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Microstructural Development of Coherent Tetragonal Precipitates in Magnesium-Partially-Stabilized Zirconia: A Computer Simulation

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A new computer simulation technique for modeling morphological pattern formation during nucleation, growth, and coarsening of coherent misfitting particles is developed. Microstructure evolution during precipitation of tetragonal phase from Mg-partially-stabilized cubic zirconia is investigated. Our computer simulation shows that during the initial stage of precipitation, the tetragonal particles formed by homogeneous nucleation display strong alignment along certain crystallographic directions, forming the so-called "tweed" pattern. During subsequent growth of the spatially correlated nuclei, an alternating band structure is observed, with each individual band consisting of lens-like-shaped tetragonal phase particles of the same orientation variant dispersed in the cubic matrix. The particles in the neighboring bands are twin-related. The microstructures obtained in our computer simulation seem to agree well with the experimental observations.

I. Introduction

One of the efficient ways to control microstructures and thus properties of advanced multiphase alloys is to control phase transformations by carefully choosing the processing parameters such as composition, temperature, and applied fields. A typical example is the transformation-toughened ZrO_2-containing ceramics.[1,2] Therefore, it is desirable to carry out computer simulations of microstructural development during a phase transformation. However, because of the complexity of the problem, e.g., the multibody and highly nonlinear characteristics, there is no comprehensive model which can deal with the rich variety of transformations underlying the microstructural development in advanced ceramics and metal alloys. Probably the most promising way to address this problem is using the phenomenological field approach based on the nonlinear deterministic Ginzburg–Landau or corresponding stochastic Langevin equations. A rich literature on the applications of this approach can be found in the recent review by Gunton *et al.*[3] The field method, unlike the conventional theory of phase transformation, does not have the problems caused by the complex boundary conditions on moving interfaces and can deal with arbitrary two-phase morphologies. It characterizes the transformation kinetics by a temporal evolution of continuum fields of concentration (for compositional heterogeneity) and long-range order (lro) parameters (for structural inhomogeneity).

There is, however, a major limitation about the current formalism of the field approach; i.e., the transformation-induced elastic strain generated by a misfit between the crystal lattices of coexisting coherent phases or their different orientation variants is generally not taken into account. This limitation drastically reduces the predicting power and realism of the theory because, in most cases, it is the accommodation of the coherency strain that dominates the microstructural development, e.g., the shapes of precipitates and their spatial arrangement.

Although there have been several attempts to include the effect of the coherency strain in the deterministic Cahn–Hilliard field equations,[4,5] they have been made only for a particular case of isostructural spinodal decomposition in cubic systems characterized by the concentration field. No attempt has been made to formulate a kinetic model which is applicable to systems like the partially stabilized ZrO_2 where, besides composition, structural rearrangement producing a low symmetry phase with several orientation variants is involved. This is in spite of the fact that the majority of phase transformations occurring in advanced engineering materials fall into this category. Furthermore, the coherency strain effect has never been introduced into the stochastic kinetic equations to study the features of nucleation in coherent solids.

The purpose of this paper is to formulate a general but still tractable stochastic field model and apply it for simulating the decomposition process in Mg-partially-stabilized ZrO_2. This model allows one to describe evolution of an arbitrary two-phase morphology without constraints on the transformation path, to take into account the coherency strain and elastic anisotropy for an arbitrary crystal lattice rearrangement, and to consider all three stages of the transformation including nucleation, growth, and coarsening within the same physical and mathematical formalism.

II. Stochastic Field Kinetic Model

The field approach to the kinetics of shape evolution of a single tetragonal phase particle coherently embedded in a cubic matrix has recently been proposed based on the time-dependent Ginzburg–Landau (TDGL) equations.[6] To describe an arbitrary particle morphology of the tetragonal phase which has a different symmetry with the cubic matrix, the continuous field of the lro parameter, $\{\eta(\mathbf{r})\}$, is introduced in addition to the concentration field, $\{c(\mathbf{r})\}$.

In this paper, we generalize this approach by introducing a multicomponent lro parameter, $\eta_\alpha(\mathbf{r})$, where $\alpha = 1, 2, 3, \ldots, \nu$, numbering independent orientation variants, and by adding the Langevin noise terms to the TDGL equations. The corresponding stochastic Langevin equations become

$$\frac{\partial c(\mathbf{r},t)}{\partial t} = M\nabla^2 \frac{\delta F}{\delta c(\mathbf{r},t)} + \xi_c(\mathbf{r},t) \tag{1a}$$

$$\frac{\partial \eta_\alpha(\mathbf{r},t)}{\partial t} = -L \frac{\delta F}{\delta \eta_\alpha(\mathbf{r},t)} + \xi_\alpha(\mathbf{r},t) \tag{1b}$$

where t is time, M and L are the kinetic coefficients characterizing the diffusional mobility and the lro relaxation, respectively, F is the total free energy including the strain energy contribution, and ξ_c and ξ_α are the Langevin noise terms which satisfy the requirements of the fluctuation-dissipation theorem:[3,7]

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$$\langle \xi_c(\mathbf{r}',t')\, \xi_c(\mathbf{r},t)\rangle = -2k_B TM\nabla^2\delta(\mathbf{r}-\mathbf{r}')\,\delta(t-t')$$

$$\langle \xi_\alpha(\mathbf{r}',t')\, \zeta_\beta(\mathbf{r},t)\rangle = 2k_B TL\delta(\mathbf{r}-\mathbf{r}')\,\delta(t-t')\delta_{\alpha\beta}$$

where k_B is the Boltzmann constant and T is the temperature. This generalization makes the model applicable to the entire microstructural development, including the nucleation, in a multiparticle decomposing system with several orientation variants of a low-symmetry phase. The problem of theoretical characterization of microstructural evolution is simply reduced to a solution of the Langevin equations (1).

According to the elasticity theory of multiphase coherent solids of Khachaturyan,[8] the elastic strain energy E_{el} generated by an arbitrary concentration or structure heterogeneity can be presented as a functional of the concentration or lro parameter fields. Particularly in the homogeneous modulus case and harmonic approximation, it can be expressed in a simple form of pairwise interaction. For example, if the strain is predominantly caused by concentration heterogeneity, the strain energy is

$$E_{el} = \frac{1}{2}\int_V\int_V W(\mathbf{r}-\mathbf{r}')_{el}\, c(\mathbf{r})\, c(\mathbf{r}')\,\frac{d\mathbf{r}\, d\mathbf{r}'}{v_0^2} \qquad (2)$$

where $W(\mathbf{r}-\mathbf{r}')_{el}$ is the strain-induced interaction potential between the finite elements at points $\mathbf{r}$ and $\mathbf{r}'$, and v_0 is the unit-cell volume of the crystal. The integration in Eq. (2) is carried out over the entire system volume V. The potential $W(\mathbf{r}-\mathbf{r}')_{el}$ can be expressed as

$$W(\mathbf{r})_{el} = \int v_0^2\,[-n_i\sigma^\circ_{ij}\Omega(\mathbf{n})_{jk}\sigma^\circ_{kl}n_l + \langle m_i\sigma^\circ_{ij}\Omega(\mathbf{m})_{jk}\sigma^\circ_{kl}m_l\rangle_{\mathbf{m}}]\exp(i\mathbf{kr})\,\frac{d\mathbf{k}}{(2\pi)^3} \qquad (3)$$

where $\sigma^\circ_{ij} = c_{ijkl}\varepsilon^\circ_{kl}$, c_{ijkl} is the elastic moduli tensor, ε°_{kl} is a tensor of the concentration expansion of the crystal lattice, $\mathbf{n}$ and $\mathbf{m}$ are unit vectors, $\Omega(\mathbf{n})_{ij}$ is a tensor which is the reverse of the tensor $\Omega(\mathbf{n})^{-1}_{ij} = c_{ijkl}n_k n_l$, and $\langle ...\rangle_{\mathbf{m}}$ is a symbol indicating averaging over all directions of $\mathbf{m}$.

If the strain effect is predominantly caused by the lro parameter heterogeneity, resulting in symmetry changes, then the strain energy is

$$E_{el} = \frac{1}{2}\sum_{\alpha\beta}\int_V\int_V W(\mathbf{r}-\mathbf{r}')^{\alpha\beta}_{el}\,\eta^2_\alpha(\mathbf{r})\,\eta^2_\beta(\mathbf{r}')\,\frac{d\mathbf{r}\, d\mathbf{r}'}{v_0^2} \qquad (4)$$

The potential $W(\mathbf{r}-\mathbf{r}')^{\alpha\beta}_{el}$ reads

$$W(\mathbf{r})^{\alpha\beta}_{el} = \int v_0^2[c_{ijkl}\,\varepsilon(\alpha)^\circ_{ij}\,\varepsilon(\beta)^\circ_{kl} - n_i\sigma(\alpha)^\circ_{ij}\,\Omega(\mathbf{n})_{jk}\,\sigma(\beta)^\circ_{kl}\,n_l]\exp(i\mathbf{kr})\,\frac{d\mathbf{k}}{(2\pi)^3} \qquad (5)$$

where $\sigma(\alpha)^\circ_{ij} = c_{ijkl}\,\varepsilon(\alpha)^\circ_{kl}$ and $\varepsilon(\alpha)^\circ_{ij}$ is the stress-free transformation strain which is related to the stress-free strain field by the equation $\varepsilon^\circ_{ij}(\mathbf{r}) = \varepsilon(\alpha)^\circ_{ij}\eta^2_\alpha(\mathbf{r})$.

The strain-induced interaction potentials $W(\mathbf{r})_{el}$ and $W(\mathbf{r})^{\alpha\beta}_{el}$ defined by Eqs. (3) and (5) depend on the separation distance as r^{-3}. This infinite-range, highly anisotropic strain-induced interaction is a key factor dominating the microstructural evolution of coherent systems.

For the purpose of microstructure characterization, the chemical free energy F_0 can be approximated by the "coarse grain" free energy functional of the concentration and lro parameter fields

$$F_0 = \int_V\left[\frac{1}{2}a_{ij}\nabla_i c\nabla_j c + \frac{1}{2}\sum_{\alpha=1}^{\alpha=\nu} b(\alpha)_{ij}\nabla_i\eta_\alpha\nabla_j\eta_\alpha + f(c;\eta_1,\eta_2,\eta_3,\ldots,\eta_\nu)\right]\frac{d\mathbf{r}}{v_0} \qquad (6)$$

where a_{ij} and $b(\alpha)_{ij}$ are the gradient energy coefficients, i and j are the indexes of Cartesian coordinates, $\nabla_i = \partial/\partial r_i$ is a differential operator, and $f(c;\eta_1,\eta_2,\eta_3,...\eta\nu)$ is the local specific free energy. The gradient energy terms in Eq. (6) describe the interactions between long-wave fluctuations in adjacent finite volumes and they provide isotropic interfacial energies.

III. Model System

The partially stabilized ZrO_2 is a focus of considerable scientific interest because of its technological significance. The interesting combination of decomposition with a cubic → tetragonal crystal lattice rearrangement makes this ceramic material an excellent generic example illustrating an important class of coherent transformations which produce several orientation variants of a low-symmetry product phase. Below we apply the generalized stochastic field model to examine the microstructural evolution during decomposition in Mg-partially-stabilized ZrO_2 (Mg-PSZ).

When Mg-PSZ is quenched from a high-temperature single cubic phase region into a two-phase field of the phase diagram consisting of cubic and tetragonal phases, three independent orientation variants of the tetragonal phase nucleate homogeneously throughout the cubic parent phase matrix. The tetragonal phase particles maintain coherency with the cubic phase matrix during their growth and coarsening and the coherency strain is accommodated by changing particle shapes and developing specific mesoscopic structure patterns. For the sake of simplicity, we start from a two-dimensional (2D) model system. In this case there are only two possible orientation variants (α = 1, 2). The specific free energy functional in Eq. (6) can be approximated by a Landau expansion polynomial[6]

$$f(c,\eta_1,\eta_2) = \frac{1}{2}A(c-c_1)^2 + \frac{1}{2}B(c-c_2)(\eta_1^2+\eta_2^2) - \frac{1}{4}D(\eta_1^4+\eta_2^4) + \frac{1}{6}G(\eta_1^2+\eta_2^2)^3 \qquad (7)$$

where η_1 and η_2 are the lro parameters which are actually the amplitudes of the symmetry breaking optical modes. They describe the two possible orientation variants of the tetragonal phase in 2D with the tetragonal axes along [10] and [01] directions, respectively. A, B, D, and G are positive constants and c_1 and c_2 are constants close to the equilibrium compositions of the cubic and tetragonal phases, respectively. The choice of these phenomenological constants should satisfy the topological requirements for free energy to characterize the first-order transition of a cubic phase into a mixture of cubic and tetragonal phases. This means that the dependence of f on η_i should provide a local minimum at $\eta_1 = \eta_2 = 0$ and global minima at $\eta_1 \neq 0$, $\eta_2 = 0$, or $\eta_1 = 0$, $\eta_2 \neq 0$. The dependence of f on c should also provide equilibrium compositions of the cubic and tetragonal phases observed experimentally. The particular set of values used in the simulations—$A = 80.0$, $B = 5.0$, $D = 0.5$, $G = 1.4$ (in the units of k_BT, where $T = 1420$°C), $c_1 = 0.16$, and $c_2 = 0.04$—meet these requirements. With these parameters, Eq. (7) provides one of the simplest free energy models, which allows one to illustrate the generic characteristics of microstructure development during phase separation with a symmetry reduction generating two orientation domains.

Minimizing the free energy (7) with respect to η_1 and η_2 at given c allows us to express the minimizing lro parameters through composition c and hence express the free energy in terms of c only. The obtained dependence of the free energy on c for the chosen set of parameters is illustrated by Fig. 1. The free energy has two branches, one describing the tetragonal phase and the other describing the cubic phase. The equilibrium compositions determined by the common tangent construction are $c_{cub} = 0.155$ and $c_{tetr} = 0.04$ for the cubic and tetragonal phases, respectively. They are close to the equilibrium compositions of Mg-PSZ at 1420°C.[9]

The gradient energy coefficient matrices in the "coarse grain" chemical free energy (6) are assumed to have the following simple forms:

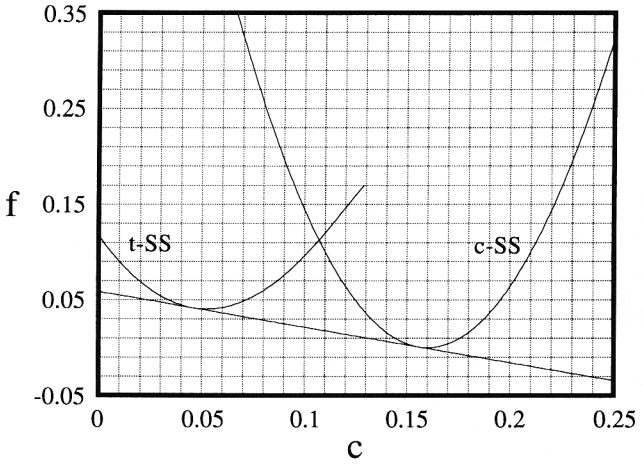

Fig. 1. Specific free energy vs composition curves for both the tetragonal and cubic solid solutions (see text for explanation).

$$a_{ij} = a\delta_{ij}$$
$$b(\alpha)_{ij} = b\delta_{ij}$$

and a and b are chosen to be 0.25 (in the units $a_0^2 k_B T$) where a_0 is a characteristic length unit of the mesh grid.

For characterization of the cubic → tetragonal transformation in PSZ, we assume that the crystal lattice parameters of the tetragonal phase are determined by the lro parameter only. In this case the strain energy (4) can be used. The elastic constants and lattice parameters of Mg-PSZ at 1420°C,[10] $c_{11} = 3.08$, $c_{12} = 0.69$, $= c_{44} = 0.36$ (10^{12} erg/cm^3) and $a_c = 5.094$, $a_t = 5.091$, and $c_t = 5.204$ (Å), are used in the calculation. A reduced time, $\tau = t/t_0$, where $t_0 = (Lk_BT)^{-1}$, is used and the ratio $M/(La_0^2)$ is fixed to 0.4 (where L and M are the kinetic coefficients in Eqs. (1)). The Langevin noise terms in Eqs. (1) are generated by a random number generator. With those chosen parameters, the kinetic equations (1) are solved numerically using a Fourier space (reciprocal space) representation for the 2D model systems consisting of 256 × 256 mesh points of a square grid. Periodic boundary conditions are applied.

IV. Results and Discussion

The initial condition for the simulation (initial condition for Eqs. (1)) is a homogeneous supersaturated cubic solid solution with an average composition $c = 0.12$. According to Eq. (7), it is in a metastable state corresponding to a local free energy minimum. Therefore, the phase transformation occurs through a nucleation process driven by the noise terms in the Langevin equations (1). Because this field approach does not impose any constraint on the transformation path, it allows the system itself to "choose" the optimal evolution path and to adopt the optimal structural and compositional configurations corresponding to a critical nucleus state. It is particularly interesting to see what these configurations look like. To do this, we artificially "turn off" the noise terms in Eqs. (2) after a certain period of time in the simulation. Under this condition, all heterogeneities corresponding to the "noncritical" fluctuations will eventually disappear and the "surviving" heterogeneities which are able to grow are actually the critical or operational nuclei. Since the infinite-ranged anisotropic strain-induced interaction between inhomogeneities is taken into account, the strain accommodation should affect the character of a nucleation process. The numerical solution of Eqs. (1) for this system shows that this is exactly the case. Figure 2(A) demonstrates a spatial pattern formed by the surviving nuclei for one of the two orientation variants of the tetragonal phase at $\tau = 5$ after switching off the noise terms. The contrast describes the value of $\eta_1(\mathbf{r})$ (the higher the value, the brighter the shade). The strong spatial correlation (alignment along the ⟨11⟩ directions) is readily seen. The simulated

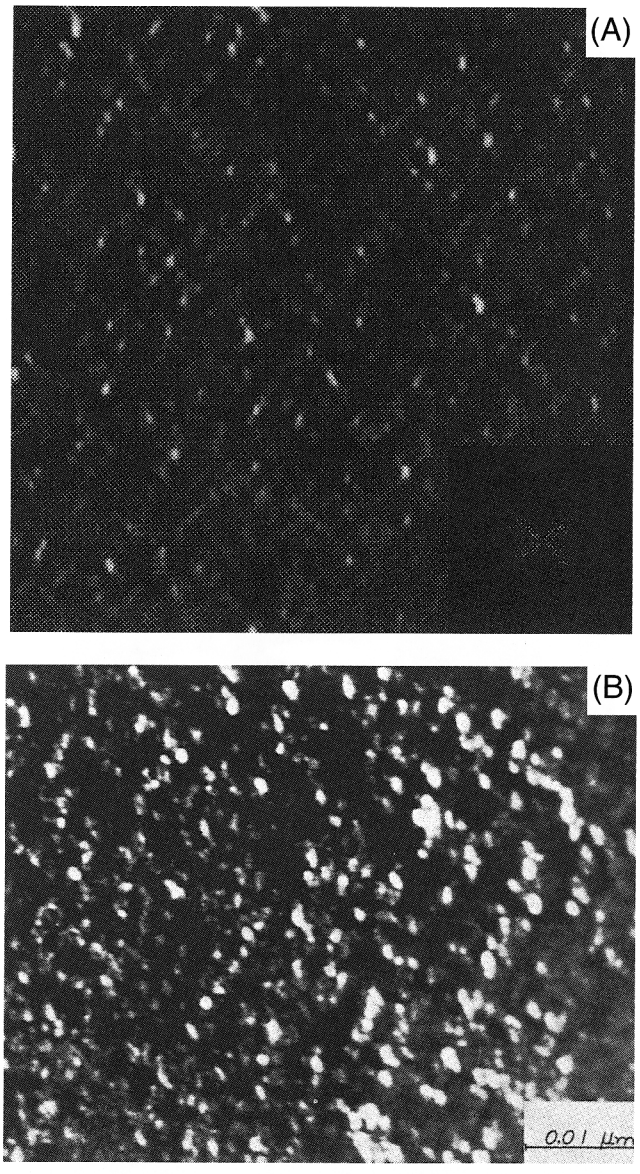

Fig. 2. (A) lro parameter fields $\eta_1(\mathbf{r})$ at $\tau = 5$ showing the tweed structures formed during homogeneous nucleation of t-ZrO_2 particles from the c-matrix. The insets represent normalized diffuse scattering intensities around the reciprocal lattice origin. (B) Homogeneously nucleated precipitates of one orientation variant during quenching of M-PSZ (courtesy of A. H. Heuer).

diffuse diffraction pattern shown in the small inset of Fig. 2(A) forms a cross about the reciprocal lattice point, which is typical for a tweed pattern.

This simulation result agrees very well with the experimental observations at an early stage of decomposition in Mg-PSZ (see Fig. 2(B).[11] It predicts a new phenomenon in a coherent system, i.e., a correlated nucleation in which a new nucleation event is affected by previously formed nuclei. This phenomenon seems not to have been reported in earlier works and it is probably quite general for coherent systems with considerable contribution of the strain energy.

The graphic visualization of solution of the Langevin equations related to later stages of microstructural development is shown in Fig. 3, where the morphology is characterized by shades of gray in accordance with the concentration profile $c(\mathbf{r},t)$. The greater the concentration, the darker is the shade. Since the strain-induced interaction becomes stronger and stronger as the nuclei grow, the tweed pattern eventually evolves into an alternating band structure with each individual band aligned along approximately the ⟨11⟩ directions (see

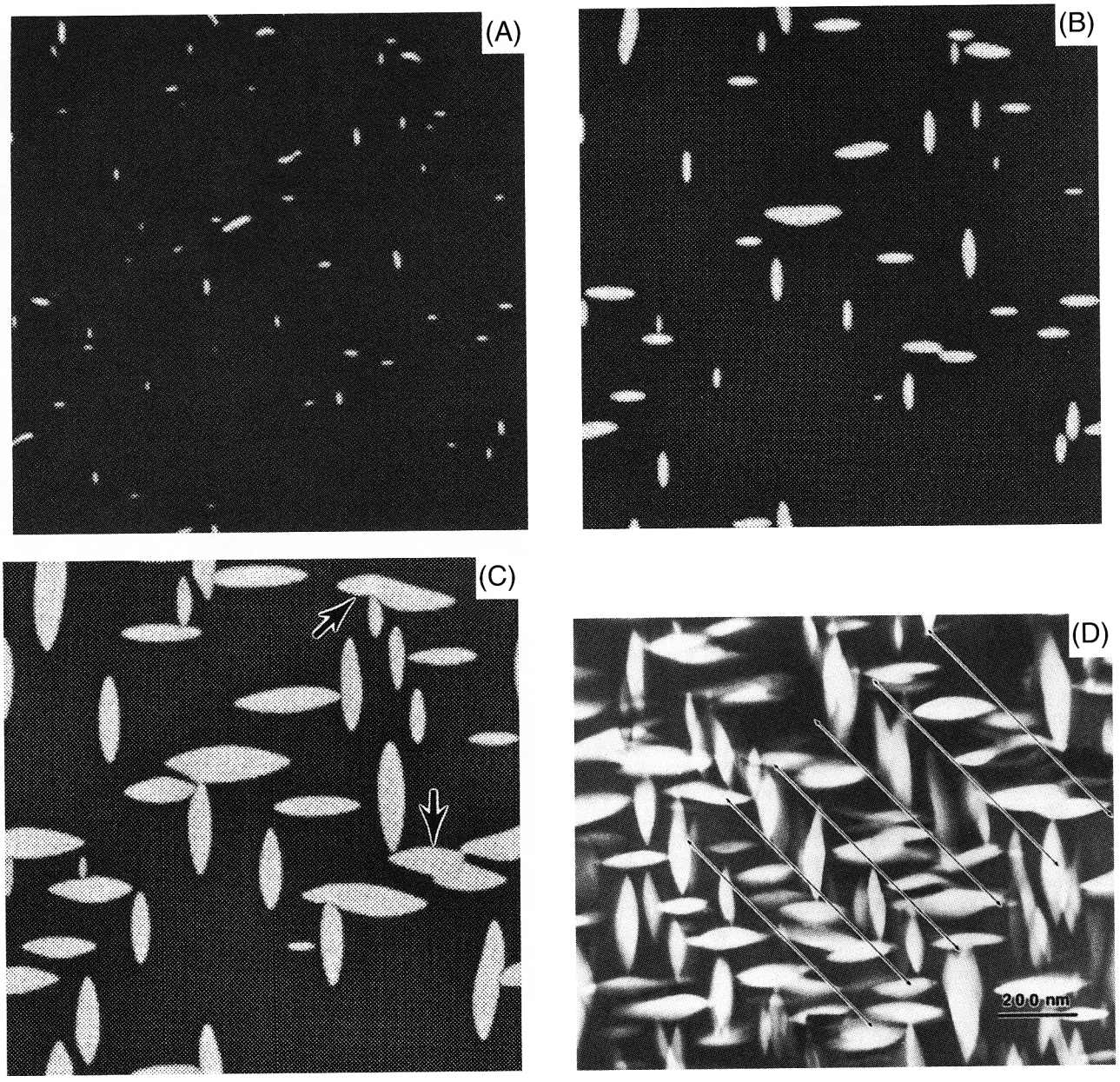

Fig. 3. Temporal and spatial evolution of two-phase morphological patterns consisting of many t-ZrO_2 particles formed by homogeneous nucleation from a supersaturated cubic matrix: (A) $\tau = 10$; (B) $\tau = 60$; (C) $\tau = 100$. (D) Bands of alternating variants observed in Mg-Y PSZ after 5 h of aging (courtesy of C. A. Bateman and M. R. Notis).

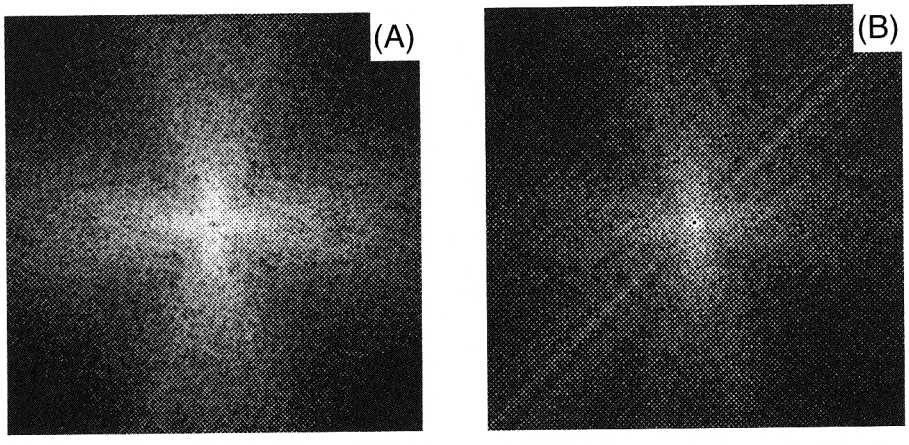

Fig. 4. (A, B) Simulated diffuse scattering patterns for the alternating band structures shown in Figs. 3(C) and (D), respectively. The parallel lines across the diagonal of (B) are due to the lines drawn on the TEM image of Fig. 3(D).

Figs. 3(A–C)). Each band contains particles of the same orientation variant. The particles in the neighboring bands are twin-related. The obtained mesoscopic structures are also in excellent agreement with the experimental observations in Mg-PSZ[10,12] (see, for example, Fig. 3(D)). A comparison between the diffuse scattering pattern calculated from the simulated structure in Fig. 3(C) and the one obtained by digitizing the electron microscopic image shown in Fig. 3(D) is given in Fig. 4. The agreement between the two patterns is excellent. Such an alternating band structure seems to be a typical pattern formed if decomposition occurs with symmetry reduction. For example, it was also observed in a metal alloy Ni-Fe-Cr[13] which undergoes a cubic → tetragonal decomposition.

The tetragonal particles shown in Figs. 3(C) and (D) have basically a lens-like shape, which is caused by strain-induced anisotropic growth; e.g., the particles grow much faster along one of the <10> directions which is parallel to their *a* axis than the other which is parallel to their *c* axis where the lattice misfit is maximal. This prediction is in a good agreement with a previous investigation of a single-particle system.[6] However, some complicated particle shapes due to the multiparticle effect are also observed (see the arrows).

V. Conclusion

A computational method for modeling microstructural development during coherent phase transformation has been developed. Because of its stochastic nature and its ability to deal with arbitrary transformation strains, it is practically applicable to all stages (nucleation, growth, and coarsening) of any coherent transformations, including diffusional precipitation and diffusionless martensitic transformation or both. Its application to a typical advanced ceramic, Mg-PSZ, shows a remarkable predicting power. For example, a new phenomenon, the correlated nucleation of different orientation variants of a low-symmetry product phase with nuclei forming special strain accommodating patterns, is predicted. The simulated alternating band structure formed by self-organized orientation variants of tetragonal phase particles is also in excellent agreement with the experimental observations. These simulation results demonstrate that the proposed model can be efficiently used for theoretical characterization and predicting microstructural evolution during processing or in service performance of complicated engineering materials.

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