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Phase-field simulations of spiral growth during directional ternary eutectic solidification

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A B S T R A C T

A wide variety of growth patterns has been observed during the directional solidification of three-phase ternary eutectics in multi-component alloys. One hypothesized pattern is the spiral growth of two rod-like phases rotating around each other, which are embedded in a matrix phase. Possible evidence of spiral growth is found experimentally in longitudinal micrographs of the ternary eutectic system Ag–Al–Cu. The phase-field method allows the study of such spatially complex microstructures in order to gain deeper insights into the three-dimensional pattern formation of ternary eutectics. Based on the simulation parameters, which produced a high tilt angle in 2D lamellar growth, determined via systematic parameter studies, multiple spirals are detected in large-scale 3D simulations. Through the presented phase-field simulations, the possibility of the existence of spiral growth in ternary eutectics, reported from experiments, is confirmed.

1. Introduction

In nature, spirals of different variations form on various length scales ranging e.g., from spiral galaxies to DNA molecules. In math, two major definitions for spirals exist. First, spiral curves in a plane, like archimedian or equiangular spirals, revolve around a center point, to which they monotonically change their distance, as it is the case for spiral galaxies. Second, three-dimensional spirals, also called helices, revolve around a rotation axis and continuously change their position in the direction parallel to this axis. Spiral curves can furthermore share the same rotation axis, like the well-known double helices found in DNA molecules. In this work, we focus on spiral growth during directional solidification of ternary eutectic alloys. The growing phases form a helix shape with a constant distance from the rotation center.

In binary alloy systems, invariant phase reactions occur when one phase transforms into two different phases, or vice versa. In alloy systems with three or more components, the additional degrees of freedom allow for invariant reactions between four or more phases. The phase diagram for a ternary alloy may contain a three-phase eutectic invariant point, where three different solid phases solidify simultaneously from a specific liquid composition upon cooling. For such multicomponent alloys, Hecht et al. summarized the current research in a review paper [1]. The microstructural patterns that can form during such three-phase eutectic solidification are far more complex and varied than the lamellar or rod-type morphologies that generally form in binary eutectics. The exact kind of pattern depends on the volume fractions, the diffusion coupling in the liquid between each of the phases and the three solid/liquid interfacial energies, which themselves depend on crystallographic orientation [2].

One of the first studies to observe pattern formation in ternary eutectics parallel to the growth direction is conducted by Cooksey and Hellawell in 1967 [3]. By analyzing cross-sections parallel to the solidification front, they report a large variety of microstructural

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features in the seven material systems Cd–Sn–(Pb, In, Tl), Al–Cu–(Mg, Zn, Ag) and Zn–Sn–Pb. Ruggiero and Rutter [4] and Lewis et al. [2] predict five idealized patterns based on geometrical considerations. The patterns consist of different arrangements of rod and lamellar phases, or in one case, two rod-like phases grow in a continuous matrix phase. They observe many of their predicted patterns in cross-sections of the systems Bi–In–Sn and Ag–Cu–Sn. However, the classification in Lewis et al. [2] only covers 2D patterns in cross-sections parallel to the front, and does not consider the evolution of pattern elements over a distance or in 3D.

McCartney et al. [5] use Ag–Al–Cu as model system for studying ternary eutectic growth. Detailed studies of the morphologies of Ag–Al–Cu are discussed by Genau and Ratke [6] as well as Dennstedt et al. [7–9] who analyze images, parallel as well as perpendicular to the growth direction. The micrographs in Fig. 1 show an example of the microstructure observed in these samples, viewed in the longitudinal (Fig. 1(a)) and transverse (Fig. 1(b)) directions. This sample relates to a velocity of 0.5 μm/s and a thermal gradient of 3 K/mm. The ternary eutectic composition is located at Al-18.1Ag-12.8Cu mol-% as determined by the Calphad database from [10,11]. The two intermetallic phases, Ag_2Al and Al_2Cu always show a close association, often forming aligned ribbons composed of alternating rod-like phases, separated by lamellar regions of a solid-solution aluminum phase, as a so-called “brick-like” structure [12]. In some regions of the sample, such as the one highlighted here, the longitudinal cross-section Fig. 1(a) indicates the two intermetallic phases spiraling around each other during their growth. Genau and Ratke report this possible evidence of spiraling eutectic rods as helices in a continuous or semi-continuous matrix phase in [6] with a figure similar to Fig. 1(a). From pure 2D images, it is impossible to conclude that the pattern is the result of spiraling. However, the three-dimensional analysis necessary to conclusively identify the microstructure is experimentally difficult and currently not available. Therefore, computational effort based on 3D phase-field simulations is investigated to determine whether such a spiral effect occurs in a three-phase eutectic system.

Spiral growth of binary eutectic structures is firstly reported in 1968 by Hellawell et al. [13] describing helices in directionally solidified binary Al–Al_2Cu and Al–Ag_2Al, as well as Al–Zn and LiF–NaF. In this case, spiral growth referred to rotation of entire grains of lamellar plates about an axis parallel to the growth direction. Other researchers discover similar effects in Pb–Sn [14–16], with spiral behavior occurring only in grains with particular crystallographic orientation relationships. This type of spiraling is attributed to certain orientation relationships between the coupled phases, forcing a misalignment between the preferred crystallographic growth direction and the growth axis, resulting in a growth component parallel to the solid–liquid interface [16].

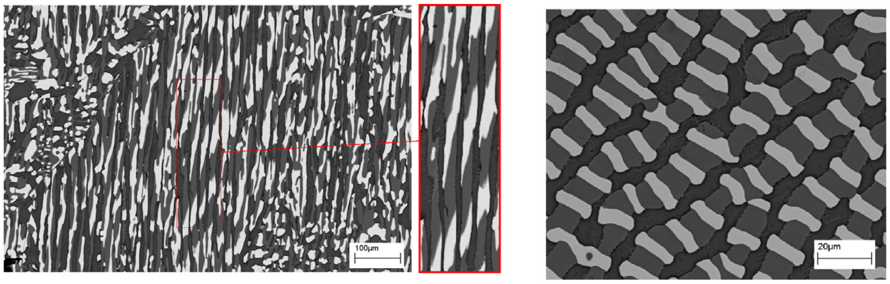
A very different type of two-phase spiral growth is reported for the first time in 2010 by Akamatsu et al. [17] in the transparent system SCN-DC-NA, a pseudo-ternary eutectic alloy based on succinonitrile-(d)camphor. During directional solidification of an univariant eutectic composition, the two solidifying phases are found to form parabolic-shaped dendrites, with the two phases spiraling around each other at the growth tip, producing a double helix pattern. Pusztai et al. [18] and Rátkai et al. [19] modeled this phenomenon using phase-field simulations, and conclude that anisotropy is a vital component to forming this type of structure.

In order to determine whether spiral growth of the type suggested by Fig. 1 can occur during directional solidification of ternary eutectic systems, phase-field simulations are utilized. Over the past years, simulations have become a powerful tool to study such spatially complex three-dimensional phenomena. Through advances in various scientific disciplines, the models became more accurate but also more computationally intensive to solve. Due to the continuing exponential increase in computational power [20], it became possible to simulate large domains with representative volume elements in a reasonable amount of time. These simulations provide the possibility of in-situ studies of the microstructure evolution, compared to micrographs which only display two-dimensional sections of the final state after completion of the solidification process.

The phase-field method has been established in a broad range of free boundary problems such as solidification processes [21–30]. In this work, we use a thermodynamically consistent phase-field model based on the grand potential approach [30–32] to study spiral growth during directional solidification of ternary eutectics. This model was already applied to eutectic solidification of idealized binary [33] and ternary systems [34–36]. Large 3D simulations for the ternary system Ag–Al–Cu are recently published by the authors in [30,37,38].

We hypothesized that spiral growth in 3D ternary eutectic structures is mechanistically related to lamellar tilted growth in a 2D configuration. Tilt in directional solidification is defined as growth with a constant angle of the solid–solid interfaces against the temperature gradient direction.

Tilted growth of lamellar eutectics is first investigated by Faivre and coworkers in 1989 [39]. The physical mechanism of tilting is further studied using a variety of experimental techniques, theoretical approaches and simulations. Several different conditions can result in growth of a 2D eutectic structure where the lamellae maintain a specific misorientation angle with respect to the growth



(a) Longitudinal cross-section showing a pattern indicative of spiral growth in the highlighted part of the micrograph.

(b) Cross-section parallel to the solidification front, showing a chain-like structure.

Fig. 1. Two micrographs of a directionally solidified ternary eutectic Ag–Al–Cu alloy. Both images are from the same sample, perpendicular to each other. The supposed spiraling phases are Ag_2Al (white) and Al_2Cu (gray) in an aluminum matrix.