MgB_2 films with very high critical current densities due to strong grain boundary pinning

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MgB_2 superconductor has a great potential for applications because of its high T_c and B_{c2}, exceeding those of any Nb-base superconductors at any temperature. It is now important to understand its flux pinning so as to raise J_c to high values over a wide field range. We show that nanometer-sized columnar-grain structure can produce J_c exceeding 5×10^6 A/cm^2. The angular dependence of J_c indicates that the strongest pinning occurs when the field is aligned parallel to the grain boundaries. Our results confirm earlier deductions that grain boundaries in MgB_2 act as effective pinning centers like those in Nb_3Sn. © *2004 American Institute of Physics*. [DOI: 10.1063/1.1805195]

Magnesium diboride (MgB_2) superconductor[1] has a great potential for applications because of its high transition temperature and upper critical fields (B_{c2}) that can exceed those of any Nb-base superconductors at any temperature. Moreover, many groups[2–17] are making films and wires of MgB_2. In order to realize the full potential of MgB_2, it is now important to understand its flux pinning so as to raise the critical current density (J_c) to high values over a wide field range. In this letter we show conclusively that 20–30 nm diameter columnar grains can produce J_c values exceeding 5×10^6 A/cm^2. By studying the angular dependence of J_c in films where the grains grow at a small angle to the substrate, we conclusively show that the strongest pinning occurs when the field is aligned parallel to the grain boundaries, even though B_{c2} is lower for this field orientation than when parallel to the Mg and B planes. Our results confirm earlier deductions[2–4] that grain boundaries in MgB_2 do not obstruct current flow as in the high-temperature cuprate superconductors, but act like those in intermetallic superconductors such as Nb_3Sn.

Intensive research work has been performed to fabricate MgB_2 thin films[5–17] with high J_c. For example, J_c values reaching 10^7 A/cm^2 at 4.2 K in zero external fields are reported.[5–13] However, magnetic field dependence of J_c is large, and J_c decreases to about 10% and 1% of self-field values in 1–3 and 4–5 T, respectively, even at low temperatures in most cases.[5–8,14,15] Further understanding and improvements in flux pinning are essential to realize J_c enhancement in fields both for thin films and wires.

We prepared MgB_2 thin films on a polished sapphire C (0001) single-crystal substrate by using electron-beam evaporation technique. We controlled the background pressure to less than 10^{-6} Pa and the substrate temperature to 543(±5) K. The evaporation flux ratio of Mg was set at four times as high as that of B, and the growth rate of the MgB_2 layer was 1 nm/s. We prepared two films with different thicknesses of MgB_2 layer of 0.30 and 0.15 μm. No heat treatment was performed after the deposition. Therefore, we examined their properties in the "as-grown" state. For the transport measurements, the films were patterned by dry-etching and electron cyclotron resonance etching processes. Both the width and length of the strip line were 1 mm. We then deposited 0.20-μm-thick gold layers for electrodes. We performed the dc four-probe transport measurements for the patterned specimens. We measured critical currents I_c as a function of the external magnetic fields B and the angle θ of the fields in liquid helium (4.2 K). Current direction was set perpendicular to the field direction and the specimen was rotated around this current axis. In this work, we define $\theta=0°$ and 90° as the fields parallel and perpendicular to the film surface (hereafter referred to as the parallel field and the perpendicular field), respectively. We employed the 1 μV/cm criterion for the I_c determination. We observed and analyzed the microstructure by transmission electron microscopy (TEM). We prepared cross-sectional foil specimens by using a focused-ion-beam instrument equipped with a micro-sampling system and performed the observation with an electron microscope accelerated at 200 kV.

Figure 1 shows J_c–B relations at 4.2 K for the films with (a) 0.30-μm-thick and (b) 0.15-μm-thick MgB_2 layers. J_c–B curve for $\theta=90°$ stays higher than that for $\theta=0°$ in the 0.30-μm-thick film. It is notable that J_c exceeds 10^6 A/cm^2 even in the perpendicular field of 4 T. On the contrary, J_c–B curve for $\theta=90°$ stays lower than that for $\theta=0°$ in the 0.15-μm-thick film. Many research papers report that B_{c2} is higher in

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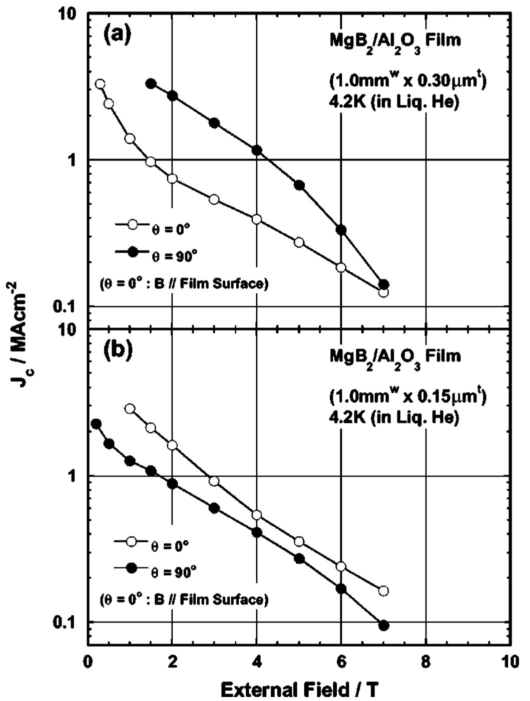

FIG. 1. J_c plots as a function of external magnetic fields for (a) 0.30-μm-thick and (b) 0.15-μm-thick MgB_2 layers at 4.2 K. $\theta=0°$ and 90° correspond to the fields parallel and perpendicular to the film surface, respectively. Open and solid circles represent J_c values at $\theta=0°$ and 90°, respectively. J_c values exceeding $5\times10^6\ A/cm^2$ in low fields could not be measured because of a thermal problem due to heat formation at the current contacts.

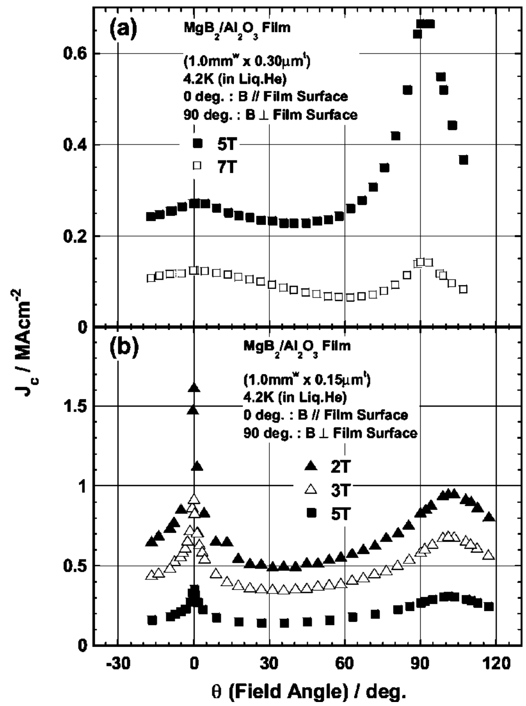

FIG. 2. J_c plots as a function of θ (angle of the fields against the film surface) for (a) 0.30-μm-thick and (b) 0.15-μm-thick MgB_2 layers at 4.2 K $\theta=0°$ and 90° correspond to the fields parallel and perpendicular to the film surface, respectively. Open and solid squares and open and solid triangles represent J_c values in 7, 5, 3, and 2 T, respectively.

the parallel field than in the perpendicular field.[5–8,14] Although the large decrease of J_c in the perpendicular high fields [see Fig. 1(a)] may be attributed to the reported angular dependence of B_{c2}, the origin of the higher J_c around $\theta=90°$ in this work cannot be explained simply in terms of the angular dependence of B_{c2}. B_{c2} (90% of normal-conducting state resistivity) of the 0.30-μm-thick film are estimated to >28 and 17 T at 4.2 K in the parallel and the perpendicular fields, respectively.

Figure 2 shows J_c–θ relations at 4.2 K for the films with (a) 0.30-μm-thick and (b) 0.15-μm-thick MgB_2 layers. The J_c–θ curves have two peaks around $\theta=0°$ and $\theta=90°$ in both films. J_c values are much higher around $\theta=90°$ than around $\theta=0°$ in the 0.30-μm-thick film. However, J_c is higher around $\theta=0°$ than around $\theta=90°$ in the 0.15-μm-thick film corresponding to the results in J_c–B relations. In addition, the peak positions vary in the two films ($\theta=92°$ and 102° for 0.30- and 0.15-μm-thick films, respectively), whereas both films show the peak at $\theta=0°$. It is remarkable that the characters of the peaks for the two films are much different. The peak around $\theta=0°$ appears rounded and the peak around $\theta=90°$ is more cusplike in the 0.30-μm-thick film. In contrast, for the 0.15-μm-thick film the peaks show the opposite character.

The microstructure of the MgB_2 layer gives the origin of the peak around $\theta=90°$. Figure 3 shows TEM bright- and dark-field images (BFI and DFI, respectively) of the cross section along with a corresponding diffraction pattern. Columnar grain growth of MgB_2 can be clearly seen in the micrographs. The columnar grains of 20–30 nm in size grow in almost perpendicular to the MgB_2/substrate interface. The diffraction pattern indicates that the MgB_2 layer has a textured microstructure and that c axes of MgB_2 crystals align with the columnar growth direction. The grain boundaries between these columnar grains may work as effective pinning centers and lead to the high J_c performance in the perpendicular fields.

These results suggest that the MgB_2 layer has two pinning mechanisms. One corresponds to the peak around $\theta=90°$ originating in grain boundaries in the columnar microstructure. The other, corresponding to the peak at $\theta=0°$, may originate in the surface of the MgB_2 layer, the MgB_2/substrate interface, or the layer structure of MgB_2 crystals. Figure 4 shows the explanation for these two pinning mechanisms and the two peaks in J_c–θ relations. This indicates that the pinning becomes strong when the field is aligned parallel to the grain boundaries. The detailed observation of the micrographs indicates that the direction of the columnar grains is shifted several degrees from the perpendicular direction. This agrees with the results that the peak position in J_c–θ

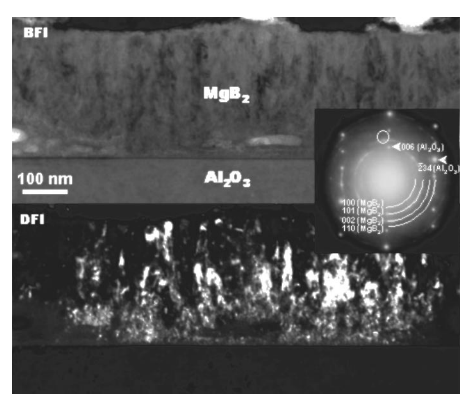

FIG. 3. TEM micrographs of the cross section of the film. (BFI and DFI, images) and a corresponding diffraction pattern are shown. The incident beam direction is [320] of the sapphire substrate.

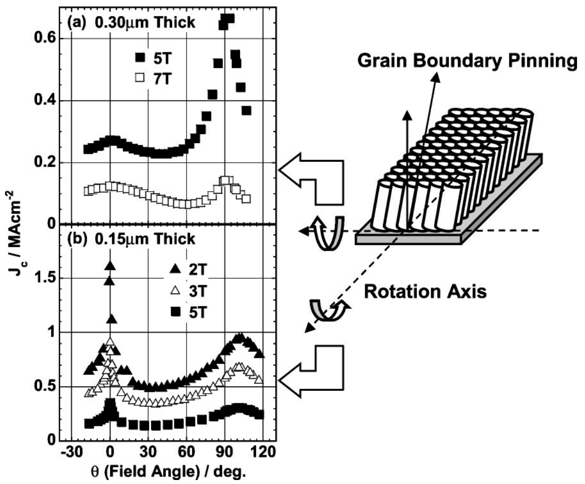

FIG. 4. Explanation for the relation between grain boundary pinning and J_c–θ profiles. J_c–θ profiles for (a) 0.30-μm-thick and (b) 0.15-μm-thick MgB_2 layers are the same as shown in Fig. 2. The difference of the peak position between the two films can be attributed to the different geometric arrangement of the columnar growth direction and the rotation axis in the measurement.

curves is shifted from the perpendicular position (see Fig. 2). We think that the direction of columnar grains growth is shifted 10°–15° from the direction perpendicular to the film surface, reflecting the geometric arrangement of the evaporation source and the substrate during the deposition process. The difference of the peak position between the two films originates in the different geometric arrangement of the columnar growth direction and the rotation axis in the measurement. J_c enhancement due to the grain boundary pinning decreases with decreasing the thickness of MgB_2 layer, comparing the peaks around $\theta=90°$. On the contrary, pinning potential in the parallel fields is not sensitive to the thickness of the MgB_2 layer. The difference of the current carrying capacity corresponding to the peak at $\theta=0°$ between the 0.15- and the 0.30-μm-thick films is small. For example, I_c values in 1.5 T are 3.19 and 2.90 A for the 0.15- and the 0.30-μm-thick films, respectively (see Fig. 1). This indicates that the mechanism and the strength of the pinning in the parallel field are similar and independent of the layer thickness. It supports the fact that the origin of pinning in the parallel field may be the surface of MgB_2 layer and/or the MgB_2/substrate interface as described before. It suggests that the volume pinning is much weaker than the surface pinning. These two pinning mechanisms also give an explanation for the different characters of the peaks in J_c–θ relations described before (see Fig. 2). The very sharp peak around $\theta=0°$ in the 0.15-μm-thick film may originate in the strong surface pinning due to very flat surface and/or interface. The broad peak around $\theta=0°$ in the 0.30-μm-thick film suggests that the surface and/or the interface in the 0.30-μm-thick film is less flat. On the other hand, the increase of thickness of the MgB_2 layer increases the pinning due to the grain boundaries and makes the peak around $\theta=90°$ more cusplike.

In summary, we can conclude based on our results that the grain boundaries between columnar grains work as effective pinning centers and lead to the high J_c performance in the perpendicular fields.

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