Boundary migration in Zn bicrystal induced by a high magnetic field

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A bicrystal of Zn with an originally flat 89° $\langle 10\bar{1}0\rangle$ symmetric tilt boundary was annealed in a magnetic field of 25 T. The boundary migrated under the action of a magnetic driving force in the direction of the grain with higher diamagnetic susceptibility. In addition, the boundary changed its crystallographic orientation, decreasing length and becoming almost perpendicular to the free surfaces. The results were interpreted in terms of magnetically forced grain boundary motion due to the anisotropy of the magnetic susceptibility in Zn. The absolute boundary mobility was measured to be about 5.1×10^{-9} $m^4/J\,s$. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1572536]

Magnetically induced grain boundary migration has been established for diamagnetic bismuth[1–3] and zinc.[4,5] It has been shown that planar boundaries in bismuth bicrystals migrate macroscopically in high fields, whereas no migration has been observed without the field. A relatively small effect of magnetic field on boundary migration has been documented in zinc bicrystals with a 86.0° $\langle 11\bar{2}0\rangle$ boundary,[4] which is close to coincident $\Sigma=13$ boundary.[6] Recently, it has been demonstrated that annealing of polycrystalline Zn–1.1%-Al alloy in a high magnetic field can result in a drastic change of crystallographic texture.[5] This change is also related to magnetically induced grain boundary migration. In magnetically anisotropic materials, the additional driving force for boundary migration or grain growth is exerted by the difference in the magnetic free energy in neighboring grains oriented differently 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 independent of crystal shape and size (the condition for this is $\chi \ll 1$), 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 magnetic field H. Although in most of the anisotropic diamagnetic materials, the difference $\Delta\chi=\chi_1-\chi_2$ is significantly smaller than in bismuth, the effect of the field can be predicted for a number of materials due to the availability of high magnetic fields and the existence of boundaries with high mobility (a proportionality factor between velocity and driving force). This effect has been demonstrated for zinc,[4,5] whose $\Delta\chi$ is about one tenth that for bismuth.[7,8] The boundary in zinc bicrystals, in which migration under the field has been observed recently,[4] is related to vicinal type.[9] However, the behavior of individual boundaries of general type remains unexplored.

In the current work, the effect of a 25-T field on the behavior of 89° $\langle 10\bar{1}0\rangle$ boundary in zinc bicrystal was studied.

Zinc bicrystal (99.995%) containing a $88.7\pm 0.5°$ $\langle 10\bar{1}0\rangle$ symmetrical tilt boundary was used [Fig. 1(a)]. A bicrystalline plate was grown by the horizontal Bridgman method from molten Zn in a boat consisting of a polished graphite plate and mica flanges in an argon atmosphere. Specimens were cut from a bicrystalline plate at an angle of 60° with respect to the boundary, using an electrical discharging machine. Basal planes show a $15\pm 1°$ deviation from the parallel and perpendicular directions to the long side of bicrystal. The damaged layer adjacent to the surfaces was removed by chemical polishing on an acid-resistant cloth. Final polishing was performed electrolytically. The experiments on magnetic annealing were carried out using a

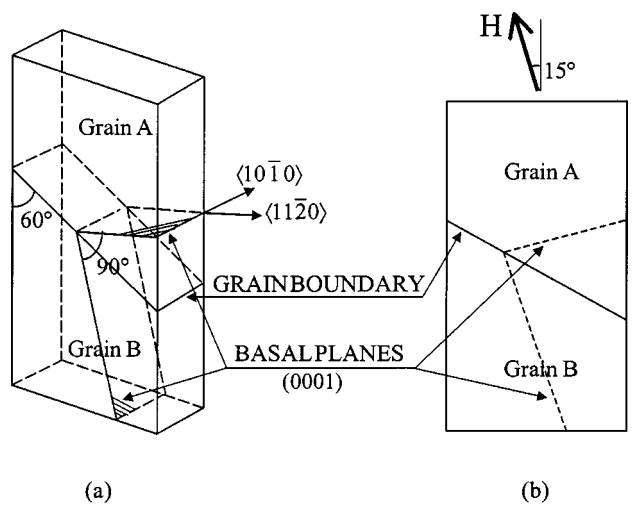

FIG. 1. Geometry of bicrystal (a) and its orientation to the field (b).

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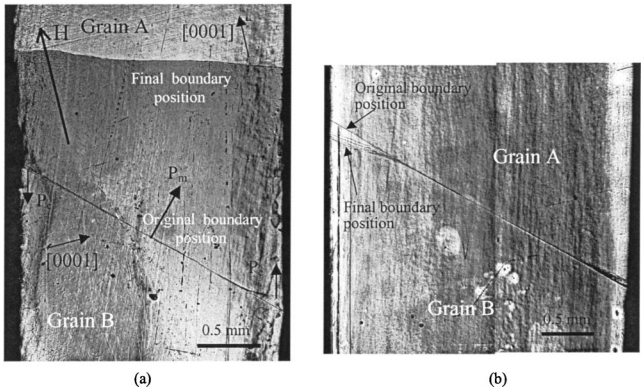

FIG. 2. Grain boundary displacement in Zn bicrystals after annealing in the field of $H=1.99\times10^7$ A/m during 5 min (a) and with no field for 100 hours (b) at $T=663$ K. A few successive boundary positions during annealing with no field were revealed by the interruption of the process for the surface observations.

resistive, steady-state, 27-T Bitter magnet with a 52-mm bore diameter. The bicrystal was first annealed with no field at a temperature of 663 K for 20 min and was then removed from the furnace for surface observations. Subsequently, it was annealed in a field with a strength of $H=1.99\times10^7$ A/m at a temperature of 663 K for 5 min. The specimen was inclined at $15\pm1°$ with respect to the field H [Fig. 1(b)], making the hexagonal axes of grains A and B almost parallel and normal to the field. Additionally, the same type of specimen was annealed at the same temperature without a magnetic field for 100 h. The annealing was interrupted after different periods for the surface observations.

Optical micrographs of bicrystals subjected to magnetic annealing and annealing with no field are shown in Fig. 2. The preliminary annealing of the bicrystal with no field for 20 min resulted in some displacement of the boundary ends near the lateral surfaces, which can be determined by the position of a boundary groove. In contrast, during magnetic annealing for 5 min, the whole boundary migrated towards grain A (Fig. 2). The distance of migration varied from one end of the boundary to the other ranging from 0.9 to 1.8 mm. The distance of migration in the middle of the boundary is equal to 1.54 mm. This was determined by averaging the measurements of migration in the middle on the front and back surfaces of the sample. Also, during migration the boundary changed its orientation in such a way that the boundary length decreased by approximately 9%. The orientation of the boundary line became almost perpendicular to the lateral surfaces. This effect was observed both on front and back surfaces of the bicrystalline samples. The position of the boundary after magnetic annealing [Fig. 2(a)] can be specified by an angle of $\psi=(\theta_B-\theta_A)/2$, which characterizes a deviation of boundary plane from its symmetry position. The magnitude of ψ is measured to be about 25°.

Figure 2(b) illustrates boundary migration and reorientation after annealing with no magnetic field for 100 h. The boundary migration started on the lateral surfaces, spreading gradually to the central part of the bicrystal. It can be seen that the angle of boundary reorientation is lower than that in the case of annealing in the field, and it takes much longer to reorient in such a way. The boundary migration rate during annealing with no field is more than two orders of magnitude lower than that in the field, and the migration stopped after 5 h of annealing. Further annealing for 95 h made almost no change in the boundary position and orientation.

Annealing of specimens with no magnetic field resulted in boundary reorientation or rotational migration mainly near the lateral surfaces [Fig. 2(b)]. It is worth noting that the boundary element, situated in the middle of the boundary length, did not move in the longitudinal direction and did not rotate. Only the peripheral parts of the boundary rotated about the axis perpendicular to the specimen plane. The motion of the boundary in this case corresponds to the well-known bicrystal technique (so called reversed-capillary technique) for measuring the grain boundary motion by applying the capillarity (reduction of boundary energy with displacement) as the driving force for grain boundary migration.[10–12] 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 $p_c=\sigma\cdot k=\sigma/R$, 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 the driving force decreases with increasing boundary displacement. Therefore, the boundary migration did not reach the center of the boundary and no rotational migration of the central part of the boundary was observed. The capillary driving force at the left and right sides of our specimens (Fig. 2), acts in opposite directions, rotating the peripheral portions of the boundary counterclockwise.

In contrast, the relatively short (5-min) annealing of the specimen in a high magnetic field leads to much larger reorientation of the boundary plane [Fig. 2(a)] and to considerable movement of the boundary in the direction of grain A or, in other words, to the growth of grain B at the expense of grain 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

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

where ϕ_A and ϕ_B are the angles between the direction of the magnetic field and the hexagonal (or c or $\langle0001\rangle$) axis in both neighboring grains, and $\Delta\chi$ 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 the investigated bicrystal. According to MacClure and Marcus,[8] the gram susceptibilities of Zn parallel and perpendicular to hexagonal (or c or $\langle0001\rangle$) axes are $\chi_{II}=-0.190\times10^{-6}$ cm^3/g and $\chi_\perp=-0.145\times10^{-6}$ cm^3/g. Conversion of this data from the Gauss unit system to SI gives us volume susceptibilities as $\chi_{II}=-1.695\times10^{-5}$ and $\chi_\perp=-1.294\times10^{-5}$. According to Eq. (2), the magnetic driving force for grain boundary migration in Zn bicrystal in a field of 1.99×10^7 A/m, and with a difference in volume susceptibilities $\Delta\chi=\chi_\perp-\chi_{II}$ of 0.401×10^{-5} is 1.00 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 the 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. 2). 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 the magnetic driving force reorients the boundary, decreasing its length, while the magnetic force acting in the direction of grain A is mostly responsible for the boundary displacement in the longitudinal direction (Fig. 2). It is easy to see that the middle of the boundary does not experience an action of the capillary driving force, since the capillary forces acting in the opposite directions compensate each other at that point. Therefore, in experiment with a magnetic field, the center of the boundary is moving under the action of the magnetic force p_m only, and its displacement can be used for 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 noting that in experiments with curved grain boundary, only the reduced mobility $A=m\cdot\sigma$, 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 bicrystal was found to be $m=5.1\times10^{-9}$ $\mathrm{m^4/J\,s}$. For comparison, the reduced mobility of the curved 86° $\langle 10\bar{1}0\rangle$ tilt boundary $A=m\cdot\sigma$ at 673 K in Zn bicrystal was measured to be $A_{\mathrm{Zn}}^{673\ \mathrm{K}}=3.2\times10^{-8}$ $\mathrm{m^2/s}$,[13] and absolute mobility (assuming $\sigma\sim0.46$ $\mathrm{J/m^2}$[14]) is $m_{\mathrm{Zn}}^{673\ \mathrm{K}}\cong7.0\times10^{-8}$ $\mathrm{m^4/J\,s}$.

In conclusion, the individual grain boundary in zinc bicrystal was moved under high magnetic field in the direction of grain with higher diamagnetic susceptibility. It was demonstrated that under the simultaneous 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. Thus, the absolute mobility of a grain boundary in Zn bicrystal was determined. The absolute mobility of 89° $\langle 10\bar{1}0\rangle$ tilt boundary at 663K is about 5.1×10^{-9} $\mathrm{m^4/J\,s}$.

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