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On the theory of grain growth in systems with anisotropic boundary mobility

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Abstract

We analyze grain growth kinetics in systems with anisotropic grain boundary mobility. In contrast to most previous studies of grain growth dynamics, we relax self-similarity assumptions that strongly constrain the dynamics and statistics during microstructural evolution in polycrystalline materials. We derive analytical expressions for the average growth rate within each topological class of *n*-sided grains as well as for the growth rate of the average grain area; we explain the results using underlying symmetries. Although anisotropic grain growth may in general be non-linear in time, we show, even in the absence of the self-similarity constraint, that the evolution kinetics obeys the von Neumann–Mullins relationship in the two limiting cases of textured and fully random microstructure with a time dependence solely determined by changes in the misorientation distribution. Our analytical results agree well with recent computer simulations using a generalized phase field approach.

Keywords: Grain growth; Anisotropic systems; Theory & modeling

1. Introduction

Grain growth and the evolution of cellular structures have been studied for decades using a variety of theoretical and experimental approaches [1–8]. Most theoretical analysis is in the isotropic limit with all boundary properties constant, i.e., orientation and inclination independent. In reality, however, both the energy and mobility of grain boundaries can be strongly anisotropic; a question of interest is how does this anisotropy influence grain growth? Since many new materials have strongly anisotropic microstructures and unique physical properties, a fundamental understanding of the effect of anisotropy on microstructural evolution is desired.

A basic aspect of the dynamics of grain growth is the possible self-similarity of microstructural evolution, under which configurations at successive times have identical statistics when transformed by a uniform linear scale of magnification. Evidence in the isotropic limit indicates that, after an initial transient, self-similarity holds for many

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aspects of grain growth [9]. In this case, the average (2D) grain area grows linearly with time and all statistical distributions are time-invariant. However, self-similarity may not develop when boundary properties like energy or mobility are anisotropic, which can result in strong nonlinearities in the growth kinetics of a polycrystalline aggregate [10]. In our recent simulations of grain growth in 2-D anisotropic systems non-self similar evolution occurred [11]. On the other hand, if self-similarity is valid in anisotropic systems, Mullins showed that the growth kinetics will be linear, as in the isotropic case [12]. The basic question we wish to address is whether self-similar evolution is necessary for the simple evolution of grain growth in anisotropic systems or only sufficient as Mullins showed.

Due to complexity of theoretical treatment of grain growth many studies use computer simulations [13–22]. These models usually consider only a few grain types or include only the misorientation or the inclination-angle dependence of the grain boundary properties but not both. Our recent computer simulations used a generalized Phase Field Model that considered the full range of misorientations as well as inclinations [11]. We found that the single grain growth rate in the presence of triple junctions and anisotropic grain boundary mobility was time dependent, in contrast to the isotropic case or the case of an island grain with mobility anisotropy. The growth exponent for the average grain area in a polycrystalline system deviated only slightly from unity, however.

In this paper we present a theoretical analysis of grain growth kinetics in two-dimensional polycrystals with anisotropic boundary mobility. Although direct application to experimental observations is limited by the restriction to isotropic grain boundary energy, the model is the first step towards the understanding of grain growth kinetics in anisotropic systems. The two-dimensional results may be relevant to columnar grain structures that are widely observed in thin films and coatings. In principle, both theoretical analysis and Phase Field modeling can be extended to 3D although technical complexity significantly increases. The major difference from the previous attempts to understand kinetics of anisotropic polycrystals is the relaxation of the self-similarity constraint and use of a grain boundary mobility that is a general function of both inclination and misorientation. Section 2 presents the analytical analysis of grain growth kinetics with mobility anisotropy. In Section 3, results of the analytical analysis are compared with Phase Field computer simulations. Major conclusions are summarized in Section 4.

2. Theoretical analysis

In this section we evaluate the effect of grain boundary mobility anisotropy on the average growth rate of grains within a given topological class of n-sided grains as well as on the growth rate of the average grain area.

Following the analysis of Mullins [23] for the isotropic case, we write the growth rate of an n-sided grain as:

$$\frac{dA_n}{dt} = -k\sum_{i=1}^{n}\int_{\beta_i^{(out)}}^{\beta_i^{(in)}} L_i(\theta_i,\beta)d\beta, \tag{1}$$

where $\beta_i^{(out)}(\beta_i^{(in)})$ is the inclination of the ith side of an n-sided grain at the triple junction, where labels out (in) indicate that the side is outgoing (incoming) to the triple junction, θ_i is the grain boundary misorientation angle (angle which measures relative grain rotation angle) of the ith boundary and k is a constant (see Fig. 1). In this study we consider tilt grain boundaries, where the axis of relative grain rotation lies in the plane of the boundary. The grain boundary inclination, β_i, introduced in Fig. 1, is defined with respect to fixed global coordinates. However, boundary properties such as energy and mobility depend on the structure of the boundary, which is determined by inclination of the boundary with respect to the local coordinates. For example, in cubic crystals with small misorientation angles, we can measure inclination from the orientation of the symmetric tilt boundary (a line which bisects the angle between equivalent crystallographic orientations of the grains, e.g. [100] axis of the two crystals), which is the most convenient choice for the local coordi-

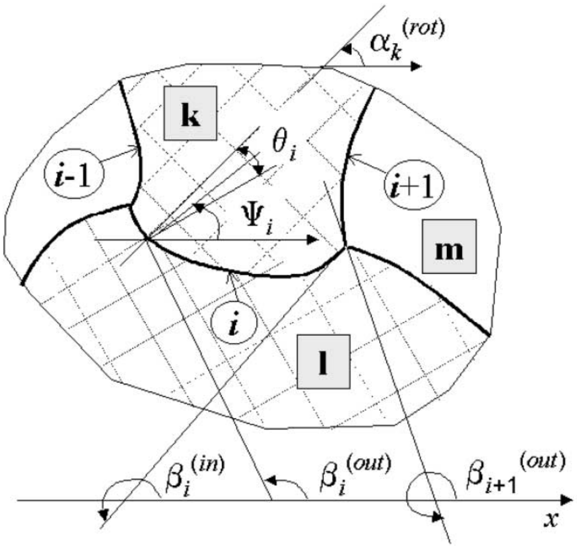

Fig. 1. Schematic drawing of an arbitrary-sided grain with triple junctions. Labels $(i-1)$, i, $(i+1)$ denote different sides of the grain. $\beta_i^{(out)}$ and $\beta_i^{(in)}$ are the outgoing and incoming inclinations of the ith grain side at the triple junctions, which are measured in the global coordinate system. ψ_i is the rotation angle of the local coordinate system associated with the ith boundary with respect to the same global system. $\alpha_k^{(rot)}$ is the rotation angle of the grain k with respect to the global system.

nates. Thus, in equation (1) the boundary mobility L_i carries a subscript i, which represents the fact that mobility of each of the boundaries is defined in its own coordinate system. We denote by ψ_i the angle of the symmetric tilt boundary in the global coordinates. Then, the local inclination of the boundary, ϕ, is $\phi = \beta - \psi_i$. Introducing $\phi_i^{(out)} = \beta_i^{(out)} - \psi_i$ and $\phi_i^{(in)} = \beta_i^{(in)} - \psi_i$, the expression for the growth rate of the n-sided grain is,

$$\frac{dA_n}{dt} = -k \sum_{i=1}^{n} \int_{\phi_i^{(out)}}^{\phi_i^{(in)}} L(\theta_i,\phi)d\phi, \tag{2}$$

where L is the mobility of the boundary as a function of its misorientation and local inclination.

For simplicity let us assume for the moment that grain boundary mobility is a function of inclination only, $L = L(\phi)$. Later, we examine the influence of the misorientation dependence of the mobility on growth kinetics. As we show below, in both cases the average growth rate of the grain area in a topological class is a sum of two terms, one of which corresponds to the von Neumann–Mullins expression observed in the isotropic limit, while the other time-dependent contribution gives the deviation of the grain growth kinetics from linear behavior. Finally, in several limiting cases we can estimate the second term using simple symmetry arguments, although we require an additional assumption when the grain boundary mobility is a function of both misorientation and inclination.

As a first step, we rewrite equation (2) in a more convenient form. Introduction of triple junctions results in discontinuous changes in the inclination at the vertex. Thus, unlike single-sided grains, $\oint d\beta L(\beta)$ is no longer equal to $2\pi \langle L \rangle_\beta$, because the integration omits some inclinations. However, to add and subtract from equation (2) the terms that complete the integration over the entire range $0 \le \phi \le 2\pi$ is useful, since, equation (2) becomes:

$$\frac{dA_n}{dt} = -2\pi k \langle L \rangle_\phi + k \sum_{i=1}^{n} \int_{\phi_i^{(in)}}^{\phi_{i+1}^{(out)}} L(\phi)d\phi, \tag{3}$$

where

$$\phi_{n+1}^{(out)} \equiv \phi_1^{(out)}.$$

Due to isotropic energy assumption the angles of the boundaries at a triple junction are all $2\pi/3$. Therefore, with our convention for incoming and outgoing lines (see Fig. 1), the rotation angle from an incoming to an outgoing line at a triple junction is, $\beta_{i+1}^{(out)} - \beta_i^{(in)} = \pi/3$. Then, using the relationship between the local and the global coordinate systems defined above, we can deduce that:

$$\delta\phi \equiv \phi_{i+1}^{(out)} - \phi_i^{(in)} = \frac{\pi}{3} - (\psi_{i+1} - \psi_i). \tag{4}$$

Even for isotropic grain boundary energy, the relationship between the *locally* defined boundary inclinations at a triple junction is no longer equal to $\pi/3$. To simplify the notation we let $\varepsilon_{i,i+1} \equiv \psi_{i+1} - \psi_i$, which measures the relative rotation of the local coordinates for the incoming and outgoing boundaries at a particular vertex and defines the deviation of $\delta\phi$ from $\pi/3$. Obviously, the value of $\varepsilon_{i,i+1}$ is invariant with respect to global rotations, since both ψ_{i+1} and ψ_i are defined relative to the global system. As a result we can always express $\varepsilon_{i,i+1}$ in terms of variables defined in local coordinates independent of the particular global choice.

Consider now the rotation of the kth grain with respect to the global system, α^k_{rot} (see Fig. 1). Then, the misorientation between grains k and l is $\theta^{kl} = |\alpha^k_{rot} - \alpha^l_{rot}|$. If we define local coordinates by the symmetrical tilt boundaries, then for the boundary between grains k and l, the rotation of the local coordinates with respect to the global coordinates is $\psi_i = (\alpha^k_{rot} + \alpha^l_{rot})/2$. Hence $\varepsilon_{i,i+1}$ depends only on the misorientations of the ith and (i+1)st boundaries and is:

$$\varepsilon_{i,i+1} = \delta^{kl}\theta^{kl} - \delta^{km}\theta^{km}, \tag{5}$$

where k, l and m are the grains meeting at a given triple junction, δ^{kl} and δ^{km} are equal to $\pm 1/2$. The sign of $\delta^{kl}(\delta^{km})$ depends on whether the rotation from grain k to grain $l(m)$ is clockwise or counterclockwise. In the following analysis we consider the counterclockwise direction to be positive.

Because of the symmetry of the underlying lattice the mobility is a periodic function of the boundary inclination with a period of $T_L = 2\pi/m$ where m is a positive integer. Thus without loss of generality:

$$L(\phi) = \langle L\rangle_\phi(1 + f(\phi)), \tag{6}$$

where $f(\phi)$ is a periodic function whose symmetry is determined by the structure and symmetry of the underlying lattice. Moreover, since $f(\phi)$ has the period of the non-negative mobility function, $f(\phi)$ must satisfy the conditions:

$$\frac{1}{2\pi}\int_0^{2\pi} f(\phi)d\phi = 0 \quad \text{and} \quad \max|f(\phi)| < 1. \tag{7}$$

Substituting equations (4)–(6) into equation (3) yields the growth rate of a single n-sided grain:

$$\frac{dA_n}{dt} = \frac{k\pi}{3}\langle L\rangle_\phi(n-6) + k\sum_{i=1}^{n}\int_{\phi_i^{(in)}}^{\phi_i^{(in)}+\frac{\pi}{3}-\varepsilon_{i,i+1}} f(\phi)d\phi \tag{8}$$

From equation (8) we can see that the growth rate of a single n-sided grain consists of two parts. The first term is identical to the von Neumann–Mullins expression obtained for the isotropic case; it is time independent and gives the grain evolution in terms of average properties. The second term contains the effect of mobility anisotropy and is in general time dependent. This term provides the difference between the growth rates of an n-sided grain for systems with isotropic and anisotropic boundary mobility. Clearly, setting $\langle L\rangle_\phi \equiv L = const$ and $f(\phi) \equiv 0$ in equation (8) recovers the isotropic limit.

To calculate the average growth rate of a single n-sided grain embedded in a polycrystalline aggregate, we introduce a vertex distribution function $g_n(\theta_1,\ldots,\theta_n,\phi_1^{(in)},\ldots,\phi_n^{(in)})$, which is the probability of finding an n-sided grain with inclinations $\phi_i^{(in)}$ of the ith grain boundary (with misorientation θ_i), incoming to a triple junction. Because of the relationship between the incoming and outgoing inclinations of the boundaries at a triple junction given by equation (4), g_n is a function of the incoming inclinations only. We choose g_n such that it satisfies the normalization condition:

$$\int_0^{\Theta}\ldots\int_0^{\Theta} d\theta_1\ldots d\theta_n \int_0^{2\pi}\ldots\int_0^{2\pi} d\phi_1^{(in)}\ldots d\phi_n^{(in)} g_n(\theta_1,\ldots,\theta_n,\phi_1^{(in)},\ldots,\phi_n^{(in)}) = 1. \tag{9}$$

Then, the average growth rate of all n-sided grains is:

$$\left\langle\frac{dA_n}{dt}\right\rangle = \int_0^{\Theta}\ldots\int_0^{\Theta} d\theta_1\ldots d\theta_n \int_0^{2\pi}\ldots\int_0^{2\pi} d\phi_1^{(in)}\ldots d\phi_n^{(in)} g_n(\theta_1,\ldots,\theta_n,\phi_1^{(in)},\ldots,\phi_n^{(in)})\frac{dA_n}{dt}. \tag{10}$$

The misorientation dependence appears only through the integration limits in equation (8) and the grain boundary mobility function depends only on the boundary inclination. Substituting equation (8) into equation (10) and evaluating the corresponding integrals we obtain the average growth rate of the n-sided grains:

$$\left\langle \frac{dA_n}{dt} \right\rangle = \left\langle \frac{dA_n}{dt} \right\rangle_0 + \delta \left\langle \frac{dA_n}{dt} \right\rangle. \tag{11}$$

Calculation of the first term is trivial due to equation (9), $\langle dA_n/dt\rangle_0 = (n-6)\langle L\rangle_\phi k\pi/3$. Here we focus on evaluating the second term, which introduces nonlinearities into the growth rate. For convenience we define an auxiliary variable $F(\phi) = \int f(\phi)d\phi$. Both $F(\phi)$ and $f(\phi)$ are periodic functions of inclination with the same period $T_f = 2\pi/m$ (see Appendix A). Then, we can write the second term in equation (11):

$$\delta\left\langle \frac{dA_n}{dt} \right\rangle = k\sum_{i-1}^{n}\int_0^{\Theta}\int_0^{\Theta} d\theta_i d\theta_{i+1}\int_0^{2\pi} d\phi g_n^{(i)} (\theta_i,\theta_{i+1},\phi)\left(F\left(\phi + \frac{\pi}{3}-\varepsilon_{i,i+1}\right)-F(\phi)\right), \tag{12}$$

where:

$$g_n^{(i)} = \int_0^{2\pi}\ldots\int_0^{2\pi} d\phi_1^{(in)}\ldots\ldots d\phi_{i-1}^{(in)} d\phi_{i+1}^{(in)}\ldots d\phi_n^{(in)}\int_0^{\Theta}\ldots\int_0^{\Theta} d\theta_1\ldots d\theta_{i-1}d\theta_{i+2}\ldots d\theta_n \times g_n(\theta_1,\ldots,\theta_n,\phi_1^{(in)},\ldots,\phi_n^{(in)}).$$

If we know the vertex distribution g_n and the boundary mobility functions, then using equation (12) we can calculate the nonlinear corrections to the average growth rate of the grains in the topological class of n-sided grains. In general, the vertex distribution is time dependent, $g_n = g_n(\phi,t)$. In a number of cases, however, we may use symmetry arguments to estimate expression (12) without detailed knowledge of the distribution functions.

2.1. Small misorientation limit

Grain growth kinetics in textured materials is an example of an important class of problems, where we can use symmetry arguments to estimate the nonlinear corrections to the growth rate. Textured materials have grains with similar orientations, $\varepsilon_{ij} \ll 1$. If the mobility is a smooth function of inclination and if:

$$\frac{F(\phi + \varepsilon_{ij})-F(\phi)}{F(\phi)} \ll 1, \tag{13}$$

then to leading order in ε_{ij}, the function F is independent of ε_{ij}, $F(\phi + \varepsilon_{ij}) = F(\phi) + o(\varepsilon_{ij})$. For example, if $\max(\varepsilon_{ij}) \leq 20^0$, then a Read–Shockley form for the grain boundary mobility $L(\theta,\phi) \propto \theta(1-\ln(\theta))(|\cos(\phi)| + |\sin(\phi)|)$ is accurate to 10%. From symmetry arguments, the vertex distribution $\tilde{g}_n(\phi)$ is also a periodic function with a period $T_g = 2\pi/m$, and the correction to the growth rate in the case of a textured material is:

$$\delta\left\langle \frac{dA_n}{dt} \right\rangle = \int_0^{2\pi} d\phi\left(\tilde{g}_n\left(\phi + \frac{\pi}{3},t\right) -\tilde{g}_n(\phi,t)\right)F(\phi), \tag{14}$$

where:

$$\tilde{g}_n(\phi) = \sum_{i=1}^{n} g_n^{(i)}(\phi).$$

Let us consider a polycrystalline aggregate with mobility anisotropy function f(ϕ) (see equation (6)). It seems reasonable that possible effects of mobility anisotropy would be to bound grain shapes by low mobility boundaries. The higher the symmetry of the function $f(\phi)$ the closer the shape of the grain should be to the one observed in the isotropic case. Therefore, the most extreme case of shape anisotropy occurs for two-fold inclination symmetry of $f(\phi)$. If we consider such a symmetry for the mobility function in the small misorientation limit, since local coordinates deviate only slightly from the global, more triple junctions will have one of their grain boundaries aligned along the x-axis of the global coordinate system than any other type of triple junctions. We have chosen the local coordinate system so that its x-axis corresponds to the lowest mobility direction, which does not affect our results. Due to the boundary energy isotropy, the angles between the inclinations of the boundaries at the triple junctions are equal to 120^0 and the vertex distribution function $\tilde{g}_n(\phi,t)$ is six-fold with respect to inclination. Thus, in the quasi-stationary regime where the time dependence of $\tilde{g}_n$ does not lead to a change of symmetry, $\tilde{g}_n(\phi,t)=\tilde{g}_n(\phi+\pi/3,t)$ and expression (14) for $\delta\langle(dA_n)/dt\rangle$ is equal to 0. Thus, the corrections to the linear term in equation (11) are vanishingly small and we can write the average growth rate of the grains in the topological class n:

$$\left\langle\frac{dA_n}{dt}\right\rangle = \frac{k\pi}{3}\langle L\rangle_\phi(n-6), \tag{15}$$

analogous to the von Neumann–Mullins expression in the isotropic limit. Our entire analysis is valid only for “quasi-stationary” evolution, i.e. for a fixed symmetry of the (possibly time dependent) inclination distribution function.

In a similar fashion we can show that the result for $\langle dA_n/dt\rangle$ is independent of the particular symmetry of the mobility function, assuming a textured (small misorientation) material with the mobility a smooth function of the inclination. The same result derives from equation (14) when the function F satisfies the relationship $F(\phi+\pi/3)=F(\phi)$, even without assumptions about the influence of mobility on the shape of the grains.

Next, we analyze the consequences of the dependence of grain boundary mobility on both inclination and misorientation. First of all, we note that equation (3) is no longer valid, since $L=L(\theta,\phi)$ and different sides of a grain, in general, have different misorientations, $L(\theta_1,\phi)\neq L(\theta_2,\phi)$; but we can write any periodic function of two variables L,

$$L(\theta,\phi) = \langle L(\theta)\rangle_\phi(1+f(\theta,\phi)), \tag{16}$$

where $f(\theta,\phi)=f(\theta,\phi+2\pi)$. Moreover, since the mobility is a positive definite function, $|f(\theta,\phi)|<1$ (compare with equation (7)). Then, we can express the growth rate of a single n-sided grain in the form:

$$\frac{dA_n}{dt} = -k\sum_{i=1}^{n}\int_{\phi_i^{(in)}}^{\phi_{i+1}^{(in)}} d\phi L(\theta,\phi) + \frac{k\pi}{3}\sum_{i=1}^{n}\langle L(\theta)\rangle_\phi + k\langle L(\theta)\rangle_\phi\sum_{i=1}^{n}\int_{\phi_i^{(in)}}^{\phi_i^{(in)}+\frac{\pi}{3}-\varepsilon_{i,i+1}} f(\theta,\phi)d\phi. \tag{17}$$

To average this expression over a topological class n, as in the previous case, we introduce a vertex distribution function $g_n(\theta_1,\ldots,\theta_n,\phi_1,\ldots,\phi_n)$, which is a function of both inclinations and misorientations of the grain boundaries. We assume this function to be normalized to unity. Taking into account the fact that $g_n(\ldots,\theta_i,\ldots,\theta_j,\ldots)=g_n(\ldots,\theta_j,\ldots,\theta_i,\ldots)$ we deduce that the average growth rate of the grains in the topological class n is:

$$\left\langle\frac{dA_n}{dt}\right\rangle = \frac{k\pi}{3}\langle L\rangle_{\theta,\phi}(n-6) + k\sum_{i=1}^{n}\int_0^{\Theta} d\theta_i d\theta_{i+1}\times\int_0^{2\pi} d\phi\left(g_n^{(i)}\left(\theta_i,\theta_{i+1},\phi+\frac{\pi}{3}-\varepsilon_{i,i+1}\right)-g_n^{(i)}(\theta_i,\theta_{i+1},\phi)\right)F(\theta_i,\phi), \tag{18}$$

where we define the functions $g_n^{(i)}$ and F similarly to the case where mobility is a function of inclination only.

We note a striking similarity between expressions (11) and (18). In both cases we can express the average growth rate of grains in a topological class in terms of the von Neumann–Mullins form [23], plus a term which describes the non-linear corrections to the constant growth rate, which can be estimated in a number of cases based on symmetry arguments.

Analyzing the small misorientation limit in equation (18) as in the case when mobility is a function of inclination only, the growth rate of the grains in the topological class n is to first approximation proportional to $(n-6)$ with corrections of order $\varepsilon_{i,i+1}$. The time dependence of the misorientation distribution function then determines time dependence of the growth rate $\langle dA_n/dt\rangle$ through $\langle L\rangle_{\theta,\phi} = \int d\theta g_n(\theta)\langle L(\theta)\rangle_{\phi}$. We can express any periodic function of two variables $g_n(\theta,\phi)$ in terms of the Fourier series:

$$g_n(\theta,\phi) = a_0(\theta) + \sum_{m=1}^{\infty} a'_m(\theta)\cos(m\phi) + a''_m(\theta)\sin(m\phi) \tag{19}$$

We determine periodicity of $g_n(\theta,\phi)$ from m_0, which is the greatest common divisor of all the values of m in the expansion (19) such that $a_m\neq 0$. Integrating equation (19) with respect to θ eliminate some of the coefficients a_m and, thus, reduce the period of $g_n(\phi)$ compared to $g_n(\theta, \phi)$. However, in the small misorientation limit, we can always choose a small interval around $\theta = 0$, where the $a_m(\theta)$ have fixed sign and the integration retains in $g_n(\phi)$ all modes present in $g_n(\theta, \phi)$. Then the period of both functions will be identical and the rest of the analysis is analogous to the case where mobility depends only on the inclination.

2.2. Fully random and intermediate cases

Another important case where we can estimate the non-linear contribution $\delta\langle dA/dt\rangle$ to the average growth rate for grains in a given topological class is the limit when grains are randomly distributed with all possible misorientations, e.g. crystals with four-fold symmetry randomly oriented in the range $[0,\pi/2]$. In this case all distribution functions are constant, including the vertex distribution function and the misorientation distribution function (giving the fraction of grain boundaries with a given misorientation). In other words, the distribution functions are periodic with period $T = 0$. If the evolution preserves the randomness as confirmed by means of computer simulations (see Section 3), then for any value of δ, $g_n(\theta,\phi + \delta) = g_n(\theta,\phi)$, and the nonlinear time-dependent corrections to the average growth rate of the grains in a topological class n vanish, independent of whether the mobility is a function of inclination only or of both inclination and misorientation. Then $\langle dA_n/dt\rangle$ is constant and, as in the textured case, is of von Neumann–Mullins type (equation (15)).

Introducing $\max(\Delta g)/g_0$, where $\Delta g = g_n(\theta,\phi + \delta)-g_n(\theta,\phi)$ and $g_0 = \langle g_n(\theta,\phi)\rangle_{\theta,\phi}$, which characterizes deviations of the vertex distribution function from its constant value in the fully random case, we can estimate the non-linear corrections to the average growth rate of the grains caused by deviations from the fully random state, due either to the initial configuration or subsequent evolution.

In the intermediate region, in contrast to the limiting cases of textured and fully random microstructures, the analysis is more complicated. The symmetry arguments no longer apply since $\varepsilon_{i,j}\sim 1$ and, generally speaking, condition (13) fails. However, we can estimate the influence of mobility anisotropy on the kinetics and the deviation of the average growth rate of grains in a topological class from von Neumann–Mullins behavior from knowledge of the vertex distribution function. In this limit the only property entering the analysis is the value of $\max(\Delta g)/g_0$. If $\max(\Delta g)/g_0$ is small compared to unity, then the mobility anisotropy has a small influence on the evolution and the growth rate in the entire range of misorientations should follow the time independent von Neumann–Mullins law. On the other hand, if the mobility anisotropy has a pronounced effect on the microstructural evolution during grain growth, then variation in the vertex distribution is no longer negligible, resulting in a time-dependent average growth rate $\langle dA_n/dt\rangle$. We evaluate inclinations in the vertex distribution function in local coordinates ("local vertex distribution") and thus the angles between the boundary inclinations, defined in the local coordinate system at a triple junction, are no longer equal to 120^0 in the global system complicating the analysis. However, as shown in Appendix B, we can calculate the local vertex distribution from the global vertex distribution without calculating it directly from the microstructure.

On the other hand, in the limit that the mobility is a function only of misorientation, as expected for large misorientations [25–27], the analysis presented above is trivial and, in the case of a time independent misorientation distribution, the expression for the average growth rate has the following form independently of the range of misorientations:

$$\left\langle \frac{dA_n}{dt} \right\rangle = \frac{k\pi}{3}\langle L\rangle_\theta(n-6), \tag{20}$$

where $\langle L\rangle_\theta = \int d\theta g_n(\theta)L(\theta)$ is the average mobility of grain boundaries in the polycrystalline aggregate. Again, any time dependence of $\langle dA_n/dt\rangle$ results only from the misorientation distribution function $g_n(\theta,t)$.

2.3. Growth rate of the average area

The analysis is less trivial if we try to estimate the growth rate of the average grain area, $d\langle A\rangle/dt$, where $\langle A\rangle = A_{tot}/N_{tot}$ is the average area of a grain in a polycrystalline aggregate with total area A_{tot} containing N_{tot} grains. Following the same line of reasoning as Mullins [12], the growth rate of the average grain is:

$$\frac{d\langle A\rangle}{dt} = \frac{2}{A_{tot}}\frac{\langle A\rangle^2}{\langle A^2\rangle}\sum_{i=1}^{N_{tot}} A_i\frac{dA_i}{dt}, \tag{21}$$

assuming the value of $(\langle A\rangle^2)/(\langle A^2\rangle)$ is time independent. Physically, this ratio characterizes the half-width of the size distribution $f(A/\langle A\rangle)$, which describes the fraction of grains with area A normalized by the average area of the grains. From the self-similarity hypothesis, in the isotropic case this ratio is always time independent [9]. If we assume that during grain growth with anisotropic grain boundary mobility, the half-width of the size distribution does not significantly change with time, then equation (21) hold up to corrections of the order of $\delta(\langle A\rangle^2/\langle A^2\rangle)$, which characterize deviations of the half-width from its average value in the quasi-stationary regime. This assumption is reasonable and has been verified in computer simulations of grain growth in anisotropic materials [11,18].

Using the above results for the average growth rate of the grains in a given topological class, we can rewrite equation (21):

$$\frac{d\langle A\rangle}{dt} = 2\frac{\langle A\rangle^2}{\langle A^2\rangle}\sum_{n=2}^{\infty} p_n\left\langle\frac{dA_n}{dt}\right\rangle, \tag{22}$$

where p_n is the fraction of the area occupied by grains of the topological class n and the sum is over different topological classes. Finally, if the distribution of p_n is time independent (which we can verify by computer simulation) the growth rate of the average grain in a polycrystalline aggregate is a constant, i.e. $\langle A\rangle$ grows linearly with time.

3. Computer simulation results

To test our theoretical predictions we have simulated grain growth with anisotropic grain boundary mobility using the Phase Field approach, described in [24]. We present detailed analysis of the simulation results, including a variety of statistical properties elsewhere [11]. Here we focus on the growth kinetics of grains in a given topological class as well as on the growth rate of the average grain area (Fig. 2).

In the simulations, we assumed a boundary mobility:

$$L(\theta,\phi) = L_1(\theta)(1-\delta_L\cos(2\phi)), \tag{23}$$

where δ_L is a phenomenological parameter controlling the degree of anisotropy. In the small misorientation limit, we have tested several functional dependencies for $L_1(\theta)$ ranging from a rapidly

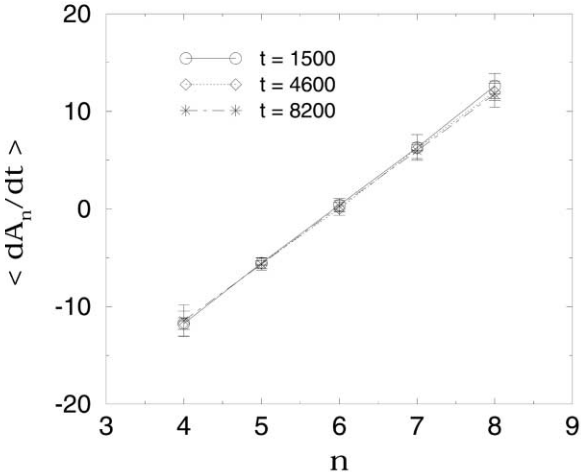

Fig. 2. Average growth rate of the grains in different topological classes as a function of the topological class. The data is averaged over three simulations.

growing $L_1 \sim \theta^5$, recently obtained by Huang and Humphreys [28] from experimental studies of subgrain growth in aluminum, to a slowly growing $L_1 \sim \theta(1-\ln(\theta))$, suggested by Read and Shockley to describe the misorientation dependence of the grain boundary energy in cubic crystals. However, the simulations show that growth kinetics is qualitatively the same in all cases considered and does not depend on the particular form of L_1. In the intermediate and fully random cases, the periodicity of the grain boundary mobility with misorientation requires a mobility of the form, $L(\theta,\phi) = L_0\sin(2\theta)(1-\delta_L\cos(2\phi))$. In all simulations we have taken the phenomenological parameter δ_L to be 0.9, which corresponds to 20 times the difference between the largest and the smallest boundary mobility for a fixed misorientation.

Using the simulations, we investigate the behavior of the vertex distribution function characterizing the fraction of boundaries at a triple junction with a particular inclination. When the grain boundary mobility function has a two-fold symmetry with respect to inclination and the boundary energy is isotropic, we argued that the vertex distribution should have six-fold symmetry with respect to inclination. Then, non-linear and time dependent corrections to the average growth rate vanish in the first approximation. Fig. 3 presents results obtained from the computer simulations which indicate, as expected, that the distribution has six distinct peaks at the angles $\phi = \pi n/3$. In addition, the vertex distribution function is time-dependent. The simulations started from initial isotropic microstructures with uniform vertex distributions. As the shape anisotropy develops, the peaks in the vertex distribution increase in magnitude without changing the symmetry of the distribution. In addition, the misorientation distribution function is time independent [11]. As long as the evolution takes place in such a "quasi-stationary" manner, our theoretical analysis remains valid and the average growth rate of the grains in a topological class n should be time independent.

Indeed, in Fig. 2, within the statistical error of the simulations, $\langle dA_n/dt \rangle$ is constant, in contrast to the time dependent growth rate of a single grain with anisotropic boundary mobility [11]. Moreover, in agreement with our theoretical predictions, $\langle dA_n/dt \rangle$ is a linear function of the topological class number n which vanishes for $n = 6$ (see equation (15)).

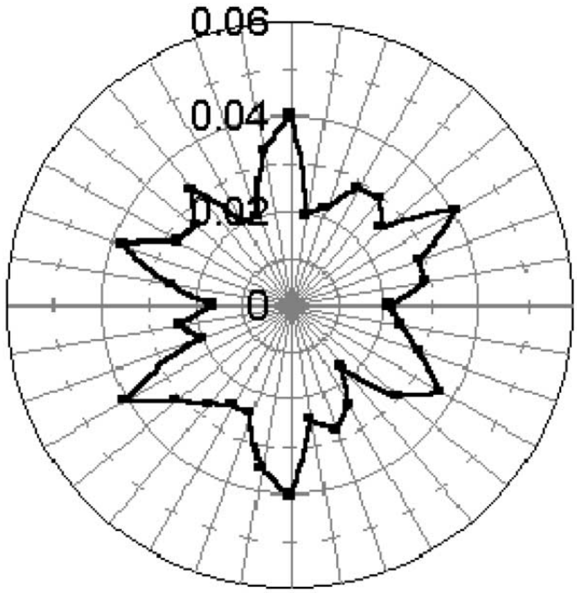

Fig. 3. Vertex distribution function (fraction of the grain boundaries at the triple junction with a particular inclination) extracted from computer simulations. The boundary mobility has been taken to be two-fold with respect to inclination. The inclination is measured in the local coordinate system, i.e. the system associated with the grain boundary. On the graph, angles are measured counterclockwise from the vertical axis.

Finally we consider the behavior of the average area growth rate for the full range of possible misorientations. Figure 4 presents the time dependence of the average grain area in two different systems: (a) a textured or small misorientation system with maximum grain rotation angle $(\alpha_{rot})_{\max} = 5^0$ (open circles), and (b) a system with an intermediate range of misorientations where $(\alpha_{rot})_{\max} = 45^0$ (solid squares). To reduce the statistical errors, both simulations were performed in a 1024×1024 system. The results for the fully random case are identical to that of the textured system and therefore are not shown separately. Following an initial transient ($150<t<800$) after the misorientation and inclination dependence of the grain boundary mobility are imposed, the average grain area in case (a) increases linearly with time, as expected from our theoretical analysis. In case (b) the

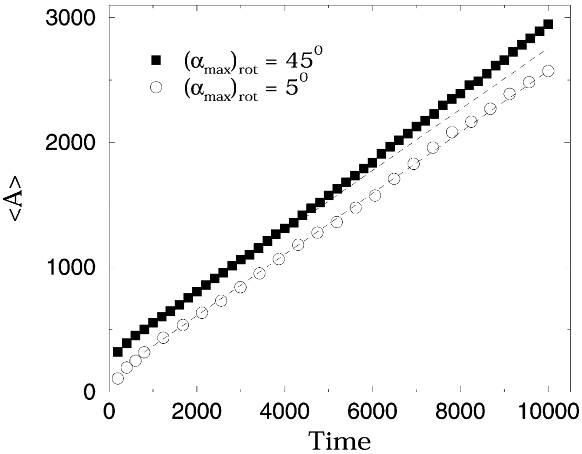

Fig. 4. Time dependence of the average grain area in the systems with (a) small grain misorientations, $(\alpha_{rot})_{max} = 5^0$ (open circles) and (b) intermediate range of misorientations, $(\alpha_{rot})_{max} = 45^0$ (solid squares). For the purpose of comparison, the slope of the textured case is normalized to be equal to the initial slope in the intemediate case (dashed line).

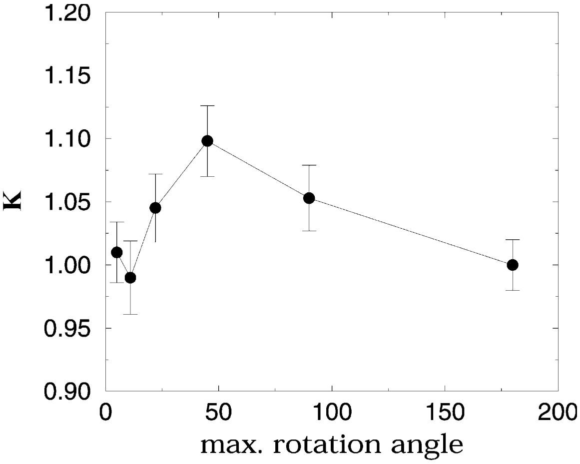

Fig. 5. Dependence of the growth exponent $k(\langle A\rangle \propto t^k)$ as a function of the maximum rotation angle of the grains $(\alpha_{rot})_{max}$ in the polycrystalline system.

growth is clearly nonlinear after the same initial transient, and can be described by the relation $\langle A\rangle^{1/k} - \langle A\rangle_0^{1/k} \propto t$ with $k = 1.086$. For the purpose of comparison the linear slope of the textured case has been normalized to be equal to the initial slope of the intermediate case (dashed line). For better visualization, the curve in the textured case is also shifted downwards along the ordinate axis. The growth exponent is determined by fitting the data in the range of times from $t_1 = 800$, which excludes the initial transient, to $t_2 \propto 10000$, where statistical effects due to decreasing number of grains ($N_{gr} \propto 300$) start to influence the kinetics of the system. Error bars estimated from the curve fits as well as repeated runs are given by the size of the data points. Figure 5 shows the dependence of the growth exponent k on $(\alpha_{rot})_{max}$, where $(\alpha_{rot})_{max} = 180^0$ corresponds to the fully random case. Maximum deviation of the growth exponent from unity occurs for intermediate range of misorientations, as predicted by our theoretical analysis. Although the size of the effect is small, of order 10 percent, the deviation from unity is clearly outside the error bars in Fig. 5, which are determined by the standard errors of the coefficients during the curve fitting. Therefore, the above analysis suggests that in the presence of mobility anisotropy the average grain area may, but does not have to, grow linearly with time, while in the textured and fully random cases self-similar microstructural evolution is not necessary for linear growth kinetics.

4. Conclusions

We studied analytically the dynamics of coarsening during grain growth in systems with anisotropic grain boundary mobility, generalizing the arguments of Mullins in the isotropic case. We analyzed the consequences of removing the self-similarity assumption, which strongly constrains microstructural evolution. We showed that the average growth rate of grains in the topological class of n-sided grains can be represented as a sum of two terms, where the first term has the well-known von Neumann–Mullins form, in which the grain boundary mobility is averaged over the entire polycrystalline aggregate, with a time dependence solely characterized by the evolution of the misorientation distribution function. The second term is, in general, time dependent and contains all information about anisotropies. In the textured and fully random limits, we showed using symmetry arguments that corrections to the von Neumann–Mullins law are negligible, implying isotropic-like evolution. From comparison with computer simulations we find that the corrections indeed vanish in the textured and fully random limits, even though evolution of the microstructure is no longer self-similar. Calculations of the growth rate of the average grain area show that the average area $\langle A \rangle$ grows linearly with time in the two extremes of textured and fully random grains. However, in the intermediate regime the growth exponent deviates from unity by up to 10 percent. Thus, self-similarity may not be a general feature of anisotropic systems and is not always necessary for linear growth kinetics.

Acknowledgements

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Appendix A

Here we show that if $f(\phi)$ is periodic function with a period $T_f = 2\pi/n_0$ where n_0 is a positive integer and $\langle f(\phi) \rangle_\phi = 0$, then its first indefinite integral $F(\phi) = \int d\phi f(\phi)$ has the same period as $f(\phi)$. Indeed, since we can expand any periodic function $f(\phi)$ as a Fourier series:

$$f(\phi) = \sum_{n=1}^{\infty} a_n \cos(n\phi) + b_n \sin(n\phi),$$

the value of n_0 which is the greatest common divisor for the values of n entering the expansion determines periodicity of this function. Integration with respect to ϕ, however, results only in the interchange $a_n \leftrightarrow -b_n$, which does not influence the structure of the expansion. As a result, the value of n_0 is unchanged after the integration and both $f(\phi)$ and $F(\phi)$ have the same period, T_f.

Appendix B

In this appendix we show how to compute the vertex distribution as a function of the angles calculated in the local coordinate system (the "local vertex distribution") based on the knowledge of the global vertex distribution, where all grain boundary inclinations are determined in the global system. We introduce the following notation: $f_l(\phi)$ is the local vertex distribution, $f_g(\varphi)$ is the global vertex distribution, and $n(\psi)$ is the distribution of the local coordinate systems as a function of their rotation with respect to the global system. Then, by definition:

$$f_g(\phi) = \int d\psi f_l(\phi + \psi) n(\psi). \tag{B1}$$

Fourier transforming this relationship becomes:

$$\tilde{f}_g(p) = 2\pi \tilde{f}_l(p) \tilde{n}(-p) \tag{B2}$$

where the quantities with *tilde* are the Fourier transforms. Solving this equation for $\tilde{f}_l(p)$ and transforming back to real space gives the expression for the local distribution function.

For example, when the triple junctions are randomly distributed in space (isotropic-like microstructure) i.e. $f_g(\varphi) = const$, then $\tilde{f}_g(p) \propto \delta(p)$. If $n(\psi)$ is an arbitrary distribution function, equation (B2) immediately implies that $\tilde{f}_l(p) \propto \delta(p)$ and $f_l(\phi) = const$. Thus, a random distribution of vertices in the global coordinates remains after transformation to local coordinates.

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