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Correlation of Transport Properties with Grain Boundary Atomic Structure in High- T_c Superconducting Films and Tapes

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CORRELATION OF TRANSPORT PROPERTIES WITH GRAIN BOUNDARY ATOMIC STRUCTURE IN HIGH T_c SUPERCONDUCTING FILMS AND TAPES.

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ABSTRACT

The critical current density of high- T_c superconducting thin films is found to be sensitive to the presence of flux-pinning defects, and particularly to the existence of grain boundaries which may act either as pinning centers for the vortices or as "weak link" junctions. Atomic resolution Z-contrast imaging and spatially resolved electron energy loss spectroscopy provide a method to obtain a direct image of the atomic grain boundary structure for correlation with its electronic structure. Using a combination of transport measurements and electron microscopy, we have begun to correlate superconducting properties with atomic scale grain boundary structure using YBCO thin films deposited on SrTiO₃ bicrystals and on Ni substrates (RABiTS).

KEY WORDS

YBCO, grain boundary, RABiTS, Z-contrast imaging, electron energy loss spectroscopy

INTRODUCTION

Since the discovery of high- T_c superconductors, there has been an intense research effort to incorporate these materials into viable devices. YBa₂Cu₃O_{7- δ} (YBCO) films grown on Rolling Assisted Biaxial Textured Substrates (RABiTS) has recently been developed [1, 2] with critical current density (J_c) over $1.2 \cdot 10^6$ A/cm². The texture of the substrate induces the presence of strain fields and grain boundaries in the YBCO film (fig. 1). The J_c of high- T_c superconducting thin films is sensitive to the presence of flux-pinning defects and to the existence of grain boundaries. The exact role of the atomic structure at grain boundaries in controlling the transport properties is a subject of debate. The J_c measured across a grain boundary generally decreases as the misorientation angle increases [3, 4]. Low-angle grain boundaries are composed of arrays of dislocations whose associated strain fields cause the material to become locally non-superconducting [5]. For high-angle grain boundaries (angles above $\sim 10^\circ$), the spacing between dislocations becomes comparable to the lattice parameter and this continuum description breaks down [6]. It has therefore generally been assumed that such boundaries will act as Josephson junctions [3, 4]. However, there have been reports that the boundaries are actually composed, according to their angle, of a defined ratio of superconducting and weak link parts and might, for certain symmetries, be free of weak links [7, 8, 9.]. Moreover, recent models for YBa₂Cu₃O_{7- δ} have shown that it is the atomic structure of the grain boundary plane that dominates J_c [10].

Electron microscopy, in particular atomic resolution Z-contrast imaging and spatially resolved electron energy loss spectroscopy (EELS) in the dedicated scanning transmission electron

fig. 1. TEM cross-sections of YBCO/YSZ/CeO₂/Ni films (RABiTS). (a) The texture of the nickel substrate induces grain boundaries in the YBCO film. (b) A significant density of extended strain fields and planar defects are observed in the YBCO layer.

microscope (STEM), provides a method to obtain a direct image of the atomic grain boundary structure for correlation with its electronic structure. Therefore, we have begun to characterize, using a combination of transport measurements and electron microscopy, the relation between superconducting properties and grain boundary structure in YBCO thin films.

EXPERIMENTAL

Using Pulsed Laser Deposition (PLD), 500 nm thick YBCO films have been deposited on SrTiO₃ bicrystals with misorientations of 24°. Transport properties across the boundary are measured through a one centimeter wide boundary to highlight small effects. Silver contacts have been deposited by sputtering and annealed at 500°C for 30 minutes in an oxygen atmosphere. Transport measurements in magnetic fields have been performed using the 4 probe method. The system allows the rotation of the sample in the magnetic field and therefore a study of the angular dependence of transport properties.

The high resolution microscopic study has been performed on a dedicated STEM (VG Microscopes HB501 UX) which allows one to perform simultaneously high resolution Z-contrast imaging and EELS (energy resolution of 0.5 eV). The cold field emission tip allows a 0.22 nm probe size to be obtained leading to an atomic column resolution both in the image and the analysis. The Z-contrast image is generated point by point using an annular detector to collect the high angle scattering which shows an intensity proportional to the Z^2 dependence of the Rutherford scattering cross section. The low angle scattering can be used for simultaneous EELS, which can be used to map hole concentrations on the scale of the coherence length [9]

RESULTS AND DISCUSSION

In figure 2, the $V(I)$ curves plotted for several fields exhibit a linear differential ohmic behavior, which extrapolated to $I = 0$ A gives a negative intercept. To explain this behavior, we have modeled the grain boundary as composed of equally sized and spaced weak links (dislocation

cores), separated by a very small fraction (x) of strong links (fig.3). The weak links have a tunneling behavior and carry J_w . The strong links might be either superconducting regions carrying a depairing current, strong links carrying the same J_c as the grains, or just Josephson junction that are much strongly coupled than the rest of the boundary, carrying $J_s = J_J$. The observed field dependence of J_c seems to exclude the case of the depairing current. In the 2 other cases, the curves exhibit behavior qualitatively similar to that of data.

The model proposed is consistent with the images recorded for such boundaries. The Z-contrast image of a near symmetric high angle boundary ($\approx 30^\circ$) presented in figure 4 shows facets composed of groups of dislocation cores, symmetric or asymmetric, separated by slightly distorted cells. As the groups of asymmetric dislocation cores are certainly acting as bad weak links, the symmetric ones and the better ordered cells might carry a better critical current. Spatial resolution EELS across the different segments of the boundary should allow a quantitative assessment of these possible scenarios.

fig. 2. $V(I)$ curves for magnetic fields varying from 117 Gauss to 3 Tesla. The small $J_c(B)$ arises from the strong links, the linear differential resistivity regime represents the grain boundary resistance, and the rapid rise at high current densities is due to the properties of the grains.

fig. 3. The grain boundary is composed of equally sized and spaced dislocation cores separated by a very small fraction of strong links (a). The simulated graph (b) exhibits qualitatively similar behavior to the data in fig. 2. for a situation where the fraction $x=0.005$, and J_c is the observed $J_c(B)$ of the grains.

fig. 4. Z-contrast image of a 30° grain boundary showing an asymmetric segment on the left, containing a slightly distorted unit cell, and a small symmetric segment on the right.

CONCLUSION

YBCO / RABiTS contain stacking faults and grain boundaries which influence their pinning and transport properties. It is necessary to investigate single $\langle 001 \rangle$ tilt grain boundaries to understand their possible effect on the overall properties of textured thin films. $V(I)$ curves measured across a 24° boundary exhibit a linear differential ohmic behavior above I_c along with a weak magnetic field dependence which can be modeled by an array of dislocation cores acting as weak links separated by a very small fraction of strong links. This model is consistent with the Z-contrast high resolution images previously recorded and spatially resolved EELS should help to determine the nature of the links.

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