Thermodynamically consistent picture of the phase-field model of vesicles: Elimination of the surface tension

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In two recent papers [D. Jamet and C. Misbah, Phys. Rev. E **76**, 051907 (2007); **78**, 031902 (2008)], we considered a thermodynamically consistent model for vesicles and membranes, where we dealt, in the first paper, with the membrane local incompressibility condition, while in the second one with the bending energy and the derivation of a constitutive law of the composite fluid: ambient fluid+membrane. This is the last paper of this series and focuses on the elimination of surface tension (inherent in phase-field models), retaining the thermodynamically consistent model. We write down the complete set of equations and the full constitutive law for membranes embedded in a Newtonian fluid.

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I. INTRODUCTION

There is an overabundance of important problems in nature that involve moving boundaries: phase transitions, liquid-liquid or liquid-gas flows, membranes, vesicles, etc. Very often, these problems are highly nonlinear, and analytical solutions are the exception rather than the rule. Resorting to numerical simulations is often mandatory. However, the numerical solution of moving boundary problems is very challenging and can prove to be very complex as far as algorithms or data structures are concerned. For instance, in the front-tracking method [1], the moving boundary can be discretized using a triangular mesh, which is very difficult to handle, especially for three-dimensional and parallel simulations. These numerical difficulties (which also exist with other numerical techniques) are at the origin of the popularity of the phase-field method. Indeed, in the phase-field approach, the sharp interface is replaced by an equivalent diffuse interface. This *a priori* extra complexity is counterbalanced by the ease of its numerical implementation. Indeed, the two-phase problem is modeled by partial differential equations that are valid in the whole space (as if the boundaries were absent), and the motion of all the interfaces present within a system is part of the solution of these equations. The set of differential equations can thus be discretized using classical numerical schemes, and this highly simplifies the numerical implementation.

Another important virtue of phase-field models is that they are generally based on the definition of an energy functional. This characteristic thus potentially ensures their thermodynamic consistency. This consistency is important since it ensures that the model is well posed physically and mathematically. This well-posedness follows from the second law of thermodynamics. From a physical point a view, the consistency should always be a requirement and, from a mathematical point of view, it ensures the existence of a Lyapunov function (i.e., the entropy), which is an important characteristic in the development of stable numerical schemes (e.g., [9]).

Phase-field formulations lead naturally to a systematic fact: the interface is endowed with a surface tension energy. In many applications (solid-liquid transitions, liquid-liquid two-phase flows, etc.), capillarity plays a key role in the physics studied, and the existence of a built-in surface tension may be regarded as an advantage. However, in other physical systems such as vesicles (or biological membranes in general), interfaces are not endowed with any surface tension and the use of the classical phase-field model implies the existence of artificial capillarity.

Note that, even if a physical system possesses a surface tension, it may prove useful to suppress it from the phase-field equation, while introducing it explicitly in the momentum balance equation (allowing thus for an extra control on the surface energy, independently from the phase field). This method was adopted by Folch *et al.* [2] in the Saffmann-Taylor problem. The idea is to subtract from the phase-field equation a contribution that naturally cancels the leading-order surface tension term inherent in the phase-field equation. Their analysis is based on an asymptotic expansion and the modification proposed ensures that the dominant contribution vanishes to leading order. This was initially adopted for vesicles in Refs. [3,4]. Despite the merit of the method, this subtraction from the phase-field equation does not lend itself to a thermodynamically consistent model, as we shall see.

Phase-field models for vesicles and membranes are subjects of increasing interest [3–6]. Our focus is to suppress the surface energy by keeping intact the thermodynamic consistency of the model. This task is not trivial, as we shall see. Furthermore, the phase-field equation is generally coupled with other equations that are of primary physical interest, such as the fluid velocity. Hence, a modification in the phase-field equation should *alter* the evolution equations of the other physical variables. How the other equations should be modified in a coherently concerted fashion is a difficult task if one does not adopt a systematic well-posed general formu-

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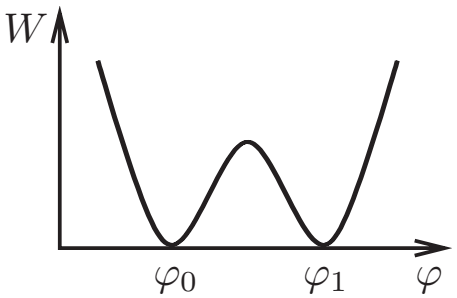

FIG. 1. Double-well function $W(\varphi)$.

lation. The present study offers a consistent framework to deal with these questions.

This paper is organized as follows. In Sec. II, the main characteristics of the classical phase-field model are presented and the model of Ref. [2] is briefly recalled. This analysis allows us to propose a model that eliminates the motion of the interface proportional to its curvature and that is thermodynamically consistent; this model is presented in Sec. III. In Sec. IV, we present a matched asymptotic expansion of the model. We show in particular that our model is equivalent to that of Folch *et al.* [2] up to second order. In Sec. V, our phase-field model is used to propose a thermodynamically consistent phase-field model for vesicles that incorporates local surface incompressibility and curvature energy.

II. CHARACTERISTICS OF THE CLASSICAL PHASE-FIELD MODEL

Let us recall the main features of the "classical" phase-field model. This will allow us to prepare the discussions of Sec. III.

We focus in this section on the phase field φ, and not on the other variables of physical interest (like the velocity $\boldsymbol{u}$, etc.). In the classical phase-field model the energy E reads

$$E^{cl}(\varphi,\boldsymbol{\nabla}\varphi) = W(\varphi) + \frac{\lambda}{2}(\boldsymbol{\nabla}\varphi)^2, \tag{1}$$

where $W(\varphi)$ is a double-well function (see Fig. 1) and λ is a positive coefficient, sometimes called the capillary coefficient (the reason for this denomination will become clear below). The function $W(\rho)$ is classically taken as a polynomial of degree 4. However, our reasoning works perfectly well with any other function, so we will not restrict the discussion to a specific form.

A. Thermodynamic equilibrium and consequences

The thermodynamic equilibrium is characterized by a minimum of energy. For a general function $E(\varphi,\boldsymbol{\nabla}\varphi)$, this condition yields[1]

$$\frac{\partial E}{\partial \varphi} - \boldsymbol{\nabla}\cdot\left(\frac{\partial E}{\partial \boldsymbol{\nabla}\varphi}\right) = 0. \tag{2}$$

For the particular case where the expression for $E(\varphi,\boldsymbol{\nabla}\varphi)$ is given by Eq. (1) and for a planar interface, the above equilibrium condition yields

$$\frac{dW}{d\varphi} - \lambda\varphi'' = 0, \tag{3}$$

where $\varphi' \equiv (d\varphi/dr)$, r being the coordinate normal to the interface. The above differential equation can be integrated once to get

$$\frac{\lambda}{2}(\varphi')^2 = W(\varphi). \tag{4}$$

This characterizes the profile $\varphi(r)$ across a planar interface at equilibrium. Provided that the function $W(\varphi)$ has adequate characteristics, this equation ensures that the phase field varies smoothly across the interface, thus defining a diffuse interfacial zone. This property is the basic attractive feature of phase-field models.

As pointed out in the Introduction, with the classical phase-field model, an interface is endowed with a surface tension energy. Let us call the total energy γ, namely,

$$\gamma = \int_{-\infty}^{+\infty}\left(W(\varphi) + \frac{\lambda}{2}(\varphi')^2\right)dr. \tag{5}$$

Using Eq. (4) one finds

$$\gamma = \int_{-\infty}^{+\infty}\lambda(\varphi')^2 dr. \tag{6}$$

This is the surface energy. As we shall see, in the classical model, the phase field is a tanh profile across the interface of width $\sqrt{\lambda}$, so that φ' is a Dirac-like function centered around the interface. Thus γ may be viewed as the excess energy due to the presence of the interface, and it is a positive quantity.[2]

Using Eq. (4), one can write

$$\gamma = \int_{\varphi_0}^{\varphi_1}\sqrt{2\lambda W(\varphi)}\,d\varphi, \tag{7}$$

where φ_0 and φ_1 are the phase-field values characteristic of the bulk phases [corresponding to the two minima of the double-well function $W(\varphi)$; see Fig. 1].

B. Dynamics: The Allen-Cahn equation

Out of equilibrium, the evolution equation for φ is the following relaxation equation (the so-called Allen-Cahn equation):

[1] We do not consider that the phase field is a conserved quantity.

[2] Note that, in the usual Gibbs definition of (extensive) surface quantities, it is not *a priori* always obvious how to fix in advance the sign of surface quantities. Contrariwise, γ, from the basic definition, must be positive.

$$\frac{\partial \varphi}{\partial t} = -\kappa \tilde{\mu}, \tag{8}$$

where the mobility κ is positive (and may depend on the thermostatic variables of the model) and $\tilde{\mu}$ is the generalized chemical potential defined by

$$\tilde{\mu} = \frac{\delta E}{\delta \varphi} = \frac{\partial E}{\partial \varphi} - \boldsymbol{\nabla} \cdot \left(\frac{\partial E}{\partial \boldsymbol{\nabla} \varphi} \right), \tag{9}$$

where $\delta / \delta \varphi$ stands for the functional derivative with respect to φ. The Allen-Cahn equation (8) is thermodynamically consistent. Indeed, on multiplying this equation by $\tilde{\mu}$, it is straightforward to show that

$$\frac{\partial E(\varphi, \boldsymbol{\nabla} \varphi)}{\partial t} = -\kappa \tilde{\mu}^2 < 0. \tag{10}$$

This inequality may be viewed as an expression of the second law of thermodynamics. Obviously, at steady state, the equilibrium condition (2) is recovered.

In the classical phase-field model where the energy $E(\varphi, \boldsymbol{\nabla} \varphi)$ is given by Eq. (1), the Allen-Cahn equation reads

$$\frac{\partial \varphi}{\partial t} = -\kappa \left(\frac{dW}{d\varphi} - \lambda \nabla^2 \varphi \right). \tag{11}$$

A requirement of phase-field models is that they should recover the sharp interface equations (i.e., a surface of discontinuity). This task is achieved by performing matched asymptotic expansions in powers of the interface width. Formally, one defines a small parameter ϵ representing the ratio (h/L), where h is the interface thickness and L is a characteristic outer length, typically the radius of an inclusion (droplet, vesicle, etc.). In the interface region φ varies abruptly (in the inner region), while it is smooth elsewhere (outer region). Using the method of matched asymptotic expansions, for the classical model where the energy is given by Eq. (1), it is by now quite well known that (see also later in this paper), at zeroth order in ϵ, φ is the equilibrium profile of a planar interface characterized by Eq. (4). At first order in ϵ, it is found that the speed of the normal displacement of the interface, v_n, obeys

$$\bar{v}_n^{(0)} \propto \bar{\mathcal{C}}^{(0)} \int_{-\infty}^{+\infty} (\bar{\varphi}^{(0)}_{,\bar{r}})^2 d\bar{r} \tag{12}$$

where $\bar{\psi}$ represents the inner solution of a quantity ψ, the superscript (0) represents the zeroth-order approximation, r is the coordinate normal to the interface, and $\mathcal{C}$ is the interface curvature (see Appendix A for details). This result shows that the classical Allen-Cahn equation induces a motion of the interface proportional to its curvature. This is a quite classical result.

Mathematically, we can view the origin of this contribution as follows. In the Allen-Cahn equation, the term $\nabla^2 \varphi$ can be decomposed as follows:

$$\nabla^2 \varphi = |\boldsymbol{\nabla} \varphi| \, \boldsymbol{\nabla} \cdot \left(\frac{\boldsymbol{\nabla} \varphi}{|\boldsymbol{\nabla} \varphi|} \right) + \left(\frac{\boldsymbol{\nabla} \varphi}{|\boldsymbol{\nabla} \varphi|} \otimes \frac{\boldsymbol{\nabla} \varphi}{|\boldsymbol{\nabla} \varphi|} \right) : \boldsymbol{\nabla} \boldsymbol{\nabla} \varphi. \tag{13}$$

The vector $\boldsymbol{\nabla} \varphi / |\boldsymbol{\nabla} \varphi|$ is the unit normal to the interface and $\boldsymbol{\nabla} \cdot (\boldsymbol{\nabla} \varphi / |\boldsymbol{\nabla} \varphi|)$ is therefore its local mean curvature $\mathcal{C}$. The curvature dependence of the interface velocity (12) is the signature of the zeroth-order approximation of the term $[|\boldsymbol{\nabla} \varphi| \boldsymbol{\nabla} \cdot (\boldsymbol{\nabla} \varphi / |\boldsymbol{\nabla} \varphi|)]$ of Eq. (13). The second term of the right-hand side of Eq. (13) represents the second derivative in the direction normal to the interface and is the equivalent of φ'' in the case of a planar interface. This term is therefore important because, combined with $W(\varphi)$, it ensures the regularity of the phase-field profile in the direction normal to the interface [see Eq. (3)].

The main idea of the model proposed by Folch *et al.* [2] is to eliminate the curvature dependence of the interface velocity *at zeroth order* by dropping the contribution $[|\boldsymbol{\nabla} \varphi| \boldsymbol{\nabla} \cdot (\boldsymbol{\nabla} \varphi / |\boldsymbol{\nabla} \varphi|)]$ from the Allen-Cahn equation (11). It is worth pointing out that their analysis [2] is a zeroth-order analysis of the interface velocity. This remark is important for the subsequent discussions. The modified Allen-Cahn equation proposed in Ref. [2] is thus the following:

$$\frac{\partial \varphi}{\partial t} = -\kappa \left[\frac{dW}{d\varphi} - \lambda \nabla^2 \varphi + \lambda |\boldsymbol{\nabla} \varphi| \, \boldsymbol{\nabla} \cdot \left(\frac{\boldsymbol{\nabla} \varphi}{|\boldsymbol{\nabla} \varphi|} \right) \right]. \tag{14}$$

However, as pointed out in the Introduction, this modified Allen-Cahn equation is not thermodynamically consistent: no energy has been found to be associated with this equation. We shall show below how this equation must be modified within a thermodynamic picture. Moreover, one must answer also the question of whether or not the modified equation will alter the other equations of the model, and, if so, how should this occur while preserving the thermodynamic consistency? This is the main goal of this paper.

III. THERMODYNAMICALLY CONSISTENT MODEL

A. Main ideas

In order to eliminate the interface velocity due to curvature, one should eliminate the surface tension energy from the classical phase-field model. However, the elimination of the surface energy should keep the diffuse nature of the interface.

The basic idea that underlies thermodynamic consistency is to rewrite the first principle of thermodynamics by eliminating the surface energy in the membrane problem. Thus, we aim at constructing an energy $E(\varphi, \boldsymbol{\nabla} \varphi)$ which we write in the following form:

$$E(\varphi, \boldsymbol{\nabla} \varphi) = E^{\mathrm{cl}}(\varphi, \boldsymbol{\nabla} \varphi) - E^{\delta}(\varphi, \boldsymbol{\nabla} \varphi), \tag{15}$$

where $E^{\mathrm{cl}}(\varphi, \boldsymbol{\nabla} \varphi)$ is the classical phase-field functional, while $E^{\delta}(\varphi, \boldsymbol{\nabla} \varphi)$ is sought for such that (1) it eliminates the surface energy due to E^{cl} and (2) it does not modify (or only very slightly modifies) the phase-field profile across the interfacial zone. It will be shown later that, from the asymptotic expansion, the model we propose recovers that presented in Ref. [2]. The novelty is that our model preserves thermodynamic consistency.

From Eqs. (8), (9), (14), and (15), and by identification, we deduce that, if the model of Ref. [2] were thermodynamically consistent, the function $E^{\mathrm{cl}}(\varphi,\nabla\varphi)$ would satisfy the following relation:

$$\frac{\partial E^{\delta}}{\partial\varphi}-\nabla\cdot\left(\frac{\partial E^{\delta}}{\partial\nabla\varphi}\right)=\lambda|\nabla\varphi|\nabla\cdot\left(\frac{\nabla\varphi}{|\nabla\varphi|}\right). \tag{16}$$

We will show later that no function $E^{\delta}(\varphi,\nabla\varphi)$ can satisfy this equation, while respecting the above two requirements 1 and 2

Since

$$\frac{\partial|\nabla\varphi|}{\partial\nabla\varphi}=\frac{\nabla\varphi}{|\nabla\varphi|}, \tag{17}$$

Eq. (16) suggests that E^{δ} should be sought for in the following form:

$$E^{\delta}(\varphi,\nabla\varphi)=\beta(\varphi,|\nabla\varphi|)|\nabla\varphi|. \tag{18}$$

At this point, the introduction of the function β is somewhat arbitrary and does not introduce any restriction in the analysis.

In the following section, we derive the general condition that the function $\beta(\varphi,|\nabla\varphi|)$ must satisfy so that the energy E^{δ} satisfies the two requirements listed above.

B. Some preliminary general results

For the sake of simplicity, let us first consider the case of a planar interface at equilibrium. This also corresponds to the zeroth-order approximation in the asymptotic analysis of the Allen-Cahn equation, which is the reference of our analysis. We shall see later how the results derived in this section can be extended to a general curved geometry.

Unlike the classical phase-field model that automatically induces a surface energy [as expressed by Eq. (5)], the introduction of an additional contribution E^{δ} offers an extra freedom that is chosen by the demand that the surface energy vanishes. We must thus, accordingly, impose

$$\int_{-\infty}^{+\infty}[E^{\mathrm{cl}}(\varphi,\varphi')-E^{\delta}(\varphi,\varphi')]dr=0. \tag{19}$$

This is the very definition of the surface energy corresponding to the modified model. We require this energy to be zero by definition.

Using Eqs. (1), (5), and (18), the above requirement is equivalent to the following condition for $\beta(\varphi,|\nabla\varphi|)$:

$$\int_{-\infty}^{+\infty}\beta(\varphi,\varphi')\varphi' dr=\gamma, \tag{20}$$

where we recall that γ is the surface energy associated with the classical phase-field model, i.e., corresponding to the sole energy E^{cl}. By the change of variable $\varphi(r)$, this condition can be replaced by the following condition:

$$\int_{\varphi_0}^{\varphi_1}\beta(\varphi,\varphi'(\varphi))d\varphi=\gamma \tag{21}$$

It is worth noting that many functions satisfy this condition. A particular choice is $\beta=\gamma/(\varphi_1-\varphi_0)$. This is the simplest function that eliminates the surface tension energy from our model.

We now turn our attention to the condition that the phase-field profile should be close to that of the classical phase-field model. For that purpose, we assume that

$$\frac{\partial E^{\delta}}{\partial\varphi}-\nabla\cdot\left(\frac{\partial E^{\delta}}{\partial\nabla\varphi}\right)=\psi(\varphi,|\nabla\varphi|)\nabla\cdot\left(\frac{\nabla\varphi}{|\nabla\varphi|}\right), \tag{22}$$

where the function $\psi(\varphi,|\nabla\varphi|)$ is general and not necessarily equal to $\lambda|\nabla\varphi|$ as in the "ideal" case (16). The general equilibrium condition (2) applied to a planar interface and to the energy (15) with (18) shows that the equilibrium profile obeys the following equation:

$$\frac{dW}{d\varphi}-\lambda\varphi''+\psi(\varphi,\varphi')\left(\frac{\varphi'}{\varphi'}\right)'=0. \tag{23}$$

The last term of the above equation vanishes for any function $\psi(\varphi,\varphi')$, so that the equilibrium profile satisfies the following differential equation:

$$\frac{dW}{d\varphi}-\lambda\varphi''=0. \tag{24}$$

This differential equation is exactly that of the classical phase-field model (3).

This result shows that, if the variational derivative of E^{δ} is of the form (22), the equilibrium profile of the phase field across a planar interface is not modified compared to that of the classical phase-field model. We must now show which condition should $\beta(\varphi,|\nabla\varphi|)$ satisfy so that the variational derivative of (sought after) E^{δ} takes the form (22).

Using the general form (18) for E^{δ}, one has

$$\frac{\partial E^{\delta}}{\partial\varphi}-\nabla\cdot\left(\frac{\partial E^{\delta}}{\partial\nabla\varphi}\right)=\frac{\partial\beta}{\partial\varphi}|\nabla\varphi|-\nabla\left(\frac{\partial\beta}{\partial|\nabla\varphi|}|\nabla\varphi|+\beta\right)\cdot\frac{\nabla\varphi}{|\nabla\varphi|}-\left(\frac{\partial\beta}{\partial|\nabla\varphi|}|\nabla\varphi|+\beta\right)\nabla\cdot\left(\frac{\nabla\varphi}{|\nabla\varphi|}\right). \tag{25}$$

By identification of the last term with the right-hand side of Eq. (16), one would impose

$$\frac{\partial\beta}{\partial|\nabla\varphi|}|\nabla\varphi|+\beta=\lambda|\nabla\varphi|\Leftrightarrow\beta(|\nabla\varphi|)=\frac{\lambda}{2}|\nabla\varphi|. \tag{26}$$

However, with this dependence, the second term of the right-hand side of Eq. (25) would not vanish and the condition (16) would not be satisfied. Moreover, with this solution, the energy of the system would simply be $E=W(\varphi)$ and the interface would no longer be diffuse.

This is the main argument to justify that, even though this solution appears to be the most intuitive, it is not possible that the function β depends on $|\nabla\varphi|$. If the function β depends only on φ, the expression (25) for the functional derivative of E^{δ} reads

$$\frac{\partial E^{\delta}}{\partial \varphi} - \boldsymbol{\nabla} \cdot \left(\frac{\partial E^{\delta}}{\partial \boldsymbol{\nabla} \varphi} \right) = \beta(\varphi) \boldsymbol{\nabla} \cdot \left(\frac{\boldsymbol{\nabla} \varphi}{|\boldsymbol{\nabla} \varphi|} \right). \tag{27}$$

By comparing this expression with the "ideal" expression (16) sought for, one sees that these expressions are very similar. In particular, they are both proportional to the interface curvature $\boldsymbol{\nabla} \cdot (\boldsymbol{\nabla} \varphi / |\boldsymbol{\nabla} \varphi|)$, which is the most important characteristic (we showed in Sec. II B that we seek to eliminate a contribution of the interface velocity proportional to this curvature). For the two expressions to be equivalent, one needs a relation between φ and $|\boldsymbol{\nabla} \varphi|$ such that

$$\beta(\varphi^{\mathrm{eq}}(x)) = \lambda |\boldsymbol{\nabla} \varphi|^{\mathrm{eq}}(x). \tag{28}$$

It is worth emphasizing that this relation is not in contradiction with our previous result stating that β cannot depend on the variable $(\boldsymbol{\nabla} \varphi)$. Indeed, this latter result is functionnal while the above relation follows from minimization of the energy, which leads naturally to a relation between the profile $\varphi^{\mathrm{eq}}(x)$ and $|\boldsymbol{\nabla} \varphi|^{\mathrm{eq}}(x)$ as in Eq. (4).

In the following section, we propose a model that satisfies this requirement.

C. Energy of the system

In the thermodynamic model, φ and $|\boldsymbol{\nabla} \varphi|$ are considered as independent variables. However, we showed in Sec. III B that, for our model, the relation (28) should be satisfied. Such a relation is not possible in general. However, for a planar interface at equilibrium, such a relation exists, as shown by Eq. (4). Based on this result it is appealing to suggest

$$\beta(\varphi) = \sqrt{2 \lambda W(\varphi)} \tag{29}$$

and

$$E^{\delta} = \sqrt{2 \lambda W(\varphi)} |\boldsymbol{\nabla} \varphi|. \tag{30}$$

The full energy thus assumes the following form:

$$E(\varphi, \boldsymbol{\nabla} \varphi) = W(\varphi) + \frac{\lambda}{2} (\boldsymbol{\nabla} \varphi)^2 - \sqrt{2 \lambda W(\varphi)} |\boldsymbol{\nabla} \varphi|. \tag{31}$$

From Eqs. (27) and (29), one has

$$\frac{\delta E^{\delta}}{\delta \varphi} = \sqrt{2 \lambda W(\varphi)} \boldsymbol{\nabla} \cdot \left(\frac{\boldsymbol{\nabla} \varphi}{|\boldsymbol{\nabla} \varphi|} \right). \tag{32}$$

We showed in Sec. III B that, since the expression (32) is of the general form (22), the phase-field profile across a planar interface is not modified compared to the classical phase-field model.

Moreover, the condition (21) to eliminate the surface tension energy yields

$$\gamma = \int_{\varphi_0}^{\varphi_1} \sqrt{2 \lambda W(\varphi)} d\varphi. \tag{33}$$

This condition is indeed satisfied since it corresponds to the definition of the surface tension energy of the classical phase-field model [see Eq. (7)].

Finally, we emphasize that the key relation on which our model is based is the following approximation that is stricly an equality for a planar interface at equilibrium:

$$\sqrt{2 \lambda W(\varphi)} \simeq \lambda |\boldsymbol{\nabla} \varphi|. \tag{34}$$

This approximation allows us to make the model of Ref. [2] thermodynamically consistent.

Since the asymptotic analysis shows that, at the dominant order, the phase-field profile across a locally spherical interface is that of a planar interface at equilibrium, i.e., where the relation (34) is actually an equality, the thermodynamic model proposed is expected to be equivalent to that of Folch *et al.* [2] at the dominant order. More precisely, the expansion at order one shows that the interface velocity depends on the phase-field profile at order zero. Since the latter is the same in our model as in that of Ref. [2], it is expected that the dominant interface velocity is nil in both models. However, at higher orders, the two models are expected to be different because the relation (34) is no longer an equality at these orders. This is why the asymptotic analysis of our model is developed up to the second order (see Sec. IV).

D. Dynamic model

The general Allen-Cahn equation is given by Eq. (8). In the particular case of our model where the expression for the free energy of the system is given by Eq. (31), it is straightforward to show that

$$\frac{\partial \varphi}{\partial t} = -\kappa \left[\frac{dW}{d\varphi} - \lambda \nabla^2 \varphi + \sqrt{2 \lambda W(\varphi)} \boldsymbol{\nabla} \cdot \left(\frac{\boldsymbol{\nabla} \varphi}{|\boldsymbol{\nabla} \varphi|} \right) \right]. \tag{35}$$

As desired, this equation is very similar to the equation of the model of Folch *et al.* [2] [see Eq. (14)]: the term $\lambda |\boldsymbol{\nabla} \varphi|$ of the latter model is simply replaced by $\sqrt{2 \lambda W(\varphi)}$ in our model.

IV. ASYMPTOTIC ANALYSIS

A. Nondimensional form of the Allen-Cahn equation

We recall that the equation studied in Ref. [2] is the following:

$$\epsilon^2 \frac{\partial \varphi}{\partial t} = f(\varphi) + \epsilon^2 \nabla^2 \varphi - \epsilon^2 |\boldsymbol{\nabla} \varphi| \boldsymbol{\nabla} \cdot \left(\frac{\boldsymbol{\nabla} \varphi}{|\boldsymbol{\nabla} \varphi|} \right). \tag{36}$$

For the sake of comparison with the above equation, it is convenient to write our Allen-Cahn equation in a nondimensional form.

We define the nondimensional energy $w(\varphi)$ such that

$$\frac{1}{\lambda} W(\varphi) = \frac{1}{h^2} w(\varphi) \tag{37}$$

where h is the interface thickness. The differential operator $\boldsymbol{\nabla}$ is made nondimensionnal through the introduction of an outer length scale L (typically the size of the entity under consideration). The Allen-Cahn equation (35) thus reads

$$-\frac{h^2}{\kappa \lambda} \frac{\partial \varphi}{\partial t} = \frac{dw}{d\varphi} - \epsilon^2 \nabla^2 \varphi + \epsilon \sqrt{2 w(\varphi)} \boldsymbol{\nabla} \cdot \left(\frac{\boldsymbol{\nabla} \varphi}{|\boldsymbol{\nabla} \varphi|} \right). \tag{38}$$

If the time scale t_c is defined by

$$t_c = \frac{L^2}{\kappa\lambda}, \tag{39}$$

one gets

$$-\epsilon^2\frac{\partial\varphi}{\partial t} = \frac{dw}{d\varphi} - \epsilon^2\nabla^2\varphi + \epsilon\sqrt{2w(\varphi)}\,\boldsymbol{\nabla}\cdot\left(\frac{\boldsymbol{\nabla}\varphi}{|\boldsymbol{\nabla}\varphi|}\right). \tag{40}$$

Our scaling is coherent with that of Ref. [2] because

$$\sqrt{2w(\varphi)} \simeq \epsilon|\boldsymbol{\nabla}\varphi|. \tag{41}$$

The details of the asymptotic analysis of the nondimensional Allen-Cahn equation (40) are presented in Appendix A. In the following sections, we recall and discuss the main results.

B. Zeroth-order solution

At zeroth order in ϵ, it is found that the phase field across the interfacial zone satisfies the following differential equation:

$$w'(\bar{\varphi}^{(0)}) - \bar{\varphi}^{(0)}{}_{,\bar{r}\bar{r}} = 0. \tag{42}$$

We have adopted the differentiation convention such that the subscript x designates the derivative with respect to x, and the subscript xx is the second derivative, and so on. This differential equation is that of a planar interface at equilibrium [see Eq. (3)]. By integration and accounting for the matching conditions, one gets

$$w(\bar{\varphi}^{(0)}) = \frac{1}{2}(\bar{\varphi}^{(0)}{}_{,\bar{r}})^2. \tag{43}$$

This equation means that, at the dominant order, the approximation (34) is actually an equality.

C. First-order solution

At first order, it is found that

$$\bar{\varphi}^{(1)}{}_{,\bar{r}\bar{r}} - w''(\bar{\varphi}^{(0)})\bar{\varphi}^{(1)} = \bar{\boldsymbol{v}}_n^{(0)}\bar{\varphi}^{(0)}{}_{,\bar{r}} + \bar{\mathcal{C}}^{(0)}[\bar{\varphi}^{(0)}{}_{,\bar{r}} - \sqrt{2w(\bar{\varphi}^{(0)})}]. \tag{44}$$

Accounting for the zeroth-order solution (43), the last two terms in square brackets of this equation cancel out, so that

$$\bar{\varphi}^{(1)}{}_{,\bar{r}\bar{r}} - w''(\bar{\varphi}^{(0)})\bar{\varphi}^{(1)} = \bar{\boldsymbol{v}}_n^{(0)}\bar{\varphi}^{(0)}{}_{,\bar{r}}. \tag{45}$$

It is worth noting that, with the model of Folch *et al.* [2], the term $\sqrt{2w(\bar{\varphi}^{(0)})}$ of Eq. (44) is replaced by $\bar{\varphi}^{(0)}{}_{,\bar{r}}$. In both cases, the consequence is the same: the term proportional to $\bar{\mathcal{C}}^{(0)}$ vanishes.

The solvability condition of Eq. (45) reads

$$\int_{-\infty}^{+\infty}\bar{\boldsymbol{v}}_n^{(0)}(\bar{\varphi}^{(0)}{}_{,\bar{r}})^2 d\bar{r} = 0, \tag{46}$$

which implies that

$$\bar{\boldsymbol{v}}_n^{(0)} = \boldsymbol{0}. \tag{47}$$

As expected, the interface velocity is nil, as in Ref. [2].

Moreover, in Appendix A 5, it is shown that

$$\bar{\varphi}^{(1)}(\bar{r}) = 0. \tag{48}$$

D. Second-order solution

At second order, it is found that

$$\bar{\varphi}^{(2)}{}_{,\bar{r}\bar{r}} - w''(\bar{\varphi}^{(0)})\bar{\varphi}^{(2)} = -\bar{\boldsymbol{v}}_n^{(1)}\bar{\varphi}^{(0)}{}_{,\bar{r}} - w'(\bar{\varphi}^{(0)}). \tag{49}$$

The solvability condition of this equation reads

$$\bar{\boldsymbol{v}}_n^{(1)}\int_{-\infty}^{+\infty}(\bar{\varphi}^{(0)}{}_{,\bar{r}})^2 d\bar{r} = \int_{-\infty}^{+\infty}\bar{\varphi}^{(0)}{}_{,\bar{r}}w'(\bar{\varphi}^{(0)})d\bar{r}. \tag{50}$$

Accounting for Eq. (42), one has

$$\bar{\boldsymbol{v}}_n^{(1)} = \boldsymbol{0}. \tag{51}$$

In order to compare this result with those of Ref. [2], we have extended their asymptotic expansion up to second order in ϵ. Surprisingly, the result is identical to ours. This was not expected *a priori* because our model is such that the equality $|\boldsymbol{\nabla}\varphi| = \sqrt{2\lambda W(\varphi)}$ is *a priori* satisfied only at equilibrium of a planar interface, i.e., at zeroth order in ϵ.

The only difference between our model and that of Folch *et al.* [2] is that the term $\epsilon^2|\boldsymbol{\nabla}\varphi|$ in the factor of $\boldsymbol{\nabla}\cdot(\boldsymbol{\nabla}\varphi/|\boldsymbol{\nabla}\varphi|)$ is their nondimensional Allen-Cahn equation is replaced by $\epsilon\sqrt{2w(\varphi)}$ in ours. We thus compare these two expressions. Since both models are equivalent at zeroth order, we use this solution to simplify the expressions.

Using

$$|\boldsymbol{\nabla}\varphi| = \frac{1}{\epsilon}\bar{\varphi}^{(0)}{}_{,\bar{r}} + \bar{\varphi}^{(1)}{}_{,\bar{r}} + O(\epsilon), \tag{52}$$

it is found that

$$\epsilon^2|\boldsymbol{\nabla}\varphi|\,\boldsymbol{\nabla}\cdot\left(\frac{\boldsymbol{\nabla}\varphi}{|\boldsymbol{\nabla}\varphi|}\right) = \epsilon(-\bar{\mathcal{C}}^{(0)}\bar{\varphi}^{(0)}{}_{,\bar{r}}) + \epsilon^2\{-\bar{\mathcal{C}}^{(0)}\bar{\varphi}^{(1)}{}_{,\bar{r}} - \bar{\varphi}^{(0)}{}_{,\bar{r}}[\bar{r}(\bar{\mathcal{C}}^{(0)})^2 + \bar{\mathcal{C}}^{(1)}]\} + O(\epsilon^3), \tag{53}$$

whereas

$$\epsilon\sqrt{2w(\varphi)}\,\boldsymbol{\nabla}\cdot\left(\frac{\boldsymbol{\nabla}\varphi}{|\boldsymbol{\nabla}\varphi|}\right) = \epsilon(-\bar{\mathcal{C}}^{(0)}\bar{\varphi}^{(0)}{}_{,\bar{r}}) + \epsilon^2\left(-\bar{\mathcal{C}}^{(0)}\frac{w'(\bar{\varphi}^{(0)})}{\sqrt{2w(\bar{\varphi}^{(0)})}}\bar{\varphi}^{(1)} - \bar{\varphi}^{(0)}{}_{,\bar{r}}[\bar{r}(\bar{\mathcal{C}}^{(0)})^2 + \bar{\mathcal{C}}^{(1)}]\right) + O(\epsilon^3). \tag{54}$$

Comparing these two expressions shows that the only difference is at second order in ϵ, where the term $\bar{\varphi}^{(1)}{}_{,\bar{r}}$ in the model of Folch *et al.* [2] is replaced by the term $\bar{\varphi}^{(1)}w'(\bar{\varphi}^{(0)})/\bar{\varphi}^{(0)}{}_{,\bar{r}}$ in ours. However, since in both cases it is found that $\bar{\varphi}^{(1)}(\bar{r})=0$, the two models are identical up to second order, which was not expected *a priori*.

E. Comments

We have seen above that to leading order it is legitimate to replace the term $\lambda|\nabla\varphi|$ by $\sqrt{2\lambda W(\varphi)}$, in which case it is possible to define an energy for the system. Our model, while recovering the result of Ref. [2], enjoys thermodynamic consistency. Note that only integral quantities enter our analysis: the definition of surface tension is an integral [see Eq. (5)] and the fact that the interface velocity is nil at first order in ϵ is also an integral condition [see Eqs. (44) and (46)]. Thus, in principle, only a condition on the integral of the energy E^δ could have been sufficient *a priori*. However, what the model does is to eliminate the effect of the surface tension locally and not only globally, which is a stronger result.

To illustrate this point, let us consider a spherical interface. We expect that, with the classical phase-field model, it is not possible to get an equilibrium profile, since the system constantly evolves to minimize its surface energy (the velocity never vanishes as long as the curvature is not nil). On the contrary, with the model of Ref. [2] as well as with ours, it is possible to get an equilibrium state for any radius of curvature. To see this, we study the equilibrium for a sphere. At equilibrium we have $(\partial\varphi/\partial t)=0$, so that the Allen-Cahn equation (35) degenerates to

$$\frac{dW}{d\varphi} - \lambda\frac{d^2\varphi}{dr^2} = \frac{2}{r}\left(\lambda\frac{d\varphi}{dr} - \sqrt{2\lambda W(\varphi)}\right), \tag{55}$$

where r is the radial coordinate.

The last term of this equation is absent in the classical phase-field model. This implies absence of equilibrium. Indeed, without that term, we have seen that we have a nonzero velocity. In the model of Ref. [2], the last term of this equation is replaced by $\lambda(d\varphi/dr)$ so that the right-hand side vanishes. Thus, the differential equation obtained is exactly that of a planar interface and (i) equilibrium is possible and (ii) the phase-field profile is thus identical to that of a planar interface[3]. In our model, the argument is more subtle, albeit the implication is the same: if the left-hand side is nil, then the right-hand side is nil as well.

In the simplest case that eliminates the surface tension energy, E^δ is constant: $E^\delta=\gamma/(\varphi_1-\varphi_0)|\nabla\varphi|$ (see Sec. III B). In this case, the elimination of the surface tension is only global and not local, in the sense that only the integral of the energy is nil but not the local energy (i.e., at each point within the interfacial zone). It follows that the above differential equation reads

$$\frac{dW}{d\varphi} - \lambda\frac{d^2\varphi}{dr^2} = \frac{2}{r}\left(\lambda\frac{d\varphi}{dr} - \frac{\gamma}{\varphi_1-\varphi_0}\right). \tag{56}$$

Now it is less obvious that this equation has a solution. Nevertheless, in the particular case where the radius of the inclusion is very large compared to the interface thickness, it is possible to make the approximation $(2/r)\simeq cte$ and it is then possible to apply the solvability condition and to show that an equilibrium solution does exist. However, one sees that the result is less general than in the more complex cases of our model.

This is why it is important to eliminate the surface tension energy "locally" and not only "globally." This follows naturally from our model.

[3]Actually, this is true only if $(d^2\varphi/dr^2)\simeq 0$ at $r=0$, a condition that is indeed satisfied as long as the radius of the interface is somewhat larger than the interface thickness.

V. THERMODYNAMICALLY CONSISTENT PHASE-FIELD MODEL FOR VESICLES

In the absence of a systematic frame, traditional phase-field equations are based on guesses, intuition, and on how an effect may be included while keeping sensible physics. The various phase-field models developed to date, including on vesicles [3], have, nevertheless, several important merits, among which is the fact that they have been able to produce quantitative numerical results. Nevertheless, it is not *a priori* obvious to see the implication of a change of a given equation on the full set of equations if one has in mind the development of models in the spirit of thermodynamics. Therefore, seeking for a thermodynamic consistency of the model is also an interesting way of thinking, since it allows one to keep overall coherence.

Let us turn to the vesicle and membrane problem and write the complete set of equations. Let us first begin with a simple case that includes only the coupling between the phase-field equation and the momentum balance equation in the absence of any curvature energy. The thermodynamically consistent model corresponds to the front-capturing method, as studied in [7] (even though the model studied in [7] is not thermodynamically consistent).

A. Hydrodynamics and surface tension force

The phase-field equation in the presence of a flow is given by

$$\frac{d\varphi}{dt} = -\kappa\left[\frac{dW}{d\varphi} - \lambda\nabla^2\varphi + \sqrt{2\lambda W(\varphi)}\,\nabla\cdot\left(\frac{\nabla\varphi}{|\nabla\varphi|}\right)\right], \tag{57}$$

where $d/dt\equiv(\partial/\partial t+\boldsymbol{u}\cdot\nabla)$ is the material derivative, and where $\boldsymbol{u}$ is the fluid velocity. By virtue of the fact that E depends only on φ and $\nabla\varphi$, it can be shown that the general form of the momentum balance equation reads as follows (see [10,11]):

$$\rho_0\frac{d\boldsymbol{u}}{dt} = -\nabla P - \nabla\cdot\left(\frac{\partial E}{\partial\nabla\varphi}\otimes\nabla\varphi\right) + \nabla\cdot\boldsymbol{\tau}^d, \tag{58}$$

where P is the pressure and $\boldsymbol{\tau}^d$ is the dissipative stress tensor (e.g., the classical Newtonian expression, where a viscosity contrast can be included, as in Refs. [3,4]). It is obvious that the right-hand side of the above equation is the divergence of a stress tensor. In comparison to the usual Newtonian stress tensor, there is an additional contribution coming from the phase-field model (second term on the right-hand side).

As shown in [10] for instance, by scalar multiplication of the above equation $\boldsymbol{u}$, multiplication of the the Allen-Cahn equation (57) by $\tilde{\mu}$, and then addition of the two equations thus obtained, one gets the following inequality:

$$\frac{d}{dt}\int_V \left(E(\varphi,\nabla\varphi)+\frac{\rho_0 \boldsymbol{u}^2}{2}\right)dV<0, \tag{59}$$

which is nothing but an expression of the second law of thermodynamics. In the particular case where the expression for $E(\varphi,\nabla\varphi)$ is given by Eq. (31), one gets

$$\rho_0\frac{d\boldsymbol{u}}{dt}=-\nabla P-\nabla\cdot\left[\left(\lambda-\frac{\sqrt{2\lambda W(\varphi)}}{|\nabla\varphi|}\right)\nabla\varphi\otimes\nabla\varphi\right]+\nabla\cdot\boldsymbol{\tau}^d. \tag{60}$$

Recalling the approximation (34), one has

$$\lambda-\frac{\sqrt{2\lambda W(\varphi)}}{|\nabla\varphi|}\simeq 0, \tag{61}$$

which implies that the surface tension force corresponding to the proposed model is approximately nil. Thus, the model proposed induces approximately no surface tension force for any configuration. However, a surface tension force (as well as a surface tension energy) may show up when the profile of the phase field across the interface is far from an equilibrium profile.

It is worth noting that the momentum balance equation (58) has the following equivalent potential form (following [8]):

$$\rho_0\frac{d\boldsymbol{u}}{dt}=-\nabla\tilde{P}+\tilde{\mu}\nabla\varphi+\nabla\cdot\boldsymbol{\tau}^d, \tag{62}$$

where $\tilde{P}=P+E$. This form has proven to be convenient for the development of numerical schemes as shown in [8,9]. In the classical phase-field model, $\tilde{\mu}$ is proportional to the interface curvature $\mathcal{C}$. However, in our model, $\tilde{\mu}$ is almost nil, owing to the fact that we have subtracted the surface energy contribution.

B. Model for vesicles and membranes

In this section, we present a thermodynamically consistent model for vesicles. Vesicle membranes have two important physical characteristics. First, the membrane is *locally incompressible*, in the sense that its local surface area is constant in the course of time. Second, it is endowed with a surface *curvature energy* proportional to $(\mathcal{C}-\mathcal{C}_0)^2$, where $\mathcal{C}_0$ is a constant called the spontaneous curvature. Due to the fact that the membrane does not exchange mass with the solution, the notion of surface energy (as for a droplet) does not make sense (the absence of a chemical potential from an underlying reservoir). The surface energy does not enter (but a tension, due to external stretching for example, may be considered in general; this is not dealt with here). In [10,11], it is shown that the incompressibility constraint and the curvature energy can be accounted for in a thermodynamically consistent phase-field approach. In Sec. III, we showed that the surface tension energy embedded in the classical phase-field model can be eliminated in the framework of a thermodynamically consistent approach.

In this section, we present a full phase-field model of vesicles and membranes that accounts for the membrane incompressibility and for a curvature energy and that does not exhibit any surface tension effect.

The energy of the model is the sum of three contributions:

$$E=E_{\mathrm{dif}}+E_{\mathrm{inc}}+E_{\mathrm{curv}}, \tag{63}$$

where E_{dif} is related to the diffuse interface model with no surface tension, E_{inc} accounts for the membrane incompressibility, and E_{curv} accounts for the curvature energy of the membrane. Their respective expression are the following:

$$E_{\mathrm{dif}}(\varphi,|\nabla\varphi|)=W(\varphi)+\frac{\lambda}{2}(\nabla\varphi)^2-\sqrt{2\lambda W(\varphi)}|\nabla\varphi|, \tag{64}$$

$$E_{\mathrm{inc}}(|\nabla\varphi|,\sigma)=\frac{\theta}{2}(|\nabla\varphi|-\sigma)^2, \tag{65}$$

where θ is the stretching modulus of the membrane and σ represents the local surface area per unit volume at equilibrium (see Ref. [10] for more details); and

$$E_{\mathrm{curv}}(|\nabla\varphi|,\mathcal{C})=\frac{\alpha}{2}(\mathcal{C}-\mathcal{C}_0)^2|\nabla\varphi|, \tag{66}$$

where α is the rigidity of the membrane.

We introduce the tensionlike field ζ defined by (see [10])

$$\zeta=-\theta(\sigma-|\nabla\varphi|). \tag{67}$$

We recall that, as shown in Ref. [10], the surface area of the membrane is conserved and is thus locally incompressible in the limit $\theta\to\infty$.

We then introduce the three corresponding generalized chemical potentials defined, respectively, as follows:

$$\tilde{\mu}_{\mathrm{dif}}=\frac{dW}{d\varphi}-\lambda\nabla^2\varphi+\sqrt{2\lambda W(\varphi)}\mathcal{C}, \tag{68}$$

$$\tilde{\mu}_{\mathrm{inc}}=-\nabla\cdot(\zeta\boldsymbol{n}), \tag{69}$$

$$\tilde{\mu}_{\mathrm{curv}}=-\frac{\alpha}{2}\{(\mathcal{C}-\mathcal{C}_0)[-\mathcal{C}(\mathcal{C}+\mathcal{C}_0)+4H]-2\nabla_s\cdot(\nabla_s\mathcal{C})\}, \tag{70}$$

where $\boldsymbol{n}$ represents the unit vector normal to the interface, $\mathcal{C}$ its mean curvature, and H its Gauss curvature. They are defined by

$$\boldsymbol{n}=\frac{\nabla\varphi}{|\nabla\varphi|}, \tag{71}$$

$$\mathcal{C}=\nabla\cdot\boldsymbol{n}, \tag{72}$$

$$H=\frac{1}{2}[(\nabla_s\cdot\boldsymbol{n})^2-\nabla_s\boldsymbol{n}:\nabla_s\boldsymbol{n}]. \tag{73}$$

The system of equations of the model is the following:

$$\nabla\cdot\boldsymbol{u}=0, \tag{74}$$

$$\frac{1}{\theta}\frac{d\zeta}{dt}=(\boldsymbol{I}-\boldsymbol{n}\otimes\boldsymbol{n}):\nabla\boldsymbol{u}+\boldsymbol{n}\cdot\nabla\left(\frac{d\varphi}{dt}\right), \tag{75}$$

$$\frac{d\varphi}{dt} = -\kappa(\tilde{\mu}_{\text{dif}} + \tilde{\mu}_{\text{inc}} + \tilde{\mu}_{\text{curv}}), \tag{76}$$

$$\rho_0 \frac{d\boldsymbol{u}}{dt} = -\boldsymbol{\nabla} P + (\tilde{\mu}_{\text{dif}} + \tilde{\mu}_{\text{inc}} + \tilde{\mu}_{\text{curv}})\boldsymbol{\nabla}\varphi + \boldsymbol{\nabla}\cdot\boldsymbol{\tau}^d, \tag{77}$$

where the simplest expression for $\boldsymbol{\tau}^d$ is that of a Newtonian fluid with a variable dynamic viscosity η,

$$\boldsymbol{\tau}^d = \eta(\varphi)[\boldsymbol{\nabla}\boldsymbol{u} + (\boldsymbol{\nabla}\boldsymbol{u})^T]. \tag{78}$$

It is worth emphasizing that the term in $\boldsymbol{\nabla}\varphi$ on the right-hand side of Eq. (77) can be written as the divergence of the stress tensor $\boldsymbol{\tau} = \boldsymbol{\tau}_{\text{dif}} + \boldsymbol{\tau}_{\text{inc}} + \boldsymbol{\tau}_{\text{curv}}$. The tensor $\boldsymbol{\tau}_{\text{dif}}$ corresponds to the energy discussed in this paper and is given by Eq. (60). The expression for the tensor $\boldsymbol{\tau}_{\text{inc}}$ is given by Eqs. (29) and (31) of Ref. [10]. Finally, the expression for the tensor $\boldsymbol{\tau}_{\text{curv}}$ is given by Eq. (32) of Ref. [11].

Equation (75) is the equation of evolution of the tension-like field that imposes local membrane incompressibility.[4] In this equation, the larger θ is, the better the condition of local incompressibility is satisfied. In the momentum balance equation, three forces normal to the interface appear. The force $(\tilde{\mu}_{\text{dif}}\boldsymbol{\nabla}\varphi)$ is very small because it represents the "surface tension force," which is almost nil (cf. Sec. III). The force $(\tilde{\mu}_{\text{inc}}\boldsymbol{\nabla}\varphi)$ represents the force due to the local incompressibility of the membrane (see [10]). Finally, the force $(\tilde{\mu}_{\text{curv}}\boldsymbol{\nabla}\varphi)$ is the Helfrich force due to the curvature energy of the membrane (see [11]).

The system of equations (74)–(78) is thermodynamically consistent. This can be checked by scalar multiplication of Eq. (77) by $\boldsymbol{u}$ and multiplication of Eq. (76) by $(\tilde{\mu}_{\text{dif}} + \tilde{\mu}_{\text{inc}} + \tilde{\mu}_{\text{curv}})$, and then addition of the equations thus obtained. It is then found that the total energy of the system $\int_V (E + \rho_0 \boldsymbol{u}^2/2)dV$ is a decreasing function of time.

VI. CONCLUSION

This paper constitutes the last of a series of three papers that deal with phase-field modeling of vesicles and membranes that retains thermodynamic consistency. The thermodynamic consistency offers a systematic frame that allows one to introduce different effects in the model. Other features, like permeations, interaction with a concentration field, nondiagonal Onsager terms, and so on, can be treated along the same lines.

In traditional phase-field models (i.e., models that do not consider thermodynamic consistency), it is sometimes found that, due to the freedom of choice of different terms and functions in the model, there is an advantage in bypassing thermodynamic consistency, which imposes a constraint that is too strong. On the one hand, from the conceptual point of view, it is more comforting to have at our disposal a consistent frame in which the various effects are included in a local thermodynamic picture systematically, and then use is made of second law of thermodynamics. The existence of this principle ensures the fact that the physical evolution of the system makes sense. On the other hand, it is likely that the thermodynamic consistency imposes some constraint on numerical schemes, and once the constraints are properly identified, the schemes may exhibit advantages over the usual methods. This point still needs further studies. We hope to report on numerical exploitation of the models in the future.

[4] In Ref. [10], the last term of the right-hand side of Eq. (75) has been omitted. Nevertheless, in Sec. IV, we have shown that with the present model, $d\varphi/dt \simeq 0$ up to second order in ϵ and this term can thus be omitted up to this order.

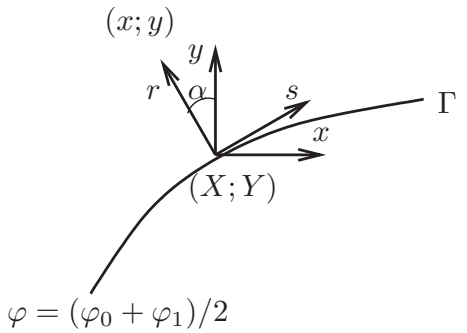

FIG. 2. System of coordinates related to the interface

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APPENDIX A: DETAILS OF THE ASYMPTOTIC ANALYSIS

In this appendix, we provide some details for the asymptotic analysis of the Allen-Cahn equation (35) corresponding to our model:

$$\frac{\partial\varphi}{\partial t} = -\kappa\left[\frac{dW}{d\varphi} - \lambda\nabla^2\varphi + \sqrt{2\lambda W(\varphi)}\,\boldsymbol{\nabla}\cdot\left(\frac{\boldsymbol{\nabla}\varphi}{|\boldsymbol{\nabla}\varphi|}\right)\right]. \tag{A1}$$

1. Geometry and scale factors

Following Folch *et al.* [2], we introduce a frame of reference linked to the interfacial zone. For the sake of simplicity, only the two-dimensional case is studied.

Let us consider a curve defined by an isocontour of the phase field. For this curve to be representative of the interface location, the isovalue of the phase field can be set to $(\varphi_0 + \varphi_1)/2$. This curve is denoted Γ. Let s be the arclength along Γ. The coordinates of a point located on Γ are denoted $(X;Y)$. Let α be the angle going from $\boldsymbol{e}_x$ to $\boldsymbol{e}_s$ (see Fig. 2). By definition, the local curvature $\mathcal{C}$ of Γ is $\mathcal{C} = \alpha_{,s}$.[5] By moving a point along Γ, one has

$$dX = \cos\alpha\, ds,$$

$$dY = \sin\alpha\, ds. \tag{A2}$$

[5] It is worth noting that $\mathcal{C}$ depends only on s.

We now consider a point located out of Γ and we denote its coordinates as $(x;y)$. The coordinates of the closest point located on Γ are denoted $(X;Y)$. One has (see Fig. 2)

$$x(s,r) = X(s) - r\sin\alpha(s),$$

$$y(s,r) = Y(s) + r\cos\alpha(s). \tag{A3}$$

This defines the change of coordinates we consider.

The corresponding scale factors are given by

$$h_r^2 = \left(\frac{\partial x}{\partial r}\right)^2 + \left(\frac{\partial y}{\partial r}\right)^2 = 1,$$

$$h_s^2 = \left(\frac{\partial x}{\partial s}\right)^2 + \left(\frac{\partial y}{\partial s}\right)^2 = (1-r\mathcal{C})^2. \tag{A4}$$

2. Expressions for the differential operators

These are

$$\boldsymbol{\nabla} a = a_{,r}\boldsymbol{e}_r + \frac{1}{1-r\mathcal{C}}a_{,s}\boldsymbol{e}_s, \tag{A5}$$

$$\boldsymbol{\nabla}\cdot\boldsymbol{b} = \boldsymbol{b}^r{}_{,r} + \frac{-\mathcal{C}\boldsymbol{b}^r + \boldsymbol{b}^s{}_{,s}}{1-r\mathcal{C}}, \tag{A6}$$

$$\boldsymbol{\nabla}^2 a = a_{,rr} - \frac{\mathcal{C}}{1-r\mathcal{C}}a_{,r} + \frac{1}{(1-r\mathcal{C})^2}a_{,ss} + \frac{r}{(1-r\mathcal{C})^3}\mathcal{C}_{,s}a_{,s}. \tag{A7}$$

3. Change of coordinates

In the method of matched asymptotic expansions, the domain is divided into an inner zone, where the phase field strongly varies, and an outer zone, where the phase-field variations are weak. Since the nondimensional width of the inner zone is of the order of ϵ (by definition), to study the inner solution, we zoom in on the interfacial region by defining the following new space variable:

$$\bar{r} = \epsilon r, \tag{A8}$$

so that

$$\frac{1}{1-r\mathcal{C}} = 1 + \epsilon\bar{r}\mathcal{C} + \epsilon^2\bar{r}^2\mathcal{C}^2 + O(\epsilon^3). \tag{A9}$$

With this change of variable, one has

$$\boldsymbol{\nabla} a = \frac{1}{\epsilon}\bar{a}_{,\bar{r}}\boldsymbol{e}_{\bar{r}} + \bar{a}_{,s}[1 + \epsilon\bar{r}\mathcal{C} + \epsilon^2\bar{r}^2\mathcal{C}^2 + O(\epsilon^3)]\boldsymbol{e}_s, \tag{A10}$$

$$\boldsymbol{\nabla}\cdot\boldsymbol{b} = \frac{1}{\epsilon}\bar{\boldsymbol{b}}^r{}_{,\bar{r}} + (\bar{\boldsymbol{b}}^s{}_{,s} - \mathcal{C}\bar{\mathbf{b}}^r)[1 + \epsilon\bar{r}\mathcal{C} + \epsilon^2\bar{r}^2\mathcal{C}^2 + O(\epsilon^3)], \tag{A11}$$

$$\nabla^2 a = \frac{1}{\epsilon^2}\bar{a}_{,\bar{r}\bar{r}} - \frac{1}{\epsilon}\mathcal{C}\bar{a}_{,\bar{r}} + (-\bar{r}\mathcal{C}^2\bar{a}_{,\bar{r}} + \bar{a}_{,ss}) + \epsilon(-\bar{r}^2\mathcal{C}^3\bar{a}_{,\bar{r}} + 2\bar{r}\mathcal{C}\bar{a}_{,ss} + \bar{r}\mathcal{C}_{,s}\bar{a}_{,s}) + O(\epsilon^2), \tag{A12}$$

$$\frac{\partial a}{\partial t} = \left.\frac{\partial a}{\partial t}\right|_{x,y=cte} = \frac{da}{dt} - \boldsymbol{v}\cdot\boldsymbol{\nabla} a = \frac{da}{dt} - \boldsymbol{v}_n\left(\frac{1}{\epsilon}\bar{a}_{,\bar{r}}\right) - \boldsymbol{v}_t(\bar{a}_{,s} + \epsilon\bar{r}\mathcal{C}\bar{a}_{,s}) + O(\epsilon^2), \tag{A13}$$

$$\frac{\partial a}{\partial t} = \frac{1}{\epsilon}(-\boldsymbol{v}_n\bar{a}_{,\bar{r}}) + \left(\frac{da}{dt} - \boldsymbol{v}_t\bar{a}_{,s}\right) + \epsilon(-\boldsymbol{v}_t\bar{r}\mathcal{C}\bar{a}_{,s}) + O(\epsilon^2), \tag{A14}$$

$$|\boldsymbol{\nabla} a| = \frac{1}{\epsilon}\bar{a}_{,\bar{r}} + \epsilon\frac{1}{2}\frac{(\bar{a}_{,s})^2}{\bar{a}_{,\bar{r}}} + \epsilon^2\bar{r}\mathcal{C}\frac{(\bar{a}_{,s})^2}{\bar{a}_{,\bar{r}}} + O(\epsilon^4), \tag{A15}$$

$$\frac{\boldsymbol{\nabla} a}{|\boldsymbol{\nabla} a|} = \left(1 - \frac{\epsilon^2}{2}\frac{(\bar{a}_{,s})^2}{(\bar{a}_{,\bar{r}})^2} - \epsilon^3\bar{r}\mathcal{C}\frac{(\bar{a}_{,s})^2}{(\bar{a}_{,\bar{r}})^2} + O(\epsilon^4)\right)\boldsymbol{e}_r + \left(\epsilon\frac{\bar{a}_{,s}}{\bar{a}_{,\bar{r}}} + \epsilon^2\bar{r}\mathcal{C}\frac{\bar{a}_{,s}}{\bar{a}_{,\bar{r}}} + O(\epsilon^4)\right)\boldsymbol{e}_s, \tag{A16}$$

$$\boldsymbol{\nabla}\cdot\left(\frac{\boldsymbol{\nabla} a}{|\boldsymbol{\nabla} a|}\right) = -\mathcal{C} + \epsilon\left[\left(\frac{\bar{a}_{,s}}{\bar{a}_{,\bar{r}}}\right)_{,s} - \frac{1}{2}\left(\frac{(\bar{a}_{,s})^2}{(\bar{a}_{,\bar{r}})^2}\right)_{,\bar{r}} - \bar{r}\mathcal{C}^2\right] + \epsilon^2\left[\left(\bar{r}\mathcal{C}\frac{\bar{a}_{,s}}{\bar{a}_{,\bar{r}}}\right)_{,s} + \bar{r}\mathcal{C}\left(\frac{\bar{a}_{,s}}{\bar{a}_{,\bar{r}}}\right)_{,s} - \left(\bar{r}\mathcal{C}\frac{(\bar{a}_{,s})^2}{(\bar{a}_{,\bar{r}})^2}\right)_{,\bar{r}} + \frac{\mathcal{C}}{2}\frac{(\bar{a}_{,s})^2}{(\bar{a}_{,\bar{r}})^2} - \bar{r}^2\mathcal{C}^3\right] + O(\epsilon^3). \tag{A17}$$

Any function $\bar{\psi}(\bar{r},s)$ is expanded in ϵ as follows:

$$\bar{\psi}(\bar{r},s) = \bar{\psi}^{(0)}(\bar{r},s) + \epsilon\bar{\psi}^{(1)}(\bar{r},s) + \epsilon^2\bar{\psi}^{(2)}(\bar{r},s) + \cdots. \tag{A18}$$

If we apply this expansion to the functions $\sqrt{w(\varphi)}$ and $w'(\varphi)$, we get

$$\sqrt{w(\varphi)} = \sqrt{w(\bar{\varphi}^{(0)})}\left(1 + \epsilon\frac{\bar{\varphi}^{(1)}}{2w(\bar{\varphi}^{(0)})}w'(\bar{\varphi}^{(0)})\right) + O(\epsilon^2), \tag{A19}$$

$$w'(\varphi) = w'(\bar{\varphi}^{(0)}) + \epsilon\bar{\varphi}^{(1)}w''(\bar{\varphi}^{(0)}) + \epsilon^2\left(\bar{\varphi}^{(2)}w''(\bar{\varphi}^{(0)}) + \frac{(\bar{\varphi}^{(1)})^2}{2}w'''(\bar{\varphi}^{(0)})\right) + O(\epsilon^3). \tag{A20}$$

Thus

$$\epsilon^2\nabla^2\varphi = \bar{\varphi}^{(0)}{}_{,\bar{r}\bar{r}} + \epsilon(\bar{\varphi}^{(1)}{}_{,\bar{r}\bar{r}} - \bar{\mathcal{C}}^{(0)}\bar{\varphi}^{(0)}{}_{,\bar{r}}) + \epsilon^2[\bar{\varphi}^{(2)}{}_{,\bar{r}\bar{r}} - \bar{\mathcal{C}}^{(0)}\bar{\varphi}^{(1)}{}_{,\bar{r}} - \bar{r}(\bar{\mathcal{C}}^{(0)})^2\bar{\varphi}^{(0)}{}_{,\bar{r}} + \bar{\varphi}^{(0)}{}_{,ss} - \bar{\mathcal{C}}^{(1)}\bar{\varphi}^{(0)}{}_{,\bar{r}}] + O(\epsilon^3), \tag{A21}$$

$$\boldsymbol{\nabla}\cdot\left(\frac{\boldsymbol{\nabla}\varphi}{|\boldsymbol{\nabla}\varphi|}\right)=-\bar{\mathcal{C}}^{(0)}+\epsilon\left\{\left(\frac{\bar{\varphi}^{(0)}{}_{,s}}{\bar{\varphi}^{(0)}{}_{,\bar{r}}}\right)_{,s}-\frac{1}{2}\left[\left(\frac{\bar{\varphi}^{(0)}{}_{,s}}{\bar{\varphi}^{(0)}{}_{,\bar{r}}}\right)^2\right]_{,\bar{r}}-\bar{r}(\bar{\mathcal{C}}^{(0)})^2-\bar{\mathcal{C}}^{(1)}\right\}+O(\epsilon^2), \tag{A22}$$

$$\epsilon\sqrt{2w(\varphi)}\,\boldsymbol{\nabla}\cdot\left(\frac{\boldsymbol{\nabla}\varphi}{|\boldsymbol{\nabla}\varphi|}\right)=\epsilon[-\bar{\mathcal{C}}^{(0)}\sqrt{2w(\bar{\varphi}^{(0)})}]+\epsilon^2\left(-\bar{\mathcal{C}}^{(0)}\frac{\bar{\varphi}^{(1)}}{\sqrt{2w(\bar{\varphi}^{(0)})}}w'(\bar{\varphi}^{(0)})\right.$$
$$\left.+\sqrt{2w(\bar{\varphi}^{(0)})}\left\{\left(\frac{\bar{\varphi}^{(0)}{}_{,s}}{\bar{\varphi}^{(0)}{}_{,\bar{r}}}\right)_{,s}-\frac{1}{2}\left[\left(\frac{\bar{\varphi}^{(0)}{}_{,s}}{\bar{\varphi}^{(0)}{}_{,\bar{r}}}\right)^2\right]_{,\bar{r}}-\bar{r}(\bar{\mathcal{C}}^{(0)})^2-\bar{\mathcal{C}}^{(1)}\right\}\right)+O(\epsilon^3). \tag{A23}$$

Like any other function, the velocity v is a function of space (and time) and thus has to be expanded in ϵ:[6]

$$\boldsymbol{v}=\bar{\boldsymbol{v}}^{(0)}+\epsilon\bar{\boldsymbol{v}}^{(1)}+O(\epsilon^2). \tag{A24}$$

Equation (A14) thus yields

$$\epsilon^2\frac{\partial\varphi}{\partial t}=\epsilon(-\bar{\boldsymbol{v}}_n^{(0)}\bar{\varphi}^{(0)}{}_{,\bar{r}})+\epsilon^2\left(\frac{d\bar{\varphi}^{(0)}}{dt}-\bar{\boldsymbol{v}}_n^{(0)}\bar{\varphi}^{(1)}{}_{,\bar{r}}-\bar{\boldsymbol{v}}_t^{(0)}\bar{\varphi}^{(0)}{}_{,s}\right.$$
$$\left.-\bar{\boldsymbol{v}}_n^{(1)}\bar{\varphi}^{(0)}{}_{,\bar{r}}\right)+O(\epsilon^3). \tag{A25}$$

The expansions in ϵ derived above are injected in the Allen-Cahn equation (A1) to get the corresponding equations at different orders in ϵ.

4. Zeroth-order solution

At zeroth order in ϵ, Eq. (A1) yields

$$w'(\bar{\varphi}^{(0)})-\bar{\varphi}^{(0)}{}_{,\bar{r}\bar{r}}=0. \tag{A26}$$

By integration and accounting for the matching conditions, we get

$$w(\bar{\varphi}^{(0)})=\frac{1}{2}(\bar{\varphi}^{(0)}{}_{,\bar{r}})^2. \tag{A27}$$

Moreover, it can be shown that $\bar{\varphi}^{(0)}$ is independent of s, so that $\bar{\varphi}^{(0)}{}_{,s}=0$ and $\bar{\varphi}^{(0)}{}_{,ss}=0$.

5. First-order solution

At first order in ϵ, Eq. (A1) yields

$$\bar{\varphi}^{(1)}{}_{,\bar{r}\bar{r}}-w''(\bar{\varphi}^{(0)})\bar{\varphi}^{(1)}=\bar{\boldsymbol{v}}_n^{(0)}\bar{\varphi}^{(0)}{}_{,\bar{r}}+\bar{\mathcal{C}}^{(0)}[\bar{\varphi}^{(0)}{}_{,\bar{r}}-\sqrt{2w(\bar{\varphi}^{(0)})}]. \tag{A28}$$

Because of the zeroth-order solution, the last term of this equation cancels out:

$$\bar{\varphi}^{(1)}{}_{,\bar{r}\bar{r}}-w''(\bar{\varphi}^{(0)})\bar{\varphi}^{(1)}=\bar{\boldsymbol{v}}_n^{(0)}\bar{\varphi}^{(0)}{}_{,\bar{r}}. \tag{A29}$$

The solvability condition reads

$$\int_{-\infty}^{+\infty}\bar{\boldsymbol{v}}_n^{(0)}(\bar{\varphi}^{(0)}{}_{,\bar{r}})^2d\bar{r}=\boldsymbol{0}, \tag{A30}$$

which implies that

$$\bar{\boldsymbol{v}}_n^{(0)}=\boldsymbol{0}. \tag{A31}$$

We now seek for $\bar{\varphi}^{(1)}(\bar{r})$, the solution of the following differential equation:

$$\bar{\varphi}^{(1)}{}_{,\bar{r}\bar{r}}-w''(\bar{\varphi}^{(0)})\bar{\varphi}^{(1)}=0. \tag{A32}$$

It is easy to check that $\bar{\varphi}^{(0)}{}_{,\bar{r}}$ and $\bar{\varphi}^{(0)}{}_{,\bar{r}}\int d\bar{r}/(\bar{\varphi}^{(0),\bar{r}})^2$ are two independent solutions of this equation. Thus, the general solution of this differential equation reads

$$\bar{\varphi}^{(1)}=C_1\bar{\varphi}^{(0)}{}_{,\bar{r}}+C_2\bar{\varphi}^{(0)}{}_{,\bar{r}}\int\frac{d\bar{r}}{(\bar{\varphi}^{(0)}{}_{,\bar{r}})^2}, \tag{A33}$$

where C_1 and C_2 are two constants that have to be determined.

It can be shown that, provided that

$$w(\varphi)\sim(\varphi-\varphi^{\pm\infty})^n\quad\text{for }\varphi\sim\varphi^{\pm\infty} \tag{A34}$$

with $n\geqslant 2$, the second particular solution does not satisfy the matching conditions that $\bar{\varphi}^{(1)}$ must satisfy. Therefore, $C_2=0$.

Moreover, the first particular solution is such that it is different from $(\varphi_0+\varphi_1)/2$ at $\bar{r}=0$. Therefore, $C_1=0$.

Therefore

$$\bar{\varphi}^{(1)}(\bar{r})=0. \tag{A35}$$

6. Second-order solution

At second order in ϵ, Eq. (A1) yields

$$\bar{\varphi}^{(2)}{}_{,\bar{r}\bar{r}}-w''(\bar{\varphi}^{(0)})\bar{\varphi}^{(2)}=-\bar{\boldsymbol{v}}_n^{(1)}\bar{\varphi}^{(0)}{}_{,\bar{r}}+\frac{(\bar{\varphi}^{(1)}{}_{,\bar{r}})^2}{2}w'''(\bar{\varphi}^{(0)})$$
$$+\bar{\mathcal{C}}^{(0)}\bar{\varphi}^{(1)}{}_{,\bar{r}}-\bar{\varphi}^{(0)}{}_{,ss}-\bar{\mathcal{C}}^{(0)}w'(\bar{\varphi}^{(0)})\frac{\bar{\varphi}^{(1)}}{\bar{\varphi}^{(0)}{}_{,\bar{r}}}. \tag{A36}$$

It is worth noting that this differential equation in $\bar{\varphi}^{(2)}$ has the same structure as that in $\bar{\varphi}^{(1)}$. The only difference comes from the expression for the right-hand side.

[6] This was not considered by Folch *et al.* [2].

The solvability condition reads

$$\bar{\boldsymbol{v}}_n^{(1)} \int_{-\infty}^{+\infty} (\bar{\varphi}^{(0)}{}_{,\bar{r}})^2 d\bar{r} = \int_{-\infty}^{+\infty} \bar{\varphi}^{(0)}{}_{,\bar{r}} \left(-\frac{(\bar{\varphi}^{(1)}{}_{,\bar{r}})^2}{2} w'''(\bar{\varphi}^{(0)}) - \bar{C}^{(0)} \bar{\varphi}^{(1)}{}_{,\bar{r}} + \bar{C}^{(0)} w'(\bar{\varphi}^{(0)}) \frac{\bar{\varphi}^{(1)}}{\bar{\varphi}^{(0)}{}_{,\bar{r}}} \right) d\bar{r}. \tag{A37}$$

Because $\bar{\varphi}^{(1)}(\bar{r})=0$, one has

$$\bar{\boldsymbol{v}}_n^{(1)} = \mathbf{0}. \tag{A38}$$

The differential equation that $\bar{\varphi}^{(2)}$ must satisfy is the following:

$$\bar{\varphi}^{(2)}{}_{,\bar{r}\bar{r}} - w''(\bar{\varphi}^{(0)}) \bar{\varphi}^{(2)} = 0. \tag{A39}$$

APPENDIX B: SOLVABILITY CONDITIONS

In this appendix, we derive the solvability conditions at first and second order in ϵ. Let us define the linear operator

$$\mathcal{L}(f) = f_{,\bar{r}\bar{r}} - w''(\bar{\varphi}^{(0)}) f$$

and the dot product

$$f_1 \cdot f_2 = \int_{-\infty}^{+\infty} \mathcal{L}(f_1) f_2 d\bar{r}.$$

It is straightforward to show that

$$\int_{-\infty}^{+\infty} \mathcal{L}(f_2) f_1 d\bar{r} = \int_{-\infty}^{+\infty} \mathcal{L}(f_1) f_2 d\bar{r} + [f_{2,\bar{r}} f_1 - f_2 f_{1,\bar{r}}]_{-\infty}^{+\infty}. \tag{B1}$$

Now, if we apply the above relation with $f_1=\bar{\varphi}^{(0)}{}_{,\bar{r}}$ and $f_2=\bar{\varphi}^{(1)}$, one has $\mathcal{L}(f_1)=0$ and

$$\int_{-\infty}^{+\infty} \mathcal{L}(\bar{\varphi}^{(1)}) \bar{\varphi}^{(0)}{}_{,\bar{r}} d\bar{r} = [\bar{\varphi}^{(1)}{}_{,\bar{r}} \bar{\varphi}^{(0)}{}_{,\bar{r}} - \bar{\varphi}^{(1)} \bar{\varphi}^{(0)}{}_{,\bar{r}\bar{r}}]_{-\infty}^{+\infty}.$$

Now the matching conditions are such that

$$\lim_{\bar{r}\to\pm\infty} \bar{\varphi}^{(0)}{}_{,\bar{r}} = 0,$$

$$\lim_{\bar{r}\to\pm\infty} \bar{\varphi}^{(0)}{}_{,\bar{r}\bar{r}} = 0, \tag{B2}$$

$$\lim_{\bar{r}\to\pm\infty} \bar{\varphi}^{(1)} = \lim_{r\to\pm 0} \varphi^{(1)} + \bar{r} \lim_{r\to\pm 0} \varphi^{(0)\prime},$$

$$\lim_{\bar{r}\to\pm\infty} \bar{\varphi}^{(1)}{}_{,\bar{r}} = \lim_{r\to\pm 0} \varphi^{(0)\prime}. \tag{B3}$$

Thus[7]

$$\int_{-\infty}^{+\infty} \mathcal{L}(\bar{\varphi}^{(1)}) \bar{\varphi}^{(0)}{}_{,\bar{r}} d\bar{r} = 0.$$

This relation corresponds to the solvability condition of the inner problem at first order.

We now study the solvability condition corresponding to the inner problem at second order. We apply the relation (B1) with $f_1=\bar{\varphi}^{(0)}{}_{,\bar{r}}$ and $f_2=\bar{\varphi}^{(2)}$. One has $\mathcal{L}(f_1)=0$ and

$$\int_{-\infty}^{+\infty} \mathcal{L}(\bar{\varphi}^{(2)}) \bar{\varphi}^{(0)}{}_{,\bar{r}} d\bar{r} = [\bar{\varphi}^{(2)}{}_{,\bar{r}} \bar{\varphi}^{(0)}{}_{,\bar{r}} - \bar{\varphi}^{(2)} \bar{\varphi}^{(0)}{}_{,\bar{r}\bar{r}}]_{-\infty}^{+\infty}.$$

Now the matching conditions are such that

$$\lim_{\bar{r}\to\pm\infty} \bar{\varphi}^{(2)} = \lim_{r\to\pm 0} \varphi^{(2)} + \bar{r} \lim_{r\to\pm 0} \varphi^{(1)\prime} + \frac{\bar{r}^2}{2} \lim_{r\to\pm 0} \varphi^{(0)\prime\prime},$$

$$\lim_{\bar{r}\to\pm\infty} \bar{\varphi}^{(2)}{}_{,\bar{r}} = \lim_{r\to\pm 0} \varphi^{(1)\prime} + \bar{r} \lim_{r\to\pm 0} \varphi^{(0)\prime\prime}. \tag{B4}$$

Because of the matching conditions (B2) and (B4), one gets

$$\int_{-\infty}^{+\infty} \mathcal{L}(\bar{\varphi}^{(2)}) \bar{\varphi}^{(0)}{}_{,\bar{r}} d\bar{r} = 0.$$

[7] $\bar{\varphi}^{(0)}{}_{,\bar{r}\bar{r}}$ is an exponentially decreasing function, whereas $\bar{\varphi}^{(1)}$ is a linearly increasing function.

[1] G. Tryggvason *et al.*, J. Comput. Phys. **204**, 708 (2001).
[2] R. Folch, J. Casademunt, A. Hernandez-Machado, and L. Ramirez-Piscina, Phys. Rev. E **60**, 1724 (1999); **60**, 1734 (1999).
[3] T. Biben and C. Misbah, Phys. Rev. E **67**, 031908 (2003).
[4] T. Biben, K. Kassner, and C. Misbah, Phys. Rev. E **72**, 041921 (2005).
[5] Q. Du, C. Liu, and X. Wang, J. Comput. Phys. **212**, 757 (2006).
[6] F. Campelo and A. Hernández-Machado, Eur. Phys. J. E **20**, 37 (2006).
[7] Y. Sun and C. Beckermann, J. Comput. Phys. **220**, 626 (2007).
[8] D. Jacqmin, J. Comput. Phys. **155**, 1 (1999).
[9] D. Jamet *et al.*, J. Comput. Phys. **182**, 262 (2002).
[10] D. Jamet and C. Misbah, Phys. Rev. E **76**, 051907 (2007).
[11] D. Jamet and C. Misbah, Phys. Rev. E **78**, 031902 (2007).