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Migration and reorientation of grain boundaries in Zn bicrystals during annealing in a high magnetic field

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Abstract

Zinc bicrystals with 89° $\langle 1\,0\,\bar{1}\,0\rangle$ symmetric tilt boundary inclined at different angles to the free surfaces are annealed in the field of 17 T. Boundaries migrate reorienting almost perpendicular to the surfaces. The observed effect is due to combination of magnetically induced and capillarity driven boundary migration. The absolute boundary mobility was measured to be about 2.5×10^{-8} $\mathrm{m^4/J\,s}$.

Keywords: Magnetic susceptibility; Boundary migration

1. Introduction

The effect of magnetic field on migrating grain boundary has been established in polycrystalline bismuth by Mullins [1]. Later Molodov et al. [2,3] have observed magnetically induced boundary migration in specially grown bismuth bicrystals with planar boundaries. It has been also found out that boundaries originally inclined to the free surfaces change inclination to become perpendicular to the free surfaces, which minimized the boundary area. Matsuzaki et al. [4] have reported that the rate of capillarity driven boundary migration in Fe–2.5%Si alloy bicrystals increases in the presence of magnetic field. In experiments with Zn bicrystals containing 86° $\langle 1\,1\,\bar{2}\,0\rangle$ tilt boundary Konijnenberg et al. [5] have shown, that also in zinc the magnetic anisotropy is pronounced well enough to drive grain boundaries in magnetic field with a strength of 20 MA/m. Recently, it has been demonstrated that crystallographic texture in Zn–1.1%Al can be affected drastically by a high magnetic field and that these texture changes are related to magnetically induced boundary migration [6]. In magnetically anisotropic materials the additional driving force for boundary migration (grain growth) is exerted by the difference in the magnetic free energy in neighboring grains differently oriented with respect to the field. If the volume density of the magnetic free energy ω in a crystal induced by a uniform magnetic field is

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independent on crystal shape and size (the condition for this is $\chi \ll 1$) then the magnetic driving force acting on the boundary of two crystals that have different magnetic susceptibilities is given by [1]:

$$p = \omega_1 - \omega_2 = \frac{\mu_0 H^2}{2}(\chi_1 - \chi_2), \tag{1}$$

where χ_1 and χ_2 are the susceptibilities of crystal 1 and 2, respectively, along the magnet field H. In most of the anisotropic diamagnetic materials the anisotropy of magnetic susceptibility is significantly smaller than in bismuth. Therefore, observations of magnetically induced boundary displacement in Zn bicrystals [5] and texture development in Zn alloy [6] have established a "new" class of diamagnetic materials whose grain boundaries are susceptible to magnetically induced migration in the direct current fields available nowadays. Drastic changes in texture of Zn–1.1%Al alloy in a high field occurs due to additional driving force for grain growth exerted by magnetic anisotropy of Zn [6]. The aim of this work is to study the behavior of originally planar individual boundaries in Zn bicrystals in a high magnetic field in the presence of capillary driving force for boundary displacement distinguishing the role of each driving force in the process of boundary migration.

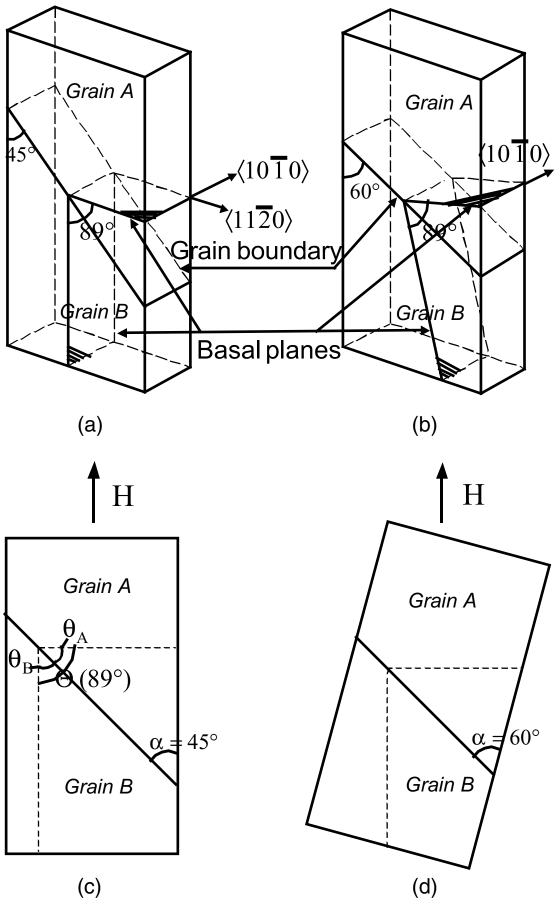

Fig. 1. Geometry of bicrystals (a, b) and their orientation to the field (c, d). Type I bicrystal (a, c), type II bicrystal (b, d).

2. Experimental procedure

Zinc bicrystals (99.995%) of two types containing a 88.7° ± 0.5° $\langle 10\bar{1}0 \rangle$ symmetrical tilt boundary were used (Fig. 1). A bicrystalline plate was grown from the melt in a boat consisting of a polished graphite plate and mica flanges by the horizontal Bridgman method in an argon atmosphere. Specimens were spark cut from a bicrystalline plate at an angle of 45° (type I specimen) and 60° (type II specimen) with respect to the boundary oriented parallel to the long side of bicrystalline plate. In the type I specimen the basal plane was oriented either normally or in parallel to its axis (Fig. 1(a) and (b)). In the type II specimen basal planes show a 15° ± 0.5° deviation from the position of basal planes in type I specimen with reference to the specimen axis. Dimensions of type I and type II bicrystalline specimens were $30 \times 4 \times 2$ and $25 \times 4 \times 2$ mm^3 respectively. The damaged layer adjacent to the surfaces was removed by chemical polishing on the acid-resistant cloth. Final polishing was performed electrolytically. The experiments on magnetic annealing were carried out using a resistive, steady-state 27 T Bitter magnet with a 52 mm bore diameter. The bicrystals were annealed preliminary without field at a temperature of 663 K during 20 min and were removed from the furnace for the surface observations. Subsequently, they were annealed in the field with strength of $H = 1.35 \times 10^7$ A/m at the temperature of 663 K during 5 min. The ramping rate of the magnet was 20 T/min. Type I specimens were oriented along the field (Fig. 1(c)). Type II specimens were inclined at 15° ± 1° with respect to the field (Fig. 1(d)). Those different orientations make the hexagonal axes of grain A and grain B parallel and normal to the field, respectively. Additionally, specimens of both types were annealed at the same temperature without magnetic field during 100 h. The annealing was interrupted after different periods for the surface observations.

3. Results

Optical micrographs of bicrystals subjected to magnetic annealing and annealing without field are shown in Fig. 2. The preliminary annealing of specimens without field during 20 min results in some displacement of the boundary ends near the lateral surfaces, which can be determined by the

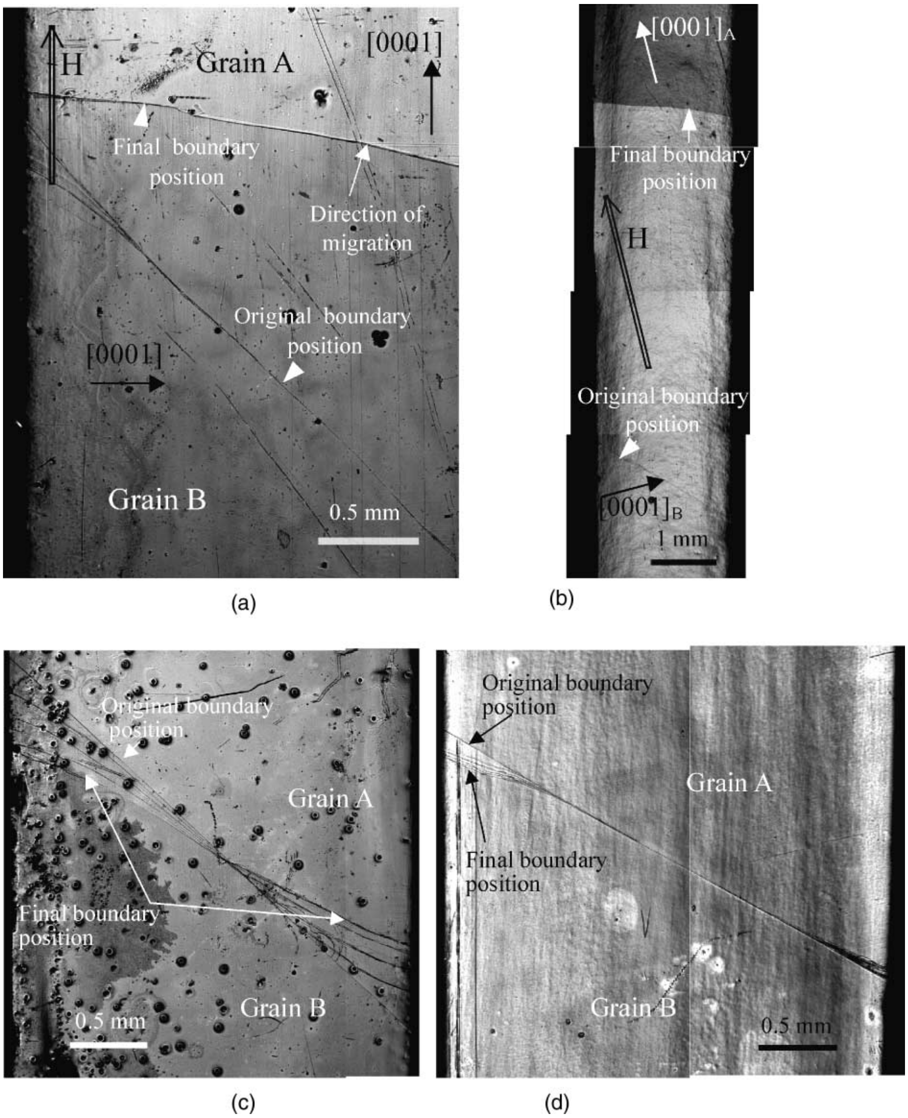

Fig. 2. Grain boundary displacement in Zn bicrystals of type I (a, c) and type II (b, d) during annealing in the field (a, b) and without the field (c, d) at $T = 663$ K. Magnetic annealing in the field of 1.35×10^7 A/m during 5 min (a, b) and annealing without magnetic field during 100 h (c, d). A few successive boundary positions are revealed by the interruption of the process for the surface observations.

position of a boundary groove. In contrast, during 5 min of magnetic annealing boundaries migrate towards grain A in the middle of specimen I for a distance of about 1.2 mm and in specimen II for a distance of 5.5 mm (Fig. 2(a) and (b)). Despite the same magnetic field the migration distance for type II bicrystal is much higher than for type I bicrystal. Also, during migration the boundaries change their orientation in such a way that boundary length decreases approximately by 29% for type I bicrystal and 9% for type II bicrystal. The orientation of boundary line comes close to the normal direction to the lateral surfaces. These effects are observed on both sides (front and back) of bicrystalline samples. The position of boundaries after magnetic annealing (Fig. 2(a) and (b)) can be specified by an angle of $\psi = (\theta_B - \theta_A)/2$ which characterizes a deviation of boundary plane from its symmetry position. For the type I specimen the magnitude of ψ is measured to be about $\psi_I \approx 36°$, whereas the final position of the boundary in the type II specimen is "less" asymmetric, namely $\psi_{II} \approx 25°$.

Fig. 2(c) and (d) illustrates boundary migration and reorientation after annealing without magnetic field during 100 h. The boundary migration starts on the lateral surfaces spreading to the central part of bicrystals. It is seen that the angle of boundary reorientation is lower than that in the case of annealing in the field and it takes much longer time to reorient in such a way. For type I bicrystals the boundary reorientation rate during annealing without the field is more than two orders of magnitude lower than that in the field. In type II bicrystals migration stops near the lateral surfaces after 5 h of annealing. Further annealing during 100 h does not make any marked change in boundary position and orientation.

4. Discussion

Annealing of investigated specimens without field results in boundary reorientation or rotational migration mainly near the lateral surfaces. Fig. 2(c) and (d) illustrates this effect after annealing during 100 h. It is worth to note, that the boundary element, situated in the middle of the boundary length, does not move in the longitudinal direction, but only rotates about the axis perpendicular to the specimen plane. The motion of our boundaries in this case corresponds to the well known bicrystal technique (so called reversed-capillary technique) for measuring the grain boundary motion applying the capillarity (reduction of boundary energy with displacement) as the driving force for grain boundary migration [7–9]. The direction of the boundary motion is normal to the boundary towards to the center of curvature. The driving force p_c is given by $p_c = \sigma k = \sigma/R$, where σ is the surface tension of the grain boundary, k the curvature, and R the radius of curvature. In the case of such geometry, and under the assumption of shape invariance during migration, the curvature is inversely proportional to the distance from the vertex of α in Fig. 1(a) and (b) and the driving force decreases with increasing boundary displacement. The capillary driving force at the left and right sides of our specimens (Fig. 2) acts in opposite directions with respect to the specimen coordinate system (Fig. 3), effectively rotating the boundary about its middle point. The boundary with initial vertex $\alpha = 45°$ (type I spec-

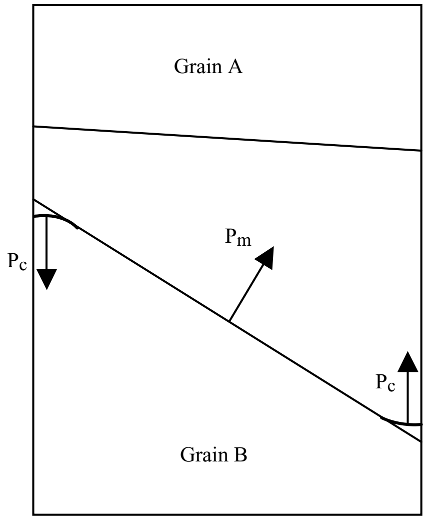

Fig. 3. Schematic of boundary migration and reorientation under the simultaneous action of magnetic and capillarity driving forces.

imen) experiences higher initial driving force, than the boundary of the type II specimen (initial $\alpha = 60°$), moves faster and, hence, covers larger distance from initial to final position (Fig. 2(c) and (d)), before the driving force becomes small being unable to move the boundary. Obviously, this capillary driving force acting on the boundaries in their final positions is almost the same for both specimens, that manifests itself in a very similar position of boundaries with respect to the lateral specimen surfaces after annealing (Fig. 2(a) and (b)).

In contrast, the relatively short (5 min) annealing of the specimens in a high magnetic field leads to much larger reorientation of boundary plane, especially for type II bicrystals (Fig. 2(a) and (b)), and to considerable movement of the boundary in the direction of grains A or, in other words, to the growth of grains B at the expense of grains A. This effect can be understood in terms of magnetic driving force for boundary migration created by the anisotropy of the magnetic susceptibility in Zn. In the case of zinc bicrystals, Eq. (1) can be transformed to:

$$p_m = \mu_0 \frac{\Delta\chi}{2} H^2 (\cos^2 \phi_A - \cos^2 \phi_B), \qquad (2)$$

where ϕ_A and ϕ_B are the angles between the direction of magnetic field and the hexagonal (or c or $\langle 0001 \rangle$ axis) in both neighboring grains, $\Delta\chi$ is the difference in susceptibilities parallel and perpendicular to the hexagonal axis. The force p_m is directed towards the grain with smaller value of ϕ which is grain A in the case of investigated bicrystals. According to [10] the gram susceptibility of Zn parallel and perpendicular to hexagonal (or c or $\langle 0001 \rangle$) axes are $\chi_\parallel = -0.190 \times 10^{-6}$ cm^3/g and $\chi_\perp = -0.145 \times 10^{-6}$ cm^3/g, respectively. Conversion of these data from Gauss unit system gives for volume susceptibility in SI units as $\chi_\parallel = -1.695 \times 10^{-5}$ and $\chi_\perp = -1.294 \times 10^{-5}$. According to Eq. (2) the magnetic driving force for grain boundary migration in Zn bicrystals ($\Delta\chi = \chi_\perp - \chi_\parallel = 0.401 \times 10^{-5}$) in the field of 1.35×10^7 amounts to 0.46 kJ/m^3.

During annealing in the field the boundary experiences simultaneous action of two different driving forces for boundary migration: magnetic driving force which moves boundary in the direction of grain A and capillary driving force, which acts in the direction of grain B on the left half of specimen and in the direction of grain A on its right half (Fig. 3). Therefore, the effective driving force for boundary motion on the left specimen side is $p_{\mathrm{eff}} = p_m - p_c$ and on its right half $p_{\mathrm{eff}} = p_m + p_c$. The action of capillary driving forces in the opposite directions at the boundary ends combined with magnetic driving force reorients the boundary decreasing its length (Figs. 2 and 3), while the magnetic force acting in the direction of grain A is mostly responsible for the boundary displacement in the longitudinal direction. As we have seen (Fig. 2) the boundary element in the middle of its length does not move in the longitudinal direction under the capillary driving force. Therefore, in experiment with a magnetic field this element can be considered as moving under the action of a magnetic force p_m only, and its displacement can be used for the measuring the grain boundary mobility.

The measurement of boundary motion under a constant magnetic driving force provides a unique opportunity to determine the absolute value of grain boundary mobility, which is given by the ratio of velocity v and driving force p_m, $m = v/p_m$. It is worth to note that in experiments with curved grain boundary only the reduced mobility $A = m\sigma$, where σ is the not exactly known boundary surface energy, can be obtained. The mobility of the investigated 88.7° $\langle 10\bar{1}0 \rangle$ tilt boundary in Zn bicrystals was found to be $m_I = 8.8 \times 10^{-9}$ m^4/J s for type I specimen and $m_{II} = 4.1 \times 10^{-8}$ m^4/J s for type II specimen. For comparison, the reduced mobility of the curved 86° $\langle 10\bar{1}0 \rangle$ tilt boundary $A = m\sigma$ at 673 K in Zn bicrystal was measured to be $A_{\mathrm{Zn}}^{673\ \mathrm{K}} = 3.2 \times 10^{-8}$ m^2/s [11], and absolute mobility (assuming $\sigma \sim 0.46$ J/m^2 [12]) is $m_{\mathrm{Zn}}^{673\ \mathrm{K}} \cong 7.0 \times 10^{-8}$ m^4/J s.

The remarkable difference in measured boundary mobility in types I and II specimens can be understood as a consequence of the different inclination of the boundary planes to their symmetrical position. As it was already noted, the final boundary position after magnetic annealing in both investigated specimens is asymmetrical, with different inclination from initial symmetrical position, $\psi_{\mathrm{I}} \approx 36°$ and $\psi_{\mathrm{II}} \approx 25°$, respectively. It was shown by Molodov et al. [3] that in bismuth bicrystals asymmetrical boundaries have lower mobility than symmetrical ones. Therefore, it is reasonable to assume that the tilt boundary with a higher degree of asymmetry moves slower, that is actually observed in the current experiment.

5. Conclusions

The grain boundary in zinc bicrystals was moved by means of annealing in a high magnetic field of 17 T. Under the common action of capillary and magnetic driving forces the initially planar symmetrical 89° $\langle 10\bar{1}0\rangle$ tilt boundary migrates reorienting its plane along the whole length nearly perpendicular to the lateral surfaces.

The mobility of 89° $\langle 10\bar{1}0\rangle$ tilt boundary in bicrystals of Zn at 663 K was determined to be about 2.5×10^{-8} $\mathrm{m^4/J\,s}$.

The mobility of grain boundaries with the same misorientation but different boundary orientations was found to be far different from each other depending on the degree of boundary asymmetry, namely, the boundary with larger inclination with respect to symmetrical boundary position moves slower.

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