

Distinguishability in Quantum Interference

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Declarations

I declare that this thesis and the work presented in it are my own. This thesis was completed while I was a candidate for a research degree at Imperial College London.

The experimental results have been obtained by me, or in collaboration with research colleagues whom I have credited appropriately. Other material resulting from collaborations has been attributed explicitly. To the best of my knowledge, I have referenced the work of others where appropriate.

London, October 2019

Alexander Edward Jones

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Abstract

Quantum interference constitutes one of the sharpest divides between our classical intuition and the weirdness of the quantum world. Whenever there are multiple paths leading to the same outcome, it is the interplay of associated probability amplitudes that determines measurement results. In the double slit experiment, wave-like interference governs the distribution of particles on a detection screen. Extending to multiple particles allows behaviour that cannot be described by a classical wave, such as the bunching of photon pairs in Hong-Ou-Mandel interference. In both these examples, the ability to distinguish the interfering paths leads to degradation of interference and a return to behaviour described by probabilities. Understanding how this transition evolves for larger multiparticle quantum systems is of fundamental interest and also vital for developing photonic quantum technologies that rely on many-photon interference.

In this thesis we investigate the role of distinguishability in the interference of three and four independent photons by using polarisation and time delays. For three photons, a new type of collective distinguishing phase appears that goes beyond a pairwise description of the similarity of quantum states. We use nonlinear photon sources and a fibre interferometer to experimentally demonstrate the ability to tune and isolate this phase in interference statistics. Extending to four photons we show that, contrary to intuition from two-photon experiments, the interference of photons prepared in distinguishable states is possible due to a four-particle phase. This is confirmed experimentally using a bulk four-mode interferometer.

We then extend our study to include the effects of state impurity in the interference of three photons. New properties of this interference enable diagnosis of realistic experimental imperfections and characterisation tasks impossible using only two photons. Finally, we link discussions of state distinguishability to the group representations underpinning the exchange symmetry of multiparticle wavefunctions.

To my family

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Author Publications

Articles

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A. E. Jones, A. J. Menssen, H. M. Chrzanowski, S. Barz & I. A. Walmsley

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“Interfering distinguishable photons”

A. E. Jones, A. J. Menssen, H. M. Chrzanowski, T. A. W. Wolterink, V. S. Shchesnovich
& I. A. Walmsley

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“Distinguishability and many-particle interference”

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“Interfering photons in orthogonal states”

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“Three photons differ in four ways”

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Contents

Declarations	2
Abstract	3
Acknowledgements	5
Author Publications	7
List of Figures	13
List of Tables	16
Abbreviations, Symbols and Constants	17
1 Introduction	19
1.1 Thesis outline	20
2 Quantum optics and interference	22
2.1 Quantum mechanics of identical particles	22
2.1.1 Identical particles	22
2.1.2 Multiparticle states and exchange symmetry	23
2.1.3 Fock space and second quantisation	24
2.1.4 Distinguishability of quantum states	25
2.1.5 Pure and mixed states	27
2.1.6 Entanglement	28
2.2 Quantum optics	29
2.2.1 Quantising the electromagnetic field	29
2.2.2 Temporal modes	31
2.2.3 Linear optics	31
2.2.4 Polarisation and the Bloch sphere	33
2.3 Interference	34
2.3.1 The superposition principle and path indistinguishability	34
2.3.2 Single-particle interference	35
2.3.3 Hong-Ou-Mandel interference	36
2.3.4 The HOM dip and distinguishability	38
2.3.5 Extending to more particles	40
2.4 Summary	43

3	Three-photon interference	44
3.1	Distinguishability of three particles	44
3.1.1	Collective distinguishability and the triad phase	44
3.1.2	Accessing the triad phase	46
3.2	Generating three photons	47
3.2.1	Single photon sources	47
3.2.2	Nonlinear optics	47
3.2.3	Spontaneous parametric down-conversion	49
3.2.4	Engineering a factorable source	51
3.2.5	Factorable SPDC in bulk crystals	52
3.2.6	SPDC source characterisation	55
3.2.7	Factorable SFWM in a silica chip	59
3.3	Three-mode interferometer	61
3.3.1	Balanced tritter	61
3.3.2	Characterisation	62
3.3.3	Interference statistics for three photons	64
3.4	Exploring distinguishability using temporal delays	65
3.4.1	State preparation and experimental setup	65
3.4.2	Results and discussion	67
3.5	Varying the triad phase	69
3.5.1	State preparation	69
3.5.2	Results and discussion	70
3.6	Isolating the triad phase	72
3.6.1	State preparation	72
3.6.2	Results and discussion	73
3.7	Conclusions	76
4	Interfering distinguishable photons	77
4.1	Graph model for distinguishability in interference	77
4.1.1	Distinguishability parameters	77
4.1.2	Graph model	78
4.2	Four-particle interference	79
4.2.1	Decomposing using triad phases	79
4.2.2	Distinguishable state interference	79
4.2.3	Pairwise distinguishable state preparation	81
4.3	Four-mode interferometer	82
4.3.1	Building a quitter	82
4.3.2	Interference statistics for a quitter	84
4.3.3	Ensuring polarisation independence	86
4.3.4	Locking the quitter phase	87
4.4	Generating four photons	88
4.4.1	Gaussian wavepackets from SPDC sources	88
4.4.2	Induced coherence and double emissions	91

4.4.3	Subtracting statistics from higher-order emissions	93
4.5	Demonstration of distinguishable state interference	95
4.5.1	Preparing polarisation and temporal modes	95
4.5.2	Results and discussion	97
4.6	Conclusions	100
5	Distinguishability and mixedness	102
5.1	Interference of mixed states	102
5.1.1	Scattering statistics for mixed states	102
5.1.2	Scattering of two and three photons	103
5.1.3	Overlaps of density matrices	104
5.2	Exploring the interference landscape	104
5.2.1	Temporal delays and identical mixed qubits	104
5.2.2	Routes from perfect to zero state overlap	106
5.3	Collective distinguishability for mixed qubits	108
5.3.1	HOM interference of mixed states	108
5.3.2	Generalising the triad phase to mixed states	108
5.4	Applications to characterisation tasks	112
5.4.1	Characterising unitaries	112
5.4.2	State tomography using interference	114
5.5	Conclusions	115
6	Symmetries and multiparticle coherence	116
6.1	Returning to first quantisation	116
6.1.1	Hilbert and Fock spaces	116
6.1.2	Partitioning the multiparticle Hilbert space	117
6.2	Capturing the symmetries of states and transformations	119
6.2.1	Representation theory and the symmetric group	119
6.2.2	Young diagrams and tableaux	120
6.2.3	Schur-Weyl duality	123
6.2.4	Unitary-unitary duality	125
6.2.5	Scattering probabilities	127
6.3	Multiparticle distinguishability in first quantisation	128
6.3.1	HOM interference	128
6.3.2	Mixed symmetry subspaces and the triad phase	130
6.4	Beyond separable states	133
6.4.1	Interfering entangled states	133
6.4.2	Simulating exotic particle statistics	135
6.5	Conclusions	137
7	Conclusions	139
7.1	Outlook	140
	Bibliography	142

A	Polarisation compensation	161
A.1	Compensation in fibre	161
A.2	Compensation in the bulk quitter interferometer	162
B	Modelling imperfections in multiphoton scattering	163
B.1	Pure states	163
B.2	Multiphoton emission and number correlations	163
B.3	Detector efficiencies and losses	164
B.4	Mixed states	166
B.5	Fluorescence and noise	166
C	Event probabilities for spatially multiplexed detectors	168
C.1	Pseudo-number resolution	168
C.2	Ideal scenario	168
C.3	Dealing with imperfections	170
C.4	Normalising to singles rates	170
D	Accessing a qutrit space using polarisation and temporal modes	173
E	Distinguishability and post-selected entanglement	175
E.1	Post-selected entanglement	175
E.2	Engineering entanglement using distinguishability	176
F	Variations in quitter twofolds from induced coherence	177
G	Data for distinguishable state interference	180
G.1	Single source arm open	180
G.2	Two source arms open	181
G.3	Three source arms open	182
H	Properties of permanents and overlaps of density matrices	183
H.1	Permanents of scattering matrices	183
H.2	Overlaps of density matrices	184

List of Figures

2.1	Partial exchange symmetry for a pair of bosons	26
2.2	Bloch sphere representation of a qubit	34
2.3	Double slit interference experiment	35
2.4	Hong-Ou-Mandel interference on a balanced beam splitter	37
2.5	Hong-Ou-Mandel dip	39
2.6	Investigating multiphoton interference	41
3.1	Pancharatnam phase in the Poincaré sphere	46
3.2	Difference-frequency generation	48
3.3	Engineering the joint spectrum	52
3.4	Birefringent phase-matching in KDP	53
3.5	Schematic of SPDC sources for generating three photons	54
3.6	SPDC sources' marginal unfiltered and filtered spectra	55
3.7	Setup for (un)heralded $g^{(2)}(0)$ correlation measurements	57
3.8	Squeezing parameter with pump power	58
3.9	HOM dips for KDP SPDC sources	58
3.10	Schematic of SFWM source for generating three heralded photons	61
3.11	Three-mode interferometer	62
3.12	Setup for probing temporal distinguishability in three-photon interference	66
3.13	Exploring three-photon interference using temporal delays	67
3.14	Probing the triad phase using polarisation	70
3.15	Testing zero triad phase with SPDC sources	71
3.16	Preparing polarisation and temporal modes to isolate the triad phase	73
3.17	Isolating the triad phase in three-photon interference	74
3.18	Phase dependence of partially bunched statistics	75
4.1	Graph model for distinguishability in interference	78
4.2	Decomposing the four-particle phase into triad phases	80
4.3	Graph model for interfering pairwise distinguishable states	80
4.4	Preparation of four photons in pairwise distinguishable states	81
4.5	Different realisations of a balanced four-mode interferometer	82

4.6	Folded bulk optic quitter	83
4.7	Phase-dependent HOM dips in a quitter	85
4.8	Ensuring the quitter phase is polarisation-independent	87
4.9	Demonstration of locking the quitter phase	88
4.10	Ideal and experimentally feasible temporal mode preparations	89
4.11	Exchange contributions and signal visibility with quitter phase	90
4.12	Induced coherence experiment by Mandel	91
4.13	Induced coherence using SPDC sources and a quitter	92
4.14	Double source emissions and fourfold coincidences	93
4.15	Filtered spectra for preparing pairwise distinguishable states	95
4.16	Calibrating the four-particle phase	96
4.17	Ensuring the temporal distinguishability of pairwise distinguishable states	97
4.18	Experimental setup for distinguishable state interference	98
4.19	Counts measured at the quitter outputs for distinguishable state interference	99
5.1	Accessing the interference landscape using temporal delays and mixedness	106
5.2	Different routes from zero to unit overlap	107
5.3	Illustrating the equivalence of mixedness and distinguishability in HOM	109
5.4	Scalar triple product of three Bloch vectors	110
5.5	Identifying the source of a reduction in pairwise overlaps	111
5.6	Interfering three qubits in the dual-rail encoding	113
6.1	Partitioning the two-particle Hilbert space	118
6.2	The irreducible representation of a group	120
6.3	Tracing out the label degree of freedom for two photons	130
6.4	Tracing out the label degree of freedom for three photons	132
6.5	Unitary evolution of a three-particle state in the system space	133
6.6	GHZ-interferometer for N -particle interference of entangled states	134
A.1	Polarisation compensation for interferometers	161
B.1	Contamination of interference statistics by multiphoton emission	164
B.2	Modelling loss at interferometer inputs	165
C.1	Using beam splitters for pseudo-number resolution	169
C.2	Probabilistic discrimination of two photons in a single mode	169
E.1	Heralding entanglement of matter systems	175
E.2	Heralding different types of entanglement	176
F.1	Labelling of sources and quitter for induced coherence	177
F.2	Interference of two sources' emissions in a quitter	178
G.1	Singles counts during distinguishable state interference run	180
G.2	Twofold quitter coincidences during distinguishable state interference run	181

G.3	Threefold quitter coincidences during distinguishable state interference run . . .	182
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List of Tables

4.1	Exchange contributions to the four-photon coincidence probability	89
4.2	Checking filtered wavepackets' coherence lengths	95
6.1	States spanning the symmetric irreps for three particles	122
6.2	States spanning the mixed symmetry irreps for three particles	123
6.3	Numbers and dimensions of irreps for S_2 and $SU(2)$	125
6.4	Numbers and dimensions of irreps for S_3 and $SU(3)$	125

Abbreviations, Symbols and Constants

List of Abbreviations

APD Avalanche Photodiode

BS Beam Splitter

CW Continuous Wave

DFG Difference-Frequency Generation

HOM Hong-Ou-Mandel (interference)

HWP Half-Wave Plate

KDP Potassium Dihydrogen Phosphate

PBS Polarising Beam Splitter

QWP Quarter-Wave Plate

SHG Second-Harmonic Generation

SMF Single-Mode Fibre

(S)FWM (Spontaneous) Four-Wave Mixing

(S)PDC (Spontaneous) Parametric Down-Conversion

Ti:Sapph Titanium Sapphire oscillator

TMSV Two-Mode Squeezed Vacuum

List of Symbols and Constants

Fundamental Constants

c Speed of light

e Charge of an electron

ε_0 Permittivity of free space

$\hbar$ Reduced Planck constant

Mathematical

$*$ Complex conjugate

$*$ Hadamard (element-wise) matrix product

$\dagger$ Hermitian conjugate

∇ Nabla gradient operator

$\oplus$ Direct sum

$\otimes$ Tensor product

$\vee$ Symmetric tensor product

i Imaginary unit

$\delta(x)$ Dirac delta function

δ_{ij} Kronecker delta

e Base of the natural logarithm

ε_{ijk} Levi-Civita symbol

$\mathcal{H}$ Hilbert space

$\text{Re}(z)$ Imaginary component of z : $\text{Im}(z) = \frac{1}{2i}(z - z^*)$

$\text{Im}(z)$ Real component of z : $\text{Re}(z) = \frac{1}{2}(z + z^*)$

$\text{Arg}(z)$ Argument of z : $z = |z|e^{i\theta} \implies \text{Arg}(z) = \theta$

$\text{Tr}(A)$ Trace of A

S_N Symmetric (permutation) group of degree N

$SU(d)$ Special unitary group of degree d

Introduction

The study of the interference of light can be traced back to Young’s double slit experiment at the start of the 19th century, where the observation of interference fringes suggested a wave description of light [1]. This was bolstered by Maxwell’s theory of light as an electromagnetic wave [2]. A century after Young’s experiment, Planck and Einstein made the conceptual leap that light comprises discrete quanta of energy: photons [3, 4]. The resulting concept of wave-particle duality heralded the end of a classical description of light and sparked the development of quantum theory.

The double slit experiment also reveals that much of the magic in the quantum world stems from indistinguishability. As long as it is impossible to discern which slit a quantum particle passes, the two possibilities interfere to give wave-like fringes on a detection screen. Between source and screen, this ignorance is captured by describing the particle as being in a superposition of both possible routes, a very non-classical notion. This new principle of physical reality is so counter-intuitive that it was famously described by Feynman as “contain[ing] the *only* mystery” of quantum mechanics [5].

Things become even more mysterious when the concept of indistinguishability is applied to larger systems. If some process involves multiple particles then, if they are identical, an equivalent process where a pair of particles is swapped is indistinguishable. These two possibilities will interfere in a way that depends on the symmetry of the particles. Bosons and fermions have different symmetries and so can lead to very different interference effects. An experiment that elegantly combines quantum interference and the bosonic symmetry of photons is the Hong-Ou-Mandel (HOM) effect [6]. Consider two independent photons incident on the two ports of a balanced beam splitter. Applying classical intuition, they are expected to emerge from different ports half the time. However if the photons are completely identical in all their properties – such as colour, polarisation and arrival time – they will *never* emerge in different output ports. The indistinguishability of the two routes to coincidences means quantum interference suppresses that outcome.

Hence even though photons by themselves do not interact, indistinguishability and quantum interference can unlock non-classical correlations. Indeed the HOM effect is at the heart of a linear optical approach to building a universal quantum computer [7, 8]. Driven by recent advances in photon source technology, multiphoton interference has found applications in quantum

simulations and boson sampling has emerged as a candidate for demonstrating a quantum advantage without requiring universality [9, 10]. The ability to manipulate large entangled states of photons also holds promise for quantum computation [11]. Clearly the exquisite control of large many-body quantum systems will underpin future quantum technologies, so it is critical to understand how large-scale interference changes with distinguishing information.

This thesis examines one of the most fundamental processes in quantum optics: the interference of independent photons. We perform a systematic investigation, both theoretically and experimentally, of extensions of Hong-Ou-Mandel interference to three and four photons. Even at this small scale, the increase in complexity means there are plenty of surprises: as well as going some way in addressing the impact of imperfections as systems grow in size, we also challenge fundamental intuitions about multiparticle interference.

1.1 Thesis outline

In Chapter 2 we present some of the theoretical background required for the rest of this thesis. We begin with the operator formalism for treating identical particles and an introduction to the distinguishability of particles' quantum states. After reviewing key concepts in linear optics, we use the double slit experiment to illustrate the concept of quantum interference. The HOM effect then reveals the links between interference, pairwise distinguishability of states and exchange symmetry. This is followed by an extension to more photons that sets the scene for subsequent chapters.

Chapter 3 concerns a new collective distinguishability parameter associated with the exchange of three photons: the triad phase. This is shown to be a fundamentally different property to that governing HOM interference. We then review the application of nonlinear optics to generating photons, with particular emphasis on a source based on spontaneous parametric down-conversion engineered to produce pairs of independent photons. A series of three-photon experiments are presented that reveal the richer interference landscape accessible for larger systems. We also demonstrate the ability to tune and isolate the three-photon exchange contribution to interference statistics using the polarisation and temporal degrees of freedom.

In Chapter 4 we describe a graph model that can be used to determine which distinguishing parameters for multiple particles are independent. If none of the participating particles' states are distinguishable from one another, then it is possible to decompose all higher-order distinguishing phases into triad phases. However we then consider a preparation of four photons for which this is not the case, and interference depending on a four-particle phase is shown to be possible despite pairs of states being distinguishable. This challenges the intuition from HOM that distinguishability is accompanied by a return to probabilistic behaviour. We then show how to build a four-mode interferometer using bulk optics and describe the preparation of four photons in suitable states. An experimental demonstration of this interference of photons in distinguishable states is presented.

The effect of impure state preparation on interference is considered in Chapter 5. We show experimentally that coherent and incoherent effects can allow access to different parts of the interference landscape for three photons. An intuitive geometric picture highlights the different

roles of distinguishability and mixedness for photons with a qubit degree of freedom. We use this to investigate the ability to diagnose the degradation of interference, and also propose applications to characterisation tasks.

In Chapter 6 we delve into the representation theory of the groups underpinning the exchange symmetry and unitary evolution of multiparticle quantum states. This is used to decompose bosonic wavefunctions from a form where particle exchange symmetry is obvious to one where symmetries of the photons' constituent degrees of freedom are explicit. In this context, distinguishability is associated with occupation of non-symmetric parts of the Hilbert spaces for each degree of freedom. We then discuss the use of entangled states to access these spaces and simulate non-bosonic particle symmetries. Finally conclusions and outlook are given in Chapter 7.

Quantum optics and interference

In this chapter we review some basic concepts used throughout the rest of this thesis, with particular emphasis on the linear algebra and symmetry at the core of quantum mechanics. We begin by describing systems of identical quantum particles in first and second quantised formalisms, and discuss how partial exchange symmetry of the wavefunction defines the concept of state distinguishability. After identifying photons as the bosonic particles in quantum optics, we show how operators can be used to describe and evolve quantum states of light. Finally the relationships between coherence, path indistinguishability, partial exchange symmetry, and their roles in quantum interference are discussed.

2.1 Quantum mechanics of identical particles

2.1.1 Identical particles

When we talk of particles being identical, we mean that all their intrinsic properties such as rest mass, charge, and spin are the same. A single quantum particle in a pure quantum state is described by a normalised vector¹ in some d -dimensional Hilbert space $\mathcal{H}^{(1)}$, defined as a complex vector space endowed with an inner product [12]. This single particle Hilbert space is given by the tensor product of the spaces for its independent degrees of freedom, for example $\mathcal{H}^{(1)} = \mathcal{H}_A \otimes \mathcal{H}_B$ for degrees of freedom A and B . Assuming that these spaces are spanned respectively by basis vectors $|\mathbf{e}_A^{(i)}\rangle, |\mathbf{e}_B^{(j)}\rangle$, then the basis states of the joint system are given by $|\mathbf{e}_{AB}^{(i,j)}\rangle = |\mathbf{e}_A^{(i)}\rangle \otimes |\mathbf{e}_B^{(j)}\rangle$. Some single particle quantum state $|\psi\rangle$ is then described by a vector

$$|\psi\rangle = \sum_{i,j} c_{ij} |\mathbf{e}_{AB}^{(i,j)}\rangle \in \mathcal{H}^{(1)}, \quad \sum_{i,j} |c_{ij}|^2 = 1. \quad (2.1)$$

The complex coefficients c_{ij} are the projections of $|\psi\rangle$ onto basis states $|\mathbf{e}_{AB}^{(i,j)}\rangle$, and $|c_{ij}|^2$ are the probabilities of the particle being found in that particular basis state.

¹More precisely it is described by a *ray*: a set of non-zero vectors differing only by complex phase factors.

2.1.2 Multiparticle states and exchange symmetry

The joint Hilbert space for a system of N identical, non-interacting particles is constructed by taking the tensor product of the identical single particle spaces:

$$\mathcal{H}^{(N)} = \left(\mathcal{H}^{(1)} \right)^{\otimes N}. \quad (2.2)$$

This space has d^N dimensions and contains all d^N -dimensional vectors, but which vector $|\Psi\rangle$ describes the physical state of N particles? The indistinguishability of identical particles means that no observable can distinguish states that are related by particle exchange [13–15]. In order to describe a physically equivalent state, the vector $|\Psi\rangle$ can therefore only change by an unobservable global phase θ when a pair of particles are swapped [16, 17].² Defining the permutation operator $\hat{P}_{i,j}$ that exchanges the particles i and j , we may write

$$\hat{P}_{i,j} |\Psi\rangle = e^{i\theta} |\Psi\rangle. \quad (2.3)$$

Permuting the particles again and using the self-inverse property of the operator³ gives

$$\begin{aligned} \hat{P}_{i,j}^2 |\Psi\rangle &= e^{2i\theta} |\Psi\rangle \equiv |\Psi\rangle \\ \implies \hat{P}_{i,j} |\Psi\rangle &= \pm |\Psi\rangle, \end{aligned} \quad (2.4)$$

where the phases $\theta = 0, \pi$ describe respectively states that are symmetric and anti-symmetric under permutation. By the spin-statistics theorem, the symmetric states with eigenvalue $+1$ describe particles of integer spin – *bosons*, that obey Bose-Einstein statistics – and the anti-symmetric states with eigenvalue -1 describe particles with half-integer spin – *fermions*, that obey Fermi-Dirac statistics [16, 24]. Wavefunction symmetry has many deep physical consequences. For example, the Pauli Exclusion Principle forbids two fermions from occupying the same quantum state [25], and manifests at hugely different scales in the Universe: at one end it defines the shell structure of electrons around the atomic nucleus and at the other it prevents the gravitational collapse of neutron stars.

The vectors describing bosons’ wavefunctions are in the symmetric part of the joint Hilbert space $\mathcal{H}_S^{(N)}$ and those describing fermions are in the anti-symmetric part $\mathcal{H}_A^{(N)}$. Before explicitly constructing these vectors we will briefly describe the symmetric group. S_N is the group of all permutations of a system of N objects (here particles) and contains $N!$ elements. The elements σ are mappings of the ordered list $(1, 2, \dots, N-1, N)$ to another list of the same length, where the i th entry is the position to which object i of the original list is mapped. For example in S_3 , $\sigma_1 : (1, 2, 3) \rightarrow (1, 2, 3)$ corresponds to the ‘do nothing’ permutation, or equivalently the identity. The element $\sigma_2 = (1, 2, 3) \rightarrow (2, 1, 3)$ corresponds to the permutation where the first

²This is a typical approach to establishing how particle indistinguishability leads to restrictions on the vectors describing multiparticle wavefunctions [17]. In fact the invariance of observables under particle permutation does not automatically imply symmetric or anti-symmetric state vectors [14, 15, 18–20]. This instead comes from the empirical ‘Symmetrisation Postulate’: the only multiparticle states of identical particles found in nature are either totally symmetric or totally anti-symmetric [13, 20]. This will be discussed in the final section of Chapter 6.

³This property of the permutation operator only holds for systems where the particles may access more than two spatial dimensions. For two-dimensional systems, the topology of a permutation can give rise to other values of the acquired phase θ [21, 22]. Particles that have phases other than $\theta = 0, \pi$ are called anyons [23].

two elements are swapped. The notation $\sigma(j)$ indicates the number to which j is permuted under the element σ . Any permutation can be decomposed into a sequence of swaps of two elements, called transpositions.

The task now is to construct the bosonic and fermionic state vectors for a collection of N identical bosons and fermions in known single particle states $|\phi_k\rangle$. In first quantised notation, the ordering of kets in a vector implicitly corresponds to the assignment of a state to a particular particle. We take an N -particle product state:

$$|\Psi_D\rangle = |\phi_1\rangle \otimes |\phi_2\rangle \otimes \dots \otimes |\phi_{N-1}\rangle \otimes |\phi_N\rangle \quad (2.5)$$

which is a vector in $\mathcal{H}^{(N)}$ that does not yet have defined exchange symmetry. Application of the symmetrising and anti-symmetrising operators $\hat{S}$ and $\hat{A}$ projects this state into respectively $\mathcal{H}_S$ and $\mathcal{H}_A$, giving the vectors:

$$\begin{aligned} |\Psi_S\rangle &= \hat{S} |\Psi_D\rangle = \mathcal{N}_S \sum_{\sigma \in S_N} \hat{P}_\sigma |\Psi_D\rangle \\ &= \mathcal{N}_S \sum_{\sigma \in S_N} |\phi_{\sigma(1)}\rangle \otimes |\phi_{\sigma(2)}\rangle \otimes \dots \otimes |\phi_{\sigma(N-1)}\rangle \otimes |\phi_{\sigma(N)}\rangle, \\ |\Psi_A\rangle &= \hat{A} |\Psi_D\rangle = \mathcal{N}_A \sum_{\sigma \in S_N} \text{sgn}(\sigma) \hat{P}_\sigma |\Psi_D\rangle \\ &= \mathcal{N}_A \sum_{\sigma \in S_N} \text{sgn}(\sigma) |\phi_{\sigma(1)}\rangle \otimes |\phi_{\sigma(2)}\rangle \otimes \dots \otimes |\phi_{\sigma(N-1)}\rangle \otimes |\phi_{\sigma(N)}\rangle. \end{aligned} \quad (2.6)$$

$\hat{P}_\sigma$ permutes particles according to σ , and the $\text{sgn}(\sigma)$ function is the parity of the permutation, taking the value $+(-)1$ for an even (odd) number of transpositions to decompose σ . The normalisation factors are given by:

$$\mathcal{N}_S = \left(N! \times \prod_k n_k! \right)^{-\frac{1}{2}}, \quad \mathcal{N}_A = (N!)^{-\frac{1}{2}}. \quad (2.7)$$

n_k is the number of particles found in state $|\phi_k\rangle$. This factor is not present in $\mathcal{N}_A$ because if any single particle state is more than singly occupied, it is impossible to construct an anti-symmetrised state – precisely the Pauli Exclusion Principle mentioned earlier. It is straightforward to verify that these vectors have the requisite symmetries under permutation that were found in equation 2.4.

2.1.3 Fock space and second quantisation

There are a few problems with this first quantised approach. It is fundamentally impossible to say which particle is in which state so labelling each particle is somewhat redundant, and expressing the symmetrisation of systems of many particles using equation 2.6 quickly becomes cumbersome. We have also so far assumed a fixed particle number which is not the case for photons, where nonlinear processes can create or destroy them.

To refine our notation we now describe physical states by counting how many particles are in each single particle state using the *occupation number representation*. Defining a set

of basis vectors $\{|\mathbf{e}_i\rangle\}$ that span the single particle space $\mathcal{H}^{(1)}$, we can write the multiparticle wavefunction by specifying the number of particles n_i in each state $|\mathbf{e}_i\rangle$. For N bosons occupying a space with M basis vectors, the multiparticle states residing in $\mathcal{H}_S^{(N)}$ have the form

$$|n_1, n_2, \dots, n_M\rangle \equiv \hat{S} \left(\bigotimes_{i=1}^M |\mathbf{e}_i\rangle^{\otimes n_i} \right), \quad \sum_i n_i = N. \quad (2.8)$$

Each partition of N is associated with a basis vector in this new representation. It is possible to decompose the single particle states $|\phi_k\rangle$ in the basis $\{|\mathbf{e}_i\rangle\}$, and then general states of fixed particle number – *Fock states* – may be written as linear combinations of the states in equation 2.8.

In order to accommodate different numbers of bosons, the space of available states is extended by constructing the *Fock space*:

$$\mathcal{F} = \mathcal{H}^{(0)} \oplus \mathcal{H}^{(1)} \oplus \mathcal{H}_S^{(2)} \oplus \mathcal{H}_S^{(3)} \oplus \dots \quad (2.9)$$

$\mathcal{H}^{(0)}$ is the space with zero particles and contains only the vacuum state $|0\rangle = |0, 0, 0, \dots\rangle$ with zero particles, that satisfies the usual normalisation condition for basis vectors $\langle 0|0\rangle = 1$. The direct sum of spaces also includes the single particle space and multiparticle symmetrised spaces. It is now possible to write down quantum states that do not have definite particle number by taking linear combinations of different Fock states.

Next we define the *bosonic creation and annihilation operators* $\hat{a}_i^\dagger, \hat{a}_i$ by their mappings between different multiparticle states:⁴

$$\begin{aligned} \hat{a}_i^\dagger |n_1, \dots, n_i, n_{i+1}, \dots\rangle &= \sqrt{n_i + 1} |n_1, \dots, n_i + 1, n_{i+1}, \dots\rangle, \\ \hat{a}_i |n_1, \dots, n_i, n_{i+1}, \dots\rangle &= \sqrt{n_i} |n_1, \dots, n_i - 1, n_{i+1}, \dots\rangle. \end{aligned} \quad (2.10)$$

These operators respectively increase and decrease by one the occupation number of the single particle state $|\mathbf{e}_i\rangle$. We also define the number operator $\hat{n}_i = \hat{a}_i^\dagger \hat{a}_i$ that counts the number of particles in the state $|\mathbf{e}_i\rangle$. Commutation relations for these operators can be used to capture the bosonic symmetry of the state:

$$[\hat{a}_i, \hat{a}_j^\dagger] = \hat{a}_i \hat{a}_j^\dagger - \hat{a}_j^\dagger \hat{a}_i = \delta_{ij}, \quad [\hat{a}_i, \hat{a}_j] = [\hat{a}_i^\dagger, \hat{a}_j^\dagger] = 0. \quad (2.11)$$

These relations completely capture the symmetry requirements for states of multiple bosons. It is now possible to describe states with an indefinite particle number by using sums of different numbers of creation operators when constructing a wavefunction.

2.1.4 Distinguishability of quantum states

Identical particles may have degrees of freedom that act as ‘labels’. These are preserved as the state evolves and so serve to distinguish one particle from another. The extent to which this is possible is called the *distinguishability* of particles’ quantum states in that label. This is

⁴It is also possible to perform this procedure for fermions. Their defining anti-commutators are $\{\hat{c}_i, \hat{c}_j^\dagger\} = \delta_{ij}$ and $\{\hat{c}_i, \hat{c}_j\} = \{\hat{c}_i^\dagger, \hat{c}_j^\dagger\} = 0$, where if $i = j$ the last one describes the Pauli Exclusion Principle: $n_i = 0, 1$ only.

intimately related to the concept of wavefunction symmetry under *partial exchange* of degrees of freedom [26]. Consider a pair of identical bosons with position and spin degrees of freedom. We start with an initial state where the particles are in non-overlapping position states: they are spatially separated and described by orthogonal state vectors $|L\rangle$ and $|R\rangle$, for left and right respectively. Their spin states, here acting as the label, are denoted by a and b so the overall state is (Figure 2.1)

$$|\psi_{LR}^{ab}\rangle = \hat{a}_{L,a}^\dagger \hat{a}_{R,b}^\dagger |0\rangle = |1_{L,a}, 1_{R,b}\rangle. \quad (2.12)$$

We have used the occupation number representation, and this state lies in $\mathcal{H}_S^{(2)}$. Now simultaneously exchanging both position and spin is equivalent to particle exchange, and here leaves the system in the state $\hat{a}_{R,b}^\dagger \hat{a}_{L,a}^\dagger |0\rangle$. Using the commutation relations of equation 2.11, this is the same as the original state and demonstrates the exchange symmetry of the bosonic wavefunction.

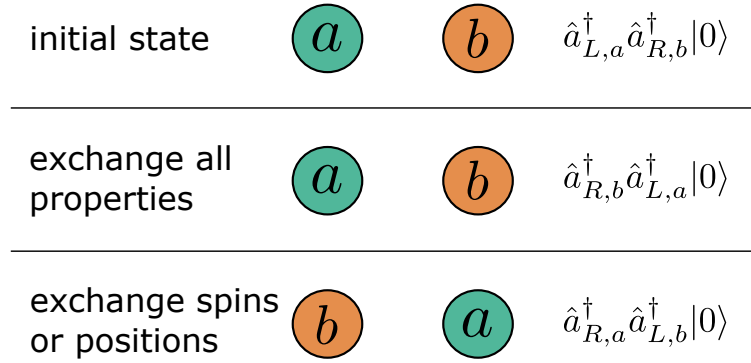


Figure 2.1: Two identical bosonic particles are prepared in a state where spin a is on the left, and spin b is on the right. The coloured circles represent the internal spin states that may or may not be the same. We consider the physical state of the system under total exchange, or partial exchange of the states for each degree of freedom.

What about if we only exchanged the positions of the particles? This leaves the system in $|\psi_{RL}^{ab}\rangle = \hat{a}_{R,a}^\dagger \hat{a}_{L,b}^\dagger |0\rangle$ which can be different from the original state if the spin states are not the same (Figure 2.1). Performing a Gram-Schmidt orthogonalisation gives an orthogonal basis for the spins, $\{|a\rangle, |a^\perp\rangle\}$. Taking the overlap between the original and partially exchanged states then gives

$$\begin{aligned} \langle \psi_{RL}^{ab} | \psi_{LR}^{ab} \rangle &= \langle 0 | \hat{a}_{R,a} \hat{a}_{L,b} \hat{a}_{L,a}^\dagger \hat{a}_{R,b}^\dagger | 0 \rangle \\ &= \langle a|b \rangle_R \langle b|a \rangle_L \\ &= |\langle a|b \rangle|^2. \end{aligned} \quad (2.13)$$

This can also be shown by explicitly casting the wavefunctions in first quantisation. So here the similarity is dictated by the squared modulus of the overlap of the internal spin states. If these are the same then the partially exchanged state is identical to the original; the overlap of the spin states is unity and we say that the particles are indistinguishable in their spin degree of freedom. On the other hand if the spin states are orthogonal, the overlap is zero and the particles are distinguishable in their spin – a measurement could tell with certainty whether exchange has occurred. It is the correlations between the position and spin properties of the particles that allow a swap of position to give information on the internal states.

When referring to distinguishability of states we implicitly assume that there is some other degree of freedom in which the states are orthogonal and that we can permute to reveal distinguishing information. In discussions that follow this will usually be the spatial modes of an interferometer. The squared modulus of the internal state overlap $|\langle a|b\rangle|^2$ is called the *pairwise distinguishability* of the particles' label states. It varies between zero and one and captures the symmetry under partial exchange. In Chapters 3 and 4 we will investigate what new multi-particle distinguishing parameters arise for larger systems, and in Chapter 6 a more detailed study of this exchange symmetry will be presented.

2.1.5 Pure and mixed states

A pure quantum state is described by a single vector $|\psi\rangle$ in a Hilbert space. Taking a normalised linear combination of multiple vectors yields another vector that also describes a valid quantum state. This is a *superposition* of multiple quantum states and the coherence of the procedure means they all have a defined relative phase relationship. Situations can arise where different pure states are prepared incoherently: for example if an experimenter makes the states $|\psi_i\rangle$ with probabilities p_i . In order to describe this ensemble we can define the *density operator*⁵ [12]

$$\hat{\rho} = \sum_i p_i |\psi_i\rangle\langle\psi_i|. \quad (2.14)$$

This object contains all information about the ensemble and we will see it can be used to calculate the probabilities of measurement outcomes. If the system evolves under a unitary operator $\hat{U}$ then the pure states become $\hat{U}|\psi_i\rangle$ and the density operator becomes $\hat{U}\hat{\rho}\hat{U}^\dagger$.

The density operator has three defining properties: it is Hermitian, it has unit trace $\text{Tr}(\hat{\rho}) = \sum_i p_i = 1$, and it is positive semi-definite. An important quantity associated with $\hat{\rho}$ is its *purity*

$$\mathcal{P} = \text{Tr}(\hat{\rho}^2) = \sum_i p_i^2. \quad (2.15)$$

A pure state can be written as $\hat{\rho} = |\psi\rangle\langle\psi|$ and has $\mathcal{P} = 1$. Anything with a purity less than one has a degree of *mixedness*. The maximally mixed state for a single particle is given by an equally weighted incoherent sum of all the basis states spanning its d -dimensional Hilbert space, and has $\mathcal{P} = 1/d$. A similar procedure can be performed for multiparticle mixed states.

This formalism is useful when describing composite quantum systems where you might not have access to some subsystem or degree of freedom. For example, suppose you have a system described by $\hat{\rho}_{AB}$ that lies in the tensor product space $\mathcal{H}_A \otimes \mathcal{H}_B$ but you can only perform measurements that resolve the A degree of freedom. We define the *reduced density matrix*

$$\hat{\rho}_A = \text{Tr}_B(\hat{\rho}_{AB}), \quad (2.16)$$

where we have performed a partial trace over B to be left with a density matrix in $\mathcal{H}_A$. This object describes the *reduced state* and can be used to assign probabilities to measurement outcomes that are insensitive to the B degree of freedom.

⁵This operator performs a mapping in a Hilbert space but, as we will often be interested in its matrix elements in a physically relevant basis, we will also use the term *density matrix* and symbol ρ interchangeably.

2.1.6 Entanglement

We have just seen how lack of access to some degree of freedom means defining a reduced density matrix that can then be used to calculate other measurement outcomes. Now consider a system of multiple identical particles, some of which we cannot access – what information is contained in the quantum state of the individual particles we do have access to? The quantum state of a *separable* system is one for which each particle is separately described by a pure state. For two particles this means that, despite lack of knowledge of one particle, you still have complete information about the other. An example of such a state is:

$$|\psi_{sep}\rangle = |L, a\rangle |R, b\rangle \quad (2.17)$$

where the state is in the Fock space, L, R denote orthogonal spatial states, and a, b label spin states. Such a state is often written just as $|a, b\rangle$ where the ordering implicitly indicates that there is another degree of freedom in which the states are orthogonal, similar to how we defined state distinguishability. We highlight this to emphasise what we mean by ignorance of a particle during a measurement. From earlier discussions regarding symmetrisation of wavefunctions, it is clear that one cannot be ignorant of a specific particle since particle labels have no physical relevance. An experimentalist can, however, be ignorant of a particular mode of a degree of freedom (here space) and as a result also be ignorant of other properties correlated with the particle there. In this example, lack of access to the particle on the right means the remaining system is described by a reduced density matrix $\hat{\rho}_{sep}^L = |a\rangle\langle a|$. This is a pure state and so contains complete information about the state of the particle on the left. Swapping which position we are ignorant about yields the same conclusion that particles at different positions have pure spin states, so results of measurements on them are independent.

Is it possible to have a state of two particles where information is lost under such a procedure? Consider an *entangled state* comprising a superposition of two-particle states:

$$|\psi_{ent}\rangle = \frac{1}{\sqrt{2}} (|a, b\rangle + |b, a\rangle), \quad (2.18)$$

again with an implicit distinguishing degree of freedom. Tracing out one of the particles leaves the other in the state $\hat{\rho}_{ent}^{R/L} = \frac{1}{2} (|a\rangle\langle a| + |b\rangle\langle b|)$. This is a mixed state that does not constitute a complete description of the particle. If a and b are orthogonal, then measuring the spin state of either particle in that basis will look random, with each outcome a, b appearing equally often. However if these outcomes for both are compared, they will be found to be perfectly correlated. Information about the system is contained non-locally in the entanglement between the particles, and so a full description of the state requires consideration as a whole⁶. It is hard to do justice to the historical, conceptual and technological importance of this phenomenon, and it finds myriad uses in quantum computing, communication, teleportation and metrology to name just a few [28].

⁶For further discussions on the distinction between entanglement in the usual sense, and the formal entanglement inherent due to wavefunction symmetrisation, see [20, 27].

2.2 Quantum optics

2.2.1 Quantising the electromagnetic field

We will now link the second quantised machinery for dealing with multiparticle wavefunctions to the bosonic quantised excitations of the electromagnetic field: photons. To begin we give a quick recap of some of the key results from a quantum treatment of the harmonic oscillator [22, 29]. Such a system is described by the Hamiltonian

$$\hat{H}_{QHO} = \frac{\hat{p}^2}{2m} + \frac{1}{2}m\omega^2\hat{x}^2 \quad (2.19)$$

where the spring constant $K = m\omega^2$, m is the mass, and ω is the angular frequency. We have defined the position operator $\hat{x}$ and momentum operator $\hat{p} = -i\hbar\frac{\partial}{\partial x}$. These obey the commutation relation $[\hat{x}, \hat{p}] = i\hbar$ that, through the Heisenberg uncertainty principle, dictates the precision to which the position and momentum of such an oscillator can be known. Solving the time-independent Schrödinger equation reveals the eigenstates of the Hamiltonian are Hermite functions $\psi_n(x)$ with $n = 0, 1, 2, \dots$. These have associated eigenvalues $E_n = \hbar\omega(n + 1/2)$ that are equally spaced in energy by $\hbar\omega$. We can define new operators

$$\begin{aligned} \hat{a} &= \sqrt{\frac{m\omega}{2\hbar}} \left(\hat{x} + \frac{i}{m\omega}\hat{p} \right), \\ \hat{a}^\dagger &= \sqrt{\frac{m\omega}{2\hbar}} \left(\hat{x} - \frac{i}{m\omega}\hat{p} \right) \end{aligned} \quad (2.20)$$

that obey the commutation relations $[\hat{a}, \hat{a}^\dagger] = 1$. The Hamiltonian can then be recast as

$$\hat{H}_{QHO} = \hbar\omega \left(\hat{a}^\dagger\hat{a} + \frac{1}{2} \right). \quad (2.21)$$

We define the number operator $\hat{n} = \hat{a}^\dagger\hat{a}$ that has eigenstates $|n\rangle$ with associated eigenvalue n , and so recover the expression for eigenenergies E_n . The actions of these operators on the eigenstates are given by

$$\begin{aligned} \hat{a}^\dagger |n\rangle &= \sqrt{n+1} |n+1\rangle, \\ \hat{a} |n\rangle &= \sqrt{n} |n-1\rangle \end{aligned} \quad (2.22)$$

They respectively increase or decrease the energy of the system by $\hbar\omega$. We now make a conceptual leap by treating these packets of fixed energy as *quantum particles*. Instead of thinking of a harmonic oscillator as having energy E_n , we consider it as possessing a harmonic mode occupied by n bosonic particles each with energy $\hbar\omega$. This is the occupation number representation from earlier and so these operators can be associated with the creation or annihilation of particles.

Now we turn to the quantisation of the electromagnetic field [30, 31]. Defining the vector potential $\mathbf{A}(\mathbf{r}, t)$ and adopting the Coulomb gauge $\nabla \cdot \mathbf{A} = 0$, the electric and magnetic fields in vacuum are given by

$$\begin{aligned} \mathbf{E}(\mathbf{r}, t) &= -\frac{\partial}{\partial t}\mathbf{A}(\mathbf{r}, t), \\ \mathbf{B}(\mathbf{r}, t) &= \nabla \times \mathbf{A}(\mathbf{r}, t). \end{aligned} \quad (2.23)$$

Along with Maxwell's equations, these show that the vector potential obeys the wave equation

$$\nabla^2 \mathbf{A}(\mathbf{r}, t) = \frac{1}{c^2} \frac{\partial^2 \mathbf{A}(\mathbf{r}, t)}{\partial t^2}. \quad (2.24)$$

Assuming we are solving for the field inside a cavity with volume $V = L^3$, a separation of the space and time variables means the spatial functions satisfy the Helmholtz equation. Periodic boundary conditions define spatial distributions that are modes of the cavity, and the time-varying part of the field is given by solutions to the harmonic oscillator. The solution is a superposition of these modes labelled by the index $\mathbf{k}$ and given by

$$\mathbf{A}(\mathbf{r}, t) = \sum_{\mathbf{k}, \lambda} \left(\mathbf{e}_{\mathbf{k}, \lambda} a_{\mathbf{k}, \lambda} e^{i(\mathbf{k} \cdot \mathbf{r} - \omega_k t)} + \mathbf{e}_{\mathbf{k}, \lambda}^* a_{\mathbf{k}, \lambda}^* e^{-i(\mathbf{k} \cdot \mathbf{r} - \omega_k t)} \right) \quad (2.25)$$

where we have included some complex coefficients $a_{\mathbf{k}, \lambda}$. The wavevector $\mathbf{k} = (k_x, k_y, k_z) = \frac{2\pi}{L}(n_x, n_y, n_z)$, with n_i taking values $0, \pm 1, \pm 2, \dots$, labels electromagnetic modes, and the angular frequency is given by the dispersion relation $\omega_k = c|\mathbf{k}|$. We have also defined the polarisation vectors $\mathbf{e}_{\mathbf{k}, \lambda}$ that satisfy $\mathbf{e}_{\mathbf{k}, \lambda} \cdot \mathbf{k} = 0$ due to the transversality of the field imposed by the Coulomb gauge. We take $\lambda \in (1, 2)$ to denote horizontal and vertical polarisations of the vector potential respectively. In order to quantise this field, the complex coefficients are promoted to operators:

$$a_{\mathbf{k}, \lambda} \rightarrow \sqrt{\frac{\hbar}{2\varepsilon_0 \omega_k V}} \hat{a}_{\mathbf{k}, \lambda} \quad (2.26)$$

and similarly the conjugate coefficients are mapped to the adjoint operators $\hat{a}_{\mathbf{k}, \lambda}^\dagger$. The normalisation ensures dimensionless operators. Knowing that the relevant particles here are going to be spin-1 photons, we also impose the bosonic commutation relations derived earlier (equation 2.11):

$$[\hat{a}_{\mathbf{k}, \lambda}, \hat{a}_{\mathbf{k}', \lambda'}^\dagger] = \delta_{\mathbf{k}, \mathbf{k}'} \delta_{\lambda, \lambda'}, \quad [\hat{a}_{\mathbf{k}, \lambda}, \hat{a}_{\mathbf{k}', \lambda'}] = [\hat{a}_{\mathbf{k}, \lambda}^\dagger, \hat{a}_{\mathbf{k}', \lambda'}^\dagger] = 0. \quad (2.27)$$

The Hamiltonian for the electromagnetic field is obtained by integrating the energy density over the volume V :

$$H_{EM} = \frac{1}{2} \int_V \left(\varepsilon_0 |\mathbf{E}(\mathbf{r}, t)|^2 + \frac{1}{\mu_0} |\mathbf{B}(\mathbf{r}, t)|^2 \right) d^3\mathbf{r}. \quad (2.28)$$

Armed with the quantised vector potential $\hat{\mathbf{A}}(\mathbf{r}, t)$ we can calculate the electric and magnetic fields in terms of operators using equation 2.23, and substituting gives the quantised Hamiltonian for the field:

$$\hat{H}_{EM} = \sum_{\mathbf{k}, \lambda} \hbar \omega_k \left(\hat{a}_{\mathbf{k}, \lambda}^\dagger \hat{a}_{\mathbf{k}, \lambda} + \frac{1}{2} \right). \quad (2.29)$$

This is the same Hamiltonian we found for the quantum harmonic oscillator. Each mode of the electromagnetic field therefore behaves like a mode of a harmonic oscillator. The mode energies can change by discrete amounts $\hbar \omega_k$, and these quanta may be identified as *photons*: they are the excitations of the electromagnetic field. The operators $\hat{a}_{\mathbf{k}, \lambda}^\dagger$ and $\hat{a}_{\mathbf{k}, \lambda}$ respectively create and destroy a photon in the mode $\mathbf{k}, \lambda$ and automatically include the bosonic symmetry of the multiparticle state.

2.2.2 Temporal modes

A full description of the wavefunction for a photon requires application of quantum field theory [32]. However for sufficiently narrowband photons it is possible to express pertinent properties by describing a photon via a wavepacket, an approach common in the literature on engineering nonlinear photon sources [33–37]. Our quantisation of the electromagnetic field provides a set of modes labelled by the wavevector $\mathbf{k}$, and for continuous wavevectors the sum in the Hamiltonian may be replaced by an integral. For now dropping the polarisation label λ , the Hamiltonian in vacuum generates the transformation $\hat{a}_{\mathbf{k}}^\dagger(t) = \hat{a}_{\mathbf{k}}^\dagger(0)e^{i\omega_{\mathbf{k}}t}$. To create an excitation that is localised in space at position $\mathbf{r}$ at time t , we define

$$\hat{a}^\dagger(\mathbf{r}, t) = \frac{1}{\sqrt{V}} \int d^3\mathbf{k} \hat{a}_{\mathbf{k}}^\dagger(t) e^{-i\mathbf{k} \cdot \mathbf{r}} \quad (2.30)$$

where V is the quantisation volume. This corresponds to taking an equally-weighted coherent superposition of plane waves $\mathbf{k}$ that are fully delocalised in space. We will always be considering wavepackets that have some finite spatial extent and are described by operators

$$\hat{\psi}^\dagger(\mathbf{r}, t) = \int d^3\mathbf{k} f(\mathbf{k}) \hat{a}_{\mathbf{k}}^\dagger(0) e^{-i(\mathbf{k} \cdot \mathbf{r} - \omega_{\mathbf{k}}t)}. \quad (2.31)$$

The normalised function $f(\mathbf{k})$ contains information about the spectral properties of the wavepacket. Time evolution of the wavepacket has been included using the transformation properties of the creation operators. Assuming propagation in the z direction so $k_x = k_y = 0, k_z = |\mathbf{k}|$, linear dispersion means $\omega_{\mathbf{k}}$ can be replaced by $\omega = c|\mathbf{k}|$ and we can relabel the operators in $\mathbf{k}$ as monochromatic operators $\hat{a}^\dagger(\omega)$. Together with the paraxial approximation, we can then write down the operator for the wavepacket as

$$\hat{\psi}^\dagger(z, t) = \int d\omega f(\omega) \hat{a}^\dagger(\omega) e^{-i\omega(\frac{z}{c} - t)}. \quad (2.32)$$

The operators $\hat{a}^\dagger(\omega)$ generate plane waves in z that are then weighted by the complex-valued spectral amplitude function $f(\omega)$ that satisfies the normalisation $\int d\omega |f(\omega)|^2 = 1$. This operator describes a wavepacket at time t after travelling a distance $z = ct$. We will describe temporal modes by the relative arrival times t_i of the centres of their wavepackets at some fixed reference position z . Hence we define broadband operators

$$\hat{A}^\dagger(t_i) = \int d\omega f(\omega) \hat{a}^\dagger(\omega) e^{i\omega t_i}. \quad (2.33)$$

If the phase of $f(\omega)$ is constant then the wavepacket is transform-limited: it has the shortest possible duration given the frequency content described by $f(\omega)$.

2.2.3 Linear optics

We may use linear optical components to manipulate states of light without changing the number of photons. Examples include mirrors, waveplates, beam splitters, and lenses, and their actions

are described by Hamiltonians that are bilinear in creation and annihilation operators [8]:

$$\hat{H}_{lin} = \sum_{ij} H_{ij} \hat{a}_i^\dagger \hat{a}_j, \quad H_{ij} = H_{ji}^*. \quad (2.34)$$

These commute with the total number operator $\hat{N} = \sum_i \hat{a}_i^\dagger \hat{a}_i$ and so conserve photon number. This expression may be interpreted as a sum over all processes where a single photon is removed from mode j , and then placed in some final mode i along with an amplitude H_{ij} . Hence each photon evolves independently of the others.

We earlier mentioned that we require a degree of freedom in which all particles are orthogonal in order to investigate distinguishability in some other property. Throughout this work we shall use linear multiport interferometers that provide photons with a set of orthogonal spatial modes or ‘ports’. The ports are all distinguishable from each other, and for some $m \times m$ multiport there are m orthogonal states corresponding to the inputs and m orthogonal states for the outputs [38].⁷ The coupled spatial modes are transverse to the direction of propagation, so we will treat evolution in these devices as a transformation on the port degree of freedom that leaves the time and z coordinates unchanged. We denote operators for the input ports by $\hat{a}_j^\dagger$, and for the output ports by $\hat{b}_k^\dagger$, where $j, k = 1, \dots, m$. Using units where $\hbar = 1$, the Schrödinger equation tells us that the evolution of a closed system under this Hamiltonian is described by the unitary operator $\hat{U} = e^{-i\hat{H}_{lin}}$ that preserves wavefunction normalisation. In the Heisenberg picture, the creation operators for the input state undergo the evolution

$$\hat{a}_j^\dagger \rightarrow \hat{U} \hat{a}_j^\dagger \hat{U}^\dagger = \sum_{k=1}^m U_{jk} \hat{b}_k^\dagger. \quad (2.35)$$

We have written the unitary transformation from input to output ports [20, 39–42]. Each input operator is transformed to a linear combination of output operators, with associated amplitudes U_{jk} that are the matrix elements of the unitary operator in this mode basis [43, 44]. Thanks to the field quantisation procedure of equation 2.26, this evolution of mode operators uses the same transformation matrix as when considering the classical fields where the output fields are linearly related to those at the inputs.

For single particle evolution, $P_{j \rightarrow k} = |U_{jk}|^2$ represents the probability of a particle in input port j ending in output port k . The matrix elements’ relative phases are important when considering interference, as we shall see in Section 2.3. Balanced multiports have the property that a particle starting in any input port has the same probability of ending in any output port, so $|U_{jk}| = 1/\sqrt{m}$. Examples are the *Fourier multiports* described by the matrices [20, 45, 46]

$$U_{jk}^{Fourier} = \frac{1}{\sqrt{m}} e^{i \frac{2\pi}{m} (j-1)(k-1)}. \quad (2.36)$$

In subsequent chapters we will encounter these matrices for $n = 2, 3, 4$. Multiport interferometers can be constructed using bulk optics, coupled fibres or integrated waveguides. We will

⁷The action of an interferometer may be treated as a rotation of the basis vectors associated with the m spatial modes that are coupled. Equivalently it may be defined as a transformation of m input modes to m output modes that are distinguished by the passage of time. We choose the latter to emphasise when evolution through an interferometer has occurred.

end up using bulk optic and fibre devices but it is the last approach that is the most appealing technologically. Any $m \times m$ unitary interferometer can be efficiently decomposed into an array of 2×2 beam splitters and phase shifters [47, 48]. The dream is to mimic the miniaturisation of silicon wafer technology in modern electronics: the fabrication of arrays of these components on an integrated platform, and with a degree of reconfigurability, could find applications in a wide range of photonic quantum technologies [49–51].

2.2.4 Polarisation and the Bloch sphere

When we quantised the field, the polarisation λ described the orientation of the electric field with respect to the direction of propagation in free space. Horizontal (H) and vertical (V) polarisations provide a basis for the associated two-dimensional Hilbert space and describe a *qubit*. It is worth noting that earlier we used the bosonic properties of photons when imposing the commutation relations, yet polarisation mimics the behaviour of a spin- $\frac{1}{2}$ particle. Whilst photons are spin-1 particles, being massless imposes constraints on the allowed spin projections. The helicity is the projection of spin onto the direction of motion, and there are two possibilities for a photon corresponding to left- and right-circular polarisations. These are states where the direction of the field precesses around the direction of propagation, and can be transformed to the $\{|H\rangle, |V\rangle\}$ basis [5, 52].

A general pure polarisation state is

$$|\psi\rangle = \cos \frac{\theta}{2} |H\rangle + e^{i\varphi} \sin \frac{\theta}{2} |V\rangle. \quad (2.37)$$

This can be represented as a point on the surface of a unit sphere by identifying θ, φ as the polar and azimuthal angles respectively (see Figure 2.2). This is called the *Bloch sphere representation*⁸ of a qubit and captures the space of all possible pure states in this two-dimensional Hilbert space. This picture can accommodate mixed states by writing some density matrix as

$$\rho = \frac{1}{2}(\mathbb{I} + \mathbf{r} \cdot \boldsymbol{\sigma}) \quad (2.38)$$

where $\mathbb{I}$ is the identity matrix, $\boldsymbol{\sigma} = (\sigma_x, \sigma_y, \sigma_z)$ is a vector of the Pauli matrices, and $\mathbf{r}$ is a *Bloch vector*. Its Cartesian coordinates are given by

$$\mathbf{r} = (r \sin \theta \cos \varphi, r \sin \theta \sin \varphi, r \cos \theta), \quad (2.39)$$

where the vector's squared length $r^2 = 2\mathcal{P} - 1$ describes the purity of the qubit. Pure qubits have Bloch vectors of unit length, and mixed states' vectors lie within the unit sphere. Orthogonal states have Bloch vectors that are on opposite sides of the Bloch sphere; for example we will usually denote $|H\rangle$ as the state at the North pole and $|V\rangle$ lying at the South pole.

Waveplates allow transformation between different polarisation states. These are disks of birefringent material that introduce a relative phase between horizontally and vertically polarised

⁸The polarisation of light may also be described by Stokes parameters or Jones vectors. The sphere used to visualise polarisation states using Stokes parameters is called the Poincaré sphere, but we will typically use the Bloch terminology familiar from quantum information.

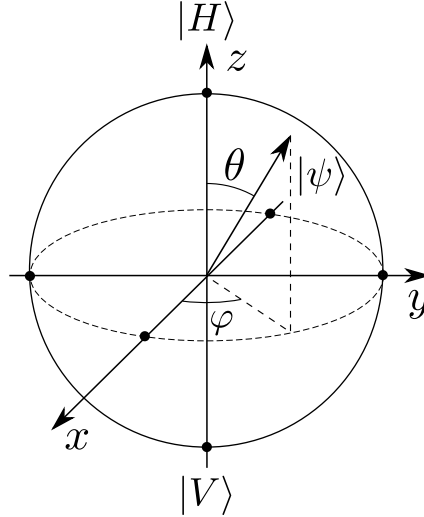


Figure 2.2: Representation of a pure qubit in the Bloch sphere. Mixedness can be described by a reduction in the length of the Bloch vector.

light: quarter- and half-wave plates introduce $\pi/2$ and π phases respectively. When oriented with their fast axis at some physical angle γ with respect to the horizontal, their action can be described by unitary matrices acting in the space spanned by $\{|H\rangle, |V\rangle\}$:

$$U_{HWP}(\gamma) = \begin{pmatrix} \cos 2\gamma & \sin 2\gamma \\ \sin 2\gamma & -\cos 2\gamma \end{pmatrix}, \quad U_{QWP}(\gamma) = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 - i \cos 2\gamma & -i \sin 2\gamma \\ -i \sin 2\gamma & 1 + i \cos 2\gamma \end{pmatrix}. \quad (2.40)$$

These can be interpreted as rotations of the Bloch sphere itself [53]. In discussions on state distinguishability we will be interested in behaviour resulting from relative orientation of qubit states and this Bloch picture will be very useful.

2.3 Interference

2.3.1 The superposition principle and path indistinguishability

A central tenet of quantum mechanics is the ability of a system to be in a *superposition*: a normalised linear combination of states is also a valid quantum state. This is automatically included in the definition of state vectors in a Hilbert space. We encountered this earlier when considering entangled states, linear optical evolution, and when constructing wavepackets from a superposition of plane waves. The superposition principle is deeply related to the idea of *indistinguishable paths* giving rise to a particular measurement outcome [16]. If there are multiple ways for an initial state to evolve and give rise to the same final detection, the system does not take just one of these paths individually, as might be expected from classical intuition, but rather evolves as a superposition of states for each path. As long as the paths are indistinguishable – there is in principle no measurement that can tell which path is being taken without destroying the superposition – final measurement outcomes are determined by the interplay of complex *probability amplitudes* associated with the paths: quantum interference [5, 54]. The coherence of the paths depends intimately on their indistinguishability [55]: information that

could in principle permit discrimination of the states in a superposition results in a collapse of the wavefunction and loss of coherence. The outcomes for the resulting mixed state then depend on classical probabilities associated with each of the paths⁹.

To capture this mathematically we use projective measurements and the Born rule to determine the probabilities of different measurement outcomes. Defining Hermitian projectors $\{\hat{\Pi}_i\}$ for some observable with outcomes i , the probability of an outcome P_i for an evolved quantum state $\hat{U}|\psi\rangle$ is

$$P_i = \langle\psi|\hat{U}^\dagger\hat{\Pi}_i\hat{U}|\psi\rangle. \quad (2.41)$$

The interference of amplitudes corresponding to the different possible paths is contained within this overlap, as is the effect of any distinguishability. This can also be extended to mixed states in order to capture any incoherence in state preparation:

$$P_i = \text{Tr}(\hat{U}\hat{\rho}\hat{U}^\dagger\hat{\Pi}_i), \quad (2.42)$$

where the trace is over all subsystems and degrees of freedom. The quantum interference of indistinguishable paths is at the very heart of quantum mechanics, and indeed most of this thesis is an investigation into how different types of distinguishability affect it.

2.3.2 Single-particle interference

The double slit experiment elegantly demonstrates the wave-particle duality of quantum particles. If single particles are repeatedly fired at a pair of slits and there is no way to tell which slit was passed, a fringe pattern in detection density develops on a screen some distance behind the slits (see Figure 2.3).

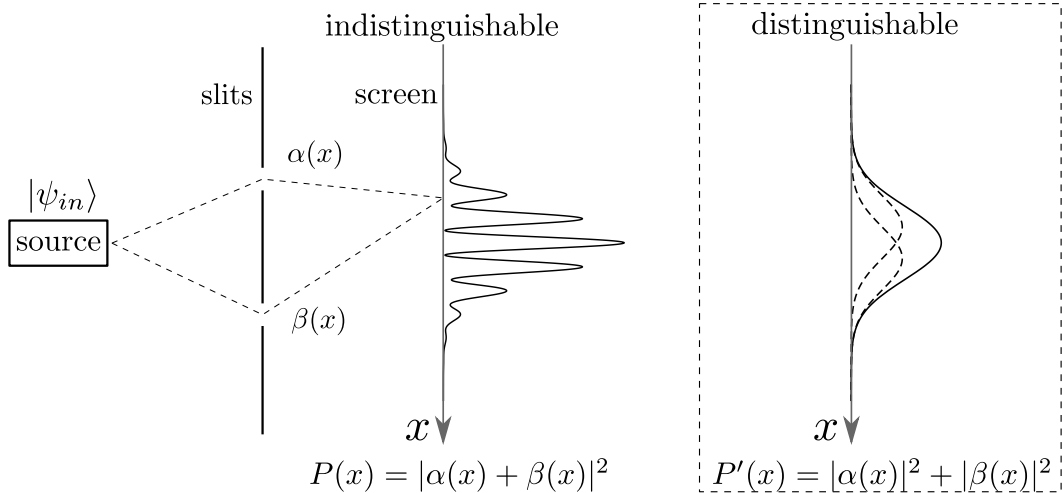


Figure 2.3: Single particles generated by a source pass through two slits that each impose Gaussian transverse modes and are then detected on a screen in the far-field. As long as the paths to position x indicated are indistinguishable, the probability of arriving at some position $P(x)$ is given by the squared modulus of the sum of the associated probability amplitudes. If the paths are distinguishable, the probability $P'(x)$ is given by the sum of the separate probabilities.

⁹Why probability amplitudes are complex numbers is a good question that we will not pursue further here, but see for example [56, 57].

Denoting the initial state of a particle from the source by $|\psi_{in}\rangle$ and describing evolution through the slits by $\hat{U}$, we can write the probability of detection at position x on the screen

$$\begin{aligned} P(x) &= \langle \psi_{in} | \hat{U}^\dagger \hat{\Pi}(x) \hat{U} | \psi_{in} \rangle \\ &= |\alpha(x) + \beta(x)|^2. \end{aligned} \tag{2.43}$$

The two paths corresponding to a detection at x have associated amplitudes $\alpha(x)$ and $\beta(x)$ from evolution under $\hat{U}$. The squared modulus of their sum determines arrival probabilities, and it is the cross-terms $\alpha^*(x)\beta(x)$, $\alpha(x)\beta^*(x)$ that describe interference and lead to behaviour outside a description based on just probabilities, given by $|\alpha(x)|^2 + |\beta(x)|^2$. The phase difference between the amplitudes varies across the screen and causes a fringe pattern that includes regions where the arrival probability is smaller or larger than if the paths are distinguishable. Thus we see that single-particle interference, or rather the interference of paths for a single particle, can here be described by a *wave* model. Superposing probability amplitudes gives the same pattern as if we had treated the quantum particle as a classical wave and measured intensities instead of discrete particles. This has been demonstrated for electrons [58], photons [59, 60], and even massive molecules [61].

Any attempt to gain information on which slit the particle took reduces path indistinguishability and results in loss of coherence. The interference visibility decreases as measurement results return to sums of classical probabilities [5, 62]. This trade-off is described by the inequality $\mathcal{D}^2 + \mathcal{V}^2 \leq 1$, where $\mathcal{D}$ describes the available amount of which-path information and $\mathcal{V}$ is the interference fringe visibility [63]. This captures the complementarity inherent in quantum mechanics: which-path information probes particle properties, at the expense of wave-like interference [64, 65]. Famously the concept of *quantum erasure* shows it is actually possible to recover interference if any such information is masked, even if this occurs after detection. An example of a “delayed-choice” eraser experiment replaces the two slits with two sources of entangled photon pairs [66]. One of the photons undergoes a wavelike interference test, whilst the other either heralds the presence of which-path information by flagging which source fired, or erases such information. Even if the pairs are detected at very different times, comparison after the experiment confirms that when which-path information is not available, a fringe pattern is observed. Conversely if the source can be identified, no interference occurs. Along with a number of other experiments and proposals [65–72], this highlighted how, despite the temptation at some point to ascribe classical properties to a propagating particle, a quantum treatment is always necessary.

2.3.3 Hong-Ou-Mandel interference

Adding another particle allows the bosonic nature of the electromagnetic field to manifest itself in quantum interference. Here we will review a famous counter-intuitive two-particle effect first demonstrated by Hong, Ou and Mandel (HOM) in 1987 [6]. Consider preparing two photons in separable indistinguishable states, and then injecting one into each input of a beam splitter. The input state is $|\psi_{in}\rangle = \hat{a}_1^\dagger \hat{a}_2^\dagger |0\rangle = |1, 1\rangle$ (see Figure 2.4a), but what happens at the outputs?

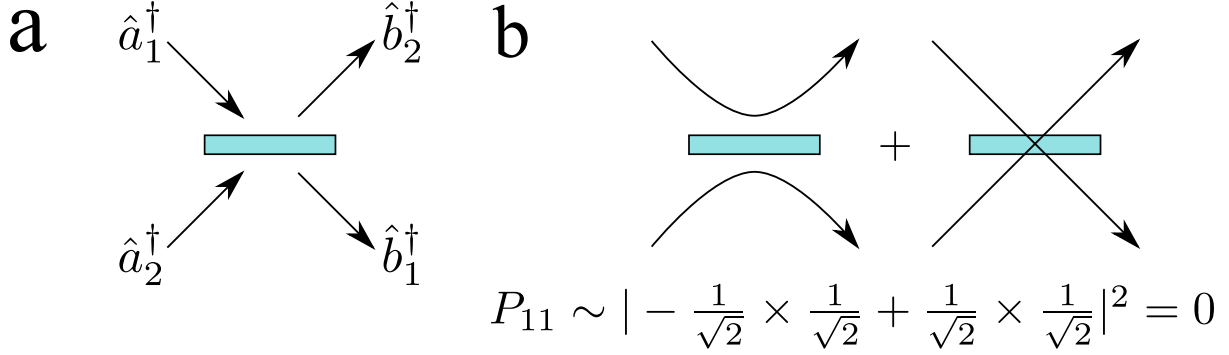


Figure 2.4: **a** Beam splitter geometry. **b** Indistinguishable paths for coincident detection interfere destructively for a balanced beam splitter so the associated probability P_{11} is zero.

A beam splitter mixes the two spatial modes and its Hamiltonian is [12, 29, 73]

$$\hat{H}_{BS} = i\theta \left(\hat{a}_1 \hat{a}_2^\dagger - \hat{a}_1^\dagger \hat{a}_2 \right). \quad (2.44)$$

This permits permutation of the port degree of freedom and has an associated unitary operator $\hat{U}_{BS} = e^{-i\hat{H}_{BS}}$. We can use this to transform creation operators as in equation 2.35 so

$$\begin{aligned} \hat{a}_i^\dagger &\rightarrow \hat{U}_{BS} \hat{a}_i^\dagger \hat{U}_{BS}^\dagger, & \hat{a}_1^\dagger &\rightarrow \cos \theta \hat{b}_1^\dagger + \sin \theta \hat{b}_2^\dagger \\ & & \hat{a}_2^\dagger &\rightarrow -\sin \theta \hat{b}_1^\dagger + \cos \theta \hat{b}_2^\dagger \end{aligned} \quad (2.45)$$

where the output port operators are labelled $\hat{b}_i$ to make the distinction from input ports, and we have used the Hadamard lemma to simplify the exponentials of operators¹⁰. This is precisely the transformation of operators discussed earlier, and is a representation of an $SU(2)$ transformation [40, 74]. For consistency with Figure 2.4a we now define the transmissivity $t = \cos \theta$ and reflectivity $r = \sin \theta$ so the unitary matrix representing the action of the beam splitter is

$$U_{BS} = \begin{pmatrix} t & r \\ -r & t \end{pmatrix}. \quad (2.46)$$

Energy conservation is satisfied since $t^2 + r^2 = 1$ and the π phase shift on reflection ensures unitarity [75]. For bulk beam splitters this phase shift arises from the Fresnel relations describing the field transformations, whilst for fibre devices it is the difference in phase velocities of the supported modes [76–79]. Applying the appropriate transformations to the input gives the output state

$$\begin{aligned} |\psi_{in}\rangle = \hat{a}_1^\dagger \hat{a}_2^\dagger |0\rangle &\rightarrow |\psi_{out}\rangle = \left(t\hat{b}_1^\dagger + r\hat{b}_2^\dagger \right) \left(-r\hat{b}_1^\dagger + t\hat{b}_2^\dagger \right) |0\rangle \\ &= \left(rt \left(-\hat{b}_1^\dagger \hat{b}_1^\dagger + \hat{b}_2^\dagger \hat{b}_2^\dagger \right) + t^2 \hat{b}_1^\dagger \hat{b}_2^\dagger - r^2 \hat{b}_2^\dagger \hat{b}_1^\dagger \right) |0\rangle. \end{aligned} \quad (2.47)$$

These four terms correspond to the different ways the photons can pass through the beam splitter: the two cases of one being transmitted and the other reflected, or both are transmitted,

¹⁰For operators $\hat{A}$ and $\hat{B}$: $e^{\hat{A}} \hat{B} e^{-\hat{A}} = \hat{B} + [\hat{A}, \hat{B}] + (1/2!)[\hat{A}, [\hat{A}, \hat{B}]] + (1/3!)[\hat{A}, [\hat{A}, [\hat{A}, \hat{B}]]] + \dots$

or both are reflected. For a balanced beam splitter, $t = r = 1/\sqrt{2}$ and then the output state is

$$\begin{aligned} |\psi_{out}\rangle &= \frac{1}{2} \left(-\hat{b}_1^\dagger \hat{b}_1^\dagger + \hat{b}_2^\dagger \hat{b}_2^\dagger + [\hat{b}_1^\dagger, \hat{b}_2^\dagger] \right) |0\rangle \\ &= \frac{1}{\sqrt{2}} (-|2, 0\rangle + |0, 2\rangle). \end{aligned} \quad (2.48)$$

In the last step we have used the commutation relation for bosons from equation 2.11. Amazingly we are left with a state where *the photons never exit in different ports*, something that goes entirely against classical intuition that they would do so half the time! Despite each photon evolving independently under a linear transformation, they end up being perfectly correlated. This *HOM interference* is a manifestation of the bosonic symmetry of the photons and, due to the phase shift on the beam splitter, of the perfect destructive interference of the two indistinguishable paths leading to coincidences (see Figure 2.4b).

2.3.4 The HOM dip and distinguishability

This bunching means that the coincidence probability for two photons in indistinguishable states $P_{11}^{indist} = |-1/2 + 1/2|^2 = 0$. If they instead have completely distinguishable states, no interference occurs and the coincidence probability is found by summing separate probabilities: 1/4 for each of both reflected or both transmitted means $P_{11}^{dist} = |-1/2|^2 + |1/2|^2 = 1/2$. What happens in the intermediate regime where the photons' states are only *partially* distinguishable? In other words there are degrees of freedom besides spatial port whose inclusion opens up the possibility of testing the partial exchange symmetry discussed in Section 2.1.4.

Assuming that the photons are in identical wavepackets characterised by some spectral amplitude $f(\omega)$, we label their temporal modes by the arrival times of the centres of the wavepackets t_1, t_2 . The input state is now $|\psi'_{in}\rangle = \hat{a}_1^\dagger(t_1)\hat{a}_2^\dagger(t_2)|0\rangle = |t_1, t_2\rangle$, where the ordering of kets denotes spatial mode (see Figure 2.5a). The beam splitter mixes spatial modes as before but leaves the temporal modes unchanged, so the total unitary evolution $\hat{U} = \hat{U}_{BS} \otimes \hat{\mathbb{I}}_{temp}$. Thus the transformation of operators from equation 2.45 can change the port a photon ends up in, but not its temporal mode:

$$\hat{a}_j(t_j) \rightarrow \sum_{k=1}^2 (U_{BS})_{jk} \hat{b}_k^\dagger(t_j). \quad (2.49)$$

The coincidence probability is now

$$\begin{aligned} P_{11}^{temp} &= \langle \psi'_{in} | \hat{U}^\dagger \hat{\Pi}_{11} \hat{U} | \psi'_{in} \rangle \\ &= \left[\frac{1}{2} \langle t_1, t_2 | - \frac{1}{2} \langle t_2, t_1 | \right] \left[\frac{1}{2} |t_1, t_2\rangle - \frac{1}{2} |t_2, t_1\rangle \right] \\ &= \frac{1}{2} \left(1 - |\langle t_1 | t_2 \rangle|^2 \right). \end{aligned} \quad (2.50)$$

The operator $\hat{\Pi}_{11} = \int d\tau_1 d\tau_2 |\tau_1, \tau_2\rangle \langle \tau_1, \tau_2|$ removes parts of the wavefunction outside the coincident subspace, and does not resolve the temporal degree of freedom. The ability of $\hat{U}_{BS}$ to exchange the spatial modes of a state probes precisely the partial exchange symmetry presented earlier [26], specifically: how similar is the state after swapping the ports but leaving all other

degrees of freedom unchanged? The coincidence probability has a dependence on the pairwise distinguishability $|\langle t_1 | t_2 \rangle|^2$ because such a symmetry test can in principle provide distinguishing information for the interfering paths of Figure 2.4b. When the arrival times are equal then $t_1 = t_2$ and the states are indistinguishable, resulting in complete destructive interference and $P_{11}^{indist} = 0$. If the wavepackets are temporally distinguishable then the overlap is zero and we recover $P_{11}^{dist} = 1/2$. In the intermediate regime, the degree of interference is controlled monotonically by the relative delay between the wavepackets and we observe a *HOM dip* (see Figure 2.5b) whose shape depends on the photons' spectra $f(\omega)$ [80–82].

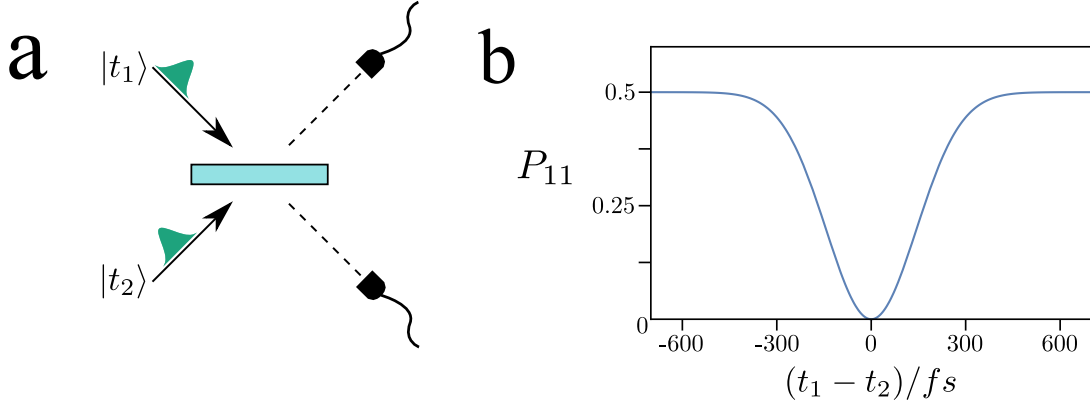


Figure 2.5: **a** Photons in identical wavepackets, delayed by times t_1 and t_2 , impinging on a balanced beam splitter whose outputs are monitored by detectors. **b** A typical HOM dip showing the probability of coincident detection as a function of the relative time delay between the photons. Here we have simulated Gaussian wavepackets with central wavelength 830nm and 3nm bandwidth, and the delay is given in femtoseconds. The probability of detecting two photons in a single output port will instead exhibit a peak due to this bunching effect [80].

In practice a HOM dip is obtained by scanning a delay stage on one of the arms before a beam splitter and recording output coincidences using photodetectors connected to a counting system. The dip visibility is given by $V = (P_{11}^{dist} - P_{11}^{indist})/P_{11}^{dist} = (N_{max} - N_{min})/N_{max}$, where N_i are the number of coincidences for some relative delay, and $V = 1$ for perfect quantum interference. Any distinguishability in degrees of freedom besides temporal mode will decrease interference strength and so reduce the visibility of the dip in coincidence counts. Hence here V is the pairwise distinguishability of the states' other modes and is ubiquitous as a measure of the indistinguishability of photons and as a benchmark for photon sources [83, 84].

The average number of photons arriving at a single detector remains constant whilst the relative delay is adjusted because only one path can contribute to the measurement of a single photon in this setup. This is true for bosons, fermions, and completely distinguishable particles: single-particle observables are unchanged by multiparticle interference, and they depend only on the elements of the linear evolution U_{jk} [20]. It can be shown that for classical fields with randomised phases that wash out single-particle (classical) interference, the maximum visibility observed in second-order intensity correlations for the same setup is 50% [85–89]. Hence an observation of $V > 0.5$ is often used to signify non-classical interference.

It is common to define a characteristic *coherence time* τ_c , and an associated *coherence length* $l_c = c\tau_c$, beyond which coherence due to indistinguishability of the paths is lost and interference disappears. This can be determined from wavepackets' spectra $f(\omega)$ and in most experiments

τ_c is many times smaller than the resolution of photodetectors, yet a dip can still be observed at the sub-picosecond scale. The temporal distinguishability that in principle is measurable but in practice is unresolved still plays a central role. It is worth highlighting here that it is the paths that interfere, not the photons themselves, and it is simply a consequence of this setup that distinguishability is tuned by relative arrival times at the beam splitter. Indeed HOM interference has been demonstrated in situations where the photons' wavepackets do not meet in space or time yet still exhibit destructive interference due to path indistinguishability [90].

There is a veritable plethora of experiments demonstrating the non-locality of two-photon interference, the use of entanglement to circumvent requirements on state indistinguishability, and the ability to restore coherence despite distinguishability prior to detection [85, 89, 91–98]. Besides being of fundamental interest and of use in benchmarking photon sources, HOM interference has also shown the enhanced sensitivity to phase differences of $N00N$ states, of which the output state of equation 2.48 is the example for $N = 2$ [94, 99]. It is also vital in routes to linear optical quantum computing: in the Knill-Laflamme-Milburn scheme, the phase difference in the output state, together with measurement induced nonlinearities and feed-forward logic, allows realisation of a heralded universal two-qubit controlled-Z gate [7, 8].

2.3.5 Extending to more particles

Things have got a lot more interesting by adding just one extra particle and extra degrees of freedom, so we would hope this trend continues. Generalisations of HOM bunching have been shown for four photons [100] and six photons [101] where the enhancement cannot be described by pairwise bunching factors alone. Furthermore suppression of certain output events in scattering experiments has been extended to more than two particles [41, 102–104]. In general these two features of HOM-like interference are different aspects of the underlying dynamics: the bunching arises from the symmetry property of bosons whilst the suppressions are a consequence of the phases of the interfering paths [20]. The two-photon case is special because the phase shift of the $SU(2)$ beam splitter reduces coincidences whilst simultaneously increasing the occurrence of bosonic bunchings; in fact as system size increases it is the coherent effects that dominate statistics [38]. HOM-like tests of indistinguishability have been extended to multiple particles, and the non-monotonicity of the quantum to classical transition has been investigated for larger systems [84, 105–107].

Our motivation for turning to larger systems is to investigate how partial exchange symmetry and multiparticle distinguishability affect large-scale interference. The general approach will be to prepare photons in separable states, evolve them in a linear network that mixes the spatial modes, and count statistics at the outputs whilst controlling distinguishing degrees of freedom in order to map out the interference landscape (see Figure 2.6). There is a wealth of theoretical work describing these systems in both first- and second-quantised formalisms, respectively [108–111] and [20, 38, 41, 112–118].

Before considering extensions, it is instructive to revisit the HOM coincidence probability of equation 2.50. Imagine preparing a separable state of a pair of photons $|\psi_{in}\rangle = |a_1, b_2\rangle$ where numbers label the inputs of an interferometer described by U_{jk} and a, b are some label degree

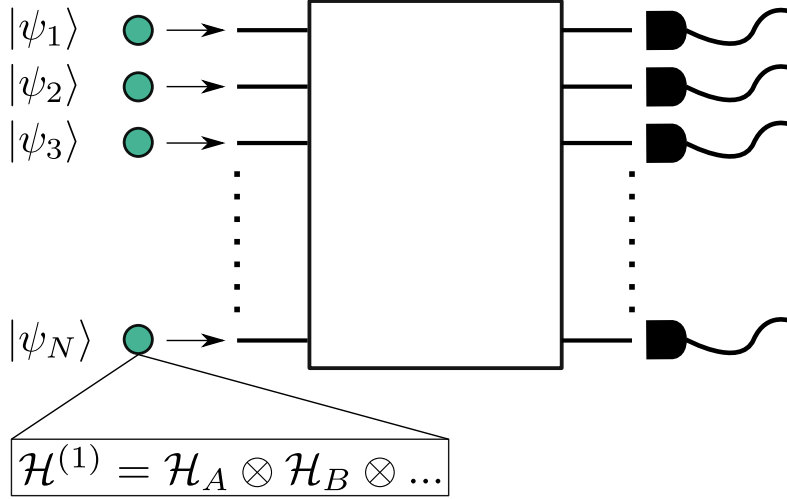


Figure 2.6: Our approach for investigating the effect of partial exchange symmetry on interference. A separable state of N photons, each with single particle Hilbert space $\mathcal{H}^{(1)}$, is injected into an interferometer that mixes spatial modes, and (possibly number-resolved) counting statistics are collected at the outputs.

of freedom unchanged by evolution. The state after the interferometer is then

$$\begin{aligned} |\psi_{out}\rangle &= \hat{U} |\psi_{in}\rangle \\ &= U_{11}U_{22} |a_1, b_2\rangle + U_{11}U_{21} |a_1, b_1\rangle + U_{12}U_{22} |a_2, b_2\rangle + U_{12}U_{21} |a_2, b_1\rangle, \end{aligned} \quad (2.51)$$

where each bosonic operator has acquired an amplitude from unitary transformation. The probability of observing a coincidence at the output ports is then

$$\begin{aligned} P_{11} &= \langle \psi_{out} | \hat{\Pi}_{11} | \psi_{out} \rangle \\ &= [U_{11}^* U_{22}^* \langle a_1, b_2 | + U_{12}^* U_{21}^* \langle a_2, b_1 |] [U_{11} U_{22} |a_1, b_2\rangle + U_{12} U_{21} |a_2, b_1\rangle] \\ &= \left(|U_{11} U_{22}|^2 + |U_{12} U_{21}|^2 \right) + |\langle a|b\rangle|^2 \times (U_{11} U_{22} U_{12}^* U_{21}^* + U_{11}^* U_{22}^* U_{12} U_{21}) \\ &= \text{perm}(U * U^*) + |\langle a|b\rangle|^2 \times \text{perm}(U * U_{(2,1),\mathbb{I}}^*). \end{aligned} \quad (2.52)$$

As earlier the action of $\hat{\Pi}_{11}$ is to remove parts of the wavefunction outside the coincident subspace without resolving the label degree of freedom. perm is the *matrix permanent*, defined in the same way as a determinant but with no alternating sign. The Hadamard product is denoted by $*$ and corresponds to the elementwise product of matrix elements, and $U_{(2,1),\mathbb{I}}$ corresponds to the unitary scattering matrix with its rows swapped. The first permanent captures the probabilistic scattering of single particles. The dependence of the second term on the pairwise distinguishability $|\langle a|b\rangle|^2$ arises from precisely the partial exchange symmetry mentioned in Section 2.1.4. The two terms may therefore be associated with elements of the symmetric group S_2 : the first corresponds to the identity element where there is no exchange contribution and particles scatter probabilistically; the second corresponds to the transposition $\sigma = (2, 1)$, and captures the distinguishability contribution associated with the partial exchange symmetry under such a permutation. Using the balanced beam splitter unitary of equation 2.46, we recover the coincidence probability in terms of temporal delays from equation 2.50.

What do scattering probabilities look like for larger systems? The procedure above could be extended to multiple particles and the distinguishability dependence derived by numerous applications of the commutation relation $[\hat{a}_{|a_i\rangle}, \hat{a}_{|b_j\rangle}^\dagger] = \delta_{ij} \langle a|b \rangle$. These calculations are condensed in the formalism developed by Tichy [41, 112]. The configuration of particles at the inputs of some m -mode interferometer is described by the list $\vec{r} = (r_1, \dots, r_m)$ where $0 \leq r_j \leq N$ is the number of particles in port j , and $\sum_j r_j = N$ is the total number of particles. Another list $\vec{s} = (s_1, \dots, s_m)$ describes the output configuration of interest in the same way. The *mode assignment list* is $\vec{d}(\vec{r}) = (d_1, \dots, d_N)$, where d_k denotes the input port of the k th particle, and is defined similarly for $\vec{d}(\vec{s})$. Given an $m \times m$ unitary scattering matrix U that describes the linear interferometer, we also define the effective $N \times N$ scattering matrix $M = U_{\vec{d}(\vec{r}), \vec{d}(\vec{s})}$ as a submatrix of U with row and column multiplicities determined respectively by $\vec{d}(\vec{r})$ and $\vec{d}(\vec{s})$. For example, if $\vec{r} = (1, 0, 2)$ then $\vec{d}(\vec{r}) = (1, 3, 3)$, and if $\vec{s} = (3, 0, 0)$ then $\vec{d}(\vec{s}) = (1, 1, 1)$, so M is the submatrix of U comprising the first row once and the third row twice, and the first column three times. This accounts for the fact that unoccupied input and output modes do not contribute to interfering paths, and also that multiple occupation of a mode is accompanied by additional amplitude factors. Finally for particles in pure states $|\phi_j\rangle$ injected into port j , the $N \times N$ Hermitian distinguishability matrix is

$$\mathcal{S}_{j,k} = \langle \phi_{\vec{d}_j(\vec{r})} | \phi_{\vec{d}_k(\vec{r})} \rangle \quad (2.53)$$

where $\mathcal{S}_{j,j} = 1$. This encodes all distinguishing information for interfering particles in degrees of freedom other than their port mode, and which we assume are unchanged on evolution. For full indistinguishability all elements are unity, whilst for full distinguishability it reduces to the $N \times N$ identity matrix. Intermediate cases will be the topic of later chapters but there are at most $(N-1)^2$ independent distinguishing parameters (see Section 4.1.1). Putting all this together means the probability of some scattering event is given by [112]

$$P(\vec{r}, \vec{s}, \mathcal{S}, U) = \frac{1}{\prod_j r_j! s_j!} \sum_{\sigma \in S_N} \left[\prod_{j=1}^N \mathcal{S}_{j, \sigma(j)} \right] \text{perm}(M * M_{\sigma, \mathbb{I}}^*). \quad (2.54)$$

σ are elements of the symmetric group S_N and $\sigma(j)$ denotes the number j is mapped to under that element. $M_{\sigma, \mathbb{I}}^*$ has had its rows permuted according to σ . The permanent appears again due to the bosonic symmetry of the particles (equation 2.6) that means all contributions have the same sign. Fermionic particle statistics are instead described by the determinant. This sum of products of matrix elements essentially captures the interference of all the different paths, related by permutations, that contribute to the same output configuration of particles. These are then weighted by elements of the distinguishability matrix that contain information on the discriminating ability inherent in the wavefunction's partial exchange symmetry under permutation σ . The order of exchange contribution is obvious from the number of overlaps in the weighting and $\sigma = \mathbb{I}$ is associated with the single particles scattering probabilistically.

For indistinguishable states this expression reduces to $P_{indist} \sim |\text{perm}(M)|^2$. If we assume that each input port is at most singly occupied, that the size of the interferometer is much larger than the number of particles $m \gg N^2$, and that the scattering unitary is drawn randomly from

the Haar measure, then sampling from this probability distribution is thought to be hard for a classical computer [10, 119]. This problem of *boson sampling* is a candidate for demonstrating a quantum advantage without requiring a universal quantum computer: the scattering of photons is naturally described by matrix permanents. Recent years have seen rapid progress in experimental demonstrations of this task [50, 120–128]. However the requirements for doing something that cannot be simulated by a classical computer have been raised by the development of approximation algorithms efficient under certain conditions [113, 115, 129–134]. Furthermore even verifying that an experiment is actually doing boson sampling becomes difficult for large systems, though various benchmarks have been proposed [135–139]. The task has also found applications in the simulation of molecular vibronic spectra [140, 141].

The scattering of photons in fully distinguishable states is described by $P_{dist} \sim \text{perm}|M|^2$ that can be efficiently approximated classically [119]. What then is the route in computational complexity between approximating the quantum and classical cases? Varying the degree of distinguishability will lead to weighted combinations of contributions of the form P^{indist} and P^{dist} , necessitating more calculations but potentially a relaxation of precision requirements because higher-order terms are polynomial in the overlap magnitude, which is less than unity when there is some distinguishability [142]. Whilst the transition from quantum interference to purely probabilistic statistics is monotonic in distinguishability for HOM, this is generally not the case in multiparticle systems: different orders of exchange contributions compete as their properties are tuned, and we shall encounter an example in the next chapter [107, 143].

2.4 Summary

In this chapter we have developed a modal description of quantum states of light and established the relationship between partial exchange symmetry of the wavefunction and distinguishability. We linked this to the path indistinguishability at the heart of quantum interference, showing how the wavefunction captures what is in principle knowable about a system, and in the process reviewed some examples of one- and two-particle interference.

The formalism developed to treat multiparticle interference will form the basis of the next two chapters where we investigate what new distinguishing parameters arise for three- and four-particle interference. In Chapter 6 we will revisit the first quantised formalism to get a handle on how multiparticle distinguishability shows up in the symmetrised wavefunctions for different degrees of freedom.

Three-photon interference

In this chapter we investigate new features of distinguishability that arise in the interference of three photons in separable states. Discussions of the partial exchange symmetry of three particles reveal that four parameters are required to describe their distinguishability. One of these is a phase that embodies a collective property not captured by pairwise distinguishabilities. We use photon sources based on nonlinear optical processes to perform three-photon interference experiments that demonstrate the rich structure of multiparticle interference. In particular, we fully map out the temporal dependence of the interference landscape and then, with the addition of polarisation, first tune and then isolate the phase-dependent three-photon contribution to scattering statistics.

The experiments performed using the four-wave mixing source (results in Sections 3.5 and 3.6) have been published in [144] and were performed in collaboration with Adrian Menssen (AJM). AJM conceived the state preparation to isolate the new distinguishing phase and set up the SFWM source. The measurements, simulations and analyses in those sections were performed by us together. All other results presented using the parametric down-conversion source were obtained by me.

3.1 Distinguishability of three particles

3.1.1 Collective distinguishability and the triad phase

What parameters are needed to fully describe the distinguishability of three particles in separable states? If we label their internal states by a, b, c , and as earlier include an auxiliary degree of freedom such as position in which they are all orthogonal, the overall state is

$$\left| \psi_{123}^{abc} \right\rangle = |a, b, c\rangle. \quad (3.1)$$

Ordering corresponds to the mutually orthogonal spatial modes numbered 1–3. We now write the complex scalar products of states in modulus-argument form: $\langle i | j \rangle = r_{ij} e^{i\varphi_{ab}}$. The modulus r_{ij} is zero for distinguishable states and one for indistinguishable states, and the argument φ_{ij} describes relative orientation in the relevant Hilbert space. The partial exchange symmetry under a binary swap of positions results in precisely the pairwise distinguishabilities encountered

earlier that were shown to control HOM interference strength. For three particles there are three such swaps in S_3 , leading to the three real numbers $r_{ab}^2, r_{ac}^2, r_{bc}^2$. The argument φ_{ij} does not affect pairwise distinguishability because it can be transformed away by including a physically irrelevant global phase factor on a single particle state, such as $|i\rangle \rightarrow e^{i\varphi_{ij}} |i\rangle$. Equivalently this can be interpreted as a rotation of the basis spanning the relevant Hilbert space such that the corresponding vectors have a real overlap.¹

S_3 has two other elements corresponding to permutations of all the particles that result in the states $|\psi_{231}^{abc}\rangle = |c, a, b\rangle$ and $|\psi_{312}^{abc}\rangle = |b, c, a\rangle$. Comparing the initial state to the first of these fully permuted ones yields

$$\begin{aligned} \langle \psi_{231}^{abc} | \psi_{123}^{abc} \rangle &= \langle c, a, b | a, b, c \rangle \\ &= \langle c | a \rangle \langle a | b \rangle \langle b | c \rangle \\ &= r_{ab} r_{bc} r_{ac} \times e^{i\varphi_{abc}}, \end{aligned} \tag{3.2}$$

where we have defined the *triad phase* $\varphi_{abc} = \text{Arg}(\langle a | b \rangle \langle b | c \rangle \langle c | a \rangle) = \varphi_{ab} + \varphi_{bc} + \varphi_{ca}$. This is a collective distinguishing parameter that is invariant under global phases applied to any individual state and is basis-independent. The overlap of the initial state with the other fully permuted one gives the complex conjugate of these overlaps, and so a phase factor $e^{-i\varphi_{abc}}$.

This quantity has a formal similarity to geometric phases acquired by a quantum system evolving adiabatically and whose value depends on the parameter space geometry of the underlying Hamiltonian [146]. The origin of such phases is distinct from dynamical phases resulting from, for example, path length differences in an interferometer, but like other relative phases they are observable in interference experiments [147–155]. These arguments of products of state overlaps are known as *Bargmann invariants* and are ubiquitous in the study of geometric phases [156–161].

When the degree of freedom involved is polarisation, the geometric phase acquired by a quantum state after some cyclic trajectory in parameter space is called the Pancharatnam phase [162]. If an initial polarisation state $|a\rangle$ is rotated in the Poincaré sphere along $|a\rangle \rightarrow |b\rangle \rightarrow |c\rangle \rightarrow |a\rangle$, the final state will have a physically observable phase. Defining the relative phase between two quantum states as $\Phi_{ij} = \text{Arg} \langle i | j \rangle$, the overall phase accumulated after tracing this path is given by the sum of the associated relative phases [163]:

$$\Phi_{abc} = \text{Arg} \langle a | b \rangle + \text{Arg} \langle b | c \rangle + \text{Arg} \langle c | a \rangle = \text{Arg}(\langle a | b \rangle \langle b | c \rangle \langle c | a \rangle). \tag{3.3}$$

For polarisation this geometric phase is given by half the solid angle Ω enclosed by the geodesic triangle defined by those states: $\Phi_{abc} = \frac{\Omega}{2} = \text{Arg}(\langle a | b \rangle \langle b | c \rangle \langle c | a \rangle)$ in a right-handed frame². It is invariant under rotation of the polarisation basis (see Figure 3.1) [164]. Hence for three particles in separable qubit states, the triad phase is the same as the geometric phase acquired by a single particle traversing a path between those three states.

¹The geodesic distance between two pure states is sometimes given by the Hilbert space angle between them, defined by $\cos \theta = |\langle a | b \rangle|$ [145].

²Solid angles are conventionally taken to be positive if the trajectory is traversed in the anti-clockwise sense.

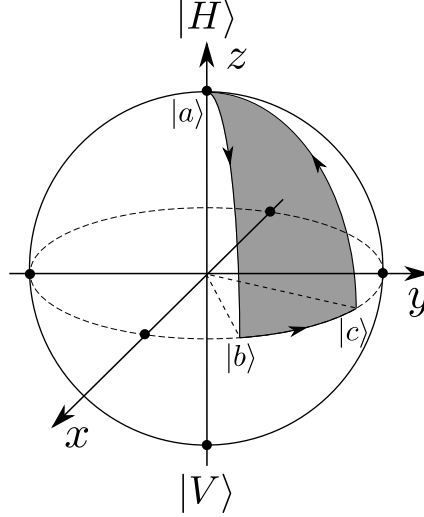


Figure 3.1: Geometric interpretation of the Pancharatnam phase. If some initial polarisation state $|a\rangle$ is rotated along the path shown, it acquires a geometric phase given by half the solid angle subtended by the shaded geodesic triangle. The same parameter quantifies the collective distinguishability of three particles prepared in qubit states $|a\rangle, |b\rangle, |c\rangle$.

3.1.2 Accessing the triad phase

How can we probe this new distinguishing parameter in an interference experiment? As before we will use permutations of orthogonal port modes to test the wavefunction symmetry. In order to access all possible distinguishabilities for three states – here four parameters – we assume, without loss of generality, a three-dimensional internal Hilbert space or ‘qutrit’. The three most general pure states in such a space are [145, 161, 165]:

$$\begin{aligned} |a\rangle &= |\mathbf{0}\rangle, \\ |b\rangle &= \cos \alpha |\mathbf{0}\rangle + \sin \alpha |\mathbf{1}\rangle, \\ |c\rangle &= \cos \gamma (\cos \beta |\mathbf{0}\rangle + e^{i\delta} \sin \beta |\mathbf{1}\rangle) + \sin \gamma |\mathbf{2}\rangle, \end{aligned} \tag{3.4}$$

where $\{|\mathbf{0}\rangle, |\mathbf{1}\rangle, |\mathbf{2}\rangle\}$ define an orthogonal basis for the qutrit space, and we have emboldened the numbers to make it clear these are not photon number states. The four real parameters $\alpha, \beta, \gamma, \delta$ control the overlaps of the states. Here we have assumed that $\langle a|b\rangle, \langle a|c\rangle \in \mathbb{R}$ and $\langle b|c\rangle \in \mathbb{C}$. γ describes the extent to which $|c\rangle$ occupies the qubit space $\{|\mathbf{0}\rangle, |\mathbf{1}\rangle\}$, with the rest residing in $|\mathbf{2}\rangle$. If $\gamma = 0$, the three states live entirely in a qubit space and then the distinguishing parameters are $r_{ab} = \cos \alpha, r_{ac} = \cos \beta$ and $\langle b|c\rangle = \cos \alpha \cos \beta + e^{i\delta} \sin \alpha \sin \beta$. Then a choice of δ fixes the final modulus r_{bc} and simultaneously defines the complex number $\langle b|c\rangle$. This can be appreciated by noting that enforcing specific pairwise overlap magnitudes for three vectors in the Bloch sphere automatically determines the enclosed solid angle. Hence the triad phase is not a free parameter for three qubits. On the other hand if γ is a free parameter, a qutrit space is accessible and the triad phase can be tuned independently. However its value is still constrained by the values of $0 \leq r_{ab}, r_{ac} \leq 1$. For the above states, the triad phase $\varphi_{abc} = \text{Arg}(\cos \alpha \cos \beta + e^{i\delta} \sin \alpha \sin \beta)$. The ability to fully tune the triad phase is only possible for certain values of overlap magnitudes, as will be seen in Section 3.6.

The addition of another photon introduces a new type of collective distinguishing parameter that goes beyond a pairwise description. There have already been some three photon interference experiments probing distinguishability but none have accessed this triad phase [108, 111, 166–168]. In the following sections we will use the temporal and polarisation degrees of freedom to give physical meaning to this parameter and to explore the new interference landscape.

3.2 Generating three photons

3.2.1 Single photon sources

The ability to generate a single photon Fock state is one of the great challenges facing quantum opticians. An ideal source would provide an on-demand stream of single photons that are each in the same indistinguishable pure state. Unfortunately some of the easiest states of light to generate in the lab do not have these number statistics: vacuum $|0\rangle$ and displaced vacuum $|\alpha\rangle = \hat{D}(\alpha)|0\rangle$, where the displacement operator $\hat{D}(\alpha) = \exp(\alpha\hat{a}^\dagger - \alpha^*\hat{a})$. The latter are coherent states generated by a laser and have a Poissonian number distribution. There are a host of approaches to building a single photon source [169], ranging from trapped single atoms, ions and molecules [170–177], to vacancy centres in diamond [178–184]. Semiconductor quantum dots acting as artificial atoms have also shown promise [185–188], with recent developments leading to impressive generation rates of indistinguishable photons and, once demultiplexed, demonstrations of boson sampling [125, 126, 189–192].

In this thesis we generate single photons using the nonlinear processes of spontaneous parametric down-conversion (SPDC) in bulk crystals and spontaneous four-wave mixing (SFWM) in a silica chip. The general principle is to probabilistically convert pump photons (one for SPDC, two for SFWM) simultaneously into a pair of daughter photons [193]. The detection of one of the daughter photons can then be used to *herald* the presence of the other. SPDC sources have been a workhorse of quantum optics since their role in early realisations of many of the interference effects described earlier, as well as a source of entangled photon pairs [194, 195]. They are still the most widespread platform for proof-of-principle experiments because they generally do not require cryogenic conditions and often comprise widely available optical components. They are ubiquitous in demonstrations of quantum computing protocols [196–199] and their development is still an active area of research, including efforts to integrate them, generate larger Fock states, and manipulate the entanglement of the generated pairs [128, 200, 200–205].

3.2.2 Nonlinear optics

The application of an electric field to a dielectric material can shift the position of its electrons and so induces electric dipole moments. The macroscopic strength of these dipoles is described by the polarisation density $\mathbf{P}(t) = -n_d e \mathbf{r}(t)$, where n_d is the electron density, e is the electric charge, and $\mathbf{r}(t)$ is the electrons' displacement. For weak fields the induced polarisation is linear in the applied field so $\mathbf{P}(t) = \epsilon_0 \chi^{(1)} \mathbf{E}(t)$, where $\chi^{(1)}$ is the linear response. This describes the effect of the refractive index $n_r = \sqrt{1 + \chi^{(1)}}$ on the speed of propagation. The advent of the first laser in 1960 provided a route to coherently drive a medium at high enough intensities for the

nonlinear mixing of fields to occur [206]. The polarisation response now contains higher-order terms in the applied field [207]:

$$\mathbf{P}(t) = \varepsilon_0 \left(\chi^{(1)} \mathbf{E}(t) + \chi^{(2)} \mathbf{E}^2(t) + \chi^{(3)} \mathbf{E}^3(t) + \dots \right), \quad (3.5)$$

where $\chi^{(n)}$ is the n th order electric susceptibility. For $\chi^{(2)}$ to be non-zero, the driven material must not have inversion symmetry. It can be shown from Maxwell's equations that these nonlinear terms in the polarisation act as source terms in the wave equation, allowing the generation of new frequencies. Physically this is due to large-scale coherent oscillations of the induced dipoles at harmonics of the applied field. Here we concentrate on the scalar response of the material to scalar fields.

Superposing two waves in a second-order nonlinear material opens up the possibility of generating new fields. Consider an electric field comprising two plane waves with frequencies ω_1 and ω_2 given by

$$\begin{aligned} E_{tot}(t) &= E_1(t) + E_2(t) \\ &= E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + \text{c.c.} \end{aligned} \quad (3.6)$$

where we have expanded out the positive- and negative-frequency components of the fields. The induced second-order polarisation response $P^{(2)} = \varepsilon_0 \chi^{(2)} E_{tot}^2(t)$ is then is given by

$$\begin{aligned} P^{(2)}(t) &= \varepsilon_0 \chi^{(2)} \left[\left(2|E_1|^2 + 2|E_2|^2 \right) + \left(E_1^2 e^{-i2\omega_1 t} + E_2^2 e^{-i2\omega_2 t} + \text{c.c.} \right) \right. \\ &\quad \left. + \left(2E_1 E_2 e^{-i(\omega_1 + \omega_2)t} + \text{c.c.} \right) + \left(2E_1 E_2^* e^{-i(\omega_1 - \omega_2)t} + \text{c.c.} \right) \right]. \end{aligned} \quad (3.7)$$

The first term in parentheses is called optical rectification and is independent of frequency. This arises because electrons in a material lacking inversion symmetry will assume some mean non-zero displacement in the presence of an applied field, and so generate a DC field. The second term contains components oscillating at frequencies $2\omega_1$ and $2\omega_2$. These can produce new fields at those frequencies in a process called second-harmonic generation (SHG) that was first demonstrated just a year after Maiman's development of the laser [208]. The third term gives rise to sum-frequency generation, and the last term results in difference-frequency generation (DFG) (see Figure 3.2). In DFG the 'pump' field at ω_1 mixes with the 'signal' field at ω_2 to produce a new 'idler' field at frequency $\omega_3 = \omega_1 - \omega_2$. Due to energy conservation this also parametrically amplifies the lower-frequency signal field.

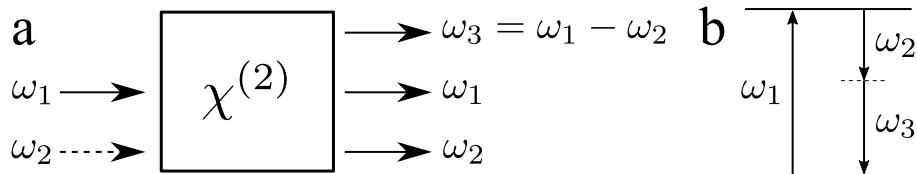


Figure 3.2: **a** Difference-frequency generation in a second-order nonlinear medium. Simultaneous application of fields at ω_1 and ω_2 can, with suitable phase-matching, generate a field at the new frequency $\omega_3 = \omega_1 - \omega_2$. At the same time the signal field at ω_2 is parametrically amplified. In the limit of vacuum signal field (dashed line), spontaneous parametric down-conversion can generate excitations at ω_2 and ω_3 . **b** Energy level diagram showing the off-resonant transitions with generated fields labelled. Adapted from [207].

A crucial requirement for efficient generation of a new field is *phase-matching*: the driving field should maintain a fixed phase relationship with respect to the material's polarisation in order to maximise the directed coherent emission of the collection of dipoles. This generally restricts the possible frequencies produced and is captured by insisting that the wavevector mismatch $\Delta k = 0$. For SHG this becomes $\Delta k = 2k(\omega_1) - k(2\omega_1) = 0$, where $k(\omega_j) = n(\omega_j)\omega_j/c$, and the refractive index $n(\omega)$ can have polarisation dependence. On the other hand, for DFG this is stated as $\Delta k' = k(\omega_1) - k(\omega_2) - k(\omega_1 - \omega_2)$.

3.2.3 Spontaneous parametric down-conversion

If the signal field is attenuated down to vacuum level then classically we would expect no DFG to occur. However in the quantum world the vacuum is not empty: there are fluctuations. With judicious choice of pump and material, these can stimulate emission into signal and idler modes by spontaneous parametric down-conversion (SPDC).

We want to find the Hamiltonian for the DFG process and then quantise it [81, 209, 210]. For a dielectric medium exposed to an electric field, the displacement field and energy density are given respectively by

$$\mathbf{D}(\mathbf{r}, t) = \varepsilon_0 \mathbf{E}(\mathbf{r}, t) + \mathbf{P}(\mathbf{r}, t), \quad U(\mathbf{r}, t) = \frac{1}{2} \mathbf{D}(\mathbf{r}, t) \cdot \mathbf{E}(\mathbf{r}, t), \quad (3.8)$$

where the spatial dependence is included explicitly. We now denote the frequencies of the pump, signal and idler fields by ω_p, ω_s and ω_i respectively to make identification of terms easier. Additionally the fields are now written in terms of their positive and negative frequency components that vary quickly in time:

$$E_j(\mathbf{r}, t) = E_j^+(\mathbf{r}, t) + E_j^-(\mathbf{r}, t), \quad E_j^-(\mathbf{r}, t) = \left(E_j^+(\mathbf{r}, t)\right)^* \quad (3.9)$$

with $j = p, s, i$. The total electric field is $E_{tot}(\mathbf{r}, t) = E_p(\mathbf{r}, t) + E_s(\mathbf{r}, t) + E_i(\mathbf{r}, t)$. The second-order polarisation response can be determined by multiplying out terms as in equation 3.7 and substitution into equations 3.8 gives the energy density $U(\mathbf{r}, t)$. This will contain terms oscillating at many different frequencies, but we now assume that the frequency of the pump is large compared to those of the signal and idler fields. All terms besides DFG oscillate very quickly and will average out when integrated over any timescales of interest later, so can be ignored. The interaction Hamiltonian is obtained by integrating the remaining terms in the energy density over the volume V of the nonlinear medium:

$$H_I(t) = \varepsilon_0 \chi^{(2)} \int_V d^3\mathbf{r} \left(E_p^+(\mathbf{r}, t) E_s^-(\mathbf{r}, t) E_i^-(\mathbf{r}, t) + \text{c.c.} \right). \quad (3.10)$$

Now we constrain the fields to be collinear plane waves propagating in the z direction. The pump field is assumed strong and so on quantisation its positive frequency component is simply replaced by a classical field

$$E_p^+(z, t) \rightarrow A_p \int d\omega_p \alpha(\omega_p) e^{i(k_p(\omega_p)z - \omega_p t)}, \quad (3.11)$$

where A_p is the pump amplitude assumed real at the crystal face $z = 0$ and $\alpha(\omega_p)$ is the normalised *pump spectral amplitude*. A similar replacement applies for the negative frequency component. As in Section 2.2.1, quantising the signal and idler fields means associating the positive and negative frequency components with annihilation and creation operators respectively [211]:

$$\begin{aligned} E_j^+(z, t) &\rightarrow iA_j \int d\omega_j \hat{a}_j(\omega_j) e^{i(k_j(\omega_j)z - \omega_j t)}, \\ E_j^-(z, t) &\rightarrow -iA_j \int d\omega_j \hat{a}_j^\dagger(\omega_j) e^{-i(k_j(\omega_j)z - \omega_j t)}, \end{aligned} \quad (3.12)$$

where $j = s, i$. We have defined constants $A_j = \left(\hbar \omega_j / 2 \varepsilon_0 n(\omega_j^0) V_Q \right)^{1/2}$ where $n(\omega_j^0)$ is the refractive index at the central frequency ω_j^0 and V_Q is the quantisation volume. These describe the strength of the electric field associated with a single signal or idler photon. Substituting these quantised fields into the interaction Hamiltonian gives

$$\begin{aligned} \hat{H}_I(t) &= \varepsilon_0 \chi^{(2)} A_p A_s A_i \int_0^L dz \int d\omega_p \int d\omega_s \int d\omega_i \alpha(\omega_p) \hat{a}_s^\dagger(\omega_s) \hat{a}_i^\dagger(\omega_i) \\ &\times \exp[i(k_p(\omega_p) - k_s(\omega_s) - k_i(\omega_i))z] \times \exp[-i(\omega_p - \omega_s - \omega_i)t] + \text{h.c.} \end{aligned} \quad (3.13)$$

L is the length of the nonlinear medium. To investigate evolution of some state between times t_1 and t_2 under this Hamiltonian, we define the unitary propagator

$$\hat{U}(t_1, t_2) = \hat{T} \exp \left(\frac{1}{i\hbar} \int_{t_1}^{t_2} dt' \hat{H}_I(t') \right). \quad (3.14)$$

$\hat{T}$ is the time-ordering operator that ensures operators on the right happen before those on the left. The pump amplitude A_p mediates interaction inside the medium, ensuring it vanishes for times outside the temporal extent of the pump wavepacket. The limits in the integral over time can therefore be extended to infinity. Its evaluation converts the second exponential in $\hat{H}_I(t)$ to the delta function $2\pi\delta(\omega_p - \omega_s - \omega_i)$, enforcing energy conservation. Performing the integral over z yields $L \times \phi(\omega_s, \omega_i)$ where the *phase-matching function*

$$\phi(\omega_s, \omega_i) = \exp \left(\frac{i\Delta k L}{2} \right) \text{sinc} \left(\frac{\Delta k L}{2} \right) \quad (3.15)$$

where $\Delta k = k_p(\omega_s + \omega_i) - k_s(\omega_s) - k_i(\omega_i)$ is the wavevector mismatch and describes the dependence on the material's dispersion and birefringence [81]. We define the *joint spectral amplitude* (JSA) as $f(\omega_s, \omega_i) = \alpha(\omega_s + \omega_i) \times \phi(\omega_s, \omega_i)$ and the integrated interaction Hamiltonian becomes

$$\frac{1}{i\hbar} \int_{t_1}^{t_2} dt' \hat{H}_I(t') = \mathcal{A} \int d\omega_s \int d\omega_i f(\omega_s, \omega_i) \hat{a}_s^\dagger(\omega_s) \hat{a}_i^\dagger(\omega_i) + \text{h.c.} \quad (3.16)$$

The parameter $\mathcal{A} = 2\pi\varepsilon_0\chi^{(2)}A_pA_sA_iL/i\hbar$ describes the interaction strength. If the pump is sufficiently weak then $\mathcal{A}$ is small and the propagator can be expanded in a Taylor series to first order. Acting this on the vacuum gives the state [209, 212]

$$|\psi(\omega_s, \omega_i)\rangle \approx |0\rangle + \mathcal{A} \int d\omega_s \int d\omega_i f(\omega_s, \omega_i) \hat{a}_s^\dagger(\omega_s) \hat{a}_i^\dagger(\omega_i) |0\rangle. \quad (3.17)$$

Thus the action of the SPDC Hamiltonian on vacuum is to generate a pair of photons in signal and idler modes with some small probability $|\mathcal{A}|^2$. It is worth noting here that the coherence between the vacuum and photon pair components led to the use of SPDC sources in early experiments testing indistinguishability in quantum interference [213–215], and its impact will be of importance in Chapter 4 when we use a pair of SPDC sources to generate four photons.

3.2.4 Engineering a factorable source

If the vacuum component of the state in equation 3.17 is heralded out then SPDC can be used as a source of single photons [216]. Any correlations present in $f(\omega_s, \omega_i)$ mean the generated pair of photons are in a spectrally entangled state [81]. Heralding on the signal photon therefore projects the idler into a mixed state and spectral filtering is required to force emission into a well-defined spectral mode. The price to pay is loss: production rates decrease and the probability that a herald click is accompanied by the arrival of its partner decreases [217, 218].

The joint spectrum is $f(\omega_s, \omega_i) = \alpha(\omega_s + \omega_i) \times \phi(\omega_s, \omega_i)$. For a Gaussian pump spectrum centred at ω_p^0 and with width σ_p , and using a uniform nonlinear medium of length L , this yields

$$f(\omega_s, \omega_i) = (\pi\sigma_p^2)^{-1/4} \exp\left(-\frac{(\omega_s + \omega_i - \omega_p^0)^2}{2\sigma_p^2}\right) \times \exp\left(\frac{i\Delta k L}{2}\right) \text{sinc}\left(\frac{\Delta k L}{2}\right). \quad (3.18)$$

Correlations between ω_s and ω_i can arise from terms in the complex phase and from the product of the real parts of each function. The task is to tune parameters in this expression to remove any such terms and ensure factorability of the joint spectrum so $f(\omega_s, \omega_i) = f_s(\omega_s)f_i(\omega_i)$. This confines emission to uncorrelated broadband modes $\hat{A}_{s(i)}^\dagger(\omega_{s(i)}) = \int d\omega_{s(i)} f_{s(i)}(\omega_{s(i)}) \hat{a}_{s(i)}^\dagger(\omega_{s(i)})$ and circumvents the need for filtering [219]. The approach for factorable sources used in this thesis involves controlling the group velocities of the fields inside the nonlinear medium.

For a scalar field j , the wavenumber can be expanded to first order around the central phase-matched frequency ω_j^0 as:

$$k_j(\omega) = k_j(\omega_j^0) + v_j^{-1} \Omega_j. \quad (3.19)$$

The wavenumber $k_j(\omega_j^0) = n_j(\omega_j^0)\omega_j^0/c$ describes the effect of the refractive index $n_j(\omega_j^0)$ on the phase velocity. The group velocity of field j is $v_j = d\omega/dk_j(\omega)|_{\omega=\omega_j^0}$ and $\Omega_j = \omega_j - \omega_j^0$. The mismatch is then given by

$$\Delta k = \Delta k_0 + (v_p^{-1} - v_i^{-1}) \Omega_i + (v_p^{-1} - v_s^{-1}) \Omega_s, \quad (3.20)$$

where the centre of the phase-matching function is set by the material's dispersion through $\Delta k_0 = k_p(\omega_s^0 + \omega_i^0) - k_s(\omega_s^0) - k_i(\omega_i^0) = 0$, and v_p, v_s, v_i are the group velocities of the pump, signal and idler respectively. The first-order terms in the expansion determine the orientation of the phase-matching function. In (Ω_s, Ω_i) space it is aligned along an axis defined by θ_{PMF} (see Figure 3.3) that depends on the group velocities as [220]

$$\tan \theta_{PMF} = -\frac{v_p^{-1} - v_s^{-1}}{v_p^{-1} - v_i^{-1}}. \quad (3.21)$$

The pump function $\alpha(\omega_s + \omega_i)$ is always anti-diagonal in (Ω_s, Ω_i) . It is then a case of choosing a pump bandwidth, the bandwidth of the phase-matching function, and the orientation θ_{PMF} so that the product of the two functions is factorable (as shown in the example in Figure 3.3).

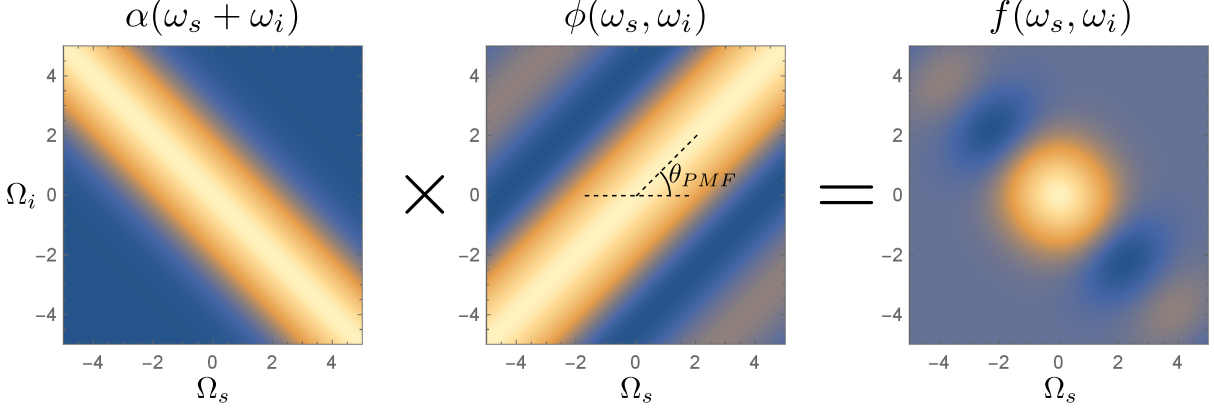


Figure 3.3: Example of engineering the joint spectrum $f(\omega_s, \omega_i)$ by controlling the pump function $\alpha(\omega_s + \omega_i)$ and phase-matching $\phi(\omega_s, \omega_i)$. We assume a Gaussian pump and the orientation of the phase-matching function θ_{PMF} is determined by the group velocities of the three fields (equation 3.21). Here the functions' widths are comparable so their product gives an approximately factorable joint with sinc lobes that can be removed using broad filters. Using narrower pump or phase-matching functions, or adjusting θ_{PMF} can correlate the generated fields.

In what follows we assume that the joint spectral amplitude has no phase correlations between the signal and idler modes. For example using a pump with a varying spectral phase would correlate the down-conversion modes and so degrade their factorability. We assume the pump employed is transform-limited and so has a flat phase. The phase-matching function in equation 3.18 gives a dependence on $\exp(i\Delta k L/2)$. In this first order expansion we ignore the effects of group velocity dispersion (GVD) arising from second derivatives of the wavenumber. GVD means that different frequencies travel at different group velocities: the result is that phases quadratic in frequency broaden the wavepackets so they are no longer transform-limited. This effect is small for wavepackets with a narrow spectrum, as is the case for both sources presented here [209, 221].

3.2.5 Factorable SPDC in bulk crystals

Orthogonal polarisations in birefringent crystals experience different refractive indices and, together with dispersion, this opens up the possibility of tuning the group velocities of fields in the crystal by adjusting their propagation directions. By equations 3.20 and 3.21 this therefore allows manipulation of the phase-matching function: *birefringent phase-matching*.

If $\phi(\omega_s, \omega_i)$ can be engineered to be a function of only one frequency then, given a pump with a Gaussian spectrum that is much broader than the bandwidth of the phase-matching function, it will dominate the joint spectrum and yield photons in factorable wavepackets [35]. This can be achieved by using a long crystal that has a correspondingly narrow phase-matching bandwidth, and then orientating its optic axis so that the group velocities of the pump and idler are equal: $\theta_{PMF} = \pi/2$ so $\phi(\omega_s, \omega_i)$ is vertical. This technique was demonstrated by the group in Oxford in 2007: Peter Mosley built a factorable type-II SPDC source using potassium dihydrogen phosphate (KDP) crystals [212, 222]. These sources are used throughout this thesis

3. Three-photon interference

so we will review why the emission is factorable and then describe the experimental setup. Further details can be found in Mosley's thesis [209].

The phase-matched frequencies are set by the first term in the wavevector mismatch (Δk_0 in equation 3.20). For this type-II process the pump and signal fields are e-polarised and the idler is o-polarised. KDP is a negative uniaxial crystal and, by changing the orientation of its optic axis θ_{OA} , the refractive index of e-polarised fields can be changed. Δk_0 vanishes for collinear degenerate down-conversion with the pump at 415nm, the signal and idler both at 830nm, and $\theta_{OA} = 67.8^\circ$ [209]. When oriented in this way, KDP's dispersion also means that the group velocities of an ultrafast e-polarised 415nm pump v_p^e and the o-polarised 830nm idler v_i^o are the same. Hence the phase-matching function is oriented vertically and only depends on Ω_s , so a factorable joint spectrum is possible. In order to appreciate the single mode character of emission we consider what happens to the fields inside the KDP crystal in Figure 3.4.

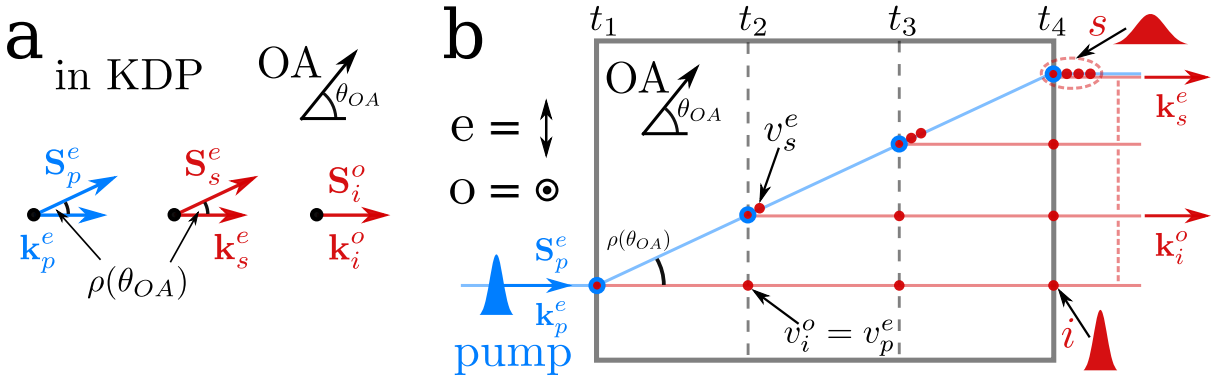


Figure 3.4: **a** The birefringence of a KDP crystal means that if its optic axis (OA) is oriented at θ_{OA} , then the Poynting vector $\mathbf{S}_{p(s)}^e$ for e-polarised light is aligned with the walk-off angle $\rho(\theta_{OA}) = \tan^{-1}[(n_o/n_e)^2 \tan \theta_{OA}] + \theta_{OA}$ instead of parallel to its wavevector $\mathbf{k}_{p(s)}^e$. The Poynting vector for o-polarised light $\mathbf{S}_i^o$ remains parallel to the wavevector $\mathbf{k}_i^o$. **b** The blue line indicates the path taken by a blue pump pulse at 415nm inside the crystal. As it propagates, possible down-conversion events at times t_i (indicated by blue circles) can generate daughter photons (red circles). The o-polarised idler photons i have the same group velocity as the pump ($v_i^o = v_p^e$) but travel in the direction of the pump's wavevector, as shown by the horizontal red lines. The idler's temporal mode is therefore constrained to be the same as that of the pump: a pure, transform-limited ultrafast wavepacket. Its transverse spatial mode out of the crystal is given by the sum of amplitudes indicated by the red vertical dashed line. Meanwhile the e-polarised signal photons s travel in the same direction as the pump but with a greater group velocity v_s^e . When the pump pulse leaves the crystal, the temporal mode of the signal photon is found by summing the amplitudes indicated by the dashed ellipse, resulting in a pure temporally broader wavepacket. The joint temporal amplitude is factorable because the emission time of the idler is determined by the pump pulse timing, and is uncorrelated with that of the signal due to temporal walk-off. Taking the Fourier transform yields a factorable joint spectral amplitude. The pump spot size is larger than the walk off in the crystal, so the generated fields have similar transverse spatial modes that can be well-matched to fibre. Adapted from [209].

The KDP experimental setup is shown in Figure 3.5. A mode-locked Ti:Sapph laser with a repetition rate of 80MHz emits 100fs pulses centred at 830nm that are focussed into a barium borate (BBO) crystal. The BBO is angle-tuned for phase-matching of type-I second-harmonic generation at 415nm. SHG requires two pump photons and so its efficiency is quadratic in pump intensity. This can be increased by using short pulse durations and tight focussing into

the crystal. The latter results in a range of angles for wavevectors incident at the crystal, and subsequently the phase-matching condition leads to spatial chirp across the wavefront of the second harmonic field.

These 415nm pulses are used to pump a KDP crystal that is angle-tuned to phase-match collinear type-II degenerate parametric down-conversion at 830nm. The o-polarised idler occupies a temporally short mode with a bandwidth of $\sim 12\text{nm}$, defined by the pump pulse's wavepacket. The e-polarised signal occupies a longer duration wavepacket with a narrower bandwidth of $\sim 3\text{nm}$ determined by the crystal length. Weak focussing into the KDP crystal eliminates the spatial chirp across the SHG wavefront and, as described in Figure 3.4, the signal and idler modes are spectrally uncorrelated, collinear and orthogonally polarised. These are spatially separated using a polarising beam splitter (PBS), and then coupled into single-mode fibres. The SPDC process thus results in probabilistic generation of pairs of signal and idler photons in spectrally uncorrelated pure spatio-temporal modes (see Figure 3.6).

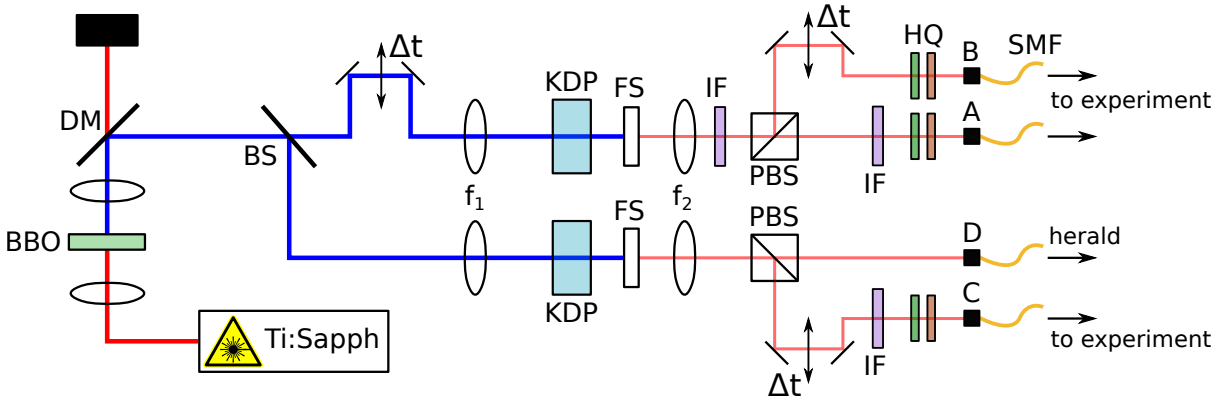


Figure 3.5: A Ti:Sapph laser (Spectra-Physics Mai Tai HP) running at a repetition rate of 80MHz generates 100fs pulses centred at 830nm, with an average power of 2.7W. These are upconverted by type-I SHG in a $700\mu\text{m}$ β -barium borate (BBO) crystal to give pulses centred at 415nm with average power of up to 650mW. The SHG is separated from the fundamental using dichroic mirrors (DM) and split on a balanced plate beam splitter (BS). It is then focussed using lenses with focal length $f_1 = 250\text{mm}$ into a pair of 8mm-long AR-coated potassium dihydrogen phosphate (KDP) crystals oriented to phase-match degenerate collinear type-II parametric down-conversion. The blue pump is removed using a stack of filters (FS) and the emission is collimated using lenses with focal length $f_2 = 150\text{mm}$. The orthogonally polarised signal and idler photons are spatially separated on polarising beam splitters (PBS) before coupling into single-mode fibres (SMF). A and D are broadband idler photons, and B and C are narrowband signal photons. Interference filters (IF) with a bandwidth $\Delta\lambda = 3\text{nm}$ ensure identical spectra for the three photons used in experiments, and D is left unfiltered to increase heralding efficiency. Motorised delay stages can apply temporal delays Δt and half- and quarter-wave plates (respectively H and Q) enable preparation of the polarisation state. Adapted from [212].

Two of these sources were built by Peter Mosley and he measured heralded HOM dips without additional filtering: the visibility for the narrowband signal photons was 95% and for the broadband idlers was 90%. In both cases the dips were consistent with transform-limited wavepackets, as expected if GVD effects are small [209]. More recent HOM dips taken after a realignment of the source are presented in Section 3.2.6.

This pair of sources can be used to deliver three indistinguishable photons: one is used in a heralded configuration, where an idler click heralds a narrowband signal photon, whilst the

3. Three-photon interference

other source has its idler mode filtered to match the signal (see Figure 3.6). The filtering does not impose any varying spectral phase and so the idler wavepacket remains transform-limited. This is also verified by the high visibility HOM interference presented later in Figure 3.9a. Post-selection on a herald click and three clicks after an interferometer enables elimination of instances when more than two photons were produced solely by the unheralded source.

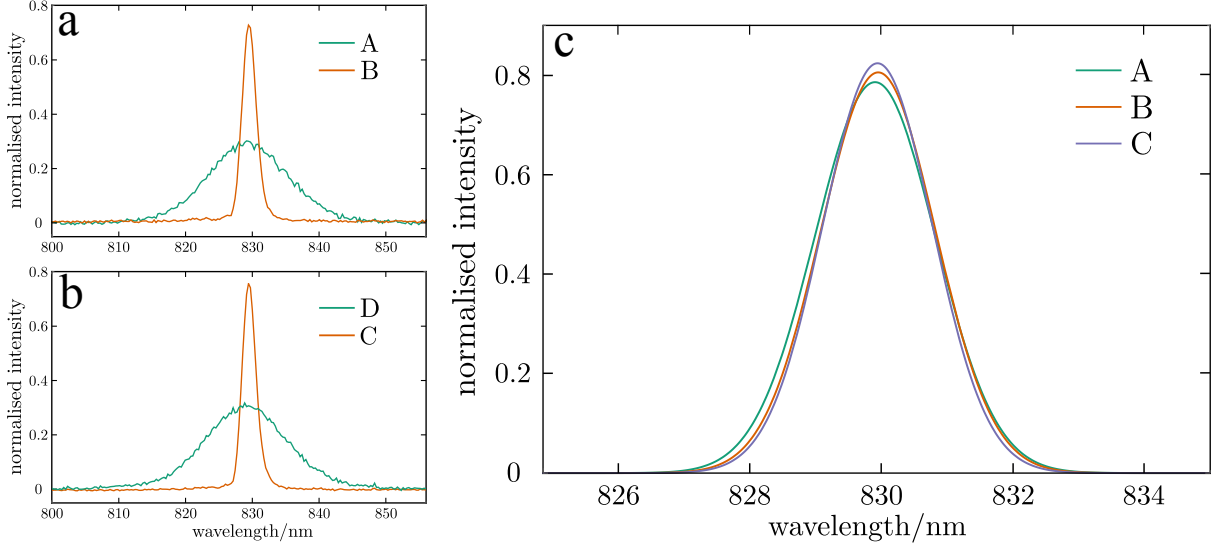


Figure 3.6: Marginal spectra of the KDP down-conversion. **a,b** The narrowband signal and broadband idler photons, with bandwidths of around 2.5nm and 12.5nm respectively. **c** With the filtering shown in Figure 3.5 it is possible to obtain three spectrally indistinguishable photons from arms A, B and C with bandwidths of ~ 2.2 nm. D is left unfiltered and used as a herald. HOM dip measurements presented in Figure 3.9 show temporal widths consistent with transform-limited wavepackets described by these spectra.

3.2.6 SPDC source characterisation

When an SPDC source has a factorable spectrum, the action of the propagator of equation 3.14 on vacuum is to produce a two-mode squeezed vacuum (TMSV) state given by [29, 223]

$$|\psi_{TMSV}\rangle = \sqrt{1 - \lambda^2} \sum_{n=0}^{\infty} \lambda^n |n_s, n_i\rangle. \quad (3.22)$$

The squeezing parameter $0 \leq \lambda < 1$ can be associated with the parameter $\mathcal{A}$ of equation 3.16. λ is assumed real and depends on the material's $\chi^{(2)}$ nonlinearity, the interaction length, and its square is linear in pump power. In contrast to the state in equation 3.17 which is a first-order approximation, we have kept all terms in the interaction Hamiltonian and accounted for time-ordering [224]. These higher-order terms result in multiple pair generation. The state has been expressed in the occupation number representation and the signal and idler modes are spatially separated broadband wavepackets. The probability of producing n pairs is given by $P(n) = (1 - \lambda^2) \times \lambda^{2n}$, and so the mean number of pairs is $\langle \hat{n} \rangle = \lambda^2 / (1 - \lambda^2)$.

Correlation functions were introduced by Glauber to quantify the degree of coherence of states of the quantised electromagnetic field [211]. First-order coherence functions test field-field correlations and it is possible to show that Fock states and coherent states are both first-order

coherent [29]. Second-order coherence functions probe intensity-intensity correlations or, in quantised language, photon number correlations. For a field in a single mode, the second-order correlation function at time τ is [29]

$$g^{(2)}(\tau) = \frac{\langle \hat{a}^\dagger \hat{a}^\dagger \hat{a} \hat{a} \rangle}{\langle \hat{a}^\dagger \hat{a} \rangle^2} = \frac{\langle \hat{n}^2 \rangle - \langle \hat{n} \rangle^2}{\langle \hat{n} \rangle^2}, \quad (3.23)$$

where expectations are evaluated at time τ . A coherent state $|\alpha\rangle$ is second-order coherent so $g^{(2)}(\tau) = 1$, reflecting random photon arrival times for a Poissonian distribution. A single photon Fock state has $g^{(2)}(0) = 0$ indicating sub-Poissonian statistics and that, given a detection of one photon, it is impossible to detect another simultaneously. Thermal states have bunched statistics with $g^{(2)}(0) = 2$ meaning that, given a detection, one is more likely to simultaneously detect another photon than for a coherent state.

If the idler mode of the state in equation 3.22 is blocked then we gain no knowledge of the number statistics and the signal is described by the reduced state

$$\rho_s = (1 - \lambda^2) \sum_{n=0}^{\infty} \lambda^{2n} |n_s\rangle \langle n_s|. \quad (3.24)$$

The number correlations result in mixture on tracing out the idler and so ρ_s is a thermal state with $g^{(2)}(0) = 2$. If an SPDC source is not factorable then a single-valued decomposition of its joint spectrum shows that the total state produced is instead a tensor product of different TMSV states [200, 225, 226]. The number statistics are then a convolution of those for separate thermal states, and $g^{(2)}(0) \approx 1 + \frac{1}{K}$ where K is the mode number. If the statistics are those of a single thermal state then $K = 1$ [227].

To measure $g^{(2)}(0)$ for the KDP sources, we block the idler arm and the signal is sent to a balanced fibre beam splitter. Singles and coincidences are recorded at the outputs using silicon avalanche photodiodes (APDs, PerkinElmer and Excelitas) connected to a signal correlator (Swabian TimeTagger 20) (see Figure 3.7). The unheralded second-order correlation is then

$$g^{(2)}(0) = \frac{N_{\text{trials}} \times C_{1,2}}{C_1 \times C_2}. \quad (3.25)$$

N_{trials} is the product of the repetition rate of the laser and the total integration time. C_1, C_2 are the number of singles recorded at the outputs of the beam splitter, and $C_{1,2}$ is the number of output coincidences recorded within a 4ns coincidence window (it must be less than the 12.5ns between laser pulses and allows for imperfect timing response of APDs). This was measured for a range of different 415nm pump powers, from 50mW to 320mW, and averaged 2.04 ± 0.07 , confirming $K \approx 1$ for this factorable source. It is worth noting that this measurement is not particularly resistant to noise in the system, so HOM measurements shown later are a better indicator of state purity [37].

We also take heralded correlation measurements where a detection on the signal arm projects out the vacuum component, leaving the idler mode in

$$\rho_s^{(h)} = \frac{1 - \lambda^2}{\lambda^2} \sum_{n=1}^{\infty} \lambda^{2n} |n_s\rangle \langle n_s|. \quad (3.26)$$

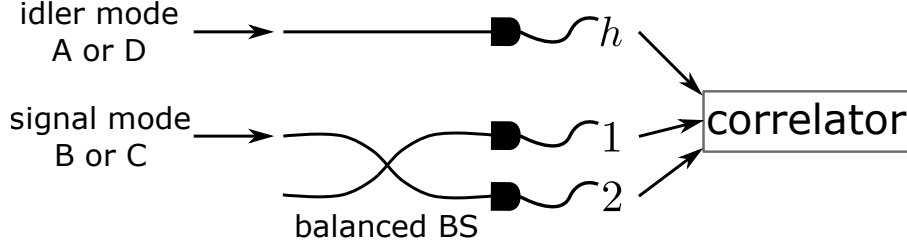


Figure 3.7: Setup for measuring second-order correlation functions using spatially multiplexed bucket detectors (here silicon APDs). The unheralded $g^{(2)}(0)$ is measured by blocking the idler mode (or just ignoring its channel at the correlator) and recording singles and coincidences at the outputs of a balanced beam splitter. The heralded correlation function is measured by conditioning on the idler mode. See main text for details.

The probability of delivering n photons given a herald click is then ideally $P(n|h) = \frac{1-\lambda^2}{\lambda^2} \times \lambda^{2n}$, giving a mean heralded photon number $\langle \hat{n}^{(h)} \rangle = 1/(1 - \lambda^2)$. The heralded correlation measurement is conditioned on a herald click that here acts as the trial counter, as opposed to the laser repetition rate previously, so

$$g_h^{(2)}(0) = \frac{C_h \times C_{h,1,2}}{C_{h,1} \times C_{h,2}}, \quad (3.27)$$

where C_h are counts on the herald arm, and $C_{h,1/2}$ are coincidences between the herald and a beam splitter output. For an ideal single photon source this should be zero, indicating perfectly anti-bunched light. The presence of threefold coincidences $C_{h,1,2}$ in the measurement introduces a sensitivity to multiphoton emissions [223]. We can use this to determine the squeezing parameter λ using [221, 228]

$$\lambda^2 = g_h^{(2)}(0) \times \frac{\eta_h}{2(1 - (1 - \eta_h)^2)}. \quad (3.28)$$

The heralding efficiency $\eta_h = (C_{h,1} + C_{h,2})/(C_1 + C_2)$ captures the fraction of SPDC pairs that are detected and indicates losses in the setup [37, 229]. These KDP sources have raw heralding efficiencies between 25–30%, independent of which mode is the herald and increasing slightly when filters are inserted. Using the heralded setup in Figure 3.7, we recorded singles and heralded coincidences to obtain the squeezing strength λ^2 as a function of pump power (see Figure 3.8).

As expected for SPDC, the squeezing strength λ^2 and generation rates are linear in pump power. At lower pump powers, $g_h^{(2)}(0) \approx 0$ and indicates an approximate single photon source. However for a maximum squeezing parameter of $\lambda \approx 0.16$, this increases to $g_h^{(2)}(0) \approx 0.1$ and the mean photon number $\langle \hat{n} \rangle \approx 1.03$. These multiphoton emissions at higher pump powers can contaminate interference statistics because the scattering of multiple photons in the same mode is different from that for single photons.

The spectra of the three photons used in experiments are matched using 3nm filters (Semrock LL01-830-12.5) inserted as shown in Figure 3.5. A spectrometer (Andor Shamrock 163 with Andor iDus CCD for single-photon sensitivity) was used to monitor their spectra and, by angle-tuning the filters, spectral indistinguishability was achieved with fitted spectral overlaps of over 99% (see Figure 3.6c). As a final verification of their indistinguishability, HOM dips were

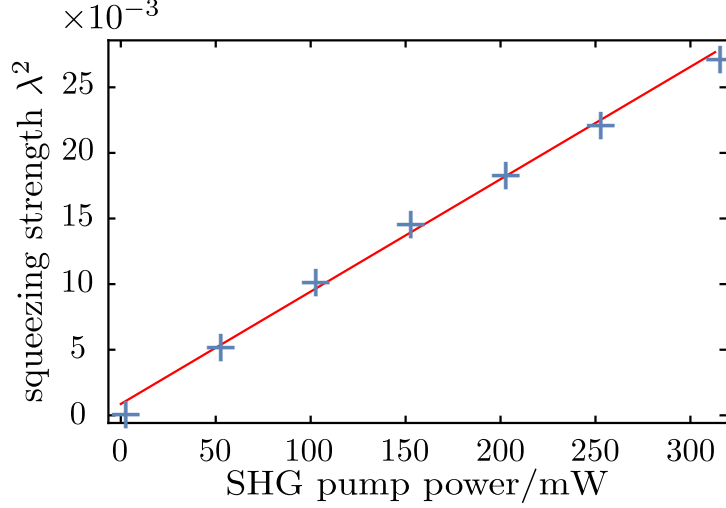


Figure 3.8: Heralded $g_h^{(2)}(0)$ measurements using the setup in Figure 3.7 were taken at different SHG pump powers and, using equation 3.28, the squeezing strength λ^2 can be calculated. As expected for weak driving, it is linear in SHG pump power: the number of blue photons available to down-convert increases linearly with pump power.

taken between pairs of photons from the same and independent sources. Photons were injected into the ports of a broadband polarisation-independent balanced fibre beam splitter. Sets of waveplates on each mode (in the order quarter-, half-, quarter-wave) were used to correct for random polarisation rotations in the SMFs and so ensure indistinguishable polarisations at the coupling region (see Appendix A). Motorised translation stages (see Figure 3.5) were used to tune the temporal distinguishability and obtain the dips shown in Figure 3.9.

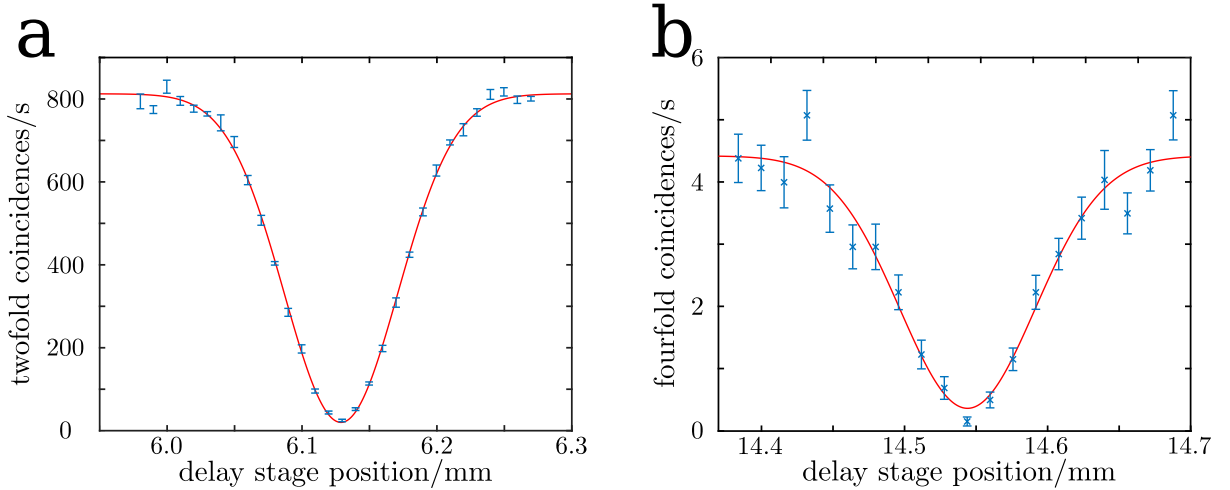


Figure 3.9: HOM dips to verify the generation of indistinguishable photons. **a** Photons from the same KDP crystal are filtered for spectral indistinguishability and yield a HOM dip with fitted visibility of $97.5 \pm 1.5\%$. **b** Two narrowband photons from two independent sources, when heralded, give a dip with visibility of $92 \pm 7\%$. In both cases, simulations verify that higher order emissions contaminate the suppression of coincidences. Error bars are from repeated sweeps.

The full width at half maximum (FWHM) of a HOM dip for photons in Gaussian wavepackets with coherence lengths (FWHM) l_1 and l_2 is

$$l_{dip} = \sqrt{l_1^2 + l_2^2}. \quad (3.29)$$

Assuming identical wavepackets, the dip width is simply $l_{\text{dip}} = \sqrt{2}l_1$. Given a transform-limited wavepacket with central wavelength λ_0 and bandwidth $\Delta\lambda$, its coherence length is

$$l_1 = \frac{2 \ln 2}{\pi} \times \frac{\lambda_0^2}{\Delta\lambda}. \quad (3.30)$$

The spectrum for the KDP sources, when filtered to have the same bandwidth (Figure 3.6), is centred at $\lambda_0 = 830\text{nm}$ and $\Delta\lambda = 2.2\text{nm}$. Therefore the transform-limited coherence length is expected to be $l_1 = 140\mu\text{m}$. From the fitted widths of the HOM dips in Figure 3.9a and b, we infer that the coherence lengths of the photons are respectively $140\mu\text{m}$ and $155\mu\text{m}$. Hence wavepackets produced from the same filtered source are essentially perfectly transform-limited, but there is some slight mismatch between those from separate sources. However the overlap of wavepackets with these slightly different widths at zero relative delay is effectively unity, so there is very little temporal distinguishability.

Despite high spectral and temporal indistinguishability, the dips do not have perfect visibility. The most likely causes are slight mismatch of the polarisation states at the coupler and higher-order terms in the TMSV state of equation 3.22. In conjunction with bucket detectors, the effect of the latter is to mix in the scattering statistics of emissions of more than one pair of photons with the desired HOM signal (see Appendix B.2). This leads to deterioration of the suppression of recorded coincidences at zero time delay. An extensive simulation code was written in *Mathematica* that models the effect of these imperfections on scattering experiments (see Appendix B). The visibilities from fits to the dips in Figure 3.9 are consistent with simulations using independent measurements of λ and system losses. We can therefore confirm that we have a source of three highly indistinguishable photons in separable pure transform-limited wavepackets, and that we can model subsequent experiments with appropriate simulations.

3.2.7 Factorable SFWM in a silica chip

The majority of the data presented in this thesis was obtained using the KDP SPDC sources, so we have reviewed their operation and characterisation in some detail. Another source was used for initial experiments probing three-photon interference and is based on spontaneous four-wave mixing (SFWM) in a silica-on-silicon chip. This source was developed by another student at Oxford, Justin Spring, in collaboration with the University of Southampton [167, 230]. Full details can be found in his thesis [221] but we will briefly review its operation and experimental setup.

Silica's $\chi^{(2)}$ is zero because it is centrosymmetric so parametric down-conversion is not possible. Instead we rely on its third-order nonlinearity $\chi^{(3)}$ and consider the process of four-wave mixing (FWM) for photon generation. The interaction Hamiltonian can be found in a similar way to that for SPDC in Section 3.2.3. Assuming a single pump p , one can expand out the fields interacting in a medium and retain the FWM term:

$$H'_I(t) = \frac{3}{4}\varepsilon_0\chi^{(3)} \int_V d^3\mathbf{r} \left((E_p^+(\mathbf{r}, t))^2 E_s^-(\mathbf{r}, t) E_i^-(\mathbf{r}, t) + \text{c.c.} \right). \quad (3.31)$$

Taking the pump to be a strong classical field, we can quantise the signal and idler fields then

follow the same steps as in equations 3.11-3.16 to obtain an approximation to the resulting SFWM quantum state:

$$|\psi'(\omega_s, \omega_i)\rangle \approx |0\rangle + \mathcal{A}' \int d\omega_s \int d\omega_i f(\omega_s, \omega_i) \hat{a}_s^\dagger(\omega_s) \hat{a}_i^\dagger(\omega_i) |0\rangle. \quad (3.32)$$

The constant $\mathcal{A}' = 3\pi\epsilon_0\chi^{(3)}A_p^2A_sA_iL/4i\hbar$ describes the interaction strength, where A_p is the pump amplitude, $A_{s(i)}$ is the strength of the signal (idler) electric field and L is the length of the medium. Energy conservation now requires that $2\omega_p = \omega_s + \omega_i$, and the pump and phase-matching functions are defined in the same way as for SPDC. The wavevector mismatch is now $\Delta k' = 2k_p(2\omega_p) - k_s(\omega_s) - k_i(\omega_i) = 0$. The output state has the same form as for SPDC earlier so we can think about group velocity matching as in Section 3.2.4.

The group velocity dispersion in silica waveguides is harder to engineer than in bulk crystals so the approach used for KDP, where a broad pump and vertical phase-matching function (from matching group velocities of pump and idler) ensured factorability, is not appropriate. Instead the orientation of the phase-matching function θ_{PMF} is chosen to be close to orthogonal to the pump function. If the bandwidths of the functions are also made equal, either by filtering the pump or choosing L , then the joint spectrum can be close to factorable, similar to the example shown in Figure 3.3. Symmetric group velocity matching where $v_s < v_p < v_i$ means that, as can be shown using equation 3.21, θ_{PMF} can be made close to 45° .

This matching requires the flexibility provided by birefringence. During fabrication, a stress between the silica and silicon layers of the chip develops and gives a uniform birefringence aligned with its axes. Its value is determined by suitable choice of dopant in the glass. UV writing is then used to cause a permanent refractive index change in the doped silica layer, thus defining waveguides that are single-mode at the desired wavelengths. The phase-matched process here converts two pump photons at 740nm to a pair of signal and idler photons at 680nm and 817nm respectively. The pump photons are polarised along the high refractive index axis, and the daughter photons are polarised along the lower index axis. An array of these waveguides was written into a single chip and three were simultaneously pumped by the same Ti:Sapph, probabilistically generating three signal-idler pairs. The signals are used to herald the arrival of three idler photons that are then used for interference experiments (see Figure 3.10).

A full characterisation of this source has been performed by Spring using the techniques presented in the previous section [167, 221]. The central wavelengths of the signal and idler are respectively 680nm and 817nm, and both have bandwidths of around 5nm. The heralded second-order correlation $g_h^{(2)}(0) = 0.08 \pm 0.01$ at the pump power used for generating three photons (130mW per waveguide) and the squeezing parameter $\lambda = 0.16$. Heralded HOM visibilities between pairs of the three waveguides used were around 90% and we estimate the spectral purity $\mathcal{P} \approx 0.95$ and residual distinguishability $r_{ij} \approx 0.95$. As well as the desired SFWM process, this source suffers from background fluorescence whose spectrum lies within the range of signal and idler emissions. It can be measured by rotating the pump's polarisation to lie along the chip's fast axis, thus turning off phase-matching of SFWM, and then monitoring the output emission on a spectrometer. The noise is thought to derive from excitation of defects in the silica induced during the UV writing process [167]. Such fluorescence has a lifetime much

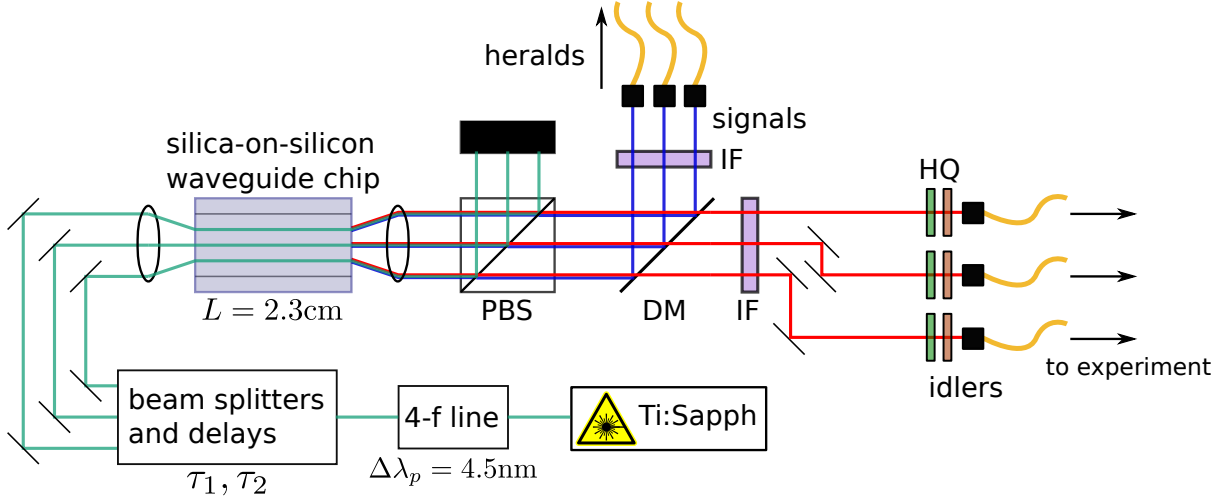


Figure 3.10: A Ti:Sapph laser (Spectra-Physics Mai Tai HP) running at a repetition rate of 80MHz generates 100fs pulses centred at 740nm. A 4-f line is used to set the pump bandwidth $\Delta\lambda_p$. A sequence of bulk beam splitters splits the pump in three, with two paths passing along motorised delay stages τ_1, τ_2 . The three beams are focussed into the silica chip using a single lens and three waveguides probabilistically generate three pairs of daughter photons. After collimation, the pump is removed using a polarising beam splitter (PBS) and then a dichroic mirror (DM) separates the signal photons at 680nm from the idler photons at 817nm. After broad filtering (IF), the signals are coupled into single-mode fibres (SMF) and detected as heralds using APDs. The idlers' polarisations are prepared using sequences of half- and quarter-wave plates before coupling into SMFs, ready to be used in experiments. Adapted from [167].

longer than the pump pulse length so much of it can be removed by setting a narrow coincidence window in correlation measurements. However the overall effect is to contaminate statistics with those involving distinguishable photons and subsequent simulations included the probabilities of generating such photons in the signal and idler arms (see Appendix B.5).

3.3 Three-mode interferometer

3.3.1 Balanced tritter

In order to test the partial exchange symmetry of three particles, we use an interferometer with three ports. Such devices have already been investigated when generalising HOM-like suppressions for three particles [102]. The interferometer that has equally strong coupling between the three ports is the balanced 3×3 Fourier multiport called a *tritter* [39, 45, 231]. Early investigations considered its application to generating high-dimensional entanglement [232, 233] and it has also found use in classical and quantum interferometry [108, 116, 166, 167, 234–237]. It is described by the unitary matrix

$$U_{trit}^{(1)} = \frac{1}{\sqrt{3}} \begin{pmatrix} 1 & 1 & 1 \\ 1 & \zeta & \zeta^* \\ 1 & \zeta^* & \zeta \end{pmatrix}, \quad \zeta = e^{i\frac{2\pi}{3}}, \quad (3.33)$$

where the probability that a single particle injected into any input port ends up in any output port is $1/3$. The matrix has been put in real-bordered form by applying phases to the input

and output modes that do not affect interference statistics for separable inputs. There is another canonical form of the tritter given by $U_{trit}^{(2)} = \left(U_{trit}^{(1)}\right)^*$ and the two are also related by a permutation of two input or output modes [232, 233, 235].

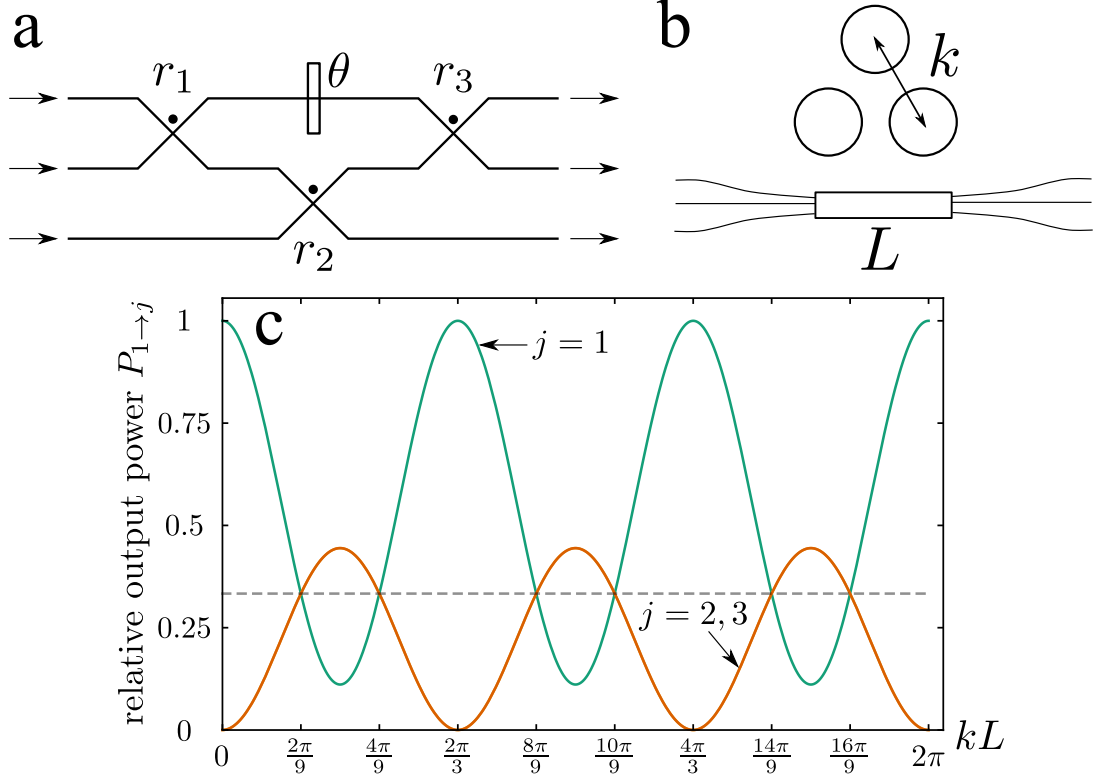


Figure 3.11: **a** Any 3×3 interferometer can be decomposed into beam splitters and a phase shifter as shown, up to input and output phases that do not affect interference of photons in separable states. For a balanced device, setting the reflectivities $r_1 = r_3 = 1/\sqrt{2}, r_2 = 1/\sqrt{3}$ and the phase $\theta = \pm \frac{\pi}{2}$ yields respectively $U_{trit}^{(1)}$ and $U_{trit}^{(2)}$. The dots indicate reflections that are accompanied by a π phase shift. **b** Three fibres with coupling constant k and interaction length L may be fused to produce a balanced tritter. **c** Power splitting across outputs j when light is injected into input 1 and varying kL . The grey dashed line indicates a balanced splitting ratio, and crossings achieve alternately $U_{trit}^{(2)}$ and $U_{trit}^{(1)}$.

Any linear interferometer can be decomposed into beam splitters and phase shifters [47], and this is shown for the tritter in Figure 3.11a. When built using free-space optics, this has the drawback of requiring stabilisation of the internal phase to maintain an equal splitting ratio [45]. Another approach is to fuse three single-mode fibres so that their evanescent fields can mix. The power splitting is determined by the coupling strength k and interaction length L [166] (see Figure 3.11b). Equal splitting is achieved when $kL = \frac{2\pi}{9}(3a - 2), \frac{2\pi}{9}(3a - 1)$, where $a \in \mathbb{Z}^+$, giving respectively $U_{trit}^{(2)}$ and $U_{trit}^{(1)}$ (see Figure 3.11c).

3.3.2 Characterisation

It is crucial to characterise a device's scattering matrix $M_{ij} = \alpha_{ij}e^{i\theta_{ij}}$ in order to predict interference statistics using equation 2.54. Two approaches are common: one using classical light and another using single photons [238, 239]. The first approach determines the matrix elements' magnitudes by injecting classical light into each input port in turn and then measuring

the distribution of power at the outputs. The ratio of these output powers gives the splitting probabilities, whose square roots give elements' magnitudes by $P_{i \rightarrow j} = |M_{ij}|^2 = \alpha_{ij}^2$. The phases of elements in the first row and first column θ_{i1}, θ_{1j} can be assumed zero by putting the matrix in real-bordered form. In order to determine the remaining phases, classical light is sent into a balanced beam splitter whose outputs are connected to the first and p th inputs of the interferometer. By adjusting the relative phase ϕ of the inputs, for example by a relative delay for a continuous-wave laser, the intensity at output q is maximised when $\theta_{pq} = 2\pi - \phi$. Repeating for all p and q permits measurement of all non-trivial phases of M_{ij} .

In the second approach, single photon transmissions are used to determine splitting probabilities in much the same way as for classical light, but using singles counts instead of power. The sensitivity of two-photon interference to interferometer phases enables determination of θ_{ij} . In particular, for two indistinguishable photons injected into inputs i, j , the HOM dip visibility at outputs k, l is given by [237, 239]

$$\begin{aligned} V_{ij}^{kl} &= \frac{P_{ij}^{kl}(\text{dist}) - P_{ij}^{kl}(\text{indist})}{P_{ij}^{kl}(\text{dist})} \\ &= -\frac{2\text{Re}[(M_{il}M_{jk}) \times (M_{ik}M_{jl})^*]}{|M_{ik}M_{jl}|^2 + |M_{il}M_{jk}|^2} \\ &= \frac{-2\alpha_{il}\alpha_{jk}\alpha_{ik}\alpha_{jl}\cos(\phi_{il} + \phi_{jk} - \phi_{ik} - \phi_{jl})}{(\alpha_{ik}\alpha_{jl})^2 + (\alpha_{il}\alpha_{jk})^2}. \end{aligned} \quad (3.34)$$

$P_{ij}^{kl}(\text{dist})(P_{ij}^{kl}(\text{indist}))$ are the measured output probabilities for temporally (in)distinguishable photons. Unlike the measurements using single photon transmissions, V_{ij}^{kl} is resistant to loss because it post-selects on events where both photons made it through the interferometer. Knowledge of HOM visibilities and element magnitudes from previous measurements thus permit calculation of the phases. There is a sign ambiguity in the cosine and so θ_{22} is usually taken to be positive, which then sets the signs of the remaining phases [239]. This means that it is not possible to discriminate between $U_{trit}^{(1)}$ and $U_{trit}^{(2)}$ using this characterisation technique, but we will see later that these canonical tritters yield different three-photon interference statistics.

We use a fused fibre tritter manufactured by Gooch & Housego. An excess loss of $\sim 0.6\text{dB}$ was measured using an attenuated pick-off from the Ti:Sapph, filtered to have the same spectrum as the down-conversion photons. We assume these losses occur at fibre connectors and that there is negligible loss in the coupler itself. The coupler is then described by a 3×3 unitary matrix that is parametrised by four real numbers (see Figure 3.11a). We used both classical and quantum techniques to characterise the tritter. In the first instance, the splitting ratios measured using the classical pick-off determine the matrix element magnitudes and, for a 3×3 unitary, these are also sufficient to fully characterise all the phases [240].³ For the second approach we record HOM dips using a single filtered SPDC source with polarisations compensated to be indistinguishable at the coupling region of the tritter. There are a total of nine dips that over-determine the four parameters of a 3×3 unitary. Their visibilities depend on matrix elements' magnitudes and phases (see equation 3.34), but are also sensitive to imperfect indistinguishability and the effect of higher-order emissions. In order to infer the tritter unitary, we perform a least-squares

³In Chapter 4 we shall see that this is not the case for a four-mode coupler.

minimisation of the difference between the measured visibilities and those obtained from a simulation that includes these imperfections, and that has the unitary's defining numbers as free parameters. The reconstructed tritter matrix was found to be the same independent of which technique was used and is given by

$$U_{trit}^{exp} = \frac{1}{\sqrt{3}} \begin{pmatrix} 1.05 & 1.02 & 0.92 \\ 1.01 & 0.94e^{i \times 0.684\pi} & 1.05e^{-i \times 0.688\pi} \\ 0.94 & 1.04e^{-i \times 0.697\pi} & 1.03e^{i \times 0.628\pi} \end{pmatrix}. \quad (3.35)$$

For an ideal tritter, the two-photon coincidence probabilities are the same for all pairs of input and output ports:

$$P_{11} = \frac{1}{9} (2 - r_{ij}^2). \quad (3.36)$$

For indistinguishable states the HOM dip visibilities are therefore $V_{ij}^{kl} = 0.5$, independent of input and output ports. For our device we measure visibilities of between 36% and 58% showing that, despite the moduli looking reasonably uniform, splitting imbalances have a large effect on two-photon interference visibility.

The excess losses, splitting probabilities, and dip visibilities were the same when using the orthogonal polarisation, verifying polarisation independence. This is critical for testing partial exchange symmetry because the internal degrees of freedom should not be mixed. This is straightforward to accomplish for temporal modes, but birefringence of components can affect the coupling of polarisation states.

3.3.3 Interference statistics for three photons

If three photons prepared in states $|a\rangle, |b\rangle, |c\rangle$ are injected into inputs 1, 2 and 3 respectively of a balanced tritter $U_{trit}^{(1)}$, then the expression for multiphoton probabilities in equation 2.54 gives the probability of detecting a threefold coincidence $\vec{s} = (1, 1, 1)$ at the outputs as

$$\begin{aligned} P_{111} &= \frac{1}{9} (2 - r_{ab}^2 - r_{bc}^2 - r_{ac}^2 + 2r_{ab}r_{bc}r_{ac} \times (e^{i\varphi_{abc}} + e^{-i\varphi_{abc}})) \\ &= \frac{1}{9} (2 - r_{ab}^2 - r_{bc}^2 - r_{ac}^2 + 4r_{ab}r_{bc}r_{ac} \times \cos \varphi_{abc}). \end{aligned} \quad (3.37)$$

In the first line there are six distinct terms and each is associated with an element of the symmetric group S_3 . In particular, the first constant term corresponds to the identity element capturing probabilistic scattering, and the next three terms are two-photon interference contributions, as for HOM, resulting from binary swaps of particles. The last two terms come from the elements of S_3 that fully permute the particles and, thanks to the associated partial exchange symmetry, they contribute a dependence on the triad phase as mentioned in Section 3.1.1. The scope for non-monotonic coincidence statistics is apparent: the two-photon interference terms act to decrease coincidences but the value of the triad phase determines whether the three-photon interference terms in $\cos \varphi_{abc}$ reinforce or compete against the lower-order contributions.

We can also determine the probabilities of detecting partially bunched outcomes where two of the three photons end up in the same output, and also for fully bunched where all emerge at

the same output:

$$\begin{aligned}
P_{012} &= P_{120} = P_{201} = \frac{1}{9} (1 + 2r_{ab}r_{bc}r_{ac} \times \cos(\varphi_{abc} + 2\pi/3)), \\
P_{021} &= P_{102} = P_{210} = \frac{1}{9} (1 + 2r_{ab}r_{bc}r_{ac} \times \cos(\varphi_{abc} - 2\pi/3)), \\
P_{003} &= P_{030} = P_{300} = \frac{1}{27} (1 + r_{ab}^2 + r_{bc}^2 + r_{ac}^2 + 2r_{ab}r_{bc}r_{ac} \times \cos \varphi_{abc}).
\end{aligned} \tag{3.38}$$

The equality of sets of statistics and the independence of partially bunched statistics on pairwise distinguishabilities derive from the high symmetry of the tritter interferometer. In practice the imbalanced tritter we use affects these expressions slightly but this can be included in simulations of experiments. Tuning the four distinguishing parameters opens up a large landscape of interference statistics, and in the following sections we map out parts of this space.

3.4 Exploring distinguishability using temporal delays

3.4.1 State preparation and experimental setup

By analogy with the original HOM experiment, the use of temporal delays to explore multi-photon distinguishability is widespread [106, 108, 241–244]. For three photons there have already been experimental investigations into how the two relative time delays between three otherwise indistinguishable photons affect interference in a balanced tritter [166, 167]. However neither of these experiments achieved strong three-photon interference: the novel non-monotonic features of the interference – that we will present shortly – did not approach the ideal visibility, mostly due to poor state indistinguishability. We improve on those results and thus confirm the high performance of our source and its ability to effectively probe multiparticle interference.

We consider preparing three photons in identical wavepackets and explore interference by tuning temporal distinguishability using the two relative delays. The three identical wavepackets generated by the SPDC sources in a semi-heralded configuration have a Gaussian spectrum:

$$f(\omega) = (\pi\sigma_\omega^2)^{-1/4} e^{-\frac{(\omega-\omega_0)^2}{2\sigma_\omega^2}}, \quad \Delta\omega = 2\sqrt{2 \ln 2} \sigma_\omega, \tag{3.39}$$

where ω_0 is the central frequency, $\Delta\omega$ is its bandwidth, and σ_ω is the standard deviation of the distribution. We can label the state of a particle with this wavepacket structure and an arrival time t_i by

$$|t_i\rangle = \hat{A}^\dagger(t_i) |0\rangle = \int d\omega e^{i\omega t_i} f(\omega) \hat{a}^\dagger(\omega) |0\rangle. \tag{3.40}$$

The overlap between two identical Gaussian wavepackets with arrival times t_i and t_j is then

$$\begin{aligned}
\langle t_i | t_j \rangle &= (\pi\sigma_\omega^2)^{-1/2} \int d\omega e^{-i\omega(t_i-t_j)} e^{-\frac{(\omega-\omega_0)^2}{\sigma_\omega^2}} \\
&= e^{-\frac{\sigma_\omega^2}{4}(t_i-t_j)^2 - i\omega_0(t_i-t_j)}.
\end{aligned} \tag{3.41}$$

We set the photon entering the second input to have a fixed arrival time t_0 . The relative delays of those entering the first and third ports, respectively τ_1 and τ_2 , are varied (see Figure 3.12).

Hence the input states and their overlaps are

$$\begin{aligned}
 |a\rangle &= |t_0 + \tau_1\rangle, & \langle a|b\rangle &= e^{-\frac{\sigma_\omega^2}{4}\tau_1^2 - i\omega_0\tau_1}, \\
 |b\rangle &= |t_0\rangle, & \langle b|c\rangle &= e^{-\frac{\sigma_\omega^2}{4}\tau_2^2 + i\omega_0\tau_2}, \\
 |c\rangle &= |t_0 + \tau_2\rangle, & \langle c|a\rangle &= e^{-\frac{\sigma_\omega^2}{4}(\tau_2 - \tau_1)^2 - i\omega_0(\tau_2 - \tau_1)}.
 \end{aligned} \tag{3.42}$$

Turning now to the scattering probabilities of equations 3.37 and 3.38, the pairwise distinguishabilities r_{ij}^2 do not have a dependence on the overlaps' arguments. For these wavepackets it is straightforward to show that phases cancel and $\langle a|b\rangle\langle b|c\rangle\langle c|a\rangle$ is real and positive, leaving the triad phase $\varphi_{abc} = 0$. It turns out that this holds as long as the spectral modes have symmetric intensity about the same mean frequency [144]. For this state preparation there are therefore just two parameters that can be adjusted independently: τ_1 and τ_2 control r_{ab} and r_{bc} respectively, and also fix r_{ac} . It is possible to use the delays to tune from completely distinguishable states where all photons are walked off temporally, to fully indistinguishable when they arrive at the interferometer simultaneously, as well as intermediate regimes where some states overlap more than others.

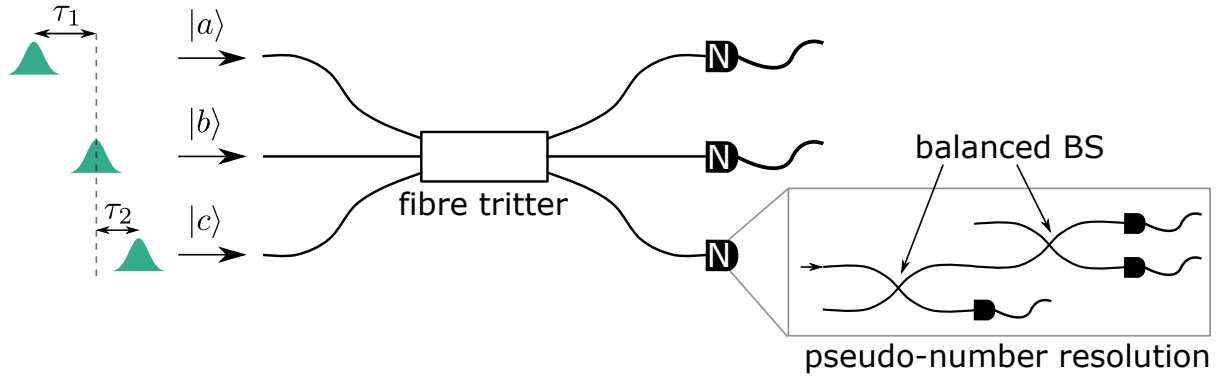


Figure 3.12: Three spectrally indistinguishable photons are injected into the ports of a fibre tritter interferometer, with polarisations matched at the coupling region using sets of paddles on the input fibres (not shown). The two relative time delays τ_1 and τ_2 are controlled using motorised delay stages to prepare the states $|a\rangle, |b\rangle, |c\rangle$. Counting statistics are recorded by spatially multiplexing APDs using pairs of balanced beam splitters (BS) at the tritter outputs, as shown in the inset. A correlator with 2ns coincidence window records heralded threefold coincidences across the nine detectors at the outputs.

Experimentally we use the three spectrally indistinguishable photons generated by the pair of SPDC sources in a heralded configuration, as shown in Figure 3.5. The source fibres are connected to the tritter interferometer and polarisation compensation ensures the polarisations are matched, up to some common unitary rotation, at the coupling region. The correspondence between source arms and prepared states is $A \rightarrow |c\rangle, B \rightarrow |a\rangle, C \rightarrow |b\rangle$. In order to determine the stage positions corresponding to full temporal overlap, source arms can be blocked to perform first a single source HOM dip for A and B, and then a dip between B and C. The latter has lower visibility because it amounts to interfering two thermal states. These measurements also tell us how large the delays must be for a certain temporal distinguishability of a pair. In order to collect full counting statistics we spatially multiplex nine APDs using layers of balanced fibre

beam splitters connected to the tritter outputs (see inset of Figure 3.12), and use a correlator (Swabian TimeTagger Ultra) to record heralded coincidences (one herald channel and nine for the output ports). This pseudo-number resolution probabilistically permits identification of partially and fully bunched output events. In Appendix C we show how to correct for this non-deterministic number resolution in order to calculate event probabilities from counting statistics.

3.4.2 Results and discussion

Using the SPDC sources, we repeatedly scan $320\mu\text{m}$ of total path difference for each of the states $|a\rangle$ and $|c\rangle$ to build up a 23×23 grid of temporal delays, including regimes for full distinguishability and indistinguishability, for which interference statistics are collected. We run the sources with squeezing parameter $\lambda \approx 0.1$ to balance collection rates with minimising contamination due to unwanted higher order emissions. A week of data collection yielded a total of over 430,000 heralded threefold events and a subset of experimentally determined probabilities are shown in Figure 3.13, along with simulated signals.

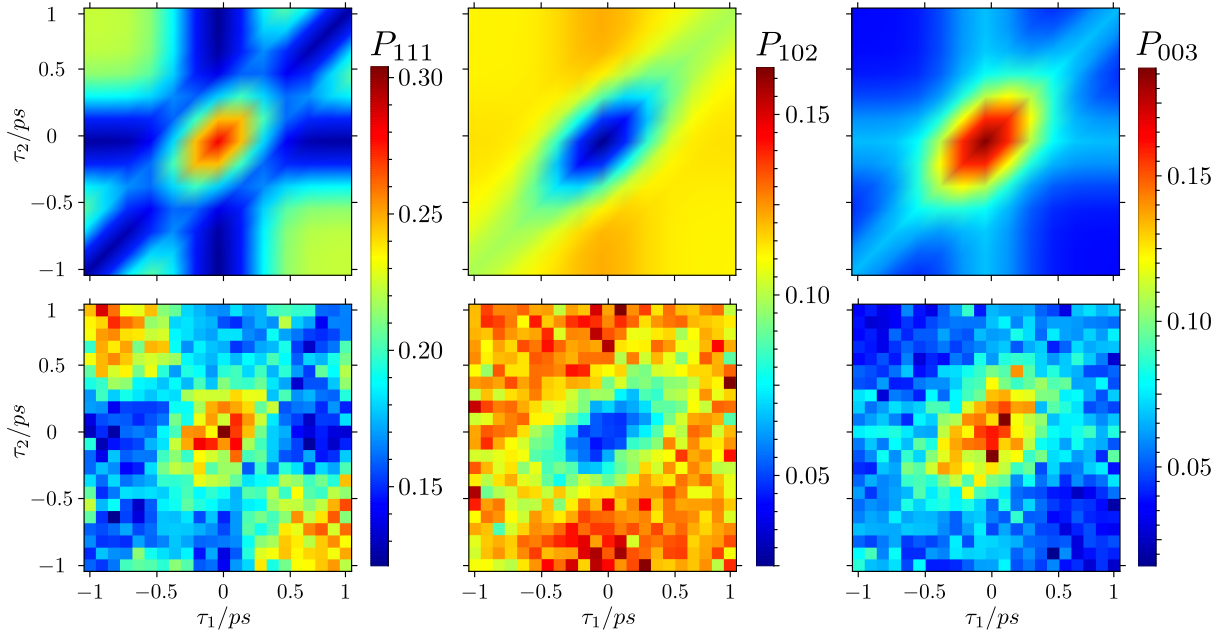


Figure 3.13: We use two SPDC sources to map the full time dependence of three-photon interference in a tritter. The top row are simulated signals using the reconstructed tritter unitary (equation 3.35) and including imperfections like distinguishability and higher-order emissions (see main text for details). The bottom row shows experimentally determined output probabilities from the fibre tritter as a function of the relative delays τ_1 and τ_2 . The probabilities plotted are those for which the most counts were recorded, respectively 100k, 80k and 20k.

The first thing to notice is the non-monotonicity of the coincidence probability P_{111} . Concentrating on these lefthand plots in Figure 3.13 and beginning at the top left, all photons are temporally distinguishable and $P_{111} \approx 0.23$ (green region of simulated plot). Tracing along the line towards the bottom right, the delays τ_1 and τ_2 decrease in magnitude until each of these wavepackets overlaps with the third. Terms in the pairwise distinguishabilities r_{ij}^2 now decrease P_{111} to ~ 0.16 (narrow blue region). Next the three-photon term starts contributing

and increases the coincidences until, when $\tau_1 = \tau_2 = 0$ at the centre, it overwhelms two-photon terms and P_{111} rises above the level for distinguishable photons (red region in the centre). This variation then repeats on the way back to full distinguishability at the bottom right (green region of simulated plot). This non-monotonicity is a well-known feature for interference of more than two photons that arises from the competition of multiphoton contributions, and reveals the richer behaviour accessible for larger systems [20, 107]. It is possible to associate the other dark blue regions with temporal delays where two-photon interference dominates.

The high symmetry of the tritter means that the partially bunched statistics, such as P_{102} in Figure 3.13, ideally have no dependence on pairwise distinguishabilities alone, and instead are only affected by interference when all three photons share some overlap. Therefore at zero relative delay, when $\tau_1 = \tau_2 = 0$, such output events are Fourier suppressed and ideally are never observed [245]. The fully bunched statistics (P_{003} in Figure 3.13) are increased by two- and three-photon interference, giving rise to the light blue and red regions in those plots.

The simulated signals plotted in the top row of Figure 3.13 include the effect of residual distinguishability $r_{ij} = 0.95$ arising from imperfect polarisation and spectral preparations, as determined from HOM dip measurements. They also include the reconstructed tritter unitary and contamination from higher-order source emissions based on $\lambda = 0.1$ for the SPDC sources with 100mW blue pump power, combined with the effect of 75% input and output transmissions for coupling to the interferometer (see Appendix B for more details on the simulation). There is excellent qualitative and quantitative agreement between simulation and experiment. For example there should be full suppression of P_{102} at zero delay given a balanced tritter and indistinguishable photons. We observe around 75% which is consistent with that predicted by our simulation. The contrast of features in the other statistics also agree with the simulation.

Another important observation is that three-photon exchange terms are modulated by $r_{ab}r_{bc}r_{ac}$, meaning signals with this dependence are very sensitive to unintended distinguishability. This is the dominant factor reducing the prominence of the associated features experimentally. On the other hand, it turns out that the features are relatively robust to imbalances in the tritter interferometer, much more so than the HOM dips mentioned in Section 3.3.2 when characterising the tritter. We attribute this to the relative sensitivities of the permanents involved in equation 2.54: those for small sub-matrices are influenced more by imbalanced elements than larger matrices, where slight imbalances tend to average out.

Using two relative time delays and three photons allows us to probe an effective qutrit space, since it is always possible to perform a Gram-Schmidt procedure to define three orthogonal basis states (see Appendix B.4). This is the Hilbert space dimension needed to access all distinguishing parameters for three photons, as shown in Section 3.1.2. However tuning the relative delays of three temporal modes allows independent control of only two of the four distinguishing parameters for three photons, and Gaussian temporal modes only access $\varphi_{abc} = 0$. In fact equations 3.36, 3.37 and 3.38 show that independent measurement of the pairwise distinguishabilities r_{ij}^2 would permit full reconstruction of these three-photon results. In order to access the triad phase we need an extra degree of freedom: we use polarisation.

3.5 Varying the triad phase

3.5.1 State preparation

The ability to manipulate the states' polarisations allows control over the triad phase as we saw earlier in Figure 3.1, where φ_{abc} was defined as half the solid angle enclosed by three vectors in the Bloch sphere. The three-photon exchange term for the coincidence probability given in equation 3.37 has a dependence on $\cos \varphi_{abc}$. We have just seen the case where $\varphi_{abc} = 0$ using identical wavepackets and, when the photons arrived simultaneously, this contribution counteracted the two-photon terms, increasing coincidences and leading to the observation of a peak in P_{111} . The other extreme occurs when $\varphi_{abc} = \pi$, where the three-photon contribution will instead act to suppress coincidences. This can be achieved by preparing polarisation states whose Bloch vectors lie at the vertices of an equilateral triangle in the Bloch sphere, as shown in Figure 3.14b. The product of the polarisation overlaps $\langle a|b\rangle \langle b|c\rangle \langle c|a\rangle$ is negative and so the argument φ_{abc} is π .

In order to highlight the different qualities of various exchange contributions in the coincidence probability P_{111} , we consider preparing either indistinguishable polarisations so $\varphi_{abc} = 0$ or those that define $\varphi_{abc} = \pi$. We then vary the relative time delays of two of the wavepackets symmetrically around the third which is left fixed, so $\tau_1 = -\frac{\tau}{2}$ and $\tau_2 = +\frac{\tau}{2}$. In this way the photons can be initialised in fully distinguishable states: the wavepackets have no overlap and the statistics are unaffected by interference. As the relative delay decreases, pairs of wavepackets will start to overlap and two-photon interference terms mediated by the pairwise distinguishability will decrease coincidences independent of φ_{abc} . Once all the wavepackets overlap, the term in $\cos \varphi_{abc}$ will contribute and either increase or decrease coincidences from the tritter depending on the polarisation configuration. The states we prepare for the two situations are:

$$\begin{aligned} |a\rangle &= \left|t_0 - \frac{\tau}{2}\right\rangle \otimes |H\rangle, & |a'\rangle &= \left|t_0 - \frac{\tau}{2}\right\rangle \otimes |H\rangle, \\ |b\rangle &= |t_0\rangle \otimes |H\rangle, & |b'\rangle &= |t_0\rangle \otimes \frac{1}{2} \left(|H\rangle + \sqrt{3}|V\rangle\right), \\ |c\rangle &= \left|t_0 + \frac{\tau}{2}\right\rangle \otimes |H\rangle, & |c'\rangle &= \left|t_0 + \frac{\tau}{2}\right\rangle \otimes \frac{1}{2} \left(|H\rangle - \sqrt{3}|V\rangle\right). \end{aligned} \quad (3.43)$$

To prepare the polarisation states $\frac{1}{2}(|H\rangle \pm \sqrt{3}|V\rangle)$ we send the emission from one of the tritter outputs through a quarter- then a half-wave plate, followed by a polarising beam splitter whose transmission is monitored using an APD. When the QWP and HWP are set at 60° and 30° respectively to the horizontal, the polarisation of state $|b'\rangle$ will be rotated to $|H\rangle$. Hence manipulation of paddles and compensation waveplates before the tritter to maximise counts after the PBS corrects for the random rotation in the input fibre of the tritter and achieves the desired polarisation state at the coupling region, up to some common unitary (see Appendix A). Similarly when these analysis waveplates are oriented at the equivalent negative angles and the same output port is monitored, the polarisation state of $|c'\rangle$ can be prepared.

3.5.2 Results and discussion

The data for this experiment was obtained using the SFWM source described in Section 3.2.7. Three herald detectors were required but the correlator used had only eight channels, so pseudo-number resolution was only performed on one output port. We prepare the polarisation states defining $\varphi_{abc} = 0, \pi$ and scan the relative delay τ using motorised delay stages whilst monitoring heralded coincidences at the tritter outputs, with results shown in Figure 3.14.

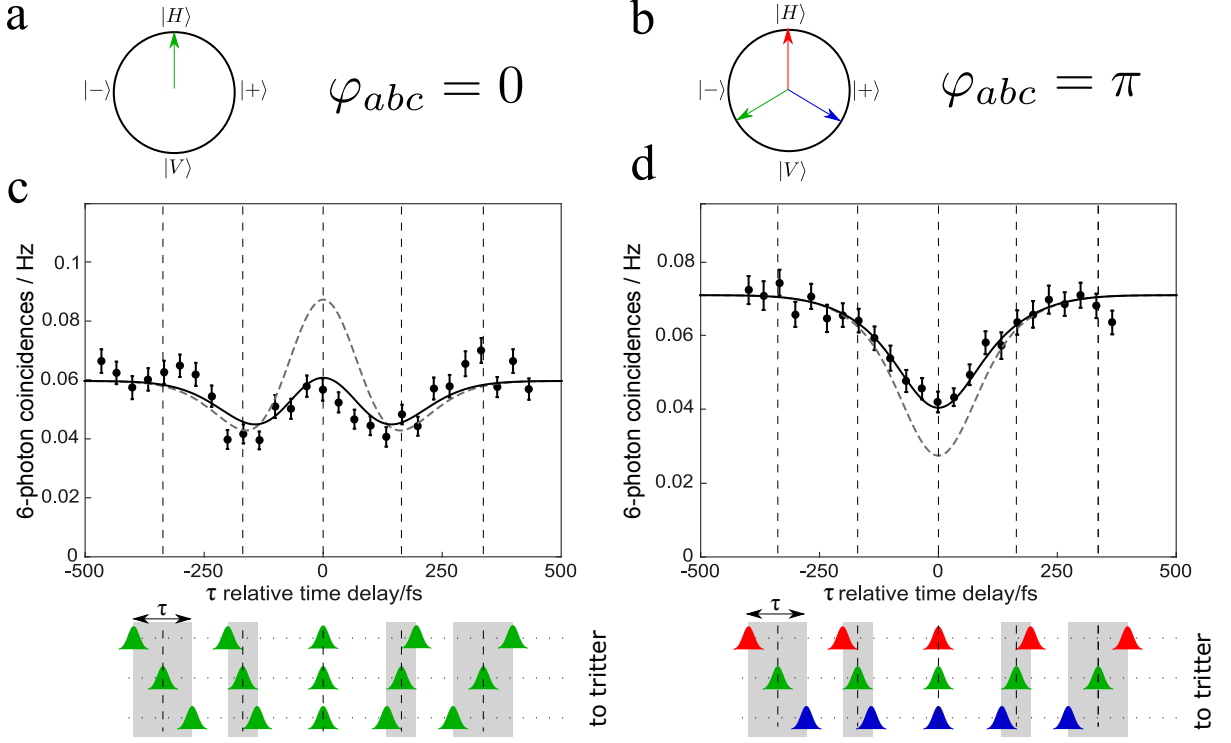


Figure 3.14: Measured threefold coincidences at the outputs of a tritter using the SFWM source and two different polarisation configurations. **a** Identical polarisations set the triad phase to zero. **b** Bloch vectors equally spaced in the plane of linear polarisation set the triad phase to π . **c,d** As the relative delay τ is varied, the behaviour of the coincidence signal can be associated with the underlying exchange contributions (see text for details). The dashed lines are curves for ideal theory and the solid lines are from simulations including various experimental imperfections mentioned in the main text. Error bars are obtained from repeated sweeps of the relative delay, and the total number of heralded counts is between 200 and 350 for **a**, and between 250 and 450 for **b**. From [144].

When the relative delay is large at the lefthand side of the two plots in Figure 3.14, the photons are temporally distinguishable and coincidence counts are constant. As τ decreases in magnitude, approaching zero for fully overlapped wavepackets, there is a dip in coincidences due to two-photon interference, mediated by the pairwise distinguishabilities r_{ij}^2 . When the relative delay is small enough that all three photons have some temporal overlap, three-photon interference starts to contribute a term cubic in the overlap magnitudes and as $\cos \varphi_{abc}$ in the triad phase. For $\varphi_{abc} = 0$ this term overwhelms the two-photon terms and a peak in counts is observed (this is precisely the same behaviour we saw when taking a diagonal slice through the P_{111} plots of Figure 3.13 earlier). On the other hand, when $\varphi_{abc} = \pi$ this term acts to decrease counts in the same way as two-photon terms, giving monotonic behaviour. This highlights how a collective phase can drastically alter the shape of interference statistics, and also that the

intuition from HOM – that indistinguishable states give the strongest contrast – does not apply to larger systems.

Whilst the different qualitative features of interference are clear in Figure 3.14, there is a marked difference between the ideal theory curves (dashed line) and the acquired data. Using the same simulation code employed in Figure 3.13, we include the effects of residual distinguishability from imperfect polarisation preparations by adding a distinguishability factor $r_{ij} = 0.95$. The three idler photons of the SFWM source pass through the same pair of bandpass filters but at different angles before interference. This means the edges of their spectral profiles are slightly shifted with respect to each other, thus limiting the ability to simultaneously filter out all undesired spectral components. As a result the sinc lobes shown in Figure 3.3 contribute some mixedness to the emission so the inferred purity $\mathcal{P} = 0.95$. Higher-order emissions are included based on $\lambda = 0.16$ and input and output coupling transmissions of 70%. An additional imperfection not present for the SPDC source but of critical importance in a simulation of the SFWM source is fluorescence. This was mentioned in Section 3.2.7 and we model it by assuming that distinguishable photons are created with probabilities P_S and P_I on the signal and idler arms respectively (see Appendix B.5 for further details). Hence in the presence of losses, faulty heralds and undesired scattering will contaminate statistics. The simulated signals are shown by solid lines in Figure 3.14 and are in excellent agreement with the experimental data.

The SPDC sources do not suffer from fluorescence noise and their emission has very high spectral purity when filtered. We can compare their statistics to those of the SFWM source for $\phi_{abc} = 0$ by selecting equivalent data from the temporal scan plots in Figure 3.13 presented earlier. By selecting points on the anti-diagonal of those plots, we obtain the signals in Figure 3.15. The peak in coincidences now rises above the background level for distinguishable photons, unlike for the SFWM source where impurity and noise suppressed it. The other counting statistics P_{102} and P_{003} also agree well with simulation.

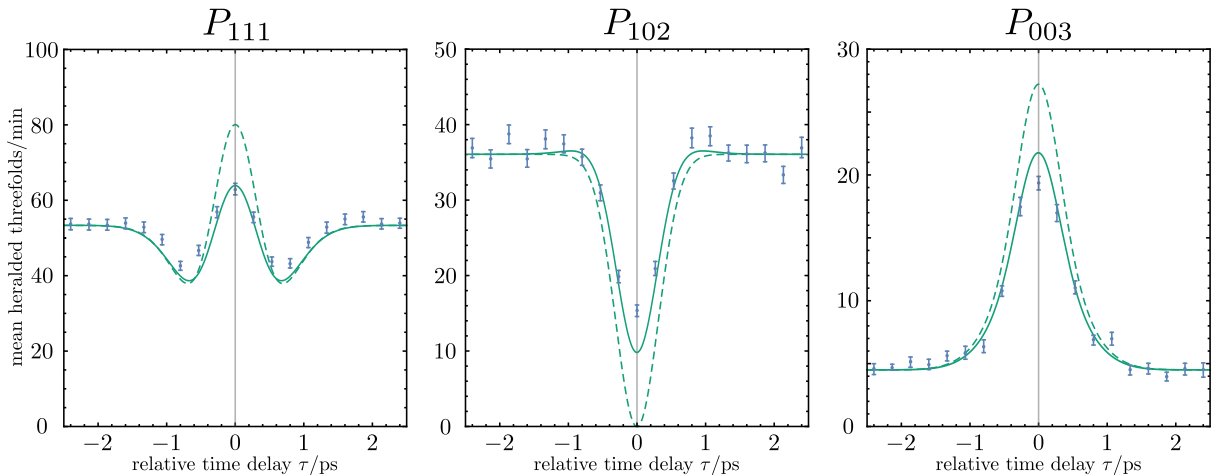


Figure 3.15: Measured threefold coincidences at the tritter output for $\varphi_{abc} = 0$ using the SPDC sources. The absence of noise photons and high spectral purity enables observation of a peak in coincidences that reaches above the background level for distinguishable photons. The dashed curves are for ideal theory and solid lines are from simulations described in Section 3.4.2. Error bars are obtained from repeated sweeps, and the total number of heralded counts are between 1200 and 1700 for P_{111} , 1000 to 400 for P_{102} and 110 to 520 for P_{003} .

As a final point here, the triad phase has been shown to have a classical counterpart that affects the interference of classical fields [246]. Rather than counting photons, normalised intensity correlation measurements are used to probe the character of interference. Surprisingly simulations of an experiment using a balanced tritter and equal intensity classical fields, prepared in the same way as for $\varphi_{abc} = 0$, do not exhibit the peak that we observe in the quantum case: our result cannot be described by a wave model. For more general three-mode interferometers and other state preparations, the shape of the signal with τ can be used to identify non-classicality of the input fields.

3.6 Isolating the triad phase

3.6.1 State preparation

The HOM dip in twofold coincidences demonstrates isolation of two-photon interference from varying single-particle contributions like those in the double slit experiment. Is it possible to observe three-photon interference independent of lower-order contributions? This would mean preparing a separable input state that allows variation of P_{111} whilst keeping two-photon interferences constant. Here that means permitting full variation of the triad phase whilst simultaneously maintaining constant pairwise distinguishabilities r_{ij}^2 (see equations 3.36 and 3.37). In Section 3.1.2 we saw that a two dimensional space is insufficient to control φ_{abc} independently of the three pairwise distinguishabilities: for that we require a qutrit. It is possible to access such a space through a combination of polarisation and temporal modes (see Appendix D).

Consider the polarisation preparation that earlier defined $\varphi_{abc} = \pi$ where the three Bloch vectors are equally spaced in the plane of linear polarisation. The triad phase can be tuned smoothly from 0 to 2π by rotating the polarisation of state $|a\rangle$ in the plane perpendicular to that of linear polarisations defined by the vectors for $|b\rangle$ and $|c\rangle$, that remain fixed (see Figure 3.16). The states and their associated overlaps are⁴:

$$\begin{aligned} |a\rangle &= |t_a\rangle \otimes (\cos 2\theta |H\rangle + i \sin 2\theta |V\rangle), & \langle a|b\rangle &= \langle t_a|t_b\rangle \times \frac{1}{2} \left(\sqrt{3} \cos 2\theta + i \sin 2\theta \right), \\ |b\rangle &= |t_b\rangle \otimes \frac{1}{2} \left(\sqrt{3} |H\rangle + |V\rangle \right), & \langle a|c\rangle &= \langle t_a|t_c\rangle \times \frac{1}{2} \left(\sqrt{3} \cos 2\theta - i \sin 2\theta \right), \\ |c\rangle &= |t_c\rangle \otimes \frac{1}{2} \left(\sqrt{3} |H\rangle - |V\rangle \right), & \langle b|c\rangle &= \langle t_b|t_c\rangle \times \frac{1}{2}. \end{aligned} \quad (3.44)$$

We set $t_b = t_c$ in order to maintain $r_{bc} = 1/2$. The symmetry of this preparation guarantees that $r_{ab}^2 = r_{ac}^2$ at all times. Consider starting with $|a\rangle$ in $|H\rangle$ and its wavepacket arrives at the interferometer slightly after the other two such that $r_{ab}^2 = r_{ac}^2 = 1/4$. As $|a\rangle$ rotates in the yz plane, φ_{abc} changes but its polarisation becomes less similar to those of $|b\rangle$ and $|c\rangle$. The associated pairwise distinguishabilities decrease, but we want these to be constant throughout variation of φ_{abc} . We now exploit the temporal degree of freedom such that the overlap of wavepackets can be used to compensate for changes in the overlaps of polarisation states and so maintain $r_{ab}^2 = r_{ac}^2 = 1/4$. This is done by exactly countering the change in polarisation overlap

⁴Compared to the states in equation 3.43 and Figure 3.14, this preparation for $\varphi_{abc} = \pi$ is reflected in the xy plane. This was done for experimental reasons, and is equivalent to swapping the definitions of $|H\rangle$ and $|V\rangle$.

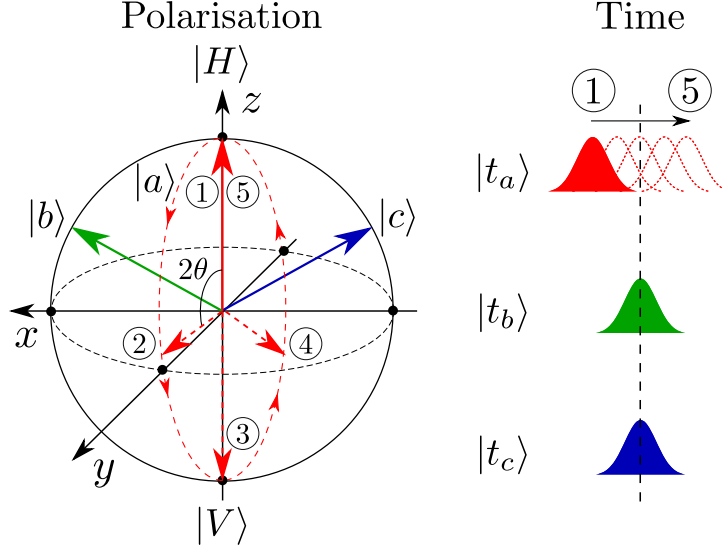


Figure 3.16: Preparation of three photons that allows variation of φ_{abc} whilst maintaining constant pairwise distinguishabilities. The triad phase is set by half the subtended solid angle in the Bloch sphere and is changed by rotating the polarisation of $|a\rangle$ in the yz plane as shown. Any change in the polarisation overlap magnitude can be compensated for by a corresponding change in the temporal overlap.

by varying the arrival time of $|t_a\rangle$ (see Figure 3.16). Specifically the overlap magnitudes are

$$\begin{aligned} r_{ab} &= \frac{1}{2} \sqrt{2 + \cos 4\theta} \times |\langle t_a | t_b \rangle|, \\ r_{ac} &= \frac{1}{2} \sqrt{2 + \cos 4\theta} \times |\langle t_a | t_c \rangle|, \\ r_{bc} &= \frac{1}{2} |\langle t_b | t_c \rangle|. \end{aligned} \quad (3.45)$$

Assuming identical Gaussian wavepackets as in equation 3.39, we now denote the variance in time $\sigma_\tau^2 = 1/\sigma_\omega^2$. Fixing $t_b = t_c$, we can now determine how to vary the delay of $|a\rangle$ as a function of θ in order to maintain all overlap magnitudes of $1/2$ by solving

$$r_{ab} = r_{ac} = \frac{1}{2} \sqrt{2 + \cos 4\theta} \times e^{-\frac{|t_a - t_b|^2}{4\sigma_\tau^2}} = \frac{1}{2}. \quad (3.46)$$

The relative delays must be varied with θ as

$$|t_a - t_b| = |t_a - t_c| = \sigma_\tau \sqrt{2 \ln(2 + \cos 4\theta)}. \quad (3.47)$$

The triad phase is given by the components in the polarisation space as

$$\varphi_{abc} = \text{Arg}(\langle a | b \rangle \langle b | c \rangle \langle c | a \rangle) = 2 \times \text{Arg}(\sqrt{3} \cos 2\theta + i \sin 2\theta). \quad (3.48)$$

3.6.2 Results and discussion

The data for this experiment was first obtained using the SFWM source described in Section 3.2.7. We prepare the polarisation of state $|a\rangle$ by sending a horizontally polarised photon through a HWP oriented at θ to the horizontal and then a QWP with its axis oriented along

the vertical. In order to correct for random rotations in the SMF to the tritter interferometer, polarisation compensation must be performed for both X and Z eigenstates to ensure this state remains in the yz plane of the Bloch sphere. The variance of the Gaussian wavepackets σ_τ^2 is determined by separate HOM dip measurements, and a motorised rotation mount and a delay stage allow variation of the waveplate angle θ and the arrival time t_a . The angle θ is varied from 0 to $\pi/2$ and the relative delay is adjusted according to equation 3.47. The heralded nature of this source means we can also monitor the situations where only two of the three sources fired, allowing independent measurement of two-photon interference. This and the three-photon results are shown in Figure 3.17.

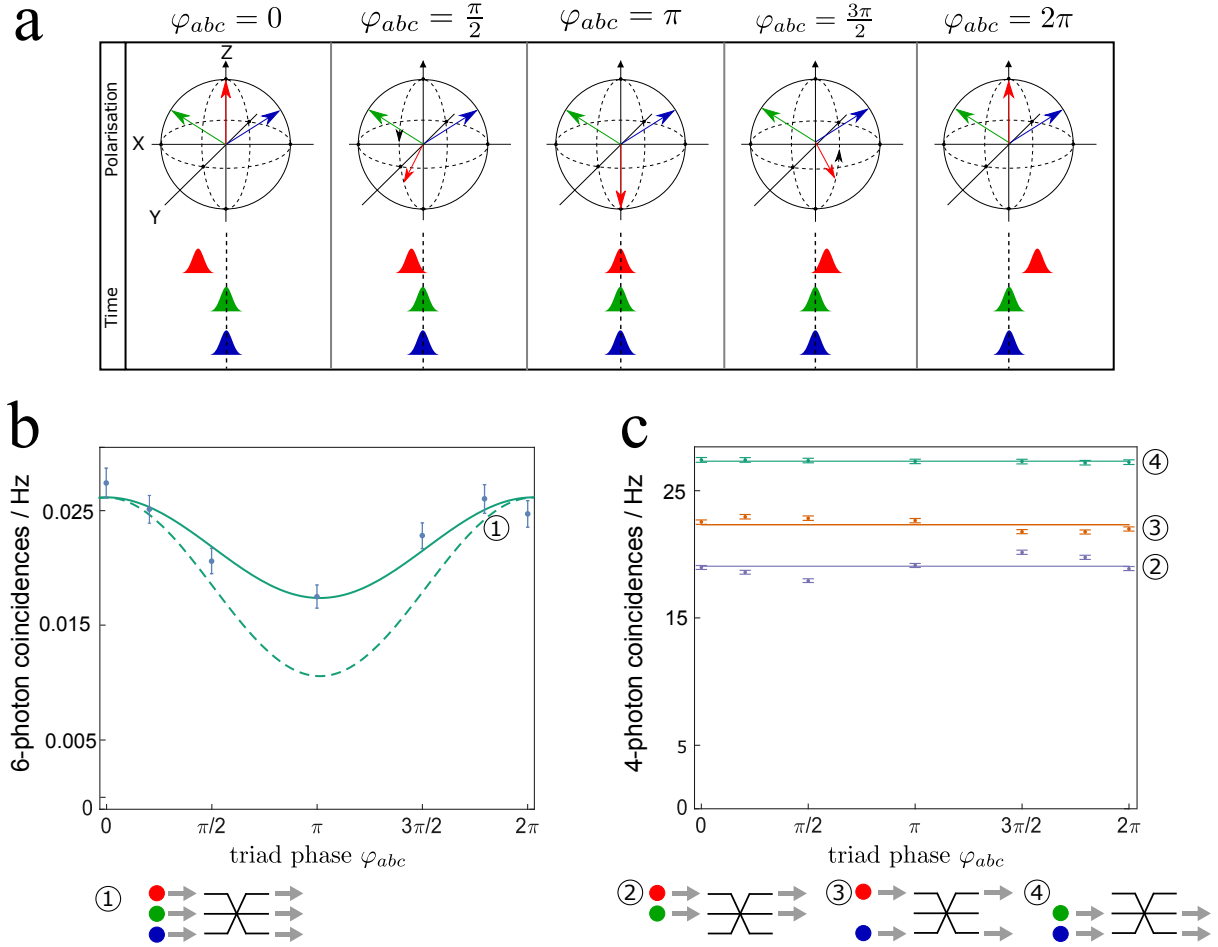


Figure 3.17: **a** Preparation of photons in polarisation and time that varies φ_{abc} but maintains constant pairwise distinguishabilities. **b** Mean heralded three photon coincidences at the outputs of the tritter interferometer using the SFWM source. As the triad phase is varied, a cosine variation is observed. The dashed curve is for ideal theory, and the solid curve is from a simulation accounting for imperfections that is described in the main text. Errors bars are obtained from repeated sweeps, and the total number of counts is between 330 and 550 per point. **c** Mean heralded twofold rates corresponding to the different input and output configurations shown. From [144].

We observe the expected cosine variation in output threefold coincidences with the triad phase but with a visibility of 34%, less than the ideal 57% (dashed line in Figure 3.17b). We simulated this SFWM experiment using the same parameters as for Figure 3.14. The signal is shown as a solid line in Figure 3.17b and is consistent with measurement.

3. Three-photon interference

Twofolds are approximately constant (see Figure 3.17c), with an average variation of below 6% that cannot be responsible for the large variation in threefold coincidences. We attribute fluctuations in the twofolds to imperfect state preparation: the polarisation of $|a\rangle$ does not rotate exactly in the yz plane, leading to sinusoidal fluctuations of the twofolds labelled ‘2’ and ‘3’ in the Figure. Singles counts varied by a maximum of 3% for the same reason.

These results highlight that distinguishability constitutes a *collective* description of the similarity of particles. Throughout the variation of the triad phase, the pairwise distinguishabilities of the states do not change and HOM measurements are close to flat. The strength of interference between a pair of photons by themselves is constant throughout, analogous to the concept of pairs of states being ‘in phase’ in the Pancharatnam connection [163]. The triad phase captures the partial exchange symmetry and distinguishability of all three photons, and enables control of three-photon interference independent of lower order contributions.

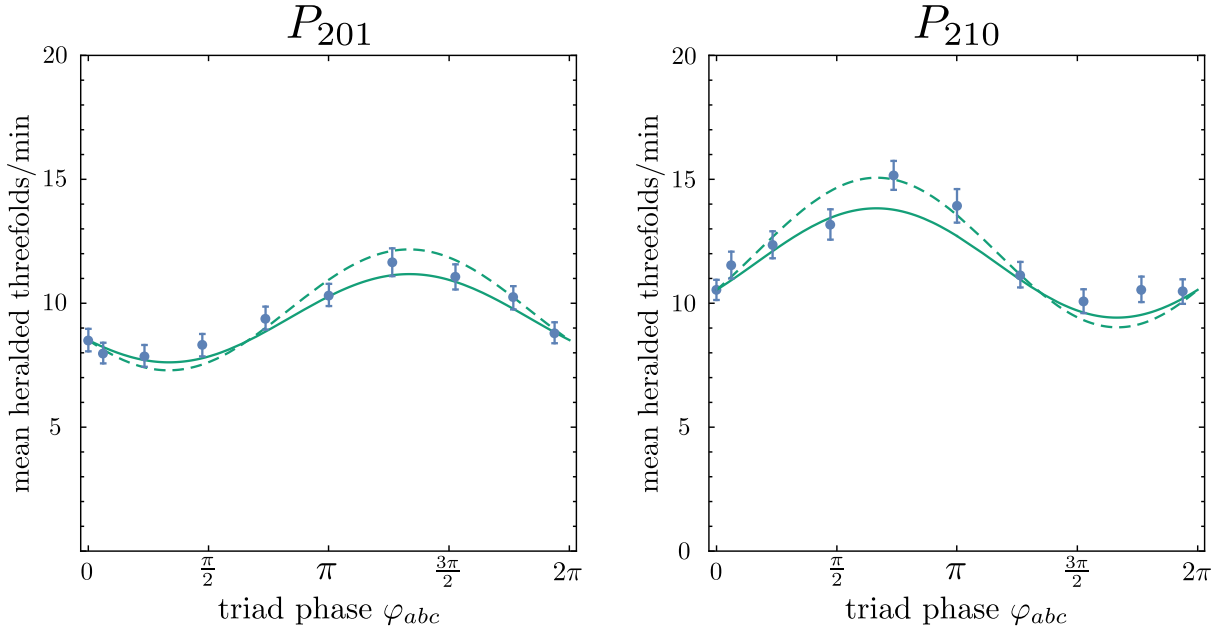


Figure 3.18: The dependence of partially bunched statistics on the triad phase, measured using SPDC sources. Unlike coincidences that have a $\cos \varphi_{abcd}$ dependence, these are predicted to exhibit a dependence on $\cos(\varphi_{abc} \pm 2\pi/3)$. The dashed curves are ideal theory and the solid curves are from a simulation of the experiment using the same parameters as for Figure 3.13. Error bars are obtained from repeated sweep, and the total number of counts are between 350 and 510 for P_{201} , and between 450 and 650 for P_{210} .

Due to losses in the system this first run did not acquire sufficient statistics to resolve features in the partially bunched counts. A second run was performed using the SPDC sources with full pseudo-number resolution on the tritter outputs. Fluctuations in the twofolds from a single source averaged 7%, again due to errors in polarisation preparation. Partially bunched threefolds recorded using pseudo-number resolved detection are shown in Figure 3.18, with visibilities of $\sim 30\%$ that cannot be attributed to changes in twofolds.

For the ideal tritter $U_{trit}^{(1)}$ given in equation 3.33, P_{201} and other partially bunched probabilities related by cyclic permutations of output occupation numbers are predicted to depend on $\cos(\varphi_{abc} + 2\pi/3)$. Meanwhile P_{210} and those related cyclically should have a $\cos(\varphi_{abc} - 2\pi/3)$ shape. These dependences swap if the interferometer is instead described by the other canonical

tritter $U_{trit}^{(2)} = \left(U_{trit}^{(1)}\right)^*$. This opens up the possibility of unambiguously discriminating between a unitary and its conjugate. As shown in equation 3.34, HOM dips can only resolve relative phases in an interferometer and so conventionally the first non-zero phase in a scattering matrix is taken to be positive [239]. For multiparticle interference, the interplay of the triad phase with the phases of the interferometer dispenses with the need to make such an assumption. The fitted shifts are $+(0.68 \pm 0.04)\pi$ and $-(0.73 \pm 0.04)\pi$ respectively for P_{201} and P_{210} , consistent with predicted values using the reconstructed unitary of equation 3.35 and confirming our choice of sign for its phases.

3.7 Conclusions

We have shown that threefold partial exchange symmetry of the wavefunction for three photons in separable states gives rise to a new distinguishing parameter: the triad phase. A full exploration of the interference landscape accessible using temporal delays of identical wavepackets was performed experimentally using a pair of SPDC sources, and demonstrated the non-monotonicity of multiparticle interference. Polarisation allows access to the triad phase and this was shown to tune the three-photon contribution to interference statistics. We also saw that photons prepared in states with low overlap can exhibit larger variations in statistics than indistinguishable states. This challenges the intuition from one- and two-photon experiments that reduced overlap causes lower interference visibility. In the next chapter this is taken to the extreme: distinguishable photons can still exhibit multiparticle interference.

The triad phase's effect in interference was isolated from terms in the pairwise distinguishability by using a combination of polarisation and temporal modes. Measured threefold output coincidences exhibit a variation with the phase that cannot be attributed to two-photon interferences, which are monitored and remain flat. This confirms that a collective description of multiparticle distinguishability is necessary, even if photons are prepared in separable states. Another approach to isolate three-photon interference using entangled states has previously been proposed [247, 248] and was demonstrated experimentally at the same time as our approach [249]. In Chapter 6 we investigate the differences between these two preparations of states of the electromagnetic field that exhibit N -fold correlations but none in lower orders.

It is clear that the introduction of a third photon opens up an enormously richer interference landscape than that accessible in two-photon experiments. The extra control afforded by such multiparticle phases could find applications in quantum state engineering, and in Appendix E we show how different types of postselected entangled states of three photons can be produced by adjusting distinguishability parameters. This control could be applied to protocols for entangling matter systems [250, 251]. Additionally the role of such phases in the computational complexity of boson sampling with partially distinguishable particles is an interesting problem to explore [132, 133]. In the next section we study the multiparticle phase defined for a separable state of four photons and demonstrate another surprising property of multiphoton interference.

Interfering distinguishable photons

Our considerations in the previous chapter have highlighted the need to treat distinguishability as a collective concept, with multiparticle phases becoming important for more than two photons. Here we use a graph model to reveal which distinguishing phases are independent parameters for particles in separable states. This leads us to the surprising discovery that, for four or more particles, multiparticle interference is possible even if some of the states involved are distinguishable. We present a preparation of the states of four photons that ensures each is pairwise distinguishable to another but allows tuning of a four-particle phase. We generate four photons in these states using a pair of SPDC sources and interfere them in a balanced bulk optic four-mode coupler. The observation of variations in interference statistics with this phase, independent of lower-order exchange contributions, confirms this effect.

This project was devised by myself, AJM and Valery Shchesnovich. AJM found the preparation of four photons' states that isolates distinguishable state interference. I performed the experiment and data analysis, with assistance from AJM on theory and simulations, and from Tom Wolterink and Thomas Hiemstra when designing the interferometer.

4.1 Graph model for distinguishability in interference

4.1.1 Distinguishability parameters

We have seen that a single parameter – the pairwise distinguishability r_{ij}^2 – quantifies the similarity of two photons in separable pure states. For three photons there are four parameters: three pairwise distinguishabilities and the triad phase. We can determine how this scales for N photons by revisiting the distinguishability matrix (equation 2.53), defined as the Hermitian $N \times N$ matrix of complex state overlaps:

$$\mathcal{S} = \begin{pmatrix} 1 & \langle \phi_1 | \phi_2 \rangle & \dots & \langle \phi_1 | \phi_N \rangle \\ \langle \phi_2 | \phi_1 \rangle & 1 & \dots & \langle \phi_2 | \phi_N \rangle \\ \vdots & \vdots & \ddots & \vdots \\ \langle \phi_N | \phi_1 \rangle & \langle \phi_N | \phi_2 \rangle & \dots & 1 \end{pmatrix}. \quad (4.1)$$

In general there are $N^2 - N$ parameters in such a matrix. However there is freedom to define physically irrelevant global phases on each input state and so $(N - 1)$ phases can be removed. This leaves $(N - 1)^2$ parameters, of which $N(N - 1)/2$ are overlap moduli r_{ij} and $(N - 1)(N - 2)/2$ are phases φ_{ij} . How do these phases combine to define independent distinguishing parameters?

4.1.2 Graph model

The role of multiparticle distinguishability in interference can be determined by choosing overlaps from the distinguishability matrix according to elements of the symmetric group (see equation 2.54). For example we have seen that for three particles the transposition $\sigma = (2, 1)$ leads to a dependence on r_{ab}^2 , and that the triad phase arises from the full permutation $\sigma = (2, 3, 1)$. These terms may be identified with *cycles* of the symmetric group. Every element of S_N has a *cycle structure* that describes the action of that element in terms of cyclic permutations of objects. For example, the element $\sigma = (2, 1, 3)$ in S_3 is described by the cycles $\mu(\sigma) = (12)(3)$. The elements in parentheses are written so that the first element goes to the position of the second, the second to that of the third, etc. until the final element takes the position of the first one. A lone element in parentheses is not permuted. A useful graph model has been developed by my colleague AJM and independently by Shchesnovich *et al.* [252] that exploits cycles to keep track of distinguishability contributions for N interfering particles (see Figure 4.1).

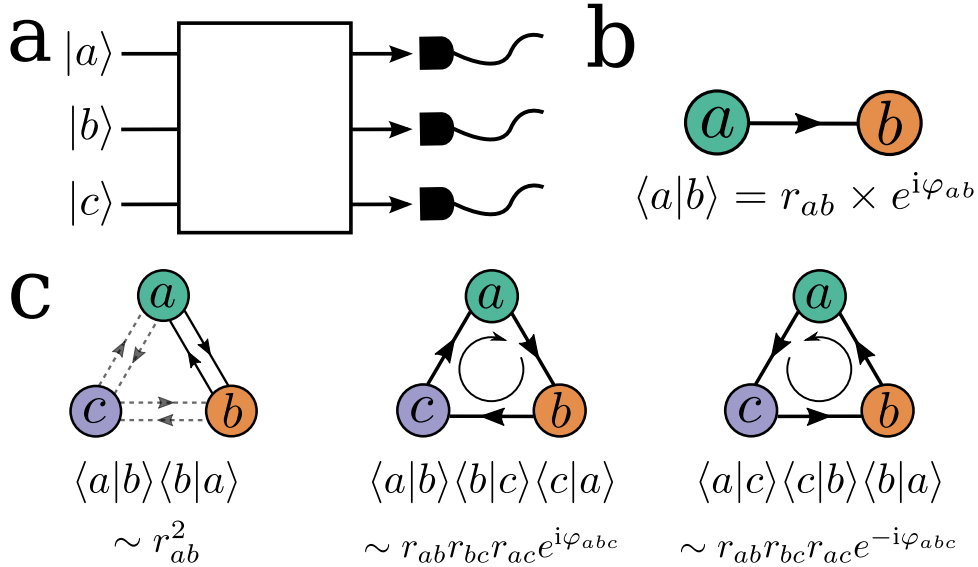


Figure 4.1: **a** Three photons prepared in separable states are injected into a fully-connected interferometer. **b** The directed edge between two vertices has a weight given by the overlap of the connected states. **c** The phase dependence of the overlaps cancels for the three HOM-type contributions like r_{ab}^2 . The elements $\sigma = (2, 3, 1)$ and $\sigma = (3, 1, 2)$ give dependences on the triad phase $\varphi_{abc} = \varphi_{ab} + \varphi_{bc} + \varphi_{ca}$, and summing these gives a $\cos \varphi_{abc}$ dependence in coincidence statistics (as seen in equation 3.37).

N vertices on a 2-dimensional graph are associated with the states of the N interfering particles. Directed edges have an accompanying weight given by the overlap of the states on the connected vertices, where the state at the starting vertex appears as a bra and the end state appears as a ket. An edge is omitted if the two states have zero overlap and this corresponds

to distinguishability in the accompanying *interfering* paths¹. Each element of S_N corresponds to a covering of the graph where closed, directed paths ensure every vertex is visited at most once. The cycle structure of the element indicates how to perform the covering. For example $\mu(\sigma) = (12)(3)$ comprises a loop over the vertices corresponding to the states in inputs 1 and 2, and the vertex for the third input is connected to itself (the loop is omitted) indicating that that particle is not exchanged so there is no dependence on its state. The product of weights for each covering gives a term in the interference statistics and describes the distinguishability parameter for that exchange contribution, exactly as in equation 2.54 (see Figure 4.1).

With this picture in mind it is straightforward to read off exchange contributions to interference. M -particle interference is captured by a closed path that traverses M vertices, and N -particle interference is possible if a loop can be drawn that visits every vertex. A caveat is that if the relevant matrix permanent (showing up in equation 2.54) is zero (either due to zeros in the matrix, or interplay of phases) then that distinguishability parameter will not appear in statistics. We saw this for the partially bunched statistics using an ideal tritter in equation 3.38, where there was no r_{ij}^2 dependence.

4.2 Four-particle interference

4.2.1 Decomposing using triad phases

The interference of four indistinguishable photons has drawn interest through comparisons with HOM interference and demonstrations of boson sampling [100, 125, 126, 253–256], and theoretical investigations have probed the non-monotonicity of statistics with distinguishability [107, 143, 257].

The interference of four photons prepared in separable states labelled a, b, c, d and injected in different ports will depend on pairwise distinguishabilities and triad phases that can be determined by paths like in Figure 4.1. Additionally full permutation leads to terms in the four-particle phase $\varphi_{abcd} = \varphi_{ab} + \varphi_{bc} + \varphi_{cd} + \varphi_{da}$ (see Figure 4.2). If all states have some mutual overlap then the four-particle phase can be decomposed into triad phases. This is shown in Figure 4.2c where the phase φ_{abcd} associated with the loop through four vertices is the same as the sum of triad phases for the loops depicted. This means that separate two- and three-photon measurements provide enough information to entirely describe such four-photon interference.

This turns out to hold for N interfering particles whose states all share some mutual overlap [252]. Intuitively any closed loops on a fully connected graph can be decomposed into triangular paths where the phases on common edges cancel. HOM measurements yield r_{ij}^2 , and three-photon experiments give the triad phases that act as primitives defining all other multiparticle phases.

4.2.2 Distinguishable state interference

What happens if the connectivity of a graph is broken? As long as there is still a closed loop over M vertices, M -photon interference will persist [252]. For example, consider breaking the

¹The term ‘path’ appears when describing interference of indistinguishable processes, and also here in graph theory. It should be clear from context which definition is appropriate.

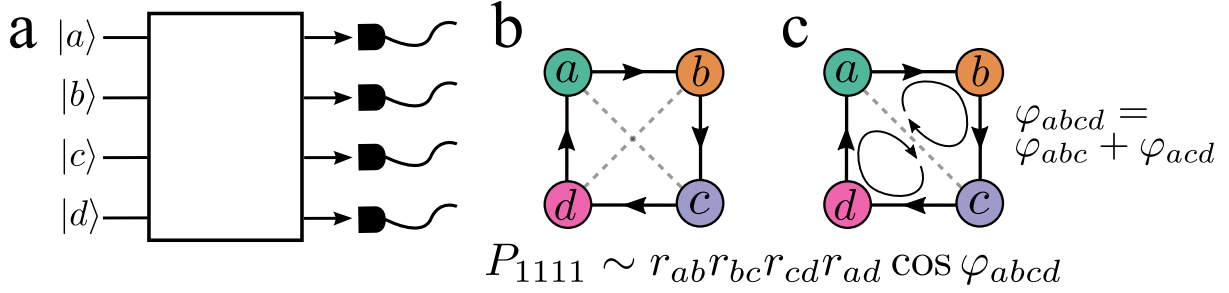


Figure 4.2: **a** Four photons prepared in separable states interfere in a multiport. **b** Graph describing four-particle interference, with a dependence on $e^{i\varphi_{abcd}}$ where $\varphi_{abcd} = \text{Arg}(\langle a|b\rangle\langle b|c\rangle\langle c|d\rangle\langle d|a\rangle)$. Traversing the graph in the other direction gives a dependence on $e^{-i\varphi_{abcd}}$, and summing these gives a $\cos \varphi_{abcd}$ dependence in the coincidence probability P_{1111} . **c** If the graph is fully connected, φ_{abcd} can be decomposed into triad phases because the phase φ_{ac} on the common edge cancels. The coincidence statistics are fully described by parameters that can be determined through two- and three-photon experiments.

connectivity of the graph for four photons by setting pairs of states to be distinguishable (see Figure 4.3). Two-photon interferences persist but there are no closed loops over three vertices. Hence triad phases are no longer defined and three-photon interference disappears, making it impossible to decompose the four-photon phase into triad phases. Four-photon interference depending on φ_{abcd} is still possible due to the loop over four vertices, and this phase is now an independent parameter. Despite some distinguishability in the states, multiparticle interference still occurs; this goes against intuition from HOM that distinguishability means no interference.

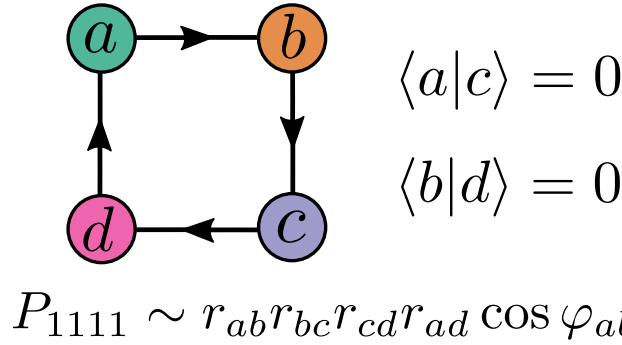


Figure 4.3: Preparing pairwise distinguishable states eliminates triad phases and three-particle interference from the system. However there is still a contribution depending on the four-particle phase φ_{abcd} .

This type of “circle-dance” interference persists for larger systems where the connectivity of the graph ensures that only two- and N -particle interference are possible. The overlap moduli r_{ij} achievable whilst imposing this distinguishability decrease for larger systems so multiparticle contributions quickly diminish. Generally as the connectivity of some graph deteriorates, the disappearance of triad phases means multiparticle phases become independent parameters [252].

Here the distinguishable state interference is solely due to the exchange symmetry of an initially separable multiparticle wavefunction. Other demonstrations with two photons have employed entangled states [95], masked distinguishing information [258, 259], or used nonlinear processes to ensure indistinguishability of interfering paths [260].

4.2.3 Pairwise distinguishable state preparation

To observe four-photon interference independent of lower-order contributions – similar to what we did for three photons in Section 3.6 – we must keep two- and three-photon distinguishabilities constant. We just saw that the only way for φ_{abcd} to be an independent parameter is by eliminating triad phases, and so we seek a preparation of states that realises the graph in Figure 4.3 and also maintains constant pairwise distinguishabilities r_{ij}^2 . To achieve $\langle a|c \rangle = \langle b|d \rangle = 0$ for the four quantum states, one could use polarisation alone: setting their Bloch vectors to be at the poles and two antipodal points on the equator in the Bloch sphere would fulfil this distinguishability condition. However in order to *also* tune φ_{abcd} we must use two degrees of freedom. We control polarisation and temporal modes to prepare the following states (see Figure 4.4):

$$\begin{aligned} |a\rangle &= |H\rangle \otimes |t_1\rangle, \\ |b\rangle &= \frac{1}{\sqrt{2}} (|H\rangle + |V\rangle) \otimes |t_2\rangle, \\ |c\rangle &= |V\rangle \otimes |t_1\rangle, \\ |d\rangle &= \frac{1}{2} (|H\rangle + e^{i\theta} |V\rangle) \otimes |t_3\rangle. \end{aligned} \tag{4.2}$$

θ is adjusted using a waveplate to rotate the polarisation of $|d\rangle$ around the equator of the Bloch sphere. The temporal modes $|t_i\rangle$ are Gaussian wavepackets with fixed arrival times t_i , and the temporal duration of $|t_1\rangle$ is approximately twice that of $|t_2\rangle$ and $|t_3\rangle$. States $|a\rangle$ and $|c\rangle$ are orthogonal in polarisation and $|b\rangle$ and $|d\rangle$ are approximately distinguishable in temporal mode since $\langle t_2|t_3 \rangle \approx 0$ (this distinguishability is discussed in Section 4.4.1).

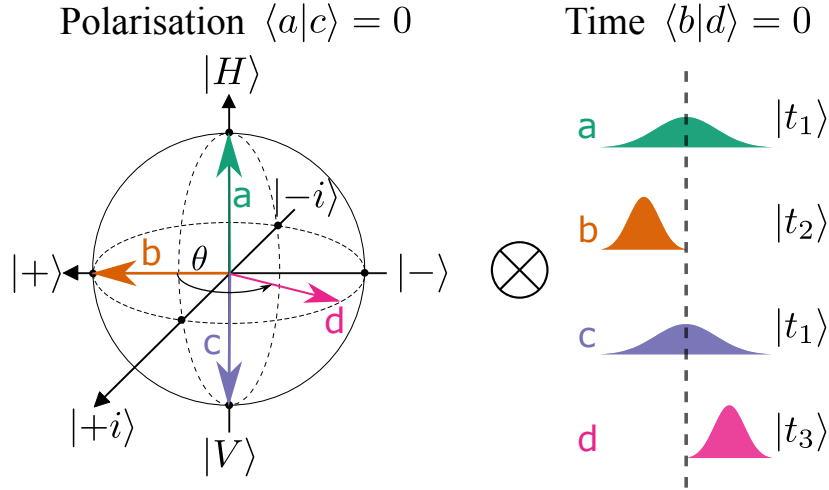


Figure 4.4: We use polarisation and temporal modes to ensure constant pairwise overlap magnitudes whilst simultaneously eliminating three-photon overlaps. The four-particle phase remains well-defined and is varied by rotating d 's polarisation in the equator of the Bloch sphere.

The non-zero overlaps are:

$$\begin{aligned} \langle a|b \rangle &= \frac{1}{\sqrt{2}} \langle t_1|t_2 \rangle, & \langle b|c \rangle &= \frac{1}{\sqrt{2}} \langle t_2|t_1 \rangle, \\ \langle c|d \rangle &= \frac{1}{\sqrt{2}} \langle t_1|t_3 \rangle e^{i\theta}, & \langle d|a \rangle &= \frac{1}{\sqrt{2}} \langle t_1|t_2 \rangle, \end{aligned} \tag{4.3}$$

corresponding to the weights of edges in Figure 4.3. All these overlaps have a constant absolute magnitude with θ and so two-photon interference terms do not vary. There is no three-photon interference due to the pairwise distinguishability removing any closed paths over three vertices. As we saw in Section 3.4, the arguments of Gaussian temporal mode overlaps cancel in cyclic products of overlaps, and here their moduli are $\sim 1/\sqrt{2}$ (see Section 4.4.1). Only the argument of the polarisation overlap in $\langle c|d \rangle$ affects the four-particle phase, so $\varphi_{abcd} = \theta$.

It is worth pointing out here that φ_{abcd} is tuned by varying the relative orientation of two states that have no mutual overlap. Moreover the state $|d\rangle$ that is rotated maintains a fixed orientation with respect to $|a\rangle$ and $|c\rangle$ at the poles, seemingly providing no reference for the angle of rotation θ . The remarkable fact that this geometric phase appears in interference arises from the ability of multiple particle to define a relative phase reference, here due to the mutual overlaps of the non-distinguishable states [261].

4.3 Four-mode interferometer

4.3.1 Building a quitter

The balanced four-mode interferometer is called a *quitter* and is described by the unitary matrix

$$U_{quit} = \frac{1}{2} \begin{pmatrix} 1 & 1 & 1 & 1 \\ 1 & 1 & -1 & -1 \\ 1 & -1 & e^{i\chi} & -e^{i\chi} \\ 1 & -1 & -e^{i\chi} & e^{i\chi} \end{pmatrix}. \quad (4.4)$$

All matrix element magnitudes are equal and so all input ports are equally coupled. There is a free internal phase χ that, unlike for the balanced 2×2 and 3×3 splitters, is not determined by the splitting ratio [240]. When $\chi = \pi/2$ this yields the 4×4 Fourier multiport. The quitter has been considered for entanglement generation [233, 262], Bell state analysis [263], optical homodyne [264, 265] and also multiphoton interference [45, 107, 143, 257, 266].

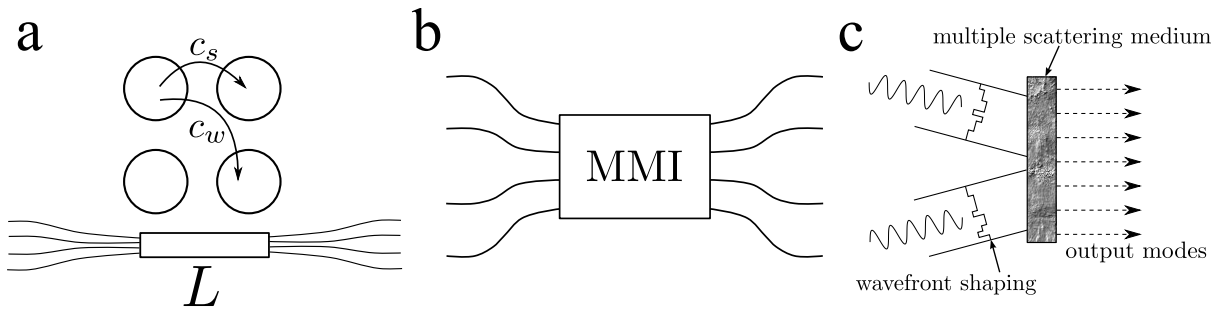


Figure 4.5: Some different realisations of a quitter. **a** Fusing four fibres with suitable choice of strong and weak coupling constants (c_s, c_w respectively) and interaction length L allows realisation of a quitter [79]. **b** Multimode interference (MMI) devices can achieve splitters that are robust to fabrication imperfections [267]. **c** Wavefront shaping of light using an SLM enables control over the large number of scattering processes in some opaque medium and thus adjustment of the coupling processes [268] (see main text for details).

There are a few ways of making this interferometer, with examples shown in Figure 4.5. The first comprises four fused fibres lying at the vertices of a square and with an interaction length

L . Due to the geometry of the fibres, those that are directly adjacent have a stronger coupling of their fields described by c_s , while those separated on the diagonal have a weaker coupling c_w . Surprisingly it is still possible to obtain a balanced splitting ratio by setting the interaction length $L = (2p + 1)\pi/4c_s, p \in \mathbb{Z}$, and this is independent of the weak coupling c_w [79]. The supported eigenmodes travel with different phase velocities so the internal phase is set by the ratio of the coupling strengths $\chi = \pi \times c_w/c_s$, though this can be difficult to engineer.

A second approach uses a multimode interference (MMI) device [266]. An MMI device supports many lateral modes that experience different refractive indices and so propagate at different speeds. The relative acquired phases result in constructive or destructive interference that, with suitable tuning of device length, achieve balanced splitting [267]. Unfortunately this property can be detrimental for observing quantum interference: it can introduce a timing jitter on the arrival times of the photons at different waveguides, thus introducing undesired temporal distinguishability. This may be countered by ensuring that the coherence length of the photons is long compared to any such timing jitter, meaning narrowband spectra. This is therefore unsuitable for the broadband modes we require for our distinguishable state preparation.

A third technique exploits the many scattering processes that occur within some opaque medium [268]. A spatial-light modulator (SLM) is used to shape the wavefront of the incoming beams so that, with appropriate phase shifts and knowledge of the transfer matrix of the scattering medium, some arbitrary scattering matrix can be programmed in. Whilst two-photon interference has been observed using this process, absorption in the medium is a significant source of loss and rates are low.

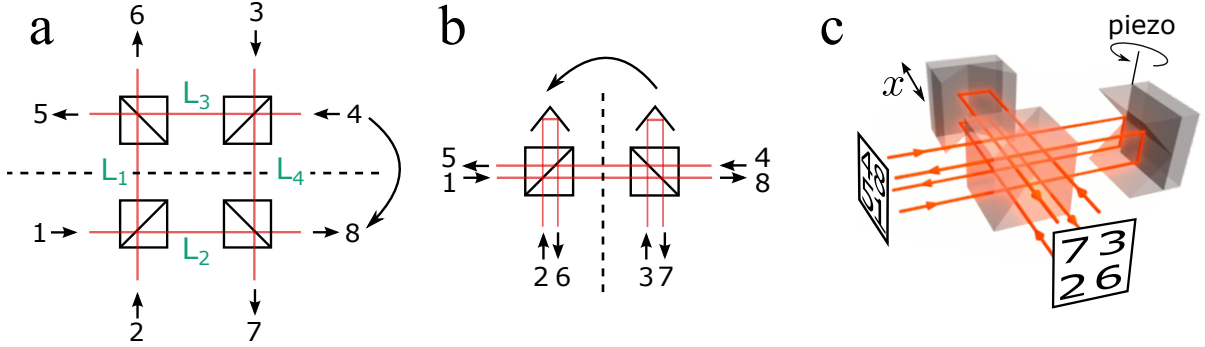


Figure 4.6: **a** Bulk optic realisation of a balanced four-port interferometer. All beam splitters have 50% reflectivity, and the numbers 1–4 label input ports and 5–8 label the outputs. Internal path lengths are labelled L_i . We fold the interferometer along the dashed line by using retroreflectors to double-pass through a pair of beam splitters at different lateral positions to give **b**. We then fold again using retroreflectors that translate the beams to a different vertical position, out of the page as shown. This results in the configuration in **c** where the beams pass through a single 50:50 beam splitter multiple times, simplifying alignment and improving stability. The corresponding input and output ports are labelled on the free faces of the beam splitter. The internal path lengths are matched using a delay stage on the lateral retroreflector (labelled x) and the vertical retroreflector is mounted on a piezo that provides control over the internal phase (see main text for details).

Our quitter is built using bulk optic components: four balanced beam splitters suitably arranged achieve equal coupling of four inputs and four outputs [45, 263] (Figure 4.6a). In order to simplify alignment, path length matching, and improve stability of the interferometer, a novel approach was taken: fold the circuit twice as shown in Figure 4.6b,c. A single 1" broadband

dielectric beam splitter (Thorlabs BS014) was passed multiple times using two retroreflecting mirrors (Thorlabs HRS1015-P01), allowing four spatial modes incident on each free face of the beam splitter to be coupled. Four input SMFs were aligned with the spatial modes labelled 1–4, and another four were aligned to the outputs labelled 5–8. The total fibre-to-fibre transmission of the device was $\sim 65\%$ and most of this loss occurs on reflection at the beam splitter.

The path lengths L_i must be matched to within the coherence length of the photons, so $L_1 \approx L_3$ and $L_2 \approx L_4$. The lateral retroreflector is mounted on a motorised delay stage and translating its position x as shown in Figure 4.6 allows $L_1 \rightarrow L_1 + 2x$ and $L_4 \rightarrow L_4 + 2x$. In order to match the paths, we send indistinguishable photons into inputs 1 and 3 and monitor χ -independent HOM dips on the pairs of outputs 5&6 and 7&8. If the path lengths are not matched, the bottoms of the dips will not line up. The position x is adjusted until this is achieved and the presence of χ -dependent HOM dips verifies correct operation (see Section 4.3.2).

The internal phase χ is determined by path length mismatch at the wavelength scale:

$$\chi = \frac{2\pi}{\lambda} \times [(L_1 + L_4) - (L_2 + L_3)]. \quad (4.5)$$

For this fine control we affix the vertical retroreflector to a computer-controlled piezo-driven mirror mount (Newport New Focus 8821). A slight tilt of the mount changes the distance $L_2 + L_3$ by an amount ϵ with minimal beam steering, so coupling into fibres outside the interferometer is unaffected. A single step of the piezo corresponds to $\epsilon \approx \lambda/20$ for 830nm light (see Figure 4.8).

4.3.2 Interference statistics for a quitter

First consider injecting two photons with internal states labelled a, b into input ports i, j and monitoring outputs k, l for coincidences. If $i = 1, j = 2$ and $k = 6, l = 7$ then, reading off the relevant submatrix from U_{quit} in equation 4.4, we see the scattering matrix for a balanced beam splitter, up to a factor of $1/\sqrt{2}$. This is also clear from Figure 4.6a: this combination of inputs and outputs just samples interference on a single beam splitter. We therefore obtain the probability

$$P_{12}^{67} = \frac{1}{8} (1 - r_{ab}^2). \quad (4.6)$$

Indistinguishable states result in suppression of this output coincidence and a HOM dip can be measured. If instead we monitor outputs $k = 5, l = 6$ then we sample a matrix whose elements are all $1/2$. Without the π phase shift, the destructive interference disappears so

$$P_{12}^{56} = \frac{1}{8} (1 + r_{ab}^2). \quad (4.7)$$

Here indistinguishable states result in an increase in coincidences and observation of a HOM ‘peak’. Again Figure 4.6a reveals what is going on: this coincidence combination is essentially performing number resolution of the two-photon bunching that occurs on the beam splitter with inputs 1 and 2, and this is expected to have exactly the opposite shape of the HOM dip in coincidences.

Things get more interesting if we inject the pair into inputs $i = 2, j = 3$ and monitor

coincidence on outputs $k = 5, l = 7$ and $k = 5, l = 8$. The coincidence probabilities now read

$$P_{23}^{57} = \frac{1}{8} (1 - r_{ab}^2 \cos \chi), \quad P_{23}^{58} = \frac{1}{8} (1 + r_{ab}^2 \cos \chi). \quad (4.8)$$

So the interference of indistinguishable states is mediated by χ , allowing observation of a HOM dip, peak, or elimination of two-photon interference entirely if $\chi = \pi/2$. Furthermore this phase cannot be measured using single photons alone: given some input port there is only one path to a given output port, so no single particle interference like in the double-slit experiment is possible. On the other hand, the two interfering amplitudes for coincidences sample all internal paths, and the sensitivity to any length mismatch manifests as a dependence on the internal phase χ . This emphasises that two-photon interference does not reduce to combinations of single-particle interference [257]. The scattering matrix here is effectively that of a lossy beam splitter, where the loss modes enable tuning of the phase that would otherwise be π by energy conservation [269, 270]. The phase-dependence of such dips has previously been demonstrated [45, 266], and we confirm it experimentally by sending a pair of photons from a single SPDC source with pairwise distinguishability $r_{ab}^2 \sim 0.9$ into the quitter (see Figure 4.7).

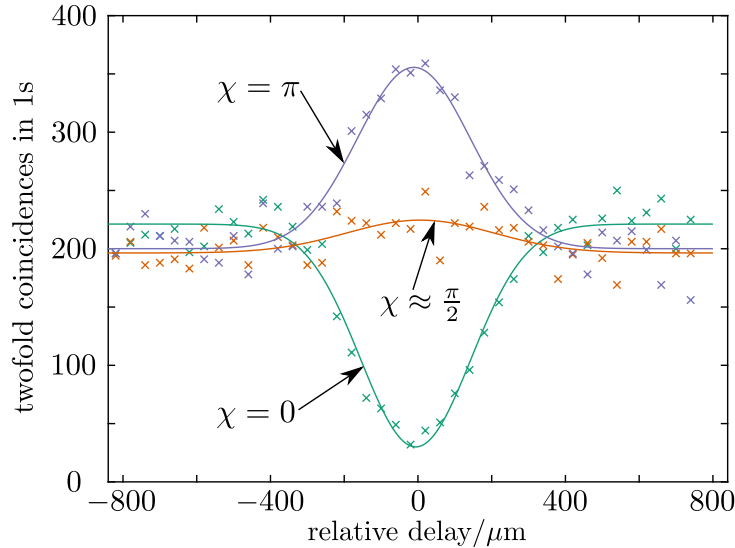


Figure 4.7: Two-photon interference in a quitter that shows the dependence of P_{23}^{57} on the internal phase χ (equation 4.8).

Now we turn to the coincidence statistics obtained when injecting four photons into the inputs of the quitter. As we have just seen, the order of injection is important when using this device. In order to observe a fourfold coincidence signal with strong dependence on the four-particle phase φ_{abcd} , we inject the pairwise distinguishable states of equation 4.2 in the order:

$$\begin{aligned} |a\rangle &\rightarrow \text{input 4,} \\ |b\rangle &\rightarrow \text{input 2,} \\ |c\rangle &\rightarrow \text{input 3,} \\ |d\rangle &\rightarrow \text{input 1.} \end{aligned} \quad (4.9)$$

Remembering $\langle a|c\rangle = \langle b|d\rangle = 0$, equation 2.54 gives the fourfold output coincidence probability

$$P_{1111} = \frac{1}{32} [3 - r_{ab}^2 - r_{bc}^2 - r_{cd}^2 - r_{ad}^2 + (\cos 2\chi + 2)(r_{ab}^2 r_{cd}^2 + r_{ad}^2 r_{bc}^2) + 2(\cos 2\chi - 2)r_{ab}r_{bc}r_{cd}r_{ad} \cos \varphi_{abcd}]. \quad (4.10)$$

It is straightforward to read off the HOM-type contributions in the pairwise distinguishabilities r_{ij}^2 , corresponding to elements of S_4 that comprise single transpositions. There are no triad phases because our state preparation eliminates the effect of threefold exchanges. The terms given by products of two pairwise distinguishabilities correspond to paths where two pairs exchange. In the graph picture these are the coverings using two closed loops over two pairs of vertices, and are due to the S_4 elements whose cycle structure is two transpositions.

The last term is the one we are after: the four-photon exchange with a dependence on φ_{abcd} . In order to maximise the contrast of this term as the phase varies from 0 to π , we want the prefactor $(\cos 2\chi - 2)$ to have the largest magnitude possible so $\chi = \pi/2$. This also minimises the flat contribution from the terms given by products of pairwise distinguishabilities $(\cos 2\chi + 2)$. This value actually eliminates the two-photon interference of equation 4.8: yet another example of how lower-order interferences do not necessarily describe higher-order effects.

4.3.3 Ensuring polarisation independence

The quitter was found to have a polarisation dependent loss: if single photons are prepared in H before the input fibre, polarisation compensation using waveplates and paddles does not guarantee H arrives at the coupling region (see Appendix A). If the input is rotated to V , the singles counts on an APD at the output were found to drop by up to 10%. We attribute this to the dielectric beam splitter having some orthogonal axes that have different transmissions: linear dichroism [271]. Hence as we rotate the polarisation state before the interferometer fibres, the intensities of H and V arriving *at the coupler* change. This means that as we rotate the state $|d\rangle = \frac{1}{\sqrt{2}}(|H\rangle + e^{i\theta}|V\rangle)$, the singles counts would fluctuate with θ , potentially swamping the variation in fourfold coincidences we are after. Hence here we had to use a different technique for polarisation compensation that involved matching the axes of polarisation to the loss axes in the beam splitter, meaning that the intensities at the beam splitter do not vary as θ is varied (see Appendix A.2). This approach achieved $< 1\%$ singles fluctuations as $|d\rangle$ was rotated.

In order to test the control over χ afforded by the piezo-driven retroreflector, we prepared two photons from one source in horizontal polarisation and injected them into inputs $i = 2, j = 3$ of the quitter. The delay stage was set at the bottom of the phase-independent HOM dip to ensure temporal indistinguishability. At the same time we also prepared two photons in vertical polarisation from the other source and injected them into inputs $i = 1, j = 4$, also setting that delay stage for temporal indistinguishability. The piezo was shifted one step at a time and the phase-dependent coincidences at outputs $k = 5, l = 7$ (equation 4.8) were monitored, with each source being opened in turn to record coincidences at the same piezo setting. These HOM probabilities should have the same dependence on χ . Results are shown in Figure 4.8a.

In both cases we observe the expected cosine shape with visibility of around 70%, consistent with the prediction from equation 4.8 and $r_{ij}^2 \approx 0.55$. However the internal phase seen by horizontally polarised light is exactly $\pi/2$ out of phase with that experienced by vertically

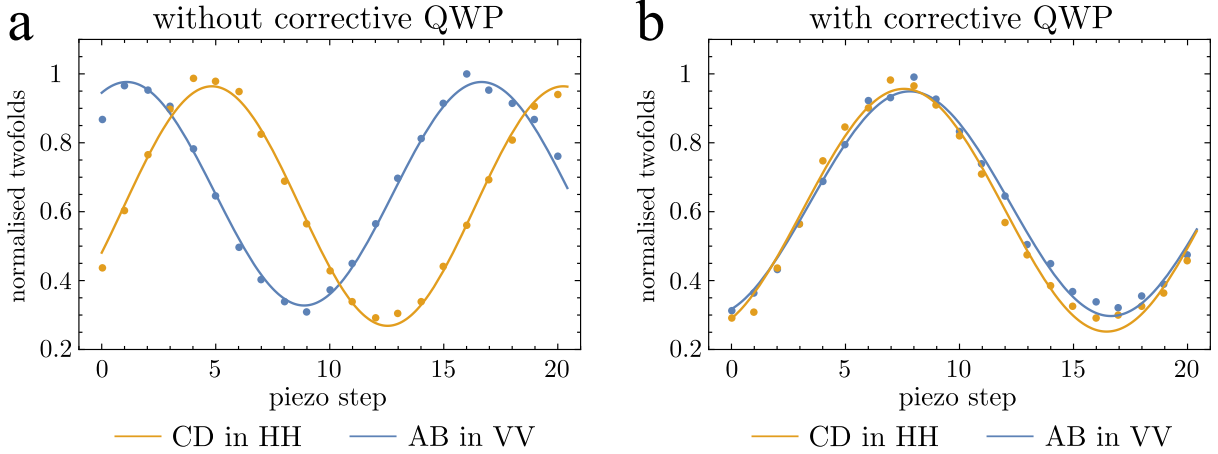


Figure 4.8: Phase-dependence of coincidences in the quitter. We use each SPDC source to prepare pairs of photons with $r_{ij}^2 \approx 0.55$ but for one source (CD) they are both horizontally polarised, and for the other (AB) they are vertically polarised. Sitting in the bottom of the HOM dips, we then shift the piezo one step at a time and open each source in turn to monitor the phase-dependent coincidences. **a** Here the coincidences reveal that horizontal and vertical polarisations experience phases offset by exactly $\pi/2$. **b** Addition of a quarter-wave plate oriented on axis inside the interferometer corrects for this shift and ensures the same phase χ is experienced independent of polarisation (see main text for details).

polarised light. We suspect this is a consequence of the Fresnel relations governing reflection and transmission for H and V polarisations at some interface [272]. We have to correct this otherwise we cannot perform a partial exchange symmetry test on a multiparticle wavefunction because the internal state is coupled to the transformation on the port modes. Additionally simulations confirmed that the visibility of the coincidence probability in equation 4.10 would be significantly reduced.

A quarter-wave plate works by introducing a $\pi/2$ phase shift between horizontal and vertical polarisations, so this can be used to exactly undo the undesired phase shift. We carefully inserted an 830nm QWP (unmounted Thorlabs WPQ05M-830), oriented at its fast-axis, on the path labelled L_2 in Figure 4.6a. With suitable adjustments of the internal path lengths to compensate for the optical path difference introduced by the quartz wave plate, the HOM dips were once again aligned and coincidences were monitored whilst stepping the piezo as before. Results are shown in Figure 4.8b, and it is clear that both polarisations now experience the same phase χ so the matrix describing the quitter is independent of polarisation.

4.3.4 Locking the quitter phase

The coincidence signal visibility of equation 4.10 is maximised when $\chi = \pi/2$ but, since this is a bulk interferometer, the phase is susceptible to variations in laboratory conditions. For example the optical table slightly expands or contracts with temperature, and humidity variations can change air's refractive index, resulting in changes of the path length within the interferometer. In the previous section we saw that the internal phase can be determined by monitoring coincidence counts and it can be changed using the piezo-mounted retroreflector. We therefore implement a locking procedure that shifts the piezo to maintain coincidences at the centre of the fringes in counts, corresponding to keeping $\chi \approx \pm\pi/2$ (see Figure 4.9).

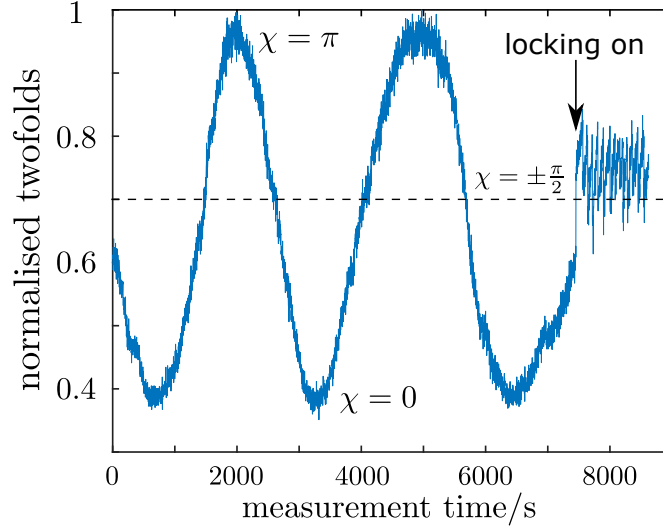


Figure 4.9: Monitoring the coincidence probability P_{23}^{57} of equation 4.8 permits measurement of the internal phase χ . Drifts in lab environment parameters with the air conditioning cycle cause slow, periodic variations of this phase. We can lock to the centre of the fringe by adjusting the piezo-driven retroreflector as shown in Figure 4.6c.

During our experiment we periodically use a single SPDC source to measure χ and then shift the piezo to achieve the desired value. Instead of constantly locking to a very narrow tolerance band, we lock to the central 1/3 of the fringe variation which, as we shall show in Section 4.4.1, only slightly reduces the visibility of the fourfold coincidence signal, whilst ensuring more time is spent recording fourfolds for significant statistics.

4.4 Generating four photons

4.4.1 Gaussian wavepackets from SPDC sources

The SPDC sources based on bulk KDP crystals are well-suited to this experiment: they generate pairs of photons with the same central wavelength but quite different bandwidths (see Figure 3.6), similar to what is shown for the distinguishable state preparation in Figure 4.4. We now consider what the ideal bandwidth ratio of the signal and idler photons should be for this experiment.

We earlier assumed the distinguishability $\langle a|c \rangle = 0$ is achieved using orthogonal polarisations and that $\langle b|d \rangle = 0$ using temporal modes. The latter would require temporal modes with top-hat profiles as shown in Figure 4.10a (assuming flat spectral phase). For this ideal preparation all the non-zero pairwise distinguishabilities $r_{ij}^2 = 1/4$ and the coincidence signal would have 30% visibility for $\chi = \pi/2$. Since the SPDC sources generate photons with Gaussian spectra, their wavepackets are also Gaussian. Distinguishability of the temporal modes $|t_2\rangle$ and $|t_3\rangle$ would therefore require a large relative time delay, which would also decrease the magnitude of the other temporal overlaps $|\langle t_1|t_2 \rangle|, |\langle t_1|t_3 \rangle|$. However from the expression for the fourfold coincidence probability in equation 4.10, the four-photon exchange contribution is given by $\sim r_{ab}r_{bc}r_{cd}r_{ad} \cos \varphi_{abcd}$ so the prefactor should be large to maximise the variations with φ_{abcd} . In order to balance this trade-off we therefore say that two Gaussian wavepackets

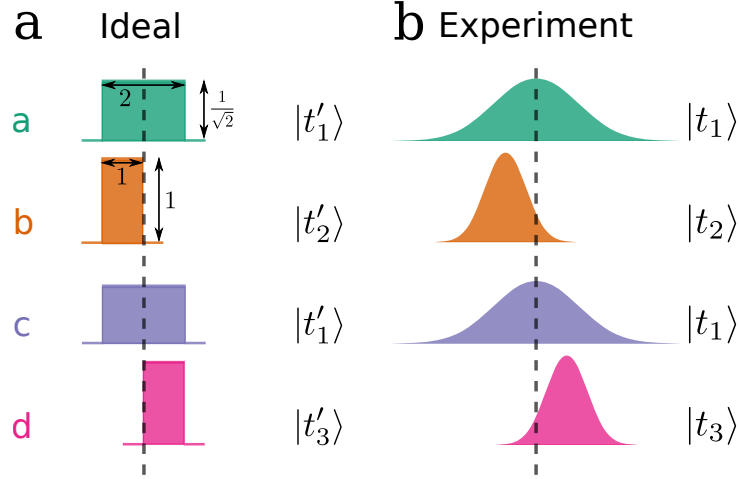


Figure 4.10: Ideal and experimentally feasible temporal mode configurations. **a** Top-hat functions with indicated relative dimensions would be ideal to maximise $|\langle t'_1 | t'_2 \rangle|$ and $|\langle t'_1 | t'_3 \rangle|$, giving the largest possible prefactor for $\cos \varphi_{abcd}$ variations, whilst also enforcing $\langle t'_2 | t'_3 \rangle = 0$. **b** The SPDC sources generate photons with Gaussian spectral, and therefore temporal, profiles. The temporal modes in this experiment have a temporal duration ratio of $\sim 2.2 : 1$, but do not achieve perfect distinguishability (see main text).

are “distinguishable” if the HOM visibility on a balanced beam splitter is at most 1%, or equivalently $|\langle t_2 | t_3 \rangle| \leq 0.1$. Subject to this constraint, it turns out that the visibility of the fourfold coincidence signal of equation 4.10 is maximised when the ratio of the temporally long to temporally short wavepackets is $\sim 2.2 : 1$ (see Figure 4.10b). This also means that the broadband idler mode still has reasonable count rates after filtering.

Due to imperfect distinguishability of temporal modes, there will be a small θ -dependent contributions to the fourfold coincidence probability from three-photon exchanges. It is important to estimate their contribution relative to the desired four-photon exchanges. We use equation 2.54 to explicitly write out the various exchange contributions to the quitter coincidence probability for the input states of equation 4.2 when using these Gaussian wavepackets (see Table 4.1).

Table 4.1: Exchange contributions to the four-photon coincidence probability

Cycle ρ	Permanent	State dependence	Cycle ρ	Permanent	State dependence
$\mathbb{I}$	$\frac{3}{32}$	1	(1,4,3)	$\frac{1}{16}$	$r_{ad}\mathbf{r}_{ac}r_{cd}e^{i\theta}$
(1,2)	$-\frac{1}{32}$	$\mathbf{r}_{bd}^2$	(2,3,4)	$\frac{1}{16}$	$r_{ab}r_{bc}\mathbf{r}_{ac}$
(1,3)	$-\frac{1}{32}$	r_{cd}^2	(2,4,3)	$\frac{1}{16}$	$r_{ab}\mathbf{r}_{ac}r_{bc}$
(1,4)	$-\frac{1}{32}$	r_{ad}^2	(1,2)(3,4)	$\frac{3}{32}$	$\mathbf{r}_{ac}^2\mathbf{r}_{bd}^2$
(2,3)	$-\frac{1}{32}$	r_{bc}^2	(1,3)(2,4)	$\frac{1}{32}(\cos 2\chi + 2)$	$r_{ab}^2r_{cd}^2$
(2,4)	$-\frac{1}{32}$	r_{ab}^2	(1,4)(2,3)	$\frac{1}{32}(\cos 2\chi + 2)$	$r_{ad}^2r_{bc}^2$
(3,4)	$-\frac{1}{32}$	$\mathbf{r}_{ac}^2$	(1,2,3,4)	$-\frac{1}{32}$	$\mathbf{r}_{ac}r_{bc}\mathbf{r}_{bd}r_{ad}$
(1,2,3)	$\frac{1}{32}$	$r_{bc}r_{cd}\mathbf{r}_{bd}e^{i\theta}$	(1,2,4,3)	$-\frac{1}{32}$	$r_{ab}\mathbf{r}_{ac}r_{cd}\mathbf{r}_{bd}e^{i\theta}$
(1,2,4)	$\frac{1}{16}$	$r_{ab}\mathbf{r}_{bd}r_{ad}$	(1,3,2,4)	$\frac{1}{32}(\cos 2\chi - 2)$	$r_{ab}r_{bc}r_{cd}r_{ad}e^{-i\theta}$
(1,3,2)	$\frac{1}{32}$	$r_{cd}r_{bc}\mathbf{r}_{bd}e^{-i\theta}$	(1,3,4,2)	$-\frac{1}{32}$	$r_{ab}\mathbf{r}_{ac}\mathbf{r}_{bd}r_{cd}e^{-i\theta}$
(1,3,4)	$\frac{1}{16}$	$\mathbf{r}_{ac}r_{cd}r_{ad}e^{-i\theta}$	(1,4,2,3)	$\frac{1}{32}(\cos 2\chi - 2)$	$r_{ab}r_{bc}r_{cd}r_{ad}e^{i\theta}$
(1,4,2)	$\frac{1}{16}$	$r_{ad}r_{ab}\mathbf{r}_{bd}$	(1,4,3,2)	$-\frac{1}{32}$	$\mathbf{r}_{ac}r_{bc}\mathbf{r}_{bd}r_{ad}$

The emboldened overlap magnitudes $\mathbf{r}_{ij}$ are those we earlier assumed were zero in order to eliminate three-photon exchange contributions. If we were to ignore these terms and sum the products of all matrix permanents with their corresponding distinguishabilities, the overall coincidence probability of equation 4.10 would be recovered. Now we here assume $r_{ac} = 0$ because orthogonality of polarisations is achievable. Preserving terms in r_{bd} that are no longer zero because $|\langle t_2|t_3 \rangle| \neq 0$, we sum the θ -dependent three- and four-photon exchanges to give respectively:

$$P^{(3)} = \frac{1}{16} r_{bc} r_{bd} r_{cd} \cos \theta, \quad P^{(4)} = \frac{1}{16} \times (2 - \cos 2\chi) \times r_{ab} r_{bc} r_{cd} r_{ad} \cos \theta. \quad (4.11)$$

$P^{(3)}$ has a $\cos \theta$ shape and $P^{(4)}$ goes as $-\cos \theta$, so the effect of three-photon exchanges is to *decrease* the signal visibility. Assuming $|\langle t_2|t_3 \rangle| = 0.1$ means $r_{bd} = 0.1 \times \sqrt{\frac{1}{2}(1 + \cos \theta)}$ and also taking $|\langle t_1|t_2 \rangle| = |\langle t_1|t_3 \rangle| = \frac{1}{\sqrt{2}}$ (a good approximation for the Gaussians in Figure 4.10), the ratio of these is:

$$P^{(4:3)} = \frac{|P^{(4)}|}{P^{(3)}} = \frac{5}{2} \times \frac{|\cos 2\chi - 2|}{\sqrt{\frac{1}{2}(1 + \cos \theta)}}. \quad (4.12)$$

The three-photon contribution is greatest when $\theta = 0$ and for this value we plot the ratio of exchange contributions with χ in Figure 4.11a. For the ideal quitter phase $\chi = \pi/2$, the three-photon term is over seven times smaller than the desired four-photon contribution. In Figure 4.11b we plot the visibility of the coincidence probability P_{1111} against χ . P_{1111} reaches a maximum when $\theta = \pi$, and a minimum when $\theta = 0, 2\pi$. As expected the signal contrast is highest when $\chi = \pi/2$. However as mentioned earlier, we want to maximise the time spent counting fourfolds and minimise as much as possible the time spent locking the phase using twofold coincidences. If we relax the tolerance of the locking algorithm so it maintains χ to be within the central third of the possible values shown in Figure 4.9, we impose $\arccos(1/3) = 1.23 \leq \chi \leq \pi - \arccos(1/3) = 1.91$ (dashed lines in Figure 4.11). This averaging reduces the visibility of the P_{1111} signal from 27.3% to 26.5%, and the maximum relative contribution of the three-photon exchange is ~ 7.4 times smaller than the four-photon term.

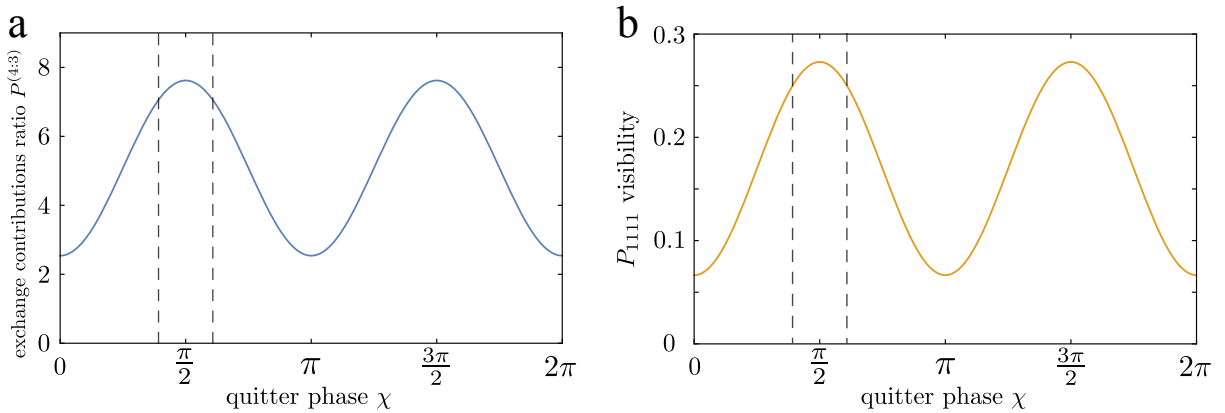


Figure 4.11: **a** The ratio of four- and three-photon exchange contributions to the fourfold coincidence probability when $\theta = 0$. On averaging over the range of χ in our experiment (shown as dashed lines), it is over seven times smaller than the four-photon term. **b** The visibility of the fourfold coincidence signal including three-photon contributions. Averaging over the indicated χ range only slightly reduces the visibility from the ideal at $\chi = \pi/2$.

4.4.2 Induced coherence and double emissions

The use of multiple SPDC sources in an unheralded configuration and pumped by the same laser opens the door to some fascinating demonstrations of the link between indistinguishability and coherence. Before addressing their role in our setup, we will first briefly consider the famously ‘mind-boggling’ experiment by Zou *et al.* [214] depicted in Figure 4.12. Two SPDC sources are pumped by the same continuous-wave laser so that the probability of pair generation is low and there is no timing information available for emissions.

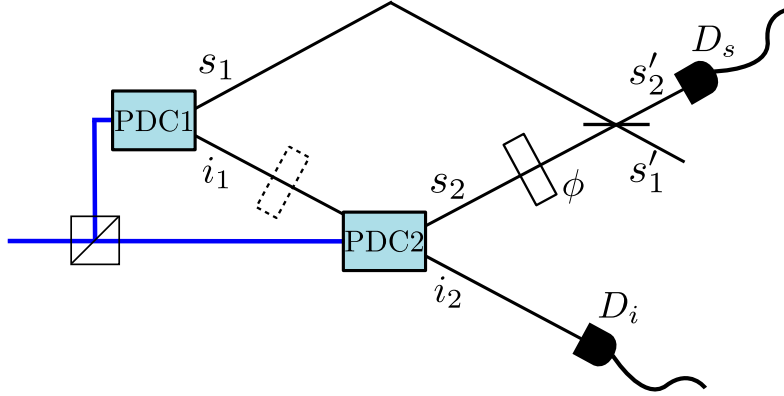


Figure 4.12: A continuous-wave laser pumps two SPDC sources whose idler modes are overlapped to be indistinguishable. Singles counts at detector D_s exhibit fringes as an applied phase ϕ is varied. If a beam block (dashed line) is inserted or the idler modes are made distinguishable, interference of the signal photons disappears (see main text). Adapted from [214].

The generated state in the signal s_1, s_2 and idler i_1, i_2 modes is

$$|\psi_{in}\rangle = \frac{1}{\sqrt{2}} (|1_{s_1}, 1_{i_1}, 0_{s_2}, 0_{i_2}\rangle + |0_{s_1}, 0_{i_1}, 1_{s_2}, 1_{i_2}\rangle). \quad (4.13)$$

Next a phase ϕ is applied to s_2 and then a balanced beam splitter mixes the signal modes. Labelling the signal modes after the beam splitter by s'_1, s'_2 , the resulting state is

$$|\psi_{out}\rangle = \frac{1}{2} \left(|1_{s'_1}, 1_{i_1}, 0_{s'_2}, 0_{i_2}\rangle + |0_{s'_1}, 1_{i_1}, 1_{s'_2}, 0_{i_2}\rangle + e^{i\phi} \left(|1_{s'_1}, 0_{i_1}, 0_{s'_2}, 1_{i_2}\rangle - |0_{s'_1}, 0_{i_1}, 1_{s'_2}, 1_{i_2}\rangle \right) \right). \quad (4.14)$$

If we consider counting singles on s'_2 , then counts on detector D_s post-select the state

$$|\psi_{out}^{D_2}\rangle = \frac{1}{2} \left(|1_{i_1}, 0_{i_2}, 1_{s'_2}\rangle - e^{i\phi} |0_{i_1}, 1_{i_2}, 1_{s'_2}\rangle \right) \quad (4.15)$$

Assuming first that the idlers i_1, i_2 are distinguishable by, for example, spatial displacement or a beam block is placed on i_1 then, even if the idler emissions are not resolved by a detector D_i and nothing disturbs the signal modes, there is no interference at D_s . The probability of a detection $P_{out}^{D_s} = \langle \psi_{out}^{D_2} | \psi_{out}^{D_2} \rangle = 1/2$.

However if the idler modes are overlapped to be indistinguishable then a detection at D_i provides no information about which source fired. Setting $i_1 = i_2$ yields $P_{out}^{D_s} = (1 - \cos \phi)/2$, showing that the paths corresponding to an emission from PDC1 and from PDC2 interfere.

The fringes will be observed even if detector D_i is ignored. This is called *induced coherence* and arises due to the indistinguishability of the idler modes and the mutual coherence of the paths with the vacuum due to a common pump laser. As soon as which-path information is in principle available due to the idler photons then, even if it is not resolved, interference of the signal photons is destroyed [29]. This effect has been investigated and demonstrated for a variety of different geometries [85, 91, 92, 147, 213, 215, 273–275].

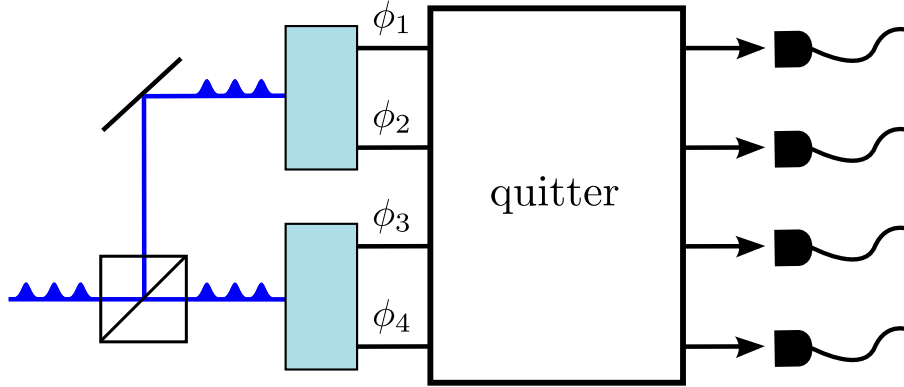


Figure 4.13: In our setup the emission from a pair of SPDC sources pumped by a pulsed laser is injected into a quitter. This interferometer erases information about which sources fired so two- and fourfold coincidences at the output ports exhibit dependence on the input phases ϕ_i .

A similar phenomenon can occur when using two SPDC sources and a quitter, with an example setup shown in Figure 4.13. The quitter erases spatial information about which source has fired and, if the internal states of the generated photons have some overlap between different sources, there will be interference effects arising due to induced coherences with the vacuum components of each TMSV. The total input state for two TMSV states with equal squeezing is

$$\begin{aligned} |\psi'_{in}\rangle &= |\psi_{TMSV}\rangle_{12} |\psi_{TMSV}\rangle_{34} \\ &= (1 - \lambda^2) \sum_{m,n=0}^{\infty} \lambda^{m+n} e^{i(m(\phi_1+\phi_2)+n(\phi_3+\phi_4))} |m, m, n, n\rangle, \end{aligned} \quad (4.16)$$

where ordering in the ket denotes input port, and we have included the phases ϕ_i on the inputs arising from path length differences before the interferometer. Restricting emission to the two-photon subspace, the input state is

$$|\psi_{in}^{(2)}\rangle = \frac{1}{\sqrt{2}} \left(e^{i(\phi_1+\phi_2)} |1, 1, 0, 0\rangle + e^{i(\phi_3+\phi_4)} |0, 0, 1, 1\rangle \right). \quad (4.17)$$

In Appendix F we show that for our chosen source and quitter configuration, and the pairwise distinguishable states labelled a, b, c, d , the twofolds coincidences at the quitter outputs can exhibit ϕ_i -dependent oscillations modulated by the quitter phase χ . At its heart the mechanism is the same as in the experiment by Zou *et al.*: coherence with the vacuum due to the indistinguishability of source emissions.

4.4.3 Subtracting statistics from higher-order emissions

When detecting four photons after the quitter it is impossible to know whether each source fired once, or one of the sources fired twice. In the four-photon subspace of the input state we have

$$|\psi_{in}^{(4)}\rangle = \frac{1}{\sqrt{3}} \left(e^{i(\phi_1+\phi_2+\phi_3+\phi_4)} |1, 1, 1, 1\rangle + e^{2i(\phi_1+\phi_2)} |2, 2, 0, 0\rangle + e^{2i(\phi_3+\phi_4)} |0, 0, 2, 2\rangle \right). \quad (4.18)$$

The density matrix in the four-photon emission space $\{|1, 1, 1, 1\rangle, |2, 2, 0, 0\rangle, |0, 0, 2, 2\rangle\}$ is then

$$\rho_{in}^{(4)} = \frac{1}{3} \begin{pmatrix} 1 & e^{-i\Delta} & e^{i\Delta} \\ e^{i\Delta} & 1 & e^{2i\Delta} \\ e^{-i\Delta} & e^{-2i\Delta} & 1 \end{pmatrix} \quad (4.19)$$

where we define $\Delta = \phi_1 + \phi_2 - \phi_3 - \phi_4$. The coincidence probability P_{1111} will generally depend on Δ in a similar way to the twofolds (see Appendix F). If these phases vary quickly and are in no way correlated with the measurement performed then the coherences will average to zero. To guarantee they do not affect interference in our experiments, we use a technique by Carolan *et al.* [136] to remove these coherences. A Pancharatnam phase is applied to the first mode by inserting a sequence of quarter-, half- and quarter-wave plates oriented at $\frac{\pi}{4}, \left(\frac{\pi}{4} - \frac{\xi}{4}\right), \frac{\pi}{4}$ respectively (precisely the Pancharatnam excursion in the Poincaré sphere shown in Figure 3.1). This leaves the state on that mode unchanged besides the addition of a phase factor $e^{i\xi}$, so Δ changes to $\Delta + \xi$. Taking an equal mixture of $\rho_{in}^{(4)}$ with $\xi = 0, \pi/2, \pi, 3\pi/2$ gives the state

$$\rho_{in,av}^{(4)} = \frac{1}{3} (|1, 1, 1, 1\rangle \langle 1, 1, 1, 1| + |2, 2, 0, 0\rangle \langle 2, 2, 0, 0| + |0, 0, 2, 2\rangle \langle 0, 0, 2, 2|). \quad (4.20)$$

The effect of the equally spaced phases ξ is to cancel out the coherences $e^{\pm i\Delta}, e^{\pm 2i\Delta}$. It is possible to measure the statistics for the same source firing twice independently by blocking each source in turn. Subtracting these from the statistics acquired with both sources open allows isolation of those corresponding to each source firing once. We use this technique and will now describe it in detail for our particular setup.

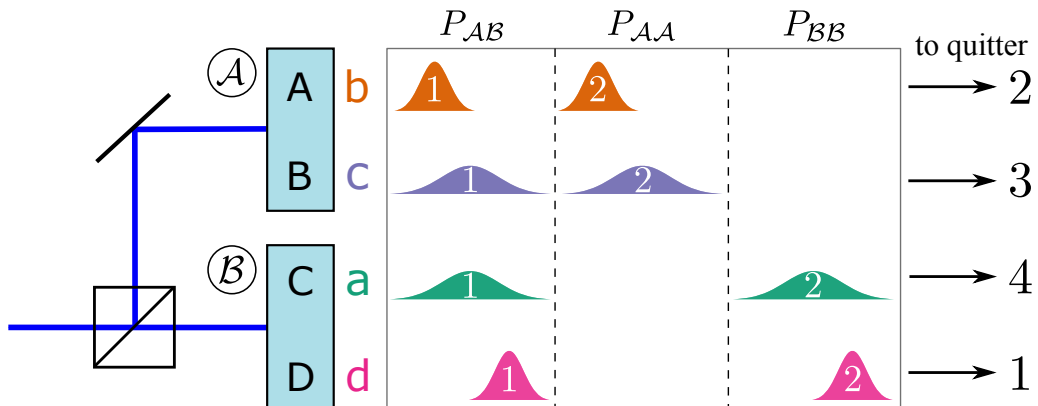


Figure 4.14: Configuration of two SPDC sources (labelled $\mathcal{A}$ and $\mathcal{B}$) to generate the pairwise distinguishable states labelled a, b, c, d from equation 4.2. The source arms A–D correspond to those of Figure 3.5 and the corresponding inputs to the quitter are shown. The inset shows the possible emissions of four photons from the sources, with the number of photons in each state labelled and P_{ij} as the probability of a particular emission event.

Our state preparation and order of injection to the quitter are shown in Figure 4.14. On averaging data at four different Pancharatnam phases, the effective input state is

$$\rho_{in}^{eff} = \mathcal{N} (P_{AB} |\Psi_{AB}\rangle \langle \Psi_{AB}| + P_{AA} |\Psi_{AA}\rangle \langle \Psi_{AA}| + P_{BB} |\Psi_{BB}\rangle \langle \Psi_{BB}|) \quad (4.21)$$

where $\mathcal{N}$ is a normalisation factor and we have allowed for the firing probabilities P_{AB}, P_{AA}, P_{BB} to be different. The first term corresponds to each source firing once and means successful preparation of the desired input state $|\Psi_{AB}\rangle = |1_{|d}\rangle, 1_{|b}\rangle, 1_{|c}\rangle, 1_{|a}\rangle$, where ordering in the ket corresponds to quitter input port. The other two terms are cases of probabilistically preparing the states $|\Psi_{AA}\rangle = |0, 2_{|b}\rangle, 2_{|c}\rangle, 0\rangle$ and $|\Psi_{BB}\rangle = |2_{|d}\rangle, 0, 0, 2_{|a}\rangle$. The overall fourfold coincidence signal includes contributions from all three terms:

$$P_{1111}^{tot} = \mathcal{N} (P_{AB} \times P_{1111}^{AB} + P_{AA} \times P_{1111}^{AA} + P_{BB} \times P_{1111}^{BB}). \quad (4.22)$$

The probability $P_{1111}^{AB} = P_{1111}$ from equation 4.10 and contains the φ_{abcd} term we want. The coincidences due to single sources firing twice depend only on pairwise distinguishabilities r_{ij}^2 :

$$\begin{aligned} P_{1111}^{AA} &= \frac{1}{32} (3 - 4r_{bc}^2 + r_{bc}^4 (2 + \cos 2\chi)), \\ P_{1111}^{BB} &= \frac{1}{32} (3 - 4r_{ad}^2 + r_{ad}^4 (2 + \cos 2\chi)). \end{aligned} \quad (4.23)$$

On averaging over our chosen range of χ these terms contribute a flat background to P_{1111}^{tot} . The firing probabilities are determined by the sources' squeezing parameters (Section 3.2.6) and input losses. For balanced pumping power and no losses they would all be given by $(1 - \lambda^2)^2 \lambda^4$, and the effect of the flat background contributions to P_{1111}^{tot} would be to decrease the visibility of the signal we are after from 26.5% to 10%. Whilst the pumping powers for each source were equal, input losses were slightly imbalanced meaning $P_{BB} \approx 1.2 \times P_{AA}$. Adjusting the normalisation $\mathcal{N}$ and the emission probabilities, this results in a very small reduction to the visibility of P_{1111}^{tot} .

During the experiment we separately record the double emission fourfolds by blocking each source in turn using shutters. It is important to note that this changes the probability of one source not firing from $(1 - \lambda^2)$ to 1. Hence the measured backgrounds are

$$P_{AA}^{bg} = \frac{P_{AA}}{(1 - \lambda^2)} \times P_{1111}^{AA}, \quad P_{BB}^{bg} = \frac{P_{BB}}{(1 - \lambda^2)} \times P_{1111}^{BB}. \quad (4.24)$$

Correcting for this factor (which is close to unity for $\lambda = 0.16$), then subtracting the independently measured backgrounds from P_{1111}^{tot} leaves the desired P_{1111}^{AB} signal that is not affected by imbalance of firing probabilities: you effectively post-select on the emission event you want.

Six-photon emissions are also possible from this pair of sources with a probability around $\lambda^2 \approx 2.5\%$ of that for four-photon emission. The possible firings are AAA, BBB, AAB, ABB and they can all lead to quitter outputs that register as fourfold coincidences. The first two depend only on pairwise distinguishabilities and again average to a flat background for our chosen χ range. Furthermore these are compensated for by the background subtraction just described. The other two possible firings – where one source fires twice and the other once – cannot be measured separately but contain a constant term and a small $-\cos \varphi_{abcd}$ term. The constant

term dominates and so these sixfold terms actually decrease the fourfold signal visibility but by a small amount, around 1.5% on the background-subtracted signal. Emissions of eight or more photons are very rare and have a negligible effect on the fourfold coincidence signal.

4.5 Demonstration of distinguishable state interference

4.5.1 Preparing polarisation and temporal modes

The bandwidths of the degenerate signal and idler photons generated by the KDP SPDC sources are 2.5nm and 12.5nm respectively, meaning the temporal duration of the signal photon's wavepacket is five times that of the idler's. We saw in Section 4.4.1 that the ideal ratio is 2.2 : 1 and so we apply spectral filtering to the down-conversion modes (see Figure 4.15).

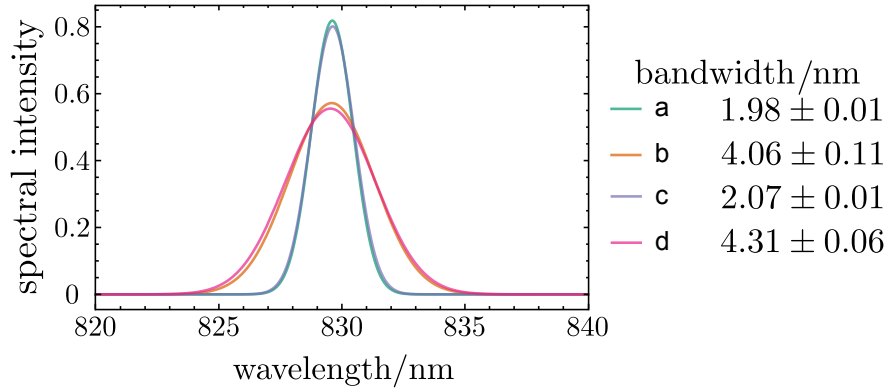


Figure 4.15: We insert 3nm filters (Semrock LL01-830-12.5) on the narrowband signal modes to achieve filtered bandwidths of 2nm for the states labelled *a*, *c*. For the broadband idler modes, a pair of bandpass filters are applied to trim each side of the spectrum (lower wavelength using Semrock FF01-857/30 and higher wavelength using Semrock FF01-832/37) and give ~ 4.2 nm bandwidths for the states labelled *b*, *d*.

As mentioned in Section 3.2.6, the width of a HOM dip between two Gaussian wavepackets of coherence lengths l_i is $l_{dip} = \sqrt{l_1^2 + l_2^2}$. It is important to verify that the filtering preserves the transform limit of the wavepackets from the KDP sources. Using the measured spectra in Figure 4.15 we predict the coherence lengths of the wavepackets and therefore also the width of the associated HOM dip, and these are presented in Table 4.2. We record HOM dips between the pairs of wavepackets shown and their full widths at half maximum are listed in the table. All measured dip widths (and also visibilities) are consistent with values predicted using the measured spectra and assuming transform-limited wavepackets.

Table 4.2: Coherence lengths and HOM dip widths

States	$l_1^{pred}/\mu\text{m}$	$l_2^{pred}/\mu\text{m}$	$l_{dip}^{pred}/\mu\text{m}$	$l_{dip}^{meas}/\mu\text{m}$
b,c	75 ± 2	147 ± 1	165 ± 1	172 ± 5
a,d	153 ± 1	70 ± 1	168 ± 1	165 ± 5
b,d	75 ± 2	70 ± 1	103 ± 2	92 ± 4
a,c	153 ± 1	147 ± 1	212 ± 1	215 ± 6

In Section 4.3.3 we discussed the procedures required to guarantee polarisation independence of the quitter. The crucial step is to check that when the state $|d\rangle = \frac{1}{\sqrt{2}}(|H\rangle + e^{i\theta}|V\rangle)$ is rotated in the equator of the Bloch sphere, the counts recorded at an output fibre that is preceded by a horizontally oriented polariser do not change. This determines the polarisation axes for the whole experiment and is used to calibrate the polarisation analysis at an output fibre to compensate all other modes in the Z basis. The positions of all delay stages corresponding to simultaneous arrival of the four photons at the quitter are found by measuring combinations of HOM dips between different arms of the SPDC sources. On rotation of the state $|c\rangle$ to vertical polarisation, the absence of a HOM dip with the horizontally polarised state $|a\rangle$ verifies the required distinguishability $\langle a|c\rangle = 0$. To prepare state $|b\rangle$ in $|+\rangle$, we first orient the state $|d\rangle$ somewhere in the equator of the Bloch sphere and calibrate the output polarisation analysis so this is the system's diagonal polarisation state. Now we send in $|b\rangle$ and manipulate its compensation waveplates to match this definition of diagonal. Since it is only relative orientation of these states in the equator that matters, this corresponds to $\varphi_{abcd} = 0$ and rotating θ varies the four-particle phase.

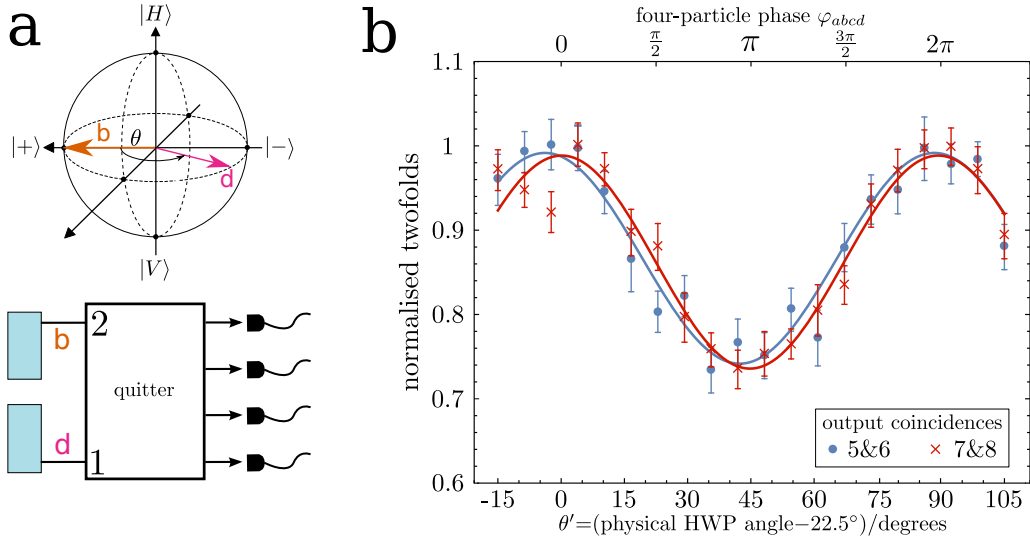


Figure 4.16: **a** In order to calibrate the waveplate angle to the four-particle phase φ_{abcd} , we temporally align the states labelled b and d and rotate the latter in the equator of the Bloch sphere whilst recording twofolds at the quitter outputs. **b** This set of output coincidences corresponds to sampling probabilities of equation 4.7 where indistinguishability is achieved when $4\theta' = \varphi_{abcd} = 0$ and the shape is $\sim \cos^2 2\theta'$. These thermal state (equation 3.24) signals are predicted to have 25% visibility. The shape of these sets of coincidences are used to calibrate the physical waveplate angle through $\theta' = \theta/4$ with the four-particle phase φ_{abcd} .

A quarter- and a half-wave plate are used to prepare the polarisation of $|d\rangle$

$$U_{QWP}\left(\frac{\pi}{4}\right) \cdot U_{HWP}\left(\theta' + \frac{\pi}{8}\right) |H\rangle = \frac{1}{\sqrt{2}} \left(|H\rangle + e^{4i\theta'} |V\rangle \right), \quad (4.25)$$

where we have inserted the physical angles of the waveplates in radians. Rotating the angle θ' from 0 to $\pi/2$ will change the angle in the Bloch sphere θ from 0 to 2π . In order to calibrate θ' to the corresponding φ_{abcd} , we temporally overlap the spectrally indistinguishable states $|b\rangle, |d\rangle$ and rotate the HWP whilst monitoring twofolds at the quitter outputs (see Figure 4.16).

These unheralded thermal state interference signals are used to associate the physical waveplate angle with the four-particle phase φ_{abcd} . If the state $|b\rangle$ is instead oriented to have horizontal polarisation then no variations are observed in the coincidences because the associated Bloch vectors remain at right angles with respect to each other.

To achieve the distinguishability of temporal modes so $\langle b|d\rangle \approx 0$, the states' polarisations are matched and the interference of thermal states is recorded as the relative delay is varied (see Figure 4.17). We ensure that $|\langle t_2|t_3\rangle| \leq 0.1$ by setting the delay stage to the indicated position. When walked off in this way and repeating the polarisation scan in Figure 4.16a, we observe no variations in output coincidences as expected.

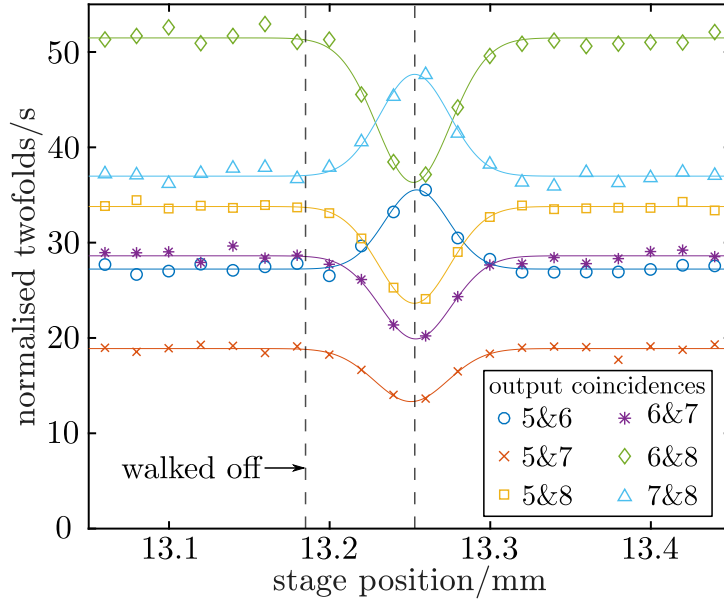


Figure 4.17: Two-photon interference between the temporally narrow independent thermal states (equation 3.24) when they have matched polarisations, giving dips with $\sim 33\%$ visibility and peaks with $\sim 25\%$ visibility. We set the delay stage to the position indicated by the dashed line on the left, for temporal overlap of 0.1 as mentioned in Section 4.4.1.

4.5.2 Results and discussion

The full experimental setup is shown in Figure 4.18. We repeatedly scan the polarisation state of $|d\rangle$ to vary φ_{abcd} across a range slightly larger than 2π , and at each setting of θ we record all singles and multiphoton coincidence counts at the quitter outputs when opening all different combinations of source arms (see Appendix G for additional data). This allows independent measurement of variations caused by lower order interference contributions and of the double emission background mentioned in 4.4.3. For every scan we use a motorised rotation mount to iterate between four different Pancharatnam phases applied to the fourth quitter input to remove the effects of induced coherence, and we carefully check there is no unintended rotation of the polarisation state injected to the quitter. The locking procedure described in Section 4.3.4 ensures that the quitter phase χ stays within a range that maximises the visibility of the desired signal. Data recorded with different source arms injected into the quitter are shown in Figure 4.19, allowing assessment of the state preparation and observation of distinguishable state interference.

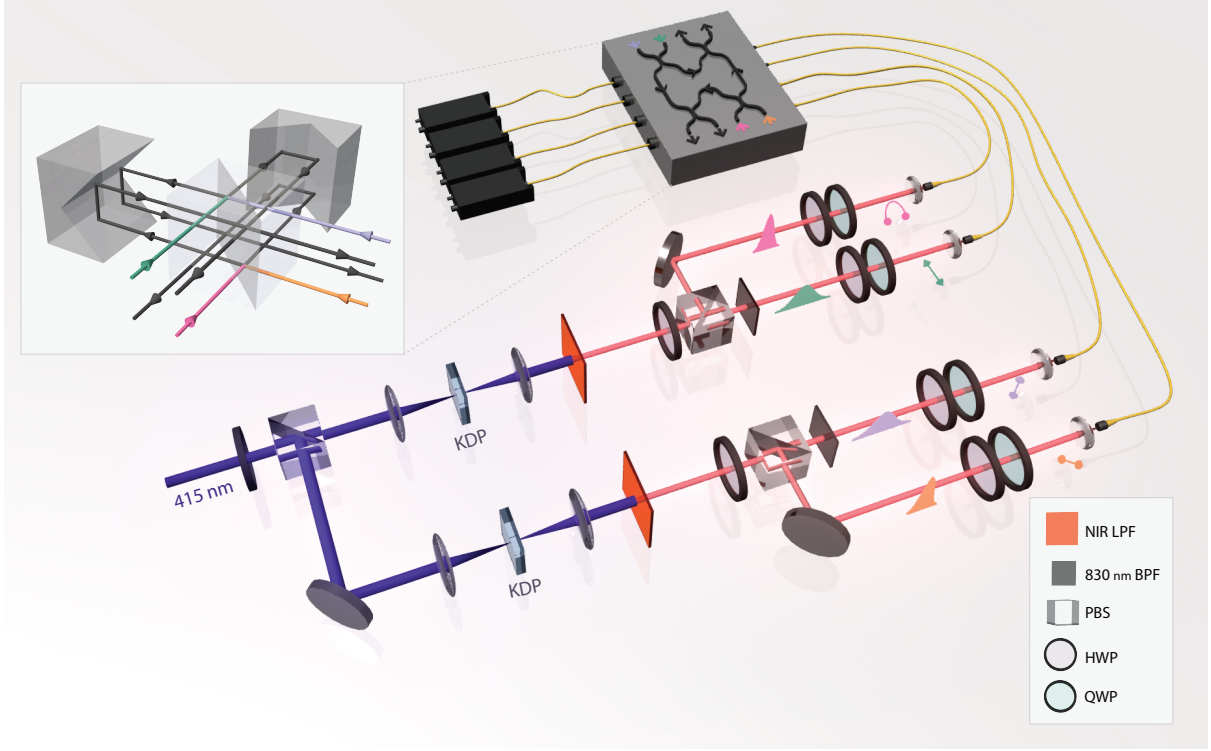


Figure 4.18: Experimental setup for distinguishable state interference. The pair of KDP sources probabilistically generate four photons that are separated from the pump using long-pass filters (NIR LPF), and spatially separated using PBSs. Band-pass filters (BPF) and waveplates are used to prepare the pairwise distinguishable states shown in Figure 4.4 before injection into the interferometer. The inset shows the free-space bulk optic quitter with inputs to the free faces of the beam splitter colour-coded to correspond to the prepared states. APDs detect photons at the quitter outputs and coincidences are recorded using a Swabian TimeTagger 20 with 4ns coincidence window. Arms of the sources are blocked during the experiment using motorised shutters. Additional waveplates for applying the Pancharatnam phase are not shown.

Figure 4.19a shows the mean singles measured at the quitter outputs when injecting $|d\rangle_1$ and blocking all other inputs. This state's polarisation is rotated around the equator of the Bloch sphere to change φ_{abcd} and it is the only preparation that varies during the experiment. Thanks to the efforts mentioned in Section 4.3.3 the effect of the quitter's polarisation-dependent loss has been minimised so these singles have a maximum variation of 1.4% and an average variation of $< 1\%$. Singles counts were also recorded when opening the other source arms individually, and are shown in Appendix G.1. Since the other states do not change during the run, the associated output singles are all constant.

In Figure 4.19b we plot the output twofolds when using a single source to inject $|d\rangle_1 |a\rangle_4$. The Bloch vector for the polarisation of $|a\rangle$ lies at right angles to that for $|d\rangle$ throughout variation of φ_{abcd} and so the pairwise distinguishability is ideally constant. The dependence of any twofold probabilities on χ averages to a constant given the locking employed. The twofold coincidences depending on this quantity are constant within error expect those on outputs 5 and 6, which have 1.6% variation due to the small variations in the underlying $|d\rangle$ singles counts. Normalising for these we see a variation of only 1.2% on this output channel. Similar data was recorded for all pairs of inputs to the quitter (see Appendix G.2) and these are constant as φ_{abcd} is varied, demonstrating constant pairwise distinguishabilities.

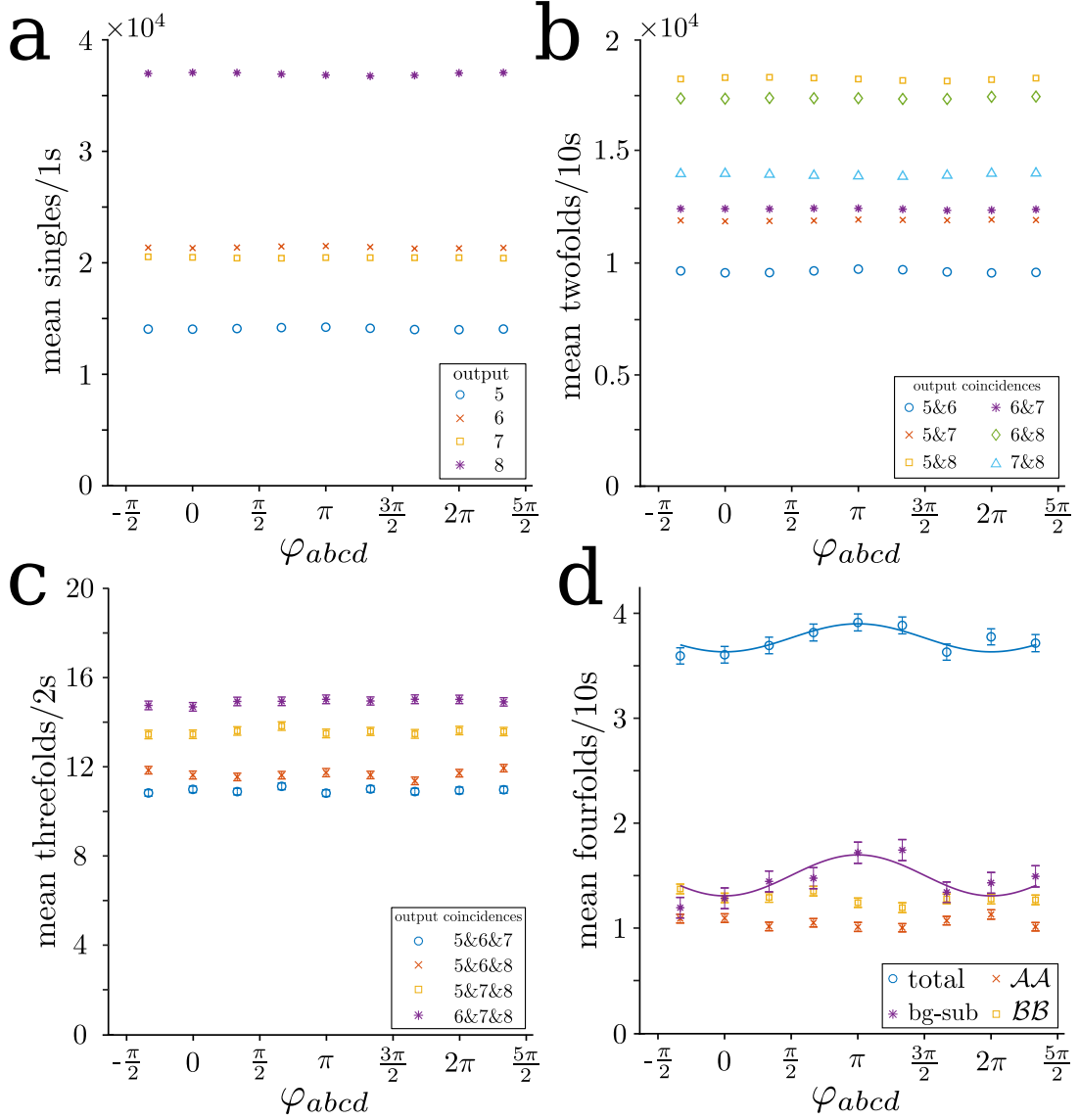


Figure 4.19: Counts measured at the quitter outputs for interference of pairwise distinguishable states when the quitter phase χ is locked to the band shown in Figure 4.11. Error bars from repeated sweeps are omitted if smaller than the markers. **a** Singles counts at the quitter outputs when sending in photons in $|d\rangle_1$ and blocking all other inputs. This is the only state whose preparation changes with φ_{abcd} . These singles exhibit an average variation of $< 1\%$. **b** Twofold output coincidences when sending in states $|d\rangle_1 |a\rangle_4$ from source $\mathcal{B}$. The largest variation is 1.6% for 5&6 coincidences and the rest vary by $< 1\%$. **c** Threefold output coincidences when sending in states $|d\rangle_1 |b\rangle_2 |c\rangle_3$ (as a representative set) are constant within error. **d** Fourfold output coincidences showing double emissions from a single source when the other is blocked ($\mathcal{AA}$, $\mathcal{BB}$), the total counts with both sources open, and the background-subtracted signal (bg-sub). The total signal exhibits the expected $-\cos \varphi_{abcd}$ dependence for P_{1111}^{tot} of equation 4.22 with a fitted visibility of $6.9 \pm 1.8\%$ and total counts per data point of between 2477 and 2275. The double emission backgrounds are constant within error and so we confirm they contribute a flat background to the total signal. Subtracting them gives the purple background-subtracted signal with $23 \pm 6\%$ visibility and counts per point of between 1104 and 756.

In order to verify that contributions from three-photon exchanges (depending on triad phases) have been removed, we plot threefold coincidences when sending in $|d\rangle_1 |b\rangle_2 |c\rangle_3$ in Figure 4.19c. These are all constant within error, as are those recorded for when different inputs are open (see Appendix G.3) and so triad phase terms have been eliminated, as expected

for this pairwise distinguishable state preparation.

Finally the data corresponding to our desired measurement of distinguishable state interference for four photons is shown in Figure 4.19d. At each φ_{abcd} setting we first open all shutters so the emission from both sources is injected into the quitter. The resulting ‘total’ output fourfold coincidences are plotted as a function of φ_{abcd} (blue data points). A cosine fit is shown in blue that has a visibility of $6.9 \pm 1.8\%$ and demonstrates the expected $-\cos \varphi_{abcd}$ dependence of P_{1111}^{tot} from equations 4.10 and 4.22. Assuming equally likely source firings, this total signal including contamination from constant double emissions is predicted to have a visibility of 7.5%. This value is determined from a simulation that includes residual distinguishability $r_{ij} = 0.975$ (from independent HOM measurements) and the effects of six-photon emissions described by the squeezing parameter $\lambda = 0.16$ and transmissions of 80% on the inputs and outputs of the quitter. For slightly different firing probabilities, the visibility is expected to decrease by around 0.5%. Therefore the experimentally measured total signal shape and visibility are both consistent with simulation. Next each source is blocked in turn and fourfolds arising from double firings are recorded independently for the same amount of time as total fourfolds. These are indicated by $\mathcal{AA}$ and $\mathcal{BB}$ in Figure 4.19d and respectively have about 675 and 800 counts per data point. These fourfolds depend only on χ and the pairwise distinguishabilities (equation 4.23). The dependence on the former averages to a constant due to locking, and the latter are constant with φ_{abcd} . Experimentally the signals are constant with φ_{abcd} and so contribute a flat background to the total signal. We then perform the background subtraction described in Section 4.4.3 to obtain the purple data in Figure 4.19d. A fitted cosine (purple curve) to this ‘bg-sub’ data has a visibility of $23 \pm 6\%$. This is consistent with a prediction of 20% from a simulation using the parameters already mentioned, where the reduction from the value of 26.5% quoted at the end of Section 4.4.1 is due to residual distinguishability, six-photon contamination and losses.

The variations in total and background-subtracted fourfolds observed in Figure 4.19d cannot be attributed to variations in the lower order exchange contributions. The cosine variation arises from a dependence on the four-particle phase φ_{abcd} defined by our preparation of pairwise distinguishable states. This demonstrates that distinguishability is not always accompanied by a return to probabilistic behaviour: multiparticle interference can persist.

4.6 Conclusions

In this chapter we introduced a graph model to treat distinguishability contributions in interference and found that triad phases can act as primitives for decomposing higher-order multiparticle phases. For four particles we found a configuration of states for which the four-particle phase cannot be expressed in this way, but is still well-defined despite pairs of the participating states being distinguishable. We then discussed the construction of a balanced four-mode interferometer and the generation and preparation of four photons in these states. This led to the demonstration of a variation in fourfold output coincidences independent of lower-order exchange contributions, and confirmed the observation of multiparticle interference of photons in pairwise distinguishable states.

Extension to more particles is possible, albeit with a smaller signal due to decreasing overlaps [252]. The example here is the first instance where triad phases are not sufficient to describe the interference. As mentioned earlier, other demonstrations of distinguishable state interference have relied on entanglement or masking of distinguishing information. Here it is purely due to the exchange symmetry of the initially separable multiparticle wavefunction. Both the double slit and HOM experiments have compelled physicists to revise what qualifies as intuition in the quantum world: distinguishability appears to be accompanied by a loss of interference and a return to probabilistic behaviour [276]. Our result shows that, on introducing another pair of independent photons, interference effects are possible that contradict even this intuition.

Distinguishability and mixedness

We have so far confined our investigations to the effect of distinguishability on interference and have developed a thorough appreciation of its role in systems of three and four particles. As the scale of photonic quantum technologies grows, it will be important to assess the effect of distinguishing information, for example arising from mismatched path lengths in some circuit, on the fidelity of some desired task. The lessons learnt in previous chapters – such as the properties of different exchange contributions, or the possibility of distinguishable state interference – provide invaluable intuition when approaching such a problem.

What other imperfections can arise in systems reliant on large-scale interference? In [Chapter 3](#) we saw that correlations can lead to spectral mixedness in the emission from heralded nonlinear sources. While it is possible to include mixedness when calculating interference statistics, simply plugging in numbers makes it difficult to grasp precisely the qualitative differences from, for example, spectral distinguishability. Here we perform a thorough analysis of some small dimensioned systems to gain an intuitive picture of the different constraints that distinguishability and mixedness introduce. As well as diagnosing the effect of realistic imperfections on interference degradation, these considerations also permit characterisation of scattering matrices impossible using single photon and HOM measurements.

In what follows, Malte Tichy (MCT) had the idea of diagnosing interference degradation by looking at which parts of the interference landscape are accessible. MCT determined the formula for mixed state scattering probabilities and conceived the comparisons presented in [Sections 5.1](#) and [5.2](#). The experimental data and remaining sections are all my own work.

5.1 Interference of mixed states

5.1.1 Scattering statistics for mixed states

We have so far dealt with photons in separable pure states but what happens to interference if the states are mixed? The loss of coherence means that rather than summing probability amplitudes for indistinguishable processes, measurement outcomes are determined by summing probabilities associated with the scattering of constituent states in the ensemble.

Suppose the photons injected into input i are described by density matrices

$$\rho_i = \sum_{j=1}^R p_{i,j} |\phi_{i,j}\rangle \langle \phi_{i,j}|. \quad (5.1)$$

R is the maximum number of states in some pure state decomposition over all input photons [165, 167]. We have diagonalised the density matrix for each input mode so $\langle \phi_{i,j} | \phi_{i,k} \rangle = \delta_{j,k}$ and, since this is done independently for each photon, the products $\langle \phi_{i,j} | \phi_{m,k} \rangle$ for $m \neq i$ are not necessarily constrained by orthogonality. Now the scattering probability for N particles in an interferometer M , from input configuration $\vec{r}$ to output $\vec{s}$ and with states given by the list $\vec{\rho} = (\rho_1, \dots, \rho_N)$ is [112]

$$P(\vec{r}, \vec{s}, M, \vec{\rho}) = \sum_{k_1, \dots, k_N=1}^R \left(\prod_{i=1}^N p_{i,k_i} \right) \times P(\vec{r}, \vec{s}, M, \mathcal{S}[\vec{k}]). \quad (5.2)$$

The probabilities on the right are those for pure states given in equation 2.54. $\mathcal{S}[\vec{k}]$ denotes distinguishability matrices with elements $\mathcal{S}_{j,l}[\vec{k}] = \langle \phi_{j,k_j} | \phi_{l,k_l} \rangle$.

5.1.2 Scattering of two and three photons

As an example, consider two photons in mixed states ρ_1 and ρ_2 interfering in a 2×2 interferometer. The output coincidence probability is:

$$\begin{aligned} P_{11}^m &= \sum_{j,k=1}^R p_{1,j} p_{2,k} \left[\text{perm}(M * M^*) + |\langle \phi_{1,j} | \phi_{2,k} \rangle|^2 \times \text{perm}(M * M_{(2,1),\mathbb{I}}^*) \right] \\ &= \text{perm}(M * M^*) + \text{Tr}(\rho_1 \rho_2) \times \text{perm}(M * M_{(2,1)}^*). \end{aligned} \quad (5.3)$$

The identity acting on the columns of M^* has been dropped for concision. It is now the trace of the product of the two density matrices that determines interference strength. This is a real number that is zero for states that are orthogonal or maximally mixed in an infinite dimensional Hilbert space, and unity for identical pure states. Just as the real pairwise distinguishability captured exchange symmetry for pure states, here the pairwise contribution is again quantified by a single parameter.

For pure states, the introduction of another photon resulted in four parameters describing their distinguishability. Turning to the coincidence statistics for three photons in mixed states, the exchange contributions corresponding to full permutation elements of S_3 are

$$\begin{aligned} P_{111}^{(2,3,1)} &= \sum_{j,k,l=1}^R p_{1,i} p_{2,j} p_{3,k} \langle \phi_{1,j} | \phi_{2,k} \rangle \langle \phi_{2,k} | \phi_{3,l} \rangle \langle \phi_{3,l} | \phi_{1,j} \rangle \times \text{perm}(M * M_{(2,3,1)}^*), \\ P_{111}^{(3,1,2)} &= \sum_{j,k,l=1}^R p_{1,i} p_{2,j} p_{3,k} \langle \phi_{1,j} | \phi_{3,l} \rangle \langle \phi_{3,l} | \phi_{2,k} \rangle \langle \phi_{2,k} | \phi_{1,j} \rangle \times \text{perm}(M * M_{(3,1,2)}^*). \end{aligned} \quad (5.4)$$

In Appendix H we show that these permanents of scattering matrices are related by complex

conjugation, meaning the coincidence probability can be written as

$$\begin{aligned}
 P_{111}^m = & \text{perm}(M * M^*) + \text{Tr}(\rho_1 \rho_2) \text{perm}(M * M_{(2,1,3)}^*) + \text{Tr}(\rho_1 \rho_3) \text{perm}(M * M_{(3,2,1)}^*) \\
 & + \text{Tr}(\rho_2 \rho_3) \text{perm}(M * M_{(1,3,2)}^*) + 2 \text{Re} [\text{Tr}(\rho_1 \rho_2 \rho_3)] \times \text{Re} [\text{perm}(M * M_{(2,3,1)}^*)] \\
 & - 2 \text{Im} [\text{Tr}(\rho_1 \rho_2 \rho_3)] \times \text{Im} [\text{perm}(M * M_{(2,3,1)}^*)].
 \end{aligned} \quad (5.5)$$

The first term is the single-particle interference contribution and the next three terms are the pairwise exchanges. The final terms depend on the complex number $\text{Tr}(\rho_1 \rho_2 \rho_3)$ so there are now five parameters affecting the coincidence probability. In Section 5.3 we will provide an intuitive interpretation of this complex number for qubits.

5.1.3 Overlaps of density matrices

From the examples above it looks like traces of products of density matrices are the parameters that govern interference strength. It has been shown that the scattering probabilities of equation 5.2 may, for singly occupied inputs, be rewritten as [112, 114]

$$P(\vec{r}, \vec{s}, M, \vec{\rho}) = \frac{1}{\prod_j s_j!} \sum_{\sigma_j \in S_N} \left[\left(\prod_{\mu_i(\sigma_j)} \text{Tr} \left[\prod_k \rho_{\mu_i^k(\sigma_j)} \right] \right) \times \text{perm}(M * M_{\sigma_j}^*) \right]. \quad (5.6)$$

$\mu(\sigma)$ is the cycle structure of that particular element, μ_i is the i th cycle, and μ_i^k is the k th element of the i th cycle. For example in S_3 , $\sigma_2 = (2, 1, 3)$ has cycle structure $\mu(\sigma_2) = (12)(3)$, meaning $\mu_1(\sigma_2) = (12)$ and $\mu_2(\sigma_2) = (3)$. The dependence on $\vec{\rho}$ is then $\sim \text{Tr}(\rho_1 \rho_2) \text{Tr}(\rho_3) = \text{Tr}(\rho_1 \rho_2)$. There is a direct correspondence between the cycle structure of elements of the symmetric group (that can be read off using the graph model presented in Section 4.1.2) and the overlaps of density matrices in this expression. These overlaps are basis invariant, cyclically invariant and, in the limit of unit purity, reduce to products of pure state overlaps. Earlier when considering the number of free parameters in the distinguishability matrix $\mathcal{S}$, we found that N photons prepared in pure states are described by $(N-1)^2$ distinguishing parameters. For an input configurations of one photon per port but now allowing mixedness, there are $N! - 1$ real parameters, consistent with the five needed for the three states in the previous section. Generalising further to an arbitrary partially distinguishable mixed state of N photons in N modes is found to require $\binom{N^2+N-1}{N} - 1$ real parameters [109].

5.2 Exploring the interference landscape

5.2.1 Temporal delays and identical mixed qubits

A realistic defect that might be encountered in integrated photonics is mismatched path lengths in a circuit. If the mismatch is comparable to the coherence length of the photons then temporal distinguishability will affect interference. Another possible imperfection is spectral mixedness from photon sources that are not perfectly factorable (equation 3.17). A simple way of modelling this is to assume mixedness over a qubit degree of freedom. In this section we investigate the

different effects these faults can have on two- and three-photon interference.

The pairwise distinguishability r_{ab}^2 and the pairwise overlap $\text{Tr}(\rho_a \rho_b)$ control interference strength for two photons in separable pure and mixed states respectively. In both cases the parameter is a real number between zero and one that can be tuned over the same range using either distinguishability or state purity. The output probabilities for a balanced beam splitter are constrained by $P_{11} + 2P_{20} = 1$ so the landscape of accessible statistics is highly restricted whatever degrees of freedom are adjusted.

We saw in Chapter 3 that these constraints are relaxed for three photons thanks to the added complexity introduced by extra exchange contributions. Unlike the pairwise exchanges, there is no simple correspondence between the products of three pure state overlaps and the overlaps $\text{Tr}(\rho_a \rho_b \rho_c)$. To highlight the different statistics accessible using pure and mixed states, we will consider two preparations for scattering in a tritter: photons whose partial distinguishability is tuned using the two relative temporal delays, and photons in the same qubit state with varying purity. We already investigated temporal distinguishability for three photons in Section 3.4 and have experimental data for the various output probabilities. For the latter we use polarisation for the qubit instead of different spectral modes, and assume photons with the state

$$\rho_p = p |H\rangle \langle H| + (1 - p) |V\rangle \langle V| \quad (5.7)$$

where the probability p is related to the state purity $\mathcal{P}$ by $p = \frac{1}{2} (1 + \sqrt{2\mathcal{P} - 1})$. When three photons prepared in this way are injected into a balanced tritter, the counting statistics are given by the weighted sum

$$\begin{aligned} P_{ijk}^m = & p^3 P_{ijk}^{HHH} + p^2(1 - p) (P_{ijk}^{HHV} + P_{ijk}^{HVV} + P_{ijk}^{VHH}) \\ & + p(1 - p)^2 (P_{ijk}^{HVV} + P_{ijk}^{VHV} + P_{ijk}^{VVH}) + (1 - p)^3 P_{ijk}^{VVV}. \end{aligned} \quad (5.8)$$

$P_{ijk}^{\alpha\beta\gamma}$ are the output probabilities for three photons prepared in pure polarisations $\alpha, \beta, \gamma \in (H, V)$ and injected into ports 1–3 respectively. Experimentally we use the setup in Figure 3.12 where two KDP sources generate three spectrally indistinguishable photons. These are temporally overlapped and their polarisations are prepared in the eight combinations of equation 5.8. Pseudo-number resolved statistics are collected at all outputs and the probabilities P_{ijk}^m are then calculated in post-processing as a function of the parameter p governing purity. Simulated signals and experimental results for accessible interference statistics are shown in Figure 5.1.

Identical mixed qubits (red in Figure 5.1) access a much smaller space than temporal delays (blue regions), which is reasonable because only the single parameter p is adjusted. Temporal distinguishability is tuned using two relative delays and accesses an effective qutrit space. In the simulations, the areas at the extreme corners of the contours where both types of statistics overlap correspond to states with large overlap. There is generally good quantitative agreement between the shapes of accessible regions for experiment and simulation, and we attribute small disparities to sensitivities of the probability scalings to losses in the system.

From these plots it is clear – unsurprisingly – that as system size grows, the regions of accessible statistics increase. Also temporal distinguishability and mixedness do not map out the same regions, and the choice of which parameters are tuned may restrict access to statistics

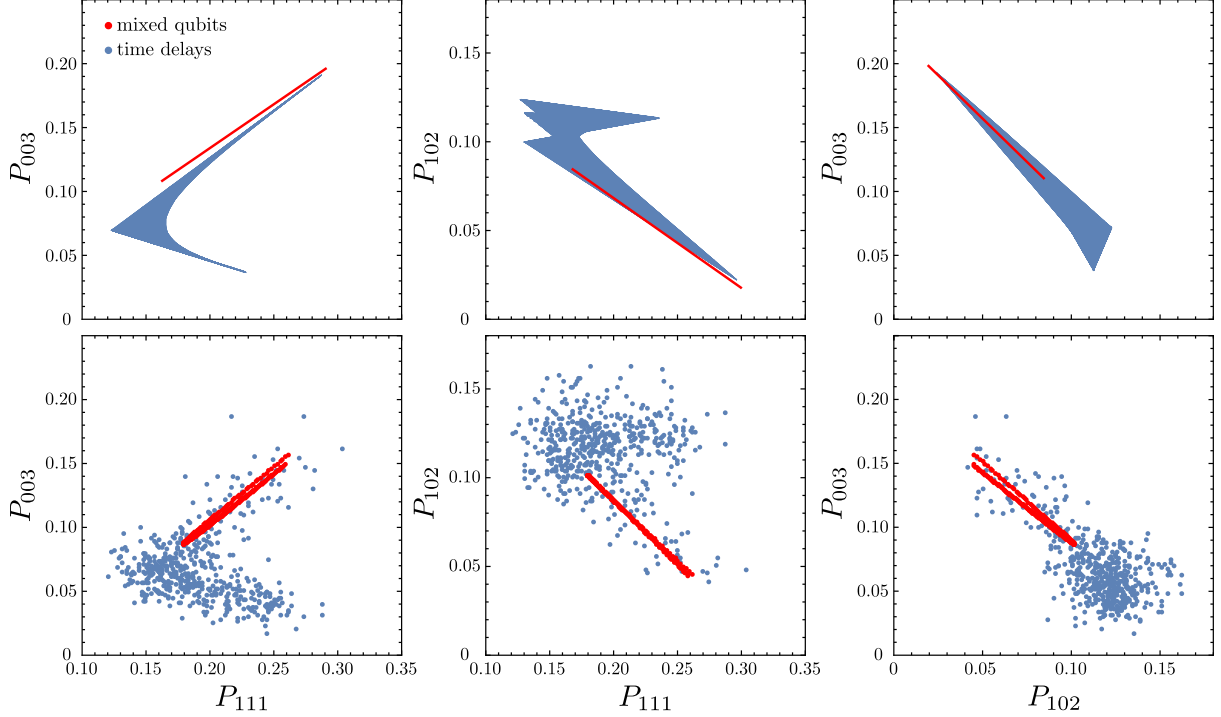


Figure 5.1: Plots of the three-photon interference statistics accessed using identical mixed qubits (red) or pure states whose distinguishability is controlled by relative delays (blue). The top row are signals simulated using the measured tritter unitary of equation 3.35 and including the effects of residual distinguishability, higher-order emissions and losses (described in detail in Section 3.4.2). The experimentally determined probabilities in the bottom row are measured by injecting three photons from the KDP sources into our fibre tritter (see main text for details). The addition of a third photon allows access to a much larger landscape.

in quite different ways. For example, there are here combinations of probabilities that can be attributed unambiguously to temporally distinguishable states as opposed to identically mixed qubits. Generally it could be possible to diagnose the source of interference degradation by detailed analysis of data from an interference experiment, something not possible for the two-photon case as will be shown in Section 5.3.

5.2.2 Routes from perfect to zero state overlap

What happens if we allow mixture over a larger dimensioned space than the qubit assumed so far? For example it might be that the emission of some heralded photon sources is actually mixed over a large number of frequency modes. We know that indistinguishability is associated with perfect interference and distinguishability, unless controlled as in Chapter 4, is typically accompanied by a return to classical behaviour. Full indistinguishability of three identical wavepackets is achieved when they overlap temporally, and they are distinguishable when the relative delays are much larger than the coherence time. It is also possible to follow the same trajectory from unit to zero overlap in the overlaps of density matrices mixed in a large space: consider injecting identical mixed states into each input of a tritter

$$\rho = \sum_j p_j |\phi_j\rangle \langle \phi_j|. \quad (5.9)$$

p_j is the probability of preparing state $|\phi_j\rangle$ in this decomposition of the mixed state and, as the Hilbert space dimension approaches infinity, can be made arbitrarily small. The overlaps governing interference strength are now

$$\text{Tr}(\rho^2) = \sum_j p_j^2, \quad \text{Tr}(\rho^3) = \sum_j p_j^3. \quad (5.10)$$

For identical pure states these are unity and there is perfect interference. On the other hand, maximally mixed states in a large space mean p_j tends to 0 and no interference occurs. Substituting these expressions into the output probabilities of equation 5.6 gives

$$\begin{aligned} P_{111} &= \frac{1}{9} (2 - 3\text{Tr}(\rho^2) + 4\text{Tr}(\rho^3)), \\ P_{\{210\}} &= \frac{1}{9} (1 - \text{Tr}(\rho^3)), \\ P_{\{300\}} &= \frac{1}{27} (1 + 3\text{Tr}(\rho^2) + 2\text{Tr}(\rho^3)). \end{aligned} \quad (5.11)$$

The configurations in braces indicate all permutations of those output occupations and the symmetry of the tritter means those of the same type are equal. Here the interference is only affected by state purity. It is difficult to experimentally access large Hilbert spaces so we perform simulations for a large set of p_j to map out a subset of these output probabilities, including cases of zero and unit overlap. These are compared to the temporally distinguishable states of the previous section in Figure 5.2.

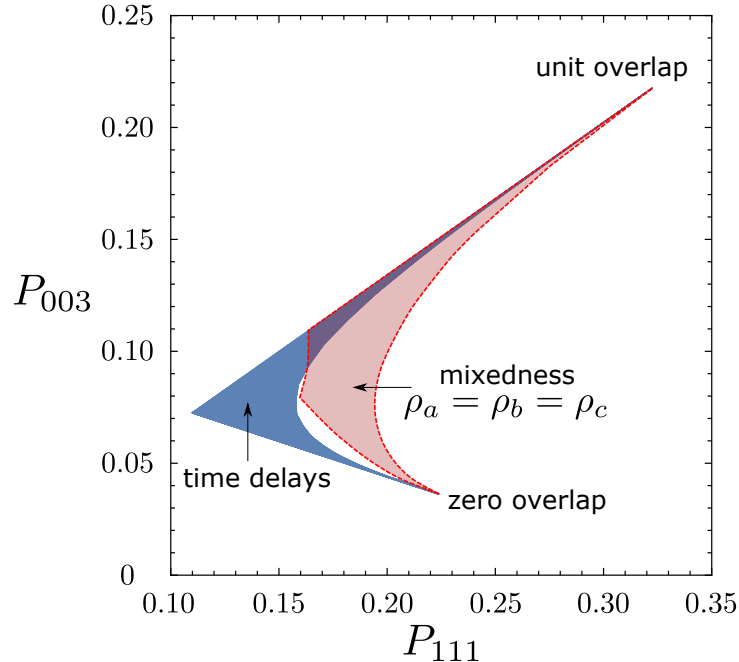


Figure 5.2: A subset of three-photon interference statistics accessible when interfering either identical mixed states of varying purity (red), or pure states with temporally tunable distinguishability (blue). The statistics accessed at the top right correspond to indistinguishable pure states with unit overlap, and those at the bottom right correspond to $r_{ij}^2 = 0$ and $\text{Tr}(\rho_i \rho_j) = 0$. Here the coherence of pure state interference permits access to parts of the landscape that are never reached for identical mixed states of any purity.

These state preparations result in very different routes from strong interference back to probabilistic behaviour. Tuning the relative delays of three wavepackets accesses a constrained output space (Appendix B.4), and it is this restriction that prevents pure state interference accessing the red region in Figure 5.2. Knowledge of the degrees of freedom accessed in an experiment is vital when determining sources of interference degradation.

5.3 Collective distinguishability for mixed qubits

5.3.1 HOM interference of mixed states

We now investigate which properties of interference can be attributed uniquely to mixedness or distinguishability. To start, consider injecting two photons prepared in states ρ_a and ρ_b into the ports of a balanced beam splitter. Using equation 5.3 for mixed state probabilities, the output coincidence probability is

$$P_{11}^m = \frac{1}{2} (1 - \text{Tr}(\rho_a \rho_b)). \quad (5.12)$$

$\text{Tr}(\rho_a \rho_b)$ governs two-particle interference strength, and reduces to the pairwise distinguishability r_{ab}^2 for pure states. It can also be written as [277]

$$\text{Tr}(\rho_a \rho_b) = \frac{1}{2} (\text{Tr}(\rho_a^2) + \text{Tr}(\rho_b^2) - \|\rho_a - \rho_b\|^2). \quad (5.13)$$

The first two terms are the individual state purities and the third is the Frobenius norm $\|A\|^2 = \text{Tr}(A^\dagger A)$. This is also known as the Hilbert-Schmidt norm and may be interpreted as a measure of distance between two states. It is zero for indistinguishable states and two when they are distinguishable. The visibility of this HOM dip would be $V = \text{Tr}(\rho_a \rho_b)$. We know that for pure states, V quantifies the pairwise distinguishability. On the other hand if the states are identical then V gives the mean purity of the states [209]. This expression shows that degradation due to mixedness and distinguishability cannot be separated in two-photon interference.

This is clear if we consider a qubit internal degree of freedom that, for two independent particles, is sufficient to probe all values of this overlap. Expressing the density matrices as $\rho_i = \frac{1}{2} (\mathbb{I} + \mathbf{r}_i \cdot \boldsymbol{\sigma})$ gives

$$\text{Tr}(\rho_a \rho_b) = \frac{1}{2} (1 + \mathbf{r}_a \cdot \mathbf{r}_b). \quad (5.14)$$

The dot product of Bloch vectors gives no information on whether it is vector lengths or relative orientation that reduce its value from unity, as shown in Figure 5.3.

5.3.2 Generalising the triad phase to mixed states

We have seen that the triad phase appears as a distinguishing quantity for three photons and for qubits it is given by half the solid angle subtended in the Bloch sphere. In the previous section we saw that the introduction of mixedness means five parameters affect interference statistics: three pairwise overlaps $\text{Tr}(\rho_i \rho_j)$ and the complex number $\text{Tr}(\rho_a \rho_b \rho_c)$. In order to appreciate the role of mixedness in three-photon interference, we restrict our discussions to qubits. This does not allow full exploration of these parameters but allows an intuitive geometric interpretation.

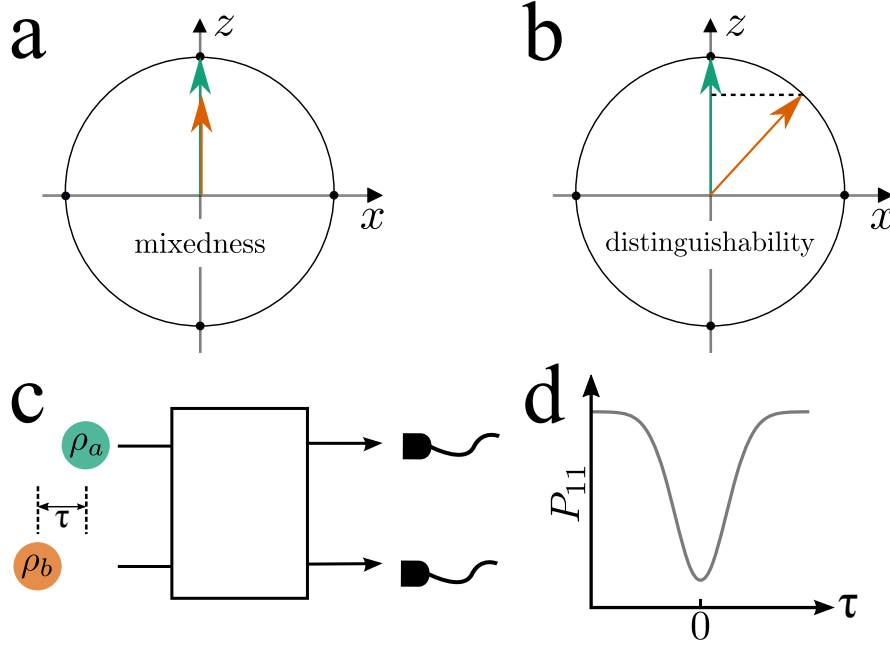


Figure 5.3: The dot product of two Bloch vectors is reduced by mixedness (a) and distinguishability (b) in the same way. c Consider injecting two photons prepared in qubit states denoted by the red and blue Bloch vectors into a balanced beam splitter with a controllable time delay τ . d The probability of coincidences at the outputs ports P_{11} will reach a minimum at zero delay, and the depth of this HOM interference dip will be the same for cases a and b.

The triple overlap for qubits can be written as (see Appendix H.2)

$$\text{Tr}(\rho_a \rho_b \rho_c) = \frac{1}{4} (1 + \mathbf{r}_a \cdot \mathbf{r}_b + \mathbf{r}_a \cdot \mathbf{r}_c + \mathbf{r}_b \cdot \mathbf{r}_c + i \mathbf{r}_a \cdot (\mathbf{r}_b \times \mathbf{r}_c)) := r_{abc} \times e^{i\varphi'}. \quad (5.15)$$

The dot products describe the pairwise overlaps of the states. The imaginary component encodes a collective description via the scalar triple product of the three Bloch vectors: $\tilde{V}_{abc} = \mathbf{r}_a \cdot (\mathbf{r}_b \times \mathbf{r}_c)$. This is a pseudoscalar with a magnitude determined by the volume of the spanned parallelepiped and a sign set by its orientation in a right-handed frame (see Figure 5.4). It is zero if the vectors are coplanar and generally contains information on areas and orientations not captured by dot products alone. The volume of the parallelepiped can be written as [278]

$$\begin{aligned} V_{abc} &= |\tilde{V}_{abc}| = |\mathbf{r}_a \cdot (\mathbf{r}_b \times \mathbf{r}_c)| \\ &= r_a r_b r_c [1 - (\hat{\mathbf{r}}_a \cdot \hat{\mathbf{r}}_b)^2 - (\hat{\mathbf{r}}_a \cdot \hat{\mathbf{r}}_c)^2 - (\hat{\mathbf{r}}_b \cdot \hat{\mathbf{r}}_c)^2 + 2(\hat{\mathbf{r}}_a \cdot \hat{\mathbf{r}}_b)(\hat{\mathbf{r}}_a \cdot \hat{\mathbf{r}}_c)(\hat{\mathbf{r}}_b \cdot \hat{\mathbf{r}}_c)]^{\frac{1}{2}} \end{aligned} \quad (5.16)$$

where r_i is the length of vector $\mathbf{r}_i$ and $\hat{\mathbf{r}}_i = \mathbf{r}_i / r_i$ are unit vectors. If $r_i=1$ for pure qubits, then the volume V_{abc} is completely determined by dot products of vectors. These can be obtained from HOM experiments (see equation 5.14) and additionally we recover $r_{abc} = |\langle a|b \rangle \langle b|c \rangle \langle c|a \rangle|$ for the appropriate pure state vectors. The argument φ' of $\text{Tr}(\rho_a \rho_b \rho_c)$ is given by

$$\tan \varphi' = \frac{\mathbf{r}_a \cdot (\mathbf{r}_b \times \mathbf{r}_c)}{1 + \mathbf{r}_a \cdot \mathbf{r}_b + \mathbf{r}_a \cdot \mathbf{r}_c + \mathbf{r}_b \cdot \mathbf{r}_c}. \quad (5.17)$$

If the qubits are pure then this expression yields $\varphi' = \Omega/2$ where Ω is the subtended solid angle [278, 279]. This is the triad phase we defined for qubits in Section 3.1.1.

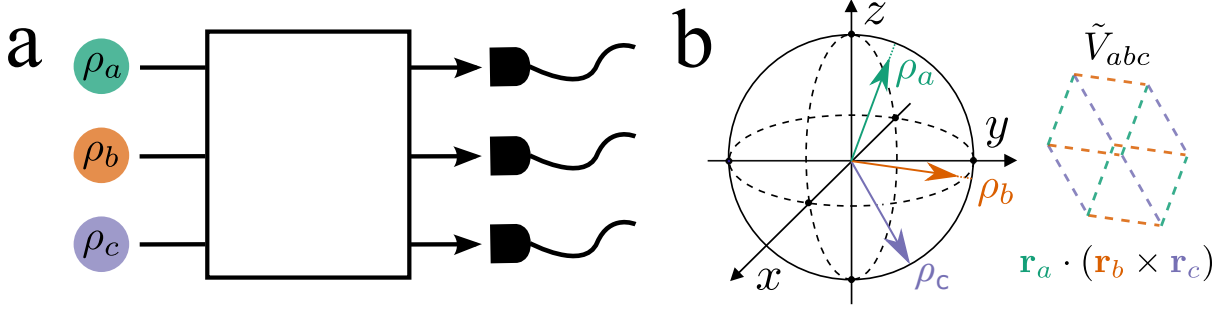


Figure 5.4: **a** Three photons with qubit internal states ρ_i are injected into the inputs of some interferometer and output statistics are collected. **b** The scattering probabilities depend on the dot products of Bloch vectors, associated with pairwise exchanges, and also on their scalar triple product. This can be interpreted geometrically as the signed volume of the spanned parallelepiped $\tilde{V}_{abc}$.

If the qubits are not pure then $\tilde{V}_{abc}$ is the appropriate collective distinguishing parameter. It can be used to identify mixedness in a way impossible using two-photon interference. If one could measure HOM visibilities and V_{abc} , then equation 5.16 would allow identification of $r_i \neq 1$. As a concrete example, suppose we have a set of three photons in qubit states that all have the same pairwise state overlap, but we do not know whether reduction from unity is due to mixedness or distinguishability. Such a situation for pure states has been considered in assessing the complexity of boson sampling [132]. The comparison could be of interest in diagnosing poor HOM interference of photons from some sources where full state tomography cannot be performed. We shall see that $\tilde{V}_{abc}$ permits discrimination between the two cases.

In the first case we consider preparing the following states as inputs for ports 1–3 respectively:

$$\begin{aligned} |a\rangle &= \cos\left(\frac{\theta}{2}\right) |H\rangle + \sin\left(\frac{\theta}{2}\right) |V\rangle, \\ |b\rangle &= \cos\left(\frac{\theta}{2}\right) |H\rangle + e^{i\frac{2\pi}{3}} \sin\left(\frac{\theta}{2}\right) |V\rangle, \\ |c\rangle &= \cos\left(\frac{\theta}{2}\right) |H\rangle + e^{i\frac{4\pi}{3}} \sin\left(\frac{\theta}{2}\right) |V\rangle. \end{aligned} \quad (5.18)$$

These have Bloch vectors of unit length and are equally spaced in azimuthal angle in the Bloch sphere. They lie at the top of the sphere when $\theta = 0$ and are equally spaced in the equator when $\theta = \pi/2$ (see Figure 5.5a). The pairwise overlaps $\text{Tr}(\rho_i \rho_j) = \frac{1}{8}(5 + 3 \cos 2\theta)$ and vary between 0.25 and 1. The spanned volume $\tilde{V}_{abc} = -3\sqrt{3}/2 \times \cos \theta \sin^2 \theta$ so V_{abc} varies from 0 to a maximum of 1 (see Figure 5.5b).

The second case involves interfering three photons in the same mixed qubit state that we considered in Section 5.2.2:

$$\rho_q = p |H\rangle \langle H| + (1-p) |V\rangle \langle V|. \quad (5.19)$$

The length of the Bloch vector $r_q = 2p - 1$ and the state purity $\mathcal{P} = \frac{1}{2}(1 + r_q^2)$ (see Figure 5.5c). Here $\text{Tr}(\rho_i \rho_j)$ can vary between 0.5 and 1, overlapping with the range possible for the pure state configuration but the spanned volume is always zero. Now given input Bloch vectors $\mathbf{r}_i$, the

scattering statistics in a tritter $U_{trit}^{(1)}$ are:

$$\begin{aligned}
 P_{111} &= \frac{1}{18} (3 + \mathbf{r}_a \cdot \mathbf{r}_b + \mathbf{r}_a \cdot \mathbf{r}_c + \mathbf{r}_b \cdot \mathbf{r}_c), \\
 P_{(120)} &= \frac{1}{36} \left(3 - \mathbf{r}_a \cdot \mathbf{r}_b - \mathbf{r}_a \cdot \mathbf{r}_c - \mathbf{r}_b \cdot \mathbf{r}_c - \sqrt{3} \tilde{V}_{abc} \right), \\
 P_{(210)} &= \frac{1}{36} \left(3 - \mathbf{r}_a \cdot \mathbf{r}_b - \mathbf{r}_a \cdot \mathbf{r}_c - \mathbf{r}_b \cdot \mathbf{r}_c + \sqrt{3} \tilde{V}_{abc} \right), \\
 P_{(300)} &= \frac{2}{3} P_{111}.
 \end{aligned} \tag{5.20}$$

Brackets around output configurations denote those related by cyclic permutation of occupation numbers. The proportionality of coincident and bunched statistics and the equality of partially bunched counts related by cyclic permutations are due to the high symmetry of the tritter. The reason $\tilde{V}_{abc}$ only appears in the latter will be explained in the next section but here shows that partially bunched statistics must be collected to measure this new quantity. In Figure 5.5d we plot these theoretical probabilities for the two state preparations.

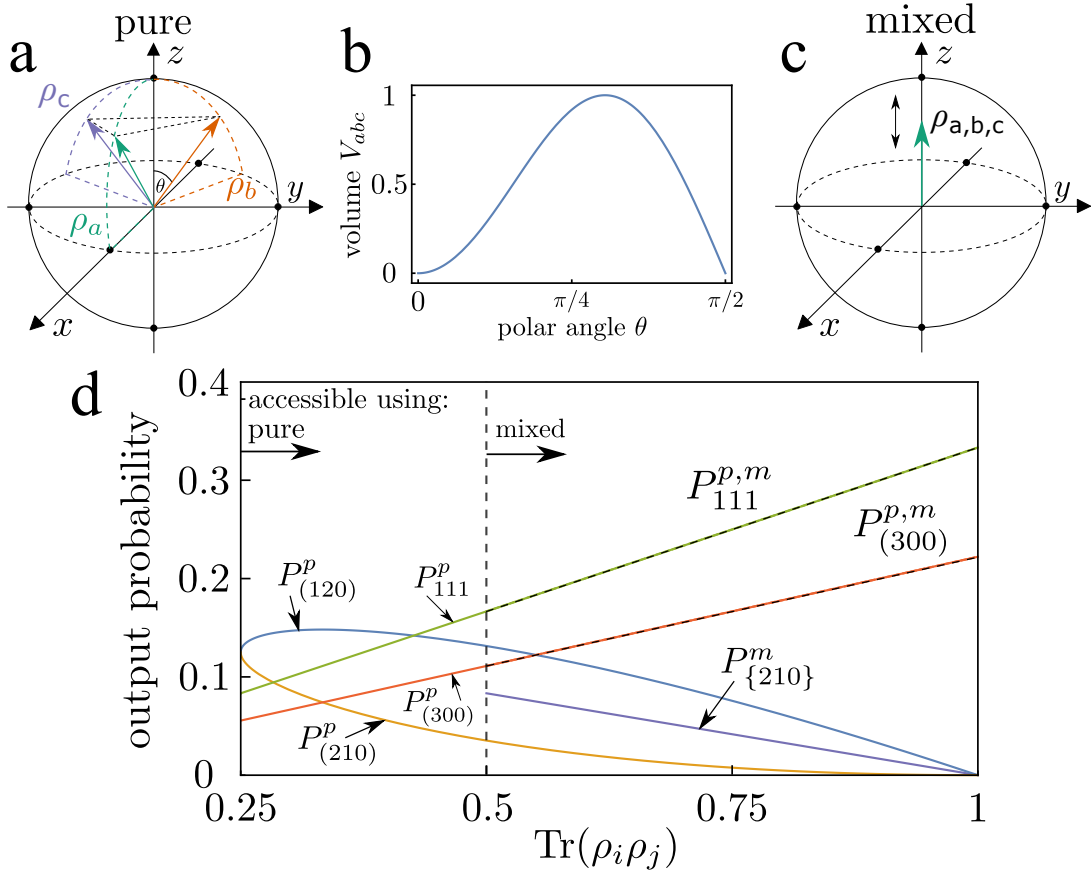


Figure 5.5: **a** The polarisation states from equation 5.18 are equally spaced in azimuthal angle and pairwise overlaps $\text{Tr}(\rho_i \rho_j)$ vary monotonically with polar angle θ . **b** The enclosed volume V_{abc} varies non-monotonically and reaches a maximum when $\theta = 1/2 \times \arccos(1/3)$. **c** Polarisation states from equation 5.19 where the Bloch vectors' lengths and pairwise overlaps are controlled by the state purity. The enclosed volume is always zero. **d** Theoretical output probabilities (equation 5.20) for photons in the states of **a** and **c** interfering in the tritter $U_{trit}^{(1)}$. p and m superscripts indicate probabilities for pure and mixed preparations respectively. The latter access a smaller range of pairwise overlaps than the former. Partially bunched statistics can identify the source of reduction of the pairwise overlaps $\text{Tr}(\rho_i \rho_j)$ (see main text for details).

If HOM dip measurements determine $0.5 \leq \text{Tr}(\rho_i \rho_j) \leq 1$ then the source of interference degradation cannot be unambiguously attributed to mixedness or distinguishability. This is also the case for P_{111} and $P_{(300)}$ that only depend on the vector dot products. For the mixed states, all partially bunched probabilities $P_{\{210\}}$ exhibit the same linear dependence on these dot products because $\tilde{V}_{abc} = 0$. However for the pure states, HOM measurements of $\mathbf{r}_i \cdot \mathbf{r}_j$ are not sufficient to fully characterise $P_{(120)}$ and $P_{(210)}$ because of their dependence on V_{abc} shown in Figure 5.5b. The observation that the partially bunched statistics are not all equal therefore indicates distinguishability and permits measurement of this distinguishing volume, thus enabling an identification impossible with lower-order interference.

What happens for more particles and more dimensions? With four qubits the parameter $\text{Tr}(\rho_a \rho_b \rho_c \rho_d)$ contains, besides dot products and scalar triple products, the quantity (Appendix H.2)

$$\begin{aligned} \text{Tr}((\mathbf{r}_a \cdot \boldsymbol{\sigma})(\mathbf{r}_b \cdot \boldsymbol{\sigma})(\mathbf{r}_c \cdot \boldsymbol{\sigma})(\mathbf{r}_d \cdot \boldsymbol{\sigma})) &= 2((\mathbf{r}_a \cdot \mathbf{r}_b)(\mathbf{r}_c \cdot \mathbf{r}_d) - (\mathbf{r}_a \cdot \mathbf{r}_c)(\mathbf{r}_b \cdot \mathbf{r}_d) \\ &\quad + (\mathbf{r}_a \cdot \mathbf{r}_d)(\mathbf{r}_b \cdot \mathbf{r}_c)). \end{aligned} \quad (5.21)$$

Hence the interference of four photons with qubit states is fully described by two- and three-photon parameters. Turning to higher dimensions, a general qutrit state is specified by the eight-component vector $\mathbf{n}$ through [280]

$$\rho_{qutrit} = \frac{1}{3} \left(\mathbb{I} + \sqrt{3} \mathbf{n} \cdot \boldsymbol{\lambda} \right). \quad (5.22)$$

$\boldsymbol{\lambda}$ is a vector of the eight Gell-Mann matrices. We earlier saw that the argument of the triple overlap φ' for qubits depends on the scalar triple product of the Bloch vectors (equation 5.17). For qutrits it is determined by a more complex geometric object amounting to a generalisation of the scalar triple product to higher dimensions [145, 161, 280]. Perhaps in some circumstances these quantities can be decomposed into smaller primitives, analogous to decomposing multiparticle distinguishing phases into triad phases.

5.4 Applications to characterisation tasks

5.4.1 Characterising unitaries

In Section 3.3.2 we discussed how to use one- and two-photon measurements to determine the scattering matrix for an interferometer. Since HOM interference only resolves the relative phases in an interferometer (equation 3.34), this method cannot discriminate between M and the conjugate M^* . Laing *et al.* mention that photon statistics are equal for these matrices so one can simply pick either matrix to describe a device [239]. This is true for fully distinguishable or indistinguishable states where interference depends on $|\text{perm} M|^2$ and $|\text{perm}(M)|^2$ respectively. However when isolating the triad phase we found the partially bunched statistics had dependences on $\cos(\varphi_{abc} \pm 2\pi/3)$ and the sign of the shift depends on whether $U_{trit}^{(1)}$ or $U_{trit}^{(2)} = (U_{trit}^{(1)})^*$ are used (equation 3.38 and Figure 3.18). Moreover in the previous section when constrained to qubits (equation 5.20), these probabilities have a dependence on $\pm \tilde{V}_{abc}$ and the direction of

the shift depends on the unitary. All of this boils down to the interplay of phases defined by the internal degrees of freedom and those of the interferometer itself.

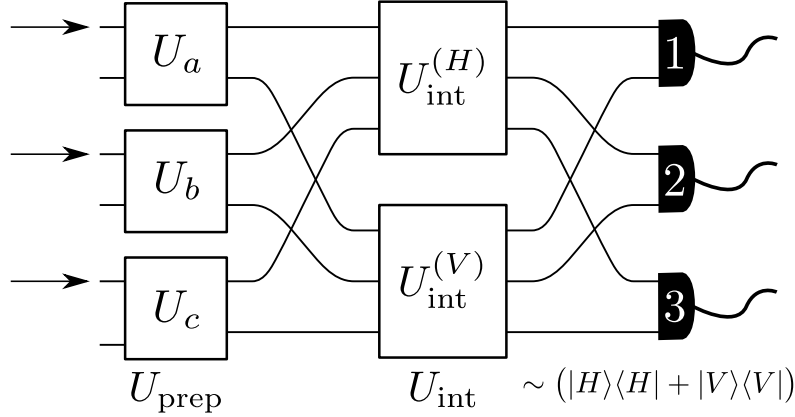


Figure 5.6: Three qubits interfering in a unitary interferometer may be recast as three indistinguishable states injected into a larger interferometer. The first layer of unitaries U_{prep} prepare the qubit states $|a\rangle, |b\rangle, |c\rangle$ in the dual-rail encoding [12]. Assuming the interferometer is polarisation independent, U_{int} acts on the H and V spaces in the same way. Polarisation-insensitive counting at the outputs is achieved by tracing over that degree of freedom. Adapted from [281].

Three photons prepared in qubit states and interfered in a 3×3 interferometer may be recast as indistinguishable states injected into a 6×6 interferometer [20, 281] (see Figure 5.6). Considering interference in the effective larger unitary $U = U_{\text{int}}U_{\text{prep}}$ makes it clear that changing U_{int} to U_{int}^* will generally influence output probabilities. In fact taking the conjugate of U_{prep} results in exactly the same change because only relative phases in the network matter, as evident from $P_{ijk} = \text{Tr}(U \rho_{in}^* U^\dagger \hat{\Pi}_{ijk}) = \text{Tr}(U^* \rho_{in}(U^*)^\dagger \hat{\Pi}_{ijk})$. Another way to see this is that conjugating the preparation unitaries for qubit states amounts to a reflection in the xz plane of the Bloch vectors and this swaps the handedness of solid angles and scalar triple products so φ_{abc} changes to $-\varphi_{abc}$, and $\tilde{V}_{abc}$ changes to $-\tilde{V}_{abc}$.

Partially bunched statistics for three photons may be used to discriminate U and U^* . Fully bunched statistics where all photons end up in the same output port k can never do this, independent of the number of photons and internal degrees of freedom and for general scattering matrix M . There is only one many-particle path contributing to that outcome, so no interplay of interferometer phases occurs:

$$\begin{aligned}
 |\psi_{\text{bunched}}\rangle &= M_{1k}M_{2k}\dots M_{Nk} \left| \psi^{(1)}, \psi^{(2)}, \dots, \psi^{(N)} \right\rangle_k \\
 P_{\text{bunched}} &= \langle \psi_{\text{bunched}} | \psi_{\text{bunched}} \rangle \\
 &= P_{1 \rightarrow k} \times P_{2 \rightarrow k} \times \dots \times P_{N \rightarrow k} \times \left\langle \psi^{(1)}, \psi^{(2)}, \dots, \psi^{(N)} \left| \psi^{(1)}, \psi^{(2)}, \dots, \psi^{(N)} \right\rangle,
 \end{aligned} \tag{5.23}$$

where N photons in states $|\psi^{(j)}\rangle$ are injected into inputs j , and the single particle transition probabilities $P_{j \rightarrow k} = |M_{jk}|^2$ are unchanged on matrix conjugation. This state overlap for photons in the same output port k but with different internal states can be calculated by applying commutation relations, and so these bunched statistics probe only the bosonic properties of photons, not the interference of paths.

So why can threefold coincidences not perform this discrimination for a 3×3 unitary?

This interplay of internal and interferometer phases is only observable when certain matrix permanents are complex. We saw this in equation 5.5 where $\text{Im}[\text{Tr}(\rho_1 \rho_2 \rho_3)]$ is multiplied by $\text{Im}\left[\text{perm}\left(M * M_{(2,3,1)}^*\right)\right]$. As long as the latter is non-zero, discrimination between a matrix and its conjugate is possible. It turns out that when M is any 3×3 unitary, this permanent is always purely real – this can be verified by casting such a unitary in terms of Euler angles [111] and is a special restriction for this small dimensioned unitary. On the other hand, the effective scattering matrix for partial bunchings in the two tritters gives $\text{Im}\left[\text{perm}\left(M * M_{(2,3,1)}^*\right)\right] = \frac{2}{9}e^{\pm i\frac{2\pi}{3}}$ and so they give different output probabilities. When the interferometer size increases, this constraint on permanents of unitaries relaxes and in general coincidence probabilities are sensitive to changing U to U^* . Another special example of when this does not hold is for the quitter presented in the previous chapter, meaning $P_{1111} \sim \cos \varphi_{abcd}$. Hence as soon as an appropriate matrix permanent is complex, three photons defining a known $\tilde{V}_{abc} \neq 0$ can be used to perform a characterisation impossible using the interference of two independent photons.

5.4.2 State tomography using interference

Most experiments in quantum mechanics involve preparing, evolving and measuring a quantum state. It is therefore crucial to have a well-characterised initial system. The no-cloning theorem prevents copying the as-yet uncharacterised quantum state [282]. Instead one can repeatedly prepare the same state and measure the expectations of a set of complete operators spanning the relevant Hilbert space. Frequency of counts are used to estimate the probabilities and the Born rule can be used to reconstruct the density matrix for the system: quantum state tomography.

A d -dimensional system has a density matrix with d^2 parameters. Accounting for the fact that probability normalisation requires normalisation of physical quantities like detector counts, this means measuring expectations of d^2 basis elements for the density matrix [53]. As a concrete example, consider the characterisation of a qubit encoded in polarisation and defined by a Bloch vector $\mathbf{r}$. The vector elements correspond to expectations of the Pauli operators $\mathbf{r} = (x, y, z) = (\langle \hat{\sigma}_x \rangle, \langle \hat{\sigma}_y \rangle, \langle \hat{\sigma}_z \rangle)$ and can be measured by sending photons through suitably oriented quarter- and half-wave plates before counting photons at the outputs of a PBS [283]. Defining the measured counts transmitted and reflected when measuring in $\hat{\sigma}_j$ as $N_T^{(j)}$ and $N_R^{(j)}$, the expectation values are

$$\langle \sigma_j \rangle = \frac{N_T^{(j)} - N_R^{(j)}}{N_T^{(j)} + N_R^{(j)}}. \quad (5.24)$$

Summing counts at the PBS outputs for each basis permits normalisation. Linear inversion then gives the reconstructed density matrix $\rho = \frac{1}{2} \left[\mathbb{I} + \sum_{j=1}^3 \langle \hat{\sigma}_j \rangle \sigma_j \right]$. This inversion often returns matrices that are not positive semi-definite, in other words not density matrices describing a physical system. It is therefore common to apply a maximum likelihood routine to ensure the three defining properties of a density matrix (Hermitian, positive semi-definite and normalised) are automatically satisfied [283].

In the preceding sections we have seen that multiparticle interference can provide information on the phases and purities of qubit states. Suppose a 3×3 unitary has been characterised using three qubits defining some known $\tilde{V}_{abc}$. Now given two photons in known qubit states ρ_a, ρ_b that do not point in the same direction in the Bloch sphere, interference statistics with some third

unknown states ρ_c may be collected. These will contain information on dot products with $\mathbf{r}_c$ and a new $\tilde{V}'_{abc}$ that together provide sufficient information to characterise ρ_c . A downside of this technique is that a large number of counts are needed to accurately reconstruct the multiphoton output probabilities but with improvement in source efficiencies this could become less of an issue. However an advantage is its independence of the degree of freedom employed, provided the interferometer is also insensitive to how the qubit is encoded. For example temporal modes have attracted attention in quantum information protocols, and reconstruction of the density matrix usually calls for active control over the measurement process [36]. This interferometric technique simply requires a 3×3 unitary¹, known partially distinguishable reference states, and then passive photon counting, with no adjustments required during the procedure.

Simulations suggest extensions to more separable photons and higher dimensions are possible using larger interferometers and more reference states. A careful analysis must be performed to verify that the number of independent counting statistics are sufficient to determine all independent parameters of the system's density matrix. Recent results considering the tomography of a general entangled state of indistinguishable photons using interference have also considered this scaling question [284]. Pairing their considerations with our observations resolving distinguishability would be an interesting avenue to explore.

5.5 Conclusions

We have considered the effect of state impurity on the interference of photons in separable states, which opens up a much larger space of accessible statistics in experiments. We considered the effect of realistic experimental imperfections, namely distinguishability and spectral mixedness, on small-scale interference. These two properties were shown to access distinct parts of the interference landscape for three photons, revealing the richer structure accessible beyond HOM interference. The ability to diagnose the source of interference degradation was investigated and, when constrained to a qubit degree of freedom, we presented a geometric quantity that includes the effect of mixedness and that recovers the triad phase of Chapter 3 in the limit of pure states. Generalisations to higher dimensions were discussed before a final comment on applications of multiparticle interference to interferometer and state characterisation.

In larger systems the possible state space grows exponentially with the number of particles, but knowledge of the system under investigation means one can constrain which parts of the space can feasibly be accessed inadvertently due to imperfections. Given strong enough statistics, one can then potentially identify and compensate for such errors. For example imagine you have a large integrated photonic chip designed to execute some simulation task that is hard to perform classically. In order to characterise the likely sources of infidelity, the chip could first be programmed to execute a known simple circuit whose output can be used to diagnose imperfections prior to running the hard task. It would be interesting to see whether this treatment can be applied to other platforms for quantum technology, such as superconducting qubits where decoherence can also be an issue.

¹The high symmetry of the tritter means that the coincident and fully bunched statistics are directly proportional to each other (equation 5.20). This means separate HOM measurements are required to determine $\mathbf{r}_i \cdot \mathbf{r}_j$. The use of a less symmetric unitary allows determination of ρ_c using only three-photon statistics.

Symmetries and multiparticle coherence

We have so far used the second quantised formalism of operators to automatically capture bosonic symmetry. This allowed identification of distinguishing parameters that affect the interference of photons in separable states. Now we return to first quantisation where exchange symmetry of the multiparticle wavefunction is explicit. The group theory underpinning the symmetries of multiparticle Hilbert spaces will be presented and consequently the role of distinguishability in partial exchange symmetry will be explored more rigorously. This should give an experimentalist a clearer picture of how parameters adjusted in the laboratory directly affect wavefunction symmetries.

The first quantised treatment of interference has been covered extensively in the literature [108, 110, 285]. The approach used here is from a paper by Stanisic and Turner [109]. In particular the sections on dualities and scattering probabilities are based on that paper. Its application to multiparticle distinguishability is my work.

6.1 Returning to first quantisation

6.1.1 Hilbert and Fock spaces

We begin with a brief review of some material presented in Chapter 2. Recall that the Hilbert space for a single particle is constructed by taking the tensor product of those for the constituent degrees of freedom, for example $\mathcal{H}^{(1)} = \mathcal{H}_A \otimes \mathcal{H}_B$. The Hilbert space for N -particle wavefunctions is then given by the tensor product of N of these single-particle spaces: $\mathcal{H}^{(N)} = (\mathcal{H}^{(1)})^{\otimes N}$. After constructing the N -particle product state $|\Psi_D\rangle = \otimes_{j=1}^N |\phi_j\rangle$, we symmetrise over all particle permutations to obtain the exchange symmetric bosonic wavefunction:

$$\begin{aligned}
|\Psi_S\rangle &= \hat{S} |\Psi_D\rangle \\
&\equiv \sum_{j=1}^N |\phi_j\rangle \\
&= \mathcal{N}_S \sum_{\sigma \in S_N} |\phi_{\sigma(1)}\rangle \otimes |\phi_{\sigma(2)}\rangle \otimes \dots \otimes |\phi_{\sigma(N-1)}\rangle \otimes |\phi_{\sigma(N)}\rangle.
\end{aligned} \tag{6.1}$$

$\mathcal{N}_S$ is a normalisation constant (equation 2.7). $\vee$ is the symmetric tensor product and $|\Psi_S\rangle$ resides in the symmetrised part of the N -particle Hilbert space: $\mathcal{H}_S^{(N)} = (\mathcal{H}^{(1)})^{\vee N}$. We defined the Fock space $\mathcal{F} = \bigoplus_{j=1}^{\infty} \mathcal{H}_S^{(j)}$ and the introduction of creation and annihilation operators, together with their commutation relations, allowed us to dispense with labelling each particle separately while automatically including the required bosonic symmetry.

This second quantised formalism is excellent at keeping track of particle exchange symmetry and its concision makes it very convenient. However sometimes the notation obscures what is going on. For example consider the state $\hat{a}_{1,H}^\dagger \hat{a}_{2,V}^\dagger |0\rangle$, where the numbers label orthogonal port modes. What is the effective state if we do not have access to the polarisation? The usual approach is to trace out that degree of freedom, but these operators are mappings between states that by definition lie in the symmetrised part of a *joint* Hilbert space. It is difficult to separate the two degrees of freedom because occupations of the separate port and polarisation spaces are correlated due to the symmetrisation procedure.¹ As we will show in the next section, it turns out that this state lies in both the symmetric and anti-symmetric parts of the two-particle port Hilbert space.

6.1.2 Partitioning the multiparticle Hilbert space

For consistency with Stanisic *et al.* [109], we denote the Hilbert space for ports resolved on detection by the ‘system’ space $\mathcal{H}_S$ and that for the unresolved internal degree of freedom by the ‘label’ space $\mathcal{H}_L$.² Hence the single-particle space $\mathcal{H}^{(1)} = \mathcal{H}_S \otimes \mathcal{H}_L$. What does the first quantised wavefunction for a pair of photons indistinguishable in their label look like? Taking this degree of freedom to be polarisation, the state for indistinguishable photons prepared in orthogonal ports ‘1’ and ‘2’ is:

$$\begin{aligned} |\psi_{12}^{HH}\rangle &= \hat{a}_{1,H}^\dagger \hat{a}_{2,H}^\dagger |0\rangle \\ &= |1, H\rangle \vee |2, H\rangle \\ &= \frac{1}{\sqrt{2}} (|1, H\rangle \otimes |2, H\rangle + |2, H\rangle \otimes |1, H\rangle). \end{aligned} \tag{6.2}$$

In the last line we have written the state explicitly in $\mathcal{H}^{(2)} = \mathcal{H}^{(1)} \otimes \mathcal{H}^{(1)}$, and the ordering of the tensor product of kets refers to the states of the first and second particles respectively. The two symbols in each ket implicitly indicate a tensor product of the associated states for each degree of freedom. In this form the exchange symmetry of the state is obvious: swapping the states of the particles leaves $|\psi_{12}^{HH}\rangle$ unchanged. We can reorganise this into separate states for the system and label degrees of freedom:

$$|\psi_{12}^{HH}\rangle = \frac{1}{\sqrt{2}} (|1, 2\rangle + |2, 1\rangle) \otimes |H, H\rangle. \tag{6.3}$$

Now the ordering of symbols in each ket refer to the states of the first and second particle respectively, and the tensor product is between the system and label spaces. This state is still

¹A combination of bosonic and fermionic operators can describe this situation for two particles [26, 286] but the more complex symmetries possible for three particles are not captured by simple commutation relations [287].

²The symmetrised part of the Hilbert space is denoted $\mathcal{H}_S$, and the system Hilbert space by $\mathcal{H}_S$.

symmetric under particle exchange, but now highlights occupation of $(\mathcal{H}_S)^{\otimes 2} \otimes (\mathcal{H}_L)^{\otimes 2}$ (see Figure 6.1) and allows us to see how the symmetry is distributed across the available degrees of freedom. Here both the system and label states are symmetric under exchange. Furthermore if we were to trace out polarisation, the wavefunction would remain in the symmetric system state because there are no correlations between the degrees of freedom.

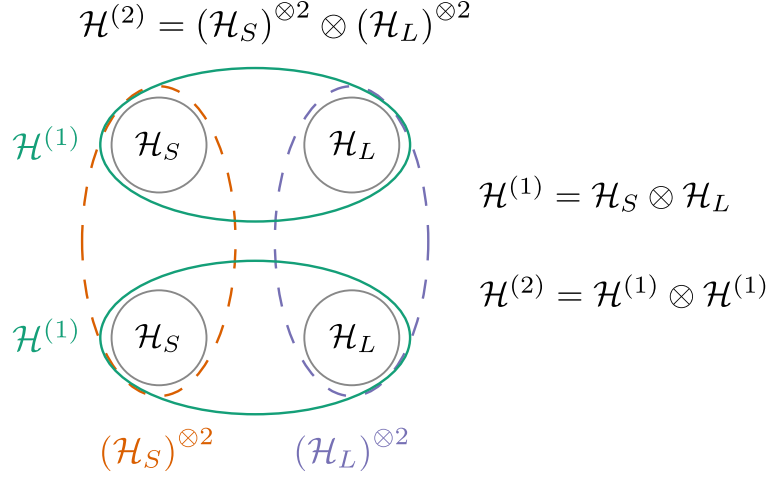


Figure 6.1: The single-particle Hilbert space is given by a tensor product of the spaces for each degree of freedom: $\mathcal{H}^{(1)} = \mathcal{H}_S \otimes \mathcal{H}_L$. The total two-particle space is $\mathcal{H}^{(2)} = \mathcal{H}^{(1)} \otimes \mathcal{H}^{(1)}$. The states in equations 6.2 and 6.4 are written to make this decomposition obvious. $\mathcal{H}^{(2)}$ may also be written as the tensor product of two-particle spaces for each degree of freedom separately: $\mathcal{H}^{(2)} = (\mathcal{H}_S)^{\otimes 2} \otimes (\mathcal{H}_L)^{\otimes 2}$. The states in equations 6.3 and 6.5 are expressed in this way.

What happens for distinguishable polarisations? Take the new input state

$$\begin{aligned} |\psi_{12}^{HV}\rangle &= |1, H\rangle \vee |2, V\rangle \\ &= \frac{1}{\sqrt{2}} (|1, H\rangle \otimes |2, V\rangle + |2, V\rangle \otimes |1, H\rangle). \end{aligned} \quad (6.4)$$

Once again the particle exchange symmetry is clear. For convenience we define the two-particle states in a single degree of freedom $|\psi_{ab}^{\pm}\rangle = (|a, b\rangle \pm |b, a\rangle)/\sqrt{2}$, where ‘+’ corresponds to the symmetric state of particles in $|a\rangle$ and $|b\rangle$, and ‘-’ to the anti-symmetric state. As before we reorganise to separate the port and polarisation (Figure 6.1):

$$|\psi_{12}^{HV}\rangle = \frac{1}{\sqrt{2}} (|\psi_{12}^{+}\rangle \otimes |\psi_{HV}^{+}\rangle + |\psi_{12}^{-}\rangle \otimes |\psi_{HV}^{-}\rangle). \quad (6.5)$$

There are now components in the *anti*-symmetric parts of $(\mathcal{H}_S)^{\otimes 2}$ and $(\mathcal{H}_L)^{\otimes 2}$. Overall the state is still symmetric under exchange, but the introduction of polarisation distinguishability means correlations between degrees of freedom [288]. This causes the state to extend outside the symmetric parts of constituent spaces. Tracing out the polarisation would now leave the system in a maximally mixed state over the symmetric and anti-symmetric system states.

In this chapter we will extend these discussions to more particles and delve into the symmetry structure of the Hilbert spaces for constituent degrees of freedom. Deriving expressions for interference statistics in this framework will shed new light on the role of distinguishability.

6.2 Capturing the symmetries of states and transformations

6.2.1 Representation theory and the symmetric group

In order to build the exchange symmetric wavefunctions corresponding to Fock states, we begin by applying representation theory to the symmetric group. A *representation* R is a mapping of a group G to linear transformations on a vector space V :

$$R : G \rightarrow GL(V). \quad (6.6)$$

Each element g in the group is mapped to an invertible matrix W , the identity element e maps to the identity matrix, and combinations of group actions are preserved:

$$\begin{aligned} g &\mapsto W(g), \\ e &\mapsto W(e) = \mathbb{I}, \\ W(g_1 \cdot g_2) &= W(g_1)W(g_2). \end{aligned} \quad (6.7)$$

We want to define a representation that captures the exchange symmetry of multiparticle wavefunctions. In quantum mechanics the vector space V is the multiparticle Hilbert space $\mathcal{H}^{(N)}$ and G is the symmetric group S_N . Permuting a pair of particles in some wavefunction is described by the action

$$\hat{P}_{i,j}(|\phi_1\rangle \otimes \dots \otimes |\phi_i\rangle \otimes \dots \otimes |\phi_j\rangle \otimes \dots \otimes |\phi_N\rangle) = (|\phi_1\rangle \otimes \dots \otimes |\phi_j\rangle \otimes \dots \otimes |\phi_i\rangle \otimes \dots \otimes |\phi_N\rangle). \quad (6.8)$$

Casting this wavefunction as a vector means the permutation can be written as a matrix and satisfies $P_{i,j}^2 = \mathbb{I}$. Any permutation of particles $\hat{P}_\sigma$ can be decomposed into products of these pairwise transpositions. This permits definition of a representation of S_N on the N -particle wavefunctions in $\mathcal{H}^{(N)}$. Recall that bosonic and fermionic wavefunctions satisfy respectively

$$\hat{P}_{i,j}|\Psi_S\rangle = |\Psi_S\rangle, \quad \hat{P}_{i,j}|\Psi_A\rangle = -|\Psi_A\rangle. \quad (6.9)$$

These define *irreps* of S_N : subspaces of the vector space that transform amongst themselves under action of any element $\sigma \in S_N$. The representation where there are no non-trivial invariant subspaces is the *irreducible representation*. The action of any $\sigma \in S_N$ is then described by a matrix that is block-diagonal, and the irreps correspond to separate blocks (see Figure 6.2). For example the two particle state $|\psi_{12}^+\rangle = |1\rangle \vee |2\rangle$ defines a one-dimensional irrep of S_2 because any $\sigma \in S_2$ leaves the state unchanged.

In Section 4.1.2 we saw that elements of S_N can be written as cycles. Elements with the same cycle structure belong to the same conjugacy class. For example $\sigma = (1, 2, 3)(4, 5)$ and $\sigma = (1, 3, 4)(2, 5)$ fall into the same conjugacy class of S_5 . A key theorem of representation theory is that the number of conjugacy classes determines the number of inequivalent irreps [12]. The irreps of S_N can be labelled by regular partitions λ of the positive integer N , denoted $\lambda \vdash N$.

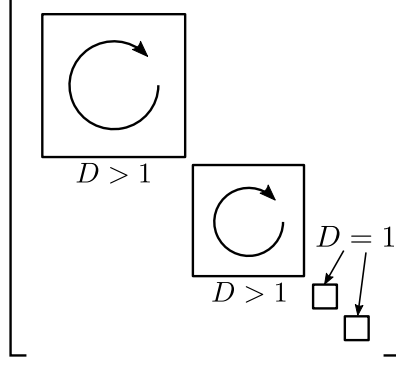


Figure 6.2: In the irreducible representation of a group G , all elements g assume block-diagonal form when mapped to matrices. The separate blocks represent subspaces of the vector space that transform onto themselves under G and are called irreps. Their dimensions D are labelled.

These are lists of positive integers $\lambda = (\lambda_1, \lambda_2, \dots, \lambda_N)$ that satisfy

$$\lambda_1 \geq \lambda_2 \geq \dots \geq \lambda_N \geq 0, \quad \sum_{i=1}^N \lambda_i = N. \quad (6.10)$$

Any $\lambda_i = 0$ are usually omitted when writing down the list. We can illustrate the correspondence between conjugacy classes and irreps using S_2 as an example. It has two conjugacy classes: the identity and the transpositions. These are associated respectively with $\lambda = (2)$ and $\lambda = (1, 1)$. The first labels the symmetric irrep that contains $|\psi_{12}^+\rangle$, and the second labels the anti-symmetric irrep that contains $|\psi_{12}^-\rangle$.

6.2.2 Young diagrams and tableaux

This association of conjugacy classes to regular partitions permits a graphical description of the irreps of S_N along with their symmetries called *Young diagrams* [289]. These comprise a collection of left-justified boxes arranged so that there are λ_i boxes in row i . The total number of boxes equals the number of objects and there is a one-to-one correspondence between Young diagrams and irreps of S_N . For $N = 2$, the two possible partitions $\lambda = (2), (1, 1)$ are represented respectively by

$$\begin{array}{|c|c|} \hline \square & \square \\ \hline \end{array} \quad \begin{array}{|c|} \hline \square \\ \hline \square \\ \hline \end{array} \quad (6.11)$$

In our discussions each box corresponds to a particle. Particles in the same row are exchange symmetric, so the first diagram corresponds to the symmetric irrep for two particles. On the other hand, particles in the same column are exchange anti-symmetric, meaning the second diagram labels the anti-symmetric irrep for two particles.

Assume the particles have a degree of freedom described by a d -dimensional Hilbert space that is spanned by orthogonal vectors numbered $1, \dots, d$. It is possible to number the boxes in Young diagrams in a way that specifies the vectors in $\mathcal{H}^{(N)}$ that span each irrep of S_N . This defines *Young tableaux* and there are two rules specifying how to label the boxes to give the ‘standard’ tableaux: the numbers across a row may not decrease and the numbers down a column must increase. The first rule prevents over-counting states and the second prevents anti-symmetrisation of N particles in a space with dimension $d < N$; for example three qubits

cannot be anti-symmetrised. For a Hilbert space with dimension $d = 2$, there are three valid fillings for the first Young diagram:

$$\begin{array}{|c|c|} \hline 1 & 1 \\ \hline \end{array} \quad \begin{array}{|c|c|} \hline 1 & 2 \\ \hline \end{array} \quad \begin{array}{|c|c|} \hline 2 & 2 \\ \hline \end{array} \quad (6.12)$$

The associated vectors are:

$$\begin{aligned} |\begin{array}{|c|c|} \hline 1 & 1 \\ \hline \end{array}\rangle &= |1, 1\rangle, \\ |\begin{array}{|c|c|} \hline 1 & 2 \\ \hline \end{array}\rangle &= |1\rangle \vee |2\rangle = \frac{1}{\sqrt{2}} (|1, 2\rangle + |2, 1\rangle), \\ |\begin{array}{|c|c|} \hline 2 & 2 \\ \hline \end{array}\rangle &= |2, 2\rangle. \end{aligned} \quad (6.13)$$

These three wavefunctions (the spin triplet) form an orthogonal basis for the symmetric part of the two-particle Hilbert space. There is only a single valid filling for the second Young diagram that gives the anti-symmetric wavefunction (spin singlet):

$$\left| \begin{array}{|c|} \hline 1 \\ \hline 2 \\ \hline \end{array} \right\rangle = \frac{1}{\sqrt{2}} (|1, 2\rangle - |2, 1\rangle). \quad (6.14)$$

The action of any element $\sigma \in S_2$ on these states leaves them invariant up to a sign. Hence the elements $\sigma \in S_2$ in this representation are matrices whose only non-zero elements appear on the diagonal. There are three one-dimensional symmetric irreps and a single one-dimensional anti-symmetric irrep.

The Young diagrams labelling irreps of S_3 can be obtained by adding another box to the diagrams in equation 6.11. These correspond to the regular partitions $\lambda = (3), (2, 1), (1, 1, 1)$ of $N = 3$ and are given by:

$$\begin{array}{|c|c|c|} \hline & & \\ \hline \end{array} \quad \begin{array}{|c|c|} \hline & \\ \hline & \\ \hline \end{array} \quad \begin{array}{|c|} \hline \\ \hline \\ \hline \\ \hline \end{array} \quad (6.15)$$

The first diagram is the fully symmetric irrep, and the last is the fully anti-symmetric one. The second ‘triangular’ diagram describes the mixed symmetry irrep. This contains states that are symmetric under exchange of two particles, and anti-symmetric under exchange of the other pair. There are two ways of obtaining this diagram when adding an extra box to the diagrams of equation 6.12, and as a result there is, as we shall see shortly, an additional multiplicity for these irreps.

If we now assume a qutrit space spanned by orthogonal modes labelled 1–3, what do the wavefunctions associated with these irreps look like? The fully symmetric states are straightforward to write down using the symmetrisation procedure of equation 6.1 and are shown in Table 6.1.

These are all one-dimensional symmetric irreps of S_3 because they transform to themselves under any $\sigma \in S_3$. There is a single anti-symmetric state spanning the anti-symmetric irrep, and using equation 2.6 is given by

$$\left| \begin{array}{|c|} \hline 1 \\ \hline 2 \\ \hline 3 \\ \hline \end{array} \right\rangle = \frac{1}{\sqrt{6}} (|1, 2, 3\rangle - |2, 1, 3\rangle + |3, 1, 2\rangle - |1, 3, 2\rangle + |2, 3, 1\rangle - |3, 2, 1\rangle). \quad (6.16)$$

The states spanning the mixed symmetry irreps require a little more work to find, and involve

Table 6.1: States spanning the symmetric irreps for three particles

Young tableau	State vector
$\begin{array}{ c c c }\hline 1 & 1 & 1 \\ \hline\end{array}$	$ 1, 1, 1\rangle$
$\begin{array}{ c c c }\hline 1 & 1 & 2 \\ \hline\end{array}$	$\frac{1}{\sqrt{3}} (1, 1, 2\rangle + 1, 2, 1\rangle + 2, 1, 1\rangle)$
$\begin{array}{ c c c }\hline 1 & 1 & 3 \\ \hline\end{array}$	$\frac{1}{\sqrt{3}} (1, 1, 3\rangle + 1, 3, 1\rangle + 3, 1, 1\rangle)$
$\begin{array}{ c c c }\hline 1 & 2 & 2 \\ \hline\end{array}$	$\frac{1}{\sqrt{3}} (1, 2, 2\rangle + 2, 1, 2\rangle + 2, 2, 1\rangle)$
$\begin{array}{ c c c }\hline 1 & 2 & 3 \\ \hline\end{array}$	$\frac{1}{\sqrt{6}} (1, 2, 3\rangle + 1, 3, 2\rangle + 2, 1, 3\rangle + 2, 3, 1\rangle + 3, 1, 2\rangle + 3, 2, 1\rangle)$
$\begin{array}{ c c c }\hline 1 & 3 & 3 \\ \hline\end{array}$	$\frac{1}{\sqrt{3}} (1, 3, 3\rangle + 3, 1, 3\rangle + 3, 3, 1\rangle)$
$\begin{array}{ c c c }\hline 2 & 2 & 2 \\ \hline\end{array}$	$ 2, 2, 2\rangle$
$\begin{array}{ c c c }\hline 2 & 2 & 3 \\ \hline\end{array}$	$\frac{1}{\sqrt{3}} (2, 2, 3\rangle + 2, 3, 2\rangle + 3, 2, 2\rangle)$
$\begin{array}{ c c c }\hline 2 & 3 & 3 \\ \hline\end{array}$	$\frac{1}{\sqrt{3}} (2, 3, 3\rangle + 3, 2, 3\rangle + 3, 3, 2\rangle)$
$\begin{array}{ c c c }\hline 3 & 3 & 3 \\ \hline\end{array}$	$ 3, 3, 3\rangle$

explicitly symmetrising states with respect to the interchange of a pair of particles and then anti-symmetrising with respect to another [111, 289, 290]. They are given in Table 6.2 and have been orthogonalised. There is now a subscript p called the *outer multiplicity* reflecting the two ways of adding a third box to make the triangular Young diagram. $p = 1$ corresponds to states that are symmetric under exchange of the first and second particles, and $p = 2$ describes states that are anti-symmetric under the same operation. These symmetries are obvious in the states listed, but the symmetries for the second and third particles are obscured due to orthogonalising the vectors. In fact it turns out that, for each filling of a Young diagram, the two outer multiplicity states $\{|\begin{array}{|c|c|c|}\hline & & \\ \hline\end{array}_1\rangle, |\begin{array}{|c|c|c|}\hline & & \\ \hline\end{array}_2\rangle\}$ form an invariant two-dimensional subspace under any $\sigma \in S_3$. For example swapping the second and third particles of a mixed symmetry vector leaves it in the subspace spanned by vectors in the same row of Table 6.2:

$$\hat{P}_{2,3} \left| \begin{array}{|c|c|c|}\hline 1 & 2 & \\ \hline 3 & & \end{array} \right\rangle_1 = \frac{1}{2} \left(- \left| \begin{array}{|c|c|c|}\hline 1 & 2 & \\ \hline 3 & & \end{array} \right\rangle_1 + \sqrt{3} \left| \begin{array}{|c|c|c|}\hline 1 & 2 & \\ \hline 3 & & \end{array} \right\rangle_2 \right). \quad (6.17)$$

This is straightforward to verify for all other $\sigma \in S_3$, and for all $\lambda = (2, 1)$ Young tableaux. There are therefore eight two-dimensional mixed symmetry irreps for S_3 .

For bookkeeping we introduce another label r called the *inner multiplicity* that specifies the two valid ways of filling the mixed symmetry Young diagram with the numbers 1–3 [109]. We associate $r = 1$ with $\begin{array}{|c|c|}\hline 1 & 2 \\ \hline 3 & \end{array}$, and $r = 2$ with $\begin{array}{|c|c|}\hline 1 & 3 \\ \hline 2 & \end{array}$ and omit it if it is unity for all Young tableaux of a given shape. Later we will consider measuring photons across k system modes, so we also define the mode occupation list $\underline{n} = (n_1, n_2, \dots, n_k)$. n_k is the number of photons in the port described by vector k and $\sum_j n_j = N$. For example $\underline{n} = (1, 1, 1)$ corresponds to one photon per port, whilst $\underline{n} = (3, 0, 0)$ means all are in port ‘1’. States can now be specified by $|\lambda, p, \underline{n}, r\rangle$, and a couple of examples are

$$\begin{aligned} |\lambda = (2), p = 1, \underline{n} = (2, 0)\rangle &= \left| \begin{array}{|c|c|}\hline 1 & 1 \\ \hline \end{array} \right\rangle, \\ |\lambda = (2, 1), p = 2, \underline{n} = (1, 1, 1), r = 2\rangle &= \left| \begin{array}{|c|c|}\hline 1 & 3 \\ \hline 2 & \end{array} \right\rangle_2. \end{aligned} \quad (6.18)$$

Given N particles, all the irreps of the symmetric group can be labelled by writing down all

Table 6.2: States spanning the mixed symmetry irreps for three particles

Young tableau	$p = 1 : (12)$ symmetric	$p = 2 : (12)$ anti-symmetric
$\begin{array}{ c c } \hline 1 & 1 \\ \hline 2 & \\ \hline \end{array}_p$	$-\frac{1}{\sqrt{6}} (1, 2, 1\rangle + 2, 1, 1\rangle - 2 1, 1, 2\rangle)$	$\frac{1}{\sqrt{2}} (1, 2, 1\rangle - 2, 1, 1\rangle)$
$\begin{array}{ c c } \hline 1 & 2 \\ \hline 2 & \\ \hline \end{array}_p$	$\frac{1}{\sqrt{6}} (1, 2, 2\rangle + 2, 1, 2\rangle - 2 2, 2, 1\rangle)$	$\frac{1}{\sqrt{2}} (1, 2, 2\rangle - 2, 1, 2\rangle)$
$\begin{array}{ c c } \hline 1 & 1 \\ \hline 3 & \\ \hline \end{array}_p$	$\frac{1}{\sqrt{6}} (1, 3, 1\rangle + 3, 1, 1\rangle - 2 1, 1, 3\rangle)$	$\frac{1}{\sqrt{2}} (1, 3, 1\rangle - 3, 1, 1\rangle)$
$\begin{array}{ c c } \hline 1 & 2 \\ \hline 3 & \\ \hline \end{array}_p$	$-\frac{1}{\sqrt{12}} (2 1, 2, 3\rangle + 2 2, 1, 3\rangle - 3, 2, 1\rangle - 3, 1, 2\rangle - 1, 3, 2\rangle - 2, 3, 1\rangle)$	$-\frac{1}{2} (1, 3, 2\rangle + 2, 3, 1\rangle - 3, 2, 1\rangle - 3, 1, 2\rangle)$
$\begin{array}{ c c } \hline 2 & 2 \\ \hline 3 & \\ \hline \end{array}_p$	$\frac{1}{\sqrt{6}} (3, 2, 2\rangle + 2, 3, 2\rangle - 2 2, 2, 3\rangle)$	$\frac{1}{\sqrt{2}} (3, 2, 2\rangle - 2, 3, 2\rangle)$
$\begin{array}{ c c } \hline 1 & 3 \\ \hline 2 & \\ \hline \end{array}_p$	$\frac{1}{2} (3, 1, 2\rangle - 3, 2, 1\rangle + 1, 3, 2\rangle - 2, 3, 1\rangle)$	$\frac{1}{\sqrt{12}} (2 1, 2, 3\rangle - 2 2, 1, 3\rangle + 3, 2, 1\rangle - 3, 1, 2\rangle + 1, 3, 2\rangle - 2, 3, 1\rangle)$
$\begin{array}{ c c } \hline 1 & 3 \\ \hline 3 & \\ \hline \end{array}_p$	$-\frac{1}{\sqrt{6}} (1, 3, 3\rangle + 3, 1, 3\rangle - 2 3, 3, 1\rangle)$	$-\frac{1}{\sqrt{2}} (1, 3, 3\rangle - 3, 1, 3\rangle)$
$\begin{array}{ c c } \hline 2 & 3 \\ \hline 3 & \\ \hline \end{array}_p$	$-\frac{1}{\sqrt{6}} (2, 3, 3\rangle + 3, 2, 3\rangle - 2 3, 3, 2\rangle)$	$-\frac{1}{\sqrt{2}} (2, 3, 3\rangle - 3, 2, 3\rangle)$

valid Young diagrams with N boxes.

6.2.3 Schur-Weyl duality

N particles each with a d -dimensional qudit define the multiparticle Hilbert space $\mathcal{H}^{(N)} = (\mathbb{C}^d)^{\otimes N}$. The action of $\sigma \in S_N$ on states $|\Psi_D\rangle$ relabels particles' states:

$$\begin{aligned} \hat{P}_\sigma |\Psi_D\rangle &= \hat{P}_\sigma (|\phi_1\rangle \otimes |\phi_2\rangle \otimes \dots \otimes |\phi_{N-1}\rangle \otimes |\phi_N\rangle) \\ &= (|\phi_{\sigma(1)}\rangle \otimes |\phi_{\sigma(2)}\rangle \otimes \dots \otimes |\phi_{\sigma(N-1)}\rangle \otimes |\phi_{\sigma(N)}\rangle). \end{aligned} \quad (6.19)$$

The evolution of states through, for example, an interferometer is usually described by elements of the special unitary group $SU(d)$ of matrices with unit determinant.³ The overall action of a unitary $\hat{U} \in SU(d)$ on the system is described by the N -fold tensor product $\hat{U}^{\otimes N}$:

$$\begin{aligned} \hat{U}^{\otimes N} |\Psi_D\rangle &= \hat{U}^{\otimes N} (|\phi_1\rangle \otimes |\phi_2\rangle \otimes \dots \otimes |\phi_{N-1}\rangle \otimes |\phi_N\rangle) \\ &= (\hat{U} |\phi_1\rangle \otimes \hat{U} |\phi_2\rangle \otimes \dots \otimes \hat{U} |\phi_{N-1}\rangle \otimes \hat{U} |\phi_N\rangle), \end{aligned} \quad (6.20)$$

where each particle evolves independently of the others [291]. A key thing to notice is that these two actions commute and this enforces a deep connection between the irreps for S_N and

³A unitary matrix $U(d)$ describing an interferometer can be written as the product of a diagonal matrix of phase factors and an $SU(d)$ matrix [111]. The phases do not affect interference for separable states so can be discarded.

$SU(d)$. The multiparticle Hilbert space of N d -dimensional states decomposes into irreducible subspaces as [109, 291, 292]

$$\left(\mathbb{C}^d\right)^{\otimes N} \simeq \bigoplus_{\lambda \vdash N} \left(\mathbb{C}^{\{\lambda\}} \otimes \mathbb{C}^{(\lambda)}\right). \quad (6.21)$$

$\mathbb{C}^{\{\lambda\}}$ carries irreps of $SU(d)$ and $\mathbb{C}^{(\lambda)}$ carries irreps of S_N . The symbol $\simeq$ means the two sides of the expression are related by a basis transformation. The dimension of the irrep (λ) determines the number of irreps $\{\lambda\}$ and vice versa [133, 293]. Hence the multiparticle Hilbert space decomposes into a direct sum of tensor products of irreps that determine each other: *Schur-Weyl duality*. This is quite abstract so we will now look at a concrete example.

From the Young tableaux of equations 6.13 and 6.14 we see that the tensor product of a pair of two-dimensional one-particle Hilbert spaces decomposes into a four-dimensional two-particle Hilbert space. This comprises the direct sum of symmetric and anti-symmetric irreps. Therefore a two-particle density matrix can be written in this four-dimensional Hilbert space as:

$$\rho^{(2)} = \begin{bmatrix} * & * & * & \\ * & * & * & \\ * & * & * & \\ & & & * \end{bmatrix} \quad (6.22)$$

where $*$ represents possibly non-zero elements and those left blank are always zero. The 3×3 block corresponds to the $\lambda = (2)$ irrep and the lone element to the orthogonal $\lambda = (1, 1)$ irrep. Now let us consider evolving this state through some $SU(2)$ unitary with elements given by U_{ij} in the single-particle basis. We can transform the unitary to the two-particle basis by calculating the elements of $U^{\otimes 2}$ in the basis of symmetric and anti-symmetric states, and find [288]

$$U^{\otimes 2} \rightarrow \begin{bmatrix} U_{11}^2 & \sqrt{2}U_{11}U_{12} & U_{12}^2 & \\ \sqrt{2}U_{11}U_{21} & \text{perm}U & \sqrt{2}U_{12}U_{22} & \\ U_{21}^2 & \sqrt{2}U_{21}U_{22} & U_{22}^2 & \\ & & & \det U \end{bmatrix}. \quad (6.23)$$

The unitary is in block-diagonal form so in this basis it decomposes into a direct sum over the *same* irreps as those for the symmetric group: $U^{\otimes 2} \simeq U^{\square\square} \oplus U^{\square}$. Its action is to mix states within subspaces of the same symmetry type: the symmetry possessed by some multiparticle state is preserved under unitary evolution, so fermions stay fermionic and bosons stay bosonic [16]. For $SU(2)$ the $\lambda = (2)$ irrep is three-dimensional, and the $\lambda = (1, 1)$ irrep that maps only to itself is one-dimensional. We can therefore associate the numbers and dimensions of the S_2 and $SU(2)$ irreps as shown in Table 6.3.

This is a hallmark of duality: two symmetries determine each other [294]. The irreps under the joint action of S_2 and $SU(2)$ in the decomposition of the multiparticle space can be labelled just by the irreps of S_2 , or using only the irreps of $SU(2)$. Here Schur-Weyl duality implies

$$\left(\mathbb{C}^2\right)^{\otimes 2} \simeq \left(\mathbb{C}^{\{2\}} \otimes \mathbb{C}^{(2)}\right) \oplus \left(\mathbb{C}^{\{1,1\}} \otimes \mathbb{C}^{(1,1)}\right). \quad (6.24)$$

Table 6.3: Numbers and dimensions of irreps for S_2 and $SU(2)$

Young tableau	S_2 irreps		$SU(2)$ irreps	
	Number	Dimension	Number	Dimension
$\square\square$	3	1	1	3
$\begin{smallmatrix} \square \\ \square \end{smallmatrix}$	1	1	1	1

Young diagrams are good labels for the irreps of both groups so we may write succinctly $\mathcal{H}^{(2)} = \mathcal{H}^{\square\square} \oplus \mathcal{H}^{\begin{smallmatrix} \square \\ \square \end{smallmatrix}}$ [111, 285, 295]. The procedure of combining boxes to make valid Young diagrams, and then filling them appropriately to find the states spanning the irreps of S_N automatically gives states that, under $SU(d)$, transform amongst subspaces of a given symmetry.

We will be revisiting the interference of three photons in a tritter, which is an $SU(3)$ transformation. Therefore the S_3 irrep states given in Tables 6.1 and 6.2, and equation 6.16 can be used as a basis to put $U \in SU(3)$ in block-diagonal form:

$$U^{\otimes 3} \simeq U^{\square\square\square} \oplus U^{\begin{smallmatrix} \square\square \\ 1 \end{smallmatrix}} \oplus U^{\begin{smallmatrix} \square \\ \square\square \end{smallmatrix}} \oplus U^{\begin{smallmatrix} \square \\ \square \\ \square \end{smallmatrix}}. \quad (6.25)$$

States with different p are not mixed by unitary evolution but, as we saw in Table 6.2 and equation 6.17, they can be mixed on action of some $\sigma \in S_3$. This can be summarised as: $SU(d)$ acts within irrep spaces, whilst S_N acts across p multiplicities [296]. A comparison of these irreps' properties is given in Table 6.4, again exhibiting their duality [290].

Table 6.4: Numbers and dimensions of irreps for S_3 and $SU(3)$

Young tableau	S_3 irreps		$SU(3)$ irreps	
	Number	Dimension	Number	Dimension
$\square\square\square$	10	1	1	10
$\begin{smallmatrix} \square\square \\ \square \end{smallmatrix}$	8	2	2	8
$\begin{smallmatrix} \square \\ \square \\ \square \end{smallmatrix}$	1	1	1	1

6.2.4 Unitary-unitary duality

The label degree of freedom must be included to study the effects of partial distinguishability. We now want to construct totally symmetric states that can be evolved using unitaries on the system or label spaces, respectively $SU(d_S)$ and $SU(d_L)$ where d_i is the dimension of the relevant degree of freedom. Given the total multiparticle Hilbert space $\mathcal{H}_{SL}^{(N)}$, and that each degree of freedom is acted on by S_N , Schur-Weyl duality permits separate decomposition of the

spaces for each degree of freedom [109, 296]:

$$\begin{aligned}\mathcal{H}_{SL}^{(N)} &= (\mathcal{H}_S)^{\otimes N} \otimes (\mathcal{H}_L)^{\otimes N} \\ &= \left(\bigoplus_{\lambda} \mathcal{H}_S^{\lambda} \right) \otimes \left(\bigoplus_{\lambda'} \mathcal{H}_L^{\lambda'} \right).\end{aligned}\tag{6.26}$$

$\mathcal{H}_i^{\lambda}$ is formed from the tensor product of irreps for $SU(d_i)$ and irreps of S_N that are labelled by the same $\lambda \vdash N$ (equation 6.21). We are after the symmetrised subspace of this larger space, where the states over both degrees of freedom satisfy bosonic symmetry. It turns out that this problem of coupling irreps for the two unitary groups leads to requiring $\lambda = \lambda'$ and $p = p'$ [109, 288, 293, 296]. Thus the totally symmetric states in $\mathcal{H}_{SL}^{(N)}$ are given by [109]

$$\left| \mathfrak{S}_{\underline{n}_S, \underline{n}_L}^{\lambda, r_S, r_L} \right\rangle_{SL} = \frac{1}{\sqrt{D_{(\lambda)}}} \sum_{p=1}^{D_{(\lambda)}} \left| \lambda, p, \underline{n}_S, r_S \right\rangle_S \otimes \left| \lambda, p, \underline{n}_L, r_L \right\rangle_L.\tag{6.27}$$

$\underline{n}_S$ and $\underline{n}_L$ denote occupations of the system and label states respectively, and $D_{(\lambda)}$ is the dimension of the irrep λ in S_N . The overall symmetry requirement means all outer multiplicity states p must have equal weighting in this basis. General symmetric states are now formed by summing over irreps

$$\left| \Psi_{SL}^{sym} \right\rangle = \sum_{\lambda, r_S, r_L} \psi_{\underline{n}_S, \underline{n}_L}^{\lambda, r_S, r_L} \left| \mathfrak{S}_{\underline{n}_S, \underline{n}_L}^{\lambda, r_S, r_L} \right\rangle_{SL}\tag{6.28}$$

where the coefficients are determined by the state being symmetrised, and are independent of p . We saw two examples of this in Section 6.1.2:

$$\begin{aligned}\left| \psi_{12}^{HH} \right\rangle &= |1, H\rangle \vee |2, H\rangle \rightarrow \left| \boxed{12} \right\rangle \otimes \left| \boxed{HH} \right\rangle, \\ \left| \psi_{12}^{HV} \right\rangle &= |1, H\rangle \vee |2, V\rangle \rightarrow \frac{1}{\sqrt{2}} \left(\left| \boxed{12} \right\rangle \otimes \left| \boxed{HV} \right\rangle + \left| \boxed{1} \right\rangle \otimes \left| \boxed{H} \right\rangle \right).\end{aligned}\tag{6.29}$$

The Young tableaux states for polarisation are obtained by substituting H and V for the number labels of the Young tableaux states in equations 6.13 and 6.14. The first state of two photons indistinguishable in polarisation only accesses the symmetric irreps of the $SU(2)$ representations for system and label with $\lambda = (2)$. The polarisation distinguishability in the second enables access to the $\lambda = (1, 1)$ anti-symmetric irreps for both spaces.

This process amounts to a generalisation of what was shown in Figure 6.1: the symmetrised tensor product of single particle spaces can be decomposed into a direct sum of products of states that lie in the invariant subspaces of $\mathcal{H}_S^{(N)}$ and $\mathcal{H}_L^{(N)}$. This also permits a definition of indistinguishability in terms of subspace symmetry [296]. Consider casting some pure state in the joint symmetrised basis of the d^N -dimensional multiparticle Hilbert space. If the label state is the same for all N particles then they are in principle indistinguishable in that degree of freedom, and the associated symmetric irrep $\lambda = (N)$ for $SU(d_L)$ is one-dimensional. Hence from the relation in equation 6.28, the label state is coupled to a single symmetric system state. The joint state is thus separable in system and label, so tracing out the latter does not affect the form of the system state. This is evident in $\left| \psi_{12}^{HH} \right\rangle$ above where the photons are indistinguishable in their polarisation and the overall state is separable in the two degrees of

freedom.

On the other hand, distinguishability means there are correlations between the system and label states. Tracing out the latter can leave the system in a mixed state with population outside the symmetric irrep of $SU(d_S)$. For example $|\psi_{12}^{HV}\rangle$ occupies both $\lambda = (2)$ and $\lambda = (1, 1)$ irreps. If one does not have access to polarisation, the correlations between system and label and the presence of non-symmetric components in the system space may be used to identify distinguishability in label [109, 296, 297]. Indeed this was instrumental in the discovery of the colour degree of freedom for quarks. The existence of baryons such as Δ^{++} with symmetric quark flavour $\boxed{u \ u \ u}$ led to the introduction of the anti-symmetric colour singlet states in order to preserve the overall fermionic symmetry [298, 299].

The action of a joint operation $\hat{U}_{SL} = \hat{U}_S \otimes \hat{U}_L \in SU(d_S \times d_L)$ in the symmetric part of the multiparticle space decomposes as

$$U_{SL}^{\otimes N} \simeq \bigoplus_{\lambda} \left(U_S^{\lambda} \otimes U_L^{\lambda} \right). \quad (6.30)$$

This is *unitary-unitary duality*: the same λ labels the irreps of both unitary groups at the same time, and unitaries act on their respective spaces independently [109, 288, 291, 294, 296]. As in equation 6.25 the unitaries will be block diagonal and so leave a state in the same irrep λ .

6.2.5 Scattering probabilities

We can now investigate the subspaces probed in interference experiments. Suppose we prepare an input state $\hat{\rho}_{SL}^{in}$ that is separable in spatial mode and has input port occupation $\underline{n}'_S$. It resides in the symmetrised part of the joint N -particle Hilbert space. After evolution under a unitary $\hat{U}_{SL} = \hat{U}_S \otimes \hat{U}_L$, we consider counting the output configuration $\underline{n}_S$ in the port degree of freedom. The scattering probability is then

$$P_{\underline{n}_S} = \text{Tr}_{S,L} \left[\left(\hat{U}_{SL}^{\otimes N} \right) \hat{\rho}_{SL}^{in} \left(\hat{U}_{SL}^{\otimes N} \right)^{\dagger} \hat{\Pi}_{SL}^{\underline{n}_S} \right]. \quad (6.31)$$

The operator $\hat{\Pi}_{SL}^{\underline{n}_S}$ projects onto the symmetrised part of the joint Hilbert space with configuration $\underline{n}_S$ in output port, and ignores the label degree of freedom. We are familiar with the requirement that, in order to test partial exchange symmetry, the evolution should not affect the label state – indeed we went to great lengths to guarantee polarisation-independence of the quitter in Section 4.3.3. This means we can identify $\hat{U}_L = \hat{\mathbb{I}}_L$. Given that our detection is insensitive to this degree of freedom, as is the case for APDs with polarisation and temporal modes, then the only non-trivial action of the trace over label in equation 6.31 is on the input state $\hat{\rho}_{SL}^{in}$. An equivalent expression for $P_{\underline{n}_S}$ could be obtained by instead considering the dynamics of the reduced matrix for the system state $\hat{\rho}_S^{in} = \text{Tr}_L(\hat{\rho}_{SL}^{in})$, specifically

$$P_{\underline{n}_S} = \text{Tr}_S \left[\left(\hat{U}_S^{\otimes N} \right) \hat{\rho}_S^{in} \left(\hat{U}_S^{\otimes N} \right)^{\dagger} \hat{\Pi}_S^{\underline{n}_S} \right]. \quad (6.32)$$

$\hat{\Pi}_S^{\underline{n}_S}$ lies outside the joint Hilbert space and so cannot generally be expressed in terms of bosonic operators. The trace is basis-invariant so we can use the jointly symmetrised basis of the

preceding sections. $U_S^{\otimes N}$ then takes the block-diagonal form of equation 6.30 and any evolution of the label state can be ignored. Given that ρ_{SL}^{in} comprises symmetric states in the joint space (equation 6.27) and has occupation $\underline{n}'_S$, the reduced state has the form [109]

$$\rho_S^{in} = \sum_{\lambda, p, r_S, r'_S} \rho_S^{\lambda, p, r_S, r'_S} \left| \lambda, p, \underline{n}'_S, r_S \right\rangle \left\langle \lambda, p, \underline{n}'_S, r'_S \right|. \quad (6.33)$$

The complex coefficients contain information on the effect of tracing over the label states and will be investigated further in the next section. Finally the counting operator $\hat{\Pi}_S^{\underline{n}_S}$ is now given by a projection on all states satisfying occupation $\underline{n}_S$:

$$\Pi_S^{\underline{n}_S} = \sum_{\lambda, p, r_S} \left| \lambda, p, \underline{n}_S, r_S \right\rangle \left\langle \lambda, p, \underline{n}_S, r_S \right|. \quad (6.34)$$

It includes population in non-symmetric irreps that can give rise to a successful detection of counting pattern $\underline{n}_S$ and is diagonal assuming an orthogonal basis [109]. Putting this all together, the output probability is

$$\begin{aligned} P_{\underline{n}_S} &= \text{Tr}_S \left[\left(\bigoplus_{\mu} U_S^{\mu} \right) \rho_S^{in} \left(\bigoplus_{\mu'} U_S^{\mu'} \right)^{\dagger} \Pi_S^{\underline{n}_S} \right] \\ &= \sum_{\substack{\lambda, p, r_S \\ r'_S, r''_S}} \rho_S^{\lambda, p, r_S, r'_S} \left\langle \lambda, p, \underline{n}_S, r''_S \right| U_S^{\lambda} \left| \lambda, p, \underline{n}'_S, r_S \right\rangle \left\langle \lambda, p, \underline{n}'_S, r'_S \right| \left(U_S^{\lambda} \right)^{\dagger} \left| \lambda, p, \underline{n}_S, r''_S \right\rangle. \end{aligned} \quad (6.35)$$

The block-diagonal form of the input state and the unitary evolution significantly simplify index summations. The contributions from the unitary reduce to permanents for $\lambda = (N)$ and determinants for $\lambda = (1, \dots, 1)$. Elements of the unitary transformation in the mixed symmetry subspaces may be calculated explicitly using expressions for the basis states (for example those in Table 6.2), or using Wigner D functions [111, 285, 300]. This expression may be used to determine which interferometers are optimal for discriminating fully indistinguishable and singly distinguishable collections of interfering photons [109]. We will use it to reveal connections between distinguishability and Hilbert space symmetries.

6.3 Multiparticle distinguishability in first quantisation

6.3.1 HOM interference

Consider the state preparation discussed in Section 5.3, namely two photons in qubit states ρ_a and ρ_b that are injected into the two ports of a beam splitter. In first quantised formalism the input state is

$$\rho_{SL}^{(2)} = (|1\rangle \langle 1| \otimes \rho_a) \vee (|2\rangle \langle 2| \otimes \rho_b). \quad (6.36)$$

The first task is to determine the form of the relevant Fock states in terms of the jointly symmetrised states defined in equation 6.27. Constrained to $\underline{n}'_S = \underline{1} = (1, 1)$, these are

$$\begin{aligned} |\mathfrak{S}_{\underline{1}, HH}^{\square\square}\rangle &= |\underline{1}\underline{2}\rangle \otimes |\overline{HH}\rangle, \\ |\mathfrak{S}_{\underline{1}, HV}^{\square\square}\rangle &= |\underline{1}\underline{2}\rangle \otimes |\overline{HV}\rangle, \\ |\mathfrak{S}_{\underline{1}, VV}^{\square\square}\rangle &= |\underline{1}\underline{2}\rangle \otimes |\overline{VV}\rangle, \\ |\mathfrak{S}_{\underline{1}, HV}^{\square}\rangle &= |\underline{1}\rangle \otimes |\overline{H}\rangle, \end{aligned} \quad (6.37)$$

where we have used Young diagrams to label the irrep, $\underline{1}$ to denote one photon in each input port, and have written the polarisation occupations explicitly instead of $\underline{n}_L$. The inner multiplicities are all unity and so are omitted here. Fock states can be expressed in this basis using equation 6.28

$$\begin{aligned} \hat{a}_{1,H}^\dagger \hat{a}_{2,H}^\dagger |0\rangle &= |1, H\rangle \vee |2, H\rangle = |\mathfrak{S}_{\underline{1}, HH}^{\square\square}\rangle, \\ \hat{a}_{1,H}^\dagger \hat{a}_{2,V}^\dagger |0\rangle &= |1, H\rangle \vee |2, V\rangle = \frac{1}{\sqrt{2}} \left(|\mathfrak{S}_{\underline{1}, HV}^{\square\square}\rangle + |\mathfrak{S}_{\underline{1}, HV}^{\square}\rangle \right), \\ \hat{a}_{1,V}^\dagger \hat{a}_{2,H}^\dagger |0\rangle &= |1, V\rangle \vee |2, H\rangle = \frac{1}{\sqrt{2}} \left(|\mathfrak{S}_{\underline{1}, HV}^{\square\square}\rangle - |\mathfrak{S}_{\underline{1}, HV}^{\square}\rangle \right), \\ \hat{a}_{1,V}^\dagger \hat{a}_{2,V}^\dagger |0\rangle &= |1, V\rangle \vee |2, V\rangle = |\mathfrak{S}_{\underline{1}, VV}^{\square\square}\rangle. \end{aligned} \quad (6.38)$$

These can be substituted into the expression for $\rho_S^{(2)}$. The reduced state $\rho_S^{(2)} = \text{Tr}_L(\rho_{SL}^{(2)})$ contains all the effects of the label degree of freedom on the partial exchange symmetry of the input state. Ordering the system basis as $\left\{ \underline{1}\underline{1}, \underline{1}\underline{2}, \underline{2}\underline{2}, \underline{1}\underline{2} \right\}$, it is given by (see Figure 6.3)

$$\rho_S^{(2)} = \frac{1}{4} \begin{bmatrix} 0 & 0 & 0 & \\ 0 & 3 + \mathbf{r}_a \cdot \mathbf{r}_b & 0 & \\ 0 & 0 & 0 & \\ & & & 1 - \mathbf{r}_a \cdot \mathbf{r}_b \end{bmatrix} \quad (6.39)$$

where $\mathbf{r}_i$ are the relevant Bloch vectors. This corresponds to population in the symmetric $|\underline{1}\underline{2}\rangle \langle \underline{1}\underline{2}|$ and anti-symmetric $|\underline{1}\underline{2}\rangle \langle \underline{2}\underline{1}|$ system states. Anything other than indistinguishability of the polarisation acts to leak population from the symmetric to the antisymmetric subspace: distinguishability and mixedness act in precisely the same way.

We earlier expressed the system unitary $U_S^{\otimes 2}$ in this basis (equation 6.23) where it takes block-diagonal form. Hence population in the symmetric state stays in the symmetric irrep, and likewise for the anti-symmetric state. Finally to calculate the coincidence probability, we construct the counting operator that is sensitive to any population with particles in both ports [109, 288]:

$$\Pi_S^1 = \begin{bmatrix} 0 & 0 & 0 & \\ 0 & 1 & 0 & \\ 0 & 0 & 0 & \\ & & & 1 \end{bmatrix}. \quad (6.40)$$

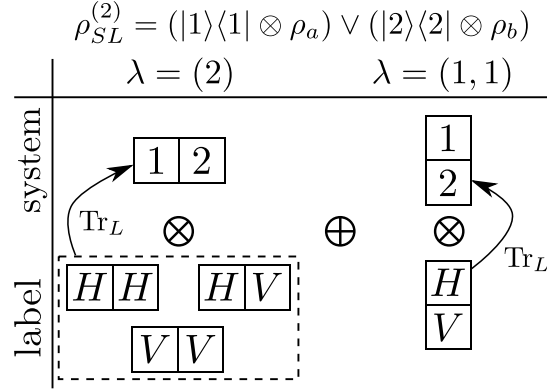


Figure 6.3: The jointly symmetrised basis for two photons with spatial and polarisation degrees of freedom (equation 6.36). States of the same symmetry, and so lying in the same irrep λ , are paired together to give overall symmetric states. On tracing out the polarisation degree of freedom (labelled Tr_L), the reduced system state is a mixture of symmetric and antisymmetric spatial states.

There are contributions from the symmetric and anti-symmetric subspaces. Putting all this together in equation 6.35 yields

$$P_{\perp}^{(2)} = \text{Tr}_S \left[\left(U_S^{\square\square} \oplus U_S^{\square\Box} \right) \rho_S^{(2)} \left(U_S^{\square\square} \oplus U_S^{\square\Box} \right)^\dagger \Pi_S^1 \right] \quad (6.41)$$

$$= \frac{1}{4} \left[(3 + \mathbf{r}_a \cdot \mathbf{r}_b) \times |\text{perm}(U_S)|^2 + (1 - \mathbf{r}_a \cdot \mathbf{r}_b) \times |\det(U_S)|^2 \right].$$

The permanent is associated with population in the symmetric irrep, and the determinant with the anti-symmetric subspace. For $\mathbf{r}_a \cdot \mathbf{r}_b = 1$ we recover $P_{\perp} = |\text{perm}(U_S)|^2$ that, for a balanced beam splitter, is zero and corresponds to HOM bunching. Distinguishable polarisation states mean $\mathbf{r}_a \cdot \mathbf{r}_b = -1$ and then $P_{\perp} = \frac{1}{2} \left(|\text{perm}(U_S)|^2 + |\det(U_S)|^2 \right) = \text{perm}|U_S|^2$ which is 1/2 for a balanced splitter. Reduction in the pairwise overlap for two particles thus decreases the symmetric component of the reduced system state. If $\mathbf{r}_a \cdot \mathbf{r}_b \neq 1$ then anti-symmetric components provide distinguishing information on the system and so lower interference visibility. Here it is clear that, even if the detection cannot resolve the label degree of freedom, the correlations between label and port affect symmetry subspace populations, and thus the counting statistics at the output ports. It is worth pointing out that the bunching probability corresponds to measuring population in, for example, the state $|\underline{11}\rangle \langle \underline{11}|$. Such statistics only probe the symmetric irrep and so are unsuitable for tasks diagnosing reduction of interference strength due to full distinguishability [109].

6.3.2 Mixed symmetry subspaces and the triad phase

Three particles are needed in order to access the mixed symmetry subspaces. Extending to a three-mode interferometer and injecting a third qubit ρ_c into the third input, we now turn to

$$\rho_{SL}^{(3)} = (|1\rangle \langle 1| \otimes \rho_a) \vee (|2\rangle \langle 2| \otimes \rho_b) \vee (|3\rangle \langle 3| \otimes \rho_c). \quad (6.42)$$

The Schur-Weyl basis states for $SU(3)$ were given in Section 6.2.3 and those with one particle per port ($\underline{n}'_S = (1, 1, 1)$) will describe the system degree of freedom. The polarisation states for three particles, by unitary-unitary duality, decompose in the same way as the system so

$$\mathcal{H}^\square \otimes \mathcal{H}^\square \otimes \mathcal{H}^\square = \mathcal{H}^{\square\square\square} \oplus \mathcal{H}^{\square\square}_1 \oplus \mathcal{H}^{\square\square}_2 \oplus \mathcal{H}^{\square}, \quad (6.43)$$

with the restriction that the anti-symmetric space is empty: it is impossible to anti-symmetrise three qubits. The basis states spanning these irreps can be obtained by taking the states in Tables 6.1 and 6.2 with the modes ‘1’ and ‘2’, and relabelling with H and V respectively. These mixed symmetry label states are inner multiplicity free because there is only one valid filling for each occupation $\underline{n}_L$. Therefore here we denote the inner multiplicity of the system states by r instead of r_S , and omit it if it is unity. The symmetrised states of equation 6.27 are here given by

$$\begin{aligned} |\mathfrak{S}_{1,HHH}^{\square\square\square}\rangle &= |\underline{123}\rangle \otimes |\underline{HHH}\rangle, \\ |\mathfrak{S}_{1,HHV}^{\square\square}\rangle &= |\underline{123}\rangle \otimes |\underline{HHV}\rangle, \\ |\mathfrak{S}_{1,HHV}^{\square\square,r=1}\rangle &= \frac{1}{\sqrt{2}} \left(|\underline{123}\rangle_1 \otimes |\underline{HH}\rangle_1 + |\underline{123}\rangle_2 \otimes |\underline{HH}\rangle_2 \right), \\ |\mathfrak{S}_{1,HHV}^{\square\square,r=2}\rangle &= \frac{1}{\sqrt{2}} \left(|\underline{123}\rangle_1 \otimes |\underline{HH}\rangle_1 + |\underline{123}\rangle_2 \otimes |\underline{HH}\rangle_2 \right), \\ |\mathfrak{S}_{1,HVV}^{\square\square}\rangle &= |\underline{123}\rangle \otimes |\underline{HVV}\rangle, \\ |\mathfrak{S}_{1,HVV}^{\square\square,r=1}\rangle &= \frac{1}{\sqrt{2}} \left(|\underline{123}\rangle_1 \otimes |\underline{HV}\rangle_1 + |\underline{123}\rangle_2 \otimes |\underline{HV}\rangle_2 \right), \\ |\mathfrak{S}_{1,HVV}^{\square\square,r=2}\rangle &= \frac{1}{\sqrt{2}} \left(|\underline{123}\rangle_1 \otimes |\underline{HV}\rangle_1 + |\underline{123}\rangle_2 \otimes |\underline{HV}\rangle_2 \right), \\ |\mathfrak{S}_{1,VVV}^{\square\square}\rangle &= |\underline{123}\rangle \otimes |\underline{VVV}\rangle. \end{aligned} \quad (6.44)$$

These can be used as a basis for the Fock states in $\rho_{SL}^{(3)}$. The states corresponding to all H or all V polarisation are given by the first and last states in the list above. The others have components in the mixed symmetry spaces, for example

$$|1, H\rangle \vee |2, V\rangle \vee |3, H\rangle = \frac{1}{\sqrt{6}} \left(\sqrt{2} |\mathfrak{S}_{1,HHV}^{\square\square}\rangle + |\mathfrak{S}_{1,HHV}^{\square\square,r=1}\rangle + \sqrt{3} |\mathfrak{S}_{1,HHV}^{\square\square,r=2}\rangle \right). \quad (6.45)$$

Constructing expressions for the other Fock states yields $\rho_{SL}^{(3)}$ in the jointly symmetrised basis. Tracing out the label degree of freedom produces the system state

$$\rho_S^{(3)} = \frac{1}{8} \begin{bmatrix} \alpha & & & & \\ & \beta & \gamma & & \\ & \gamma^* & \delta & & \\ & & & \beta & \gamma \\ & & & \gamma^* & \delta \end{bmatrix}, \quad \begin{aligned} \alpha &= 4 + \frac{4}{3} (\mathbf{r}_a \cdot \mathbf{r}_b + \mathbf{r}_a \cdot \mathbf{r}_c + \mathbf{r}_b \cdot \mathbf{r}_c), \\ \beta &= 1 - \frac{2}{3} (\mathbf{r}_a \cdot \mathbf{r}_c + \mathbf{r}_b \cdot \mathbf{r}_c - \frac{1}{2} \mathbf{r}_a \cdot \mathbf{r}_b), \\ \gamma &= \frac{1}{\sqrt{3}} (\mathbf{r}_a \cdot \mathbf{r}_c - \mathbf{r}_b \cdot \mathbf{r}_c + i\tilde{V}_{abc}), \\ \delta &= 1 - \mathbf{r}_a \cdot \mathbf{r}_b, \end{aligned} \quad (6.46)$$

where the bases are ordered as $\left\{ \underline{123}, \underline{12}_1, \underline{12}_2, \underline{13}_1, \underline{13}_2 \right\}$. The coefficients in the matrix are the same for the outer multiplicities $p = 1, 2$. There are two things to note: firstly in

contrast to the two particle case, there are now coherences induced by the trace over label modes; secondly the coherences have a real part given by pairwise dot products, and an imaginary part given precisely by the quantity $\tilde{V}_{abc}$ of equation 5.16. The three-particle distinguishability is contained entirely in the imaginary coherences (see Figure 6.4). In the previous chapter we presented sets of pure distinguishable and identical mixed states that had the same $\text{Tr}(\rho_i \rho_j)$ but gave different three-photon scattering statistics (Figure 5.5). Whilst all Bloch vector dot products are identical, the only difference between the situations is the presence of the imaginary coherence for pure states.

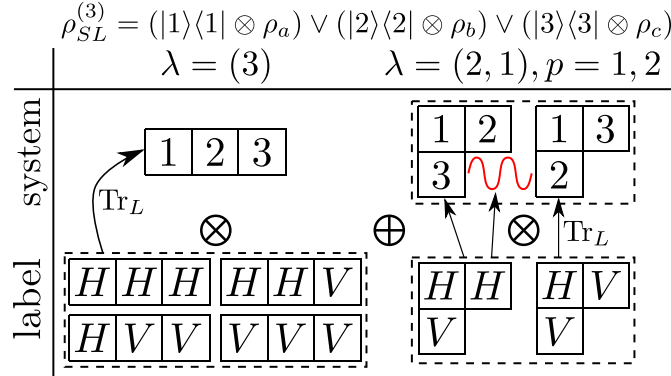


Figure 6.4: The jointly symmetrised basis for three photons with spatial and polarisation degrees of freedom. On tracing out the polarisation, the reduced system state has population in the symmetric and mixed symmetry coincident subspaces. A new feature is the development of coherences between the mixed symmetry states of different inner multiplicity (red wavy line). The same process occurs for both outer multiplicities p .

This input reduced system state resides in the coincident subspace of the system Hilbert space (Figure 6.5a). The unitary causes the state to evolve outside the coincident subspace, but the total population within a given irrep is constant throughout due to the block-diagonal form of $U_S^{\otimes 3}$ in this basis (Figure 6.5b). Ignoring any contribution from the unpopulated anti-symmetric irrep, the probability of detecting photons with output occupation $\underline{n}_S$ is then

$$P_{\underline{n}_S}^{(3)} = \text{Tr}_S \left[\left(U^{\square\square\square} \oplus U^{\square\square_1} \oplus U^{\square\square_2} \right) \rho_S^{(3)} \left(U^{\square\square\square} \oplus U^{\square\square_1} \oplus U^{\square\square_2} \right)^\dagger \Pi_S^{\underline{n}_S} \right]. \quad (6.47)$$

It can be shown that the elements $U^{\square\square_1}$ are identical to those of $U^{\square\square_2}$, so the $p = 1$ and $p = 2$ components of $\rho_S^{(3)}$ contribute in precisely the same way. Explicit calculation of these probabilities using the appropriate matrix elements and counting operators recovers the output probabilities for three photons with qubit states in the previous chapter, in equation 5.20. Bunched statistics are not sensitive to $\tilde{V}_{abc}$ because they only probe the symmetric irrep containing dot products $\mathbf{r}_i \cdot \mathbf{r}_j$. Whilst the operator for coincident detection does probe the mixed symmetry irreps $\lambda = (2, 1)$, the products of matrix elements that show up in equation 6.35 are real thanks to a constraint for 3×3 unitaries. Just as we saw in Section 5.4.1 this leads to an insensitivity to $\tilde{V}_{abc}$. However partially bunched statistics that count population in the mixed symmetry irreps (orange elements in Figure 6.5) are sensitive to this quantity.

When interfering three particles in qubit states, the pairwise distinguishabilities sift popula-

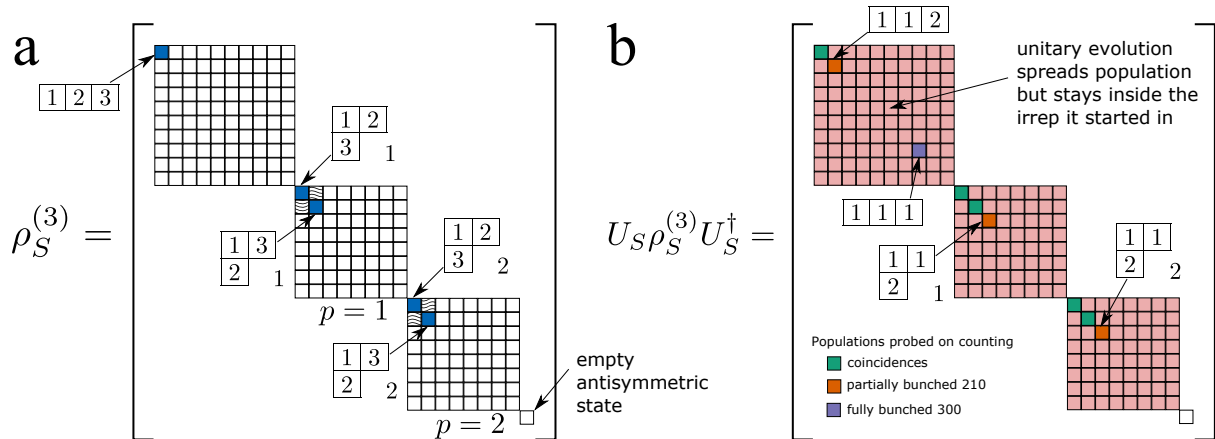


Figure 6.5: **a** The reduced density matrix $\rho_S^{(3)}$ occupies parts of the system Hilbert space with one particle per port (equation 6.46) and the $SU(3)$ irreps are listed in Table 6.4. There are populations in the symmetric and mixed symmetry irreps (blue) as well as coherences between the two inner multiplicity states in the mixed symmetry irreps (wavy lines). The anti-symmetric irrep is empty by unitary-unitary duality and the fact that three qubits cannot be anti-symmetrised. **b** Evolution under U_S spreads population across the irrep it starts in (pink). Counting photons in output configuration $\underline{n}_S$ corresponds to probing populations in the coloured boxes as shown.

tion out of the symmetric irrep for the reduced states and can generate real coherences between mixed symmetry states. The collective distinguishing parameter $\tilde{V}_{abc}$ shows up as an imaginary coherence in the mixed symmetry subspace. Adding more particles and a larger label space can lead to more of these coherences between subspaces of more complex symmetries, corresponding to the multiparticle distinguishing phases discussed in Chapter 4. While this approach is not an easy way of determining interference statistics for separable states, it does provide insight into how particle symmetry is distributed across the constituent degrees of freedom, as well as the effect of distinguishability across different subspace symmetries. In fact this formalism is quite general, and in the next section we will discuss its application to entangled states.

6.4 Beyond separable states

6.4.1 Interfering entangled states

In the absence of entanglement, separable states have the property that each particle is separately described by a pure quantum state. This has allowed us to develop a measure of distinguishability by appending a degree of freedom in which the photons are orthogonal, and then permuting this label to test the state's partial exchange symmetry. However photons in an entangled state do not have a well-defined state so it is impossible to make a statement about state distinguishability in this way. For example, consider two photons in the polarisation Bell state

$$|\Psi^+\rangle = \frac{1}{\sqrt{2}}(|H_1, V_2\rangle + |V_1, H_2\rangle) \quad (6.48)$$

where the state is cast in second quantised form, and the subscripts denote orthogonal spatial modes. Ignorance of the photon in the second spatial mode leaves the other photon in a

maximally mixed state in polarisation, so it is impossible to assign it a pure quantum state. What happens if we perform a HOM interference test on this state? We can substitute the expressions for Fock states in first quantisation from equation 6.38, and this Bell state then takes the form:

$$|\Psi^+\rangle = |\overline{12}\rangle \otimes |\overline{HV}\rangle. \quad (6.49)$$

The reduced system state is $\rho_S^+ = |\overline{12}\rangle \langle \overline{12}|$ which lies entirely in the symmetric $SU(2)$ irrep. Evolution through a balanced beam splitter, put into block-diagonal form as in equation 6.23 where $\text{perm}(U_S) = 0$, shows that this state will exhibit HOM bunching [286]. The ability to quantify distinguishability in a label degree of freedom breaks down here because it is not possible to treat the particles as each occupying a single pure state. The indistinguishability leading to interference now arises because the state is a superposition of two distinct two-particle states. Despite the photons being distinguishable in either component of the superposition, the overall state is still symmetric under exchange of spatial mode or polarisation. Destructive interference of the paths leading to coincidences then causes the photons to bunch. This has been demonstrated for SPDC emission where the signal and idler are in a spectrally entangled state, and the JSA is symmetric under exchange [301, 302].

In Chapters 3 and 4 we presented preparations of photons in separable states that isolated respectively the three- and four-photon exchange contributions to interference. This meant keeping terms in lower-order distinguishing parameters constant. Free from the constraints imposed by using separable states, there are approaches towards isolating N -particle interference independent of $m < N$ contributions using entangled states and so-called N -particle GHZ interferometers [247, 248]. The three-particle incarnation is shown in Figure 6.6.

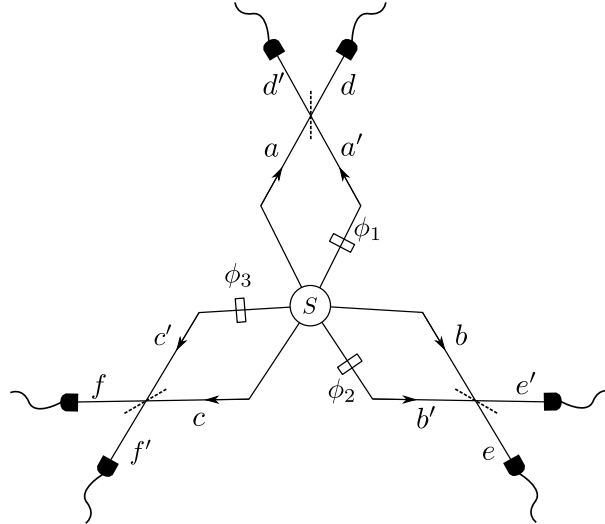


Figure 6.6: A source S generates three photons in an entangled state over paths a, b, c and a', b', c' (the state $|GHZ_3\rangle$ in the main text). Phase shifts are applied to the interferometer arms labelled with primes, and balanced beam splitters mix the pairs of paths as shown. This erases which-path information and, along with the coherence of the generated state, permits phase-sensitive interference for coincident detection (equation 6.50). Adapted from [248].

A source generates the entangled state $|GHZ_3\rangle = \frac{1}{\sqrt{2}}(|a, b, c\rangle + |a', b', c'\rangle)$ and phase shifts are applied to the primed paths. Pairs of paths for each of the three particles are overlapped on

balanced beam splitters as shown, and phases ϕ_i are applied to a subset of the paths. Subsequent detection of three photons at the outputs yields the probabilities

$$\begin{aligned} P_{\text{even}} &= \frac{1}{8} (1 + \sin(\phi_1 + \phi_2 + \phi_3)), \\ P_{\text{odd}} &= \frac{1}{8} (1 - \sin(\phi_1 + \phi_2 + \phi_3)), \end{aligned} \quad (6.50)$$

where P_{even} applies for coincidences with an even number of primed outputs, and P_{odd} for odd. Fringes will be observed whilst varying the phases ϕ_i and indicate three-photon interference. At the same time, it can be shown that two-photon coincidences remain flat and there is no two-photon interference [247]. This setup has been realised using polarisation GHZ states to contradict local realism in quantum mechanics [303]. More recently the approach was used to demonstrate another type of three-photon interference using energy-time entangled photon triplets and three spatially separated Franson interferometers that erase information of emission times [91, 249]. High visibility fringes in threefold coincidences are observed as phases within the Franson interferometers are varied and, unlike in our demonstration in Chapter 3, the spectral distinguishability of three photons has no impact on this type of interference. Extensions to more particles are possible by increasing the number of particles in the GHZ state and adding more arms to the interferometer in Figure 6.6.

6.4.2 Simulating exotic particle statistics

We saw that pairwise distinguishability means the reduced state of two particles occupies both the symmetric *and* anti-symmetric irreps for the system degree of freedom (Figure 6.3). Entanglement permits cleaner access to these subspaces. For example consider the two-particle state

$$\begin{aligned} |\psi_{HV}^\theta\rangle &= \frac{1}{\sqrt{2}} (|H_1, V_2\rangle + e^{i\theta} |V_1, H_2\rangle) \\ &\equiv \cos \frac{\theta}{2} |\overline{11}\overline{2}\rangle \otimes |\overline{H}\overline{V}\rangle - i \sin \frac{\theta}{2} \left| \frac{1}{2} \right\rangle \otimes \left| \frac{H}{V} \right\rangle. \end{aligned} \quad (6.51)$$

We have used the expressions in equation 6.38 that cast Fock states in the symmetrised states defined using unitary-unitary duality. The reduced state of the system is obtained by tracing out polarisation, giving

$$\rho_S^\theta = \cos^2 \frac{\theta}{2} |\overline{11}\overline{2}\rangle \langle \overline{11}\overline{2}| + \sin^2 \frac{\theta}{2} \left| \frac{1}{2} \right\rangle \left\langle \frac{1}{2} \right|. \quad (6.52)$$

The angle θ directly controls the subspace populations: when $\theta = 0$ the state looks bosonic, and when $\theta = \pi/2$ it looks fermionic. Measuring the counting statistics after propagation through some interferometer therefore permits simulation of fermionic statistics with bosonic particles [304]. Equivalently this control permits variation between a HOM dip and a HOM peak, purely by adjusting the exchange symmetry of the input state. Other values of θ simulate *anyons* that have neither integer nor half-integer spin, and define representations of the braid group [305]. The symmetry properties of the spectral entanglement from an SPDC source can be controlled to probe two-particle symmetries in a similar way [301, 302].

Another species of particle that would be interesting to simulate are *paraparticles* [287, 306–

308]. In Chapter 2 we invoked the Symmetrisation Postulate to assert that the only particles found in nature are bosons and fermions. Using the approach developed in this chapter, we would say that all particles either reside completely in the totally symmetric irrep or the totally anti-symmetric irrep of the symmetric group S_N . For example, a bosonic state of three particles with states labelled a, b, c is given by $|\overline{abc}\rangle$ and, together with a global phase, corresponds to a ray in $\mathcal{H}^{(3)}$ that spans the one-dimensional $\lambda = (3)$ irrep of S_3 . Similarly for fermions, assuming the three states are orthogonal, the anti-symmetric state is associated with a ray spanning the one-dimensional $\lambda = (1, 1, 1)$ irrep. These wavefunction symmetries derived from our assumption that permuting pairs of particles can only invoke a sign change on some multiparticle wavefunction: $\hat{P}_{i,j}|\Psi\rangle = \pm|\Psi\rangle$. This defines the symmetric and anti-symmetric irreps that are both one-dimensional. Therefore we tacitly assume that every physical state of N identical particles corresponds to just *one* ray in $\mathcal{H}^{(N)}$. However the theory of identical particles asserts only that expectation values for any physical observable are invariant under permutations of the particles in some quantum state – it does not necessarily rule out multiple rays being associated with the same quantum state (though this appears to be the case in nature). Indeed any two vectors may be said to represent the same quantum state if they give the same expectation values for all physical observables [306, 308]. We saw something reminiscent of this in the previous section: three-particle scattering probabilities had identical contributions from the $p = 1$ and $p = 2$ mixed symmetry irreps. In particular, the mixed symmetry irrep of S_3 for particles in orthogonal states 1, 2, 3 is spanned by the four orthonormal vectors

$$\left\{ \left| \overline{\begin{smallmatrix} 1 & 2 \\ 3 \end{smallmatrix}}_1 \right\rangle, \left| \overline{\begin{smallmatrix} 1 & 2 \\ 3 \end{smallmatrix}}_2 \right\rangle, \left| \overline{\begin{smallmatrix} 1 & 3 \\ 2 \end{smallmatrix}}_1 \right\rangle, \left| \overline{\begin{smallmatrix} 1 & 3 \\ 2 \end{smallmatrix}}_2 \right\rangle \right\}. \quad (6.53)$$

The first two vectors define a two-dimensional irrep of S_3 labelled by the inner multiplicity $r = 1$, and they both essentially describe the same quantum state for three paraparticles. For example we earlier saw they gave the same matrix elements $\langle \overline{\begin{smallmatrix} 1 & 2 \\ 3 \end{smallmatrix}}_i | U_S | \overline{\begin{smallmatrix} 1 & 2 \\ 3 \end{smallmatrix}}_i \rangle$ for $i = 1, 2$. Analogously the second pair define another two-dimensional irrep labelled $r = 2$. In contrast to bosons and fermions, the quantum state of paraparticles is not labelled by a unique ray but rather by the specified mixed symmetry irrep of the symmetric group, that in general is spanned by multiple vectors [306]. The unusual symmetry properties of $m < N$ paraparticles drawn from a total state of N paraparticles have attracted some theoretical interest [306, 308, 309]. While such particles have not been observed, it would be interesting to investigate the ability to simulate their behaviour by combining multiple degrees of freedom in entangled states of photons.

Closely related to paraparticles are another set of hypothetical particles called *immanons* [310]. Their states are described by vectors with a more general symmetry than bosons and fermions. For N single-particle states $|\phi_i\rangle$, the multiparticle immanon wavefunction is

$$|\Psi_\lambda\rangle = \frac{1}{\sqrt{N!}} \sum_{\sigma \in S_N} \chi_\lambda(\sigma) |\phi_{\sigma(1)}\rangle \otimes |\phi_{\sigma(2)}\rangle \otimes \dots \otimes |\phi_{\sigma(N-1)}\rangle \otimes |\phi_{\sigma(N)}\rangle. \quad (6.54)$$

The function χ_λ is called the *character* and maps elements of the symmetric group to complex numbers, and tables can be found in [311]. For the special case of the symmetric irrep $\lambda = (N)$, χ_λ is unity for all permutations and we recover the symmetrised state for bosons. On the

other hand, for the anti-symmetric irrep $\lambda = (1, \dots, 1)$, the character reduces to $\text{sgn}(\sigma)$ that is $+(-)1$ for an even (odd) number of transpositions when decomposing σ . This leads to the anti-symmetrised state for fermions, given earlier in equation 2.6. The role of χ_λ becomes more interesting for immanons described by mixed symmetry irreps, and the interference of indistinguishable immanons leads to scattering probabilities given by [310]

$$P_\lambda(\vec{s}) = \frac{1}{N! \prod_{j=1}^N s_j!} \sum_{\rho \in S_N} \left| \sum_{\sigma \in S_N} \chi_\lambda(\sigma) \prod_{j=1}^N M_{\sigma_j, \rho_j} \right|^2. \quad (6.55)$$

The quantity whose squared-modulus must be calculated is the *matrix immanant* $\text{imm}_\lambda(M_\rho)$, where M_ρ has its columns permuted according to ρ . This reduces to the permanent for $\lambda = (N)$ and to the determinant for $\lambda = (1, \dots, 1)$. We encountered quantities similar to the immanant earlier when calculating three-photon scattering statistics by looking at occupation of different subspaces (equation 6.47). It is possible to identify:

$$\text{imm}_{\lambda=(2,1)}(U) = \left\langle \begin{array}{|c|c|} \hline 1 & 2 \\ \hline 3 & \\ \hline \end{array} \right| U \left| \begin{array}{|c|c|} \hline 1 & 2 \\ \hline 3 & \\ \hline \end{array} \right\rangle + \left\langle \begin{array}{|c|c|} \hline 1 & 3 \\ \hline 2 & \\ \hline \end{array} \right| U \left| \begin{array}{|c|c|} \hline 1 & 3 \\ \hline 2 & \\ \hline \end{array} \right\rangle. \quad (6.56)$$

For fully distinguishable states, sums of elements in a mixed symmetry irrep reduce to immanants [109]. Immanants can also be used to express scattering probabilities in a similar way to the approach using permanents described in Chapter 2 [108, 110, 111, 285]. In equation 6.52 we saw it is possible to simulate purely fermionic statistics using entangled states of photons. Extending this to simulating pure immanon statistics of three or more particles would be an elegant demonstration of the power of this first quantised formalism.

6.5 Conclusions

We have followed the treatment by Stanisic *et al.* that applies Schur-Weyl and unitary-unitary dualities to the representations of the symmetric and special unitary groups [109]. The multiparticle wavefunction can then be written in a way that separates the degree of freedom resolved in an experiment from that which is traced out on detection, and also makes explicit the correlations between symmetries of the associated Hilbert spaces. Indistinguishability in a label degree of freedom means that the state for the resolved degree of freedom lies fully in the symmetric subspace. However distinguishability in the HOM interference of two photons was shown to degrade this perfect port exchange symmetry by mixing in components from the anti-symmetric subspace. The introduction of more photons introduces subspaces with more complex symmetries, and distinguishability is associated with populations and coherences in the non-symmetric subspaces. The collective distinguishing property for three particles was shown to correspond to an imaginary coherence between these mixed symmetry subspaces, and we showed how counting statistics can be used to probe this quantity. Including entanglement in interference meant dispensing with the concept of distinguishability in a degree of freedom, and we briefly reviewed another approach to isolating N -particle interference from lower-order contributions. Finally we applied this formalism to entangled states to demonstrate how photons can be used to simulate statistics of particles with non-bosonic symmetry.

Efforts to demonstrate boson sampling in realistic experiments have motivated investigations into how distinguishability degrades the hardness of sampling from the associated distribution [132, 133, 312]. We showed this for the straightforward case of two photons in Section 6.3.1: when the reduced system state is a maximal mixture over the symmetric and anti-symmetric irreps, the coincidence statistics are a balanced sum of permanent and determinant contributions. This simplifies to $P_{\underline{1}} = \text{perm}|U_S|^2$, and describes scattering of distinguishable photons with no interference contributions. For a state of N distinguishable particles, the reduced state is given by a balanced mixture over irreps of all symmetries and can be written as [109]

$$\rho_d = \frac{1}{N!} \sum_{\lambda, p, r_S} |\lambda, p, \underline{n_S}, r_S\rangle \langle \lambda, p, \underline{n_S}, r_S|. \quad (6.57)$$

This is related to the projector $\Pi_{\underline{S}}^{n_S}$ defined in equation 6.34 to determine output probabilities: $\rho_d = \Pi_{\underline{S}}^{n_S}/N!$. Summing the contributions from all the irreps to some output probability (given by a permanent, a determinant and immanants) will then reduce to the permanent of a real matrix, as seen for HOM and also from the expression for scattering probabilities using elements from the distinguishability matrix $\mathcal{S}$ given in equation 2.54. It would be interesting to isolate the role of multiparticle coherences presented here in the transition between different degrees of computational complexity. Another related endeavour is to generalise the concept of wave-particle to many-body quantum systems. In the double slit experiment there is an intrinsic trade-off between fringe visibility and which-path information [63]. In multiparticle scattering, the label degree of freedom can provide this type of information and similar complementarity relations can be used to bound expected signal visibilities [313].

Conclusions

In this thesis we have concentrated on the quantum interference of independent photons, one of the most fundamental effects in quantum optics. We performed natural extensions of the two-photon Hong-Ou-Mandel experiment and investigated how and why distinguishability and mixedness affect interference.

In Chapter 2 we reviewed the double slit and HOM experiments, where distinguishing information is accompanied by a loss of interference and a return to classical probabilistic behaviour. We identified distinguishing parameters that describe how the partial exchange symmetry of the multiparticle wavefunction provides which-path information, and reviewed the theoretical tools needed to extend this study to larger numbers of particles.

In Chapter 3 the distinguishability associated with a system of three particles was shown to depend on a collective phase that goes beyond a pairwise description: the triad phase. We experimentally demonstrated that temporal distinguishability leads to richer behaviour in three-photon interference experiments, and used the polarisation degree of freedom to access the triad phase. By varying this parameter, we challenged the idea that reduction in state overlap generally lowers the contrast of interference signals. Careful preparation of three photons' states in polarisation and temporal modes then allowed isolation of the triad phase associated with three-particle exchange, independent of single- and two-particle contributions. Results were found to be consistent with a comprehensive simulation of realistic experiments.

We began Chapter 4 by relating multiparticle phases to the higher-order exchange contributions of quantum interference. In some cases these can be decomposed into triad phases, and then two- and three-photon measurements are sufficient to characterise a multiparticle interference experiment. However it is possible to define phases that cannot be decomposed in this way by preparing photons in distinguishable states. We presented such a preparation for four photons in polarisation and temporal modes and observed interference that depends on a four-particle phase despite pairwise distinguishability of states, and independently of lower-order exchange contributions. This runs counter to assumptions from one- and two-particle experiments that distinguishability means a return to classical dynamics.

Next we expanded our treatment to include the effects of impure state preparation in Chapter 5. This opens up a much larger parameter space, so we concentrated on small-dimensioned systems with small numbers of particles. A three-photon experiment using a tritter

highlighted the different effects of distinguishability and mixedness, in contrast to HOM where they cause a similar degradation of interference. A geometric interpretation of the two effects was derived for three qubit states and shown to permit identification of state impurity. We showed how to resolve a shortcoming of a standard technique for characterising interferometers using quantum light, and discussed the application of controlled quantum interference to state tomography.

Chapter 6 saw us dispense with the second quantised formalism and revert to explicit expressions for multiparticle wavefunctions. We applied group theoretic methods to isolate the exchange symmetries of the wavefunctions for independent degrees of freedom. In this picture the effect of distinguishability is to spread population and coherences out of the fully symmetric subspaces. Multiparticle collective phases were linked to the richer symmetry structure of larger systems' Hilbert spaces. We concluded by discussing applications of this formalism to simulating particles with exotic statistics.

7.1 Outlook

An obvious question after reading this thesis is what happens for more than four photons? While there are currently no plans to continue this investigation, we suspect that many of the qualitatively new features are already captured by our considerations of four or fewer photons: pairwise distinguishabilities, multiparticle phases, and the possibility of distinguishable state interference look to be the obvious properties of distinguishability in the graph model, and the role of these phases in mixed symmetry Hilbert spaces is clear from our treatment of three photons. Of course larger systems with more particles and modes contain many more paths, and the boson sampling route to quantum advantage relies on this inherent computational complexity. However in this thesis we have seen that there is plenty of complexity (though not the computational kind) even in small extensions of HOM interference.

Large-scale interference will likely underpin future photonic quantum technologies for simulation and computing. As systems grow in size it is important to remember that intuition at the small scale does not necessarily still hold. For example we have seen surprises like how indistinguishability does not guarantee the strongest interference contrast and distinguishability does not always mean probabilistic scattering. Moreover we saw a version of Mandel's mind-boggling induced coherence in a difference guise when using the quitter – I admit that seeing the output twofolds suddenly start fluctuating when opening both SPDC sources was initially completely unexpected! This goes with the common pitfall of assuming a system delivers exactly what is needed for an experiment (in this case four independent photons). In reality there will often be subtleties easily overlooked (here interferences associated with multiple sources) or imperfections that when scaled can affect the fidelity of some task in unexpected ways.

Distinguishability in a system is often undesirable and we have seen some ways of diagnosing its presence at the small scale, but are there any situations where it could be useful? In Appendix E we considered the possible postselectable entangled states from some linear network given a preparation of photons that are not all indistinguishable. Turning the problem on its head, this opens up the possibility of using non-orthogonal measurement bases to herald

or witness different types of entanglement. It would be interesting to see if combining this observation with the symmetry discussions of Chapter 6 permits engineering of entangled states robust to certain types of errors.

It is an exciting time for quantum technology: enormous amounts of effort and resources are pushing systems to scales where quantum advantage is feasible relatively soon. Given the rich behaviour we have already seen at the small scale, it will be fascinating to see what new surprises these larger systems hold.

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Appendices

Appendix A

Polarisation compensation

A.1 Compensation in fibre

On propagation through single-mode fibres, random stresses induce birefringence and so cause rotation of the polarisation states of light injected into one end. By the time light arrives at the coupling region, its polarisation state will be scrambled and the desired interference will not be observed. It is possible to compensate for these random unitary rotations so that all polarisation states arrive at the coupling region of an interferometer, up to some common unitary rotation (see Figure A.1).

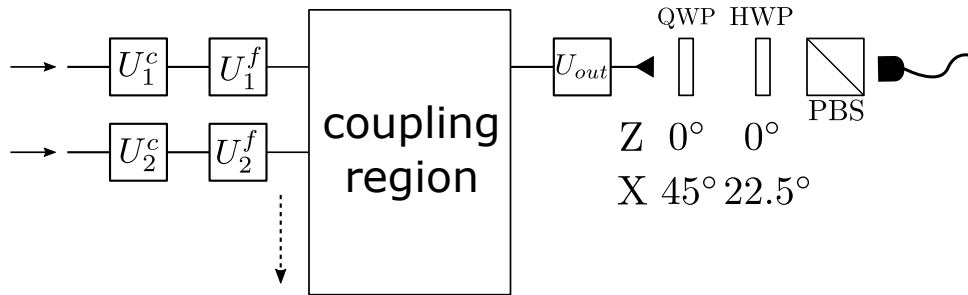


Figure A.1: Photons with defined polarisation states are injected into the inputs of some interferometer. We model components used to compensate for rotations, such as extra waveplates or fibre paddles, by compensation unitaries U_i^c . The random rotations in fibres up to the coupling region are denoted U_i^f . The random rotation in the monitored output fibre is U_{out} . We then consider analysing the emerging photons in either the Z or X bases by suitable preparation of a QWP, a HWP and a PBS whose output is monitored.

Consider first the situation where we want to interfere two photons in H polarisation. Setting the analysis waveplates to measure in the Z basis, we first manipulate U_1^c to maximise counts on the APD, thus achieving

$$U_{out}.U_1^f.U_1^c|H\rangle = |H\rangle. \quad (\text{A.1})$$

Then the state at the coupling region is given by $|\psi_1\rangle = U_{out}^{-1}|H\rangle$. Repeating the same process on the second input port we similarly find the state at the coupling region is $|\psi_2\rangle = U_{out}^{-1}|H\rangle$. The pairwise overlap of the states at the coupler is then $\langle\psi_1|\psi_2\rangle = \langle H|U_{out}U_{out}^{-1}|H\rangle = 1$, demonstrating that the common unitary cancels.

If vertical polarisation states are injected we find

$$U_{out}.U_1^f.U_1^c|V\rangle = e^{i\alpha}|V\rangle. \quad (\text{A.2})$$

Then the overall unitary acting on a polarisation state from input to output is

$$U_{out}.U_1^f.U_1^c = \begin{pmatrix} 1 & 0 \\ 0 & e^{i\alpha} \end{pmatrix}. \quad (\text{A.3})$$

If we were to inject $|+\rangle = \frac{1}{\sqrt{2}}(|H\rangle + |V\rangle)$ into an input, the output state would have some random phase α between the polarisation components. In order to correct for this undesired phase, one must iterate between compensation in the Z and X bases. This ensures preservation of the relative phase between H and V polarisations, and so allows compensation up to some common unitary of an arbitrary input polarisation: $U_{out}.U_1^f.U_1^c = \mathbb{I}$.

A.2 Compensation in the bulk quitter interferometer

The bulk quitter interferometer of Chapter 4 has a polarisation-dependent loss. If we performed the compensation described above, the input state $|d\rangle = \frac{1}{\sqrt{2}}(|H\rangle + e^{i\theta}|H\rangle)$ would arrive at the coupling region as

$$\begin{aligned} |\psi_{coup}\rangle &= U_{out}^{-1}|d\rangle \\ &= \begin{pmatrix} U_{11} & U_{12} \\ U_{21} & U_{22} \end{pmatrix} \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ e^{i\theta} \end{pmatrix} \\ &= \frac{1}{\sqrt{2}} \begin{pmatrix} U_{11} + U_{12}e^{i\theta} \\ U_{21} + U_{22}e^{i\theta} \end{pmatrix}. \end{aligned} \quad (\text{A.4})$$

The probability of a photon being horizontally polarised is then

$$P_H = \frac{1}{2} \left[|U_{11}|^2 + |U_{12}|^2 + 2|U_{11}||U_{12}|\cos(\theta + \beta) \right], \quad (\text{A.5})$$

where $\beta = \text{Arg}(U_{12}) - \text{Arg}(U_{11}^*)$. A similar expression may be derived for vertical polarisation probability P_V . This means that as θ rotates, the probabilities of being horizontally or vertically polarised with respect to the input axes will change. This would not matter for a polarisation independent device: as long as the modes stay orthogonal, the interference would be the same. However due to the polarisation dependent loss of the quitter, this would then result in fluctuations of the singles counts at the quitter's outputs as θ covers the range $0 \rightarrow 2\pi$.

To circumvent this problem we instead perform polarisation compensation by inserting a polariser oriented to transmit horizontal polarisation *inside* the interferometer, just before coupling into the first output fibre. We then adjust the compensation components before the input fibre for $|d\rangle$ and monitor the singles counts on the first output free from any analysis polarisation optics. If a rotation of θ does not change the transmitted singles counts, we have ensured that the axes we define for polarisation are precisely aligned with the axis of the beam splitter, and so $P_H = P_V = 1/2$ throughout. This then defines the output unitary rotation that we must compensate on the other inputs.

Modelling imperfections in multiphoton scattering

There are a number of imperfections in the experiments presented here that merit thorough investigation to see how they affect interference statistics. Adrian Menssen and I wrote a comprehensive package in *Mathematica* to simulate realistic scattering experiments. Here we give an overview of different effects and how they are modelled.

B.1 Pure states

As mentioned in Section 2.3.5, we use the formalism by Tichy [112] to determine scattering probabilities. The input configuration of photons to an interferometer is described by the vector $\vec{r}$; the distinguishabilities of the separable pure states in which photons are prepared are contained in the distinguishability matrix $\mathcal{S}$; the effective scattering matrix M is determined from the scattering matrix U with appropriate row and column multiplicities; the output configuration of interest is denoted by the vector $\vec{s}$. The probability of that scattering event is then given by

$$P(\vec{r}, \vec{s}, \mathcal{S}, U) = \frac{1}{\prod_j r_j! s_j!} \sum_{\sigma \in S_N} \left[\prod_{j=1}^N \mathcal{S}_{j, \sigma(j)} \right] \text{perm} (M * M_{\sigma, \mathbb{I}}^*) \quad (\text{B.1})$$

where perm is the matrix permanent, $*$ is the element-wise Hadamard matrix product, and σ are elements of the symmetric group S_N that are used to permute rows in $M_{\rho, \mathbb{I}}$.

B.2 Multiphoton emission and number correlations

In all experiments presented here, we use nonlinear sources to generate photons. If engineered to have a factorable joint spectrum, they ideally generate a two-mode squeezed vacuum state:

$$|\psi_{TMSV}\rangle = \sqrt{1 - \lambda^2} \sum_{n=0}^{\infty} \lambda^n |n_s, n_i\rangle. \quad (\text{B.2})$$

Using an APD to herald on the idler mode allows the signal to be used as an approximation to a single photon Fock state. Since an APD is a bucket detector (it only clicks if at least one photon was present and is described by $\hat{\Pi}_{APD} = \mathbb{I} - |0\rangle\langle 0|$), the signal mode is left in the mixed state:

$$\rho_s^{(h)} = \frac{1 - \lambda^2}{\lambda^2} \sum_{n=1}^{\infty} \lambda^{2n} |n_s\rangle\langle n_s|. \quad (\text{B.3})$$

The probability of heralding an n photon Fock state is then $P(n|h) = \frac{1-\lambda^2}{\lambda^2} \times \lambda^{2n}$. If multiple heralded sources are used to inject photons into an interferometer, there could be instances where a two-photon Fock state occupies a single input mode. From equation B.1 this means the associated path amplitude is the square of that for a single photon Fock state. If the output detectors cannot register that there are more photons emerging from the interferometer than intended, for example because they are bucket detectors, the measured statistics are a weighted sum of the desired single photon statistics and contaminations from higher orders (see Figure B.1). Similar contamination occurs when using two sources in the semi-heralded configuration used to generate three photons shown in Figure 3.5, and its effect is modelled in the same way with suitable adjustments to the generation probability if the source is not heralded. These processes can also be included for three heralded sources by considering, for example, the contamination to tritter statistics due to inputting the configuration $\vec{r} = (2, 1, 1)$.

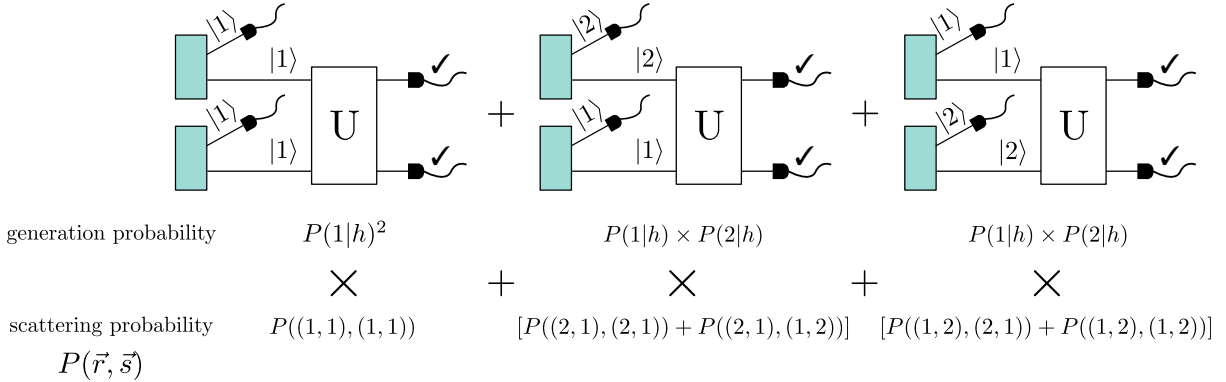


Figure B.1: Illustration of how multiphoton emission by heralded sources can contaminate statistics. The scattering probabilities can be calculated using equation B.1 and the generation probability with equation 3.22. The first contribution gives the desired signal resulting from single photon inputs to some interferometer U . The other diagrams represent the (less likely) events where one of the sources fired twice, and the bucket detectors register three photons at the outputs as a valid coincidence. The measured coincidences are therefore a sum of the desired statistics with contamination from multiphoton events. The squeezing strength λ^2 determines how likely each multiphoton component is, and so how many terms in this series should be included.

B.3 Detector efficiencies and losses

Undesired scattering off optical components or absorption lead to the loss of photons. In conjunction with multiphoton source emission, losses before and after an interferometer affect measurements in quite different ways.

B.3.1 Output losses

We combine the loss after an interferometer and the detector's finite efficiency into a single parameter $0 \leq \eta \leq 1$ for each output mode. This overall efficiency is modelled by a beam splitter with transmissivity $\sqrt{\eta}$ in front of a perfect bucket detector. The probability of a 'click' given one photon is $P(\text{click}|1) = \eta$. If instead there are two photons in this output mode, for example due to a higher order source emission like in Figure B.1, the probability of a detector click is then $P(\text{click}|2) = 1 - P(\text{no click}) = 1 - (1 - \eta)^2 \approx 2\eta$ when $\eta \ll 1$. In other words the undesired interferences are twice as likely to trigger a detector than the ideal signal, thus increasing the relative weighting of the contaminations in Figure B.1.

More generally, consider some output port with N total photons and a detector sensitive up to n photons (for example, a transition-edge sensor or pseudo-number resolved detectors with the same efficiencies, see Appendix C). The probability of a successful detection of n photons in the presence of losses is given by $P(\text{click}) = \sum_{i=0}^{N-n} \binom{N}{i} \eta^{N-i} (1 - \eta)^i$, where i is the number of photons lost. The effect of output losses is therefore to weight statistics by a combinatorial factor that is straightforward to include in a simulation. Losses on the heralding arms can be simulated in the same way.

B.3.2 Input losses

Input losses can change what interference takes place. Taking the example in Figure B.1, we can model the effect of input loss by inserting beam splitters with transmission coefficients given by the throughput t_i of each mode to the interferometer. These couple the inputs to loss modes whose occupations are traced out since we never know how many photons we lost (see Figure B.2).

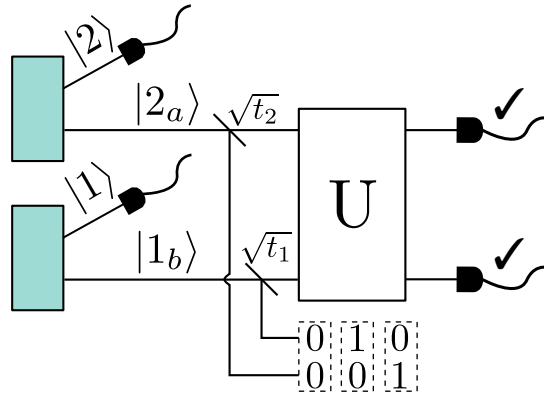


Figure B.2: We imagine two heralded sources preparing the states $|a\rangle$ and $|b\rangle$. Given a higher order emission from one source, we can model the effect of input losses by imagining beam splitters before the interferometer. These have transmissivity given by the throughput of each input mode, and they couple the inputs to loss modes whose photon numbers are not resolved. The possible valid configurations of lost photons are shown in dashed boxes, leaving enough photons to still be able to trigger a coincidence at the output.

If no photons are lost then we are left with the same contamination discussed in the previous section. If one photon is lost from the top source, the dynamics follow from interference of a photon in state $|a\rangle$ and another in $|b\rangle$. On the other hand, if one photon is lost from the bottom

source, no interference takes place and the probability of two photons prepared in $|a\rangle$ triggering a coincidence is determined solely by combinatorics in the interferometer. In simulations we account for all combinations of lost photons that can still lead to the desired detection event.

B.4 Mixed states

If there is residual spectral entanglement in the non-linear photon sources then heralding will leave the other photon in a spectrally mixed state. As mentioned in Chapter 5 and [112, 114], the scattering probability for inputs that are singly occupied by mixed states is

$$P_{\text{mix}}(\vec{r}, \vec{s}, \vec{\rho}, U) = \frac{1}{\prod_j r_j! s_j!} \sum_{\sigma_j \in S_n} \left[\left(\prod_{\mu_i(\sigma)} \text{Tr} \left[\prod_k \rho_{\mu_i^k(\sigma_j)} \right] \right) \times \text{perm}(M * M_{\sigma_j}^*) \right] \quad (\text{B.4})$$

where ρ is a vector of density matrices for the photons injected into each input mode and σ_j is an element of the symmetric group S_n . $\mu(\sigma)$ is the cycle structure of that particular element, μ_i is the i^{th} cycle, and μ_i^k is the k^{th} element of the i^{th} cycle. For example $\sigma_3(S_3) = (2, 1, 3)$, so the cycle structure $\mu(\sigma_3) = (12)(3)$ and then $\mu_1(\sigma_3) = (12)$.

As a concrete example we here consider the situation from the Appendix of [144]: the experiment using the SFWM source presented in Section 3.6. We model state mixedness by assuming the total density matrix for each photon is a tensor product of the desired pure state (in temporal and polarisation degrees of freedom) and some mixed state [167]:

$$\rho_i = \rho_{i,\text{pure}} \otimes \rho_{i,\text{mixed}}, \quad \rho_{i,\text{pure}} = \rho_{i,\text{pol}} \otimes \rho_{i,\text{temp}}, \quad (\text{B.5})$$

where the pure state is given by the tensor product of pure polarisation and temporal states. An orthogonal basis for temporal modes $\{|\tau_j\rangle\}$ can be found by a Gram-Schmidt procedure. Three general temporal modes $|t_k\rangle$ can be written in this basis as [20, 144, 228]

$$\begin{aligned} |t_1\rangle &= |\tau_1\rangle, \\ |t_2\rangle &= \langle t_1|t_2\rangle |\tau_1\rangle + \sqrt{1 - |\langle t_1|t_2\rangle|^2} |\tau_2\rangle, \\ |t_3\rangle &= \langle t_1|t_3\rangle |\tau_1\rangle + \alpha |\tau_2\rangle + \sqrt{1 - |\alpha|^2 - |\langle t_1|t_3\rangle|^2} |\tau_3\rangle, \end{aligned} \quad (\text{B.6})$$

where $\alpha = \frac{\langle t_2|t_3\rangle - \langle t_1|t_2\rangle \langle t_1|t_3\rangle}{\sqrt{1 - |\langle t_1|t_2\rangle|^2}}$. For simplicity, we assume that the mixed state $\rho_{i,\text{mixed}}$ occupies some two dimensional space so that each input photon is prepared in a classical mixture across two modes [167].

B.5 Fluorescence and noise

As mentioned in Section 3.2.7 and [167], the SFWM source suffers from fluorescence processes that contaminate the emission. We model this by assuming that distinguishable photons are created with probabilities P_S and P_I on the signal and idler respectively. The probability of creating zero such photons is given by $(1 - P_S)(1 - P_I)$, and the probability of producing k in the signal mode, and l in the idler mode is then $(1 - P_S)P_S^k(1 - P_I)P_I^l$. The emission of a single

source is then described by the density matrix

$$\rho_{FWM} = (1 - \lambda^2)(1 - P_S)(1 - P_I) \sum_{n,k,l=0}^{\infty} \lambda^{2n} P_S^k P_I^l |n_s, n_i, k_s, l_i\rangle \langle n_s, n_i, k_s, l_i|. \quad (\text{B.7})$$

The total number of photons $2n + k + l$ now includes possible noise photons that propagate without interference. For the simulation in Section 3.6 we use $\lambda = 0.16$ as determined using heralded second-order correlation measurements. $P_S = 0.009$ and $P_I = 0.035$ are determined by monitoring the signal and idler emissions on a spectrometer when the phase-matching of SFWM is switched off, and then integrating the relative signals. The spectral purity is estimated as $\mathcal{P} = 0.95$. Including an input transmission of 70% and residual distinguishability of $r_{ij} = 0.95$, we obtain the simulated curves shown in Figures 3.14 and 3.17. The same values are used in both sets of simulations, and provide good agreement with measured data.

Event probabilities for spatially multiplexed detectors

C.1 Pseudo-number resolution

There are true number-resolving detectors based on thin films of material held at their superconducting transition: transition-edge sensors (TESs). The heat imparted by single photons is sufficient to cause a measurable change in the material’s resistance, and its dependence on the deposited energy enables number resolution [314–316]. However these detectors require cryogenic conditions and usually operate at much less than the 80MHz repetition rate of the Ti:Sapph we use for the SPDC and SFWM sources.

For the experiments presented in Chapter 3, we instead spatially multiplex silicon APDs to achieve pseudo-number resolution (see Figure C.1). These are ‘bucket’ detectors that only tell you whether *at least* one photon was detected, but this scheme allows the probabilistic discrimination of events where multiple photons were in the same spatial mode (in our case, the same output fibre of a tritter). Full resolution on all tritter outputs was only possible using the SPDC sources; the additional herald detectors required for the SFWM source meant only five correlator channels were free for output statistics.

Here we denote the true rate for a particular output event by R_{ijk} . For example, R_{111} is the true number of coincidences per second at the tritter outputs, and R_{300} is the true rate for when all photons emerged in the third tritter port. Our job is to determine the conditional probabilities of these events being successfully identified by the bucket detectors so that we can compensate in post-processing. Failures correspond to two or more photons ending up at the same APD.

C.2 Ideal scenario

To begin we assume no losses, ideal balanced beam splitters, and perfect detector efficiencies. We can consider each type of output event in turn. The factors we determine hold for all counts of that type.

Coincidence counts: 111

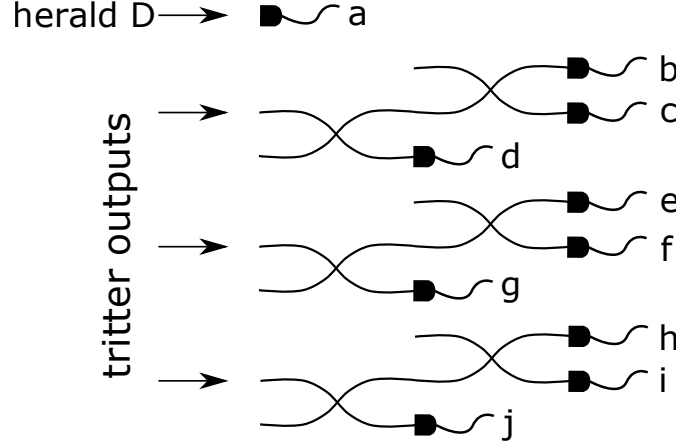


Figure C.1: For the data presented in Section 3.4, the three output fibres of the tritter interferometer were connected to layers of balanced beam splitters as shown. The herald photon triggers an APD connected to channel ‘a’ and the other 9 APDs are connected to 9 more channels on a Swabian TimeTagger Ultra. Heralded threefold coincidences may be identified with particular outputs events and allow probabilistic number resolution.

Since only one photon enters each set of cascaded beam splitters there will never be missed coincidence events. There are $3^3 = 27$ different heralded threefold channels that contribute to the total number of coincidence counts, specifically $a(bcd)(efg)(hij)$ where a single channel is taken from each set of three contained in parentheses. In order to calculate R_{111} it is sufficient, in this ideal case, to sum the counts over all of these channel combinations to give $C_{111} = R_{111}$.

Partially bunched counts: 210

The photon emerging at the second port will always be detected at one of e, f, g . The two photons emerging from the first port must not end up at the same detector:

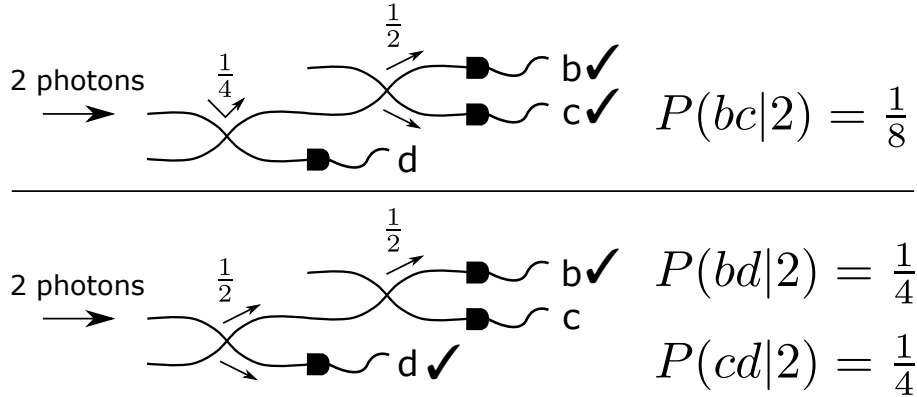


Figure C.2: Three ways that two photons in a single tritter output can be successfully identified using APDs, with their associated probabilities for balanced beam splitters.

The total probability of successfully discriminating two photons in the same port is given by the sum of those shown in Figure C.2, giving $P(210 \text{ success}) = \frac{5}{8}$. When summing the counts for appropriate output detector combinations, given by combinations from $a((bc)(bd)(cd))(efg)$, we thus find the total recorded counts $C_{210} = \frac{5}{8} \times R_{210}$. Therefore to determine the true rate of such partially bunched events, we multiply the total recorded counts by $\frac{8}{5}$.

Fully bunched counts: 300

To identify three photons in the first tritter output, we seek the output event $abcd$. The

probability of success is $P(300 \text{ success}) = \left[\binom{3}{1} \times \frac{1}{2} \times \left(\frac{1}{2}\right)^2 \right] \times \frac{1}{2} = \frac{3}{16}$. The term in square brackets captures the ways that one photon ends up at b , and the other two continue on to the second beam splitter. The final factor gives the probability that those two then split on the second beam splitter. We then record $C_{300} = C_{abcd} = \frac{3}{16} \times R_{300}$. Multiplication of the total recorded counts by $\frac{16}{3}$ therefore corrects for the probabilistic discrimination.

In order to determine relative event probabilities, we determine the true rates R_{ijk} and then find $P_{ijk} = R_{ijk} / \sum R_{ijk}$.

C.3 Dealing with imperfections

In reality we have detectors with imperfect efficiencies. Relative APD efficiencies were determined using attenuated Ti:Sapph light and a balanced fibre beam splitter. Singles were recorded using APDs connected to each output: one used as a reference, and another to be calibrated compared to that reference. Swapping the two detectors around and correcting for dark counts allows determination of relative efficiencies η_i , which equals one for the detector with the best efficiency. Another source of error in counting arises from losses in the components after the tritter. We measured the coupling losses from the tritter output all the way to the final layer of detectors by using single photons from an SPDC source and correcting for η_i . We also measured the actual splitting ratios of all the fibre beam splitters used. This results in slight adjustment of the success probabilities given in the previous section, but turns out to have a relatively small effect.

Data processing comprises first scaling the measured counts (combinations of fourfold coincidences) to correct for the effects of non-uniform APD efficiencies and the losses across the tritter outputs. These corrected counts can then be scaled using newly calculated identification probabilities in order to determine true rates and the overall event probabilities in the presence of these imperfections. These are plotted in Figure 3.13. It is worth noting that this method is reasonably sensitive to the losses used to correct the counts, and between runs there is a risk of changing the coupling if any fibres are unplugged.

C.4 Normalising to singles rates

This method involves knowing the relative rates of singles $\bar{R}_j$ from each of the interferometer outputs $j = 1, 2, 3$ *before* adding layers of splitters and detectors for pseudo-number resolution. These should be normalised to the maximum rate to give numbers $0 \leq \bar{R}_j \leq 1$. The rate of singles is usually far larger than the rates of multiphoton events because of losses and the probabilistic nature of the sources we use. Once the singles rates have been determined, the layers of splitters can be plugged in and any losses accumulated in these fibre-to-fibre connections can be commuted and absorbed by the total channel efficiencies at the end η_i where $i \in \{b, c, d, e, f, g, h, i, j\}$. Assuming again ideal balanced beam splitters at the outputs, we can

then express the recorded singles on each detector in the form $\bar{R}_i \times P(i|j)$

$$\begin{aligned} C_b &= \bar{R}_1 \times \frac{1}{4} \times \eta_b, & C_e &= \bar{R}_2 \times \frac{1}{4} \times \eta_e, & C_h &= \bar{R}_3 \times \frac{1}{4} \times \eta_h, \\ C_c &= \bar{R}_1 \times \frac{1}{4} \times \eta_c, & C_f &= \bar{R}_2 \times \frac{1}{4} \times \eta_f, & C_i &= \bar{R}_3 \times \frac{1}{4} \times \eta_i, \\ C_d &= \bar{R}_1 \times \frac{1}{2} \times \eta_d, & C_g &= \bar{R}_2 \times \frac{1}{2} \times \eta_g, & C_{jd} &= \bar{R}_3 \times \frac{1}{2} \times \eta_j. \end{aligned} \quad (\text{C.1})$$

In the presence of losses and detection inefficiencies, and neglecting higher-order emissions in these heralded threefolds, we can revise the success probabilities from earlier to give

$$\begin{aligned} P'(111 \text{ success}) &= \frac{1}{2} \left[\frac{1}{2}(\eta_b + \eta_c) + \eta_d \right] \times \frac{1}{2} \left[\frac{1}{2}(\eta_e + \eta_f) + \eta_g \right] \times \frac{1}{2} \left[\frac{1}{2}(\eta_h + \eta_i) + \eta_j \right], \\ P'(210 \text{ success}) &= \frac{1}{4} \left[\eta_b \eta_d + \eta_c \eta_d + \frac{1}{2} \eta_b \eta_c \right] \times \frac{1}{2} \left[\frac{1}{2}(\eta_e + \eta_f) + \eta_g \right], \\ P'(300 \text{ success}) &= \frac{3}{16} \times \eta_b \eta_c \eta_d. \end{aligned} \quad (\text{C.2})$$

Each term in these expressions can be associated with a particular coincidence counting channel. In the limit of ideal detectors and no losses we recover the success probabilities given in the previous section.

For the event where one photon emerges from each output port, consider the contributing channel combination $abeh$. If we normalise the recorded coincidences to the product of the corresponding singles we obtain

$$\frac{C_{abeh}}{C_b C_e C_h} = \frac{R_{111} \times P(abeh|111)}{\bar{R}_1 \bar{R}_2 \bar{R}_3 \times P(b|1)P(e|2)P(h|3)}. \quad (\text{C.3})$$

The required success probabilities are given in equations C.1 and C.2 and here we find

$$\begin{aligned} \frac{C_{abeh}}{C_b C_e C_h} &= \frac{R_{111} \times \left(\frac{1}{4}\right)^3 \eta_b \eta_e \eta_h}{\bar{R}_1 \bar{R}_2 \bar{R}_3 \times \left(\frac{1}{4}\right)^3 \eta_b \eta_e \eta_h} \\ &= \frac{R_{111}}{\bar{R}_1 \bar{R}_2 \bar{R}_3}, \end{aligned} \quad (\text{C.4})$$

where the success probabilities and losses have all cancelled. For the other channels contributing to $P'(111 \text{ success})$ the factors from beam splitters cancel in the same way and so these normalised coincidences directly yield the true R_{111} up to some common factor given by the product of relative singles rates.

Now consider a channel contributing to the partially bunched event 210, for example $abce$:

$$\begin{aligned} \frac{C_{abce}}{C_b C_c C_e} &= \frac{R_{210} \times P(abce|210)}{\bar{R}_1 \bar{R}_1 \bar{R}_2 \times P(b|1)P(c|1)P(e|2)} \\ &= \frac{R_{210} \times \frac{1}{8} \eta_b \eta_c \times \frac{1}{4} \eta_e}{\bar{R}_1 \bar{R}_1 \bar{R}_2 \times \left(\frac{1}{4}\right)^3 \times \eta_b \eta_c \eta_e} \\ &= 2 \times \frac{R_{210}}{\bar{R}_1 \bar{R}_1 \bar{R}_2}. \end{aligned} \quad (\text{C.5})$$

Again all the channels contributing to $P'(210 \text{ success})$, when normalised to singles, give the same normalised true rate.

Finally we can investigate a fully bunched channel like $abcd$:

$$\begin{aligned} \frac{C_{abcd}}{C_b C_c C_d} &= \frac{R_{300} \times P(abcd|300)}{\bar{R}_1 \bar{R}_1 \bar{R}_1 \times P(b|1)P(c|1)P(d|1)} \\ &= 6 \times \frac{\bar{R}_{300}}{\bar{R}_1 \bar{R}_1 \bar{R}_1}. \end{aligned} \tag{C.6}$$

Similar expressions hold for all the different coincident channel combinations contributing to a particular counting pattern.

It is also possible to compensate for unbalanced beam splitters by appropriate changes of the various success probabilities. As before these normalised rates R_{ijk} can be used to determine event probabilities P_{ijk} . This technique is robust to losses and is experimentally straightforward to implement. However we did not record the average singles direct from the tritter prior to the data run, so results presented use the previous method.

Accessing a qutrit space using polarisation and temporal modes

In Section 3.1.2 we discussed the constraints on the triad phase given some states in a qutrit space:

$$\begin{aligned} |a\rangle &= |\mathbf{0}\rangle, \\ |b\rangle &= \cos \alpha |\mathbf{0}\rangle + \sin \alpha |\mathbf{1}\rangle, \\ |c\rangle &= \cos \gamma \left(\cos \beta |\mathbf{0}\rangle + e^{i\delta} \sin \beta |\mathbf{1}\rangle \right) + \sin \gamma |\mathbf{2}\rangle. \end{aligned} \tag{D.1}$$

The triad phase is then

$$\varphi_{abc} = \text{Arg} \left(\cos \alpha \cos \beta + e^{i\delta} \sin \alpha \sin \beta \right). \tag{D.2}$$

Clearly the values accessible depend on the parameters α, β, δ . In Section 3.6 we saw how a combination of temporal and polarisation modes allowed independent control over the triad phase, meaning the states span a qutrit space. Here we show explicitly the correspondence between the states above and those prepared in our experiment from equation 3.44, that we here relabel for ease of comparison with the qutrit states above:

$$\begin{aligned} |a'\rangle &= \frac{1}{2} \left(\sqrt{3} |H\rangle + |V\rangle \right), \\ |b'\rangle &= \frac{1}{2} \left(\sqrt{3} |H\rangle - |V\rangle \right), \\ |c'\rangle &= \cos \gamma (\cos 2\theta |H\rangle + i \sin 2\theta |V\rangle) + \sin \gamma |\mathbf{2}\rangle, \end{aligned} \tag{D.3}$$

and we have included the effect of distinguishable temporal modes by leaking amplitude into the third qutrit state $|\mathbf{2}\rangle$, with $\cos \gamma = \exp \left(-\frac{|t_a - t_b|^2}{4\sigma_\tau^2} \right)$. Now we proceed by expressing these primed states in the form of equation D.1. We define

$$\begin{aligned} |\mathbf{0}\rangle &= |a'\rangle = \frac{1}{2} \left(\sqrt{3} |H\rangle + |V\rangle \right), \\ |\mathbf{1}\rangle &= |\mathbf{0}^\perp\rangle = \frac{1}{2} \left(|H\rangle - \sqrt{3} |V\rangle \right). \end{aligned} \tag{D.4}$$

In this basis we now write

$$\begin{aligned}
|a'\rangle &= |\mathbf{0}\rangle, \\
|b'\rangle &= \frac{1}{2} \left(|\mathbf{0}\rangle + \sqrt{3} |\mathbf{1}\rangle \right), \\
|c'\rangle &= \frac{1}{2} \cos \gamma \left[\left(\sqrt{3} \cos 2\theta + i \sin 2\theta \right) |\mathbf{0}\rangle + \left(\cos 2\theta - \sqrt{3} i \sin 2\theta \right) |\mathbf{1}\rangle \right] + \sin \gamma |\mathbf{2}\rangle \\
&= \frac{1}{2} \cos \gamma \left[\sqrt{2 + \cos 4\theta} e^{i\theta'} |\mathbf{0}\rangle + \sqrt{2 - \cos 4\theta} e^{i\theta''} |\mathbf{1}\rangle \right] + \sin \gamma |\mathbf{2}\rangle \\
&\rightarrow \cos \gamma \left[\frac{1}{2} \sqrt{2 + \cos 4\theta} |\mathbf{0}\rangle + \frac{1}{2} \sqrt{2 - \cos 4\theta} e^{i(\theta'' - \theta')} |\mathbf{1}\rangle \right] + \sin \gamma |\mathbf{2}\rangle,
\end{aligned} \tag{D.5}$$

where we have defined $\theta' = \arctan\left(\frac{\tan 2\theta}{\sqrt{3}}\right)$ and $\theta'' = \arctan(-\sqrt{3} \tan 2\theta)$, and in the last step we have factored out a global phase and redefined the basis state $|\mathbf{2}\rangle$ accordingly. The angles of equation D.1 can be identified as $\alpha = \pi/3$, $\cos \beta = \frac{1}{2} \sqrt{2 + \cos 4\theta}$, $\cos \gamma = (\sqrt{2 + \cos 4\theta})^{-1}$ and $\delta = \theta'' - \theta'$. From equation D.2, we then recover the expression for φ_{abc} with θ from equation 3.48.

Distinguishability and post-selected entanglement

E.1 Post-selected entanglement

The ability of linear optical components to erase which-path information and hence generate entangled states is well known [317–320]. The general idea for, for example, two photons is to prepare a separable state such as $|\psi_{in}\rangle = |H_1, V_2\rangle$ where here numbers label the input ports of a balanced beam splitter (see Figure E.1a). The state emerging from such a device is

$$|\psi_{out}\rangle = \frac{1}{2} (|H_1, V_1\rangle + |H_1, V_2\rangle - |V_1, H_2\rangle - |H_2, V_2\rangle) \quad (\text{E.1})$$

where now numbers denote output ports. Post-selecting on one photon per output port then yields the entangled state $|\psi_{ps}\rangle = \frac{1}{\sqrt{2}} (|HV\rangle - |VH\rangle)$, where the ordering of states implies orthogonal port modes. This can be used to violate a Bell inequality.

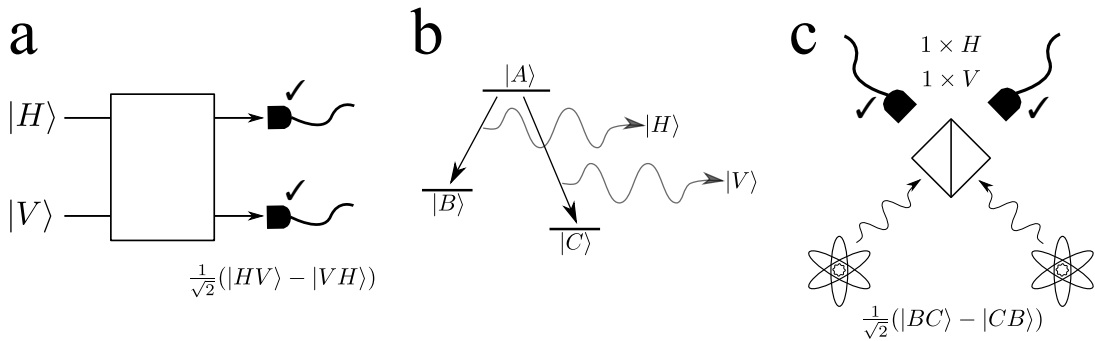


Figure E.1: **a** Injecting one horizontal and another vertical photon into a balanced beam splitter allows post-selection of an entangled state of two photons with success probability $P_{succ.} = \frac{1}{2}$. **b** Toy model of the level structure of some ion, where decay from an excited state $|A\rangle$ to either $|B\rangle$ or $|C\rangle$ is accompanied by a photon in either $|H\rangle$ or $|V\rangle$ respectively. **c** If the emission of two excited ions is sent through a balanced beam splitter, polarisation-resolved detection of H on one detector and V on the other projects the ions into an entangled state.

This can also be used to herald entanglement of matter systems. Imagine some excited state of an ion that can either emit an H photon to end in state $|B\rangle$, or alternatively emit a

V photon and end in state $|C\rangle$ that is orthogonal to $|B\rangle$ (Figure E.1b). Routing the emission from two excited ions through a beam splitter erases information about the internal states of the ions. Detection of one horizontal and another vertical photon then projects the ions into an entangled state (Figure E.1c). A similar method was used to prepare nitrogen vacancy centres in an entangled state for demonstration of the first loophole-free Bell test [251].

E.2 Engineering entanglement using distinguishability

Injecting $M - 1$ horizontally polarised photons and one vertically polarised photon into an $M \times M$ Fourier multiport permits post-selection of an M -photon W-state [321]. The example for three photons is shown in Figure E.2a and, as for two photons, this technique can be used to project three ions into a W-state with success probability $1/9$ (see Figure E.2b).

Now consider photons prepared in the states defining $\varphi_{abc} = \pi$ from equation 3.43:

$$\begin{aligned} |a\rangle &= |H\rangle, \\ |b\rangle &= \frac{1}{2} (|H\rangle + \sqrt{3}|V\rangle), \\ |c\rangle &= \frac{1}{2} (|H\rangle - \sqrt{3}|V\rangle). \end{aligned} \quad (\text{E.2})$$

When these are injected into a balanced tritter, the post-selected state at the output is

$$\begin{aligned} |\psi_{post}^{\varphi_{abc}=\pi}\rangle &= \frac{1}{2} (-|HHH\rangle + |HVV\rangle + |VHV\rangle + |VVH\rangle) \\ &= \frac{1}{\sqrt{2}} (-|H\rangle |\Phi^-\rangle + |V\rangle |\Psi^+\rangle) \end{aligned} \quad (\text{E.3})$$

where $|\Phi^-\rangle, |\Psi^+\rangle$ are Bell states, and the success probability is $1/12$. This is a linear cluster state where measurement of the first photon's polarisation heralds the presence of a post-selected Bell state on the other ports, whose type depends on the first measurement outcome [322]. This could be used to herald an equivalent entangled state of ions (Figure E.2d). This technique of erasing emitter information and then measuring photons in non-orthogonal bases, with insight from multiparticle distinguishing phases, could be an interesting avenue to explore for heralding or witnessing different types of entanglement.

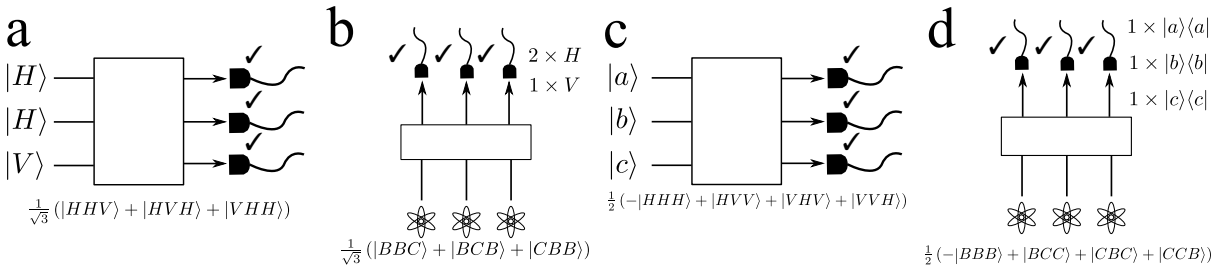


Figure E.2: **a** Extending the scheme shown earlier, injecting two horizontally polarised photons and one vertically polarised allows post-selection of a W-state. **b** This can be used to herald a W-state of three ions. **c** Injecting the partially distinguishable states of equation E.2 permits post-selection of a different type of entanglement. **d** Changing the measurement bases at the output of some interferometer allows manipulation of the heralded entanglement of three ions.

Appendix F

Variations in quitter twofolds from induced coherence

As mentioned in Section 4.4.2, the inability to tell which source has fired can lead to interference from induced coherence. This phenomenon occurs when measuring twofolds at the outputs of the quitter interferometer when two SPDC sources emit into its inputs. The setup is shown in Figure F.1. The quitter unitary matrix is

$$U_{quit} = \frac{1}{2} \begin{pmatrix} 1 & 1 & 1 & 1 \\ 1 & 1 & -1 & -1 \\ 1 & -1 & e^{i\chi} & -e^{i\chi} \\ 1 & -1 & -e^{i\chi} & e^{i\chi} \end{pmatrix}. \quad (\text{F.1})$$

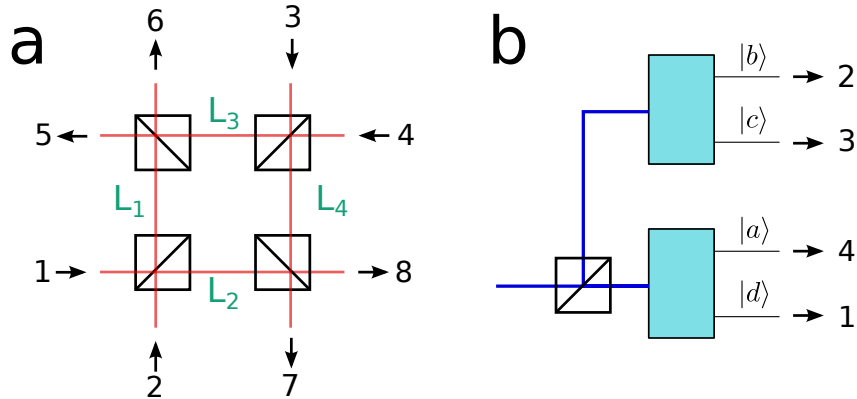


Figure F.1: **a** Quitter construction where inputs 1-4 correspond to the rows of the matrix in equation F.1, and outputs 5-6 correspond to its columns. **b** Pair of SPDC sources with inputs to the quitter and internal states from equation 4.2 labelled.

If one source is opened at a time, the corresponding twofold coincidence probabilities are

$$\begin{aligned} P_{23}^{56} &= \frac{1}{8}(1 - r_{bc}^2), & P_{14}^{56} &= \frac{1}{8}(1 - r_{ad}^2), \\ P_{23}^{57} &= \frac{1}{8}(1 - r_{bc}^2 \cos \chi), & P_{14}^{57} &= \frac{1}{8}(1 - r_{ad}^2 \cos \chi). \end{aligned} \quad (\text{F.2})$$

When both sources are open, the twofolds are *not* simply the sum of the separate statistics. Assuming a phase ϕ_i on input i of the quitter, the input state in the two-photon subspace is

$$|\psi^{(2)}\rangle = \frac{1}{\sqrt{2}} \left(e^{i(\phi_2+\phi_2)} |0, b, c, 0\rangle + e^{i(\phi_1+\phi_4)} |d, 0, 0, a\rangle \right). \quad (\text{F.3})$$

Evolving this through the quitter and monitoring twofolds, one finds

$$\begin{aligned} P_{both}^{56} &= \frac{1}{16} (2 - (r_{ad}^2 + r_{bc}^2) - 2r_{ab}r_{cd} \cos \Phi) \\ P_{both}^{57} &= \frac{1}{16} (2 - \cos \chi (r_{bc}^2 + r_{ad}^2) + 2r_{ab}r_{cd} \cos \chi \cos \Phi), \end{aligned} \quad (\text{F.4})$$

where $\Phi = (\phi_1 + \phi_4 - \phi_2 - \phi_3 + \theta_{cd} - \theta_{ab})$ and the phases $\theta_{ij} = \text{Arg}(\langle i|j \rangle)$. If the phases ϕ_i fluctuate randomly, the new terms will average out over time and the total statistics are given by the sum of the separate coincidences from each source. In an early run of this experiment, the path length differences before the quitter varied at the wavelength scale quickly. We plot some data showing interference from these induced coherences in Figure F.2.

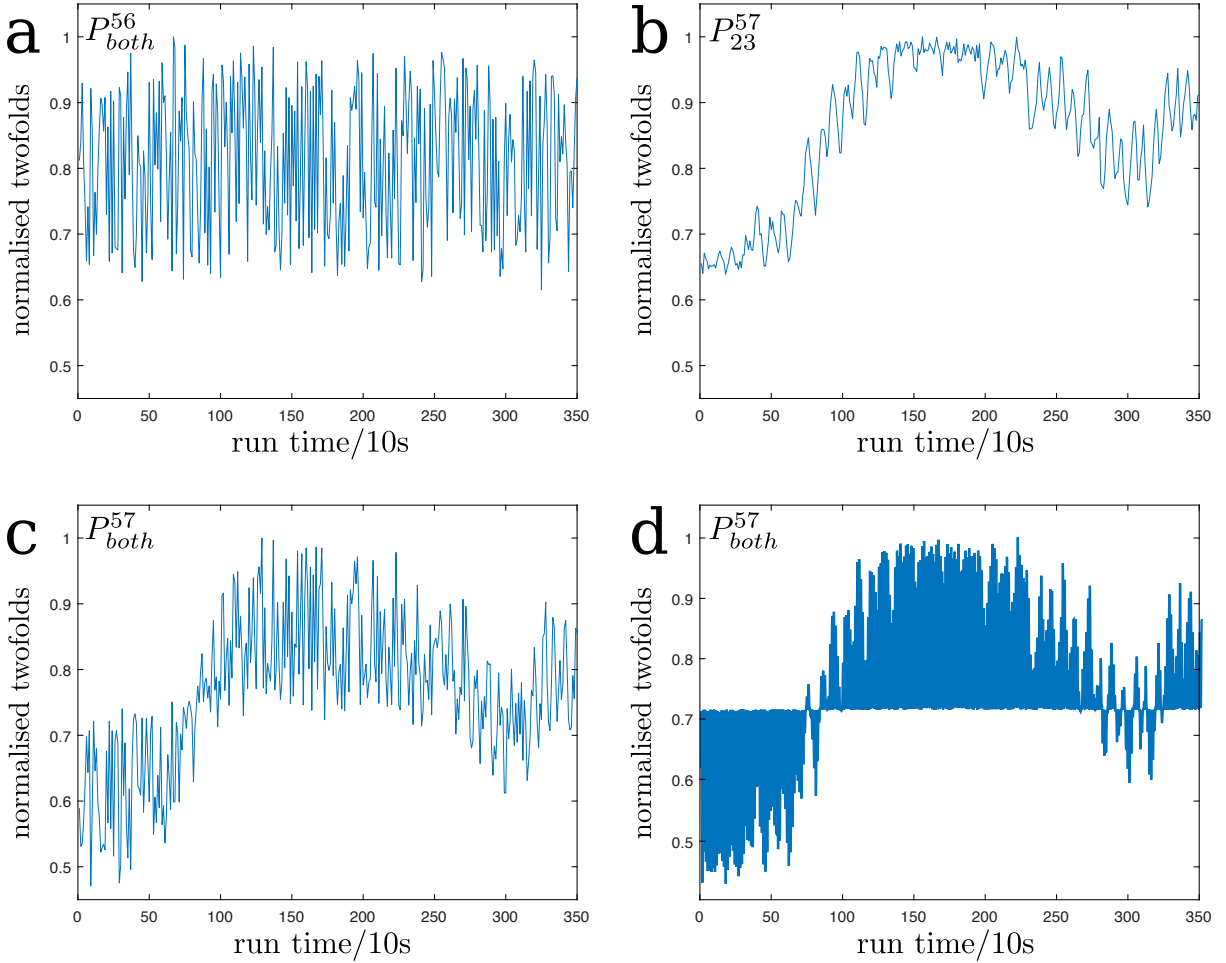


Figure F.2: **a** Measured twofolds at outputs 5 and 6 of the quitter when both sources are open. **b** Measured twofolds at outputs 5 and 7 when injecting a single source into the quitter, showing a dependence on the slowly varying quitter phase χ . **c** Measured twofolds at outputs 5 and 7 when both sources are open, exhibiting fast ϕ_i -dependent oscillations modulated by χ . **d** A simulation of the signal in **c** using values of χ inferred from **b** and $r_{ij} = 0.5$.

In Figure F.2a the P_{both}^{56} statistics vary with the cosine of this fast moving phase with no dependence on χ , and with a visibility of around 40%. Figure F.2b are the twofolds recorded when only one source is open and can be used to infer the quitter phase χ . Figure F.2c are the twofolds measured at outputs 5 and 7 when both sources are open, exhibiting fast oscillations modulated by χ variations. We perform a simulation using the inferred quitter phase χ from P_{23}^{57} measurements, the ideal r_{ij} value, and assuming quickly (and here for simplicity, also periodically) oscillating ϕ_i phases to obtain the predicted signal in Figure F.2d that shows good agreement with the observed statistics.

When collecting the data presented in Section 4.5.2, the beam path through the experiment had been simplified and stabilised. As a result the oscillations of the phases ϕ_i from path length differences before the quitter were slower. The method of applying a controlled Berry phase described in Section 4.4.2 ensures that these phases do not affect the four-photon signal.

Appendix G

Data for distinguishable state interference

During the experiment described in Section 4.5.2 shutters periodically block different arms of the SPDC sources to allow independent measurement of variations in lower order exchange contributions whilst φ_{abcd} is adjusted. In the following we plot counts for the different configurations, where the states listed in the plots' titles denote the prepared state and associated input port.

G.1 Single source arm open

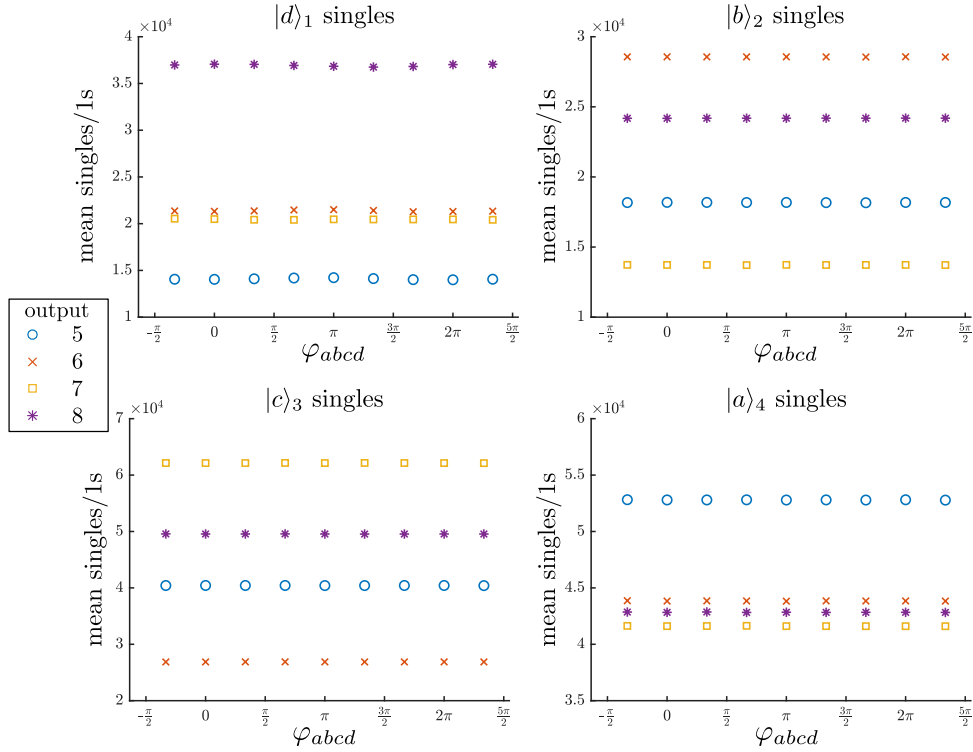


Figure G.1: Output singles for each individual quitter input recorded by blocking other arms. Error bars are smaller than markers so are omitted. See main text for details.

Only the state $|d\rangle$ is varied during the experiment: its polarisation rotates in the Bloch sphere to change the four-particle phase φ_{abcd} . The singles at the quitter outputs when injecting only

this state are shown in the top left plot. Those at outputs 5 and 6 exhibit small variations of 1.4% and 1% respectively that we attribute to slight mismatch of the polarisation axes with those of the quitter (see Appendix A.2). The other outputs exhibit $< 1\%$ variation with φ_{abcd} .

G.2 Two source arms open

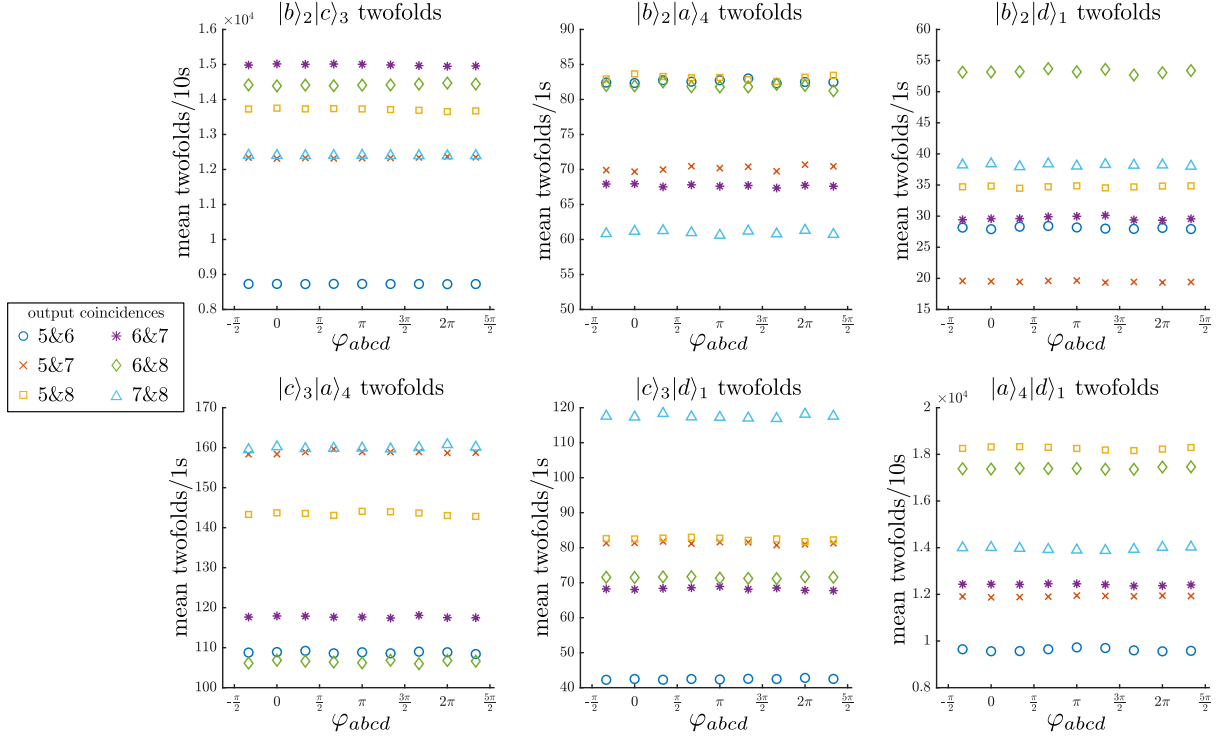


Figure G.2: Output twofold coincidences for each pair of quitter inputs recorded by blocking other arms. Error bars are smaller than markers so are omitted. High counts correspond to twofolds when a single source is used, and lower counts are twofolds between distinct sources. See main text for details.

We expect any variations in the output twofolds to involve the varying state $|d\rangle$. The $|a\rangle_4|d\rangle_1$ twofolds correspond to counts when a single source is injected into the quitter. The coincidences at outputs 5 and 6 exhibit 1.6% variation. Some of this is due to the underlying singles count variations observed in Figure G.1. The effect of these can be removed from the twofold counts by appropriate normalisation.

Consider some true rate of singles and twofolds R from a single source and denote $\eta_d, \eta_a, \eta_5, \eta_6$ the losses on the two inputs and two outputs of interest respectively. We can write down the measured singles and twofold counts at the detectors:

$$\begin{aligned} C_5^d &= \eta_d \eta_5 \times R \times P_{1 \rightarrow 5} \\ C_6^a &= \eta_a \eta_6 \times R \times P_{4 \rightarrow 6} \\ C_{56}^{ad} &= \eta_a \eta_d \eta_5 \eta_6 \times R \times P_{14}^{56}, \end{aligned} \tag{G.1}$$

where $P_{i \rightarrow j}$ is the single photon scattering probability from input i to output j , and P_{14}^{56} is the HOM scattering probability for this set of input states in the quitter. Dividing the twofolds by

the singles gives:

$$\frac{C_{56}^{ad}}{C_5^d \times C_6^a} = \frac{1}{R \times P_{1 \rightarrow 5} \times P_{4 \rightarrow 6}} \times P_{14}^{56}. \quad (\text{G.2})$$

This procedure removes the effect of any (possibly time-varying) losses at the inputs and outputs, thus returning a clean HOM signal up to some normalisation. Applying this to the twofolds that display a 1.6% raw variation of counts yields a normalised HOM signal with 1.2% variation that we attribute to a very small change in r_{ad}^2 as φ_{abcd} is varied. This could arise from a waveplate being 0.5° off the intended angle. This small variation in r_{ad}^2 could cause a maximum of around 1.4% variation in the associated double emission fourfolds for that source, and has a negligible impact on the background-subtracted fourfold signal visibility.

The remaining twofold outputs for the input $|a\rangle_4 |d\rangle_1$ have $< 1\%$ variation. The other (thermal) HOM signals between $|d\rangle$ and the other inputs are constant within error, as are all other signals with inputs not involving $|d\rangle$. Therefore any variations in the pairwise distinguishabilities are either zero or have negligible impact on the fourfold signal.

G.3 Three source arms open

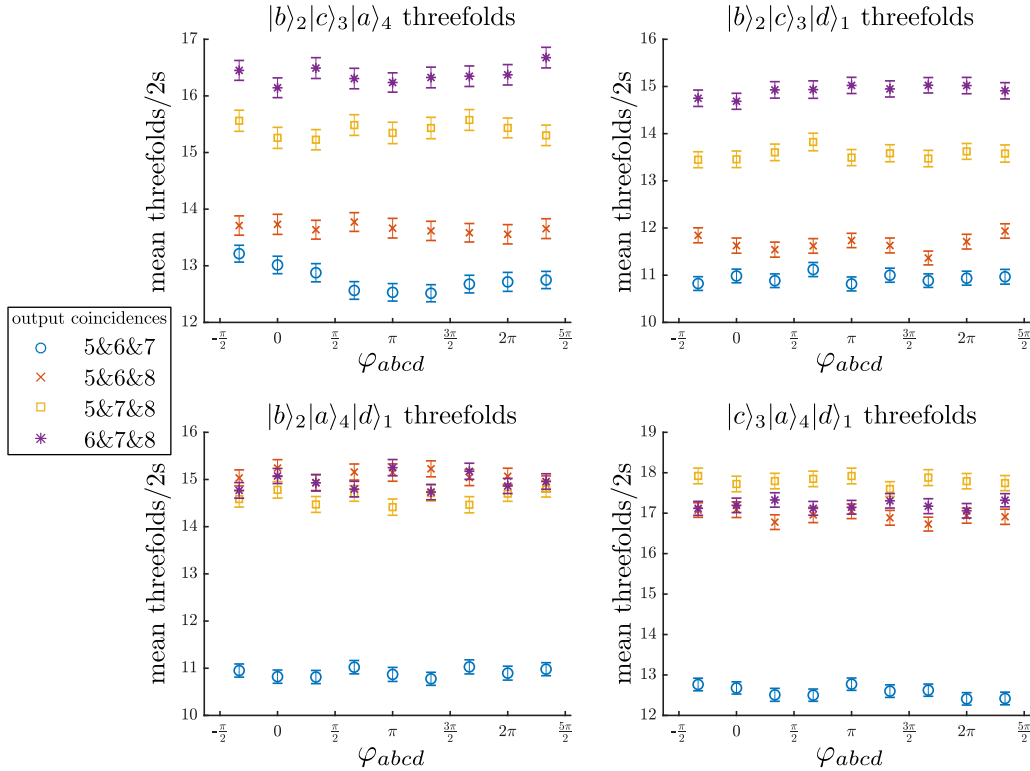


Figure G.3: Output threefold coincidences for each set of three quitter inputs recorded by blocking a fourth arm. Error bars are from repeated sweeps. See main text for details.

The count rates here are lower so the error bars are slightly larger than for other measurements. Again only signals involving $|d\rangle$ are expected to vary (if at all). All these measurements are constant with φ_{abcd} to within error which is good evidence that the pairwise distinguishabilities are constant and any triad phase-dependent contributions arising from small temporal overlap of wavepackets are very small, as expected for our distinguishable state preparation.

Appendix H

Properties of permanents and overlaps of density matrices

H.1 Permanents of scattering matrices

H.1.1 Permutational invariance

The permanent of a matrix A is defined as

$$\text{perm}(A) = \sum_{\sigma_i \in S_N} \prod_{j=1}^N A_{\sigma_i(j),j} \quad (\text{H.1})$$

where σ_i are elements of the symmetric group S_N , and $\sigma_i(j)$ denotes the number that j is mapped to by that element. In the main text we occasionally dispensed with the subscript where the summation was obvious, but here we retain it for clarity.

We define a permutation matrix $\mathcal{P}_\rho$ that reorders the i th rows of some matrix to new rows $\rho(i)$ according to the single element $\rho \in S_N$. Its elements are given by $\mathcal{P}_\rho = \delta_{i,\rho(i)}$. Now consider taking the permanent of A after it has been permuted in this way:

$$\begin{aligned} \text{perm}(\mathcal{P}_\rho A) &= \sum_{\sigma_i \in S_N} \prod_{j=1}^N (\mathcal{P}_\rho A)_{\sigma_i(j),j} \\ &= \sum_{\sigma_i \in S_N} \prod_{j=1}^N A_{\sigma_i \cdot \rho(j),j} \\ &= \sum_{\tau_i \in S_N} \prod_{j=1}^N A_{\tau_i(j),j} = \text{perm}(A), \end{aligned} \quad (\text{H.2})$$

where in the last step we have defined $\tau_i = \sigma_i \cdot \rho$ and, since the sum is still over all elements of the symmetric group, we recover the same permanent despite the permutation ρ .

H.1.2 Permanents related by inverse permutations

The scattering probabilities of equation 2.54 contain permanents like

$$\begin{aligned} \text{perm}(M * M_{\rho, \mathbb{I}}^*) &= \sum_{\sigma_i \in S_N} \prod_{j=1}^N (M * M_{\rho, \mathbb{I}}^*)_{\sigma_i(j), j} \\ &= \sum_{\sigma_i \in S_N} \prod_{j=1}^N M_{\sigma_i(j), j} M_{\sigma_i \cdot \rho(j), j}^* \end{aligned} \quad (\text{H.3})$$

where M is the effective scattering matrix and $\rho \in S_N$. These can be related to the permanents where the rows of the second matrix are instead permuted according to the inverse ρ^{-1} :

$$\begin{aligned} \text{perm}(M * M_{\rho^{-1}, \mathbb{I}}^*) &= \sum_{\sigma_i \in S_N} \prod_{j=1}^N (M * M_{\rho^{-1}, \mathbb{I}}^*)_{\sigma_i(j), j} \\ &= \sum_{\sigma_i \in S_N} \prod_{j=1}^N ((\mathcal{P}_{\sigma_i \cdot \rho \cdot \sigma_i^{-1}}) M * M_{\rho^{-1}, \mathbb{I}}^*)_{\sigma_i(j), j} \\ &= \sum_{\sigma_i \in S_N} \prod_{j=1}^N M_{\sigma_i \rho(j), j} M_{\sigma_i(j), j}^* \\ &= [\text{perm}(M * M_{\rho, \mathbb{I}}^*)]^*. \end{aligned} \quad (\text{H.4})$$

Since transpositions are their own inverse, all permanents associated with pairwise exchanges are real.

H.2 Overlaps of density matrices

H.2.1 Real and complex overlaps

Two-particle exchanges depend on $\text{Tr}(\rho_i \rho_j)$. These are real due to the Hermiticity of density matrices:

$$\begin{aligned} [\text{Tr}(\rho_1 \rho_2)]^* &= \left[\sum_{i, j} (\rho_1)_{ij} (\rho_2)_{ji} \right]^* \\ &= \sum_{i, j} (\rho_1^*)_{ij} (\rho_2^*)_{ji} \\ &= \sum_{i, j} (\rho_1^T)_{ij} (\rho_2^T)_{ji} \\ &= \sum_{i, j} (\rho_1)_{ji} (\rho_2)_{ij} \\ &= \text{Tr}(\rho_1 \rho_2). \end{aligned} \quad (\text{H.5})$$

Three-particle interference has a dependence on $\text{Tr}(\rho_1\rho_2\rho_3)$ and on $\text{Tr}(\rho_1\rho_3\rho_2)$ due to the full permutation elements of S_3 . These are related by complex conjugation

$$\begin{aligned}
[\text{Tr}(\rho_1\rho_2\rho_3)]^* &= \left[\sum_{i,j,k} (\rho_1)_{ij} (\rho_2)_{jk} (\rho_3)_{ki} \right]^* \\
&= \sum_{i,j,k} (\rho_1^*)_{ij} (\rho_2^*)_{jk} (\rho_3^*)_{ki} \\
&= \sum_{i,j,k} (\rho_1)_{ji} (\rho_2)_{kj} (\rho_3)_{ik} \\
&= \text{Tr}(\rho_1\rho_3\rho_2).
\end{aligned} \tag{H.6}$$

H.2.2 Scalar triple product from overlap of three qubit density matrices

We express a qubit density matrix using its Bloch vector:

$$\rho_i = \frac{1}{2} (\mathbb{I} + \mathbf{r}_i \cdot \boldsymbol{\sigma}). \tag{H.7}$$

$\boldsymbol{\sigma}$ is a vector of the Pauli matrices and these obey the commutation relations $[\sigma_\alpha, \sigma_\beta] = 2i\varepsilon_{\alpha\beta\gamma}\sigma_\gamma$. A number of other relations can be derived from these:

$$\begin{aligned}
(\mathbf{r}_i \cdot \boldsymbol{\sigma})(\mathbf{r}_j \cdot \boldsymbol{\sigma}) &= (\mathbf{r}_i \cdot \mathbf{r}_j)\mathbb{I} + i(\mathbf{r}_i \times \mathbf{r}_j) \cdot \boldsymbol{\sigma}, \\
\text{Tr}(\sigma_a \sigma_b) &= 2\delta_{ab}, \\
\text{Tr}(\sigma_a \sigma_b \sigma_c) &= 2i\varepsilon_{abc}, \\
\text{Tr}(\sigma_a \sigma_b \sigma_c \sigma_d) &= 2(\delta_{ab}\delta_{cd} - \delta_{ac}\delta_{bd} + \delta_{ad}\delta_{bc}).
\end{aligned} \tag{H.8}$$

Turning to the overlap of three density matrices describing qubits:

$$\begin{aligned}
\text{Tr}(\rho_1\rho_2\rho_3) &= \text{Tr} \left(\frac{1}{2}(\mathbb{I} + \mathbf{r}_1 \cdot \boldsymbol{\sigma}) \frac{1}{2}(\mathbb{I} + \mathbf{r}_2 \cdot \boldsymbol{\sigma}) \frac{1}{2}(\mathbb{I} + \mathbf{r}_3 \cdot \boldsymbol{\sigma}) \right) \\
&= \frac{1}{8} \text{Tr} \left(\mathbb{I} + \mathbf{r}_1 \cdot \boldsymbol{\sigma} + \mathbf{r}_2 \cdot \boldsymbol{\sigma} + \mathbf{r}_3 \cdot \boldsymbol{\sigma} + (\mathbf{r}_1 \cdot \boldsymbol{\sigma})(\mathbf{r}_2 \cdot \boldsymbol{\sigma}) + (\mathbf{r}_1 \cdot \boldsymbol{\sigma})(\mathbf{r}_3 \cdot \boldsymbol{\sigma}) \right. \\
&\quad \left. + (\mathbf{r}_2 \cdot \boldsymbol{\sigma})(\mathbf{r}_3 \cdot \boldsymbol{\sigma}) + (\mathbf{r}_1 \cdot \boldsymbol{\sigma})(\mathbf{r}_2 \cdot \boldsymbol{\sigma})(\mathbf{r}_3 \cdot \boldsymbol{\sigma}) \right) \\
&= \frac{1}{8} \left(2 + 2\mathbf{r}_1 \cdot \mathbf{r}_2 + 2\mathbf{r}_1 \cdot \mathbf{r}_3 + 2\mathbf{r}_2 \cdot \mathbf{r}_3 + 2i\varepsilon_{abc}\mathbf{r}_1^a\mathbf{r}_2^b\mathbf{r}_3^c \right) \\
&= \frac{1}{4} (1 + \mathbf{r}_1 \cdot \mathbf{r}_2 + \mathbf{r}_1 \cdot \mathbf{r}_3 + \mathbf{r}_2 \cdot \mathbf{r}_3 + i\mathbf{r}_1 \cdot (\mathbf{r}_2 \times \mathbf{r}_3)).
\end{aligned} \tag{H.9}$$

There is now a dependence on the scalar triple product of the Bloch vectors.