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Numerical Studies of Thermal Hydrodynamics

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A Thesis submitted to the
Faculty of Graduate Studies and Research
in partial fulfillment of the requirements for the degree of
Doctor of Philosophy

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Abstract

We introduce and study new lattice gas models of hydrodynamics to simulate thermal fluids in non-equilibrium. The new approach consists in modifying the conventional Lattice Gas Cellular Automata method for fluids by removing the constraints of the Fermi exclusion principle and by introducing internal potential energy levels for the particles. These modifications allow the effective employment of Monte-Carlo dynamics for the evolution of the models, so that the temperature is defined in a natural way, and the introduction of interactions becomes straightforward.

Because the transport properties of fluids determine their behavior in non-equilibrium, we study in detail the effect of our modifications on transport coefficients. We derive expressions for these coefficients in two ways: from Chapman-Enskog expansions and from linear response theory. Because of the potential energy, the transport properties are more similar to those of real fluids than are conventional LGCA models with kinetic energy only: the bulk viscosity is non-zero and thermal diffusion is well defined over a range of densities and temperatures.

We construct several models to illustrate the advantages and implications of our approach. One model is used to study the local temperature distribution in a simulation of Rayleigh Bénard convection. Another is used to introduce interactions between particles and to simulate the 1st order phase separation of a fluid into the regions of low and high density. This latter model is also used to study the dynamics of interfaces between the phases. We observe the effects of inertia at the interface and demonstrate that the mean square width of an initially flat interface scales as $t^{3/2}$ for late times t .

Résumé

On introduit et étudie des nouveaux modèles de gazes sur grille pour l'hydrodynamique, pour étudier des fluides thermiques hors d'équilibre. La nouvelle approche consiste à modifier la méthode conventionnelle de Automates Cellulaires De Gaz De Treillis pour fluide en enlevant la contrainte reliée au principe d'exclusion de Pauli et en introduisant des niveaux d'énergie interne pour les particules. Ces modifications permettent l'utilisation efficace de la dynamique de Monte-Carlo pour l'évolution du modèle, ce qui permet une définition naturelle pour la température, et l'introduction d'interaction devient directe.

Étant donné que les propriétés de transport d'un fluide déterminent son comportement hors d'équilibre, on étudie l'effet de nos changements sur les coefficients de transport. On trouve des expressions pour ces coefficients de deux façons: à partir de l'expansion de Chapman-Enskog et de la théorie de réponse linéaire. À cause de l'énergie potentielle, les propriétés de transport sont plus similaires à celles des vrais fluides que celles des modèles conventionnels LGCA avec l'énergie cinétique seulement; la viscosité en bloc est différente de zéro et la diffusion thermique est bien définie sur un intervalle des densités et des températures.

On construit plusieurs modèles qui illustrent les avantages et les implications de notre approche. Un modèle est utilisé pour étudier la distribution de température locale dans une simulation de convection de Rayleigh Bernard. Un autre modèle est utilisé pour introduire des interactions entre les particules et pour simuler la séparation de phase de premier ordre d'un fluide entre des régions de basses et hautes densités. Ce dernier modèle est aussi utilisé pour étudier la dynamique des interfaces entre les phases. On observe les effets de l'inertie aux interfaces et on démontre que la largeur moyenne d'une interface qui est initialement plate change par un facteur d'échelle $t^{3/2}$ pour des temps t plus tard.

Statement of Originality

In this thesis, I introduce a new approach for modeling thermal hydrodynamic systems with applications to non-equilibrium statistical mechanics. The new features of the Lattice Gas method for modeling fluids are the removal of the requirements for Fermi statistics, the introduction of internal potential energy levels and the use of Monte-Carlo algorithms for time evolution. These features allow the construction of numerically effective models with well defined thermal properties and the incorporation of interactions between particles in a manner consistent with thermodynamics. I use the new models, the generalized thermal lattice gases (GTLG), to study non-equilibrium phenomena in fluids.

Specifically, the following aspects are original:

- In Section 3.5: the analytical expressions for the bulk viscosity ζ (3.34) and for the thermal conduction (3.42) for GTLG in single relaxation time approximation.
- In Section 3.6: the Green-Kubo type expressions for the transport coefficients of GTLG models.
- In Section 4.3: the combination of Lattice-Boltzmann dynamics with the many-demons Monte-Carlo algorithm.
- In Section 4.3: the results of simulations of Rayleigh Bénard convection.
- In Chapter 5: the new model with interactions, its analysis and the description of thermodynamic equilibrium.
- In Section 6.3: observation of a new scaling regime (the width of an initially flat interface growing with time as $t^{3/2}$ at late times) in numerical simulations of interface roughening using the model of Chapter 5.
- In Section 6.4.2: theoretical explanation of the $t^{3/2}$ growth law for the interface.

My contributions to this study include:

- In Chapter 3: the development of the single-relaxation-time approximation description of GTLG and the derivation of Green-Kubo type expressions for transport coefficients.

- In Chapter 4: the numerical analysis of the 13-bit model; numerical simulation of Rayleigh Bénard convection: identification of relevant values of parameters.
- In Chapter 5: the development of all the necessary code to carry out the simulations of the model with interactions; all numerical studies and analysis of equilibrium states of the model.
- In Chapter 6: simulations of interface roughening.

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Numerical Studies of Thermal Hydrodynamics

Introduction

1.1 Numerical Methods in Fluid Dynamics

Numerical methods for fluid dynamics now occupy an important place in the study of complex systems. A major stimulus to the field has been the large number of applications in which fluid flows play a crucial role, and this has spurred the interest of numerous physicists, computational engineers and mathematicians.

Numerical methods have proved to be an important tool to investigate the physics of fluids. In many cases numerical experiments are the only ones that allow the study of physical systems in particular regimes. Unlike many other areas of applicability of numerical methods, fluids, or hydrodynamics systems, are characterized by high complexity. The basic equation that describes fluids is the Navier-Stokes equation, (discussed later in Section 2.1). It is a non-linear, second-order differential equation, that is difficult to solve analytically even for simple boundary conditions. For most of the interesting applications (mixture of fluids, solidification, phase-separation, driven systems, complex geometry of flow, etc) this equation must be coupled to other complex equations so that direct numerical integration becomes ineffective. Often, the path of evolution of fluid systems is not known or is the subject of investigation, so that the construction of appropriate equations becomes the main difficulty.

In physics, numerical methods can be placed in one of two major groups:

1. computing the quantities of interest directly from the equations that describe them, assuming we can write the correct (or approximately correct) equations.
2. the simulation of phenomena using a model. The computer here is a kind of experimental laboratory, designed to explore the phenomena of interest.

In this thesis we develop and use an approach, *the Lattice Gas (LG) model of fluids*, that is part of second group. We construct a simple model that allows us to simulate the behavior of real systems. The model implies the set of parameters or variables that describe the state of system at any given time, as well as the rules to describe the evolution of this set. For fluid simulations the model should consist of a macroscopic number of particles (molecules, states, etc.) that necessarily possess (the attributes of) coordinates, mass and momentum. Also, depending on the complexity of the phenomena to be modeled, it can include other attributes: energy, orientation, internal structure, etc. The rules of evolution necessarily imply the conservation of mass and momentum as a prerequisite for recovering hydrodynamics in the behavior of system.

Lattice gas models are one of the two major approaches for simulating fluids: the other is the method of Molecular Dynamics (MD). The main difference between the two is that the MD methods imply continuous values for coordinates and time, while lattice gases deal with discrete space and time. In MD methods the evolution is determined by the forces that act on each particle from all other particles through some potential of interaction (often a Lennard-Jones potential (Verlet [67])). In the LG approach the evolution is defined by a set of rules for collisions and propagation. As a consequence, LG models are more computationally effective. While conventional Molecular Dynamic simulations (Frenkel and Smit [96]; Nielsen [99]), involve experiments that have, at best, $500 - 10^3$ particles, the lattice gas models discussed in this thesis can deal with about 10^6 particles for the same time-span of an experiment.

1.2 *Non-Equilibrium Phenomena in Fluids*

The computational efficiency of the lattice gas method for fluids makes it an important tool for studying systems where hydrodynamics is of importance (Manneville et al. [89]; Chopard and Droz [98]). Since the method was first described by Frisch, Hasslacher and Pomeau [86] in 1986, it has been successfully used in many areas of fluid physics - flows through porous media, phase separation and interface dynamics. Besides their relevance to hydrodynamics, lattice gas automata are also suitable for

the analysis of fundamental issues in statistical mechanics¹. **In this thesis we focus on the lattice gas method as applied to non-equilibrium phenomena in fluids, where temperature plays an important role in the dynamic evolution of the system.** The topic is especially interesting and important because of considerable progress made recently in the area of non-equilibrium statistical mechanics for non-hydrodynamic systems, progress which is not as evident for hydrodynamic systems. Similarly, it is of important experimental and theoretical interest and has wide interdisciplinary applications (see for example Family and Vicsek [91]). Examples can be divided into two categories: 1) single phase fluids in non-equilibrium, for example, the thermal instability of a layer of fluid heated from below (Rayleigh-Bénard convection, Getling [98]), or the dynamics of thin liquid films in the presence of surface-tension gradients (Maragoni convection, Ludviksson and Lightfoot [71]), and 2) phase separating systems in non-equilibrium (one or both phases being fluids), for example, the roughening of interfaces or spinodal decomposition².

The second category of multi-phase fluids is especially interesting from both theoretical and applied points of view. In their evolution (towards equilibrium), phase-separated systems are characterized by some time dependent characteristic length, the average width of the interface, for example, or the average domain size in spinodal decomposition. If this length grows as a power law in time over certain time-periods, then the evolution is scale-invariant (Bray [94]; Krug [97]). Certain characteristics, such as the growth exponent from the above power law, for example, then determine a universality class that reflects basic similarities in the mechanism of evolution of systems in the class. The mechanisms of evolution in non-equilibrium processes are least well understood in fluids, although some scenarios have been suggested (for example Siggia [79]; Grant and Elder [99] for spinodal decomposition phenomena). Most of the understanding, however, comes from the results of computer simulations, which allow direct study of the non-equilibrium phenomena of interest and of the scale-invariance

¹One of the successful examples is so-called 'long-time-tail' problem (Colvin, Ladd and Alder [88]; Frenkel and Ernst [89]). Here, the cellular automata models allowed the effective measurements of velocity autocorrelation function that confirmed existing mode-coupling theory.

²The process of decay towards equilibrium in a multi-phase region of a phase diagram constrained by particle conservation, as in quenches of binary mixtures.

of the selected mechanisms of evolution. Reviewing the existing attempts in this area we notice that:

1. Simulations done with lattice gas models of fluids are superior to other hydrodynamic models, because of their computational efficiency and, hence, ability to investigate non-equilibrium behavior over a much broader range of time and length scales than is possible with molecular dynamics.
2. In most cases the LG models used are more representative of dynamical systems than of thermal systems, *i.e.* the models used do not possess a thermodynamically correct temperature.
3. In the remaining situations, *i.e.* when LG models do possess a thermodynamically correct temperature, the models are not capable of modeling *situations where variations of the local temperature are important.*

This last observation (item [3.]) implies that the existing (thermodynamically correct) LG models of fluids have not yet been adapted to simulate a spatial temperature distribution dependent on boundary conditions in such non-equilibrium phenomena as Rayleigh-Bénard convection (fluid in a gravitational field heated at the lower boundary), or the evolution of a system with a 1st order phase transition where latent heat is produced at an interface. Thus, in this thesis, we address the challenge of non-equilibrium phenomena in fluids in three parts: 1) the development of a numerical method that is free from the disadvantages described above, 2) numerical simulations of some non-equilibrium scenarios and 3) a new theoretical approach for studying non-equilibrium systems in fluids with latent heat.

In all these situations we limit our investigations to lattice gas fluid models in two dimensions. In principle, the extension to three dimensions is straightforward, however, the computational simplicity of models in two dimensions makes them appropriate for the preliminary studies described in the thesis.

1.3 Outline of the Thesis

The thesis is summarized in Figure 1.1; it is organized as follows. First, in Chapter 2, we introduce lattice gas models of fluids. Basic concepts, definitions, notations and

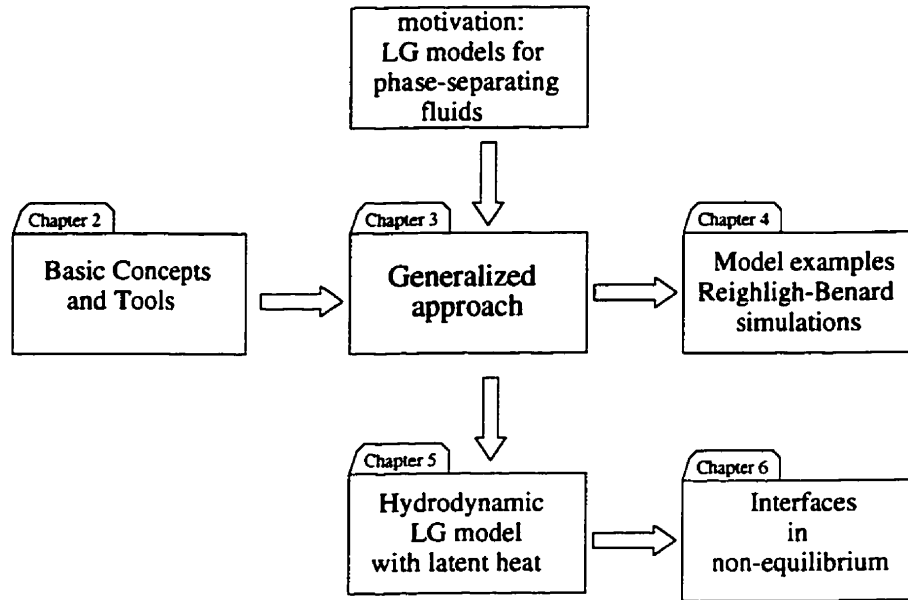


Figure 1.1: Flow-chart for thesis organization.

tools are discussed here.

In Chapter 3, we introduce and discuss the new class of Cellular Automata Lattice Gas Models (LGCA) for Thermal Hydrodynamics. Two novel features in constructing new models for hydrodynamics are the presence of internal structure or energetic states for each particle that constitute a gas, and the use of rules of evolution that do not necessarily imply conventional LGCA Fermi-Dirac statistics. In the macroscopic limit these models obey the Navier-Stokes equation of hydrodynamics. Statistical-mechanical equilibrium is described in terms of conserved quantities and the generalized expressions for transport coefficients are derived. Linear response theory for lattice gases is revised to accommodate the new features of our models.

In Chapter 4, we illustrate the feasibility of our approach with two examples. In the first example we study the transport properties of a model with 2 energy levels and with the use of unconventional statistics. In the second example, Maxwell-Boltzmann statistics is employed to construct a simple model with 3 energy levels and with two possible speeds for the particles. We show that the standard Monte-Carlo methods of statistical mechanics are easily adapted to our class of models. We report the results of a Rayleigh-Bénard convection experiment simulated using the our model adapted to the Lattice-Boltzmann method.

In Chapter 5, we expand the formalism of the LGCA approach to models that include short-range interactions between particles. We construct a model with 2 energy levels, one speed for the particles, Maxwell-Boltzmann statistics, and with a microscopic Hamiltonian that resembles the Ising Hamiltonian. Mean-field arguments are presented to confirm the presence of a first-order phase transition. The algorithm is discussed and the results for simulations of single-phase and two-phase states in equilibrium are presented. The phase diagram is obtained. The density correlation function (structure function) for two different phases that can coexist at certain temperature is shown to have the expected structure.

In Chapter 6, we use the model with interactions to study the interface between two fluids in phase equilibrium. First, the theoretical arguments about physical mechanisms that drive the interface between two phases are discussed and the general scaling behavior with time of the width of interface, initially flat, is derived. The effect of hydrodynamics on the interface evolution and the appearance of a new $t^{3/2}$ regime are presented, followed by a detailed study of the new inertial regime for interface evolution.

Lattice Gas models of Hydrodynamics

2.1 Differential Equation of Hydrodynamics

There are two ways to look at fluids. On the basic, *microscopic* level, fluids are interacting molecules obeying the basic laws of physics *i.e.* conservation of mass, momentum and energy. On the *macroscopic scale* a fluid may be described as a smoothly varying continuum. In this latter case, the relevant variable is the *velocity* $\mathbf{u}$ (a vector) of flow at different points, and the conservation equations take the form of partial-differential equations. For example, the (microscopic) mass conservation law takes the form of the *equation of continuity*

$$\frac{\partial \rho}{\partial t} + \nabla \cdot \rho \mathbf{u} = 0 \quad (2.1)$$

where ρ is the local density. The (microscopic) momentum conservation can take different (macroscopic) forms, depending on the properties of the fluid. The simplest form is known as the *Euler equation*

$$\frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u} \cdot \nabla) \mathbf{u} = -\frac{1}{\rho} \nabla p \quad (2.2)$$

where p is the pressure. The Euler equation coupled to the continuity equation (2.1) describes *inviscid compressible flow*. In the case of incompressible fluids, the density may be taken to be constant. In this case the continuity equation reduces to

$$\nabla \cdot \mathbf{u} = 0 \quad (2.3)$$

Equations (2.2) and (2.3) then are a set of equations for *inviscid incompressible flow*¹.

¹Unlike for compressible fluids, there is no need to employ an equation of state for incompressible flow. The pressure here is completely determined from equations (2.2) and (2.3), *i.e.* it adjusts so that the flow follows the incompressibility condition (2.3).

However, fluids are in reality *viscous*, *i.e.* they react to shear stress (or a velocity gradient) and to compression by establishing a momentum flux. The strength of this reaction is determined by the *kinematic shear viscosity* ν and the *bulk viscosity* ζ . The equation of evolution then becomes

$$\rho \left[\frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u} \cdot \nabla) \mathbf{u} \right] = -\nabla p + \rho \nu \nabla^2 \mathbf{u} + \left(\zeta + \frac{1}{d} \rho \nu \right) \nabla (\nabla \cdot \mathbf{u}) \quad (2.4)$$

Here $d = 2$ is a dimension. Equation (2.4) (also known as *Navier-Poisson equation*) describes momentum conservation in a compressible viscous fluid. It is considerably simplified for incompressible fluids. In this case ρ is constant and condition (2.3) eliminates the last term in (2.4) resulting in the *Navier-Stokes* (NS) equation of incompressible viscous hydrodynamics:

$$\frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u} \cdot \nabla) \mathbf{u} = -\frac{1}{\rho} \nabla p + \nu \nabla^2 \mathbf{u} \quad (2.5)$$

The above equations govern the dynamics of fluids in our everyday experience, and their solution is the main goal in applied problems of fluid dynamics (as well for our work in this thesis). Of special interest is the Navier-Stokes equation (2.5), since most of the real fluids are incompressible or weakly compressible in practical situations¹.

For the derivation and deeper understanding of equations of hydrodynamics we refer to any book on hydrodynamics (for example, Hansen and McDonald [89]; Landau [95]; Resibois and de Leener [77], etc). Here we note that the LG methods discussed in this thesis allow us to effectively simulate Navier-Stokes fluids in situations where the thermodynamic state of the fluid in equilibrium is controlled by the temperature. The nonequilibrium scenarios, our prime interest of study (Section 1.2), are then controlled by transport coefficients (for instance, viscosities, that enter the NS equation, as well as thermal diffusion and sound attenuation coefficients that will be discussed in Section 2.4) defined for the fluid in equilibrium, in so far as our non-equilibrium situation is not really far from equilibrium².

¹The interparticle potential, Lennard-Jones for example, has a local minimum at some interparticle distance and a very strong (almost infinite) potential barrier that prevents the particles from getting closer under a compression force.

²The relation between transport properties in equilibrium and non-equilibrium constitute the well known *fluctuation dissipation theorem*.

In what follows in this Chapter, we introduce the essential elements of simple Lattice Gas (LG) models of hydrodynamics capable of simulating NS fluids, and discuss the essential tools and methods for describing the equilibrium state of the LG and its transport properties.

For a more detailed review of the theory, we refer to the corresponding sections in the literature: Manneville et al. [89], Rothman and Zaleski [94], Baran [95], Rothman and Zaleski [97], Chopard and Droz [98].

2.2 The Example of the FHP Model

The classic and illustrative example of Lattice Gas fluids is the FHP model (Frisch, Hasslacher and Pomeau [86]). It is defined on a two-dimensional hexagonal lattice, lattice spacing $a = 1$. Each site of the lattice can be occupied by a number of some fictitious particles, Figure 2.1. Each particle is characterized by its discrete location

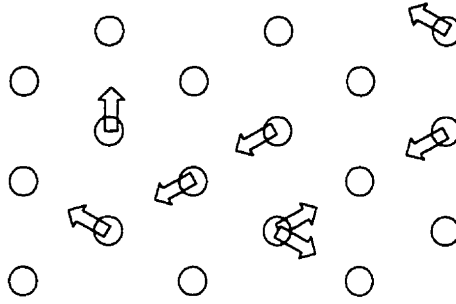


Figure 2.1: Two dimensional hexagonal lattice (sites shown as circles), populated by fictitious particles (shown as arrows) in FHP model

$\mathbf{r}$ (or site number) and discrete velocity $\mathbf{c}_i$ such that the absolute value of the velocity c is always constant, and the direction i of velocity can only point to the nearest neighbor sites. Because there are $z = 6$ nearest neighbors in a hexagonal lattice, we have six allowed velocities: $|\mathbf{c}_i| = c, i = 1..6$. Further, the velocity value c is equal to $a/\Delta t$, so that after some unit of time $\Delta t = 1$, called *one timestep*, the particle finds itself at the neighboring site. Thus, the system with N particles on a lattice with V sites evolves with time. The evolution consists of a *collision* step and a *propagation* step. The collisions are local and occur according to particular rules. These rules are conveniently represented by a *collision table*: a list of diagrams depicting all possible *precollisional states* at a site with the corresponding *postcollisional* state(s) as shown

on Figure 2.2 for the simplest FHP model. Both two- and three-body collisions are

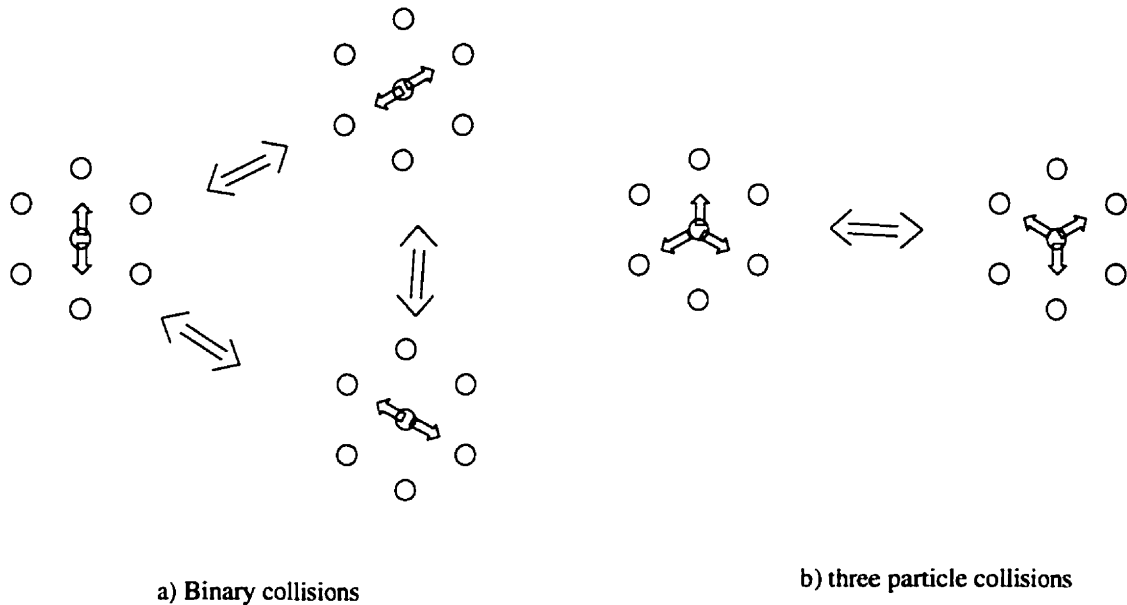


Figure 2.2: Typical set of collision rules defined on the hexagonal lattice. Arrows indicate the occupancy of the site by a particle with velocity modulo 1 and direction indicated by this arrow. Pre-collision and post-collision configurations are connected by the $\rightleftharpoons$ sign. Momentum is always conserved. Collision sets a) and b) constitute the microscopic description of the simplest FHP model.

necessary to avoid extra spurious conservation laws. (We refer to Section 2.4.3 for more discussion on unwanted *spurious* invariants in LG models and other artifacts of discreteness.)

Collisions occur simultaneously at all sites of the lattice. A computer algorithm for a lattice gas requires sequential visiting all of the sites and changing the pre-collision configurations to post-collision configurations according to Figure 2.2.

Collisions are then followed by the propagation of particles. Each particle moves to a neighboring site according to the direction of its velocity. When all sites and particles have been updated according to these collision and propagation rules, the system has evolved over *one time-step*.

In an FHP model the occupation of velocity states has a Boolean nature. The state is either occupied or not. This restriction came about because the invention of the FHP model was much inspired by progress in studying Cellular Automata systems (Toffoli and Margolus [87]). These are dynamic models in which the fully discrete universe is made up of cells which consist of finite number of bits with possible values 1

or 0. Thus, an FHP model is characterized by the *Fermi-Dirac exclusion principle* for the distribution of particles among states. This principle implies that there cannot be two particles in the system that have the same location and the same velocity. This imposes restrictions on the initial distribution, as well as on the collision step in evolution: a collision occurs if and only if the output configuration is allowed by the exclusion principle.

Thus, all of the above defines on a microscopic level the FHP model of a simple fluid. As we see, it is far simpler than the microscopic description of any natural fluid, but, remarkably, at a macroscopic scale it flows just like such a fluid, following the route determined by the Navier-Stokes equation (2.5).

We will refer to the FHP model, and its variants that differ from simplest FHP model by the set of collision rules, in many places in this thesis because many of its elements are present in the other LG models that we will consider. The detailed analysis, both analytical and numerical, is available in the classic papers of Frisch, Hasslacher and Pomeau [86] and Frisch et al. [87]. In what follows, we present the basic elements and results of such analysis, not for the FHP model, but for a larger class of models to which the FHP model belongs.

2.3 Thermal LGCA

The lattice-gas models of hydrodynamics have evolved since their first form as the FHP model (Section 2.2). Now they are found in several different categories depending on the type of specific properties that must be present or enhanced in the fluid system of interest.

A simple and useful modification of the FHP model may be obtained by adding *rest particles* with zero velocity to the model. The resulting 7-velocity model (also known as the FHP-II or FHP-III model, the name depending on the number of mass and momentum conserving collisions considered in their evolution) then has a higher collision rate. It yields a faster relaxation to statistical equilibrium and a lower viscosity.

The LG models capable of simulating the thermal properties of a fluid are of special interest to us (see Section 1.2). They are called *thermal* models. The FHP model

introduced in previous Section is *athermal*, without the notion of temperature. The ability of a model to simulate a fluid with temperature is determined by the energy conservation constraint that is independent of momentum conservation (Ernst and Das [92]). The key ingredient of many known thermal models is the presence of a velocity distribution instead of one single speed. In this velocity distribution the *modulus* of the particle velocity may vary in addition to its direction. The typical example of a *multi-speed* thermal model is the 19-velocity model of Grosfils et al. [93] where four species of particles move on a hexagonal lattice: the “fastest” particles travel two lattice spacings in one timestep, “fast” particles travel to second nearest neighbor sites, normal, speed-one particles to nearest neighbor sites and, finally, rest particles move not at all. The choice of collision rules must conserve mass, momentum and kinetic energy independently.

There are as well other approaches for constructing LG models with thermal properties (Shan [97] and, recently, Swift, Osborn and Yeomans [95]; Orlandini, Swift and Yeomans [95]; Swift et al. [96]; Gonella, Orlandini and Yeomans [97]; Wagner and Yeomans [98]). These approaches employ the so-called Lattice-Boltzmann method (Section 2.5).

We note that a novel, alternative, method for constructing thermal LGs will be presented and discussed in Chapter 3 of this thesis. However the basic concepts and investigation tools that are valid for multi-speed thermal LG models constitute the necessary theoretical background for the introduction of our method and will be the subject of the following discussion in this Chapter.

2.3.1 Models with Kinetic Energy Conservation

Let us consider the class of models on a lattice, where the velocity of fictitious particles can take a limited number of discrete values $\mathbf{c}_i, i = 1..b$, where the bit-number b now is not (necessarily) the number of nearest neighbors (z), but is the total number of allowed different velocities. Comparing to the FHP model of Section 2.2, the velocity can point not only to nearest neighbors, but to next nearest neighbors and so on. Obviously, to cover larger than the nearest neighbor distance in one timestep, a particle must have a larger velocity, and hence, larger momentum and kinetic energy. Also, rest particles with zero velocity can be present in the system, and can

change their velocity during a collision step. In such a system, the collision table with collisions that conserve mass and momentum is significantly large. As a purely combinatorial exercise, we could build a collision table that includes collisions that conserve mass, momentum and kinetic energy. An illustrative example of a model with kinetic energy conservation is the 19-bit ($b = 19$) model of Grosfils, Boon and Lallemand [92]. In Figure 2.3 the elementary collision rules for this model are grouped

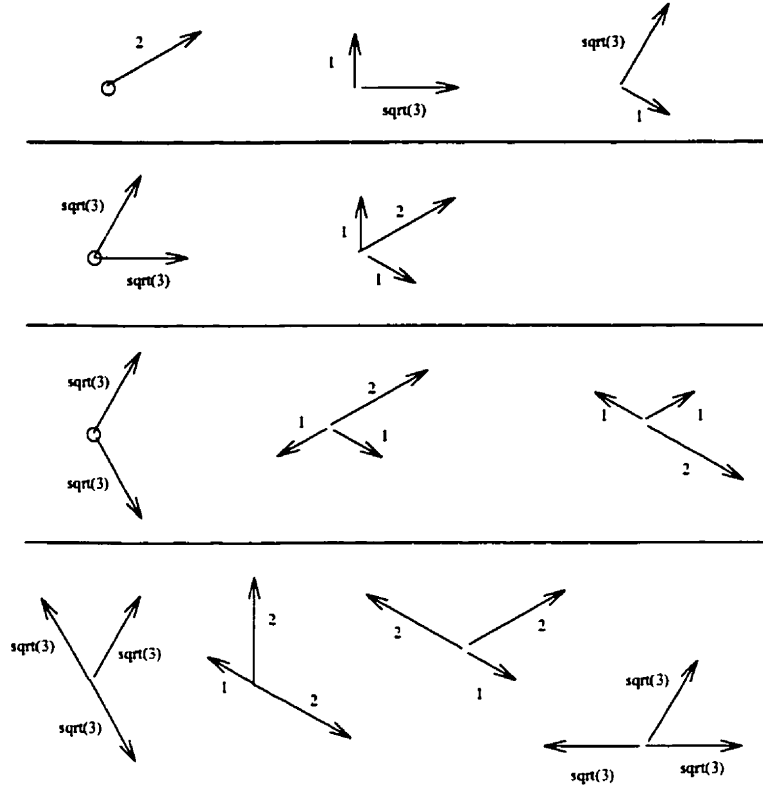


Figure 2.3: Elementary collisions for the 19-bit model that transfer energy. The circle marks rest particle. The arrows with labels 1 , $\sqrt{3}$, 2 indicate moving particles with the corresponding speeds.

in classes according to the values of the invariants. The actual number of states in each class is large - up to 257 (Grosfils, Boon and Lallemand [92]).

2.3.2 Microscopic Dynamic Equation of LGCA

The space state of all Lattice Gas Cellular Automata (LGCA) models at time t is defined by the occupation numbers $n(\mathbf{r}, t)$ (integer), namely the number of particles at any given site $\mathbf{r}$ of the lattice. As discussed before, each particle has its own (discrete) momentum state, determined by its velocity $\mathbf{c}_i, i = 1..b$. Suppose, for a

moment, that the particles simply propagate without colliding. Then the dynamics would be described by the following free streaming equation:

$$n_i(\mathbf{r} + \mathbf{c}_i \Delta t, t + \Delta) = n_i(\mathbf{r}, t) \quad (2.6)$$

where Δt is one time-step of evolution, as described in Section 2.2. Let us for convenience set $\Delta t = 1$ unit of time. In rest of the thesis we will use this convention. The addition of collisions introduces a *collision operator* on the right-hand side of the above equation. That is, we have

$$n_i(\mathbf{r} + \mathbf{c}_i, t + 1) = n_i(\mathbf{r}, t) + \Omega_i(n_*(\mathbf{r}, t)), \quad (2.7)$$

where the collision operator, Ω_i , describes the change in n_i due to collisions, and is a nonlinear function of a limited set of occupation numbers $n_*(\mathbf{r}, t)$ in the small neighborhood of a given site $\mathbf{r}$ (*i.e* particles flow from this neighborhood to form a precollision state and flow back to it after collision). In other words, the collision operator depends on the occupation function n for all possible values of the index represented by the asterisk. In general, the collision process at a fixed lattice site and timestep can be fully specified by a *transition matrix* A , whose element $A(s \rightarrow s')$ is the probability that a local configuration $s = \{n_i(\cdot), i = 1, \dots, b\}$ (input state) is transformed into a configuration s' (output state). For instance, in the FHP model, Figure 2.2 shows that the probability of the transition

$$s = (0, 1, 0, 0, 1, 0) \rightarrow (0, 0, 1, 0, 0, 1) = s' \quad (2.8)$$

is

$$A(s \rightarrow s') = \frac{1}{2} \quad (2.9)$$

where zeros and ones are the absence and presence of a particle with a particular velocity (out of six). Clearly, there are $2^6 = 64$ input states and 64 possible output states, and thus A is a 64×64 matrix for the FHP model. In general, because of the requirements of conservation laws, A is a sparse matrix (with the majority of matrix elements being zeros). Since each incoming state gives rise to exactly one outgoing state, it is required that

$$\sum_{s'} A(s \rightarrow s') = 1. \quad (2.10)$$

The relationship of this matrix to the collision operator Ω will be shown in Section 2.3.6.

2.3.3 Conserved Quantities

A distinguishing feature of lattice gases is the presence of some number of conserved quantities. The presence of conserved quantities is the key argument in understanding why we should expect the bulk behavior of particles moving and colliding on a lattice to be that of a fluid. On a more theoretical level, conserved quantities ensure the existence of respective conjugate thermodynamic variables and the existence of an equilibrium state of the fluid corresponding to these quantities.

The quantities that are conserved may differ from one LGCA model to another. In the present example of a thermal LGCA we have

$$\text{number of particles:} \quad \rho(\mathbf{r}, t) = \sum_i n_i(\mathbf{r}, t) \quad (2.11)$$

$$\text{momentum:} \quad \mathbf{g}(\mathbf{r}, t) = \sum_i \mathbf{c}_i n_i(\mathbf{r}, t) \quad (2.12)$$

$$\text{kinetic energy:} \quad \varepsilon(\mathbf{r}, t) = \sum_i \frac{1}{2} c_i^2 n_i(\mathbf{r}, t) \quad (2.13)$$

More explicitly the total number of particles $N = \sum_{\mathbf{r}} \rho(\mathbf{r}, t)$, the total momentum of the system $P = \sum_{\mathbf{r}} \mathbf{g}(\mathbf{r}, t)$ and the total kinetic energy $H = \sum_{\mathbf{r}} \varepsilon(\mathbf{r}, t)$ are constants of the motion.

Because the local occupations $n_i(\mathbf{r}, t)$ can change only through free streaming but not through collisions, the collision term in equation (2.7) must satisfy the relation:

$$\sum_i q(c_i) \Omega_i(n_*(\mathbf{r}, t)) = 0 \quad (2.14)$$

where $q(c_i) = \{1, \mathbf{c}_i, \frac{1}{2} c_i^2\}$.

Finally we note that if energy is conserved, one has a *thermal* lattice gas model in which temperature can be defined in accord with thermodynamics. If the energy is not conserved or is simply a multiple of the particle density (as it is, for example in the FHP model, Section 2.2), one has an *athermal* lattice gas model.

2.3.4 Statistical Averages and The Boltzmann Equation

The precise knowledge of the location and state of each and every particle in the system is often more information than one really desires. So we now consider some

statistical aspects of LGCA theory. Let us introduce the probability $f_i(\mathbf{r}, t)$ of finding a particle in a state with velocity $\mathbf{c}_i$ at site $\mathbf{r}$ at time t as

$$f_i(\mathbf{r}, t) = \langle n_i(\mathbf{r}, t) \rangle \quad (2.15)$$

By angular brackets $\langle \rangle$ we denote the ensemble average. Note that while the n_i 's are integer (binary for LGCA), the f_i 's have values in the set of real numbers.

A coarser description, such as a closed set of kinetic equations for the one-particle distribution function defined in (2.15) is then a more appropriate description of the system. From this description we expect to get the hydrodynamic behavior of the system.

To obtain a closed set of equations for the f_i , we take the ensemble average of (2.7). We are immediately confronted by the fact that the collision operator is generally a nonlinear function of the occupation numbers n_i . Thus, the simplest approximation that we can make to close the system of equations for the n_i is to assume that the incoming occupation numbers, n_i , are uncorrelated. This is the discrete version of the famous *Boltzmann molecular chaos assumption*. From this assumption, it follows that

$$\langle \Omega_i(n_*) \rangle \approx \Omega_i(\langle n_* \rangle) = \Omega_i(f_*), \quad (2.16)$$

In this way, we get the lattice version of the *Boltzmann equation*,

$$f_i(\mathbf{r} + \mathbf{c}_i, t + 1) = f_i(\mathbf{r}, t) + \Omega_i(f_*(\mathbf{r}, t)) \quad (2.17)$$

2.3.5 Detailed Balance Condition and Boltzmann Equilibrium

One of the important characteristics of LGCA is the existence (or non-existence) of thermodynamically correct equilibrium states.

Let us define the matrix of transition probabilities $A_{ss'}$:

$$A_{ss'} = \langle A(s \rightarrow s') \rangle \quad (2.18)$$

where $A(s \rightarrow s')$ is the transition matrix introduced in Section 2.3.2.

The equilibrium state of an LGCA is the solution of equation (2.17) at late times. This solution must depend only on globally conserved quantities, as discussed in

Section 2.3.3. The standard statistical-mechanical argument (see, for example, Frisch et al. [87]) sets the following condition for the existence of an equilibrium solution

$$\sum_s A_{ss'} = 1 \quad (2.19)$$

It is called the *semi-detailed balance condition*. For the LGCA with a Fermi-exclusion principle for the occupation numbers (Section 2.2), the equilibrium distribution then takes the form of the Fermi-Dirac distribution (Frisch et al. [87])

$$f_i = \frac{1}{1 + e^{-\sum_\lambda b_\lambda q_\lambda(c_i)}} \quad (2.20)$$

where the q are collisional invariants as in (2.14), and the set of thermodynamic fields $b(\mathbf{r}, t)$ includes the chemical potential $\mu(\mathbf{r}, t)$, the reciprocal thermodynamic temperature $\beta(\mathbf{r}, t)$ and the conjugate to the momentum $\gamma(\mathbf{r}, t)$. If the semi-detailed balance condition (2.19) is not satisfied, the equilibrium distributions are not simply functions of the invariants, and depend explicitly on the transition rules. Such models cannot be described in terms of correct thermodynamics, and hence are not suited to our goals (Section 1.2).

We note, that because the evolution of the LG models considered in this thesis is defined by a Monte-Carlo algorithm (see Chapter 3), our models obey the even stronger condition of *detailed balance*

$$A_{s's} = A_{ss'} \quad (2.21)$$

requiring that the forward and backward collision rates be equal. On account of (2.10), this satisfies the semi-detailed balance condition (2.19).

2.3.6 Linear Approximation to the Boltzmann Equation

The dynamics of a lattice gas (both in equilibrium and non-equilibrium) within acceptable approximations is governed by the Boltzmann equation (2.17). However, because of the nonlinearity and complexity of the collision operator¹ the practical use of this equation is difficult.

¹In Section 4.2 we show an example of building the collision operator from collision-rule table.

Here we therefore outline the method of linear approximation to solve the Boltzmann equation (2.17) for models with a well defined equilibrium state¹. The linear approximation to the collision operator is then used in a Chapman-Enskog expansion (Section 2.4.1) to recover the hydrodynamic equations as a solution of the Boltzmann equation with mass and momentum conservation constraints. It is also used in the evaluation of the transport properties of LGCA models from a Structure Factor analysis (Section 2.4.2).

The approach assumes that the distribution function f_i differs only slightly from the equilibrium form f_i^0 . Thus we can make an approximation:

$$f_i = f_i^0 + \Phi_i \quad \left(\frac{|\Phi_i|}{f_i^0} \ll 1 \right) \quad (2.22)$$

With this approximation the collision term Ω_i in the Boltzmann transport equation may be approximated by a power series expansion in Φ_i

$$\Omega_i = \sum_j \Omega_{ij}^{(1)} \Phi_j + \sum_{jk} \Omega_{ijk}^{(2)} \Phi_j \Phi_k + \dots \quad (2.23)$$

where $\Omega^{(1)}$ is the linearized collision operator. In a spatially uniform system close to equilibrium one may use a linear approximation to the Boltzmann equation

$$f_i(\mathbf{r} + \mathbf{c}_i, t + 1) - f_i(\mathbf{r}, t) = \sum_j \Omega_{ij}^{(1)} \Phi_j \quad (2.24)$$

This equation can be solved in terms of the eigenvalues α_λ and eigenvectors $|\Psi_\lambda\rangle$ of the matrix $\Omega^{(1)}$ i.e.,

$$\Omega^{(1)} |\Psi_\lambda\rangle = \alpha_\lambda |\Psi_\lambda\rangle \quad (2.25)$$

Then, the Φ_i may always be written (Wolfram [86]) as sums of pieces proportional to each of the orthogonal eigenvectors $|\Psi_\lambda\rangle$

$$\Phi_i = \sum_\lambda B_\lambda |\Psi_\lambda\rangle \quad (2.26)$$

and the general solution of (2.24) is given in terms of equation (2.26) by

$$B_\lambda(t) = B_\lambda(0)e^{\alpha_\lambda t} \quad (2.27)$$

¹It is also valid for the systems without statistical-mechanical equilibrium in the sense of (Section 2.3.5), and for the general class of systems which possess solutions of the Boltzmann equation converging to steady probability distributions.

In Section 2.4.2 (see discussion after equation (2.44)) we will show that all nonzero α_λ have negative real parts. Hence, the associated B_λ must decay exponentially with time. Only the combinations of Φ_i associated with zero eigenvalues survive at large times.

It is easy to show that the set of eigenfunctions Ψ_λ with zero eigenvalues is proportional to the set of conserved quantities $q_\lambda(c_i) = \{1, c_i, \frac{1}{2}c_i^2, \dots\}$ defined in (2.14). Since $\Omega_i(f^0(b_\lambda)) = 0$ for any set of b_λ conjugate to the conserved quantities q_λ , it follows that

$$0 = \sum_j \frac{\partial \Omega_i}{\partial b_\lambda} = \sum_j \frac{\partial \Omega_i}{\partial f_j} \frac{\partial f_j^0}{\partial b_\lambda} = \sum_j \Omega_{ij}^{(1)} \frac{\partial f_j^0}{\partial b_\lambda} \quad (2.28)$$

The derivatives $\frac{\partial f_j}{\partial b_\lambda}$, in turn, are proportional to the conserved quantities

$$\frac{\partial f_j}{\partial b_\lambda} = k_j q_\lambda(c_j) \quad (2.29)$$

For example, for the equilibrium distribution (2.20) we have

$$\frac{\partial f_j^0}{\partial b_\lambda} = q_\lambda(c_i) f_j^0 (1 - f_j^0) \quad (2.30)$$

Because of (2.28) the $q_\lambda(c_j)$ are indeed eigenvectors of $\Omega_{ij}^{(1)}$ with zero eigenvalues. Hence corresponding eigenvectors with zero eigenvalue play a special role and are associated with the conservation laws (Section 2.3.3).

This result supports the local equilibrium assumption used for the derivation of hydrodynamic equations. It implies that regardless of the initial average densities Φ_i , collisions bring the system to an equilibrium that depends only on the values of the macroscopic conserved quantities.

2.4 Transport Properties of LGCA

The most interesting physics of fluids appears when a system is driven away from its equilibrium state¹. The response of a system to an external perturbation is known to be related to transport phenomena. The strength of this response is characterized by transport coefficients, some of which, such as the shear and bulk viscosities, were already introduced in Section 2.1. Others include the thermal diffusion coefficient D_T

¹Equilibrium in the general sense, with zero or non-zero total velocity flow $\mathbf{u}$.

as the measure of response to thermal perturbations, and the attenuation coefficient Γ for sound waves.

In general, the transport coefficients $L_{qq'}$ define the linear response of a system to an external perturbation (McLennan [89])

$$J_q(\mathbf{r}, t) = - \sum_{q'} L_{qq'} \nabla b_{q'}(\mathbf{r}, t) \quad (2.31)$$

where $\{\nabla b_{q'}\}$ is the set of thermodynamic driving forces. $J_q(\mathbf{r}, t)$ is the current associated with the conserved density $q(\mathbf{c}_i) = \{1, \mathbf{c}_i, \frac{1}{2}c_i^2, \dots\}$

$$J_q(\mathbf{r}, t) = \sum_i c_{ix} q(\mathbf{c}_i) \delta n_i(\mathbf{r}, t) \quad (2.32)$$

where the direction of the current is assumed parallel to the x axis. In terms of the above equations:

- the *heat conductivity* λ (the thermal diffusion coefficient D_T) is defined as the coefficient of proportionality between the dissipative heat current and the temperature (reciprocal temperature) gradient, *i.e.*,

$$\mathbf{q}^D = -\lambda \nabla T \equiv L_{TT} \nabla \beta \quad \text{with} \quad L_{TT} = D_T = k_B T^2 \lambda \quad (2.33)$$

- the transport phenomena related to the gradient of the vector flow field $\mathbf{u}$ involves the fourth-rank viscosity tensor $\nu_{\alpha\beta\gamma\delta}$

$$\sigma_{\alpha\beta}^D = -\rho \nu_{\alpha\beta\gamma\delta} \nabla_\gamma u_\delta \quad (2.34)$$

where $\alpha\beta\gamma\delta = \{x, y, z, \dots\}$ label Cartesian components, $\sigma_{\alpha\beta}^D$ is a dissipative part of the stress tensor and summation over repeated indices is implied. We restrict ourselves to those models which provide a fourth-rank tensor $\nu_{\alpha\beta\gamma\delta}$ having the full fluid symmetry¹ so that it can be expressed in terms of two independent scalars. We then obtain the shear viscosity $\nu = \nu_{\alpha\beta\alpha\beta}$ and the bulk viscosity $\zeta = (\frac{1}{d^2}) \nu_{\alpha\alpha\beta\beta}$ in the well known form from continuous fluid theory

$$\nu_{\alpha\beta\gamma\delta} = \nu(\delta_{\alpha\gamma}\delta_{\beta\delta} + \delta_{\alpha\delta}\delta_{\beta\gamma} - \frac{2}{d}\delta_{\alpha\beta}\delta_{\gamma\delta}) + \zeta\delta_{\alpha\beta}\delta_{\gamma\delta} \quad (2.35)$$

¹The requirement of the full symmetry, or *isotropy* is the main reason that most of the 2D LG models are defined on the hexagonal lattice (as opposed to a square lattice, for example). This will be discussed in more detail in Section 2.4.3.

As in real fluids, there are several ways to evaluate the transport properties of lattice gas models of hydrodynamics. Here we distinguish two ways for doing so. Figure

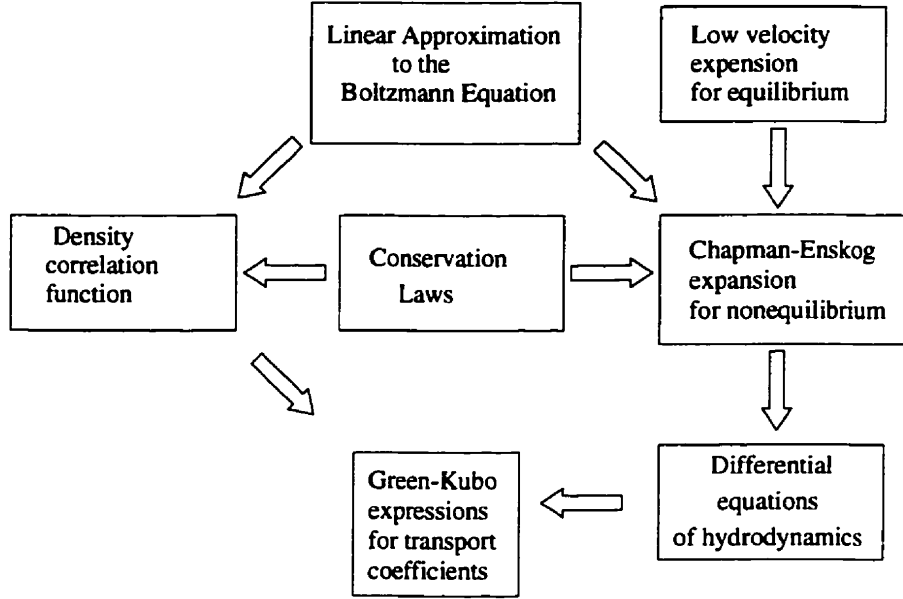


Figure 2.4: Chart of different ways to get Transport Coefficients in LGCA

2.4 summarizes them. Both of them use the idea of the linear approximation to the Boltzmann equation (Section 2.3.6). The Chapman-Enskog expansion (right-column part of Figure 2.4) allows us to recover the differential equations of hydrodynamics (Section 2.1) and will be discussed later in Section 2.4.1. The density fluctuation analysis (left-column part of Figure 2.4) is often a more convenient approach for the models where an explicit form of the collision matrix is relatively easy to obtain. This approach allows us to obtain the Dynamic Structure Factor, as will be outlined in Section 2.4.2.

2.4.1 The Chapman-Enskog Expansion

The traditional Chapman-Enskog method (Chapman and Cowling [70]) for real fluids and its lattice-gas adaptation (Frisch et al. [87]; Chen et al. [91a], also, Section 3.5 of this thesis) involves the series expansion of solutions of the Boltzmann equation (equation (2.17) for lattice gases) around the equilibrium solution¹ and recovers the macroscopic equation of hydrodynamics (Section 2.1) from conservation laws. The

¹In equation (2.22) the expansion in terms of function Φ_i .

starting point here is the expansion of the equilibrium solution in terms of a not-so-large flow-velocity vector $\mathbf{u}^1$. In general, the equilibrium distribution is a function of the invariants q_λ and their conjugate thermodynamic fields b_λ . For the Fermi-Dirac distribution, (2.20), we may write:

$$f_i^0 = \mathcal{F}(h + \mathbf{q} \cdot \mathbf{c}_i) \quad (2.36)$$

$$h(\rho, \mathbf{u}) = h_0 + h_2 u^2 + \mathcal{O}(u^4) \quad (2.37)$$

$$\mathbf{q}(\rho, \mathbf{u}) = q_1 \mathbf{u} + \mathcal{O}(u^3) \quad (2.38)$$

The condition of invariance under the reflections $\mathbf{u} \rightarrow -\mathbf{u}$, $\mathbf{c}_i \rightarrow -\mathbf{c}_i$ implies that h must be an even function of $\mathbf{u}$ and $\mathbf{q}$ must be odd. A Taylor expansion around $u = 0$ then gives

$$f_i^0 = \mathcal{F}(h_0) + \mathcal{F}'(h_0)(q_1 \mathbf{u} \cdot \mathbf{c}_i + h_2 u^2) + \frac{1}{2} \mathcal{F}''(h_0) q_1^2 (\mathbf{u} \cdot \mathbf{c}_i)^2 + \mathcal{O}(u^3) \quad (2.39)$$

This expression can be simplified using the conservation conditions (2.11),(2.12). Thus, for the Fermi-Dirac distribution one arrives (Frisch et al. [87]) at

$$f_i^0 = \frac{\rho}{6} + \frac{\rho}{3c} \mathbf{c} \cdot \mathbf{u} - \frac{1}{3} \rho g(\rho) \frac{u^2}{c^2} + \frac{2}{3} \rho g(\rho) c_{i\alpha} c_{i\beta} \frac{u_\alpha u_\beta}{c} \quad (2.40)$$

where $\alpha\beta$ are the Cartesian coordinate indices and the factor g given by

$$g(\rho) = \frac{3 - \rho}{6 - \rho} \quad (2.41)$$

is so-called Galilean factor, discussed in Section 2.4.3.

From the microscopic conservation laws one obtains the macroscopic equations of hydrodynamics (Section 2.1). The transport coefficients then are obtained as parameters in front of the corresponding terms in the macroscopic hydrodynamic equations. In Section 3.5 of this thesis we will follow the Chapman-Enskog method to arrive at the macroscopic equations of hydrodynamics and at transport coefficients for a new generalized class of lattice gas models.

¹The condition of validity of the results based on low-velocity expansion is $\frac{u}{c_s} \ll 1$, where c_s is the speed of sound, which is typically $\mathcal{O}(c_i)$.

2.4.2 Linear Response and Dynamic Structure Factor

In this section we introduce the formal definition and general properties of a time-correlation function, namely the dynamic structure factor, and thus establish the link between spontaneous time-dependent fluctuations and the response of a lattice gas to an external probe. The importance of density fluctuations in LGCA models lies in the fact that LGCA fluctuations, as well as those in real fluids, can be measured with a high degree of accuracy using computer simulations and light scattering experiments respectively (Hansen and McDonald [89], p.10). Thus the value of an LGCA as a model of a real fluid can be easily seen.

The Dynamic Structure Factor $S(\mathbf{k}, \omega)$ is defined as the double Fourier transform with respect to space and time of the correlation function of density fluctuations $\delta\rho(\mathbf{r}, t)$ around the equilibrium state:

$$\rho S(\mathbf{k}, \omega) = \sum_{\mathbf{r}} \sum_{t=-\infty}^{\infty} e^{-i\omega t - i\mathbf{k} \cdot \mathbf{r}} \langle \delta\rho(\mathbf{r}, t) \delta\rho(0, 0) \rangle = \sum_{t=-\infty}^{\infty} e^{-i\omega t} V^{-1} \langle \delta\rho(\mathbf{k}, t) \delta\rho^*(0, 0) \rangle \quad (2.42)$$

where $\rho(\mathbf{k}) = \sum_{\mathbf{r}} \exp(-i\mathbf{k} \cdot \mathbf{r}) \rho(\mathbf{r})$ is the spatial Fourier transform of the density and V is the number of sites in the d -dimensional lattice.

The Fourier transformation of the linearized lattice-gas Boltzmann equation (2.24) yields, to first order in the fluctuation δn_j :

$$\delta n_i(\mathbf{k}, t+1) = \sum_j e^{-i\mathbf{k} \cdot \mathbf{c}_i} (\delta_{ij} + \Omega_{ij}^{(1)}) \delta n_j(\mathbf{k}, t) \quad (2.43)$$

Hence we can define an important quantity in the non-equilibrium description of LGCA, namely the kinetic propagator Γ , as:

$$\Gamma_{ij}(\mathbf{r}, t) k_j = \langle \delta n_i(\mathbf{k}, t) \delta n_j^*(\mathbf{k}, 0) \rangle = [e^{-i\mathbf{k} \cdot \mathbf{c}} (\mathbf{1} + \Omega^{(1)})]_{ij}^t k_j \quad (2.44)$$

where k_j is the equal-time correlation function of the δn_i 's given by (2.29). This form of the kinetic propagator as the t -th power of $e^{-i\mathbf{k} \cdot \mathbf{c}} (\mathbf{1} + \Omega^{(1)})$ immediately implies that the eigenvalues of the matrix $(\mathbf{1} + \Omega^{(1)})$ must have modulus < 1 ¹. Otherwise the expression (2.44) diverges.

¹This justifies the argument (in Section 2.3.6) about the determining role of zero eigenvalues and eigenvectors in the evolution of the model.

Summing equation (2.44) over (i, j) and taking the temporal Fourier transform yields the dynamic structure factor (2.43) in Boltzmann approximation in a form that is too complicated to be expressed analytically. However, when one is interested in the dynamic structure factor in the *hydrodynamic regime*, then the method of Landau and Placzek (Resibois and de Leener [77]) allows the calculation of $S(\mathbf{k}, \omega)$ analytically in the limit of $\mathbf{k} \rightarrow 0, \omega \rightarrow 0$.

We refer to Grosfils et al. [93] for the detailed derivation of the lattice gas version of the structure factor in the hydrodynamic limit. Here we note that in such a derivation one uses the eigenvalue equation for the non-symmetric matrix $e^{-i\mathbf{k}\cdot\mathbf{c}}(\mathbf{1} + \Omega^{(1)})$,

$$e^{-i\mathbf{k}\cdot\mathbf{c}}(\mathbf{1} + \Omega^{(1)}) | \Psi_\lambda(\mathbf{k}) \rangle = e^{z_\lambda(\mathbf{k})} | \Psi_\lambda(\mathbf{k}) \rangle \quad (2.45)$$

Because of (2.25) and (2.29) the eigenvalues (or eigenmodes) with $z_\lambda(\mathbf{k}) = 0$ at $k = 0$ are called hydrodynamic modes and the corresponding eigenvectors $| \Psi_\lambda(\mathbf{k}) \rangle$ and eigenvalues $z_\lambda(\mathbf{k})$ can be expanded in a Taylor-series with ik as a small parameter

$$\Psi_\lambda(k) = \Psi_\lambda^{(0)} + ik\Psi_\lambda^{(1)} + (ik)^2\Psi_\lambda^{(2)} \quad (2.46)$$

$$z_\lambda(\mathbf{k}) = ikz_\lambda^{(0)} + (ik)^2z_\lambda^{(2)} \quad (2.47)$$

The coefficients of the expansion can be expressed in terms of thermodynamic derivatives such as the speed of sound c_s , and transport coefficients such as the kinematic viscosity ν , the heat diffusivity D_T , and the sound damping constant Γ . It is shown¹ that the dominant contributions to the structure factor in the small k approximation come from hydrodynamic modes: the shear mode ($\lambda = \perp$), the heat mode ($\lambda = T$) and the two sound modes ($\lambda = \sigma = \pm$). In particular, the expansion (2.47) reduces to

$$\begin{aligned} z_\perp(k) &= -\nu k^2 \\ z_T(k) &= -D_T k^2 \\ z_\sigma(k) &= -i\sigma c_s k - \Gamma k^2 \end{aligned} \quad (2.48)$$

The sound damping constant Γ is found to be dependent on the other coefficients as:

$$\Gamma = \frac{1}{2}\nu + \frac{1}{2}(\gamma - 1)D_T \quad (2.49)$$

¹In Section 3.6 we present in more detail the linear response theory for a new generalized class of models.

Here γ is the ratio of squares of the speeds of sound, adiabatic and isothermal respectively¹. The relation (2.49) differs from the one for real (two dimensional) fluids in the absence of a bulk viscosity: because the particles possess only kinetic energy, the bulk viscosity is zero (Ernst and Das [92]). However, we will show in Section 3.6 that for a class of models studied in this thesis the non-zero bulk viscosity changes the relation (2.49) to a new one, (3.64).

Finally, in terms of the above coefficients of the small k and ω expansions, one arrives at the expression for the dynamic structure factor in the hydrodynamic regime, known in the literature as the Landau-Placzek approximation:

$$\begin{aligned} \frac{S(\mathbf{k}, \omega)}{S(\mathbf{k})} &= \left(\frac{\gamma-1}{\gamma}\right) \frac{2D_T k^2}{\omega^2 + (D_T k^2)^2} + \frac{1}{\gamma} \sum_{\pm} \frac{\Gamma k^2}{(\omega \pm c_s k)^2 + (\Gamma k^2)^2} \\ &+ \frac{1}{\gamma} [\Gamma + (\nu-1)D_T] \frac{k}{c_s} \sum_{\pm} \frac{(c_s k \pm \omega)}{(\omega \pm c_s k)^2 + (\Gamma k^2)^2} \end{aligned} \quad (2.50)$$

The first two terms in (2.50) represent three symmetric Lorentzians. The first one is the Rayleigh peak with a width $D_T k^2$ and the other two Doppler shifted ($\pm c_s k$) lines are the Brillouin peaks with a width Γk^2 . The last term in (2.50) describes asymmetric contributions to the Brillouin lines (there are no asymmetric corrections to the Rayleigh line).

The power spectrum (2.50) consisting of the Rayleigh line and the asymmetric Brillouin lines, derived here from the Boltzmann equation for the lattice gas, is completely analogous to the results of the Landau-Placzek theory for continuous isotropic fluids (Boon and Yip [91]).

2.4.3 Artifacts of Discreteness

There is always a penalty for trying to model complex systems with simple models. In the LGCA method special concern must be devoted to the effect of the discreteness of space, time, velocity, etc. on the macroscopic behavior of the system.

First, the discreteness of space may affect the *isotropy of fluid behavior*. The isotropy turns out to be dependent on the symmetry of the lattice. For the square lattice in two dimensions one gets orientation-dependent transport properties (Das and Ernst [92]). Isotropic fourth-order tensors $\sum_i c_{i\alpha} c_{i\beta} c_{i\gamma} c_{i\delta}$, where $\alpha\beta\gamma\delta$ are Cartesian

¹We will define these later in Section 3.3.

coordinates, play a crucial role in the lattice gas models of hydrodynamics because of the convective nonlinear term $\mathbf{u} \cdot \nabla \mathbf{u}$ in the Navier-Stokes equation (2.5). Such tensors are isotropic for the hexagonal lattice in 2D and this is the reason why *a hexagonal lattice is used for hydrodynamic models, rather than a square lattice*.

The other consequence of discreteness is the possible *lack of Galilean invariance* of the models. It is manifested in the presence of the additional density dependent factor $g(\rho)$ (2.41) in front of the inertial term in the Navier-Stokes equation obtained for LGCA models. The convective derivative operator

$$\frac{D}{Dt} \equiv \frac{\partial}{\partial t} + g(\rho) \mathbf{u} \cdot \nabla \quad (2.51)$$

and hence the corresponding differential equation of hydrodynamics, is not invariant under the transformation of coordinates $x' = x - \mathbf{V}t$, $t' = t$, unless the factor $g(\rho)$ is exactly unity. There are several ways to fix this problem. For incompressible flow, for which ρ and hence $g(\rho)$ are constant, the easiest solution is to scale $\mathbf{u}$ and the pressure P by $g(\rho)$. Another approach introduces additional particle velocities to force $g(\rho)$ to unity (Gunstensen and Rothman [91]). Also, the lattice Boltzmann method, discussed in following Section, allows us to eliminate non-Galilean invariance completely (Chen, Chen and Matthaeus [92]), since in this method we control the form of the equilibrium distribution. In this thesis we propose new models of fluid that similarly may not possess Galilean invariance. However, these models can be constructed either as particle-based models or as lattice Boltzmann models: the second choice is obviously preferable in applications where Galilean invariance is of special concern.

Further, the discreteness of a model's variables of state can cause the existence of global conserved quantities that are not intentionally built into the model. Examples include momentum conserved along each lattice line and each lattice column in some square lattice models, or conservation related to the checkerboard structure of the lattice (*staggered invariants*, Zanetti [89]). If the spurious invariant is a consequence of the incompleteness of a collision table, it shows up when performing the Chapman-Enskog expansion¹. For the other situations, no evidence of significant effects have

¹The inclusion of three-particle collisions in the FHP model (Section 2.2) is an example of removing a *harmful* invariant. Indeed, a two-particle collision removes a pair of particles with zero total momentum and moves them to another lattice direction. Therefore, it conserves momentum along

been detected in fluid flow simulations. On the other hand, the influence of a spurious invariant on the physical behavior of a cellular automata fluid can be minimized by properly choosing the initial state so that the total unwanted invariant vanishes exactly.

One more concern that follows from dealing with Boolean quantities in LGCA is significant “noise” in the simulation. In order to obtain the macroscopic behavior of a system, one has to average the state of each point in volume over a rather large patch of sites around it (for example a square of 25×25 sites) and over several consecutive timesteps. For b states per site and for coarse-grained averages over patches of N sites, the noise is of order $\approx 1/\sqrt{bN}$. This slows down the speed of the simulation and requires a large system (Orszag and Yakhot [86]). The alternative lattice gas method for simulating fluids - the Lattice Boltzmann method - completely eliminates the statistical fluctuations that are inherent in LGCA. This method will be discussed next.

2.5 The Lattice Boltzmann Method

One of the important and successful variations of LG models for fluids is the so called *Lattice Boltzmann* (LB) method. It is closely related to the LG Cellular Automata method described in previous sections. The distinct feature of the method is that it simulates not the particles but their probabilities of presence. The direct simulation on a computer of the lattice Boltzmann equation (2.17) turns out to be very effective for situations where correlations can be neglected and the Boltzmann molecular chaos assumption is valid. The advantage comes from the fact that we average the microdynamics *before* simulating it rather than after. As mentioned previously, the main drawback is that this approach neglects fluctuations¹.

The variables of state are no longer Boolean variables but probabilities (2.15). Further, useful simplifications are possible for the collision term in the Boltzmann equation (2.17), as long as particle number and momentum are conserved. In its sim-

each lattice direction. The presence of three particle collisions destroys this (unwanted) conservation.

¹Also, due to the roundoff error inherent in manipulations of real numbers on a computer, the LB method no longer maintains the conservation laws exactly. This may have some negative effects on the stability of numerical simulations.

plest form, the lattice Boltzmann dynamics can be written as a relaxation equation, in the spirit of Bhatnagar, Gross and Krook [54]

$$f_i(\mathbf{r} + \Delta t \mathbf{c}_i, t + \Delta t) - f_i(\mathbf{r}, t) = \frac{1}{\tau} (f_i^{(0)}(\mathbf{r}, t) - f_i(\mathbf{r}, t)) \quad (2.52)$$

where τ is some single relaxation time, Δt is 1 time step and the equilibrium solution $f_i^{(0)}$ can be chosen in an appropriate way so as to produce a given behavior. Although this is an uncontrolled approximation, it is useful in our context because it is simple and effective. A natural choice for specifying $f_i^{(0)}$ as a function of ρ and $\rho \mathbf{u}$ would be the expression similar to that obtained for known LGCA models, *i.e.* Equation (2.40). Thus, in the LB method we can choose the solution as

$$f_i^{(0)} = A\rho + \frac{B}{c^2} \rho (\mathbf{c}_i \cdot \mathbf{u}) + E\rho \frac{u^2}{c^2} + \rho \frac{H}{c^4} c_{i\alpha} c_{i\beta} u_\alpha u_\beta \quad (2.53)$$

where A , B , E , and H are coefficients which are determined from the constraints of mass, momentum conservation and the condition of Galilean invariance (see Section 2.4.3). Further, because of the similarity between solutions in the LGCA and LB methods, (2.40) and (2.53), the derivation of the Navier-Stokes equation in the LB scheme is for the most part identical to that for the FHP model. For the detailed derivation of lattice Boltzmann dynamics and basic applications we refer to Benzi, Succi and Vergassola [92].

Recent developments of LB methods (Swift, Osborn and Yeomans [95]; Orlandini, Swift and Yeomans [95]; Swift et al. [96]; Gonella, Orlandini and Yeomans [97]; Wagner and Yeomans [98]) have ensured the thermodynamic consistency of the method, and introduced the possibility of modeling isothermal hydrodynamics for two-phase systems. We will discuss these methods in Section 5.1, Chapter 5, in the context of building our own LG model with interactions.

Generalized Thermal Lattice Gases

3.1 Introduction

In the previous chapter we considered the theoretical aspects of LG models of fluids with correct thermodynamics. The known models of that type (Ernst and Das [92]; Grosfils, Boon and Lallemant [92]; Grosfils et al. [93]; Chen et al. [89]; Chen et al. [91a]) are ones with strict conservation of kinetic energy. They contain particles with different velocity moduli, and hence different kinetic energies. The rules for the time-evolution of models with this energy conservation constraint are often very complex: the model of Grosfils, Boon and Lallemant [92], for example (see Section 2.3.1), is an extension of the FHP model to 4 species of particles with carefully chosen momenta and energies, and in Chen et al. [89], the authors have to introduce unequal masses for particles moving in different channels, in order to ensure a sufficient number of allowed collisions.

Further, the kinetic energy models have few of the prerequisites for the effective simulation of non-ideal fluids. One of the reasons is a limited range of possible densities because of the Fermi-Dirac restriction on occupation numbers. Chen et al. [89], for example, build an FHP-type model where two colliding particles may form a zero velocity “bound particle”. The short-range interaction between bound particles and the process of their creation and annihilation is determined by the temperature $1/\beta$ so that creation or annihilation is associated with an energy cost ΔE , and occurs with probability $\exp(-\beta\Delta E)$. They showed that at certain values of β a phase separation occurs: the high-density phase is occupied mostly by bound-particles, and the low-density phase is mostly the FHP-type fluid. However, because of the exclusion principle, the density of each phase is not optimal for hydrodynamic behavior, and

for the high-density phase the existence of hydrodynamic properties is questionable. In other attempts to include interactions in the existing LGCA models (see Section 5.1 for review of known models with interactions) the consistency with statistical mechanics is broken because of the violation of the semi-detailed balance condition (2.19).

Thermal LB models (Section 2.5) are in principle less complex because they are expressed in terms of the distribution functions for the fictitious particles and allow flexibility in choosing the equilibrium distribution. Alexander, Chen and Sterling [93] have offered a LB model that uses a small number of discrete velocity states and is capable of modeling the thermo-hydrodynamic transport of a (non-interacting) mono-atomic gas. More recent models (Swift, Osborn and Yeomans [95]; Orlandini, Swift and Yeomans [95]; Swift et al. [96]; Gonella, Orlandini and Yeomans [97]; Wagner and Yeomans [98]) allow the incorporation of the pressure term (5.1) in the equilibrium solution so that isothermal hydrodynamics of non-ideal systems can be simulated. However, such models are not easily adapted for situations where variations in the local temperature are important (see Section 1.2).

Indeed, much of the interest in the study of thermal phenomena, particularly those of a non-equilibrium nature (heat conduction) or involving instabilities (convection), concerns the spatial variation of temperature. For these situations it is not possible without modification (Shan [97]) to apply existing LG models.

For non-hydrodynamic systems, on the other hand, models that effectively simulate both equilibrium and non-equilibrium thermal phenomena for systems with interactions are frequently implemented using Monte-Carlo (MC) algorithms (Binder and Heermann [97]). It would be then very advantageous to have a LG model of hydrodynamics that can be simulated with such algorithms.

In this chapter we show that it is possible to build thermal models with MC evolution, and hence, to simulate interesting non-equilibrium phenomena (Section 1.2). Thus, we introduce and analyze a class of thermal models which have well-defined thermodynamics and which permit the inclusion of inter-particle interactions and the simulation of fluids with phase transitions.

3.2 Generalized Thermal Lattice Gas Models

3.2.1 Internal Energy in Particles

The key feature of our new approach in constructing thermal LG models is **the existence of an energy level structure for each particle, independent of the kinetic energy**. In analogy with the basic LGA model (Frisch et al. [87]), we define a class of thermal models in which several species of fictitious particles move on a two-dimensional hexagonal lattice. Some of the particles (rest particles) remain at rest. During each time step, the particles interact (“collide” like billiard balls) (see, for example Figure 2.2) and then propagate ballistically (Figure 2.1) from one site of the lattice to a neighboring site. The collision step rigorously conserves the number of particles and their total momentum, but both the collision steps and the propagation steps can involve changes in the energy of the system.

Energy is defined as the sum of the kinetic and potential energies of the individual fictitious particles, and it is essential that there be at least two species of particles with distinct energies for there to be well-defined thermal properties (Ernst [92]; Ernst [90]; Ernst and Das [92]).

The occupation of energy levels of the different particles is then determined by the thermodynamic temperature in the manner known from the Monte-Carlo literature (see for example Binder and Heermann [97]; Frenkel and Smit [96]). The evolution of the models within the MC framework will be discussed in Sections 3.2.3 and 3.2.4.

3.2.2 Statistics of Occupation Numbers

Further, the **choice of Fermi-Dirac statistics is not essential for our generalized approach**. Historically, it was convenient for coding purposes to represent occupation numbers as binary variables. Other statistics were considered in the literature (Bagnoli, Rechtman and Zanetti [93]; Baran, Wan and Harris [97]; Boghosian et al. [97]) but rarely employed for practical purposes. We exploit the other statistics in two ways. First, we obtain greater flexibility in adjusting the density on which the hydrodynamic transport properties depend. Further, as will be illustrated in Chapter 5, the non-Fermi statistics lead to models which are more efficient for building in interactions and simulating non-ideal fluids with phase-separation behavior. Indeed, in

as much as Lattice Boltzmann methods (Section 2.5) have achieved better success in application to non-ideal fluids with different equilibrium distributions, so our models have the same advantages because of their less restrictive statistics.

3.2.3 Monte-Carlo Algorithms for Evolution

Since our models have well defined energy structures (see Section 3.2.1), the Monte-Carlo method can readily be used to simulate the equilibrium state or relaxation towards equilibrium. This method implies the generation of the different microconfigurations or states (denoted by s) of the system with a probability $\rho(s)$. In a system described by a Hamiltonian $H(s)$, one possibility for the probability $\rho(s)$ is: $\rho(s) = \exp(-\beta H(s))/Z$, where $Z \equiv \sum_s \exp(-\beta H(s))$ is the partition function, and β is the inverse temperature. When the algorithm explores the entire equilibrium phase space (ergodic sampling) to generate states with the probability $\rho(s)$, it correctly simulates the equilibrium distribution function for the canonical ensemble.

One way to ensure that the algorithm generates the states with the correct probability is the use of the Metropolis Monte Carlo scheme (Metropolis et al. [53]), where the probability for accepting a trial move from a microconfiguration s to a microconfiguration s' is given by

$$p(s \rightarrow s') = \min(1, \exp(-\beta \Delta H)), \quad (3.1)$$

where ΔH is the difference in energy between the microconfigurations s and s' . It is straightforward to show (Frenkel and Smit [96]) that the acceptance criteria (3.1) provides detailed balance in equilibrium: the number of accepted trial moves that result in the system leaving state s must be exactly counterbalanced by the number of accepted trial moves from all other states to state s .

The sampling probability $\rho(s)$ in the above example of the Monte Carlo algorithm is defined according to the canonical ensemble. Thus, the temperature is a set parameter and it is not useful to use this algorithm for simulations of local temperature variations, or for the study of phase separating systems at the transition temperature¹. However, there are many other effective Monte Carlo algorithms that

¹In this last case the problem is an inability to control the (relative fraction of the) two phases.

use sampling of states that correspond to other ensembles. One of them, the multi-demon algorithm, has been extensively employed for applications to non-equilibrium thermal phenomena. It is discussed next.

3.2.4 The Multi-Demon Algorithm

The so-called *multi-demon algorithm* of evolution is an alternative Monte Carlo algorithm that allows the study of local temperature variations. The idea was introduced by Creutz [83], and was exploited in a series of papers applying Ising-style lattice gas models to non-equilibrium interface problems (Harris, Jörgenson and Grant [92]; Harris and Grant [90]; Jörgenson, Harris and Grant [89]).

Conservation of energy is enforced with the aid of the so-called demons, one at each site¹. A demon acts as a kind of energy reservoir, permitting collisions in a manner which satisfies detailed balance². At each site, a collision process may proceed *either* if it produces surplus (or zero) energy *or* if the local demon can provide the requisite energy deficit. In the former case, the demon absorbs the surplus. Demons are required always to have positive energy, and as a result serve also to regulate the occurrence of collision processes³. As shown in Appendix A.2 (and demonstrated by Jörgenson, Harris and Grant [89]), the demons thus act as thermometers, measuring the local temperature by virtue of their own statistically averaged energy. It is also possible to use the demons to control the local temperature: if this is done at the boundaries of a sample, then, for example, a temperature gradient can be set up across the sample, permitting a measurement of thermal conductivity, Harris and Grant [88], or the establishing of Rayleigh-Bénard convection (Section 4.3).

A useful way to understand the role of the demons is to consider them as a species of rest particle. Rest particles of the conventional kind can be created or annihilated in collision processes so as to satisfy the conservation laws. Demons are neither created nor annihilated, but act to satisfy the conservation of energy. Conventional rest-particles (usually) have only one energy level, but demons have many such levels

¹In principle it is possible to envisage demons which move with the fluid. However, the interpretation of local temperature fluctuations would then be problematic.

²We return to more details of the implementation of this algorithm in Sections 4.3 and 5.4.

³A process requiring energy which the demon does not possess cannot occur.

- although in simple situations one might imagine demons with only a small number of distinct levels, Creutz [86].

Over the course of time, there will be distribution functions for each local demon energy, which have the form $P(E_D) \sim \exp(-\beta E_D)$, where $\beta = 1/T$ is the inverse of the local temperature. If each E_D were a continuously varying quantity, its average value $\langle E_D \rangle$ would be equal to the local temperature. In practice, however, since the Hamiltonian for a given model may correspond to two or more energy scales, the demon distribution functions are more difficult to interpret: this is particularly true for the interacting model described in Chapters 5 and 6. Details of the analysis for this case are given in Appendix A.2.

3.3 Statistical Equilibrium

As a first step in deriving expressions for the transport coefficients we introduce a notation which generalizes that used in Section 2.3.

As before, sites in the hexagonal lattice are labeled by an index i and by a site vector $\mathbf{r}_i$. The vectors radiating from a site i to its (not-necessarily) nearest neighbors define different possible directions for the motion of fictitious particles at that site. The particles of different total energy can be considered as different species particles. For particles of species I , we have the notation for its velocity $\mathbf{c}_{a,I}$, $a = 1, 2, \dots, b_I$, where b_I is the number of distinct neighbors at distance c_I . This distance also defines the “speed” of the species I , so that the kinetic energy of each fictitious moving particle is $\frac{1}{2}c_I^2$. Each particle of species I also has potential energy ϵ_I , so that its total energy is $E_I = \frac{1}{2}c_I^2 + \epsilon_I$. Any rest particle has total energy zero.

The fictitious particles occupy the available states $\{i, a, I\}$ (or location, direction, total energy) according to particular statistics, and the detailed balance property of the collision rules guarantees that the system possesses a state of statistical equilibrium. In general we write the occupation number of the state $\{i, a, I\}$ as $n_{i,a,I}$, and the ensemble-averaged distribution function for this state as $f_{i,a,I} = \langle n_{i,a,I} \rangle$ (as in (2.15)). It is convenient to represent rest particles in the same way by $n_{i,0}$ and $f_{i,0}$.

In an equilibrium state for which there is no net motion of the fluid, the functions f have the values f_I^{eq} . It is now possible to define thermodynamic variables in terms

of the equilibrium distributions.

The most important of these are the density ρ and the pressure, P . The density is just

$$\rho = M f_0^{eq} + \sum_{a,I} f_I^{eq} \equiv \rho_0 + \sum_I \rho_I \quad (3.2)$$

where M is the number of rest-particle states per site. In principle, the pressure should be derived from the free energy. However, as is well known in the lattice gas literature (Frisch, Hasslacher and Pomeau [86]; Ernst [92]), the quantity which plays the role of pressure in the hydrodynamic equations is

$$P = \frac{1}{2} \sum_I c_I^2 \rho_I \quad (3.3)$$

sometimes known as the “kinetic pressure”. It is useful to define the analogous partial pressures $P_I = \frac{1}{2} c_I^2 \rho_I$, so that $P = \sum_I P_I$. We will also require the energy density $U = \sum_I E_I \rho_I$ ¹, and the isothermal and adiabatic speeds of sound, which are respectively (Frisch et al. [87]; Das, Bussemaker and Ernst [93]; Grosfils et al. [93])

$$c_T^2 = \frac{1}{2} \sum_I \rho_I c_I^2 / \rho \quad (3.4)$$

and

$$c_s^2 = \frac{1}{2} \sum_I f_I^\mu c_I^4 / \sum_I f_I^\mu c_I^2 \quad (3.5)$$

The derivatives f_I^μ of the equilibrium distribution functions f_I^{eq} are defined as $f_I^\mu = T \left(\frac{\partial f_I}{\partial \mu} \right)_T$ where T is the temperature and μ is the chemical potential².

3.4 Time Evolution

As in a traditional LGA (Frisch, Hasslacher and Pomeau [86]), time evolution proceeds in two distinct steps. First, particles at a given site, (both rest particles and

¹Strictly speaking, the energy per unit area (rather than per site) requires an additional factor of $\sqrt{3}/2$. This same factor would appear in the expressions for the thermal conductivity and for the specific heats.

²This is not the definition employed in Frisch et al. [87], although the change in the algebra is minimal. The reason for the change is that it is useful to introduce the thermodynamic variables appropriate to thermal LGA models (Ernst [90]). Because the volume of the system is by definition fixed, it is the density which provides the link with continuum models. This suggests that the notation of the grand canonical ensemble is the most appropriate, so that we will employ the temperature T and the chemical potential μ .

moving particles in any energy level), interact with each other (“collide”) following predetermined rules (Frisch, Hasslacher and Pomeau [86], Figure 2.2). Let the energy conservation be ensured by the local demon, as described previously in Section 3.2, and detailed balance is rigorously observed. Conservation of particles, momentum and energy on each site can be written (in a way similar to equations (2.11), (2.12) and (2.13), but in new notations) explicitly as

$$\sum_{a,I} f_{i,a,I}(t) + f_{i,0}(t) = \text{constant} \quad (3.6)$$

$$\sum_{a,I} \mathbf{c}_a f_{i,a,I}(t) = \text{constant} \quad (3.7)$$

$$\sum_{a,I} f_{i,a,I}(t) E_I + E_{D,i}(t) = \text{constant} \quad (3.8)$$

where t is the (discrete) time. $E_{D,i}$ is the demon energy.

After each collision step, there is a propagation step, in which each moving particle moves distance c_I in the direction a of its velocity, while the rest particles and the demon remain unchanged. Thus the kinetic equations for the particle and demon variables, including both collisions and propagation, are:

$$f_{j,a,I}(t+1) - f_{i,a,I}(t) = \Omega_I, \quad a = 1, \dots, b_I \quad (3.9)$$

$$f_{i,0}(t+1) - f_{i,0}(t) = \Omega_0,$$

$$E_{D,i}(t+1) - E_{D,i}(t) = - \sum_I \sum_a^{b_I} \Omega_I E_I \quad (3.10)$$

where the index j is defined by $\mathbf{r}_j = \mathbf{r}_i + \mathbf{c}_{\mathbf{I}a}$. Ω is the ensemble averaged collision operator. Strictly speaking, $E_{D,i}$ should also be replaced by its ensemble average, $\langle E_{D,i} \rangle$.

3.5 Chapman Enskog Analysis

As discussed in Section 2.4 Chapman-Enskog analysis allows us to recover the continuum equations for a thermal fluid and obtain the transport coefficients. The procedure involves rather lengthy and tedious algebra when carried out for our generalized LGCA with a collision operator Ω that accounts for factor of b_I^2 more possible output states for binary collisions and b_I^3 for three-particles collisions, compared to the

athermal FHP model¹. For the purposes of evaluating the expressions for transport coefficients and recovering the Navier-Stokes (2.5) behavior in our generalized approach we have chosen to carry out the Chapman Enskog expansion within a Lattice Boltzmann approximation, introduced in Section 2.5. Later, in the following section we present the alternative approach to arrive at Green-Kubo expressions for transport coefficients in terms of the exact form of the collision operator.

The lattice Boltzmann approximation to the collision matrix directly employs the existence of equilibrium distribution functions. Any distribution function which deviates from its equilibrium value can be written in a way, similar to (2.22), as $f = f^{eq} + g$, where g is hopefully small. If we assume that there exists a single characteristic relaxation time τ (Bhatnagar, Gross and Krook [54]), then in terms of g , and in our notations, the Boltzmann approximation (2.52), takes the form

$$f_{j,a,I}(t+1) - f_{i,a,I}(t) = -g_{i,a,I}/\tau, \quad a = 1, \dots, b_I \quad (3.11)$$

$$f_{i,0}(t+1) - f_{i,0}(t) = -g_{i,0}/\tau \quad (3.12)$$

These expressions will be employed to simplify the analysis in the subsequent parts of the analysis.

When there exists a net local flow velocity $\mathbf{u}$ it is still possible to define equilibrium distribution functions. In the low velocity limit $u \ll c$, they can be expanded in powers of $\mathbf{u}$. Following Frisch, Hasslacher and Pomeau [86], but with a slightly generalized notation, we obtain to first order²

$$f_{a,I}^{eq}(\mathbf{u}) = f_I^{eq} + q\rho f_I^\mu \mathbf{c}_{a,I} \cdot \mathbf{u} \quad (3.13)$$

The constant q is determined by requiring that the expansions be consistent with the total momentum defined as

$$\rho \mathbf{u} = \sum_{a,I} \mathbf{c}_{a,I} f_{a,I}^{eq} \quad (3.14)$$

¹An example of writing the explicit form of Collision Operator Ω for rather simple scenario with $b_I = 2$ for an example 13-bit model can be found in Section 4.2.2 of the thesis.

²The second order term, as in (2.53), leads to the convective term $(\mathbf{u} \cdot \nabla) \mathbf{u}$ in the Navier-Stokes equation. We have chosen here not to get into lengthy algebraic derivations with all possible terms in the low-velocity equilibrium solution, but to provide only these parts of the analysis that determine the transport coefficients.

We obtain

$$q = 2 / \sum_I b_I f_I^\mu c_I^2 \quad (3.15)$$

$$(3.16)$$

and it is convenient to define $q_I = q f_I^\mu$ so that

$$f_{a,I}^{eq}(\mathbf{u}) = f_I^{eq} + \rho q_I \mathbf{c}_{a,I} \cdot \mathbf{u} \quad (3.17)$$

and $\frac{1}{2} \sum b_I q_I c_I^2 = 1$. Note that to first order in $\mathbf{u}$, there are no corrections to f_0^{eq} or to $P(E_D)$.

We will follow the procedure of Frisch et al. [87]. The first step is to replace the rate equations (3.11), (3.12) by their continuum versions. In the limit where spatial and temporal changes are small and/or slow, the left-hand sides of these equations yield derivatives which can be replaced by $\partial_t = \epsilon \partial_{t_1} + \epsilon^2 \partial_{t_2}$, $\nabla r = \epsilon \nabla r_1$, where ϵ is a small parameter. The distribution functions can also be expanded in terms of ϵ . To order ϵ^2 , this gives:

$$f_{a,I} = f_{a,I}^{(0)} + \epsilon f_{a,I}^{(1)} + \epsilon^2 f_{a,I}^{(2)},$$

and

$$f_{0,I} = f_{0,I}^{(0)} + \epsilon f_{0,I}^{(1)} + \epsilon^2 f_{0,I}^{(2)}$$

where the zeroth order terms in the expansion are just the equilibrium distributions, and the higher order terms are just g : $f^{eq} \equiv f^{(0)}$ and $g \equiv \epsilon f^{(1)} + \epsilon^2 f^{(2)}$. Necessarily, the conservation of mass, momentum and energy require that

$$f_0^{(\alpha)} + \sum_{a,I} f_{a,I}^{(\alpha)} = 0, \quad (3.18)$$

$$\sum_{a,I} \mathbf{c}_a f_{a,I}^{(\alpha)} = 0 \quad (3.19)$$

and

$$E_D^{(\alpha)} + \sum_{a,I} E_I f_{a,I}^{(\alpha)} = 0 \quad (3.20)$$

where $\alpha = 1, 2$.

Substituting into equations (3.11)-(3.12), and equating terms for each order of ϵ , we obtain a hierarchy of Boltzmann equations. The first order equations are:

$$D_{a,I}^{(1)} f_{a,I}^{(0)} \equiv (\partial_{t_1} + \mathbf{c}_{a,I} \cdot \nabla_1) f_{a,I}^{(0)} = -\frac{1}{\tau} f_{a,I}^{(1)}, \quad (3.21)$$

$$\partial_{t_1} f_0^{(0)} = -\frac{1}{\tau} f_0^{(1)}, \quad (3.22)$$

$$\partial_{t_1} E_D^{(0)} = -\frac{1}{\tau} E_D^{(1)}. \quad (3.23)$$

Using (3.18) and (3.19) we then obtain

$$\partial_{t_1}(\rho) + \nabla_1 \cdot (\rho \mathbf{u}) = 0 \quad (3.24)$$

and

$$\partial_{t_1}(\rho \mathbf{u}) + \nabla_1 P = 0 \quad (3.25)$$

where P is the kinetic pressure as defined earlier. Similarly, since these relations are true for any values of the distribution functions $f^{eq}(\mathbf{u} = 0)$, there must also be analogous relations for the partial densities ρ_I and partial pressures P_I , namely

$$\partial_{t_1}(\rho_I) + \frac{\rho_I}{\rho} \nabla_1 \cdot (\rho \mathbf{u}) = 0 \quad (3.26)$$

and

$$\partial_{t_1}(\rho_I \mathbf{u}) + \nabla_1 P_I = 0 \quad (3.27)$$

The expressions for the shear viscosity ν , the bulk viscosity ζ and the thermal conduction coefficient λ arise from the second order terms in ϵ . After some reduction, the corresponding equations become

$$\partial_{t_2}(\rho) + \frac{1}{2} \sum_{a,I} D_{a,I}^{(1)} D_{a,I}^{(1)} f_{a,I}^{(0)} = 0 \quad (3.28)$$

and

$$\begin{aligned} \partial_{t_2}(\rho \mathbf{u}) + \frac{1}{2} \sum_{a,I} D_{a,I}^{(1)} D_{a,I}^{(1)} \mathbf{c}_{a,I} f_{a,I}^{(0)} \\ + \sum_{a,I} \mathbf{c}_{a,I} \mathbf{c}_{a,I} \cdot \nabla_1 f_{a,I}^{(1)} = 0 \end{aligned} \quad (3.29)$$

where the terms preceded by the factor $\frac{1}{2}$ are second order in the Taylor expansion of the derivatives. These terms can be reduced by making use of the equations (3.18) and (3.21). Although equation (3.28) reduces to the simple result $\partial_{t2}(\rho) = 0$, equation (3.29) is more interesting. We simplify its second term to read $\sum_{a,I} \mathbf{c}_{a,I} \cdot \nabla_1 D_{a,I}^{(1)} \mathbf{c}_{a,I} f_{a,I}^{(0)}$, and then substitute for $f_{a,I}^{(1)}$ from (3.21) to obtain

$$\partial_{t2}\rho(\mathbf{u}) = (\tau - \frac{1}{2}) \sum_{a,I} \mathbf{c}_{a,I} \cdot \nabla_1 D_{a,I}^{(1)} \mathbf{c}_{a,I} f_{a,I}^{(0)} \quad (3.30)$$

The final step is to substitute (3.17), and use (3.26) and (3.27) to write

$$\partial_{t2}(\rho\mathbf{u}) = (\tau - \frac{1}{2}) \sum_I (X_I \nabla^2 \rho\mathbf{u} + Y_I \nabla \nabla \cdot \rho\mathbf{u}) \quad (3.31)$$

where

$$X_I = \frac{1}{4} b_I q_I c_I^4$$

and

$$Y_I = \frac{1}{2} (b_I q_I c_I^4 - \rho_I c_I^2 / \rho)$$

Combining with (3.25), this becomes

$$\partial_t(\rho\mathbf{u}) = -\nabla P + (\tau - \frac{1}{2})(\nu \nabla^2 \rho\mathbf{u} + \xi \nabla \nabla \cdot \rho\mathbf{u}) \quad (3.32)$$

Equation (3.32) is in fact the differential equation (2.4) for a compressible viscous fluid, apart from difference in the convective term $(\mathbf{u} \nabla) \mathbf{u}$ ¹. Thus, the shear viscosity ν is

$$\nu = \frac{1}{2} c_s^2 (\tau - \frac{1}{2}) \quad (3.33)$$

and the bulk viscosity ζ is determined from

$$\xi \equiv \frac{\zeta}{\rho} + \frac{1}{2} \nu = (c_s^2 - c_T^2) (\tau - \frac{1}{2}) \quad (3.34)$$

Here we used the expressions for the sound speeds c_T and c_s given earlier. Note that the bulk viscosity vanishes for any model having only one value of the speeds c_I : this includes FHP model, (Frisch, Hasslacher and Pomeau [86]), as a special case.

The procedure to obtain the thermal conduction coefficient, λ , is very similar to that described, for example, in Huang [87]. A key feature is the necessity to subtract

¹As mentioned before, the convective term can be only recovered if we take into account the second order term in low-velocity expansion (3.13).

out of the energy current that part which depends on the net flow of the fluid. This is the origin of the “subtracted current” described by Ernst (Ernst and Das [92]; Ernst [90]), but neglected by Chen et al. [91a]. We account for this effect by replacing the energies E_I by $\tilde{E}_I$ such that

$$\sum_{a,I} \mathbf{c}_{a,I} \tilde{E}_I f_{a,I}^{eq}(\mathbf{u}) = 0 \quad (3.35)$$

or

$$\sum_I b_I f_I^\mu c_I^2 \tilde{E}_I = 0 \quad (3.36)$$

Writing $\tilde{E}_I = E_I - E_S$, we obtain

$$\begin{aligned} E_S &= \sum_I b_I q_I E_I c_I^2 / \sum_I b_I q_I c_I^2 \\ &= \sum_I b_I f_I^\mu E_I c_I^2 / \sum_I b_I f_I^\mu c_I^2 \end{aligned} \quad (3.37)$$

The first order equation describing the time evolution of the local energy density $\tilde{U} = \sum_{a,I} f_{a,I}^{eq}(\mathbf{u}) \tilde{E}_I$ is then

$$\partial_{t1}[\tilde{U} + E_D] = 0 \quad (3.38)$$

which gives no new information. However, after using equations (3.20)-(3.21), the second order equation becomes

$$\partial_{t2}[\tilde{U} + E_D] = (\tau - \frac{1}{2}) \sum_{a,I} \tilde{E}_I \mathbf{c}_{a,I} \cdot \nabla D_{I,a}^{(1)} f_{I,a}^{eq}(\mathbf{u}) \quad (3.39)$$

Using the explicit form of $f_{I,a}^{eq}(\mathbf{u})$, equation (3.17), and the definition of $\tilde{E}$, equation (3.36), we therefore obtain

$$\partial_t[\tilde{U} + E_D] = (\tau - \frac{1}{2}) \sum_I \tilde{E}_I c_I^2 \nabla^2 \rho_I \quad (3.40)$$

It remains to express the gradient explicitly in terms of the thermal gradient. In the absence of a net flow of particles, the chemical potential μ is not a function of position, and so $\nabla^2 f_I = f_I^T \nabla^2 T$, where $f_I^T = \left(\frac{\partial f_I}{\partial T} \right)_\mu$, and the coefficient of thermal conduction is

$$\lambda = (\tau - \frac{1}{2}) \sum_I b_I f_I^T c_I^2 \tilde{E}_I / 2 \quad (3.41)$$

It is convenient to write this also in the form

$$\lambda = (\tau - \frac{1}{2}) \frac{1}{T^2} \sum_I b_I f_I^\beta c_I^2 \tilde{E}_I / 2 \quad (3.42)$$

where $\beta = \frac{1}{T}$ and f_I^β is given by $f_I^\beta = -\left(\frac{\partial f_I}{\partial \beta}\right)_\mu$. The high temperature behavior of λ is dominated by the $\frac{1}{T^2}$ prefactor.

For the particular case of the Fermi-Dirac distribution, it is easy to show that this is precisely the expression given by Ernst [90]. We write $f_I^\beta = -(\mu - E_I) f_I^\mu$ and therefore obtain

$$\begin{aligned} \lambda &= (\tau - \frac{1}{2}) \frac{1}{T^2} \sum_I b_I f_I^\mu c_I^2 \tilde{E}_I (E_I - \mu) / 2 \\ &\equiv (\tau - \frac{1}{2}) \frac{1}{T^2} \sum_I b_I f_I^\mu c_I^2 \tilde{E}_I^2 / 2 \end{aligned} \quad (3.43)$$

where the last step comes from the identity (3.36).

Expressions (3.33), (3.34) and (3.42) are the results for basic transport coefficients in single relaxation time approximation for a wide class of generalized thermal lattice gas models.

3.6 Linear Response Theory

Here we present an alternative approach to the evaluation of transport coefficients of our generalized LG method in terms of the eigenvalues of the linearized collision operator (Figure 2.4). As mentioned before, this method works best for models where a linearized collision operator can easily be obtained analytically¹. The method is outlined in Section 2.4.2. For a multi-speed thermal LGCA the Green-Kubo transport expressions were obtained within this method by Grosfils et al. [93]. In the generalization to our new class of models we take into account the following distinctions from multi-speed thermal LGCA:

- 1) The collision matrix is not symmetric anymore² (this means we treat separately the equations for left and right eigenvalues of collision matrix).

¹When the dimension of the collision matrix is a three digit number, it is still possible to write all the matrix elements, but, obviously, not worth doing so!

²We shall see this later from definitions (3.44.) and (3.45)

- 2) Because of the existence of internal (potential) energy in particles (Section 3.2.1) the different basis set of orthogonal eigenvectors (2.26) must be considered. We shall see that this will lead to non-zero bulk viscosity.

Thus, the idea is to use the lattice gas small k expansions (2.47) to arrive at Green-Kubo type expressions for transport coefficients. As mentioned in Section 2.4.2, the transport properties of LGCA in the hydrodynamic regime are fully determined by the “soft” modes of the kinetic equation (2.45) for the linearized collision operator $\Omega^{(1)}$. The eigenvalues z_λ (2.48) define the transport coefficients of the model: the shear viscosity ν ; the bulk viscosity ζ ; the thermal diffusion coefficient D_T (which applies only to models with strict energy conservation - the adiabatic models); and the attenuation coefficient for sound waves Γ (which is a composite of ν , ζ and D_T).

As discussed in Section 3.3, the state of a particle at any given site is given by the direction and energy labels a, I . Let us denote the total number of states at each site by b . The linearized collision matrix Ω^1 then has dimension² b . We write the analog of the equation (2.14) for our models with conserved kinetic plus potential energy e_i :

$$\begin{aligned} \langle q_n | \Omega = 0 \\ \langle q_n | = q_n(c_i) = \{ \langle \rho |, \langle c_l |, \langle c_\perp |, \langle e | \} = \{ 1, c_i, e_i \}; \quad i = 1, \dots, b \end{aligned} \quad (3.44)$$

Here c_l and $c_\perp$ are two component of a particle's momentum, which we shall call the longitudinal and the perpendicular components. Thus, the collisional invariants determine the left eigenvectors of Ω with zero eigenvalues. To define the right eigenvectors of the collision matrix with zero eigenvalues we use the equation (2.28) adapted to the present models:

$$| q_n \rangle = \{ | \rho \rangle, | c_l \rangle, | c_\perp \rangle, | e \rangle \} = \frac{df_i^{(0)}}{db_n} = k_i^n q_n(c_i) \quad i = 1, \dots, b \quad (3.45)$$

We have assumed the existence of an equilibrium distribution function $f_i^{(0)}$ which is fully defined by the thermodynamic parameters $b_n = \nu, \gamma_l, \gamma_\perp, 1/T$, conjugate to the

¹Here and in the following analysis we omit the superscript (1) introduced in definition of linearized collision matrix 2.23.

²For completely non-restrictive statistics with arbitrary number of particles per site, such as Maxwell-Boltzmann or Bose statistics, b is of order of the number of particles N .

conserved quantities. The difference between equations (3.45) and (2.28) is that in the former the k -factor is “mode-dependent” (with super-script n). This means, that unlike the specific case of Fermi-Dirac statistics (2.20), the collision matrix in general is not symmetric.

We substitute the Taylor-series expansions for the hydrodynamic modes $\Psi_\lambda(\mathbf{k})$ and eigenvalues $z_\lambda(\mathbf{k})$ (2.47) into (2.48) and its left-eigenvector version and obtain

$$\begin{aligned} \Omega | \Psi_\lambda^{(0)} \rangle &= 0; & \langle \Psi_\lambda^{(0)} | \Omega &= 0 \\ \Omega | \Psi_\lambda^{(1)} \rangle &= (c_l + z_\lambda^{(1)}) | \Psi_\lambda^{(0)} \rangle; & \langle \Psi_\lambda^{(1)} | \Omega &= \langle \Psi_\lambda^{(0)} | (c_l + z_\lambda^{(1)}) \\ \Omega | \Psi_\lambda^{(2)} \rangle &= (c_l + z_\lambda^{(1)}) | \Psi_\lambda^{(1)} \rangle & \langle \Psi_\lambda^{(2)} | \Omega &= \langle \Psi_\lambda^{(1)} | (c_l + z_\lambda^{(1)}) \\ &+ [z_\lambda^{(2)} + \frac{1}{2}(c_l + z_\lambda^{(1)})^2] | \Psi_\lambda^{(1)} \rangle; & &+ \langle \Psi_\lambda^{(1)} | [z_\lambda^{(2)} + \frac{1}{2}(c_l + z_\lambda^{(1)})^2] \end{aligned} \quad (3.46)$$

Because of (2.48), it is the eigenvalues to second order $z_\lambda^{(2)}$ which constitute the transport coefficients. Let us define the currents for each hydrodynamic mode λ

$$j_\lambda \equiv (c_l + z_\lambda^{(1)}) \Psi_\lambda^{(0)} \quad (3.47)$$

The second equation of (3.47) then has the following solution¹

$$| \Psi_\lambda^{(1)} \rangle = \frac{1}{\Omega} | j_\lambda \rangle \quad (3.48)$$

By inserting (3.48) into the third equation of (3.47) and multiplying it on the left by $\langle \Psi_\lambda^{(0)} |$, we obtain a Green-Kubo type equation for the transport coefficients

$$z_\lambda^{(2)} = -\langle j_\lambda | \frac{1}{\Omega} + \frac{1}{2} | j_\lambda \rangle / \langle \Psi_\lambda^{(0)} | \Psi_\lambda^{(0)} \rangle \quad (3.49)$$

To obtain the expressions for currents (3.47) and eigenvectors $\Psi_\lambda^{(0)}$ we proceed as follows. As in (2.25), the zero order function $\Psi_\lambda^{(0)}$ can be written as a linear combination of the eigenvectors of matrix Ω which have zero eigenvalues.

$$| \Psi_\lambda^{(0)} \rangle = \sum_{n=1}^b B_n | q_n \rangle \quad \langle \Psi_\lambda^{(0)} | = \sum_{n=1}^b B_n^* \langle q_n | \quad (3.50)$$

It is more convenient, however, to expand it in space of orthogonal eigenvectors. To establish the orthogonal set we introduce entropy and pressure vectors as linear

¹Equation (3.48) is the partial solution of (3.47). As was noted by Grosfils et al. [93], the general solution includes also the terms that depend on the set of $| \Psi_\lambda^{(0)} \rangle$ vectors. However, these terms do not contribute to the transport coefficients.

combinations of energy and density collisional invariants. We require that the entropy vector s be orthogonal to the kinetic pressure vector $p_k = \frac{1}{2}c_l^2$ and that the pressure vector p be orthogonal to the entropy vector. Thus

$$\begin{aligned} |s\rangle &= |e\rangle - h |p\rangle; & \langle s| &= \langle e| - h^* \langle p| \\ h &= \frac{\langle p_k | e \rangle}{\langle p_k | p \rangle}; & h^* &= \frac{\langle e | p_k \rangle}{\langle p | p_k \rangle} \end{aligned} \quad (3.51)$$

$$\begin{aligned} |p\rangle &= |e\rangle - \mu |s\rangle; & \langle p| &= \langle e| - \mu^* \langle s| \\ \mu &= \frac{\langle s | e \rangle}{\langle s | p \rangle}; & \mu^* &= \frac{\langle e | s \rangle}{\langle p | s \rangle} \end{aligned} \quad (3.52)$$

so that the following orthogonality relations are satisfied

$$\langle a_m | a_n \rangle = \delta_{mn} \langle a_n | a_n \rangle \quad m, n = p, s, c_l, c_\perp \quad (3.53)$$

We determine the coefficients B_n (B_n^*) in (3.50) and the eigenvalues $z_\lambda^{(1)}$ by multiplying the second set of equations in (3.47) to the left (right) with $\langle a_m |$ ($| a_m \rangle$). This yields

$$\begin{aligned} \sum_n \langle a_m | c_l + z_\lambda^{(1)} | a_n \rangle B_n &= 0 \\ \sum_n \langle a_n | c_l + z_\lambda^{(1)} | a_m \rangle B_n^* &= 0 \end{aligned} \quad (3.54)$$

Here λ counts the different solutions of the system of equations (3.55). The coefficients in front of the vector B_n form the following matrices:

$$\begin{pmatrix} z_\lambda^{(1)} \langle p | p \rangle & 0 & \langle p | c_l^2 \rangle & 0 \\ 0 & z_\lambda^{(1)} \langle s | s \rangle & 0 & 0 \\ \langle c_l^2 | p \rangle & 0 & z_\lambda^{(1)} \langle c_l | c_l \rangle & 0 \\ 0 & 0 & 0 & z_\lambda^{(1)} \langle c_\perp | c_\perp \rangle \end{pmatrix}$$

The determinants of these matrices have to be equal zero. This yields $z_T^{(1)} = z_p^{(1)} = 0$ and

$$(z_\sigma^{(1)})^2 = \frac{1}{\langle c_l | c_l \rangle} \frac{\langle p | c_l^2 \rangle \langle c_l^2 | p \rangle}{\langle p | p \rangle} = c_0^2 \quad (3.55)$$

here T labels the heat mode, $\perp$ the transverse momentum or shear mode, and $\sigma = \pm 1$ the two sound modes.

Thus the eigenvalues to first order, $z_\lambda^{(1)}$, and the hydrodynamic modes to zeroth order, $\Psi_\lambda^{(0)}$, are

$$\begin{aligned} z_\sigma^{(1)} &= -\sigma c_0; & |\Psi_\sigma^{(0)}\rangle &= \frac{\langle p | p_k \rangle}{\langle p | p \rangle} |p\rangle + \sigma c_0 |c_l\rangle \\ z_\perp^{(1)} &= 0; & |\Psi_\perp^{(0)}\rangle &= |c_\perp\rangle \\ z_T^{(1)} &= 0; & |\Psi_T^{(0)}\rangle &= |s\rangle \end{aligned} \quad (3.56)$$

Using the general solution (3.49) we obtain the following expressions for the transport coefficients:

$$z_T^{(2)} = D_T = -\langle \Psi_T^{(0)} | c_l | \frac{1}{\Omega} + \frac{1}{2} | c_l \Psi_T^{(0)} \rangle / \langle \Psi_T^{(0)} | \Psi_T^{(0)} \rangle \quad (3.57)$$

$$z_\perp^{(2)} = \nu = -\langle c_\perp | c_l | \frac{1}{\Omega} + \frac{1}{2} | c_l c_\perp \rangle / \langle c_\perp | c_\perp \rangle \quad (3.58)$$

$$z_\sigma^{(2)} = \Gamma = -\langle \Psi_\sigma^{(0)} | (c_l - \sigma c_0) | \frac{1}{\Omega} + \frac{1}{2} | (c_l - \sigma c_0) \Psi_\sigma^{(0)} \rangle / \langle \Psi_\sigma^{(0)} | \Psi_\sigma^{(0)} \rangle \quad (3.59)$$

To simplify the expression for $\Gamma = z_\sigma^{(2)}$ we rewrite the corresponding current (3.47):

$$|j_\sigma\rangle = |j_T\rangle + \sigma c_0 |\tau_{xx}\rangle + |j'_\sigma\rangle \quad (3.60)$$

$$|j'_\sigma\rangle = |c_l(\alpha p - s - c_0^2)\rangle + \sigma c_0(|p_k\rangle - \alpha |p\rangle) \quad (3.61)$$

where $\alpha = \frac{\langle p | p_k \rangle}{\langle p | p \rangle}$ and $\tau_{xx} = \frac{1}{2}(c_l^2 - c_\perp^2)$.

Let us further define the so-called γ factor (the ratio of specific heats C_P/C_V)

$$\gamma \equiv \frac{1}{c_0^2} \frac{\langle s | s \rangle}{\langle c_l | c_l \rangle} + 1 \quad (3.62)$$

Then, on account of the following relations

$$\langle \Psi_\sigma^{(0)} | \Psi_\sigma^{(0)} \rangle = 2c_0^2 \langle c_l | c_l \rangle \quad (3.63)$$

$$\langle \Psi_T^{(0)} | \Psi_T^{(0)} \rangle = (\gamma - 1)c_0^2 \langle c_l | c_l \rangle$$

we obtain an expression for the sound damping constant in terms of viscosity $\nu = z_\perp^{(2)}$ and the heat diffusivity $D_T = z_T^{(2)}$,

$$\Gamma = \frac{1}{2} \{ (\gamma - 1) D_T + (\nu + \xi) \} \quad (3.64)$$

where the coefficient ξ that determines the bulk viscosity ζ , (3.34), is given by

$$\xi = -\frac{\langle j'_\sigma | \frac{1}{\Omega} + \frac{1}{2} | j'_\sigma \rangle}{c_0^2 \langle c_l | c_l \rangle} \quad (3.65)$$

We have used the fact that on a lattice with triangular symmetry a fourth rank tensor is isotropic (see Section 2.4.3)

$$\langle \tau_{xy} | \frac{1}{\Omega} + \frac{1}{2} | \tau_{xy} \rangle = \langle \tau_{xx} | \frac{1}{\Omega} + \frac{1}{2} | \tau_{xx} \rangle; \quad \tau_{xy} = j_{\perp} = c_l c_{\perp} \quad (3.66)$$

We note that our approach is consistent with the results for multi-speed models without potential energy when $e = p = p_k$. Specifically, the coefficient (3.65) vanishes in this limit and the γ -factor (3.62) is the ratio of the squares of the adiabatic to the isothermal speed of sound, (3.5) and (3.4).

Thus, we have derived the Green-Kubo expressions (3.57), (3.58) and (3.59) for transport coefficients in linear response theory and recovered the general relation (3.64) between transport coefficients for a model with non-zero bulk viscosity. In Section 4.2 we illustrate the applicability of this approach to determine the transport coefficients of an example of a thermal model.

Model Examples of the Generalized Approach

4.1 Introduction

Here we illustrate the ideas of the previous chapter with two model examples. In the first one, Section 4.2, we illustrate the use of statistics other than Fermi-Dirac. The analysis of transport coefficients in this first example is done using linear response theory (Section 3.6). In the second example, Section 4.3, we illustrate the ability of our approach to simulate nonequilibrium systems with spatially varying temperature with an application to Rayleigh Bénard convection.

Before introducing our first example, we make a few remarks as to the main criteria for constructing suitable models. Ideally we are looking for maximum simplicity combined with basic features of thermal hydrodynamics¹. Starting from an athermal FHP model, we can build in the thermal properties in three ways:

1. adding more allowed velocities; this is the “classical” way, and the resulting models are multi-speed thermal LGCAs, discussed in Section 2.3,
2. keep a single velocity ($c = 1$), but add (potential) energy levels for each particle,
3. build a model with both variable kinetic and potential energy.

Because there is a potential energy level structure, it is natural to model the time evolution by Monte-Carlo algorithms (Sections 3.2.3 and 3.2.4), and this is our approach for simulating fluids in non-equilibrium.

¹Spatial temperature variation, the temperature dependence of transport coefficients, non-zero (but small) bulk viscosity, are examples of those properties of realistic thermal fluids that are important in many applications and yet are not modeled by “conventional” LGCA models.

The second way is the simplest. However, the transport coefficients (viscosity, sound speed, etc) are not temperature dependent. Indeed, the particles' speeds are constant $c_I = c$, which leads to the following expressions for the isothermal and adiabatic speeds of sound (3.4), (3.5) and the shear and bulk viscosities (3.33), (3.34)

$$c_T^2 = c_s^2 = \frac{1}{2}c^2; \quad \nu = \frac{1}{4}c^2(\tau - \frac{1}{2}); \quad \xi = 0 \quad (4.1)$$

The thermal conduction $\lambda = (\tau - \frac{1}{2})\frac{1}{2}c^2\langle(\Delta E)^2\rangle$ depends on the statistics of occupation. In Chapter 5 we will use a model with a single speed and two potential energy levels as a basis to build the interactions.

The third way promises simplicity if we have two speeds: regular $c = 1$ plus rest particles $c = 0$. This is our choice for the examples in Sections 4.2 and 4.3.1. Whenever we have rest particles in the model, we expand the collision table of the FHP model by at least one type of collision which involves a rest particle (and conserves momentum, of course). The most obvious choice for such a collision is shown in Figure 4.1.

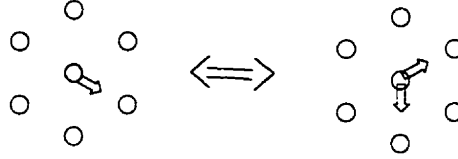


Figure 4.1: Typical collision involving a rest particle (located at the site shown as a filled circle)

4.2 A 13-bit Model with Three Energy Levels

4.2.1 The Model

We define an FHP model (Section 2.2) with rest particles and with a potential energy structure shown in Figure 4.2. The moving particles can be of two types, which are characterized by two distinct *potential* energies, E_A and E_B . We take the zero energy of the rest particles as reference (and refer to it as $E_G = 0$), and choose the two energy levels of the moving particles to be $E_A = \epsilon$ and $E_B = 2\epsilon$ respectively.

Thus, we have a 13-bit model with 12 distinct states for the moving particles and 1 state for rest particles. However, we choose to allow only one moving particle, either A or B , to be present in any one velocity channel at any one time, which implies

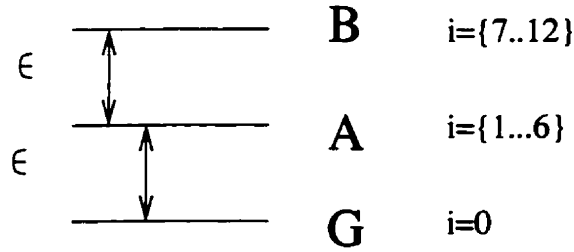


Figure 4.2: Energy level structure in each particle. Ground level G is populated by rest particles only. Moving particles can populate either A or B levels.

that the occupancy of level A for a given bond i *prevents* the occupancy of this bond by a particle of type B and vice versa. Hence, for the system in equilibrium, the probability of finding the particle of energy E_I ; $I = G, A, B$, which we shall denote by f_I , does not take the form of the Fermi-Dirac distribution.

To obtain the equilibrium distribution we will enumerate the number of ways such that energy levels G , A and B (Figure 4.2) are populated by N_G , N_A , N_B particles respectively. Let W_{AB} be the number of ways the particles can populate either level A or B and W_G be the number of ways to have N_G particles in level G

$$W_{AB} = \frac{N!}{N_A!N_B!(N - N_A - N_B)!} \quad (4.2)$$

$$W_G = \frac{N!}{N_G!(N - N_G)!} \quad (4.3)$$

Here $N = N_G + N_A + N_B$ is the total number of particles. The entropy of the system $S = k \ln(W_{AB}W_G)$ is maximum when

$$\frac{\partial S}{\partial N_I} - \alpha - \beta E_I = 0 \quad I = G, A, B \quad E_I = 0, \epsilon, 2\epsilon \quad (4.4)$$

where the Lagrange multipliers α and β have the physical meaning of chemical potential and inverse temperature respectively. Using Stirling's approximation we obtain

$$f_A = \langle N_A \rangle = \frac{zx}{1 + zx(1 + x)} \quad (4.5)$$

$$f_B = \langle N_B \rangle = \frac{zx^2}{1 + zx(1 + x)} \quad (4.6)$$

$$f_G = \langle N_G \rangle = \frac{z}{1 + z} \quad (4.7)$$

where z is $e^{-\alpha}$ and x is $e^{-\beta\epsilon}$. Differentiating (4.7) with respect to the chemical potential α and inverse temperature β gives the equal-time correlation functions for

the density fluctuations and energy fluctuations respectively¹

$$k_i^\alpha = \begin{cases} f_G(1 - f_G) & i = 0; \\ f_A(1 - f_H) & i = 1, \dots, 6; \\ f_B(1 - f_H) & i = 7, \dots, 12. \end{cases} \quad (4.8)$$

$$k_i^\beta = \begin{cases} 0 & i = 0; \\ f_A \bar{f}_H - f_A f_B & i = 1, \dots, 6; \\ 2f_B \bar{f}_H + f_A f_B & i = 7, \dots, 12. \end{cases} \quad (4.9)$$

with $f_H = f_A + f_B$ and $\bar{f}_H = 1 - f_A - f_B$.

4.2.2 Collision Matrix

We define the collision rules to be of FHP type. Thus, we have binary collisions (Figure 2.2(a)) and three-particle collisions (Figure 2.2(b)) for particles in levels A and B plus the collisions involving rest particles (Figure 4.1). Within the Monte-Carlo type evolution (Section 3.2.3) the energy need not be conserved during collisions. This allows, in principle, a much greater number of collisions than in a conventional FHP model. For example, a two-particle output state has $2^2 = 4$ equally probable distinct versions, and a three particle configuration has $2^3 = 8$ versions.

To analyze the model within linear response theory (Ernst [90], also see Section 3.6) we need the linearized collision matrix $\Omega^{(1)}$ which is defined through (2.23). Rather than giving the complete derivation of $\Omega^{(1)}$, we will show some essential steps of the linearization using the example of the collisions involving the rest particle.

The rate $\dot{f}_a^I$ ($I = G, A, B$ and $a = 1 \dots 6$) of the change of the occupation function of G-, A- and B-levels within the Monte-Carlo evolution (Section 3.2.3) defines the first, second-to-seventh and eighth-to-thirteenth rows of the $\Omega^{(1)}$ matrix respectively. We consider the $\{1, 0\} \rightarrow \{2, 6\}$ process² shown in Figure 4.1. For the changes in occupation of direction 1 we have the following expressions

$$\begin{aligned} \dot{f}_1^A = \frac{1}{4} \{ & -f^G f_1^A \bar{f}_2 \bar{f}_6 x + f_2^A f_6^A \bar{f}_0 \bar{f}_1 \\ & -f^G f_1^A \bar{f}_2 \bar{f}_6 x^2 + f_2^A f_6^B \bar{f}_0 \bar{f}_1 \end{aligned}$$

¹These are the same quantities f_i^μ and f_i^T that enter equations (3.5) and (3.41) respectively.

²The number 0 labels the state of rest particle, the numbers 1...6 label six velocity directions.

$$\begin{aligned}
& -f^G f_1^A \bar{f}_2 \bar{f}_6 x^2 + f_2^B f_6^A \bar{f}_0 \bar{f}_1 \\
& -f^G f_1^A \bar{f}_2 \bar{f}_6 x^3 + f_2^B f_6^B \bar{f}_0 \bar{f}_1 \} (f_A)^2 \bar{f}_G \bar{f}_H
\end{aligned} \tag{4.10}$$

$$\begin{aligned}
f_1^B = \frac{1}{4} \{ & -f^G f_1^B \bar{f}_2 \bar{f}_6 + f_2^A f_6^A \bar{f}_0 \bar{f}_1 \\
& -f^G f_1^B \bar{f}_2 \bar{f}_6 x + f_2^A f_6^B \bar{f}_0 \bar{f}_1 \\
& -f^G f_1^B \bar{f}_2 \bar{f}_6 x + f_2^B f_6^A \bar{f}_0 \bar{f}_1 \\
& -f^G f_1^B \bar{f}_2 \bar{f}_6 x^2 + f_2^B f_6^B \bar{f}_0 \bar{f}_1 \} (f_A)^2 \bar{f}_G \bar{f}_H
\end{aligned} \tag{4.11}$$

We use the notations $\bar{f}_0 = 1 - f^G$, $\bar{f}_a = 1 - f_a^A - f_a^B$, $a = 1, 2, \dots, 6$, $\bar{f}_H = 1 - f_A - f_B$. The Boltzmann factors x , as defined in (4.7), as well as the identities

$$x f_G \bar{f}_H = f_A \bar{f}_G \quad f_B = x f_A \tag{4.12}$$

ensure the zero-order cancellation for f_1^A or the detailed balance condition for the model. Note, that the first line of (4.11) is the contribution from energy-conserving process.

Linearization gives the following expressions for the second and eighth rows of $\Omega^{(1)}$

$$\begin{aligned}
\Omega_{1i}^{(1)} = K \quad & \{ -(\frac{1}{f_G} + \frac{1}{f_G}), \\
& -(\frac{1}{f_A} + \frac{1}{f_H}), \quad (\frac{1}{f_A} x_1 + \frac{1}{f_H}), \quad 0, \quad 0, \quad 0, \quad (\frac{1}{f_A} x_1 + \frac{1}{f_H}), \\
& -(\frac{1}{f_H}), \quad (\frac{1}{f_B} x_2 + \frac{1}{f_H}), \quad 0, \quad 0, \quad 0, \quad (\frac{1}{f_B} x_2 + \frac{1}{f_H}) \} \\
\Omega_{7i}^{(1)} = K \quad & \{ -(\frac{1}{f_G} + \frac{1}{f_G}), \\
& -(\frac{1}{f_H}), \quad (\frac{1}{f_A} x_1 + \frac{1}{f_H}), \quad 0, \quad 0, \quad 0, \quad (\frac{1}{f_A} x_1 + \frac{1}{f_H}), \\
& -(\frac{1}{f_B} + \frac{1}{f_H}), \quad (\frac{1}{f_B} x_2 + \frac{1}{f_H}), \quad 0, \quad 0, \quad 0, \quad (\frac{1}{f_B} x_2 + \frac{1}{f_H}) \} \\
x_1 = \frac{1+x}{1+2x+x^2}; \quad & x_2 = x x_1; \quad K = (f_A)^2 \bar{f}_G \bar{f}_H (1+2x+x^2)
\end{aligned} \tag{4.13}$$

Other matrix elements follow from the symmetry relations:

$$\begin{aligned}
\Omega_{i=\{2,\dots,6\} j=0} &= \Omega_{i=1 j=0} & \Omega_{i=\{8,\dots,12\} j=0} &= \Omega_{i=7 j=0} \\
\Omega_{i=\{2,\dots,6\} j=\{1,\dots,6\}} &= \text{circ}\{\Omega_{i=1 j=\{1,\dots,6\}}\} \\
\Omega_{i=\{8,\dots,12\} j=\{1,\dots,6\}} &= \text{circ}\{\Omega_{i=7 j=\{1,\dots,6\}}\} \\
\Omega_{i=\{2,\dots,6\} j=\{7,\dots,12\}} &= \text{circ}\{\Omega_{i=1 j=\{7,\dots,12\}}\} \\
\Omega_{i=\{8,\dots,12\} j=\{7,\dots,12\}} &= \text{circ}\{\Omega_{i=7 j=\{7,\dots,12\}}\} \\
\Omega_{0i} &= \sum_{j=1}^{12} \Omega_{ij}
\end{aligned} \tag{4.14}$$

where *circ* designates a circulant matrix¹. The full matrix of basic set of collisions is obtained by adding the other processes: $\{2, 6\} \rightarrow \{1, 0\}$, $\{1, 4\} \rightarrow \{2, 5\}$, $\{1, 4\} \rightarrow \{3, 6\}$, $\{1, 3, 5\} \rightarrow \{2, 4, 6\}$ and their rotational variations.

Conservation of the number of particles and momentum implies the following relations for the matrix elements

$$\sum_{i=0}^{12} \Omega_{ij} = 0 \quad \sum_{i=0}^{12} \Omega_{ij} c_{ix} = 0 \quad (4.15)$$

As mentioned earlier, within the Monte-Carlo scheme, energy is not conserved when the system is in the canonical ensemble (Section 3.2.3) or with the many-demons algorithm (Section 3.2.4). Because all updates occur according to a set constant temperature in the first case, or because the demons serve to maintain a uniform temperature in the second case, we call these models “isothermal”. It is also useful to consider the model with the subset of collisions that strictly conserve energy. For example, in the $\{1, 0\} \rightarrow \{2, 6\}$ process, considered above, we distinguish a situation when a moving particle in B level collides with a rest particle resulting in two particles in level A. The energy before the collision, $E_B + E_G$, is equal to the energy after the collision, $E_A + E_A$. For this “adiabatic” model, the linearized collision matrix satisfies the condition

$$\sum_{i=0}^{12} \Omega_{ij} E_i = 0 \quad (4.16)$$

4.2.3 Transport Coefficients

The important physical characteristics of the model are contained in its transport properties. In turn, these determine the utility of the model for the study of non-equilibrium processes. We have chosen to obtain the transport coefficients here by analyzing the hydrodynamic dispersion relations for spatial fluctuations in the occupation of single-particle states, which are defined through the linearized collision matrix $\Omega^{(1)}$, obtained in Section 4.2.2, and eigenvalue problem (2.45), Section 3.6. We also use the Green-Kubo expression, (3.58), for the shear viscosity.

¹In the circular matrix each row, except the first one, is derived from the the previous row by the procedure of shifting the location of each matrix element one column to the right with last element taking its place in the first column.

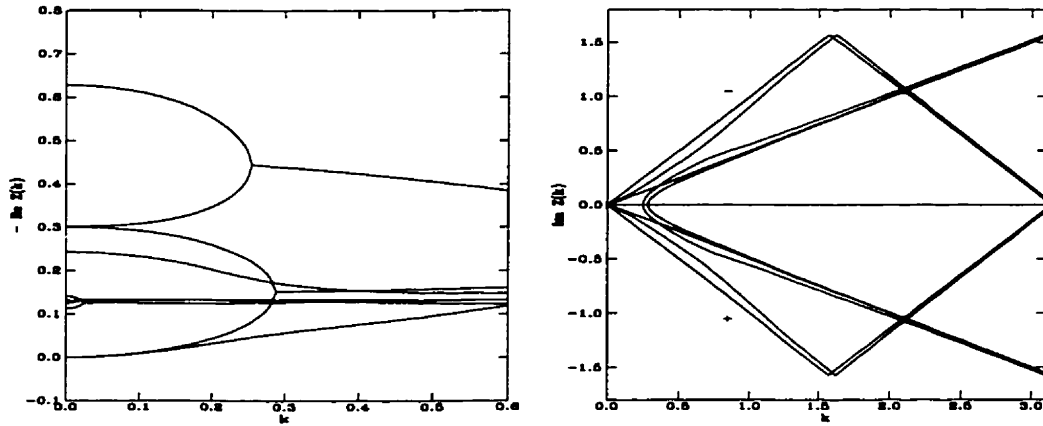


Figure 4.3: Real (left) and imaginary (right) parts of the solution of (2.45) for isothermal 13 bit model.

Figure 4.3 shows the real and imaginary parts of the solution of the eigen-value problem (2.45). The hydrodynamic modes z_λ , that determine transport coefficients, have zero eigenvalues at $k = 0$ (Section 2.3.6). We evaluated the transport coefficients from the expansions (2.48). Figure 4.4 shows $\text{Re}(z_\perp(\mathbf{k}))/k^2$ for $\mathbf{k} \parallel \hat{x}$ and $k \rightarrow$

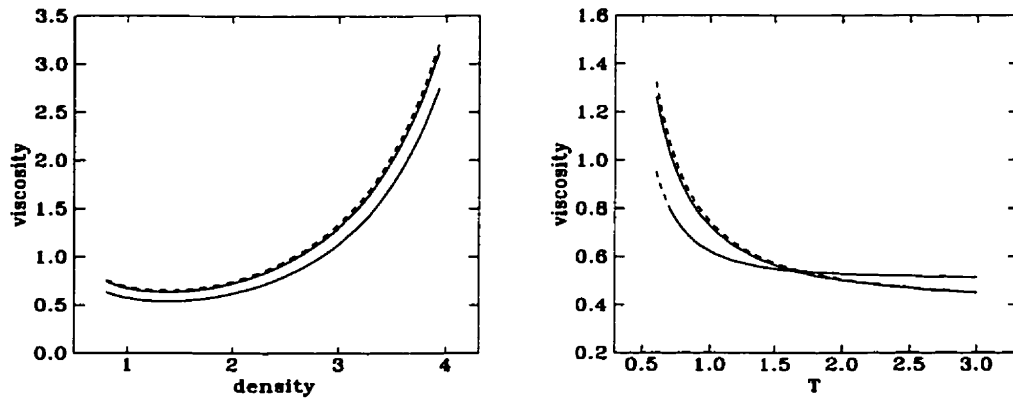


Figure 4.4: Left: Shear viscosity dependence on density (in units of particle per site) for $T=\epsilon$. Right: shear viscosity dependence on temperature (in units of ϵ) for $\rho = 2$. Upper curves at low temperatures, adiabatic model; lower curves, isothermal model.

0 as a function of density and temperature respectively. Calculations were done for both the adiabatic and isothermal models (the solid lines) and compared to the direct evaluation of the Green-Kubo expression for the shear viscosity (3.58) (the dashed lines). Both methods of calculation give identical results. For the adiabatic model, we also computed $\text{Re}(z_T(\mathbf{k}))$, and derived the thermal diffusion coefficient. Its

dependence on density and temperature are shown in Figure 4.5.

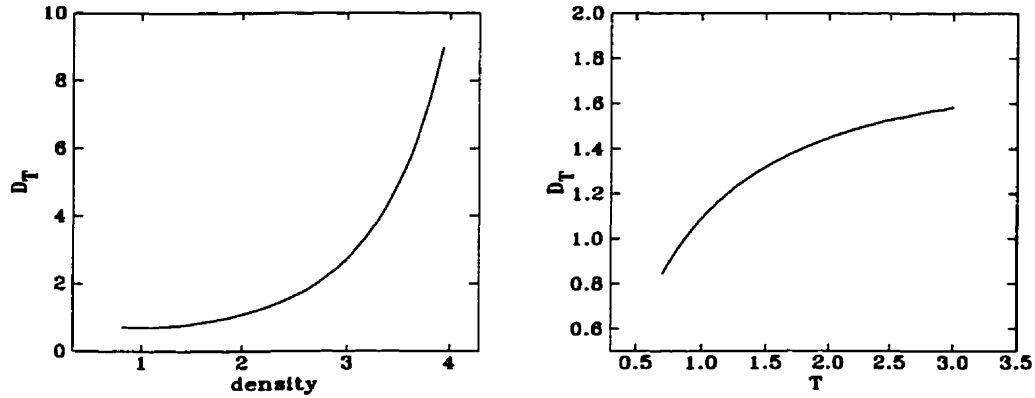


Figure 4.5: Left: thermal diffusion dependence on density for $T=\epsilon$. Right: thermal diffusion dependence on temperature (in units of ϵ) for $\rho = 2$.

4.2.4 Conclusion

This thermal lattice gas model has a number of interesting features. It avoids the complexity of existing energy-conserving models by introducing two possible energy states for each moving particle. This permits us to employ collision rules which are similar to those of the original FHP model, so that analysis becomes more transparent. In principle, this also leads to considerable computational efficiency. More important, however, is that the model has well-defined transport coefficients over a range of temperatures and densities. When the evolution of the model is governed by the multi-demon algorithm (Section 3.2.4), the model is potentially useful in non-equilibrium situations where, for example, the local temperature is a dynamically varying parameter, not fixed as in a conventional lattice gas simulation. We will test this idea on an example of a model with similar features in our next example, Section 4.3.

4.3 Rayleigh Bénard Convection

In this section we illustrate the applicability of GTLG to simulate a non-equilibrium phenomenon, such as Rayleigh Bénard convection. When the fluid is subjected to both a temperature gradient and to gravity, as shown in Figure 4.6, the convective

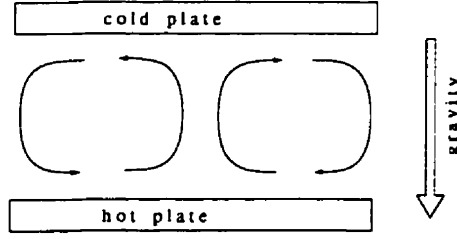


Figure 4.6: The Rayleigh Bénard convection

instability known as Rayleigh Bénard convection can occur (Chandrashekar [81]). The temperature distribution in the fluid is the major factor in the evolution of this instability. Thus, simulation is possible using our GTLG models with the multi-demon algorithm.

4.3.1 A Model

The model we build for our simulations of Rayleigh Bénard convection has many features of the 13-bit model studied in Section 4.2. However, it is advantageous to have some new features for two reasons:

1. To enhance the “tunability” of the model, and hence, to expand the parameter space so that the instability can be more easily detected. An example here is the change to even less restrictive Maxwell-Boltzmann statistics, so that the density can be varied over a greater range.
2. To further exploit the various possibilities of the GTLG approach: here, the degeneracy of energy levels.

Thus, we choose to employ Maxwell-Boltzmann statistics for a simple model with 3 energy levels. The lowest level, with energy $E_0 = 0$, is M -fold degenerate, so that there are at most M fictitious particles with zero velocity (“rest particles”). The other two levels both correspond to the same speed c , with the same 6-fold symmetry as in the FHP models, but their energies are respectively E_A and $E_B = E_A + \Delta$. In our numerical calculations, we take $E_A = 0.6$ and $E_B = 1.2$ in units of c^2 , and $M = 2$.

Assuming Maxwell-Boltzmann statistics, we obtain (3.13)

$$f_0(\mathbf{u}) = e^{\beta\mu}; \quad f_{I,a}(\mathbf{u}) = e^{\beta(\mu - E_I)}(1 + q\rho\mathbf{u} \cdot \mathbf{c}_a), \quad I = A, B \quad (4.17)$$

where $q = \rho e^{-\beta\mu}/3c^2(e^{-\beta E_A} + e^{-\beta E_B})$, and $\rho e^{-\beta\mu} = [M + 6(e^{-\beta E_A} + e^{-\beta E_B})]$. This simple form for the velocity expansion results because $f_I^\mu = f_I^{eq}$. The densities ρ and ρ_0 are defined in (3.2).

Because there is only one speed in the model, the expressions for the sound velocities and for the shear and bulk viscosities again become particularly simple. From (3.4) and (3.5) we obtain

$$\begin{aligned} c_T^2 &= \frac{1}{2}c^2 \frac{\sum_{A,B} \rho_I}{\rho} = 3c^2 \left[\frac{e^{-\beta E_A} + e^{-\beta E_B}}{M + 6(e^{-\beta E_A} + e^{-\beta E_B})} \right]; \\ c_s^2 &= \frac{1}{2}c^2 \end{aligned} \quad (4.18)$$

and

$$\begin{aligned} \nu &= (\tau - \frac{1}{2})c^2/4; \\ \xi &= (\tau - \frac{1}{2})\frac{c^2}{2}\frac{\rho_0}{\rho} \\ &= (\tau - \frac{1}{2})\frac{c^2}{2}\frac{M}{M + 6(e^{-\beta E_A} + e^{-\beta E_B})} \end{aligned} \quad (4.19)$$

The temperature-independence of ν is an artifact of the model, but the temperature dependence of the coefficient ξ , (3.34), is typical of the viscosity of a liquid. For a typical value of the density, ξ is displayed in Figure 4.7 as a function of temperature.

The model also leads to a simple expression for the thermal conduction coefficient λ :

$$\lambda = (\tau - \frac{1}{2})\frac{3c^2\Delta^2}{T^2}\frac{f_A^{eq}f_B^{eq}}{f_A^{eq} + f_B^{eq}}$$

For the purpose of illustration, it is convenient to relate this to the thermal diffusion coefficient, defined as $D_T = \lambda/\rho C_p$, which is also plotted in Figure 4.7. Although the temperature dependence of λ is dominated by the $\frac{1}{T^2}$ prefactor at high temperatures, $T > \Delta$, this behavior is exactly compensated by the temperature dependence of the specific heat.

For completeness, Figure 4.7 also shows the temperature variation of the sound attenuation coefficient Γ , defined in the standard way, (3.64).

4.3.2 Numerical Results

With the model defined above, we carried out simulations of Rayleigh-Bénard convection (Figure 4.6). We used the Lattice-Boltzmann method (Section 2.5) with an

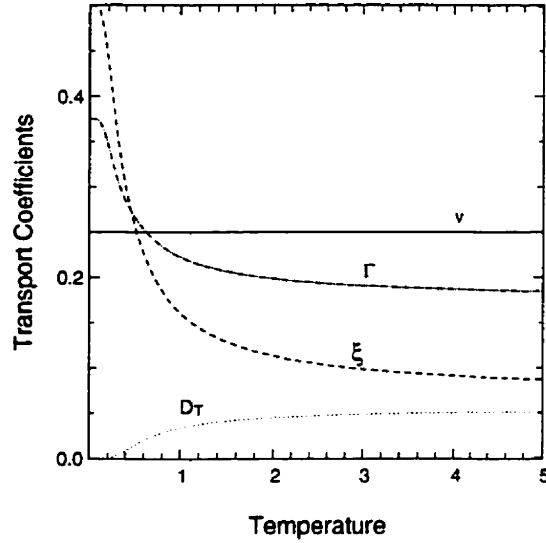


Figure 4.7: Temperature variation of the transport coefficients for the model described in the text with density $\rho = 3.6$. Solid line: bulk viscosity. Dashed line: shear viscosity. Dotted line: thermal diffusion. Dash-dotted line: sound attenuation. All coefficients are in units of $\tau - \frac{1}{2}$.

equilibrium distribution given by (4.17). Since the central factor in this experiment is the temperature field, the relevant algorithm of simulation is again the multi-demon algorithm (Section 3.2.4). The algorithm was adapted for this purpose, so that relaxation occurred towards a local distribution function defined by an instantaneous average local temperature.

We considered a two dimensional cell with horizontal length L and vertical height H . Periodic boundary conditions were imposed in the horizontal direction, with two rigid walls at the top and the bottom of the cell. The demon energies on the upper and lower walls were fixed at values T_1 and T_2 , respectively. The force of gravity was implemented by changing the vertical momentum of the particles at a constant rate, while keeping the horizontal momentum unchanged. Thus, both the temperature difference δT and the force could be tuned continuously. We averaged velocity fields over 10×10 regions in order to evaluate the local distribution functions.

We studied a system with $L = 400$ lattice units and $H = 100 \times \sqrt{3}$ units, so that the aspect ratio $L : H$ was near $2 : 1$, with a particle density of 3.6 per site. The energy levels were $E_A = 0.6$, $E_B = 1.2$ in units of c^2 , and the temperatures at the lower and upper boundaries were $T_1 = 6.0$, $T_2 = 0.8$ in the same units. Gravity was

set to 0.002 in units of “unit of velocity per unit of time”. We chose the relaxation time τ to be 1 unit.

Initial conditions were a uniform density and a uniform temperature gradient, and with the local velocity everywhere zero, so that we were able to observe the onset and subsequent evolution of the convective instability. With these parameters, the evolution of the system was sufficiently slow that “snapshots” of the velocity field could be obtained by time averages over as many as 50 time-steps.

Figure 4.8 shows the evolution of the temperature distribution, and of the corresponding velocity fields for these parameters. At first four convection rolls are clearly seen, but subsequently these collapse into two. Evidently, the model has captured the essential features of the physical phenomenon.

According to the classic linear stability analysis, (Chandrashekar [81]), the critical parameter in Rayleigh Bénard convection is so-called Rayleigh number

$$\mathcal{R} = \frac{\alpha_P g H^3 \delta T \rho C_v}{\nu \lambda} \quad (4.20)$$

where α_P is the thermal expansion coefficient, g is the gravitational field, H is the height of the cell, and δT is the temperature difference between the two boundaries. The convection occurs when the Rayleigh number exceeds a critical value of order 10^3 .

In our experiment we observe the difference of density of order 0.2 between hot and cold part of fluid. Thus, in “rough” approximation we have $\alpha_P = \Delta\rho/\Delta T \simeq 0.04$. This gives the estimation of Rayleigh number $\mathcal{R} \simeq 10^5$ which exceeds the critical value. Even though we are far from the linear regime, the value of $\mathcal{R}$ is a good indicator of the stability of the convective phenomenon. Thus, for the simulation shown in the figure, the convection rolls should be extremely stable, as is indeed the case. Indeed, our results are strikingly similar to those of a previous detailed study (Shan [97]), obtained with a modified LB model which represented the temperature as a “convected passive scalar field”.

An interesting feature of our results is the observation of four rolls before the final stable state with two rolls is reached. Linear analysis predicts two rolls (since the wavenumber of the instability should be around π/H where H is the height of the cell). Our tentative explanation is that the velocity field is first established near the

lower (hot) boundary, so that the effective height of the cell is considerably smaller than H . We therefore expect the wavenumber of the instability to be larger than π/H , and the periodic boundary conditions select $\sim 2\pi/H$. (In principle, with different geometry, we might expect to see yet higher initial wavenumbers.) The study of this transient regime, and the subsequent stabilization of the two rolls, would be an interesting topic for further study.

However, in general terms, this illustration of Rayleigh-Bénard convection demonstrates the feasibility of our Lattice Boltzmann method for non-equilibrium thermal lattice-gases. Our approach can be applied to a variety of phenomena for which the statistical mechanics of the gas are critically important.

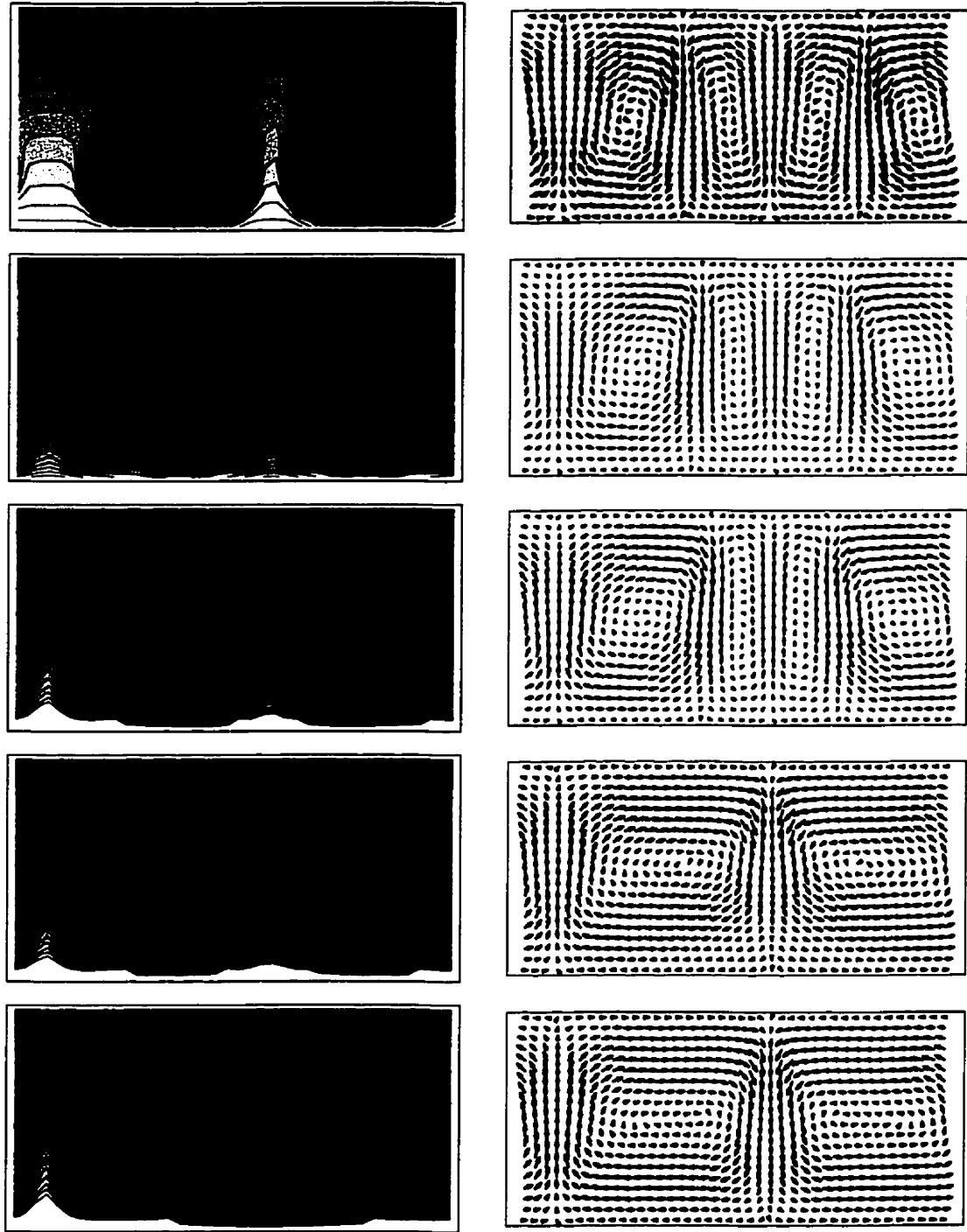


Figure 4.8: Contour plots showing the time evolution of the temperature distribution (left panels) and of the velocity field (right panels). From top to bottom, data is shown at times 90,000, 180,000, 270,000, 360,000 and 450,000 units respectively. One time unit is defined as one update of every site in the lattice. In each panel, the hot boundary is at the bottom.

Thermohydrodynamic Model with Interactions

5.1 Introduction

Motivated by the possibility of constructing thermodynamically correct models of thermal hydrodynamics for the simulation of out-of-equilibrium phenomena in complex fluids (Section 1.2) we have already demonstrated significant progress. Indeed, the generalized lattice gas approach, introduced in Chapter 3, allows us to construct models where the correct thermodynamic temperature controls (and is part of) the evolution of a fluid, as illustrated for Rayleigh-Bénard convection in Section 4.3. On the other hand, we cannot yet use our approach to simulate more interesting phenomena involving phase-separating fluids. However, our approach turns out to have one more important advantage - the ability to include the interaction between particles in a straightforward way consistent with correct thermodynamics and hydrodynamics. This will be the main focus of this chapter.

There has been continuous interest in lattice gas models of hydrodynamics with interactions ever since LGA were proposed. Modified LGCA and LB models with interactions have been described by various authors:

Rothman and Keller [88] have introduced the *immiscible lattice gas* (ILG) model. In their model, two types of particles, labeled as “red” and “blue”, undergo FHP type collisions and propagation, but with a modification in the collision step: the collision rule changes the configuration of particles so that, as much as possible, red particles are directed towards sites with majority red particles, and blue particles are directed towards sites with majority blue particles. The model is effective in simulating the phase separation of a binary mixture. Indeed, if the system is initialized as a random mixture of blue and red particles, then it evolves towards distinct domains of one

color, similar to water-oil mixtures, for example. However, ILG are not suitable for modeling thermal phenomena: there is no distribution of particles among energy levels to define a temperature.

Further, the fundamental problem of this and other LGCA models with interaction concerns the non-existence of a correct thermodynamic equilibrium state and, hence, the non-validity of statistical mechanics. For ILG models this happens because they violate the semi-detailed balance condition (2.19).

Despite this criticism, the model and its derivatives (Chen et al. [91b]; Boghosian, Coveney and Emerton [96]; Maillet and Coveney [00]) turn out to be useful in studying the kinetics of some hydrodynamic systems (Emerton, Coveney and Boghosian [97]; Weig, Coveney and Boghosian [97]; Maillet and Coveney [00])

Similar problems with semi-detailed balance occur with other approaches. Appert and Zaleski [90]; Gerits, Ernst and Frenkel [93], for example, have added one extra collision step to an FHP-type model. Pairs of particles, a fixed distance apart (not one lattice unit), reverse their momenta if the Fermi-Dirac statistics so allows. Unlike the ILG, phase separation in this model manifests itself not as the segregation of two species of particles into separate regions, but as the separation of a single species of particle into regions of high and low density.

The Lattice Boltzmann (LB) method (Section 2.5) underwent a rather different evolution to include interactions. Gunstensen et al. [91], for instance, adapted the ILG model of Rothman and Keller [88] to the Lattice Boltzmann scheme. Using the equilibrium distribution for both blue and red matter as in equation (2.53) they modified the Boltzmann operator for situations determined by the maximum of the color gradient. The violation of the semi-detailed balance condition and consequent inconsistent thermodynamics have then exactly the same basis as in particle-based ILG.

In another approach, Shan and Chen [93] redefined the equilibrium velocity distribution so as to simulate a fluid with a non-ideal equation of state and an arbitrary number of phases or components. The short-range interparticle potential introduced in this model adds an attractive or repulsive element to collisions. Although this model and its modifications (Shan and Chen [94]; Shan [97]) involve the local non-

conservation of momentum, and, again, inconsistent thermodynamics (in references Shan and Chen [94]; Shan [97], for example, authors talk about the kinetic properties of the model as a function of temperature-like parameter) yet the interactions ensure the correct interfacial properties.

The problem of thermodynamic inconsistency in LG models of hydrodynamics with interaction was resolved for the first time by Swift, Osborn and Yeomans [95]; Orlandini, Swift and Yeomans [95]; Swift et al. [96]. They showed that it is indeed possible to set up a Lattice Boltzmann method modeling isothermal hydrodynamics for two-phase systems. They introduced into the collision operator the equilibrium pressure tensor for a non-ideal fluid by adding additional term $G_{\alpha\beta}c_{i\alpha}c_{i\beta}$ to the general solution for the distribution function (2.53). This term requires an additional constraint for the pressure tensor to be taken into consideration

$$\sum_i f_i^{(0)} c_{i\alpha} c_{i\beta} = P_{\alpha\beta} + \rho u_\alpha u_\beta \quad (5.1)$$

where the pressure tensor $P_{\alpha\beta}$ is set to correspond to the equilibrium pressure tensor for the fluid and can be associated with a free energy functional. Thus an appropriate choice of the pressure tensor (or free energy functional) can lead to a model for simulating phase-transitions. This approach have been successfully applied to thermal phenomena in non-equilibrium (Gonella, Orlandini and Yeomans [97]; Wagner and Yeomans [98]).

As we see, LB methods for non-ideal fluids have made considerable progress. However, because of their nature, LB methods cannot be used in situations where fluctuations play an important role (although Ladd [94] has introduced thermal fluctuations as Gaussian noise in the lattice Boltzmann equation for non-interacting models, and Stelitano and Rothman [00] have used a Gaussian noise term in more complex simulations of phase separations). However, *the above mentioned thermodynamically consistent LB method cannot model the temperature as a local variable dependent on boundary conditions, as, for example, in our model for Rayleigh-Bénard convection in Section 4.3.*

In this chapter we introduce a new approach for the construction of interacting thermal LG models that has the advantages of particle-based models and is explicitly consistent with thermodynamics. In previous chapters we have shown that proper

thermal hydrodynamic behavior remains valid when the collision step is performed in the manner of a Monte-Carlo simulation. For that to be possible, we allowed each particle to have its own “potential” energy structure. We will show, below, that such models possess all the necessary conditions for including short-range interactions in a way that is entirely transparent.

5.2 Model of Ising Spins with 1-st Order Phase Transition

The compatibility of our approach with statistical-mechanical Monte-Carlo methods makes it easy to adopt features of the simplest interacting lattice model - the Ising model - to our hydrodynamically correct model. To validate the later choice of interaction we review here a modified Ising model that simulates a 1-st order phase transition. This model has been discussed extensively by Harris, Jörgenson and Grant [92] both in the context of equilibrium and nonequilibrium problems. It has been shown to reliably model thermal diffusion processes (Jörgenson, Harris and Grant [89]; Jörgenson and Harris [92]), steady states (Harris and Grant [88]) and thermal instabilities (Harris, Jörgenson and Grant [92]; Jörgenson et al. [93]).

The $N = V$ spins $\sigma_i = \pm 1, i = 1..V$ are located on V sites of some lattice with coordination number z , one spin at a site. The Hamiltonian, closely related to the Ising Hamiltonian, is

$$H = -\frac{J}{2} \sum_i \sum_{j=\langle ij \rangle} \sigma_i \sigma_j - \Delta \sum_i \sigma_i \quad (5.2)$$

where J is the spin coupling, the interactions are with the nearest neighbors and Δ represents a uniform external field. In the absence of Δ the second-order phase transition is at some temperature T_c (for our triangular lattice $T_c = 3.6408J$, Lavis and Bell [99], p.211). In order to induce a first-order transition (and a stable region of phase coexistence) the state $\sigma = -1$ is given the degeneracy $\delta > 1$. For the particular case $\delta = 2$ the model becomes a special case of the Blume-Emery-Griffiths Hamiltonian (Blume, Emery and Griffiths [71]).

Harris [85] has shown that the system, described by Hamiltonian (5.2) with a degenerate upper state displays a first-order phase transition at $T_m < T_c$ with latent heat of transition Λ of order 2Δ ; the melting temperature is

$$T_m = 2\Delta / \ln(\delta) \quad (5.3)$$

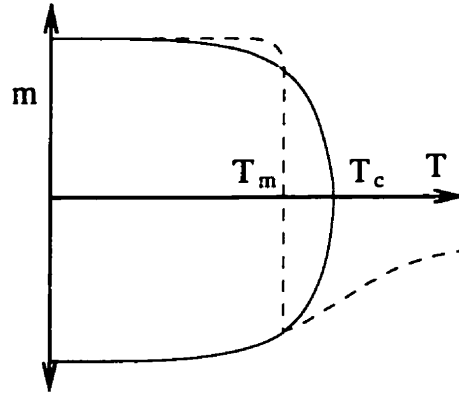


Figure 5.1: Schematic phase diagram of the modified Ising model in the temperature-magnetization plane, adapted from Harris, Jörgenson and Grant [92]. The solid line is the plot of the solution of an Ising model with a second-order phase-transition at T_c . The dashed line is a solution of the modified Ising model that exhibit a first-order phase transition at $T_m < T_c$.

Figure 5.1 illustrates the situation by superposing the plot of m via T on the phase diagram of the Ising model.

5.3 Defining the Hydrodynamic Model with Latent Heat

The next step in building the hydrodynamic model of an interacting fluid is to combine the Ising-like interaction (Section 5.2) with our generalized lattice gas approach for thermal hydrodynamics (Chapter 3). Thus, we set the basic frame-work for our model as follows:

- i. A two-dimensional hexagonal lattice with six ($z=6$) nearest neighbors for each site.
- ii. Particles occupy the sites of lattice and have only one speed $|c| = 1$. The direction of the velocity can take six values in compliance with the directions to neighboring sites.
- iii. The particles are distinguishable and obey Maxwell-Boltzmann statistics (Pathria [78], pp.143-144) with no restrictions as to multiple occupancy of different single-particle states.
- iv. Each particle has two potential energy states, separated by 2Δ in energy units. We label the lower energy level as a $\sigma = +1$ spin state and the higher energy level as a $\sigma = -1$ spin state (see Figure 5.2).

- v. Each of the energy levels can be degenerate: we set $\delta = 2$ to be the degeneracy of the $\sigma = -1$ level in the spirit of the model discussed in Section 5.2.

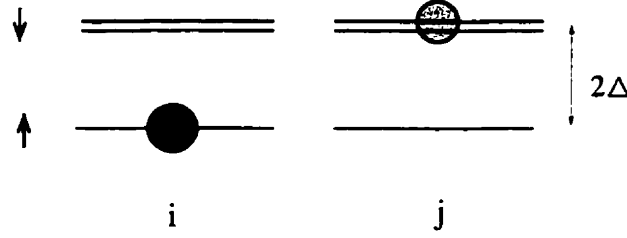


Figure 5.2: The two-energy level structure for each particle. Left: the particle located at site i in its lower energy state (spin $\sigma_{ik} = +1$); right: the particle is located at site j in one of two higher energy states (spin $\sigma_{ik} = -1$). The velocity attributes are not shown.

The choices [i] and [ii] are obvious in terms of a minimal, isotropic model of hydrodynamics (Section 2.3). Choice [iii] is natural for modeling interacting fluids. In the absence of interaction Fermi-Dirac statistics was important because it, in a sense, represents the hard core repulsion potential, present in real fluids. On the other hand, Fermi-Dirac statistics sets a limit on the average density of the system: in combination with Monte Carlo selection rules for updates it reduces the allowed overall density to a range within which phase-separation may not be possible. Thus, our choice of Maxwell-Boltzmann statistics combines a simplicity of numerical implementation and the advantage of no restriction on allowed density.

Further, choices [iv] and [v] allow us to build the interactions in the following way. Let us introduce the following quantities:

- n_i is the number of particles at a given site i , $i = 1, 2, \dots, V$, V is the total number of sites
- σ_{ik} is the particle spin variable (+1 for lower energy state, -1 for higher energy state of particle), $k_i = 1, \dots, n_i$ labels particles on site i .
- S_i is a site-spin variables defined as:

$$S_i = \sum_{k=1}^{n_i} \sigma_{ik} \quad (5.4)$$

In terms of the above variables we define the following Hamiltonian for the system:

$$H = -\frac{J}{2} \sum_i \sum_{j=\langle ij \rangle} S_i S_j - \Delta \sum_i S_i + \frac{1}{2} U \sum_i n_i (n_i - 1) \quad (5.5)$$

where $j = \langle ij \rangle$ are sites neighboring site i . The first term contains a constant of spin interaction J , when it is positive, the interactions are “ferromagnetic”. Thus, this term drives separation into a spin-plus phase and a spin-minus phase.

The second term favors the spin-plus phase so that at low temperature this phase is the ground state. However, at higher temperature the entropy associated with the degenerate energy levels will cause a transition to the spin-minus phase. As described in Section 5.2 the degeneracy induces the first-order phase transition.

Finally, the last term in (5.5) represents an on-site repulsion between particles. With the translational degrees of freedom and Maxwell-Boltzmann statistics, the model allows any number of particles at a site. As in any model for gases or fluids, we require the presence of a strong short-range repulsion between particles. The strength of this repulsion is determined by the following arguments. For the limiting case of a one-phase state of the system (all spins plus or all spins minus), we have: $S_i S_j = n_i n_j$. Let $\rho = n_i = n_j$ be the local (uniform) density (number of particles per site). The terms in (5.5) which are second-order in ρ then read: $H_{local} = -\frac{1}{2}Jz\rho^2 + \frac{1}{2}U\rho^2$, where $z = 6$ is the number of nearest neighbors for each site. This sets the following condition for U :

$$U > Jz \quad (5.6)$$

Should this not be satisfied, the attraction wins over the repulsion, resulting in the piling up of all the particles at one site.

Thus, we have defined a model that can be viewed as a mixture of a cellular automaton and a modified Ising model. There have been a few attempts in the literature that propose similar ideas. Chopard and Droz [98], for example, describe “an Ising cellular automata fluid” where particles of a standard FHP model can have two spin states $s = +1$ and $s = -1$ interacting as in Ising model. Fermi-Dirac statistics is preserved. The updating rule for spin dynamics is also taken from the Monte-Carlo method and defines the temperature in a natural way. However, the spin variables in this model are not coupled to energy, so that transport properties are not expected to be dependent on temperature. Below the Ising critical temperature the model simulates a binary fluid. The same criticism can be addressed to the LB model for magnetic fluids proposed by Sofonea [94]. In another attempt, Yezpez [96]

exploited the idea of Monte-Carlo type dynamics in the model of a lattice-gas with long-range interactions first introduced by Appert and Zaleski [90]. In his model the role of potential energy is taken by an effective potential of interaction, deduced from the collision rules. The tracking of this interaction energy is difficult, and the use of Monte-Carlo algorithms for simulations of non-equilibrium phenomena is questionable.

5.4 Evolution

The evolution of our model is described using the languages of both LGCA and of Monte-Carlo technique. In terms of a LGCA we have a propagation step, a collision step, and a collision table which ensures the conservation of momentum.

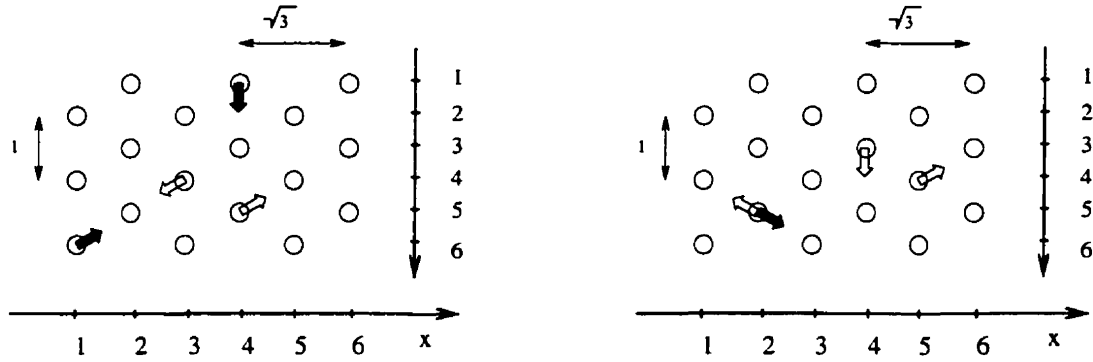


Figure 5.3: One time-step of evolution: initial configuration of four particles (left) evolves to its new state (right). Circles are the sites of hexagonal lattice, they have fixed coordinates (x,y) labeled on horizontal and vertical axes. Dark-grey arrows radiating from sites are spin-plus particles, light-grey are spin-minus particles. In the left-bottom corner of the initial configuration we have a pre-collision situation: a particle at site $(3,4)$ is heading towards a particle at site $(1,6)$. The other two particles, $(4,1)$ and $(4,5)$ are “ballistically” propagating through the lattice.

Figure 5.3 shows the “top-view” of one time-step of evolution for four particles on a lattice with 18 sites. The evolution of particles is similar to or different from the evolution of FHP-type model in the following ways:

- unlike for LGCA models we have a Monte-Carlo type selection rule for the propagation step. A particle does not necessarily propagate to its neighboring site, but attempts to do so. For each attempt, there is a cost of propagation. For a particle moving from site i (with neighboring sites j) to site i' (neighboring

sites j') with a change of spin from σ_{ik} to $\sigma_{i'k'}$ according to (5.5) this cost is:

$$\Delta E = -(J(\sum_{j'} S_{j'} - \sigma_{ik}) + \Delta)\sigma_{i'k'} + (J\sum_j S_j + \Delta)\sigma_{ik} + U(n_{i'} - n_i + 1) \quad (5.7)$$

In Figure 5.3 the particle initially at site (4,1) has six different possible output states: three of them with the particle ending up on the same (4,1) site, and three others with it ending up at the neighboring site (4,3). Ending up at the same site in one of three states, the singly degenerate spin-up state or the two degenerate spin-down state, may also be viewed not as part of propagation step, but as part of some “thermal equilibration” step. Similarly, the associated cost of energy for changing spin value from σ_{ik} to value σ'_{ik} at site i (neighboring sites j) is

$$\Delta E = -(J\sum_j S_j + \Delta)(\sigma'_{ik} - \sigma_{ik}) \quad (5.8)$$

- the collision step involves binary collisions only. From the “kinetic point of view” the rules are shown in Figure 2.2-a. However, the spin labels of the particles make for 9 energetically different outputs for each (again, out of 9) initial spin state of two colliding particles. The cost associated with σ_{i1k1} and σ_{i2k2} becoming σ'_{i1k1} and σ'_{i2k2} is

$$\Delta E = -(J\sum_j S_j + \Delta)(\sigma'_{i1k1} - \sigma_{i1k1} + \sigma'_{i2k2} - \sigma_{i2k2}) \quad (5.9)$$

The following immediate concerns must be addressed in the above scheme of evolution.

First of all, the new interaction part inevitably affects the kinetic part of the model: the particles propagate to their neighboring sites if, and only if, the Monte-Carlo selection rule (Sections 3.2.3 and 3.2.4) allows the move. While from the statistical mechanical point of view it is (thermodynamically) correct, from a kinetic point of view the particles seems NOT to undergo the displacement that is implied by their (labeled) velocity. However, we do not find this contradictory. We define *one Monte-Carlo Step (MCS)* as the time during which all of the particles are given a chance both to propagate and to collide (including the “thermal equilibration” subset of updates). Although all of the particles have speed equal $|c| = 1$, only a fraction δ_c of them

actually propagate to a new location during 1 MCS. In order, in statistical terms, that all particles are displaced to neighboring sites, we need to wait a time $1/\delta_c > 1$ MCS. In other words, all particles have some *effective velocity* $c_{eff} = \delta_c$, (dependent on temperature and other parameters, as we will see in the next sections) that makes the propagation step continuous and thermodynamically correct. Although the collision step includes only binary (head-on) collisions as a minimal set, the model does not suffer “spurious” conservation of momentum along lattice directions (Section 2.2) because of the probabilistic nature of the propagation step.

5.5 Numerical Simulation of Phase Coexistence

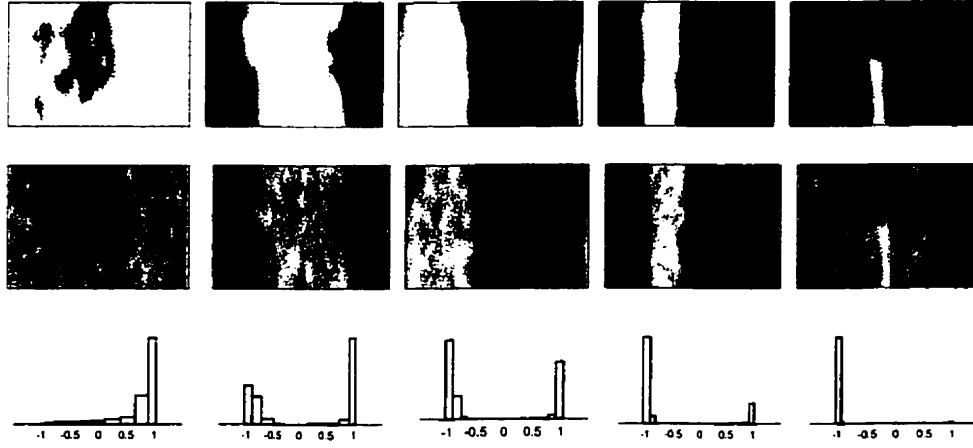


Figure 5.4: Magnetization (1st row), density (2nd row) , and magnetization histograms (3d row) for equilibrium distributions for a system of size $V = 64 \times 64 \times 2$ and numbers of particles $N = 4.13 \times V$. The total energy of the sample is increasing from left to right and takes the following values: 31.46, 34.87, 38.11, 41.41, 44.38 in units of J/V .

In this section we present the results of an equilibrium simulation of our model which show that it is indeed capable of simulating a phase transition and the coexistence of two phases. The parameter space is rather large: Ising attraction J , strength of repulsion U , half-width of energy separation between two spin-levels Δ and overall density ρ can all be varied¹. Our choice was as follows. We set $J = 1$ to set the energy scale (thus, all other energy variables are in units of J). Further, we set $U = 10J$ to satisfy condition (5.6). Performing then a systematic scan of a

¹In principle, the degeneracy of the spin-down level δ is also a parameter which can take any real number value $\delta \geq 1$.

now 2-dimensional parameter space $\{\rho, \Delta\}$, we discover phase separation behavior for $\Delta = 3.5J$ and overall density of the sample $\rho \simeq 4$ when the temperature is $T \sim 13J$.

We did the simulations with the multi-demon algorithm because it allows to better control and study the coexistence of phases, Section 3.2.4. Figure 5.4 shows the process of melting the spin-up phase into the spin-down phase by adding extra energy to a sample with a fixed number of sites and particles. The energy is the total energy of spin-system plus demons, and is therefore precisely constant. In this figure the values of magnetization and density were obtained by averaging around the area that includes nearest and next-nearest neighbors for each site. Thus, the phase-separation manifests itself in separation into different magnetization AND different density regions, with the low density (LD) phase having negative magnetization and the high density (HD) phase having positive magnetization. Figure 5.5 shows the energy dependence for magnetization and density for the same sample. Note that

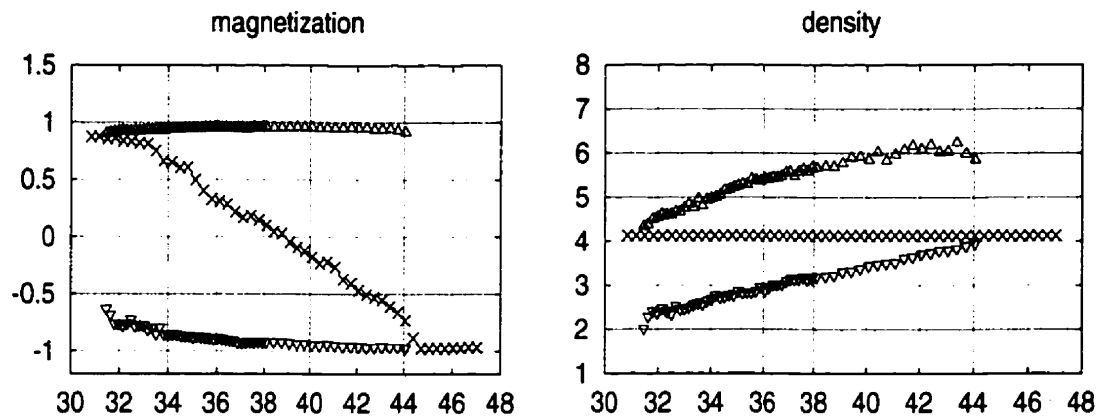


Figure 5.5: Magnetization and density versus total energy (in units of J/V). Angle-up triangles mark data for the HD phase, angle-down triangles mark data for the LD phase, and crosses mark the average values for the total sample.

the magnetization of the HD phase is always close to $+1$ and that of the LD phase close to -1 . However, the densities of the two phases vary in an approximately linear fashion as the energy of the system is increased.

The temperatures of the samples were measured in the manner discussed in Section 3.2.4 and Appendix A.2. The demons are in thermal equilibrium with spin system, so that the distribution of their energy tends to obey the exponential law $P(E_D) \sim \exp(E_D/T)$. Figure 5.6 thus shows the log plot of the measured distribution of demon

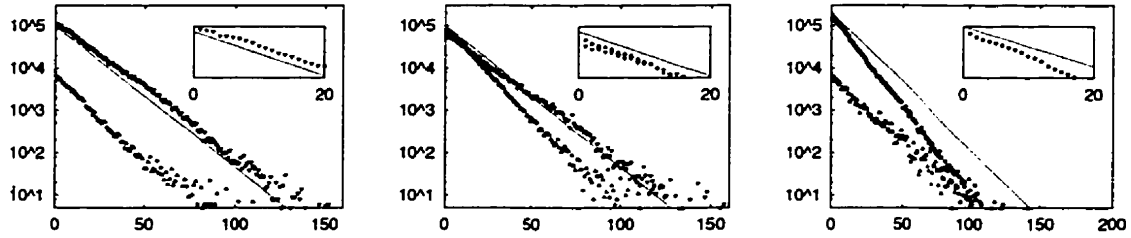


Figure 5.6: Log plot of demon distributions for configurations with: LD phase in minority ($E = 31.46 J/V$, left figure); approximately equal volume fraction of phases ($E = 38.11 J/V$, middle figure); HD phase in minority ($E = 44.38 J/V$, right figure). Angle-up triangles mark HD phase measurements, angle-down triangles mark LD phase measurements. Solid line shows the $\exp(-E_D/13)$ distribution. Samples are the same as used to obtain left, middle and right plots in Figure 5.4.

energies for three samples with different volume fractions of each phase compared with a slope of $T = 13.0J$. All these measured distributions are consistent with this slope - for energies below around $20J$ (as is seen from the enlargements in the upper corners of the figures)¹. Using the low-energy distributions we have measured the temperature along the whole path of the experiment (Figure 5.5) including the one-phase samples where the total energy is just a little outside the phase-coexistence region. To within 5% of accuracy we found the temperature to be constant at $T \simeq 13.0J$.

Thus, we have a situation with two phase coexistence in which the total energy of the system (which is controlled by the experiment) determines the volume fraction of each phase. Further, the energy must have at the same time a direct effect on the pressure in the system since the density of each phase varies with energy (subject to the total conservation of the number of particles and the volume): the fluid responds to energy changes just as any compressible fluid responds to pressure changes. These results confirm the ability of our particle-based model to simulate a two-phase fluid.

In next section we will try to understand the behavior of our model from mean field considerations.

¹As discussed in Appendix A.2, and confirmed in the simulations, the high energy tails of the distribution have tendency to decrease faster than $e^{(E_D/13)}$ for the LD phase, and slower in the HD phase.

5.6 Mean-Field Thermodynamics

To apply the mean field method to our model, we must combine features of its application to the Ising model, on the one hand, and to the Van-der-Waals model of a gas, on the other. The discreteness of space, time and spin-energy levels makes the computation of the partition function straightforward. However, since particles are moving from site to site, there is a distribution for the site occupation number n_i . The number of nearest neighbor particles is not fixed, in fact, it could vary from 0 to the total number of particles N . The energy levels available for each particle are thus almost continuous and infinite in number, with a density of states that reflects the actual site-distribution of particles.

The combination of configurational and translational degrees of freedom must therefore to be accounted for in a proper manner.

5.6.1 Mean Field Free Energy Functional

The mean field approximation implies replacing the site occupation number n_i and the spin-variable σ_{ik} in the microscopic Hamiltonian (5.5) by their position independent averages $\rho = \langle n_i \rangle = N/V$ and $m = \langle \sigma_{ik} \rangle$ respectively. Thus, the configurational mean-field energy of the system of spin-particles reads

$$E = N \left(\frac{U}{2} (\rho - 1) - \lambda m \right) \quad (5.10)$$

$$\lambda = \frac{1}{2} J z m \rho + \Delta \quad (5.11)$$

As before, $z = 6$ is a number of neighbors for each site of the lattice. The factor $\frac{1}{2}$ in (5.11) is needed to account for the double counting of exchange energy.

To arrive at the free energy functional, and, eventually, to calculate the thermodynamic characteristics of the model we next calculate the entropy. The number of ways Γ that correspond to the energy (5.10) has “kinetic” Γ_k and “configurational” Γ_c contributions. The kinetic part reflects the fact that each particle (out of N) can have V possible locations. Thus,

$$\Gamma_k = \frac{V^N}{N!} \quad (5.12)$$

This definition correspond to the rule of “correct Boltzmann counting”, Huang [87] p. 182. Further, the division of N particles into $N_\uparrow$ spin-up particles and $N_\downarrow$ spin-down

particles can be achieved in the following number of ways

$$\Gamma_c = \frac{N! \delta^{N_\downarrow}}{N_\uparrow! N_\downarrow!} \quad (5.13)$$

where $N = N_\uparrow + N_\downarrow$, $(N_\uparrow - N_\downarrow)/N = m$ and the degeneracy δ of states for spin-down particles is taken into account. The entropy then is defined as

$$\begin{aligned} S &= \ln(\Gamma_k \Gamma_c) = S_k + S_c \\ S_k &= -N(\ln(\rho) - 1) \end{aligned} \quad (5.14)$$

$$\begin{aligned} S_c &\simeq N(-n_\uparrow \ln(n_\uparrow) - n_\downarrow \ln(n_\downarrow) + n_\downarrow \ln(\delta)) \\ &= N \frac{1}{2} \left(\ln\left(\frac{4\delta}{1-m^2}\right) - m \ln\left(\delta \frac{1+m}{1-m}\right) \right) \end{aligned} \quad (5.15)$$

where $n_{\uparrow(\downarrow)} = N_{\uparrow(\downarrow)}/N$ and the Stirling approximation $\ln(A!) \simeq A \ln(A) - A$ has been used. It is useful to write explicitly the derivatives of energy and entropy with respect to magnetization

$$\frac{\partial S}{\partial m} = -N \frac{1}{2} \ln\left(\delta \frac{1+m}{1-m}\right) \quad (5.16)$$

$$\frac{\partial E}{\partial m} = -N(\rho z J m + \Delta) \quad (5.17)$$

It is to be noted, that the entropy (5.14) can take negative values for large densities. This should not cause concern, as it is well known that Maxwell-Boltzmann statistics lead to the violation of the third law of thermodynamics. For an ideal Maxwell gas, for example, the entropy takes the form of Sackur-Tetrode equation, Huang [87] p.184, which diverges to minus infinity as temperature goes to zero. This is due to the existence of a ground state which is multiply degenerate under the permutation of particles.

The free energy $F = E - TS$ then is a function of N , V and m . In spin systems the derivative from free energy over the magnetization defines the external magnetic field H . In our case $H = 0$, thus, the conditions $\frac{\partial F}{\partial m}|_{\{N,V,T\}} = 0$ determines the configurational equation of states or magnetization-density relation, which, using derivatives (5.16) and (5.17) takes the following form:

$$T = \frac{2(\rho z J m + \Delta)}{\ln\left(\delta \frac{1+m}{1-m}\right)} \quad (5.18)$$

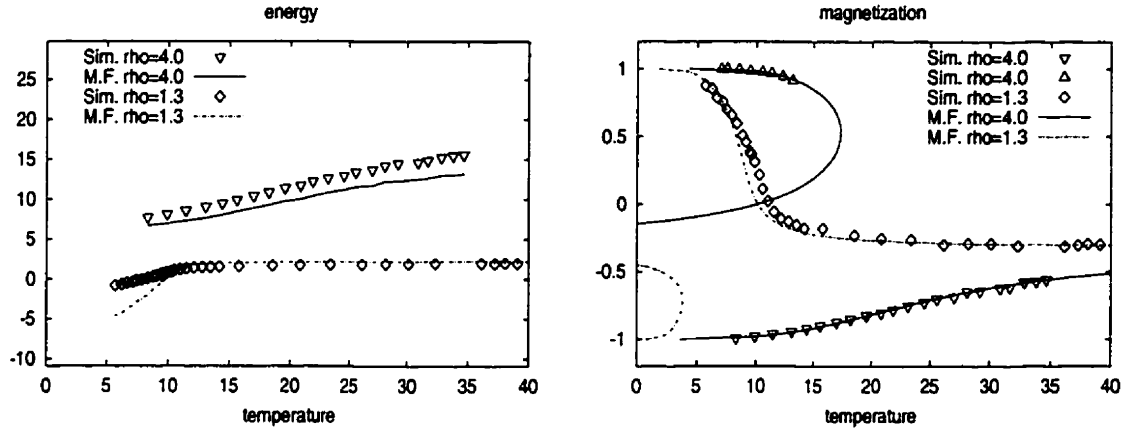


Figure 5.7: Energy of the spin systems in energy units per particle (left) and magnetization (right) versus temperature for two systems with total density $\rho = 4$ and $\rho = 1.3$ particles per site. The simulation results are plotted over the mean-field solutions (5.10) and (5.18). For the simulation results, the error-bars are vertical and of order of the size of the symbols at high temperatures, and horizontal and of order of 2-3 sizes of the symbols at low temperatures.

The mean-field equation of state (5.18) turns out to describe the system, when not in the coexistence region, very well. Figure 5.7 (right) shows the mean-field plots of magnetization-temperature dependence for two different densities, $\rho = 4$ and $\rho = 1.3$, and the “working” set of parameters $J = 1$, $z = 6$, $\Delta = 3.5$, $\delta = 2$, $U = 10$ plotted over the results of numerical simulations. The high-temperature limit of magnetization does not depend on density

$$m_c \equiv \lim_{T \rightarrow \infty} m = \frac{1 - \delta}{1 + \delta} = -\frac{1}{3} \quad (5.19)$$

When the system approaches its high-temperature limit (5.19) from the state with $m < m_c$, the denominator of expression (5.18) approaches zero with negative values. Hence the numerator also must be negative: $\rho z J m < \Delta$. Similarly, the system approaches its high-temperature state with $m > m_c$ only when $\rho z J m > \Delta$. Thus, the form of solution must depend on the relation of the density of the system to the critical density:

$$\rho_c = \frac{\Delta}{z J m_c} = \frac{\Delta(\delta + 1)}{z J (\delta - 1)} = 1.75 \quad (5.20)$$

Indeed, the solutions for two representative densities $\rho = 4 > \rho_c$ and $\rho = 1.3 < \rho_c$, plotted in Figure 5.7 (right), are different: while the lower density system can be heated from the zero-temperature state $m = +1$ to the high-temperature state (5.19) along a continuous path of equilibrium solutions, the high density system shows a

discontinuity. Thus, expression (5.20) defines the critical density, above which phase-separation is possible.

The results of simulations, also shown in Figure 5.7 are in good agreement with mean-field. For both densities, we started from the high-temperature state and cooled the system down by gradually removing energy from the system. For density $\rho = 4$ the system gets easily into a metastable supercooled state (for temperatures $T < 13 J$). It is only when we force a small patch of spins to have positive magnetization, that the low-temperature phase develops and, after further cooling, replaces the high-temperature phase.

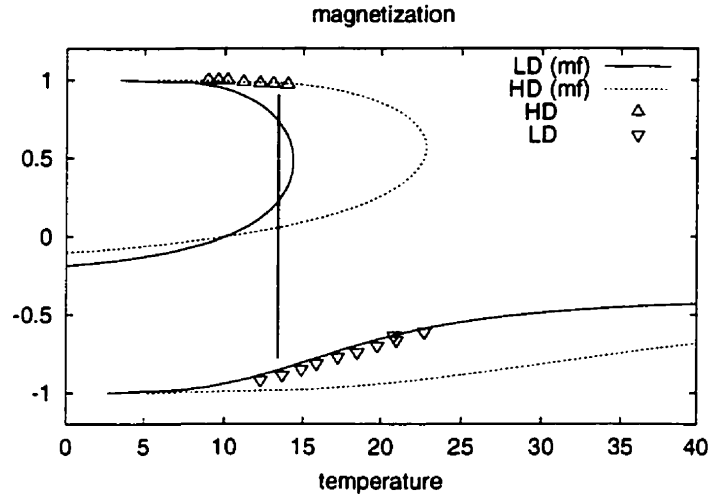


Figure 5.8: The equilibrium simulations of two samples: $\rho = 3.11$ (LD) $\rho = 5.61$ that can coexist at $T = 13 J$ (marked by vertical line). The error-bars here are horizontal and are approximately double the size of the symbol. Solid lines mark the mean field solutions for each density.

In Figure 5.8 we show the superimposed results of simulations for two samples with densities that can coexist in a state of equal volume fraction for each phase (as shown in the middle column in Figure 5.4). Again, the agreement with mean-field calculations is excellent.

To explore the phase coexistence region in more detail, we can calculate the mean-field thermodynamic pressure $P = -\frac{\partial F}{\partial V}|_{\{N,m,T\}}$ and the chemical potential $\mu = \frac{\partial F}{\partial N}|_{\{V,m,T\}}$ as derivatives of the free-energy. The necessary derivatives are

$$\frac{\partial E}{\partial V}|_{\{N,m,T\}} = -\frac{1}{2}\rho^2(U - Jzm^2) \quad (5.21)$$

$$\frac{\partial E}{\partial N}|_{\{V,m,T\}} = U\rho - \frac{U}{2} - m(Jzm\rho + \Delta) \quad (5.22)$$

$$\frac{\partial S_k}{\partial V}|_{\{N,m,T\}} = \rho; \quad \frac{\partial S_k}{\partial N}|_{\{V,m,T\}} = -\ln(\rho); \quad \frac{\partial S_c}{\partial N}|_{\{V,m,T\}} = \frac{S_c}{N} \quad (5.23)$$

The final expressions for pressure and chemical potential then read:

$$P = T\rho + \frac{1}{2}\rho^2(U - Jzm^2) \quad (5.24)$$

$$\mu = T \ln(\rho) + U\rho - \frac{U}{2} - \frac{T}{2} \ln\left(\frac{4\delta}{1-m^2}\right) \quad (5.25)$$

The first terms in the above equations are the ideal-gas terms that result from the “kinetic” contribution to the entropy (5.14).

It is immediately evident that the pressure defined by equation (5.24) cannot be the same for both the LD and HD phases in equilibrium in the coexistence region. Since m^2 is (almost) the same for both phases, and since T , U and J are constant, the pressure is a monotonically increasing function of the density. Seemingly, this invalidates the mean-field approach. However, in LGCA theory, the pressure that actually enters the hydrodynamics macroscopic equations is the so called “kinetic” pressure (3.3) (also discussed in Section 3.3) that differs from the “thermodynamic” pressure (Ernst [90]; Gerits, Ernst and Frenkel [93])¹. For an interacting model, the definition of “kinetic pressure” is not obvious. However, we argue that the kinetic pressure is proportional to the (average) rate of momentum transfer from any one site to any one of its neighbors. Thus, we write $P_k \sim rc$ where c is the momentum carried by each particle in our one-speed model (mass is defined to be unity) and r is the (average) transition rate from one site to another². However, because of detailed balance, the rate r is the same for each and every site, irrespective of the density at that site. P_k is therefore the same everywhere, and, in particular, must be the same for two coexisting phases. Indeed, in one simulation, we counted directly the numbers of momentum transfers during one MCS separately in the LD ($\rho = 3.11$) and HD ($\rho = 5.61$) samples of equal volume ($V = 32768$) and at equal temperature

¹In reference Gerits, Ernst and Frenkel [93] authors cite the (unpublished) work of Cercignany who showed, that the difference between kinetic and thermodynamics pressure is related to the limited number of velocity states in LGCA models and the proper description of system must be in terms of kinetic pressure.

²The transition rate is a product of density and effective velocity c_{eff} discussed in Section 5.4.

($T = 13 J$). These numbers, divided by volume, give the estimates for the transition rates r in each phase. To within 5% of accuracy they are equal: $r_{LD} \simeq r_{HD} \simeq 0.64$.

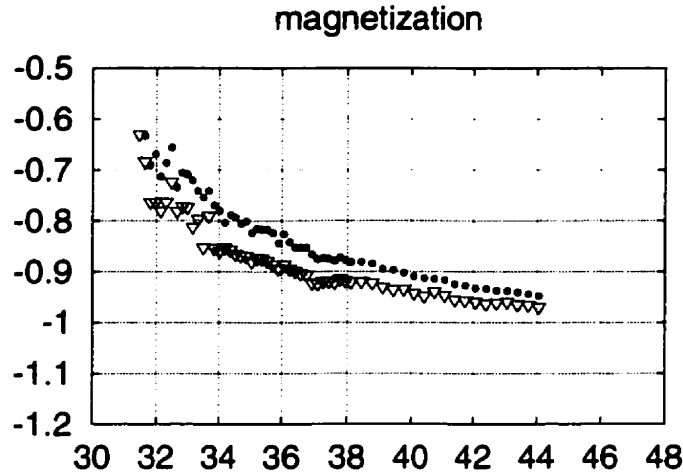


Figure 5.9: Mean-field (black dots) and measured (triangles) magnetizations for low densities in the experiment, described in Section 5.5. Because of $\ln(1 - m^2)$ term in the mean-field expression for chemical potential, the difference between two magnetizations makes mean-field evaluation for low densities inaccurate.

The situation with the chemical potential is more straightforward. The value given by equation (5.25) depends critically on the $\ln(1 - m^2)$ term, which is the only negative term (apart from the unimportant constant, $-U/2$). As we have already seen, however, the mean-field calculation gives values for m which are not in precise agreement with the data. This is most clearly evident for low densities, as in Figure 5.9. It is therefore not unexpected that the values of μ from the mean-field expression are not the same for coexisting phases. Although small adjustments to the values of m might perhaps bring about equality, this is unlikely to be successful when ρ is small - *i.e.* close to the critical value 1.75 identified by (5.20). For such densities, the fluctuations of ρ and of m are likely to be important - they seem to be visible in the left-most panel of Figure 5.4 - so that mean-field estimates will be quite unreliable.

Quantitative understanding of the coexistence region of our model is thus not possible within a mean field approach. Nevertheless, since the phases are well-defined, we can proceed to measure their transport properties, in preparation for exploring non-equilibrium phenomena.

5.7 The Density Correlation Function

To examine and evaluate the transport properties of the fluids simulated by our model, we have measured the density correlation function (structure factor) (2.43) for the two phases¹ and obtained some of the transport coefficients (such as the heat diffusivity D_T , and the sound damping constant Γ , see Section 2.4.2) by fitting to the Landau-Placzek form of the structure factor (2.50).

Our measurements of the structure factor were done first for two different samples that represented the two phases of the fluid that can coexist in equilibrium at temperature $T = 13 J$ (as in the experiment described in Section 5.5).

Each sample (of size 128×128) was prepared with a distribution of magnetization, density and demons sampled from the phases in coexistence as shown in the third column of Figure 5.4. As before, we used the index LD for the low-density negative magnetization phase sample and HD for the high-density positive magnetization phase sample. The two samples, with $\rho_{LD} = 3.11$, $\rho_{HD} = 5.61$, were then allowed to evolve for extra time (10,000 MCS) to ensure their equilibrium state.

Figure 5.10 shows the three-dimensional plots of the structure factor for the two samples, evolved over 4096 MCS. The dependence on k is obtained after averaging

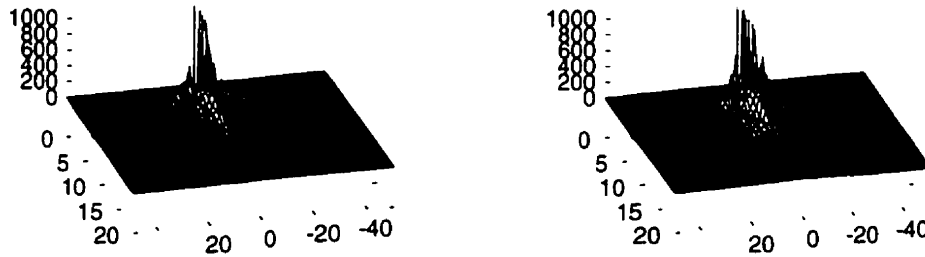


Figure 5.10: Structure Factor $S(k, \omega)$ for the low-density phase $\rho_{LD} = 3.11$ (left) and the high density phase $\rho_{HD} = 5.61$ (right).

over angle, to reduce noise. The figures show only small k ($0 < |k| < 20$ in units of $k_0 = 2\pi/128$) and small ω ($-40 < \omega < 40$ in units of $\omega_0 = 2\pi/4096$). The constant k cross-section of the measurements are plotted in Figure 5.11 for $k = 4k_0$. In the

¹With our “default” set of the parameters: $J = 1$, $U = 10$, $\Delta = 3.5$, $\delta = 2$.

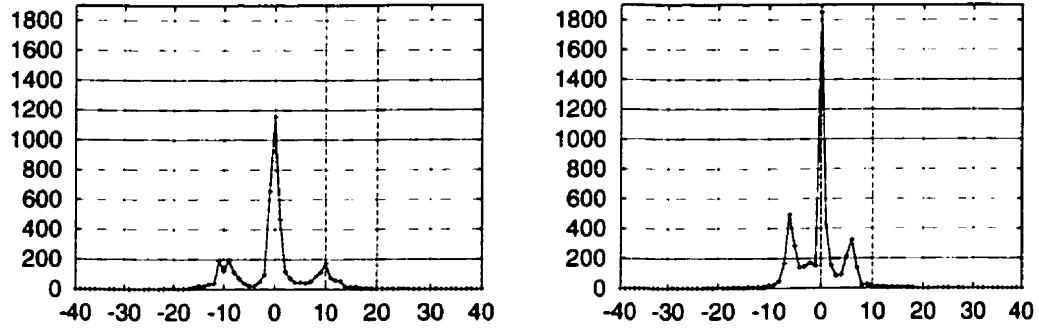


Figure 5.11: Structure Factor $S(\omega)$ for the low-density phase $\rho_{LD} = 3.11$ (left) and high density phase $\rho_{HD} = 5.61$ (right). $k = 42\pi/128$

low-density phase the location of the sound peaks is estimated as $\omega_s^{LD} = \pm 10\omega_0$ (with a tolerance of 5%) which gives a value for the sound velocity as $c_s^{LD} = \omega_s^{LD}/k = 10/4 (\omega_0/k_0) \simeq 0.08 \pm 0.02$. Similar analysis for the high-density phase gives $c_s^{HD} = \omega_s^{LD}/k = 6/4 (\omega_0/k_0) \simeq 0.05 \pm 0.01$.

The thermal diffusion coefficient D_T and the sound damping coefficient Γ can be estimated from the structure factor measurements. The width of the Rayleigh peak $\Delta\omega_T$ is about $(2 \pm 0.5)\omega_0$ in the LD phase and $(1.5 \pm 0.5)\omega_0$ in the HD phase. The width of the sound peaks $\Delta\omega_c$ are $\simeq (4 \pm 0.5)\omega_0$ and $\simeq (2 \pm 0.5)\omega_0$ respectively, resulting in the estimates:

$$D_T^{LD} = \frac{\Delta\omega_T}{k^2} = \frac{2\omega_0}{(4k_0)^2} \simeq 0.08 \pm 0.02 \quad (5.26)$$

$$D_T^{HD} = \frac{1.5\omega_0}{(4k_0)^2} \simeq 0.06 \pm 0.02 \quad (5.27)$$

$$\Gamma^{LD} = \frac{\Delta\omega_c}{k^2} = \frac{4\omega_0}{(4k_0)^2} \simeq 0.16 \pm 0.02 \quad (5.28)$$

$$\Gamma^{HD} = \frac{2\omega_0}{(4k_0)^2} \simeq 0.08 \pm 0.02 \quad (5.29)$$

Because the samples are close to the first order liquid-gas-type transition, we observe the domination of the Rayleigh peak in our measurements. This is because the intensity of the Rayleigh peak is determined by ratio of specific heats $\gamma = \frac{C_p}{C_v}$ (see expression (2.43), for example). Because of the relation

$$C_p - C_v = \frac{T}{\rho^2} \left(\frac{\partial p}{\partial T} \right)_\rho^2 \left(\frac{\partial \rho}{\partial p} \right)_T \quad (5.30)$$

the constant-pressure specific heat must diverge at transition temperature, and $\gamma \rightarrow \infty$. The γ -factor can therefore be estimated from the structure function. Indeed, the direct integration of the first three terms of (2.43) gives

$$\frac{I_R}{2I_s} = \gamma - 1 \quad (5.31)$$

where I_R and I_s are the integrated intensities of the Rayleigh and sound peaks respectively or the areas defined by the respective Lorentzians. The direct numerical evaluation of these areas from the results on Figure 5.11 yields

$$\gamma^{LD} \simeq \gamma^{HD} \sim 3.0 \pm 0.5 \quad (5.32)$$

We summarize many of the transport properties of the two phases at $T = 13 J$ in Table 6.1 in Section 6.5.1.

Figure 5.12 shows the constant k cross-section of the structure factor for the LD phase at two different temperatures: $T = 24 J$ and $T = 70 J$. We observe the shift of

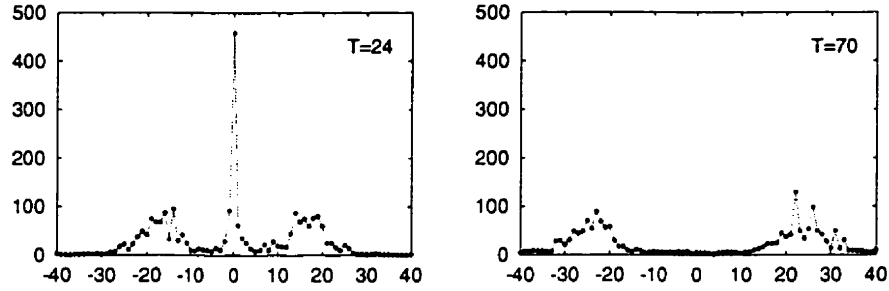


Figure 5.12: Structure Factor for the low-density phase $\rho = 3.11$ at temperatures $T = 24$ (left) and $T = 70$ (right). $k = 4k_0$.

the sound peaks with temperature so that the sound velocity increases with temperature, and, as expected, the Rayleigh peak gradually disappears. This is because at high temperatures the model becomes effectively “athermal”: the interactions have no effect on the kinetics of one-speed particles. Thus, the value of the speed of sound approaches the limit of sound speed for the FHP model¹: $c_s(T \rightarrow \infty) = c/\sqrt{2} \simeq 0.35$. We have measured (Figure 5.12) $c_s(T = 70 J) \simeq 0.25$.

¹In our definition of 1 MCS, Section 5.4, the effective speed of particles at high temperatures is $c = 0.5$.

Interfaces in fluids with a 1st order phase transition

6.1 *Introduction*

In this chapter, we demonstrate the application of our model to a simple example of the complex phenomena that exist at surfaces between two phases of a fluid: the roughening of the interface. Thus we will illustrate the advantages of our approach and use our model to study an interesting non-equilibrium phenomenon. The roughening here is an evolution with time of an initially flat boundary between two fluid phases. As a part of surface science, fluid-on-fluid roughening is interesting because of the insight it may give about the role of hydrodynamics in the mechanisms for surface evolution (Shinozaki [93]). These mechanisms are also expected to be present in more interesting non-equilibrium experiments: spinodal decomposition, driven hydrodynamic systems.

A wide variety of methods and models deals with solid-on-solid or gas-on-solid roughening (Godrèche [92]; Family and Vicsek [91]). A great deal of progress has been made in these cases because of the large number of experiments and simulations that can be performed on solids or gases. Both real and computer experiments with fluid-fluid interface in equilibrium are more complicated. In the first case it is the short values of relaxation times for interface-driving forces that cause difficulties in the control and analysis of the interface. In the second case, the known models for simulations with hydrodynamics are computationally costly, for example molecular dynamics, or they involve the violation of the semi-detailed balance condition (2.19) and inconsistent thermodynamics (Flekkøy and Rothman [96]; Starr et al. [96]; Stelitano and Rothman [00]). However, with our new model for simulating the thermal hydrodynamics of a system with a first-order phase transition, we are well placed

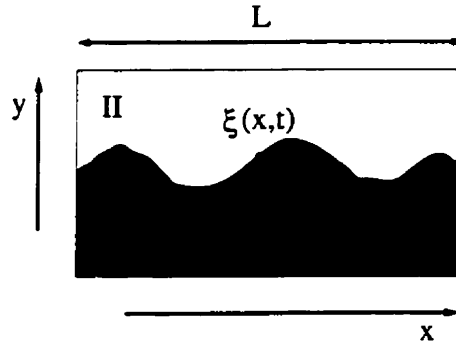


Figure 6.1: The rough interface

to learn more about the interface between fluids.

This chapter is presented in three parts: [1.] a short overview of the theory of interface roughening where we define and describe the quantities of interest, [2.] the results of computer simulations of interface roughening for the two-phase fluid in equilibrium using the model with interactions as introduced in Chapter 5, and [3.] theoretical arguments for the explanation of the observed scaling regimes.

6.2 Theory of Interface Roughening

The equilibrium between two phases in a system of some size L (Figure 6.1) is the state where the temperature, pressure and chemical potential of each phase are equal. In addition, the interface between the phases is stable and rough. This means that the interface is such that all the forces acting on it are in equilibrium. Let the interface be parallel to the x -axis. We define the state of the interface by defining the y -coordinate of each point on it to be a function of x and time t . We call this function the displacement variable $\xi(x, t)$, with the reference point in the y direction chosen so that $\langle \xi(x, t) \rangle = 0$. The qualitative description of roughness, then, is the width of the interface W , defined as

$$W = \sqrt{\langle \xi(x, t)^2 \rangle} \quad (6.1)$$

Now, if we force the interface out of its rough equilibrium state (by setting it straight and flat, for example) the interface will move back towards its equilibrium state. The equation of motion then relates the velocity of the surface $v = \dot{\xi}$ to the interface curvature $\kappa \sim \frac{1}{R}$ where R is the local radius of curvature. In its simplest form it

takes the form

$$v = D\kappa + \mu \quad (6.2)$$

where D is a constant coefficient in a Taylor series for small κ , and μ is a stochastic driving term. With details which depend on the particular model, this equation becomes the Langevin equation with appropriate boundary conditions. With the usual approximation of an “almost flat interface” with no overhangs or bubbles, $W \ll L$, the instantaneous position of the interface $\xi(x, t)$ is a single-valued function of the coordinate x . The Langevin equation involves the stochastic driving term μ , which is a random Gaussian variable with $\langle \mu \rangle = 0$ and $\langle \mu(s, t) \mu(s', t) \rangle = B \delta(s - s') \delta(t - t')$ where B is a constant, and s denotes positions along the arc length of the interface. Clearly, if μ is Gaussian, so is ξ . So we only need compute the second moment of ξ to have a complete solution.

Assuming at $t = 0$ that we have completely flat interface, a Fourier transform gives a formal solution that can be put in the general form (Grant [99])

$$\langle \xi(k, t) \xi(k', t) \rangle = \hat{G}(k, t) (2\pi)^{d-1} \delta(k + k') \quad (6.3)$$

where

$$\hat{G}(k, t) = \frac{B}{2Dk^{2-\mu}} (1 - e^{-2Dk^z t}) \quad (6.4)$$

and μ and z are some model-dependent scaling exponents. For linear theories $\mu = 0$ always. The late time solution must be the equilibrium solution which can be obtained independently from minimizing the free energy $F = \sigma L^{d-1}$ of the interface. Here σ is the surface tension coefficient. The comparison between these solutions gives the relation $B = 2D \frac{k_B T}{\sigma}$, where $k_B = 1$ is the Boltzmann constant and T is the temperature. Thus, the function $\hat{G} = \langle \xi_k^2 \rangle$ must be

$$\frac{T}{\sigma k^2} (1 - e^{-t/\tau}); \quad \tau = \frac{1}{2Dk^z} \quad (6.5)$$

where we have defined a relaxation time τ for a given mode k .

In terms of this function the interface width W is computed as

$$W^2 = \int \frac{dk}{(2\pi)^{d-1}} \hat{G}(k, t) \quad (6.6)$$

where $\frac{2\pi}{L} < |k| < \frac{2\pi}{a}$, with a the lattice constant. Straightforward manipulation gives

$$W^2 = \frac{1}{z} (Dt)^{\frac{1}{z}} \int_{x_L}^{x_a} dx \frac{1 - \exp(-x)}{x^{\frac{1+z}{z}}} \quad (6.7)$$

where $x_L = (\frac{2\pi}{L})^z Dt$ and $x_a = (\frac{2\pi}{a})^z Dt$. When the above integral is evaluated for $L \rightarrow \infty$, $a \rightarrow 0$ and finite time t , we see that the roughening of the interface is described by

$$W^2 \sim t^{\frac{1}{z}} \quad (6.8)$$

Thus, the scaling exponent for “slowing down”, z , characterizes an interface relaxing towards its equilibrium state. The equation that connects the relaxation time τ and the k -vector is called the dispersion relation. The dispersion relation allows us to determine z for a particular model with boundary conditions. More details on scale invariance in growth processes can be found in Krug [97].

6.3 Numerical Simulations of Interface Roughening

Here we present the results of numerical simulations for the evolution of the interface, initially flat, with time. We use the model studied in Chapter 5. We choose to run

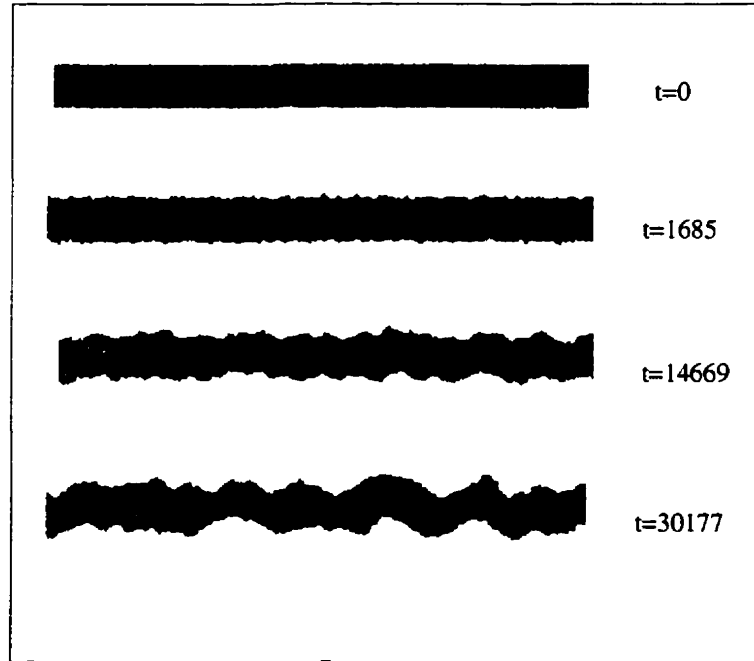


Figure 6.2: Evolution of initially flat interface with time. Grey color shows the low-density (LD) (negative magnetization) fluid phase

our model with the multi-demon algorithm (Section 3.2.4) so that we do include thermal diffusion effects in the evolution of the interface. With the “default” set of the parameters: $U = 10, J = 1, \Delta = 3.5, \delta = 2$, and with periodic boundary conditions, we created two initial flat interfaces in a sample of size 1024×128 .

The two-phase sample at $T = 13$ (see Section 5.5) with equal volume fraction of each phase provided the distributions for the density, magnetization and demons of the two phases for this sample. This initial configuration was then run for 50,000 MCS, storing intermediate configurations from which were extracted the width of the interface (6.1) and other characteristics. The numerical algorithm for determining the location of interfaces is described in Appendix A.3.

Figure 6.2 shows profiles of the interfaces for different times. Correspondingly,

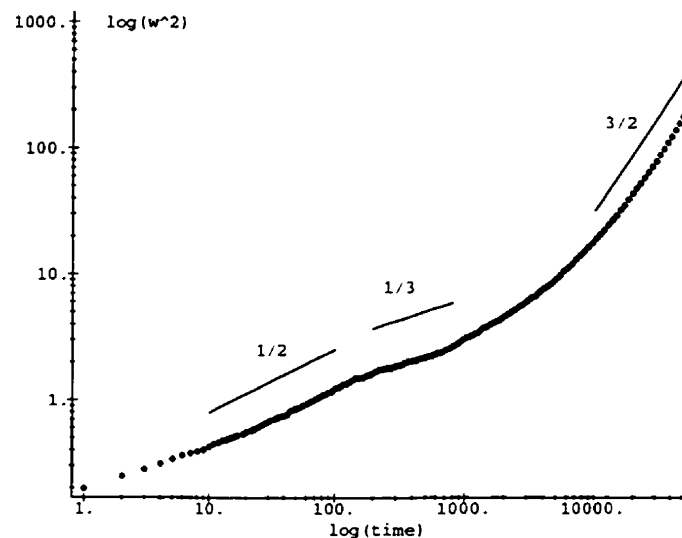


Figure 6.3: Log-Log plot of square of the width as a function of time. The solid lines, guides to the eye, have slopes $1/2$, $1/3$ and $3/2$ respectively.

Figure 6.3 shows the log-log-plot of the square of the width of the interface as a function of time. Assuming the power-law dependence (6.8) we observe three regimes in the roughening of the interface:

- early time $t^{\frac{1}{2}}$ regime ($z = 2$).
- $t^{\frac{1}{3}}$ growth ($z = 3$) observed in time-span 100-1000 MCSs.
- late time $t^{\frac{3}{2}}$ growth ($z = 2/3$)

It is not surprising that we observe the first two regimes for the growth of the interface. They are known in the literature as a consequence of the stabilizing effects of surface tension and of the thermal diffusion of the latent heat produced at the interface (see Section 6.4.1). However, a new $t^{\frac{3}{2}}$ regime at late time indicates the presence of a new mechanism for interface evolution. We call it an “inertial” regime, since we believe the new mechanism is a consequence of coupling the interface evolution to hydrodynamics. To validate our belief, we seek for evidence of velocity correlations of the fluid phases along the interface.

A few interesting observations are very suggestive. Figure 6.4 shows the results

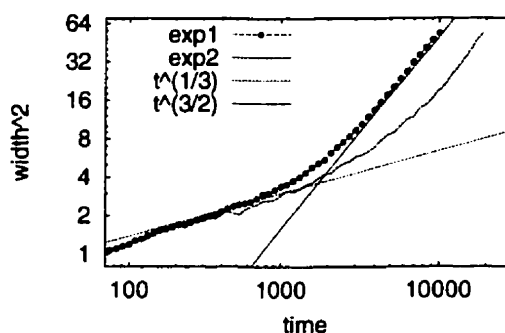


Figure 6.4: Log-Log plot of square of the width as a function of time for a sample of size 1024×64 , for two different ways of preparing the initial sample, as discussed in the text.

of roughening simulations for two different ways of creating the initial sample. In experiment 1 (shown as line-dots) the sample was re-initialized 5 times from the results of previous runs by modifying the near-interface regions so that the interface again became flat. Thus, each of the initial samples preserved the velocity correlations of the bulk phases. The results of these five runs were averaged. The presence of similar velocity correlations in the initial samples is seen from comparison of the profiles of the interfaces for these samples at late times. Two such profiles are shown in Figure 6.5. It seems clear that their resemblance is due to the persistence of velocity

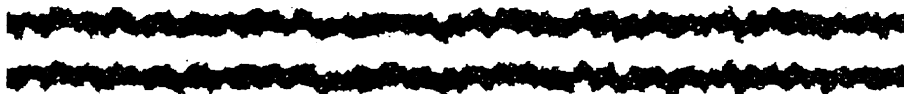


Figure 6.5: The “independent” runs still show the similar profile in long-wavelength regime.

correlations.

In the second experiment (shown as the solid line in Figure 6.4) the initial sample was first prepared as in the first experiment, but we then randomized the direction of the velocities for each particle in the sample. As we clearly see, the onset of the inertial $t^{3/2}$ regime in the latter case is delayed. This strongly indicates that the velocity correlations are the important factors in the inertial regime.

However, the most convincing argument for the existence of hydrodynamic effects in the late time regime is a the plot of the average velocity field in the sample at late times. The noise inherent to LGCA (Section 2.4.3) requires averages over space and/or time. The choice of temporal and spatial scales must be much larger than one time step and one lattice unit, but still small enough that the mass flux $\sum_i \langle n_i \rangle c_i$ varies slowly and the macroscopic description of the fluid (2.5) is valid. We also have an upper limit on spatial scale: it cannot exceed the (longest) wavelength of the interface or the correlation between interface and velocity field will be averaged out¹.

Figure 6.6 shows the measured velocity field in part of the sample at late time. The result was obtained by averaging over the area defined by a hexagon with side-length of 3 lattice units (which includes 19 sites) and over a time of 500 MCSs². We also superimposed the magnetization field plot so the interface location is seen. The figure clearly shows the presence of fluid motion in the LD phase along the interface.

Motion of the fluid which is coherent with the interface results, in our view, from the HD phase invading the LD phase and pushing the LD-phase fluid sideways along the interface. We do not observe the similar velocity correlations in HD phase, although they may be present: in this phase the different density and viscosity (see Table 6.1) require different time- and length- scales for the appropriate macroscopic description.

¹We can also argue that as soon as the wavelength of the interface profile exceeds the spatial scale of the average, correlations between the interface and the fluid velocity become possible. We will return to this argument in Section 6.4.2.

²With this spatial scale we tried different timescales. Only for the timescales above 300 MCSs did correlations in the velocity become visible through the noise.

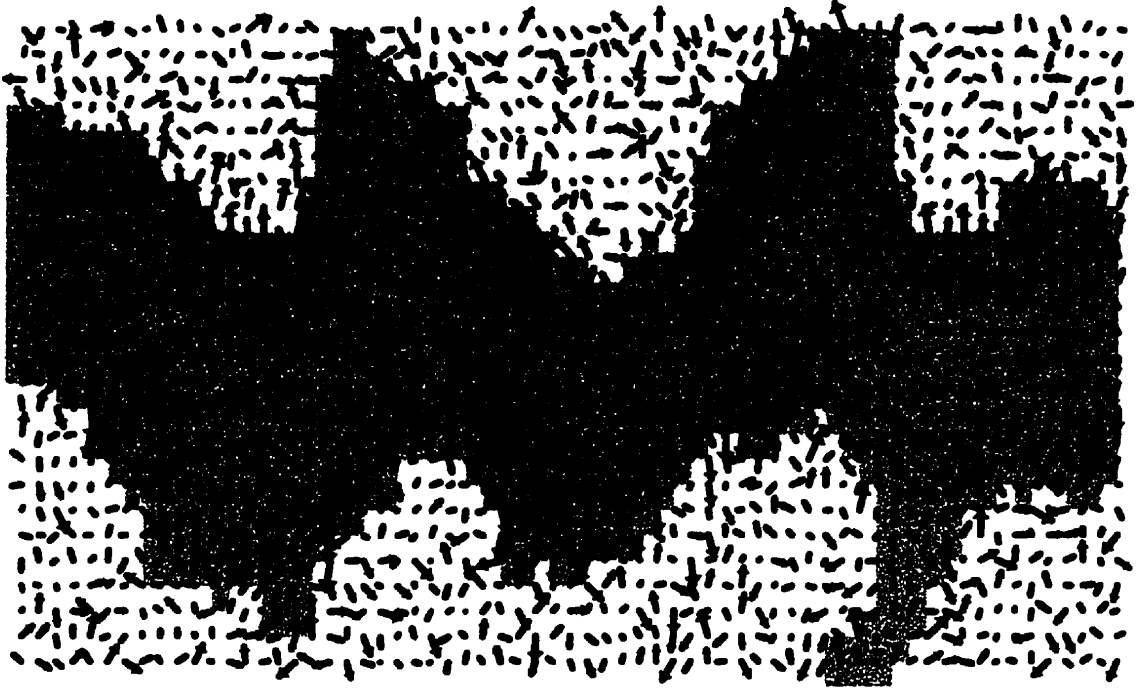


Figure 6.6: The velocity field in the late time configuration of the system. The aspect ratio is not preserved. Dark grey corresponds to the low-density (LD) negative magnetization phase.

6.4 Theoretical Predictions

In this section we explain the observed behavior of the interface.

6.4.1 The Model with Latent Heat and Diffusion in the Bulk

As mentioned earlier, the first two regimes ($z = 3$ and $z = 2$) in interface evolution are well described by a model in which the relaxation of the interface towards equilibrium is controlled by surface tension, thermal fluctuations and by the production and absorption of latent heat at the interface. This model was studied both theoretically and numerically by Harris, Jörgenson and Grant [92] and we adopt it here as a starting point for the full model with hydrodynamics.

We begin by writing the determining equation and boundary condition for the interface motion. These are (Langer [80])

$$\bar{v} = -d_0\kappa \quad (6.9)$$

$$D_T \nabla^2 \bar{v} = \frac{\partial \bar{v}}{\partial t} \quad (6.10)$$

$$v_n = D_T [\nabla \bar{v}_I - \nabla \bar{v}_{II}] \cdot \hat{n} \quad (6.11)$$

where $\bar{v}$ (upsilon with overline) is the dimensionless under-cooling, defined in terms of the temperature T as $(\Lambda/C_p)\bar{v} = T - \Lambda/C_p$, with Λ/C_p the ratio of the latent heat to the specific heat at constant pressure. As in Langer [80], we make an approximation for the specific heat¹ $C_p^I = C_p^{II} = C_p$. Further, κ is the curvature of the interface², d_0 is the capillary length (defined later as (6.14)), and D_T is the diffusion constant. The velocity component v_n is perpendicular to the interface and directed into phase II .

The second equation (6.10) is the diffusion of heat near the boundary. In general, we must write this equation separately for phase I and phase II , as they will have different values of D_T . However, for the qualitative incorporation of the mechanism of diffusion it suffices to assume them equal.

The third equation (6.11) is the condition of heat conservation at a point on the moving interface. A more familiar form (Langer [80]) of this equation is $\Lambda v_n = [D_T^I C_p^I (\nabla T)_I - D_T^{II} C_p^{II} (\nabla T)_{II}] \cdot \hat{n}$, where the left-hand side is the rate at which heat is generated at the interface, and the right-hand side is the rate at which this heat flows into the bulk phases on either side.

The first of the equations (6.9) assumes the surface tension to be the crucial stabilizing force. It is derived from the Gibbs-Thomson relation, which relates the pressure difference across an interface to the surface tension. The more usual form of this equation is

$$P_I - P_{II} = \sigma \kappa, \quad (6.12)$$

where P_I and P_{II} are the pressures on either side of the interface. The difference in pressure along the phase coexistence line is related to the temperature difference via the Clausius-Clapeyron equation

$$\frac{\Delta P}{\Delta T} = \frac{\Lambda}{T_m \Delta \mathcal{V}} \quad (6.13)$$

Evidently, T_m is the melting temperature, and $\Delta \mathcal{V}$ the difference in specific volume between the two phases. In linear approximation we replace the pressure difference

¹Similar approximations will be made later for the thermal diffusion coefficients $D_T^{I(II)}$ and densities $\rho_{I(II)}$ of two phases: the qualitative errors resulting from these approximations have no effect on the results of the theory.

²The curvature of the interface κ is understood to be positive if the center of curvature lies on side I of the interface, that is, if the phase I bulges into the phase II .

by (6.12) and the temperature difference by $T - \Lambda/C_p$. We arrive at (6.9) with the following expression for the capillary length

$$d_0 = \frac{\sigma T_m C_p}{\Lambda^2} \quad (6.14)$$

For an interface which almost coincides with the x -axis, the equation can be linearized by Fourier transforming the displacement ξ

$$\xi = \xi_k e^{ikx + \omega t} \quad (6.15)$$

and the temperature near the interface

$$\bar{v} = \bar{v}_k e^{ikx + \omega t \pm qy} \quad (6.16)$$

with the $\pm$ signs corresponding to the opposite sides of the interface. The three equations then become (Harris, Jörgenson and Grant [92])

$$\bar{v}_k = -d_0 k^2 \xi_k \quad (6.17)$$

$$D_T(-k^2 + q^2) = \omega \quad (6.18)$$

$$\omega \xi_k = 2D_T q \bar{v}_k \quad (6.19)$$

and can be solved to give the dispersion relation

$$\tau = -\frac{1}{\omega} = d_0^2 \left[(1 + k^2 d_0^2)^{1/2} + k d_0 \right] / (D_T k^3 d_0^3) \quad (6.20)$$

This expression for the relaxation time has two asymptotic forms: $1/2D_T d_0 k^3$ for $k \ll 1/d_0$ and $1/D_T k^2$ for $k \gg 1/d_0$. According to the discussion in Section 6.2 (see equation (6.7) for example) this defines the dynamic exponent z as $z = 2$ for early times and $z = 3$ late times, because short wavelengths dominate at early times and vice-versa. Thus, the roughening undergoes a transition from a $t^{1/2}$ regime to a $t^{1/3}$ regime at some crossover time τ_0 defined by the capillary length d_0

$$\tau_0 \sim \frac{d_0^2}{D_T} \quad (6.21)$$

As discussed in Harris, Jörgenson and Grant [92], the crossover behavior is a consequence of the competition between the rate of generation of latent heat at the interface and the rate of heat diffusion in the bulk phases. When, at early times,

the latent heat produced (absorbed) at the interface is efficiently diffused away, then the thermal fluctuations will grow unhindered with $z = 2$ (which, in the language of Hohenberg and Halperin [77], is the behavior of model-type A with a non-conserved order parameter). Then, at later times, the rate of heat generation is very much larger than the rate of heat diffusion. The relaxation then will be retarded by the coupling to the energy field and the interface will grow with $z = 3$. This corresponds to the so-called model-type C, namely, a system with a non-conserved order parameter (the magnetization) coupled to a conserved field (the energy). Thus, equation (6.20) describes both model A and model C situations and predicts the form of the crossover¹.

6.4.2 Including Hydrodynamics

We now proceed to the inclusion of hydrodynamic terms in the mechanism of surface evolution. In our simulations (Section 6.3) we detected the presence of coherent fluid flow along the interface (Figure 6.6) in late time of the evolution. As a first approximation, we assume this flow is inviscid and the fluid is incompressible so the liquid velocity then satisfies the Euler equation (2.2) on each side of the interface. However, we can use the linearized form of this equation

$$\frac{\partial \mathbf{v}_l}{\partial t} = -\nabla \frac{P_l}{\rho_l}; \quad l = I, II \quad (6.23)$$

because the non-linear term $(\mathbf{v} \cdot \nabla)\mathbf{v}$ is negligible compared to $\frac{\partial \mathbf{v}}{\partial t}$ if the width of the interface is small compared to the characteristic wavelength of the interface profile. Indeed, in a time τ the fluid particles travel an average distance W , the width of the interface. Their velocity v then is of order W/τ . This velocity varies along the interface with the characteristic wavelength of the interface profile. Thus, the gradient of velocity is of order $\frac{v}{\lambda}$ and the condition $(\mathbf{v} \cdot \nabla)\mathbf{v} \ll \frac{\partial \mathbf{v}}{\partial t}$ is equivalent to $\frac{1}{\lambda} \left(\frac{W}{\tau}\right)^2 \ll \frac{W}{\tau} \frac{1}{\tau}$, or $W \ll \lambda$.

¹The crossover time roughly correspond to the fluctuations with characteristic wavelength d_0 . Indeed, the two asymptotics of equation (6.20) correspond to the same time scale τ_0 when

$$\tau_0 = \frac{1}{D_T k_0^2} = \frac{1}{2D_T d_0 k_0^3} \quad (6.22)$$

or $k_0 = \frac{1}{2d_0}$. Then, $\tau_0 = \frac{4d_0^2}{D_T}$, as stated in (6.21).

The absence of the nonlinear term in the Euler equation permits the introduction of the velocity potential ϕ and the stream function ψ (Landau [95])

$$\mathbf{v}_\ell = \nabla \phi_\ell; \quad v_x^I = -\frac{\partial \psi_\ell}{\partial y}; \quad v_y^I = -\frac{\partial \psi_\ell}{\partial x}; \quad \ell = I, II \quad (6.24)$$

To show this we apply the operation $(\nabla \times)$ to both sides of (6.23) and arrive at $\nabla \times \mathbf{v}_\ell = 0$, which is the necessary condition for potential flow. Equation (6.23) then reads

$$\frac{\partial \phi}{\partial t} = -\frac{P_\ell}{\rho_\ell}; \quad \frac{\partial \psi}{\partial t} = 0 \quad (6.25)$$

When the inertia of the fluids is taken into account, it is the Gibbs-Thomson condition, equation (6.17), that must be modified. Not only does the pressure difference across the interface incorporate the surface tension, but, for it must also account for the motion of the two fluids. Instead of equation (6.12) we write

$$P_I - P_{II} = \sigma \kappa + \rho_I \frac{\partial \phi_I}{\partial t} - \rho_{II} \frac{\partial \phi_{II}}{\partial t} \quad (6.26)$$

As before, we introduce the temperature by writing $P_I - P_{II} = -\bar{v}/d_0$, and then linearize the equation by setting

$$\begin{aligned} \phi_I &= \phi_k e^{ikx + \omega t - ky} \\ \phi_{II} &= -\phi_k e^{kx + \omega t + ky} \end{aligned} \quad (6.27)$$

This manner of linearizing is based on the assumption of the presence of continuous convective flow along the interface as shown in Figure 6.7 (left), caused by moving interface, Figure 6.7 (right). The assumption was confirmed earlier in the numerical simulations, Figure 6.6. The minus sign in front of the expression for ϕ_{II} reflects the symmetry of the velocity fields in the two phases: at the point where ϕ_{II} is maximum (flows directed from this point), ϕ_I is minimum (flows directed to this point). In the reference frame of the leading edge of the interface, the phase *I* (*II*) fluid encounters the edge with velocity $\frac{\partial \xi}{\partial t}$ and turns sideways. Thus,

$$ik\phi_k = -\omega\xi_k \quad (6.28)$$

In other words, the velocity of fluid is out of phase with that of the interface. The modified Gibbs-Thomson equation becomes

$$\bar{v}_k = -d_0 k^2 \xi_k - id_0 \frac{\rho}{\sigma} \frac{\omega^2}{k} \xi_k \quad (6.29)$$

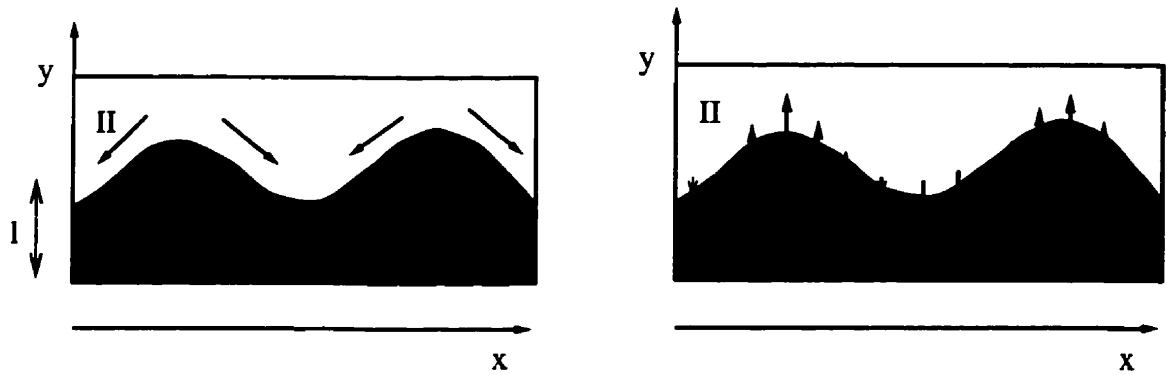


Figure 6.7: The fluid velocity $\nabla_x(\phi)$ along interface (left) and interface velocity $\frac{\partial \xi}{\partial t}$ (right)

where the approximation $\rho_I = \rho_{II} = \rho$ has been made. The solution to the linearized equations (6.29) has three distinct regimes, of which the first two have been identified in Section 6.4.1. The third, inertial, regime is obtained by including only the inertial term, not the Gibbs-Thomson term from equation (6.29). We then obtain

$$k^2 = -\frac{\omega^3 \beta^2}{D_T (\omega^2 \beta^2 + 1)} \quad (6.30)$$

with $2 d_0 D_T \rho / \sigma = \beta$. For late times $\omega^2 \beta^2 \ll 1$. Expanding the right hand side of equation (6.30) in powers of $x = \omega^2 \beta^2$ (or using the known formula $\frac{x}{1+x} = x - x^2 + x^3 - \dots$) and keeping the leading term only, we obtain the final dispersion relation

$$\omega^3 = -\frac{1}{\beta^2} D_t k^2 \quad (6.31)$$

This identifies the “slowing down” exponent as $z = \frac{3}{2}$ and the growth regime as $t^{3/2}$. This is a simplified illustration of the existence of $t^{3/2}$ growth regime. The result is confirmed by a more robust analysis of equation (6.29) (together with the boundary conditions (6.18) and (6.19)) in Appendix A.4.

6.4.3 Growth Regimes and Crossover Times

Figure 6.8 shows the cartoon of the different regimes for the growth of the width of the interface. There are three characteristic cross-over times. The first is τ_0 as defined by (6.21). The next is the crossover between the early time $t^{1/2}$ regime and the late time $t^{3/2}$ regime. The former is described by the dispersion relation $\omega = -D_T k^2$ (see equation (6.20) and discussion afterwards) and the latter by the relation (6.31). The crossover time corresponds to the situation where both these expressions make equal

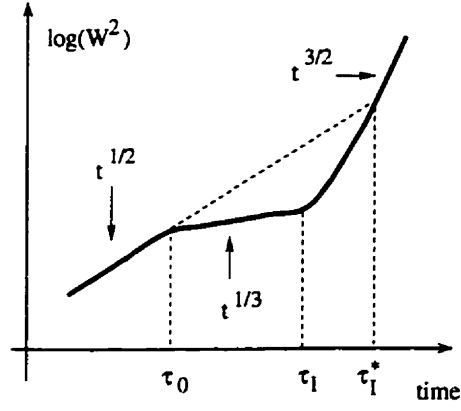


Figure 6.8: The three growth regimes as predicted by our theory.

contributions. Thus, eliminating k^2 from above dispersion relations determined the time

$$\tau_I^* \sim \beta = 2D_T d_0 \frac{\rho}{\sigma} \quad (6.32)$$

As we see from Figure 6.8 the onset of the $t^{3/2}$ regime happens at a time $\tau_I < \tau_I^*$. This time is obtained similarly from the dispersion relation $\omega = -2D_T d_0 k^3$ and the relation (6.31) yielding

$$\tau_I \sim \beta \left(\frac{\tau_0}{\beta} \right)^{\frac{1}{7}} \quad (6.33)$$

However, the crossover times (6.32) and (6.33) defined by time-scale β assume that flow coherent with the interface, Figure 6.7, is established from the very beginning of the simulations. In practice this is not the case, because of the finite viscosity of the fluid which is primarily responsible for the transfer of momentum within the fluid and the establishment of the flow.

By dimensional analysis, the effect of the viscosity in this case is characterized by an effective hydrodynamic time scale

$$\tau_h = l^2 / \nu \quad (6.34)$$

where l is a characteristic length defined by the geometry of the experiment, Figure 6.7, and ν is a viscosity. Thus, when $\tau_h > \beta$ expression (6.34) defines the crossover to the inertial $t^{3/2}$ regime.

6.5 Numerical Estimates

To make the connection of our numerical results (Section 6.3) with our theoretical predictions (Section 6.4.2) we must evaluate the properties of the bulk phases and the interface that are relevant to the evaluation of the cross-over times (6.21), (6.33) and (6.34).

6.5.1 Thermodynamic and Transport Properties of the Bulk

Table 6.1 summarizes the thermodynamic and transport properties of the two phases used in the roughening experiment. The average density and magnetization values in each phase are measured directly from the sample data. Temperature was measured from the distribution of demons, Figure 5.6. The thermal diffusion coefficients D_T were estimated from structure function measurements in Section 5.7. Also, from the structure factor we use the estimation of the ratio of specific heats γ (5.32) and the relation (3.64) to determine the estimate for the viscosity.

The specific heat c_V at constant volume is estimated in mean-field approximation as:

$$C_V = \rho \frac{\partial E}{\partial T} \big|_{\{N,V\}} = \rho \frac{\partial E}{\partial m} \frac{\partial m}{\partial T} \big|_{\{N,V\}} = \rho \frac{(\rho z J m + \Delta)^2}{T \left(\frac{T}{1-m^2} - \rho z J \right)} \quad (6.35)$$

and the specific heat at constant pressure is $C_P = \gamma C_V$. The entropy per particle S_N is estimated from the mean-field expressions (5.14) and (5.15). We note, that for the entropy in the HD phase the mean-field gives a negative value: this should cause no concern, as discussed in Section 5.6.1. In fact, we are interested in the difference in entropy of the two phases which determines the latent heat (Section 6.5.2).

phase	ρ	m	T	C_V	C_P	S_N	D_T	Γ	$\nu + \xi$
$I(\text{LD})$	3.11	-0.918	13.	0.69	2.0	0.70	0.08	0.16	0.16
$II(\text{HD})$	5.61	0.966	13.	3.48	10.0	-0.63	0.06	0.08	0.04

Table 6.1: Bulk properties of the two phases in the roughening experiment.

6.5.2 Interface Parameters

The interface is characterized by the surface tension σ , the latent heat Λ , and the capillary length d_0 . We estimate the surface tension of a *sharp interface* in mean-field

approximation. In the hexagonal arrangement, Figure 6.9 (left), each particle that

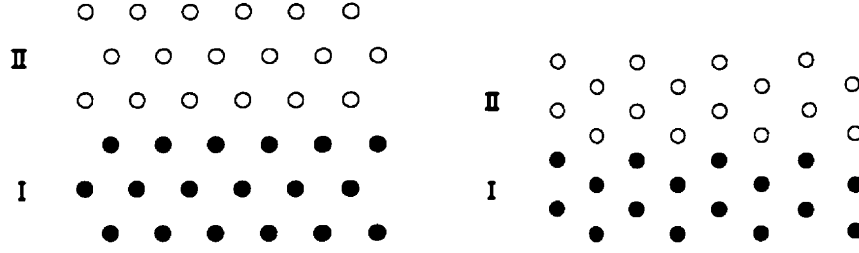


Figure 6.9: Two different arrangement of sharp interface: left picture shows the interface parallel to nearest neighbor line, right picture shows interface parallel to next-nearest neighbor line

sits on the interface line has two near-neighbor links to the sites of opposite phase (broken bonds) and four same-phase links. The averaged exchange energy E_1 of two sites on opposite sides of interface is

$$E_1 = -\frac{1}{2}J\rho_I m_I(4\rho_I m_I + 2\rho_{II} m_{II}) - \frac{1}{2}J\rho_{II} m_{II}(4\rho_{II} m_{II} + 2\rho_I m_I) \quad (6.36)$$

If the two sites are in the bulk, their combined energy E_2 is

$$E_2 = -\frac{1}{2}J6(\rho_I m_I)^2 - \frac{1}{2}J6(\rho_{II} m_{II})^2 \quad (6.37)$$

The difference between these two energies gives an estimate of the surface tension

$$\sigma = \frac{\Delta E}{a} = \frac{1}{a}(E_2 - E_1) = J(\rho_{II} m_{II} - \rho_I m_I)^2 \simeq 68 J \quad (6.38)$$

where we have used the measured values of density and magnetization from Table 6.1 and $a = 1$ is the lattice spacing. For the interface, parallel to the next-nearest-neighbor line (Figure 6.9 right), the estimate is similarly straightforward. It yields an expression for the surface tension that differs from (6.38) by a factor of $\frac{2}{\sqrt{3}} \simeq 1$ (within acceptable limits for our “rough” estimate).

The latent heat in mean-field is estimated per particle as

$$\Lambda = T(S_{II}\rho_{II} - S_I\rho_I)/N \simeq 70 J \quad (6.39)$$

The value of the capillary length (6.14) is then¹

$$d_0 \simeq 1 \quad (6.40)$$

¹We use the averaged specific heat: $C_p^I \simeq C_p^{II} \simeq C_p = 6$

6.5.3 Crossover Times

The above data result in the following estimations of crossover times (6.21), (6.32) and (6.34)

$$\tau_0 \simeq 10 \quad (6.41)$$

$$\tau_I^* \simeq 0.1 \quad (6.42)$$

$$\tau_h \simeq 10^4 \quad (6.43)$$

In last estimate we used the viscosity estimate $\nu \simeq 0.1$ (see Table 6.1) and the characteristic length $l \simeq 64$, the width of the HD phase. Because $\tau_h > \tau_I^*$, the estimate (6.43) is the crossover time to the $t^{3/2}$ regime.

We now compare the two crossover times, (6.41) and (6.43), with the result of numerical simulations, as estimated from data in Figure 6.3.

The estimate for the first crossover time, (6.41), is lower than the value observed in the simulations, which is $\sim 10^2$. However, the difference is small enough to be related to the accumulation of errors in the estimate of the capillary length (6.14) which depends on quantities that are evaluated in mean-field.

Our estimate of the crossover time to the inertial $t^{3/2}$ regime, $\tau_h \simeq 10^4$, agrees well with the results of the simulation for the sample of size 1024×128 (Figure 6.3).

We note, that expression (6.34) predicts different crossover times to inertial regime for different interface-to-interface distances l . We have tested this dependence in samples of different vertical sizes $L_y = 2l$. Figure 6.10 shows the superimposed results for the growth of the interface for three samples of different sizes¹: 1024×64 , 1024×128 and 1024×256 . The approximate location of crossovers, marked by arrows in Figure 6.10, complies with a quadratic dependence of the crossover time on the vertical size of the sample

$$\frac{\tau_h(L_y = 256)}{\tau_h(L_y = 128)} \simeq \frac{\tau_h(L_y = 128)}{\tau_h(L_y = 64)} \simeq 4 \quad (6.44)$$

We have not found significant difference in the growth of the interface for different horizontal sizes L_x .

¹In order to effectively simulate systems of large size ($V > 500,000$) we adapted our numerical code to be run on the Beowulf cluster belonging to the Centre for the Physics of Materials.

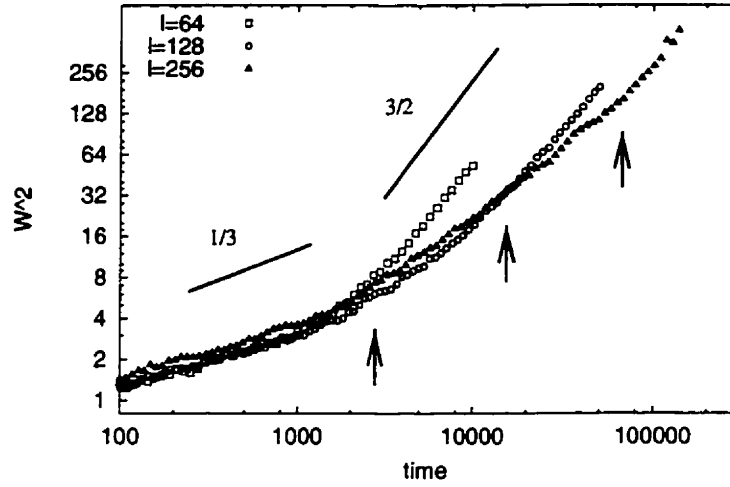


Figure 6.10: The interface growth for samples of three different sizes. The solid lines, guides to the eye, have slopes $1/3$ and $3/2$ respectively. Arrows indicate the approximate location of crossover to inertial $t^{3/2}$ regime for each sample.

Further, because of (6.34), the flow along the interface must *first* establish itself in the phase of higher viscosity. This indeed is observed in the numerical study of the velocity field, Figure 6.6.

There is another way in which this data seems to be consistent with conventional hydrodynamics. To characterize the stability of the flow of fluid in the inertial regime we evaluate the Reynolds number, defined as

$$R = \frac{\tau_l}{\tau_h} \quad (6.45)$$

where the effective time of laminar flow $\tau_l = l/u$ is determined by the velocity u of flow and τ_h is defined in (6.34). For our estimate we use the fact that the velocity of flow is of order of the averaged velocity of the interface. From the data, shown in Figure 6.3, for example, we noticed that between times $t = 30,000$ and $t = 50,000$ the width of the interface changes from $W^2 = 78$ to $W^2 = 175$. Hence $u \simeq 10^{-4}$ and $\tau_l \simeq 10^5$ where the characteristic length is $l = 64$. Thus, $R \sim 10$, which characterizes the flow as stable and non-turbulent (Landau [95]). This is certainly consistent with our observations.

In summary:

- the $t^{3/2}$ growth regime is the result of stable laminar flow of fluid along the

interface. This flow affects the growth of the interface because it alters the pressure close to the interface.

- the flow along the interface results from specific boundary conditions: as the interface grows, it couples to the motion of the fluid between two interfaces, distance l apart. The time needed for this fluid flow to be established is in turn determined by the effective time (6.34) for the transfer of momentum across this distance in the viscous fluid.

General Conclusions

In the present thesis we have investigated a new class of microscopic models for two-dimensional fluid systems with well defined thermal properties. Taking the Lattice Gas Cellular Automata approach as a starting point, we developed a new approach that is better suited for simulating thermal fluids in equilibrium and non-equilibrium. Our modifications of the LGCA method include:

- The removal of requirements for the Fermi-exclusion principle on occupation numbers which is present in all original LGCA models. The advantage is a better fine-tuning of the models' behavior when dealing with a greater range of densities. As well, the greater number of allowed collisions for less restrictive statistics allows us to recover the hydrodynamic behavior more easily. In terms of computer implementation, the code spends less time dealing with collisions that are not allowed to happen.
- Allowing the particles that constitute our models to have an internal degree of freedom, a potential energy, by means of the introduction of a set of discrete energy levels. Again this leads to enhanced collision rates. More importantly, this modification allowed us to model the evolution of the systems using Monte-Carlo methods and, hence, to transfer the properties of simple LGCA models of hydrodynamics to the more interesting thermal models.

We have studied the advantages and implications of the above modifications both theoretically and numerically. We analyzed the transport properties of the new models using the Chapman-Enskog method and linear response theory and obtained generalized expressions for all the transport coefficients.

The first particular model investigated, Section 4.2, was studied analytically to illustrate the new features developed earlier. In our second example, Section 4.3, we build a minimal model within the Lattice Boltzmann scheme for studying a non-equilibrium phenomenon, Rayleigh Bénard convection. In this simulation we illustrated the advantages of Monte-Carlo methods for time-evolution: the use of the multidemon algorithm effectively simulated the necessary local temperature distribution.

The main advantage of the new approach, is the possibility of including the interactions between particles (in a straightforward way) to study phase-separating fluids. We have built a model which is a combination of a lattice-gas automaton and a modified Ising lattice gas that can model a two-phase fluid with a 1st order phase transition and a latent heat. The Monte-Carlo dynamics allow us to define temperature in a natural way. For appropriate parameters, we studied the the phase-separation of the fluid into the regions of low and high density.

The model was used to simulate the non-equilibrium phenomena of two-phase fluids with latent heat. In particular, the relaxation of the interface towards equilibrium was simulated and a new scaling law for the averaged width of interface was observed. We explained the origin of this scaling and showed that it is a consequence of our specific boundary conditions, *i.e.* the geometry of the experiment.

There would seem to be many possible further applications of our approach for the two-phase fluid with latent heat in particular and for the generalized lattice gas in general. Our model with interactions (Chapter 5) has many advantages for the study of nonequilibrium phenomena in fluids. Special interest is presented by the simulation of spinodal-decomposition, the nucleation of the droplets of a new phase in an under-cooled fluid. Ongoing research in this area (Grant and Elder [99], for example) concerns the effects of hydrodynamics on such nucleation. To simulate this phenomena with our model we would need to work in a different statistical ensemble: the traditional constant $\{N, V, E(T)\}$ ensemble is not appropriate since the nucleation of a new phase changes the density of the under-cooled fluid. Nevertheless, a Monte-Carlo method (Frenkel and Smit [96]) that keeps the chemical potential constant (by insertion of particle at boundaries at a rate consistent with thermodynamics) is

readily adapted to our model.

Another area of nonequilibrium physics that can be studied with our model is the solidification phenomenon: we have done some preliminary work in this direction. A simple modification of the evolution algorithm involves changing the mobility of a particle depending on its spin-value. For example, we can modify our model so that spin-up particles cannot propagate to new locations. The immobile $m \sim +1$ phase is then characterized as the solid phase. Figure 7.1 shows the time evolution of a nucleated solid phase in such an under-cooled fluid: we observe the development of a “snowflake”. However, more theoretical work needs to be done to properly

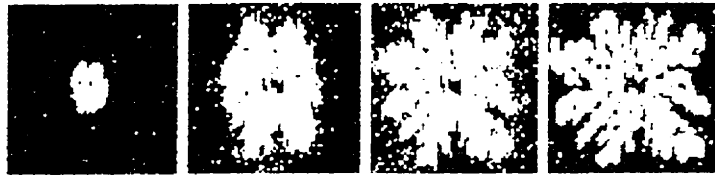


Figure 7.1: Nucleation of snowflake. The time-sequence is: 50, 500, 1000, 2500 MCSs. Grey color shows the low-density (LD) (negative magnetization) fluid phase, white color is the solid phase.

characterize the two phases in this model of solidification.

Appendices

A.1 Hexagonal Lattice

This appendix illustrates notation for the two-dimensional hexagonal lattice and the convention for the size of the lattice. Throughout the thesis the hexagonal lattice of size $L_x \times L_y$ means L_x sites along the line of second-nearest neighbors, x , and L_y sites along the direction perpendicular to this, as shown in Figure A.1. Thus, in terms of lattice constant $a = 1$, the area A covered by a lattice of size $L_x \times L_y$ is

$$A = L_x L_y \sqrt{3} a^2 \quad (\text{A.1})$$

Since there are $L_x \times L_y \times 2$ sites in this area we obtain the area per site as $\sqrt{3}/2a^2$.

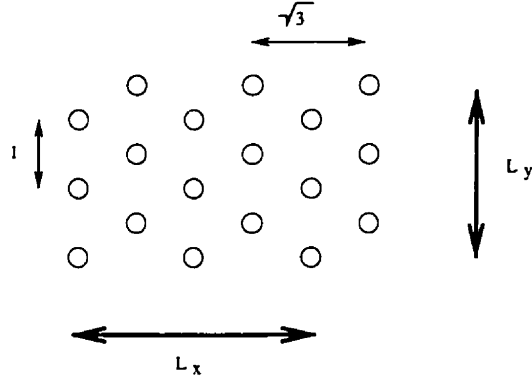


Figure A.1: The two-dimensional Hexagonal Lattice. Here $L_x = 3$ in units of $\sqrt{3}$ and $L_y = 3$ in units of 1 lattice spacing.

A.2 Temperature Measurements in Multi-Demon Algorithm

The multi-demon algorithms (Creutz [86]; Harris and Grant [88]; Harris, Jörgenson and Grant [92]) do not utilize the temperature as an input parameter. Like in molecular dynamics calculations, the temperature is found from analyzing the configurations of the system. For example, the average kinetic energy of a given molecule should

be $\frac{1}{2}T$ per degree of freedom, because of the equipartition theorem. In our multi-demon algorithm, each site is tied to a so-called demon variable E_D . Demons serve to transfer energy to/from neighboring sites in two ways: 1. through the intrinsic Ising interaction (5.5), and 2. via propagating particles¹. As mentioned in Section 3.2, because the demons are in local thermal equilibrium with the system, their energies will become exponentially distributed for energies small compared with the local system energy². This allows, in certain cases, the determination of the inverse temperature from the average value of the demon's energy. We show this first for a simple Ising model (one spin at a site) with $\Delta = 0, U = 0$. We first write the partition function for a single demon:

$$Z_i = \sum_{E_D=0}^{\infty} e^{-E_D/T} \quad (\text{A.3})$$

where the demon energy ranges from 0 to ∞ in increments of q ; the increment q is the smallest possible exchange of energy between a demon and system and is equal to $2J$. Since demons are non-interacting, the partition function for the entire system of demons is a multiple of the one-demon partition function $Z_D = Z_i^V$. From standard canonical ensemble analysis we obtain the expression for the average energy of the demons

$$V \langle E_D \rangle = V \frac{\partial}{\partial \frac{1}{T}} \frac{1}{T} \ln Z_i = V \frac{q}{e^{q/T} - 1} \quad (\text{A.4})$$

When q/T is small we can expand $e^{(q/T)}$ to first order and (A.4) becomes

$$\langle E_D \rangle \simeq T \quad (\text{A.5})$$

In other words, measuring the average demon energy at any site means measuring local temperature. The more general case of an Ising model with $\Delta > 0$ for was con-

¹The algorithm effectively simulates the micro-canonical sum

$$\Theta = \sum_C \sum_{E_D} \delta(H(s) + K(P) + E_D - E_{fixed}) \quad (\text{A.2})$$

where s denotes the configurational spin state of the lattice with energy given by (5.5), $K(P)$ is the kinetic energy associated with the momenta P , $\delta()$ denotes the delta-function and E_{fixed} is an initially determined total energy.

²Thus, if we observe deviations from exponential distribution in our measurements, the low-energy part of distribution must be used for temperature determination. This argument was used in Section 5.5.

sidered by Jörgenson [92]. This was done by separating the non-interacting demons into two groups, those associated with up-spins and those associated with down spins. Jörgenson [92] showed that the field Δ introduces a bias into the definition of temperature from averaged demon's energy: $\langle E_{D_\uparrow} \rangle - \langle E_{D_\downarrow} \rangle \sim 2\Delta$. Similarly, in the case of our model with moving spins and $U > 0$ we should consider separately rather large number of groups: those demons that serve sites with 1 particle at a site, those on sites with 2,3,..., etc particles. Clearly, the analysis gets complicated in this case.

A.3 Numerical Algorithm for Interface Recognition

In this Appendix we describe the pattern-recognition algorithm used in our numerical analysis of interfaces in the simulations that are discussed in Section 6.3. As mentioned in Section 6.2, the interface can be completely defined by a so-called displacement variable $\xi(x, t)$ for each of the two interfaces in our roughening experiments¹. The x-axis is parallel to the flat interface at $t = 0$ and in our initial configurations it is parallel to a line connecting second-nearest neighbors, as in Figure A.1. The displacement variable is a single-valued function of x for times when overhangs have not developed.

In our lattice model, x is a discrete variable. As is seen from Figure A.1, it corresponds to a column of sites. The columns are separated by $\sqrt{3}/2a$ where $a = 1$ is the lattice constant. Thus for each column x we scan the sites belonging to this column to determine the location (or y-coordinate) of the *maximum gradient of phase parameter* ϕ (either magnetization or density). To reduce the noise, the gradient $\nabla\phi$ at any point y is calculated as follows:

$$\nabla\phi(x, y) = \sum_i^W (\phi(x, y+i) - \phi(x, y-i)) \quad (\text{A.6})$$

We set $W = 6$. Let $\phi_I < \phi_{II}$, where I and II refer to two different phases. Then, the location of both interfaces, $y1(x)$ between phases I and II , and $y2(x)$ between phases II and I , is determined from the following numerical procedures:

$$\nabla\phi(x, y1(x)) = \max \{ \nabla\phi(x, y) \}; \quad y = 1..L_2 \quad (\text{A.7})$$

$$\nabla\phi(x, y2(x)) = \min \{ \nabla\phi(x, y) \}; \quad y = 1..L_2 \quad (\text{A.8})$$

¹We need two flat interfaces in order to build a two-phase sample with periodic boundary conditions.

Further subtracting the common reference point for each of the interfaces yields the displacement variable $\xi(x) = y(x) - \langle y(x) \rangle$, see Figure 6.1.

A.4 Dispersion Relation for the Full Model

In this appendix we analyze in more (analytical and numerical) details the modified Gibbs-Thomson equation (6.29) of Section 6.4.2. To obtain the general dispersion relation (with both surface tension term and inertial term) we proceed as follows. Let us write again the system of equations that has to be solved:

$$v_k = -d_0 k^2 \xi_k - i d_0 \frac{\rho}{\sigma} \frac{\omega^2}{k} \xi_k \quad (\text{A.9})$$

$$D_T(-k^2 + q^2) = \omega \quad (\text{A.10})$$

$$\omega \xi_k = 2 D_T q v_k \quad (\text{A.11})$$

First, we substitute ξ_k from (A.11), $\xi_k = 2 \frac{D_T q v_k}{\omega}$, into (A.9) and solve for q :

$$q = -\frac{1}{2d_0 D_T \frac{k^2}{\omega} + i\beta \frac{\omega}{k}} \quad (\text{A.12})$$

where, again, $\beta = 2d_0 D_T \frac{\rho}{\sigma}$. Then we substitute (A.12) into (A.10) to obtain the dispersion relation in general form

$$D_T \left(-k^2 + \frac{1}{(2d_0 D_T \frac{k^2}{\omega} + i\beta \frac{\omega}{k})^2} \right) = \omega \quad (\text{A.13})$$

Let us check the asymptotics of (A.13) for large and small k :

$$D_T \left(-k^2 + \frac{1}{4d_0^2 D_T^2} \frac{\omega^2}{k^4} - i \frac{\rho}{2\sigma d_0^2 D_T^2} \frac{\omega^4}{k^7} + \mathcal{O}\left(\frac{1}{k^{10}}\right) \right) = \omega, \quad k \rightarrow \infty \quad (\text{A.14})$$

$$D_T \left(-k^2 \left(1 + \frac{1}{\omega^2 \beta^2} \right) - i \frac{2\sigma}{\beta^2 \rho} \frac{k^5}{\omega^4} + \mathcal{O}(k^8) \right) = \omega, \quad k \rightarrow 0 \quad (\text{A.15})$$

The leading terms of (A.14) yield the dispersion relation (6.20) and the leading term of (A.15) yields the dispersion relation (6.30).

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