Flux pinning properties of MgB$_2$ thin films on Ti buffered substrate prepared by molecular beam epitaxy

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Abstract

Transport properties of the MgB$_2$ thin films on Si, MgO and ZnO substrates with Ti buffer layer prepared by molecular beam epitaxy were investigated to clarify effects of the substrates and the Ti buffer layer on flux pinning. The critical current density $J_c$ of each sample shows different dependence on magnetic fields parallel to c-axis. However, the scaling analysis of the macroscopic pinning force for all the measured samples implies that the grain boundaries work as the dominant pinning centers for $B//c$. The pinning parameter for MgB$_2$/Ti/Si estimated from the electric field $E$ vs. the current density $J$ characteristics shows the highest value among all the measured samples. This result is attributed to the high density of grain boundaries caused by the effect of both the Ti buffer and Si substrate in the growth process. Therefore, the selection of substrates and buffer layer strongly affects the flux pinning properties of MgB$_2$ thin films and plays an important role in the determination of performance for superconducting devices and wires.

Keyword: critical current density, Ti buffer layer, grain boundary
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Introduction

MgB$_2$ has the highest critical temperature $T_c$ of 39 K among metallic superconductors [1]. This superconductor has some advantages which are usable in liquid hydrogen, a simple crystal structure, a low material cost and long coherence length. Therefore, MgB$_2$ superconductors are expected to be applied to various fields, such as wires and electronic devices.

High quality MgB$_2$ thin films are prepared by molecular beam epitaxy (MBE) [2] and electron beam evaporation (EBE) [3]. As-grown MgB$_2$ thin films deposited at low temperature by MBE and EBE have a high critical current density $J_c$. It is well known that dominant pinning centers of MgB$_2$ thin films on Al$_2$O$_3$ deposited by MBE and EBE are grain boundaries [4, 5]. The grain boundaries grow up perpendicular to surface in most cases. Then, the grain boundaries are effective pinning centers in magnetic field perpendicular to the surface. For this reason, MgB$_2$ thin films prepared by MBE and EBE show high critical current density $J_c$ in the magnetic field perpendicular to the surface. However, further improvement of $J_c$ in MgB$_2$ thin films are required for the high field application. Therefore, we need to introduce more effective pinning centers to MgB$_2$ thin films or to enhance pinning force of MgB$_2$ grain boundaries.
In this study, the MgB$_2$ thin films were prepared on various Ti buffered substrates.

In order to investigate influence of Ti buffer layer and substrates on flux pinning properties, we measured the magnetic field dependence of $J_c$, field angular dependence of $J_c$ and the electric field $E$ vs. current density $J$ characteristics. The vortex glass-liquid transition temperature $T_g$ and the pinning parameter $m$ were estimated from $E - J$ characteristics. So, we can discuss the flux pinning properties of MgB$_2$ thin films on Ti buffered substrates in the viewpoint of the pinning strength distribution from the magnetic field dependence of $m$.

**Experimental**

MgB$_2$ thin films were deposited on MgO(100), Si(111) and ZnO(001) substrates with Ti buffer layer and MgO substrate without Ti buffer layer by MBE [6]. The substrates were set in the deposition chamber, and heated at 200 °C by tantalum heater. Base pressure of deposition chamber was very low about less or equal to $4 \times 10^{-10}$ Torr. The evaporation sources were a pure magnesium block (99.99%) and stuffed granular boron (99.9%). The flux rate of B was 0.03 nm/s and the flux rate of Mg was from eight to nine-fold of that of B. The thicknesses of MgB$_2$ thin films are 150 nm (MgB$_2$/Ti/MgO, MgB$_2$/Ti/Si) and 300 nm (MgB$_2$/Ti/ZnO, MgB$_2$/MgO) respectively.

The x-ray diffraction analysis shows that the all samples are oriented along $c$-axis. The
Ti buffer layer was deposited on a substrate before the deposition of MgB$_2$, and thicknesses of Ti buffer layers are 50 nm ($\text{MgB}_2$/Ti/MgO, $\text{MgB}_2$/Ti/Si) and 20 nm ($\text{MgB}_2$/Ti/ZnO), respectively. The Ti layer for protection was deposited on surface of MgB$_2$ thin films. The thickness of protection layer is about 3–5 nm. Table 1 summarizes the deposition parameters for each sample.

The lattice mismatching of MgB$_2$ for ZnO and Ti is small with 5.24% and 4.47%, respectively. On the other hand, the lattice mismatching of MgB$_2$ for MgO(100) and Si(111) are very large with 30.8% and 21.78%. However, it has been known that a 45° in-plane rotation of the MgB$_2$ lattice with MgO results in the lattice mismatching of only ~3% for two unit cells of MgB$_2$ on a MgO unit cell [7,8].

In order to measure the transport properties, the samples were patterned into a microbridge shape of 30 µm wide and 1 mm long by photolithography. To obtain good contact, Au contact pads were deposited on the films by RF sputtering after cleaning the film surfaces. The transport properties were measured by a four probe method. The magnetic field was applied by a 20 T superconducting magnet. The angle of external magnetic field was changed by rotating a sample holder in the superconducting magnet. The temperature was stabilized within ±0.03 K. The value of $J_c$ was evaluated from the transport properties with the criterion of the electric field $E_c = 1 \mu \text{Vcm}^{-1}$. A magnetic
field angle is defined such that $B/c$ is $\theta = 0^\circ$ and transport currents are always perpendicular to the $c$-axis and the magnetic fields.

**Results and discussion**

Fig. 1 shows the temperature dependence of resistivity $\rho$. MgB$_2$/Ti/MgO, MgB$_2$/Ti/Si, MgB$_2$/MgO and MgB$_2$/Ti/ZnO have the critical temperatures $T_c$ of 32.7 K, 34.3 K, 33.6 K and 34.9 K, respectively. The resistivity of MgB$_2$/Ti/Si is highest among the all samples. The resistivity of MgB$_2$/Ti/Si is about four times higher than the lowest resistivity of MgB$_2$/Ti/ZnO. Although MgB$_2$/Ti/MgO and MgB$_2$/MgO are the same substrate, the resistivity of MgB$_2$/Ti/MgO is higher than that of MgB$_2$/MgO.

The $B - T$ phase diagram is estimated from the $\rho - T$ curves in the magnetic fields. Fig. 2 shows the temperature dependence of upper critical current field $B_{c2}$ and irreversibility field $B_{irr}$ for MgB$_2$/Ti/MgO and MgB$_2$/Ti/Si. The values of $B_{c2}$ are defined as the mid points of resistivity $\rho$ in the transition regions of $\rho - T$ curves up to 15 T. The $B_{irr}(T)$ curves are determined by the temperatures when the resistivity becomes 0.1% of resistivity immediately before the transition at each magnetic field. $B_{c2}$ and $B_{irr}$ of MgB$_2$/Ti/Si are higher than those of MgB$_2$/Ti/MgO in the high magnetic field. It is considered that the higher $B_{c2}$ in MgB$_2$/Ti/Si is due to the diffusion of Si into the MgB$_2$ thin film. This tendency was also reported by Harada *et al* [9]. We cannot confirm that
Si in the MgB$_2$ thin film becomes the impurity or the second phase material. However, it is considered that the highest resistivity of MgB$_2$/Ti/Si is caused by the diffusion of Si into the MgB$_2$ thin film.

Fig. 3 shows the dependence of $J_c$ on the magnetic fields applied in the direction parallel to the $c$-axis at 20 K. In the low magnetic fields, MgB$_2$/Ti/ZnO shows the highest $J_c$ of all samples. In the magnetic fields from 3 T to 8 T, MgB$_2$/Ti/MgO shows the highest $J_c$ and MgB$_2$/Ti/Si shows the highest $J_c$ over 8 T. The value of $J_c$ of MgB$_2$/Ti/MgO is higher than that of MgB$_2$/MgO in all magnetic fields.

Fig. 4 shows the normalized macroscopic pinning force density, $f = F_p/F_{p_{\text{max}}}$, as a function of the normalized $b = B/B_{\text{irr}}$. The normalized pinning force density is fitted to the expression $f = \beta A B^p (1 - B/B_{\text{irr}})^q$. In Fig. 4, the solid line denotes in the case of $p = 0.9$ and $q = 2.1$. The data of the all samples is fitted to the solid line. When the dominant pinning centers are the grain boundaries, the parameters are $p = 0.5$~$1.0$ and $q \approx 2.0$ [10,11]. Then the effective pinning centers of all samples would be grain boundaries.

Fig. 5 shows the field angular dependence of $J_c$ at 20 K for 3 T. Then a large peak at $\theta = 0^\circ$ appears in the all samples. This result indicates that the all samples have $c$-axis correlated pinning centers which are grain boundaries as mentioned above. The value of $J_c$ of MgB$_2$/Ti/MgO is higher than that of MgB$_2$/MgO in all angles in spite of the same
substrate. Small peaks at $\theta = 90^\circ$ are due to the intrinsic pinning of MgB$_2$.

Although the grain boundaries act as dominant pinning centers in all samples, there is a difference in the $J_c$. The highest $J_c$ in the low magnetic fields was observed in MgB$_2$/Ti/ZnO. This result would be caused by the low density of grain boundaries, which means that MgB$_2$/Ti/ZnO has large MgB$_2$ grains. The large MgB$_2$ grains originate from a little lattice mismatching between MgB$_2$ and Ti buffered ZnO(001) substrate. The highest $J_c$ was observed in MgB$_2$/Ti/MgO from 3 to 8 T. This result indicates the higher density of grain boundaries. The high density of grain boundaries is caused by small MgB$_2$ grains. It is considered that the higher density of grain boundaries is caused by the effect of both Ti buffer layer and MgO substrate in growth process. Furthermore, MgB$_2$/Ti/Si shows the highest $J_c$ above 8T. Then it is predicted that the density of grain boundaries in MgB$_2$/Ti/Si would be higher than that in MgB$_2$/Ti/MgO. In addition, it was reported that the small MgB$_2$ grains cause high $B_{c2}$ [12]. Therefore, the high $B_{c2}$ for MgB$_2$/Ti/Si may be due to both the diffusion of Si into the MgB$_2$ thin film and small MgB$_2$ grains.

The isothermal $E$ - $J$ curves of MgB$_2$/Ti/MgO at 1 T for $B//c$ are shown in Fig.6. The $E$ - $J$ curves at a low temperature region have negative curvatures in a log-log plot. As the temperature increases, the $E$ - $J$ curves vary from a negative curvature to a positive
curvature on reaching a certain temperature. This temperature is called the glass-liquid transition temperature $T_g$. It has been reported that the $E - J$ characteristics of high-$T_c$ superconductors are expressed by the percolation transition model. According to this model [13], the probability function of local current density $J_{cl}$ is given by the Weibull distribution function as

$$P(J_{cl}) = \left(\frac{J_{cl} - J_{cm}}{\Delta J_c}\right)^{m-1} \exp\left[-\left(\frac{J_{cl} - J_{cm}}{\Delta J_c}\right)^m\right].$$

where $J_{cm}$ is the minimum value of the distribution of the local current density, $\Delta J_c$ is the half width of the distribution, and $m$ is the pinning parameter which characterizes the shape of the distribution function. When the distribution of pinning strength is more uniform, the value of the pinning parameter $m$ becomes larger. Using the distribution function Eq. (1), the $E - J$ characteristics are given as follow:

$$E = \rho_{FF} \int_{J_{cm}}^J \left(\frac{J - J_{cl}}{\Delta J_c}\right)^{m-1} \exp\left[-\left(\frac{J - J_{cm}}{\Delta J_c}\right)^m\right] \, dJ_{cl}.$$  

$$= \begin{cases} 
\rho_{FF} \int_{J_{cm}}^J \left(\frac{J - J_{cl}}{\Delta J_c}\right)^{m-1} \exp\left[-\left(\frac{J - J_{cm}}{\Delta J_c}\right)^m\right] \, dJ_{cl} & \text{for } T < T_g, \\
\rho_{FF} \Delta J_c \left(\frac{J}{\Delta J_c}\right)^{m-1} & \text{for } T = T_g, \\
\rho_{FF} \Delta J_c \left(\frac{J}{\Delta J_c}\right)^{m-1} - \left(\frac{J_{cm}}{\Delta J_c}\right)^m & \text{for } T > T_g, 
\end{cases}$$

where $\rho_{FF}$ is the flux flow resistivity. We fitted the theoretical expression Eq. (2) to the $E - J$ curves of MgB$_2$ thin films. The pinning parameters $J_{cm}, \Delta J_c$ and $m$ were determined as the error between experimental data and fitting theoretical curves makes minimum.
As shown in Fig.6, the theoretical curves are in excellent agreement with the experimental data in all measured temperatures. In the same way, the theoretical curves are in good agreement with the experimental data of MgB$_2$/Ti/Si and MgB$_2$/MgO.

Fig.7 shows the magnetic field dependence of $m$ for MgB$_2$/Ti/MgO, MgB$_2$/Ti/Si and MgB$_2$/MgO. In all magnetic fields, MgB$_2$/Ti/Si shows the highest $m$-values. This result indicates that the distribution of pinning strength is very uniform. The high $m$ value probably originates from the high density of grain boundaries in MgB$_2$/Ti/Si. This result coincides with the magnetic field dependence of $J_c$. The value of $m$ of MgB$_2$/Ti/MgO is higher than that of MgB$_2$/MgO. This result indicates that the density of grain boundaries is higher than that of MgB$_2$/MgO.

**Conclusions**

In this study, we deposited MgB$_2$ thin films on Ti buffered substrates. In order to investigate the effects of various substrates and Ti buffer layer, the temperature dependence of resistivity in the magnetic field, the magnetic field dependence of $J_c$, field angular dependence of $J_c$, and $E - J$ characteristics were measured. In the $B - T$ phase diagram, MgB$_2$/Ti/Si shows the high $B_{c2}$ at low temperature. MgB$_2$/Ti/ZnO shows the highest $J_c$ in the low magnetic fields, MgB$_2$/Ti/MgO shows the highest $J_c$ from 3 to 8 T and MgB$_2$/Ti/Si shows the highest $J_c$ above 8 T, respectively. The scaling analysis of
the macroscopic pinning force density shows that the grain boundaries are the dominant pinning centers in all samples. It is confirmed that the all samples have the $c$-axis correlated pinning centers due to the grain boundaries from the field angular dependence of $J_c$. The highest $J_c$ of MgB$_2$/Ti/ZnO in the low magnetic field originates from the low density of grain boundaries by a little lattice mismatching between MgB$_2$ and the Ti buffered ZnO(001) substrate. On the other hand, The highest $J_c$ of MgB$_2$/Ti/MgO in 3 to 8 T is due to the high density of grain boundaries by the effect of both Ti buffer layer and MgO(100) substrate. The highest value of $J_c$ of MgB$_2$/Ti/Si over 8 T is caused by the high density of grain boundaries and its high $B_{c2}$. This result coincides with the highest $m$-value of MgB$_2$/Ti/Si. Therefore, MgB$_2$/Ti/Si has a uniform distribution of pinning strength.

Therefore, we can control the density of grain boundaries in MgB$_2$ thin films using the various substrates and Ti buffer. This method is useful for the application of electronic devices and wires since the value of $J_c$ changes with grain size.
References


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Table 1 Samples in this study
Figure captions

**Fig.1** Temperature dependence of resistivity.

**Fig.2** $B - T$ phase diagram.

**Fig.3** Magnetic field dependence of $J_c$ at 20 K for $B//c$.

**Fig.4** Scaling analysis of $F_p - B$ at 20 K for $B//c$.

**Fig.5** Field angular dependence of $J_c$ at 20 K for 3 T.

**Fig.6** $E - J$ characteristics for MgB$_2$/Ti/MgO at 1 T.

**Fig.7** Magnetic field dependence of $m$ at $B//c$. 
$f = A b^p (1 - b)^q$

- $p = 0.9$
- $q = 2.1$
- $A = 6.25$

$B // c$

$T = 20K$
$I_c [\text{A/m}^2]$ vs. $B/c$, $\theta [\text{deg.}]$, and $B/ab$ for different samples:
- $T = 20K$:
  - MgB$_2$/Ti/MgO
  - MgB$_2$/Ti/Si
  - MgB$_2$/MgO
  - MgB$_2$/Ti/ZnO

$B = 3T$.