

Thesis submitted for the degree of PhD

Infrared spectroscopy of multi-quantum well microcavities

including a study on Q switching the 3 micron erbium laser

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Declaration of Originality

I, Francis John Murphy, declare that the work submitted in this thesis is my own. Contributions made by others have been properly acknowledged.

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Dedicated to my parents

Abstract

Two projects are presented in this thesis. The first project is an investigation into linewidth-narrowing phenomena of intersubband cavity polaritons (ICPs) in a microcavity/multi-quantum well sample through angle-resolved laser spectroscopy. Strong coupling of the vacuum field of the microcavity, and the intersubband transition (ISBT) of the multi-quantum well led to vacuum Rabi splitting of 12.4 meV of the ICP modes at room temperature in the absorbance spectra.

The linewidths of the ICPs were found to be substantially narrowed (4.2 meV, at room temperature) at the anticrossing point, narrower than the bare ISBT and empty microcavity linewidths at room temperature (6.2 and 6 meV, respectively, at room temperature), and narrower than existing theory predicted. The same effect was observed at cryogenic temperatures. Narrowing was explained by the light effective mass of the ICP, rendering the ICP unaffected by interface roughness scattering of the multi-quantum well. The measurement of the narrow linewidths was made possible by the superior angular resolution of laser spectroscopy compared to previously-used thermal light sources.

The second project consisted of the development of a Q switched $\text{Er}^{3+}, \text{Cr}^{3+}:\text{YSGG}$ laser, of 3 μm wavelength, with 76 mJ fundamental mode pulses in free-running mode.

Q switching of the laser was investigated in order to produce short (<400 ns) laser pulses, using two ‘slow’ Q switch methods. The first, a rotating mirror Q switch, was found to produce pulses of ~ 10 mJ, with an optimum mirror rotation rate of 300 rotations per second.

The second Q switch method, a ‘polygon chopper Q switch’ produced single pulses of energy ~ 4 mJ, using a rotating polygon as an optical chopper. From this Q switch, it was deduced that the longest Q switching time possible for $\text{Er}^{3+}, \text{Cr}^{3+}:\text{YSGG}$, with *any* method of Q switching, was ~ 30 μs for single pulse operation, and ~ 80 μs for multiple pulses.

Publications

F. J. Murphy, A. O. Bak, M. Matthews, E. Dupont, H. Amrania, and C. C. Phillips, *Linewidth-narrowing phenomena with intersubband cavity polaritons*. Phys. Rev. B, 89, 205319 (2014).

E. A. Arbabzadah, **F. J. Murphy**, C. C. Phillips, and M. J. Damzen, *High repetition rate diode-pumped, mechanically Q-switched Er:YSGG lasers at $3\mu\text{m}$* . To be published (see Appendix sec. A.4)

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Foreword

It was found convenient to divide the thesis into two Parts. Part I details the midinfrared laser spectroscopy of a multi-quantum well sample displaying strong coupling of light and matter. Part II details the investigation of Q switching the $\lambda \simeq 3 \mu\text{m}$ erbium laser. The unifying trait of the two parts is the midinfrared region of the spectrum.

For convenience, conversions between commonly used units are given:

$$\lambda[\mu\text{m}] = 10000/\bar{\nu}[\text{cm}^{-1}],$$

$$\bar{\nu}[\text{cm}^{-1}] = E[\text{meV}]/0.123984,$$

$$E[\text{meV}] = 1239.84/\lambda[\mu\text{m}].$$

A list of abbreviations and nomenclature follows the Appendix.

Part I.

**Infrared spectroscopy of
multi-quantum well microcavities:
linewidth narrowing of the
intersubband cavity polariton**

Summary of Part I

Analysis of the absorbance spectra of intersubband cavity polaritons and their linewidths is presented. The sample studied is a multi-quantum well slab embedded in a microcavity waveguide with n-doped GaAs mirrors. The fundamental cavity mode in the microcavity strongly couples with the intersubband transition (ISBT) in the conduction band of the quantum wells; this leads to the formation of a coupled mode quasi-particle of a light-matter mixture known as an intersubband cavity polariton (ICP). We find that when the ICP is approximately equal parts light and matter its linewidth is reduced to below that of the predictions of analytical and numerical models.

The ISBT has several sources of broadening, and we investigate two mechanisms that would allow the ICP to overcome this broadening. It was found, aided by analysis of the low temperature linewidths, that the interface roughness scattering broadening of the ISBT is reduced by the ICP having a far lighter effective mass than the ISBT. Furthermore, the ICP also has a greater spatial extent than the ISBT and thus averages over the roughness more.

1. Introduction to Part I

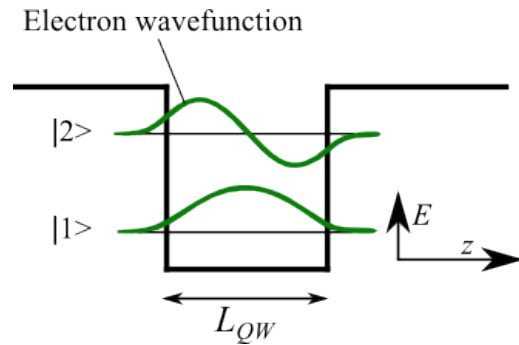
We will begin by introducing the quantum well and the intersubband transition. Then, the subject of microcavities and quantisation of the cavity field will be introduced. These two subjects will then be brought together to introduce the field of intersubband cavity polaritons, the main subject of this thesis. Finally, the sample studied in this thesis, herein referred to as the ‘NRC sample’, is given a brief introduction.

1.1. Quantum wells and the intersubband transition

1.1.1. The particle in a box - becoming a reality

The quantum mechanics of matter, developed in the early 20th Century, led to the realisation that confinement of an electron to dimensions on the order of the electron wavelength, would lead to quantisation of the electron’s energies, in the well-known ‘particle in a box’ scenario (see Fig. 1.1) [1]. The de Broglie wavelength of a non-relativistic electron at room temperature is of the order of 10 nm and fabricating on such small scales remained a challenge for much of the 20th Century. The first

Figure 1.1.: The particle in a box. For example, an electron in a finite potential well, confined to a length, L_{QW} , of the same order as its de Broglie wavelength. Its energies become quantised, here to two levels, the ground state $|1\rangle$ and first excited state $|2\rangle$.



observations of artificially-induced electron energy quantisation were in the inversion layers of Si-MOSFETs [2, 3].

It was not until the end of the 1960s and the development of molecular beam epitaxy (MBE) [4] that the prospect of designable *quantum wells* (QWs) became a reality. MBE allowed layers of semiconductor to be grown at thicknesses of nanometres, with abrupt interfaces between materials. In such a semiconductor QW, the well layer is composed of a semiconductor sandwiched between barrier layers of another semiconductor with a larger bandgap (see Fig. 1.2 b)). The electrons in the conduction band of the well are the ‘particles in a box’ and, if the well width is sufficiently narrow, the electrons will have their continuous energy dispersion quantised into *subbands*. They are subbands rather than discrete levels, because the lack of confinement in two of the 3 dimensions gives the electrons an energy dispersion, with the square of in-plane wavevector, $k_{\parallel}^2$. (See sec. 2.1.)

In order to increase the interaction strength with coupled radiation, many QWs are often grown adjacently to form a multi-quantum well (MQW); the barrier must be sufficiently thick to prevent tunnelling between the wells which would reduce confinement and broaden the subbands¹. Fig. 1.2 shows a typical MQW structure, where the MQW stack is grown atop a much thicker substrate material, then capped. The direction of layer growth is defined as along the z -axis.

MBE is a viable technique for many materials systems, but started out with the III-V semiconductor GaAs (as the well) and AlGaAs (as the barrier) system. This remains, arguably, the major materials system for QW fabrication to this day, and indeed is the system utilised in this thesis for the studied NRC sample.

1.1.2. The quantum well interband transition

Quantisation of electron energies in semiconductor QWs was demonstrated in 1974 at Bell Labs [5]. There, QWs were grown by MBE and consisted of GaAs wells and AlGaAs barrier layers. The evidence of energy quantisation was in the form of ‘steps’ seen in the absorption spectra of the QWs. Whereas in bulk semiconductors, absorption is continuous above the bandgap, in the QW case the absorption spectra

¹In the case of tunnelling to adjacent wells the MQW system is known as a superlattice, and the subbands become broader *minibands*.

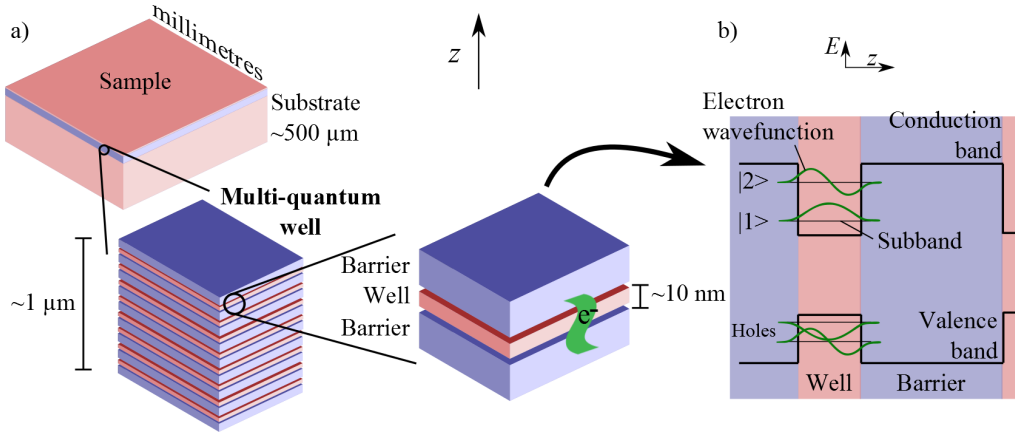


Figure 1.2.: a) Typical growth scheme of an MQW. An MQW stack is grown on top of a thicker substrate. Each QW is of the order of an electron de Broglie wavelength, and the barrier layer material has a wider bandgap compared to the well material, thereby confining electrons within the well. b) An example of a resulting band structure. Electron (and hole) energy becomes quantised forming subbands.

had peaks at energies corresponding to the quantised transitions from the valence band to each subband in the conduction band².

1.1.3. The quantum well intersubband transition

1.1.3.1. General properties of the ISBT

As MBE growth technology matured, it became possible to observe the transitions between the subbands themselves, the *intersubband transitions* (ISBTs): through Raman spectroscopy in 1979 [6], then later through directly observed photon absorption, in 1985 [7]. To observe ISBTs the lower subband must be populated; to this end, MQWs are doped such that the Fermi energy of the electrons is above the ground subband state energy. Through the choice of materials, and band structure engineering, the ISBT is able to access wavelengths continuously in the considerably wide range of 1.55 to 200 μm [8]. Furthermore, the ($k_{\parallel}^2$ -dependent) energy dispersions of the subbands are close to parallel, which leads to a sharp ISBT absorption resonance, almost atomic-like in form. See Fig. 1.3, a).

Since the electrons are only confined in the growth direction (z), in the other two dimensions they behave as a 2D electron gas (2DEG) (Fig. 1.3, b)). This leads to

²These results also gave evidence of the quantisation of the valence band into subbands.

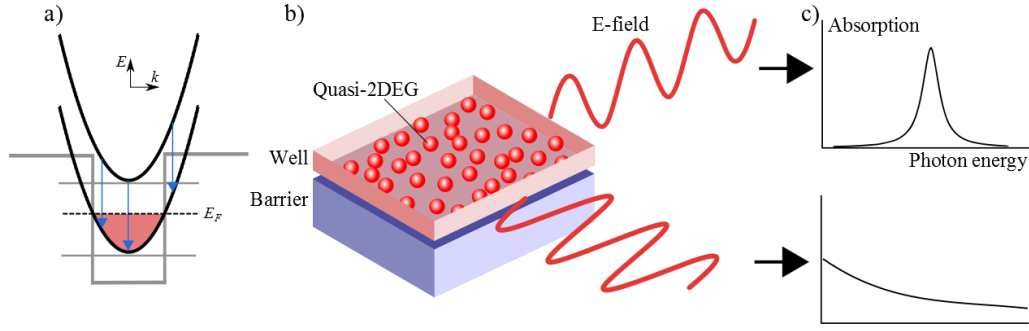


Figure 1.3.: a) The Fermi energy, E_F , is above the lowest subband, thus populating it with electrons. The $k_{||}^2$ -dependent dispersion curves for the subbands are near-parallel giving narrow absorption and emission characteristics. b) The formation of a 2D electron gas (2DEG) in a QW, and its polarisation-dependent interaction with an electromagnetic field. c) Due to the selection rules of the ISBT, only light with polarisation in the growth direction couples to the ISBT (top). Light with polarisation in the plane of the 2DEG feels the response of an electron Drude gas (bottom).

the ISBT selection rule where only light with polarisation in the growth direction of the MQW (TM-polarised) can interact with the ISBT, in which case the 2DEG gives a Lorentzian-shaped absorption profile. On the other hand, light with in-plane polarisation will simply oscillate the electrons in the plane with the response of a Drude gas (Fig. 1.3, c)). Furthermore, coupling between adjacent QWs via the Coulomb interaction leads to collective oscillations, causing all the layers to effectively act as one. The electron gas is hence sometimes described as a quasi-2DEG.

1.1.3.2. ISBT sample geometries

The above selection rule leads to certain constraints for the geometries of MQW ISBT samples, since normal-incidence light will not couple to the ISBT. See Fig. 1.4. Direct coupling into the MQW layer is often performed at Brewster's angle to minimise reflections of the interacting TM-polarisation orientation; however, because of the high refractive index of the typical semiconductor ($n \gtrsim 3$), the magnitude of polarisation in the z -direction is small, and coupling is low. To solve this, light can be coupled into the substrate and then from the substrate into the MQW. Then, to increase the number of interactions between the light and the MQWs, it is common for the substrate to be capped with a reflective layer such that the light is bounced back into the MQW, sometimes multiple times.

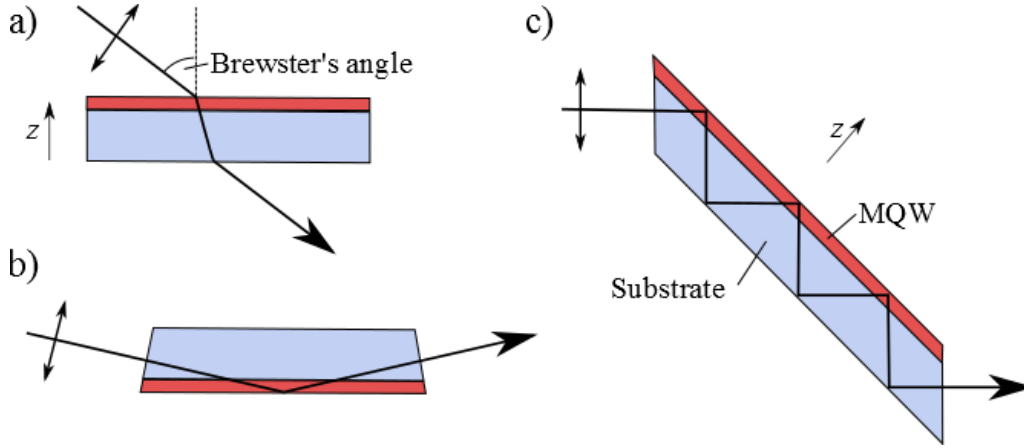


Figure 1.4.: Side view of typical MQW samples showing possible orientations of coupling light into and out of MQW samples: a) light is incident directly onto the MQW at Brewster's angle to minimise reflection of the TM-polarised light, and is transmitted through the substrate. b) Light is coupled through substrate ends and achieves two passes through the MQW. c) A multi-pass geometry. Shown is the path of the probe light through the sample, and the polarisation orientation (TM-polarised).

1.1.3.3. Applications of the ISBT

There are two major applications of the ISBT; the detection and the emission of infrared light.

Quantum well ISBT photodetector

ISBTs were demonstrated for use in photodetectors in 1983 [9] and what is now known as a quantum well infrared photodetector (QWIP) was demonstrated in 1987 [10]. It was soon thereafter incorporated into a focal plane array (FPA) [11, 12], going on to become a well established, commercialised technology, with sensitivity in the near to midinfrared region, with uses in chemical sensing and heat detection.

The principle of the QWIP is the photoexcitation of electrons from the first subband into the *continuum* of states above the barrier energy height, or alternatively into the adjacent well. These electrons then flow under bias to the collector layer which is of the same material as the well. The ISBTs in a QWIP can be categorised into three groups depending on the position of the upper excited state. Bound to bound, bound to continuum and bound to quasi-bound, the choice of which affects the photon frequency bandwidth and dark current characteristics of the detector.

QWIPs are generally grown with thick barriers to prevent tunnelling between wells and with many QW repeats, sometimes in excess of 100. The NRC sample studied in this thesis was designed as a bound to quasi-bound QWIP, though it was never used in this capacity.

Quantum cascade laser

The ISBT as a light emitter was theoretically proposed as early as 1971 [13], and emission from ISBTs was seen in the Si-MOSFET inversion layer system in 1976 [14]. However, precise control of laser level scattering rates was not achievable without the precision afforded by MBE. Intensive research into a GaAs/AlGaAs quantum well emitter resulted in the observation of intersubband emission in 1988 [15, 16]. It was not until 1994, 25 years after the original proposal, that ISBT lasing was demonstrated, through the *quantum cascade laser* (QCL) [17]. Like many other proposals of ISBT lasers at the time, the basic idea was to have a repeating staircase-type band structure design, in which an electron would emit a photon in an active, lasing region, then tunnel through to the next lasing region to repeat the process, cascading down the staircase. See Fig. 1.5.

The significant difference to the 1994 QCL, however, was the control of population inversion. Population inversion is not inherent to the ISBT system, with all subbands above the optical phonon emission energy sharing roughly the same scattering time. In order to lengthen the non-radiative scattering time from the upper laser level to the lower laser level, the band structure was engineered such that the states of the two subband levels had reduced spatial overlap, requiring a large momentum transfer in order to undergo transition, thereby lengthening the scattering time of the upper laser level. Subsequent scattering time out of the lower laser level was then of the order of an ordinary fast phonon emission, and the subsequent tunnelling to the next lasing region, faster still.

The QCL is a significant implementation of the ISBT, and remains a lucrative and intensive field of study. The QCL has enabled laser emission throughout the region from the near ($\lambda \sim 3 \mu\text{m}$) [18] to the far infrared, terahertz (THz) region ($\lambda \sim 300 \mu\text{m}$) [19]. Thanks to band structure engineering it is possible to virtually arbitrarily choose the wavelength of emission. Commercial QCLs are widely available which have continuous tunability in the midinfrared, operating at room temperature; indeed QCLs feature in this current thesis, acting as the spectroscopic light sources.

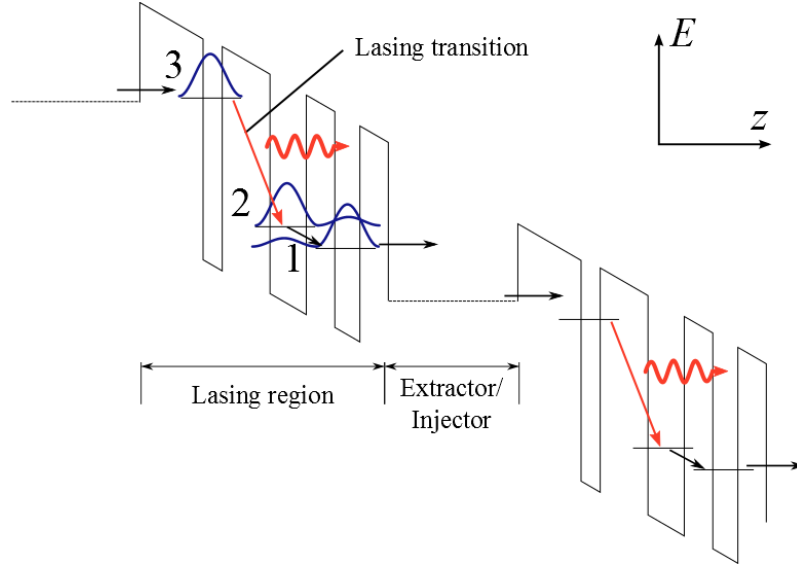


Figure 1.5.: The staircase bandstructure of the QCL when under appropriate bias. The lasing transition is between levels 3 and 2. Low spatial overlap of these two levels ensures a longer scattering time for level 3 allowing population inversion to build up. The electron is extracted from the lower lasing level via phonon scattering, and then tunnels into the extractor/injector region whereafter it is injected into the next lasing region. Single electrons repeat the lasing process for all the periods of the structure; there are typically ~ 30 repeats. See Ref. [17].

Work to bring THz QCLs to room temperature operation is ongoing; since THz photon energies are of the order of typical phonon energies, they are hampered by the self-heating mechanism which makes population inversion difficult to achieve at room temperature. One possible way to achieve THz emission from an ISBT is to exploit the properties of the cavity polariton (see sec. 1.4.2).

1.2. Light-matter interactions in semiconductors

Light may couple with the excitations in semiconductors and was first treated theoretically with respect to the interaction between light and the exciton. This leads to the concept of the quasi-particle known as the polariton.

1.2.1. Physical picture of the exciton

During an interband excitation an electron is excited from the valence band into the conduction band forming an electron-hole pair. Due to the mutual Coulom-

bic attraction between the electron and the hole, a bound state may be created, which moves through the semiconductor crystal as a single quasi-particle, the exciton. Since the exciton is a bound state, it is longlived, lasting from hundreds of picoseconds, to nanoseconds [20], so the absorption profile is far sharper than that of a plain interband transition. Since the effective mass of the hole is greater than that of the electron, the exciton is similar to a hydrogen atom, with the electron approximately orbiting the hole with an associated Bohr radius and binding energy³ [20]. See Fig.1.6. Note that excitons also exist in bulk semiconductors, not exclusively in QWs.

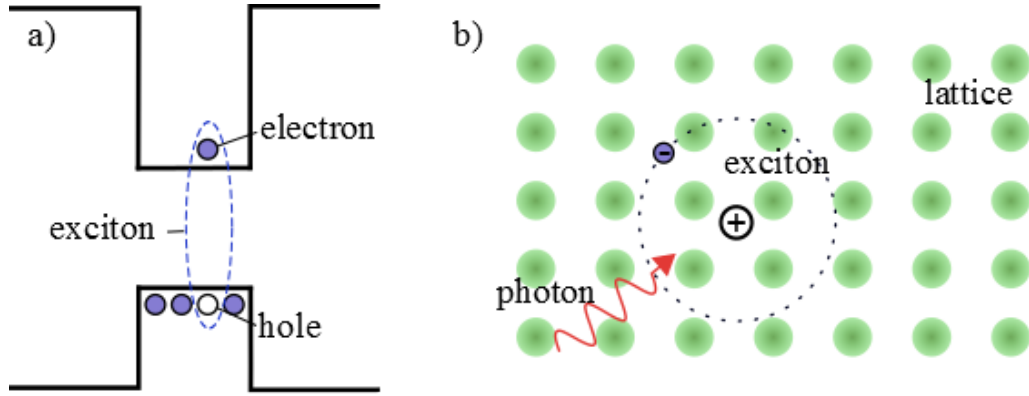


Figure 1.6.: a) The formation of an exciton from a bound electron and hole in a QW. b) In real space, the exciton is like a hydrogen atom of reduced mass in the crystal lattice, which may interact with incident light.

1.2.2. Interaction of light with exciton - the exciton polariton

The interaction of the exciton with light was analysed by Hopfield in 1958, treating the interaction of the radiation field with the polarisation of an exciton, with a quantum electrodynamic description. What resulted was the prediction of exciton energy mode-splitting and the emergence of the mixed light-matter eigenstate now known as the *exciton polariton* [21].

The energy in the light-matter interaction is exchanged periodically between photons and the exciton at a frequency known as the Rabi frequency, Ω_R (Fig.1.7, a)). If the Rabi frequency is rapid enough - in a sufficiently intense, resonant field - it is

³Specifically in reference to the Wannier-Mott exciton, rather than the smaller-radius Frenkel exciton.

possible to split the exciton energy levels (Fig. 1.7, b))⁴. The amount of splitting of the exciton excited state is equal to the Rabi frequency. The split states are new quasi-particles, exciton polaritons - their eigenstates are composed of the light and matter's uncoupled eigenstates, but unfactorisable. Exciton polaritons were seen in fluorescence spectra of GaAs as early as 1977 [23].

In solid state physics the general term *polariton* refers to a quasi-particle of strong coupling of light and matter. As such it is also used in the description of the interaction of light and the ISBT.

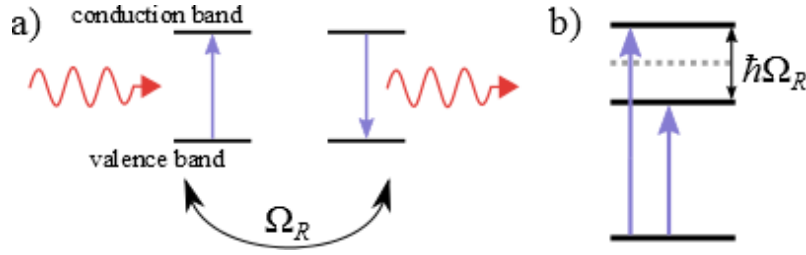


Figure 1.7.: Formation of the exciton polariton. a) An electron absorbs a photon thus creating an exciton. The exciton de-excites, emitting a photon, before another photon re-excites the electron. The energy in the system cycles between excitonic and photonic form at the Rabi frequency, Ω_R . b) At a sufficiently high Rabi frequency the excitation becomes *dressed*, with the exciton energy level splitting from the original resonant energy by $\pm\frac{1}{2}\hbar\Omega_R$. The new energy levels represent mixed light-matter eigenstates called exciton polaritons.

1.2.3. Interaction of light with the ISBT

The parallel dispersion curves of subbands, in principle, give the ISBTs a naturally sharp resonance and, with the improvement in growth purity, has come the ability of the ISBT to mimic the transitions that occur in excitons and in atomic vapours. The linewidth of the ISBT needs to be sufficiently narrow, in order to see the subtle effects of the light-matter interaction. Fano interference [24], ac Stark splitting [25], and gain without inversion [26], have all been demonstrated in ISBT systems.

Rabi splitting of ISBT states has been demonstrated in an intense field, corresponding to the formation of ISBT polaritons [26]. However, lasers providing the requisite intense fields are not commonplace in the wavelength region of the ISBT (mid to

⁴In quantum optics, the split states are said to be *dressed* (i.e. dressed by photons) [22].

far-infrared). Alternatively, the light-matter interaction with the ISBT can be studied without an intense field, by instead coupling the ISBT with the quantised field of an optical cavity, in the field of *cavity quantum electrodynamics*.

1.3. Cavity quantum electrodynamics

Cavity quantum electrodynamics (cQED) is the study of the interaction of a quantised cavity field with excited states of matter. This section introduces the quantised field, some cQED phenomena, and the cavity polariton.

1.3.1. Quantisation of the electromagnetic field

When the dimensions of an optical cavity are of the same scale as a wavelength of the confined radiation, quantisation of the field occurs, with only half-integer multiples of wavelength (resolved in the confinement direction) permitted within the cavity. A simple case is when confinement is in just one dimension, as in Fabry-Perot resonators with parallel mirrors. See Fig. 1.8. Should the radiation be at an oblique angle to the mirrors, then it will flow parallel to the mirror surfaces, the mirrors thereby acting as a *slab waveguide*. At a given angle, only the resonant modes will be permitted to flow within the waveguide: the waveguide modes are quantised. (See sec. 2.3.)

Microcavities

To create mode quantisation on a scale of an optical or infrared wavelength requires the fabrication of cavities on the scale of microns. Cavities of this size, when containing semiconductor material, are called *microcavities*.

When microcavities contain within them MBE-grown semiconductor, the mirrors of the microcavity might themselves be grown in the same process to ensure high precision and optical quality. Typically, distributed Bragg reflectors are used, formed from multiple layers of semiconductor.

Metallic Fabry-Perot reflectors may also be grown with ease on top of an MQW. Alternatively, sufficiently n-doped semiconductor will confine and guide radiation

propagating at an internal angle, via total internal reflection; in this case it would be a *waveguide microcavity*.

Other microcavity schemes range from the fairly conventional, e.g. diffraction grating cavity mirrors, implemented to some success in THz microcavities; to micro-toroidal resonators for ultra high Q with whispering-gallery modes [27]. See also sec. 1.4.4.2.

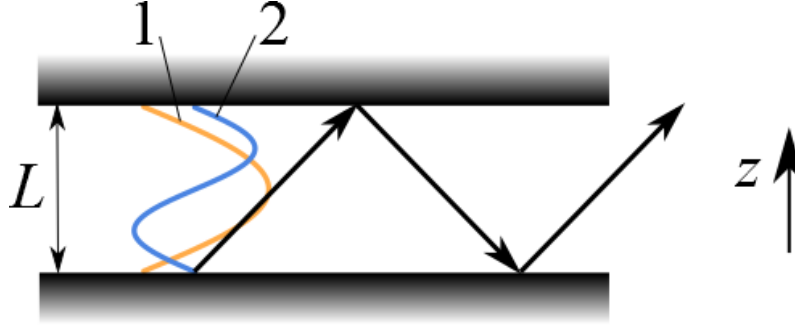


Figure 1.8.: The quantisation of the electromagnetic field in a cavity of similar dimensions to the light's wavelength. Only wavelengths resonant (when resolved in the direction of confinement) with the cavity length L can exist in the cavity. Non-resonant wavelengths will be suppressed in the cavity. A cavity may be used as a waveguide for resonant wavelengths, which in the ray picture corresponds to the light reflecting multiple times from the mirrors with the effect of travelling parallel to the mirrors. Shown are first- and second-order modes, 1 and 2.

1.3.2. cQED phenomena

The quantisation of the electromagnetic field by a cavity affects the behaviour of optically-emitting processes within. Here are presented some cQED phenomena, categorised according to the strength of the coupling between cavity field and matter. The optically-emitting process is, for now, treated as a general entity, i.e. any optically-active electronic transition.

1.3.2.1. Weak coupling of light and matter

Modification of spontaneous emission - The Purcell effect

Quantisation of the electromagnetic field modifies the spontaneous emission properties of optical transitions within the cavity. Spontaneous emission is akin to stimulated emission, but the field which couples to the emitting transition is the vacuum

field. When a cavity quantises the vacuum field, spontaneous emission will be enhanced for optical transitions resonant with the cavity, and suppressed for transitions away from resonance. This effect is known as the Purcell effect, discovered in 1946, heralding the beginning of cQED [28].

The Purcell effect is an example of a weak coupling phenomenon. The first treatment of spontaneous emission enhancement, by Purcell [29], regarded transitions in nuclear magnetic resonance, with spontaneous emission probability enhanced by a factor proportional to $Q\lambda_0^3/V$ where Q is the resonator quality factor, λ_0 the resonant wavelength of the cavity, and V the volume of the resonator. For the effect to be reasonable the volume of the cavity needs to be on the same scale as the resonant wavelength, $V \approx \lambda_0^3$, in which case the spontaneous emission enhancement is roughly equal to the resonator quality factor, Q , which can vary from unity to the order of 10^3 and beyond. This enhancement originates from the increase in mode density within the cavity, which increases the Einstein coefficient for spontaneous emission, A_{21} .

Demonstration of enhanced spontaneous emission occurred as early as 1983, when Goy *et al.* reduced the emission lifetime of a single sodium Rydberg atom in a high- Q superconducting cavity [30]. The Purcell effect can also be responsible for the inhibition of spontaneous emission if the optically emitting transition is not resonant with the cavity frequency. This was demonstrated in 1968 where fluorescence decay lifetime of a molecule was increased when placed by a mirror, due to the standing wave pattern of the vacuum field formed between the molecule and the mirror⁵ [32].

The suppression and enhancement of spontaneous emission was demonstrated in solid state systems at the end of the 1980s by Yablonovitch *et al.* in a GaAs/AlGaAs double heterostructure [33]. A GaAs layer of a thickness on the order of the GaAs bandgap wavelength, sandwiched between AlGaAs layers (which acted as a cavity) exhibited a modified electron-hole recombination rate⁶.

The Purcell effect is not thought to enhance emission by more than a factor of 1.5 in

⁵The cavity field is also capable of affecting the emission of virtual photons from zero-point fluctuations. The well known Casimir effect is such an outcome of quantisation of the vacuum field. Between two closely separated plates, with a vacuum between them, only resonant virtual photons may exist, whereas outside the plates the vacuum field is continuous; this exerts an attractive force between the two plates [31].

⁶Whether the recombination was enhanced or inhibited depended on the number of carriers injected into the GaAs, which brought the interband resonance in and out of the photonic band-gap.

planar microcavities with bulk dielectric therewithin [34, 35], unless the oscillating dipole of the emitting transition is perpendicular to the plane (as is the case for ISBTs), in which case the Purcell factor scales as $Q\lambda_0/L_{MC}$ where L_{MC} is the thickness of the microcavity. This has led to observations of spontaneous emission enhancement in ISBT QCL microcavities [36].

1.3.2.2. Strong coupling regime and the vacuum Rabi frequency

Beyond the weak coupling of optical transitions and cavity modes, is the strong coupling regime (SCR). If the light-matter interaction is strong enough, the interacting photon will be absorbed, emitted and reflected multiple times by the optical transition and cavity before it is lost to the continuum through the mirrors. The energy oscillates between the electronic and photonic form at a frequency known as the *vacuum Rabi frequency* (here denoted $\Omega_{R,k_{\parallel,res}}$, occurring at the resonant wavevector, $k_{\parallel,res}$). This is similar to the Rabi frequency of dressed atomic states in quantum optics [37] and in exciton polaritons (see sec. 1.2.2), however, this splitting does not require an intense field and can occur in the cavity vacuum field.

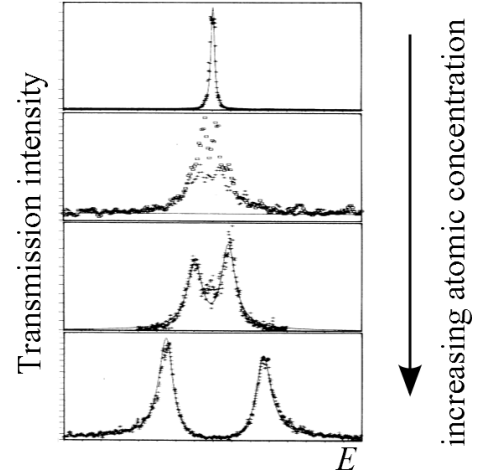
The condition for being in SCR is if this splitting is resolvable in spectra (see sec. 2.4.2). This splitting was seen in atom-cavity experiments in 1983 through superradiance ringing [38], and later in the 1980s, through transmission spectra [39] (see Fig. 1.9); in both cases, the observations were seen from sodium atoms beamed through a Fabry-Perot cavity. The mode splitting of single atoms is not substantial, but, fortunately, the splitting was found to scale with the number of atoms in the cavity, i.e. coupling strength is enhanced by increasing the number of individual oscillators within the cavity.

In terms of dispersion relations, this splitting will appear as an *anticrossing* (or avoided crossing) of the resonant cavity mode and the electronic transition mode. Plotted against in-plane wavevector⁷, $k_{\parallel}$, when the cavity mode is tuned towards resonance with the cavity mode, rather than cross and become degenerate in energy, the modes will anticross, thereby forming new modes⁸. The vacuum Rabi frequency dictates the minimum energy separation of these new modes. See Fig. 1.10. These modes represent new eigenstates of the system, and in solid state physics are re-

⁷That is, in the plane of the MQW and cavity, with direction (x, y) .

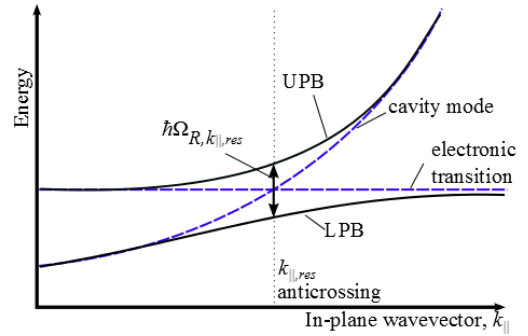
⁸Akin to the phenomenon of level repulsion, and similar to the case of classical coupled oscillators [40].

Figure 1.9.: The splitting of the transmission mode of an optical cavity due to the introduction of atoms coupled with the cavity mode, with increasing atomic concentration from top to bottom. Reprinted figure with permission from Ref. [39]. Copyright (1989) by the American Physical Society.



ferred to as *cavity polaritons*, comprising an upper polariton branch (UPB) and a lower polariton branch (LPB). The photon and electron, interacting coherently for a meaningful amount of time, form a mixed light-matter eigenstate at frequencies split from the resonant frequency. See Fig.1.11, and sec.2.4 for theoretical treatment.

Figure 1.10.: An electronic transition and a cavity mode (dashed lines) undergo anticrossing where the two uncoupled modes are resonant, at an in-plane wavevector of $k_{\parallel, res}$. The new modes form the UPB and LPB (solid lines). The minimum separation, at the anticrossing, is the vacuum Rabi frequency, $\Omega_{R, k_{\parallel, res}}$.



1.3.3. Exciton cavity polariton

The first observation of strong coupling in a quantum well cavity, and the corresponding *exciton cavity polariton*, was in 1992 when splitting of the energy mode of an exciton was observed by Weisbuch *et al.* in GaAs [41]. Fig.1.12 shows the anticrossing of modes of this work seen in reflectivity spectra. This first observation

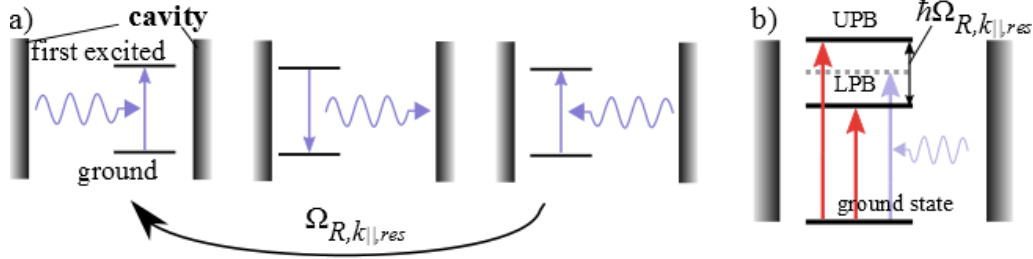
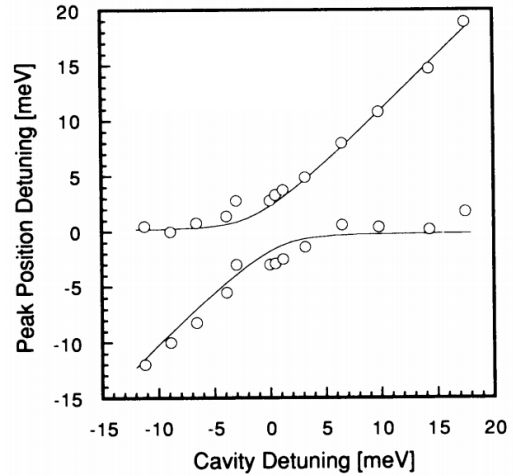


Figure 1.11.: Coupling between a cavity mode and an optically-active electronic transition. a) An initial photon excites the transition. The photon is then re-emitted into the cavity, where it is resonant. The photon then interacts again with the transition. The energy in the system cycles between photonic and electronic form at a frequency known as the vacuum Rabi frequency, $\Omega_{R,k_{||,res}}$. b) As with the case of excitons in a strong field, the energy levels split by the magnitude of $\Omega_{R,k_{||,res}}$. In semiconductors, the mixed photonic and electronic states become *cavity polaritons*.

of the cavity polariton was in an MQW sample, rather than in bulk semiconductor, due to the greater density of states and oscillator strength of MQW electrons, compared to bulk excitons, leading to a stronger coupling. Note that it was necessary for the cavity polariton to be composed of an exciton rather than a plain interband transition, thanks to the increased sharpness of the former [42].

Figure 1.12.: First measurement of the anticrossing of the cavity mode and exciton mode peaks in an MQW microcavity. The cavity mode was tuned by selecting different parts of a wedged microcavity containing an MQW. This was the first observation of a cavity polariton. Reprinted figure with permission from Ref. [41]. Copyright (1992) by the American Physical Society.



1.4. A decade of intersubband cavity polaritons

The ISBT of an MQW, like the exciton, is able to couple to a suitable resonant mode of a microcavity. The first observations of the *intersubband cavity polariton* (ICP) came in 2003, from two groups, Dini *et al.* [43] (see Fig. 1.13), and Dupont *et al.* [44], our collaborators.

We focus, in this section, on the wider relevance of ICPs, and their possible applications. Theoretical treatment of ICPs will be found in sec. 2.4.

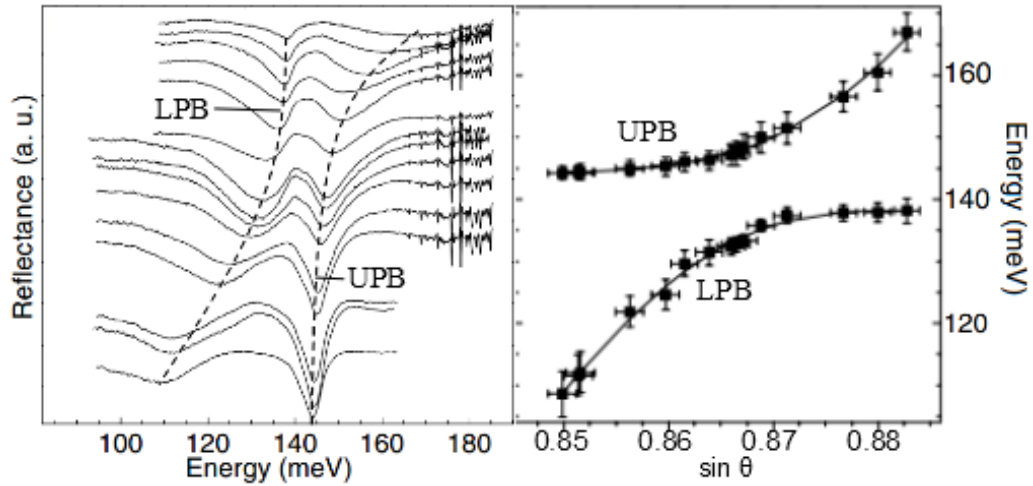


Figure 1.13.: First measurement of the strong coupling of ISBT and microcavity mode, as reported by Dini *et al.* [43]. (Left) Reflection spectra, with dips corresponding to the polariton branches. These were plotted as dispersion curves (right) showing the anticrossing, plotted against the sine of the coupling angle into the microcavity, which is proportional to the in-plane wavevector of the radiation. Reprinted figure with permission from [43]. Copyright (2003) by the American Physical Society.

1.4.1. New avenues opened by the ICP

Though similar to the exciton cavity polariton system, the ICP differs in a number of ways making it a valuable system to explore in its own right. The optically active region of ISBTs, as detailed, ranges from the infrared to the THz region of the spectrum. The band gap engineering and atomic-like transition of the ISBT essentially permits the creation of designer atoms giving many degrees of control to the experimenter. This generates interest in the design of devices including coherent

light emitters, and, in terms of fundamental explorations, the ICP offers new grounds to expand upon the successes of the exciton cavity polariton, and to discover new effects unique to the ICP.

1.4.2. Light emission from ICPs

One of the main applications of ISBTs is the QCL (sec. 1.1.3.3). These of course (being lasers) employ cavities, but not necessarily on the length scale of the emitting wavelength, and so the question of strong coupling is not relevant. Light emission from ICPs, on the other hand, offers different properties compared to the QCL system. For example, QCLs, in extending the radiative lifetime of the upper level (ten to a hundred nanoseconds) in order to induce population inversion, suffer from a poor radiative efficiency, due to the presence of faster nonradiative phenomena (e.g. LO phonon scattering, picoseconds). ICPs, on the other hand, have radiative lifetime properties which can be dominated by the vacuum Rabi oscillation rather than nonradiative events, leading to a greater efficiency [45]. ICP emitters offer the prospect of lower thresholds than QCLs, as well as lower operating temperatures, and emission at wavelengths otherwise difficult to achieve.

1.4.2.1. Quantum cascade electroluminescent ICP devices

Light emission from ICPs necessitates inherently different radiative lifetime properties of the subbands compared to within a QCL. QCLs require a population inversion to operate; however, a system with population inversion cannot achieve strong coupling since the ICP requires the exchange of energy between photonic and ISBT modes - during full population inversion a photon cannot be absorbed and is more likely to trigger stimulated emission; the ISBT becomes transparent and no ICP can be formed. Hence it is intrinsically difficult to have polaritons lasing in the traditional sense [46].

To that end, devices have been designed [47] and demonstrated [45] incorporating the staircase principle of the QCL, but with the relaxation lifetime of the lower subband *longer* than the radiative lifetime of the excited subband. Injector states inject electrons into the quantum well at the ICP energy (whether at upper or lower polariton energy is through design). The electron undergoes spontaneous emission, then strongly couples to the cavity to form the ICP. The favoured mode

of the spontaneous emission is that which conserves in-plane momentum in the microcavity; this mode will have a lateral propagation, and so can be coupled out of the microcavity via frustrated total internal reflection⁹. See Fig. 1.14.

The larger the vacuum Rabi splitting, the more efficient the injection of electrons into polariton states rather than the bare ISBT. Enhancement of spontaneous emission is also thanks to a Purcell-type effect [48].

Tunability of the output radiation wavelength may also be achieved in this scheme since the dispersion curve of the ICP has a variable energy. By varying the voltage across the cascade structure the electron injection level itself varies, and the energy of the injected electron determines the energy of the ICP. In this way, tunability of $\sim 20\%$ can be achieved from such a device [49]. See Fig. 1.14.

Recently, a room temperature THz electroluminescent device has been reported based on ICPs¹⁰ [50]. THz QCLs, on the other hand, currently suffer from thermal issues, and are not yet able to reach room temperature operation.

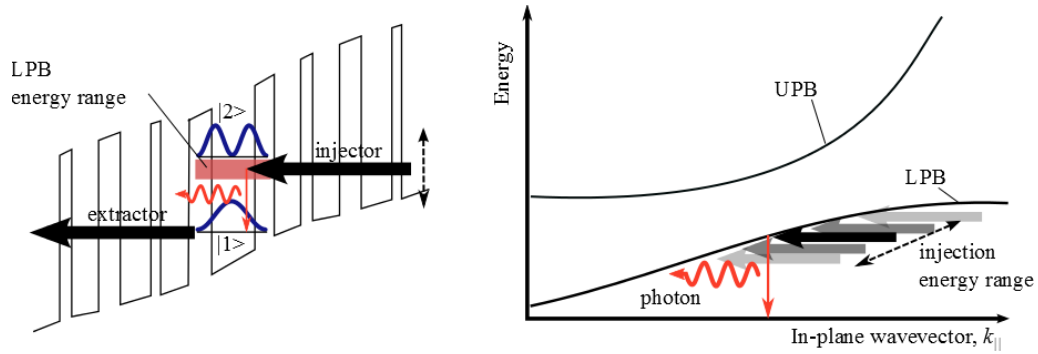


Figure 1.14.: Cascade structure (left) of an electroluminescent ICP device. The electron injection energy can be chosen by the applied voltage, and so can create ICPs in a range of energies (dashed arrow) in accordance with the position on the ICP (here, the LPB) dispersion curve (right).

1.4.2.2. Other ICP light emission schemes

Further schemes have been proposed for ICP-based light emission, such as phonon-assisted stimulated scattering in a scheme similar to resonant difference frequency

⁹See sec. 2.3.1.2.

¹⁰This particular electroluminescent device employs parabolic QWs, which have several subbands equally spaced in energy. Hence when the temperature of the device raises, and higher subbands become populated, the active ISBT will still have the same transition energy, and the ICP will be unchanged.

generation in nonlinear optics: a resonant pump laser populates the upper polariton state, which then undergoes stimulated LO phonon scattering into the lower polariton state whereupon a ‘signal’ photon is emitted [51]. The signal photons emitted stimulate further interactions and emissions of signal photons, creating laser-like emission, without population inversion. See Fig. 1.15, a).

Another scheme proposed would be for stimulated photon emission from upper to lower polariton [52]. Usually a forbidden interaction, this is not the case for quantum wells asymmetric in z . Upper and lower polariton branches are separated typically by energies corresponding to photons in the THz frequency range; this scheme therefore offers another alternative to the THz emitter.

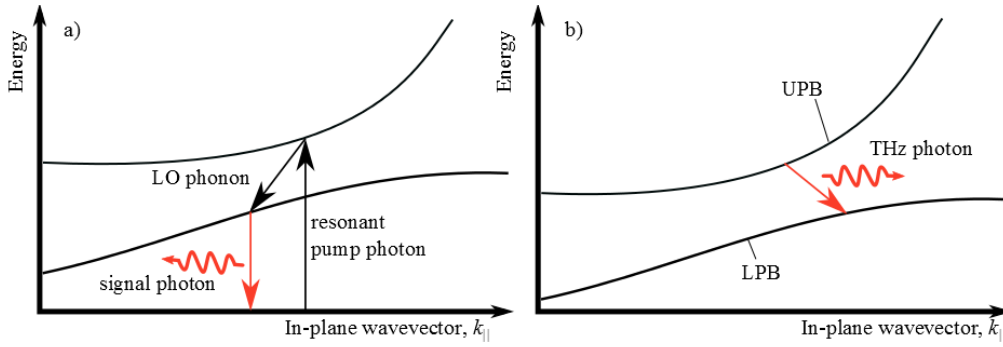


Figure 1.15.: Schemes for ICP light emission. a) Pump photon resonant with the upper polariton stimulates an emission of an LO phonon to the lower polariton which then emits a signal photon [51]. b) Asymmetric QWs allow the transition from upper to lower polariton branch, typically emitting THz photons [52].

1.4.3. Ultrastrong coupling and correlated photons

An additional coupling regime to those mentioned in sec. 1.3.2 is the *ultrastrong coupling regime* (UCR). This is when the vacuum Rabi energy is of the order of the optical transition energy, and further terms must be included in the Hamiltonian of the polariton [21, 53]. It is possible to reach the UCR at longer-wavelength ICPs, since these have wider well layers, which therefore create a larger dipole. The larger resultant vacuum Rabi splitting invokes additional, *anti-resonant* Hopfield terms in the quantum description of the polariton state (see Ref. [53] for details). Due to these extra terms, the ground state is no longer the ordinary vacuum, but rather a *squeezed state* consisting of virtual photons. These virtual photons cannot escape the vacuum unless the Rabi frequency is non-adiabatically switched to a value close

to zero where the ground state becomes the ordinary vacuum. At this point the virtual photons will be ‘unbound’, and emitted¹¹ [55, 56, 57]. The emitted photons will be entangled pairs, correlated in frequency, with practical applications in, for example, quantum communications. The required non-adiabatic change of vacuum state must occur on sub-cycle time scales, i.e. before the polariton has a chance to dissipate (tens of femtoseconds) [58, 59, 60].

1.4.4. Future of ICPs

As the ICP enters its second decade, we take a look at some possible avenues of development.

1.4.4.1. Exciton cavity polariton phenomena

Many potentially interesting applications of the ICP depend on whether the ISBT can be approximated as a boson. The exciton is an approximate boson since it is a bound state between two fermions, a hole and an electron. ISBTs on the other hand do not represent a bound state, but are still composed of an excited electron and a hole in the Fermi sea of lower subband electrons, and so can still be described as composite bosons [51] - at least up until reasonable upper subband carrier densities [61].

Should the boson description hold, then theoretically the same avenues taken by the exciton cavity polariton system should be accessible to the ICP. For example, the creation of polariton Bose-Einstein condensates, demonstrated in 2006 [62]. There, polaritons in the lower branch scatter downwards towards the energy minimum, at the point of zero momentum, to achieve massive ground state population.

Despite being bosons themselves, plain excitons cannot form condensates; but, by coupling to light, the polariton has a small effective mass and therefore a large spatial coherence, increasing probability of wavefunction overlap and condensation.

It is expected that ICPs, too, will demonstrate effects due to the increased spatial coherence associated with the polariton state.

¹¹This effect is similar to the so-called Dynamical Casimir Effect, the modulated version of the Casimir effect. Relativistic oscillation of the mirrors creates an emission of the virtual photons into measurable photons, as recently demonstrated in Ref. [54].

1.4.4.2. ICPs in other systems

ISBTs are not exclusive to MQWs, and therefore ICPs can equally occur in other systems. The quantum dot, and dot in well concepts confine the electron in more than one direction and are possible candidates for development of ICPs beyond the MQW.

Further variations on the ICP come from varying the form of the confining cavity. ISBT polaritons have been reported in metallic photonic crystals, metallic gratings where the quantised plasmon waves of the grating strongly couple with MQWs to form polaritons in geometries more accessible to pump and probe lasers [60].

Inductor-capacitor (LC)-type ‘microresonators’ have also been employed in order to enhance the electric field localisation and therefore the strong coupling [50], especially in the THz region. Furthermore, *metamaterials* have recently been combined with ICPs, with a resonant cavity designed such that all its dimensions (rather than just in the z -direction) are sub-wavelength, a so called ‘nanocavity’ [63]. Further innovations in cavity design and ISBT implementation will be necessary to fulfill the potential of ICPs.

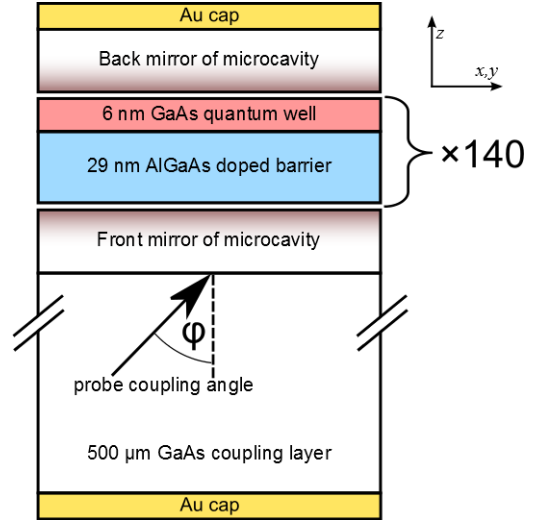
1.5. Motivation, my work and the structure of this Part

The subject of the thesis is the absorbance properties of the ICP. Here, the NRC sample to be studied and objectives of the work are briefly summarised.

1.5.1. Sample to be studied in this thesis

The NRC sample to be studied in this thesis is an MQW within a waveguide microcavity, in which the ISBT of the MQW is strongly coupled with the fundamental mode of the microcavity, thereby forming ICPs. The MQW is made up of 140 repeats of a GaAs/AlGaAs quantum well, in which the sole ISBT is a bound to quasi-bound transition; that is, the upper level is close to the top of the well barrier potential.

Figure 1.16.: Schematic of the NRC sample. 140 periods of a GaAs/AlGaAs QW sit within a waveguide microcavity. Probe radiation is coupled into the NRC sample from the substrate layer via frustrated total internal reflection. The NRC sample is capped top and bottom with gold to aid waveguiding of radiation. (A more detailed version of this diagram is shown in Fig. 3.1.)



1.5.2. Object of work and motivation

The object of the thesis is to investigate the linewidths of ICPs by laser spectroscopy, paying particular attention to the linewidths at the resonance, when the ICP is equal parts matter and light. Low temperature measurements also represent the first cryogenic studies of ICP linewidths.

The motivation is to gain further insight into the properties of the ICP. Being a relatively new field, it is crucial to understand the linewidth (and therefore lifetime) and other properties of these quasi-particles. This is important for the development of ICP-based technology, in particular the new field of ICP light emitters, where sub-band lifetimes, and sharpness of emission linewidths play critical roles. Additionally, the ICP may yield further technologies, for example in infrared detection.

From a fundamental point of view, studies of light-matter interactions alone are significant enough to warrant the investigation of the ICP here.

1.5.3. The NRC sample in the literature

The NRC sample studied in this thesis has previously appeared in the literature under the name of ‘NT3557’. The first time was in 2003, in a paper from our collaborators Dupont *et al.*, at the National Research Council of Canada [44], which was one of the first two reports of the observation of ICPs. The NRC sample was one of four studied in the paper which displayed the strong coupling of ISBT

and waveguide cavity modes. The NRC sample was also studied previously in this group as part of the thesis of Mary Matthews [64]. There, Matthews performed laser spectroscopy with a frequency-converted midinfrared light source; however, the inherent noise and difficulty of using that laser system hindered the acquisition of a large set of data, ultimately halting progress. The major finding of Matthews' work was that ICP linewidths were narrower than predicted by the standard model describing ICP linewidths in the SCR. The aim of this thesis is to continue that work with a more stable laser source, and investigate the linewidths further, and establish a description of the ICP's behaviour.

More recently (2013), Załużny and Zietkowski [65] have performed a detailed semi-classical theoretical study of a sample based on the NRC sample (denoted there as 'Structure $\mathcal{B}$ '). Their analytical treatment is referred to in sec. 2.4.

2. Theory of intersubband cavity polaritons

This chapter gives a theoretical treatment of the ICP. Beginning with the mathematical description of the modes of MQWs and microcavity waveguides, the chapter continues with a treatment of the ICP and the absorption linewidth broadening mechanisms affecting the ICP.

2.1. Theory of the multi-quantum well

We begin with the theory of the MQW, and the broadening mechanisms of the ISBT absorption linewidth.

2.1.1. Subbands of the quantum well conduction band

The total wavefunction of an electron in the conduction band of a QW can be described as a combination of the periodicity of the semiconductor crystal, the free electron motion in the plane of growth (x, y) , and the electron confinement in the direction of growth (z -direction):

$$\psi(\mathbf{r}) = u_\nu(\mathbf{r})f_{\mathbf{k}_\parallel}(x, y)\varphi_n(z), \quad (2.1)$$

where $u_\nu(\mathbf{r})$ is the Bloch function of band ν (which we assume that the quantum well has been appropriately doped such that only the conduction band is optically active). The envelope function in the plane of the quantum well is the form of a plane wave $f_{\mathbf{k}_\parallel}(x, y) = \frac{1}{\sqrt{S}}e^{i\mathbf{k}_\parallel \cdot \mathbf{r}}$, with $\mathbf{k}_\parallel = (k_x, k_y)$ the in-plane wavevector, and S

the sample area. The behaviour of the wavefunction in the z -direction is described by the function $\varphi_n(z)$, with n the associated quantum number. When the Bloch function is assumed the same in all constituent materials, Schrödinger's equation describing the system is

$$-\frac{\hbar^2}{2m}\nabla^2[f_{\mathbf{k}_{\parallel}}\varphi_n] + V(\mathbf{r})[f_{\mathbf{k}_{\parallel}}\varphi_n] = E_{n,\mathbf{k}_{\parallel}}[f_{\mathbf{k}_{\parallel}}\varphi_n], \quad (2.2)$$

where $V(\mathbf{r})$ is the well and barrier potential, and m^* is the effective mass of the electron. The effective mass is a mathematical way to encompass the perturbation of the $\mathbf{k} \cdot \mathbf{p}$ term from application of the Hamiltonian to equation 2.1. The use of an effective mass relates to how accurate the final energy level calculation will be. Furthermore, m^* is anisotropic to an extent, and depends on the parabolicity of the dispersion curve.

When the z -varying parts are separated from the (x, y) -varying parts, Schrödinger's equation may be derived for the electron in the quantum well:

$$-\frac{\hbar^2}{2m^*}\frac{d^2\varphi_n}{dz^2} + V(z)\varphi_n = E_n\varphi_n. \quad (2.3)$$

This equation has well known solutions for the square potential well, and can be solved easily for the case of the infinite potential well. Simultaneously, the component of the wavefunction moving in the plane of growth, $f_{\mathbf{k}_{\parallel}}$, has the energy of a free electron, but with an effective mass, and is described by a parabolic dispersion curve at low energies. Therefore, the energies of the electron in the (infinite) potential well can be described with

$$E_{n,\mathbf{k}_{\parallel}}^{\infty} = E_n + E_{\mathbf{k}_{\parallel}} = \frac{n^2\pi^2\hbar^2}{2m^*L_{QW}^2} + \frac{\hbar^2k_{\parallel}^2}{2m^*}, \quad (2.4)$$

where L_{QW} is the thickness of the well and n is the integer quantum number denoting the level of the electron (see Fig. 2.1). The term 'subband' is used because the in-plane energy is not discrete, in that, because the in-plane momentum is free to vary continuously at each n , so too can the energy. Despite this dispersion of energies,

transitions between subbands (ISBTs) remain quantised since the dispersion curves are parallel, and $k_{\parallel}$ is conserved during the transition. So the energy of the transition $|n'\rangle \rightarrow |n\rangle$ is

$$E_{n'n}^{\infty} = \frac{\pi^2 \hbar^2}{2m^* L_{QW}^2} (n'^2 - n^2). \quad (2.5)$$

(Deviations from this ideal case of ISBT energy are given in sec. 2.1.4.)

The electron wavefunction in the z -direction of an infinite potential well (with well extent from $z = 0$ to $z = L_{QW}$) is

$$\varphi_n(z) = \sqrt{\frac{2}{L_{QW}}} \sin\left(\frac{n\pi z}{L_{QW}}\right). \quad (2.6)$$

For the case of the finite square well the subband energies are reduced, and the wavefunctions tunnel (measurably) some way into the barrier layers. The most important aspect of the QW in terms of this thesis is the ISBT energy, rather than the exact form of the electron wavefunction. The ISBT energy may be calculated without knowledge of the wavefunction, so no further development beyond equation 2.6 will be given for the z -direction wavefunction.

2.1.2. Intersubband transitions

2.1.2.1. Fermi's golden rule for ISBTs

The transition rate of the interaction with an incident field can be described with Fermi's golden rule:

$$W_{if} = \frac{2\pi}{\hbar} \left| \left\langle \psi_i \left| H_I \right| \psi_f \right\rangle \right|^2 d(\omega, \omega_{fi}) \quad (2.7)$$

where H_I is the interaction Hamiltonian, which, in the dipole approximation, is $H_I = -(e/m^*)\mathbf{A} \cdot \mathbf{p}$. $\mathbf{A}$ is the vector potential, $\mathbf{p}$ the momentum operator, e the

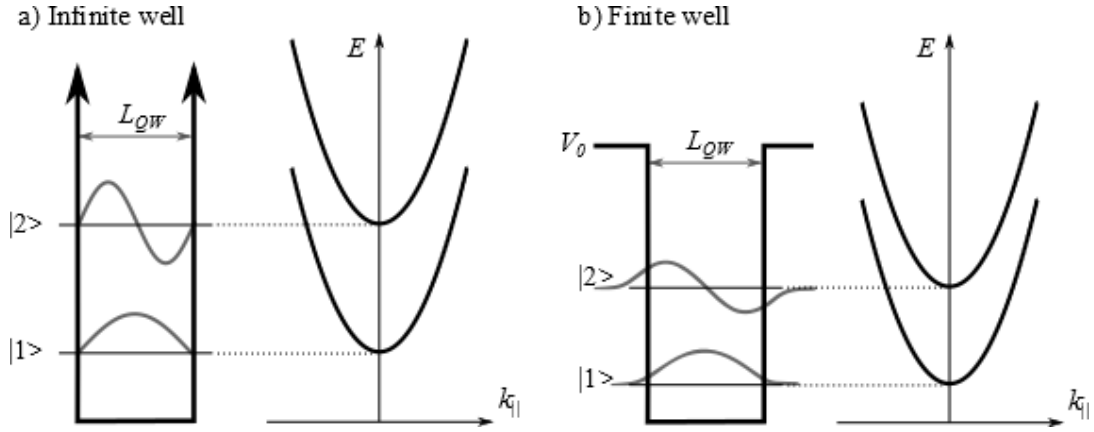


Figure 2.1.: a) The first two levels of an infinite well, of width L_{QW} , and the associated electron wavefunctions. The electron is confined in just one dimension leading to electron energies forming subbands rather than discrete levels, as shown on the right. b) The same width well, but with finite barrier, height V_0 , where electron wavefunctions tunnel some way into the barrier. Energy subbands are shifted downwards in energy leading to a smaller $|2\rangle \rightarrow |1\rangle$ transition energy.

charge of the electron, and ω the angular frequency of incident radiation. $d(\omega, \omega_{fi})$ describes the final density of states of the transition, and to a first approximation may be taken as a delta function positioned at the transition frequency, ω_{fi} . (Its actual form will be introduced in sec. 2.1.3.)

By substituting the appropriate relation between the vector potential, $\mathbf{A}$, and the electric field, $\mathbf{E}$, equation 2.7 becomes

$$W_{if} = \frac{2\pi}{\hbar} \frac{e^2 |E_0|^2}{4m^* \omega^2} \left| \left\langle \psi_i \left| \mathbf{e} \cdot \mathbf{p} \right| \psi_f \right\rangle \right|^2 d(\omega, \omega_{fi}), \quad (2.8)$$

where E_0 is the magnitude of the electric field, and $\mathbf{e}$ the polarisation vector of $\mathbf{E}$.

2.1.2.2. ISBT selection rule

The matrix element of equation 2.8 may be evaluated by substituting equation 2.1 into it. Acknowledging that the Bloch function u_ν is unchanged when the interaction

is within one band (here, the conduction band) simplifies the matrix element to

$$\left\langle \psi_i \left| \mathbf{e} \cdot \mathbf{p} \right| \psi_f \right\rangle = \mathbf{e} \cdot \left\langle f_{\mathbf{k}_{\parallel}} \varphi_n \left| \mathbf{p} \right| f_{\mathbf{k}'_{\parallel}} \varphi_{n'} \right\rangle, \quad (2.9)$$

where n' is the index of the final subband and $\mathbf{k}'_{\parallel}$ the final in-plane wavevector. Evaluating this matrix element gives:

$$\begin{aligned} \mathbf{e} \cdot \left\langle f_{\mathbf{k}_{\parallel}} \varphi_n \left| \mathbf{p} \right| f_{\mathbf{k}'_{\parallel}} \varphi_{n'} \right\rangle &= \frac{1}{S} \int e^{-i\mathbf{k}_{\parallel} \cdot \mathbf{r}} \varphi_n^* [e_x p_x + e_y p_y + e_z p_z] e^{i\mathbf{k}'_{\parallel} \cdot \mathbf{r}} \varphi_{n'} d^3r \\ &= \frac{1}{S} \left((e_x \hbar k_x + e_y \hbar k_y) \delta_{n,n'} \delta_{k_{\parallel},k'_{\parallel}} + e_z \delta_{k_{\parallel},k'_{\parallel}} \int \varphi_n^* p_z \varphi_{n'} dz \right). \end{aligned} \quad (2.10)$$

Inspection of this equation shows that in order to undergo a transition from n' to n , the incident radiation must have non-zero z -polarisation. This expression also shows that transitions between states that match parity (e.g. $|1\rangle \rightarrow |3\rangle$) are dipole forbidden. These are the selection rules for the ISBT. The last term of equation 2.10 is the matrix element which determines the intersubband absorption: $\int \varphi_n^* p_z \varphi_{n'} dz = \langle n | p_z | n' \rangle$.

A commonly used quantity related to optical transitions is the dimensionless *oscillator strength*, which is used to compare the relative likelihood of a transition between subbands:

$$f_{nn'} = \frac{2}{m^* \hbar \omega_{n'n}} \left| \langle n | p_z | n' \rangle \right|^2, \quad (2.11)$$

with the property that $\sum_n f_{nn'} = 1$.

Typically, the $|1\rangle \rightarrow |2\rangle$ transition is the most important since usually only $|1\rangle$ is occupied; indeed in our system $|1\rangle$ and $|2\rangle$ are the only subbands that exist, so therefore $f_{12} \approx 1$.

2.1.3. Absorption coefficient for ISBTs

To calculate the absorbance properties of an MQW, the absorption of a single quantum well layer, α_{2D} , must be calculated. α_{2D} is a dimensionless quantity and is simply the fraction of electromagnetic energy absorbed by a single layer. To that end it is expressed as the ratio between absorbed intensity and incident intensity:

$$\alpha_{2D} = \frac{\hbar\omega N_s W_{if}}{\frac{1}{2}c\sqrt{\varepsilon_w} |E_0|^2}, \quad (2.12)$$

where N_s is the sheet number density of electrons and ε_w is the relative permittivity of the well material. Substituting the expression for W_{if} (equation 2.8), we get

$$\alpha_{2D}(\omega) = \frac{N_s}{2c\sqrt{\varepsilon_w}} \frac{e^2}{m^*} f_{12} \frac{\pi\gamma_{ISBT}/2}{(\omega_{21} - \omega)^2 + \gamma_{ISBT}^2/4} e_z^2 \quad (2.13)$$

where ω_{21} is the oscillator transition frequency of the ISBT. Additionally, in the derivation of this expression, the density of states $d(\omega, \omega_{21})$ has been replaced by a Lorentzian profile of FWHM linewidth γ_{ISBT} , and that $\omega \approx \omega_{21}$ (narrow linewidth approximation). This expression is for the case of $T = 0$ K; with increasing temperature, absorption broadens, and peak absorption drops.

The inclusion of a Lorentzian profile as the density of states is justified for the bare ISBT oscillation when it is thought of as a driven, damped harmonic oscillator, with a damping constant determining the decay of oscillations. The damping constant encompasses all the homogeneous contributions of broadening of the ISBT, the major ones of which will be introduced in sec. 2.2.

Since there are many layers of QW, each thin compared to the radiation wavelength, the incremental reduction in radiation intensity, I , as the radiation passes through an infinitesimal portion of the layers, dN , can be expressed as

$$\frac{dI}{I} = -\alpha_{2D} dN, \quad (2.14)$$

which integrates to give the familiar Beer-Lambert type law [66]

$$I = I_0 \exp(-\alpha_{2D} N_{QW}), \quad (2.15)$$

where I_0 is the intensity incident on the MQW. The *absorbance* of the full MQW is therefore

$$\alpha_{MQW} = \alpha_{2D} N_{QW}. \quad (2.16)$$

The absorbance of the NRC sample is the quantity to be measured in this thesis. Other modifications to equation 2.15 can be made to take into account angle of propagation of light through sample, and additional absorption not from the MQWs.

2.1.4. Energy levels in real MQWs

Until this point in our treatment, the QW has been taken as an infinite potential well, which provides a good idea of the behaviour of the ISBT and the electron wavefunctions. Detailed below are some significant modifications to the ISBT energy in the real system.

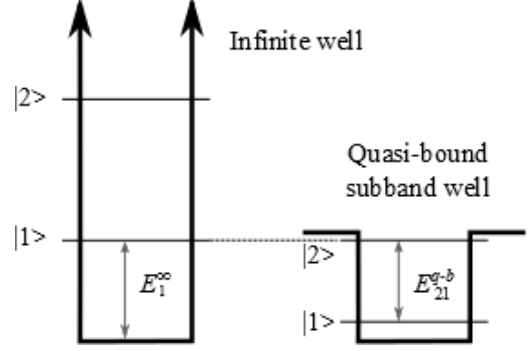
2.1.4.1. Finite potential well with quasi-bound ISBT

The first correction is that the wells are in fact finite, which becomes especially pertinent for bound to quasi-bound transitions, such as the ISBT of the NRC sample of this thesis.

Relationship between infinite well approximation and quasi-bound well ISBT

The lowering of the potential barrier, from infinity to a finite value, shifts the energy levels downwards, thereby creating a redshift in the ISBT; the energy levels in the well also become progressively fewer in number. One consequence is that the energy of the topmost level of a quasi-bound, finite quantum well, E_n^{q-b} , is the same as

Figure 2.2.: The relationship between an infinite well and finite well of the same width when the finite well has a quasi-bound first excited state, $|2\rangle$.



the energy of the level numbered below in the equivalent infinite well, E_{n-1}^∞ . For example, $E_2^{q-b} = E_1^\infty$, as in the case of our NRC sample. See Fig. 2.2.

From this fact we can make a first estimate of the ISBT energy of the quasi-bound state: $E_{21}^{q-b} \lesssim E_1^\infty(k_\parallel = 0)$. As a consequence, using equation 2.5, the actual quasi-bound $|2\rangle \rightarrow |1\rangle$ ISBT energy is less than a third of the infinite well approximation: $E_{21}^{q-b} \lesssim \frac{1}{3} E_{21}^\infty$.

Solving the finite well ISBT

To calculate the actual energy levels of the finite potential well, one follows the procedure outlined in, for example, Ref. [1]. This procedure requires knowledge of the depth of the well, V_0 , which is related to the difference between the barrier and well material bandgaps, and the band offset. The band offset between GaAs and $\text{Al}_x\text{Ga}_{1-x}\text{As}$ gives well depth of approximately 0.6 times that of the band gap difference [67]:

$$V_0 \approx 0.6 \times (E_g^{\text{AlGaAs}}(x) - E_g^{\text{GaAs}}), \quad (2.17)$$

where the bandgap of $\text{Al}_x\text{Ga}_{1-x}\text{As}$, $E_g^{\text{AlGaAs}}(x)$, depends on the proportion of Al atoms, x , in the alloy. This allows continuous control of the QW potential within the range of 0-337 meV.

Notation

From now on, the energy of the $|2\rangle \rightarrow |1\rangle$ transition is denoted without superscripts pertaining to whether infinite, finite or quasi-bound quantum wells. We assume a generic finite potential, of energy $E_{21} = \hbar\omega_{21} = E_{2,k_\parallel} - E_{1,k_\parallel}$.

2.1.4.2. Hartree potential (Self-consistent Coulomb potential)

The next modification to the energy of an ISBT regards the shape of the MQW conduction band.

To create an active ISBT requires the occupation of the lower subband of the transition. QWs are therefore doped, at 3D concentration N_D , to increase the carrier concentration and Fermi energy: $E_F \approx N_s \hbar^2 \pi / m^*$, where N_s is the electron sheet density (2D density) in the QW. In a QW, the charge distribution of the donated electrons is determined by the confined electron wavefunction, which will differ to the dopant ions' charge distribution. The resulting charge separation contributes to the potential of the wells, and a z -varying potential term in the system's Hamiltonian known as the Hartree potential.

When the well layers are themselves doped, the spatial charge distribution is not so disparate, resulting in a small blueshift in ISBT energies. However, this doping leads to increased scattering of the electrons with the dopant ions, leading to unwanted broadening of the ISBT absorbance linewidth. To avoid this, it is possible to dope the barrier layer from where the electrons transfer to the well layers. This satisfactorily decreases the electron-ion scattering, but does lead to less overlap of the dopant ion and electron charge distribution which increases the Hartree potential. The effect is shown in Fig. 2.3, where it can be seen that the bending of the conduction band upwards effectively reduces the well depth. The effect can redshift the ISBT energy by a significant amount, more than 5% in some cases. The Hartree potential is often referred to as the self-consistent Coulomb potential due to the manner in which the potential must be solved. A Hartree potential term, V_H , is included in equation 2.2 to account for this, an expression for which may be found in Ref. [8].

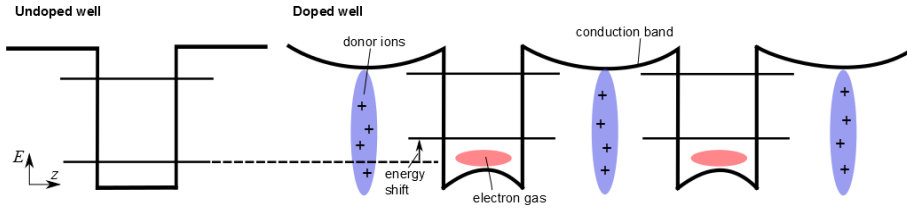


Figure 2.3.: Conduction band bending as a result of the Hartree potential in barrier-doped MQW. The well is effectively made shallower leading to a redshift in ISBT energy.

2.1.4.3. Many-body effects I - exchange and correlation

In a real QW the carrier concentration is high ($\sim 10^{12} \text{ cm}^{-2}$), and the Coulomb interaction between the large number of electrons will have a non-negligible effect on the energy levels. So called *many-body* effects act to shift the energy of the ISBT.

An effect termed *correlation* encompasses electron screening of the dopant ion charge, which decreases the Coulomb interaction, and the potential of the well. This acts to counter the energy level shift from the Hartree potential which was calculated for electrons without screening.

Additionally, electron *exchange* lowers the Coulomb attraction via the Pauli exclusion principle: electrons with the same spin will be spread out in space, lowering the concentration of charge, the mutual repulsion, and the effective potential in the well.

These two effects are strongest where there is higher electron density, therefore the ground subband is lowered further than the excited subbands, resulting in a blue-shift of the ISBT energy. As with the Hartree potential, the combined exchange-correlation potential is included as an additional term, $V_{xc}(N_s(z))$, in the Hamiltonian of equation 2.2:

$$\left[-\frac{\hbar^2}{2m^*} \frac{d^2}{dz^2} + V(z) + V_H(N_s(z)) + V_{xc}(N_s(z)) \right] \psi_{\mathbf{k}_{\parallel},n} = E_{n,\mathbf{k}_{\parallel}} \psi_{\mathbf{k}_{\parallel},n}. \quad (2.18)$$

See Refs. [8, 68] for discussion on methods of obtaining an expression for $V_{xc}(N_s(z))$.

2.1.4.4. Many-body effects II - depolarisation and exciton shift

There are two additional many-body effects of significance which adjust the ISBT energy $E_{21} = \hbar\omega_{21}$, as calculated from equation 2.18.

Arguably, the most important of the many-body effects is the depolarisation shift, in terms of the energy shift it induces. It is well established that the optical response of the MQW quasi-2DEG is like that of a collective oscillator [69, 70]. The Coulomb interaction between the QWs couples the layers, such that the charge density modulated in one layer will induce a correlated response in adjacent layers.

Because each QW layer is orders of magnitude thinner than the incident radiation wavelength, the MQW can be treated as an effective medium, capable of supporting 3D plasma oscillations. This plasma oscillation, known as the QW plasma frequency, ω_P , can itself couple with the collective ISBT oscillation. This results in a blueshifted coupled absorption frequency, the depolarisation-shifted frequency [71, 72]:

$$\omega_{dep} = \sqrt{\omega_{21}^2 + \omega_P^2}. \quad (2.19)$$

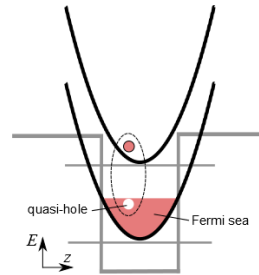
The QW plasma frequency ω_P , is given by

$$\omega_P = \sqrt{\frac{4\pi N_s e^2}{\varepsilon_w L_{QW}^{eff} m^*}} \quad (2.20)$$

where L_{QW}^{eff} is the effective QW width [8, 65]. The QW plasma frequency is a plasma oscillation where the MQW is effectively a quasi-homogeneous medium, with the electron density of the entire medium set to the value of the actual electron density of just the well layers, N_s/L_{QW}^{eff} .

On top of the depolarisation shift, there is the exciton shift which is due to the interaction of the excited electron with the quasi-hole left behind in the ground state subband Fermi sea (see Fig. 2.4). The Coulomb interaction between the electron and the quasi-hole lowers the electron energy, modifying the local Hartree potential and inducing a redshift.

Figure 2.4.: Exciton-like interaction between excited ISBT electron and quasi-hole left in the Fermi sea in ground state subband.



The transition frequency, ω_{21} (calculated from equation 2.18), is adjusted by the

exciton and depolarisation shifts to give the final, measurable ISBT frequency

$$\omega_{ISBT}^2 = \omega_{dep}^2 - \beta\omega_{21}^2, \quad (2.21)$$

where $\beta > 0$ is the factor pertaining to the exciton shift [8].

2.1.4.5. Total shift to finite potential ISBT energy

After calculating the energy levels for a finite, barrier-doped MQW, the combined additional energy shifts from all sources is in fact minimal. This is because the two largest energy shift mechanisms, the energy redshift due to the Hartree potential and the depolarisation blueshift, largely balance each other out.

The other mechanisms, smaller in magnitude, also cancel each other out to an extent [8, 73]. See Tab. 2.1.

The details of the energy shifts have mainly been listed because temperature-dependent properties of each mechanism differ, and our experiments involve cryogenic temperatures.

Table 2.1.: The total resonant energy shift on the ISBT of an 8 nm wide GaAs/Al_{0.3}Ga_{0.7}As quantum well as described by Ref. [8]. Barrier doping at a concentration of $N_D = 5 \times 10^{18} \text{ cm}^{-3}$, 4 nm from the well, gives an electron sheet density of $N_s = 5 \times 10^{18} \text{ cm}^{-2}$ in the well. Total shift to the bare finite potential ISBT is $\sim 1\%$.

	Resonance position
Bare finite potential	111.2 meV
+Hartree	-6.8 meV
+exchange correlation	+2.3 meV
+depolarisation	+8.8 meV
+exciton	-3.1 meV
Total potential	112.4 meV

2.2. The ISBT linewidth

The ISBT is subject to several broadening mechanisms which affect the relaxation lifetime of the excitation, and the sharpness of its absorbance linewidth.

Broadening mechanisms can be categorised into homogeneous and inhomogeneous contributions, where homogeneous broadening is typically a result of intrinsic scattering properties of the ISBT electron, or small length scale extrinsic scattering. Inhomogeneous broadening is a result of growth variations.

We follow the treatment of Unuma *et al.* [73] in the following sections.

2.2.1. Homogeneous broadening in MQWs

We first analyse the homogeneous broadening mechanisms which affect the ISBT linewidth.

2.2.1.1. Total summation of broadening mechanisms

The lifetime characteristics of electrons in semiconductors are inherently more complicated compared to free electrons in vacuum. This is due to the nature of the crystal in which the electrons reside providing multiple routes for relaxation, especially via phonons. Additionally, electrons in QWs face scattering mechanisms due to the heterostructure interface itself, where different semiconductor types meet.

The Lorentzian FWHM frequency linewidth of an ISBT, as introduced in equation 2.13, is related to the total relaxation lifetime τ_{ISBT} via

$$2\gamma = \frac{1}{\tau_{ISBT}}. \quad (2.22)$$

This is the general relationship between absorption linewidths and relaxation lifetimes, and for the rest of the thesis, linewidth will be used predominantly rather than lifetime. γ_{ISBT} incorporates all the scattering and relaxation rates that the ISBT is subject to (in terms of absorbance) via

$$\gamma_{ISBT} = \frac{1}{2} \left(\gamma_{ISBT}^{(intra)} + \gamma_{ISBT}^{(inter)} \right), \quad (2.23)$$

where the contribution has been split into intrasubband scattering $\gamma^{(intra)}$ (i.e. scattering which broadens the subband energy itself) and intersubband scattering $\gamma^{(inter)}$

[74]. Here, the intrasubband and intersubband linewidths for a two level ISBT are calculated for elastic scattering as

$$\gamma^{(intra)} = \frac{2\pi}{\hbar} \sum_{\mathbf{k}'} \left\langle \left| (1\mathbf{k}_{\parallel}' | V_{scat} | 1\mathbf{k}_{\parallel}) - (2\mathbf{k}_{\parallel}' | V_{scat} | 2\mathbf{k}_{\parallel}) \right|^2 \right\rangle \delta [E(\mathbf{k}_{\parallel}) - E(\mathbf{k}_{\parallel}')], \quad (2.24)$$

$$\gamma^{(inter)} = \frac{2\pi}{\hbar} \sum_{\mathbf{k}'} \left\langle \left| (1\mathbf{k}_{\parallel}' | V_{scat} | 2\mathbf{k}_{\parallel}) \right|^2 \right\rangle \delta [E(\mathbf{k}_{\parallel}) - E(\mathbf{k}_{\parallel}') + E_{21}] \quad (2.25)$$

where V_{scat} is the potential of a given scattering mechanism, $E(\mathbf{k}) = \hbar^2 k_{\parallel}^2 / 2m^*$, $|n\mathbf{k}_{\parallel}\rangle$ is the state vector of the electron with subband index n and in-plane wavevector $\mathbf{k}_{\parallel}$, and $\langle \dots \rangle$ denotes an average over the distribution of scatterers [73]. The delta functions, in their given form, ensure energy and momentum conservation, but for inelastic scattering would require an additional term in the delta function to take into account substantial energy losses. For example, for intrasubband longitudinal optical phonon emission: $\delta [E(\mathbf{k}_{\parallel}) - E(\mathbf{k}_{\parallel}')] \rightarrow \delta [E(\mathbf{k}_{\parallel}) - E(\mathbf{k}_{\parallel}') \pm E_{LO}]$.

The relative contribution and behaviour of scattering mechanisms may be evaluated by substituting appropriate scattering potentials into equations 2.24 and 2.25. The mechanisms which dominate the scattering are: longitudinal optical (LO) and longitudinal acoustic (LA) phonon scattering; interface roughness scattering (IRS); alloy disorder scattering (AD); and, ionised impurity scattering (ION).

These each contribute to γ_{ISBT} via¹:

$$\gamma_{ISBT} \approx \gamma_{LO} + \gamma_{LA} + \gamma_{IRS} + \gamma_{AD} + \gamma_{ION} \quad (2.26)$$

with $\gamma_{LO} = \gamma_{LO}^{(intra)} + \gamma_{LO}^{(inter)}$ etc. Each factor of equation 2.26 in principle has intersubband and intrasubband broadening mechanisms calculable from equations 2.24 and 2.25.

Ref. [73] gives approximate values for broadening from these various mechanisms in a typical GaAs QW, which are listed in Tab. 2.2. IRS is estimated to be the most

¹Note that some authors contest the additiveness of broadening, arguing that some mechanisms cancel, to an extent [75].

dominant broadening mechanism, followed by the two modes of phonon broadening (LO and LA). The additional broadening mechanisms (AD, ION, and others) are not treated beyond here, due to their smaller contribution.

Table 2.2.: Magnitudes of absorption linewidth broadening mechanisms of the ISBT, for GaAs quantum wells, width $\lesssim 10$ nm, with $T = 0$ K according to Ref. [73].

Broadening mechanism	FWHM contribution (meV/ $\hbar$)
IRS (intra/inter)	$\sim 1-10/\sim 1$
LO (intra/inter)	$\sim 0.1/\sim 1$
LA	~ 1
AD	~ 0.5
ION	~ 0.001

2.2.1.2. Interface roughness scattering

In the case of MQWs with especially narrow wells, the roughness of the well-barrier interface becomes significant. The ISBT electron, mobile in 2 dimensions, feels the roughness between the layers of semiconductor due to imperfect growth conditions, and is thus scattered: this is IRS. See Fig. 2.5. Even in ideal growth conditions of the MQW, IRS will still arise because, at the interface between well and barrier, there are two different crystals with different constituent atoms. Lattice matching techniques are refined, but will never be able to fully eliminate, for example, migration of the atoms at the interface [73].

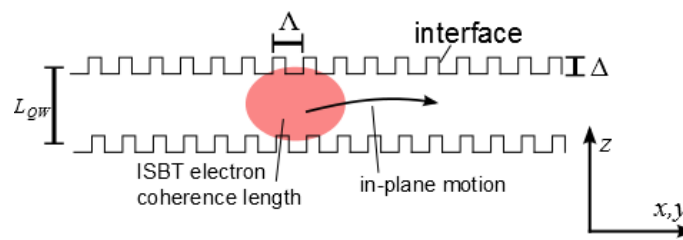


Figure 2.5.: Illustration of IRS. When the height of the scattering Δ becomes comparable to the (average) well width $\langle L_{QW} \rangle$, IRS becomes important, which broadens the ISBT through intrasubband in-plane scattering from roughness. The ISBT electron has a coherence length which, for GaAs/AlGaAs quantum wells [73], is larger than the correlation length, Λ , of the roughness, meaning that IRS gives a contribution to the homogeneous component of the linewidth.

It is assumed in Ref. [73] that IRS has a mean characteristic height, Δ , and a correlation length, Λ [see Fig. 2.5, a)], related via a correlation function

$$\langle \Delta(\mathbf{r})\Delta(\mathbf{r}') \rangle = \Delta^2 \exp\left(-\frac{|\mathbf{r} - \mathbf{r}'|^2}{\Lambda^2}\right), \quad (2.27)$$

with $\mathbf{r} = (x, y)$.

Making appropriate substitutions [73] into equation 2.24, gives an intrasubband IRS broadening described by

$$\gamma_{IRS}^{(intra)} = \frac{m_{scat}^* \Delta^2 \Lambda^2}{\hbar^3} \int_0^\pi \left[\frac{1}{S(q, T)} \frac{\partial E_1}{\partial L_{QW}} - \frac{\partial E_2}{\partial L_{QW}} \right]^2 d\theta \exp(-q^2 \Lambda^2 / 4), \quad (2.28)$$

where m_{scat}^* is the effective mass of the scattered electron, $q^2 = 2k_{\parallel}^2(1 - \cos \theta)$ is the 2D scattering vector, $S(q, T) \sim 1$ is a screening correction factor, and E_1 is the energy of the 1st subband, etc. Equation 2.28, the intrasubband linewidth, simplifies to

$$\gamma_{IRS}^{(intra)} = \frac{m_{scat}^* \Delta^2 \Lambda^2}{\hbar^3} \left[\frac{\partial E_1}{\partial L_{QW}} - \frac{\partial E_2}{\partial L_{QW}} \right]^2 \int_0^\pi d\theta \exp(-q^2 \Lambda^2 / 4). \quad (2.29)$$

The integral in this equation has a value converging to π when the in-plane momentum is small. The *intersubband* linewidth is of a similar form, dependent on the same variables, but roughly an order of magnitude smaller.

IRS becomes significant when the QW thickness, L_{QW} , approaches the scale of the roughness height $\Delta \lesssim 1$ nm, around $L_{QW} \lesssim 10$ nm in GaAs/AlGaAs QWs [76]. The width of the QWs studied in this thesis is 6 nm.

A note on IRS homogeneity in GaAs/AlGaAs

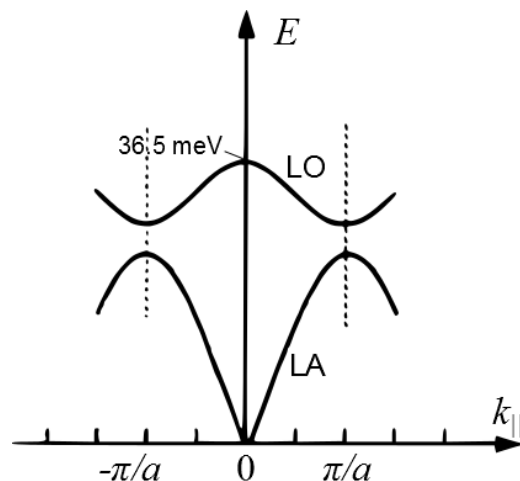
In the derivation of the above expressions for IRS, the correlation length, Λ , is presumed to be smaller than the ISBT electron mean free path, or in other words the coherence length. This is thought to be true of GaAs/AlGaAs MQWs where the

migration distance of Al atoms is short, and GaAs surfaces are virtually flat [73]. When this is the case, IRS contributes homogeneous broadening, since the electron responds to a moving average of the full roughness variation, which will not change significantly as the electron changes position.

2.2.1.3. Phonon scattering

In diatomic crystals, such as GaAs, the phonon branch splits into longitudinal optical (LO) and longitudinal acoustic (LA) modes [77]. In optical modes the adjacent oscillating atoms (which are of differing charge) move in anti-phase thereby creating a dipole moment which can potentially interact with an optical field. For acoustic modes, which determine how sound waves propagate in the crystal, adjacent atoms oscillate in phase. Fig. 2.6 shows a sketch of the dispersion of LO and LA phonons in GaAs.

Figure 2.6.: Phonon dispersion curves for the diatomic crystal GaAs showing splitting of the phonon branch into longitudinal optical and acoustic modes [77].



LO phonon scattering and emission

LO phonons in GaAs have an energy, near $k_{\parallel} = 0$, of $\hbar\omega_{LO} = 36.5$ meV [73] which is largely independent of $k_{\parallel}$. At room temperature there will exist a self-thermalised LO phonon population ($\frac{3}{2}k_B T \sim 39$ meV $>$ 36.5 meV), capable of intersubband scattering of the electrons. Intersubband LO phonon spontaneous emission is also a means of relaxation, provided the ISBT energy is greater than the LO phonon energy, $\hbar\omega_{ISBT} > \hbar\omega_{LO}$, as is the case for the NRC sample studied in this thesis (see Fig. 2.7). At $T = 0$ K, only intersubband spontaneous emission may occur; when

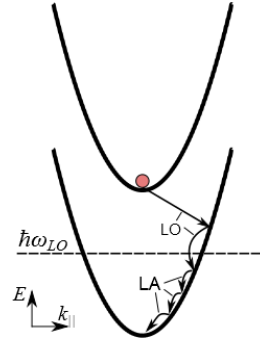
in-plane kinetic energy is sufficient, intrasubband phonon emission may also occur at this temperature.

Broadening is between 1 and 2 meV for typical QWs. These scattering and emission events occur with a typical time constant of ~ 1 ps [8] and provide the major means of electron intersubband relaxation. See Tab. 2.2.

LA phonon scattering and emission

Compared with the LO phonon, the LA phonon has a longer typical scattering time of 100 ps. Furthermore, the energy of a LA phonon is typically much smaller than that of the LO phonon (see Fig. 2.6), with values close to 0 meV near $k_{\parallel} = 0$. The LA phonon therefore is active in intrasubband relaxation of electrons, as they scatter downwards towards the subband minimum, losing momentum and energy (See Fig. 2.7). The LA phonon would have no way of causing an intersubband relaxation except in the narrower THz-frequency ISBTs, where it becomes an issue in the production of THz QCLs (see sec. 1.1.3.3). LA phonons contribute to both intrasubband and intersubband broadening, with a total of $\lesssim 1$ meV/ $\hbar$ for QWs [73]. This is comparable to the LO phonon broadening, despite the orders of magnitude of difference in scattering time. LA phonons have a similar broadening contribution to the total ISBT linewidth possibly because of the increased number of times intrasubband scattering may occur in the lower subband in a given relaxation.

Figure 2.7.: Relaxation of ISBT electron via LO phonon emission to the lower subband followed by LO and LA phonon scattering and emission.



2.2.2. Inhomogeneous broadening from growth fluctuations

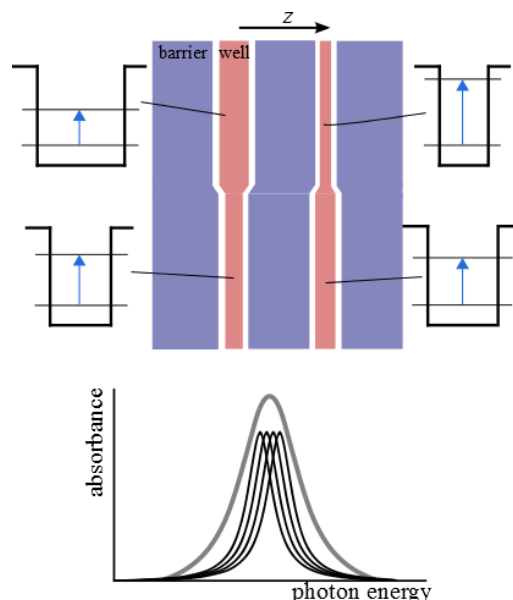
We now analyse the inhomogeneous broadening mechanisms that affect the ISBT linewidth.

2.2.2.1. Are ISBTs inhomogeneously broadened?

For non-ideal semiconductor growth, there exists the possibility of inhomogeneous broadening. For example, should the width of the QWs differ from one well layer to another (well width fluctuation) then there will be a spread in ISBT energies. Similarly, if there is well width variation within a single well layer, on a lateral length scale larger than the coherence length of the electron, then there will again be a spread in ISBT absorption energies. See Fig. 2.8. Furthermore, doping concentrations may be inhomogeneously distributed across QW layers, and would contribute to additional inhomogeneity.

The ISBT energies are presumed, to a first approximation, to have a Gaussian spread about a mean energy². Since each ISBT has a Lorentzian absorbance profile, the total absorbance profile will be a convolution of the Lorentzian and the Gaussian profiles - a *Voigt* profile³.

Figure 2.8.: (Top) Depiction of intra-well well width variation on an inhomogeneous scale, and inter-well width variation (well width fluctuation), leading to inhomogeneous broadening. (Bottom) The total absorbance profile of the ISBTs together will therefore be a convolution of absorption profiles and therefore broaden.



The magnitude of this inhomogeneous broadening will of course depend on the quality of the sample grown, and indeed, with the maturation of MBE, there has come

²It is more likely that the *well widths*, L_{QW} , rather than the ISBT energy itself, will have a Gaussian spread about a mean value. Since ISBT energy is proportional to $1/L_{QW}^2$, the spread in energy will not be exactly Gaussian, and will have a longer high-energy tail.

³The convolution of a Gaussian and a Lorentzian profile has long been known to the field of atomic spectroscopy, where the Doppler shift of atomic energy levels leads to a broadening of emission/absorbance linewidths. Here the convolution is known as the ‘plasma dispersion function’.

a reduction in the inhomogeneous broadening seen in samples. However, whether or not inhomogeneous broadening can be fully eliminated is still up for debate. For example, Beadie *et al.* [78] found evidence for inhomogeneous broadening in MQW, whilst on the other hand, previous work in our own group by Vodopyanov *et al.* [79] found no evidence of spectral hole-burning in an MQW sample, a fact which would usually indicate a lack of inhomogeneous broadening.

In the presence of inhomogeneous broadening, the FWHM of the ISBT absorbance profile (equation 2.13) is modified to reflect the Voigt profile shape; the linewidth cannot be parameterised by solely the Lorentzian FWHM, γ_{ISBT} .

2.2.2.2. Voigt profile (Gaussian-Lorentzian convolution)

The Voigt profile is defined as [80]

$$V(x; \sigma, \gamma) = \int_{-\infty}^{\infty} G(x, \sigma) L(x - x', \gamma) dx' \quad (2.30)$$

where $G(x; x_0, \sigma)$ is the Gaussian profile with parameter σ , centred at x_0 :

$$G(x; x_0, \sigma) = \frac{1}{\sigma\sqrt{2\pi}} \exp\left(-\frac{(x - x_0)^2}{2\sigma^2}\right), \quad (2.31)$$

and $L(x; x_0, \gamma)$ is the Lorentzian profile with FWHM, γ , and centred at x_0 :

$$L(x; x_0, \gamma) = \frac{\gamma/2}{\pi((x - x_0)^2 + \gamma^2/4)}. \quad (2.32)$$

The Voigt profile FWHM is approximated by [81]

$$\delta_{Voigt} \simeq 0.5346\gamma + \sqrt{0.2166\gamma^2 + \xi^2} \quad (2.33)$$

where ξ is the Gaussian FWHM, with $\xi = 2\sigma\sqrt{2\ln(2)}$. The behaviour of the Voigt profile with varying Gaussian FWHM is shown in Fig. 2.9. Note from that figure

the way that at the wings of the profiles the Voigt resembles the pure Lorentzian regardless of the magnitude of the Gaussian contribution as it is varied from $\xi = 0$ to $\xi > \gamma$. This fact is significant for sec. 2.4.4.2.

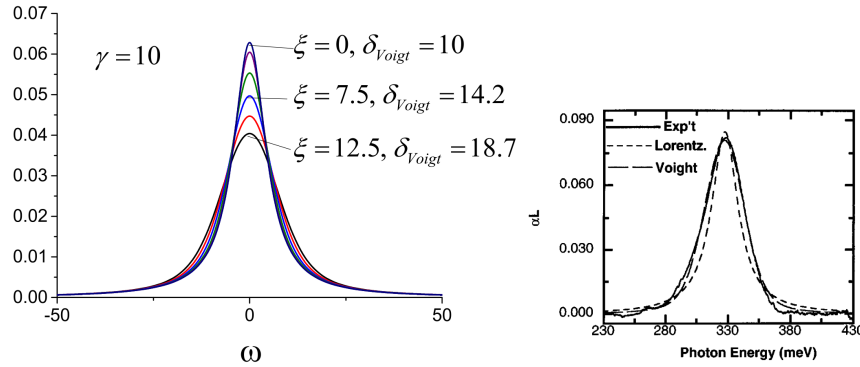


Figure 2.9.: (Left) The Voigt profile when Gaussian FWHM ξ is varied and the Lorentzian FWHM held constant throughout ($\gamma = 10$). (Right) Experimental fitting of 4 nm, inhomogeneously broadened $\text{In}_{0.45}\text{Ga}_{0.55}\text{As}/\text{Al}_{0.45}\text{Ga}_{0.55}\text{As}$ MQW ISBT absorbance profile, showing good agreement from Voigt profile fit, compared to Lorentzian fit. Reprinted figure with permission from [78]. Copyright (1997) by the American Physical Society.

The Voigt profile would then be incorporated into the ISBT absorption profile (of equation 2.13) as

$$\alpha_{2D}(\omega) = \frac{\hbar n_s}{2\varepsilon_0 c \eta} \frac{q^2}{m^*} f_{12} e_z^2 V(\omega; \gamma_{ISBT}, \xi_{ISBT}), \quad (2.34)$$

with the Gaussian parameter, ξ_{ISBT} , controlled by the amount of well width variation. The FWHM of the ISBT absorption profile would therefore be $\delta_{Voigt}(\gamma_{ISBT}, \xi(\sigma_{ISBT}))$.

The question of ISBT inhomogeneity will be examined closely in the course of this thesis because of the implication it may have on the behaviour of the linewidth of the ICP.

2.3. Microcavities and mode quantisation

Having focussed up to now on the ISBT, we must now turn our attention to the other component of the ICP: the cavity mode. The NRC sample studied in this

thesis is an MQW situated within a waveguide microcavity. This is the same thing as a flat-flat Fabry-Pérot cavity, except that in this case mode propagation parallel to the mirrors must be taken into account, since coupling into the cavity is at an angle.

2.3.1. Quantisation of the field within a waveguide

2.3.1.1. Waveguide with ideal mirrors

If a slab of dielectric is placed between two dielectric layers of lower refractive index then propagating light may be confined to the slab via total internal reflection (TIR), thus forming a waveguide. The two dielectric layers act as mirror layers. In the ray picture of light, light reflects between the mirror layers at an angle of incidence lower than the critical angle, $\varphi < \varphi_c$, and the interference of wavefronts causes a standing wave pattern across the slab, which is subject to boundary conditions. See Fig. 2.10. For TM-polarised light this requires the y -direction magnetic field, H_y , to be continuous across the boundary between mirrors and slab. In the case of ideal mirrors, for which there is no transmission into the mirrors, at the edges of the microcavity ($z = 0$ and $z = L_{MC}$): [82]

$$H_y = \begin{cases} A \sin(k_{n,z}z), & 0 < z < L_{MC} \\ 0, & z < 0, z > L_{MC} \end{cases}, \quad (2.35)$$

where

$$k_{n,z} = \frac{\pi n}{L_{MC}}, \quad n = 1, 2, 3... \quad (2.36)$$

is the wavevector in the z -direction, with mode index n , and A represents the magnitude of H_y , and is a constant. See Fig. 2.10. This simply expresses the necessity for only half-integer wavelengths of light being able to resonate within the waveguide when resolved in the confinement direction. If light is coupled into the waveguide at an angle φ , then only particular wavelengths (or, equivalently, photon frequencies) will be resonant with the waveguide's modes. Very similar solutions exist for the

case of transverse electric- (TE-) polarised light, wherein it is the electric field, E_y , which must satisfy the boundary conditions.

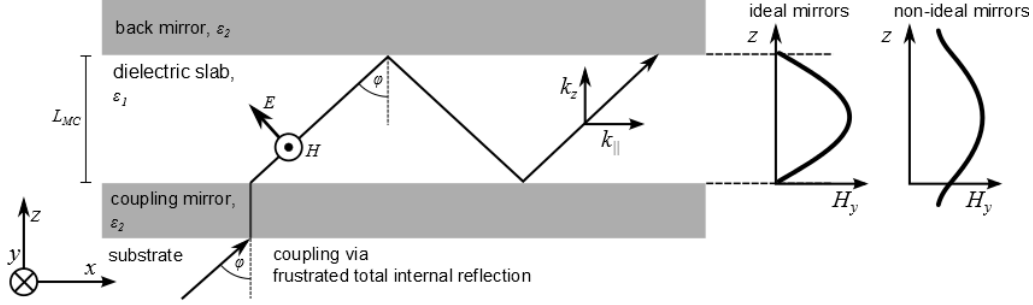


Figure 2.10.: Illustration of TM-polarised light propagating in a waveguide microcavity, of thickness L_{MC} . Shown is the coupling angle, φ , of the light, which will be equal to the angle within the slab should the substrate have the same dielectric constant. The wavevector of the light in the waveguide is shown resolved into x and z directions, with the x -direction wavevector known as the in-plane wavevector $k_{||}$. To the right is shown the resonant H_y field of the fundamental mode in the case of ideal mirrors, and non-ideal mirrors.

The wavevector of the light within the waveguide, parallel to the mirrors (x -direction), is

$$k_{x,n} \equiv k_{||,n} = \sqrt{K^2 \varepsilon_1 - k_{n,z}^2}, \quad (2.37)$$

where $\varepsilon_1 = n_1^2$ is the permittivity of the slab medium, which, for now, is presumed isotropic and lossless (n_1 is the refractive index of the slab medium, and permeability is taken as $\mu_1 = 1$). The quantity $K = (\frac{\omega}{c})$ is the light's wavevector in vacuum. The so-called *radiation region* is bound by the line $\omega = k_{||}c/\sqrt{\varepsilon_2}$ on the dispersion curve, corresponding to where the waveguide can no longer confine the radiation through TIR [83]. At high $k_{||}$, modes converge with the free-space line for the waveguide slab medium, $\omega = k_{||}c/\sqrt{\varepsilon_1}$. Examples of these modes are plotted in Fig. 2.11.

The corresponding supported frequency modes of the waveguide are

$$\bar{\omega}_n^2(k_{||}) = \omega_{||}^2(k_{||}) + n^2 \omega_z^2 \quad (2.38)$$

where $\omega_{||} = k_{||}c/\sqrt{\varepsilon_1}$ and $\omega_z = \frac{\pi c}{L_{MC}\sqrt{\varepsilon_1}}$. The bar above $\bar{\omega}_n$ denotes that this is for a

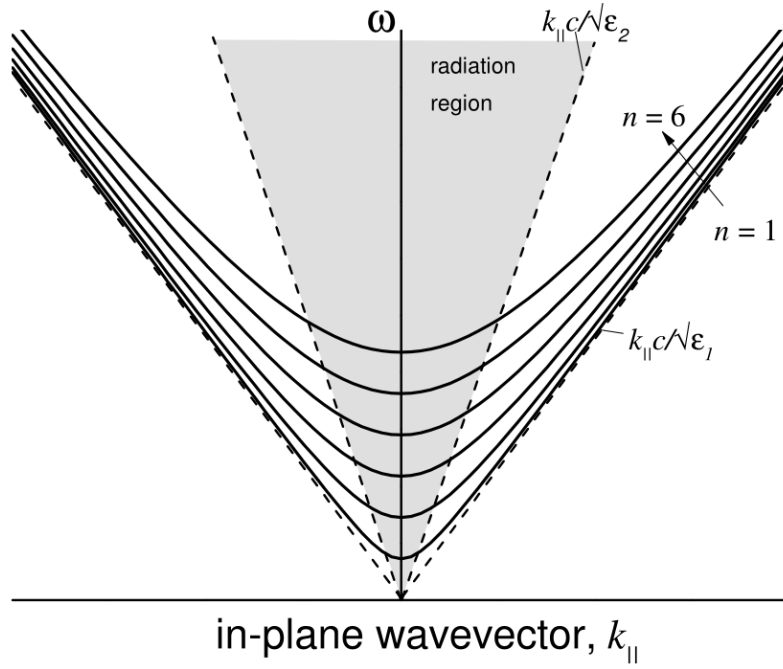


Figure 2.11.: Dispersion of the first 6 modes in an ideal waveguide microcavity (TM and TE are equivalent when with ideal mirrors), plotted as frequency versus in-plane wavevector. The radiation region is shown, within which there is no propagation in the waveguide.

cavity empty of MQWs, with ideal mirrors.

Experimentally, dispersion curves are measured against incidence angle, φ (see Fig. 2.10). Dispersion relations are transformed from $k_{||}$ to φ by [65]

$$k_{||} = \frac{\sqrt{\varepsilon_1}}{c} \omega(\varphi) \sin \varphi, \quad (2.39)$$

which, combined with equation 2.37, gives

$$\bar{\omega}_n = \frac{\pi n c}{L_{MC} \sqrt{\varepsilon_1} \cos \varphi} \quad (2.40)$$

for the φ -dependent dispersion relation. Equation 2.40 gives dispersion curves as shown in Fig. 2.12 . Here it can be seen that the radiation region is bound by the critical angle of the TIR confining the light to the waveguide, below which the

dielectric mirror layers become leaky. The implication is that $k_{\parallel} \approx 0$ modes cannot be reached in a TIR-based waveguide.

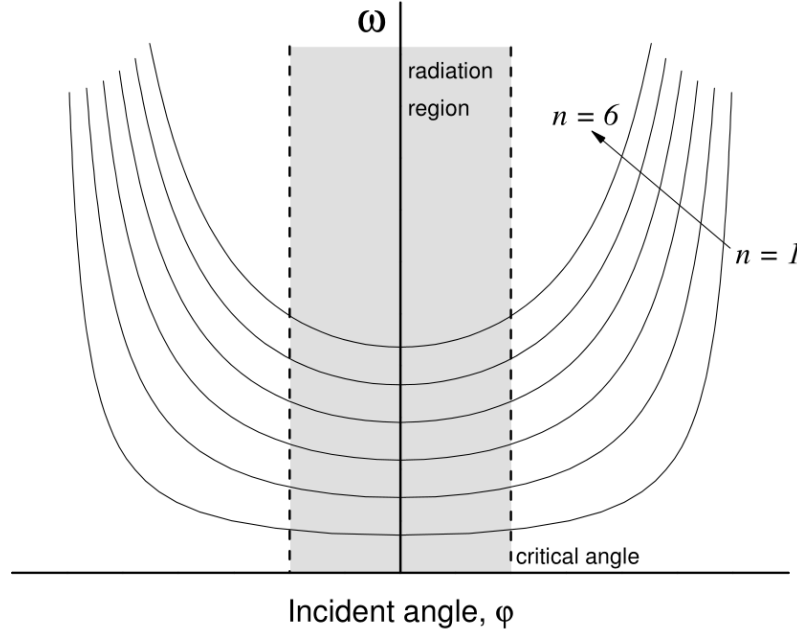


Figure 2.12.: Dispersion of the first 6 modes in a slab waveguide, frequency, ω versus coupling angle, φ , with same range of $k_{\parallel}$ and ω as Fig.2.11. Shown is the position of the critical angle below which there is no TIR, and therefore no confined waveguide mode: the radiation region.

2.3.1.2. Waveguide with non-ideal mirrors

In the case of non-ideal mirrors (reflectivity < 1) the standing wave pattern of the field penetrates some way into the mirrors, decaying exponentially away from the slab (see Fig. 2.10). The model solution for the resonant magnetic field is

$$H_y = \begin{cases} A \exp(-\eta(z - L_{MC})) & z > L_{MC} \\ B \sin(k_{n,z}z) & 0 < z < L_{MC} \\ C \exp(\eta(z)) & z < 0. \end{cases} \quad (2.41)$$

The extinction parameter for the mirrors, η , the mode wavevector, $k_{n,z}$, and constants A, B, C are found by applying boundary conditions, and solving transcendental equations, as with the electron in finite quantum well problem. This effect

does not change the basic feel of the solution to the propagating modes, but does effectively extend the cavity length, thereby shifting the dispersion curves.

The form of $k_{n,z}$ must now take into account the phase shift and reflectivity of the mirrors, so is no longer $k_{n,z} = \pi n/L_{MC}$ (i.e. equation 2.36).

The resonant condition equation for $k_{n,z}$ is rederived to take this into account [65], through multibeam analysis or transfer matrix formalism (see Ref. [84]) to give

$$k_{n,z}L_{MC} = \pi \left[n - \frac{\Phi_{\text{mirr}}}{2\pi} + i \frac{\varrho_{\text{mirr}}}{2\pi} \right] \quad (2.42)$$

where Φ_{mirr} is the total phase shift of both mirrors, $\varrho_{\text{mirr}} = \ln(R)$ is the total reflectivity of both mirrors, with the reflection coefficient $R < 1$, and n is the mode index integer (as before). In terms of the complex frequency of the waveguide

$$\tilde{\omega}_{MC}(n, \varphi) = \bar{\omega}_1 \left[n - \frac{\Phi_{\text{mirr}}(\omega)}{2\pi} + i \frac{\varrho_{\text{mirr}}(\omega)}{2\pi} \right] \quad (2.43)$$

where the substitution $k_{n,z} = \frac{\omega\sqrt{\epsilon_1}}{c} \cos \varphi$ has been made, and $\bar{\omega}_1$ is the fundamental mode in the ideal-mirror case (equation 2.40). Hence, the waveguide modes are redshifted for positive mirror dispersion.

Another outcome is the imaginary component to the wavevector and frequency which takes into account mirror losses, and thereby broadening of the photonic modes. (See sec. 2.3.2.)

2.3.1.3. Frustrated total internal reflection for coupling

The exponential decay into the non-ideal mirrors ($z > L_{MC}$ and $z < 0$ in equation 2.41) is of vital importance from an experimental point of view, since it allows light to be coupled into and out of the cavity via frustrated TIR. Analogous to quantum mechanical tunnelling, if the mirror ends before the exponentially decreasing field diminishes, it can become oscillatory on the other side of the mirror and propagate. See Fig. 2.13.

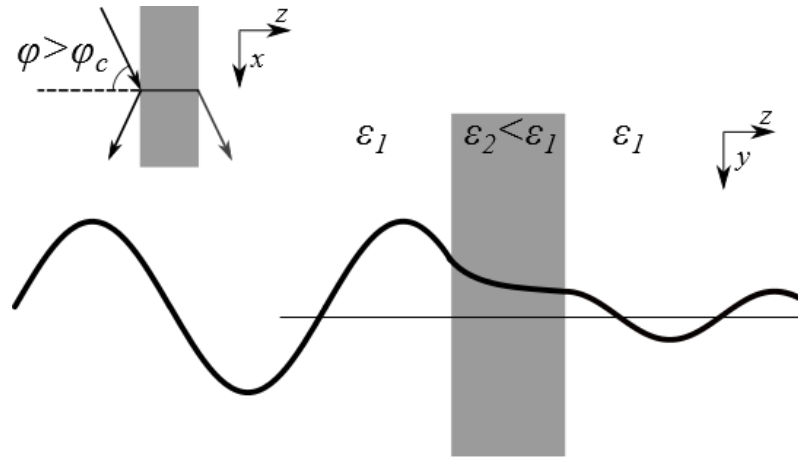


Figure 2.13.: Electric field in a medium of refractive index $n_1 = \sqrt{\epsilon_1}$ incident on a layer of refractive index $n_2 < n_1$. Angle of incidence is larger than the critical angle (inset), hence the field is totally internally reflected; however, the reflecting medium is sufficiently thin such that the electric field does not fully diminish across its length. The electric field is therefore able to become oscillatory on the other side of the layer, and thereby propagate again. This is known as frustrated TIR. This is the means of coupling into and out of the MQW microcavity as shown in Fig. 2.10.

2.3.1.4. Measuring waveguide modes

In terms of experimental measurements, at a given angle, φ , if the photon energy of the beam does not correspond to a resonant mode of the waveguide then it will couple back out and be detected. However, when the photon energy does create a resonant cavity mode, the mode will flow parallel to the mirrors. Less light will be coupled out and detected, so this will appear as absorbance from the waveguide. This is in contrast with non-waveguide planar cavities, with normal incidence radiation, where at the cavity resonance, the transmission through the cavity is enhanced, not diminished [85]. It is thus important to distinguish between an ordinary Fabry-Pérot cavity and a waveguide cavity for this reason.

2.3.2. Waveguide microcavity linewidth

Here we treat the broadening of the waveguide cavity absorbance linewidths.

2.3.2.1. Imaginary component of the complex frequency

The mirror losses of a microcavity introduce a characteristic lifetime to photons resonating within. This finite lifetime translates as a broadening in the resonance frequency of the cavity. In general, the FWHM broadening, γ , is related to the complex frequency $\tilde{\omega}$ by

$$\gamma = -2\text{Im}(\tilde{\omega}). \quad (2.44)$$

Substituting equation 2.43 gives the broadening of the microcavity resonant mode as⁴

$$\gamma_{MC} = \frac{c}{2L_{MC}\sqrt{\varepsilon_1}\cos\varphi} \left(-\ln\sqrt{R}\right). \quad (2.45)$$

Larger coupling angles and narrower microcavities give wider linewidths. Here we can also see that the dispersion in the mirrors could become significant, i.e. if $R = R(\omega, \varphi)$, there will be asymmetry in the cavity linewidth, and further dependence on φ .

2.4. Formation of intersubband cavity polaritons from the strong coupling of ISBT and microcavity modes

In the preceding two sections we have introduced the ISBT mode and its linewidth, and the microcavity mode and its linewidth. We now consider the case when these modes are resonant with each other, and strongly couple. Detailed below largely follows a semi-classical approach based on the work of Załuźny and Zietkowski [65] to describe the MQW microcavity system. For detailed quantum mechanical treatments of vacuum Rabi splitting and ICPs, see Ref. [22], chapter 6B and Ref. [53].

⁴A similar expression is gained from treating the waveguide as a Fabry-Pérot etalon [86].

2.4.1. Theoretical description of intersubband cavity polaritons

To model the coupling of the ISBT and waveguide modes, it is possible to consider the complex waveguide mode, as in sec. 2.3, but replace the dielectric constant of the slab medium ϵ_1 by an anisotropic permittivity tensor ϵ_{MQW} with components $\epsilon_{xx} = \epsilon_{yy} = \epsilon_{\parallel} \neq \epsilon_{zz}$, where the z -direction permittivity incorporates the absorption properties of the ISBT.

2.4.1.1. Effective medium approximation

Since each quantum well thickness is much smaller than the wavelength of incident light, it is possible to calculate the average response of the MQW taken as a single layer, dramatically simplifying calculations. This is the effective medium approximation (EMA). See Fig. 2.14.

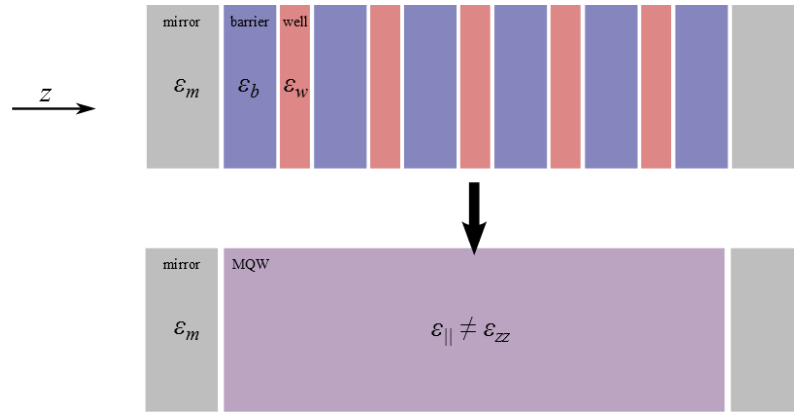


Figure 2.14.: The effective medium approximation. Since QW layers are thinner than the wavelength of incident light, the whole MQW can be approximated as a single layer with anisotropic permittivity.

For anisotropic media, such as the MQW, solving the wave equation for plane waves gives, for the z -direction wavevector [87]

$$k_z^{MQW} = \left[\epsilon_{xx} K^2 - (\epsilon_{xx}/\epsilon_{zz}) k_{\parallel}^2 \right]^{1/2}, \quad (2.46)$$

where again, $K = \left(\frac{\omega}{c}\right)$. When the approximation is taken that the barrier and well materials have equal permittivity, $\epsilon_b = \epsilon_w$, the permittivity tensor, as calculated in

the EMA, has the components [65]

$$\varepsilon_{xx} = \varepsilon_{yy} = \varepsilon_w + i \frac{4\pi}{d_{MQW}\omega} \sigma_{xx}(\omega), \quad (2.47)$$

where d_{MQW} is the MQW period, and we have introduced the in-plane conductivity

$$\sigma_{xx}(\omega) = \frac{N_s e^2}{m^*} \frac{i}{\omega + i\gamma_{ISBT}^{(intra)}}, \quad (2.48)$$

where N_s is the electron sheet density, and $\gamma_{ISBT}^{(intra)}$ is the intrasubband broadening of sec. 2.2, here representing the *in-plane* broadening. This expression of in-plane permittivity encompasses the Drude response of the MQW 2DEG (see sec. 1.1.3).

For the z -direction permittivity constant, we have

$$\frac{1}{\varepsilon_{zz}} = \frac{1}{\varepsilon_w} - \frac{i}{\varepsilon_w^2} \frac{4\pi}{d_{MQW}\omega} \sigma_{zz}(\omega), \quad (2.49)$$

where the conductivity in the z -direction is

$$\sigma_{zz}(\omega) = \frac{N_s e^2 f_{21}}{m^*} \frac{i\omega}{\omega^2 - \omega_{ISBT}^2 + i2\omega\gamma_{ISBT}^{(inter)}}, \quad (2.50)$$

and $\gamma_{ISBT}^{(inter)}$ is the homogeneous intersubband broadening (the FWHM absorbance linewidth broadening) of the ISBT⁵.

A full expression for k_z^{MQW} can be obtained from the transfer matrix formalism (see Ref. [84]), though much simpler analytical solutions can be obtained if the microcavity mirrors are taken as ideal. We get a result similar to equation 2.36, but

⁵The relationship between absorption as derived in equation 2.13 and the permittivity is straightforward in isotropic media, i.e. $\alpha = \text{Im}(\varepsilon)$. However, in anisotropic media, it is not so simple, with the absorption peak being shifted with respect to the permittivity peak. See Ref. [88] Figure 1c) for more details.

acknowledging that $L_{MC} \simeq L_{MQW}$:

$$k_z^{MQW} = \frac{n\pi}{L_{MQW}}, \quad n = 1, 2, 3... \quad (2.51)$$

Equating this with equation 2.46 and substituting in equations 2.47 to 2.50, and for now eliminating the broadening ($\gamma_{ISBT}^{(intra)} = \gamma_{ISBT}^{(inter)} = 0$), we get the following ‘exact’ eigenfrequency relationship for the system:

$$\omega^2 - \bar{\omega}_n^2 = \frac{\omega_{\parallel}^2 f_{12} \omega_p^2}{\omega^2 - \omega_{ISBT}^2} + \frac{n^2 \omega_z^2 \omega_p^2}{\omega^2 - \omega_p^2} \quad (2.52)$$

where we have re-introduced in-plane and perpendicular (z -direction) frequencies, as used in equation 2.38. We have also introduced the frequency of the MQW plasma, ω_p , which considers the entire MQW as a uniform plasma with the average value of electron density:

$$\omega_p = \sqrt{\frac{4\pi N_s e^2}{\varepsilon_w d_{MQW} m^*}} \quad (2.53)$$

where d_{MQW} is the period of the MQW. This differs from the previously-introduced QW plasma frequency, ω_P , of equation 2.20, which considered the plasma frequency of the MQW, but with an electron density equal to that of the well layer only. These two plasma frequencies are related via $\frac{\omega_p}{\omega_P} = \left(\frac{L_{QW}^{eff}}{d_{MQW}} \right)^{1/2}$.

To simplify the calculation of the eigenfrequency from equation 2.52 we can make the simplification that the in-plane permittivity is simply that of the well, $\varepsilon_{xx} = \varepsilon_w$, effectively ignoring the intrasubband plasma oscillations. This reduces equation 2.52 to

$$(\omega^2 - \bar{\omega}_n^2) (\omega^2 - \omega_{ISBT}^2) = \omega_{\parallel}^2 f_{12} \omega_p^2. \quad (2.54)$$

Finally, if we take the system at resonance, i.e. $k_{\parallel} \approx k_{\parallel, res}$ for the first waveguide

mode, which implies $\omega \approx \bar{\omega}_1 \approx \omega_{ISBT}$ then the above reduces to

$$4\omega^2 (\omega - \bar{\omega}_1) (\omega - \omega_{ISBT}) = \omega_{\parallel}^2 f_{12} \omega_p^2 \quad (2.55)$$

which gives

$$(\omega - \bar{\omega}_1) (\omega - \omega_{ISBT}) = \frac{\pi N_s e^2 f_{12}}{\varepsilon_w m^* d_{MQW}} \frac{k_{\parallel}^2}{k_{\parallel}^2 + k_z^2} = \Omega_{R, k_{\parallel}}. \quad (2.56)$$

Here we have arrived at the *simple two coupled oscillator* (STCO) equation for solving the polariton modes of the system. We have also now formally introduced the coupling strength, $\Omega_{R, k_{\parallel}}$, which will be equal to the vacuum Rabi frequency at the resonance position, $k_{\parallel, res}$ (see sec. 2.4.2).

The STCO (equation 2.56) gives a feel for the behaviour of the supported polariton modes, but bear in mind the simplifications made in arriving here: no intrasubband effects, ideal mirrors, ISBT and cavity mode close to resonance, equal well and barrier permittivities ($\varepsilon_w = \varepsilon_b$), and no absorption linewidth broadening.

2.4.1.2. Voigt profile permittivity

Should the MQW have inhomogeneous broadening rather than purely homogeneous, and therefore the ISBT have a Voigt absorption profile, the term for z -direction permittivity is modified to reflect this [89]:

$$\frac{1}{\varepsilon_{zz}^{Voigt}} = \frac{1}{\varepsilon_w} - \frac{N_s e^2 f_{12} \hbar \pi}{2m^* \varepsilon_w^2 \omega} \left(\frac{i \text{Re}(w(u)) + \text{Im}(w(u))}{\sigma \sqrt{2\pi}} \right) \quad (2.57)$$

where

$$w(u) = e^{-z^2} \text{erfc}(-iu) \quad (2.58)$$

is the Faddeeva function, and $\text{erfc}(-iu)$ is the complimentary error function, with

$$u = \frac{(\omega_{21} - \omega) + i\gamma_{ISBT}^{(inter)}/2}{\sigma\sqrt{2}}. \quad (2.59)$$

This Voigt permittivity converges with the previous expression for permittivity, equation 2.49, when $\sigma = 0$. The Voigt profile is given by

$$\text{Re}(w(u))/\sigma\sqrt{2\pi} = V(x; \gamma, \sigma). \quad (2.60)$$

Analytical results with this expression of permittivity are not possible, though equation 2.57 will be required to model inhomogeneity in any numerical model, as in sec. 3.7.

2.4.2. Simple two coupled oscillator model

The full waveguide analysis of sec. 2.4.1, after simplifications, leads to the STCO expression of equation 2.56. We can take this as a starting point for further analysis, and here reintroduce the broadening terms of the ISBT and the microcavity, γ_{ISBT} and γ_{MC} [84]. The STCO equation is valid if the ISBT modes are taken as a collective mode, and if the higher order cavity modes are ignored (single mode cavity approximation).

We are then able to treat the coupled system with the STCO model, the characteristic equation of which is [84]

$$(\tilde{\omega}_{ISBT} - \omega) (\tilde{\omega}_{MC}(k_{\parallel}) - \omega) = \frac{\Omega_{R,k_{\parallel}}^2}{4} \quad (2.61)$$

where $\tilde{\omega}_{ISBT} = \omega_{ISBT} - i\gamma_{ISBT}/2$ is the complex ISBT frequency and $\tilde{\omega}_{MC}(k_{\parallel}) = \bar{\omega}_1(k_{\parallel}) - i\gamma_{MC}(k_{\parallel})/2$ is the complex microcavity (fundamental) mode frequency. Only homogeneous broadening can be considered in this equation. $\Omega_{R,k_{\parallel}}$ is the coupling

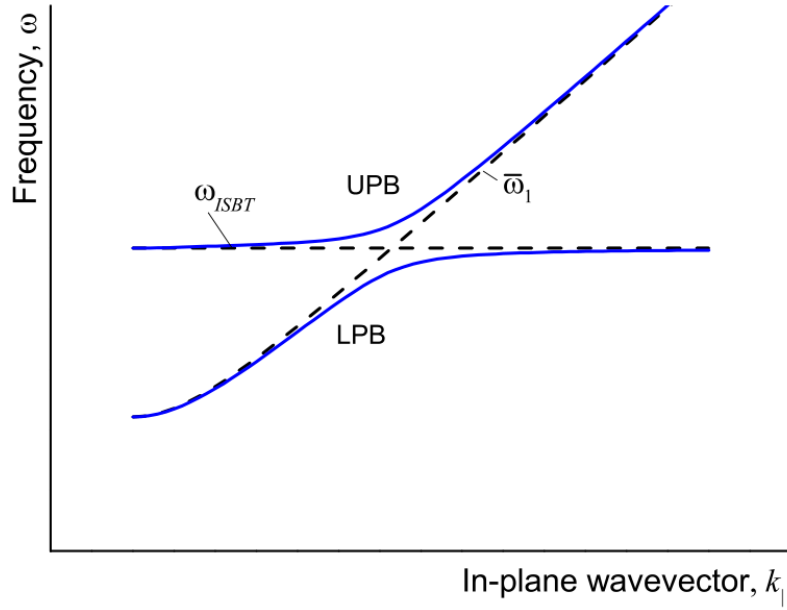


Figure 2.15.: Resonance of the ISBT mode and the fundamental cavity mode leads to formation of two new modes, the upper and lower polariton branches (UPB/LPB) which anticross at the resonance.

strength of the two oscillators, which through comparison with equation 2.56 is

$$\Omega_{R,k_{||}} = \sqrt{\frac{4\pi e^2 N_s f_{12}}{m^* \varepsilon_w d_{MQW}} \frac{k_{||}^2}{(k_{||}^2 + k_{z,1}^2)}}, \quad (2.62)$$

which becomes the *vacuum Rabi frequency* when at resonance ($k_{||} = k_{||,res}$): $\Omega_{R,k_{||,res}}$.

There are two eigenfrequency solutions to equation 2.61:

$$\begin{aligned} \tilde{\omega}_{UPB,LPB} = & \frac{\omega_{ISBT} + \omega_{MC} - i(\gamma_{ISBT} + \gamma_{MC})/2}{2} \\ & \pm \frac{1}{2} \sqrt{\Omega_{R,k_{||}}^2 + (\omega_{ISBT} - \omega_{MC} - i(\gamma_{ISBT} - \gamma_{MC})/2)^2} \end{aligned} \quad (2.63)$$

where the upper polariton branch (UPB) takes the positive root, and lower polariton branch (LPB) takes the negative root. Collectively, the UPB and LPB polaritons are the ICPs.

2.4 Formation of intersubband cavity polaritons from the strong coupling of ISBT and microcavity modes

The behaviour of equation 2.63 is shown in Fig. 2.15, where it can be seen that the ICP dispersions, instead of following the microcavity and ISBT modes, do not cross at the resonance wavevector, $k_{\parallel, res}$. This is known as the anticrossing point.

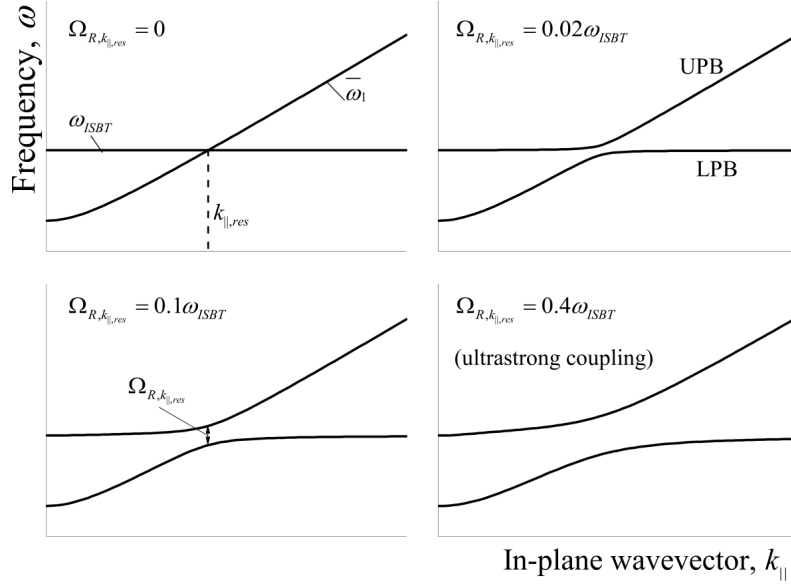


Figure 2.16.: The anticrossing of the ISBT mode and the fundamental cavity mode increases as the vacuum Rabi frequency ($\Omega_{R, k_{\parallel, res}}$) is increased. The magnitude of the Rabi splitting of the UPB and LPB branches at resonance is $\Omega_{R, k_{\parallel, res}}$, following equation 2.64.

The stronger the coupling between the microcavity and ISBT modes, the larger the minimum splitting between the UPB and LPB branches (see Fig. 2.16). At $k_{\parallel} = k_{\parallel, res}$ where $\omega_{ISBT} = \bar{\omega}_1(k_{\parallel, res})$, the splitting is given by

$$\text{Re}(\tilde{\omega}_{UPB} - \tilde{\omega}_{LPB}) = \sqrt{\Omega_{R, k_{\parallel, res}}^2 - (\gamma_{ISBT} - \gamma_{MC})^2 / 4} \approx \Omega_{R, k_{\parallel, res}}. \quad (2.64)$$

Here at the resonance the ISBT energy is said to be *Rabi split*, with new resonance energies at $\sim \pm \frac{1}{2}\Omega_{R, k_{\parallel, res}}$ from the uncoupled ISBT energy. See Fig. 2.17. $\Omega_{R, k_{\parallel, res}}$ is the frequency at which the energy in the system cycles between electronic and photonic form, at resonance (sec. 1.3.2.2.).

We see from equation 2.64 that Rabi splitting only occurs if

$$\Omega_{R, k_{\parallel}} > \frac{|\gamma_{ISBT} - \gamma_{MC}|}{2}, \quad (2.65)$$

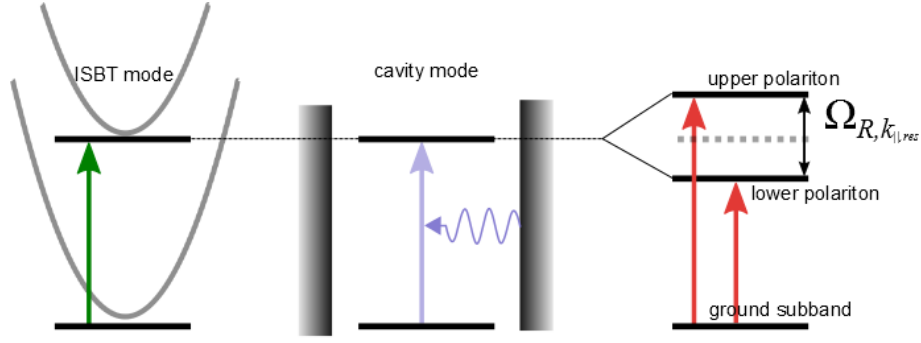


Figure 2.17.: Resonant coupling between the ISBT mode and cavity mode leads to a splitting of the excited state at energies $\sim \pm \frac{1}{2}\Omega_{R,k_{||,res}}$ from the original excited state energy.

which is the condition for the system to be in the strong coupling regime.

As mentioned, with respect to the waveguide modes, dispersion of modes is actually measured experimentally on the $\varphi - \omega$ plane, and conversion to the $k_{||} - \omega$ plane is through equation 2.39. On the $\varphi - \omega$ plane the modes are sometimes given the prefix ‘virtual’.

In the $\varphi - \omega$ plane it is possible to describe the UPB and LPB in terms of eigenfrequencies with an equation practically identical to equation 2.63, but with the variables dependent on φ , not $k_{||}$:

$$\begin{aligned} \tilde{\omega}_{UPB,LPB} = & \frac{\omega_{ISBT} + \omega_{MC}(\varphi) - i(\gamma_{ISBT} + \gamma_{MC})/2}{2} \\ & \pm \frac{1}{2}\sqrt{\Omega_{R,\varphi}^2 + (\omega_{ISBT} - \omega_{MC}(\varphi) - i(\gamma_{ISBT} - \gamma_{MC})/2)^2} \quad (2.66) \end{aligned}$$

In this equation it is vital to note that the coupling strength is not the same magnitude as in the $k_{||} - \omega$ plane, with $\Omega_{R,\varphi} > \Omega_{R,k_{||}}$. Therefore at resonance angle, φ_{res} , the quantity $\Omega_{R,\varphi_{res}}$ does *not* correspond to the vacuum Rabi frequency, and does not parameterise the Rabi oscillations (flopping) of energy levels, though it could be considered a *virtual* vacuum Rabi frequency. Nonetheless, $\Omega_{R,\varphi}$ still characterises the Rabi splitting as measured in spectra and will thus be referred to in the experimental section; but note that it is physically not the same thing as the vacuum Rabi frequency.

2.4.3. Expression of ICP linewidth

As introduced in the waveguide cavity modes section (sec. 2.3.2), the imaginary part of a mode's complex frequency has a correspondence to the mode linewidth:

$$\gamma_{UPB(LPB)} = -2\text{Im}(\tilde{\omega}_{UPB(LPB)}). \quad (2.67)$$

At resonance ($\omega_{ISBT} = \bar{\omega}_1$) the linewidth is simply the arithmetic average:

$$\gamma_{UPB} = \gamma_{LPB} = \frac{1}{2}(\gamma_{ISBT} + \gamma_{MC}). \quad (2.68)$$

More generally, the ICP linewidth is found by substituting equation 2.63 into equation 2.67, leading to a weighted average of the ISBT and microcavity linewidths [84]:

$$\begin{aligned} \gamma_{UPB} &= \mathcal{X}^2(k_{\parallel})\gamma_{ISBT} + \mathcal{C}^2(k_{\parallel})\gamma_{MC} \\ \gamma_{LPB} &= \mathcal{C}^2(k_{\parallel})\gamma_{ISBT} + \mathcal{X}^2(k_{\parallel})\gamma_{MC} \end{aligned} \quad (2.69)$$

where $\mathcal{X}$ and $\mathcal{C}$ are the *Hopfield coefficients* which take the values [84]

$$\mathcal{X}^2(k_{\parallel}) = \frac{\omega_{ISBT} - \omega_{MC}(k_{\parallel}) + \sqrt{(\omega_{ISBT} - \omega_{MC}(k_{\parallel}))^2 + \Omega_{R,k_{\parallel}}^2}}{2\sqrt{(\omega_{ISBT} - \omega_{MC}(k_{\parallel}))^2 + \Omega_{R,k_{\parallel}}^2}} \quad (2.70)$$

and

$$\mathcal{C}^2 + \mathcal{X}^2 = 1. \quad (2.71)$$

Hence we see that, for example, when the UPB dispersion is close to the ISBT mode frequency, $\mathcal{X} \rightarrow 1$ and $\mathcal{C} \rightarrow 0$; the UPB thus has a linewidth similar to that of

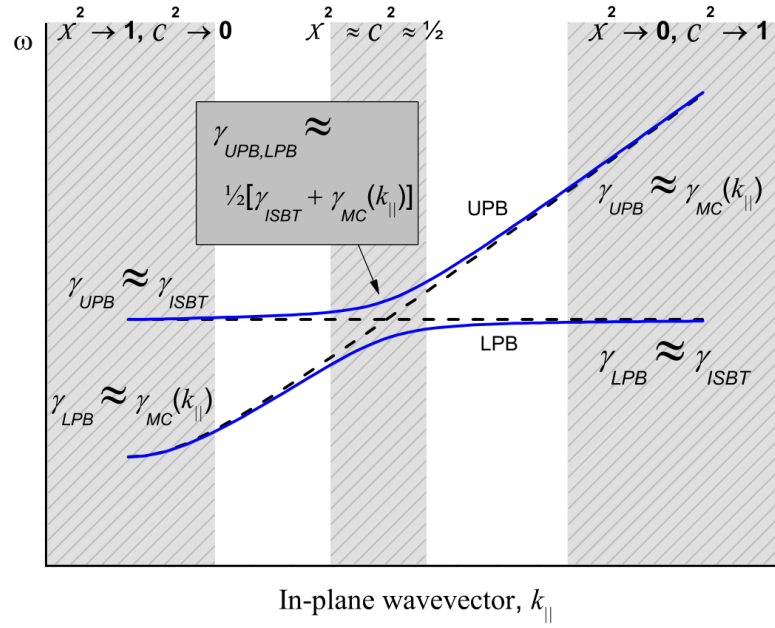


Figure 2.18.: The behaviour of Hopfield coefficients with varying $k_{\parallel}$. The central region, around the anticrossing, has approximately equal Hopfield coefficients, with the ICP (UPB and LPB) linewidths the arithmetic average of the ISBT and microcavity linewidths. The left and right hand regions correspond to the ICPs becoming dominated by either their electronic (ISBT) or their photonic components. For example, in the left hand region the UPB closely follows the ISBT mode, and thus has the linewidth properties of the uncoupled, bare ISBT.

the bare ISBT, with minimal or no contribution from the microcavity linewidth. Fig. 2.18 summarises the behaviour of equation 2.69.

The Hopfield coefficients weight the proportions of which the ICP is ISBT-like or photonic. Thus it has a wider implication to the nature of the ICP quasi-particle, and has a direct role in the quantum description of the ICP. In fact, the Hopfield coefficients describe the probability of finding the ICP in the photonic or ISBT state [22, 53]:

$$\begin{aligned} |\Psi_{LPB}\rangle &= \mathcal{X} |a, n\rangle + \mathcal{C} |b, n-1\rangle \\ |\Psi_{UPB}\rangle &= -\mathcal{C} |a, n\rangle + \mathcal{X} |b, n-1\rangle \end{aligned} \quad (2.72)$$

where a refers to the ground state of the ISBT, and b the excited state. n is the photon number. Thus, the physical properties of the ICP are dictated by the

Hopfield coefficients.

2.4.4. Cavity polariton linewidth narrowing

2.4.4.1. Previous ICP linewidth studies

Previous work on our NRC sample by Mary Matthews [64] indicated that ICP linewidths were narrowed compared to the expected values from the STCO model with the bare ISBT and empty cavity mode linewidths. This was not the first time that ICP linewidths had been seen to be narrowed with respect to predictions; our collaborators (Dupont *et al.*) had in fact seen ICPs with particularly narrowed linewidths in similar ICP samples, active at longer wavelengths, in 2007 [55].

The explanation given by Dupont *et al.* followed the lead of observations in the exciton cavity polariton system. In the mid-90s, it was found that the linewidths of exciton cavity polaritons at resonance were often narrower than the value predicted from the bare exciton and bare cavity linewidths. The prevailing explanation given for these observations was based on the exciton energy distribution having an inhomogeneous broadening. The theory is outlined below, though we do not presume in advance that it applies to the NRC sample. It is, however, the prevailing cavity polariton linewidth narrowing theory, and so is included so that it may be properly considered.

2.4.4.2. Houdré's theory of narrowing due to inhomogeneity in excitonic systems

In the theoretical work by Houdré *et al.* [90, 91], the exciton linewidth, δ_e , was described as a Voigt profile, with FWHM as equation 2.33, composed of a homogeneous Lorentzian linewidth, γ_e , and an inhomogeneous Gaussian linewidth, ξ_e . It was found that if the splitting of the polariton modes was substantial, the exciton would only contribute its Lorentzian (homogeneous) linewidth component to the final cavity polariton linewidth. In effect, the inhomogeneous broadening, ξ_e , was eliminated from the equation. The polariton linewidth was therefore described by the weighted average of γ_e , and the cavity linewidth, γ_{cav} (as in equation 2.68 for

the case of ICPs):

$$\delta_{pol}^{>} = \frac{1}{2}(\gamma_e + \gamma_{cav}), \quad (2.73)$$

where the superscript $>$ indicates that this is the case of larger polariton vacuum Rabi splitting.

It was then shown that, should the cavity polariton vacuum Rabi splitting be *smaller* than the exciton broadening (but still in strong coupling regime), the *full* Voigt FWHM of the bare exciton would contribute to the polariton linewidth. The polariton linewidth at resonance was given in Ref. [90] as

$$\delta_{pol}^{<} \approx \frac{1}{2}(\delta_e + \gamma_{cav}), \quad (2.74)$$

though this form of averaging is only strictly valid for Lorentzian linewidths. The superscript $<$ indicates the case of smaller Rabi splitting. Nevertheless, equation 2.74 is the average of the bare, uncoupled linewidths of the exciton and microcavity for this system. Therefore, the linewidth given by equation 2.73 is *subaverage*, and may be considered *narrowed* with respect to the estimated linewidth. The result is that

$$\delta_{pol}^{>} < \delta_{pol}^{<}. \quad (2.75)$$

i.e. the large polariton vacuum Rabi splitting leads to a reduction in the cavity polariton linewidth, compared to the same system, but with small Rabi splitting. See Fig. 2.19.

The theory predicted that not only were the cavity polaritons narrower than the arithmetic average of the exciton and cavity linewidths, but could even be narrower than the bare exciton and cavity linewidths individually, i.e. $\delta_{pol}^{>} < \delta_e$ and $\delta_{pol}^{>} < \gamma_{cav}$. Hence, by forming a cavity polariton with large Rabi frequency, it became possible to extract narrow linewidths from systems with large broadening. Experimental evidence for this effect came soon after the theoretical treatment [91].

The explanation forthcoming from the model was that, when the cavity polariton

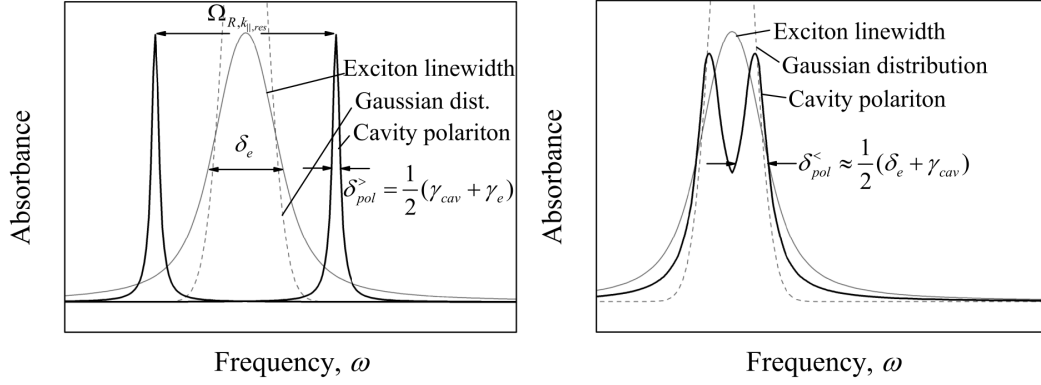


Figure 2.19.: The behaviour of cavity polariton absorbance linewidths in the presence of the exciton broadening, of total linewidth, δ_e , in the strong coupling regime at resonance, with vacuum Rabi frequency, $\Omega_{R,k_{||,res}}$. When the interaction is stronger (left hand graph), the polariton frequencies do not overlap with the inhomogeneous (Gaussian) broadening component of the exciton linewidth, and therefore are not influenced by it. The polaritons only feel a contribution from the homogeneous component (γ_e) of the exciton linewidth, and their linewidths reflect this (see expression in figure). When the interaction is weaker (right hand graph), the polariton frequencies are within the frequency range of the inhomogeneous broadening; the polariton linewidths therefore are the average of the cavity linewidth (γ_{cav}) and the total exciton linewidth. The polariton linewidths are therefore narrower when the vacuum Rabi splitting is larger. See Ref. [90].

Rabi splitting was substantial, its frequency ω coincided with the tails of the exciton absorption profile.

A property of the Voigt profile (if the exciton were inhomogeneously broadened) is that in the tails of the distribution, the form of the curve is increasingly indistinguishable from the constituent Lorentzian distribution. This occurs because Gaussian distributions tail off far quicker than the Lorentzian, even when the Gaussian and Lorentzian FWHMs are similar. (As seen previously in Fig. 2.9.)

It was hypothesised that a cavity polariton in the vicinity of the tails of the exciton absorption profile would effectively only be interacting with the homogeneously-broadened exciton. The cavity polariton could not possibly see the effects of inhomogeneous disorder and its Gaussian broadening - the information about the Gaussian broadening was effectively hidden. Since the Lorentzian FWHM is narrower than the Voigt FWHM, this results in a narrower cavity polariton than expected from a simple average of the ISBT and waveguide cavity linewidths.

2.4.4.3. Applicability to the ICP system

The aim of the thesis is to assess the behaviour of the ICP linewidths. Linewidth narrowing was indeed found, as Mary Matthews had found previously, and so the applicability of the above theory will be tested in sec. 3.7. Whether the above theory is valid depends mainly on whether the ISBT MQW is inhomogeneously broadened, which, as discussed, is not necessarily so.

Otherwise, the observations of linewidth narrowing will require a different explanation.

3. Experimental observation of linewidth-narrowing in ICPs

Here, the NRC sample and experiment is described in detail. The NRC sample was grown by the team of Emmanuel Dupont at the National Research Center of Canada, and was first reported in Ref. [44], denoted there as ‘NT3557’.

The particular result of importance is the observation of a linewidth-narrowing effect acting on the ICPs. Our results were reported in Ref. [92].

3.1. MQW waveguide microcavity sample

The NRC sample is an MQW slab within a waveguide microcavity, grown on a 500 μm -thick semi-insulating GaAs substrate. See Fig. 3.1. It comprised a 0.8 μm -thick n-GaAs coupling mirror layer doped at $N_D = 2 \times 10^{18} \text{ cm}^{-3}$; a 140 repeat MQW; followed by a back mirror comprising a 0.394 μm -thick n-GaAs layer, with the same doping as the coupling mirror. The whole sample was capped, top and bottom, with a 200 nm/9 nm Au/Ti layer.

Each period of the MQW was composed of a 6 nm GaAs well of equivalent 2D electron density $N_s = 2 \times 10^{10} \text{ cm}^{-2}$, and a 29 nm $\text{Al}_{0.24}\text{Ga}_{0.76}\text{As}$ barrier, with the central 20 nm Si-doped at a concentration of 10^{16} cm^{-3} . The QWs were of depth $V_0 \approx 180 \text{ meV}$ and the ISBT was a bound to quasi-bound transition, which at room temperature was of frequency $\omega_{ISBT} = 129.1 \text{ meV}/\hbar$ (see sec.3.3.4). The QW is shown in Fig.3.2 and the calculated oscillator strength is $f_{12} = 0.96$ (see model, sec.3.7). The MQW slab filled the entire microcavity, and had a total thickness of $L_{MQW} \simeq L_{MC} = 4.9 \mu\text{m}$.

The NRC sample was cleaved to a length of $2 \times n_{GaAs} \times d \approx 3.3 \text{ mm}$, where $n_{GaAs} = \sqrt{\epsilon_s} \approx 3.3$ is the refractive index of the GaAs substrate and d is the thickness of

Figure 3.1.: Schematic of the NRC sample. 140 periods of a GaAs/AlGaAs QW sit within a waveguide microcavity. Probe radiation is coupled into the NRC sample from the substrate layer via frustrated TIR, at angle φ . The NRC sample is capped top and bottom with gold to aid waveguiding of radiation.

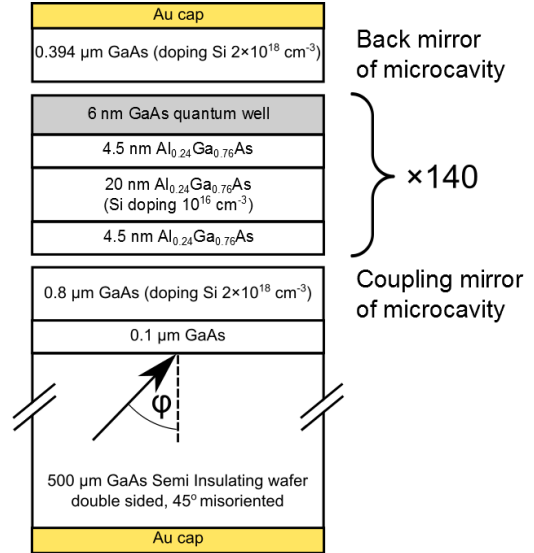
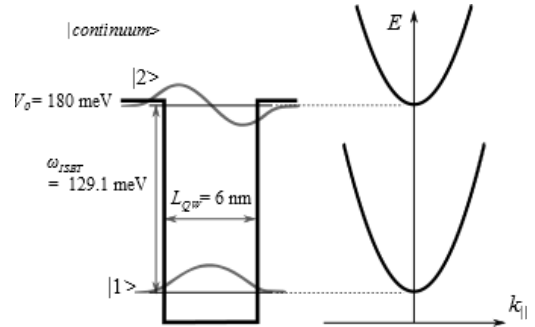


Figure 3.2.: Band structure of QW showing bound to quasi-bound ISBT. Also shown is the energy dispersion from this ISBT.



the entire NRC sample. The end facets of the NRC sample were polished to allow coupling of light into and out of the 500 μm -thick substrate. The NRC sample's length allowed light to make two reflections within the sample, so that the light would be co-linear before and after the sample: one reflection from the Au cap, and the other reflection from the coupling mirror of the microcavity waveguide, at incident angle φ (see Fig. 3.3). The reflection from the coupling mirror was due to total internal reflection (TIR), owing to the high doping of the waveguide mirrors lowering the refractive index of the GaAs. The coupling mirror is sufficiently narrow for the occurrence of frustrated TIR (see sec. 2.3.1.2, Fig. 2.13) and radiation thereby couples into the waveguide [93]. It is approximated that the angle within the waveguide microcavity is equal to the coupling angle from the substrate, φ . This assumes that the refractive index is the same in the substrate and the MQW, which is not entirely true due to the anisotropic nature of the MQW, but will suffice as an

approximation.

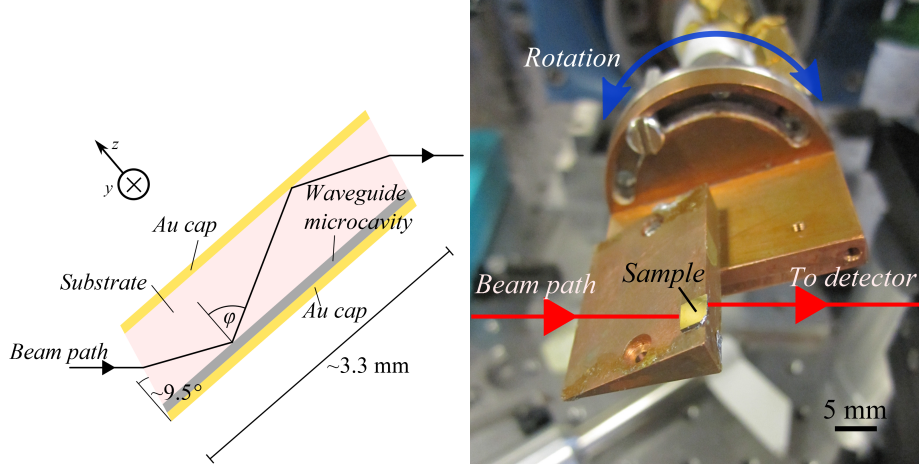


Figure 3.3.: (Left) Diagram of laser beam path through sample, showing sample structure, including position of waveguide and the $\sim 9.5^\circ$ cut. (Right) Photograph of sample, as mounted on cryostat coldfinger, with beam path and rotation orientation shown.

The GaAs mirror layers confine radiation to the waveguide as described in sec. 2.3, by TIR. The waveguide's fundamental cavity mode, $\bar{\omega}_1$, in accordance with equation 2.40 (which applies to the ideal mirror case), has a continuous range of values and is resonant with the ISBT mode, ω_{ISBT} , when the incident angle spans $\varphi \approx 70 - 80^\circ$. Due to the high 2D electron concentration of the QWs, N_s , the vacuum Rabi frequency is substantial ($\sim 3 \text{ meV}/\hbar$), since $\Omega_{R,k_{\parallel,res}} \propto \sqrt{N_s}$ (see equation 2.62). The coupling between ISBT and fundamental cavity mode is thus designed to be strongly-coupled, as described in sec. 2.4, and the two modes will therefore anticross.

To access the anticrossing range of φ requires that the end facets of the NRC sample, where the light couples into the substrate, be angled. Therefore facets were polished at $\sim 9.5^\circ$ to the z -direction, giving the NRC sample a parallelogram shape when viewed from the side. See Fig. 3.3.

3.2. Experimental procedure

3.2.1. Summary of laser spectroscopy

The laser spectroscopy setup is shown in Fig. 3.4. A laser beam from a QCL was raised to experiment height with a periscope whose upper mirror yaw was 45° to the lower mirror. This had the effect of rotating the polarisation of the QCL beam (which was initially vertical) to 45° to the bench.

This beam was then directed to a BaF_2 lens of focal length $f = 100$ mm. At the focus of the beam was positioned the NRC sample, attached with silver paste to a copper wedge, itself attached to the cold finger of the cryostat (see Fig. 3.3). The NRC sample was orientated with its y -axis parallel to the bench, and the cryostat cold finger allowed rotation of the NRC sample around an axis parallel to the y -axis.

Upon leaving the NRC sample, the beam was directed through a BaF_2 polariser which selected either vertically or horizontally polarised light. The vertical light was TM-polarised with respect to the MQW microcavity, and the horizontal was TE-polarised. Only the TM-polarised light interacted with the MQW's ISBT; the TE-polarised light took the same path and so was used as the reference spectrum.

The light was detected by a liquid nitrogen-cooled MCT detector. The laser was modulated so the signal could be improved with a digital lock-in-amplifier (LIA).

With the NRC sample at a given angle, the laser was tuned through a range of photon frequencies, and at each photon frequency the TM-polarised and TE-polarised signals were measured. The NRC sample angle was changed and the laser once more tuned through the range of photon frequencies. The process was carried out a room temperature (300 K), 77 K and 13 K.

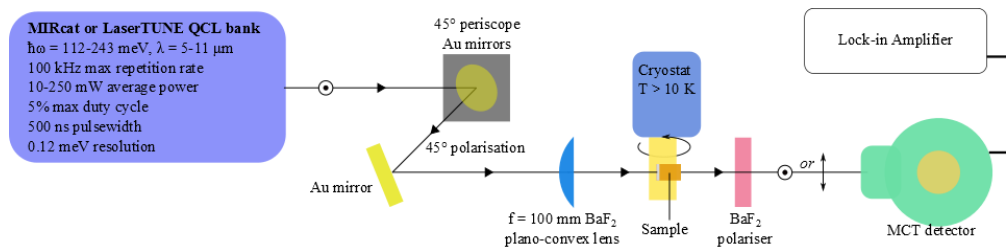


Figure 3.4.: Set up of QCL-based laser spectroscopy. The solid line indicates the path of the QCL beam from laser to detector.

3.2.2. Experimental equipment

3.2.2.1. QCL source and beam

The QCL

As first introduced in sec. 1.1.3.3, the QCL is itself a product of band structure engineering and the MQW ISBT. Recently, the development of this technology into the midinfrared spectral region has allowed a number of companies to commercialise tunable midinfrared laser sources. A QCL with a large-bandwidth ISBT may be tunable if the external cavity resonance is tunable.

Commercialised QCL-based lasers often have several QCL chips within them to extend the tuning range further still. Two such ‘QCL banks’ were used for taking the spectra of the NRC sample, primarily the MIRcat from Daylight Solutions and to a lesser extent the LaserTune 510 from Block Engineering, which had similar properties to the MIRcat and so will not be described in detail below.

MIRcat beam properties

The MIRcat laser contained four QCL chips covering the range from 111.6 - 243.1 meV/ $\hbar$ ($\lambda = 11.11\text{-}5.1\ \mu\text{m}$, $\bar{\nu} = 1961\text{-}9001\ \text{cm}^{-1}$), with a photon frequency resolution of $\sim 0.12\ \text{meV}/\hbar$. The output was pulsed, up to a repetition rate of 100 kHz. Pulses widths were 500 ns, and the maximum duty cycle was 5%. Operation was at room temperature, with the QCL chips cooled thermoelectrically (TEC), with optional water-cooling to increase uninterrupted experiment time and QCL lifespan.

The time-averaged power spectrum of the four MIRcat QCL chips (QCL1-QCL4) in pulsed operation is shown in Fig. 3.5. Maximum average power is around 25 mW for a 5% duty cycle; maximum power of a pulse is therefore $\sim 500\ \text{mW}$. However, this is not typical of the entire range, and, especially at lower photon frequencies, the average power is below 10 mW. A lock-in-amplifier was thus necessary in order to measure reliable signal, since the lower photon frequencies of the QCL were relevant to our NRC sample. Power was measured with a Thor labs S302C thermal absorber with the PM100D power meter.

The beam, across most of the photon frequency range, was a Gaussian TEM₀₀ mode with $1/e^2$ radius $\sim 3\ \text{mm}$, a circular beam and $M^2 \approx 1$. Viewing the beam on an infrared camera (CEDIP, see Fig. 3.6) revealed that the beam had some lateral

Figure 3.5.: Time-averaged power in pulsed operation versus photon energy for the entire range of the MIRcat QCL, as measured with the Thor S302C thermal absorber power meter.

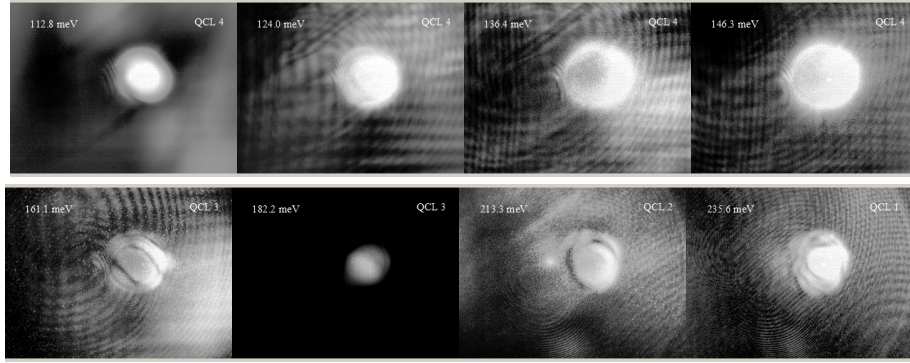
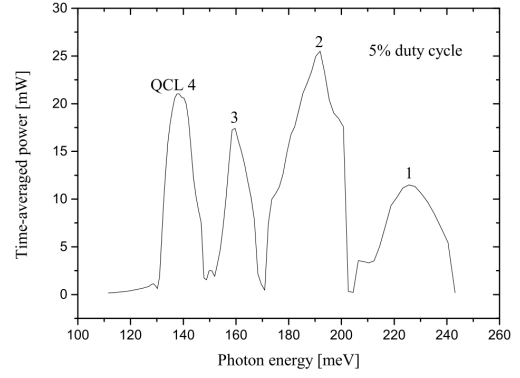


Figure 3.6.: CEDIP images of the MIRcat laser beam profile for the four QCL chips. It can be seen that the QCL4 beam was a well-behaved TEM_{00} , though with slight size variation and some positional variation. QCL3 showed some signs of multimodal operation, though not for the entire range. QCL2 and 1, which were less important for the experiment, occasionally showed some suspected multimodal behaviour.

movement as the photon frequencies were tuned through, though only by fractions of millimetres, after a metre of travel. The beam radius changed slightly (< 0.5 mm) across the photon frequency range, and, at some photon frequencies, showed signs of possible multimodal behaviour, though not on the QCL chip most important to the experiments conducted (QCL4).

This variation in position and radius required that the NRC sample position be reoptimised periodically, when tuning through the photon frequencies, for best signal. Repositioning was necessary also because the position of focus was altered with wavelength variation (due to the lens material dispersion). A red laser diode was used to colour the QCL beam, and also used to determine the angle of the NRC sample, as an optical lever.

Some drift of the QCL power was also noted after long periods of use. To deal with this, TM and TE-polarised signals were taken in succession such that the output properties of the laser and detector were as constant as possible for each point in the spectrum.

Improved angular resolution with laser spectroscopy

Owing to the TEM_{00} and $M^2 \approx 1$ properties of the beam, the size of the focussed spot was constrained merely by the focal length of the lens employed, $f = 100$ mm. It was important to focus the beam to a radius smaller than half the NRC sample substrate thickness (~ 250 μm), but not too small so as to decrease the Rayleigh range of the beam, z_R . The Rayleigh range is the distance from the beam waist to where the beam radius has increased by a factor of $\sqrt{2}$. A focussed Gaussian beam has a total angular spread (twice the *divergence*) of $2\varphi_{div} = 2\lambda/\pi w_0$ where λ is the wavelength and w_0 is the beam waist. However, within the Rayleigh range, the beam has a narrower spread and is closer to being a parallel beam.

The beam waist, w_0 , was measured with the knife-edge technique (see sec. A.1), and the resultant focussed beam radii for a handful of photon frequencies is shown in Fig. 3.7 (for the beam in air). The data have been fitted with the Gaussian-beam radius equation for an $M^2 \approx 1$ beam, and show that the beam waist is $w_0 \approx 100$ μm for the various photon frequencies. The Rayleigh range in air is longer than the NRC sample length. The Rayleigh range is even longer within the substrate; for example, at the frequency $112.8 \text{ meV}/\hbar$ the Rayleigh range is $z_R \approx 19.2$ mm (the NRC sample is ~ 3.3 mm long). This gives an angular spread within the substrate of $\Delta\varphi \sim 0.01^\circ$, whereas the Gaussian beam as a whole had an angular spread of $2\varphi_{div} = 0.8^\circ$.

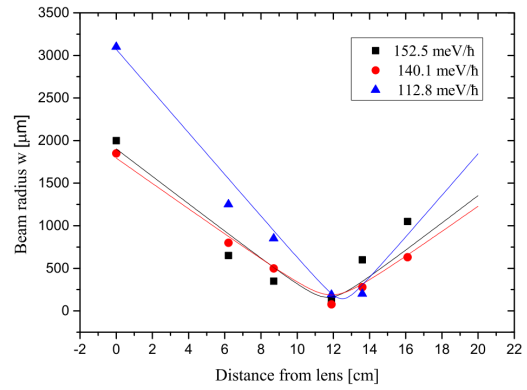
The angular spread of the beam is of critical importance to the spectral resolution of the measurement, i.e. for measuring linewidths. The frequency of a waveguide cavity mode, for example, depends on the coupling angle/angle of propagation in the waveguide. The spatial centre of a focussed beam will resonate with a particular frequency of the mode. However, at the edges of the beam's angular spread the beam will be incident on the waveguide at a different angle, which will therefore resonate with a higher or lower frequency mode. The result is that, in a spectrum, the resonant mode is a Gaussian convolution of the resonant peaks across a finite angular range. See Fig. 3.8.

Therefore, the linewidth of a peak depends somewhat on the angular spread of the focussed beam, $\Delta\varphi$, and on the steepness of the dispersion, $\frac{d\omega}{d\varphi}$. As a simple estimate, the contribution to a mode's spectral resolution is

$$\Delta\omega = \frac{d\omega}{d\varphi} \Delta\varphi. \quad (3.1)$$

Non-laser light sources, where the beam may not be Gaussian, do not focus in as well-behaved a manner, and $\Delta\varphi$ will be substituted in equation 3.1 for the divergence of the focussed light, $2\varphi_{div}$. Since, typically, $2\varphi_{div} > \Delta\varphi$, the resolution of a non-laser light source will be lower, i.e. $\Delta\omega$ will be larger, than with a laser light source. This is a *critical* feature of the work in this thesis, that, by using a laser source for spectroscopy, we can measure the linewidths of the ICPs to a higher resolution than previously achievable.

Figure 3.7.: Beam radius for three different photon frequencies, fitted as $M^2 \approx 1$ Gaussian modes, as measured with the knife-edge technique.



A note on intensity dependence of ISBTs

The peak intensity of the MIRcat, when focussed to this size, does not exceed $\sim 1 \text{ MWcm}^{-2}$, and is around 100 kWcm^{-2} for photon frequencies pertinent to the NRC sample. Previous work by Mary Matthews indicated that at values ~ 100 times this intensity, saturation of the ISBTs begins to affect the strength of the coupling, and the vacuum Rabi splitting drops [64]. At the MIRcat's intensities, the carriers are still, to a good approximation, all in the lower subband, therefore the ISBT did not saturate (and become transparent); therefore the vacuum Rabi splitting did not change magnitude.

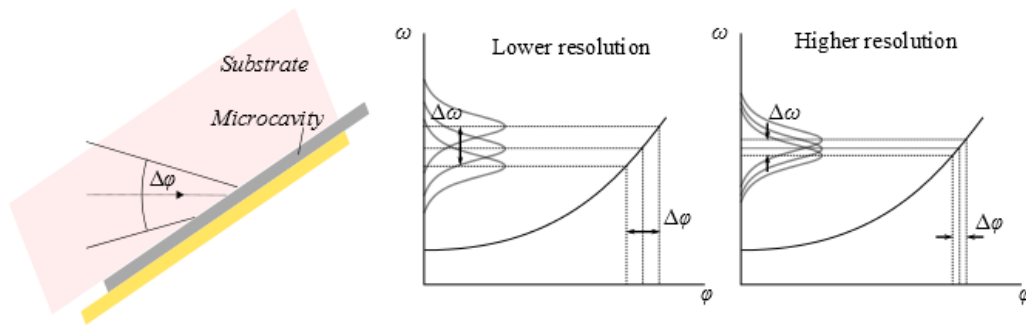


Figure 3.8.: Illustration of the effect of angular divergence on the energy resolution when measuring the optical modes of a waveguide microcavity. The spread in beam angle, $\Delta\varphi$, results in a spread of mode frequencies probed, $\Delta\omega$, at any one angle.

3.2.2.2. Cryostat

In order to probe the temperature dependence of the ICPs, the NRC sample is housed within a cryostat. Temperature-dependent properties of the semiconductors composing the NRC sample are expected to have an influence on the ICPs, in particular, through the freeze-out of carriers at low temperature, and the effect of temperature on phonon populations and other broadening mechanisms.

The cryostat was a nitrogen-compressing CTI-Cryogenics Model 22 refrigerator, controlled by LakeShore 331 temperature controller. It was capable of temperatures as low as 10 K, with a thermoelectric heater regulating the cooling with a PID algorithm. Any wires within the cryostat were bound down with PTFE tape to avoid thermal contact, and care was taken to avoid the trapping of air within the tape bindings. The cold finger was shrouded with a cold shield to reduce radiative heating effects. The outer casing had a series of BaF_2 windows to allow the passage of the laser beam. The chamber was pumped down to $\sim 10^{-5}$ atm with a BOC Edwards RV5 turbo vacuum pump, prior to cooling. The process of cooling to 13 K took approximately 2 hours.

3.2.2.3. Detector and lock-in amplifier

The detector used for midinfrared detection was an MCT (HgCdTe) direct band gap detector¹ with a large active area ($\sim 9 \text{ mm}^2$), and a sensitivity range span-

¹The MCT detector element is formed from the alloy of the semimetal HgTe (with zero band gap) and the semiconductor CdTe (with 1.5 eV direct band gap); the band gap is tunable with the

ning 49-413 meV/ $\hbar$ (peaking at 113 meV), with IR detector figure of merit $D^* \approx 10^{10} \text{ cm}\sqrt{\text{Hz}}\text{W}^{-1}$. The detector bandwidth was sufficient to detect the 100 kHz repetition rate of the QCL laser. The detector responded linearly below an output of 1.4 V, a voltage which corresponded to incident laser powers much higher than used in the experiment.

The detector was a Stanford Research Systems (model SR844) digital LIA. This was set to a time constant of 1 s and a cut off filter of 12 dB/octave. The LIA was capable of locking onto signals at frequencies of up to 1 MHz.

The LIA was connected to computer via GPIB serial port, and could thereby be linked to lab automation software, LabVIEW.

3.2.3. Spectral acquisition

The laser signal arriving at the detector was a measure of the transmissivity of the whole sample (and everything else between laser and detector), and the reflectivity of the microcavity waveguide within the NRC sample. The desired part of this signal was the reflectivity of the microcavity waveguide, therefore the transmissivity part needed to be ratioed out of the signal, since it was merely background.

For the NRC sample, we exploited the fact that the ISBT is only active to one polarisation orientation (TM-polarised). The TE-polarised beam could therefore be used as the background reference, provided its intensity was the same as the TM-polarised beam. The laser beam used here had both TE and TM polarisations in equal intensity, selectable with an IR polariser. The reflectance-absorbance of the microcavity waveguide (a unitless quantity) was thus calculated through the Beer-Lambert law, similar to equation 2.15, as

$$\alpha_{ICP} = -\ln\left(\frac{I_{TM}}{I_{TE}}\right) \quad (3.2)$$

where $I_{TM(TE)}$ is the magnitude of the TM- (TE-) polarised beam at the detector. External angles from air to substrate were in the range of 20° angle of incidence - this is far from the Brewster angle between GaAs and air (~74.2°), and so the

proportion of Cd, and so midinfrared detection is possible to photon frequencies of 1.5 eV/ $\hbar$.

reflectance of TM and TE polarisations is very similar, being around 0.3 in both cases.

An absorbance measurement of a waveguide such as this will include the effects of a resonant photon frequency being transmitted in the plane of the waveguide, and therefore away from the reflected beam path. The resonant mode does not couple out of the cavity because the field has nodes at the edge of the cavity.

3.3. ICP spectra and discussion

We begin by looking at the spectra of the ICPs at room and low temperatures, and derive the ICP dispersion curves from these. From these spectra we also observe the linewidth-narrowing effect of the ICPs, which is covered in sec. 3.5.

3.3.1. Room-temperature spectra

The room temperature spectra are presented in Fig. 3.9 across a range of coupling angles spanning the anticrossing of the ISBT and the fundamental cavity mode. Spectra are separated by $\varphi \sim 0.3^\circ$ which corresponded to steps of 1° for the external angle of incidence. In these absorbance spectra, it is possible to see the peaks of the upper polariton branch (UPB) on the high energy side and the peaks of the lower polariton branch (LPB) on the low energy side. The anticrossing is clearly distinguishable and the vacuum Rabi splitting substantial, so the system is thus in the strongly coupled regime.

Derived from these spectra is the dispersion curve in the $\varphi - \omega$ plane, and, using equation 2.39, the $k_{\parallel} - \omega$ plane. From these dispersion curves can be derived the minimum ICP separation, which in the case of the $k_{\parallel}$ -dependent dispersion corresponds to the vacuum Rabi frequency, $\Omega_{R,k_{\parallel},res}$, as shown more closely in Fig. 3.12. The vacuum Rabi splitting of ICP modes is exaggerated in the $\varphi - \omega$ plane compared to in the $k_{\parallel} - \omega$ plane.

The UPB tends towards the frequency of the bare ISBT at low angles, and in fact is fully ISBT-like far enough away from the anticrossing point (at the lowest angles), in accordance with the Hopfield coefficients (sec. 2.4.3, equation 2.70). In this way, the bare ISBT position is represented by the UPB at $\varphi = 62.0^\circ$, and the position of this ‘bare’ ISBT is marked by a dashed line. See also sec. 3.3.4.

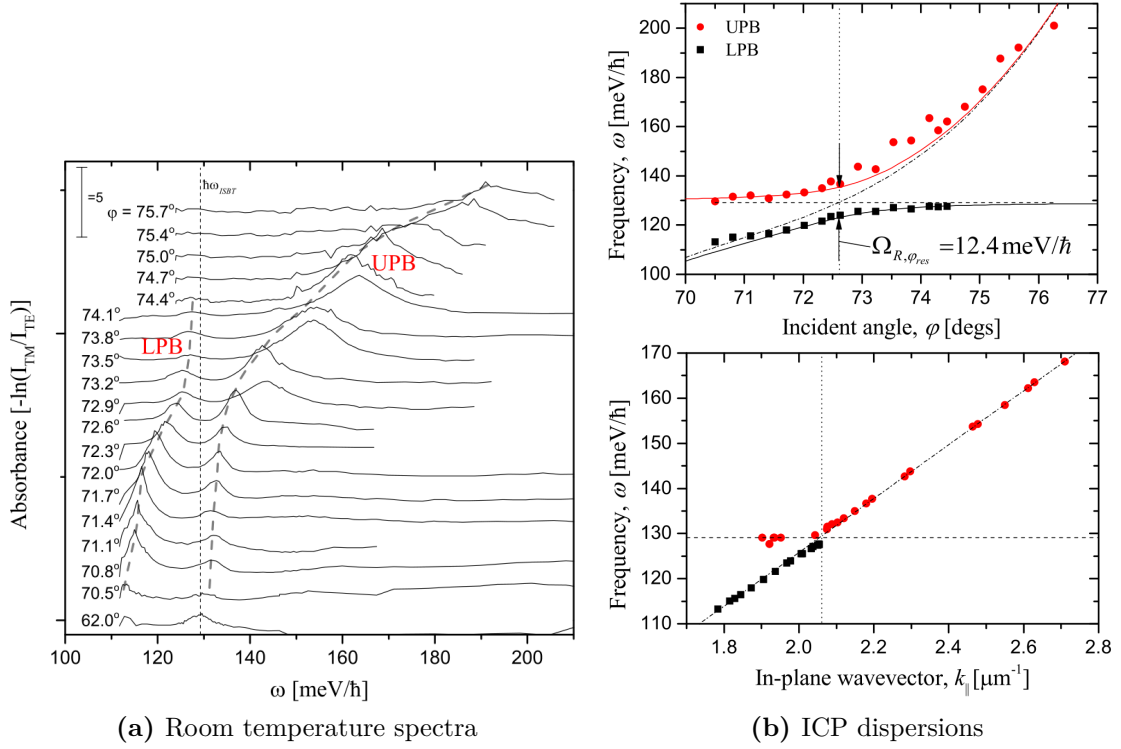


Figure 3.9.: a) Room temperature spectra, separated by $\varphi \sim 0.3^\circ$, and shown with a 1.6 offset for clarity. Also shown is the bare ISBT mode, with its peak position represented by the vertical dashed line. LPB peaks are on the low energy side of this line and UPB peaks on the high. Anticrossing of the peaks can be clearly seen. b) Related dispersion curves in $\varphi - \omega$ plane (upper) and $k_{||} - \omega$ plane (lower), with anticrossing point represented by dotted line, bare ISBT resonance, ω_{ISBT} , with a dashed line and the empty microcavity resonance, ω_{MC} , with a dash-dotted line, calculated from model described in sec. 3.4. For the $\varphi - \omega$ dispersion, the STCO model is fitted to the data (solid lines) and the minimum vacuum Rabi splitting extracted. See Fig. 3.12 for the equivalent in the $k_{||} - \omega$ plane.

In the same way, at higher angles, $\varphi > 74.4^\circ$, according to the Hopfield coefficients, the LPB is ISBT-like. However, the LPB is not visible on the absorption spectra here. One possible explanation is to do with the coupling of the ISBT with the electron plasma. As discussed in sec. 2.1.4.4 the ISBT measured is that of the pure harmonic transition coupled with the 3D plasma oscillation, leading to the depolarisation shift. At larger angles, this 3D plasma oscillation is less capable of forming due to the layered nature of the MQW restricting electron movement. This would suggest that, for strong absorption of radiation, the ISBT needs to be effectively coupled with the plasma; i.e. the many-body effect is *required*.

3.3.2. Low temperature spectra

The low temperature spectra are shown in Fig. 3.10 and Fig. 3.11, with, again, the derived dispersion curves. They are of similar form as the room temperature spectra and dispersions. Because the ISBT shifts its position, so too does the resonance of the ISBT and the cavity mode (see sec. 3.3.4). Furthermore, the cavity mode will shift because the mirror properties are dependent on the density of free carriers. At lower temperatures the number of available electrons from the donor atom decreases due to carrier freeze-out, thereby increasing the refractive index.

Linewidths also appear sharper than at room temperature, and will be dealt with in sec. 3.5. There is also a large absorption feature at higher energies, thought to be the bound-continuum transitions.

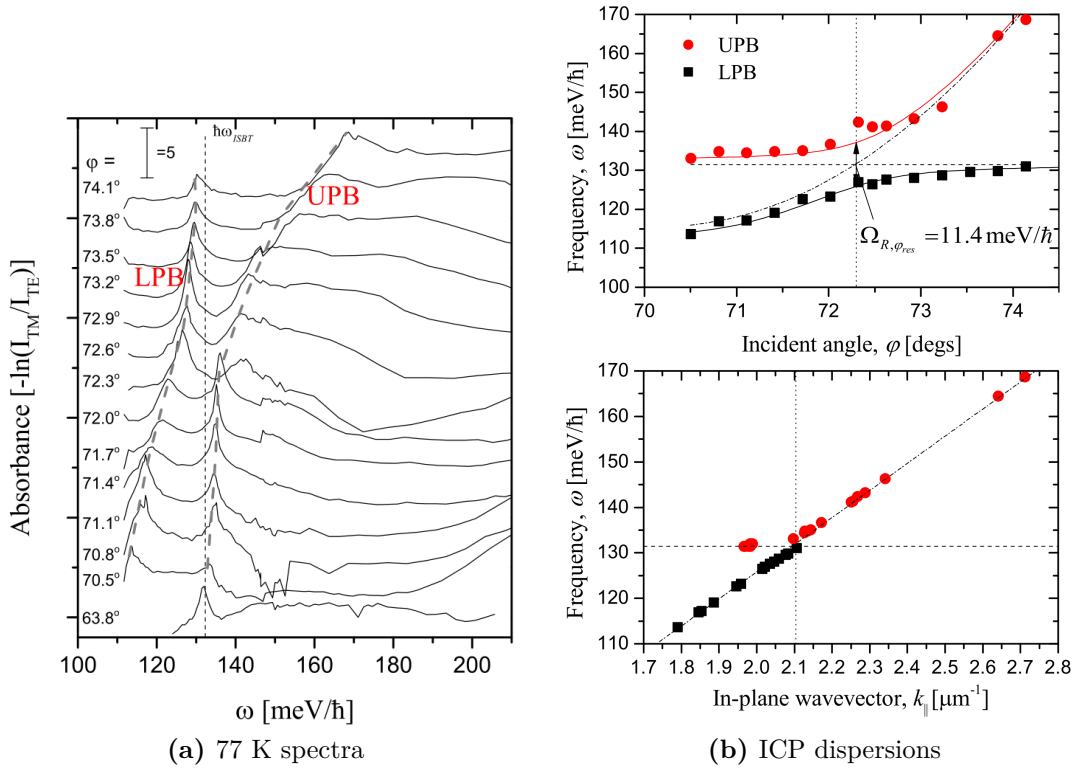


Figure 3.10.: As for Fig. 3.9, but with $T = 77 \text{ K}$.

3.3.3. Fitting of dispersion data with STCO model

The dispersion data can be fitted with the STCO model of sec. 2.4.2, with the micro-cavity waveguide mode, ω_{MC} , derived from the waveguide simulation described in

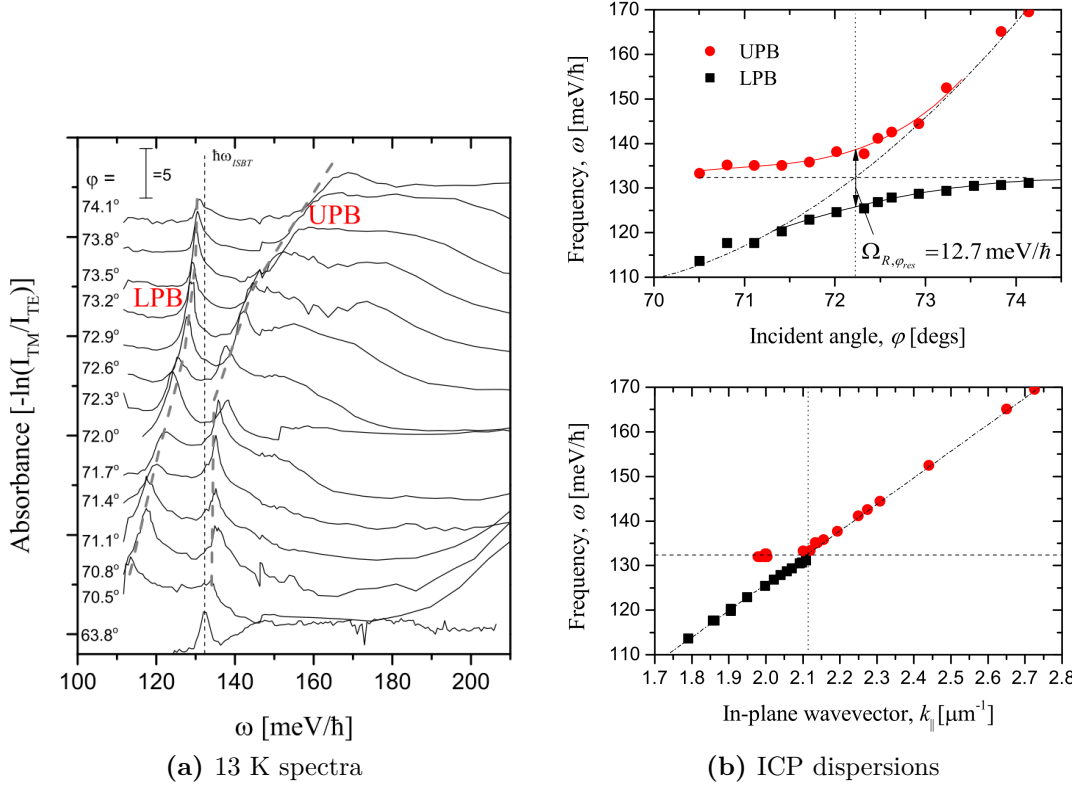


Figure 3.11.: As for Fig. 3.9, but with $T = 13 \text{ K}$.

sec. 3.4. The minimum separation and the vacuum Rabi frequency may be extracted from these fits. See Fig. 3.12. Values of the vacuum Rabi frequency, $\Omega_{R,k_{\parallel,res}}$, derived from the dispersion curves, are given in Tab. 3.1. Also shown in the table are the wavevectors and angles of anticrossing ($k_{\parallel,res}$ and φ_{res}), and the vacuum Rabi splitting magnitude at resonance in the $\varphi - \omega$ plane ($\Omega_{R,\varphi_{res}}$).

At room temperature (300 K) the vacuum Rabi frequency is $2.9 \pm 0.3 \text{ meV}/\hbar$ (0.70 THz) which is $\sim 2.2\%$ of the ISBT frequency. This confirms that whilst the system is in the strong coupling regime, it is not in the ultrastrong coupling regime. Similar behaviour of the splitting is seen at low temperature, with the vacuum Rabi frequency dropping to $2.5 \pm 0.3 \text{ meV}/\hbar$ at cryogenic temperatures; this is expected due to the freeze-out of carriers, and the resultant lowering of 2D electron density, N_s .

However, there is a slightly different story for vacuum Rabi splitting in the $\varphi - \omega$ plane (see Fig. 3.9); firstly, it is much larger, at $12.4 \pm 0.2 \text{ meV}/\hbar$ at room temperature, which is around 10% of the ISBT frequency. This larger splitting is down to

the transformation between $k_{\parallel}$ and φ (equation 2.39). The vacuum Rabi splitting initially decreases to a value of 11.4 ± 0.2 meV/ $\hbar$ at 77 K, but then increases again at 13 K to 12.7 ± 0.4 meV/ $\hbar$.

The cause of the minimum value being at 77 K (rather than at 13 K) is unknown, but may indicate an interaction between the ICPs and the phonon population. This behaviour is revealed in the $\varphi - \omega$ plane and not the $k_{\parallel} - \omega$ plane because in the latter the LPB and UPB do not overlap much in $k_{\parallel}$ and so the data are less reliable, and have larger error. We would require to know the splitting at more temperatures to understand the full temperature-dependent properties of the vacuum Rabi splitting, and discover a cause for this observation.

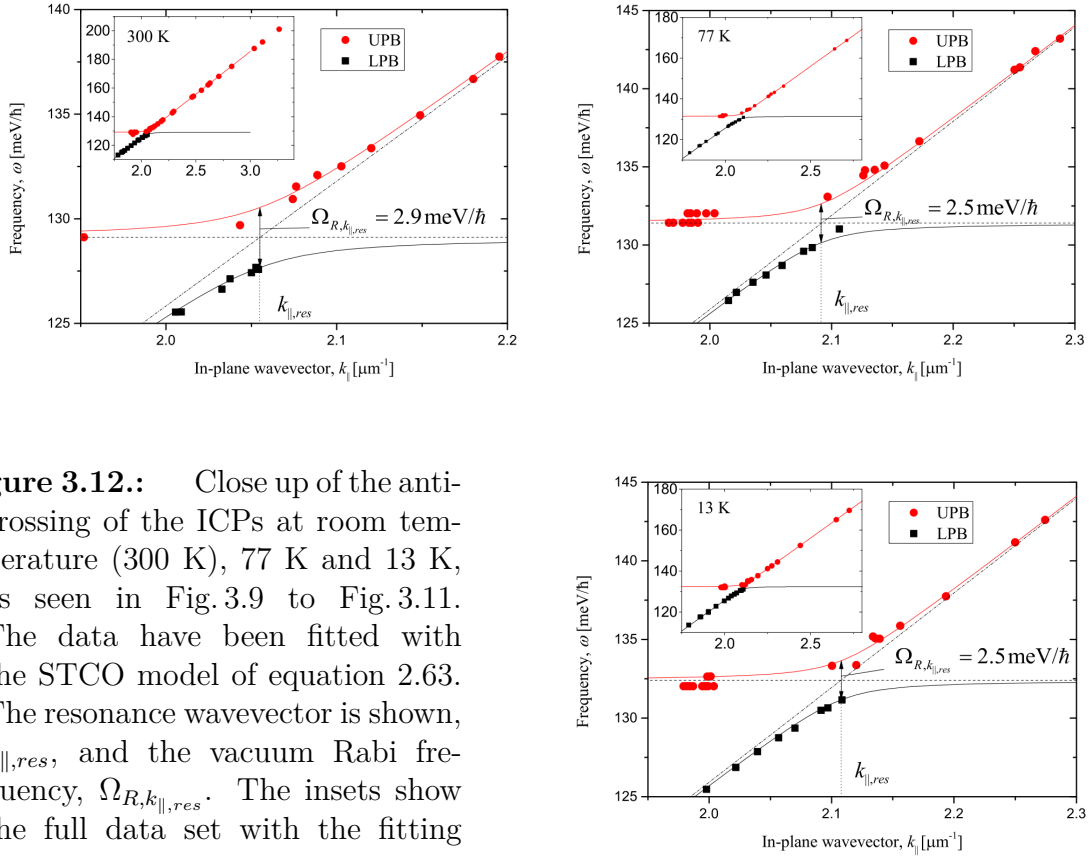


Figure 3.12.: Close up of the anti-crossing of the ICPs at room temperature (300 K), 77 K and 13 K, as seen in Fig.3.9 to Fig.3.11. The data have been fitted with the STCO model of equation 2.63. The resonance wavevector is shown, $k_{\parallel, \text{res}}$, and the vacuum Rabi frequency, $\Omega_{R, k_{\parallel, \text{res}}}$. The insets show the full data set with the fitting lines.

Table 3.1.: Derived quantities from dispersion curves of Fig. 3.9 to Fig. 3.12.

T [K]	ω_{ISBT} [meV/ $\hbar$]	$k_{\parallel, res}$ [μm^{-1}]	$\Omega_{R, k_{\parallel, res}}$ [meV/ $\hbar$]	φ_{res} [°]	$\Omega_{R, \varphi_{res}}$ [meV/ $\hbar$]
300	129.1 ± 0.5	2.06 ± 0.02	2.9 ± 0.4	72.6 ± 0.2	12.4 ± 0.2
77	131.4 ± 0.4	2.09 ± 0.03	2.5 ± 0.4	72.3 ± 0.2	11.4 ± 0.2
13	132.4 ± 0.4	2.11 ± 0.01	2.5 ± 0.4	72.2 ± 0.2	12.7 ± 0.2

3.3.4. Bare ISBT properties

Fig. 3.13 shows the UPB spectra at $\varphi = 62^\circ$, which may be used to obtain the properties of the ‘bare’ ISBT resonance. Shown also are Lorentzian-curve fittings of the peaks (fitted using the program SciDAVis), which show good agreement with the data, apart from perhaps on the high-energy side where the peak magnitude tails fall off slower than on the low-energy side, i.e. the peak appears slightly asymmetric. In Fig. 3.14 is an alternative measurement, carried out with a Fourier transform infrared spectrometer (FT/IR), by the team of our collaborator Emmanuel Dupont (who manufactured the NRC sample). There, they used the Brewster angle method of coupling into an equivalent of the NRC sample, which had been grown without the enclosing Au caps. Their measurement was at 77 K and corroborates the position and linewidth of our own 77 K bare ISBT measurement. Dupont’s team found that it was not possible to perform the same measurement at room temperature with this sample, due to insufficient absorption from the ISBTs due to the near-normal propagation through the MQWs; our own room temperature measurement is quite noisy, but the peak is discernible.

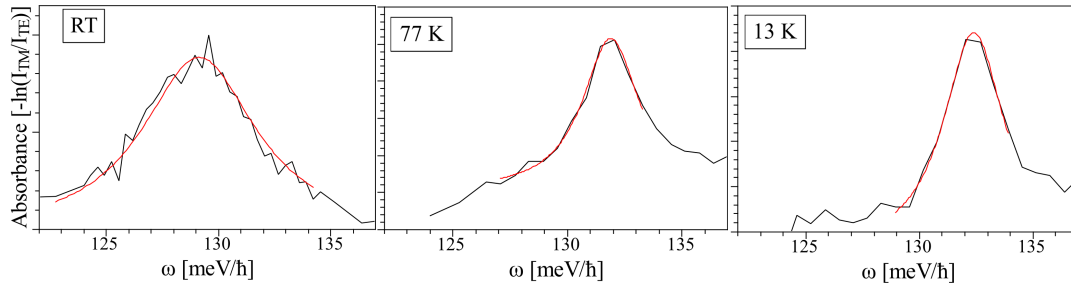


Figure 3.13.: The bare ISBT absorbance peaks as measured from the UPB at $\varphi = 62^\circ$. The peaks are fit very well with a Lorentzian profile, apart from perhaps the high-energy side of the peak. The 77 K peak may be compared to that of Fig. 3.14. The absorbance axes differ in magnitude for each graph; at room temperature the data is very noisy and absorbance is lower because of decreased occupation of $|1\rangle$, unlike in the low temperature cases.

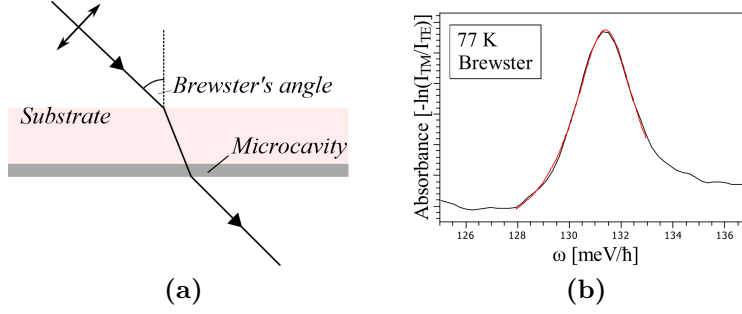


Figure 3.14.: a) Orientation of the Brewster angle measurement of the ISBT peak, showing beam path and polarisation direction through the NRC sample. There were no Au caps for this measurement. b) The resultant FT/IR spectrum and fitting of bare ISBT at 77 K .

The temperature behaviour of the bare ISBT is shown in Fig. 3.15 a), which shows that as temperature is decreased, the resonance position shifts upwards in energy². This is possibly due to the freeze-out of carriers, which reduces the Hartree potential, V_H (sec. 2.1.4.2); with less charge separation, band-bending is reduced and the ISBT is increased.

The linewidth behaviour with temperature is shown in Fig. 3.15 b), and shows the reduction in linewidth with decreasing temperature. In fact, the linewidth more than *halves* in the temperature range, from ~ 6.2 meV to ~ 2.6 meV. This may be attributed to the reduction in the phonon population (both LO and LA) thereby reducing scattering in the NRC sample as temperature is reduced. According to the literature, at 10 K, total phonon broadening, $\gamma_{\text{phonon}} = \gamma_{LO} + \gamma_{LA}$, is reduced to around ~ 1 meV [73].

The other major contributor to ISBT broadening is IRS (γ_{IRS} , sec. 2.2.1). Assuming, for the time being, that there is no inhomogeneous broadening, and that IRS and phonon broadening are the only broadening mechanisms at work ($\gamma_{ISBT} \approx \gamma_{IRS} + \gamma_{\text{phonon}}$), we can make a phenomenological estimation of their temperature-dependent behaviour. These are shown graphically in Fig. 3.15 b).

The justification for these estimations is as follows. Of the total 13 K ISBT broadening of $\gamma_{ISBT} \approx 2.6$ meV, because $\gamma_{\text{phonon}} \approx 1$ meV/ $\hbar$ is attributable to phonons [73], there is around $\gamma_{IRS} \approx 1.6$ meV/ $\hbar$ attributable to IRS.

IRS broadening is predicted to decrease with increasing temperature (but not by

²The effect is not due to the variation of band gap with temperature; the QW does not change its depth since GaAs and AlGaAs have the same temperature dependence.

more than 10% between 10 K and 300 K) because the ISBT electron is less localised at higher temperature and thus averages over the roughness more [73]. Therefore, IRS broadening, γ_{IRS} , is shown in Fig. 3.15 b) as a linear progression beginning at $\sim 1.6 \text{ meV}/\hbar$ and decreasing slightly over the temperature range.

At room temperature, to account for the full $\gamma_{ISBT} \approx 6.2 \text{ meV}/\hbar$ of broadening, this implies that phonon broadening is $\gamma_{phonon} = 4.6 \pm 0.5 \text{ meV}$.

Therefore, in Fig. 3.15 b), γ_{phonon} is estimated as a linear progression from $\sim 1 \text{ meV}/\hbar$ to $\sim 4.6 \text{ meV}/\hbar$ across the temperature range.

These two lines are added together to give the fitting to the data points shown in Fig. 3.15 b). Of course, in reality, other broadening mechanisms will make a contribution, and temperature dependences of phonon and IRS broadening are unlikely to be linear, but this estimation gives a good idea of the temperature behaviour of ISBT broadening mechanisms, and will be significant later, when the ICP linewidth is modelled (sec. 3.6).

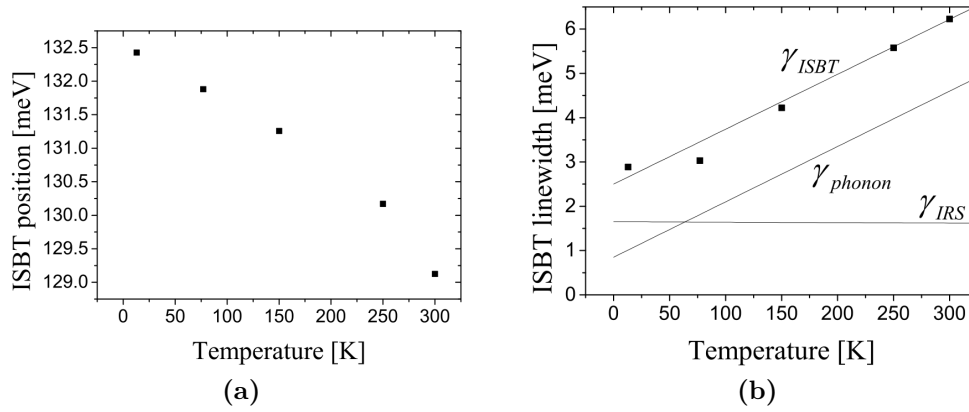


Figure 3.15.: a) Bare ISBT position against temperature. b) Bare ISBT linewidth against temperature. Shown are the experimental values of the linewidth, and a fitting based on approximate values of the two main contributors to ISBT linewidth: phonon broadening, γ_{phonon} , and IRS broadening, γ_{IRS} .

3.4. Empty microcavity dispersion and linewidths from transfer matrix formalism model

For assessing the ICP linewidths, the linewidth of the empty cavity mode, γ_{MC} , is also required. The empty cavity linewidth is given by equation 2.45, which contains a

dependence on $1/\cos \varphi$, explicitly giving an angular dependence, but also a $-\ln \sqrt{R}$ dependence. The reflectivity, R , may be taken, for simplicity, as a constant, but will itself have a φ -dependence, in which case the cavity would need to be modelled to properly determine the complete linewidth behaviour.

Here we introduce a transfer matrix formalism (TMF)-based model written in MATLAB by a member of the group, Alex Bak. Capable of modelling the full microcavity waveguide, it was also used to model the empty microcavity. The effective medium approximation used (see sec. 2.4.1.1) treated the MQW as a single anisotropic layer. Then, to model the empty cavity, the MQWs were effectively emptied of their electrons rendering the ISBTs ‘invisible’, whilst maintaining the same background dielectric properties of the microcavity. The mirror layers were modelled with a Drude dielectric response, with electron scattering time of $\tau = 1$ ps (from LO phonon scattering) and a doping concentration of $N_D = 2.6 \times 10^{18} \text{ cm}^{-3}$ (as per the NRC sample’s actual doping). The waveguide thicknesses were as in the NRC sample, and the Au cap was included. The cavity spectra were modelled against φ (giving a dispersion shown in Fig. 3.9) and the linewidths were extracted and shown in Fig. 3.17 and Fig. 3.18, as dash-dotted lines alongside the data. More details of the model are given where it is used in its full capacity, i.e. when modelling the full MQW microcavity, in sec. 3.7.

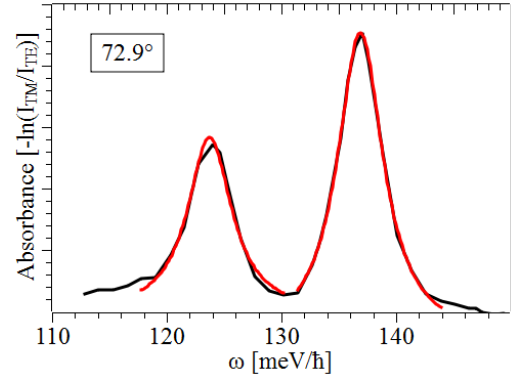
3.5. Observation of linewidth narrowing of ICPs

According to the STCO model (sec. 2.4.3), ICP linewidths should follow the Hopfield coefficient-weighted average of the bare ISBT linewidth and the empty cavity mode. However, we find that this is *not* the case for our system, prompting the subsequent development of the theory to explain the narrowed linewidths, to be detailed in sec. 3.6 and sec. 3.7.

For measuring linewidths, it is only practical to do so from spectra in the $\varphi - \omega$ plane (at constant values of φ). To measure linewidths in the $k_{\parallel} - \omega$ plane would require a very fine angle step between each spectra, and would not give as smooth, continuous peaks as seen in the $\varphi - \omega$ plane; it is also unnecessary. All the mathematics governing the properties of ICPs in $k_{\parallel} - \omega$ have equivalents in $\varphi - \omega$. The expression for the complex ICP frequency, $\tilde{\omega}_{ICP}(\varphi)$, from the STCO model, for example, was given in equation 2.66, with the ICP dispersion given by $\omega_{ICP} = \text{Re}(\tilde{\omega}_{ICP})$, and the ICP

linewidth $\gamma_{ICP} = -2\text{Im}(\tilde{\omega}_{ICP})$. Therefore, this section deals with the matter of linewidth entirely in $\varphi - \omega$, with no loss of generality. Linewidths are extracted from Lorentzian profile fittings of the ICP peaks, an example of which is shown in Fig. 3.16.

Figure 3.16.: Fitting of Lorentzian profiles to the ICP peaks at room temperature, and 72.9° angle of incidence. Linewidth is extracted from these fittings. Fittings are made in the program SciDAVis.



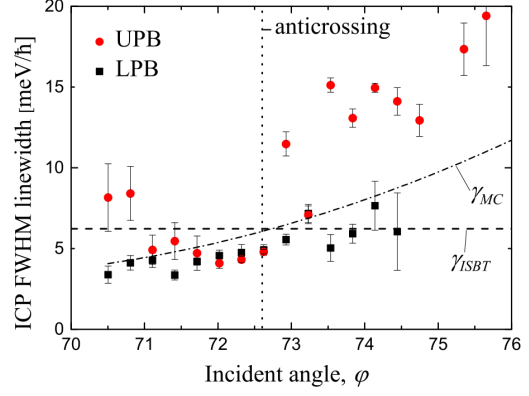
3.5.1. Room temperature ICP linewidths

The room temperature ICP linewidths are extracted from the spectra of Fig. 3.9 to give the linewidths shown in Fig. 3.17. Indicated are the angle of anticrossing, the bare ISBT linewidth (γ_{ISBT}) and the empty cavity mode linewidth (γ_{MC}). The empty cavity linewidth was derived from a model of the cavity as detailed in sec. 3.4.

3.5.1.1. Observation of subaverage ICP linewidths

By far the most significant result from this data is that, at the anticrossing point, the ICP linewidths (both UPB and LPB) are *below* the values of both γ_{MC} and γ_{ISBT} . In accordance with equation 2.68, at the anticrossing point, the ICP linewidths should be the arithmetic average of the bare ISBT and empty cavity linewidths. We see here that, at the anticrossing point, both ICP linewidths are *below* this average: they are said to be *subaverage*. This means that the STCO model is incomplete as far as describing ICP linewidths goes, and so, to predict the ICP linewidths at the anticrossing point, additional theory must be implemented. Such theory will be detailed in sec. 3.6 and sec. 3.7.

Figure 3.17.: ICP linewidths versus angle at room temperature. At the anticrossing point the measured linewidths are lower than both the bare ISBT linewidth, γ_{ISBT} , and the empty cavity mode, γ_{MC} . This means that linewidths are subaverage around the anticrossing point.



3.5.1.2. ICP linewidths away from the anticrossing point

We take a quick aside to explain that the data better follows the STCO model away from the anticrossing point. At low angles the UPB takes the linewidth properties of the ISBT, as expected, and the LPB takes on the linewidth properties of the empty cavity. At higher angles the LPB follows the linewidth of the bare ISBT and the UPB takes on the characteristics of the empty cavity mode - though we see that the measured UPB does in fact become wider than the modelled microcavity linewidth. This could indicate that, in fact, the angular resolution of the laser is wider than presumed in sec. 3.2.2.1. The UPB has the steepest dispersion in this region, so this is where inferior angular resolution would be revealed.

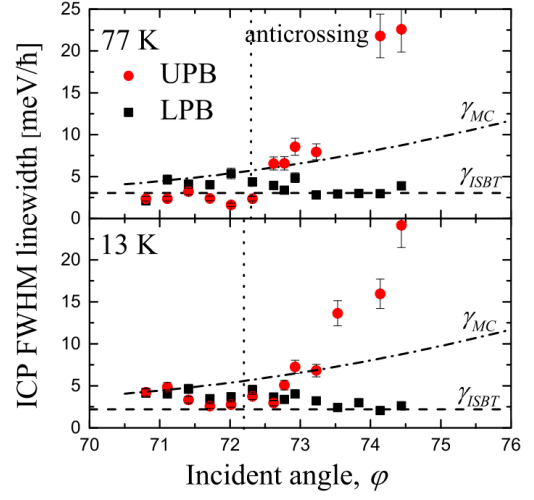
3.5.2. Low temperature ICP linewidths

The derived ICP linewidths at low temperature are shown in Fig. 3.18. The linewidths show the same trend as the room temperature measurements; however, the linewidths are not as markedly narrowed at the anticrossing point, as they were at room temperature.

The bare ISBT linewidths, γ_{ISBT} , are shown, and, within error, the ICP linewidths do not drop far below this. The same empty cavity linewidth, γ_{MC} , is shown as for the room temperature case. The reduction of carriers, as the NRC sample is cooled, would make the mirror more lossy, thereby decreasing the photon lifetime and increasing the linewidth. On the other hand, the reduction in phonons would increase the scattering time and decrease the linewidth. Overall, the effect of cooling is unknown, so we retain the room temperature empty cavity linewidth model.

At low temperatures, the ICP linewidth at the anticrossing point is, again, subaverage. As stated, to understand the subaverage linewidths requires the implementation of models beyond the STCO.

Figure 3.18.: Linewidth versus φ at low temperature. The modelled empty cavity linewidth γ_{MC} and the bare ISBT linewidth γ_{ISBT} are shown (dash-dotted and dashed lines, respectively). Data show subaverage behaviour around linewidths, but not such marked narrowing as seen at room temperature.



3.5.3. Previous work regarding ICP linewidths

Confirmation of Mary Matthews' work

These results confirm the observation of subaverage linewidths as first detected in the NRC sample by Mary Matthews [64], who performed laser spectroscopy with a high-intensity laser (see Ref. [94] for details of this laser). However, the linewidths there did not follow any discernible trend and so formulating a theory or fitting to a model was problematic. Furthermore, given the high pulse-pulse variation of the laser used, it was never certain that the narrow linewidths were not just an artefact of the laser itself. With these new data we are able to confirm and investigate in more detail the linewidth narrowing of the NRC sample.

Emmanuel Dupont's first paper regarding the NRC sample

The very first published results of the NRC sample did not detect any subaverage linewidths [44]. There, our collaborators used an FT/IR to measure spectra, with a $f/10$ numerical aperture giving an $1/e$ -width angular distribution of $\pm 6.3^\circ$. Therefore, the measurements were convolutions of a Gaussian distribution of angles. Later on, the same group managed to improve the spectral resolution of their FT/IR and reported subaverage linewidths of the UPB in a distinct, though similar ICP sample [55]. This demonstrates that using a light source with smaller angular distribution, the QCL laser, has given us a truer account of the ICP linewidths.

Załuźny and Zietkowski modelling of the NRC sample

A recent theoretical paper from Załuźny and Zietkowski [65], analytically probed the dispersion and linewidths of the NRC sample studied here ('Structure $\mathcal{B}$ ' in their paper corresponds to the NRC sample). In that work, the UPB linewidth follows that of the empty cavity, and the LPB linewidth follows that of the bare ISBT mode. This contradicts the STCO model, and looks similar to our measured linewidths at low temperature, seen in Fig. 3.18. But, the model did not predict linewidths to ever be narrower than the bare ISBT, as seen for our data at room temperature and 77 K. This suggests that their model, whilst thorough, did not take into account a particular aspect of the ICP, possibly the fact that it is a quasi-particle, and not just a light mode in a waveguide.

3.5.4. Shortcomings of STCO linewidth model

Fig. 3.19 shows the fitting of the data with the STCO model. This demonstrates in a more formal way the *subaverage* nature of the ICP linewidths, and the need for a development beyond the STCO model. The parameters of the STCO model were as derived previously; ISBT linewidths, γ_{ISBT} , were as per Fig. 3.15, and microcavity linewidths, γ_{MC} , were those of the TMF model, as shown by dash-dotted lines in Fig. 3.17 and Fig. 3.18.

The room temperature STCO model overestimates the magnitude of the linewidths at the anticrossing point, therefore confirming that these linewidths are subaverage. Elsewhere, the data is followed by the model quite closely, although we see that the linewidths are underestimated at larger angles. At room temperature, the model ICP linewidths simply trace the paths of the bare ISBT and empty cavity linewidths, though ‘exchanging labels’ when crossing.

At low temperature, we can see that the data, particularly the LPB, follows the model, but again, at the anticrossing point, the ICP linewidths are lower. This indicates that there is a need for a development of the STCO model, and a corresponding physical explanation for these subaverage linewidth observations.

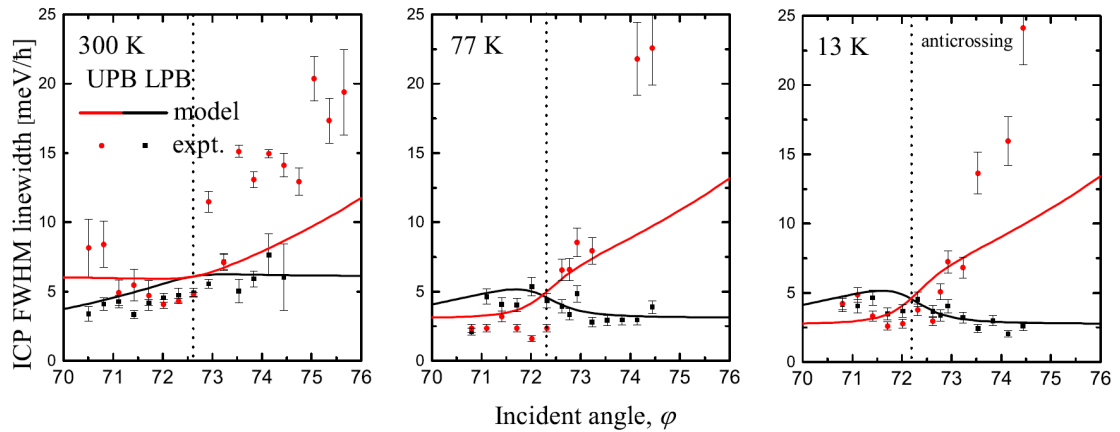


Figure 3.19.: Linewidth data with STCO model, showing fair agreement, but not predicting the subaverage linewidths at the anticrossing point (dotted line). Parameters in the model for the ISBT linewidth and cavity linewidth are as shown in Fig. 3.15 to Fig. 3.18.

3.5.5. Two models to consider for the explanation of subaverage linewidths

When subaverage linewidths were seen, in the mid 1990s, in the exciton cavity polariton system (see sec. 2.4.4) the cause was assigned to the inhomogeneity of the sample. However, it cannot be said, to any degree of certainty, that the NRC sample is inhomogeneously broadened. The absorption peak, for example, is very closely fit with a Lorentzian curve (notwithstanding the asymmetry, Fig. 3.13), and previous work by the group heavily suggested that the ISBT of well-grown, modern-day MQW samples is not typically inhomogeneously broadened (see sec. 2.2.2).

Nevertheless, we now consider two models which have two independent linewidth-narrowing explanations. Model I assumes the NRC sample is only homogeneously broadened, whereas Model II considers the case where the NRC sample is inhomogeneously broadened. The results and implications of these two Models are assessed and a conclusion is drawn as to which Model is the more likely to explain the data.

3.6. Model I - homogeneous broadening of ISBT linewidth only

We first consider the scenario where there is no inhomogeneous broadening of the ISBTs. The broadening mechanisms are those introduced in sec. 2.2, which affect the ISBT and the cavity linewidths. We must suppose that the effect of one or several broadening mechanisms is reduced at and around the resonance point. Around the resonance point the ICP is in its most mixed state, i.e. it is half light and half matter, in accordance with the Hopfield coefficients.

The theoretical background for this model borrows from a linewidth-narrowing theory known as *motional narrowing*, formulated by Whittaker *et al.* [95, 96]. This theory was originally applied to explain linewidth-narrowing of exciton cavity polaritons with inhomogeneous broadening. However, part of the theory can be applied to the homogeneously-broadened ICP system, and results in a new and *distinct* explanation of linewidth-narrowing.

3.6.1. Expression of ICP mass

To understand why the linewidth of the ICP is narrowed we must consider the broadening effects relevant to the constituent ISBT, in proximity to the resonance position. The broadening from phonons is mainly dependent on temperature of the sample (phonon scattering), and the energy of the ISBT (phonon emission). On the other hand, the IRS is dependent on a number of factors pertinent to the ISBT electron being scattered, in particular, its mass. The ICP quasi-particle has a reduced effective mass compared to the ISBT electron, which can be estimated from the masses of the ISBT electron and the photon. At equal parts light and matter (at the anticrossing point) [96]

$$\frac{1}{m_{ICP}} = \frac{1}{2} \left(\frac{1}{m^*} + \frac{1}{m_{photon}} \right), \quad (3.3)$$

where m^* is the ISBT electron effective mass, and the photon effective mass is defined as

$$\frac{1}{m_{photon}} = \frac{1}{\hbar^2} \frac{d^2(\hbar\bar{\omega}_1)}{dk^2}. \quad (3.4)$$

From equation 2.38, the photon energy in a microcavity is approximately quadratic which allows the effective photon mass in the microcavity to be calculable [96]:

$$m_{photon} \approx \frac{\hbar\pi\sqrt{\varepsilon_w}}{cL_{MC}}. \quad (3.5)$$

The photon does not have an actual mass, in the same way that in a semiconductor, the electron mass does not actually change - the photon can simply be described as if it had a mass within the waveguide. This gives $m_{photon} \approx 10^{-5}m_0$, where m_0 is electron mass. Since this is much smaller than the effective mass of the electron ($m^* = 0.063m_0$), the ICP mass will be approximately that of the photon, in accordance with equation 3.3: $m_{ICP} \approx m_{photon}$.

3.6.2. Small effective ISBT mass implies that ICP is unaffected by IRS

3.6.2.1. Small effective mass ICP does not scatter from interface roughness

If we inspect the equation for IRS (equation 2.29), we see that IRS is proportional to the mass of the scattered particle, the square of the mean characteristic height and the square of the correlation length of the roughness, $\gamma_{IRS}^{(intra)} \propto m_{scat}^* \Delta^2 \Lambda^2$. When the electron mixes with the photon and becomes the ICP quasi-particle, the mass of the scattered particle is reduced by at least 4 orders of magnitude ($m_{scat}^* = m_{ICP}$), thus we can expect the IRS broadening to be reduced by the same factor - this would virtually eliminate the IRS: $\gamma_{IRS}|_{anticrossing} \rightarrow 0$.

3.6.2.2. Increased spatial extent of ICP averages over IRS

Furthermore, this smaller effective mass decreases the localisation of the quasi-particle and the ICP's wavefunction is therefore spread out further than a pure ISBT³. The ISBT electron already has a fairly well-spread wave-like wavefunction of $\Lambda_{ISBT} \sim 10$ nm, but the ICP will have a spatial extent spreading even further according to [96]

$$\Lambda_{ICP} \approx \frac{2\pi\hbar}{\sqrt{2m_{ICP}E_{ICP}}} \quad (3.6)$$

which gives $\Lambda_{ICP} \approx 40$ nm. The increased length scale of the ICP, Λ_{ICP} , progressively averages over a larger area and reduces the magnitude of roughness height, Δ , in equation 2.29.⁴

3.6.3. STCO model with variable ISBT linewidth

The end result is that, at the anticrossing point, the IRS broadening is virtually eliminated, leaving just phonon broadening (and other, minor, effects - Tab. 2.2)

³This effect in the exciton cavity polariton system can aid Bose-Einstein condensation.

⁴This is closer to the motional narrowing theory of Whittaker *et al.* [96] in which the increased spatial extent of a polariton was able to average over the disorder in a microcavity exciton cavity polariton system.

affecting the ISBT: $\gamma_{IRS}|_{\text{anticrossing}} \rightarrow 0$, $\gamma_{ISBT}|_{\text{anticrossing}} \approx \gamma_{\text{phonon}}$.

We can thus work this into the STCO model, using the figures from the bare ISBT measurement (sec. 3.3.4), e.g. $\gamma_{\text{phonon}} \approx 4.6 \text{ meV}/\hbar$ and $\gamma_{IRS} \approx 1.6 \text{ meV}/\hbar$, at room temperature. The total ISBT linewidth thus has a variation with φ , where, away from the anticrossing point, it is $\gamma_{ISBT} \approx 6.2 \text{ meV}$, and at the anticrossing is $\gamma_{ISBT} \approx 4.6 \text{ meV}$. The form of the bare ISBT linewidth can be phenomenologically estimated as

$$\gamma_{ISBT}(\varphi) = \gamma_{\text{phonon}} + (\mathcal{C}^2 - \mathcal{X}^2)^2 \gamma_{IRS}, \quad (3.7)$$

where the term in brackets containing Hopfield coefficients $\mathcal{C}$ and $\mathcal{X}$ goes to zero at the anticrossing point.

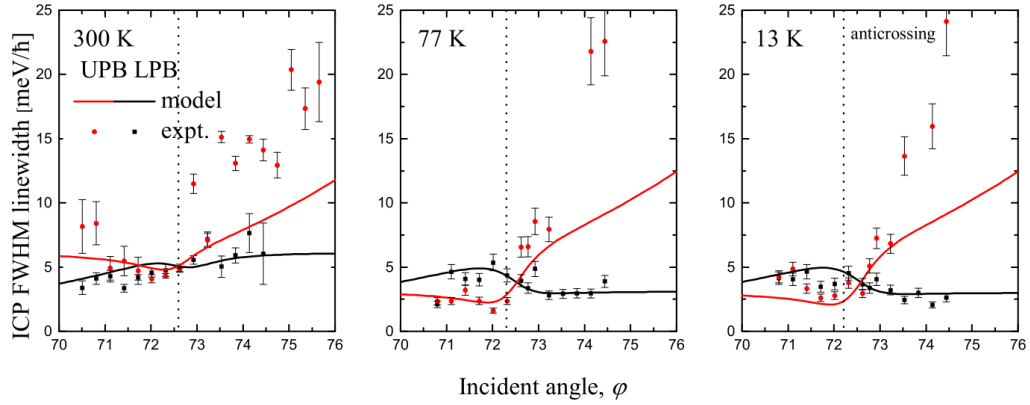


Figure 3.20.: Fitting of the data with Model I, described in sec. 3.6, with a variable γ_{ISBT} in which the IRS broadening is eliminated at the anticrossing point. The model shows good agreement with the data.

Substituting this into equation 2.69, and plotting alongside the data, gives the fits shown in Fig. 3.20, where there is now good agreement between model and data. Crucially, the data is now no longer consistently narrower than the model predicts: the model fits the subaverage linewidths. Parameters used in the model for all temperatures are given in Tab. 3.2.

Thus we have seen that an explanation of the subaverage linewidths could be due to the small mass of the ICP, and the effect this has to eliminate IRS broadening from the ISBT linewidth.

This concludes the interpretation of the linewidth data based on the ISBT being homogeneously broadened only.

Table 3.2.: Parameters used to model the variable γ_{ISBT} in Fig. 3.20 for Model I.

T [K]	γ_{phonon} [meV/ $\hbar$]	γ_{IRS} [meV/ $\hbar$]
300	4.6	1.6
77	1.8	1.6
13	1	1.6

3.7. Model II - inhomogeneous broadening

Now we turn our attention to the hypothetical situation where the MQW growth is inhomogeneous, and the ISBT linewidth is thus inhomogeneously broadened.

3.7.1. ISBT linewidth described by the Voigt profile

As seen in sec. 2.2.2, the presence of inhomogeneous broadening naturally leads to cavity polariton linewidth narrowing, if the vacuum Rabi splitting of the modes is larger than the bare ISBT linewidth. We make the assumption that the ISBT linewidth does have inhomogeneous broadening.

Inhomogeneous broadening gives the ISBT absorption peak a Voigt profile rather than a Lorentz profile. This is more challenging to include in analytical equations for the ICP linewidth, so we therefore employ the TMF model first introduced in sec. 3.4. To include the Voigt profile, the dielectric response of the MQW in the growth direction, via the effective medium approximation (sec. 2.4.1.1), is given by equation 2.57.

The model requires a total ISBT linewidth (equation 2.33), repeated here for convenience, of

$$\delta_{ISBT} = 0.5346 \times \gamma_{ISBT} + \sqrt{0.2166 \times \gamma_{ISBT}^2 + \xi_{ISBT}^2} \quad (3.8)$$

where γ_{ISBT} and ξ_{ISBT} are the Lorentzian and Gaussian FWHM linewidths, respectively. The Gaussian FWHM linewidth is related to the Gaussian parameter σ via: $\xi_{ISBT} = 2\sigma\sqrt{2\ln 2}$.

Starting from the fact that we know from separate measurements the total ISBT linewidth, $\delta_{ISBT} = 6.2 \text{ meV}/\hbar$, we can then vary the Lorentzian and Gaussian linewidths in the model until the model ICP linewidths fit the ICP data. This will then characterise the extent of the homogeneous and inhomogeneous broadening.

We found at room temperature that the best fit to the linewidth data was given by $\xi_{ISBT} = 3.1 \pm 0.3 \text{ meV}/\hbar$ and $\gamma_{ISBT} = 4.6 \pm 0.3 \text{ meV}/\hbar$. The dispersion relation resulting from this model is shown, in Fig. 3.21, as a colourmap, and is compared to the room temperature dispersion of the data first presented in Fig. 3.9. The data and model dispersion relations show good agreement.

Figure 3.21.: Room temperature colourmap disperions of Model II and experimental data showing good agreement between the two. The Model is, as detailed in sec. 3.7, a TMF model with the MQW represented by the effective medium approximation.

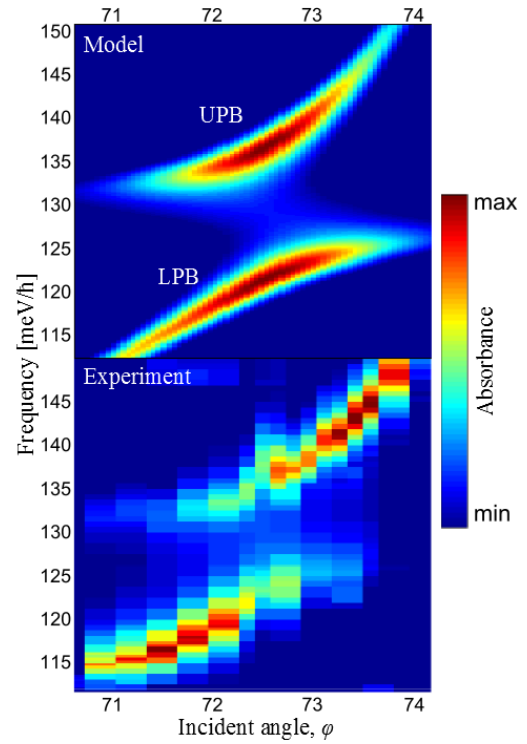
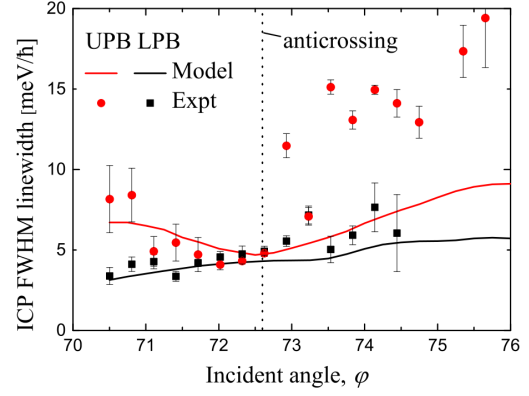


Fig. 3.22 shows that there is good agreement between the linewidth data and the model. As with Model I, there is the discrepancy between UPB data and model at angles above the anticrossing angle, possibly due to the spectral resolution of the laser, but otherwise the data and model are quite harmonious.

As stated, this narrowing effect at the anticrossing point has happened purely by virtue of supposing the ISBT to be inhomogeneously broadened. Whether the ISBT is inhomogeneously broadened or not is open to scrutiny.

Figure 3.22.: Data and modelled linewidths of ICPs with inhomogeneous broadening of bare ISBT (Model II). Bare ISBT linewidth is described by a Voigt profile, with the inhomogeneous broadening of $\xi_{ISBT} = 3.1 \pm 0.3 \text{ meV}/\hbar$ and homogeneous broadening of $\gamma_{ISBT} = 4.6 \pm 0.3 \text{ meV}/\hbar$. The data follows the linewidths well except at higher angles for the UPB.



3.7.2. Low temperature ICP linewidth narrowing with Voigt profile ISBT

3.7.2.1. Calculating inhomogeneous broadening from low temperature data

The TMF model was unable to model the low temperature behaviour of the ISBT. We believe that at low temperatures the bound to quasi-bound nature of the ISBT becomes important; the TMF model was not equipped to calculate the resulting transition energy.

It is, however, possible to analyse the low temperature data specifically at the anticrossing point. At the anticrossing point, the Gaussian contribution is removed from the ISBT linewidth, $\xi_{ISBT} \rightarrow 0$, and we are left with purely a Lorentzian ISBT mixing with a Lorentzian cavity mode, since now $\delta_{ISBT}|_{anticrossing} = \gamma_{ISBT}$. As such, the ICP linewidth becomes the arithmetic average

$$\gamma_{ICP}|_{anticrossing} = \frac{1}{2}(\gamma_{ISBT} + \gamma_{MC}|_{anticrossing}), \quad (3.9)$$

implying that, at the anticrossing point, the system follows the STCO model.

3.7.2.2. The issue with inhomogeneous broadening at low temperature for NRC sample

Unrealistic behaviour of inhomogeneous broadening

From the 13 K data, the ICP linewidths are approximately $\gamma_{ICP} \approx 3 - 4 \text{ meV}/\hbar$. Given that, according to the empty microcavity model, $\gamma_{MC}|_{\text{anticrossing}} \approx 6 \text{ meV}/\hbar$, we therefore can calculate the homogeneous ISBT broadening using equation 3.9, giving $\gamma_{ISBT} \approx 2 \text{ meV}/\hbar$.

Then, given that the full ISBT linewidth, $\delta_{ISBT}(13 \text{ K}) \approx 2.6 \text{ meV}/\hbar$, we can substitute values back into equation 3.8 to ascertain the inhomogeneous broadening at 13 K. Doing so gives an inhomogeneous contribution of $\xi_{ISBT}(13 \text{ K}) = 1.9 \pm 0.7 \text{ meV}/\hbar$.

This is problematic, because this value of inhomogeneous broadening differs substantially from the previously-calculated room temperature value ($\xi_{ISBT}(300 \text{ K}) = 3.1 \pm 0.3 \text{ meV}/\hbar$). Inhomogeneous broadening should *not* be temperature-dependent, since it originates from physical variation of well width which is temperature independent.

Conclusion that Model II does not explain linewidth narrowing in the NRC sample

We can therefore conclude that the linewidth-narrowing is *not* explicable via inhomogeneous ISBT broadening. It would appear that the NRC sample has minimal inhomogeneous broadening, or none whatsoever, which is reflected in the fact that the bare ISBT absorbance peaks of Fig. 3.13 look almost entirely Lorentzian.

This therefore suggests that Model II does *not* apply to our NRC sample. The observed linewidth narrowing in the NRC sample is explained by Model I: the light effective mass of the ICP being unaffected by IRS.

Conclusion to Part I

The spectra of ICPs have been studied at room and cryogenic temperatures. It was found that the linewidths of the ICPs are, near the anticrossing point, subaverage. That is, they do not follow a simple two coupled oscillator model, nor do they follow more detailed analytical models from other research groups. Two possible causes were considered for the narrowing of the linewidths.

The first cause (described by Model I) was that the linewidth of the ISBT reduces when the ISBT is in the quasi-particle state (the ICP) because the lighter effective mass of the ICP scatters less from the interface roughness, and additionally has a larger spatial extent which averages over a larger area of roughness, when compared to the ISBT electron. This reduction in the IRS broadening reduces the linewidth of the ICP.

The second explanation of narrowing investigated (Model II) was one previously established in exciton cavity polariton systems in which the inhomogeneity of the ISBT distribution is eliminated under strong vacuum Rabi splitting. This behaviour appeared to be supported by a transfer matrix formalism-based model which incorporated the inhomogeneity, accounting for the narrowing at room temperature. However, low temperature measurements implied that the inhomogeneous broadening *reduced* with sample cooling, which was not the expected behaviour, therefore suggesting that this explanation did *not* apply to our NRC sample.

Hence, the first explanation (Model I) was deemed the dominant mechanism for the narrowing of ICP linewidths. This conclusion was supported by the fact that the ISBT linewidth follows the homogeneous, Lorentzian fitting very strongly.

The work demonstrated that, by using laser sources to perform spectroscopy of highly dispersive systems, it is possible to attain a sharper spectral resolution when compared to wide-angled sources typical of fourier transform infrared spectrometers, and thereby study the linewidths more precisely. Future development of this work would involve a more rigorous theoretical treatment of the ICP wavefunction and

mass to properly track the behaviour of the IRS scattering as the ISBT mixes with the cavity photon. Experimentally, this linewidth narrowing phenomenon should be investigated in other systems; those with different geometry, and those systems displaying ultrastrong coupling. This current work adds to the study of the quantum effects of the ICP, and the insight gained could be used where the understanding of ICP coherent lifetimes is important.

Part II.

A study on Q switching the 3 micron erbium laser

Summary of Part II

Investigations into the $\lambda \simeq 3 \mu\text{m}$ $\text{Er}^{3+}, \text{Cr}^{3+}:\text{YSGG}$, flashlamp-pumped, Q switched laser are presented.

The thermal lens in the laser rod has been characterised, and the laser cavity designed to support fundamental mode (TEM_{00}) lasing of high energy, free-running output pulses ($\sim 76 \text{ mJ}$ per pulse).

Q switching of the laser was then investigated in order to produce short ($< 400 \text{ ns}$) laser pulses, using two ‘slow’ Q switching methods: a rotating mirror Q switch, and a polygon chopper Q switch. The rotating mirror Q switch was found to produce pulses of $\sim 10 \text{ mJ}$, with an optimum mirror rotation rate of 300 rotations per second. Faster rotation rates gave lower pulse energies because the low-loss period of the Q switch at these rates became shorter than the flashlamp pulse rise time.

The polygon chopper Q switch, which used a rotating polygon as an optical chopper, produced single pulses of energy $\sim 4 \text{ mJ}$. From this Q switch, it was deduced that the longest Q switching time possible for $\text{Er}^{3+}, \text{Cr}^{3+}:\text{YSGG}$ for any Q switching method was $\sim 30 \mu\text{s}$ for single pulses, and $\sim 80 \mu\text{s}$ for any Q switching whatsoever.

Finally, an airtight container for the laser was constructed, and a gaseous nitrogen purge applied to the polygon chopper Q switched laser, demonstrating increased laser pulse energy of 29%. This was attributed to the removal of water vapour from the atmosphere, a strong absorber at $\lambda \approx 3 \mu\text{m}$.

4. Introduction and theory to Part II

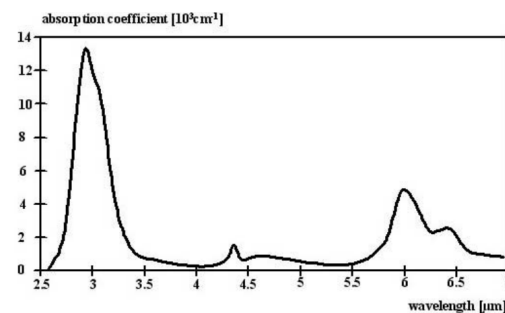
This chapter contains an overview of the uses of the 3 μm erbium laser, theoretical details of the lasing operation, and details of the current state of the art.

4.1. Uses of 3 μm radiation

4.1.1. Direct uses of 3 μm radiation

The 3 μm erbium laser has gained much attention in recent years mainly due to the presence of strong water absorption at this wavelength. Because water makes up a large proportion of biological matter, pulsed 3 μm radiation is used in a variety of settings to ablate human tissue by the rapid heating of the human cell's constituent water. Erbium lasers are already widely used in cosmetic skin treatment, ablating damaged, superficial tissue to allow healing [97]. Erbium lasers find a similar use in dentistry [98], and also have potential in ophthalmic surgery in reshaping the cornea, and removal of cataracts, amongst other uses [99, 100]. See Fig. 4.1.

Figure 4.1.: Absorption coefficient spectrum for water in corneal tissue. Other tissue types with high water content will have a similar spectrum. Reprinted figure with kind permission from [101]. Copyright (2003) Springer Science+Business media.



Erbium lasers also find surgical uses inside the body, where fibre-delivered light is able to make incisions during keyhole surgery, and also can be used to vapourise

tumours and other unwanted matter [102]. Incisions made by erbium lasers cauterise better than the more widespread midinfrared CO₂ medical laser, and produce less collateral damage. Collateral damage is minimised further with shorter, singular pulses, and with precise beam delivery. Methods of shortening beam pulses (specifically, Q switching) and improving beam quality (specifically, thermal lens compensation) are investigated in this thesis for the 3 μm erbium laser.

4.1.2. 3 μm radiation for generation of midinfrared light

Whilst surgical applications have provided the main incentives for the commercial development of high-energy erbium lasers, intense, 3 μm laser light finds use elsewhere.

Given sufficient intensities, 3 μm radiation can be down-converted to light of wavelengths spanning the midinfrared, from $\lambda = 3$ to beyond 20 μm . The common method for down conversion, the optical parametric oscillator (OPO), has been implemented utilising a variety of crystals including ZnGeP₂, CdGeAs₂, CdSe, AgGaS₂ and AgGaSe₂. See Ref. [103] for a summary on suitable materials and wavelength ranges covered. OPOs operating in the midinfrared are important due to the current lack of laser sources in this wavelength range, especially sources of high intensity.

4.1.2.1. Spectroscopy of gases and organic species

A continually-tunable midinfrared source, such as the above-mentioned OPO, finds use in spectroscopy, since almost all polyatomic molecules have vibrational modes in the midinfrared [104]. Fig. 4.2 shows the absorption ranges for the functional groups of a number of common molecules, and the positions of absorption peaks for some environmentally important molecules are shown below that [105]. The presence and quantity of a given air-borne gas can be detected with midinfrared sources, thanks to the individual spectral *fingerprints* of a particular molecule. In the same way, midinfrared sources can be used in biological spectroscopy, including the analysis of DNA and proteins [106, 107].

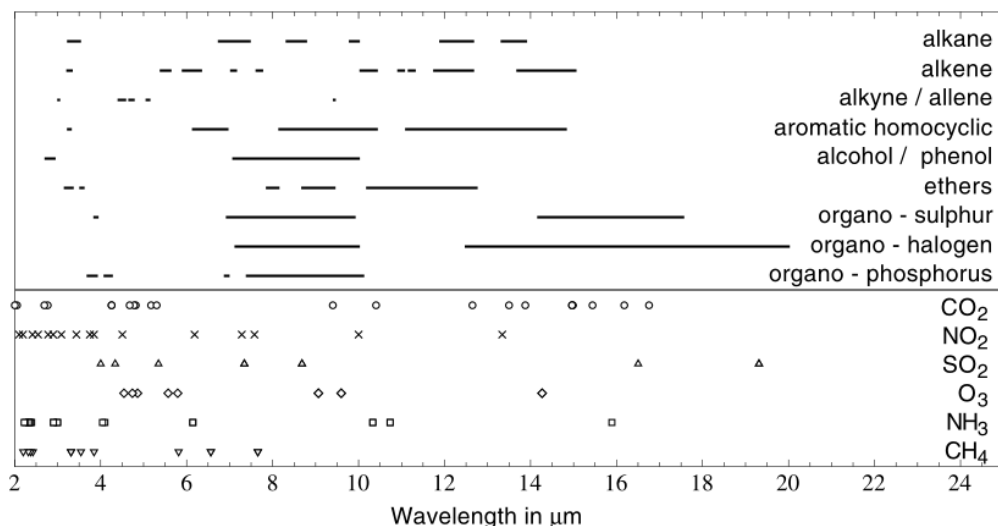


Figure 4.2.: Absorption ranges of important functional groups of molecules (top) and environmentally-significant molecules (bottom). Reprinted figure with kind permission from [105]. Copyright (2003) Springer Science+Business media.

4.1.2.2. Spectroscopy of multi-quantum wells

Midinfrared sources can also be used in semiconductor spectroscopy, in particular, for multi-quantum well samples. As seen in Part I of this thesis, the intersubband transitions in quantum wells are often active in the midinfrared, and so a laser source capable of probing these transitions is useful, particularly if high intensity and high angular resolution are required.

4.2. Theory of the 3 μm erbium laser transition

4.2.1. Er^{3+} optical transitions

The trivalent erbium atom, Er^{3+} , has several transitions capable of lasing in the visible and infrared. Like certain other rare-earth ions¹, these transitions in erbium occur in the $4f^{11}$ subshell, which is split by the noncentrosymmetric electron-electron interaction, the spin orbit interaction, and the Stark interaction (the crystal field effect) [109]. The partial energy level diagram is given in Fig. 4.3, without the Stark splitting.

¹For example, various lanthanides e.g. Ho, Yb and Tm have successfully had infrared lasers developed based on similar energy levels [108].

We see in Fig. 4.3 some of the possible optical transitions in Er^{3+} which lase from the visible to midinfrared regions. The transition which actually lases depends largely on the host material of the Er^{3+} ions, its properties such as transparency range, the interaction between the host phonon population and the transition energies, and the concentration of Er^{3+} ions. Furthermore, to encourage certain lasing transitions, the host crystal can be co-doped with different elements to enhance the filling of specific energy levels.

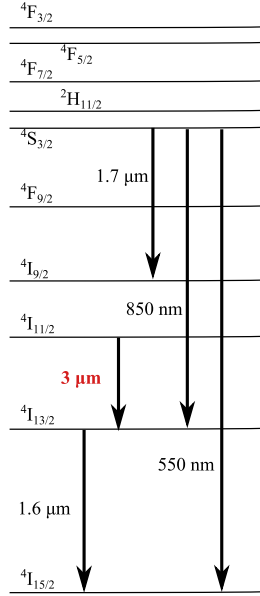


Figure 4.3.: Energy-level scheme of trivalent erbium resulting from the splitting of the $4f^{11}$ configuration due to electron-electron interaction and spin-orbit interaction. Further Stark splitting due to crystal field of the host is not shown. A selection of notable optical transitions in the visible and infrared are shown, with the pertinent $3\ \mu\text{m}$ transition being highlighted in red.

One transition of particular note is the $\lambda = 1.5\text{-}1.6\ \mu\text{m}$ transition, between $4I_{13/2} \rightarrow 4I_{15/2}$ which is accessible in Er:glass fibre lasers [108]. This wavelength has widespread use in telecoms since common optical fibres have minimal loss around $1.5\ \mu\text{m}$, and fibre amplifiers can easily be implemented as signal boosters. Other transitions shown in Fig. 4.3 harnessed in lasers emit at wavelengths of $550\ \text{nm}$ [110], $850\ \text{nm}$ [111], and $1.7\ \mu\text{m}$ [112]. These laser transitions are not, however, studied in this thesis.

4.2.2. $3\ \mu\text{m}$ lasing dynamics in erbium

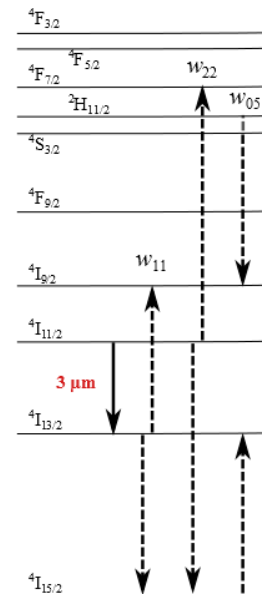
This $\sim 3\ \mu\text{m}$ lasing transition, pertinent to our system, is the transition occurring between $4I_{11/2} \rightarrow 4I_{13/2}$. A notable property of this laser transition is that the upper laser level has a shorter spontaneous emission lifetime than the lower laser level ($1.3\ \text{ms}$ and $3.4\ \text{ms}$, respectively for Er,Cr:YSGG - see sec. 4.3.1), which causes the

self-termination of lasing action. Self-termination prevents cw lasing, and emission of continuous long-duration pulses [113].

More notable in Er^{3+} , however, is the presence of certain energy up-conversion processes which counteract the imbalance of the laser level spontaneous emission lifetimes, which must be considered when formulating rate equations describing level populations. Indeed, these energy up-conversion processes in fact permit cw emission for the 3 μm laser transition, given the appropriate laser pumping rate [113, 114].

The three major energy up-conversion processes are inter-ionic interactions which cooperatively act to rapidly deplete the lower laser level, and are shown in Fig. 4.4. Two are energy-transfer up-conversion (ETU) processes, mediated by dipole-dipole interactions; the third interaction is a thermally-coupled cross-relaxation (CR).

Figure 4.4.: Energy up-conversion processes affecting the laser levels of the 3 μm transition in Er^{3+} . The ratio between w_{11} and w_{22} , energy-transfer up-conversion processes, should be maximised for most effective attainment of population inversion. The cross-relaxation, w_{05} , encourages the w_{11} process.



The first ETU, denoted w_{11} , depopulates the lower laser level, with a non-radiative transition from $^4\text{I}_{13/2} \rightarrow ^4\text{I}_{15/2}$ in one ion, exciting an electron in a neighbouring ion from $^4\text{I}_{13/2} \rightarrow ^4\text{I}_{9/2}$, which then relaxes down to populate the upper laser level. The second ETU, denoted w_{22} , removes electrons from the upper laser level, with one electron going to the ground state, and the other to the $^4\text{F}_{9/2}$ level. To encourage population inversion, w_{11} should be maximised with respect to w_{22} , the ratio being dependent on the host and further controlled by Er^{3+} ion concentrations - see Tab. 4.1 [109]. The CR process, w_{05} , consists of the relaxation from $^2\text{H}_{11/2}$ or $^4\text{S}_{3/2} \rightarrow ^4\text{I}_{9/2}$, thermally exciting an electron from the ground state $^4\text{I}_{15/2}$ to $^4\text{I}_{13/2}$. The

excitation to this level encourages the w_{11} process, and thereby increases population inversion [115].

4.3. Design considerations for 3 μm erbium laser

In the designing of a 3 μm erbium laser, there are a number of considerations depending on the pumping mechanism, the efficiency required and the exact wavelength to be emitted, etc. We outline them here, making reference to our own system where appropriate.

4.3.1. Choice of host material

4.3.1.1. Common erbium hosts

The host material for the Er^{3+} ions in this thesis is YSGG (yttrium scandium gallium garnet, $\text{Y}_3\text{Sc}_2\text{Ga}_3\text{O}_{12}$), co-doped with trivalent chromium, Cr^{3+} (see sec. 4.3.2.2), giving Er,Cr:YSGG. Er^{3+} is typically doped between 30-50 at.%, and Cr^{3+} at <2 at.%, the dopants replacing ions of Y and ScGa, respectively [116, 117].

This host was chosen over the more commonplace Er:YAG (yttrium aluminium garnet, $\text{Y}_3\text{Al}_5\text{O}_9$), which is the major material for commercial medical-grade erbium lasers [101], thanks to possible superior efficiency and energy-storage prospects of the YSGG [113, 115, 118]. Another bulk host which has achieved some success is Er:YLF (LiYF_4) [119, 120].

4.3.1.2. Other host materials

Other possible hosts of Er^{3+} which have been demonstrated, but not developed further, include other garnets: GGG [113], GSGG [121], GSAG [122], $\text{Er}_x\text{Y}_{1-x}\text{AG}$ [123], $\text{Er}_x\text{Lu}_{1-x}\text{AG}$ [124]; as well as various fluorides: CaF_2 [125], SrF_2 [126], BaY_2F_8 [127], and BaLuF_8 [128].

There have been recent breakthroughs in fibre lasers, in particular, the fibre known as ZBLAN [112], in terms of efficiency and output power - see sec. 4.4.1.

Table 4.1.: Comparison of spectroscopic and thermal properties of erbium doped in YSGG and YAG. (a)[116], (b)[113], (c)[118], (d)[117]

	Er(30%):YSGG	Er(50%):YAG
Primary laser wavelength [μm]	2.794 (a)	2.937 (b)
Upper laser level lifetime [ms] (b)	1.3	0.12
Lower laser level lifetime [ms] (b)	3.4	7.25
Stimulated emission cross-section [10^{-21}cm^2] (c)	2.8	2.8
w_{11} [$10^{-22}\text{m}^3\text{s}^{-1}$] (c)	2.3	0.25
w_{22} [$10^{-22}\text{m}^3\text{s}^{-1}$] (c)	7.3	0.3
Thermal expansion coefficient [10^{-6}K^{-1}] (d)	8.1	9.0
dn/dT [10^{-6}K^{-1}] (d)	12.3	7
Thermal conductivity [$\text{W}\cdot\text{mK}^{-1}$] (d)	8	7.6

4.3.1.3. Effect of host on lasing transition

Stark sublevels

The host crystal field and phonon population interacts with the erbium ions and affects the Stark sublevels involved in the lasing; this interaction is responsible for most of the variation across hosts in the lasing wavelength from, for example, 2.7 μm in ZBLAN, to 2.94 μm in YAG.

In YSGG, for example, the possible transitions between the Stark sublevels range in wavelength from 2.608 to 2.923 μm . However, in YSGG, the Stark sublevels of the upper and lower lasing levels are usually 2 and 4, respectively, with lasing at $\lambda = 2.794 \mu\text{m}$ [116]. See Fig. 4.5.

The lasing transition that does occur is influenced by the macroscopic Boltzmann distribution of the electrons within the Stark sublevel manifolds, with the upper laser level typically being a low Stark sublevel, and the low laser level a high Stark sublevel. The filling of specific lasing Stark sublevels must be accounted for in any rate equation formulation of this laser, and is dependent somewhat on the host material.

Spontaneous emission lifetimes

The choice of host material also affects the spontaneous emission lifetimes of the lasing levels, with some host materials affording the erbium ions superior energy storage and efficiency. Er:YSGG has an upper laser level lifetime some ten times

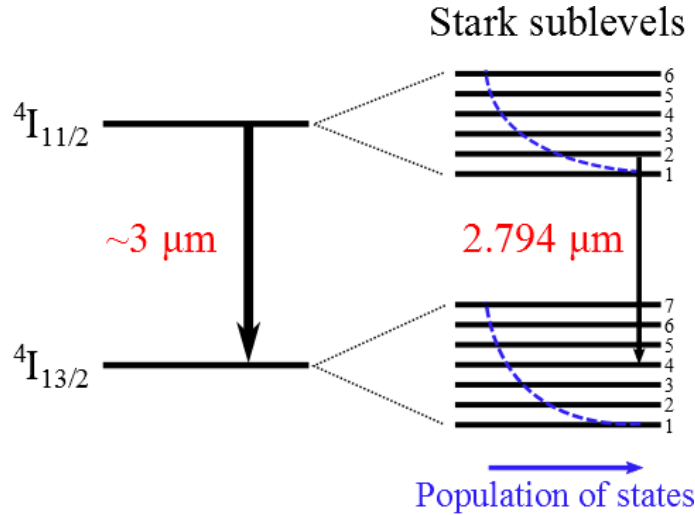


Figure 4.5.: The Stark sublevels of the Er,Cr:YSGG laser levels, showing the typical lasing transition of $\lambda = 2.794 \mu\text{m}$ between sublevels 2 and 4. Also shown is the macroscopic population of electron states in the sublevels distributed according to the Boltzmann distribution.

as long as that of Er:YAG (1.3 ms compared to 0.12 ms [113]), and an upper:lower level lifetime ratio of 0.38 compared to 0.017. See Tab.4.1. This superiority has been demonstrated theoretically in Ref. [118] and experimentally for free-running laser mode in Ref. [115].

Thermal properties

The choice of host material also impacts on the various thermal properties of the laser, for example, the thermal expansion coefficient, the thermal conductivity, and the temperature-dependent refractive index change. See Tab.4.1 for thermal properties of YSGG and YAG. This is of importance in the consideration of thermal lensing, and general heat management - see chapter 5.

4.3.2. Choice of pumping mechanism

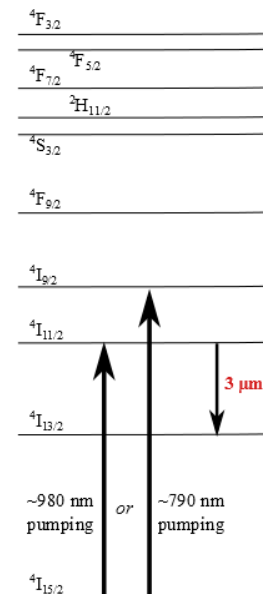
There are two major pumping mechanisms possible for erbium. Flashlamp pumping and diode pumping. (It is also possible to pump with Ti:Sapphire [121] or another suitable laser, though diode pumping is more convenient.)

4.3.2.1. Diode pumping

Direct level diode pumping has the advantage of having minimal quantum defect, which greatly increases efficiency and reduces excessive heating of the host, which in turn increases efficiency even further. For fibre lasers it is the preferred method of pumping, and for cw lasers the only sensible choice.

For the lowest quantum defect, diodes are chosen to pump the erbium at $\lambda \approx 980 \text{ nm}$ [115, 129], the absorption transition from the ground state directly to the upper laser level, $^4\text{I}_{11/2}$. This pumping scheme is a three-level laser system. Alternatively, the $^4\text{I}_{9/2}$ level can be populated with a laser operating at $\lambda \approx 790 \text{ nm}$, in a four-level laser scheme, to the detriment of the quantum defect [116, 129, 130]. See Fig. 4.6.

Figure 4.6.: Energy level diagram for 3 μm lasing in Er^{3+} showing direct-level pumping by laser diodes, or other sources, of the upper laser level, $^4\text{I}_{11/2}$, by $\lambda \approx 980 \text{ nm}$ radiation for a 3-level laser system, or of the $^4\text{I}_{9/2}$ level with $\lambda \approx 790 \text{ nm}$ radiation, for a 4-level laser.

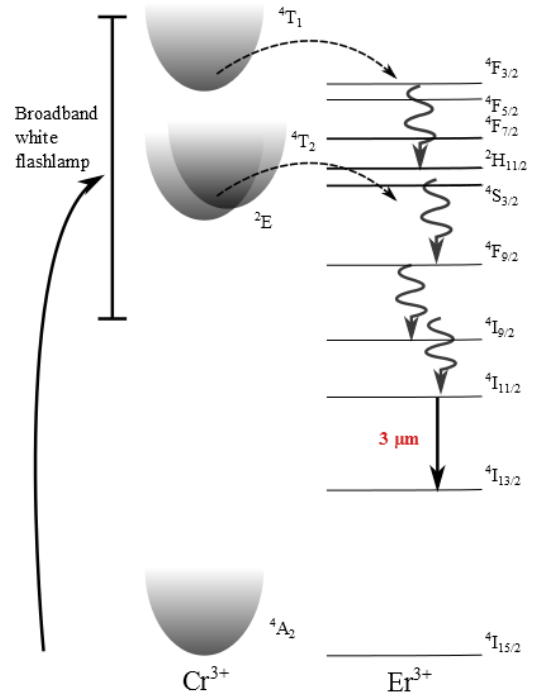


4.3.2.2. Flashlamp pumping

For pulsed operation it is possible to use a broad-spectrum white flashlamp (for example, of xenon gas). This excites the electron population into high energy levels from where they relax downward towards the upper laser level.

In the YSGG host it is possible to increase sensitivity to broadband flashlamp light with chromium doping. Cr^{3+} ions act as sensitizers possessing two broad absorption bands in the blue-green (centred at $\lambda \approx 450 \text{ nm}$) and red wavelength (centred at

Figure 4.7.: Energy level diagram demonstrating xenon flashlamp excitation of the Er^{3+} ion, via direct excitation, and also via the broadly absorbing levels of the Cr^{3+} sensitizer ions, which transfer excitation energy to the Er^{3+} ions via dipole-dipole interaction.



$\lambda \approx 640 \text{ nm}$) regions, a result of vibronic transitions to the 4T_1 and 4T_2 energy levels. These bands will absorb a large amount of broadband xenon flashlamp emission which may then be transferred to Er^{3+} via dipole-dipole nonradiative processes with $\sim 100\%$ efficiency. Cr^{3+} is rarely co-doped in $\text{Er}:\text{YAG}$ crystals due to the larger crystal field of YAG slowing the nonradiative transition. Furthermore, unlike YAG, large lattice garnets such as YSGG and GSGG are also able to accept large concentrations of Cr^{3+} [108, 116]. See Fig. 4.7.

The envelope of a flashlamp flash is not readily controllable, in terms of shape and duration. This is in contrast to diode pumping wherein the pump radiation amplitude can be shaped (allowing a top-hat profile, for example), and varied precisely in duration to find the optimum pumping time to optimise level filling, and to avoid excess heating, etc.

4.3.2.3. Influence of pumping mechanism on gain design

The shape of the gain crystal itself must be considered, but this is mostly influenced by the type of pumping. Fig. 4.8 shows two common methods of pumping.

Flashlamp-pumped rods must be designed to accept light from a multi-directional

pump source. Hence, a cylindrical rod is best, with the rod placed at one focal point of an elliptical reflective pump chamber, and the flashlamp at the other focal point. Flashlamp light enters the rod at all angles, uniformly illuminating the rod interior. Pumped in this way, cylindrical laser rods are well-suited for producing fundamental mode lasing, thanks to the inherent circular symmetry of the pumping.

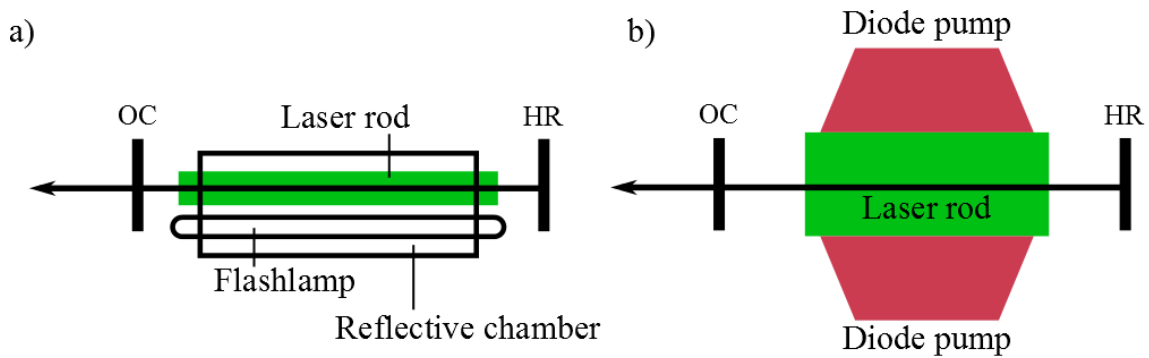


Figure 4.8.: The two main options for pumping the erbium laser: a) flashlamp pumping, where broadband white light from a flashlamp (usually xenon) is reflected within an elliptical reflective chamber into a cylindrical rod; and b) diode pumping, where monochromatic laser diode (usually around 970 nm wavelength) is focussed into the sides of a laser rod. Both schemes are shown within a laser resonator consisting of a high reflecting back mirror (HR) and an output coupler (OC). Variations of both schemes exist, e.g. Brewster cut rod faces, and internal reflections of the laser mode within the rod.

For diode pumping, the pump radiation is more directional. Diode-pumped laser rods are therefore often rectangular in cross-section such that the pump radiation can be effectively focussed through the side of the rod. Diode-pumped lasers often also employ total internal reflections within the laser cavity, to homogenise the laser mode (averaging through the regions of different temperature), tracing a zig-zag through the medium, or making a single reflection.

In both pumping cases, the ends of the rods can be plane-cut, often with appropriate anti-reflection coating; they can be Brewster-angled to minimise reflection for the p-polarisation; or they can be concave to counteract thermal lensing in the rod. In this thesis, the Er,Cr:YSGG laser rod was cylindrical, with Brewster-angled ends, and was flashlamp-pumped.

4.3.3. Cavity design for fundamental mode lasing

To design a cavity which lases in the fundamental mode (TEM_{00}), it is important to match the cross-sectional size of the fundamental mode with the gain region (population-inverted region) of the laser rod. This may be done with intracavity apertures, to effectively limit the size of the gain region. Alternatively, intracavity lenses may be used, or concave or convex cavity mirrors, to increase the size of the fundamental mode.

However, to design a cavity with a maximised fundamental mode cross-section, one must take into account the gain medium thermal lens. This is an issue inherent to solid state lasers, especially flashlamp-pumped ones; the high pumping power induces a temperature gradient in the gain medium, and therefore a gradient index lens is formed, alongside other effects (see sec. 5.2). Since this lens changes its value depending on the input pulse energy and pulse repetition rate, its behaviour in a given medium must be carefully characterised. Only then can reliable fundamental mode operation be achieved. This is the aim of chapter 5, with respect to the flashlamp-pumped Er,Cr:YSGG system.

4.3.4. Pulsewidth and intensity of $\lambda \simeq 3 \mu\text{m}$ erbium laser

In terms of decreasing the pulsewidth of the laser, and increasing its pulse intensity, common choices are Q switching, gain switching and modelocking [131]. Q switching is considered in this thesis.

Q switching is the technique whereby the quality factor, Q , of the laser cavity is held temporarily low during pumping. This allows population inversion to build up to a high value, at which point the Q is *switched* to a high value. This causes rapid depopulation of the upper laser level resulting in a short, intense pulse of light. There are many techniques for Q switching, categorised into fast and slow methods. These are expanded upon in sec. 6.1. In this thesis we investigate two methods of slow Q switching the erbium laser, one of which is a method not previously implemented in the erbium laser - an optical chopper Q switch.

4.3.5. Atmospheric considerations

A final factor affecting the laser's design is the question of intracavity, atmospheric loss to 3 μm radiation. The very factors that make midinfrared lasers useful (i.e. the presence of absorption bands in the midinfrared of countless molecules) also hinder their performance. An erbium laser, operating at $\lambda \simeq 3 \mu\text{m}$, will suffer from intracavity loss from water vapour absorption.

Previous research showed that removing the moisture from an Er:YAG cavity as much as doubled output pulse energy [132]. In this thesis we investigate the effect of purging the atmosphere of the surroundings of the Er,Cr:YSGG laser with pure nitrogen.

4.4. Erbium laser performance in the literature

4.4.1. Current state of the art

The highest energy Q switched erbium laser was reported by Yang *et al.* to be a xenon flashlamp-pumped Er:YAG laser with 226 mJ pulses of 62 ns pulsewidth, giving 3.6 MW peak power, running at 3 Hz, in multiple transverse modes [133]. Q switching was achieved electrooptically (see sec. 6.1.2.1). This laser also output ~ 1 J per pulse in free-running mode, giving a 3 W average power.

The best Q switching performance with fundamental mode operation mode was a flashlamp-pumped Er:YAG laser, with a rotating mirror Q switch (see sec. 6.1.2.2) giving 30 mJ pulses at 25 Hz, with pulsewidths of less than 300 ns - free-running pulses were of the order of 100 mJ [134].

In terms of cw operation, Er:YLF (LiYF_4) has the highest reported cw power for a bulk solid state laser, at 4 W, diode pumped at $\lambda = 980 \text{ nm}$ [120]. The highest power reported from YAG was quasi-CW at 3.2 W consisting 32 mJ pulses at 100 Hz repetition rate (962 nm pump wavelength) [135].

More recently, ZBLAN fibre-based, 3 μm erbium lasers have become more powerful [136], and in fact an Er:ZBLAN laser holds the current record for highest average power, with a 24 W cw laser, pumped at $\lambda = 975 \text{ nm}$ in a multimode fibre [137], and elsewhere, a 10 W fundamental mode laser [138]. Q switching in Er:ZBLAN also looks promising with reports of 100 μJ energy pulses with 90 ns pulsewidths, at

a 120 kHz repetition rate, in a multimode fibre. The efficiency of the ZBLAN host is due partly to the fact that in this host $w_{11} > w_{22}$, unlike in YSGG and YAG, greatly favouring energy recycling upconversion (see sec. 4.2.2) [109].

Reports of the diode pumping of Er^{3+} at $\lambda \approx 790$ nm, corresponding to pumping of the $^4\text{I}_{9/2}$ level, are less widespread; lower-power cw and pulsed operation, are reported in Refs. [119, 139].

4.4.2. YSGG as host performance

Lasing from Er,Cr:YSGG, the gain medium studied in this thesis, was first attained in 1986 [140], and has since been studied and developed as an alternative to YAG. The long upper laser level lifetime, and the superior upconversion ratio w_{11}/w_{22} can potentially give YSGG a lower threshold, higher slope efficiency, and better Q switch energy storage capacity compared to YAG in a comparable system [118].

The highest free-running pulse energies from the Er,Cr:YSGG laser are reported as 120 mJ in fundamental mode, or 580 mJ in multimode operation, from a flashlamp-pumped system [116].

The highest power free-running diode-pumped Er:YSGG laser² comes from our collaborators at Imperial College (Mike Damzen group) with ~ 55 mJ pulses at a repetition rate of ~ 36 Hz, pumped at $\lambda = 966$ nm, giving a 2 W output power [141]. See sec. 6.6.

The best reported Q switched performance from the YSGG host was from a flashlamp-pumped system giving 55 mJ pulses, with 43 ns pulsewidth, surpassing 1 MW pulse power, by Högele *et al.* Interestingly, it was a $\lambda = 2.70$ μm transition that was made to lase, rather than the more commonplace 2.79 μm transition [142]. Q switching was achieved with a piezoelectric-modulated prism pair. Q switching will be covered in more detail in sec. 6.1.2.

Reports of cw lasing with a YSGG host are less numerous than in other hosts [113] - Waarts *et al.* reported 900 mW cw action from a monolithic microchip laser [143]. This power was recently matched in an Er:YSGG/YSGG composite crystal, which benefitted from superior thermal management [144].

²Co-doping with Cr^{3+} is ineffectual when diode-pumping.

4.5. Motivation for this work

Most laser applications require fundamental mode operation (TEM_{00}) for optimal spatial properties of the beam, which gives enhanced focussing capability, and convenient intensity profile. Additionally, in pulsed operation, the pulsewidth is of importance to applications where higher intensity is required, for example to drive nonlinear optical processes, or ablation of biological tissue.

Our system consists of a flashlamp pumped Er,Cr:YSGG laser, with fundamental mode operation and Q switched pulses. Chapter 5 details the characterisation of the thermal lens and achieving fundamental mode operation. Chapter 6 introduces Q switching of the system, and the results of various Q switching methods, including one method never previously implemented in the erbium system. In that same chapter is also detailed the improvement of the laser output through nitrogen purging of the surrounding laser atmosphere.

5. Fundamental mode operation of the Er,Cr:YSGG laser

As is common in solid state lasers, Er,Cr:YSGG laser rods suffer from thermal effects which have focussing and depolarising effects on the cavity modes. The ‘thermal lens’ is strong enough to significantly affect the stability of the cavity and the modes resonating within. This chapter describes the theory of the thermal lens within the Er,Cr:YSGG rod and the experimental determination of its strength; the chapter then describes the design of the cavity taking into account this thermal lens and achieving fundamental mode emission in free-running mode.

5.1. The Er,Cr:YSGG laser rod

The active element in the laser is a rod of YSGG ($\text{Y}_3\text{Sc}_2\text{Ga}_3\text{O}_{12}$) doped with Er^{3+} and Cr^{3+} at concentrations of $4.7 \times 10^{21} \text{ cm}^{-3}$ and $2.0 \times 10^{20} \text{ cm}^{-3}$, respectively. This is an Er concentration of around 33 at.% relative to Y, and a chromium concentration of 2.4 at.% relative to ScGa [116, 117]. The gain rod is $\text{Ø}4 \text{ mm} \times 90 \text{ mm}$ and is Brewster-cut to select p-polarised light in the cavity. See Fig. 5.1.

The rod is flashlamp-pumped by a xenon flashlamp within a water-cooled ($\sim 17^\circ\text{C}$), diffusely-scattering pump chamber (of BaSO_4 interior), at repetition rates of up to 12 pulses per second, at energies per flashlamp flash up to 60 J.

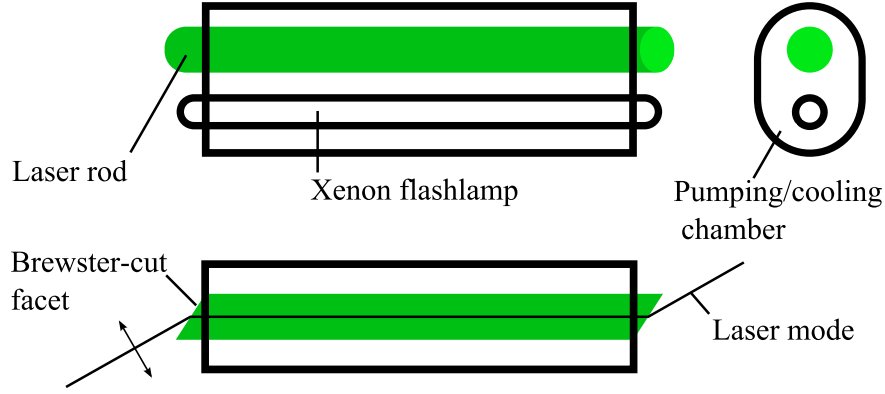


Figure 5.1.: Plan view of the Er,Cr:YSGG laser rod and Xe flashlamp within the pumping and cooling chamber, and the direction and polarisation of the laser mode with the laser rod's Brewster-cut facets.

5.2. Theory of thermal lenses in solid state laser gain rods

5.2.1. Intrinsic pump-to-laser efficiency

5.2.1.1. Quantum defect in Er,Cr:YSGG

The amount of input energy from the pump light that is converted into output laser light is fundamentally limited by the quantum defect of the laser system. For direct-level pumping by diode laser, the energy lost in non-radiative, heating transitions is minimised. Even so, for our system, direct pumping of the upper laser level, $^4I_{11/2}$ ($\lambda \simeq 980$ nm), has a quantum defect of $\sim 67\%$ of the pump photon energy. For direct pumping to a higher level, the quantum defect is higher; for example, for direct pumping of level $^4I_{9/2}$ ($\lambda \simeq 790$ nm), the quantum defect is $\sim 74\%$.

So for a flashlamp-pumped system, with a white, broadband spectrum, the range of quantum defects will be higher still. The amount of pump energy that does not go towards lasing, instead heating the laser rod, was estimated to be above 80%.

5.2.1.2. Effect of up-conversion processes on heating

The existence of energy up-conversion processes introduced in sec. 4.2.2 can complicate any straightforward calculation of pump energy transferred to heat based purely on the quantum defect. The effects of w_{11} and w_{05} would decrease the amount of

energy lost to phonons, whereas the effect of w_{22} would increase the amount of energy lost to phonons. The combined effects of the three up-conversion processes is to decrease the amount of heating, but only when those same processes are at work improving the efficiency of the laser. For example, when in cw mode, rather than when in self-terminating mode.

5.2.2. Thermal lens formulation

When surface-cooling a gain medium, the heat is removed from the gain rod via the rod's surface; furthermore, the focus of the pump energy is at the centre of the rod. The rod is therefore hottest at the centre and a temperature gradient will be formed between the centre and surface of the rod. The temperature gradient across the rod can be modelled via the thermal diffusion equation to give, in the steady state case:

$$\Delta T(r) = - \left(\frac{H}{4\kappa} \right) r^2 \quad (5.1)$$

where T is temperature, r is radial distance from the rod's centre, κ is thermal conductivity and

$$H = \frac{P_h}{V_{rod}} \quad (5.2)$$

where P_h is total heat power from the flashlamp and V_{rod} is the volume of the rod that is being pumped.

The thermo-optic effect gives the laser rod a temperature-dependent refractive index, parameterised via the thermo-optical coefficient, which for YSGG is $\left(\frac{dn}{dT} \right)_{YSGG} = 12.3 \times 10^{-6} \text{K}^{-1}$ [117], where n is the refractive index.

Hence, equation 5.1 can be re-expressed to give the change in refractive index as a function of distance from the centre of the rod [108]:

$$\Delta n = - \left(\frac{H}{4\kappa} \right) \left(\frac{dn}{dT} \right) r^2. \quad (5.3)$$

Light travelling further from the centre of the rod will experience a smaller refractive index and therefore travel faster than light at the centre of the rod, therefore parallel wavefronts entering the rod will be curved when leaving the rod and will therefore focus. This forms a gradient index (GRIN) lens, and, when formed by a thermal gradient, the lens is simply known as a *thermal lens*. The dioptric power is given by

$$\frac{1}{f} = \frac{P_h l}{2\kappa V_{rod}} \left(\frac{dn}{dT} \right) \quad (5.4)$$

where l is length of the rod. See Fig. 5.2.

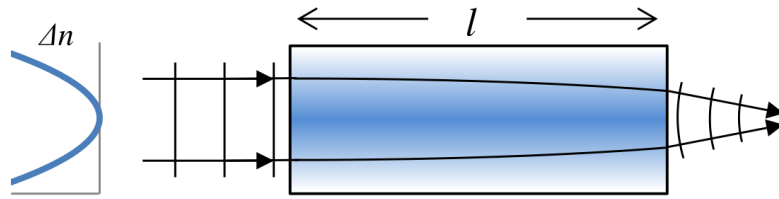


Figure 5.2.: Focussing effect of thermal GRIN lens on parallel rays due to the parabolic refractive index change within the rod. Light travelling further from the rod centre travels faster, bending the wavefronts of the light, focussing the light.

Equation 5.4 shows that the dioptric power of the thermal lens is proportional to P_h , the heat power absorbed by the rod; this is proportional to the total power delivered to the rod from the flashlamp: $P_h \propto P_{in}$, assuming that the heat transferred is due solely to the quantum defect. Thus

$$\frac{1}{f} \propto P_{in}. \quad (5.5)$$

The thermal lens is characterised by the constant of proportionality in this equation.

5.2.3. Other thermal effects affecting the laser rod

There are two additional thermal effects.

The first is due to thermal expansion and the inhomogeneous heating of the rod. Since the centre of the rod is hotter, this will expand more than the outside of

the rod, and hence the rod ends will bulge outwards, and thereby create a separate focussing effect. This effect can be treated alongside the GRIN aspect of the thermal lens introduced above, representing a minor deviation from the linearly proportional relationship of equation 5.5 [108].

The second additional effect is due to the stress-induced birefringence caused by the different rates of expansion across the rod cross-section. Since the birefringence has an angular dependence, off-axis light will lose its linear polarisation. This is of significance in cavities with polarisation-selective elements, and has been shown to be correctable with appropriate optics (see Yang *et al.*, introduced in sec. 4.4.1) [133], though no such correction is considered in this present work.

5.3. Thermal lens approximated by thin lens

To assess the effect of the thermal lens on the cavity, it is convenient to consider the thermal lens as a simple thin lens in between two half-rods of length $l/2$ of homogeneous refractive index, as shown in Fig. 5.3.

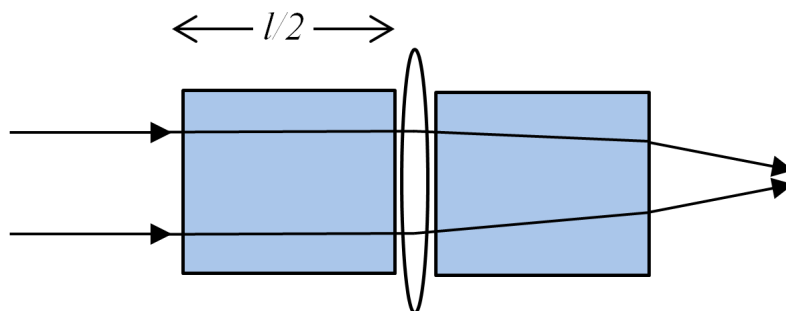


Figure 5.3.: The thermal lens may be approximated by a thin lens in between two rods of homogeneous refractive index, each one half the length of the full rod.

5.3.1. Equivalent cavity containing thermal lens

The behaviour of the thermal lens in the laser rod within a cavity can be simplified and represented by just a thin lens within a cavity of equivalent optical path length [145]. We consider the rod placed within a simple flat-flat cavity, with a thermal lens strong enough to focus the mode onto both mirrors, as shown in figure Fig. 5.4 a). The thermal lens can then be approximated as a thin lens between two rod

halves of homogeneous refractive index, as in Fig. 5.4 b). The refraction due to the thin lens and the rods can then be considered as all occurring at the principal planes of focus, at distances $h = l/2n$ from the rod ends (see Fig. 5.4 c)).

The distances from the principal planes to the cavity mirrors are L_1 and L_2 . The rod halves may then be considered removed, with the principal planes superimposed onto each other, and the cavity shortened to make the equivalent cavity of total length $L = L_1 + L_2$, as shown in Fig. 5.4 d).

This is an equivalent cavity with respect to longitudinal mode behaviour. Any cavity lengths mentioned during the analysis and investigation of the thermal lens, refer to the equivalent rather than the actual cavity length, unless otherwise stated.

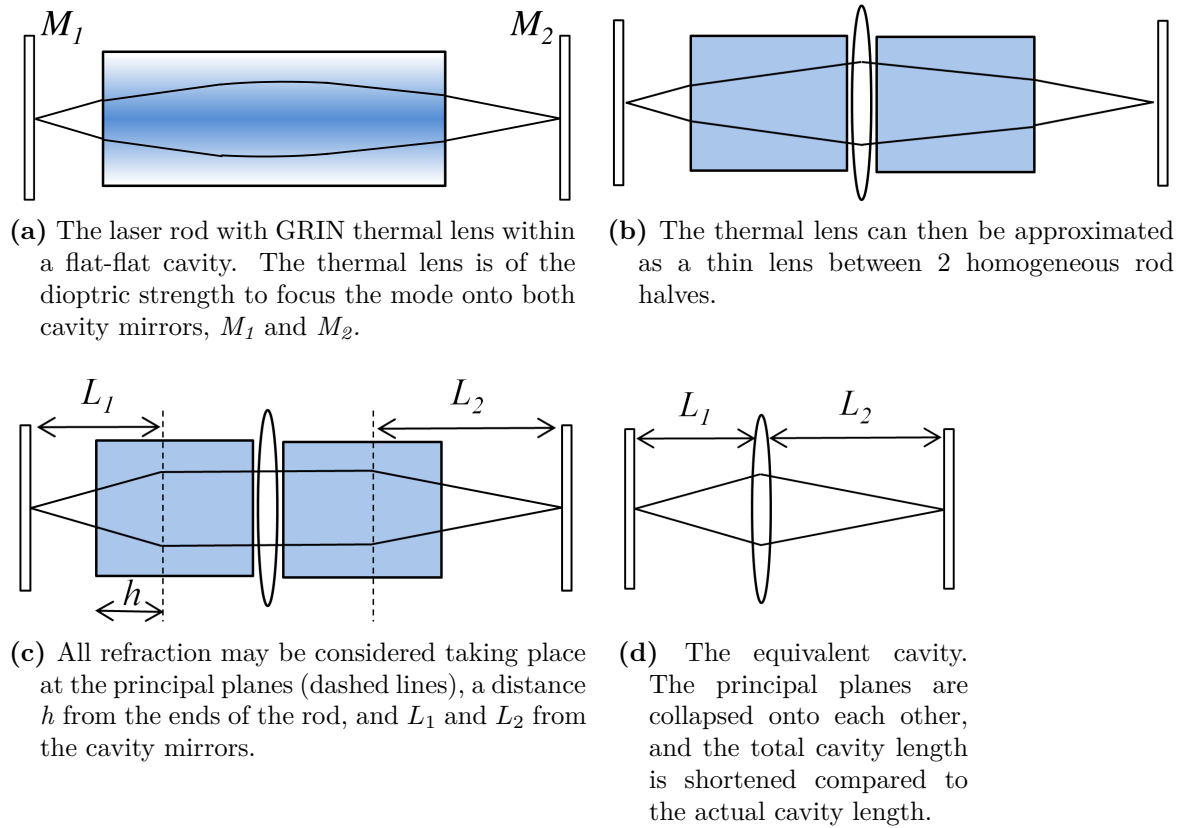


Figure 5.4.: The derivation of the equivalent cavity to be used during thermal lens investigations. A GRIN thermal lens is represented by a thin lens in an equivalent, shorter cavity.

5.3.2. Stability of modes in a flat-flat cavity containing a variable thin lens

Stability of a laser cavity *without* an intracavity lens is usually assessed with the use of g parameters of the cavity mirrors:

$$g_{1(2)} = 1 - \frac{L}{R_{1(2)}} \quad (5.6)$$

where $R_{1(2)}$ is the radius of curvature of the two cavity mirrors. A cavity is considered stable if the resonated beam size is refocussed during a round trip, and not allowed to grow uncontrollably in radius. This stability is expressed in the condition:

$$0 \leq g_1 g_2 \leq 1. \quad (5.7)$$

This is typically plotted in Cartesian coordinates; Fig. 5.5 shows the $g_1 - g_2$ plane with region shaded corresponding to stable cavities.

When an intracavity lens is introduced, this stability condition still applies. However, the g parameters are now modified to take into account the strength of the lens:

$$g_{1(2)} = 1 - \frac{L_{2(1)}}{f} - \frac{L'}{R_{1(2)}} \quad (5.8)$$

where L' is an effective cavity length (defined in Ref. [145]). Since, for simplicity, we are considering flat-flat cavities, $R_{1(2)} \rightarrow \infty$, and the last term of equation 5.8 goes to zero leaving:

$$g_{1(2)} = 1 - \frac{L_{2(1)}}{f}. \quad (5.9)$$

As the flashlamp power is increased, the dioptric power of the thermal lens will increase (equation 5.5) and the g parameters will change. Fig. 5.5 shows how the

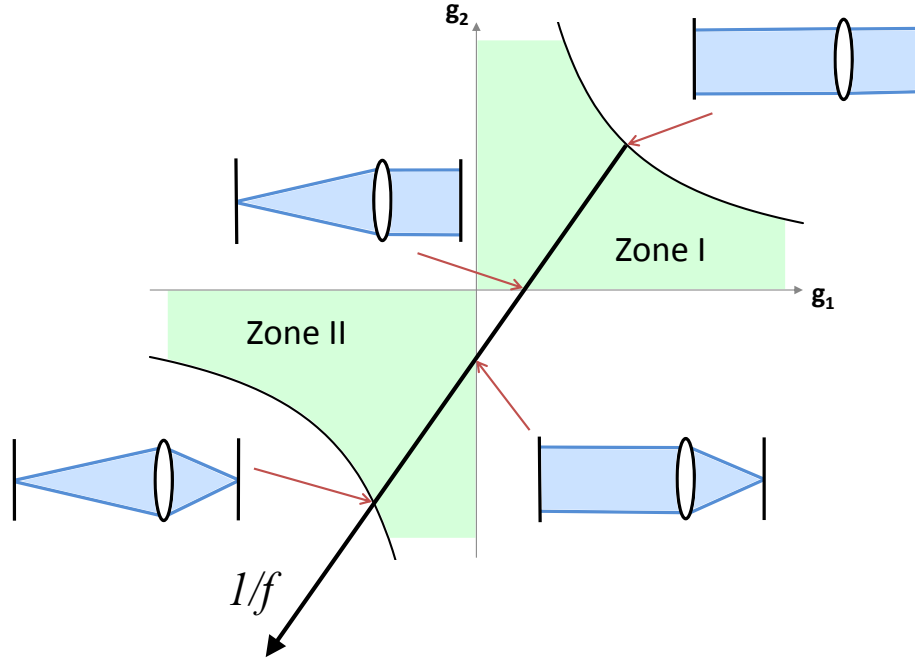


Figure 5.5.: The $g_1 - g_2$ plane showing shaded region indicating the 2 stability zones for cavities. The arrow indicates the path of the flat-flat cavity as its thermal lens increases in dioptric power. The cavity moves in and out of stability zones, with the corresponding longitudinal cavity modes given at the stability zone boundaries. This is the case of $L_1 > L_2$ [145].

increase in dioptric power of the thermal lens moves the position of an asymmetric flat-flat cavity across the $g_1 - g_2$ plane. When the dioptric power is zero, $g_{1(2)} = 1$ and the cavity takes the position of an ordinary flat-flat cavity with no thermal lens. As the thermal lens increases in power, the mode begins to focus, but the light stays within the cavity, the cavity remaining stable. At a point where the thermal lens focal length is equal to the longer cavity arm length ($f = L_1$ in this case), the cavity leaves stability zone I (when $g_2 = 0$). The cavity will remain in the unstable region until the point whereupon the thermal lens focal length equals the length of the shorter cavity arm ($f = L_2$ in this case, giving $g_1 = 0$). After this point, increasing the thermal lens strength will keep the cavity in stability zone II, until the dioptric strength at which the lens focusses the cavity mode onto both cavity mirrors ($\frac{1}{f} = \frac{1}{L_1} + \frac{1}{L_2}$). Then, if the thermal lens is stronger than this, all modes existing in the cavity will always have their focal point within the cavity, hence, upon reflection from the cavity mirrors, will diverge outside the cavity, leading to instability and no lasing.

5.4. Beam radius in cavity

To achieve efficient fundamental mode operation, the fundamental Gaussian cavity mode must be sufficiently wide throughout the gain rod. When pumping, population inversion will take place across a certain cross-section in the rod, an inversion region, and this is where lasing will be achievable. See Fig. 5.6. However, if the fundamental Gaussian mode of the cavity is small compared to the inversion region, then energy will be lost to higher order modes which have larger cross-sections.

It is possible to describe the beam radius ($1/e$) at the centre of the rod (length-wise, at the thermal lens position) as [145]:

$$w_3^2 = \frac{\lambda}{\pi} \left[\frac{4u_1u_2g_1g_2 + (u_1 - u_2)^2}{(1 - g_1g_2)g_1g_2} \right]^{\frac{1}{2}} \quad (5.10)$$

where $u_{1(2)} = L_{1(2)} \left(1 - \frac{L_{1(2)}}{R_{1(2)}} \right)$, which is simplified in the case of flat cavity mirrors to $u_{1(2)} = L_{1(2)}$.

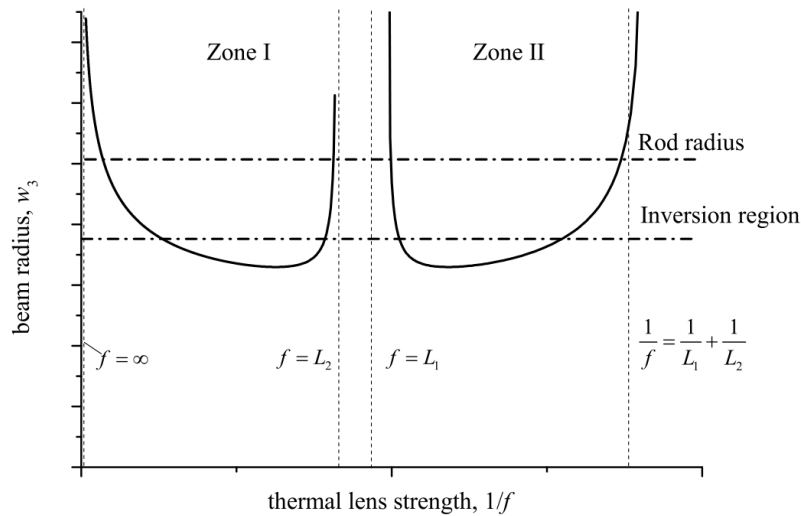


Figure 5.6.: Beam radius ($1/e$) in centre of laser rod (length-wise, at thermal lens position) for a flat-flat cavity for fundamental Gaussian mode. The two stability zones are seen, corresponding to those of Fig. 5.5, which are separated by a region where the beam radius diverges where there is no lasing. Shown in the dash-dotted lines are the population inversion region, which should ideally be matched with the beam radius, and the rod radius, within which all beam sizes must fit.

Equation 5.10, the fundamental mode radius, w_3 , is plotted in Fig. 5.6 against thermal lens strength, $1/f$. It shows two stability zones, I & II, corresponding to those of Fig. 5.5. Outside of these stability zones the beam radius diverges to larger than the rod radius and thus lasing is unstable and unlikely to occur.

The equivalent graphs for the higher order modes would have larger outer beam radii¹. If a higher-order mode radius is closer to the inversion region radius, the higher-order mode will likely out-compete the fundamental mode. The higher-order mode can be prevented from lasing with an intracavity aperture; however, this wastes much of the energy stored in population inversion. It is preferable to enlarge the fundamental mode to match the inversion region.

The minimum fundamental mode radius, w_{30} , is given by

$$w_{30}^2 = \frac{2\lambda}{\pi} \max(|u_1|, |u_2|) \quad (5.11)$$

$$= \frac{2\lambda}{\pi} \frac{1}{\left|\Delta \frac{1}{f}\right|} \quad (5.12)$$

where $\Delta \frac{1}{f}$ is the width of the stability zones, which is the same value for both zones. It is beneficial to match w_{30} to the inversion region radius, so that the laser will be more stable under fluctuations of the thermal lens power. The power of the thermal lens will vary with time - the flashlamp does not transfer heat continuously, and so it could be presumed that the temperature gradient of the rod has fluctuations, and so too, therefore, the thermal lens [108]. Because the minima of the stability zones are fairly flat, fluctuations in lens power will not cause a great change in the mode radius. This concept is termed *dynamic stability* [145]. Dynamic stability works better with wider stability zones (larger $\Delta \frac{1}{f}$), but enlargening $\Delta \frac{1}{f}$ will simultaneously decrease w_{30} (equation 5.11), which is unwanted. Therefore, an optimum midpoint must be found.

The size of w_{30} may be manipulated by changing the length of the cavity and the curvature of the mirrors, $R_{1(2)}$, whilst using equations 5.10 and 5.11. Otherwise, intracavity lenses may be inserted and ‘ABCD formalism’ used to determine w_3 ,

¹The *boundaries* of the stability regions would be the same since this is dictated by the cavity length and thermal lens strength

though this is not done in this present work.

5.5. Experimental measurement of thermal lens

To control the fundamental mode size in our system, it is necessary to ascertain the thermal lens strength for typical flashlamp energies and flash repetition rates. At the edges of stability zones, the focal length depends on the lengths of the arms L_1 and L_2 . The relationship between f and input power P_{in} can thus be extracted.

5.5.1. Simple cavity design

To assess the thermal lens strength, the laser rod was placed within a flat-flat cavity and operated in free-running mode, as shown in Fig. 5.7. The cavity is flat-flat, with the back mirror a high-reflectivity dielectric mirror, and the output coupler an $R = 60\%$ dielectric mirror. The distances of the mirror to the rod are continually variable. The fact that the laser rod facets are Brewster-cut does not modify the arguments made in sec. 5.3 about the thermal lens, and the equivalent cavity length.

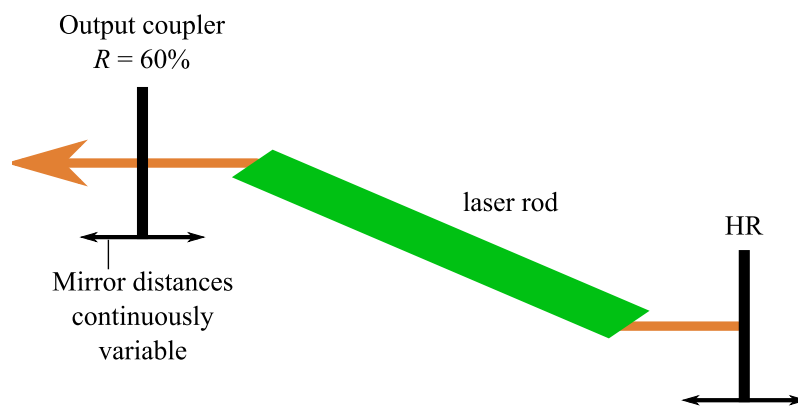


Figure 5.7.: Cavity setup for measurement of thermal lens strength.

5.5.2. Procedure for thermal lens measurement

At numerous cavity lengths, the output signal was measured from the laser over a range of pulse repetition rates. The flashlamp pump energy was kept constant, and only the repetition rate varied. Passing through a full range of pumping powers, the

two stability regions (roughly possessing the same width) could clearly be seen in asymmetric cavities, as shown in Fig. 5.8.

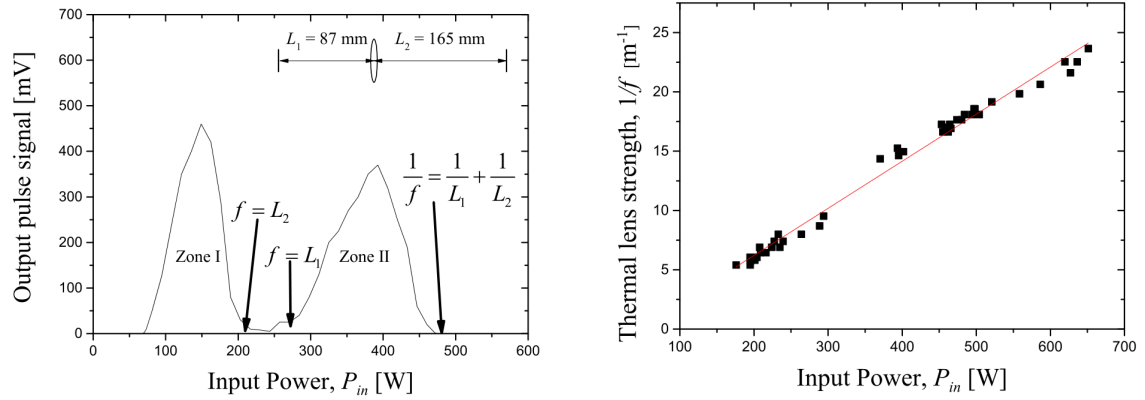


Figure 5.8.: (Left) The output pulse signal of the laser in the given configuration, against input power, showing the two stability regions of lasing. (Right) Thermal lens power $1/f$ versus power in P_{in} , showing linear relationship, with slope of $0.035 \pm 0.003 \text{ m}^{-1}\text{W}^{-1}$.

The edges of the stability zones corresponded to certain values of $1/f$, e.g. the higher-power edge of stability zone II corresponds to $\frac{1}{f} = \frac{1}{L_1} + \frac{1}{L_2}$. Fig. 5.8 shows the linear relationship between $1/f$ and P_{in} , which gave:

$$\frac{1}{f}/P_{in} = 0.035 \pm 0.003 \text{ m}^{-1}\text{W}^{-1}. \quad (5.13)$$

The thermal lens strength thus varied from $\frac{1}{f} \approx 5$ to 25 m^{-1} , or $f \approx 40$ to 200 mm as the input power varied from $P_{in} \approx 200$ to 700 W . P_{in} is related to the input voltage of the flashlamp, V_{in} , by

$$P_{in} = \frac{1}{2} C V_{in}^2 \times f_{pps} \quad (5.14)$$

and f_{pps} is the pulse per second (pps) of the flashlamp, $C = 120 \mu\text{F}$ the capacitance of the flashlamp capacitor banks, and $V_{in} \leq 1000 \text{ V}$. Input energy is limited to $E_{in} \leq 60 \text{ J}$, and pulse per second to $f_{pps} \lesssim 12 \text{ pps}$. The heating of the rod is taken as proportional to the input power, $P_h \propto P_{in}$ (see sec. 5.2).

5.5.3. Comparison of thermal lens strength with the literature

The behaviour of the thermal lens agrees, to within 5%, with a previous measurement [146] of the Er,Cr:YSGG thermal lens in a rod of similar dimensions ($\varnothing 5 \text{ mm} \times 80 \text{ mm}$). There, the thermal lens was measured from the variation of the focal length of a Nd:YAG laser passing through the rod. Ref. [147], too, found strong thermal lensing in Er,Cr:YSGG, of $6.67\text{--}10 \text{ m}^{-1}$ at a pumping power of 500 W in a shorter rod.

The strength of the thermal lens in Er,Cr:YSGG is higher than that of Er:YAG, where, for instance Ref. [148] measures, in a rod of similar dimensions to ours, a ratio between dioptric power and input power: $\frac{1}{f}/P_{in} \approx 0.005 \text{ m}^{-1}\text{W}^{-1}$. This indicates that, for similar pump powers, the YSGG rod will produce a thermal lens seven times the dioptric power as YAG would. Elsewhere, Ref. [149] calculated a focal lens power for Er:YAG of $\sim 3 \text{ m}^{-1}$ for 600 W pumping. In our system, at 600 W of pumping, the focal lens power is $\sim 20 \text{ m}^{-1}$.

Equation 5.4 shows that the dioptric power of the thermal lens is proportional to κ and $\frac{dn}{dT}$ which are properties of YSGG. The two garnets, YAG and YSGG, have similar thermal properties (see Tab. 4.1) [117]. So, the difference in thermal lens strengths is probably associated with the presence of Cr^{3+} in our YSGG rod, permitting more absorption of radiation, and more dissipation of thermal energy into the rod.

5.6. Obtaining Gaussian fundamental mode in laser rod

Equipped with the range of YSGG thermal lens strengths, it is possible to design cavities using equations 5.8 and 5.10, to give an optimum size of w_3 . Any cavity design must have laser stability zones residing within the P_{in} range of our system ($\lesssim 700 \text{ W}$).

5.6.1. Fundamental mode size with variation of cavity length

We have seen that the minimum value of the fundamental mode radius, w_{30} , is proportional to the length of the longest cavity arm in the flat-flat cavity configura-

tion (equation 5.11). Therefore, it should be possible to achieve fundamental mode operation with the appropriate length of cavity arms.

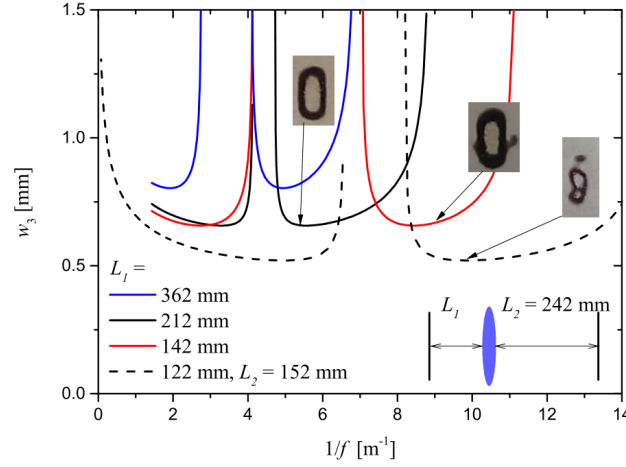


Figure 5.9.: Theoretical behaviour of w_3 in flat-flat cavity when one cavity arm, length L_1 , is varied, and the other kept constant. w_3 will increase if L_1 exceeds L_2 (blue line). Varying $L_1 (< L_2)$ will only shift the position of zone II in $1/f$, and not increase w_{30} . Note also that the larger the value of w_3 , the narrower the stability zone (smaller $\Delta \frac{1}{f}$). Beam profiles are also shown at three points.

Fig. 5.9 shows the theoretical behaviour of the mode size as the cavity length is altered, along with beam profiles measured from heat-sensitive paper². It can be seen that a short cavity (dashed line), with $L_1 = 122$ mm and $L_2 = 152$ mm, gave a TEM₀₂, multi-mode laser pulse output. This cavity configuration corresponded, theoretically, to a minimum fundamental mode radius of $w_{30} \approx 0.5$ mm. The mode shapes are elliptical due to the Brewster-cut facets of the laser rod forming a limiting horizontal aperture.

It can be seen that, with the right hand cavity arm length increased to $L_2 = 242$ mm, the fundamental mode minimum was increased to $w_{30} \approx 0.66$ mm. With the left hand arm length set to $L_1 = 142$ mm, the cavity mode appears close to fundamental mode, but somewhat unclean. When L_1 is increased to $L_1 = 212$ mm, we see that the mode is much cleaner and apparently fundamental mode, which is corroborated in sec. 5.6.2.1. This suggests that to achieve fundamental mode lasing, it is not just the value of w_{30} which is important, but also the overall length of the cavity. It is possible that unwanted, higher order modes are not stable in longer cavities due to their higher divergence. Cavities with $L_2 = 242$ mm are further investigated in

²i.e. receipt paper.

sec. 5.6.2.1.

Also shown in Fig. 5.9 is the value of w_3 for a longer cavity ($L_1 = 362$ mm, $L_2 = 242$ mm), where $w_{30} \approx 0.8$ mm. This cavity configuration, when attempted, was found to not lase with sufficiently high output energy, probably due to the increased instability at long lengths, and increased intracavity absorption from atmospheric moisture.

5.6.2. Laser pulse output energy

It is important that fundamental mode lasing is accompanied by adequate output pulse energy. Fig. 5.10 shows a survey of output laser pulse energies, E_{out} , against input power, P_{in} , with fixed right hand cavity arm length ($L_2 = 242$ mm) and variable left hand cavity arm length ($L_1 = 72, 142, 212$ mm), and varying input flashlamp pulse energy, E_{in} . (The beam profiles shown in Fig. 5.9 are partially taken from this survey.)

5.6.2.1. High-energy, fundamental mode lasing

At $L_1 = 212$ mm, the cavity arms are of similar length, hence zone I and II are merged. The peak output energy was $E_{out} = 76$ mJ, occurring when $E_{in} = 60$ J, which constituted the highest output energy from our free-running laser. Furthermore, at this cavity length, fundamental mode lasing was seen for all of the values of E_{in} . Fundamental mode lasing was corroborated by knife-edge beam profile measurements (see sec. A.1), as shown in Fig. 5.11, and measurements of the beam radius across the laser waist (focussed, external to the cavity, by a BaF₂ lens), as shown in Fig. 5.12. These revealed that in the horizontal (x) and vertical (y), the beam quality factor was unity, $M_x^2 = M_y^2 \approx 1$.

This was the final configuration of the free-running laser cavity, to be Q switched in chapter 6: $L_1 = 212$ mm and $L_2 = 242$ mm, with peak output pulse energy, $E_{out} = 76$ mJ.

5.6.2.2. Output behaviour of shorter cavities

As shown in Fig. 5.10, when $L_1 = 142$ mm, peak output energy was $E_{out} = 67$ mJ, occurring in zone I for $E_{in} = 60$ J. As discussed (sec. 5.6.1) this had the appearance of

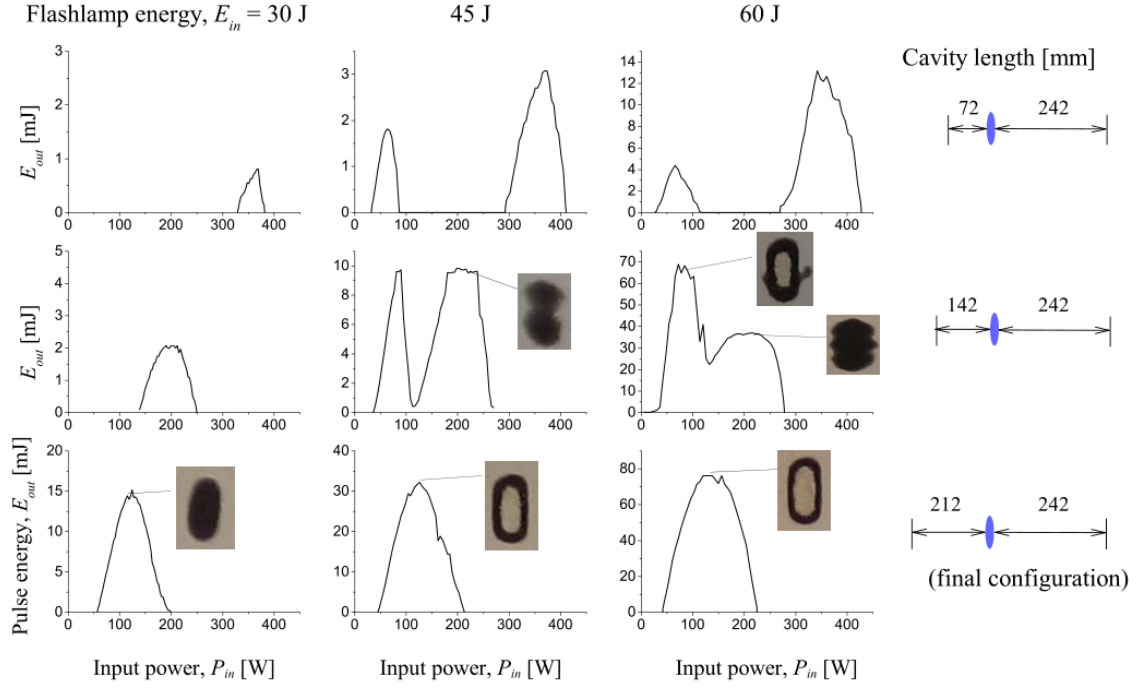


Figure 5.10.: A survey of laser pulse energy, E_{out} , versus input power, P_{in} , with variation of left hand cavity arm length, L_1 , in a flat-flat cavity, and input flashlamp energy, E_{in} . Images of the beam profiles scorched onto thermal paper are shown, demonstrating the transverse mode at particular points. Highest energy pulses (>70 mJ) were recorded for the cavity with 212 mm left hand cavity arm, which also corresponded to the best, fundamental mode emission - this mode configuration was the final configuration settled on. Modes are elliptical due to the Brewster-cut rod forming an aperture in the horizontal direction; consequently, any multimode behaviour is seen only in the vertical.

an untidy fundamental mode. At this same input energy the zone II mode produced less energy, and was also evidently TEM_{02} . At a lower input energy, $E_{in} = 45$ J, the two stability zones had the same output energy as each other ($E_{out} \approx 10$ mJ), and the zone II mode was TEM_{01} - this is presumably because the inversion region radius of the $E_{in} = 45$ mJ pulse was smaller than with $E_{in} = 60$ mJ.

Reducing L_1 further, to $L_1 = 72$ mm, made the cavity more asymmetric, and lowered the output pulse energy to $E_{out} < 12$ mJ. This was too low to record a beam profile on heat-sensitive paper. The two stability zones are truly separate at this cavity configuration.

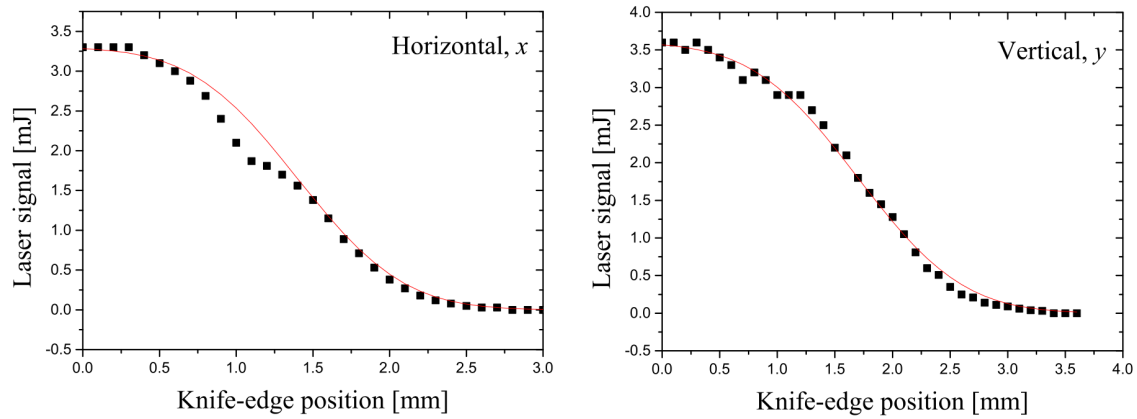


Figure 5.11.: Knife-edge measurement of laser mode in horizontal (x) and vertical (y), 560 mm from the laser output coupler, black squares, for the laser configured as for the ‘final configuration’ in Fig. 5.10. The red lines are the fittings which correspond to Gaussian mode of equivalent width.

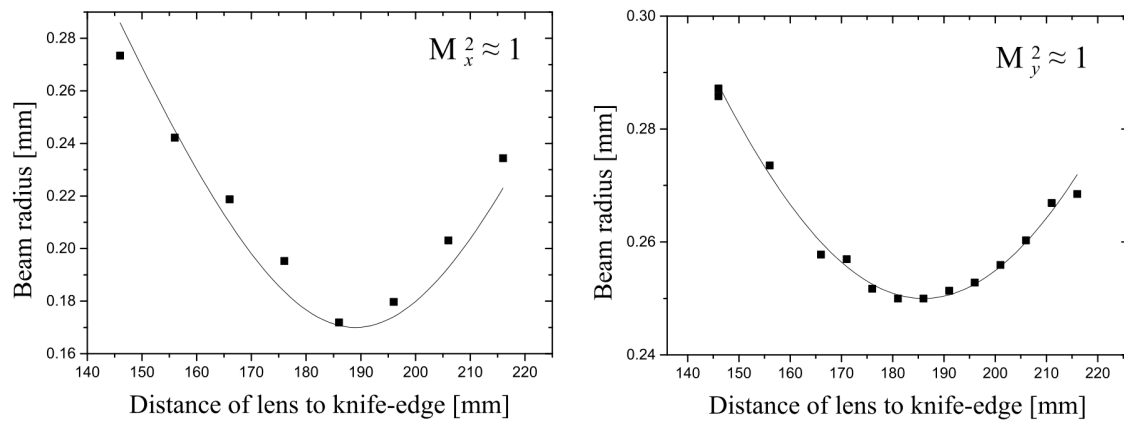


Figure 5.12.: M^2 measurements for vertical and horizontal directions of laser beam, demonstrating $M_x^2 = M_y^2 \approx 1$ for the laser configured as for the ‘final configuration’ in Fig. 5.10.

6. Slow Q switching the erbium laser

This chapter presents an investigation into Q switching the Er,Cr:YSGG laser. Presented is the theory of Q switching, and our own investigations into slow Q switching the $\lambda \simeq 3 \mu\text{m}$ erbium laser, including methods new to the Er,Cr:YSGG system.

6.1. Theory of Q switching

We now outline the theory of Q switching, how the $3 \mu\text{m}$ erbium laser Q switches, and the various Q switching methods that have been reported.

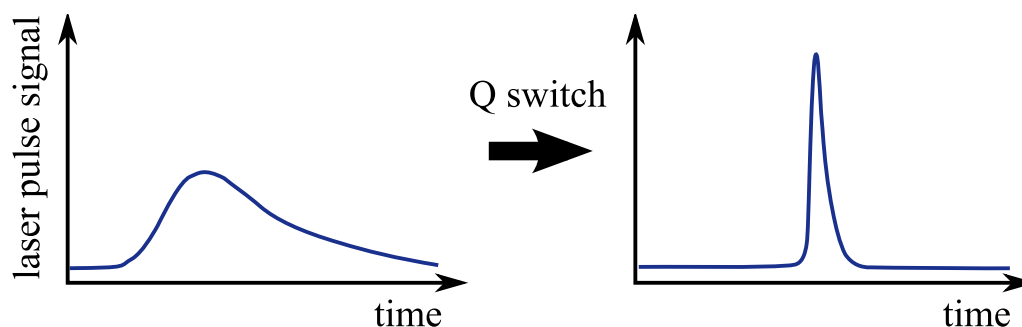


Figure 6.1.: Demonstration of the effect of Q switching. A free-running pulse, without Q switching (left), and the laser pulse after Q switching (right). Q switching shortens pulsewidth and increases pulse intensity.

6.1.1. Q switching overview

Q switching is a common method for creating short, high-energy laser pulses, and was demonstrated in 1962, not long after the invention of the laser itself [150]. Q switching involves introducing a loss mechanism into a laser cavity during pumping; population inversion increases, but no lasing occurs since the optical feedback mechanism of the cavity is defeated. When population inversion is high, the loss

mechanism is removed; the quality factor, Q , is ‘switched’ to its maximum value in a time t_Q .¹ At this point, cavity radiation builds up from spontaneous emission, and when lasing occurs, because the upper laser level population is at a higher level than usually permitted, stimulated emission of a giant pulse happens, in a short space of time [151]. See Fig. 6.2.

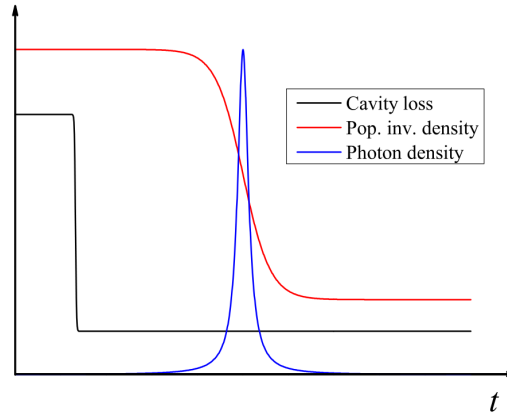


Figure 6.2.: A diagrammatic explanation of Q switching. Population inversion density (red line) is pumped to a high value whilst the cavity losses (black line) are high. Suddenly switching the cavity loss to a low value (Q switching) brings the cavity to above the threshold condition. This allows the photon density (blue line) to build up, and a giant laser pulse to be emitted.

6.1.2. Methods of Q switching

There are various methods of Q switching, typically categorised into whether they are active or passive (requiring input energy to function, or not), and if they are fast or slow (pertaining to Q switching time relative to laser pulse build-up time).

6.1.2.1. Active, fast Q switching

Some of the common active methods of Q switching do not require moving parts, and generally fall into the category of *fast* Q switches, with Q switching times, t_Q , of under hundreds of nanoseconds.

Electrooptic Q switch

¹ Q is defined as the ratio of the energy stored in the cavity to the energy loss per cycle.

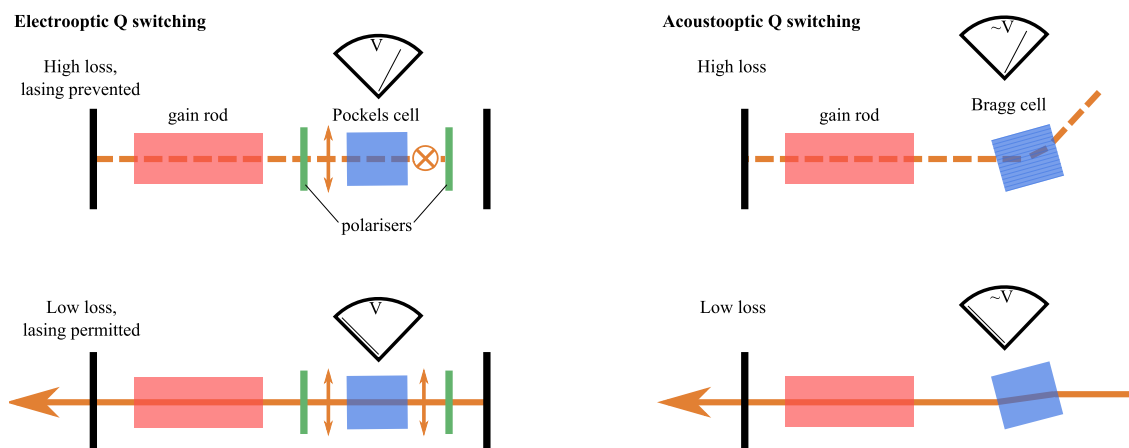


Figure 6.3.: Examples of active methods of fast Q switching. Back mirror and output coupler are black lines, and laser mode is the orange line. (Left) Electrooptic Q switch, with Pockels cell. When a high voltage is applied across the Pockels cell, it acts as a half-wave plate. Two linear polarisers of the same orientation will block radiation that has passed through the half-wave plate, creating high loss, preventing lasing. With zero voltage across the cell, the polarisers select the polarisation of the lasing mode, and the cavity has low loss; lasing is permitted. (Right) Acoustooptic Q switch with Bragg cell. Piezoelectric transducer transmits high frequency vibration into the Bragg cell which deflects the laser mode outside of the cavity, causing high loss. With no voltage applied, the system has low loss.

In the electrooptic Q switch (EOQS), a transparent, optical cell is aligned within the laser cavity. With no voltage applied across the cell, the laser mode passes through the cell with only transmission losses depleting its magnitude. This corresponds to the cavity with low loss.

When a voltage is applied to the cell, the refractive index of the cell is altered², such that the cell becomes a quarter- or half-wave plate (half wave plate case shown in Fig. 6.3) - the polarisation of the laser mode is rotated as it passes through the cell. Another element within the laser cavity, with polarisation-dependent loss, prevents lasing of the mode in the altered polarisation. This corresponds to the high loss mode.

The application time of voltage, and therefore the switching time of the EOQS, can be very short, under one hundred nanoseconds, allowing rapid switching.

The electrooptic cell can be either a Kerr cell or a Pockels cell. The change in

²The cell becomes birefringent.

refractive index, specifically of the birefringence, of a Kerr cell is proportional to the square of the applied electric field, whereas in a Pockels cell the refractive index is *linearly* proportional to the electric field. Kerr cells, therefore, require slightly smaller voltages (<1 kV) compared to those required of Pockels cells (>1 kV).

Both types of cell have been applied to the erbium laser system. Ref. [152] has a direct comparison of the Kerr and Pockels cell methods, with the Pockels cell faring better in terms of energy output. The Pockels cell is the more commonplace Q switch in the erbium system, and is investigated as part of this thesis. (See sec. 6.2 for more theory and examples of existing work.)

Acoustooptic Q switch

For the acoustooptic Q switch, an acousto-optic Bragg cell is placed into the cavity, and a piezoelectric transducer transmits high frequency vibrations into the cell. The standing wave within the cell creates alternating high and low regions of refractive index which forms a type of diffraction grating in the cell. The defraction grating deflects the lasing mode, thus introducing high cavity losses. See Fig. 6.3. Workers reporting in Ref. [153] utilised a germanium Bragg cell to Q switch an Er:YAG and Er,Cr:YSGG laser citing ~ 10 μ J of energy in ~ 100 ns pulsewidths. Elsewhere, the Er:ZBLAN system was Q switched acoustooptically to produce 100 μ s pulses at a repetition rate of 120 kHz [154]. Q switching times are typically 100 ns or less.

6.1.2.2. Active, slow Q switching

Other active Q switch methods require rapidly moving parts, so called *mechanical* methods. Because of practical limitations of mechanical movement, these are typically *slow* Q switches. They are called ‘slow’ because Q switching time, t_Q , is slow with respect to the laser pulse build up time, t_d .

Rotating mirror Q switch

A popular method of slow Q switching is the rotating mirror Q switch (RMQS). In this scheme a cavity (or intracavity) mirror is rotated at a high rate; when the mirror is unaligned with the cavity the loss is high and there is no lasing. When the mirror is aligned with the cavity, the loss is temporarily minimised, and lasing may occur [155, 156]. See Fig. 6.4.

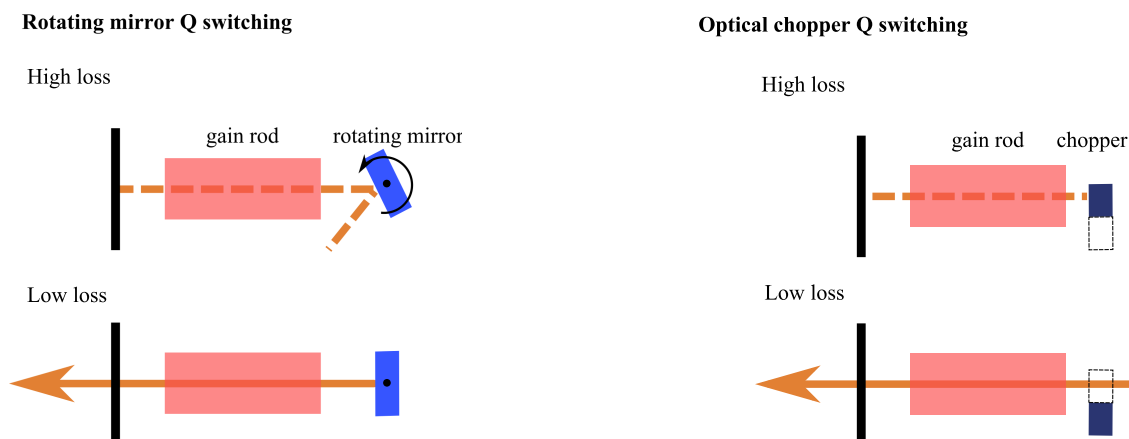


Figure 6.4.: Active, slow methods of Q switching. (Left) Rotating mirror Q switch. One of the cavity mirrors is rotated at high frequency. When the rotating mirror is not aligned, the cavity has high loss. (Right) Optical chopper Q switch. A chopper blocks the laser beam giving high loss, and is rapidly moved out the way for the low loss period.

Switching time from high to low loss is not as quick as the fast methods of Q switching, being limited to rotation rates of mirrors, which may only realistically go to around 500 rotations per second (rps). At this rate the mirror may sweep through 10 mrad in about 8 μ s. This constitutes a rough estimate for the Q switching time, t_Q , for this system, assuming that 10 mrad of mirror misalignment is sufficient to prevent lasing and allow population inversion growth. This itself depends on the properties of the cavity, and the pumping power.

As such, the RMQS favours laser materials with longer upper lasing level lifetimes, so that population inversion is not lost during the slow Q switching time.

The RMQS has been well developed for Er:YAG laser (see sec. 4.4.1). In this thesis, the RMQS will be implemented into the Er,Cr:YSGG system. (See sec. 6.3.)

Optical chopper Q switch

The optical chopper Q switch (OCQS) consists of the insertion of a high frequency chopper into the cavity. When the chopper is obscuring the path of the cavity mode the losses are very high. When the chopper is not obscuring the path of the cavity mode, the loss is at its minimum, and lasing can occur. See Fig. 6.4.

Previous examples of the OCQS exist in CO₂ lasers [157, 158] and certain fibre lasers [159]. Instances do not appear widespread, possibly because of the difficulty

in producing a chopper fast enough to provide sufficiently quick switching time for most laser media.

A form of the OCQS will be implemented for the first time in the Er,Cr:YSGG system in this thesis. (See sec. 6.4).

Frustrated total internal reflection Q switch

The final active Q switch to be introduced here is based upon frustrated total internal reflection (TIR), and is denoted here as the ‘FTQS’. The amount of frustrated TIR occurring in a prism reflector can be modulated by varying the distance between the reflecting prism and a second, adjacent prism. When the prisms are far apart, TIR occurs in the reflecting prism. When the prisms are close, frustrated TIR occurs, splitting the path of the light, thereby introducing a loss mechanism to the cavity [160].

This technique is a promising method of Q switching, since the modulator can be made of virtually any material that is transparent to $\lambda \simeq 3 \mu\text{m}$. The laser designer is free to choose whatever material for the Q switch to minimise insertion loss and the chance of optical damage, for example, a YAG-based Q switch (see Högele *et al.*, sec. 4.4.2).

6.1.2.3. Passive Q switching

Passive methods of Q switching make use of intracavity saturable absorbers which have intensity-dependent transmission. At low photon intensity, transmission is low and therefore cavity loss is high. At higher photon intensity, which occurs when population inversion is high, transmission of the saturable absorber is high and therefore cavity loss is low.

There are many examples of suitable saturable absorbers for Q switching, with the requirement being that the material absorbs at the laser wavelength, and has a fairly step-like intensity saturation action.

For the erbium laser, saturable absorber materials implemented in the literature include water [153], ethanol [161] and InAs [94, 160]. The absorber is often susceptible to damage since it must take a certain thermal load, and as such its use is not widespread. Passive Q switching also offers less control of pulse timings as does active Q switching. For more, see Ref. [108].

6.1.3. Q switched laser pulse properties

The losses experienced by a Q switched cavity can be expressed as

$$\gamma(t) = \ln \frac{1}{R} + \gamma_0 + \gamma_Q(t) \quad (6.1)$$

where R is the reflectivity of the output coupler (assuming back mirror is of high-reflectivity), and γ_0 is the background cavity loss, including the Q switch insertion losses. $\gamma_Q(t)$ is the time-varying loss of the Q switch, which has a high of γ_{Qmax} and a low of 0. The constant, minimum loss can be expressed as $\gamma_{min} = \ln \frac{1}{R} + \gamma_0$, such that $\gamma(t) = \gamma_{min} + \gamma_Q(t)$.

The lasing threshold can be expressed as

$$2\sigma l n_i \geq \gamma(t) \quad (6.2)$$

where σ is the stimulated emission cross-section and l is the gain medium length.

Population inversion density condition for achieving Q switching

The value of the initial population inversion density after pumping, n_i , depends on how lossy the high-loss state is: the more lossy, the harder the laser can be pumped. The maximum value attainable of n_i , whilst maintaining Q switching, is

$$n_{imax} = \frac{1}{2\sigma l} \left(\ln \frac{1}{R} + \gamma_0 + \gamma_{Qmax} \right). \quad (6.3)$$

Simultaneously, n_i must surpass the threshold population inversion density of the laser in its low-loss state, otherwise no lasing will occur at all. The threshold population inversion density is:

$$n_t = \frac{1}{2\sigma l} \left(\ln \frac{1}{R} + \gamma_0 \right) = \frac{1}{2\sigma l} \gamma_{min}. \quad (6.4)$$

Thus, the condition for Q switched lasing is

$$n_{imax} > n_i > n_t. \quad (6.5)$$

Q switched laser pulse output energy

Theoretical treatments [108, 155] give the total laser pulse energy as

$$E_{out} = \frac{1}{m} S l \hbar \omega (n_i - n_f) \frac{\ln(1/R)}{\ln(1/R) + \gamma_0} \quad (6.6)$$

where $\hbar \omega$ is the laser photon energy, S the effective beam cross-sectional area, and m a factor which equals 1 for four-level lasers, and 2 for three-level lasers.

The effectiveness of a Q switch in creating high-energy pulses thus depends on the maximisation of the change between initial and final inversion population density, $n_i - n_f$.

n_f is related to n_i and n_t in a transcendental equation, as follows:

$$n_i - n_f = n_t \ln \frac{n_i}{n_f}. \quad (6.7)$$

A lower value of n_t will result in a lower n_f , and therefore a greater $n_i - n_f$, which will lead to an increase in pulse energy. n_t is lowered by having low background loss, γ_0 , and having higher-reflectivity output coupler, R . However, the R -dependence in equation 6.6 shows that a lower R will lead to higher pulse energy. As such, there is an optimum value of R for the highest pulse energy.

Q switched laser pulsewidth

The pulsewidth of the laser pulse (FWHM pulsewidth) is, likewise, partially determined by the magnitude of $n_i - n_f$, and is expressed as [108]

$$t_p = t_r \frac{n_i - n_f}{n_i - n_t [1 + \ln(n_i/n_t)]}. \quad (6.8)$$

Firstly, minimising the round-trip time, t_r , will shorten the pulsewidth. Secondly, the minimum pulsewidth for a given system requires a fine balancing of n_t with respect to n_i , such that, in equation 6.8, the numerator is as small, and the denominator as large, as possible.

6.1.4. Theoretical analysis of slow Q switching

A brief mathematical treatment of slow Q switching is presented below.

6.1.4.1. Definition of slow Q switching

Q switches are categorised as either fast or slow. Fast Q switches, as introduced in sec.6.1.2.1, are those in which the switching time, t_Q , is effectively instantaneous, $\gamma(t)$ a step function. The build-up of the photon population in the cavity and of the laser pulse happens entirely after the cavity has switched to minimum loss. See Fig. 6.5 (left).

In a slow Q switch, the cavity loss, $\gamma(t)$, changes gradually. As such, the Q switching time, t_Q , of the cavity loss changes on a time scale similar to the time it takes for the pulse to emit after the threshold condition (equation 6.2) has been satisfied, a time known as the build-up time, t_d .

See Fig. 6.5 (right). Mechanical, active Q switches, as introduced in sec. 6.1.2.2, are slow Q switches - the RMQS, for example, is constrained by the speed of the motor moving the rotating mirror into alignment with the cavity.

6.1.4.2. Laser pulse emission with slow Q switching

At some point, as $\gamma(t)$ drops in a slow Q switch, the threshold condition (equation 6.2) will be satisfied and photon density will begin to grow. At a time t_d later, a pulse will be emitted when the photon density reaches a critical value, a value cited as typically a twentieth of the maximum photon density of the pulse [155].

To the detriment of energy efficiency, whilst photon density is growing, the cavity loss will not yet be at its minimum, γ_{min} , resulting in some loss in stored energy. In an RMQS, for example, the lasing threshold condition will be satisfied whilst the mirror is a number of milliradians away from perfect optimal alignment.

Coinciding the end of build-up time with minimum cavity loss

Simultaneously, it is crucial, for efficient slow-regime Q switching, that the laser pulse emission coincides with the onset of minimum cavity losses, e.g. when the cavity mirror is perfectly aligned in an RMQS. See Fig. 6.6, a). The build-up time,

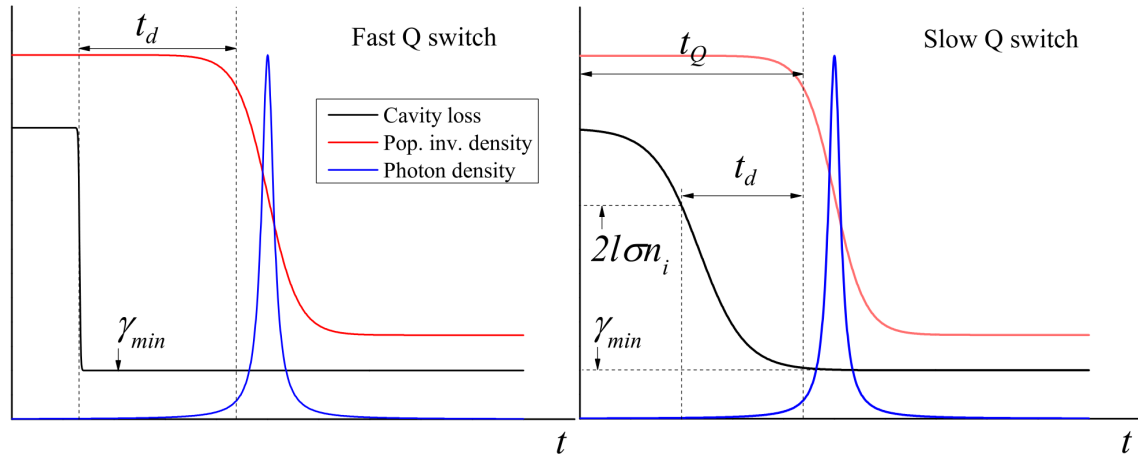


Figure 6.5.: Two speeds of Q switching. (Left) Diagrammatic explanation of fast Q switch operation. Initially high cavity loss prevents lasing, holding population inversion high. Cavity loss is reduced to a minimum quasi-instantaneously and at a build-up time, t_d , later the laser pulse is emitted, whilst the population inversion density is depleted. (Right) Slow Q switch operation, where the Q switching time, t_Q , is comparable to the build-up time, t_d , of the laser pulse. Build-up begins when cavity loss is equal to the laser gain, $2l\sigma n_i$. Ideally, the emission of the pulse coincides with when the cavity loss is at its minimum value, γ_{min} .

t_d , is dependent on the round trip time of the cavity, the cavity losses, and the spontaneous emission lifetime of the upper laser level, t_f . Thus the suitability of a given slow Q switch depends on certain cavity design constraints, and on the properties of the laser medium - typically, only lasers with longer upper laser level lifetimes can tolerate slow Q switching.

Breakthrough lasing from pumping too hard with respect to Q switch speed

The performance of slow Q switches are also dependent on the amount of pumping. The harder the gain is pumped, the higher n_i will be. The threshold condition will thus be satisfied at a larger value of $\gamma(t)$, earlier in time. Therefore, if the laser pulse is to coincide with the minimum cavity loss, the Q switch will have to be switched faster. The formation of a pulse before the onset of minimum $\gamma(t)$ is known as breakthrough lasing. See Fig. 6.6, b). Furthermore, secondary pulses may be emitted after the initial pulse. For a given pumping power, and associated initial population inversion, n_i , there is an optimum Q switching time, t_Q .

Late pulse emission from insufficiently short low loss interval

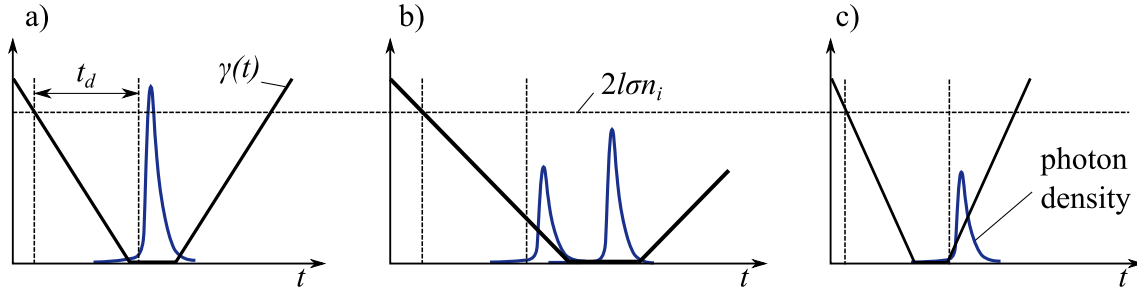


Figure 6.6.: Different slow Q switching scenarios for various Q switch speeds for a given laser system. Pulse build up time, t_d , begins when $\gamma(t) \leq 2l\sigma n_i$, i.e. the threshold condition. a) Ideally, the Q switch reaches minimum cavity loss when the pulse is emitted. b) If the Q switch is too slow, breakthrough pulses will occur before $\gamma(t)$ is at its minimum, and additional pulses may occur to fully deplete the population inversion. c) If the Q switch is too quick, the laser pulse may be too late for the period of minimum loss (right). $\gamma(t)$ is given a linear trend for simplicity [155].

Another potential problem for a slow Q switch may arise if the interval, or window, of low loss is short. See Fig. 6.6, c). If, for example, the rotating mirror is rotating too rapidly, the cavity may pass in and out of low loss interval during the laser pulse build-up, resulting in the pulse being emitted when there are higher losses, or not emitted at all. See Fig. 6.6.

6.1.4.3. Expression for slow Q switching pulse energy

The Q switched laser pulse energy, described by equation 6.6, may be re-expressed as an analytical equation with a substitution. If n_i is not too much greater than n_t , then the transcendental equation (6.7) can be simplified to [155]

$$n_i - n_f \approx 2(n_i - n_t). \quad (6.9)$$

This assumption is often true of slow Q switches, due to the increased risk of breakthrough lasing if there is excessive pumping (i.e. higher values of n_i). n_i is generally closer in value to n_t than for the case of fast Q switching. Substituting equation 6.9 into equation 6.6, and assuming the pulse is emitted at the cavity loss minimum,

gives a pulse energy out of

$$E_{out} = \frac{S\hbar\omega t_d |d\gamma/dt|}{m\sigma} \frac{\ln(1/R)}{\ln(1/R) + \gamma_0} \quad (6.10)$$

where $|d\gamma/dt|$ is the rate of change of cavity loss, the Q switch speed.

Evidently, a faster Q switch will achieve the highest energy pulses. Also, a lower stimulated emission cross-section, σ , favours high-energy output - having this property allows a higher population inversion to be reached.

Using the approximation of equation 6.9 means that, according to equation 6.10, R should be minimised to maximise E_{out} . In practical terms, one would not desire a value of R too low because the laser threshold would be too high to be practically functional. But for maximum pulse energy, a lower R should be considered for a slow Q switch laser.

6.1.4.4. Previous slow Q switching of erbium

Georgescu and Lupei [129] examined the case of Er:YAG with an idealised, fast (step-function) Q switch versus a slow Q switch with a Q switching time of $t_Q = 1.5 \mu\text{s}$, corresponding to the FTQS Q switching time of Högele *et al.* [142] (see sec. 4.4.2). They found little theoretical difference in the output energy between the idealised and the slow Q switch when the cavity length exceeded ~ 500 mm in length, which happens to be the length of our own cavity (see sec. 6.2.1).

As introduced in sec. 4.4.2, the work of Högele *et al.* yielded Q switched laser pulses of 55 mJ with 43 ns pulsewidths from Er,Cr:YSGG. They found that the long fluorescence time of the upper laser level (1.3 ms) in Er,Cr:YSGG, and the small stimulated emission cross-section ($2.8 \times 10^{-21} \text{ cm}^2$) allowed the slow Q switch to successfully produce high-energy pulses.

Slow Q switching has also been thoroughly demonstrated in Er:YAG, with the RMQS [134] and, again, the FTQS [162]. Er:YAG has an upper laser level lifetime (0.12 ms) some 10 times shorter than in Er,Cr:YSGG, but has the same stimulated emission cross-section, σ . This would suggest that σ is the more important property in determining the performance of the slow Q switch, rather than the laser level lifetimes.

6.1.4.5. Intrinsic limiting efficiency of 3 μm erbium in Q switched regime

Theoretical calculations by Georgescu *et al.* [163] have examined the efficiency of the Q switch regime in 3 μm erbium lasers. They point out that the energy up-conversion processes, introduced in sec.4.2.2, which facilitate the cw regime, are less effective in the Q switch regime. This is because up-conversion does not occur in the timeframe of a single Q switch pulse, thus cannot replenish the upper laser level and enhance the pulse energy. Thus, once the lower laser level has been filled, and the Q switched pulse has been emitted, there still remains a large amount of stored energy which was not extracted by the laser pulse ($\sim 90\%$).

6.1.4.6. Theoretical optimum pumping wavelength for 3 μm erbium

Georgescu and Lupei [129] theoretically examined the performance of the Q switching Er:YAG laser, when pumped with discrete wavelengths, and with broadband flashlamp light. They found that flashlamp pumping was the least efficient method, even predicting lower output pulse energy than when directly pumping the *lower* laser level. The distributed large quantum defect when flashlamp pumping was cited as the cause for the inefficiency. The most efficient wavelength was the direct pumping of the upper laser level ($\lambda \approx 970$ nm, to $^4\text{I}_{11/2}$), followed by the pumping of the $^4\text{I}_{9/2}$ level ($\lambda \approx 790$ nm). Nevertheless, the flashlamp pumping of erbium is more developed, and, as reported in the literature, is able to deliver more energy and make up for the lower efficiency.

6.2. Preliminary Q switching results

6.2.1. General cavity design for all Q switched cavities

The laser cavity which will be Q switched is as introduced in the previous chapter (sec. 5.6.2.1), which achieved high-energy (< 76 mJ), fundamental mode lasing. It is shown again for convenience in Fig. 6.7. The back mirror is high-reflectivity dielectric and the output coupler is an $R = 60\%$ dielectric mirror. The effective cavity arm lengths are $L_1 = 212$ mm and $L_2 = 242$ mm. These lengths corresponds to actual distances from rod-end to cavity mirror of 190 and 220 mm, respectively, giving a

total cavity length of 500 mm. The Q switches will be placed to the right of the laser rod.

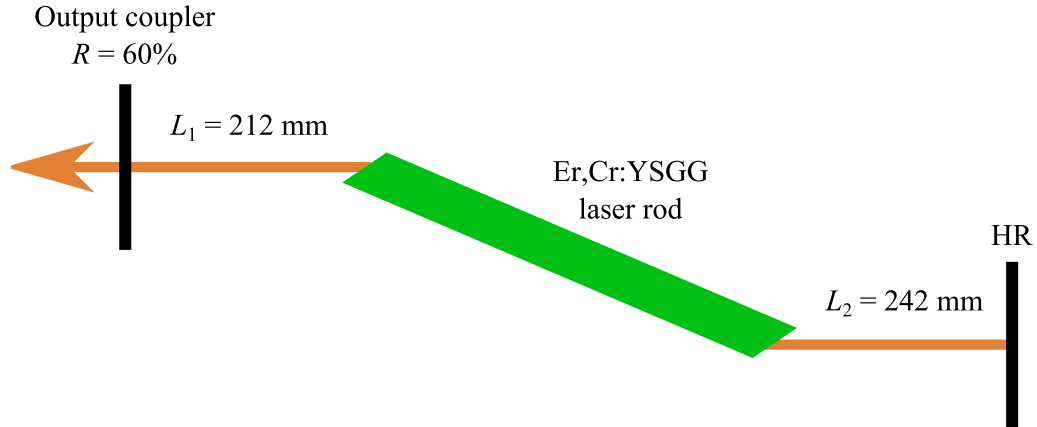


Figure 6.7.: Basic cavity set up, as determined in sec.5.6.2.1. Shown are the equivalent lengths of the cavity arms, L_1 and L_2 , from the centre of the rod (position of thermal lens approximation) and cavity mirrors. Actual distances from rod-end to cavity mirror are 190 and 220 mm, respectively. The laser rod is 9 mm long, and total cavity length is therefore 500 mm.

6.2.2. Laser pulse of free-running erbium

Before Q switching the erbium laser, it is important to look at the behaviour of the free-running pulse. Fig.6.8 shows the flashlamp pulse and its relation to the free-running laser pulse. Emission of the laser pulse begins as the flashlamp reaches its peak emission, around 150 μs after the beginning of flashlamp emission. The flashlamp pulse has a duration of around 400 μs . The free-running laser pulse lasts around 200 μs .

A closer look at the laser pulse is shown in Fig.6.9, as measured with a Molectron P3-01 pyroelectric detector. Evidently, the free-running laser emission is a pulse train consisting of ~ 20 individual pulses of ~ 400 ns pulsewidth, at 10 μs intervals.

This demonstrates the self-terminating nature of the transition when flashlamp-pumped [130]. A pulsetrain occurs because the long flashlamp pulse continues to replenish the upper laser level after the first laser pulse, but, because of the bottleneck at the lower laser level, re-absorption of photons by ground state electrons is delayed.

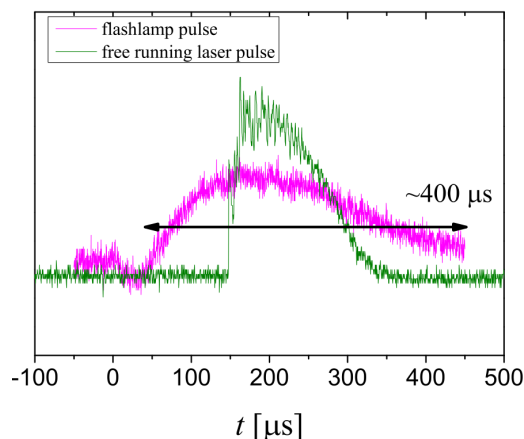


Figure 6.8.: Relationship between the flashlamp pulse and the free-running laser pulse. Flashlamp is triggered at $t = 0$. Rise time of the flashlamp pulse is $\sim 130 \mu\text{s}$, and has a $\sim 400 \mu\text{s}$ total duration (fixed for all input voltages). At approximately the flashlamp's peak emission does the laser pulse begins to emit, itself lasting for $\sim 200 \mu\text{s}$.

6.2.3. Fast Q switching of Er,Cr:YSGG

The system presented below is an EOQS Er,Cr:YSGG laser. Electrooptic Q switching of this gain medium (and of Er:YAG) is well documented and developed (sec. 4.4.1). Because the EOQS is close to the ideal Q switch, it can be used to ascertain the optimum build-up time for the system, and the optimum laser pulse emission time with respect to the flashlamp pulse.

6.2.3.1. EOQS system design

In our system, the Er,Cr:YSGG gain medium was Brewster-cut giving a linear polarisation to the laser mode, parallel to the optical bench (details in sec. 5.1). The LiNbO₃ Pockels cell, too, was Brewster-cut to further select this polarisation above other polarisations. Upon application of a high voltage ($\sim 1 \text{ kV}$) perpendicular to the Pockels cell, the Pockels cell became a half-wave plate, and polarisation was rotated to vertical with respect to the optical bench. It thus suffered loss upon leaving the Pockels cell, and again when re-entering the Pockels cell after reflection, with the losses preventing lasing. See Fig. 6.10.

To switch to low loss and permit lasing, a second voltage was applied in the opposite sense across the Pockels cell. The second voltage had a rise time of $< 100 \text{ ns}$ (see Fig. 6.11), and its magnitude, to cancel the electric field of the first voltage,

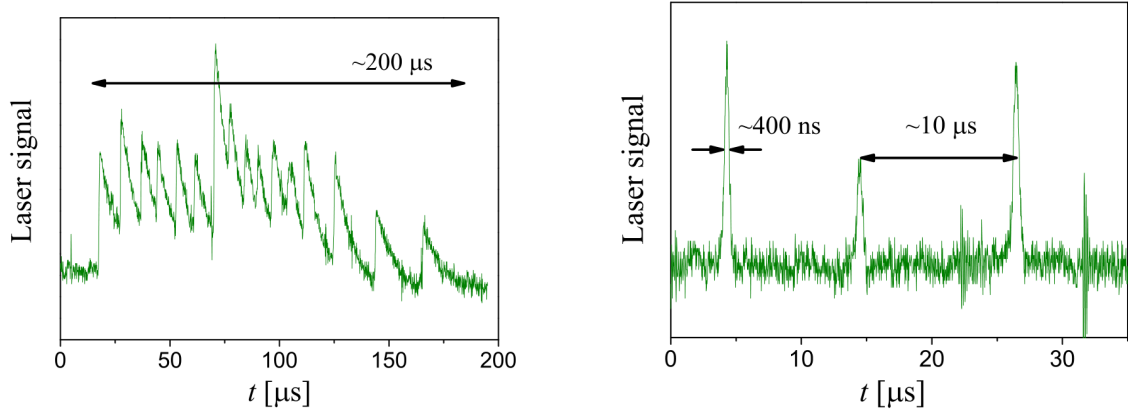


Figure 6.9.: Experimental pulse trace of the free-running, flashlamp-pumped Er,Cr:YSGG laser. A pulse train is emitted of ~ 20 pulses (left). The individual pulses of the pulse train are of ~ 400 ns duration, in ~ 10 μs intervals.

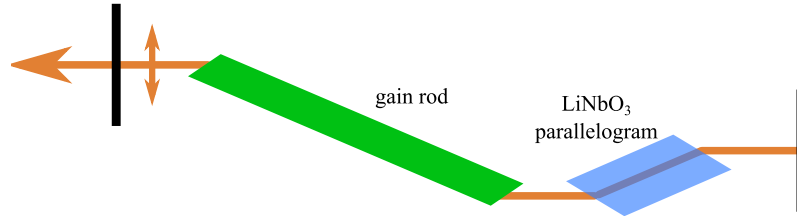


Figure 6.10.: Electrooptic, Brewster-cut, LiNbO₃, Pockels cell Q switch. With no voltage applied the laser mode travels through the Q switch, unperturbed. With the application of a high voltage the Pockels cell becomes a half-wave plate. The Brewster cut facets provide the loss mechanism to the rotated polarisation modes.

was ~ 1 kV. The second voltage, and therefore the low loss interval, was of ~ 3.5 μs duration, enough time to permit the formation and emission of the laser pulse.

Triggering of the system was with respect to the flashlamp pulse. The Q switch voltage pulse is applied $\lesssim 350$ μs after the flashlamp trigger, corresponding to laser pulse emission after the peak of the flashlamp pulse. See Fig. 6.11. After the near-instantaneous onset of low cavity loss, pulse build-up time was $t_d = 2.2$ μs , which can be taken as an intrinsic build-up time for this cavity, gain medium and ‘ideal’ Q switching. The time between flashlamp trigger and laser emission was $t_e \approx 350$ μs , which can be taken as an optimum time for this system, corresponding to emitting at the moment of critical population inversion.

Pulses emitted were of pulsewidth $t_p \approx 246$ ns, the precise value depending on the cavity length. The example pulse shown in Fig. 6.11 was for the cavity of total length 500 mm described at the beginning of this section. Output pulses were typically in

the range of ~ 10 mJ when pumping at 45 J, a wall-plug efficiency of $\sim 0.02\%$.

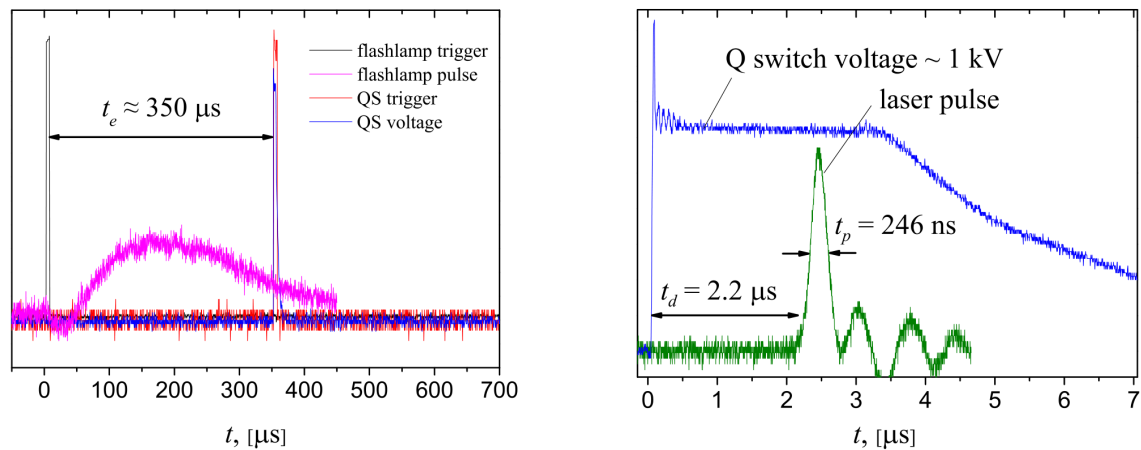


Figure 6.11.: (Left) Timing dynamics of the laser showing delay between flashlamp trigger, flashlamp pulse, and the Q switch trigger and Q switch voltage. (Right) The Q switch voltage and the emitted laser pulse, with timing aspects shown.

6.2.3.2. Laser damage of Pockels cell in EOQS

It was found that the LiNbO_3 Pockels cells suffered irreparable laser damage not too long after initial installation. The occurrence of this damage of the LiNbO_3 Pockels cell was corroborated by Skórczakowski *et al.*, working in the Er:YAG system [148], who cited high amounts of damage after $\sim 10^5$ pulses.

The cause of the damage appeared to be photoablation of the surface of the crystal, resulting in ‘pockmarks’ on the surface, and possible melting of the bulk, resulting in a clouding of the crystal’s usual transparency. The laser-induced damage threshold for LiNbO_3 is given in the literature, in terms of fluence, as $5 \times 10^4 \text{ J/cm}^2$ for $\lambda = 1.06 \mu\text{m}$ pulses of 10 ns pulsewidth [164]. In terms of intensity, damage threshold is cited as $\sim 5 - 30 \text{ MW/mm}^2$ for thermal damage, depending on where damage was seen in the crystal [165]. Given that, in our system, $\gtrsim 10$ mJ of energy was focussed into areas of $\sim 1 \text{ mm}^2$, in a duration of $\gtrsim 100 \text{ ns}$, the fluence in the cavity was $\sim 1 \text{ J/cm}^2$ and the intensity was 100 kW/mm^2 , which is lower than the above-mentioned thresholds. Though the fluence and intensity values of the laser will be greater at the spatial and temporal centre of the laser pulse, the values still fall somewhat short of the cited laser damage threshold for LiNbO_3 .

Other groups utilising the LiNbO_3 Pockels cell in high-fluence Er:YAG lasers have not reported the same laser damage [133, 166]. It is unknown whether high-quality

polishing of the crystal mitigated the effect of high intracavity fluence damage in those cases.

This weakness of the EOQS with LiNbO₃ Pockells cell was part of the motivation for investigating alternative Q switching methods for Er,Cr:YSGG, such as the slow Q switching methods detailed in this thesis.

6.3. Rotating mirror Q switching

In this section we investigate slow Q switching of the Er,Cr:YSGG laser, implementing the RMQS, only previously applied to the Er:YAG system [134] with regards to erbium.

6.3.1. RMQS laser setup

6.3.1.1. RMQS laser cavity configuration

Fig. 6.12 shows the configuration of the RMQS cavity, the rotating mirror replacing the back cavity mirror of Fig. 6.7. The rotating polygon mirror used in this case was a decagon, shown in Fig. 6.12, with all ten facets gold-plated and polished, and of area $20 \times 6 \text{ mm}^2$ (though only one facet was involved in Q switching at a time). The rotating polygon mirror was the model SOS-SA24C from Lincoln Laser, capable of rotating at 100-400 rps. It was powered with 24 V DC, drawing 4 A, and was controlled by the BMC 3.4 controller, also provided by Lincoln Laser, as well as some homemade electronics which also served as the interface with the flashlamp electronics.

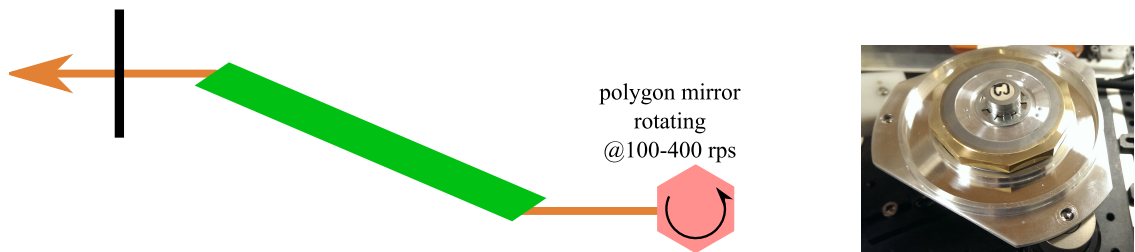


Figure 6.12.: (Left) Rotating polygon mirror Q switch, operating as per Fig. 6.4. The polygon in our system is a decagon. (Right) Photo of the gold-faceted, decagonal rotating polygon mirror used as the RMQS.

6.3.1.2. Control sequence of the RMQS

Fig. 6.13 shows the control flow chart for the RMQS. The sequence controlling the rotating mirror was initiated by a function generator, which set the rps of the mirror, f_{rps} . The mirror motor's electronics generated its own once-per-revolution signal (of identical frequency), which was then frequency-divided by homemade electronics to an appropriate pulse per second (pps) rate, f_{pps} , for the flashlamp (not exceeding ~ 12 pps). The once-per-revolution signal could be divided by powers of 2 (1 to 256):

$$f_{pps} = f_{rps}/2^n \quad (6.11)$$

where n is a selectable integer from 0 to 8. For example, with the mirror rotating at 400 rps, the flashlamp could have a pulse per second rate of $400 \div 64 = 6.25$ pps, or $400 \div 128 = 3.125$ pps, but not the values between³.

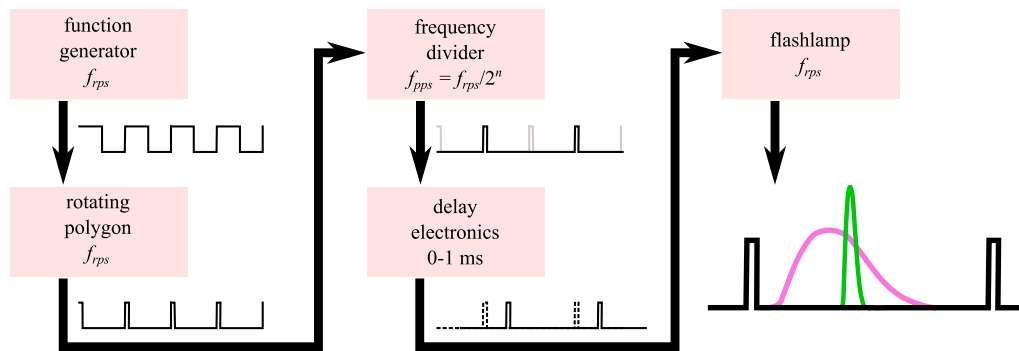


Figure 6.13.: RMQS control signal flow chart. A function generator outputs a TTL signal controlling the rps rate, f_{rps} , of the rotating mirror. The rotating mirror produces its own once-per-revolution signal which is fed to a frequency divider. The frequency divider is capable of dividing the signal by powers of 2 (1 to 256). The divided frequency, f_{pps} , is then sent to a delay box which outputs a signal with a selectable delay of 0-1 ms. The delayed signal then triggers the flashlamp. Fine-tuning of the delay allows for optimum timing of laser pulse output.

Before the frequency-divided pulse was sent to trigger the flashlamp, it passed through homemade delay electronics, capable of outputting a secondary signal with an arbitrary delay of 0-1 ms with respect to the input signal. This secondary signal

³'rps' and 'pps' are used as units instead of 'Hz' to avoid ambiguity between the two repetition rates described.

was used to trigger the flashlamp. The amount of delay was optimised for each mirror rps rate, and flashlamp power.

6.3.2. Optimising the RMQS mirror rotation rate

The rotation rate of the mirror had to be optimised to obtain the best Q switching time, t_Q , for the Er,Cr:YSGG system. Simultaneously, the number of pulses per second, and therefore the power in, , had to also satisfy the stability conditions of the cavity, as determined in sec. 5.6.2.1.

6.3.2.1. Pulse per second range of cavity

As shown in sec. 5.6.2.1, the cavity operates with an input power range of $P_{in} \approx 50 - 225$ W. For a laser pulse rate of, say, $f_{pps} \approx 3$ pps, this corresponds to input pulses of energy $E_{in} \approx 17 - 75$ J. Given that the system is limited to $E_{in} \leq 60$ J, pulse rate values of $f_{pps} \gtrsim 3$ pps were chosen.

6.3.2.2. Optimisation of mirror rotation rate

Fig. 6.14 shows the laser pulse energy output for four different mirror rotation rates: 100, 200, 300 and 400 rps, each lasing at 3.125 pps, except for the 300 rps case which lased at 4.69 pps. For each rotation rate the range of lasing fell within the stability zone of the cavity.

The best performing rotation rates was 300 rps followed by 200 rps, both with pulse energies approaching 10 mJ. Pulse energies could have reached higher levels, but there were fears over the damage of the gold rotating mirror facets, and so pulse energies were kept low. The output energies were comparable to those of the EOQS of sec. 6.2, with similar wall-plug efficiencies ($\sim 0.02\%$). The slope efficiency, η_{slope} , was best for the 300 rps data, in the region of $\eta_{slope} \approx 0.06\%$. Slope efficiency for 200 rps was $\eta_{slope} \approx 0.04\%$, and lower yet for 400 rps, at $\eta_{slope} \approx 0.015\%$. Slope efficiency for 100 rps was complicated by the prominent peak feature; before the peak, slope efficiency was $\eta_{slope} \approx 0.08\%$, and above 43 J it was $\eta_{slope} \approx 0.06\%$.

The peak feature of the 100 rps data showed a possible effect of the laser stability zones, with a peak at where the centre of the stability zone was, though similar peak features did not appear for the other three rps values studied. Alternatively, the

peak could possibly be explained by the rotation rate being at the optimum for the given pump energy, and the subsequent fall in energy being a result of breakthrough lasing. The equation describing laser pulse energy, E_{out} , for the slow Q switch (equation 6.10) states that the faster the mirror rotation speed, the higher E_{out} will be. However, 300 rps appears to be the optimum rotation rate, rather than the faster 400 rps.

This could be because of the oscillatory nature of the RMQS. The mirror does not remain at the angle of minimal loss indefinitely; if the rotation rate is too rapid then before the build-up time $t_d \approx 2.2 \mu s$ is completed, the Q switch could have passed in and out of the minimum loss value, and thus the pulse will emit during non-minimal cavity loss. This situation is described diagrammatically in Fig. 6.6 c).

An alternative explanation for why 300 rps yields the best output pulse energies, rather than 400 rps, for this decagonal mirror is investigated in the next section, sec. 6.3.3.

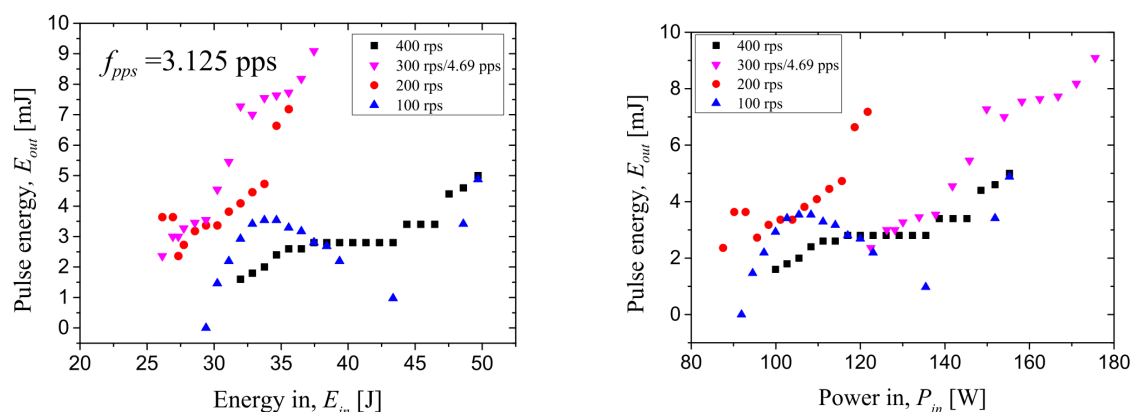


Figure 6.14.: (Left) Energy in versus energy out for the laser at various mirror rotation rates, all with flashlamp repetition rate of ~ 3 pps. The 300 rps rotation rate is the best tested here; this rotation rate was at a slightly higher flashlamp pulse repetition rate (4.69 pps). (Right) The same data plotted against power in, P_{in} , to demonstrate the extent of the stability zones of the laser.

6.3.3. Properties of pulse emission from the RMQS laser

6.3.3.1. Pulse emission timings for the RMQS laser

The switching time of the slow Q switch is of the order of 1-10 μs , depending on mirror speed and cavity length. This Q switching time is similar to the build up

time of the laser pulse ($t_d \approx 2.2 \mu\text{s}$). But, the Q switching time is quick with respect to the time taken for the system to reach maximum population inversion ($\sim 150 \mu\text{s}$). Therefore, compared to the flashlamp pulse, the Q switch is virtually a step function.

It was found that the optimal time between flashlamp trigger and laser emission, t_e , varied with mirror rps rate. For the 300 rps and 200 rps cases, it was found that $t_e \approx 350 \mu\text{s}$, which was much the same as for the EOQS of sec. 6.2 - this can be taken as the optimum value of t_e , with the pulse emitting at the moment of optimum population inversion density. See Fig. 6.15.

Decagon limits population inversion

On the other hand, for 400 rps, the emission time was $t_e \approx 300 \mu\text{s}$. See Fig. 6.15. To understand why this is the case, we look at the fact that the polygon mirror is a decagon. Rotating at 400 rps means there are $250 \mu\text{s}$ between the intervals of low loss. Thus, if the flashlamp is triggered for the optimum time for the EOQS, such that the pulse emits at $t_e \approx 350 \mu\text{s}$, then $100 \mu\text{s}$ after the trigger, the cavity will have an interval of low loss. This coincides with the rising edge of the flashlamp pulse. We have seen that for the free-running laser, the pulse emits at $t_e \approx 120 \mu\text{s}$, so this instance of low loss will deplete the growing population inversion. Therefore the optimum pulse emission is $\sim 50 \mu\text{s}$ earlier such that the early low-loss interval does not occur when the population inversion is high. This then helps to explain why 400 rps is not the highest-performing mirror rotation speed.

To solve this, only one facet of the polygon should be made reflective, and the rest of low reflectivity. Commercial systems often use flat mirrors of minimum possible radius, rather than polygon mirrors.

6.3.3.2. Pulseswidth properties for the RMQS laser

Shown in Fig. 6.16 is the laser pulsetrace of the 300 rps case with $E_{in} = 45 \text{ J}$ pumping energy. The pulseswidth is $t_p = 176 \text{ ns}$. It was found that pulseswidth was dependent on the pulse energy of the given pulse, and ranged from $\sim 126\text{--}375 \text{ ns}$ across the laser pulse energy range. This is mostly because the population inversion is depoulated quicker when there is more intracavity energy, as described by equation 6.8.

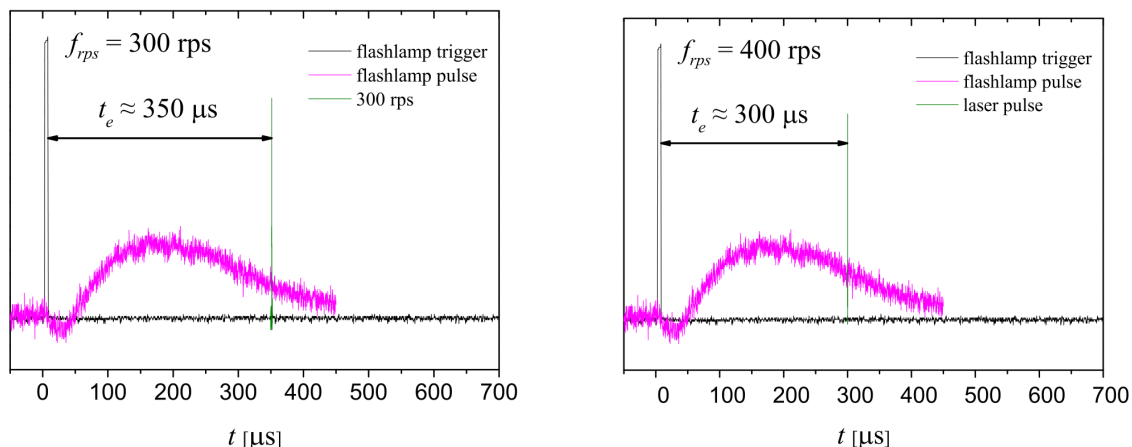


Figure 6.15.: Composite pulse trace demonstrating timing of flashlamp trigger, flashlamp light pulse, and laser pulse of RMQS laser, for 300 rps (left) and 400 rps (right). Note that the time t_e is longer for the 300 rps case, leading to high energy output.

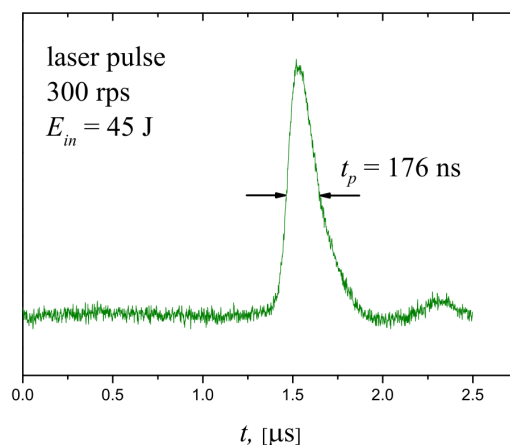


Figure 6.16.: Pulsetrace of the laser pulse of the RMQS laser.

6.4. Polygon chopper Q switching

An alternate method of Q switching the Er,Cr:YSGG laser was developed, which constituted, to the best of the author's knowledge, the first of its kind for the $\lambda \simeq 3 \mu\text{m}$ erbium system.

6.4.1. Using rotating polygon as chopper

The same rotating polygon of sec. 6.3 was repurposed to become an optical chopper, the function of which is shown diagrammatically in Fig. 6.17. An optical chopper Q

switch with a polygon chopper is herein referred to as a polygon chopper Q switch (PCQS).

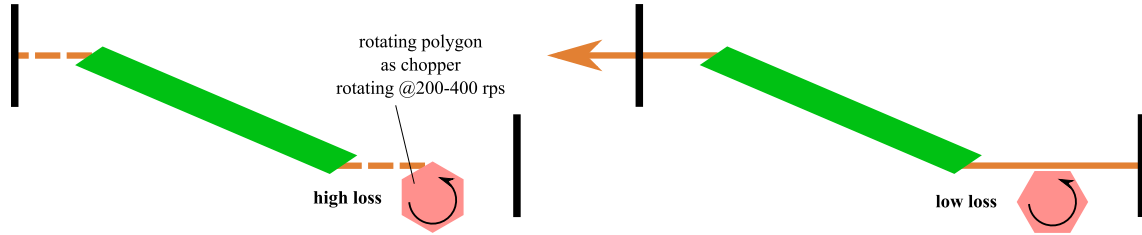


Figure 6.17.: Polygon chopper Q switch. A spinning polygon acts as a chopper for the optical mode. For low loss the polygon edge is parallel and not obstructing the laser mode. When the vertices of the polygon block the laser mode (even partially) the cavity has high loss.

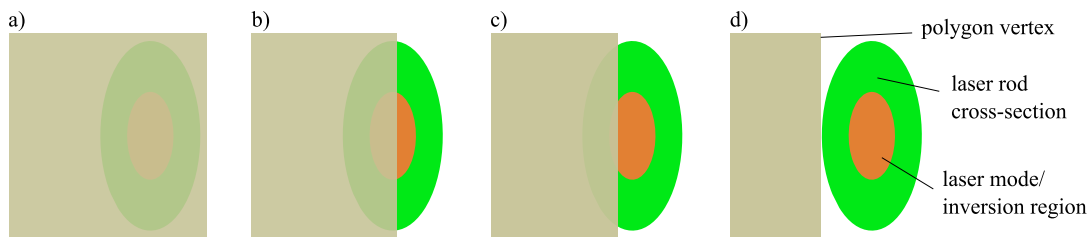


Figure 6.18.: Cross-sectional view of laser rod (green) and cavity mode (orange) as the polygon vertex (grey) makes the transition from obscuring the mode and inducing high cavity loss (a), to not obscuring the mode at all, giving low loss (d). In between these two points is (b) where the polygon obscures enough of the laser mode to prevent lasing, and (c) where the polygon obscures part of the laser mode, but smaller, unwanted laser modes may lase.

The polygon is positioned with an edge parallel to the laser mode, and adjacent to, but not obscuring, the laser mode. Then, when the polygon rotates, the vertices move in and out of the laser mode path, and the polygon thus acts as an optical chopper.

Fig. 6.18 shows the path of the polygon vertex as it moves across the cross-section of the laser rod, and obscures the laser mode. When fully obscuring the mode, the cavity loss, $\gamma(t)$, is effectively infinite. Even with the polygon obscuring just half of the intracavity laser mode's path, the loss will remain too high for lasing to occur. As the centre of the cavity becomes unobscured, small-radius modes will begin to build up in photon density, and these smaller modes may breakthrough and lase before the full-sized mode is fully unobscured. This will deplete population

inversion from the main laser mode. Therefore, the polygon vertex must be clear of the laser mode in as short a time as possible for effective Q switching.

For a decagon, as used here, with an outer radius of 33 mm, the outer extent of the decagon traces a path, back and forth, of ~ 1.6 mm. The outer extent is thus able to oscillate between obscuring and permitting the intracavity mode (horizontal radius $\lesssim 1$ mm). A photo of the PCQS cavity is shown in Fig. A.2, page 212.

6.4.2. Q switching time of the PCQS

In the process of designing the PCQS laser, it was unknown if the Q switching time, t_Q , would be short enough. Though the PCQS is a slow Q switch by nature, there is still a limit to *how* slow it can be and still successfully Q switch the laser.

To calculate the Q switching time of the PCQS, we evaluate the speed with which the chopper is opened. With the polygon rotating at its maximum speed, 400 rps, the chopper shutter will be opened and closed at a rate of 4000 times per second, i.e. opened once and closed again in $250\text{ }\mu\text{s}$. So an upper estimate for Q switching time is $t_Q = 125\text{ }\mu\text{s}$, for the 400 rps case, which is considerably slower than the RMQS switching time ($t_Q < 10\text{ }\mu\text{s}$).

A better estimate for the Q switching time comes from substituting the laser mode for an external laser, and measuring how quickly the external laser signal changes from minimum to maximum value. A HeNe laser was used here, with its beam waist matched to that of the erbium laser mode, with a silicon detector positioned on the opposite side of the polygon.

Fig. 6.19 shows the (inverted) signal of the HeNe laser beam whilst passing close by the polygon, with the polygon rotating at 400 rps. The signal periodically oscillates between maximum (high loss) and minimum (low loss), with a period of $250\text{ }\mu\text{s}$. The transition from high to low loss is a truer estimate of the Q switching time, and this gives us $t_Q = 30\text{ }\mu\text{s}$.

6.4.3. Optimal positional alignment of PCQS

The lateral position of the PCQS in the cavity is critical for successful Q switching. Fig. 6.20 shows pulsetraces of the laser as the PCQS is moved from a position

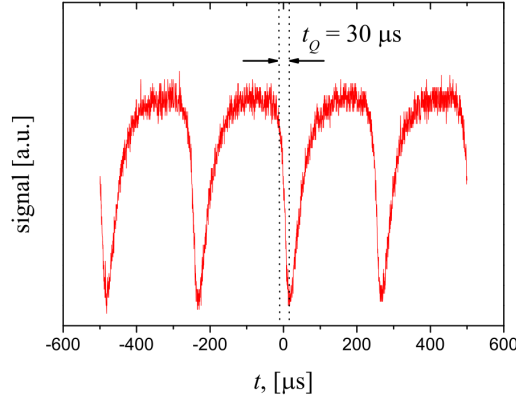


Figure 6.19.: Lateral position of polygon chopper vertex as measured with HeNe beam. Peaks correspond to the polygon chopper fully obscuring the beam, and troughs correspond to the beam unobscured. The time it takes to transition from peak to trough provides an estimation of the Q switching time of $t_Q = 30 \mu\text{s}$. PCQS is rotating at 400 rps.

not obscuring the laser mode at all, to an intermediate position where it partially obscures the beam, and finally to a position where full Q switching occurs.

Note from Fig. 6.20 that in an intermediate position, the PCQS causes emission of two separate pulses, the second corresponding to emission during the subsequent low-loss interval, $250 \mu\text{s}$ later (a one-tenth rotation of the decagon, rotating at 400 rps). This signifies that not all the energy is expended in the initial pulse. Also in the intermediate PCQS position, it is possible to see that there is a greater delay to the laser pulse after the flashlamp trigger, showing that the emission is delayed by the rotating polygon, but not quite by enough to cause Q switching. For the final, optimised position, a single pulse is emitted.

On occasion, if the PCQS was not optimally positioned, the period of low loss was too long and a short pulse train of 2 or 3 pulses could occur.

6.4.4. Optimal polygon rotation rates and energy performance

Until this point, only the 400 rps PCQS speed has been considered. At lower speeds (350 rps and below), Q switching did still occur, but with the occurrence of breakthrough pulses, as shown in Fig. 6.21. Lasing began with a smaller pulse, emitted whilst the loss was not yet at minimum, followed by a second, stronger pulse, coinciding with the minimum cavity loss. This is the scenario shown in Fig. 6.6 b).

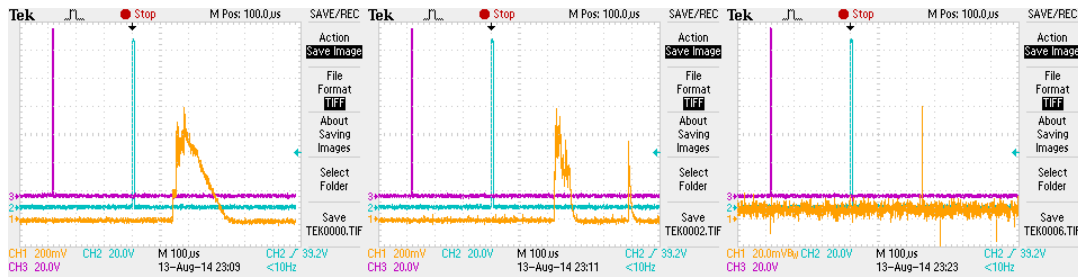


Figure 6.20.: Change of the laser pulse as the Q switch is progressively moved into position, from left to right. The purple line is the polygon trigger, the blue is the flashlamp trigger, and the orange is the laser signal measured on the Molectron P3-01 detector.

This indicates that the Q switching time of the 400 rps polygon, $t_Q = 30 \mu\text{s}$, is the limiting slow Q switch time for the Er,Cr:YSGG system. Any Q switching times longer than this will lead to breakthrough and multiple pulse emission. This is believed to be true of any Q switching method applied to Er,Cr:YSGG.

At 150 rps, reliable Q switching of the laser ceased to occur completely, corresponding to a Q switching time of $t_Q \approx 80 \mu\text{s}$. Again, this is thought to be universally true of Q switching Er,Cr:YSGG.



Figure 6.21.: Breakthrough pulse preceding larger pulse when PCQS rotating at 350 rps and below (200 rps case shown here).

Fig. 6.22 shows the energy in versus energy out for the PCQS laser, with the polygon rotating at various rates of revolution. The best performing rate was 400 rps, giving maximum output of $\sim 3.8 \text{ mJ}$ at 60 J input, a 0.006% wall-plug efficiency. The slope efficiency for 400 rps was $\eta_{\text{slope}} = 0.014\%$, which was also true for the 350 rps case. We remind ourselves that the energy output for rates of revolution lower than 400 rps are not single pulses, so are not fully comparable with the 400 rps case.

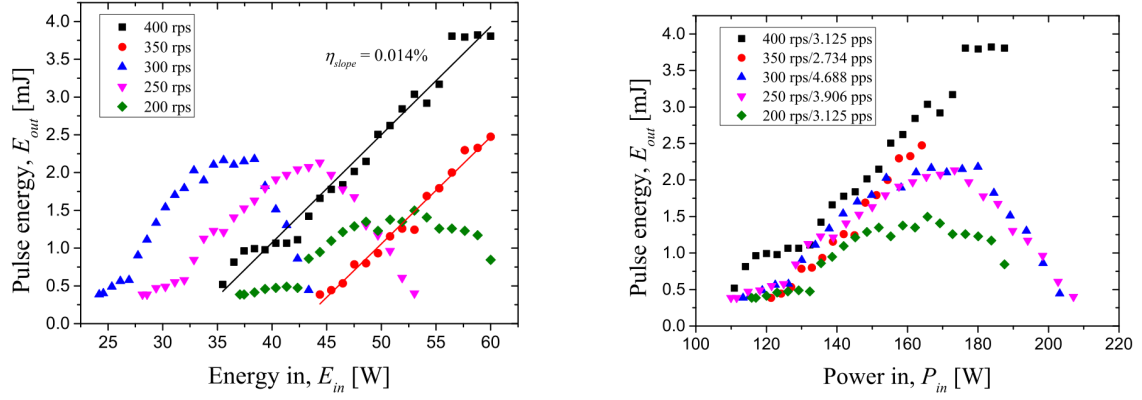


Figure 6.22.: (Left) Energy in versus energy out for 4 different PCQS rps rates, with two slope efficiencies of the same value shown. (Right) The same data, but plotted against power in, P_{in} , demonstrating the thermal lens stability zone effects.

This slope efficiency and maximum energy is lower than the $\sim 0.06\%$ and ~ 10 mJ achieved with the RMQS. This is a consequence of the slower Q switching time of the PCQS, and the fact that the polygon is specifically a decagon - see next section (sec. 6.4.5) for more details.

Fig. 6.22 also shows the power in versus energy out, showing the edges of the thermal lens stability region of sec. 5.6.2.1, Fig. 5.10. The consequence of the stability zones is that, pumping higher than $E_{in} = 60$ J will not gain much more output energy, since the laser will leave the stability region. This could be remedied by halving the lasing rate from 3.125 to 1.5625 pps, and increasing the input flashlamp energy, E_{in} , beyond the capability of the current system.

6.4.5. Laser pulse properties and emission timing with PCQS

Fig. 6.23 shows the relationship between the flashlamp trigger, flashlamp pulse, laser pulse, and position of the polygon vertex for the 400 rps rotation rate. The laser pulse emission coincides with the chopper being fully retracted from the path of the cavity mode.

The laser pulse is emitted about $t_e = 300$ μ s after the flashlamp trigger, resulting in less energetic pulses than seen in the EOQS and RMQS. This is because the optimum emission time is $t_e \approx 350$ μ s for Er,Cr:YSGG Q switching. This earlier emission time is as with the RMQS at 400 rps (see sec. 6.3.3), and the same explanation applies as to why.

The $t_e \approx 300 \mu\text{s}$ delay is longer than the repetition period of the loss ($250 \mu\text{s}$), and so if $t_e = 350 \mu\text{s}$ instead then there will be a low-loss interval during the rising edge of the flashlamp, which will spoil population inversion growth. Thus, the optimum emission time is reduced to $300 \mu\text{s}$ to avoid this, but at the cost of not emitting at the optimum moment with respect to population inversion.

Lower rotation rates will not circumvent this problem, because, as we have seen, dual pulses will occur if the PCQS is slowed down; a rotation speed of 400 rps is necessary to guarantee single pulse emission. Thus the PCQS emits lower-energy pulses.

Also shown in Fig. 6.23 is the trace of the pulse, with pulsewidth of $t_p = 305 \text{ ns}$ for the 400 rps case. This is comparable to the RMQS, but slightly longer, possibly because of the slower Q switching time and lower pulse energies.

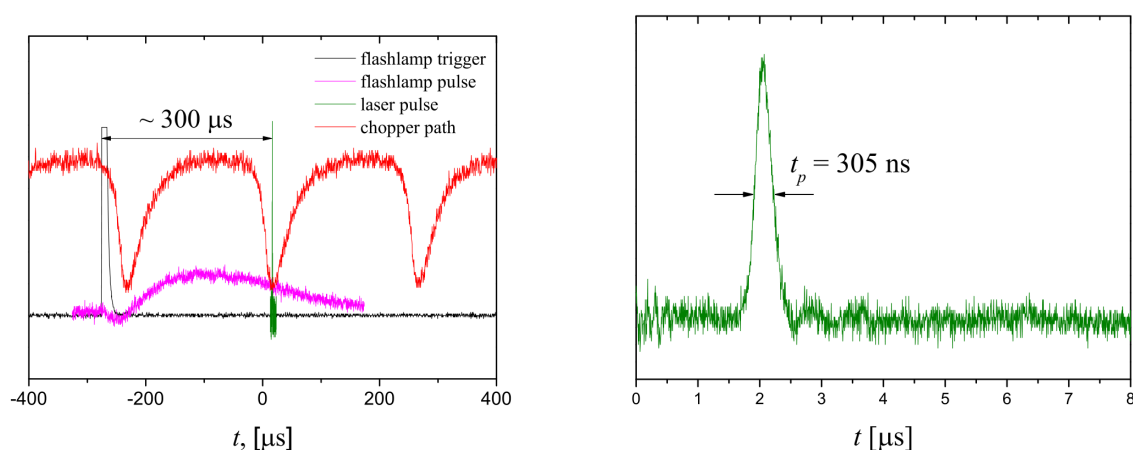


Figure 6.23.: (Left) Timing of PCQS and flashlamp emission, and laser pulse emission. (Right) The pulse trace of Q switched pulse for 400 rps PCQS.

6.5. Improving performance of PCQS with nitrogen purging

6.5.1. Energy and efficiency improvement of PCQS laser

The strong absorption of $\lambda \approx 3 \mu\text{m}$ radiation by water provides the erbium laser one of its most prevalent uses, in medicine. However, this same water absorption can

hinder the efficiency of the erbium laser, due to the water vapour present within the laser cavity constituting a loss mechanism.

To this end, we encased the erbium laser in an air-tight box, which could be purged of its atmosphere with gaseous N_2 . See Fig. 6.24 and Fig. A.3. The dimensions of the box were $65 \times 35 \times 25$ cm. Time taken to fully purge was 3 minutes, and the purge held for an hour at a time, or longer if a small positive pressure of N_2 was maintained. A BaF_2 window was inserted for the laser beam output.

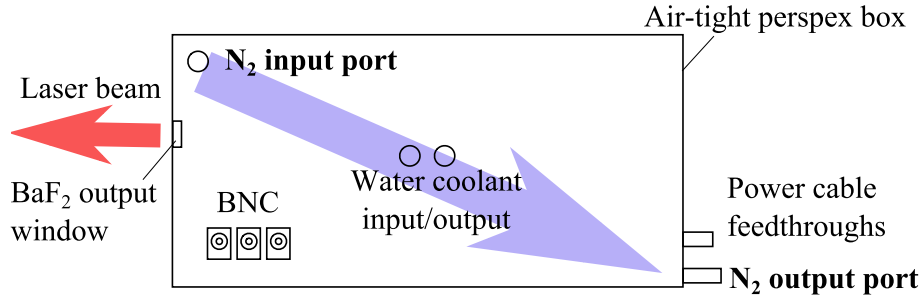


Figure 6.24.: Schematic of purging box, with the laser within (not shown). Purple arrow shows the direction of the N_2 through the box, as it purges the atmosphere within. The laser beam exits the box through a BaF_2 window. Other bulkheads were fitted to allow BNC cables, water coolant, and power cables to enter and exit the box.

Our results compare the PCQS laser energy output for the laser box unpurged and purged, shown in Fig. 6.25. Purging increased the slope efficiency to $\eta_{slope} \approx 0.016\%$; the maximum energy output achieved was $E_{out} = 4.9$ mJ, a 29% increase compared to before purging. Prior to purging in the presented case, the atmosphere had a 41% relative humidity, as monitored with an ATP hand-held humidity purging. After purging, relative humidity was $< 0.1\%$. The exact amount of improvement from purging depended on the ambient humidity before purging; on a dry day (relative humidity $< 20\%$) purging caused less of an improvement to the output energy, if any at all.

Previous work by Livingston *et al.* [132], working with the Er:YAG laser, detailed the removal of water vapour with a dehumidifying air circuit, and found that output laser pulse energy was doubled. This is a greater improvement compared to what was found here - though it was undisclosed what the humidity of their unpurged atmosphere was, so direct comparison is difficult.

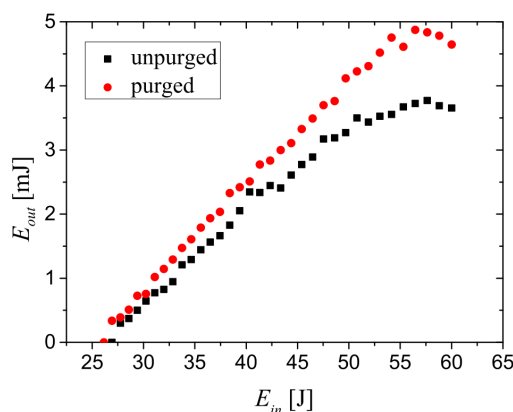


Figure 6.25.: Output laser pulse energy versus energy in, with and without N_2 purging, of the 400 rps PCQS laser. N_2 purging increases the slope efficiency 0.016% such that maximum energy increases by some 29%. When unpurged, relative humidity = 41%.

6.5.2. Additional benefits of purging

Besides improvement in efficiency and output energy, purging the laser also reduced the likelihood of laser-induced damage of optical components. The purge box helped keep the environment of the laser free of dust; impurities such as dust constitute heating-centres around which optical damage can nucleate. Furthermore, a low-oxygen atmosphere benefits the optical components; optical coatings are susceptible to oxygen absorption which form oxides which change the optical properties, and constitute impurities which would expedite laser damage.

Finally, the perspex box, opaque to midinfrared radiation, improves the safety of the laser in the lab setting.

6.6. Diode-pumped, bounce geometry Er:YSGG laser

6.6.1. Design of bounce-geometry laser

Simultaneous to the development of the flashlamp-pumped system detailed here, collaborators in the Photonics Group here at Imperial College (Emma Arbabzadah, under the direction of Mike Damzen) developed a diode-pumped version of the system [115], planned as an eventual successor to the flashlamp-pumped system. There, the laser was in ‘bounce-geometry’, where the pumping takes place at the

edge of the gain medium, and the laser mode completes a reflection from the pumped edge during its traversal of the cavity. This is shown in Fig. 6.26 where the gain medium is also Brewster-cut giving the laser gain a trapezoidal shape. Diode banks of $\lambda = 966$ nm were used, pumping the $^4I_{11/2}$ level, the upper laser level. The laser in free-running mode constituted the highest-pulse energies of a diode-pumped Er:YSGG system to date (see sec. 4.4.2), of ~ 55 mJ pulse energy.

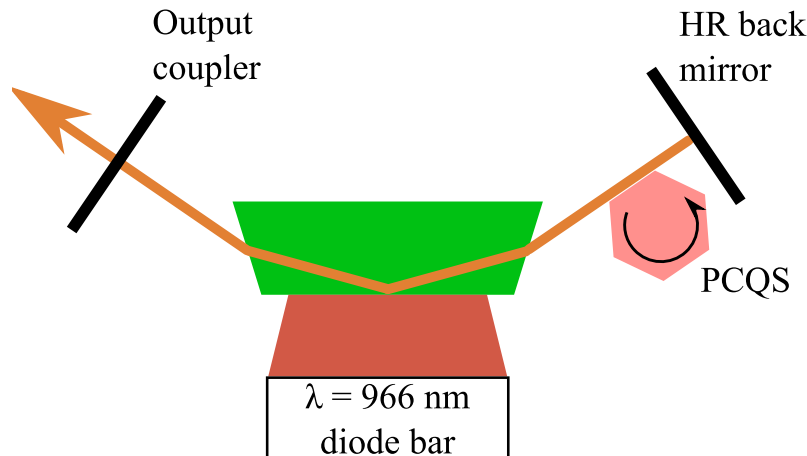


Figure 6.26.: Schematic of the diode-pumped, bounce-geometry laser, with PCQS.

6.6.2. Diode-pumped Q switched laser output energy

The system was augmented to include a Q switch. All three of the mentioned Q switching schemes were attempted: EOQS [141], RMQS and PCQS. Q switched pulse energies in all Q switch methods were < 0.5 mJ, with repetition rate of up to 200 pps in the case of the PCQS method. By comparison, the flashlamp-pumped system, outputting 76 mJ pulses, had Q switched pulses of ~ 10 mJ, with repetition rate of up to 12 pps.

It is possible that direct-pumping of higher energy levels, (e.g. the $^4I_{9/2}$ level, at a diode wavelength of ~ 790 nm) would improve laser output pulse energies, by being a closer emulation of the flashlamp-pumped case. Investigations into increasing the diode-pumped Q switching power are on-going. The diode-pumped system is desired due to the prospect of increased efficiency, and reduced thermal issues allowing higher repetition rate, and more compact cavities.

6.7. Comparison of Q switching methods

Tab. 6.1 contains a comparison of the various Q switch methods presented in this thesis. The hypothetical optimum Q switched erbium laser would take its output pulse properties from each of the different systems, e.g. the output pulse energy of the flashlamp-pumped EOQS system, and the repetition rate of the diode-pumped PCQS system. However, transverse fundamental mode lasing is difficult to achieve in non-cylindrical, diode-pumped lasers, so the method of Q switching, and type of gain medium should be chosen specific to the application of the laser.

Future development of the PCQS could possibly result in a high pulse energy system, benefitting from having no insertion losses and less susceptibility to laser damage of cavity optical components. There is also much to explore in the diode-pumped system, which will develop alongside and possibly eventually supercede the flashlamp-pumped system.

Table 6.1.: Comparison between different methods of Q switching presented in this thesis.

	EOQS	RMQS	PCQS	D-P, PCQS
Maximum E_{out} [mJ]	>10	~10	~4	0.55
Minimum pulsewidth, t_p [ns]	~246	126-375	~305	154
Max. repetition rate, f_{pps} [pps]	~12	~12	~12	200

Conclusion to Part II

The flashlamp-pumped, $\lambda \simeq 3 \mu\text{m}$ Er,Cr:YSGG laser has been investigated, firstly in terms of thermal lensing properties, and secondly in terms of Q switching. The overall aim was to create a high-energy, Q switched, fundamental mode (TEM_{00}) laser.

The thermal lens strength of the Er,Cr:YSGG gain rod was measured by varying pulse repetition rate and cavity length, with the boundaries of the laser's stability zones indicating the strength of the thermal lens. The thermal lens focal length was found to vary between $f \approx 5$ to 25 m, a stronger thermal lens than in equivalently-sized Er:YAG rods. The difference was attributed to the additional thermal absorption from the Cr^{3+} doping. Fundamental mode lasing was achieved by increasing the size of the fundamental mode within the gain rod to better match the cross-section of the population-inverted atoms. The fundamental mode radius was increased by lengthening the cavity, and in this mode pulse energies of up to 76 mJ could be achieved.

Next, Q switching of the Er,Cr:YSGG laser was investigated. Two mechanical, slow Q switches were implemented and characterised. The first was a rotating mirror Q switch. This yielded laser pulse energies of ~ 10 mJ, with pulsewidths of 176-375 ns, and slope efficiencies up to 0.06% when at the optimum mirror rotation rate, 300 rps. When the rotation rate was faster (400 rps), the low-loss repetition period of the rotating mirror was too short for the population inversion growth rate, resulting in loss of some population inversion before optimum population inversion.

The second Q switch implemented was based on the optical chopper Q switch concept. Here, the vertex of a rotating polygon acted as a chopper, as it passed into and out of the path of the intracavity mode; this was called a polygon chopper Q switch. This is a first implementation of any optical chopper Q switch in an erbium laser. With a Q switching time of $t_Q = 30 \mu\text{s}$, single pulse operation was achieved, with energies of up to 3.8 mJ, and pulsewidths of ~ 300 ns, and a slope efficiency of $\sim 0.014\%$.

When the Q switching time was longer than $t_Q = 30 \mu\text{s}$, breakthrough pulses were detected. Thus, this Q switching time constitutes the limit of well-behaved slow Q switching of Er,Cr:YSGG by *any* method. No Q switching whatsoever occurred for Q switching times of above $t_Q \approx 80 \mu\text{s}$, which represents the limit of slow Q switching Er,Cr:YSGG.

Nitrogen purging of the laser, which eliminated the loss-inducing water vapour from the cavity, was shown to increase the output pulse energy by $\sim 29\%$, the exact improvement depending on the ambient humidity before purging.

Future development of the PCQS laser could lead to a reliable, long-lifetime erbium medical laser. It could also be implemented in a midinfrared-wavelength-generating optical parametric oscillator for high intensity midinfrared radiation. Diode-pumped systems are capable of higher pulse repetition rates, but fundamental mode lasing is difficult to achieve. The flashlamp-pumped system, on the other hand, benefits from having cylindrical laser rods, allowing fundamental mode lasing to be more easily achievable. However, it is expected that, with continued development, diode-pumped systems will eventually supercede flashlamp-pumped systems, simply because of the superior efficiency and simpler thermal management issues.

What is certain, however, is the growing importance of the erbium laser in the field of medical surgery and dentistry, justifying continued research into this laser system.

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A. Appendix

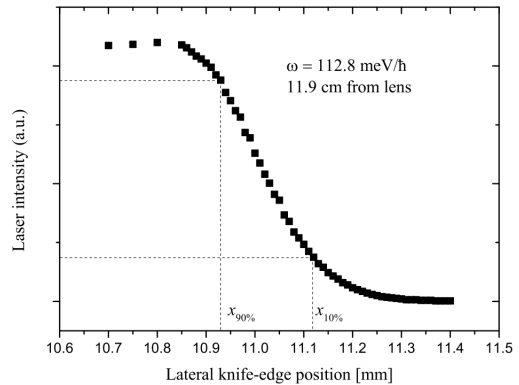
A.1. Knife edge beam radius measurement

The properties of the focussed beam of the MIRcat laser, and of the Er,Cr:YSGG laser were measured with the knife-edge method. In this method, a razor blade incrementally obscures the path of the beam whilst laser intensity at the detector is recorded against razor blade position. The resulting curve is that of the error function (see Fig. A.1) and the Gaussian profile of the beam can be extracted by fitting to this curve. If just the beam width is required, one can use the relationship between the error function and the Gaussian profile [80], and by measuring, for example, the position of 10% beam transmission ($x_{10\%}$) and 90% beam transmission ($x_{90\%}$), obtain the beam radius at a given point with

$$w(z) = \frac{x_{90\%} - x_{10\%}}{1.28}, \quad (\text{A.1})$$

where, for this equation only, z is the optical beam direction (rather than the sample growth direction).

Figure A.1.: Knife-edge measurement of the focussed MIRcat laser beam, showing error function curve of laser intensity with razor blade position.



A.2. Photos for Part II

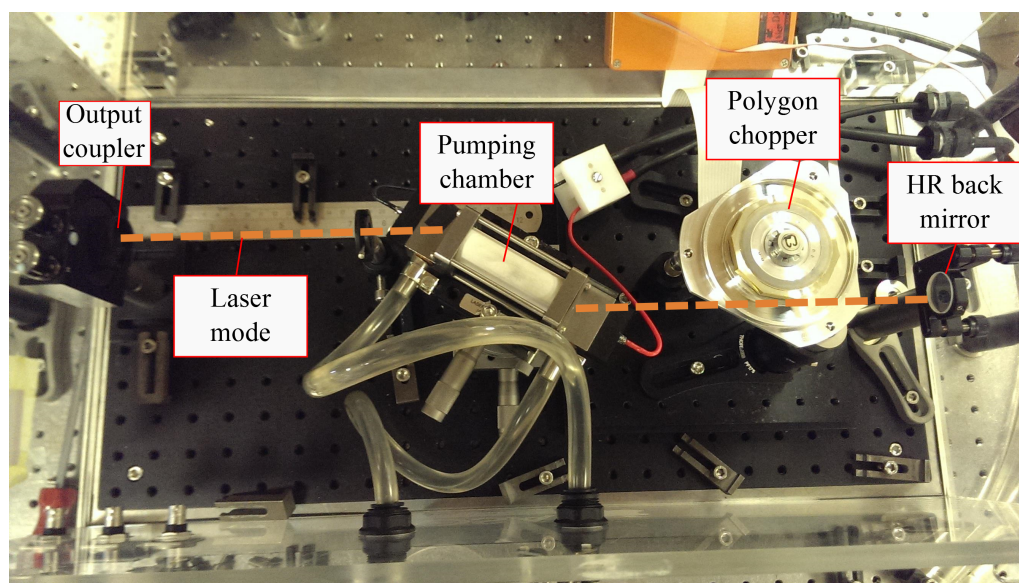


Figure A.2.: Photo of PCQS laser cavity of sec. 6.4.

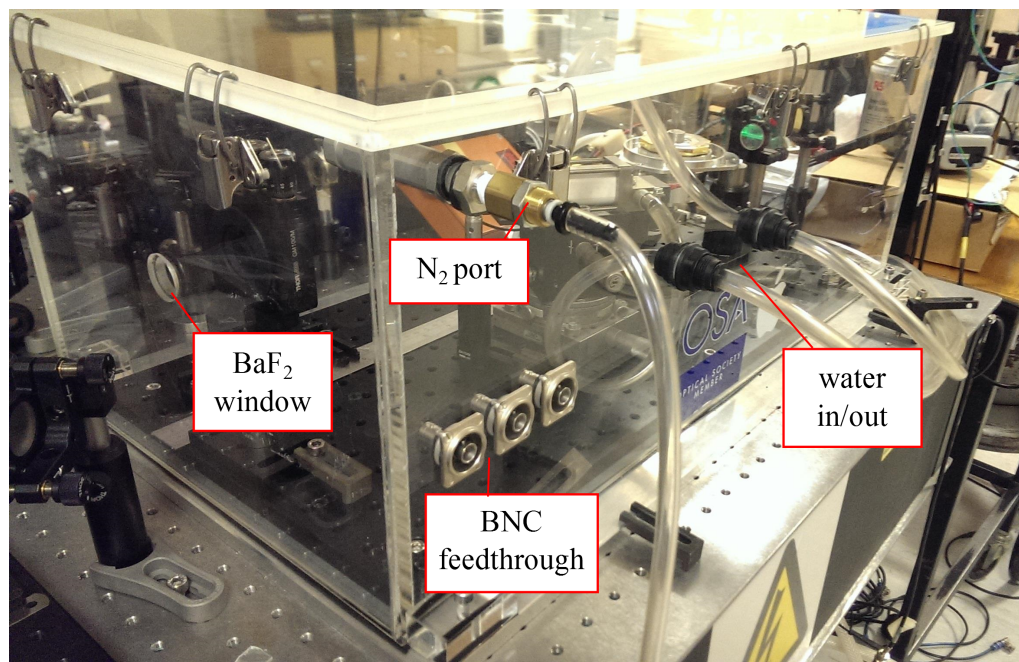


Figure A.3.: Photo of N₂ purging box exterior, as described in sec. 6.5.

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September 9, 2014

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- Fig. 1, Phys. Rev. B 55, 9731 – Published 15 April 1997

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Daniel Hofstetter, Jérôme Faist

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A.4. Draft of paper to be published for Part II

High repetition rate diode-pumped, mechanically Q-switched Er:YSGG lasers at 3 μ m

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Abstract: In this work, high repetition rate Q-switching of diode-pumped Er:YSGG lasers is presented. Pulsed operation is achieved using a rotating polygon mirror as a mechanical Q-switch which is employed in two configurations; as a rotating back mirror or as an intra-cavity chopper of the laser mode. Pulse energies >0.5mJ at 100Hz and operation at repetition rates of 200Hz are obtained at a laser wavelength of 2.797 μ m. Spectroscopy of the characteristic green fluorescence from a diode pumped 50at.% Er:YSGG laser crystal is also examined as a pre-cursor to the laser work.

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1. Introduction

Pulsed erbium-doped lasers operating at mid-IR wavelengths close to 3 μ m are useful in a range of medical and biological applications owing to strong absorption by water in this region [1]. The majority of research and commercial development on the erbium laser has focused on lamp pumped systems with high pulse energy, but which are usually limited to low repetition rates due to detrimental thermal effects caused by the inefficiency of the pump scheme [2]. Although the higher efficiency offered by diode pumping offers a potential solution to this problem, very few studies have been undertaken into Q-switched diode pumped erbium-doped lasers at the 3 μ m transition [3-5]. In this work we present high repetition rate mechanical Q-switching of diode-pumped Er:YSGG lasers utilising the bounce amplifier geometry, with pulse energies >0.5mJ at 100Hz and operation at repetition rates of 200Hz at a laser wavelength of 2.797 μ m. As a pre-cursor to this, spectroscopy of the fluorescence from a diode pumped Er:YSGG crystal is presented in Section 2 alongside a description of the 3 μ m transition in erbium-doped lasers.

2. Spectroscopy of Er:YSGG

Figure 1 is a simplified Er^{3+} energy level diagram showing diode pumping at $\sim 970\text{nm}$, lasing on the $3\mu\text{m}$ transition, important energy transfer processes in the system and the transitions which produce the fluorescence investigated in this section (curved lines). In the diagram, level 0 is the ground state ($^4I_{15/2}$) and the excited states are numbered as levels 1 to 6. The $3\mu\text{m}$ lasing transition occurs between level 2 ($^4I_{11/2}$) and level 1 ($^4I_{13/2}$) and diode pumping at a wavelength $\sim 970\text{nm}$ (bold red arrow) produces direct excitation from the ground state to the $3\mu\text{m}$ upper laser level (level 2).

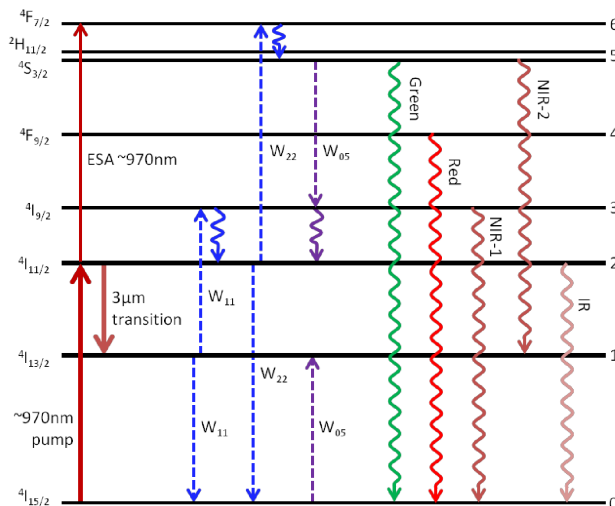


Fig. 1. Simplified energy level diagram for the trivalent erbium ion. The $3\mu\text{m}$ transition occurs between the energy levels shown in bold. The blue dashed arrows represent the ETU processes W_{11} and W_{22} and the green dashed arrow illustrates the cross relaxation W_{05} .

A peculiarity of the $3\mu\text{m}$ transition of the trivalent erbium ion is that the lower laser level lifetime is longer than the upper laser level lifetime, which should cause erbium-doped lasers to operate only in a pulsed, self-terminating mode [6]. However, long pulse and continuous wave (CW) operation is attainable due primarily to the complex non-radiative energy transfer processes which especially take place in highly doped erbium lasers are shown in figure 1 as blue and purple dashed lines with rate coefficients W_{11} , W_{22} and W_{05} . Two of these, W_{11} and W_{22} , are energy transfer upconversion (ETU) processes, while W_{05} is a cross relaxation process. The first ETU process, W_{11} , involves two erbium ions in level 1 (the $3\mu\text{m}$ lower laser level) interacting such that one ion is promoted to level 3 and the other is de-excited to the ground state. The ion in level 3 then undergoes rapid multiphonon decay to level 2, which is the $3\mu\text{m}$ upper laser level. The effect of this is to remove two excitations from the lower laser level and add one to the upper laser level [7]. It is this process that has been shown to stop the self termination of the $3\mu\text{m}$ transition [8]. The second ETU process, W_{22} , originates from level 2 (the $3\mu\text{m}$ upper laser level) and causes depletion of the population of this level and excitation to level 7 ($^4F_{7/2}$) with fast multiphonon decay to populate level 5 ($^4S_{3/2}$). It is emission from this latter level that is responsible for producing the characteristic green fluorescence from erbium-doped laser crystals [9, 10]. The cross relaxation process, W_{05} , involves ions in level 5 and the ground state (level 0) and acts to repopulate both of the levels involved in the $3\mu\text{m}$ laser transition (1 and 2). In highly doped Er^{3+} crystals this W_{05} cross relaxation process causes fast decay of the level 5 population. We note that excited state absorption (ESA) from level 2 ($^4I_{11/2}$), denoted by the red dashed arrow in figure 1, can occur and also generates population in level 7 ($^4F_{7/2}$), although this process is considered to be small in highly doped erbium crystals relative to the W_{22} ETU process.

We have measured the spectrum of the characteristic green fluorescence from a diode pumped 50at.% Er:YSGG laser crystal and this is presented in figure 2. The spectral features labeled Green, Red, NIR-1 and IR in

figure 2 correspond to the similarly labeled curved lines in figure 1 which illustrate the transitions responsible for this fluorescence.

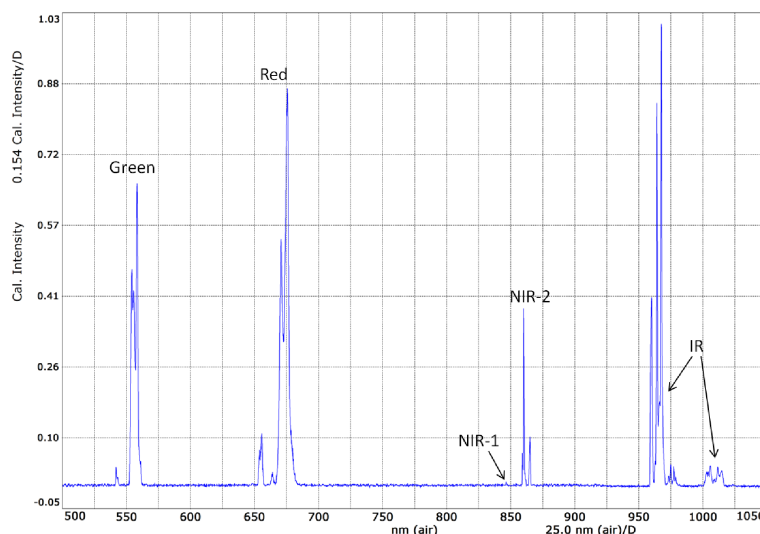


Fig. 2. Spectrum of fluorescence from 50at.% Er:YSGG crystal pumped by laser diode at ~ 970 nm with pump repetition rate of 50Hz, pump duration 100 μ s and pump pulse energy ~ 25 mJ.

The green signal, which gives the fluorescence its characteristic appearance, is produced by a transition beginning in level 5 and ending in the ground state. As the ETU process W_{22} and ESA from level 2 are both possible population mechanisms for level 5, further investigation of this transition may give insight into their strength/role in the erbium system. The red fluorescence originates in level 4, which is mainly bypassed in the erbium system, and terminates in level 0. The NIR fluorescence originates in level 5 (from which the green fluorescence also originates) and terminates in the $3\mu\text{m}$ lower laser level (level 1). The wavelengths of 860/865nm correspond to transitions from the lowest (and most populated) Stark sub-level of the $^4S_{3/2}$ state to the two highest (and least populated) Stark sub-levels of the $^4I_{13/2}$ state. The IR fluorescence is produced by a transition which begins in level 2 and terminates in the ground state. Study of this signal could provide a direct probe of the $3\mu\text{m}$ upper laser level population. It should also be noted that very weak features at ~ 418 nm (level 8 to level 1) and ~ 845 nm (level 3 to level 0) were also observed when using a longer integration time.

The existence of these numerous fluorescence signals demonstrates the complexity of the erbium system. Although the mechanisms that they evidence can greatly enhance long-pulse or CW operation of the erbium laser, this is not necessarily the case in the short-pulse, Q-switched regime. This is because on the tens-of-nanoseconds timescale of the Q-switch pulses, the energy transfer process, W_{11} , responsible for the high efficiency in the long-pulse/CW regime, is too slow to recycle the lower laser level population. It is therefore not clear what the effect of these mechanisms is on the energy storage potential of the $3\mu\text{m}$ upper laser level and for Q-switching of these lasers.

3. Mechanical Q-switching of Er:YSGG lasers

Pulsed operation of the Er:YSGG laser was achieved through mechanical Q-switching using a rotating polygon mirror. This approach is attractive compared to electro-optic (EO) switching as it eliminates the high voltage switching requirement and the cavity insertion loss which is particularly detrimental in these low gain lasers.

The experimental set-ups for the mechanically Q-switched Er:YSGG lasers are shown in figure 2. Mechanical Q-switching was achieved by means of a gold plated, ten-sided polygon mirror mounted onto a motor and

electronically synchronised to the diode pump module. This unit was incorporated using two different Q-switching arrangements. The first, shown in figure 2(a), used the polygon as a rotating back mirror. In this set-up laser action was inhibited during the pump pulse due to mirror misalignment and the end of the pump pulse is synchronised to coincide with re-alignment of the laser cavity and release of a Q-switched pulse. Figure 2(b) illustrates the second approach for Q-switching, which used the rotating polygon as an intra-cavity chopper for the laser mode. Here the polygon was positioned so that as it rotated the laser cavity was successively blocked and unblocked. As in the first case, the rotation of the polygon was synchronised with the diode pumping to prevent lasing during the pump pulse but allow build up and release of a Q-switched pulse at the end of pumping. Both arrangements utilized the diode side pumped bounce geometry [11], in which the laser mode takes a grazing incidence total internal reflection off the pump face of the laser crystal. This provides access to the region of highest inversion just below the pump surface, as well as spatial averaging of gain and thermal non-uniformities. Additionally, this geometry allows straightforward and effective delivery of radiation from high power diode bars and stacks to the laser crystal.

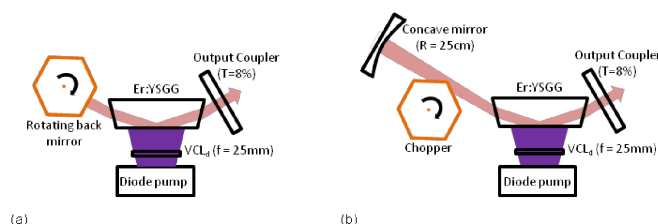


Fig. 3. Experimental set-up of Q-switched Er:YSGG laser using rotating polygon as a) the back mirror and b) an intra-cavity chopper.

The laser crystals investigated in this work were a 50at.% Er:YSGG slab and a 38at.% Er:YSGG slab. The 50at.% Er:YSGG slab had dimensions of 20mm x 5mm x 2mm and was held in a water-cooled copper mount. This crystal was transversely cooled from its large (20mm x 5mm) top and bottom faces and side pumped via the 20mm x 2mm face. The laser faces were cut to be at Brewster's angle for the laser mode when operating in the bounce geometry. The 38at.% Er:YSGG slab had dimensions of 20mm x 2mm x 5mm and was used in a face-cooled regime. Here heat is removed directly from the (20mm x 5mm) pump face, which is also the area of greatest heating, which can significantly improve the thermal performance of the laser. For this, a sapphire block (with high thermal conductivity) was optically contacted to the 20mm x 5mm pump face of the laser crystal and these were held in a copper mount sitting on a water-cooled copper block. The laser faces were also cut at the Brewster's angle for the laser mode as with the other crystal. Diode pumping of the laser crystal was achieved using either a 3-bar diode stack with nominal wavelength of 971nm, or two single diode bars with nominal wavelength of 966nm angularly combined to overlap at the pump face of the crystal (which we denote as dual diode pumping). In both schemes a vertical cylindrical lens (VCL_v) with focal length $f=25\text{mm}$ was used to focus the pump radiation to a horizontal line ($\sim 100\mu\text{m} \times 10\text{mm}$) at the crystal face.

3.1 Q-switching results from the rotating back mirror set-up

In the set-up of figure 2(a) the transversely cooled 50at.% Er:YSGG laser crystal was used in conjunction with dual diode pumping. The rotating polygon was positioned such that when one of its reflecting surfaces was aligned it formed a resonator of length $\sim 80\text{mm}$, with a distance of 65mm between the polygon surface and centre of the pump face of the laser crystal. The output coupler was a plane mirror with transmission of 8%. The polygon mirror was operated at a rotation rate of 395 rps and a laser cavity was formed 10 times during each rotation by its ten reflecting faces (3950Hz). A synchronisation signal was sent on each rotation to an adjustable divider circuit before triggering the diode pump module. The electronics allowed adjustable delay for the optimum Q-switching time relative to pump pulse. With a pump pulse repetition rate of 30Hz, with QCW pump power of 290W for a duration of 200 μs (58mJ), this set-up yielded Q-switched pulse energy of 0.47mJ and pulse duration of 83ns before rapidly degrading over a period of a few seconds. This degradation in performance was due to laser damage caused by the Q-switching which was observed on the polygon facet after this result was obtained. Incrementally moving the polygon mirror to an undamaged location produced a similar result with laser damage again causing a fall-off in the pulse energy. This problem could be solved or improved through use of a better coating with higher damage threshold. The pulse rate that this system was operated at was relatively low because thermal lensing ruined stability of the simple plane-plane cavity and prevented operation at higher repetition rates. Decreasing the distance between the polygon surface and the laser crystal

facet could help here as the effect would be to decrease diffractive losses and aid resonator stability. However this was unfortunately not possible owing to the dimensions of the motor. It also may be possible to counteract this effect with additional cavity optics but this line of investigation was not pursued in this work especially in view of the mirror damage issue.

3.2 Q-switching results from the intra-cavity chopper set-up

With the aim of achieving higher repetition rates and eliminating laser induced damage to the coated mirror surface, the set-up of figure 2(b) was implemented. A concave mirror with radius of curvature $R=25\text{cm}$ acted as the back mirror of the laser cavity (length $\sim 28\text{cm}$), positioned both to match the fundamental mode size to the narrow vertical gain region and to maintain cavity stability despite strong thermal lensing. The rotating polygon blocked the laser cavity at a distance of 4cm from centre of the pump face of the laser crystal and 10 times during each rotation. The motor was again operated at a rotation rate of 395 rps. Both the transversely cooled 50at.% Er:YSGG crystal and the face-cooled 38at.% Er:YSGG crystal were tested in this configuration, with pumping by the 3-bar stack.

Figure 3 shows Q-switched results obtained with the 50at.% Er:YSGG crystal. The Q-switched pulse energy (left axis, black data points) and Q-switched pulse duration (right axis, red data points) obtained at pump pulse repetition rate of 100Hz and pump pulse duration of $100\mu\text{s}$ are presented. The highest QS pulse energy of 0.55mJ and shortest QS pulse duration of 154ns were achieved at the maximum pump energy of 40mJ . The standard deviation of the energy stability was $\sim 2\%$ and of the pulse duration stability was $\sim 1\%$, showing good pulse-to-pulse repeatability even for this mechanical Q-switching technique. The longer pulse duration obtained with this set-up compared to the first system (154ns compared to 83ns) was caused by the increased cavity length (280mm compared to 80mm).

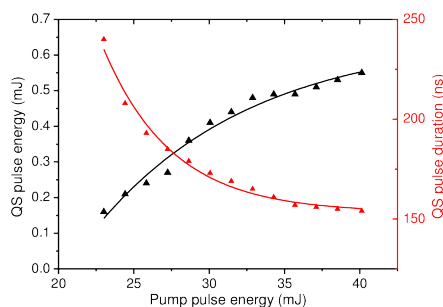


Fig. 4. Q-switching results from the diode pumped, mechanically Q-switched 50at.% Er:YSGG laser, operating at 100Hz using the rotating polygon as intra-cavity chopper.

Replacing the transversely cooled 50at.% Er:YSGG crystal by the face-cooled 38at.% Er:YSGG crystal in the intra-cavity chopper configuration enabled even higher repetitions rates to be achieved. Q-switched pulses with energy of 0.19mJ and duration of 171ns were achieved at repetition rate of 200Hz, pump duration of $70\mu\text{s}$ and pump energy of 28mJ .

4. Conclusion

Mechanical Q-switching of diode-pumped Er:YSGG lasers operating at the $3\mu\text{m}$ transition has been presented. A rotating polygon mirror was used to achieve pulsed operation and two experimental configurations were tested. In the first set-up the rotating polygon mirror was used as the back mirror of the laser cavity, which produced Q-switched pulses with 0.47mJ pulse energy and 83ns pulse duration at 30Hz. Unfortunately laser damage on the surface of the rotating polygon mirror (caused by the Q-switching) led to rapid degradation of this result. In the second set-up the rotating polygon mirror was used as an intra-cavity chopper for the laser mode. High repetition rate Q-switching was achieved with this configuration, with pulses of 0.55mJ energy and 154ns duration achieved at 100Hz with the transversely cooled 50at.% Er:YSGG crystal and pulses of 0.19mJ energy and 171ns duration achieved at 200Hz. Spectroscopy of fluorescence signals from the diode-pumped 50at.% Er:YSGG crystal demonstrates the complicated nature of the system.

A.5. Other photos



Figure A.4.: Me (pre laser-eye surgery), the view from the lab (Huxley 901) of the Queen's Tower, and the view of Huxley 901 from the top of the Queen's Tower.

Nomenclature

δ_{ISBT}	FWHM linewidth of inhomogeneously-broadened ISBT
$\gamma^{(inter)}$	FWHM linewidth pertaining to the intersubband broadening
$\gamma^{(intra)}$	FWHM linewidth pertaining to the intrasubband broadening
γ_{IRS}	FWHM linewidth pertaining to total IRS broadening
γ_{phonon}	FWHM linewidth pertaining to total phonon broadening
$\Omega_{R,\varphi_{res}}$	vacuum Rabi splitting at resonance in $\varphi - \omega$ space
$\Omega_{R,k_{\parallel,res}}$	vacuum Rabi frequency, the splitting at $k_{\parallel,res}$
φ_{res}	incident angle where ISBT and microcavity mode resonate
ξ_{ISBT}	FWHM inhomogeneous contribution to broadening
$k_{\parallel,res}$	in-plane wavevector where ISBT and microcavity mode resonate
L_{MC}	microcavity length
m_{scat}^*	effective mass of particle scattered by IRS
n_f	final population inversion density
n_i	initial population inversion density
n_t	threshold population inversion density
t_d	laser pulse build-up time
t_e	time between flashlamp trigger and laser emission
t_p	FWHM pulsewidth of laser

t_Q	Q switching time
t_r	round trip time of laser cavity
2DEG	2D electron gas
cQED	cavity quantum electrodynamics
CR	cross-relaxation
EMA	effective medium approach
EOQS	electro optic Q switch
ETU	energy-transfer upconversion
FT/IR	fourier transform infrared spectrometer
FTQS	frustrated total internal reflection Q switch
FWHM	full width at half maximum
ICP	intersubband cavity polariton
IRS	interface roughness scattering
ISBT	intersubband transition
LA	longitudinal acoustic phonon
LIA	lock-in amplifier
LO	longitudinal optical phonon
LPB	lower polariton branch
MBE	molecular beam epitaxy
MCT	mercury cadmium telluride infrared detector
microcavity	an optical cavity on the length scale of a micron, designed to strongly quantise light of optical and infrared wavelengths
MQW	multi-quantum well

NRC sample	the ICP sample studied in this thesis
p-polarised	parallel polarised - electric field is parallel to the plane of incidence. See TM-polarised
PCQS	polygon chopper Q switch
pps	pulses per second
Q-factor	the ratio of the energy stored in the cavity to the energy loss per cycle
QCL	quantum cascade laser
QW	quantum well
RMQS	rotating mirror Q switch
rps	rotations per second
SCR	strongly coupled regime
STCO	simple two coupled oscillator model for the ICP
subaverage	a description of ICP linewidths which are narrower than the STCO model predicts
TE-polarised	Transverse Electric-polarised - in the case of waveguides, a mode of light with the electric field transverse to the propagation direction
TIR	total internal reflection
TM-polarised	Transverse Magnetic-polarised - in the case of waveguides, a mode of light with the magnetic field transverse to the propagation direction (see p-polarised)
TMF	transfer matrix formalism
UCR	ultrastrong coupling regime
UPB	upper polariton branch
YAG	yttrium aluminium garnet
YSGG	yttrium scandium gallium garnet

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My first thanks go to Prof Chris Phillips for giving me the opportunity to work in his laboratory, for having such a welcoming open-door policy, and for creating a working environment conducive to producing results and also exploring our own ideas. My funding source, EPSRC, are also thanked for their support.

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FJM