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Interface width and bulk stability: Requirements for the simulation of deeply quenched liquid-gas systems

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Simulations of liquid-gas systems with interface terms evaluated by central difference discretizations are observed to fail to give accurate results for two reasons: the interface can get "stuck" on the lattice or a density overshoot develops around the interface. In the first case, the bulk densities can take a range of values, dependent on the initial conditions. In the second case, inaccurate bulk densities are found. We derived the minimum interface width required for the accurate simulation of liquid-gas systems with a diffuse interface. This criterion is demonstrated for lattice Boltzmann simulations of a van der Waals gas. Combining this criterion with predictions for the bulk stability defines the parameter range for stable and accurate simulation results even for high density ratios of over 1000.

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Application of lattice Boltzmann (LB) methods to liquid-gas systems was one of their early successes. Three very different algorithms were developed to do this: Swift *et al.* developed an algorithm based on implementing a pressure tensor [1], Shan and co-workers developed an algorithm based on mimicking microscopic interactions [2], and Gunstensen *et al.* developed an algorithm specifically for strong phase separation [3]. These algorithms have been successfully applied to simulations of phase separation [4], drop collisions [5,6], wetting dynamics and spreading [7], and the study of dynamic contact angles [8,9]. Only recently has it been shown that including higher-order corrections into the Shan *et al.* approach makes it thermodynamically consistent, i.e., stationary states have a constant pressure [10].

Currently only heuristic predictions exist for parameter ranges leading to accurate simulation results. Close to the critical point, thin interfaces can lead to nonunique interface profiles [10]. Further from the critical point, density overshooting is observed at the interfaces, in combination with bulk densities inconsistent with thermodynamic theory [10]. Below, we present a criterion to predict the range of acceptable values for the interface width for algorithms that have a constant pressure in the steady state. The accuracy of the algorithms rapidly deteriorates when this limit is exceeded. The second, LB-specific, contribution is a more general definition of the equation of state. The usual LB methods recover the ideal gas equation of state with a pressure of $p=\rho/3$ when the gas is dilute. While relaxing this requirement has no effect on the interfacial properties, it allows us to adjust the bulk stability of the LB method. Taken together, this determines the parameters for which deep quenches can be accurately simulated. This is an important result since LB methods for van der Waals gases were previously believed to be limited to density ranges of about 20. This led to the development of hybrid methods [5], the examination of different equations of state [11], as well as the early work by Gunstensen *et al.* [3], which allows only strong separation.

The key to a successful simulation of liquid-gas systems is the faithful representation of the interface [10]. In particular, we need to obtain a constant pressure across a flat interface. For a standard Landau free energy of $F=\int \psi_0+(\kappa/2)\times(\nabla\rho)^2$, where ρ is the density, we have

$$P_{\alpha\beta}=p_0\left\{\left[p(\rho)-\kappa\left(\rho\nabla^2\rho+\frac{1}{2}\boldsymbol{\nabla}\rho\cdot\boldsymbol{\nabla}\rho\right)\right]\delta_{\alpha\beta}+\kappa\nabla_\alpha\rho\nabla_\beta\rho\right\}, \tag{1}$$

where $p_{(\rho)}=\rho\partial_\rho\psi_0-\psi_0$ is the bulk pressure. Note that we introduced a scale factor p_0 with the pressure. Such a scale factor clearly is not expected to change the equilibrium behavior. However, as we will see below, it can have a profound effect on the bulk stability of the system. Most previous approaches [2,12] correspond to a choice of $p_0=1$ in Eq. (5). Only for this choice will the lattice Boltzmann method recover the standard LB method for ideal gases in the limit of small densities. In equilibrium the normal pressure is constant across an interface. For the equilibrium profile $\rho(x)$ corresponding to a bulk pressure of p_B, this implies

$$\kappa=\frac{p(\rho)-p_B}{\rho\partial_x^2\rho-\frac{1}{2}\partial_x\rho\partial_x\rho}. \tag{2}$$

For simulations, the density derivatives are replaced by discrete derivatives. For simplicity, we will limit our analysis to the one-dimensional case here. We consider discrete derivatives of central difference form $\partial_x\rho=0.5[\rho(x+1)-\rho(x-1)]$ and $\partial_x^2\rho=\rho(x+1)+\rho(x-1)-2\rho(x)$. As shown below, this puts severe limits on the allowable values for κ.

We now estimate the minimum value κ_m. For any point on the interface with density ρ_s, we consider two neighboring points, one with a smaller density ρ_- and one with a larger density ρ_+. If ρ_l is the liquid density and ρ_g is the gas density, we deduce a lower limit for the smallest possible value $\kappa_m(\rho_s)$ by varying the values of ρ_+ and ρ_-, but not allowing overshooting:

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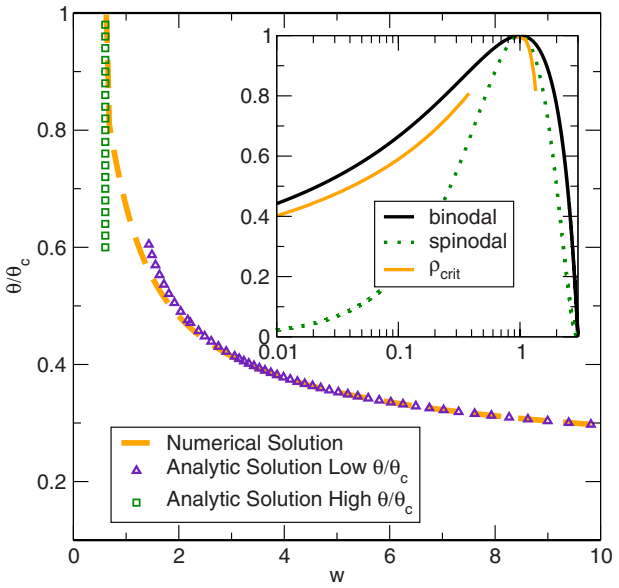

FIG. 1. (Color online) The two limiting cases for which we can obtain an analytical approximation to the $w(\kappa_m)$ relation. The inset shows the value of the critical density ρ_{crit}, which is the value ρ_s takes when (3) is maximized. Note that there is a discontinuity.

$$\kappa_m(\rho_s) = \min_{\substack{\rho_g<\rho_-<\rho_s \\ \rho_s<\rho_+<\rho_l}} \frac{p(\rho)-p_b}{\rho_s(\rho_- - 2\rho_s + \rho_+) - (\rho_+ - \rho_-)^2/8}. \quad (3)$$

Since we need to allow any value of ρ_s between ρ_g and ρ_l, the minimum allowable value of κ is then given by

$$\kappa_m = \max_{\rho_g<\rho_s<\rho_l} \kappa_m(\rho_s). \quad (4)$$

As an example we use a van der Waals gas with

$$p(\rho) = \left(\frac{\rho}{3-\rho} - \frac{9}{8}\rho^2\theta_c\right). \quad (5)$$

A good approximation for the equilibrium interface shape that becomes exact close to the critical point [where Eq. (8) holds] is given by

$$\rho^{init}(x) = \rho_g + \frac{\rho_l - \rho_g}{2}\left[1 + \tanh\left(\frac{x}{w(\kappa, \theta/\theta_c)}\right)\right], \quad (6)$$

where ρ_l and ρ_g are the equilibrium gas and liquid densities and $\theta=1/3$. The interface width is given by

$$w(\kappa, \theta/\theta_c) = \sqrt{\frac{2\kappa}{\theta_c/\theta - 1}}. \quad (7)$$

This profile is not the exact analytical solution to the differential equation $\nabla P=0$, but it is very close to it.

We first examine numerically which values of ρ_s in Eq. (3) lead to the most restrictive constraint, i.e., the largest value of $\kappa_m(\rho_s)$. The orange (gray) line in the inset of Fig. 1 shows how this density ρ_{crit} varies as a function of temperature. Near to the critical temperature, the orange (gray) line in the inset in Fig. 1 lies close to the high-density spinodal curve. This is because $P-p_b$ has its highest magnitude here, therefore helping to maximize κ_m within this region. An analytical estimate for κ_m can be obtained by expanding the pressure around the critical density, giving

$$p - p_b = -\frac{9}{4}(\theta_c - \theta)(\rho_s - 1) + \frac{3}{16}(\rho_s - 1)^3. \quad (8)$$

Within this regime, $p-p_b$ is large and negative, and therefore ρ_- and ρ_+ must be chosen to make the denominator in (3) as negative as possible. A suitable choice is $\rho_- = \rho_g$ and $\rho_+ = \rho_s$. We assume that the critical value of ρ_s lies on the spinodal curve $\rho_{spin}=1+2\sqrt{\theta_c-\theta}$. This allows us to obtain κ_m, and substituting this expression into (7) gives a minimum interface width of

$$w_{min} = \frac{1}{\sqrt{1+\sqrt{3}}}. \quad (9)$$

As the temperature is decreased in the inset of Fig. 1, the critical density ρ_{crit} makes a discontinuous jump to a regime in which it lies close to the gas density ρ_g. The minimum interface width can, in this case, be analytically obtained by expanding densities around ρ_g. We define $\rho_s=\rho_g+\delta\rho$ and $\rho_+=\rho_-+\Delta\rho$. Since $p-p_b$ is a positive quantity, a suitable choice for ρ_- is $\rho_-=\rho_s$. Substituting these expressions into Eq. (3) gives

$$\kappa_m = \max_{\delta\rho>0}\min_{\Delta\rho>0} \frac{\theta\delta\rho}{(\rho_g+\delta\rho)\Delta\rho - \Delta\rho^2/8}. \quad (10)$$

Minimizing this with respect to $\Delta\rho$ leads to $\rho_s=\Delta\rho/4$. Resubstituting this result back into Eq. (10) and maximizing with respect to $\delta\rho$, we finally obtain $\rho_{crit}=2\rho_g$. Using this we can calculate the minimum interface width,

$$w_{min} = \frac{1}{\sqrt{4\rho_g(\theta_c - \theta)}}, \quad (11)$$

as shown by the triangles in Fig. 1. This closely follows the numerical result at low temperatures. This means that we need wide interfaces for deep quenches because of the unfortunate cancellation of the discrete derivative and Laplace operator for low densities in the denominator of (3).

Most previous LB simulations approached the simulation of nonideal systems by using the ideal gas equation of state $p=\rho\theta=\rho/3$, as a starting point. Interactions are then included to allow the simulation of nonideal systems. The speed of sound $c_s=\sqrt{\partial_\rho p}$ will then recover the ideal gas value of $1/\sqrt{3}$ in the dilute limit. For a van der Waals gas with a critical density of 1, a temperature of $\theta=1/3$, and an interfacial free energy of $\int(\kappa/2)(\nabla\rho)^2$, the pressure tensor used by previous approaches matched the ideal gas equation of state in the dilute limit, leading to $p_0=1$. For the van der Waals gas the speed of sound increases rapidly for high densities. A problem arises when the speed of sound becomes larger then the lattice velocity $|v_i|$, because information cannot be passed on at speeds larger than the lattice velocity. When the speed of sound is increased above 1 the simulation becomes unstable. This problem is exacerbated by the presence of the gradient terms in the pressure tensor. These terms further decrease the stability, as shown in a previous analysis of the pressure

method by Pooley for one-, two-, and three-dimensional LB methods [13]. In the notation of this Rapid Communication the linear stability condition is

$$c_s < \sqrt{1 - 4p_0\kappa\rho} \tag{12}$$

for a homogeneous, one-dimensional system with density ρ. This implies a restriction for both the maximum quench depth and the maximum interface width. This is shown in Fig. 3 below as solid lines for different values of p_0. This suggests that, at least as far as the stability of the bulk phase is concerned, the most stable solutions should be found for $\kappa=0$.

For simplicity we demonstrate the constraints of liquid-gas lattice Boltzmann simulations by the common one-dimensional projection of one-, two-, and three-dimensional models commonly known as the D1Q3 model, i.e., a one dimensional model with a velocity set $v_i=\{-1,0,1\}$. The lattice Boltzmann equation for densities f_i corresponding to velocity v_i is given by

$$f_i(\mathbf{x}+\mathbf{v}_i,t+1) = f_i(\mathbf{x},t) + \frac{1}{\tau}[f_i^0(\mathbf{x},t) - f_i(\mathbf{x},t)]. \tag{13}$$

The f_i^0 are the equilibrium distributions and are given by

$$f_{-1} = -\rho u/2 + \Pi/2, \quad f_0 = \rho - \Pi, \quad f_1 = \rho u/2 + \Pi/2,$$

and $\Pi=\rho u^2+P+\nu u\partial_x\rho$ [1,10] where $\nu=(\tau-0.5)/3$. To second order the resulting equations of motion are, as usual, the continuity equation

$$\partial_t\rho + \partial_x(\rho u) = 0 \tag{14}$$

and the Navier-Stokes equation

$$\partial_t(\rho u) + \partial_x(\rho u^2) = -\partial_x P + \partial_x(2\nu\rho\partial_x u). \tag{15}$$

To lower the speed of sound in the liquid phase, we now reduce the value of p_0 in (5). This decreases the speed of sound in the liquid by a factor of $\sqrt{p_0}$. This also increases the range of stability for κ in (12). We now expect that lowering the speed of sound by a large enough factor will reduce the speed of sound sufficiently to simulate systems with arbitrarily low temperature ratios θ/θ_c.

To test this idea we performed simulations with near-equilibrium profiles by defining an initial density profile that is given by two domains with densities ρ_l and ρ_g, respectively, connected by a near equilibrium interface given by Eq. (6). By initializing the simulation with this profile we test the linear stability of the method around a near equilibrium profile to good accuracy. Note that the shape of a stable interfacial profile is independent of p_0.

In Fig. 2 we see that, by lowering p_0, the method is now able to simulate very small values of the reduced temperature θ/θ_c for interface width $w>1$, but that significantly larger widths are required to recover an accurate phase diagram for deep quenches. For values of θ/θ_c between 0.9 and 1, we also find nonunique solutions for small values of κ, as is discussed in more detail in a previous paper [10].

We now test the predictions of the accuracy of extended interface liquid-gas simulations using the LB implementation presented in [10].

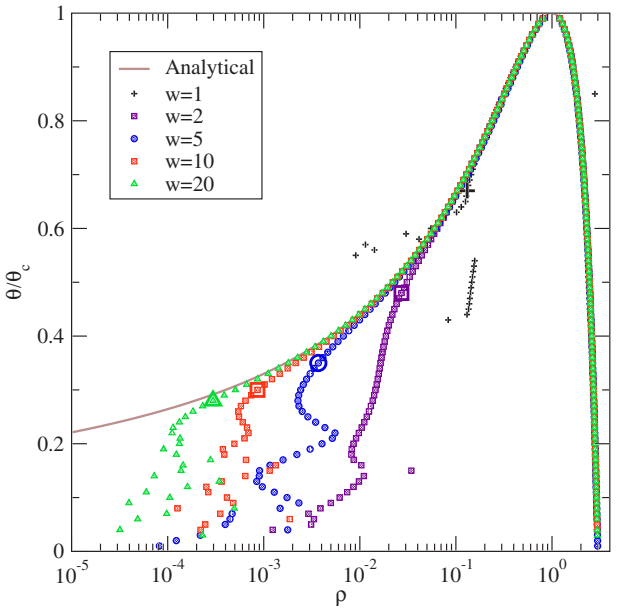

FIG. 2. (Color online) The van der Waals phase diagram is recovered to very good approximation for interfaces wider than the minimum width. The large symbols represent the points at which the simulations are predicted to become inaccurate by Eq. (3). The value of p_0 affects only the bulk stability and values from 1 to 10^{-7} were used for increasing quench depth.

We performed a scan of the parameter space w and θ/θ_c initializing the simulation with a near equilibrium profile for different values of p_0. We accept simulations that are stable, accurate and unique. The criterion of accuracy is defined to be $\log_{10}(\rho_{min})-\log_{10}(\rho_g)<0.1$. As can be seen in Fig. 2, the

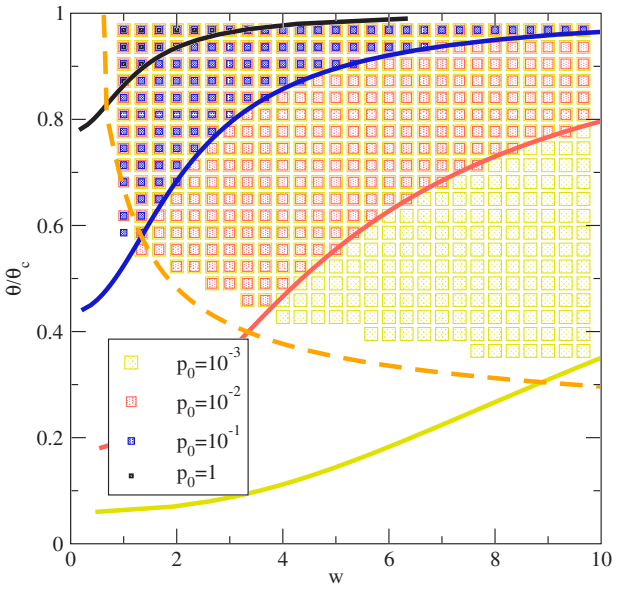

FIG. 3. (Color online) Existence of accurate solutions for different values of p_0 and w. Symbols indicate parameter combinations that lead to stable, accurate, and unique solutions. Solid lines are the bulk stability limits for the pressure method given by Eq. (12). The dashed line is the line for an accurate interface representation given by Eq. (3).

results are not very sensitive to the exact value of the cutoff. For values of the interface width $w<1.5$, we also test the uniqueness of the simulation by using initial profiles with bulk densities corresponding to the pressure at the spinodal points [10]. Our criterion for uniqueness is that all simulations lead to the same minimum density to within $\Delta\rho<0.01$.

Comparing (3), shown as a dashed line in Fig. 3, and the numerical results for stable, accurate, and unique solutions shows excellent agreement. The bulk stability of Eq. (12) gives the second limit for the acceptable parameter range for the pressure method. We performed a similar analysis for the forcing method of [10] and obtained nearly identical results except for a slightly ($\sim 10\%$) larger range of bulk stability. Multirelaxation time (MRT) LB methods recover the Bhatnagar-Gross-Krook form for equal relaxation times. We use $\tau=1$ in this paper and MRT methods will therefore have identical stability properties for this (natural) choice. Note that previous LB simulations use $p_0=1$, which corresponds to the area under the black line in Fig. 3. This is why it was assumed that standard LB simulations of van der Waals gases are limited to a maximum density ratio of about 10 [5].

The interface constraint (3) is remarkably successful at predicting the acceptable simulation parameters. It predicts how thin is too thin for an interface. It thereby detects when nonunique solutions occur and when solutions for deep quenches fail to deliver accurate results.

For simulation methods it is important to be aware of the acceptable parameter ranges. Lattice Boltzmann simulations are often believed to have the nice property of becoming unstable before they become inaccurate [14]. This is not the case for thin interfaces in multiphase simulations. In this case the simulation can remain stable and become inaccurate. This makes it necessary to find some criterion that determines whether a set of parameters will lead to an accurate simulation. Such a criterion was presented for the interface width (controlled by κ) in this Rapid Communication.

The second contribution presented in this paper appears trivial at first: it consists of a simple prefactor for the pressure. This breaks with the idea that the standard LB method for ideal gases should always be recovered in the dilute limit. This prefactor has profound implications for the stability of the bulk phases, which can be seen using an important result about the bulk stability from [13].

Combining these two components, we were able to show that the lattice Boltzmann method is indeed able to simulate very deep quenches for liquid-gas cases. This analysis was general and will be applied to other equations of state [11] as well as other discretizations of the interfacial terms. One important approach here is to use a forcing term that utilizes $\nabla P=\rho\nabla\mu$ [12], which has been found to allow the simulation of higher density ratios. Combining this may yet yield significant further advances for the development of multiphase LB methods.

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