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Phase-field study on the effects of process and material parameters on the tilt angle during directional solidification of ternary eutectics

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ABSTRACT

In the directional solidification of molten alloys, the configuration of growing phases plays a vital role in the properties of the resulting materials. During directional eutectic solidification, the growth direction and the velocity can be controlled by an imprinted temperature gradient. For this kind of process, different instabilities can occur. One observed instability is tilted growth of the solid phases against the direction of the imprinted temperature gradient. In ternary eutectics this phenomenon can occur in systems with isotropic phases. To investigate the effects of the different process and material parameters, simulation studies with a thermodynamically consistent phase-field model based on the grand potential approach, are conducted. An idealized system with different isotropic interfacial energies, diffusion coefficients, lamellar spacings, solidification velocities and slopes of the imposed temperature gradient is systematically studied. The effects on tilting are analyzed and discussed. To quantitatively determine the tilt angle automatized, an unsupervised measurement method is developed. Based on the results of the measurements, a fitting function is derived to predict the tilt angles.

1. Introduction

The improvement of material properties is crucial for different technical applications [1]. The final microstructure influences the macroscopic material properties [2], like its mechanical, electrical, optical, acoustic, tribological, thermal and magnetic properties. Scientific investigations of the influence of the melt composition and of the process parameters on the evolving microstructure have improved the materials and the understanding of solidification in the last decades.

Besides experiments and analytic approaches, simulation methods have been established in recent years, to study the microstructure evolution during solidification. Especially the possibility to study the effect of different physical parameter and process parameters in complex spatial arrangements using the phase-field method allows to gain new insights into the solidification process [1,3]. During diffusion controlled directional solidification of a eutectic composition, different phases evolve in a coupled manner and form various arrangements in the microstructure [4]. In eutectic alloys different instabilities in the microstructure, such as bifurcation [5,6], growth of eutectic colonies [7,3,8,9], oscillations in the phase widths [5,10–13] and tilting in sections perpendicular to the growth direction [15,14,16–18], can be observed. In the case of tilting, the phases evolve with a deviation between the solid-solid interfaces and the direction of the imprinted temperature gradient. This leads to an undefined direction of the evolving phases and hence to different macroscopic material properties. Therefore, it is important to gain a better insight into the fundamental principles of tilted growth.

The two dimensional phenomenon of tilted growth for binary systems is investigated experimentally in [14,16], theoretically in [17] and numerically with phase-field simulations in [16,18–20]. It is found that tilted growth in binary eutectics is a result of anisotropic interface energies. Further, it is observed that the tilt angle does increase when the lamellar spacing is increased [9,13,14,21,22]. However Ghosh et al. do not confirm this behavior in [18,19] with their phase-field simulations and dynamic boundary integral studies. For these investigations, slow growth velocities and small Péclet numbers are used in the vicinity of the lamellar spacing with the minimum undercooling. In [9], Akamatsu et al. illustrate the dependence of the tilt angle on the lamellar spacing with a second order polynomial fit.

In multicomponent multiphase systems further effects besides anisotropy can cause tilted growth. For phase-field simulations of ternary eutectics, tilting is reported for crystallographic isotropic

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systems [23,24]. Apel et al. [23] describe the behavior of tilting for an asymmetric ternary phase diagram with a three phase sequence of $\alpha\beta\delta$. In contrast to the observed modes in binary alloys, tilted growth is a result of the lack of mirror symmetries in the diffusion field of the ternary system. Tilted growth due to different but isotropic interface energies is investigated for various parameters by Hötzer et al. [24]. By varying the ratio of the interface energies between the solid phases α and β and the interface energies of the solid-liquid phase boundaries ($\alpha\ell$), tilting can be observed.

Further, the phase equilibria can influence the tilt angle experimentally and in phase-field simulations, as shown for the ternary system $CBr_4 - C_2Cl_6$ [22]. For this system, tilting occurs as a response of different growth velocities [25], demonstrating the influence of the process conditions on the tilt angle.

To improve the understanding of the mechanisms that influence the tilt angle during the directional solidification of ternary eutectics, different physical parameters and process parameters are systematically varied in this work. For this purpose, extensive studies of 2D phase-field simulations are conducted, and the resulting tilt angles are measured with an unsupervised and automatized method. First, the applied model and simulation setup is introduced, and a quantitative method is given to measure the tilt angle. The effects of different lamellar spacings, diffusion coefficients, interface energy ratios, temperature gradients and growth velocities on the resulting tilt angle are analyzed. Based on these results, a general fitting approach is then derived to predict the tilt angle depending on the investigated parameters.

2. Methods

In this section the applied phase-field model is introduced, the setup for the simulations is illustrated and the variation ranges of the analyzed parameters are explained. Further, the method for measuring the resulting tilt angles in the simulations is described.

2.1. Phase-field model

To investigate tilted growth, the phase-field model based on the thermodynamic grand potential approach is derived in [26,27] and is presented in [28]. The model is spatially discretized with a finite difference scheme and the temporal evolution is calculated by an explicit Euler scheme, as described e.g. in [29]. The implementation of the model into the massive parallel framework waLBerla[2] [30] and the used optimizations are presented in [28,31,32]. Quantitative investigations of different aspects of directional solidification of ternary alloys with the applied model are shown in [24,28,33–38].

For the simulation of directional solidification, the coupled set of partial differential equations of the form

$$\frac{\partial T}{\partial t} = \frac{\partial}{\partial t}(T_0 + G(z - vt)) = -Gv \tag{3}$$

are solved. The evolution Eqs. (1) for the $N = 4$ phase-fields $\phi_{\hat{\alpha}}$ are derived with an Allen-Cahn type approach from the grand potential functional. The Lagrange multiplier Λ is utilized to fulfill the constraint $\sum_{\hat{\alpha}=1}^{N} \partial\phi_{\hat{\alpha}}/\partial t = 0$ and $\hat{\alpha}, \hat{\beta}$ and $\hat{\gamma}$ represent phase indices. To calculate the driving force for the phase transition, the grand potentials $\psi_{\hat{\alpha}}$ of the system are determined. While the kinetics of the diffuse interface is described by the parameter τ, its thickness is related to the parameter ϵ. The diffuse interface is modeled with the gradient energy density a, given as

$$a(\boldsymbol{\phi}, \nabla\boldsymbol{\phi}) = \sum_{\hat{\alpha}<\hat{\beta}}^{N,N} \gamma_{\hat{\alpha}\hat{\beta}} |q_{\hat{\alpha}\hat{\beta}}|^2 \tag{4}$$

and the potential energy ω is expressed as an obstacle-type potential energy by

$$\omega(\boldsymbol{\phi}) = \frac{16}{\pi^2} \sum_{\hat{\alpha}<\hat{\beta}}^{N,N} \gamma_{\hat{\alpha}\hat{\beta}} \phi_{\hat{\alpha}} \phi_{\hat{\beta}} + \sum_{\hat{\alpha}<\hat{\beta}<\hat{\delta}}^{N,N,N} \gamma_{\hat{\alpha}\hat{\beta}\hat{\delta}} \phi_{\hat{\alpha}} \phi_{\hat{\beta}} \phi_{\hat{\delta}}. \tag{5}$$

Both a and ω depend on the isotropic interface energies $\gamma_{\hat{\alpha}\hat{\beta}}$. The parameter $\gamma_{\hat{\alpha}\hat{\beta}\hat{\delta}}$ is introduced to suppress the occurrence of third or higher order phases in binary interfaces [39,29].

Based on the assumption of mass conservation and on Fick's laws, the evolution Eqs. (2) for the $K = 3$ chemical potentials $\boldsymbol{\mu}$ are derived. The interpolation function $h_{\hat{\alpha}}(\boldsymbol{\phi})$ is used to scale the values in the diffuse interface.

To balance the effects of the artificially enlarged interface, an anti-trapping current, $\boldsymbol{J}_{at}$, is introduced [27,40,41]. The mobilities of the chemical potentials $\boldsymbol{\mu}$ are given by the matrix $\boldsymbol{M}$, which is described as

$$\boldsymbol{M}(\boldsymbol{\phi}, \boldsymbol{\mu}, T) = \sum_{\hat{\alpha}=1}^{N} \boldsymbol{D}_{\hat{\alpha}} \frac{\partial \boldsymbol{c}_{\hat{\alpha}}(\boldsymbol{\mu}, T)}{\partial \boldsymbol{\mu}} h_{\hat{\alpha}}(\boldsymbol{\phi}) \tag{6}$$

with the diffusion matrix $\boldsymbol{D}_{\hat{\alpha}}$. The concentration vector $\boldsymbol{c}_{\hat{\alpha}}(\boldsymbol{\mu}, T)$ contains the values of the K chemical elements in the corresponding phase $\hat{\alpha}$. As presented in [42], the derivative $\partial \boldsymbol{c}_{\hat{\alpha}}/\partial\boldsymbol{\mu}$ is calculated from the parabolic Gibbs energies.

The evolution of the analytic temperature field, depending on the base temperature T_0 and on the applied gradient G, with the velocity v in the growth direction z, is described by Eq. (3). The approximation of the temperature by means of an analytical approach relies on the assumption that heat diffusion is multiple times faster than mass diffusion, resulting in a significant reduction of the computational effort.

$$\tau\epsilon \frac{\partial \phi_{\hat{\alpha}}}{\partial t} = \underbrace{\underbrace{-\epsilon T \left(\frac{\partial a(\boldsymbol{\phi}, \nabla\boldsymbol{\phi})}{\partial \phi_{\hat{\alpha}}} - \nabla \cdot \frac{\partial a(\boldsymbol{\phi}, \nabla\boldsymbol{\phi})}{\partial \nabla \phi_{\hat{\alpha}}} \right) - \frac{T}{\epsilon} \frac{\partial \omega(\boldsymbol{\phi})}{\partial \phi_{\hat{\alpha}}}}_{\text{interface contribution}} - \underbrace{\frac{\partial \psi(\boldsymbol{\phi}, \boldsymbol{\mu}, T)}{\partial \phi_{\hat{\alpha}}}}_{\text{bulk contribution}}}_{:= rhs_{\hat{\alpha}}} + \underbrace{\frac{1}{N} \sum_{\hat{\beta}=1}^{N} rhs_{\hat{\beta}}}_{\Lambda}, \tag{1}$$

$$\frac{\partial \boldsymbol{\mu}}{\partial t} = \left[\sum_{\hat{\alpha}=1}^{N} h_{\hat{\alpha}}(\boldsymbol{\phi}) \left(\frac{\partial \boldsymbol{c}_{\hat{\alpha}}(\boldsymbol{\mu}, T)}{\partial \boldsymbol{\mu}} \right) \right]^{-1} \cdot \left(\nabla \cdot \left(\boldsymbol{M}(\boldsymbol{\phi}, \boldsymbol{\mu}, T) \nabla \boldsymbol{\mu} - \boldsymbol{J}_{at}(\boldsymbol{\phi}, \boldsymbol{\mu}, T) - \sum_{\hat{\alpha}=1}^{N} \boldsymbol{c}_{\hat{\alpha}}(\boldsymbol{\mu}, T) \frac{\partial h_{\hat{\alpha}}(\boldsymbol{\phi})}{\partial t} - \sum_{\hat{\alpha}=1}^{N} h_{\hat{\alpha}}(\boldsymbol{\phi}) \left(\frac{\partial \boldsymbol{c}_{\hat{\alpha}}(\boldsymbol{\mu}, T)}{\partial T} \right) \frac{\partial T}{\partial t} \right) \right), \tag{2}$$

[2] www.walberla.net.