Investigation of critical properties in MgB$_2$/SiC/Si thin films prepared under varied conditions

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Abstract. Critical magnetic properties of MgB$_2$ thin films grown on SiC/Si substrate are investigated with different preparation conditions. MgB$_2$ films were prepared by sequential evaporation of boron and magnesium on SiC buffered Si substrate. The substrate temperature and the amount of supplied boron was varied and influence of varied conditions is investigated. The maximum superconducting transition temperature obtained is 34.5 K. The upper critical field $H_{c2}$ is estimated from the onset of AC diamagnetic susceptibility. The temperature dependence is generally linear, being fitted with $|dH_{c2}/dT|$ around 3 ~ 7 kOe/K depending on the substrate temperature during in situ annealing as well as the magnetic field direction. The critical current density is evaluated from DC magnetization hysteresis to be more than $10^6$ A/cm$^2$ at lower temperatures. Irreversibility field scales with $[1 - (T/T_c)]^n$ with critical exponents n of 3 ~ 7. Correlations among critical properties, crystal qualities and sample preparation conditions are discussed.

1. Introduction
Among many investigations on the binary metallic MgB$_2$ superconductor[1], the form of thin films is of particular interest from basic as well as applied points of view[2, 3].

In our previous articles[4, 5, 6, 7], investigations of MgB$_2$ thin films on Si substrate are reported with different buffer layers: NbN and SiC. We obtained larger upper critical field $H_{c2}$ with SiC buffer than NbN buffer, while obtained smaller irreversibility field $H_{irr}$ with SiC than NbN. Since varieties of factors, such as film morphology, crystallinity, impurities could influence superconducting critical parameters, it cannot hastily be concluded that these variations in $H_{c2}$ and $H_{irr}$ are simply due to difference in the buffer layer.

In order to further elucidate influence of preparation conditions on critical properties, we investigate in this report MgB$_2$/SiC/Si thin films with different preparation conditions.

2. Experimental procedure
MgB$_2$ thin films were prepared by sequential evaporation of boron and magnesium bilayers on SiC buffered Si substrates followed by in situ annealing[8]. The precursor Mg-B bilayers were deposited at room temperature substrates by electron beam evaporation. The amount of B was varied so as to result in different thickness of stoichiometric MgB$_2$ film after reaction with the
excess Mg top layer. As-deposited films were in situ heated to 280°C for 30 min in an argon atmosphere at a pressure of 0.06 Pa. Subsequently, the pressure was increased up to 16 Pa and the temperature was increased to maximum temperature of either 750°C or 830°C and kept there for 10 min. The samples were then cooled down to room temperature at 10³ Pa Ar pressure. According to cross sectional study, the obtained thickness of MgB₂ was about 10 and 50 nm. These films were cut into several pieces and stacked together with insulating PTFE tape so as to fit into the sample holder for magnetic measurements. Because of limited space of this report we concentrate our attention on the results for 50 nm films here.

AC and DC magnetizations were measured with magnetic fields parallel and perpendicular to the film using PPMS magnetometer (Quantum Design). The onset transition temperature $T_{c2}(H)$ of AC diamagnetic susceptibility at each field $H$ was evaluated and temperature dependence of the upper critical field $H_{c2}(T)$ was deduced. The critical current density $J_c$ was estimated from DC magnetization hysteresis with the Bean critical state model, $J_c = 30 \Delta M/r$, where $\Delta M$ is the height of the magnetization loop and $r \sim 0.15$ cm is the sample half-width. Irreversibility field $H_{irr}$ was also estimated from diminishing magnetization hysteresis.

3. Results and discussion

In Figure 1 we indicate onsets of the AC diamagnetic susceptibility $\chi'$ under various magnetic fields perpendicular to the MgB₂ film annealed at maximum temperature of 750°C. As indicated by arrows, onset transition temperatures $T_{c2}(H)$ vary linearly in general with applied fields, from which we obtain temperature dependent upper critical fields $H_{c2}(T)$.

Figure 1. Onsets of the AC diamagnetic susceptibility $\chi'$ under various magnetic fields perpendicular to the film surface of MgB₂/SiC/Si annealed at 750°C.

Figure 2 represents thus obtained $H_{c2}$ as a function of temperature $T$ by open and full diamonds for the MgB₂/SiC/Si film annealed at 750°C, comparing them to $H_{c2}$ for the film annealed at 830°C with open and full circles[6]. Open circles and open diamonds correspond to magnetic fields parallel to the film surface, while full circles and full diamonds correspond to magnetic fields perpendicular to the surface.

We here observe certain anisotropy of $H_{c2}$ due to the field direction for both films annealed at 750°C and 830°C. This can be explained by the preferred orientation of nanocrystals in the films, which is observed by the Selected Area Diffraction (SAD) patterns with Transmission Electron Microscopy (TEM)[9]. In Table 1 we summarize values of temperature derivative of
the upper critical field, $|dH_{c2}^\parallel/dT|$ for the parallel field and $|dH_{c2}^\perp/dT|$ for the perpendicular field estimated from the linear fitting to the data in Figure 2, together with corresponding values for MgB$_2$/NbN/Si film annealed at 700°C[5]. Apparent anisotropy parameter $\gamma' = \frac{|dH_{c2}^\parallel/dT|}{|dH_{c2}^\perp/dT|}$ is also given in Table 1. The inspection of Table 1 indicates a tendency the higher the annealing temperature, the larger the anisotropy. However, difference of $\gamma'$ in SiC-buffered films annealed at 750°C and 830°C is rather small, while the anisotropy in MgB$_2$/NbN/Si is only little. Therefore, it is inferred that preferred orientation of nanocrystals is more readily realized with SiC than NbN buffer layer.

### Table 1. Summary of critical parameters of MgB$_2$ thin films prepared under various conditions.

| buffer layer | annealing temperature | $T_c$ | $|dH_{c2}^\parallel/dT|$ | $|dH_{c2}^\perp/dT|$ | $\gamma'$ |
|-------------|----------------------|------|----------------|----------------|------|
| SiC         | 830°C                | 32.0 K | 7.4 kOe/K  | 4.1 kOe/K  | 1.8  |
| SiC         | 750°C                | 34.5 K | 4.9 kOe/K  | 3.1 kOe/K  | 1.6  |
| NbN         | 700°C                | 31.0 K | 4.9 kOe/K  | 4.4 kOe/K  | 1.1  |

Next, we examine the coherence length $\xi^\parallel$: characteristic length of the order parameter variation in the direction parallel to the film, which is related to $H_{c2}^\parallel$ by the equation[10]:

$$H_{c2} = \frac{\Phi_0}{2\pi\xi^\parallel}$$

with the flux quantum $\Phi_0 = h/2e$.

When we compare the values of the temperature derivative $|dH_{c2}^\perp/dT|$ in the SiC buffered films (Table 1), it is known that $\xi^\parallel$ in the film annealed at 830°C is shorter than the one annealed at 750°C. This is probably explained by the shorter mean free path due to some impurities produced by higher annealing temperature of 830°C. We actually observed interdiffusion[9] between SiC buffer layer and MgB$_2$ film. It is natural that the higher the annealing temperature rises, the more readily the interdiffusion takes place. Increased amount of SiC or Si as intergranular materials should reduce the intergrain coupling and then shorten the electron mean free path and $\xi^\parallel$. Impurity phase like Mg$_2$Si can also affect $\xi^\parallel$ as discussed previously[6, 7].

As for the NbN buffered film annealed at 700°C, since the geometrical average of 4.9 and 4.4 kOe/K is larger than that of the SiC buffered film annealed at 750°C, $\xi^\parallel$ in the NbN buffered film may be shorter than that of the SiC buffered film annealed at 750°C. This is not unusual too, because the melting temperature of NbN (2050°C) is lower than that of SiC (2830°C). In practical applications, either shorter or longer $\xi^\parallel$ will be desired depending on the type of applications (power system, electronics, etc.). It is suggested that variety of values of $\xi^\parallel$ can be obtained by combining different buffer layers and various annealing temperatures.

Other important parameter for the practical applications is the critical current density $J_c$, which is shown in Figure 3 for respective temperatures as a function of magnetic field perpendicular to the surface of MgB$_2$/SiC/Si annealed at 750°C. We note that $J_c$ at lower temperatures and lower fields well exceeds 10$^6$ A/cm$^2$, providing plenty of margin for high current applications. This $J_c$ is larger than that of MgB$_2$/SiC/Si annealed at 830°C. This is again explained by the better crystallinity with less impurity (suppressed interdiffusion between MgB$_2$ and SiC or Si) due to lower annealing temperature of 750°C. The remaining issue is decrease of $J_c$ at higher magnetic field, which is related to the irreversibility field.

Then we estimate the irreversibility field $H_{irr}$ by taking $J_c = 60$ kA/cm$^2$ (1/100 of the maximum $J_c$) as the criterion of reversibility with magnetic field perpendicular to the film. The estimated $H_{irr}$ for MgB$_2$/SiC/Si annealed at 750°C is plotted with full diamonds in Figure 4.
Figure 3. Critical current density $J_c$ as a function of magnetic field perpendicular to the surface of MgB$_2$/SiC/Si annealed at 750°C.

Figure 4. Irreversibility field $H_{irr}$ in MgB$_2$/SiC/Si film annealed at 750°C (fulldiamond) and in the film annealed at 830°C (●) as a function of $1 - t^2$ with $t = T/T_c$.

as a function of $1 - t^2$. Here $t = T/T_c$ is the reduced temperature with $T_c = 34.5$K. $H_{irr}$ for MgB$_2$/SiC/Si annealed at 830°C is also plotted (●), which is reanalyzed by the same criterion of 1/100 of the maximum $J_c$.

We find interesting difference in the temperature variation of $H_{irr}$, between the films annealed at 830°C and annealed at 750°C. As for the film annealed at 830°C, $H_{irr}$ decreases rapidly at higher temperatures probably due to weak links between the crystal grains with diffused impurities[6]. This is consistent with the above examinations on $|dH_{cd}/dT|$ and $\xi_\parallel$ concerning Figure 2. On the other hand, $H_{irr}$ in the film annealed at 750°C keeps steady values even at higher temperatures and follows the scaling low: $H_{irr} \propto (1 - t^2)^n$, with exponents $n$ of 3 at all temperatures. This also indicates the good crystal quality of the 750°C annealed film.

In conclusion, critical properties are compared among MgB$_2$ films with different buffer layers and different annealing temperatures. The lower temperature annealing resulted in better crystal quality with larger $\xi_\parallel$, $J_c$ and $H_{irr}$.

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