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Study of magnetization and pinning mechanisms in MgB_2 thin film superconductors: effect of heavy ion irradiation

Anurag Gupta[1], **H Narayan**[1], **D Astill**[2], **D Kanjilal**[3], **C Ferdeghini**[4], **M Paranthaman**[5] **and A V Narlikar**[6]

[1] National Physical Laboratory, Dr K S Krishnan Marg, New Delhi 110 012, India
[2] IRCS, Cavendish Laboratory, West Cambridge Site, Madingley Road, Cambridge, UK
[3] Nuclear Science Centre, PO Box 10502, Aruna Asaf Ali Marg, New Delhi 110 067, India
[4] INFM, Dipartimento di Fisica, Via Dodecaneso 33, 16146 Genova, Italy
[5] Oak Ridge National Laboratory, Oak Ridge, TN 37831-6100, USA
[6] IUC for DAE Facilities, University Campus, Khandwa Road, Indore-452 001, India

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Abstract
We report magnetization studies on MgB_2 superconducting thin films in a temperature range 4.2–40 K and magnetic field range 0–6 T. Thin films prepared by both pulsed laser deposition (PLD) and electron beam evaporation (EBE) methods were investigated. In addition, both films were studied before and after heavy ion irradiation by 200 MeV Ag ions with a dose of 10^{11} ions cm^{-2}. Variation of sweep rates during the measurement of the magnetization loop reveals the presence of flux creep in both films. The PLD film, after irradiation, shows a severe degradation of T_c, critical current densities (J_c) in low fields and irreversibility line ($B^*(T)$). In contrast, the EBE film shows a slight enhancement in T_c, and nearly no change in $J_c(B)$ and the position of irreversibility line after irradiation. For both pristine films, the obtained volume pinning forces F_p versus reduced field $b = B/B^*$ shows a good scaling for $T \leqslant 10$ K, which matches well with the theoretical curve based on the flux line shear (FLS) pinning model. These and other results can be interpreted in terms of grain boundaries in MgB_2 films acting as FLS channels.

1. Introduction

The recently discovered [1] MgB_2 superconductor, besides being a simple bimetallic compound, looks promising for applications due to encouraging values of its superconducting critical temperature (T_c) and coherence length (ξ). Its $T_c = 39$ K is higher than that of 'conventional' low T_c superconductors (LTSC) and much lower as compared to that of high T_c superconductors (HTSC). The value of ξ ranging (due to anisotropy in MgB_2) from 23 Å to 68 Å [2] is comparable to that of the known practically useful LTSCs such as NbTi and Nb_3Sn, and much larger than that of anisotropic HTSC with ξ typically ranging from 1–4 Å to 20–40 Å. Thus it may be that in MgB_2, like LTSC, ξ is just sufficiently large to overcome grain boundaries as a limitation (the well known weak-link problem in HTSC) for high critical current densities (J_c). The fact that this is the case has been noted in several recent reports [3, 4]. Another advantage of typical values of T_c and ξ in MgB_2 may be that the flux creep, which also limits the J_c, may be less prominent when compared to HTSC. In the present work we investigate these issues by magnetization studies on MgB_2 thin films. To throw light on the dependence of J_c on microstructure, two differently synthesized MgB_2 films were studied. Further, both films were also irradiated by heavy energetic ions (200 MeV Ag^{+17} ions) to study its effect on their irreversible magnetization.

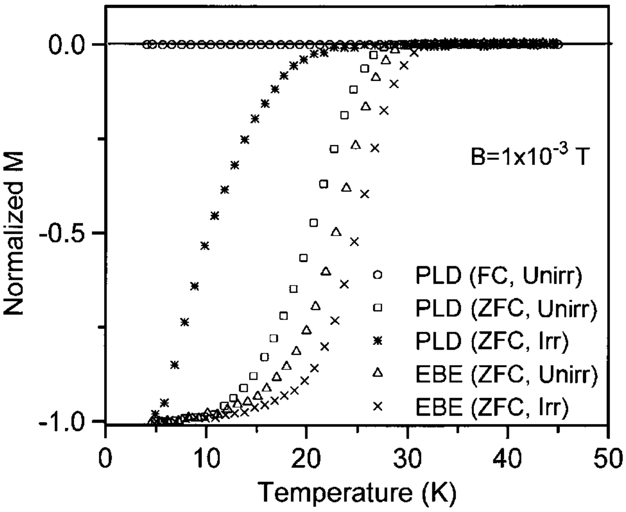

Figure 1. Normalized M as a function of T for both EBE and PLD films. 'Unirr' stands for unirradiated and 'irr' stands for irradiated.

2. Experimental details

Two MgB_2 thin films prepared by pulsed laser deposition (called the 'PLD' film) [5] and electron beam evaporation (called the 'EBE' film) [6] have been used for the present investigations. The films are c-axis oriented with randomly oriented grains in the ab-plane. The PLD film has a much smaller grain size (~100 nm) than that found in the EBE film (grain size ~500 nm). Also, the PLD films typically have a much larger normal state resistivity (ρ_n) as compared to EBE films. The presently used PLD and EBE films had a $T_c = 29$ K and 31 K, respectively. Two pieces of each of the films, with a typical size of 4–5 mm × 4–6 mm × 250–550 nm, were cut from a single larger piece or selected from the same batch of films. One piece of each of the films was used as a pristine sample whereas the other piece was irradiated (ion beam parallel to the c-axis) at a temperature of 80 K with 200 MeV Ag^{+17} ions at a dose of 1×10^{11} ions cm^{-2}. The irradiation was carried out at the Nuclear Science Centre, New Delhi, using the 15 UD Pelletron Accelerator facility. The possible creation of defects in MgB_2 samples by heavy ion irradiation has been reported by some of us earlier [7]. The magnetization (M) was measured by VSM both as a function of temperature (=4.2 K to $T > T_c$) with constant applied dc magnetic field (B), and as a function of B (=0–6 T) with fixed temperature.

3. Results and discussion

Figure 1 shows M as a function of T, with applied $B = 1 \times 10^{-3}$ T, for PLD and EBE films both before and after heavy ion irradiation. For all the samples, the zero field cooled (ZFC) $M(T)$ measurements show the expected diamagnetic transition whereas, within the resolution of VSM, none of the samples show a diamagnetic signal in the field cooled (FC) measurement. Note that neither of the pristine films has an optimum T_c. Several reasons such as disorder at Mg sites, formation of Mg vacancies, presence of impurities at B site and oxidation of Mg have been invoked to understand the variation of T_c in MgB_2 material [8–10]. After irradiation, in the case of the EBE film the onset temperature (defined as T_c) of the ZFC diamagnetic signal is found to increase, whereas in the case of the PLD film, the T_c shows a severe degradation (see figure1). These results indicate that heavy ion irradiation (see also [11, 12]) can both improve and degrade the superconducting properties depending on the preparation method of MgB_2 films.

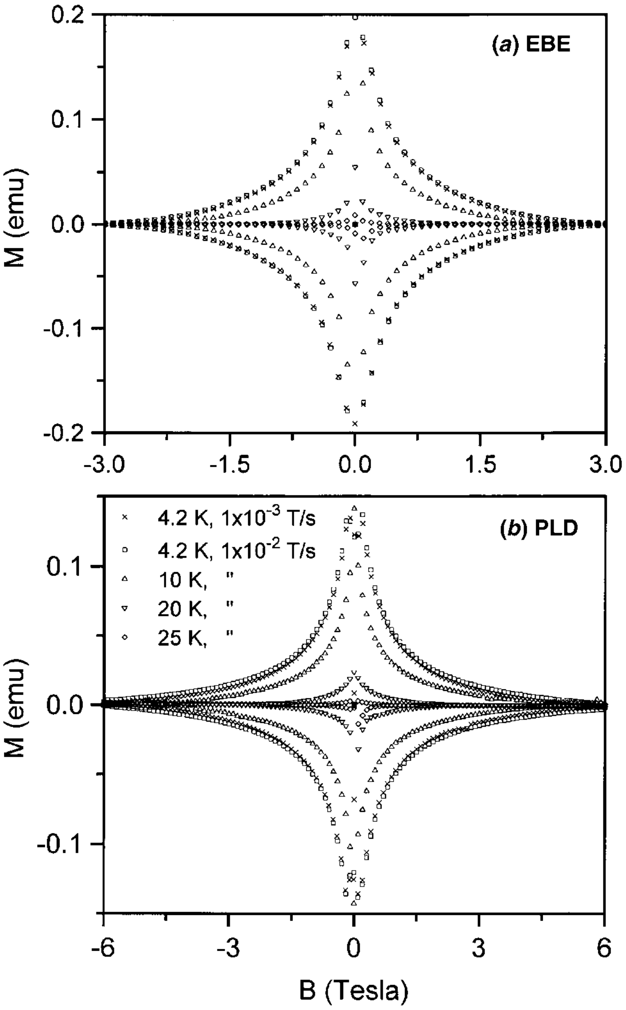

Figure 2. M–B loops measured at different temperatures for both EBE and PLD pristine films.

Figures 2(a) and (b) show M–B loops at different fixed temperatures for both EBE and PLD pristine films, respectively. For both films the loops become larger with a decrease in temperature. For the same temperature, in comparison to the EBE film, the loops in the PLD film close at higher fields (compare the x-axis scale of figures 2(a) and (b)). This shows that magnetic irreversibility in the PLD film persists in much higher magnetic fields than in the EBE film. The M–B loops were measured with two different sweep rates 1×10^{-3} T s^{-1} and 1×10^{-2} T s^{-1}. Figures 2(a) and (b) also show the same for both films at $T = 4.2$ K. A careful look at the data reveals that the loops become smaller for a lower sweep rate, which is a signature of flux creep in both films.

To make the data of M–B loops more comprehensible, $J_c(B,T)$ values were estimated for all the samples by using Bean's critical state model. J_c is related to the width of

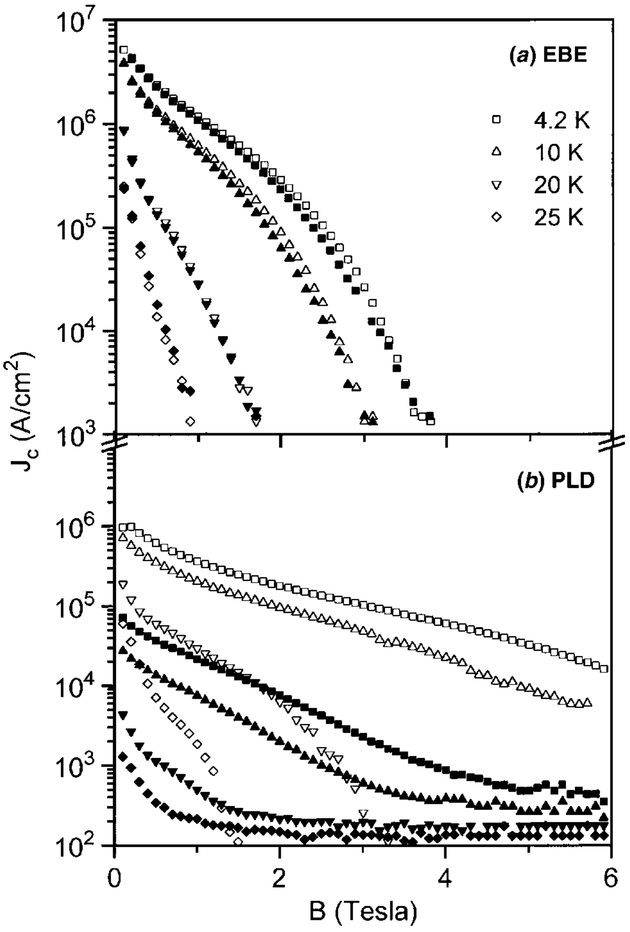

Figure 3. J_c as a function of B at different temperatures for both EBE and PLD films. The open and closed symbols correspond to before and after irradiation, respectively.

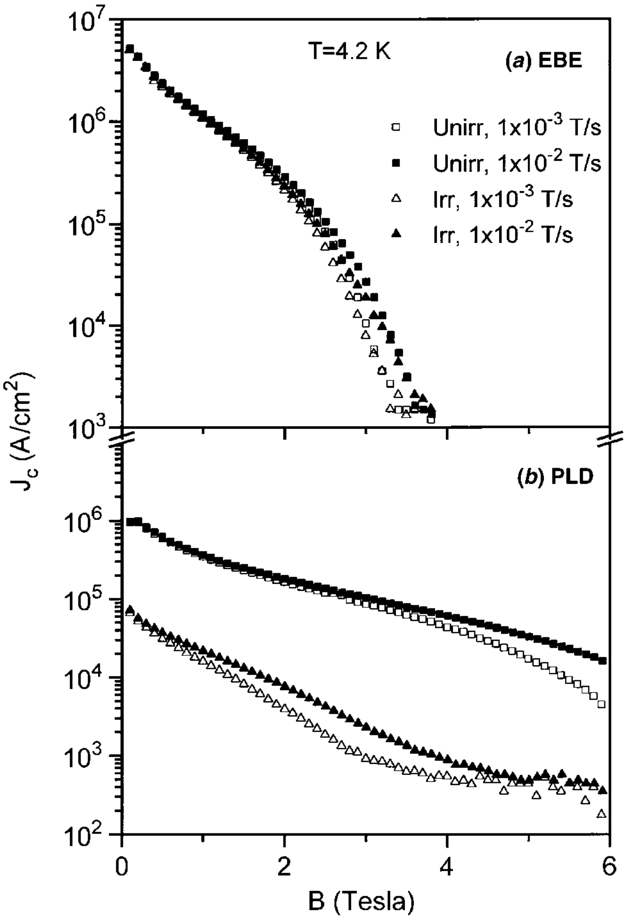

Figure 4. J_c as a function of B at different sweep rates for both EBE and PLD films.

the magnetization loop by the formula $J_c = (20\Delta M/Va)(1 - a/3b)$, where V is the superconducting volume, and a and b are the length and breadth of the film respectively [13]. The estimated $J_c(B)$ at different temperatures for both EBE and PLD films before and after irradiation are shown in figures 3(*a*) and (*b*) respectively. The high values of J_c for both EBE and PLD pristine films reflect their high quality. At lowest field, the $J_c(4.2\text{ K}) = 5 \times 10^6$ A cm^{-2} for unirradiated EBE film is found to be much higher than $J_c(4.2\text{ K}) = 1 \times 10^6$ A cm^{-2} observed for unirradiated PLD film. However, at high fields say $B = 3$ T, the former shows a $J_c(4.2\text{ K}) = 1 \times 10^4$ A cm^{-2}, which is much lower than $J_c(4.2\text{ K}) = 1 \times 10^5$ A cm^{-2} observed for the latter. Note also that in case of the former film, the J_c becomes nearly zero for $B > 3.5$ T, whereas in case of the latter film the $J_c > 1 \times 10^4$ A cm^{-2} even for B as high as 6 T (see figures 3(*a*) and (*b*)). Note that, in comparison to the EBE film, smaller grain size, higher ρ_n and lower T_c of the PLD film apparently improve the supercurrent carrying capacity in high fields. After irradiation, in the case of the EBE film (see figure 3(*a*)), at low fields J_c does not change much. At high fields and especially at high temperatures, J_c shows a slight increase with irradiation. However, since T_c also improves with irradiation, the latter observation needs to be taken only cautiously. In contrast to the EBE film, the PLD film shows a different behaviour after irradiation. Firstly, just like the degradation in T_c, severe reduction in J_c is also observed after irradiation (see figure 3(*b*)). For all temperatures, the J_c at low fields decreases by more than an order of magnitude after irradiation. Secondly, note that before irradiation the J_c steeply tends to zero for $B > 3.7$ T (at 20 K) and $B > 1.7$ T (at 25 K) (see figure 3(*b*)). However, after irradiation, for all temperatures the J_c after an initial rapid degradation does not show any tendency to become zero with increasing magnetic fields (see figure 3(*b*)). This shows that although heavy ion irradiation leads to a considerable degradation of superconducting properties in PLD film, the same irradiation also creates supercurrent channels that survive at very high fields.

Figures 4(*a*) and (*b*) show $J_c(B, 4.2\text{ K})$ at two different sweep rates for both EBE and PLD films before and after irradiation, respectively. In case of both films, smaller $J_c(B)$ values are observed at lower sweep rates. This clearly reveals the presence of flux creep that leads to relaxation of irreversible magnetization responsible for the formation of M–B loops. At lower sweep rates, irreversible magnetization at any fixed value of applied B in the M–B loop (see figure 2) gets more time to relax and proportionately the loop width is smaller (i.e. the $J_c(B)$ is smaller). As seen from figure 4, in the case of the EBE film, the J_c decreases by around a factor 3.5 at $B = 3$ T, whereas in the case of the PLD film the decrease in J_c is nearly negligible at the same field. After irradiation, the suppression

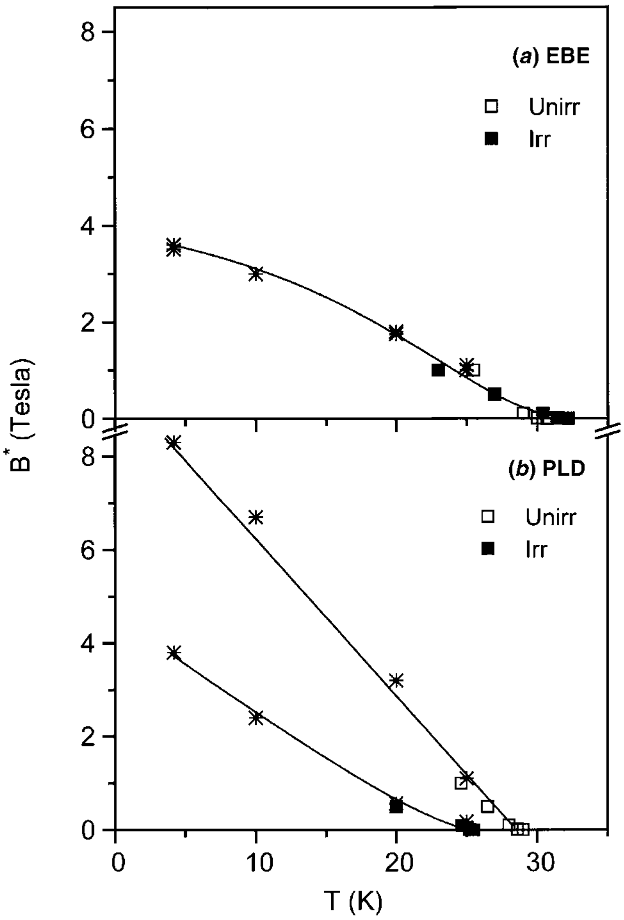

Figure 5. Irreversibility field B^* as a function of T for both EBE and PLD films. The 'square' symbols represent the diamagnetic onset temperatures as seen in ZFC $M(T)$. The 'star' symbols represent the $J_c = 1 \times 10^3$ A cm^{-2} criterion.

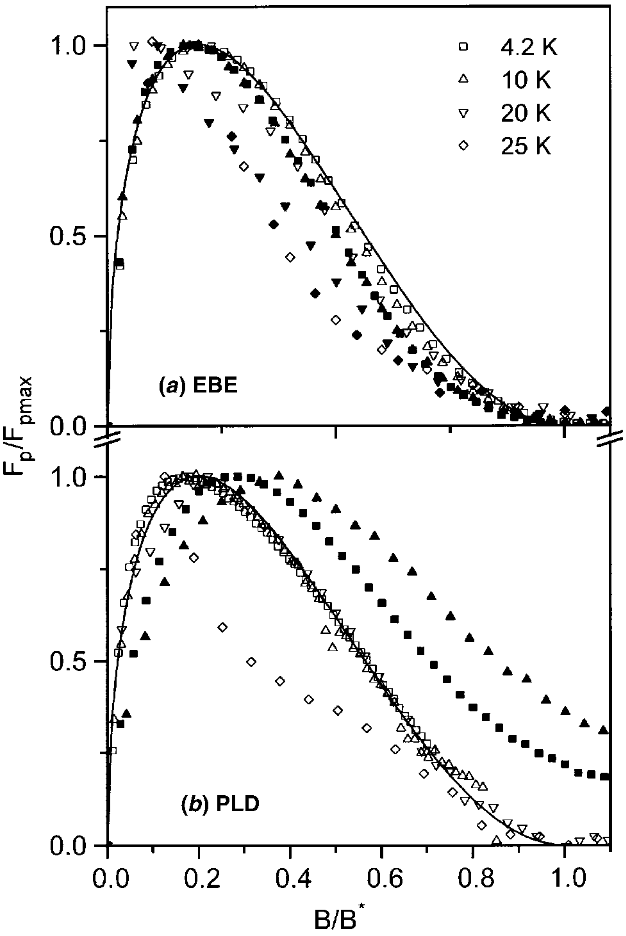

Figure 6. Reduced volume pinning forces as a function of reduced field at different temperatures for both EBE and PLD films. The open and closed symbols correspond to before and after irradiation, respectively. The solid curve represents the theoretical function $b^{0.5}(1 - b)^2$.

in $J_c(B)$ due to flux creep is nearly unchanged in the former and becomes much higher in the latter. Comparing these results with the observed differences of J_c in both films and effects of irradiation in them (figure 3), it may be concluded that flux creep increases in samples with smaller J_c. As also seen from figure 4, for both films, the flux creep is found to enhance with increase in magnetic field. The latter result indicates that the barrier for flux creep decreases with increasing magnetic field. In other words, as in HTSC [14], the pinning barrier in MgB_2 also decreases with an increase in magnetic field.

To highlight the differences in superconducting properties of EBE and PLD films arising from their different T_c and heavy ion irradiation, we plot in figures 5(*a*) and (*b*) the irreversibility line $B^*(T)$. The characteristic values of $B^*(T)$ were identified as the temperature where, within the resolution of VSM, the ZFC value of $M(T)$ measured in a constant applied field merges with the background zero line (see figure 1). These values of $B^*(T)$ were found to match well with the constant $J_c(B,T) = 10^3$ A cm^{-2} criterion in figures 3(*a*) and (*b*). The characteristic $B^*(T)$ points using the latter constant $J_c(B,T)$ criterion are also included in figures 5(*a*) and (*b*). As seen clearly from the figure, after irradiation, $B^*(T)$ shows no significant change for the EBE film, whereas for the PLD film $B^*(T)$ shows a strong suppression. This result is in line with the above mentioned behaviour of T_c and J_c after irradiation in these films.

Now we look into the nature of pinning that may exist in the films. This information can be obtained from the observed scaling of the volume pinning forces $F_p(B) = BJ_c(B)$ measured at different temperatures. The $F_p(B)$ curves can be scaled by plotting the reduced $F_p(B)/F_{pmax}$ as a function of reduced magnetic field $b = B/B^*$. In figures 6(*a*) and (*b*) we depict such plots for EBE and PLD films, respectively, both before and after irradiation. We would like to state here that in case of the PLD film, after irradiation, $J_c(B)$ becomes independent of B at $T \geqslant 20$ K (see figure 3(*b*)) and the corresponding $F_p(B)$ data do not even show a bell shape and are not included in figure 6(*b*). As seen from figure 6, for both pristine films EBE at $T \leqslant 10$ K and PLD at $T \leqslant 20$ K the data fall on the same curve. The scaled data can be reasonably fitted (see figures 6(*a*) and (*b*)) by the theoretical curve $f = Ab^p\ (1 - b)^q$ (where A is a numerical constant) with $p = 0.5$ and $q = 2$ that is well known for flux line shear (FLS) as a pinning model [15, 16]. Within this model, in the presence of few strong pinning centres, channels with weaker superconducting properties across the sample can allow the flux lines to shear flow along them and determine the $F_p(B)$. For the pristine films EBE at $T \geqslant 20$ K and PLD at $T = 25$ K, the data deviate from the FLS-model curve (see figures 6(*a*) and (*b*)). After irradiation the data also show deviation that increases with increasing temperature. Within the FLS mechanism such deviation at high fields ($b > b_{\max}$ in figures 6(*a*) and (*b*)) indicates the presence of distribution in the FLS channel width and its superconducting properties, i.e., identical FLS channels cannot describe the system.

Finally, we try to show that the grain boundaries (GBs) are a strong candidate for constituting FLS channels in the films. In polycrystalline MgB_2 it was recently [17] observed that the typical width of the GB lies in the range 5–20 nm and has a metallic nature. Both PLD and EBE films also show a GB/intergrain-channel width ~20 nm [18]. Considering also that GB width $\geqslant \xi_{ab}$ (~7 nm), the GBs may thus be proximity-coupled superconductor with weak pinning [19] and act as FLS channels. The observed deviation from an ideal FLS model at high temperatures and with irradiation may be understood now. Since the GBs are the regions of inhomogeneities/disorder, both high temperatures and irradiation can lead to a further distribution in channel properties. The deviation at low fields can also occur due to the presence of additional Josephson-coupled GBs in the samples. The grain size of the films will also influence the J_c. For instance, in comparison to the EBE film, the PLD film with much smaller grain size is expected to have a higher FLS channel cross-sectional area. Secondly, the effect of heavy ion irradiation in MgB_2 seems to be also dependent on the grain size. The irradiated PLD film shows a typical flow pattern indicative of significant mass movement on the surface [18] that may explain the severe degradation of T_c and J_c in it.

4. Conclusions

We studied the variation of $J_c(B)$ and the pinning mechanism in two differently synthesized MgB_2 thin films before and after heavy ion irradiation. In comparison to electron beam evaporated film, the pulsed laser deposited film is found to be more robust at high temperatures and high applied magnetic fields with regard to $J_c(B)$. We find that heavy ion irradiation has nearly no effect on $J_c(B)$ and $B^*(T)$ of the EBE grown MgB_2 film, whereas it severely degrades similar properties in the PLD grown film. The underlying pinning mechanism in both films seems to be governed by the flux line shear model. The grain boundaries in these *c*-axis oriented films can act as flux line shear flow channels. Higher temperatures and heavy ion irradiation can result in distribution of the channel width and its superconducting properties. We may thus conclude that though the J_c might not be weak-link limited, still the proximity-coupled large width GBs may limit the J_c at high temperatures in MgB_2.

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