

Lattice Boltzmann Simulation of Nonideal Fluids

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A lattice Boltzmann scheme able to model the hydrodynamics of phase separation and two-phase flow is described. Thermodynamic consistency is ensured by introducing a nonideal pressure tensor directly into the collision operator. We also show how an external chemical potential can be used to supplement standard boundary conditions in order to investigate the effect of wetting on phase separation and fluid flow in confined geometries. The approach has the additional advantage of reducing many of the unphysical discretization problems common to previous lattice Boltzmann methods.

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The hydrodynamics and kinetics of two-component fluids present a wealth of physical problems of both fundamental and technological importance [1]. There is much current interest in the relevance of hydrodynamics to spinodal decomposition [2] and the effects of substrates with different wetting properties on phase separation and domain growth [3]. In addition, the flow properties of multicomponent systems, particularly in porous media, have been intensively studied and are of great relevance to oil recovery [4].

Conventional methods for simulating two-phase flow include numerical integration of the Navier-Stokes equations and molecular dynamics simulations [5]. These techniques are extremely computationally intensive and particularly difficult to implement in random geometries. A newer approach, the lattice Boltzmann method, has recently proved competitive [6]. Here a set of distribution functions defined on a lattice is allowed to relax to equilibrium via a Boltzmann equation, discrete in both space and time. The correct choice of equilibrium distribution ensures that in the long wavelength limit the Navier-Stokes equations are recovered.

Several authors have set up lattice Boltzmann schemes for two-phase systems. In most approaches interface formation has been introduced phenomenologically by modifying the Boltzmann collision operator to impose phase separation [7]. Recent work by Shan and Chen [8] has attempted to relate phase separation to microscopic interactions by redefining the equilibrium velocity distribution so as to simulate a fluid with a nonideal equation of state. However, their approach leads to inconsistent thermodynamics unless a particular equation of state is chosen. In addition, all current schemes reach equilibrium distributions which have unphysical velocity fluctuations within the interfacial region [9].

In this Letter we show for the first time that it is possible to set up a lattice Boltzmann scheme modeling isothermal hydrodynamics for two-phase systems. This is achieved by introducing directly into the collision operator the equilibrium pressure tensor for a nonideal fluid. The resulting phase transition is pressure driven,

as pertinent to a liquid-vapor system quenched to well below the critical point [10]. The fluid reaches the correct thermodynamic equilibrium as determined by the equation of state and a Maxwell construction.

We first summarize the relevant results from the van der Waals formulation of quasiloccal thermodynamics for a two-component fluid in thermodynamic equilibrium at a fixed temperature [11]. The free energy functional is taken to be

$$\Psi(\mathbf{r}) = \int \left\{ \frac{\kappa}{2} |\nabla n(\mathbf{r})|^2 + \psi(n(\mathbf{r})) \right\} d\mathbf{r}, \quad (1)$$

where the first term gives the contribution from any density gradients and the second describes the bulk free energy density. The nonlocal pressure is defined by

$$p(\mathbf{r}) = n \frac{\delta \Psi}{\delta n} - \Psi(\mathbf{r}) = p_0 - \kappa n \nabla^2 n - \frac{\kappa}{2} |\nabla n|^2, \quad (2)$$

where $p_0 = n\psi'(n) - \psi(n)$ is the equation of state of the fluid. To obtain the full pressure tensor in a nonuniform fluid, nondiagonal terms must be added [12]:

$$P_{\alpha\beta}(\mathbf{r}) = p(\mathbf{r}) \delta_{\alpha\beta} + \kappa \frac{\partial n}{\partial x_\alpha} \frac{\partial n}{\partial x_\beta}. \quad (3)$$

In equilibrium the components of the pressure tensor define the surface tension in inhomogeneous regions of the fluid.

We next describe how a lattice Boltzmann simulation can be set up to ensure that these equilibrium conditions are met while incorporating the dynamical behavior pertinent to fluids, namely, order-parameter conservation and hydrodynamic transport. To illustrate our method, we choose to work in two dimensions on a triangular lattice, the simplest geometry that allows us to reproduce the Navier-Stokes equations. Let $f_i(\mathbf{x}, t)$ be a non-negative real number describing the distribution function of the fluid density at site \mathbf{x} at time t moving in direction \mathbf{e}_i , $i = 1, \dots, 6$. The unit vectors $\mathbf{e}_i = \{\cos[2\pi(i-1)/6], \sin[2\pi(i-1)/6]\}$ are the velocity vectors along the links of the lattice. With each site is

also associated a function $f_0(\mathbf{x}, t)$, which corresponds to the component of the distribution with zero velocity.

The distribution functions evolve according to a Boltzmann equation which is discrete in both space and time

$$f_i(\mathbf{x} + \mathbf{e}_i, t + 1) - f_i(\mathbf{x}, t) = \Omega_i(\mathbf{x}, t). \quad (4)$$

The most convenient choice for Ω_i is a single relaxation time form [13]

$$\Omega_i = -\frac{1}{\tau} (f_i - f_i^{\text{eq}}). \quad (5)$$

The density n and macroscopic velocity \mathbf{u} are defined by

$$n = \sum_i f_i, \quad (6)$$

$$nu_\alpha = \sum_i f_i e_{i\alpha}, \quad (7)$$

and the equilibrium distribution f_i^{eq} is chosen so as to reproduce the correct dynamic equations for n and \mathbf{u} .

For a one-component fluid with an ideal gas equation of state, f_i^{eq} is expanded as a power series in the local velocity and the coefficients determined by local conservation of mass and momentum and by the constraints of Galilean invariance and isotropy of the pressure tensor. In order to include the correct nonlocal thermodynamic properties of a nonideal fluid, additional nonlocal terms are needed in the expansion for f_i^{eq} . We thus define

$$f_i^{\text{eq}} = A + B e_{i\alpha} u_\alpha + C u^2 + D u_\alpha u_\beta e_{i\alpha} e_{i\beta} + F_\alpha e_{i\alpha} + G_{\alpha\beta} e_{i\alpha} e_{i\beta}, \quad (8)$$

$$f_0^{\text{eq}} = A_0 + C_0 u^2, \quad (9)$$

where the coefficients, now functions of n and its derivatives, can be determined by three macroscopic constraints.

The first two are, as for the noninteracting case, local conservation of mass and momentum

$$\sum_i f_i^{\text{eq}} = n, \quad (10)$$

$$\sum_i f_i^{\text{eq}} e_{i\alpha} = nu_\alpha. \quad (11)$$

The third constraint is that the pressure tensor takes the form

$$\sum_i f_i^{\text{eq}} e_{i\alpha} e_{i\beta} = P_{\alpha\beta} + nu_\alpha u_\beta. \quad (12)$$

The constraints (10), (11), and (12), together with the equilibrium thermodynamic definitions (1)–(3), are sufficient to determine the coefficients in the expansions

(8) and (9)

$$A_0 = n - 2(p_0 - \kappa n \nabla^2 n),$$

$$A = (p_0 - \kappa n \nabla^2 n)/3,$$

$$B = n/3, \quad C_0 = -n,$$

$$C = -n/6, \quad D = 2n/3,$$

$$F_\alpha = 0,$$

$$G_{xx} = -G_{yy} = \frac{\kappa}{3} \left[\left(\frac{\partial n}{\partial x} \right)^2 - \left(\frac{\partial n}{\partial y} \right)^2 \right],$$

$$G_{xy} = 2 \frac{\kappa}{3} \frac{\partial n}{\partial x} \frac{\partial n}{\partial y}, \quad (13)$$

where, on the lattice, derivatives are simply expressed by finite difference approximations.

The continuum hydrodynamic equations modeled by this dynamic scheme can be determined by performing a Chapman-Enskog expansion on the Boltzmann equation (4). To second order, the usual continuity and Navier-Stokes equations result

$$\frac{\partial n}{\partial t} + \frac{\partial (nu_\alpha)}{\partial x_\alpha} = 0, \quad (14)$$

$$\frac{\partial nu_\alpha}{\partial t} + \frac{\partial nu_\beta u_\alpha}{\partial x_\beta} = -\frac{\partial p_0}{\partial x_\alpha} + \nu \nabla^2 nu_\alpha + \frac{\partial}{\partial x_\alpha} [\lambda(n) \nabla \cdot n\mathbf{u}], \quad (15)$$

where $\nu = (2\tau - 1)/8$ and

$$\lambda(n) = \left(\tau - \frac{1}{2} \right) \left(\frac{1}{2} - \frac{\partial p_0}{\partial n} \right). \quad (16)$$

Note that to this order, the only difference between Eq. (15) and the Navier-Stokes equation for an ideal fluid is the appearance of the nonideal pressure p_0 .

To test the correctness and applicability of the approach described above we performed simulations on a van der Waals fluid for which

$$\psi = nT \ln \left(\frac{n}{1 - nb} \right) - an^2. \quad (17)$$

We first consider the equilibrium configuration for a system with periodic boundary conditions and zero net flow velocity. The inset of Fig. 1 shows the coexistence curve as a function of temperature T , calculated from Eq. (2) using a Maxwell construction. The points show simulation data obtained from lattices of size 256×256 , equilibrated for 10 000 time steps. Because the correct equilibrium thermodynamics is inherent in the model, the bulk phases reached in the simulations obey the Maxwell construction. Note the wide range of coexisting densities that can be reached by the simulation before significant finite difference errors come into play as the temperature is lowered.

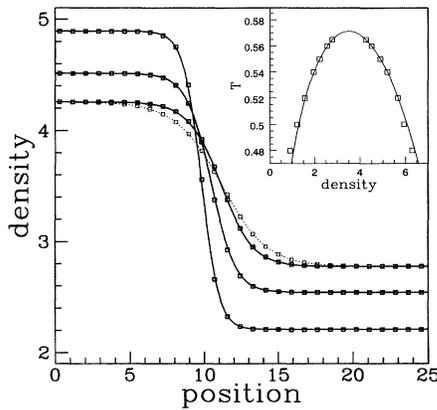


FIG. 1. Equilibrium density profiles normal to a flat interface for a van der Waals fluid for the three highest values of T shown on the coexistence curve (inset). The solid lines are numerical solutions of the continuum thermodynamic equations, while the points are from the lattice Boltzmann simulations. The parameter values are $a = \frac{9}{49}$ and $b = \frac{2}{21}$, while $\kappa = 0.01$ for the bold curves and 0.02 for the dashed curve.

Figure 1 also shows the equilibrium interface density profiles which are seen to depend, as expected, on κ and the temperature. Again the agreement with a direct integration of the continuum thermodynamic equations, derived by minimizing Eq. (1), is excellent. The interfacial width can be varied, typically between ~ 2 and 30 lattice sites. This ensures that lattice anisotropy effects can be made unimportant. Our mechanism for interface formation also reduces the magnitude of the microscopic velocities in the interfacial region. For example, we find a reduction in the velocity by a factor of $\sim 10^3$ [14] in comparison with the liquid-vapor model of Shan and Chen [8]. This is because our scheme conserves momentum locally and, thus, there are no spurious corrections to the continuity equation, Eq. (14). Furthermore, this eliminates the need to redefine the fluid velocity in the interfacial region [15].

In Fig. 2 we emphasize the consistency between the mechanical definition of surface tension

$$\sigma = R\Delta P, \tag{18}$$

where ΔP is the pressure difference between the inside and outside of a spherical domain of radius R , and the thermodynamic definition

$$\sigma = \kappa \int \left(\frac{\partial n}{\partial z} \right)^2 dz \tag{19}$$

for a flat interface. Agreement is seen as $R \rightarrow \infty$ with the expected curvature correction to Eq. (18) appearing for $R \lesssim 10$ lattice units.

Far from the interface, the fluid obeys the usual Navier-Stokes equation common to other lattice Boltzmann schemes. However, the dynamical behavior of the interface itself is of importance if domain growth is to be

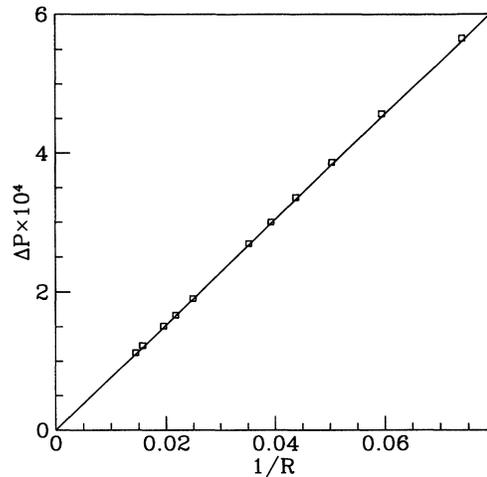


FIG. 2. ΔP plotted vs $1/R$ as a test of Laplace's law. The solid line has a gradient calculated using Eq. (19).

correctly described by the model. In Fig. 3 the dispersion relation for capillary waves is displayed [16] giving a best fit of $\omega \sim k^{1.6}$. We attribute the slight discrepancy from the expected dispersion relation $\omega^2 \sim k^3$ to curvature corrections to Laplace's law. These results were obtained by imposing a sine wave of a given wave vector on an interface that had reached equilibrium in a 128×128 system and observing the period of the subsequent oscillations for, typically, 500 time steps.

Finally, we demonstrate how the addition of an external chemical potential at the surfaces of a confined system can be used to supplement the usual bounce-back boundary conditions, allowing us to change the substrate properties and hence study wetting. Gradients in the chemical potential $\mu_{ex}(\mathbf{r})$ act as a thermodynamic force on the

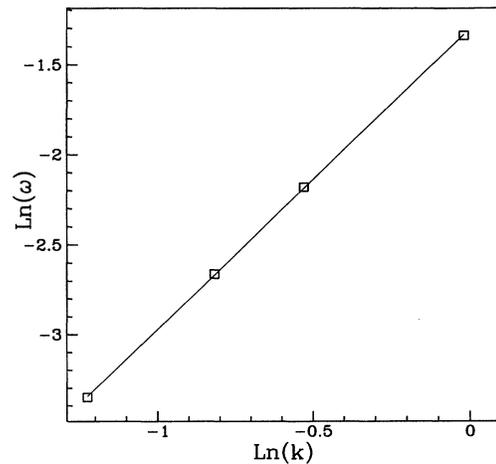


FIG. 3. The dispersion curve for capillary waves on an interface, plotted on a log-log scale. The best fit line has gradient 1.6 ± 0.05 .

fluid and can be included within the lattice Boltzmann framework by modifying Eq. (11)

$$\sum_i f_i^{\text{eq}} = nu_\alpha - \tau n \frac{\partial \mu_{\text{ex}}}{\partial x_\alpha}, \quad (20)$$

which, in turn, introduces a force into the collision operator, Eq. (8),

$$F_\alpha = -\frac{\tau n}{3} \frac{\partial \mu_{\text{ex}}}{\partial x_\alpha}. \quad (21)$$

As an illustrative example, by letting $\mu_{\text{ex}}(\mathbf{r})$ differ from zero only at the boundary sites, the affinity of the boundaries for each of the phases can be tuned in a simple, physically appealing way. Results in Fig. 4 show how the fluid configuration develops in a one-dimensional pore for a situation when the black, dense, fluid (a) wets and (b) does not wet the walls. By modifying the functional form of $\mu_{\text{ex}}(\mathbf{r})$, for example, by introducing long-range interactions, equilibrium phases comprised of plugs, tubes, and capsules discussed by Liu *et al.* [17] can be reached. In addition, an important consequence of this formalism is that the fluid-boundary interface is diffuse. This allows for a reduction of many of the problems associated with surface orientation and standard bounce-back boundary conditions [18].

To summarize, we have described a lattice Boltzmann scheme, the main new features of which are the direct introduction of a nonideal pressure tensor and an external chemical potential. This enables us to obtain an isothermal model of phases separation which correctly describes bulk and interfacial dynamics at low temperatures. The method also provides a convenient, physically motivated way of tuning boundary conditions, giving a new approach to situations when flow and phase separation are affected by fluid-substrate interactions. Moreover, unphysical velocity oscillations at surfaces and interfaces are substantially reduced.

The simplicity of the method and the ease of implementation suggests that our approach may be a valuable

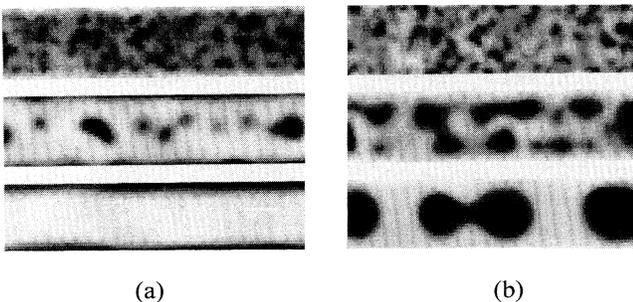


FIG. 4. Time evolution (vertically downwards) of phase separation in a narrow capillary. In (a) the dense (dark) fluid wets the surfaces while in (b) nonwetting is illustrated. The simulations were performed on a 128×32 lattice at $T = 0.56$ with (a) $\mu_{\text{ex}} = -0.1$ and (b) $\mu_{\text{ex}} = 0.5$.

tool in the study of multiphase hydrodynamical systems. Furthermore, the introduction of a temperature parameter in our mechanism for interface formation should allow the formalism to be extended to nonisothermal situations where heat transfer is important [19]. We are currently in the process of investigating such a model.

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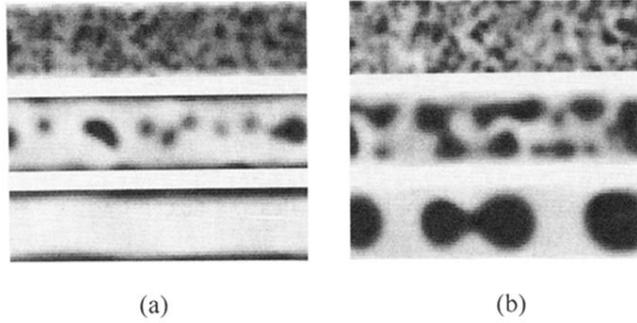


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