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Directional solidification of inclined structures in thin samples

Jihene Ghmadh [a], Jean-Marc Debierre [a,*], Julien Deschamps [b], Marc Georgelin [b], Rahma Guérin [a], Alain Pocheau [b]

[a] Aix-Marseille Université, CNRS, Université de Toulon, IM2NP UMR 7334, 13397 Marseille, France
[b] Aix-Marseille Université, CNRS, Centrale Marseille, IRPHE UMR 7342, 13384 Marseille, France

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Abstract

We address the directional solidification of inclined structures by combining numerical and experimental studies performed in situations capable of yielding a detailed relevant comparison between them. We especially seek to determine the growth directions and the stability of microstructures at various Péclet numbers when the crystal axes and the thermal gradient involve a misorientation. For this we perform experiments and simulations in the closest possible conditions referring to similar physical parameters and to a monocrystal growing in a thin sample by a single layer of homogeneously spaced microstructures. Implementing a 3D phase-field numerical code proves necessary to accurately model the solidification structures. A quite satisfactory agreement, both on qualitative and quantitative grounds, is found between experiments and 3D simulations, on both the growth directions of microstructures and the transition to the degenerate mode. This agreement provides a confirmation of the growth direction law evidenced experimentally and a fine validation of the 3D phase-field numerical model.

Keywords: Solidification microstructures; Crystallographic misorientations; Phase-field modeling; Dendritic solidification

1. Introduction

As compared to idealized descriptions of directional solidification, many extra features may influence and even sometimes dominate real processes [1]. For instance, casting is largely conditioned by misorientations between the direction of the local heat gradient and the crystal axes of the growth structures. This is definitely the dominant effect in equiaxed growth, when the rate of nucleation is large, so that many structures solidify simultaneously with quite different crystal orientations. Important effects result that concern both the structure shapes and their dynamics, as asymmetric branching, structure stability, grain drift, grain texture evolution, etc.

Directional solidification experiments in thin samples and transparent dilute alloys may provide a simplified situation where all the structures grow from a single crystal and experience a uniform temperature gradient. Growth then occurs with planar isotherms and the same misorientation angle θ_0 on the whole sample. An experiment in CBr_4 showed that, at $\theta_0 = 30^\circ$ and 40°, the growth angle θ depends on the spacing λ and the velocity V through a combination corresponding to the Péclet number $\mathrm{Pe} = \lambda V/D$ [2]. Another experiment in succinonitrile studied the response at many different θ_0 [3,4]. Noticing a symmetry in the database led to the identification of the $\theta(\mathrm{Pe}, \theta_0)$ law which will be referred to as the DGP law in the following. An analytical model based on solute interaction in given tip geometries

* Corresponding author. Tel.: +33 491288735.
E-mail addresses: jihene.ghmadh@im2np.fr (J. Ghmadh), jean-marc.debierre@im2np.fr (J.-M. Debierre), deschamps@irphe.univ-mrs.fr (J. Deschamps), marc.georgelin@irphe.univ-mrs.fr (M. Georgelin), rahma.guerin@im2np.fr (R. Guérin), alain.pocheau@irphe.univ-mrs.fr (A. Pocheau).

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proposed relationships for the growth angle which are in agreement with experiments and simulations [5].

To date, the numerical methods used to study misorientation effects on solidification were systematically two-dimensional (2D) [2,6–8]. Those based on a boundary element method showed that the growth angle is always smaller than θ_0 and that it increases less than linearly with either θ_0 or the velocity V [6]. Those relying on a sharp interface code showed for a given misorientation $\theta_0 = 30°$ that the growth angle varies with the Péclet number alone [2]. Recent phase-field simulations developed to study competing grain orientations reproduced qualitatively the DGP law for $\theta_0 = 30°$ [7]. Even more recently, comparable results were found for a wide range of misorientations [8]. Besides being limited to 2D systems, these numerical studies addressed too few Pe or θ_0 values (with the exception of Ref. [8]) to enable the determination of the $\theta(\mathrm{Pe}, \theta_0)$ law and its comparison to experiments.

On the other hand, recent simulations of solidification without misorientation showed marked differences between 2D and 3D structures [9], and it is likely that comparable differences also exist for inclined structures. However, up to now, numerical results in 3D including misorientation were only reported for confined growth of a pure melt [10]. The goal of the present work is thus twofold: first develop a 3D phase-field model of directional solidification capable of addressing large misorientation angles, and then use it to perform a direct comparison with experiment.

To enhance the close relationship between the model and the experiment, the construction of our numerical model has constantly made reference to the experimental situation. The benefit of this approach has been to finely validate the model with respect to the experiment and thus give confidence in its extensions to more complex systems in the future. To reach a relevant comparison, our strategy has been to define a well-controlled experimental setup and a model that can be used for the exact same geometry and physical parameters. Then, a range of control parameters has been scanned and attention has been focused on the main evolutions concerning the structure shapes, their orientation and their stability. This program has been fulfilled here for directional solidification in thin samples of a succinonitrile-based dilute alloy and 3D phase-field simulations. A quantitative agreement has been found for the orientation response and the stability limits, while the shapes and their evolution displayed a qualitative agreement. In addition, new results have been obtained, showing that the orientation response is rather insensitive to a number of parameters.

2. Experimental

The setup is designed to achieve directional solidification of a layer of microstructures in homogeneous and controlled conditions capable of allowing both the study of misorientation and the visualization of microstructures [3,4]. For this a thin sample is pushed at a controlled speed in between heaters and coolers that set a uniform thermal gradient (Fig. 1a). The sample thickness is chosen so as to allow the growth of a single layer of microstructures and observation is achieved by ombroscopy. Control of misorientation is obtained by selecting a single crystal from the whole sample and by tuning the direction of the thermal gradient.

Heaters and coolers are made of top and bottom metallic blocks which sandwich the sample. They are electronically regulated at 100 °C and 10 °C, respectively, to an accuracy better than 0.1°. The gap between them, set by spacers, yields a thermal gradient of 140 K cm^{-1} on the solidification interface. The pushing stage is provided by a microstepper motor which drives a linear ball-screw attached to a translating stage. Pushing velocities up to 50 μm s^{-1} may be achieved with a relative accuracy better than ±3% for long durations.

Samples are made of two glass plates separated by 50 μm thick spacers and filled with the mixture being solidified. We use succinonitrile with acrylonitrile as solute. Samples are wide (4.5 cm) and long (15 cm) enough to provide solidification domains far from boundary disturbances. Their thickness was chosen to be small enough to avoid the emergence of a second layer and large enough to ensure a 3D behavior of microstructures as opposed to the 2D ribbon-like behavior displayed for excessive squeezing [9]. The sample transparency allows visualization of the solidification interface by following the slight optical aberrations undergone by a parallel light beam that crosses it. Images are recorded by a camera with a resolutions of 768 × 512 pixels. This usually corresponds to a width of 2 mm on the solidification interface.

To improve the microstructure homogeneity and allow a definite misorientation to be studied, care was taken to prepare the sample in a single-crystal state. This has been achieved by selecting a grain and by making it invade the whole sample by a spatial control of fusion/solidification. Analysis of grain orientation has been performed from the morphology of both freely growing germs and rapidly growing dendrites. The former ones developed a cross shape with a 4-fold symmetry and the latter ones displayed side-branches which, apart from those directed on the sample plane, were aligned on the sample normal. These features indicate that the [0 1 0] crystal axis is normal to the sample plane, and the two remaining principal axes $\vec{a}$ and $\vec{a}'$ are lying in this plane (Fig. 1b). Misorientation of the solidification configuration is then given by their angles with the thermal gradient $\vec{G}$. The misorientation angle $\theta_0 = (\vec{a}, \vec{G})$ represents the smallest of them (Fig. 1a), the direction $\vec{a}$ then being the one actually followed by rapidly growing dendrites. This direction usually corresponds to a crystallographic axis although atypical directions may sometimes occur [11,12]. Depending on the pushing velocity, it is observed that microstructures grow in a direction that lies in between the thermal gradient direction $\vec{G}$ and the principal axis direction $\vec{a}$. We call $\vec{V}_g$ their growth