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Can diffuse-interface models quantitatively describe moving contact lines?

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Abstract. The three-phase contact line is a long-standing problem in the physics and hydrodynamics of interfaces. The traditional sharp-interface Navier-Stokes formulation encounters a non-integrable stress singularity, which is commonly avoided by introducing slip at the contact line. In recent years, diffuse-interface models have emerged as an alternative method. They are attractive in regularizing the singularity in a more rational manner, and in the meantime supplying a means for describing the interfacial motion on the large scale. Although a number of groups have carried out diffuse-interface computations of moving contact lines, a closer inspection shows that some fundamental questions remain to be answered. For example, how can a sharp-interface limit be realized to produce a solution that is independent of the interfacial thickness? How to determine model parameters so as to match a specific experiment? Finally, is it possible to make quantitatively accurate predictions of the moving contact line using diffuse-interface models? Using the Cahn-Hilliard model as an example, we describe these issues and suggest solutions.

1 Introduction

The three-phase contact line is perhaps the most significant unresolved problem in multiphase flows. Consider the steady-state displacement of fluid 2 by fluid 1 on a solid wall (Fig. 1), with the reference frame attached to the contact point. A straightforward formulation of the problem runs into a singularity at the contact line, where the velocity should be zero as the contact line is stationary in this reference frame, but in the meantime should equal the wall velocity U by virtue of the no-slip boundary condition. It is generally agreed that the difficulty arises from the fact that microscopic physics dominates the local behavior at the contact line, and the continuum Navier-Stokes formulation fails to represent that. A number of comprehensive reviews have

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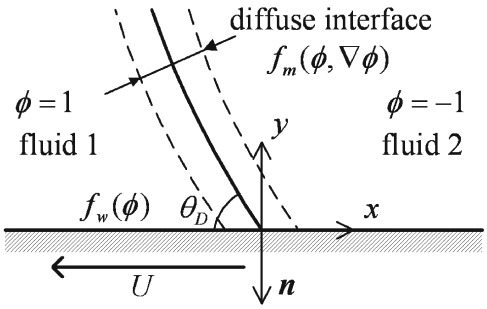

Fig. 1. A dynamic contact line represented in a diffuse-interface framework. $\boldsymbol{n}$ is the outward normal to the wall. The phase-field variable ϕ represents the two fluid bulks by $\phi = \pm 1$ and the fluid-fluid interface by $\phi = 0$. The dynamic contact angle θ_D is defined by the slope at which the interface intersects the substrate.

summarized the efforts to resolve this over the past several decades, e.g., [1–4]. We will focus on a relatively recent aspect of the problem, and will not try to be comprehensive in reviewing the literature.

To describe the local dynamics at the contact line, one may take either a hydrodynamic or a molecular-kinetic viewpoint [3]. The first is exemplified by the Voinov-Cox matched asymptotic solution [5,6]. An ad hoc slip length is introduced to carve out the immediate neighborhood of the contact line from the hydrodynamic domain. Thus a fluid-mechanical solution can be obtained outside. The key results from this type of analysis are the following. First, the outer solution is insensitive to the slip models. The upside of this recognition is that a faithful description of the local dynamics is unnecessary for an accurate solution of the macroscopic flow. The downside is that measurements, limited to larger length scales, would be ineffective in elucidating the local dynamics. Second, the apparent contact angle θ_A is determined by viscous bending in such a manner [6]:

$$g(\theta_A) = g(\theta_S) + Ca \ln(\delta^{-1}), \tag{1}$$

where g is a complex but known function given by Cox [6], θ_S is the static contact angle, and $\delta = l_s/W$ is the ratio between the slip length and the macroscopic length. The capillary number $Ca = \mu U/\sigma$ indicates the strength of viscous bending relative to the surface tension σ. This formula has been corroborated by experimental observations of drop spreading on a highly wettable surface [7,8], but fails to represent data of liquid-liquid displacement [9]. Such asymptotic solutions are limited to vanishing Ca and simple geometries. For complex large-scale flow computations, ad hoc slip conditions have been used to bypass the contact line difficulty. These often use the Navier condition [10,11] and sometimes a so-called "numerical slip" [12,13].

The molecular-kinetic approach [14], on the other hand, treats the motion of the contact line as the consequence of molecules jumping from one interface to another. This allows one to predict the contact line speed or dynamic contact angle in terms of the intensity of jumping. The results differ from those of the hydrodynamic predictions, and the possibility of reconciling and perhaps integrating the two approaches has been discussed by Blake [3].

The diffuse-interface model attempts to integrate the micro- and macroscopic scales in a single framework. The model treats the interface as a diffuse and continuous layer. By expanding the density distribution function and then truncating the nonlocal intermolecular forces, one coarse-grains the microscopic physics at the contact line into fluid-fluid and fluid-substrate interfacial energies. Via a variational procedure, these are then incorporated into the continuum hydrodynamic model [15]. On this

mesoscopic scale, the motion of the contact line is effected by diffusion across the interface and there is no singularity. Being a continuum theory, the diffuse-interface model is amenable to macroscopic flow simulations in complex geometry. So far, the Cahn-Hilliard version of the model, called the CH model hereafter, has been especially popular in computing contact line problems [16–23].

These efforts have produced results with correct qualitative features of the contact line. Though promising, fundamental questions remain about this approach. The first and most obvious concerns the thickness of the diffuse interface. Real interfaces between immiscible fluids are of molecular dimensions. In computing a macroscopic flow, on the other hand, current computing powers can reach down to perhaps micrometer scales. Are the results relevant to real physical situations? Second, how to determine the phenomenological model parameters in order to match experiments with specific fluids and solid substrates? Finally, once these model parameters are determined, are they to be taken as material properties? In other words, will the same model parameters predict the correct solution in different geometries, where the contact line is driven differently? In this Discussion & Debate piece, we suggest tentative answers to these questions and point out remaining puzzles in this difficult but fascinating problem.

2 Formulation of the Cahn-Hilliard model

It is necessary for the ensuing discussions to first list the governing equations, boundary conditions and the parameters of the Cahn-Hilliard model. More detailed presentations can be found in the literature, e.g., [4,17,24,25]. For the system depicted in Fig. 1, we introduce a phase field variable ϕ that takes on bulk values of ± 1 in the two fluids and varies sharply but continuously across the interface. We assume zero inertia and equal viscosity between the two fluids. In reality, of course, both inertia and viscosity contrast can influence contact line motion. These have been investigated elsewhere [21,24, e.g.] and are not important to the focus of this article. We write the governing equations as

$$\nabla \cdot \boldsymbol{v} = 0, \tag{2}$$

$$\nabla p = \mu \Delta \boldsymbol{v} + G \nabla \phi, \tag{3}$$

$$\frac{\partial \phi}{\partial t} + \boldsymbol{v} \cdot \nabla \phi = \nabla \cdot (\gamma \nabla G), \tag{4}$$

where the bulk chemical potential $G = \lambda \left[-\nabla^2 \phi + (\phi^2 - 1)\phi/\epsilon^2 \right]$, λ being the interfacial energy density and ϵ a capillary width indicating the interfacial thickness. In equilibrium, the fluid-fluid interfacial tension is given by

$$\sigma = \frac{2\sqrt{2}}{3} \frac{\lambda}{\epsilon}. \tag{5}$$

Equation (4) is the Cahn-Hilliard equation, in which γ is the mobility parameter, taken to be a constant here. These are supplemented by the following boundary conditions on the solid surface:

$$\boldsymbol{v} = \boldsymbol{U}, \tag{6}$$

$$\boldsymbol{n} \cdot \nabla G = 0, \tag{7}$$

$$\frac{\partial \phi}{\partial t} + \boldsymbol{v} \cdot \nabla \phi = -\Gamma L, \tag{8}$$

where $L = \lambda \boldsymbol{n} \cdot \nabla \phi + f_w'(\phi)$ is the surface chemical potential, which arises from the variation of f_w, the interfacial energy between the wall and the fluids:

$$f_w(\phi) = -\sigma \cos\theta_S \frac{\phi(3-\phi^2)}{4} + \frac{\sigma_{w1} + \sigma_{w2}}{2}. \tag{9}$$

Here σ_{w1} and σ_{w2} are the fluid-solid interfacial tensions for the two fluids, related to the static contact angle θ_S through Young's equation $\sigma_{w2} - \sigma_{w1} = \sigma \cos\theta_S$. Equation (6) asserts no slip on the solid substrate. Although Cahn-Hilliard diffusion has removed the contact line singularity, a slip velocity could be included here as well [18,26]. Equation (7) implies zero flux through the wall. Finally, Eq. (8) allows relaxation of the wall layer as driven by the surface potential L, Γ being a rate constant [17,26].

Of the four model parameters, λ, ϵ, γ and Γ, ϵ will be determined by the requirement of attaining the sharp-interface limit, and then λ is constrained by Eq. (5). Although γ and Γ may be viewed as material properties related to parameters in molecular-kinetic theory [17,27], their values are not known for any specific fluid-solid combinations. How to choose them to match specific experiments will be one of the issues to be discussed in the following.

3 Sharp-interface limit

A fundamental question in diffuse-interface models is about the thickness of the interface. This has an analytical aspect and a computational one. Mathematically, if one takes the limit of $\epsilon \to 0$, does the model converge to the conventional formalism of a Navier-Stokes system with a sharp interface? Computationally, how thin must the interface be to produce results that no longer depend on ϵ and therefore effectively correspond to the sharp-interface limit?

For a *closed interface*, such as one enclosing a drop, both of these questions have been answered. The existence of the sharp-interface limit has been proved mathematically [28], and numerical experimentation has established guidelines on maximum allowable ϵ values [29,30]. These are typically in terms of the Cahn number $Cn = \epsilon/W$, the ratio between the capillary width and the macroscopic length scale of the problem. Thus, one can obtain numerical results at an artificially large ϵ, typically with $Cn \sim 0.01$, that accurately reflect reality having molecular-scale interfaces.

If a fluid interface intersects a solid wall and forms a three-phase wetting line, the situation is much murkier. No mathematical proof is available. Because the very motion of the contact line is via interfacial diffusion, a sharp-interface limit with zero diffusion would produce a pinned contact line. Instead, we should seek a limit with a finite diffusive flux across the interface. Such a limit is obtained by keeping the Cahn-Hilliard mobility γ fixed while reducing ϵ.

First, such a limiting process can be rationalized by scaling arguments [24,26]. Balancing the flow effect in convecting the interface and its relaxation by CH diffusion, Yue *et al.* [24] showed that the chemical potential G varies not over ϵ but a larger *diffusion length* $l_d = (\gamma\mu)^{1/2}$, and that the variation of G across the contact line scales with $\mu U/l_d$. Thus, as $\epsilon \to 0$ at a fixed γ, both l_d and the variation of G stay constant, resulting in a finite diffusive flux that, of necessity, produces the slip velocity U.

Moreover, this conceptualization for the sharp-interface limit is supported by numerical evidence. We computed the steady-state solutions for an interface sheared between parallel plates or driven by a pressure gradient in a capillary tube [24]. Over all the capillary numbers tested, the interface approaches a definite limit as $\epsilon \to 0$ while all other parameters, especially the mobility γ, are fixed. This is illustrated by

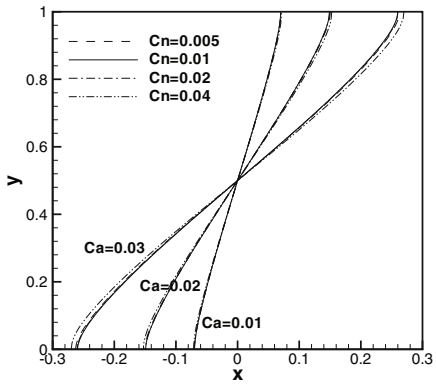

Fig. 2. Convergence of the moving contact line to the sharp-interface limit with decreasing Cn, shown for steady interfacial shapes in a Couette flow at three Ca values. $\theta_S = 90^\circ$. The γ value corresponds to $S = l_d/W = 0.01$. Adapted from Yue *et al.* [24] with permission, ©2010 Cambridge University Press.

Fig. 2 in the special case of instant wall relaxation ($\Gamma = \infty$), but is generally true. We also observed that if γ is reduced simultaneously toward zero, the contact line becomes pinned on the substrate. Conversely, if γ is increased, say as ϵ^{-1}, complete slip prevails at the contact line.

Another interesting finding is that the diffusion length l_d can be related to the slip length in the asymptotic solutions [6]. This is informed by the velocity field in the neighborhood of the contact line. Over all the parameters tested, the stagnation point always occurs at a distance of $2.5l_d$ above the wall. Since in the reference frame of Fig. 1, the fluid interface is stationary and the wall is moving, graphically it is natural to take this distance to be the conventional slip length:

$$l_s = 2.5l_d. \tag{10}$$

This equation highlights a connection between the CH model and the Cox solution, first suggested by Jacqmin [17] on analytical grounds. In our view, this is not a coincidence but reflects the fact that the macroscopic features of the solution are insensitive to details of the contact-line model. Thus, two phenomenological models such as the Cox and Cahn-Hilliard yield pictures that overlap to a great extent.

Base on numerical data, we proposed an empirical criterion for attaining the sharp-interface limit in computations:

$$Cn \leq 4S, \tag{11}$$

where $S = l_d/W$. Therefore, the interfacial thickness has to be smaller than a multiple of the diffusion length. This can be rationalized thus: a faster diffusion is more capable of maintaining the integrity of the interfacial profile, rendering it more tolerant to distortions by flow even on relatively thick interfaces. With the concept of the sharp-interface limit, we have taken a first step toward answering the question raised in the title of this article. But further problems remain.

4 Difficulty with small slip lengths

The connection between the diffusion and slip lengths, Eq. (10), appears to offer a means to determine the value of γ in simulating a real system. However, estimations

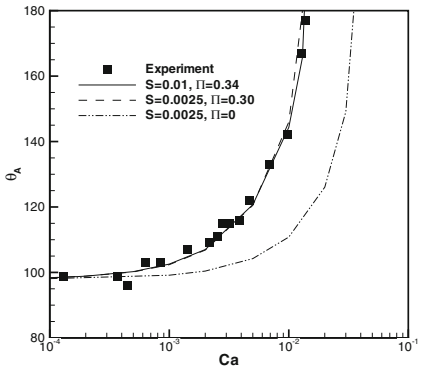

Fig. 3. Apparent contact angle in liquid-liquid displacement in a capillary tube. The symbols are experimental data for a glycerin-silicone oil system with $\theta_S = 98^\circ$ and receding-to-advancing viscosity ratio of 0.9 [9]. The lines are computations at artificially large diffusion lengths. $Cn = 0.005$.

typically put the slip length l_s on the order of a few nanometers for smooth surfaces [9,31]. Then to attain the sharp-interface limit, Eq. (11) again requires us to reduce the interfacial thickness to nanometers, putting us back in the same difficulty that we started with.

Drawing on ideas in analysis of asymptotic solutions [32, e.g.], we have suggested a strategy for circumventing this difficulty [25]. Since the macroscopic solution is insensitive to the actual physics on the order of the slip length l_s, we can re-parametrize the boundary value problem into one that has a larger *artificial slip length* $\bar{l}_s$ or diffusion length $\bar{l}_d$, as well as an artificial wall relaxation parameter $\bar{\Gamma}$ that is chosen so as to yield the accurate macroscopic solution.

This strategy relies on two factors. The first is the large gap between the length scales l_s and W. This makes it possible to obtain a solution on an intermediate length scale $\bar{l}_s$: $l_s \ll \bar{l}_s \ll W$. Thus $\bar{l}_s$ is large enough as to be computable, yet still negligibly small on the scale of W so that the solution captures the smallest length of interest in a real hydrodynamic system. The second factor is an apparent competition between Cahn-Hilliard diffusion (indicated by γ) and wall energy relaxation (indicated by $1/\Gamma$). This has been explained in detail in [25]. Briefly, γ promotes relaxation of the interfacial profile toward the equilibrium one, thus resisting the viscous bending by flow. As a result, γ tends to reduce the dynamic contact angle θ_D. Conversely, through the boundary condition of Eq. (8), wall relaxation tends to rotate the interface as if it is hinged at the contact line. Smaller Γ (slower wall relaxation) produces a larger dynamic contact angle θ_D.

We have derived analytical formulas for the dependence of θ_D on Γ, and for the amount of wall relaxation needed to compensate for using a large $\bar{\gamma}$ or $\bar{l}_d$ [25]. These are further confirmed by numerical computations. As an example, Fig. 3 compares simulations using two pairs of $\bar{\gamma}$ and $\bar{\Gamma}$ values in fitting an experimental data set of Fermigier and Jenffer [9]. The experimental system has a slip length below $10^{-4}W$, much too small for the numerical resolution. Using a computable diffusion length $\bar{l}_d$ corresponding to $S = 2.5\times10^{-3}$, not surprisingly, produces a gross underprediction of the apparent contact angle θ_A (dot-dash curve in Fig. 3). Introducing wall relaxation at $\Pi = (\mu\bar{\Gamma}W)^{-1} = 0.3$ brings the prediction at the same $\bar{l}_d$ to overlap the data closely. Furthermore, equally accurate representation of the data can be obtained at an even larger $\bar{l}_d$ ($S = 0.01$) by using a slower wall relaxation ($\Pi = 0.34$). This compensation

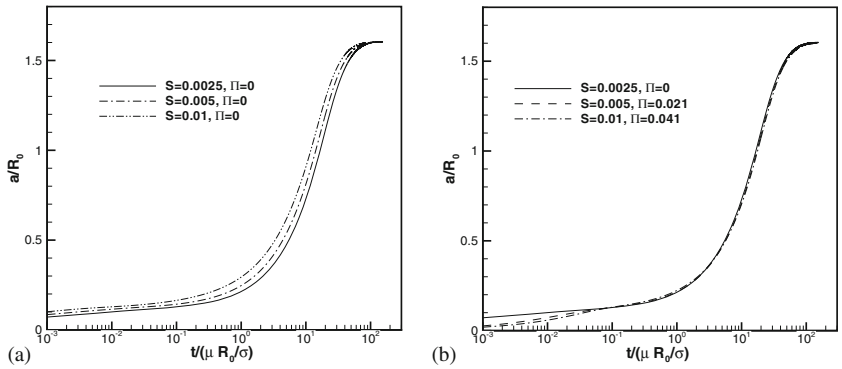

Fig. 4. Simulations of drop spreading on a partially wetting substrate with $\theta_S = 60°$, $Cn = 0.005$. a is the radius of the contact circle and R_0 is the intial radius of the spherical drop. Time is scaled by the capillary time $\mu R_0/\sigma$. (a) Increasing the CH diffusion S accelerates spreading. (b) A universal curve is obtained by compensating the larger S by slower wall energy relaxation (larger Π).

between $\bar{\gamma}$ and $\bar{\Gamma}$ conforms to the formula derived by Yue *et al.* [25]. The same effect is observed in drop spreading, as shown in Fig. 4. Simulations at different diffusion lengths collapse onto a master curve if the proper amount of wall relaxation is used to compensate for the stronger diffusion.

Returning now to the question raised in the Introduction on how to determine model parameters, we realize that we would need the *true* values of γ (or equivalently the diffusion length l_d or slip length l_s) and Γ before we can use the "compensation" strategy to determine $\bar{\Gamma}$ at a larger $\bar{l}_d$. But such true values are not known in general; there are estimations and measurements of slip lengths, but it is unclear how Γ can be determined experimentally. Thus, we suggest the following protocol for simulating an experiment. First, we select the smallest ϵ or Cn that is computationally affordable. Since the interfacial profile requires roughly 10 mesh points to resolve [29], a thinner interface leads to more grid points and a larger numerical problem. We then select a $\bar{\gamma}$ value according to Eq. (11) to ensure that the sharp-interface limit is reached. Finally, we fit one experimental data point to determine the wall relaxation $\bar{\Gamma}$. For drop sliding or spreading on substrates, we may fit an instantaneous position of the contact line. For interfaces being driven at a constant speed, we may fit the apparent contact angle.

With the CH model parameters thus determined, can we use them to predict moving contact lines in other geometries for the same fluid and substrate materials as have provided the data? In other words, are parameters $\bar{\gamma}$ and $\bar{\Gamma}$ material properties that are independent of the flow geometry and speed? This is the question to which we turn next. Of course, $\bar{\gamma}$ and $\bar{\Gamma}$ are *numerical parameters* devised to avoid resolving the small slip length l_s; they could be *material parameters* only in the sense that they are traceable to the true, albeit unknown, γ and Γ.

5 Universality

The computational strategy discussed above amounts to compensating the use of a larger $\bar{l}_d$ by imposing a larger dynamic contact angle θ_D, determined from the wall energy relaxation. Thus, the question of whether $\bar{\gamma}$ and $\bar{\Gamma}$ are material parameters is

connected to the issue of *material boundary conditions* discussed in the context of the matched asymptotic solution [33,34]. The solution of Cox [6] predicts the interface slope at an intermediate length r between l_s and W:

$$g[\theta(r)] = g(\theta_w) + Ca \ln\left(\frac{r}{l_s}\right), \tag{12}$$

where g is the same function as in Eq. (1) and θ_w is the microscopic or actual contact angle at the contact line. If θ_w and l_s are both material constants, independent of the outer geometry and Ca, then the angle $\theta(r)$ can be imposed at a distance $r \gg l_s$ as a universal boundary condition for computing any outer flow involving the same materials.

Marsh *et al.* [33] presented an interesting experimental investigation of the above hypothesis using a glass rod that is thrust along its axis into a quiescent pool of silicone oil, with perfect wetting ($\theta_S = 0$). During flow, the actual contact angle θ_w is assumed fixed at θ_S, which is by definition independent of the outer flow conditions. Therefore, the question boils down to whether the slip length l_s is a material constant. By varying the angle and speed of the glass rod, they showed that indeed l_s is independent of the geometry (in this case represented by the inclination angle of the rod relative to the free surface), but it decreases mildly with Ca. This dependence on Ca apparently dashes the hope for universality. In computing a drop sliding down a slope, for example, Ca cannot be prescribed a priori, and neither could a material boundary condition.

In the diffuse-interface context, the corresponding question—is the *diffusion length* l_d a material constant—has never been explicitly asked before. At present we have very few results that bear on that question. For one, Fig. 3 shows that the $(\bar{\gamma}, \bar{\Gamma})$ pair fits the experimental data extremely well over the entire Ca range, up to the point of wetting failure with $\theta_A \to 180°$. This seems to suggest an Ca-independence of the model parameters. It is not clear how to rationalize the apparent discrepancy with Marsh *et al.*'s conclusion of Ca dependence. On the experimental side, one might note that their l_s data are noisy with large error bars, and the dependence on Ca is a very weak one that essentially vanishes for large Ca. On the computational side, the apparent contact angle θ_A is insensitive to the slip length l_s for $Ca \ll 1$ according to Eq. (1). Similarly for the CH model, θ_A might simply be too insensitive to reflect the small variation of l_s with Ca suggested by Marsh *et al.*'s experiment.

The question of *geometry independence* of CH parameters can be investigated in the following way. Select the proper $\bar{\gamma}$ and $\bar{\Gamma}$ values to fit an experimental data set, say for displacement in a capillary tube. Now use the same parameter values to predict the flow in a different geometry, say for drop spreading or sliding. Will this prediction agree with an experiment in this geometry, using exactly the same fluid pair and solid surface as in the first experiment? Such a scheme is hampered at present by a lack of suitable experiments. In the literature we have not found a single pair of experiments in different geometries using precisely the same materials. Different groups report different wetting angles for nominally the same materials. It is well known that minute details in surface handling and cleaning may have significant effects on contact angles and wettability. The dearth in experimental benchmark is exacerbated by the fact that the CH formalism used here does not lend itself well to static contact angles close to 0° and 180°. For interfaces nearly parallel to the substrate, small errors in the ϕ field induces much larger ones in the contact angle. This rules out the completely wetting glass-silicone combination commonly used in experiments [7,9,35, e.g]. Therefore, we have to leave open the question of the universality of CH model parameters pending more suitable experimental benchmarks.

6 Concluding remarks

In this article, we have summarized our own investigations into the use of Cahn-Hilliard (CH) model for moving contact lines. A series of fundamental questions are raised, for some of which, but not all, we have suggested answers. For instance, for the simulation results to have any physical relevance at all, they have to be independent of the numerical interfacial thickness. This can be ensured by attaining the sharp-interface limit. The numerical difficulty in resolving a typically small diffusion or slip length is bypassed by introducing wall relaxation that compensates for the use of an artificially large slip length. Based on these, the answer to the question raised in the title is probably cautiously affirmative.

But we hasten to emphasize the issues that remain to be addressed. So far the sharp-interface limit is established by scaling arguments and empirical evidence from numerical results. There is no mathematical proof comparable to that for closed interfaces without three-phase contact lines. Similarly, the strategy of using wall relaxation to bypass the resolution of small slip lengths is based on a limited set of numerical results. It remains to be tested by applications in a wide range of geometries and parameter values. Finally and significantly, the question of the physical universality of the model parameters remains open. From a kinetic-molecular viewpoint, it would not be surprising if the slip length turns out not to be a material property but to depend on the velocity and the geometry of the flow. After all, these conditions determine how the interface is being driven at the contact line, and hence how molecules would jump from one interface to another. From a continuum viewpoint, a non-universal slip length is inconvenient to say the least, and may even compromise the predictive power of a theory such as the CH model.

Needless to say, the phenomenological nature of the CH model underlies the entire discussion. The idea of interfacial diffusion is based on a postulation instead of a physical law, at least in the current context of multiphase fluid dynamics. In this regard, the model is similar and indeed connected to the slip-based models due to Cox and others. For instance, the Cahn-Hilliard diffusion length is related to the slip length, and the idea of applying a "material boundary condition" at a larger length scale has a counterpart in our proposed strategy for handling the small slip length. In the end, neither escapes the need for empirical inputs; it is the slip length for the Cox model, and the mobility and wall relaxation parameters for the CH model. This again raises the question of whether these are universal material properties. However, the CH model does enjoy several advantages over the slip models. It fits the data better [25], allows large-scale computations, and has a certain elegance in regularizing the contact line singularity and capturing the macroscopic interface using the same phase field. These are the rewards for its relative complexity.

It is possible to add further to the version of the CH models presented here. For instance, the generalized Navier boundary condition of Qian *et al.* [4,26] combines the Navier slip and CH diffusion. This further increases the number of model parameters, but reflects the observation of slip in molecular-dynamics simulations. It will be interesting to explore how the extra slip improves the CH model, both in a theoretical sense and in large-scale simulations. This can be done only after a clear understanding of the simpler diffusion-only CH model, which is what we set out to do.

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