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Enhancement of J_c in MgB_2 thin films on Si substrate with pinning centers introduced by deposition in O_2 atmosphere

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As-grown MgB_2 thin films on Si substrates with high J_c under magnetic fields were prepared by electron-beam evaporation. The value of J_c has been enhanced by the deposition of MgB_2 thin film in an O_2 atmosphere. The MgB_2 thin film deposited in the O_2 atmosphere (O_2 -doped film) has exhibited considerably higher J_c in magnetic fields among MgB_2 thin films reported before. It has been found that the high J_c of the O_2 -doped film is attributable to the flux pinning with grain boundaries strengthened by an introduction of MgO along grain boundaries. In a high magnetic field, a peculiar behavior of E - J characteristics where E - J curves vary in two stages was observed. This behavior also originates from the flux pinning with strengthened grain boundaries. © 2007 American Institute of Physics. [DOI: [10.1063/1.2794722](https://doi.org/10.1063/1.2794722)]

Much effort has been put into the development aiming for high current applications and electronic devices using a magnesium diboride (MgB_2) since the highest critical temperature T_c of 39 K among metallic superconductors was discovered.¹ In addition to the high T_c , MgB_2 has attractive features such as a simple crystal structure, a long coherent length, and low material costs.

For electronic devices, high quality as-grown MgB_2 thin films prepared by low temperature processes such as molecular-beam epitaxy²⁻⁴ (MBE) and sputtering⁵ are needed. Although as-grown thin films with high T_c and high critical current density J_c had not been obtained in early years since the discovery of MgB_2 ,⁶ as-grown thin films with high J_c prepared by MBE and electron-beam evaporation (EBE) have been reported recently.⁷⁻¹⁰ These thin films prepared by a low temperature process are suitable for fabricating junction and multilayered structure.

Enhancement of J_c is one of the most important topics for practical applications of MgB_2 thin films. It is essential to increase J_c especially for applications in high magnetic fields such as a superconducting probe coil for NMR.

In this paper, MgB_2 thin films with high J_c in magnetic fields have been prepared by EBE. The value of J_c has been enhanced by deposition in an O_2 atmosphere. We have clarified the flux pinning mechanism of the MgB_2 thin films by

the investigation of J_c and electric field versus current density (E - J) characteristics.

MgB_2 thin films were deposited on Si (100) substrates by EBE.⁹ A substrate was set in a deposition chamber with base pressure of 5×10^{-7} Pa and heated at 250 °C by a halogen lamp heater. The flux rates of Mg and B were controlled at 3.0 and 0.35 nm s⁻¹, respectively. The thickness of the film was 280 nm. Moreover, another MgB_2 thin film with artificial pinning centers was prepared. After the base pressure had reached at 5×10^{-7} Pa, the oxygen gas was flowed into the deposition chamber, where the oxygen partial pressure was 8.4×10^{-7} Pa. The flux rates of Mg and B and the substrate temperature were the same with those of the above sample. The thickness of the film was 250 nm.

The T_c values of MgB_2 thin films deposited in the high vacuum and the O_2 atmosphere (hereafter referred to as non-doped film and O_2 -doped film, respectively) were 35.3 and 35.0 K, respectively. The x-ray diffraction analyses showed that both the films were oriented along the c axis. To measure transport properties, the films were patterned into micro-bridge shapes with the strip dimension of 50 μm wide and 1 mm long by photolithography. The dc transport properties in magnetic fields were measured by a four probe method. The direction of the external magnetic field was changed by rotating a sample holder. The angle θ of the magnetic field direction was defined as an angle from the c axis direction. The transport current is always orthogonal to the magnetic field. The temperature was stabilized within ± 0.02 K.

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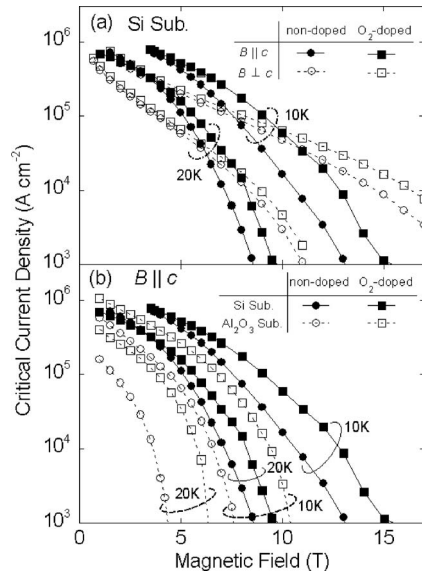


FIG. 1. Magnetic field dependences of J_c : (a) MgB_2/Si thin films in magnetic fields applied parallel and perpendicular to the c axis, and (b) Comparison of J_c between MgB_2/Si and $\text{MgB}_2/\text{Al}_2\text{O}_3$ in the magnetic field applied parallel to the c axis, where circle symbols indicate nondoped films and square symbols indicate O_2 -doped films.

The dependences of J_c on magnetic fields applied in the direction parallel ($B\parallel c$) and perpendicular ($B\perp c$) to the c axis for the MgB_2 thin films deposited on Si substrates are shown in Fig. 1(a). The values of J_c of the O_2 -doped film are higher than those of the nondoped film in both the magnetic field directions. This fact indicates that effective pinning centers have been introduced by the deposition in the O_2 atmosphere. The reduction of J_c with increasing magnetic field for $B\perp c$ is more slowly than that for $B\parallel c$, although the values of J_c for $B\perp c$ is lower than those for $B\parallel c$ in low magnetic fields. This is because that the upper critical magnetic field B_{c2} for $B\perp c$ is higher than that for $B\parallel c$ due to the anisotropic crystal structure of MgB_2 .

Figure 1(b) shows a comparison of magnetic field dependences of J_c between the present MgB_2 thin films deposited on Si substrates (MgB_2/Si) and MgB_2 thin films deposited on Al_2O_3 substrates ($\text{MgB}_2/\text{Al}_2\text{O}_3$) reported previously.¹⁰ The values of J_c of MgB_2/Si exceed those of $\text{MgB}_2/\text{Al}_2\text{O}_3$ greatly.

It has been reported that an amorphous layer is formed at the interface between the Al_2O_3 substrate and the MgB_2 layer in $\text{MgB}_2/\text{Al}_2\text{O}_3$, because the Al_2O_3 substrate would not heat up immediately to desired temperature due to the transparency of Al_2O_3 in the early stage of MgB_2 formation by EBE using a lamp heater.¹¹ The amorphous layer causes a degradation of the current carrying capacity. In case of the Si substrate, on the other hand, it has been confirmed by transmission electron microscopy (TEM) observations that MgB_2 grains grow immediately above the substrate. The O_2 -doped film deposited on the Si substrate has exhibited substantially high J_c in the magnetic fields exceeding J_c for MgB_2 thin films prepared by carbon doped hybrid physical-chemical vapor deposition and MgO incorporating *ex situ* annealing method in which these MgB_2 thin films have higher J_c among MgB_2 thin films reported so far.^{12,13}

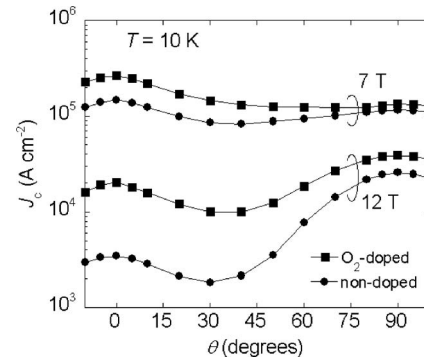


FIG. 2. Angular dependence of J_c at 10 K, on $B=7$ and 12 T.

To investigate the anisotropy of flux pinning properties in high magnetic fields, the angular dependences of J_c have been measured as shown in Fig. 2. Peaks of J_c were observed at $\theta=0^\circ$ ($B\parallel c$) in both of the nondoped and the O_2 -doped films. These peaks are attributable to the grain boundary pinning with columnar growth MgB_2 grains.^{8,10}

The values of J_c of the O_2 -doped film are higher than those of the nondoped film in the whole angle region. This result indicates that isotropic pinning centers which work effectively in every magnetic field direction have been introduced by deposition in the O_2 atmosphere. The isotropic pinning centers are MgO particles which are produced by oxidation of Mg flux at the deposition process. The enhancement of J_c by deposition in the O_2 atmosphere is especially large at $\theta=0^\circ$. This fact indicates that pinning forces of grain boundaries have been strengthened since MgO is introduced not only into grains but also along grain boundaries.

It is important to understand E - J characteristics for the design of superconducting equipments. We measured E - J characteristics in various magnetic fields and temperatures, and evaluated the flux pinning mechanism. The E - J characteristics of the O_2 -doped film are shown in Fig. 3. Figure 3(a) shows E - J characteristics in the low magnetic field of 3 T and $\theta=0^\circ$. An E - J curve at a low temperature has a negative curvature in a log-log plot. As the temperature increases, the curvature of the E - J curve varies from negative to positive on reaching a certain temperature which is called as the vortex glass-liquid transition temperature T_g .¹⁴

Figure 3(b) shows E - J characteristics in the high magnetic field of 8 T and $\theta=0^\circ$. A noticeable behavior of E - J characteristics has been observed where E - J curves vary in two stages. This noticeable behavior has not been observed in the nondoped film even in high magnetic fields. The O_2 -doped film has strong pinning centers which are grain boundaries strengthened by MgO and other weak pinning centers. In low magnetic fields, even weakly pinned fluxoids can not easily escape from pinning centers, because these fluxoids are supported by strong pinning centers with the fluxoid-fluxoid interaction. In high magnetic fields, on the other hand, weakly pinned fluxoids would disengage from pinning centers and flow in a low applied current density region. Subsequently, strongly pinned fluxoids gradually flow as the applied current density increases. Because the fluxoid-fluxoid interaction is weakened due to the reduction

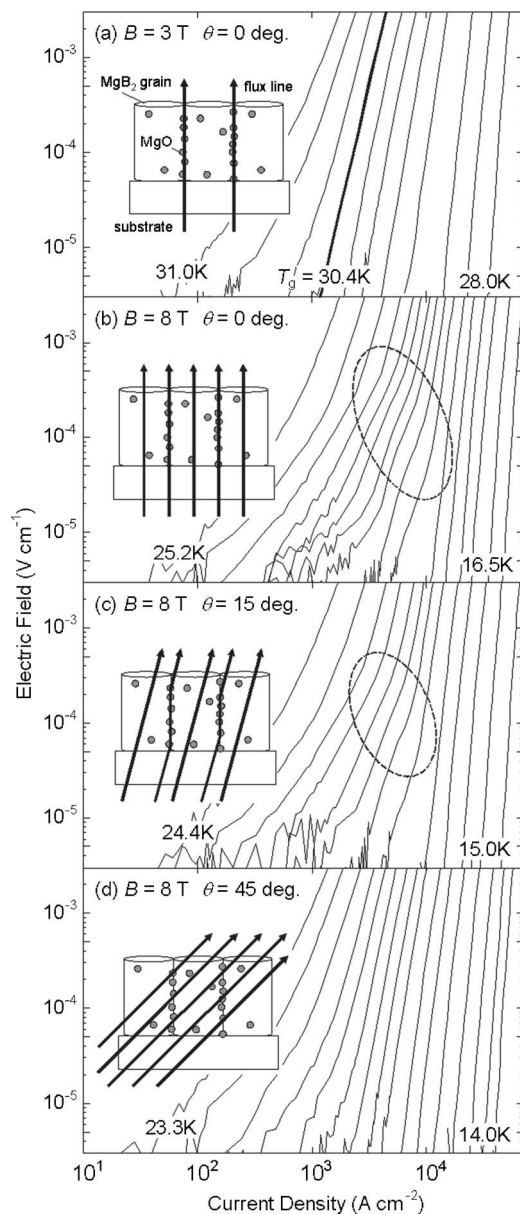


FIG. 3. E - J characteristics of O_2 -doped MgB_2 thin film: (a) $B=3$ T, $\theta=0^\circ$, (b) $B=8$ T, $\theta=0^\circ$, (c) $B=8$ T, $\theta=15^\circ$, and (d) $B=8$ T, $\theta=45^\circ$, where insets show schematic illustration of flux pinning.

of the shear modulus C_{66} with increasing magnetic field. Consequently, E - J curves vary in two stages.

Figures 3(c) and 3(d) show E - J characteristics in $B=8$ T, $\theta=15^\circ$, and $\theta=45^\circ$, respectively. Notice that the kink of E - J characteristics has been small with increasing θ , and the kink has disappeared at $\theta=45^\circ$. The pinning effect by grain boundaries becomes small as the direction of magnetic field c axis direction as shown in the insets of Fig. 3. At θ

$=45^\circ$, all fluxoids may be pinned weakly because the grain boundary pinning dose not work effectively. This remarkable behavior of E - J characteristics is attributable to the grain boundary pinning strengthened by dispersed MgO .

In summary, the as-grown MgB_2/Si thin films were prepared by EBE. We have achieved the improvement of J_c of the MgB_2 thin film under magnetic fields by the deposition in the O_2 atmosphere. The values of J_c of the present MgB_2/Si have been enhanced in comparison with the previous MgB_2/Al_2O_3 . The O_2 -doped film has exhibited considerably higher J_c in magnetic fields among MgB_2 thin films reported before. It has been found that the high J_c of the O_2 -doped film is attributable to the flux pinning with grain boundaries strengthened by the introduction of MgO . In high magnetic fields, we observed the peculiar behavior of E - J characteristics varying in two stages. This behavior originates from the flux pinning with grain boundaries strengthened by dispersed MgO .

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