Investigation of critical behaviour of MgB$_2$ thin films on SiC/Si substrate

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Abstract. Critical magnetic behaviour of MgB$_2$ thin films grown on SiC-buffered Si substrate was investigated in comparison with MgB$_2$ films on NbN-buffered Si substrate. MgB$_2$ films were prepared by sequential evaporation of boron and excess magnesium followed by in situ annealing in an Ar atmosphere. The upper critical field $H_{c2}$ estimated from the onset of AC diamagnetic susceptibility was larger in films with SiC buffer than those with NbN buffer. The temperature dependence was almost linear with $|dH_{c2}/dT|$ up to 8 kOe/K, especially under parallel magnetic field to the film surface. Although the critical current density evaluated from DC magnetization hysteresis approached 1 MA/cm$^2$ at the lowest temperatures, the maximum value was smaller than that in MgB$_2$/NbN/Si. The irreversibility field also inferred effects of weak links, possibly due to some impurity phases.

1. Introduction

Many investigations have been carried out on the binary metallic MgB$_2$ superconductor[1] with various forms of samples[2, 3, 4, 5, 6, 7, 8, 9, 10, 11]. Among these, thin film samples are of particular interest from basic as well as applied point of view.

We previously reported[12, 13] on critical properties of MgB$_2$ thin films prepared on NbN-buffered Si substrate, and observed the critical current density $J_c$ exceeding 1 MA/cm$^2$ which is fairly large compared with sintered bulk[2, 4, 5] and single crystalline[6, 7, 8] samples. On the other hand, the lower critical field $H_{c1}$ and the irreversibility field $H_{irr}$ were rather small compared with bulk and crystals.

Since varieties of factors, e.g. film morphology, crystallinity, substrate, impurities, can contribute to these critical values, it is important to investigate superconducting properties in varieties of samples even within a category of thin films. In this work, in order to examine difference of buffer layers, we investigate critical properties ($H_{c2}$, $J_c$ and $H_{irr}$) of MgB$_2$ thin films prepared on SiC-buffered Si substrate, and compare them with the results in NbN-buffered films.

2. Experimental procedure

The MgB$_2$ thin films were prepared by sequential evaporation of boron and magnesium bilayers on SiC-buffered Si substrates followed by in situ annealing[14]. The precursor Mg-B bilayers
were deposited at room temperature substrates by electron beam evaporation. The thickness of the B layer was adjusted so as to result in 200 nm 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 830°C and kept there for 10 min. Application of SiC as a buffer layer enabled to raise this annealing temperature higher compared with the previous annealing temperature of 700°C with NbN buffer layer[15]. The samples were then cooled down to room temperature at 10$^3$ Ar pressure. Examination with transmission electron microscopy revealed a nanogranular character of the prepared films[14].

The obtained film was cut into several pieces and stacked together so as to fit into the sample holder for magnetic measurements. AC and DC magnetizations were measured with magnetic fields parallel and perpendicular to the film surface 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 $r = 0.11$ cm is the sample half-width. Irreversibility field $H_{irr}$ was also estimated from diminishing magnetization hysteresis.

3. Results and discussion
In Fig. 1 We indicate onsets of the AC diamagnetic susceptibility $\chi'$ under various magnetic fields perpendicular to the MgB$_2$ film surface. As indicated by arrows, onset transition temperatures $T_{c2}(H)$ vary almost linearly with applied fields, from which we obtain temperature dependent upper critical fields $H_{c2}(T)$.

![Figure 1](image1.png)

**Figure 1.** Onsets of the AC diamagnetic susceptibility $\chi'$ in MgB$_2$/SiC/Si under various magnetic fields perpendicular to the film surface as indicated by arrows.

In Fig. 2 we show thus obtained $H_{c2}$ as a function of temperature $T$ in MgB$_2$ on SiC/Si substrate by solid circles for the field perpendicular to the film surface and by open circles for the field parallel to the surface. We note that $H_{c2}$ for the parallel field is fairly larger (almost by a factor of 2) than that for the perpendicular field. This behaviour is different from almost identical $H_{c2}$ for both directions of the field in MgB$_2$ on NbN/Si substrate[13], their average values being shown by the dashed line in Fig. 2.

![Figure 2](image2.png)

**Figure 2.** Upper critical field $H_{c2}(T)$ under parallel (○) and perpendicular (●) fields for MgB$_2$/SiC/Si, which is larger than average $H_{c2}$ for MgB$_2$/NbN/Si (---).
X-ray diffraction measurements on our MgB$_2$/SiC/Si samples indicated no clear peaks assignable to the MgB$_2$ phase. This implies a nanogranular character of our film, which is also confirmed by Transmission Electron Microscopy (TEM)[14]. Selected Area Diffraction (SAD) patterns with TEM indicated diffraction rings originated from the MgB$_2$ phase, where each ring was not completely uniform but involved bright spots within the ring. This infers that respective granules correspond to oriented nanocrystals, and such orientation of nanocrystals is not completely random throughout the entire film. The observed difference in $H_{c2}$ between perpendicular and parallel fields may be explained by such preferred orientation. If the crystal $ab$ planes are preferably oriented parallel to the film surface, $H_{c2}^\parallel$ becomes closer to $H_{c2}^{ab}$ and larger than $H_{c2}^\perp$ which is closer to $H_{c2}^{ab}$. In the case of MgB$_2$/NbN/Si, the \textit{in situ} annealing temperature was not so high and the film was more like amorphous (with random orientation), which resulted in similar $H_{c2}$ for both directions of the field.

Secondly, we note that the present $H_{c2}$ in MgB$_2$/SiC/Si is larger than that in MgB$_2$/NbN/Si. Especially, the temperature derivative, $|dH_{c2}^\perp/dT|$ of around 8 kOe/K seems to be among the highest ever reported[3, 4, 7, 8]. Although concrete mechanism for such enhancement is not known, possible impurity phases such as Mg$_2$Si or MgO[14] might play roles to shorten the coherence length $\xi$ and then enhance $H_{c2}$. This can also explain the similar values of $|dH_{c2}^\perp/dT|$ in SiC- and NbN-buffered MgB$_2$, \textit{i.e.} coincidental balance between enhancement due to short $\xi$ and suppression due to anisotropy.

The critical current density $J_c$ is another important parameter for practical applications, and the result in MgB$_2$/SiC/Si with magnetic fields perpendicular to the film surface is indicated in Fig. 3. It is noted that $J_c$ at lower temperatures exceeds $10^5$ A/cm$^2$: a common benchmark for practical applications, and approaches 1 MA/cm$^2$ at the lowest temperatures (self field). However, absolute values seem to be smaller by a factor of about 3 than $J_c$ in MgB$_2$/NbN/Si. The reason for this may be related to different sample characteristics of the prepared films. As reported previously[15], MgB$_2$ films on NbN-buffered Si are fairly homogeneous and uniform like amorphous without any impurities, while MgB$_2$ films on SiC-buffered Si are of oriented nanocrystals with impurity phases, probably due to higher reaction temperature. For example, Mg$_2$Si grains with a size of about $10 \times 40$ nm$^2$ were found especially at the surface of
MgB$_2$/SiC/Si[14]. Thus, such developed grain boundaries and impurities might reduce the intergrain coupling and lower $J_c$ in the present film.

Finally, we estimate the irreversibility fields $H_{irr}$ by taking $J_c = 8000$ A/cm$^2$ as the criterion of reversibility[13] and plot them against $1 - t^2$ for fields parallel (○) and perpendicular (●) to the surface in Fig. 4 with reduced temperature $t = T/T_c$ ($T_c = 32$ K). The solid lines indicate the scaling law: $H_{irr} \propto (1 - t^2)^n$, with exponents $n$ of 2 (for the parallel field) and 3 (for the perpendicular field). This scaling behaviour is quite consistent with those observed in MgB$_2$/NbN/Si, concerning not only their exponents but also their absolute values of $H_{irr}$. The value of $n \approx 2$ for the parallel field is fairly close to the previously reported value for the powder sample[16] and is consistent with the bulk flux pinning character. On the other hand, the value of $n \approx 3$ for the perpendicular field suggests additional pinning mechanism such as surface or grain boundary.

However, at higher temperatures for the perpendicular field, $H_{irr}$ deviates from the scaling law with $n \approx 3$ and seems to change to the law with the exponent $n \approx 6$. Such large $n$ value infers effects of some weak links possibly due to impurity phases such as Mg$_2$Si.

4. Conclusions

In conclusion, AC and DC magnetic measurements were performed in MgB$_2$/SiC/Si and compared with MgB$_2$/NbN/Si. The upper critical field in MgB$_2$/SiC/Si was larger than that in MgB$_2$/NbN/Si, reflecting the shorter coherence length probably due to impurities yielded by the higher reaction temperature. The temperature derivative, $|dH_{c2}/dT|$ of around 8 kOe/K was among the highest ever reported. On the other hand, although $J_c$ approached 1 MA/cm$^2$ at the lowest temperatures, the maximum value was smaller than that in MgB$_2$/NbN/Si. The irreversibility field $H_{irr}$ also inferred some effects of weak links. According to these investigations, it will be necessary to further search for appropriate impurity phases that can enhance $H_{c2}$ and act as the flux pinning centers but do not reduce the intergrain coupling.

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