

Modelling of electrokinetic flow using the lattice-Boltzmann method

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Abstract

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The lattice-Boltzmann method is used to model flow in electrokinetic systems. A modelling approach based on the coupling of Navier-Stokes, Nernst-Planck and Poisson's equation of electrostatics is utilised. Three lattice-Boltzmann methods are formulated for the three equations respectively.

The method is implemented in C++ with the aim of being high performing. Topics as locality, instruction pipelines and parallel computing are considered. The implementation is tested for a number of classic examples with known solutions, e.g. Taylor-Green vortex flow, an Helmholtz equation and an advection-diffusion situation. The computed solutions agree well with the analytic solutions.

The physical systems modelled consists mainly of various charged channel flows of ionic solutions. Electrokinetic effects, such as electroosmosis and the electroviscous effect are studied. This is done in thin channels where the thickness of the electrical double layers is comparable to the channel dimension. The electroviscous effect is shown to slow the flow down and a local minimum is found in the velocity profile for thick enough double layers. Other more complicated systems are also studied; electroosmotic flow in a channel with heterogeneously charged walls and flow in an array of charged squares.

Keywords: lattice-Boltzmann, electrokinetics, electrohydrodynamics, Nernst-Planck, Poisson-Boltzmann, high performance computing.

1 Introduction

This thesis deals with modelling of physical problems in the interdisciplinary field of hydrodynamics and electrostatics. The tool used for realising this is the new and promising but somewhat immature lattice-Boltzmann method. This is a method that is still under development but is today used in practical applications both in industry and academy.

1.1 Background

There is currently an ongoing project at the mathematics faculty of Chalmers University in producing a modelling package that should be able to deal with transport of various liquids and particles through complicated structures. The method of choice has fallen upon the lattice-Boltzmann method for its suitable characteristics in the systems of interest.

This work aims to investigate the possibility and procedure for taking electrical effects into account in the modelling of charged fluids. More theoretical questions about the method itself and of the physics involved is of interest as well as how the method may be effectively implemented on a computer.

From both industry and academy, there is a demand on the modelling of this kind of physics. For instance, in medical sciences, accurate modelling of transport of charged fluids is a fundamental ingredient in understanding biological systems and to be able to manipulate them. As a consequence of the always so present desire of more environmental friendly ways of using the planet, automotive industry are now engineering electrical cars. A great challenge is to produce high performing and durable batteries, the ability to accurately model the electrolytes in the batteries is indeed an advantage in achieving this.

1.2 Outline

The text is structured in five main chapters. In chapter ??, the physics involved and the equations of interest are presented. This is followed by chapter 2.7 where the lattice-Boltzmann method is formulated for the different equations of interest. Also an introduction to the method as well as some discussion on different boundary conditions is given here. In chapter ??, the implementation of the method is discussed together with some general aspects that is important to have in mind in order to produce a high performing code. The implementation is then tested for classic examples with known solutions in chapter ??. Finally some results in electrokinetics are presented and discussed in chapter ??. Here, the focus is rather on the physics of the simulated systems than on LBM aspects of the problems. These aspects, such as grid dimensions, how LBM parameters relate to physical quantities etc. are discussed for the problems in chapter ??.

1.3 Previous work

An extensive treatment of both theory and experiments in the field of electrokinetics is carried out in [?]. Mainly the Poisson-Boltzmann model is used in the modelling but

also in some situations, the model used in this work based on the coupling of Navier-Stokes, Nernst-Planck and Poisson's equation of electrostatics is discussed. Also in [?], this modelling approach is used. However, the computational model is not the Lattice-Boltzmann method (LBM).

There are a lot of formulations of the LBM for the Navier-Stokes equations as the method typically is used in the modelling of fluid dynamics. Not so common are formulations for the Nernst-Planck and Poisson's equation. However there are a few, e.g. in [?] and [?] formulations for the Poisson's equation is discussed. In [?] a complete formulation for the three equations are presented together with some example simulations of electrokinetic systems. The formulation presented in [?] will not be completely the same as the one used in this work as is discussed in later chapters of this text.

2 Physical model

Electrohydrodynamics involves the study of electric phenomena on fluid flow. How fluids carrying electrical charges (electrolytes) react upon external electrical fields or interact with charged objects are examples of problems that arise in this field.

As a charged object is brought into contact with an electrolyte it is, qualitatively, easily deduced that ions with a sign of charge opposite to that of the object will be attracted to the object and ions with the same sign of charge will be repelled. These two distinct categories of ions will from hereon be referred to as counter- and co-ions respectively. In this case, for a neutral electrolyte, a surplus of counter-ions will be present in the direct vicinity of the object and a surplus of co-ions will be present at some other location further from the object. The part of the net charged fluid nearest to the charged boundary is often referred to as an electrical double layer (EDL) [?].

Traditionally, the physical description of EDLs and electrokinetics in general is based on the Poisson-Boltzmann model. However the PB model includes some crude assumptions on the system and is sometimes not an accurate choice, see section 2.4 for a brief description. Therefore, more sophisticated modelling approaches has been proposed e.g. in [?]. This paper is based on such an approach which is described further in this section.

2.1 Model overview

To model the fluid motion of a charged fluid under influences of electrostatic forces, a coupling between different models is considered.

The electric field and potential in the system are obtained from solving Poisson's equation (PE) for electrostatics (section 2.2) with a given charge density. This charge density is obtained from a set of Nernst-Planck (NP) equations (section 2.3) by including effects on the charge distribution from the electric field previously mentioned, diffusion and advection. One NP equation is solved for each different ion species in the solution. For instance in a 1:1 solution, two equations are solved one for the positive and one for the negative ions respectively. advective charge flux is given from the velocity field in the fluid that is obtained by solving the Navier-Stokes (NS) equations (section 2.5).

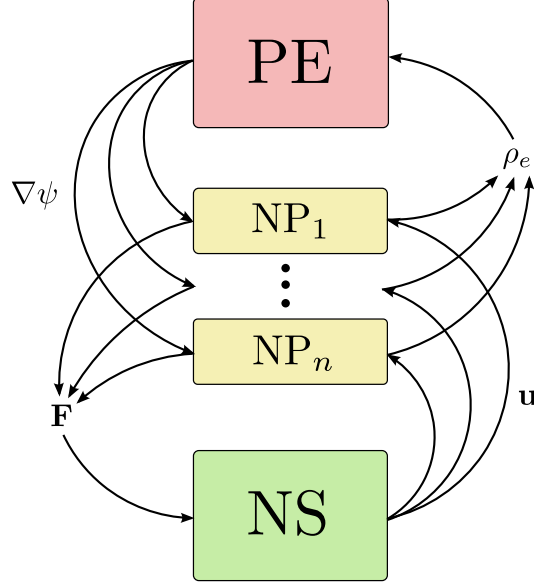


Figure 1: Visualisation of the coupling between the three equations present in the model. Poisson’s equation (PE), the set of Nernst-Planck equations (NP₁ ... NP_n) for the different ion species and the Navier-Stokes equations (NS). The dependencies have also be marked with arrows indicating what quantities for a certain equation that are needed from an other.

Forces due to present electric fields on net charged areas of the fluid also couples the NS equations to the NP equation. More about the force coupling is discussed in sections. 2.6 and 2.7. The coupling between the different equations are visualised in fig. 1.

2.2 The potential - Poisson’s equation

To be able to model the flow dynamics of liquids in a channel with present EDLs, the potential and charge distribution in the channel must be determined. These quantities are mutually related through Poisson’s equation for electrostatics:

$$\nabla^2\psi = -\frac{\rho_e}{\epsilon_r\epsilon_0} \quad (1)$$

where ψ is the electrical potential, ρ_e the electrical charge density, ϵ_r is the relative permittivity and ϵ_0 the vacuum permittivity. Under certain assumptions, the charge density may be explicitly determined as a function of the potential distribution, one such result is the so called Poisson-Boltzmann equation, further discussed in section 2.4.

2.2.1 Boundary conditions

At the charged boundaries, most physical situations may be covered by either specifying the potential or the surface charge density. The former would be a boundary condition

of Dirichlet type:

$$\psi(\mathbf{x}) = \zeta(\mathbf{x}) , \quad \mathbf{x} \in \Gamma \quad (2)$$

and the latter a boundary condition of Neumann type:

$$\nabla\psi(\mathbf{x}) \cdot \mathbf{n} = -\frac{\sigma(\mathbf{x})}{\epsilon_0\epsilon_r} , \quad \mathbf{x} \in \Gamma \quad (3)$$

where Γ denotes the boundary of the domain and \mathbf{n} is the normal to the boundary surface. [?]

2.3 The transport of charges - Nernst-Planck equation

The charge concentration in an electrolyte is indeed affected by its environment. In the model proposed here, influences from: advection of the electrolyte, diffusion due to concentration gradients and effects from the electric field originating from charged objects placed at the border or in the flow is considered. Charge conservation without any external sources of the ion density, $c(\mathbf{x}, t)$, gives:

$$\frac{\partial c}{\partial t} + \nabla \cdot \mathbf{J} = 0 \quad (4)$$

where $\mathbf{J}(\mathbf{x}, t)$ is the net flux induced by the effects described above. Explicit expressions for the fluxes due to advection and diffusion respectively are

$$\mathbf{J}_{adv} = c\mathbf{u} \quad (5)$$

and

$$\mathbf{J}_{dif} = -D\nabla c \quad (6)$$

where \mathbf{u} is the advective velocity and D is a diffusion coefficient. The ionic flux due to the presence of an electric potential, $\psi(\mathbf{x}, t)$, is given by the Nernst equation [?]:

$$\mathbf{J}_{ele} = -\frac{zq_e D}{k_B T} c \nabla \psi \quad (7)$$

where z is the relative charge of the ion species, q_e is the fundamental charge, k_B is the Boltzmann constant and T is the temperature of the fluid.

Summing up the fluxes and putting them into eq. (4) gives

$$\frac{\partial c}{\partial t} = \nabla \cdot \left[D\nabla c - c\mathbf{u} + \frac{zq_e D}{k_B T} c \nabla \psi \right] \quad (8)$$

which is a known result often referred to as the Nernst-Planck equation. This is the equation for the transport of *one* species of ions, if several are present one NP equation for each species needs to be solved. The advective velocity, \mathbf{u} , and the potential gradient, $\nabla\psi$, are obtained from couplings to the Navier-Stokes and Poisson's equation respectively. More about the coupling between the equations is discussed in section 2.1.

2.3.1 Boundary conditions

Depending on the physical situation being modelled, different conditions may be imposed at the boundaries of the domain. Throughout this work, at hard boundaries (walls), the charge flux through the boundary is set to zero, i.e.:

$$\mathbf{J} \cdot \mathbf{n} = 0, \quad \mathbf{x} \in \Gamma \quad (9)$$

where \mathbf{n} denotes the normal to the surface and Γ is the boundary of the domain.

2.4 Poisson-Boltzmann equation

Consider a system consisting of an electrolyte in contact with a (flat) charged wall. Under certain assumptions, it is possible to explicitly determine the charge density in eq. (8) as a function of the electric potential. E.g. if there is no advection present and if the system has reached a steady state, i.e. $\partial c / \partial t = 0$ and $\mathbf{u} = \mathbf{0}$ we have:

$$D \nabla c + \frac{z q_e D}{k_B T} c \nabla \psi = \mathbf{J}_0 \quad (10)$$

where \mathbf{J}_0 is a constant flux. Due to the steady state assumption, what the equation above actually says is that the net flux of charge in the system is constant. Since no charges are wanted to flow through the wall boundary, the flux is set to zero on the wall and since the flux is constant it will therefore be zero everywhere in the liquid, i.e. $\mathbf{J}_0 = \mathbf{0}$.

Considering only a one-dimensional situation with a position variable y varying in a direction out from the wall into the liquid, eq. (10) reads

$$\frac{1}{c} \frac{dc}{dy} + \frac{z q_e}{k_B T} \frac{d\psi}{dy} = 0. \quad (11)$$

The charge density is determined by solving eq. (11) for c , i.e. integrating the equation. In order to avoid introducing additional unknown quantities, the equation is integrated to far away from the wall where the potential from the EDL is assumed to have decreased to zero and where the concentrations, c^∞ , of the electrolyte is known.

$$\int_y^\infty d \ln(c(y')) = -\frac{z q_e}{k_B T} \int_y^\infty d\psi(y') \quad (12)$$

This gives an expression for $C(y)$:

$$c(y) = c^\infty \exp \left(-\frac{z q_e \psi(y)}{k_B T} \right). \quad (13)$$

In a general case, there may be several species of ions in the electrolyte, the net charge density, ρ_e , is then given by simply summing up the contributions from the different species:

$$\rho_e = q_e \sum_i z_i c_i. \quad (14)$$

Summarising eqs. (1), (13) and (14) gives the Poisson-Boltzmann equation in one dimension

$$\frac{d^2\psi(y)}{dy^2} = -\frac{q_e}{\epsilon_r\epsilon_0} \sum_i z_i c_i^\infty \exp\left(-\frac{z_i q_e \psi(y)}{k_B T}\right). \quad (15)$$

2.4.1 The Debye–Hückel approximation

Historically, the non-linear nature of eq. (15) complicated for those wanting to solve it. This was a major difficulty in the past when the computational power at hands were rather limited. A linearisation is therefore sometimes done, this linear version of the PB equation is often referred to as the Debye–Hückel approximation. The solution of the linearisation gives, something to compare with and is usually used when defining a characteristic length scale of the EDL.

For a 1:1 electrolyte solution with an equal amount of positive and negatively charged ions, eq. (15) reduces to

$$\frac{d^2\psi(x)}{dx^2} = \frac{2c^\infty q_e z}{\epsilon_r\epsilon_0} \sinh\left(\frac{z q_e \psi(x)}{k_B T}\right). \quad (16)$$

and the linearised equation is

$$\frac{d^2\psi(x)}{dx^2} = \frac{2c^\infty q_e^2 z^2}{\epsilon_r\epsilon_0 k_B T} \psi(x) = \kappa^2 \psi(x) \quad (17)$$

where κ^{-1} is the Debye length which is where the exponential solution has decayed to e^{-1} of the boundary value. This quantity gives therefore a measure for the characteristic thickness of the EDL.

2.5 The velocity field - Navier-Stokes equations

The Navier-Stokes equations are among the most fundamental corner stones of hydrodynamics. They describe the motion of a fluid under the influence of various internal and external forces.

For later convenience and for reference when it comes to deriving the Lattice-Boltzmann formulation of the NS equation, a brief sketch of a derivation will here be presented. A most general form of the Navier-Stokes equation follows from momentum conservation

$$\frac{\partial(\rho \mathbf{u})}{\partial t} + \nabla \cdot (\rho \mathbf{u} \otimes \mathbf{u}) + \mathbf{Q} = 0 \quad (18)$$

where, ρ is fluid density, \mathbf{u} is velocity, \otimes represents the outer product and \mathbf{Q} is a momentum source term (force per volume). Expanding the time derivative and the divergence terms respectively gives

$$\mathbf{u} \left(\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{u}) \right) + \rho \left(\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} \right) + \mathbf{Q} = 0. \quad (19)$$

To assure mass conservation (without sources) we have

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{u}) = 0 \quad (20)$$

and eq. (19) reduces to

$$\rho \left(\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} \right) + \mathbf{Q} = 0 \quad (21)$$

which together with eq. (20) is a general formulation of the Navier stokes equations.

The force term \mathbf{Q} , is determined by the physical properties of the fluid and from its environment. In this work, only incompressible ($\rho = \text{constant}$) Newtonian fluids will be studied. The force contribution to \mathbf{Q} involved in that case is limited to viscous forces, pressure gradients in the fluid and to external force fields. Putting this into eqs. (20) and (21) gives

$$\nabla \cdot \mathbf{u} = 0 \quad (22)$$

and

$$\rho \left(\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} \right) = -\nabla P + \mu \nabla^2 \mathbf{u} + \mathbf{F} \quad (23)$$

where P is the pressure, μ the kinematic viscosity and \mathbf{F} is the contributions from external forces.

2.5.1 Boundary conditions

At hard boundaries (walls), the boundary conditions to eqs. (22) and (23) are set on the velocity of either a Dirichlet or Neumann type. In most physical situations the Dirichlet condition is used which corresponds to that there is a friction between the fluid and the wall, usually full friction, i.e. when no relative movement between fluid and wall is present and the velocity at the wall boundary is set to zero, i.e.

$$\mathbf{u} = 0, \quad \mathbf{x} \in \Gamma \quad (24)$$

where Γ denotes the boundary. The Neumann type conditions are used for no-friction walls where the normal component of the derivative of the velocity is specified, usually to zero.

At wet boundaries, inlets and outlets, of the domain various boundary conditions may be set. For instance the pressure or the velocity could be fixed. In the case of a fixed pressure boundary, a flow direction must also be specified for completeness. [?]]

2.6 Pressure-driven electrokinetic flow

As a charged fluid is driven by a pressure gradient, a movement of charges, i.e. an electrical current will be induced. Due to the charge flux, a potential gradient will build up along the flow direction. This potential is usually referred to as the streaming potential, $\phi(\mathbf{x})$, and its magnitude is determined from the induced current through Ohm's law

$$\mathbf{J} = -\sigma \nabla \phi \quad (25)$$

where σ is the conductivity of the fluid. In a perfectly conducting fluid there will be no potential differences. Also a complete neutral solution will carry no net current and also in this case there will be no potential differences.

Charges under the influence of an electric field will be affected by a force. Charges moving due to this force will, in a liquid, also pull liquid (uncharged) molecules with them. In a macroscopic limit, the force density affecting the charges in the liquid is assumed to affect the liquid as a whole. The volumetric force affecting the fluid from the presence of the streaming potential is then given by:

$$\mathbf{F} = -\rho_e \nabla \phi \quad (26)$$

where ρ_e is the charge density. This is an example of how the charge density from the Nernst-Planck equation may couple to the force term in Navier-Stokes equations.

This force will always be affecting the fluid in a direction opposite to the net flux of charge, i.e. the force will slow the fluid down, this is illustrated in fig. 2. This effect that a moving net charged fluid is slowed down is called the *electroviscous effect*. The name originates from that a similar effect might be achieved by increasing the viscosity of the fluid.

2.7 Electroosmotic flow

Instead of driving the fluid flow through a pressure drop, a net charged fluid may be driven by an external electric field. This may be seen as the opposite case to that in section 2.6 where a current is induced by a pressure drop.

The volumetric force on the fluid from the external field, \mathbf{E}_{ext} , is given by

$$\mathbf{F} = \rho_e \mathbf{E}_{ext} \quad (27)$$

where ρ_e is the charge density. If the electric field is constant (or at least has the same direction) everywhere, the sign of the force is not in the same direction for a net charged positive area of the fluid as for a net charged negative. Thus the fluid may be either slowed down or sped up. This is a qualitative difference to pressure driven situation and is illustrated in fig. 3.

The electroviscous effect is in the case of pure electroosmotic flow usually neglected as the field due to the streaming potential is, in most physical cases, small in comparison to the applied external field. [?]

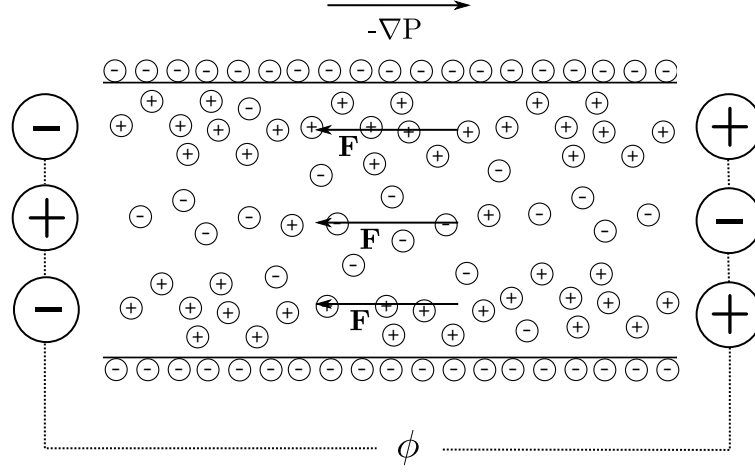


Figure 2: Example of an electroviscous system. The fluid is driven by a pressure gradient, ∇P . The directions of the forces on the fluid are always opposite to the flow direction. The force originates from the potential difference, ϕ , that builds up along the channel. The force is always opposite to the flow direction, thus slowing the flow down.

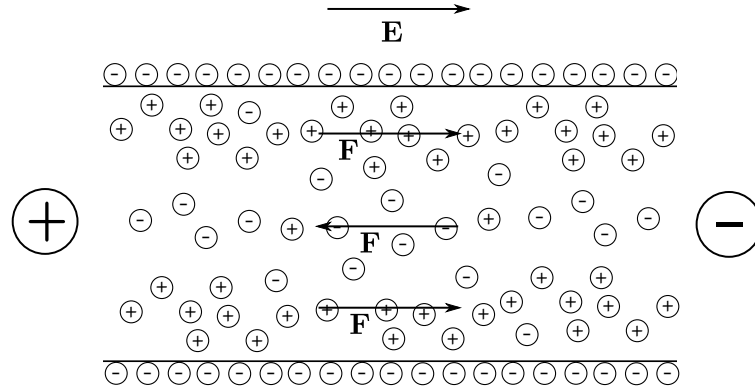


Figure 3: Example of an electroosmotic system. The fluid is driven by an external electric field, \mathbf{E} . The directions of the forces on the fluid from the electric field are indicated with arrows. Note however that the fluid does not necessarily has to flow in the direction of the force, this due to viscous effects in the fluid.

The lattice-Boltzmann method Rather than modelling on a macroscopic or microscopic scale, the lattice-Boltzmann method (LBM) operates at a scale in between those, often referred to as a mesoscopic scale. Nowadays, the method is most frequently used in modelling of fluid dynamics, i.e. computing solutions of the macroscopic Navier-Stokes equations. However, the lattice-Boltzmann method is not limited to this case and may be used to model other macroscopic systems as well. In this chapter a LBM approach will, in addition to the Navier-Stokes equations, also be formulated for the Nernst-Planck and Poisson's equation.

3 Historical overview

With the introduction of electronic calculating machines came also completely new possibilities of tackling difficult problems. New fields of computational science was born and methods for solving both new and traditional problems were developed.

The idea of using a discrete and simplified version of the Boltzmann-equation dates back to the mid 60's [?] with an experimental attempt to model simple gas dynamics. However, at the time, this kind of statistical computational approaches was not considered a serious alternative for the modelling of more sophisticated and complex systems such as fluid behaviour. It was first in the mid 80's when Frisch, Hasslacher and Pomeau showed that a lattice automaton that conserved mass and momentum in the collisions and with a lattice of certain symmetry, reproduced the Navier-Stokes equations in a macroscopic limit. It was by their work and the always increasing computational power that made the idea of fluid modelling on a mesoscopic scale a serious research topic. [?]

The lattice gas automata (LGA) approach was not perfect and suffered from some notable flaws, e.g. that the boolean nature of the method introduced statistical noise and that lack of symmetry in the lattices used made the advection non-isotropic. The statistical noise was usually dealt with by averaging which resulted in a coarsened domain and the advection issue was handled by introducing lattices of higher symmetry. Another consequence of the boolean variables is that only one particle per state was allowed which resulted in an equilibrium state from Fermi-Dirac statistics rather than the desired Maxwell-Boltzmann statistics.

As the flaws of the LGA approach was resolved one by another, the method evolved into what we today know as the lattice-Boltzmann method, with the crucial refinement of using continuous distributions over boolean variables. [?]

Today, the lattice-Boltzmann method is in many situations indeed a competitor to more traditional CFD methods. For example with advantages when it comes to parallelisation or implementing boundary conditions in complex geometries. One major downside with the LBM is the lack of theoretical work done and lack of literature compared to the case with more traditional methods such as finite element/volume methods. [?]

4 Statistical background

Consider one litre of air. At STP, the volume will contain in the order of 10^{22} molecules. In order to model this system microscopically, 6 variables per molecule will be needed to describe the microstate of the system. Just to store the state of the system in a computer would require more space than the estimated size of the whole world wide web times one million [?]. Thus, for these kind of systems, the microscopic approach is somewhat impractical.

Statistical approaches have been developed for these types of problems. A fundamental quantity used for describing the system is a continuous probability density distribution, f . This distribution may be regarded as an average over the microstates. Consider a volume of $d^3\mathbf{x}d^3\mathbf{p}$ in phase space, the number of molecules, dN in this volume is then given through the density distribution, f as

$$dN(\mathbf{x}, \mathbf{p}, t) = f(\mathbf{x}, \mathbf{p}, t)d^3\mathbf{x}d^3\mathbf{p}. \quad (28)$$

Thus $f(\mathbf{x}, \mathbf{p}, t)$ is a measure of the number of particles at location \mathbf{x} with momentum \mathbf{p} and at time t . Macroscopic variables are obtained by summing, e.g. the particle density, n , is obtained from

$$n(\mathbf{x}, t) = \int f(\mathbf{x}, \mathbf{p}, t)d^3\mathbf{p} \quad (29)$$

and the macroscopic momentum density of the system is determined by

$$\rho(\mathbf{x}, t)\mathbf{u}(\mathbf{x}, t) = \int \mathbf{p}f(\mathbf{x}, \mathbf{p}, t)d^3\mathbf{p} \quad (30)$$

where $\rho = mn$ is the mass density. Multiplying f by a power k of \mathbf{p} or $\mathbf{v} = \mathbf{p}/m$ and integrating is often referred to as taking the k :th moment of f and is a term that will be used throughout this chapter.

In the late 19th century, Boltzmann developed a model for the time evolution of f . To do this he had to make several assumptions. First, only collisions between two particles are considered, this makes the equation mostly applicable to dilute gases. Second, the two particles colliding are assumed to be uncorrelated before the collision. Third, external forces are assumed not to affect the collisions [?]. The equation is named after its father to the Boltzmann (transport) equation and reads:

$$\partial_t f + \frac{\mathbf{p}}{m} \cdot \nabla_{\mathbf{x}} f + \frac{\mathbf{F}}{m} \cdot \nabla_{\mathbf{v}} f = Q(f, f) \quad (31)$$

where f is the distribution function for a single species collection of particles of mass m , \mathbf{F} is external forces, \mathbf{p} is momentum, $\nabla_{\mathbf{x}}$ and $\nabla_{\mathbf{v}}$ are the gradients in location and velocity space respectively. The right-hand side contains the so called collision term which in the general case is expressed as an integral. This integral states how the distribution function changes after a two particle collision. However, the structure of this integral is in most physical situations too complicated to be used directly. Therefore, a number of simplifications have been proposed during the years.

When designing these approximations, at least two main properties of the collision integral must be kept. [?]

1. The same quantities that are conserved under collisions in the collision integral must also be conserved in the approximation.
2. Boltzmann's H-theorem must be fulfilled for the approximated collision operator.

Without being too specific, the H-theorem states that the entropy computed from f is always increasing with time and that the maximum entropy is obtained for a so called Maxwellian distribution in momentum/velocity space. Boltzmann used an other quantity denoted by H closely related to entropy, thus the name of the theorem. The Maxwellian distribution in two dimensions that f tends towards is given by

$$f^{(M)}(\mathbf{x}, \mathbf{v}, t) = n \left(\frac{m}{2\pi k_B T} \right) \exp \left(-\frac{m}{2k_B T} (\mathbf{v} - \mathbf{u})^2 \right) \quad (32)$$

where $\mathbf{u}(\mathbf{x}, t)$ is the mean velocity of the particles in the system and $n(\mathbf{x}, t)$ is the number of particles at location \mathbf{x} . In section 6, one of the most widely used approximations of the collision integral will be presented.

5 Basic idea of the LBM

As previously noted, the lattice-Boltzmann method is a mesoscopic method. This means that the modelling is neither done on a microscopic (molecular) level nor by direct solving of the macroscopic equations involved. The aim, in most situations with the lattice-Boltzmann method is indeed to solve some macroscopic equation but not direct. Instead a statistical model is used with various mesoscopic variables that, in some limit, reproduces the macroscopic variables. It is also possible to ensure that these variables (to some extent) fulfil a certain macroscopic equation by using a certain scheme.

Basically the lattice-Boltzmann method solves a discretised version of eq. (31) for the distribution functions from which the macroscopic quantities may be determined. Both the spatial positions and the velocity space is discretised allowing the distributions to "sit" only at certain positions and to stream to neighbouring locations only in certain directions. A naive way to visualise the evolution of f is to consider the distribution functions at the lattice nodes as pseudo particles that move along the lattice and collide.

Usually in two dimensions the velocity space is discretised into 9 distinct velocities, more about the choice of lattice is discussed in section 7. In this case 9 distribution functions are needed per node which might correspond to one or two macroscopic variables but is indeed fewer than the number of variables needed for a microscopic approach.

The discretised Boltzmann equation is referred to as the lattice-Boltzmann equation (LBE) and is one of the fundamental corner stones in the lattice-Boltzmann method, it reads:

$$f_i(\mathbf{x} + \mathbf{c}_i \delta_t, t + \delta_t) - f_i(\mathbf{x}, t) = \Omega_{ij}(\mathbf{x}, t) \quad (33)$$

where f_i denotes the distribution function for direction \mathbf{c}_i , δ_t is the time step and Ω_{ij} is the (for now non-specified) collision operator. An implicit sum over the second velocity index j is assumed. Various forms of collision operators exist and will be further discussed in section 6.

5.1 Computational algorithm

In order to solve eq. (33) the distribution functions must be set to some initial value. The choice of initial value is in most cases crucial with respect to stability and accuracy of the method. More about the initialisation will be discussed in later sections of this chapter.

When a proper initialisation has been performed, the time evolution of the distribution functions is determined iteratively by the explicit scheme in the LBE, eq. (33). The update in each time step is usually divided into two computational tasks. First, the new value that later will be propagated to a neighbouring node is computed, i.e.

$$f_i^*(\mathbf{x}, t + \delta_t) = f_i(\mathbf{x}, t) + \Omega_{ij}(\mathbf{x}, t) \quad (34)$$

This step will be referred to as the collision step since it is here the “collision” is computed. The second step consists of propagating the distribution functions to the neighbouring node in its corresponding direction, i.e.

$$f_i(\mathbf{x} + \mathbf{c}_i \delta_t, t + \delta_t) = f_i^*(\mathbf{x}, t + \delta_t) \quad (35)$$

This step will be referred to as the streaming step.

In the case of a finite domain, certain rules (boundary conditions) must be specified at the boundaries. Typically the distribution functions that are going to be streamed out of the domain is used to define the unknown ones that will “enter” the domain. More about boundary conditions in section 13. Thus, at each time step, the boundary conditions must also be handled.

This is broadly the whole computational algorithm behind the LBM, in fig. 4, a flow scheme of the algorithm is shown.

6 The BGK collision operator

The collision term in the LBE is the main ingredient in what determines the physics of the system that is being modelled. Here the desired interaction of the pseudo particles is stated. In section 4, two necessary properties to approximations of the full collision integral was stated.

One of the simplest collision operators that fulfil conditions (1) and (2) in section 4 is the BGK operator (BGK from its creators: Bhatnagar, Gross and Krook). It was proposed in 1954 and is today one of the most commonly used collision operators both in the case of the lattice-Boltzmann and the continuous Boltzmann equation. It is based on the principle of relaxing f towards a Maxwellian distribution. The relaxation is also

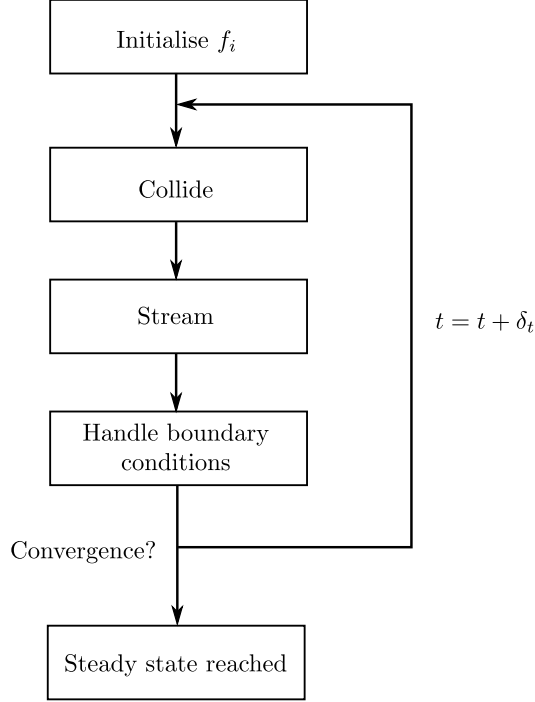


Figure 4: Flowchart of the most fundamental parts in an implementation of the LBM. The convergence is usually tested for a macroscopic variable.

performed in such a way that the collision invariants are preserved. In the discrete case, eq. (33), the operator is given by:

$$\Omega_{ij} = \Omega_i = -\omega \left[f_i(\mathbf{x}, t) - f_i^{(eq)}(\mathbf{x}, t) \right] \quad (36)$$

where ω is a parameter determining the relaxation rate and $f_i^{(eq)}$ should be an equilibrium distribution that makes sure that the necessary conditions are fulfilled. In the discrete case, a truncated expansion of eq. (32) is typically used [?]. This gives for instance

$$f_i^{(eq)} = w_i \rho \left[1 + \frac{\mathbf{c}_i \cdot \mathbf{u}}{c_s^2} + \frac{(\mathbf{c}_i \cdot \mathbf{u})^2}{2c_s^4} - \frac{\mathbf{u}^2}{2c_s^2} \right] \quad (37)$$

where w_i is a lattice specific weight, ρ is the zeroth moment of f_i and \mathbf{c}_i is a unit velocity in the discretised velocity space.

The BGK operator is due to its simplicity both when it comes to theoretical treatment and implementation a popular choice. However in some physical situations, e.g. multi-phase or high Reynolds-number flows, more sophisticated alternatives are required [?]. Throughout this work, the BGK operator will be used.

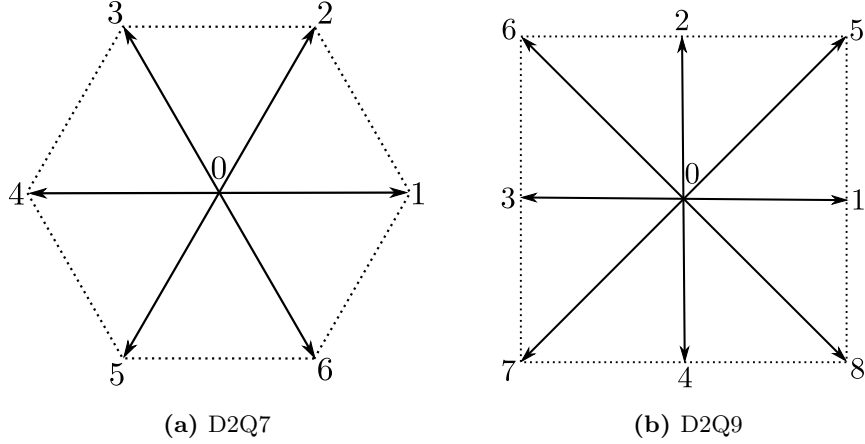


Figure 5: Two different unit cells for lattices used in the LBM in two dimensions. In (a) the D2Q7 seven speed lattice is shown and in (b) the nine speed D2Q9 lattice. The numbering at the edges is the usual naming convention for the different velocities.

7 The lattice

The discrete spatial coordinates together with the discrete velocity coordinates forms a lattice. Spatial coordinates are neighbourwise connect through the discrete velocities. The discretisation in the LBM must be performed in such a way that the lattice may be produced from translating a single unit cell. This gives that the spatial resolution is constant for the whole domain. In other approaches, e.g. finite element methods the mesh may locally be refined at interesting regions of the domain which is a strength to these methods over the LBM.

There exist a convention for naming different lattices. A lattice of dimension d and with q distinct velocities is denoted by $DdQq$. For example a lattice with 9 velocities in dimension 2 is denoted D2Q9. An example of two different lattices is presented in fig. 5. The D2Q9 lattice is also the one that will be used in this work, the velocities \mathbf{c}_i are given by

$$\{\mathbf{c}_i\} = \left\{ \begin{bmatrix} 0 \\ 0 \end{bmatrix}, \begin{bmatrix} 1 \\ 0 \end{bmatrix}, \begin{bmatrix} 0 \\ 1 \end{bmatrix}, \begin{bmatrix} -1 \\ 0 \end{bmatrix}, \begin{bmatrix} 0 \\ -1 \end{bmatrix}, \begin{bmatrix} 1 \\ 1 \end{bmatrix}, \begin{bmatrix} -1 \\ 1 \end{bmatrix}, \begin{bmatrix} -1 \\ -1 \end{bmatrix}, \begin{bmatrix} 1 \\ -1 \end{bmatrix} \right\} \quad (38)$$

To be able to retrieve the desired equations in the macroscopic limit of the LBE, the lattice used must possess a certain degree of isotropy. Thus the choice of lattice is not arbitrary. For instance, for obtaining the Navier-Stokes equations, the lattice velocities must at least posses the following properties:

$$\sum_i w_i = 1 \quad (39a)$$

$$\sum_i w_i c_{i\alpha} = 0 \quad (39b)$$

$$\sum_i w_i c_{i\alpha} c_{i\beta} = c_s^2 \delta_{\alpha\beta} \quad (39c)$$

$$\sum_i w_i c_{i\alpha} c_{i\beta} c_{i\gamma} = 0 \quad (39d)$$

$$\sum_i w_i c_{i\alpha} c_{i\beta} c_{i\gamma} c_{i\delta} = c_s^4 (\delta_{\alpha\beta} \delta_{\gamma\delta} + \delta_{\alpha\gamma} \delta_{\beta\delta} + \delta_{\alpha\delta} \delta_{\beta\gamma}) \quad (39e)$$

where the sums are over the discrete velocities \mathbf{c}_i , w_i are lattice specific weights c_s is the speed of sound for the lattice and δ_{ij} is a Kroenecker delta.

The weights are introduced to compensate for the fact that different velocity vectors are of different length. See for example the D2Q9 lattice in fig. 5b where three lengths are present. In this case, three different weights will be needed. From the relations in eqs. (39) follow that these weights are:

$$w_i = \begin{cases} 4/9 & \text{if } i = 0 \\ 1/9 & \text{if } i = 1, 2, 3, 4 \\ 1/36 & \text{if } i = 5, 6, 7, 8 \end{cases} \quad (40)$$

The quantity c_s which often is referred to as the speed of sound may be thought of as an effective propagation velocity of the lattice and is determined, for the D2Q9 lattice, to

$$c_s = c/\sqrt{3} \quad (41)$$

where $c = |\mathbf{c}_{1,2,3,4}| = \delta_x/\delta_t$.

8 Asymptotic analysis

Methods from asymptotic analysis will, in this section, be used to investigate the macroscopic limit of the general LBE. More detailed and specific analyses for the three different equations considered will be presented in sections 9.1, 10.1 and 11.1 respectively.

Asymptotic analysis is basically about describing mathematical objects in some limit, e.g. how a function behaves for large or small values of some variable or parameter. Consider for example the series S_ϵ :

$$S_\epsilon = a^{(0)} + a^{(1)}\epsilon + a^{(2)}\epsilon^2 + a^{(3)}\epsilon^3 + \mathcal{O}(\epsilon^4) \quad (42)$$

It is clear that for sufficiently small values of ϵ , the terms of higher order is of negligible magnitude to those of lower order and the series may be truncated at some point and

still be a good approximation of S_ϵ . For example if ϵ is small, then $S_\epsilon \approx a^{(0)}$ and we say that if $a^{(1)} \neq 0$ that this approximation is of first order accuracy.

There are different approaches to go from the discrete LBE to a continuous macroscopic equation. The most frequently applied one to obtain the Navier-Stokes equations is the Chapman-Enskog method [?], which will reproduce the compressible equations. Another method, often employed by M. Junk and his associates, e.g. in [?], is a method based on regular asymptotic expansions, this is also the method that will be utilised in this work and will in the case of Navier-Stokes reproduce the incompressible equations. A brief discussion of the differences between the Chapman-Enskog and the regular expansion approaches will be carried out at the end of this chapter.

The basic idea behind the analysis is to expand the distribution function f_i in some small parameter, ϵ . Also this parameter will be related to the spatial and time scales. The macroscopic limit is obtained by taking the Taylor expansion of the discrete LBE and comparing terms of equal order in ϵ . Together with the fact that certain quantities are invariant under collisions, macroscopic differential equations are obtained. Now follows the part of the analysis which is common for the three equations, the more equation specific analysis is carried out in sections 9.1, 10.1 and 11.1 respectively.

8.1 Motivation of the choice of expansion parameter

A most desired property of the expansion parameter is that it should be a small and dimensionless number. If the lattice is dense enough with respect to the characteristic length scale of the system, a suitable choice is the Knudsen number, ϵ , which is defined as the ratio of the mean free path, δ_x , and the characteristic length of the system under consideration, ℓ_0 , i.e. $\epsilon = \delta_x/\ell_0$. To be able to perform the asymptotic analysis we must also relate the time scale to this parameter. From the fact that the lattice speed $c = \delta_x/\delta_t$ and by introducing a characteristic speed, $u_o = \ell_0/t_0$, we have

$$\epsilon = \frac{\delta_x}{\ell_0} = \frac{c}{u_o} \frac{\delta_t}{t_0} \quad (43)$$

It is now clear that what determines the relation between the timescale and the parameter ϵ is the ratio of the characteristic speed and the lattice speed which is usually referred to as the Mach number, Ma . In our particular case we will operate in the incompressible limit, i.e. $\text{Ma} \ll 1$ and a suitable choice is a small number, thus $\text{Ma} = \epsilon$ is chosen [?]. The discretisation of the space and time step is then related through

$$\delta_x'^2 = \delta_t' = \epsilon^2 \quad (44)$$

where the primes denote dimensionless variables. This particular scaling is usually referred to as diffusive scaling.

8.2 Expanding the LBE

The LBE, eq. (33), with dimensionless variables and the BGK collision operator reads:

$$f_i(\mathbf{x}' + \epsilon \mathbf{c}'_i, t' + \epsilon^2) - f_i(\mathbf{x}', t') = -\omega \left[f_i(\mathbf{x}', t') - f_i^{(eq)}(\mathbf{x}', t') \right]. \quad (45)$$

The primes denoting dimensionless variables will, for readability reasons, from hereon be dropped. If nothing else is stated we always consider dimensionless variables.

To obtain a differential equation, the difference equation in eq. (45) is Taylor expanded, which gives

$$\epsilon(\mathbf{c}_i \cdot \nabla f_i) + \epsilon^2(\partial_t f_i + (\mathbf{c}_i \cdot \nabla f_i)^2/2) + \epsilon^3(\partial_t(\mathbf{c}_i \cdot \nabla f_i) + (\mathbf{c}_i \cdot \nabla f_i)^3/6) + \mathcal{O}(\epsilon^4) = -\omega \left[f_i - f_i^{(eq)} \right] \quad (46)$$

Expanding also f_i and $f_i^{(eq)}$ in the parameter ϵ :

$$f_i = f_i^{(0)} + \epsilon f_i^{(1)} + \epsilon^2 f_i^{(2)} + \epsilon^3 f_i^{(3)} + \mathcal{O}(\epsilon^4) \quad (47)$$

$$f_i^{(eq)} = f_i^{(eq,0)} + \epsilon f_i^{(eq,1)} + \epsilon^2 f_i^{(eq,2)} + \epsilon^3 f_i^{(eq,3)} + \mathcal{O}(\epsilon^4) \quad (48)$$

and inserting these expressions into eq. (46) gives an equation with terms of varying orders of ϵ . Separating this equation in equations of common orders allows for an analysis of what happens at different scales of ϵ . For the four leading orders in ϵ we have:

$$\epsilon^0 : 0 = -\omega \left[f_i^{(0)} - f_i^{(eq,0)} \right], \quad (49)$$

$$\epsilon^1 : \mathbf{c}_i \cdot \nabla f_i^{(0)} = -\omega \left[f_i^{(1)} - f_i^{(eq,1)} \right], \quad (50)$$

$$\epsilon^2 : \mathbf{c}_i \cdot \nabla f_i^{(1)} + \partial_t f_i^{(0)} + (\mathbf{c}_i \cdot \nabla f_i^{(0)})^2/2 = -\omega \left[f_i^{(2)} - f_i^{(eq,2)} \right] \quad (51)$$

and

$$\epsilon^3 : \mathbf{c}_i \cdot \nabla f_i^{(2)} + \partial_t f_i^{(1)} + (\mathbf{c}_i \cdot \nabla f_i^{(1)})^2/2 + \partial_t(\mathbf{c}_i \cdot \nabla f_i^{(0)}) + (\mathbf{c}_i \cdot \nabla f_i^{(0)})^3/6 = -\omega \left[f_i^{(3)} - f_i^{(eq,3)} \right]. \quad (52)$$

The idea is now that for an equation of a particular order in ϵ , use collision invariants and eliminate unknown $f_i^{(n)}$ by using equations of lower order in ϵ . This will in the end, result in differential equations for macroscopic variables, given by moments of the f_i 's.

9 LBM for the Nernst-Planck equation

The method presented here is based on representing the Nernst-Planck equation, eq. (8), as an equation of advection-diffusion type. Considering the quantity:

$$\bar{\mathbf{u}} = \mathbf{u} - \frac{z q_e D}{k_B T} \nabla \psi \quad (53)$$

as an effective advective velocity, we have:

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\bar{\mathbf{u}}\rho - D\nabla\rho) = 0 \quad (54)$$

which is a mass conservation equation with fluxes from diffusion and from advection respectively. The letter c for denoting the charge concentration has in this section been replaced by the letter ρ to avoid the risk of confusing it with the lattice velocities which traditionally are denoted by \mathbf{c}_i .

A collision operator of BGK type, eq. (36) will be used together with a D2Q9 lattice. The lattice-Boltzmann equation then reads:

$$f_i(\mathbf{x} + \mathbf{c}_i\delta_t, t + \delta_t) - f_i(\mathbf{x}, t) = -\omega \left[f_i(\mathbf{x}, t) - f_i^{(eq)}(\mathbf{x}, t) \right] \quad (55)$$

with $\{\mathbf{c}_i\}_{i=0}^{Q-1}$ for the D2Q9 lattice as in eq. (38). The equilibrium function, $f_i^{(eq)}$, is chosen as [?]:

$$f_i^{(eq)} = w_i \rho \left(1 + \frac{\mathbf{c}_i \cdot \bar{\mathbf{u}}}{c_s^2} \right) \quad (56)$$

with the weights, w_i , as in eq. (40). The charge density and charge flux density is obtained by taking the zeroth and first moments of the distribution function respectively, i.e:

$$\rho = \sum_i f_i \quad (57)$$

and

$$\mathbf{j} = \sum_i f_i \mathbf{c}_i \quad (58)$$

The diffusion constant, D , is related to the relaxation parameter ω through

$$D = c_s^2 \left(\frac{1}{\omega} - \frac{1}{2} \right). \quad (59)$$

9.1 Asymptotic analysis

To motivate the appearance of the above suggested method for solving eq. (54) and for showing under what premises the method is valid, the macroscopic limit of the discrete scheme will now be analysed in an asymptotic manner. Note that for the advection diffusion equation mass is but flux is not conserved.

From the expansion of f_i in eq. (47) and from eqs. (74) and (75) follow the expansions of the mass density and flux respectively as

$$\rho = \rho^{(0)} + \epsilon \rho^{(1)} + \epsilon^2 \rho^{(2)} + \epsilon^3 \rho^{(3)} + \mathcal{O}(\epsilon^4) \quad (60)$$

and

$$\mathbf{j} = \mathbf{j}^{(0)} + \epsilon \mathbf{j}^{(1)} + \epsilon^2 \mathbf{j}^{(2)} + \epsilon^3 \mathbf{j}^{(3)} + \mathcal{O}(\epsilon^4) \quad (61)$$

The advective velocity is also expanded as:

$$\bar{\mathbf{u}} = \bar{\mathbf{u}}^{(0)} + \epsilon \bar{\mathbf{u}}^{(1)} + \epsilon^2 \bar{\mathbf{u}}^{(2)} + \epsilon^3 \bar{\mathbf{u}}^{(3)} + \mathcal{O}(\epsilon^4) \quad (62)$$

By plugging these expansion into the equilibrium distribution eq. (56), the expansion in eq. (48) is obtained. The terms of order zero is used in the zeroth order equation of the LBE, eq. (49), which gives

$$f_i^{(0)} = w_i \rho^{(0)} \left(1 + \frac{\mathbf{c}_i \cdot \bar{\mathbf{u}}^{(0)}}{c_s^2} \right). \quad (63)$$

However, since we are only considering advection in the low Mach limit, i.e. $|\bar{\mathbf{u}}| \sim \epsilon$, we will in this analysis assume that $\bar{\mathbf{u}}^{(0)} = 0$. $\bar{\mathbf{u}}$ will then be of order ϵ to leading order. It is possible to show [?] that this assumption holds if $\bar{\mathbf{u}}$ is initialised properly, i.e. small and if no major momentum sources are present. Thus the expression for $f_i^{(0)}$ reduces to

$$f_i^{(0)} = w_i \rho^{(0)}. \quad (64)$$

We now continue to the equation of order one in ϵ , eq. (50). Taking the zeroth moment gives the equation $0 = 0$ which indeed is true but not very useful. Note that the right hand side vanishes due to mass conservation. $f_i^{(1)}$ will be needed in the next step and is, by using eq. (64):

$$f_i^{(1)} = -\frac{1}{\omega} (\mathbf{c}_i \cdot \nabla) (w_i \rho^{(0)}) + w_i \left(\rho^{(1)} + \rho^{(0)} \frac{\mathbf{c}_i \cdot \bar{\mathbf{u}}^{(1)}}{c_s^2} \right). \quad (65)$$

Taking the first moment of $f_i^{(1)}$ gives the leading order in the flux ($\mathbf{j}^{(0)} = 0$ since $\bar{\mathbf{u}}^{(0)} = 0$):

$$\mathbf{j}^{(1)} = \rho^{(0)} \bar{\mathbf{u}}^{(1)} - c_s^2 / \omega \nabla \rho^{(0)} \quad (66)$$

Continuing to the equation of order two in ϵ , eq. (51) and taking the zeroth moment of the equation gives

$$\nabla \cdot \mathbf{j}^{(1)} + \partial_t \rho^{(0)} + c_s^2 / 2 \nabla^2 \rho^{(0)} = 0 \quad (67)$$

and by inserting the expression for $\mathbf{j}^{(1)}$ we end up with

$$\partial_t \rho^{(0)} + \nabla \cdot \left[\rho^{(0)} \bar{\mathbf{u}}^{(1)} - c_s^2 \left(\frac{1}{\omega} - \frac{1}{2} \right) \nabla \rho^{(0)} \right] = 0 \quad (68)$$

which is an advection diffusion equation with a diffusion constant as in eq. (59). Since $\rho^{(0)}$ fulfils the equation of interest, the solution ρ that we get from the lattice-Boltzmann method is at least of first order accuracy. To determine the exact order of accuracy, higher order terms, $\rho^{(k>0)}$, must be determined. If also all those terms would be zero the method would be exact, unfortunately that is not the case. The analysis of higher order terms will not be performed here, but it is possible to show that $\rho^{(1)}$ is zero only under certain premises, i.e. for a proper initialisation [?]. $\rho^{(2)}$ is however in general non-zero and the obtained solution is thus second order accurate.

10 LBM for the incompressible Navier-Stokes

The most frequent use of the LBM is to solve the Navier-Stokes equations. In this work only the incompressible case, eqs. (22) and (23) will be considered. The LBM may however be used to reproduce weak compressibility [?]. The incorporation of forces will be treated in section 10.2. We recall the incompressible Navier-Stokes equations without external forces present from chapter ?? as:

$$\nabla \cdot \mathbf{u} = 0 \quad (69)$$

and

$$\rho \left(\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} \right) = -\nabla P + \rho \nu \nabla^2 \mathbf{u} \quad (70)$$

where ν is the kinematic viscosity related through the dynamic viscosity, μ , by

$$\mu = \rho \nu. \quad (71)$$

A LBM will now be formulated for eqs. (69) and (70). The LBE with a BGK collision operator is given by

$$f_i(\mathbf{x} + \mathbf{c}_i \delta_t, t + \delta_t) - f_i(\mathbf{x}, t) = -\omega \left[f_i(\mathbf{x}, t) - f_i^{(eq)}(\mathbf{x}, t) \right] \quad (72)$$

where

$$f_i^{(eq)} = w_i \rho \left[1 + \frac{\mathbf{c}_i \cdot \mathbf{u}}{c_s^2} + \frac{(\mathbf{c}_i \cdot \mathbf{u})^2}{2c_s^4} - \frac{\mathbf{u}^2}{2c_s^2} \right] \quad (73)$$

where w_i are the weights in eq. (40).

The density, ρ , and the mass flux, $\rho \mathbf{u}$, is determined from f_i by taking the zeroth and first moments respectively:

$$\rho = \sum_i f_i \quad (74)$$

and

$$\rho \mathbf{u} = \sum_i f_i \mathbf{c}_i. \quad (75)$$

The kinematic viscosity is related to the relaxation parameter, ω

$$\nu = c_s^2 \left(\frac{1}{\omega} - \frac{1}{2} \right). \quad (76)$$

10.1 Asymptotic analysis

Partially based on [?], an asymptotic analysis of the above suggested method will be performed. In most literature available, when reproducing the Navier-Stokes equations in the macroscopic limit, a Chapman-Enskog expansion is performed. Note that this is *not* what is done here.

From the expansion of f_i in eq. (47) follows the expansion of the macroscopic mass and velocity as

$$\rho = \rho^{(0)} + \epsilon \rho^{(1)} + \epsilon^2 \rho^{(2)} + \epsilon^3 \rho^{(3)} + \mathcal{O}(\epsilon^4) \quad (77)$$

and

$$\mathbf{u} = \mathbf{u}^{(0)} + \epsilon \mathbf{u}^{(1)} + \epsilon^2 \mathbf{u}^{(2)} + \epsilon^3 \mathbf{u}^{(3)} + \mathcal{O}(\epsilon^4). \quad (78)$$

These expansions are plugged into the equilibrium distribution in eq. (73) and from the equation of order zero in ϵ , eq. (49) gives:

$$f_i^{(0)} = w_i \rho^{(0)} \quad (79)$$

Here $\mathbf{u}^{(0)}$ has been assumed to be zero by the same argumentation as in the Nernst-Planck analysis, section 9.1. Continuing to the equation of order 1 in ϵ , eq. (50) and taking the first moment gives

$$\nabla \rho^{(0)} = 0. \quad (80)$$

Further, by using this fact that $\rho^{(0)}$ is constant in space and that

$$f_i^{(1)} = -\frac{1}{\omega} \mathbf{c}_i \cdot \nabla f_i^{(0)} + f_i^{(eq,1)} \quad (81)$$

where

$$f_i^{(eq,1)} = w_i \left[\rho^{(1)} + \rho^{(0)} \frac{\mathbf{c}_i \cdot \mathbf{u}^{(1)}}{c_s^2} \right] \quad (82)$$

gives when taking the zeroth moment of the equation of order two in ϵ , eq. (51)

$$\partial_t \rho^{(0)} + \nabla \cdot (\rho^{(0)} \mathbf{u}^{(1)}) = 0. \quad (83)$$

This equation states the conservation of mass for $\rho^{(0)}$. Since we are considering only systems in the incompressible limit, $\rho^{(0)}$ will be assumed to also be constant in time and we have

$$\nabla \cdot \mathbf{u}^{(1)} = 0. \quad (84)$$

Taking the first moment of eq. 51 gives

$$\nabla \rho^{(1)} = 0. \quad (85)$$

It can be showed [?] that if $\rho^{(1)}$ is initialised properly, then it does not change in time. Therefore, if $\rho^{(1)}$ is initialised to zero, it will also remain zero for all time.

As we now continue to the equation of order three in ϵ , things will get a bit more technical as we will make use of the fourth order lattice isotropy, eq. (39). The zeroth moment of the equation gives

$$\nabla \cdot \mathbf{u}^{(2)} = 0. \quad (86)$$

which may be used to show that $\mathbf{u}^{(2)} = 0$ [?], it will not be shown here. Instead we continue by taking the first moment of the equation, as it gets a bit more technical now, some intermediate steps in the calculation will explicitly be written out. First from the equation of order two in ϵ we have that

$$f_i^{(2)} = -\frac{1}{\omega} \mathbf{c}_i \cdot \nabla f_i^{(1)} - \frac{1}{\omega} \partial_t f_i^{(0)} - \frac{1}{2\omega} (\mathbf{c}_i \cdot \nabla f_i^{(0)})^2 + f_i^{(eq,2)} \quad (87)$$

where

$$f_i^{(eq,2)} = w_i \left[\rho^{(2)} + \rho^{(0)} \frac{(\mathbf{c}_i \cdot \mathbf{u}^{(1)})^2}{2c_s^4} - \rho^{(0)} \frac{(\mathbf{u}^{(1)})^2}{2c_s^2} \right] \quad (88)$$

where the fact that $\mathbf{u}^{(2)} = 0$ have been used. Inserting the expression for $f_i^{(2)}$ into eq. (52) gives, in index notation

$$\begin{aligned} c_{i\alpha} c_{i\beta} \partial_\beta \left[-\frac{\rho^{(0)}}{\omega c_s^2} c_{i\gamma} \partial_\gamma c_{i\delta} u_\delta^{(1)} + w_i \rho^{(2)} + \frac{\rho^{(0)} w_i}{2c_s^4} (c_{i\gamma} c_{i\delta} u_\gamma^{(1)} u_\delta^{(1)} - (u^{(1)})^2) \right] + \\ + \partial_t w_i \rho^{(0)} u_\alpha^{(1)} + \frac{\rho^{(0)}}{2c_s^2} c_{i\alpha} c_{i\beta} \partial_\beta c_{i\gamma} \partial_\gamma c_{i\delta} = 0 \end{aligned} \quad (89)$$

For brevity, the terms that in the end will equal zero have been omitted. The indices α, β, γ and δ may take the x or y component respectively. Summing eq. (89) over the directional index i and by using the isotropy properties of the lattice, eq. (39) gives:

$$\begin{aligned} c_s^2 \rho^{(0)} \left(\frac{1}{2} - \frac{1}{\omega} \right) \partial_\beta (\delta_{\alpha\beta} \delta_{\gamma\delta} + \delta_{\alpha\gamma} \delta_{\beta\delta} + \delta_{\alpha\delta} \delta_{\beta\gamma}) + c_s^2 \partial_\beta \delta_{\alpha\beta} \rho^{(2)} + \\ + \frac{\rho^{(0)}}{2} ((\delta_{\alpha\beta} \delta_{\gamma\delta} + \delta_{\alpha\gamma} \delta_{\beta\delta} + \delta_{\alpha\delta} \delta_{\beta\gamma}) u_\gamma^{(1)} u_\delta^{(1)} - \delta_{\alpha\beta} (u^{(1)})^2) + \rho^{(0)} \partial_t u_\alpha^{(1)} = 0 \end{aligned} \quad (90)$$

On the form it is written now, it is not totally clear whether the above equation is the incompressible Navier-Stokes momentum equation or not. A simplification is in order and for brevity only the simplification of one term is shown here.

By noting that $(u^{(1)})^2$ may be written as $\delta_{\gamma\delta} u_\gamma^{(1)} u_\delta^{(1)}$ we have

$$\begin{aligned}\partial_\beta \left[(\delta_{\alpha\beta}\delta_{\gamma\delta} + \delta_{\alpha\gamma}\delta_{\beta\delta} + \delta_{\alpha\delta}\delta_{\beta\gamma}) u_\gamma^{(1)} u_\delta^{(1)} - \delta_{\alpha\beta} (u^{(1)})^2 \right] = \\ = \partial_\beta \left[(\delta_{\alpha\beta}\delta_{\gamma\delta} + \delta_{\alpha\gamma}\delta_{\beta\delta}) u_\gamma^{(1)} u_\delta^{(1)} \right]\end{aligned}\quad (91)$$

and by summing over γ and δ the above expression reduces to:

$$\partial_\beta \left[u_\alpha^{(1)} u_\beta^{(1)} + u_\beta^{(1)} u_\alpha^{(1)} \right] = 2\partial_\beta u_\alpha^{(1)} u_\beta^{(1)} \quad (92)$$

and from incompressibility ($\partial_\beta u_\beta^{(1)} = 0$), eq. (84) we end up with

$$2u_\beta^{(1)} \partial_\beta u_\alpha^{(1)}. \quad (93)$$

By analogous simplification of the remaining terms in eq. (90) we get

$$\rho^{(0)} \left(\partial_t u_\alpha^{(1)} + u_\beta^{(1)} \partial_\beta u_\alpha^{(1)} \right) = -\partial_\beta (c_s^2 \rho^{(2)}) + \rho^{(0)} \nu \partial_\beta \left(\partial_\beta u_\alpha^{(1)} + \partial_\alpha u_\beta^{(1)} \right) \quad (94)$$

where ν is defined as in eq. (76), and if the quantity $c_s^2 \rho^{(2)}$ is interpreted as the pressure, the above equation is indeed the incompressible Navier-Stokes momentum equation, eq. (70). It is satisfied by $\mathbf{u}^{(1)}$ in the expansion of \mathbf{u} , since $\mathbf{u}^{(2)} = 0$, the error will be of order ϵ^3 and we say that the LB formulation is of second order accuracy in velocity. $\rho^{(0)}$ satisfies the mass equation and since $\rho^{(1)} = 0$ we say that the method is second order in density as well [?]. Since the pressure is included as a term in the density expansion, problems with keeping the incompressibility might arise if large enough pressures are present.

10.2 Forcing schemes

Several methods have been proposed to add the forcing term in eq. (23) to the LBE. In [?] five of these methods have been compared. To briefly summarise the article; for single phase flows, the methods achieve comparable.

Due to its simplicity, a method formulated by Shan and Chen [?] was used in this work. It consists of modifying the equilibrium distribution by computing it with the modified velocity

$$\mathbf{u}^{(eq)} = \frac{1}{\rho} \left(\sum_i f_i \mathbf{c}_i + \frac{\mathbf{F} \delta_t}{\omega} \right) \quad (95)$$

where \mathbf{F} is the external force. The physical velocity, \mathbf{u} , is computed by

$$\mathbf{u} = \frac{1}{\rho} \left(\sum_i f_i \mathbf{c}_i + \frac{\mathbf{F} \delta_t}{2} \right). \quad (96)$$

11 LBM for Poisson's equation

As Poisson's equation, eq. (1), is considered, a fundamental difference to the Nernst-Planck and Navier-Stokes equations is immediately noted. It is not a differential equation of parabolic but of elliptic type. If we consider what has been said about the LBM so far in this chapter, it seems that it is a method that deals with parabolic equations and not elliptic. However, by introducing a time derivative to the Poisson's equation and only considering the steady state solution the LBM may be used even for this equation. A diffusion-like equation with a source term is then obtained

$$\partial_t \rho + \nabla^2 \rho = R \quad (97)$$

where R is the right-hand side of the Poisson's equation.

A LBM for this equation will now be formulated. The LBE to be solved in this case is, an equation of the form:

$$f_i(\mathbf{x} + \mathbf{c}_i \delta_t, t + \delta_t) - f_i(\mathbf{x}, t) = -\omega \left[f_i(\mathbf{x}, t) - f_i^{(eq)}(\mathbf{x}, t) \right] + G_i(R) \quad (98)$$

where $G_i(R)$ is an addition to the BGK collision operator to account for the source term R in eq. (97). The equilibrium distribution is given by

$$f_i^{(eq)} = w_i \rho \quad (99)$$

where w_i are the weights in eq. (40).

The quantity ρ which in this work is interpreted to electric potential in the steady state is determined by

$$\rho = \sum_i f_i \quad (100)$$

and the quantity which, to us, is of even more interest, i.e. the electric field, is given by

$$\nabla \rho = -\frac{\omega}{c_s^2 \delta_x} \sum_i f_i \mathbf{c}_i. \quad (101)$$

Finally, the additional source term in the collision operator, $G_i(R)$, is given by

$$G_i(R) = w_i c_s^2 \left(\frac{1}{2} - \frac{1}{\omega} \right) R. \quad (102)$$

11.1 Asymptotic analysis

The given method for solving eq. (97) will now be motivated by performing an asymptotic analysis of the suggested LBE.

f_i is expanded as in eq. (47) which gives an expansion of ρ as in eq. (60). Further, the source term R is expanded:

$$R = R^{(0)} + \epsilon R^{(1)} + \epsilon^2 R^{(2)} + \epsilon^3 R^{(3)} + \mathcal{O}(\epsilon^4) \quad (103)$$

which also gives an expansion for the LBE source term as

$$G_i = G_i^{(0)} + \epsilon G_i^{(1)} + \epsilon^2 G_i^{(2)} + \epsilon^3 G_i^{(3)} + \mathcal{O}(\epsilon^4). \quad (104)$$

The equilibrium distribution is expanded by plugging in the expansion of ρ from eq. (60). This gives for the zeroth order equation in ϵ , eq. (49) that

$$f_i^{(0)} = w_i \rho^{(0)} + \frac{1}{\omega} G_i^{(0)} \quad (105)$$

However, taking the first moment of the equation gives $G_i^{(0)} = 0$ and the above expression for $f_i^{(0)}$ reduces to

$$f_i^{(0)} = w_i \rho^{(0)}. \quad (106)$$

Continuing to the equation of first order in ϵ , eq. (50) and taking the zeroth moment gives that also $G_i^{(1)} = 0$. Taking the first moment of the same equation gives

$$\nabla \rho^{(0)} = -\frac{\omega}{c_s^2} \sum_i f_i^{(1)} \mathbf{c}_i \approx -\frac{\omega}{c_s^2 \delta x} \sum_i f_i \mathbf{c}_i \quad (107)$$

Here, for the approximation, the expansion of f_i is used together with the fact that $\epsilon = \delta x$ in dimensionless variables.

The next equation in ϵ , i.e. the one of order two, will eventually give us what we are looking for. Taking the zeroth moment gives

$$-\frac{c_s^2}{\omega} \nabla^2 \rho^{(0)} + \partial_t \rho^{(0)} + \frac{c_s^2}{2} \nabla^2 \rho^{(0)} = \sum_i G_i^{(2)}(R^{(2)}) \quad (108)$$

From this equation, it is deduced that in order to obtain Poisson's equation in a steady state situation, i.e. when $\partial_t \rho^{(0)} = 0$ the right-hand side must fulfill the following condition

$$\sum_i G_i^{(2)}(R^{(2)}) = c_s^2 \left(\frac{1}{2} - \frac{1}{\omega} \right) R^{(2)} \quad (109)$$

One such $G_i^{(2)}(R)$ is the one described in eq. (102).

12 Algorithm/Scheme for solving the coupled equations

In this section, the scheme to solve the coupled Poisson's, Nernst-Planck and Navier-Stokes equations is described. The dependencies between the equations are shown in fig. 1.

Following an initialisation of the velocity field and charge density, an iteration in time is performed. At each time step, a potential and electric field is computed by solving Poisson's equation for the present charge density. This is followed by updating the charge density from the new potential and then the velocity field is updated with the presence of a force related to the charge density. The main ingredients in the iterative algorithm are shown in fig. 6.

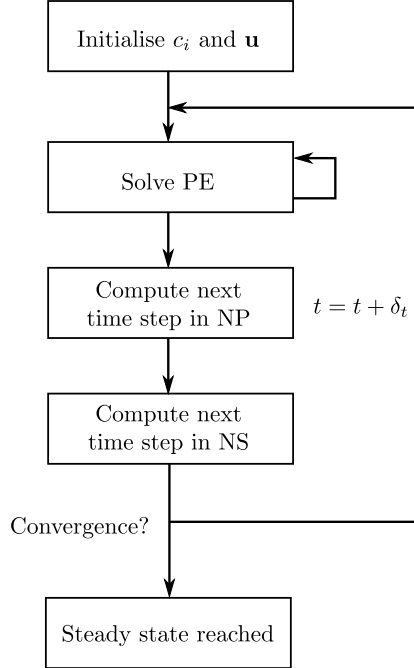


Figure 6: Flow scheme of the algorithm for solving the coupled equations.

13 Boundary conditions

In all physical situations, when solving a differential equations on a domain, conditions for what is happening on the boundary of the domain must be specified. So far, only the update rule for f_i on the interior of this domain has been treated. In this section we will also define rules for the boundaries of the domain. The nodes in the interior and the boundary will be referred to as *interior nodes* and *boundary nodes* respectively. Typically, a boundary condition in a macroscopic variable, e.g. velocity, is specified from the physical problem. This condition must be translated into a condition for the distribution function, f_i , on the statistical level. In this section some, to this work, useful boundary conditions will be formulated and discussed.

13.1 Bounce-back boundaries

The lattice-Boltzmann approach is often praised for its rather straight-forward easiness of implementing boundary conditions. One of the simplest and most commonly used is the bounce-back rule. It is a mesoscopic rule for implying a Dirichlet condition on the first moment. In the case of specifying boundary conditions for Navier-Stokes, it may be used to set a velocity at a boundary, typically at wall boundaries the velocity is set to zero.

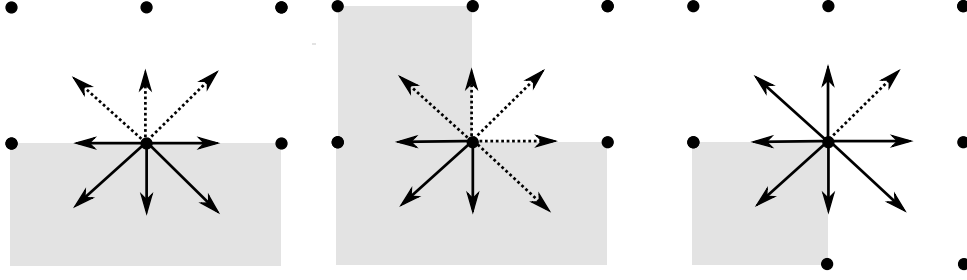


Figure 7: Three distinct boundary situations that in general has to be treated differently. From left to right, a straight boundary, a corner and an edge. Grey areas are outer of the domain. The solid arrows corresponds to known directions of the distribution function and dotted lines to unknown.

The bounce-back rule for setting the first moment to zero reads

$$f_i^{(bb)} = f_{i^*} \quad (110)$$

where \mathbf{c}_{i^*} is the direction opposite to \mathbf{c}_i . This justifies the name of the rule, all pseudo particles are propagated in a direction opposite to where they came from, i.e. bounced back. If the boundary nodes are updated between the collision and streaming step, all directions should be updated with its opposite counterpart. However, if the update is performed after the streaming step, only the unknown directions are to be updated, see fig. 7. These two schemes are referred to full-way and half-way bounce-back. If a non-zero Dirichlet condition is desired, it is possible to add some momentum to suitable directions.

It is possible to show that, in the case of a straight boundary, the actual boundary is with second order accuracy located half a node-node distance into the computational domain [?]. Thus, the boundary will not be located at the boundary node.

As suggested earlier in this section, this type of local boundary conditions allows for easy implementation of more or less arbitrary boundaries. In fig. 8, an example of 2D flow in a complicated domain is shown. At the walls in the figure, bounce-back conditions are used.

13.2 Slip boundaries

A zero friction (slip) boundary condition may be imposed by the following rule for the straight boundary situation in fig. 7. Instead of making all pseudo particles “bounce back”, they are reflected in a mirror-like manner, see fig. 9b. This gives that the component normal to the boundary of the first moment is zero while the tangential component is unchanged. In this work, this condition is used to set the ion flux through the boundary to zero, eq. (9).

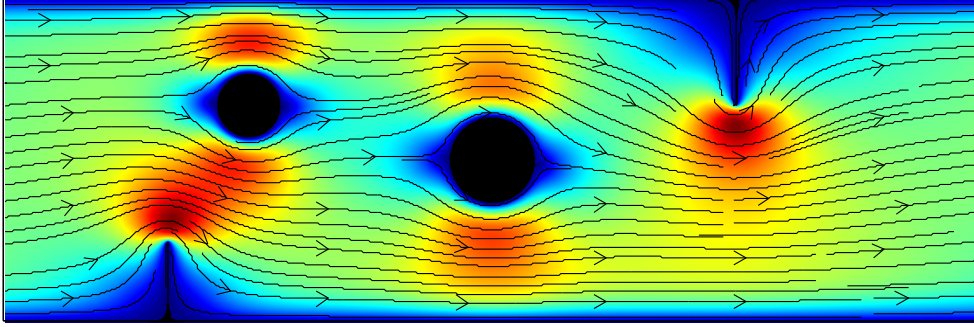


Figure 8: Example of flow in a non-trivial geometry where bounce-back boundaries are used.

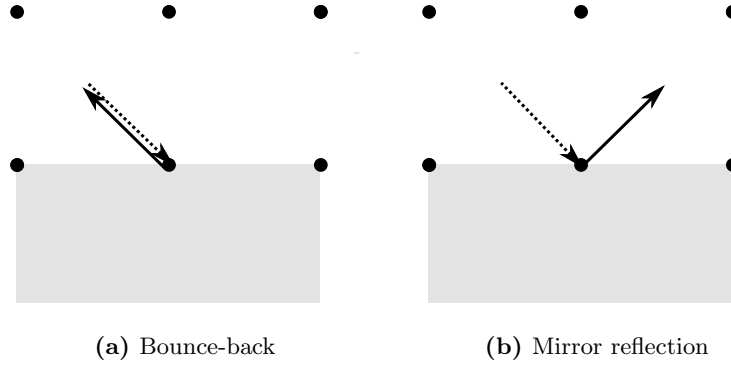


Figure 9: Intuitive scheme for an incoming pseudo particle for two common boundary conditions in the LBM. The bounce-back rule (a) and mirror reflection (b).

It is not as straight-forward to define rules at corner and edge nodes, as for the straight boundary nodes. From tests, it has been shown that adequate accuracy may be obtained by treating the corner or edge as two perpendicular straight boundaries and mirror reflect half of the distributions at each plane respectively.

13.2.1 With momentum addition

In the case with the fixed surface charge boundary condition, eq. (3), we do not wish to set the normal component to zero but to some value of the surface charge. This is realised by adding some “momentum” in the three directions pointing in to the domain. The total surface charge may be divided between the directions in different proportions, in this work it is however evenly distributed, i.e. one third per direction.

14 Physical and lattice units

A physical system may in its physical units contain quantities whose numerical values may differ a lot from each other. It is therefore, from a numerical point of view, a very bad idea and in the case with lattice-Boltzmann it will in general not work to just solve the equation in physical units. The equation is therefore scaled to dimensionless form by introducing characteristic quantities. For example a quantity A is scaled by A_0 to a dimensionless quantity A' through

$$A' = A/A_0 \quad (111)$$

Usually the dimensionless quantities are wanted to be of order one.

In the case with Navier-Stokes equation, three characteristic quantities are needed to put it on dimensionless form. I.e. one for the density (ρ_0), velocity (u_0) and length (ℓ_0). The non-dimensional incompressible N-S will contain one dimensionless parameter, i.e. the Reynolds number $Re = \rho_0 u_0 \ell_0 / \mu$.

For the advection-diffusion equation, in addition to a characteristic length and velocity, a characteristic concentration (c_0) is needed. One dimensionless parameter arise when non-dimensionalising the equation, i.e. the Peclet number, $Pe = u_0 \ell_0 / D$ where D is the diffusion coefficient.

Poisson's equation is non-dimensionalised by using a length and a characteristic voltage (V_0). The RHS then gets multiplied by factor ℓ_0^2/V_0 in the non-dimensional form.

When the non-dimensional form of the equations are determined, the domain is discretised. The non-dimensional length of the system is now typically 1 and $\delta_x = 1/N$ where N is the number of lattice cells used. From the diffusive scaling used, $\delta_t = \delta_x^2$ is usually a good choice.

15 Chapman-Enskog vs. regular expansion analysis

When reading literature and articles about the LBM, different approaches may be used to derive the macroscopic behaviour of the LBM. The aim of this section is to bring some clarity and briefly explain the main differences between two of these.

The method used in this work is referred to as regular (error) expansion analysis [?] and is used with one time scale. In the case of analysing the macroscopic behaviour of the LBM for the Navier-Stokes equations, the incompressible equations are obtained. The mass and momentum equations are exactly satisfied by $\rho^{(0)}$ and $\mathbf{u}^{(1)}$ from the regular expansions of ρ and \mathbf{u} respectively.

An other approach is by using the so called Chapman-Enskog analysis [?]. This is a traditional method in kinetic theory and is the most frequently used in literature about the LBM. Here two time scales are used, on faster (convective) and one slower (diffusive). This gives that in the macroscopic limit of the LBE, the compressible Navier-Stokes equations are obtained. In the C-E analysis, the full quantities ρ and \mathbf{u} are showed to satisfy, not the exact, but the N-S equations with higher order error terms.