



Phase-field simulations and geometrical characterization of cellular solidification fronts



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ABSTRACT

The structure and dynamics of cellular solidification fronts produced during the directional solidification of dilute binary alloys are studied by phase-field simulations. A quantitative phase-field model in conjunction with a multi-scale simulation algorithm allows us to simulate arrays with 10–40 cells in three dimensions on time scales that are long enough to allow for a significant reorganization of the array. We analyze the geometry of the complex two-phase structure (mushy zone) and extract the fraction of solid and the connectivity of the two phases as a function of depth. We find a transition from stable arrays at high values of the crystalline anisotropy to unsteady arrays at low anisotropy that continuously exhibit tip splitting and cell elimination events.

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1. Introduction

The solidification of alloys can produce patterns of great geometrical complexity, such as dendrites, cells, or multi-phase composites. The study of these structures is a classic subject in metallurgy, because their presence leads to an inhomogeneous distribution of alloy components (microsegregation), which has an important influence on the properties of the finished material [1,2]. Besides this practical aspect, microstructure development is also an example for spontaneous pattern formation out of equilibrium [3]. Therefore, solidification experiments can be used to investigate fundamental aspects of self-organization.

One of the simplest situations that can be studied is the directional solidification of a dilute binary alloy: a temperature gradient G is set up between two furnaces, and the alloy sample is pulled with constant velocity V from the hot to the cold zone. For dilute alloys, a well-known sequence of morphological transitions takes place with increasing velocity V . Below a critical value V_c , the interface remains planar, whereas above V_c , a morphological instability (Mullins-Sekerka instability [4]) takes place and leads to the emergence of complex patterns. For a certain range of velocities, the interface adopts a cellular morphology, that is, smooth “fingers” of solid are separated by grooves of liquid. If the velocity is further increased, the cells become more pointed, and finally develop into dendrites that exhibit lateral branches. For even higher velocities, there are further transitions back to

cells and finally to a planar interface (absolute stability) [1]. This high-velocity regime will not be addressed here.

The properties of these various morphologies, as well as the transitions between them, have been extensively studied by experiments, theory, and modelling. On the experimental side, a large amount of work has been devoted to the solidification of transparent organic alloys in thin samples [5–16]. This technique allows for a direct *in situ* observation by optical microscopy. Furthermore, if the sample thickness is smaller than the typical spacing between the cells or dendrites, two-dimensional models are often a good approximation which facilitates both theoretical analysis [17,18] and numerical modelling [13,19–25]. For a theoretical analysis, a good starting point are the spatially periodic solutions of the growth problem, characterized in two dimensions by a single parameter—the spacing. Such solutions generally exist over a large range of spacings, and are stable within a certain spacing interval that depends on the control parameters G and V [9,16,21]. Outside of this stable range, various secondary instabilities can occur, for example cell elimination for small spacings and oscillatory instabilities for large spacings [8,11,21]. In numerical simulations it was found (both in two and three dimensions) that the stability limits strongly depend on the anisotropy of surface free energy that is due to the crystal structure [12,21,26].

Since there is a range of possible spacings for each set of control parameters, it is natural to ask which one will be obtained in experiments. To answer this question, it turns out that it is necessary to follow the front dynamics from its starting point. In experiments, the initial condition is usually the equilibrium state of the system at rest, that is, an equilibrium interface at its equilibrium liquidus isotherm. Theories and models often start from the unstable steady state of the planar interface, because the

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initial stages of the dynamics (the development of the linear instability) are well understood for this case. In both scenarios, for typical parameters used in experiments, the final cell spacing is much larger than the wavelength of the fastest growing unstable mode. As a result, many small cells develop at the onset of the instability, and then the front undergoes numerous cell eliminations until it reaches a steady state. This scenario, which was analyzed more quantitatively by Warren and Langer [27], is in qualitative agreement with the thin-sample experiments [9,11,16], and was also confirmed by large-scale two-dimensional phase-field simulations [24]. These works also clarified that the final spacing can to some degree be controlled by suitable changes of the growth velocity during the experiment.

Much less information is available for three-dimensional fronts, for various reasons. In experiments with bulk samples, convection is always present, which considerably complicates the interpretation of the results. Furthermore, a direct *in situ* visualization is much more difficult than in thin samples. Both of these problems can be circumvented by experiments carried out in microgravity and equipped with innovative optical instruments [28,29]. On the modelling side, three-dimensional phase-field simulations of extended systems are still computationally costly, and only a few studies are currently available. Of these, Refs. [30,26] were carried out with phase-field models that do not describe alloy solidification well, because the solute diffusivity was taken identical in both phases. Works that use quantitative models for alloy solidification are either limited to the vicinity of the cellular bifurcation [31] or to confined systems [32–34].

Here, we present preliminary results of three-dimensional phase-field simulations of dilute binary alloys in domains that are large enough to contain between 15 and 40 cells, such that the effect of confinement (if present) is weak. The simulations are carried out using an efficient quantitative model for alloy solidification [35] and a multi-scale simulation algorithm that uses a cloud of random walkers to represent the large-scale diffusion field far from the solidification front [36,37]. This allows us to reach simulation times that are large enough for significant reorganization of the cellular array to occur. As a result, the pattern reaches either a steady state or a dynamical state in which the average geometry of the cells (spacing, tip position, etc.) does not evolve in time any more.

Various geometric properties of the front are then extracted. Obviously, the first interesting property is the number and shape of cells. For a reasonably complete characterization of the front geometry, properties such as the statistics of distances between the cell tips, of the number of neighbors, or of the segment lengths of the minimal spanning tree (see for example Ref. [29]) would be of interest. However, our domains are still too small for such statistical analyses to be significant. Therefore, we only extract the average spacing and compare its behavior to the available standard scaling theory [2].

We also study the fraction of solid and the connectivity of the solid phase as a function of depth. While these quantities are highly important for the macroscopic description of mushy zones, they have been little studied so far because they are difficult to obtain from experiments. In particular, only when the flanks of the cells have coalesced, a continuous “skeleton” of solid is formed that can sustain mechanical stresses. We determine the depth at which the solid percolates in the plane perpendicular to the growth direction. This point marks the transition from a liquid-like to a solid-like behavior and can therefore give an indication for the extension of the mushy zone.

Finally, we also study the influence of the crystalline anisotropy on the cellular patterns. We confirm the earlier results obtained with a qualitative model [26]: only for relatively high anisotropies, the front rapidly converges to a steady state. For the values of the

anisotropy typically found in metals [38], the front continually exhibits cell elimination and cell splitting events and does not reach a steady state on the time scale of our simulations. This raises the question whether stable arrays of deep cells can exist in experiments.

The remainder of the paper is organized as follows: in Section 2, we briefly present the sharp-interface and phase-field models. In Section 3, we display our results and discuss their implications in the light of available literature, and Section 4 is devoted to conclusions and perspectives for future work.

2. Models

Both the free-boundary problem and the phase-field model for dilute alloy solidification have been discussed in detail in previous publications [35,31]; therefore, we will limit ourselves here to a summary description.

2.1. Free-boundary problem

We consider the solidification of a dilute binary alloy made of substances A and B, with an idealized phase diagram that consists of straight liquidus and solidus lines of slopes m and m/k , respectively, where k is the partition coefficient. The concentrations c_s and c_l (in molar fractions) of impurities B at the solid and liquid side of the interfaces satisfy the partition relation,

$$c_s = kc_l. \quad (1)$$

For the directional solidification of a sample with composition c_∞ , at a flat steady-state interface we have $c_s = c_\infty$ and $c_l = c_\infty/k$ because of global mass conservation. In the absence of kinetic effects, the interface temperature is equal to $T_0 = T_m - |m|c_\infty/k$ (the solidus temperature), where T_m is the melting temperature of pure A. We choose T_0 as reference temperature, and the corresponding concentration $c_0 = c_\infty/k$ as reference concentration.

We use the “frozen-temperature approximation”, in which the temperature field in the sample is externally imposed,

$$T(z) = T_0 + G(z-Vt), \quad (2)$$

where the temperature gradient G is aligned with the z direction and the sample is pulled with a constant speed V . Furthermore, we neglect convection in the liquid. Hence, the solidification dynamics is entirely governed by solute diffusion.

For convenience, we use a rescaled concentration field

$$U = \frac{c-c_0}{(1-k)c_0}. \quad (3)$$

In the one-sided model, where solute diffusion is neglected in the solid, the dynamics of the solid–liquid interface and the concentration field in the liquid are given by the following free boundary problem:

$$\partial_t U = D\nabla^2 U, \quad (4)$$

$$[1 + (1-k)U]V_n = -D\partial_n U|_l, \quad (5)$$

$$U_{\text{int}} = -d_0 \sum_{i=1}^2 \left[a(\hat{n}) + \frac{\partial_2 a(\hat{n})}{\partial \theta_i^2} \right] \frac{1}{R_i} - \frac{z-Vt}{l_T}. \quad (6)$$

Here, Eq. (4) describes solute diffusion in the liquid with a constant diffusivity D . Eq. (5) is the Stefan condition at the solid–liquid interface which expresses mass conservation: the amount of solute rejected by the solid, which grows with normal velocity V_n , must be balanced by the solute flux on the liquid side; ∂_n denotes the derivative normal to the interface. Finally, Eq. (6) is the anisotropic form of the Gibbs–Thomson condition. The first term on the right hand side of Eq. (6) describes the capillary effect,

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