

An inventory of Lattice Boltzmann models of multiphase flows

Erik Aurell^{1,2,3} and Minh Do-Quang²

30th August 2017

¹ SICS, Swedish Institute of Computer Science AB, SE-164 29 Kista, Sweden

² Dept. of Numerical Analysis and Computer Science, Kungliga Tekniska Högskolan, SE-100 44 Stockholm, Sweden

³ NORDITA, Blegdamsvej 17, DK-2100 Copenhagen, Denmark

Abstract

This document reports investigations of models of multiphase flows using Lattice Boltzmann methods. The emphasis is on deriving by Chapman-Enskog techniques the corresponding macroscopic equations. The singular interface (Young-Laplace-Gauss) model is described briefly, with a discussion of its limitations. The diffuse interface theory is discussed in more detail, and shown to lead to the singular interface model in the proper asymptotic limit. The Lattice Boltzmann method is presented in its simplest form appropriate for an ideal gas. Four different Lattice Boltzmann models for non-ideal (multi-phase) isothermal flows are then presented in detail, and the resulting macroscopic equations derived. Partly in contradiction with the published literature, it is found that only one of the models gives physically fully acceptable equations. The form of the equation of state for a multiphase system in the density interval above the coexistence line determines surface tension and interface thickness in the diffuse interface theory. The use of this relation for optimizing a numerical model is discussed. The extension of Lattice Boltzmann methods to the non-isothermal situation is discussed.

Key words: Multiphase flows, Lattice Boltzmann methods, Chapman-Enskog expansions, diffuse interface theory, Cahn-Hilliard theory

Work supported by the Parallel Scientific Computing Institute (PSCI) at NADA/KTH as project 24082-61206.

Preface

This document reports on a project carried out by the authors in March-August, 2000. While waiting for renewed funding the results were not properly written up, but left in the form found here. It now being clear that the project has definitively been discontinued, the unfinished report is deposited as e-Print as is, in the hope that some of it may be of some use to someone somewhere.

As a short guide, the paper contains first a survey of the diffuse interface models, also known as Cahn-Hilliard models. None of this is new, but is here collected in one place. It is pointed out that two different versions of the diffuse interface theory are used in the literature. The paper thereafter contains an introduction the Lattice Boltzmann equations along standard lines, and then a rederivation of the model of Swift et al. Again, nothing here is new, but the calculations are spelled out in more detail than in Swift et al.

The paper then contains a survey of other proposed LBE models (isothermal), chapters 5–7, where we reach different results than in the published literature. In fact, we find that one such model not only does not give the correct momentum equation, but even displays a spurious density diffusion term in the continuity equation! On the other hand, we find that one other model presented in the literature actually gives the correct continuity and momentum equations.

Section 8 contains a discussion of thermal multiphase Lattice Boltzmann equations. The investigations reported are not conclusive, and probably along the wrong path. The general setting, and the references, could however be of use. We believe that constructing thermal multiphase Lattice Boltzmann equations is a difficult problem. We doubt it can be done in the standard simplifying single relaxation-time approximation.

Section 9 contains a discussion of optimization issues for the model described in section 7. By changing the equation of state in the interface region, one may change any of either three properties, keeping the others constant. The properties are the characteristics of the stable phases, the surface tension and the interface thickness. If, for instance, one wants to simulate an interface on a given grid, then the interface thickness in the model must be significantly larger. Hence, by changing only interface thickness, one could simulate multiphase fluids with given density contrast and surface tension on different grids. The numerical results do not quite illustrate this fact, which we nevertheless believe should be important in practical use.

The work in sections 4 and 5 were done in collaboration with Massimo Vergassola, Observatoire de Nice, to whom we direct our heartfelt thanks for patient instruction and very useful advice on the calculations.

Contents

1	Introduction	4
2	Models of multiphase flows	6
2.1	The singular interface models	6
2.2	The diffuse interface theory: statics	7
2.3	The diffuse interface theory: dynamics	8
2.4	Derivation of the singular interface theory	11
3	Lattice Boltzmann equation models in brief	12
4	Non-equilibrium thermodynamics model	17
4.1	Method of successive approximations	18
4.2	Summary	24
5	Luo model: LBE for non-ideal dense gases	25
5.1	Chapman-Enskog analysis	26
5.2	Summary	31
6	Intermolecular interaction model	32
6.1	Summary	35
7	An LBE scheme with high order accuracy	36
7.1	Summary	41
8	Thermal lattice Boltzmann	42
8.1	General considerations	42
8.2	Non-isentropic Lattice Boltzmann	44
8.3	An attempt to generalize section 7	44
9	Optimizing a diffuse interface model	50
9.1	Summary	53
10	List of symbols and conventions used	55

1 Introduction

Multiphase flows are difficult because they involve thermodynamics (co-existing phases) kinetics (nucleation, phase transitions), and hydrodynamics (inertial effects). The nature of a liquid-vapour interface is still partly an open problem from the physical point of view. Mathematical descriptions of a multiphase flow have hence the status of models, and can be classified as i) singular interface models, which go back to Young, Laplace and Gauss; ii) diffuse interface models, which were first developed by Maxwell, Gibbs, Van der Waals [47] and Korteweg [23], and in a more modern setting by Cahn & Hilliard [6], and iii) fully detailed statistical mechanics models, which are still under development [15].

In numerical work the most common are methods based on the first kind of models, mainly because they do not require the resolution of the interface, and can therefore be simulated with less effort. We begin below with a brief discussion of such models, and in which situations they are likely to be inadequate. We then turn to the diffuse interface theory, from which one can derive the singular interface theory as an asymptotic limit.

If one is only interested in phenomena where the singular interface model is expected to be correct, the diffuse interface theory can, from the computational point of view, be considered a method of solving a moving boundary problem by solving a corresponding set of reaction-diffusion equations. The difference between a purely numerical procedure and the diffuse interface theory is that the latter is also a continuum thermodynamic model of the interface, and can therefore be compared with a wider set of data.

Lattice Boltzmann methods are computational schemes to solve macroscopic equations by solving the equations of a microscopic world, which is described by the desired macroscopic equations in an asymptotic limit. An review of early work in the area is [3], for a comparatively recent review, see [10]. If the microscopic world would be the real world, we would have a method akin to Molecular Dynamics, inevitably an expensive and cumbersome way to solve the macroscopic equations. What makes Lattice Boltzmann an interesting method is that we have freedom in choice of the microscopic model. In fact, we can make a rather drastic departure from the real world, and consider a microworld where velocities only take values in a discrete set. If such a model is correctly set up, the resulting macroscopic equations will still be the desired ones.

From the PDE standpoint, Lattice Boltzmann methods would seem to be wasteful, since they require resolution of scales smaller than hydrodynamic. In fact, however, Lattice Boltzmann methods do not require resolution of very small scales, and should rather be thought of as finite difference schemes with auxiliary variables. In this regard Lattice Boltzmann methods are superior to Lattice Gas models, which use Boolean variables, and require an additional averaging, and hence a significantly larger scale separation. The fluctuations on the other hand make Lattice Gases advantageous for some thermodynamic stability problems, such as those recently investigated by Boghosian and coworkers [9].

The competitiveness of Lattice Boltzmann methods with standard PDE methods – at present, and in the future – will mostly be determined by how large an overhead the extra variables in Lattice Boltzmann represent, and by the overall versatility and stability of the method. We will not attempt such a comparison in this report. A fair summary of the literature is that the opinion is still divided, but that Lattice Boltzmann methods have so far mainly been used in the physics community, where the interpretation in terms of a fictitious microworld is considered more of an advantage than a draw-back. A related point of view is that Lattice Boltzmann methods are closer to the physics, in the same sense as kinetics is compared with hydrodynamics. In flows with complicated physics the hydrodynamical descriptions may not have been worked out, or may be unfamiliar, and in both cases using Lattice Boltzmann methods could then be a more effective solution. The example usually given in this context is flow in porous media [10].

Given the reasons why Lattice Boltzmann methods for multiphase could be interesting, it still remains to construct such models. In this report we will go through models proposed in the literature, and find them lacking in several respects. Some of this work, presented below in sections 4-6 repeats calculations in the literature, while some of it is new. In section 7 we show that one of the schemes proposed in the literature on the other hand gives correct continuity and momentum equations for an isothermal multiphase flow. The construction involves an auxiliary quantity, and an expansion which mixes orders for this auxiliary quantity. In section 8 we show that the model of section 7 can be generalized to give non-isothermal multiphase flows. We note that other Lattice Boltzmann models for non-isothermal flows have been introduced in the literature, but do not review them in detail. In section 9 we discuss optimizing issues by modifying the free energy density in the interface region.

2 Models of multiphase flows

2.1 The singular interface models

In the singular interface model a surface is prescribed separating the two phases. The free energy includes a term proportional to the area of the surface, and spatially located with support on the surface, hence the name singular interface model.

For definitiveness, assume that the fluid obeys Navier-Stokes equations in both phases, and that the interface is an impermeable separating surface. A necessary boundary condition is then that the normal velocity components are continuous across the interface

$$\vec{v} \cdot \hat{n}|_- = \vec{v} \cdot \hat{n}|_+ = v_n \quad (2.1)$$

If the fluids are viscous we also have that the tangential components are continuous:

$$(\vec{v} - (\vec{v} \cdot \hat{n})\hat{n})|_- = (\vec{v} - (\vec{v} \cdot \hat{n})\hat{n})|_+ \quad (2.2)$$

If the velocity gradients at the interface vanish, so that the interface and its surroundings are locally at rest, and if the surface tension does not depend on the position along the surface, then there is a pressure jump across the interface according to Laplace's law

$$P_+ - P_- = 2\sigma\mathcal{K} \quad (2.3)$$

where σ is the surface tension and \mathcal{K} is the mean curvature of the surface, oriented such that the $+$ side is on the inside and the $-$ side is on the outside. If the fluid is not at rest, to the right hand side of (2.3) should be added the difference of viscous stresses, and if σ depends on the position on the interface, e.g. through a dependence on temperature, there is a further term involving the gradient of σ (see [24], § 60). In general, singular interface models lead to free boundary problems with a stress balance at the boundary, and can include both heat and mass flow through the interface [14].

Singular interface models are likely to be incorrect if applied to processes on length scales similar to the thickness of the interface. The standard list of such phenomena is (see e.g. [2]):

- motion near a critical point, where the interface thickness is large;
- motion of a contact line between a liquid and its vapour along a solid wall, see also [42];

- changes in topology, such as coalescence of liquid droplets or breakup of bubbles;
- spontaneous formation of bubbles in overheated liquids (nucleation);
- spontaneous separation into thermodynamically stable phases after a rapid quench from a higher temperature, where only one phase is stable (spinodal decomposition).

The first three of these examples are from near-equilibrium statistical mechanics, while in the last two inertial effects are weak, at least initially. The physics of systems that are both far from equilibrium and coupled to macroscopic transport is not systematically known, but is likely to give rise to new phenomena. One example could be bubble formation (nucleation) in the presence of strong temperature gradients, as in a liquid close to a heated wall.

2.2 The diffuse interface theory: statics

In modern terms, the diffuse interface theory is a Landau mean-field theory. In the isothermal setting, as considered by Van der Waals and Cahn-Hilliard, the (Helmholtz) free energy functional is assumed to depend on density and density gradients as

$$\mathcal{F} = \int_V \rho f(\rho, T) + \frac{1}{2} K |\nabla \rho|^2 dV \quad (2.4)$$

where $f(\rho, T)$ is the bulk free energy density (per unit mass) of a homogeneous phase with density ρ at temperature T , and K is a function of ρ and T . For simplicity we will in the following discussion take K a constant.

A thermodynamically stable state at fixed mass is a minimum of the grand potential $\mathcal{A} = \mathcal{F} - \mu M$, where μ is the chemical potential per unit mass. If there is a single spatially homogeneous global minimum at $\bar{\rho}$, the free energy is

$$\mathcal{F} = V \bar{\rho} f(\bar{\rho}, T) \quad (\text{homogeneous phase}) \quad (2.5)$$

The thermodynamic pressure is $P = -\frac{\partial \mathcal{F}}{\partial V}|_{(M, T)}$ (derivative at constant mass and temperature). The chemical potential per unit mass is related to the free energy by $\mu = \frac{\partial \mathcal{F}}{\partial M}|_{(V, T)}$ (derivative at constant volume and temperature). Since $M = \bar{\rho} V$, (2.5) leads to

$$P = \bar{\rho}^2 f'(\bar{\rho}) \quad \mu = f(\bar{\rho}) + \bar{\rho} f'(\bar{\rho}) \quad (2.6)$$

Alternatively, if $\psi = \rho f$ is the bulk free energy per unit volume, then

$$P = \bar{\rho}\psi'(\bar{\rho}) - \psi(\bar{\rho}) \quad \mu = \psi'(\bar{\rho}) \quad (2.7)$$

If, however, the grand potential has more than one global minimum, i.e. if there are two or more coexisting phases, then the minimum of is found by a Euler-Lagrange variational equation:

$$\mu = (\rho f)'_{\rho} - K \nabla^2 \rho \quad (2.8)$$

In one dimension, this equation can be integrated once to give

$$(\rho f - \rho \mu) = \frac{1}{2} K |\partial_z \rho|^2 - P \quad (2.9)$$

We expect there to be solutions of these equations which consist of domains where ρ is almost constant, and equal to one of the minima, separated by flat interfaces. Wherever $\nabla^2 \rho$ can be neglected, the Lagrange multiplier μ is equal to $\rho f' + f$, which by (2.6) is the chemical potential. If density varies but in one direction, then (2.9) is generally valid, and when then $|\nabla \rho|^2$ can be neglected, the constant of integration P on the right hand side of is by (2.6) equal to the pressure. The thermodynamic rules that the chemical potential and the pressure in two coexisting phases in equilibrium are equal, can then be seen as conditions that the solutions of (2.8) and (2.9) are of the desired type.

If we multiply (2.8) with the gradient of ρ , we see that it is equivalent to

$$\partial_{\alpha} T_{\alpha\beta} = 0 \quad T_{\alpha\beta} = \rho(f - \mu)\delta_{\alpha\beta} - K \left(\partial_{\alpha} \rho \partial_{\beta} \rho - \frac{1}{2} |\nabla \rho|^2 \delta_{\alpha\beta} \right) \quad (2.10)$$

where T is a capillary stress tensor. In one dimension T has only one component, and (2.10) reduces to (2.9).

2.3 The diffuse interface theory: dynamics

A diffuse interface model of a multiphase flow is a set of equations for density (ρ), momentum density (ρu_{α}) and energy density per unit mass (ε), that reduces to the static case just discussed, if velocity is zero. These equations will be of the form

$$\partial_t \rho + \partial_{\alpha}(\rho u_{\alpha}) = 0 \quad (2.11)$$

$$\rho D_t u_{\beta} = \partial_{\alpha}(T_{\alpha\beta} + \sigma_{\alpha\beta}^{(v)}) \quad (2.12)$$

$$\rho D_t \varepsilon = \partial_{\alpha}((T_{\alpha\beta} + \sigma_{\alpha\beta}^{(v)})u_{\beta}) - \partial_{\alpha} q_{\alpha} \quad (2.13)$$

where $T_{\alpha\beta}$ is the reversible part of a stress tensor, $\sigma_{\alpha\beta}^{(v)}$ the hydrodynamic viscous stress tensor, and q an energy flux. Using the first (continuity) equation the left hand side of the two others can be rewritten $\partial_t(\rho u_\alpha) + \partial_\beta(\rho u_\alpha u_\beta)$ and $\partial_t(\rho \varepsilon) + \partial_\alpha(\rho \varepsilon u_\alpha)$. The energy density consists of two parts: a kinetic energy ($\frac{1}{2}\rho u^2$), and an internal energy density $\rho \varepsilon_I$. The equations (2.11), (2.12) (2.13) must be supplemented with an equation of state, and a constitutive relation for the energy flux, which according to [2] contains a heat flux, and a “non-classical” term of interstitial working:

$$q_\alpha = -\kappa \partial_\alpha T + K \rho (\nabla \cdot u) \partial_\alpha \rho \quad (2.14)$$

The appearance of the second term in the energy flux (but not in the entropy flux) can be traced back to an assumption by Van der Waals, that the gradient term in the free energy in fact stems entirely from the internal energy. It allows for some simplifications, in that the equations for the internal energy density ε_I and entropy density s read

$$\rho D_t \varepsilon_I = \partial_\alpha (\kappa \partial_\alpha T) - p (\nabla \cdot u) + \sigma_{\alpha\beta}^{(v)} \partial_\alpha u_\beta \quad (2.15)$$

$$\rho T D_t s = \partial_\alpha (\kappa \partial_\alpha T) + \sigma_{\alpha\beta}^{(v)} \partial_\alpha u_\beta \quad (2.16)$$

where p is a pressure, related to density and temperature by the equation of state (2.6). If $f[\rho, T, \nabla \rho]$ is the free energy functional density in (2.4) of a system at rest, $\varepsilon_I[\rho, T, \nabla \rho]$ the internal energy functional density, and $s(\rho, T)$ the entropy density, then $f = \varepsilon_I - Ts$. The equation of heat transport (equation (2.16)) is therefore not independent, but can be derived from (2.15), the equation of state and thermodynamic identities, see [24] § 49. The recent review [2] lists a number of problems in which diffuse interface models have been used successfully, for other recent papers, see [31] and [21].

Different dynamic diffuse interface models differ by the choice of q and the stress tensor T . Gibbs et al [48], who credit Lovett and Lebowitz & Percus, use for T the stress tensor in (2.10). This choice has the perhaps undesired feature that it is not entirely local, since it depends on the chemical potential μ : it assumes that some part of the fluid is at thermal equilibrium and at rest, so that μ can be measured there. Another choice is to use equation (2.8) to solve for μ , and substitute that expression in (2.10): this is the form used in e.g. [2]. We note that equation (2.8) holds for a fluid at rest, but not necessarily for a fluid in motion, compare (2.12).

We will not discuss further which of the most common forms in the literature gives the correct description of the dynamics of a diffuse interface. As a historic aside, it is however interesting to note that Korteweg in his 1901 paper [23] derives by

symmetry arguments the following more general form of the inviscid stress tensor ([23], p. 12, equation (20) includes also the viscous terms)

$$T_{\alpha,\beta} = \delta_{\alpha\beta} (p + a|\nabla\rho|^2 - c\nabla^2\rho) + b\nabla_\alpha\rho\nabla_\beta\rho + d\nabla_{\alpha\beta}^2\rho \quad (2.17)$$

with four undetermined coefficient functions (a , b , c and d) to describe the capillary forces. Korteweg's stress tensor reduces to the one of [48] if one takes p equal to $\rho(f - \mu)$, b equal to $-K$, a equal to $K/2$, and $c = d = 0$. Korteweg further derives a surface tension (compare (2.22)

$$\sigma = \int \left(b + \frac{\partial d}{\partial \rho} \right) \left| \frac{\partial \rho}{\partial z} \right|^2 dz \quad (2.18)$$

which shows that the diagonal components of the stress tensor do not show up in the surface tension.

We end this brief introduction by discussing the limits of validity of the Cahn-Hilliard free energy (2.4). If K is a constant, it follows that the surface tension decreases to zero along the critical line as $\sigma \sim (T_c - T)^\mu$ with $\mu = \frac{3}{2}$. The correct value of this critical exponent is however $\mu \approx 1.28$ [51], which reflects the fact that a Landau theory does not correctly describe this second order phase transition. To match the observed dependence of the thickness one may take K a parametric function of temperature and density. It is still however an open question if such a modified Cahn-Hilliard theory correctly describes the interface close to a critical point.

Along the coexistence line away from the critical point, the Cahn-Hilliard theory predicts a sharp interface, on the order of a few atomic diameters. This is in agreement with experiments, but nevertheless a conceptual problem, because if the interface is so sharp there is no reason why higher order terms should be relatively small. Since the singular interface theory can be derived from the diffuse interface theory, and since both have been applied to widely different flows, there should however be some meaning to truncating (2.4) after the first gradient term, perhaps as some sort of normal form. For a detailed discussion of gradient expansion resummation techniques of first principle statistical mechanics models, and of possible problems with the diffuse interface models which go outside the range of phenomena of interest to us here, see [15].

2.4 Derivation of the singular interface theory

For completeness we derive here in the simplest case a singular interface model from a diffuse interface theory.

If K is small and density gradients are close to zero everywhere but in a small part of the volume, then that part can be considered a fuzzy interface. Let us suppose that this interface passes through point P and is there approximated by a surface given parametrically by $z = \frac{1}{2}(r_1x^2 + r_2y^2)$. The two principal components of the curvature tensor (r_1 and r_2) need not have the same sign. Let us further assume that density is only a function of $z - \frac{1}{2}(r_1x^2 + r_2y^2)$. A unit vector normal to the interface is formed by the density gradient

$$\hat{n} = \frac{\nabla \rho}{|\nabla \rho|} = \frac{(-r_1x, -r_2y, 1)}{\sqrt{1 + r_1^2x^2 + r_2^2y^2}} \quad (2.19)$$

and points for small x and y approximately in the \hat{z} direction. The stress tensor reads

$$\sigma_{\alpha\beta} = \left(-P + \frac{1}{2}p_1\right) \delta_{\alpha\beta} - p_1 \hat{n}_\alpha \hat{n}_\beta \quad (2.20)$$

where $P = \rho(\mu - f)$ is the thermodynamic pressure in a homogeneous phase at density ρ , and p_1 is $K(\frac{\partial \rho}{\partial a})^2$, evaluated at $a = z - \frac{1}{2}(r_1x^2 + r_2y^2)$.

On the z axis the only non-zero component of the force field $\partial_\alpha \sigma_{\alpha\beta}$ is in the z direction:

$$\partial_\alpha \sigma_{\alpha\beta}(0, 0, z) = \hat{z} (-\partial_z P - \partial_z p_1 + p_1(r_1 + r_2)) \quad (2.21)$$

On the other hand, we know that at rest $\partial_\alpha \sigma_{\alpha\beta} = 0$. If z_0 and z_Q are two points on the z axis on opposite sides of the interface we therefore have

$$0 = \int_{z_0}^{z_Q} \partial_\alpha \sigma_{\alpha\beta}(0, 0, z) dx_\beta = -(P(z_Q) - P(z_0)) + (r_1 + r_2) \int_{z_0}^{z_Q} K |\partial_z \rho|^2 dz \quad (2.22)$$

The pressure difference is proportional to the mean curvature ($\frac{r_1+r_2}{2}$) and to the gradient contribution to the free energy in (2.4). We can therefore identify that term as a free energy proportional to the area of the surface, i.e. a surface tension.

3 Lattice Boltzmann equation models in brief

A physical system can be described on different levels, each of which corresponding to different resolutions in time and space. In statistical mechanics of essentially classical systems, such as ordinary liquids and gases, there are three important levels of description. The first (most detailed) level considers the motion of individual atoms and molecules. In a real liquid at room temperature and atmospheric pressure this implies spatial scale of about 10^{-9} m, and temporal scale of about 10^{-12} s. Considerations at this scale are important e.g. in modelling chemical reactions in solution.

The second level of description becomes valid when the actual motion can be substituted with the mean number of particles to have momentum in a range $[\vec{\xi}, \vec{\xi} + \Delta\vec{\xi}]$ at positions $[\vec{x}, \vec{x} + \Delta\vec{x}]$. The condition for this level to be accurate is that fluctuations of this number are relatively small, or that the mean number is much larger than one. The system is then described by a chain of distribution functions, where $f(\vec{\xi}, \vec{x}, t)$ is the probability density of finding a particle at position \vec{x} with momentum $\vec{\xi}$ at time t , $f_2(\vec{\xi}_1, \vec{\xi}_2, \vec{x}_1, \vec{x}_2, t)$ is the joint probability of finding one particle at \vec{x}_1 with momentum ξ_1 and another at \vec{x}_2 with momentum ξ_2 , and so on. A description of a gas or a liquid in terms of distribution functions is called kinetic theory. The simplest such description, valid for a very rare gas, is Boltzmann's equation, which in the single relaxation time approximation reads:

$$\frac{\partial f}{\partial t} + \vec{\xi} \cdot \vec{\nabla} f = -\frac{1}{\tau}(f - f^{eq}) \quad (3.1)$$

where f^{eq} would be the distribution function of a liquid or a gas in thermal equilibrium with the same density, velocity and temperature. Boltzmann's equation can include a force field (acceleration term) and of course Boltzmann's full collision operator Ω , quadratic in f . In this paper we will however in accordance with most of the Lattice Boltzmann equation literature stay with the (simpler) single relaxation time approximation for Ω .

The first three moments of the distribution function f are respectively the density

(ρ), the momentum density, (ρu) and the kinetic energy density per unit mass (ε_K):

$$\int f(\vec{\xi}, \vec{x}, t) d\vec{\xi} = \rho(\vec{x}, t) \quad (3.2a)$$

$$\int \vec{\xi} f(\vec{\xi}, \vec{x}, t) d\vec{\xi} = \rho(\vec{x}, t) u(\vec{x}, t) \quad (3.2b)$$

$$\int \frac{\xi^2}{2m} f(\vec{\xi}, \vec{x}, t) d\vec{\xi} = \rho(\vec{x}, t) \varepsilon_K(\vec{x}, t) \quad (3.2c)$$

where m is the mass of a particle. We can introduce a local temperature field T such that $\varepsilon_K = \frac{1}{2}u^2 + \frac{D}{2}RT$, where D is dimension of space (2 or 3 in the cases of interest to us) and R the gas constant. The space integrals of density, momentum density and total energy density are integrals of the motion, namely total mass, total momentum and total energy. Large-scale modulations of these densities in space therefore have slow dynamics in time, compared to the other degrees of freedom on the kinetic level. A hydrodynamics of a physical system refers in general to a description only in terms of such slow, large-scale modes [8], which are therefore called hydrodynamic modes. Hydrodynamics in the ordinary sense, e.g. to describe the motion of water, is an example of such a description, albeit more complicated than for some other systems, since total energy is implicitly given by velocity, density and temperature through the equation of state.

A Lattice Boltzmann equation is a kinetic level description of a fictitious microworld with the same conserved quantities as the real world. If properly constructed (see below), it will then give rise to the same hydrodynamics. The smallest scale that needs to be resolved to compute the hydrodynamic equations in this manner is only such that hydrodynamics is valid for the scale of the object of interest. For practical purposes, Lattice Boltzmann equations are therefore a sort of finite difference schemes, which contain auxiliary variables that stand for purely kinetic (not hydrodynamic) modes. The number of auxiliary modes is typically 2-5 more than the number of hydrodynamic modes, which represents an overhead.

A quick way to introduce the Lattice Boltzmann equations is to discretize space on a regular lattice, and momentum to a set of vectors in the dual lattice. Equation (3.1) can then be solved to first-order accuracy in time

$$f_i(\vec{x} + e_i, t + \Delta t) - f_i(\vec{x}, t) = -\frac{1}{\tau}(f_i - f_i^{eq}) \quad (3.3)$$

where the indices i label the set of momentum vectors, f_i is the probability to be at lattice point \vec{x} with momentum e_i at time t , and time Δt is such that a particle with

velocity e_i moves from \vec{x} to $\vec{x} + e_i$. The hydrodynamic variables are defined in a way similar to the continuum, i.e.

$$\sum_i w_i f_i = \rho \quad (3.4a)$$

$$\sum_i w_i f_i e_i = \rho u \quad (3.4b)$$

$$\frac{1}{2} \sum_i w_i f_i |e_i - u|^2 = \rho \frac{D}{2} RT \quad (3.4c)$$

where the w_i are a set of weights. A given Lattice Boltzmann model is then determined by equation (3.3) (or a generalization thereof), and how the equilibrium distribution function f_i^{eq} depends on ρ , u and T .

The a priori conditions for the same hydrodynamical equations to result from a Lattice Boltzmann model as from a real physical system, is that the symmetry of the lattice is sufficiently close to the full rotational symmetry of space. To capture correctly dissipative terms one needs that the invariant tensors form from the lattice vectors up to fourth order are isotropic:

$$\sum_i w_i = 1 \quad (3.5a)$$

$$\sum_i w_i e_{i\alpha} = 0 \quad (3.5b)$$

$$\sum_i w_i e_{i\alpha} e_{i\beta} = \Upsilon^{(2)} \delta_{\alpha\beta} \quad (3.5c)$$

$$\sum_i w_i e_{i\alpha} e_{i\beta} e_{i\gamma} = 0 \quad (3.5d)$$

$$\sum_i w_i e_{i\alpha} e_{i\beta} e_{i\gamma} e_{i\theta} = \Upsilon^{(4)} (\delta_{\alpha\beta} \delta_{\gamma\theta} + \delta_{\alpha\gamma} \delta_{\beta\theta} + \delta_{\alpha\theta} \delta_{\beta\gamma}) \quad (3.5e)$$

Greek indices ($\alpha, \beta \dots$) here label spatial directions, i the lattice vectors and $\Upsilon^{(2)}$ and $\Upsilon^{(4)}$ are lattice constants. Natural choices of lattices share with space the property that all odd order invariant tensors are zero, and the condition on second-order is easily satisfied. The technical difficulty overcome in the 80'ies (in the context of Lattice Gases) was to choose lattices with isotropic fourth order tensors, such as the hexagonal lattice in two dimensions. For faster convergence to the hydrodynamics it has sometimes been proposed to use lattices with isotropic sixth or higher order tensors [36, 12].

One presently popular model uses a lattice of 9 velocities in 2 dimensions, and is therefore known as the $D2Q9$ lattice. The velocities are the rest state (velocity $e_0 = 0$), along the vectors to the four nearest neighbours in a simple cubic lattice (velocities e_1-e_4), and along the vectors to the four next-nearest neighbour (e_5-e_8). If lattice length is $c\Delta t$, the velocities and weights are given in the following table:

Direction	Lattice vector	Weight
e_0	$(0, 0)$	$w_0 = 4/9$
e_1	$c(1, 0)$	$w_1 = 1/9$
e_2	$c(0, 1)$	$w_2 = 1/9$
e_3	$c(-1, 0)$	$w_3 = 1/9$
e_4	$c(0, -1)$	$w_4 = 1/9$
e_5	$c(1, 1)$	$w_5 = 1/36$
e_6	$c(-1, 1)$	$w_6 = 1/36$
e_7	$c(-1, -1)$	$w_7 = 1/36$
e_8	$c(1, -1)$	$w_8 = 1/36$

It is easily checked that all odd order invariant tensors formed from this lattice indeed vanish, and that it satisfies (3.5c) and (3.5e) with $\Upsilon^{(2)} = \frac{c^2}{3}$ and $\Upsilon^{(4)} = \frac{c^4}{9}$.

For the simplest applications it is sufficient to choose the equilibrium distribution function as

$$f_i^{eq} = \rho \left[1 + \frac{(e_i \cdot \mathbf{u})}{\Upsilon^{(2)}} + \frac{(e_i \cdot \mathbf{u})^2}{2\Upsilon^{(4)}} - \frac{\mathbf{u}^2}{2\Upsilon^{(2)}} \right] \quad (3.6)$$

from which we can derive

$$\sum_i w_i f_i^{eq} = \rho \quad (3.7a)$$

$$\sum_i w_i f_i^{eq} e_{i\alpha} = \rho u_\alpha \quad (3.7b)$$

$$\sum_i w_i f_i^{eq} e_{i\alpha} e_{i\beta} = \Upsilon^{(2)} \rho \delta_{\alpha\beta} + \rho u_\alpha u_\beta \quad (3.7c)$$

$$\sum_i w_i f_i^{eq} e_{i\alpha} e_{i\beta} e_{i\gamma} = \frac{\Upsilon^{(4)}}{\Upsilon^{(2)}} \rho (\delta_{\alpha\beta} u_\gamma + \delta_{\alpha\gamma} u_\beta + \delta_{\beta\gamma} u_\alpha) \quad (3.7d)$$

From (3.7c) it is natural to identify $\Upsilon^{(2)} \rho$ with a pressure term p . This means that the velocity of sound, $v_s = \sqrt{\frac{\partial p}{\partial \rho}}$, should be $\sqrt{\Upsilon^{(2)}}$ (a constant). For the

$D2Q9$ models this constant is $c/\sqrt{3}$. The temperature RT is on the other hand $\frac{1}{D\rho} \sum_i w_i f_i^{eq} (e_{i\alpha} - u_\alpha)(e_{i\alpha} - u_\alpha)$, a constant equal to $\Upsilon^{(2)}$. A model using (3.1) and (3.6) can therefore only describe isothermal hydrodynamics with an ideal gas equation of state, $p = RT\rho$. This is sufficient for deriving e.g. incompressible Navier-Stokes dynamics in a small Mach number expansion, but otherwise somewhat limited. Two important issues in the following will be to generalize (3.3) or (3.6) or both, to allow for flows which do not have the ideal gas equation of state, and which are not restricted to be isothermal.

The usual presentation of Lattice Boltzmann methods (as above) uses a uniform grid in space. This limitation is not a principal one, by varying in an appropriate manner the weights w_i it is possible to use also non-uniform grids. These issues have not been investigated extensively in the literature, see however [32] and [17].

4 Non-equilibrium thermodynamics model

This model was developed by Swift and collaborators [44, 45]. The model has later been used in numerical work by several teams, for contributions from authors of this report, see [33] and [30]. The basic idea is to assume that the distributions relax to a local equilibrium distribution function, which in a one-phase region coincides with the equilibrium in ordinary one-phase flows. In the interface region the local equilibrium is however assumed to have a pressure (stress) tensor as in the diffuse interface theory. The model uses the D2Q9 lattice, equation (3.3) and assumes an isothermal situation. The equilibrium distribution function is a generalization of (3.6)

$$\begin{cases} f_i^{eq} = A + Be_{i\alpha}u_\alpha + Cu^2 + Du_\alpha u_\beta e_{i\alpha}e_{i\beta} + G_{\alpha\beta}e_{i\alpha}e_{i\beta} \\ f_0^{eq} = A_0 + C_0u^2 \end{cases} \quad (4.1)$$

The coefficients A , B , C , D , $G_{\alpha,\beta}$ and A_0 and C_0 are chosen such that the zeroth and first weighted moments are ρ and ρu , while the second moment is prescribed to be

$$\sum_i w_i f_i^{eq} e_{i\alpha}e_{i\beta} = P_{\alpha\beta} + \rho u_\alpha u_\beta \quad (4.2)$$

where second order tensor P (in [44] referred to as a non-equilibrium pressure tensor) is chosen to be a stress tensor in a diffuse interface model:

$$P_{\alpha\beta}(\vec{r}) = p(\vec{r})\delta_{\alpha\beta} + \kappa\partial_\alpha\rho\partial_\beta\rho \quad (4.3)$$

The diagonal component $p(\vec{r})$ of the tensor P is

$$p(\vec{r}) = p_0 - \kappa\rho\nabla^2\rho - \frac{\kappa}{2}|\vec{\nabla}\rho|^2 \quad (4.4)$$

and p_0 is the pressure in a Van der Waals equation of state

$$p_0 = \frac{\rho RT}{1 - b\rho} - a\rho^2 \quad (4.5)$$

where T is temperature (assumed constant). The partial derivatives of the density are evaluated as discrete differences over neighbouring lattice points. In terms of a real system, this introduces a coupling between neighbouring sites, i.e. a potential energy.

The kinetic energy density computed from the distribution function (4.1) would

in fact by (4.2) give rise to a local temperature field $T(\vec{r}) = \frac{p_0}{\rho} - \kappa \nabla^2 \rho$. The equilibrium distribution in this model is however computed not with this variable temperature, but with a global constant parameter. The use of the model is therefore restricted to phenomena where the back action of a temperature gradient on density and momentum can be neglected, basically a isothermal setting.

The model starting from (4.3) will give rise to Gallilean non-invariant terms in the hydrodynamic equations. It will become clear in the following that of the various discretization errors that appear in the Swift et al. model, there are three which are more serious, and should be considered of the same order as the diffusive/viscous terms. To the model will therefore be added a term $\mathcal{F}_{\alpha\beta}$ which eliminates two of these.

$$P_{\alpha\beta}(\vec{r}) = p(\vec{r})\delta_{\alpha\beta} + \kappa\partial_\alpha\rho\partial_\beta\rho + \mathcal{F}_{\alpha\beta} \quad (4.6)$$

One result of [45] is that the mass and momentum equations are:

$$\frac{\partial\rho}{\partial t} + \nabla \cdot (\rho \mathbf{u}) = 0 \quad (4.7)$$

and

$$\begin{aligned} \frac{\partial(\rho \mathbf{u})}{\partial t} + (\mathbf{u} \cdot \nabla)(\rho \mathbf{u}) = & -\nabla p_0 + \nu \Delta(\rho \mathbf{u}) + \nabla \cdot [\lambda(\rho) \nabla \cdot (\rho \mathbf{u})] \\ & - \delta_t \frac{dp_0}{d\rho} \nabla \cdot (\nabla \cdot (\rho \mathbf{u}) + (\rho \mathbf{u}) \cdot \nabla) \end{aligned} \quad (4.8)$$

in which the kinematic viscosity ν and bulk viscosity λ of the fluid are determined by:

$$\begin{aligned} \nu = & \delta_t \frac{2\tau - 1}{2} \frac{\Upsilon^{(4)}}{\Upsilon^{(2)}} \\ \lambda(\rho) = & \delta_t \left(\tau - \frac{1}{2} \right) \left(\frac{2\Upsilon^{(4)}}{\Upsilon^{(2)}} - \frac{\partial p_0}{\partial \rho} \right) \end{aligned}$$

In fact, as we will see, equation (4.8) is only correct with the additional assumption that the fluid is close to incompressible, something which is also found in [44].

4.1 Method of successive approximations

Following [44] we will derive continuity equation and the Navier-Stokes equation by the method of successive approximations. The discussion in this section does

not contain other material than in [44], but is in places carried out in more detail.

We start from equation (3.3) with a Taylor expansion of the l.h.s:

$$\Omega_i \equiv -\frac{1}{\tau}(f_i - f_i^{eq}) = \sum_{k=1}^{\infty} \frac{1}{k!} \delta_t^k (\partial_t + e_{i\alpha} \partial_\alpha)^k f_i = \sum_{k=1}^{\infty} \frac{1}{k!} \delta_t^k \mathcal{D}^k f_i \quad (4.9)$$

with $\mathcal{D} \equiv (\partial_t + e_{i\alpha} \partial_\alpha)$. We assume that the distribution function f is close to the equilibrium distribution function f^{eq} and expand in δ_t .

$$f_i = f_i^{eq} + (f_i - f_i^{eq}) = f_i^{eq} - \tau \left(\delta_t \mathcal{D} f_i + \frac{\delta_t^2}{2} \mathcal{D}^2 f_i \right) + \mathcal{O}(\delta_t^3) \quad (4.10)$$

Substituting (4.10) to r.h.s of (4.9) and retaining terms to order δ_t^2

$$\begin{aligned} \frac{\Omega_i}{\delta_t} &= \mathcal{D} \left[f_i^{eq} - \tau \left(\delta_t \mathcal{D} f_i + \frac{\delta_t^2}{2} \mathcal{D}^2 f_i \right) \right] \\ &\quad + \frac{\delta_t}{2} \mathcal{D}^2 \left[f_i^{eq} - \tau \left(\delta_t \mathcal{D} f_i + \frac{\delta_t^2}{2} \mathcal{D}^2 f_i \right) \right] + \mathcal{O}(\delta_t^2) \\ &= \mathcal{D} f_i^{eq} - \tau \delta_t \mathcal{D}^2 f_i + \frac{\delta_t}{2} \mathcal{D}^2 f_i^{eq} + \mathcal{O}(\delta_t^2) \\ &= \mathcal{D} f_i^{eq} - \delta_t \left(\tau - \frac{1}{2} \right) \mathcal{D}^2 f_i^{eq} + \mathcal{O}(\delta_t^2) \end{aligned} \quad (4.11)$$

So

$$\frac{\Omega_i}{\delta_t} = (\partial_t + e_{i\alpha} \partial_\alpha) f_i^{eq} - \delta_t \left(\tau - \frac{1}{2} \right) (\partial_t + e_{i\alpha} \partial_\alpha)^2 f_i^{eq} + \mathcal{O}(\delta_t^2) \quad (4.12)$$

By projecting on the hydrodynamic modes we will now derive the macroscopic equations to lowest and next lowest order in δ_t . To obtain the continuity equation (4.7) to lowest order, we sum over i and use (3.4a) and (3.4b), which gives

$$0 = \sum_i w_i \frac{\Omega_i}{\delta_t} = \partial_t \sum_i w_i f_i^{eq} + \partial_\alpha \sum_i w_i e_{i\alpha} f_i^{eq} - \mathcal{O}(\delta_t) \quad (4.13a)$$

Or

$$\partial_t \rho + \partial_\alpha (\rho u_\alpha) = \mathcal{O}(\delta_t) \quad (4.13b)$$

To the next order in δ_t :

$$= \delta_t \left(\tau - \frac{1}{2} \right) \left[\partial_t \partial_t \sum_i w_i f_i^{eq} + 2 \partial_t \partial_\alpha \sum_i w_i e_{i\alpha} f_i^{eq} + \partial_\alpha \partial_\beta \sum_i w_i e_{i\alpha} e_{i\beta} f_i^{eq} \right] \quad (4.14)$$

$$= \delta_t \left(\tau - \frac{1}{2} \right) \left[\partial_t (\partial_t \rho + \partial_\alpha (\rho u_\alpha)) + \partial_\alpha \left(\partial_t (\rho u_\alpha) + \partial_\beta \sum_i w_i e_{i\alpha} e_{i\beta} f_i^{eq} \right) \right] \quad (4.15)$$

Substituting in eqn.(4.15), eqn.(4.13b) and (4.18) derived below we see that the $\mathcal{O}(\delta_t)$ terms are actually $\mathcal{O}(\delta_t^2)$. The continuity equation is therefore:

$$\partial_t \rho + \partial_\alpha (\rho u_\alpha) = \mathcal{O}(\delta_t^2) \quad (4.16)$$

This means that there will not be any density diffusion terms in our equations, as indeed there should not be on general grounds [24].

To obtain the momentum equation, (4.8), we multiply eqn.(4.12) by $e_{i\beta}$ and sum over i :

$$0 = \partial_t \sum_i w_i e_{i\beta} f_i^{eq} + \partial_\alpha \sum_i w_i e_{i\alpha} e_{i\beta} f_i^{eq} - \mathcal{O}(\delta_t) \quad (4.17)$$

$$0 = \partial_t (\rho u_\beta) + \partial_\alpha (P_{\alpha\beta} + \rho u_\alpha u_\beta) - \delta_t \left(\tau - \frac{1}{2} \right) \quad (4.18)$$

$$\left[\underbrace{\partial_t^2 (\rho u_\beta)}_{\mathcal{A}_\beta} + \underbrace{\partial_\alpha \partial_\gamma \left(\sum_i w_i e_{i\alpha} e_{i\beta} e_{i\gamma} f_i^{eq} \right)}_{\mathcal{B}_\beta} + \underbrace{2 \partial_t \partial_\alpha (P_{\alpha\beta} + \rho u_\alpha u_\beta)}_{\mathcal{C}_\beta} \right] + \mathcal{O}(\delta_t^2)$$

To lowest order we get the Euler equation which can be substituted back into the last parenthesis in eqn.(4.15), to prove that the continuity equation holds to order δ_t^2 . Expanding $P_{\alpha\beta}$ by equation (4.6) we have

$$\begin{aligned} \partial_t (\rho u_\beta) + \partial_\alpha (\rho u_\alpha u_\beta) &= -\partial_\beta p_0 + \kappa \partial_\beta (\rho \Delta \rho) + \frac{\kappa}{2} \partial_\beta |\nabla \rho|^2 \\ &\quad - \partial_\alpha (\kappa \partial_\alpha \rho \partial_\beta \rho) - \partial_\alpha \mathcal{F}_{\alpha\beta} + \mathcal{A}_\beta + \mathcal{B}_\beta + \mathcal{C}_\beta \end{aligned} \quad (4.19)$$

Equation (4.19) shows terms on the Euler level, where some terms have been simplified using (4.2). We will now simplify further the three viscous-level terms denoted \mathcal{A}_β , \mathcal{B}_β and \mathcal{C}_β . Term \mathcal{A}_β can be computed by substituting back eqn.(4.19)

$$\begin{aligned}\partial_t^2(\rho u_\beta) &= \partial_t(\partial_t(\rho u_\beta)) \\ &= -\partial_t(\partial_\alpha(P_{\alpha\beta} + \rho u_\alpha u_\beta)) + \mathcal{O}(\delta_t)\end{aligned}\quad (4.20)$$

In the following manipulations, we always imply all equalities to hold up to terms of order δ_t . Therefore,

$$\begin{aligned}\mathcal{A}_\beta + \mathcal{C}_\beta &= \partial_t(\partial_\alpha(P_{\alpha\beta} + \rho u_\alpha u_\beta)) \\ &= \partial_t \partial_\beta p_0 - \partial_t \partial_\beta(\kappa \rho \Delta \rho) - \partial_t \partial_\beta \left(\frac{\kappa}{2} |\nabla \rho|^2 \right) + \\ &\quad \partial_t \partial_\alpha (\kappa \partial_\beta \rho \partial_\alpha \rho) + \partial_\alpha \partial_t(\rho u_\alpha u_\beta) + \partial_\alpha \partial_t \mathcal{F}_{\alpha\beta}\end{aligned}\quad (4.21)$$

We see that there are very many terms to order δ_t . It will be useful to concentrate on the terms with smallest number of derivatives acting on the density. We will therefore, following Swift, neglect terms with as least as many derivatives as these:

$$\begin{aligned}\partial_t \partial_\beta(\kappa \rho \Delta \rho) &\approx 0 \\ \partial_t \partial_\beta \left(\frac{\kappa}{2} |\nabla \rho|^2 \right) &\approx 0 \\ \partial_t \partial_\alpha (\kappa \partial_\beta \rho \partial_\alpha \rho) &\approx 0 \\ \partial_\alpha \partial_t \mathcal{F}_{\alpha\beta} &\approx 0\end{aligned}$$

With this simplification,

$$\mathcal{A}_\beta + \mathcal{C}_\beta = \underbrace{\partial_t \partial_\beta p_0}_{\mathbf{AC1}_\beta} + \underbrace{\partial_\alpha [u_\beta \partial_t(\rho u_\alpha)]}_{\mathbf{AC2}_\beta} + \underbrace{\partial_\alpha [(\rho u_\alpha) \partial_t u_\beta]}_{\mathbf{AC3}_\beta} \quad (4.22)$$

These terms can be further simplified by

$$\mathbf{AC1}_\beta = \partial_t \partial_\beta p_0 = \partial_\beta \frac{\partial p_0}{\partial \rho} (\partial_t \rho) = -\partial_\beta \left(\frac{\partial p_0}{\partial \rho} \partial_\alpha (\rho u_\alpha) \right) \quad (4.23)$$

$$\mathbf{AC2}_\beta = -\partial_\alpha [u_\beta \partial_\gamma (P_{\alpha\gamma} + \rho u_\alpha u_\gamma)] \quad (4.24)$$

Neglected furthermore these terms in equation (4.24)

$$\begin{aligned}\partial_\alpha \partial_\gamma (\kappa \rho \Delta \rho) &\approx 0 \\ \partial_\alpha \partial_\gamma \left(\frac{\kappa}{2} |\nabla \rho|^2 \right) &\approx 0 \\ \partial_\alpha \partial_\gamma (\kappa \partial_\theta \rho \partial_\delta \rho) &\approx 0 \\ \partial_\alpha \partial_\gamma \mathcal{F} &\approx 0\end{aligned}$$

we have

$$\mathbf{AC2}_\beta = -\partial_\alpha[u_\beta\partial_\alpha p_0 - u_\beta\partial_\gamma(\rho u_\alpha u_\gamma)] \quad (4.25)$$

The term $\mathbf{AC3}$ can be computed by using

$$\partial_t(\rho u_\beta) = u_\beta\partial_t\rho + \rho\partial_t u_\beta = \rho\partial_t u_\beta - u_\alpha\partial_\gamma(\rho u_\gamma) \quad (4.26)$$

So

$$\rho\partial_t u_\alpha = -\partial_\beta(P_{\alpha\beta} + \rho u_\alpha u_\beta) + u_\alpha\partial_\beta(\rho u_\alpha) \quad (4.27)$$

Therefore

$$\begin{aligned} \mathbf{AC3}_\beta &= \partial_\alpha[u_\alpha(\rho\partial_t u_\beta)] \\ &= -\partial_\alpha\{u_\alpha[\partial_\gamma(P_{\gamma\beta} + \rho u_\gamma u_\beta) + u_\beta\partial_\gamma(\rho u_\gamma)]\} \\ &= -\partial_\alpha\{u_\alpha\partial_\beta p_0 - \rho u_\alpha u_\gamma\partial_\gamma u_\beta\} \end{aligned} \quad (4.28)$$

Substituting equations (4.23), (4.25) and (4.28) back into (4.22) gives

$$\begin{aligned} \mathcal{A}_\beta + \mathcal{C}_\beta &= -\partial_\beta\left(\frac{\partial p_0}{\partial\rho}\partial_\alpha(\rho u_\alpha)\right) \\ &\quad -\partial_\alpha[u_\beta\partial_\alpha p_0 + u_\alpha\partial_\beta p_0 - u_\beta\partial_\gamma(\rho u_\alpha u_\gamma) - \rho u_\alpha u_\gamma\partial_\gamma u_\beta] \\ &= -\partial_\beta\left(\frac{\partial p_0}{\partial\rho}\partial_\alpha(\rho u_\alpha)\right) - \partial_\alpha(u_\beta\partial_\alpha p_0 + u_\alpha\partial_\beta p_0) + \partial_\alpha\partial_\gamma(\rho u_\alpha u_\beta u_\gamma) \\ &= -\partial_\beta\left(\frac{\partial p_0}{\partial\rho}\partial_\alpha(\rho u_\alpha)\right) - \partial_\alpha\left[\frac{\partial p_0}{\partial\rho}(u_\beta\partial_\alpha\rho + u_\alpha\partial_\beta\rho)\right] + \partial_\alpha\partial_\gamma(\rho u_\alpha u_\beta u_\gamma) \end{aligned} \quad (4.29)$$

The term \mathcal{B}_β in the equation (4.18) can be computed by using the equilibrium distribution function (4.1)

$$\begin{aligned} \partial_\alpha\partial_\gamma\left(\sum_i w_i f_i^{eq} e_{i\alpha} e_{i\beta} e_{i\gamma}\right) &= \partial_\alpha\partial_\gamma\left(\sum_i w_i B u_\alpha e_{i\alpha} e_{i\beta} e_{i\gamma} e_{i\theta}\right) \\ &= \partial_\alpha\partial_\gamma B \Upsilon^{(4)}(u_\gamma\delta_{\alpha\beta} + u_\beta\delta_{\alpha\gamma} + u_\alpha\delta_{\beta\gamma}) \end{aligned} \quad (4.31)$$

In eqn.(4.31), the only free index is β , while α and γ are summed over. The value of B given by Swift et al is $\rho/\Upsilon^{(2)}$ ¹ and therefore

$$\mathcal{B}_\beta = \frac{\Upsilon^{(4)}}{\Upsilon^{(2)}}(2\partial_\beta\partial_\alpha(\rho u_\alpha) + \Delta(\rho u_\beta)) \quad (4.32)$$

¹ The expression given by Swift et al is for the two-dimensional hexagonal lattice, for which $\Upsilon^{(2)} = 3c^2$, $\Upsilon^{(4)} = 3c^4/4$ and hence $B\Upsilon^{(4)} = \rho c^2/4$.

Substituting now equations (4.30) and (4.32) into (4.19) we have

$$\begin{aligned}
\partial_t(\rho u_\beta) + \partial_\alpha(\rho u_\alpha u_\beta) = & -\partial_\beta p_0 + \kappa \partial_\beta(\rho \Delta \rho) + \frac{\kappa}{2} \partial_\beta(|\nabla \rho|^2) \\
& - \kappa \partial_\alpha(\partial_\alpha \rho \partial_\beta \rho) - \partial_\alpha \mathcal{F}_{\alpha\beta} + \underbrace{\Delta \left(\frac{\delta_t(2\tau-1)}{2} \frac{\Upsilon^{(4)}}{\Upsilon^{(2)}} \rho \mathbf{u} \right)}_{\nu \rho \mathbf{u}} \\
& + \partial_\beta \left[\underbrace{\delta_t \left(\tau - \frac{1}{2} \right) \left(2 \frac{\Upsilon^{(4)}}{\Upsilon^{(2)}} \partial_\alpha(\rho u_\alpha) - \frac{\partial p_0}{\partial \rho} \partial_\alpha(\rho u_\alpha) \right)}_{\lambda(\rho) \partial_\alpha(\rho u_\alpha)} \right] \\
& - \delta_t \left(\tau - \frac{1}{2} \right) \partial_\alpha \left[\frac{\partial p_0}{\partial \rho} (u_\beta \partial_\alpha \rho + u_\alpha \partial_\beta \rho) + \partial_\gamma(\rho u_\alpha u_\beta u_\gamma) \right] \quad (4.33)
\end{aligned}$$

Some terms in the equation (4.33) can be further simplified as:

$$\begin{aligned}
& \kappa \partial_\beta(\rho \Delta \rho) + \frac{\kappa}{2} \partial_\beta(|\nabla \rho|^2) - \kappa \partial_\alpha(\partial_\alpha \rho \partial_\beta \rho) \\
& = \kappa \partial_\beta(\rho \Delta \rho) + \kappa \partial_\beta \rho (\partial_\alpha \partial_\beta \rho) - \kappa (\partial_\alpha \partial_\beta \rho) \partial_\beta \rho - \kappa (\partial_\beta \rho) \Delta \rho \\
& = \kappa \rho \partial_\beta(\Delta \rho) \quad (4.34)
\end{aligned}$$

So, the momentum equation is:

$$\begin{aligned}
\partial_t(\rho u_\beta) + \partial_\alpha(\rho u_\alpha u_\beta) = & -\partial_\beta p_0 + \nu \Delta(\rho u_\beta) + \partial_\beta[\lambda(\rho) \partial_\alpha(\rho u_\alpha)] \\
& - \delta_t \left(\tau - \frac{1}{2} \right) \frac{dp_0}{d\rho} \partial_\alpha (u_\alpha \partial_\beta \rho + u_\beta \partial_\alpha \rho) - \partial_\alpha \mathcal{F}_{\alpha\beta} + \kappa \rho \partial_\beta(\Delta \rho) \quad (4.35)
\end{aligned}$$

The extra viscous terms in the momentum equation (4.35) are not Galilean invariant when density gradients are present. Comparing one sees that all of these terms can be derived from spatial derivatives acting on the pressure, i.e. term \mathcal{C}_β in (4.18). A possible interpretation is therefore that the Swift et al. condition on the second moments of the distribution function, i.e. equation (4.6), effectively involves an evaluation of a force term in Boltzmann's equation (compare section 6 below), which has to be done to better than linear order if unphysical viscous terms are to be avoided.

Some of the non-Galilean invariant terms in (4.35) can be removed by an additional term $\mathcal{F}_{\alpha\beta}$ in the pressure tensor (equation (4.6)). First, we define:

$$\xi \equiv 2\nu - \lambda = \delta_t \left(\tau - \frac{1}{2} \right) \frac{dp_0}{d\rho} \quad (4.36)$$

The equation (4.35) can then be rewritten:

$$\begin{aligned}\partial_t(\rho u_\beta) + \partial_\alpha(\rho u_\alpha u_\beta) = & -\partial_\beta p_0 + \nu \partial_\alpha(\rho \partial_\alpha u_\beta) + \partial_\beta(\lambda \rho \partial_\alpha u_\alpha) \\ & + \partial_\alpha[(\nu - \xi)u_\beta \partial_\alpha \rho - \xi u_\alpha \partial_\beta \rho] \\ & + \partial_\beta[\lambda u_\alpha \partial_\alpha \rho] + \partial_\alpha \mathcal{F}_{\alpha\beta} + \kappa \rho \partial_\beta(\Delta \rho)\end{aligned}\quad (4.37)$$

By a suitable choice of the term \mathcal{F} we can eliminate two of the three terms with one derivative of ρ

$$\mathcal{F}_{\alpha\beta} = \xi(u_\beta \partial_\alpha \rho - u_\alpha \partial_\beta \rho) - \lambda u_\gamma \partial_\gamma \rho \delta_{\alpha\beta} \quad (4.38)$$

and with this choice the momentum equation is becomes

$$\begin{aligned}\partial_t(\rho u_\beta) + \partial_\alpha(\rho u_\alpha u_\beta) = & -\partial_\beta p_0 + \nu \partial_\alpha(\rho \partial_\alpha u_\beta) + \partial_\beta(\lambda \rho \partial_\alpha u_\alpha) \\ & + \nu \partial_\alpha(u_\beta \partial_\alpha \rho) + \kappa \rho \partial_\beta(\Delta \rho)\end{aligned}\quad (4.39)$$

With this modified model Swift et al. found qualitatively correct dynamics of a bubble in a liquid. This should be considered an experimental fact, difficult to explain theoretically.

4.2 Summary

The model of Swift et al. has some disadvantages:

- The model is not Galilean invariant when the density gradients are present. By introducing the extra term \mathcal{F} , we can remove some of the non-Galilean invariant terms, but not all.
- It is difficult to introduce the force terms into this model. We have to modify the continuity equation with a force terms \mathbf{F} inside the derivative of $\rho \mathbf{u}$.

$$\partial_t \rho + \nabla \cdot (\rho \mathbf{u} + \mathbf{F}) = 0 \quad (4.40)$$

Therefore, this method can only be used if the force term \mathbf{F} is negligibly small.

5 Luo model: LBE for non-ideal dense gases

The model of Li-Shi Luo starts from Boltzmann equation for dense gases [27], also known as the Enskog equation:

$$\frac{\partial f}{\partial t} + \xi \cdot \nabla f + \mathbf{a} \cdot \nabla_{\xi} f = \Omega \quad (5.1)$$

where \mathbf{a} and Ω are the acceleration and collision operators. With the expansion of Ω in a Taylor series, using a single relaxation time approximation, and assuming the fluid to be isothermal and incompressible, Luo obtains the following equation

$$\frac{\partial f}{\partial t} + \xi \cdot \nabla f + \mathbf{a} \cdot \nabla_{\xi} f = -\frac{g}{\lambda}(f - f^{eq}) + J \quad (5.2a)$$

$$J = -f^{eq} b \rho g (\xi - \mathbf{u}) \cdot \nabla \ln(\rho^2 g) \quad (5.2b)$$

where b is the second virial coefficient of the equation of state for the hard-sphere system, and g is the radial distribution function. The equilibrium distribution function is discussed by Luo in the continuum form

$$f^{eq} = \frac{\rho}{(2\pi RT)^{D/2}} \exp \left[-\frac{(\xi - \mathbf{u})^2}{2RT} \right] \quad (5.3)$$

but is used in the discretized form (3.6).

Solving (5.2) to first order in time we have

$$\begin{aligned} f(x + \xi \delta_t, \xi, t + \delta_t) - f(x, \xi, t) = & -\frac{1}{\tau} (f(x, \xi, t) - f^{eq}(x, \xi, t)) \\ & + J(x, \xi, t) \delta_t - \mathbf{a} \cdot \nabla_{\xi} f(x, \xi, t) \delta_t \end{aligned} \quad (5.4)$$

with a force term, $\mathbf{a} \cdot \nabla_{\xi} f$, written [27]

$$\mathbf{a} \cdot \nabla_{\xi} f = -\rho \omega(\xi) \xi_T^{-2} [(\xi - \mathbf{u}) + \xi_T^{-2} (\xi \cdot \mathbf{u}) \xi] \cdot \mathbf{a} \quad (5.5)$$

The auxiliary variable ξ_T is equal to \sqrt{RT} ($\frac{c}{\sqrt{3}}$ in the $D2Q9$ model), and the weight $\omega(\xi)$ denotes the weight w_i for the momentum $\xi = e_i$. The force term J is proportional to the gradient of the function of density $\rho^2 g$, which as in the Swift model is evaluated by discrete differences over lattice points. The interpretation is also that of a potential term. Equations (5.4) and (5.5) together with (3.6), (3.4a), (3.4b), and a choice of a lattice, define Luo's Lattice Boltzmann model.

To proceed further we want to evaluate moments with respect to momentum of the two “forces” (the interior force J , and the exterior force through the acceleration a). The forcing term of eqn.(5.5) is

$$F_i = -w_i \rho \left[\frac{1}{\Upsilon^{(2)}} (e_i - \mathbf{u}) + \frac{(e_i \cdot \mathbf{u})}{\Upsilon^{(4)}} e_i \right] \cdot \mathbf{a} \quad (5.6)$$

and satisfies the following constraints:

$$\sum_i F_i = 0 \quad (5.7a)$$

$$\sum_i (F_i e_{i\alpha}) = -\frac{\rho}{\Upsilon^{(2)}} \sum_i (w_i e_{i\alpha} e_{i\beta}) \cdot \mathbf{a} = -\rho a_\alpha \quad (5.7b)$$

$$\begin{aligned} \sum_i (F_i e_{i\alpha} e_{i\beta}) &= - \left[-\frac{\rho u_\gamma}{\Upsilon^{(2)}} \sum_i (e_{i\alpha} e_{i\beta} w_i) + \frac{\rho}{\Upsilon^{(4)}} \sum_i ((\mathbf{e}_i \cdot \mathbf{u}) e_{i\alpha} e_{i\beta} e_{i\gamma} w_i) \right] \cdot \mathbf{a} \\ &= \rho (u_\alpha a_\beta - u_\beta a_\alpha) \end{aligned} \quad (5.7c)$$

The interior force term J_i satisfies the following constraints:

$$\begin{aligned} \sum_i J_i &= -b\rho g \left(\sum_i (f_i^{eq} e_{i\alpha}) - \sum_i (f_i^{eq} u_\alpha) \right) \cdot \nabla \ln(\rho^2 g) \\ &= -b\rho g (\rho u - \rho u) \cdot \nabla \ln(\rho^2 g) = 0 \end{aligned} \quad (5.8a)$$

$$\begin{aligned} \sum_i (J_i e_{i\alpha}) &= -b\rho g \left(\sum_i (f_i^{eq} e_{i\alpha} e_{i\beta}) - \sum_i (f_i^{eq} e_{i\alpha}) u_\beta \right) \partial_\beta \log(\rho^2 g) \\ &= -b\Upsilon^{(2)} \partial_\alpha (\rho^2 g) \end{aligned} \quad (5.8b)$$

$$\begin{aligned} \sum_i (J_i e_{i\alpha} e_{i\beta}) &= -b\rho g \left(\sum_i (f_i^{eq} e_{i\alpha} e_{i\beta} e_{i\gamma}) - \sum_i (f_i^{eq} e_{i\alpha} e_{i\beta}) u_\gamma \right) \cdot \nabla \ln(\rho^2 g) \\ &= b[u_\alpha u_\beta \mathbf{u} \cdot \nabla - \Upsilon^{(2)} (u_\alpha \partial_\beta + u_\beta \partial_\alpha)] (\rho^2 g) \end{aligned} \quad (5.8c)$$

5.1 Chapman-Enskog analysis

In this section we will derive the macroscopic equations with the Chapman-Enskog method. The earlier presented method of successive approximations can be considered a special case of the more general and more flexible Chapman-Enskog method.

First, we apply the Taylor expansion to equation (5.4)

$$\sum_{n=1}^{\infty} \frac{\epsilon^n}{n!} \mathcal{D}_t^n f_i(\mathbf{x}, t) = -\frac{g}{\tau} (f_i(\mathbf{x}, t) - f_i^{eq}(\mathbf{x}, t)) + J_i \epsilon - F_i \epsilon \quad (5.9)$$

where $\epsilon = \delta_t$ and $\mathcal{D} \equiv (\partial_t + e_{i\alpha} \partial_\alpha)$. We now introduce the following expansion:

$$f_i = \sum_{n=0}^{\infty} \epsilon^n f_i^{(n)} \quad (5.10a)$$

$$\partial_t = \sum_{n=0}^{\infty} \epsilon^n \partial_{nt} \quad (5.10b)$$

$$\partial_\alpha = \partial_{0\alpha} \quad (5.10c)$$

The meaning of the expansion of f in powers of ϵ is clear. The meaning of the formal expansion of the time derivatives in derivatives with respect to different “times” is that f is assumed to have support in the frequency domain on bands that are separated by powers of ϵ . We will look for an equation involving a “slow time”, that is low-frequency components of f . Such components are Fourier transforms of f , where sufficiently fast oscillations are averaged out. A rigorous theory of these procedures can be given in homogenization theory [25]. In general one can also assume there to be fast and slow spatial variables (small and large scales). For the present problem it is however enough to assume one spatial scale, hence (5.10c)

With ϵ^2 order accuracy, equation (5.9) can be rewritten:

$$f_i \equiv f_i^{(0)} + \epsilon f_i^{(1)} + \epsilon^2 f_i^{(2)} = f_i^{eq} - \frac{\tau}{g} \left(\epsilon \mathcal{D}_t + \frac{\epsilon^2}{2} \mathcal{D}_t^2 \right) f_i + \frac{\tau}{g} J_i \epsilon - \frac{\tau}{g} F_i \epsilon \quad (5.11)$$

Group the right-hand side of the eqn.(5.11) by the order of ϵ .

$$\begin{aligned} f_i^{(0)} + \epsilon f_i^{(1)} + \epsilon^2 f_i^{(2)} &= f_i^{eq} - \epsilon \frac{\tau}{g} \mathcal{D}_t (f_i^{(0)} + \epsilon f_i^{(1)} + \epsilon^2 f_i^{(2)}) \\ &\quad - \epsilon^2 \frac{\tau}{2g} \mathcal{D}_t^2 (f_i^{(0)} + \epsilon f_i^{(1)} + \epsilon^2 f_i^{(2)}) \\ &\quad + \epsilon \frac{\tau}{g} J_i - \epsilon \frac{\tau}{g} F_i \end{aligned} \quad (5.12)$$

To successive orders in ϵ we find:

$$\epsilon^0 : \quad f_i^{(0)} = f_i^{eq} \quad (5.13a)$$

$$\epsilon^1 : \quad f_i^{(1)} = -\frac{\tau}{g} \mathcal{D}_{t_0} f_i^{(0)} + \frac{\tau}{g} J_i - \frac{\tau}{g} F_i \quad (5.13b)$$

$$\epsilon^2 : \quad f_i^{(2)} = -\frac{\tau}{2g} \mathcal{D}_{t_0}^2 f_i^{(0)} - \frac{\tau}{g} \partial_{t_1} f_i^{(0)} - \frac{\tau}{g} \mathcal{D}_{t_0} f_i^{(1)} \quad (5.13c)$$

The goal of the calculation is to project on the hydrodynamic modes of density and momentum. Equation (5.13a) shows that the zeroth order distribution function agrees with the equilibrium distribution function, which shares the same density and momentum. The higher order distribution functions therefore obey the following constraints:

$$\sum_i f_i^{(n)} = 0 \quad (n > 0) \quad (5.14)$$

$$\sum_i (f_i^{(n)} e_{i\alpha}) = 0 \quad (n > 0) \quad (5.15)$$

Summing equation (5.13b) over i , using equations (5.7a), and (5.8a), gives

$$\partial_{t_0} \rho + \partial_{0\alpha} (\rho u_\alpha) = 0 \quad (5.16)$$

To second order in ϵ , we sum equation (5.13c) over i , substituting the first order solution (5.13b) which gives

$$0 = -\frac{1}{2} \left[\sum_i \mathcal{D}_{t_0} J_i - \sum_i \mathcal{D}_{t_0} F_i \right] + \left(\frac{1}{2} - \frac{g}{\tau} \right) \sum_i \mathcal{D}_{t_0} f_i^{(1)} - \partial_{t_1} \sum_i f_i^{(0)} \quad (5.17)$$

or

$$\partial_{t_1} \rho = -\frac{1}{2} \partial_\alpha (\rho a_\alpha - b \Upsilon^{(2)} \partial_\alpha (\rho^2 g)) \quad (5.18)$$

Combining the first and the second order results for f_i by $\partial_t = \partial_{t_0} + \epsilon \partial_{t_1}$ and recalling that $\epsilon = \delta_t$, we have a continuity equation with an (unphysical) density diffusion term:²

$$\partial_t \rho + \partial_\alpha (\rho u_\alpha) = -\frac{\delta_t}{2} \partial_\alpha (\rho a_\alpha - b \Upsilon^{(2)} \partial_\alpha (\rho^2 g)) \quad (5.19)$$

The first order of the momentum equation can be produced by multiplying equation (5.13b) with $e_{i\beta}$, then summing over i :

$$0 = \sum_i \mathcal{D}_{t_0} (f_i^{(0)} e_{i\beta}) + \sum_i (J_i e_{i\beta}) - \sum_i (F_i e_{i\beta})$$

²The author of [27] claims there is no such density diffusion term, but we find that it is there.

By using equations (5.14), (5.8b), and (5.7b), we have

$$\begin{aligned}\partial_{t_0}(\rho u_\beta) + \partial_\alpha(\rho u_\alpha u_\beta) &= -\Upsilon^{(2)}\partial_\beta\rho - b\Upsilon^{(2)}\partial_\beta(\rho^2 g) + \rho a_\beta \\ &= -\partial_\beta P + \rho a_\beta\end{aligned}\quad (5.20)$$

where

$$P = \Upsilon^{(2)}\rho(1 + b\rho g)$$

is a non-ideal pressure. Equation (5.20) is hence Euler's equation. The second order momentum equation is

$$0 = \partial_{t_1} \sum_i f_i^{(0)} e_{i\beta} + \frac{1}{2} \mathcal{D}_{t_0} \sum_i (J_i e_{i\beta} - F_i e_{i\beta}) + \left(1 - \frac{g}{2\tau}\right) \mathcal{D}_{t_0} \sum_i (f_i^{(1)} e_{i\beta})$$

Substituting the equations (5.15), (5.7) and (5.8) into above we have

$$\partial_{t_1}(\rho u_\beta) = \left(\frac{g}{2\tau} - 1\right) \partial_\alpha \Pi_{\alpha\beta}^{(1)} - \frac{1}{2} \mathcal{D}_{t_0} \sum_i (J_i e_{i\beta} - F_i e_{i\beta}) \quad (5.21)$$

where the first-order momentum flux tensor $\Pi_{\alpha\beta}^{(1)}$ is $\sum_i f_i^{(1)} e_{i\alpha} e_{i\beta}$ and therefore

$$\begin{aligned}\partial_\alpha \Pi_{\alpha\beta}^{(1)} &= \partial_\alpha \sum_i f_i^{(1)} e_{i\alpha} e_{i\beta} \\ &= -\frac{\tau}{g} \partial_\alpha \sum_i \mathcal{D}_{t_0} \underbrace{f_i^{(0)} e_{i\alpha} e_{i\beta}}_{\Pi_{\alpha\beta}^{(0)}} + \frac{\tau}{g} \partial_\alpha \sum_i (J_i e_{i\alpha} e_{i\beta} - F_i e_{i\alpha} e_{i\beta})\end{aligned}\quad (5.22)$$

where $\Pi_{\alpha\beta}^{(0)}$ is the zeroth-order momentum flux tensor. Furthermore

$$\begin{aligned}\mathcal{D}_{t_0} \Pi_{\alpha\beta}^{(0)} &= \partial_{t_0} (\Upsilon^{(2)} \rho \delta_{\alpha\beta} + \rho u_\alpha u_\beta) + \Upsilon^{(2)} [\partial_\alpha(\rho u_\alpha) \delta_{\alpha\beta} + \partial_\alpha(\rho u_\beta) + \partial_\beta(\rho u_\alpha)] \\ &= \Upsilon^{(2)} [\partial_{t_0} \rho + \partial_\alpha(\rho u_\alpha)] \delta_{\alpha\beta} + \partial_{t_0}(\rho u_\alpha u_\beta) + \Upsilon^{(2)} [\partial_\alpha(\rho u_\beta) + \partial_\beta(\rho u_\alpha)] \\ &= \partial_{t_0}(\rho u_\alpha u_\beta) + \Upsilon^{(2)} [\partial_\alpha(\rho u_\beta) + \partial_\beta(\rho u_\alpha)] + \mathcal{O}(\delta_t)\end{aligned}\quad (5.23)$$

Substituting back the above results into (5.21), we have,

$$\begin{aligned}\partial_{t_1}(\rho u_\beta) &= \Upsilon^{(2)} \frac{2\tau - g}{2g} \partial_\alpha [\partial_\alpha(\rho u_\beta) + \partial_\beta(\rho u_\alpha)] + \frac{2\tau - g}{2g} \partial_\alpha \partial_{t_0}(\rho u_\alpha u_\beta) + \\ &\quad \frac{2\tau - g}{2g} \partial_{t_0} \sum_i (J_i e_{i\beta} - F_i e_{i\beta}) + \frac{1}{2} \partial_\alpha \sum_i (J_i e_{i\alpha} e_{i\beta} - F_i e_{i\alpha} e_{i\beta})\end{aligned}\quad (5.24)$$

Combining the first and the second order results, equations (5.20) and (5.24), for f_i by $\partial_t = \partial_{t_0} + \epsilon \partial_{t_1}$ and recalling that $\epsilon = \delta_t$, we then have the Navier-Stokes level momentum equation,

$$\partial_t(\rho u_\beta) + \partial_\alpha(\rho u_\alpha u_\beta) = -\partial_\beta P + \nu \partial_\alpha [\partial_\alpha(\rho u_\beta) + \partial_\beta(\rho u_\alpha)] + \rho a_\beta + \mathcal{E}_\beta \quad (5.25)$$

where the kinematic viscosity is

$$\nu = \Upsilon^{(2)} \frac{2\tau - g}{2g} \delta_t \quad \left(= c^2 \frac{2\tau - g}{6g} \delta_t \text{ in the } D2Q9 \text{ model} \right)$$

and various extra viscous-order terms are grouped together in \mathcal{E}_β :

$$\begin{aligned} \mathcal{E}_\beta &= \delta_t \frac{2\tau - g}{2g} \partial_\alpha \partial_{t_0}(\rho u_\alpha u_\beta) + \\ &\quad \delta_t \frac{2\tau - g}{2g} \partial_{t_0} \sum_i (J_i e_{i\beta} - F_i e_{i\beta}) + \delta_t \frac{1}{2} \partial_\alpha \sum_i (J_i e_{i\alpha} e_{i\beta} - F_i e_{i\alpha} e_{i\beta}) \\ &= \delta_t \frac{2\tau - g}{2g} \partial_\alpha \partial_{t_0}(\rho u_\alpha u_\beta) + \delta_t \frac{2\tau - g}{2g} \partial_{t_0} \left[\frac{c^2}{3} \partial_\beta(\rho^2 g) - \rho a_\beta \right] + \\ &\quad \delta_t \frac{1}{2} \partial_\alpha \left\{ b[u_\alpha u_\beta \mathbf{u} \cdot \nabla - \Upsilon^{(2)}(u_\alpha \partial_\beta + u_\beta \partial_\alpha)](\rho g^2) + \rho(u_\alpha a_\beta + u_\beta a_\alpha) \right\} \quad (5.26) \end{aligned}$$

This momentum equation is not Galilean invariant when density gradients are present, and contains some unphysical terms. If we assume that Mach number is small (M formally of the same order as ϵ in the expansion), then the extra term $\partial_\alpha \partial_{t_0}(\rho u_\alpha u_\beta)$ in \mathcal{E}_β can be further simplified as:

$$\begin{aligned} \partial_\alpha \partial_{t_0}(\rho u_\alpha u_\beta) &= \partial_\alpha \left\{ u_\alpha [\partial_{t_0}(\rho u_\beta) - u_\beta \partial_{t_0} \rho] + u_\beta \partial_{t_0}(\rho u_\alpha) \right\} \\ &= \partial_\alpha \left\{ -u_\alpha \partial_\gamma(\rho u_\beta u_\gamma) + u_\alpha u_\beta \partial_\gamma(\rho u_\gamma) - u_\beta \partial_\gamma(\rho u_\alpha u_\gamma) \right. \\ &\quad \left. - (u_\alpha \partial_\beta P + u_\beta \partial_\alpha P) + \rho(u_\beta a_\alpha + u_\alpha a_\beta) \right\} + \mathcal{O}(\delta_t) \quad (5.27) \end{aligned}$$

Terms of cubic order in Mach number ($\mathcal{O}(\mathbf{u}^3)$) can now be neglected, and we simplify to

$$\begin{aligned} \partial_\alpha \partial_{t_0}(\rho u_\alpha u_\beta) &= -\Upsilon^{(2)} \partial_\alpha (u_\alpha \partial_\beta \rho + u_\beta \partial_\alpha \rho) - b \Upsilon^{(2)} \partial_\alpha [u_\alpha \partial_\beta(\rho^2 g) + u_\beta \partial_\alpha(\rho^2 g)] \\ &\quad + \partial_\alpha [\rho(u_\beta a_\alpha + u_\alpha a_\beta)] + \mathcal{O}(\delta_t) + \mathcal{O}(\mathbf{u}^3) \quad (5.28) \end{aligned}$$

which allows the extra terms in the momentum equation to be rewritten as with

$$\mathcal{E}_\beta = \nu \partial_\alpha (u_\alpha \partial_\beta(\rho^2 g)) - \delta_t \frac{2\tau - g}{2g} \partial_{t_0}(\rho a_\beta) + \delta_t \tau \partial_\alpha [\rho(u_\beta a_\alpha + u_\alpha a_\beta)] \quad (5.29)$$

5.2 Summary

- This model is correct to first order (inviscid equations), but incorrect to second order (viscous equations). The continuity equation contains density diffusion terms, and the momentum equation various unphysical terms, somewhat similar to the ones appearing in the model of Swift and collaborators.

6 Intermolecular interaction model

This model was introduced by Qian, D’Humières and Lallemand [35] and further developed by He et al [19] and Chen et al. [11]. The starting point is the Boltzmann equation with a force field \mathbf{F} in the single relaxation time approximation (compare (5.2))

$$\frac{\partial f}{\partial t} + \xi \cdot \nabla f + \mathbf{F} \cdot \nabla_{\xi} f = -\frac{f - f^{eq}}{\lambda} \quad (6.1)$$

The force \mathbf{F} is not specified. In the case at hand we want eventually to recover a diffuse interface theory, and \mathbf{F} should therefore include density gradients. For different proposals, see [19] and [11]. The ansatz made in [35] is that the gradient $\nabla_{\xi} f$ can be approximated by $\nabla_{\xi} f^{eq}$. Since in the continuum we have a Maxwell-Boltzmann equilibrium distribution function.

$$f^{eq} = \frac{\rho}{(2\pi RT)^{D/2}} \exp \left[-\frac{(\xi - \mathbf{u})^2}{2RT} \right] \quad (6.2)$$

we further assume $\nabla_{\xi} f \approx -\frac{\xi - \mathbf{u}}{RT} f^{eq}$ and obtain an approximate Boltzmann equation in the form

$$\frac{\partial f}{\partial t} + \xi \cdot \nabla f = -\frac{f - f^{eq}}{\lambda} + \frac{\mathbf{F} \cdot (\xi - \mathbf{u})}{RT} f^{eq} \quad (6.3)$$

By discretizing on a lattice and integrating over one time step to first order we obtain a Lattice Boltzmann scheme as

$$f_i(\mathbf{x} + \mathbf{e}_i \delta_t, t + \delta_t) - f_i(\mathbf{x}, t) = -\frac{f(\mathbf{x}, t) - f^{eq}(\mathbf{x}, t)}{\tau} + \frac{\mathbf{F} \cdot (\mathbf{e}_i - \mathbf{u})}{\Upsilon^{(2)}} f_i^{eq} \delta_t \quad (6.4)$$

since in the isothermal models Lattice Boltzmann models $RT = \Upsilon^{(2)}$. Comparing we see that Luo’s model only differs from (6.4) by a definite choice of the force term F , and by a slightly different form of the external force term in (5.5).

The continuity and Navier-Stokes equations will be derived by the method of suc-

cessive approximations. We note the constraints

$$\begin{aligned}\sum_i \mathcal{F}_i &= \frac{F_\alpha}{\Upsilon^{(2)}} \left(\sum_i e_{i\alpha} f_i^{eq} - u_\alpha \sum_i f_i^{eq} \right) \\ &= \frac{F_\alpha}{\Upsilon^{(2)}} (\rho u_\alpha - u_\alpha \rho) = 0\end{aligned}\tag{6.5a}$$

$$\begin{aligned}\sum_i e_{i\beta} \mathcal{F}_i &= \frac{F_\alpha}{\Upsilon^{(2)}} \left(\sum_i e_{i\alpha} e_{i\beta} f_i^{eq} - u_\alpha \sum_i e_{i\beta} f_i^{eq} \right) \\ &= \frac{F_\alpha}{\Upsilon^{(2)}} (\Upsilon^{(2)} \rho \delta_{\alpha\beta}) = \mathbf{F} \rho\end{aligned}\tag{6.5b}$$

$$\begin{aligned}\sum_i e_{i\beta} e_{i\gamma} \mathcal{F}_i &= \frac{F_\alpha}{\Upsilon^{(2)}} \left(\sum_i e_{i\alpha} e_{i\beta} e_{i\gamma} f_i^{eq} - u_\alpha \sum_i e_{i\beta} e_{i\gamma} f_i^{eq} \right) \\ &= F_\alpha \left(\frac{\Upsilon^{(4)}}{(\Upsilon^{(2)})^2} \rho (\delta_{\alpha\beta} u_\gamma + \delta_{\alpha\gamma} u_\beta + \delta_{\beta\gamma} u_\alpha) - u_\gamma \rho \delta_{\alpha\beta} - \frac{\rho}{\Upsilon^{(2)}} u_\alpha u_\beta u_\gamma \right)\end{aligned}\tag{6.5c}$$

For the $D2Q9$ model the last expression simplifies to

$$\rho F_\beta u_\gamma + \rho F_\gamma u_\beta - \frac{3}{c^2} (\mathbf{F} \cdot \mathbf{u}) \rho u_\gamma u_\beta\tag{6.6}$$

We now sum the moments of the collision operator $\Omega_i \equiv -1/\tau (f_i - f_i^{eq})$ over i . The zeroth moment to first order reads

$$\partial_t \rho + \partial_\alpha (\rho u_\alpha) = \mathcal{O}(\delta_t)\tag{6.7}$$

and the next

$$\begin{aligned}&= \delta_t \left(\tau - \frac{1}{2} \right) \left[\partial_t \partial_t \sum_i f_i^{eq} + 2 \partial_t \partial_\alpha \sum_i e_{i\alpha} f_i^{eq} + \partial_\alpha \partial_\beta \sum_i e_{i\alpha} e_{i\beta} f_i^{eq} \right] - \\ &\quad \delta_t \tau \left(\partial_t \sum_i \mathcal{F}_i + \partial_\alpha \sum_i \mathcal{F}_i e_{i\alpha} \right)\end{aligned}\tag{6.8a}$$

$$\begin{aligned}&= \delta_t \left(\tau - \frac{1}{2} \right) \left[\partial_t (\partial_t \rho + \partial_\alpha (\rho u_\alpha)) + \partial_\alpha \left(\partial_t (\rho u_\alpha) + \partial_\beta \sum_i e_{i\alpha} e_{i\beta} f_i^{eq} \right) \right] - \\ &\quad \delta_t \tau \partial_\alpha (\rho F_\alpha)\end{aligned}\tag{6.8b}$$

Substituting in eqn.(6.8b), eqn.(6.7) and (6.10) derived below, we see that,

$$\begin{aligned}\mathcal{O}(\delta_t) &= -\delta_t \left(\tau - \frac{1}{2} \right) [\mathcal{O}(\delta_t) + \mathcal{O}(\delta_t) + \partial_\alpha(\rho F_\alpha)] - \delta_t \tau \partial_\alpha(\rho F_\alpha) \\ &= \mathcal{O}(\delta_t^2) - \frac{\delta_t}{2} \partial_\alpha(\rho F_\alpha)\end{aligned}$$

with this, the continuity equation becomes,

$$\partial_t \rho + \partial_\alpha(\rho u_\alpha) = -\frac{\delta_t}{2} \partial_\alpha(\rho F_\alpha) + \mathcal{O}(\delta_t^2) \quad (6.9)$$

To obtain the momentum equation, we multiply Ω_i by $e_{i\beta}$ and sum over i . To lowest order we get an Euler equation

$$\begin{aligned}0 &= \partial_t \sum_i e_{i\beta} f_i^{eq} + \partial_\alpha \sum_i e_{i\alpha} e_{i\beta} f_i^{eq} - \sum_i \mathcal{F}_i e_{i\beta} + \mathcal{O}(\delta) \\ \partial_t(\rho u_\beta) + \partial_\alpha(\rho u_\alpha u_\beta) &= -\partial_\beta P_0 + \rho F_\beta + \mathcal{O}(\delta)\end{aligned} \quad (6.10)$$

where $P_0 = \Upsilon^{(2)} \rho$ is the ideal gas pressure, P_1 given by $\rho F_\beta = -\partial_\beta P_1$ is a correction, and the total pressure is $P = P_0 + P_1$. We note that P can contain density gradient terms by a suitable choice of F . To next order in δ_t we find

$$\begin{aligned}\partial_t(\rho u_\beta) + \partial_\alpha(\rho u_\alpha u_\beta) &= -\partial_\beta P_0 + F_\beta \rho + \delta_t \left(\tau - \frac{1}{2} \right) \underbrace{(\partial_t + e_{i\alpha} \partial_\alpha)^2 \sum_i f_i^{eq} e_{i\beta}}_{\mathcal{A}_\beta} \\ &\quad - \delta_t \tau [\partial_t(\rho F_\beta) + \partial_\alpha(\rho F_\beta u_\alpha + \rho F_\alpha u_\beta)] + \mathcal{O}(\delta^2)\end{aligned} \quad (6.11)$$

In equation (6.11), a term $\partial_\alpha(\rho u_\alpha u_\beta \mathbf{F} \cdot \mathbf{u})$ has been neglected because the order of this term is $\mathcal{O}(\mathbf{u}^3)$, and F will eventually also have to be taken small. We have

$$\mathcal{A}_\beta = \partial_t^2 \sum_i f_i^{eq} e_{i\beta} + 2\partial_\alpha \partial_t \sum_i f_i^{eq} e_{i\beta} e_{i\alpha} + \partial_\alpha \partial_\gamma \sum_i f_i^{eq} e_{i\beta} e_{i\alpha} e_{i\gamma} \quad (6.12a)$$

$$= \partial_t^2(\rho u_\beta) + 2\partial_\alpha \partial_t (\Upsilon^{(2)} \rho \delta_{\alpha\beta} + \rho u_\alpha u_\beta) + \frac{\Upsilon^{(4)}}{\Upsilon^{(2)}} \partial_\alpha \partial_\gamma (\Delta_{\alpha\beta\gamma\theta}^{(4)} u_\theta) \quad (6.12b)$$

$$\begin{aligned}&= \partial_t [\partial_t(\rho u_\beta) + \partial_\alpha(\rho u_\alpha u_\beta)] + 2\Upsilon^{(2)} \partial_\beta [\partial_t \rho + \partial_\alpha(\rho u_\alpha)] + \\ &\quad \partial_\alpha \partial_t(\rho u_\alpha u_\beta) + \frac{\Upsilon^{(4)}}{\Upsilon^{(2)}} \partial_\alpha \partial_\alpha(\rho u_\beta) + 2 \left(\frac{\Upsilon^{(4)}}{\Upsilon^{(2)}} - \Upsilon^{(2)} \right) \partial_{\beta\alpha}^2(\rho u_\alpha)\end{aligned} \quad (6.12c)$$

For the $D2Q9$ model $\Upsilon^{(4)} = (\Upsilon^{(2)})^2$, and the last parenthesis in above vanishes. By substituting the continuity equation (6.7) and momentum equation (6.10), with the

accuracy to order of δ_t , into (6.12c) we have

$$\mathcal{A}_\beta = -\Upsilon^{(2)}\partial_t\partial_\beta\rho + \partial_t(\rho F_\beta) + \partial_\alpha\partial_t(\rho u_\alpha u_\beta) + \frac{\Upsilon^{(4)}}{\Upsilon^{(2)}}\partial_\alpha\partial_\alpha(\rho u_\beta) + \quad (6.12d)$$

$$= 2\left(\frac{\Upsilon^{(4)}}{\Upsilon^{(2)}} - \Upsilon^{(2)}\right)\partial_{\beta\alpha}^2(\rho u_\alpha) + \mathcal{O}(\delta_t) \quad (6.12e)$$

Substituting \mathcal{A}_β into (6.11) we have the momentum equation. For the $D2Q9$ model, as used by [35, 19, 11], we have some simplifications, and finally

$$\begin{aligned} \partial_t(\rho u_\beta) + \partial_\alpha(\rho u_\alpha u_\beta) &= -\frac{c^2}{3}\partial_\beta\rho + \nu\partial_\alpha[\partial_\beta(\rho u_\alpha) + \partial_\alpha(\rho u_\beta)] + F_\beta\rho + \\ \delta_t\left(-\frac{1}{2}\partial_t(\rho F_\beta) + \left(\tau - \frac{1}{2}\right)\partial_\alpha\partial_t(\rho u_\alpha u_\beta) - \tau\partial_\alpha(\rho F_\beta u_\alpha + \rho F_\alpha u_\beta)\right) \end{aligned} \quad (6.13)$$

with

$$\nu = \frac{c^2\delta_t}{3}\left(\tau - \frac{1}{2}\right) = \frac{\delta_x^2}{\delta_t}\left(\tau - \frac{1}{2}\right)$$

The unwanted terms in (6.13) are those of the second row, all of viscous order. Mach number M is the ratio of fluid velocity and speed of sound, which in Lattice Boltzmann models is proportional to lattice spacing c . In the method of successive approximations we have no expansion parameter ϵ with which to scale Mach number, but we can collect the terms in \mathcal{A}_β with smallest Mach number, and therefore neglect the term in (6.13) proportional to $(\tau - \frac{1}{2})$. The momentum equations up to viscous terms hence read

$$\begin{aligned} \partial_t(\rho u_\beta) + \partial_\alpha(\rho u_\alpha u_\beta) &= -\frac{c^2}{3}\partial_\beta\rho + \nu\partial_\alpha(\rho\partial_\beta u_\alpha + \rho\partial_\alpha u_\beta) + F_\beta\rho + \\ \frac{\delta_t}{2}(-\partial_t(\rho F_\beta) - \tau\partial_\alpha(\rho F_\beta u_\alpha + \rho F_\alpha u_\beta)) &+ \mathcal{O}(\delta_t^2) + \mathcal{O}(M^3) \end{aligned} \quad (6.14)$$

6.1 Summary

- This model is Galilean consistent with small Mach number only.
- Extra terms involving the force appear in the continuity and momentum equations. To have correct Navier-Stokes equations we therefore have to assume that \mathbf{F} is small, that is that we are close to the ideal gas.

7 An LBE scheme with high order accuracy

The purpose of this section is to show that it suffices to slightly modify the Lattice Boltzmann model of section 6 to get correct viscous order equations. The model which we will describe was introduced in [19] with a specific choice for the force term \mathcal{F} . In [19] it is not stated very explicitly that these modifications solve the problem of unphysical viscous-order terms, and we therefore belabour this point here.

We assume an isothermal situation. The starting point is to integrate equation (6.3) to better than linear order for \mathcal{F}

$$f_i(\mathbf{x} + \mathbf{e}_i \delta_t, t + \delta_t) - f_i(\mathbf{x}, t) = - \int_t^{t+\delta_t} \frac{f_i - f_i^{eq}}{\lambda} dt + \int_t^{t+\delta_t} \frac{\mathbf{F} \cdot (\mathbf{e}_i - \mathbf{u})}{RT} f_i^{eq} dt \quad (7.1)$$

Applying the trapezoidal rule to the second integral of right-hand side (but not to the first), we have the Lattice Boltzmann model (compare (6.4))

$$f_i(\mathbf{x} + \mathbf{e}_i \delta_t, t + \delta_t) - f_i(\mathbf{x}, t) = - \frac{f_i - f_i^{eq}}{\tau} \Big|_t + \frac{\delta_t}{2} \left[\frac{\mathbf{F} \cdot (\mathbf{e}_i - \mathbf{u})}{RT} f_i^{eq} \Big|_{t+\delta_t} + \frac{\mathbf{F} \cdot (\mathbf{e}_i - \mathbf{u})}{RT} f_i^{eq} \Big|_t \right] \quad (7.2)$$

where $\tau = \lambda/\delta_t$ is the non-dimensional relaxation time. The right-hand side involves the quantities evaluated at $t + \delta_t$. To eliminate this implicitness, we introduce the new variable:

$$h_i = f_i - \frac{\mathbf{F} \cdot (\mathbf{e}_i - \mathbf{u})}{2RT} f_i^{eq} \delta_t \quad (7.3)$$

in term of which the Lattice Boltzmann equation (7.2) is:

$$h_i(\mathbf{x} + \mathbf{e}_i \delta_t, t + \delta_t) - h_i(\mathbf{x}, t) = - \frac{h(\mathbf{x}, t) - h^{eq}(\mathbf{x}, t)}{\tau} + \frac{\mathbf{F} \cdot (\mathbf{e}_i - \mathbf{u})}{RT} f_i^{eq} \delta_t \quad (7.4)$$

Another interpretation of this procedure is that we evaluated all terms in equation (7.2) to the same order, with a collision operator for the original variables f_i that was actually not of the single relaxation time form. Instead it was something different that gave the single relaxation time form for the auxiliary variables h_i .

The equilibrium distribution for h contains terms up to three velocities, and reads

$$h_i^{eq} = \left[1 - 3 \frac{\mathbf{F} \cdot (\mathbf{e}_i - \mathbf{u})}{2c^2} \delta_t \right] f_i^{eq} \quad (7.5)$$

where f_i^{eq} is the standard choice of (3.6). The macroscopic variables given by eqn.(7.3) become

$$\rho = \sum_i h_i \quad (7.6)$$

$$\rho \mathbf{u} = \sum_i h_i \mathbf{e}_i + \frac{1}{2} \rho \mathbf{F} \delta_t \quad (7.7)$$

We now wish to compute the macroscopic equations by Chapman-Enskog expansions. We will for notational ease use both h_i and f_i in the expansions. Since these two quantities differ by a term to order ϵ (i.e. equations (7.3) and (7.5)), a choice has to be made for which of the two in which we count powers of ϵ . The natural choice is to count in orders of ϵ for f_i , but to perform the expansion in h_i , since the equations for h_i are explicit. This will mix orders for h_i , but in an unambiguous manner, as the expansion can be mapped back to order by order in ϵ for f_i . To ϵ^2 order accuracy we have

$$h_i \equiv h_i^{(0)} + \epsilon h_i^{(1)} + \epsilon^2 h_i^{(2)} = h_i^{eq} - \tau \left(\epsilon \mathcal{D}_t + \frac{\epsilon^2}{2} \mathcal{D}_t^2 \right) h_i + \epsilon \tau \mathcal{F}_i \quad (7.8)$$

where each term $h_i^{(n)}$ includes a term of order ϵ , and where

$$\mathcal{F}_i = \frac{\mathbf{F} \cdot (\mathbf{e}_i - \mathbf{u})}{RT} f_i^{eq}$$

Regrouping the right-hand side of eqn.(7.8) by the leading order of ϵ we have

$$\begin{aligned} h_i^{(0)} + \epsilon h_i^{(1)} + \epsilon^2 h_i^{(2)} &= h_i^{eq} - \epsilon \tau \mathcal{D}_t (h_i^{(0)} + \epsilon h_i^{(1)} + \epsilon^2 h_i^{(2)}) \\ &\quad - \epsilon^2 \frac{\tau}{2} \mathcal{D}_t^2 (h_i^{(0)} + \epsilon h_i^{(1)} + \epsilon^2 h_i^{(2)}) \\ &\quad + \epsilon \tau \mathcal{F}_i \end{aligned} \quad (7.9)$$

which is

$$\epsilon^0 : \quad h_i^{(0)} = h_i^{eq} \quad (7.10a)$$

$$\epsilon^1 : \quad h_i^{(1)} = -\tau \mathcal{D}_{t_0} h_i^{(0)} + \tau \mathcal{F}_i \quad (7.10b)$$

$$\epsilon^2 : \quad h_i^{(2)} = -\frac{\tau}{2} \mathcal{D}_{t_0}^2 h_i^{(0)} - \tau \partial_{t_1} h_i^{(0)} - \tau \mathcal{D}_{t_0} h_i^{(1)} \quad (7.10c)$$

Equation (7.10a) here determines $h_i^{(0)}$ to be of mixed zeroth and first order in ϵ , i.e. given by (7.5). Since the hydrodynamic modes belong to equilibrium distribution functions, we have for n greater than zero the constraints

$$\sum_i h_i^{(n)} = 0 \quad (7.11)$$

$$\sum_i h_i^{(n)} e_{i\alpha} = 0 \quad (7.12)$$

For the lowest order $h_i^{(0)} = h_i^{eq}$ we have on the other hand

$$\sum_i h_i^{(0)} = \sum_i \left[f_i^{eq} - \frac{\epsilon}{2} \mathcal{F} \right] = \rho \quad (7.13)$$

$$\sum_i h_i^{(0)} e_{i\alpha} = \sum_i f_i^{eq} e_{i\alpha} - \frac{\epsilon}{2} \sum_i \mathcal{F}_i e_{i\alpha} = \rho \mathbf{u} - \frac{\epsilon}{2} \rho \mathbf{F} \quad (7.14)$$

$$\begin{aligned} \sum_i h_i^{(0)} e_{i\alpha} e_{i\beta} &= \sum_i f_i^{eq} e_{i\alpha} e_{i\beta} - \frac{\epsilon}{2} \sum_i \mathcal{F}_i e_{i\alpha} e_{i\beta} \\ &= \Upsilon^{(2)} \rho \delta_{\alpha\beta} + \rho u_\alpha u_\beta - \frac{\epsilon}{2} \left[\rho F_\alpha u_\beta + \rho F_\beta u_\alpha - \frac{1}{\Upsilon^{(2)}} (F \cdot u) \rho u_\alpha u_\beta \right] \end{aligned} \quad (7.15)$$

Summing equation (8.21), using equations (7.13), (7.14) and (6.5a), gives

$$\partial_{t_0} \rho + \partial_\alpha (\rho u_\alpha) = \frac{\epsilon}{2} \partial_\alpha (\rho F_\alpha) \quad (7.16a)$$

To second-order in ϵ we get by summing (7.10c)

$$0 = -\frac{1}{2} \mathcal{D}_{t_0} \sum_i \mathcal{F}_i + \left(\frac{1}{2} - \tau \right) \mathcal{D}_{t_0} \sum_i h_i^{(1)} - \partial_{t_1} \sum_i h_i^{(0)} \quad (7.17a)$$

The first and the third terms is substituted from equations (6.5a), (6.5b) and (7.13). The second term is zero, and the two others give

$$\partial_{t_1} \rho = -\frac{1}{2} \partial_\alpha (\rho F_\alpha) \quad (7.17b)$$

Combining the first and the second order results for h_i by $\partial_t = \partial_{t_0} + \epsilon \partial_{t_1}$ we have the continuity equation:

$$\partial_t \rho + \nabla \cdot (\rho \mathbf{u}) = 0 \quad (7.18)$$

valid up to order ϵ^2 . In other words, there is no density diffusion term in this equation.

To first order, the momentum equation can be computed by multiplying equation (8.21) with $e_{i\beta}$, then summing over i , and using (7.15):

$$\partial_{t_0}(\rho u_\beta) + \partial_\alpha \left(\sum_i e_{i\alpha} e_{i\beta} h_i^{(0)} \right) = \frac{\epsilon}{2} \partial_{t_0}(\rho F_\beta) + \rho F_\beta \quad (7.19a)$$

which is

$$\begin{aligned} \partial_{t_0}(\rho u_\beta) + \partial_\alpha(\rho u_\alpha u_\beta) = & -\Upsilon^{(2)} \partial_\beta \rho + \frac{\epsilon}{2} \partial_\alpha(\rho F_\alpha u_\beta + \rho F_\beta u_\alpha) \\ & - \frac{\epsilon}{2\Upsilon^{(2)}} \partial_\alpha[(F \cdot u) \rho u_\alpha u_\beta] + \frac{\epsilon}{2} \partial_{t_0}(\rho F_\beta) + \rho F_\beta \end{aligned} \quad (7.19b)$$

The second order momentum equation are obtained by multiplying equation (7.10c) with $e_{i\beta}$ and summing over i , which leads to

$$0 = \partial_{t_1} \left(\sum_i h_i^{(0)} e_{i\beta} \right) + \frac{1}{2} \mathcal{D}_{t_0} \sum_i \mathcal{F}_i e_{i\beta} - \left(\frac{1}{2\tau} - 1 \right) \mathcal{D}_{t_0} \sum_i (h_i^{(1)} e_{i\beta}) \quad (7.20a)$$

Substituting in equations (7.14), (6.5b) and (6.5c), we have

$$\begin{aligned} \partial_{t_1}(\rho u_\beta) = & \left(\frac{1}{2\tau} - 1 \right) \partial_\alpha \sum_i (h_i^{(1)} e_{i\alpha} e_{i\beta}) \\ & - \frac{1}{2} \partial_{t_0}(\rho F_\beta) - \frac{1}{2} \partial_\alpha \left(\rho F_\alpha u_\beta + \rho F_\beta u_\alpha - \frac{1}{\Upsilon^{(2)}} (\mathbf{F} \cdot \mathbf{u}) \rho u_\alpha u_\beta \right) \end{aligned} \quad (7.20b)$$

Combining the first and the second order results, equations (7.19b) and (7.20b), for h_i by $\partial_t = \partial_{t_0} + \epsilon \partial_{t_1}$, we have,

$$\partial_t(\rho u_\beta) + \partial_\alpha(\rho u_\alpha u_\beta) = -\Upsilon^{(2)} \partial_\beta \rho + \epsilon \left(\frac{1}{2\tau} - 1 \right) \partial_\alpha \Pi_{\alpha\beta}^{(1)} + \rho F_\beta \quad (7.21)$$

where $\Pi_{\alpha\beta}^{(1)} = \sum_i e_{i\alpha} e_{i\beta} h_i^{(1)}$ is the first-order momentum flux tensor. We have

$$\begin{aligned} \Pi_{\alpha\beta}^{(1)} = & \sum_i e_{i\alpha} e_{i\beta} h_i^{(1)} = -\tau \sum_i e_{i\alpha} e_{i\beta} \mathcal{D}_{t_0} h_i^{(0)} + \tau \sum_i e_{i\alpha} e_{i\beta} \mathcal{F}_i \\ = & -\tau \left(\partial_{t_0} \sum_i e_{i\alpha} e_{i\beta} h_i^{(0)} + \partial_\gamma \sum_i e_{i\alpha} e_{i\beta} e_{i\gamma} h_i^{(0)} \right) + \tau \sum_i e_{i\alpha} e_{i\beta} \mathcal{F}_i \end{aligned}$$

and in which the sums can be simplified to

$$\sum_i e_{i\alpha} e_{i\beta} h_i^{(0)} = \sum_i e_{i\alpha} e_{i\beta} f_i^{(0)} - \frac{\epsilon}{2} \sum_i e_{i\alpha} e_{i\beta} \mathcal{F}_i \quad (7.22a)$$

$$\sum_i e_{i\alpha} e_{i\beta} e_{i\gamma} h_i^{(0)} = \sum_i e_{i\alpha} e_{i\beta} e_{i\gamma} f_i^{(0)} - \frac{\epsilon}{2} \sum_i e_{i\alpha} e_{i\beta} e_{i\gamma} \mathcal{F}_i \quad (7.22b)$$

The second terms in equations (7.22) are order of ϵ . Therefore, when we substitute them back into (7.21), these terms become ϵ^2 , and should be neglected. With this simplification $\Pi_{\alpha\beta}^{(1)}$ can be rewritten,

$$\Pi_{\alpha\beta}^{(1)} = -\tau \underbrace{\left(\partial_{t_0} \sum_i e_{i\alpha} e_{i\beta} h_i^{(0)} + \partial_\gamma \sum_i e_{i\alpha} e_{i\beta} e_{i\gamma} h_i^{(0)} \right)}_{\mathcal{D}_{t_0} \Pi_{\alpha\beta}^{(0)}} + \tau \sum_i e_{i\alpha} e_{i\beta} \mathcal{F}_i$$

where $\Pi_{\alpha\beta}^{(0)}$ is zeroth-order momentum flux tensor, and the convective derivative has been computed above in (5.23)

$$\mathcal{D}_{t_0} \Pi_{\alpha\beta}^{(0)} = \partial_{t_0} (\rho u_\alpha u_\beta) + \Upsilon^{(2)} [\partial_\alpha (\rho u_\beta) + \partial_\beta (\rho u_\alpha)] + \mathcal{O}(\delta_t) \quad (7.23)$$

$$= \Upsilon^{(2)} (\rho \partial_\alpha u_\beta + \rho \partial_\beta u_\alpha) + \rho (u_\alpha F_\beta + u_\beta F_\alpha) + \mathcal{O}(\delta_t) + \mathcal{O}(\mathbf{u}^3) \quad (7.24)$$

The first-order momentum flux tensor is therefore in the small Mach number approximation and to lowest order in ϵ equal to

$$\Pi_{\alpha\beta}^{(1)} = -\tau \Upsilon^{(2)} (\rho \partial_\alpha u_\beta + \rho \partial_\beta u_\alpha) \quad (7.25)$$

Substituting back the above results into (7.21) we have the momentum equation,

$$\partial_t (\rho u_\beta) + \partial_\alpha (\rho u_\alpha u_\beta) = -\Upsilon^{(2)} \partial_\beta \rho + \nu \partial_\alpha (\rho \partial_\alpha u_\beta + \rho \partial_\beta u_\alpha) + \rho F_\beta \quad (7.26)$$

where

$$\nu = \Upsilon^{(2)} \delta_t \left(\tau - \frac{1}{2} \right)$$

The final equation is the Navier-Stokes equation of an ideal gas with a force field. This force field can be an external force, but it can also be an internal force which describes capillary forces and non-ideal corrections. By comparison with (2.12) we see that it suffices to choose $\rho F_\beta = \Upsilon^{(2)} \partial_\beta \rho + \partial_\alpha T_{\alpha\beta}$ to describe isothermal flow with reversible stress tensor $T_{\alpha\beta}$. If we for instance wish to have the capillary stress tensor of (2.10) we should choose $\mathbf{F} = (\Upsilon^{(2)} + (\rho(f - \mu)' - K \nabla^2 \rho) \nabla \log \rho$, where the Laplacian and the gradient can be computed by discretization on the grid as in [44].

7.1 Summary

- This modified second-order Lattice Boltzmann equation gives rise to correct continuity and momentum equations to viscous order. The local and nonlocal pressure terms in a diffuse interface model can be introduced via the force field F .
- As in previous sections, a small Mach number expansion has been used, such that terms with three velocities are ignored. If desired this can be improved by using lattices with higher isotropy, following [36, 12].
- The model is isothermal.

8 Thermal lattice Boltzmann

8.1 General considerations

The equations of non-isentropic fluid flow are the continuity equation, the momentum equation, an equation of state and either the energy or entropy transport equations. In the context of diffuse interface theory, the energy and entropy transport equations are (2.15) and (2.16) above. One difficulty in constructing a Lattice Boltzmann scheme for non-isentropic flow is that we have so far not defined the internal energy or the entropy in terms of the distribution function.

Let us start by remarking that temperature can be defined in terms of the thermal kinetic energy as

$$\rho\varepsilon_T = \frac{1}{2} \sum_i f_i (e_{i\alpha} - \mathbf{u})^2 \quad (8.1)$$

where ρ is the mass density, and $\varepsilon_T = \frac{D}{2} RT$. In the rest of this section we will assume units have been chosen such that the gas constant R is one. A first requirement of a nonisotropic Lattice Boltzmann model is that temperature is not a constant, i.e. that the equilibrium distribution function (3.6) is modified to depend on T .

In equilibrium, the internal energy and entropy densities per unit volume are related by ([8], note that ε_I and s here stand for densities per unit mass)

$$d(\rho\varepsilon_I) = Td(\rho s) + \mu d\rho \quad (8.2)$$

The free energy density and the pressure on the other obey

$$\rho f = \rho(\varepsilon_I - Ts) \quad p = -\rho(f - \mu) \quad (8.3)$$

from which follows

$$d\varepsilon_I = -\frac{p}{\rho^2} d\rho + Tds \quad df = -\frac{p}{\rho^2} d\rho - sdT \quad (8.4)$$

If we with slight abuse of notation denote the free energy functional density per unit mass in the Cahn-Hilliard theory by

$$f[\rho, \nabla\rho, T] = f(\rho, T) + \frac{K}{2\rho} |\nabla\rho|^2 \quad (8.5)$$

where $f(\rho, T)$ is the free energy density of a homogeneous phase at density ρ and temperature T , then

$$p = -\rho^2 \frac{\delta f[\cdot]}{\delta \rho} \Big|_T = -\rho^2 \frac{\partial f}{\partial \rho} - \frac{K}{2} |\nabla \rho|^2 - K \rho \nabla^2 \rho \quad (8.6)$$

$$s = -\frac{\delta f[\cdot]}{\delta T} \Big|_\rho = -\frac{\partial f}{\partial T} \quad (8.7)$$

We note that p is equal to the diagonal component of the stress tensor in the second formulation discussed in section 2. We also note that in equilibrium, as we assume here, the non-diagonal terms of the stress tensor are zero, and the stress tensor can therefore be considered a non-equilibrium generalization of pressure. Entropy density in diffuse interface theory is just a function of ρ and T , while internal energy density depends on density gradients, i.e.

$$\varepsilon_I[\rho, \nabla \rho, T] = f[\cdot] + Ts = \varepsilon_I(\rho, s) + \frac{K}{2\rho} |\nabla \rho|^2 \quad (8.8)$$

where (ε_I, s) and (f, T) forms a Legendre transform pair:

$$\varepsilon_I(\rho, s) = f(\rho, T) + sT \quad s = -\frac{\partial f}{\partial T} \quad (8.9)$$

Thermodynamics in a system with mass motion follows from the free energy (see [8], section 8.4)

$$F(T, V, M, \mathbf{v}) = E - TS - \mathbf{P} \cdot \mathbf{v} \quad (8.10)$$

where \mathbf{P} is total momentum, and \mathbf{v} is the velocity of the rest frame of the fluid relative to the laboratory. We have $\mathbf{P} = M\mathbf{v}$, and the internal energy with total momentum \mathbf{P} is

$$E(S, V, M, \mathbf{P}) = E(S, V, M, 0) + \frac{1}{2} \frac{P^2}{M} \quad (8.11)$$

from which follows

$$F(T, V, M, \mathbf{v}) = F(T, V, M, 0) - \frac{1}{2} M \mathbf{v}^2 \quad (8.12)$$

The internal energy ε_I , the free energy density f and momentum density per unit mass ξ^3 in a system in motion are therefore related by

$$f(\rho, T, v) = \varepsilon_I(\rho, s, \xi) - Ts - \mathbf{v}^2 \quad \varepsilon_I(\rho, s, \xi) = \varepsilon_I(\rho, s, 0) + \frac{1}{2} \xi^2 \quad (8.13)$$

³In equilibrium the average momentum density per unit mass is the velocity v .

8.2 Non-isentropic Lattice Boltzmann

The equilibrium distribution function can be generalized to depend on temperature T as⁴

$$f_i^{eq} = \rho \left[1 + \frac{(e_i \cdot \mathbf{u})}{T} + \frac{(e_i \cdot \mathbf{u})^2 \Upsilon^{(2)}}{2T \Upsilon^{(4)}} - \frac{\mathbf{u}^2}{2T} \right] \quad (8.14)$$

We consider the continuous-time Boltzmann equation in the form

$$\frac{\partial f}{\partial t} + \xi \cdot \nabla f + \mathbf{F} \cdot \nabla_\xi f = \Omega(f) \quad (8.15)$$

where we identify $\xi = \partial_\xi \varepsilon(\rho, s, \xi)$ and $\mathbf{F} = -\partial_x \varepsilon(\rho(x), s(x), \xi)$. The left-hand side of (8.15) is Liouville's equation.

Here is a list of unanswered questions, that we feel is pertinent:

- What quantities should the collision operator Ω conserve ? Presumably mass, momentum and energy ?
- Can this then be done with a single relaxation time scheme ?
- The internal energy density ε_I is a function of ρ and s , but s is a function of ρ and T . Hence we can consider ε_I a function of ρ and T , both quantities which are naturally defined for the LBE. More explicitly, on the lattice, we could have

$$\varepsilon_I(x, t) = \varepsilon_I^0(\rho(x, t), T(x, t)) + \frac{K}{2\rho(x, t) \Upsilon^{(2)}} \sum_i w_i (\rho(x + e_i, t) - \rho(x, t))^2 \quad (8.16)$$

where $\varepsilon_I^0(\rho(x, t), T(x, t))$ stands for the bulk contribution to the internal energy, and the rest is a discrete approximation to the gradient terms.

How to construct an LBE scheme that conserves such a beast?

8.3 An attempt to generalize section 7

The LBE models in the previous sections are all isothermal. The objective of the present section is to describe work towards adapting the model described in section 7 to non-isothermal flow in a non-ideal fluid. The procedure follows the one for

⁴We assume units such that $R = 1$.

heat flow in an one-phase fluid with an ideal gas equation of state introduced by Alexander et al [1]. This procedure has a limitation, in that the Prandtl number can not be varied.

Several Lattice Boltzmann models of fluids with heat and mass flow have been introduced in the literature. Most of these share a defect of the one presented here, in that an equation is derived for the variation of temperature. The general equation of heat transfer (2.16) involves the variation of entropy density, and only reduces to an equation for temperature in the incompressible limit, see [24] § 50. We will not attempt a review of these models, but refer to [28, 29, 20, 12, 34]. The work of Palmer & Rector [34] deserves however to be singled out, as it uses a different approach where the Prandtl number can be changed, and simulates the energy transport equation (2.15), albeit without the effects of viscous heating and interstitial working.

Let us start with defining the thermal kinetic energy as

$$\rho\varepsilon = \frac{1}{2} \sum_i f_i (e_{i\alpha} - \mathbf{u})^2 \quad (8.17)$$

where ρ is the mass density, and $\varepsilon = \frac{D}{2}RT^5$. The equilibrium distribution function (3.6) only leads to isothermal flows, as ε is then $\Upsilon^{(2)}$. However, by changing the equilibrium function to be a function of ρ , u and T as in [1], we can have the

⁵ As an aside, let us note that a natural definition of entropy density per unit mass would be

$$\rho s = - \sum_i f_i \log f_i$$

where in equilibrium s is a function of ρ and T . A Lattice Boltzmann scheme starting from such a definition would naturally lead to a convective derivative of entropy density, as on the left-hand side of (2.16). The right hand side of (2.16) however only follows if the equilibrium function is chosen in an suitable form: this work is not attempted here.

following set of constraints:

$$\sum_i f_i^{eq} = \rho \quad (8.18a)$$

$$\sum_i f_i^{eq} e_{i\alpha} = \rho u_\alpha \quad (8.18b)$$

$$\sum_i f_i^{eq} e_{i\alpha} e_{i\beta} = \rho u_\alpha u_\beta + \rho RT \delta_{\alpha\beta} \quad (8.18c)$$

$$\sum_i f_i^{eq} e_{i\alpha} e_{i\beta} e_{i\gamma} = \rho u_\alpha u_\beta u_\gamma + \rho RT (\delta_{\alpha\beta} u_\gamma + \delta_{\alpha\gamma} u_\beta + \delta_{\beta\gamma} u_\alpha) \quad (8.18d)$$

$$\begin{aligned} \sum_i f_i^{eq} e_{i\alpha}^2 e_{i\beta} e_{i\gamma} = & \rho u_\alpha^2 u_\beta u_\gamma + \\ & (D+2)\rho(RT)^2 \delta_{\beta\gamma} + \rho(RT) u_\alpha^2 \delta_{\beta\gamma} + (D+4)\rho(RT) u_\beta u_\alpha \end{aligned} \quad (8.18e)$$

We now introduce an auxiliary quantity h_i as in section 7 which obeys

$$\sum_i h_i^{eq} = \rho \quad (8.19a)$$

$$\sum_i h_i^{eq} e_{i\alpha} = \rho u_\alpha - \frac{1}{2\Upsilon(2)} F_\alpha \rho(RT) \delta_t \quad (8.19b)$$

$$\sum_i h_i^{eq} e_{i\alpha} e_{i\beta} = \rho u_\alpha u_\beta + \rho RT \delta_{\alpha\beta} - \frac{1}{2\Upsilon(2)} (F_\beta u_\alpha + F_\alpha u_\beta) \rho RT \delta_t \quad (8.19c)$$

and a force term in the Lattice Boltzmann equations (7.4) which obeys

$$\sum_i \mathcal{F}_i = 0 \quad (8.20a)$$

$$\sum_i \mathcal{F}_i e_{i\beta} = \frac{1}{\Upsilon(2)} \rho RT F_\beta \quad (8.20b)$$

$$\sum_i \mathcal{F}_i e_{i\beta} e_{i\gamma} = \frac{1}{\Upsilon(2)} \rho RT (F_\beta u_\gamma + F_\gamma u_\beta) \quad (8.20c)$$

$$\sum_i \mathcal{F}_i e_{i\alpha}^2 e_{i\beta} = \frac{1}{\Upsilon(2)} ((D+2)\rho(RT)^2 F_\beta + \rho RT u_\alpha^2 F_\beta + 2\rho RT F_\alpha u_\alpha u_\beta) \quad (8.20d)$$

We compute the macroscopic equations with Chapman-Enskog expansions as in section 7. The constraints we use to close the equations are the zeroth, first and second moments of the collision operator, that is

$$\sum_i (f_i - f_i^{eq}) = \sum_i (f_i - f_i^{eq}) e_{i\alpha} = \sum_i (f_i - f_i^{eq}) e_{i\alpha} e_{i\alpha} = 0 \quad (8.21)$$

The first and second order in expansion parameter ϵ of the continuity equation read:

$$\partial_{t_0}\rho + \partial_\alpha(\rho u_\alpha) = \epsilon \frac{1}{2\Upsilon^{(2)}} \partial_\alpha(F_\alpha \rho RT) \quad (8.22a)$$

$$\partial_{t_1}\rho = -\frac{1}{2\Upsilon^{(2)}} \partial_\alpha(F_\alpha \rho RT) \quad (8.22b)$$

Combining the first and the second order results by $\partial_t = \partial_{t_0} + \epsilon \partial_{t_1}$ we have the continuity equation:

$$\partial_t \rho + \nabla \cdot (\rho \mathbf{u}) = 0 \quad (8.22c)$$

The first-order in ϵ of the momentum equation is:

$$\begin{aligned} \partial_{t_0}(\rho u_\beta) + \partial_\alpha(\rho u_\alpha u_\beta) = & -\partial_\beta P_0 + \epsilon \frac{1}{2\Upsilon^{(2)}} (F_\beta u_\alpha + F_\alpha u_\beta) \rho RT + \\ & \epsilon \frac{1}{2\Upsilon^{(2)}} \partial_{t_0}(F_\beta \rho RT) + \frac{1}{\Upsilon^{(2)}} F_\beta \rho RT \end{aligned} \quad (8.23)$$

with the ideal part of the pressure $P_0 = \rho RT$. The first-order in ϵ of the temperature equation is calculated by taking the second moment of (8.21).

$$\begin{aligned} \partial_{t_0} \left(\frac{D}{2} \rho RT + \frac{1}{2} \rho u_\alpha^2 \right) = \\ -\frac{1}{2} \partial_\beta \left(\rho u_\alpha^2 u_\beta + \frac{(2+D)}{\rho} RT u_\beta \right) + \frac{1}{\Upsilon^{(2)}} \rho RT F_\beta u_\beta + \frac{\epsilon}{4} \mathcal{D}_{t_0} \sum_i \mathcal{F}_i e_{i\alpha}^2 \end{aligned} \quad (8.24a)$$

To second-order, the temperature equation can be computed by the same way from equation (7.10c).

$$\begin{aligned} \partial_{t_1} \left(\frac{D}{2} \rho RT + \frac{1}{2} \rho u_\alpha^2 \right) = \\ \frac{1}{2} \left(\frac{1}{2\tau} - 1 \right) \mathcal{D}_{t_0} \sum_i h_i^{(1)} e_{i\alpha}^2 - \frac{1}{4} \mathcal{D}_{t_0} \sum_i \mathcal{F}_i e_{i\alpha}^2 + \frac{\epsilon}{4} \partial_{t_1} \sum_i \mathcal{F}_i e_{i\alpha}^2 \end{aligned} \quad (8.25a)$$

Combining the first and the second order of energy equations (8.24a),(8.25a) by $\partial_t = \partial_{t_0} + \epsilon \partial_{t_1}$, we have,

$$\begin{aligned} \partial_t \left(\frac{D}{2} \rho RT + \frac{1}{2} \rho u_\alpha^2 \right) = & -\frac{1}{2} \partial_\beta \left(\rho u_\alpha^2 u_\beta + \frac{(2+D)}{\rho} RT u_\beta \right) + \\ & \frac{1}{2} \left(\frac{1}{2\tau} - 1 \right) \epsilon \mathcal{D}_t \sum_i h_i^{(1)} e_{i\alpha}^2 + \frac{3}{c^2} \rho \epsilon F_\beta u_\beta \end{aligned} \quad (8.26)$$

In the equation above, a term with the second-order of ϵ : $\frac{\epsilon^2}{4}\partial_{t_1}\sum_i\mathcal{F}_ie_{i\alpha}^2$ is neglected. By using the first order of continuity and momentum equations (8.22a), (8.23) we have

$$\partial_t\left(\frac{1}{2}(\rho u_\alpha^2)\right) = -u_\alpha\partial_\alpha P + \frac{1}{2}u_\alpha^2\partial_\alpha(\rho u_\alpha) - u_\alpha\partial_\beta(\rho u_\alpha u_\beta) + \mathcal{O}(\epsilon) \quad (8.27)$$

where P stands for the full effective pressure given by $\partial_\alpha P = \partial_\alpha(\rho RT) - \frac{1}{\Upsilon(2)}\rho RT F_\alpha$.

Substituting equation (8.27) back into (8.26), we have,⁶

$$\begin{aligned} \partial_t\left(\frac{D}{2}\rho RT\right) + \partial_\beta(\rho \varepsilon u_\beta) = & -P\partial_\beta u_\beta + \frac{1}{2}\left(\frac{1}{2\tau} - 1\right)\epsilon\mathcal{D}_t\sum_i h_i^{(1)}e_{i\alpha}^2 + \\ & \frac{3}{c^2}\rho\varepsilon F_\beta u_\beta \end{aligned} \quad (8.28)$$

With the assumption that, the local energy is conservative. the second term in r.h.s of equation above can be rewritten as,

$$\mathcal{D}_t\sum_i h_i^{(1)}e_{i\alpha}^2 = -\tau\partial_\beta\left(\partial_t\sum_i h_i^{eq}e_{i\alpha}^2e_{i\beta} + \partial_\gamma\sum_i h_i^{eq}e_{i\alpha}^2e_{i\beta}e_{i\gamma} - \sum_i \mathcal{F}_ie_{i\alpha}^2e_{i\beta}\right)$$

The high-order terms of ϵ can be neglected as same as (7.22),

$$\mathcal{D}_t\sum_i h_i^{(1)}e_{i\alpha}^2 = -\tau\partial_\beta\left(\underbrace{\partial_t\sum_i f_i^{eq}e_{i\alpha}^2e_{i\beta}}_{d\Pi_\beta^{(3)}} + \underbrace{\partial_\gamma\sum_i f_i^{eq}e_{i\alpha}^2e_{i\beta}e_{i\gamma}}_{d\Pi_{\beta\gamma}^{(4)}} - \sum_i \mathcal{F}_ie_{i\alpha}^2e_{i\beta}\right)$$

Some terms in the equation above can be further simplified as:

$$\begin{aligned} \partial_t(\rho u_\alpha^2 u_\beta) = & u_\alpha u_\beta \partial_t(\rho u_\alpha) + u_\alpha u_\beta [\partial_t(\rho u_\alpha) - u_\alpha \partial_t \rho] + u_\alpha^2 [\partial_t(\rho u_\beta) - u_\beta \partial_t \rho] \\ = & -\partial_\gamma(\rho u_\alpha^2 u_\beta u_\gamma) - 2u_\alpha u_\beta \partial_\alpha P - u_\alpha^2 \partial_\beta P + \\ & \frac{12}{c^2 D}\rho\varepsilon u_\alpha u_\beta F_\alpha + \frac{6}{c^2 D}\rho\varepsilon u_\alpha^2 F_\beta \end{aligned} \quad (8.29)$$

⁶ From here on the calculations have not been rechecked thoroughly. The reader beware that the likelihood of accidental is not negligible, and, in particular, that there may be some confusion below in the use of RT and ε .

$$\begin{aligned}
\partial_t(\rho\varepsilon u_\beta) &= u_\beta \partial_t(\rho\varepsilon) + \varepsilon[\partial_t(\rho u_\beta) - u_\beta \partial_t \rho] \\
&= -\partial_\alpha(\rho\varepsilon u_\alpha u_\beta) - \frac{2}{D}\rho\varepsilon u_\beta \partial_\alpha u_\alpha - \frac{2}{D}\varepsilon \partial_\beta(\rho\varepsilon) + \\
&\quad \frac{6}{c^2 D}\rho\varepsilon F_\alpha u_\alpha u_\beta + \frac{6}{c^2 D}\rho\varepsilon^2 F_\beta
\end{aligned} \tag{8.30}$$

By using the equations (8.29), (8.30) and (8.20d), we have,

$$\begin{aligned}
\mathcal{D}_t \sum_i h_i^{(1)} e_{i\alpha}^2 &= -\tau \partial_\beta \left[\frac{4}{D}\rho\varepsilon u_\alpha (\partial_\beta u_\alpha + \partial_\alpha u_\beta) + \frac{4(D+2)}{D^2}\rho\varepsilon \partial_\beta \varepsilon - \right. \\
&\quad \left. \frac{8}{D^2}\rho\varepsilon u_\beta \partial_\alpha u_\alpha - \frac{6(D+4)}{c^2 D^2}\rho\varepsilon^2 F_\beta + \frac{6}{c^2 D}\rho\varepsilon F_\alpha u_\alpha u_\beta \right]
\end{aligned} \tag{8.31}$$

Substituting back equation (8.31) into (8.28), we have the energy equation,⁷

$$\begin{aligned}
\partial_t(\rho\varepsilon) + \partial_\beta(\rho\varepsilon u_\beta) &= -P\partial_\beta u_\beta + \partial_\beta(\kappa\partial_\beta \varepsilon) + \partial_\beta[\mu(\partial_\beta u_\alpha + \partial_\alpha u_\beta)u_\alpha] + \\
&\quad \partial_\beta(\lambda u_\beta \partial_\alpha u_\alpha) + \frac{3}{2}(\mu - \lambda)\partial_\beta(\varepsilon F_\beta) + \frac{3}{2}\mu\partial_\beta(F_\alpha u_\alpha u_\beta) + \frac{3}{c^2}\rho\varepsilon F_\beta u_\beta
\end{aligned} \tag{8.33}$$

where,

$$\mu = \frac{2}{D}\rho\varepsilon \left(\tau - \frac{1}{2} \right) \tag{8.34}$$

$$\lambda = -\frac{4}{D^2}\rho\varepsilon \left(\tau - \frac{1}{2} \right) \tag{8.35}$$

$$\kappa = \frac{2(2+D)}{D^2}\rho\varepsilon \left(\tau - \frac{1}{2} \right) \tag{8.36}$$

⁷ Footnote added: the equation of heat transfer in an incompressible fluid is (compare [24], eq. 50.2)

$$\rho D_t T = (\kappa/c_p)\Delta T + (\mu/2c_p)(\partial_\alpha v_\beta + \partial_\beta v_\alpha)^2 \tag{8.32}$$

where c_p is the specific heat at constant pressure. Equation (8.31) definitely contains more terms than this. Among these are terms of the right hand side of the energy equation (2.15), but also other terms. The meaning and validity of equation (8.31) is unclear. The calculations of this section however well illustrate that constructing thermal multiphase LBE models is not trivial.

9 Optimizing a diffuse interface model

We have seen that a diffuse interface theory of a multiphase flow involves a free energy density, which in the isothermal situation is a function of density. For a real fluid this energy density is an observable quantity in a homogeneous phase. In the interface, the real form of the free energy density is less constrained by experimental data, and one has some freedom of choice in the model.

The numerical difficulty in solving a problem of a multiphase flow with an interface by a Lattice Boltzmann, or other diffuse interface method, depends on the interface thickness ξ , since the grid size Δx cannot be less than ξ ⁸. A typical density profile in an interface is shown in Fig. 1. The interface thickness is approximatively related

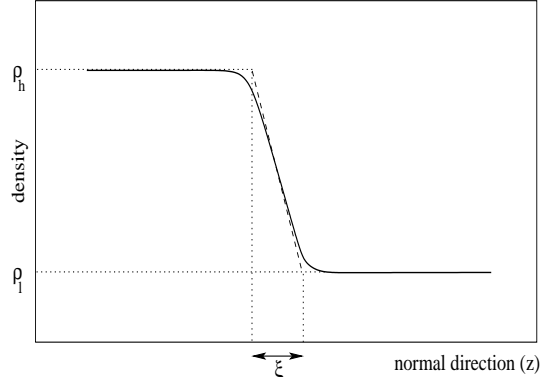


Figure 1: Density profile

to the density gradient and the density difference between the two phases as

$$\frac{d\rho}{dz} \sim \frac{\rho_h - \rho_l}{\xi} \quad (9.1)$$

In a typical application $(\rho_h - \rho_l)$ is something we would like to keep fixed. Similarly, surface tension, and the physical properties of the two phases, are also things we would like to keep fixed.

From (2.9) and (2.18) (or (2.22)) follow that the surface tension can be written

$$\sigma = 2 \int_{z_-}^{z_+} (\rho f - \rho \mu + P) dz \quad (9.2)$$

⁸We assume a homogeneous grid, although that is not necessary in the Lattice Boltzmann method. An adaptive grid would involve some sort of front-tracking, and lead to problems outside the scope of this paper.

where z is a coordinate in the normal direction to an interface. The thermodynamic rule that the pressure and the chemical potential in two coexisting phases are equal, implies that the two phases (at densities ρ_1 and ρ_2 and bulk free energy densities ψ_1 and ψ_2) are related by the the double tangent construction of Fig.2. Let us introduce

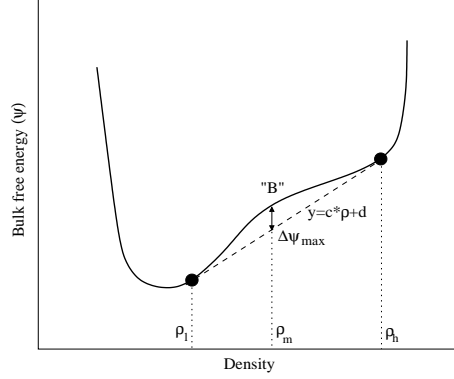


Figure 2: A typical bulk free energy for a one component system

$\tilde{\psi}$, a linear interpolation of ψ , and write

$$\psi(\rho) = \tilde{\psi}(\rho) + \Delta\psi \quad \tilde{\psi} = \psi_1 + \mu(\rho - \rho_1) \quad (9.3)$$

With P the pressure in one of the homogeneous phases (for instance the 1 phase) we have in the interface region

$$\Delta\psi = \psi - \rho\mu + P \quad (9.4)$$

Equations (9.4) and (9.2) express that the excess free energy per unity of surface area of the interface is the excess free energy density in the interface region, integrated over the normal direction to the surface.

By using (9.1) we have

$$\sigma \sim \overline{\Delta\psi} \xi \quad (9.5)$$

where $\overline{\Delta\psi}$ is the mean excess free energy density in the interface region. Equation (2.9) on the other hand means

$$\overline{\Delta\psi} \sim K \frac{(\Delta\rho)^2}{\xi^2} \quad (9.6)$$

If the parameters to be kept fixed are σ and $\Delta\rho$, the one to be taken large is ξ , and the two parameters to be freely adjusted are K and $\overline{\Delta\psi}$ we have

$$\overline{\Delta\psi} \sim \frac{\sigma}{\xi} \quad K \sim \frac{\sigma\xi}{(\Delta\rho)^2} \quad (9.7)$$

To have ξ large, all else constant, one should therefore take $\overline{\Delta\psi}$ small and K large in the same proportion. The optimization problem addressed here is that of maximizing the density difference $\Delta\rho$, while keeping σ and ξ constant. $\overline{\Delta\psi}$ should then be held constant, while K should be inversely proportional to the square of $\Delta\rho$.

If we use a standard equation of state, such as the Van der Waals

$$P = \frac{\rho RT}{1 - b\rho} - a\rho^2 \quad \psi = \rho RT \ln \left(\frac{\rho}{1 - b\rho} \right) - a\rho^2 \quad (9.8)$$

the interface is thin, except close to the critical point, where however also the density difference between the two phases is small. Let us now consider the following “stretched” Van der Waals free energy, where the free energy curve is a straight line in the interval $[\rho_m - \Delta\rho, \rho_m + \Delta\rho]$, see Fig.2

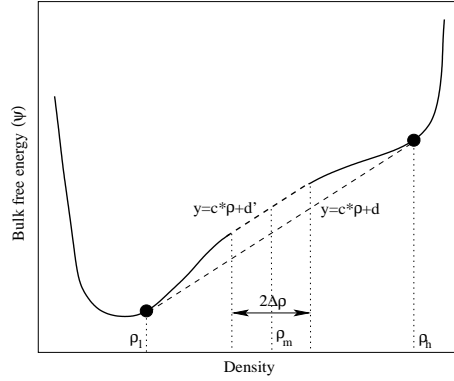


Figure 3: A modification of bulk free energy for a one component system

$$\begin{cases} \psi_{new}(\rho) = \psi(\rho + \Delta\rho) & \rho \leq \rho_m - \Delta\rho \\ \psi_{new}(\rho) = \Delta\psi(\rho_m) + \mu(\rho - \rho_m) & \rho_m - \Delta\rho < \rho < \rho_m + \Delta\rho \\ \psi_{new}(\rho) = \psi(\rho - \Delta\rho) & \rho \geq \rho_m + \Delta\rho \end{cases} \quad (9.9)$$

Here ρ_m is the point of maximum excess free energy in the Van der Waals energy, and $\Delta\psi(\rho_m)$ is that excess energy. The effect of the perturbation is hence to move

apart the two densities of the coexisting phases.

The pressure in the stretched region is constant, and equal to the pressure in the two coexisting phases, see Fig.4 With this method, the density difference between

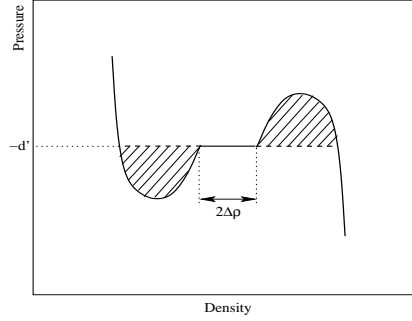


Figure 4: A modification of equation of state

two phases can be changed quite easily, see Fig.5

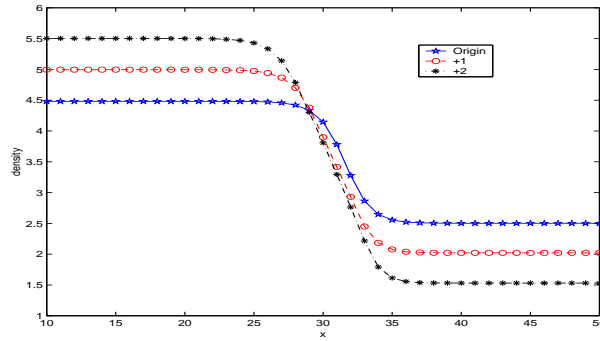


Figure 5: Solutions with the different modification of $\Delta\rho = 0, 1, 2$

9.1 Summary

The form of the free energy in the interface determines surface tension and interface thickness. In a physical liquid these two quantities vary together with temperature and pressure in the phase diagram, and the interface is only thick close to the critical point. In a numerical procedure that demands the resolution of the interface, the interface thickness sets a limit on the smallest grid size that can be used.

The free energy in the interface can be modified, so as to e.g. keep surface tension and interface thickness constant, while increasing the density difference. From a numerical point of view, this procedure amounts to smearing the interface, keeping all else constant, i.e., in some sense the opposite approach to the use of singular interface theory. The advantages of the procedure are that problems which are difficult in the singular interface theory can be readily simulated (see list on page 4), in particular there is no difficulty with changes in topology. The drawback is that the simulations will be of a fictitious liquid with thick interfaces, and the validity of the simulation results would in the end have to be established from comparison with experiments.

10 List of symbols and conventions used

c_s	speed of sound
\mathbf{e}_i	lattice (particle) discrete velocities
f_i	discrete one-particle distribution function
P, P_{ij}	pressure and pressure tensor
R	gas constant
T	temperature
\mathbf{u}	macroscopic velocity
ρ	density
ε_I	internal energy
ε_K	kinetic energy
ϵ	expansion parameter
$\mathcal{O}(\dots)$	on the order of ...
w_i	weight of different sub-lattices
α, β	space coordinates
$\delta_{\alpha\beta}, \Delta^{(2n)}$	Kronecker delta symbols
δt	time scale
δx	length scale
η	first viscosity
κ	elastic stress coefficient
λ	bulk viscosity
ν	kinetic viscosity
ξ	second viscosity
σ	surface tension
τ	relaxation time
ψ	bulk free-energy density
Ψ	non-local free-energy functional
Ω_i	collision operator
\mathbf{F}	force term
Π	momentum flux tensor
$\Upsilon^{(2)}$	Lattice constant, second order tensors
$\Upsilon^{(4)}$	Lattice constant, fourth order tensors

Greek indices (α, β, \dots) generally label spatial coordinates, while Latin indices (i, j, \dots) label lattice vectors. The Einstein convention of summing repeated spatial indices has been used throughout the paper. Hence, if $\mathbf{a} = \{a_1, \dots, a_D\}$ and $\mathbf{b} =$

$\{b_1, \dots, b_D\}$ are two vectors, then $a_\alpha b_\alpha = \mathbf{a} \cdot \mathbf{b} = \sum_{\alpha=1}^D a_\alpha b_\alpha$. Repeated indices labeling lattice coordinates are not summed over, unless so indicated.

Acknowledgements

We thank PSCI for financial support, and Profs. T. Bohr, A. Nepomnyaschy and S. Succi for comments and communications.

We thank again Massimo Vergassola for many valuable discussions, useful advice, and for taking his time to do the calculations of sections 4 and 5 above with us.

References

- [1] Alexander, F. J., Chen, S., Sterling, J.D., Lattice Boltzmann thermohydrodynamics *Physical Review E*, 1993, v. 47, 4, pp. R2249-R2252
- [2] Anderson D.M., McFadden G.B. & Wheeler A.A., “Diffuse-interface methods in fluid mechanics” *Ann. Review Fluid Mech.*, 1998, **30**, 139-65.
- [3] Benzi, R., Succi, S., Vergassola, M., The lattice Boltzmann equation: Theory and applications, *Physics Reports*, 1992, v. 222, 3, pp.145-197.
- [4] B. M. Boghosian, P. V. Coveney, A particulate basis for an immiscible lattice-gas model, *cond-mat/9911340*
- [5] B. M. Boghosian, J. Yepez, P. V. Coveney, A. J. Wagner, Entropic lattice Boltzmann methods, *cond-mat/0005260*
- [6] Cahn, J.W. and Hilliard, J.E., Free energy of a nonuniform system. I. Interface free energy *Journal Chemical Physics*, 1958, v.58, pp.258.
- [7] Cao, N., Chen, S., Jin, S. Martinez, D., Physical symmetry and lattice symmetry in the lattice Boltzmann method, *Physical Review E*, 1997, v.55, 1, pp.R21.
- [8] Chaikin & Lubensky Principles of Condensed Matter Physics.
- [9] Chen, H., Boghosian, B. M., Coveney P. V. A Ternary Lattice Boltzmann Model for Amphiphilic Fluids *cond-mat/9910369*
- [10] Chen, S., and Doolen, G.D., Lattice Boltzmann Method for Fluid Flows, *Annual Reviews of Fluid Mechanics*, 1998, v.30, 4, pp.329-364.
- [11] Chen, Y., Teng, S., Shukuwa, T., Ohashi, H., Lattice-Boltzmann simulation of two-phase fluid flows, *International Journal of Modern Physics C*, 1998, v.9, 8, pp.1383-1391
- [12] Chen, Y., Ohashi, H., Akiyama M. Thermal lattice Bhatnagar-Gross-Krook model without nonlinear deviations in macrodynamic equations *Phys. Rev. E*, 1994, v.50, pp.1776-2783.
- [13] Cornubert, R., D’Humières, D., Levermore, D., A Kundsén layer theory for lattice gas, *Physica D* 47, 1991, pp. 241-259

- [14] Delhay J.M., 1974. Jump conditions and entropy sources in two-phase systems. Local instant formulation *Int. J. Multiphase Flow* **1**, 395-409.
- [15] Evans, R., The nature of the liquid-vapour interface and other topic in the statical mechanics of non-uniform, classical fluids, *Advances in physics*, 1979, v.28, pp.143-200
- [16] Gonnella, G., Orlandini, E., and Yeomans, J.M., Lattice-Boltzmann Simulations of Complex Fluids, *International Journal of Modern Physics C*, v.8, 4, pp.783-792.
- [17] He, X., and Dembo, M., 1996, Some Progress in Lattice Boltzmann Method. Part I. Nonuniform Mesh Grids, *Journal of Computational Physics*, v.129, pp.357-363.
- [18] He, X. and Luo, L., Lattice Boltzmann model for the incompressible Navier-Stokes equation, *Journal of Statistical Physics*, 1997, v.88, 3/4, pp.927-944.
- [19] He, X., Shan, X., Doolen, G., Discrete Boltzmann equation model for non-ideal gases, *Physical Review E*, 1998, v.57, 1, pp.R13-R16.
- [20] Ihle T. & Kroll D.M., Thermal lattice-Boltzmann methods for non-ideal gases with potential energy- *Comp. Phys. Commun.*, 2000, v.129, 1-12.
- [21] Jasnow D. & Vinals J., Course-grained description of thermo-capillary flow *Phys. Fluids*, 1996, v.8, 1, 660-668.
- [22] Hou., S., Shan, X., Zou, Q., Doolen, G.D., and Soll, W.E., Evaluation of Two Lattice Boltzmann Models for Multiphase Flows, *Journal of Computational Physics*, v.138, 4, pp.695-713.
- [23] Korteweg D.J., 1901, Sur la forme que prennent ..., *Arch. Néérl. Sci. Extactes Nat. Ser. II*, v.6, pp.1-24
- [24] Landau, L.D., and Lifschitz, E.M., Theoretical Physics, v.VI, "Fluid Mechanics", Second Edition, 1987, Pergamon.
- [25] D. Mc Laughlin, G.C. Papanicolaou & O. Pironneau, *SIAM Journal of Appl. Math.*, 1985, v.45, 780
- [26] Liboff, R.L., *Kinetic theory*, Second Edition, John Wiley & Sons, 1998.
- [27] Li-Shi Luo, Unified Theory of Lattice Boltzmann Models for Nonideal Gases, *Physical Review Letters*, 24 August 1998, v.81, 8, pp.1618-1621.

- [28] Malevanets A. & Kapral R. *J. Chem. Phys.*, 1999, v.110, 8605.
- [29] McNamara G.R., Carcia A.L. & Alder B.J., *J. Stat. Phys.*, 1995, v.81, 395.
- [30] Minh Do-Quang Master thesis, Parallelized Lattice Boltzmann Modeling of Bubble Dynamics, *Royal Institute of Technology*, December 1999
- [31] Nadiga, B.T., Zaleski, S., Investigation of a two-phase fluid model *Eur. Journal Mechanics*, 1996, v.15, pp.885
- [32] Nannelli, F. and Succi, S., 1992, The Lattice Boltzmann Equation on Irregular Lattices, *Journal of Statistical Physics*, v.68, 3/4, pp.401-407.
- [33] Nourgaliev, R.R., Dinh, T.N., Hattori, S., and Sehgal, B.R., Free-energy based lattice Boltzmann model of phase transition in isothermal non-ideal fluid, *CD-ROM Proceedings of the Ninth International Topical Meeting on Nuclear Reactor Thermal Hydraulics (NURETH-9)*, San-Fransisco, California, October 3-8, 1999.
- [34] Palmer B.J. and Rector D.R. Lattice-Boltzmann algorithm for simulating thermal two-phase flow *Physical Rev. E*, 2000, v.61, 5, pp.5295-5306.
- [35] Qian, Y.H., D'Humières, D., and Lallemand, P., Lattice BGK Models for Navier-Stokes Equation, *Europhysics Letters*, v.17, 6, pp.479-484.
- [36] Qian, Y.H., Orszag S. *Europhysics Lett.*, 1993, v.21, pp. 225.
- [37] Rothman D.H., Keller J.M. *Phys. Rev. Lett.*, 1988, v.56, pp. 889.
- [38] Rowlinson, J.S., and Widom, B., Molecular Theory of Capillarity, *The International Series of Monographs on Chemistry*, Claderon Press, Oxford, 1982.
- [39] Shan, X. and Chen, H., Lattice Boltzmann Model for Simulating Flows with multiple Phases and Components, *Physical Review E*, March 1993, v.47, 3, pp.1815-1819.
- [40] Shan, X. and Doolen, G., Multicomponent Lattice-Boltzmann Model with Interparticle Interactions, *Journal of Statistical Physics*, v.81, 1/2, pp.379-393.
- [41] Shan, X. and Doolen, G., Diffusion in a Multicomponent Lattice Boltzmann Equation Model, *Physical Review E*, October 1996, v.54, 4, pp.3614-3620.

- [42] Shikhmurzaev Yu.D., Moving contact lines in liquid/liquid/solid systems *J. Fluid Mech.*, 1997, v.334, pp.211-249.
- [43] Sterling, J. D. and Chen, S., Stability Analysis of Lattice Boltzmann Methods, *Journal of Computational Physics*, **123**, 196-206.
- [44] Swift, M.R., Osborn, W.R. and Yeomans, J.M., Lattice Boltzmann Simulations of Nonideal Fluid, *Physical Review E*, November 1996, v.54, 5, pp.5041-5052.
- [45] Swift, M.R., Orlandini, E., Osborn, W.R. and Yeomans, J.M., Lattice Boltzmann Simulations of Liquid-Gas and Binary Fluid Systems, *Physical Review Letters*, 31 July 1995, v.75, 5, pp.830-833.
- [46] Teng, S., Chen, Y., Ohashi, H., Lattice Boltzmann simulation of multiphase fluid flows through the total variation diminishing with artificial compression scheme, *Int. Journal of Heat and fluid flow*, 2000, v.21, pp.112-121
- [47] Van der Waals, J.D., The thermodynamic theory of capillarity under the hypothesis of a continuous variation of density, (English translation) *Journal of statistical physics*, 1979, v.20, pp.197-244.
- [48] Yang, A.J.M., Fleming, P.D, Gibbs, J.H, Molecular theory of surface tension, *Journal of Chemical Physics*, 1976, v.64(9), pp.3732-3747
- [49] Ziegler, D.P, Boundary conditions for lattice Boltzmann simulations, *Journal of Statistical Physics*, 1993, v.71(5/6), pp.1171-1177.
- [50] Wagner, A.J., and Yeomans, J.M., Effect of Shear on Droplets in a Binary Mixture, *International Journal of Modern Physics C*, v.8, 4, pp.773-782.
- [51] Widom, B., Phase transition and critical phenomena, 1972, vol.2, pp.79.
- [52] Wolfram, S., Cellular Automata Fluids 1: Basic Theory, *Journal of Statistical Physics*, v.45(3/4), pp.471-526.