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Three-dimensional phase-field simulations of directional solidification

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Abstract

The phase-field method has become the method of choice for simulating microstructural pattern formation during solidification. One of its main advantages is that time-dependent three-dimensional simulations become feasible, which makes it possible to address long-standing questions of pattern stability and pattern selection. Here, a brief introduction to the phase-field model and its implementation is given, and its capabilities are illustrated by examples taken from the directional solidification of binary alloys. In particular, the morphological stability of hexagonal cellular arrays and of eutectic lamellar patterns is investigated.

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1. Introduction

The phase-field method has been used for numerical modelling of solidification microstructures for more than 20 years (for a recent review, see Ref. [1]). While in the beginning its predictive capabilities were limited, in recent years progress both in the formulation of the models and in their implementation has made it possible to obtain quantitative results in three dimensions that can be directly compared to experiments. Therefore, this method can now be used to gain new insights into problems of microstructure formation and morphological stability. The goal of the present paper is to provide a brief introduction to the phase-field method and its implementation in numerical simulations, and to illustrate its capabilities and limitations by a few examples taken from the directional solidification of binary alloys.

Problems of microstructure formation and morphological stability have been traditionally formulated as *free boundary problems*, in which the solid and liquid phases are separated by mathematically sharp boundaries which move in time and have often very complicated shapes (for reviews, see Refs. [2,3]). The implementation of this formulation in numerical simulations, however, is cumbersome since these boundaries have to be explicitly tracked. The phase-field method avoids this problem by introducing one or several supplementary scalar fields, the phase fields, which indicate the local state of the system for each space point. They take fixed values in the bulk and vary continuously through interfaces of a characteristic finite width W. The phase fields can be seen as order parameters, and the equations of motion, derived from Ginzburg–Landau type free energy functionals, are standard partial differential equations which are simple to solve; the shape of the interfaces is then implicitly given by a certain level curve of the phase field.

The price to pay for this simplicity is the introduction of the new scale W into the problem. The thickness of a typical rough solid–liquid interface is of the order of a nanometer, whereas typical microstructural patterns have features on the micron scale, and the entire sample is even much larger. Clearly, it is unfeasible to resolve numerically all these scales at the same time, even with modern supercomputers. In order to simulate microstructural patterns efficiently, the interface thickness has to be artificially enlarged in the model. In general, this introduces a dependence of the simulation results on the interface thickness W. A major progress has been the development of phase-field models for specific simple physical situations in which the dominant contribution to this dependence can

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be eliminated either by a suitable choice of parameters or by the addition of correction terms in the equations which cancel the undesired effects [4–9]. It has been demonstrated in detailed benchmark simulations that the simulation results are virtually independent of the interface thickness as long as it remains about an order of magnitude smaller than the smallest structural length scale. These models will be called "quantitative phase-field models" in the remainder of this paper. All the simulation results presented here are obtained with such models.

The results presented here concern microstructures formed during the directional solidification of dilute and eutectic binary alloys. The typical structures observed in dilute alloys are cells or dendrites, whereas in eutectics lamellar or rod-like two-phase composite patterns arise. A classic question is then which type of structure and which spacings can be observed for given conditions, and if there is a mechanism which leads to the selection of a particular structure or spacing [10]. This question has been extensively studied in thin samples, where patterns are quasi two-dimensional and convection in the liquid is suppressed. The results of both experimental and numerical investigations show that in general there exists, for given processing conditions, a range of stable spacings. The maximum and minimum stable spacings are set by the occurrence of dynamical instabilities that break one or several symmetry elements of the original patterns. If these spacings are plotted as the function of the control parameters, the so-called *stability balloon* of the periodic pattern is obtained. Different spacings inside this stable balloon can then be selected, either by the growth history or by boundary effects. These general features are common to many pattern-forming systems [11,12].

Whereas the situation is fairly well understood in thin samples, much remains to be learned about fully three-dimensional samples. The situation is complicated both by the effects of convection which is always present in bulk samples (except in microgravity) and by the fact that the structure and dynamics of three-dimensional patterns (two-dimensional fronts) are far richer than in two dimensions (one-dimensional fronts). Numerical simulations can be of great help to provide a useful starting point for the analysis: the situation can be first analyzed without convection and in a setting that is well controlled. As an example, it is shown here how the stability balloons for hexagonal cells in binary alloys and for lamellar eutectic patterns can be obtained. In particular, for cells the influence of crystalline anisotropy is studied and compared to the known results in two dimensions [13,14]. For eutectics, a zig-zag instability recently observed in experiments [15] is characterized, and it is shown that this is the only instability that is experimentally observable.

The remainder of the paper is organized as follows. In Section 2, the basic free boundary problem of directional solidification is outlined and the corresponding phase-field model is presented. Sections 3 and 4 are devoted to the stability of cellular and eutectic patterns, respectively, followed by conclusions and an outlook in Section 5.

2. Phase-field model

2.1. Free-boundary problem

The classic free-boundary problem and its phase-field representation have been discussed extensively in the literature [1–3]. The purpose of the present exposition is therefore not to give a detailed derivation or a review of different approaches to construct phase-field models; rather, its intention is to recall the main points with an emphasis on the ingredients needed in the context of directional solidification. A detailed presentation of the phase-field models used for the present work can be found in Refs. [4,7,9].

The directional solidification of a dilute binary alloy made of substances A and B is considered. For a complete modelling of the whole process, the thermal, concentration, and flow fields would have to be included. However, even though phase-field models including hydrodynamics [5] or thermal and solutal diffusion [8] have been developed, they remain to date computationally too costly to carry out systematic studies as intended here. Therefore, convection is neglected, and the "frozen-temperature approximation" is used, in which the temperature field in the sample is externally imposed,

$$T(z) = T_0 + G(z - V_{\rm p}t), \tag{1}$$

where the temperature gradient G is aligned with the z direction and the sample is pulled with a constant speed $V_{\rm p}$.

A dilute alloy is considered which has an idealized phase diagram consisting of straight liquidus and solidus lines of slopes m and m/k, respectively, where k is the partition coefficient. The concentrations $c_{\rm s}$ and $c_{\rm l}$ (in molar fractions) of impurities B at the solid and liquid sides of the interfaces satisfy the partition relation

$$c_{\rm s} = kc_{\rm l}. \tag{2}$$

For a sample with composition c_∞, the solidus temperature $T_0 = T_{\rm m} - |m|c_\infty/k$ (where $T_{\rm m}$ is the melting temperature of pure A) and the corresponding equilibrium liquid concentration $c_0 = c_\infty/k$ are chosen as reference temperature and composition, respectively.

With the temperature field fixed by Eq. (1), the dynamics of the solidification front is governed by the redistribution of solute. The classic free boundary problem is

$$\partial_t c = D_{\rm s,l}\nabla^2 c, \tag{3}$$

$$(c_{\rm l} - c_{\rm s})V_n = \hat{n}\cdot[D_{\rm s}\vec{\nabla}c|_{\rm s} - D_{\rm l}\vec{\nabla}c|_{\rm l}], \tag{4}$$

$$\frac{c_{\rm l} - c_0}{\Delta c} = -d_0\sum_{i=1}^{2}\left[a(\hat{n}) + \frac{\partial_2 a(\hat{n})}{\partial\theta_i^2}\right]\frac{1}{R_i} - \beta(\hat{n})V_n - \frac{z - V_{\rm p}t}{l_T}. \tag{5}$$