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Theory of dendritic growth in the presence of lattice strain

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We discuss elastic effects due to lattice strain which are a new key ingredient in the theory of dendritic growth for solid-solid transformations. Both thermal and elastic fields are eliminated by Green's function techniques, and a closed nonlinear integro-differential equation for the evolution of the interface is derived. We find dendritic patterns even without the anisotropy of the surface energy required by classical dendritic growth theory. In this sense, elastic effects serve as a new selection mechanism.

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Solvability theory has been very successful in predicting certain properties of dendritic growth and a number of related phenomena (see, for example, [1,2]). The solution of two-dimensional steady-state growth is described by a needle crystal, which is assumed to be close in shape to the parabolic Ivantsov solution [3]. If anisotropic capillary effects are included, a single dynamically stable solution is found for any external growth condition. This theory has also been extended to the three-dimensional case [4,5]. The capillarity is a singular perturbation, and the anisotropy of the surface energy is a prerequisite for the existence of the solution. In the case of isotropic surface energy, a dendritic solution does not exist and instead the so-called doublon structure is the solution of the problem [6,7].

Usually, the structural phase transitions in solids are accompanied by small lattice distortions leading to elastic deformations (for a review, see [8,9]). One of the well-known consequences is a thermodynamic elastic hysteresis—i.e., the splitting of the phase equilibrium point into two points: the point of the direct and inverse transition. This is mainly due to the coherency at the interface boundary, meaning that the lattice layers remain continuous through the boundary. Correspondingly, the hysteresis disappears without interface coherence [10,11]. However, the systematic investigation of the growth *kinetics* of such phase transitions is much less developed. Recently, pattern formation processes controlled by interface kinetics have been considered in [12] and the growth of spherical inclusions under elastic and thermal influences was investigated in [13] by means of the phase-field model.

In this article, we discuss the influence of elastic strain on dendritic growth in solids controlled by heat diffusion. Significant progress in the description of dendritic growth was made by the elimination of the thermal field using the Green's function technique. This allows one to obtain a closed equation for the interface evolution (see, for example, [14]). The crucial point of the present analysis is that the elastic field can also be eliminated by the corresponding Green's function technique. By these means, we derive here, as in classical dendritic growth theory, a single integro-differential equation for the shape of the interface which takes into account elastic effects. Then, we consider two simple examples of dilatational and shear transformations. We show that in the case of a pure dilatation, elastic effects lead only to a trivial shift of the transition temperature. However, for the case of shear transitions, we find dendritic patterns even without the anisotropy of the surface energy required by classical dendritic growth theory. In this sense, elastic effects serve as a new selection mechanism.

Let us consider the growth of a new β phase inside of an unbounded mother α phase. We denote the characteristic lattice strain (also known as the stress-free strain tensor), associated with the phase transition, by ϵ_{ik}^{0}. The free energy density in the initial α phase is

$$F_\alpha = F_\alpha^0(T) + \frac{\lambda}{2}\epsilon_{ii}^2 + \mu\epsilon_{ik}^2, \tag{1}$$

where $F_\alpha^0(T)$ is the free energy density without elastic effects, which depends only on the temperature T, ϵ_{ik} are the components of the strain tensor, and λ and μ are the elastic moduli of isotropic linear elasticity. The free energy density of the growing β phase is given by

$$F_\beta = F_\beta^0(T) + \frac{\lambda}{2}(\epsilon_{ii} - \epsilon_{ii}^0)^2 + \mu(\epsilon_{ik} - \epsilon_{ik}^0)^2. \tag{2}$$

Here, we neglect the difference between the elastic coefficients in the two phases. We also assume that the elastic effects are small—i.e., $\epsilon_{ik}^0 \ll 1$. Since in our description the reference state for both phases is the undeformed initial phase, the coherency condition at the interface reads $u_i^{(\alpha)} = u_i^{(\beta)}$, where u_i is the displacement vector. Mechanical equilibrium at the interface demands $\sigma_{nn}^{(\alpha)} = \sigma_{nn}^{(\beta)}$ and $\sigma_{n\tau}^{(\alpha)} = \sigma_{n\tau}^{(\beta)}$, $\sigma_{ns}^{(\alpha)} = \sigma_{ns}^{(\beta)}$. Here the indices n and (τ, s) denote the normal and tangential directions with respect to the interface; the stress tensor is defined as

$$\sigma_{ik} = \frac{1}{2}\left(\frac{\partial F}{\partial \epsilon_{ik}} + \frac{\partial F}{\partial \epsilon_{ki}}\right).$$

The condition of phase equilibrium requires the continuity of a new potential

$$\tilde{F} = F - \sigma_{nn}\epsilon_{nn} - 2\sigma_{n\tau}\epsilon_{n\tau} - 2\sigma_{ns}\epsilon_{ns}$$

across the flat interface [15], which takes into account the coherency constraint. In the general case of curved interfaces, the surface energy γ has also to be incorporated, and the phase equilibrium condition for each interface point in the case of isotropic surface energy reads

$$\tilde{F}_\alpha - \tilde{F}_\beta - \gamma\kappa = 0, \tag{3}$$

where κ is the local curvature of the interface.

Let us denote by $\tilde{\sigma}_{ik}$ the stress tensor which is related to the strain field ϵ_{ik} by the usual Hooke's law. Then, the stress in the new β phase can be written as $\sigma_{ik}^{(\beta)}=\tilde{\sigma}_{ik}^{(\beta)}-\sigma_{ik}^{0}$, while it remains unchanged in the mother α phase: $\sigma_{ik}^{(\alpha)}=\tilde{\sigma}_{ik}^{(\alpha)}$. The tensor σ_{ik}^{0} is related to the lattice strain ϵ_{ik}^{0} by Hooke's law

$$\sigma_{ik}^{0}=\frac{E}{1+\nu}\left(\epsilon_{ik}^{0}+\frac{\nu}{1-2\nu}\delta_{ik}\epsilon_{ll}^{0}\right), \tag{4}$$

where E is the Young's modulus and ν is the Poisson ratio. Thus, the mechanical equilibrium conditions at the interface require us to introduce the interface force density in the equilibrium equation $\partial\tilde{\sigma}_{ik}/\partial x_k=f_i$:

$$f_i=(\tilde{\sigma}_{ik}^{(\beta)}-\tilde{\sigma}_{ik}^{(\alpha)})n_k=\sigma_{ik}^{0}n_k, \tag{5}$$

where the normal vector $\mathbf{n}$ points from the β phase into the α phase. Because the forces act only at the interface, the displacement field can be written as an integral over the interface surface:

$$u_i(\mathbf{r})=\int G_{ik}(\mathbf{r},\mathbf{r}')f_k(\mathbf{r}')dS',$$

where $G_{ik}(\mathbf{r},\mathbf{r}')$ is the so-called Green's tensor (see, for example, [16]). Then, the strain field is determined by

$$\epsilon_{ik}(\mathbf{r})=\frac{1}{2}\int\left(\frac{\partial G_{km}(\mathbf{r},\mathbf{r}')}{\partial x_i}+\frac{\partial G_{im}(\mathbf{r},\mathbf{r}')}{\partial x_k}\right)f_m(\mathbf{r}')dS'. \tag{6}$$

This strain is fully defined by the corresponding Green's tensor and Eqs. (4) and (5). The strain components $\epsilon_{\tau\tau}$, $\epsilon_{s\tau}$, and ϵ_{ss} are continuous across the interface, while the other components are discontinuous:

$$\epsilon_{nn}^{(\beta)}-\epsilon_{nn}^{(\alpha)}=\epsilon_{nn}^{0}+\frac{\nu}{1-\nu}(\epsilon_{\tau\tau}^{0}+\epsilon_{ss}^{0}),$$

$$\epsilon_{n\tau}^{(\beta)}-\epsilon_{n\tau}^{(\alpha)}=\epsilon_{n\tau}^{0},\qquad \epsilon_{ns}^{(\beta)}-\epsilon_{ns}^{(\alpha)}=\epsilon_{ns}^{0}.$$

Taking these jumps of the strain field at the interface into account, one can find the elastic contribution to the local equilibrium condition, Eq. (3). A tedious but straightforward calculation leads to

$$\delta\tilde{F}^{(el)}=\tilde{F}_{\alpha}^{(el)}-\tilde{F}_{\beta}^{(el)}=\sigma_{ik}^{(0)}\epsilon_{ik}^{(\alpha)}-\frac{E[(\epsilon_{\tau\tau}^{0})^2+(\epsilon_{ss}^{0})^2+2\nu\epsilon_{ss}^{0}\epsilon_{\tau\tau}^{0}+2(1-\nu)(\epsilon_{s\tau}^{0})^2]}{2(1-\nu^2)}, \tag{7}$$

where $\epsilon_{ik}^{(\alpha)}$ is the strain in the α phase at the interface. Note that the expression above is a complicated integro-differential functional of the interface shape.

For simplicity, we consider transitions in pure materials and assume that the heat diffusion constants are equal in both phases (the so-called symmetrical model). Without loss of generality, we assume that the β phase is the low-temperature phase. We introduce the dimensionless temperature field $w=c_p(T-T_\infty)/L$, where L is the latent heat, c_p is the heat capacity, and T_∞ is the temperature in the α phase far away from the interface. The temperature field w obeys the following heat diffusion equation and boundary conditions:

$$D\nabla^2 w=\partial w/\partial t, \tag{8}$$

$$v_n=D\mathbf{n}(\nabla w_\beta|_{int}-\nabla w_\alpha|_{int}), \tag{9}$$

$$w|_{int}=\Delta-d_0\kappa+T_{eq}c_p\delta\tilde{F}^{el}/L^2, \tag{10}$$

where $d_0=\gamma T_{eq}c_p/L^2$ is the capillarity length, κ is the curvature of the interface, which assumed to be positive for convex interfaces, D is the thermal diffusion constant, and T_{eq} is the equilibrium temperature for the flat interface without elastic effects—i.e., it is determined by the condition $F_{\alpha}^{0}(T_{eq})=F_{\beta}^{0}(T_{eq})$. We also introduce the dimensionless undercooling $\Delta=c_p(T_{eq}-T_\infty)/L$. The physics underlying Eqs. (8)–(10) is quite simple. The interface moving with normal velocity v_n releases latent heat. The requirement of heat conservation at the interface gives Eq. (9). The local thermodynamical equilibrium at the interface, Eq. (3), implies Eq. (10). This equation then gives the equilibrium value of the temperature at the interface, taking into account the curvature corrections and elastic effects, while additional dissipation due to the interface kinetics is neglected. The thermal field can be eliminated by using Green's function techniques (see, for example, [14]), and consequently together with a proper Green's tensor $G_{ik}(\mathbf{r},\mathbf{r}')$ for the elastic field, the set of equations (6)–(10) can be incorporated into a single integro-differential equation for the shape of the solid-solid interface.

First, we consider the dilatational case $\epsilon_{ik}^{0}=\varepsilon\delta_{ik}$, where the bond lengths of the new phase are uniformly longer or shorter in all directions in comparison to the original phase. In this case, the elastic contribution to the local equilibrium condition at the interface, $\delta\tilde{F}^{(el)}=-\varepsilon^2E/(1-\nu)$, is a constant along the interface for any interface shape. This result corresponds to the elastic hysteresis mentioned above, and it can be obtained using the analogy of this elastic problem to the problem of thermal expansion for a given temperature field [16]. As a consequence, the equilibrium interface temperature is shifted by a constant value and the problem is equivalent to the problem of the classical dendritic growth. The anisotropy of the surface energy is a prerequisite for the existence of the dendritic solution in this case, as we have already mentioned above. Note that this exact result is valid only under the assumptions of our model—i.e., the isotropic elasticity and equal elastic moduli in both phases. It also serves as a nontrivial check of our numerical code.

Let us consider now a simple type of transition in hexagonal crystals involving shear strain. For the transitions lowering the symmetry from C_6 to C_2, the shear strain in the basic plane appears. This is the case, for example, in hexagonal-orthorhombic transitions in ferroelastics (see [17] and references therein). Let the principal axis C_6 be orientated in the z direction. Although the general approach presented above is valid in the three-dimensional case, we assume from now on that the system obeys translational invariance in this direction, and thus, it is effectively two dimensional. By proper choice of the crystal orientation around the main axis in the

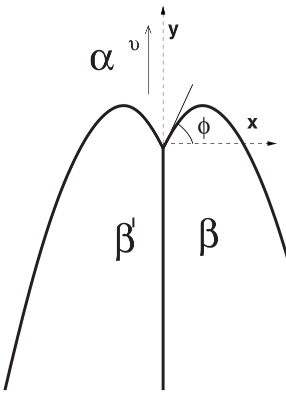

FIG. 1. Steady-state growth of a bicrystal (β/β'). The structure propagates with a constant velocity v along the y axis. The β phase corresponds to $\theta=2\pi/3$, and the β' phase corresponds to $\theta=-2\pi/3$.

initial phase, we obtain the new phase in three possible states, having the following nonvanishing components of the strain tensor ϵ^0_{ik}:

$$\epsilon^0_{xx} = -\epsilon^0_{yy} = \varepsilon \cos 2\theta, \quad \epsilon^0_{xy} = \varepsilon \sin 2\theta, \tag{11}$$

where the angle θ has three possible values: $\theta=0$, $\pm 2\pi/3$. The situation of $\theta=0$ corresponds to single-crystal growth and $\theta=\pm 2\pi/3$ to bicrystal growth (Fig. 1). Because the elasticity of hexagonal crystals is axisymmetric in the harmonic approximation and $\epsilon^0_{iz}=\epsilon_{iz}=0$, we can use the isotropic theory of elasticity—i.e., expressions for the free energy, Eqs. (1) and (2), remain valid [16]. The moduli of the effective isotropic elasticity, λ and μ, can be expressed in terms of the elastic constants of the original hexagonal crystal. The two-dimensional Green's tensor for isotropic materials is [18]

$$G_{ik}(\mathbf{r}) = \frac{1+\nu}{4\pi(1-\nu)E}\left(\frac{x_i x_k}{r^2} - (3-4\nu)\delta_{ik}\ln(r)\right). \tag{12}$$

Eliminating the thermal field, we obtain the steady-state equation for the shape of the solid-solid interface. In the comoving frame of reference, this equation reads

$$\Delta - \frac{d_0\kappa}{R} + \frac{T_{eq}c_p\delta\tilde{F}^{el}}{L^2} = \frac{p}{\pi}\int dx' \exp\{-p[y(x)-y(x')]\}K_0(p\eta), \tag{13}$$

where $\eta=\{(x-x')^2+[y(x)-y(x')]^2\}^{1/2}$, K_0 is the modified Bessel function of the third kind in zeroth order, and $p=vR/2D$ is the Peclet number. All lengths are reduced by the radius of the curvature, R, of asymptotic Ivantsov parabola.

In the asymptotic region $(|x|\to\infty)$, the strain ϵ_{ik} decays and the local contribution to $\delta\tilde{F}^{el}$ [second term in Eq. (7)] has a constant value, $\delta\tilde{F}^{el}=-E(\epsilon^0_{yy})^2/2(1-\nu^2)$. It follows from this relation that the temperature shift (elastic hysteresis) for the growth of a single crystal is 4 times larger than for a bicrystal. Although both bicrystal configurations $(\beta/\beta'$ and $\beta'/\beta)$ are energetically equivalent far from the tip, the symmetry is broken by the choice of the propagation direction. Therefore, in the following we will discuss the most favorable configuration of bicrystals [12], as presented in Fig. 1.

Let us introduce the shifted, due to the elastic hysteresis, undercooling

$$\tilde{\Delta} = \Delta - \Delta_{el}, \quad \Delta_{el} = T_{eq}c_p E\varepsilon^2/8(1-\nu^2)L^2.$$

The dimensionless parameter Δ_{el} describes the strength of the elastic effects. The relation between this shifted undercooling $\tilde{\Delta}$ and the Peclet number is given by the two-dimensional Ivantsov formula [3] $\tilde{\Delta}=\sqrt{p\pi}\exp(p)\mathrm{erfc}(\sqrt{p})$.

The presence of the twin (β/β') boundary leads to additional effects. First, calculating the strain field ϵ_{ik}, which enters in the expression for $\delta\tilde{F}^{(el)}$, in addition to the integrals along α/β and α/β' interface, the integration should also be performed along the twin boundary β/β'. The force density at this boundary is $f_x=0$, $f_y=E\sqrt{3}/(1+\nu)$. Second, the equilibrium angle ϕ at the triple junction (Fig. 1) is given by Young's law: $2\gamma\sin\phi=\gamma_b$, where γ is the surface energy of the β/α interface and γ_b is the surface energy of the twin boundary.

Equation (13) is a complicated nonlinear integro-differential equation for the interface shape. We should find a solution of this equation which has a proper angle ϕ at the triple junction and which is close to the Ivantsov parabola $(y=-x^2/2)$ in the tail region. Note that without elastic effects, this problem is equivalent to the classical dendritic growth problem with isotropic surface tension. The latter does not have a solution with angles $\phi\geqslant 0$ [1,2]. This statement can be expressed in the following form. For any given positive values of the Peclet number p and the so-called stability parameter $\sigma=d_0/pR$, the symmetric solution which is close to the Ivantsov parabola in the tail region has an angle at the tip $\phi=f(\sigma,p)<0$. The limit $\sigma=0$ and $\phi=0$ is a singular limit for that problem. For example, Meiron [19] calculated the angle ϕ as a function of σ for several values of the Peclet number with isotropic surface tension numerically and found that the angle $\phi<0$ for any positive σ.

Now, we discuss the numerical results obtained by the solution of Eq. (13) in the spirit of Ref. [19]. In the important regime of small Peclet numbers, the eigenvalue $\sigma=\sigma^*(\phi,\Delta_{el},p)$ depends only on the ratio Δ_{el}/p for a fixed angle ϕ. While the strength of the elastic effects is assumed to be small, $\Delta_{el}\ll 1$, the control parameter Δ_{el}/p can be varied in a wide region in the limit of small p. The eigenvalue σ^* as a function of Δ_{el}/p for two values of the angle, $\phi=0$ and $\phi=\pi/6$, is shown in Fig. 2. The situation with $\phi\approx 0$ is realized if $\gamma_b\ll\gamma$, while $\phi\approx\pi/6$ corresponds to $\gamma_b\approx\gamma$. The Poisson ratio was fixed at $\nu=1/3$.

The most remarkable feature of these results is that we do find dendritic solutions for the isotropic surface tension in the presence of elastic effects. In this sense, elastic effects serve as a new selection mechanism. We note that σ^* becomes large for large values of Δ_{el}/p, while in classical dendritic growth σ^* is always small, being controlled by tiny

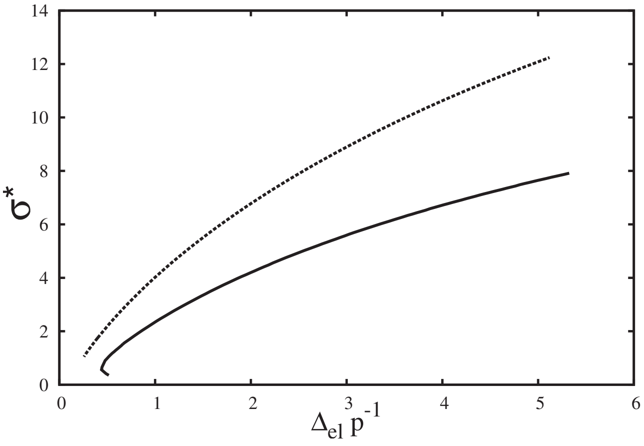

FIG. 2. Stability parameter σ^* versus Δ_{el}/p for two values of ϕ: the dashed line corresponds to $\phi=0$, and the solid line corresponds to $\phi=\pi/6$.

anisotropy effects. Thus, the growth velocity $v=2D\sigma^* p^2/d_0$ can be much larger due to elastic effects, compared to classical dendritic growth.

In the case $\phi=\pi/6$, the solution exists only beyond some critical value of the control parameter, Δ_{el}/p. The lower branch of this solution has a negative curvature near the triple junction, and it is presumably unstable. The general structure of the theory suggests that in the case $\phi=0$, the curve should start from the origin ($\sigma=0$, $\Delta_{el}/p=0$) which is a singular point of the problem. However, numerics becomes very difficult in the vicinity of this point.

We have also performed several runs for single-crystal growth [$\theta=0$ in Eq. (11)]. As in classical dendritic growth, we have not found solutions with a smooth tip, $\phi=0$. Moreover, for negative values of ϕ, where solutions exist and correspond to the growth along a grain boundary in the mother α phase [20], the selected stability parameter σ^* is a decreasing function of Δ_{el}/p. This is in strong contrast with the results for bicrystal growth.

In summary, we present a theory of dendritic growth for solid-solid transformations where lattice strain is a key ingredient. We find that, in this case, dendritic patterns are selected even without the anisotropy of the surface energy required by classical dendritic growth theory. Of course, the discussed elastic effects also introduce an "effective anisotropy" of the system. However, the physics and structure of selection theory for the two mechanisms, anisotropy of surface energy and elastic effects, are fundamentally different. Moreover, elastic effects lead to a much more robust selection mechanism compared to the tiny effects of anisotropy of the surface energy. We hope that our results will stimulate new experimental and theoretical investigations in this interesting field. Specifically, it would be interesting to observe bicrystal growth in systems exhibiting a hexagonal-orthorhombic transition and also to measure the value of the stability parameter, which should be much larger compared to classical dendritic growth theory.

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