

# Chimera Multiscale Simulation of Complex Flowing Matter

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## Abstract

We discuss a unified mesoscale framework for the simulation of complex states of flowing matter across scales of motion which requires no explicit coupling between different macro-meso-micro levels. The idea is illustrated through selected examples of complex flows at the micro and nanoscale.

## 1 Multiscale without hand-shaking: the chimera concept

Multiscale modeling is the natural response to the hierarchical organization of the interactions which shape up the complexity of the world around us. Traditional multiscale computing is based on the notion of *overlapping* between the four basic levels of the BBGKY hierarchy, macro, meso, micro and quantum [1]. The overlap between these levels is sustained by the continued advances in computer hardware and modelling techniques. Yet, the gaps remain daunting, easily ten decades in space (say Angstroms to meters) and twice as many in time (femtoseconds to hours). In addition, the practical implementation of the hand-shaking interfaces across the different levels remain laborious and technically intensive [2], [3], [4]. Under such state of affairs, it is highly desirable to explore different multiscale strategies based on a *unified* treatment of multiple levels of the hierarchy without handshaking interfaces. In this context, the concept of overlapping is replaced by the idea of *morphing*, i.e. the unified scheme should be able to morph into the desired level, say micro and continuum, wherever and whenever needed "on demand". This is what we shall refer to as to *chimera* multiscale. At variance with the original mythological meaning, to us the chimera is not a monster, but a graceful and flexible creature!

With the above premises, it is clear that mesoscale representations, and particularly kinetic theory, lie at a vantage point to implement the chimera strategy, since they offers a natural intermediate description between the micro and macro descriptions. Indeed, Boltzmann kinetic theory is based on a continuum field, the probability distribution function, propagating along particle-like trajectories. This offers a natural particle-field duality which can prove pretty useful for multiscale purposes. For instance, instead of coupling continuum hydrodynamics to atomistic models, the mesoscale chimera would morph, so to say, into both these levels by suitable "mutations" which take it close to Navier-Stokes on the upper end and to Newtonian mechanics on the lower one. By doing so, the need for handshaking between different levels is lifted altogether. Of course there are limits to this strategy,

In the sequel of this paper, we shall provide a cursory description of the mutations which promote this chimera behaviour, and discuss its potential and limitations.

## 2 Universality and Molecular Individualism: Weakly Broken Universality

Before doing so, let us address a natural question: how far can we push the (unconventional) idea of chimera multiscale? In other words, how close can one take the mesoscale description to the upper macro and lower micro levels, before hitting the ceiling (floor) where handshaking can no longer be postponed? The ceiling part is solid: it is known that, under the appropriate conditions (weak departure from local equilibrium), the Boltzmann equation converges to the Navier-Stokes equation. Here the morphing is guaranteed to be convergent. The floor side, on the other hand, tells another story. It is indeed obvious that Boltzmann's kinetic theory cannot be made equal to molecular dynamics. How close the two can be brought together is controlled by the degree of universality exposed by each given problem. More precisely, by the degree of *broken* universality, meaning

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by this the departure from universal behaviour, the one associated with the continuum description of moving matter. On the other hand, complex moving matter is characterized, and we might say even *defined*, by the coexistence of Universality and Molecular Individualism, to an extent which changes from problem to problem. It is hereby surmised that the chimera approach is well suited to capture the fascinating physics stemming from such coexistence. If universality is only weakly broken (WBU), i.e. the physics under exploration requires more descriptors, fields and parameters, than continuum hydrodynamics, but still no strict knowledge of molecular details. Under such conditions, the Chimera approach can provide major computational returns. Otherwise, conventional hand-shaking cannot be helped. In the following, we shall substantiate the above program by means of concrete examples based on a specific mesoscale technique, known as Lattice Boltzmann (LB) method, which we next proceed to briefly illustrate.

### 3 Lattice Boltzmann in a nutshell

The LBE, in single-relaxation form for simplicity [5], reads as follows [6, 7, 8, 9]

$$f_i(\vec{x} + \vec{c}_i \Delta t, \vec{c}_i; t + \Delta t) - f_i(\vec{x}, \vec{c}_i; t) = \omega(f_i^{eq} - f_i) + \gamma S_i \quad (1)$$

where the index  $i$  labels the set of discrete velocities. In the above,  $f_i^{eq}$  denotes a lattice version of the Maxwell-Boltzmann local equilibrium, parametrised by the fluid density  $\rho = \sum_i f_i$  and velocity  $\vec{u} = \sum_i \vec{c}_i f_i / \rho$ , both functions of space and time.

The parameter  $\omega = \Delta t / \tau_c$  is the ratio of free-streaming versus collision timescales,  $S_i$  is a source term associated with external/internal forces and  $\gamma = \Delta t / \tau_S$  is a measure of the strength of the external versus collisional relaxation.

In other terms,  $\omega$  measures the strength of collisions versus free propagation, so that  $\omega \rightarrow \infty$  connotes the strongly-coupled hydrodynamic regime, whereas  $\omega \rightarrow 0$  denotes the weakly-coupled non-hydrodynamic regime leading to free molecular streaming. Likewise,  $\gamma \rightarrow \infty$  denotes strong interaction with external (or internal) driving sources, while  $\gamma \rightarrow 0$  indicates weak coupling to the source. For the case of sources of momentum, the parameter  $\gamma$  is proportional to the Froude number  $Fr \equiv \frac{v_{th} \tau_c}{a}$ , where  $v_{th}$  is the thermal speed and  $a = F/m$  the acceleration due to the force  $F$ . Linear stability imposes the constraint  $0 < \omega < 2$ , which secures positive kinematic viscosity through the relation  $\nu = c_s^2 (1/\omega - 1/2) \frac{\Delta x^2}{\Delta t}$ . Likewise, the parameter  $\gamma$  should be taken well below the unit value, on pain of ruining the stability of the scheme, as we are going to detail in the following.

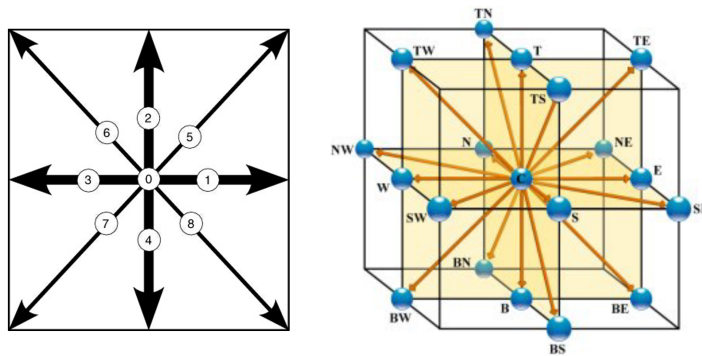


Figure 1: The D2Q9 lattice, i.e. nine discrete velocities in two spatial dimensions and the D3Q19 lattice, i.e. nineteen discrete velocities in three spatial dimensions. These lattices provide fourth order isotropy and permit to recover the isothermal, quasi-incompressible Navier-Stokes equations.

#### 3.1 Interacting fluids

One of the main merits of the LB formalism is its flexibility towards the inclusion of complex physics beyond the realm of Navier-Stokes hydrodynamics, through the source term  $S_i$ . The typical case are force fields, representing either coupling with the external environment, as well as internal forces resulting from inter-molecular interactions. A very general and fruitful expression of LB force fields is [10]:

$$\vec{F}(\vec{r}; t) = \psi(\vec{r}; t) \sum_i \vec{r}_i G_i \psi(\vec{r} + \vec{c}_i \Delta t; t) \quad (2)$$

where the sum extends to a suitable neighborhood of the lattice site  $\vec{r}$  and  $G_i$  is the coupling strength of the interactions along the  $i$ -th link [11]. A suitable tuning of the local functional  $\psi(\vec{r}; t) \equiv \psi[\rho(\vec{r}; t)]$  permits to

recover the main features of interacting fluids, namely non-ideal equation of state, surface tension and various type of dispersion forces.

The key observation is that such forces can be reabsorbed into a shifted-local equilibrium of the form:

$$f^{eq} = f_M(\vec{c}), \quad \vec{c} \equiv \frac{\vec{v} - \vec{u} - \frac{\vec{F}\tau}{m}}{v_{th}},$$

where  $f_M$  denotes the Maxwell-Boltzmann distribution and  $v_{th} = \sqrt{\frac{k_B T}{m}}$  is the thermal speed.

As a result, the powerful stream-collide paradigm of the force-free formulation is kept intact: the new physics is simply encoded in the force shift. This permits to incorporate a broad class of (weakly) interactions beyond hydrodynamics, without loosing the major computational perks of the force-free formalism.

Of course, this is limited to weakly coupled fluids, with Froude numbers well below unity,  $Fr \ll 1$ , a situation which covers nonetheless a broad variety of soft matter flows.

In the following, we shall provide a few concrete examples drawn by recent and past experience in the field.

## 4 Chimera in action: nano and micro-flows

LB can be taken to the nanoscale, provided fluctuations are properly reinserted through the source term  $S_i$ , this time a stochastic source compliant with the fluctuation dissipation theorem. On the other hand, for flow quantities resulting from time and ensemble averaging over sufficiently long intervals as compared with the collisional scales, one may stay with deterministic forces expressing just the proper time-averaged interactions. This permits to simulate a host of interesting nano-fluidic phenomena which are not directly amenable to a continuum description and yet too demanding for molecular dynamics.

### 4.1 Super-hydrophobicity

Super-hydrophobicity is the counterintuitive phenomenon by which fluids may experience less friction by flowing on corrugated surfaces than on smooth ones. The reason for this "paradox" is that, under proper physico-chemical conditions, the development of a vapour film between the solid wall and the flowing liquid proves able of lowering the free-energy budget required to sustain the three phase (Solid-Liquid-Vapour). Formally:

$$\sigma_{VS}S_{VS} + \sigma_{LV}S_{LV} < \sigma_{LS}S_{LS}$$

where  $\sigma_{AB}$  denotes the surface tension between phases  $A$  and  $B$ , inclusive of the associated contact angle and  $S_{AB}$  is the area of the corresponding contact surface. Super-hydrophobicity was pointed out by molecular dynamics simulations of microscale flows over nanometric corrugations [12] (see Fig. 2, left panel). In particular a non-monotonic relation between the pressure deficit between the liquid and vapour phases as a function of the size of the corrugation was computed and shown to exhibit a loop structure, supporting a so-called de-wetting transition: the fluid in contact with the wall transits to the vapour phase, while the bulk remains liquid. If the physics of super-hydrophobicity is controlled only by transport parameters such as surface tension and contact angle, there are good reasons to believe that molecular dynamics should not be needed to unravel it. On the other hand, it is not immediately obvious that the above nanoscale features can be readily included within a continuum description. Under such state of affairs, it may argued that a mesoscale approach, equipped with suitable interfacial interactions, should be able to capture superhydrophobic behaviour at a tiny fraction of the cost as compared to MD.

This is indeed the case, as first shown in LB simulations [13], which were able of reproducing the pressure-geometry "equation of state" to quantitative accuracy as compared to MD data.

#### 4.1.1 What did we gain?

It is of interest to point out that such simulations required a sub-nanometric mesh-spacing,  $\Delta x_{LB} = 0.3$  nm, i.e. basically the range of molecular interaction.

One might thus wonder what have we gained in the process of using LB instead of MD.

The advantage is arguably three-fold.

First, even though the LB mesh-spacing is the same as the molecular range, the LB time-step still is at least an order of magnitude larger:  $\Delta t_{LB} \sim \frac{\Delta x}{c_s} \sim \frac{r_0}{c_s}$  versus  $\Delta t_{MD} \sim 0.1 \frac{r_0}{c_s}$ . This is because LB is stable up to  $\Delta t_{LB} < 2\tau_c$ , where  $\tau_c \sim r_0/c_s$ , with mass and momentum conserved to machine accuracy, while MD requires a time-step much smaller than  $\tau_c$  in order to secure accurate energy conservation.

Second, updating a single LB degree of freedom (the discrete population  $f_i$ ) is significantly faster than the update of a single MD molecule. This is because the LB interactions are lattice-bound, hence do not require

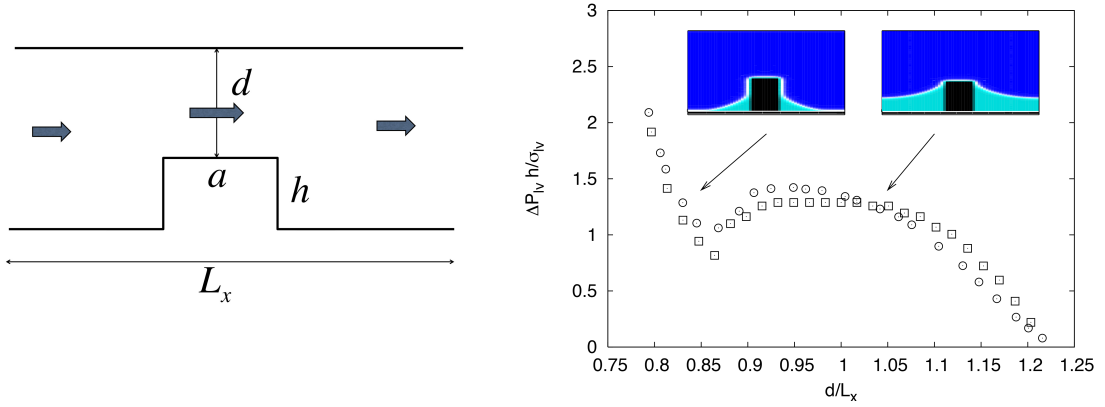


Figure 2: Super-hydrophobic nanoflow. Left: the geometrical set-up of the flow with nano-corrugations. Right: Pressure drop between the liquid and vapor phase as a function of the distance  $d$  between the top of the nano-corrugation and the upper wall of the micro-channel. The liquid within the nano-corrugation de-wets from solid wall leading to a vapour phase which sustains the liquid flowing on the top. The liquid then flows on the vapour with less friction than on the smooth solid wall. From [13].

the construction of a dynamic link-list to identify the interacting neighbours. This counts easily another order of magnitude in favour of LB.

Finally, LB requires no statistical average, since, by definition, the distribution function represents a large number of molecules.

Taken all together, this gives a two or three orders of magnitude LB/MD speed-up, for the same spatial size of the problem. In other words, LB performs a substantial coarse-graining in time.

Of course, this is **not** MD, since MD would account for molecular fluctuations completely neglected in the present LB formulation. However, for the purpose of investigating the physics of superhydrophobic transport, such fluctuations are inessential, thus setting a needless tax on the MD simulations.

## 4.2 Suppression of fast water transport in graphene oxide

The same effect has been highlighted again for the study of fast water transport in graphene oxide nano flows. While pure graphene sustains slip flow, with mass flow rates far exceeding the hydrodynamic value, graphene oxide, exhibiting hydroxyl groups protruding from the walls, is believed to suppress such fast water transport (FWT) regime. The debate on whether the nano flow is hydro on molecular is pursued by either Navier-Stokes with slip boundary condition or molecular dynamics. In order to mimic the friction effect due to the protruding molecules, a mesoscale LB model including a frictional interaction at the interface has been developed. The frictional force takes the following form

$$F_x(y) = \gamma_0 e^{-y/w} u_x(y)$$

where  $\gamma_0$  is a typical collision frequency between the water molecule and the oxydril groups,  $w$  the size of the hydroxyl molecules and  $u_x$  the water flow along the mainstream direction  $x$ ,  $y$  being the crossflow direction.

This model proves capable of predicting the breakdown of the FWT regime and also highlighted a subtle coexistence of collective motion in the bulk and individual molecular motion in the near-wall region. The LB model runs about two orders of magnitude faster than MD simulation.

## 4.3 The Knudsen paradox

A typical manifestation of non-equilibrium effects beyond the hydrodynamic picture is the so called Knudsen paradox. According to hydrodynamics, the mass flow rate should scale inversely with the Knudsen number

$$Q_{hydro} = \frac{1}{12} \frac{g}{\nu h^2} \propto \frac{1}{Kn}$$

where  $g = \nabla p / \rho$  is the pressure drive along the channel of width  $h$  and the Knudsen number is defined as  $Kn = \frac{\lambda}{h}$   $\lambda$  being the mean-free path. Since the Knudsen number is proportional to the kinematic viscosity small Knudsen imply low dissipation hence high mass flow, and viceversa. The viceversa part, however, is deceiving, since the above proportionality only holds if  $Kn \ll 1$ .

Indeed, actual experiments show that beyond a critical value, around  $Kn \sim 1$ , the mass flow starts to increase with the Knudsen number, a typical signature of individual molecular behaviour, i.e. free-streaming.

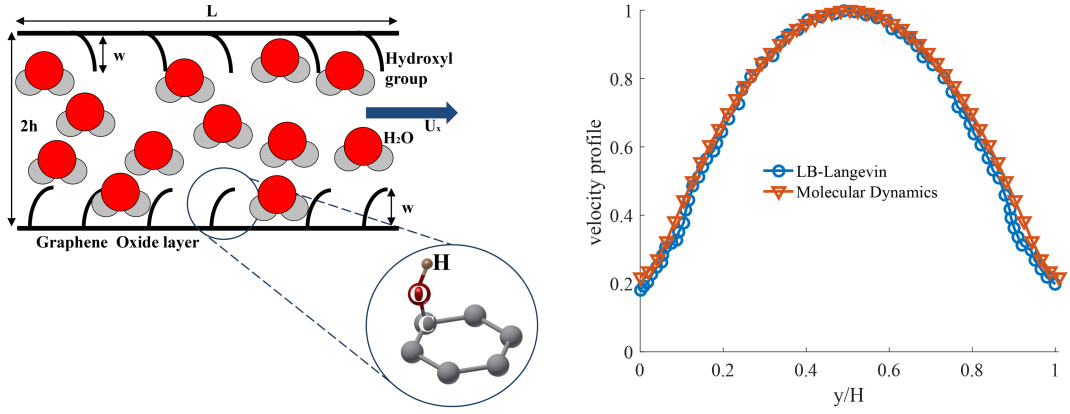


Figure 3: Examples of WBU nanoflow. Left: Hydroxyl groups protruding from graphing oxide walls in a nanometric channel. Right, the velocity profile computed with LB (blue) and MD (Right).

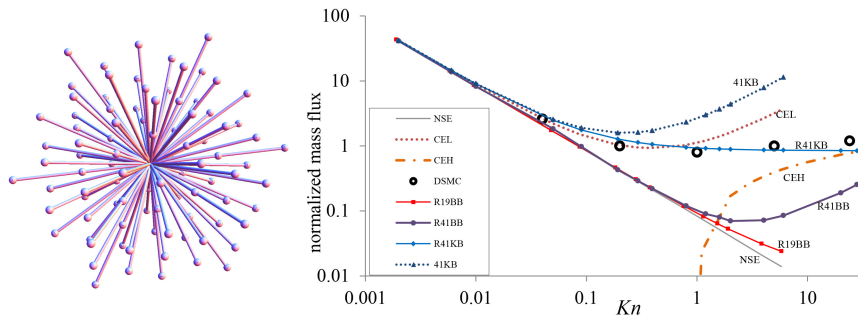


Figure 4: Left: a high order lattice with 93 discrete speeds, featuring eighth order isotropy. Right: the mass flow rate as a function of the Knudsen number for a two-dimensional channel flow. The open circles refer to the DSMC solution, which is taken as a benchmark reference. CEL and CEH refer to analytical asymptotic solutions obtained by Cercignani in the low (CEL) and high (CEH) regimes, respectively. NSE reports the Navier-Stokes solution, while the remaining curves refer to assorted variants of the LB method: KB refers to kinetic boundary conditions versus bounce-back (BB). The former allow for slip flow at the wall, while the latter does not. The prefix R denotes the Regularized version and finally 19 and 41 refer to the number of discrete velocities. As one can see, the R41KB model comes very close to DSMC, providing a concrete example of LB morphing into Boltzmann. The morphing is not perfect and it applies only to a global quantity, the mass flow rate, but still encouraging. From [15].

Such kind of behaviour cannot quantitatively reproduced by the Navier-Stokes equations, neither by the standard LB. However, Regularized LB with higher order lattices has proven capable of reproducing this prototypical non-equilibrium phenomena. In a nutshell, the regularisation consists in projecting out the higher order non-equilibrium moments after the streaming step, so as to minimise their effect on the lower order kinetic moments governing the transport property of the flow [14].

As one can appreciate, a suitable LB variant (R41KB, see caption) model comes very close to DSMC, providing a concrete example of LB morphing into Boltzmann. The morphing is not perfect and it applies only to a global quantity, the mass flow rate, but still encouraging. Further work is needed to assess whether the morphing reaches up to the details of the spatial structure of the velocity profile within the Knudsen layer, thereby relieving the need of the full multiscale LB/DSMC procedure described above

## 5 Future perspectives

Many fascinating challenges lie ahead of the chimera approach, all being aimed enhancing the degree of molecular individualism within the stream-collide lattice kinetic harness. Here we mention just a few relevant instances.

### 5.0.1 Lattice BBGKY

The idea is to develop a lattice version of the two-body Liouville equation for the joint distribution  $f(\vec{r}_1, \vec{r}_2, \vec{v}_1, \vec{v}_2; t)$ . This would permit to replace heuristic pseudo-potentials with atomistic potentials. The computational cost is steep: a very modest 32 space resolution demands of the order of  $20 \times 20^6 \sim 0.4$  trillion degrees of freedom! In

principle this can be attempted, at least as a proof of concept. The physical issue is how to concoct a realistic two-body local equilibrium in regions with strong inhomogeneities, say sharp interfaces, where the two-body correlation is neither homogeneous nor isotropic.

### 5.0.2 LB with internal degrees of freedom

Here the idea is to enrich LB with the statistical dynamics of internal degrees of freedom, say descriptors of the molecular structure (shape) such as angular variables. Each internal degree of freedom adds an extra-dimension to phase-space, and consequently economic discretizations are mandatory.

### 5.0.3 LB with strong fluctuations

Define a chimera distribution blending the Boltzmann distribution with particles (Klimontovich distribution),  $\Psi(\vec{r}, \vec{v}; t) = w f(\vec{r}, \vec{v}; t) + (1-w) \delta[\vec{r} - \vec{r}_p(t), \vec{v} - \vec{v}_p(t)]$ . The regular component carries the hydrodynamic content, while the singular one takes care of non-equilibrium fluctuations, so as to absorb strong fluctuations which would violate the positivity of the regular one. This is similar to standard LB/DSMC coupling, except that the weight  $w$  can be made a function of space and time, so that both regular ( $f$ ) and singular ( $\delta$ ) components are evolved concurrently. The advantage is that the particle component can be kept much more economical than stand-alone, since the low-order moments are in charge of the smooth part  $f$ .

## 6 Summary

Lattice Boltzmann is broad and flexible, it can navigate across many scales of motion with relatively minor enrichments, all of which can be cast in the powerful stream-collide paradigm. LB is not perfect, whenever the lattice spacing approaches relevant physical scales, close scrutiny of lattice artefacts is necessary and higher order lattices required. Nevertheless, it provides handy access to complex states of moving matter characterised by the coexistence of Universality and Molecular Individualism, a hallmark of soft matter at the Biology/Chemistry/Physics interface.

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