Mesoscale Modeling of Dynamic Wetting Phenomena

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The modeling of fluid flows using 'mesoscopic' methods is becoming increasingly popular as researchers attempt to bridge the gap between incomplete continuum-based models and resource-limited molecular dynamics simulations. Mesoscopic methods are particle methods, but rather than capturing the behavior of individual molecules, they are based on the dynamics of statistically representative 'pseudo-particles', which may be free or be constrained to move on a lattice. Examples of such methods include 'Smoothed particle hydrodynamics' (SPH) [1], 'Dissipative particle dynamics' (DPD) [2], 'direct simulation Monte Carlo' (DSMC) methods [3], and discretizations of the Boltzmann equation, for example the 'lattice Boltzmann method' (LBM) [4]. This paper considers only applications of the latter.

With roots in the kinetic theory of gases, the LBM is a special discretisation of the Boltzmann equation in which the molecular velocity distributions are coupled to a structured lattice. The pseudo-particles in this method are probability density distribution functions which, during each time step, stream along the links between lattice nodes and then interact with each other locally at each node through a collision model involving the relaxation towards an equilibrium probability distribution. The fact that the LB method is based on local interactions rather than global partial differential equations provides it with two major advantages. Firstly, there is no large matrix to invert so the method is well suited to parallel execution with excellent scalability. For large-scale simulations (say for a coating flow over several centimeters), parallel execution is in fact essential since the required lattice (i.e. mesh) densities are generally rather higher than those in a corresponding finite element (FE) simulation. The second advantage, arising for the same reason, is that topologically complex, multiply-connected domains are easily

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accommodated. An obvious application exploiting the latter feature is flow in porous media, or spreading flows over rough surfaces. A potential disadvantage of the LB method, however, is that it is inherently time-dependent. Thus, for the solution of steady-state flows, it may not be as efficient as a steady-state FE method. However, for coating flows, where the onset of instabilities and other time-dependent effects are of interest, steady-state simulations are of limited use anyway.

Note that, though the LBM can be derived from the Boltzmann equation, which is based on assumptions germane to rarefied gases, it can be shown via a multiple-scale spatio-temporal expansion procedure (the 'Chapman-Enskog' expansion) that solving the lattice Boltzmann equation in the limit of small Knudsen and Mach numbers is equivalent to solving the incompressible Navier-Stokes equations. Hence the LBM can be considered as an alternative (and in some cases more flexible) Navier-Stokes solver.

Several different multiphase/multicomponent lattice Boltzmann methods were developed in the 1990s [5-8], and this is a field still in active development, e.g. [9], as the limitations of existing models are addressed. In all the multiphase LBMs, liquid-fluid interfaces are diffuse, i.e. they have a non-zero thickness. This has advantages and disadvantages. On the positive side, such interface descriptions allow the straightforward and automatic simulation of flows involving interface break-up and coalescence, and they do not suffer from the mathematical singularities present in most *ad hoc* continuum-based wetting models. On the downside, the diffusivity of the interface introduces an additional length scale into the simulation. If the physical interface thickness is to be captured, the resulting constraint on the lattice cell size severely restricts the size of problem that can be tackled. Inevitably one has to accept that the simulated interface thickness will be (possibly several orders of magnitude) thicker than the actual one if simulations are to be feasible; the LBM is a mesoscopic method after all. In the context of wetting, a consequence of artificially thick interfaces is that LBM simulations tend to over-predict spreading rates, though there is no evidence that the predicted dynamics are qualitatively incorrect [10]. A further issue concerning diffuse interface methods is the appearance of spurious 'microcurrents' in the interface, i.e. rogue interfacial velocities which persist even when a supposedly steady state has been achieved. In general such currents are small, and can be reduced by careful selection of discretization schemes.

Multiphase LBMs have naturally been applied to wetting flows. In an early example, Blake *et al.* [11] showed that the velocity dependence of the dynamic contact angle predicted by a 'chromodynamic' LBM is consistent with the molecular kinetic theory. The 'pseudo-potential' model of Shan & Chen [6] has been a popular choice for wetting simulations, due to its convenience for dealing with fluid-solid attractions/repulsions [12]. Progress towards a thermodynamically consistent wetting model was made by Briant *et al.* [13] using the 'free-energy' LBM [7], where a wetting potential can be added to the total free energy. The approach is consistent with Young's equation at equilibrium. In this short space it is not possible to provide a comprehensive overview of LBM models of wetting, but it is hoped the above offers some pointers for interested readers. One of the appealing features of the LBM, as far as this author is concerned, is the potential to simulate the wetting of solid surfaces which are more realistic in nature, i.e. they may be rough, porous or chemically inhomogeneous. In the literature there are many examples of LBM studies of droplet dynamics on such surfaces, e.g. [14].

A further example is offered here, using the 'finite-density' LBM [8], which derives from Enskog's modification to the Boltzmann equation to account for the finite size of molecules and the consequent 'excluded volume' effect [15]. The underlying equation of state governing phase separation is the Carnahan-Starling equation, and wetting effects are included using the 'surface affinity' idea of Iwahara *et al.* [16], which is a normalized density defined as

$$\alpha_s = \frac{\rho - \overline{\rho}}{\rho_L - \overline{\rho}}, \quad \text{where } \overline{\rho} = \tfrac{1}{2}(\rho_L + \rho_G) \tag{1}$$

with ρ being fluid density, and ρ_L and ρ_G being the equilibrium densities of the liquid and gas phases respectively. It can be shown that the surface affinity is related to the static contact angle via Young's equation,

$$\cos\theta_s = \frac{\sigma_{SL} - \sigma_{SG}}{\sigma_{LG}} = \tfrac{1}{2}\alpha_s(3 - \alpha_s^2)\,. \tag{2}$$

Recasting equation (1) to give ρ in terms of α_s and using equation (2) provides a means of specifying θ_s by setting the density on the solid surface. This approach easily allows spatial variations in α_s and is therefore a simple means of mimicking chemical inhomogeneities on the

surface. By way of example, the motion of a droplet on an inclined plane is considered in figure 1. The surface is patterned with a sinusoidal variation in α_s:

$$\alpha_s = \overline{\alpha}_s + \Delta\alpha \sin(2\pi x / \Lambda) \tag{3}$$

where $\Delta\alpha$ and Λ are respectively the amplitude and wavelength of the variation, and $\overline{\alpha}_s$ is the average surface affinity. Of interest here is the effect of the patterning wavelength (expressed in lattice units) on the motion of the droplet on the incline. Figure 1 plots the position of the droplet versus time and shows that as Λ increases, eventually a value is found where the resulting surface non-uniformity is able to prevent the droplet from running down the incline. It is envisaged that the inclusion of real surface effects into simulations will be of great benefit to coating flows, where wetting is a forced rather than a natural process.

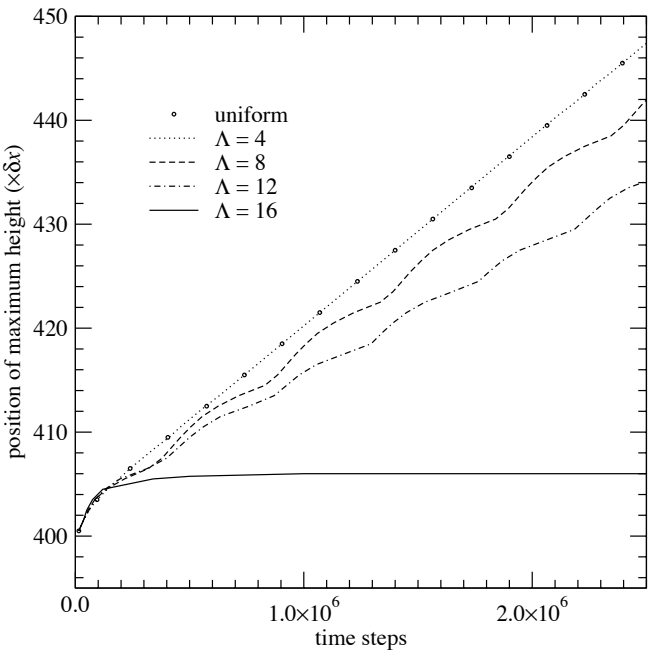

Figure 1: Position of a droplet on a non-uniform solid surface inclined at 5° to the horizontal. The parameter Λ is the wavelength of the non-uniformity in lattice units. For comparision, the interface thickness is roughly 4 lattice units.

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