Properties of MgB_2 Films With Very High Transport Critical Current Densities

Hitoshi Kitaguchi, Toshiya Doi, Yuki Kobayashi, Akiyoshi Matsumoto, Harini Sosiati, Satoshi Hata, Masao Fukutomi, and Hiroaki Kumakura

***Abstract*—Magnesium diboride, MgB_2, thin films are fabricated through two different kinds of process. One is an in-situ process by using electron beam deposition and the other is an ex-situ process by using the combination of pulsed laser deposition and heat treatment. The critical current density, J_c, is investigated as a function of external magnetic field in the range of 0–7 T and/or temperatures ranging from 4.2 K to the critical temperature by using dc 4-probe transport method. The in-situ processed film shows very high J_c, e.g., 7.1×10^6, 1.2×10^6, and 1.4×10^5 A/cm^2 in 0, 4, and 7 T (perpendicular fields), respectively. Angular dependence of J_c is much different between the two kinds of films. J_c-angle (magnetic field) curves of the in-situ processed film show two peaks; one is around the field perpendicular to the film surface and the other is around the parallel field. On the contrary, the curves of the ex-situ processed film have only one peak around the parallel field. Microstructure analyses show that the in-situ film has columnar grains aligned perpendicular to the film surface and that the ex-situ one has granular grains with random orientations. These results indicate that the grain boundaries between columnar grains act as effective pinning centers and enhance J_c in the perpendicular fields.**

***Index Terms*—Critical current densities (J_c), irreversibility fields, MgB_2, pinning, thin films.**

I. Introduction

MAGNESIUM DIBORIDE, MgB_2, superconductor [1] has been studied extensively by many research groups in the world since its discovery aiming for high current applications and superconducting electronic devices. The advantages of MgB_2 may exist in its simple crystal structure, long coherence length, and so on. These characteristics lead to high critical current density, J_c, due to high transparency at grain boundaries for the current flow. Intensive research works have been performed to fabricate MgB_2 thin films with high J_c [2]–[14]. For example, J_c reaching 10^7 A/cm^2 at 4.2 K in zero external fields is reported [2]–[10]. Magnetic field dependence of J_c is large even at low temperatures in most cases [2]–[5], [11], [12]. For example, J_c decreases to about 10% of self-field value in 1–3 T and to about 1% in 4–5 T. Improvements in flux pinning properties leading to J_c enhancement in the fields are expected. We have confirmed [15] earlier deductions [16]–[18] that the grain boundaries are effective as pinning sites. We have also reported that the columnar grain growth perpendicular to the MgB_2/substrate interface enhances J_c in the field perpendicular to the film surface [15].

In this paper, we report on the performance of two kinds of MgB_2 thin films with different microstructures. The comparison of the characteristics between these two films will give further understanding of the pinning in MgB_2 films.

II. Experiments

We prepared two kinds of MgB_2 thin films using two different processes. We fabricated MgB_2 films on polished sapphire C (0001) single-crystal substrates through in-situ process using electron beam deposition, EB, or through ex-situ process using the combination of pulse laser deposition, PLD, and heat treatment (hereafter referred to as “in-situ EB film” and “ex-situ PLD film”, respectively.) The details of sample preparation procedure have been reported elsewhere [15], [18]. The in-situ EB film experienced no heat treatment after the deposition, so the characterization was performed in “as-grown” state. The ex-situ PLD film was heat treated in pure argon gas at 883 K for 30 min. in order to form MgB_2 phase after the deposition of the precursor layer. The thickness of the MgB_2 layer was measured by using surface texture measuring instruments. These values are 0.30 and 0.40 μm for the in-situ EB and the ex-situ PLD films, respectively. A strip line of 1.0–1.2 mm width and 1.0–2.0 mm length was formed in each film for the dc four-probe transport measurements. Critical currents, I_c, were determined with the electric field criterion of 1 $\mu V/cm$ and then J_c values were calculated based on the width of the strip line and the thickness of the MgB_2 layer. I_c measurements were performed in fields of 0–7 T and at temperatures between 4.2 K and the critical temperature, T_c. The samples were cooled in the liquid He or temperature controlled He gas in 4.2 K- or high-temperature-measurements, respectively. We also measured I_c as a function of field direction at 4.2 K. The current direction was set perpendicular to the field direction and the sample was rotated around this current axis. In this paper, we define the field direction, $\theta, = 0°$ and $90°$ as the fields parallel and perpendicular to the film surface (hereafter referred to as “parallel field” and “perpendicular field”), respectively. Upper critical fields, B_{c2}, and irreversibility fields, B_{irr}, were investigated with the temperature dependence of resistance, R. B_{c2} and B_{irr} were determined with the criteria of 90% and 0.1% of the normal-state resistance in the R-T curves of the T_c transition, respectively.

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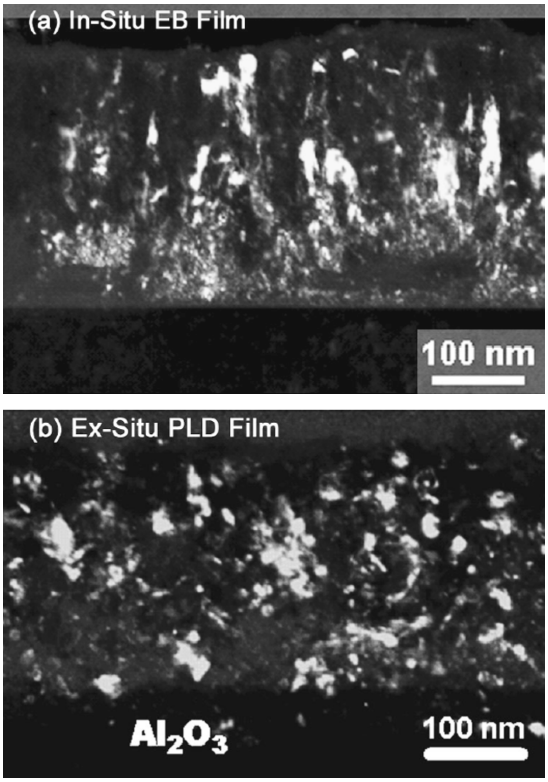

Fig. 1. TEM micrographs of the cross section of (a) the in-situ EB film and (b) the ex-situ PLD film. Dark-field images are shown in order to indicate the difference in their microstructures. The in-situ EB film has a columnar grain structure aligned perpendicular to the MgB_2/substrate interface. TEM analyses revealed that c axes of MgB_2 crystals align with the column direction. Contrary, the ex-situ PLD film has a granular structure without any texture.

III. Results and Discussion

Fig. 1 shows the microstructure of the cross section of the films. Dark-field images obtained in TEM observation are shown in order to indicate the difference in these microstructures. The two films have very different microstructure. The in-situ EB film has a columnar grain structure where the columnar grains are aligned perpendicular to the MgB_2/substrate interface. Typical diameter of the columnar grains is 20–30 nm. TEM analyses revealed that c axes of MgB_2 crystals align with the column direction. In contrast, the ex-situ PLD film has a granular structure without any texture.

Fig. 2 shows the field dependence of J_c for these two films. J_c of the film in zero-external-field reaches $7.1 \times 10^6\ \mathrm{A/cm^2}$. J_c of the in-situ EB film exceeds $10^6\ \mathrm{A/cm^2}$ even in a perpendicular field of 4 T. J_c values are 1.2×10^6 and $1.4 \times 10^5\ \mathrm{A/cm^2}$ in 4 and 7 T perpendicular field, respectively. It is notable that the in-situ EB film has higher J_c in perpendicular field than in parallel field. Contrary, the ex-situ PLD film shows higher J_c in parallel field than in perpendicular field. The global pinning force, $F_p, = J_c \times B$ was calculated using the data shown in Fig. 2 and $F_p - B$ relations are plotted in Fig. 3. F_p has a peak around 2–3 T for the in-situ EB film in perpendicular field. The columnar grain structure of the in-situ EB film gives a good explanation for these results. Strong pinning at the boundaries between columnar grains aligned perpendicular to the surface leads to high J_c values and large pinning force in perpendicular fields. The columnar grain size of 20–30 nm and the spacing between magnetic flux lines (35 nm in 2 T and 20 nm in 6 T) show a good agreement.

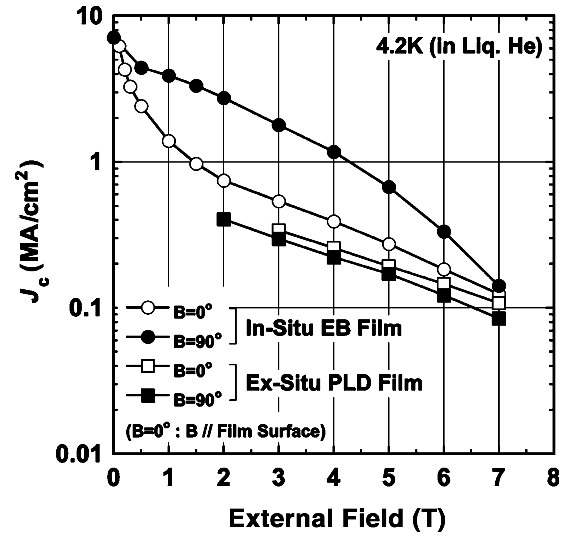

Fig. 2. $J_c - B$ relations at 4.2 K for the in-situ EB and the ex-situ PLD films. $J_c - B$ curve of the in-situ EB film in the perpendicular field stays higher than in the parallel field. On the contrary, J_c of the ex-situ PLD film is higher in the parallel field than in the perpendicular field.

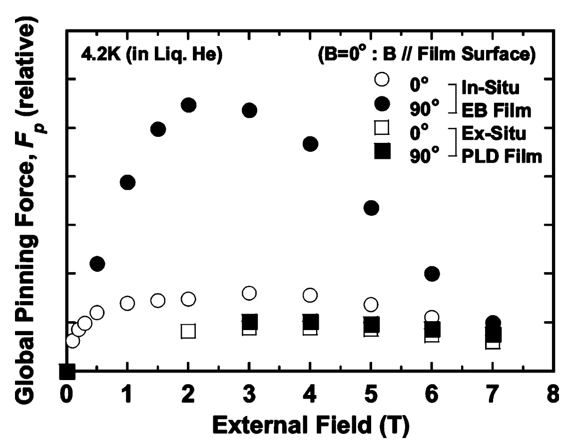

Fig. 3. Magnetic field dependence of global pinning force, F_{p}, for the in-situ EB and the ex-situ PLD films.

In Fig. 4, J_c values are plotted as a function of θ for the two films. The $J_c - \theta$ curve for the in-situ EB film has two peaks around $\theta = 0°$ and $\theta = 90°$. J_c values are much higher around $\theta = 90°$ than around $\theta = 0°$. The higher peak around $\theta = 90°$ can be attributed to the grain boundary pinning originating in the columnar grain structure. The possible origin of the peak around $\theta = 0°$ maybe the surface pinning and/or geometric effects. On the other hand, the $J_c - \theta$ curve for the ex-situ PLD film has only one peak is around $\theta = 0°$. Although J_c is higher around $\theta = 0°$ than around $\theta = 90°$ for the ex-situ PLD film corresponding to the results in $J_c - B$ relations (see Fig. 2,) the difference in J_c values is small. The decrease of J_c around $\theta = 90°$ is only 13% of the maximum J_c value. The granular structure with randomly oriented grains in the ex-situ PLD film does not cause such an anisotropic behavior in pinning as due to the columnar grain structure and leads to the low and broad peak in the $J_c - \theta$ relation.

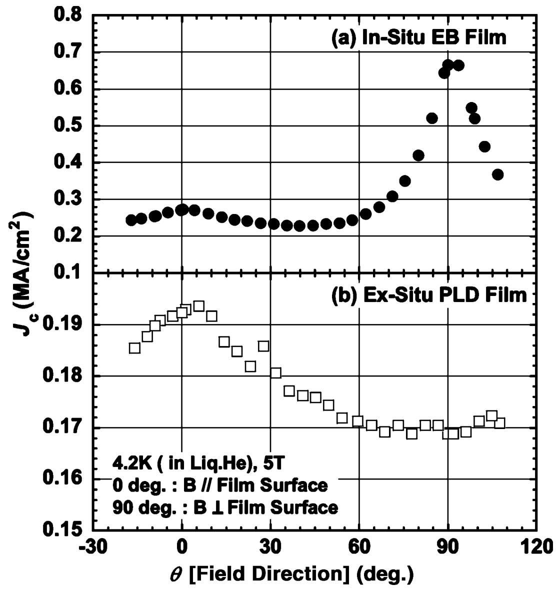

Fig. 4. J_c plots as a function of θ (angle of the fields against the film surface) for (a) the in-situ EB film and (b) the ex-situ PLD film at 4.2 K, 5 T. $\theta = 0°$ and 90° correspond to the field parallel and perpendicular to the film surface, respectively.

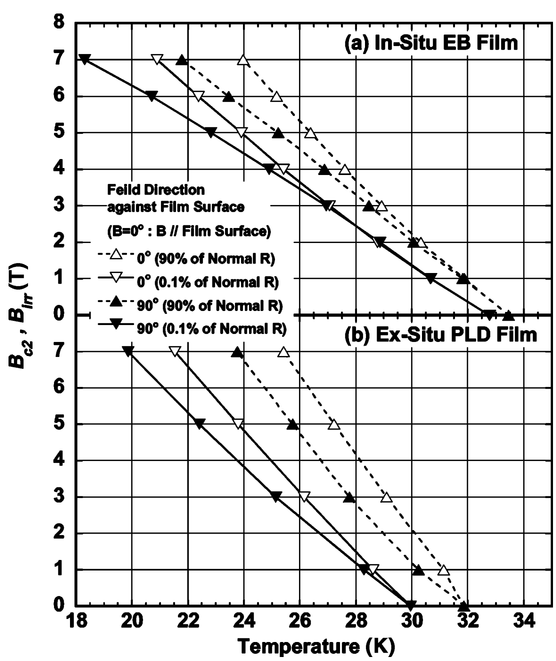

Fig. 5. B_{c2} and B_{irr} as a function of temperature for (a) the in-situ EB film and (b) the ex-situ PLD film. B_{c2} and B_{irr} stay higher in the parallel field than in the perpendicular field.

Fig. 5 shows the temperature dependence of B_{c2} and B_{irr} for the both films. B_{c2} and B_{irr} stay higher in parallel field than in perpendicular field. The angular dependence of B_{c2} and B_{irr} gives an explanation for $J_c - \theta$ relation (see Fig. 4) of the ex-situ PLD film and for the peak around $\theta = 0°$ of the in-situ EB film. However, the origin of the peak around $\theta = 90°$ in the in-situ EB film cannot be explained in terms of the angular dependence of B_{c2} and B_{irr}. These results strongly support the discussion that the origin of high J_c values of the in-situ EB film in perpendicular field is the pinning at the grain boundaries between columnar grains.

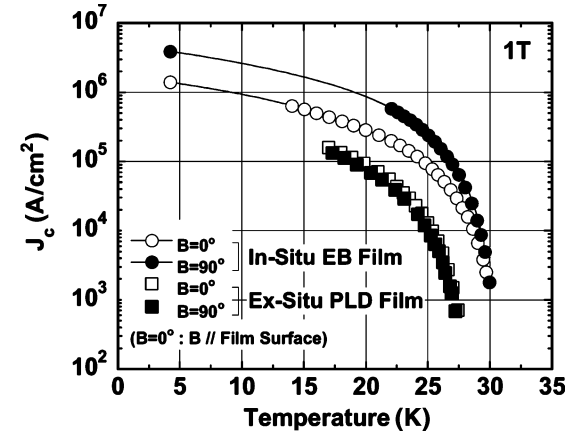

Fig. 6. Temperature dependence of J_c in 1 T for the in-situ EB and the ex-situ PLD films. The in-situ EB film has J_c nearly $10^6\ A/cm^2$ even at 20 K.

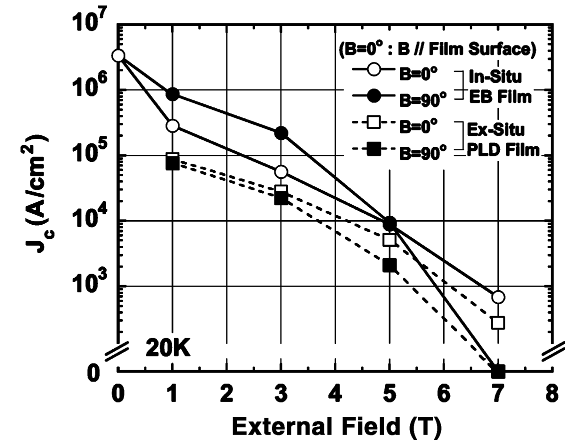

Fig. 7. $J_c - B$ relations at 20 K for the in-situ EB and the ex-situ PLD films. $J_c - B$ curve of the in-situ EB film in the perpendicular field stays higher than in the parallel field below 5 T.

Figs. 6 and 7 show the temperature dependence of J_c in 1 T. It is remarkable that the in-situ EB film shows J_c of nearly $10^6\ A/cm^2$ in perpendicular field even at 20 K. J_c enhancement due to columnar grain boundaries is confirmed at this temperature. For both films in perpendicular field, J_c decreases steeply above 5 T and becomes zero at 7 T. This is attributed to the fact that their B_{irr} are lower than 7 T at 20 K.

IV. Conclusion

We have compared the characteristics of in-situ-EB-and ex-situ-PLD-MgB_2/sapphire films. The results are summarized conclusively as follows:

(1) The in-situ EB film has an excellent performance. J_c values at 4.2 K are 7.1×10^6, 1.2×10^6, and $1.4 \times 10^5\ A/cm^2$ in 0, 4, and 7 T (perpendicular fields), respectively. It shows J_c of nearly $10^6\ A/cm^2$ in the perpendicular field of 1 T even at 20 K.

(2) The in-situ EB film and the ex-situ PLD film have columnar and granular grain structure, respectively.

(3) The in-situ EB film has the larger pinning force and thus the higher J_c in perpendicular field than in parallel field due to the strong pinning at the grain boundaries

between the columnar grains aligned perpendicular to the film surface.

(4) The ex-situ PLD film has the higher J_c in parallel field than in perpendicular field reflecting its granular grain structure and angular dependence of B_{c2} and B_{irr}.

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