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Eutectic colony formation in systems with interfacial energy anisotropy: A phase field study



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ABSTRACT

Instability of a binary eutectic solidification front to morphological perturbations due to rejection of a ternary impurity leads to the formation of eutectic colonies. Whereas, the instability dynamics and the resultant microstructural features are reasonably well understood for isotropic systems, several experimental observations point to the existence of colonies in systems with anisotropic interfaces. In this study, we extend the understanding of eutectic colonies to anisotropic systems, where certain orientations of the solid-liquid or solid-solid interfaces are associated with a lower free energy than the others. Through phase field simulations in 2D and 3D, we have systematically probed the colony formation dynamics and the resulting microstructures, as functions of the pulling velocity and the relative orientation of the equilibrium interfaces with that of the imposed temperature gradient. We find that in 2D, stabler finger spacings are selected with an increase in the magnitude of anisotropy introduced, either in the solid-liquid or in the solid-solid interface. The fingers have a well-defined orientation for the case of anisotropy in the solid-liquid interface, with no fixed orientations for the lamellae constituting the colony. For the case where anisotropy exists in the solid-solid interface, the lamellae tend to orient themselves along the direction of the imposed temperature gradient, with tilted solid-liquid interfaces from the horizontal. The 3D simulations reveal existence of eutectic spirals which might become tilted under certain orientations of the equilibrium interfaces. Our simulations are able to explain several key features observed in our experimental studies of solidification in Ni-Al-Zr alloy.

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1. Introduction

Two-phase growth in a ternary alloy where the two phases exchange two components and reject an impurity into the liquid, results in the formation of a boundary layer of this component ahead of the solidification front. This interface is then unstable to morphological perturbations much in the same manner as in a Mullins-Sekerka instability [1] (henceforth will be referred to as MS instability) of the solidification front during single phase growth. The amplification of these instabilities leads to the formation of eutectic colonies (also called two-phase fingers) which are cells made up of two-phase lamellae.

Experimentally, eutectic colonies have been extensively studied, as in [2–8], including a study on their dynamics during directional solidification of thin samples [9]. Furthermore, Akamatsu et al. [10] are the first to observe and characterize the helical arrangement of two eutectic solids about a finger axis, which they anoint as "eutectic spirals". They also point out the presence of such structures in studies that predate their observation, like the one in Al-Cr-Nb systems carried out by Souza et al. [11].

Theoretical understanding of this problem begin with the study by Plapp and Karma [12], where they perform linear stability analysis to establish that the instability leading to colony formation is oscillatory compared to the one operating on a single phase binary solid-liquid interface. The experimental observation that a spiral tip radius (ρ) scales linearly with the lamellar width (λ) [10] leads to an analytical establishment of the scaling of ρ with $V^{-0.5}$, where V is the spiraling dendrite tip growth velocity [13].

Numerical computations performed to study eutectic colony formation dynamics augment theoretical understanding in regimes outside the purview of linear analysis. In this regard, phase field simulations of eutectic colonies by Plapp and Karma [14] not only validate their theory in [12], but also highlight the lack of a stable cellular morphology under isotropic conditions.

While the studies in the previous cases concentrate on isotropic eutectics, alloys systems in general contain phases which either have anisotropic solid-liquid interfaces or where the interfacial



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boundaries between the solid phases have a preferred alignment of crystallographic planes giving rise to defined orientation relationships for these interfaces. Experimentally, anisotropic interfacial energies of the solid-solid interfaces have been found to result in spirals in binary eutectic alloys [15,16].

Ni-Al-Zr is another exemplary system consisting of two solid intermetallic phases (Ni₃Al and Ni₇Zr₂) whose crystallographic planes share a well defined orientation relationship as revealed by the TEM diffraction patterns in Fig. 1(a) and (b). The two-phase eutectic in this alloy is also a monovariant reaction and is therefore unstable to morphological perturbations. Detailed characterization of the colonies shows that the central stem of the colonies have well aligned lamellar feature as seen in Fig. 1(c). Further resolution of the colony microstructures at the interface between two colonies reveals features resembling spiraling of two solid phases.

The influence of a well defined orientation relationship between the eutectic solids in a binary system on its steady-state growth morphologies has been studied theoretically, numerically and experimentally in [17–19]. Pusztai et al. [20] and Rátkai et al. [21], investigate the influence of kinetic anisotropy in stabilizing the spiral microstructures during two-phase eutectic colony formation in a ternary alloy by conducting phase field simulations. However, no studies exist which systematically investigate the influence of anisotropy in the interfacial energies on the colony morphology arising out of the destabilization of steady-state two-phase growth interface, in either thin-film geometry or during bulk solidification.

This motivates the two principal aims in our paper. Firstly, we perform phase field simulations to investigate the influence of the anisotropy of the different interphase interfaces on eutectic colony morphologies in thin-film geometry. Secondly, through phase-field simulations in three-dimensions we characterize the influence of anisotropy both in the solid-solid and solid-liquid interfaces on the morphologies of the spirals and the colony structures.

In what follows, we perform phase-field simulations of the following directionally solidifying systems: one where the interfacial energy is isotropic, followed by systems with anisotropic solidliquid and solid-solid interfacial energies in 2D. Our simulations in this regard can be thought to be representative of the solidification experiments carried out for thin samples [9]. The colony formation dynamics and the resultant lamellar morphologies for each of these situations are studied as functions of the anisotropy strength and the sample pulling velocity.

We also perform 3D simulations in order to understand the effect of an introduction of a third dimension on the lamellar structures in directionally solidified systems with anisotropic solid-liquid and solid-solid interfacial energies. Our simulations are numerical studies of the eutectic spiraling observed experimentally in [10]. The computational cost involved in these simulations restricts us to a single choice in both the pulling velocity and the strength of anisotropy.

We begin with a discussion of 2D isotropic systems.

2. 2D: isotropic system

We begin our discussion with the isotropic system where we briefly review the mathematical model developed by Plapp and Karma [14]. The colony formation dynamics and the resulting lamellar and cellular morphologies are also discussed here.





Fig. 1. The anisotropy in solid-solid interfacial energy is indicated by the existence of a well defined orientation relationship between the two phases in (a) and (b). The orientation of the lamellae along the axis of the colony finger along with some spiral like features is displayed in (c). The two eutectic phases are identified from their contrast in (d).

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