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Phase-field modeling of precipitate evolution dynamics in elastically inhomogeneous low-symmetry systems: Application to hydride precipitation in Zr

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Abstract

A phase-field model was developed within the framework of heterogeneous elasticity theory to study the precipitation of particles with trigonal symmetry in a hexagonal matrix. The model is first calibrated and successfully compared with previous analytical calculations performed to explain the effect of symmetry-breaking transformations on precipitate morphology. Secondly, the model was adapted to study the precipitation of the coherent ζ hydride phase in zirconium. The results are consistent with the well-established experimental observation of the existence of acicular precipitates aligned along the dense directions in the basal plane. Moreover, original kinetic pathways are implied by the presence of a threefold axis of symmetry, leading to the emergence of original morphological bifurcations not previously reported and probably related to the inconsistency between the threefold symmetry and the inversion properties of the B function introduced by Khachaturyan. In spite of its simplicity (only one order parameter is taken into account), the present phase-field model gives rise to very complex morphological sequences.

Keywords: Precipitation; Phase-field models; Hydrides

1. Introduction

The equilibrium particle shape during the early stages of precipitation is the result of competing effects of the interfacial and elastic energies giving rise to a high diversity of morphologies. Moreover, the kinetic path followed by the precipitates can go through different transient shapes before reaching the equilibrium shape. For example, a wide range of γ' morphologies (doublets, octets or chaplets of cuboids) are reported in the literature concerning nickel-based superalloys, some of them clearly induced by kinetic effects, as shown by numerical investigations using finite-element [1] or phase-field [2] calculations.

Equilibrium shapes can be investigated using the micro-elasticity theory developed by Khachaturyan [3], which easily incorporates strain-induced effects during the nucleation, growth and coarsening. Most particularly, this theory establishes a very useful criterion to determine the habit plane of well-developed coherent precipitates assuming that the interfacial energy plays a negligible part. According to this theory, the normal to equilibrium habit plane $\mathbf{n_0}$ is the direction that minimizes the following function B:

$$B(\mathbf{n}) = \sigma^0_{ij}\varepsilon^0_{ij} - n_i\sigma^0_{ij}\Omega_{jm}(\mathbf{n})\sigma^0_{mn}n_n \quad (1)$$

This function includes all the elastic parameters of the system: the stress-free strain tensor ε^0_{kl}, which is associated with the stress-free volume and shape change of the matrix due to the transformation, $\sigma^0_{ij} = C_{ijkl}\varepsilon^0_{kl}$ with the elastic constants of the system assumed to be elastically homogeneous (the matrix and precipitate have the same elastic constants), and the inverse Green function tensor $\Omega^{-1}_{jm}(\mathbf{n}) = C_{jklm}n_kn_l$ where n_i are the Cartesian components of $\mathbf{n}$. This

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criterion remains valid even in the case of inhomogeneous elasticity conditions (distinct elastic constants between the matrix and the precipitate), by considering only the elastic constants of the precipitate. Note that all the crystallographic symmetry information is included in function *B*.

Despite the huge impact of this criterion, it does not incorporate the interfacial effect which may induce morphological bifurcations. Different approaches, classified in Ref. [4], have therefore been proposed to study the morphological evolution of a misfitting particle inside a matrix under the combined elastic and interfacial effects. The first category consists in restricting the possible precipitate shapes to a particular geometrical family (for example, ellipses in two dimensions and ellipsoids in three dimensions). It offers the advantage of giving analytical solutions to morphological bifurcation problems, precisely determining the influence of the various material parameters [5,6]. However, some possible shapes may be ignored by this method, which is its main drawback. Other numerical approaches (called nonrestricted methods, or NRM) allow no restriction on transient or equilibrium particle morphologies [7–16]. Moreover, they can simulate shape evolution dynamics during growth and coarsening. On the latter aspects, inhomogeneous elastic constants in the system may have a significant effect on shape instabilities. For example, the application of the discrete atom method [10] to an initially soft circular isotropic inclusion in an isotropic matrix ($\mu^p = \mu^m/2$, with μ^p and μ^m the shear moduli of the precipitate and matrix, respectively) reveals that it is unstable and develops topological waves at its surface before reaching its equilibrium elliptical shape. The corresponding simulation was achieved in Ref. [9] with a phase-field model (PFM) taking into account elastic inhomogeneities. This feature was incorporated via the algorithm presented in Ref. [17] and based on the perturbation method of Ref. [18]. Both methods, although developed at two distinct scales, give the same evolution of the precipitate shape, meaning that the shape destabilization mechanism is not model dependent but has a true physical origin. Moreover, despite the simple isotropy assumption of the elastic constants in both matrix and precipitate, these studies show that elastic inhomogeneities can induce significant shape instabilities.

A thorough survey of the literature shows that there is a lack of information on systems with lower symmetry than cubic. This paper proposes an extension of the study of shape instabilities, leading to the formation of transient or equilibrium shapes, to the case of hexagonal systems in inhomogeneous elasticity conditions. Furthermore, to illustrate the possible impacts of the study on technological alloys, an example of great industrial interest is chosen as the reference case: the precipitation of ζ hydrides in an αZr matrix [6,19,20], for which a crystallographic symmetry break occurs, since ζ hydrides belong to the trigonal point group *3m* whereas the αZr matrix belongs to the hexagonal point group *6/mmm*. The formation of three different orientation variants is experimentally observed in the basal plane during precipitation of the trigonal precipitates inside the hexagonal matrix. In order to determine whether such variants can be elastically induced, Eshelby's equivalent inclusion method was adopted in a previous study [6], allowing us to conclude, under the heterogeneous elasticity assumption, that the most stable shape was elliptic in the basal plane, with the preferential orientation of the trigonal precipitates along the dense directions, this orientation being dictated by the extra elastic constant C_{15} of the precipitate.

Although useful, this approach is based on the assumption that the precipitate is ellipsoidal, though no argument can ascertain whether it is a good approximation of the exact equilibrium shape. Moreover, this method does not allow an exhaustive approach of all the possible transient or equilibrium morphologies that can be encountered in this type of system. Indeed, by analogy with homogeneous cubic systems with purely dilatational misfit, it is expected that two kinds of shape transition will be observed in the system (Fig. 1): symmetry-conserving (resp. -breaking) shape transition, for which the symmetry of the precipitate shape is equal to or greater (resp. less) than the symmetry resulting from the intersection of the point group of the precipitate and matrix phases. By definition, the circle–ellipse bifurcation occurring in the basal plane and studied in Ref. [6] is a symmetry-breaking transition. Indeed, applying the Curie principle, the resulting shape of a symmetry-conserving transition should contain at least a three-fold axis of symmetry $\left(\frac{6}{m}\frac{2}{m}\frac{2}{m}\cap\bar{3}\frac{2}{m}=\bar{3}\frac{2}{m}\right)$, which is not the case of an ellipse with distinct semi-axes. It was established (under assumptions specified in Ref. [6]) that this transition occurs if $L > 4$ (Fig. 1), where L is given by:

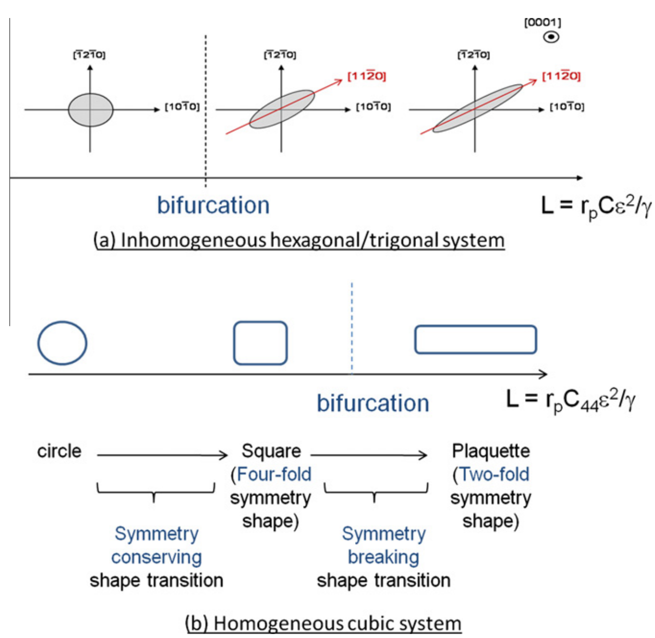

Fig. 1. Diagram representing the 2-D bifurcation of (a) a misfitting trigonal precipitate inside the basal plane of a hexagonal matrix (inhomogeneous elasticity) and (b) a particle in a homogeneous cubic system with a purely dilatational misfit.