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Influence of interphase boundary anisotropy on bulk eutectic solidification microstructures

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ABSTRACT

The solidification of eutectic alloys generally produces two-phase microstructures. Their morphology is determined by the dynamics of the solid-liquid interfaces at the crystallization front. At the triple lines, where the liquid and the two solid phases meet, solid-liquid and solid-solid surface free energies are in local equilibrium. We perform three-dimensional phase-field simulations with a multi-phase-field model in which the surface free energies can be independently controlled. We find that an anisotropy of the interphase boundary (solid-solid) energy has a strong effect on the microstructural patterns. The lamellae tend to align with directions of minimal interphase boundary energy. For a two-fold anisotropy, as generally expected for interphase boundaries between two cubic crystals, regular lamellar arrays are formed, in strong contrast to the labyrinth patterns observed in isotropic systems. If two different grains compete, the one with the lowest interphase boundary energy always overgrows the other. These results are consistent with observations in bulk metallic eutectic samples, namely, the frequent occurrence of large regular lamellar arrays and the prevalence of grains with special orientation relationships.

1. Introduction

Eutectic alloys solidify naturally into two-phase composites that find application in many engineering domains, particularly in high-strength materials [1,2]. During the crystallization, the two phases, which are of different chemical compositions, exchange solute by diffusion through the liquid region adjacent to the growth front. The geometric arrangement of the two phases results from a pattern formation process that is governed by the interplay between diffusion and capillarity. Many features of this coupled growth mode are well understood since the works of Zener [3], Hillert [4], and Jackson and Hunt [5]. Two basic patterns of the phases are widespread: if the volume fractions of the two solid phases are comparable, they self-organize in alternating platelets (lamellae); otherwise, rods of the minority phase are dispersed in a continuous matrix of the other phase [5—7]. Other patterns that exhibit more complex symmetries or are disordered have also been investigated in many experiments and numerical simulations [8—14].

There are number of experimental observations, however, that are not accounted for by these theories and simulations. One example is the fact that lamellae often grow tilted, that is, they form a finite angle with respect to the direction of the temperature gradient. This and other phenomena can be linked to crystallographic effects, which have largely been omitted in theories and models up to now. A eutectic composite generally consists of *eutectic grains*, in which the orientation of the crystallographic axes is the same for all the domains (lamellae, rods) of a given phase [15]. This implies that the relative orientation of the two solid phases is also fixed in each (eutectic) grain; it may vary, however, between different grains of the same sample.

Regular eutectics have solid-liquid interfaces that are rough on the atomic scale, which are expected to be only weakly anisotropic. In contrast, the solid-solid interfaces (interphase boundaries, IB) can be strongly anisotropic and may exhibit facets even in regular eutectics. It has been found that the solidification dynamics strongly depends on the properties of the IB [16]. So-called *floating grains*, in which the IB are weakly anisotropic, conform to the theoretical predictions made by the theories with isotropic interfaces and grow parallel to the temperature gradient; in contrast, in *locked grains*, the IB follow specific crystallographic directions, so that lamellae can be strongly tilted with respect to the direction of

the temperature gradient [17].

Recently, this situation was examined more quantitatively in experiments using a rotating directional solidification setup [17], and was also analyzed theoretically [18]. The theory predicts that anisotropy of the IB alone (whereas the solid-liquid interfaces remain isotropic) can lead to the observed tilted growth. This is quite remarkable, since the anisotropy of the IB enters the classic free-boundary problem of eutectic growth only through the local equilibrium condition at the trijunction point. The prediction for the growth direction was verified by direct numerical simulations of eutectic growth in two dimensions, using both boundary-integral and phase-field methods [19,20].

Here, we present three-dimensional phase-field simulations of eutectic solidification performed with the same model as in Ref. [19]. Eutectic solidification with anisotropic interfaces has already been studied recently by phase-field modelling [21,22], but with models in which the anisotropy of solid-liquid and solid-solid interfaces cannot easily be controlled separately. In our model, we can independently set the anisotropy of each interface, which allows us to probe the fundamental mechanisms of pattern formation more precisely. We focus here on systems with isotropic solid-liquid and anisotropic solid-solid interfaces. With respect to Ref. [19], the main new question that we address here is the effect of an anisotropy in the plane of the isotherms (azimuthal plane), which can obviously not be investigated by two-dimensional simulations. Such an anisotropy breaks the rotational symmetry of the problem in the azimuthal plane, and can thus be expected to have a strong effect. Here, we limit ourselves to anisotropy functions with simple regular m-fold symmetry ($m = 2, 4$) and to a model eutectic alloy with symmetric phase diagram, and focus on qualitative differences between eutectic growth with isotropic and anisotropic IB.

We find that the dynamics of pattern formation is strongly altered by the presence of anisotropy. The lamellae tend to align with directions of minimal IB energy. For a four-fold anisotropy, this leads to two families of lamellae that are perpendicular to each other; for two-fold symmetry, a single orientation is favored, and a well-ordered lamellar state is rapidly reached. This is in strong contrast to the "labyrinth" structures observed in isotropic systems [11,13]. The rate of ordering increases with the strength of the anisotropy.

We also perform simulations in which two grains with different anisotropy functions compete, in order to study the question of grain selection. Anisotropic grains with low-energy IB tend to rapidly outgrow the others. These results are compatible with the experimental observations that eutectic grains with special orientation relationships between the phases are frequently observed in bulk samples, and that regions of ordered and perfectly parallel lamellae are very frequent [23–27].

The structure of the article is as follows. In Sec. 2, we briefly describe the main elements of the phase-field model used to simulate anisotropic eutectic growth. Then, we outline the details of the simulation procedure, the choice of parameters and some details of our data analysis. In Sec. 3, we present the results and discuss the mechanisms of the microstructural evolution. Following an overall discussion in Sec. 4, we give conclusions and perspectives in Sec. 5.

2. Methods

2.1. Phase-field framework

Phase-field models have become a standard tool for the modelling of microstructure evolution during solidification and solid-state transformations [28–30]. The principle of this method is to describe the geometry of the evolving structures by time-dependent scalar phase fields that take uniform values in the bulk and vary continuously through the interfaces. Thus, front tracking is avoided, and the model equations can be integrated in time by simple algorithms that are easy to parallelize. Here, we have used a recent multi-component grand-canonical phase-field formulation that has been detailed elsewhere [19,31,32]. We will briefly describe the ingredients that are most relevant to the anisotropic growth of a binary eutectic alloy solidifying into α and β solids.

We assume that there is no convection in the liquid; solute is transported in the liquid by diffusion only, while the diffusion in the solids is neglected (one-sided model). $N = 3$ phase fields, namely, ϕ_α, ϕ_β and ϕ_l, equivalent to local volume fractions, describe the structures, and obey the constraint $\sum_{i=1}^{N} \phi_i = 1$.

The starting point is a grand-canonical free energy functional,

$$\Omega = \int_V \varepsilon a(\phi, \overrightarrow{\nabla}\phi) + \frac{1}{\varepsilon} \mathscr{W}(\phi) + \omega(\mu, T, \phi), \qquad (1)$$

where ε is a length scale parameter related to the numerical interface thickness, and $a(\phi, \overrightarrow{\nabla}\phi)$ is the gradient energy density given by

$$a(\phi, \overrightarrow{\nabla}\phi) = \sum_{i<j}^{N} \gamma_{ij} \left[a_c^{ij}\left(\hat{q}_{ij}\right) \right]^2 |\overrightarrow{q}_{ij}|^2. \qquad (2)$$

Here, $\overrightarrow{q}_{ij} = \phi_i \overrightarrow{\nabla}\phi_j - \phi_j \overrightarrow{\nabla}\phi_i$ is a vector normal to the i-j interface, $\hat{q}_{ij} = \overrightarrow{q}_{ij}/|\overrightarrow{q}_{ij}|$ the corresponding unit vector, and $a_c^{ij}(\hat{q}_{ij})$ is the anisotropy function of the i-j interface. For isotropic interfaces, $a_c^{ij} \equiv 1$; γ_{ij} is the isotropic part (mean value) of the interface energy. Moreover, $\mathscr{W}(\phi)$ in Eq. (1) is a multi-obstacle potential,

$$\mathscr{W}(\phi) = \begin{cases} \dfrac{16}{\pi^2} \displaystyle\sum_{i<j}^{N} \gamma_{ij}\phi_i\phi_j + \displaystyle\sum_{i<j<k}^{N} \gamma_{ijk}\phi_i\phi_j\phi_k & \text{if} \quad \phi \in \Sigma \\ \infty \text{ elsewhere}. \end{cases} \qquad (3)$$

The simplex Σ is bounded by $\phi_i \geq 0$ and $\sum_i^N \phi_i = 1$, and γ_{ijk} is a third-order potential term which prevents the appearance of any unwanted "foreign" phases in the binary interfaces.

The function $\omega(\mu, T, \phi)$ in Eq. (1) is the grand-canonical potential, where μ is the diffusion potential (the thermodynamic conjugate of the concentration field c). For each phase, ω_i is the Legendre transformation of the concentration-dependent Helmholtz free energy density, f_i. The complete function ω is then obtained by an interpolation between the phases,

$$\omega(\mu, T, \phi) = \sum_{i=1}^{N} \omega_i(\mu, T) h_i(\phi) \qquad (4)$$

$$\omega_i(\mu, T) = f_i - \mu c, \qquad (5)$$

with $h_i(\phi) = \phi_i^2(3 - 2\phi_i) + 2\phi_i\phi_j\phi_k$ a weight function that satisfies $\sum_{i=1}^{N} h_i(\phi) = 1$. For the free energies, f_i, we take parabolas with equal curvatures, as detailed in Refs. [19,33].

The diffusion potential obeys a diffusion equation with a source term. An anti-trapping current $\overrightarrow{j}_{at}$ is added to the diffusion currents to counteract spurious solute trapping at the interfaces and thus guarantee the correct thin interface limit [32,34,35]: