

TOPICS IN STATISTICAL MECHANICS

A THESIS PRESENTED FOR THE DEGREE OF
DOCTOR OF PHILOSOPHY OF IMPERIAL COLLEGE LONDON
AND THE
DIPLOMA OF IMPERIAL COLLEGE
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MARCH 2019

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Topics in Statistical Mechanics

Johannes Pausch

ABSTRACT

This thesis aims to showcase the versatility of statistical mechanics. It splits into two parts: firstly, applications of Doi-Peliti field theory to filament growth and branching processes, and secondly, wetting phenomena on structured surfaces.

In Chapter 1, I derive the Doi-Peliti field theory; the derivation starts with stochastic processes that obey the Markov property and goes on to link them to the master equation. It is then recast as a partial differential equation of the probability generating function, the solution of which is found perturbatively with the Doi-Peliti path integral.

This path integral formulation is used in Chapter 2 to model continuous-time branching processes. The critical behaviour of these processes is studied analytically and corroborated with simulations. The results were also published in [51] and show a natural link between the mathematics of stochastic processes and Doi-Peliti field theory.

The field theory is then applied to the reaction-diffusion process of filament growth in Chapter 3. The filaments are polymers found in cells of living organisms, which assemble by incorporating their building blocks from the environment. However, if the environment does not contain enough building blocks, the filaments disassemble. The stochastic switching between disassembly and assembly leads to intriguing dynamics. Some of the results are part of a published article [118].

Statistical mechanics is also concerned with equilibrium phenomena. A class of these phenomena is studied in Chapter 4 in the context of wetting. Wetting occurs when a fluid forms a liquid film on a solid surface. If the surface is *structured*, i.e. has edges or corners, the occurrence of phase transitions of the liquid film depends on its specific structure. The structured surfaces studied in this thesis are the wedge and the apex; the results for them were also published in [133].

To my family, old and new.

ACKNOWLEDGMENTS

This thesis would not have been written without the help and support of many people. First and foremost, I thank my supervisors Andy Parry and Gunnar Pruessner who did not get tired of motivating me. Their enthusiasm is infectious and their constructive criticism paired with their wit guided me through highs and lows. They always lend an ear and looked at the products of my work with open eyes – even when I presented utterly erroneous ones. I cannot imagine finding better supervisors.

Many of my friends, PhD siblings and colleagues also played huge rôles in the rational and emotional rollercoaster of my thesis. Some supported me as office mates or SIAM chapter colleagues, other more directly through collaborations or with helpful discussions, proofreading and feedback at my Early Stage Assessment or Late Stage Review: Benjamin Walter, Rosalba Garcia-Millan, Ignacio Bordeu Weldt, Paul Gefert, Soairse Armarteifio, Nanxin Wei, Geina Maziya, Simon Schöller, Hemant Khatri, Alexis Arnaudon, Chiara Taranto, Luca Cocconi, Ziluo Zhang, Adam Collins, Marianna Chatzakou, Alexandr Malijevský, Carlos Rascón, Thomas Surrey, Robert Endres, Guillaume Salbreux, Eva-Maria Gräfe, Ryan Barnett and many more. Anderson Santos also deserves a huge thank you, as he made everyone's life in the department more enjoyable through his uplifting spirit and dedication to his students.

Supportive for decades or only for a few years have also been my family of which I can only name a few here: Hannah Sparrowhawk, my parents Annette and Guntram Pausch, my brother Richard Pausch, my grandparents Anni and Udo Kasulke, Brigitte and Dankfried Pausch.

Thank you.

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The difficulty lies, not in the new ideas, but in escaping from the old ones [...].

John Maynard Keynes [85]



INTRODUCTION

Statistical mechanics is a versatile toolbox that has its origins in thermodynamics [75, 110, 72, 140, 89]. The following brief account of its development reflects my personal viewpoint rather than a balanced overview.

The phenomena that statistical mechanics tries to explain have been studied decades and centuries before it was formally recognized as a field. The phase transitions of melting and solidifying were used to make metal objects of particular shapes more than 7,000 years ago [30, 129], and in the 1st century, liquid-gas transitions were used to distill alcohol [163] and run steam powered machines [39]. A critical point was first scientifically observed in 1822 by *Charles Cagniard de la Tour* [19, 8] using alcohol.

Explaining the rich phenomenology of materials took many years and numerous contributions from scientists of which many remain famous today: *Rudolf Clausius*, *Ludwig Boltzmann*, *James Clerk-Maxwell*, *Willard Gibbs*, *Johannes van der Waals* to name a few. In fact in Chapter 4, I will use a graphical construction analogous to *Maxwell's* original equal areas construction from 1875, which he used to explain the first-order phase transition from gas to liquid [27]. Notions like partition function, ensembles, entropy and kinetic theory originated in this time before the first World War [89].

With the introduction of quantum mechanics and the understanding of the spin of a

particle, a qualitatively new kind of model entered statistical mechanics: the lattice spin model. In 1925, *Wilhelm Lenz* and *Ernst Ising* introduced the first of many attempts to link macroscopic magnetism to the order or disorder of microscopic spins [75, 4]. In 1943, *Lars Onsager* was able to show analytically that the Ising model exhibits a phase transition using the transfer matrix method [110]. This result ignited a discovery of many more lattice spin models, including the Potts model [125] and the XY-model [93], of which only a few have been solved exactly today [4]. Understanding the universal behaviour around second order phase transitions became a new focus. Their associated critical exponents became accessible through the study of scaling laws, notably by *L. Kadanoff* in 1966 [79], and renormalization group procedures by *K. Wilson* in 1971 [169, 170].

Up until this point, the centre of interest was equilibrium statistical mechanics. With the introduction of statistical dynamics on the Ising model by *R.J. Glauber* in 1963 [58] and *K. Kawasaki* in 1966 [83], non-equilibrium statistical mechanics on lattices became more prominent. Shortly after in 1973, *P.C. Martin*, *E.D. Siggia* and *H.A. Rose* [98] contributed a dynamical statistical field theory, followed by an almost complete characterization of the involved critical phenomena by *P.C. Hohenberg* and *B.I. Halperin* in 1977 [67].

Since then, statistical mechanics has spread its wings and invaded many fields which are rarely associated with physics, including:

- the mathematical theory of stochastic processes ([160, 77, 104] and Chap. 2),
- biology ([9, 32, 42] and Chap. 3),
- material science ([162] and Chap. 4),
- finance [14, 80], and even
- social sciences [78, 97],

to name only a few.

Its success can be explained by its ability to combine complex stochastic behaviour through rigorous tools in order to extract emergent phenomena. This thesis attempts

to add a few drops to the ocean of statistical mechanics. Rather than aligning with a common storyline, the individual chapters try to showcase the versatility of the field.

As Chapter 2 and 3 are based on the same method, Doi-Peliti field theory, I have decided to arrange ample space for its derivation in Chapter 1. My hope is that by exposing many of the details of its inner workings, its possible extensions can be added more easily.

The first, theory-focused chapter is followed by a chapter that attempts to link statistical mechanics to the mathematical theory of stochastic processes, which is in turn followed by a chapter that tries to link statistical mechanics to biology. The final chapter goes back to the roots of statistical mechanics with an application to material science and chemistry.

Each chapter starts with an abstract, where I point out which parts are original contributions, and ends with an outlook on future research. Almost all the presented work is based on collaborations and I will use the word *I* only to highlight personal choices for the exposition of the results. *We* is used as *the reader and I* when referring to arguments and equations on preceding pages, sections and chapters.

[...] the infinite answers are all too prevalent and confusing.

Richard P. Feynman, PhD thesis [45]

1

DOI-PELITI FIELD THEORY

ABSTRACT

Doi-Peliti field theory is the main method used in Chapters 2 and 3 for analysing stochastic processes. Its fundamentals are derived in this chapter and assumed in the following chapters. The three main steps of its derivation are: modelling reaction-diffusion systems with a master equation, which is an infinite system of coupled ordinary differential equations; transforming it into a single partial differential equation (PDE) for the probability generating function using the language of second quantization; solving the PDE using a path integral formulation. Open questions and an outlook on my future research is presented in this chapter's conclusion, Sec. 1.6.

1.1 INTRODUCTION

The path integral is an elegant tool which allows its user to systematically find approximate – and sometimes even complete – n -point correlators for quantum or classical stochastic processes [148, 174, 92, 121, 154]. Its origins trace back to *Paul Dirac* [36] and *Richard Feynman* [45]. Various derivations have been presented since, with varying degree of mathematical rigour. However, despite being widely accepted in physics, it evades acceptance in mathematical communities [153, 29].

Doi-Peliti field theory is a class of Field Theories which builds on work by *Masao Doi* [37] and *Luca Peliti* [120]. Typically, field theories in this class are derived from a class of models which are cast as master equations of microscopic Poisson processes.

In the following, I present a derivation of Doi-Peliti Field Theory. Furthermore, I translate the objects that are considered the results or observables of a field theory to objects in the mathematics of stochastic processes. I point out areas of interest into which these field theories could extend in the future, as well as concerns of mathematical rigour which are typically ignored in the physics communities. Furthermore, I point out which master equations cannot be turned into a Doi-Peliti field theory.

Over the last decades, Doi-Peliti field theory has proven itself to be a useful tool in non-equilibrium statistical mechanics and it has been applied to percolation [156, 66, 77], reaction-diffusion fronts [69], predator-prey models [155], unidirectionally coupled systems [60], branching random walks [23, 71], the voter model [70] and neuroscience [18].

1.2 MASTER EQUATION

The following derivations and explanations are based on Chapters III, IV, and V of N.G. van Kampen’s textbook *Stochastic Processes in Physics and Chemistry* [162].

A *stochastic process* is a discrete or continuous sequence of random variables $(N_t)_{t \in I}$ each of which map from the same sample space Ω to the same co-domain Y :

$$\forall t \in I : N_t : \Omega \rightarrow Y. \tag{1.1}$$

In the following, the index set I of the sequence is interpreted as time $I = \mathbb{R}$ and is equipped with the euclidian distance function $|t_1 - t_2|$. The probability that the random variable N_t is equal to $n \in Y$ is denoted by $P(n, t)$ or $P(N(t) = n)$. The probability that $N_t = n$ under the condition that $N_s = m$ is denoted by $P(n, t|m, s)$ or $P(N(t) = n|N(s) = m)$.

In Chapter 2 on continuous-time branching, N_t represents a single particle number, i.e. $\Omega = Y = \mathbb{N}_0$, but there is no space for movements involved. Here, other random variables are derived from N_t such as the time of death $T = \inf\{t \in \mathbb{R}_{>0} : N_t = 0\}$ or the dimensionless trajectory size $S = \int_0^T s N_t dt$, where s is the system's internal rate.

In Chapter 3 on filament growth, N_t is interpreted as a pair of particle numbers, one for monomers and one for filament tips on a lattice $\mathcal{X} = \mathbb{Z}^3$ for movement, with $\Omega = Y = (\mathbb{N}_0^2)^\mathcal{X}$. In both chapters, the goal is to derive and understand the properties of the distribution P such as expectation values, and higher moments.

1.2.1 MARKOV PROCESSES

Markov processes are stochastic processes whose conditional probabilities satisfy the Markov property:

$$\begin{aligned} \forall k \geq 1 \forall j \in \{1, \dots, k\} \forall t_j < t : \\ P(n, t|m_1, t_1; \dots; m_k, t_k) &= P(n, t|m_{j_{\max}}, t_{j_{\max}}) \\ \text{with } j_{\max} = j \in \{1, \dots, k\} : t_j &= \max\{t_1, \dots, t_k\} \end{aligned} \tag{1.2}$$

A process that has the Markov property is interpreted as memoryless because in a sequence of random events, only the most recent one determines the likelihood of the next.

Two examples of stochastic processes which have the Markov property are Markov Chains in discrete time and Poisson processes in continuous time. In particular, reaction-diffusion processes are an example of a Markov process with discrete number of particles in continuous time.

1.2.2 CHAPMAN-KOLMOGOROV EQUATION

For a general stochastic process, the joint probability of three events at times $t_1 \geq t_2 \geq t_3$ can be rewritten as a sequence of conditional events

$$\begin{aligned} P((n_1, t_1) \wedge (n_2, t_2) \wedge (n_3, t_3)) &= P(n_1, t_1 | (n_2, t_2) \wedge (n_3, t_3)) P((n_2, t_2) \wedge (n_3, t_3)) \\ &= P(n_1, t_1 | n_2, t_2 \wedge n_3, t_3) P(n_2, t_2 | n_3, t_3) P(n_3, t_3) \end{aligned} \quad (1.3)$$

If this process has the Markov property (1.2), the last line can be simplified to

$$P((n_1, t_1) \wedge (n_2, t_2) \wedge (n_3, t_3)) = P(n_1, t_1 | n_2, t_2) P(n_2, t_2 | n_3, t_3) P(n_3, t_3) \quad (1.4)$$

This seemingly subtle difference can be exploited by integrating / summing over n_2 and dividing by $P(n_3, t_3)$ to obtain the Chapman-Kolmogorov equation (CKE) [86]

$$P(n_1, t_1 | n_3, t_3) = \sum_{n_2 \in Y} P(n_1, t_1 | n_2, t_2) P(n_2, t_2 | n_3, t_3). \quad (1.5)$$

The Chapman-Kolmogorov equation can also be formulated for random variables with a continuous range Y and its conditional probability density functions $f_N(n, t | m, t')$:

$$f_N(n_1, t_1 | n_3, t_3) = \int_{n_2 \in Y} f_N(n_1, t_1 | n_2, t_2) f_N(n_2, t_2 | n_3, t_3) dn_2 \quad (1.6)$$

The Chapman-Kolmogorov equation can be interpreted as follows: The probability of the transition from state n_3 to state n_1 is given by the sum / integral over the probabilities of transition to and from all intermediate states n_2 . The Markov property allows separating the probability of transition between the initial state n_3 and the intermediate state n_2 from the probability of transition between the intermediate state n_2 and the final state n_1 .

The Chapman-Kolmogorov eq. (1.5) can also be regarded as the origin of a path integral by considering paths of the stochastic process. Starting at state n_s at time t_s , what is the probability to find the system in the final state n_f at time t_f ? In discrete time, the answer can be formulated as a sum over all intermediate paths $n(t) : I \rightarrow Y$, with

$n(t_s) = n_s$ and $n(t_f) = n_f$:

$$P(n_f, t_f | n_s, t_s) = \sum_{\{n(t)\}} \prod_{t=t_s}^{t_f-\Delta t} P(n(t+\Delta t), t+\Delta t | n(t), t), \quad (1.7)$$

where Δt is the time step size of the index set of the stochastic process $I = \Delta t \mathbb{Z}$. I call this a path sum because the set of all intermediate paths $\{n(t)\}$ is countable. However, at this point, it is not clear how to formulate a continuous time version of this path sum. In particular when looking naively at the product in Eq. (1.7), the continuum limit appears to imply that an infinite product of numbers between 0 and 1 is taken. Then, every term in the path sum would surely be equal to zero, which in turn would imply the contradiction that all probabilities $P(n_f, t_f | n_s, t_s)$ must equal zero. I therefore conclude that the continuum limit must be taken very carefully. Progress can be made by introducing a differential form of the Chapman-Kolmogorov equation.

1.2.3 DIFFERENTIAL FORM OF THE CKE

If the index set of the stochastic process is $\Delta t \mathbb{Z}$, then it is of interest to ask what happens if $\Delta t \rightarrow 0$. The probability $P(n, t)$ becomes a function of the real variable t , and I assume that it is differentiable. Thus, the Chapman-Kolmogorov equation (1.5) can be differentiated w.r.t. t_1 :

$$\frac{\partial P(n_1, t_1 | n_3, t_3)}{\partial t_1} = \sum_{n_2} \frac{\partial P(n_1, t_1 | n_2, t_2)}{\partial t_1} P(n_2, t_2 | n_3, t_3) \quad (1.8)$$

The derivatives can be understood better, if $t_1 - t_2 =: \tau$ is assumed to be small. With the differentiability assumption, a Taylor expansion to first order in time of the conditional probabilities can be expressed as:

$$P(n_1, t + \tau | n_2, t) = P(n_1, t | n_2, t) + \tau \frac{\partial P(n_1, t' | n_2, t)}{\partial t'} \Big|_{t'=t} + \mathcal{O}(\tau^2). \quad (1.9)$$

While $P(n_1, t | n_2, t) = \delta_{n_1, n_2}$, the derivative of P splits into two parts: $n_2 \neq n_1$ and $n_2 = n_1$. The former case describes the change of probability over time for a transition from state $n_2 \neq n_1$ to n_1 and will be denoted by $W(n_1 | n_2)$. The latter describes the change of probability over time to stay in $n_2 = n_1$. The probability to stay in state n_1

over a time τ is 1 minus the probability of leaving during time τ :

$$\text{probability to stay in } n_1 \text{ for time } \tau = 1 - \sum_{n' \neq n_1} W(n'|n_1)\tau + \mathcal{O}(\tau^2). \quad (1.10)$$

Thus, the time derivative of the conditional probability is identified as

$$\left. \frac{\partial P(n_1, t'|n_2, t)}{\partial t'} \right|_{t'=t} = W(n_1|n_2)(1 - \delta_{n_1, n_2}) - \sum_{n' \neq n_1} W(n'|n_1)\delta_{n_1, n_2}. \quad (1.11)$$

The $W(\cdot|\cdot)$ quantify infinitesimal changes in the conditional probabilities and are therefore associated with transitions caused by independent, microscopic processes. Using Eq. (1.11), the differentiated Chapman-Kolmogorov Eq. (1.8) can be written for all $t_1 > t_2 > t_3$ as

$$\boxed{\frac{\partial P(n_1, t_1|n_3, t_3)}{\partial t_1} = \sum_{n_2 \neq n_1} \left(W(n_1|n_2)P(n_2, t_2|n_3, t_3) - W(n_2|n_1)P(n_1, t_2|n_3, t_3) \right)} \quad (1.12)$$

This differential form of the Chapman-Kolmogorov equation (CKE) is the master equation. While the sum of $W(n_1|n_2)$ over n_2 is called *gain*, the sum of $W(n_2|n_1)$ over n_2 is termed *loss*. Often, the explicit mentioning of the condition on the system to have been in state n_3 at time t_3 is omitted for a shorter notation and I will occasionally use this abbreviation in the following sections and chapters. However, the condition on the initial state (n_3 at time t_3) must be kept in mind.

Going back to the sum over all paths, Eq. (1.7), the Taylor expansion Eq. (1.9) can be used for $\tau = \Delta t$ to obtain

$$P(n_f, t_f|n_s, t_s) = \sum_{\{n(t)\}} \prod_{t=t_s}^{t_f-\Delta t} \left(P(n(t), t|n(t), t) + \Delta t \left. \frac{\partial P(n(t'), t'|n(t), t)}{\partial t'} \right|_{t'=t} \right), \quad (1.13)$$

where I assume that $n(t)$ is continuous from the right, i.e. for all $t \in \mathbb{R}$, $n(t) = \lim_{t' \rightarrow t^+} n(t')$ (called *càdlàg*). This assumption can be interpreted as: new values of $n(t)$ occur at a specific time. In the branching process, this means that particles are created or become extinct at a specific time. With this assumption, $\lim_{\Delta t \rightarrow 0} P(n(t + \Delta t), t + \Delta t|n(t), t) = 1$

and Eq. (1.13) becomes an exponential function in the limit $\Delta t \rightarrow 0$.

Even when the values of $n(t)$ are discrete, the set of all paths $\{n(t)\}$ becomes a continuum as the time becomes uncountable. Hence, the sum over all paths is formally replaced by an integral over all paths:

$$P(n_f, t_f | n_s, t_s) = \int \exp \left(\int_{t_s}^{t_f} \frac{\partial P(n(t'), t' | n(t), t)}{\partial t'} \bigg|_{t'=t} dt \right) \mathcal{D}n(t) \quad (1.14)$$

$$= \int \exp \left(- \int_{t_s}^{t_f} \sum_{n' \neq n(t)} W(n' | n(t)) dt \right) \mathcal{D}n(t). \quad (1.15)$$

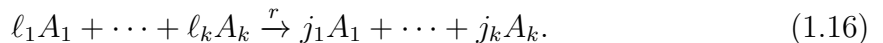
This integral over all paths from state n_s at time t_s to state n_f at time t_f has as its integrand a weight of paths. A path has a small weight if there are many and likely ways of transitioning to a different path. A path has a higher weight, if there are fewer and less likely ways of transitioning to a different path.

The path integral (1.15) is rather impractical because it is not clear how the the measure $\mathcal{D}n(t)$ for a specific n_s and n_f should be defined or normalized.

The time derivative of the conditional probabilities $W(n_1 | n_2)$ can be time-dependent in general. An example of this case is shown in Sec. 2.9.1.

What are the time derivatives of conditional probabilities, $W(n_1 | n_2)$? In principle, any time distribution could be used to derive specific expressions for $W(n_1 | n_2)$. Here, I consider reaction-diffusion processes which are Markov processes with exponentially distributed waiting times. Hence, $W(n_1 | n_2)$ is typically a product of a base rate and binomial coefficients. While the base rate characterizes the underlying time scale for a reaction to occur, the binomial coefficients incorporate that the reaction can happen between any of the available and suitable reactants, i.e. the binomial coefficients are used to calculate the number of independent reaction processes.

Example: Let a reaction occur between particle types $A_1, \dots, A_k$ with base rate r :



The reaction requires ℓ_1 particles of type $A_1, \dots$, and ℓ_k reactants of type A_k and

produces j_1 particles of type $A_1, \dots$, and j_k particles of type A_k . Before the reaction, the system is in state n_2 with particle numbers $n_2 = \{m_1, \dots, m_k\}$ with $m_i \geq \ell_i$, i.e. the initial state must contain at least the particles necessary for the reaction. After the reaction, the system is in state n_1 with particle numbers $n_1 = \{m_1 - \ell_1 + j_1, \dots, m_k - \ell_k + j_k\}$. Then, $W(n_1|n_2)$ equals

$$W(n_1|n_2) = r \binom{m_1}{\ell_1} \cdots \binom{m_k}{\ell_k}, \quad (1.17)$$

i.e. we choose ℓ_i particles of type A_i necessary for the reaction. The product of the binomials is equal to the number of particle subsets in the initial state n_2 that can react. Each subset waits a $\sim \text{Exp}(r)$ -distributed time to react. Because they are independent, the overall waiting rate is the sum of the individual rates, equaling $W(n_1|n_2)$.

If the process contains several independent reactions, then for each reaction a transition rate $W(n_1|n_2)$ is derived and, due to the assumed independence of the processes, they are all added up for the overall transition rate.

Processes whose transition rates $W(\cdot|\cdot)$ are not of the form (1.17) cannot be transformed into a Doi-Peliti field theory: for example if $W(\cdot|\cdot)$ depended on $1/m_1$, the formalism which is introduced in the following sections would not work.

1.3 THE PROBABILITY GENERATING FUNCTION

From now on, I assume that the range of the considered stochastic process is the space of vectors of non-negative integers representing the number of particles of various types of reactants ($A_1, \dots, A_k$) in the system. Let us assume there are k particles types, then a state n of the system is described by a list of numbers of particles $m_1, \dots, m_k$. The probability generating function is defined as

$$\mathcal{M}_o(z, t) = \sum_{\{m_1, \dots, m_k\}} P(m_1, \dots, m_k, t) z_1^{m_1} \cdots z_k^{m_k}, \quad (1.18)$$

where the sum goes over the entire (countable) set $\{m_1, \dots, m_k\} = \mathbb{N}_0^k$ of possible particles numbers. Each particle type A_j with $j \in \{1, \dots, k\}$ is represented by its own auxiliary complex variable z_j . Definition (1.18) is the original definition for probability

generating functions. However, they can also be defined for marginal and conditional probabilities. In the following, I am always going to use the probability generating function for probabilities conditioned to a specific initialization of the system at time t_0 :

$$\mathcal{M}(z, t) = \sum_{\{m_1, \dots, m_k\}} P(m_1, \dots, m_k, t | n_1, \dots, n_k, t_0) z_1^{m_1} \dots z_k^{m_k}. \quad (1.19)$$

Here, the condition / initialization is the state with n_1 particles of type A_1 , $\dots$, and n_k particles of type A_k at time t_0 . In the following, I always assume an initialization but might use the right hand side of Eq. (1.18) as a shorter notation. In fact, Eq. (1.19) is already simplified by assuming that all the $n_1 + \dots + n_k$ particles are initialized at the same time. In general, each of the $n_1 + \dots + n_k$ particles could be initialized at a different time.

The master equation (1.12) can be used to find a differential equation of the (conditional) probability generating function (1.19) by applying a time derivative to the latter and replacing the resulting time derivatives of the conditional probabilities by the expressions of the former:

$$\begin{aligned} \frac{d}{dt} \mathcal{M}(z, t) = & \sum_{\{m\}} \sum_{m' \neq m} \left(W(m|m') P(m', t | n, t_0) \right. \\ & \left. - W(m'|m) P(m, t | n, t_0) \right) z_1^{m_1} \dots z_k^{m_k}, \end{aligned} \quad (1.20)$$

where I abbreviated $m_1, \dots, m_k$ by m , $m'_1, \dots, m'_k$ by m' , and $n_1, \dots, n_k$ by n .

In the case of reaction-diffusion systems, we previously found in Eq. (1.17), that the transition rates of the conditional probabilities $W(m|m')$ are expressed in terms of binomial coefficients. For simplicity, I consider only one reaction in the system. Hence, for the *gain* term, $W(m|m')$ can be replaced by derivatives with respect to z and multiplications with z :

$$\begin{aligned} W(m|m') z_1^{m_1} \dots z_k^{m_k} &= r \binom{m'_1}{\ell_1} \dots \binom{m'_k}{\ell_k} z_1^{m_1} \dots z_k^{m_k} \\ &= r \left(\frac{z_1^{j_1}}{\ell_1!} \frac{d^{\ell_1}}{dz_1^{\ell_1}} \right) \dots \left(\frac{z_k^{j_k}}{\ell_k!} \frac{d^{\ell_k}}{dz_k^{\ell_k}} \right) z_1^{m'_1} \dots z_k^{m'_k}, \end{aligned} \quad (1.21)$$

where $m_i = m'_i - \ell_j + j_i$, i.e. the reaction allows a transition from state m' to state m , see Eq. (1.16). For the *loss* term, $W(m'|m)$ can also be expressed with powers of z and derivatives w.r.t. z :

$$\begin{aligned} W(m'|m)z_1^{m_1} \dots z_k^{m_k} &= r \binom{m_1}{\ell_1} \dots \binom{m_k}{\ell_k} z_1^{m_1} \dots z_k^{m_k} \\ &= r \left(\frac{z_1^{\ell_1}}{\ell_1!} \frac{d^{\ell_1}}{dz_1^{\ell_1}} \right) \dots \left(\frac{z_k^{\ell_k}}{\ell_k!} \frac{d^{\ell_k}}{dz_k^{\ell_k}} \right) z_1^{m_1} \dots z_k^{m_k} \end{aligned} \quad (1.22)$$

The important differences between Eq. (1.21) and Eq. (1.22) are the powers of z , both in the brackets and at the end.

Thus, the master equation, Eq. (1.20), can be expressed as a linear partial differential equation:

$$\frac{d}{dt} \mathcal{M}(z, t) = r \underbrace{\left(z_1^{j_1} \dots z_k^{j_k} - z_1^{\ell_1} \dots z_k^{\ell_k} \right) \left(\frac{1}{\ell_1!} \frac{d^{\ell_1}}{dz_1^{\ell_1}} \right) \dots \left(\frac{1}{\ell_k!} \frac{d^{\ell_k}}{dz_k^{\ell_k}} \right)}_{:= \mathcal{H}\left[z, \frac{d}{dz}\right] \dots \text{differential operator of the reaction}} \mathcal{M}(z, t) \quad (1.23)$$

If there are more types of reactions happening simultaneously and independently in the system, i.e. with different ℓ s and j s in Eq. (1.16), then their corresponding transition rates $W(\cdot|\cdot)$ are added, and ultimately, their differential operators are added in Eq. (1.23).

While expression (1.23) is for a general reaction and appears to be quite complicated, in many situations, it turns out to be short and simple. For an extinction process, a creation process and a coagulation process of a single reactant A , the differential operators are

$$\text{extinction} \quad A \xrightarrow{\epsilon} \emptyset \quad \epsilon(1-z) \frac{d}{dz} \quad (1.24)$$

$$\text{creation} \quad \emptyset \xrightarrow{\gamma} A \quad \gamma(z-1) \quad (1.25)$$

$$\text{coagulation} \quad 2A \xrightarrow{c} A \quad cz(1-z) \frac{d^2}{dz^2} \quad (1.26)$$

At this stage, it is worth pausing for a moment and think about Eq. (1.23). In the first bracket is a difference between monomials, which implies that the probability generating

function has a fixed point in its time evolution that is given by $z_1 = \dots = z_k = 1$. This can also be seen in the three explicit examples in Eqs. (1.24) to (1.26). What is the meaning of this fixed point? Going back to the definition of $\mathcal{M}(z, t)$ in Eq. (1.19), we see that $\mathcal{M}(1, t) = 1$ because a probability distribution is normalized. Hence, the existence of this fixed point simply means that the total probability is conserved.

It is reassuring that the conservation of probability is encoded in the PDE of the probability generating function. But it also hints at a redundancy in Eq. (1.23). This redundancy can be eliminated by shifting the variable z by 1 and introducing the new, so-called *Doi-shifted*, variable $\tilde{z} = z - 1$. Using this Doi-shifted variable, a different formulation of the model can be established, which will be used whenever it seems more convenient. At the level of generating functions, a corresponding Doi-shifted generating function can be defined as $\widetilde{\mathcal{M}}(\tilde{z}, t) := \mathcal{M}(\tilde{z} + 1, t)$, which obeys an analogous PDE:

$$\frac{d}{dt} \widetilde{\mathcal{M}}(\tilde{z}, t) = \underbrace{\mathcal{H} \left[\tilde{z} + 1, \frac{d}{d\tilde{z}} \right]}_{:= \widetilde{\mathcal{H}} \left[\tilde{z}, \frac{d}{d\tilde{z}} \right]} \widetilde{\mathcal{M}}(\tilde{z}, t), \quad (1.27)$$

with $dz = d\tilde{z}$. The operators of the example processes, extinction, creation and coagulation in Eqs. (1.24) to (1.26) become with the Doi-shift

$$\text{extinction} \quad A \xrightarrow{\epsilon} \emptyset \quad - \epsilon \tilde{z} \frac{d}{d\tilde{z}} \quad (1.28)$$

$$\text{creation} \quad \emptyset \xrightarrow{\gamma} A \quad \gamma \tilde{z} \quad (1.29)$$

$$\text{coagulation} \quad 2A \xrightarrow{c} A \quad - c \tilde{z} (\tilde{z} + 1) \frac{d^2}{d\tilde{z}^2}. \quad (1.30)$$

At this point, the original description, Eq. (1.23), and the Doi-shifted one, Eq. (1.27), are simply equivalent descriptions. Which of the two has an advantage over the other, is not clear yet. At a later point, I will show that the Doi-shift has advantages in many situations and I will therefore often include the Doi-shifted alternatives in the following derivations. But for now, I focus on the operators that can be applied to $\mathcal{M}$ and their interpretations.

The examples above do not include any spatial component. Given that I want to model reaction-*diffusion* systems, how can a process in physical space be included in such a theory? This answer is: using a discrete space. I demonstrate this with classical

diffusion because a random walker can be regarded as a discrete version of a diffusive particle. In Chapter 3, we will also encounter directed movements of particles.

Let the physical space be the d -dimensional lattice $h\mathbb{Z}^d$ with lattice constant h . For each coordinate $j \in \mathbb{Z}^d$ on the grid, a particle species A_j is introduced. In this discrete space, a random walk is a set of reactions that has one particle at site j as reactant and one particle at a neighboring site i as reaction product: $A_j \xrightarrow{D(h)} A_i$ with reaction rate $D(h)$, which is interpreted as a hopping rate. For a lattice, the conditional probability generating function has a variable z_j for every coordinate $j \in \mathbb{Z}^d$. All the random walker reactions (i.e. for all neighbors) for all the sites have to be summed up to obtain the differential operator for the random walk

$$\text{random walk } A_j \xrightarrow{D(h)} A_i \quad \frac{D(h)}{2} \sum_{j \in \mathbb{Z}^d} \sum_{\substack{i \in \mathbb{Z}^d \\ \text{with} \\ |i-j|=1}} (z_i - z_j) \left(\frac{d}{dz_j} - \frac{d}{dz_i} \right), \quad (1.31)$$

where the factor $1/2$ appears due to the symmetry between hopping from j to i and i to j . The term $|i - j|$ is the distance between position i and j . It is set to 1 for nearest neighbor hopping. In the Doi-shifted version, the operator representing the random walk has exactly the same functional form because $(z_i - z_j) = (\tilde{z}_i - \tilde{z}_j)$.

What is the (conditional) probability generating function good for? If it is known (e.g. by solving the linear PDE (1.23) or its Doi-shifted version Eq. (1.27)), all (conditional) moments, all (conditional) factorial moments and (conditional) probabilities are in principle accessible:

$$\ell\text{th moment} \quad \mathbb{E}[m^\ell | n] = \left(z \frac{d}{dz} \right)^\ell \mathcal{M}(z, t) \Big|_{z=1} \quad (1.32)$$

$$\ell\text{th factorial moment} \quad \mathbb{E}[(m)_\ell | n] = \frac{d^\ell}{dz^\ell} \mathcal{M}(z, t) \Big|_{z=1} \quad (1.33)$$

$$\text{probability} \quad P(m | n) = \frac{1}{m!} \frac{d^m}{dz^m} \mathcal{M}(z, t) \Big|_{z=0}, \quad (1.34)$$

where ℓ and m are used as multiindices such that $z^\ell = z_1^{\ell_1} \cdots z_k^{\ell_k}$ and $dz^m = dz_1^{m_1} \cdots dz_k^{m_k}$, and $(m)_\ell = m(m-1) \cdots (m-\ell+1)$ is the falling factorial. In particular, probabilities and factorial moments feature as coefficients of the Taylor expansion of $\mathcal{M}(z, t)$ at different points. The former at $z = 0$, the latter at $z = 1$. For the Doi-shifted gener-

ating function $\widetilde{\mathcal{M}}(\widetilde{z}, t)$, the probabilities appear as coefficients of the Taylor expansion at $\widetilde{z} = -1$, and the factorial moments are the coefficients of the Taylor expansion at $\widetilde{z} = 0$:

$$\widetilde{\mathcal{M}}(\widetilde{z}, t) = \sum_{\ell=0}^{\infty} \frac{\mathbb{E}[(m)_{\ell}|n]}{\ell!} \widetilde{z}^{\ell}. \quad (1.35)$$

As the modelling of stochastic processes using Doi-Peliti field theory lies in the intersection of mathematics and theoretical physics, we should remind ourselves how the different communities use different notation: $\langle m^{\ell} \rangle = \mathbb{E}[m^{\ell}]$. In Chapter 2, I will use the mathematical notation with $\mathbb{E}[\cdot]$ for expectations, whereas in Chapter 3, I will use the physics notation with $\langle \cdot \rangle$ for expectations. However, n -point correlation functions of the Doi-Peliti field theory will always be written with angular brackets.

By definition, all moments – if finite – can be derived from the probability distribution. However, here, we find that all moments can be used to recover all probabilities, because the (conditional) probability generating function is analytic. This statement is not true in general if the probability generating function is not analytic or if the range of the random variable is continuous.

Moments can be calculated from the factorial moments using Stirling numbers of the second kind

$$\mathbb{E}[m^{\ell}|n] = \sum_{k=0}^{\ell} \left\{ \begin{matrix} \ell \\ k \end{matrix} \right\} \mathbb{E}[(m)_k|n], \quad (1.36)$$

and vice versa, using the Stirling number of the first kind

$$\mathbb{E}[(m)_{\ell}|n] = \sum_{k=0}^{\ell} (-1)^{\ell-k} \left[\begin{matrix} n \\ k \end{matrix} \right] \mathbb{E}[m^{\ell}|n] \quad (1.37)$$

I use this relation in Sec. 2.4 to obtain the full probability distribution from the factorial moments of continuous-time branching processes.

Only the Stirling numbers of the second kind will be need, which are defined as:

$$\left\{ \begin{matrix} n \\ k \end{matrix} \right\} := \frac{1}{k!} \sum_{i=0}^k (-1)^i \binom{k}{i} (k-i)^n \quad (1.38)$$

$$(1.39)$$

I formally solve Eq. (1.23) in order to extract moments using the operators in Eq. (1.32). The formal solution of Eq. (1.23) is

$$\mathcal{M}(z, t) = e^{\mathcal{H}t} \mathcal{M}(z, 0) \quad (1.40)$$

What are typical initial conditions for the (conditional) probability generating function? If the system is initiated with one particle at time $t_0 = 0$, then

$$\mathcal{M}(z, 0) = z, \quad (1.41)$$

if it is initiated with k particles, then

$$\mathcal{M}(z, 0) = z^k. \quad (1.42)$$

Probabilistic superpositions of initialized states, similar to quantum mechanical situations like in the Stern-Gerlach experiment [56], are also realisable. For example, if the system contains k particles with probability p_k , then

$$\mathcal{M}(z, 0) = \sum_{k=0}^{\infty} p_k z^k \quad (1.43)$$

Initializations, like Eq. (1.41), time evolutions like Eq. (1.40), derivation of moments, like Eq. (1.32) can all be regarded as operators acting on the ground state of the system, i.e. the empty system $\mathcal{M}(z, t) = 1$. Alternatively, I could have formally solved the Doi-shifted PDE, Eq. (1.27), to find $\widetilde{\mathcal{M}}(\widetilde{z}, t) = e^{\widetilde{\mathcal{H}}t} \widetilde{\mathcal{M}}(\widetilde{z}, 0)$. Then, conditions such as $\widetilde{\mathcal{M}}(\widetilde{z}, 0) = \widetilde{z}$ initialize the system with specific values of its factorial moments. However, interpreting specific values of factorial moments is not easy and I therefore keep the non-Doi-shifted viewpoint when considering initializations of the system.

This idea of formulating everything explicitly in terms of operators can be achieved with a simplified notation, the second quantization.

1.4 SECOND QUANTIZED EQUATION

The language of second quantization shifts the focus of operators from the point of view of analysis to a viewpoint which readily allows interpreting the action of operators in terms of particle numbers. It was originally introduced in quantum mechanics by *P. Dirac* in 1939 [35], where the focus shifted to energy eigenstates rather than particles numbers.

First, I define the *ladder operators**

$$a := \frac{d}{dz} \quad \text{and} \quad a^\dagger := z. \quad (1.44)$$

and the ground state, which represents a system with zero particles

$$|0\rangle := 1. \quad (1.45)$$

The ladder operators do not commute, i.e.

$$1 = aa^\dagger - a^\dagger a =: [a, a^\dagger]. \quad (1.46)$$

States which include more particles can be created by applying $a^\dagger$ to the ground state, which is therefore called creation operator. The state with k particles is written as

$$|k\rangle := a^{\dagger k} |0\rangle = z^k, \quad (1.47)$$

analogously to Eq. (1.42). The vectors $|k\rangle$ are called *ket*-vectors. Thus, the action of

*These definitions of ladder operators are specific to the context of Doi-Peliti field theory. In other contexts, ladder operators have different definitions. Example: for the quantum harmonic oscillator, the ladder operators are defined as $a = \sqrt{\frac{m\omega}{2\hbar}} \left(x + \frac{\hbar}{m\omega} \frac{d}{dx} \right)$ and $a^\dagger = \sqrt{\frac{m\omega}{2\hbar}} \left(x - \frac{\hbar}{m\omega} \frac{d}{dx} \right)$, where m is the mass of the particle, ω is the frequency of the oscillator and $\hbar$ is the reduced Planck constant. They act on wave functions $|n\rangle = (2^n n!)^{-1/2} (m\omega/\pi\hbar)^{1/4} e^{-m\omega x^2/2\hbar} H_n(x\sqrt{m\omega/\hbar})$, where H_n is the n th Hermite polynomial. [153]

the other ladder operator can be derived as

$$a|k\rangle = k|k-1\rangle, \quad (1.48)$$

and it is therefore called annihilation operator, because it reduces the number of particles in the system. In addition, the Doi-shifted creation operator is defined as $\tilde{a} = a^\dagger - 1$. The probability generating function $\mathcal{M}(z, t)$, Eq. (1.40), is expressed as linear combinations of the *ket*-vectors with time dependent coefficients:

$$\mathcal{M}(z, t) = \sum_{m=0}^{\infty} P(m, t|n, 0)|m\rangle =: |\mathcal{M}(t)\rangle, \quad (1.49)$$

where n is the initial state at time $t_0 = 0$.

The vector space over $\mathbb{C}$ spanned by the states $|k\rangle$ is the space of analytic functions in z . Given that the *ket*-vectors $|0\rangle, |1\rangle, |2\rangle, \dots$ form a basis, its dual basis, spanning the dual space, can be formally identified as $\langle 0|, \langle 1|, \langle 2|, \dots$, defined by their action such that

$$\langle k|n\rangle = \delta_{k,n}. \quad (1.50)$$

The dual vectors $\langle k|$ are called *bra*-vectors. On a compact domain, the functions $|n\rangle$ belong to a complex Hilbert space with inner product

$$\langle g|f\rangle = \int \overline{g(z)}f(z)dz, \quad (1.51)$$

where $\overline{g(z)}$ is the complex conjugate of the function $g(z)$.

Since $|n\rangle$ represents the function z^n , by the Fréchet-Riesz representation theorem [47, 134], the *bra*-vector $\langle k|$ can be represented as an integral [120]

$$\frac{(-1)^k}{k!} \int \delta^{(k)}(z)z^n dz = \delta_{k,n} = \langle k|n\rangle, \quad (1.52)$$

where $\delta^{(k)}(z)$ is the k th derivative of the Dirac delta-function. The *bra*-vectors are evaluating functions and their derivatives at $z = 0$. Hence, the probabilities $P(m, t|n, 0)$

can be found by using a and $\langle 0|$:

$$P(m, t|n, 0) = \frac{1}{m!} \langle 0|a^m|\mathcal{M}(t)\rangle = \langle m|\mathcal{M}(t)\rangle. \quad (1.53)$$

However, in order to find moments and factorial moments, the probability generating function and its derivatives have to be evaluated at $z = 1$, Eqs. (1.32) and (1.33). How can an evaluation at $z = 1$ be achieved? If a function $f(z)$ is analytic in z , then a complete Taylor series has an infinite radius of convergence and is exact at any evaluation point. Hence

$$\sum_{k=0}^{\infty} \langle k|f\rangle = \sum_{k=0}^{\infty} \frac{(-1)^k}{k!} \int \delta^{(k)}(z) f(z) dz \quad (1.54)$$

$$= \sum_{k=0}^{\infty} \frac{f^{(k)}(0)}{k!} = f(1) = \int \delta(z-1) f(z) dz \quad (1.55)$$

Given that $\mathcal{M}(z, t)$, as a probability generating function, Eq. (1.19), is analytic in z (but possibly not in time t), and since the multiplication operator $a^\dagger = z \cdot$ and differentiation operator $a = d/dz$ do not change the analyticity, the ℓ th moment can be expressed in its second quantized form as

$$\sum_{k=0}^{\infty} \langle k|(a^\dagger a)^\ell|\mathcal{M}(t)\rangle = \mathbb{E}[m^\ell|n] = \left(z \frac{d}{dz}\right)^\ell \mathcal{M}(z, t) \Big|_{z=1}, \quad (1.56)$$

and the n th factorial moment can be expressed as

$$\sum_{k=0}^{\infty} \langle k|a^\ell|\mathcal{M}(t)\rangle = \mathbb{E}[(m)_\ell(t)|n] = \frac{d^\ell}{dz^\ell} \mathcal{M}(z, t) \Big|_{z=1}. \quad (1.57)$$

The evaluation operator $\sum_k \langle k|$ for evaluating a function at $z = 1$ is also called *abyss* [128] and is abbreviated by

$$\sum_{k=0}^{\infty} \langle k| = \langle 0|e^a =: \langle \star|. \quad (1.58)$$

If I wanted to evaluate an analytic function $f(z)$ at any other point $\xi \in \mathbb{C}$, Eq. (1.54)

could be adjusted such that, in second quantized form

$$\sum_{k=0}^{\infty} \langle k | \xi^k | f \rangle = f(\xi). \quad (1.59)$$

In order to complete the translation from the probability generating function, Eq. (2.7) to a second quantized representation, I have to understand how the system is initialized. If there is one particle in the system at time $t_0 = 0$, then the probability generating function is $\mathcal{M}(z, 0) = z$, because $P(n, 0) = \delta_{n,1}$, Eq. (1.41). In second quantized form, this is represented by $|1\rangle = a^\dagger|0\rangle = |\mathcal{M}(0)\rangle$. Analogously, if the system is initialized with k particles, then $\mathcal{M}(z, 0) = z^k$, Eq. (1.42), because $P(n, 0) = \delta_{n,k}$, which is represented by $|k\rangle = a^{\dagger k}|0\rangle = |\mathcal{M}(0)\rangle$.

While the transition to a second quantized form may be regarded as completed at this stage, the time evolution of the probability distribution, Eq. (1.12), and the probability generating function, Eq. (1.40), is still unsolved. Although the second quantised form of the master equation (1.23) is a complicated differential equation in z , it is a first order, linear differential equation in time t . Hence, a formal solution of the time evolution is, analogously to Eq. (1.40)

$$|\mathcal{M}(t)\rangle = e^{\mathcal{H}t}|\mathcal{M}(0)\rangle, \quad (1.60)$$

where the differential operator of the reaction $\mathcal{H} = \mathcal{H}[a, a^\dagger]$ is expressed using the ladder operators a and $a^\dagger$ and is called Hamiltonian. In the following, I will always assume that the Hamiltonian is *normal ordered*, i.e. it can be written as a power series, where each term has the creation operators on the left and the annihilation operators on the right:

$$H[a, a^\dagger] = \sum_{\gamma, \sigma} h_{\gamma, \sigma} a^{\dagger \gamma} a^\sigma, \quad (1.61)$$

with $h_{\gamma, \sigma} \in \mathbb{R}$, $\gamma, \sigma \in \mathbb{N}_0$. For reaction-diffusion systems, this is always possible, see Eq. (1.23). When $a^\dagger$ is replaced by $\tilde{a}$, a corresponding Doi-shifted Hamiltonian operator $\tilde{\mathcal{H}}[\tilde{a}, a]$ is found.

In general, calculating the exponential of the Hamiltonian $\mathcal{H}$ is difficult and I will

introduce the path integral to achieve it perturbatively.[†]

From here, a way forward is the consideration of the left and right eigenfunctions of the ladder operators a and $a^\dagger$, called *coherent states*. The right eigenfunctions of operator a and the left eigenfunctions of operator $a^\dagger$ are

$$|\phi\rangle := e^{\phi a^\dagger} |0\rangle \quad \langle\phi^\dagger| := \langle 0| e^{\phi^\dagger a}, \quad (1.62)$$

respectively, with arbitrary eigenvalues $\phi, \phi^\dagger \in \mathbb{C}$, i.e. $a|\phi\rangle = \phi|\phi\rangle$ and $\langle\phi^\dagger|a^\dagger = \langle\phi^\dagger|\phi^\dagger$. There are no left eigenfunctions for a , and no right eigenfunctions for $a^\dagger$.[‡] The *bra*-vector $\langle\phi^\dagger|$ is also an eigenvector of the Doi-shifted operator $\tilde{a}$, however, its eigenvalue is $\phi^\dagger - 1 =: \tilde{\phi}$. The eigenvalues $\phi, \phi^\dagger, \tilde{\phi}$ must not be functions of z but can be functions of t , which will be used in the following.

Since $|0\rangle, |1\rangle, \dots$ is an orthonormal basis, an identity operator can be expressed using them

$$\mathbb{I} = \sum_{k=0}^{\infty} |k\rangle\langle k|. \quad (1.63)$$

This identity can be also rewritten using coherent states:

$$\mathbb{I} = \int \frac{d\text{Re}\phi d\text{Im}\phi}{\pi} e^{-\phi^\dagger \phi} |\phi\rangle\langle\phi^\dagger|, \quad (1.64)$$

where $\text{Re}\phi$ and $\text{Im}\phi$ are the real and imaginary parts of ϕ , and $\phi^\dagger$ is the complex conjugate of ϕ . Eq. (1.64) is shown in the appendix, Sec. A.1. This identity operator can be superficially made time-dependent: Eq. (1.64) is true even when $\phi = \phi(t)$ are time-dependent functions. This identity will be denoted by $\mathbb{I}_t$.

[†]For comparison: the time evolution operator in quantum mechanics has the form $e^{-i\mathcal{H}t}$ and is used to calculate probability *amplitudes*.

[‡]This can be seen using the following argument: suppose $|\chi\rangle$ is a right eigenfunction of $a^\dagger$, then it can be written as a linear combination of the basis vectors $|n\rangle$, i.e. $|\chi\rangle = \sum_{n \in \mathbb{N}_0} b_n |n\rangle$. Let $n^* \in \mathbb{N}_0$ be the smallest $n \in \mathbb{N}_0$ such that $b_{n^*} \neq 0$, then the vector $a^\dagger |\chi\rangle$ can be written as a linear combination of the basis $|n\rangle$ as well, i.e. $|\chi\rangle = \sum_{n \in \mathbb{N}} c_n |n\rangle$, with $c_n = b_{n-1}$. However, here $|n^*\rangle$ is not part of this representation as $c_{n^*} = b_{n^*-1} = 0$. Hence, there cannot be such a right eigenfunction $|\chi\rangle$ of $a^\dagger$. An analogous argument dismisses the existence of a left eigenfunction for a .

1.5 PATH INTEGRAL DERIVATION

In this section, I follow the lecture notes by *John Cardy* [22] and *Gunnar Pruessner* [128]. Originally, the path integral of the Doi-Peliti field theory was derived by *Luca Peliti* in 1985 [120].

1.5.1 THE EXPONENTIAL OF THE HAMILTONIAN OPERATOR

The formal solution to the second quantised form of the master equation is Eq. (1.60), which is the exponential of the Hamiltonian operator $\mathcal{H}[a, a^\dagger]$. The exponential can be discretized in time by introducing time steps of length Δt such that

$$e^{\mathcal{H}t} = \lim_{\Delta t \rightarrow 0} (1 + \Delta t \mathcal{H})^{\frac{t}{\Delta t}} = \lim_{\Delta t \rightarrow 0} \mathbb{I}_t \prod_{t'=0}^{t-\Delta t} (1 + \Delta t \mathcal{H}) \mathbb{I}_{t'}, \quad (1.65)$$

where in the last step, identities were inserted between the factors of $(1 + \Delta t \mathcal{H})$. In the notation above, each factor contains an identity operator on the right. I reorder the notation of the product such that each identity is split up notationally into the *bra*-vector part and the *ket*-vector part:

$$e^{\mathcal{H}t} = \lim_{\Delta t \rightarrow 0} \int \cdots \int |\phi(t)\rangle \left(\prod_{t'=0}^{t-\Delta t} \underbrace{\langle \phi^\dagger(t' + \Delta t) | (1 + \Delta t \mathcal{H}) | \phi(t') \rangle}_{=: \Xi(t')} \right) \langle \phi^\dagger(0) | \quad (1.66)$$

$$e^{-\phi^\dagger(t)\phi(t)} \cdots e^{-\phi^\dagger(0)\phi(0)} \frac{d\text{Re}\phi(t) d\text{Im}\phi(t)}{\pi} \cdots \frac{d\text{Re}\phi(0) d\text{Im}\phi(0)}{\pi},$$

where I grouped the factors $e^{-\phi^\dagger(t')\phi(t')}$ because they commute with $\mathcal{H}$. An analogous express is found for the Doi-shifted evolution operator $e^{\tilde{\mathcal{H}}t}$, where $\mathcal{H}$ is replaced by $\tilde{\mathcal{H}}$, and $\tilde{\Xi}(t')$ is analogously defined to $\Xi(t')$. The terms $\Xi(t')$ and $\tilde{\Xi}(t')$ are analysed in the following:

1. The term $\langle \phi^\dagger(t' + \Delta t) | \phi(t') \rangle$ is equal to

$$\langle \phi^\dagger(t' + \Delta t) | \phi(t') \rangle = \langle 0 | e^{\phi^\dagger(t' + \Delta t)a} | \phi(t') \rangle \quad (1.67)$$

$$= \langle 0 | \sum_{k=0}^{\infty} \frac{\phi^{\dagger k}(t' + \Delta t)}{k!} a^k | \phi(t') \rangle \quad (1.68)$$

$$= \langle 0 | \sum_{k=0}^{\infty} \frac{\phi^{\dagger k}(t' + \Delta t)}{k!} \phi^k(t') | \phi(t') \rangle \quad (1.69)$$

$$= e^{\phi^\dagger(t' + \Delta t)\phi(t')}, \quad (1.70)$$

where from line (1.68) to line (1.69), I used that $|\phi(t')\rangle$ is an eigenvector of a with eigenvalue $\phi(t')$.

2. Assuming that the Hamiltonian operator $\mathcal{H}$ is normal ordered, each of its terms will be of the form $a^{\dagger\gamma}a^\sigma$ for some $\gamma, \sigma \in \mathbb{N}_0$. Hence, $\langle \phi^\dagger(t' + \Delta t) | a^{\dagger\gamma}a^\sigma | \phi(t') \rangle$ is equal to

$$\langle \phi^\dagger(t' + \Delta t) | a^{\dagger\gamma}a^\sigma | \phi(t') \rangle = \phi^{\dagger\gamma}(t' + \Delta t) \phi^\sigma(t') \langle \phi^\dagger(t' + \Delta t) | \phi(t') \rangle \quad (1.71)$$

$$= \phi^{\dagger\gamma}(t' + \Delta t) \phi^\sigma(t') e^{\phi^\dagger(t' + \Delta t)\phi(t')}, \quad (1.72)$$

where I used that $\langle \phi^\dagger(t' + \Delta t) |$ is an eigenvector of $a^\dagger$, and $|\phi(t')\rangle$ is an eigenvector of a , and Eq. (1.70). If $\mathcal{H} = \mathcal{H}[a^\dagger, a]$ is normal ordered, then each $a^\dagger$ is replaced by $\phi^\dagger(t' + \Delta t)$ and each a is replaced by $\phi(t')$, resulting in $\mathcal{H}[\phi^\dagger(t' + \Delta t), \phi(t')]$. Particularly noteworthy is, that the complex functions $\phi^\dagger(t)$ and $\phi(t)$ commute, i.e. $[\phi, \phi^\dagger] = 0$.

If the Doi-shifted Hamiltonian operator $\tilde{\mathcal{H}}[\tilde{a}, a]$ is used, the arguments are almost identical. The difference is that $\langle \phi^\dagger(t' + \Delta t) |$ is an eigenvector of $\tilde{a}$ with eigenvalue $\phi^\dagger(t' + \Delta t) - 1 = \tilde{\phi}(t' + \Delta t)$, see the comment after Eq. (1.62). Thus,

$$\langle \phi^\dagger(t' + \Delta t) | \tilde{\mathcal{H}}[\tilde{a}, a] | \phi(t') \rangle = \tilde{\mathcal{H}}[\tilde{\phi}(t' + \Delta t), \phi(t')] e^{\phi^\dagger(t' + \Delta t)\phi(t')}. \quad (1.73)$$

3. The function $\Xi(t')$ is equal to

$$\Xi(t') = e^{\phi^\dagger(t' + \Delta t)\phi(t')} (1 + \Delta t \mathcal{H}[\phi^\dagger(t' + \Delta t), \phi(t')]), \quad (1.74)$$

for the non-Doi-shifted PDE, Eq (1.60) and for the Doi-shifted one $\tilde{\Xi}$ equals

$$\tilde{\Xi}(t') = e^{\phi^\dagger(t'+\Delta t)\phi(t')} \left(1 + \Delta t \tilde{\mathcal{H}}[\tilde{\phi}(t' + \Delta t), \phi(t')] \right). \quad (1.75)$$

Hence, the exponential of the Hamiltonian operator can be written as

$$e^{\mathcal{H}t} = \lim_{\Delta t \rightarrow 0} \int \dots \int e^{-\phi^\dagger(t)\phi(t)} \dots e^{-\phi^\dagger(0)\phi(0)} | \phi(t) \rangle \left(\prod_{t'=0}^{t-\Delta t} e^{\phi^\dagger(t'+\Delta t)\phi(t')} (1 + \Delta t \mathcal{H}[\phi^\dagger(t' + \Delta t), \phi(t')]) \right) \langle \phi^\dagger(0) | \quad (1.76)$$

$$\frac{d\text{Re}\phi(t) d\text{Im}\phi(t)}{\pi} \dots \frac{d\text{Re}\phi(0) d\text{Im}\phi(0)}{\pi} \\ = \lim_{\Delta t \rightarrow 0} \int \dots \int \frac{d\text{Re}\phi(t) d\text{Im}\phi(t)}{\pi} \dots \frac{d\text{Re}\phi(0) d\text{Im}\phi(0)}{\pi} \\ e^{-\phi^\dagger(t)\phi(t) + \sum_{t'=0}^{t-\Delta t} (\phi^\dagger(t'+\Delta t) - \phi^\dagger(t'))\phi(t')} \quad (1.77) \\ | \phi(t) \rangle \left(\prod_{t'=0}^{t-\Delta t} (1 + \Delta t \mathcal{H}[\phi^\dagger(t' + \Delta t), \phi(t')]) \right) \langle \phi^\dagger(0) |,$$

Next, I take the limit $\Delta t \rightarrow 0$. What happens to the different terms?

1. The product over t' converges to an exponential again, see Eq. (1.65):

$$\lim_{\Delta t \rightarrow 0} \prod_{t'=0}^{t-\Delta t} (1 + \Delta t \mathcal{H}[\phi^\dagger(t' + \Delta t), \phi(t')]) = e^{\int_0^t \mathcal{H}[\phi^\dagger(t'), \phi(t')] dt'} \quad (1.78)$$

2. The sum over t' becomes a Riemann integral:

$$\lim_{\Delta t \rightarrow 0} \sum_{t'=0}^{t-\Delta t} (\phi^\dagger(t' + \Delta t) - \phi^\dagger(t')) \phi(t') = \int_0^t \left(\frac{d}{dt'} \phi^\dagger(t') \right) \phi(t') dt' \quad (1.79)$$

3. The measures become a path integral measure:

$$\lim_{\Delta t \rightarrow 0} \int \dots \int \frac{d\text{Re}\phi(t) d\text{Im}\phi(t)}{\pi} \dots \frac{d\text{Re}\phi(0) d\text{Im}\phi(0)}{\pi} = \int \mathcal{D}[\phi^\dagger, \phi] \quad (1.80)$$

Thus, the exponential of the Hamiltonian operator has been rewritten as

$$e^{\mathcal{H}t} = \int \mathcal{D}[\phi^\dagger, \phi] e^{-\phi^\dagger(t)\phi(t) + \int_0^t \left(\frac{d}{dt'} \phi^\dagger(t') \right) \phi(t') + \mathcal{H}[\phi^\dagger(t'), \phi(t')] dt'} |\phi(t)\rangle \langle \phi^\dagger(0)|, \quad (1.81)$$

and an integration by parts results in

$$e^{\mathcal{H}t} = \int \mathcal{D}[\phi^\dagger, \phi] e^{-\phi^\dagger(0)\phi(0) + \int_0^t \overbrace{\phi^\dagger(t') \left(-\frac{d}{dt'} \phi(t') \right) + \mathcal{H}[\phi^\dagger(t'), \phi(t')]}^{=: \mathcal{A}'[\phi^\dagger, \phi]} dt'} |\phi(t)\rangle \langle \phi^\dagger(0)|, \quad (1.82)$$

where $\mathcal{A}'[\phi^\dagger, \phi]$ is introduced as the (non-Doi-shifted) action.

Alternatively, the function $\phi^\dagger$ can be replaced by $\phi^\dagger = \tilde{\phi} + 1$ (*Doi-shift*), and $\mathcal{H}$ by $\tilde{\mathcal{H}}$ to find

$$e^{\tilde{\mathcal{H}}t} = \int \mathcal{D}[\phi^\dagger, \phi] e^{-\phi(t) - \tilde{\phi}(0)\phi(0) + \int_0^t \overbrace{\tilde{\phi}(t') \left(-\frac{d}{dt'} \phi(t') \right) + \tilde{\mathcal{H}}[\tilde{\phi}(t'), \phi(t')]}^{=: \mathcal{A}[\tilde{\phi}, \phi]} dt'} |\phi(t)\rangle \langle \phi^\dagger(0)|, \quad (1.83)$$

where $\mathcal{A}[\tilde{\phi}, \phi]$ is introduced as the Doi-shifted action functional, i.e. it the dimensionless quantity whose exponential $e^{\tilde{\mathcal{A}}[\tilde{\phi}, \phi]}$ is the likelihood of the realization ϕ .

Both time evolution operators and $e^{\mathcal{H}t}$ and $e^{\tilde{\mathcal{H}}t}$ are the same; they are simply written in different representations. Because the Doi-shifted action $\mathcal{A}$ will be used almost everywhere, I use the notation $\mathcal{A}$ instead of the more intuitiv one $\tilde{\mathcal{A}}$. The non-Doi-shifted action will be rarely used and is therefore given the notation $\mathcal{A}'$.

1.5.2 THE INITIALIZATION

The system is initialized by setting a particle distribution at $t_0 = 0$, see Eqs. (1.41), (1.42) and (1.43), represented by $|\mathcal{M}(0)\rangle$. Either the time evolution operator $e^{\mathcal{H}t}$, Eq. (1.82), is applied to this initial state, or $|\mathcal{M}(0)\rangle$ is Doi-shifted and the time evolution operator $e^{\tilde{\mathcal{H}}t}$, Eq. (1.83), is applied to the shifted initialization. Both $e^{\mathcal{H}t}$ and $e^{\tilde{\mathcal{H}}t}$ have the *bra*-vector $\langle \phi^\dagger(0)|$ on their right side. Hence, to transfer the initialization of the system from the operator viewpoint to the field-theory viewpoint, $\langle \phi^\dagger(0)|\mathcal{M}(0)\rangle$ has to be calculated. Three cases are considered

- The system is initialized with k particles at time t_0 . Therefore, $|\mathcal{M}(0)\rangle = |k\rangle$, and the initialization is equal to

$$\langle \phi^\dagger(0) | \mathcal{M}(0) \rangle = \phi^{\dagger k}(0) = \left(\tilde{\phi}(0) + 1 \right)^k. \quad (1.84)$$

- The system is initialized with a particle distribution p_k at time $t_0 = 0$. Here, $|\mathcal{M}(0)\rangle = \sum_k p_k |k\rangle$, and the initialization is equal to

$$\langle \phi^\dagger(0) | \mathcal{M}(0) \rangle = \sum_{k=0}^{\infty} p_k \phi^{\dagger k}(0) = \sum_{k=0}^{\infty} p_k \left(\tilde{\phi}(0) + 1 \right)^k. \quad (1.85)$$

- The system is initialized with several particles at different times. This situation is more difficult because the particles which have been initialized earlier evolve according to the stochastic process before the other particles are initialized. This problem is addressed in Sec. 1.5.4.

Thus, after applying $e^{\mathcal{H}t}$ or $e^{\tilde{\mathcal{H}}t}$ to the initial state $|\mathcal{M}(0)\rangle$, the result is $|\mathcal{M}(t)\rangle$ or $|\tilde{\mathcal{M}}(t)\rangle$, respectively. These states are the same, there are simply written in a different basis.

1.5.3 THE OBSERVABLE

In this section, the advantage of using the Doi-shift will become apparent. Up until now, the Doi-shifted versions of observables, generating functions, time evolution operators and actions, simply represented an alternative way of writing the same objects. However, when calculating moments and factorial moments, the Doi-shift makes practical computations easier, as I will show in the following.

Observables are typically moments or factorial moments, which are measured using the number operator $a^\dagger a$ or higher powers of it. However, the general case is that the observable is a functional $\mathcal{O}$ of the probability generating function $\mathcal{M}(z, t)$. I assume, this functional can be expressed as an operator series of $z^\alpha \frac{d^\beta}{dz^\beta}$ applied to $\mathcal{M}(z, t)$ and

evaluated at $z = 1$:

$$\mathcal{O}[\mathcal{M}(z, t)] = \sum_{\alpha, \beta} c_{\alpha, \beta} z^\alpha \frac{d^\beta}{dz^\beta} \mathcal{M}(z, t) \Big|_{z=1}. \quad (1.86)$$

Hence, it can be represented by some combination of the ladder operators $a^\dagger$ and a , which is assumed to be normal ordered. Then the operator of the observable is a sum of terms of the form $a^{\dagger\alpha} a^\beta$ with $\alpha, \beta \in \mathbb{N}_0$. For moments and factorial moments, the evaluation of the probability generating function $|\mathcal{M}(t)\rangle$ at $z = 1$ was realised by applying the evaluation operator $\langle \star |$, Eq. (1.58). Therefore, $\langle \star | a^{\dagger\alpha} a^\beta | \phi(t) \rangle$ has to be evaluated:

$$\left(z^\alpha \frac{d^\beta}{dz^\beta} \mathcal{M}(z, t) \right) \Big|_{z=1} = \langle \star | a^{\dagger\alpha} a^\beta | \phi(t) \rangle = \phi^\beta(t) e^{\phi(t)}, \quad (1.87)$$

where I used that $\langle \star |$ is an eigenvector of $a^\dagger$ with eigenvalue 1 and $|\phi(t)\rangle$ is an eigenvector of a with eigenvalue $\phi(t)$. The remaining term was already found in Eq. (1.70).

The *Doi-shift advantage*: If I write the time evolution operator in the Doi-shifted version, the exponential $e^{\phi(t)}$ cancels with the exponential $e^{-\phi(t)}$ in $e^{\tilde{\mathcal{H}}t}$, see Eq. (1.83):

$$\langle \star | a^{\dagger\alpha} a^\beta e^{\tilde{\mathcal{H}}t} = \int \mathcal{D}[\phi^\dagger, \phi] \phi^\beta(t) e^{\mathcal{A}[\tilde{\phi}, \phi]} \langle \phi^\dagger(0) | \quad (1.88)$$

In particular, if I assume that the system was initialized with k particles at time $t_0 = 0$, i.e. $\mathcal{M}(z, 0) = z^k$, the observable equals

$$\langle \star | a^{\dagger\alpha} a^\beta e^{\tilde{\mathcal{H}}t} | \mathcal{M}(0) \rangle = \int \mathcal{D}[\phi^\dagger, \phi] \phi^\beta(t) e^{\mathcal{A}[\tilde{\phi}, \phi]} \phi^{\dagger k}(0) =: \langle \phi^\beta(t) \phi^{\dagger k}(0) \rangle, \quad (1.89)$$

where I used the result in Eq. (1.84) and introduced a shorthand notation for the path integral expression in the last equality. The number α does not play a rôle in this calculation.

The *disadvantage when not using the Doi-shift*: If the Doi-shift is not used in the time evolution operator, the result is still given by Eq. (1.87). However, the factor $e^{\phi(t)}$ does

not cancel with anything in $e^{\mathcal{H}t}$. Hence, the expectation of the observable is

$$\begin{aligned}\langle \star | a^{\dagger\alpha} a^{\beta} e^{\mathcal{H}t} | \mathcal{M}(0) \rangle &= \sum_{\ell=0}^{\infty} \frac{1}{\ell!} \int \mathcal{D}[\phi^{\dagger}, \phi] \phi^{\beta+\ell}(t) e^{\mathcal{A}'[\phi^{\dagger}, \phi]} \phi^{\dagger k}(0) \\ &=: \sum_{\ell=0}^{\infty} \frac{1}{\ell!} \langle \phi^{\beta+\ell}(t) \phi^{\dagger k}(0) \rangle_{\text{no-Doi}},\end{aligned}\tag{1.90}$$

where $\langle \dots \rangle_{\text{no-Doi}}$ represents the path integral based on the non-Doi-shifted action.

More explicitly, if I want to calculate the expected number of particles at time t , given that there were k particles at time $t_0 = 0$, then I have to calculate:

$$\mathbb{E}[n, t | k, 0] = \langle \star | a^{\dagger} a e^{\tilde{\mathcal{H}}t} | k \rangle = \langle \phi(t) \phi^{\dagger k}(0) \rangle \tag{1.91}$$

$$= \langle \star | a^{\dagger} a e^{\mathcal{H}t} | k \rangle = \sum_{k=0}^{\infty} \frac{1}{k!} \langle \phi^{1+\ell}(t) \phi^{\dagger k}(0) \rangle_{\text{no-Doi}}. \tag{1.92}$$

This indicates that using the Doi-shift can reduce the number of terms that have to be calculated. This advantage become more apparent in Sec. 1.5.7 on Feynman diagrams.

From now on, I will always assume that the Doi-shift is used and that $\langle \star |$ is used as the *bra*-vector.

The expression in Eq. (1.89) to an initialization at time t_0 and the measurement of an observable at time t . But what if the observable requires evaluations at several times? How can the system be observed at one time, then let to continue to evolve and then be observed again at a later time? Does observing the system change the system, like in quantum mechanics? These problems are addressed in the following, in Sec. 1.5.4.

1.5.4 INITIALIZING AND OBSERVING AT SEVERAL TIMES

In this section, I explain how more initializations and observations at several times can be treated within the path integral.

Adding a particle to a system that has already evolved from an earlier initialization of particles is achieved at the level of the probability generating function $\mathcal{M}(z, t)$ by multiplying with z , see Eq. (1.42) and Eq. (1.47). If, just prior to the creation of a particle at time t , the probability for n particles in the system was $P(n, t' \rightarrow t_1^-)$, then

at time t_1 it is equal to the probability for $n + 1$ particles in the system:

$$\text{adding a particle at time } t_1 \quad P(n + 1, t_1) = \lim_{t' \rightarrow t_1^-} P(n, t'). \quad (1.93)$$

For the entire probability generating function, this means

$$\text{adding a particle at time } t_1 \quad \mathcal{M}(z, t_1) = z \cdot \lim_{t' \rightarrow t_1^-} \mathcal{M}(z, t'), \quad (1.94)$$

which is written in the language of second quantisation as

$$\text{adding a particle at time } t_1 \quad |\mathcal{M}(t_1)\rangle = a^\dagger \cdot \lim_{t' \rightarrow t_1^-} |\mathcal{M}(t')\rangle. \quad (1.95)$$

Now that I have expressed several initializations, how do they eventually link to the observable? If the expectation of an observable $\mathcal{O}$ is calculated at a time $t_2 > t_1$ after the latest part of the initialization, the state of the system continues to evolve from t_1 to t_2 :

$$|\mathcal{M}(t_2)\rangle = e^{\tilde{\mathcal{H}}(t_2-t_1)} a^\dagger e^{\tilde{\mathcal{H}}t_1} |\mathcal{M}(t_0)\rangle \quad (1.96)$$

In the path integral formulation, the $a^\dagger$ acts on $|\phi(t_1)\rangle$ on its right, and it acts on $\langle\phi^\dagger(t_1)|$ on its left, see Eq. (1.83). Since $\langle\phi^\dagger(t_1)|$ is an eigenvector of $a^\dagger$, this produces a factor $\phi^\dagger(t_1)$. The remaining term $\langle\phi^\dagger(t_1)|\phi(t_1)\rangle$ was found in Eq. (1.70) to be equal to $e^{\phi^\dagger(t_1)\phi(t_1)}$. After the *Doi* shift and the integration by parts, see step from Eq. (1.81) to Eq. (1.83), $e^{\phi^\dagger(t_1)\phi(t_1)}$ cancels with the term produced by the intergration by parts. Finally, the integrals in the exponential, one from t_0 to t_1 and the other one from t_1 to t_2 merge:

$$|\mathcal{M}(t_2)\rangle = \int \mathcal{D}[\phi^\dagger, \phi] e^{-\phi(t_2) - \tilde{\phi}(t_0)\phi(t_0) + \int_{t_0}^{t_2} \tilde{\phi}(t') \left(-\frac{d}{dt'} \phi(t')\right) + \tilde{\mathcal{H}}[\tilde{\phi}(t'), \phi(t')] dt'} \quad (1.97)$$

$$|\phi(t_2)\rangle \phi^\dagger(t_1) \langle\phi^\dagger(t_0)| \mathcal{M}(t_0)\rangle,$$

which illustrates how to incorporate several initialization times.

Using this insight, I can ‘initialize’ the system with zero particles at time $t_0 = -\infty$, let it evolve and then implement a ‘real’ initialization at time $t_1 = 0$, i.e. with particles. If

I assume that my system is empty at $t_0 = -\infty$, then the term $\tilde{\phi}(-\infty)\phi(-\infty) = 0$ and the lower integral boundary of the action starts at $t_0 = -\infty$.

Analogous results hold also for the non-Doi-shifted action, Eq. (1.82).

How can an observable, which requires a measurement of the system at several times, be included in the path integral? First, the observable is expressed in terms of operators a and $a^\dagger$. As it requires evaluations at several times, it is split up into observables for specific time points, each of which is expressed in terms of the operators a and $a^\dagger$, and a time evolution $e^{\tilde{\mathcal{H}}t}$ is included between each part. This operator expression is transformed in the field theory to path integrals with actions which are split up between the times of the pieces of the observable. Then each piece of the observable, say $a^{\dagger\alpha}a^\beta$, is bracketed in between $\langle\phi^\dagger(t)|$ and $|\phi(t)\rangle$, just as in the step from Eq. (1.96) to Eq. (1.97). Hence, a single-time piece of the observable $a^{\dagger\alpha}a^\beta$ is replaced by its corresponding fields $\phi^{\dagger\alpha}\phi^\beta$ and the exponential $e^{\phi^\dagger(t)\phi(t)}$. The exponential functions of previous and future time evolutions merge to form one unified action, just as for the initialization at several times.

As an explicit example, let's consider an initialization of k_0 particles at time t_0 and an additional k_1 particles at t_1 followed by an observable that has the operator $a^{\dagger\alpha_2}a^{\beta_2}$ at time t_2 and the operator $a^{\dagger\alpha_3}a^{\beta_3}$ at time t_3 . Then, the expectation of the observable given the chosen initialization equals

$$\begin{aligned} \left\langle \star \left| a^{\dagger k_3} a^{k_3} e^{\tilde{\mathcal{H}}(t_3-t_2)} a^{\dagger k_2} a^{k_2} e^{\tilde{\mathcal{H}}(t_2-t_1)} a^{\dagger k_1} e^{\tilde{\mathcal{H}}(t_1-t_0)} a^{\dagger k_0} \right| 0 \right\rangle &= \\ \int \mathcal{D}[\phi^\dagger, \phi] \phi^{\beta_3}(t_3) \phi^{\dagger\alpha_2}(t_2) \phi^{\beta_2}(t_2) \phi^{k_1}(t_1) \phi^{k_0}(t_0) e^{\mathcal{A}[\tilde{\phi}, \phi]} &= \\ =: \langle \phi^{\beta_3}(t_3) \phi^{\dagger\alpha_2}(t_2) \phi^{\beta_2}(t_2) \phi^{k_1}(t_1) \phi^{k_0}(t_0) \rangle, \end{aligned} \quad (1.98)$$

where the last equality uses a shorthand notation for the path integral, see in Eq. (1.89).

Further, the arguments about observables at several times is also valid if I include the observable $\mathcal{O}_{\text{final}} = 1$ at the final time $t = \infty$. Thus, the upper boundary of the action

functional $\mathcal{A}$ can be replaced by the time ∞ , and the action is

$$\mathcal{A}[\tilde{\phi}, \phi] = - \int_{-\infty}^{\infty} \left(\tilde{\phi}(t) \frac{d}{dt} \phi(t) - \tilde{\mathcal{H}}[\tilde{\phi}(t), \phi(t)] \right) dt \quad (1.99)$$

This extension to $+\infty$ assumes however, that the functions $\phi(t)$ and $\tilde{\phi}(t)$ can be integrated on this domain. If the number of particles is diverging, then, the extension of the upper boundary to $+\infty$ cannot work.

1.5.5 CONTINUUM LIMIT IN SPACE

In Eq. (1.31), a random walk was considered on a d -dimensional lattice $h\mathbb{Z}^d$. The Hamiltonian operator associated with the random walk on this lattice is

$$\tilde{\mathcal{H}} = \frac{D(h)}{2} \sum_{j \in \mathbb{Z}^d} \sum_{\substack{i \in \mathbb{Z}^d \\ \text{with} \\ |i-j|=1}} (\tilde{a}_i - \tilde{a}_j) (a_j - a_i). \quad (1.100)$$

As this Hamiltonian is already normal ordered, it can be transferred directly to its corresponding expression in terms of $\tilde{\phi}_j$ and ϕ_j , see Eq. (1.71):

$$\tilde{\mathcal{H}}[\tilde{\phi}_j, \phi_j] = \frac{D(h)}{2} \sum_{j \in \mathbb{Z}^d} \sum_{\substack{i \in \mathbb{Z}^d \\ \text{with} \\ |i-j|=1}} (\tilde{\phi}_i - \tilde{\phi}_j) (\phi_j - \phi_i). \quad (1.101)$$

The continuum limit can be achieved by taking $h \rightarrow 0$ and assuming that the hopping rate $D(h)$ depends on the lattice constant h as $D(h) = D/h^2$, where D is the diffusion constant. The h^2 in the denominator corresponds to a lattice Laplacian which converges to derivatives in $\tilde{\phi}_j$ and ϕ_j as $h \rightarrow 0$. However at the same time, ϕ_j is also turned into

a density which is denoted by ϕ :

$$\tilde{\mathcal{H}}[\tilde{\phi}_j, \phi_j] = \frac{D}{2} \sum_{\substack{j \in \mathbb{Z}^d \\ \rightarrow \int_{\mathbb{R}^d}}} \sum_{\substack{i \in \mathbb{Z}^d \\ \text{with } |i-j|=1}} \underbrace{\frac{(\tilde{\phi}_i - \tilde{\phi}_j)}{h}}_{\rightarrow -2\nabla \tilde{\phi}} \underbrace{\frac{(\frac{\phi_j}{h^d} - \frac{\phi_i}{h^d})}{h}}_{\rightarrow 2\nabla \phi} \underbrace{h^d}_{\rightarrow d^d x}. \quad (1.102)$$

The factors 2 in front of the gradient ∇ appear because in the sum over nearest neighbors (with index i) the difference ratio appears twice for every pair of nearest neighbors. Thus, the continuum limit is

$$\tilde{\mathcal{H}}[\tilde{\phi}, \phi] = - \int_{\mathbb{R}^d} D \left(\nabla \tilde{\phi} \right) (\nabla \phi) d^d x, \quad (1.103)$$

where $\tilde{\phi}$ and ϕ are functions of $x \in \mathbb{R}^d$, ϕ is a density. If the diffusion constant D is independent of x , then integration by parts results in

$$\tilde{\mathcal{H}}[\tilde{\phi}, \phi] = \int_{\mathbb{R}^d} D \tilde{\phi} \Delta \phi d^d x, \quad (1.104)$$

where Δ is the Laplace operator.

1.5.6 PROPAGATORS

The action $\mathcal{A}$ splits into two parts: a bilinear part $\mathcal{A}_{\text{lin}}$ (i.e. terms that are each both linear in ϕ and linear in $\tilde{\phi}$), and an interaction part, $\mathcal{A}_{\text{int}}$ which contains all other terms.

By construction, the bilinear part contains a time derivative. If there are n particle types, represented by several fields ϕ_j and $\tilde{\phi}_j$, all fields can be collectively written as vectors ϕ and $\tilde{\phi}$ of fields and the bilinear part is expressed using a matrix $A_{t'}$:

$$\mathcal{A}_{\text{lin}} = \int \left(\tilde{\phi}_1, \dots, \tilde{\phi}_n \right) A_{t'} \begin{pmatrix} \phi_1 \\ \vdots \\ \phi_n \end{pmatrix} dt' = \int \tilde{\phi}^T A_{t'} \phi dt', \quad (1.105)$$

where the index of A_t is a reminder that the matrix contains the time derivative. In

order to calculate expectations of observables, two auxiliary fields (or vectors of fields), J and $\tilde{J}$ are introduced in the path integral of the bilinear action:

$$\langle \bullet \rangle_0 = \int \mathcal{D}[\tilde{\phi}, \phi] \bullet e^{\mathcal{A}_{\text{lin}} + \int \tilde{J}\phi + \tilde{\phi}J dt} \Big|_{\substack{J=0 \\ \tilde{J}=0}} \quad (1.106)$$

Thus, any observable can be expressed using (functional) derivatives w.r.t. J and $\tilde{J}$. For example, the expectation of the observable $\phi(t)\tilde{\phi}(0)$ is written as

$$\langle \phi(t)\tilde{\phi}(t_0) \rangle_0 = \frac{\delta}{\delta \tilde{J}(t)} \frac{\delta}{\delta J(t_0)} \int \mathcal{D}[\tilde{\phi}, \phi] \phi(t)\tilde{\phi}(t_0) e^{\mathcal{A}_{\text{lin}} + \int \tilde{J}\phi + \tilde{\phi}J dt} \Big|_{\substack{J=0 \\ \tilde{J}=0}}, \quad (1.107)$$

where the functional derivative $\frac{\delta}{\delta J(t)}$ is the functional derivative w.r.t. J with the variation $\delta(t - t')$, and the functional derivative $\frac{\delta}{\delta \tilde{J}(t_0)}$ is the functional derivative w.r.t. $\tilde{J}$ with the variation $\delta(t_0 - t')$, see appendix, Sec. A.2.

The path integral (1.106) is a Gaussian integral. By discretizing the $\tilde{\phi}, \phi$ -space, this indeed becomes a Gaussian integral, which can be calculated. After the calculation, the continuum limit can be taken again and it turns out the result is equal to:

$$\int \mathcal{D}[\tilde{\phi}, \phi] e^{\mathcal{A}_{\text{lin}} + \int \tilde{J}\phi + \tilde{\phi}J dt'} = e^{\int \tilde{J} A_{t'}^{-1} J dt'} \quad (1.108)$$

As $A_{t'}$ contains a time derivative, it is not entirely clear what its inverse is. To avoid dealing with inverses of differential operators, it is useful to cast the problem in Fourier space. The observable $\phi(t)\tilde{\phi}(t_0)$ (or any other observable) can be Fourier transformed, see Sec. A.3 for the used conventions. For a single type of particle, $A_{t'}$ is a 1×1 matrix, which is typically of the form $-\frac{d}{dt'} - r$, and $\mathcal{A}_{\text{lin}}$ can be written as

$$\mathcal{A}_{\text{lin}} = \int \tilde{\phi}(t') \left(-\frac{d}{dt'} - r \right) \phi(t') dt' = - \int \tilde{\phi}(-\omega') \underbrace{(-i\omega' + r)}_{=: A_{\omega'}} \phi(\omega') d\omega'. \quad (1.109)$$

Thus, in Fourier space, $A_{t'}$ becomes $A_{\omega'}$, which can be inverted. However, we have to keep the changed sign of the argument of $\tilde{\phi}$ in mind. Hence, the path integral is

$$\int \mathcal{D}[\tilde{\phi}, \phi] \cdot e^{\mathcal{A}_{\text{lin}} + \int \tilde{J}(-\omega')\phi(\omega') + \tilde{\phi}(\omega')J(-\omega') d\omega'} = e^{\int \tilde{J}(-\omega') A_{\omega'}^{-1} J(-\omega') d\omega'} \quad (1.110)$$

Hence, in the example of the observable $\phi(t)\tilde{\phi}(t_0)$, Eq. (1.107), its expectation is expressed as

$$\langle \phi(t)\tilde{\phi}(0) \rangle_0 = \int \langle \phi(\omega)\tilde{\phi}(\omega_0) \rangle e^{-i\omega t - i\omega_0 t_0} d\omega d\omega_0 \quad (1.111)$$

and the Fourier transformed expectation of the observable, $\langle \phi(\omega)\tilde{\phi}(\omega') \rangle$ can be calculated using the Fourier transformed path integral, Eq. (1.110):

$$\langle \phi(\omega)\tilde{\phi}(\omega') \rangle_0 = \frac{\delta}{\delta \tilde{J}(-\omega)} \frac{\delta}{\delta J(-\omega_0)} e^{\int \tilde{J}(-\omega') A_{\omega'}^{-1} J(-\omega') d\omega'} \Big|_{\substack{J=0 \\ \tilde{J}=0}} \quad (1.112)$$

$$= \frac{\delta}{\delta \tilde{J}(-\omega)} \frac{\delta}{\delta J(-\omega_0)} \int \tilde{J}(-\omega') A_{\omega'}^{-1} J(-\omega') d\omega' \Big|_{\substack{J=0 \\ \tilde{J}=0}} \quad (1.113)$$

$$= \frac{\delta}{\delta \tilde{J}(-\omega)} \frac{\delta}{\delta J(-\omega_0)} \int \tilde{J}(-\omega'') A_{\omega'}^{-1} J(-\omega') \delta(\omega' + \omega'') d\omega' d\omega'' \Big|_{\substack{J=0 \\ \tilde{J}=0}} \quad (1.114)$$

$$= \int \delta(\omega + \omega'') A_{\omega'}^{-1} \delta(\omega_0 + \omega') \delta(\omega' + \omega'') d\omega' d\omega'' \quad (1.115)$$

$$= A_{-\omega_0}^{-1} \delta(\omega_0 + \omega). \quad (1.116)$$

Hence, for the example that $A_{t'} = -\frac{d}{dt'} - r$, the expectation of $\phi(t)\tilde{\phi}(t_0)$ is equal to

$$\langle \phi(t)\tilde{\phi}(t_0) \rangle = \int \frac{e^{-i\omega t - i\omega_0 t_0} \delta(\omega + \omega_0) d\omega d\omega_0}{i\omega_0 + r} = \Theta(r(t - t_0)) e^{-r(t - t_0)} \quad (1.117)$$

If the system includes diffusion, the bilinear part of the action, Eq. (1.109) will contain a $\tilde{\phi}(x, t) D \Delta \phi(x, t)$ term and the derivation of the propagator involves spatial Fourier transforms which lead to the following bare propagator in Fourier space:

$$\langle \phi(k, \omega) \tilde{\phi}(k_0, \omega_0) \rangle = \frac{\delta(k + k_0)}{-i\omega + Dk^2 + r}, \quad (1.118)$$

whose Fourier transform is a Gaussian in space with additional decay:

$$\langle \phi(x, t) \tilde{\phi}(x_0, t_0) \rangle = \frac{\Theta(r(t - t_0))}{\sqrt{4\pi D(t - t_0)}} e^{-\frac{(x - x_0)^2}{4D(t - t_0)} - r(t - t_0)}, \quad (1.119)$$

which is the Green's function of Brownian motion [44].

1.5.7 FEYNMAN DIAGRAMS

Usually, the action of a reaction-diffusion system will not only consist of a bilinear part but also contain an interaction part. In order to incorporate the interactions, a perturbative expansion around the bilinear part is performed. The expansion is ordered according to their power of each coefficients ('coupling') in the interaction. The non-zero terms appearing in the expansion are represented by Feynman diagrams.

Example 1: Let's assume the interaction part of the action, A_i , only consists of a term $q_2 \tilde{\phi}^2 \phi$ and the bilinear part of the action as the same as in Eq. (1.109) (as will be the case in Chapter 2). Then the entire path integral is

$$\langle \mathcal{O} \rangle = \int \mathcal{D}[\tilde{\phi}, \phi] \mathcal{O} e^{\mathcal{A}} = \sum_{k=0}^{\infty} \int \mathcal{D}[\tilde{\phi}, \phi] \mathcal{O} e^{\mathcal{A}_{\text{lin}}} \frac{\mathcal{A}_i^k}{k!}, \quad (1.120)$$

assuming that the order of the integral and the sum can be exchanged.

First, let's assume, that we want to calculate the (non-bare) propagator, i.e. $\mathcal{O} = \phi(\omega) \tilde{\phi}(\omega_0)$, then

$$\langle \phi(\omega) \tilde{\phi}(\omega_0) \rangle = \sum_{k=0}^{\infty} \int \mathcal{D}[\tilde{\phi}, \phi] \phi(\omega) \tilde{\phi}(\omega_0) e^{\mathcal{A}_{\text{lin}}} \frac{\left(q_2 \int \tilde{\phi}(\omega_a) \tilde{\phi}(\omega_b) \phi(\omega_c) \mathfrak{d}\omega_{a,b,c} \right)^k}{k!}, \quad (1.121)$$

where $\mathfrak{d}\omega_{a,b,c} = \delta(\omega_a + \omega_b + \omega_c) \mathfrak{d}\omega_a \mathfrak{d}\omega_b \mathfrak{d}\omega_c$. Since the integral contains a Gaussian part (i.e. $e^{\mathcal{A}_{\text{lin}}}$), all cases where an odd number of fields is multiplied with the Gaussian are zero.

For the case when an even number of fields is multiplied to the Gaussian, I follow the same strategy as in Eq. (1.107), i.e. I use functional derivatives of auxiliary fields J and $\tilde{J}$. In the end, however, I set the auxiliary fields to zero. A close inspection of the calculation in Sec. 1.5.6, reveals that all terms which are generated with an unequal number of J - and $\tilde{J}$ -derivatives will be zero once J and $\tilde{J}$ are set to zero. This happens because the auxiliary fields J and $\tilde{J}$ always appear in pairs, see Eq. (1.110). Since for any $k \geq 1$ in Eq. (1.121), the fields $\tilde{\phi}$ and ϕ will appear in equal numbers, the functional derivatives w.r.t. J and $\tilde{J}$ will appear in unequal numbers. Hence, all terms

in Eq. (1.121) are zero except the term for $k = 0$. From Eq. (1.116), I conclude

$$\langle \phi(\omega) \tilde{\phi}(\omega_0) \rangle = \langle \phi(\omega) \tilde{\phi}(\omega_0) \rangle_0 = \frac{\delta(\omega + \omega_0)}{-i\omega + r} \quad (1.122)$$

$$\hat{=} \text{---} \quad (1.123)$$

which is represented by one straight line because it contains only one bare propagator.

Example 2: Next, I calculate the expectation of the observable $\mathcal{O} = \phi(\omega_1)\phi(\omega_2)\tilde{\phi}(\omega_0)$ using the same action as in Example 1 above:

$$\langle \phi(\omega_1)\phi(\omega_2)\tilde{\phi}(\omega_0) \rangle = \sum_{k=0}^{\infty} \int \mathcal{D}[\tilde{\phi}, \phi] \phi(\omega_1)\phi(\omega_2)\tilde{\phi}(\omega_0) e^{\mathcal{A}_{\text{lin}}} \frac{\left(q_2 \int \tilde{\phi}(\omega_a) \tilde{\phi}(\omega_b) \phi(\omega_c) \mathfrak{d}\omega_{a,b,c} \right)^k}{k!}, \quad (1.124)$$

with $\mathfrak{d}\omega_{a,b,c} = \delta(\omega_a + \omega_b + \omega_c) \mathfrak{d}\omega_a \mathfrak{d}\omega_b \mathfrak{d}\omega_c$. I adjust the arguments in Example 1:

1. If the Gaussian $e^{\mathcal{A}_{\text{lin}}}$ is multiplied with an odd number of fields, the path integral is zero. However, in the present case, the conclusion is that k has to be odd.
2. Since I'm using auxiliary fields J and $\tilde{J}$ to calculate the path integral, I can conclude that only terms with equal numbers of ϕ and $\tilde{\phi}$ will be non-zero. Therefore $k = 1$ is the only contributing term.

$$\langle \phi(\omega_1)\phi(\omega_2)\tilde{\phi}(\omega_0) \rangle = \iint \mathcal{D}[\tilde{\phi}, \phi] \phi(\omega_1)\phi(\omega_2)\tilde{\phi}(\omega_0) e^{\mathcal{A}_{\text{lin}}} q_2 \tilde{\phi}(\omega_a) \tilde{\phi}(\omega_b) \phi(\omega_c) \mathfrak{d}\omega_{a,b,c}. \quad (1.125)$$

Next, I introduce the auxiliary fields J and $\tilde{J}$, see Eq. (1.106) and I use functional derivatives w.r.t. J and $\tilde{J}$ to calculate the path integral, see Eq. (1.116):

$$\begin{aligned} \langle \phi(\omega_1)\phi(\omega_2)\tilde{\phi}(\omega_0) \rangle &= \\ &= q_2 \int \frac{\delta}{\delta J_0} \frac{\delta}{\delta \tilde{J}_1} \frac{\delta}{\delta \tilde{J}_2} \frac{\delta}{\delta J_a} \frac{\delta}{\delta J_b} \frac{\delta}{\delta \tilde{J}_c} \int \mathcal{D}[\tilde{\phi}, \phi] e^{\mathcal{A}_{\text{lin}} + \int \tilde{J}\phi + \tilde{\phi}J} \mathfrak{d}\omega_{a,b,c} \Big|_{\substack{J=0 \\ \tilde{J}=0}} \end{aligned} \quad (1.126)$$

$$= 2q_2 \int \delta(\omega_a + \omega_b + \omega_c) \frac{\delta(\omega_1 + \omega_a)}{-i\omega_1 + r} \frac{\delta(\omega_2 + \omega_b)}{-i\omega_2 + r} \frac{\delta(\omega_0 + \omega_c)}{i\omega_0 + r} \mathfrak{d}\omega_a \mathfrak{d}\omega_b \mathfrak{d}\omega_c \quad (1.127)$$

where $J_0 = J(-\omega_0)$, $\dots$, and $\tilde{J}_c = J(-\omega_c)$. The factor 2 appears because, in the functional derivatives two such terms appear: one with the pairing of ω_1 to ω_a and ω_2 with ω_b as above and one term with ω_1 paired up with ω_b and ω_2 with ω_a . However, both terms are equal since ω_a and ω_b can be simply renamed. Eq. (1.127) can be evaluated for ω_a , ω_b and ω_c , and the inverse Fourier transform for ω_1 , ω_2 and ω_3 can be taken to find $\langle \phi(t_1)\phi(t_2)\tilde{\phi}(t_0) \rangle$, see Sec. 2.3.1 and Sec. 2.7.1 in Chapter 2, and in the appendix, Sec. B.1 for the explicit calculation.

Eq. (1.127) contains 3 bare propagators (which happen to be the full propagators in my example as well) which are ‘sown’ together. Eq. (1.127) is therefore represented as a Feynman diagram with 3 lines. These lines are connected because of the term $\tilde{\delta}(\omega_a + \omega_b + \omega_c)$:

$$\langle \phi(\omega_1)\phi(\omega_2)\tilde{\phi}(\omega_0) \rangle \hat{=} \begin{array}{c} \text{---} \text{---} \text{---} \\ \diagup \quad \diagdown \end{array} \quad (1.128)$$

By convention, the lines that are connected to the initialization fields (here $\tilde{\phi}(\omega_0)$) are placed on the right of the diagram, while the lines corresponding to the remaining fields of the observable (here $\phi(\omega_1)$ and $\phi(\omega_2)$) are placed on the left.

Example 3: While the previous examples only resulted in one Feynman diagram representing the observable, in the next example, I will show loop corrections. Let’s assume the same bilinear part as before and an interaction which contains two terms $\lambda\tilde{\phi}\phi^2$ and $\tau\tilde{\phi}^2\phi$, where λ and τ are parameters, called *couplings*, which in this example have the unit of rates. As in Example 1, I want to calculate the propagator:

$$\begin{aligned} \langle \phi(\omega)\tilde{\phi}(\omega_0) \rangle &= \sum_{k=0}^{\infty} \sum_{\ell=0}^{\infty} \int \mathcal{D}[\tilde{\phi}, \phi] \phi(\omega)\tilde{\phi}(\omega_0) e^{\mathcal{A}_{\text{lin}}} \\ &\quad \frac{\left(\tau \int \tilde{\phi}(\omega_a)\tilde{\phi}(\omega_b)\phi(\omega_c) d\omega_{a,b,c} \right)^k}{k!} \frac{\left(\lambda \int \tilde{\phi}(\omega_d)\phi(\omega_e)\phi(\omega_f) d\omega_{d,e,f} \right)^\ell}{\ell!}, \end{aligned} \quad (1.129)$$

The arguments used in Example 1 and 2 can be used again to conclude

1. Only terms with even numbers of fields contribute, hence $k + \ell$ must be even.
2. ϕ and $\tilde{\phi}$ must appear in equal numbers, hence, $k = \ell$.

This leaves still infinitely many terms and they all contribute to the full propagator. The case $k = \ell = 0$ was calculated in Example 1 and corresponds to the bare propagator. For $k = \ell = 1$, we find

$$\langle \phi(\omega) \tilde{\phi}(\omega_0) \rangle \Big|_{k=\ell=1} = \int \mathcal{D}[\tilde{\phi}, \phi] \phi(\omega) \tilde{\phi}(\omega_0) e^{\mathcal{A}_{\text{lin}}} \quad (1.130)$$

$$\begin{aligned} & \left(\tau \int \tilde{\phi}(\omega_a) \tilde{\phi}(\omega_b) \phi(\omega_c) \mathfrak{d}\omega_{a,b,c} \right) \left(\lambda \int \tilde{\phi}(\omega_d) \phi(\omega_e) \phi(\omega_f) \mathfrak{d}\omega_{d,e,f} \right) \\ &= 2\lambda\tau \int \frac{\delta(\omega + \omega_d)}{-i\omega + r} \frac{\delta(\omega_e + \omega_a)}{-i\omega_f + r} \frac{\delta(\omega_f + \omega_b)}{-i\omega_e + r} \frac{\delta(\omega_0 + \omega_c)}{i\omega_0 + r} \mathfrak{d}\omega_{a,b,c} \mathfrak{d}\omega_{d,e,f}, \end{aligned} \quad (1.131)$$

where $\mathfrak{d}\omega_{a,b,c} = \delta(\omega_a + \omega_b + \omega_c) \mathfrak{d}\omega_a \mathfrak{d}\omega_b \mathfrak{d}\omega_c$, $\mathfrak{d}\omega_{d,e,f}$ is defined analogously. The factor 2 appears because the term appears twice: once in the version above and once with ω_e paired to ω_b and ω_f paired to ω_a instead.

The expression contains 4 bare propagators and is therefore depicted by a Feynman diagram with 4 lines. The Dirac δ -functions $\delta(\omega_a + \omega_b + \omega_c)$ and $\delta(\omega_d + \omega_e + \omega_f)$ are represented as vertices:

$$\langle \phi(\omega) \tilde{\phi}(\omega_0) \rangle \Big|_{k=\ell=1} \hat{=} \text{---} \bigcirc \text{---} \quad (1.132)$$

For $k = \ell = 2$, there will be three topologically different diagrams:

$$\langle \phi(\omega) \tilde{\phi}(\omega_0) \rangle \Big|_{k=\ell=2} \hat{=} \text{---} \bigcirc \text{---} \bigcirc \text{---} \quad (1.133)$$

$$+ \text{---} \bigcirc \text{---} \quad (1.134)$$

$$+ \text{---} \bigcirc \text{---} \quad (1.135)$$

and for $k = \ell = 3, 4, 5$, there will be more and more topologically different Feynman diagrams.

In summary:

- Every term in a perturbative expansion of the path integral is depicted as a *Feynman* diagram,
- each bare propagator in each term is depicted as a line,

- each interaction term is depicted as a vertex,
- the power of the coupling associated with the interaction equals the number of how many times that vertex appears in the diagram,
- given a number of lines and vertices, each topologically possible diagram will appear.
- Advice: each type of field should be represented by a differently designed line. (see for example Chapter 3)

Not only can the Feynman diagrams be derived from the explicit perturbative expansion, but for a given Feynman diagram, the corresponding term of the expansion can be found by following the rules above vice versa. In particular, due to the functional derivatives w.r.t. J and $\tilde{J}$ in Fourier space, for each line in the Feynman diagram, a bare propagator is attached as a factor. Therefore, in real space, different parts of a Feynman diagram are connected via convolutions.

It is thus more convenient to draw an expansion in terms of its representation in Feynman diagrams, then write down its expression in Fourier space variables and then perform an inverse Fourier transformation.

1.6 CONCLUSION

Doi-Peliti field theory allows modelling complex stochastic reaction-diffusion systems. It solves the associated master equation, Eq. (1.12) indirectly by solving the implied linear partial differential equation for the probability generating function, Eq. (1.23). Although the machinery of the field theory appears to be complicated and difficult, once accustomed to it, many specific problems can be solved elegantly and systematically. This chapter laid the ground for the following two chapters, in which I showcase some of the field theory's benefits. Although it did not contain original work, there are some open questions to which I will turn in my future research. They are outlined in the following.

1.6.1 OUTLOOK: SUPERCRITICAL REGIME

A major problem of the Doi-Peliti field theories is its restriction to subcritical behaviour. This issue can be seen in the derivation of the bare propagator, Sec. 1.5.6, and in particular Eq. (1.117). Its solution in real space is an exponential decay which is forward in time if $r > 0$ and appears to be backward in time if $r < 0$. However, this apparent backward propagation in time is unphysical and a sign of the inability of Doi-Peliti field theory to deal with supercritical dynamics.

One step towards dealing with supercritical dynamics is the realization that the intuitively expected supercritical dynamics often contains exponential growth, which cannot be described by distributions (generalized functions), and in particular not tempered distributions which I assume in using Fourier transforms. The crucial step towards this catastrophe was Eq. (1.83) and the assumption that the integral boundaries can be taken to $\pm\infty$. Thus, the space of eligible functions ϕ over which the path integral sums cannot contain exponential growth.

Hence, the apparent impasse of supercritical behaviour can be overcome by stepping back to Eq. (1.83) and keeping the integral boundaries as they are. Then, in the derivation of the propagators, the Fourier series are used instead of Fourier transforms.

1.6.2 OUTLOOK: FINITE CARRYING CAPACITY

When reaction-diffusion systems are interpreted as models of other objects, like populations, mutations, or abstract stochastic processes, additional constraints of the system are often interesting. One important constraint is the carrying capacity. Can Doi-Peliti field theory describe a system where each site has a maximal number of particles it can hold? The carrying capacity can be interpreted as finite available space for a population, or – in the extreme case of a carrying capacity of one particle per site – as a fermionic constraint. Implementing such a constraint has been attempted in [167, 104]. Unfortunately, both approaches lead to complicated interactions and loop corrections. Therefore, alternative approaches are desirable.

One such approach is to change the identity operator in Eq. (1.63) to a projection

operator $\mathbb{P}_n$ onto a finite particle subspace, say carrying at most n particles:

$$\mathbb{P}_n = \sum_{k=0}^n |k\rangle\langle k|, \quad (1.136)$$

then, the identity operator which is expressed in terms of coherent states has to be adjusted as well, see appendix, Sec. A.1. Pseudo coherent states can be introduced as

$$|_n\phi\rangle := \sum_{k=0}^n \frac{\phi^k a^{\dagger k}}{k!} |0\rangle \quad \text{and} \quad \langle_n\phi| := \sum_{k=0}^n \langle 0| \frac{\phi^k a^k}{k!}, \quad (1.137)$$

which obey the following pseudo eigenstate property:

$$a|_n\phi\rangle = \phi|_{n-1}\phi\rangle \quad \text{and} \quad \langle_n\phi|a^\dagger = \langle_{n-1}\phi|\phi \quad (1.138)$$

Then, the projection in Eq. (1.136) can be expressed as

$$\mathbb{P}_n = \int \frac{d\text{Re}\phi d\text{Im}\phi}{\pi} e^{-\phi^\dagger\phi} |_n\phi\rangle\langle_n\phi|, \quad (1.139)$$

and used to calculate the exponential of the Hamiltonian operator, instead of the identity, see Eq. (1.65). The entire derivation of the path integral has to be repeated with the identities replaced by projections and coherent states replaced by pseudo coherent states. Hopefully, the resulting path integral has an action that can be worked with in practice, but this is for future research.

God did give the damn physicists all the solvable problems.

Philip E. Tetlock [157]

2

CONTINUOUS-TIME BRANCHING

ABSTRACT

In this chapter, continuous time branching processes are analyzed using a Doi-Peliti field theory. This research project was in collaboration with R. Garcia-Millan, B. Walter and G. Pruessner. Its results were published in *Phys. Rev. E* in 2018 [51]. My contribution to this project consisted of conducting and being involved with all analytics, in particular the moments, Sec. 2.3, the particle distribution, Sec. 2.4, the survival probability and time, Sec. 2.5, time covariances, Sec. 2.7 and total size characterization, Sec. 2.8. I was not involved with the simulations that were part of the publication [51]. However, for this thesis, I simulated the particle distribution, Fig. 2.4 and the expected time of death, Fig. 2.7, which were not part of the original project. In the conclusion, I present an outlook on my future research on coupled branching processes and noisy parameters, Sec. 2.9.

2.1 INTRODUCTION

Branching is the stochastic process of reproduction where an entity creates K copies of itself, which in turn repeat the same process independently. Different interpretations of this process have found numerous applications ranging from survival of family names [165] to nuclear chain reactions [119, 168]. One of the standard references for branching processes is the textbook by *T.E. Harris* (1964) [63]. An overview of its wide ranging applications can be found in [144].

There are many variations of branching processes. If offspring is created in discrete time steps, members of the population at a specific time are called a generation. In continuous time, two members of the population can produce offspring at different times, making the referral to generations more complicated. In particular, at a specific time, two members of the population might belong to different generations. Here, the focus lies on continuous time branching processes and the notion of generations is not used.

The number of created offspring K that a member produces is not fixed; it is a random variable and follows a discrete offspring distribution p_k . An example for such distributions is the binary offspring distribution

$$K = \begin{cases} 0 & \text{with probability } p_0 \in (0, 1), \\ 2 & \text{with probability } p_2 = 1 - p_0, \end{cases} \quad (2.1)$$

where p_0 is the probability that the population member has no offspring and dies, and p_2 is the probability that the member has 2 offspring and dies. An alternative interpretation for p_2 is that the member has one offspring and continuous to live. An example of a realization of a branching process with binary offspring distribution is shown in Fig. 2.1.

Another typical offspring distribution is the geometric distribution, where the possible number of offspring is unbound

$$K = \begin{cases} 0 & \text{with probability } p_0 \in (0, 1), \\ k & \text{with probability } p_k = p_0(1 - p_0)^k. \end{cases} \quad (2.2)$$

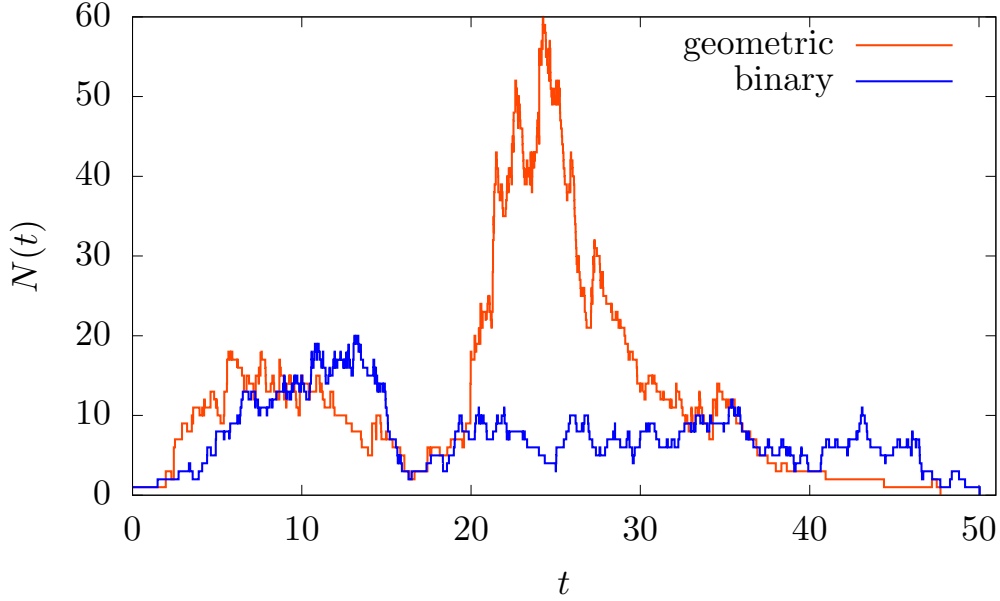


Figure 2.1: Simulated trajectories $N(t)$ of the branching process with binary offspring distribution (blue line) and geometric offspring distribution (red line) at criticality. Time t is in units of $1/s$, the average waiting time for a branching event of a single particle, see Eq. (2.3). Figure from [51].

While any non-negative integer can occur as the number of offspring, all the moments of the geometric distribution are well defined. An example of a realization of a branching process with geometric offspring distribution is shown in Fig. 2.1. In principle, offspring distributions with diverging higher moments could be considered for the branching process. However, the presented model below cannot treat such cases and they are therefore ignored. In the derivation below, the origin of this limitation will be pointed at, just after Eq. (2.21).

Some research on branching processes considers spatial aspects as well. This includes neuronal networks [7], earthquakes [99], and abstract networks [59]. In this chapter, the branching does not have a spatial component. It can be considered as a stochastic process in zero (space) dimensions. Only the time dimension plays a rôle.

In summary, this chapter describes a field theoretic formulation of continuous-time branching processes in zero spatial dimensions following offspring distributions with finite moments. The model and results have been published in *Phys. Rev. E*, **98**, 062107 (2018) together with co-authors Rosalba Garcia-Millan, Benjamin Walter and

Gunnar Pruessner [51]. Figures of this article are reproduced here; please note the copyright and licence statement on page 224.

2.2 DERIVATION OF THE MODEL

A continuous-time branching process can be regarded as a chemical reaction where reactants of type A spontaneously split into K copies of itself



with an average rate s . The number of offspring K is itself a random variable and can take any natural number value including zero. Hence, the number of particles $N(t)$ at time t is also a random variable with values $n \in \mathbb{N}_0$. Each reactant waits an exponentially distributed waiting time t , before it branches or dies. The parameter of that exponential distribution is s , i.e. $t \sim \text{Exp}(s)$.

Under assumptions of homogenous concentrations and vast available amounts, such chemical reactions are often modeled by rate equations. The rate equation corresponding to the chemical reaction (2.3) is

$$\frac{d}{dt}c_A(t) = \underbrace{s(\mathbb{E}[K] - 1)}_{=:-r} c_A(t), \quad (2.4)$$

where $c_A \geq 0$ is the expected concentration of reactant A , and a new rate $r = s(1 - \mathbb{E}[K])$ is introduced. The assumption of homogenous concentration eliminates considerations of spatial aspects. The rate r divides the phase space into a subcritical regime $r > 0$ and a supercritical regime $r < 0$. In the subcritical regime, the expected concentration converges to 0, i.e. $\lim_{t \rightarrow \infty} c_A(t) = 0$, while in the supercritical regime, the expected concentration diverges, i.e. $\lim_{t \rightarrow \infty} c_A(t) = \infty$.

The advantage of the rate-equation-approach (2.4) is that it can be solved analytically for $c_A(t)$ to be an exponential function:

$$c_A(t) = c_A(0)e^{-rt} \quad (2.5)$$

However, it entails several caveats: Inaccessible are higher moments of the number of

reactants, the probability distribution of the number of reactants, the distribution of the extinction time of the population of A , correlations between several time points, and many other observables. In fact, all aspect of the stochastic process are lost except the expected concentration c_A .

Realising these drawbacks, what are the alternative approaches? Under the assumption that all observables are smooth functions, casting the reaction (2.3) as a master equation safeguards all stochastic properties of the process.

In the following, a master equation, as in Eq. (1.12), is set up for the branching process. Let n denote the number of particles in the system and let $P(n, t) = P(N(t) = n)$ be the probability that the system contains n particles at time t . In the system, each particle independently undergoes the branching process. Because of this independence, the entire system waits an exponentially distributed waiting time until one of the n particles branches. The parameter of the exponential distribution of the entire system's waiting time is ns . Hence, if there are n particles in the system, the probabilistic transition rate to go in one branching event to a system with $n + k$ particles is nsp_k .

If a particle has no offspring, it is said to have died and it is lost from the system. This is equivalent to an extinction of particles and its rate is sp_0 for a single particle and nsp_0 for the extinction of one particle in a system containing n particles.

Thus, all the probabilistic transition rates are identified and the master equation can be set up as follows:

$$\begin{aligned} \frac{d}{dt}P(n, t|n_0, t_0) = \\ =s \sum_{k=0}^{\infty} \left((n - k + 1)p_k P(n - k + 1, t|n_0, t_0) - np_k P(n, t|n_0, t_0) \right). \end{aligned} \quad (2.6)$$

The system is initialised with one particle $n_0 = 1$ at time $t_0 = 0$, i.e. $P(1, 0) = 1$ and $P(n \neq 1, 0) = 0$. The probabilities for negative particle numbers are set to zero, i.e. $P(n < 0, t) = 0$. From Eq. (2.6), the unique absorbing state $n = 0$ can be identified.

The derivation of the corresponding Doi-Peliti field theory, as outlined for a general master equation from Sec. 1.3 to Sec. 1.5 is briefly repeated for the branching process in the following.

The time-dependent probability generating function of the branching process is

$$\mathcal{M}(z, t) = \sum_{n=0}^{\infty} P(n, t | n_0, t_0) z^n. \quad (2.7)$$

Using the master equation, the time evolution of the probability generating function can be expressed using the differentiation operator d/dz and multiplication operator $z \cdot$ as follows

$$\frac{d}{dt} \mathcal{M}(z, t) = s \sum_{k=0}^{\infty} \left(p_k z^k \frac{d}{dz} - p_k z \frac{d}{dz} \right) \mathcal{M}(z, t), \quad (2.8)$$

which is a first-order, linear partial differential equation.

In order to get insight into the solution of the master equation (2.6) and the corresponding PDE of the probability generating function (2.8), it is transferred into a Doi-Peliti field theory. As a first step, an equivalent, second-quantized form is derived by introducing the ladder operators a and $a^\dagger$, which follow the commutation rule $[a, a^\dagger] = aa^\dagger - a^\dagger a = 1$, and which act on *bra*- and *ket*-vectors $\langle n|$ and $|n\rangle$ as follows

$$a|n\rangle = n|n-1\rangle \quad (2.9)$$

$$a^\dagger|n\rangle = |n+1\rangle \quad (2.10)$$

$$\langle n|a = \langle n+1|(n+1) \quad (2.11)$$

$$\langle n|a^\dagger = \langle n-1|. \quad (2.12)$$

The *bra-ket*-vectors form an orthonormal basis:

$$\langle m|n\rangle = \delta_{m,n}, \quad (2.13)$$

where $\delta_{m,n}$ is the Kronecker- δ . The *bra-ket*-vectors represent how many particles the system contains. The vector $|n\rangle$ indicates that there are n particles present.

The probability generating function (2.7) can be written with *bra-ket*-vectors, as

$$|\mathcal{M}(t)\rangle = \sum_{n=0}^{\infty} P(n, t | n_0, t_0) |n\rangle. \quad (2.14)$$

The PDE (2.8) is expressed using the ladder operators as follows

$$\frac{d}{dt}|\mathcal{M}(t)\rangle = s \sum_{k=0}^{\infty} (p_k a^{\dagger k} a - p_k a^{\dagger} a) |\mathcal{M}(t)\rangle. \quad (2.15)$$

Eq. (2.15) is called the second quantized form of the master equation (2.6). The term on the right hand side, in front of $|\mathcal{M}(t)\rangle$ is the associated Hamiltonian $\mathcal{H}$.

From Sec. 1.4 to Sec. 1.5.6, it is shown how to derive a path integral formulation of a second quantized form of a master equation. The result is that the action of the field theory can be derived by replacing the operators a and $a^{\dagger}$ in the Hamiltonian by time dependent fields $\phi(t)$ and $\phi^{\dagger}(t)$ in the integral of the action. For the branching process and the Hamiltonian in Eq. (2.15), this is

$$\mathcal{A}[\phi, \phi^{\dagger}] = s \int_{\mathbb{R}} \sum_{k=0}^{\infty} (p_k \phi^{\dagger k}(t) \phi(t) - p_k \phi^{\dagger}(t) \phi(t)) + \tilde{\phi}(t) \frac{\partial}{\partial t} \phi(t) dt. \quad (2.16)$$

As the Hamiltonian is dimensionless and s has the unit of a rate, it can be derived that ϕ and $\phi^{\dagger}$ are both dimensionless too.

In Doi-Peliti field theory, the creation field has to be shifted $\tilde{\phi}(t) = \phi^{\dagger}(t) - 1$, which results in the shifted action

$$\mathcal{A}[\phi, \tilde{\phi}] = s \int_{\mathbb{R}} \sum_{k=0}^{\infty} \left(p_k \sum_{j=1}^k \binom{k}{j} \tilde{\phi}^j(t) \phi(t) - p_k (1 + \tilde{\phi}(t)) \phi(t) \right) + \tilde{\phi}(t) \frac{\partial}{\partial t} \phi(t) dt. \quad (2.17)$$

By introducing rates q_j and r with

$$q_j = s \sum_{k=0}^{\infty} p_k \binom{k}{j} \quad \text{and} \quad r = (s - q_1), \quad (2.18)$$

the action can be written compactly as

$$\mathcal{A}[\phi, \tilde{\phi}] = \int_{\mathbb{R}} \sum_{j=2}^{\infty} q_j \tilde{\phi}^j(t) \phi(t) - \tilde{\phi}(t) \left(\frac{\partial}{\partial t} + r \right) \phi(t) dt. \quad (2.19)$$

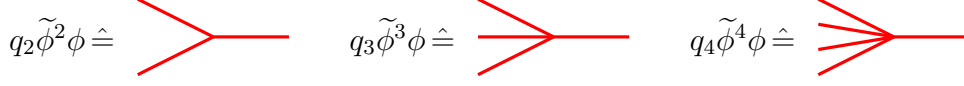


Figure 2.2: Representation of the first three interaction terms as vertices in Feynman diagrams. In general, the interaction term $q_k \tilde{\phi}^k \phi$ is represented by a vertex with one incoming leg and k outgoing legs.

For the binary and geometric offspring distributions, Eqs. (2.1) and (2.2), I find the following explicit expressions for q_j and r

$$\text{binary} \quad q_2 = sp_2, \quad \forall j \geq 3 : q_j = 0 \quad r = s(p_0 - p_2) \quad (2.20)$$

$$\text{geometric} \quad q_j = s \frac{(1 - p_0)^j}{p_0^j} \quad r = s \left(2 - \frac{1}{p_0} \right) \quad (2.21)$$

However, if the offspring distribution did not have finite moments, the q_j s would not be well-defined and therefore the action (2.19) wouldn't be well-defined either.

The bilinear part $\mathcal{A}_{\text{lin}}$ of the action $\mathcal{A}$ is identified as

$$\mathcal{A}_{\text{lin}}[\phi, \tilde{\phi}] = - \int_{\mathbb{R}} \tilde{\phi}(t) \left(\frac{\partial}{\partial t} + r \right) \phi(t) dt \quad (2.22)$$

Using Fourier transforms (A.11), the bilinear part of the action (2.22) is rewritten in frequency space as

$$\mathcal{A}_{\text{lin}}[\phi, \phi^\dagger] = - \int_{\mathbb{R}^2} \tilde{\phi}(\omega') (-i\omega + r) \phi(\omega) \delta(\omega + \omega') d\omega d\omega', \quad (2.23)$$

where $d\omega = 2\pi d\omega$ and $\delta(\omega + \omega') = 2\pi \delta(\omega + \omega')$.

The interaction part of the action $\mathcal{A}$ includes all the terms that are not bilinear. They are represented by vertices in Feynman diagrams, shown in Fig. 2.2. In particular, the action does not allow for loops in the Feynman diagrams and therefore, all connected Feynman diagrams are tree-like.

Furthermore, the system will always be initialized with a single particle $n_0 = 1$ at time t_0 in the following Secs. 2.3 to 2.8. Therefore, all Feynman diagrams will have a single incoming leg.

The expectation of an observable $\mathcal{O}$ can be calculated using the path integral

$$\mathbb{E}[\mathcal{O}] = \int \mathcal{O} e^{\mathcal{A}[\phi, \tilde{\phi}]} \mathcal{D}[\phi, \tilde{\phi}], \quad (2.24)$$

where the measure $\mathcal{D}$ is defined such that the expectation of observable 1 is 1:

$$\mathbb{E}[1] = \int e^{\mathcal{A}[\phi, \tilde{\phi}]} \mathcal{D}[\phi, \tilde{\phi}] = 1. \quad (2.25)$$

With this choice of the measure $\mathcal{D}$, the path integral describes a probability distribution of all paths that the stochastic process $N(t)$ might take.

2.3 MOMENTS

After initialising the system with a single particle at time $t_0 = 0$, particles branch and go extinct randomly. The time dependent moments of the particle distribution are observables and can be calculated directly using the path integral (2.24). The n th moment of the number of particles in the system, after an initialization with one particle at time $t_0 = 0$ is

$$\mathbb{E}[N^n(t) | N(0) = 1] = \left\langle \star \left| (a^\dagger a)^n e^{\mathcal{H}[a^\dagger, a]t} a^\dagger \right| 0 \right\rangle \quad (2.26)$$

$$= \sum_{k=1}^n \left\{ \begin{matrix} n \\ k \end{matrix} \right\} \left\langle \star \left| a^{\dagger k} a^k e^{\mathcal{H}[a^\dagger, a]t} a^\dagger \right| 0 \right\rangle \quad (2.27)$$

$$= \sum_{k=1}^n \left\{ \begin{matrix} n \\ k \end{matrix} \right\} \left\langle \star \left| a^k e^{\mathcal{H}[a^\dagger, a]t} a^\dagger \right| 0 \right\rangle \quad (2.28)$$

$$= \sum_{k=1}^n \left\{ \begin{matrix} n \\ k \end{matrix} \right\} \left\langle \phi^k(t) \tilde{\phi}(0) \right\rangle \quad (2.29)$$

where I used the Stirling number of the second kind to obtain normal ordering [102]

$$\left\{ \begin{matrix} n \\ k \end{matrix} \right\} = \frac{1}{k!} \sum_{i=0}^k (-1)^i \binom{k}{i} (k-i)^n \quad (1.38)$$

The term $\langle \phi^k(t) \tilde{\phi}(0) \rangle$ is represented by all Feynman diagrams which consist of one ingoing leg and k outgoing legs.

In particular, Eq. (2.29) is exactly the relation between moments and factorial moments (see also Eqs. (1.36) and (1.37)), which is the (bijective) Stirling transform. Therefore, I can identify the factorial moments as

$$\mathbb{E}[(N(t))_k | N(0) = 1] = \langle \phi^k(t) \tilde{\phi}(0) \rangle. \quad (2.30)$$

2.3.1 FIRST AND SECOND MOMENT

The first and second moment can be calculated directly, they only involve r and q_2 . If $r > 0$, the first moment is:

$$\mathbb{E}[N(t) | N(0) = 1] = \langle \phi(t) \tilde{\phi}(0) \rangle = \int_{\mathbb{R}} \frac{e^{-i\omega t}}{-i\omega + r} d\omega = \Theta(t) e^{-rt}, \quad (2.31)$$

where $\Theta(t)$ is the Heaviside function and $d\omega = d\omega/(2\pi)$. Its corresponding Feynman diagram is a straight line

$$\langle \phi(t) \tilde{\phi}(0) \rangle \hat{=} \text{---}. \quad (2.32)$$

There are no corrections to this result because there is no perturbative expansion for the expected particle number.

The expected number of particles is zero before $t_0 = 0$, then discontinuously jumps to $N(0) = 1$, followed by an exponential decay back to zero. The viewpoint of the response field formalism is that at $t_0 = 0$, the system was perturbed and then relaxes back to its ground state $N = 0$.

Ignoring the derivation in Sec. 1.5, blindly replacing the positive r in Eq. (2.31) by a negative r leads to the following result

$$\text{if } r < 0 : \quad \int_{\mathbb{R}} \frac{e^{-i\omega t}}{-i\omega + r} d\omega = \Theta(-t) e^{-rt}, \quad (2.33)$$

which is an exponential increase prior to the initialization where it is abruptly stopped.

This result contradicts the common understanding of how cause and effect are placed in time. It is hinting at a significant drawback of the field theory. The intuitively expected result for any r is

$$\mathbb{E}[N(t)|N(0) = 1] = \Theta(t)e^{-rt} \quad (2.34)$$

I will address this issue in Sec. 2.4 and restrict myself to the case $r > 0$ for the time being, occasionally taking the limit $r \rightarrow 0^+$. Unless stated otherwise, all expressions are assumed to be for the subcritical case $r > 0$.

Following Eq. (2.29), the second moment is $\mathbb{E}[N^2(t)|N(0) = 1] = \langle \phi(t)\tilde{\phi}(0) \rangle + \langle \phi^2(t)\tilde{\phi}(0) \rangle$. It is represented by two Feynman diagrams

$$\mathbb{E}[N^2(t)|N(0) = 1] \hat{=} \text{---} + \text{---} \text{---} \text{---}, \quad (2.35)$$

of which the first part was calculated in Eq. (2.31) and the second part is the 2nd factorial moment $\langle \phi^2(t)\tilde{\phi}(0) \rangle$, which is equal to

$$\langle \phi^2(t)\tilde{\phi}(0) \rangle = \Theta(t)2\frac{q_2}{r}e^{-rt}(1 - e^{-rt}) \quad (2.36)$$

whose detailed calculation is presented in the appendix, Sec. B.1.

Hence, for $r > 0$, the second moment equals

$$\mathbb{E}[N^2(t)|N(0) = 1] = \Theta(t)e^{-rt} \left(1 + 2\frac{q_2}{r}(1 - e^{-rt}) \right). \quad (2.37)$$

Therefore, the variance of the particle number is equal to

$$\text{Var}(N(t)|N(0) = 1) = \Theta(t)e^{-rt} \left(1 + 2\frac{q_2}{r} \right) (1 - e^{-rt}). \quad (2.38)$$

As expected, the variance is zero at the initialization $t_0 = 0$ because the particle number is fixed. The variance reaches its maximum at $t_{\max} = (\ln 2)/r$, i.e. the time point of the maximum variance does not depend on higher moments of the offspring distribution and it diverges to $+\infty$ as $r \rightarrow 0$. The variance is shown in Fig. 2.10 for the binary offspring distribution.

When the critical point is approached, $r \rightarrow 0^+$, the expected particle number becomes

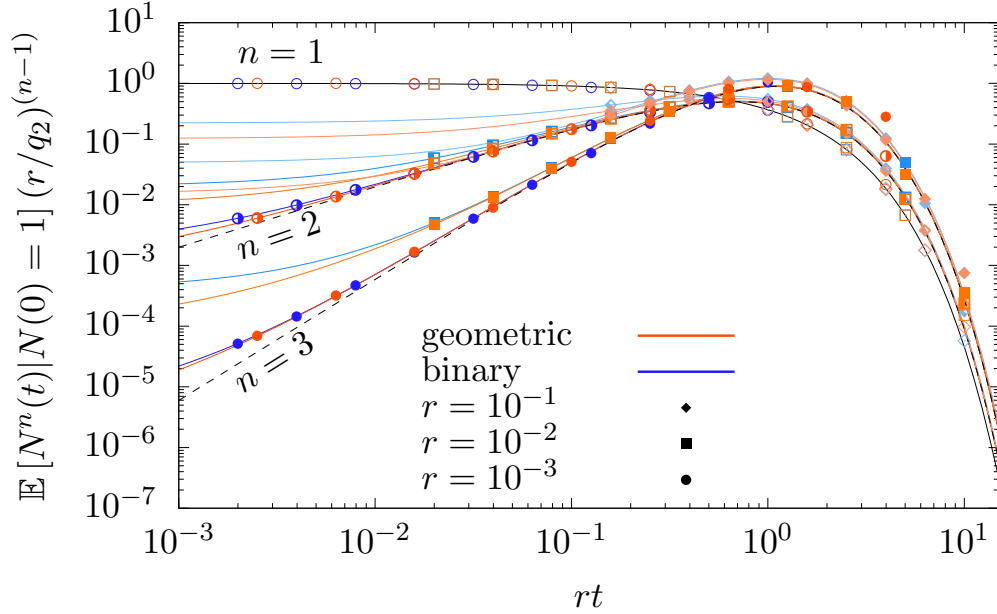


Figure 2.3: First, second and third moments ($n = 1, 2, 3$) of branching processes with binary and geometric offspring distributions. As both offspring distributions are one-parameter distributions, the coefficient r is sufficient to completely define the distribution. Markers: simulation results. Straight lines: Exact analytical solutions, Eqs. (2.31), (2.37), and (2.42). Dashed line: analytical approximations for $n = 2, 3$, Eq. (2.44). The scaling of the y -axis by $(r/q_2)^{n-1}$ and of the x -axis by r results in a collapse of $g_n(t)$ for all values of r . Figure adapted from [51].

a constant $\lim_{r \rightarrow 0^+} \mathbb{E}[N(t), N(0) = 1] = 1$, while the variance diverges as $\propto r^{-1}$.

Simulations of the first and second moment of branching processes with binary and geometric offspring distributions are shown alongside these analytical results in Fig. 2.3. It shows that the analytical results agree very well with the simulations.

2.3.2 THIRD MOMENT

When considering the third moment, an additional complication arises: higher order interactions. As shown in Eqs. (2.26)–(2.29), the third moment contains the terms $\langle \phi(t) \tilde{\phi}(0) \rangle$, $\langle \phi^2(t) \tilde{\phi}(0) \rangle$, and $\langle \phi^3(t) \tilde{\phi}(0) \rangle$. While I studied the first two terms in the previous subsection 2.3.1, the last term has not been considered so far. It is represented

by the following Feynman diagrams

$$\left\langle \phi^3(t) \tilde{\phi}(0) \right\rangle \doteq \underbrace{\text{diagram}}_{\doteq I_3} + \underbrace{\text{diagram}}_{\doteq g_3} \quad (2.39)$$

I split the terms of $\langle \phi^3(t) \tilde{\phi}(0) \rangle$ into two parts: I_3 is the term which has a factor q_3 and g_3 is the term that has the factor q_2^2 . If the offspring distribution is binary, $q_3 = 0$ and therefore $I_3 = 0$.

For I_3 and g_3 , I find

$$I_3(t) = \Theta(t) \frac{3q_3}{r} e^{-rt} (1 - e^{-2rt}) \quad (2.40)$$

$$g_3(t) = \Theta(t) 3! e^{-rt} \left(\frac{q_2}{r} (1 - e^{-rt}) \right)^2 \quad (2.41)$$

whose detailed calculation can be found in the appendix, Sec. B.2.

All the terms for calculating the third moment are known now and I combine Eq. (2.40), (2.41), (2.36), and (2.31) in Eq. (2.29), to find

$$\begin{aligned} \mathbb{E}[N^3(t) | N(0) = 1] = & \Theta(t) 6e^{-rt} \left(1 + \frac{q_2}{r} (1 - e^{-rt}) + \frac{q_3}{2r} (1 - e^{-2rt}) \right. \\ & \left. + \left(\frac{q_2}{r} (1 - e^{-rt}) \right)^2 \right) \end{aligned} \quad (2.42)$$

The third moment, Eq. (2.42), is shown in Fig. 2.3 for binary and geometric offspring distributions alongside simulation results, which agree very well.

Since the third moment is already quite complicated and since it is expected that higher moments will be even more complicated, it is reasonable to ask if there are useful approximations for the moments. Characterising the moments close to the critical point $r = 0$ is useful if I want to describe the universal properties of branching processes. This approach is explored in the following subsection.

2.3.3 n TH MOMENT APPROXIMATION

Close to the critical point $r = 0$, the terms of the n th moment which have the highest power of r^{-1} will dominate the expression of the moment, i.e. its amplitude will be largest. These terms can be identified by employing a dimensional argument when considering all involved Feynman diagrams. It goes as follows:

- Following the normal ordering in Eq. (2.29), the n th moment is described by Feynman diagrams with 1 incoming leg and $m \leq n$ outgoing legs.
- For fixed m , with $3 \leq m \leq n$, diagrams contain ℓ internal legs (i.e. legs connecting two vertices). There can be between 0 and $m - 1$ internal legs. There are zero internal legs if the ingoing leg splits into m , using coupling q_m . There are $m - 1$ internal legs if all vertices are dyadic, i.e. only the coupling q_2 is used and it appears as a factor q_2^{m-1} .
- As the terms represented by the Feynman diagrams are parts of moments, they are dimensionless. The q_j s have the unit of inverse time and only appear as factors with powers equal to the number of vertices with j outgoing legs. The only parameter in the system which can dimensionally compensate is r which also has the unit of inverse time.
- Therefore, a diagram with
 - ▷ k_2 vertices with 2 outgoing legs each,
 - ▷ k_3 vertices with 3 outgoing legs each,
 - ▷ $\dots$,
 - ▷ k_j vertices with j outgoing legs each

has a prefactor $q_2^{k_2} \dots q_j^{k_j}$ and a prefactor $r^{-k_2} \dots r^{-k_j}$, which makes the result dimensionless.

- Hence, the term represented by the Feynman diagrams with the highest number of vertices has the greatest power of r^{-1} .

- The n th moment is dominated by the Feynman diagrams with n outgoing legs and with the highest number of vertices, i.e. those which only contain dyadic vertices using coupling q_2 only.

Hence, I use the following approximation for $r \rightarrow 0^+$

$$\mathbb{E}[N^n(t)|N(0) = 1] = \underbrace{\left\langle \phi^n(t) \tilde{\phi}(0) \right\rangle \Big|_{\substack{j \geq 3 \\ q_j = 0}}}_{=: g_n(t)} + \mathcal{O}(r^{-(n-2)}), \quad (2.43)$$

where $\mathcal{O}(\cdot)$ refers to the (Bachmann-Landau) Big O notation [3, 88]. In this expression, all diagrams that contain vertices with more than 2 outgoing legs are zero by setting $q_j = 0$ for $j \geq 3$.

Furthermore, the same dimensional argument also holds when applied to factorial moments with the difference that g_n is an even better approximation for the n th factorial moments than for the n th moment, because less terms are disregarded: While the n th moment contains terms which are represented by Feynman diagrams with less than n outgoing legs and which are disregarded in our approximation, the n th factorial moment only contains terms which are represented by Feynman diagrams with exactly n outgoing legs.

In the case of the binary branching distribution, g_n is not an approximation for the n th factorial moment, but it is exact, i.e. $g_n(t) = \mathbb{E}[(N(t))_n | N(0) = 1]$, because for $j \geq 3$, $q_j = 0$ and only dyadic vertices occur, see Eq. (2.20).

In the previous subsections 2.3.1 and 2.3.2, I identified

$$\begin{aligned} g_1(t) &= \Theta(t)e^{-rt}, & g_2(t) &= \Theta(t)2e^{-rt}\frac{q_2}{r}(1 - e^{-rt}), \\ g_3(t) &= \Theta(t)3!e^{-rt}\left(\frac{q_2}{r}(1 - e^{-rt})\right)^2, \end{aligned} \quad (2.44)$$

from which it can be deduced that

$$g_n(t) = \Theta(t)n!e^{-rt}\left(\frac{q_2}{r}(1 - e^{-rt})\right)^{n-1} \quad (2.45)$$

Its correctness can be proved by induction: I consider the following dyadic Feynman

diagrams

$$g_n(t) \hat{=} \sum_{k=1}^{n-1} \binom{n}{k} \left(\begin{array}{c} k \vdots \\ \text{---} \bullet \\ \text{---} \bullet \\ n-k \vdots \end{array} \right) \quad (2.46)$$

This Feynman diagram consists of two branches: the upper one has k outgoing legs and represents g_k and the lower has $n - k$ outgoing legs and represents g_{n-k} . Both branches are connected by stem, representing their convolution with g_1 . This is indeed a convolution because in Fourier space it is a product of the corresponding Fourier-transforms of g_k , g_{n-k} and g_1 , as was derived in Chapter 1, Sec. 1.5.7.

Therefore $g_n(t)$ can be expressed as a convolution

$$\begin{aligned} g_n(t) &= \sum_{k=1}^{n-1} \binom{n}{k} q_2 \int_0^t e^{-r(t-t')} g_k(t') g_{n-k}(t') dt' \\ &= n! e^{-rt} \left(\frac{q_2}{r} (1 - e^{-rt}) \right)^{n-1}, \end{aligned} \quad (2.47)$$

which completes the induction. The details of the second equality are shown in the appendix in Sec. B.3.

Therefore, the n th moment, and the n th factorial moment are equal to

$$\mathbb{E}[N^n(t) | N(0) = 1] = \Theta(t) n! e^{-rt} \left(\frac{q_2}{r} (1 - e^{-rt}) \right)^{n-1} + \mathcal{O}(r^{-(n-2)}), \quad (2.48)$$

whose maximum occurs as $t_{\max} = (\ln n)/r$. As criticality is approached $r \rightarrow 0$, $t_{\max} \rightarrow \infty$ and the n th moment and n th factorial moment diverge as $\propto r^{-n+1}$.

Close to the critical point $r = 0$, branching processes with different offspring distributions will have different moments because q_2 differ between different offspring distributions. However, the ratios of moments are equal to

$$\frac{\mathbb{E}[N^k(t)] \mathbb{E}[N^\ell(t)]}{\mathbb{E}[N^{k+m}(t)] \mathbb{E}[N^{\ell-m}(t)]} \approx \frac{g_k(t) g_\ell(t)}{g_{k+m}(t) g_{\ell-m}(t)} = \frac{k! \ell!}{(k+m)! (\ell-m)!}, \quad (2.49)$$

for $k, \ell, m \in \mathbb{N}$ and $\ell > m$, where all expectations are conditioned on $N(0) = 1$. There

are independent of q_2 and of time, and therefore reflect the universal behaviour of branching processes near the critical point.

2.4 PROBABILITY DISTRIBUTION $P(N(t)|N(0) = 1)$

Factorial moments and probabilities are connected through the probability generating function, Eqs. (1.32), (1.33) and (1.34): if the range of the random variable is non-negative integers, then the probability generating function and the factorial moment generating function are equal: the probabilities feature as coefficients of the Taylor expansion at $z = 0$, while the factorial moments feature as coefficients of the Taylor expansion at $z = 1$.

If all factorial moments are known, the probability distribution $P(N(t)|N(0) = 1)$ itself can be recovered by writing the probability generating function in terms of the factorial moments and then take derivatives and evaluations at $z = 0$ to obtain the probabilities:

$$P(N(t) = m|N(0) = 1) = \frac{1}{m!} \frac{d^m}{dz^m} \mathcal{M}(z, t) \Big|_{z=0} \quad (2.50)$$

$$= \frac{1}{m!} \frac{d^m}{dz^m} \sum_{k=0}^{\infty} \frac{(z-1)^k \mathbb{E}[(N(t))_k|N(0) = 1]}{k!} \Big|_{z=0} \quad (2.51)$$

$$= \begin{cases} 1 - \frac{e^{-rt}}{1 + \frac{q_2}{r}(1-e^{-rt})} + \mathcal{O}(r^{-1}) & \text{if } m = 0 \\ \frac{e^{-rt} \left(\frac{q_2}{r}(1-e^{-rt}) \right)^{m-1}}{\left(1 + \frac{q_2}{r}(1-e^{-rt}) \right)^{m+1}} + \mathcal{O}(r^{-1}) & \text{for } m > 0, \end{cases} \quad (2.52)$$

where the step from Eq. (2.51) to Eq. (2.52) is presented in the appendix, Sec. B.4. Eq. (2.52) is exact for binary offspring distributions. This result was also found in [119] using other means. The probabilities for $m \in \{0, \dots, 6\}$ are shown for a binary and geometric offspring distributions in Fig. 2.4 alongside simulation results. While the analytical results and simulation results agree well for binary offspring distributions, there is a significant disagreement for the geometric offspring distribution for small times $st < 1$. This disagreement for small times is due to the approximation of the n th factorial moment, Eq. (2.48), where terms involving $q_3, q_4, \dots$ were ignored. These parameters characterize how often a particle splits into 3, 4, $\dots$ particles in one event. Between initialization and time $st = 1$, typically one offspring event occurs. Therefore, the approximation overestimates the probability to split into 2 particles and

underestimates the probability to split into 3, 4, ... particles before time $st = 1$.

By considering the probability distribution (2.52), it is found that the probabilities obey universal ratios:

$$\text{for all } k + \ell = m + n : \quad \frac{P(N(t) = k)P(N(t) = \ell)}{P(N(t) = m)P(N(t) = n)} = 1, \quad (2.53)$$

$$\text{for all } k + \ell = m + n : \quad \frac{P(N(t) > k)P(N(t) > \ell)}{P(N(t) > m)P(N(t) > n)} = 1, \quad (2.54)$$

with $k, \ell, m, n > 0$ for the first ratio, and where all probabilities are actually conditioned on $N(0) = 1$. At criticality, these ratios are exact for all offspring distributions.

Now that we have the distribution $P(N(t) = m | N(0) = 1)$, we can check that it is a solution of the original master equation, Eq. (2.6), with binary offspring distribution. It turns out, that Eq. (2.52) also solves the master equation for $r < 0$. Since the master equation is a linear ordinary differential equation, it has a unique solution – even in the supercritical regime $r < 0$. Therefore, Eq. (2.52) is the solution of the branching process with binary offspring distributions in the sub- and supercritical regime.

2.5 SURVIVAL PROBABILITY AND TIME OF DEATH

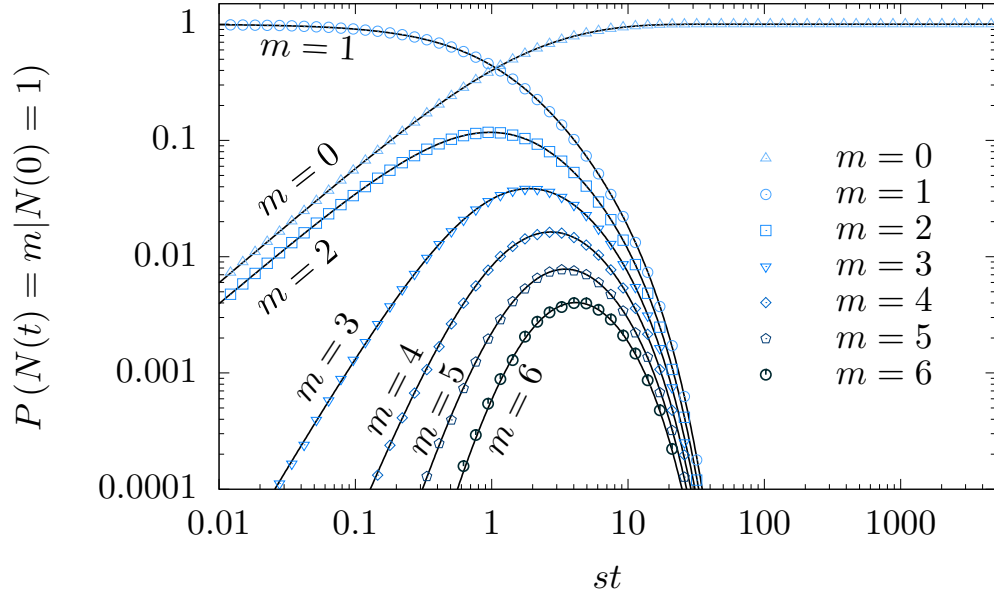
When considering branching processes, the fact that there exists the unique absorbing state $N = 0$ naturally leads to the questions

- How likely is it that a branching process has not been absorbed / died yet at time t , if it was initiated with a single particle at time $t_0 = 0$?
- What is the distribution of times of deaths and what is the expected time of death?

The answer to the first question can be found by realising that the probability of extinction before time t , $P(N(t) = 0 | N(0) = 1)$, is 1 minus the probability of survival until time t , $P_s(t) := P(N(t) > 0 | N(0) = 1)$:

$$P_s(t) = \frac{e^{-rt}}{1 + \frac{q_2}{r}(1 - e^{-rt})}. \quad (2.55)$$

(a)



(b)

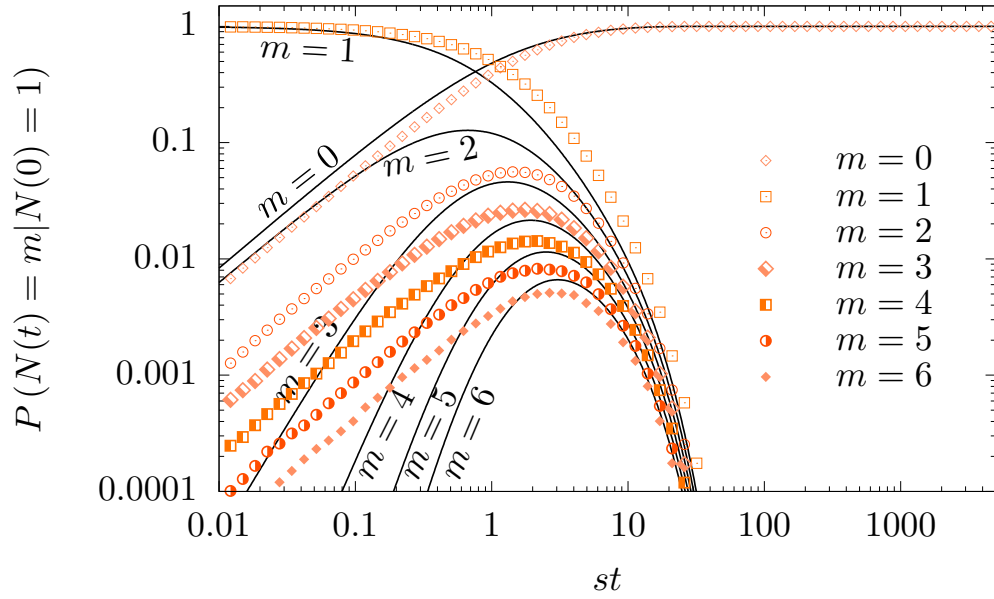


Figure 2.4: Probabilities $P(N(t) = m | N(0) = 1)$ for $m \in \{0, 1, 2, 3, 4, 5, 6\}$ for a binary offspring distribution with $r/s = 0.2$. Lines: analytical results, Eq. (2.52). Markers: simulation results. Top (a): binary offspring distribution. Bottom (b): geometric offspring distribution.

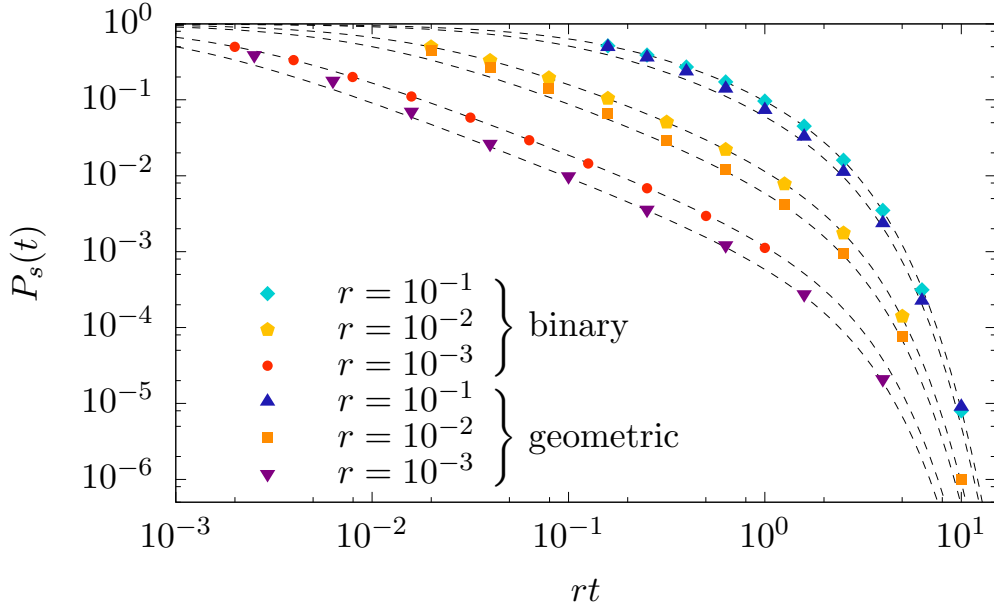


Figure 2.5: Survival probability for binary and geometric offspring distributions. Dashed lines: analytic results, which are exact for binary offspring distributions and which are approximations for geometric offspring distributions. Markers: simulation results. Figure adapted from [51].

It is shown for binary and geometric offspring distributions in Fig. 2.5 alongside simulation results, with which it agrees well. In particular, since it is derived from the probability distribution in Eq. (2.52), it also holds in the supercritical regime $r < 0$. How likely is it that a branching process survives indefinitely? For $r > 0$, when taking the limit $t \rightarrow \infty$, this eternal survival probability is found to be zero

$$r \geq 0 \quad \lim_{t \rightarrow \infty} P_s(t) = 0, \quad (2.56)$$

which means, in the subcritical regime, $r > 0$, all trajectories go extinct with probability 1. However, in the supercritical regime $r < 0$, the limit is

$$r < 0 \quad \lim_{t \rightarrow \infty} P_s(t) = -\frac{r}{q_2} \in (0, 1), \quad (2.57)$$

which is equal to $1 - p_0/p_2$ for binary offspring distributions (in the supercritical regime, $p_2 > p_0$). Hence, there is a finite probability of survival.

In order to find the answers to the second question, I can consider the time of death T

as a random variable with range $(0, \infty)$. For a specific realisation, if $T < t$, then the particle number is zero at time t , i.e. $N(t) = 0$. In particular

$$P(T < t) = P(N(t) = 0 | N(0) = 1), \quad (2.58)$$

which means that the probability that there is no particle in the system at time t is equal to the cumulative probability that the process died sometime before t . Hence, the probability density function $f_T(t)$ of the time of death is found by differentiating with respect to t :

$$r \geq 0 : \quad f_T(t) = \frac{d}{dt} P(T < t) = \frac{r e^{-rt} \left(1 + \frac{q_2}{r}\right)}{\left(1 + \frac{q_2}{r} (1 - e^{-rt})\right)^2} + \mathcal{O}(r^{-1}), \quad (2.59)$$

where the last equality is true for binary branching offspring distributions, but it is only an approximation for other offspring distributions. The probability density function $f_T(t)$ of the distribution of the times of death is plotted alongside simulation results in Fig. 2.6 for the binary offspring distributions. Simulations and analytical results agree well.

In the supercritical regime $r < 0$, $f_T(t)$ in Eq. (2.59) cannot be a probability density function as some trajectories survive forever. However, this only means that $f_T(t)$ is not correctly normalized in the supercritical regime and that $f_T(t)$ tacitly assumes the condition of eventual death of all trajectories. Hence, in the supercritical regime, among the trajectories that die, the time of death follows a distributions $f_T(t)$:

$$r < 0 : \quad f_T(t) = \frac{q_2}{q_2 + r} \frac{r e^{-rt} \left(1 + \frac{q_2}{r}\right)}{\left(1 + \frac{q_2}{r} (1 - e^{-rt})\right)^2}, \quad (2.60)$$

where the first fraction normalizes the probability density function and is equal to the inverse of the probability of death for any trajectory in the supercritical regime.

For the subcritical regime $r > 0$, using the probability density function f_T , Eq. (2.59), the expected time of death is calculated (using *Mathematica*):

$$r > 0 : \quad \mathbb{E}_T[T] = \frac{1}{q_2} \ln \left(1 + \frac{q_2}{r}\right), \quad (2.61)$$

which diverges at the critical point $r \rightarrow 0^+$ logarithmically. Eq. (2.61) is exact for

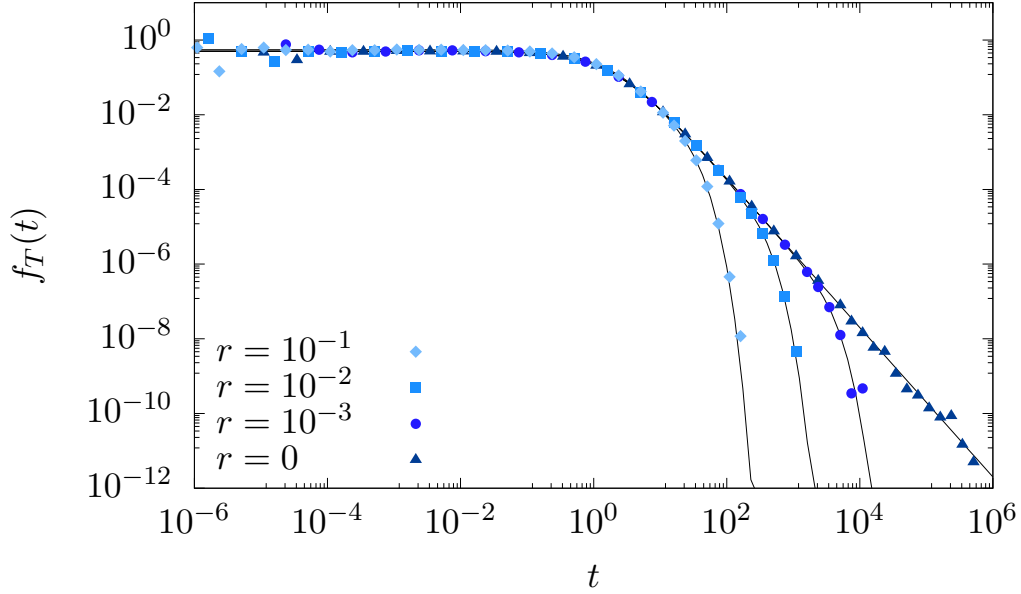


Figure 2.6: Distribution of the times of death for branching processes with several binary offspring distributions $r/s \in \{10^{-1}, 10^{-2}, 10^{-3}, 0\}$. Dashed line: analytical results, Eq. (2.59). Markers: simulation results. Figure adapted from [51].

binary offspring distributions. For other offspring distributions, it is an approximation. The expected time of death is shown in Fig. 2.7 alongside simulations for a wide range of values of $r/s \in [10^{-4}, 10^{-1}]$. Analytical results and simulations agree well for binary offspring distributions. However, for geometric offspring distributions, a systematic error can be observed. This error is due to omitted terms which are represented by Feynman diagrams with non-dyadic vertices. These terms include the parameters $q_3, q_4, \dots$ and take branching events into account where a particle splits into more than 2 particles. Because such events are under-accounted for, the analytic approximation underestimates the time of death. Nonetheless, the analytic approximation consistently tracks the simulated result and the relative error decreases (and is expected to vanish) as the critical point $r = 0$ is approached.

For the supercritical regime, among the trajectories that eventually die, the expected time of death is calculated using their conditional probability density function $f_T(t)$

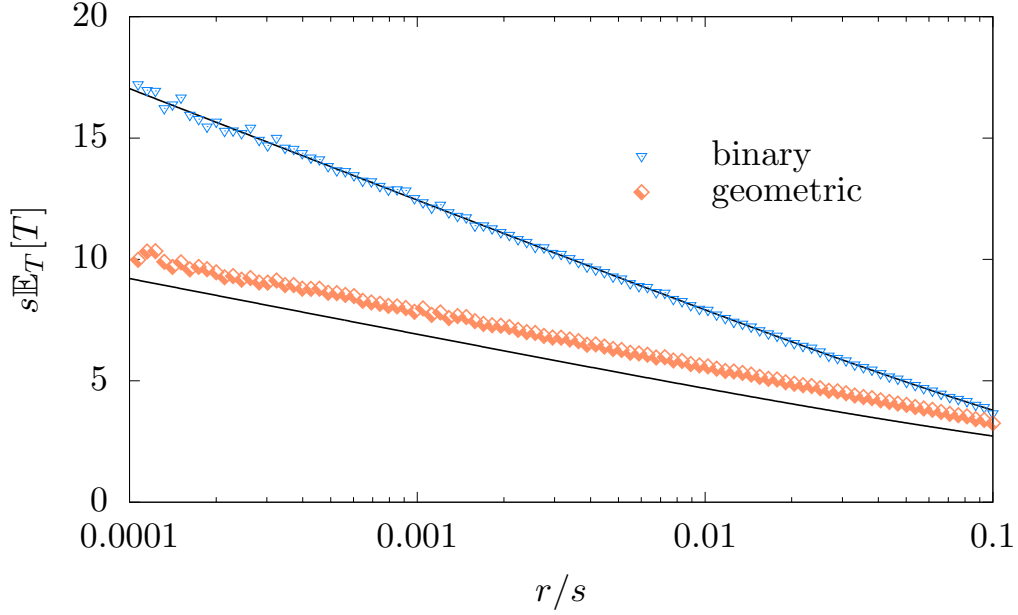


Figure 2.7: Rescaled expected time of death $s\mathbb{E}_T[T]$ over $r/s \in [10^{-4}, 10^{-1}]$ for binary and geometric offspring distributions. Straight line: analytical results, Eq. (2.61). Markers: simulation results.

from Eq. (2.60) (using *Mathematica*):

$$r < 0 : \quad \mathbb{E}_T[T] = \frac{1}{q_2 + r} \ln \left(-\frac{q_2}{r} \right), \quad (2.62)$$

which diverges logarithmically as $r \rightarrow 0^-$, i.e. as more and more long-lived trajectories eventually die.

2.6 TRAJECTORY SHAPE

In the subcritical regime, $r > 0$, all trajectories go extinct with probability 1 (i.e. almost surely), see Eq. (2.56). Depending on the choice of offspring distribution, the number of particles in the system can increase significantly in one instance, while it can decrease by only one particle at a time through the extinction process. Does this mean that particle numbers rise quickly and die slowly in a typical trajectory? Or do all the possible offspring events even out on average? In order to answer these questions, the expected trajectory shapes are studied in this section.

What is an expected trajectory shape and why is it different to the expected particle number, Eq. (2.31)? Here, I define the expected trajectory shape as the expected particle number under the condition of extinction at a specific time of death T . A trajectory shape is specific to the chosen time of death, for different times of deaths, different shapes occur.

How can the system be conditioned on a specific time of death? In deriving the formulation of the probability generating function in the language of the second quantization in Sec. 1.4, the *bra*-vector $\langle \star |$ was introduced in order to evaluate the probability generating function and its derivatives at $z = 1$, and thus calculate the moments of the random variable. If I want to retrieve only those trajectories which have gone extinct by time T , $\langle \star |$ has to be replaced by $\langle 0 |$. This *bra*-vector acts like a filter that only allows taking into account those realizations which have no particle in the system at the time when $\langle 0 |$ is imposed. Hence, the probability that a trajectory has gone extinct by time T is

$$P_e(T) = \langle 0 | e^{\mathcal{H}T} a^\dagger | 0 \rangle = 1 - P_s(T), \quad (2.63)$$

which I found using a different approach above, Eq. (2.55). The expected number of particles within the ensemble of trajectories which have gone extinct by time T is therefore

$$\mathbb{E}[N(t) | N(T) = 0, N(0) = 1] = \frac{\langle 0 | e^{\mathcal{H}(T-t)} a^\dagger a e^{\mathcal{H}t} a^\dagger | 0 \rangle}{\langle 0 | e^{\mathcal{H}T} a^\dagger | 0 \rangle}. \quad (2.64)$$

However, this includes trajectories which have gone extinct any time between initiation at time $t_0 = 0$ and time T . I would like to focus on those which have died exactly at time T . Hence, the infinitesimal difference between having died at time T and time

$T + \Delta T$ has to be considered:

$$\begin{aligned} \mathbb{E}[N(t)|N(T) = 0, \forall t' < T : N(t') > 0, N(0) = 1] = \\ = \lim_{\Delta T \rightarrow 0} \frac{\langle 0 | e^{\mathcal{H}(T+\Delta T-t)} a^\dagger a e^{\mathcal{H}t} a^\dagger | 0 \rangle - \langle 0 | e^{\mathcal{H}(T-t)} a^\dagger a e^{\mathcal{H}t} a^\dagger | 0 \rangle}{\langle 0 | e^{\mathcal{H}(T+\Delta T)} a^\dagger | 0 \rangle - \langle 0 | e^{\mathcal{H}T} a^\dagger | 0 \rangle} \end{aligned} \quad (2.65)$$

$$= \frac{\frac{d}{dT} \langle 0 | e^{\mathcal{H}(T-t)} a^\dagger a e^{\mathcal{H}t} a^\dagger | 0 \rangle}{f_T(T)} \quad (2.66)$$

$$=: V(t, T), \quad (2.67)$$

where I also introduced a shorter notation for the expected avalanche shape, $V(t, T)$.

Since the *bra*-vector $\langle \star |$ was used to derive the field-theoretic formulation, how can the vector $\langle 0 |$ be represented in the field theory? Both vectors are related as follows

$$\langle 0 | = \langle \star | e^{-a}, \quad (2.68)$$

which translates in the field theory to

$$\langle 0 | e^{\mathcal{H}(T-t)} a^\dagger a e^{\mathcal{H}t} a^\dagger | 0 \rangle = \left\langle e^{-\phi(T)} \phi^\dagger(t) \phi(t) \tilde{\phi}(0) \right\rangle. \quad (2.69)$$

Its calculation is explained in the following.

First, the exponential of the field $\phi(T)$ in Eq. (2.69) is written as a series whose terms are (up to a prefactor)

$$\left\langle \phi^k(T) \phi^\dagger(t) \phi(t) \tilde{\phi}(0) \right\rangle = \underbrace{\left\langle \phi^k(T) \phi(t) \tilde{\phi}(0) \right\rangle}_{I_A(k)} + \underbrace{\left\langle \phi^k(T) \tilde{\phi}(t) \phi(t) \tilde{\phi}(0) \right\rangle}_{I_B(k)}, \quad (2.70)$$

which splits into terms $I_A(k)$ and $I_B(k)$. The terms $I_A(k)$ can be represented by Feyn-

man diagrams as follows

$$I_A(k) \hat{=} \sum_{\ell=1}^k \sum_{j_1 \dots j_\ell} \binom{k}{j_1 \dots j_\ell} \left(\begin{array}{c} j_1 \vdots \\ j_\ell \vdots \end{array} \begin{array}{c} \bullet \\ \bullet \end{array} \begin{array}{c} \ell \vdots \\ \bullet \end{array} \right) \quad (2.71)$$

$$= \sum_{\ell=1}^k \sum_{j_1 \dots j_\ell} \binom{k}{j_1 \dots j_\ell} \frac{1}{\ell!} g_{j_1}(T-t) \dots g_{j_\ell}(T-t) g_{\ell+1}(t) \quad (2.72)$$

$$= k! e^{-rt} \left(\frac{q_2}{r} \right)^k (1 - e^{-rT})^k \left(\frac{u^2(k-1)}{(1+u)^2} + \frac{2u}{1+u} \right), \quad (2.73)$$

where the auxiliary variable u is introduced as

$$u = \frac{e^{-r(T-t)} - e^{-rT}}{1 - e^{-r(T-t)}}. \quad (2.74)$$

Line (2.72) is only exact for binary offspring distributions. For other offspring distributions, the ignored terms are of the order $\mathcal{O}(r^{-k+1})$, while the included ones are of the order $\mathcal{O}(r^{-k})$. Hence, this expression is only exact in the limit of criticality, $r \rightarrow 0^+$. The step from Eq. (2.72) to Eq. (2.73) is explained in the appendix, Sec. B.5.1.

Similarly, $I_B(k)$ is represented by the following Feynman diagram

$$I_B(k) \hat{=} \sum_{\ell=1}^k \sum_{j_1 \dots j_\ell} \binom{k}{j_1 \dots j_\ell} \left(\begin{array}{c} j_1 \vdots \\ j_{\ell-1} \vdots \\ j_\ell \vdots \end{array} \begin{array}{c} \bullet \\ \bullet \\ \bullet \end{array} \begin{array}{c} \ell-1 \vdots \\ \bullet \end{array} \right) \quad (2.75)$$

$$= \sum_{\ell=1}^k \sum_{j_1 \dots j_\ell} \binom{k}{j_1 \dots j_\ell} \frac{1}{(\ell-1)!} g_{j_1}(T-t) \dots g_{j_\ell}(T-t) g_\ell(t) \quad (2.76)$$

$$= \frac{k! e^{-rt}}{1 - e^{-rt}} \left(\frac{q_2}{r} \right)^{k-1} (1 - e^{-rT})^k \left(\frac{u^2(k-1)}{(1+u)^2} + \frac{u}{1+u} \right) \quad (2.77)$$

The step from Eq. (2.76) to Eq. (2.77) is explained in the appendix, Sec. B.5.2. The equality in line (2.76) is exact for binary offspring distributions. For other offspring distributions, the ignored terms are of the order $\mathcal{O}(r^{-k+2})$, while the included ones are of the order $\mathcal{O}(r^{-k+1})$. Hence, this expression is only exact at criticality, $r \rightarrow 0^+$.

Both results can be combined to obtain the expected number of particles in trajectories which die exactly at time T :

$$V(t, T) = 1 + 2\frac{q_2}{r} (1 - e^{-rt}) \left(1 - \frac{P_s(T)}{P_s(t)}\right). \quad (2.78)$$

Its calculation is explained in the appendix, Sec. B.5.3. The shapes $V(t, T)$ for different times of death T can be rescaled $\tau = t/T$ such that all rescaled shapes start at $\tau = 0$ and end at $\tau = 1$. For fixed q_2 and r the rescaled shapes change with T , which is shown in Fig. 2.8 for $r/s = 10^{-1}$ and $q_2/s = 0.45$. As the time of death is increased, the rescaled shapes approach an upper boundary:

$$\lim_{T \rightarrow \infty} V\left(\frac{T}{2}, T\right) = 1 + 2\frac{q_2}{r}, \quad (2.79)$$

which is exact for binary offspring distributions and an approximation for other offspring distributions. As the critical point $r = 0$ is approached, the upper boundary diverges as $\propto r^{-1}$.

At criticality, $r = 0$, the shape becomes a parabola:

$$\lim_{r \rightarrow 0^+} V(t, T) = 1 + 2\frac{q_2^2 T^2}{1 + q_2 T} \left(1 - \frac{t}{T}\right) \frac{t}{T}, \quad (2.80)$$

whose maximum at $t = T/2$ diverges linearly in T as $T \rightarrow \infty$.

Comparing analytical results to simulations of the expected shapes of trajectories under the condition of a specific time of death is very difficult because a specific time of death never occurs twice in simulations, almost surely, see Eq. (2.59). How can a comparison still be achieved?

If simulated shapes are

1. recorded, then

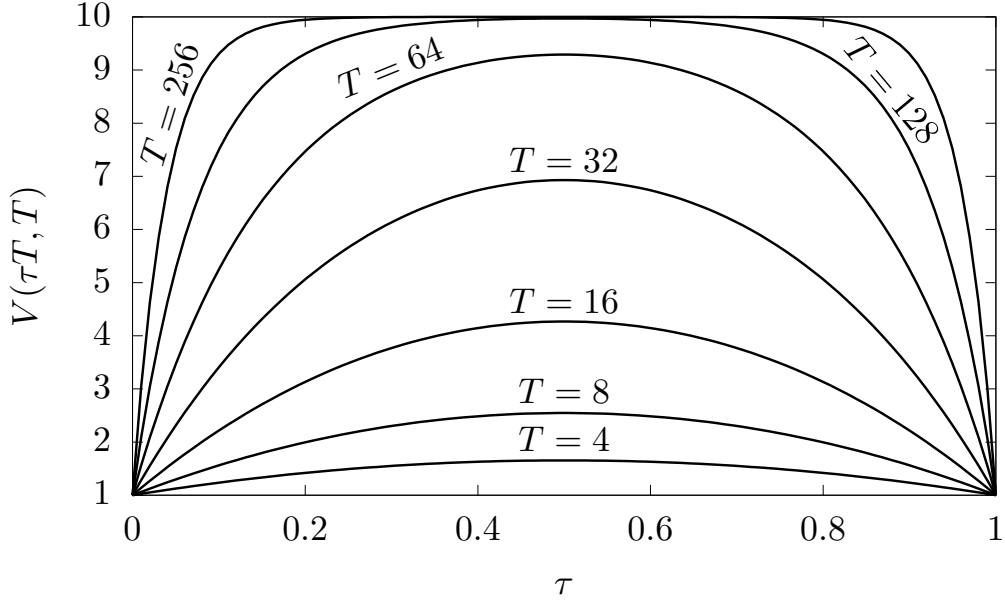


Figure 2.8: Expected shape of trajectory given death at time T for different $T/s \in \{4, 8, 16, 32, 64, 256\}$. Time is rescaled with $\tau = t/T$ such that time of death is at $\tau = 1$. Chosen parameters are $r/s = 10^{-1}$ and $q_2/s = 0.45$. As the time of death is increased, the shapes approach an upper boundary $\lim_{T \rightarrow \infty} V(\frac{T}{2}, T) = 1 + 2\frac{q_2}{r}$. Figure from [51].

2. rescaled with their time of death $\tau = t/T$, next
3. averaged over all times of death, and finally
4. normalized by area,

the resulting shapes can be compared to analytical results as follows. The likelihood of a time of death T is $f_T(T)$, Eq. (2.59). Hence, the T -averaged shape is proportional to

$$\mathbb{E}_T[V(\tau T, T)] \propto \int_0^\infty f_T(T) V(\tau T, T) dT \quad (2.81)$$

As I want the shapes to be normalized by area, the proportionality constant is $1/N_V$

with

$$N_V = \int_0^1 \int_0^\infty f_T(T) V(\tau T, T) dT d\tau. \quad (2.82)$$

Using *Mathematica*, the resulting averaged expected shapes can be expressed using the Gaussian hypergeometric functions ${}_2F_1(a, b, c, z)$

$${}_2F_1(a, b, c, z) = \sum_{k=0}^{\infty} \frac{a^{(k)} b^{(k)}}{c^{(k)}} \frac{z^k}{k!}, \quad (2.83)$$

where $a^{(k)} = a(a+1) \cdots (a+k-1)$ is the rising factorial:

$$\mathbb{E}_T[V(\tau T, T)] = \frac{1}{N_V} + \tau(1-\tau) \frac{q_2 F(\tau, q_2, r)}{(q_2 + r) N_V} \quad (2.84)$$

$$\text{with } F(\tau, q_2, r) = \frac{{}_2F_1\left(1, 2-\tau, 3-\tau, \frac{q_2}{q_2+r}\right)}{\tau-2} - \frac{{}_2F_1\left(1, 1+\tau, 2+\tau, \frac{q_2}{q_2+r}\right)}{\tau+1} \quad (2.85)$$

The normalisation constant and the function F diverge at the critical point, with the limit (obtained using *Mathematica*)

$$\lim_{r \rightarrow 0^+} \frac{F(\tau, q_2, r)}{N_V} = 6. \quad (2.86)$$

Hence, at criticality $r = 0$, the averaged expected shape is

$$\lim_{r \rightarrow 0^+} \mathbb{E}_T[V(\tau T, T)] = 6\tau(1-\tau). \quad (2.87)$$

The analytical result in Eq. (2.84) is compared to simulations in Fig. 2.9 for several values of r for the branching process with binary offspring distribution. The figure shows that the analytical results and the simulations agree well.

2.7 TIME-COVARIANCES

Other observables of interest are time-covariances. Intuitively, when a trajectory has a large number of particles at some point in time, it is very likely that it also had a large

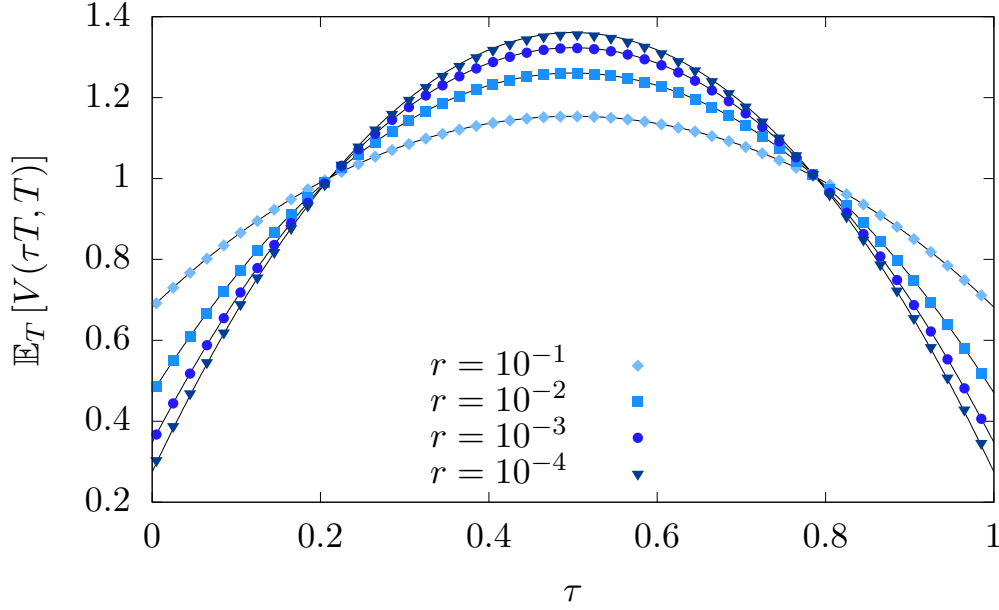


Figure 2.9: Rescaled trajectory shapes averaged over all possible times of death $T \in (0, \infty)$ for several $r/s \in \{10^{-1}, 10^{-2}, 10^{-3}, 10^{-4}\}$ using binary offspring distributions. Dashed lines: analytical result Eq. (2.84), Markers: simulation results. Figure adapted from [51].

number shortly before and will still have a similarly large number of particles shortly after. Is this intuition correct? Are the covariances symmetric before and after? How quickly do different time points in a trajectory become independent?

In order to investigate these questions, the 2- and 3-time covariances are calculated. The n -point covariance is defined as

$$\begin{aligned} \text{cov}\left(N(t_1), \dots, N(t_n) \middle| N(0) = 1\right) \\ = \mathbb{E}\left[\left(N(t_1) - \mathbb{E}[N(t_1)]\right) \cdots \left(N(t_n) - \mathbb{E}[N(t_n)]\right) \middle| N(0) = 1\right], \end{aligned} \quad (2.88)$$

where the inner expectations are also conditioned on $N(0) = 1$.

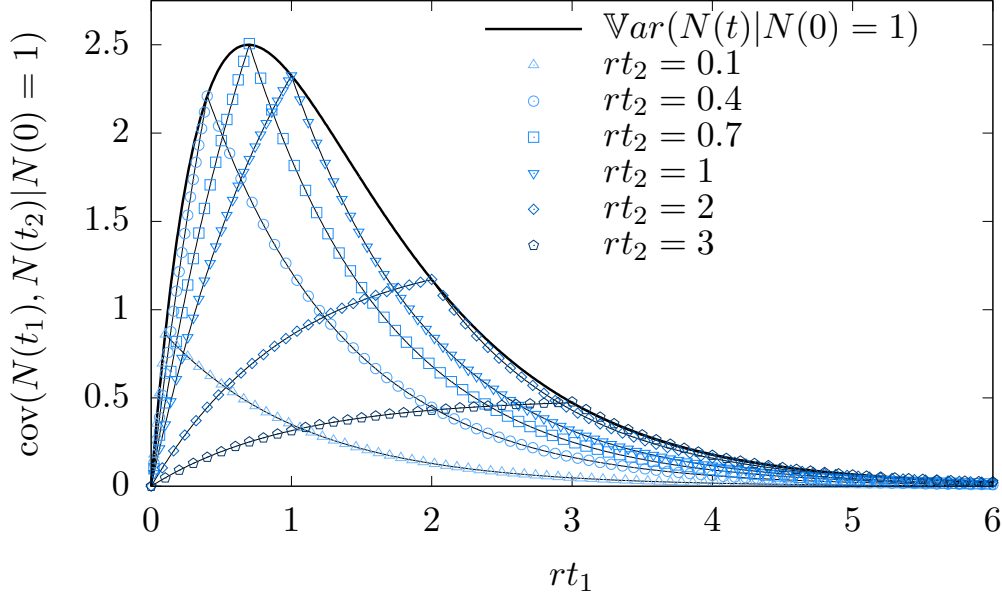


Figure 2.10: Variance (straight black line) and 2-point covariance (dashed lines) for several times of the branching process with binary offspring distribution. Markers: simulation results. Figure adapted from [51].

2.7.1 2-TIME COVARIANCE

For the 2-point covariance, the term $\mathbb{E}[N(t_1)N(t_2)|N(0) = 1]$ has to be calculated, which, for $t_2 > t_1$, is equal to

$$\mathbb{E}[N(t_1)N(t_2)|N(0) = 1] = \langle \star | a^\dagger a e^{\mathcal{H}(t_2-t_1)} a^\dagger a e^{\mathcal{H}t_1} a^\dagger | 0 \rangle \quad (2.89)$$

$$= \langle \phi(t_2) \tilde{\phi}(t_1) \rangle \langle \phi(t_1) \tilde{\phi}(0) \rangle + \langle \phi(t_2) \phi(t_1) \tilde{\phi}(0) \rangle. \quad (2.90)$$

While the first part is known from the calculation of the first moment, Eq. (2.31), the second part can be determined to be equal to

$$\langle \phi(t_2) \phi(t_1) \tilde{\phi}(0) \rangle = \Theta(t_1) \Theta(t_2) 2 \frac{q_2}{r} e^{-r(t_1+t_2)} (e^{r \min\{t_1, t_2\}} - 1) \quad (2.91)$$

for any relation between t_1 and t_2 . The details of its derivation are shown in the appendix, Sec. B.1.

Hence, the 2-point covariance is equal to

$$\begin{aligned} \text{cov}\left(N(t_1), N(t_2) | N(0) = 1\right) &= \\ &= \Theta(t_1)\Theta(t_2)e^{-r \max\{t_1, t_2\}} \left(1 + 2\frac{q_2}{r}\right) (1 - e^{-r \min\{t_1, t_2\}}), \end{aligned} \quad (2.92)$$

which has an unsurprising similarity to the expression for the variance, Eq. (2.38). The analytical results for the covariance are shown for several times t_1 and t_2 in Fig. 2.10 for binary offspring distribution alongside simulation results. The figure shows that simulations and analytical results agree very well.

2.7.2 3-TIME COVARIANCE

The 3-time covariance is defined as (all expectations are assumed to be conditioned on $N(0) = 1$):

$$\begin{aligned} \mathbb{E}\left[\left(N(t_1) - \mathbb{E}[N(t_1)]\right)\left(N(t_2) - \mathbb{E}[N(t_2)]\right)\left(N(t_3) - \mathbb{E}[N(t_3)]\right)\right] &= \\ &= \mathbb{E}[N(t_1)N(t_2)N(t_3)] - \mathbb{E}[N(t_1)N(t_2)]\mathbb{E}[N(t_3)] \\ &\quad - \mathbb{E}[N(t_1)N(t_3)]\mathbb{E}[N(t_2)] - \mathbb{E}[N(t_2)N(t_3)]\mathbb{E}[N(t_1)] \\ &\quad + 2\mathbb{E}[N(t_1)]\mathbb{E}[N(t_2)]\mathbb{E}[N(t_3)] \end{aligned} \quad (2.93)$$

which is even more complicated once each term is expressed in the field theory. For example

$$\begin{aligned} \mathbb{E}[N(t_1)N(t_2)N(t_3)] &= \langle \phi(t_3)\phi(t_2)\phi(t_1)\tilde{\phi}(0) \rangle \\ &\quad + \langle \phi(t_3)\phi(t_2)\tilde{\phi}(t_1)\phi(t_1)\tilde{\phi}(0) \rangle \\ &\quad + \langle \phi(t_3)\tilde{\phi}(t_2)\phi(t_2)\phi(t_1)\tilde{\phi}(0) \rangle \\ &\quad + \langle \phi(t_3)\tilde{\phi}(t_2)\phi(t_2)\tilde{\phi}(t_1)\phi(t_1)\tilde{\phi}(0) \rangle \end{aligned} \quad (2.94)$$

for the time ordering $t_3 > t_2 > t_1$. Furthermore, each of these terms is lengthy. For example, the first term in Eq. (2.94) is equal to

$$\begin{aligned} \left\langle \phi(t_3)\phi(t_2)\phi(t_1)\tilde{\phi}(0) \right\rangle &= \Theta(t_1)\Theta(t_2)\Theta(t_3)e^{-r(t_1+t_2+t_3)} \left(\right. \\ &\quad \frac{4q_2^2}{r^2} \left((e^{r \min\{t_1, t_2, t_3\}} - 1) (e^{r \min\{t_1, t_2\}} + e^{r \min\{t_1, t_3\}} + e^{r \min\{t_2, t_3\}}) \right. \\ &\quad \left. \left. - \frac{3}{2} (e^{2r \min\{t_1, t_2, t_3\}} - 1) \right) + \frac{3q_3}{r} (e^{2r \min\{t_3, t_2, t_1\}} - 1) \right) \end{aligned} \quad (2.95)$$

Its derivation is shown in the appendix, Sec. B.2. Therefore, I conclude that useful approximations are needed.

2.7.3 n -TIME COVARIANCE APPROXIMATION

In the previous subsection, it became clear that expressions for the n -time covariances can be quite complicated and lengthy. Therefore, I seek an approximation, analogously to the approximation of higher moments in Sec. 2.3.3 for $r \rightarrow 0^+$. In fact, I can repeat the argument in Sec. 2.3.3 and find that close to criticality $r = 0$, the n -time covariance is dominated by

$$\begin{aligned} \mathbb{E}[(N(t_1) - \mathbb{E}[N(t_1)]) \cdots (N(t_n) - \mathbb{E}[N(t_n)])] &= \left\langle \phi(t_n) \cdots \phi(t_1) \tilde{\phi}(0) \right\rangle \Big|_{\substack{j \geq 3 \\ q_j = 0}} \\ &\quad + \mathcal{O}(r^{-n+2}) \end{aligned} \quad (2.96)$$

where all expectations are conditioned on $N(0) = 1$.

The expression on the right hand side is denoted by $\zeta_n(t_1, \dots, t_n)$ and is represented by

the following Feynman diagram

$$\zeta_n(t_1, \dots, t_n) \hat{=} \sum_{k=1}^{n-1} \sum_{\substack{\sigma \subset \{t_1, \dots, t_n\} \\ |\sigma|=k}} \left(\begin{array}{c} \sigma : \\ \sigma^c : \end{array} \begin{array}{c} \text{Feynman diagram with two blue nodes and red lines} \end{array} \right) \quad (2.97)$$

$$\begin{aligned} &= q_2 \sum_{k=1}^{n-1} \sum_{\substack{\sigma \subset \{t_1, \dots, t_n\} \\ |\sigma|=k}} \int_0^{t_{\min}} \zeta_k(t_{\sigma(1)} - t', \dots, t_{\sigma(k)} - t') \\ &\quad \cdot \zeta_{n-k}(t_{\sigma^c(k+1)} - t', \dots, t_{\sigma^c(n)} - t') e^{-rt'} dt', \end{aligned} \quad (2.98)$$

where $t_{\min} = \min\{t_1, \dots, t_n\}$.

While expression (2.98) is difficult to calculate explicitly, it can be used to calculate the distribution of the sizes of trajectories, which is shown in the next section.

2.8 TOTAL SIZE

The final question of the pure branching process that I want to address is: How large are trajectories, what is their typical size? The notion of size combines two properties of trajectories: its number of particles at different times and its length, i.e. time of death. I define the size of a trajectory as follows

$$S = \int_0^\infty s N(t) dt, \quad (2.99)$$

where the factor s is used to make the result independent of the branching event frequency, i.e. independent of a rescaling of time. S is a random variable and the aim of this section is to derive its distribution.

First, I calculate the moments of S , i.e. $\mathbb{E}[S]$, $\mathbb{E}[S^2]$, $\dots$:

$$\mathbb{E}[S^n] = s^n \int_0^\infty \dots \int_0^\infty \mathbb{E}[N(t_1) \dots N(t_n) | N(0) = 1] dt_n \dots dt_1. \quad (2.100)$$

Going back to the dimensional arguments in Sec. 2.3.3, close to the critical point,

$\mathbb{E}[N(t_1) \cdots N(t_n) | N(0) = 1]$ will be dominated by those terms which are represented by dyadic Feynman diagrams:

$$\mathbb{E}[N(t_1) \cdots N(t_n) | N(0) = 1] \approx \zeta_n(t_1, \dots, t_n) \quad (2.101)$$

Hence, the n th moment of the total size of the trajectories is

$$\mathbb{E}[S^n] \approx s^n \int_0^\infty \cdots \int_0^\infty \zeta_n(t_1, \dots, t_n) dt_n \cdots dt_1 \quad (2.102)$$

$$= \frac{q_2}{r} \sum_{k=1}^{n-1} \binom{n}{k} \mathbb{E}[S^k] \mathbb{E}[S^{n-k}] \quad (2.103)$$

$$= \frac{s^n q_2^{n-1}}{r^{2n-1}} 2^{n-1} (2n-3)!!, \quad (2.104)$$

where $(2n-3)!! = (2n-3)(2n-5) \cdots 3 \cdot 1$ is the double factorial of $(2n-3)$. The calculation from Eq. (2.102) to Eq. (2.103) and to Eq. (2.104) is shown in the appendix, Sec. B.6. This result is only exact in the limit $r \rightarrow 0^+$.

Analogously to the universal ratios of moments in Eq. (2.49) and the universal ratios of probabilities in Eq. (2.53), universal ratios of the total size of the trajectories are found:

$$\frac{\mathbb{E}[S^k] \mathbb{E}[S^\ell]}{\mathbb{E}[S^{k+m}] \mathbb{E}[S^{\ell-m}]} = \frac{(2k-3)!!(2\ell-3)!!}{(2(k+m)-3)!!(2(\ell-m)-3)!!}, \quad (2.105)$$

with $k, \ell, m \in \mathbb{N}$ and $m < \ell$.

Hence, the moment generating function of S is

$$M_S(z) = \mathbb{E}[e^{zS}] \simeq 1 + \frac{r - \sqrt{r^2 - 4sq_2z}}{2q_2}, \quad (2.106)$$

which is derived in the appendix, Sec. B.6.1. From moment generating function, the probability density function of S , $f_S(x)$, can be found by calculating the inverse Laplace transform of $M(-z)$:

$$f_S(x) \simeq \frac{1}{2} \sqrt{\frac{s}{q_2\pi}} x^{-\frac{3}{2}} e^{-\frac{r^2 x}{4q_2 s}}, \quad (2.107)$$

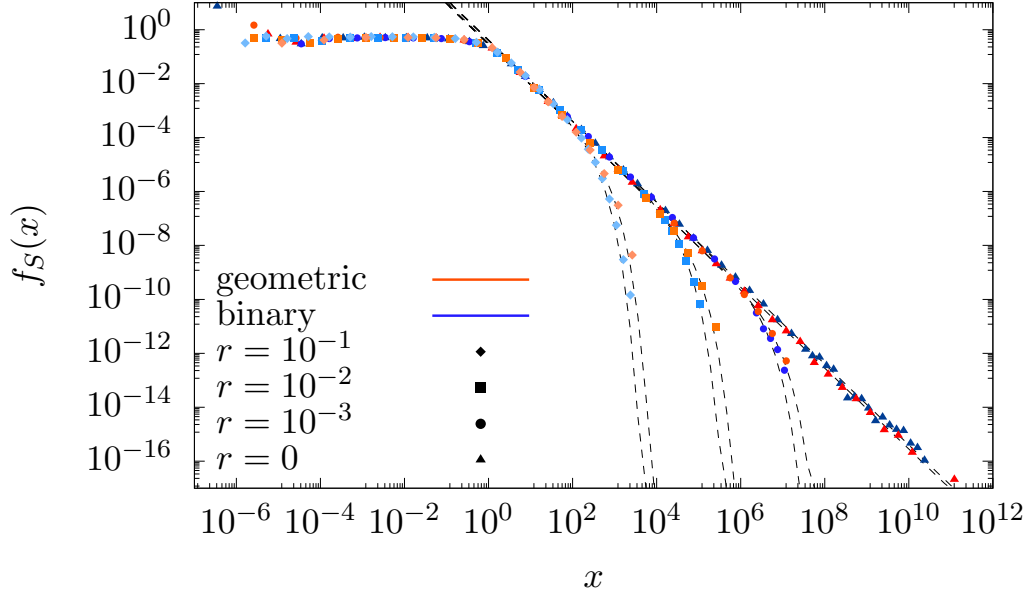


Figure 2.11: Distribution of the total size of trajectories for binary and geometric offspring distributions for $r/s \in \{10^{-1}, 10^{-2}, 10^{-3}, 0\}$. Dashed line: analytical approximation. Markers: simulation results. Figure adapted from [51].

which is shown for several rates r for binary and geometric offspring distributions in Fig. 2.11. The probability density function $f_S(x)$ is an approximation which is expected to be good for $x \geq 1$, while it is expected to be bad for $x < 1$. The case $x > 1$ is dominated by trajectories that actually branch into more than one particle before dying, which is captured well by dyadic diagrams. However, the case $x < 1$, is dominated by trajectories that were initialized, never branched and then died. These trajectories are captured by the terms represented by disconnected diagrams, which I omitted in the approximation in Eq. (2.101).

Nevertheless, the approximation of $f_S(x)$ captures two important characteristics: the exponential decay in the subcritical regime $r > 0$ and the power law scaling with power $-3/2$ at the critical point $r = 0$.

2.9 CONCLUSION

Continuous-time branching processes are modelled in the subcritical regime using a Doi-Peliti field theory. A variety of properties of the branching processes were analyzed, of which an overview is given in Table. 2.1 on page 108. All observables are exact asymptote for the critical limit $r \rightarrow 0^+$. Most analytical results are corroborated with simulations of binary and geometric offspring distributions.

An important result is that close to criticality $r \rightarrow 0^+$, all the studied observables are determined, up to rescaling of time, by the first and second moment of the offspring distribution only, which are part of the parameters r and q_2 . Higher moments of the offspring distribution become negligible at criticality. At the basis of this result is the dimensional argument presented in Sec. 2.3.3.

The Doi-Peliti field theory for the continuous-time branching process is also at the basis of future work, of which I outline two projects in the following subsections.

2.9.1 OUTLOOK: NOISY PARAMETERS

Branching processes are used as models for a variety of applications. Their interpretation as real-life systems has the drawback that most systems are noisy. For example, when interpreted as the reproduction in populations, the branching process set out in this Chapter cannot account for seasonal variations, the influence of epidemics, food supply fluctuations or other influences on fertility rates.

Therefore, it is desirable to model branching processes whose offspring distribution fluctuates in time. In order to achieve this, the *Martin-Siggia-Rose-Janssen-DeDominicis* (MSRJD) response field formalism can be used [98, 31, 76]. This formalism can be used to transform the description of a stochastic process by a Langevin equation to a description by a field theory. For example, the Ornstein-Uhlenbeck process [160] can be described by the following Langevin equation:

$$\frac{d}{dt}y(t) = -\beta y(t) + \xi, \quad (2.108)$$

where $y(t)$ is the time-dependent random variable of interest, $\beta > 0$ is the return frequency modelling a harmonic potential and ξ is time-dependent white noise with

mean zero and covariance $\text{cov}(\xi(t), \xi(t')) = D\delta(t - t')$, $D > 0$.

Following the MSRJD formalism as described in *Täuber's* book [154], a path integral is derived with the following action:

$$\mathcal{A}[\tilde{\psi}, \psi] = \int \tilde{\psi} \left(-\frac{d}{dt} - \beta \right) \psi + D\tilde{\psi}^2 dt, \quad (2.109)$$

where ψ describes the value of the Ornstein-Uhlenbeck field and $\tilde{\psi}$ is its response field.

Such a noise field can be coupled to parameters of the branching process. However, it is unclear how such noisy parameters influence the branching process. Does the critical point shift, do observables change? These are questions to be answered in future work.

2.9.2 OUTLOOK: COUPLED BRANCHING PROCESSES

A common observation in neuroscience is that activities in the brain, measured as local field potentials at electrodes inserted into the live brain or into brain slices [7, 126, 127], show dynamics similar to trajectories of branching processes close to criticality. Different activities such as deep sleep or wakefulness lead to different statistics of the trajectories and the dynamics appear to be closer or further away from a critical point. Different areas of the brain might be in different activity states. However, they are connected and can influence each other. It therefore seems natural to ask how several branching processes, at different distances from the critical point, influence each other when coupled.

A different interpretation of coupled branching processes arises in the context of mutations within a population. Different genetic mutations can lead to different fertility rates. However, mutations are also reversible. Thus two branching processes, randomly exchanging particles can be used to model the competition between alleles [106].

The coupling of branching processes can be achieved within the framework of Doi-Peliti field theory. If the processes are simply exchanging particles as a Poisson process, then the exchange is analogous to a continuous-time Markov chain. Assuming that there are m different branching processes with their offspring distributions $p_{k,m}$, their Poissonian

exchange of particles can be described by the following master equation:

$$\begin{aligned} \frac{d}{dt}P(\{n\}, t|\{n_0\}, t_0) = & \sum_{i \neq j} \lambda_{ij} \left((n_i + 1)P(\{n_i + 1, n_j - 1\}, t|\{n_0\}) \right. \\ & \left. - n_i P(\{n\}, t|\{n_0, t_0\}) \right) \\ & + \sum_{i=1}^m (\text{branching process } i), \end{aligned} \quad (2.110)$$

where $n_1, \dots, n_m$ are the particle numbers of the m branching processes, $\{n_0\}$ is the initial state at time $t_0 < t$ and $\{n\}$ is a shorthand for $n_1, \dots, n_m$. The rates λ_{ij} are the rates for the exponentially distributed waiting time of one particle in branching process i to transfer to branching process j .

The master equation (2.110) is transformed into the following action of a Doi-Peliti field theory:

$$\mathcal{A} = \sum_{i=1}^m \int \tilde{\phi}_i \left(-\frac{d}{dt} - r_i - \sum_{j \neq i} \lambda_{ij} \right) \phi_i + \sum_j \lambda_{ij} \tilde{\phi}_j \phi_i + \sum_{\ell=2}^{\infty} q_{\ell,i} \tilde{\phi}_i \phi_i dt, \quad (2.111)$$

where $q_{\ell,i}$ and r_i are the coefficients q_ℓ and r of branching process i , see Eq. (2.18).

Future work will focus on determining the critical hypersurface of these coupled branching processes.

Other ways to couple branching processes are also feasible. For instance, instead of exchanging particles, the branching processes can also influence each others branching probabilities, which will be investigated in the future as well.

Furthermore, the connection to evolutionary dynamics, modelling competitions of alleles, raises another prospect for future research: evolutionary dynamics is modelled often using games theory [106, 107, 135]. Therefore, future work will focus on: Can field theory be a useful tool for game theory?

Table 2.1: Observables of the continuous time branching process, which were calculated, including their corresponding section and equation numbers. The accuracy of the results is labeled in several ways: Is the result exact for all offspring distributions (OD)? Is it exact for binary offspring distribution (BOD) only? Is it asymptotically exact (AE) $r \rightarrow 0^+$ for all offspring distributions? Was the asymptotic controlled (AC), i.e. is the next term of the order of some power in r^{-1} ? Were the analytical results directly corroborated with simulations (S)? Table adapted from [51].

Observable	Sec.	Eq.	Fig.	OD	BOD	AE	AC	S
1st moment	2.3.1	(2.31)	2.3	✓	✓	✓	✓	✓
2nd moment	2.3.1	(2.37)	2.3	✓	✓	✓	✓	✓
3rd moment	2.3.2	(2.42)	2.3	✓	✓	✓	✓	✓
n th moment	2.3.3	(2.48)	2.3	-	-	✓	✓	✓
n th factorial moment	2.3.3	(2.48)	-	-	✓	✓	✓	-
probability of particle number	2.4	(2.52)	2.4	-	✓	✓	✓	✓
survival probability	2.5	(2.55)	2.5	-	✓	✓	✓	✓
probability density of trajectory duration	2.5	(2.59)	2.6	-	✓	✓	✓	✓
expected time of death	2.5	(2.61)	2.7	-	✓	✓	-	✓
expected trajectory shape	2.6	(2.78)	2.8	-	✓	✓	-	-
averaged expected trajectory shape	2.6	(2.84)	2.9	-	✓	✓	-	✓
2-time covariance	2.7.1	(2.92)	2.10	✓	✓	✓	✓	✓
3-time covariance approximation	2.7.2	(2.95)	-	-	-	✓	✓	-
n -time-covariance approximation	2.7.3	(2.98)	-	-	-	✓	✓	-
moments of total size	2.8	(2.104)	-	-	✓	✓	✓	-
moment generating function of total size	2.8	(2.106)	-	-	✓	✓	-	-
probability density of total size	2.8	(2.107)	2.11	-	✓	✓	-	✓

3

FILAMENT SELF-ASSEMBLY

ABSTRACT

In this chapter, the reaction-diffusion system of actin filament and microtubule self-assembly is studied using Doi-Peliti field theory. The results up to Sec. 3.3.2 are also part of an article, written in collaboration with G. Pruessner, which was published in the *Journal of Statistical Mechanics: Theory and Experiment* on 23 May 2019 [118]. I was involved in all calculations presented here.

New about the research results is that they describe the interplay of reactions at and diffusion around the filament tip and characterizes emergent phenomena, which provide a better explanation for specific experimental results, see Sec. 3.3.3, and suggest further observables for experimental verification, see Sec. 3.6.

3.1 INTRODUCTION

The smallest units of life are cells. They occur as single-cell organisms with relatively simple modes of operation and as part of large, multi-cellular organisms with complex behaviours. Typically, cells span tens of micrometers. However, some specialized cells can have extensions that are longer than one meter. Their internal organisation is similar across many species. One of the parts that appears, in different variants, in all cells are filaments, which are polymer structures and which are often grouped into three types: long microtubules, short actin filaments and intermediate filaments. While microtubules are found in all organisms, actin filaments are not found in bacteria, and intermediate filaments are only found, with few exceptions, in larger, multi-cellular organisms. In cells, filaments grow and shrink dynamically and are involved in many vital processes inside the cell. This includes: maintaining or changing the cell shape, movement of the cell, cell division and transport processes between different parts of the cell [124]. In some specialized cells, filaments are part of the specialized structure. For example in sperm cells, microtubules are part of the tail of sperm cells, responsible for the swimming of the cell. In neurons, i.e. nerve cells, dynamic actin filament growth is important for connecting two neurons to form a neural network. Furthermore, neural signals are propagated as electric potentials along microtubules that form long extensions, called axons, of the cell [81]. In addition, microtubules are also studied as elements in active fluids [150]. The building blocks of microtubules are tubulin dimers which are about 8 nm long and form protofilaments of which 13 arrange into a cylinder-like structure [96]. The building blocks of actin filaments are actin monomers which have a length of about 7 nm and which form two strands, which together make up the actin filament [40].

The self-assembly of actin filaments and microtubules has been studied intensively experimentally [54, 50, 123, 87, 49] and theoretically [103, 48, 25, 26, 164, 5, 108]. However, open questions about their self-assembly dynamics remain. One central question is: is their self-assembly limited by diffusion of their building blocks to the tip [108, 38] or by the reactions for their incorporation into the filament [5, 38]? This question is addressed in this chapter.

In general, the building blocks of the cell's filaments are moving diffusively inside the cell plasma. They attach and detach from the filament tip at random, which creates a

random growth and shrinking process of which the mean and variance are of particular interest. In microtubules, there is the additional phenomenon of catastrophes, which are sudden, very fast shrinking events. During a catastrophe, a microtubule might reduce its length by more than 50% within a short time. However, catastrophes occur rarely and this chapter is only concerned with the filament dynamics between catastrophes. Actin filaments do not exhibit catastrophes. [103]

The growth speed v of the filaments is observed to scale linearly with the bulk density ζ of the building blocks. However, there is a bulk density where stochastic assembly and disassembly balance each other, on average. The maximal disassembly speed is measured in an environment that does not contain any building blocks. This linear relationship is written as

$$\langle v \rangle = h(k_{\text{on}}\zeta - k_{\text{off}}), \quad (3.1)$$

where h is the average step size of the growth and shrinking per incorporated particle. The parameters k_{on} and k_{off} are the incorporation rate per bulk density and the release rate. Eq. (3.1) is the relation ship that is observed and k_{on} and k_{off} are determined in experiments. Also observed is the variance of the self-assembly by measuring the mean square displacement of the filament tip [54] or by creating histograms of the length fluctuations [50, 87, 54]. It approximately grows linearly in time and its proportionality constant is described as an effective diffusion constant D_{eff} .

An important goal has been to understand both the growth speed and length fluctuations with one model. There have been two major competing assumptions: either the system is reaction-limited or the system is diffusion-limited.

The assumption that the system is reaction-limited implies that the supply of the building blocks (i.e. actin monomers or tubulin dimers) by diffusion is sufficiently fast, such that local depletion can be neglected. In particular, this means that the time scale of the reactions (proportional to the inverse of the reaction rates k^{-1}) is slower than the average time it takes a particle to move the average distance between two particles, which is proportional to $D^{-1}\zeta^{-2/3}$, where D is the diffusion constant and ζ is the bulk density of the particles. Hence, a system can be placed closer to reaction-limited dynamics if the bulk density ζ is increased or if the diffusion constant is increased (e.g. by changing the viscosity [166]).

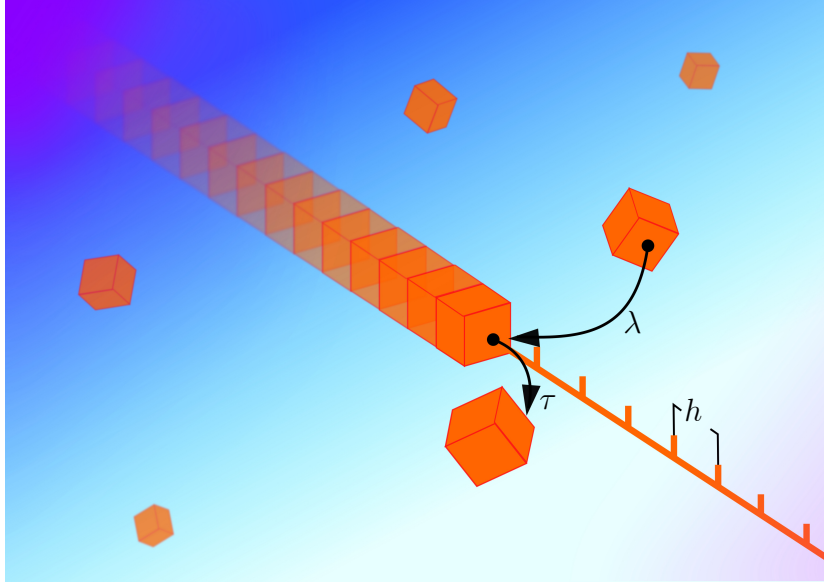


Figure 3.1: A schematic of the microtubule self-assembly process. Tubulin / actin (red blocks) moves diffusively in $\mathbb{R}^3$, while the microtubule / actin filament tip is fixed on a lattice with spacing h . Tubulin / actin can be incorporated in the microtubule / actin filament (coefficient λ) and released from its tip (rate τ). Figure from [118].

In the reaction-limit, the incorporation and release of particles from the filament tip are independent Poisson processes with exponential waiting times, whose rates are denoted by $\lambda\zeta$ for incorporation and τ for release. Incorporation is proportional to the bulk particle density ζ . The competing processes together form a Skellam process with a Skellam distribution [145]. The resulting mean growth speed is equal to

$$\langle v \rangle_R = h(\lambda\zeta - \tau), \quad (3.2)$$

where h is the average step size of the growth and shrinking per incorporated or released particle. The variance of the position grows linearly in time with effective diffusion constant D_{eff} :

$$D_{\text{eff},R} = h^2(\lambda\zeta + \tau). \quad (3.3)$$

If the system were reaction limited, then the measured coefficients k_{on} , k_{off} and D_{eff} would be equal to λ , τ and $D_{\text{eff},R}$. However, the experimentally determined variance is

much larger than expected [50, 87, 54] in reaction-limited dynamics.

The alternative, common assumption is that the system is diffusion-limited. Here, average reaction times are assumed to be much faster than the time to diffusively fill a depleted area with particles from the bulk. Growth of the filament is determined by waiting times for new building blocks to arrive at the filament tip. On a mean field level, transport in a diffusive environment only goes from higher concentrations to lower concentrations. As the filament tip is acting as a sink of actin monomers or tubulin dimers, in the mean field picture transport only goes to the tip and is described by a diffusion equation. Diffusion does not only refer to position in space but also to orientation of the molecules in phase space and can be accommodated by an effective reaction radius in position space [10, 105].

In this scenario, growth speed $\langle v \rangle_D$ is determined by the diffusive flux J to the reaction sphere around the tip of effective radius R :

$$\langle v \rangle_D = 4\pi DRh\zeta. \quad (3.4)$$

This is equivalent to the result of Smoluchowski for coagulation [146] if any orientation of the building blocks would lead to incorporation into the tip. However, the orientation does matter. To take this into account, the effective reaction radius is considered to be smaller than the actual particle size [10]. If the system is diffusion-limited, then the effective incorporation rate is equal to the volume flux $k_{\text{on}} = 4\pi DR$. However, this limit cannot accommodate an effective release rate k_{off} as any released particle would immediately be pushed back to the tip by the diffusive flux. Thus a perfect diffusion limit cannot explain the observations, Eq. (3.1).

Furthermore, following the Stokes-Einstein relation for small Reynolds numbers [151], viscosity and incorporation rate should be inversely proportional without offset. This relationship was experimentally tested in [38] for actin filaments and in [166] for microtubules. In both cases, offsets for the relationship were found, implying that the dynamics is not purely diffusion-limited. However, the incorporation rate does change with the viscosity, which also disproves the reaction-limited dynamics assumption.

In addition, the diffusion equation, as a mean-field theory, does not give any information about fluctuations of the flux. It is therefore impossible to derive the effective diffusion constant D_{eff} of the filament length fluctuations.

In order to go beyond mean-field approaches and combine the stochasticity of reactions and diffusion, we set up a Doi-Peliti field theory, which is explained in the following.

3.2 DERIVATION OF THE MODEL

The dynamic process of microtubule and actin filament self-assembly can be modelled on a three-dimensional lattice and described by a master equation, see appendix, Eq. (C.3). This master equation model is transformed into a Doi-Peliti field theory, which is explained in general in Secs. 1.2 to 1.5. The main steps for obtaining the action of the field theoretic model of filament self-assembly are outlined in the appendix, Sec. C.1 and C.2.

In our model, tubulin dimers, actin monomers, the microtubule tip and actin filament tip are represented by point-particle fields, which are interpreted as time-dependent probability distributions of their positions in space. Because of the representation as fields, they do not have a size. However, the finite size of particles is an important element of the step-wise filament growth. To accommodate this step-wise growth, the model uses a hybrid, three dimensional space. Particles move in a continuous space $\mathbb{R}^3$ and filaments grow along a discrete line in $\mathbb{Z}^3$ with lattice constant h , see Fig. 3.1 for a schematic of the situation. That the growth is forced on a perfectly straight line can be justified by considering the filament's persistence length and rigidity. The persistence length P and flexural rigidity K have been measured to be $P \approx 5200 \mu\text{m}$ and $K \approx 2 \cdot 10^{-23} \text{ Nm}^2$ for microtubules and $P \approx 18 \mu\text{m}$ and $K \approx 7 \cdot 10^{-26} \text{ Nm}^2$ for actin filaments [57]. The origin, directions and lengths of the continuous space and the lattice coincide. As we are modelling only a single polymer instead of the 13 protofilaments that are typically found in a microtubule [96] or 2 strands in actin filaments [40], we interpret the lattice spacing h as the effective length by which a filament grows when a single particle attaches, which corresponds to the actual particle size h_{real} divided by thirteen for microtubules or divided by 2 for actin filaments.

Tubulin dimers and actin monomers will be jointly referred to as particles, while the microtubule and the actin filament will be simply called filament.

The particle fields and the filament tip fields exist in two versions: an annihilation field and a creation field. For particles, they are denoted by $\varphi(x, t)$ and $\varphi^\dagger(x, t)$, respectively,

with spatial argument $x \in \mathbb{R}^3$ and time dependence t . For the filament tip, they are denoted by $\psi_j(t)$ and $\psi_j^\dagger(t)$, respectively, with time dependence t and spatial argument $j \in \mathbb{Z}^3$. Both creation fields also appear as Doi-shifted fields [37], e.g. $\varphi^\dagger(x, t) = \tilde{\varphi}(x, t) + 1$. In addition, the particle annihilation field is shifted to measure deviations from the bulk density ζ , i.e. $\varphi(x, t) = \check{\varphi}(x, t) + \zeta$.

The fields' propagation and interactions represent six microscopic processes of the reaction-diffusion system. In the following list of these processes, the units of their coefficients are denoted by [...]. A unit of time is written as T ; a unit of length is written as L .

- *Particle diffusion* in $\mathbb{R}^3$ with diffusion constant D , $[D] = T^{-1}L^2$.
- *Particle adsorption* by the filament tip with coefficient λ and subsequent movement of the tip by distance h in the $+z$ direction, $[\lambda] = T^{-1}L^3$. This is the assembly process.
- *Particle release* from the filament tip with rate τ and subsequent movement of the tip by distance h in the $-z$ direction, $[\tau] = T^{-1}$. This is the disassembly process.
- *Creation of a particle* with coefficient γ , $[\gamma] = T^{-1}L^{-3}$. This process is not part of the original self-assembly set-up, but it is included to maintain a constant bulk density of particles, see comment below.
- *Extinction of a particle* with rate r , $[r] = T^{-1}$. This process is used to enforce causality and to maintain the constant bulk density.
- *Extinction of the filament tip* with rate ϵ , $[\epsilon] = T^{-1}$, which is used to enforce causality.

The creation and extinction of particles is balanced such that a constant bulk density $\zeta = \gamma/r$ is created. The two extinction processes are included in the field theory to enforce causality. After calculations, we let parameters γ, r and ϵ tend to zero while keeping the ratio $\gamma/r = \zeta$ constant and positive. Thus, the spontaneous extinction and creation are removed while a bulk density remains included.

The processes above are encoded in the action functional $\mathcal{A}$ which splits up into a bilinear part and an interaction part $\mathcal{A} = \mathcal{A}_{\text{lin}} + \mathcal{A}_{\text{int}}$. The diffusion and extinction of

the building blocks is represented in the propagator part for the particles

$$\mathcal{A}_{\text{lin-P}} = \int_{\mathbb{R}^4} \tilde{\varphi}(x, t) (-\partial_t + D\Delta - r) \check{\varphi}(x, t) d^3x dt, \quad (3.5)$$

where Δ is the spatial Laplace operator in 3 dimensions.

The filament is stationary without the processes of incorporation or release of particles. It is described by the stationary propagator part for the filament tip

$$\mathcal{A}_{\text{lin-F-stat}} = \int_{\mathbb{R}} \sum_{j \in \mathbb{Z}^3} \tilde{\psi}_j(t) (-\partial_t - \epsilon) \psi_j(t) dt. \quad (3.6)$$

Due to particle incorporation and release, the propagator part of the filament tip action functional includes jumps along the set line of growth in steps of h . For convenience, I chose the z direction as assembly and disassembly direction.

$$\begin{aligned} \mathcal{A}_{\text{lin-F-mov}} = \int_{\mathbb{R}} \sum_{j \in \mathbb{Z}^3} & \left(\lambda \zeta (\tilde{\psi}_{j+1_z} - \tilde{\psi}_j) \psi_j \right. \\ & \left. + \tau (\tilde{\psi}_{j-1_z} - \tilde{\psi}_j) \psi_j \right) dt, \end{aligned} \quad (3.7)$$

where I omitted the time argument of the fields for better readability.

The first part, with prefactor $\lambda \zeta$ describes the filament assembly in the particle bulk, while the second part, with prefactor τ corresponds to the disassembly of the filament. The index $j + 1_z$ refers to the next lattice site in $+z$ direction, and the index $j - 1_z$ signifies the next lattice site in $-z$ direction.

All three bilinear actions together make up $\mathcal{A}_{\text{lin}}$

$$\mathcal{A}_{\text{lin}} = \mathcal{A}_{\text{lin-P}} + \mathcal{A}_{\text{lin-F-stat}} + \mathcal{A}_{\text{lin-F-mov}}. \quad (3.8)$$

The interaction part of $\mathcal{A}$ has the form

$$\begin{aligned} \mathcal{A}_{\text{int}} = \int_{\mathbb{R}} \sum_{j \in \mathbb{Z}^3} & \left[\lambda \left(\underbrace{(\tilde{\psi}_{j+1z} - \tilde{\psi}_j) \psi_j \check{\varphi}(hj)}_{(a)} - \underbrace{\tilde{\psi}_j \psi_j \check{\varphi}(hj) \check{\varphi}(hj)}_{(b)} - \underbrace{\psi_j \check{\varphi}(hj) \check{\varphi}(hj)}_{(c)} \right) \right. \\ & \left. + \underbrace{(\tau \tilde{\psi}_{j-1z} - \lambda \zeta \tilde{\psi}_j) \psi_j \check{\varphi}(hj)}_{(d)} + \underbrace{(\tau - \lambda \zeta) \check{\varphi}(hj) \psi_j}_{(e)} \right] dt, \end{aligned} \quad (3.9)$$

where the time argument is omitted for better readability.

The different parts of the interaction describe the following processes:

- (a) the filament assembles, i.e. the filament tip moves one step in the positive z direction upon incorporating a particle;
- (b) as a particle is incorporated into the tip, the particle density is reduced, resulting in anticorrelations of the tip and particle density;
- (c) the particle density is reduced by incorporation into the tip;
- (d) in the presence of a tip, the particle density is increased by spontaneous release (τ) and decreased by incorporation ($\lambda\zeta$), leading to corresponding correlations of tip and particle densities;
- (e) prefactor τ : the particle density is increased because the filament releases a particle; prefactor $\lambda\zeta$: the particle density is decreased because the filament incorporates a particle.

Both, propagation and interactions are schematically represented by Feynman diagrams. Particle propagation is drawn as a straight red line, filament propagation is depicted as a curly blue line, and interactions are illustrated by vertices, see Fig. 3.2.

The interplay between propagation and interaction in a system governed by the action $\mathcal{A}$ can be calculated using the path integral. The system may be initialised by placing a filament tip at position $hj_0 = 0$ at time $t_0 = 0$. Then, the system evolves and particle concentrations, the filament tip positions, or higher moments of their distributions can be measured at a later point in time as n -point correlation functions. In general, if the observable $\mathcal{O}(t)$ that we want to measure is represented by a combination of fields ,

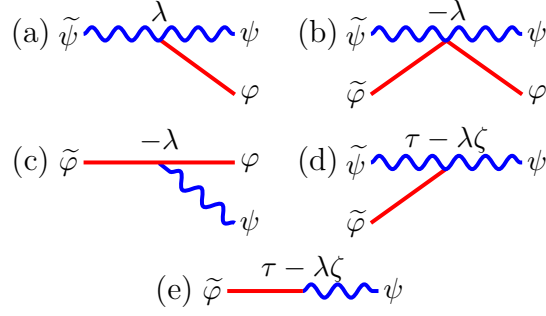


Figure 3.2: The stochastic processes that appear in the interaction part of the action $\mathcal{A}_{\text{int}}$ are represented as amputated vertices in Feynman diagrams. All Feynman diagrams should be read from right to left. Figure from [118].

then its time-dependent, spatial probability distribution is given by the following path integral (see e.g. [154] for a detailed review and Sec. 1.5)

$$\langle \mathcal{O}(t) \psi_0^\dagger(0) \rangle := \sum_{\ell=0}^{\infty} \int \mathcal{D}[\varphi, \psi] \mathcal{O}(t) \psi_0^\dagger(0) e^{-\mathcal{A}_{\text{prop}}} \frac{(-\mathcal{A}_{\text{int}})^\ell}{\ell!}, \quad (3.10)$$

where the system, which contains a bulk of particles, is initialized with a filament tip at the origin $j_0 = 0$ at time $t_0 = 0$.

Formally, this integral is summing all variations of all fields involved of all stochastic processes possibly occurring. The path integral is normalised such that

$$\langle 1 \rangle = \sum_{\ell=0}^{\infty} \int \mathcal{D}[\varphi, \psi] e^{-\mathcal{A}_{\text{prop}}} \frac{(-\mathcal{A}_{\text{int}})^\ell}{\ell!} = 1. \quad (3.11)$$

The ℓ -th term of the series is the contribution of all processes with ℓ interactions*. When I approximate the observable in Eq. (3.10) by truncating the series at ℓ , I use the notation $\langle \dots \rangle_\ell$. When I use a Dyson sum approximation, I use the notation $\langle \dots \rangle_{\text{Dy}}$. *A priori*, it is not clear which truncation of the path integral is a good approximation of the observable. However, a good agreement of the approximate result with experimental data indicates that the processes which were not included in the calculation rarely occur under experimental conditions.

*These interactions are interactions in the field-theoretic sense. In fact, the zeroth order term (i.e. $\ell = 0$) of $\langle \psi_j(t) \psi_0^\dagger(0) \rangle$ includes already an arbitrary number of chemical interactions.

In the following, $\langle \dots \rangle_0$ denotes contributions of order $\ell = 0$ to observables such as in Eq. (3.10). I will refer to the process devoid of any non-linearities as the zeroth order process. Finally, steady state solutions will be indicated by subscript s , while the remainder by subscript r for relaxation.

3.3 GROWTH SPEED AND VARIANCE

Once the system is initialised with a filament tip, interactions with bulk particles allow growing and shrinking of the filament. In the vicinity of the filament tip, particles are depleted or replenished. Asymptotically for large times, the expected averaged filament growth speed becomes constant and, in the moving frame of the tip, the shape of the particle depletion zone may attain a stationary profile. The characteristic time to reach this asymptotic steady state is the relaxation time of the system.

3.3.1 STEADY STATE FILAMENT GROWTH SPEED

Placing a filament tip at time $t_0 = 0$ at the origin $hj_0 = 0$, the system relaxes towards a steady state. The zeroth order process and its first correction are represented by the following Feynman diagrams:

$$\langle \psi_j(t) \tilde{\psi}_0(0) \rangle_2 \hat{=} \text{---} \text{wavy blue line} \text{---} \quad (3.12)$$

$$+ \text{---} \text{wavy blue line} \text{---} \text{red arc} \text{---} \text{wavy blue line} \text{---} \quad (3.13)$$

The zeroth order process is a mean field description of the filament tip moving in the particle bulk (top diagram). The first correction at the bottom takes into account the growth or the absence thereof either by particles being released and reincorporated (due to release with rate τ) or by particles being absorbed and thus being no longer available at a later point in time (with rate $\lambda\zeta$). Diagrams are to be read from right to left.

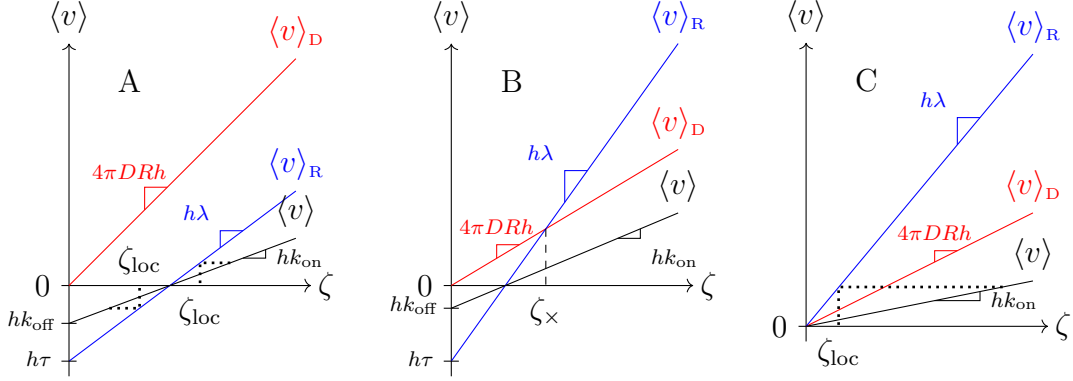


Figure 3.3: Three scenarios for filament growth speed. Red lines: diffusion-limited growth speed. Blue lines: reaction-limited growth speed. Black lines: effective growth speed. A: The effective reaction-limited growth speed is slower than the diffusion-limited growth speed at all bulk densities ζ . B: There is a cross-over bulk density $\zeta_\times$ above which diffusion-limited growth is slower than reaction-limited growth. C: Diffusion-limited growth is slower than reaction-limited growth at all bulk densities ζ . Figure from [118].

Hence, the averaged filament growth speed in the steady state regime is

$$\langle v \rangle_{2,s} = h(\lambda\zeta - \tau) \left(1 - \underbrace{\lambda \left(\frac{\Lambda}{2\pi^2 D} - \frac{h|\lambda\zeta - \tau|}{8\pi D^2} \right)}_{\text{Loop correction}} \right), \quad (3.14)$$

$\langle v \rangle_{2,s}$ is not the instantaneous speed but the stationary speed defined as the expected growth length at time t divided by t in the limit $t \rightarrow \infty$. The derivation of $\langle v \rangle_{2,s}$ outlined in the appendix, Sec. C.3. The loop correction term accounts for the local depletion of particles at the filament tip due to the polymerisation of the filament.

Such interactions result in a non-linear dependence of the average growth speed on the underlying parameters, in particular the particle bulk density ζ . Furthermore, a cutoff Λ appears whose inverse is linearly related to the reaction radius R of the filament tip.

Without the loop correction, this results reflects the chemical picture of rate equations. It corresponds to effective reaction limited growth. However, the loop correction term takes into account that, due to diffusive transport, the effective reaction-limited speed is reduced because the locally depleted reactants have to reach the reaction radius. Considering the first term of the loop correction, if the diffusion is strong, or the reaction radius large, the chemical reactions are less hindered by slow transport and the growth speed is closer to its reaction-limit. This diffusion correction is itself corrected in the

second term of the loop correction, which describes how quickly the tip reaches regions that are not depleted. If the filament were stationary, i.e. $\mathcal{A}_{\text{lin-F-mov}} = 0$, then the loop correction would only consist of its first term.

If the loop correction term has a similar magnitude compared to 1, then further loop corrections have to be taken into account and the growth process is further away from reaction-limited behaviour and closer to diffusion-limited dynamics.

In order to capture corrections to any order in λ and τ , I calculate the steady state Dyson sum approximation, which is represented by Daisy-chain-like Feynman diagrams:

$$\langle v \rangle_{\text{Dy},s} \hat{=} \text{wavy line} \quad (3.15)$$

(3.16)

$$\text{Diagram 1} + \dots \quad (3.17)$$

$$= \frac{h(\lambda\zeta - \tau)}{1 + \lambda \left(\frac{1}{4\pi DR} - \frac{h|\lambda\zeta - \tau|}{8\pi D^2} \right)}. \quad (3.18)$$

The above result involves the Dyson sum of the the position of the tip $\langle \psi_j(t) \tilde{\psi}_0(0) \rangle_{\text{Dy}}$ divided by time t . It is non-linear in the bulk density ζ , which has not been observed so far. Therefore, I assume that $\hbar|\lambda\zeta - \tau|/(8\pi D^2)$ is small compared to the other terms in the denominator. I approximate the the speed as

$$\langle v \rangle_{\text{Dy},s} \approx \frac{h(\lambda\zeta - \tau)}{1 + \frac{\lambda}{4\pi DR}}. \quad (3.19)$$

If the above speed approximation is equal to the measured speed, Eq. (3.1), the coefficients λ and τ can be determined from the measured coefficients k_{on} and k_{off}

$$\lambda = k_{\text{on}} \frac{4\pi DR}{4\pi DR - k_{\text{on}}} \quad \text{and} \quad \tau = k_{\text{off}} \frac{4\pi DR}{4\pi DR - k_{\text{on}}}. \quad (3.20)$$

In particular, it implies that $k_{\text{on}}/\lambda < 1$ and $k_{\text{on}}/(4\pi DR) < 1$, as well as $k_{\text{off}}/\tau < 1$, see Fig. 3.3 for comparison.

The crossover bulk density $\zeta_\times$ in panel B of Fig. 3.3 is defined as

$$\zeta_\times = \frac{k_{\text{off}}}{2k_{\text{on}} - 4\pi DR}, \quad (3.21)$$

above which the diffusion process becomes more limiting to the effective growth speed than the reactions.

3.3.2 VARIANCE OF THE FILAMENT LENGTH

In our model, the variation $\text{Var}_0(\ell)$ of the average steady state filament length can be estimated for times larger than the relaxation time and in the mean-field approximation, to be

$$D_{\text{eff},0} = h^2(\lambda\zeta + \tau). \quad (3.22)$$

This is not the variance of the instantaneous length but the variance of the average steady state length taken after time t . Hence it grows linear in t . Its derivation is outlined in the appendix, Sec. C.4.

Its first loop correction is

$$D_{\text{eff},2} = h^2(\lambda\zeta + \tau) \left(1 - \frac{\lambda}{4\pi DR} \right) + \lambda \frac{h|\lambda\zeta - \tau|}{2D\pi} \quad (3.23)$$

$$\approx h^2(k_{\text{on}}\zeta + k_{\text{off}}) + \lambda \frac{h|\lambda\zeta - \tau|}{2D\pi}. \quad (3.24)$$

3.3.3 EXPERIMENTAL EVIDENCE

In experiments with microtubules and actin filaments, many factors play a rôle which are not part of our model. For example, if the experiment is *in vivo*, other proteins such as microtubule associated proteins [68, 53, 100] accelerate or slow down the self-assembly or even break up the entire structure. There are different types of tubulin dimers [54] and actin monomers [50]. Also important is the question whether the filament is free floating in a solution or if one end is attached. In some experimental settings, the filament is pushing against a load, simulating the cell wall [33].

Table 3.1: Experimental data extracted from three references [50, 87, 54]: effective incorporation rate k_{on} , effective release rate k_{off} , bulk density of actin / tubulin ζ , and measured effective diffusion $D_{\text{eff-m}}$. The second column specifies specific variants of the used actin monomers or tubulin dimers. The data marked by * is taken from Fig. 6C in [54] and is unusually low. The typical values are marked by † and are taken from table S1 in [54]. The second-to-last column is calculated from column k_{on} and k_{off} , following Eq. (3.3). Table from [118].

Reference		k_{on} $\mu\text{M}^{-1}\text{s}^{-1}$	k_{off} s^{-1}	ζ μM	$D_{\text{eff-m}}$ nm^2s^{-1}	$D_{\text{eff-R}}$ nm^2s^{-1}	R nm
Fujiwara et al. [50]	<i>Ca</i>	6.1	0.85	0.14	366	12.4	0.02
	<i>Mg</i>	10	0.64	0.06	421	9.3	0.03
Kuhn, Pollard [87]	<i>Mg</i>	7.4	0.8	0.16	226	14.5	0.02
Gardner et al. [54]	GMPCPP	5.1	3.9	1.5	28.8	4.2	0.13
	GTP	1.5*	0.5*	7.0	111	4	0.04
	GTP	4.8 †	15 †	7.0	111	17.5	0.13

Despite the numerous special circumstances of every experiment, the comparison of the model with data can be achieved. The steps of the comparison are

1. Extract for each experiment the measured values of the effective incorporation rate k_{on} , effective release rate k_{off} and effective diffusion rate $D_{\text{eff-m}}$ at a specific bulk density ζ .
2. Compare the measured effective diffusion constant $D_{\text{eff-m}}$ with the one from Eq. (3.3), where the reaction-limit is assumed, i.e. $k_{\text{on}} = \lambda$ and $k_{\text{off}} = \tau$. The result is shown in the second-to-last column in Table 3.1. The result of this comparison is that the reaction-limited dynamics do not explain the measured data well.
3. Use Eq. (3.20) and Eq. (3.24) to match k_{on} , k_{off} and $D_{\text{eff-m}}$ by choosing a suitable effective reaction radius R , shown in last column in Table 3.1. Are the resulting effective reaction radii plausible?

For the calculations, a diffusion constant of $D = 5 \cdot 10^7 \text{ nm}^2\text{s}^{-1}$ is chosen for actin in water [101, 90, 152] and $D = 5.9 \cdot 10^7 \text{ nm}^2\text{s}^{-1}$ for tubulin in sea urchin cytoplasm [142]. As effective growth step size h , I use $h = 0.6 \text{ nm}$ for microtubules and $h = 2.7 \text{ nm}$ for actin filaments, as has been done in [50].

In Table 3.1, the measured effective diffusion constant $D_{\text{eff-m}}$ is significantly greater than the expected one on the basis of reaction-limited dynamics, $D_{\text{eff-R}}$. A proposed

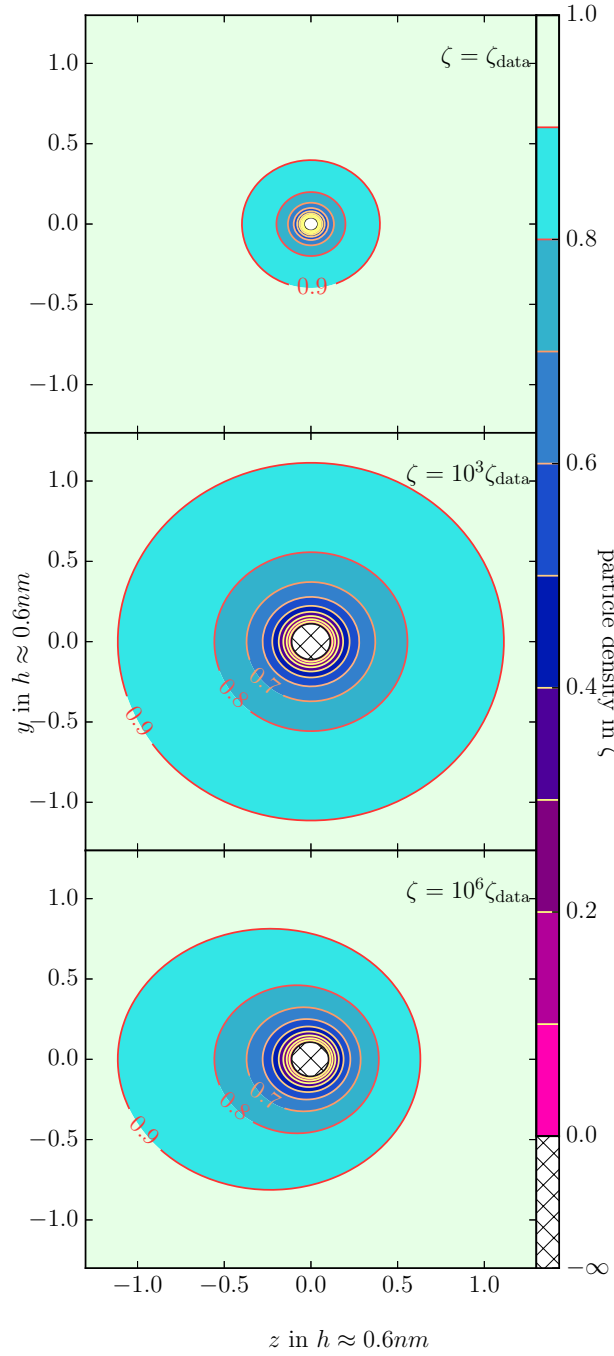


Figure 3.4: Density of particles in the moving frame in the section $\tilde{x} = 0$. The particle density is depicted compared to the bulk density ζ for three different bulk densities ζ . All other parameters are taken from Table 3.2, with $\lambda = k_{on}$ and $\tau = k_{off}$. The depletion zones change from a spherically symmetric shape to a asymmetric oval shape. However, the oval shape only appears for unrealistically large densities.

Table 3.2: Parameters and their typical values for microtubule self-assembly. These are not taken from any specific experiments but are approximate values of the data found in [54]. The effective tubulin dimer size takes into account that microtubules consists of 13 protofilaments, while we consider only a single polymer. These parameter values are used for the plots in Figs. 3.4, 3.5, 3.6 and 3.7. For actin filaments, the effective release rate k_{off} is about a factor 5 to 10 smaller, and typical densities are about a factor 10 to 15 smaller, see Table 3.1 for comparison.

parameter	value
diffusion constant D	$10\mu m^2 s^{-1}$
tubulin dimer density ζ_{data}	$5\mu M$
on-rate k_{on}	$5\mu M^{-1} s^{-1}$
off-rate k_{off}	$15 s^{-1}$
size of tubulin dimer h_{real}	$8nm$
effective tubulin dimer size h	$\frac{8nm}{13}$

Only the zeroth order stochastic process is represented in Eq. (3.25). It is a mean field description of the filament tip locally depleting ($-\lambda\zeta$) or fueling (τ) the particle bulk. Diagrams have to be read from right to left.

The particle density and its depletion can be calculated as the following observable

$$\langle \varphi(x, t) \psi_0^\dagger(0) \rangle_{0,s} = \zeta + \frac{\tau - \lambda\zeta}{4\pi D |\tilde{x}|} e^{-\frac{h(\lambda\zeta - \tau)}{2D} (|\tilde{x}| + \tilde{x}_z)}, \quad (3.26)$$

where the coordinate of the moving frame is $\tilde{x}$ with $\tilde{x}_x = x_x$, $\tilde{x}_y = x_y$ and $\tilde{x}_z = x_z + h(\lambda\zeta - \tau)t$. This result recovers the transport picture as it is described by *Odde* (1997) [108], however it also directly relates its constituent elements to the chemical picture.

Fig. 3.4 depicts the particle depletion of example parameters (Table 3.2) set alongside two more examples of significantly higher particle densities. Clearly, oval depletion zones shapes are reached only as the particle density is of the order of a million times higher than in typical experimental settings.

The expected number of particles n_V in a volume $V \subset \mathbb{R}^3$ is hence

$$\langle n_V \rangle_{0,s} = \int_V \langle \varphi(\tilde{x}, t) \psi_0^\dagger(0) \rangle_{0,s} d\tilde{x}. \quad (3.27)$$

Far away from the filament tip, the number of particles in the volume V is $|V|\zeta$, where $|V|$ is the volume size of V . If $\lambda\zeta - \tau < 0$, then the particle number can also be calculated for arbitrary volumes that include the filament tip. However, if $\lambda\zeta - \tau > 0$, the solution (3.26) predicts a negative particle density and therefore unphysical results. If V is a volume in the shape of a ball centered at the filament tip, we can numerically calculate its minimal radius ρ_0 such that the particle number is non-negative, using Eq. (3.27). For $h^2|\lambda\zeta - \tau|/D \ll 1$, we can analytically approximate the minimal radius ρ_0 to

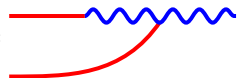
$$\rho_0 \approx \frac{3|\lambda\zeta - \tau|}{8\pi\zeta D} \quad (3.28)$$

For the example particle density ζ_{data} , the minimal radius equals approximately 0.01nm. For smaller volumes within this minimal radius, the approximation breaks down and we interpret the expected particle number as being equal to zero.

A more detailed derivation of these results can be found in the appendix, Sec. C.5.

3.4.1 VARIANCE OF STEADY STATE PARTICLE DEPLETION

Although the expected particle number drops to zero in the vicinity of the filament tip, there are significant fluctuations of the particle density in the steady state. In order to estimate these fluctuations, the variance of the number of particles is calculated in the following. First, the spatial correlations of the particle density are calculated:

$$\langle \varphi(x_1, t) \varphi(x_2, t) \tilde{\psi}_0(0) \rangle_0 \hat{=} \text{Diagram} \quad (3.29)$$


After initialisation of a filament tip (curly blue line), the particle density (straight red line) is measured twice at the same time but possibly at different positions. Only the zeroth order stochastic processes is represented in form of Feynman diagrams. The zeroth order process is a mean field description of the filament tip locally depleting ($-\lambda\zeta$) or fueling (τ) the particle concentration. Diagrams are to be read from right to left.

Next, the covariance of the particle density in a system with a filament tip is calculated to zeroth order

$$\begin{aligned}
\text{cov}_{0,s}(\varphi(\tilde{x}, t)\varphi(\tilde{y}, t)|\psi_0^\dagger(0)) &= \underbrace{\frac{(\tau - \lambda\zeta)^2 \left(e^{-\frac{h|\lambda\zeta - \tau|}{2D}(|\tilde{x}| + x_z)} + e^{-\frac{h|\lambda\zeta - \tau|}{2D}(|\tilde{y}| + y_z)} \right)}{2(4\pi D|\tilde{x}|)(4\pi D|\tilde{y}|)}}_{\text{Reaction-diffusion part} =: \text{cov}_{0,s,\text{RD}}(\dots)} \\
&\quad + \underbrace{\delta(\tilde{x} - \tilde{y}) \left(\zeta + \frac{(\tau - \lambda\zeta) e^{-\frac{h|\lambda\zeta - \tau|}{2D}(|\tilde{x}| + \tilde{x}_z)}}{4\pi D|\tilde{x}|} \right)}_{\text{Poissonian part} =: \text{cov}_{0,s,\text{Poi}}(\dots)},
\end{aligned} \tag{3.30}$$

where only the the stochastic process represented by the Feynman diagram shown in Eq. 3.29 was taken into account.

The covariance of the particle density splits into two parts. The first one is due to the dynamics of the reaction-diffusion processes in the system, while the second is due to the Poissonian spatial distribution of the particles. The covariance of the number of particles n_{V_1} and n_{V_2} in volumes V_1 and V_2 is given by the integral

$$\text{cov}_{0,s}(n_{V_1}, n_{V_2}) = \int_{V_1 \times V_2} \text{cov}_{0,s}(\varphi(\tilde{x}, t)\varphi(\tilde{y}, t)|\psi_0^\dagger(0)) d\tilde{x} d\tilde{y}, \tag{3.31}$$

where the integrand is stated in Eq. (3.30). This is a conditioned covariance. Its condition is the initialization of the system with a filament tip at position $j_0 = 0$ at time $t_0 = 0$. The Poissonian part plays a rôle only if the volumes V_1 and V_2 have a non-empty intersection, as distinct Poissonian-distributed particles are independent.

In order to calculate the variance $\text{Var}_0(n_V)$ of the number of particles n_V in a volume V , we set volumes V_1 and V_2 equal to V .

$$\text{Var}_0(n_V) = \text{Var}_{0,\text{RD}}(n_V) + \text{Var}_{0,\text{Poi}}(n_V), \tag{3.32}$$

$$\text{Var}_{0,\text{RD}}(n_V) = \int_V \text{cov}_{0,s,\text{RD}}(\varphi(\tilde{x}, t)\varphi(\tilde{x}, t)|\psi_0^\dagger(0)) d\tilde{x}, \tag{3.33}$$

$$\text{Var}_{0,\text{Poi}}(n_V) = \int_V \text{cov}_{0,s,\text{Poi}}(\varphi(\tilde{x}, t)\varphi(\tilde{x}, t)|\psi_0^\dagger(0)) d\tilde{x}. \tag{3.34}$$

In that case, there are two regions which are dominated by different fluctuations. Far away from the depletion zone, the variance is dominated by the Poissonian part

$\text{Var}_{0,\text{Poi}}(n_V)$, while the reaction-diffusion part $\text{Var}_{0,\text{RD}}(n_V)$ vanishes. If V is an open finite ball around $\tilde{x}$, we find:

$$\lim_{|\tilde{x}| \rightarrow \infty} \text{Var}_{0,\text{Poi}}(n_V) = |V|\zeta = \lim_{|\tilde{x}| \rightarrow \infty} \langle n_V \rangle_0 \quad (3.35)$$

$$\lim_{|\tilde{x}| \rightarrow \infty} \text{Var}_{0,\text{RD}}(n_V) = 0 \quad (3.36)$$

In this limit, the expected number of particles and its variance are equal as expected for purely Poissonian fluctuations.

However, the variance of the particle density in the neighbourhood of the filament tip is also important because large fluctuations around the tip can lead to large fluctuations of the growth speed. We calculate the variance for the ball of minimal radius ρ_0 , Eq. (3.28), centered at the filament tip in the approximation $h^2|\lambda\zeta - \tau|/D \ll 1$ and find

$$\text{Var}_0(n_{V_{\rho_0}}) \approx \frac{|\lambda\zeta - \tau|^2}{4D^2} \rho_0^4. \quad (3.37)$$

As the expected particle number in V_{ρ_0} is zero, this result indicates the non-Poissonian character of the fluctuations.

Comparing the standard deviation $\sigma_0(n_{V_{\rho_0}})$ with the expected particle number at the bulk $\zeta|V_{\rho_0}|$, we find

$$\frac{\sigma_0(n_{V_{\rho_0}})}{\zeta|V_{\rho_0}|} \approx 1. \quad (3.38)$$

Hence, we conclude that even in the particle depleted-volume in the neighbourhood of the filament tip, the fluctuations of the particle density are significant. In particular, even if the depletion zone of the filament tip increases with particle density and thus growth speed, particle density fluctuations will allow the filament tip to grow faster for a short period of time than the mean-field limit, such as presented in [108].

Details of the calculation can be found in the appendix, Sec. C.6.

3.5 RELAXATION TOWARDS STEADY STATE

Many models [65, 103, 24, 26, 164, 5, 108] assume that, given a fixed particle density, filaments are growing or shrinking in a steady state fashion. This means that filament growth speed, shrinking speed, and particle depletion in the moving frame of the filament tip are time-independent. However, putting a filament seed into a bath of particles, mechanically changing the filament position, or changing the particle density changes the steady state of the system. I call the convergence of the system towards its steady state *relaxation*. I want to estimate the typical time scales of such relaxations.

3.5.1 RELAXATION OF THE FILAMENT GROWTH SPEED

If I initialize the system with one filament tip at the origin $j_0 = 0$ at time $t_0 = 0$, and calculate the expected filament tip position, we find that the calculation splits naturally into two parts. One part is linear in time and determines the steady state growth, the other part is time-dependent and tends towards zero for large times. This second part is the relaxation I want to characterise in the following. It is given by

$$\begin{aligned} \langle v \rangle_{2,r} &:= \langle v \rangle_2 - \langle v \rangle_{2,s} \\ &= \frac{h}{t} \left(\lambda(\tau - \lambda\zeta) \left(\frac{-\sqrt{t} e^{-\frac{h^2(\lambda\zeta - \tau)^2 t}{4D}}}{4(\pi D)^{3/2}} + \right. \right. \\ &\quad \left. \left. + \frac{\left(1 + \frac{h^2(\lambda\zeta - \tau)^2 t}{2D}\right) \operatorname{erfc}\left(\sqrt{\frac{h^2(\lambda\zeta - \tau)^2 t}{4D}}\right) - 1}{4\pi D h |\lambda\zeta - \tau|} \right) \right) \quad (3.39) \end{aligned}$$

for large times $t \sim \begin{cases} \frac{\lambda}{4\pi D t} \operatorname{sign}(\lambda\zeta - \tau) & \text{Relaxation 1} \\ \frac{\lambda(\lambda\zeta - \tau)h}{4(\pi D)^{3/2}\sqrt{t}} & \text{Relaxation 2.} \end{cases}$

The relaxation of the filament growth speed occurs only in interaction with the relaxing particle bulk. The feedback from the particle bulk is taken into account by loop corrections. Therefore, the relaxation behaviour appears only beyond the mean-field approximation.

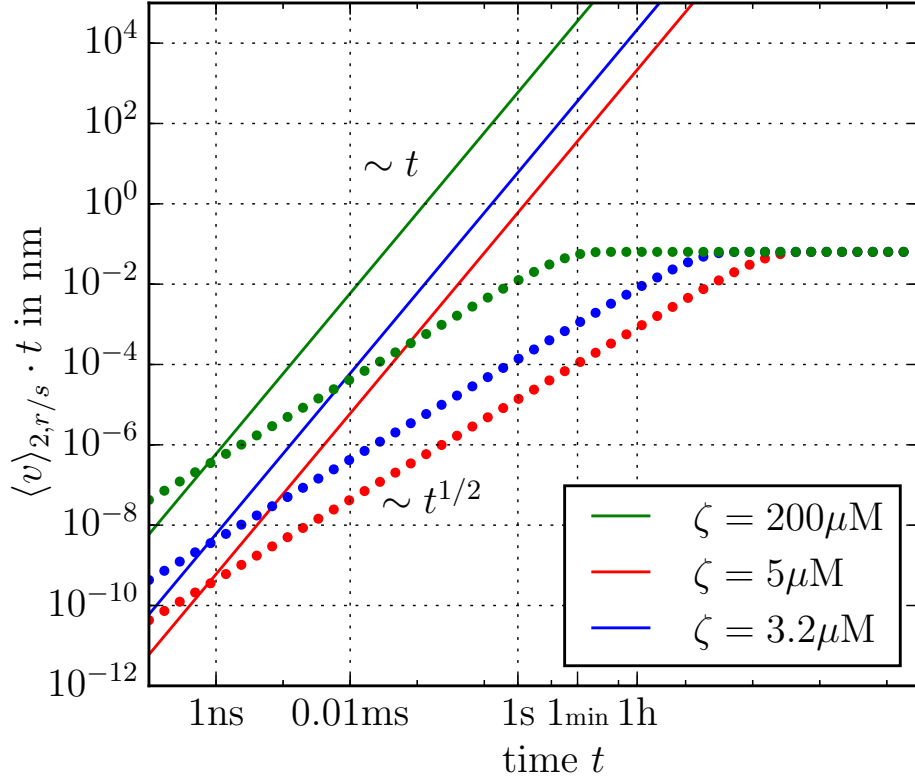


Figure 3.5: Steady state (line) and relaxation (dotted) parts of the growth length are shown for three different example densities ζ . All other parameters are taken from Table 3.2 with $\lambda = k_{\text{on}}$ and $\tau = k_{\text{off}}$.

The relevant asymptotic behaviour of the relaxation depends on the parameters and is given by Eq. (3.39). Although ultimately, the Relaxation 1 term will dominate, we have to estimate at what point in time this term becomes dominant and how it compares at that point to the amplitude of the steady state solution.

In Fig. 3.5 the filament growth length is shown as a function of time t . Independent of the particle density, the relaxation part drops to 1% of the steady state part within approximately $10\mu\text{s}$. Hence, we conclude that the relaxation is not important for measuring the filament growth speed.

A detailed derivation of this result can be found in the appendix, Sec. C.7.

In experiments with filaments, it does not seem feasible to measure a relaxation time of the growth speed of $10\mu\text{s}$. Hence, this result would not be verifiable. However, it

also hints at a drawback of the used model. The model uses a microtubule tip of size zero, i.e. the tip does not have a diameter. Hence, the depletion area around the tip is unrealistically small and only takes a short time to reach the steady state. In reality, a microtubule consists of 13 protofilaments which are arranged in a cylinder of about 24nm diameter. Because of the size of the tip, the depletion area around the tip would also be larger, its relaxation to the steady state would take longer, and therefore the relaxation of the growth speed would also take longer. A better model will have to include more protofilaments within one microtubule or a different implementation of the tip size in order to accurately predict the relaxation of the growth speed.

3.5.2 RELAXATION OF THE PARTICLE DENSITY

Our model is set up such that the system is initially filled with a uniform particle density ζ . In order to initialize dynamics in the system, we can put a filament tip into the bulk and calculate the relaxation of the particle bulk around the filament tip towards a steady state distribution. The relaxation part of the particle density is given by

$$\begin{aligned}
\langle \varphi(\tilde{x}, t) \psi_0^\dagger(0) \rangle_{0,r} &= \langle \varphi(\tilde{x}, t) \psi_0^\dagger(0) \rangle_0 - \langle \varphi(\tilde{x}, t) \psi_0^\dagger(0) \rangle_{0,s} \\
&= \exp\left(-\frac{h(\lambda\zeta - \tau)\tilde{x}_z}{(2D)}\right) \int_t^\infty \frac{(\lambda\zeta - \tau) e^{-\frac{\tilde{x}^2}{4Dt'} - \frac{h^2(\lambda\zeta - \tau)^2}{4D}t'}}{(4\pi Dt')^{\frac{3}{2}}} dt' \\
&\sim \begin{cases} \frac{1}{4\pi Dht} \text{sign}(\tau - \lambda\zeta) & \text{Relaxation 1} \\ \frac{\tau - \lambda\zeta}{(4\pi D)^{3/2}\sqrt{t}} & \text{Relaxation 2.} \end{cases}
\end{aligned} \tag{3.40}$$

The relaxation of the particle density already occurs at mean-field level because the initialised filament tip directly impacts the particle bulk. A more detailed derivation of this result can be found in the appendix, Sec. C.8.

3.6 SPEED CORRELATIONS

As the filament grows, it interacts with the particle bulk and the tip will move through regions of higher and lower particle density. Such inhomogeneities are caused by fluctuations due to the diffusion of Poisson distributed particles and as a feedback to the incorporation and release of particles (see Sec. 3.4.1). These interactions lead to temporal correlations of the filament growth speed. As a measure for these correlations we define the speed correlation coefficient C which depends on the time difference Δt at which the speed of the filament growth is measured:

$$C(t, \Delta t) = \frac{\langle v(t + \Delta t)v(t) \rangle_0}{\langle v(\Delta t) \rangle_0 \langle v(t) \rangle_0} - 1. \quad (3.41)$$

This correlation coefficient drops to zero when the growth speed $v(t + \Delta t)$ at time $t + \Delta t$ is uncorrelated to the speed $v(t)$ at time t .

Part of this correlation coefficient is the temporal correlation function $\langle v(t + \Delta t)v(t) \rangle_0$ which requires to measure the position of the filament tip twice:

$$\langle \psi_{j_2}(t + \Delta t) \tilde{\psi}_{j_1}(t) \psi_{j_1}(t) \tilde{\psi}_0(0) \rangle_0 \hat{=} \text{diagram} \quad (3.42)$$


After initialisation of a filament tip (curly blue line), the particle (straight red line) interacts with the filament tip. The same filament tip is measured twice, but at different times. The interaction leads to local particle depletion ($-\lambda\zeta$) or fueling (τ) by the filament tip. Diagrams are to be read from right to left.

In the large time limit, when the growth is in a steady state, the coefficient becomes independent of time t . This limit, denoted by $C_s(\Delta t) = \lim_{t \rightarrow \infty} C(t, \Delta t)$ is

$$C_s(\Delta t) = \lambda \left(\frac{e^{-\frac{h^2(\lambda\zeta - \tau)^2}{4D}\Delta t}}{4(\pi D)^{3/2}\sqrt{\Delta t}} - \frac{h|\lambda\zeta - \tau|}{8\pi D^2} + \right. \\ \left. + \frac{(\frac{1}{\Delta t} + \frac{h^2(\lambda\zeta - \tau)^2}{2D})\text{erf}\left(\sqrt{\frac{h^2(\lambda\zeta - \tau)^2\Delta t}{4D}}\right)}{4\pi D h |\lambda\zeta - \tau|} \right), \quad (3.43)$$

where the stochastic process shown as the Feynman diagram in Fig. 3.42 had to be

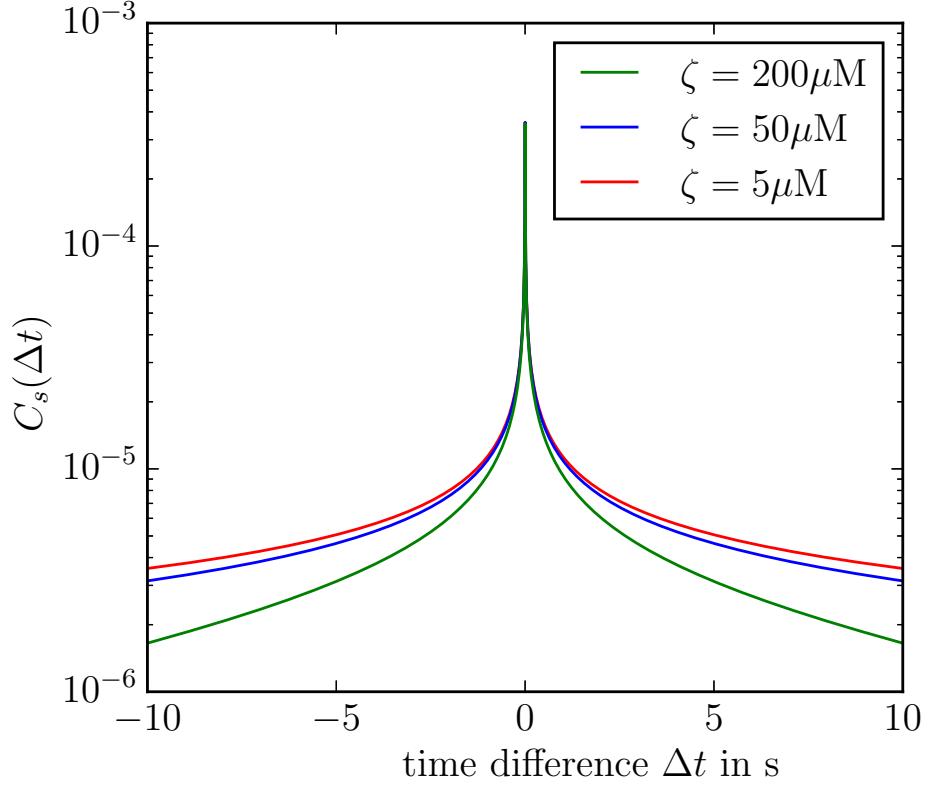


Figure 3.6: The correlation function $C_s(\Delta t)$ is depicted for three different particle densities. All other parameter values were taken from Table 3.2.

taken into account.

For $\Delta t \rightarrow 0$ the coefficient diverges to infinity because the average speed goes linearly to zero while the growth due to interactions with the particle bulk only vanishes with the square root of Δt . Hence, it diverges as $\Delta t^{-1/2}$. As expected, for long time differences Δt the growth speeds become independent and the correlation coefficient decays to zero. This decay is divided into two regimes. For $\Delta t < 4D/(h^2(\lambda\zeta - \tau)^2)$, the decay follows $\Delta t^{-1/2}$. For larger $\Delta t > 4D/(h^2(\lambda\zeta - \tau)^2)$, the decay is exponential.

In experiments, the time and space resolution are limited, which can be captured by a convolution with a time window. As $C_s(\Delta t)$ is locally integrable, the resulting function is a smooth function without a divergence at $\Delta t = 0$. It is bell-shaped decaying $\Delta t^{-1/2}$ in the first regime, and an exponential decay in the second regime. The stationary

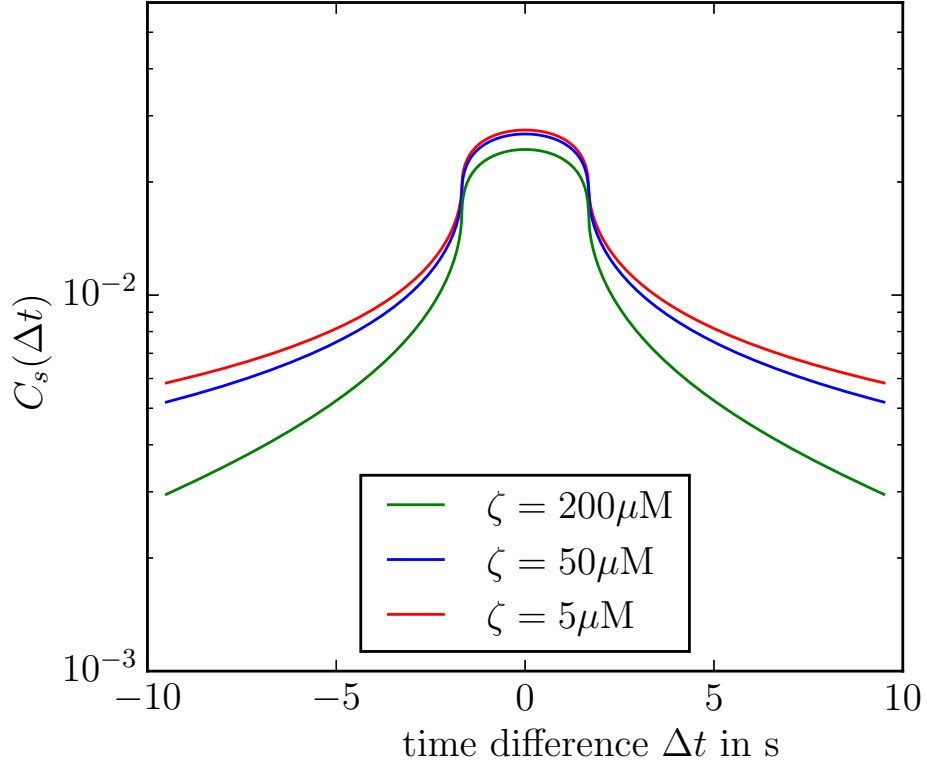


Figure 3.7: A convolution of the correlation function $C_s(\Delta t)$ with a time window of $1.5s$ is depicted for three different particle densities ζ . All other parameter values were taken from Table 3.2. The convolution imitates the effect of a low temporal resolution as would be expected in experiments.

correlation coefficient $C_s(\Delta t)$ and a convolution with a time window of a characteristic time are depicted in Fig. 3.6 and Fig. 3.7.

Details of the derivation can be found in the appendix, Sec. C.9.

3.7 CONCLUSION

A field-theoretic model for the self-assembly of actin filaments and microtubules in a diffusive environment was derived and analysed. The focus of the approach was to capture beyond-mean field behaviour of the reaction-diffusion system.

While the reaction-limited mean-field behaviour, Eq. (3.2) and Eq. (3.3), and the diffusion-limited mean-field behaviour, Eq. (3.4), are unable to capture important aspects of the

self-assembly dynamics, in particular its fluctuations, our model is able to provide an explanation for their emergence out of the superposition of the two stochastic processes, see Sec. 3.3.3.

Capturing analytically the stochasticity of reaction processes whose supply of reactants is undermined by diffusion comes at the prize of other simplifications. They include the hydrolysis of the tip's GTP tubulin to GDP tubulin for microtubules [52] and the ATP actin hydrolyzing to ADP actin in the tip of actin filaments [87]. Furthermore, the geometric structure was simplified too: the 13 microtubule protofilaments and the 2 strands of actin filaments were simplified to a single polymer line. Nevertheless, the quantitative comparison in Table 3.1 shows reasonable results.

In order to verify the approach, a complementary experimental comparison of growth speed correlations was suggested in Sec. 3.6.

3.7.1 OUTLOOK: HYDROLYSIS OF THE CAP – DYNAMIC INSTABILITY OF MICRO-TUBULES

In microtubules and in actin filaments, the GTP / ATP of tubulin / actin hydrolyze to GDP / ADP [123]. While actin filaments remain stable [50], for microtubules, this is assumed to be the main cause of an instability due to which large parts of the microtubule fall apart [164]. During the unstable phase, microtubules exhibit great release rates that are typically much larger than the assembly rates during the stable phase [109]. However, many questions about its stochastic nature remain open: what is the first-passage time for a microtubule to switch to its unstable state, in which it rapidly disassembles and what is the mean disassembly distance before it is rescued, i.e. before it switches back to its stable state?

Despite its common observation, the statistics of the dynamic instability are poorly understood and rarely precisely modelled. Significant simulation results were obtained in [161], an important analytic attempt was made in [175] using *Doi's* approach of modelling the master equation in a second quantized language [37].

Future research will include an extension of the Doi-Peliti field theory model, which allows for the hydrolysis dynamics in the microtubule cap by introducing a second kind of field for the tip, one represents the stable tip, the other one the unstable tip.

3.7.2 OUTLOOK: STOP-AND-GO DYNAMICS OF TRANSPORT ALONG MICROTUBULES

Microtubules and actin filaments form a network in cells which is also used for transport [61]. Motorproteins move along the filaments and transport organelles and other large-scale components within the cell [68]. Typically, these motorproteins have a preferred direction of travel, rarely change the protofilament they are walking along, and can get stuck in a traffic-jam-like condensation process. The process resembles an asymmetric exclusion process [147, 2, 28, 73] on 13 lanes with different particles: those which can go in only one direction (2 types) and those which can go in both (1 type). The study of this asymmetric exclusion process would be of interest both with regards to non-equilibrium statistical mechanics as well as cell biology, and is left for future research.

In the meantime the mathematicians, guided by that instinct which teaches them to store up for others the irrepressible secretions of their own minds, had developed with the utmost generality the dynamical theory of material systems.

James Clerk-Maxwell [27]

4

WETTING ON STRUCTURED SURFACES

ABSTRACT

In this chapter, a model of equilibrium wetting phenomena on structured surfaces is presented. After introducing the surface tension and the contact angle of liquids on solid surfaces, Sec 4.1, I present several phase transitions of liquid layers on flat surfaces, wedges and apices, Secs. 4.3-4.6. The original work related to my research is presented in Sec. 4.5 and Sec. 4.6. The project was in collaboration with A.O. Parry and C. Rascón and the results, including Figs. 4.6 and 4.9 to 4.16 were published in *Soft Matter* in 2018 [133]. I contributed to all mean-field calculations. The article [133] also involved calculations of the interfacial fluctuation effects, which were performed solely by my collaborators and are therefore not presented here. At the end of the chapter, I present future research goals in Sec. 4.7.1.

4.1 INTRODUCTION

Wetting phenomena surround us in our everyday life. They become visible in condensation and drying, and can be felt as humidity. That is probably why theoretic research on this topic goes back more than 200 years to *Thomas Young* and *Pierre-Simon LaPlace* [173, 91]. Wetting phenomena still play an important rôle in science today and are intensively experimentally studied [55, 158], in particular in connection with growing crystals from fluids [6, 13]. The theoretical work in this area also remains very active, and is continuously extended in new directions [122, 82, 74].

At the outset of the study of wetting is the phenomenon that liquids form droplets on solid surfaces and rise in capillaries against gravity, which cannot be explained by hydraulics. Already at the time of *Isaac Newton*, it was ruled out by *Francis Hauksbee* that gravitational attraction between the capillary walls and the liquid are responsible for the phenomenon by observing its independence on the thickness of the walls [64].

The important observable of wetting was the contact angle θ , i.e. the angle which is formed between the liquid-gas boundary and the liquid-solid boundary, shown in Fig. 4.1. It varies for different materials and with temperature [173, 139]. For example, the contact angle of mercury on teflon increases between temperatures of 25°C and 150°C from $\theta = 157^\circ$ to $\theta = 178^\circ$ [41]. At about 20°C , water has a contact angle of 9° on biotite but a contact angle of 40° on calcite [43].

The contact angle and the surface tension are closely related. For the derivation of this relation, I follow the book by *Rowlinson* and *Widom* [139].

Let us assume there are two volume elements dV_1 and dV_2 of possibly different materials with particle number densities ρ_1 and ρ_2 . Between the molecules of the two materials acts an attractive force $F(r)$ with finite and short range d_{12} . If the molecules are closer than distance $r < d_{12}$, the force is attractive. Beyond this distance, the force is zero. Hence, the attractive force between the volume elements is

$$F_{dV_1, dV_2} = F(r)\rho_1 dV_1 \rho_2 dV_2, \quad (4.1)$$

where r is the distance between dV_1 and dV_2 .

The volume elements dV_1 and dV_2 are part of bigger volumes V_1 and V_2 whose attrac-

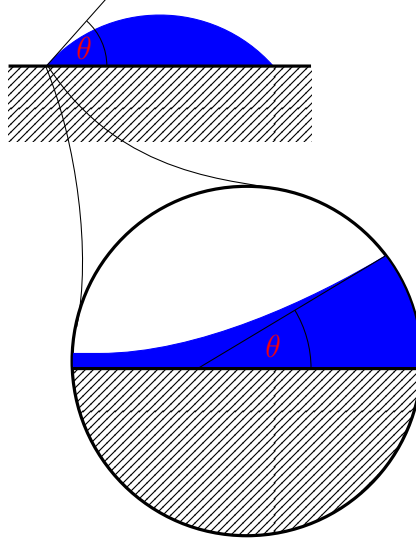


Figure 4.1: Macroscopic picture (top) and mesoscopic picture (bottom) of a liquid droplet on a solid surface, surrounded by gas. The contact angle θ is formed between the macroscopic liquid-gas and solid-liquid boundaries. In the mesoscopic picture, a liquid layer coats the entire solid surface.

tive force to each other can be calculated by integrating F_{dV_1, dV_2} over their volumes. However, the force has a finite and short range. For large volumes in contact, only their contact surface will effectively enter into the overall attractive force. Hence, an attractive force per area is a more useful object.

Therefore, the volume element dV_1 is split into its area element dA_1 along the surface and its normal element dN_1 . Thus, the attractive force f per unit area of volume V_1 to the entire volume V_2 is

$$f_{V_1, V_2} = \iint F(r) dN_1 dV_2. \quad (4.2)$$

The work H_{12} per unit area that it takes to separate the two volumes is hence the integral over f_{dV_1, dV_2} from the volume elements' current distance r to the edge of the force's range d_{12} :

$$H_{12} = \iiint_r^{d_{12}} F(r') dr' dN_1 dV_2 \quad (4.3)$$

Let's assume the two volumes are in contact. If the two materials are actually both the same liquid, then separating them creates two new liquid-gas surfaces, costing work $H_{\ell\ell}$. Thus, $H_{\ell\ell}$ can be interpreted as a *tension* that tries to keep the liquid surface together. It must be equal to the tensions of the two newly created surfaces, i.e. $2\sigma_{\ell g} = H_{\ell\ell}$. Similarly, if the materials are liquid and solid, then separating the solid-liquid interface produces a solid-gas interface with surface tension σ_{sg} and a liquid-gas interface, hence

$$H_{sl} = \sigma_{\ell g} + \sigma_{sg} - \sigma_{sl}. \quad (4.4)$$

Surface tensions are naturally associated with the direction of the normal vector of the surface. Any equilibrium configuration must balance all surface tensions. Considering Fig. 4.1, the shape of the droplet must balance the tensions of the three interfaces such that the gas is not intruding the interface between the solid and the liquid. This is achieved by an angle θ between the interfaces and captured in *Young's equation* [173]

$$0 = \sigma_{\ell g} \cos \theta + \sigma_{sl} - \sigma_{sg} \quad (4.5)$$

Using surface tensions to explain contact angles and capillary phenomena also comes with the difficulty of trying to understand what a solid-gas surface tension actually means. Separating such an interface would normally imply that pressure and temperature have to be taken into account. Nevertheless, *Young's equation* has provided a satisfying explanation for a long time and is still commonly found in textbooks.

In order to understand capillary and wetting phenomena better, more detailed models have to be considered. In general, the models can be classified into macroscopic, mesoscopic and microscopic models. *Young's equation* is a macroscopic model. The involved objects can be seen with the naked eye. Microscopic models take into account the varying densities of particles of the materials involved and their interactions. Such fluctuations are not visible and it takes a lot of effort, usually by using simulations, to connect them to macroscopic phenomena [95, 172]. Mesoscopic descriptions lie on scales in between. They do not take microscopic fluctuations into account, but treat macroscopic phenomena more accurately than macroscopic models.

Such a mesoscopic model is the subject of the following sections. It will be used to understand some of the rich phenomenology of wetting on structured surfaces.

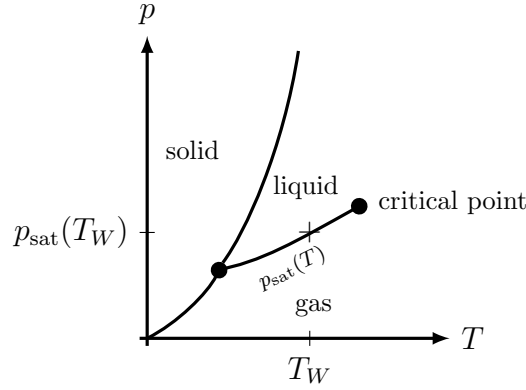


Figure 4.2: Typical phase diagram of a material with liquid-gas coexistence line $p_{\text{sat}}(T)$. Along this line is the wetting transition point $(T_W, p_{\text{sat}}(T_W))$, where the contact angle θ vanishes. At the end of the coexistence line, there is a critical point beyond which liquid and gas are indistinguishable.

4.2 WETTING TRANSITIONS ON GENERAL STRUCTURED SURFACES

The study of wetting phenomena is concerned with the formation of liquid layers of a fluid on solid surfaces. They exhibit a rich phenomenology [11, 141] and are studied using models with varying degree of detail [136, 115, 114].

Central to the study of wetting are phase transitions between different heights or shapes of the liquid layer. The wetting transition is the transition *on* (not across) the liquid-gas coexistence line $p_{\text{sat}}(T)$, see Fig. 4.2, where the contact angle vanishes. This phenomenon has been extensively studied [149, 12]. For the remainder of this chapter, I assume that this is a first-order phase transition. This means that the contact angle θ changes discontinuously at a wetting temperature T_W along $p_{\text{sat}}(T)$. Above the temperature T_W , macroscopic amounts of liquid are adsorbed on the surface. Below T_W , only a mesoscopic liquid layer extends on the solid.

Furthermore, the phenomenon of pre-wetting can appear close to the coexistence line $p_{\text{sat}}(T)$ in the gas phase, because the wetting transition is of first-order [136, 172]. Pre-wetting is the transition between two mesoscopic liquid heights, i.e. a thin and a thick layer. Its transition line originates at the wetting transition point and extends into the gas phase where it ends at its critical point, see Fig. 4.3.

These phase transitions play an important role in creating small structures on integrated

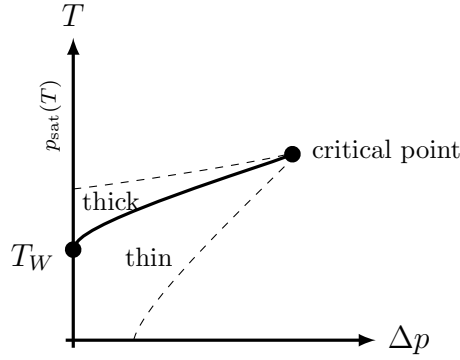


Figure 4.3: Phase diagram of a fluid exposed to a solid surface. The fluid's liquid-gas coexistence line $p_{\text{sat}}(T)$ is aligned with the y -axis. The x -axis shows $\Delta p = p_{\text{sat}}(T) - p$. The pre-wetting transition line extends from the wetting transition point $(p_{\text{sat}}(T_W), T_W)$ into the gas phase, along the (solid) line $(\Delta p, T_{PW}(\Delta p))$. Above the pre-wetting line, a thick liquid layer forms on the solid surface, below the pre-wetting line, a thin liquid layer forms. Both phases merge at a critical point. Dashed lines: spinodals of the pre-wetting transition. Between the upper spinodal and the pre-wetting transition line, the thin configuration exists as a meta-stable state. Analogously, between the lower spinodal and the pre-wetting transition line, the thick configuration exists as a meta-stable state.

circuits, sensors and displays. During the manufacturing process, gaseous and liquid layers condensate and freeze onto structured surfaces. This procedure, called epitaxy, creates solids of different materials with the desired structure and electrical properties. Later on, parts of the structure are removed using acids during the etching phase. If the etching is anisotropic, the surface structure is used to remove materials with specified orientations. Observing such procedures experimentally during a wet or gaseous phase is very difficult. Often, the results can only be observed once the involved materials have been removed or have become solid because the measurements are done using electron microscopes [171, 159]. Another method consists of measuring the mass of the liquid film by analysing the vibrations of the solid substrate and liquid film confirming the predicted complete wetting phase transition [16, 17]. Alternative approaches use large polymers [137] and colloid-polymer mixtures [1] to quantify fluctuations of the liquid layers. However, direct experimental verification of pre-wetting transitions on structured surfaces are still not available and are currently being researched.

What are the considered surfaces? In the context of wetting phenomena, structured surfaces are simplified models of solid surfaces which are defined by their shape and interaction with a liquid forming a layer on it. Their microscopic characteristics are

assumed to be irrelevant. Temperature and pressure changes are considered for the liquid, gas, and their effective interaction with the solid surface, however, they are deemed unimportant for the solid surface's shape or position. Henceforth, structured surfaces are determined by their profile, given as a function, and an effective potential density W of the interaction with the liquid height.

Let $z(x, y)$ be the function describing the profile of the structured surface. It marks the boundary between the solid which fills the space $\{< z\}$ and the fluid filling the space $\{> z\}$. Let $\ell(x, y)$ be the boundary between the liquid on the solid and the gas above the liquid, and let $\eta = \ell - z$ be the vertical height of the liquid layer, see Fig. 4.6.

In this setup, an effective interaction potential density $W(\eta(x, y))$ describes the local energy density landscape of the liquid height [34, 136, 113]:

$$W(\eta) = \frac{A}{\eta^2} - \frac{B}{\eta^3} + \frac{D}{\eta^4} + \Delta p \eta, \quad (4.6)$$

with the Hamaker constants $A(T) > 0$, $B > 0$ and $D > 0$ where the latter are assumed to be independent of temperature and pressure [62]. The pressure difference to the pressure p_{sat} at liquid-gas coexistence is denoted by Δp and is positive in the gas phase. Although W is a potential *density*, I will simply refer to it as potential in the following. For this analysis, the Hamaker constant A is assumed to be proportional to the temperature distance from the wetting transition $A(T) - A(T_W) \propto T - T_W$, at which the contact angle vanishes [131].

The potential W describes long-range interactions inducing a first-order wetting transition, as will be discussed in Sec. 4.3. It could be replaced by a short-range potential where the terms $1/\eta^k$ are replaced by $e^{-k\eta}$ [15]. A potential for a continuous wetting transition would equal $W_c = -A/\eta^2 + B/\eta^3 + \Delta p \eta$ [113]. However, I am not considering such cases in the following.

The minima of W are at energetically favourable liquid heights. The effective free energy of the liquid is

$$F[\ell] = \int \left(\frac{\sigma}{2} (\nabla \ell)^2 + W(\ell - z) \right) dx dy, \quad (4.7)$$

with the surface tension σ and the gradient $\nabla \ell$ of the liquid-gas boundary ℓ [113].

On a flat surface with constant profile function $z_F(x, y) = \text{const.}$, a liquid height can attain the local energy minimum everywhere and the complete phase diagram is given in Fig. 4.3. However, on a more complexly structured surface, the liquid height will balance local energy costs and achieve a global energy minimum. It will balance minimising the bending of the liquid layer (enforced by its surface tension) and keeping its height as close as possible to the minimum of W .

Minimising the free energy Eq. (4.7) with respect to η leads to the differential equation

$$\sigma (\eta_{xx} + \eta_{yy} + z_{xx} + z_{yy}) = W'(\eta), \quad (4.8)$$

where the indices represent derivatives with respect to x or y , e.g. η_{xx} is the second derivative of η w.r.t. x and where W' denotes the first derivative of the potential W with respect to the liquid height η . Eq. (4.8) is a second order, non-linear partial differential equation which is difficult to solve in general. However, some advances were made in [132]. Its derivation, using variational calculus, can be found in the appendix, see Sec. D.1. In Secs. 4.3 to 4.6, simple examples are presented that can be solved almost completely.

The simplifications used in these sections build on transforming the partial differential equation Eq. (4.8) into an ordinary differential equation by choosing structured surfaces which are translation invariant in the y -direction. This assumption will make the liquid height η also y -invariant. A further simplification is achieved by only considering piecewise linear structured surfaces whose second derivatives are zero. With these simplifications, Eq. (4.8) becomes

$$\sigma \eta_{xx} = W'(\eta), \quad (4.9)$$

which is an ordinary, non-linear differential (Euler-Lagrange) equation. Superficially, Eq. (4.8) is analogous to Newton's second law, force equals mass times acceleration. Here, W' acts like a force that bends the liquid-gas boundary proportionally to its stiffness, the surface tension σ , and the profiles coordinate x plays the rôle of time.

The Euler-Lagrange equation Eq. (4.8) remains valid for any y -translation-invariant structured surface whose profile consists of segments of straight lines. It is wrong for bent surfaces, i.e. for profiles with $z_{xx} \neq 0$. It is also not valid for surfaces which are

not translation invariant in the y -direction.

Furthermore, Euler-Lagrange problems require fixed boundary conditions, such that functional variations are constrained to vanish at the boundary. Here, the boundary is at $x \rightarrow \pm\infty$ and its associated condition is that the liquid height converges to the height η_π where the interaction potential W reaches its global minimum.

When multiplied with η_x and integrated over x , it is transformed into

$$\frac{\sigma}{2}\eta_x^2 = W(\eta) + C, \quad (4.10)$$

where C is an integration constant. Eq. (4.10) can be transformed to

$$\boxed{\eta_x = \pm \sqrt{\frac{2\Delta W(\eta)}{\sigma}}}, \quad (4.11)$$

with $\Delta W(\eta) = W(\eta) + C$. Eq. (4.11) is the main equation for finding wetting phase transitions on structured surfaces. It is discussed and interpreted for the plane, wedge, and apex in Secs. 4.3, 4.4, 4.5 and 4.6. In general, the differential equation Eq. (4.11) cannot be solved analytically. Nevertheless, phase transitions can be identified using a graphical analysis, which is similar to Maxwell's equal area construction [27], and which was picked up by *Cahn* in the context of wetting [20]. The main steps of this graphical construction are explained in the following.

Depending on the chosen structured surface and its symmetry, additional conditions will apply to η_x . These symmetries narrow down the possible values of η_x at points $x_a, x_b, \dots$ and, via Eq. (4.11), link them to a few possible values of η , say $\eta_{1a}, \eta_{2a}, \dots, \eta_{1b}, \eta_{2b}, \dots$. For the plane, all points are symmetry points and any could be chosen. For the apex and wedge, the corner point, say $x = 0$, is the unique symmetry point.

Let's assume there is only one such point $x_a = 0$. Once the height at x_a is fixed, the rest of the liquid height function $\eta(x)$ is determined by the differential equation Eq. (4.11), i.e. $\eta_1, \eta_2, \dots$ act as possible boundary conditions to the differential equation. Thus, I find several eligible liquid profiles. But which one is the correct one? From a physics perspective, the profiles with the lowest free energy will be preferred and assumed by the system.

The free energy $F(x_a, x_b)$ of a specific profile segment from $x = x_a$ to x_b is calculated based on Eq. (4.7) as follows:

$$F(x_a, x_b) = \int_{x_a}^{x_b} \frac{\sigma}{2} \ell_x(x)^2 + W(\eta(x)) dx \quad (4.12)$$

$$= \int_{x_a}^{x_b} \frac{\sigma}{2} \left(\eta_x(x)^2 + 2\eta_x(x) \underbrace{z_{W_x}(x)}_{=\alpha} + z_{W_x}(x)^2 \right) + W(\eta(x)) dx, \quad (4.13)$$

where α is the slope of the structured surface between x_a and x_b . For the plane, $\alpha = 0$, but for the wedge and apex the slope is non-zero and the segments are $(-\infty, 0)$ and $(0, \infty)$.

The goal is to find the minimal energy configuration for the liquid height η . Therefore, any part of the integral that does not depend on η can be ignored. Here, the term $z_{W_x}(x)^2$ is ignored in further calculations.

Eq. (4.11) can be used to simplify the integral and by replacing variable x by variable η with $\eta_x dx = d\eta$. For profile k with $\eta(x_a) = \eta_{ka}$ and $\eta(x_b) = \eta_{jb}$, the free energy is equal to:

$$F_k(x_a, x_b) = \sigma \int_{\eta_{ka}}^{\eta_{jb}} \left(\sqrt{\frac{2\Delta W(\eta)}{\sigma}} - \alpha \right) d\eta. \quad (4.14)$$

The constant α cannot be omitted because the integral's boundaries depend on η . Eq. (4.14) shows the free energy of the profile segment from $x = x_a$ to x_b . For other segments, an analogous calculation can be made and their free energies can be summed up to obtain the free energy of the entire profile from $x = -\infty$ to $x = +\infty$. At infinity, the liquid will behave like on a plane and hence, it will assume the liquid height η_π where the potential W is minimal. Once the free energy of a liquid profile is obtained, it can be compared to the energies of other possible liquid profiles.

4.3 WETTING AND PRE-WETTING TRANSITION OF THE PLANE

In order to understand the rich phenomenology of wetting and pre-wetting on structured surfaces, I first introduce these transitions for the plane. Eq. (4.11) can be used to find the liquid profile. As the plane is translation-invariant, the liquid layer profile should show the same symmetry and also be translation-invariant. This has three implications:

- because of translation-invariance, $\eta_x = 0$,
- $\forall x : \eta(x) = \eta_\pi := \operatorname{argmin}(\Delta W(\eta))$, because in thermal equilibrium, the energetically minimal configuration is assumed,
- the integration constant C must achieve that $\sqrt{\frac{2\Delta W(\eta_\pi)}{\sigma}} = 0$.

What are the possible cases for the liquid height $\eta = \eta_\pi$? For $\Delta p = 0$ at $T = T_W$, η_π is either a finite height or it is at $\eta = +\infty$. The transition can be seen in plots of η_x over η as the transition of the global minimum from a finite η to $\eta = +\infty$, shown in Fig. 4.4. In the dry phase $T < T_W$, only a mesoscopic liquid layer of height $\eta_\pi < \infty$ forms. However, at $T = T_W$, a discontinuous transition occurs: a finite and an infinite liquid layer height are equally likely. In the wet phase $T > T_W$, the global minimum of $\sqrt{2\Delta W(\eta)/\sigma}$ is at $\eta = \infty$ and a macroscopic liquid layer forms on the solid. This is the wetting transition on the plane.

The *pre-wetting* transition occurs in the gas phase, i.e. for $\Delta p > 0$. The influence of the changed pressure according to Eq. (4.6) is shown in Fig. 4.5. Here, the global minimum of η_x is always at a finite liquid height $\eta_\pi < \infty$. However, there is a discontinuous transition where the global minimum changes from one local minimum to the other. This is the pre-wetting transition, where the mesoscopic liquid height changes from a thin to a thick phase. The phase space points where both local minima are also global minima form the transition line $T = T_{PW}(\Delta p)$, shown in Fig. 4.3.

In the thick phase at constant temperature, the height of the thick layer diverges as $\Delta p \rightarrow 0$, connecting the thick phase of the pre-wetting transition to the wet phase of the wetting transition. Hence, the pre-wetting transition line extends from the wetting transition point into the gas phase. The pre-wetting transition line ends in a critical point where the two global minima merge into one and the thin and thick phase become indistinguishable.

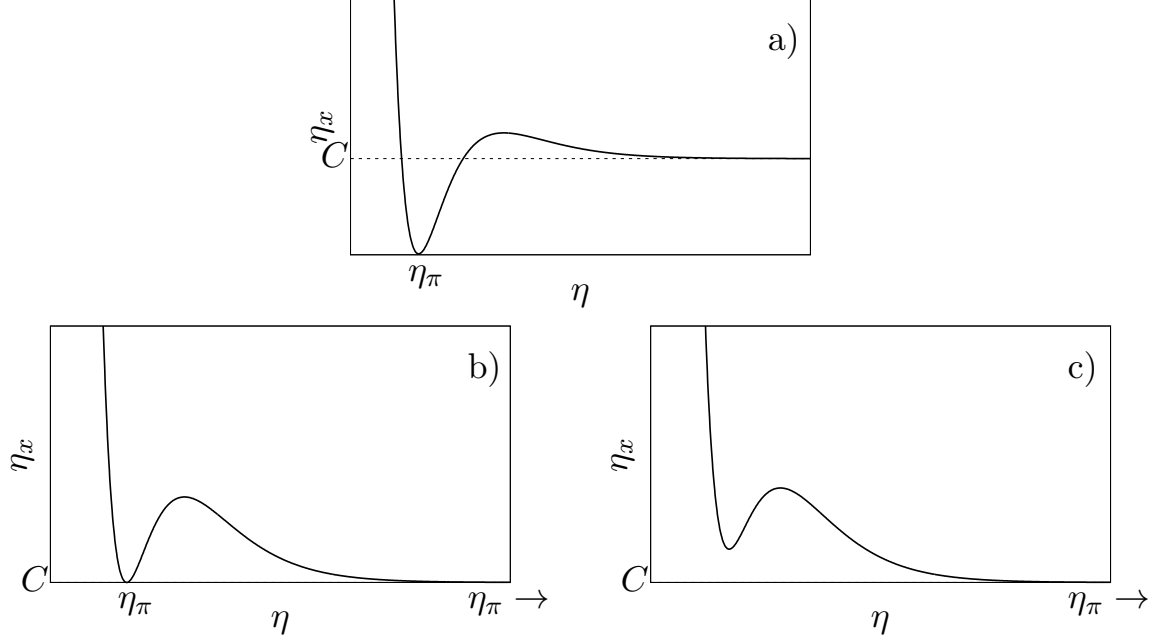


Figure 4.4: Sketch of the three different scenarios for Eq. (4.11) for the wetting on a plane. η_π marks the global minimum of the function Eq. (4.11). a) Dry phase $T < T_W$, the liquid layer assumes height η_π and $C > 0$. b) Wetting transition point $T = T_W$. Two liquid heights are energetically equal, finite height and infinite height. $C = 0$. c) Wet phase $T > T_W$. An infinite liquid layer is energetically preferable. $C = 0$.

Furthermore in the $(T, \Delta p)$ phase space, the two minima of $\eta_x = \sqrt{2\Delta W(\eta)/\sigma}$ only exist in an area which is bound by three lines: $\Delta p = 0$ and two lines, called spinodals, shown in Fig. 4.3 as dashed lines. Here, one phase is the equilibrium state, while the other exists as a meta-stable state. Outside of this area, only one local minimum exists.

Similar arguments and discussions of curves will be used to understand the possible phase transitions of the wedge and apex in the following sections.

4.4 FILLING TRANSITION OF THE WEDGE

In this section, the influence of a simple structure of a surface on the wetting transition (i.e. $\Delta p = 0$) is explored using the example of the wedge, shown in Fig. 4.6. The liquid height at the wedge point is denoted by η_w . The wedge and its filling and pre-filling transition have been studied previously in [136], their fluctuations in [113, 112, 130], and using density functional theory in [94, 95]. It also has been studied with a different

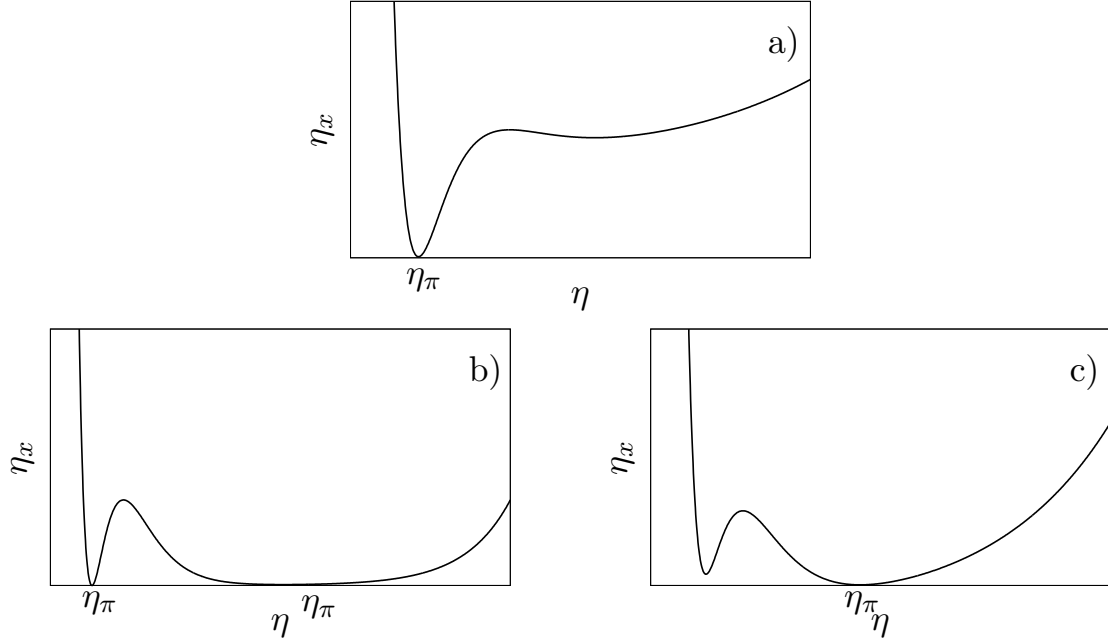


Figure 4.5: Sketch of the three different scenarios of Eq. (4.11) for pre-wetting on a plane. η_π marks the global minimum of the function Eq. (4.11). a) Thin phase $T < T_{PW}(\Delta p)$, the liquid layer assumes a thin height. b) Pre-wetting transition point $T = T_{PW}(\Delta p)$. Two liquid heights are energetically equal, thin and thick. c) In the thick phase $T > T_{PW}(\Delta p)$. A thick liquid layer is energetically preferable.

orientation as a corner in [116]. The profile of the wedge is given by

$$z_W(x, y) = z_W(x) = \alpha|x|, \quad (4.15)$$

with $\alpha > 0$.

As the surface profile is symmetric around $x = 0$, it is expected that the profile of the liquid layer shows the same symmetry. Furthermore, I assume that the liquid layer profile $\ell(x)$ is continuously differentiable. Hence, $\ell_x(0) = 0$, which implies the following conditions on the liquid height $\eta = \ell - z_W$:

$$\lim_{x \rightarrow 0^-} \eta_x = \alpha \quad (4.16)$$

$$\lim_{x \rightarrow 0^+} \eta_x = -\alpha \quad (4.17)$$

Following Eq. (4.11) and its plots in Fig. 4.7, I can see where $\alpha = \eta_x$ can occur.

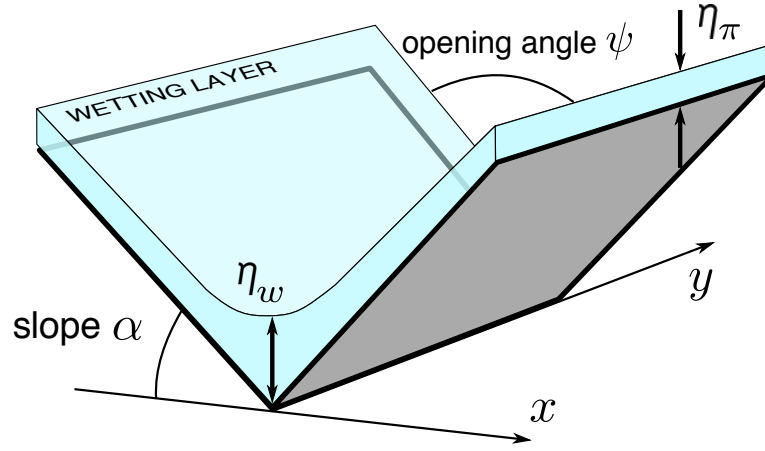


Figure 4.6: Setup of the wedge with slope $\pm\alpha$. Due to the symmetry of the solid surface, the liquid layer is symmetric too. The liquid profile is assumed to be continuously differentiable at the wedge $x = 0$, where its height equals η_w . Far away from the wedge, the liquid layer assumes the energetically minimising height η_π . Figure adapted from [133].

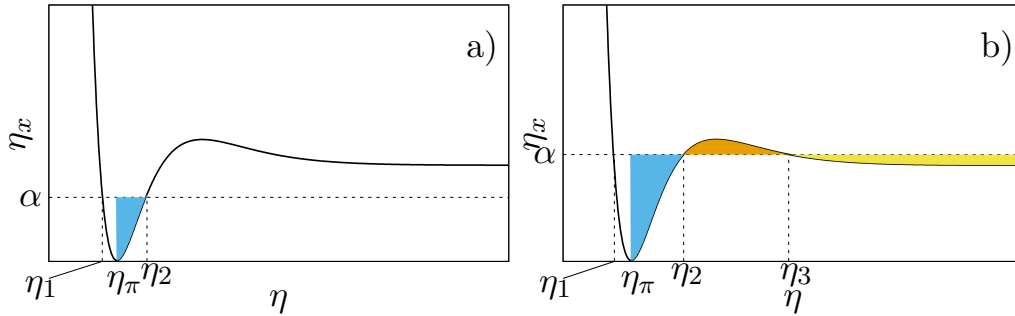


Figure 4.7: Sketch of $\eta_x = \sqrt{2\Delta W/\sigma}$ and $\eta_x = \alpha$ for the wedge filling transition. a) empty phase, b) filled phase. The free energy of the possible solutions can be compared by considering the coloured areas: Above the line $\eta_x = \alpha$, areas count positive for the free energy, below the line, areas count negative for the free energy. η_1 is unphysical as a solution. The negative blue area represents the free energy of the liquid profile with liquid height η_2 at $x = 0$, given that away from the wedge, the liquid height tends to η_π . The orange minus the blue area represents the free energy of the liquid profile with liquid height η_3 at $x = 0$, given that away from the wedge, the liquid height tends to η_π . However, the free energy of a completely filled wedge is even lower, represented by the yellow area. Hence, for the shown wedge slope α in b), the system is in the filled phase of the filling transition. The empty phase appears where α is small enough so that the third solution η_3 does not exist, see a).

I start by assuming that the system is in the dry phase, see Fig. 4.4 a).

Case $\alpha < C$: For $x \leq 0$, I have to consider the condition in Eq. (4.16). There are two liquid heights η such that $\eta_x = \alpha$: one smaller than η_π , η_1 , and one larger than η_π , η_2 . How can I decide which one is the solution? As I am assuming that ℓ and ℓ_x are continuous, η must also be continuous. Hence, I can read off the liquid height and its slope directly from the graph. Starting at $x = 0$, η will assume one of the two possible heights $\eta_{1/2}$, i.e. in configuration space, the system is at $(\eta_{1/2}, \eta_x = \alpha)$. Then, as x goes to $-\infty$, in configuration space the system (η, η_x) moves along the graph to $(\eta_\pi, 0)$, the global minimum of $\sqrt{2\Delta W(\eta)}/\sigma$.

But I can also imagine the opposite direction: starting at $x = -\infty$, going to $x = 0$, the system starts in configuration $(\eta_\pi, 0)$ and moves along the graph to configuration $(\eta_{1/2}, \eta_x = \alpha)$. As $\eta_1 < \eta_\pi$ but η_x is positive, I conclude that the solution η_1 is unphysical and the correct solution must be η_2 .

For $x \geq 0$, the same arguments apply but with changed signs. I have to consider $\eta_x = -\sqrt{2\Delta W(\eta)}/\sigma$ and the condition in Eq. (4.17). At $x = 0$, the system is at $(\eta_2, \eta_x = -\alpha)$. For $x \rightarrow \infty$, the system's configuration moves along the graph to configuration $(\eta_\pi, 0)$.

Case $\alpha \geq C$: Here a third solution for $\eta_x = \alpha$, η_3 , appears, and if α large enough, the two greater solutions merge and disappear at and above the local maximum of $\sqrt{2\Delta W(\eta)}/\sigma$. The smallest solution for $\eta_x = \alpha$, η_1 was already ruled out above because it is unphysical. But which of the two greater solutions, η_2 and η_3 is the right one? To answer this question, the free energies have to be compared, see Eq. (4.14). For the two possible liquid heights at $x = 0$, η_2 and η_3 , the free energies F_2 and F_3 are calculated, respectively. They can be compared by looking at the graph of Fig. 4.7. In this plot, coloured areas below the $\eta_x = \alpha$ line represent negative contributions to the free energy, above they represent positive free energy. The result is that η_2 (the smaller liquid height) is favoured over η_3 . However, there is a third option. What if the liquid profile would have an even larger liquid height, replacing η_π by $\eta > \eta_3$ and completely filling the wedge? Such a configuration (represented by the yellow area in the plot) would have a smaller free energy and therefore would be preferred over the other (finite) liquid heights. This is the hallmark of the filled phase.

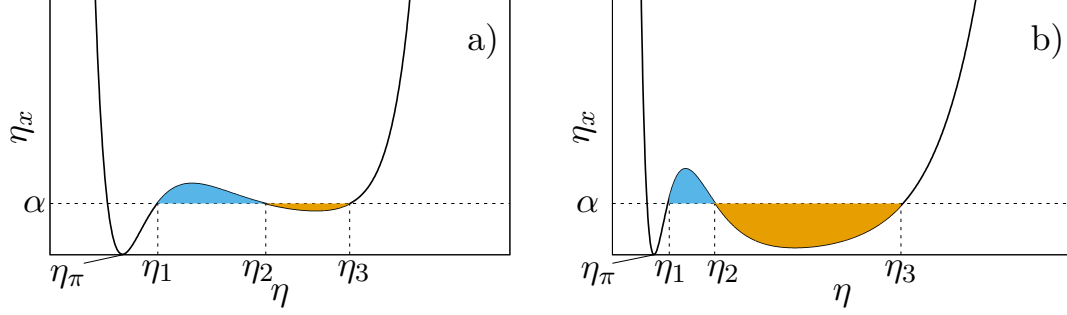


Figure 4.8: Sketch of the solutions for the pre-filling transition on a wedge. η_π marks the global minimum of the function Eq. (4.11). The spinodals are defined as the points in $(\Delta p, T)$ -space, where the second local minimum touches the $\eta_x = \alpha$ line (dashed horizontal line). a) If the blue area is larger than the orange area, then the liquid height will be η_1 at $x = 0$. b) If the orange area is larger than the blue, the liquid height will be η_3 at $x = 0$, see Eq. (4.18). The line in $(\Delta p, T)$ -space along which the blue and orange areas are equal is the pre-filling transition line.

The exact slope where $\alpha = \lim_{\eta \rightarrow \infty} \sqrt{2\Delta W(\eta)}/\sigma$ is the transition point between the empty and the filled phase of the wedge. Conversely, for each slope α , there exists a temperature $T_F(\alpha)$ where the filling phase transition occurs.

4.5 WEDGE PRE-FILLING TRANSITIONS

In this section, a phase transition of the liquid layer is explored which only can appear on a structured surface and not on a plane. The prototype of this transition can be seen on the wedge, which is used in this section as an example. The basic parameters for the wedge and its liquid layer are depicted in Fig. 4.6.

As the wedge profile is symmetric around $x = 0$, the liquid layer will also be symmetric w.r.t. $x = 0$. In addition, the liquid-gas transition line is a smooth function because of the surface tension. Therefore, on the symmetry axis, its slope should vanish, $\ell_x(0) = 0$. Translated to the liquid height η , its slope has the discontinuities at $x = 0$ expressed in Eqs. (4.16) and (4.17).

For the wedge, considering Fig. 4.8, there are two or four solutions for $\eta_x = \alpha$. In case there are only two, one of the two possible solutions is a liquid height $\eta_w < \eta_\pi$ which was found to be unphysical in Sec. 4.4. Hence, in this case, the greater solution of $\eta_x = \alpha$

is the right one. If there are four solutions possible, how can I choose the correct one? The energetically preferred solution is the one with the lowest free energy. The three possible, physical solutions for $\eta_w = \eta(0)$ are denoted by η_1 , η_2 , and η_3 , see Fig. 4.8. In Eqs. (4.12) to (4.14), it was explained how the free energies of the different solutions can be calculated and compared. Building on Eq. (4.14), the difference in free energy is computed by

$$F_1 - F_2 = 2\sigma \int_{\eta_2}^{\eta_1} \left(\sqrt{\frac{2\Delta W(\eta)}{\sigma}} - \alpha \right) d\eta, \quad (4.18)$$

and analogous expressions hold for $F_1 - F_3$ and $F_2 - F_3$. Therefore, the differences in free energy correspond to the coloured areas in Fig. 4.8.

Thus, it can be read off the plots in Fig. 4.8, which of the free energies F_1 , F_2 or F_3 is the smallest, by comparing the areas above and below the line $\eta_x = \alpha$. It follows that η_2 is never the preferred solution. Which of F_1 and F_3 is smaller depends on the exact position in phase space $(\Delta p, T)$. The points $(\Delta p, T_{PF}(\Delta p))$, where $F_1 = F_3$ is the transition line for the pre-filling phase transition.

Why is it called pre-filling? In the gas phase, the wedge cannot undergo the filling transition described in Sec. 4.4. However, in the pre-filled phase, a partial thick layer forms around the wedge point $x = 0$. The closer the system is to the filling phase, the higher and longer the thick shoulder extends along the walls. In the limit $\Delta p \rightarrow 0$, the layer thick layer's height and the length h diverge to ∞ . Example profiles of this extension of the thick layer for fixed Δp are shown in Fig. 4.9. The transition from the pre-filled wedge to the thick phase is continuous, while the transition from the thin phase to the pre-filled wedge is a first-order transition. The pre-filling line extends from the filling transition point $(\Delta p = 0, T_F(\alpha))$ to its own critical point which lies on the spinodal of the pre-wetting transition.

The pre-filling transition lines can be traced for a variety of wedge slopes α . There are two cases. If $\alpha > \alpha^*$, the pre-filling transition line is a continuous line from the filling transition point to the pre-wetting spinodal. However, if $\alpha < \alpha^*$, the pre-filling line intersects the pre-wetting line, shown in Fig. 4.10. The critical slope α^* is to be determined.

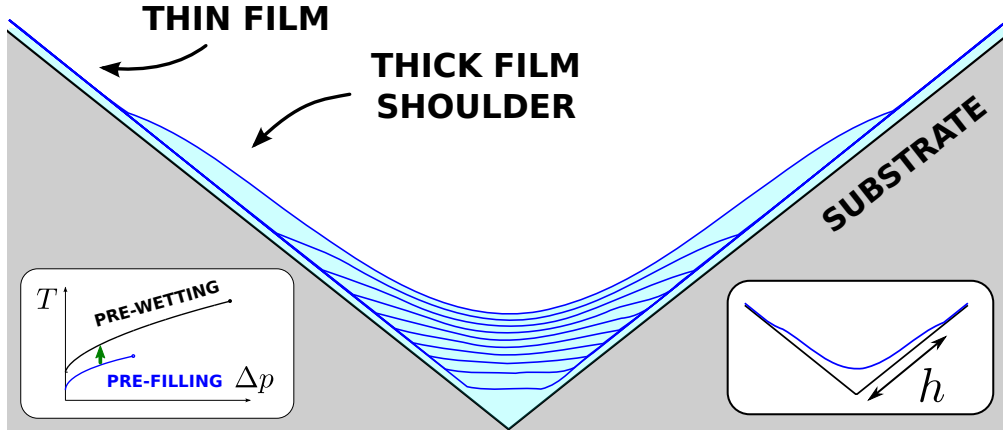


Figure 4.9: Main Figure: Profiles (thin blue lines) of the liquid layer of the wedge close to the pre-filling transition. At the pre-filling transition the liquid layer at the wedge $x = 0$ discontinuously changes to a thick liquid height. At higher temperatures, this thick layer extends along the walls until it covers the entire solid surface at the pre-wetting transition. Left inset: the phase diagram, black: pre-wetting transition line, blue: pre-filling transition line. The extension of the thick layer increases in the direction of the green arrow. Right inset: The extension h of the thick layer diverges as the pre-wetting line is approached. Figure from [133].

Whether the pre-filling line intersects the pre-wetting line or not depends on the slope of the wedge α . The temperatures of the two intersection points T_1^* and T_2^* along the pre-wetting line can be determined numerically for each value of α and plotted as a graph of a function $\tilde{\alpha}(T)$, shown in Fig. 4.11. At the critical slope α^* , both temperatures merge to one, and for slopes steeper than α^* , the pre-filling line does not intersect the pre-wetting line.

The value of α^* can be determined numerically in terms of the parameters B and D of the potential W as

$$\alpha^* \approx 0.10873 \sqrt{\frac{B^3}{16D^4\sigma}}, \quad (4.19)$$

see Eq. (4.6).

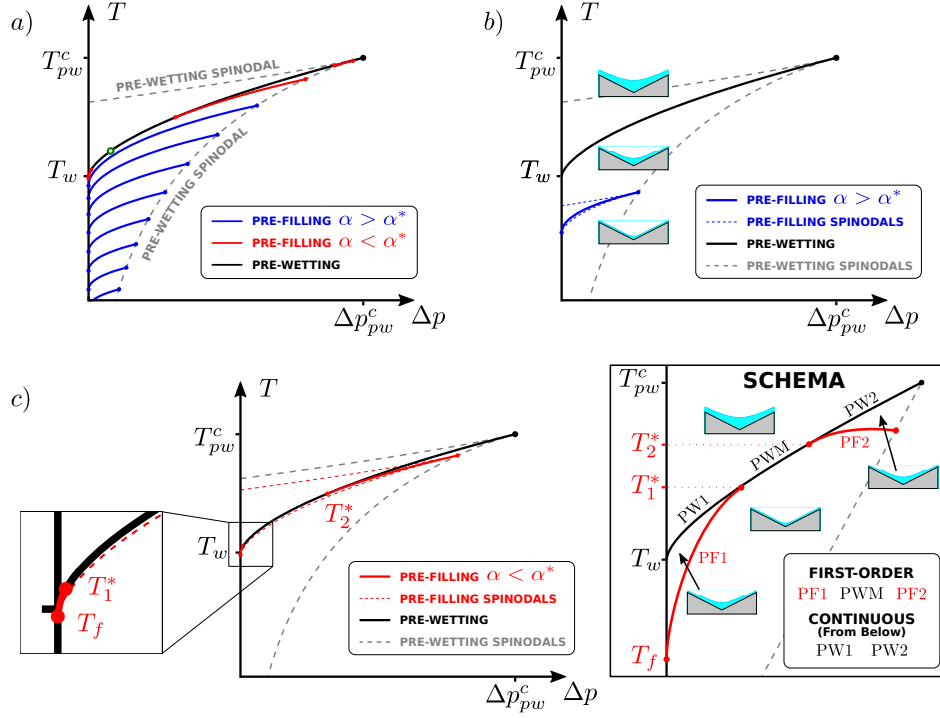


Figure 4.10: Phase diagram of the pre-filling transition. a) Several pre-filling transition lines with varying values of the wedge slope α . Blue lines: if $\alpha > \alpha^*$, the pre-filling line is continuous from the filling transition point to the pre-wetting spinodal. Red lines: if $\alpha < \alpha^*$, the pre-filling line intersects the pre-wetting line. b) Schematic of the different phases: below the pre-filling line, the liquid layer is thin everywhere, between pre-filling and pre-wetting line, the layer is thin at $x = \pm\infty$ but thick at the wedge $x = 0$, above the pre-wetting line, the liquid layer is thick everywhere. The pre-filling transition also has spinodals (dashed blue and red lines), between which the other phase exists as a meta-stable configuration. c) The discontinued pre-filling line has two parts and splits the pre-wetting line into three segments PW1, PWM and PW2, of which two are continuous transitions, while the middle segment is a first order transition. Figure from [133].

4.6 APEX UNBENDING

The focus of this section is on the implications of the pre-wetting transition for the apex.

While the liquid layers on an apex have been studied previously on the liquid-gas coexistence line [111], they have not been studied in the pre-wetting region until our publication [133]. The shape of the apex is determined by a profile function z_A which

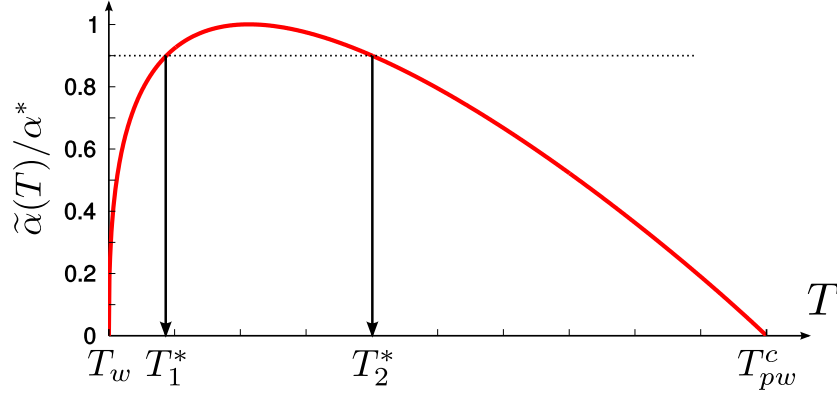


Figure 4.11: The two temperatures T_1^* and T_2^* of the intersection of the pre-filling line with the pre-wetting line depending on the wedge slope $\alpha = \tilde{\alpha}(T)$. For each choice of α (the example shown is for $\alpha = 0.9 \cdot \alpha^*$), there are two intersection points, except for the critical slope α^* , where they merge into one point. In the limit of vanishing slope $\alpha = 0$, the two intersection points converge to the wetting transition point and the end of the pre-wetting line because the wedge transforms into a plane, which does not exhibit a pre-filling transition. Figure from [133].

is independent of the y -coordinate. The profile function of the apex is

$$z_A(x, y) = z_A(x) = -\alpha|x|. \quad (4.20)$$

The parameter $\alpha > 0$ describes how steep the apex is. The solid fills the space underneath $\{< z_A\}$ the surface profile, whereas the space above $\{> z_A\}$ is filled with the fluid. A liquid film will form on the surface and its boundary to the gas is described by the function $\ell(x, y) = \ell(x)$. The situation for the apex is shown in Fig. 4.12. The liquid height at the apex point is denoted by η_a .

Compared to the pre-filling transition on the wedge, what changes for the apex? The conditions for the slope of the liquid height are different because the slope of the underlying solid surface is different:

$$\lim_{x \rightarrow 0^-} \eta_x = -\alpha \quad (4.21)$$

$$\lim_{x \rightarrow 0^+} \eta_x = \alpha. \quad (4.22)$$

However, as in the case of the wedge, there are two or four solutions for $\eta_x = \alpha$ and $\eta_x = \sqrt{2\Delta W(\eta)/\sigma}$, see Fig. 4.13. Which ones are the correct ones? While for the wedge, the smallest of the possible heights was found to be unphysical, for the apex,

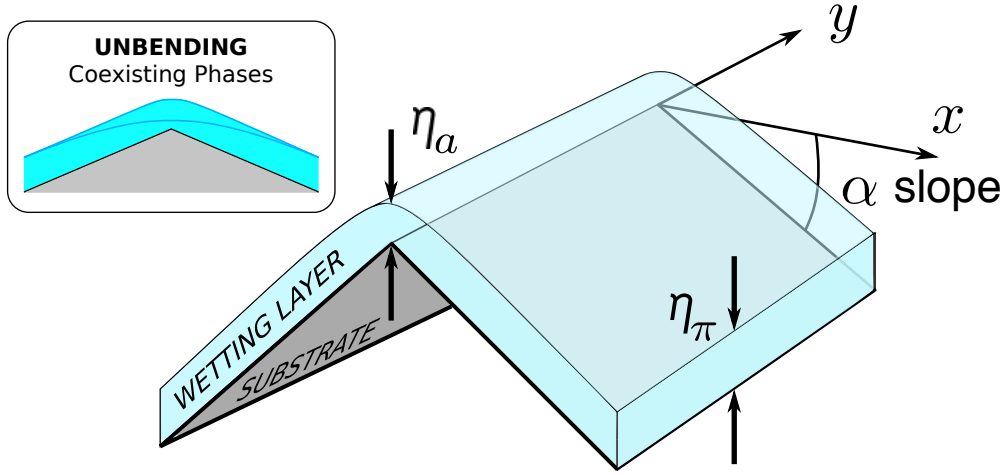


Figure 4.12: Wetting of an apex with slope $\pm\alpha$. Far away from the apex, the liquid height is given by the global minimum of $\sqrt{2\Delta W(\eta)/\sigma}$, η_π . At the apex, the liquid height is η_a and can have two different, physically plausible solutions. Figure adapted from [133].

it is the largest of the possible heights which is unphysical. The remaining (possibly three) solutions are denoted by η_1 , η_2 and η_3 , see Fig. 4.13.

As for the wedge pre-filling transition, the energetically preferred configuration for the apex can be found by considering the free energy. Following Eq. (4.12) to Eq. (4.14) and Eq. (4.18), the difference in free energy for the possible solutions with apex heights η_1 , η_2 and η_3 can be calculated as

$$F_1 - F_2 = 2\sigma \int_{\eta_1}^{\eta_2} \left(\sqrt{\frac{2\Delta W(\eta)}{\sigma}} - \alpha \right) d\eta, \quad (4.23)$$

where the integral boundaries are switched compared to pre-filling case on the wedge because the possible heights η_1 , η_2 are smaller than η_π . For $F_1 - F_3$ and $F_2 - F_3$, analogous expressions hold. The relation between F_1 , F_2 and F_3 can be therefore read off the plot in Fig. 4.13 by comparing the areas between $\eta_x = \alpha$ and $\eta_x = \sqrt{2\Delta W(\eta)/\sigma}$. Thus, η_2 is never the preferred configuration at $x = 0$, but between η_1 and η_3 , the decision depends on the location in phase space. The line along which $F_1 = F_3$ is the unbending phase transition line, shown in Fig. 4.14.

For temperatures above the unbending line, the apex has a thick liquid layer. At the

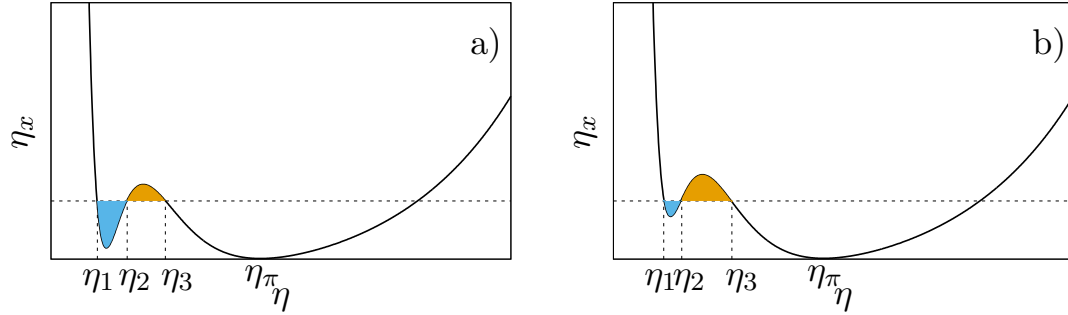


Figure 4.13: Sketch of the solutions for the unbending transition on an apex. η_π marks the global minimum of the function Eq. (4.11). The spinodals are defined as the points in $(\Delta p, T)$ -space, where the second local minimum touches the $\eta_x = \alpha$ line (dashed horizontal line). a) If the blue area is larger than the orange area, then the liquid height will be η_1 at $x = 0$. b) If the orange area is larger than the blue, the liquid height will be η_3 at $x = 0$, see Eq. (4.14). The line in $(\Delta p, T)$ -space along which the blue and orange areas are equal is the unbending transition line.

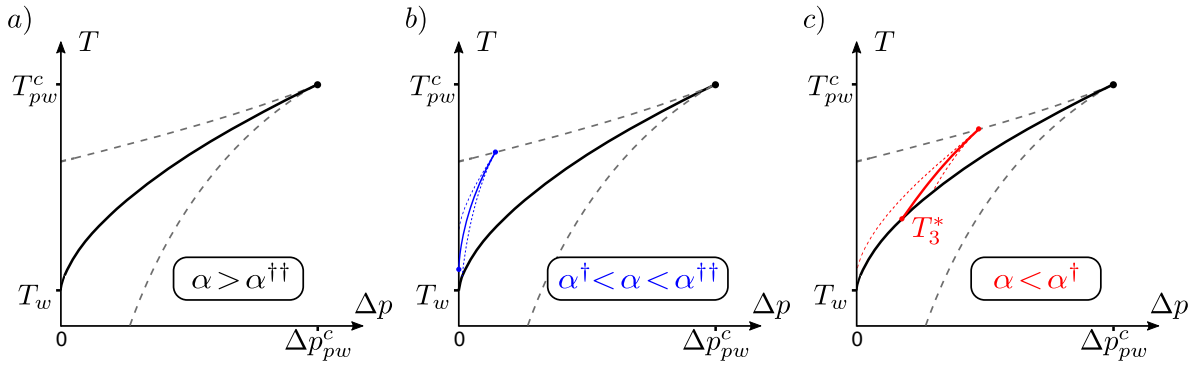


Figure 4.14: Phase diagram of the unbending transition. Black lines: pre-wetting line (solid) and its spinodals (dashed). Blue and red lines: unbending transition lines (solid) and their spinodals (dashed). The unbending only exists if $\alpha < \alpha^{\dagger\dagger}$. b) For $\alpha^\dagger < \alpha < \alpha^{\dagger\dagger}$, the unbending line starts at the liquid-gas transition line and ends at the pre-wetting spinodal. c) For $\alpha < \alpha^\dagger$, the unbending line starts at the pre-wetting line (at temperature T_3^*) and also ends at the pre-wetting spinodal. Figure from [133].

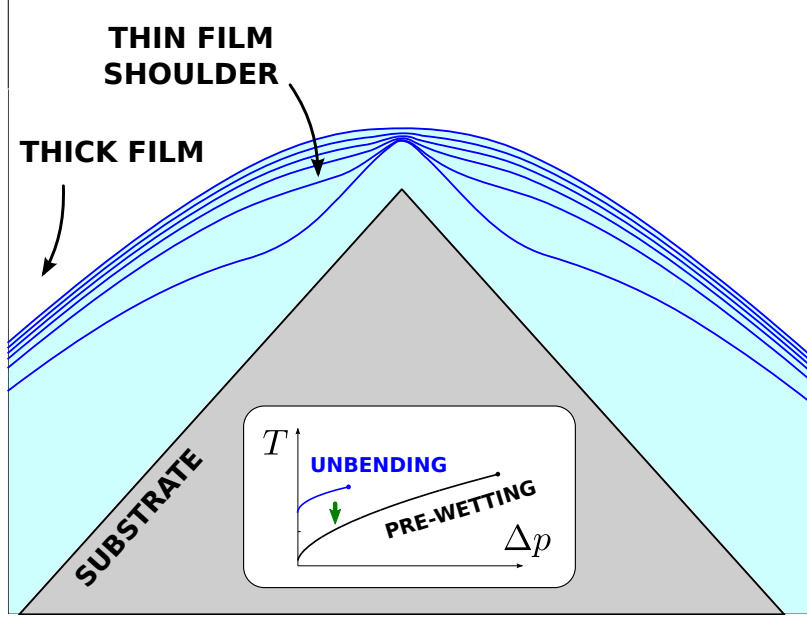


Figure 4.15: Main Figure: Profiles (thin blue lines) of the liquid layer on an apex. At the unbending transition, the height of the liquid layer discontinuously changes from thick to thin. At lower temperatures, this thin layer extends along the walls until it covers the entire solid surface at the pre-wetting transition. Inset: Pre-wetting line (black) and unbending line (blue). The extension of the thin layer diverges in the direction of the green arrow. Figure from [133].

unbending transition, the liquid height at $x = 0$ decreases discontinuously. This is a first order phase transition. For lower temperatures, a shoulder of thin liquid height extends along the apex walls, and as the pre-wetting line is approached, the length h of the thin layer diverges. At the pre-wetting line, the entire liquid layer is thin. This is a continuous phase transition. Several example profiles are shown in Fig. 4.15.

If α is small enough, the unbending line intersects the pre-wetting line once at a temperature T_3^* . The relation between α and T_3^* can be determined numerically and plotted as a function $\bar{\alpha}(T)$, shown in Fig. 4.16.

If α is large enough, no unbending transition exists. The critical slope $\alpha^{\dagger\dagger}$, above which unbending is lost, is defined as the largest height of all inflection points of $\sqrt{2\Delta W(\eta)}/\sigma$ on the pre-wetting spinodal of the thick phase.

The values $\alpha^\dagger$ and $\alpha^{\dagger\dagger}$ can be determined numerically in terms of α^* as $\alpha^\dagger \approx 2.7127\alpha^*$ and $\alpha^{\dagger\dagger} \approx 4.2242\alpha^*$.

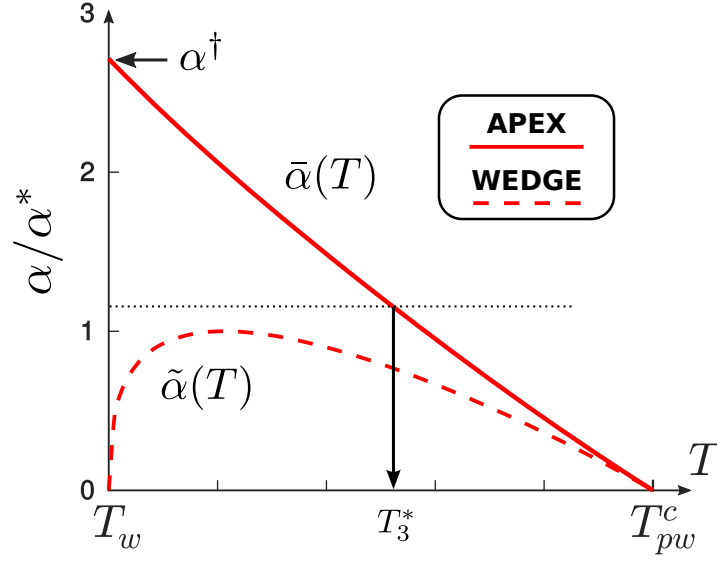


Figure 4.16: The temperature T_3^* of the intersection of the unbending line with the pre-wetting line as a function $\bar{\alpha}(T)$ (solid red line). For a specific choice of α (here as example $\alpha = 1.1 \cdot \alpha^*$), there is at most one temperature T_3^* for which an intersection is possible. If $\alpha > \alpha^\dagger$, no such temperature exists and the unbending line intersects the liquid-gas transition line instead. For comparison, the function $\tilde{\alpha}(T)$ for the wedge is shown as well (red dashed line). Figure from [133].

4.7 CONCLUSION

In this chapter, several phase transitions of liquid layers on structured surfaces were studied. These phase transitions are:

- the first order wetting transition on the liquid-gas coexistence line,
- the pre-wetting transition in the gas phase,
- the first-order filling transition of a wedge,
- the first-order pre-filling transition near a wedge, and
- the first order unbending transition near an apex.

These phase transitions only exist because the wetting transition is assumed to be of first order. The pre-wetting transition is a first order transition on the plane. However on the wedge and apex, it can be a continuous phase transition because the new liquid height

(thick / thin) spreads along the walls. Its extend diverges as the system approaches the pre-wetting transition line.

The wetting phenomena on the wedge and on the apex exemplify the potential myriad of phase transitions on more complexly structured surfaces. In particular, the apex and wedge can be regarded as the building blocks of all structured surfaces which have piecewise linear profiles and are translation invariant in one direction. These structured surfaces will be investigated in future research.

However, an important question is whether the possible phenomena are simply a concatenation of the transitions found for the wedge and apex or whether there is qualitatively new behaviour. A potential candidate for such new behaviour, which does not obey the translation invariance in one direction, is described in the following outlook.

4.7.1 OUTLOOK: SECONDARY PRE-WETTING

A ratchet is a structured surface that combines elements of the wedge and apex, see Fig. 4.17. The apex side will exhibit an unbending transition and the wedge side will show a pre-filling transition. This setup leads to a coexistence of a completely thick liquid layer extending from the wedge and a completely thin layer extending from the apex. The exact position in phase space of this coexistence line depends on the slopes and the distance L between the wedge and apex points, shown in Fig. 4.17.

Such a transition can be predicted directly from the apex and wedge unbending and pre-filling transition. The phase diagram will show two transition lines. One will be on the thick phase side of the pre-wetting line, marking the unbending transition on the apex shoulder. The other one will be on the thin phase side of the pre-wetting line, marking the pre-filling transition on the wedge shoulder. Thus, for the ratchet, the pre-wetting area is divided into four phases. This phase space, although containing rich phenomena, is not qualitatively new in itself.

However, it can be used in a truly three-dimensional arrangement where two small ratchets connect to a large ratchet, shown schematically in Fig. 4.17. In the figure, the orientation of the structure is turned such that it looks like an arrangement of upright steps: two small steps and one large step. Let's assume that the system is at the phase space point where the two small ratchets have thick liquid layers on both shoulders but

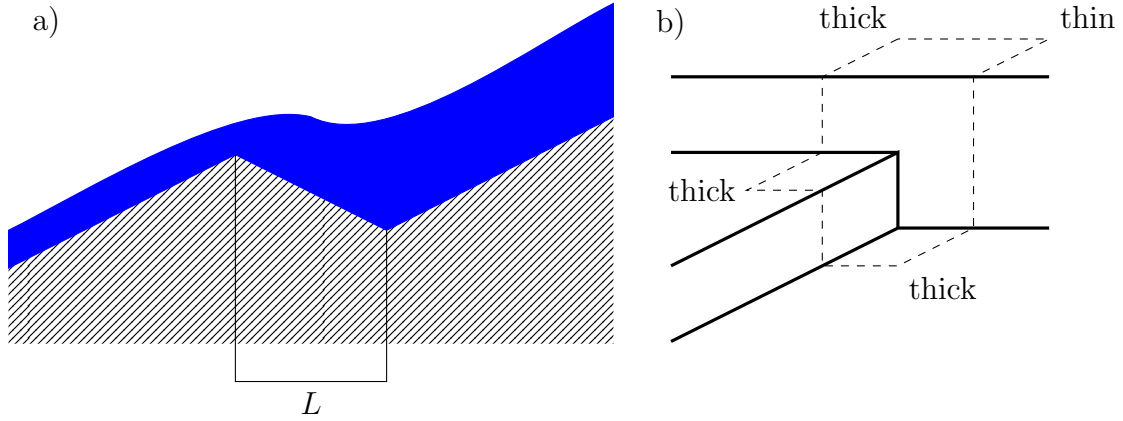


Figure 4.17: a) Sketch of liquid layer on ratchet at the coexistence of the thin layer on the apex shoulder and the thick layer on the wedge side. The apex and wedge points are a distance L apart. Where this coexistence occurs depends on L as well as the slopes of the wedge and apex parts. b) Sketch of the double ratchet shown in an upright orientation (solid lines). First small ratchet: front right to front left, Second small ratchet: front left to back left. Large ratchet: front right to back right. Along the dashed line, starting at the front right corner going clockwise: thick phase on bottom transitions to thick phase on intermediate step, which transitions to thick phase on top. However, if I go along the dashed line, starting at front right corner and go anti-clockwise, the thick phase on bottom transitions to a thin phase on the very top. Thus, there must be a line on the top half plane along which the thin and thick layers meet. In particular, there will be an angle at the edge associated with the transition.

the large ratchet is exactly at the coexistence of the thin layer on top and the thick layer on the bottom. Then, the half plane on top must have an area with a thin layer and an area with a thick layer. Therefore, on the top half plane must be a transition between the thin and thick layer, which I call secondary pre-wetting. Its transition line will intersect the edge where it will form an angle which can be interpreted as a contact angle, analogously to the contact angle of droplets.

In order to analyze this system, the previous assumption of y -translation invariance does not hold anymore and the partial differential equation Eq. (4.8) has to be considered instead. However, this is left to future research.

5

CONCLUSION

Statistical mechanics is a wide ranging field of which three topics were presented in this thesis. Its link to mathematical stochastic processes was studied in the example of continuous-time branching processes (Chapter 2). Its usefulness in biophysics was illustrated by modelling filament self-assembly and discovering emergent stochastic behaviour that can be matched with experiments (Chapter 3). Its effectiveness was demonstrated in the field of material science by exploring a plethora of phase transitions of liquid layers on structured surfaces (Chapter 4).

Stochastic processes' theory naturally connects to statistical mechanics because the essence of the latter is the probabilistic behaviour of physical objects. In the Ising model, each spin is identically distributed but not independent. In the branching process (Chapter 2), the particles' probabilities to produce offspring are identically distributed and independent. In the context of filament growth, each building block of the polymer (actin monomers or tubulin dimers) are probabilistically behaving in the same way (Chapter 3). When considering wetting phenomena (Chapter 4), each liquid profile element is under the same energetic constraints that allowed predicting the expected liquid heights. The microscopic, probabilistic rules are given in these cases. However, it is the collective behaviour of *all* the random variables involved that is of interest. In particular, whether this collective behaviour is 'more' than the sum of the

individual dynamics. Hence, statistical mechanics is the theory of collective stochastic processes. Exploring the connections between the physical and mathematical viewpoints has proven to be greatly beneficial and I will continue to pursue research in this area.

Biophysics is the perfect playground for statistical mechanics. The essential processes of life are not unique. In each cell, each tissue, each individual, each species and each evolutionary strand, they repeat again and again. These processes are probabilistic: mutations in evolution, cooperation in populations, the development of cells in a tissue, the expression of genes in a cell, and the transport processes within a cell, to name a few. Filament growth (Chapter 3) is a prime example: it can be broken down into the microscopic stochastic processes of chemical reactions and diffusion. Individually, both processes are well understood; however, new behaviour emerges in their superposition and can be observed as anomalous fluctuations and correlations.

Equilibrium material science is a classical application of statistical mechanics, where it shows its strengths immediately. It allows the explanation and prediction of wetting phenomena (Chapter 4) with effective potentials and graphical arguments. Thus, a complex phase space was explored and several phase transitions discovered. Furthermore, the considered apex and wedge structures allow the prediction of phase transitions occurring on more complexly structured surfaces by combining apex and wedge elements.

In summary, statistical mechanics has proven to be a versatile toolbox whose range of applications continues to grow and deserves more exploratory research. Therefore, I will focus my future work on joining the physical and mathematical viewpoints and on other applications in finance, biology and material science.

REFERENCES

- [1] Dirk G.A. L. Aarts, Matthias Schmidt, and Henk N.W. Lekkerkerker. “Direct Visual Observation of Thermal Capillary Waves”. In: *Science* 304.5672 (2004), pp. 847–850. DOI: [10.1126/science.1097116](https://doi.org/10.1126/science.1097116).
- [2] D.A. Adams, R.K. P. Zia, and B. Schmittmann. “Power Spectra of the Total Occupancy in the Totally Asymmetric Simple Exclusion Process”. In: *Phys. Rev. Lett.* 99.020601 (2007), pp. 1–4. DOI: [10.1103/PhysRevLett.99.020601](https://doi.org/10.1103/PhysRevLett.99.020601).
- [3] Paul Bachmann. *Zahlentheorie. Versuch einer Gesamtdarstellung dieser Wissenschaft in ihren Hauptteilen*. B.G. Teubner, 1894, p. 401.
- [4] Rodney J. Baxter. *Exactly Solvable Models in Statistical Mechanics*. Academic Press, 1982. ISBN: 0-12-083180-5.
- [5] Peter Bayley. “Why Microtubules Grow and Shrink”. In: *Nature* 363.6427 (1993), p. 309. DOI: [10.1038/363309a0](https://doi.org/10.1038/363309a0).
- [6] D. Beaglehole. “Profiles of the precursor of spreading drops of siloxane oil on glass, fused silica, and mica”. In: *J. Phys. Chem.* 93.2 (1989), pp. 893–899. DOI: [10.1021/j100339a067](https://doi.org/10.1021/j100339a067).
- [7] John M. Beggs and Dietmar Plenz. “Neuronal Avalanches in Neocortical Circuits”. In: *J. Neurosci.* 23.35 (2003), pp. 11167–11177. DOI: [10.1523/JNEUROSCI.23-35-11167.2003](https://doi.org/10.1523/JNEUROSCI.23-35-11167.2003).
- [8] Bertrand Berche, Malte Henkel, and Ralph Kenna. “Critical Phenomena: 150 Years since Cagniard de la Tour”. In: *J. Phys. Stud.* 13.3 (2009), pp. 1–4.
- [9] Howard C. Berg and Edward M. Purcell. “Physics of Chemoreception”. In: *Biophys. J.* 20.2 (1977), pp. 193–219. DOI: [10.1016/S0006-3495\(77\)85544-6](https://doi.org/10.1016/S0006-3495(77)85544-6).
- [10] Otto G. Berg and Peter H. von Hippel. “Diffusion-controlled macromolecular interactions”. In: *Ann. Rev. Biophys. Chem.* 14 (1985), pp. 131–160. DOI: [10.1146/annurev.bb.14.060185.001023](https://doi.org/10.1146/annurev.bb.14.060185.001023).

- [11] Kurt Binder. “Modeling of Wetting in Restricted Geometries”. In: *Annu. Rev. Mater. Res.* 38.1 (2008), pp. 123–142. DOI: [10.1146/annurev.matsci.38.060407.130302](https://doi.org/10.1146/annurev.matsci.38.060407.130302).
- [12] Daniel Bonn et al. “Wetting and spreading”. In: *Rev. Mod. Phys.* 81.2 (2009), pp. 739–805. DOI: [10.1103/RevModPhys.81.739](https://doi.org/10.1103/RevModPhys.81.739).
- [13] P. Bönsch et al. “UltrasMOOTH V-Grooves in InP by Two-step wet chemical etching”. In: *J. Electrochem. Soc.* 145.4 (1998), pp. 1273–1276. DOI: [10.1149/1.1838450](https://doi.org/10.1149/1.1838450).
- [14] J.-P. Bouchaud and R. Cont. “A Langevin approach to stock market fluctuations and crashes”. In: *Eur. Phys. J. B* 6 (1998), pp. 543–550. DOI: [10.1007/s100510050582](https://doi.org/10.1007/s100510050582).
- [15] E. Brézin, B.I. Halperin, and S. Leibler. “Critical Wetting in Three Dimensions”. In: *Phys. Rev. Lett.* 50.18 (1983), pp. 1387–1390. DOI: [10.1103/PhysRevLett.50.1387](https://doi.org/10.1103/PhysRevLett.50.1387).
- [16] L. Bruschi, A. Carlin, and G. Mistura. “Complete Wetting on a Linear Wedge”. In: *Phys. Rev. Lett.* 89.16 (2002), pp. 1–4. DOI: [10.1103/PhysRevLett.89.166101](https://doi.org/10.1103/PhysRevLett.89.166101).
- [17] L. Bruschi et al. “Crossover effects in the wetting of adsorbed films in linear wedges”. In: *Phys. Rev. E* 68.2 (2003), pp. 1–5. DOI: [10.1103/PhysRevE.68.021606](https://doi.org/10.1103/PhysRevE.68.021606).
- [18] Michael A. Buice and Jack D. Cowan. “Field-theoretic approach to fluctuation effects in neural networks”. In: *Phys. Rev. E* 75.5 (2007), pp. 1–14. DOI: [10.1103/PhysRevE.75.051919](https://doi.org/10.1103/PhysRevE.75.051919).
- [19] Charles Cagniard De La Tour. “Exposé de quelques résultats obtenues par l’action combinée de la chaleur et de la compression sur certains liquides, tels que l’eau, l’alcool, l’éther sulfurique et l’essence de pétrole rectifiée”. In: *Annales de Chimie et de Physique* 21 (1822), pp. 127–132.
- [20] John W. Cahn. “Critical point wetting”. In: *J. Chem. Phys.* 66.8 (1977), pp. 3667–3672. DOI: [10.1063/1.434402](https://doi.org/10.1063/1.434402).
- [21] David Callan. “A combinatorial Survey of Identities for the Double Factorial”. In: *arXiv:0906.1317v1* (2009), pp. 1–29.

- [22] John Cardy. *Lecture Notes on Field Theory and Nonequilibrium Statistical Mechanics*. 1998.
- [23] John L. Cardy and Uwe C. Täuber. “Field Theory of Branching and Annihilating Random Walks”. In: *J. Stat. Phys.* 90.1/2 (1998), pp. 1–56. DOI: [10.1023/A:1023233431588](https://doi.org/10.1023/A:1023233431588).
- [24] M. F. Carlier et al. “Synchronous oscillations in microtubule polymerization”. In: *Proc. Natl. Acad. Sci. Unit. States Am.* 84.15 (1987), pp. 5257–5261. DOI: [10.1073/pnas.84.15.5257](https://doi.org/10.1073/pnas.84.15.5257).
- [25] Marie-France Carlier, Dominique Pantaloni, and Edward D. Korn. “Evidence for an ATP Cap at the Ends of Actin Filaments and Its Regulation of the F-actin Steady State”. In: *J. Biol. Chem.* 259.16 (1984), pp. 9983–9986.
- [26] Yi-der Chen and Terrell Hill. “Theoretical studies on oscillations in microtubule polymerization”. In: *Proc. Natl. Acad. Sci. Unit. States Am.* 84.23 (1987), pp. 8419–8423. DOI: [10.1073/pnas.84.23.8419](https://doi.org/10.1073/pnas.84.23.8419).
- [27] James Clerk-Maxwell. “On the dynamical evidence of the molecular constitution of bodies”. In: *Nature* (1875), pp. 357–359.
- [28] L. Jonathan Cook, R.K.P. Zia, and B. Schmittmann. “Competition between multiple totally asymmetric simple exclusion processes for a finite pool of resources”. In: *Phys. Rev. E* 80.031142 (2009), pp. 1–12. DOI: [10.1103/PhysRevE.80.031142](https://doi.org/10.1103/PhysRevE.80.031142).
- [29] Kevin Costello. *Renormalization and Effective Field Theory*. American Mathematical Society, 2011.
- [30] P.T. Craddock. “From Hearth to furnace: evidences for the earliest metal smelting technologies in the eastern mediterranean”. In: *Paléorient* 26.2 (2000), pp. 151–165. DOI: [10.3406/paleo.2000.4716](https://doi.org/10.3406/paleo.2000.4716).
- [31] C. De Dominicis. “Techniques de renormalisation de la théorie des champs et dynamique des phénomènes critiques”. In: *J. Phys. Colloques* 37 (1976), pp. 247–253. DOI: [10.1051/jphyscol:1976138](https://doi.org/10.1051/jphyscol:1976138).
- [32] P.-G. de Gennes. “Chemotaxis: the role of internal delay”. In: *Eur. Biophys. J.* 33 (2004), pp. 691–693. DOI: [10.1007/s00249-004-0426-z](https://doi.org/10.1007/s00249-004-0426-z).

- [33] Damien Démoulin et al. “Power transduction of actin filaments ratcheting in vitro against a load”. In: *Proc. Natl. Acad. Sci.* 111.50 (2014), pp. 17845–17850. DOI: [0.1073/pnas.1414184111](https://doi.org/10.1073/pnas.1414184111).
- [34] S. Dietrich. *Phase Transitions and Critical Phenomena. Wetting Phenomena*. Ed. by C. Domb and J.L. Lebowitz. Vol. 12. Academic Press, 1988, p. 1. ISBN: 0122203127.
- [35] Paul Dirac. “A new notation for quantum mechanics”. In: *Math. Proc. Camb. Philos. Soc.* 35.3 (1939), pp. 416–418.
- [36] Paul Dirac. “The Lagrangian in Quantum Mechanics”. In: *Physikalische Zeitschrift der Sowjetunion* 3 (1933), pp. 312–320.
- [37] Masao Doi. “Second quantization representation for classical many-particle system”. In: *J. Phys. A: Math. Gen.* 9 (1976), pp. 1465–1477.
- [38] Detlev Drenkhahn and Thomas D. Pollard. “Elongation of Actin Filaments is a Diffusion-limited Reaction at the Barbed End and Is Accelerated by Inert Macromolecules”. In: *J. Biol. Chem.* 261 (1986), pp. 12754–12758.
- [39] Editors of Encyclopaedia Britannica. *Aeolipile*. 2016. URL: <https://www.britannica.com/technology/aeolipile> (visited on 03/22/2019).
- [40] Edward H. Egelman. “The structure of F-actin”. In: *J. Muscle Res. Cell. Motil.* 6 (1985), pp. 129–151. DOI: [10.1007/BF00713056](https://doi.org/10.1007/BF00713056).
- [41] A.H. Ellison et al. “Contact Angles of Mercury on Various Surfaces and the Effect of Temperature”. In: *J. Chem. Eng. Data* 12.4 (1967), pp. 607–609. DOI: [10.1021/je60035a037](https://doi.org/10.1021/je60035a037).
- [42] Jeremy L. England. “Statistical Physics of self-replication”. In: *J. Chem. Phys.* 139.121923 (2013), pp. 1–8. DOI: [10.1063/1.4818538](https://doi.org/10.1063/1.4818538).
- [43] Edgar F. Ethington. “Interfacial contact angle measurements of water, mercury, and 20 organic liquids on quartz, calcite, biotite, and Ca-montmorillonite substrates”. In: *US Department of the interior, geological survey* 90.409 (1990), pp. 1–18.
- [44] Lawrence C. Evans. *Partial Differential Equations*. 2nd ed. AMS, 2010. ISBN: 978-0-8218-4974-3.

- [45] Richard P. Feynman. “Principles of least action in quantum mechanics”. PhD thesis. Princeton University, 1942.
- [46] Gerald B. Folland. *Quantum Field Theory. A tourist guide for mathematicians*. AMS, 2008. ISBN: 978-0-8218-4705-3.
- [47] Maurice Fréchet. “Sur les ensembles de fonctions et les opérations linéaires”. In: *Comptes Rendues* 144 (1907), pp. 1414–1416.
- [48] Carl Frieden. “Actin and Tubulin Polymerization: The Use of Kinetic Methods to Determine Mechanism”. In: *Annu. Rev. Biophys. Bio.* 14 (1985), pp. 189–210.
- [49] Ikuko Fujiwara, Dimitrios Vavylonis, and Thomas D. Pollard. “Polymerization kinetics of ADP- and ADP-P_i-actin determined by fluorescence microscopy”. In: *Proc. Natl. Acad. Sci. Unit. States Am.* 104 (2007), pp. 8827–8832.
- [50] Ikuko Fujiwara et al. “Microscopic analysis of polymerization dynamics with individual actin filaments”. In: *Nat. Cell Biol.* 4 (2002), pp. 666–673.
- [51] Rosalba Garcia-Millan et al. “Field-theoretic approach to the universality of branching processes”. In: *Phys. Rev. E* 98 (062107 2018), pp. 1–11.
- [52] Melissa K. Gardner, Marija Zanic, and Jonathon Howard. “Microtubule catastrophe and rescue”. In: *Current Opinion in Cell Biology* 25 (2013), pp. 14–22. DOI: [10.1016/j.ceb.2012.09.006](https://doi.org/10.1016/j.ceb.2012.09.006).
- [53] Melissa K. Gardner et al. “Depolymerizing Kinesins Kip3 and MCAK Shape Cellular Microtubule Architecture by Differential Control of Catastrophe”. In: *Cell* 147 (2011), pp. 1092–1103. DOI: [10.1016/j.cell.2011.10.037](https://doi.org/10.1016/j.cell.2011.10.037).
- [54] Melissa K. Gardner et al. “Rapid Microtubule Self-Assembly Kinetics”. In: *Cell* 146 (2011), pp. 582–592.
- [55] S. Garoff et al. “The effects of substrate roughness on ultrathin water films”. In: *J. Chem. Phys.* 90.12 (1989), pp. 7505–75015. DOI: [10.1063/1.456184](https://doi.org/10.1063/1.456184).
- [56] Walther Gerlach and Otto Stern. “Der experimentelle Nachweis der Richtungsquantelung im Magnetfeld”. In: *Z. Phys.* 9.1 (1922), pp. 349–352.
- [57] Frederick Gittes et al. “Flexural Rigidity of Microtubules and Actin Filaments Measured from Thermal Fluctuations in Shape”. In: *J. Cell Biol.* 120 (1993), pp. 923–934.

- [58] Roy J. Glauber. “Time-dependent statistics of the Ising model”. In: *J. Math. Phys.* 4.2 (1963), pp. 294–307. DOI: [10.1063/1.1703954](https://doi.org/10.1063/1.1703954).
- [59] James P. Gleeson and Rick Durrett. “Temporal profiles of avalanches on networks”. In: *Nat. Com.* 8 (2017), pp. 1–13.
- [60] Yadin Goldschmidt et al. “Nonequilibrium critical behavior in unidirectionally coupled stochastic processes”. In: *Phys. Rev. E* 59.6 (1999), pp. 6381–6408. DOI: [10.1103/PhysRevE.59.6381](https://doi.org/10.1103/PhysRevE.59.6381).
- [61] Anne E. Hafner et al. “Run-and-Pause dynamics of cytoskeletal motor proteins”. In: *Sci. Rep.* 6.37162 (2016), pp. 1–12. DOI: [10.1038/srep37162](https://doi.org/10.1038/srep37162).
- [62] H.C. Hamaker. “The London-Van der Waals attraction between spherical particles”. In: *Physica* 4.10 (1937), pp. 1058–1072. DOI: [10.1016/S0031-8914\(37\)80203-7](https://doi.org/10.1016/S0031-8914(37)80203-7).
- [63] Theodore E. Harris. *The Theory of Branching Processes*. RAND, 1964.
- [64] F. Hauksbee. *Physico-Mechanical Experiments on Various Subjects. Containing an Account of several surprizing Phenomena touching light and electricity, producible on the attrition of bodies with many other remarkable appearances , not before observ’d*. R. Brugis, 1709.
- [65] Terrell L. Hill. “Introductory analysis of the GTP-cap phase change kinetics at the end of a microtubule”. In: *Proc. Natl. Acad. Sci. Unit. States Am.* 81 (1984), pp. 6728–6732.
- [66] H. Hinrichsen and M. Howard. “A model for anomalous directed percolation”. In: *Eur. Phys. J. B* 7 (1999), pp. 635–643. DOI: [10.1007/s100510050656](https://doi.org/10.1007/s100510050656).
- [67] P.C. Hohenberg and B.I. Halperin. “Theory of dynamic critical phenomena”. In: *Rev. Mod. Phys.* 49.3 (1977), pp. 435–479. DOI: [10.1103/RevModPhys.49.435](https://doi.org/10.1103/RevModPhys.49.435).
- [68] Jonathon Howard. *Mechanics of Motor proteins and the cytoskeleton*. Sinauer, 2001. ISBN: 0-87893-334-4.
- [69] M. Howard and J. Cardy. “Fluctuation effects and multiscaling of the reaction-diffusion front for $A + B \rightarrow OE$ ”. In: *J. Phys. A: Math. Gen.* 28.13 (1995), pp. 3599–3621. DOI: [10.1088/0305-4470/28/13/007](https://doi.org/10.1088/0305-4470/28/13/007).

- [70] M. Howard and C. Godrèche. “Persistence in the Voter model: continuum reaction-diffusion approach”. In: *J. Phys. A: Math. Gen.* 31.11 (1998), pp. L209–L215. DOI: [10.1088/0305-4470/31/11/001](https://doi.org/10.1088/0305-4470/31/11/001).
- [71] Martin Howard, Per Fröjdh, and Kent Bækgaard Lauritsen. “Surface critical behavior in systems with nonequilibrium phase transitions”. In: *Phys. Rev. E* 61.1 (2000), pp. 167–183. DOI: [10.1103/PhysRevE.61.167](https://doi.org/10.1103/PhysRevE.61.167).
- [72] Kerson Huang. *Statistical Mechanics*. 2nd ed. John Wiley & Sons, 1963. ISBN: 0-471-81518-7.
- [73] Shingo Ichiki, Jun Sato, and Katsuhiko Nichinari. “Totally Asymmetric simple exclusion process on a periodic lattice with Langmuir kinetics depending on the occupancy of the forward neighboring site”. In: *Eur. Phys. J. B* 89.135 (2016), pp. 1–5. DOI: [10.1140/epjb/e2016-70192-5](https://doi.org/10.1140/epjb/e2016-70192-5).
- [74] J.O. Indekeu. “Line tension near the wetting transition: results from an interface displacement model”. In: *Phys. A: Stat. Mech. its appl.* 183.4 (1992), pp. 439–461. DOI: [10.1016/0378-4371\(92\)90294-Z](https://doi.org/10.1016/0378-4371(92)90294-Z).
- [75] Ernst Ising. “Beitrag zur Theorie des Ferromagnetismus”. In: *Z. Phys.* 31.1 (1925), pp. 253–258. DOI: [10.1007/BF02980577](https://doi.org/10.1007/BF02980577).
- [76] Hans-Karl Janssen. “On a Lagrangian for Classical Field Dynamics and Renormalization Group Calculations of Dynamical Critical Properties”. In: *Z. Physik B* 23.4 (1976), pp. 377–380. DOI: [10.1007/BF01316547](https://doi.org/10.1007/BF01316547).
- [77] Hans-Karl Janssen and Uwe C. Täuber. “The field-theory approach to percolation processes”. In: *Ann. Phys.* 315 (2005), pp. 146–192. DOI: [10.1016/j.aop.2004.09.011](https://doi.org/10.1016/j.aop.2004.09.011).
- [78] N.F. Johnson et al. “New online ecology of adversarial aggregates: ISIS and beyond”. In: *Science* 352.6292 (2016), pp. 1459–1463. DOI: [10.1126/science.aaf0675](https://doi.org/10.1126/science.aaf0675).
- [79] Leo P. Kadanoff. “Scaling Laws for Ising models nears T_c ”. In: *Phys. Physique Fizika* 2.6 (1966), pp. 263–272. DOI: [10.1103/PhysicsPhysiqueFizika.2.263](https://doi.org/10.1103/PhysicsPhysiqueFizika.2.263).
- [80] Kiyoshi Kanazawa et al. “Derivation of the Boltzmann Equation for Financial Brownian Motion: Direct Observation of the Collective Motion of High-Frequency Traders”. In: *Phys. Rev. Lett.* 120.13 (2018), pp. 1–6. DOI: [10.1103/PhysRevLett.120.138301](https://doi.org/10.1103/PhysRevLett.120.138301).

- [81] Eric R. Kandel et al. *Principles of Neuroscience*. 5th ed. McGraw-Hill, 2013.
- [82] Mehran Kardar and Joseph O. Indekeu. “Wetting of fractally rough surfaces”. In: *Phys. Rev. Lett.* 65.5 (1990), p. 662. DOI: [10.1103/PhysRevLett.65.662](https://doi.org/10.1103/PhysRevLett.65.662).
- [83] Kyozi Kawasaki. “Diffusion Constants near the Critical Point for Time-Dependent Ising Models I”. In: *Phys. Rev.* 145.1 (1966), pp. 224–230. DOI: [10.1103/PhysRev.145.224](https://doi.org/10.1103/PhysRev.145.224).
- [84] Jacob W.J. Kerssemakers et al. “Assembly dynamics of microtubules at molecular resolution”. In: *Nature* 442 (2006), pp. 709–712. DOI: [10.1038/nature04928](https://doi.org/10.1038/nature04928).
- [85] John Maynard Keynes. *The General Theory of Employment, Interest and Money*. The Economic Journal, 1936.
- [86] A. Kolmogoroff. *Grundbegriffe der Wahrscheinlichkeitsrechnung*. Chelsea Publishing Company, 1933.
- [87] Jeffrey R. Kuhn and Thomas D. Pollard. “Real-Time Measurements of Actin Filament Polymerization by Total Internal Reflection Fluorescence Microscopy”. In: *Biophys. J.* 88 (2005), pp. 1387–1402.
- [88] Edmund Landau. *Handbuch der Lehre von der Verteilung der Primzahlen. II*. B.G. Teubner, 1909, p. 883.
- [89] L.D. Landau and E.M. Lifshitz. *Statistical Physics*. 3rd ed. Elsevier, 1980. ISBN: 978-0-7506-3372-7.
- [90] Frederick Lanni, D. Lansing Taylor, and B.R. Ware. “fluorescence photobleaching recovery in solutions of labeled actin”. In: *Biophys. J.* 35 (1981), pp. 351–364.
- [91] Pierre Simon Marquis De Laplace. *Traité de Mécanique Céleste. Supplément au dixième livre – Sur l’action capillaire*. Vol. 4. Courcier, 1805.
- [92] Michel Le Bellac. *Quantum and Statistical Field Theory*. Oxford Science Publications, 1991.
- [93] Elliott Lieb, Theodore Schultz, and Daniel Mattis. “Two Soluble Models of an Antiferromagnetic Chain”. In: *Ann. Phys.* 16.3 (1961), pp. 407–466. DOI: [10.1016/0003-4916\(61\)90115-4](https://doi.org/10.1016/0003-4916(61)90115-4).

- [94] Alexandr Malijevský and Andrew O. Parry. “Critical Point Wedge Filling”. In: *Phys. Rev. Lett.* 110.16 (2013), pp. 1–5. DOI: [10.1103/PhysRevLett.110.166101](https://doi.org/10.1103/PhysRevLett.110.166101).
- [95] Alexandr Malijevský and Andrew O. Parry. “Filling transitions in acute and open wedges”. In: *Phys. Rev. E* 91.5 (2015), pp. 1–8. DOI: [10.1103/PhysRevE.91.052401](https://doi.org/10.1103/PhysRevE.91.052401).
- [96] Eckhard Mandelkow, Joseph Thomas, and Carolyn Cohen. “Microtubule structure at low resolution by x-ray diffraction”. In: *Proc. Natl. Acad. Sci.* (1977), pp. 3370–3374. DOI: [10.1073/pnas.74.8.3370](https://doi.org/10.1073/pnas.74.8.3370).
- [97] Pedro D. Manrique et al. “Generalized Gelation Theory Describes Onset of Online Extremist Support”. In: *Phys. Rev. Lett.* 121.4 (2018), pp. 1–5. DOI: [10.1103/PhysRevLett.121.048301](https://doi.org/10.1103/PhysRevLett.121.048301).
- [98] P.C. Martin, E.D. Siggia, and H.A. Rose. “Statistical Dynamics of Classical Systems”. In: *Phys. Rev. A* 8.1 (1973), pp. 423–437. DOI: [10.1103/PhysRevA.8.423](https://doi.org/10.1103/PhysRevA.8.423).
- [99] Warner Marzocchi and Anna Maria Lombardi. “A double branching model for earthquake occurrence”. In: *J. Geophys. Res.* 113 (2008), pp. 1–12.
- [100] Sebastian P. Maurer et al. “EB1 Accelerates Two Conformational Transitions Important for Microtubule Maturation and Dynamics”. In: *Curr. Biol.* 24.4 (2014), pp. 372–384. DOI: [10.1016/j.cub.2013.12.042](https://doi.org/10.1016/j.cub.2013.12.042).
- [101] Koshin Mihashi. “Molecular characteristics of G-ADP actin”. In: *Arch. Biochem. Biophys.* 107 (3 1964), pp. 441–448.
- [102] V.V. Mikhailov. “Normal ordering and generalised Sterling numbers”. In: *J. Phys. A: Math. Gen.* 18 (1985), pp. 231–235.
- [103] Tim Mitchison and Marc Kirschner. “Dynamic Instability of Microtubule Growth”. In: *Nature* 312 (1984), pp. 237–242.
- [104] S. Nekovar and G. Pruessner. “A field-theoretic approach to the Wiener Sausage”. In: *J. Stat. Phys.* 163 (2016), pp. 604–641. DOI: [10.1007/s10955-016-1483-2](https://doi.org/10.1007/s10955-016-1483-2).
- [105] Scott H. Northrup and H.P. Erickson. “Kinetics of protein-protein association explained by Brownian dynamics computer simulations”. In: *Proc. Natl. Acad. Sci.* 89.8 (1992), pp. 3338–3342. DOI: [10.1073/pnas.89.8.3338](https://doi.org/10.1073/pnas.89.8.3338).

- [106] Martin A. Nowak. *Evolutionary Dynamics. Exploring the Equations of Life*. Cambridge, MA, USA: The Belknap Press of Harvard University Press, 2006. ISBN: 0-674-02338-2.
- [107] Martin A. Nowak. “Five Rules for the Evolution of Cooperation”. In: *Science* 314.5805 (2006), pp. 1560–1563. DOI: [10.1126/science.1133755](https://doi.org/10.1126/science.1133755).
- [108] David J. Odde. “Estimation of the Diffusion-Limited Rate of Microtubule Assembly”. In: *Biophys. J.* 73 (1997), pp. 88–95.
- [109] David Odde, Lynne Cassimeris, and Helen M. Buettner. “Kinetic of Microtubule Catastrophe assessed by probabilistic analysis”. In: *Biophys. J.* 69 (1995), pp. 796–802.
- [110] Lars Onsager. “Crystal Statistics. I. A Two-Dimensional Model with an Order-Disorder Transition”. In: *Phys. Rev.* 65.3 and 4 (1944), pp. 117–149. DOI: [10.1103/PhysRev.65.117](https://doi.org/10.1103/PhysRev.65.117).
- [111] A.O. Parry, M.J. Greenall, and J.M. Romero-Enrique. “Fluid Adsorption near an Apex: Covariance between Complete and Critical Wetting”. In: *Phys. Rev. Lett.* 90.4 (2003), pp. 1–4. DOI: [10.1103/PhysRevLett.90.046101](https://doi.org/10.1103/PhysRevLett.90.046101).
- [112] A.O. Parry, C. Rascón, and A.J. Wood. “Critical Effects at 3D Wedge Wetting”. In: *Phys. Rev. Lett.* 85.2 (2000), pp. 345–348. DOI: [10.1103/PhysRevLett.85.345](https://doi.org/10.1103/PhysRevLett.85.345).
- [113] A.O. Parry, C. Rascón, and A.J. Wood. “Universality for 2D Wedge Wetting”. In: *Phys. Rev. Lett.* 83.26 (1999), pp. 5535–5538. DOI: [10.1103/PhysRevLett.83.5535](https://doi.org/10.1103/PhysRevLett.83.5535).
- [114] A.O. Parry et al. “Derivation of non-local interfacial Hamiltonian for short-ranged wetting II: General diagrammatic structure”. In: *J. Phys.: Condens. Matter* 19.41 (2007), pp. 1–21. DOI: [10.1088/0953-8984/19/41/416105](https://doi.org/10.1088/0953-8984/19/41/416105).
- [115] A.O. Parry et al. “Derivation of non-local interfacial Hamiltonian for short-ranged wetting: I. Double-parabola approximation”. In: *J. Phys.: Condens. Matter* 18.28 (2006), pp. 6433–6451. DOI: [10.1088/0953-8984/18/28/001](https://doi.org/10.1088/0953-8984/18/28/001).
- [116] A.O. Parry et al. “Universal Phase Boundary Shifts for Corner Wetting and Filling”. In: *Phys. Rev. Lett.* 87.19 (2001), pp. 1–4. DOI: [10.1103/PhysRevLett.87.196103](https://doi.org/10.1103/PhysRevLett.87.196103).

- [117] Frédéric Paugam. *Towards the Mathematics of Quantum Field Theory*. Springer, 2014. ISBN: 978-3-319-04563-4. DOI: [10.1007/978-3-319-04564-1](https://doi.org/10.1007/978-3-319-04564-1).
- [118] Johannes Pausch and Gunnar Pruessner. “Is Actin Filament and Microtubule Growth Reaction- or Diffusion-Limited?” In: *J. Stat. Mech.: Theory Exp.* 2019.053501 (2019), pp. 1–18. DOI: [10.1088/1742-5468/ab081c](https://doi.org/10.1088/1742-5468/ab081c).
- [119] I. Pázsit and L. Pál. *Neutron Fluctuations: A Treatise on the Physics of Branching Processes*. Elsevier, 2007.
- [120] L. Peliti. “Path integral approach to birth-death processes on a lattice”. In: *J. Phys.-Paris* 46 (1985), pp. 1469–1483.
- [121] Michael E. Peskin and Daniel V. Schroeder. *An Introduction to Quantum Field Theory*. CRC Press, 1995.
- [122] P. Pfeifer et al. “Multilayer Adsorption on a fractally rough surface”. In: *Phys. Rev. Lett.* 62.17 (1989), pp. 1997–2000. DOI: [10.1103/PhysRevLett.62.1997](https://doi.org/10.1103/PhysRevLett.62.1997).
- [123] Thomas D. Pollard. “Polymerization of ADP-Actin”. In: *J. Cell Biol.* 99 (1984), pp. 769–777.
- [124] Thomas D. Pollard and William C. Earnshaw. *Cell Biology*. 2nd ed. Elsevier, 2004.
- [125] Renfrey B. Potts. “The mathematical investigation of some cooperative phenomena”. PhD thesis. Oxford University, 1951.
- [126] Viola Priesemann, Matthias H.J. Munk, and Michael Wibral. “Subsampling effects in neuronal avalanche distributions recorded in vivo”. In: *BMC Neurosci.* 10.40 (2009), pp. 1–20. DOI: [10.1186/1471-2202-10-40](https://doi.org/10.1186/1471-2202-10-40).
- [127] Viola Priesemann et al. “Neuronal Avalanches Differ from Wakefulness to Deep Sleep – Evidence from Intracranial Depth Recordings”. In: *PLoS Comput. Biol.* 9.3 (2013). DOI: [10.1371/journal.pcbi.1002985](https://doi.org/10.1371/journal.pcbi.1002985).
- [128] Gunnar Pruessner. *Lecture Notes on Non-equilibrium Statistical Mechanics*. 2011.
- [129] Miljana Radivojević et al. “On the origins of extractive metallurgy: new evidence from Europe”. In: *J. Archaeol. Sci.* 37.11 (2010), pp. 2775–2787. DOI: [10.1016/j.jas.2010.06.012](https://doi.org/10.1016/j.jas.2010.06.012).
- [130] C. Rascón and A.O. Parry. “Covariance for Cone and Wedge Complete Filling”. In: *Phys. Rev. Lett.* 94.9 (2005). DOI: [10.1103/PhysRevLett.94.096103](https://doi.org/10.1103/PhysRevLett.94.096103).

- [131] C. Rascón and A.O. Parry. “Surface Phase diagrams for wetting on heterogenous substrates”. In: *J. Chem. Phys.* 115.11 (2001), pp. 5258–5271. DOI: [10.1063/1.1380377](https://doi.org/10.1063/1.1380377).
- [132] C. Rascón, A.O. Parry, and A. Sartori. “Wetting at nonplanar substrates: Unbending and unbinding”. In: *Phys. Rev. E* 59.5 (1999), pp. 5697–5700. DOI: [10.1103/PhysRevE.59.5697](https://doi.org/10.1103/PhysRevE.59.5697).
- [133] C. Rascón, J. Pausch, and A.O. Parry. “First-order wedge wetting revisited”. In: *Soft Matter* 14 (2018), pp. 2835–2845.
- [134] Michael Reed and Barry Simon. *Methods of Modern Mathematical Physics Vol. I: Functional Analysis*. Revised and Enlarged Edition. Academic Press, 1980, p. 43.
- [135] Johannes G. Reiter et al. “Crosstalk in concurrent repeated games impedes direct reciprocity and requires stronger levels of forgiveness”. In: *Nat. commun.* 9.555 (2018), pp. 1–8. DOI: [10.1038/s41467-017-02721-8](https://doi.org/10.1038/s41467-017-02721-8).
- [136] K. Rejmer, S. Dietrich, and M. Napiórkowski. “Filling transition for a wedge”. In: *Phys. Rev. E* 60.4 (1999), pp. 4027–4042. DOI: [10.1103/PhysRevE.60.4027](https://doi.org/10.1103/PhysRevE.60.4027).
- [137] L. Rockford et al. “Polymers on Nanoperiodic, heterogeneous surfaces”. In: *Phys. Rev. Lett.* 82.12 (1999). DOI: [10.1103/PhysRevLett.82.2602](https://doi.org/10.1103/PhysRevLett.82.2602).
- [138] Nicholas J. Rose. *Mathematical Maxims and Minims*. Rome Press, 1988.
- [139] J.S. Rowlinson and B. Widom. *Molecular Theory of Capillarity*. Oxford University Press, 1982. ISBN: 0-19-855612-8.
- [140] David Ruelle. *Statistical Mechanics: Rigorous Results*. Addison-Wesley, 1969. ISBN: 0-201-09416-9.
- [141] W.F. Saam. “Wetting, Capillary Condensation and More”. In: *J. Low. Temp. Phys.* 157 (2009), pp. 77–100. DOI: [10.1007/s10909-009-9904-0](https://doi.org/10.1007/s10909-009-9904-0).
- [142] E.D. Salmon et al. “Diffusion Coefficient of Fluorescein-labeled Tubulin in the Cytoplasm of Embryonic Cells of a Sea Urchin: Video Image Analysis of Fluorescence Redistribution after Photobleaching”. In: *J. Cell Biol.* 99 (1984), pp. 2157–2164. DOI: [10.1083/jcb.99.6.2157](https://doi.org/10.1083/jcb.99.6.2157).
- [143] Henry T. Schek et al. “Microtubule Assembly Dynamics at the Nanoscale”. In: *Curr. Biol.* 17.17 (2007), pp. 1445–1455. DOI: [10.1016/j.cub.2007.07.011](https://doi.org/10.1016/j.cub.2007.07.011).

- [144] M.V. Simkin and V.P. Roychowdhury. “Re-inventing Willis”. In: *Phys. Rep.* 502 (2011), pp. 1–35.
- [145] J. G. Skellam. “The Frequency Distribution of the Difference Between Two Poisson Variates Belonging to Different Populations”. In: *J. Roy. Stat. Soc.* 109 (1946), p. 296.
- [146] M. v. Smoluchowski. “Versuch einer mathematischen Theorie der Koagulationskinetik kolloider Lösungen”. In: *Z. Phys. Chem.* 92 (1917), pp. 129–168.
- [147] Frank Spitzer. “Interaction of Markov Processes”. In: *Adv. Math.* 5.2 (1970), pp. 246–290. DOI: [10.1016/0001-8708\(70\)90034-4](https://doi.org/10.1016/0001-8708(70)90034-4).
- [148] H. Eugene Stanley. *Introduction to Phase Transitions and Critical Phenomena*. Oxford Science Publications, 1971.
- [149] D.E. Sullivan. “Van der Waals model of adsorption”. In: *Phys. Rev. B* 20.10 (1979), pp. 3991–4000. DOI: [10.1103/PhysRevB.20.3991](https://doi.org/10.1103/PhysRevB.20.3991).
- [150] Thomas Surrey et al. “Physical Properties Determining Self-Organization of Motors and Microtubules”. In: *Science* 292.5519 (2011), pp. 1167–1171. DOI: [10.1126/science.1059758](https://doi.org/10.1126/science.1059758).
- [151] William Sutherland. “A Dynamical Theory of Diffusion for Non-Electrolytes and the Molecular Mass of Albumin”. In: *Phil. Mag.* 9 (1905), pp. 781–785.
- [152] Jonathan F. Tait and Carl Frieden. “Polymerization and Gelation of Actin Studied by Fluorescence Photobleaching Recovery”. In: *Biochemistry* 21.15 (1982), pp. 3666–3674. DOI: [10.1021/bi00258a022](https://doi.org/10.1021/bi00258a022).
- [153] Leon Armenovich Takhtajan. *Quantum Mechanics for Mathematicians*. American Mathematical Society, 2008.
- [154] Uwe C. Täuber. *Critical Dynamics*. Cambridge University Press, 2014.
- [155] Uwe C. Täuber. “Population oscillations in spatial stochastic Lotka-Volterra models: a field-theoretic perturbational analysis”. In: *J. Phys. A: Math. Theor.* 45.40 (2012), pp. 1–34. DOI: [10.1088/1751-8113/45/40/405002](https://doi.org/10.1088/1751-8113/45/40/405002).
- [156] Uwe C. Täuber, Martin J. Howard, and Haye Hinrichsen. “Multicritical Behavior in Coupled Directed Percolation Processes”. In: *Phys. Rev. Lett.* 80.10 (1998), pp. 2165–2168. DOI: [10.1103/PhysRevLett.80.2165](https://doi.org/10.1103/PhysRevLett.80.2165).

- [157] Philipp E. Tetlock. *Expert Political Judgement*. Princeton University, 2005, p. 87.
- [158] I.M. Tidswell et al. “Complete Wetting of a Rough Surface: An X-Ray Study”. In: *Phys. Rev. Lett.* 66.16 (1991), pp. 2108–2111. DOI: [10.1103/PhysRevLett.66.2108](https://doi.org/10.1103/PhysRevLett.66.2108).
- [159] M. Trau et al. “Microscopic patterning of orientated mesoscopic silica through guided growth”. In: *Nature* 390.6661 (1997), pp. 674–676. DOI: [10.1038/37764](https://doi.org/10.1038/37764).
- [160] G.E. Uhlenbeck and L.S. Ornstein. “On the theory of the brownian motion”. In: *Phys. Rev.* 36.5 (1930), pp. 823–841. DOI: [10.1103/PhysRev.36.823](https://doi.org/10.1103/PhysRev.36.823).
- [161] Vincent Van Buren, David J. Odde, and Lynne Cassimeris. “Estimates of lateral and longitudinal bond energies within the microtubule lattice”. In: *Proc. Natl. Acad. Sci. Unit. States Am.* 99 (2002), pp. 6035–6040.
- [162] N.G. van Kampen. *Stochastic Processes in Physics and Chemistry*. Elsevier, 1992.
- [163] Edmund O. von Lippmann. *Entstehung und Ausbreitung der Alchemie; mit einem Anhang: zur älteren Geschichte der Metalle; ein Beitrag zur Kulturgeschichte*. Springer, 1919, pp. 46–49.
- [164] R.A. Walker et al. “Dynamic Instability of Individual Microtubules Analyzed by Video Microscopy: Rate Constants and Transition Frequencies”. In: *J. Cell Biol.* 107.4 (1988), pp. 1437–1448. DOI: [10.1083/jcb.107.4.1437](https://doi.org/10.1083/jcb.107.4.1437).
- [165] H. W. Watson and Francis Galton. “On the probability of the extinction of families”. In: *J. Anthropol. Inst. G. B. Irel.* 4 (1875), pp. 138–144.
- [166] Michal Wieczorek, Sami Chaaban, and Gary J. Brouhard. “Macromolecular Crowding Pushes Catalyzed Microtubule Growth to Near the Theoretical Limit”. In: *Cell. Mol. Bioeng.* 6 (2013), pp. 383–392.
- [167] Frédéric van Wijland. “Field Theory for reaction-diffusion processes with hard-core particles”. In: *Phys. Rev. E* 63.2 (2001), pp. 1–4. DOI: [10.1103/PhysRevE.63.022101](https://doi.org/10.1103/PhysRevE.63.022101).
- [168] M. M. R. Williams. *Random Processes in Nuclear Reactors*. Elsevier, 2013.
- [169] Kenneth Wilson. “Renormalization Group and Critical Phenomena. I. Renormalization Group and the Kadanoff Scaling Picture”. In: *Phys. Rev. B* 4.9 (1971), pp. 3174–3183. DOI: [10.1103/PhysRevB.4.3174](https://doi.org/10.1103/PhysRevB.4.3174).

- [170] Kenneth Wilson. “Renormalization Group and Critical Phenomena. II. Phase-Space Cell Analysis of Critical Behavior”. In: *Phys. Rev. B* 4.9 (1971), pp. 3184–3205. DOI: [10.1103/PhysRevB.4.3184](https://doi.org/10.1103/PhysRevB.4.3184).
- [171] Younan Xia et al. “Microcontact printing of alkanethiols on copper and its application in microfabrication”. In: *Chem. Mater.* 8.3 (1996), pp. 601–603. DOI: [10.1021/cm950464+](https://doi.org/10.1021/cm950464+).
- [172] P. Yatsyshin, A.O. Parry, and S. Kalliadasis. “Complete Prewetting”. In: *J. Phys.: Condens. Matter* 28.275001 (2016), pp. 1–12. DOI: [10.1088/0953-8984/28/27/275001](https://doi.org/10.1088/0953-8984/28/27/275001).
- [173] Thomas Young. “An essay on the cohesion of fluids”. In: *Phil. Trans. R. Soc. Lond.* 95 (1805), pp. 65–87.
- [174] Jean Zinn-Justin. *Quantum Field Theory and Critical Phenomena*. Oxford Science Publications, 1989.
- [175] Chenghang Zong et al. “Nonequilibrium self-assembly of linear fibers: microscopic treatment of growth, decay, catastrophe and rescue”. In: *Physical Biology* 3 (2006), pp. 83–92. DOI: [10.1088/1478-3975/3/1/009](https://doi.org/10.1088/1478-3975/3/1/009).



DERIVATION OF DOI-PELITI FIELD THEORY

A.1 THE IDENTITY OPERATOR IN TERMS OF COHERENT STATES

Following derivations in [22, 128], the identity operator, $\mathbb{I}$, can be expressed using coherent states as:

$$\mathbb{I} = \sum_{k=0}^{\infty} |k\rangle\langle k| \quad (\text{A.1})$$

$$= \sum_{k=0}^{\infty} \sum_{\ell=0}^{\infty} \delta_{k,\ell} |k\rangle\langle \ell| \quad (\text{A.2})$$

$$= \sum_{k=0}^{\infty} \sum_{\ell=0}^{\infty} \int \frac{e^{-\phi^\dagger \phi}}{\pi} \phi^{\dagger \ell} \frac{\phi^k}{k!} d\text{Re}\phi d\text{Im}\phi |k\rangle\langle \ell| \quad (\text{A.3})$$

$$= \int \frac{e^{-\phi^\dagger \phi}}{\pi} |\phi\rangle\langle \phi^\dagger| d\text{Re}\phi d\text{Im}\phi, \quad (\text{A.4})$$

where the step from Eq. (A.2) to Eq. (A.3) is based on the following expression of the Kronecker- δ :

$$\int \frac{e^{-\phi^\dagger \phi}}{\pi} \phi^{\dagger \ell} \frac{\phi^k}{k!} d\text{Re}\phi d\text{Im}\phi = \int_0^\infty \int_0^{2\pi} \frac{e^{-\rho^2}}{\pi} \frac{\rho^{k+\ell+1} e^{i(k-\ell)\theta}}{k!} d\theta d\rho, \quad (\text{A.5})$$

where ϕ and its complex conjugate $\phi^\dagger$ were expressed in polar coordinates (ρ, θ) in the complex plane. The θ -integral is zero iff $k \neq \ell$. However, when $k = \ell$, the integral equals

$$\int \frac{e^{-\phi^\dagger \phi}}{\pi} \phi^{\dagger \ell} \frac{\phi^k}{k!} d\text{Re}\phi d\text{Im}\phi = \delta_{k,\ell} \int_0^\infty \int_0^{2\pi} \frac{e^{-\rho^2}}{\pi} \frac{\rho^{2k+1}}{k!} d\theta d\rho \quad (\text{A.6})$$

$$= \delta_{k,\ell} \frac{2}{k!} \int_0^\infty e^{-x} x^k dx \quad (\text{A.7})$$

$$= \delta_{k,\ell} \frac{1}{k!} \Gamma(k+1) \quad (\text{A.8})$$

$$= \delta_{k,\ell} \quad (\text{A.9})$$

A.2 FUNCTIONAL DERIVATIVES

Following [46, 117], I use as definition for the derivative of the functional $F[g]$ with argument $g \in \mathcal{S}'$ and variation $h \in \mathcal{S}'$

$$\frac{\delta F}{\delta g}(h) = \lim_{\epsilon \rightarrow 0} \frac{F[g + \epsilon h] - F[g]}{\epsilon}, \quad (\text{A.10})$$

where I assume that F is regularized with a suitable test function from the Schwartz space $\mathcal{S}$ ($\mathcal{S}'$ is its dual space, the space of tempered distributions). The reason for choosing tempered distributions (instead of distributions in $\mathcal{D}'$) is that, its Fourier transforms are well defined.

A.3 DEFINITION OF FOURIER TRANSFORM

The following conventions for Fourier transforms are used for functions in time only

$$\begin{aligned}\phi(\omega) &:= \mathcal{F}[\phi(t)](\omega) = \int_{\mathbb{R}} \phi(t) e^{i\omega t} dt, \\ \mathcal{F}^{-1}[\phi(\omega)](t) &= \int_{\mathbb{R}} \phi(\omega) e^{-i\omega t} d\omega.\end{aligned}\tag{A.11}$$

For functions in time and continuous space $\mathbb{R}^3$, the conventions are

$$\begin{aligned}\varphi(\omega, k) &:= \mathcal{F}[\varphi(t, x)](\omega, k) = \int_{\mathbb{R}^4} \varphi(t, x) e^{i\omega t - ikx} d^3x dt, \\ \mathcal{F}^{-1}[\varphi(\omega, k)](t, x) &= \int_{\mathbb{R}^4} \varphi(\omega, k) e^{-i\omega t + ikx} d^3k d\omega.\end{aligned}\tag{A.12}$$

For functions in time and discrete space $(h\mathbb{Z})^3$, the conventions are

$$\begin{aligned}\psi(\omega, k) &:= \mathcal{F}[\psi_j(t)](\omega, k) = \int_{\mathbb{R}} \sum_{j \in \mathbb{Z}^3} \psi_j(t) e^{i\omega t - ikh j} dt, \\ \mathcal{F}^{-1}[\psi(\omega, k)](\omega, k) &= h^3 \int_{\mathbb{R}} \int_{[0, \frac{2\pi}{h}]^3} \psi(\omega, k) e^{-i\omega t + ikh j} d^3k d\omega,\end{aligned}\tag{A.13}$$

with $d\omega = d\omega/(2\pi)$ and $d^3k = d^3k/(2\pi)^3$. Also used are the shorthands $\delta(\omega - \omega') = 2\pi\delta(\omega - \omega')$, $\delta^3(k - k') = (2\pi)^3\delta^3(k - k')$ and $\delta_c^3(k - k') = \sum_{j \in \mathbb{Z}^3} \delta^3(k - k' + j2\pi/h)$, where $\delta(\cdot)$ is the Dirac δ -function in one dimension and $\delta^3(\cdot)$ in three dimensions. Furthermore, $\Theta(t)$ denotes the Heaviside function:

$$\Theta(t) = \begin{cases} 0 & t < 0 \\ 1 & t \geq 0 \end{cases}\tag{A.14}$$

B

BRANCHING PROCESS

B.1 SECOND MOMENT AND 2-TIME COVARIANCE

In order to calculate the second moment of the particle number, the correlation function $\langle \phi(t_2)\phi(t_1)\tilde{\phi}(0) \rangle$ has to be calculated. It is part of the 2-time covariance and, if $t_1 = t_2$, it is also part of the second moment. The following Feynman diagram represents this term

$$\langle \phi(t_2)\phi(t_1)\tilde{\phi}(0) \rangle \hat{=} \begin{array}{c} t_1 \\ \text{ } \\ t_2 \end{array} \begin{array}{c} \text{ } \\ \diagup \quad \diagdown \\ q_2 \end{array} \text{---} t_0 = 0 \quad (\text{B.1})$$

In the following calculation (for $r > 0$), the variables ω_0 , ω_1 , and ω_2 have been assigned to the legs indicated in the following diagram

$$\langle \phi(\omega_2)\phi(\omega_1)\tilde{\phi}(\omega_0) \rangle \hat{=} \begin{array}{c} \omega_1 \\ \text{ } \\ \omega_2 \end{array} \begin{array}{c} \text{ } \\ \diagup \quad \diagdown \\ \omega_0 \end{array} \quad (\text{B.2})$$

The term is equal to

$$\langle \phi(t_2)\phi(t_1)\tilde{\phi}(0) \rangle = \int_{\mathbb{R}^3} \frac{2q_2 e^{-i\omega_1 t_1 - i\omega_2 t_2} \delta(\omega_1 + \omega_2 + \omega_0)}{(-i\omega_1 + r)(-i\omega_2 + r)(i\omega_0 + r)} d\omega_1 d\omega_2 d\omega_0 \quad (\text{B.3})$$

$$= \int_{\mathbb{R}^2} \frac{2q_2 e^{-i\omega_1 t_1 - i\omega_2 t_2}}{(-i\omega_1 + r)(-i\omega_2 + r)(-i(\omega_1 + \omega_2) + r)} d\omega_1 d\omega_2 \quad (\text{B.4})$$

$$= \Theta(t_2) 2q_2 e^{-rt_2} \int_{\mathbb{R}} \frac{e^{-i\omega_1(t_1-t_2)} - e^{-i\omega_1 t_1}}{(-i\omega_1 + r)(i\omega_1)} d\omega_1. \quad (\text{B.5})$$

Formally, the pole at $\omega_1 = 0$ is irrelevant which can be made apparent by introducing the auxiliary variable t' :

$$\langle \phi(t_2)\phi(t_1)\tilde{\phi}(0) \rangle = \Theta(t_2) 2q_2 e^{-rt_2} \int_{\mathbb{R}} \int_0^{t_2} \frac{e^{-i\omega_1(t_1-t')}}{-i\omega_1 + r} dt' d\omega_1 \quad (\text{B.6})$$

$$= \Theta(t_2) 2q_2 e^{-r(t_1+t_2)} \int_0^{t_2} \Theta(t_1 - t') e^{rt'} dt'. \quad (\text{B.7})$$

While the t' integral goes up to t_2 , the Heaviside function $\Theta(t_1 - t')$ possibly cuts the integral at t_1 if $t_1 < t_2$. Hence, the upper boundary is replaced by $\min\{t_1, t_2\}$:

$$\langle \phi(t_2)\phi(t_1)\tilde{\phi}(0) \rangle = \Theta(t_1)\Theta(t_2) 2q_2 e^{-r(t_1+t_2)} \int_0^{\min\{t_1, t_2\}} e^{rt'} dt' \quad (\text{B.8})$$

$$= \Theta(t_1)\Theta(t_2) 2 \frac{q_2}{r} e^{-r(t_1+t_2)} (e^{r \min\{t_1, t_2\}} - 1) \quad (\text{B.9})$$

Hence, if $t_1 = t_2$, then

$$\langle \phi(t_2)\phi(t_1)\tilde{\phi}(0) \rangle = \Theta(t) 2 \frac{q_2}{r} e^{-rt} (1 - e^{-rt}) \quad (\text{B.10})$$

B.2 THIRD MOMENT AND 3-TIME COVARIANCE

In order to calculate the third moment and the 3-time correlation function, the term $\langle \phi(t_3)\phi(t_2)\phi(t_1)\tilde{\phi}(0) \rangle$ has to be calculated, which splits into two further terms, $I_3(t_3, t_2, t_1)$

and $g_3(t_3, t_2, t_1)$.

$$\left\langle \phi(t_3)\phi(t_2)\phi(t_1)\tilde{\phi}(0) \right\rangle \hat{=} \underbrace{\begin{array}{c} t_1 \\ t_2 \\ t_3 \end{array} \xrightarrow{q_3}}_{\hat{=} I_3} + 2 \underbrace{\begin{array}{c} t_1 \\ t_2 \\ t_3 \end{array} \xrightarrow{q_2}}_{\hat{=} g_3} \quad (\text{B.11})$$

While for I_3 , the placement of t_1 , t_2 and t_3 in the Feynman diagram is symmetric under the permutation, in g_3 the symmetry is different. There are three choices for which time is attached to the leg that first branches off and there are $2 \cdot 2$ choices for the order in the dyadic branches. I call $g'_3(t_3, t_2, t_1)$ the term in g_3 where t_3 is attached to the first leg to branch off (as shown in Eq. (B.11)). The term $I_3(t_3, t_2, t_1)$ can be calculated for $r > 0$:

$$I_3(t_3, t_2, t_1) = \int_{\mathbb{R}^4} \frac{6q_3 e^{-i(\omega_1 t_1 + \omega_2 t_2 + \omega_3 t_3)} \delta(\omega_0 + \omega_1 + \omega_2 + \omega_3)}{(-i\omega_1 + r)(-i\omega_2 + r)(-i\omega_3 + r)(i\omega_0 + r)} \mathrm{d}\omega_1 \mathrm{d}\omega_2 \mathrm{d}\omega_3 \mathrm{d}\omega_0 \quad (\text{B.12})$$

$$= \int_{\mathbb{R}^3} \frac{6q_3 e^{-i(\omega_1 t_1 + \omega_2 t_2 + \omega_3 t_3)} \mathrm{d}\omega_1 \mathrm{d}\omega_2 \mathrm{d}\omega_3}{(-i\omega_1 + r)(-i\omega_2 + r)(-i\omega_3 + r)(-i(\omega_1 + \omega_2 + \omega_3) + r)} \quad (\text{B.13})$$

$$= \Theta(t_3) 6q_3 e^{-rt_3} \int_{\mathbb{R}^2} \frac{e^{-i(\omega_1 t_1 + \omega_2 t_2)} (e^{i(\omega_1 + \omega_2) t_3} - 1) \mathrm{d}\omega_1 \mathrm{d}\omega_2}{(-i\omega_1 + r)(-i\omega_2 + r)(i(\omega_1 + \omega_2))} \quad (\text{B.14})$$

The pole at $\omega_1 + \omega_2 = 0$ is irrelevant, which can be shown by introducing a auxiliary variable t' :

$$I_3(t_3, t_2, t_1) = \Theta(t_3) 6q_3 e^{-rt_3} \int_{\mathbb{R}^2} \int_0^{t_3} \frac{e^{-i(\omega_1 t_1 + \omega_2 t_2)} e^{i(\omega_1 + \omega_2) t'} \mathrm{d}t' \mathrm{d}\omega_1 \mathrm{d}\omega_2}{(-i\omega_1 + r)(-i\omega_2 + r)} \quad (\text{B.15})$$

$$= \Theta(t_3) 6q_3 e^{-rt_3} \int_{\mathbb{R}} \int_0^{t_3} \Theta(t_2 - t') \frac{e^{-r(t_2 - t')} e^{-i\omega_1(t_1 - t')} \mathrm{d}t' \mathrm{d}\omega_1}{(-i\omega_1 + r)} \quad (\text{B.16})$$

$$= \Theta(t_3) 6q_3 e^{-rt_3} \int_0^{t_3} \Theta(t_2 - t') \Theta(t_1 - t') e^{-r(t_2 - t')} e^{-r(t_1 - t')} \mathrm{d}t' \quad (\text{B.17})$$

While the t' integral has as its upper boundary t_3 , the Heaviside functions $\Theta(t_2 - t')$ and $\Theta(t_1 - t')$ reduce the upper boundary to $\min\{t_1, t_2, t_3\}$:

$$I_3(t_3, t_2, t_1) = \Theta(t_3)\Theta(t_2)\Theta(t_1)6q_3e^{-r(t_1+t_2+t_3)} \int_0^{\min\{t_3, t_2, t_1\}} e^{2rt'} dt' \quad (\text{B.18})$$

$$= \Theta(t_3)\Theta(t_2)\Theta(t_1)\frac{3q_3}{r}e^{-r(t_1+t_2+t_3)} (e^{2r\min\{t_3, t_2, t_1\}} - 1) \quad (\text{B.19})$$

So for the 3rd moment, when $t_1 = t_2 = t_3$, I find

$$I_3(t) = \Theta(t)\frac{3q_3}{r}e^{-rt} (1 - e^{-2rt}) \quad (\text{B.20})$$

The term $g'_3(t_3, t_2, t_1)$ can be calculated for $r > 0$ to be equal to

$$g'_3(t_3, t_2, t_1) = \int_{\mathbb{R}^5} \frac{4q_2^2 e^{-i(\omega_1 t_1 + \omega_2 t_2 + \omega_3 t_3)}}{(-i\omega_1 + r)(-i\omega_2 + r)(-i\omega_3 + r)} \quad (\text{B.21})$$

$$\frac{\delta(\omega_1 + \omega_2 - \omega')\delta(\omega' + \omega_3 + \omega_0)\mathfrak{d}\omega_0\mathfrak{d}\omega_1\mathfrak{d}\omega_2\mathfrak{d}\omega_3\mathfrak{d}\omega'}{(-i\omega' + r)(i\omega_0 + r)}$$

$$= \int_{\mathbb{R}^3} \frac{4q_2^2 e^{-i(\omega_1 t_1 + \omega_2 t_2 + \omega_3 t_3)}}{(-i\omega_1 + r)(-i\omega_2 + r)(-i\omega_3 + r)(-i(\omega_1 + \omega_2) + r)} \quad (\text{B.22})$$

$$\frac{\mathfrak{d}\omega_1\mathfrak{d}\omega_2\mathfrak{d}\omega_3}{(-i(\omega_1 + \omega_2 + \omega_3) + r)}$$

$$= \Theta(t_3)4q_2^2 e^{-rt_3} \int_{\mathbb{R}^2} \frac{e^{-i(\omega_1 t_1 + \omega_2 t_2)} (e^{i(\omega_1 + \omega_2)t_3} - 1) \mathfrak{d}\omega_1 \mathfrak{d}\omega_2}{(-i\omega_1 + r)(-i\omega_2 + r)(i(\omega_1 + \omega_2))(-i(\omega_1 + \omega_2) + r)}. \quad (\text{B.23})$$

The pole at $\omega_1 + \omega_2$ is irrelevant, and can be reduced by introducing the auxiliary variable t' :

$$g'_3(t_3, t_2, t_1) = \Theta(t_3)4q_2^2 e^{-rt_3} \int_{\mathbb{R}^2} \int_0^{t_3} \frac{e^{-i(\omega_1 t_1 + \omega_2 t_2)} e^{i(\omega_1 + \omega_2)t'} dt' \mathfrak{d}\omega_1 \mathfrak{d}\omega_2}{(-i\omega_1 + r)(-i\omega_2 + r)(-i(\omega_1 + \omega_2) + r)} \quad (\text{B.24})$$

$$= \Theta(t_3)4q_2^2 e^{-r(t_2+t_3)} \int_{\mathbb{R}} \int_0^{\min\{t_2, t_3\}} \frac{e^{rt' - i\omega_1 t_1} (e^{i\omega_1 t_2} - e^{i\omega_1 t'}) dt' \mathfrak{d}\omega_1}{(-i\omega_1 + r)(i\omega_1)}. \quad (\text{B.25})$$

The pole at $\omega_1 = 0$ is irrelevant and can be reduced by introducing another auxiliary variable t'' :

$$g'_3(t_3, t_2, t_1) = \Theta(t_2)\Theta(t_3)4q_2^2 e^{-r(t_2+t_3)} \int_{\mathbb{R}} \int_0^{\min\{t_1, t_2, t_3\}} \int_{t'}^{t_2} \frac{e^{rt'} e^{i\omega_1(t''-t_1)} dt'' dt' d\omega_1}{(-i\omega_1 + r)} \quad (\text{B.26})$$

$$= \Theta(t_1)\Theta(t_2)\Theta(t_3)4q_2^2 e^{-r(t_1+t_2+t_3)} \int_0^{\min\{t_1, t_2, t_3\}} \int_{t'}^{\min\{t_1, t_2\}} e^{rt'} e^{rt''} dt'' dt' \quad (\text{B.27})$$

$$= \Theta(t_1)\Theta(t_2)\Theta(t_3) \frac{4q_2^2}{r^2} e^{-r(t_1+t_2+t_3)} \left((e^{r \min\{t_1, t_2, t_3\}} - 1) e^{r \min\{t_1, t_2\}} - \frac{1}{2} (e^{2r \min\{t_1, t_2, t_3\}} - 1) \right) \quad (\text{B.28})$$

However, g_3 contains three terms like g'_3 , with the difference that t_3 is replaced by t_1 or t_2 . Hence,

$$g_3(t_3, t_2, t_1) = \Theta(t_1)\Theta(t_2)\Theta(t_3) \frac{4q_2^2}{r^2} e^{-r(t_1+t_2+t_3)} \left((e^{r \min\{t_1, t_2, t_3\}} - 1) (e^{r \min\{t_1, t_2\}} + e^{r \min\{t_1, t_3\}} + e^{r \min\{t_2, t_3\}}) - \frac{3}{2} (e^{2r \min\{t_1, t_2, t_3\}} - 1) \right) \quad (\text{B.29})$$

Thus, for the third moment $t_3 = t_2 = t_1 = t$, $g_3(t)$ is equal to

$$g_3(t) = \Theta(t)3!e^{-rt} \left(\frac{q_2}{r} (1 - e^{-rt}) \right)^2 \quad (\text{B.30})$$

In summary, $\langle \phi^3(t) \tilde{\phi}(0) \rangle$ is equal

$$\langle \phi^3(t) \tilde{\phi}(0) \rangle = \Theta(t) \frac{3q_3}{r} e^{-rt} (1 - e^{-2rt}) + \Theta(t)3!e^{-rt} \left(\frac{q_2}{r} (1 - e^{-rt}) \right)^2 \quad (\text{B.31})$$

B.3 n TH MOMENT APPROXIMATION

The convolution relation is

$$g_n(t) = \sum_{k=1}^{n-1} \binom{n}{k} q_2 \int_0^t e^{-r(t-t')} g_k(t') g_{n-k}(t') dt' \quad (\text{B.32})$$

$$= \sum_{k=1}^{n-1} \binom{n}{k} q_2 \int_0^t e^{-r(t+t')} k!(n-k)! \left(\frac{q_2}{r} (1 - e^{-rt'}) \right)^{n-2} dt'. \quad (\text{B.33})$$

The factors $k!(n-k)!$ cancel with the binomial coefficient such that the $n-1$ summands are independent of k .

$$g_n(t) = n! e^{-rt} q_2 \int_0^t (n-1) e^{-rt'} \left(\frac{q_2}{r} (1 - e^{-rt'}) \right)^{n-2} dt' \quad (\text{B.34})$$

$$= n! e^{-rt} \left(\frac{q_2}{r} (1 - e^{-rt}) \right)^{n-1}. \quad (\text{B.35})$$

Hence, the proof by induction is complete.

B.4 PROBABILITY DISTRIBUTION

In order to derive the probability $P(N(t) = \ell | N(0) = 1)$, I calculate the probability $P(N(t) \leq \ell | N(0) = 1)$ first.

$$P(N(t) \leq \ell | N(0) = 1) = \sum_{k=0}^{\ell} P(N(t) = k | N(0) = 1) \quad (\text{B.36})$$

$$= \sum_{k=0}^{\ell} \frac{d^k}{dz^k} \frac{M_{(N)}(z)}{k!} \Big|_{z=0} \quad (\text{B.37})$$

$$= \sum_{k=0}^{\ell} \frac{d^k}{dz^k} \sum_{m=0}^{\infty} \frac{(z-1)^m}{m!k!} \quad (\text{B.38})$$

$$= \sum_{k=0}^{\ell} \sum_{m=k}^{\infty} \frac{m!(-1)^{m-k}}{m!k!(m-k)!} \langle \phi^m(t) \phi^\dagger(0) \rangle \quad (\text{B.39})$$

$$= \underbrace{1}_{m=0} + \underbrace{0}_{m=1} + \sum_{k=0}^{\ell} \sum_{\substack{m=k \\ m \geq 2}}^{\infty} \frac{(-1)^{m-k}}{k!(m-k)!} \underbrace{m! e^{-rt} \left(\frac{q_2}{r} (1 - e^{-rt}) \right)^{m-1}}_{=g_m(t), \text{ only dyadic diagrams}} \quad (\text{B.40})$$

$$= 1 + \sum_{m \geq 2}^{\infty} (-1)^m e^{-rt} \left(\frac{q_2}{r} (1 - e^{-rt}) \right)^{m-1} \underbrace{\sum_{k=0}^{\ell} \binom{m}{k} (-1)^k}_{\substack{=0 \text{ if } \ell \geq m \\ \neq 0 \text{ if } \ell < m}} \quad (\text{B.41})$$

$$= 1 - e^{-rt} \sum_{m=\ell+1}^{\infty} \left(-\frac{q_2}{r} (1 - e^{-rt}) \right)^{m-1} (-1)^\ell \binom{m-1}{\ell} \quad (\text{B.42})$$

where in line (B.41), I used the convention that the binomial coefficient is zero if the upper number is strictly smaller than the lower number (i.e. choosing more items than there are available is not possible). The sum over k was obtained using *Mathematica*. The equality in line (B.40) is only true in the case of the binary offspring distribution. For other offspring distributions, this has corrections of the order of $\mathcal{O}(r^{-m+2})$. In the following, I continue assuming that the offspring distribution is the binary one and keep using equality signs. The sum over m can also be computed using *Mathematica*,

it gives:

$$P(N(t) \leq \ell | N(0) = 1) = 1 - e^{-rt} (-1)^\ell \left(-\frac{q_2}{r} (1 - e^{-rt}) \right)^\ell \left(1 + \frac{q_2}{r} (1 - e^{-rt}) \right)^{-(\ell+1)} \quad (\text{B.43})$$

$$= 1 - \frac{e^{-rt} \left(\frac{q_2}{r} (1 - e^{-rt}) \right)^\ell}{\left(1 + \frac{q_2}{r} (1 - e^{-rt}) \right)^{\ell+1}}, \quad (\text{B.44})$$

which satisfies its master equation for binary branching and from which follows also:

$$P(N(t) > \ell | N(0) = 1) = \frac{e^{-rt} \left(\frac{q_2}{r} (1 - e^{-rt}) \right)^\ell}{\left(1 + \frac{q_2}{r} (1 - e^{-rt}) \right)^{\ell+1}}. \quad (\text{B.45})$$

For $\ell = 0$, $P(N(t) > 0 | N(0) = 1)$ is the survival probability.

The probability $P(N(t) = \ell | N(0) = 1)$ is found by subtracting $P(N(t) \leq \ell - 1 | N(0) = 1)$ from $P(N(t) \leq \ell | N(0) = 1)$.

B.5 AVALANCHE SHAPE

In order to calculate the avalanche shape, two terms have to be calculated, $I_A(k)$ and $I_B(k)$. First, I show the calculation of $I_A(k)$. I introduce the auxiliary variable u as

$$u = \frac{e^{-r(T-t)} - e^{-rT}}{1 - e^{-r(T-t)}}. \quad (2.74)$$

I will also encounter $1 + u$, which is

$$1 + u = \frac{1 - e^{-rT}}{1 - e^{-r(T-t)}} \quad (\text{B.46})$$

B.5.1 DERIVATION OF $I_A(k)$

$$I_A(k) \hat{=} \sum_{\ell=1}^k \sum_{j_1 \dots j_\ell} \binom{k}{j_1 \dots j_\ell} \left(\begin{array}{c} j_1 \vdots \\ j_\ell \vdots \end{array} \begin{array}{c} \bullet \\ \bullet \end{array} \begin{array}{c} \ell \vdots \\ \bullet \end{array} \right) \quad (2.71)$$

$$= \sum_{\ell=1}^k \sum_{j_1 \dots j_\ell} \binom{k}{j_1 \dots j_\ell} \frac{1}{\ell!} g_{j_1}(T-t) \cdots g_{j_\ell}(T-t) g_{\ell+1}(t), \quad (2.72)$$

where $j_1 + \cdots + j_\ell = k$ and where the factor $1/\ell!$ appears because there are $\ell!$ ways of reordering the $j_1, \dots, j_\ell$. The last equality is exact for binary offspring distributions. It is an approximation for other offspring distributions which becomes exact in the criticality limit $r \rightarrow 0^+$. Next, the definition of $g_n(\cdot)$ is used:

$$I_A(k) = \sum_{\ell=1}^k \sum_{j_1 \dots j_\ell} \binom{k}{j_1 \dots j_\ell} \frac{1}{\ell!} j_1! \cdots j_\ell! e^{-\ell r(T-t)} \left(\frac{q_2}{r} (1 - e^{-r(T-t)}) \right)^{j_1 + \cdots + j_\ell} \quad (B.47)$$

$$\begin{aligned} & (\ell+1)! e^{-rt} \left(\frac{q_2}{r} (1 - e^{-rt}) \right)^\ell \\ &= \sum_{\ell=1}^k \sum_{j_1 \dots j_\ell} k! (\ell+1) (1 - e^{-r(T-t)})^{k-\ell} \\ & \quad e^{-rt} \left(\frac{q_2}{r} \right)^k (e^{-r(T-t)} - e^{-rT})^\ell, \end{aligned} \quad (B.48)$$

where the identity $j_1 + \cdots + j_\ell = k$ was used, and the factors $j_1! \cdots j_\ell!$ were canceled with the multinomial coefficient. Next, I use that the number of compositions of k into ℓ parts is equal to the binomial coefficient of $\ell-1$ out of $k-1$, and I use the definition of u , Eq. (2.74):

$$I_A(k) = k! e^{-rt} \left(\frac{q_2}{r} \right)^k (1 - e^{-r(T-t)})^k \sum_{\ell=1}^k \binom{k-1}{\ell-1} (\ell+1) u^\ell \quad (B.49)$$

$$= k! e^{-rt} \left(\frac{q_2}{r} \right)^k (1 - e^{-r(T-t)})^k \sum_{\ell'=0}^{k-1} \binom{k-1}{\ell'} (\ell'+2) u^{\ell'+1}, \quad (B.50)$$

where the index $\ell = \ell' + 1$ was shifted in the last line. Next, I recognise polynomials in u and use the following identity:

$$\sum_{\ell'=0}^{k-1} \binom{k-1}{\ell'} \ell' u^{\ell'+1} = u^2 \frac{\partial}{\partial u} \sum_{\ell'=0}^{k-1} \binom{k-1}{\ell'} u^{\ell'} = u^2 \frac{\partial}{\partial u} (1+u)^{k-1} \quad (\text{B.51})$$

to obtain

$$I_A(k) = k! e^{-rt} \left(\frac{q_2}{r} \right)^k (1 - e^{-r(T-t)})^k u \left(2 + u \frac{\partial}{\partial u} \right) (1+u)^{k-1} \quad (\text{B.52})$$

$$= k! e^{-rt} \left(\frac{q_2}{r} \right)^k \frac{(1 - e^{-rT})^k}{(1+u)^k} u (2(1+u)^{k-1} + u(k-1)(1+u)^{k-2}) \quad (\text{B.53})$$

$$= k! e^{-rt} \left(\frac{q_2}{r} \right)^k (1 - e^{-rT})^k \left(\frac{u^2(k-1)}{(1+u)^2} + \frac{2u}{1+u} \right), \quad (2.73)$$

where, I used the identity for $1+u$, Eq. (B.46). Eq. (2.73) is the result for $I_A(k)$.

B.5.2 DERIVATION OF $I_B(k)$

The calculation of $I_B(k)$ follows a very similar path to the one for $I_A(k)$:

$$I_B(k) \hat{=} \sum_{\ell=1}^k \sum_{j_1 \dots j_\ell} \binom{k}{j_1 \dots j_\ell} \left(\begin{array}{c} j_1 \vdots \\ j_{\ell-1} \vdots \\ j_\ell \vdots \end{array} \begin{array}{c} \bullet \\ \bullet \\ \bullet \end{array} \begin{array}{c} \bullet \\ \bullet \\ \bullet \end{array} \right) \quad (2.75)$$

$$= \sum_{\ell=1}^k \sum_{j_1 \dots j_\ell} \binom{k}{j_1 \dots j_\ell} \frac{1}{(\ell-1)!} g_{j_1}(T-t) \cdots g_{j_\ell}(T-t) g_\ell(t), \quad (2.76)$$

where the factor $1/(\ell - 1)!$ accounts for the number of permutations of the branches which don't have the disconnected propagator line. Next the definition of $g_n(\cdot)$ is used:

$$I_B(k) = \sum_{\ell=1}^k \sum_{j_1 \dots j_\ell} k! \ell e^{-r\ell(T-t)} \left(\frac{q_2}{r}\right)^{k-1} (1 - e^{-r(T-t)})^{k-\ell} e^{-rt} (1 - e^{-rt})^{\ell-1} \quad (\text{B.54})$$

$$= k! \left(\frac{q_2}{r}\right)^{k-1} \frac{e^{-rt}}{1 - e^{-rt}} (1 - e^{-r(T-t)})^k \sum_{\ell=1}^k \binom{k-1}{\ell-1} \ell u^\ell, \quad (\text{B.55})$$

where I used the definition of u , Eq. (2.74) and the fact that the number of compositions of k into ℓ parts is equal to the binomial coefficient of $\ell - 1$ out of $k - 1$. Next, I shift the index $\ell = \ell' + 1$ and recognise two polynomials in u . Then, I use Eq. (B.51) to obtain

$$I_B(k) = k! \left(\frac{q_2}{r}\right)^{k-1} \frac{e^{-rt}}{1 - e^{-rt}} (1 - e^{-r(T-t)})^k u \sum_{\ell'=0}^k \binom{k-1}{\ell'} (\ell' + 1) u^{\ell'} \quad (\text{B.56})$$

$$= k! \left(\frac{q_2}{r}\right)^{k-1} \frac{e^{-rt}}{1 - e^{-rt}} (1 - e^{-r(T-t)})^k u ((1 + u)^{k-1} + u(k-1)u^{k-2}) \quad (\text{B.57})$$

$$= \frac{k! e^{-rt}}{1 - e^{-rt}} \left(\frac{q_2}{r}\right)^{k-1} (1 - e^{-rT})^k \left(\frac{u^2(k-1)}{(1+u)^2} + \frac{u}{1+u} \right), \quad (2.77)$$

where, I used the identity for $1 + u$, Eq. (B.46). Eq. (2.77) is the result for $I_B(k)$.

B.5.3 COMBINING $I_A(k)$ AND $I_B(k)$ TO OBTAIN $V(t, T)$

The shape of the avalanches $V(t, T)$ requires to calculate $\langle e^{-\phi(T)} \phi^\dagger(t) \phi(t) \tilde{\phi}(0) \rangle$, which splits up into contributions of $I_A(k)$ and $I_B(k)$:

$$\left\langle e^{-\phi(T)} \phi^\dagger(t) \phi(t) \tilde{\phi}(0) \right\rangle = \langle \phi(t) \tilde{\phi}(0) \rangle + \sum_{k=1}^{\infty} \frac{(-1)^k}{k!} (I_A(k) + I_B(k)) \quad (\text{B.58})$$

$$= e^{-rt} + e^{-rt} \sum_{k=1}^{\infty} (-1)^k \left(\frac{q_2}{r}\right)^{k-1} (1 - e^{-rT})^k \left(\left(\frac{q_2}{r} + \frac{1}{1 - e^{-rt}} \right) \frac{u^2(k-1)}{(1+u)^2} + \frac{u}{1+u} \left(\frac{2q_2}{r} + \frac{1}{1 - e^{-rt}} \right) \right), \quad (\text{B.59})$$

where I used Eqs. (2.31), (2.73), and (2.77). In the next step, I assume $q_2 < r$, which is true in the subcritical regime of the binary and geometric offspring distributions, and calculate the geometric series:

$$\left\langle e^{-\phi(T)} \phi^\dagger(t) \phi(t) \tilde{\phi}(0) \right\rangle = \quad (\text{B.60})$$

$$= e^{-rt} - (1 - e^{-rT}) \left(\frac{q_2}{r} + \frac{1}{1 - e^{-rt}} \right) \frac{u^2 e^{-rt} q_2}{(1 + u)^2} \frac{\partial}{\partial q_2} \frac{1}{1 + \frac{q_2}{r} (1 - e^{-rT})} \quad (\text{B.61})$$

$$- e^{-rt} \frac{u}{1 + u} \left(\frac{2q_2}{r} + \frac{1}{1 - e^{-rt}} \right) \frac{(1 - e^{-rT})}{1 + \frac{q_2}{r} (1 - e^{-rT})}$$

$$= e^{-rt} - \frac{q_2}{r} e^{-rt} \left(1 + \frac{q_2}{r} (1 - e^{-rt}) \right) \frac{(e^{-r(T-t)})^2 (1 - e^{-rt})}{\left(1 + \frac{q_2}{r} (1 - e^{-rT}) \right)^2} \quad (\text{B.62})$$

$$- e^{-rt} \left(1 + 2 \frac{q_2}{r} (1 - e^{-rt}) \right) \frac{e^{-r(T-t)}}{1 + \frac{q_2}{r} (1 - e^{-rT})}$$

$$= e^{-rt} - P_s(T) \left(1 + \frac{q_2}{r} (1 - e^{-rt}) \left(2 - \frac{P_s(T)}{P_s(t)} \right) \right), \quad (\text{B.63})$$

where in the second-to-last step, the definition of u , Eq. (2.74) was used, and in the last step I identified the survival probability at times t and T

$$P_s(t) = \frac{e^{-rt}}{1 + \frac{q_2}{r} (1 - e^{-rt})} \quad (2.55)$$

We recall the definition of the shape $V(t, T)$, Eq. (2.67), and calculate the occurring derivative w.r.t. T which results in the sought expression, Eq. (2.78).

B.6 TOTAL SIZE

I am interested in calculating the n th moment of the total size in the approximation of dyadic Feynman diagrams. I denote $\min\{t_1, \dots, t_n\} = t_{\min}$:

$$\mathbb{E}[S^n] = \int_{\mathbb{R}_+^n} s^n \zeta_n(t_1, \dots, t_n) dt_1 \dots dt_n \quad (\text{B.64})$$

$$\begin{aligned}
&= \sum_{m=1}^{n-1} \sum_{\substack{\sigma \subset \{t_1, \dots, t_n\} \\ |\sigma|=m}} s^n q_2 \int_{\mathbb{R}_+^n} \int_0^{t_{\min}} \zeta_m(t_{\sigma(1)} - t', \dots, t_{\sigma(m)} - t') \\
&\quad \cdot \zeta_{n-m}(t_{\sigma^c(m+1)} - t', \dots, t_{\sigma^c(n)} - t') e^{-rt'} dt' dt_1 \dots dt_n
\end{aligned} \tag{B.65}$$

Now, I redefine $t'' = t'/t_{\min}$

$$\mathbb{E}[S^n] = \sum_{m=1}^{n-1} \sum_{\substack{\sigma \subset \{t_1, \dots, t_n\} \\ |\sigma|=m}} s^n q_2 \int_0^1 \int_0^1 \zeta_m(t_{\sigma(1)} - t'' t_{\min}, \dots, t_{\sigma(m)} - t'' t_{\min}). \quad (\text{B.66})$$

$$\cdot \zeta_{n-m}(t_{\sigma^c(m+1)} - t''t_{\min}, \dots, t_{\sigma^c(n)} - t''t_{\min}) e^{-rt''t_{\min}} t_{\min} dt'' dt_1 \dots dt_n \quad (\text{B.67})$$

Next, I define $t'_n = t_n - t''t_{\min}, \dots, t'_1 = t_1 - t''t_{\min}$. The integrands are $dt_n = dt'_n, \dots, dt_1 = dt'_1$, except for the one that is the minimum, for which I get $dt_{\min} = dt'_{\min}/(1 - t'')$.

$$\mathbb{E}[S^n] = \sum_{m=1}^{n-1} \sum_{\substack{\sigma \subset \{t_1, \dots, t_n\} \\ |\sigma|=m}} s^n q_2 \int \int_{\mathbb{R}_+^n} \zeta_m(t'_{\sigma(1)}, \dots, t'_{\sigma(m)}) \quad (\text{B.68})$$

$$\cdot \zeta_{n-m}(t'_{\sigma^c(m+1)}, \dots, t'_{\sigma^c(n)}) \frac{t'_{\min} e^{-\frac{rt''t'_{\min}}{1-t'_{\min}}}}{(1-t'')^2} dt'' dt'_1 \dots dt'_n$$

$$= \sum_{m=1}^{n-1} \sum_{\substack{\sigma \subset \{t_1, \dots, t_n\} \\ |\sigma|=m}} \frac{s^n q_2}{r} \int_{\mathbb{R}_+^n} \zeta_m(t'_{\sigma(1)}, \dots, t'_{\sigma(m)}) \zeta_{n-m}(t'_{\sigma^c(m+1)}, \dots, t'_{\sigma^c(n)}) dt'_1 \dots dt'_n \quad (\text{B.69})$$

where the lower bounds of the integrals remain 0 because the ζ_m s contain Heaviside functions that restrict their arguments to be positive. The remaining integral is the definition of the m th and $n - m$ th moment of the total size which is summed over the possible time subsets of order m .

$$\mathbb{E}[S^n] = \frac{q_2}{r} \sum_{m=1}^{n-1} \binom{n}{m} \mathbb{E}[S^m] \mathbb{E}[S^{n-m}] = \frac{s^n q_2^{n-1}}{r^{2n-1}} 2^{n-1} (2n-3)!! = \frac{s^n q_2^{n-1}}{r^{2n-1}} \frac{(2n-2)!}{(n-1)!} \quad (\text{B.70})$$

which is consistent with the directly calculated results I obtained for $\mathbb{E}[S^2]$ (see previous section) and $\mathbb{E}[S^3]$ (see next section). Interestingly, $(2n-3)!!$ is also the number of leave-labelled, rooted, binary trees. The ratio of the moments is then ($n \geq 1$)

$$\frac{\mathbb{E}[S^{n+1}]}{\mathbb{E}[S^n]} = \frac{s q_2}{r^2} (4n-2) \quad (\text{B.71})$$

In particular, it turns out that certain moment ratios are independent of r and q_2 :

$$\forall k, \ell \geq 1, m \in \{0, \dots, k-1\} :$$

$$\frac{\mathbb{E}[S^k] \mathbb{E}[S^\ell]}{\mathbb{E}[S^{k-m}] \mathbb{E}[S^{\ell+m}]} = \frac{(2k-3)!!(2\ell-3)!!}{(2(k-m)-3)!!(2(\ell+m)-3)!!} \quad (\text{B.72})$$

$$= \frac{(2k-2)!(2\ell-2)!(k-m-1)!(\ell+m-1)!}{(k-1)!(\ell-1)!((2(k-m)-2))!(2(\ell+m)-2)!} \quad (\text{B.73})$$

$$= \frac{\binom{2k-2}{k-1} \binom{2\ell-2}{\ell-1} \binom{k-1}{m}}{\binom{2(k-m)-2}{k-m-1} \binom{2(\ell+m)-2}{\ell+m-1} \binom{\ell+m-1}{m}} \quad (\text{B.74})$$

I proof Eq. (B.70) by induction. I showed above that Eq. (B.70) is correct for $\mathbb{E}[S]$ and $\mathbb{E}[S^2]$. Now, I verify the induction step:

$$\mathbb{E}[S^n] = \frac{q_2}{r} \sum_{m=1}^{n-1} \binom{n}{m} \left(\frac{s^m q_2^{m-1} 2^{m-1} (2m-3)!!}{r^{2m-1}} \right) \left(\frac{s^{n-m} q_2^{n-m-1} 2^{n-m-1} (2(n-m)-3)!!}{r^{2(n-m)-1}} \right) \quad (\text{B.75})$$

$$= \frac{s^n q_2^{n-1}}{r^{2n-1}} 2^{n-2} \sum_{m=1}^{n-1} \binom{n}{m} (2m-3)!! (2(n-m)-3)!! \quad (\text{B.76})$$

Now, I use the following identity which is proved in [21]:

$$\sum_{k=0}^{n-1} \binom{n}{k+1} (2k-1)!! (2(n-k)-3)!! = (2n-1)!! \quad (\text{B.77})$$

I define $m = k + 1$ and subtract the term for $m = n$ from the sum:

$$\sum_{m=1}^{n-1} \binom{n}{m} (2m-3)!! (2(n-m)-1)!! = (2n-1)!! - (2n-3)!! = 2(n-1)(2n-3)!! \quad (\text{B.78})$$

Next, I divide by $(n-1)$ and look at the terms $(2(n-m)-1)!!/(n-1)$:

$$\frac{(2(n-m)-1)!!}{(n-1)} = (2(n-m)-3)!! \left(\frac{2(n-m)-1}{n-1} \right) = (2(n-m)-3)!! \underbrace{\left(1 + \frac{n-2m}{n-1} \right)}_{X(m)} \quad (\text{B.79})$$

In the sum over m , each term appears twice: once at $m = \ell$ and $m = n - \ell$, except if n is divisible by 2. Then, the term $m = n/2$ appears only once. For the terms that appear twice, I find that $X(\ell) = -X(n - \ell)$, hence they cancel. While, if n is divisible by 2, the term $X(n/2) = 0$. Thus I find

$$\sum_{m=1}^{n-1} \binom{n}{m} (2m-3)!! (2(n-m)-3)!! = 2(2n-3)!! \quad (\text{B.80})$$

and the resulting Eq. (B.70).

B.6.1 MOMENT GENERATING FUNCTION AND PROBABILITY DENSITY FUNCTION OF THE TOTAL SIZE

I calculate the moment generating function of the total size distribution as follows

$$\mathcal{M}(z) = \sum_{n=0}^{\infty} \mathbb{E}[S^n] \frac{z^n}{n!} = 1 + \sum_{n=1}^{\infty} \frac{s^n q_2^{n-1}}{r^{2n-1}} \frac{(2n-2)!}{(n-1)!n!} z^n \quad (\text{B.81})$$

$$= 1 + \sum_{n=1}^{\infty} \frac{s^n q_2^{n-1}}{r^{2n-1}} \binom{2(n-1)}{n-1} \frac{z^n}{n} \quad (\text{B.82})$$

$$= 1 + \frac{zs}{r} \sum_{n=0}^{\infty} \left(\frac{sq_2 z}{r^2} \right)^n \underbrace{\binom{2n}{n} \frac{1}{n+1}}_{=C_n \text{ Catalan number}} \quad (\text{B.83})$$

$$= 1 + \frac{zs}{r} \frac{1 - \sqrt{1 - 4 \frac{sq_2 z}{r^2}}}{2 \frac{sq_2 z}{r^2}} \quad (\text{B.84})$$

$$= 1 + \frac{r - \sqrt{r^2 - 4sq_2 z}}{2q_2} \quad (\text{B.85})$$

Given the above moment generating function, can I deduce the corresponding probability density function? Given that the total size is a continuous random variable $S \in [0, \infty)$ with probability density function $f_S(x)$, the moment generating function of S can be written as

$$\mathcal{M}(z) = \int_0^{\infty} f_S(x) e^{xz} dx \quad (\text{B.86})$$

Hence, it can be interpreted as a Laplace transform $\mathcal{L}$ of f_S :

$$\mathcal{M}(-z) = \int_0^{\infty} f_S(x) e^{-xz} dx = \mathcal{L}[f_S](z) \quad (\text{B.87})$$

The probability density function f_S is then given by the inverse Laplace transform (using Mathematica)

$$f_S(x) = \mathcal{L}^{-1}[\mathcal{M}(-z)](x) = \delta(x) \left(1 + \frac{r}{2q_2} \right) + \frac{e^{-x \frac{r^2}{4sq_2}}}{\sqrt{4\pi \frac{q_2}{s} x^3}} \quad (\text{B.88})$$

Surprisingly, Mathematica says that the above probability density function integrates to 1 (for $x \in [0, \infty)$) if the Heaviside Θ function is zero at the origin, i.e. $\Theta(0) = 0$.



FILAMENT SELF-ASSEMBLY

C.1 MASTER EQUATION

The master equation includes six processes:

- 1) diffusion of particles (constant D),
- 2) particle absorption by the tip (constant λ) and subsequent movement of the tip in z direction,
- 3) particle release (rate τ),
- 4) creation of particles (rate γ),
- 5) extinction of particles (rate r), and
- 6) extinction of a filament tip (rate ϵ).

The master equation describes in continuous time and on a discrete spatial lattice $h\mathbb{Z}^3$ how many filament tips (m_j) and how many particles (n_j) are at position $j \in \mathbb{Z}^3$. Let $\{m\}$ denote the entire filament tip occupation configuration in $h\mathbb{Z}^3$, and $\{n\}$ denote the respective particle population. Then, we denote by $\mathcal{P}(\{m\}, \{n\}, t)$ the probability to

find these configurations at time t . Furthermore, we use the shorthand 1_j for occupation of one filament tip / particle at position j . Now, we can write the master equation as follows:

$$\begin{aligned}
\partial_t \mathcal{P}(\{m\}, \{n\}, t) = \sum_{j \in \mathbb{Z}^3} \Big[& \\
1) \quad & D \sum_{|i-j|=1} \left((n_j + 1) \mathcal{P}(\{m\}, \{n + 1_j - 1_i\}, t) - n_j \mathcal{P}(\{m\}, \{n\}, t) \right) \\
2) \quad & + \lambda \left((m_j + 1)(n_j + 1) \mathcal{P}(\{m + 1_j - 1_{j+e_z}\}, \{n + 1_j\}, t) - m_j n_j \mathcal{P}(\{m\}, \{n\}, t) \right) \\
3) \quad & + \tau \left((m_j + 1) \mathcal{P}(\{m_j + 1_j - 1_{j-e_z}\}, \{n - 1_j\}, t) - m_j \mathcal{P}(\{m\}, \{n\}, t) \right) \\
4) \quad & + \gamma \left(\mathcal{P}(\{m\}, \{n - 1_j\}, t) - \mathcal{P}(\{m\}, \{n\}, t) \right) \\
5) \quad & + r \left((n_j + 1) \mathcal{P}(\{m\}, \{n + 1_j\}, t) - n_j \mathcal{P}(\{m\}, \{n\}, t) \right) \\
6) \quad & + \epsilon \left((m_j + 1) \mathcal{P}(\{m + 1_j\}, \{n\}, t) - m_j \mathcal{P}(\{m\}, \{n\}, t) \right) \Big].
\end{aligned} \tag{C.1}$$

C.2 SECOND QUANTIZED MODEL

As outlined by Doi [37], we transform the classical many particle equation into a second quantized version. Therefore, we introduce time-independent occupation states $|\{m\}, \{n\}\rangle$ which represent the particle configuration, i.e. they tell us where we find how many filament tips ($\{m\}$) and particles ($\{n\}$). Furthermore, we introduce ladder operators $a_j, a_j^\dagger$ for filament tips and $b_j, b_j^\dagger$ for particles. Their commutation rules are $[a_j, a_i^\dagger] = [b_j, b_i^\dagger] = \delta_{ij}$. All other commutators are zero. Their action on occupation states is defined as

$$a_j |\{m\}, \{n\}\rangle = m_j |\{m - 1_j\}, \{n\}\rangle, \quad a_j^\dagger |\{m\}, \{n\}\rangle = |\{m + 1_j\}, \{n\}\rangle, \tag{C.2}$$

$$b_j |\{m\}, \{n\}\rangle = n_j |\{m\}, \{n - 1_j\}\rangle, \quad b_j^\dagger |\{m\}, \{n\}\rangle = |\{m\}, \{n + 1_j\}\rangle. \tag{C.3}$$

Now, we define the state of the system as

$$|\phi(t)\rangle = \sum_{\{m\}, \{n\}} \mathcal{P}(\{m\}, \{n\}, t) |\{m\}, \{n\}\rangle. \quad (\text{C.4})$$

The time derivative of $|\phi(t)\rangle$ can be written as

$$\begin{aligned} \partial_t |\phi(t)\rangle = \sum_{j \in \mathbb{Z}^3} \bigg[& D \sum_{|i-j|=1} (b_i^\dagger b_j - b_j^\dagger b_i) \\ & + \lambda (a_{j+e_z}^\dagger a_j b_j - a_j^\dagger a_j b_j^\dagger b_j) + \tau (a_{j-e_z}^\dagger a_j b_j - a_j^\dagger a_j) \\ & + \gamma (b_j^\dagger - 1) + r (b_j - b_j^\dagger b_j) + \epsilon (a_j - a_j^\dagger a_j) \bigg] |\phi(t)\rangle. \end{aligned} \quad (\text{C.5})$$

As described by Peliti [120], the second quantized state equation (C.5) can be transformed into a field theory in path integral formulation. The result is presented in the main text of the article.

C.3 EXPECTED FILAMENT GROWTH LENGTH

For the calculations of expectancy and variances, we use that higher moments of distributions can be obtained by differentiating the moment generating function and setting the respective Fourier space variable to zero.

The zeroth order calculation is

$$\begin{aligned} \langle h j_z \rangle_0 &= i \partial_{k_z} \int_{\mathbb{R}} \langle \psi(\omega, k) \tilde{\psi}(\omega', k') \rangle_0 d\omega \Big|_{k_z=0} \\ &= i \partial_{k_z} \int_{\mathbb{R}} \frac{e^{-i\omega t} d\omega}{-i\omega + \lambda \zeta (1 - e^{-i h k_z}) + \tau (1 - e^{i h k_z}) + \epsilon} \Big|_{k_z=0} \\ &= h (\lambda \zeta - \tau) t e^{-\epsilon t} \Theta(t). \end{aligned} \quad (\text{C.6})$$

If we now let the creation and extinction coefficients tend to zero while keeping their

ratio $\zeta = \gamma/r$ constant, we find

$$\lim_{\epsilon \rightarrow 0} \langle h j_z \rangle_0 = h(\lambda\zeta - \tau)t\Theta(t). \quad (\text{C.7})$$

Hence, the zeroth order average filament growth speed is $\langle v \rangle_0 = \langle h j \rangle_0/t = h(\lambda\zeta - \tau)$.

For the first order correction, we consider the process which is represented by the one-loop Feynman diagram:

$$\begin{aligned} \langle h j_z \rangle_1 &= i \partial_{k_z} \int_{\mathbb{R}^5} \langle \psi(\omega, k) \tilde{\psi}(\omega', k') \rangle_1 d\omega d\omega_b dk_b \Big|_{k_z=0} \\ &= i \partial_{k_z} \int_{\mathbb{R}^5} \frac{\lambda(e^{-ihk_z} - 1)(\tau e^{ihk_z} - \lambda\zeta)e^{-i\omega t}}{(-i\omega + \lambda\zeta(1 - e^{-ihk_z}) + \tau(1 - e^{ihk_z}) + \epsilon)^2(-i\omega_b + Dk_b^2 + r)} \\ &\quad \frac{d^3 k_b d\omega d\omega_b}{(i(\omega_b - \omega) + \lambda\zeta(1 - e^{-ih(k-k_b)_z}) + \tau(1 - e^{ih(k-k_b)_z}) + \epsilon)} \Big|_{k_z=0} \\ &\approx \lambda(\tau - \lambda\zeta)ht \left(\frac{\Lambda}{2\pi^2 D} - \frac{h|\lambda\zeta - \tau|}{8\pi D^2} \right) \Theta(t), \end{aligned} \quad (\text{C.8})$$

where ω_b and k_b are the loop's free frequency and momentum. Here, we approximated $(1 - e^{\pm ih(k_b - k)_z})$ by $\mp ih(k_b - k)_z$, as well as $(1 - e^{\pm ihk_z})$ by $\mp ihk_z$. Furthermore, we let r, ϵ tend to zero and we introduced a cutoff for the k_b integral such that $|k_b| < \Lambda$.

Then, the calculation splits into a steady state part (shown here) and a relaxation part (shown in section C.7), where the latter tends to zero for large times t . What exactly large times mean is analysed and estimated in Section 3.5.1 of the main text and Section C.7. Hence, the average expected (first order) speed is $\langle v \rangle_1 = \langle h j_z \rangle_1/t$.

Both, zeroth and first order correction are graphically represented by Feynman diagrams in Fig. 3.12 of the main text.

C.4 VARIATION OF FILAMENT GROWTH LENGTH

In Section 3.3.1 of the main text, we find that the loop correction is negligible for the filament growth speed. Therefore, we only calculate the variance of the average growth speed using the zeroth order processes, i.e. the first of the two Feynman diagrams in Fig. 3.12. The second moment, i.e. the mean square displacement, of the growth length

is given by

$$\begin{aligned}\langle (hj_z)^2 \rangle_0 &= -\partial_{k_z}^2 \int_{\mathbb{R}} \langle \psi(\omega, k) \tilde{\psi}(\omega', k') \rangle_0 d\omega \Big|_{k_z=0} \\ &= h^2 \Theta(t) e^{-\epsilon t} \left((\lambda\zeta + \tau)t + (\lambda\zeta - \tau)^2 t^2 \right).\end{aligned}\tag{C.9}$$

The variance is equal to

$$\begin{aligned}\text{Var}_0(hj_z) &= \langle (hj_z)^2 \rangle_0 - \langle hj_z \rangle_0^2 \\ &= h^2 \Theta(t) (\lambda\zeta + \tau)t,\end{aligned}\tag{C.10}$$

where we took the limit $\epsilon \rightarrow 0$. Hence, the variance of the average speed $\langle v \rangle_0$ decreases as $1/t$

$$\text{Var}_0(v) = \frac{\text{Var}_0(hj_z)}{t^2} = \frac{h^2(\lambda\zeta + \tau)}{t}.\tag{C.11}$$

C.5 STEADY STATE PARTICLE DEPLETION

We want to find the particle density at time t and position x , given that a filament seed was set at position $j_0 = 0$ at time $t_0 = 0$. In Section 3.3.1, we found that loop corrections are negligible. Therefore, we focus on the zeroth order process:

$$\langle \varphi(t, x) \psi_0^\dagger(0) \rangle_0 = \langle \varphi(t, x) \rangle_0 + \langle \varphi(t, x) \tilde{\psi}_0(0) \rangle_0,\tag{C.12}$$

where $\langle \varphi(t, x) \rangle_0 = \zeta$. Hence, we focus on the second part:

$$\begin{aligned}\langle \varphi(t, x) \tilde{\psi}_0(0) \rangle_0 &= \int_{\mathbb{R}^4} \frac{(\tau - \lambda\zeta) e^{-i\omega t + ikx} d\omega d^3k}{(-i\omega + Dk^2 + r)(-i\omega + \lambda\zeta(1 - e^{-ikh_z}) + \tau(1 - e^{ikh_z}) + \epsilon)} \\ &\approx \int_{\mathbb{R}^3} \frac{(\tau - \lambda\zeta) (e^{-\epsilon t - i h k_z (\lambda\zeta - \tau)t} - e^{-(Dk^2 + r)t}) e^{ikx} d^3k}{Dk^2 - i h k_z (\lambda\zeta - \tau) + r - \epsilon},\end{aligned}\tag{C.13}$$

where we approximated $(1 - e^{\mp i h k_z})$ by $\pm i h k_z$ in the last line.

Now, we define $\tilde{k}_z = k_z - ih(\lambda\zeta - \tau)/(2D)$, while keeping the other directions unchanged $\tilde{k}_x = k_x$, $\tilde{k}_y = k_y$. Then, we let $r, \epsilon \rightarrow 0$. Finally, we change into the moving frame of the filament tip $x_z = \tilde{x}_z + h(\lambda\zeta - \tau)t$, $x_x = \tilde{x}_x$, $x_y = \tilde{x}_y$, and find

$$\langle \varphi(t, x) \tilde{\psi}_0(0) \rangle_0 \approx \int_{\mathbb{R}^3} \frac{(\tau - \lambda\zeta)(1 - e^{-D\tilde{k}^2 t}) e^{i\tilde{k}\tilde{x}} \tilde{d}^3 \tilde{k}}{D\tilde{k}^2 + h^2(\lambda\zeta - \tau)^2/(4D)} e^{-h(\lambda\zeta - \tau)\tilde{x}_z/(2D) - \frac{h^2(\lambda\zeta - \tau)^2}{4D} t}. \quad (\text{C.14})$$

We split this integral into two pieces: the steady state part $\langle \varphi(t, x) \tilde{\psi}_0(0) \rangle_{0,s}$ and the relaxation part $\langle \varphi(t, x) \tilde{\psi}_0(0) \rangle_{0,r}$ of which we treat the former here and the latter in Section C.8. For $t \rightarrow \infty$, the steady state remains constant, while the term in blue of the relaxation part vanishes.

$$\begin{aligned} \langle \varphi(t, x) \tilde{\psi}_0(0) \rangle_{0,s} &\approx \int_{\mathbb{R}^3} \frac{(\tau - \lambda\zeta) e^{i\tilde{k}\tilde{x}} \tilde{d}^3 \tilde{k}}{D\tilde{k}^2 + h^2(\lambda\zeta - \tau)^2/(4D)} e^{-h(\lambda\zeta - \tau)\tilde{x}_z/(2D) - \frac{h^2(\lambda\zeta - \tau)^2}{4D} t} \\ &= (\tau - \lambda\zeta) \frac{e^{-\frac{h|\lambda\zeta - \tau|}{2D} |\tilde{x}|}}{4\pi D |\tilde{x}|} e^{-h(\lambda\zeta - \tau)\tilde{x}_z/(2D)}. \end{aligned} \quad (\text{C.15})$$

We note, that this expression is independent of time! In Section C.8, we will see that the remaining part $\langle \varphi(t, x) \tilde{\psi}_0(0) \rangle_{0,r}$ decays with time.

We can calculate the number of particles in a ball around the origin of radius ρ_0 :

$$\begin{aligned} \mathbb{E}(n_V) &= \int_0^{\rho_0} \int_0^\pi \int_0^{2\pi} \left(\zeta + \frac{\tau - \lambda\zeta}{4\pi D \rho} e^{-\frac{h|\lambda\zeta - \tau|}{2D} (\rho + \rho \cos \theta)} \right) \rho^2 \sin \theta d\phi d\theta d\rho \\ &= \frac{4\pi \zeta \rho_0^3}{3} + \frac{\text{sign}(\tau - \lambda\zeta)}{h} \left(\rho_0 + D \frac{e^{-\frac{h|\lambda\zeta - \tau|}{D} \rho_0} - 1}{h|\lambda\zeta - \tau|} \right). \end{aligned} \quad (\text{C.16})$$

This result is unphysical because the density over which we integrate has a negative part. However, in order to calculate the standard deviation at the origin, we need to consider the mathematically correct but unphysical particle density.

If we assume $h^2|\lambda\zeta - \tau|/(D) \ll 1$, we can approximate the expected particle number to be

$$\mathbb{E}[n_V] \approx \frac{4\pi \zeta \rho_0^3}{3} + \text{sign}(\tau - \lambda\zeta) \frac{|\lambda\zeta - \tau|}{2D} \rho_0^2. \quad (\text{C.17})$$

Then, the smallest positive radius ρ_0 for which the expected number of particles is equal to zero is given by

$$\rho_0 \approx \frac{3|\lambda\zeta - \tau|}{8\pi\zeta D}. \quad (\text{C.18})$$

C.6 STEADY STATE PARTICLE VARIANCE

C.6.1 RELEVANT FEYNMAN DIAGRAM FOR THE COVARIANCE OF THE PARTICLE DENSITY

From the second quantised model (Eq. (C.5)), we derive that the spatial covariance of the density is given by

$$\langle \varphi(t, x) \varphi(t, y) \psi_0^\dagger(0) \rangle_0 + \delta(x - y) \langle \varphi(t, x) \psi_0^\dagger(0) \rangle_0. \quad (\text{C.19})$$

We already calculated the second part in Section C.5, and therefore concentrate on the first. It splits up into

$$\langle \varphi(t, x) \varphi(t, y) \psi_0^\dagger(0) \rangle = \langle \check{\varphi}(t, x) \check{\varphi}(t, y) \tilde{\psi}_0(0) \rangle + \zeta \langle \check{\varphi}(t, x) \tilde{\psi}_0(0) \rangle + \zeta \langle \check{\varphi}(t, y) \tilde{\psi}_0(0) \rangle + \zeta^2. \quad (\text{C.20})$$

Only the first expression, we haven't calculated before. If no loops are considered, only the Feynman diagram shown in Fig. 3.29 of the main article is relevant. It corresponds to the following integral:

$$\begin{aligned} \langle \check{\varphi}(t, x) \check{\varphi}(t, y) \tilde{\psi}_{j_0}(t_0) \rangle_0 &= \frac{1}{2} \int_{\mathbb{R}^8} \frac{(\tau - \lambda\zeta)(\tau e^{ih(k_1+k_2)} - \lambda\zeta)}{(-i\omega_1 + Dk_1^2 + r)(-i\omega_2 + Dk_2^2 + r)} \\ &\cdot \frac{(e^{-i(\omega_1+\omega_2)t+ik_1x+ik_2y} + (x \leftrightarrow y)) \, \mathrm{d}^3k_1 \mathrm{d}^3k_2 \mathrm{d}\omega_1 \mathrm{d}\omega_2}{(-i\omega_2 + (\lambda\zeta - \tau)ihk_2 + \epsilon)(-i(\omega_1 + \omega_2) + (\lambda\zeta - \tau)ih(k_1 + k_2) + \epsilon)}, \end{aligned} \quad (\text{C.21})$$

where approximated in the denominator $(1 - e^{\mp i h k})$ by $\pm i h k$. The symbol $(x \leftrightarrow y)$ signifies that the same integral is repeated only with x and y interchanged. The reason for the symmetrisation is that the process depicted in Fig. 3.29 of the main chapter is not symmetric in the particle fields, however, the covariance is.

Furthermore, we approximate $\tau e^{ih(k_1+k_2)}$ by τ , we define $\tilde{k}_{1z} = k_{1z} - ih(\lambda\zeta - \tau)/(2D)$, as well as $\tilde{k}_{2z} = k_{2z} - ih(\lambda\zeta - \tau)/(2D)$, while keeping $\tilde{k}_{1x} = k_{1x}$, $\tilde{k}_{1y} = k_{1y}$, $\tilde{k}_{2x} = k_{2x}$, and $\tilde{k}_{2y} = k_{2y}$ unchanged. In addition, we change into the moving frame for both x and y by defining $\tilde{x}_z = x_z - (\lambda\zeta - \tau)t$ and $\tilde{y}_z = y_z - (\lambda\zeta - \tau)t$ and keeping $\tilde{x}_x = x_x$, $\tilde{x}_y = x_y$, $\tilde{y}_x = y_x$, and $\tilde{y}_y = y_y$ unchanged. Finally, after integration over ω_1 and ω_2 , we let r and ϵ tend to zero.

Thus, $\langle \check{\varphi}^2(t, x) \tilde{\psi}_{j_0}(t_0) \rangle_0$ can be simplified and approximated by

$$\begin{aligned} \langle \check{\varphi}(t, \tilde{x}) \check{\varphi}(t, \tilde{y}) \tilde{\psi}_{j_0}(t_0) \rangle_0 \approx & \Theta(t) \int_{\mathbb{R}^6} (\tau - \lambda\zeta)^2 \frac{e^{i\tilde{k}_1\tilde{x} + i\tilde{k}_2\tilde{y}} + e^{i\tilde{k}_1\tilde{y} + i\tilde{k}_2\tilde{x}}}{2} e^{-\frac{h(\lambda\zeta - \tau)(\tilde{x} + \tilde{y})}{2D}} \\ & \cdot \left(\frac{\left(1 - e^{-(D\tilde{k}_1^2 + r + \frac{h^2(\lambda\zeta - \tau)}{4D})t}\right)}{(D\tilde{k}_1^2 + ih\tilde{k}_1(\lambda\zeta - \tau) - \frac{h^2(\lambda\zeta - \tau)^2}{4D})(D\tilde{k}_2^2 + \frac{h^2(\lambda\zeta - \tau)^2}{4D})} + \right. \\ & \left. + \frac{\left(1 - e^{-(D\tilde{k}_1^2 + D\tilde{k}_2^2 + \frac{h^2(\lambda\zeta - \tau)^2}{2D})t}\right)}{(D\tilde{k}_1^2 + D\tilde{k}_2^2 + \frac{h^2(\lambda\zeta - \tau)^2}{2D})(D\tilde{k}_2^2 + \frac{h^2(\lambda\zeta - \tau)^2}{4D})} \right) d^3\tilde{k}_1 d^3\tilde{k}_2. \end{aligned} \quad (C.22)$$

The steady state solution is found for $t \rightarrow \infty$, which leads to the convergence of the terms in blue to zero:

$$\begin{aligned} \langle \check{\varphi}(t, \tilde{x}) \check{\varphi}(t, \tilde{y}) \tilde{\psi}_0(0) \rangle_{0,s} \approx & \frac{\Theta(t)(\tau - \lambda\zeta)^2}{(4\pi D|\tilde{x}|)(4\pi D|\tilde{y}|)} \cdot \\ & \cdot \left(\frac{e^{-\frac{h|\lambda\zeta - \tau|}{2D}(|\tilde{y}| + \tilde{y}_z)} + e^{-\frac{h|\lambda\zeta - \tau|}{2D}(|\tilde{x}| + \tilde{x}_z)}}{2} + e^{-\frac{h|\lambda\zeta - \tau|}{D}(|\tilde{x}| + \tilde{x}_z + |\tilde{y}| + \tilde{y}_z)} \right). \end{aligned} \quad (C.23)$$

C.6.2 COVARIANCE OF THE PARTICLE NUMBER

We initialised a filament tip at position $j_0 = 0$ at time $t_0 = 0$ and wait until steady state is reached. Let V_1 and V_2 be compact volumes in $\mathbb{R}^3$ which are moving with the filament tip in steady state. Then, the zeroth order covariance of the number of particles in V_1

and V_2 is equal to

$$\begin{aligned}
\text{cov}_s(n_{V_1}, n_{V_2} | \psi_0^\dagger) &= \mathbb{E}[(n_{V_1} - \mathbb{E}[n_{V_1}])(n_{V_2} - \mathbb{E}[n_{V_2}])] = \mathbb{E}[n_{V_1} n_{V_2}] - \mathbb{E}[n_{V_1}] \mathbb{E}[n_{V_2}] \\
&= \int_{V_1} \int_{V_2} \left(\langle \varphi(\tilde{x}) \varphi(\tilde{y}) \psi_0^\dagger \rangle_{0,s} + \delta(x-y) \langle \varphi(\tilde{x}) \psi_0^\dagger \rangle_{0,s} - \langle \varphi(\tilde{x}) \psi_0^\dagger \rangle_{0,s} \langle \varphi(\tilde{y}) \psi_0^\dagger \rangle_{0,s} \right) d^3 \tilde{x} d^3 \tilde{y} \\
&= \int_{V_1} \int_{V_2} \frac{(\tau - \lambda \zeta)^2}{(4\pi D |\tilde{x}|)(4\pi D |\tilde{y}|)} \frac{1}{2} \left(e^{-\frac{h(\lambda \zeta - \tau)}{2D}(|\tilde{x}| + \tilde{x})} + e^{-\frac{h(\lambda \zeta - \tau)}{2D}(|\tilde{y}| + \tilde{y})} \right) d^3 \tilde{x} d^3 \tilde{y} \\
&\quad + \int_{V_1 \cap V_2} \left(\zeta + (\tau - \lambda \zeta) \frac{e^{-\frac{h(\lambda \zeta - \tau)}{2D}(|\tilde{x}| + \tilde{x})}}{4\pi D |\tilde{x}|} \right) d^3 \tilde{x}.
\end{aligned} \tag{C.24}$$

The variance of the number of particles in a volume V is then found by setting $V_1 = V_2 = V$ in the expression for the covariance. If V is a ball of radius ρ_0 , centered at the origin of the moving frame, the variance can be calculated to be equal to

$$\text{Var}_0(n_V) = \frac{4\pi \zeta \rho_0^3}{3} + \left(\text{sign}(\tau - \lambda \zeta) + \frac{|\tau - \lambda \zeta| \rho_0^2}{2D} \right) \frac{1}{h} \left(\rho_0 + D \frac{e^{-\frac{h|\lambda \zeta - \tau|}{D} \rho_0} - 1}{h|\lambda \zeta - \tau|} \right). \tag{C.25}$$

In the approximation $h^2 |\lambda \zeta - \tau| / D \ll 1$, we can calculate that the variance of the minimal radius ρ_0 , for which the expected number of particles is zero, equals

$$\text{Var}_0(n_V) \approx \frac{|\lambda \zeta - \tau|^2}{4D^2} \rho_0^4. \tag{C.26}$$

C.7 RELAXATION OF THE FILAMENT GROWTH SPEED

Following the discussion in Section C.3, we also would like to calculate the short time behaviour of the filament growth and its relaxation towards the steady state speed. The zeroth order process does not show any relaxation. Therefore, we look at the relaxation

part $\langle hj_z \rangle_{1,r}$ that we dismissed in Eq. (C.8)

$$\begin{aligned} \langle hj_z \rangle_{1,r} = & e^{-\frac{h^2(\lambda\zeta-\tau)^2}{4D}t} \int_{\mathbb{R}} \frac{\lambda(\tau - \lambda\zeta)e^{-D\eta^2 t}(-2iD\eta)\mathfrak{d}\eta}{(D\eta^2 + \frac{h^2(\lambda\zeta-\tau)^2}{4D})^2} \left(\frac{\Lambda}{2\pi^2 D} + \frac{i\eta}{4\pi D} \right) \\ & + \frac{\lambda(\lambda\zeta - \tau)}{4\pi Dh|\lambda\zeta - \tau|}. \end{aligned} \quad (\text{C.27})$$

The integration interval is symmetric around the origin, hence $\langle hj_z \rangle_{1,r}$ can be reduced to its even part and solved using *Mathematica*:

$$\begin{aligned} \frac{\langle hj_z \rangle_{1,r}}{\lambda(\tau - \lambda\zeta)} = & \frac{-\sqrt{t}e^{-\frac{h^2(\lambda\zeta-\tau)^2}{4D}t}}{4(\pi D)^{3/2}} + \frac{\left(1 + \frac{h^2(\lambda\zeta-\tau)^2}{2D}t\right)\text{erfc}\left(\sqrt{\frac{h^2(\lambda\zeta-\tau)^2}{4D}t}\right)}{4\pi Dh|\lambda\zeta - \tau|} \\ & - \frac{1}{4\pi Dh|\lambda\zeta - \tau|}. \end{aligned} \quad (\text{C.28})$$

C.8 RELAXATION OF THE PARTICLE DEPLETION

In Eq. (C.14) of Section C.5, we found that the relaxation part of the particle density in the moving frame of the filament tip equals

$$\langle \varphi(t, x) \tilde{\psi}_0(0) \rangle_{0,r} \approx \int_{\mathbb{R}^3} \frac{(\lambda\zeta - \tau)e^{-D\tilde{k}^2 t} e^{i\tilde{k}\tilde{x}} \mathfrak{d}^3 \tilde{k}}{D\tilde{k}^2 + h^2(\lambda\zeta - \tau)^2/(4D)} e^{-h(\lambda\zeta - \tau)\tilde{x}_z/(2D) - \frac{h^2(\lambda\zeta - \tau)^2}{4D}t}. \quad (\text{C.29})$$

It can be reformulated in a more convenient time integral as follows:

$$\langle \varphi(t, x) \tilde{\psi}_0(0) \rangle_{0,r} = \int_t^\infty \frac{(\lambda\zeta - \tau)e^{-\frac{|\tilde{x}|^2}{4Dt'} - \frac{h^2(\lambda\zeta - \tau)^2}{4D}t'}}{(4\pi Dt')^{\frac{3}{2}}} dt' e^{-h(\lambda\zeta - \tau)(\tilde{x})_z/(2D)}. \quad (\text{C.30})$$

We can test that for $t = 0$, we find the correction to the bulk density ζ when a filament is put into the system

$$\langle \varphi(0, x) \tilde{\psi}_0(0) \rangle_{0,r} = (\lambda\zeta - \tau) \frac{e^{-\frac{h|\lambda\zeta - \tau|}{2D}|\tilde{x}|}}{4\pi D|\tilde{x}|} e^{-h(\lambda\zeta - \tau)\tilde{x}_z/(2D)}. \quad (\text{C.31})$$

$$\langle \psi_{j_2}(t + \Delta t) \tilde{\psi}_{j_1}(t) \psi_{j_1}(t) \tilde{\psi}_0(0) \rangle_0 = \text{~~~~~} \text{~~~~~}$$

Figure C.1: After initialisation of a filament tip (curly blue line), the filament tip is measured twice, but at different times t and $t + \Delta t$. At zeroth order, there is no interaction with particles. Diagrams should be read from right to left.

C.9 CORRELATIONS

A filament tip is set at position $j_0 = 0$ at time $t_0 = 0$. After time t , the position of the filament tip is measured but immediately reinitialised again at the same position it was measured at. Now, after a second small time period Δt , the position of the filament tip is measured again. All the positions on the lattice will be assumed to be on the z -axis, i.e. we write j_1 for j_{1z} . We are interested in the properties of the joint probability function of j_1 and j_2 , which is given by

$$\langle \psi_{j_2}(t + \Delta t) \psi_{j_1}^\dagger(t) \psi_{j_1}(t) \psi_0^\dagger(0) \rangle = \langle \psi_{j_2}(t + \Delta t) \tilde{\psi}_{j_1}(t) \psi_{j_1}(t) \tilde{\psi}_0(0) \rangle. \quad (\text{C.32})$$

We want to quantify how j_1 and j_2 are related in terms of Δt . Therefore, we consider the expected product of the growth lengths $\langle (hj_2)(hj_1) \rangle$. For comparison with the uncorrelated case, we then subtract the product of the expected growth lengths $\langle hj_2 \rangle \langle hj_1 \rangle$.

C.9.1 ZEROth ORDER CORRELATIONS

The Feynman diagram in Fig. C.1 depicts the zeroth order process. It corresponds to the following integral

$$\begin{aligned} \langle \psi_{j_2}(t_2) \tilde{\psi}_{j_1}(t_1) \psi_{j_1}(t_1) \tilde{\psi}_0(0) \rangle_0 &= \\ &= h^2 \int_{[0, \frac{2\pi}{h}]^2} \int_{\mathbb{R}^2} \frac{e^{-i\omega_2 \Delta t - i\omega'_1 t} e^{ik_2 h(j_2 - j_1) + ik'_1 h j_1}}{(-i\omega_2 + \lambda \zeta(1 - e^{-ihk_2}) + \tau(1 - e^{ihk_2}) + \epsilon)} \\ &\quad \cdot \frac{dk'_1 dk_2 d\omega'_1 d\omega_2}{(-i\omega'_1 + \lambda \zeta(1 - e^{-ihk'_1}) + \tau(1 - e^{ihk'_1}) + \epsilon)} \end{aligned} \quad (\text{C.33})$$

which is the joint probability distribution of the filament tip positions j_1 and j_2 .



Figure C.2: After initialisation of the filament at time t_0 , its cap interacts with the particle bulk or releases a particle. Then, the filament length is measured at time t_1 before the free particle is recaptured. After the recapturing, the filament length is measured again at time t_2 . In Fourier space, the four intermediate time evolutions are described by the four frequencies ω_0 (from initiation to first interaction), ω'_1 (from first interaction to first length measurement), ω_1 (from measurement to second interaction), and ω_2 (from second interaction to final length measurement).

We multiply by and integrate over $h(j_2 - j_1)$ and $h(j_1 - j_0)$. Then, the correlation function of the two positions is

$$\begin{aligned} \langle h(j_2 - j_1)h(j_1 - j_0) \rangle_0 &= h^2 \int_{\mathbb{R}^2} \frac{(\lambda\zeta - \tau)^2 e^{-i\omega_2(t_2-t_1) - i\omega'_1(t_1-t_0)} d\omega_2 d\omega'_1}{(-i\omega_2 + \epsilon')^2 (-i\omega'_1 + \epsilon')^2} \\ &= h^2 \delta_{(j'_1, j_0)_{yz}} (\lambda\zeta - \tau)^2 (t_2 - t_1)(t_1 - t_0) e^{-\epsilon'(t_2-t_0)} \Theta(t_2 - t_1) \Theta(t_1 - t_0). \end{aligned} \quad (\text{C.34})$$

In order to detect correlations, we have to subtract the part that appears if they were independent:

$$\langle h(j_2 - j_1)h(j_1 - j_0) \rangle_0 - \langle h(j_2 - j_1) \rangle_T \langle h(j_1 - j_0) \rangle_0 = 0 \quad (\text{C.35})$$

This means that at tree-level, the growth at one point in time and space is independent from the growth at a later point in time anywhere in space. This is expected because the tree level approximations are mean field approximations and hence ignore correlations and local fluctuations.

C.9.2 ONE LOOP CORRECTION

In Fig. C.2, the Feynman diagram is depicted that has to be considered if we are interested in one-loop corrections. This corresponds to

$$\begin{aligned}
& \langle \psi_{j_2}(t_2) \tilde{\psi}_{j_1}(t_1) \psi_{j_1}(t_1) \tilde{\psi}_0(0) \rangle_1 = \\
& = h^4 \int_{[0, \frac{2\pi}{h}]^4} \int_{\mathbb{R}^8} \frac{\lambda(e^{-ihk_2} - 1)(\tau e^{-ihk_0} - \lambda\zeta)}{(-i\omega_2 + \lambda\zeta(1 - e^{-ihk_2}) + \tau(1 - e^{-ihk_2}) + \epsilon')} \\
& \quad \frac{\delta(\omega_2 + \omega_1 - \omega_a) \delta_c(k_2 + k_1 - k_a)}{(i\omega_1 + \lambda\zeta(1 - e^{-ihk_1}) + \tau(1 - e^{-ihk_1}) + \epsilon')} \\
& \quad \frac{\delta(\omega'_1 + \omega_0 + \omega_a) \delta_c(k'_1 + k_0 + k_a)}{(-i\omega'_1 + \lambda\zeta(1 - e^{-ihk'_1}) + \tau(1 - e^{-ihk'_1}) + \epsilon')} \\
& \quad \frac{e^{-i\omega_2 t_2 - i(\omega_1 + \omega'_1)t_1 - i\omega_0 t_0} e^{ik_2 h j_2 + ih(k_1 + k'_1)j_1 + ik_0 h j_0}}{(i\omega_0 + \lambda\zeta(1 - e^{-ihk_0}) + \tau(1 - e^{-ihk_0}) + \epsilon')(-i\omega_a + Dk_a^2 + r)} \\
& \quad d\omega_2 d\omega_1 d\omega'_1 d\omega_0 d\omega_a dk_a^3 dk_2 dk_1 dk'_1 dk_0.
\end{aligned} \tag{C.36}$$

First, we evaluate all the δ functions. Next, we are interested in the expectation of $h(j_2 - j_1)$ and $h(j_1 - j_0)$, hence we derive with respect to k_2 and $-k_0$, and set them to zero:

$$\begin{aligned}
& \langle h(j_2 - j_1) h(j_1 - j_0) \rangle_1 = \\
& = h^2 \int_{\mathbb{R}^3} \int_{\mathbb{R}^3} \frac{\lambda}{(-i\omega_2 + \epsilon')(i(\omega_a - \omega_2) + \lambda\zeta(1 - e^{-ihk_a}) + \tau(1 - e^{-ihk_a}) + \epsilon')} \\
& \quad \frac{1}{(i(\omega_a + \omega_0) + \lambda\zeta(1 - e^{-ihk_a}) + \tau(1 - e^{-ihk_a}) + \epsilon')(i\omega_0 + \epsilon')} \\
& \quad \left(\frac{-(\lambda\zeta - \tau)^2}{i\omega_0 + \epsilon'} + \frac{(\tau - \lambda\zeta)(\lambda\zeta e^{-ihk_a} - \tau e^{-ihk_a})}{i(\omega_a + \omega_0) + \lambda\zeta(1 - e^{-ihk_a}) + \tau(1 - e^{-ihk_a}) + \epsilon'} - \tau \right) \\
& \quad \frac{e^{-i\omega_2(t_2 - t_1) - i\omega_0(t_0 - t_1)}}{-i\omega_a + Dk_a^2 + r} d\omega_2 d\omega_0 d\omega_a dk_a.
\end{aligned} \tag{C.37}$$

Next, we consider the poles of ω_2 , ω_0 and ω_a , and calculate their integral, following the residue theorems. In this calculation, the only viable option is $(t_1 > t_0)$. Hence, the

integral can be written as

$$\begin{aligned}
\langle h(j_2 - j_1)h(j_1 - j_0) \rangle_1 &= \Theta(t_1 - t_0)\Theta(t_2 - t_1) \cdot \\
&\cdot h^2 \int_{\mathbb{R}^3} \lambda e^{-\epsilon'(t_2-t_1)} \left(\frac{1 - e^{-(Dk_a^2+r)(t_2-t_1)-(t_2-t_1)(\lambda\zeta(1-e^{ihk_a})+\tau(1-e^{-ihk_a}))}}{(Dk_a^2 + \lambda\zeta(1 - e^{ihk_a}) + \tau(1 - e^{-ihk_a}) + r)} \right) \\
&e^{-\epsilon'(t_1-t_0)} \left(\frac{(\lambda\zeta - \tau)^2(t_1 - t_0)}{(Dk_a^2 + \lambda\zeta(1 - e^{ihk_a}) + \tau(1 - e^{-ihk_a}) + r)} \right. \\
&+ \frac{(\lambda\zeta - \tau)^2}{(Dk_a^2 + \lambda\zeta(1 - e^{ihk_a}) + \tau(1 - e^{-ihk_a}) + r)^2} \\
&- \frac{(\lambda\zeta - \tau)^2 e^{-(Dk_a^2+r)(t_1-t_0)-(t_1-t_0)(\lambda\zeta(1-e^{ihk_a})+\tau(1-e^{-ihk_a}))}}{(Dk_a^2 + \lambda\zeta(1 - e^{ihk_a}) + \tau(1 - e^{-ihk_a}) + r)^2} \\
&+ \frac{(\tau - \lambda\zeta)(\lambda\zeta e^{ihk_a} - \tau e^{-ihk_a})}{(Dk_a^2 + \lambda\zeta(1 - e^{ihk_a}) + \tau(1 - e^{-ihk_a}) + r)^2} \\
&+ \frac{(\lambda\zeta - \tau)(\lambda\zeta e^{ihk_a} - \tau e^{-ihk_a})(t_1 - t_0) e^{-(Dk_a^2+r)(t_1-t_0)-(t_1-t_0)(\lambda\zeta(1-e^{ihk_a})+\tau(1-e^{-ihk_a}))}}{-Dk_a^2 - \lambda\zeta(1 - e^{ihk_a}) - \tau(1 - e^{-ihk_a}) - r} \\
&+ \frac{(\lambda\zeta - \tau)(\lambda\zeta e^{ihk_a} - \tau e^{-ihk_a}) e^{-(Dk_a^2+r)(t_1-t_0)-(t_1-t_0)(\lambda\zeta(1-e^{ihk_a})+\tau(1-e^{-ihk_a}))}}{(Dk_a^2 + \lambda\zeta(1 - e^{ihk_a}) + \tau(1 - e^{-ihk_a}) + r)^2} \\
&\left. - \tau \frac{1 - e^{-(Dk_a^2+r)(t_1-t_0)-(t_1-t_0)(\lambda\zeta(1-e^{ihk_a})+\tau(1-e^{-ihk_a}))}}{Dk_a^2 + \lambda\zeta(1 - e^{ihk_a}) + \tau(1 - e^{-ihk_a}) + r} \right) \mathrm{d}k_a.
\end{aligned} \tag{C.38}$$

Next, we approximate

$$e^{ihk_a} \approx 1 + i h k_a \quad \text{and} \quad e^{-ihk_a} \approx 1 - i h k_a, \tag{C.39}$$

and we find

$$\langle h(j_2 - j_1)h(j_1 - j_0) \rangle_1 = \Theta(t_1 - t_0)\Theta(t_2 - t_1). \quad (\text{C.40})$$

$$\begin{aligned} & \cdot h^2 \int_{\mathbb{R}^3} \lambda e^{-\epsilon'(t_2-t_1)} \left(\frac{1 - e^{-(Dk_a^2+r)(t_2-t_1)+(t_2-t_1)(\lambda\zeta-\tau)ihk_a}}{(Dk_a^2 - (\lambda\zeta - \tau)ihk_a + r)} \right) \\ & e^{-\epsilon'(t_1-t_0)} \left(\frac{(\lambda\zeta - \tau)^2(t_1 - t_0)}{(Dk_a^2 - (\lambda\zeta - \tau)ihk_a + r)} \right. \\ & + \frac{(\lambda\zeta - \tau)^2(1 - e^{-(Dk_a^2+r)(t_1-t_0)+(t_1-t_0)(\lambda\zeta-\tau)ihk_a})}{(Dk_a^2 - (\lambda\zeta - \tau)ihk_a + r)^2} \\ & + \frac{(\tau - \lambda\zeta)(\lambda\zeta + \tau)ihk_a}{(Dk_a^2 - (\lambda\zeta - \tau)ihk_a + r)^2} \\ & + \frac{(\lambda\zeta - \tau)(\lambda\zeta + \tau)ihk_a(t_1 - t_0)e^{-(Dk_a^2+r)(t_1-t_0)+(t_1-t_0)(\lambda\zeta-\tau)ihk_a}}{-Dk_a^2 + (\lambda\zeta - \tau)ihk_a - r} \\ & + \left. \frac{(\lambda\zeta - \tau)(\lambda\zeta + \tau)ihk_a e^{-(Dk_a^2+r)(t_1-t_0)+(t_1-t_0)(\lambda\zeta-\tau)ihk_a}}{(Dk_a^2 - (\lambda\zeta - \tau)ihk_a + r)^2} \right. \\ & \left. - \tau \frac{1 - e^{-(Dk_a^2+r)(t_1-t_0)+(t_1-t_0)(\lambda\zeta-\tau)ihk_a}}{Dk_a^2 - (\lambda\zeta - \tau)ihk_a + r} \right) \mathrm{d}k_a. \end{aligned} \quad (\text{C.41})$$

We take the limit $r, \epsilon' \rightarrow 0$. and approximate $(t_1 - t_0)$ to be large. Then, the dominating part is:

$$\begin{aligned} & \langle h(j_2 - j_1)h(j_1 - j_0) \rangle_1 \sim \\ & \sim \Theta(t_1 - t_0)\Theta(t_2 - t_1)h^2(\lambda\zeta - \tau)^2(t_1 - t_0) \cdot \\ & \cdot \int_{\mathbb{R}^3} \lambda \left(\frac{1 - e^{-Dk_a^2(t_2-t_1)+(t_2-t_1)(\lambda\zeta-\tau)ihk_a}}{(Dk_a^2 - (\lambda\zeta - \tau)ihk_a)^2} \right) \mathrm{d}k_a. \end{aligned} \quad (\text{C.42})$$

Now, we shift the k_a vector in z direction by $k_{az} = \tilde{k}_{az} + ih(\lambda\zeta - \tau)/(2D)$, and go into spherical coordinates with $\rho = \sqrt{\tilde{k}_a^2}$, this reads

$$\begin{aligned} \langle h(j_2 - j_1)h(j_1 - j_0) \rangle_A &\sim \\ &\sim \frac{1}{\pi} \Theta(t_1 - t_0) \Theta(t_2 - t_1) h^2(\lambda\zeta - \tau)^2 (t_1 - t_0) \cdot \\ &\cdot \int_0^\infty \lambda \rho^2 \left(\frac{1 - e^{-(D\rho^2)(t_2-t_1) - \frac{h^2(\lambda\zeta-\tau)^2}{4D}(t_2-t_1)}}{(D\rho^2 + \frac{h^2(\lambda\zeta-\tau)^2}{4D})^2} \right) d\rho. \end{aligned} \quad (\text{C.43})$$

With Mathematica, we can solve this integral and find:

$$\begin{aligned} \int_0^\infty \rho^2 \frac{1 - e^{-D\rho^2(t_2-t_1) - \frac{h^2(\lambda\zeta-\tau)^2}{4D}(t_2-t_1)}}{(D\rho^2 + \frac{h^2(\lambda\zeta-\tau)^2}{4D})^2} d\rho &= \\ &= \frac{e^{-\frac{h^2(\lambda\zeta-\tau)^2}{4D}(t_2-t_1)} \sqrt{t_2 - t_1}}{4\sqrt{\pi} D^{3/2}} - \frac{h|\lambda\zeta - \tau|(t_2 - t_1)}{8D^2} \\ &+ \frac{(1 + 2\frac{h^2(\lambda\zeta-\tau)^2}{4D}(t_2 - t_1)) \operatorname{erf}\left(\sqrt{\frac{h^2(\lambda\zeta-\tau)^2(t_2-t_1)}{4D}}\right)}{4Dh|\lambda\zeta - \tau|}, \end{aligned} \quad (\text{C.44})$$

which is the sought result.

D

Wetting

In this appendix, details of the calculations from Chapter 4 are explained and important results are derived.

D.1 FREE ENERGY OF GENERAL STRUCTURED SURFACE

Given the profile of a structured surface $z(x, y)$, and the effective interaction potential $W(\ell - z)$ for a liquid layer with liquid/gas boundary $\ell(x, y)$, the free energy

$$F[\ell] = \int \frac{\sigma}{2} (\nabla \ell)^2 + W(\ell - z) dx dy \quad (\text{D.1})$$

has to be minimised in order to find the equilibrium configuration. Its first functional derivative with respect to $\eta := \ell - z$ and variation $h(x, y)$ is

$$\frac{\delta F}{\delta \eta}[\eta, h] = \lim_{\epsilon \rightarrow 0} \frac{F[\eta + \epsilon h] - F[\eta]}{\epsilon} \quad (\text{D.2})$$

$$= \int \sigma ((\eta_x + z_x)h_x + (\eta_y + z_y)h_y) + W'(\eta)h dx dy \quad (\text{D.3})$$

where $\epsilon \in \mathbb{R}$. Integration by parts allows to make the dependence on h simpler:

$$\frac{\delta F}{\delta \eta}[\eta, h] = - \int (\sigma(\eta_{xx} + z_{xx} + \eta_{yy} + z_{yy}) - W'(\eta)) h dx dy \quad (\text{D.4})$$

$$+ \underbrace{[\sigma(\eta_x + z_x + \eta_y + z_y) h]_{\text{boundary}}}_{=0}, \quad (\text{D.5})$$

where the absolute term is zero because the variation h does not vary the boundary conditions.

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Mathematics is a game played according to certain rules with meaningless marks on paper.

David Hilbert [138]

LIST OF MATHEMATICAL SYMBOLS

$:$	such that
$;$	and
$\mathcal{A}$	action
$\delta(\cdot)$	Dirac delta function
$\delta^{(m)}(\cdot)$	m th derivative of $\delta(\cdot)$
$\tilde{\delta}(\cdot)$	$2\pi\delta(\cdot)$
$\tilde{\delta}_c(\cdot)$	$\sum_j \delta(\cdot + 2\pi j/h)$
$\delta_{m,n}$	Kronecker delta
$\mathrm{d}t$	$(2\pi)^{-1}\mathrm{d}t$
d^3k	$(2\pi)^{-3}\mathrm{d}^3k$
$\frac{\delta}{\delta J}$	functional derivative w.r.t. J
$\mathbb{E}[\cdot]$	expectation
$\mathcal{F}$	Fourier operator
η_x, η_y	$\frac{\mathrm{d}}{\mathrm{d}x}\eta, \frac{\mathrm{d}}{\mathrm{d}y}\eta$
ℓ_x, ℓ_y	$\frac{\mathrm{d}}{\mathrm{d}x}\ell, \frac{\mathrm{d}}{\mathrm{d}y}\ell$
$\mathcal{M}$	generating function
$(m)_\ell$	Pochhammer symbol, falling factorial
$m^{(\ell)}$	Pochhammer symbol, rising factorial or ℓ th derivative, depending on context
$\mathbb{N}_0$	non-negative integers
$\mathcal{O}(\cdot)$	Landau big-O notation
P	probability
$\Theta(t)$	Heaviside function
$W(\cdot \cdot)$	probability transition rate
$W(\cdot)$	effective wetting interaction potential
$\mathbb{Z}$	integers

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