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Systematics of transport critical-current-density hysteresis in polycrystalline Y-Ba-Cu-O

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We have characterized the dependence of the transport critical current density of the polycrystalline $YBa_2Cu_3O_{7-x}$ superconductors on magnetic field and temperature history as a function of the applied field. Large hysteretic enhancements in the critical current density are observed upon changing the field direction. The enhancements are a strong function of the maximum applied field. Further, a novel field dependence of the critical current density is observed in field-cooled samples. The results indicate that currents circulating within the grains strongly influence the transport current.

INTRODUCTION

Transport critical current densities, J_c's, of bulk ceramic high-temperature superconductors (HTS) have generally been restricted to values ≤ 1000 A/cm^2 at 77 K in self-field because of weak intergranular current transfer. In small magnetic fields (~ 100 Oe) the bulk J_c is rapidly depressed to values ≤ 10 A/cm^2, and $J_c(B)$ has been successfully described by a model consisting of an array of strongly superconducting grains joined by Josephson junctions.[1] Recently, several reports[2-4] have described observations of hysteresis in the bulk $J_c(B)$ measured on ceramic HTS materials at low magnetic fields that are in disagreement with the well-established critical-state model for the magnetic behavior of type-II superconductors[5] and with its extensions to granular superconductors.[6] In all of these reports the transport $J_c(B)$ is strongly enhanced over the value measured by monotonically increasing the applied magnetic field from zero to the measuring field H_m, if the field is first increased above H_m and then reduced to the value of the measurement. As shown below, these enhancements can exceed an order of magnitude at low magnetic fields and depend upon the maximum field to which the sample has been cycled.

For type-II superconductors in the mixed state, the intrinsic J_c is determined by the volume pinning force P_v, which stabilizes the vortex structure in the presence of a Lorentz force $\mathbf{J}\times\mathbf{B}$ caused by the interaction of an applied field with the gradient in the vortex density associated with the transport current. J_c is defined by the relation $\mathbf{J}_c\times\mathbf{B}=\mathbf{P}_v$ at which the Lorentz force can just be supported by the pinning structure. Phenomenological models of $P_v(B,T)$ generally begin with the assumption that $P_v(B,T)$ is a unique function depending only on the material microstructure and thermodynamic properties of the superconductor. This assumption would seem to rule out dependence of $J_c(B,T)$ on the path taken to arrive at (B,T). Nevertheless, small history-dependent effects have been observed on bulk conventional superconductors and have been reviewed by Kupfer and Gey.[7] These effects have been ascribed to the interrelation between the regularity of the flux lattice and the number of random pinning centers that can be occupied by flux lines in a given volume. Since the degree of perfection of the flux lattice may depend upon the path, history dependence of $J_c(B,T)$ is predicted. However, both predictions and experimental results on conventional superconductors indicate that J_c should be depressed after cycling to higher fields, a result opposite to the observations reported below. Hysteretic effects similar to those seen in ceramic HTS materials have been seen in granular aluminum, indium, and niobium thin-film bridges, but were not given an explanation.[8]

The purpose of this investigation is to determine the dependence of the critical current density on the detailed magnetic field and temperature history. It will be shown, in agreement with previous studies, that hysteretic effects are related to penetration of flux into strong pinning intragranular regions, and that the presence of intragranular critical currents enhances intergranular critical currents when the applied field is reduced from higher values. Because of the strong dependence of these hysteretic effects on the intragranular pinning, the most striking hysteretic effects are observed in field-cooled samples. We report new results on the field dependence of the critical current density in these field-cooled samples. The systematics on these history-dependent effects will be discussed in terms of their implications for the interrelation between intergranular and intragranular currents in high-temperature superconductors.

EXPERIMENTAL DETAILS

Samples were prepared from commercial[9] $YBa_2Cu_3O_7$ powder of average grain size 10 μm cold-pressed to 50 000 psi (3.4 kbar) and then fired in oxygen. The annealing cycle consists of a ramp to 965 °C at 200 °C/h, holding at 965 °C for 10 h, cooling to 400 °C at 25 °C/h, holding at 400 °C for 20 h, and then cooling to room temperature at

25 °C/h. A sample bar for critical-current-density measurements was cut with a 0.03-cm^2 cross section.

The room-temperature resistivity of this material is about 2600 $\mu\Omega$ cm with a superconducting transition temperature midpoint of 92 K, a 10% to 90% width of 2.5 K, and zero resistance near 89 K. dc susceptibility measurements (shielding) in a 100-Oe field showed a 93-K onset, a 74-K midpoint, and 95% flux exclusion at 7 K. The Meissner effect in 100 Oe was 24% at 7 K.

Magnetization (M) versus magnetic field (H) loops were measured at 7, 65, and 75 K in a superconducting quantum interference device magnetometer. Hysteresis in these data is indicative of intragranular critical currents. Values of the intragranular J_c determined using the Bean model,[5] $J_c = 15\Delta M/R$ where ΔM is the hysteresis in emu/cm^3, R is a typical particle size in cm ($\sim 10^{-3}$ cm for the results reported here), and J_c is in A/cm^2, are 1.8×10^6 at 7 K, 6.8×10^3 at 65 K, and 3.4×10^3 at 75 K, all at $H=0$. These values are similar to those measured in single crystals,[10] although with a more rapid fall off with increasing temperature. The position of the peak in the zero-field-cooled magnetization, consistent with complete penetration of the grains by flux, was also measured to be 1500, 200, and 100 Oe at 7, 65, and 75 K, respectively.

Transport critical current measurements were made with a short sample immersed in either a liquid-He or N_2 cryogen and with the current direction perpendicular to H. The contacts to the sample were made by soldering Ag wires to plasma-sprayed Ag pads 0.2 cm in diameter and 0.01 cm thick. Contact resistivities were in the 10^{-8} $\Omega\,cm^2$ range and caused no observable Joule heating. A 1-μV criterion was used for defining J_c; this corresponds to about 3 μV/cm or about 10^{-5} of the normal-state resistance at $T=75$ K and $H=0$. The current was increased to J_c in about 30 s while the voltage across the sample was monitored with a nanovoltmeter. Low-field measurements (< 1 kOe) were performed in a copper solenoid, and higher-field measurements were done in a superconducting magnet. The measurements presented here were all taken at 4 K where the hysteretic effects were maximized. However, we also observed hysteresis in the transport $J_c(H)$ over the range 65–75 K.

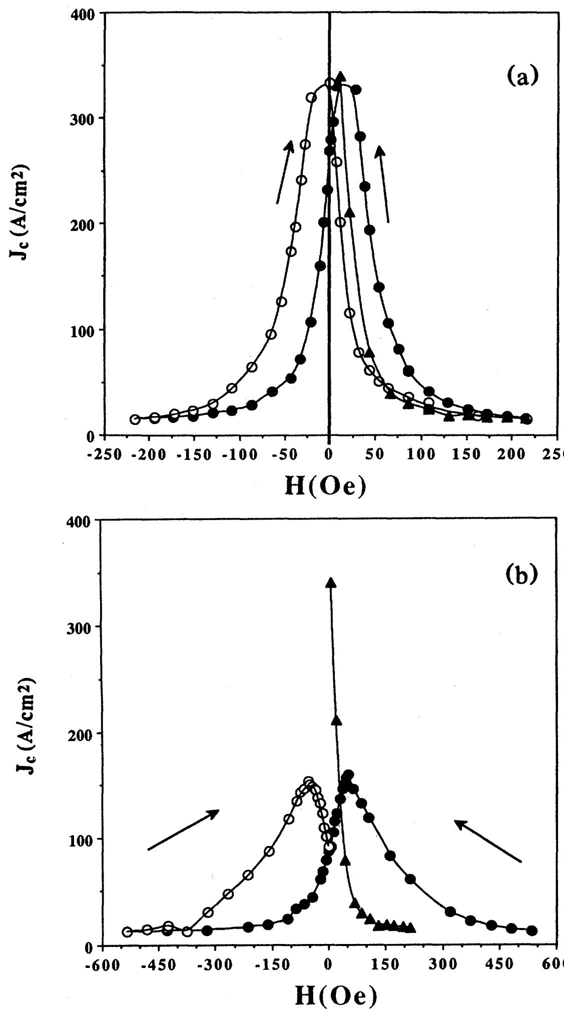

FIG. 1. Hysteresis in the critical current density J_c of polycrystalline Y 1:2:3 cooled in zero field to 4 K. (a) Virgin curve showing $J_c(H)$ in an increasing field (triangles), and decreasing- (filled circles) and increasing-field (open circles) branches after cycling to $H_{max}=225$ Oe. (b) Virgin curve (triangles), decreasing- (filled squares) and increasing-field (open squares) branches after cycling to $H_{max}=550$ Oe.

RESULTS AND DISCUSSION

Figure 1 illustrates the systematics of the hysteretic response of the transport J_c as a function of applied field for $T=4$ K for a zero-field-cooled (ZFC) specimen. The virgin curve critical current density at zero field is extrapolated because it exceeded the power supply capacity. The critical current density for this virgin curve drops precipitously ($\sim 1/B$) with increasing applied field as shown in Fig. 1(a). These field-increasing results are typical of ceramic superconductors and have been given a detailed analysis in terms of a Josephson-junction–Airy-current pattern by Peterson and Ekin.[1].

After increasing the field monotonically to $H_{max}=225$ Oe, J_c was measured as a function of decreasing field until $H=-225$ Oe, along the branch labeled J_{c1}(down), as shown in Fig. 1(a). Notable in this branch is the increased value of J_c, by as much as a factor of 3:1, as compared with the data for the virgin curve. The critical current density is seen to peak on decreasing the field at $H=\sim 20$ Oe, so that for $H=0$, the J_c value is substantially reduced as compared to the virgin ZFC measurement. J_c continues to decrease monotonically as H is decreased through negative field values. This curve asymptotically approaches the mirror image of the virgin curve as the applied field becomes more negative. Increasing the applied field from -225 Oe to positive values of H, the curve J_{c1}(up) is obtained. This curve is near to being the mirror image of J_{c1}(down). In actuality, mirror symmetry exists around the vertical line $H=\sim 6$ Oe, which does not appear to be a function of temperature or maximum applied field. This value is consistent with estimates of the self-field.

The curves shown in Fig. 1(b) illustrate the dependence of the hysteresis in the critical current density on the maximum applied field H_{max}. These curves are the result of cycling to fields of 550 and -550 Oe, respectively. Notable in this figure are the increased values of J_c at high fields and a significant increase in the field at which the peak in J_c occurs ($|H| \sim 60$ Oe). Thus for higher H_{max}, the behavior of the $J_c(H)$ peak is characterized by progressive broadening, shifting to higher fields, and a decreasing height. It should be noted that measures of the hysteresis, such as the position of the peak in J_c, do change monotonically with the maximum applied field, but the peak position appears to change most dramatically for applied fields which approximate the average H_{c1} (~ 200 Oe) of the grains. This emphasizes the importance of intragranular currents on the transport critical-current-density values. It suggests that the establishment of the intragranular currents and consequent phase relationships between grains acts to strongly influence the transfer of current between grains.

As first demonstrated by Evetts and Glowacki[4] (EG), these and other systematics of history-dependent bulk $J_c(B)$ behavior can be modeled by a Josephson array in which the magnetic field acting on the junctions is a sum of two terms, one the applied field and the other contributed by the intragranular flux gradients. On increasing the magnetic field from a ZFC condition, magnetic flux penetrates through the weak link network suppressing current flow through the Josephson junctions. The resulting transport $J_c(B)$ curve reflects an envelope function of the individual Fraunhofer diffraction patterns. However, upon exceeding H_{c1} for the grains, flux penetrates the grains in accordance with the critical-state model[5] (CSM). Diamagnetic flux gradients (currents) are established on the increasing field portion of the cycle. Upon reducing the field from H_{max}, the flux gradients and currents will be reversed, initially at the surface, and then extending further into the grains as the field is reduced. Because this reverse current produces a paramagnetic contribution to the magnetic moment of the grain, we will refer to it as a paramagnetic current. If diamagnetic *intragranular* currents add to the effect of the *intergranular* fields in suppressing J_c and paramagnetic currents subtract, then the general features of the observed $J_c(B)$ systematics are obtained.

In the EG model, the contribution from the grains is assumed to come simply from the dipole fields generated by the intragranular currents. This contribution to the field acting on the junctions is then proportional to the magnetic moment of the grains which adds to the applied field with the field increasing and subtracts on decreasing the field. At the peak the two contributions roughly cancel. Under this model, field increasing and decreasing curves should be simple translations of one another. In measurements beginning in a ZFC state, the flux gradients on decreasing from H_{max} will be both diamagnetic and paramagnetic, and their effect on J_c will be only partially canceling. In the field-cooled state the initial flux distribution is flat (for $T > T_c$) while a negative flux gradient is obtained below T_c, for decreasing fields, as flux is expelled. Therefore, measurements taken while decreasing H from an initially field-cooled (FC) state should be particularly revealing in that, according to CSM predictions, only paramagnetic currents will be present and cancellation of the applied field should be enhanced. This expected enhancement of hysteretic effects should provide a more sensitive test of the EG model.

Figure 2 shows measurements of the transport critical current density as a function of decreasing applied field at 4 K after the sample has been field cooled in various low magnetic fields. Points joined by a smooth curve correspond to the same FC value, which is represented by the point furthest to the right. Apparent from the figure is the significant enhancement of J_c in the FC state as compared to values obtained in the ZFC measurements, both in the field-increasing and field-decreasing runs. This supports the idea that intragranular diamagnetic currents in ZFC field-increasing measurements are contributing substantially to the suppression of J_c. These are largely absent in the initial FC state. A small Meissner flux expulsion, for $H > 200$ Oe, which was observed in FC magnetometer measurements on this sample, implies that only small flux gradients are present in the FC state.

On decreasing H from the FC value, J_c is observed to rise steeply to a peak at $H_{peak} \sim H_{FC} - (50\text{–}60$ Oe) and then to decrease as B is reduced to zero. If the peak in J_c represents a rough cancellation of the intergranular and intragranular effects, then the steep initial rise indicates that intragranular effects dominate the behavior of J_c. Furthermore, the weak-field dependence of $H_{FC} - H_{peak}$, as shown in the inset of Fig. 2, suggests that the intragranular contribution is not simply related to the average paramagnetic moment of the grains, in contrast to that predicted by the EG model.

Figure 3 shows FC results for the higher fields of 2000 and 3000 Oe, respectively. In both cases J_c rises steeply

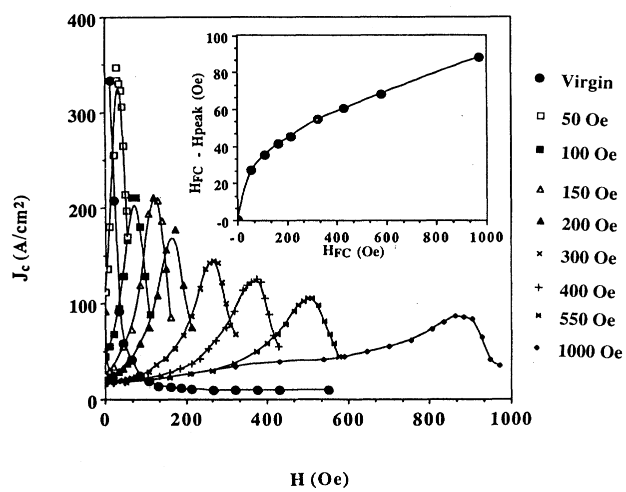

FIG. 2. Hysteresis in the critical current density of polycrystalline Y 1:2:3 material cooled in field to 4 K. Zero-field-cooled initial curve and field-cooled branches are shown in decreasing fields from starting values $H_{FC}=50$, 100, 150, 200, 300, 400, 550, and 1000 Oe. The inset shows the field dependence of $H_{FC} - H_{peak}$, where H_{peak} is the position of the maximum in J_c.

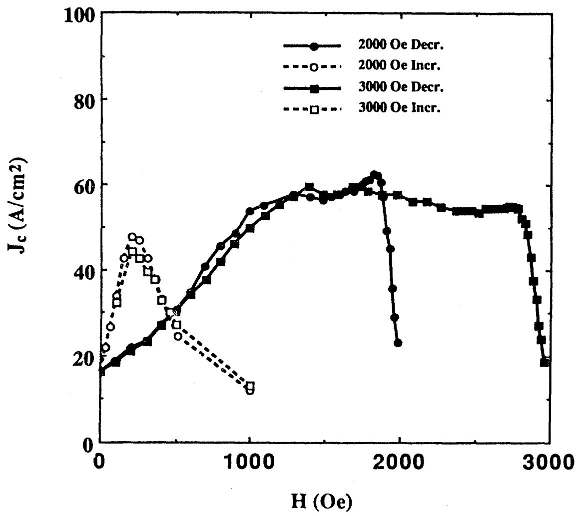

FIG. 3. $J_c(H)$ for samples field cooled at 2 and 3 kOe, with H decreasing to zero and then increasing to 1 kOe.

from its FC value and then reaches a plateau as the field is further decreased. Evident in the figure is also a saturation effect in that the results are identical in decreasing fields below ~1800 Oe. The initial steep rise for these high-field results indicates that paramagnetic currents generated at the surface of the grains are dominant in determining the transport J_c. These results are difficult to reconcile with the EG model in which dipolar fields generated by a ~200 Oe decrease in B would have to compensate for a 2800-Oe applied field. The heavy weighting of intragranular surface currents is further emphasized by the low-field turnaround behavior shown in Fig. 3 where after decreasing H to zero and then increasing, J_c again rises rapidly to a peak. In this situation the majority of the intragranular volume is still filled with paramagnetic currents and only the surface currents have been reversed.

These effects point to certain shortcomings in the EG model which originate in the simple superposition of the external field and the dipole fields of the grains. These results do seem to be adequately addressed by a recently developed coupled-phase Josephson-junction model in which the details of the intragranular critical state and its contribution to a composite Josephson-junction phase angle are addressed.[11] It should also be noted that these new systematics observed in the FC measurements are only characteristic of low-temperature data where the large intragranular J_c's are operative. A full discussion of the temperature dependence of these effects will be given in a subsequent work.

SUMMARY AND CONCLUSIONS

New results on magnetic-history-dependent transport critical current densities, in granular Y-Ba-Cu-O, have been presented for field-cooled measurements. These results are consistent with a mechanism which attributes the hysteresis in J_c to the effect of gradients in the pinned flux stored within the individual grains that obey the Bean critical-state model.[5] This interrelation between intragranular and intergranular currents has been previously observed by several groups. The field-cooled results presented here reveal new features not previously observed in ZFC studies. The peak in the transport J_c, generally attributed to a compensation of the applied field by the effect of paramagnetic intragranular currents generated in decreasing fields, is observed to move to higher fields close to the FC value. The steep rise to a maximum, the weak-field dependence of $H_{FC} - H_{peak}$, and the enhancement of the bulk J_c in the FC state all suggest that intragranular currents dominate the effect of intergranular fields upon transport J_c's at low temperatures. These new systematics are difficult to reconcile with the EG model, which is based upon a vectorial addition of the applied field and dipolar fields proportional to the magnetic moment of the grains.

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