Phase-field simulations for drops and bubbles

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Recently we proposed a phase field model to describe Marangoni convection in a compressible fluid of van der Waals type far from criticality [Eur. Phys. J. B **44**, 101 (2005)]. The model previously developed for a two-layer geometry is now extended to drops and bubbles. A randomly distributed initial density evolves towards phase separation and single droplet formation. For a two-component liquid-liquid system we report on numerical simulations for drop Marangoni migration in a vertical thermal gradient.

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Drops and bubbles are encountered in our everyday life. They appear in many industrial situations involving material processing, manufacturing, boiling, storage, and management of liquids. The word drop represents an object which contains liquid and the word bubble is used to designate an object which contains gas or vapor. In experiments carried out in reduced gravity the gravitational force acting on particles, drops, or bubbles becomes very small or completely vanishes. This is the motivation for studying the motion of these objects due to forces other than gravity. One can imagine few mechanisms independent of gravity that will cause the motion of a droplet (drop or bubble). For example, an electric or magnetic field can be used to move an object. The most common mechanism that has been used is, however, the application of a temperature gradient, because it is easy to produce temperature variations in a fluid. Such gradients also occur naturally in many material processing applications, because of the use of heating or cooling as an integral part of the process. A droplet placed in a temperature gradient tends to move towards the hotter wall, "attracted" by hot objects. This is the motion of the droplet relative to the shearing Marangoni flow induced along its surface by surface tension gradients. This phenomenon is called thermocapillary migration or Marangoni migration and has been experimentally discovered by Young *et al.* in 1959 [1]. It has been widely theoretically investigated, using classical models, for quasistationary states of incompressible spherical droplets (see Refs. [2–6], and references therein). The classical models consider the hydrodynamic basis equations (energy and Navier-Stokes equations) for each phase, shear stress balance (Marangoni condition) and continuity of the heat fluxes along the droplet interface.

The purpose of this paper is to examine the same phenomenon using a phase field model. Here, one introduces an additional field to the usual set of state variables to distinguish between the different phases. With the help of this phase field all the system parameters can be expressed as functions varying continuously from one medium to the other. Therefore, the problem is treated like an entire one phase problem and the interface conditions will be substituted by some extra terms in the Navier-Stokes equation. Because they reduce the system of equations (they don't need different equations for each medium) and eliminate the explicit interface conditions, the phase field models are suitable for problems with complex geometries and are very attractive in view of their numerical simplicity.

The present work extends the phase field model previously elaborated for describing Marangoni convection in two-layer systems [7–11] to drops and bubbles and presents some phase field simulations for Marangoni migration in two-component fluids. We first study an isothermal system without gravity—a liquid in equilibrium with its own vapor—a situation for which the most natural phase field variable is the density ρ, scaled by the liquid density. So $\rho=1$ designates the liquid phase and $\rho\approx 0$ the vapor bulk. For a two-phase system with diffuse interface and without evaporation phenomena the Helmoltz free-energy functional is given by [12,13]

$$\mathcal{F}[\rho]=\int_V \left[f(\rho)+\frac{\mathcal{K}(\vec{\nabla}\rho)^2}{2}\right] dV, \tag{1}$$

where the first term in Eq. (1) represents the free-energy density for the homogeneous phases and the second term is associated with variations of density and contributes to free-energy excess of the interface

$$\gamma=\int_{-\infty}^{+\infty} \mathcal{K}(\vec{\nabla}\rho)^2 dz.$$

As already shown in Refs. [7,8], minimizing the free-energy functional (1) one can derive the nonclassical phase field terms which has to be included in the Navier-Stokes equation for assuring the shear stress balance at the droplet interface

$$\rho\frac{d\vec{v}}{dt}=-\vec{\nabla}p+\rho\vec{\nabla}[\vec{\nabla}\cdot(\mathcal{K}\vec{\nabla}\rho)]+\vec{\nabla}\cdot(\eta\vec{\nabla}\vec{v})+\vec{\nabla}(\lambda\vec{\nabla}\cdot\vec{v})+\rho\vec{g},\quad \lambda\approx\frac{\eta}{3}. \tag{2}$$

The fluid density ρ obeys the continuity equation

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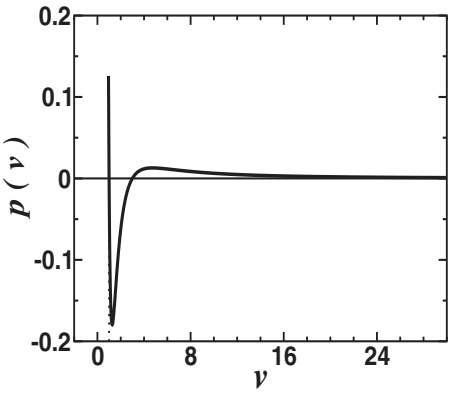

FIG. 1. The pressure corresponding to the free-energy density (5) as function of $v=1/\rho$. The unit volumes for liquid and vapor phase are $v_l=1$ and $v_v=\infty$, respectively.

$$\frac{\partial \rho}{\partial t} + \vec{\nabla} \cdot (\rho \vec{v}) = 0, \tag{3}$$

so that the mass conservation is fulfilled. For a system in equilibrium and without interfacial mass exchange the free-energy density has to be a symmetrical double-well potential with two minima corresponding to the two alternative phases: $\rho=1$ for the liquid and $\rho=0$ for the vapor state. We choose the free-energy density given by

$$f(\rho) = \frac{C}{2}\rho^2(\rho-1)^2. \tag{4}$$

If one represents the thermodynamical pressure $p(\rho)=\rho\frac{\partial f}{\partial \rho} - f(\rho)$ against the unit volume $1/\rho$ for the free-energy density (4) one observes a curve of van der Waals type (see Fig. 1). With Eq. (4), the Navier-Stokes equation (2) admits an analytical solution for the stationary state

$$\rho_0(z) = 1 \Big/ \left[1 + \exp\frac{(z-1)}{l}\right], \quad l \propto \sqrt{\mathcal{K}/C}.$$

The above parameter l describes the thickness of the interface. For small enough values of l this solution describes two superposed liquid-vapor layers with the liquid boundary at $z=0$, the vapor boundary at $z=2$, and the diffuse interface around $z=1$. Thermocapillary convection in two planar layers vertically heated was investigated in Refs. [9,11] in the frame of the phase field model. A linear stability analysis and a comparison with the classical models were done in Ref. [9]. The fully nonlinear evolution for the same problem with evaporation was described in Ref. [11].

Now we wish to use the Eqs. (2) and (3) to treat drops and bubbles. For this new geometry one has no analytical solution for the stationary state and no linear stability analysis can be done. Instead, one can solve the problem numerically starting from an initial noise density

$$\rho_{\text{initial}}(x,z) = C_m \xi(x,z), \tag{5}$$

where C_m is the noise intensity and ξ is a random distribution between 0 and 1. The constant C_m controls the total mass of the system. Depending on its value, the asymptotically stable

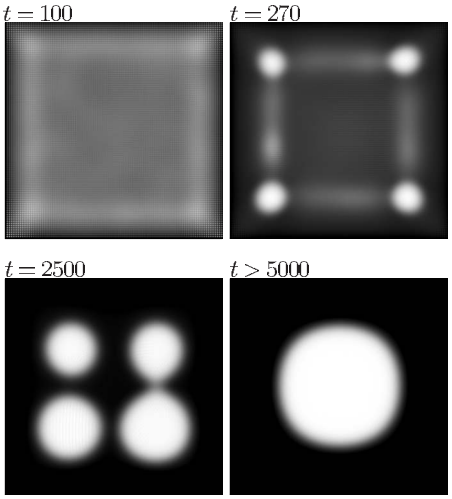

FIG. 2. Time series in (x,z) plane for the formation of a liquid drop in a vapor atmosphere for a system without gravity and without external heating ($\bar{\rho}=0.27$).

state of lowest free energy corresponds to a single liquid drop in a vapor atmosphere (small C_m) or a gas bubble in a liquid (large C_m). The noise character of Eq. (5) may act as seeds for phase separation in the unstable or metastable regime of Fig. 1. In the latter, drops or bubbles are found by nucleation and need a finite initial disturbance. In both cases, the dynamical process is dominated by coarsening and relaxes towards one of the "fixed points" described above.

The material parameters η, λ, κ and c are considered linearly coupled to the density. For numerical simulations we developed a numerical code in two spatial dimensions based on a finite difference method with 200×200 mesh points for water-vapor parameters [14]. The interface is about 3% from the length of the box, that means a resolution around 7 points in the diffuse interface. No-slip conditions for the velocity field were imposed at the wall boundaries ($\vec{v}=0$).

For low total mass small liquid drops are coalescing forming larger and larger drops as the time evolves. Figure 2 displays time series for an average density $\bar{\rho}=0.27$ (the time indicated in the labels is scaled by d^2/χ where d is the length of the box and $\chi=\kappa/\rho c$ is the liquid thermal diffusivity). The density distributions are emphasized in a grey scale, where the white regions describe the maxima of fluid density, the dark regions the minima. When the saturation state is reached ($t>5000$) a single liquid drop remains in the vapor system. Increasing in the following the mass of the system the final drop becomes bigger and bigger till an intermediate situation appears, represented in Fig. 3 for $\bar{\rho}=0.56$. For $0.42\leqslant\bar{\rho}\leqslant0.81$ the system hardly decides between drops and bubbles and therefore a planar liquid-vapor system may persist for very long times. For $\bar{\rho}\geqslant0.82$ the formation of a vapor bubble in a liquid is energetically favorable, as shown in Fig. 4. We have to remark that the boundary conditions for the density field at the solid walls play an important role for the contact angle at the solid surface and determine the position of the droplet. In our model we have controlled the contact angle through the density at the solid boundary. The influence of the boundary conditions on the droplet contact

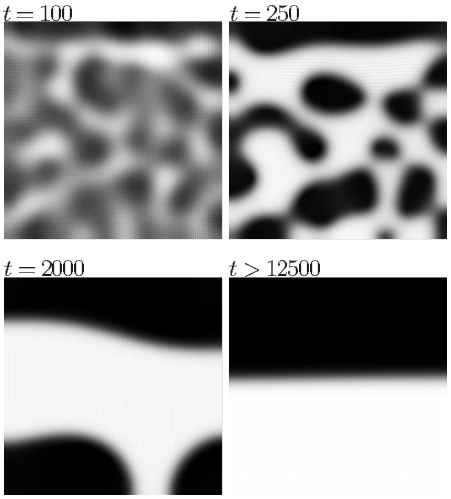

FIG. 3. Time series in (x,z) plane for the formation of a planar liquid-vapor system ($\bar{\rho}=0.56$) under microgravity conditions.

angle will be described in more detail elsewhere.

Next we concentrate on a two-component system in a gravitational field heated from above. We simulate quantitatively the experimental results given by Savino *et al.* in Ref. [4], concerning Marangoni migration on Fluorinert FC43 drops in a silicone oil (3 Cs). Fluorinert FC43 and silicone oil are two liquids with the densities 1.88 g/cm^3 and 0.89 g/cm^3, respectively. A complete list of the physical properties of the two investigated liquids is given in Ref. [4]. We scale the density to the heavier liquid density. So $\rho=1$ designates now the Fluorinert FC43 and $\rho=0.47$ the silicon oil. Under these conditions the bulk potential (4) takes the form

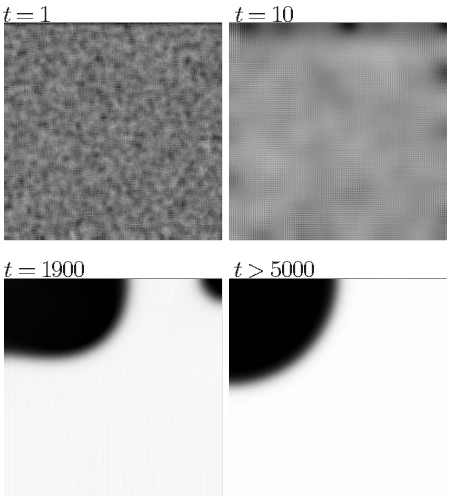

FIG. 4. Time series in (x,z) plane for the formation of a vapor bubble in a liquid ($\bar{\rho}=0.83$). The boundary conditions for the density field at the solid walls favor the bubble to be in contact with the solid surface.

$$f(\rho) = \frac{C}{2}(\rho - 0.47)^2(\rho - 1)^2.$$

No interfacial mass exchange appears between the two liquids, therefore we can again use the free energy functional (1).

As in the one-component system presented above, depending on the total mass, one can have either a Fluorinert drop in silicone oil or a silicon oil drop in Fluorinert. We focus on the formation of a Fluorinert droplet under the gravitational field. One obtains a drop falling down under a sedimentation force, which is the resultant between the gravity and the Archimedian force [see Fig. 5(a)]. Additionally we apply an external heating at the upper wall. In order to describe thermocapillary convection in the frame of the phase field model we have to consider the generalized surface tension coefficient $\mathcal{K}$ weakly depending on temperature $\mathcal{K}=\mathcal{K}_0-\mathcal{K}_T T(\mathcal{K}_T>0)$. The temperature field from Eq. (2) is described by the energy equation

$$\rho c\left(\frac{\partial T}{\partial t} + (\vec{v}\cdot\vec{\nabla})T\right) = \vec{\nabla}\cdot(\kappa\vec{\nabla}T) + q \tag{6}$$

with c as the specific heat capacity, κ as the thermal conductivity, and q as the internal heat generation rate per unit volume. In our system without interfacial mass exchange, no evaporation or condensation phenomena appear, and the dissipative heat production can surely be neglected. That means, we can assume $q=0$.

We consider a Fluorinert drop of diameter $D\approx 0.13$ mm inside a box 0.25 mm $\times$ 0.25 mm filled with silicone oil. The temperature differences top-bottom are about $\Delta T\approx(2-10)$ K. The temperature gradient generates a surface tension gradient along the droplet interface. The lowering of surface tension at its leading pole—hotter than the rear pole—induces Marangoni flows inside and outside of the drop [see the streamlines plotted in Fig. 6(b)]. Shearing around the droplet surface creates a net force on the drop, a Marangoni pushing force $\vec{F}_M$ directed upwards, towards the hotter wall. This force is the resultant between the viscosity and the pressure forces along the drop [4]:

$$(F_M)_i = \int_S n_j\sigma_{ij}dS - \int_S pn_i dS$$

(σ is the viscous stress tensor).

For a sufficiently high temperature gradient (sufficient Marangoni stress) the Marangoni pushing force $\vec{F}_M$ can balance the sedimentation force caused by the gravitational field. Hence in the steady state a floating liquid drop can occur, as depicted in Fig. 5(b) for $\Delta T=2$ K. If $|\vec{\nabla}T|$ increases further, the drop moves up and finds its equilibrium at a higher position [see in Fig. 5(c) the droplet position at $\Delta T=9$ K].

Recent studies on Marangoni migration in one-component fluids show that, latent heat released or absorbed at the interface drastically changes the hydrodynamic flow around the droplet [6]. As result the temperature becomes almost

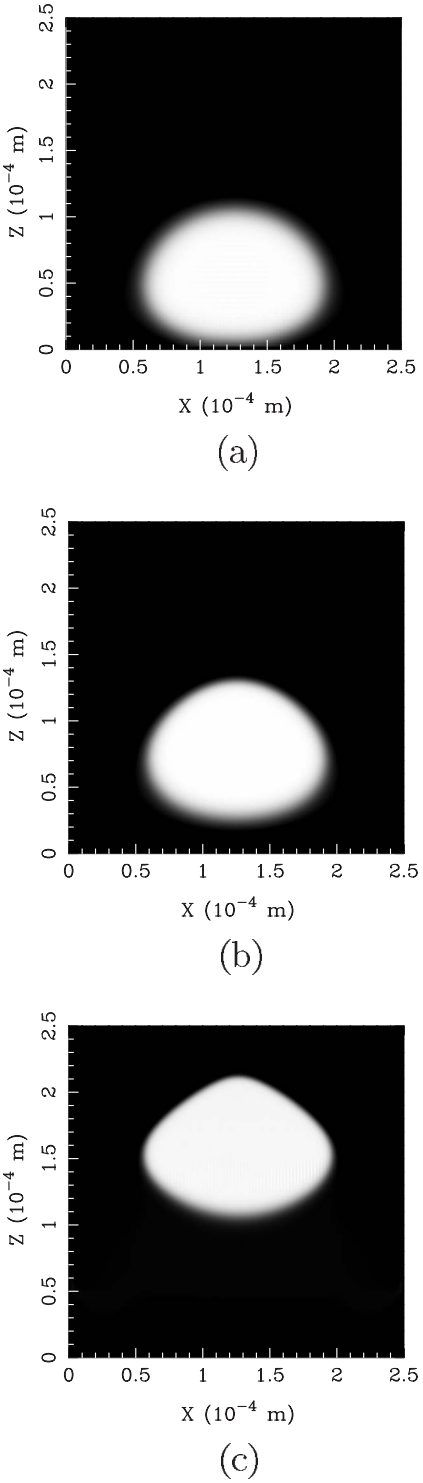

FIG. 5. Position of Fluorinert FC43 droplet (D=0.13 mm) in 3 Cs silicon oil 3 in (x,z) plane (0.25 mm×0.25 mm): (a) isothermal conditions, (b) ΔT=2 K, (c) ΔT=9 K.

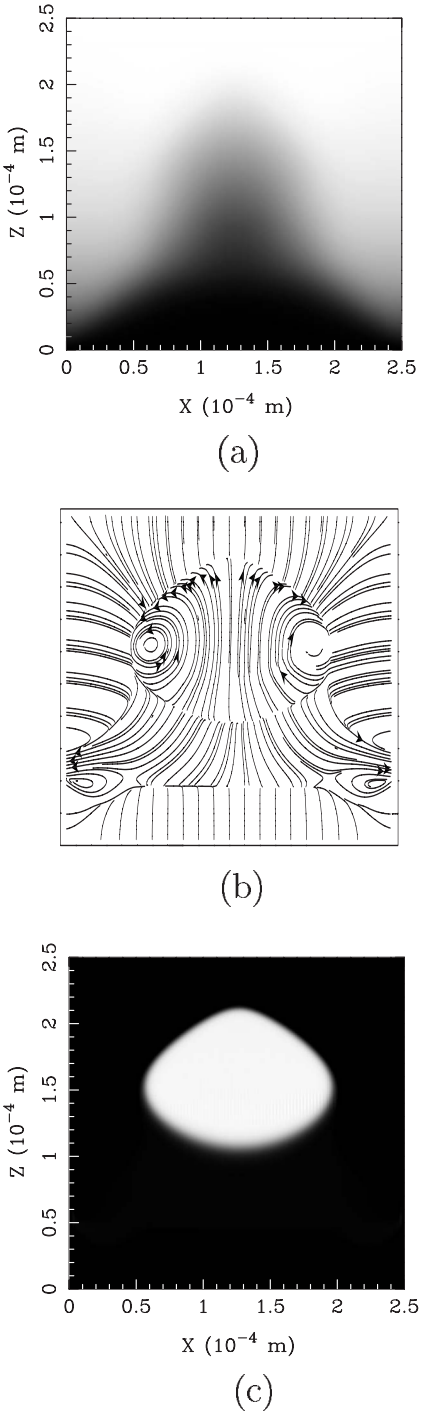

FIG. 6. The balance between Marangoni and gravity effects leads to a floating Fluorinert droplet in silicone oil (d=0.25 mm, D=0.13 mm, ΔT=9 K). Frame (a) shows the profile of temperature in (x,z) coordinates (the system is heated from above), (b) the streamlines, and (c) the density.

homogeneous inside the droplet and the Marangoni effect arising from the surface tension gradient is much suppressed. For example, for a liquid droplet in CO_2 the droplet velocity is much decelerated and completely vanishes for large enough droplets. We plan to extend our phase field modelling to one-component systems with interfacial mass exchange and to examine the latent heat effects close to water drops in vapor atmosphere at usual room temperatures.

Summarizing, we developed a scheme to study drops and bubbles using the phase field formalism. A randomly distributed initial density evolves to phase separation and single droplet formation. For two-component liquid-liquid systems we have performed numerical simulations for Marangoni mi-

gration in a vertical temperature gradient in the presence of a gravitational field. A good agreement between our phase field simulations and the experiments done by Savino *et al.* in Ref. [4] is obtained.

Simple, flexible and elegant, the actual model can become a useful tool for describing different phenomena with large applications in material and chemical engineering as Marangoni migration, chemically driven running drops, drop spreading on a solid surface, drop motion on an inclined substrate under gravity effects, or oscillatory thermocapillary convection around bubbles heated from above at very large Marangoni numbers.

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[1] N. O. Young, J. S. Goldstein, and M. J. Block, J. Fluid Mech. **6**, 350 (1959).
[2] R. Balasubramaniam and S. R. Subramanian, Phys. Fluids **12**, 733 (2000).
[3] L. Zhang, S. R. Subramanian, and R. Balasubramaniam, J. Fluid Mech. **448**, 197 (2001).
[4] R. Savino, R. Monti, and G. Alterio, Phys. Fluids **13**, 1513 (2001).
[5] R. Balasubramaniam and S. R. Subramanian, Phys. Fluids **16**, 3131 (2004).
[6] A. Onuki and K. Kanatani, Phys. Rev. E **72**, 066304 (2005).
[7] R. Borcia and M. Bestehorn, Phys. Rev. E **67**, 066307 (2003).
[8] R. Borcia, D. Merkt, and M. Bestehorn, Int. J. Bifurcation Chaos Appl. Sci. Eng. **14**, 4105 (2004).
[9] R. Borcia and M. Bestehorn, Eur. Phys. J. B **44**, 101 (2005).
[10] R. Borcia and M. Bestehorn, J. Optoelectron. Adv. Mater. **8**, 1037 (2006).
[11] R. Borcia, D. Merkt, and M. Bestehorn, Int. J. Bifurcation Chaos Appl. Sci. Eng. **16**, 2705 (2006).
[12] D. Jasnow and J. Viñals, Phys. Fluids **8**, 660 (1996).
[13] L. M. Pismen and Y. Pomeau, Phys. Rev. E **62**, 2480 (2000).
[14] J. P. Burelbach, G. S. Bankoff, and S. H. Davis, J. Fluid Mech. **195**, 463 (1988).