Toward a thermodynamically consistent picture of the phase-field model of vesicles: Curvature energy

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We extend our recent work on phase-field model for vesicles [D. Jamet and C. Misbah, Phys. Rev. E **76**, 051907 (2007)]—where only the membrane local incompressibility was treated—to the situation where the bending forces and spontaneous curvature are included. We show how the general phase-field equations can be derived within a thermodynamic consistent picture. We analyze a general form of the bending energy, where the Helfrich bending force is treated as a special case. The dynamical evolution equation derived here for the velocity field allows one to write down a constitutive law of the composite fluid: The ambient fluid plus the membrane. This constitutive law has a viscoelastic form, the viscous part arises from the fluid, while the elastic one represents the action of the membrane. It is shown that the elastic stress tensor is not symmetric, owing to bending torque, inherent to a diffuse membrane model.

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I. INTRODUCTION

Vesicles and, more generally, phospholipid membranes constitute a very active field of research in different disciplines. One major reason is related to the fact that they play an essential role in biology. For example, cytoplasmic membranes contain a large amount of lipid bilayer. Red blood cell membranes may contain up to 75% of phospholipids. In the cytoplasm, submicron vesicles are produced by the Golgi apparatus (lipidic layers assembly) in order to carry proteins from the interior of the cell toward the surrounding environment. The hope is that understanding some basic physical processes of the behavior of pure lipid membranes may help to identify some features encountered in more complex situations.

Giant vesicles (in contrast to submicron vesicles), produced in the laboratory, constitute an attractive model system for the viscous and viscoelastic properties of real cells [1]. For example, several features known for vesicles (such as tank-treading motion under shear flow, tumbling, or more complex oscillatory motion) have been identified on red blood cells as well.

At equilibrium the vesicle shapes are obtained from an energy minimization (the Helfrich bending energy). Contrariwise, under flow the shape results from a subtle interplay between various forces [2]: Hydrodynamic forces, tension field within the membrane, bending energy, etc.

The nonequilibrium problem is highly nonlinear and nonlocal, and in its full generality numerical treatment seems to be necessary in many circumstances. Several numerical tools have been introduced: (i) Methods based on the integral representation, by means of the Green's function technique (valid for Stokes flow) [3]. This technique has been used for capsules [4] (an elastic membrane made of polymers, and considered as a simple model mimicking red blood cells), and vesicles [5,6], (ii) techniques based on dynamically triangulated models [7] or particle-based mesoscale solvent, multiparticle collision dynamics [8], or their combination [9], (iii) phase-field [10–13] and level set models [14], (iv) the so-called immersed boundary method [15], used to model some features of red blood cells [16], (v) Lattice-Boltzmann methods [17].

In this work we are interested in a phase-field approach. This paper is a continuation of a previous work [18] in which we have suggested a formulation which is consistent thermodynamically. There we have treated only the first subtle part of the vesicle problem, namely local membrane incompressibility, and have shown how a phase-field model could be built, but we have completely disregarded bending forces. In this paper we show how to deal with the bending Helfrich forces in a thermodynamically consistent picture.

The scheme of the paper is as follows. In Sec. II we introduce the bending energy and derive some corresponding quantities that will be useful later. In Sec. III we derive the general dynamical equations, discuss the constitutive law, and the symmetry of the stress tensor. Section IV is devoted to a summary and a general discussion. Appendix A contains a technical derivation of the asymmetry of the stress tensor, and Appendix B reports on some surface invariants.

II. BENDING ENERGY

Instead of defining, as is traditionally done, an interface in a sharp manner, the idea of the phase-field approach is to encode the interface position in a rapid variation of a field φ that is defined in the entire space and is function of time. This field takes a constant value in each of the coexisting phases, but it varies quite abruptly from one phase to the other. Typically $\varphi \sim \tanh(r/\sqrt{\lambda})$, where r is the coordinate normal to the interface, and $\sqrt{\lambda}$ is a small parameter representing the width of the interfacial region. All the interfacial properties are weighted by $|\nabla\varphi|$, which is a smeared Dirac

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δ function. With the classical phase-field model, $|\nabla\varphi|^2 \propto [1-\tanh^2(r/\sqrt{\lambda})]$. However, we could construct a phase-field model for which $|\nabla\varphi|^2$ would be, for example, a Gaussian function. What matters is that the physical properties of the membrane are localized and the classical phase-field model considered in this paper fulfills this requirement. In addition, this classical model follows naturally from the Landau-Ginzburg double-well potential. The advantage of the phase-field approach is that the evolution equations are defined in the whole domain without explicit reference to an interface. The virtue of the phase-field approach is that it enables a straightforward numerical implementation, together with a systematic account for topology changes.

The typical bending energy introduced by Helfrich [19] is written in our phase-field approach in terms of its density (energy per unit volume, and not per unit area, as is treated in the sharp boundary limit):

$$E = \frac{\alpha}{2}(\mathcal{C}-\mathcal{C}_0)^2|\nabla\varphi|, \tag{1}$$

where α is the rigidity of the membrane, $\mathcal{C}$ is the mean curvature of the membrane, and $\mathcal{C}_0$ represents the spontaneous curvature. In this work, we consider that $\mathcal{C}_0$ is a constant. Implicit in this assumption is the fact that we assume that the two monolayers form a single entity. It would be an interesting task for future work to study the general case of nonuniform $\mathcal{C}_0$.

It is the term $|\nabla\varphi|$ (that tends toward a Dirac δ function in the limit of an extremely thin boundary) that allows one to pass from an energy per unit area to an energy per unit volume. Note that since we are not interested here in a change of topology, we do not include the classical contribution coming from the Gauss curvature in (1) as introduced by Helfrich [19], since it is a topological invariant, owing to the Gauss-Bonnet theorem [20].

The membrane position corresponds to a particular contour line of the phase-field φ. Thus $\nabla\varphi/|\nabla\varphi|$ represents the normal vector to contour lines. The mean curvature $\mathcal{C}$ is defined by [21]

$$\mathcal{C} = \nabla\cdot\left(\frac{\nabla\varphi}{|\nabla\varphi|}\right). \tag{2}$$

$\mathcal{C}$ is a function of $\nabla\varphi$ and $\nabla\nabla\varphi$. Indeed, expanding expression (2) one can write

$$\mathcal{C}(\nabla\varphi,\nabla\nabla\varphi) = \frac{1}{|\nabla\varphi|}\nabla\nabla\varphi:\left(\boldsymbol{I}-\frac{\nabla\varphi}{|\nabla\varphi|}\otimes\frac{\nabla\varphi}{|\nabla\varphi|}\right), \tag{3}$$

where $\boldsymbol{I}$ is the identity tensor. In a subscripted form, this yields

$$\mathcal{C}(\varphi_{,i},\varphi_{,ij}) = \frac{1}{|\nabla\varphi|}\left(\varphi_{,ij}\delta_{ij}-\varphi_{,ij}\frac{\varphi_{,i}}{|\nabla\varphi|}\frac{\varphi_{,j}}{|\nabla\varphi|}\right). \tag{4}$$

We can thus formally write

$$E = E(\nabla\varphi,\nabla\nabla\varphi). \tag{5}$$

From this point of view, the present model differs from a classical phase-field model where the energy depends on φ and $\nabla\varphi$ only.

We rewrite the Helfrich energy density (1) formally as follows (by showing the independent variables, and their composition):

$$E(\nabla\varphi,\nabla\nabla\varphi) = E[|\nabla\varphi|,\mathcal{C}(\nabla\varphi,\nabla\nabla\varphi)]$$
$$= \frac{\alpha}{2}[\mathcal{C}(\nabla\varphi,\nabla\nabla\varphi)-\mathcal{C}_0]^2|\nabla\varphi|. \tag{6}$$

Actually the three writings (6) correspond to three hierarchies of models. (i) The writing on the left-hand side signifies any general expression that depends on the two arguments. This is the upper hierarchy, referred to as model 1. (ii) The middle writing is a model of lower hierarchy. It shows that $(\nabla\varphi,\nabla\nabla\varphi)$ enter via the mean curvature, while the dependence on the scalar $|\nabla\varphi|$ is arbitrary. This is referred to as model 2. Finally, the writing on the right-hand side is the explicit expression of the Helfrich model, referred to as model 3. It turns out that several results derived here are valid not only for model 3, but also for model 2 and/or model 1. Keeping the general form does not induce any special complication for the derivation. Therefore, we provide the general proof and, when necessary, we explicitly specify the validity of the results with regard to the above three models.

The energy is an isotropic function. Thus, its form should be frame invariant. This constraint reduces the degrees of freedom for the dependence of E on $(\nabla\varphi,\nabla\nabla\varphi)$, even in the most general case, i.e., model 1. Indeed, it can be shown (e.g., Ref. [22]) that an isotropic function of $\nabla\varphi$ and $\nabla\nabla\varphi$ can depend only on the following six scalars: $(\nabla\varphi)^2$, $(\nabla^2\varphi)$, $(\nabla\varphi\cdot\nabla\nabla\varphi\cdot\nabla\varphi)$, $(\nabla\varphi\cdot\nabla\nabla\varphi\cdot\nabla\nabla\varphi\cdot\nabla\varphi)$, $(\nabla\nabla\varphi:\nabla\nabla\varphi)$, and $[(\nabla\nabla\varphi\cdot\nabla\nabla\varphi):\nabla\nabla\varphi]$ [23]. For model 3, it becomes clear that among the six scalars listed above, only $(\nabla\varphi)^2$, $(\nabla^2\varphi)$, and $(\nabla\varphi\cdot\nabla\nabla\varphi\cdot\nabla\varphi)$ will enter [see Eqs. (3) and (6)].

Let us now present some results regarding the bending energy, which we will need later. In the most general case (model 1), the differential of E reads as

$$dE = \frac{\partial E}{\partial\nabla\varphi}\cdot d\nabla\varphi + \frac{\partial E}{\partial\nabla\nabla\varphi}:d\nabla\nabla\varphi. \tag{7}$$

To simplify the notations, we define the vector $\boldsymbol{\phi}$ and the tensor $\mathcal{T}$ as follows:

$$\boldsymbol{\phi} = \frac{\partial E}{\partial\nabla\varphi}, \tag{8}$$

$$\mathcal{T} = \frac{\partial E}{\partial\nabla\nabla\varphi}, \tag{9}$$

so that

$$dE = \boldsymbol{\phi}\cdot d\nabla\varphi + \mathcal{T}:d\nabla\nabla\varphi. \tag{10}$$

Our first aim below is to determine the explicit expressions for $\mathcal{T}$ and $\boldsymbol{\phi}$ defined in Eqs. (8) and (9). For this goal we shall confine ourselves to model 3. However, the derivation of the general phase-field model will be valid for the three models, as long as we are not interested in writing explicitly $\mathcal{T}$ and $\boldsymbol{\phi}$.

A. Expression for $\mathcal{T}$

By definition

$$\mathcal{T}_{ij} = \frac{\partial E}{\partial \varphi_{,ij}} = \frac{\partial E}{\partial \mathcal{C}} \frac{\partial \mathcal{C}}{\partial \varphi_{,ij}}. \tag{11}$$

From Eqs. (4) it is straightforward to show that

$$\frac{\partial \mathcal{C}}{\partial \varphi_{ij}} = \frac{1}{|\boldsymbol{\nabla}\varphi|}\left(\delta_{ij} - \frac{\varphi_{,i}}{|\boldsymbol{\nabla}\varphi|}\frac{\varphi_{,j}}{|\boldsymbol{\nabla}\varphi|}\right) \tag{12}$$

or, in a tensorial form

$$\frac{\partial \mathcal{C}}{\partial \boldsymbol{\nabla}\boldsymbol{\nabla}\varphi} = \frac{1}{|\boldsymbol{\nabla}\varphi|}\left(\boldsymbol{I} - \frac{\boldsymbol{\nabla}\varphi}{|\boldsymbol{\nabla}\varphi|} \otimes \frac{\boldsymbol{\nabla}\varphi}{|\boldsymbol{\nabla}\varphi|}\right). \tag{13}$$

For the particular case of model 3 where the expression for E is given by Eq. (6), one has $(\partial E/\partial \mathcal{C}) = \alpha(\mathcal{C} - \mathcal{C}_0)$, so that

$$\mathcal{T}_{ij} = \alpha(\mathcal{C} - \mathcal{C}_0)\left(\delta_{ij} - \frac{\varphi_{,i}}{|\boldsymbol{\nabla}\varphi|}\frac{\varphi_{,j}}{|\boldsymbol{\nabla}\varphi|}\right) \tag{14}$$

or, in a tensorial form

$$\mathcal{T} = \alpha(\mathcal{C} - \mathcal{C}_0)\left(\boldsymbol{I} - \frac{\boldsymbol{\nabla}\varphi}{|\boldsymbol{\nabla}\varphi|} \otimes \frac{\boldsymbol{\nabla}\varphi}{|\boldsymbol{\nabla}\varphi|}\right). \tag{15}$$

It is worth noting that this expression shows that $\mathcal{T}$ is proportional to the projection operator on the tangent plane to the membrane.

B. Expression for $\boldsymbol{\phi}$

By definition

$$\boldsymbol{\phi} = \frac{\partial E}{\partial \boldsymbol{\nabla}\varphi} = \frac{\partial E}{\partial |\boldsymbol{\nabla}\varphi|}\frac{\partial |\boldsymbol{\nabla}\varphi|}{\partial \boldsymbol{\nabla}\varphi} + \frac{\partial E}{\partial \mathcal{C}}\frac{\partial \mathcal{C}}{\partial \boldsymbol{\nabla}\varphi}. \tag{16}$$

The determination of the expression for $(\partial \mathcal{C}/\partial \varphi_{,i})$ is a little bit technical and is not detailed here. It is found that

$$\frac{\partial \mathcal{C}}{\partial \boldsymbol{\nabla}\varphi} = -\frac{3\mathcal{C}}{|\boldsymbol{\nabla}\varphi|^2}\boldsymbol{\nabla}\varphi + 2\frac{\boldsymbol{\nabla}^2\varphi}{|\boldsymbol{\nabla}\varphi|^3}\boldsymbol{\nabla}\varphi - \frac{2}{|\boldsymbol{\nabla}\varphi|^3}\boldsymbol{\nabla}\boldsymbol{\nabla}\varphi \cdot \boldsymbol{\nabla}\varphi. \tag{17}$$

Moreover, it is easy to show that

$$\frac{\partial |\boldsymbol{\nabla}\varphi|}{\partial \boldsymbol{\nabla}\varphi} = \frac{\boldsymbol{\nabla}\varphi}{|\boldsymbol{\nabla}\varphi|}.$$

Therefore, for the particular case of model 3 where the expression for E is given by Eq. (6), one has

$$\boldsymbol{\phi} = -\frac{\alpha}{2}(\mathcal{C} - \mathcal{C}_0)(5\mathcal{C} + \mathcal{C}_0)\frac{\boldsymbol{\nabla}\varphi}{|\boldsymbol{\nabla}\varphi|} - 2\alpha\frac{\mathcal{C} - \mathcal{C}_0}{|\boldsymbol{\nabla}\varphi|^2}[(\boldsymbol{\nabla}\boldsymbol{\nabla}\varphi) \cdot \boldsymbol{\nabla}\varphi - (\boldsymbol{\nabla}^2\varphi)\boldsymbol{\nabla}\varphi]. \tag{18}$$

III. EQUATIONS OF MOTION

In this section, we determine the form of the evolution equations. These equations will involve, when using the second law of thermodynamics, the expression (1) for the energy. The derivation is first given for the general model 1 corresponding to any function $E(\boldsymbol{\nabla}\varphi, \boldsymbol{\nabla}\boldsymbol{\nabla}\varphi)$. The particular case where the energy is given by (1) is studied in Sec. III C.

A. General case

In this whole section we consider the general model 1. To simplify the analysis, we consider a system of uniform density ρ_0 (for example, we could think of a local density change in the vicinity of the membrane). The velocity field is thus solenoidal. This constraint on the flow is accounted for in the momentum balance equation, as usual, by the pressure. The general form of the equations of motion thus read as

$$\boldsymbol{\nabla} \cdot \boldsymbol{u} = 0, \tag{19}$$

$$\rho_0 \frac{d\boldsymbol{u}}{dt} = -\boldsymbol{\nabla}P + \boldsymbol{\nabla} \cdot \boldsymbol{\tau}, \tag{20}$$

where d/dt is the material derivative and $\boldsymbol{\tau}$ is the stress tensor whose expression must be determined. Note that here we have, in general, a complex fluid, and not a Newtonian fluid. Indeed the stress tensor $\boldsymbol{\tau}$ contains the usual Newtonian contribution, as well as a contribution coming from the membrane, whose expression is to be determined. In our derivation below we shall be able to provide this new constitutive law, which is a class of a general law for a complex fluid.

In addition to these equations, an equation of evolution for φ should be provided. It will be shown in this section that the evolution equations follow from the second law of thermodynamics.

Since the system is supposed to be isothermal, the second law of thermodynamics dictates that the free energy is a nonincreasing function of time, which reads as

$$\frac{d}{dt}\left(E + \rho_0\frac{\boldsymbol{u}^2}{2}\right) = -\boldsymbol{\nabla} \cdot \boldsymbol{q} + \boldsymbol{\nabla} \cdot (\boldsymbol{u} \cdot \boldsymbol{\tau}^T - P\boldsymbol{u}) - \Delta, \tag{21}$$

where $\boldsymbol{q}$ is the heat flux, $\boldsymbol{\tau}$ is the stress tensor, $\boldsymbol{\tau}^T$ its transpose and Δ is the energy dissipation. The first term on the right-hand side of (21) is an energy flux (to be determined). The second term is also an energy flux which is quite usual: It corresponds to the work of pressure as well as the work of viscous and elastic forces, both of them are hidden in the total stress tensor $\boldsymbol{\tau}$. Of course as long as $\boldsymbol{q}$ (as well as $\boldsymbol{\tau}$; but this tensor has a mechanical interpretation so that one can anticipate on the corresponding flux) is not determined there is a certain arbitrariness in the way Eq. (21) is written. Nevertheless, we write the equation in order to comply with common usage of irreversible thermodynamics.

From Eqs. (20) and (21), it is easy to show that

$$\frac{dE}{dt} = -\boldsymbol{\nabla} \cdot \boldsymbol{q} + \boldsymbol{\tau}^T : \boldsymbol{\nabla}\boldsymbol{u} - \Delta. \tag{22}$$

Now, from Eq. (7) we have

$$\frac{dE}{dt} = \boldsymbol{\phi} \cdot \frac{d\boldsymbol{\nabla}\varphi}{dt} + \mathcal{T} : \frac{d\boldsymbol{\nabla}\boldsymbol{\nabla}\varphi}{dt}. \tag{23}$$

We now seek the expressions for $(\boldsymbol{\phi} \cdot d\boldsymbol{\nabla}\varphi/dt)$ and $(\mathcal{T} : d\boldsymbol{\nabla}\boldsymbol{\nabla}\varphi/dt)$ as functions of $(d\varphi/dt)$.

It can be shown that

$$\boldsymbol{\phi}\cdot\frac{d\boldsymbol{\nabla}\varphi}{dt}=\boldsymbol{\nabla}\cdot\left(\boldsymbol{\phi}\frac{d\varphi}{dt}\right)-(\boldsymbol{\nabla}\cdot\boldsymbol{\phi})\frac{d\varphi}{dt}-(\boldsymbol{\nabla}\varphi\otimes\boldsymbol{\phi}):\boldsymbol{\nabla}\boldsymbol{u}, \tag{24}$$

$$\mathcal{T}:\frac{d\boldsymbol{\nabla}\boldsymbol{\nabla}\varphi}{dt}=\boldsymbol{\nabla}\cdot\left(\mathcal{T}\cdot\frac{d\boldsymbol{\nabla}\varphi}{dt}-\frac{d\varphi}{dt}\boldsymbol{\nabla}\cdot\mathcal{T}\right)+\frac{d\varphi}{dt}\boldsymbol{\nabla}\cdot(\boldsymbol{\nabla}\cdot\mathcal{T})+(\boldsymbol{\nabla}\varphi\otimes\boldsymbol{\nabla}\cdot\mathcal{T}-\boldsymbol{\nabla}\boldsymbol{\nabla}\varphi\cdot\mathcal{T}):\boldsymbol{\nabla}\boldsymbol{u}. \tag{25}$$

Equations (23)–(25) entail that

$$\frac{dE}{dt}=\boldsymbol{\nabla}\cdot\left((\boldsymbol{\phi}-\boldsymbol{\nabla}\cdot\mathcal{T})\frac{d\varphi}{dt}+\mathcal{T}\cdot\frac{d\boldsymbol{\nabla}\varphi}{dt}\right)-\frac{d\varphi}{dt}\boldsymbol{\nabla}\cdot(\boldsymbol{\phi}-\boldsymbol{\nabla}\cdot\mathcal{T})-[\boldsymbol{\nabla}\varphi\otimes(\boldsymbol{\phi}-\boldsymbol{\nabla}\cdot\mathcal{T})+\boldsymbol{\nabla}\boldsymbol{\nabla}\varphi\cdot\mathcal{T}]:\boldsymbol{\nabla}\boldsymbol{u}, \tag{26}$$

Substituting this expression for (dE/dt) into Eq. (22) provides the following expression for the dissipation Δ:

$$\Delta=-\boldsymbol{\nabla}\cdot\left(\boldsymbol{q}-(\boldsymbol{\phi}-\boldsymbol{\nabla}\cdot\mathcal{T})\frac{d\varphi}{dt}-\mathcal{T}\cdot\frac{d\boldsymbol{\nabla}\varphi}{dt}\right)+\frac{d\varphi}{dt}\boldsymbol{\nabla}\cdot(\boldsymbol{\phi}-\boldsymbol{\nabla}\cdot\mathcal{T})+[\boldsymbol{\tau}^{T}+\boldsymbol{\nabla}\varphi\otimes(\boldsymbol{\phi}-\boldsymbol{\nabla}\cdot\mathcal{T})+\boldsymbol{\nabla}\boldsymbol{\nabla}\varphi\cdot\mathcal{T}]:\boldsymbol{\nabla}\boldsymbol{u}. \tag{27}$$

In order to guarantee the positivity of Δ, the following choice can be made:

$$\frac{d\varphi}{dt}=\kappa\boldsymbol{\nabla}\cdot(\boldsymbol{\phi}-\boldsymbol{\nabla}\cdot\mathcal{T}), \tag{28}$$

$$\boldsymbol{q}=(\boldsymbol{\phi}-\boldsymbol{\nabla}\cdot\mathcal{T})\frac{d\varphi}{dt}+\mathcal{T}\cdot\frac{d\boldsymbol{\nabla}\varphi}{dt}, \tag{29}$$

$$\boldsymbol{\tau}^{T}=-\boldsymbol{\nabla}\varphi\otimes(\boldsymbol{\phi}-\boldsymbol{\nabla}\cdot\mathcal{T})-\boldsymbol{\nabla}\boldsymbol{\nabla}\varphi\cdot\mathcal{T}+(\boldsymbol{\tau}^{D})^{T},$$

$$\boldsymbol{\tau}=-(\boldsymbol{\phi}-\boldsymbol{\nabla}\cdot\mathcal{T})\otimes\boldsymbol{\nabla}\varphi-\mathcal{T}\cdot\boldsymbol{\nabla}\boldsymbol{\nabla}\varphi+\boldsymbol{\tau}^{D}, \tag{30}$$

where $\boldsymbol{\tau}^{D}=\eta[\boldsymbol{\nabla}\boldsymbol{u}+\boldsymbol{\nabla}\boldsymbol{u}^{T}]$ is the dissipative stress tensor such that $\boldsymbol{\tau}^{D}:\boldsymbol{\nabla}\boldsymbol{u}\geq 0$ (η is the dynamical viscosity of the ambient fluid, which may be taken as a function of φ in order to account, for example, for a viscosity contrast [24]).

Some general remarks are in order. The choice of the fluxes in (28)–(30) is not unique. As discussed, for example, in Ref. [25], different sets of fluxes and forces can represent equally well the system under study. As long as we do not include nondiagonal elements (in the opposite case, the problem may become nontrivial), there is no special care to be taken, since what matters is (besides conservation laws) that the choices comply with the second law of thermodynamics. Our choice of the flux (28) is motivated by the fact that we want our equation to reduce to the Allen-Cahn equation in the traditional situation (say for fluid-fluid interfaces). If another choice for the flux (28) had been made, then this would have affected the definition of $\boldsymbol{q}$. The first two contributions in Eq. (27) (the one appearing as a divergence and the next one) can influence each other if another definition were adopted. As soon as we use an Allen-Cahn type of spirit, then $\boldsymbol{q}$ is fixed. The last contribution in (27) does not interfere with the two other terms, owing to Curie's principle (fluxes and forces of different tensorial character do not couple for an isotropic system).

The expressions (28)–(30) are generalizations of the classical case where the energy depends only on $\boldsymbol{\nabla}\varphi$ and not on $\boldsymbol{\nabla}\boldsymbol{\nabla}\varphi$. Indeed, the evolution equation of φ (28) is a generalization of the classical Allen-Cahn equation, the expression (29) for $\boldsymbol{q}$ is a generalization of the classical interstitial working [26], and the expression (30) for the stress tensor shows that its nondissipative part is a generalization of the classical Korteweg tensor.

However, it is worth pointing out that the energy E^{cl} associated to the classical phase-field model is not accounted for in the above presentation,

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

where $W(\varphi)$ is a double-well function and λ is the capillary coefficient.

This choice is made in order to emphasize the theoretical issues related to the sole Helfrich energy (1). The purpose of the energy E^{cl} is to ensure that the phase-field profile across the interface remains constantly locally as a hyperbolic tangent profile. In the complete model, the energy E^{cl} should be simply added to the Helfrich energy (1), which would then give rise to the classical terms of phase-field models in the Allen-Cahn equation and in the expression for the stress tensor.

B. Analysis of the stress tensor

Relation (30) is the general form of the constitutive law that relates the stress tensor to the velocity field and the phase field. A similar relation has been derived by following another spirit (close to that presented in Ref. [11]) [14]. Note also that in the sharp boundary model, the recent constitutive law derived for vesicles contains the velocity field and the shape function of the vesicle [27]. Here the shape is accounted for by the phase field.

Note that expression (30) contains a dissipative part $\boldsymbol{\tau}^{D}$ (which arises from viscous friction), and a nondissipative one $\boldsymbol{\tau}^{C}$ (arising from membrane forces; it is an elastic type of contribution). Thus the constitutive law (30) is viscoelastic. Finally, as will be shown below, the elastic stress tensor $\boldsymbol{\tau}^{C}$ is nonsymmetric.

Let $\boldsymbol{\tau}^{C}$ denote the stress tensor associated to the bending energy,

$$\boldsymbol{\tau}^{C}=-(\boldsymbol{\phi}-\boldsymbol{\nabla}\cdot\mathcal{T})\otimes\boldsymbol{\nabla}\varphi-\mathcal{T}\cdot\boldsymbol{\nabla}\boldsymbol{\nabla}\varphi. \tag{32}$$

The expression for this stress tensor is not easy to interpret. We thus analyze the corresponding force, i.e., $\boldsymbol{\nabla}\cdot\boldsymbol{\tau}^{C}$. It can straightforwardly be shown that

$$\boldsymbol{\nabla}\cdot\boldsymbol{\tau}^{C}=-\boldsymbol{\nabla}E-[\boldsymbol{\nabla}\cdot(\boldsymbol{\phi}-\boldsymbol{\nabla}\cdot\mathcal{T})]\boldsymbol{\nabla}\varphi. \tag{33}$$

The gradient term of the above expression can be absorbed into the pressure term of the momentum balance equation and is thus not really relevant. The second term shows that the effective force due to the energy is a force normal to the

membrane (i.e., parallel to $\nabla\varphi$). This force, denoted $\boldsymbol{F}^C$, is proportional to $\nabla\cdot(\boldsymbol{\phi}-\nabla\cdot\mathcal{T})$, which is the term that also appears in the Allen-Cahn equation (28). To analyze this term, we seek for an interpretation of it in terms of classical interfacial geometric properties (such as the curvature or the normal to the interface).

C. Particular case of the Helfrich energy

In this section we consider the explicit form of the Helfrich energy. We thus consider that the expression for the energy $E(\nabla\varphi,\nabla\nabla\varphi)$ is given by Eq. (1), which corresponds to model 3. We focus our attention on the term $\nabla\cdot(\boldsymbol{\phi}-\nabla\cdot\mathcal{T})$ that appears in the Allen-Cahn equation as well as in the expression for the interfacial force [see Eqs. (28) and (33)].

After lengthy algebra, it can be shown from the expression (15) of $\mathcal{T}$ that

$$\nabla\cdot\mathcal{T}=\alpha\nabla\mathcal{C}\cdot\left(\boldsymbol{I}-\frac{\nabla\varphi}{|\nabla\varphi|}\otimes\frac{\nabla\varphi}{|\nabla\varphi|}\right)-\alpha\frac{\mathcal{C}-\mathcal{C}_0}{|\nabla\varphi|^2}[\nabla\nabla\varphi\cdot\nabla\varphi-(\nabla^2\varphi)\nabla\varphi]-2\alpha\mathcal{C}(\mathcal{C}-\mathcal{C}_0)\frac{\nabla\varphi}{|\nabla\varphi|}. \tag{34}$$

By using expression (18) for $\boldsymbol{\phi}$, one finds immediately

$$\boldsymbol{\phi}-\nabla\cdot\mathcal{T}=-\frac{\alpha}{2}(\mathcal{C}^2-\mathcal{C}_0^2)\frac{\nabla\varphi}{|\nabla\varphi|}-\alpha\frac{\mathcal{C}-\mathcal{C}_0}{|\nabla\varphi|^2}[\nabla\nabla\varphi\cdot\nabla\varphi-(\nabla^2\varphi)\nabla\varphi]-\alpha\nabla\mathcal{C}\cdot\left(\boldsymbol{I}-\frac{\nabla\varphi}{|\nabla\varphi|}\otimes\frac{\nabla\varphi}{|\nabla\varphi|}\right). \tag{35}$$

The second term of this expression is rather difficult to interpret. We therefore seek for another expression for this term.

After lengthy algebra, one finds that

$$\nabla\nabla\varphi\cdot\nabla\varphi-(\nabla^2\varphi)\nabla\varphi=-|\nabla\varphi|^2\mathcal{C}\frac{\nabla\varphi}{|\nabla\varphi|}+|\nabla\varphi|\nabla|\nabla\varphi|\cdot\left(\boldsymbol{I}-\frac{\nabla\varphi}{|\nabla\varphi|}\otimes\frac{\nabla\varphi}{|\nabla\varphi|}\right). \tag{36}$$

Thus

$$\boldsymbol{\phi}-\nabla\cdot\mathcal{T}=\frac{\alpha}{2}(\mathcal{C}-\mathcal{C}_0)^2\boldsymbol{n}-\frac{\alpha}{|\nabla\varphi|}\nabla[(\mathcal{C}-\mathcal{C}_0)|\nabla\varphi|]\cdot(\boldsymbol{I}-\boldsymbol{n}\otimes\boldsymbol{n}), \tag{37}$$

where $\mathbf{n}$ is the unit normal to the interface defined by

$$\boldsymbol{n}=\frac{\nabla\varphi}{|\nabla\varphi|}. \tag{38}$$

After several algebraic manipulations, one finds that

$$\nabla\cdot(\boldsymbol{\phi}-\nabla\cdot\mathcal{T})=\frac{\alpha}{2}(\mathcal{C}-\mathcal{C}_0)[-\mathcal{C}(\mathcal{C}+\mathcal{C}_0)+4H]-\alpha\nabla_s\cdot(\nabla_s\mathcal{C}), \tag{39}$$

where H is the Gauss curvature defined by

$$H=\frac{1}{2}[(\nabla_s\cdot\boldsymbol{n})^2-\nabla_s\boldsymbol{n}:\nabla_s\boldsymbol{n}]. \tag{40}$$

The expression (39) combined with Eq. (33) gives rise to the following form for the force $\boldsymbol{F}^C$ deriving from the Helfrich energy:

$$\boldsymbol{F}^C=-\frac{\alpha}{2}\{(\mathcal{C}-\mathcal{C}_0)[-\mathcal{C}(\mathcal{C}+\mathcal{C}_0)+4H]-2\nabla_s\cdot(\nabla_s\mathcal{C})\}\nabla\varphi. \tag{41}$$

This latter expression is exactly that found in Ref. [11]. However, there is an important difference. Indeed, the derivation in Ref. [11] is based on an approximation of the force expressed in its divergence form $\nabla\cdot\boldsymbol{\tau}^C$, where $\boldsymbol{\tau}^C$ is given by Eq. (32). This first-order approximation in $\varepsilon=\mathcal{C}\,h$, where h is the equilibrium interface thickness, assumes that the profile of the phase-field across the interface is the equilibrium profile. Here we have proven that the form of the force is the same even far from equilibrium. This means that the total force (before any approximation) derived in [11] [Eq. (5)], can be written as a full divergence of some quantity (the quantity is nothing but $\boldsymbol{\tau}^C$ of the present paper). This has indeed been recently shown [14]. Nevertheless, the present general approach shows that the force can be written as the divergence of a tensor, not only in the particular case of model 3, as considered in [14], but even in the general case of model 1.

Some additional remarks are in order. In our analysis, the interface has an implicit representation through the continuous function $\varphi(\boldsymbol{x},t)$. This function can be any function and not necessarily a function whose variation is of hyperbolic tangent type as in phase-field models: it could be a signed distance function as in the level set method, for instance. In this regard, the mean and Gauss curvatures [28], defined, respectively, by Eqs. (2) and (40), do represent the actual curvatures of the membrane and should not be considered only as approximations. In contrast, in sharp interface models, the representation of the interface is explicit. Thus, in an effort to compare the present model to a sharp interface model, the main issue is to determine whether the displacement of the interface is the same with both models. In our model, the displacement of the interface is accounted for through the equation of evolution of the phase field, i.e., the Allen-Cahn equation (28) [along with Eq. (39)]. Currently, the method used to derive the properties of this equivalent surface of discontinuity is the matched asymptotic method, as used in [11]. However, this study is beyond the purpose of this paper.

D. On the asymmetry of the stress tensor

In this section, we discuss an issue that is triggered by the phase-field approach. Indeed, contrary to the classical description of a Newtonian fluid, the stress tensor $\boldsymbol{\tau}$ is not symmetric. This feature can easily be shown. From basic concepts of physics of continuous media, it is known that this means that internal torque must exist within the system. One is naturally led to ask the following questions: (i) Is it physically relevant and (ii) does one have to account also for the

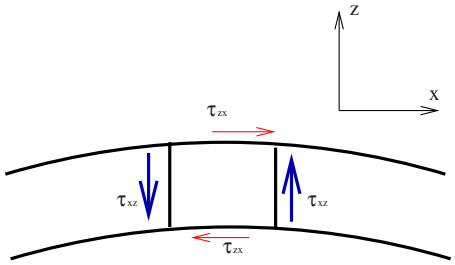

FIG. 1. (Color online) Schematic view of the forces. The force contribution shown in red (horizontal) vanishes while the one in blue (vertical) is nonzero (at least it is different, in a more general situation; see text).

angular momentum balance equation? Indeed, in continuum mechanics, the angular momentum balance equation is in general necessary in order to have a closed set. However, in the particular case where the stress tensor is symmetric, it turns out that this equation is equivalent to the momentum balance equation and thus does not provide any additional information. However, when the stress tensor is not symmetric, the angular momentum balance equation should be accounted for.

Actually, this problem is akin to that of thin plates in elasticity theory. In our case the plate means a fluid membrane which does not resist shear, but which is endowed with a bending energy (as solid plates). The symmetry of the stress tensor does not hold as soon as one takes the limit of a thin plate. To illustrate this let us consider a portion of a thin plate as shown in Fig. 1 (which is macroscopically curved). We consider only the nondiagonal elements of the stress tensor $\boldsymbol{\tau}_{ij}$ (i.e., we assume for simplicity that $\boldsymbol{\tau}_{xx}=\boldsymbol{\tau}_{yy}=\boldsymbol{\tau}_{zz}=0$). Thus, the contribution $\boldsymbol{\tau}_{zx}dA_z$ designates the force which is directed along Ox and is exerted on the upper face [29]. If the plate is in contact with a fluid at rest (this assumption is not restrictive), then $\boldsymbol{\tau}_{zx}=0$. The term $\boldsymbol{\tau}_{xz}dA_x$ is the force exerted on the lateral side, and is directed along z. It is clear that this force (for a curved plate) is nonzero [30], as it is an internal force; it is the stress generated internally by the plate bending. It follows thus that $\boldsymbol{\tau}_{xz}\neq\boldsymbol{\tau}_{zx}$. As a consequence of this, the torque around the Oy axis is nonzero.

IV. CONCLUSION

This paper is a natural continuation of a paper [18] (referred to hereafter as paper I). Here we have extended the model in order to treat bending forces (written in a more general form). In paper I we have treated the local incompressibility of the membrane. The full model equations, including both effects, is obtained just by a simple superposition of the two effects. Namely, the full model is obtained by adding to the bending energy, the energy associated with compressibility, as introduced in paper I. This is why we have not felt it worthwhile to write here the full set of equations. We have now provided a thermodynamically consistent formulation including the two main effects of the physics of membranes: Bending forces and local incompressibility. Actually, in addition to the above (physical) forces, it is well known that the term $(\boldsymbol{\nabla}\varphi)^2$ of the classical phase-field model produces an artificial surface tension. A membrane has intrinsically not such contribution, and one must thus suppress this effect. A possibility has been suggested in Ref. [31] for fluid-fluid [32] interfaces, and has been adopted for vesicles [10,11]. If one must follow the same strategy, this would destroy the thermodynamical consistency of the model. What is needed is to develop a model where the suppression of surface tension originating from the term $(\boldsymbol{\nabla}\varphi)^2$ would be contained in an energy functional. This issue is currently under investigation.

Interesting facts have followed from the present study. First, we have been able to express the total stress tensor (of the fluid and membrane) in a form where viscoelasticity is simply exhibited. In Ref. [14], it has been shown also that the membrane contribution appears in the momentum balance equation under a divergence (of a second-order tensor). Note, however, that the two treatments, the one presented here and that of Ref. [14], have two different starting points. Here, it is postulated from the very beginning that the total stress tensor must be absorbed under a divergence (in order to comply with the general spirit of traditional continuum media), and then the derivation is based on the first and second laws of thermodynamics, together with conservation laws. In Ref. [14], as well as in [11], the membrane force was added in the momentum equation as a functional derivative of the membrane displacement, multiplied by $|\boldsymbol{\nabla}\varphi|$ (a Dirac-like function enforcing localization of the membrane force). It did not appear obvious that the membrane force in [11] could simply be written as a divergence (it has been shown now [14] that this is indeed the case). The present method has the advantage of deriving in a systematic way the composite fluid constitutive law. Indeed its starting point complies with general continuum media theory, in that any kind of membrane force (be it of bending, or of any other nature) is automatically embedded within a divergence of a rank-2 tensor, and then the force is derived *a posteriori* from the first and second laws of thermodynamics.

Second, we have shown that the stress tensor associated with the bending forces is not symmetric. Finally, we hope that after having solved the last issue regarding (artificial) surface tension, we will have at our disposal a thermodynamically consistent phase-field model for studying numerically nonequilibrium problems pertaining to membranes, vesicles, and interactions with flows in various geometries and circumstances.

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APPENDIX A: STUDY OF THE SYMMETRY OF THE CURVATURE STRESS TENSOR

The general expression for the curvature stress tensor $\boldsymbol{\tau}^C$ is given by Eq. (32). In this section, we study the general properties of symmetry of this tensor. We show in particular

that in the case of the Helfrich expression (1), $\boldsymbol{\tau}^C$ is not symmetric.

Our study is restricted to the case where the energy of the system is an isotropic function of the variables $(\boldsymbol{\nabla}\varphi, \boldsymbol{\nabla}\boldsymbol{\nabla}\varphi)$, which limits the dependence of the energy on only six scalars. However, we showed in Sec. II that in the particular case of the Helfrich energy (1), the energy depends only on the three following scalars:

$$a = (\boldsymbol{\nabla}\varphi)^2, \quad b = \boldsymbol{\nabla}^2\varphi, \quad c = \boldsymbol{\nabla}\varphi \cdot \boldsymbol{\nabla}\boldsymbol{\nabla}\varphi \cdot \boldsymbol{\nabla}\varphi. \tag{A1}$$

Therefore, we limit our analysis to the case where the energy is a general function of these three scalars, $E(a,b,c)$.

After some algebraic manipulations, it can be shown that the general expression for the stress tensor $\boldsymbol{\tau}^C$ is the following:

$$\begin{aligned}\boldsymbol{\tau}^C = &\left[-2\frac{\partial E}{\partial a} + \boldsymbol{\nabla}\left(\frac{\partial E}{\partial c}\right)\cdot\boldsymbol{\nabla}\varphi + \frac{\partial E}{\partial c}(\boldsymbol{\nabla}^2\varphi)\right]\boldsymbol{\nabla}\varphi\otimes\boldsymbol{\nabla}\varphi \\ &- \frac{\partial E}{\partial b}\boldsymbol{\nabla}\boldsymbol{\nabla}\varphi - \frac{\partial E}{\partial c}[(\boldsymbol{\nabla}\varphi\cdot\boldsymbol{\nabla}\boldsymbol{\nabla}\varphi)\otimes\boldsymbol{\nabla}\varphi + \boldsymbol{\nabla}\varphi \\ &\otimes(\boldsymbol{\nabla}\varphi\cdot\boldsymbol{\nabla}\boldsymbol{\nabla}\varphi)] + \boldsymbol{\nabla}\left(\frac{\partial E}{\partial c}\right)\otimes\boldsymbol{\nabla}\varphi.\end{aligned} \tag{A2}$$

It is straightforward to show that the first three tensors appearing on the right-hand side of the above equation are symmetric. Thus, we now study the symmetry properties of the tensor $\boldsymbol{\nabla}(\partial E/\partial c)\otimes\boldsymbol{\nabla}\varphi$. It can be shown that

$$\begin{aligned}\boldsymbol{\nabla}\left(\frac{\partial E}{\partial c}\right)\otimes\boldsymbol{\nabla}\varphi = &2\frac{\partial^2 E}{\partial a\,\partial c}(\boldsymbol{\nabla}\varphi\cdot\boldsymbol{\nabla}\boldsymbol{\nabla}\varphi)\otimes\boldsymbol{\nabla}\varphi + \frac{\partial^2 E}{\partial b\,\partial c}\boldsymbol{\nabla}(\boldsymbol{\nabla}^2\varphi) \\ &\otimes\boldsymbol{\nabla}\varphi + \frac{\partial^2 E}{\partial c^2}[2(\boldsymbol{\nabla}\varphi\cdot\boldsymbol{\nabla}\boldsymbol{\nabla}\varphi)\cdot\boldsymbol{\nabla}\boldsymbol{\nabla}\varphi + (\boldsymbol{\nabla}\varphi \\ &\otimes\boldsymbol{\nabla}\varphi):\boldsymbol{\nabla}\boldsymbol{\nabla}\boldsymbol{\nabla}\varphi]\otimes\boldsymbol{\nabla}\varphi.\end{aligned} \tag{A3}$$

It is straightforward to show that none of the three tensors appearing on the right-hand side of the above relation are symmetric.

Therefore, the tensor $\boldsymbol{\tau}^C$ is not symmetric as soon as $(\partial E/\partial c)$ is not constant.

In the particular case of the Helfrich energy (1), one has

$$\frac{\partial E}{\partial c} = -\alpha(\mathcal{C} - \mathcal{C}_0). \tag{A4}$$

Now, the curvature $\mathcal{C}$ depends on the three variables (a,b,c) [see Eq. (3)], so that $\boldsymbol{\nabla}(\partial E/\partial c) \neq 0$. This shows that the curvature tensor $\boldsymbol{\tau}^C$ is not symmetric.

APPENDIX B: SURFACE INVARIANTS

The two parameters of a surface are the two main radii of curvature R_1 and R_2. Two other parameters are important: The mean curvature $\mathcal{C}$ and the Gauss curvature H. These are related to the R_1 and R_2 by

$$\mathcal{C} = \frac{1}{R_1} + \frac{1}{R_2}, \tag{B1}$$

$$H = \frac{1}{R_1}\frac{1}{R_2}. \tag{B2}$$

It is possible to relate $\mathcal{C}$ and H to the invariants of the tensor $\boldsymbol{\nabla}_s\boldsymbol{n}$. Indeed, it can be shown that

$$\mathcal{C} = \boldsymbol{\nabla}_s\cdot\boldsymbol{n}, \tag{B3}$$

$$H = \frac{1}{2}[(\boldsymbol{\nabla}_s\cdot\boldsymbol{n})^2 - \boldsymbol{\nabla}_s\boldsymbol{n}:\boldsymbol{\nabla}_s\boldsymbol{n}]. \tag{B4}$$

The third invariant of $\boldsymbol{\nabla}_s\boldsymbol{n}$ is nil [33].

[1] *Structure and Dynamics of Membranes*, edited by R. Lipowsky and E. Sackmann, Handbook of Biological Physics (Elsevier, Amsterdam, 1995), Vol. 1A and Vol. 1B.

[2] U. Seifert, Adv. Phys. **46**, 13 (1997).

[3] O. A. Ladyzhenskaya, *The Mathematical Theory of Viscous Incompressible Flow*, 2nd ed. (Gordon and Breach, New York, 1969).

[4] C. Pozrikidis, *Boundary Integral and Singularity Methods for Linearized Viscous Flow* (Cambridge University Press, Cambridge, 1992).

[5] M. Kraus, W. Wintz, U. Seifert, and R. Lipowsky, Phys. Rev. Lett. **77**, 3685 (1996).

[6] I. Cantat and C. Misbah, Phys. Rev. Lett. **83**, 880 (1999).

[7] G. Gompper and D. Kroll, J. Phys.: Condens. Matter **9**, 8795 (1997).

[8] A. Malevanets and R. Kapral, J. Chem. Phys. **110**, 8605 (1999).

[9] H. Noguchi and G. Gompper, Phys. Rev. Lett. **93**, 258102 (2004); H. Noguchi and G. Gompper, Proc. Natl. Acad. Sci. U.S.A. **102**, 14159 (2005).

[10] T. Biben and C. Misbah, Phys. Rev. E **67**, 031908 (2003).

[11] T. Biben, K. Kassner, and C. Misbah, Phys. Rev. E **72**, 041921 (2005).

[12] Q. Du, C. Liu, and X. Wang, J. Comput. Phys. **212**, 757 (2006).

[13] F. Campelo and A. Hernández-Machado, Eur. Phys. J. E **20**, 37 (2006).

[14] E. Maitre, C. Misbah, P. Peyla, and A. Raoult (unpublished).

[15] C. S. Peskin, J. Comput. Phys. **25**, 220 (1977).

[16] C. D. Eggleton and A. S. Popel, Phys. Fluids **10**, 1834 (1998).

[17] Li Huabing, Yi Houhui, Shan Xiaowen, and Fang Haiping, e-print arXiv:physics/0607074.

[18] D. Jamet and C. Misbah, Phys. Rev. E **76**, 051907 (2007).

[19] W. Helfrich, Z. Naturforsch. [C] **29c**, 510 (1974).

[20] If one has in mind a more general form of the Gaussian contribution $f(H)$ (with f an arbitrary nonlinear function of H), then the topological invariance does not hold anymore. We shall not, however, consider this general case. Note also that even with the linear function H, the topological invariance is not fulfilled in the strict sense within phase field, since we

would have $\int H|\nabla\varphi|dV$, and not just $\int HdA$, where dA is the area element. However, in the asymptotic limit where the interface width tends to zero, we should recover the topological invariance.

[21] Actually, the mean curvature is defined by $\nabla_s \cdot \boldsymbol{n}$, where ∇_s is the gradient along the contour (s refers to surface). However, since $|\boldsymbol{n}|=1$, it is straightforward to show that $\nabla_s \cdot \boldsymbol{n} = \nabla \cdot \boldsymbol{n}$.

[22] P. Seppecher, Ph.D. thesis, University Paris VI, 1987.

[23] The invariants of a tensor $\boldsymbol{T}$ are $\mathrm{Tr}(\boldsymbol{T})$, $\mathrm{Tr}(\boldsymbol{T}^2)$, and $\mathrm{Tr}(\boldsymbol{T}^3)$ [it can be shown that these invariants are also $\mathrm{Tr}(\boldsymbol{T})$, $\mathrm{Tr}(\boldsymbol{T}^2)$, and $\det(\boldsymbol{T})$]. In the particular case where $\boldsymbol{T}=\nabla\nabla\varphi$, it is straightforward to show that $\mathrm{Tr}(\boldsymbol{T})=\nabla^2\varphi$, $\mathrm{Tr}(\boldsymbol{T}^2)=(\nabla\nabla\varphi:\nabla\nabla\varphi)$, and $\mathrm{Tr}(\boldsymbol{T}^3)=[(\nabla\nabla\varphi\cdot\nabla\nabla\varphi):\nabla\nabla\varphi]$, which are thus the three invariants of the tensor $\nabla\nabla\varphi$. This result is coherent with that provided in Ref. [22].

[24] It is worth noting that the application of thermodynamics of irreversible processes shows that, because the thermodynamic variable $\nabla\varphi$ is a vector, the dynamic viscosity is in general a tensor with five independent components. This feature accounts for the anisotropic character of the viscosity within the interfacial region due to the introduction of a special direction $\nabla\varphi$. However, in the present work, we restrict the model to the purely isotropic case, which is believed to capture the essential ingredient of viscous effects.

[25] S. R. De Groot and P. Mazur, *Non-Equilibrium Thermodynamics* (Wiley, New York, 1962).

[26] J. E. Dunn and J. Serrin, Arch. Ration. Mech. Anal. **88**, 88 (1965).

[27] G. Danker and C. Misbah, Phys. Rev. Lett. **98**, 088104 (2007).

[28] S. Osher and R. Fedkiw, *Level Set Methods and Dynamic Implicit Surface* (Springer, New York, 2003).

[29] The choice of the relative position of the subscripts x, z is consistent with the notations in this paper. We must keep in mind that sometimes (i.e., in several literature) the reverse subscript order is chosen.

[30] Strictly speaking $\tau_{zx}=0$ only at the surface of the plate. But since the plate is thin, it should remain small within the plate. Here we have taken the elementary width of the volume element in Fig. 1 to be that of the plate itself.

[31] R. Folch, J. Casademunt, A. Hernández-Machado, and L. Ramirez-Piscina, Phys. Rev. E **60**, 1724 (1999).

[32] While a surface tension does exist for fluid interfaces, the authors in [31] were interested in not including this phase-field surface tension in the phase-field evolution itself, but rather in the momentum balance equation.

[33] It is straightforward to show that $\boldsymbol{n}\cdot(\nabla_s\boldsymbol{n})=0$ and $(\nabla_s\boldsymbol{n})\cdot\boldsymbol{n}=0$ so that the determinant of $\nabla_s\boldsymbol{n}$, which is the third invariant, is nil. If we set $\boldsymbol{A}\equiv\nabla_s\boldsymbol{n}$, then the ith components in the products above are defined as $(\boldsymbol{n}\cdot\boldsymbol{A})_i=\boldsymbol{n}_j\boldsymbol{A}_{ji}$, $(\boldsymbol{A}\cdot\boldsymbol{n})_i=\boldsymbol{A}_{ij}\boldsymbol{n}_j$.