# TRANSPORT PROPERTIES ACROSS HIGH-ANGLE BICRYSTALS OF MELT-TEXTURED $YBa_2Cu_3O_{6+x}$

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*Abstract*--We have investigated the transport properties of eight individual bicrystals in melt textured $YBa_2Cu_3O_{6+x}$ in low and high fields at 4K and 77K. Bicrystals were cut from samples produced by the melt texture process with 10% and 20% added $Y_2BaCuO_5$. Misorientations between the crystals were determined by pole figure analysis, backscatter channeling patterns and light microscopy. The bicrystals exhibited a wide range of misorientations in both the ab planes and c axis direction and all boundaries were high angle. Contrary to many reported expectations for melt textured materials, all of the grain boundaries were weakly coupled. However, the strength of the coupling varied with misorientation angle and a weaker field dependence of $J_c$ was observed in the samples with lower misorientation angle and lower grain boundary resistivity.

## I. INTRODUCTION

The exact role that high angle grain boundaries (HAGB) play in current transport in polycrystalline $YBa_2Cu_3O_{6+x}$ (123) is still not clear. One view is that grain boundaries with misorientation angles (θ) > 5-10° (i.e. HAGB) are inherently weak linked due to the intrinsic sensitivity of superconductivity in these materials to small scale disorder. Strong evidence for this view was presented in the thin film studies of Dimos et al.[1] Our own experiments on flux grown bicrystals support the overall nature of these findings, but we have also observed that some HAGB are strongly coupled [2,3]. Babcock et al.[2] and Eom et al.[4] established that certain, approximately 90° [100], grain boundaries are free from such weak link behavior, and have $J_c(B)$ dependencies akin to single crystals. Larbalestier et al.[3] showed that grain boundaries in θ[001] flux grown bicrystals can be weak-link free for θ values up to 22° θ[001]. They also reported that the $J_c(B)$ characteristics of some HAGB were sensitive to extended oxygenation anneals. This aspect has been studied extensively by Moeckly et al.[5]

The goal of this work was to investigate high angle grain boundaries in Melt Textured (MT) 123. One purpose of melt texturing [6-8] was to improve $J_c$ by creating large, well-oriented grains (which contain significant amounts of an insulating second phase, $Y_2BaCuO_5$ (211)) separated by low angle grain boundaries. Many authors [7-9] have reported significant transport current densities (>$10^4$A/cm$^2$ at 77K) within the individual MT grains. There has also been some suggestion that melt-textured HAGB are not necessarily detrimental to the passage of large critical current densities [10,11] and that deliberately added 211 may clean such grain boundaries, thus promoting better coupling [12]. Evidence, however, on this point [12-15] has been conflicting.

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In this work, we measured directly the properties of HAGBs to ascertain if they were weak-link free. In particular we were interested in the effects of the misorientation on the boundary properties. A number of studies of both MT material [10,11,16] and sintered bicrystals [17] have addressed aspects of this problem. Here we report on the first measurements of the high and low field $J_c$ characteristics across individual HAGB in MT bicrystals with known misorientation relationships. Our results bear directly on the nature of the coupling in such materials, permitting a direct comparison with our earlier measurements on flux grown bicrystals [2,3].

## II. EXPERIMENTAL PROCEDURES

The melt textured samples were fabricated from 123 and 211 powders. Samples of 123, containing 10 and 20mol% of 211 phase were melt textured using the method developed by Salama et al.[7] S. Jin provided MT material with 20% added 211 that was grown by a similar process [14]. Bicrystals were isolated and shaped by careful cutting, grinding, and polishing. Final samples were ~50 μm thick. Individual crystals were identified by polarized light microscopy. The relative contrast across the HAGB was used as a first estimate of the misorientation (Table 1). Both sides were polished and twin traces compared to verify that the samples were only one grain thick.

### A. *Electromagnetic Experiments*

Individual bicrystals were affixed to sapphire substrates with silver epoxy. Gold and/or silver leads of 25 μm or 50 μm diameter were attached to the sample surface with silver epoxy. The curing time and temperature was ~2 min. per lead at 200°C followed by an anneal at 400°C in flowing oxygen for 1 hr. Final contact resistivities were on the order of $10^{-8}\Omega cm^2$. Electromagnetic experiments performed on the samples include the following: resistive $T_c$ with high sensitivity AC measuring current, voltage vs. current traces in low fields (0-100G) at 4K and 77K and in high fields (0-7T) at 4K and 77K. Fields were applied orthogonal to the sample surface normal for the samples listed in Table 1. The field to c-axis orientation was thus different for each crystal of each sample. However, some measurements were made with the field parallel to the surface normal. Their $J_c(B)$ characteristics were qualitatively similar to those reported in this paper.

### B. *Misorientation Determination*

General crystallographic misorientation first was estimated by polarized light microscopy in order to preselect highly misoriented crystals. Quantitative misorientation information was obtained primarily by an x-ray pole figure technique. Crystallographic information for very small (surface area <200μmx-200μm) samples was deduced from backscattered electron channeling patterns obtained in a Scanning Electron Microscope (SEM).

The crystallographic misorientation is represented by the two angles shown in Fig. 1. $\gamma$ is the angle subtended by the c-axes of crystals 1 and 2. $\beta$ is the angle formed by the b(a) axis of crystal 1 and the projection of the b(a) axis of crystal 2 onto the (001) plane of crystal 1. Thus, $\gamma$ describes the "kink angle" formed by the high conductivity copper oxide planes at the boundary. $\beta$ then describes the remaining component of the misorientation relationship. This description is used rather than the standard (common axis, angle) pair description because it is believed to illustrate the important physical features of the misorientation relationship more directly.

## III. RESULTS

The bicrystals exhibited a variety of behavior which is illustrated by the data shown in Figs. 2-6. Fig. 2 and Table 1 show that all samples had linear, metallic normal state behavior with $T_c$(R=0)=89-91K, except for sample MB4B.1, which had a grain boundary $T_c$ of only 48K. A small breadth to the transition frequently was observed (see inset to Fig. 2) which we attribute to the boundary. The V-I traces were Josephson-like at low fields (Fig. 3), showing an abrupt transition behavior, rather than exhibiting the rounded behavior of flux flow observed in fully coupled boundaries [2,3]. The $J_c$(B) characteristics at 77K are shown in Figs. 4 and 5. The low field characteristics fall into two distinct classes. One class has relatively weak field dependence in fields up to 10mT. These boundaries have lower values of $\gamma$ and $\beta$. Other boundaries, having larger $\gamma$ and $\beta$ values, exhibit a strong field dependence and smaller boundary $J_c$. Both $J_c$(B) characteristics are much more field dependent than the fully coupled flux-grown bicrystals [2,3]. In Fig. 5, the high field 77K behavior of two bicrystals from the smaller misorientation group are compared. They exhibit similar exponential slopes in fields of 1-7T. At 4K (Fig. 6) the shapes of the high field curves of both types of samples are similar and all bicrystals appear to have a plateau-like characteristic between ~1 and 7T. In all cases the samples having lower misorientation tended to have the higher $J_c$, a weaker field dependence and a smaller grain boundary resistivity.

Table 1. Sample details (nm: not measured). All samples had 20% added 211 except MB4B.1 which had 10%. The last column gives the field sensitivity of $J_c$.

| Sample | Misor -ient | $\gamma$ | $\beta$ | $T_c$ (R=0) | $\rho_n d$ ($\Omega\mu m^2$) | $J_c$(B) 77K |
|---|---|---|---|---|---|---|
| MB4B.1 | high | 27° | 5° | 48K | 7.9 | - |
| SJ3151B3 | high | 18° | 41° | 91K | 1.3 | weak |
| MB4C.1 | high | 44° | 40° | 90K | 2.8 | strong |
| MB4C.3a | low | 7° | 6° | 89K | 0.3 | weak |
| MB4C.3b | low | 7° | 6° | 90K | 0.3 | weak |
| MB4C.4 | high | nm | nm | 89K | 8.0 | strong |
| MT4C.2#1 | low | nm | nm | 90K | 0.3 | weak |
| MT4C.2#2 | high | nm | nm | 90K | 8.7 | strong |

## IV. DISCUSSION

In the present experiments we selected 8 HAGB from MT samples and searched for strong-coupled behavior across the grain boundaries. We did not find such behavior. The V-I characteristics and the field sensitivity are both more characteristic of Josephson-coupled than strongly-coupled boundaries.

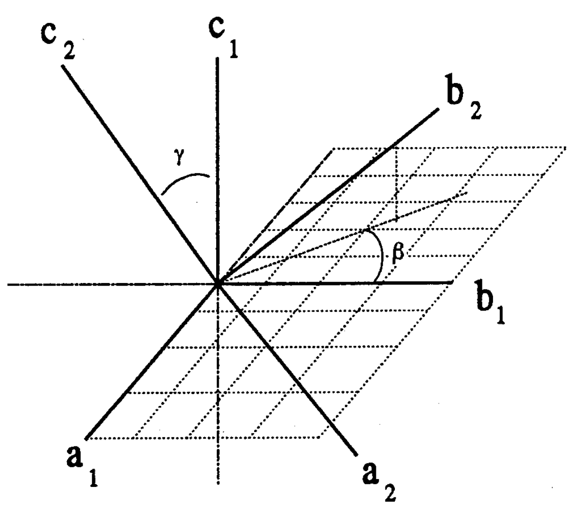


Fig.1 Definition of misorientation angles $\gamma$ and $\beta$

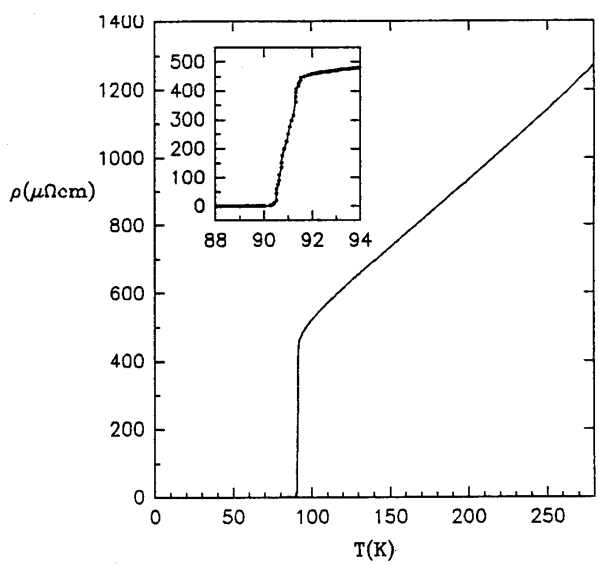


Fig. 2 AC Resistive $T_c$ trace for sample MB4C.3b.

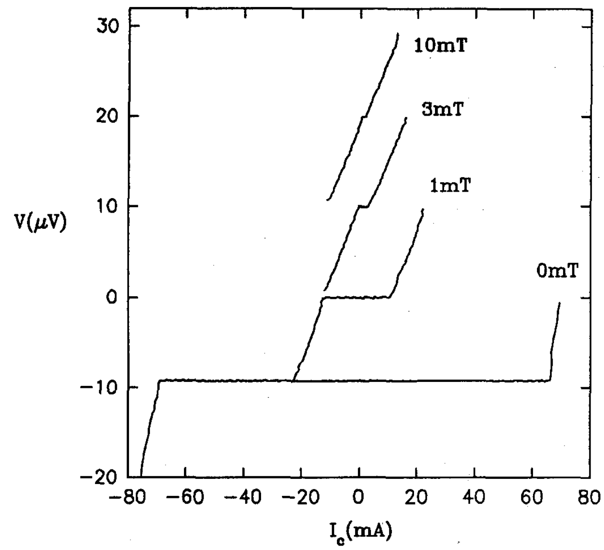


Fig. 3 V-I traces at 77K for sample MB4C.4.

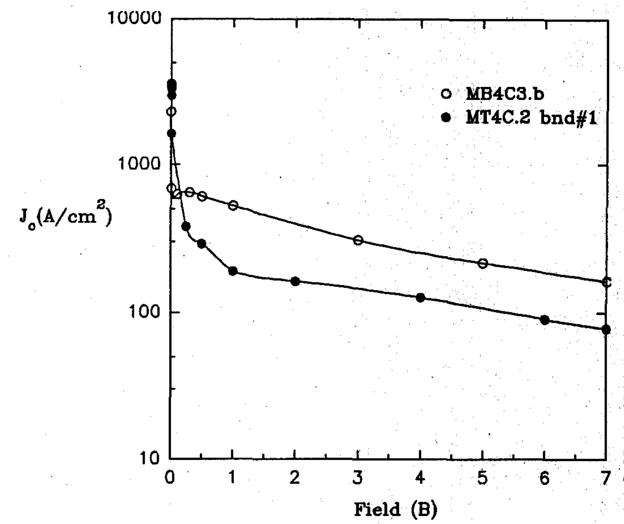


Fig. 5 High field behavior at 77K.

The values of grain boundary resistivity ($\rho_n d$) derived from measurements of the dynamic resistance (R) at 77K and the grain boundary area S ($\rho_n d$=RS, where d is the grain boundary width) are also characteristic of weakly coupled boundaries. They range from ~0.3 to 9$\Omega\mu m^2$, values quite comparable to those (~1 to 90$\Omega\mu m^2$) earlier reported by Larbalestier et. al. [3] for weakly-coupled flux grown bicrystals and values of ~0.02 to 8$\Omega\mu m^2$ reported by Dimos et. al. [1] on thin films.

The two boundaries found in an in-line tricrystal, MT4C.2, were tested. Fig. 4 shows that the higher misorientation boundary (MT4C.2 bnd#2) has almost the most sensitive $J_c(B)$ characteristic, while the lower misorientation (bnd#1) has almost the least sensitive. Thus the misorientation appears to dominate both the $\rho_n d$ value (8.7 and 0.3$\Omega\mu m^2$ respectively) and the $J_c(B)$ characteristics. This result was also observed in thin films [1,5].

Another point of interest is the appearance of a largely field-independent $J_c(B)$ in fields above about 1T at 4K (Fig. 6). A somewhat similar characteristic is observed at 77K (Fig. 5). In polycrystals this has sometimes been attributed to a percolative path [18], albeit of very small effective cross section. More recently, it has been suggested [19] that percolation need not be responsible for this behavior. These results support the latter suggestion in that a field independent $J_c(B)$ is seen even for bicrystals. Somewhat similar characteristics recently have been observed by Daumling et. al. [20] on a 25° [001] symmetrical tilt thin film bicrystal at 5K whose characteristics in zero field can be modeled as a resitively-shunted Josephson junction. Thus it appears that a finite high field, quasi-plateau character might be attributed to Josephson junction behavior, a possibility originally discussed by Yanson [21].

## V. CONCLUSIONS

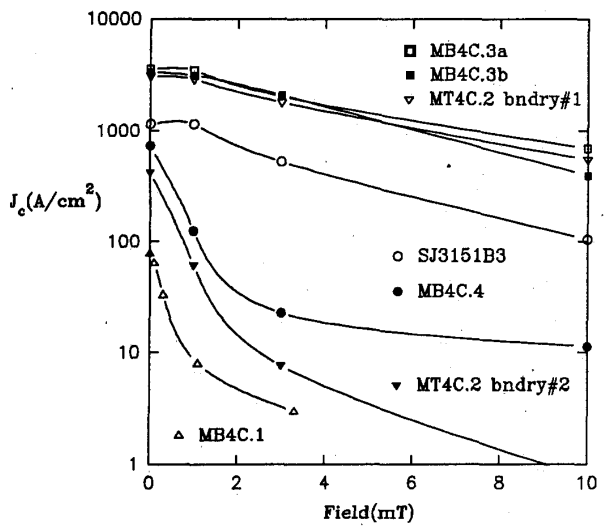


Fig. 4 Low field behavior at 77K.

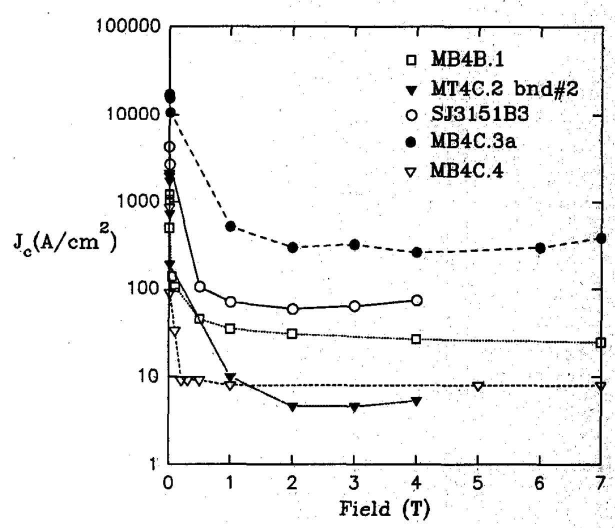


Fig. 6 High field behavior at 4K.

We have measured the $J_c(B)$ characteristics of 8 high angle grain boundaries in melt-textured $YBa_2Cu_3O_{6+x}$ bicrystals. All were found to exhibit weak coupling characteristics. The grain boundaries with lower misorientations had better electromagnetic coupling. All boundaries, regardless of misorientation, had a finite $J_c$ at high fields.

## ACKNOWLEDGEMENTS

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