1100 Wayne Avenue, Suite 1100
Silver Spring, Maryland 20910
301/587-8202

MANUFACTURED TO AIIM STANDARDS
BY APPLIED IMAGE, INC.

58916-00-105
49191-20-105
85088-24-105

Manuscript

January 1993

ANL/MSD/PP- - 78801

**TlBa₂CaCu₂O_x Epitaxial Films:
Irreversibility, 2-D Pinning, and Critical Currents***

J.D. Hettinger, D.J. Miller, K.E. Gray, D.H. Kim
Materials Science Division, Argonne National Laboratory, Argonne, IL 60439

J. DeLuca, P. Karas, J.E. Tkaczyk
General Electric Corporate Research and Development, Schenectady, NY 12301

M. Eddy
Superconductor Technologies, Inc., Santa Barbara, CA 93111

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.

DISCLAIMER

jmc

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.

MASTER
878

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

*Work supported by the U.S. Department of Energy, Division of Basic Energy Sciences-Materials Sciences, Conservation and Renewable Energy as part of a program to develop electric power technology (DJM, KEG), the Superconductivity Pilot Center (JDH) under contract #W-31-109-ENG-38 and the National Science Foundation-Office of Science and Technology Centers for Superconductivity contract #DMR-91-20000 (DHK).

RECEIVED

JUN 10 1994

OSTI

TlBa₂CaCu₂O_x Epitaxial Films: Irreversibility, 2-D pinning, and Critical Currents

J. D. Hettinger, D. J. Miller, K. E. Gray, D. H. Kim
Argonne National Laboratory, Argonne, IL 60439

J. DeLuca, P. Karas, J. E. Tkaczyk
General Electric Corporate Research and Development, Schenectady, NY 12301
and
M. Eddy
Superconductor Technologies, Inc, Santa Barbara, CA 93111

Abstract

Post-annealed epitaxial thin films of TlBa₂CaCu₂O_x (Tl-1212) have been fabricated using sputtered precursors. Values of the transport critical current (J_c) for Tl-1212 epitaxial thin films in magnetic fields up to 7 tesla and at temperatures ranging from 25 to 77K are reported. These values are compared with those for Tl-2212 epitaxial films, Tl-2212 polycrystalline films, and Tl-1223 polycrystalline thick films. This comparison explicitly demonstrates the roles of grain boundaries and anisotropy in limiting the transport J_c . Analysis also supports the existence of a universal pinning defect in every highly anisotropic high-temperature superconductor, with a lower bound on the size of the defect being approximately 12Å in diameter.

Single thallium layer compounds ($\text{TlBa}_2\text{Ca}_n\text{Cu}_{n+1}\text{O}_x$, $n=1, 2$, Tl-1212 and Tl-1223) have received a large amount of attention due to their relatively small anisotropies which yield better intragranular dissipationless current carrying properties than double layer compounds (Tl-2212 and Tl-2223) in the presence of a magnetic field[1-4]. Optimistic electrical transport properties of single layer materials in polycrystalline form have also promoted interest in these materials[3]. For fundamental studies of the single layer compounds and determination of the ultimate ability of these materials for current transport, one must eliminate all weak-link defects which are detrimental to the dissipationless current carrying process. It is therefore, important to grow epitaxial films or single crystals. Every high temperature superconductor (HTS) which has been grown in the geometry of an epitaxial film so far has had very large critical current densities[5,6], possibly approaching the optimum performance of these materials.

We describe a technique for fabricating Tl-1212 epitaxial films and the results of transport measurements on these films. The small-magnetic field critical currents are found to be enhanced over those of polycrystalline Tl-1223 and Tl-2212, while the samples are able to carry a critical current to much larger magnetic fields than Tl-2212 due to their enhanced range of irreversibility. By carefully characterizing the irreversibility line of Tl-1212 and comparing to the results for Tl-2212, Tl-1223, and Bi-2212, we demonstrate why the differences in the irreversible behavior for all highly anisotropic HTS are primarily determined by the coupling between Cu-O planes, and only slightly by pinning due to defects in the planes. It is a consequence of the similarity of "naturally occurring" defects in the Cu-O planes of all of these materials. This represents the first explicit evidence that "universal" pinning sites predominate in all of these highly anisotropic materials.

Epitaxial films were fabricated using sputtered precursors with a $\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ stoichiometry on LaAlO_3 . These films were annealed using a two zone reaction furnace with a sample temperature of 865C and a Tl_2O_3 source at 745C. The resulting films were nearly phase pure Tl-1212 with a small impurity phase of Tl-1223 (<5%). Rocking curves indicate a FWHM of 0.8° . Epitaxy was confirmed using electron backscattered channelling patterns. Although epitaxy could not be confirmed by large area scans due to surface morphology, patterns generated at single spots consistently yield a larger proportion of

images showing the four-fold symmetry of the [001] axis and these patterns are parallel (a and b axes are also parallel). Thus, we conclude that although there are some misoriented grains and small impurity phases, there exists a dominant underlying epitaxial film.

Microbridges were defined to be $120\mu\text{m} \times 1\text{mm} \times 2500\text{\AA}$. Two samples of different thickness were measured yielding very similar results in spite of a slightly different surface morphology size scale. Transport measurements were made using standard four-terminal techniques. The irreversibility line was determined by AC transport using a current density of $30\text{A}/\text{cm}^2$ and a resistive criterion of $\rho/\rho_n=10^{-5}$. The critical currents (J_c) were determined as a function of magnetic field for the field directed along the c-axis using DC transport. An electric-field (E_c) criterion of $1\mu\text{V}/\text{cm}$ was used for all J_c measurements.

Figure 1 shows the critical current density as a function of magnetic field comparing it to several other samples of Tl containing superconductors with varying microstructures all at similar reduced temperatures ($T/T_c \sim 0.75$). An E_c of $12.5\mu\text{V}/\text{cm}$ was used in the case of the Tl-1223 thick polycrystalline film, whereas $E_c=1\mu\text{V}/\text{cm}$ for all other samples. In comparison with polycrystalline samples of Tl-1223 and Tl-2212, the low magnetic field critical currents of epitaxial Tl-1212 are enhanced by a factor of ten. The J_c of the polycrystalline sample are probably reduced due to the existence of weak-link grain boundaries. Although we can not entirely rule out the existence of high-angle-grain-boundaries in the Tl-1212 samples since our measurements did not extend to very small magnetic fields, we do not observe the usual evidence for grain boundaries in the shape of the electric field versus current density (E-J) characteristic at elevated currents and magnetic fields, i.e. if phase fluctuations across grain boundaries dominate the measured dissipation, the $\log(E)$ - $\log(J)$ characteristic displays negative curvature, or in other words, $\frac{d^2\log(E)}{d(\log(J))^2} < 0$, even at very large magnetic fields [7]. The high-magnetic field J_c of Tl-1212 epitaxial films are greatly enhanced in comparison with the polycrystalline and epitaxial Tl-2212 films. A comparison to polycrystalline Tl-1223 near the irreversibility line (where the J_c reduces toward 0), reveals a very comparable performance. This result is probably due to the similar anisotropies in each of the single thallium layer compounds[1].

Figure 2 shows the critical current density (electric field criterion, $E_c=1\mu\text{V}/\text{cm}$) as a function of magnetic field for several temperatures. Data was collected as E-J characteristics at a single temperature

while varying the magnetic field. This allowed an evaluation of the E-J characteristic in the region of the $1\mu\text{V}/\text{cm}$ dissipation level used as E_c in these measurements. At magnetic fields larger than those displayed in fig. 2, the E-J characteristic demonstrated ohmic behavior at the E_c used. Therefore, our samples do not support a dissipationless current by any reasonable definition at those fields and temperatures. The J_c 's shown are always extracted from non-linear E-J characteristics, i.e. $\frac{d\log(E)}{d(\log(J))} > 1$. As was shown in fig. 1, the rapid reduction of the J_c near the irreversibility line is an improvement over Tl-2212 which has considerably weaker interplanar coupling. The J_c is only weakly magnetic field dependent at small fields, lending support that critical currents are not limited by grain boundaries in these samples. The large J_c values at small fields may be attributed to the pinning of 3-D vortex lines since the interplanar coupling is expected to be large in this region of parameter space.

Resistance versus temperature data collected in the presence of a magnetic field are shown as an inset of fig.3. Figure 3 also shows the irreversibility line measured at small current densities. These values were extracted from the data of the inset using the criterion $\rho/\rho_n=10^{-5}$. Irreversibility lines chosen using different criteria yield very similar qualitative features while modifying the fitting parameters described below only slightly.

In a model which describes the dimensional crossover of the vortices which we will apply to our irreversibility data, there are two energies considered[1]. One is the Josephson coupling energy between the superconducting Cu-O bi- or tri-layers, which may be written as,

$$E_{cj} = \frac{\pi h \Delta(T)}{2e^2 R_N} \tanh\left(\frac{\Delta(T)}{2k_B T}\right),$$

where R_N is the normal state resistance of the junction and $\Delta(T)$ is the temperature-dependent energy gap.

This energy scale determines the crossover from three-dimensional (3-D) vortex lines to two-dimensional (2-D) pancake-like vortices. The material parameter which determines the strength of the coupling is the distance between Cu-O bi- or tri-layers in these materials denoted in ref. [1] as d_j . The energy coupling the Cu-O planes has been shown to depend exponentially on d_j , as expected for tunneling. Values of d_j are 9.57, 9.59, 11.55, and 12.29 Å, for Tl-1212, Tl-1223, Tl-2212, and Bi-2212 respectively. Since this is not the energy scale which will be focused on in the following analysis, a detailed discussion will not be presented here since one may be found in ref. 1.

The second energy scale which must be considered is that which determines 2-D vortex motion. Three-dimensional vortex lines are assumed to be strongly pinned and are therefore not considered in the model. A possible description of the 2-D vortex motion is that an individual 2-D vortex must overcome pinning forces due to defects which are normal or exhibit reduced superconductivity. This may be explicitly written in terms of the pinning energy, E_p , as

$$E_p = (1-Q)\pi \frac{d^2}{4} s \frac{B_c^2(T)}{2\mu_0} (1-b)^2, \text{ for } d < 2\xi_{ab},$$

where $1-Q$ is related to the "depth" of a potential well and accounts for the possibility for regions of reduced, but non-zero, superconductivity contributing to the pinning. The value d is the defect diameter, s is the unit cell thickness, which is the relevant length of defect along the flux line, and the term $(1-b)^2$ ($b \equiv B/B_{c2}(T)$) is introduced to account for the loss of condensation energy due to the applied magnetic field. The value $(1-Q)$ is related to the pinning factor, α_p , in ref. [1,10] by the expression $\alpha_p = (1-Q)(\frac{d}{2\xi_{ab}})^2$.

The total pinning energy which must be overcome for activated motion of a single vortex is $2E_{cj} + E_p$, where the coefficient 2 is included to account for the interaction with both adjacent Cu-O bi- or tri-layers. Therefore, when $2E_{cj} + E_p = k_B T_{irr}$, vortex motion occurs and the magnetization is expected to be reversible at or above T_{irr} . We assume the Josephson coupling energy to have a temperature dependence of $(1-t)$.

The solid line in fig. 3 is a fit of the data to the model described above. The parameter which comes out of this fit that is relevant to the forthcoming discussion is $d_{eff} \frac{B_c^2(0)}{2\mu_0}$, where we have introduced the effective defect size, $d_{eff} = \sqrt{1-Q} d$, which is the defect size if $Q=0$. In order to determine a value for the parameter d_{eff} , we must know $B_c(0)$. One way to independently estimate $B_c(0)$ is to assume that it is proportional to the zero field T_c and scale to results for fully oxygenated $YBa_2Cu_3O_7$ [8]. This allows us to extract a value of $12.5\text{\AA}$ for d_{eff} in the Tl-1212 films.

Now, a comparison will be made with values of d_{eff} for other materials from work reported elsewhere. Using the expression mentioned earlier relating the values of the defect size, d , to α_p and inserting the definition of d_{eff} gives $d_{eff} = 4\alpha_p \xi_{ab}$. Again, extracting the thermodynamic critical field as

described earlier, calculated d_{eff} for several materials yields 12.5, 11.5, 12.5, and 11.5 Å for Tl-1223 polycrystalline thick films[3], Tl-2212 epitaxial films[1], Bi-2212 crystal #1, and Bi-2212 crystal #2 [9,10] respectively. This indicates that the defect size is similar for each of these materials, implying similar pinning sites. The average value of 12 Å is a very reasonable value, e.g., it corresponds to a cluster of nine nearest neighbors unit cells in the ab-plane. This is a very important result since it indicates that the low temperature irreversibility region of each of these materials is dominated by pinning to the same types of reasonably plentiful defects. Therefore, the factor which determines good materials for applications is the Josephson coupling between Cu-O bi- or tri-layers.

In summary, we have fabricated Tl-1212 epitaxial films and demonstrated that all transport characteristics of the material are dominated by fundamental properties, not by extrinsic grain boundaries. At small fields the critical currents approach those found for epitaxial Tl-2212, while at larger fields, the irreversibility line approaches that of Tl-1223. The enhanced irreversibility region is due to the relatively small anisotropy of Tl-1212 which is similar to that of Tl-1223. Based on this analysis, we anticipate that the intragranular J_c in Tl-1223 will have a similar dependence on reduced field and temperature as the Tl-1212 results presented. We have also shown that the pinning centers responsible for pinning 2-D vortices after the planes are decoupled may be similar for all materials studied here regardless of microstructure indicating that the main difference determining the dissipationless current carrying capacity of these materials is the Josephson coupling between Cu-O bi- or tri-layers.

We acknowledge the support of the D.O.E., Divisions of Basic Energy Sciences-Materials Sciences and Conservation and Renewable Energy as part of a program to develop electric power technology (DJM, KEG), and the Superconductivity Pilot Center (JDH), under contract #W-31-109-ENG-38, and the NSF-Office of Science and Technology Centers under Contract #DMR 91-20000(DHK).

References

1. 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 C* **177**, 431 (1991).
2. R. S. Liu, D. N. Zheng, J. W. Loram, K. A. Mirza, A. M. Campbell, P. P. Edwards, *Appl. Phys. Lett.* **60**, 1019(1992).
3. John A. DeLuca, Pamela L. Karas, J. E. Tkaczyk, Peter J. Bednarczyk, Mary F. Garbauskas, Clyde L. Briant, and Donald B. Sorensen, Preprint, submitted to *Physica C*, accepted.
4. D.S. Ginley, J. F. Kwak, E. L. Venturini, B. Morosin, and R. J. Baughman, *Physica C* **140**, 42 (1989).
5. J. D. Hettinger, D. H. Kim, K. E. Gray, U. Welp, and R. T. Kampwirth, *Appl. Phys. Lett.* **60**, 2153 (1992).
6. J. D. Hettinger, A. G. Swanson, W. J. Skocpol, J. S. Brooks, J. M. Graybeal, P. M. Mankiewich, R. E. Howard, B. L. Straughn, E. G. Burkhardt, *Phys. Rev. Lett.* **62**, 2044 (1989).
7. J. D. Hettinger, D. H. Kim, D. J. Miller, J. G. Hu, K. E. Gray, J. E. Sharping, K. Daly, C. Pettiette-Hall, J. E. Tkaczyk, and J. Deluca, *IEEE Transactions on Applied Superconductivity*, Vol. 3.
8. K. E. Gray, D. H. Kim, B. W. Veal, G. T. Seidler, T. F. Rosenbaum, and D. E. Farrell, *Phys. Rev. B.* **45**, 10071 (1992). The fitting of the irreversibility line in ref. 7 assumes a single α_p for all samples with varying coherence lengths. This would be the case if the defects are always larger than 2ξ . This is somewhat surprising since the lowest T_c sample has $\xi \sim 90\text{\AA}$, making $d > 180\text{\AA}$. However, this proportionality is justified within BCS theory and by many experimentally determined values of T_c and B_c .
9. J. R. Thompson, Y. R. Sun, H. R. Kerchner, B. C. Sales, B. C. Chakoumakos, A. D. Marwick, L. Civale and J. O. Thompson, *Appl. Phys. Lett.* **60**, 2306 (1992).
10. D. H. Kim, K. E. Gray, and J. D. Hettinger, Preprint, submitted to *Phys. Rev. B.* July (1992).

Figure Captions

Fig. 1: Plot comparing J_C of various materials with different microstructures explicitly demonstrating the role of grain boundaries in polycrystalline materials and the separation between Cu-O planes in determining the irreversibility line. All J_C 's were determined using an electric field criterion of $1\mu\text{V}/\text{cm}$ except the Tl-1223 sample which used $12.5\mu\text{V}/\text{cm}$.

Fig. 2: Plot of the magnetic field dependent J_C using an electric field criterion of $1\mu\text{V}/\text{cm}$. In particular, notice the insensitivity of J_C to small applied fields indicating that weak-link grain boundaries do not play an important role in limiting the J_C .

Fig. 3: Plot of the irreversibility data (solid points) for Tl-1212 epitaxial films (as defined in the text) and a fit (solid line) to the vortex dimensional crossover model of Kim et. al. [Ref. 1]. *Inset:* Plot of the resistivity versus temperature data.

Fig. 1

Fig. 2

Fig. 3

DATE

FILMED

7/7/94

END