Low-field scaling behaviors of global flux pinning in Tl-Ba-Ca-Cu-O thin films

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The scaling behaviors of global pinning force density ($\mathbf{F}_p = \mathbf{J}_c \times \mathbf{B}$) in dc sputtered Tl-Ba-Ca-Cu-O superconducting thin films as functions of both the reduced magnetic field and temperature were investigated by direct transport measurements for fields up to 4 kOe. It was found that, depending on the film granularities (originating from different deposition and annealing conditions and characterized by the temperature and magnetic-field dependencies of critical currents), vastly different scaling behaviors were manifested. For films with very high critical current densities, where the intragranular behaviors prevailed, the conventional surface core pinning by some high-density normal pinning centers combined with a significant flux creep appeared to be the predominant limiting mechanisms for the critical current densities, whereas in more granular films, suggestive collective-pinning effects arising from the weakly pinned fluxoids in the intergranular networks were observed.

Critical current densities (J_c) typically observed in thin films of high-T_c oxide superconductors are very high, much higher than those in bulk samples and somewhat higher even than those in single crystals. It is clearly of interest to establish the origin of the flux pinning in these films to understand the underlying mechanisms of these high critical current densities. Recent efforts of theoretical anlayses and direct imaging by scanning tunneling microscopy (STM) on $YBa_2Cu_3O_{7-\delta}$ (YBCO) thin films[1,2] have suggested that a high density of both point and line defects as the predominant pinning centers probably could account for the high critical current densities observed in these materials. However, quantitative correlations between these metallurgical defects and the critical current densities are still far from conclusive. Alternatively, studies of flux pinning by measuring the macroscopic pinning force density (F_p) as functions of the applied field and temperature to delineate the overall interactions between the fluxoid lattice and the pinning centers have been well established as an effective method of understanding the active pinning mechanisms for type-II superconductors.[3] Such studies have been carried out by various authors in both LaSrCuO[4] and $YBa_2Cu_3O_{7-\delta}$ systems.[5,6] Unfortunately, these experiments were either carried out in a relatively high-field regime,[4,5] where the bare vortex-defect interaction is complicated by the interactions between vortices, or by magnetic measurements,[6] making it difficult for a direct comparison with existing models.

Recently, with the success of making high quality Tl-Ba-Ca-Cu-O superconducting thin films which exhibited a zero-field critical current density exceeding 10^7 A/cm^2 at 87 K by a refined two-step (deposition + annealing) process, we have been able to perform a preliminary study on flux pinning in these films.[7] It is interesting to find not only that none of the weak-link models were sufficient to account for the observed magnetic and transport properties but also that the scaling behavior of the global pinning force density of these films showed a strong resemblance to conventional type-II superconductors with a flux-creep kinetic factor of about 10 times larger.[7] In this paper, additional results on the scaling behaviors of pinning force density derived from direct four-probe transport measurements for both high and lower critical current density Tl-Ba-Ca-Cu-O thin films will be reported together with detailed comparisons. It was found that, depending on the film granularities (originated from different deposition and annealing conditions and were depicted by the temperature and magnetic-field dependencies of the critical current densities), vastly different scaling behaviors were manifested. Since only relatively small magnetic fields were applied, the results presumably reflect the bare vortex-defect interactions and can reveal important facts about the flux-pinning processes active in these materials.

Thin films with a typical thickness of 1 μm were prepared by a single-target dc sputtering method previously reported.[8] Despite the stoichiometric composition used for preparing the targets, the films initially deposited were amorphous and not superconducting, as commonly obtained in various deposition processes used for preparing Tl-Ba-Ca-Cu-O thin films.[8-10] More interestingly, the final quality of the films obtained was found to be have a strong dependence on the subsequent annealing conditions employed. The details of the preparation conditions can be found in some of our previous publications.[7,8] It is interesting to note that although the final film properties can be varied vastly by the detailed annealing conditions all the films produced exhibited c-axis oriented characteristics as revealed by thin film x-ray diffractions. All the films used in this study were patterned into a 10-μm-wide, 125-μm-long bridge by standard lithographic processes[11] for subsequent transport measurements by the usual four-probe method with the magnetic field applied perpendicular to the film surface. To determine the critical current densities of the films a practical criterion of 1 μV/cm has been adopted in all cases.

In Fig. 1 the temperature dependence of zero-field critical current $[J_c(T)]$ for two different type of films were shown for comparison. As is evident from Fig. 1(a) for films with better qualtities, typically with T_c higher than 100 K and $J_c(77\ \text{K}) > 5\times10^6$ A/cm^2, the $J_c(T)$ shows a striking linear behavior over a wide range of temperatures. As we have pointed out earlier,[7] instead of attributing this to the usual weak-link limiting models for an explanation, this could well be interpreted as a consequence of creep-limited[12,13] manifestation of J_c. In the Anderson-Kim model,[12,13] J_c should vary as

$$J_c \approx J_{c0}(1-\alpha t-\beta t^2) \qquad (1)$$

for $t=T/T_c \ll 1$. Here the quadratic term is based largely on the temperature dependence of thermodynamic quantities, while the linear term is determined by the kinetics of flux creep. In this model,

$$\alpha \approx [kT_c/F_p(0)]\ln(\nu_0/\nu_{\min}) , \qquad (2)$$

where $\nu_{\min}$ is the minimum creep velocity detectable with the sensitivity of the voltage measurement and $F_p(0)=(H_c^2/8\pi)(4\pi\xi^3/3)(4\ln\kappa)^{3/2}$ is the maximum pinning energy of a point center with dimension of ξ. The slopes estimated from the inset of Fig. 1(a) gave $\alpha \approx 1.1-1.2$, about an order of magnitude larger than that in conventional superconductors,[12] indicating that flux creep is playing a much more important role in these materials. In contrast to these better quality films, films with poorer grain connectivities, i.e., more granular (as seen from the grain morphologies by SEM), vastly different $J_c(T)$ were observed. As shown in Fig. 1(b), for these more granular films, not only the absolute J_c values are orders of magnitude lower but the $J_c(T)$ shows a much more complicated behavior similar to that observed in granular $YBa_2Cu_3O_{7-\delta}$ films by Jones *et al.*,[14] where the $J_c(T)$ was attributed to the combined effects of the SNS weak links and thermally activated flux creep. Details of the analyses in terms of this model and their physical implications will be reported separately. In any case, it is interesting to note here that the extremely similar $J_c(T)$ behavior exhibited by the films shown in Fig. 1(a) suggests that essentially the same pinning mechanisms are active in these films, whereas for films shown in Fig. 1(b) totally different pinning mechanisms are clearly acting and further studies are clearly needed.

In Fig. 2, the typical field dependencies of critical current densities $[J_c(H)]$ at several temperatures for the two types of films were shown to elucidate further the differences among them. It is noted immediately that the $J_c(H)$ for the high-J_c films were reversible in field, whereas that for granular films showed significant hysteresis. The insets in both Figs. 2(a) and 2(b) were plotted to demonstrate the more detailed features. While the $J_c(H)$ hysteresis found in most granular high-T_c materials were well accounted for by the effects of trapped magnetic flux in the intergranular region during field cycling,[15] the reversible $J_c(H)$ remained to be explained. Intuitively, it can be interpreted as a result of the absence of pinning and/or weak-linked regions. Hence, no trapped flux is available to give rise to the $J_c(H)$ hysteresis. As is evident from the high-J_c values and the $J_c(T)$ behavior shown in Fig. 1(a), it is indicative that strong intragranular pinning could be predominant in these high-quality films.

To further delineate exactly what kinds of pinning are active, it is always instructive to go through the practice of analyzing the scaling behaviors of pinning force densi-

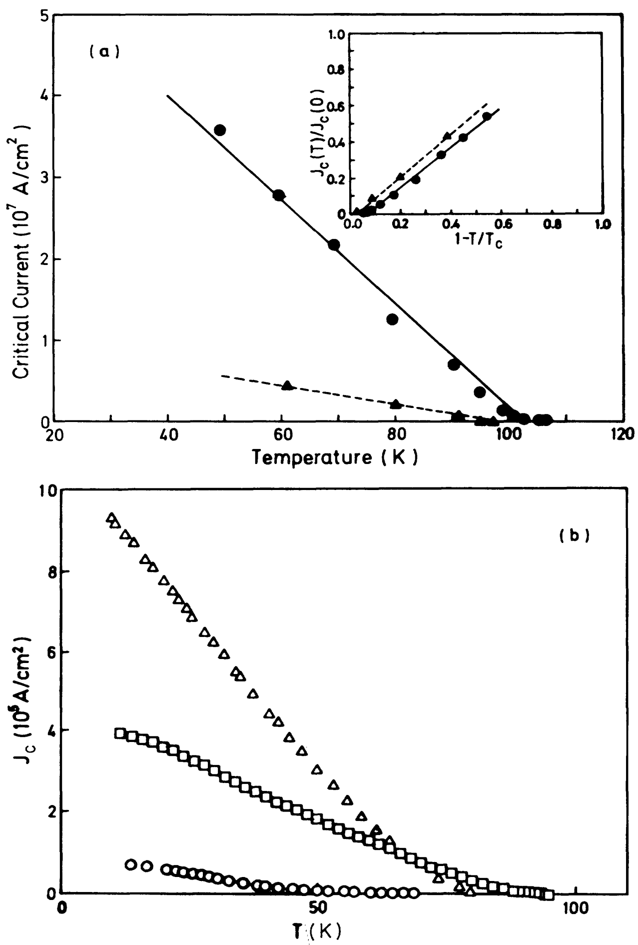

FIG. 1. (a) The $J_c(T)$ results for two of the high-J_c Tl-Ba-Ca-Cu-O films plotted in linear scales, showing the striking linear behavior in a wide range of temperatures. The inset depicts the normalized values of $J_c(T)/J_{c0}$ as a function of reduced temperatures. The curves are the fits to Eq. (1) with slopes of $\alpha \approx 1.1$ and $\alpha \approx 1.17$ for solid and dashed curves, respectively. (b) The $J_c(T)$ results for three of the lower J_c granular films prepared with a similar method but slightly different annealing conditions.

ties as functions of temperature and applied field.[3,16] The basic idea for the scaling studies is to separate the effects of temperature and magnetic field on the interactions between the pinning centers and quantized fluxoids existent in the mixed-state of type-II superconductors. For conventional superconductors with well-defined upper critical fields, H_{c2}, and pinning centers, F_p was found to scale as

$$F_p \approx A[H_{c2}(T)]^{2.5} f(h) \tag{3}$$

by Fietz and Webb.[17] Here A is a constant related to material parameters, $f(h)$ is a function determined by the detailed pinning mechanism in operation, and h is the reduced field defined as $h = H_{\text{appl}}/H_{c2}(T)$. With the usual reducing processes,[7,16] the typical results of the pinning force density scaling for two types of films studied are shown in Fig. 3. The choices of $H_{c2}(T)$ can be obtained by extrapolating the $J_c(H)$ data to $J_c(H,T)=0$ or by fitting the high-field data to an assumed pinning mechanism (e.g., $F_p \approx aH + b$ for individual pinning and $F_p \approx aH^2 + b$ for flux-line-lattice shearing).[16] It is noted here that the choice of a particular extrapolation method does not affect the apparent scaling observed. Thus, the well-behaved scaling behaviors with both temperature and field exhibited in Fig. 3(a) for high-quality (Tl)-films clearly demonstrate an unique pinning mechanism is predominant in the H-T domains studied. A logarithm-logarithm plot for F_p(max) as a function of $H_{c2}(T)$ gives a slope of 2.6 in fairly agreement with that predicted by Fietz and Webb,[17] further reinforcing the above statement. To see exactly what sort of pinning is acting, we further analyzed the data following the procedure suggested by Dew-Hughes.[18] The solid curve shown in Fig. 3(a) shows a fit with a function of $F_P \approx 5h^{0.6}(1-h)^{2.2}$,

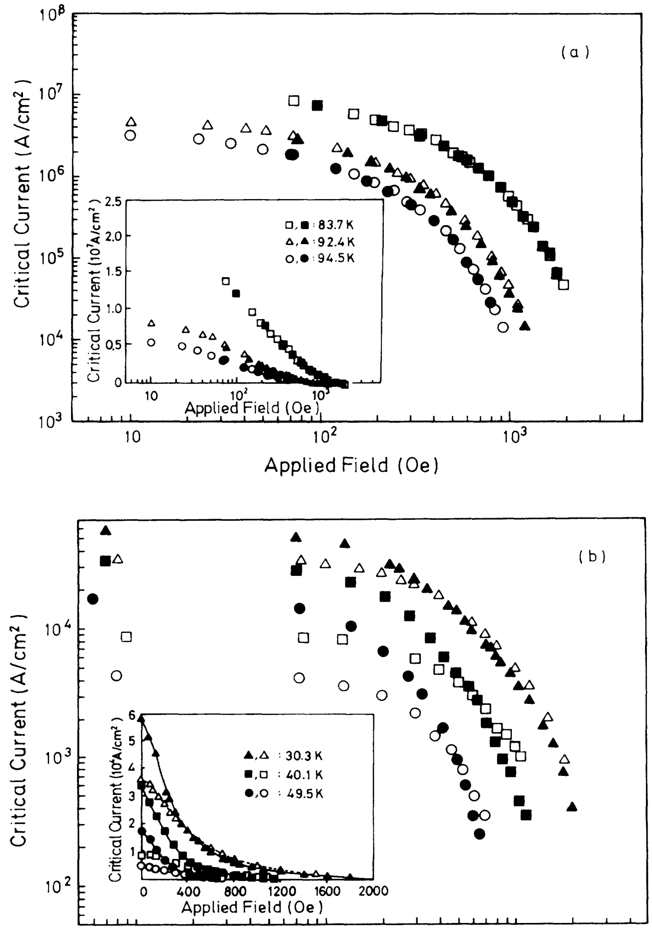

FIG. 2. (a) $J_c(H)$ for one of the high-J_c films at three different temperatures for one applied field cycle. The solid symbols represent the results obtained when the field was increased whereas the open symbols are for the reversed field. The inset shows the same data in semilogarthmic scale to delineate the possible hysteretic behavior. It is evident that there is no apparent $J_c(H)$ hysteresis in these films. (b) Similar results as (a) for one of the lower J_c films. The significant hysteretic behaviors in $J_c(H)$ are clearly evident.

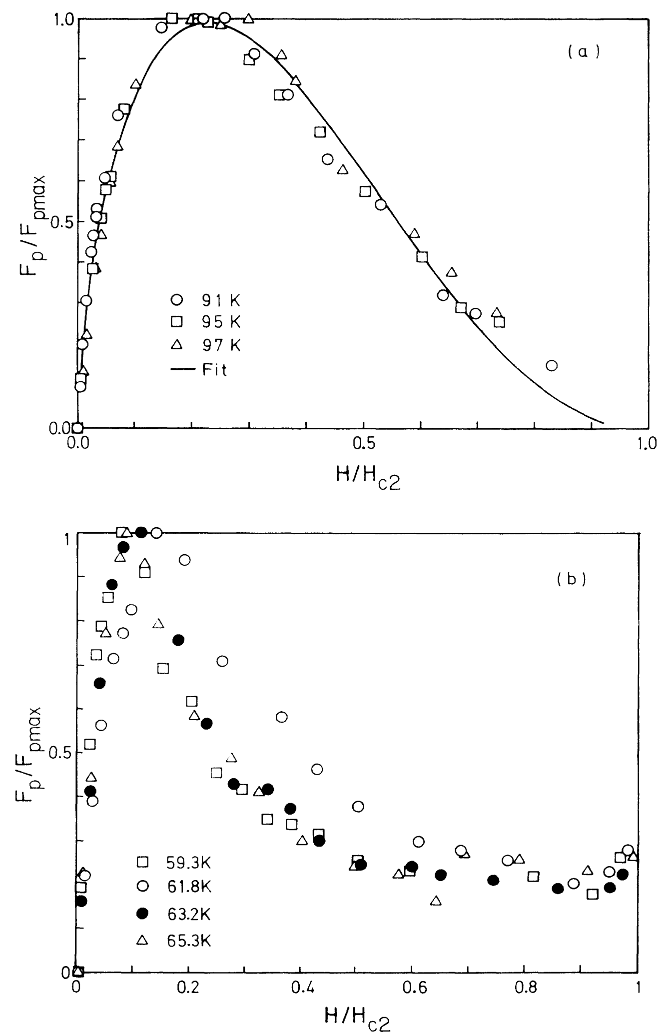

FIG. 3. (a) The normalized pinning force density as a function of reduced field for one of the high-J_c films at three different temperatures. The solid curve shows the fit to the data with a function of $F_p \approx 5h^{0.6}(1-h)^{2.2}$. (b) Similar results as (a) for one of the granular films.

similar to that found in single crystalline $YBa_2Cu_3O_{7-\delta}$ films by Nishizaki *et al.*[19] The peak position (at $h \approx 0.2$) and the functional form suggest that the observed scaling could well be manifestations by surface core pinning with an interaction length of the order of ξ of some normal pinning centers. The small discrepancies between the obtained fitting function and the model at high fields presumably was a consequence of the large creep kinetics factor discussed previously and the relative high measuring temperatures studied. In any case, direct microstructure analyses for identifying the specific defects existing in these films is clearly helpful for clarifying this issue and is currently undertaken.

In contrast to the behavior described above, the scaling of F_p for the granular films is clearly absent, as shown in Fig. 3(b). We evidently found that the F_p did not scale with the $H_{c2}(T)$, either. The results suggest not only pinning mechanisms for these films can be vastly different from the conventional superconductors but the definition of the "$H_{c2}(T)$" may need to be reconsidered, as well. As is suggested by the $J_c(H)$ data shown in Fig. 2(b), the intergranular features for these films play an important role in transport properties. As a result, we have replotted the data by using the procedures shown in the inset of Fig. 4. The "$H_{c2}(T)$" was then redefined as the separation point field-cooled and zero-field-cooled J_c at each temperature and field. It is noted here that since the measurements were performed by direct transport measurements, the "$H_{c2}(T)$" thus obtained may have totally different physical meaning from the "irreversibility lines" commonly obtained by magnetic relaxation measurements.[20] Nonetheless, the well-behaved scaling in F_p obtained (Fig. 4) by this procedure is astonishing and representing yet another predominant pinning mechanism that is active in these granular films. As was evident from the $J_c(T)$ and $J_c(H)$ results shown in Figs. 1 and 2, these films were well described by SNS weak links, their transport properties are better understood in terms of the weak-link systems in the presence of giant Josephson vortices.[14,21] Indeed, if one simply compares the J_c values in the same field regimes studied in the present work, about 2 orders of magnitude difference in J_c obtained implying about the same order of magnitude difference in pinning energies were prevailing, which in qualitative agreement with that observed in totally epitaxial and in granular $YBa_2Cu_3O_{7-\delta}$ films by Jones *et al.*[14] In the language of flux pinning, a system with such a weak and randomly distributed pinning networks should give rise to collective-pinning effects on the fluxoids existent in the mixed state.[22] The peak position of F_p at around $h \approx 0.1$ and the peak effects near the "$H_{c2}(T)$"

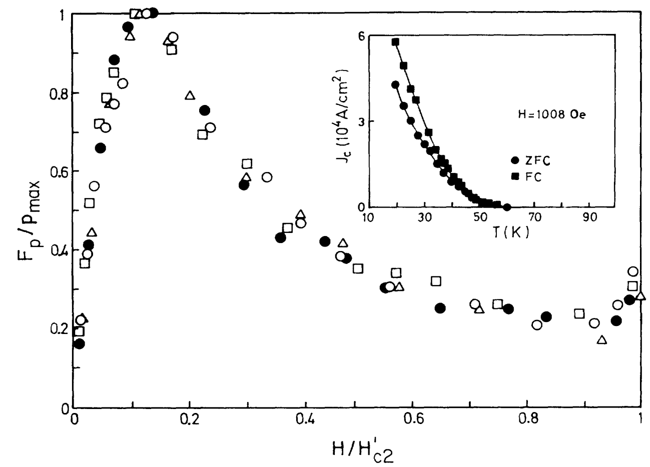

FIG. 4. Same results as shown in Fig. 3(b) but with a different definition of the $H_{c2}(T)$. The inset shows how the $H_{c2}(T)$ was determined. The results clearly replicate the major features of collective-flux-pinning effects.

manifested by these films (Fig. 4) clearly replicate the main features of collective pinning observed in the model systems of amorphous $Zr_{70}Cu_{30}$ and Nb_3Ge.[23,24] We note here that due to the flux-creep effects, the peak effects around the critical field were smeared out significantly. Detailed quantitative analyses on the parameters, such as the correlation length scales between the fluxoids and grain sizes in these granular films within the frameworks of the collective-pinning models, will be reported elsewhere.

In summary, in this paper, two distinct pinning mechanisms, as depicted by the detailed studies of the scaling behaviors of global pinning force densities in the relatively low field regimes, were found to be responsible for determining the critical current densities of Tl-Ba-Ca-Cu-O films with different granularities. For films with very high-critical current densities, where the intragranular behaviors prevailed, the conventional surface core pinning of some high-density normal pinning centers combined with significant flux-creep effects appeared to be the predominant limiting mechanisms for the critical current densities, whereas in more granular films, suggestive collective-pinning effects on the fluxoids due to weakly pinned intergranular networks with creep effects were found to be the predominant limiting factors.

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