

**Grain Boundary Transport Properties in  $\text{YBa}_2\text{Cu}_3\text{O}_x$  Coated Conductors**

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# Grain Boundary Transport Properties in $\text{YBa}_2\text{Cu}_3\text{O}_x$ Coated Conductors

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**Abstract**—Critical current data obtained as a function of magnetic field on an isolated grain boundary (GB) of a coated conductor and two other types of bicrystal GBs of  $\text{YBa}_2\text{Cu}_3\text{O}_x$  show a peak in the critical current and an unusual hysteresis. These results provide support for a new mechanism for enhanced GB critical currents, arising from interactions of GB vortices with pinned Abrikosov vortices in the banks of a GB, as suggested by Gurevich and Cooley. A substantial fraction of this enhancement, which can exceed a factor of ten, also occurs upon surpassing the critical current of the grains after zero field cooling. A bulk GB and thin film GBs show qualitatively identical results.

**Index Terms**—High Temperature Superconductors, Critical Current, Grain Boundaries.

## INTRODUCTION

THERE is evidence that the critical current density,  $J_c$ , of grain boundaries (GB) in high-temperature superconductors (HTS) does not drop as quickly [1] with magnetic field,  $H$ , as might have been expected from a simple Josephson junction model, in which the envelope of the Fraunhofer pattern goes as  $1/H$ . In very low fields, pinning of Josephson vortices by the meandering of thin-film, [001] tilt, bicrystal GBs in  $\text{YBa}_2\text{Cu}_3\text{O}_7$  has been demonstrated [2] to enhance  $J_c$ . However, as the spacing between Josephson vortices decreases in higher fields, this long wavelength pinning potential due to meandering becomes less effective (the meander size is  $\sim 0.5 \mu\text{m}$ ). Recently, Gurevich and Cooley [3] proposed a new mechanism for an enhanced GB critical current arising from pinned Abrikosov vortices in the banks of a GB which present a static, quasiperiodic pinning potential to pin GB vortices. Their calculations, that predict [3], e.g., a peak in  $J_c(H)$ , are in the low field limit, but the central concept can be extrapolated to higher fields. This pinning mechanism exhibits optimal effectiveness if the Abrikosov and Josephson vortices have the same spacing, i.e. when the magnetic flux density in the GB and the banks are

equal. In that case there is one potential well for pinning per Josephson vortex. A peak in  $J_c(H)$  is not uncommon in melt-textured and single crystalline  $\text{YBa}_2\text{Cu}_3\text{O}_7$  which are made without intentional GBs, but we are unaware of such direct experimental evidence in GB transport [4].

This paper reports critical currents, that are extracted from current-voltage curves of bicrystal GBs that show a peak in the GB critical current,  $I_{cb}(H)$ , and an unusual hysteresis that give considerable support to the central concept of the Gurevich-Cooley model [3]. At high fields, this support comes from the history dependence of  $I_{cb}(H)$  and the field profiles found in these bulk materials. We have measured  $I_{cb}(H)$  of the GB after either: (1) field cooling (FC) the sample in an applied field,  $H$ , to a temperature,  $T$ , from above the transition temperature,  $T_c$ ; or (2) increasing  $H$  after zero-field cooling (ZFC) to  $T$ . In low fields, the GBs exhibit a larger  $I_{cb}$  for FC, which is just opposite to the usual hysteresis for the grains of bulk materials (in which the larger internal fields associated with FC decrease the pinning and thus  $I_{cg}$ ). However, this is exactly the expectation of the Gurevich-Cooley model for GBs, since FC provides a larger Abrikosov vortex density in the banks that can more strongly pin GB vortices. Magnetization data obtained from one of the samples are consistent with features of the  $I_{cb}$  hysteresis interpreted in this framework, including the irreversibility field, above which, the internal flux profiles are nearly the same for FC and ZFC. Above the irreversibility field, a necessary expectation of Ref. 3 is that the GB transport should be indistinguishable between FC and ZFC and our data confirm this. Finally, in the ZFC case, after a sufficiently large current is applied such that vortices can both be injected in the banks and exhibit flux creep, the  $I_{cb}$  of the GBs is permanently increased thereafter, and by a considerable amount. This is consistent with the additional flux penetration caused by the supercritical current in the banks, although the full  $I_{cb}$  value for FC has not been achieved.

## EXPERIMENTAL

We have investigated a naturally formed GB occurring in a sample made by the RABiTS process [5], artificial boundaries formed by laser deposition on a bicrystal substrate and bulk boundaries formed by dual seeded melt texture growth.

The RABiTS process results in films with grain sizes that are typically  $100 \mu\text{m}$  allowing the study of isolated GB using photolithographic patterning techniques. An example of the alignment of a track with a single grain boundary is illustrated in Fig. 1. The grain boundary angle was estimated from electron backscatter Kikuchi patterns to be 11 degrees. A  $200 \text{ nm}$  thin  $\text{YBa}_2\text{Cu}_3\text{O}_x$  film was grown by laser ablation on a  $\text{SrTiO}_3$  bicrystal substrate with a 24 degree [001] tilt GB.

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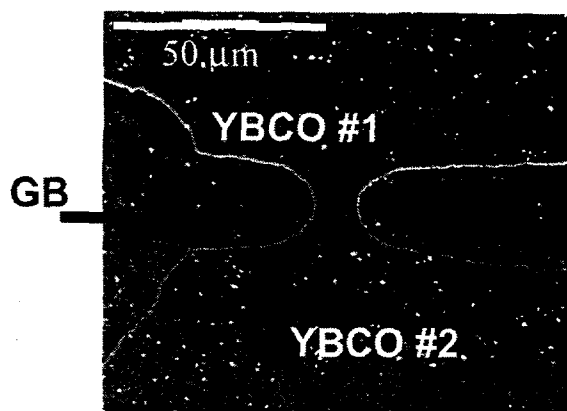


Figure 1. Isolated grain boundary in a  $\text{YBa}_2\text{Cu}_3\text{O}_x$  film made by the RABiTS process [5] with a GB angle of 11 degrees, close to [001] tilt. The GB was isolated from the coated conductor sample by the fabrication of a 13  $\mu\text{m}$  wide track containing the GB (arrow) using photolithography.

tilt GB. Photolithography was used to pattern narrow tracks crossing the boundary. The bulk GBs were grown by the cubic-seed-growth melt-texture processing which is described in detail elsewhere [6]. Sections containing  $90^\circ$  [100] symmetric tilt GBs were thinned to  $\sim 75 \mu\text{m}$  and the misorientation angle was verified by electron backscatter Kikuchi patterns to be  $90^\circ \pm 1^\circ$ . A solid state Nd-YAG laser was used to cut these plate-like sections into the shape sketched in the inset of Fig. 5. Four electrical contacts with resistance  $\sim 1 \text{ ohm}$  were attached with silver epoxy, as indicated, with the outermost ones used for applied current,  $I$ , and the innermost for voltage,  $V$ . For this L-shaped sample, the *macroscopic* applied current direction (thick arrows) is parallel to the Cu-O planes. The widths (lengths) of the arms are  $\sim 300 \mu\text{m}$  ( $\sim 600 \mu\text{m}$ ).

Transport properties were measured in a He-gas flow cryostat, initially using current pulses of 150 ms duration to reduce heating while retaining sufficient voltage sensitivity (see further discussion of heating below). The transition temperatures of the samples in ambient magnetic fields ( $< 1 \text{ Oe}$ ), were close to 90 K. The current-voltage measurements in a magnetic field were obtained after two specific field temperature sequences. In the first sequence, known as field cooling (FC), data were taken after cooling the sample in a field, which was applied above the transition temperature,  $T_c$ . Using the same field orientation, a second sequence increases the field after cooling from a temperature above  $T_c$  to  $T$  in zero field (ZFC). During the I-V measurements the current was perpendicular to the magnetic field. In the thin film samples the field was parallel to the c-axis, however, in the bulk sample  $H$  was perpendicular to  $c$ . The three samples studied here show a sharp onset of dissipation at the critical current, characteristic of a GB response (grains show a gradual development of the voltage as the current is increases due to flux creep effects). In the bulk sample a second kink in the IV data marks the grain critical current  $I_{cg}$  [10]. GB critical currents were determined at 1  $\mu\text{V}$  for the film boundaries, whereas a 0.1  $\mu\text{V}$  criterion was used for the bulk melt-textured GB sample. The differences in I(V) between FC and ZFC are shown in Fig. 2 for 77 K and 100 Oe in the coated conductor sample.

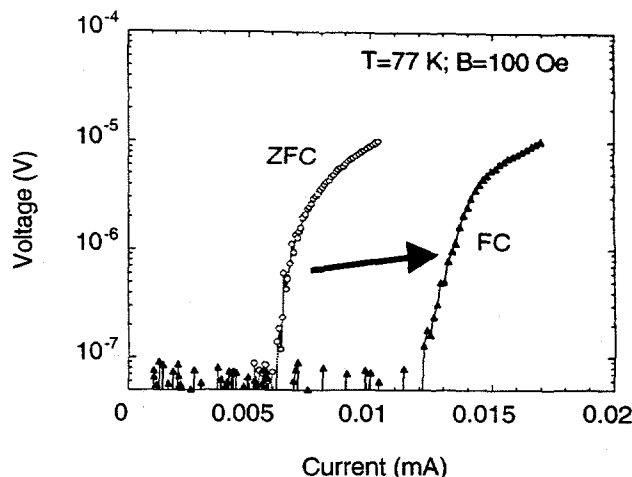


Figure 2. Dependence of current-voltage curves on the magnetic field history of the grain boundary in a coated conductor shown in Fig. 1. The GB critical current determined at 1  $\mu\text{V}$  increases from 6.8 mA after zero field cooling (ZFC) to 13.3 mA after field cooling (FC).

#### CRITICAL CURRENT ENHANCEMENT BY FIELD COOLING

The critical current data as function of the magnetic field are shown in Figs. 3-5 for the three samples investigated here. Though the samples were prepared in very different ways, the overall critical current behavior is remarkably similar. The difference between field cooling and zero field cooling are dramatic in low fields where the GBs exhibit a larger  $I_{cb}$  for FC. In particular the  $24^\circ$  [001] tilt GB samples shows a factor 10 increase in low fields. The larger critical current for FC is just opposite to the usual result for bulk materials (i.e., the larger internal fields associated with FC decrease the pinning and thus  $I_{cg}$ ). In addition, a broad peak in  $I_{cb}$  is seen in the ZFC branch for  $\mu_0 H \sim 0.05-0.2 \text{ T}$ .

There is a strong correlation of some of the distinctive features of the data in Fig. 5 with the bulk magnetization of the grains (banks), measured on one half of a GB sample at 77 K and shown in Fig. 6. The transport data at 77 K was qualitatively the same as Fig. 5, except the characteristic fields were about a factor of 2 larger, in excellent agreement with Fig. 6. Thus the deviation from the Meissner-like magnetization at  $\sim 0.01 \text{ T}$  signals the entry of flux into the grain and this corresponds to the beginning of the upturn of the grain boundary  $I_{cb}$  in the ZFC case. The grains are reversible above about 2 T and so is the GB. These results strongly hint that the GB  $I_{cb}$  is connected to the magnetic flux density in the grains.

The bulk, flat  $90^\circ$  [100] symmetric tilt GBs are somewhat special ones that are relevant to step-edge film junctions (devices), but not so important for coated-conductor applications. In addition, the field was applied *parallel to the ab-planes* resulting in an anisotropic pinning potential for the Abrikosov vortices [7]. However, the generality of the Gurevich-Cooley concept (i.e., the enhancement of  $I_c$  by decorating the banks of GBs with Abrikosov vortices) is clearly shown by the data from a coated conductor sample made by the RABiTS process [5] in Fig. 3 and the laser ablated sample in Fig. 4. For these samples the applied field was *parallel to the c-axis*, implying an isotropic pinning

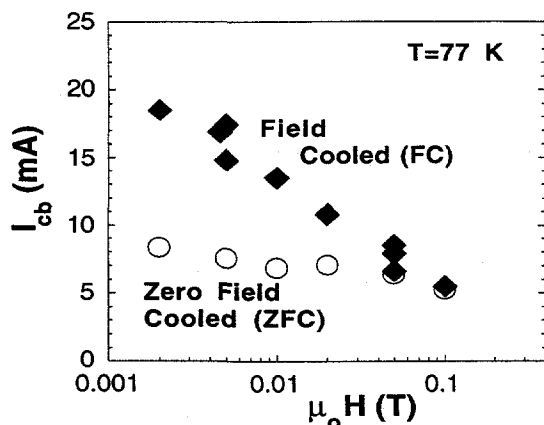


Figure 3. Critical current data,  $I_{cb}$ , taken from the coated conductor GB given in Fig. 1. From current voltage data such as presented in Fig. 2, taken as a function of field, the  $I_{cb}$  for FC (solid symbols) and ZFC (open symbols) were obtained.

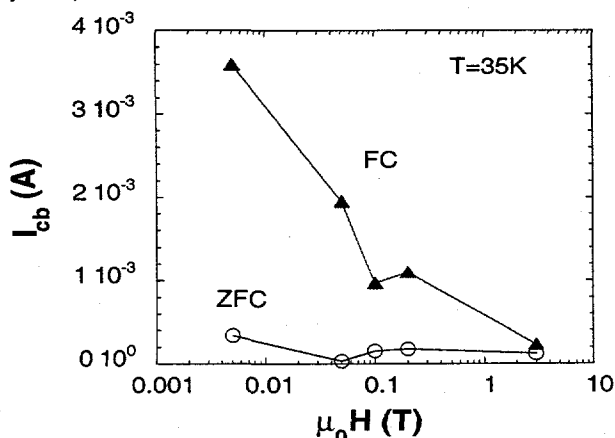


Fig. 4 Critical current density of a 24 degree [001] tilt grain boundary in a 200 nm thin film deposited using laser ablation.

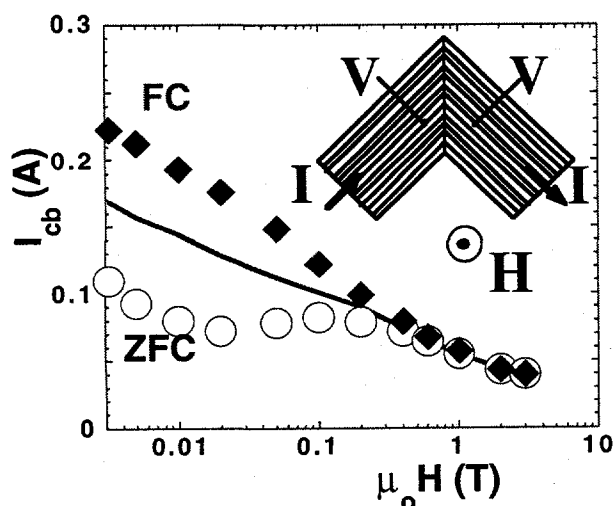


Figure 5. The  $I_{cb}$  data for FC (solid symbols) and ZFC (open symbols) at 84 K are shown for a grain boundary formed by melt texturing. The solid line is the irreversible (i.e., permanent) change in  $I_{cb}$  found upon exceeding  $I_{cg}$  after ZFC. The inset shows the L-shaped sample geometry.

potential for the Abrikosov vortices (note that the current flow is still perpendicular to the field). The data, shown in Fig. 3, are remarkably similar to those of Fig. 5 for the bulk, flat 90° [100] symmetric tilt GB, so this mechanism appears to be a general property of GBs. In addition, previous data [8] on artificial thin-film bicrystal GBs showed the same qualitative hysteresis as in Figs. 3-5.

## DISCUSSION

We propose [9] that the Gurevich-Cooley model [3] can explain this remarkable inverse hysteresis in terms of pinning of Josephson-like GB vortices by Abrikosov vortices pinned nearby in the banks. By a Josephson-like vortex, we mean the usual Josephson vortex in low applied fields, but as their density increases, neighboring GB and Abrikosov vortices overlap to significantly alter their structure, e.g., shape [6]. Gurevich and Cooley [3] proposed that sufficiently well-pinned vortices in the banks of a GB present a static, quasiperiodic pinning potential to pin such GB vortices, but it also automatically has the optimal spacing at each field. This magnetic interaction provides additional longitudinal pinning [3] to that resulting from inhomogeneities [2] of the Josephson current along a GB.

The following scenario provides a possible explanation for the detailed features of the data in Fig. 3 and 5. For ZFC, the field penetrates first into the GB, even if  $H < H_{c1g}$ , where  $\mu_0 H_{c1g} \sim 0.01$  T is the critical flux-entry field of the grains (banks). Then the initial decrease of  $I_{cb}$  (for  $\mu_0 H < 0.01$  T) is likely due to the diluting the average pinning strength as the GB vortex density increases. For  $H > H_{c1g}$ , the surface barrier is overcome so vortices can enter the grains. It is those situated next to the GB that can provide pinning by the Gurevich-Cooley mechanism. It is not clear if these vortices are injected at the outer surfaces of the grains and migrate to the GB, or if their origins are the GB vortices themselves, such that they are injected at the GB interfacial surface (this could be relevant for non-uniform critical-state flux profiles in the grains). However, it is these vortices that likely cause the increase in  $I_{cb}$  with field shown most clearly in Fig. 5 for ZFC and  $\mu_0 H$  between 0.02 and 0.1 T. For the FC curve

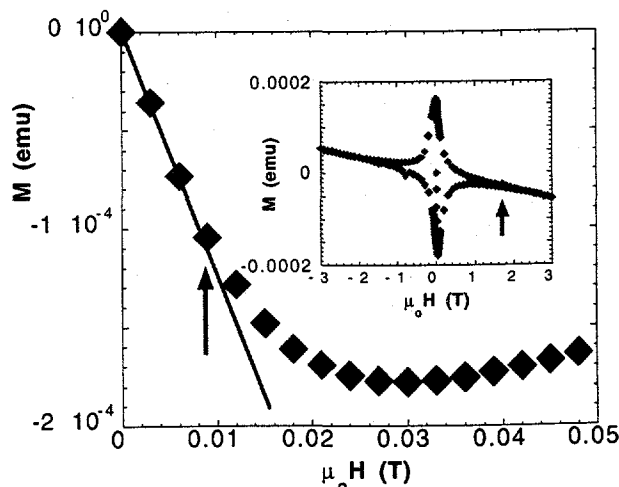


Figure 6. The bulk magnetization of one half of a GB measured at 77 K. The deviation from the Meissner-like magnetization at  $\sim 0.01$  T (arrow) signals the entry of flux into the grain and the inset shows that the grains are reversible above about 2 T (arrow).

the vortex density in the grains is near or at its maximum, so the ZFC curve cannot cross it, but instead merges with it as the irreversibility of each individual grain disappears. For FC, the decrease of the GB  $I_{cb}$  with  $H$  could result from a dilution of the pinning potential as the vortices move closer together, somewhat analogous to the reduction of the shear modulus in an Abrikosov vortex lattice at high fields.

One may think of two curves of  $I_{cb}(H)$ : one is the actual FC data in which the field in the grains is at a maximum and approximately equal to  $H$ ; the other is a hypothetical curve for no vortices in the grains. The latter  $I_{cb}(H)$  curve is determined only by inhomogeneities [2] of the Josephson current along a GB and it may be roughly parallel to the first, but exhibits lower  $I_{cb}$  than the FC data due to the absence of pinning by Abrikosov vortices in the banks. The beginning of this latter curve is seen as the ZFC curve below  $\sim 0.01$  T. The ZFC data from  $\sim 0.01$  T to  $\sim 0.5$  T, including the peak, is then the transition between the two  $I_c(H)$  curves as Abrikosov vortices populate the grains and provide pinning for the GB vortices.

An alternative explanation which shares some of the characteristics noted above is flux focusing along the GB, caused by field expulsion from the banks. While flux focusing can explain the low-field ( $\sim 0.01$  T) hysteresis in granular materials [10], the hysteresis in our data extends to much higher fields. Interpreting our data as due to flux focusing requires mapping our ZFC data points onto the FC curve (at a presumed higher GB field, that is amplified by flux focusing). This implies, e.g., at  $I_{cb} \sim 0.1$  A in Fig. 5, that a focused field of  $\sim 0.2$  T is found at the GB for applied fields of only  $\sim 0.004$  T. Our direct bulk magnetization data in Fig. 6, and also measurements with Hall-effect microprobes, dispel that possibility. In addition, the peak in  $I_c$  seen in our GBs for ZFC was not seen in the earlier study [10], and flux focusing offers no obvious explanation of it. Thus we suggest that flux focusing cannot explain our data.

We have discovered another method, besides FC, to introduce Abrikosov vortices into the grains and enhance  $I_{cb}$ . Starting from the ZFC case, if the current exceeds the threshold for flux creep, i.e., at  $I_{cg}$ , Abrikosov vortices are injected into the bulk grains [10]. These can play that same pinning role as the Abrikosov vortices introduced by FC, and thus increase  $I_{cb}$  of the GB. The increase, shown as the solid line in Fig. 5, is irreversible (i.e., permanent) and can be a considerable fraction ( $\sim 1/2$ ) of the increase found by FC. However, by analyzing the temporal voltages during current pulses that exceeded  $I_{cg}$ , evidence was found for heating effects. Thus the injection of vortices into the grains could be akin to FC. Shorter pulses ( $\sim 2$   $\mu$ sec) eliminated heating, so the much smaller, but definitive, enhancements of  $I_{cb}$  must be due to Lorentz-force driven vortex injection. The enhancements depended mostly on the pulse current magnitude (up to 1.4 A) and only weakly on the number of pulses.

In summary, we have presented strong support for the conceptual model of Gurevich and Cooley [3] in which GB vortices are pinned by Abrikosov vortices in the banks of the GB. This conclusion has some interesting and possibly important consequences. It provides a mechanistic basis to understand the high-field behavior of granular high- $T_c$  superconductors. It also points to the potential for improved performance (i.e., higher  $I_c$ ), in applications where  $I_c$  is

affected by GBs, by decorating the GB banks with pinned Abrikosov vortices.

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