

Achieving realistic interface kinetics in phase-field models with a diffusional contrastG. Boussinot^{1,2} and Efim A. Brener¹¹*Peter Grünberg Institut, Forschungszentrum Jülich, D-52425 Jülich, Germany*²*Computational Materials Design Department, Max-Planck Institut für Eisenforschung, D-40074 Düsseldorf, Germany*

(Received 5 February 2014; published 30 June 2014)

Phase-field models are powerful tools to tackle free-boundary problems. For phase transformations involving diffusion, the evolution of the nonconserved phase field is coupled to the evolution of the conserved diffusion field. Introducing the kinetic cross coupling between these two fields [E. A. Brener and G. Boussinot, *Phys. Rev. E* **86**, 060601(R) (2012)], we solve the long-standing problem of a realistic description of interface kinetics when a diffusional contrast between the phases is taken into account. Using the case of the solidification of a pure substance, we show how to eliminate the temperature jump at the interface and to recover full equilibrium boundary conditions. We confirm our results by numerical simulations.

DOI: [10.1103/PhysRevE.89.060402](https://doi.org/10.1103/PhysRevE.89.060402)

PACS number(s): 81.10.Aj, 05.70.Ln, 64.70.dm, 81.30.Fb

Introduction. The phase-field method is one of the most important and powerful tools for modeling interface dynamics and pattern formation processes. In particular, it has proven its efficiency for the description of microstructure evolution during phase transformations that are coupled to bulk diffusion. However, anomalous interface kinetic effects exist when the diffusion coefficients in the two phases differ [1–4]. Eliminating them remains a long-standing problem for a realistic phase-field modeling.

The phase-field method tackles free-boundary problems with the use of fields that are continuous across the interface. They exhibit an intrinsic length scale, the interface width W , that has to be much smaller than the length scales of the emerging pattern. For computationally tractable simulations, W is usually chosen significantly larger than the actual width of the physical interface. However, this leads to an artificial enhancement of the interfacial kinetic effects (for an extended discussion, see Refs. [1–4]). Therefore it is important to eliminate these artificial effects by a proper adjustment of the model parameters. For example, for the solidification of a pure substance where the growth is coupled to the heat transport, one may look for an elimination of the jump of temperature at the interface (Kapitza jump). This is the usual approximation in thermal problems. A more particular goal is to recover full equilibrium boundary conditions (no kinetic effects), a situation that corresponds to the limit of small driving forces and that is often met in experiments.

Classical phase-field models do not contain a kinetic cross coupling between the nonconserved phase field and the conserved diffusion field, and in this respect are diagonal (model C in the Hohenberg-Halperin nomenclature [5]). Karma and Rappel developed the so-called thin interface analysis to connect a phase-field model and the corresponding macroscopic description [1]. They found that if the diffusivity does not depend on the phase field, i.e., is the same in the two phases, the Kapitza jump is automatically eliminated. They calculated the growth kinetic coefficient and gave the relation between the parameters of the phase-field model to recover equilibrium boundary conditions. However, Almgren showed that, in the presence of a diffusional contrast, the Kapitza jump cannot be eliminated without producing anomalous corrections to the energy conservation law at the interface [2]. For isothermal phase transformations in alloys, the same

problem exists concerning the jump of the diffusion chemical potential (relevant to the solute trapping effect). For the solidification of alloys where the diffusion flux is neglected in the solid (one-sided model), the so-called antitrapping model was developed to tackle this problem [3,4]. In Ref. [6], an attempt was made to extend this model to a finite diffusion in the solid. Although, in this case, the diffusional fluxes on the two sides of the interface and the interface velocity are related by only one equation (the conservation law), the authors used an additional relation between them [Eq. (5.1) in Ref. [6]]. Thus, they imposed an unjustified linear relation between two quantities that are, in fact, linearly independent [7].

Recently, a kinetic cross coupling between the nonconserved phase field and the conserved diffusion field was introduced in the phase-field equations of motion [8–10]. With this cross coupling, in general allowed by Onsager symmetry, the phase-field model becomes nondiagonal and possesses an additional kinetic velocity scale compared to classical diagonal models. This provides a full correspondence between the number of independent kinetic parameters in the phase field and the macroscopic descriptions. It was shown that the cross coupling has a crucial importance for some kinetic effects, such as solute trapping in alloys [8] and the Ehrlich-Schwoebel effect in molecular-beam epitaxy [10]. Moreover, it was also successfully adapted to the case of the one-sided model [9,10] recovering a full thermodynamical consistency of the antitrapping model that originally does not obey Onsager symmetry.

In this Rapid Communication, we use a nondiagonal phase-field model [8,10] to address the long-standing problem of achieving realistic simulations in the presence of a diffusional contrast. In particular, we show that the introduction of the kinetic cross coupling allows one to eliminate the Kapitza jump and, in addition, to recover full equilibrium boundary conditions at the interface. This provides a generalization of the results by Karma and Rappel [1] in the presence of a diffusional contrast. We test numerically these results using a simple but illustrative example.

Variational phase-field model with kinetic cross coupling. Complementary to our previous studies on alloys [8,10], we focus here on the solidification of a pure material where the phase transformation is coupled to the heat transport. We measure all energy densities in units of the latent heat of the

transformation $L = T_M(S_L - S_S)$, where T_M is the melting temperature and S_L (S_S) is the entropy of the liquid (solid) phase at T_M . We start from an entropy functional S ,

$$S = \int_V dV \{s[e(T, \phi), \phi] - H[(1 - \phi^2)^2/4 + (W\nabla\phi)^2/2]\}, \quad (1)$$

where e is the dimensionless internal energy density that depends on the temperature T and on the phase field ϕ . For simplicity we assume that the cost of an interface is provided by the variations of the phase field only and is described by H . The entropy production is

$$\dot{S} = \int_V dV \left[\frac{\delta S}{\delta \phi} \dot{\phi} + \frac{\delta S}{\delta e} \dot{e} \right] = \int_V dV \left[\frac{\delta S}{\delta \phi} \dot{\phi} + \nabla \frac{\delta S}{\delta e} \cdot \mathbf{J} \right], \quad (2)$$

where the flux of dimensionless energy \mathbf{J} enters the continuity equation

$$\dot{e} = -\nabla \cdot \mathbf{J}. \quad (3)$$

Then the variational equations of motion relate $\delta S/\delta \phi$ and $\nabla(\delta S/\delta e)$ to $\dot{\phi}$ and \mathbf{J} through the Onsager linear relations,

$$\frac{c_P T_M^2}{L^2} \frac{\delta S}{\delta \phi} = \tau \dot{\phi} + (MW\nabla\phi) \cdot \mathbf{J}, \quad (4)$$

$$\frac{c_P T_M^2}{L^2} \nabla \frac{\delta S}{\delta e} = (MW\nabla\phi) \dot{\phi} + \frac{\mathbf{J}}{D(\phi)}, \quad (5)$$

where c_P is the specific heat at $T = T_M$ that is assumed to be independent of the phase (independent of ϕ). For simplicity, we consider constant τ and M . The thermal diffusivity $D(\phi)$ depends on ϕ to account for a diffusional contrast. While the variational formulations without cross coupling ($M = 0$) have been well known in the literature for a long time (see, for example, Ref. [11] and references therein), cross terms were introduced only recently [8–10]. They obey Onsager symmetry and are naturally written proportional to $W\nabla\phi$. This vector is normal to the interface and is of magnitude of order unity in the interface region and zero in the bulk. It is also important to make a proper link between scalar and vectorial quantities (we refer to Refs. [8,10] for more details). A positive entropy production $\dot{S} > 0$ requires

$$\tau > 0, D(\phi) > 0, \Delta = 1 - (MW\nabla\phi)^2 D(\phi)/\tau > 0. \quad (6)$$

The variational derivatives $\delta S/\delta \phi = [\partial s/\partial \phi]_e + H[\phi(1 - \phi^2) + W^2\nabla^2\phi]$ and $\delta S/\delta e = [\partial s/\partial e]_\phi$ are evaluated using the thermodynamical relations $[\partial s/\partial \phi]_e = -(L/T)[\partial f(T, \phi)/\partial \phi]_T$ and $[\partial s/\partial e]_\phi = L/T$ [11]. The dimensionless free energy $f = e - T_S/L$ is chosen equal to zero at $T = T_M$. We interpolate the entropies at T_M as

$$\frac{T_M}{L} s(T = T_M, \phi) = \sigma(\phi) = \frac{\sigma_S + \sigma_L}{2} - \frac{\sigma_L - \sigma_S}{2} p(\phi), \quad (7)$$

where $\sigma_S = T_M S_S/L$, $\sigma_L = T_M S_L/L$, and hence $\sigma_L - \sigma_S = 1$. To be explicit, we choose the switching function $p(\phi) = 15(\phi - 2\phi^3/3 + \phi^5/5)/8$ that has the properties $p(\phi = \pm 1) = \pm 1$, $p(\phi) = -p(-\phi)$, and $p'(\phi = \pm 1) = p''(\phi = \pm 1) = 0$. Then, near $T = T_M$, the dimensionless internal

energy e and free energy f read

$$e(T, \phi) = u + \sigma(\phi), \quad f(T, \phi) = -\frac{L}{c_P T_M} \sigma(\phi) u, \quad (8)$$

with the dimensionless temperature $u = c_P(T - T_M)/L$. Finally, using the above considerations, we present Eqs. (3)–(5) in the form of coupled equations of motion for ϕ and u ,

$$\Delta \tau \dot{\phi} = \tilde{H}[\phi(1 - \phi^2) + W^2\nabla^2\phi] - \frac{p'(\phi)}{2} u + MW D(\phi) \nabla\phi \cdot \nabla u \quad (9)$$

and

$$\dot{u} = \nabla \cdot \{D(\phi)[\nabla u + MW\dot{\phi}\nabla\phi]\} + \frac{p'(\phi)}{2} \dot{\phi}, \quad (10)$$

where $\tilde{H} = (c_P T_M^2/L^2)H$.

At equilibrium where $u = 0$ and $\dot{\phi} = 0$, the phase field obeys $\phi_{\text{eq}}(x) = -\phi_{\text{eq}}(-x) = -\tanh(x/W\sqrt{2})$ for a solid phase at $x = -\infty$ and a liquid phase at $x = +\infty$, and $W^2[\phi'_{\text{eq}}(x)]^2 = [1 - \phi_{\text{eq}}^2(x)]^2/2$. We also define $\sigma_{\text{eq}}(x) = \sigma[\phi_{\text{eq}}(x)] = (\sigma_S + \sigma_L)/2 - p[\phi_{\text{eq}}(x)]/2$.

Macroscopic description and thin interface limit. In the macroscopic description, one discusses the thermal diffusion equation in the bulk with boundary conditions at the sharp interface. First, the energy conservation at the interface reads

$$D_S \nabla u|_S \cdot \mathbf{n} + V \sigma_S = D_L \nabla u|_L \cdot \mathbf{n} + V \sigma_L = J_E, \quad (11)$$

where D_S (D_L) is the thermal diffusivity in the solid (liquid) phase, \mathbf{n} is the vector normal to the interface, and $\nabla u|_S$ ($\nabla u|_L$) is the gradient of u on the solid (liquid) side of the interface. V is the normal velocity of the interface and J_E is the normal flux of dimensionless energy through the interface. Second, the temperature at the interface is T_S (T_L) on the solid (liquid) side and deviates from T_M due to interfacial kinetic effects. The kinetic boundary conditions relate, through linear Onsager relations, the difference of chemical potential or free energy across the interface δf and the Kapitza temperature jump δu to their conjugate fluxes V and J_E . These relations read [12,13]

$$(c_P T_M/L) \delta f = \sigma_S u_S - \sigma_L u_L = \bar{A} V + \bar{B} J_E + d_0 \kappa, \quad (12)$$

$$\delta u = u_L - u_S = \bar{B} V + \bar{C} J_E, \quad (13)$$

where $u_S = c_P(T_S - T_M)/L$ and $u_L = c_P(T_L - T_M)/L$. The Gibbs-Thomson correction proportional to the curvature κ of the interface is parametrized by the capillary length $d_0 = \gamma c_P T_M/L^2$, where the interface energy is related to the phase-field parameters through $\gamma = \alpha H T_M W$ with $\alpha = W \int_{-\infty}^{\infty} dx [\phi'_{\text{eq}}(x)]^2 = 2\sqrt{2}/3$ [1]. \bar{A} is the inverse growth kinetic coefficient (usually denoted by β when the other kinetic coefficients are absent), \bar{C} is the Kapitza resistance, and \bar{B} is the cross coefficient. A physically motivated procedure for the thin interface limit that involves the entropy production (dissipation function) in the interface region was introduced in Ref. [10]. It is equivalent to the more mathematically oriented asymptotic matching [1,2]. It allows one to express the macroscopic kinetic coefficients $\bar{A}, \bar{B}, \bar{C}$ in terms of the parameters of the phase-field model τ, M and the

thermal diffusivity $D(\phi)$ [compare with Eqs. (19)–(21) in Ref. [10]]:

$$\bar{A} = \int_{-\infty}^{\infty} dx [\tau - 2MW\sigma_{\text{eq}}(x)][\phi'_{\text{eq}}(x)]^2 + \int_{-\infty}^{\infty} dx \left[\frac{\sigma_{\text{eq}}^2(x)}{D[\phi_{\text{eq}}(x)]} - \frac{\sigma_S^2}{2D_S} - \frac{\sigma_L^2}{2D_L} \right], \quad (14)$$

$$\bar{B} = \int_{-\infty}^{\infty} dx MW[\phi'_{\text{eq}}(x)]^2 - \int_{-\infty}^{\infty} dx \left[\frac{\sigma_{\text{eq}}(x)}{D[\phi_{\text{eq}}(x)]} - \frac{\sigma_S}{2D_S} - \frac{\sigma_L}{2D_L} \right], \quad (15)$$

$$\bar{C} = \int_{-\infty}^{\infty} dx \left[\frac{1}{D[\phi_{\text{eq}}(x)]} - \frac{1}{2D_S} - \frac{1}{2D_L} \right]. \quad (16)$$

As we see, here there is a larger flexibility for tuning kinetic coefficients compared to classical phase-field models where $M = 0$.

Elimination of the Kapitza jump and full equilibrium boundary conditions. The models for the solidification of a pure material usually assume $\delta u = 0$. Since V and J_E are linearly independent fluxes, this requires $\bar{B} = \bar{C} = 0$. For this purpose we use

$$\frac{1}{D(\phi)} = \left(\frac{1}{2D_S} + \frac{1}{2D_L} \right) + g(\phi) \left(\frac{1}{2D_S} - \frac{1}{2D_L} \right), \quad (17)$$

where $g(\phi)$ obeys $g(\phi = \pm 1) = \pm 1$ and $g(\phi) = -g(-\phi)$. This choice automatically eliminates the Kapitza resistance $\bar{C} = 0$ [14]. Then

$$\bar{B} = \alpha M - \frac{\gamma W}{2} \left(\frac{1}{2D_S} - \frac{1}{2D_L} \right), \quad (18)$$

where

$$\gamma = \int_{-\infty}^{\infty} \frac{dx}{W} \{1 - p[\phi_{\text{eq}}(x)]g[\phi_{\text{eq}}(x)]\}. \quad (19)$$

The condition $\bar{B} = 0$ is therefore provided by

$$M = M^* = \frac{\gamma W}{2\alpha} \left(\frac{1}{2D_S} - \frac{1}{2D_L} \right). \quad (20)$$

We understand at this point why the kinetic cross coupling was not needed ($M^* = 0$) for the Kapitza jump to be eliminated when $D(\phi) = D_S = D_L$ [1].

Using these choices of $D(\phi)$ and M , we find the remaining kinetic coefficient \bar{A} ,

$$\bar{A} = \frac{\alpha\tau}{W} - \frac{\beta W}{4} \left(\frac{1}{2D_S} + \frac{1}{2D_L} \right), \quad (21)$$

where

$$\beta = \int_{-\infty}^{\infty} \frac{dx}{W} \{1 - p^2[\phi_{\text{eq}}(x)]\} \simeq 1.40748. \quad (22)$$

Finally, kinetic effects may be fully eliminated ($\bar{A} = \bar{B} = \bar{C} = 0$) by the choice

$$\tau = \tau^* = \frac{\beta W^2}{4\alpha} \left(\frac{1}{2D_S} + \frac{1}{2D_L} \right). \quad (23)$$

The condition of stability $\Delta > 0$ sets an upper bound for the diffusional contrast if one requires equilibrium boundary conditions. Using the relation $(W\phi'_{\text{eq}})^2 = (1 - \phi_{\text{eq}}^2)^2/2$ and defining the contrast $\nu = (D_L - D_S)/(D_L + D_S)$, the inequality $\Delta = 1 - (M^*W\nabla\phi)^2 D(\phi)/\tau^* > 0$ reads close to equilibrium

$$1 < \frac{2\beta\alpha}{\gamma^2\nu^2} \min \frac{1 + \nu g(\phi_{\text{eq}})}{(1 - \phi_{\text{eq}}^2)^2}. \quad (24)$$

This means $|\nu| < \nu_{\text{max}}$, where ν_{max} is model dependent through $g(\phi)$ and the coefficient γ . In the case of a larger contrast ($D_S \ll D_L$), which is important for the solidification of alloys, one may use a one-sided model (see the relevant discussion in Ref. [10]).

Two remarks are in order. First, Almgren has shown that the correction to the conservation law due to the interface stretching effect is eliminated if $\int_{-\infty}^{\infty} dx [\sigma_{\text{eq}}(x) - \sigma_S/2 - \sigma_L/2] = 0$ [2]. This integral represents the equilibrium interface ‘‘adsorption’’ and vanishes as soon as $p(\phi) = -p(-\phi)$, which is the usual choice in phase-field models. In Ref. [15], the authors effectively add an even part to $p(\phi)$ in order to tune the kinetic coefficient \bar{B} , thus allowing for some interface adsorption. However, it is not physically sound to tune the kinetic effects using a thermodynamical degree of freedom of the model. Here, we tune the kinetic cross coefficient \bar{B} with the help of the cross coupling term M . Second, surface diffusion is another effect that alters the conservation law at the interface. In order to remove this effect, $D(\phi)$ should satisfy $\int_{-\infty}^{\infty} dx [D(\phi_{\text{eq}}) - D_S/2 - D_L/2] = 0$ [2]. The switching function should in this case be written $g(\phi) = g_0(\phi) + ag_1(\phi)$, where $g_0(\pm 1) = \pm 1$ and $g_1(\pm 1) = 0$, both being odd in order to recover $\bar{C} = 0$. Then the surface diffusion effect is eliminated with an appropriate choice $a = a^*(|\nu|)$ [2].

We now shortly summarize how the nondiagonal phase-field model is designed. With the switching function $p(\phi)$, one tunes the thermodynamic adsorption property of the interface that causes a kinetic correction to the conservation law due to interface stretching. With the diffusivity $D(\phi)$, one tunes the kinetic coefficient \bar{C} and the kinetic correction to the conservation law due to surface diffusion. With the cross coupling coefficient M , one tunes the kinetic coefficient \bar{B} . With the relaxation time τ , one tunes the kinetic coefficient \bar{A} . Here we have presented the procedure to eliminate *simultaneously* all the kinetic effects. Let us note that an interface anisotropy may be introduced in a routine way through the orientation dependences $W(\mathbf{n})$, $\tau(\mathbf{n})$ [1], and $M(\mathbf{n})$.

Numerical test. We now perform a numerical validation of the presented theory using a simple but illustrative example. We discuss the solidification of a thin film that may exchange heat with an environment at temperature $T_E < T_M$. Averaging the temperature in the film over its thickness, the problem becomes two dimensional. Moreover, we discuss the propagation of a flat solidification front, and the average dimensionless temperature in the film then obeys the one-dimensional diffusion equation

$$\dot{u}(x) = D_k u''(x) - \frac{u(x) - u_E}{\tau_v} \quad (25)$$

in phase $k = L, S$. We model the heat exchange with the environment using a simple linear law valid for small temperature differences between the film and the environment, i.e., for $-u_E = -c_P(T_E - T_M)/L \ll 1$. Here τ_v is the characteristic time for this exchange.

We focus on the case without Kapitza jump, $\bar{B} = \bar{C} = 0$, that requires $M = M^*$. Then, according to Eqs. (12) and (13), and due to the fact that $\sigma_L - \sigma_S = 1$, we have $u_S = u_L = u_i = -\bar{A}V$, where \bar{A} is given by Eq. (21).

In a comoving frame of reference with the interface located at the origin $x = 0$, phase S occupying $x < 0$ and phase L occupying $x > 0$, the solution in the quasistatic approximation ($\dot{u} = 0$) reads

$$\begin{aligned} u(x < 0) &= u_E + (u_i - u_E) \exp(x/\sqrt{D_S \tau_v}), \\ u(x > 0) &= u_E + (u_i - u_E) \exp(-x/\sqrt{D_L \tau_v}). \end{aligned} \quad (26)$$

Using the energy conservation at the interface, i.e., $V = D_S u'(x = 0^-) - D_L u'(x = 0^+)$, we find

$$V = V_{\text{eq}} \left(1 - \frac{\bar{A}(J_S + J_L)}{1 + \bar{A}(J_S + J_L)} \right), \quad (27)$$

where $J_S = \sqrt{D_S/\tau_v}$ and $J_L = \sqrt{D_L/\tau_v}$, and with

$$V_{\text{eq}} = -u_E(J_S + J_L) \quad (28)$$

being the velocity for equilibrium boundary conditions, i.e., when $u_i = 0$. Let us note that the quasistatic approximation that allows one to use Eqs. (26) is justified for $-u_E \ll 1$.

We perform phase-field simulations where the temperature evolution equation (10) is replaced by

$$\dot{u} = \nabla \cdot \{ D(\phi) [\nabla u + MW \phi \nabla \phi] \} + \frac{p'(\phi)}{2} \dot{\phi} - \frac{u - u_E}{\tau_v} \quad (29)$$

that takes into account the heat exchange with the environment. We use $u_E = -0.1$, $D_S \tau_v / W^2 = 200$, and different ratios $D_L/D_S = (1 + \nu)/(1 - \nu)$. We choose $g(\phi) = \phi$ (no need to eliminate surface diffusion here) for which $\nu_{\text{max}} \simeq 0.71$ [for example, for the choice $g(\phi) = p(\phi)$, one has $\nu_{\text{max}} \simeq 0.86$]. In Fig. 1, we present $(V - V_{\text{eq}})/V_{\text{eq}}$ as a function of $\mathcal{A}(J_S + J_L)$, where V is obtained from the simulations (symbols) and V_{eq} is given by Eq. (28). We compare with the analytical prediction (line) with V given by Eq. (27). The excellent quantitative agreement between simulations and analytics provides a strong support for the theory.

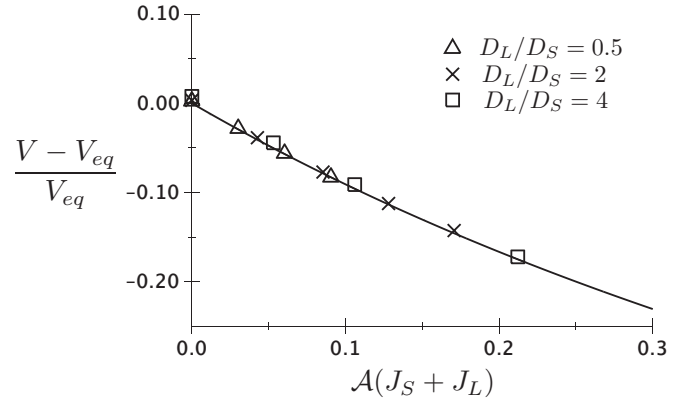


FIG. 1. $(V - V_{\text{eq}})/V_{\text{eq}}$ as a function of $\mathcal{A}(J_S + J_L)$ for different values of D_L/D_S from simulations (triangles, crosses, squares) and from analytics (line).

We have also performed two-dimensional simulations of the relaxation of a corrugated interface to its flat equilibrium, a situation that involves surface diffusion. The resulting spectrum has been compared with the analytical solution of the macroscopic problem. The numerical results agree well with the analytical predictions. These results will be published elsewhere.

Summary. In this Rapid Communication, we have achieved a realistic description of interface kinetics in a phase-field model with a diffusional contrast. This is enabled by a kinetic cross coupling (nondiagonal model) between the conserved and the nonconserved fields that is not present in classical diagonal models. This cross coupling provides a full correspondence between the number of independent kinetic parameters in the phase field and the macroscopic descriptions. This correspondence allows one to tune the kinetic effects as desired and, for example, allows one to eliminate them partly or fully. Using the case of the solidification of a pure substance, we first eliminate the Kapitza temperature jump at the interface and, second, fully eliminate interfacial kinetic effects. We obtain an excellent quantitative agreement between phase-field simulations and the analytical solution of the corresponding macroscopic approach using a simple but illustrative example. The extension of our results to multiphase systems where triple junctions are present might be challenging.

Acknowledgments. We are grateful to R. Spatschek for useful discussions. E.A.B. acknowledges the support of the Deutsche Forschungsgemeinschaft under Project No. SFB 917.

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