Structure and Transport Properties of [001] Tilt Grain Boundaries in YBa$_2$Cu$_3$O$_x$*

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STRUCTURE AND TRANSPORT PROPERTIES OF [001] TILT GRAIN BOUNDARIES IN YBa$_2$Cu$_3$O$_x$

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ABSTRACT

We have studied the microstructure and transport properties of nominal [001] tilt grain boundaries in order to correlate microstructural features with transport properties. Single grain boundaries were prepared by epitaxial growth of YBa$_2$Cu$_3$O$_x$ onto bi-crystal SrTiO$_3$ substrates. Transport measurements were used to characterize the electrical behavior across the boundary while TEM was used to characterize the structure of the boundary. The transport measurements are consistent with grain boundary behavior, but detailed comparisons show anomalies which may be resolved by microstructural characterization. Microstructural characterization showed that these artificially induced grain boundaries are periodically decorated by impurity phases. In addition, the boundaries are not always straight, but instead frequently meander away from the path of the underlying boundary in the substrate, and the various facets produced by the meandering show varying degrees of tilt and twist character. These structural variations suggest one potential explanation for the reduced effective coupling area, although the correlation is not unambiguous.

INTRODUCTION

The weak link behavior of high angle grain boundaries has been documented for many high-temperature superconductors.[1-5] Mechanisms based on structural models proposed to explain this behavior suggest that the specific grain boundary plane must play an important role. For example, dislocations along a boundary have been cited as a plausible explanation for the rapid decrease in $I_c$ as a function of misorientation.[6] Meilikho$^v$[7] further suggested that asymmetric facets may be more strongly coupled than symmetric facets based on the difference in strain fields around edge and screw dislocations.

An implicit assumption in studies of well-defined, artificially induced grain boundaries is that the grain boundary in the superconducting film is defined by the bi-crystal interface (grain boundary) in the underlying substrate. Previously, it has been shown by Alarco, et al.[8] that in special cases, the boundary in the film can deviate from the boundary in the substrate, and this has been further demonstrated in a more general case by Traeholt, et al. [9]. Furthermore, the transport measurements indicate that the effective coupling area is much smaller than the total grain boundary area. These results illustrate the difficulty in making an insightful correlation between grain boundary structure and properties.

EXPERIMENT AND RESULTS

The YBCO films were deposited by in situ growth onto 24° SrTiO$_3$ bi-crystal substrates, using sputtering at a growth rate of ~4 Å/min. Photolithography was used to define microbridges for transport measurements. Samples were examined by TEM in the plane-view orientation.

Figure 1 shows the sample resistance as a function of temperature for two intergrain sections and one intragrain section, each 110μm wide. The resistances of
the intergrain sections differ above $T_c$ due to the different lengths of the sections, but the "normal-state" resistances of the grain boundaries themselves agree extremely well. This indicates the uniformity of the grain boundary for transport measurements over $110 \mu m$ wide regions.

Figure 2 shows extended range V-I curves for a $36\mu m$ wide intergrain section measured as a function of temperature from 88 K to 81 K. At high temperatures, the data show linear (Ohmic) behavior over the full current range. At lower temperatures, there is linear behavior only in the low- and high- current limits, with a crossover between these. The data are very well fit by the Ambegaokar-Halperin (A-H) model for Josephson coupled superconductors\[10\], and this provides a model for quantitative description of the grain boundary behavior. These fits contain three parameters: $R_N$ is the grain boundary normal state resistance, $\gamma$ is the ratio of the Josephson coupling energy $E_J$ to the thermal energy $k_B T$, and $I_{A-H}$ is a critical current.

Using this information, we find that the values $I_{A-H}$ obtained directly from the fits are a factor of ~16 larger than the current $eE_J/h$ which was calculated from the fit values of $\gamma$. Considering the geometry of the junction, there are several possibilities which might explain this behavior. The section may behave as a long junction with a distribution of pinning centers in the barrier. Alternatively, defects along the grain boundary may divide the section into a number of independent junctions, as in proposed models of grain boundaries.[11-13]

![Figure 1](image1.png)  
![Figure 2](image2.png)

Figure 1 Resistance versus temperature for one intragrain and two intergrain sections, each $110 \mu m$ wide. Note the agreement in the "normal state" resistance for the two grain boundary sections.

Figure 2 Extended V-I curves for a $36\mu m$ wide intergrain section for temperatures between 88 K and 81 K.

The typical structure of the boundary is shown in the TEM image in Fig. 3. Typically, second phases decorate the grain boundary, and these can also be observed by optical microscopy. TEM imaging also shows that the boundary in the YBCO film is not straight, but "meanders," as the two orientations of the YBCO film grow over the substrate boundary in places. Both parts of the film grow into each other and that the degree of the meandering is about equal on both sides of the underlying substrate boundary, in this film reaching $250 \ nm$ in places. In the case of the films discussed here, the rotation angle was $24^\circ$. Thus, any part of the film which overgrows the substrate boundary finds itself in an orientation which has very poor matching with the substrate and little tendency for epitaxial growth. Thus, the observation of meandering in these films suggests that this meandering is a general phenomenon.

It is likely that island-like growth of the YBCO films is responsible for the "meandering" of the boundary. In this situation, changes in the growth rate are likely to result in changes in the magnitude of the meandering. The much smaller magnitude of the meandering observed in a film grown under a much lower deposition rate, together with a correlation between island-size and the magnitude of the meandering confirms this mechanism.\[14\]
Figure 3 Bright-field TEM micrograph showing the typical "meandering" nature of grain boundaries in YBCO thin films deposited onto SrTiO₃ bi-crystal substrates.

An important implication of this meandering is that the grain boundary plane varies along the length of the boundary, as shown in Fig. 4 in which various segments of the boundary possess a different macroscopic grain boundary plane. Furthermore, the various "segments" exhibit varying degrees of tilt and twist character. As noted above, structural models suggest that the specific grain boundary plane must play a role in transport behavior. Thus, the observation that the grain boundary plane varies suggests the potential for significant local differences in transport properties along the boundary.

Figure 4 High-resolution TEM micrograph which illustrates how the grain boundary plane, and, in particular, the relative tilt and twist components, vary as a result of the "meandering."

CONCLUSIONS

In summary, structural variations in artificially induced grain boundaries have been observed. Specifically, the boundaries have been observed to "meander" and, as a consequence, the grain boundary plane varies along the length of the boundary. In addition, transport measurements have shown consistent grain boundary behavior, but a detailed comparison with short junction behavior has revealed some anomalies in Josephson coupling behavior.

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