



## Grain Boundary Dynamics: A Novel Tool for Microstructure Control\*

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**Abstract.** The reaction of grain boundaries to a wide spectrum of forces is reviewed. Curvature, volume energy and mechanical forces are considered. The boundary mobility is strongly dependent on misorientation, which is attributed to both grain boundary structure and segregation. In magnetically anisotropic materials grain boundaries can be moved by magnetic forces. For the first time a directionality of boundary mobility is reported. Flat boundaries can also be moved by mechanical forces, which sheds new light on microstructure evolution during elevated temperature deformation. Curvature driven and mechanically moved boundaries can behave differently. A sharp transition between the small and large angle boundary regime is observed. It is shown that grain boundary triple junctions have a finite mobility and thus, may have a serious impact on grain growth in fine grained materials. The various dependencies can be utilized to influence grain boundary motion and thus, microstructure evolution during recrystallization and grain growth.

**Keywords:** grain boundary motion, triple junctions, magnetic effects, stress effects, compensation effect

### Introduction

Defects play an essential role in microstructure evolution, in particular 2D defects, i.e. internal surfaces between similar and dissimilar phases. Interfaces between equal phases but different crystallographic orientations are referred to as grain boundaries. Grain boundaries have the unique property that they react to exerted forces by a change of position. Grain boundary dynamics, i.e. the motion of grain boundaries under the action of forces is the main subject of this paper. A more detailed account of this and related topics can be found in a recent review by the current authors [1].

Commercial materials are polycrystalline, i.e. they consist of a large number of crystallites (grains) which are separated by grain boundaries. Each crystallite is surrounded by more than one grain which implies that grain boundaries form a spatial boundary network comprising boundaries, triple lines and quadruple junctions. If all boundaries and their junctions had equal properties, respectively, the temporal change of grain structure evolution would be sufficient to characterize the kinetic properties of grain boundaries. Fortunately, nature has chosen to make boundaries different, which gives us a powerful tool for microstructure control. Therefore, by definition, there is no average representative grain boundary, and grain boundary properties are most appropriately measured on specific individual grain boundaries, i.e. in bicrystals. Equivalently, measurement of junction properties requires tricrystal experiments, etc.

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### Measurement of Grain Boundary and Triple Junction Velocity

The measurement of grain boundary velocity seems to be a trivial problem, since only the displacement with time needs to be recorded. Unfortunately, metallic materials are opaque so that the grain boundary position can only be observed on the surface. A boundary in contact with the surface, however, feels the surface tension of the free surface and thus, tends to form a groove to balance the surface tension. A boundary groove, however, impedes grain boundary motion and thus, interferes with the free motion of the boundary, and a jerky motion of the boundary is the result. Therefore, boundary velocity measurements by stepwise annealing can give erroneous results as shown in the past [2]. Instead, continuous tracking of boundaries without interference with the migration process is necessary. This can be accomplished by utilizing the discontinuity of orientation dependent properties at the boundary, e.g. the intensity of diffracted X-rays or the intensity of backscattered electrons in a scanning electron microscope. Most measurements so far have been conducted with the XICTD (X-Ray Interface Continuous Tracking Device). It allows measurements under atmospheric pressure and inert gas atmosphere, which retards groove formations [3].

For the investigation of grain boundary systems (triple junctions, networks) the orientation dependence of backscattered electron intensity in an SEM can be used [4]. In materials with non cubic crystal symmetry, the anisotropy of reflectivity of visible light can be exploited for orientation contrast to reveal the boundary position [5].

### Forces on Grain Boundaries

A grain boundary of area  $F$  experiences a force  $P$  per unit area (at constant temperature  $T$  and pressure  $p$ ) when its displacement  $dx$  results in a decrease of the Gibbs free energy  $dG$

$$P = -\frac{dG}{F dx} = -\frac{dG}{dV} \quad (1)$$

where  $V$  is the swept volume. The gain of free energy can result from a volume free energy difference across the boundary e.g. by the anisotropy of free energy density in a magnetic or elastic field, or from a change of the grain boundary energy due to a reduc-

tion of the grain boundary area in case of curved grain boundaries.

### Grain Boundary Mobility

Grain boundary motion is comprised by a non-zero net flux of lattice sites across the grain boundary. Atoms crossing the boundary will change their energy state (Fig. 1) which results in an asymmetry of the atomic migration energy and thus an imbalance of the jump rate across the boundary in both directions.

Each atom of volume  $\Omega_a \approx b^3$  will gain the free energy  $Pb^3$  when becoming attached to the growing grain but has to expend this free energy when moving in the opposite direction. Correspondingly, with a boundary displacement  $b$  per atom the boundary velocity reads

$$v = b \left( e^{-\frac{G_m}{kT}} - e^{-\frac{G_m + Pb^3}{kT}} \right) \quad (2)$$

For all practical cases, including recrystallization in heavily cold worked metals,<sup>1</sup>  $Pb^3 \ll kT$  at temperatures where boundaries are observed to move ( $T \geq 0.3T_m$ ) and, therefore,

$$v = \frac{b^4 v_D}{kT} e^{-\frac{G_m}{kT}} \cdot P \equiv m_b \cdot P \quad (3)$$

where  $m_b$  is referred to as grain boundary mobility.

This very simple model may be refined by assuming the detachments to occur in a sequence of steps or that thermal grain boundary vacancies have to assist

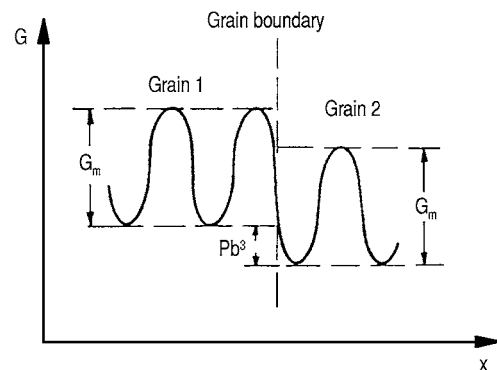


Figure 1. The free energy of a moving atom changes by the driving force  $Pb^3$  when it crosses the boundary.  $G_m$  is the free energy barrier for bulk diffusion.

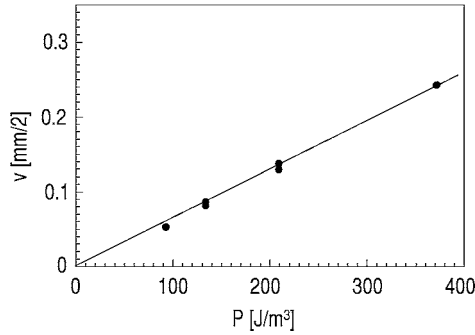


Figure 2. Measured grain boundary migration rate vs. driving force of a flat boundary in a bicrystal of Bi exposed to a magnetic field.

diffusion [6]; however, these modifications will only affect the preexponential factor  $m_0$  and the activation enthalpy  $H$  of the mobility  $m_b = m_0 e^{-H/kT}$ , but the proportionality between migration rate  $v$  and driving force  $P$  remains unchanged.

Despite some previous confusion due to improperly conducted experiments as shown in [7], relation (6) has been confirmed by many experiments with different driving forces (Fig. 2). Hence, the important kinetic property of a grain boundary with regard to grain boundary motion is the mobility. It depends very sensitively on grain boundary crystallography and grain boundary chemistry. According to recent findings, the mobility may even not be a unique scalar property of a given boundary, as it may depend on the direction of motion for asymmetric boundaries. For sake of simplicity, however, we shall consider the grain boundary mobility as a specific boundary property except otherwise noted.

### Kinetic Parameters of Grain Boundary Mobility

Grain boundary migration is a thermally activated process. Thus, its kinetics follow an Arrhenius type temperature dependence

$$v = v_0 \exp\left(-\frac{H_m}{kT}\right) \quad (4)$$

Since the driving force is essentially independent of temperature, the temperature dependence of  $v$  is the temperature dependence of the grain boundary mobility

$$m_b = v/P = m_0 \exp(-H_m/kT) \quad (5)$$

where  $H_m$  is the activation enthalpy of grain boundary migration and  $m_0$  the corresponding preexponential mobility factor.

Since most results referred to in the following were obtained for the migration of curved grain boundaries, we introduce for simplicity the reduced mobility

$$A_b = m_b \cdot \sigma_b = A_0 \exp\left(-\frac{H_m}{kT}\right) \quad (6)$$

where  $\sigma_b$  is the grain boundary surface tension.

Any influence on grain boundary mobility will be reflected in a change of  $H_m$ , and/or  $m_0$  (resp.  $A_0$ ). Therefore, in the following we will discuss the dependence of these kinetic parameters on the structure and chemistry of grain boundaries.

### Orientation Dependence of Mobility

Figure 3 shows the dependence of the activation energy for the mobility of  $\langle 100 \rangle$  tilt grain boundaries on misorientation in Al with different content of impurities. For small angle boundaries the activation energy is seen to decrease with increasing misorientation. For high angle boundaries ( $\geq 20^\circ$ ) the activation energy depends strongly on impurity level. For both ultrapure (99.99995%) and low purity (99.98%) material, the activation energy does not depend on angle of rotation, but the absolute value of the activation energy is by a factor of three higher for the impure material. For less than ultrapure material (99.992%, referred to as high purity material), the activation energy oscillates and attains minima for special angles of rotation, which correspond to low  $\Sigma$  CSL rotations. For these special misorientations the activation energy is practically

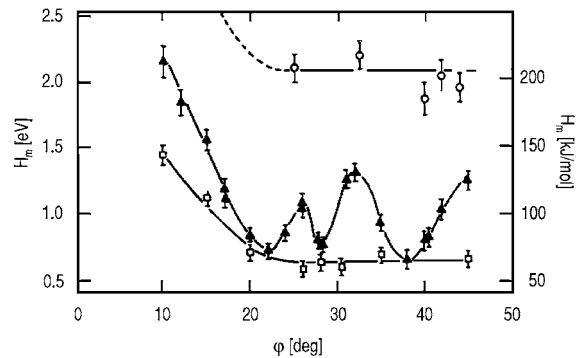


Figure 3. The dependence of the activation enthalpy of migration for  $\langle 100 \rangle$  tilt grain boundaries in Al of different purity:  $\circ$ —99.99995 at.%;  $\blacktriangle$ —99.9992 at.%;  $\circ$ —99.98 at.%.

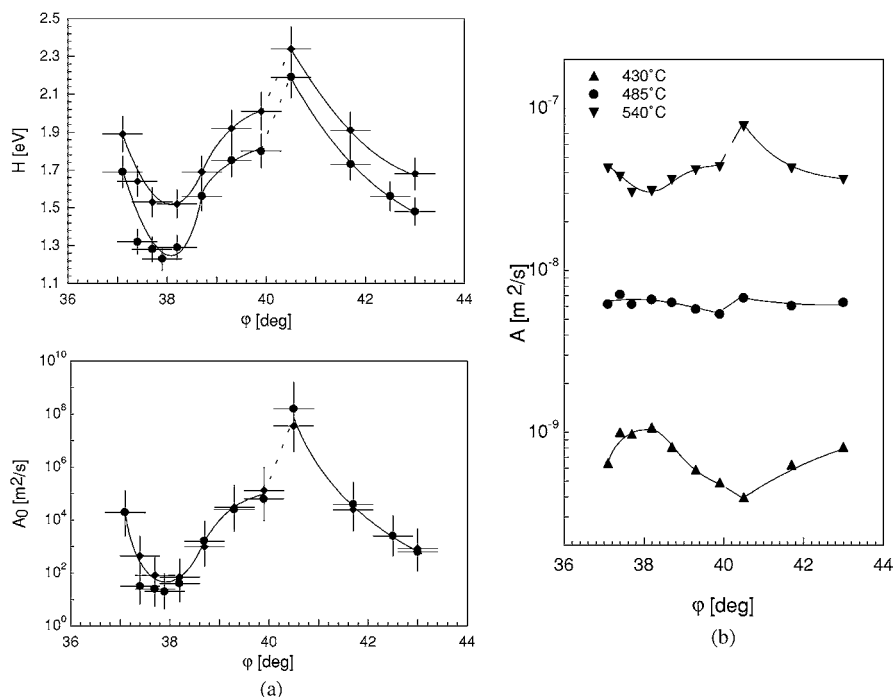


Figure 4. (a) Activation enthalpy  $H$  and preexponential factor  $A_0$  for  $\langle 111 \rangle$  tilt boundaries in pure Al of different origin ( $\bullet$ —Al I;  $\blacklozenge$ —Al II); (b) Mobility dependence of  $\langle 111 \rangle$  tilt grain boundaries on rotation angle in pure Al.

identical for high purity and ultrapure material. Thus, experimental results on single grain boundaries qualitatively confirm the findings on polycrystals [8–10] that in high purity materials special boundaries have a higher mobility than random boundaries.

It is customary to associate the absolute value of the migration rate with the magnitude of the activation enthalpy only, such that the highest activation enthalpy indicates the lowest migration rate. This is not true, however, especially for interfaces due to the so-called compensation effect, which allows one to use annealing temperature as a selective tool for microstructure control.

Studies of the grain boundary mobility in Al bicrystals [11] have shown that tilt grain boundaries with  $\langle 111 \rangle$  rotation axis and rotation angle of about  $40^\circ$  have the highest mobility. This is commonly understood such that grain boundaries with highly periodic coincidence structure (so called low  $\Sigma$  or special boundaries) move faster than off-coincidence (random) boundaries, i.e. the special  $\Sigma 7$  ( $38.2^\circ \langle 111 \rangle$ ) tilt boundary was identified as the fastest boundary in Al. However, from growth selection experiments [12–14] it was known that the rotation angle of the fastest boundary was distinctly larger than  $38.2^\circ$ .

We investigated the misorientation dependence of grain boundary mobility on a fine scale in the angular interval  $37^\circ$ – $43^\circ$   $\langle 111 \rangle$  with angular spacing  $0.3^\circ$ – $0.6^\circ$ . The experiments reveal that both the activation enthalpy and the preexponential factor are at maximum for a misorientation angle  $\phi = 40.5^\circ$  and at minimum for the exact  $\Sigma 7$  orientation (Fig. 4(a)). Therefore, one is tempted to conclude that the  $\Sigma 7$  boundary has the highest mobility. However, the mobility of boundaries with different misorientation angles do have a different temperature dependence, and there is a temperature, the so-called compensation temperature  $T_c$ , where the mobilities of all investigated boundaries of differently misoriented grains are the same. As a result, for  $T > T_c$ , the mobility is higher for grain boundaries with higher activation energy, in particular it is at maximum for  $\phi = 40.5^\circ$ , while for  $T < T_c$  the exact  $\Sigma 7$  boundary moves fastest (Fig. 4(b)).

This result explains the apparent contradiction between growth selection experiments and recrystallization experiments. The problem resulted only from the wrong tacit assumption that the preexponential factor is essentially independent of misorientation so that only the activation enthalpy controls mobility. Growth selection experiments have to be conducted at very high

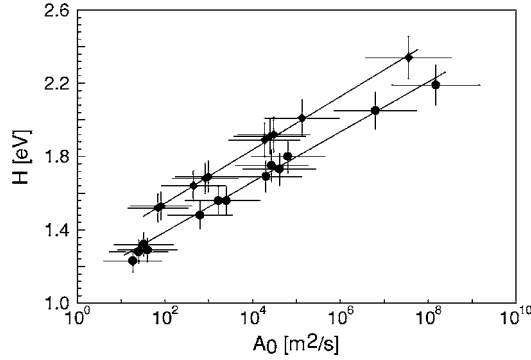


Figure 5. Dependence of migration activation enthalpy on preexponential mobility factor for (111) tilt grain boundaries in Al of different purity (● 0.4 ppm, ◆ 1 ppm).

temperatures (above 600°C), i.e. in the temperature regime, where, according to results of the current study, the mobility of the 40.5°(111) boundary is the highest due to its high preexponential factor. The reason for the changing maximum mobility orientation in different temperature regimes is obviously the orientation dependence of both, the activation enthalpy and the preexponential factor. In fact, both are related to each other in a linear fashion (Fig. 5), i.e.

$$H = \alpha \ln A_0 + \beta \quad (7)$$

where  $\alpha$  and  $\beta$  are constants. This correlation is referred to as the compensation effect [15]. The value of  $\alpha = kT_c$  defines the compensation temperature  $T_c$ . Effectively the compensation effect states that at high temperatures ( $T > T_c$ ), i.e. above the compensation temperature  $T_c$ , the process with the highest activation energy proceeds fastest, while at low temperatures ( $T < T_c$ ), grain boundaries with the lowest activation energy exhibit the highest mobility and at  $T = T_c$  all boundaries move at the same rate. It is important to keep this in mind, when discussing the temperature dependence of grain boundary mobility.

### Magnetic Driving Forces

A curved grain boundary implies that its structure changes along the boundary, since it is composed of different grain boundary planes. The mobilities obtained cannot, therefore, be related to a specific grain boundary structure. Although experimentally more complicated, also a planar boundary can be forced to move, if a bicrystal with grains that have some orientation dependent property, like elastic constants or magnetic susceptibility, are utilized. Under the impact of a respective

directed external field the property anisotropy will generate a free energy difference between adjacent grains that creates a driving force for grain boundary displacement. This driving force does not depend on boundary properties and moves a boundary from the grain with lower free energy toward the one with higher free energy. Such conditions, in particular, can be obtained by the action of a magnetic field on a bicrystal of a material with anisotropic magnetic susceptibility.

The origin of the driving force for grain boundary migration in a magnetically anisotropic material was considered by Mullins [16]. The expression for the driving force, as applied to bismuth, reads

$$P = \mu_0 \frac{\Delta\chi}{2} H^2 (\cos^2 \Theta_1 - \cos^2 \Theta_2) \quad (8)$$

where  $H$  is the magnetic field strength,  $\Delta\chi$  the difference of the susceptibilities parallel and perpendicular to the trigonal axis,  $\Theta_1$  and  $\Theta_2$  are the angles between the magnetic field and the trigonal axes in both grains of the Bi-bicrystal.

The grain boundary motion was measured by applying a magnetic field to a specially prepared bicrystal, i.e. to a system that was in equilibrium without an applied magnetic field [17, 18]. The experiments were carried out on specifically grown bicrystals of high purity (99.999%) bismuth. A 90°(112) boundary was examined. The misorientation angle between trigonal axis in both crystals of the bicrystal was chosen to be 90° in order to gain the maximum possible magnetic driving force (Eq. (8)). Prior to their exposure to a magnetic field (the field strength used was  $1.63 \cdot 10^7$  A/m) the samples were annealed for 10 hours at 230°C in vacuum. To ensure sufficient boundary mobility the magnetic field was imposed on the samples at a temperature of 255°C ( $0.97T_m$ ).

The experiments unambiguously confirmed that grain boundaries in Bi-bicrystals actually move under the action of a magnetic driving force. The observed linear dependence of boundary displacement on annealing time proves the free character of its motion (Fig. 2). In contrast to the symmetric tilt boundary for the asymmetric tilt boundary the measured boundary mobilities were found to be distinctly different for motion in opposite directions (Fig. 6). For the chosen crystallography of the bicrystals the boundary was less mobile, when the  $c$ (111) axis in the growing grain was perpendicular to the direction of motion ( $m_{\perp} = 8.0 \times 10^{-8}$  m<sup>4</sup>/J·s) and moved faster, when the trigonal  $c$  axis in the growing grain was close to the direction

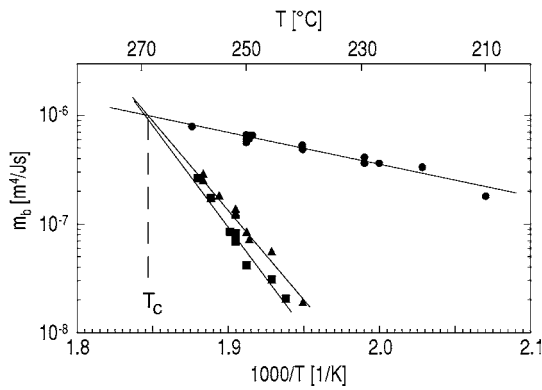


Figure 6. Temperature dependence of symmetrical (●) and asymmetrical (■, ▲) tilt boundaries in Bi.

of motion ( $m_{||} = 1.3 \times 10^{-7} \text{ m}^4/\text{J} \cdot \text{s}$ ). In this respect it is interesting that the symmetric tilt boundary exhibited a much higher mobility than the asymmetric tilt boundary and did not show a dependence of boundary mobility on the sense of motion (Fig. 6).

In any event, if this asymmetry of grain boundary mobility holds also for other metals, it would 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.

### Effect of Impurities

If there is an interaction energy  $U$  (energy gain) between boundary and impurity atoms, these solute atoms will tend to segregate to the boundary. In fact, if thermal equilibrium could be established at all temperatures, and the boundary could adsorb an unlimited number of impurities—like in the case of the Henry isotherm—all solute atoms would end up in the boundary at  $T = 0 \text{ K}$ . Owing to thermal agitation (entropy effect), for  $T > 0$  the concentration in the boundary will be

$$c_b = c_0 \exp\left(\frac{U}{kT}\right) \quad (9)$$

where  $c_0$  is the volume impurity concentration. When the grain boundary moves, the segregated atoms will attempt to remain in the boundary, i.e. the boundary has to drag its impurity load and can only migrate as fast as the slowly moving impurities. Hence, grain boundary motion is understood to be slowed down by the presence of impurities, and this is also the prediction of theories of impurity drag as derived by Lücke and Detert [19], Cahn [20], and Lücke and Stüwe [21].

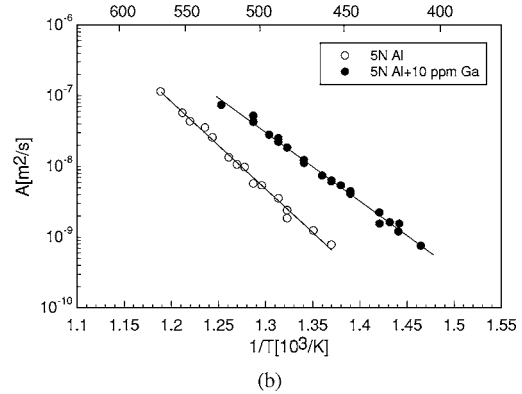
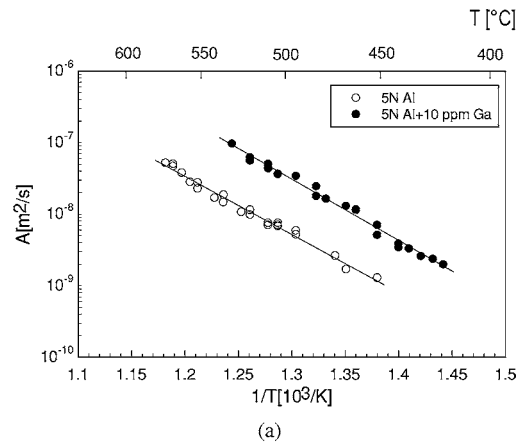


Figure 7. Arrhenius plot of mobility of (a)  $38.2^\circ$  and (b)  $40.5^\circ$  (111) tilt grain boundaries in pure Al and pure Al doped with 10 ppm Ga.

All known experiments on bicrystals and polycrystals confirm that solute atoms reduce the rate of boundary motion. However, it is important to realize that solute atoms not necessarily hinder grain boundary motion as evident from the addition of minor amounts of gallium to aluminum (Fig. 7). The experiments were carried out on bicrystals of both pure Al and the same Al doped with 10 ppm Ga. Irrespective of the type of boundary, whether special or nonspecial, 10 ppm gallium in aluminum substantially increases grain boundary mobility, which means that it substantially speeds up recrystallization kinetics. Addition of 10 ppm Ga effectively increases the mobility of both investigated  $38.2^\circ$  and  $40.5^\circ$  (111) tilt boundaries.

### Effect of Mechanical Stresses

In 1952 Washburn, Parker et al. [22, 23] investigated planar low angle boundaries in Zn under the influence

of an external shear stress and observed the motion with polarized light in an optical microscope. This observation method does not work for cubic crystal structures, however, and only recently a method was developed for in-situ observation of grain boundary motion in aluminum [3]. Investigations were conducted on bicrystals with  $\langle 112 \rangle$  and  $\langle 111 \rangle$  tilt grain boundaries with misorientation angles in a range from  $4^\circ$  to  $32^\circ$ .

Symmetrical low angle tilt boundary consist of periodic arrangements of a single set of edge dislocations. An external shear stress perpendicular to the boundary plane will cause a force on each dislocation and in summary a driving force on the boundary. The samples were exposed to a shear stress ranging from  $10^{-1}$  MPa to  $10^{-3}$  MPa. In aluminum (purity 99.999%) the yield stress is 15–20 MPa [24], hence the applied shear stress is definitely in the elastic range.

The dependence of the grain boundary velocity on the applied mechanical shear stress for two different  $\langle 112 \rangle$ -tilt boundaries is shown in Fig. 8. Obviously, the low angle grain boundary moves under the influence of the shear stress and the grain boundary velocity changes in proportion to the stress. Contrary to expectation, also the high angle grain boundary is forced to move by the exerted mechanical stress. Figure 8 shows the dependence of the activation enthalpy on misorientation angle for different tilt axes and impurity content. For low angle grain boundaries we find a constant activation enthalpy of  $\Delta H = 1.28$  eV and for high angle grain boundaries  $\Delta H = 0.85$  eV. The transition from low angle to high angle grain boundaries is revealed by a conspicuous step of the activation enthalpy at a misorientation angle of  $13.6^\circ$ .

From Fig. 9 we conclude that  $\langle 112 \rangle$ - and  $\langle 111 \rangle$ -tilt boundaries move with the same activation enthalpies

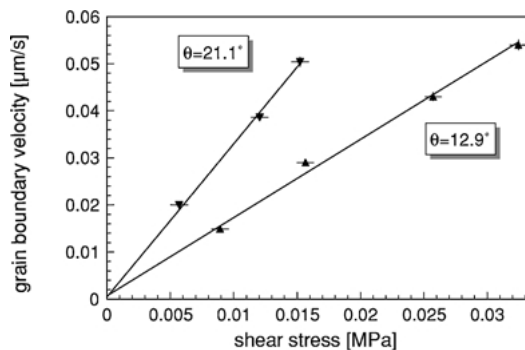


Figure 8. Dependency of the grain boundary velocity on the external shear stress for two symmetrical  $\langle 112 \rangle$  tilt boundaries.

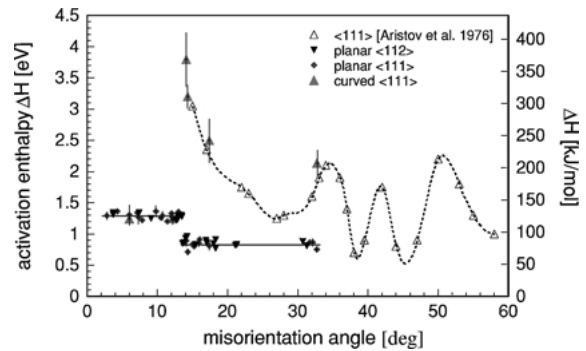


Figure 9. Dependency of the activation enthalpy on misorientation angle for curvature driven and planar  $\langle 111 \rangle$  grain boundaries, and planar  $\langle 112 \rangle$  grain boundaries (open symbols [29]; filled symbols this work).

when exposed to a mechanical stress. This holds for low angle as well as for high angle symmetrical tilt boundaries.

As shown previously, experiments on curvature driven  $\langle 111 \rangle$  tilt boundaries in aluminum bicrystals showed a strong misorientation dependence of the activation enthalpies. For comparison, we conducted curvature driven boundary migration experiments on  $\langle 111 \rangle$  tilt boundaries. The driving force was a constant capillary force,  $p = \sigma/a$  where  $a$  is the width of the shrinking grain.

In Fig. 9 the dependence of the activation enthalpy on the misorientation angle for the curved and the planar grain boundaries is shown. For the curvature driven grain boundaries our results are in good agreement with previous experimental data [11, 25], and one can see a strong dependency of the activation enthalpy on the misorientation angle, i.e. on grain boundary structure. There is also a clear difference between the activation enthalpies for the stress induced motion of the planar high angle grain boundaries and the curvature driven migration of the curved high angle grain boundaries.

The results prove that grain boundaries can be driven by an applied shear stress irrespective whether low or high angle boundaries. The activation enthalpy for the low angle grain boundaries amounts to  $\Delta H = 1.28$  eV and is comparable to the activation enthalpy of self diffusion in aluminum. For the high angle grain boundaries we found an activation enthalpy of  $\Delta H = 0.85$  eV, which is comparable to the activation enthalpy for grain boundary diffusion in aluminum.

As a result it is noted that strong effects are expected where boundaries are exposed to high internal stresses.

This may be instrumental for nucleation and growth during recrystallization or high temperature deformation of polycrystals.

### Steady State Motion of Triple Junctions

Polycrystals comprise a grain boundary network composed of grain boundaries and their junctions. A specific mobility of triple junctions was first introduced by Galina et al. [26], who considered the steady-state motion of a grain boundary system with a triple junction (Fig. 10). The problem was considered in the quasi-two-dimensional approach for a uniform grain boundary model (the boundaries of the system are considered identical, in particular their surface tension  $\sigma$  and their mobility  $m_b$ . Furthermore, it is assumed that  $m_b$  and  $\sigma$  are independent of the inclination of the grain boundaries). These assumptions define the problem to be symmetric with regard to the  $x$ -axis. With these simplifications some very important features of the motion of this system can be established. As was shown in [5, 26], a steady-state motion of the whole system is possible indeed.

In the following a boundary system as shown in Fig. 10 with two identical curved boundaries (GB I and II) and a different straight boundary (GB III) will be considered.

The steady state velocity of GB I and II is:

$$v^{\text{GB}} = \frac{2\theta m_b \sigma}{a} \quad (10)$$

For the situation given in Fig. 10 the triple junction velocity  $v^{\text{TJ}}$  can be expressed [27] by

$$v^{\text{TJ}} = m_j (2\sigma \cos \theta - \sigma_3) \quad (11)$$

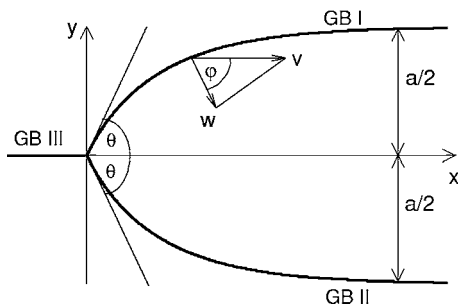


Figure 10. Geometry of a grain boundary system with triple junction.

where  $m_j$  is the junction mobility. In the case of steady state motion of the boundary system the velocity of the triple junction equals the velocity of the grain boundaries. Therefore, the steady-state value of the angle  $\theta$  is determined by:

$$\frac{2\theta}{2 \cos \theta - \sigma_3/\sigma} = \frac{m_j a}{m_b} = \Lambda \quad (12)$$

The dimensionless criterion  $\Lambda$  reflects the drag influence of the triple junction on the migration of the system. One can distinguish two limiting cases:

- $\Lambda \rightarrow 0$ : In this case the angle  $\theta$  tends to zero, i.e. the motion of the entire boundary system is governed by the mobility of the triple junction and the corresponding driving force.
- $\Lambda \rightarrow \infty$ : In this case the angle  $\theta$  tends to the value of thermodynamic equilibrium:  $\theta = \arccos \sigma_3/2\sigma = \theta_{\text{eq}}$ . The motion of the system is independent of the triple junction mobility and is governed only by the grain boundary mobility and the corresponding driving force. The velocity of the boundary system in this case is given by Eq. (11) ( $\theta = \theta_{\text{eq}}$ ).

Corresponding experiments were carried out on zinc-tricrystals with a grain boundary geometry as shown in Fig. 10. The tricrystals were produced of high purity Zn (99.999 at.%) by a technique of directional crystallisation. For measuring the migration rate and the geometry of the grain boundary system during the motion at elevated temperatures a modified optical microscope operating with polarised light and a hot stage was used [5].

The motion of four different triple junctions with a grain boundary configuration as in Fig. 10 were investigated in the temperature range between 330°C and 405°C. The triple junctions of samples 1 and 2 consisted of two nearly identical high angle tilt grain boundaries (curved boundaries, GB I and II) and a low angle tilt boundary (straight boundary, GB III). For both types of triple junctions the shape of the moving grain boundary system was similar to the shape predicted by the theory.

For all samples the velocities  $v$  and the angles  $\theta$  were found to be constant for a given temperature over the entire investigated temperature range. Evidently, the assumption of a steady state motion of the entire grain boundary system was justified. The angle  $\theta$  increased



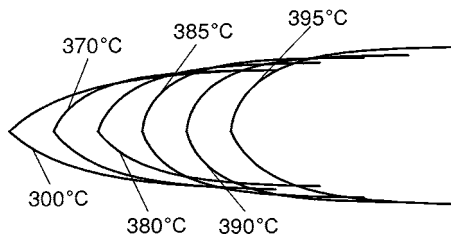


Figure 11. Evolution of the shape of a grain boundary system with triple junction with increasing temperature.

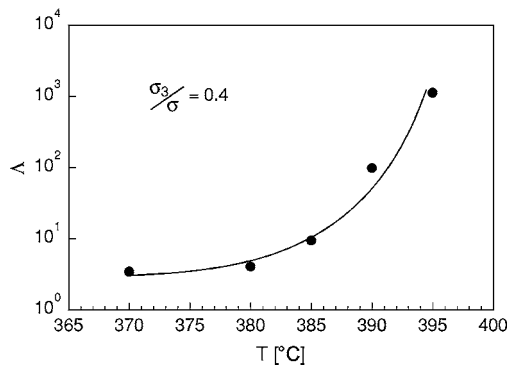


Figure 12. Temperature dependence of the criterion  $\Lambda$  for a grain boundary system with a triple junction.

with increasing temperature. In particular for the symmetrical triple junction the change of  $\theta$  was drastic (Fig. 11).

In accordance with the temperature dependence of  $\theta$ , the criterion  $\Lambda$ , determined by Eq. (12) was found to be constant for a given temperature, but increased with increasing temperature (Fig. 12). At low temperatures  $\Lambda$  was on the order of unity and increased with rising temperature up to 3 orders of magnitude. For the calculation of  $\Lambda$  for symmetrical triple junctions the ratio  $\sigma_3/\sigma$  was determined under the assumption that for temperatures near the melting point the value of  $\theta$  reaches the thermodynamic equilibrium value.

For the first time two different regimes of coupled triple junction—grain boundary motion were observed, indicated by a change of the angle  $\theta$  with temperature. At low temperatures, where  $\Lambda$  is on the order of unity (Fig. 12), the motion of the boundaries is dragged by the hardly mobile triple junction. Accordingly, the angle  $\theta$  is smaller than predicted by the equilibrium of grain boundary surface tensions, and the motion of the entire boundary system is controlled by the mobility of the triple junction. With increasing temperature the triple junction becomes more mobile compared to the grain

boundary mobility as indicated by an increasing value of  $\Lambda$  (Fig. 12).

The current investigation unambiguously proves the existence of a specific mobility of triple junctions, which usually differs from the mobility of the adjoining grain boundaries. As a consequence, the transition temperature between the two kinetic regimes, where the reduced mobilities  $A^{\text{GB}}$  and  $A^{\text{TJ}}$  are comparable, changes with grain size. For a large grain size, as characteristic for grain growth experiments, the mobility of grain boundaries is comparable to the mobility of triple junctions at relatively low temperatures. The kinetics in the temperature regime, which is characteristic for commercial recrystallization treatments, is controlled by the more slowly moving grain boundary. For a very small grain size, as characteristic for ultrafine grained material, e.g. nanocrystalline material, the situation is opposite.

The results are of particular importance for grain growth. During the motion of the grain boundary systems controlled by the mobility of grain boundaries the grain boundary velocity  $v$  is proportional to the grain boundary curvature ( $\sim 1/a$ ) (Eq. (12)) while in the case of triple junction controlled motion the velocity  $v$  is constant (Eq. (14)). The kinetics of the evolution of the mean grain size in the former case will be determined by the dependency  $v = \frac{da}{dt} \sim \frac{1}{a} \Rightarrow \langle a \rangle \sim \sqrt{t}$ , i.e. the mean grain size increases in proportion to the square root of the annealing time. In contrast, in the latter case  $v = \frac{da}{dt} \sim \text{const.} \Rightarrow \langle a \rangle \sim t$ , i.e. the mean grain size increases in proportion to the time of annealing. Experimental data relevant to such kinetics were actually obtained for grain growth of nanocrystals [28] and confirm a linear growth law.

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### Note

1. For heavily cold worked metals  $P \approx 10$  MPa. For Al  $kT \cong 10^{-20}$  J at  $T = 0.8T_m \cong 450^\circ\text{C}$ , i.e. with  $b = 3 \times 10^{-10}$  m;  $Pb^3/kT \cong 0.01$ . It is noted at this point that in molecular dynamics simulations where very high driving forces are applied to make the boundary noticeably move in the allowed small time interval, this approximation may not hold.

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