Thermodynamic Characterisation of Nonequilibrium Steady States and Study of Phase Transitions

A Thesis submitted for the degree of

Doctor of Philosophy (Science)
in
Physics (Theoretical)

by
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Dedicated to

My parents

whom no words ever invented can ornament

Sukumar Sir

for unfolding before me the beautiful realm of Physics

and,

The teachers

teaching science as the method to not only discover but also establish truth

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Publications

Included in the thesis

- Additivity property and emergence of power laws in nonequilibrium steady states, Arghya Das, Sayani Chatterjee, Punyabrata Pradhan, and Pradip Kumar Mohanty, Phys. Rev. E **92**, 052107 (2015).

- Spatial correlations, additivity and fluctuations in conserved-mass transport processes, Arghya Das, Sayani Chatterjee, and Punyabrata Pradhan, Phys. Rev. E **93**, 062135 (2016).

- Einstein relation and hydrodynamics of nonequilibrium mass transport processes, Arghya Das, Anupam Kundu, and Punyabrata Pradhan, Phys. Rev. E **95**, 062128 (2017).

Other works

- Hydrodynamics, density fluctuations and universality in conserved Manna sandpiles, Sayani Chatterjee, Arghya Das, and Punyabrata Pradhan (submitted).

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1. Introduction : Statistical mechanics for many particle systems

Macroscopic bodies may be acted upon by various external agents, or influences, which could be of thermal, mechanical, or chemical nature. Empirically, equilibrium is defined to be a state where the forces exerted on a system by some external agents keep the matter in the system in a perfect *balance* such that, on average, there is *no* net current of the matter; in equilibrium, fluctuations are time-stationary and averages of observables (bulk properties) are independent of time. Classical thermodynamics deals with equilibrium systems and provides a unified framework to describe time-independent bulk properties of such macroscopic bodies in nature. What is truly remarkable is that, according to thermodynamics, such macroscopic systems, irrespective of the details of microscopic degrees of freedom and interactions, are completely specified by only a finite set of thermodynamic variables. More specifically, in an isolated system in equilibrium, the macroscopic properties are characterised by a thermodynamic variable called entropy, which is expressed as a function of various extensive thermodynamic variables (conserved) such as energy, volume and particle number. Any open system interacting with a much larger system, called environment or bath, through exchange of some of the conserved quantities can be described using an appropriate thermodynamic variable, called free energy (instead of entropy), and can be associated with an *intensive* thermodynamic variable, corresponding to each of the conserved quantities exchanged with the environment or bath. If a system exchanges energy with the bath and remains in equilibrium, the appropriate thermodynamic variable would be 'Helmholtz free energy' and the corresponding intensive thermodynamic variable would be temperature. The second law of thermodynamics implies that entropy (free energy) must be convex (concave) functions of the extensive observables.

In other words, with the knowledge of the above mentioned thermodynamic variables and their functional forms, bulk characterisation of the concerned system is complete. For example, at a given temperature and pressure, the density of water is fixed and can be expressed, in principle, as a function (equation of state) of temperature and pressure. One can, using such characterisation, explain the changes in the state of the system under infinitely slow (quasi-static) changes in the external agents. This specific mode of categorisation is known to be thermodynamic characterisation of matter in equilibrium; in this thesis, we aim to generalise this thermodynamic framework to systems out of equilibrium.

Having its foundation in empirical facts, classical thermodynamics provides a robust understanding of matter in equilibrium. However there are important questions which thermodynamics cannot answer: It does not explain the microscopic origin of entropy, and other thermodynamic potentials. This is because constructing such macroscopic thermodynamic theory does not as such take into account the underlying microscopic discrete constituents of matter and therefore is unable to describe fluctuations, which are inherent in any systems. Clearly, classical thermodynamics cannot prescribe how to calculate thermodynamic potentials such as entropy or free energy, starting from microscopic properties of interacting microscopic constituents of matter. Thus it is apprehensible that thermodynamics must be an emergent feature of laws governing internal 'microscopic' structure and dynamics of matter, that should not only explain, preferably from first principles (i.e., microscopic interactions), the laws of thermodynamics but also account for other bulk phenomena outside its purview. Statistical mechanics provides such a framework, connecting microscopic properties to the macroscopic ones.

1.1. Statistical mechanics in equilibrium

Interaction among the microscopic constituents in a system obligates us to consider matter as a dynamically evolving system, which is traversing its all possible *microscopic* configurations continuously with time. As a consequence, macroscopic observables, directly depending on its microscopic configurations, do fluctuate, though always satisfying the constraints of various conservation laws. Unlike classical thermodynamics, understanding fluctuations is therefore fundamental to the formulation of statistical mechanics. Within such a statistical mechanical formulation, the classical thermodynamic characterisation of equilibrium state of bulk matter can be comprehended readily. Specifically, according to the Boltzmann-Gibbs formulation of statistical mechanics, to understand fluctuation properties the concept of *a-priori* (equal) probabilities of microscopic state is introduced and consequently equilibrium state in a system is stated as the one (maximum probable state) where these probabilities are maximized provided the constraints of the conservation laws. To elaborate further, since the microscopic configurations are ever-changing, the condition for equilibrium necessitates that the corresponding probabilities must be a monotonic (but bounded) function of time, and therefore equilibrium could be understood as the state with maximum probability consistent with the given constraint of conservation laws (in spirit, that is the second law of thermodynamics). What is implied here is that the equilibrium probability for a macroscopic state can be in principle derived from the microscopic Boltzmann-Gibbs probability weight, subject to a suitable choice of ensemble. Consequently, equilibrium fluctuations in extensive variables, e.g., in energy or number of particles, can be characterized by an *intensive* thermodynamic variable like temperature or chemical potential, respectively; this statistical mechanical theory gives us back the classical thermodynamic theory for equilibrium systems.[1] We should remark here that the fluctuations in equilibrium are time-stationary.[2]

The above statistical characterisation, known as equilibrium statistical mechanics (ESM), therefore enables one to quantify the fluctuations in *any* extensive observable and consequently any macroscopic property of matter. However, this equilibrium characterisation will be incomplete without the understanding of a system's response to change in external stimuli, which must describe inherent properties of the concerned equilibrium system. It indeed turns out that, in ESM, there exists a class of general relations between response to a change in the intensive variable (the external stimuli) and the corresponding equilibrium fluctuations in the conserved quantity. These relations, collectively called the fluctuation-response (FR) relations or alternatively the fluctuation-dissipation theorems (FDTs), essentially characterize the fluctuations of any conserved quantity in an equilibrium system. Indeed, the FDTs can be proved using linear-response theory around equilibrium state having the Boltzmann-Gibbs distribution [2].

We specially mention the Einstein relation (ER) [3,4], also known as the Einstein-Smoluchowski

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[1] This description, although necessary, is by no means sufficient. The probabilities and fluctuations must not only be time independent, but at the same time must be very small compared to the average. Otherwise there is no thermodynamics. We shall see later that, for macroscopic bodies a law of large number (LLN) is satisfied that takes care of this condition.

[2] To be consistent, time-independence means symmetry about all operations involving time, i.e. time translation invariance as well as time reversal invariance. As discussed later, for 'stochastic' systems the second criterion for equilibrium is implemented by an explicit statement of microscopic reversibility or detailed balance. (In chapter 7 we discuss stochastic models which violate detailed balance, but gives rise to macroscopic reversibility in the steady state!) However for 'truly' microscopic dynamics described by explicitly time independent interactions, microscopic reversibility is always satisfied; although in such deterministic dynamics the validity of detailed balance is not immediately obvious, but we intuitively expect it to hold since microscopic reversibility eventually leads to equilibrium which itself is also a macroscopic time reversible state.

relation, which is one of the earliest known forms of FDTs . It connects, quite unexpectedly, two seemingly unrelated transport coefficients, bulk-diffusion coefficient $D(\rho)$ and conductivity $\chi(\rho)$, as $D(\rho) = \chi(\rho)/\sigma_{eq}^2(\rho)$ where $\sigma_{eq}^2(\rho) = \lim_{v\to\infty}(\langle n_v^2\rangle_{eq} - \langle n_v\rangle_{eq}^2)/v$ is scaled variance of particle-number n_v in a sub-volume v much smaller than the system volume and ρ is local number density (angular bracket $\langle\cdot\rangle_{eq}$ denoting equilibrium average). Here the diffusion coefficient $D(\rho)$ is defined from Fourier's law for diffusive current $J_D = -D(\rho)\partial\rho/\partial x$ where $\partial\rho/\partial x$ is spatial density gradient in a particular direction, say along $x-$axis. The conductivity $\chi(\rho)$ is defined from Ohm's law for drift current $J_d = \chi(\rho)F/k_BT$, due to a small external biasing force F also along $x-$axis, with k_B and T being the Boltzmann constant and temperature, respectively. For system obeying the detailed balance condition, the ER is universal and is independent of the microscopic details. It turns out that the ER is more generally valid and have very important role in the fluctuating hydrodynamics description.

At this point it is useful to have a qualitative look at equilibrium phenomena in real lives. It is well known that, no real physical system is ever in equilibrium! For example, how do we explain a bowl of water kept on a horizontal table, the whole set up being in a room for some time with temperature of air reading 300K and pressure 1 atm? This question does not have a definite answer. Of course the glass of water looks fairly in equilibrium. There is no force imbalance. We can carefully measure the mass now, and 10 minutes later, and we expect to get same result. If we put a thermometer into the water, it would read 300K. However, if we leave the system by itself and measure the same quantities, say, 3 days later, the results may change significantly. So in a time scale of hours, we have equilibrium description of water, whereas this description fails in a time scale of days. It is more appealing to a hypothetical observer who blinks in a period of say 1 day. Then he or she will see the water in a highly nonequilibrium state: The water vaporises in a few blinks! A human observer will have the same conclusion for rectified spirit left open. Therefore concept of equilibrium is intimately related to the time scale of observation, and it is the property of the material and its interactions that determines the scale. Apart from the example of rectified spirit given above, we may consider the case of the bowl. Although the water vaporised in a matter of days, the bowl will remain intact even after years. Empirically one can conclude that, given a real bulk matter interacting with a certain environment, there exists a time scale in which the properties of matter is described by equilibrium properties, and what we are observing is really a ‘local equilibrium’ or ‘quasi-equilibrium’ state. We conceptually idealise the situation to understand the basic principles and quantify the properties. Results of statistical mechanics that we concern here are for such idealised situations. A relevant question would be whether the conclusions of this idealised theory describes the real properties observed in this ‘local/quasi-equilibrium’ within the desired degree of accuracy? Equilibrium statistical mechanics is a remarkable success in this regard.[3] And we expect that, even when the systems are far from equilibrium, the qualitative understanding gained, the quantitative results obtained, and conclusions reached using the corresponding ideal theory will hold in case of real out of equilibrium situations.

In this context, one may qualitatively summarise the principles on which statistical mechanics works. Equilibrium statistical mechanics, although having a much broad purview, does not really provide the ideal first principle description of matter in equilibrium. This is because, to construct the whole structure of equilibrium statistical mechanics we need the three additional postulates - principle of equal a priori probability, the Boltzmann-Gibbs law, and principle of additivity (closely linked to the Boltzmann-Gibbs law, except for systems with long range interaction). In addition, to facilitate quantitative prediction, we have to assume that the observed results in equilibrium, which are essentially average values over very large microscopic durations, must be given by the average taken over the whole phase space (i.e., the set of all possible configurations of the microscopic degrees of freedom).

This idea of phase space averaging is important. This embodies the concept of ‘ensemble’, which is an infinite collection of independent and identically prepared systems differing from each other through its instantaneous microscopic configurations. Probability distribution, averages and fluctuations are assumed to be calculated over the whole ensemble - which is the usual ‘statistical’ description. However, it is indeed strange that, such a theory does explain, that too with a remarkable accuracy, the observed quantities in real physical systems. For example, while measuring the temperature of the water in the glass, we are

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[3] In fact, for macroscopic objects with number of microscopic degrees of freedom in orders of Avogadro number, ESM predicts results within a precision as small as $\mathcal{O}(10^{-11})$ for bulk quantities. The inaccuracy stemming from surface effects is within $\mathcal{O}(10^{-8})$.

measuring a large but single system in a large but really finite time![4] Secondly, the answer to why the long-time description and ensemble description would be equivalent even in ideal situations is, to our knowledge, not yet agreed upon. In fact, in his famous book on probability and statistics, Feller argued that in Bernoulli trials the statistics of a given infinite sequence and and infinite ensemble of independent trials differ. Consequently the statistics of a single Brownian particle (as a simplest model of a particle moving in a heat bath) over large time will not be mimicked by the statistics of large collection of independent Brownian particles [5]. It appears desirable to develope a theory for the single system at hand. However, as Ma argued, interactions are crucial for the postulates of ESM and the ensemble picture to operationally work as a valid calculational strategy (chapters 5, 12 of [6]; some more discussion in next chapter). And interactions are indeed present in physical systems; even ideal gas molecules must collide.

The thesis is concerned to obtain the idealised theoretical description of nonequilibrium processes, for which we have adopted the usual statistical description of probability and averages. The answer to questions like why this description works, how reversible microscopic dynamics gives rise to irreversible macroscopic behaviour in the first place, etc., constitute the foundation of statistical mechanics which to our knowledge is still unanswered and is an important area of research (a few very intriguing discussions will be found in [6–10]).

1.1.1. Equilibrium phase transitions

Evaporation and melting are ubiquitous natural phenomena. Usually, at a given macroscopic external condition, the properties of a substance is uniquely determined. At a temperature of 300K and in 1 atm pressure, density of water is unique. As we increase the temperature (or reduce the pressure) very slowly such that the substance remains almost in equilibrium throughout the duration of the change, the density smoothly decreases. However, across 373K, the smoothly decreasing density experiences a jump to a much lower

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[4] We measure it over a large time because we measure macroscopic quantities. The time scale of measurement of equilibrium properties depends upon the size of the system. This is related to the equilibration/relaxation time scale. For example, we cannot measure the temperature of a glass of water in, say, a few nanoseconds. The water need to equilibrate with the air, and thermometer, itself a macroscopic body, need to equilibrate with the water. After that we need to wait for some more duration, large enough, so as to allow the instrument to 'register' the data with fluctuations averaged out, ensuring that the observed average value is not changing. Therefore there are really two time scales for equilibrium, lower one for the given equilibrium condition to set in, and the upper and preferably much larger one for the equilibrium to sustain .

value, and the whole of the substance moves from liquid phase to a gaseous phase (at 373K, the phase is not unique), signifying a phase transition. Similar phenomena takes place across 273K when water forms ice. It turns out that, when the substance is not allowed to exchange particles with the environment, instead of this stark phase transition what we observe is coexistence of sharply defined phases with molecules from one phase moving freely to the other phase. For example, in a closed container at room temperature, liquid water and water vapour coexist. In the figure Fig. 1.1, the phase diagram of typical materials and that of water is given.

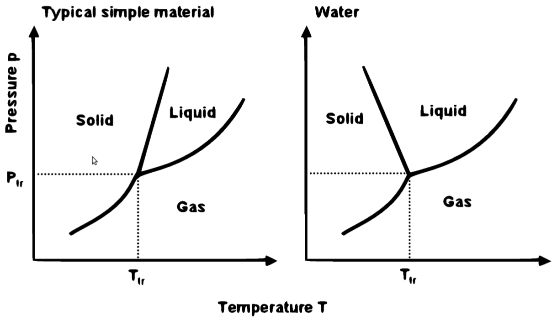

Figure 1.1.: The phase diagram of water and that of typical materials in with pressure and temperature as external tuning parameters. The red lines, known as the phase coexistence curve, signify the first order transition. Note that, the liquid-gas coexistence curve actually ends at a point, known as the critical point. [Image courtesy : Google (ref: C Rose Petruck, Brown University, Jan 99, Chem 201#1)]

Phase transition of the kind mentioned above is known as the 'first order phase transition', which is related to discrete jump in the observable properties (like density) with the change of external conditions like temperature or pressure. In ESM, this kind of phase transition is understood in the thermodynamic limit ($N, V \to \infty$, N/V = finite) through discontinuities in the first derivative of some appropriate free energy, usually Gibbs free energy; whereas the phase coexistence for conserved particle number could be best understood with Helmholtz free energy becoming non-concave in certain values of the external conditions. The regularisation by Maxwell construction then quantifies the phase coexistence and provides the thermodynamic characterisation by ensuring that, these phases can in principle separately exist with the same density at the same values of external control parameters.

Apart from this sharp transitions, there is the other kind of transition, known to be 'continuous phase transition'. Let us take the example of water, as given in the phase diagram in Fig. 1.1. As we increase the pressure, the evaporation to gaseous phase will occur at a higher temperature, and interestingly, the density difference of the coexisting phases will gradually decrease. At a certain temperature and pressure, known as the critical point (approximately 647K and 218 atm for water), the density difference will continuously go to zero, and beyond this point there is no distinction of liquid and gas and a homogeneous 'fluid' phase is attained. Similar phenomena occurs in a magnet; as the temperature is increased, the magnetisation continuously reduced to zero.[5] In the thesis we only concern phase coexistence in far from equilibrium situations.

Our aim is to for possible extension of the lessons from ESM and seek a general framework for characterising nonequilibrium states.

1.2. Statistical mechanics far from equilibrium

1.2.1. Defining features

Equilibrium can be defined in terms of time-translational invariance and time-reversal symmetry, in both microscopic and macroscopic scales, which physically implies the absence of all conserved currents, namely the particle, energy or probability current. Nonequilibrium is defined as a situation where this perfect balance is violated in some way or other. All time-varying states are necessarily nonequilibrium states. So are the states which are time-stationary, but carry mass current (or, more generally, probability current in the configuration space). Unlike in equilibrium, there is no unified principle to characterize fluctuations in nonequilibrium. The added complexity in nonequilibrium is the existence of currents. Characterisation of nonequilibrium system therefore amounts to a characterisation, both static and dynamic, of fluctuation of mass and of currents. Among the vast class of nonequilibrium systems, an important subclass is those having a nonequilibrium steady state. A system in a nonequilibrium steady state, unlike in equilibrium, has a nonzero

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[5] Physically phase transitions are the result of competition between thermodynamic forces. The Helmholtz free energy $F = E - TS$ is minimised by minimising the internal energy at low temperatures, implying that the microscopic configuration will be ordered. However at higher temperatures entropy will start dominating, and eventually the minimum free energy will be obtained by maximising the entropy resulting into disordered microscopic configurations. Similar considerations in presence of mechanical and chemical forces will determine the conditions for phase transitions.

current but its macroscopic properties, like in equilibrium, still remains invariant under time-translation.

1.2.2. Few examples and physical implications

All phenomena we observe in nature are nonequilibrium phenomena in general. One can easily identify blowing wind, flowing stream, moving herds. We can further identify innumerable natural processes like cloud formation, condensation, fragmentation and self-assembly in various substances, traffic flow, flow of charged particles when the light is switched on, transport of sands in the riverbed, particles (like pollen) moving in a stochastic environment, formation of various spatially and temporally varying patterns, all forms of life, economy, and so on, each involving numerous constituent parts whose interactions inherently carry the sense of nonequilibrium. In fact, systems apparently in equilibrium loses this description in a certain time scale. The questions of characterising the macroscopic transient properties of many particle system obeying deterministic Hamiltonian dynamics, explanation of the emergence of thermodynamics and the onset of phase coexistence etc. are, to the best of our knowledge, important open questions in physics, which by construction fall into the realm of nonequilibrium statistical mechanics.

Apart form transient features obeyed by relaxing systems, the physical origin of nonequilibrium phenomena could be viewed as a competition between drive and dissipation. The drive might be external, for example a bulk drive (like gravity) associated to each constituent, or some potential gradient imposed through the boundaries; the drive might be internal as well, as for active (living) systems converting chemical energy to mechanical motion through internal mechanism. All these results into currents, inhomogeneity etc. leading to widely varying bulk properties of systems far away from equilibrium: power laws in the macroscopic distributions is a generic feature; several systems show generic long-range spatial and temporal correlations; bulk properties depend on details of microscopic parameters as well as on the details of interaction with the environment, and remarkably, these violate ensemble equivalence where conserved and nonconserved versions of the same system exhibits strikingly different macroscopic properties and is sensitive to the interaction with bath, thus implying the absence of state functions and equations of state [11–14]. Even when some bulk property (say, pressure P) could be identified as a state functions for certain models, the equations of state $P(\rho)$ is qualitatively very different (for example in active brownian spheres [15]). More exotic events include pattern formation and self organised criticality, where the microscopic dynamics drive the system to macroscopic spatial and temporal features - for example power law correlation - resembling critical behaviour near continuous phase transition, but without external tuning of parameters like temperature. Then there are nonequilibrium phase transitions, which include usual ones like condensation transition, and as well as novel phase transitions unique to nonequilibrium processes. The first kind, although resembles equilibrium phase coexistence, at times have very different statistical mechanical behaviour; for example, even if an intensive thermodynamic parameter for individual phases could be identified, that generically cannot describe coexistence phenomena since it does not offer a Maxwell construction [12, 15, 16] (a feature reminiscent to ensemble inequivalence). The second category include continuous transitions, like the widely studied and debated absorbing phase transition(APT) [17], where below the critical density, the system is trapped into a state (actually one of several such possible states) with no dynamics at all. Dynamic phase transition concern nonanalytic change in the properties of macroscopic current fluctuations.

Despite these wide and rich variety of features which defy ESM-like general universal characterisation, it is rather astonishing that, a large class of nonequilibrium systems possess bulk properties that actually allow a remarkably unified understanding. It will be explored in later chapters of the thesis.

1.2.3. Stochastic process

Characterisation of this innumerable physical phenomena and an understanding of the bulk properties poses a formidable challenge. Although Hamiltonian systems defy much progress, there is considerable development in understanding stochastic processes. In a mesoscopic scale, stochasticity could be regarded an emergent feature of the deterministic dynamical laws, and for physical systems the stochastic description is what matters most of the time. While studying the motion of pollens in a fluid, a grain is the unit constituent and one need not consider either the complicated and mostly unknown underlying structure and interactions within each grain, or the details of each microscopic collisions of the fluid molecule with the pollen particle for characterising its overall motion. This is true for most of the systems of interest. In the thesis, we study processes which are stochastic in nature.

A stochastic processes is defined by the transition probabilities $T(C,t|C',t';C'',t'';...;C_0,0)$ ($t > t' > t'' \cdots$) between different configurations $\{C\}$ and supplemented by their initial distribution $P(C,0)$. The basic question we are interested in is: given a stochastic process, what is the probability $P(C,t)$ of finding some particular configuration 'C' in a particular time? There are additional questions like whether there is a steady state having time independent probability distribution, what are the properties of average values of relevant observables (which are functions of some configuration) and their fluctuations, etc. To answer these questions are in general very difficult. However, for Markov processes, which are one of the simplest and so far most relevant class of stochastic processes for modelling physical systems, there is a well developed formalism [18].

Markov processes are stochastic processes that obey the Markov property, i.e., the transition probability (or rate, when the process is continuous in time) from one configuration to other does not depend on the path by which the current configuration is achieved. In other words,

$$T(C,t|C',t';C'',t'';...;C_0,0) = T(C,t|C',t' < t) \equiv T(C' \to C,t).$$

The consequence is the 'master equation' that governs the probability of finding a given state in an instant,

$$\frac{dP(C,t)}{dt} = \sum_{C' \neq C} [P(C',t)T(C' \to C,t) - P(C,t)T(C \to C',t)]. \tag{1.1}$$

Given the transition rates and the initial condition, the solution of this equation in principle gives the complete microscopic description of the process concerned, and thereby one can quantify various observables. Of particular interest is the processes having jump rates independent of time, $T(C \to C',t) = T(C \to C')$.

In the thesis we consider mass transport processes, a class of Markov processes, that constitute a broad paradigm in modelling ubiquitous physical phenomena. We proceed to characterise simple interacting models defined on a lattice, carrying a locally conserved discrete or continuous variable (called mass). The interaction is local and is governed by stochastic rules of transfer of mass from one lattice site to other through chipping, diffusion and coalescence. It turns out that such simple models give rise to rich nonequilibrium properties (some of which outlined in earlier subsection) and one expects that a unified characterisation of such simple model systems will lead to a better understanding of more general nonequilibrium phenomena.

1.2.4. Nonequilibrium steady states (NESS)

The master equation, which is a system of linear equations written in the form of an eigenvalue problem, $\frac{d|P\rangle}{dt} = \mathcal{T}|P\rangle$, $\mathcal{T}$ being the stochastic matrix with vanishing column sum having an eigenstate $|P_0\rangle$ corresponding to the *largest* eigenvalue 0, the general time dependent solution of the master equation is the linear combination of all such eigenstates. In general, the transient contributions from all non-zero eigenvalues vanish with time, and the system eventually settles into a steady state with configurations occuring with probabilities, $P_{\text{ss}}(C) = \lim_{t\to\infty} P(C,t) = P_0(C)$.

We further note that, since total probability is conserved, the master equation could be perceived as a continuity equation in the configuration space. The steady state could be interpreted as the condition when the total probability flux across an arbitrary state C vanishes, i.e., $\sum_{C'\neq C}[P_0(C')T(C'\to C) - P_0(C)T(C\to C')] \equiv \sum_{C'\neq C} J_{CC'} = 0$. There is a very special situation satisfying the above stationarity criteria, where every element $J_{CC'}$ of the sum individually vanishes, that is, the incoming and outgoing probability flux within each and every pair of states balance each other. This is the ‘detailed balance’ condition, and the corresponding special steady state is known as equilibrium. Note that, this condition for equilibrium could be stated as,

$$\frac{P_0(C')}{P_0(C)} = \frac{T(C\to C')}{T(C'\to C)} \equiv e^{-[E(C')-E(C)]},$$

which is reminiscent of the Gibbs-Boltzmann law of equilibrium.

Given a Markov process, there is another way to check whether the steady state is equilibrium or not, without solving for $P_0(C)$. Given an arbitrary loop of configurations ($C_1 \rightarrow C_2 \rightarrow \cdots \rightarrow C_n \rightarrow C_1$), the necessary and sufficient condition for detailed balance to be satisfied is,

$$\frac{T(C_1 \rightarrow C_2)T(C_2 \rightarrow C_3)\cdots T(C_n \rightarrow C_1)}{T(C_n \rightarrow C_{n-1})T(C_{n-1} \rightarrow C_{n-2})\cdots T(C_2 \rightarrow C_1)} = 1. \tag{1.2}$$

Known as the Kolmogorov criterion [19], this an explicit statement of microscopic reversibility of 'paths' in configuration space.

While detailed balance provides a constraint that is sufficient to characterise the equilibrium states, there is no such universal property in more general nonequilibrium steady states (NESS). Calculation of static correlations, quantitative characterisation of macroscopic observables in steady states, and a complete microscopic description of NESS for physical systems are important open issues.

1.2.5. Dynamical properties

The situation is far more challenging and less understood when the problem involves time varying quantities. The understanding of transient properties in deterministic as well as stochastic systems is required for a holistic understanding of the behaviour of matter. From a macroscopic viewpoint, it is very important to understand currents (which are time dependent quantities by construction) since these define a system to be out of equilibrium, and therefore constitute inherent far from equilibrium property. The understanding of averages, fluctuations, correlations, and eventually complete distribution of currents within and out of steady states involve detailed understanding of time dependent properties of the models concerned. The phenomena of dynamic phase transitions that involve abrupt change in the nature of macroscopic current fluctuation is important in characterising various phases of nonequilibrium processes [20].

Dynamic characterisation is important in another perspective. As discussed in section 1.1, the applicability of statistical mechanics in describing macroscopic features lies with understanding the dynamical nature (for example dynamic correlations) of the system under consideration. On other hand, knowledge of relaxation to the steady state, which involves the second largest eigenvalue of the stochastic matrix T, is crucial for having a stationary description at all. Systems relaxing infinitely slowly reveal exotic properties like ageing, where averages depend upon the time scale of observation [21], and a local stationary description is non-existent. Thus the detailed understanding of dynamic properties of systems is the broad goal of nonequilibrium statistical mechanics.

1.3. Aim : towards a unified macroscopic description

Albeit mainly in a model specific manner, there have been extensive studies to understand the bulk properties of mass transport processes during last few decades. Despite the efforts, leading to very significant developments, the overall progress towards a broad understanding is still largely limited. Correlations in a limited category of systems allowing factorised or matrix-product steady states could be calculated [22–25]. We have results on spatio-temporal structure for simple model systems [26]. Mass distributions have been calculated mainly at a single site level, that too mostly in mean field [27–31], or otherwise for cases where the joint distribution factorises [32–35]. The results are important in understanding the features of mass distributions and scaling relations [30] various exponents obey in steady states. Within statistical mechanics, several models for self-organised criticality have been proposed [36–38]. Exotic phase transitions, including in one dimension, have been hypothesised and explained for certain models. We have models for condensation phenomena [29, 39–41]. A huge literature is there for the APTs. The common thing is, all these have defied unification, even within their sub-domains.

On a more general footing, near equilibrium fluctuation and relaxation properties [2, 42–44] have been generalised to nonequilibrium fluctuation theorems for Hamiltonian systems [9, 45] and is an active area of current research. Further generalisation took place with the discovery of the full fluctuating hydrodynamics for certain class of stochastic systems [46, 47]. In the static case, a fluctuation response relation is discovered for systems obeying an approximate but quite general factorisation scheme [48–50].

For systems in or close to equilibrium, there is a well-defined thermodynamic formulation, which helps us to universally characterize various systems irrespective of details of microscopic interactions among constituent particles or with the reservoir. However, in spite of the important progress outlined above, no such unified understanding is still there for the huge variety of nonequilibrium phenomena. Thus a systematic understanding of macroscopic properties of systems in or out of equilibrium is very much desirable and fundamentally important for constructing a suitable framework from general statistical mechanics considerations.

1.4. Challenges

We have seen, for systems far away from equilibrium, there are wide spectra of systems exhibiting widely varying features. In this myriad of new information and insights, challenging new questions are coming by in this rapidly developing arena. However, a crucial necessity towards building up a general statistical framework is to discover a method for characterising this pile of (some model specific and some more general) results based upon a unifying underlying physical principle, at least for a broad class of systems.

Nonequilibrium steady states (NESSs), which are the closest counterpart of equilibrium and which one expects to be relatively simpler to understand, is the automatic choice to be looked upon. Indeed an important problem in nonequilibrium statistical mechanics is the characterisation of systems having nonequilibrium steady states. Even then, it remains a great challenge to construct a universal statistical mechanics framework for NESSs in general. For example, as briefed in earlier subsection, characterisation of steady states could be accomplished so far for systems having simple product measure, or within mean field; correlations could be calculated either assuming a priori forms of the microscopic measures, or with particular parameter values. Regarding condensation phenomena, a certain characterisation for simple model systems have been put forth in [49]; however their general thermodynamic characterisation far from equilibrium is debated [16], with the centre of confusion lying in understanding steady states in contact (amounting to discovering a counterpart of zeroth law for NESS; there are some important recent developments, in [12, 51, 52]). Therefore, where we are yet very far from a general understanding of detailed microscopic distributions, the aim of characterising macroscopic features look even distant. And then, a general characterisation of dynamical properties seem hopeless.

Despite all these, the situation certainly improved over last couple of decades. Important progress took place in the lines of an additivity property in describing static features, whereas a macroscopic fluctuating theory emerged as a promising candidate to categorise macroscopic spatio-temporal fluctuations for a broad class of systems. With these, a silhouette of a more unified structure has been emerging. However, the identification of the unifying physical property and concrete construction of the framework based on it are the challenging open questions we would like to address here.

1.5. What we have broadly done in the thesis

In this thesis, we propose a unified statistical mechanics framework that we believe can explain a large variety of nonequilibrium static and dynamic properties of *macroscopic* observables in a broad class of stochastic mass transport processes. The basic principle is outlined in the next chapter, and further elaborated in chapters 3 and 6; the applications to characterise concrete far from equilibrium systems are the contents in chapters 4, 5, and 7.

First we focussed on the question whether driven systems having a NESS can have an equilibrium-like thermodynamic structure and can be characterized through fluctuation relations analogous to the fluctuation dissipation theorems (FDT) in equilibrium. To this end, we have explicitly studied a broad class of one dimensional conserved-mass transport processes, which have become a paradigm in nonequilibrium statistical physics. The models we studied are defined with generic parameter values having non-zero spatial correlations. We demonstrated that an equilibrium-like additivity property forwarded in [48–50] can lead one to characterise the subsystem distribution from the knowledge on an intensive quantity, namely an equilibriumlike chemical potential, thus enabling a thermodynamic characterisation for these nonequilibrium systems (detailed in Chapters 3 to 5). We provide an explanation, based on an additivity property, for the scaling forms and power laws widely observed throughout the literature of nonequilibrium phenomena [14].

Moreover, we have exactly calculated two-point spatial correlations for another conserved-mass (continuous) chipping processes in the steady state and demonstrated that, even in the absence of the knowledge of detailed microscopic probability weights for such systems, which have nontrivial spatial correlations, macroscopic distributions could still be determined using additivity [14].

Characterisation of phase transition in interacting many-particle systems is an important problem in statistical physics. In this work [14], we have addressed the condensation phenomena in various mass aggregation models having a NESS [29]. We found that such kind of transitions are consequence of multi-pole singularity in the variance as function of density. Using additivity property, one identifies an equilibriumlike free energy function and a resulting Maxwell construction readily explains the well-known critical power law behaviour of the steady-state mass distribution and phase coexistence (Chapter 4).

Furthermore, the important problem of obtaining hydrodynamic evolutions in interacting many-particle systems on large spatial and temporal scales is addressed at the end. In this thesis, we have derived a fluctuating hydrodynamic description for the above mentioned conserved-mass transport processes, characterised the joint current and density profile distribution by explicitly calculating two transport coefficients - diffusivity and conductivity, and finally calculated the density large deviation function [53] using a recently developed macroscopic fluctuation theory [47]. It is shown that the macroscopic characterisation obtained through this dynamic set up is consistent to that previously obtained by us using additivity.

2. Motivation : Background of present thesis work

2.1. Near equilibrium statistical mechanics: Linear response theory

The response of a system to an external stimuli is important to understand its properties, and in fact it is the most obvious way to experimentally determine the properties of a substance. It is simultaneously true that, external perturbation to a system in equilibrium will necessarily cause a deviation from it (this is why the equilibrium response properties of matter is obtained in the limit of small external bias). On the other hand, this raises a question of principle. Since the equilibrium response is an inherent property of matter in equilibrium, it *should* be theoretically understandable in terms of the system's inherent parameters within equilibrium configurations.

The equilibrium fluctuation-response (FR) relations (alternatively fluctuation-dissipation theorems) obtained within linear response theory is precisely the recognition of the above requirement [2, 42]. It states that, given that some arbitrary but *small* departure from the most probable state took place, the system cannot distinguish whether it is created due to small external perturbation or due to equilibrium fluctuation. The mathematical statement is as follows. Let, at $t = 0$, a *small* external force $F(t)$ that is conjugate to an observable $A(C)$ is applied on a system (having configurations $\{C\}$) in equilibrium. For Hamiltonian system this changes the energy by $-A(C)F(t)$. Then, the change in the average value of an observable $B(C)$ over a time t is,

$$\langle \Delta B \rangle = \int_0^t dt' R_{BA}(t-t')F(t'),$$

$R(t)$ is the response function. With the help of Boltzman law, the FDT then relates this response to a small external stimuli with the equilibrium two point correlation functions through the following relation,

$$R_{BA}(t) = \beta \langle \dot{A}(0) B(t) \rangle_{\text{eq.}} = -\beta \langle A(0) \dot{B}(t) \rangle_{\text{eq.}}, \tag{2.1}$$

the second equality arising from time translation symmetry in equilibrium. Such FDTs can be proved for stochastic processes as well. As a simple illustration, let $B = A$, for which the response function is just proportional to the time derivative of equilibrium autocorrelation function $c(t) = \langle \dot{A}(0)A(t)\rangle_{\text{eq.}}$. Now if we choose the external force to be independent of time for all $t > 0$, the total change in the observable A in $t \to \infty$, i.e. the static susceptibility, is simply given by the equilibrium variance in the absence of external bias,

$$\gamma_A \equiv \langle \Delta A\rangle/F = \int_0^\infty R_A(t)dt = \beta[c(0) - c(\infty)] = \beta\sigma_A^2(F = 0), \tag{2.2}$$

which, a well known result in ESM, is a form of the Green-Kubo relation. For example, given A be the particle density, and F arising from a gradient in chemical potential, the lhs will equal the particle number fluctuation. Applying the above formulation to macroscopic quantities, such as densities and currents of conserved quantities, and identifying the external perturbation with 'thermodynamic forces', such as local gradients in pressure, temperature, or chemical potential, gives the bulk transport coefficients as the integrated time-correlation of respective conjugate fluxes [2]. Consider, for example, conductivity that characterises the mass current $J = \chi\nabla(\mu/T)$ arising in response to chemical potential gradient. In equilibrium it is equal to the time integrated correlation of equilibrium current fluctuation,

$$\chi = \beta \int_0^\infty dt \langle J(0)J(t)\rangle_{\text{F=0}}. \tag{2.3}$$

It is remarkable that, the quantities in last two equations are not independent. Kubo argued that, when the particles are not bound and interactions are short ranged, the fluctuation and conductivity will be finite and nonzero in the long wavelength (hydrodynamic) limit and are connected by the Einstein relation.

A very important dynamical property is the near equilibrium relaxation. Since the system cannot distinguish whether a small departure from the equilibrium state is due to the small external stimuli or equilibrium fluctuation, the nature of the irreversible relaxation of a macroscopic observable to its equilibrium value after the stimuli is switched off must be identical to spontaneous equilibration of its 'small' equilibrium fluctuation. This principle, known to be the 'Onsager regression hypothesis' [44], is contentwise essentially a particular case of the linear response theory as in Eq. (2.1). In consequence, the near equilibrium relaxation properties could be understood in terms of the same transport coefficients as in

Eq. (2.3), i.e., by the equilibrium time integrated correlations.

One can see this in a thermodynamic set up using locally defined densities. We note that, the nonzero rate of net entropy production, which is the defining property of irreversible relaxation, is linear in the rate of changes of other thermodynamic quantities near equilibrium. For example, in case of energy fluctuation alone, $\frac{\partial s_{tot}}{\partial t} = \frac{\partial s}{\partial t} + \nabla . J_s = \frac{1}{T}\frac{\partial \rho_e}{\partial t} + \nabla . \left(\frac{J_e}{T}\right) = J_e . \nabla\left(\frac{1}{T}\right)$, the last equality following from continuity of energy. Now, close to equilibrium, the energy current is linearly driven by temperature gradient, $J_e = -\kappa \nabla T = \kappa T^2 \nabla(1/T)$, κ being the thermal conductivity. Therefore, $\frac{\partial s_{tot}}{\partial t} = \kappa T^2 \nabla(1/T).\nabla(1/T)$, where κ is given by the Green-Kubo relation.

The transport coefficients enumerated by equilibrium fluctuation properties thus also characterise the near equilibrium relaxation behaviour. However, the analysis is not so obvious when it involves several forces inducing currents of several conserved quantities (or, alternatively, when one needs to characterise simultaneous fluctuation in several conserved variables). Usually in these cases, current of one variable is influenced by the force conjugate to another variable; or fluctuation in one influences the fluctuation of other. For example, in thermoelectric effect, potential gradient across a junction of two materials not only cause an electric current, but also creates a temperature difference causing energy flow. In a more day to day experience, sugar dissolves faster when the tea is hotter, implying that, apart form heat flow, temperature difference can also affect particle current. In general, the currents could be expressed in the form,

$$J_\alpha = \chi_\alpha F_\alpha + \sum_{\beta \neq \alpha} L_{\alpha\beta} F_\beta;$$

F_α are the thermodynamic forces which could be considered as local gradients in the intensive quantities causing fluxes to the conjugate conserved quantities, say K_α, so that the rate of local entropy production is given by $\frac{\partial s_{tot}}{\partial t} = \sum_\alpha \chi_\alpha F_\alpha . F_\alpha + \sum_{\alpha \neq \beta} L_{\alpha\beta} F_\alpha . F_\beta$. Onsager showed that, these new transport coefficients are in fact connected to the equilibrium temporal cross-correlation of the currents of these conserved quantities, and with the additional assumption that the systems are governed by reversible microscopic dynamics, these are shown to satisfy the 'reciprocity relation', $L_{\alpha\beta} = L_{\beta\alpha}$ [43, 44]. Note that, as in case of conductivity, these coefficients could be understood by a Green-Kubo relation for equilibrium current fluctuations in the form [54],

$$L_{\alpha\beta} \propto \int_0^\infty dt \langle J_\alpha(0) J_\beta(t) \rangle_{\mathrm{F}=0}, \tag{2.4}$$

and the reciprocity is established as follows. First, the microscopic reversibility implies, $\theta J(t) \equiv -J(-t) = J(t)$, θ being the time reversal operator. Consequently, $L_{\alpha\beta} \propto \langle J_\alpha(0) J_\beta(t) \rangle$ equals its time reversed value, $\theta L_{\alpha\beta} \propto \langle J_\alpha(0) J_\beta(-t) \rangle$. Then we perform a translation in time, $t' \to t' + t$ $(\forall t')$, and using the time translational symmetry in equilibrium, we obtain, $\langle J_\alpha(0) J_\beta(-t) \rangle = \langle J_\alpha(t) J_\beta(0) \rangle$, which is nothing but $L_{\beta\alpha}$.

So far we are concerned with the current due to external bias. The question is to understand the irreversible relaxation in the linear response regime. The regression hypothesis says that, the law governing this relaxation could be understood in terms of the spontaneous equilibration of small equilibrium fluctuation. In the language of ESM, these fluctuations δK_α will be Gaussian distributed, and the entropy change δS_{tot} (negative) due to fluctuation will be quadratic in them, $\delta S_{tot} = -\sum_{\alpha\beta} A_{\alpha\beta} \delta K_\alpha \delta K_\beta$. We have earlier seen that, the entropy production rate is also quadratic in the forces. Taken together, it suggests what one intuitively expects near equilibrium, that, the fluctuations δK_α are linear in the forces F_α. Then conjugate variables, defined as $k_\alpha = \frac{\partial(\delta S_{tot})}{\partial(\delta K_\alpha)}$ will then be a linear combination of all δK_α's. These two together imply that, k_α's are linear in F_α. Finally, using the linear form of currents J_α in the continuity equation for the conserved variables, $\frac{\partial \rho_\alpha}{\partial t} = -\nabla . J_\alpha$, and integrating it over the local region, we obtain, the rate of equilibration of the fluctuations δK_α as linear sum of F_αs. Combining all these, one writes,

$$\delta \dot{K}_\alpha = -\sum_\beta G_{\alpha\beta} k_\beta,$$

which relates the near equilibrium equilibration rate of the intensive quantities to the fluctuation of the corresponding fluxes. Using microscopic reversibility, Onsager showed that, the kinetic coefficients, $G_{\alpha\beta}$, satisfy the reciprocal relation, i.e., $G_{\alpha\beta} = G_{\beta\alpha}$ [44].

At this stage, it is worth mentioning that, although the above results are valid for close to equilibrium conditions, the approach, both in the static and the dynamic case, could be remarkably generalised for nonlinear and far from equilibrium conditions. Particularly, during last couple of decades, the way of analysing time-dependent behaviour in Onsager's theory is successfully applied and extended in understanding general macroscopic properties of transient density and current fluctuation in the framework of macroscopic fluctuation theory [47].

2.2. Beyond linear response : Fundamentals of large deviation theory

In the Onsager theory, the relaxation properties are understood when the fluctuation in thermodynamic quantities are small and satisfy a Gaussian distribution. Evans et al. have argued that, the FDTs, particularly the Green-Kubo relations, could be understood within central limit theorem, thus exhibiting the connection of the linear response theory with the 'typical' Gaussian fluctuations [55]. The aim is to understand physical properties when the fluctuation is very large, or the internal/external forces are such that these permanently drive the system away from equilibrium. Large deviation theory provides a useful tool to address such questions.

2.2.1. Large deviation and breakdown of central limit theorem

Let us revisit the idea of ensembles. An ensemble is the collection of all possible results of a measurement done on the system. In other words, results of infinite number of independent ideal measurements on identically prepared systems constitutes an ensemble. In this sense the probability we are concerned here is interpreted as the statistical probability. In an alternative formulation, we employ the concept of ensemble as an infinite collection of independent identically distributed (iid) random variables while doing the phase space average in statistical mechanics. Such collection of iids have important properties having direct consequences in this branch of physics. If the mean and variance of the iids $\{X_i\}$ be μ and σ^2 respectively, the mean value and the standard deviation of the *sample average* $M_n = \frac{1}{n}\Sigma_{i=1}^{n} X_i$ are respectively given by μ and $\frac{\sigma}{\sqrt{n}}$. The *Central Limit Theorem* (CLT) states that, given that the first two cumulants of X_i exist, they are sufficient to characterise the distribution of the sample average. In fact,

The distribution of M_n will be a Gaussian with mean μ and standard deviation $\frac{\sigma}{\sqrt{n}}$ at the limit of infinitely large sample size:

$$\lim_{n\to\infty} P\left(M_n = \frac{1}{n}\Sigma_{i=1}^{n} X_i\right) = \frac{1}{\sqrt{2\pi\frac{\sigma^2}{n}}} e^{-\frac{(M_n-\mu)^2}{2\sigma^2/n}} \tag{2.5}$$

Said differently, if W_i be the scaled deviation from mean, we can write, for each member of the iid, $X_i = \mu + \sigma W_i$, and,

$$M_n = \mu + \frac{\sigma}{\sqrt{n}} Z$$

where $Z = \frac{1}{\sqrt{n}}\Sigma_{i=1}^{n} W_i$ is dispersion scaled to order unity that follows a normal distribution with mean 0 and variance 1. We conclude that, as the sample size n is increased, the *error* in the sample average decreases as $n^{-1/2}$. In the limit of $n \to \infty$, the sample average is no longer a random variable and in fact, $M_n \to \mu$, which is the law of large numbers (LLN).

However, in central limit theorem we are really concerned with lower cumulants of the sample average M_n. CLT essentially makes an a priori assumption that the fluctuation Z itself goes as the *typical error*, namely, the sample standard deviation, which lies within $\mathcal{O}(\sqrt{n})$ off the sample mean, such that cumulants of higher orders are not important. The question is to find the probability of an arbitrary fluctuation. More specifically, if the fluctuation takes values of $\mathcal{O}(n)$, what would be the corresponding probability?

At such *large deviations*, the plausible form of scaled dispersion Z is $\frac{1}{n}\Sigma_{i=1}^{n} W_i$, and therefore the distribution away from the maxima will take the form,

$$\lim_{n\to\infty} P(M_n = x) \sim e^{-n\phi(x)}$$

in leading order. This motivates the mathematical statement of the *large deviation principle*,

$$-\lim_{n\to\infty} \frac{1}{n} \ln P(M_n = x) = \phi(x) \tag{2.6}$$

In this large n limit, the characteristic function is governed by the maximum of the integrand, and consequently the large deviation function $\phi(x)$ is the corresponding *optimum and concave* profile satisfying a Legendre-Fenchel transform (Gartner-Ellis Theorem) [56],

$$\phi(x) = \sup_s [xs - \lambda(s)] \tag{2.7}$$

$\lambda(s)$ being the cumulant generating function. The 'central limit' of CLT is suppressed to a point in the infinite n limit, and the Gaussian distribution is recovered only after a Taylor expansion to second order around the minima of $\phi(x)$. Note that, the law of large number is still valid, since for large n the distribution is sharply peaked about the mode and the distinction of mean and mode is only of order $1/n$.

Before going into the demonstrations for LDF, we briefly ponder upon the physical relevance of the above ideas in statistical mechanics. The ensemble picture indeed provides a strong statistical method giving statistical mechanics its remarkable predictive power. But it is of course not a physical picture even for an idealised isolated system. Secondly, in statistical mechanics we encounter quantities which are actually "correlated". However, there is an inherent and remarkable simplification in the physical picture of large systems that allow the concept of sample averages and the law of large numbers to arise naturally. We note that, it is essentially the sample averages that are observed as macroscopic quantities. For example, density of some quantity in a macroscopic volume is in fact the sample average of fluctuating mass in a huge number of microscopic cells or sites. Further, as motivated in the previous chapter, macroscopic quantities are associated with corresponding large time scales of observation. Consequently, the observed value is also the sample average over a large temporal duration. As it occurs in equilibrium and in a huge variety of nonequilibrium systems, the two-point spatial (and in many cases temporal) correlations of underlying microscopic degrees of freedom are either short-ranged, i.e. of order of microscopic length and time, or weak, so that the concerned observables defined over *macroscopic subsystems* and observed over different macroscopic intervals are almost uncorrelated. Homogeneity in these macroscopic scales imply such quantities to be identically distributed. These dynamical conditions are at the bottom of additivity property explored in detail in chapter 3. LLN is well established for such random variables [57] [1]. Powered by LLN and with the

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[1] In [58, 59], it is shown that, even strongly correlated random variables having diverging sample variance can

identification of entropy and other thermodynamic potentials as the large deviation functions, large deviation theory enables statistical mechanics to not only form the microscopic basis for equilibrium thermodynamics [56] but also to provide a framework to explore more general macroscopic features.

Side by side, in an operational perspective, this large and almost independent observation time picture can also be considered as a repeated and almost independent measurements of macroscopic quantities, which is reminiscent of measurement over an ensemble [2]. The large deviation property then imply the large fluctuations to be exponentially rare in the system size and measurement time, and thus repeated measurements will almost surely yield values 'close' to the typical value with deviations limited within the central limit. This explains why, although the phase space itself is exponentially large in system size while exponentially large measurement time is never feasible in practice, measurements in a mere macroscopic duration (of orders in simple powers of N), gives the same statistical averages that one otherwise expects to obtain over exponentially large number of trials [6]. Not only simulations are done with this assumption, but the real experimental *observation* of time stationary states like equilibrium bears the validity of these considerations.

Hydrodynamics is a remarkable example of application of the above considerations to nonequilibrium. Given that the conditions for short-ranged or weak correlations are satisfied, we may as well consider the above considerations to hold at individual subsystem (microscopically large but macroscopically small) level. Then, this subsystem, behaving as statistically an almost independent system, acquires the local/quasi equilibrium properties in an appropriate macroscopic yet small time. This is the core idea employed for obtaining the hydrodynamic description [60].

As one last comment in this subsection, we would like to mention that, the very proposition of short-ranged and/or weakly correlated random variables for describing statistical

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satisfy LLN under certain restrictions.

[2] Putting it in the reverse order, the success of ensemble picture imply that, in most circumstances, the physical value, that is nothing but the average measured over large observation time, is indeed given by the statistical average calculated over allowed phase space (or ensembles). This strongly suggests that, the measurements over different intervals are statistically almost uncorrelated.

properties of macroscopic quantities bring forth another remarkable physical consequence, the full strength of which is understood only recently and is the theme of the chapters on additivity and of fluctuating hydrodynamics using MFT. It is shown that, given that the system is homogeneous, correlations are short-ranged, and the microstate is specified by the configuration of a locally conserved quantity, the full static as well dynamic large deviation probability of *arbitrary* fluctuation (beyond CLT) of a macroscopic quantity can still be completely characterised by the knowledge of its first two moments.

2.2.2. An example : random walk in one dimension

Assume that in a lattice $[-L, L]$, a random walker has taken N steps starting from the origin and have reached a site x. The probability of taking a step along the positive axis is p, and that along the negative direction is $q = 1 - p$. We assume, $L \gg N$ always. Two cases:

(1) when N is *infinitely* large, x is finite/small;

(2) when x can be comparable to N.

The probability of (x, N) is given by the binomial distribution,

$$P(x,N) = \frac{N!}{\frac{N+x}{2}!\,\frac{N-x}{2}!}\, p^{\frac{N+x}{2}} q^{\frac{N-x}{2}} \tag{2.8}$$

Using the definition in Eq. (2.6), for $N,\ N+x,\ N-x$, all very large, we make the Stirling approximation, $N! \approx \sqrt{2\pi N} N^N e^{-N}$ and obtain,

The LDF:

$$\phi\left(\frac{x}{N} = r\right) = \frac{1}{2}(1+r)\ln p + \frac{1}{2}(1-r)\ln(1-p) + \ln 2 - \frac{1}{2}(1+r)\ln(1+r) + \frac{1}{2}(1-r)\ln(1-r) \tag{2.9}$$

and get the result for (2).

Expanding Eq. (2.9) upto quadratic order around its minima, we obtain, after putting back $r = \frac{x}{N}$,

$$P(x,N) \approx \frac{1}{\sqrt{8\pi pqN}} e^{-\frac{[x-(p-q)N]^2}{8pqN}} \tag{2.10}$$

which is the well known result for random walk in case (1).

2.3. Application of large deviation theory

2.3.1. In equilibrium

While studying bulk physical properties of stochastic systems, we care about fluctuations which actually affects the bulk measurement, and therefore we need to characterise the atypical fluctuations given by large deviation functions.

The natural description of equilibrium thermodynamics is given by LDF. In equilibrium the partition function in some suitable ensemble is given by, $Z = \Sigma_N e^{-\beta E(N)}$, N being the fluctuating extensive variable and $E(N)$ being the total energy. The free energy defined by $Z = e^{-\beta F} \sim e^{-\beta V f(N*/V)}$ is dominated by the most probable density $\rho*$ (having typical fluctuation of $O(N^{-1/2})$). However, the distribution for *any n* in a macroscopic subsystem of size v conditioned to a total particle content N is given by,

$$P_v(n) = \frac{Z_v(n) Z_{V-v}(N-n)}{Z_V(N)} \approx e^{-v\phi(\frac{n}{v})}$$

to the leading order, with $\phi(r) \approx \beta\left[f(r) - f(\rho) - (r-\rho)f'(\rho)\right]$, $\rho = N/V$ being the equilibrium large deviation function for subsystem density. However, when the condition is relaxed, the probability in question is obtained simply by replacing ρ by $\rho*$ in the above equation. This could be extended to write down the joint probability distribution of the subsystem number fluctuation giving the equilibrium *large deviation functional* for the whole density profile,

$$\mathcal{F}([\rho(\vec{x})]) = \beta\left[f(\rho) - f(\rho^*) - (\rho-\rho^*)f'(\rho^*)\right]. \tag{2.11}$$

This expression could be simply understood by assuming the concerned system to be a part of a much larger conserved system having density $\rho*$. The above expression assumes the form

$$\mathcal{F}([\rho(\vec{x})]) = \beta \int dv[f(\rho) - f(\rho^*)] \tag{2.12}$$

for conserved systems. We can see from Eq. (2.6) that, in the microcanonical ensemble where the postulate of equal a priori probability is valid, $\phi(x)$ is identical to the entropy density which is an LDF in equilibrium microcanonical ensemble. In fact, once we identify the conjugate quantities, the most general equilibrium fluctuations could be written in the form of Einstein fluctuation equation,

$$P \approx exp(-\Delta W) = exp(\Delta S - \beta\Delta U - P\Delta V - \mu\Delta N - \cdots).$$

The whole of equilibrium statistical mechanics built upon the properties of free energies is naturally expressed by the large deviation theory [56]. The free energies consistent to different ensembles and interpreted as the corresponding large deviation functions can be seen to be related by Legendre-Fenchel transforms in equilibrium, i.e. the most probable state (for example, the Helmholtz free energy is the transform of of entropy, and the Gibbs free energy is another transform of Helmholtz free energy), restricting them to be concave functions, which is physically reminiscent to the laws of thermodynamics. The equilibrium phase transitions could be understood when the large deviation function fails to be strictly concave, and the thermodynamic behaviour is characterised by its concave envelope giving rise to Maxwell construction.

2.3.2. Away from equilibrium

The example of random walk one dimension itself constitutes an example of a nonequilibrium process. It is possible in general to characterize systems having a equilibrium state as well as a non equilibrium steady state in terms of the large deviation function that generalises the concept of *free energy density* to nonequilibrium. It could capture various macroscopic properties of the systems. Therefore we aim to study the LDFs in general for various systems of interest, and the purpose of this thesis is to characterise such large deviation functions for various nonequilibrium systems and find out its physical consequences. Prior to that, it would be instructive to look into few known examples.

Let us take for example the *boundary driven Symmetric Simple Exclusion Process*. The model is defined on a 1 dimensional lattice with two ends connected with particle reservoirs at densities ρ_a and ρ_b respectively. At the boundaries, the system and reservoir can exchange particles at certain rates to cope with the reservoir densities. In the bulk, the particle can jump to any of its neighbouring sites with rate 1 if it is empty. It is shown that [1], at the limit of large system size, the probability of observing a density profile $\rho(x)$ will be given by the large deviation functional,

$$\mathcal{F}([\rho(x)],\rho_a,\rho_b)=\int_0^1 dx\left[B(\rho(x),F(x))+\ln\frac{F'(x)}{\rho_b-\rho_a}\right] \tag{2.13}$$

where $F(x)$ is given by the *monotonous* solution of the equation,

$$\rho(x)=F+\frac{F(1-F)F''}{F'^2},$$

and

$$B=(1-\rho)\log\left(\frac{1-\rho}{1-F}\right)+\rho\log\frac{\rho}{F}$$

is the large deviation function when the densities of the two baths are identical. It is manifestly obvious that, in presence of global density gradient, the large deviation function is *nonlocal*, that the local density at some position is directly affected by the distribution everywhere else. This system thus exhibits long range correlation that can be calculated from the knowledge of the large deviation function.

In the following sections there are other examples of nonequilibrium large deviation functions.

2.4. Previous attempts to construct 'thermodynamics of NESS'

In lines of equilibrium, an understanding of nonequilibrium steady states (NESSs) within a universal macroscopic framework is a longstanding question. Taking a phenomenological approach to provide a macroscopic characterisation, akin to thermodynamics for equilibrium, constitutes the idea of 'steady state thermodynamics (SST)'. The goal is to build up a theory for macroscopic processes based on some unifying principle(s) that can actually extend the laws of thermodynamics and incorporate both equilibrium and nonequilibrium phenomena in its purview. This resulted in an 'operational' approach for generalising the thermodynamic quantities and principles to SST is undertaken for last couple of decades. We outline few main results in this direction in conjunction to the present thesis.

Oono and Paniconi in [61] proposed a phenomenological generalisation of the concepts of thermodynamic variables that specify macroscopic states, quasi-steady processes, heat, entropy, and thermodynamic potentials, and went on to propose coherent empirical laws for steady states of a macroscopic system in lines with thermodynamics. However, a very important distinction with equilibrium is that, to *maintain* a NESS, we need to supply energy to balance the irreversible dissipation, and therefore need to define a 'housekeeping heat' Q_h (which vanishes if we have equilibrium as the steady state). Therefore, to realise a nonequilibrium process if a total Q_{tot} amount of energy is dissipated, one must consider the excess heat $Q_{ex} = Q_{tot} - Q_h$ while describing the phenomenological laws. For example, the generalised second law of SST for a generalised entropy and temperature will state that, $T\Delta S \geq -Q_{ex}$. Subsequently, this formalism supplemented by the microscopic inputs from a generalised Jarzynski equality was utilised to derive a concrete form of nonequilibrium entropy and the second law for a Langevin system [62].

As equilibrium statistical mechanics gave a microscopic and statistical understanding of the laws of thermodynamics, the ideas of SST were similarly explored by Sasa and Tasaki in more quantitative manner with a statistical mechanics viewpoint to SST. Their approach was to take operational thermodynamic definitions of macroscopic quantities understood via appropriately modelled macroscopic interactions among steady states [63]. For example, they have defined a nonequilibrium chemical potential that balances the particle flow in two parts of the fluid due to potential difference, exactly as in equilibrium thermodynamics, $\mu(\rho_1) + V_1 = \mu(\rho_2) + V_2$ (eq. 1.1 of their paper), and broad thermodynamic characterisation follow. We specially mention this equation because, as we understood so far and will present in the thesis, a broad class of nonequilibrium systems do have such a equilibriumlike thermodynamic characterisation, and the notions of chemical potential and equations like this come as consequence.

However, when there comes the question of interaction of steady states, or the system-bath interaction in the SST framework, questions rise on the important notion of zeroth law, thereby questioning the existence of a thermodynamics at all. Actually, Oono-Paniconi in [61] argued that, a zeroth law is not needed for SST as long as the different intensive parameters could be related by a smooth scale factor. However, in that case, phase transition, in particular phase coexistence, could not be defined within the thermodynamic framework - a factor much emphasized by Dickman. It is well known that, generic interaction rules of two systems violate zeroth law and ensemble equivalence. On the other hand, he argues that if we choose a special rule - the Sasa-Tasaki (ST) rule that fixes the exchange rate to the state of the donor only - of interaction among the system and bath (both taken to be uniform), then some (rather ad hoc) intensive thermodynamic parameter could be identified that satisfy the zeroth law [64]. However even the ST rule fails when the system is inhomogeneous, for example between the two phases in case of a coexistence [65]. In such case, the parameter values at which the coexistence takes place will be different from the value taken when separate phases of the same system are connected by the Sasa-Tasaki interaction; in such case unique thermodynamic characterisation of the system will not be possible.

In an alternative development, as will be elaborated in the next chapter, Chatterjee *et al.* provided very general conditions for a system to have a description in terms of extensive variables like nonequilibrium free energy, and conjugate intensive chemical potential. Remarkably, in [52], they showed that, if two such systems interact with rates connected by the exponential of the free energy difference of the contact region, $\frac{u_{12}}{u_{21}} = e^{-\Delta F}$, not only the zeroth law but an equilibriumlike fluctuation-response (FR) relation holds - thereby providing a complete thermodynamic description of interacting NESSs. The results provide a concrete definition of the intensive variable, and the validity of FR allows a Maxwell construction for the coexisting phases, thereby proposing an immediate solution of the problem put forward by Dickman.

This takes us to the context of additivity property in steady states, that allow the study of fluctuations of a conserved quantity via a nonequilibrium counterpart of intensive thermodynamic parameters, and provides a consistent and complete thermodynamic characterisation for a broad class of models. Additivity is a well known property in equilibrium. While studying the hydrodynamic limit of driven KLS model, Eyink *et al.* identified the scale separation required for having a hydrodynamics with the joint steady state probability measure to be approximately separable, $P(n_1, n_2) \approx \sup_{n_1, n_2, n_1+n_2=n}[P(n_1)P(n_2)]$, identified a concave quantity resembling a free energy, and also identified an FR relation [48]. Bertin *et al.* argued that, for a broad class of models, an additivity property holds, which enable the identification of an intensive variable, such that, it takes uniform value for different subsystems. It is further shown to be related to the subsystem mass fluctuation by an equilibriumlike FR relation, and demonstrated for systems like ZRP having a product-measure steady state [49, 66]. Subsequently, it was shown that, this additivity property, which must be valid for subsystems much larger than correlation length, is in principle enough to provide *complete* distribution of subsystem mass fluctuation of a conserved model, provided the functional form of subsystem mass variance is known [50].

With this intriguing developments, universal characterisation of broad class of stochastic model systems could be understood, as will be discussed subsequently.

2.5. Characterisation of model systems having a NESS

2.5.1. Mass transport processes

The mass transport processes have been studied intensively in the last couple of decades and have become a paradigm in nonequilibrium statistical physics of interacting many particle systems. Actually a huge variety of natural phenomena spanning a wide range of length scales (many of which outlined in last chapter) can be modelled using such simple mass transport processes. They represent, for example, formation of clouds [67], river networks [68], gels [69,70] and planets [71], formation of lipid droplets on cell surface [72], fragmentation and self-assembly in various materials [73], condensation of fluids on cold substrates [74], traffic flow [75], wealth distribution [76] and migration and formation of cities [77], and so on.

In the thesis we broadly consider models with conserved total mass. Such model was introduced as the Hammersley process [78] and as a model of force fluctuations in a pack of granular beads [79,80]. They were consequently generalized to various stochastic processes, called random average processes (RAPs) [27, 81–85] or, equivalently, called mass chipping models (MCMs) [28, 31, 50]. There are also several other variants of these mass transport processes, which we call mass exchange models (MEMs), where neighboring sites across a bond exchange among themselves a random fraction of their added masses [86, 87]. In Fig.

2.1, a schematic of mass chipping models is provided.

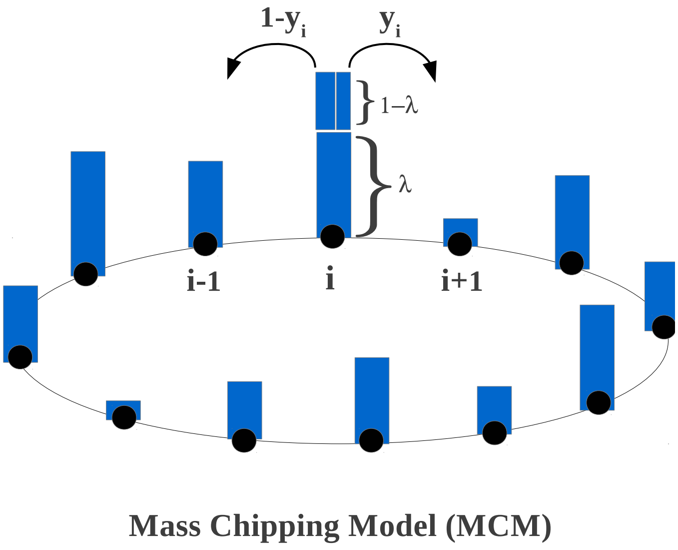

Figure 2.1.: Schematic of one dimensional conserved mass chipping processes. Details given in chapter 5.

While in the above models the mass variables are continuous, there are a wide variety of models having discrete mass variables. They include the Katz-Lebowitz-Spohn (KLS) model and several exclusion processes which have been studied widely in last decades, few of which can be found in references [1, 20, 48, 88–91]. Of models allowing arbitrary number of particles include Zero Range Process (ZRP) [23, 49], models having pair factorised steady states [40, 41] and more recently studied finite range processes [22], and several others. A large class of models (that include many of the above) show condensation phenomena, of which an important variety is the conserved mass aggregation models defined with competing dynamics [29]. A broad class of models are dedicated for the study of self organised criticality [36, 37]; their widely studied conserved-mass versions, known as fixed energy sandpiles (FES), predict a novel nonequilibrium phenomena, viz., active-absorbing phase transitions [38, 92–94]. Of this wide variety of systems, we will mainly concern the broad subcategory of continuous mass transport processes and the mass aggregation models. A schematic of a symmetric mass aggregation models is given below.

It is interesting to note that, these models are defined with simple stochastic dynamics involving a single conserved quantity, and the interactions are limited within the nearest

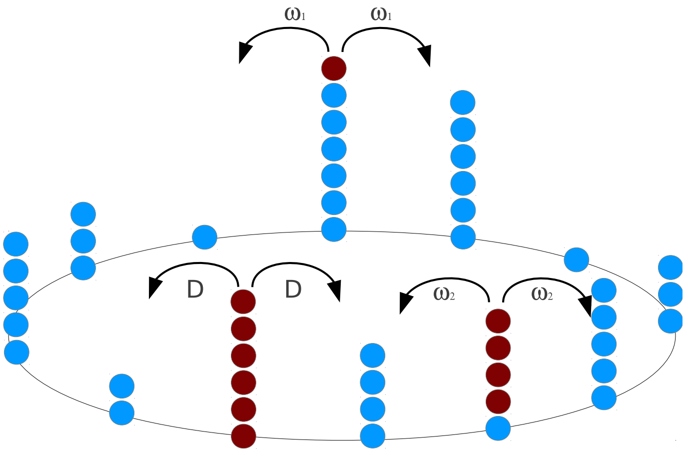

Figure 2.2.: Schematic of one dimensional conserved mass aggregation models. Discussed in section 2.5.2, and in chapter 4.

neighbours only. These are perhaps some simplest possible model systems, but they give rise to a rich variety of features, whose characterisation pose a great challenge.

2.5.2. Violation of detailed balance

Our claim that these models represent nonequilibrium processes could be substantiated by observing that, these models violate the Kolmogorov criterion for detailed balance. It is manifestly obvious for asymmetric models where there is a mass current in the steady state. Even the symmetric versions, although there is no net mass current, this violation is in general true for generic parameter values. Here, as a demonstration, let us consider the conserved mass aggregation model as shown in Fig. 2.2. Here, the total mass at an arbitrarily chosen site can either diffuse to any of its neighbours with a rate D, a single particle can chip off and coalesce to any of the neighbours with a rate w_1, and all but one particle can fragment and merge with any of the neighbouring mass with a rate w_2. Notice that, after a diffusion event at site i having a mass m_i, the new configuration necessarily have a site with mass $m_i' = 0$, and a neighbouring site with mass $m_{i+1}' = m_i + m_{i+1}$. For arbitrary m_i and m_{i+1}, the reverse process simply does not exist, and therefore there is no Kolmogorov criterion.

Similar arguments are valid for the continuous mass chipping processes and will be detailed in Chapter 5.

2.5.3. Steady state mass distributions in above processes

Calculation of the mass distribution for these wide variety of conserved mass transport processes, both in a single site and at a subsystem level, is a difficult problem, and it is in general not known barring few exceptions. However, through intense works in last few decades, some remarkably features came up. It is found that, based on asymptotic mass distribution at a single site level, there are three broad categorisation. There are are models that approximately obey gamma distribution [27, 28]: $P(m) \propto m^{\eta-1} e^{-\eta m/rho}$. The second category of models follow a particular scaling form: $m^{-\tau} e^{-rho^{\delta} m}$ [29, 30]. The third category exhibits condensation transition at a finite critical density, below which the asymptotic mass distribution obeys the above scaling form, and at and above criticality, there exists a condensate carrying phase, while the coexisting fluid obeying a pure power law. Interestingly, the power law exponents below and above criticality were reported as different in [29], which differ from the prediction by other works [14]).

At this point it is worth noting that, the familiar notion of no thermodynamic phase transition in 1D is obviously not working. Actually, these systems violate the van Hove/Ruelle's assumptions for having no phase transition, by allowing, for example, indefinite number of particles in a single site. Even the one dimensional symmetric zero range process which obey detailed balance demonstrates condensation.

2.6. Dynamic characterisation

Dynamic characterisation of currents and the time dependent properties of physical observables are integral to the complete understanding of nonequilibrium phenomena. Although not addressed in the thesis, we must mention that, dynamic correlations in transient as well as steady states are very important quantity for understanding dynamical properties. Not only it is needed for characterisation of dynamic fluctuations and currents, but it might also be useful for the hydrodynamic description. Despite the necessity there are not many comprehensive results except in few cases, like the Random average processes [26,82]. The present discussion will be confined to macroscopic current distributions, for which, it turned out that, knowledge of microscopic dynamic correlation is not a priori needed so far for deriving quantitative results.

2.6.1. Characterisation of current distribution and additivity principle

Behaviour of macroscopic systems that carries a steady current is a central problem of the study of non equilibrium statistical mechanics. An open system in contact with two particle reservoirs at different density is one paradigm of central interest. The other paradigm is of course steady current in mass transport processes when the system satisfies periodic boundary conditions.

In many cases the LDFs for current and density distribution are *nonlocal functionals* of the density and current profiles [1, 46]. However in the absence of long range correlation, an additivity principle for the current fluctuations can be formulated. Being a very important development, we shall elaborate the hypothesis and the steps involved, and also mention the main results. The way the problem is tackled is of direct relevance to the considerations in the following chapters.

Before proceeding further, we note that, conservation of particles imply that, the local current and number density is related : $\frac{\partial \rho}{\partial t} = -\frac{\partial j}{\partial x}$. Therefore the complete characterisation must include fluctuation of current and number density simultaneously.

Let us take a 1D system in contact with particle reservoirs of densities ρ_a and ρ_b respectively at the two ends. Also assume that, $\rho_a > \rho_b$. We want to calculate the probability of the number of particles $Q = jt$ being transmitted through the system for a large time t.

We want to calculate the large deviation function of average integrated current,

$$P\left(\frac{Q_t}{t} = j\right) \sim e^{-tF_L(j,\rho_a,\rho_b)} \tag{2.14}$$

L, the system size, is taken to be large and we assume diffusive dynamics in the local coarse grained scale. Let us break the system into two subsystems of lengths L and L' respectively. From Fig. 2.3, we see that, the total number of particles (integrated current) passing through the whole system of length $L + L'$ is also passing through each of the subsystems. Additivity principle states that,

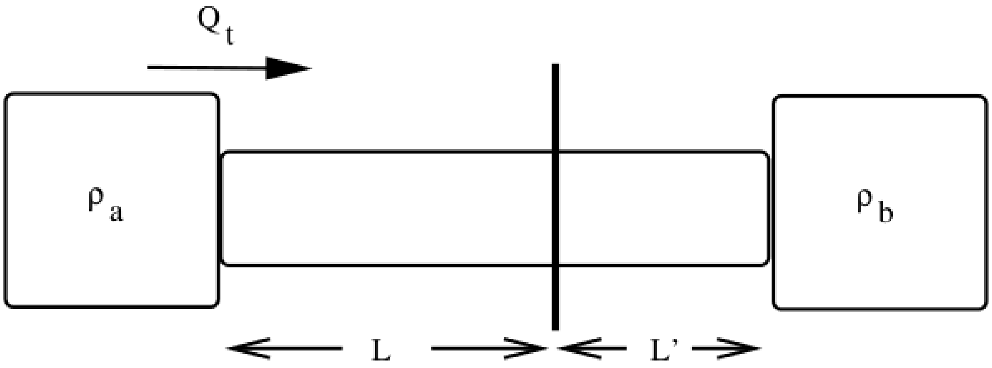

Figure 2.3.: Additivity Principle : Current of a system in terms of current through the subsystems. Image taken from [1].

The probability of transporting a current j *over a length* $L+L'$ *is the maximised product of the respective probabilities of carrying the same current through the subsystems.*

In other words, the density ρ at the contact of the subsystems is adjusted such a way that,

$$P_{L+L'}(j,\rho_a,\rho_b) = \{\max_{\rho} P_L(j,\rho_a,\rho)P_{L'}(j,\rho,\rho_b\}) \tag{2.15}$$

$$\Rightarrow F_{L+L'}(j,\rho_a,\rho_b) = \min_{\rho}\{F_L(j,\rho_a,\rho)+F_{L'}(j,\rho,\rho_b)\} \tag{2.16}$$

The idea is to divide the whole system into large number of such subsystems with size much greater than correlation length and apply the above principle at every contact point. This gives,

$$F_L(j,\rho_a,\rho_b) = \min_{\rho_a,\rho_1,\rho_2\cdots\rho_i,\cdots\rho_b}\{\Sigma_{i=1}^{k-1}F_{\Delta L_i}(j,\rho_i,\rho_{i+1})\}, \tag{2.17}$$

$k \gg 1$ being the number of subsystems. In the $k \to \infty$ limit the discrete sum is converted to integral and the minimization is done over the whole density profile.

For diffusive dynamics we introduce the local diffusion coefficient as,

$$\frac{\langle Q_t\rangle}{t} = D(\rho)\frac{\Delta\rho}{\Delta L} \tag{2.18}$$

We introduce another quantity equivalent to local conductivity through the local variance of current under the condition of vanishing gradient,

$$\frac{\langle Q_t^2 \rangle}{t} = \frac{1}{\Delta L}\sigma(\rho) \tag{2.19}$$

Since L is very large, the current $j \sim \frac{1}{L}$ is small. In this limit, we can assume the density is locally uniform within the subsystem of size ΔL. The local diffusive profiles will then satisfy a Gaussian distribution,

$$\begin{aligned} P_{\Delta L}(j,\rho,\rho-\Delta\rho) &\approx e^{-\frac{(Q-\langle Q \rangle)^2}{2V_Q}} \\ &= e^{-t\frac{\left(j+D(\rho)\frac{\Delta\rho}{\Delta L}\right)^2}{2\sigma(\rho)/\Delta L}} \end{aligned} \tag{2.20}$$

We now rescale the current to $\mathcal{O}(1)$, $J = jL$, and define $x = \frac{l}{L}$, l being the distance of the concerned subsystem from the left reservoir. With these definitions, we can rewrite the local current distribution as,

$$P_{\Delta x}(j,\rho,\rho-\Delta\rho) \approx e^{-\frac{t}{L}\frac{(J+D(\rho)\rho')^2}{2\sigma(\rho)}\Delta x} \tag{2.21}$$

According to the additivity principle, the probability of a current j through the whole system after a very large time is given, in the continuum limit, by,

$$P_L(J,\rho_a,\rho_b) = \max_{\rho(x)}\left[e^{-\frac{t}{L}\int_0^1 dx\frac{(J+D(\rho)\rho')^2}{2\sigma(\rho)}}\right] \tag{2.22}$$

Therefore the current large deviation functional is,

$$F_L(J,[\rho(x)]) = \frac{1}{L}\min_{\rho(x)}\left[\int_0^1 dx\frac{(J+D(\rho)\rho')^2}{2\sigma(\rho)}\right] \tag{2.23}$$

From Eq. (2.23) it is observed that, the current large deviation functional F_L scales as L^{-1},

$$F_L(J,[\rho(x)]) = \frac{1}{L}G(J,[\rho(x)])$$

As expected the average profile with no fluctuation corresponding to the one for which the time dependence of the current distribution vanishes is given by $F_L(J_s,[\rho_s(x)])=0$, or,

$$J_s + D(\rho_s(x))\rho'_x = 0 \tag{2.24}$$

On the other hand, The *optimal density profile* conditioned to a given J and satisfying Eq. (2.23) with the boundary condition $\rho(0)=\rho_a$ and $\rho(1)=\rho_b$ is obtained from the Euler-Lagrange equation,

$$\begin{aligned} D^2(\rho)\left(\frac{d\rho}{dx}\right)^2 &= J^2(1+2K\sigma(\rho)) \qquad (2.25)\\ \Rightarrow J = \frac{J_s}{\sqrt{1+2K\sigma(\rho)}} & \end{aligned}$$

'K' being the constant of integration to be adjusted by the boundary conditions. It can be shown that the cumulant generating function $\mu_L(\lambda)$ is obtained through eliminating K from the following equations,

$$\lambda = G = \int_{\rho_b}^{\rho_a} d\rho \frac{D(\rho)}{\sigma(\rho)}\left[1-\frac{1}{\sqrt{1+2K^*\sigma(\rho)}}\right] \tag{2.26}$$

$$\mu = -\frac{K^*}{L}\left[\int_{\rho_b}^{\rho_a}\frac{d\rho D(\rho)}{\sqrt{1+2K^*\sigma(\rho)}}\right]^2 + o(1/L) \tag{2.27}$$

Consequentially, large deviation functional $G(J,[\rho(x)])$ can be obtained in principle using a Legendre-Fenchel Transform [56].

Violation of additivity principle

Phase transition: The diffusion equation allows a scaling symmetry,

$$t \to \frac{t}{\lambda^2}\ ,\ x \to \frac{x}{\lambda}$$

These imply, for a large system of size L and at a large time $\sim L^2$, the time dependent density and current will have the scaling relations,

$$\rho(i,t) = \hat{\rho}\left(\frac{i}{L},\frac{t}{L^2}\right) = \hat{\rho}(x,\tau) \ \text{ and } \ \mathrm{Q(i,t) = L\hat{Q}\left(\frac{i}{L},\frac{t}{L^2}\right) = L\hat{Q}(x,}\tau) \tag{2.28}$$

x, τ being the coarse grained *macroscopic* position and time.
Bertini et al showed, using macroscopic fluctuation theory [1, 47, 95], that the LDF corresponding to the probability of observing the average rescaled current $J = \frac{1}{\tau}\int_0^\tau d\tau' \int_0^1 dx\hat{j}(x,\tau')$ will be of the form,

$$F(J) = \lim_{\tau\to\infty} \frac{1}{\tau} \min_{\hat{\rho}(x,\tau),\hat{j}(x,\tau)} \left[\int_0^\tau d\tau \int_0^1 dx \frac{(\,\hat{j}(x,\tau) + D(\hat{\rho}(x,\tau)\rho')^2}{2\sigma(\hat{\rho}(x,\tau))} \right] \tag{2.29}$$

along with the constraints of particle conservation, $\frac{\partial \rho}{\partial t} = -\frac{\partial j}{\partial x}$ and the definition of average current J.

The identical structures of the equations imply that, Eq. (2.29) will give the same expression as in Eq. (2.23) when the optimal profiles of density and current are time independent. However, as both Bertini and Derrida [1] pointed, under certain conditions $F(J)$ of Eq. (2.29) is no longer a convex function [47, 95] and we cannot use the additivity principle. In this case a unique time independent optimal solution does not exist and the true optimal profile becomes time dependent. Such a situation here imply a *phase transition*.

Non-local LDF: For systems exhibiting long range correlation, the large deviation functional is non-local, and the notion of breaking systems into independent subsystems and hence additivity in principle breaks down.

However, for boundary driven SSEP, we can see from Eq. (2.13) that the LDF exhibits non-local behaviour, since $F(x)$ involves value of the density of all points. Therefore it exhibits a very weak but long range correlation. However it is shown that this system do obey the results given by additivity principle [1]. Here long range correlation is the consequence of different boundary densities, and this does not affect the bulk dynamics; local current continues to depend only on local dynamics that continues to be purely diffusive and considerations like local equilibrium and local Gaussian description holds. Systems exhibiting strong enough correlation over a long range will violate this additivity principle.

These violations, and the question of large deviation property of general time dependent profiles for general characterisation of the bulk properties of nonequilibrium fluids, necessitates a consistent coarse-graining procedure in the continuum limit, which is the aim of fluctuating hydrodynamics.

2.6.2. Fluctuating hydrodynamics

Fluctuating hydrodynamics brings forth an important difference in approach compared to Derrida's additivity, that is, the former aims to arrive at a macroscopic and dynamic description of matter starting from microscopic laws of motion, and therefore conforms to the spirit of statistical mechanics in a purely dynamic set up. To our present understanding, a hydrodynamic description is possible if dynamic quantities like $\frac{\partial\rho}{\partial t}$ could be connected to macroscopic average quantities having a local stationary description (details in section 6.1). Although achieving such a description is of fundamental importance, for most of the deterministic and stochastic many particle systems it remains largely elusive. The situation, however, improved gradually over last couple of decades, and a broad framework has emerged only recently with the advent of Macroscopic Fluctuation Theory (MFT).

2.7. Introduction to macroscopic fluctuation theory

MFT signifies an important progress towards obtaining a fluctuating hydrodynamics description for a broad class of driven diffusive systems with microscopic changes governed by simple non-trivial stochastic dynamical rules. As a very significant development over the near equilibrium statistical mechanics, let us briefly point out the main features.

First of all the formalism is not restricted to small fluctuations. Actually, it provides within its framework a method to calculate the nonlinear large deviation probabilities. Secondly, within Onsager theory, the small current was linearly dependent on the fluctuation of the conserved quantities, and a separate characterisation of the current was out of place, which is generalised in MFT and a method for studying features of current distribution is given. In this context, the consequence of the time reversal operation to 'macroscopic' distributions have been explored, and a variational principle for stationary distribution is discovered. It turns out that, irrespective of details of microscopic dynamics, if time reversal is a symmetry of the steady state macroscopic fluctuations, the Einstein relation will be a consequence.

To go beyond the linear response regime, we need to mention a remarkable and quite general property of Markov processes, that, for such processes a small external perturbation can actually lead to large and arbitrary fluctuation. In other words, a Markovian system cannot distinguish whether an *arbitrary* configuration is attained owing to pure fluctuation or by an external stimuli. This enables one to in principle *condition* an appropriate bias to the rare fluctuations of an unperturbed system and study their behaviour [47, 96], which, have been crucial in building up the MFT framework.

Detailed discussion of the above matters is outside the scope of the thesis and the review by Bertini *et al.* is referred [47]. However, it would be pertinent to mention that, despite this significant development, the MFT could be used and the fluctuating hydrodynamics is actually obtained for only a handful of systems, most of which either obey detailed balance, or allow a factorised steady state leading to important simplifications.

2.8. Plan of the thesis

As mentioned in last chapter, unifying these variety of results in these large class of models based on a common underlying principle in a single framework is both a necessity and challenge, and over last couple of decades, we believe the progress along the lines of additivity property and MFT gives rise to a concrete and possible approach towards a synthesis.

With this in view, the later chapters in the thesis are organised as follows. Three chapters in Part I are concerned with static characterisation of mass transport processes. In chapter 3, the additivity property for conserved-mass systems is defined and the consequences are discussed. It is demonstrated that this property alone leads to a equilibriumlike thermodynamic characterisation of macroscopic subsystem, and the knowledge of static two-point correlations is in principle sufficient to have quantitative characterisation of NESS. We have also briefly discussed the nonconserved models, while a concrete example of nonconserved system is discussed in Appendix A. Chapter 4 addresses the question of the asymptotic scaling forms commonly observed in widely varying mass-transport processes. How additivity leads to the observed scaling forms starting from the power law nature in asymptotic forms of the mass variance is explained. It is further shown that, a scaling relation proposed earlier [30] between the exponents of the variance and that in the mass distribution is actually a very general feature. On other hand, the thermodynamic characterisation of nonequilibrium condensation transition is developed, and quantitative analytical results (in excellent agreement with numerical data) are obtained for a broad class of mass aggregation models assuming additivity to be valid at a single site level (one can numerically check the separation of two point correlations). For a large class of nonequilibrium continuous mass transport processes having nontrivial correlations, analytical results for exact steady state two point correlation functions are calculated in chapter 5. The correlations being short-ranged, we went on to calculate the subsystem mass distribution. For each model we demonstrated the numerical data with gamma distribution as predicted by additivity.

In Part II, a detailed dynamic characterisation of macroscopic quantities for mass transport processes is discussed. Chapter 6 provides an introductory discussion of macroscopic fluctuation theory (MFT). The form of steady state distributions in large spatial scales, the coarse graining procedures, and the formulation of MFT are detailed. In chapter 7 we discuss the application of MFT in systems inherently far from equilibrium. At first, linear response around a NESS is discussed. Then the formalism is applied to one dimensional nonequilibrium conserved mass transport processes; the transport coefficients are exactly calculated and the fluctuating hydrodynamics description is built up. This formalism is then extended to some extent for processes with asymmetric mass transfer. Finally the density fluctuations in the steady state limit are characterised and results compared with the predictions of additivity. A summary of the thesis is given in chapter 8.

Part I.

STATICS

3. The additivity property

Nonequilibrium steady states, although is the closest counterpart of equilibrium, neither have a counterpart of Boltzmann law nor of the equal a-priori probability. These states violate detailed balance. However, the same may not be true regarding additivity. As was discussed in section 2.2 of previous chapter, this property has its foundation on certain dynamical conditions allowing a description of macroscopic quantities in terms of almost uncorrelated random variables. It is expected that such conditions and therefore the additivity property will hold for a broad class of nonequilibrium processes. We will seek the consequences.

Let us discuss additivity first in the context of equilibrium. For equilibrium systems having an energy function E with short-range interactions, the microscopic weight of a configuration C can be written in terms of the Boltzmann distribution $P(C) \sim \exp[-\beta E(C)]$ where β is the inverse temperature. It is well known that, in the thermodynamic limit, such an equilibrium system can be divided into many large subsystems which, being large and thus statistically almost independent, can be characterized using thermodynamic potentials like entropy or free energy function. For example, joint distribution $P[\{N_k\}]$ of subsystem particle-numbers $\{N_1, N_2, \ldots, N_\nu\}$ in a system of volume V, which is kept in contact with a heat bath of inverse temperature β and has a fixed total particle-number N, can be obtained from free energy function $F(N_k, v)$ of the individual subsystems of volume v,

$$P[\{N_k\}] \simeq \frac{\prod_{k=1}^{\nu} e^{-\beta F(N_k,v)}}{e^{-\beta F(N,V)}} \delta\left(\sum_{k=1}^{\nu} N_k - N\right), \tag{3.1}$$

where N_k is the number of particles in the ith subsystem. The free energy function $F(N,V) = -\ln\{\sum_C \exp[-\beta E(C)]\}$ can in principle be calculated from the Boltzmann weights. The property that the joint subsystem distribution $P[\{N_k\}]$ for a system can be approximately written as a product (i.e., subsystems are almost independent) of individual subsystem weight factors $\exp[-\beta F(N_k)]$ is called additivity property, which remains to be the corner-stone in equilibrium thermodynamics. In equilibrium, additivity thus implies that, provided the interaction is short-ranged, the whole system can be divided into statistically almost independent macroscopic subsystems. In other words, we can define *extensive* observables, for example entropy characterising the distribution for the subsystems, which in the most probable state add up to give the the total entropy characterising the equilibrium distribution for the whole system: $S(E,N) \approx S_1(E_1,N_1) + S_2(E_2,N_2) + \ldots.$ The equivalent statement using the Boltzmann law and equal a-priori probability is, $P[E_1, N_1; E_2, n_2; ...] \simeq \frac{1}{e^{S(E,N)}} e^{S_i(E_i,N_i)}.\delta(E - \sum E_i)\delta(N - \sum N_i)$, which upon maximisation over $\{E_i, N_i\}$ gives the additive form of the total entropy along with the condition of uniform value of the associated intensive thermodynamic parameter, viz. temperature and chemical potential in equilibrium. The 'thermodynamic characterisation' of macroscopic states follow.

We note that the whole structure of equilibrium statistical mechanics could be built precisely because we have the other two laws, the Boltzman law and the eual a priori probability, with which the additivity property could be supplemented. In nonequilibrium we don't have this privilege! For systems having a NESS, there is usually no internal energy function, which can lead to the microscopic probability weights of the steady-state configurations, nor there is any well-defined notion of thermodynamic potentials as in equilibrium. In fact, for most of these nonequilibrium systems, the steady-state weights are not a-priori known and, to find them, one usually requires to explicitly obtain the time-independent solution of the Master equation (here we consider only the systems, which are governed by stochastic Markovian dynamics). Precisely at this stage, the difficulty arises as, in a driven many-particle system, it is often a formidable task to find these detailed microscopic weights.

However, as demonstrated recently in Refs. [14, 50], to characterize the fluctuation properties of a macroscopic quantity, such as the distribution of mass in a large subsystem, one may not actually need to calculate the weights of all microscopic configurations; rather, obtaining coarse-grained probability weights on a larger scale would suffice for this purpose, provided additivity, as discussed below (see Eq. 6.1), holds. It turns out that this sole property, coupled with large deviation principle, gives rise to a robust alternative thermodynamic framework that completely characterises the steady states of a broad class of systems under consideration. Another notable difference with usual ESM formalism is that, in ESM one must have the information about entropy $S(E, N)$ derived from microscopic dynamics, while in the present formalism we require, as the microscopic input, the second moment as a function of the conserved quantity.

3.1. Formal definition and consequences

For characterising the fluctuation of a macroscopic quantity, such as the distribution of mass in a large subsystem, in general we need to know the weights of all microscopic configurations. Additivity asserts that this is not required and, provided some certain conditions are met, we can directly obtain coarse-grained probability weights and characterise the fluctuations uniquely [50, 97].

The property states that, large subsystem masses should be statistically almost independent irrespective of whether the system is in equilibrium or in a NESS. This is physically plausible provided that the subsystems are much larger than the spatial correlation length, so that the boundary correlations between the subsystems could be ignored. Given that this is true and assuming spatial homogeneity in NESS, the joint probability distribution of subsystem masses $\{M_1, M_2, \ldots, M_\nu\}$, to the leading order, can be written in a product form,

$$P[\{M_k\}] \simeq \frac{\prod_{i=1}^{\nu} W_v(M_k)}{Z(M,V)} \delta\left(\sum_{k=1}^{\nu} M_k - M\right), \tag{3.2}$$

where the weight factor $W_v(M_k)$, given by the Boltzman weight in equilibrium, but unknown in more general nonequilibrium states, is assumed to depend only on the respective subsystem mass M_k (spatial homogeneity); $v(>> \xi)$ is the volume of each subsystem, $\nu = \frac{V}{v}$ is total number of subsystems, and the total mass $M = \Sigma_k M_k$ is conserved. In the above equation, the normalization constant, or the partition sum, can be written as

$$Z(M,V) = \left[\prod_k \int dM_k W_v(M_k)\right] \delta\left(\sum_k M_k - M\right) \approx \exp[-V f(\rho)], \tag{3.3}$$

$f(\rho)$ is a nonequilibrium counterpart of free energy density, which can be shown to be related to the large deviation function for the subsystem mass fluctuation in the following subsections. Note that, the following considerations make no distinction or reference to equilibrium or nonequilibrium, and therefore the consequences will be applicable for both.

3.1.1. Nonequilibrium steady states are extensive

Given additivity, existence of free energy like extensive quantities that characterises the distributions is a direct consequence. In the following we give a proof of the second equality in Eq. (3.3) which is a direct and rather intuitive consequence of Eq. (6.1). The generating function for $Z(M,V)$ is given by $\tilde{Z}(s,V) = \int_0^\infty Z(M,V) exp(-sM) dM$. Thus taking the Laplace transform of both sides of the first part of Eq. (3.3), we obtain,

$$\tilde{Z}(s,V) = [\tilde{W_v}(s)]^{\frac{V}{v}}, \tag{3.4}$$

where,

$$\tilde{W_v}(s) = \int_0^\infty W_v(m) exp(-sm) dm.$$

For all functions for which $w(s) \equiv [\tilde{W_v}(s)]^{\frac{1}{v}}$ exists, the partition sum $Z(M,V)$ is calculated by inverting $\tilde{Z}(s,V) = [w(s)]^V$. Now, we can always write $[w(s)] \equiv exp(-\lambda(s))$. This, for V large, proves the second equality of Eq. (3.3), where $f(\rho)$ is the Legendre-Fenchel transform,

$$f(\rho) = \sup_s [sm - \lambda(s)].$$

Therefore we interpret, in lines of ESM,

$$f(\rho) = -\frac{1}{V} lnZ(M,V) \tag{3.5}$$

to be the nonequilibrium free energy density.

It can be further shown that, to leading order, W_v itself is extensive for large subsystem size. From Eq. (3.4),

$$\tilde{W_v}(s) = [\tilde{Z}(s,V)]^{\frac{v}{V}} = [w(s)]^v = exp(-v\lambda(s)).$$

Now since $f(x)$ is the Legendre-Fenchel transform of $\lambda(s)$), for large v the inverse Laplace transform of $\tilde{W_v}(s)$ gives,

$$W_v(m) \simeq exp(-vf(\hat{\rho})),\ \hat{\rho} = m/v \tag{3.6}$$

being the fluctuating local density. This macroscopic characterisation of fluctuating states provide an exact equilibriumlike interpretation of NESS. We see that, the joint subsystem distribution takes the form,

$$P[\{M_k\}] \simeq \frac{\prod_{i=1}^{\nu} exp(-vf(\hat{\rho}))}{exp(-Vf(\rho))} \delta\left(\sum_{k=1}^{\nu} M_k - M\right) \tag{3.7}$$

The product form in Eq. (4.1) amounts to an additivity property analogous to that in equilibrium, in the sense that a free energy function $\sum F(m_k, v)$, with $F = \ln W_v$, is minimized in the (maximum probable state). And indeed in the macrostate the total nonequilibrium free energy is the sum of subsystem free energies, characterised by an equilibriumlike condition of uniform value of a quantity, $\mu(\rho) = \frac{df}{d\rho}$. [1]

While this structure emerging from the properties of almost independent random variables can be regarded as a self-contained and unified thermodynamic description of equilibrium as well as nonequilibrium steady states, this is still a formal structure only. Quantitative prediction of physical observables including the free energy itself requires supplementary information from microscopic dynamics. In this regard we find that, unlike in ESM where fluctuation-response relations arise as 'consequences', the present alternative approach takes up FR relations as indispensable part and parcel of the characterisation of time stationary properties of matter.

3.1.2. Nonequilibrium FR relations and intensive thermodynamic variables

Now, the probability that the mass M_k in the kth subsystem takes values in the interval $(m, m + dm)$ is expressed as $\text{Prob}[M_k \in (m, m + dm)] \equiv P_v(m)dm$, where the probability density function can be formally written as

$$P_v(m) \simeq \frac{1}{\mathcal{Z}} W_v(m) e^{\mu(\rho)m} \tag{3.8}$$

Which is nothing but the marginal distribution. Here $\mu(\rho) = \frac{df}{d\rho}$ is an equilibrium-like chemical potential, $\rho = M/V = \langle M_k \rangle / v$ the mass density and

$$\mathcal{Z}(\mu) = \int_0^\infty W_v(m) e^{\mu m} dm \tag{3.9}$$

the normalization constant. Therefore our task is reduced to find the weight factor $W_v(m)$ and chemical potential $\mu(\rho)$, both of which can be determined from a equilibrium-like fluctuation-response (FR) relation between nonequilibrium compressibility $d\rho/d\mu$ and the

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[1] Also note that, Eqs. (3.5) and (3.6) resemble Boltzmann law.

subsystem particle-number fluctuation,

$$v\frac{d\rho}{d\mu} = \sigma_v^2(\rho), \tag{3.10}$$

which is a direct consequence of the above form of the subsystem distribution as discussed below [48–51]; here, $\sigma_v^2(\rho) = \langle M_k^2\rangle - \langle M_k\rangle^2$ is the variance, or the standard deviation, of mass in the kth subsystem and is a function of density ρ. The above nonequilibrium FR relation has indeed a very close resemblance with the familiar fluctuation dissipation theorem (FDT) in equilibrium, where compressibility is related to particle number fluctuation in a system. At this stage, it is not difficult to see why the quantity $\mu(\rho)$ can be interpreted as an equilibriumlike chemical potential even for systems having a NESS. In fact, the FR relation can be proved using Eqs. (3.8) and (3.9). The proof is as follows. We first note that the mean and the variance of subsystem mass $\langle M_k\rangle$ and $\langle M_k^2\rangle - \langle M_k\rangle^2$, respectively, can be written as

$$\langle M_k\rangle = v\rho = \frac{d\ln\mathcal{Z}}{d\mu}, \tag{3.11}$$

$$\langle M_k^2\rangle - \langle M_k\rangle^2 = \frac{d^2\ln\mathcal{Z}}{d\mu^2}. \tag{3.12}$$

Now taking derivative of Eq. (3.11) w.r.t. chemical potential and then using Eq. (3.12), we obtain the FR as in Eq. (3.10).

3.1.3. Complete thermodynamic characterisation of NESS

As illustrated later in various model systems, the variance of subsystem mass as a function of density can be obtained from the knowledge of two-point spatial correlations of microscopic mass variables at two lattice sites. From the definitions, it follows that, the variance $\sigma_v^2(\rho)$ of subsystem mass $M_k = \sum_{i=0}^{v-1} m_i$ can be written as,

$$\sigma_v^2 = vc_0 + 2(v-1)c_1 + 2(v-2)c_2 + \cdots + 2c_{v-1} \approx v\Sigma_{k=-v}^{v} c_k \tag{3.13}$$

for large v. Here, to write the last step we have used the fact that, on ring the steady state will be spatially homogeneous and the correlation between mass of two sites will depend only on the distance between the sites. Thus, once we calculate two-point spatial correlations of microscopic mass variables at two lattice sites and thereby obtain the functional dependence of the variance $\sigma_v^2(\rho)$ on mass density ρ, we immediately have expressions for chemical potential,

$$\mu(\rho) = \int \frac{1}{\sigma^2(\rho)} d\rho + \alpha, \tag{3.14}$$

where we define scaled variance $\sigma^2(\rho) = \sigma_v^2(\rho)/v$. To calculate the weight factor $w_v(m)$, we proceed as follows. We integrate $\mu(\rho)$ to obtain the nonequilibrium free energy density function,

$$f(\rho) = \int \mu(\rho) d\rho + \alpha\rho + \beta, \tag{3.15}$$

α and β being arbitrary constants of integration. Then, as already explained in section 3.2.1, the Laplace transform of the weight factor,

$$\tilde{W}_v(s) = \int_0^\infty W_v(m) e^{-sm} dm \equiv e^{-\Lambda_v(s)}, \tag{3.16}$$

which is related to the Laplace Transform of $Z(M,V)$, can be obtained from the function $\Lambda_v(s)$ which is Legendre transform of nonequilibrium free energy density [14, 50, 56],

$$\Lambda_v(s) = v[\mathbf{inf}_\rho\{f(\rho) + s\rho\}] = v[f(\rho^*) + s\rho^*], \tag{3.17}$$

where $\rho^*(s)$ is the solution of $s = -f'(\rho^*)$, i.e.,

$$s = -\mu(\rho^*). \tag{3.18}$$

Now, performing inverse Laplace transform of $\tilde{w}_v(s)$, we get the weight factor $W_v(m) \approx exp(-vf(m/v))$ (up to some power law correction) for large subsystem size [14]. Thereafter, substituting $\mu(\rho)$ obtained from Eq. (3.14) in Eq. (3.8), we obtain the probability density function $P_v(m)$ for subsystem mass, and the description is in principle complete.

So, following is the prescription. First, once the stochastic model is defined, calculate the two point correlations. Check whether it is either short-ranged or weak. If it is, then calculate the subsystem variance. Then following the procedures just described, go on to calculate the nonequilibrium chemical potential $\mu(\rho)$ and free energy $f(\rho)$, and finally $W_v(m)$.

In the models, we have calculated the variance $\sigma_v^2(\rho)$ of mass in a subsystem of size v as a function of mass density ρ in a broad class of conserved-mass transport processes. The variance for such mass transport processes usually take the following forms :

$$\begin{aligned} \sigma^2(\rho) &\simeq \rho^{(1-\delta)}, \quad \delta < 0 \text{ and } \neq -1 & (3.19)\\ &\simeq \rho^{(1-\delta)}, \quad \delta = -1 & (3.20)\\ &\simeq (\rho_c - \rho)^{(1-\delta)}, \; \delta > 1, \; \rho_c < \infty & (3.21) \end{aligned}$$

In the first case, using the additivity property, we found that subsystem mass distribution obeys a scaling form,

$$P(m) \sim m^{-\tau} e^{-vf(m/v)+\mu(\rho)m}, \tag{3.22}$$

where $\mu(\rho) \simeq -\rho^{\delta}$ and the two exponents are related by a *scaling relation*, $\delta(\tau - 2) = 1$. Whereas in the third case, an identical scaling form and scaling relation is obtained below ρ_c. However, we can see that, in this case the variance has a multi-pole singularity at this *critical point*, and we will look into it in more detail while studying condensation phenomena.

Interestingly, the borderline case in Eq. (3.20) with $\delta = -1$ generates gamma distribution [50], where the exponents appearing in the distribution does not depend on the power of density ρ in the functional form of the variance but instead depends on the constant of proportionality $1/\eta$. The probability distribution function for subsystem mass has the following form,

$$P_v(m) \propto m^{v\eta-1} e^{-\eta m/\rho}, \tag{3.23}$$

which can be recast in the form of a large deviation function, or a rate function [56],

$$P_v(m) \simeq \text{const.} e^{-vh(m/v)}, \tag{3.24}$$

with $h(x) = -\eta \ln x - \mu x$ and a $1/m$ leading order correction term. Such power laws and gamma distributions are common features in mass transport processes.

Note. Non-conserved systems satisfying additivity : Nonconserved systems, for example a system of particles attached to a particle reservoir of uniform density, allows a equilibrium thermodynamic description. We expect the additivity property to hold for such systems, and for several other models having a NESS. It turns out that, additivity alone cannot characterise such processes completely. However, the consequence can be formulated in lines similar to the conserved processes.

In this case, the joint subsystem mass distribution is given by,

$$\mathcal{P}[\{m_v(i)\}] \simeq \prod_i P_v(m_v(i)), \tag{3.25}$$

$P_v(m)$ being the unknown subsystem mass distribution. Consequently it could be shown that, the *total* mass fluctuation occurs with a large deviation probability,

$$P(M) = \int \delta(M - \textstyle\sum_i m_v(i))\, \mathcal{P}[\{m_v(i)\}] \simeq e^{-Vh(\rho)}.$$

Here ρ, the global density, is a fluctuating quantity and the function $h(\rho)$ could be interpreted as an equilibriumlike grand free energy that resembles (but not necessarily identical to) $\phi(r)$ in section 2.3.1 in the previous chapter. The relation of the total mass distribution and the subsystem mass distribution is explicitly observed in the Laplace space, $\tilde{P}(s,V) = [\tilde{P}(s,v)]^{\nu}$. Given that an *effective* canonical free energy is identified, the density profile distribution could be written of the form of Eq. (2.11), and a nonequilibrium FR relation (Eq. 3.10) would hold for the optimal density.

We note that, since the total mass is not conserved, additional input is required for characterising the distribution of total mass M. Unlike equilibrium, this usually involves the details of the bulk interaction of the system with the reservoir. Once $P(M)$ is known, one can calculate the subsystem mass distribution [2]. In Appendix A., we gave an example of a nonconserved mass aggregation model within mean field; the total mass distribution $P(M)$ could be understood with a detailed balance criterion, and consequently the single site mass distribution, and the condensation phenomena [14] are characterised.

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[2] As mentioned in last chapter, when this bulk mass exchange rule follows some certain constraints [52], then other details of interaction does not affect the nature of bulk fluctuations, and in fact the form of the fluctuation and the FR relation as obtained in the conserved case is preserved. However, for general interactions this does not hold.

4. Power law scaling in nonequilibrium steady states and condensation

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[1] This chapter is based on the paper, Additivity property and emergence of power laws in nonequilibrium steady states, Arghya Das, Sayani Chatterjee, Punyabrata Pradhan, and P. K. Mohanty, Phys. Rev. E **92**, 052107 (2015).

Power laws are ubiquitous in nature [98]. They appear in the distributions of drainage area of rivers [68], droplet size [67, 99], size of clusters formed in polymerization processes [69], rain size [100], size of fragments in fractured solids [101], population and wealth [86, 102], and in stock market fluctuations [103], etc. Power laws, which are usually associated with criticality through emergence of a diverging length scale, are observed evidently in widely unrelated systems, suggesting existence of some broad underlying principle. Recent evidence that living systems, independent of most of the microscopic details, might be operating in the vicinity of a critical regime [104] indeed invoke further questions - how and why systems adapt to near-criticality.

4.1. Emergence of power laws

There have been several attempts to reveal the origin of the power laws in nature, through studies of paradigmatic nonequilibrium models - most appealing being sandpile models [36, 105, 106] and mass aggregation models [107]. Many of these models, where there is a conservation law or, in case of violation, the law is weakly violated in the sense that the systems are slowly driven, are intimately connected to each other. For example, the mass aggregation models [29,108,109] are connected to directed abelian sandpile [110] or to models of river network [68].

In this Letter, we argue that power law distributions in out-of-equilibrium systems can arise simply from additivity property, the tenet of equilibrium thermodynamics. We find that the divergence in the response function is the key. Diverging fluctuations could arise from distributions other than power laws[2]; this is however prohibited if one imposes additivity and consequent fluctuation-response (FR) relation. The response function determines the full scaling form of the distribution, at and away from criticality; critical exponents originate from the singularity in the response function. To demonstrate this, we consider mass aggregation models which are known to have nonequilibrium steady state with scale invariant structures. The distribution function $P_v(m)$ of mass m in a subsystem of volume

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[2] For example, a normalizable distribution $P_v(m) \sim \frac{1}{m_1^*}\Phi_1(\frac{m}{m_1^*}) + \frac{1}{(m_2^*)^2}\Phi_2(\frac{m}{m_2^*})$, near criticality, can have a finite mean $\langle m \rangle = \rho$ but diverging second moment $\langle m^2 \rangle$ for a suitable choice of a finite $m_1^*(\rho)$ and a diverging $m_2^*(\rho) \sim (\rho - \rho_c)^{-n}$; a simple choice could be $\Phi_{1,2}(x) = \exp(-\kappa_{1,2} x^{\delta_{1,2}})$ with $\kappa_{1,2}, \delta_{1,2} > 0$.

v, which is obtained solely from the FR relation at *all* mass densities, has a scaling form $P_v(m) \sim m^{-\tau}\exp(\mu m)$. The quantity $\mu(\rho)$, inverse of a cut-off mass $m^*(\rho) = -1/\mu(\rho)$, is an analogue of equilibrium-like chemical potential and provides a useful thermodynamic interpretation of the emergence of power laws in nonequilibrium systems. We show that the exponent τ and the critical properties of $\mu(\rho)$ arise from a multiple-pole or branch-cut singularity in the variance at a critical mass density ρ_c. As the critical density is approached from below $\rho \to \rho_c^-$, nonequilibrium chemical potential vanishes $\mu(\rho) \to 0$, leading to pure power laws. Beyond the critical density $\rho > \rho_c$, there is a gas-liquid like phase coexistence.

The above result immediately provides answer to why $m^{-5/2}$ power law, at or away from criticality, appears so often in mass aggregation models - especially in higher dimensions - at all densities and irrespective of that the motion of the diffusing masses is biased or not [29, 111, 112]. Interestingly, the same power law appears in k-mer distribution in the classic Flory-Stockmayer [113] theory of polymerization and also in particle number distribution in three dimensional ideal Bose gas near critical point [114], thus *indicating a universality* irrespective of whether the systems are in or out of equilibrium. We show that the $m^{-5/2}$ law is a consequence of a simple-pole singularity in the variance. This analysis is extended also to nonconserved mass aggregation models. We validate the results by explicitly calculating mass distributions in previously studied mass aggregation models and their variants and comparing them with simulations.

Theory. - The instrument of our analysis will be the additivity property introduced in last chapter that a wide class of systems, irrespective of that they are in or out of equilibrium, could possess. Note that, in this chapter the models broadly considered possess discrete mass variables. The application of the consequences of additivity to such mass variables is straightforward. If the subsystems are large (much larger than correlation length), they could be considered statistically almost independent. In that case, the joint subsystem mass distribution in the steady state can be written in a product form,

$$\mathcal{P}[\{m_k\}] \simeq \frac{\prod_{k=1}^{\nu} w_v(m_k)}{Z(M,V)}\delta\left(\sum_k m_k - M\right). \tag{4.1}$$

where $Z = \prod_k \int dm_k w_v(m_k)\delta(\sum_k m_k - M) \equiv \exp[-Vf(\rho)]$ the partition sum, $f(\rho)$ a nonequilibrium free energy density and $\rho = M/V$ mass density. Eq. (4.1) leads to the probability distribution function $\text{Prob}[M_k \in (m, m+dm)] = P_v(m)dm$ for subsystem mass where $P_v(m) = w_v(m)e^{\mu m}/\mathcal{Z}$ with $\mu(\rho)$ a nonequilibrium chemical potential, and $\mathcal{Z}$ the normalization constant. The weight factor $w_v(m)$ and chemical potential $\mu(\rho) = df/d\rho$ can be obtained using a fluctuation-response relation,

$$\frac{d\rho}{d\mu} = \sigma^2(\rho), \tag{4.2}$$

where the scaled variance $\sigma^2(\rho) = (1/v)(\langle M_k^2\rangle - \langle M_k\rangle^2)$. How the FR relation could lead to the complete derivation of the weight factor and the full statistics of the mass distribution is detailed in preceding chapter. In the discrete case, the weight factor can be calculated as $w(m) = (1/2\pi i)\int_C \tilde{w}(z)/z^{m+1}dz$ where $\tilde{w}(z) = \sum_{m=0}^{\infty} z^m w(m)$ is obtained from $\tilde{w}(s)$ by substituting $s = -\ln z$ with C a suitably chosen contour in the complex z-plane ($\tilde{w}(s)$ being the Laplace transform of the weight factor).

Importantly, the distribution function $P_v(m)$ is determined solely by the functional form of the scaled variance $\sigma^2(\rho)$. We argue below that singular response functions generate power law distributions. Other functional form of $P_v(m)$ with diverging moments is also possible, which, we show, however are not allowed if the FR relation holds. We start with a multi-pole singularity at a finite density ρ_c,

$$\sigma^2(\rho) = \begin{cases} \frac{g(\rho)}{(\rho_c-\rho)^n} & \text{for } \rho < \rho_c, \\ \infty & \text{otherwise.} \end{cases} \tag{4.3}$$

This form, with $n > 0$, is relevant in the context of a wide class of mass aggregation models. The analytic part $g(\rho)$ is not particularly relevant in determining the asymptotic form of the distribution $P_v(m)$, however it contributes to the exact form of $P_v(m)$. In fact, other kinds of singularities, such as $\sigma^2(\rho) \sim \log(\rho_c - \rho)(\equiv n = 0)$, $\exp[(\rho_c - \rho)^{-m}](\equiv n \to \infty)$, $1/|\rho - \rho_c|^n$, and the case with $n < 0$ can also arise. One can show that they all lead to power laws.

To analyse the behaviour of $\lambda_v(s)$, the Legendre transform of the free energy density $f(\rho)$, we integrate Eq. (4.2) near $\rho = \rho_c$ and obtain

$$\mu(\rho) \simeq -\frac{(\rho_c - \rho)^{n+1}}{(n+1)g(\rho_c)}\left[1 + \mathcal{O}(\rho - \rho_c)\right] + \alpha, \tag{4.4}$$

which gives $(\rho^* - \rho_c) = [(n+1)g(\rho_c)(s+\alpha)]^{1/(1+n)}$ by using Eqs. (4.28) and (4.4). Integrating $\mu(\rho)$, we get

$$f(\rho) \simeq \frac{(\rho - \rho_c)^{n+2}}{(n+1)(n+2)g(\rho_c)} + \alpha\rho + \beta. \tag{4.5}$$

Note that, this form is valid below ρ_c. Above that, the fluctuation is infinite, implying that the chemical potential is constant α, and the free energy developes a linear branch, thereby losing concavity and leading to phase coexistence, as will be described later. Now, writing $\lambda_v(s) = v[f(\rho^*) + s\rho^*]$, we obtain, in leading order,

$$\lambda_v(s) \simeq v[a_0 + a_1(s+\alpha) + a_2(s+\alpha)^{\frac{n+2}{n+1}}], \tag{4.6}$$

where a_0, a_1, a_2 are constants. Thus, $\tilde{w}(s) = \exp[-\lambda_v(s)] \simeq \text{const.} \times [1 + va_1(s+\alpha) + va_2(s+\alpha)^{1+1/(1+n)}]$, implying $w(m) \sim \exp(-\alpha m)/m^\tau$, with $2 < \tau = [2 + 1/(1+n)] < 3$, for *small* v and large subsystem masses $m \gg v$. This translates into the mass distribution having a scaling form

$$P_v(m) \propto \frac{1}{m^\tau} e^{\tilde{\mu}(\rho)m}. \tag{4.7}$$

On the other hand, for very large subsystem size, following Eq. (3.22), the steady state subsystem mass distribution in the limit of large subsystem mass takes the form,

$$P_v(m) \propto \frac{1}{m^\tau} e^{-vf(m/v) + \tilde{\mu}(\rho)m}, \tag{4.8}$$

where $\tilde{\mu}(\rho) = \int_{\rho_c}^{\rho} 1/\sigma^2(\rho) d\rho = \mu(\rho) - \mu(\rho_c)$ is a chemical potential, inverse of which gives a cut-off $m^* = 1/\tilde{\mu}$ in the distribution. Later, we explicitly calculate $\tilde{\mu}(\rho)$ in specific cases. Note that $\tilde{\mu}(\rho_c) = 0$ at *critical point* $\rho = \rho_c$ and consequently $P_v(m)$ becomes a pure power law. Moreover, by defining a critical exponent $\delta = 1 + n$ as $\tilde{\mu}(\rho) \sim (\rho_c - \rho)^\delta$, we get a scaling relation $\delta(\tau - 2) = 1$.

Power laws without singular variance : It is instructive to consider the case $n = 0$ (Eq.

4.3) where the variance diverges as $\sigma^2(\rho) \sim \rho^{1-\delta}$ at large densities, with $\delta < -1$. Using Eq. (4.2), we get $\mu(\rho) \sim \rho^{\delta}$ (we set integration constant $\alpha = 0$ without loss of any generality) and consequently $\tilde{w}(s) \simeq a_0 + a_1 s^{1+1/\delta}$ which leads to the mass distribution having a scaling form $P_v(m) \propto m^{-\tau} h(m\rho^{\delta})$ where the scaling function $h(x) = \exp(-x)$ and power law exponent $\tau = 2+1/\delta$ with $1 < \tau < 2$ as $\delta < -1$, leading to a scaling relation $\delta(\tau-2) = 1$. This scaling form was numerically observed in mass aggregation models with mass-dependent diffusion [30]. Note that the case with $\delta = -1$ generates gamma distributions [50].

4.2. Thermodynamic characterisation of condensation transition

The form of the variance as in Eq. (4.3) indeed have broad implications, not only in conserved mass transport processes but also in the nonconserved versions (Appendix A.). Note that, for $n > 0$ the FR relation in Eq. (4.2) implies that the free energy density $f(\rho)$ is not a strictly concave function of ρ and has a linear branch of slope $\mu(\rho_c)$ for $\rho \geq \rho_c$. Moreover, $f''(\rho = \rho_c) = \mu'(\rho = \rho_c) = 0$ (prime denotes derivative w.r.t. ρ) implies a point of inflection in $f - \rho$ curve at $\rho = \rho_c$. Consequently, the Legendre transform of $\lambda_v(s)$ develops a singularity in the nth order derivative at $s = s_c$.

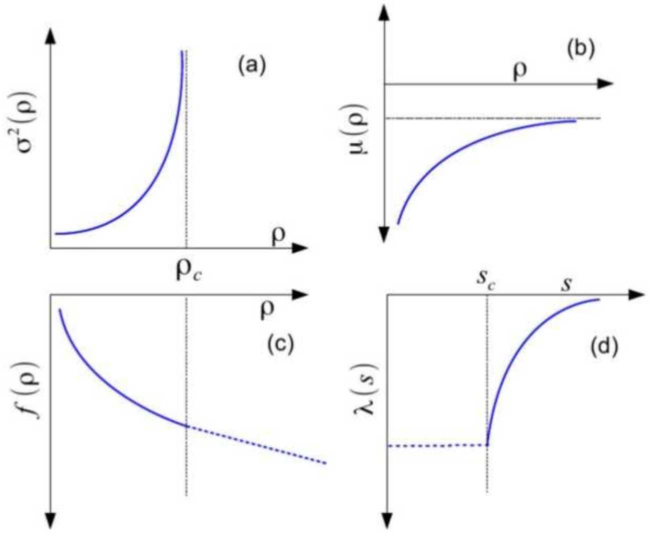

Figure 4.1.: Schematic representation of condensation transition: Panel (a) - $\sigma^2(\rho)$ as a function of ρ, panel (b) - $\mu(\rho)$ as a function of ρ, panel (c) - $f(\rho)$ as a function of ρ, and panel (d) - $\lambda(s)$ as a function of s.

This construction of a nonequilibrium free energy function $f(\rho)$ from a general thermodynamic consideration readily explains the phase coexistence between a fluid and a condensate, as observed in the past in many out-of-equilibrium systems. Indeed, the FR relation imply that, beyond criticality, the *excess* macroscopic fraction of total mass amounting to $V(\rho - \rho_c)$ aggregates in a single site (due to dominance of diffusion), and the chemical potential must equalise for the condensate carrying site and the rest of the fluid which remains in the critical state, implying that a thermodynamic Maxwell construction is valid as graphically presented in Fig. 4.1. Note that, when $n = 0$, there cannot be any condensation transition as there is no singularity in the variance at any finite density.

4.2.1. Models and illustration

We now substantiate the above claims in a broad class of nonequilibrium model systems which were studied intensively in the last couple of decades. Now onwards, for the purpose of illustrations, we specifically consider the case with $n = 1$ and $v = 1$ (i.e., factorisation of steady state mass distribution at a single site level, which we call the mean field description). We define a generalized version of conserved mass aggregation models (CMAM) studied in [29,107,115], for simplicity on a one dimensional lattice of V sites, where masses (discrete) diffuse, fragment and coalesce stochastically with nearest-neighbour masses according to the following dynamical rules: (1) diffusion of mass m_i at site i to $i \pm 1$ with mass-dependent rate $D(m_i)$ where $m_i \to 0$ and $m_{i\pm1} \to m_{i\pm1} + m_i$ and (2) fragmentation of a discrete mass Δ at site i, provided $\Delta \leq m_i$, and coalescence of the mass to either of the sites $i\pm1$ with mass-dependent rate $w(\Delta)$ where $m_i \to m_i - \Delta$ and $m_{i\pm1} \to m_{i\pm1} + \Delta$ with $\Delta = 1, 2, \dots$. Total mass $M = \sum_{i=1}^{V} m_i$ is conserved in this process. It has been argued in the second chapter that, although there is no average mass current in the steady state, these models, in presence of diffusion ($D \neq 0$), violate the Kolmogorov criterion for equilibrium and are inherently far from equilibrium. Even for these simple dynamical rules, the steady state weight in general is therefore not exactly known. However, we have numerically checked two-point spatial correlations to be small ($\sim 1/V$), and the additivity property as in Eq. (4.1), to a good approximation, is expected to hold.

We calculate the variance $\sigma^2(\rho)$ of mass at a single site in various special cases, using

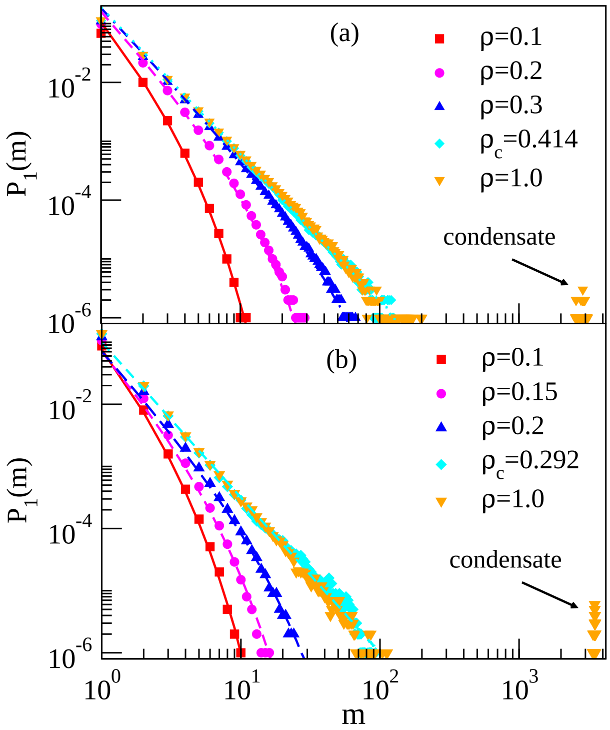

Figure 4.2.: Single site distribution (points - simulations) in conserved mass aggregation models is compared with analytic expression in Eq. (4.7) (lines - theory). Panel (a) - $w_1 = 1$, $w_2 = 0$ and panel (b) - $w_1 = 0$, $w_2 = 1$. System size $L = 5000$.

the additivity property Eq. (4.1) with $v = 1$. We take diffusion rate $D(m_i) = 1$, independent of mass m_i, $w(\Delta = 1) = w_1$ (rate of single particle chipping), $w(\Delta = m_i - 1) = w_2$ (rate of all-but-one particle chipping) and $w(\Delta) = 0$ otherwise. Case I. - For $w_1 = 1$ and $w_2 = 0$, the model becomes the one studied in [29]. For $\rho \leq \rho_c$, using additivity property, we exactly calculated the variance and consequently chemical potential with the critical density $\rho_c = \sqrt{2} - 1$ For large mass, the mass distribution function is calculated to be $P_1(m) \propto m^{-5/2} \exp[(\mu(\rho) - \mu(\rho_c))m]$. In top panel of Fig. 4.2 we plot $P_1(m)$ obtained from simulations for various values of ρ and compared them with the analytical results which are in a very good agreement with the simulation results. Regarding the subsystem mass distribution $P_v(m)$ for large v, we find the same power law tail with $\tau = 5/2$ for $m \gg v$. In top panel of Fig. 4.3, we plot $P_v(m)$ obtained from simulations and compared them with the analytical results, showing a quite good agreement.

Case II. - Similarly for $w_1 = 0$ and $w_2 = w$ and $\rho \leq \rho_c$, the variance and chemical potential can be exactly obtained using the additivity property The critical density in this case is $\rho_c =$

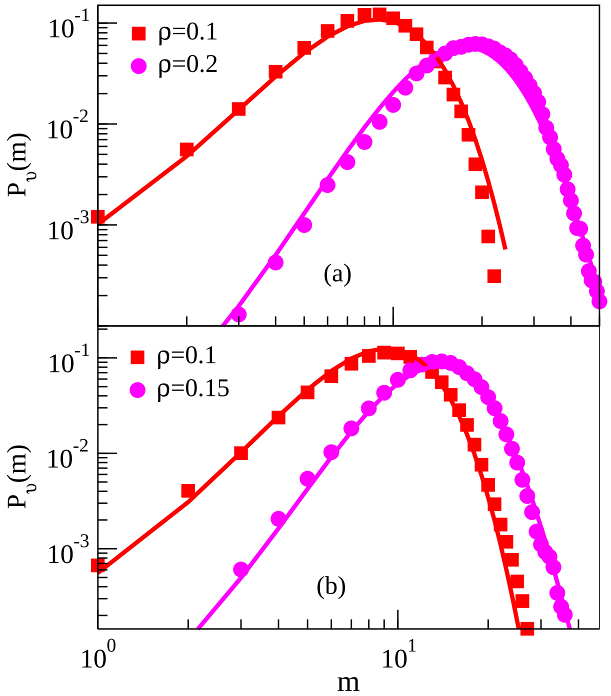

Figure 4.3.: Subsystem mass distribution functions Pv (m) (points represent simulations) in CMAMs compared with the analytic expression in Eq. (4.8) (lines represent theory) for various densities and cutoff mass (a) mass chipping rates w1 = 1 and w2 = 0 and a 20 and (b) mass chipping rates w1 = 0 and w2 = 1 and cutoff mass a 25. In all cases, the mass diffusion rate D = 1, the system size L = 105 , and the subsystem size v = 100

$1-1/\sqrt{2}$. In the bottom panels of Figures 4.2 and 4.3, we see that the simulation results agree remarkably well with the analytical scaling forms in Eqs. (4.7) and (4.8). In simulations, we have also studied various other cases (all with $D = 1$): Case III. - $w(\Delta = 1) = w_1$, $w(\Delta = 2) = w_2$ and $w(\Delta) = 0$ otherwise, Case IV. - a discrete-mass model with $w(\Delta) = \exp(-\Delta)$ and Case V. - a continuous-mass model with $w(\Delta) = \exp(-\Delta)$. In these cases, in the absence of an analytical expression of $\sigma^2(\rho)$, we checked in simulations (Fig. 4.4) that the variance indeed has the same behaviour near critical point $\sigma^2 \sim (\rho_c - \rho)^{-n}$, with $n = 1$,

which therefore leads to the same power law exponent $\tau = 5/2$. For continuous-mass model in Case V. we demonstrate in Fig. 4.5 this power law exponent for the distribution of large mass at a single site level, and the critical point and condensation.

Interestingly, the same exponent $\tau = 5/2$ appears also in the distribution of particle numbers in ideal Bose gas in three dimensions (3D) near the critical point where Bose-Einstein condensation occurs. This could be easily understood from the fact that particle-number fluctuation in the case of 3D Bose gas has the same critical behaviour $\sigma^2(\rho) \sim (\rho_c - \rho)^{-n}$,

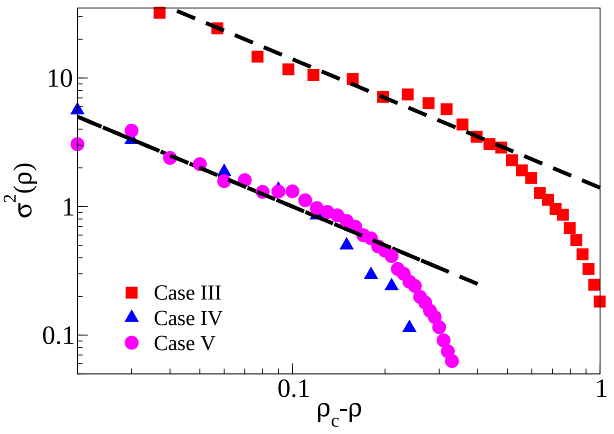

Figure 4.4.: $\sigma^2(\rho)$ *vs.* $(\rho_c - \rho)$. Black line is const. $\times (\rho_c - \rho)^{-n}$ with $n = 1$. Red rectangles are for Case III (one and two particle fragmentation), blue triangles are for Case IV [Δ is discrete with fragmentation rate $w(\Delta) = \exp(-\Delta)$] and red circles are for case V [Δ is continuous with fragmentation rate $w(\Delta) = \exp(-\Delta)$]. Diffusion rate $D = 1$ throughout.

with $n = 1$, as in these 'mean-field' nonequilibrium systems having negligible spatial correlations. That, on a mean-field level, the nonequilibrium aggregation models belong to the universality class of equilibrium Bose gas in 3D, so far has not been realized.

4.2.2. Details of the calculation for specific models

Mass Aggregation Models (CMAM):

We define here a class of conserved mass aggregation models (CMAM) on a one dimensional lattice with periodic boundary and calculate the variance of mass at a single site in the steady state, assuming that the additivity property holds at a single site level. For, simplicity, we consider only the discrete mass case.

The mass at each site undergoes either diffusion (where whole of the mass is transferred to either of neighbouring sites) or chipping, with certain transition rates; in the models considered below, there are two types of chipping process. The diffusing mass or the chipped-off mass are coalesced with the mass at either of the neighbouring sites with a pre-assigned rates. In this process, the total mass of the system is conserved.

Provided a site i is occupied, particles hop to either of the nearest neighbour sites accord-

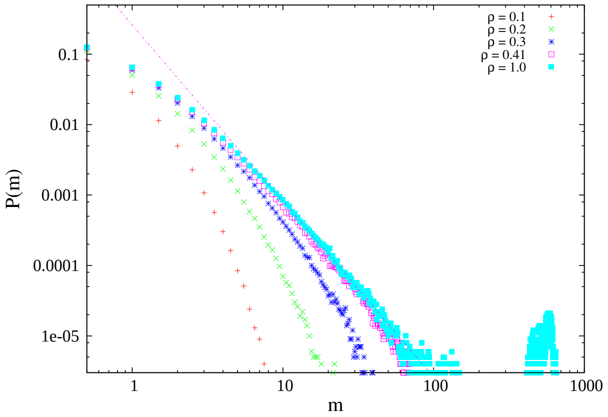

Figure 4.5.: Single site mass distribution at different densities and condensation in case V [Δ is continuous with fragmentation rate $w(\Delta) = \exp(-\Delta)$]. Diffusion rate $D = 1$.

ing to the transition rates specified below.

A. Diffusion with rate 1: All particles at a site i hop with rate 1 to left or right, i.e., $m_i \to 0$ and $m_{i\pm1} \to m_{i\pm1} + m_i$.

B. Chipping with rate w_1: This chipping process involves a particle at site i being chipped off and thrown to left or right neighbour, i.e., $m_i \to (m_i - 1)$ and $m_{i\pm1} \to m_{i\pm1} + 1$.

C. Chipping with rate w_2: This chipping process involves $m_i - 1$ particles going to either left or right neighbour and the rest of the particles remaining at site i, i.e., $m_i \to 1$ and $m_{i\pm1} \to m_{i\pm1} + m_i - 1$.

Loss terms at site i:

$$m_i(t+dt) = 0 \qquad \text{with probability } (dt) \tag{4.9}$$

$$= m_i(t) - 1 + \delta_{m_i(t),0} \qquad \text{with probability } (w_1 dt) \tag{4.10}$$

$$= 1 - \delta_{m_i(t),0} \qquad \text{with probability } (w_2 dt) \tag{4.11}$$

Gain terms from $(i-1)^{th}$ site:

$$\begin{aligned} m_i(t+dt) &= m_i(t) + m_{i-1}(t) && \text{with probability } (dt/2) && (4.12)\\ &= m_i(t) + 1 - \delta_{m_{i-1}(t),0} && \text{with probability } (w_1 dt/2) && (4.13)\\ &= m_i(t) + m_{i-1}(t) - 1 + \delta_{m_{i-1}(t),0} && \text{with probability } (w_2 dt/2) && (4.14) \end{aligned}$$

Gain terms from $(i+1)^{th}$ site:

$$\begin{aligned} m_i(t+dt) &= m_i(t) + m_{i+1}(t) && \text{with probability } (dt/2) && (4.15)\\ &= m_i(t) + 1 - \delta_{m_{i+1}(t),0} && \text{with probability } (w_1 dt/2) && (4.16)\\ &= m_i(t) + m_{i+1}(t) - 1 + \delta_{m_{i+1}(t),0} && \text{with probability } (w_2 dt/2) && (4.17) \end{aligned}$$

Mass remains unchanged at site i:

$$m_i(t+dt) = m_i(t) \qquad \text{with probability } (1 - 2dt - 2w_1 dt - 2w_2 dt) \qquad (4.18)$$

Define $\langle (1-\delta_{m_j,0}) \rangle = \mathcal{S}(\rho)$, the probability that a site is occupied. We deal with steady state throughout. We assume that the additivity property is valid at single site level and therefore n- point ($n \geq 2$) correlation factorizes.

***n*-th moment equation:** The n-th moment $\langle m_i^n \rangle$ can be calculated in the steady state where $\langle m_i^n(t+dt) \rangle = \langle m_i^n(t) \rangle$ as given below,

$$\begin{aligned} \langle m_i^n(t+dt) \rangle = \langle m_i^n(t) \rangle &= \langle [m_i(t) - 1 + \delta_{m_i(t),0}]^n \rangle w_1 dt + \langle [1 - \delta_{m_i(t),0}]^n \rangle w_2 dt + \langle [m_i(t) + m_{i-1}(t)]^n \rangle \frac{dt}{2}\\ &+ \langle [m_i(t) + 1 - \delta_{m_{i-1}(t),0}]^n \rangle w_1 \frac{dt}{2} + \langle [m_i(t) + m_{i-1}(t) - 1 + \delta_{m_{i-1}(t),0}]^n \rangle w_2 \frac{dt}{2} + \langle [m_i(t) + m_{i+1}(t)]^n \rangle \frac{dt}{2}\\ \langle [m_i(t) + 1 - \delta_{m_{i+1}(t),0}]^n \rangle w_1 \frac{dt}{2} &+ \langle [m_i(t) + m_{i+1}(t) - 1 + \delta_{m_{i+1}(t),0}]^n \rangle w_2 \frac{dt}{2} + \langle m_i^n(t) \rangle (1 - 2dt - 2w_1 dt - 2w_2 dt). \end{aligned}$$

However, we are only interested in calculating the variance, which can be done as follows.

2^{nd} moment equation: If we put $n = 2$ in the above equation, the second moment $\langle m_i^2 \rangle$

however cancels out from the above equation. Using factorization of two-point correlation, i.e., $\langle m_i m_j \rangle \approx \rho^2$ for $i \neq j$, we get an expression for the occupation probability $\mathcal{S}$ as a function of mass density ρ,

$$\rho^2(1+w_2) = (w_1+w_2)(\rho - \mathcal{S}) - (w_1 - w_2)\rho\mathcal{S} \tag{4.19}$$

which, after simplification, gives

$$\mathcal{S}(\rho) = \frac{(w_1+w_2)\rho - (1+w_2)\rho^2}{(w_1+w_2)+(w_1-w_2)\rho}. \tag{4.20}$$

3$^{\text{rd}}$ moment equation: Similarly for $n = 3$, we obtain an equation where the third moment $\langle m_i^3 \rangle$ cancels out and we actually get, using factorization of both two-point and three-point correlation, a relation for the second moment

$$\langle m^2 \rangle = \rho \frac{(w_1+w_2)(1+\mathcal{S}) - 2w_2\rho}{(w_1+w_2) - 2(1+w_2)\rho - (w_1-w_2)\mathcal{S}} \tag{4.21}$$

Using the expression of occupation probability in Eq. (4.20), we obtain

$$\langle m^2 \rangle = \rho \frac{(w_1+w_2)^2 + 2(w_1^2 - w_2^2)\rho - (w_1+w_2+3w_1w_2-w_2^2)\rho^2}{(w_1+w_2)^2 - 2(w_1+w_2)(1+w_2)\rho - (w_1-w_2)(1+w_2)\rho^2} \tag{4.22}$$

which leads to the desired expression of the variance as a function of density,

$$\sigma^2(\rho) = \rho \frac{(w_1+w_2)^2 + (w_1+w_2)(w_1-3w_2)\rho + (w_1+w_2-w_1w_2+3w_2^2)\rho^2 + (w_1-w_2)(1+w_2)\rho^3}{(w_1+w_2)^2 - 2(w_1+w_2)(1+w_2)\rho - (w_1-w_2)(1+w_2)\rho^2}. \tag{4.23}$$

The variance $\sigma^2(\rho)$ has a *singularity* at $\rho = \rho_c$, i.e., it diverges at a critical density $\rho = \rho_c$. The critical density can be obtained by solving the following equation

$$(w_1+w_2)^2 - 2(w_1+w_2)(1+w_2)\rho_c - (w_1-w_2)(1+w_2)\rho_c^2 = 0, \tag{4.24}$$

i.e., by putting the denominator of Eq. (4.23) zero. This gives a *simple pole* at the critical density

$$\rho_c = \frac{\sqrt{1+\frac{w_1-w_2}{1+w_2}} - 1}{\frac{w_1-w_2}{w_1+w_2}}. \tag{4.25}$$

Nonequilibrium free energy function can be calculated by integrating nonequilibrium chemical potential w.r.t. density ρ

$$\mu(\rho) = \frac{df}{d\rho} \Rightarrow f(\rho) = \int \mu(\rho) d\rho. \tag{4.26}$$

The function $\lambda_v(s) = -\ln \tilde{w}(s)$, which is the Legendre transform of the free energy density $f(\rho)$, can be obtained as given below,

$$\lambda_v(s) = v[f(\rho^*) + s\rho^*], \tag{4.27}$$

where ρ^* is the solution of

$$s = -\mu(\rho^*). \tag{4.28}$$

I: CMAM with $w_1 = 1, w_2 = 0$ For $w_1 = 1, w_2 = 0$, i.e., the model studied in [29] by generating function method, we obtain the variance as given below

$$\sigma^2(\rho) = \frac{\rho(1+\rho)(1+\rho^2)}{(1-2\rho-\rho^2)} = \frac{\rho(1+\rho)(1+\rho^2)}{(\rho_c-\rho)(\sqrt{2}+1+\rho)} \quad \text{with } \rho_c = (\sqrt{2}-1), \tag{4.29}$$

for which one can obtain a chemical potential $\mu(\rho)$ and free energy function $f(\rho)$, by integrating the fluctuation-response relation,

$$\begin{aligned} \mu(\rho) &= \int \frac{1}{\sigma^2(\rho)} d\rho \\ &= -2\tan^{-1}\rho + \ln\left(\frac{\rho}{1+\rho}\right) + \alpha \end{aligned} \tag{4.30}$$

and, upon one more integration,

$$\begin{aligned} f(\rho) &= \int \mu(\rho) d\rho \\ &= -2\rho\tan^{-1}\rho + \rho\ln\left(\frac{\rho}{1+\rho}\right) - \ln\left(\frac{1+\rho}{1+\rho^2}\right) + \alpha\rho + \beta \end{aligned} \tag{4.31}$$

where α and β are two arbitrary constants of integration.

II: CMAM with $w_1 = 0, w_2 = 1$ For $w_1 = 0, w_2 = 1$, we obtain the variance

$$\sigma^2(\rho) = \frac{\rho(1-\rho)(2\rho^2-2\rho+1)}{2\rho^2-4\rho+1} = \frac{\rho(1-\rho)(2\rho^2-2\rho+1)}{(\rho_c-\rho)(2+\sqrt{2}-2\rho)}. \tag{4.32}$$

There is a simple pole at the critical density $\rho_c = 1 - 1/\sqrt{2}$. By integrating fluctuation-response relation, we get

$$\mu(\rho) = 2\tan^{-1}(1-2\rho) - \ln\left[\frac{1}{2\rho(1-\rho)} - 1\right] \tag{4.33}$$

$$f(\rho) = 2\rho\tan^{-1}(1-2\rho) + (1-\rho)\ln\rho[1+(1-2\rho)^2] + \rho\ln(1-2\rho). \tag{4.34}$$

Mass distribution and phase coexistence:

The results above suggests that, for systems having no upper bound of mass at a single site, the competition between diffusion that tends to aggregate the mass at one site and chipping that tends to break it leads to the condensation transition at a finite critical density. Given an additivity property holds, this phenomena has a thermodynamic explanation in terms of a chemical potential and Maxwell construction of nonconcave nonequilibrium free energy, as explained earlier and shown in Fig. 4.1.

In subsequent calculations, we provide the essential steps to obtain single-site (i.e., $v = 1$) mass distribution function $P_1(m) \propto w_1(m)\exp[\mu(\rho)m]$ where $w_1(m)$ is the single-site weight factor and $\mu(\rho)$ is a chemical potential. We first analyse the behaviour of $\lambda_1(s)$ near the singularity at $s = s_c$ by expanding $\mu(\rho)$ and $f(\rho)$ near critical density in the power series of $\rho - \rho_c$ where $\rho - \rho_c < 0$ is small,

$$\mu(\rho) = \mu(\rho_c) + \frac{\mu''(\rho_c)}{2}(\rho-\rho_c)^2 + \ldots \tag{4.35}$$

$$f(\rho) = f(\rho_c) + \mu(\rho_c)(\rho-\rho_c) + \frac{f'''(\rho_c)}{3!}(\rho-\rho_c)^3 + \ldots \tag{4.36}$$

where we have used Eq. (4.26) and $\mu'(\rho_c) = f''(\rho_c) = 0$ (see Fig. 4.1). Using Eq. (4.28) in Eq. (4.35) where $s + \mu(\rho_c) \simeq -\mu''(\rho_c)(\rho^* - \rho_c)^2/2$, we get

$$(\rho^* - \rho_c) = -\sqrt{\frac{2}{|\mu''(\rho_c)|}}(s - s_c)^{1/2} \tag{4.37}$$

where $s_c = -\mu(\rho_c)$ and $\mu''(\rho_c) < 0$. Therefore $\lambda_1(s) = f(\rho^*) + s\rho^*$ near $s = s_c$, in the leading order of $(s - s_c)$, can be approximated as

$$\begin{aligned} \lambda_1(s) &\simeq \left[f(\rho_c) - s_c(\rho^* - \rho_c) + \frac{f'''(\rho_c)}{3!}(\rho^* - \rho_c)^3 \right] + s\rho^* \\ &= \lambda_1(s_c) + \rho^*(s - s_c) + \frac{f'''(\rho_c)}{3!}(\rho^* - \rho_c)^3 \\ &= \left[a_0 + a_1(s - s_c) + a_2(s - s_c)^{3/2} \right] \end{aligned} \tag{4.38}$$

where $a_0 = \lambda_1(s_c) = f(\rho_c) + s_c\rho_c$, $a_1 = \rho_c$ and $a_2 = -(2/3)\sqrt{2/|\mu''(\rho_c)|}$. The inverse Laplace transform of the weight factor $w_1(m)$ can be written as

$$\tilde{w}_1(s) = e^{-\lambda_1(s)} \simeq e^{-a_0}[1 - a_1(s - s_c) - a_2(s - s_c)^{3/2}] \tag{4.39}$$

which, for $m \gg 1$, translates into

$$w_1(m) \sim \frac{e^{s_c m}}{m^{5/2}}. \tag{4.40}$$

Consequently the mass distribution can be written as

$$P_1(m) \sim \frac{e^{s_c m}}{m^{5/2}} e^{\mu(\rho)m} = \frac{e^{-(\alpha + \mu_0(\rho_c))m}}{m^{5/2}} e^{(\mu_0(\rho) + \alpha)m} \tag{4.41}$$

$$P_1(m) \sim \frac{1}{m^{5/2}} e^{[\mu_0(\rho) - \mu_0(\rho_c)]m}. \tag{4.42}$$

Note that effective chemical potential $\tilde{\mu}(\rho) = \mu_0(\rho) - \mu_0(\rho_c)$ is zero at the critical density $\rho_c = (\sqrt{2} - 1)$. The mass distribution in Eq. (4.42) is precisely what was found in [29] at $\rho = \rho_c$ and describes the simulation data remarkably well (see Fig. 4.2).

4.3. Concluding remarks

In this chapter we argued that an additivity property can explain why simple power-law scaling appears generically in nonequilibrium steady states with short-range correlations. We demonstrate that the existence of a fluctuation-response relation, a direct consequence of additivity, with a singular response function leads to power-law distributions with non-trivial exponents. The simplest form of the singularity, a simple pole, gives rise to the exponent 5/2, which was often observed in the past in apparently unrelated systems. We substantiate the claims by analytically calculating the response function, which diverges as critical point is approached, in paradigmatic nonequilibrium mass aggregation models and the corresponding single-site and subsystem mass distributions. Most remarkably, the analysis, being independent of dynamical rules in a particular system, equally extends to critical properties in equilibrium and nonequilibrium.

Thermodynamic characterization of phase coexistence in driven systems is a fundamental problem in statistical physics and has been addressed in the past [27,28,39–43], either numerically or analytically for exactly known steady states mostly having a product measure. From that perspective, it is quite encouraging that, even when steady-state weights are a priori not known, our analytical method not only gives insights into the steady-state structure but can also be applied to identify a chemical potential, which equalizes in the coexisting phases and whose vanishing at criticality gives rise to pure power laws.

Note that, in our formulation, the mass distribution functions, though approximate, have been calculated solely from the knowledge of the variance. This formulation is perhaps not surprising in equilibrium where the free energy function (or entropy, for an isolated system) essentially determines the fluctuation properties of a system. However, in the nonequilibrium scenario, it is a priori not clear that such an equilibrium thermodynamic approach can indeed be applied in systems having a steady state with nontrivial spatial structure. Here it is worth mentioning that one requires, in principle, all the moments to specify a probability distribution function. However, the additivity property, provided it holds, puts a strong constraint on the mass distribution function Pv (m) through a fluctuation-response relation and thus helps to uniquely determine Pv (m), only from the knowledge of the variance as a function of density.

We believe that our analysis, being based on a general thermodynamic principle, would be applicable in many other driven systems where phase coexistence is known to occur (e.g., in active matters [44,45]). As a concluding remark, we mention that the additivity property is expected to be quite generic for systems having short-range correlations and therefore it would be indeed interesting to actually verify additivity, through the predictions concerning fluctuations, on a case-by-case basis. Also, it remains to be seen whether the principle of additivity can be extended to systems having long-range spatial correlations, at least in the cases where the strength of these correlations is weak.

5. Spatial correlations and subsystem mass distributions in mass transport processes

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[1] This chapter is based on the paper, Spatial correlations, additivity and fluctuations in conserved-mass transport processes, Arghya Das, Sayani Chatterjee, and Punyabrata Pradhan, Phys. Rev. E **93**, 062135 (2016).

5.1. Why two-point correlation

Characterizing spatial structure in interacting many-particle systems having a nonequilibrium steady state (NESS) is a fundamental problem [24, 25, 116], though a difficult one, in statistical physics. In fact, the difficulty arises primarily because the exact steady-state weights of microscopic configurations, in most cases, are not known. In this chapter, we study a broad class of one dimensional conserved-mass transport processes involving chipping, diffusion and coalescence of continuous mass variables and demonstrate that the processes possess, quite remarkably, an equilibriumlike thermodynamic structure.

Though dynamical rules governing these processes are quite simple, they can give rise to nontrivial spatial structure in the steady state. In fact, even in one dimension which we consider in this chapter, the exact steady-state weights, except for a few cases [27,28,31,83–85], are not known. Notwithstanding the difficulty in obtaining the exact steady-state weights, there have been some progress in the past in calculating the two-point correlations in a few specific model systems [26–28, 82, 85]. However, the spatial correlations for generic parameter values are still mostly unexplored. Moreover, another important quantity in these processes, the subsystem mass distribution when the subsystem size is large, or equivalently the large deviation function for subsystem mass, has not been studied when there are finite spatial correlations in the systems; single-site mass distributions have been calculated in the past, though for systems having a product-measure steady state [27, 83] or within mean field theory [28, 31, 80].

Here, we characterize the steady-state spatial structure of these conserved-mass transport processes, by exactly calculating the two-point spatial (equal-time) correlations between masses at any two sites. Moreover using the additivity property and corresponding fluctuation-response (FR) relation, the knowledge of only the two-point correlation functions is sufficient for obtaining the probability distribution function of mass in a subsystem, which is much larger than the spatial correlation length in the system (as detailed in chapter 3). In other words, in the conserved-mass transport processes, we use the additivity formulation to actually calculate the large deviation probability of subsystem masses. In lines to the discussions in chapter 3, the logarithm of the large deviation probability can be interpreted as a nonequilibrium free energy function, which governs the density fluctuations in these nonequilibrium processes and thus immediately connects to the standard statistical mechanics framework.

The organization of the rest of the chapter is as follows. In next subsection we discuss how the additivity can be used to obtain subsystem mass distributions, in particular the gamma distribution widely observed in nonequilibrium mass-transport processes mentioned above. In section 5.2, we exactly calculate the two-point spatial correlations in the three variants of the mass chipping models (MCMs) - model IV (MCM IV) in section III.A, model I (MCM I) in section III.B and model II (MCM II) in Sec. III.C. In next section, we calculate two-point spatial correlations in model III, the mass exchange model (MEM) and then we summarize with some concluding remarks.

5.1.1. Additivity and gamma distribution

In chapter 3, we explained how additivity property can be used to calculate steady state subsystem mass distribution when the subsystem size is large, much larger than the static correlation length, irrespective of whether the system is in or out of equilibrium. We have, in fact, used the methods to successfully explain the widely observed scaling forms, and the phase coexistence, in a thermodynamic framework, and demonstrated for models assuming additivity to be valid even at a single site level. In this chapter we encounter, as we shall see, models having a correlation, and at present there is no hope to know the microscopic configuration. The challenge is to still characterise the macroscopic distributions of these models with the knowledge of the correlations, as we have claimed in chapter 3. To recapitulate briefly, the variance of subsystem mass as a function of density can be obtained from the knowledge of two-point spatial correlations of microscopic mass variables at two lattice sites, and provided additivity holds, the variance as a function of mass density is enough to characterise the mass distribution completely.

In the subsequent sections, we calculate the variance $\sigma_v^2(\rho)$ of mass in a subsystem of size v as a function of mass density ρ in a broad class of conserved-mass transport processes. Interestingly, in all these cases, we find that the variance $\sigma_v^2(\rho)$ of subsystem mass has the following functional dependence on mass density ρ,

$$\sigma_v^2(\rho) = v\frac{\rho^2}{\eta}, \tag{5.1}$$

i.e., the variance of subsystem mass is proportional to the square of mass density, where the factor η depends on microscopic parameters of the particular model systems. In that case, chemical potential and free energy density can be immediately obtained from Eqs. (3.14) and (3.15),

$$\mu(\rho) = -\frac{\eta}{\rho} + \alpha, \tag{5.2}$$

$$f(\rho) = -\eta \ln \rho + \alpha\rho + \beta, \tag{5.3}$$

which, using Eqs. (3.16), (3.17) and (3.18), respectively, lead to the following expressions,

$$s = \frac{\eta}{\rho^*} - \alpha, \tag{5.4}$$

$$\Lambda_v(s) = \text{const.} + \ln\left[(s+\alpha)^{\eta v}\right], \tag{5.5}$$

$$W_v(s) = \text{const.}(s+\alpha)^{-v\eta}. \tag{5.6}$$

Now, performing inverse Laplace transform of $W_v(s)$, we obtain the weight factor,

$$W_v(m) = \text{const.} m^{v\eta-1} e^{-\alpha m}, \tag{5.7}$$

and the corresponding probability distribution function for subsystem mass,

$$P_v(m) \propto m^{v\eta-1} e^{-\eta m/\rho}, \tag{5.8}$$

which has the form of gamma distribution. The above subsystem mass distribution can be immediately recast as given below,

$$P_v(m) \simeq \text{const.} e^{-vh(m/v)}, \tag{5.9}$$

in the form of a large deviation function, or a rate function [56], $h(x) = -\eta \ln x - \mu x$.

5.2. Models

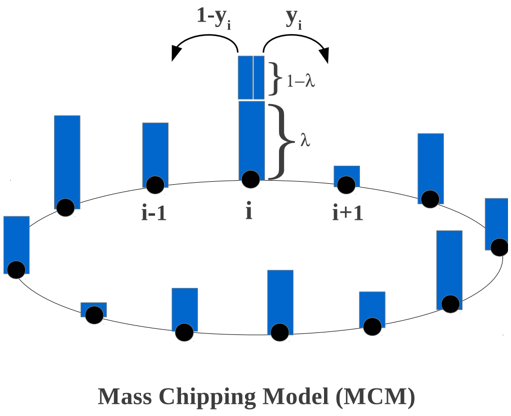

Figure 5.1.: Schematic representation of the mass chipping models (MCMs): $(1-\lambda)$ fraction of mass m_i at a site i is chipped off. Then, one or both of the fractions, y_i and $(1-y_i)$, of the chipped-off mass diffuse and coalesce with one of the nearest-neighbor masses, depending on whether the mass transfer rule to the neighbors is asymmetric or symmetric. Random number $y_i \in [0,1]$ is drawn from a distribution $\phi(y_i)$.

In this section, we study mass chipping models (MCMs), which are defined on a one dimensional ring of L sites, having a continuous mass variable $m_i \geq 0$ at site i where total mass $M = \sum_{i=1}^{L} m_i$ remains conserved [26–28, 31, 81–84]. The dynamics involves chipping, diffusion and coalescence of masses. In the process of chipping, a site i keeps a certain fraction λ of its mass m_i and the rest of the mass $(1-\lambda)m_i$ is chipped off. Then, a random fraction y_i of the chipped-off mass, where y_i is chosen from a probability density function $\phi(y_i)$ with $y_i \in [0,1]$, is transferred to one of its nearest neighbors. The rest of the chipped-off mass is either deposited back to the departure site or transferred to its other nearest neighbor. The mass-chipping processes are schematically represented in Fig. 5.1. Depending on the details of these dynamical rules, there can be several variants of the MCM, as discussed below.

In Model I (MCM I), which were introduced in Ref. [31], a random fraction y_i of the chipped-off mass goes to the right neighbor and the other fraction goes to the left neighbor. In model II (MCM II), which is a generalized version of the models studied in Refs. [27,28,83], the mass transfer rule is as follows. A random fraction y_i of the chipped-off mass goes to *either* of the nearest neighbors, with equal probability 1/2; the rest of the chipped-off mass is deposited back to the departure site. Model III (MEM), which generalises the

KMP process [32] on a ring and several other wealth distribution processes [76, 77, 86, 117], is a bit different in the sense that, here instead of a single site, one bond of neighbouring sites is randomly selected, mass is chipped off from both of the sites and then randomly redistributed within these two sites. In the last variant, which we call Model IV (MCM IV) - particular versions of which were studied in the context of asymmetric random average processes (ARAP) in Refs. [27, 28, 83, 84], mass is transferred completely asymmetrically in a particular direction (say, clockwise) as following. A random fraction y_i of the chipped-off mass, i.e., $y_i(1-\lambda)m_i$, is transferred *only* to the *right* nearest neighbor and the rest of the mass, i.e., $(1-y_i)(1-\lambda)m_i$, comes back to the departure site.

Note that, in MCM IV, mass is transferred completely asymmetrically to the right nearest neighbor; consequently, there is a mass-current in the system and the model is manifestly out of equilibrium. On the contrary, in MCM II, mass is transferred completely symmetrically, with equal probability, to either of its nearest neighbors; in this case, there is *no* net current in the system. However, in MCM I and MEM, the mass transfer can be effectively either symmetric or asymmetric, depending on the form of the probability density function $\phi(y)$. For a symmetric probability density $\phi(y) = \phi(1-y)$, the mass transfer will be indeed symmetric (therefore, no net current in the system); otherwise, the mass transfer is effectively asymmetric and therefore there can be a net current in a particular direction.

Note that, in the symmetric versions of the models, mass transfers take place, without any preference, to the right or (and) to the left nearest neighbor(s); consequently, net mass currents are zero in the nonequilibrium steady states. However, as shown later, the systems with the symmetric transfers still remain far from equilibrium as the dynamics in the configuration space violates Kolmogorov criterion and thus also detailed balance [19]. For asymmetric mass transfers (see section 7.4), the violation is evident as there would be nonzero mass current in the systems. Kolmogorov criterion, which provides a necessary and sufficient condition for detailed balance to hold in a system, says the following. If, for each and every possible loop generated by the dynamics in the configuration space, the probability of a forward path and that of the corresponding reverse path are equal, detailed balance is satisfied, and *vice versa*. As a consequence, if a reverse path corresponding to a forward path in a particular transition in the configuration space does not exist, it suffices to say that Kolmogorov criterion, and therefore detailed balance, is violated. Indeed, in the absence of the knowledge of exact steady-state measures in these mass transport processes, Kolmogorov criterion helps one to check whether detailed balance is satisfied or not.

Stochastic updates in all these variants of the MCMs are done according to either random sequential update (RSU) or parallel update (PU).

5.2.1. Model I (MCM I)

Random Sequential Update (RSU)

In MCM I with random sequential update (RSU), a site i is chosen randomly and a certain fraction $\tilde{\lambda} = 1 - \lambda$ of mass m_i at site i is chipped off. Then, a random fraction y_i of the chipped-off mass, i.e., $\tilde{\lambda} y_i m_i$ is transferred to the right nearest neighbor and the rest of the chipped-off mass, i.e., $\tilde{\lambda}(1 - y_i)m_i$, is transferred to the left nearest neighbor [31]. The stochastic update is given by,

$$m_i(t+dt) = \begin{cases} \underline{\text{value:}} & \underline{\text{prob.:}} \\ \lambda m_i(t) & dt \\ m_i(t) + \tilde{\lambda} y_{i-1} m_{i-1}(t) & dt \\ m_i(t) + \tilde{\lambda}(1 - y_{i+1}) m_{i+1}(t) & dt \\ m_i(t) & (1 - 3dt) \end{cases} \tag{5.10}$$

where the mass value in the first column of r.h.s. is assigned to the mass $m_i(t+dt)$ at a particular site i at time $t+dt$ with the corresponding probability given in the second column and $y_i \in [0,1]$ is a random variable having a probability density $\phi(y_i)$. The first two moments of the probability density function $\phi(y)$ are denoted as

$$\theta_1 = \int_0^1 y\phi(y)dy, \tag{5.11}$$

$$\theta_2 = \int_0^1 y^2\phi(y)dy. \tag{5.12}$$

For the purpose of demonstration, we choose throughout in simulations a particular probability density $\phi(y) = 1$, i.e., a uniform distribution in the unit interval of $y \in [0,1]$, providing

$\theta_1 = 1/2$ and $\theta_2 = 1/3$.

Breakdown of Kolmogorov criterion.– In this model with random sequential update, at any instant of time, mass is chipped off from a single departure site and then it arrives at its two nearest-neighbor destination sites. Clearly, the reverse path, where mass would have been simultaneously chipped from two departure sites $i-1$ and $i+1$ and would have arrived at a single destination site i, is not allowed by the actual dynamics as given in Eq.(7.9). Therefore, Kolmogorov criterion is violated and consequently there is no detailed balance even when there is as such no external biasing force.

We now define two-point correlation function as $c_r = \mathcal{C}_r - \rho^2$ where $\mathcal{C}_r = \langle m_i m_{i+r} \rangle$ with $r \in \{0, 1, \dots, L-1\}$. Note that, for $r = 0$, the quantity $\mathcal{C}_0$ is actually the second moment of mass at any site.

Using the above time-evolution equation and the steady-state condition $d\mathcal{C}_r/dt = 0$, we get the following relations between two-point functions

$$\mathcal{C}_0 = \frac{1}{\lambda + (1-\lambda)(\theta_1 - \theta_2)} \mathcal{C}_1, \tag{5.13}$$

$$\mathcal{C}_2 - 2\mathcal{C}_1 + \lambda \mathcal{C}_0 = 0, \tag{5.14}$$

$$\mathcal{C}_3 - 2\mathcal{C}_2 + \mathcal{C}_1 + (1-\lambda)(\theta_1 - \theta_2)\mathcal{C}_0 = 0, \tag{5.15}$$

$$\mathcal{C}_{r+1} - 2\mathcal{C}_r + \mathcal{C}_{r-1} = 0. \tag{5.16}$$

The method of obtaining these equations is demonstrated for Model IV (MCM IV) for calculational simplicity. Solving the above equations, we obtain the two-point correlation function

$$c_r = \mathcal{C}_r - \rho^2 = \begin{cases} \frac{(1-\lambda)[1-2(\theta_1-\theta_2)]}{\lambda+2(1-\lambda)(\theta_1-\theta_2)} \rho^2 & \text{for } r = 0, \\ -\frac{(1-\lambda)(\theta_1-\theta_2)}{\lambda+2(1-\lambda)(\theta_1-\theta_2)} \rho^2 & \text{for } r = 1, \\ 0 & \text{otherwise.} \end{cases} \tag{5.17}$$

It is important to note that the relations in Eqs. (5.13) to (5.16) involve only two-point, not any higher order, correlations. This is because, in this process (as well as in the other processes considered later) the probability (or, equivalently, the transition rate) with which each mass-chipping event occurs in an infinitesimal time dt depends neither on the mass of departure site nor on that of destination site (e.g., see the transition probabilities given in the respective column in Eqs. (5.10). This is also true in general for n-point correlations, i.e., a particular n-point correlation involve only other n-point or lower order correlations, not n+1 or higher order correlations. In other words, the Bogoliubov-Born-Green-Kirkwood-Yvon (BBGKY) hierarchy for the correlation functions closes for these mass transport processes. this allows one to exactly solve the hierarchy and calculate the steady state correlations.

The variance of subsystem mass $m = \sum_{k=0}^{v-1} m_k$ can be written as $\sigma_v^2 = \langle m^2 \rangle - v^2\rho^2$ where

$$\sigma_v^2 = vc_0 + 2(v-1)c_1 + 2(v-2)c_2 + \cdots + 2c_{v-1}. \tag{5.18}$$

Using Eq. (5.18), we calculate the variance of subsystem mass $\sigma_v^2(\rho) = v\rho^2/\eta$ as a function of density ρ where $\eta = \frac{\lambda+2(1-\lambda)(\theta_1-\theta_2)}{(1-\lambda)[1-2(\theta_1-\theta_2)(2-1/v)]}$.

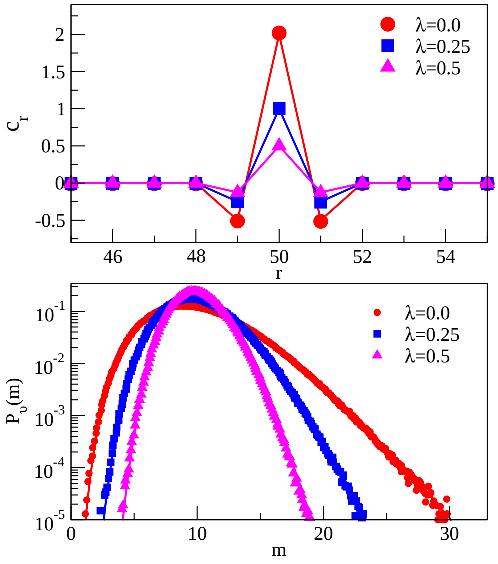

Figure 5.2.: Model I (MCM I) with random sequential update (RSU) for $\lambda = 0$, 0.25 and 0.5. In top panel, two point correlation function $c_r = \langle m_i m_{i+r} \rangle - \rho^2$ is plotted as a function of distance r. In bottom panel, the probability density function $P_v(m)$ for mass in a subsystem size v as a function of subsystem mass m. In all cases, system size $L = 5000$, subsystem size $v = 10$ and mass density $\rho = 1$; points are simulations, lines are theory as in Eq. (5.8).

Therefore, the subsystem mass distribution is given by gamma distribution as in Eq. (5.8) with the above expression of $\eta(\lambda, \theta_1, \theta_2)$. In Fig. 5.2, we have compared our analytical and the simulation results for the two-point correlation function c_r and the probability density function $P_v(m)$ for various values of $\lambda = 0$, 0.25 and 0.5 and for system size $L = 5000$, $\rho = 1$ and subsystem size $v = 10$. Analytic and simulation results show very good agreement.

Parallel Update (PU)

In MCM I with parallel update (PU), the amount of masses which are transferred to the left and right are the same as in the case of RSU (see section III.B.1), but now all sites are simultaneously updated in parallel [31]. The update rule in this case is given below,

$$m_i(t+1) = \lambda m_i(t) + \tilde{\lambda} y_{i-1} m_{i-1}(t) + \tilde{\lambda}(1 - y_{i+1}) m_{i+1}(t) \tag{5.19}$$

where $\tilde{\lambda} = 1 - \lambda$ and $y_i \in [0, 1]$ is a random variable having probability density $\phi(y_i)$.

Breakdown of Kolmogorov criterion.– In the case of parallel update, the breakdown of Kolmogorov criterion, though not quite obvious, can be straightforwardly shown for generic parameter values $\lambda \neq 0$. For example, consider a configuration having two sites $i-1$ and i, with masses m_{i-1} finite and m_i infinitesimal (say, $m_i = 0$, just for the sake of argument), respectively. Then, a chunk of mass is transferred from site $(i-1)$ to site i so that $m_{i-1} \to m'_{i-1} > 0$ and $m_i \to m'_i > 0$. In the next time step, since at least a λ fraction of mass m'_i must be retained at site i, the reverse path where the whole mass m'_i would have been transferred back to $i-1$ from site i is not possible, implying breakdown of Kolmogorov criterion and thus violation of detailed balance. This simple, though not rigorous, argument can be readily extended to any configuration with sufficiently large difference of masses in any two neighbouring sites so that there cannot be a reverse path corresponding to a particular possible path of mass transfer. Note that, in this argument, we consider only the unbiased process $(F = 0)$. Let us consider transitions $\{m_i\} \to \{m'_i\}$ and $\{m'_i\} \to \{m''_i\}$ at two consecutive time steps. In the second transition, one must have $m''_i > \lambda m'_i$, i.e., the mass retained at site i must be at least $\lambda m'_i$. Now, if the amount of mass $\lambda m'_i$ is greater than mass m_i, the value of mass at site i at the initial step, clearly the path cannot be reversed. Therefore the condition for which a process cannot be reversed is simply $\lambda m'_i > m_i$, which, after using Eq. (7.17)

$m'_i = \lambda m_i + \tilde{\lambda} r_{i-1} m_{i-1} + \tilde{\lambda}\tilde{r}_{i+1} m_{i+1}$, leads to the condition

$$r_{i-1} m_{i-1} + \tilde{r}_{i+1} m_{i+1} - \frac{1+\lambda}{\lambda} m_i > 0. \tag{5.20}$$

Therefore, for $\lambda \neq 0$, indeed there are configurations (a finite set in the configuration space) which satisfy the above inequality. This implies breakdown of Kolmogorov criterion and that the steady state is far from equilibrium even in the absence of any biasing force ($F = 0$). Analysis for $\lambda = 0$ requires more effort and is omitted here.

In the steady state, the two-point correlations can be calculated using the above dynamics from the steady-state condition $\langle m_i(t+1) m_{i+r}(t+1)\rangle = \langle m_i(t) m_{i+r}(t)\rangle$, which gives the following relations between the $\mathcal{C}_r$'s: For $r = 0$,

$$(\lambda + (1-\lambda)\beta)\, \mathcal{C}_0 - \lambda \mathcal{C}_1 - (1-\lambda)\alpha\, \mathcal{C}_2 = 0, \tag{5.21}$$

for $r = 1$,

$$\lambda \mathcal{C}_0 - (2\lambda + (1-\lambda)\alpha)\, \mathcal{C}_1 + \lambda \mathcal{C}_2 + (1-\lambda)\alpha\, \mathcal{C}_3 = 0, \tag{5.22}$$

and, for $r \geq 2$,

$$\begin{gathered}(1-\lambda)[\alpha + (\beta - \alpha)\delta_{r,2}]\mathcal{C}_{r-2} + \lambda \mathcal{C}_{r-1} \\ -2[\lambda + (1-\lambda)\alpha]\mathcal{C}_r + \lambda \mathcal{C}_{r+1} + (1-\lambda)\alpha\, \mathcal{C}_{r+2} = 0\end{gathered} \tag{5.23}$$

where $\alpha = \theta_1(1-\theta_1)$ and $\beta = \theta_1 - \theta_2$. To solve the above set of equations, we define a generating function,

$$G(z) = \sum_{r=1}^{\infty} \mathcal{C}_r z^r, \tag{5.24}$$

within a range of $|z| < 1$. Multiplying Eq. (5.23) by z^r and summing over r, we obtain, using Eqs. (5.21) and (5.22),

$$G(z) = \frac{z}{(1-z)} \frac{z[\epsilon(1+z) + 2(\kappa - 1)]\mathcal{C}_0 + (1+z)\mathcal{C}_1}{(z-z_1)(z-z_2)} \tag{5.25}$$

where

$$\epsilon = \frac{\beta}{\alpha};\ \kappa = 1 + \frac{\lambda}{2\alpha(1-\lambda)},\ z_1 = -\kappa + \sqrt{\kappa^2 - 1};\ z_2 = \frac{1}{z_1}. \tag{5.26}$$

The quantities $\mathcal{C}_0$ and $\mathcal{C}_1$ can be obtained along the lines of arguments as in Ref. [28]. Note that, in the limit of large r, $\mathcal{C}_r = \rho^2$ and, therefore, the asymptotic expression of the generating function is given by

$$\lim_{z\to 1} G(z) = \frac{\rho^2}{1-z}, \tag{5.27}$$

which, using Eq. (5.25), immediately leads to a relation between $\mathcal{C}_0$ and $\mathcal{C}_1$

$$(\epsilon + \kappa - 1)\mathcal{C}_0 + \mathcal{C}_1 = (1+\kappa)\rho^2. \tag{5.28}$$

Moreover, at $z = z_1$, which is within the radius of convergence of generating $G(z)$, the function $G(z)$ appears to diverge. However, this cannot be the case as $G(z)$ must remain finite for $z_1 < 1$, implying that the numerator in the r.h.s. of Eq. (5.25) must vanish at z_1, leading to the second relation between $\mathcal{C}_0$ and $\mathcal{C}_1$,

$$z_1[\epsilon(1+z_1) + 2(\kappa-1)]\mathcal{C}_0 + (1+z_1)\mathcal{C}_1 = 0. \tag{5.29}$$

The last two equations give,

$$\mathcal{C}_0 = \frac{1}{\epsilon + \sqrt{\frac{\kappa-1}{\kappa+1}}(1-\epsilon)}\,\rho^2, \tag{5.30}$$

and we obtain the variance of mass at a single site,

$$\sigma_1^2 = (1-\epsilon)\frac{1-\sqrt{\frac{\kappa-1}{\kappa+1}}}{\epsilon + \sqrt{\frac{\kappa-1}{\kappa+1}}(1-\epsilon)}\rho^2. \tag{5.31}$$

Therefore, using Eq. (5.29) and the expression of $\mathcal{C}_0$ in (5.25), the final expression of the generating function is calculated to be,

$$G(z) = \epsilon\,\mathcal{C}_0\,\frac{z}{1-z}\,\frac{\left(1+\frac{2\kappa-1}{\epsilon(1+z_1)}\right)+z}{z-z_2} \tag{5.32}$$

After some algebraic manipulations, the two-point correlation function $c_r = \mathcal{C}_r - \rho^2$ can be expressed as

$$c_r = \begin{cases} (1-\epsilon)\frac{1-\sqrt{\frac{\kappa-1}{\kappa+1}}}{\left[\epsilon+\sqrt{\frac{\kappa-1}{\kappa+1}}(1-\epsilon)\right]}\rho^2 & \text{for } r=0, \\ -\frac{1}{1+\sqrt{\frac{\kappa+1}{\kappa-1}}\frac{\epsilon}{1-\epsilon}}z_1^r\rho^2 & \text{otherwise,} \end{cases} \tag{5.33}$$

where $z_1 = -\kappa + \sqrt{\kappa^2 - 1}$. The magnitude of the correlation function shows exponential decay $c_r \sim \exp(-r/\xi)$ where the correlation length ξ is given by

$$\xi = -\frac{1}{\log|z_1|}. \tag{5.34}$$

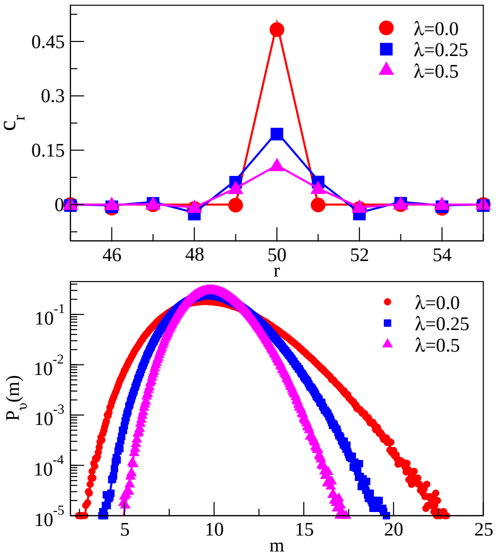

Figure 5.3.: Model I (MCM I) with parallel update (PU) for $\lambda = 0$, 0.25 and 0.5. In top panel, two-point correlation function $c_r = \langle m_i m_{i+r}\rangle - \rho^2$ is plotted as a function of distance r. In bottom panel, the probability density function $P_v(m)$ for mass in a subsystem size v as a function of subsystem mass m. In all cases, system size $L = 5000$, subsystem size $v = 10$, and mass density $\rho = 1$; points are simulations, lines are theory as in Eq. (5.8).

The variance of subsystem mass is obtained using Eq. (5.18),

$$\sigma_v^2 = v\frac{2(1-\epsilon)}{\kappa+1}\frac{\left[1 - \frac{1}{2v}\sqrt{\frac{\kappa-1}{\kappa+1}}\,(1-z_1^v)\right]\rho^2}{\left[\epsilon + \sqrt{\frac{\kappa-1}{\kappa+1}}(1-\epsilon)\right]} \equiv v\frac{\rho^2}{\eta} \tag{5.35}$$

where

$$\eta \approx \frac{\kappa+1}{2}\left[\frac{\epsilon}{1-\epsilon} + \sqrt{\frac{\kappa-1}{\kappa+1}}\right]\left[1 + \frac{1}{2v}\sqrt{\frac{\kappa-1}{\kappa+1}}\right]. \tag{5.36}$$

Consequently, the subsystem mass distributions are described by gamma distribution as in Eq. (5.8) with the above expression of $\eta(\lambda, \theta_1, \theta_2)$. In Fig. 5.3, we have compared our analytical and the simulation results for the two-point correlation function c_r and the probability density function $P_v(m)$ for various values of $\lambda = 0, 0.25$ and 0.5 and for system size $L = 5000$, $\rho = 1$ and subsystem size $v = 10$. Analytic and simulation results show very good agreement.

5.2.2. Model II (MCM II)

Random Sequential Update (RSU)

In MCM II with random sequential update, a site i is chosen randomly and a certain fraction $\tilde{\lambda} = \lambda$ of mass m_i at site i is chipped off. Further, a random fraction y_i of the chipped-off mass, i.e., $\tilde{\lambda} y_i m_i$, is transferred either to the left or to right with equal probability 1/2 and the rest of the chipped-off mass, i.e., $\tilde{\lambda}(1-y_i)m_i$, is deposited back to the site i. The stochastic time evolution in infinitesimal time dt is given below,

$$m_i(t+dt) = \begin{cases} \underline{\text{value:}} & \underline{\text{prob.:}} \\ \lambda m_i(t) + \tilde{\lambda}(1-y_i)m_i(t) & dt \\ m_i(t) + \tilde{\lambda} y_{i+1} m_{i+1}(t) & dt/2 \\ m_i(t) + \tilde{\lambda} y_{i-1} m_{i-1}(t) & dt/2 \\ m_i(t) & (1-2dt) \end{cases} \tag{5.37}$$

$y_i \in [0,1]$ is a random variable having probability density $\phi(y_i)$.

Breakdown of Kolmogorov criterion.– First let us show that the process in the absence of any external bias ($F = 0$) violates Kolmogorov criterion and therefore also detailed balance. Let us consider transitions $\{m_i\} \to \{m_i'\}$ and $\{m_i'\} \to \{m_i''\}$ at two consecutive time steps where

$$m_i' = (1-\tilde{\lambda} r_i)m_i \;;\; m_{i+1}' = m_{i+1} + \tilde{\lambda} r_i m_i,$$
$$m_i'' = (1-\tilde{\lambda} r_i)m_i' \;;\; m_{i+1}'' = m_{i+1}' + \tilde{\lambda} r_i m_i',$$

Now the conditions, $m_i'' = m_i$ and $m_{i+1}'' = m_{i+1}$, for the existence of a reverse path leads to an equality, $r_{i+1}' = r_i m_i/(m_{i+1} + \tilde{\lambda} r_i m_i)$. Or equivalently, an inequality $m_{i+1} \geq \lambda r_i m_i$, as $r_{i+1}' \leq 1$, must be satisfied for the existence of a reverse path. Said differently, the condition for which a reverse path will not exist can be written as the following inequality on the ratio of neighboring masses,

$$\frac{m_i}{m_{i+1}} > \frac{1}{\lambda r_i}.$$

The above condition is satisfied by a finite set in the configuration space and will then imply the steady state to be far from equilibrium even in the absence of any external bias ($F = 0$).

By putting $d\mathcal{C}_r/dt = 0$ in the steady state, we obtain the following relations for two-point functions: For $r = 1$,

$$[\theta_1 - (1-\lambda)\theta_2]\mathcal{C}_0 - 2\theta_1\mathcal{C}_1 + \theta_1\mathcal{C}_2 = 0, \tag{5.38}$$

and, for $r \geq 2$,

$$\mathcal{C}_{r+1} - 2\mathcal{C}_r + \mathcal{C}_{r-1} = 0, \tag{5.39}$$

implying $\mathcal{C}_r$ = constant for $r \geq 2$. Finally, using the steady-state condition $d\mathcal{C}_0/dt = 0$ and Eq. (5.37), we obtain

$$\mathcal{C}_0 = \frac{\theta_1}{\theta_1 - (1-\lambda)\theta_2}\mathcal{C}_1. \tag{5.40}$$

Combining the above relations, we finally have the two-point correlation function,

$$c_r = \mathcal{C}_r - \rho^2 = \begin{cases} \frac{\theta_2(1-\lambda)}{\theta_1-\theta_2(1-\lambda)}\rho^2 & \text{for } r = 0, \\ 0 & \text{otherwise,} \end{cases} \tag{5.41}$$

which is interestingly identical to the results obtained for asymmetric mass chipping model with random sequential update. Accordingly, the variance of mass in a subsystem of size v is given by

$$\sigma_v^2 = vc_0 \equiv v\frac{\rho^2}{\eta}, \tag{5.42}$$

where

$$\eta = \frac{\theta_1 - \theta_2(1-\lambda)}{\theta_2(1-\lambda)}.$$

Therefore, the subsystem mass distribution is given by gamma distribution as in Eq. (5.8) with the above expression of $\eta(\lambda, \theta_1, \theta_2)$. In Fig. 5.3, we have compared our analytical and the simulation results for the two-point correlation function c_r and the probability density function $P_v(m)$ for various values of $\lambda = 0$, 0.25 and 0.5 and for system size $L = 5000$, $\rho = 1$ and subsystem size $v = 10$. Analytic and simulation results show excellent agreement.

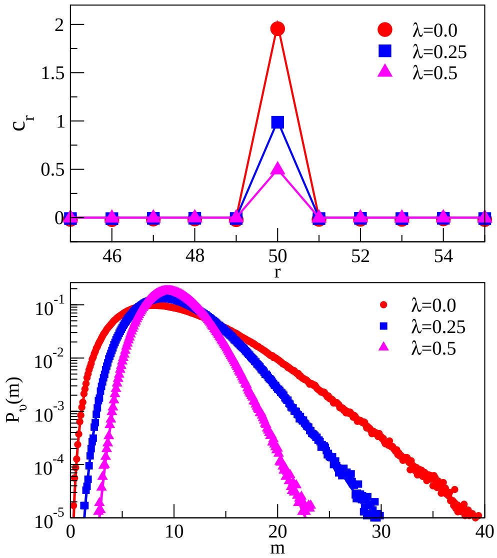

Figure 5.4.: Mass chipping model II (MCM II) with random sequential update (RSU) for $\lambda = 0$, 0.25 and 0.5. In top panel, two-point correlation function $c_r = \langle m_i m_{i+r}\rangle - \rho^2$ is plotted as a function of distance r. In bottom panel, the probability density function $P_v(m)$ for mass in a subsystem size v as a function of subsystem mass m. In all cases, system size $L = 5000$, subsystem size $v = 10$, and mass density $\rho = 1$; points are simulations, lines are theory as in Eq. (5.8).

Parallel update (PU)

In MCM II with parallel update, the amount of mass which is transferred from a site i is the same as in the case of RSU (see Sec. III.C.1), but now all sites are simultaneously updated in parallel. The discrete time-evolution is given below,

$$\begin{aligned} m_i(t+1) = [\lambda + \tilde{\lambda}(1-y_i)]m_i(t) + \tilde{\lambda} s_{i-1} y_{i-1} m_{i-1}(t) \\ + \tilde{\lambda}(1-s_{i+1}) y_{i+1} m_{i+1}(t), \end{aligned} \tag{5.43}$$

where we have introduced a random variable s_i which takes discrete values 0 and 1, each with probability 1/2. When the chipped-off mass moves to the right, $s_i = 1$ and otherwise $s_i = 0$, implying $\langle s_i^n \rangle = 1/2$ for $n \neq 0$.

Breakdown of Kolmogorov criterion.– In this model, the breakdown of Kolmogorov criterion, for generic parameter values $\lambda \neq 0$, can be shown along the lines of arguments as given in the case of parallel update for model I in section 7.2.1. As before, let us consider transitions $\{m_i\} \to \{m'_i\}$ and $\{m'_i\} \to \{m''_i\}$ at two consecutive time steps. Provided that the mass $(1-\tilde{\lambda} r'_i)m'_i$, the least amount of mass retained at site i after second transition, is greater than the initial mass m_i, there cannot be a reverse path. Using dynamical rule in Eq. (7.27), it can be shown that the condition of inequality $(1-\tilde{\lambda} r'_i)m'_i > m_i$ leads to a condition on the initial masses,

$$s_{i-1}r_{i-1}m_{i-1} + (1-s_{i+1})r_{i+1}m_{i+1} - \left(\frac{1}{\lambda} + r_i\right)m_i > 0.$$

The above condition is satisfied for a finite set of configurations in the configuration space and will then imply violation of Kolmogorov criterion, and thus also detailed balance, and that the steady state is far from equilibrium even in the absence of any biasing force ($F = 0$).

To calculate the two-point correlations, we use the steady-state condition

$\langle m_i(t+1)m_{i+r}(t+1)\rangle = \langle m_i(t)m_{i+r}(t)\rangle$, to obtain, for $r = 0$,

$$4[(1-\lambda)\epsilon - 1]\mathcal{C}_0 + 4\alpha\mathcal{C}_1 + (1-\alpha)\mathcal{C}_2 = 0, \tag{5.44}$$

for $r = 1$,

$$4[1-(1-\lambda)\epsilon]\mathcal{C}_0 - (1+7\alpha)\mathcal{C}_1 + 4\alpha\mathcal{C}_2 + (1-\alpha)\mathcal{C}_3 = 0, \tag{5.45}$$

and, for $r \geq 2$,

$$\begin{aligned}(1-\alpha)(1-\delta_{r,2})\mathcal{C}_{r-2} + 4\alpha\mathcal{C}_{r-1} - 2(1+3\alpha)\mathcal{C}_r \\ +4\alpha\mathcal{C}_{r+1} + (1-\alpha)\mathcal{C}_{r+2} = 0,\end{aligned} \tag{5.46}$$

where $\epsilon = \theta_2/\theta_1$ and $\alpha = 1-(1-\lambda)\theta_1$. As in the MCM II in Sec. III.B.2, one can readily solve these equations using the method of generating function $G(z) = \sum_{r=1}^{\infty} \mathcal{C}_r z^r$ as given below,

$$G(z) = \frac{1}{1-\alpha}\frac{z}{1-z}\frac{4[1-(1-\lambda)\epsilon]z\mathcal{C}_0 + (1-\alpha)(1+z)\mathcal{C}_1}{(z-z_1)(z-z_2)} \tag{5.47}$$

where

$$z_1 = -\frac{1-\sqrt{\alpha}}{1+\sqrt{\alpha}}; \;\; z_2 = \frac{1}{z_1}. \tag{5.48}$$

Now, we obtain

$$2[1-(1-\lambda)\epsilon]\mathcal{C}_0 + (1-\alpha)\mathcal{C}_1 = 2\rho^2 \tag{5.49}$$

as $\lim_{z\to 1} G(z) = \rho^2/(1-z)$ and

$$4z_1[1-(1-\lambda)\epsilon]\mathcal{C}_0 + (1-\alpha)(1+z_1)\mathcal{C}_1 = 0 \tag{5.50}$$

as the numerator of $G(z)$ is zero $z = z_1$. Eliminating $\mathcal{C}_1$ from the above two equations and using the expression of z_1, we obtain,

$$\mathcal{C}_0 = \frac{\sqrt{\alpha}}{1-(1-\lambda)\epsilon}\rho^2 \tag{5.51}$$

The expression of $G(z)$ in Eq. (5.47) then reduces to

$$G(z) = \frac{1}{1-\alpha}\frac{z}{1-z}\frac{4[1-(1-\lambda)\epsilon]\mathcal{C}_0 + (1-\alpha)\mathcal{C}_1}{z-z_2}, \tag{5.52}$$

which, from Eq. (5.50), is further reduced to

$$G(z) = \frac{2}{1-\alpha}\frac{z}{1-z}\frac{[1-(1-\lambda)\epsilon]\mathcal{C}_0 + \rho^2}{z-z_2}.$$

Using $\mathcal{C}_0$ from Eq. (5.52), we finally obtain

$$G(z) = \frac{z}{1-z}\frac{1-z_1}{1-zz_1}\rho^2, \tag{5.53}$$

implying $\mathcal{C}_r = (1-z_1^r)\rho^2$ and therefore the two-point correlation function $c_r = \mathcal{C}_r - \rho^2$ can be written as

$$c_r = \begin{cases} \frac{(1-\lambda)\epsilon-(1-\sqrt{\alpha})}{1-(1-\lambda)\epsilon}\rho^2 & \text{for } r = 0, \\ -z_1^r\rho^2 & \text{otherwise.} \end{cases} \tag{5.54}$$

Consequently, using Eq. (5.18), the variance of subsystem mass is given by

$$\sigma_v^2(\rho) = v\left[\frac{(1-\lambda)\sqrt{\alpha}\epsilon}{1-(1-\lambda)\epsilon} - \frac{1-\alpha}{2v}(1-z_1^v)\right]\rho^2 \equiv v\frac{\rho^2}{\eta} \tag{5.55}$$

where

$$\eta \approx \frac{1-(1-\lambda)\epsilon}{\sqrt{\alpha}(1-\lambda)\epsilon - \frac{1-\alpha}{2v}[1-(1-\lambda)\epsilon]}.$$

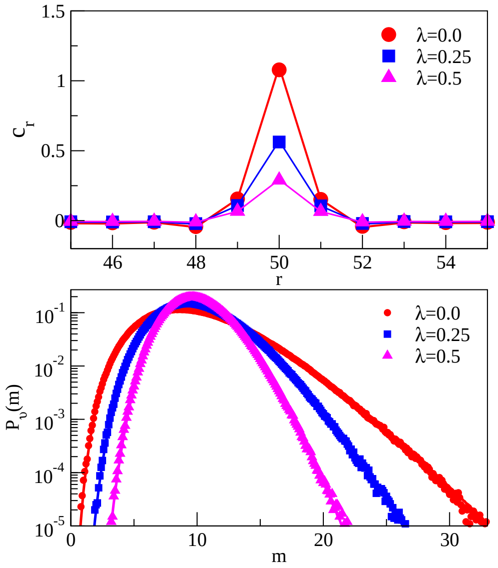

Figure 5.5.: Model II (MCM II) with parallel update (PU) for $\lambda = 0$, 0.25 and 0.5. In top panel, two-point correlation function $c_r = \langle m_i m_{i+r}\rangle - \rho^2$ is plotted as a function of distance r. In bottom panel, the probability density function $P_v(m)$ for mass in a subsystem size v as a function of subsystem mass m. In all cases, system size $L = 5000$, subsystem size $v = 10$, and mass density $\rho = 1$; points are simulations, lines are theory as in Eq. (5.8).

Therefore, the subsystem mass distributions are described by gamma distribution as in Eq. (5.8) with the above expression of $\eta(\lambda, \theta_1, \theta_2)$. In Fig. 5.5, we have compared our analytical and the simulation results for the two-point correlation function c_r and the probability density function $P_v(m)$ for various values of $\lambda = 0$, 0.25 and 0.5 and for system size $L = 5000$, $\rho = 1$ and subsystem size $v = 10$. Analytic and simulation results show excellent agreement.

5.2.3. Model III : Mass exchange model (MEM)

Mass exchange models have been studied throughout the past couple of decades [76, 87, 117, 118], usually on a mean field level - on a graph where all sites interact with each other. Earlier, to consider the effect of a lattice structure on these processes, we studied the MEM on a one dimensional lattice [50], where only neighboring masses can interact by exchanging certain fraction of masses among themselves. The lattice variant of the MEM gives rise to nontrivial spatial correlations, where the exact steady-state weights of the microscopic

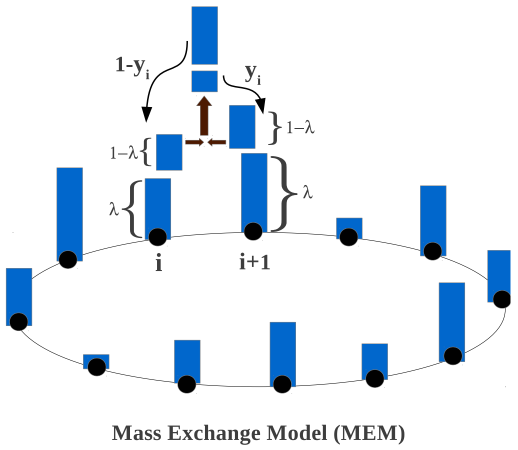

Figure 5.6.: Schematic representation of the mass exchange model (MEM): $(1-\lambda)$ fractions of masses m_i and m_{i+1} at two nearest neighbor sites i and $i+1$ are chipped off and are added up. Then, y_i and $(1-y_i)$ fractions of the added-up masses are assigned to one of the sites $i+1$ and i, respectively. Random number $y_i \in [0,1]$ is drawn from a distribution having a density function $\phi(y_i)$.

configurations, even in one dimension, are still unknown.

In this section, we exactly calculate the two-point spatial correlations for the MEM on a one dimensional periodic lattice of L sites. The dynamical rules for the MEM are as follows. A bond between any two neighboring sites i and $i+1$ is chosen randomly. Certain $\tilde{\lambda} = 1-\lambda$ fraction of their masses, i.e., $\tilde{\lambda} m_i$ and $\tilde{\lambda} m_{i+1}$, are chipped-off and added up. Then, y_i and $1-y_i$ fractions of this added-up mass, where $y_i \in [0,1]$ is drawn from a distribution having a density function $\phi(y_i)$, are exchanged between the sites i and $i+1$. Equivalently, the dynamical rules can be written as

$$m_i(t+dt) = \begin{cases} \underline{\text{value:}} & \underline{\text{prob.:}} \\ \lambda m_i(t) + \tilde{\lambda}(1-y_i) m_{i,i+1}(t) & dt \\ \lambda m_i(t) + \tilde{\lambda} y_{i-1} m_{i-1,i}(t) & dt \\ m_i(t) & (1-2dt) \end{cases} \tag{5.56}$$

where we define a bond-variable $m_{i,i+1} = m_i + m_{i+1}$ being total mass at the bond $(i, i+1)$. For the MEM, we consider only random sequential update as, in this case, parallel update is not well defined.

Violation of Kolmogorov criterion.– Again, let us consider transitions $\{m_i\} \to \{m'_i\}$ and $\{m'_i\} \to \{m''_i\}$ at two consecutive time steps where, by denoting $\mu_{i,i+1} = m_i + m_{i+1}$,

$$m'_i = \lambda m_i + \tilde{\lambda} r_i \mu_{i,i+1} \; ; \; m'_{i+1} = \lambda m_{i+1} + \tilde{\lambda}\tilde{r}_i \mu_{i,i+1},$$
$$m''_i = \lambda m'_i + \tilde{\lambda} r_i \mu_{i,i+1} \; ; \; m''_{i+1} = \lambda m'_{i+1} + \tilde{\lambda}\tilde{r}_i \mu_{i,i+1}.$$

The condition, $m''_i = m_i$ and $m''_{i+1} = m_{i+1}$, of having a reverse path can be written as an equality $r'_i = (1+\lambda)m_i/\mu_{i,i+1} - \lambda r_i$, or alternatively, as an inequality (as $r'_i \leq 1$) on the ratio of neighboring masses $m_i/m_{i+1} \leq (1+\lambda r_i)/\lambda\tilde{r}_i$. Said differently, for $m_i/m_{i+1} > (1+\lambda r_i)/\lambda\tilde{r}_i$, Kolmogorov criterion and detailed balance are violated, and thus the steady state is far away from equilibrium even in the absence of any biasing force ($F = 0$).

Now using the above time-evolution equations and the steady-state condition $d\mathcal{C}_r/dt = 0$, we obtain the following relations. For $r = 0$, the second moment of the distribution of mass at a single site is given by

$$\mathcal{C}_0 = \frac{1 - 2(1-\lambda)(\theta_1 - \theta_2)}{\lambda + 2(1-\lambda)(\theta_1 - \theta_2)}\mathcal{C}_1, \tag{5.57}$$

and, for $r = 1$,

$$2\mathcal{C}_1 = \mathcal{C}_2 + [\lambda + 2(1-\lambda)(\theta_1 - \theta_2)]\mathcal{C}_0 + 2(1-\lambda)(\theta_1 - \theta_2)\mathcal{C}_1. \tag{5.58}$$

The above two relations imply, $\mathcal{C}_1 = \mathcal{C}_2$. Furthermore, for $r \geq 2$, we have

$$\mathcal{C}_{r+1} - 2\mathcal{C}_r + \mathcal{C}_{r-1} = 0, \tag{5.59}$$

implying $\mathcal{C}_r = \rho^2$ for $r \geq 2$. Combining the all of the above relations, we finally obtain

$$c_r = \mathcal{C}_r - \rho^2 = \begin{cases} \frac{(1-\lambda)[1-4(\theta_1-\theta_2)]}{\lambda+2(1-\lambda)(\theta_1-\theta_2)}\rho^2 & \text{for } r = 0, \\ 0 & \text{otherwise,} \end{cases} \tag{5.60}$$

and consequently the variance of subsystem mass,

$$\sigma_v^2 = v c_0 \equiv v\frac{\rho^2}{\eta}, \tag{5.61}$$

where

$$\eta = \frac{\lambda + 2(1-\lambda)(\theta_1 - \theta_2)}{(1-\lambda)[1 - 4(\theta_1 - \theta_2)]}.$$

When the random number $y_i \in [0,1]$ is chosen from a uniform distribution $\phi(y_i) = 1$, $\theta_1 = 1/2$ and $\theta_2 = 1/3$ and therefore $\eta(\lambda) = (1+2\lambda)/(1-\lambda)$. This particular expression of $\eta(\lambda)$ was obtained earlier within mean field approximation $\langle m_i m_{i+r}\rangle \approx \rho^2$ [50], which, as we have shown in this section, is indeed exact due to the fact all the neighboring correlations vanish, i.e., $c_r = \langle m_i m_{i+r}\rangle - \rho^2 = 0$ for $r \geq 1$. As demonstrated in the previous simulations [50], the subsystem mass distributions for various values of λ are indeed described by gamma distribution.

5.2.4. Model IV (MCM IV)

Random Sequential Update (RSU)

In MCM IV with random sequential update (RSU), a site i is chosen randomly. A fraction $\tilde{\lambda} = (1-\lambda)$ of mass m_i at site i is chipped off and then a random fraction y_i of this chipped-off mass $\tilde{\lambda} m_i$ is transferred to the right nearest neighbor; the rest, $\tilde{\lambda}(1-y_i)m_i$, of the chipped-off mass is deposited back to the site i. The dynamics in infinitesimal time dt can be written as given below,

$$m_i(t+dt) = \begin{cases} \underline{\text{value:}} & \underline{\text{probability:}} \\ \lambda m_i(t) + \tilde{\lambda}(1-y_i)m_i(t) & dt, \\ m_i(t) + \tilde{\lambda} y_{i-1} m_{i-1}(t) & dt, \\ m_i(t) & (1-2dt), \end{cases} \tag{5.62}$$

Using the update rules as in Eq. (5.62), infinitesimal time-evolution of the second moment $\langle m_i(t)^2\rangle$, up to order dt, can be written as

$$\begin{aligned} \langle m_i^2(t+dt)\rangle = \langle m_i^2(t)\rangle(1-2dt) \\ +\langle[\lambda + \tilde{\lambda}(1-y_i)]^2 m_i^2(t)\rangle dt \\ +\langle[m_i(t) + \tilde{\lambda} y_{i-1} m_{i-1}(t)]^2\rangle dt + \mathcal{O}(dt^2), \end{aligned} \tag{5.63}$$

or, equivalently,

$$\begin{aligned}\frac{d\mathcal{C}_0}{dt} = \frac{d\langle m_i^2(t)\rangle}{dt} &= -2\langle m_i^2(t)\rangle + \langle[\lambda+\tilde{\lambda}(1-y_i)]^2 m_i^2(t)\rangle \\ &\quad + \langle[m_i(t)+\tilde{\lambda}y_{i-1}m_{i-1}(t)]^2\rangle.\end{aligned} \tag{5.64}$$

By using the steady-state condition $d\mathcal{C}_0/dt = 0$ and that the fact that y_i and m_i are independent random variables, we have

$$\mathcal{C}_0 = \frac{\theta_1}{\theta_1 - \theta_2(1-\lambda)}\mathcal{C}_1. \tag{5.65}$$

The time evolution of two-point correlations C_r, for $r = 1$ and $r \geq 2$, in infinitesimal time dt can be written as

$$m_i m_{i+1}(t+dt) = \begin{cases} \underline{\text{value:}} & \underline{\text{prob.:}} \\ [m_i(t)+\tilde{\lambda}y_{i-1}m_{i-1}(t)]m_{i+1} & dt, \\ m_i[\lambda m_{i+1}(t)+\tilde{\lambda}(1-y_{i+1})m_{i+1}(t)] & dt, \\ [m_{i+1}(t)+\tilde{\lambda}y_i m_i(t)][\lambda m_i(t)+\tilde{\lambda}(1-y_i)m_i(t)] & dt, \\ m_i(t)m_{i+1}(t) & (1-3dt), \end{cases} \tag{5.66}$$

and,

$$m_i m_{i+r}(t+dt) = \begin{cases} \underline{\text{value:}} & \underline{\text{prob.:}} \\ [m_i(t)+\tilde{\lambda}y_{i-1}m_{i-1}(t)]m_{i+r}(t) & dt, \\ [\lambda+\tilde{\lambda}(1-y_i)]m_i(t)m_{i+r}(t) & dt, \\ [m_{i+r}(t)+\tilde{\lambda}y_{i+r-1}m_{i+r-1}(t)]m_i(t) & dt, \\ [\lambda+\tilde{\lambda}(1-y_{i+r})]m_i(t)m_{i+r}(t) & dt, \\ m_i(t)m_{i+r}(t) & (1-4dt), \end{cases} \tag{5.67}$$

which, using the steady-state condition $d\mathcal{C}_r/dt = 0$, lead to

$$\mathcal{C}_2 - 2\mathcal{C}_1 + \frac{\theta_1 - \theta_2(1-\lambda)}{\theta_1}\mathcal{C}_0 = 0, \tag{5.68}$$

for $r = 1$ and

$$\mathcal{C}_{r+1} - 2\mathcal{C}_r + \mathcal{C}_{r-1} = 0, \tag{5.69}$$

for $r \geq 2$. The above relations imply $\mathcal{C}_2 = \mathcal{C}_1$ and $\mathcal{C}_n = \rho^2$ for $r \geq 2$ and can be combined to finally obtain the following,

$$c_r = \mathcal{C}_r - \rho^2 = \begin{cases} \frac{\theta_2(1-\lambda)}{\theta_1-\theta_2(1-\lambda)}\rho^2 & \text{for } r = 0, \\ 0 & \text{otherwise.} \end{cases} \tag{5.70}$$

As $c_r = 0$ for $r \neq 0$, we obtain the variance of the subsystem mass,

$$\sigma_v^2 = v c_0 = v \frac{\theta_2(1-\lambda)}{\theta_1 - \theta_2(1-\lambda)}\rho^2 \equiv v \frac{\rho^2}{\eta} \tag{5.71}$$

where

$$\eta = \frac{\theta_1 - \theta_2(1-\lambda)}{\theta_2(1-\lambda)}.$$

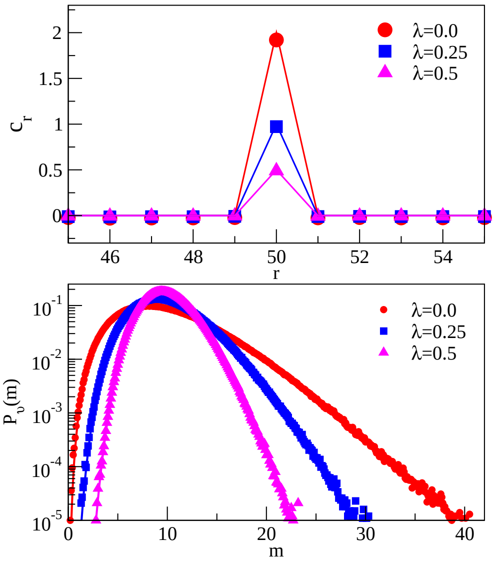

Figure 5.7.: Mass chipping model IV (MCM IV) with random sequential update (RSU) for $\lambda = 0$, 0.25 and 0.5. In top panel, two-point correlation function $c_r = \langle m_i m_{i+r}\rangle - \rho^2$ is plotted as a function of distance r. In bottom panel, the probability density function $P_v(m)$ for mass in a subsystem size v as a function of subsystem mass m. In all cases, system size $L = 5000$, subsystem size $v = 10$ and mass density $\rho = 1$; points are simulations, lines are theory as in Eq. (5.8).

Note that the variance is proportional to the square of the mass density. As derived in section II., this particular functional dependence of the variance on density, along with additivity, implies that the subsystem mass distribution has the form of gamma distribution,

$$P_v(m) = \frac{1}{\Gamma(v\eta)}\left(\frac{\eta}{\rho}\right)^{v\eta} m^{v\eta-1} e^{-\eta m/\rho}, \tag{5.72}$$

as in Eq. (5.8) with the above expression of $\eta(\lambda, \theta_1, \theta_2)$. In Fig. 5.7, we have compared our analytical results with the simulation results, where we numerically calculated the two-point correlation functions c_r and the subsystem mass distributions $P_v(m)$ for various values of $\lambda = 0$, 0.25 and 0.5 and for system size $L = 5000$, $\rho = 1$ and subsystem size $v = 10$. Analytic and simulation results show very good agreement.

Parallel Update (PU)

In MCM IV with parallel update (PU), the amount of mass $\tilde{\lambda} y_i m_i(t)$, which is transferred from a site i to the right nearest neighbor at a time step t, is the same as in the previous case of MCM IV with RSU in Sec. III.A.1, but now all lattice sites are simultaneously updated in parallel. The discrete time stochastic dynamics with parallel update is given by,

$$m_i(t+1) = \lambda m_i(t) + \tilde{\lambda}(1-y_i)m_i(t) + \tilde{\lambda} y_{i-1} m_{i-1}(t) \tag{5.73}$$

where $y_i \in [0,1]$ is a random variable having probability density $\phi(y_i)$. The steady-state correlations can be calculated using the dynamical rules as in Eq. (5.73). We write below explicitly the discrete-time evolution of the two-point correlations $\langle m_i m_{i+r}\rangle$,

$$\begin{aligned}
\langle m_i^2(t+1)\rangle &= \lambda^2\langle m_i^2(t)\rangle + \tilde{\lambda}^2\langle(1-y_i)^2 m_i^2(t)\rangle + 2\lambda\tilde{\lambda}\langle(1-y_i)m_i^2(t)\rangle + \tilde{\lambda}^2\langle y_{i-1}^2 m_{i-1}^2\rangle \\
&\quad + 2\langle[\tilde{\lambda}(\lambda + \tilde{\lambda}(1-y_i))y_{i-1}]\langle m_i(t)m_{i-1}(t)\rangle \\
\langle m_i(t+1)m_{i+1}(t+1)\rangle &= \langle[\lambda m_i(t) + \tilde{\lambda}(1-y_i)m_i(t) + \tilde{\lambda} y_{i-1} m_{i-1}(t)] \\
&\quad \times[\lambda m_{i+1}(t) + \tilde{\lambda}(1-y_{i+1})m_{i+1}(t) + \tilde{\lambda} y_i m_i(t)]\rangle \\
\langle m_i(t+1)m_{i+r}(t+1)\rangle &= \langle[\lambda m_i(t) + \tilde{\lambda}(1-y_i)m_i(t) + \tilde{\lambda} y_{i-1} m_{i-1}(t)] \\
&\quad \times[\lambda m_{i+r}(t) + \tilde{\lambda}(1-y_{i+r})m_{i+r}(t) + \tilde{\lambda} y_{i+r-1} m_{i+r-1}(t)]\rangle
\end{aligned}$$

for $r = 0$, 1 and $r \geq 2$, respectively. Now using the steady-state condition $\langle m_i(t+1)m_{i+r}(t+1)\rangle = \langle m_i(t)m_{i+r}(t)\rangle$ in the above equations, we obtain the following relations between the correlation functions $\mathcal{C}_r$'s: For $r=0$,

$$\mathcal{C}_0 = \frac{\lambda\theta_1 + (1-\lambda)\theta_1(1-\theta_1)}{\lambda\theta_1 + (1-\lambda)(\theta_1-\theta_2)}\mathcal{C}_1, \tag{5.74}$$

for $r=1$,

$$\mathcal{C}_2 - 2\mathcal{C}_1 + \frac{\lambda\theta_1 + (1-\lambda)(\theta_1-\theta_2)}{\lambda\theta_1 + (1-\lambda)\theta_1(1-\theta_1)}\mathcal{C}_0 = 0 \tag{5.75}$$

and, for $r \geq 2$,

$$\mathcal{C}_{r+1} - 2\mathcal{C}_r + \mathcal{C}_{r-1} = 0. \tag{5.76}$$

Now combining Eqs. (5.74), (5.75), and (5.76), we obtain the two-point correlation function,

$$c_r = \mathcal{C}_r - \rho^2 = \begin{cases} \frac{(1-\lambda)(\theta_2-\theta_1^2)}{\lambda\theta_1+(1-\lambda)(\theta_1-\theta_2)}\rho^2 & \text{for } r=0 \\ 0 & \text{otherwise.} \end{cases}, \tag{5.77}$$

and the subsystem mass variance, $\sigma_v^2 = vC_0 \equiv v\rho^2/\eta$, where $\eta = \frac{\lambda\theta_1+(1-\lambda)(\theta_1-\theta_2)}{(1-\lambda)(\theta_2-\theta_1^2)}$.

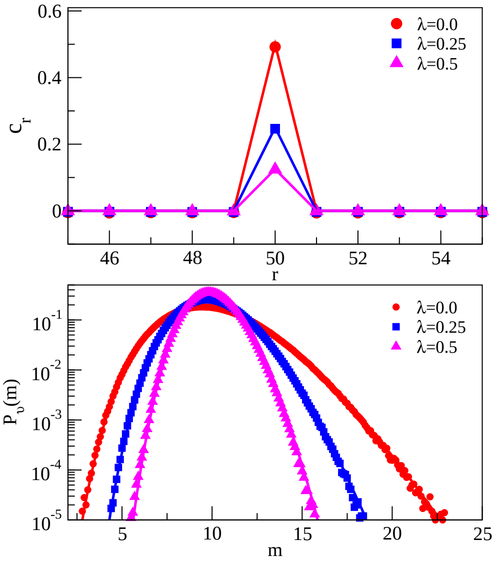

Figure 5.8.: Model IV (MCM IV) with parallel update (PU) for $\lambda = 0$, 0.25 and 0.5. In top panel, two-point correlation function $c_r = \langle m_i m_{i+r}\rangle - \rho^2$ is plotted as a function of distance r. In bottom panel, the probability density function $P_v(m)$ for mass in a subsystem size v as a function of subsystem mass m. In all cases, system size $L = 5000$, subsystem size $v = 10$, and mass density $\rho = 1$; points are simulations, lines are theory as in Eq. (5.8).

The subsystem mass distribution is given by gamma distribution as in Eq. (5.8) with the above expression of $\eta(\lambda,\theta_1,\theta_2)$. In Fig. 5.8, we have compared our analytical and the simulation results for the two-point correlation function c_r and the probability density function $P_v(m)$ for various values of $\lambda = 0$, 0.25 and 0.5 and for system size $L = 5000$, $\rho = 1$ and subsystem size $v = 10$. Analytic and simulation results show very good agreement.

Note that, for both random sequential and parallel update dynamics in MCM IV, the two-point correlations vanish, $c_r = 0$ for $r \neq 0$. In other words, spatial correlation length ξ over which $c_r \sim \exp(-r/\xi)$ decays is essentially zero, i.e., $\xi = 0$. In that case, additivity is expected to hold even on the single-site level, which is indeed the case as verified in [50] where the distribution of mass at any single site was shown to be well approximated by gamma distribution. In fact, as we have shown here, the variance calculated in [50] within mean field approximation is indeed exact as all the neighboring correlations vanish, i.e., $c_r = 0$ for $r = 1$.

5.3. Concluding remarks

We have characterized spatial structure in a broad class of nonequilibrium conserved-mass transport processes, which represent a wealth of natural phenomena concerning fragmentation, diffusion and coalescence of masses. Except for a few spacial cases which have a product measure [27, 28, 31, 83, 85], e.g., mass chipping model IV with parallel update and with $\lambda = 0$ [31], these processes in general give rise to nontrivial steady-state structure, which, in most of the cases, are not exactly known [84]. Here, we have demonstrated that even the perfectly symmetric versions of these systems violate conditions for detailed balance and therefore the steady states are inherently far from equilibrium. In the thermodynamic limit, we have exactly calculated the two-point spatial (equal-time) correlation functions, which are found to be short-ranged. Remarkably, these processes possess an equilibriumlike thermodynamic structure: They have an additivity property and, consequently, there exists a fluctuation-response (FR) relation (Eq. 3.10) between the compressibility and the fluctuations, analogous to the equilibrium fluctuation dissipation theorem (FDT). To substantiate our claims, we have used additivity and the corresponding FR relation to obtain, in the thermodynamic limit, the probability distribution function, including the large-deviation probability and the corresponding large deviation function, of subsystem mass.

In all the cases studied here, the variance of subsystem mass is proportional to square of the mass density. This particular functional dependence of the variance of subsystem mass on mass density, together with additivity and the FR relation, leads to the subsystem mass distributions having the form of gamma distribution. Quite interestingly, gammalike distributions have been observed in various experiments in the past [73, 79], which could be understood in the light of the results of this work.

We note that the main reason due to which the two-point spatial correlations can be calculated in these mass transport processes is that the Bogoliubov-Born-Green-Kirkwood-Yvon (BBGKY) hierarchy involving correlation functions here closes, e.g., the two-point correlations do not depend on the three-point (or higher order) correlations, etc. Nevertheless, the full characterization of three-point and higher order spatial correlations is still lacking and remains to be an interesting open issue, understanding of which could shed some light on the exact microscopic steady-state structure in these systems.

In summary, we have demonstrated that a broad class of conserved-mass transport processes have an equilibriumlike thermodynamic structure. That is, like in equilibrium, the fluctuations in these processes can be characterized in terms of thermodynamic potentials, such as a nonequilibrium free energy function and a chemical potential. Our results could be significant, considering that it is not often that, in driven interacting-particle systems, two-point correlations [23, 25, 26] and, especially, the mass distributions [109, 119] can be calculated exactly. From an overall perspective, our work leaves open the possibility of a unified thermodynamic framework for driven systems in general.

Part II.

DYNAMICS

6. Dynamic approach towards bulk characterisation : Macroscopic Fluctuation Theory

For systems in equilibrium where detailed balance is obeyed, the Einstein relation (ER, introduced in Chapter 1) is universal, irrespective of the details of inter-particle interactions or that of whether the systems are liquids or gases, etc. The ER involves two bulk transport coefficients $D(\rho)$ and $\chi(\rho)$, defined on a macroscopic level from the two phenomenological laws of transport - Fourier's and Ohm's law. One way to understand such macroscopic phenomenological fluctuation relations is to derive, from microscopic dynamics, a hydrodynamic description of the systems on a large space and time scales. However, such a task, for classical deterministic (or quantum) dynamics, is quite difficult. On the other hand, for systems governed by stochastic dynamics, there has been considerable progress made in this direction in last few decades [60, 120, 121].

In the thesis we shall calculate, using macroscopic fluctuation theory (MFT), the fluctuating hydrodynamics of a broad class of one-dimensional conserved-mass transport processes introduced in earlier chapters. Before that, it will be useful to have a detailed introduction to the MFT formalism, as discussed in the following sections.

6.1. Fluctuating hydrodynamics : General considerations

In the earlier chapters we discussed some very general aspects of fluctuations in steady states and have obtained results for a broad class of mass-transport processes within the framework of additivity. It is important to understand their connection to hydrodynamics. It is emphasized that, although the exact steady-state probability weights are not known, at a coarse-grained level we have a much simpler description of matter. To recapitulate, we wrote the joint subsystem mass distribution for conserved processes in the form,

$$\mathcal{P}[\{M_k\}] \simeq \frac{\prod_k W_v(M_k)}{Z(V,M)} \delta\left(\sum_k M_k - M\right), \tag{6.1}$$

have identified that the weight factor $W_v(M_k)$ can be characterized by a large deviation 'density' function $f(\rho_k)$ (or 'rate' function; equivalent to 'nonequilibrium free energy' density) as $W_v(M_k) \simeq \exp[-v f(\rho_k)]$ where $\rho_k = M_k/v$ is fluctuating subsystem mass density [14]. The immediate consequence is a fluctuation-response relation analogous to equilibrium fluctuation-dissipation theorems,

$$f''(\rho) = \frac{d\mu}{d\rho} = \frac{1}{\sigma^2(\rho)}, \tag{6.2}$$

where $\mu(\rho) = f'(\rho)$ is defined to be a chemical potential and $\rho = \langle \rho_k \rangle$ is average local mass density, which equals $\rho_0 = M/V$ for homogeneous systems. Now, instead of subsystem mass variables $\{M_k\}$, additivity property [Eq. (6.1)] can be written in terms of subsystem density variables $\{\rho_k = M_k/v\}$, or equivalently, in terms of coarse-grained fluctuating density profile in the continuum limit $\{\rho(x)\}$. Then, one can write the joint subsystem density distribution, or large-deviation probability of a given density profile $\{\rho(x)\}$, as

$$\mathcal{P}[\rho(x)] \simeq e^{-\mathcal{F}[\rho(x)]},$$

where $\mathcal{F}[\rho(x)]$ is called large deviation function (LDF), given by,

$$\mathcal{F}[\rho(x)] = \int_V dx \, \{f(\hat{\rho}) - f(\rho) - \mu(\rho)(\hat{\rho} - \rho)\}, \tag{6.3}$$

Which, for conserved models, take the form,

$$\mathcal{F}[\rho(x)] = \int_V dx \, \{f(\hat{\rho}) - f(\rho)\}, \tag{6.4}$$

The FR in Eq. (6.2) can be verified from Eqs. (6.3) and (6.4).

Thus, additivity property helps one to construct a statistical mechanical framework for macroscopic characterisation of these conserved-mass transport processes, through a free energy density $f(\rho)$ and a chemical potential $\mu(\rho)$, which however describes only the static properties of *steady-state* mass fluctuations. To have such a macroscopic description for relaxing systems is precisely the aim of fluctuating hydrodynamics. At this stage, one could further ask whether the above steady-state LDFs can be derived in a dynamical setting, or whether fluctuation relations like FDTs in equilibrium could be calculated for systems possessing a NESS. To address these issues, here we formulate, within recently developed macroscopic fluctuation theory (MFT) [47, 122], a statistical mechanical description of fluctuations for these processes. The formulation provides a dynamical description of mass fluctuations at a coarse-grained level, i.e., a fluctuating hydrodynamics valid in large length and time scales [see Eq. (7.37)].

Since mass remains conserved locally under the microscopic evolution, one must keep the mass conservation valid also at the hydrodynamic scales. Therefore, the hydrodynamic equation must be written in the form of a continuity equation,

$$\partial_\tau \rho(x,\tau) + \partial_x J(\rho(x,\tau)) = 0, \tag{6.5}$$

which governs the time evolution of density field $\rho(x,\tau)$ with x and τ being suitably rescaled position and time, respectively. Since, the class of processes we consider here are of 'gradient type' (i.e., local diffusive current can be expressed as a gradient in local observables) [123] with respect to their microscopic evolutions, one would expect a nonlinear hydrodynamics in the diffusive scaling limit, where the current $J(\rho(x,\tau))$ is sum of two parts $J = J_D + J_d$. The first part $J_D = -D(\rho)\partial_x\rho$ is the diffusive current with $D(\rho)$ being the diffusion coefficient and the second part $J_d = \chi(\rho)F$ is the drift current due to a small slowly varying biasing field $F(x)$ (conjugate to conserved mass variable) with $\chi(\rho)$ being the conductivity.

According to the hydrodynamic Eq. (6.5), along with a constitutive relation for the current $J(\rho) = -D(\rho)\partial_x\rho + \chi(\rho)F$, the density field $\rho(x,\tau)$ evolves deterministically in time. However, to study any dynamical aspects of fluctuations, one requires to add a suitable noise term. Clearly, as the noise in this case should maintain the local mass conservation, one must add a noise term ζ to the deterministic part of the current $J(x,\tau) \to J(x,\tau) + \zeta(x,\tau)$, making the total current now a fluctuating one. But the question here is what properties the noise ζ would have. As we shall see, within MFT [see Eq. (7.37)] the fluctuating part ζ of the total current can be represented in terms of a weak multiplicative Gaussian white noise, whose strength explicitly depends on the conductivity $\chi(\rho)$. So the problem of formulating a theory of large fluctuations in these processes essentially boils down to finding the functional dependence of the diffusion coefficient $D(\rho)$ and the conductivity $\chi(\rho)$ on local density ρ.

Before proceeding with the models and derivation of the results, we summarise the details of the formulation of MFT.

6.1.1. The MFT formulation

Given a stochastic Markovian system we ask the question, what is the large deviation probability of observing a nonequilibrium macroscopic current and density profile? For systems that allow certain simplifications and which have equilibrium as the time stationary state, Bertini et al. developed the following approach. For stochastic systems having Markov property, it is in principle possible to bias the system conditioned to its rare configuration [96, 124]. In other words, the non-stationary large deviation current and density of the original unbiased system becomes the typical current and density for the biased one. To quantify the effect of such a bias, they hypothesised that, the system satisfies a *local detailed balance* condition, i.e., the biased transition rates from one configuration to other mimics an equilibrium-like detailed balance criterion. It is further hypothesised that, even in the non-stationary state, the system allows a scale separation such that, in large length and time scales a *local equilibrium* description where the local average values of observables in a macroscopically infinitesimal but microscopically infinitely large region is given by the averages taken over the equilibrium ensemble consistent to the local density, is obeyed. These two hypotheses will be elaborated shortly.

With these, the framework of Macroscopic Fluctuation Theory has been developed. It is shown that, when the system obeys the gradient condition[1], we can identify a diffusion coefficient $D(\rho)$ and a mobility $\chi(\rho)$ from microscopic dynamics, which is related to the equilibrium free energy density $f(\rho)$ via the Einstein relation,

$$\frac{1}{\sigma^2(\rho)} = f''(\rho) = \frac{D(\rho)}{\chi(\rho)}, \tag{6.7}$$

where $j(x,\tau)$ and $\rho(x,\tau)$ being the fluctuating macroscopic current and density respectively. The strategy to obtain the large deviation probability is the following. In presence of a weak

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[1] It is crucial that in the models considered there is only a single conserved quantity, namely, the mass, and that the dynamics is completely defined given the mass configuration is specified. This allows one to consider all relevant macroscopic observables to be functions of (local) density only. However there is a further and most crucial restriction in the usual formulation of MFT, that, we can write a consistent coarse grained fluctuating hydrodynamics equation if and only if the average instantaneous local current satisfy a gradient condition in a local observable,

$$q_{ij} = u_i - u_j, \tag{6.6}$$

where $u_i \equiv u(\rho_i)$, ρ_i being the local density.

bias F the 'typical' (i.e. average as well as most probable) current will have the form,

$$j_m = j_0 + \chi(\rho)F. \tag{6.8}$$

The statement that the force F is conditioned to a fluctuation of the original system have the following quantitative interpretation. In the absence of F, $j_0 = -D(\rho)\frac{\partial\rho}{\partial x}$ would have been the typical current of the unbiased system and j_m being the observed large deviation current. However this j_m is also the typical current of conditionally biased system which undergoes typical Gaussian fluctuation around this j_m. With this identification we can translate the large fluctuations in the current of the original system into the fluctuation in the external bias F, which, being a typical fluctuation, must be a Gaussian (by CLT),

$$\begin{aligned} Prob(\rho, j) &\approx exp\left[-\frac{V}{4}\int d\tau \int dx F.\chi(\rho)F\right] \\ &= exp\left[-\frac{V}{4}\int d\tau \int dx \frac{\left(j + D(\rho)\frac{\partial\rho}{\partial x}\right)^2}{\chi(\rho)}\right] \end{aligned} \tag{6.9}$$

where the first equality is obtained through a Boltzman-Gibbs distribution for the 'work done' by the the external bias for building up the observed atypical profile, while the last equality is obtained by substituting F from Eq. (6.8). Thereafter we can integrate out current under the constraint of continuity to obtain the density profile distribution, and vice versa. To calculate the probability of observing some time-averaged current q over a certain time T (in the spirit of Derrida's additivity), we have to integrate out ρ along with another constraint given by the mathematical definition of time-average, $q = \frac{1}{T}\int_0^T j d\tau$. In this way can in principle quantify various macroscopic properties within MFT.

Before proceeding further, we identify an alternative way of stating the distribution in 6.9. In the spirit of fluctuating hydrodynamics, it is illuminating to identify the macroscopic noise in the coarse-grained current of the original system,

$$j(\rho) = -D(\rho)\frac{\partial\rho}{\partial x} + \sqrt{\chi(\rho)}\eta(x,\tau), \tag{6.10}$$

$\eta(x,\tau)$ being the Gaussian white noise characterized by its two point correlation,

$$\langle \eta(x,\tau)\eta(x',\tau')\rangle = \frac{1}{V}\delta(x-x')\delta(\tau-\tau').$$

Eq. (6.10) is the coarse-grained fluctuating hydrodynamics for the systems under consideration. This equation, added with the continuity Eq. (6.5) completes the description.

It can be shown (to be elaborated in next chapter) that, if the transport coefficients obey the Einstein relation, the Eq. (6.3) is the time stationary solution for density profile distribution obtained from Eq. (6.9). We find that, a broad class of nonequilibrium systems, quite surprisingly, satisfy both ER and the Eq. (6.9), and consequently their steady state density profiles must be distributed according as (6.3), which is precisely the additivity property.

It is worth mentioning that, although the above discussion refers to a local equilibrium hypotheses and the existence of equilibrium free energy, the approach is much more general that encompasses the macroscopic and dynamic characterisation of a broad class of systems exhibiting only nonequilibrium steady states. In fact, in the next chapter it will be argued that, for all the models considered so far, the additivity property allows a remarkable identification of the role of steady state large deviation function as a nonequilibrium free energy, and the local equilibrium must be understood more generally as a *local steady state*; the ER is satisfied allowing a fluctuating hydrodynamics of the form 6.10 for these models. Prior to that that, in rest of the present chapter, we would like to elaborate the hypotheses made, coarse-graining procedures taken, and the challenges posed by systems having a NESS.

Note I. Biasing the system and local detailed balance: We perturb the system by a weak and slowly varying force field F_i in, say, the clockwise direction, such that the symmetry of the transition is broken and the mass transfer from site i to $i+1$ is preferred compared to the move from i to $i-1$. Note that, *the effect of the field is only to change the mass transfer rates*, while the quantity of transferred mass remain unchanged. *Local detailed balance* condition quantifies this change:

For some mass transport process let us call c_{ij} to be the rate of transfer of mass from site

i to j. For models with random sequential update this rate is identical to the transition probability among different configurations. If we bias the system by a field F_i, the mass transfer rate is modified to some c^F_{ij}. In equilibrium where detailed balance condition is satisfied, we have the following,

$$\frac{c_{ij}/c^F_{ij}}{c_{ji}/c^F_{jj}} = e^{-W_{ij}}.$$

Considering mass transfer rates form i to j and form j to i in equal footing, we claim,

$$c^F_{ij} = c_{ij} e^{W_{ij}/2}, \tag{6.11}$$

$W_{ij} = \Delta m_{ij} F_i.(j-i)$ being the work done by the external field for a transfer of mass Δm_{ij} from site i to site j. [2]

Obviously, since the above equations connect the transition rates, they would be applicable even in the transient state. Moreover, although in the mass transport processes under consideration, the time-stationary state itself is a NESS far from equilibrium, we proceed with the local detailed balance assumption. For weak external bias and with $|i-j|/L \ll 1$, in subsequent calculations we will approximate Eq. (6.11) by,

$$c^F_{ij} \approx c_{ij}\left[1 + \frac{W_{ij}}{2}\right]. \tag{6.12}$$

Note II. Local steady state : Ideally, the continuum description is associated with taking a continuum limit along with coarse-graining. Taken together, the whole procedure states the following: We embed the lattice system of L sites and total mass M on a finite interval, say, 1, and define subsystems of size v that are *macroscopically infinitesimal but microscopically infinitely large* (i.e., $\xi \ll v(i) \ll L$, ξ is the correlation length). The continuum limit is defined by taking limits $L \to \infty$, $M \to \infty$ such that $M/L = \rho$ finite. While coarse graining we shall also consider the thermodynamic limit, namely, $L \gg v \to \infty$.

Thus in the coarse-grained description, we look at the system at lengths of unit $\mathcal{O}(L)$ in a time of unit $\mathcal{O}(L^z)$, z being the dynamic exponent characterising the time scale of

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[2] We should really replace $F_i.(j-i)$ by $\int_i^j F(x)dx$. However, the interaction being short ranged and the field being slowly varying, the expression used for quantifying W_{ij} justified.

relaxation of global fluctuations. To quantify hydrodynamic observables, we first choose a neighborhood $v(i) \equiv \Delta x = v(i)/L$ around a site $i \equiv x = \frac{i}{L}$. Then we choose, in similar lines, a macroscopically infinitesimal but microscopically infinitely large time interval [i.e. $T \geq t_{\mathrm{r} \equiv \Delta\tau \approx \mathrm{t}_{\mathrm{r}/\mathrm{L}^{\mathrm{z}},\ \mathrm{t}_{\mathrm{r}}}}$ being the time scale of relaxation of local fluctuation. We argue that, in time $\Delta\tau$, large number of updates take place within the neighborhood $v(i)$ but the macroscopic quantities remain almost same, and any conceivable local fluctuations have died out. In other words, there are intermediate scales in which only infinitesimal macroscopic changes occur so that, the *local* neighbourhood behave as if they are in *homogeneous steady state* consistent to local density. Thereafter, taking the limit of $\Delta x \to 0$, we obtain the required hydrodynamic equations.

In view of this discussion we can quantify the above hypotheses by stating that, the average value of an observable $A(m_i)$ over neighborhood $v(i)$ and time T is equal to the steady state average consistent to the value of local density:

$$\lim_{\Delta x \to 0} \frac{1}{v(i)} \sum_{v(i)} \overline{A(m_i)} = \lim_{\Delta x \to 0} \frac{1}{v(i)} \sum_{v(i)} \langle A(m_i) \rangle = \langle A(m_i) \rangle_{\mathrm{lss}} \,, \tag{6.13}$$

and,

$$\langle A(m_i) \rangle_{\mathrm{lss}} \equiv \psi(\rho_i) = \psi\left(\lim_{\Delta x \to 0} \frac{1}{v(i)} \sum_{v(i)} \langle m_i \rangle \right), \tag{6.14}$$

The last equality in Eq. (6.14) follows by replacing A in Eq.(6.13) by m_i, whereas the first one follows from the fact that, by definition of the dynamics we can write average of any observable as functions of the single conserved quantity, viz. mass density, in steady state.

Note that, the local stationarity in Eq. (6.13) is conceived as consequence of the law of large numbers [60] in presence of only short range correlations. For correlation time much smaller than T, $\overline{A(m_i)}$ is a sample average of almost independent random variables $A(m_i, t)$ and could be replaced by $\langle A(m_i) \rangle$ by LLN giving us the first equality of Eq. (6.13). The spatial randomness in $\langle A(m_i) \rangle$ is taken care of by LLN for the average over the subsystem in the thermodynamic limit, which, provided T is also larger than the relaxation time of local fluctuation, will give us the second equality.

6.2. Demonstration with KMP model on a ring

The well-studied Kipnis-Marchioro-Presutti (KMP) model on a ring, which is nothing but the Model III (defined in last chapter) with $\lambda = 0$, provides a perfect platform to demonstrate the methods. Steady state of this model is known to be product measure and it is exactly solved. Moreover, this model obeys the detailed balance condition for equilibrium and therefore must satisfy ER in macroscopic scales.

In the following, starting from from the microscopic stochastic dynamics for this model, we shall derive the two transport coefficients in the framework of MFT and will also check whether the ER is indeed satisfied in the coarse-grained limit.

The update rule for this model is given by setting $\lambda = 0$ in 5.56, i.e., the whole of the mass of two neighbouring sites are mixed and then redistributed randomly. For non-driven KMP model in one dimenson, for example that with both boundaries attached to baths of same mass density or on a ring, the steady state joint probability density for a given configuration m_i is given by [125],

$$\mathcal{P}(\{m_i\}) = \prod_i \frac{1}{\rho} e^{-m_i/\rho}. \tag{6.15}$$

Using local detailed balance and the following expressions (the methods detailed in [47] and the next chanper),

$$\begin{aligned}\Delta m_{i+1\to i} &= r_i m_{i+1}(t) - (1-r_i) m_i(t)\\ \Delta m_{i-1\to i} &= (1-r_{i-1}) m_{i-1} - r_{i-1} m_i(t),\end{aligned}$$

we have the following evolution for density $\rho_i \equiv \langle m_i \rangle$,

$$\begin{aligned}\frac{d\rho_i}{dt} &= \frac{\tilde{\lambda}}{2}[\rho_{i+1} - 2\rho_i + \rho_{i-1}].\\ &- \frac{1}{2}\Big\langle (\Delta m^2_{i+1\to i} Fa - \Delta m^2_{i-1\to i} Fa) \Big\rangle,\end{aligned}$$

To calculate the averages we make use of the local equilibrium prescription and Eq. (6.15) to obtain,

$$\frac{d\rho_i}{dt}$$
$$= \frac{1}{2}[\rho_{i+1} - 2\rho_i + \rho_{i-1}]$$
$$-\frac{1}{6}\left[2(\rho_{i+1}^2 - \rho_{i-1}^2) - 1(\rho_{i+1}\rho_i - \rho_i\rho_{i-1})\right]Fa$$

Thereafter, taking the continuum limit by rescaling space and time as $i \to x = i/L$, $t \to \tau = t/L^2$ and $a \to 1/L$ for this diffusive system, we obtain the hydrodynamic equation for the density field : $\partial_t\rho(x,\tau) + \partial_x J(\rho(x,\tau)) = 0$ where $J = J_d + J_D$, with

$$J_d(\rho) = \frac{1}{2}\rho^2 \ ; \ J_D(\rho) = -\frac{1}{2}\frac{\partial\rho}{\partial x}.$$

The transport coefficients for this process is, therefore,

$$\chi(\rho) = \frac{1}{2}\rho^2 \ ; \ D(\rho) = \frac{1}{2}.$$

The ER in Eq. (6.7) that implies $\sigma^2(\rho) = \rho^2$ can be verified directly from Eq. (6.15).

6.3. Challenges posed by systems having NESS

However, in systems having a NESS, one generally does not expect the ER or FDTs to be valid. Because, unlike in equilibrium, they violate detailed balance and usually cannot be described by the Boltzmann-Gibbs distribution. In fact, in most cases, microscopic probability weights in the steady state are *not* known. Even for simplest stochastic systems having a NESS, the steady-state probability weights are not in general known and tackling the problem analytically in such systems, especially when there are nonzero finite spatial correlations, remains to be a challenging one [126]. As the demonstration in last section emphasizes, these microscopic inputs, i.e. the knowledge of the steady state moments and correlations, are essential so that the formalism is useful at all. Perhaps not surprisingly, so far there are not many nonequilibrium interacting-particle systems for which exact hydrodynamic descriptions have been derived. In fact, the difficulty arises primarily because fluctuation, diffusion coefficient and conductivity, which would appear in ER (if valid at all) in such systems, must be calculated, not in or around equilibrium, but in a steady state far from equilibrium that have mostly defied analytical characterisation in terms of microscopic distributions.

Quite interestingly recent studies [14, 47–51, 97, 122, 123, 127–130], many of which discussed in Part I, have indicated that, even in broad class of NESSs, there can be fluctuation relations analogous to the FDTs in equilibrium. In particular, the ER has been found, mostly numerically, in several model systems [131–133] having a NESS. These provide a footstep to study the fluctuating hydrodynamics far from equilibrium.

7. Einstein Relation and fluctuating hydrodynamics for nonequilibrium systems

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[1] This chapter is based on the paper, Einstein relation and hydrodynamics of nonequilibrium mass transport processes, Arghya Das, Anupam Kundu, and Punyabrata Pradhan, Phys. Rev. E **95**, 062128 (2017).

In present chapter, we study the fluctuating hydrodynamics of a broad class of nonequilibrium conserved-mass transport processes on a ring defined in chapter 4. To recapitulate briefly, these processes are governed by chipping, diffusion and coalescence of neighboring masses with total mass in the system being conserved, and have become paradigm in nonequilibrium statistical physics of driven many-particle systems [78, 81]. Indeed, throughout the last couple of decades, these have been explored intensively to model a huge variety of natural phenomena, such as, formation of clouds [67] and gels [28,70], force fluctuation in packs of granular beads [79,80], transport of energy in solids [32], dynamics of interacting particles on a ring [27], self-assembly of molecules in organic and inorganic materials [72,73], and distribution of wealth in a society [76], etc.

The challenge before deriving the hydrodynamics of the above mentioned one-dimensional conserved-mass transport processes are that, these have nontrivial spatial correlations (nonzero and finite), with their steady-state weights in most cases not known. For these processes, we explicitly calculate the two transport coefficients as a function of local mass density ρ – the bulk-diffusion coefficient $D(\rho)$ and the conductivity $\chi(\rho)$, which characterize the hydrodynamics. Remarkably, we found that, for this class of models, the two transport coefficients satisfy an equilibriumlike Einstein relation,

$$D(\rho) = \frac{\chi(\rho)}{\sigma^2(\rho)}, \tag{7.1}$$

where

$$\sigma^2(\rho) = \lim_{v\to\infty} \frac{\langle m^2\rangle - \langle m\rangle^2}{v}, \tag{7.2}$$

is scaled variance of mass m in a large subsystem (much smaller than the system) of volume v with $\rho = \langle m\rangle/v$ is average local mass density. The diffusion coefficient $D(\rho)$ and the conductivity $\chi(\rho)$ are suitably defined on a hydrodynamic level from diffusive current $J_D = -D(\rho)\partial\rho/\partial x$ and drift current $J_d = \chi(\rho)F$, respectively, where $\partial\rho/\partial x$ is gradient in local mass density and F is the magnitude of a small biasing force coupled locally to conserved mass variable and applied in a particular direction. For all the processes considered in this paper, we find bulk diffusion coefficient $D(\rho) =$ const. and conductivity $\chi(\rho) \propto \rho^2$, indicating that the processes, on hydrodynamic level, belong to the class of Kipnis-Marchioro-Presutti (KMP) processes on a ring [32]. Moreover, we use the two transport coefficients to find probabilities of large deviations of mass in a subsystem in the framework of recently developed macroscopic fluctuation theory (MFT) [46, 122]. The mass large-deviation functions (LDFs) completely agree with that in Refs. [50, 97], which were derived earlier using an additivity property.

We have further extended this method for asymmetric versions of the models and have shown that, these models defined on a ring allow a consistent hydrodynamic description within the framework of MFT.

The chapter is organised as follows. In section 7.1, we present a linear-response analysis around a nonequilibrium steady state, which is implemented to calculate the transport coefficients in the model-systems discussed later. We introduce, in section 7.2 (symmetric versions) and section 7.4 (asymmetric versions), a broad class of conserved-mass transport processes (called models I, II and III) and derive hydrodynamics of these systems in terms of two transport coefficients - the diffusion coefficient and the conductivity. In section 7.3 and 7.4, we discuss how the density large deviation functions in all these models can be calculated using a macroscopic fluctuation theory. In section 7.5, we summarize with some concluding remarks.

In the following section, we explain the general theory. In the subsequent sections we will explicitly calculate the two transport coefficients, $D(\rho)$ and $\chi(\rho)$, in a broad class of conserved-mass transport processes. Remarkably, in all cases studied here, we find that the two transport coefficients obey an Einstein relation Eq. (7.1). We present below the details of computations for different models separately.

7.1. Theory: Linear Response around nonequilibrium steady states

Before proceeding to the calculations of the transport coefficients in the nonequilibrium mass transport processes mentioned in the previous section, we first present a proof of the Einstein relation (ER), which is valid in or, strictly speaking, around equilibrium state of a system, in the limit of an external force vanishingly small. In equilibrium, an external force field $\vec{F}$ (here taken to be constant, for simplicity), or equivalently an external potential, can be directly related to chemical potential of the system. For example, consider a one dimensional system whose two halves are kept at two different external potentials, say, first half at potential V_1 and second half at potential V_2 where $V_2 - V_1 = \Delta V = -\int F dx$ with the force field $\vec{F} = F\hat{x}$. The fact that effective chemical potentials of the two halves equalize implies

$$\mu(\rho_1) + V_1 = \mu(\rho_2) + V_2,$$

where ρ_1 and ρ_2 are densities of the first and second halves, respectively, $\mu(\rho) = df/d\rho$ is chemical potential (canonical) and $f(\rho)$ free energy density (canonical) in the absence of any external potential. In other words, across a spatial interval Δx, we have the following relation $\Delta\mu/\Delta x = -\Delta V/\Delta x = F$, or

$$\frac{d\mu}{dx} = F, \tag{7.3}$$

in the limit of $\Delta x \to 0$. Now, in the limit of small force $F \to 0$, drift current $J_d = \chi(\rho)F$ due to the force F and the diffusion current $J_D = -D(\rho)d\rho/dx$ must balance each other so that there is no net current in the system. That is, we must have $J_d + J_D = 0$, which, along with the equality $F = d\mu/dx = (d\mu/d\rho)(d\rho/dx)$ [from Eq. (7.3)] and the equilibrium fluctuation-response relation between compressibility and fluctuation $d\rho/d\mu = \sigma^2(\rho)$ [Eq. (6.2)], immediately leads to the ER.

On the other hand, in nonequilibrium, though detailed balance is violated on a microscopic level, the macroscopic mass current in the steady state could still be zero, e.g., in the case of the mass-transport processes with symmetric mass transfer rules. In that case, one would perhaps expect, for a suitably chosen biasing force, an ER even in nonequilibrium. Interestingly, we see later that an ER holds in the cases of both symmetric and asymmetric mass transfers. The issue essentially revolves around the crucial question whether Eq. (7.3) would hold in such cases, which could be addressed by checking if there is an ER. In fact, provided it holds, an ER would then imply a LDF of the form as in Eq. (6.3) where $f''(\rho) = D(\rho)/\chi(\rho)$.

To explore the issue further, we perform a linear-response analysis of the conserved-mass transport processes in the presence of a small constant biasing force field $\vec{F} = F\hat{x}$, which is now applied in the system, with $\hat{x}$ being a unit vector along $+ve$ x axis. The force field $\vec{F}$, somewhat like a gravitational one, is conjugate to the conserved-mass variables (external force is coupled to local masses at the individual sites) and is chosen as following. The biasing force $\vec{F}$ modifies the original mass transfer rates $c_{i\rightarrow j}$, from site i to j, to biased rates $c^F_{i\rightarrow j}$ (which are now effectively asymmetric) [46],

$$c^F_{i\rightarrow j} = c_{i\rightarrow j}\Phi(\Delta e_i), \tag{7.4}$$

where $\Phi(\Delta e_i) > 0$ is non-negative function of

$$\Delta e_i = \Delta m_{i\rightarrow j}(\vec{F}.\delta\vec{x}_{ij}). \tag{7.5}$$

The quantity Δe can be physically interpreted as extra energy cost (due to the biasing force $\vec{F}$), for transferring or displacing mass $\Delta m_{i\rightarrow j}$ from site i to j in a particular direction with the mass displacement vector $\delta\vec{x}_{ij} = (j-i)a\hat{x}$ and a being the lattice constant. We explicitly write the lattice constant, which would be required later for taking diffusive scaling limit. Clearly, $\Phi|_{F=0} = 1$ as $c^{F=0}_{i\rightarrow j} = c_{i\rightarrow j}$.

In the case of only nearest-neighbor mass transfer (more generalized version is described below), the mass displacement vector $\delta\vec{x}_{ij}$ can take, depending on the direction of the mass transfer, one of the two values $\delta\vec{x} = \pm a\hat{x}$. Consequently, the form of rates in Eq. (7.4) makes the modified forward and backward mass-transfer rates across a bond asymmetric and therefore induces a small net current in the system.

To check the ER, we consider, somewhat analogous to equilibrium, the function Φ to have a form $\Phi(\Delta e) = \exp(\Delta e/2)$ [46]. However, note that, in the following linear analysis for small force F where we require only the leading order term $\mathcal{O}(F)$ (or $\mathcal{O}(\Delta e)$), the whole analysis goes through even for a general functional form of Φ. We expand Φ in $\mathcal{O}(F)$,

$$\Phi(\Delta e_i) \simeq 1 + \left[\frac{d\Phi}{d(\Delta e)}\right]_{\Delta e=0} \Delta e_i = 1 + \frac{1}{2}\Delta m_{i\rightarrow j}(\vec{F}.\delta\vec{x}_{ij}). \tag{7.6}$$

For example, see the biased mass-transfer rates $c^F_{i\to j}$ as in Eqs. (7.23) and (7.24). In the above equation, without any loss of generality, we put $2[d\Phi/d\Delta e]_{\Delta e=0} = 1$, which essentially implies a rescaling of the applied force $F \to [2d\Phi/d(\Delta e)]_{\Delta e=0} \times F$.

It is possible that several fractions $\Delta m_{i_n \to j_{n'}}$, where $n = 1, 2, \dots, K$ and $n' = 1, 2, \dots, K'$, of masses from K number of sites $\{i_n\} \equiv i_1, i_2, \dots i_K$ are transferred, at the same instant of time, to K' number of sites $\{j'_n\} \equiv \{j_1, j_2, \dots j_{K'}\}$. For example, see the modified rates for Model I in Eq. (7.10) where $K = 1$ and $K' = 2$ and in Eq. (7.19) where $K = K' = L$. The original rate $c_{\{i_n\}\to\{j_{n'}\}}$, for mass transfer from sites $\{i_n\}$ to $\{j_{n'}\}$, and the corresponding modified biased rate $c^F_{\{i_n\}\to\{j_{n'}\}}$ are related as

$$c^F_{\{i_n\}\to\{j_{n'}\}} = c_{\{i_n\}\to\{j_{n'}\}} \Phi(\Delta e), \tag{7.7}$$

where the total extra energy cost, due to the biasing, can be written by summing over all individual energy costs corresponding to each and every pair $\langle n, n' \rangle$ of departure site n and destination site n' as

$$\Delta e = \sum_{\langle n,n' \rangle} \Delta m_{i_n \to j_{n'}} (\vec{F}.\delta\vec{x}_{i_n j_{n'}}). \tag{7.8}$$

In the next, we use this modified biased rate $c^F_{\{i_n\}\to\{j_{n'}\}}$ [as in Eq. (7.7)] alongwith Eqs. (7.6) and (7.8) for the three models (I, II, III) to derive a hydrodynamic equation like in Eq. 6.5 and hence, in turn, we compute the diffusivity $D(\rho)$ and the conductivity $\chi(\rho)$.

7.2. models and results: symmetric mass transfers

In this section, we recapitulate the symmetric versions of the models, first in the absence of any biasing force, where masses are transferred symmetrically, without any preferential direction, to the nearest neighbors. Consequently, there is no net mass current in the systems. However, it is important to note that, even in that case, detailed balance condition is still not satisfied. In fact, it would be quite instructive to explicitly show that, for generic values of parameters in the models, Kolmogorov criterion and therefore detailed balance is strongly violated, in the sense that, for a transition (say, forward) from one configuration to another while mass being transferred from a site to its neighbor, the corresponding reverse path of transition may not exist.

Therefore, even in the absence of any biasing force, the system eventually reaches a steady state, which is inherently far from equilibrium, and cannot be described by the equilibrium Boltzmann-Gibbs distribution. To calculate conductivity in such a nonequilibrium steady state, we need to apply a biasing (constant, for simplicity) force field, which would essentially modify the original mass-transfer rates in the systems, inducing a mass current, and then we calculate the current in the limit of biasing force being small.

7.2.1. Model I

This particular class of models has been introduced to study mass transport processes accounting for stickiness of masses while fragmenting and diffusing [31]. These processes are variants of various previously studied mass transport processes, such as random average processes (RAP), etc. [27, 28, 81].

Random Sequential Update

In Model I with random sequential update (RSU), three sites are updated simultaneously where two random fractions of the chipped-off mass from site i are shared randomly with the nearest neighbour sites $i-1$ and $i+1$. The stochastic time evolution of mass $m_i(t)$ at time t after an infinitesimal time dt can be written as

$$m_i(t+dt) = \begin{cases} \lambda m_i(t) & \text{prob. } dt \\ m_i(t) + \tilde{\lambda} r_{i-1} m_{i-1}(t) & \text{prob. } dt \\ m_i(t) + \tilde{\lambda}\tilde{r}_{i+1} m_{i+1}(t) & \text{prob. } dt \\ m_i(t) & \text{prob. } (1-3dt) \end{cases} \tag{7.9}$$

where $r_j \in (0,1)$s are independent and identically distributed (i.i.d.) random variables, having a probability density $\phi(r)$ and $\tilde{\lambda} = 1-\lambda$ and $\tilde{r}_{i+1} = 1 - r_{i+1}$. Throughout the paper, we denote the first and the second moments of $\phi(r)$ as

$$\theta_1 = \int_0^1 r\phi(r)dr; \;\; \theta_2 = \int_0^1 r^2\phi(r)dr,$$

respectively. Note that, if the probability density $\phi(r)$ is not symmetric around $r = 1/2$, it can be shown that, in the hydrodynamic equation for density field, drift dominates diffusion unless the asymmetry is small and comparable to the diffusive contribution. In that case, the analysis would lead to hyperbolic hydrodynamic equations for density field (hydrodynamics of such systems are discussed in section 7.4). Here we consider the density function $\phi(r)$, which is symmetric around $r = 1/2$, i. e., $\phi(r) = \phi(1-r)$, thus $\theta_1 = 1/2$ is taken throughout; but the probability density $\phi(r)$ is otherwise arbitrary.

Dynamics when $F \neq 0$. – Let us now bias the system by applying a small constant biasing force field $\vec{F} = F\hat{x}$, say, along the clockwise direction, which affects the mass transfer rates according to Eq. (7.7). Since, at every instant of time, two fractions of the chipped-off mass from site i could be simultaneously transferred, to the two neighboring sites $i \pm 1$, the modified biased rate in this case is written as

$$c^F_{i\to\{i+1,i-1\}} = c_{i\to\{i+1,i-1\}}\left[1 + \frac{\Delta e_i}{2}\right], \tag{7.10}$$

where $c_{i\to\{i+1,i-1\}} = 1$ and $\Delta e_i = Fa(\Delta m_{i\to i+1} - \Delta m_{i\to i-1})$ with $\Delta m_{i\to i+1} = \tilde{\lambda} r_i m_i$ and $\Delta m_{i\to i-1} = \tilde{\lambda}(1-r_i)m_i$. For notational simplicity, we denote the biased rate as $c^F_{i\to\{i+1,i-1\}} \equiv c^F_i$, which can be explicitly written as $c^F_i = 1 + \tilde{\lambda}(2r_i - 1)m_i Fa/2$, with $\tilde{\lambda} = 1 - \lambda$. We now write the modified dynamics,

$$m_i(t+dt) = \begin{cases} \lambda m_i(t), \quad \text{prob. } c^F_i dt \\ m_i(t) + \tilde{\lambda} r_{i-1} m_{i-1}(t), \; \text{prob. } c^F_{i-1} dt \\ m_i(t) + \tilde{\lambda} \tilde{r}_{i+1} m_{i+1}(t), \; \text{prob. } c^F_{i+1} dt \\ m_i(t), \; \text{prob. } (1 - (c^F_i + c^F_{i-1} + c^F_{i+1})dt). \end{cases} \tag{7.11}$$

Consequently the time evolution of the first moment of mass $m_i(t)$ in the infinitesimal time dt can be written as,

$$\begin{aligned} \langle m_i(t+dt)\rangle &= \langle \lambda m_i(t) c^F_i\rangle dt \\ &+ \langle [m_i(t) + \tilde{\lambda} r_{i-1} m_{i-1}(t)] c^F_{i-1}\rangle dt \\ &+ \langle [m_i(t) + \tilde{\lambda} \tilde{r}_{i+1} m_{i+1}(t)] c^F_{i+1}\rangle dt \\ &+ \langle m_i(t)[1 - (c^F_i + c^F_{i-1} + c^F_{i+1})dt]\rangle \end{aligned}$$

After simplifying the above expression, the time evolution of average mass, or mass density, $\langle m_i \rangle \equiv \rho_i$ at site i, can be rewritten as

$$\begin{aligned}
\frac{d\rho_i}{dt} &= \tilde{\lambda}\langle r_{i-1} m_{i-1} c^F_{i-1} + (1-r_{i+1}) m_{i+1} c^F_{i+1} - m_i c^F_i \rangle \\
&= \frac{\tilde{\lambda}}{2}(\rho_{i-1} + \rho_{i+1} - 2\rho_i) \\
&\quad + \frac{\tilde{\lambda}^2}{2}(2\theta_2 - 1/2)[\langle m^2_{i-1}\rangle Fa - \langle m^2_{i+1}\rangle Fa]. \qquad (7.12)
\end{aligned}$$

Note that the time evolution of the first moment of local mass, i.e., the density $\rho_i = \langle m_i \rangle$, depends on the second moments $\langle m^2_{i\pm1}\rangle$ of neighboring masses, and so on. Thus the hierarchy between the local density and the local fluctuation does not close.

Hydrodynamics– However, we are interested in the hydrodynamic description of the density field at large space and time scales, called diffusive scaling limit as described below. Importantly, on the large spatio-temporal scales, local observables are expected to be slowly varying functions of space and time. Therefore, we could safely assume that a local steady state is achieved throughout the system such that average of any local observable $g(m_i)$ could be replaced by its exact local steady-state average $\langle g(m_i)\rangle_{st}$, which in that case would be a function of the local density ρ_i only. In other words, we assume $\langle g(m_i)\rangle \approx \langle g(m_i)\rangle_{st}$. Thus, for the average of the quantity $g(m_i) = m_i^2$, i.e., the second moment of local mass, we have replaced the average by the its local steady-state average,

$$\langle m_i^2 \rangle \approx \langle m_i^2 \rangle_{st} = \frac{1}{1-2\tilde{\lambda}\theta_2}\rho_i^2. \qquad (7.13)$$

The above steady-state expression of the second moment has been exactly calculated before in Ref. [97]. Now substituting Eq. (7.13) in Eq. (7.12) and then taking the diffusive scaling limit of Eq. (7.12), $i \to x = i/L$, $t \to \tau = t/L^2$ and $a \to 1/L$, we obtain the hydrodynamic equation for the density field, $\partial_\tau \rho(x,\tau) + \partial_x J = 0$, where current $J(\rho(x,\tau))$ is given by

$$J = \frac{\tilde{\lambda}^2}{2}\frac{4\theta_2 - 1}{1-2\tilde{\lambda}\theta_2}\rho^2 F - \frac{\tilde{\lambda}}{2}\frac{\partial\rho}{\partial x}. \qquad (7.14)$$

In the above equation, we break the current $J = J_d + J_D$ into two parts, drift current $J_d = [\tilde{\lambda}^2(4\theta_2 - 1)/2(1-2\tilde{\lambda}\theta_2)]\rho^2 F$ and diffusive current $J_D = -(\tilde{\lambda}/2)(\partial\rho/\partial x)$, to identify the conductivity and the diffusion coefficient, respectively, as

$$\chi(\rho) = \frac{\tilde{\lambda}^2}{2}\frac{(4\theta_2 - 1)}{(1 - 2\tilde{\lambda}\theta_2)}\rho^2 \; ; \; D(\rho) = \frac{\tilde{\lambda}}{2}. \tag{7.15}$$

Now the scaled variance $\sigma^2(\rho)$ of sub-system mass [as defined in Eq. (7.2)] can be calculated by summing over the microscopic correlation function $c(n) = \langle m_i m_{i+n}\rangle - \rho^2$,

$$\sigma^2(\rho) = \sum_{n=-\infty}^{\infty} c(n) = \frac{\tilde{\lambda}(4\theta_2 - 1)}{(1 - 2\tilde{\lambda}\theta_2)}\rho^2 \equiv \frac{\rho^2}{\eta}, \tag{7.16}$$

where $c(n)$ has been exactly calculated in Ref. [97],

$$\begin{aligned} c(n) &= \frac{2\tilde{\lambda}\theta_2}{(1 - 2\tilde{\lambda}\theta_2)}\rho^2 && \text{for } n = 0 \\ &= -\frac{\tilde{\lambda}}{2}\frac{(1 - 2\theta_2)}{(1 - 2\tilde{\lambda}\theta_2)}\rho^2 && \text{for } n = 1 \\ &= 0 && \text{for } n \geq 2, \end{aligned}$$

and $\eta = (1 - 2\tilde{\lambda}\theta_2)/\tilde{\lambda}(4\theta_2 - 1)$. Using Eqs. (7.15) and (7.16), one can readily verify that the ER as in Eq. (7.1) is indeed satisfied. We emphasize that the nearest- neighbor spatial correlations here (and in the models discussed later) are actually finite and our hydrodynamic analysis takes into account the effects of the finite microscopic spatial correlations.

Parallel Update

In Model I with parallel update (PU), fractions of masses to be transferred to the two nearest neighbors are the same as in the case of random sequential update. However, at a discreet time t, the mass variables at all sites are updated simultaneously according to the following rule,

$$m_i(t+1) = \lambda m_i(t) + \tilde{\lambda} r_{i-1} m_{i-1}(t) + \tilde{\lambda}\tilde{r}_{i+1} m_{i+1}(t), \tag{7.17}$$

where $\tilde{\lambda} = 1 - \lambda$, $\tilde{r}_i = 1 - r_i$ and $r_i \in (0,1)$ is a symmetrically distributed random variable, having a probability density $\phi(r_i)$.

The time evolution equation in the configuration space $\{m_i\} \equiv \{m_1, m_2, \ldots, m_L\}$ can be written as

$$\mathcal{P}[\{m_i\}, t+1] = \left[\prod_j \int dm_j\right] \times \Gamma[\{m_j\} \to \{m_i\}] \mathcal{P}[\{m_j\}, t], \tag{7.18}$$

where $\mathcal{P}[\{m_i\}, t]$ is the probability of a configuration $\{m_i\}$ at time t and

$$\Gamma[\{m_j\} \to \{m_i\}] = \prod_i \phi(r_i)$$

is the transition probability, per unit time, from a configuration $\{m_j\}$ to another configuration $\{m_i\}$.

Dynamics when $F \neq 0$. – Let us now consider the process in the presence of an externally applied biasing force, $F \neq 0$. Once the random fractions ($\tilde{\lambda} r_i m_i$ and $\tilde{\lambda}(1 - r_i) m_i$) of mass m_i at a site i are chosen at time t they are transferred, at the next discrete time step, to the nearest neighbour sites $i+1$ and $i-1$, respectively, with probability 1 and this is done simultaneously for all sites. That is, in this case, the mass transfer rate, or the transition probability per unit time, can be written as $c_{\{i_n\} \to \{j_{n'}\}} = 1$, which we modify, in the presence of biasing force, as $c^F_{\{i_n\} \to \{j_{n'}\}} = c_{\{i_n\} \to \{j_{n'}\}} \prod_i \exp(\Delta e_i/2)$, according to Eq. (7.7). Here we put $\Phi(\Delta e) = \exp(\Delta e/2)$ with $\Delta e = \sum_i \Delta e_i$ and $\Delta e_i = Fa(\Delta m_{i \to i+1} - \Delta m_{i \to i-1}) = \tilde{\lambda}(2r_i - 1) m_i F a$. The time evolution Eq. (7.18) can now be written by replacing the original transition probability Γ with the modified one Γ^F,

$$\Gamma^F[\{m_j\} \to \{m_i\}] = \prod_j \left[\frac{\phi(r_j) e^{\Delta e_j/2}}{\gamma(m_j, F)}\right], \tag{7.19}$$

where $\gamma(m_j, F)$ is a normalization constant, ensuring that the transition probability $\Gamma^F[.]$ is suitably assigned from a normalized probability density function where $(\prod_j \int dr_j)\Gamma^F = 1$. As the probability density $\phi(r)$ is considered to be symmetric about $r = 1/2$, we have the following expansion in powers of F,

$$\begin{aligned} \gamma(m_i, F) &= \int_0^1 \phi(r_i) e^{\tilde{\lambda}(2r_i-1)m_i Fa/2} dr_i \\ &= 1 + \frac{(\tilde{\lambda} m_i)^2 \theta_2}{8} (Fa)^2 + \ldots, \end{aligned}$$

implying that the leading order term is quadratic $\mathcal{O}(F^2)$ in the biasing force F and therefore, to linear order of F, we can take $\gamma(m_i, F) \approx 1$ in the following analysis (see also section 7.2.2).

The expression for the average of mass m_i at site i can now be written as

$$\begin{aligned} \langle m_i(t+1) \rangle &= \left[\prod_j \int dm_j \right] m_i \mathcal{P}[\{m_j\}, t+1] \\ &= \left\langle \left[\lambda m_i(t) + \tilde{\lambda} r_{i-1} m_{i-1}(t) + \tilde{\lambda}(1 - r_{i+1}) m_{i+1}(t) \right] \right\rangle, \end{aligned}$$

where the angular brackets $\langle \cdot \rangle$ denote average over both random numbers $\{r_j\}$ and the mass variables $\{m_j\}$. Explicitly writing the terms, we get

$$\begin{aligned} \langle m_i(t+1) \rangle = & \left\langle \lambda m_i(t) \int \frac{\phi(r) e^{\tilde{\lambda}(2r-1)m_i Fa/2}}{\gamma(m_i, F)} dr \right\rangle \\ & + \left\langle \tilde{\lambda} m_{i-1}(t) \int r \frac{\phi(r) e^{\tilde{\lambda}(2r-1)m_{i-1} Fa/2}}{\gamma(m_{i-1}, F)} dr \right\rangle \\ & + \left\langle \tilde{\lambda} m_{i+1}(t) \int (1-r) \frac{\phi(r) e^{\tilde{\lambda}(2r-1)m_{i+1} Fa/2}}{\gamma(m_{i+1}, F)} dr \right\rangle, \end{aligned}$$

which, in leading order in F, leads to

$$\begin{aligned} \rho_i(t+1) - \rho_i(t) = & \frac{\tilde{\lambda}}{2} (\rho_{i-1} + \rho_{i+1} - 2\rho_i) \\ & + \frac{\tilde{\lambda}^2}{2} (2\theta_2 - 1/2) [\langle m_{i-1}^2 \rangle Fa - \langle m_{i+1}^2 \rangle Fa]. \end{aligned}$$

Hydrodynamics.– Now taking the diffusive scaling limit $i \to x = i/L$, $t \to \tau = t/L^2$ and $a \to 1/L$ and substituting $\langle m_i^2 \rangle$ by the following expression of second moment [97], within the assumption of local steady-state,

$$\langle m_i^2 \rangle_{st} = \frac{1}{\epsilon + (1-\epsilon)\sqrt{\frac{\kappa-1}{\kappa+1}}} \rho_i^2,$$

we obtain the hydrodynamic equation for the density field, $\partial_\tau \rho(x,\tau) + \partial_x (J_d + J_D) = 0$, where the drift current $J_d(\rho(x,\tau))$ and the diffusive current $J_D(\rho(x,\tau))$ are given by

$$J_d = \frac{\tilde{\lambda}^2}{2} \frac{4\theta_2 - 1}{\epsilon + (1-\epsilon)\sqrt{\frac{\kappa-1}{\kappa+1}}} \rho^2 F \ ; \ J_D = -\frac{\tilde{\lambda}}{2} \frac{\partial \rho}{\partial x}, \tag{7.20}$$

respectively. Then, the conductivity $\chi(\rho)$ and the diffusion coefficient $D(\rho)$ can be expressed as

$$\chi(\rho) = \frac{\tilde{\lambda}^2}{2} \frac{4\theta_2 - 1}{\epsilon + (1-\epsilon)\sqrt{\frac{\kappa-1}{\kappa+1}}} \rho^2 \ ; \ D(\rho) = \frac{\tilde{\lambda}}{2}, \tag{7.21}$$

where $\epsilon = 2 - 4\theta_2$ and $\kappa = (1+\lambda)/(1-\lambda)$. The Einstein relation Eq. (7.1) can be immediately verified using the expression of the scaled variance,

$$\sigma^2(\rho) = \frac{\tilde{\lambda}(4\theta_2 - 1)}{\epsilon + (1-\epsilon)\sqrt{\frac{\kappa-1}{\kappa+1}}} \rho^2,$$

which was exactly calculated earlier in Ref. [97]. We mention here that the microscopic spatial correlations, as in the case of Model I (RSU), are also finite and have been accounted exactly in the above analysis.

7.2.2. Model II

The class of models studied in this section is a generalized version of previously known Hammersley process [78] and a variant of random average processes [81]. These models were studied in the past to understand force fluctuations in granular beads [79, 80] and dynamics of driven interacting particles on a ring [27, 83], etc.

Random Sequential Update

In Model II with random sequential update, two nearest neighbor sites are updated in an infinitesimal time dt: A random fraction of mass at site i is chipped off and transferred either to site $i-1$ or to site $i+1$, each with probability $(1/2)dt$, i.e., the mass transfer rates $c_{i\rightarrow i-1} = 1/2$ and $c_{i\rightarrow i+1} = 1/2$.

Dynamics when $F \neq 0$. – However, in the presence of a biasing force $F \neq 0$, the dynamics is modified as

$$m_i(t+dt) = \begin{cases} \lambda m_i(t) + \tilde{\lambda}(1-r_i)m_i(t) & \text{prob. } dt \\ m_i(t) + \tilde{\lambda} r_{i+1} m_{i+1}(t) & \text{prob. } c^F_{i+1\to i}dt \\ m_i(t) + \tilde{\lambda} r_{i-1} m_{i-1}(t) & \text{prob. } c^F_{i-1\to i}dt \\ m_i(t) & \text{prob. } [1-(1+c^F_{i+1\to i}+c^F_{i-1\to i})dt] \end{cases} \tag{7.22}$$

where $\tilde{\lambda} = 1-\lambda$ and the modified mass transfer rates, $c^F_{i\to i\pm1} = \exp(\pm\Delta m_{i\to i\pm1}Fa/2)$ with transported mass $\Delta m_{i\to i\pm1} = \tilde{\lambda} r_i m_i(t)$, can be written, in leading order of F, as

$$c_{i-1\to i} = \frac{1}{2} + \frac{\tilde{\lambda}}{4} r_{i-1} m_{i-1} Fa, \tag{7.23}$$

$$c_{i+1\to i} = \frac{1}{2} - \frac{\tilde{\lambda}}{4} r_{i+1} m_{i+1} Fa. \tag{7.24}$$

Clearly, $F = 0$ reproduces the original unbiased dynamics. Now, the time evolution of average mass or density at site i is given by,

$$\frac{d\langle m_i\rangle}{dt} = \tilde{\lambda}\langle [r_{i-1}m_{i-1}c^F_{i-1\to i} + r_{i+1}m_{i+1}c^F_{i+1\to i} - r_i m_i c^F_i]\rangle$$

which, in leading order of F, can be written as

$$\frac{d\rho_i}{dt} = \frac{\tilde{\lambda}}{2}\theta_1(\rho_{i-1}+\rho_{i+1}-2\rho_i) + \frac{\tilde{\lambda}^2}{4}\theta_2[\langle m^2_{i-1}\rangle Fa - \langle m^2_{i+1}\rangle Fa].$$

Hydrodynamics– Taking the diffusive scaling limit $i \to x = i/L$, $t \to \tau = t/L^2$ and $a \to 1/L$ and using the local steady-state expression for the second moment,

$$\langle m_i^2\rangle = \frac{\theta_1}{\theta_1 - \tilde{\lambda}\theta_2}\rho_i^2,$$

we obtain the hydrodynamic equation governing the density field, $\partial_\tau \rho(x,\tau) + \partial_x(J_d + J_D) = 0$.

Here the drift current $J_d(\rho(x,\tau))$ and the diffusive current $J_D(\rho(x,\tau))$ are given by

$$J_d = \frac{\tilde{\lambda}^2}{2}\frac{\theta_1\theta_2}{\theta_1 - \tilde{\lambda}\theta_2}\rho^2 F \; ; \; J_D = -\frac{\tilde{\lambda}}{2}\theta_1\frac{\partial\rho}{\partial x}. \tag{7.25}$$

Therefore, the conductivity $\chi(\rho)$ and the diffusion coefficient $D(\rho)$ are given by

$$\chi(\rho) = \frac{\tilde{\lambda}^2}{2}\frac{\theta_1\theta_2}{\theta_1 - \tilde{\lambda}\theta_2}\rho^2 \; ; \; D(\rho) = \frac{\tilde{\lambda}}{2}\theta_1. \tag{7.26}$$

The Einstein relation Eq. (7.1) can now be verified by using the expression of scaled variance,

$$\sigma^2(\rho) = \frac{\tilde{\lambda}\theta_2}{\theta_1 - \tilde{\lambda}\theta_2}\rho^2,$$

which was obtained earlier in Ref. [97].

Parallel Update

In Model II with parallel update, at each discrete time step, masses at all sites are updated simultaneously according to the following rule,

$$\begin{aligned} m_i(t+1) &= (1-\tilde{\lambda}r_i)m_i(t) + \tilde{\lambda}r_{i+1}m_{i+1}(t) \\ &+ \tilde{\lambda}[s_{i-1}r_{i-1}m_{i-1}(t) - s_{i+1}r_{i+1}m_{i+1}(t)] \end{aligned} \tag{7.27}$$

where $\tilde{\lambda} = 1-\lambda$. Here we have introduced a set of discrete i.i.d. random variables $\{s_i\}$: When the chipped-off fraction of mass moves to right, $s_i = 1$ and otherwise $s_i = 0$. As each of the values $s_i = 0$ and $s_i = 1$ occurs with probability 1/2, we have $\langle s_i^n \rangle = 1/2$ for $n > 0$.

Dynamics when $F \neq 0$. – In the presence of a biasing force $F \neq 0$, the transition probability $\Gamma[\{m_j\} \to \{m_k\}]$ from a configuration $\{m_j\}$ to another configuration $\{m_k\}$ is modified as

$$\Gamma^F[\{m_j\} \to \{m_k\}] = \prod_i \left[\frac{1}{\gamma(m_i,F)}\phi(r_i)e^{\Delta e_i/2}\right], \tag{7.28}$$

where $\Delta e_i = [s_i - (1-s_i)]\tilde{\lambda} r_i m_i Fa$ and the normalization factor

$$\begin{aligned}\gamma(m_i, F) &= \Sigma_{s_i} P(s_i) \int_0^1 \phi(r_i) e^{(2s_i-1)\tilde{\lambda} r_i m_i Fa/2} dr_i \\ &= 1 + \frac{(\tilde{\lambda} m)^2 \theta_2}{8} (Fa)^2 + \cdots \approx 1,\end{aligned}$$

to the linear order of F. The time evolution of the average mass or density at site i is given by

$$\begin{aligned}\langle m_i(t+1)\rangle &= \Big\langle (1-\tilde{\lambda} r_i) m_i(t) \Big\rangle + \Big\langle \tilde{\lambda} s_{i-1} r_{i-1} m_{i-1}(t) \Big\rangle \\ &\quad + \Big\langle \tilde{\lambda}(1-s_{i+1}) r_{i+1} m_{i+1}(t) \Big\rangle \qquad (7.29)\end{aligned}$$

where the above angular brackets denote averaging over all three random variables, $\{r_i\}$, $\{s_i\}$ and $\{m_i\}$. Equivalently, we can write

$$\begin{aligned}&\langle m_i(t+1)\rangle = \left\langle (1-\tilde{\lambda} r_i) m_i(t) \int \frac{\phi(r_i) e^{\Delta e_i/2}}{\gamma(m_i, F)} dr_i \right\rangle \\ &+ \left\langle \tilde{\lambda} s_{i-1} r_{i-1} m_{i-1}(t) \int \frac{\phi(r_{i-1}) e^{\Delta e_{i-1}/2}}{\gamma(m_{i-1}, F)} dr_{i-1} \right\rangle \qquad (7.30) \\ &+ \left\langle \tilde{\lambda}(1-s_{i+1}) r_{i+1} m_{i+1}(t) \int \frac{\phi(r_{i+1}) e^{\Delta e_{i+1}/2}}{\gamma(m_{i+1}, F)} dr_{i+1} \right\rangle,\end{aligned}$$

where, in the second step, we have explicitly written the averaging over the i.i.d. random variables $\{r_i\}$. Next, doing the averaging over the i.i.d. random variables $\{s_i\}$, we obtain, in linear order of F, the time evolution equation for density $\rho_i = \langle m_i \rangle$ at site i,

$$\begin{aligned}\rho_i(t+1) - \rho_i(t) &= \frac{\tilde{\lambda}}{2} \theta_1 (\rho_{i-1} + \rho_{i+1} - 2\rho_i) \\ &+ \frac{\tilde{\lambda}^2}{4} \theta_2 [\langle m_{i-1}^2 \rangle Fa - \langle m_{i+1}^2 \rangle Fa]. \qquad (7.31)\end{aligned}$$

Hydrodynamics.– Now rescaling the space and time by $i \to x = i/L$, $t \to \tau = t/L^2$ and $a \to 1/L$, and using the expression for second moment of m_i in the local steady state [97],

$$\langle m_i^2 \rangle = \frac{\sqrt{\alpha}}{1-(1-\lambda)\epsilon} \rho_i^2,$$

we obtain the hydrodynamic equation of density field, $\partial_\tau \rho(x,\tau) + \partial_x(J_d + J_D) = 0$ where the drift $J_d(\rho(x,\tau))$ and diffusive currents $J_D(\rho(x,\tau))$, respectively, can be written as

$$J_D = -\frac{\tilde{\lambda}}{2}\theta_1 \frac{\partial \rho}{\partial x} \; ; \; J_d = \frac{\tilde{\lambda}^2}{2}\theta_2 \frac{\sqrt{\alpha}}{1-(1-\lambda)\epsilon}\rho^2 F.$$

The above expressions of currents immediately gives the diffusion coefficients and the conductivity as a function of density,

$$\chi(\rho) = \frac{\tilde{\lambda}^2}{2}\theta_2 \frac{\sqrt{\alpha}}{1-(1-\lambda)\epsilon}\rho^2 \; ; \; D(\rho) = \frac{\tilde{\lambda}}{2}\theta_1, \tag{7.32}$$

respectively, with $\epsilon = \theta_2/\theta_1$, $\alpha = (1+\lambda)/2$. Now, by using the exact expression of scaled variance [97],

$$\sigma^2(\rho) = \frac{\tilde{\lambda}\sqrt{\alpha}\epsilon}{1-\tilde{\lambda}\epsilon},$$

one can verify that the ER as in Eq. (7.1) is indeed satisfied. Note that, as in the case of Model I, the microscopic spatial correlations are also finite here and have been taken into account in deriving hydrodynamics.

7.2.3. Model III

This class of models have been studied intensively in the past to understand distribution of wealth in a population [76, 86, 117]. In this model, each site keeps a λ fraction (usually called "saving propensity" in the literature) of its own mass, and the remaining mass of two neighboring sites are mixed and are distributed randomly among themselves. Here we study only the random sequential update dynamics, which can be written in an infinitesimal time dt as follows,

$$m_i(t+dt) = \begin{cases} \lambda m_i(t) + \tilde{\lambda} r_i [m_i(t) + m_{i+1}(t)] & \text{prob. } c_{i+1\to i}dt \\ \lambda m_i(t) + \tilde{\lambda} \tilde{r}_{i-1} [m_i(t) + m_{i-1}(t)] & \text{prob. } c_{i-1\to i}dt \\ m_i(t) & \text{prob. } [1-(c_{i+1\to i} + c_{i-1\to i})dt] \end{cases} \tag{7.33}$$

where $\tilde{\lambda} = 1 - \lambda$, $\tilde{r}_i = 1 - r_i$, $m_i(t)$ is mass at site i at time t, $r_i \in (0,1)$ is a i.i.d. random variable having a probability density $\phi(r_i)$ (symmetric around 1/2) and the mass transfer rate $c_{i\to j} = 1$ (here $j = i \pm 1$).

Dynamics when $F \neq 0$. – In the presence of a biasing force, the mass transfer rates are modified as

$$\begin{aligned} c^F_{i+1\to i} &= e^{-\Delta m_{i+1\to i}Fa/2} \approx 1 - \frac{1}{2}\Delta m_{i+1\to i}Fa, \\ c^F_{i-1\to i} &= e^{\Delta m_{i-1\to i}Fa/2} \approx 1 + \frac{1}{2}\Delta m_{i-1\to i}Fa \end{aligned}$$

where

$$\Delta m_{i+1\to i} = \tilde{\lambda} r_i m_{i+1}(t) - \tilde{\lambda}(1 - r_i)m_i(t) \tag{7.34}$$

and

$$\Delta m_{i-1\to i} = \tilde{\lambda}(1 - r_{i-1})m_{i-1} - \tilde{\lambda} r_{i-1} m_i(t). \tag{7.35}$$

The time evolution of the first moment of local mass or density $\rho_i = \langle m_i \rangle$ at site i can be written as

$$\begin{aligned} \langle m_i(t+dt)\rangle = &\Big\langle [\lambda m_i(t) + \tilde{\lambda} r_i (m_i(t) + m_{i+1}(t))] c^F_{i+1\to i} dt \Big\rangle \\ &+ \Big\langle [\lambda m_i(t) + \tilde{\lambda}(1 - r_{i-1})(m_i(t) + m_{i-1}(t))] c^F_{i-1\to i} dt \Big\rangle \\ &+ \Big\langle m_i(t)[1 - (c^F_{i-1\to i} + c^F_{i+1\to i})dt] \Big\rangle. \end{aligned}$$

After substituting $\langle m_i \rangle = \rho_i$ and some simplifications, we have the following evolution for density ρ_i,

$$\begin{aligned} \frac{d\rho_i}{dt} &= \frac{\tilde{\lambda}}{2}[\rho_{i+1} - 2\rho_i + \rho_{i-1}] \\ &- \frac{1}{2}\Big\langle (\Delta m^2_{i+1\to i}Fa - \Delta m^2_{i-1\to i}Fa) \Big\rangle, \end{aligned}$$

which leads to

$$\frac{d\rho_i}{dt} = \frac{\tilde{\lambda}}{2}[\rho_{i+1} - 2\rho_i + \rho_{i-1}] - \frac{\tilde{\lambda}^2}{2}\left[\theta_2 \frac{\lambda + 2\tilde{\lambda}\theta_2}{1 - 2\tilde{\lambda}\theta_2}(\rho_{i+1}^2 - \rho_{i-1}^2) - (1 - 2\theta_2)(\rho_{i+1}^2 - \rho_i^2)\right] Fa$$

In the last step, following the assumption of local steady-state, we have used Eqs. (7.34) and (7.35) and, subsequently, used the expression of second moment of local mass as well as the expression of nearest-neighbor mass-mass correlation [97],

$$\langle m_i^2 \rangle = \frac{1 - \tilde{\lambda}(1 - 2\theta_2)}{\lambda + \tilde{\lambda}(1 - 2\theta_2)}\rho_i^2 \; ; \; \langle m_{i-1} m_i \rangle = \rho_i^2.$$

Hydrodynamics.– Finally, we take the diffusive limit, by rescaling space and time as $i \to x = i/L$, $t \to \tau = t/L^2$ and $a \to 1/L$, and obtain the hydrodynamic equation for the density field as $\partial_t \rho(x, \tau) + \partial_x J(\rho(x, \tau)) = 0$ where $J = J_d + J_D$, with

$$J_d(\rho) = \frac{\tilde{\lambda}^2}{2}\frac{4\theta_2 - 1}{1 - 2\tilde{\lambda}\theta_2}\rho^2 \; ; \; J_D(\rho) = -\frac{\tilde{\lambda}}{2}\frac{\partial \rho}{\partial x}.$$

The above functional forms of currents imply that the diffusion coefficient and the conductivity, respectively, have the following expressions,

$$\chi(\rho) = \frac{\tilde{\lambda}^2}{2}\frac{4\theta_2 - 1}{1 - 2\tilde{\lambda}\theta_2}\rho^2 \; ; \; D(\rho) = \frac{\tilde{\lambda}}{2}. \tag{7.36}$$

The ER as in Eq. (7.1) can now be verified by using the previously obtained expression of scaled variance [97],

$$\sigma^2(\rho) = \frac{\tilde{\lambda}(4\theta_2 - 1)}{1 - 2\tilde{\lambda}\theta_2}\rho^2.$$

7.3. Density large deviations

The evolution in Eq. (6.5), in fact, describes the evolution of the average density profile. As mentioned earlier, our microscopic models are, however, stochastic by nature, which gives rise to fluctuations in the density and the associated current fields. According to the macroscopic fluctuation theory (MFT) [46], the fluctuations in these two fields can be introduced by adding a random current field $\zeta(x,\tau)$ to the deterministic one $J(x,\tau)$ as following. The total current can now be written as $j(x,\tau) = J(x,\tau) + \zeta(x,t)$, where $\zeta(x,\tau)$ is a weak Gaussian multiplicative white noise, whose mean is zero and strength depends on local density through conductivity $\chi(\rho)$,

$$\langle \zeta(x,\tau)\rangle = 0\ ;\ \langle \zeta(x,\tau)\zeta(x',\tau')\rangle = \frac{1}{L}\chi(\rho)\delta(x-x')\delta(\tau-\tau').$$

Thus, one obtains the following fluctuating-hydrodynamic time-evolution of the density field,

$$\partial_\tau \rho(x,\tau) + \partial_x\left[-D(\rho)\partial_x\rho(x,\tau) + \chi(\rho)F + \zeta(x,\tau)\right] = 0. \tag{7.37}$$

Starting from the stochastic microscopic dynamics, and using the Markov properties of the evolution, one can actually prove the above stochastic hydrodynamic equation (7.37) [46]. Then, using Eq. (7.37), one can, in principle, find the joint probability of any given time-trajectories of the full density $\rho(x,\tau)$ and current $j(x,\tau)$ profiles, starting from an arbitrary initial condition.

However, here, we are interested in the steady-state probabilities of density large deviations. According to MFT, the probability of an arbitrary density profile $\rho(x)$ in the steady state, which corresponds to Eq. (7.37) with zero external bias $F = 0$, is given by the following large deviation probability $\mathcal{P}[\rho(x)] \approx e^{-\mathcal{F}[\rho(x)]}$, where the large deviation function $\mathcal{F}[\rho(x)]$ satisfies [46]

$$\int dx \left[\partial_x\left(\frac{\delta\mathcal{F}}{\delta\rho}\right)\chi(\rho)\,\partial_x\left(\frac{\delta\mathcal{F}}{\delta\rho}\right) - \frac{\delta\mathcal{F}}{\delta\rho}\partial_x J_D(\rho)\right] = 0. \tag{7.38}$$

After performing a partial integration in the second term, one can readily check that the above equation is satisfied by the LDF $\mathcal{F}[\rho(x)]$ which satisfies the following conditions,

$$\partial_x\left(\frac{\delta\mathcal{F}}{\delta\rho(x)}\right) = \partial_x\left(f'(\rho(x)) - f'(\rho_0)\right), \tag{7.39}$$

$$\frac{1}{f''(\rho)} = \frac{\chi(\rho)}{D(\rho)}. \tag{7.40}$$

Here, ρ_0 is the average or typical local mass density (which in our case turns out to be the same as the global density since the systems are homogeneous) at which the LDF $\mathcal{F}[\rho]$ has a minimum equal to $\mathcal{F}[\rho(x) = \rho_0] = 0$. Equation (7.39), together with this minimum condition, gives the following expression of the LDF,

$$\mathcal{F}[\rho(x)] = \int_{-\infty}^{\infty} dx\{f(\rho) - f(\rho_0) - f'(\rho_0)(\rho - \rho_0)\}. \tag{7.41}$$

Note that the above functional form of the LDF implies the FR as in Eq. (6.2). Now substituting Eq. (6.2) in Eq. (7.40), one immediately obtains the Einstein relation Eq. (7.1). Moreover, using Eqs. (7.40) and (6.2), one can easily see that the LDF in Eq. (7.41) is exactly the same as in Eq. (6.3), which was earlier obtained directly from additivity and the FR Eq. (6.2). Particularly, for the conserved-mass transport processes considered here, one recovers free energy density $f(\rho)$ and chemical potential $\mu(\rho) = f'(\rho)$, as in Eqs. (3.15) and (3.14), by explicitly using the expressions of conductivity $\chi(\rho)$ and diffusion coefficients $D(\rho)$ derived in sections 7.2.1, 7.2.2 and 7.2.3.

7.4. results: asymmetric mass transfers

In the asymmetric mass transport processes, masses are transferred preferentially in a particular direction, say, counter-clockwise. Consequently, there is, on average, a nonzero mass current and detailed balance is manifestly broken in the system. However, even in the case of such asymmetric mass transfer, we explicitly show below that the bulk-diffusion coefficient $D(\rho)$ and the conductivity $\chi(\rho)$ can be defined and calculated within MFT and the resulting quantities still satisfy an ER. The conductivity (differential) $\chi(\rho) = [\partial J_d/\partial F]_{F=0}$ here can be defined with respect to a small perturbing biasing force field $\vec{F}$ around the nonzero current-carrying steady state. For simplicity, only the random sequential update rule is considered here; the results can be straightforwardly generalised to the parallel update rules.

To illustrate how one could incorporate asymmetry in transfer of masses, let us now consider a particular model, say, model I where the dynamics is described by Eq. (7.9) in section 7.2.1. In this case, model I becomes one having asymmetric transfer of masses, provided that the probability density function $\phi(r_i)$ is not symmetric around $r_i = 1/2$. Clearly, the asymmetric mass-transfer gives rise to an inherent bias towards a particular direction. Note that asymmetry can be incorporated in several other ways also, but, for simplicity, we confine our discussions to the cases considered below.

Now, in the above mentioned asymmetric version of model I, the time-evolution of the first moment $\langle m_i(t)\rangle = \rho_i(t)$ of mass at site i is governed by

$$\begin{aligned}\frac{d\rho_i}{dt} &= \tilde{\lambda}\langle r_{i-1}m_{i-1} + (1-r_{i+1})m_{i+1} - m_i\rangle \\ &+\frac{\tilde{\lambda}^2}{2}(2\theta_2-\theta_1)a\Big[\langle m_{i-1}^2\rangle - \langle m_{i+1}^2\rangle\Big]F \\ &+\frac{\tilde{\lambda}^2}{2}(2\theta_1-1)a\Big[\langle m_{i+1}^2\rangle - \langle m_i^2\rangle\Big]F\end{aligned}$$

Let us define strength of asymmetry $\alpha = [1-2\theta_1]$, which in a particular case may depend on system size L through the first moment θ_1 of probability density function $\phi(r_i)$. The parameter α helps us in obtaining concisely the hydrodynamic equation, which can be applicable to both weakly and strongly asymmetric cases, depending on α. We now rescale Eq. (7.42) by $i \to x = i/L$, $t \to \tau = t\alpha/L$ and $a \to 1/L$ and, using the expression $\langle m_i^2\rangle = \rho_i^2/[\lambda + 2\tilde{\lambda}(\theta_1-\theta_2)]$ [97], we obtain the hydrodynamic equation,

$$\frac{\partial\rho}{\partial\tau} = -\tilde{\lambda}\frac{\partial\rho}{\partial x} + \nu D\frac{\partial^2\rho}{\partial x^2} - \frac{\partial}{\partial x}[\nu\chi(\rho)F], \tag{7.42}$$

where $\nu = 1/\alpha L$ and

$$\chi(\rho) = \frac{\tilde{\lambda}^2}{2}\frac{1-4(\theta_1-\theta_2)}{\lambda+2\tilde{\lambda}(\theta_1-\theta_2)}\rho^2\ ;\ \ D(\rho) = \frac{\tilde{\lambda}}{2}.$$

There is now an additional drift current $\tilde{\lambda}\rho$ appearing in the hydrodynamic equation. However, one can immediately verify that, the diffusivity and mobility are indeed connected by the Einstein relation as in Eq. (7.1). Note that conductivity now depends on the strength of asymmetry α through $\theta_1 = (1-\alpha)/2$.

In the case of weak asymmetry where $\alpha(L) = \text{const.}/L$ is $\mathcal{O}(1/L)$, the above rescaling of time ($\tau \sim t/L^2$) leads to diffusive hydrodynamics with conductivity $\nu\chi(\rho)$ and diffusion coefficient $\nu D(\rho)$ both being finite. Whereas, in the case of strong asymmetry where $\alpha = \text{const.}$ is $\mathcal{O}(1)$, the above rescaling of time ($\tau \sim t/L$) gives hyperbolic hydrodynamics with conductivity $\nu\chi(\rho)$ and diffusion coefficient $\nu D(\rho)$ both being infinitesimally small as $\nu \to 0$ in the hydrodynamic limit. However, the MFT is still expected to describe the density fluctuation in both cases [46] and density field $\rho(x,\tau)$ would then satisfy the following stochastic hydrodynamic equation with a Gaussian multiplicative noise-current $\zeta(x,\tau)$,

$$\frac{\partial\rho}{\partial\tau} = -\partial_x\left[\tilde{\lambda}\rho - \nu D\frac{\partial\rho}{\partial x} + \zeta(x,\tau)\right], \tag{7.43}$$

where $\langle\zeta(x,\tau)\rangle = 0$ and $\langle\zeta(x,\tau)\zeta(x',\tau')\rangle = [\nu\chi(\rho)/L]\delta(x-x')\delta(\tau-\tau')$. Note that the structure of stochastic hydrodynamics for asymmetric cases remains quite similar to Eq. (7.37), where J_D is now replaced by $J_D + \tilde{\lambda}\rho$ and D and χ are now replaced by νD and $\nu\chi$, respectively. Consequently, the density large deviation function can be obtained by solving a slightly modified version of Eq. (7.38),

$$\int dx\left[\partial_x\left(\frac{\delta\mathcal{F}}{\delta\rho}\right)\nu\chi(\rho)\,\partial_x\left(\frac{\delta\mathcal{F}}{\delta\rho}\right) + \frac{\delta\mathcal{F}}{\delta\rho}\partial_x\nu D(\rho)\partial_x\rho\right] + \tilde{\lambda}\int dx\frac{\delta\mathcal{F}}{\delta\rho}\partial_x\rho = 0. \tag{7.44}$$

By noting that the last term in the l.h.s. of the above equation is identically zero when integration is performed over a periodic boundary, Eq. (7.41), along with Eq. (7.40), provides the density LDF, having the same functional form as in Eq. (6.3). One could check that the same LDF can also be recovered by directly using additivity. The only difference in the two cases of symmetric and asymmetric mass transfers is that the exact expressions of free energy $f(\rho)$ may differ as it is directly obtained from the ratio (related to parameter η) of conductivity $\chi(\rho)$ and diffusion coefficient $D(\rho)$ (or, from the mass fluctuation $\sigma^2(\rho)$) and the ratios may be different in these two cases. Indeed, the LDFs in the cases of symmetric and strongly asymmetric mass transfer are different as the conductivity $\chi(\rho)$ is different in these two cases. However, the LDFs are the same in symmetric and weakly asymmetric cases, which is somewhat expected.

In Figs. 7.1 and 7.2, we have plotted steady-state probability distribution $P_v(m)$ of mass m in a subsystem of volume $v = 10$ as a function of m for $\lambda = 0$, 0.25 and 0.5 and $L = 5000$, which are in excellent agreement with fluctuating hydrodynamics Eq. (7.43) as well as addi-

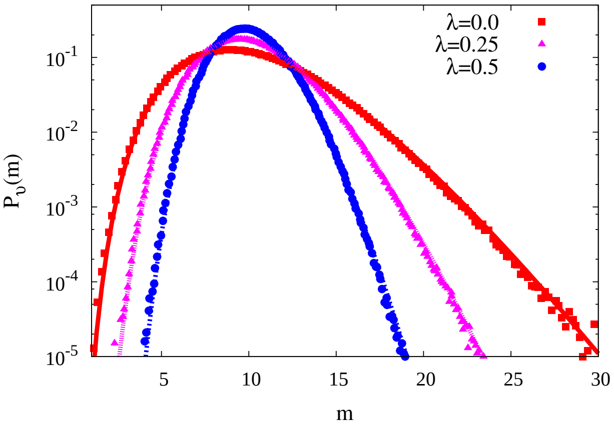

Figure 7.1.: Weakly asymmetric mass transfers; Model I (random sequential update): Steady-state probability distribution $P_v(m)$ is plotted as a function of subsystem mass m for $\lambda = 0$, 0.25 and 0.5 and subsystem volume $v = 10$.

tivity property in Eq. (6.1). It should be noted that, for a particular value of λ, the subsystem mass distributions are different for weak and strong asymmetry, depending on the parameters θ_1 (or α, the strength of asymmetry) and θ_2.

We have also considered asymmetric versions of models II and III, leading to similar conclusions as above (results not presented).

7.5. Concluding remarks: MFT and additivity

In this chapter, we have derived hydrodynamics of paradigmatic conserved-mass transport processes on a one dimensional ring-geometry, which have been intensively studied in the last couple of decades. In these processes, we have calculated two transport coefficients – diffusion coefficient $D(\rho)$ and conductivity $\chi(\rho)$. In all cases studied here, we find that the diffusion coefficient D is independent of mass density ρ and the conductivity $\chi(\rho) \propto \rho^2$ is proportional to the square of the mass density ρ. Interestingly, for all of these continuous conserved mass transport processes studied here, the analytically obtained functional dependence of the two transport coefficients $D(\rho)$ and $\chi(\rho)$ on density indicates that, on large space and time scales, these mass transport processes belong to the class of Kipnis-Marchioro-Presutti (KMP) processes demonstrated in section 6.2 of last chapter. However, unlike the KMP processes on a ring, the processes studied in this paper generally have a

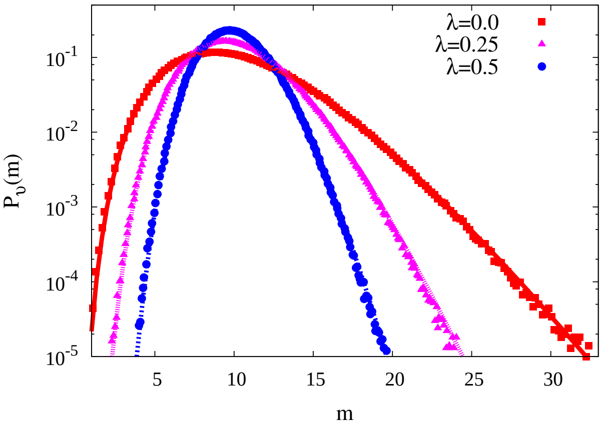

Figure 7.2.: Strongly asymmetric mass transfers; Model I (random sequential update): Steady-state probability distribution $P_v(m)$ is plotted as a function of subsystem mass m for $\lambda = 0$, 0.25 and 0.5 and subsystem volume $v = 10$.

nontrivial spatial structure in their steady states. That is, they have finite spatial correlations in the steady state. Not surprisingly, the exact probability weights of microscopic configurations in the steady-state, except for a few special cases [27, 28, 80, 83], are not yet known. In fact, precisely due to this nontrivial spatial steady-state structure in out-of-equilibrium interacting-particle systems, finding hydrodynamics in such systems poses a great challenge. This is because, in the absence of knowledge of the exact steady-state weights, it is usually difficult to calculate averages of local observables (e.g., moments of local mass variables, which have been actually used here to derive hydrodynamics of these processes).

However, as noted in Ref. [97], there is an important feature in these conserved-mass transport processes (with zero external bias $F = 0$), arising from the fact that the Bogoliubov-Born-Green-Kirkwood-Yvon (BBGKY) hierarchy involving $n-$point spatial correlations in the steady states closes. In other words, $n-$point spatial correlations in the steady state do not depend on $(n+1)-$point or any higher-order spatial correlations. This particular property previously enabled us to exactly calculate the steady-state $2-$point spatial correlations and, consequently, the second moment $\langle m_i^2 \rangle$ of local mass at site i [97]. Indeed, the second moment of local mass, which appears in the hydrodynamic equations [e.g., see Eq. (7.12)], determines the functional dependence of the conductivity $\chi(\rho)$ on density ρ.

Remarkably, the two transport coefficients satisfy an equilibriumlike Einstein relation Eq. (7.1) even when the microscopic dynamics violate detailed balance. Moreover, using these two transport coefficients, a fluctuating hydrodynamic framework for these processes have been set up here, following a recently developed macroscopic fluctuation theory (MFT). The MFT has helped us to calculate density large deviation function (LDF), which is analogous to an equilibriumlike free energy density function. The LDFs completely agree with that obtained previously in Refs. [50, 97] solely using an additivity property Eq. (6.1) [2]. Taken together, the additivity and MFT formalism provide a broad and consistent framework to understand macroscopic static and dynamic properties of far from equilibrium processes. Indeed, using these two together, a remarkable characterisation of a number of fixed energy sandpile (FES) models has been recently put forward with important consequences in the near critical behaviour [134].

Finally, it is worth mentioning that, unlike in equilibriumm, microscopic dynamics in the mass transport processes considered here in general do not satisfy detailed balance. Even for the processes with symmetric mass transfers, we have explicitly shown that Kolmogorov criterion, and thus detailed balance, is violated (even in the absence of a biasing force) and the microscopic dynamics is not time reversible. That is, for a forward path in the configuration space, there may not exist a reverse path. However, in spite of the lack of any microscopic reversibility in the dynamics of the processes, the observed Einstein relation suggests that these mass transport processes possess a kind of time-reversibility on a coarse-grained macroscopic (hydrodynamic) level. As discussed here, this macroscopic time-reversibility can be understood in the light of a macroscopic fluctuation theory (MFT) [47], which indeed correctly predicts the probabilities of density large-deviations obtained earlier in Refs. [50, 97]. From an overall perspective, we believe our study could provide some useful insights in characterizing fluctuations in many other driven many- particle systems, e.g., various driven lattice gases [50, 121], where a fluctuating hydrodynamic description is yet to be obtained.

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[2] This agreement is not a mere coincidence. Note that, as explained in chapters 2, 3, and 6, both additivity and MFT are valid under similar properties of macroscopic variables; both require scale separation, or weak correlation.

8. Summary

Based on very general considerations of properties of fluctuating macroscopic observables, we demonstrated that, for a broad class of stochastic processes, irrespective of the details of the underlying dynamics and independent of whether detailed balance satisfied or not, the steady state could be completely characterised using an equilibriumlike additivity property. In fact, a detailed thermodynamic characterisation of the macroscopic distributions for a variety of nontrivial model systems, viz. conserved mass transport processes, could be established with this property alone supplemented with the knowledge of static correlations – as required by the theory. Further, we obtained exact results for transport coefficients and found macroscopic current-density profiles in a dynamic set up for these models in the framework of macroscopic fluctuation theory. It is remarkable that, the results from dynamics are in excellent agreement with the predictions of additivity in the steady state limit, thereby indicating towards a more general underlying unity of the static and dynamic perspectives for nonequilibrium processes in general.

In chapter 3, it is explicitly shown that, irrespective of whether the time stationary state is equilibrium or not, an additivity property alone leads to the unified characterisation of macroscopic quantities in terms of an *intensive* thermodynamic variable. The result is further astonishing in the sense that, the full large deviation probability for arbitrary processes could be obtained with the knowledge of only of the second moment, a conclusion used to be considered as restricted to central limit regime only. Although the analysis is made and demonstrations are given for restricted but important category of conserved systems, a brief headway for understanding nonconserved systems is also provided. In this and the subsequent chapters, we argue that this property can explain why power laws appear generically in nonequilibrium steady states. We demonstrate that the existence of a fluctuation-response relation with a singular (power law in general) response function leads to power-law distributions with nontrivial exponents. The simplest form of the singularity, a simple pole, gives rise to the exponent 5/2, which was often observed in the past in apparently unrelated systems. We substantiate the claims by analytically calculating the response function, which diverges as critical point is approached, and the corresponding mass distributions in paradigmatic nonequilibrium mass aggregation models defined on a one dimensional ring. Most remarkably, the analysis equally applies to critical properties in both equilibrium and nonequilibrium and is independent of detailed dynamical rules.

Thermodynamic characterization of phase coexistence in driven systems is a fundamental problem in statistical physics. From that perspective, it is quite interesting that, these nonequilibrium systems having a variance diverging at finite density does admit an equilibrium-like chemical potential, which equalizes in the coexisting phases and vanishes at criticality - giving rise to a thermodynamic characterisation of phase coexistence in terms of Maxwell construction, and leads to pure power laws at criticality. We believe that the analysis, although done for a particular kind of aggregation models in a mean field approach (which we we believe to be an excellent approximation in these cases), is based on very general principle of additivity, and would be applicable in many other driven systems where phase coexistence is known to occur.

In the subsequent chapter, we have characterized spatial structure in a broad class of continuous conserved-mass transport processes defined on a one dimensional ring. Although defined by simple dynamical rules, except for few spacial cases having a product measure, the steady state structure is widely unknown. We have shown that, for generic parameter values all these systems violate the Kolmogorov condition for detailed balance and therefore the steady states are inherently far from equilibrium. However, for these models allow one to exactly calculated the static two-point spatial correlation functions, which are found to be short-ranged. Consequently, using additivity, the distribution of mass in macroscopic subsystems is calculated. Note that, in all these models, the variance of subsystem mass is in fact found to be quadratic in the mass density leading to the subsystem mass distributions in the form of gamma distribution. We substantiated our claims using simulation.

The mathematical simplicity of these nontrivial models gives rise to an expectation that, the full characterization of three and higher order spatial correlations, which is still lacking and remains to be an interesting open issue, could indeed be possible, that eventually could lead to complete understanding of microscopic steady-state structure in these systems. Nevertheless, apart from providing a much needed thermodynamic structure to the understanding of NESS, our results mark a significant progress in that, it have added to the rather sparse knowledge of exact results of two-point correlations and, in particular, the macroscopic mass distributions for nonequilibrium mass transport processes.

In dynamic set up, we have derived the fluctuating hydrodynamics of these paradigmatic continuous conserved-mass transport processes. In these processes, we have analytically calculated two transport coefficients – diffusion coefficient $D(\rho)$ and conductivity $\chi(\rho)$. In all cases studied here, we find that the diffusion coefficient D is independent of mass density ρ and the conductivity $\chi(\rho) \propto \rho^2$, resembling Kipnis-Marchioro-Presutti (KMP) processes on a ring. However, unlike the KMP on a ring, the models have finite spatial correlations in the steady state, and the exact probability weights of microscopic configurations are unknown. In fact, precisely due to this lack of knowledge of the exact steady-state weights, the calculation of the averages of local observables (e.g., moments and correlations of local mass variables, which appears during the derivation of hydrodynamics) and thereby the hydrodynamics in general pose enormous difficulties.

However, as we have already mentioned, the steady-state 2−point functions for these particular models are exactly solved, which this time allow us to determine the functional dependence of the conductivity $\chi(\rho)$ on density ρ. Remarkably, the two transport coefficients are seen to satisfy an equilibriumlike Einstein relation Eq. (7.1) even though the microscopic dynamics violate detailed balance. Moreover, using these two transport coefficients, a fluctuating hydrodynamics have been set up for these processes following the recently developed macroscopic fluctuation theory (MFT). The MFT has helped us to calculate macroscopic current-density large deviation function (LDF) and the steady state distribution of the density profile. It turns out that, the LDFs completely agree with that obtained previously using additivity.

Finally, note again that, unlike in equilibrium, microscopic dynamics in the mass transport processes considered here do not satisfy detailed balance. In fact, the asymmetric models which are also considered here have finite mass current in the steady state. Despite the lack of microscopic reversibility in the microscopic dynamics, the observed Einstein relation suggests that these processes possess a kind of emergent time-reversibility on a coarse-grained macroscopic (hydrodynamic) level, which can be characterised in the light of a macroscopic fluctuation theory (MFT). Indeed, these closely connected formalisms of MFT and additivity, taken together, provide a framework characterising broad class of far from equilibrium processes.

In a overall perspective, though we could touch upon a broad still only a fraction of the immense variety of nonequilibrium phenomena, we believe, the whole discussion in the thesis provides a headway that could be useful in understanding in a unified framework the wide spectrum of macroscopic static as well as dynamic properties of systems far from equilibrium.

Part III.

Appendices

A. Condensation in models without conservation

We discuss a nonconserved version of the mass aggregation models where systems exchange mass, though weakly, with environment. In this case, in addition to the two processes (1) diffusion and (2) fragmentation of masses, a particle now can be adsorbed with rate q and desorbed at a site with rate p, provided the site is occupied, where $p, q \to 0$ with the ratio $r = q/p$ finite. Due to adsorption and desorption processes, total mass in the system is not conserved. This model is related to several models studied in the past for finite p and q [29, 108, 115, 135]. Interestingly, in the limit of $p, q \to 0$, mass fluctuation in the nonconserved model can be obtained from the occupation probability in its conserved version (i.e., $p = q = 0$). Let us first define, in the space of total mass M, a transition rate $T_{M+1,M}$ from mass M to $M+1$. In the steady state, the probability $P(M)$ that the system has mass M satisfies a balance condition $P(M)T_{M+1,M} = P(M+1)T_{M,M+1}$ where the mass distribution $P(M)$ can be obtained as

$$P(M+1) = \left[\prod_{M=0}^{M} \frac{T(M \to M+1)}{T(M+1 \to M)} \right] P(0). \tag{A.1}$$

As the ratio of transition rates can be written as $T_{M+1,M}/T_{M,M+1} = \frac{q}{p\mathcal{S}(\rho)}$ where $\mathcal{S}(\rho)$ is the occupation probability and $\rho = M/V$, the distribution function can be written, upto a normalization factor, as

$$P(M) \propto e^{\sum_M [\ln(q/p\mathcal{S})]} \simeq e^{-V \int_0^\rho d\rho [\mu(\rho) - \mu_0]} \tag{A.2}$$

where $\mu_0 = \ln(q/p)$ an effective chemical potential and $f(\rho) = \int d\rho \mu(\rho) = \int d\rho \ln \mathcal{S}(\rho)$ an effective free energy (canonical) density function. The stationary macroscopic density ρ_0 as a function of adsorption to desorption ratio $r = q/p$ can be obtained by minimizing the grand potential or the large deviation function for the density fluctuation $h(\rho) = f(\rho) - \mu_0 \rho$, leading to the relation $\mathcal{S}(\rho_0) = r$.

Till now, the analysis is exact. However, it may not always be possible to exactly calculate the occupation probability $\mathcal{S}(\rho)$. For demonstration, let us proceed by considering a model with diffusion and fragmentation rate as in Case I. We obtain an approximate expression, obtained within mean- field theory, of $\mathcal{S}(\rho) = \rho(1-\rho)/(1+\rho)$. [1] Eq. A.2 implies the subsystem mass distribution having a form $P_v(m) \propto w_v(m)\exp(\mu m)$ and consequently a FR relation as in Eq. 4.2 follows. Then one can calculate the scaled variance as $\sigma^2(\rho) = (d\mu_0/d\rho)^{-1} = \rho_0(1-\rho_0)(1+\rho_0)/(1-2\rho_0-\rho_0^2)$ for $\rho_0 < \rho_c$, where critical macroscopic density $\rho_c = \sqrt{2}-1$, or equivalently for a $r < r_c$. Note that, unlike the conserved case, here the mass density is a fluctuating quantity, and the macrosctate and the fluctuations are governed by the 'external' parameter, viz. the chemical potential $\mu_0(r)$. The functional form of variance here is different from that in the conserved-mass case, implying that the canonical and grand canonical ensembles are not equivalent [13, 14]. However, the nature of singularity in the variance remains the same near criticality where $\sigma^2(\rho) \sim (\rho_c-\rho)^{-n}$ with $n = 1$. Therefore the additivity property leads to the same power law scaling in the single-site mass distribution $P_1(m) \sim m^{-\tau}\exp(\tilde{\mu}m)$, for large m, where $\tau = 5/2$ and $\tilde{\mu} = \mu_0 - \ln\mathcal{S}(\rho_c) = \ln(r/r_c)$ with $r_c = \mathcal{S}(\rho_c)$.

The calculational steps are as follows. The probability distribution function $P(M)$ of total mass M can be written, up to a normalization factor, is given by equation A.2,

$$P(M) = \text{const.} \times e^{-V\int_0^\rho d\rho[\mu(\rho)-\mu_0]}$$

Now, if we assume that the joint mass distribution $\mathcal{P}[\{m_i\}]$ has a product form on single-site level ($v = 1$), i.e., product of single-site mass distribution function $p(m_i)$,

$$\mathcal{P}[\{m_i\}] = \prod_{i=1}^{V} p(m_i), \tag{A.3}$$

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[1] In a single site level, mean field calculation from the update rule gives, $\mathcal{S}(\rho_0) = \rho(1-\frac{\rho_0}{1+p})/(1+\rho_0)$, ρ_0 is the average single site density that should equal with most probable global density. The expression we used in the text is valid for very small p.

the probability distribution function $P(M)$ of mass M in the system can be written as

$$P(M) = \prod_{i=1}^{V} \left[\int dm_i p(m_i) \right] \delta \left(M - \sum_i m_i \right). \tag{A.4}$$

From the Laplace transform $\tilde{P}(s) = \int dMP(M)\exp(-sM) = [\tilde{p}(s)]^V$ of the mass distribution $P(M)$, the Laplace transform $\tilde{p}(s) = \int dm_i p(m_i)\exp(-sm_i)$ of single-site mass distribution $p(m)$ can be written as

$$\tilde{p}(s) = \text{const.} \times e^{-\lambda_1(s)}, \tag{A.5}$$

where

$$\lambda_1(s) = \mathbf{inf}_\rho [h(\rho) + s\rho]. \tag{A.6}$$

Here we have used inverse transform

$$\tilde{P}(s) = \text{const.} \times \int d\rho e^{-V[h(\rho)+s\rho]}, \tag{A.7}$$

which has been obtained from Eq. A.2 and where grand potential or the large deviation function for density fluctuation $h(\rho) = f(\rho) - \mu_0\rho = \int_0^\rho [\mu(\rho) - \mu_0] d\rho$ and chemical potential $\mu(\rho) = \ln \mathcal{S}(\rho) = \ln[\rho(1-\rho)/(1+\rho)]$, as written earlier. Note that the function $\mathcal{S}(\rho)$ is the occupation probability in the conserved mass aggregation model and has been obtained by putting $w_1 = 1$ and $w_2 = 0$ in Eq. 4.20.

Now the function $\lambda_1(s)$, Legendre transform of grand potential $h(\rho)$, can be written as

$$\lambda_1(s) = h(\rho^*) + s\rho^*, \tag{A.8}$$

where ρ^* is the root of the equation $d[h(\rho) + s\rho]/d\rho = 0$ or $\mu(\rho^*) - \mu_0 + s = 0$, i.e., ρ^* is the root of

$$\ln \left[\frac{\rho^*(1-\rho^*)}{1+\rho^*} \right] = \mu_0 - s. \tag{A.9}$$

The critical density is obtained by putting scaled variance as $\sigma^2(\rho) = (d\mu_0/d\rho)^{-1} = \infty$ or $1/\sigma^2(\rho) = 0$,

$$\frac{(1 - 2\rho_c - \rho_c^2)}{\rho_c(1-\rho_c)(1+\rho_c)} = 0, \tag{A.10}$$

and thus $\rho_c = \sqrt{2} - 1$. In the macrostate (most probable state), we have $\mathcal{S}(\rho_0) = r$, implying that the critical density is related to the ratio $r = q/p$ through $\mathcal{S}(\rho_c) = r_c$, i.e., when the large deviation function vanishes $h(\rho_c) = 0$. To obtain the large-mass behaviour, we expand $\mu(\rho)$ around $\rho = \rho_c$,

$$\mu(\rho) = \mu(\rho_c) + \frac{\mu''(\rho_c)}{2}(\rho - \rho_c), \tag{A.11}$$

to obtain

$$(s - s_c) \simeq \frac{|\mu''(\rho_c)|}{2}(\rho^* - \rho_c)^2, \tag{A.12}$$

$$\lambda_1(s) \simeq a_0 + a_1(s - s_c) + a_2(s - s_c)^{3/2}, \tag{A.13}$$

in leading order in $(\rho * - \rho_c)$ where $s_c = \mu_0 - \mu(\rho_c)$, leading to the desired result in the main text,

$$p(m) \sim \frac{1}{m^{5/2}} e^{s_c m} = \frac{1}{m^{5/2}} e^{[\mu_0 - \mu(\rho_c)]m}. \tag{A.14}$$

The above results are indeed consistent with what was found for general p and q, on the mean-field level, in the *'in-out'* model of [115] which is a special case of the nonconserved model with $w = 0$. One can interpret this phenomenon in the light of equilibrium Bose-Einstein conensation (BEC). The critical density here actually signifies that, for $r > r_c = \mathcal{S}(\rho_c) = 3 - 2\sqrt{2}$, there is a condensate (BEC- like) which, in the grand-canonical setting (due to the absence of mass conservation), implies a phase with a diverging mass density, similar to the 'Takayasu phase' where mass density eventually diverges. For p and q finite, form of the subsystem mass distribution as in Eq. A.2 remains the same, but the expression of $\mathcal{S}(\rho)$, due to the presence of spatial correlations, is different. However, the physical picture remains the same as in the Bose-Einstein condensation.

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