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Grain boundary migration: misorientation dependence

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Abstract

The ability of grain boundaries (GB) to move has been found to be strongly dependent on crystallography, i.e. misorientation of the adjacent grains and orientation (inclination) of the GB in a crystal. Boundary mobility is rate-controlling in recrystallization and grain growth and thus, affects microstructure evolution and texture formation. This paper deals with recent advances in our understanding of misorientation and inclination dependence of grain boundary migration.

A most important peculiarity of grain boundaries is their ability to move. This grain boundary (GB) property has been found to be strongly dependent on grain boundary crystallography, i.e. misorientation of the adjacent grains and orientation (inclination) of the GB in a crystal. Boundary mobility is rate-controlling in recrystallization and grain growth and thus, affects microstructure evolution and texture formation. Recent achievements in our understanding of misorientation and inclination dependence of grain boundary migration constitute the subject of this paper.

The mobility m_b is a quantitative measure of the kinetic properties of a grain boundary and thus, the principal parameter of the process of GB migration. It is defined as GB velocity v per unit of driving force p:

$$m_b = \frac{v}{p}$$

A driving force for GB migration arises when a boundary displacement leads to a reduction of the total energy of the system. It is necessary to stress that the system need not be limited to adjacent grains and a GB only, but may include external elastic, electrical or magnetic fields as well. There are two ways by which this driving force arises. The first uses the free energy of a GB itself, the other utilizes a free energy difference of the adjacent grains ('the system'), or the work expended by an external field.

The great majority of experimental measurements of GB mobility of single GBs was carried out by the so-called capillary driven motion technique, in which a curved GB moves under the action of GB curvature, and the driving force p is provided by the GB surface tension γ. Since the true value of γ is commonly not known, a reduced GB mobility $A_b = m_b\gamma$ is usually used. The dimension of A_b is $[m^2/s]$, i.e. the same as the diffusion coefficient. An inherent feature of GB mobility is that it depends, apart from the conventional thermodynamic variables (temperature, pressure, etc.), on the misorientation of the adjacent grains and GB orientation. A precise measurement and thus, examination of the misorientation dependence of GB mobility was made possible by tracking techniques of GB migration in bicrystals. The distinctive properties of such techniques are: controlled driving force, continuous tracking of GB displacement, accuracy and reproducibility of GB crystallography [**1]. As a first milestone, from measurements with these techniques materials scientists became aware that properties of GBs with different misorientation can be essentially different. In particular, it was established that GB mobility and its parameters are changing in a non-motonic way with the angle of misorientation.

For special misorientations (low-Σ boundaries) the activation enthalpy H_m of GB migration assumes a minimum. An example is shown in Fig. 1, where the misorientation dependence of GB mobility $\underline{A}_b = A_0 \exp(-H_m/kT)$ and its parameters for tilt GBs in pure aluminium (99.9995 at%) is presented [**1]. The similarity between the misorientation dependence of the migration activation enthalpy H_m and the logarithmic pre-exponential factor $\ln A_0$ is obvious. This similarity, which frequently has been observed experimentally, manifests itself quantitatively in a linear relation between the activation enthalpy

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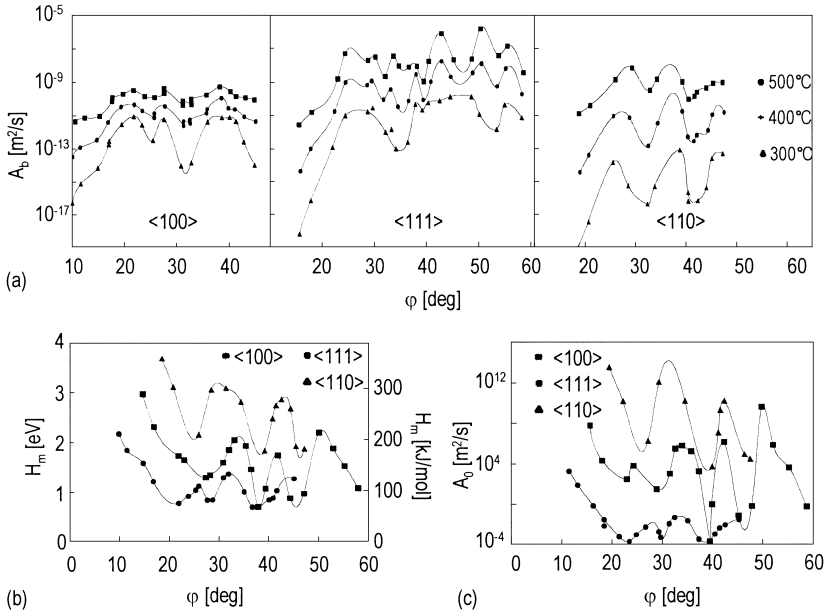

Fig. 1. Temperature and misorientation dependency of the reduced GB mobility A_b and its parameters for ⟨100⟩, ⟨111⟩, and ⟨110⟩ tilt grain boundaries in aluminium at 500, 400 and 300°C [**1]. (a) Reduced mobility; (b) enthalpy of activation; (c) pre-exponential factor.

of migration and the logarithm of the pre-exponential factor [**1,**2]. The relation is referred to as the compensation effect, since it strongly moderates the impact of a variation of activation enthalpy on the value of GB boundary migration rate *v*. As can be seen, the linear dependency leads to the existence of a so-called compensation temperature T_c, which is determined by the slope of the plot of H_m vs. $\ln \underline{A}_0$. At T_c all GB mobilities are equal and the kinetic lines in Arrhenius coordinates intersect at this temperature. This behavior has an important consequence. When the temperature is above T_c, grain boundaries with higher activation enthalpy assume higher values of the mobility. Conversely, when a mobility-controlled experiment is conducted at a temperature below T_c, grain boundaries with maximum values of activation enthalpy exhibit the minimum GB mobility.

The compensation effect can be associated with the fact that the activated state is not a random energy fluctuation in space and time but a defined and thus reproducible although unstable state, which is described by its respective thermodynamic functions [**2]. The linear compensation relation and the expression for the compensation temperature can be derived under these conditions [**2]. In particular, the compensation temperature can be expressed as:

$$T_c = \frac{dH_m}{dS_m}\Big|^{\lambda=\lambda_0}$$

where the parameter λ denotes some intensive structural or chemical specification, like angle of misorientation, composition, etc. In this approach T_c is the equilibrium temperature between the ground state and an activated state.

The compensation effect plays an important role in grain growth and grain structure evolution, since it establishes the relationship between the mobilities of grain boundaries in the granular system at different temperatures and pressures. In particular, the compensation effect concept provides the basis for an explanation of the classical contradiction — which was discussed for several decades — between the misorientation dependence of GB mobility obtained in bicrystal experiments and the results of growth selection experiments carried out by Lücke and co-workers in the 1950s. Actually, those experiments showed convincingly that the fastest boundaries were not the special ⟨111⟩ tilt boundaries Σ7, but non-special boundaries with a misorientation angle of slightly more than 40°. This contradiction was resolved in [*3,4], when the misorientation dependence of GB mobility for ⟨111⟩ tilt boundaries was investigated on a fine scale (Fig. 2). The experiments reveal that the mobilities of differently oriented boundaries have different temperature dependencies (Fig. 3). Namely, at 'low' temperatures ($T < T_c$) the exact Σ7 GB moves fastest, while at relatively high temperatures ($T > T_c$) the 40.5° ⟨111⟩ GB is the most mobile (Fig. 4). It is notable that for the first time the bicrystal and growth selection data had been reconciled.

The possibility to analyse experimental results by an independent method, in particular by computer simulation, is one of the principal advantages of a pure physical experiment. Atomistic simulations of GB motion were performed using molecular dynamics [5] for, what is of importance, the same geometry as was used in experiment to ensure steady-state, curvature driven boundary migration [**1] as a function of temperature and GB misorientation. These simulations represent the first systematic

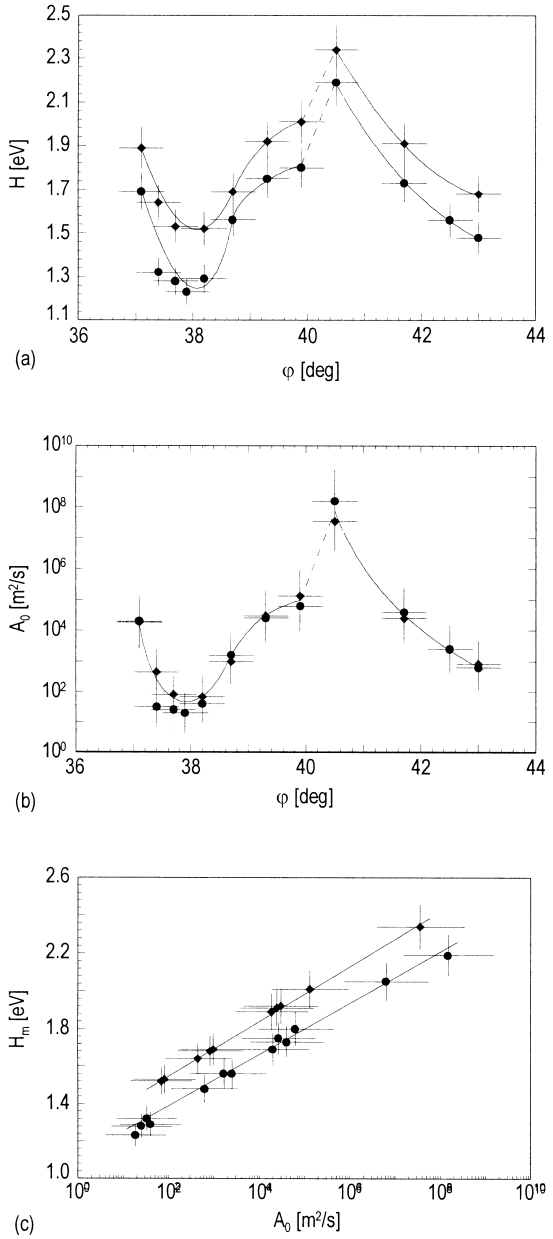

Fig. 2. Misorientation dependency of migration activation enthalpy H_m and pre-exponential reduced mobility factor A_0 for ⟨111⟩ tilt GBs in the vicinity of the special misorientation Σ7 in high purity Al of two charges with different impurity content: Al I, 0.4 ppm (●); Al II, 1.0 ppm (♦) [**2,*3]. (a) Misorientation dependence of migration activation enthalpy; (b) misorientation dependence of reduced mobility preexponential factor; (c) dependence of migration activation enthalpy H_m on the pre-exponential factor A_0 for the investigated ⟨111⟩ tilt GBs.

computational study of the dependence of GB mobility on misorientation (Fig. 5) and, therefore, probably the only feasible study of intrinsic (impurity free) boundary mobility since experimentally it is virtually impossible to fabricate bicrystals of perfect purity. Excellent agreement between simulations and experiments was obtained in almost all respects. Specifically, the activation energy and the logarithm of the pre-exponential factor of GB mobility

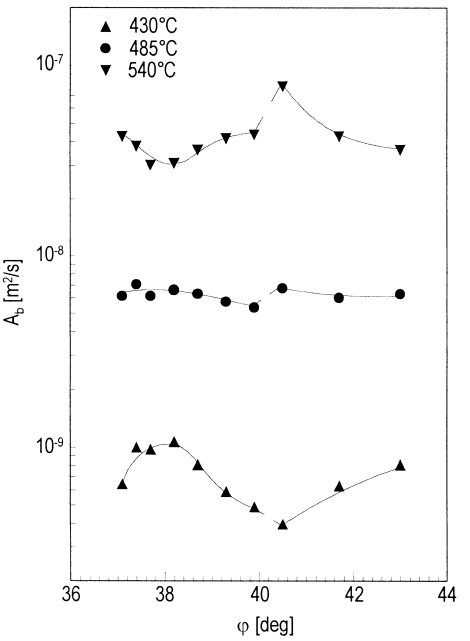

Fig. 3. Misorientation dependence of the reduced mobility A_b for ⟨111⟩ tilt GBs in Al II (see Fig. 2) at different temperatures [*3].

exhibited very similar variations with misorientation, including the presence of distinct cusps at low Σ misorientations.

It seems unnecessary to address the question whether the discussed misorientation relationships depend on the kind of driving force. In Refs. [6,7], a method to activate and investigate the migration of planar symmetrical and asymmetrical ⟨111⟩ and ⟨112⟩ tilt boundaries in aluminium bicrystals with 1 or 7.7 ppm impurity contents under the action of an external shear stress was introduced. It was shown that low-angle as well as high-angle tilt boundaries can be moved by such shear stress (Fig. 6). Surprisingly, the migration kinetics of grain boundaries driven by the shear stress are essentially different from those obtained in experiments with capillary driven boundaries (Fig. 6). The migration activation enthalpy obtained in experiments with

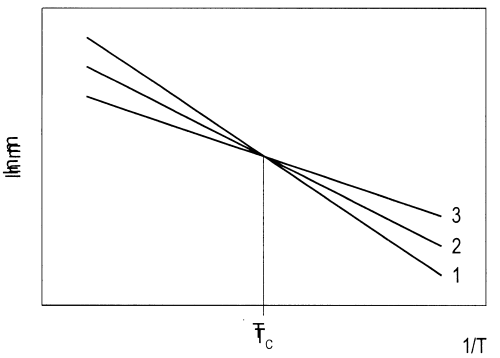

Fig. 4. Schematic sketch demonstrating the impact of the compensation effect on GB mobility m_b above and below T_c [**1]. Lines 1,2 and 3 represent Arrhenius dependencies of grain boundaries with different activation enthalpies H_m.

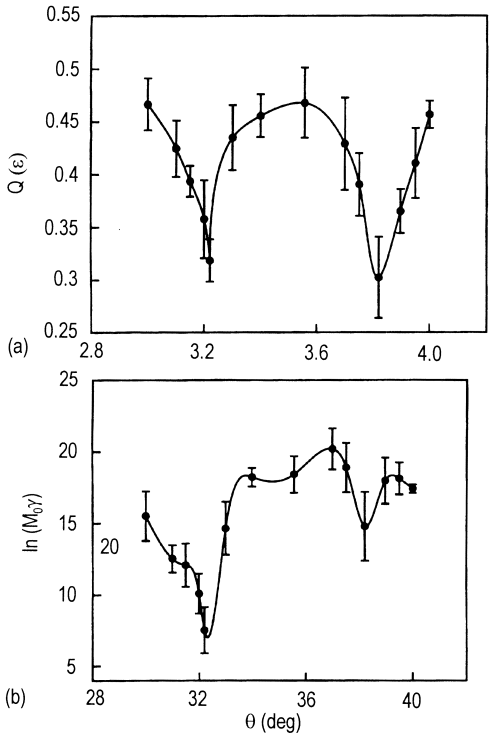

Fig. 5. The activation energy for GB migration (a) and the logarithm of the pre-exponential factor (b) of the reduced mobility as a function of boundary misorientation found in simulations of grain boundary motion [5].

shear-stress-driven boundaries is independent of misorientation angle except for a step at the transition from low- to high-angle boundaries (Fig. 5). In other words, a shear stress driven GB does not 'feel' the special misorientation, or generally speaking, the boundary structure, except for the difference between low and high angle boundaries. The migration activation enthalpy for stress driven motion of low and high angle boundaries was attributed to the activation enthalpy of bulk and GB self-diffusion correspondingly [*6,7]. The transition from low-angle to high-angle boundaries was found to manifest itself as a sharp step of the activation energy at a misorientation

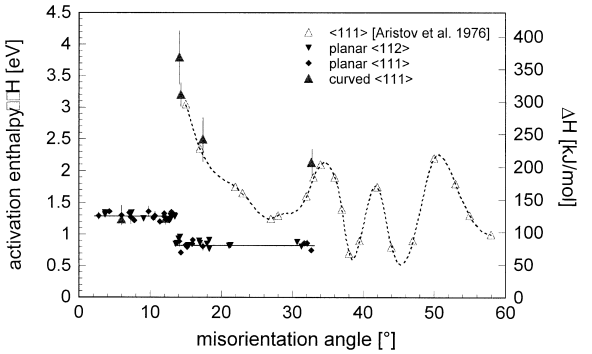

Fig. 6. Dependency of the activation enthalpy on misorientation angle for (curved) curvature driven and stress driven planar ⟨111⟩ and ⟨112⟩ tilt grain boundaries (filled symbols [*6], open symbols [8]) [*6].

angle of 13.6±0.5° which, in the framework of experimentally obtained data, seems to be independent of tilt axis, impurity content and tilt boundary plane. It has been common to assume the border between low and high angle boundaries to be in the region of 15° for structural reasons, expressed by Brandon [Acta Metall. 14 (1966) 1429]. The measurements on stress driven boundaries can be considered as the first determination of this border afforded by a difference in GB property, namely GB mobility.

The motion of a single dislocation under a shear stress is textbook knowledge, the motion of a low-angle grain boundary under the same stress was demonstrated in the well known experiments of Washburn and Parker [9], but the migration of a high-angle boundary under the action of a mechanical stress comes as a surprise. Actually, according to traditional conception, a mechanical stress does not couple with a high-angle GB since it is not considered to carry a strain field. Of course, if a high angle grain boundary is associated with an undercooled melt, as was assumed in some early models of grain boundary structure, there should be no place for a residual stress field. More recent concepts of GB structure however, propose to conceive high-angle boundaries as being composed of periodic arrangements of primary and secondary GB dislocations [10]. The relaxed structure of these arrangements as predicted by computer simulations corresponds quite well to HREM images of GB structure [11]. Nevertheless, the relaxation of the core structure of GB dislocations is also tacitly assumed to relax the stress field of structural dislocations. The experimental results reported in [*6,7] imply that this fundamental hypothesis is wrong. No matter what the misorientation angle is, a resolved shear stress on the GB will couple with the residual strain field of its (incompletely) relaxed dislocation structure. From this point of view the effect of a resolved shear stress on a GB is a force acting on the dislocation structure of the GB. Consequently, the experimentally observed dependencies, in particular the migration activation enthalpy of low angle GBs, have to be considered in terms of dislocation properties [*6,7].

Finally, we would like to discuss the impact of the orientation of the GB plane on its mobility. As mentioned above, the motion of a curved GB — the curvature is essential to create the capillary driving force — prevents us from associating the obtained mobilities with a specific GB structure. Rather, the motion of flat boundaries has to be investigated. The motion of a planar GB under the action of a magnetic field in bicrystals of a material with anisotropic magnetic susceptibility (Bi) was investigated in [12,13]. The experiments were carried out on bicrystals of high-purity (99.999%) bismuth. Symmetrical and asymmetrical ($\Psi = 45°$) pure tilt GBs with 90° ⟨112⟩ misorientation were examined [12,13]. It was found that the motion of asymmetric GBs can be very different from symmetrical GBs. Firstly, the migration activation enthalpy for asymmetrical GBs is nearly one order of magnitude (!)

higher than for symmetrical GBs: 3.4–3.8 and 0.51 eV, respectively (Fig. 7a) with a compensation temperature close to the melting temperature of Bi. The most surprising feature is that in contrast to the symmetric boundary for an asymmetric tilt boundary the measured GB mobility was found to be distinctly different for the motion in opposite directions (Fig. 7b). There are several potential reasons for this anisotropy. First, there is an essential difference in the distance between the crystallographic planes on each side of the boundary. An estimation shows that this factor may change the velocity of grain boundary motion, however, the influence of it is unlikely to affect the velocity of GB motion by more than 20%, which is distinctly less than the observed effect [12,*13]. Second, because GB motion in Bi-bicrystals may be influenced by impurity drag, the difference in the diffusivity of impurities in two opposite directions in the anisotropic structure of Bi should be taken into account. Finally, as shown recently, the motion of a grain boundary in a magnetic field can be considered as a motion of a conductor in a magnetic field, or more strictly, as a motion of a region with a conductivity different from that of the surrounding matrix in a magnetic field. Such a motion causes an electromotive force and as a consequence, an additional dissipation of energy in a magnetic field [14]. This dissipation should, on the one hand, be different for symmetric and asymmetric GBs and, on the other hand, for asymmetric GBs be different for GB motion in opposite directions [14].

In any event, if this asymmetry of GB mobility holds also for other metals, it will have a serious impact on our understanding of grain boundary motion, since the mobility of a grain boundary is commonly conceived as not dependent on its direction of motion.

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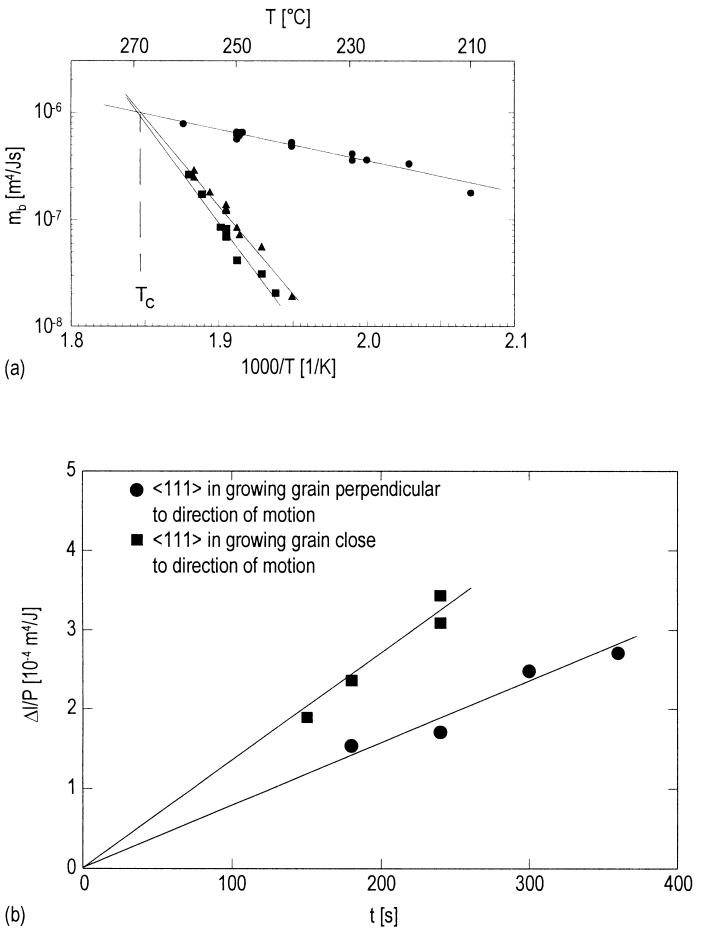

Fig. 7. (a) Temperature dependence of the mobility of 90° ⟨112⟩ symmetrical (●) and asymmetrical (▲, △, ■, □) tilt GBs in Bi-bicrystals, moving in opposite directions [12,13]. (▲ and △) Trigonal axis in the growing grain is parallel to the growth direction; (■ and □) trigonal axis in the growing grain is perpendicular to the growth direction. (b) Normalized displacement vs annealing time for asymmetrical tilt boundaries moving in opposite directions.

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