49192-20-105
85088-24-105
58916-00-105

MANUSCRIPT

ANL/CP--77430

DE93 000610

**CURRENT TRANSPORT ACROSS GRAIN BOUNDARIES IN $\text{TLBa}_2\text{Ca}_2\text{Cu}_3\text{O}_x$
THICK POLYCRYSTALLINE FILMS AND $\text{YBa}_2\text{Cu}_3\text{O}_7$ STEP EDGES***

J.D. Hettinger, D.H. Kim, D.J. Miller, J.G. Hu, K.E. Gray, J.E. Sharping

*Materials Science Division and Science & Technology Center for Superconductivity
Argonne National Laboratory, Argonne, Illinois 60439*

K. Daly, C. Pettiette-Hall

TRW Space and Technology Group, One Space Park, Redondo Beach, Ca 90278

J.E. Tkaczyk and J. Deluca

General Electric Research and Development, Schenectady, NY 12301

The submitted manuscript has been authored by a contractor of the U.S. Government under contract No. W-31-109-ENG-38. Accordingly, the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or allow others to do so, for U.S. Government purposes.

1992 Applied Superconductivity Conference, Chicago, IL, August 24-28, 1992

jmc

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

*Work supported by the U.S. Department of Energy, Division of Basic Energy Sciences-Materials Sciences and Conservation and Renewable Energy as a part of a program to develop electric power technology under contract #W-31-109-ENG-38 (JDH, DJM, JGH, KEG, JES) and the National Science Foundation Office of Science and Technology Center for Superconductivity contract #DMR-8809854 (DHK).

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

for

CURRENT TRANSPORT ACROSS GRAIN BOUNDARIES IN $\text{TLBa}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ THICK POLYCRYSTALLINE FILMS AND $\text{YBa}_2\text{Cu}_3\text{O}_7$ STEP EDGES

J. D. Hettinger, D. H. Kim, D. J. Miller, J. G. Hu, K. E. Gray, and J. E. Sharping

Argonne National Laboratory, Argonne, IL 60439, U.S.A.,

K. Daly and C. Pettiette-Hall, TRW Space and Technology Group, One Space Park, Redondo Beach, CA 90278, U.S.A.,

J. E. Tkaczyk and J. Deluca, General Electric Research & Development, Schenectady, NY 12301, U.S.A.

Abstract--We have performed transport measurements in magnetic fields (H/c) up to 10T on a series of $\text{YBa}_2\text{Cu}_3\text{O}_7$ grain boundaries induced by epitaxial growth on a substrate containing a series of step edges. For this material we find an activation energy which goes as $U(T) \sim (1-t)$. Electric field versus current density curves taken at a constant temperature while varying the magnetic field allows a determination of the magnetic field dependence of the activation energy using the Ambegaokar-Halperin model. These values agree with results extracted from the resistive transitions. Similar characterizations of a sample of $\text{TiBa}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ are also presented. The temperature dependence of the activation energy for this system may be summarized as $U(T) \sim (1-t)^2$, suggesting that the grain boundaries behave as SNS junctions.

I. INTRODUCTION

Large scale applications of high-temperature superconductors (HTS) for current transport and magnetic applications necessitate the use of polycrystalline materials. Processes suitable for large scale production which could possibly render the resulting bulk material textured have been investigated [1,2]. These studies have yielded optimistic results regarding the usefulness of textured materials at low temperatures for applications, particularly for HTS containing bismuth. For this reason, it is very important to understand the types of grain boundaries that play a detrimental role in the dissipationless conduction process. Recently there has been a large body of research directed at investigating these properties. The first work was performed on $\text{YBa}_2\text{Cu}_3\text{O}_7$ (Y-123) films grown on bi-crystal substrates by Dimos and coworkers [3]. These results indicated that all high-angle-grain-boundaries (HAGB) behave the same, as weak-links with Josephson-junction-like properties. This has been exploited for electronic device fabrication [4]. In fact, much of the work which has followed these initial investigations has been geared toward device fabrication. There has not yet been a high field, systematic study of a well defined grain boundary or system of grain boundaries.

Work by Babcock and coworkers on flux grown bi-crystals of Y-123 indicated that not all HAGB were detrimental to the dissipationless conduction process [5]. However, the critical current densities, as defined by a electric-field criterion, in the

materials forming the electrodes on either side of the junctions were not particularly impressive. Therefore, questions still remain regarding the existence of strongly coupled HAGB.

Although these efforts represent significant and important progress, they are rather unrealistic models of true polycrystalline materials where many grain boundaries, both high and low angle, in parallel and series, are sampled simultaneously. However, in the other extreme of polycrystalline materials, studies suffer from a lack of knowledge about the grain boundary structure [6,7]. Therefore, it is difficult to determine if all or a few grain boundaries of a certain type are contributing to the observed dissipation in a transport measurement.

The work presented here is an attempt to bridge the gap between single grain boundary studies and studies of polycrystalline materials. With that as a goal, results from two systems are discussed. First, a textured thick film of $\text{TiBa}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ (Ti-1223), a single thallium layer compound which has been shown to be the best alternative so far to Y-123 for high field and high temperature applications [8]. Secondly, results from transport measurements on Y-123 grown on a substrate consisting of a series of step edges. In what follows, the microstructure of these materials will be described and then related to the various transport measurements performed.

II. MATERIALS AND MICROSTRUCTURE

Microstructural characterization of a Y-123 sample, grown in-situ by laser ablation on the substrate with a series of step edges, was also performed in order to verify the types of grain boundaries present. Figure 1 shows a TEM image of a typical [100] tilt boundary. The lattice fringes observed in this micrograph represent the direction of the Cu-O planes. This micrograph is similar to results of other work using single step edge substrates [9]. This sample contains a series of very well defined grain boundaries which are continuous across the conduction path. The thickness of the film is approximately the depth of the ion milled channels or 3000Å (see inset of Fig. 3A). Each channel creates four grain boundaries of a similar type. The repeat distance is about 13µm, indicating that for our measurements, where we sample a 1mm length of the material, we are actually sampling around 300 similar grain boundaries. The regions between the HAGB are epitaxial to the substrate (Many different areas of the film were imaged with very similar results).

The Ti-1223 sample was grown at General Electric using a thallium free precursor film of composition $\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ which was deposited using a spray coating technique. The film was then annealed in a two zone reactor where the

This work was supported by the United States Department of Energy, Division of Basic Energy Sciences-Materials Sciences, and Conservation and renewable Energy as a part of a program to develop electric Power Technology, under Contract No. W-31-109-ENG-38 (JDH,DJM,JH,KEG, JS) and by the National Science Foundation as part of the Science and Technology Center for Superconductivity under Contract No. DMR 88-09854 (DHK).

thallium vapor pressure could be controlled. The resulting films are phase pure, highly textured Tl-1223. The sample studied here is a film of typical $2.7\mu\text{m}$. The average grain size is roughly $1\text{-}5\mu\text{m}$ as determined from scanning electron microscopy.

Fig. 1. A TEM micrograph showing a grain boundary in a Y-123 film grown on a substrate containing many step edges.

In order to verify the textured microstructure and to determine the types of grain boundaries present in materials grown in this manner, TEM studies were initiated. Figure 2 shows a TEM image of a few typical grain boundaries in a section of the textured Tl-1223 thick film. From this micrograph, we can observe several different grain boundaries. At the point labeled A, the lattice fringes abruptly discontinue at the interface with the adjacent grain. The point denoted as B in the graph shows a very low angle grain boundary (LAGB). This demonstrates that the sample consists of a distribution of grain boundaries, both high and low angle. Since different types of grain boundaries exist in series and in parallel, it is expected that at small current densities, the current path will consist of LAGB with the strongest superconducting properties. However, there could exist regions where a single or series of HAGB extend across a large portion of or the entire conducting path. At zero field, the HAGB may be good superconductors. But as the magnetic field is increased, the critical current density is expected to decrease significantly.

III. EXPERIMENTS

We report electrical transport measurements using the standard four probe technique. The temperature was controlled in a standard gas-flow type cryostat using a calibrated carbon-glass resistive thermometer and a metal film heater in a cold bore superconducting solenoid capable of reaching fields of 12 T. The resistance of the thermometer was corrected for magnetoresistive effects. The AC resistive transition measurements were performed using standard lock-in detection techniques and the current density used in the measurements was $3.3\text{A}/\text{cm}^2$. The DC electric-field vs. current density (E-J)

measurements were performed using a Keithley 224 digital current supply and a Keithley 182 digital sensitive voltmeter, typically capable of measuring voltage levels around 20-30 nV. Temperature control in these measurements was stable to less than 30 mK. Electrical contact was made to the materials using sputtered gold pads and pressed indium contacts.

Fig. 2. A TEM micrograph showing a few grain boundaries in a thick-polycrystalline film of Tl-1223. Point A denotes a HAGB and point B is a LAGB.

IV. RESULTS

A. Y-123 Step Edge Samples

Figure 3A is a plot of the resistive transition for the polycrystalline Y-123 sample grown with well defined [100] tilt grain boundaries. This data is closely fit to the activation model with a temperature dependence of $U_0(T) \sim (1-t)$ which is valid for a material with SIS junctions. These fits are also shown on the same plot. At low temperatures and current densities, it is assumed that the dissipation is from the junction with the weakest superconducting properties.

Figure 3B is a plot of a family of E-J curves at 70K for the Y-123 sample. At larger current levels, one can observe the negative curvature observed for materials with much larger critical current densities. At lower temperatures, a family of E-J curves generated in the same way, show only negative curvature. In what follows we will try to understand this behavior and compare it to the results for the resistive transitions.

B. $\text{TlBa}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ Thick Film

Figure 4A shows the resistive transition for Tl-1223 on a semi-log plot vs. inverse temperature. Also shown on the plot are fits using an activation model, $\rho \sim \rho_0 \exp(-U_0(T)/k_B T)$, where $U_0(T) \sim (1-t)^2$ which is valid if the grain boundaries behave as SNS junctions [10]. Often, in a polycrystalline

system, a double transition is observed, one attributed to the intragranular material and one attributed to the intergranular coupling. Notice that there are not two well defined resistive transitions for this sample. This indicates that the grain boundaries are dominating the resistivity of the system. This temperature dependence of the activation energy is very similar to that observed for polycrystalline Tl-2212 thin films [10].

Fig. 3. (A) Resistivity vs. $100/T$ (symbols) for a Y-123 sample with a series of HAGB across the conducting bridge as well as lines indicating a $(1-t)$ dependence of the activation energy. (B) A family of E-J curves generated at 75K by varying the magnetic field on a log-log plot. Curves between those labeled are evenly spaced in magnetic field.

Figure 4B shows a family of E-J curves at 40 K for the Tl-1223 sample. Here we can observe an effect which may be directly attributed to HAGB. At small applied fields and large current densities, the E-J curves shift very noticeably as the field is increased. However, at slightly higher fields, the critical current density, as defined by an electric field criterion, is nearly magnetic field independent. The field independence extends from 1T to 4T at this temperature. Also notice that at this temperature, there always exists negative curvature in the E-J plot when viewed on a log-log scale.

V. DISCUSSION

The data in Fig. 3B are reminiscent of data generated from

the Ambegaokar-Halperin (A-H) model [12]. Realizing this, it is possible to fit the general shape of the E-J curve to the A-H model. In order to accomplish this, it is necessary to choose a value for the normal state resistance of the junction. In the analysis shown here, we chose a temperature dependent normal state resistance. This was dictated by the data which showed nearly ohmic behavior at 10T and at temperatures of 75 and 78K, although the resistance was noticeably smaller at 75K. This is a fairly important assumption since this value dictates the critical current, I_c , in the absence of thermal fluctuations, a parameter extracted from the model. This quantity will be shown here as an activation energy which will be compared to those extracted from the resistivity transitions show in Fig. 3A.

Fig. 4. (A) Resistivity vs. $100/T$ (solid lines) for a textured Tl-1223 thick film as well as fits (symbols) to an activated model as described in the text. (B) E-J curves on a log-log plot showing a sharp reduction in critical current at small applied fields. Also note the negative curvature as viewed on this type of plot which is characteristic of all polycrystalline materials.

The quality of the fits to the A-H model are not very good over the full range of data collected. This is probably due to non-uniformity over all grain boundaries. Nonetheless, with

these deficiencies aside, we can extract values for $I_c(B,T)$ which are related to the activation energies extracted from the R vs. T data using $U(H)=hI_c/c(1-t)k_B$. We get the results which are shown in Fig. 5. Given the uncertainties in extracting I_c from the E - J curves, it is very surprising that we get such good agreement for the activation energy extracted in two different ways. Furthermore, the form of the data is very similar to results shown recently for the field dependence of the activation energy for a Nb-AlO_x-Nb junction, where the coherent area of the Josephson junction is not the physical dimension, but a field dependent area, which at large fields is $U(H)\sim 1/H$ [12] corresponding to the area of a single flux quantum.

Fig. 5. Activation energies for a Y-123 sample containing uniform HAGB determined from two independent measurements as indicated in the text.

The ρ vs. $100/T$ curves shown in Fig. 4A may be fit at the low dissipation end with an activation energy which has a temperature dependence of $U(T)\sim(1-t)^2$ as stated earlier. This is consistent with results for polycrystalline Tl-2212 [10]. The field dependence of the activation energies extracted in this manner can be summarized as $U\sim B^{-0.9}$. This is very close to that measured for Y-123 with HAGB at large fields. We do not put much emphasis on this similarity since the orientation of the grain boundary with respect to the applied field, is not similar in the two systems.

In Fig. 4B, the E - J curves for the single thallium layer compound Tl-1223 shows only a region with "negative curvature" when viewed on a log-log plot. We have also noted a similar behavior in Tl-2212 polycrystalline films. In contrast, epitaxial Tl-2212 films have shown positive curvature in the same region of parameter space. Therefore, we believe this behavior is a result of the grain boundaries. Similar curvature is also observed for the Y-123 E - J curve at lower temperatures. This supports our assertion that the curvature is a result of the grain boundary properties.

The sharp reduction in the measured J_c at small applied fields can be attributed to the destruction of the

superconducting properties of HAGB. The field independent region is a result of the strong superconducting properties of the remaining low-angle grain boundaries as well as substantial pinning in the intragranular materials. Both types of grain boundaries could be observed in Fig. 2. This explanation is further supported by the magnetization hysteresis measurements. After magnetic flux has reached the center of the sample, the hysteresis collapses very quickly as the applied field increases eventually reaching a relatively field independent region. Subdividing the sample into four sections and measuring the magnetic hysteresis again shows reasonable scaling at small fields and very little change in the high field hysteresis, even at 10 K.

V. CONCLUSIONS

We have found agreement of activation energies attained from two independent transport measurements for a Y-123 containing a series of very similar [100] tilt grain boundaries. This indicates that the dissipation and the shape of the E - J characteristic is dominated by grain boundaries in a polycrystalline material. This is assumed to be the origin of the negative curvature in polycrystalline Tl-1223. Further work is required to isolate individual grain boundaries in order to determine specific detrimental structures.

REFERENCES

- [1] S. Jin, R. B. van Dover, T. H. Tiefel, J. E. Graebner, and N. D. Spencer, *Appl. Phys. Lett.* **58**, 868(1991).
- [2] Y. Yamada, B. Obst, and R. Flukiger, *Supercond. Sci. Technol.* **4**, 165 (1991).
- [3] D. Dimos, P. Chaudhari, and J. Mannhart, *Phys. Rev. B* **41**, 4038(1990).
- [4] see for example J. S. Martens, V. M. Hietala, T. E. Zipperian, G. A. Bawter, D. S. Ginley, C. P. Tigges, T. A. Plut, and G. K. G. Hohenwarter, *Appl. Phys. Lett.* **60**, 1013 (1992).
- [5] S. E. Babcock, X. Y. Cai, D. L. Kaiser, and D. C. Larbalestier, *Nature* **347**, 167(1990).
- [6] A. C. Wright, T. K. Xia, and A. Erbil, *Phys. Rev. B* **45**, 5607(1992).
- [7] J. W. Ekin, H. R. Hart, Jr., and A. R. Gaddipati, *J. Appl. Phys.* **68**, 2285(1990).
- [8] D. H. Kim, K. E. Gray, R. T. Kampwirth, J. C. Smith, D. S. Richeson, T. J. Marks, J. H. Kang, J. Talvacchio, and M. Eddy, *Physica (Amsterdam)* **177C**, 431(1991).
- [9] C.L. Jia, B. Kabius, K. Urban, K. Herrmann, J. Schubert, W. Zander, and A. I. Braginski, preprint.
- [10] D. H. Kim, K. E. Gray, R. T. Kampwirth, and D. M. McKay, *Phys. Rev. B* **42**, 6249(1990).
- [11] V. Ambegaokar and B. I. Halperin, *Phys. Rev. Lett.* **22**, 1364 (1969).
- [12] D. H. Kim, K. E. Gray, and J. H. Kang, *Phys. Rev. B* **45**, 7563(1992).

END

**DATE
FILMED**

12 / 8 / 92