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Effect of grain boundary faceting on kinetics of grain growth and microstructure evolution

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The Von-Neumann-Mullins relationship for two-dimensional grain growth is modified for the case of grain boundary faceting. It is shown that the anisotropy of grain boundary energy alone slows down the rate of normal grain growth. For highly mobile facets, however, the acceleration of the growth process is possible, accompanied by development of anisotropic microstructure. It is shown that the mean-field approach to the problem of grain growth in highly anisotropic polycrystal results in parabolic growth law similar to that for isotropic systems, with the facet mobility and maximal torque substituting the grain boundary mobility and grain boundary energy in isotropic systems.

1. Introduction

Improving the properties of polycrystal by increasing the fraction of coincidence site lattice (CSL) grain boundaries (GBs) is the quintessence of the concept of Grain Boundary Engineering [1]. The conventional wisdom says that the CSL GBs exhibit higher corrosion resistance and fracture energy and lower diffusivity than their random counterparts and, therefore, increasing the fraction of CSL GBs improves mechanical properties and corrosion resistance of polycrystal. This concept gained momentum with the development of orientation image microscopy (OIM) technique that allows to characterize easily the misorientational degrees of freedom (DOFs) of thousands of GBs and to determine the fraction of different CSL GBs in polycrystal.

However, this approach suffers from one serious drawback: while three misorientational DOFs determine completely the type of CSL GB, a considerable variation of GB properties can be caused by remaining two inclinational DOFs. The latter are difficult to determine experimentally, and there are only few works in which the GB properties are correlated with all five geometric DOFs [2, 3]. The importance of the inclinational DOFs can be demonstrated on example of the $\Sigma 3$ (here Σ is a reciprocal density of coincident lattice sites) CSL GB in Cu: while the symmetric coherent twin boundary exhibits particularly low energy and no segregation of impurities, symmetrical incoherent twin that can be obtained from the coherent one by 90 deg rotation around $\langle 011 \rangle$ axis behaves similarly to the random non-CSL GBs [4]. Strong inclinational anisotropy of the GBs leads to the phenomenon of GB faceting. There are numerous observations in the literature showing that the CSL GBs are particularly prone to faceting [5]. It is the GB faceting that is responsible for the characteristic shape of $\Sigma 3$ twins in Cu.

An essential element of any GB engineering process is the grain growth after recrystallization. Indeed, the computer simulations confirm that the fraction of low energy, low mobility GBs considerably increases in the course of grain growth [6]. The aim of the present work is to investigate the effect of GB faceting on grain growth and evolution of the population of the anisotropic GBs in polycrystals.

In the recent computer simulation study based on the phase field model [7] the dependencies of both GB energy and mobility on misorientation and inclination angles were taken into account. It was shown that, in contrast with the expectations based on the common sense, the anisotropy of GB mobility has little effect on parameters of grain growth process, while the anisotropy of GB energy can lead to substantial deviations from the parabolic growth behavior and random misorientation distribution characteristic for isotropic systems. However, the phase field model with a weak anisotropy considered in [7] cannot account for the sharp edges and flat facets of GBs observed in a number of experimental works. The observations of Yoon and co-workers [8–10] indicate that such a faceting may play a crucial role in normal grain growth and in nucleation of abnormal grain growth that leads to survival of anisotropic faceted GBs in the final microstructure. In the present work we will base our analysis on the assumption of strong inclinational anisotropy of the GB energy that ultimately leads to faceting.

2. Von Neumann-Mullins relationship for faceted GBs

Von Neumann-Mullins relationship for 2-D grain growth represents a remarkable exact result that expresses the rate of area change of an individual grain as

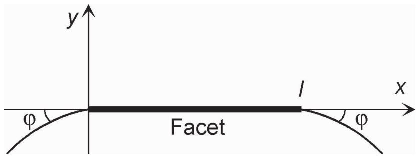

Figure 1 Geometry of a facet (bold line) connected with non-singular GBs at sharp edges.

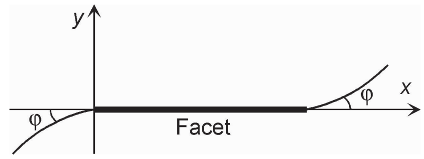

Figure 2 A facet that does not contribute to grain growth.

a function of the number of grain sides [6]. It served as a starting point for numerous computer simulations of 2-D grain growth [11]. In what follows we will consider how the GB faceting modifies Von Neumann-Mullins relationship.

Let us consider a faceted, flat GB that is joined with two non-singular GBs of identical isotropic energy γ_b at the sharp edges (Fig. 1). Such GB shape corresponds to the sharp narrow cusp at facet inclination in otherwise spherically symmetrical GB γ-plot. The relationship between γ_b and the facet energy, γ_o, can be obtained by considering a small shift of the sharp edge in the x-direction, which should not change the total interfacial energy of equilibrated system:

$$\gamma_0 = \gamma_b \cos\varphi \tag{1}$$

where φ is the angle at which the non-singular GB joins the facet at the sharp edge. At the first glance, since the flat facet does not exhibit any mathematical curvature, the driving force for facet migration should be zero and it should not contribute to the shrinking or growing of grains. However, as was discussed by Herring [12] and later by Taylor [13] the relevant quantity for microstructure evolution of faceted structures is not mathematical, but weighted mean curvature. The latter is defined as the negative of change of total interfacial energy due to the small facet displacement, divided by the volume swept by the facet. According to this definition, the weighted mean curvature, *WMC*, (and, hence, the driving force for facet migration) for the facet in Fig. 1 is

$$WMC_1 = \frac{2\gamma_b \sin\varphi}{l} \tag{2}$$

where l is the facet length. Since the migration rate of the facet is its mobility, M_0, multiplied by the driving force, WMC_1, the contribution of the facet to the rate of grain shrinking/growth (in geometry of Fig. 1) is:

$$\left(\frac{dS}{dt}\right)_f = -2\gamma_b M_0 \sin\varphi \tag{3}$$

where S is the area of the grain bounded by the facet in Fig. 1 and t is the annealing time. To proceed with our model we need further simplifications. We will assume that for all grains with the number of sides $n < 6$ all facets are of the type shown in Fig. 1, i.e., they contribute only to the grain shrinking. For the large grains with $n > 6$ we will assume that the non-singular GBs are concave, i.e., their centers of curvature lie outside the grain. In this situation facet migration contributes to the grain growth. At this point we will disregard the possibility that the facet joins the non-singular GBs or other facets at the triple junction. This should be a good approximation for short facets and for $\varphi \ll \pi/3$. For all situations we will discard the facets of the geometry shown in Fig. 2, since their weighted mean curvature is zero and they do not contribute to grain growth anyway. We will also consider the situation of maximum one facet for each grain side. The generalization for the more realistic case of multiply faceting is straightforward, but in this paper we would like to demonstrate the main effects of faceting for the simplest possible situation. Another assumption is that the number of singular GB orientations, X, is a constant for the given grain. Strictly speaking this is not the case since in the process of grain growth the grain under consideration changes its neighbors and, consequently, the symmetry of corresponding GBs also changes. However, there is some evidence that the GB which is parallel to a low-index plane in just one of two neighboring grains exhibits local cusp in energy and is singular. This fact provides some justification to the assumption $X = \text{const}$, since the orientations of the grains do not change in the course of grain growth for sufficiently large grains.

Let us first consider the situation $n < 6$. The angular range of inclinations swept by all n sides of the grain is $2\pi - n\pi/3$. Here we assumed that all the GBs meeting at the triple junctions are the non-singular ones and have equal energy of γ_b. The probability that all X singular orientations will fall inside this interval is $X(1 - n/6)$. This is also the number of facets for the given grain. We can now write down the expression for the rate of grain area change:

$$\frac{dS}{dt} = -\oint K\gamma_b M ds - 2X\left(1 - \frac{n}{6}\right)\gamma_b M_0 \sin\varphi \tag{4}$$

where ds is the element of length along the GBs, K is the GB curvature and M is the mobility of non-singular GBs. The first and the second term in the RHS of Equation 4 represent the contributions of the non-singular GBs and of the facets, respectively, to the shrinking of the grain. The loop integral in the RHS of Equation 4 would be $2\pi\gamma_b M$ for the circular grain embedded in the single crystalline matrix. For the real grain the result of loop integration of the curvature will be reduces by $\pi/3$ for every triple junction and by 2φ for every facet. Taking this into account we arrive at the main result of this section:

$$\frac{dS}{dt} = \frac{M\gamma_b\pi}{3}(n-6)\left\{1 - \frac{36X}{\pi}\left(\varphi - \frac{M_0}{M}\sin\varphi\right)\right\} \tag{5}$$

The identical result can be obtained also for $n > 6$. Equation 5 demonstrates that with the simplifications made the role of GB anisotropy in modifying the Von Neumann-Mullins relationship is reduced merely to the renormalization of GB mobility:

$$M' \to M\left\{1 - \frac{36X}{\pi}\left(\varphi - \frac{M_0}{M}\sin\varphi\right)\right\} \quad (6)$$

It is instructive to consider first the case of identical mobilities $M = M_0$. Since the inequality $\varphi > \sin\varphi$ is valid for all non-zero values of φ we come to the conclusion that in this case the GB anisotropy always reduces the rate of normal grain growth. Developing $\sin\varphi$ in Equation 6 and $\cos\varphi$ in Equation 1 in Taylor series and keeping terms up to the third order in φ we arrive at the approximate result for renormalized mobility:

$$M' \approx M\left\{1 - 5.4X\left(1 - \frac{\gamma_0}{\gamma_b}\right)^{3/2}\right\} \quad (7)$$

Equation 7 demonstrates that the effect of GB anisotropy on slowing down the process of grain growth is relatively strong: for the two-fold symmetry ($X = 2$) and mere 10% anisotropy in GB energy ($\gamma_0 = 0.9\gamma_b$) the renormalized GB mobility is reduced by approx. 34%. For larger values of GB anisotropy M' can be further reduced or even turned negative, however, at such large anisotropies the assumptions made during derivation of Equation 5 are not valid anymore.

The above results are consistent with the results of phase field simulations of Kazaryan *et al.* [7], where the slowing down of the rate of normal grain growth resulting from the energy anisotropy alone was found. However, no direct comparison between the results of two works is possible. In Ref. [7] the GB energy was the function of both GB misorientation and inclination angles. The slowing down of grain growth in [7] can be associated with the increase of the fraction of low-angle, low-energy GBs in the course of growth. In the present work, the GB energy depends on the GB inclination only, and the cusps on γ-plot associated with the singular orientation are assumed to be narrow and sharp. Therefore, in the present work the effect of only inclinational GB anisotropy on grain growth is emphasized. It is remarkable that this inclinational energy anisotropy alone slows down the rate of grain growth.

We have made an assumption $M = M_0$ for illustrative purpose only since there is little physical justification for assuming that the mobility of singular GB coincides exactly with the mobility of its random, isotropic counterpart. Indeed, while it is commonly agreed that the migration of random isotropic GB is realized by uncorrelated jumps of individual atoms across the GB, the migration mechanism of a singular facet can be totally different [14]. For example, the hot stage *in-situ* transmission electron microscopy (TEM) study of the migration of $\Sigma5$ singular GB in gold has shown that its migration occurs by the shuffling of groups of atoms directly across the GB [15]. The difference in migration mechanisms causes the difference in mobilities that can reach one-two orders of magnitude [14]. While the situation $M_0 < M$ will just lead to further decrease of the rate of normal grain growth (see Equation 5), the opposite situation $M_0 > M$ can result in *increased grain growth rate due to faceting*. It should be noted that Equation 5 describes the time evolution of the area of the grain and does not tell anything about the grain shape. It is clear that the presence of highly mobile facets will result not only in acceleration of the overall rate of grain growth, but also in anisotropic microstructure: the grains will be elongated in the direction of fast facets. Such microstructures are often observed during the grain growth in ceramics [16, 17].

One of the most important assumptions made during the derivation of Equation 5 was that the facets are sufficiently short and do not reach the triple junctions of the grains. In the next section we will show that with increasing M_0 the length of the facets increases, so that at some point they inevitably reach the triple junctions. In this situation the extension of Von Neumann-Mullins relationship (Equation 5) will loose its validity. In what follows we will develop an alternative approach based on mean field approximation of Burke and Turnbull [6].

3. Steady state migration of the faceted GB

Let us consider the steady-state migration of GB half-loop that exhibits a singular facet for the inclination which is parallel to the y-axis (Fig. 3). In the steady-state regime both the non-singular portion of the GB and the facet move as a whole in the x-direction with a unique migration rate V and the overall shape of the GB does not depend on time. Though the GB migration during grain growth is never steady state, the geometry of half-loop is often employed for the experimental studies of capillary-driven GB migration [14] and, therefore, it is instructive to consider how the difference in mobilities of the non-singular GB and the facet affects the geometry of the half-loop. The equations governing the migration of the non-singular GB have been derived elsewhere [14] and here we will write down the general solution for the shape of this portion of the half-loop:

$$y = y_0 + \frac{M\gamma_b}{V}\arccos\left\{\exp\left(-\frac{(x - x_0)V}{M\gamma_b}\right)\right\} \quad (8)$$

where x_0 and y_0 are the integration constants. These constants, together with the values of V and facet length l can be determined from the obvious boundary conditions:

$$y(0) = \frac{l}{2} \quad (9)$$

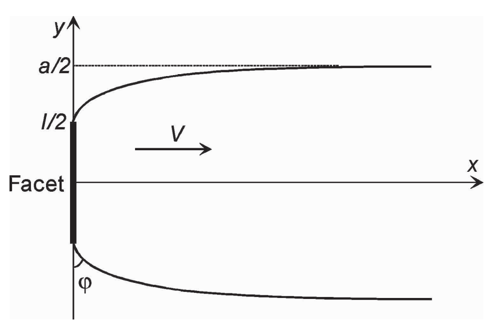

Figure 3 Geometry of capillary-driven migration of the faceted GB.

$$y(\infty) = \frac{a}{2} \tag{10}$$

and

$$y'(0) = \frac{1}{\tan\varphi} \tag{11}$$

The last condition can be obtained using the same weighted mean curvature arguments for migration of the facet that led to Equations 2 and 3:

$$V = \frac{2\gamma_b M_0 \sin\varphi}{l} \tag{12}$$

Equations 8–12 define the geometry of the half-loop completely. After some algebra we obtain:

$$V = \frac{\gamma_b M}{a}\left\{2\frac{M_0}{M}\sin\varphi - 2\varphi + \pi\right\} \tag{13}$$

and

$$l = \frac{a}{1 + \frac{\frac{\pi}{2} - \varphi}{\frac{M_0}{M}\sin\varphi}} \tag{14}$$

The dependence of the dimensionless facet length, l/a, on the ratio of mobilities, M_0/M, for several different values of φ is shown in Fig. 4. It is remarkable that even for a very weak anisotropy ($\varphi = 0.15$ corresponds, according to Equation 1, to a mere 1.1% difference in energies) the length of the facet rapidly increases with increasing mobility and reaches 50% of the half-loop width already for $M_0/M \approx 10$. Fig. 4 expresses the main message of this section for the analysis of grain growth in 2-D polycrystals: sufficiently mobile facets are so long that they will probably extend up to the closest triple junctions and form the entire grain sides. In this case the analysis of Section 2 looses its validity. The GBs in such situation do not exhibit any mathematical curvature and conventional wisdom says that the grain growth should stagnate. In the next section we will demonstrate that this is not the case.

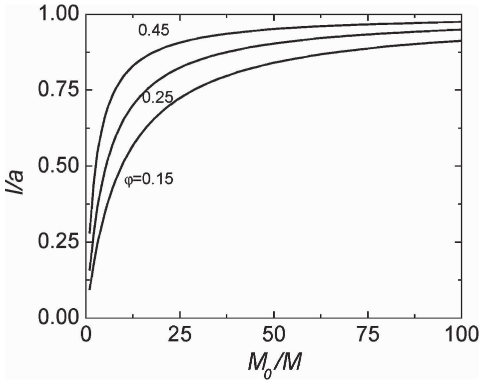

Figure 4 The dependence of dimensionless facet length on the reduced facet mobility for three different values of φ.

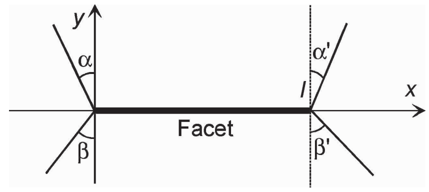

Figure 5 Geometry of a facet (bold line) joining four non-singular GBs at two triple junctions.

4. Grain growth in highly anisotropic polycrystals

Let us consider a singular facet joined by four non-singular isotropic GBs at two opposite triple junctions (see Fig. 5). It follows from the vector ξ-thermodynamics of Cahn and Hofman [18, 19] that the normal force acting on the facet is a sum of two torques at the triple junctions. This normal force causes the migration of the facet and the growth/shrinkage of respective grains. We will give below a more detailed derivation of the fact that the torques at the triple junction represent a sole driving force for facet migration. If the non-singular GBs joining the facet are isotropic the conditions of equilibrium analogous to Equation 1 can be written in the form:

$$\gamma_0 = \gamma_b(\sin\alpha + \sin\beta) \tag{15a}$$

$$\gamma_0 = \gamma_b(\sin\alpha' + \sin\beta') \tag{15b}$$

The torques acting on a facet in the direction of y-axis are:

$$T = \gamma_b(\cos\alpha - \cos\beta) \tag{16a}$$

$$T' = \gamma_b(\cos\alpha' - \cos\beta') \tag{16b}$$

The weighted mean curvature of the facet, WMC_5, which in turn is the driving force for facet migration can be defined using a small displacement of the facet along the y-axis:

$$WMC_5 = \frac{T + T'}{l} \tag{17}$$

Both T and T' vary from zero to some maximal value T_{max}. T_{max} is the maximal torque that can be sustained by the facet without spontaneous bowing out. During the grain growth T and T' may be of the opposite signs and partly compensate each other. We will assume that all facets can be characterized by one value of T_{max} and that the total torque can be written in the form δT_{max}, where δ is a numerical coefficient of the order of one. In the spirit of Burke-Turnbull theory of normal grain growth we will assume that the averaged grain diameter scales with the averaged length of the facets, $\bar{l}$, that fully or partly bound the grains:

$$\frac{d\bar{l}}{dt} = M_0\frac{\delta T_{max}}{\bar{l}} \tag{18}$$

This equation has an obvious parabolic solution

$$\bar{l}^2 - \bar{l}_0^2 = 2M_0\delta T_{max}t \tag{19}$$

where $\bar{l}_0$ is the averaged grain diameter for $t = 0$. Equation 19 is strikingly similar to the classical parabolic growth law for the system of isotropic GBs, with the isotropic GB mobility substituted by M_0 and the isotropic GB energy substituted by T_{max}. The amplitude of T_{max} depends both on the depth and width of the cusp on the γ-plot of respective GB and its amplitude may be comparable with γ_b. The value of M_0 may be both lower and higher than the mobility of non-singular GBs, M. For example, the coherent $\Sigma 3$ twin GBs are known to exhibit very low mobility, whereas the mobilities of singular facets of the CSL GBs with the relatively high energy may be much higher than the mobility of their non-singular counterparts [6]. Therefore, contrary to the conventional wisdom, the rate of normal grain growth in the ensemble of fully faceted GBs can be both lower and higher than the rate of grain growth in isotropic system.

5. Conclusions

From the results of the present work the following conclusions can be drawn:

1. The GB faceting that is caused by the presence of sharp cusps on GB energy vs. GB orientation plot slows down the rate of normal grain growth. Under some simplifying assumptions (i.e., that the GB facets do not reach triple junctions) the faceting modifies Von Neumann-Mullins relationship for 2-D grain growth by mere renormalization of GB mobility. In the approximation of equal mobilities and of weak energy anisotropy, the reduction of the "effective" GB mobility is proportional to the difference of energies of the non-singular GB and the facet power 3/2.

2. If the mobility of the facet is higher than the mobility of non-singular GBs, the grain growth can be accelerated and the anisotropic microstructure should develop.

3. The problem of capillary-driven migration of faceted GB in the half-loop geometry was solved. It was shown that the length of the facet increases with increasing mobility. It was concluded that in the polycrystal highly mobile facets should directly connect the neighbouring triple junctions, thus invalidating the Von Neumann-Mullins relationship.

4. An analog of Burke-Turnbull analysis of normal grain growth for the highly anisotropic microstructure was suggested. The parabolic grain growth law followed, with the maximal torque and the facet mobility substituting the GB energy and the GB mobility, respectively, in the parabolic growth law for isotropic system. Depending on the ratio between the torque and the facet mobility the faceting can both slow down and accelerate the process of normal grain growth.

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