Unambiguous Gibbs dividing surface for nonequilibrium finite-width interface: Static equivalence approach

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The definition of all properties of the nonequilibrium interface depends on the choice of the position of the dividing surface. However, the definition of its position has been an unsolved problem for more than a century. A missing principle to unambiguously determine the position of the Gibbs' dividing surface is found: the principle of static equivalence. A sharp interface (dividing surface) is statically equivalent to a nonequilibrium finite-width interface with distributed tensile stresses if it possesses (a) the same resultant force, equal to the interface energy, and (b) the same moment, which is zero about the interface position. Each of these conditions determines the position of a sharp interface, which may be contradictory. This principle is applied to resolve another basic problem: the development of a phase field approach to an interface motion that includes an expression for interface stresses, which are thermodynamically consistent, and consistent with a sharp-interface limit. Using an analytical solution for a curved propagating interface, it is shown that both conditions determine the same dividing surface, i.e., the theory is self-consistent. The expression for the interface energy is also consistent with the expression for the velocity of the curved sharp interface. Applications to more complex interfaces that support elastic stresses are discussed.

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I. INTRODUCTION

In his pioneering work on interface properties [1], Gibbs introduced a concept of a sharp interface or dividing surface that is aimed to substitute an actual finite-width interface [Fig. 1(a)]. The excess of any properties at the interface between phases 1 and 2, in particular, the free energy per unit undeformed volume ψ, is defined with respect to the corresponding properties in bulk, ψ_1 and ψ_2, and the total interface excess energy per unit area is defined as

$$\gamma := \int_{-\infty}^{\varsigma_{ds}} (\psi - \psi_1) d\varsigma + \int_{\varsigma_{ds}}^{\infty} (\psi - \psi_2) d\varsigma, \qquad (1)$$

where ς_{ds} is the position of the dividing surface. It is clear that the excess quantity for a nonequilibrium interface ($\psi_1 \neq \psi_2$) depends on the choice of ς_{ds}, which was not strictly defined either in Ref. [1] or later. Alternative definitions of the excess properties, which do not involve a dividing surface, were suggested for different purposes [1–9]. Still, as it is mentioned in Refs. [1–8], arbitrariness in some definitions persists, and there is no direct connection between the distribution of properties for a finite-width interface and interface excess properties. Also, most of the definitions are based on the reversible work to change area of a stationary interface. They are valid for an equilibrium interface only, because a nonequilibrium interface propagates and dissipates energy. However, for an equilibrium interface, $\psi_1 = \psi_2$, and the interface energy can be determined without knowledge of the position of the dividing surface. Thus, the necessity for a conceptual definition of the position of the dividing surface is of fundamental and applied importance for nonequilibrium interfaces. This longstanding problem is resolved here by utilizing the *principle of static equivalence* [Fig. 1(a)]: A finite-width interface with distributed tensile stresses is statically equivalent to the sharp interface if it possesses the same resultant force and the same moment about any chosen point [10]. For a sharp interface, (a) the resultant force is equal to γ and (b) the moment about the interface position is zero. Each of these conditions determines a sharp-interface position. Ironically, for more than a century there were no conditions to determine the dividing surface. With the help of undergraduate statics, we found two conditions, which may be potentially inconsistent with each other. Utilizing the phase field approach, we will demonstrate that they are consistent, at least for the model considered here.

The phase field approach is broadly used for the modeling of the structure of evolving interfaces including interface stresses [11–17]. Phase evolution is described by an order parameter η, which varies from 0 in phase 1 to 1 in phase 2, and is obtained by the solution of the Ginzburg-Landau equation. However, introducing interface stresses in a phase field approach is a nontrivial problem. As it was shown in Ref. [14], the results given in Refs. [11–13] are not consistent with a sharp-interface limit because the interface tension contains an additional hydrostatic pressure, or it spreads in the bulk for a nonequilibrium interface [11]. In Refs. [15,16] interface stresses were properly introduced for solid-solid and solid-melt equilibrium interfaces. In Refs. [14,17] results were extended for a nonequilibrium interface for a fourth-degree polynomial thermodynamic potential. Since the approach in Refs. [14,17] utilizes an analytical solution for a propagating interface, it depends on a specific potential. However, a fourth-degree potential is symmetric with respect to an exchange of phases 1 and 2, the distribution of interface stresses is symmetric, and the dividing surface in Eq. (1) was trivially determined by the condition $\eta_{ds} = 0.5$. Here, we will develop a similar approach for the sixth-degree thermodynamic potential, which is not symmetric and whose interface stress distribution is

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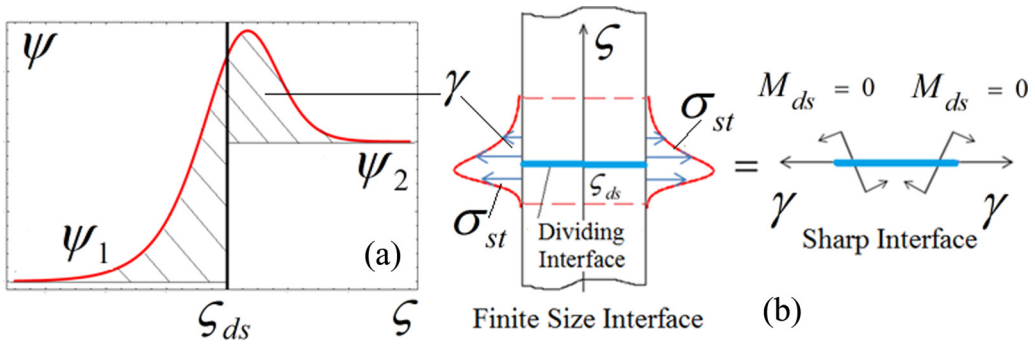

FIG. 1. (Color online) Principle of static equivalence. (a) Distribution of the free energy across a nonequilibrium interface and definition of an interface excess energy as an area of dashed regions. (b) Principle of static equivalence of distributed interface stresses for a finite-width interface and the resultant force placed at the dividing surface. Each of the conditions, the equality of the interface energy in (a) and the resultant force in (b) to γ and zero moment in (b), determines the position of the dividing surface.

also not symmetric. Thus, without an explicit definition of the position of the dividing surface, this theory cannot be completed. We found that conditions (a) and (b) result in the same sharp-interface position, and all other conditions for interface stresses are satisfied, i.e., the developed phase field approach is fully self-consistent.

II. PRINCIPLE OF STATIC EQUIVALENCE

We will start with an interface between a liquid and vapor or two liquid phases, which does not support elastic stresses. Such an interface is subjected to interface stresses (surface tension), which represent biaxial tension σ_{st} with the magnitude of the resultant force equal to the nonequilibrium interface energy γ (which depends on ς_{ds}). The following problem formulation was considered (Fig. 1). Let a finite-width curved interface propagating with velocity c_k (which depends on the mean interface curvature K) in direction r along its unit normal $\boldsymbol{k}$ and distribution of the excess stress tensor,

$$\boldsymbol{\sigma}_{st} = \sigma_{st}(\varsigma)(\boldsymbol{I} - \boldsymbol{k} \otimes \boldsymbol{k}), \quad \boldsymbol{k} = \nabla\eta/|\nabla\eta|, \quad \varsigma = r - c_k t, \tag{2}$$

corresponding to biaxial tension, as well as energy $\psi(\varsigma)$, be given. Here t is time, $\otimes$ designates the dyadic product of vectors, ∇ is the gradient operator in the deformed (current) state, and $\boldsymbol{I}$ and $\boldsymbol{I} - \boldsymbol{k} \otimes \boldsymbol{k}$ are the unit tensor and the two-dimensional unit tensor within a diffuse interface, respectively. For the radius of curvature $1/K$ that is much larger than the interface width δ (thin interface), K can be considered as independent of r within an interface. One needs to substitute such a finite-width interface with an equivalent sharp interface at location ς_{ds}.

Equivalence consists of several points:

(1) The sharp interface possesses an energy defined in Eq. (1) (energetic equivalence).

(2) The tangential force per unit interface length is

$$T := \int_{-\infty}^{\infty} \sigma_{st} d\varsigma = \gamma, \tag{3}$$

which is part of the static equivalence. Note that if the stress and energy distributions are known, the combination of Eqs. (1) and (3) represents the missing equation with respect to ς_{ds}, which, however, was not used for such purposes.

(3) The stress tensor is defined as

$$\boldsymbol{\sigma}_{st} = \gamma(\boldsymbol{I} - \boldsymbol{k} \otimes \boldsymbol{k})\delta_D(\varsigma_{ds}) \tag{4}$$

(stress state equivalence), which satisfies Eq. (3) due to the Dirac delta function δ_D property, $\int_{-\infty}^{\infty} \delta_D(\varsigma_{ds})d\varsigma = 1$.

(4) The static equivalence of two systems of force (stress) means that they not only possess the same resultant force, but also the same moment about any chosen point [10]. For a sharp-interface moment with respect to ς_{ds}, $M_{ds} = 0$. Indeed, neither interface nor liquid or gas phases support moments. Then the same is true for distributed stresses for a finite-size interface:

$$M_{ds} := \int_{-\infty}^{\infty} (\varsigma - \varsigma_{ds})\sigma_{st}(\varsigma)d\varsigma = 0. \tag{5}$$

Geometrically, Eq. (5) means that ς_{ds} is the ς coordinate of the centroid of the area below the $\sigma_{st}(\varsigma)$ curve [10]. In particular, if $\sigma_{st}(\varsigma)$ is symmetric with respect to axes passing through some point, this point is ς_{ds}. Thus, we found two conditions which may be potentially inconsistent with each other. Utilizing the phase field approach, we will demonstrate that they are consistent.

III. PHASE FIELD APPROACH

A. Helmholtz free energy

We accept the following expression for the Helmholtz free energy:

$$\begin{aligned} \psi(\boldsymbol{\varepsilon},\eta,\theta,\nabla\eta) &= \psi^e + J\check{\psi}^\theta + \tilde{\psi}^\theta + J\psi^\nabla, \\ \psi^\nabla &= 0.5\beta|\nabla\eta|^2, \\ \tilde{\psi}^\theta + \check{\psi}^\theta &= f(\theta,\eta) = \Delta G^\theta \eta^4(3 - 2\eta^2) \\ &\quad + 0.5A\eta^2(1 - \eta^2)^2, \end{aligned} \tag{6}$$

where θ is the temperature, ψ^e, f, and ψ^∇ are the elastic, thermal, and gradient contributions to the energy, respectively, $J = 1 + \varepsilon_0$, ε_0 is the volumetric strain, $\boldsymbol{\varepsilon}$ is the strain tensor, $\Delta G^\theta(\theta)$ is the difference between the thermal parts of the energies of phases 2 and 1, and $A(\theta)$ and $\beta(\theta)$ are the double-well energy and gradient energy coefficients, respectively. Elastic energy and strains can be treated in the same way as in Ref. [14], which will not be repeated here. Function

$f(\eta)$ can be found, e.g., in Ref. [18]. Functions $\tilde{\psi}^\theta$ and $\breve{\psi}^\theta$ separately will be found below. As it was demonstrated in Refs. [14,17], the multiplication of functions $\breve{\psi}^\theta$ and ψ^∇ by J and the dependence of ψ^∇ on the gradient $\nabla\eta$ in the *deformed* state leads to the appearance of the interface stresses of the following structure:

$$\boldsymbol{\sigma}_{st} = \beta|\nabla\eta|^2(\boldsymbol{I} - \boldsymbol{k}\otimes\boldsymbol{k}) + (\breve{\psi}^\theta - 0.5\beta|\nabla\eta|^2)\boldsymbol{I}. \quad (7)$$

Thus, one has to find proper partitioning of function f into $\tilde{\psi}^\theta$ and $\breve{\psi}^\theta$ so that, for the propagating interface, the second term in Eq. (7) disappears and interface stresses reduce to the desired biaxial tension Eq. (2).

B. Analytical solution for curved nonequilibrium interfaces

The linear relationship between generalized thermodynamic forces and rates results in the Ginzburg-Landau equation

$$\frac{1}{L}\frac{\partial\eta}{\partial t} = -\frac{\partial\psi}{\partial\eta} + \nabla\cdot\left(\frac{\partial\psi}{\partial\nabla\eta}\right) = -\frac{\partial\psi}{\partial\eta} + \beta\nabla^2\eta, \quad (8)$$

where L is the kinetic coefficient and ∇^2 is the Laplacian operator. For a curved interface one can introduce a local curvilinear orthogonal coordinate system with a coordinate r along the normal to the interface. When gradients along the interface are neglected, the Laplacian operator in this curvilinear system is [19]

$$\nabla^2 := \frac{\partial^2}{\partial r^2} + 2K\frac{\partial}{\partial r}, \quad (9)$$

where K is the mean curvature of the interface. For the radius of curvature $1/K$ that is much larger than the interface width δ, K can be considered as independent of r within an interface. Then the Ginzburg-Landau equation (8) in the local curvilinear system takes the following form for $J = 1$:

$$\frac{1}{L}\frac{\partial\eta}{\partial t} = -\frac{\partial\psi}{\partial\eta} + \beta\frac{\partial^2\eta}{\partial r^2} + 2\beta K\frac{\partial\eta}{\partial r}. \quad (10)$$

Substituting in Eq. (10) an ansatz for the moving interface, $\eta = f(\varsigma)$ with $\varsigma = r - c_k t$, where c_k is the curvature-dependent interface velocity, one obtains

$$-\left(\frac{c_k}{L} + 2\beta K\right)\frac{\partial\eta}{\partial\varsigma} = -\frac{\partial\psi}{\partial\eta} + \beta\frac{\partial^2\eta}{\partial\chi^2}. \quad (11)$$

For a plane interface, $K = 0$, and Eq. (11) simplifies to

$$-\frac{c}{L}\frac{\partial\eta}{\partial\varsigma} = -\frac{\partial\psi}{\partial\eta} + \beta\frac{\partial^2\eta}{\partial\chi^2}. \quad (12)$$

An analytical solution to Eq. (12) for a plane nonequilibrium interface is [18]

$$\eta_{in} = (1 + e^{-\zeta})^{-0.5}, \quad \zeta = k(r - ct) = k\varsigma, \quad (13)$$

$$k = 2\sqrt{B/\beta}, \quad B = (A - 4\Delta G^\theta), \quad (14)$$

$$c = \mu\Delta G^\theta, \quad \mu = 8L/k, \quad (15)$$

where μ is the interface mobility. Since Eq. (11) for a curved interface coincides with Eq. (12) for a plane interface for $c = c_k + 2\beta KL$, one can rewrite the solutions Eqs. (13)–(15) for the thin curved interface:

$$\eta_{in} = (1 + e^{-\zeta})^{-0.5}, \quad \zeta = k(r - c_k t), \quad (16)$$

$$k = 2\sqrt{B/\beta}, \quad B = (A - 4\Delta G^\theta), \quad (17)$$

$$c_k = \mu\Delta G^\theta - 2\beta KL = \mu\left(\Delta G^\theta - \frac{k\beta K}{4}\right), \quad \mu = 8L/k. \quad (18)$$

Thus, the thin curved interface has the same profile and, consequently, energy and width, but the interface velocity has an additional contribution due to curvature.

C. Energy and interface stress tensor for a nonequilibrium interface

We limit ourselves to thermodynamic parameters and a time interval in which the solution Eqs. (16)–(18) is stable with respect to possible fluctuations. The most important property of the solution Eq. (16) is

$$d\eta_{in}/d\zeta = 0.5\eta_{in}\left(1 - \eta_{in}^2\right). \quad (19)$$

Using it and the definition of k in Eq. (17), we obtain a key relationship for points of a propagating interface:

$$\psi_{in}^\nabla = 0.5\beta|\nabla\eta_{in}|^2 = 0.5\beta k^2(d\eta/d\zeta)^2 = 0.5B\eta_{in}^2\left(1 - \eta_{in}^2\right)^2. \quad (20)$$

To obtain biaxial interface tension in Eq. (7) for the propagating interface, one has to define for an arbitrary distribution of η,

$$\breve{\psi}^\theta := \psi_{in}^\nabla = 0.5B\eta^2(1 - \eta^2)^2, \quad (21)$$

where Eq. (20) was used. Substituting Eq. (21) in Eq. (7), we obtain for a propagating interface

$$\boldsymbol{\sigma}_{st} = \sigma_{st}(\boldsymbol{I} - \boldsymbol{k}\otimes\boldsymbol{k}), \quad \sigma_{st} = \beta|\nabla\eta|^2 = 2\breve{\psi}^\theta. \quad (22)$$

Evidently, σ_{st} is localized at the diffuse interface, as required. The desired stress state as in Eq. (2) is obtained. Also,

$$\tilde{\psi}^\theta = f(\theta,\eta) - \breve{\psi}^\theta = \Delta G^\theta(\theta)\eta^2(2 - \eta^2). \quad (23)$$

We still need to prove that the resultant force per unit interface length is equal to the interface energy γ. This includes the definition of the dividing surface. Setting without loss of generality $\psi_1 = f(\theta,0) = 0$ and $\psi_2 = f(\theta,1) = \Delta G^\theta(\theta)$, substituting an expression for ψ from Eq. (6) and the solution for the interface Eqs. (16) and (17) into Eq. (1) and integrating, we obtain for the gradient Ψ^∇ and local Ψ^l parts of γ:

$$\Psi^\nabla = \frac{k\beta}{16} = \frac{\sqrt{\beta B}}{8}, \quad \Psi^l = \Psi^\nabla + \frac{Z\Delta G^\theta}{k}, \quad Z = 1 + \ln\left(\frac{\bar{\eta}^2}{1 - \bar{\eta}^2}\right), \quad (24)$$

with $\bar{\eta} := \eta(\varsigma_{ds})$. While the gradient part is obviously independent of the dividing surface, local energy depends on the value $\bar{\eta}$ for the dividing surface. Substituting σ_{st} from Eq. (22) and η_{in} from Eqs. (16) and (17) in zero-moment condition

Eq. (5), we obtain $\bar{\eta} = (1+e)^{-0.5} = 0.5186$ and $Z = 0$, as well as for the interface energy,

$$\gamma = \Psi^l + \Psi^\nabla = 2\Psi^l = 2\Psi^\nabla = k\beta/8 = \sqrt{\beta B}/4. \quad (25)$$

Thus, the application of one of the principle of static equivalence conditions Eq. (5) leads to explicit position of the dividing surface and to equality of the local and gradient energies for the nonequilibrium interface, which is similar to the result for the equilibrium interface.

It is evident that another static equivalence condition Eq. (3) is satisfied. Indeed, if stress σ_{st} in Eq. (22) is equal to the double gradient energy at each point, then the total force is equal to the double total gradient energy, which is interface energy γ. Thus, the position of the dividing surface, interface energy, and interface stresses that satisfy all the formulated conditions are obtained in a self-consistent way. This means that zero-moment condition (5) and a combination of Eqs. (1) and (3) results in the same sharp-interface position.

Moreover, an important point is that Eqs. (25) and (18) result in the equation for interface velocity,

$$c_k = \mu(\Delta G^\theta - 2\gamma K), \quad (26)$$

which is consistent with the sharp-interface approach. Note that one can use Eq. (26) as an alternative definition of the nonequilibrium interface energy (which, however, cannot be applied to a plane interface). Then a combination of Eqs. (26) and (18) leads to the correct equation for the interface energy Eq. (25) without involving a dividing surface.

After a consistent expression for γ is found, there is no problem determining other interface excess functions using the same equations as for bulk functions. For example, for entropy excess,

$$s_i = -\frac{\partial\gamma}{\partial\theta} = -\frac{\partial}{\partial\theta}\left(\int_{-\infty}^{\varsigma_{ds}}(\psi-\psi_1)d\varsigma + \int_{\varsigma_{ds}}^{\infty}(\psi-\psi_2)d\varsigma\right)$$
$$= \int_{-\infty}^{\varsigma_{ds}}(s_i - s_{i1})d\varsigma + \int_{\varsigma_{ds}}^{\infty}(s_i - s_{i2})d\varsigma, \quad (27)$$

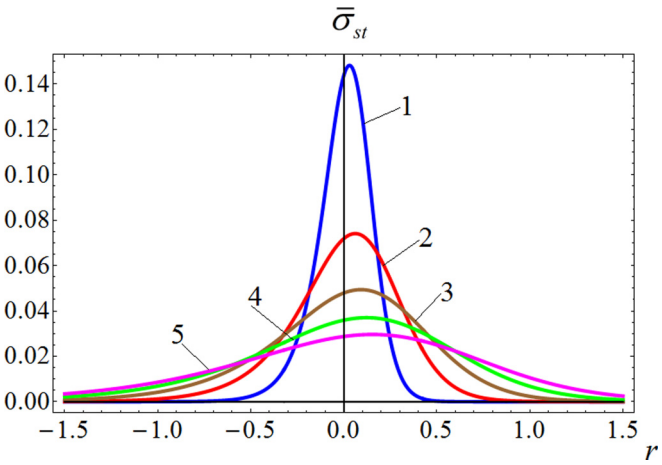

FIG. 2. (Color online) Distribution of the dimensionless biaxial interface stress $\bar{\sigma}_{st}(r)$ for the nonequilibrium interface for different interface widths δ shown near the curves.

one obtains a definition consistent with the same position of the dividing surface ς_{ds}.

The diffuse interface width is defined as $\delta := 10/k = 5\sqrt{\beta/B} = 5\beta/(4\gamma)$, where a factor of 10 is an approximate width of the diffuse interface $\eta_{in}(\zeta)$ in terms of ζ in Eq. (17). The distribution of the normalized biaxial interface stresses is

$$\bar{\sigma}_{st} = \sigma_{st}/(20B\gamma) = \eta_{in}^2\left(1-\eta_{in}^2\right)^2/\delta. \quad (28)$$

In Fig. 2, the distribution of $\bar{\sigma}_{st}$ is plotted after the substitution of $e^{-\zeta} = e^{-10r/\delta+1}$, which provides that $r = 0$ coincides with the dividing surface. Since γ is the same in all plots, the resultant force, i.e., area below each curve, is also the same.

IV. CONCLUDING REMARKS

To summarize, a strict solution to the longstanding problem in the interface and surface science, formulated by Gibbs, is found: how to define the position of the sharp interface equivalent to the finite-width interface. We utilized the principle of static equivalence and even obtained two solutions, which fortunately coincide at least for the phase field model considered here. It is also consistent with the expression for the velocity of the curved sharp interface. The next question is whether this is the case for other phase field polynomials and theories and whether there is a way to prove the equivalence of both conditions for an arbitrary thermodynamic potential. Note that the principle of static equivalence is the fundamental principle of mechanics (and, consequently, physics) and it must not be violated. It is surprising that it was neglected in the interface and surface science. It is applicable to any interface (not just liquid-liquid and liquid-gas) interfaces; however, the results for different interfaces can be different. In particular, for solid-solid and solid-melt interfaces that support elastic stresses, the resultant force T is always determined by the first Eq. (3) and the resultant moment is always determined by the first Eq. (5); if the interface stresses are anisotropic, i.e., depend on $\boldsymbol{k}$ and the direction within an interface, these equations are valid for any direction. However, with elastic interface stresses, $T \neq \gamma$, so this condition should not be satisfied. In most works [1–5,7,8,20] no bending moments (couples) are present for sharp interfaces, which means that the condition $M_{ds} = 0$ is inexplicitly assumed. In this case, it can be used to determine the position of the dividing surface. If the dividing surface determined from Eq. (5) is located outside of an interface or is significantly shifted from the point with $\bar{\eta} = 0.5$ (which is the case for stresses at solid-liquid interfaces in Ref. [20]), this is the sign that one must introduce couples in the sharp-interface theory, and such formal theories are known [21]. This may be true for large transformation strain that varies across an interface from zero to the finite value in phase 2, which causes bending of a diffuse interface. Also, for an anisotropic interface and unequal normal stresses in two principle directions, if the position of an interface is chosen from a zero-moment condition for one of the principle directions, for another one the moment is generally not zero and should be included in the sharp-interface model. In these cases, the finite-width distribution (obtained, e.g., using the phase field approach or molecular dynamics) can be used to determine constitutive equations for a sharp interface, including couple stresses. The developed phase field approach can be generalized for higher

order polynomials, for multiple order parameters, using the potential developed in Ref. [22] for twinning and multivariant transformations in solids, for interfaces with intermediate phases [23], for finite-width external surfaces [24,25], and for conservative order parameters described by the Cahn-Hilliard equation.

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