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Lattice kinetic theory as a form of supra-molecular dynamics for computational microfluidics

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Abstract. We present a review of recent technical developments in Lattice Boltzmann Equations, as applied to single-phase flows with and without slip lenghts at the wall and for multi-phase flows in presence of hydrophobic walls. The interplay between roughness and hydrophobicity is discussed for microfluidics application. The issue of finite Knudsen effects is also addressed.

Key words: lattice kinetic theory, supra-molecular dynamics, computational microfluidics.

1. Introduction

The lattice Boltzmann (LB) method was proposed nearly two decades ago, with the main intent of providing an alternative to the discretization of the Navier-Stokes equations of continuum mechanics for the numerical simulation of complex flow phenomena [1,2].

To date, the LB method has indeed been developed mostly in the form of a competitive alternative to Navier-Stokes solvers for macroscopic flows. As current science and technology relentlessly move down the micro-nano-bio chain, there is great incentive in going 'back' to the atomistic roots and develop new LB models capable of dealing with micro and nanoscale fluid phenomena. The notion of LB as a 'catalyzer' of multiscale applications, either accelerating the overlap between the continuum and atomistic worlds, or providing a smoother marriage between the two (a sort of preconditioner) is tightly connected to this direction. A bolder possibility is to explore whether, at least for some applications, LB can thought as a unified approach to multiscale applications, i.e. an alternative to hybrid (continuum plus atomistic) approaches. In the final part of this paper, we shall provide a cursory view of the potential and limitations of the kinetic approach to the simulation of microfluids.

2. Lattice Boltzmann equation as a form of supramolecular dynamics

The mainstream strategy in the numerical simulation of microfluids is to solve the continuum equations of fluid mechanics, or generalizations thereof, and whenever these prove untenable on physical grounds, jump directly to the atomistic level and turn to Molecular Dynamics simulations. Kinetic theory, and most notably, the Boltzmann equation, are skipped altogether. Indeed, for most problems involving dense fluids, the Boltzmann equation is perceived either as inappropriate (the Boltzmann equation is formally derived for dilute gases), or, even accepting its finite-density extensions as physically viable, nearly as complicated as the Newton equations of motion (Molecular Dynamics) to be solved on digital computers. As a result, when it comes to the quantitative simulation of dense fluids and liquids, kinetic theory is often regarded as a kind of 'computational desert' between the continuum and atomistic descriptions. Over the last decade, however, this time-honoured view has been challenged by the advent of a minimal form of Boltzmann equations, known as Lattice Boltzmann (LB) equation. Under hydrodynamic conditions (negligible particle mean-free path over minimal macroscopic length scale), the LB is known to provide a very efficient alternative to the discretization of the Navier-Stokes equations. On the other hand, contrary to a rather widespread belief [3], LB is not restricted to dilute gases, nor to ideal fluids, the reason being that short mean free paths, as well many effects of potential energy interactions, can be accomodated through free-tunable transport coefficients and short-range density-dependent effective potentials, much in the spirit of density functional theory.

The result is that a number of microfluidic phenomena, and precisely those which do not depend on molecular specificity (henceforth dubbed 'supramolecular') present an ideal ground for mesoscopic kinetic methods. By supramolecular, we intend phenomena that escape a purely macroscopic description (for instance slip-flow) and yet do not depend on the specific detail of the interaction potential, but only on some of its integral properties, such as the ratio of potential to thermal energy

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ϵ/kT, or surface tension/contact angle. Whenever these integral properties can be mapped one-to-one onto the free parameters of the mesoscopic LB equation, the latter offers several orders of magnitude acceleration over atomistic methods, thereby providing de-facto a literal form of 'supra-molecular dynamics'.

A few simple considerations help illustrating the picture. Even assuming that LB operates at the same spatial scale as MD, its timestep is typically hundred times larger, because MD needs be advanced in time in timesteps much shorter than the typical interaction time $\tau_{MD} = \sigma/\sqrt{2\epsilon/m}$, whereas, thanks to its built-in conservation properties, LB can march in time steps of the same order as the collisional time, $\Delta t_{LB} \sim \tau_{LB}$. In the above, τ_{LB} is the timescale between two subsequent collisions, which in a dense fluid is comparable to the duration of the collision itself, τ_{MD}, the time to traverse a distance equal to the interaction range σ. The update of a MD degree of freedom typically involves the calculation of the force exerted by a few tens of neighbours, while the LB update requires the evaluation of the local fluid speed, which involves of the order of twenty coupled populations (in 3d) living on the same spatial cell. As a result, the update of a single LB degree of freedom is less expensive than the update of a single MD degree of freedom. Even under these restrictive assumptions, same spatial resolution and same computational cost per degree of freedom, LB would be about two orders of magnitude faster than MD. Further orders of magnitude are gained due to the fact that, at variance with MD, LB evolves a pre-averaged distribution function, which needs no form of enesemble averaging. The result is that LB is easily 4–5 orders of magnitude faster than MD, hence represents a method of choice for supra-molecular microfluidics. Of course, other forms of supramolecular dynamics have been developed before, such as Lattice Gas Cellular Automata [4], with the goal of accelerating hydrodynamic simulations at all scales, not only micro or nano-fluidics. These, however, still belong to the class of (stylized) many-body simulations, whereas in the sequel we should like to keep focus on kinetic theory at the level of one-body Boltzmann distributions.

Before delving into an illustration of these matters, a short survey of the basics of microfluidics is in order.

2.1. Basics of microflows. Micro and nanometric flows play a crucial role in many emerging applications in material science, chemistry, micro-engineering and biology [5]. A deeper understanding of the physics of fluids at the micro and nanoscale is paramount to these applications. The major aspect of micro/nanoflows is the substantial increase of surface/volume effects due to their reduced size. The immediate consequence is a significant enhancement of dissipative versus inertial effects (low Reynolds flows) which configures microfluidics as characterized by the competition between dissipative effects and pressure drive. This competition is formalized by the Stokes-equation for incompressible flows

$$\mu \Delta \boldsymbol{u} = -\nabla P \tag{1}$$

$$\nabla \cdot \boldsymbol{u} = 0 \tag{2}$$

plus boundary conditions. Due again to the high surface/volume ratioes, it is clear that boundary conditions become paramount to the overall dynamics of the microflow. In particular, they heavily bear on the capability of driving the flow across microchannels at an affordable energetic costs. A simple calculation makes the point. By adopting the standard no-slip boundary conditions, $u_{wall} = 0$, the Stokes equations in two-dimensional planar channel deliver the well-know Poiseuille parabolic profile

$$u(y) = 4U_c \left(1 - \frac{y}{H}\right) \frac{y}{H}$$

where

$$U_c = \frac{\Delta P H^2}{8\mu L}$$

is the centerline speed and H and L are the channel width and length respectively, while μ is the dynamic viscosity of the fluid. The flow rate across the channel is then given by

$$Q = UH^2$$

where $U = 2U_c/3$ is the average speed. This shows that, at a given pressure drop ΔP, the flow rate scales with the *fourth* power of the inverse surface to volume ratio, $H \sim V/S$. This power-4 law represents a very steep dissipative barrier to fluid motion at microscales, and the search for ways out of this barrier constitute one of the leitmotifs of microfluidic research. The second central issue of microfluidics is the potential breakdown of the continuum representation, as the size of the domain becomes comparable with the molecular mean free-path, that is, when the Knudsen number is no longer negligible. This issue is particular serious in gas microflows, where l_{mfp} is typically of the order of 1 micron, so that Knudsen numbers around $Kn \sim 1$ may be reached. Much less so in liquids, where molecules move just fractions of their size away from their equilibrium configuration. Yet, finite-Knudsen issues may become relevant also for liquid flows, whenever they develop thin gas films near the walls. The breakdown of the continuum approximation implies that the flow is no longer close to local thermodynamic equilibrium, so that, in principle, the full Boltzmann equation should be used. This clearly casts a serious question mark on the possibility of using LB for microfluidics. In fact, such possibility, was categorically ruled out in [6], on the basis of the fact that LB does not provide enough symmetry in velocity space to quantitatively describe the evolution of higher order moments of the distribution excited at finite-Knudsen regimes. Fortunately, this pessimistic view has proven over-restrictive, as we shall discuss in the following.

2.2. Slip-flow. We have pointed out that boundary conditions play a crucial role in microfluidics. From a macro-

scopic point of view, the physics of fluid-solid interactions is conveyed into the specification of appropriate boundary conditions, reflecting the gross features of molecular interactions at solid-fluid interfaces. The common tenet in continuum fluid dynamics is that fluid molecules in the immediate vicinity of a solid wall should move at the same speed of the wall: the so-called no-slip boundary conditions. However, it is well known that the no-slip boundary onditions do not follow from any basic principles of the physics of fluids. In fact they are often presented as a postulate of continuum mechanics. Indeed, it is well recognized that a variety of interesting fluids do exhibit a net motion relative to the solid wall, a phenomenon known as slip motion [7]. Slip motion may have dramatic consequences on the overall mass-flow rate, since it turns the 4-th power law into a quadratic one, thus implying a significant gain in efficiency, as typically measured by the flow gain factor

$$G = Q/Q_P$$

Q being the actual flow rate and Q_P the one associated with Poiseuille flow.

An important measure of slip motion is the slip length, l_s, defined as the extrapolated distance from the wall where the fluid speed matches exactly the wall speed. The slip length is generally proportional to the molecular mean-free-path, but it is known that for the case of specularly reflecting walls, the constant of proportionality may become anomalously large, so that significant deviations from hydrodynamics can result in the vicinity of the wall. Since the operation of many micro-devices depends crucially on fluid-wall interactions, it is important to model the effects of a non-zero slip coefficient on the transport properties of such devices. In this work we shall be concerned with the effects of a non-zero slip coefficient on the conversion efficiency of catalytic devices of millimetric size. Nano and micro-hydrodynamic flows have made the object of intense studies in the recent years, mainly by means of molecular dynamics (MD) simulations [8]. This MD information forms the basis for the set-up of correct boundary conditions to be used in macroscopic models of fluid flows. Since current MD simulations cannot reach scales beyond a few tens of nanometers, the coupling between MD and fluid models must necessarily proceed through a huge gap in space and time scales. Mesoscopic models are very appealing because they help reducing this gap considerably. In the bulk flow, LBE is essentially an efficient Navier-Stokes solver in disguise. At the solid interface, however, the mesoscopic nature of LBE reveals itself, because boundary conditions must be imposed on the particle distributions rather than on fluid quantities. This a mixed blessing: on the one side, it is a great advantage because boundary conditions keep being formulated on particle populations obeying the very same equation, rather than to additional fields obeying higher order partial differential equations. On the other hand, this leaves some mathematical ambiguity because there are generally more populations to fix than boundary constraints. This ambiguity has however been largely overcome by recent developments, as we shall illustrate in the sequel.

3. Lattice Boltzmann models for microflows

Possibly, the earliest LB simulation of microflows was performed by Nie *et al.* [9]. These authors pursued what one may call a minimal LB approach to microfluidics, that is, keep LB completely unchanged except for fixing the dynamic viscosity, $\mu = \rho\nu$, rather than the kinematic one, to a constant value. This change is needed in long-thin channels, where density changes along the streamwise coordinate cannot be ignored (experimental set-ups work typically with 100:1 aspect ratioes, with density drops $\rho_{out}/\rho_{in} \simeq 1$). Apart from this, anything else in the LB is left unchanged, and the Knudsen number is raised by simply taking large values of the relaxation time, according to the usual expression of the kinematic viscosity,

$$\nu = c_s^2(\tau - \frac{\Delta t}{2})$$

and then impose $\rho\nu = \mu_0$, which yields

$$\tau(\rho) = \frac{\Delta t}{2} + \frac{\mu_0}{\rho c_s^2}.$$

At solid walls, the standard bounce-back boundary condition was used. Nie *et al.* [9] report excellent agreement with experimental data, both in terms of the slip-velocity, density and pressure profiles. However, succesful match with the experimental data requires an adjustment factor, a, in the definition of the Knudsen number

$$Kn = ac_s(\tau - \Delta t/2)/H$$

where H is the channel height. The fact that such a minimal micro-LB does yield satisfactory agreement with (some) experimental data, is pleasing but also somewhat puzzling, since it is known from analytical solutions of LB in straight channel geometries, that the use of bounce-back boundary conditions (see below) leads to an artificial quadratic dependence of the slip length on the Knudsen number, while continuum kinetic theory gives a linear one, plus quadratic corrections. In fact, He *et al.* [10] show that the bounce-back condition leads to a quadratic dependence of the slip length on the Knudsen number

$$\frac{l_s}{H} = a + bKn^2$$

where the coefficients a and b depend on τ. This contradicts the linear Maxwell relation, as well as its second-order generalization due to Cercignani, that is:

$$\frac{l_s}{H} = 1.15Kn + 0.92Kn^2.$$

However, for moderate Knudsen, below unity, the numerical data do not differ significantly. Since microflows are characterized by high surface/volume ratioes, it is only

natural to expect that new types of boundary conditions are called for to quantitatively describe them. These were developed shortly later by a number of authors. Essentially, two classes of boundary conditions have been proposed: i) kinetic boundary conditions, patterned after the continuum kinetic theory of rarefied gases, ii) atomistic boundary conditions, in which fluid-wall interactions are explicitly included in the LB dynamics.

3.1. Kinetic boundary conditions. Due to the large surface/volume ratios the dynamics of microfluids is crucially affected by fluid-wall interactions. These can be decribed at several levels of detail, from the specification ofthe detail atomistic potentials, up to general statements on the velocity field at the wall, such as no-slip boundary conditions.

The mesoscopic description interpolates between the two. While molecular specifity lies beyond the realm of a mesoscopic treatment, it is nonetheless possible to formulate kinetic boundary conditions both in terms of adjustable accomodation coefficients, like in rarefied gas dynamics, or, closer to the atomistic scale, through the specification of coarse-grained pseudo-potentials in which the space-dependence of (soft) interactions is carried through the intermediate of the density field $\rho(r)$. The situation can be summarized as follows:

- Microscopic: atomistic potentials $V(r)$
- Mesoscopic/micro: pseudo-potentials $\Psi[\rho(r)]$
- Mesoscopic/macro: accomodation coefficients (r, s)
- Hydrodynamic: slip-length l_s

At the meso/macro level, a key control parameter is the Knudsen number, so that the accomodation coefficients must be adjusted mainly (but not exclusively) on the basis of this parameter. In the meso/macro description, the strength of the (effective) interaction, as well as its range can be specified in fully analogy with molecular dynamics practice. In the sequel, a few examples of both strategies will be provided.

A correct set of boundary conditions for low-Knudsen flows has been formulated by Ansumali-Karlin [11]. These authors developed a lattice transcription of the well-known 'full-accomodation' model in rarefied gas dynamics, known also as diffuse boundary conditions. The idea, dating back to Maxwell [12], is that molecules impinging on the wall loose complete track of their incoming speed, and consequently, they are reinjected along a random direction with a velocity drawn from a Maxwellian at the local wall speed and temperature. The fact that the lattice transcription of these conditions proves realizable, represents a major progress in the application of LB techniques to microfluidics.

3.2. Partial-accomodation kinetic boundary conditions. An empirical generalization of the bounce-back rule, allowing for partial slip, besides reflection, at the wall, was introduced in [13] (this has been studied before in the context of LGCA's [14]).

To discuss the slip-reflection rule, let us remind that the no-slip boundary condition, $\vec{u} = 0$, is typically imposed by reflecting the outgoing populations back into the fluid domain via the so-called bounce-back rule. A useful generalization consists of making allowance for a mix of bounce-back and specular reflections. For instance, with reference to a planar 'north' wall, we have ($f_{\searrow}$ indicates a south-east moving population, and similar for other directions):

$$f_{\searrow}(x, y, H+1) = r f_{\nwarrow}(x+1, y, H) + s f_{\nearrow}(x-1, y, H) \quad (3)$$

where r is the reflection coefficient and $s = 1 - r$ is the slip-coefficient.

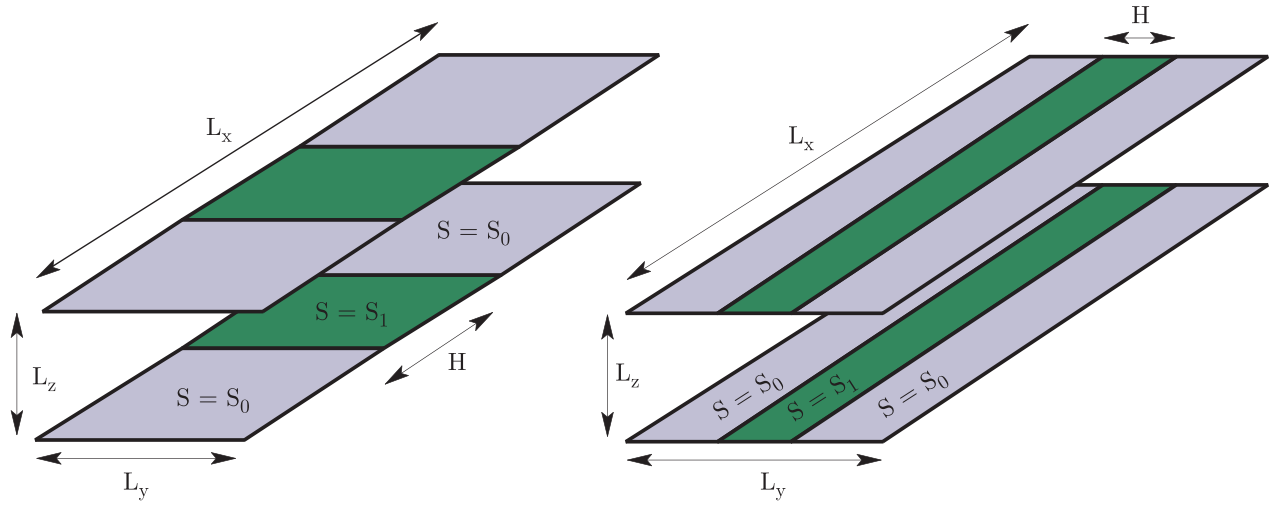

Fig. 1. Typical geometry of a microchannel configuration. We have periodic boundary conditions along the stream-wise, $\hat{x}$, and span-wise $\hat{y}$ directions. The two rigid walls at $z = 0, L_z$ are covered by two strips of width H and $L - H$, where $L = L_x$ for transversal strips (left panel) and $L = L_y$ for longitudinal strips (right panel). The two strips have different slippage properties identified by the values s_0 and s_1. The ratio $\xi = H/L$ identifies the fraction of hydrophobic material deposited on the surface. Typical sizes used in the LBE simulations are $L_x = L_y = 64$ grid points and $L_z = 84$ grid points. This would correspond, for example, for an ordinary gas at $Kn = 10^{-3}$, to a microchannel of height of the order of $100\,\mu$m. Details are after Ref. 16

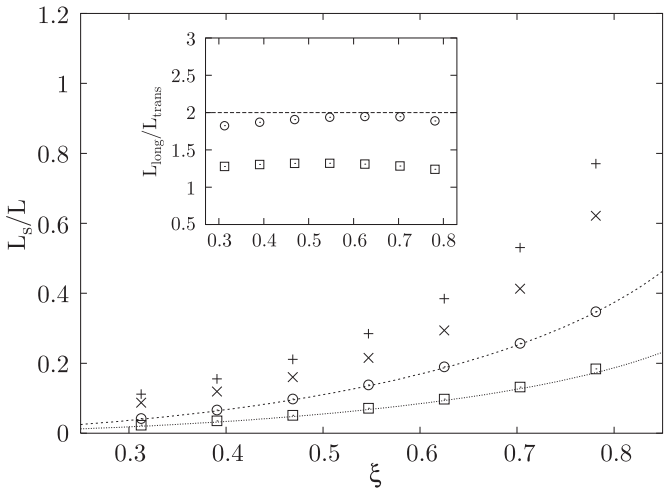

Fig. 2. Normalized slip length for transversal and longitudinal strips with $s_1 = 1$, $s_0 = 0$. We plot the normalized slip length as a function of the slip percentage ξ. The system's dimensions are those of Fig. (1). A first set of LBE simulation is carried out at small Knudsen, $Kn = 1.10^{-3}$ for transversal (□) and longitudinal strips (○). These results are compared with the analytical estimates of [20] (dashed line) and [19] (continuous line). Notice the perfect agreement with the analytical results in the hydrodynamic limit [19]. Another set of simulations is carried out with much larger Knudsen, $Kn = 5.10^{-2}$ to highlight the effect of rarefaction on the system for both traversal (×) and longitudinal (+) strips. In the inset we show the ratio between the slip lengths for parallel and longitudinal strips for $Kn = 1.10^{-3}$ (○) and $Kn = 5.10^{-2}$ (□). Here we notice how by increasing the Knudsen number the orientation of the strip region with respect to the mean flow becomes less important. Details are after Ref. 16

This three-site interaction is more complicated than pure bounce-back, but permits to model free slip motion at the solid interface. Within this approach, the reflection/slip coefficient must be regarded as a free parameter, to be calibrated through comparison with experimental data. In particular, the amount of slip-flow is a very sensitive function of s in the near vicinity of $s = 1$, where it diverges like $(1 - s)^{-1}$. Although the linear superposition given by eq. (3) might not be the most general one (we are grateful to an anonymous referee for bringing this point to our attention), it is nonetheless flexible enough to permit the development of sensible and experimentally realizable generalizations of purely hydrodynamic boundary conditions, as it is shown in the sequel.

The realizability of the reflect-slip boundary conditions has been recently proven analytically by Sbragaglia and Succi [15,16], who could also show that it correspond to a lattice transcription of the popular Cercignani-Lampis scattering kernel [17], for the case of athermal flows. This model could reproduce experimental data up to second order in the Knudsen number. This scheme has been succesfully extended to heterogeneus conditions, i.e. with r, s depending on the local position on the wall, to compute slip flows over micro-patterned walls [16,18], in excellent agreement with analytical solutions [19] (see Figs. 1 and 2).

Summarizing, Ansumali and Karlin identified the correct framework to formulate lattice boundary conditions [20] for non-zero Knudsen numbers. Their model was confined to the case of full-accomodation. Generalizations to partial-accomodation (slip-reflect) were formulated first on an empirical basis in [13], and subsequently backed up by analytical work in [15].

3.3. Volumetric boundary conditions. General boundary conditions capable of dealing with arbitrary complex geometries have been proposed years ago by H. Chen [21] and succesfully implemented in commercial LB software [22]. These are based on a volumetric formulation of the LB scheme, which permits to handle the fluid-wall interactions in terms of coupled dynamics of volume and surface elements (voxels and surfels respectively). These boundary conditions can handle the complex boundary layers associated with turbulent flows. Very recently, these have also proven capable of successfully reproducing microflow behaviour, such as slip-flow and the Knudsen paradox [23]. Since these boundary conditions are explicitly designed to handle boundary-layers, it is perhaps not surprising that they can handle slip-flow situations too.

3.4. Fluid-wall potentials. A more microscopic approach consists of introducing non-local fluid-wall potentials in the LBE, typically in exponential form [24]:

$$V_{FW}(y) = G_{fw} e^{-y/w}$$

where y is the normal-to-wall coordinate and w the range of the wall potential, typically a few lattice sites, and G_{fw} is the coupling strength. With explicit inclusion of wall potentials, the onset of slip-flow onset proceeds as follows [25]. The repulsive wall-fluid potential gives rise to a depletion layer near the wall, a so-called 'dry-layer'. Due to this density drop, the dynamic viscosity also drops down, so that the near-wall fluid can slide away as compared to the bulk region. This scenario provides the following expression for the slip length [25]:

$$l_s/w = e^{G_{fw} w/T}.$$

This expression clearly shows that, given the nanoscopic value of w, unrealistically high ratioes of potential to thermal energy are required to account for micrometric or even millimetric slip lengths reported in experiments and molecular dynamics situations. Among many other possibilities, whose discussion is beyond the scope of the present work, a way out of this problem is provided by the synergistic cooperation between fluid-wall and fluid-fluid potential interactions in the bulk, which can lead to dramatic magnification of the effective fluid-wall coupling (for details, see [25]), thereby permitting to fill the gap between nano and micrometric distances. The robustness of such 'inflationary' scenario remains to be assessed.

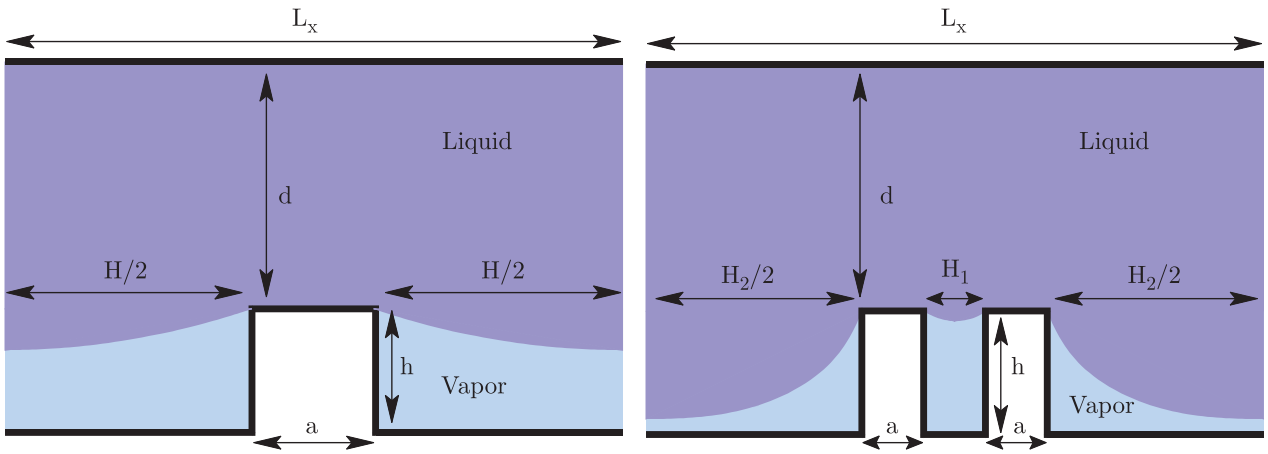

Fig. 3. Left: homogeneous roughness. A groove with depth $h = 33\Delta x$ and width $H = L_x - a$ (with $a = 10\Delta x$) is introduced on the bottom wall and periodic boundary conditions are assumed along x. In this configuration, the presence of vapor pockets inside the groove changes the "effective" boundary conditions felt by the bulk fluid, with a net decrease of drag when a pressure drop is applied. Right: channel with heterogeneous roughness. Two grooves of width $H_1 = 40\Delta x$ and $H_2 = 70\Delta x$ are present. The two grooves are filled separately at different values in the pressure/density diagram. The lattice spacing corresponds to $\Delta x \sim 0.3nm$. Details are after Ref. 29

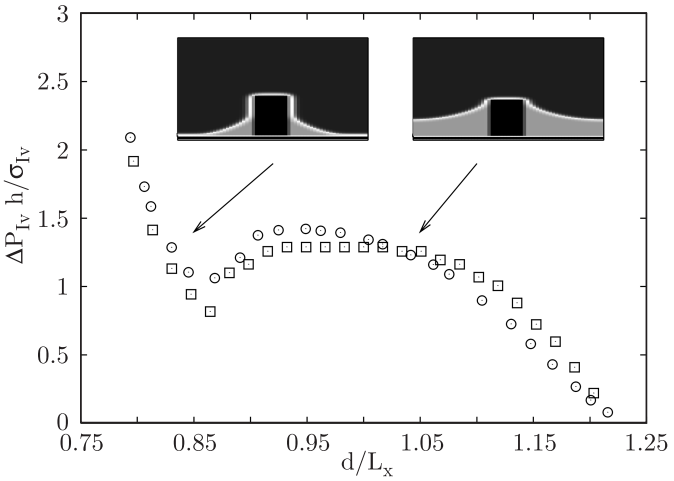

Fig. 4. The normalized pressure drop, $\Delta P_{lv} h/\sigma_{lv}$ between the two bulk phases, is shown as a function of the normalized distance d/L_x (see Fig. 3). LBE (□) results have been obtained with a contact angle $\theta = 160^o$. MD results with the same contact angle are plotted with (○) [30]. The two insets represent the density configuration at the onset of the wetting/dewetting transition (right) and for a wetted configuration (left). The plateaux in the pressure curve defines the capillary pressure, P_{cap}. Details are after Ref. 29

3.5. Fluid-wall pseudo-potentials. A rich variety of complex microscopic flows is being simulated with LB schemes for non-ideal fluids, both in pseudo-potential [26] and free-energy versions [27]. Here, the versatility of LB proves very valuable. For instance, by introducing separate fluid-fluid and fluid-wall pseudo-potentials, it is possible to provide a seamless and powerful description of droplet interactions with chemically heterogeneus surfaces, micro-patterned surfaces, and many other phenomena of great interest to chemical engineering [24,27]. Very recently, a similar approach based on the extension of the Shan-Chen pseudo-potential model, has proved capable of quantitatively reproducing the major features of finite-size dewetting transitions and related super-hydrophobicity effects [29] (see Figs. 3 and 4 where a direct comparison between LBE and MD is shown for the case of a microflows in a corrugated device).

4. Success and points of criticism

To date, a growing body of results indicates that LB methods can quantitatively (that does not necessarily mean accurately in the sense of numerical analysis...) predict a number of non-trivial features of microflows under fairly complex situations, and sometimes even in the finite-Knudsen regime. The latter result contradicts some pessimistic no-go statements, according to which LB can only be used strictly within the low-Knudsen hydrodynamic regime. Essentially, the point is that in the bulk, the LB errors scale linearly with the Knudsen number, so that they remain numerically negligible up to $Kn \sim 1$. Since most interesting applications for microfluidics are in the range $Kn \sim 0.1$, finite-Knudsen errors in the bulk are not a serious threaten, unless something real wrong takes place at the boundary. In addition, for athermal flows, it can be shown analytically that the LB scheme would eventually miss the Knudsen layer in the velocity profile, but still get the correct slip flow [30,31]. In essence, this indicates that LB can work well whenever the slip layer does not couple significantly with the bulk solution. On the other hand, a few points of criticism still remain.

4.1. Anisotropic Knudsen layers and spurious currents. As proved long ago by Cornavin et al, [32], solid walls not aligned with the lattice grid are known to excite spurious and highly anisotropic Knudsen-layers, which may contaminate the physical solution in the inside region

of the flow. The question remains as to whether a sufficiently accurate discretization, jointly with a volumetric formulation of the LB scheme, may significantly dampen the problem.

Strictly related is the issue of spurious currents near curved interfaces, which are also due to the lack of symmetry of the lattice at the higher orders (fourth-order symmetry is sufficient to recover correct hydrodynamic behaviour in the bulk). Although it has recently been shown [33,28] that these spurious currents do eventually vanish in the limit of infinite resolution and/or perfect isotropy of the lattice, the practical implementation of these results appears problematic.

The problem of spurious currents is particularly felt for dynamic problems, such as the moving-contact-line problem in immiscible flows. It is well known that a purely hydrodynamic treatment of the moving contact line problem, i.e. a gas-liquid interface sliding over a solid wall, meets with a singular behaviour at the contact point between the three phases. Of course, this singular behaviour signals the inadequacy of a continuum treatment, and indeed molecular dynamics studies have shown that the singularity is regularized at atomistic scales. Despite some preliminary investigations [34], it is still not clear whether LB, as we know it, can achieve a similar, quantitatively correct, regularization. Here again, the issue is obscured by spurious currents, which may well be of the same order of magnitude of the physical effects, e.g. rolling motion at the interface, often invoked to resolve the singularity issue. Much detailed work is needed to understand whether LB can indeed provide quantitative insights into these complex microfluidic phenomena beyond the hydrodynamic realm.

4.2. Mapping to physical units. It is well known that LB interfaces cannot be made thinner than a few lattice spacings. Since real interfaces are just a few nanometers wide, realistic resolution of such interfaces would force the LB mesh spacing to a few nanometers, thus rendering the method totally unpractical for realistic microfluidic computations, extending to a few millimiters in size. This is particularly apparent for the fluid-wall interatomic potential approach, in which the Boltzmann distribution experiences an explicit space-dependent potential. The pseudo-potential approach, however, is much less exposed to this criticism, because pseudo-potentials and free-energy LB's are -by default- coarse-grained versions of the atomsitic potentials, hence they operate on supra-molecular scales.

More generally, it should be reminded that, by design, mesoscopic method make strong advocacy to universality. By this, we refer to situations in which the relevant physics is governed by dimensionless groups of variables, rather than by the physical values of the variables themselves.

4.3. Thermal effects. A substantial criticism regards thermal effects: since heat exchange plays no negligible role in microflows, some authors claim that realistic LB models for microflows must necessarily account for thermal phenomena. This is a serious issue, for it is known that, even for ideal fluids, thermo-hydrodynamic LBE's are way less robust than their a-thermal counterparts. Entropic LB schemes with thermal capabilities have been developed by Ansumali and Karlin [35]. However, these schemes go necessarily off-lattice and require delicate interpolations. A further thermal model has been developed to study the behavior of nonideal fluid systems with two phases of liquid-vapor type [36]. In general, the dynamics of a multi-phase mixture below the critical temperature requires the use of a full set of macroscopic equations which take into account the conservation of mass, momentum and energy. The heat equation has to be explicitly taken into account if realistic situations where the temperature is not constant have to be simulated. Indeed, non-isothermal situations occur for example when the system is placed in contact with two walls at different temperatures or when adiabatic quenches need to be considered.

Finally, to the best of these author's knowledge, most LB microfluidic simulations to date are performed in simple geometries. It remains to be seen whether microflows in complex geometries can still be handled without requiring higher-order lattices. For sure, there will be a lot to learn on this fast-moving front of LB research in the coming years.

5. Conclusions

As discussed in the Introduction, the LB was born to address the numerical simulation macro-fluid dynamic problems. Nearly twenty years on, with LB applications mushrooming across most subfields of fluid-dynamics, it is only fair to say that it has largely succeeded in fulfilling the task. More recently, an increasing number of investigators have started to explore the LB capabilities back to where LB comes from, i.e. sub-hydrodynamic scales, namely micro and, to a lesser extent, nano-fluidics as well. Here, a number of formal/theoretical no-go issues immediately arise, since the LB theory is only backed-up in the low-Knudsen, macroscopic limit, where the Chapman-Enskog analysis applies. Whence, the restrictive view that LB can only be applied there, where the Chapman-Enskog analysis holds. Fortunately, this restrictive view has proven to be over-pessimistic, and, as we speak, dozens of papers have shown that LB continues to provide useful information also in microfluidic situations beyond the realm of continuum hydrodynamics. This opens up the exciting prospect of LB, and more generally, kinetic theory, as an effective form of Supra-Molecular Dynamics. Of course, assessing the actual capability of LB in this direction requires a wide body of detailed and quantitative work for the future. Prospects look bright.

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