# Critical currents across grain boundaries in YBCO : The role of grain boundary structure

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**Abstract**

Measurements across single grain boundaries in YBCO thin films and bulk bicrystals have been used to demonstrate the influence of grain boundary structure on the critical current carried across the grain boundary. In particular, we show that one role of grain boundary structure is to change the degree of pinning along the boundary, thereby influencing the critical current. This effect can be used to explain the large difference in critical current density across grain boundaries in thin films compared to that for bulk bicrystal. These differences illustrate the distinction between the intrinsic mechanism of coupling across the grain boundary that determines the maximum possible critical current across a boundary and the measured critical current which is limited by dissipation due to the motion of vortices.

*Keywords* : Critical current, grain boundary

## I. Introduction

The critical current across grain boundaries in $YBa_2Cu_3O_x$ (YBCO) has been measured in a large number of studies using a variety of types of samples. In each of these studies, it has been shown that the critical current carried across a grain boundary is a strong function of the misorientation angle between the grains that form the boundary. However the magnitude of the critical current across a grain boundary of a given misorientation can vary widely from study to study. This variation implies that there must be some differences between boundaries prepared in different ways. A variety of models have been proposed that attempt to explain the fundamental dependence of critical current on misorientation angle. [1,2-5] However, the critical current ($I_c$) and critical current density ($J_c$) of a grain boundary are macroscopic quantities that reflect not only the intrinsic coupling across the boundary (which would determine electron conduction) but also external parameters, such as flux motion along the boundary (which contributed to measured dissipation). Thus, both factors are important in determining the transport properties of a grain boundary.

In this work, we compare critical current density measurements across thin film and bulk grain boundaries. This comparison reveals that significant differences in the transport $J_c$ can be related to differences in structure of the boundary. Furthermore, those structural variations lead to differences in pinning of vortices along the boundary. Thus, the variation in pinning along the two types of boundary contributes to the large differences in transport properties.

## II. EXPERIMENTAL

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Thin film bicrystals for these studies were prepared by off-axis sputter deposition onto commercially available $SrTiO_3$ bicrystal substrates so that the misorientation angle and axis in the YBCO bicrystal is determined by the substrate. Bulk bicrystals of YBCO were prepared using a dual-seeded melt textured growth process that has been described fully elsewhere. [8] In short, two $NdBa_2Cu_3O_x$ seed crystals are used to nucleate the growth of two domains in a YBCO pellet when subjected to a melt-texturing process, resulting in a bicrystal with the misorientation angle and axis determined by the seeding method. The YBCO pellet contains YBCO plus about 25 vol. % $Y_2BaCuO_x$ (211), so that the YBCO grains in the bicrystal also contain small 211 particles. Transmission electron microscopy (TEM) has shown that the 211 is uniformly distributed throughout the sample and is not segregated to the boundary.

Transport measurements were carried out using a 4- or 6-terminal configuration at a temperature of 77 K. Magnetic fields up to 0.1 T were also applied. In most cases, the field was applied with $H$ parallel to the $c$-axis of the grains. The dependence of $J_c$ on field orientation was measured for one 90° [100] tilt boundary by rotating the field in the plane of the boundary.

## III. RESULTS

Many of the studies of transport properties across single grain boundaries, and all of those discussed in this work, have been carried out on what are described as [001] tilt boundaries. The misorientation angle, $\theta$, defines the rotation angle between the two grains. Unfortunately, this is an incomplete representation of the grain boundary, since the specific grain boundary plane must also be specified. In the case of symmetric [001] tilt boundaries, the boundary plane is implied. However, it has been shown that the boundaries in thin film bicrystals are not well defined but instead meander significantly, leading to a grain boundary plane that changes as a function of position along the boundary. In contrast, the grain boundaries in the bulk bicrystals have been shown to be highly planar over a wide range of length scales. An example of the typical

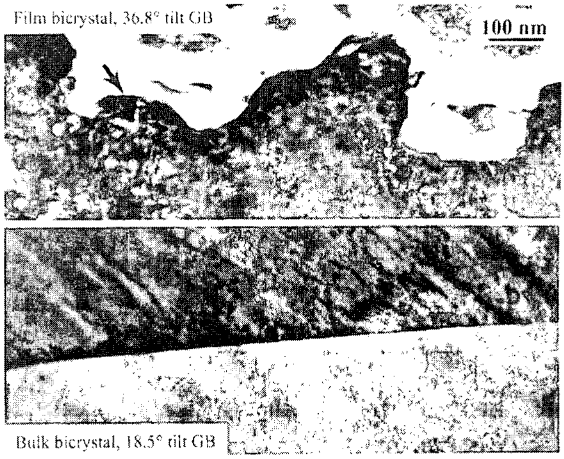


Fig. 1 Bright-field TEM micrographs showing the typical microstructure of a thin film [001] tilt grain boundary (top) and a bulk [001] tilt boundary.

microstructures for each type of boundary is shown in Fig. 1. Thus, when comparisons are made between different measurements of grain boundary properties, it cannot be assured that the grain boundaries are identical or, as in the case of comparing thin film with bulk bicrystals, even similar.

An illustration of this point is shown in the transport properties plotted in Fig. 2. In this figure, previously reported [6-8] results of $J_c$ measured across single grain boundaries in thin film bicrystals and in bulk bicrystals are plotted as a function of the misorientation angle between the grains. All of the data represents measurements on nominally identical boundaries, that is [001] tilt grain boundaries. If these boundaries were indeed identical, one would expect the same value of $J_c$ for a given value of $\theta$. However, there are two very significant differences in $J_c$. Firstly, the absolute magnitude of $J_c$ measured for grain boundaries in the bulk samples is significantly lower than those measured for the thin film counterparts. Secondly, the functional dependence of $J_c$ on $\theta$ is weaker in the case of the bulk samples compared to the thin films, with the slope of $J_c(\theta)$ for the bulk samples approximately half of that for the thin films. In the very low angle regime ($\theta$ less than $\approx$10°), the difference in $J_c$ may be ascribed to the significantly lower $J_c$ in the grains on each side of the boundary in the bulk samples compared to relatively high $J_c$ values typical of high quality thin films. Since each

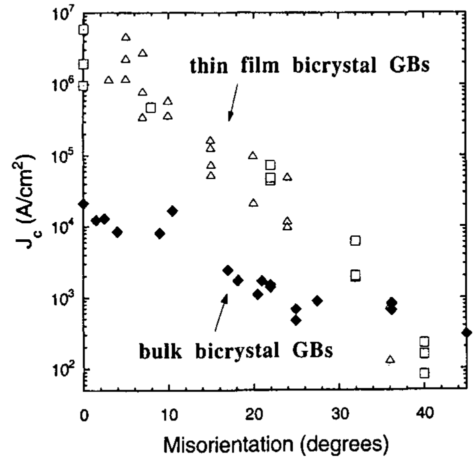


Fig. 1. Critical current density ($J_c$) at 77 K, $B$ = 0, as a function of misorientation angle, $\theta$, for single grain boundaries in thin film and bulk bicrystals. The data from Ivanov [6], Heinig [7] are for thin film grain boundaries and the data from, Todt [8] is for bulk bicrystal boundaries. All are for nominal [001] tilt boundaries.

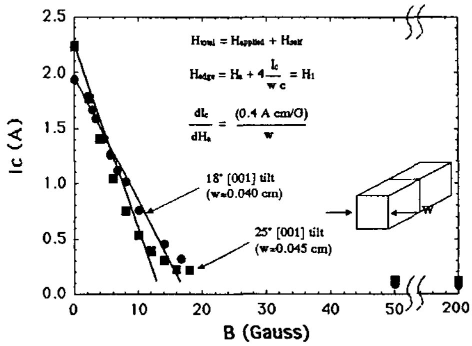


Fig. 2. Critical current ($I_c$) at 77 K as a function of applied magnetic field. The applied field was parallel to the $c$-axis of each grain.

transport measurement of a grain boundary includes some contribution from the grains on each side due to the placement of voltage contacts, any dissipation in the grains contributes to the measured voltage. As a result, the values measured for the grain boundary $J_c$ in the bulk samples are influenced by the $J_c$ for the grains (to a degree influenced by the spacing of the voltage taps for an electric field criterion) and should in fact be somewhat lower if the boundary also contributes to the dissipation as well. However, in the higher angle regime, the $J_c$ values clearly reflect dissipation due to the grain boundary in both the thin film and bulk bicrystal cases. For example, looking at the values for $\theta \approx 20°$, it is seen that in both cases the $J_c$ values measured are nearly an order of magnitude lower than the low $\theta$ values, suggesting the strong influence of the boundary. In this situation, it is difficult to explain the difference in critical current between the two types of boundaries that are nominally identical. One can conclude from this that, although the boundaries are nominally identical, there must be significant differences between them and, as shown in Fig. 1. The most obvious difference is in the morphology of the grain boundary.

Recently, Gray, et al.[9] have proposed a model that considers the pinning of vortices along the grain boundary due to differences in the structure of the boundary to explain the discrepancies in this data. In this model, it is assumed that the highly meandering grain configuration of thin film grain boundaries is effective in pinning a Josephson vortex while the smooth, planar configuration of the bulk boundaries would pose little resistance to motion. The presence of Josephson vortices is assumed as a consequence of the weak coupling across the boundaries, and the Josephson critical current, $J_{cJ}$, follows an exponential decay with the misorientation angle, $\theta$. Under these circumstances, the $J_c$ measured for the thin film boundaries should reflect the degree of pinning and scale linearly with $J_{cJ}$. In the case of the bulk bicrystals, $J_c$ occurs once a vortex enters the boundary since it may then be driven along the boundary rather freely, leading to dissipation. In this situation, $J_c$ is expected to scale as $J_{cJ}^{1/2}$. Based on this model, the difference in the magnitude of $J_c$ as well as the difference in the slope of $J_c(\theta)$ in Fig. 2 can be accurately predicted.

Microstructural characterization of the bulk boundaries reveals no features that would be effective in pinning vortices. Under these circumstances, $J_c$ is determined once the magnetic field reaches $H_{c1}$ and vortices can penetrate the boundary. A consequence of this factor is that $I_c$ should scale inversely with applied magnetic field

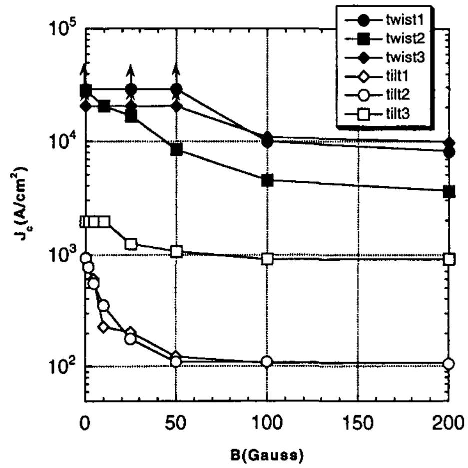


Fig. 3. $J_c(B, 77\text{ K})$ for 90° [100] twist and symmetric tilt samples. The twist samples are in black symbols, the symmetric tilt are the white symbols. The arrows on the symbols for samples twist1 and twist3 represent minimum $J_c$ values, limited by sample heating

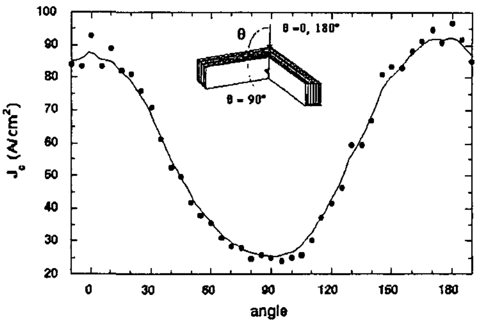


Fig. 4. Critical current density ($J_c$) at 77 K as a function of applied magnetic field orientation, $\theta$. The applied field of 0.1 T was oriented in the plane of the boundary as shown in the inset.

since the total field is equal to the applied field plus the self-field generated as a result of the current passing through the sample. Figure 3 shows data of $I_c$ as a function of small applied fields for two different bulk grain boundaries. In each case, the initial decrease obeys the expected relationship. Note that $I_c(0)$ for the 25° boundary is higher than that for the 18° boundary and that they cross over at approximately 5 Gauss. This behavior is based on the self-field generated by the sample, which is dependent on the sample dimensions. As a further test of consistency, we can use the slop of $I_c(B)$ to calculated the dimensions of the sample measured and, as noted in the figure, the agreement between this calculated value and the actual dimensions of the sample is within about 25%, supporting the concept that $J_c$ is determined by flux penetration along the boundary for this orientation.

In contrast, the highly meandering configuration of thin film boundaries exhibits defects (facets) along the boundary of a spacing that is closely correlated to the characteristic length predicted for Josephson vortices along the boundary. In this case pinning would be expected to be strong. For example, the grain boundary plane changes as a function of position, and recently we have been able to measure the effect of the boundary plane on $J_c$ using 90° [100] grain boundaries [10]. In these measurements, we have found a substantial difference in $J_c$ based on the boundary plane, with 90° [100] twist boundaries supporting a $J_c$ nearly an order of magnitude higher than their 90° [100] symmetric tilt counterpart, as shown in Fig. 4. Thus, the idea that pinning of Josephson vortices in the thin films boundaries can be strong is reasonable based on a boundary consisting of a series of different planes. The higher slope as f($\theta$) for the thin film data of Fig. 2, by about a factor of two over that for the bulk samples, is consistent with that predicted from this model.

A final demonstration of the influence of vortex pinning along the boundary is shown in Fig. 4. In this case, the transport measurements are across a 90° [100] symmetric tilt boundary. These boundaries are produced in similar manner and exhibit the same highly planar boundary structure as the [100] tilt boundaries discussed to this point. As shown in the inset of Fig. 4, the applied field (0.1 T) was rotated in the plane of the boundary. The data of Fig. 4 shows a smooth variation of $I_c$ with field orientation. The maximum corresponds to a field oriented parallel to the *ab*-planes of the grains ($\theta \approx 0°$, 180°) while the minimum occurs when the field is oriented so that it is symmetrically inclined from the *c*-axes of the grains by 45° ($\theta \approx 90°$). The absence of any sharp peaks in the curve supports the idea that these planar boundaries are uniform and do not contain discrete pinning centers. However, the general shape of the curve is similar to that measured for single crystals or thin films in which a maximum in $J_c$ is measured when the field is oriented along the *ab*-planes and a minimum in $J_c$ is measured when the field is parallel to the *c*-axis. Those results are interpreted on the basis on strong pinning of Abrikosov vortices due to the layered structure of YBCO when the field is parallel to *ab* and relatively weaker pinning when the field is parallel to the *c*-axis. Thus, these results suggest that even though only coreless Josephson vortices are expected along the boundary, pinning can play a role in determining $J_c$, even for these straight grain boundaries.

## IV. DISCUSSION

The data shown here can be consistently analyzed on the basis of vortex pinning along the grain boundary. In very low angle grain boundaries, where the vortices may exist as Abrikosov vortices, pinning based on some microstructural features has been shown previously [11]. In this discussion, we have concentrated on higher misorientation angles where it is Josephson vortices that must be pinned. An important aspect of pinning is that the length scale of the pinning center should be of the same order as the characteristic length scale of the vortex. For Josephson vortices, the characteristic length is the Josephson penetration length, $\lambda_J$, which is inversely proportional to the strength of coupling for the junction, and therefore is expected to vary as a function of $\theta$. However, $\lambda_J$ is expected to be rather long for the higher angle boundaries considered here, on the order of a few hundred nanometers to microns. This length scale is consistent with the characteristic period of meanders in thin film boundaries, which would lead to relatively strong pinning in that case. In the bulk grain boundaries, however, there are no features found along the boundary on this length scale and consequently pinning is expected to be weak.

The data of Fig. 4 suggests that the motion of Josephson vortices can be impeded in certain directions, however, The maxima in $J_c$ correspond to field orientations for which the vortices must move across the *ab*-planes of the sample. Certain pinning mechanisms cannot be completely excluded as the basis for this behavior due to the geometry of the sample. For example, in this pure symmetric tilt boundary, the dislocations are expected to run parallel to the *ab*-planes, and thus they may contribute to pinning. In addition, the Abrikosov vortices in the grains themselves are also more strongly pinning, at least in one direction, and so pinning due to a rigid lattice [12] is also a possibility. However, the dependence of $J_c$ on angle can also be explained based on the drag force induced by the *intrinsic anisotropy* of the YBCO grain. The circulation currents that define the Josephson vortex pass through the YBCO grains that consist of superconducting and insulting layers. In an idealized system, the currents will be passed as supercurrent and Josephson current in each of the layers, respectively. In this highly overdamped system, viscous drag is expected to impose a resistance to motion in the direction of the layering [13]. Note that motion parallel to the layer is not subject to this friction and thus is expected to be free of pinning, as in the case of the [001] tilt boundaries discussed previously.

In this work, we consider the presence of vortices and their as key aspect of dissipation. The intrinsic condition across a short length of the boundary is also an essential factor in determining $J_c$. A number of models based on the structure of the boundary or pairing symmetry have been proposed that may reasonably explain this dependence. [2-5] However, none of these models can accurately describe the large variations in transport data between measurements on different sets of thin films or the more systematic differences between thin film and bulk bicrystal data if those boundaries are indeed identical. In order to understand those large differences, we must invoke some difference in the coupling across the different types of boundaries. In this work, we have shown that indeed there are physical differences. In addition, we suggest that these physical differences may even lead to different degree of coupling. However, it is variation in coupling from point to point that can lead to a high degree of pinning. Thus, we suggest that there may be some fundamental relationship between the Josephson critical current, $J_{cJ}$, and $\theta$ but it is vortex motion that dominates the relationship between the *measured* critical current, $J_c$, and $\theta$.

## V. CONCLUSION

The significant differences in the critical current density carried across higher angle [001] tilt grain boundaries in thin films compared to bulk samples can be explained on the basis of pinning of Josephson vortices. A model based on pinning can quantitatively predict the difference in magnitude of $J_c$ for equivalent boundaries as well as the difference in the slope of $J_c(\theta)$ and the dependence of $I_c$ on applied field. A strong basis for pinning is the meandering grain boundary plane typical of thin film bicrystals. In contrast, the straight, highly planer boundaries typical of bulk bicrystals provide very weak pinning. However, it is found that even along these planar boundaries the intrinsic anisotropy of YBCO can lead to pinning of Josephson vortices.

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