Microdiffraction measurements of the effects of grain alignment on critical current in high temperature superconductors

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ABSTRACT

While single crystals and epitaxial thin films of high temperature superconductors can carry large current densities, devices useful for applications such as power transmission and magnets cannot be produced because polycrystalline material cannot carry sufficient current densities. Efforts are underway to produce polycrystalline material in which grains are aligned to carry high current densities. We report x-ray and electron microdiffraction measurements of local grain alignment and models of how this grain alignment affects the critical current densities. $\text{TiCa}_2\text{Sr}_2\text{Cu}_3\text{O}_x$ samples can be grown on polycrystalline substrates with good c axis alignment but no overall a axis alignment. In $\text{TiCa}_2\text{Sr}_2\text{Cu}_3\text{O}_x$, high critical current occurs in regions in which there is local a axis alignment.

X-ray microdiffraction measurements of grain orientation were made with a monochromatic, 100 μm diameter beam produced by inserting a pinhole at the focus of an NSLS bending magnet beamline. Local grain orientation was measured by observing Bragg reflection as the sample was rotated. While x-ray data was taken at this low resolution over large areas, the orientation of individual grains was measured over small regions by measuring the Kikuchi pattern produced by inelastic scattering from a 100 nm electron beam. In both cases, the sample position was scanned to map grain orientation. With advanced x-ray optics currently under development, high-resolution maps of grain orientation will be available without the elaborate surface preparation required for electron diffraction. This will facilitate study of samples prepared in a wider variety of forms.

Keywords: x-ray, electron, microdiffraction, superconductor, microstructure, texture

2. INTRODUCTION

High-temperature superconductors have been shown to carry large current densities. Single crystals of $\text{YBa}_2\text{Cu}_3\text{O}_7$, for example, exhibit a critical current density (J_c) of $\sim 10^6 \text{ A/cm}^2$ at a temperature of 4 K in a magnetic field of 5 T.¹ Such high current densities can be carried only by single crystals and epitaxial thin films in which all grains are aligned.² For sintered $\text{YBa}_2\text{Cu}_3\text{O}_7$, in which grains are randomly oriented, $J_c \sim 10^2 \text{ A/cm}^2$ under similar conditions.³ Grain alignment is a key factor in the development of high-temperature superconductors for high-current applications.

Single crystal and epitaxial thin films of high-temperature superconductors are readily prepared in $\sim 1 \text{ cm}$ sizes, and may be suitable for many purposes. However, applications such as power transmission, motors, and magnets will require conductors $\sim 1 \text{ km}$ in length. One technique has been developed to the point of producing prototypes of this length.⁴ A silver tube is filled with $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ (Bi-2223), and the powder-in-tube assemblage is rolled into a flat tape, yielding a superconductor with partial grain alignment which carries $\sim 10^5 \text{ A/cm}^2$ at low temperatures. The tetragonal Bi-2223 grains are arranged with their c axes aligned normal to the tape, but with a axes in random directions in the plane of the tape, as shown schematically in Fig. 1. This has been called the 'brick wall' microstructure, based on the appearance of cross-sections [Fig 1(b,c)]. Grain boundaries consist predominantly of (001) twist boundaries [horizontal lines in Fig. 1(b)] and [001] tilt boundaries [vertical lines in Fig. 1(b)].⁵

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Both types of boundaries in the brick wall microstructure are typically large angle. The high J_c of this microstructure has been attributed to the favorable character of (001) twist boundaries: the misorientation of neighboring grains at such a boundary can be accommodated with minimal distortion of the superconducting CuO_2 planes. Two idealized models have been proposed for conduction. In the 'brick wall' model [Fig. 1(b)], intergranular current flows in the [001] direction across the twist boundaries.⁶ The 'switchyard' model [Fig. 1(c)] assumes that intragranular current flows strictly in the higher-conductivity (001) planes. In this model, intergranular current flow requires that the twist boundaries be slightly off the (001) planes.⁷ A third model proposes that current flows through a percolative path of small angle and special grain boundaries. Because such boundaries have lower energy than random boundaries, they will occur more frequently than dictated by chance alone.^{8,9}

Bi-2223 is intrinsically limited to low temperature applications: intragranular J_c falls to $\sim 10^3 \text{ A/cm}^2$ at 77 K and 1 T.⁴ High-current application at such a high temperature must use materials such as $\text{Tl}_1\text{Ca}_2\text{Sr}_2\text{Cu}_3\text{O}_x$ (Tl-1223), with intragranular $J_c \sim 10^5 \text{ A/cm}^2$ at 77 K and 1 T. A process has recently been developed for the production of high J_c Tl-1223 films using techniques which appear amenable to scaling to production of long lengths; an aerosol solution is sprayed on a polycrystalline substrate, followed by reaction in a furnace. While J_c is high, it is also highly variable, ranging from $10^3 - 10^5 \text{ A/cm}^2$ at 77 K in zero field and $10^2 - 10^4 \text{ A/cm}^2$ at 77 K in 1 T, in similarly prepared samples, and even in different regions of the same sample.¹⁰

In this paper, we report x-ray and electron microdiffraction measurements which show that while Tl-1223 has the same 'brick wall' appearance as Bi-2223 films, it has a distinctive microstructure in which 'colonies' of grains are locally aligned in-plane (i.e., both the a and c axes are aligned). We show that variations in J_c are correlated with the nature of local grain alignment, and we develop models for current flow which quantitatively account for both the mean value and the variance of J_c .

3. EXPERIMENTAL

3.1 Sample preparation

Details are provided elsewhere.¹⁰ Briefly, an aerosol of Ba-Ca-Cu-Ag nitrates is sprayed onto $8 \times 12 \text{ mm}^2$ polycrystalline yttria-stabilized zirconia (YSZ) wafers. These substrates are fine-grained and exhibit no preferred orientation. The samples are heated in O_2 , converting nitrates to oxides. The critical step is the thallination, which is carried out in a two-zone furnace. One zone controls the sample temperature, another the temperature of a supply of Tl_2O_3 , which in turn controls the pressure of Tl_2O . Formation of high J_c material depends on three factors. Firstly, the sample must be raised to the reaction temperature of 860° C before it is exposed to Tl_2O vapor. Secondly, sufficient Tl must be added. The two-zone

Fig. 1. Schematic of Bi-2223 brick wall microstructure. Dashed lines illustrate the current path in the (b) brick wall and (c) switchyard conduction models.

furnace is used to satisfy these first two conditions. Thirdly, the deposited mixture must include a small quantity of Ag, although much of the Ag precipitates out of the superconductor. Microscopic examination of the films suggests that these growth conditions result in a transient melting of the film, leading to formation of the colony microstructure.¹¹ The films are 3 μm thick; in cross-section they have a 'brick wall' appearance, with grains 3-10 μm wide and 0.5 - 2 μm thick.⁵

3.2 Critical current

Sample were patterned for measurement of J_c as shown in Fig. 2. Results for five samples prepared under similar conditions are given in Table 1. J_c for 4 mm segments (AE in Fig. 2) is equal to the minimum J_c for the four 1 mm segments AB, BC, CD, and DE; for this reason the J_c for 1 mm segments is both higher and more variable. Both J_c and x-ray measurements were made on two samples; J_c values for each segment of these samples are given in Table 2. Details of J_c measurement are given elsewhere.¹⁰

3.3 X-ray microdiffraction

Measurements were made at beamline X-14 at the NSLS.¹² Bending magnet radiation was monochromatized by sagittally focussing Si(111) crystals, with wavelength 1.409 $\text{\AA}$, chosen just below the energy of the Cu K absorption edge so that absorption from the 3 μm film thickness is small.

The scattering geometry is shown in Fig. 3. A 0.2 x 0.2 mm^2 square slit is used to insure that the ion chamber downstream of this slit is a reasonably accurate monitor of the x-ray flux passing through the 0.1 mm diameter pinhole downstream of the ion chamber. This pinhole, in a Pt foil, is placed 30 mm upstream of the sample. The incident beam divergence is 4.5 mrad in the horizontal and 0.2 mrad in the vertical direction; convolved with the circular pinhole aperture, this results in a spot size at the sample of 0.13 mm horizontal and 0.09 mm vertical FWHM.

A four-circle diffractometer is used to orient the sample for diffraction. Out-of-plane alignment is measured by rocking curves through the (0010) Bragg peak of the Tl-1223 films, which is $\sim 2^\circ$ FWHM and does not depend strongly on

Table 1. Mean and standard deviation of J_c measured at 77 K and zero magnetic field for five 4 mm segments with a total of twenty 1 mm segments and calculated based on measured map of grain orientations.

length x width (mm^2)	measured J_c (10^4 A/cm^2)		calculated J_c (10^4 A/cm^2)	
	mean	S.D.	mean	S.D.
1 x 0.2	4.0	1.3	3.7	0.9
4 x 0.2	2.7	0.7	2.6	0.4

Fig. 2. Pattern for J_c measurement. Current is passed from A to E while voltage is measured along 1 x 0.2 mm^2 segments AB, BC, CD, and DE.

Fig. 3. Scattering geometry for x-ray microdiffraction.

Table 2. J_c for 1 mm segments of 4 x 0.2 mm test bridges, measured at 77 K in zero magnetic field.

sample	J_c (A/cm ²)			
	AB	BC	CD	DE
SS-463	28302	16038	39623	10378
SS-467	44118	18873	88235	85784

Fig. 4. Typical scans of Tl-1223 in-plane alignment in which (a) and single colony, and (b) two colonies are illuminated.

spot size. In-plane alignment is determined by scanning the (111 $\bar{8}$) Bragg peak. Because [001] crystal axes are well-aligned with the sample normal, this is performed by first rotating the sample 17° off the [001] orientation and then rotating the sample about its surface normal. Using the notation of Busing and Levy,¹³ the sample is mounted with its surface normal along the ϕ axis, χ is set to the inclination of the (111 $\bar{8}$) planes, ω is held at zero, and ϕ is scanned from 0 to 360°. An alternative geometry is discussed elsewhere.¹⁴ The beam is incident on the sample 38° off-normal, resulting in a 26% increase in spot size.

Typical scans are shown in Fig. 4. In Fig. 4(a), the beam illuminates a single colony. Although ~1000 grains diffract, all have the same in-plane orientation within a 10° FWHM distribution. The four-fold pattern reflects the tetragonal crystal symmetry. In Fig. 4(b), the beam falls on the boundary between two colonies. The unequal intensities of the four reflections from each colony are caused by both the non-circular shape of the x-ray beam's footprint on the sample and the limited precision of the diffractometer; the sample's center of rotation coincides with the center of the x-ray beam only to within 0.1 mm.

Diffraction was used to analyze in-plane orientation for both patterned and unpatterned samples. Fig. 5 illustrates the variation in orientation as a function of position along the 0.2 x 4 mm² test bridge of two patterned samples. Each circle on the plot corresponds to the position of a peak in a scan such as those shown in Fig. 4. Since the crystal has 90° in-plane periodicity, only a 90° sector is shown. In this representation, a random in-plane orientation would produce ~30,000 circles uniformly covering the plots, while a perfectly aligned film would produce a single horizontal row of circles. These samples are an intermediate case,

Fig. 5. Peak positions for in-plane orientation of patterned Tl-1223 samples.

exhibiting good local alignment with a continuous change in alignment within colonies [e.g. between C and E in Fig. 5(a)], with abrupt shifts in alignment between colonies [e.g. near point C in Fig. 5(a)].

On unpatterned samples large areas could be scanned. Fig. 6 is a map of the most common in-plane orientation for each of 546 scans over a $6 \times 8 \text{ mm}^2$ area. The two-dimensional colony microstructure is apparent in Fig. 6, and it may be quantified by considering the orientational correlation function

$$C(r, \phi) = \frac{\langle [I(r_0, \phi_0) - \bar{I}][I(r_0 + r, \phi_0 + \phi) - \bar{I}] \rangle}{\bar{I}^2}, \quad (1)$$

where $I(r, \phi)$ is the diffracted intensity at position r and angle ϕ , $\bar{I}$ is the average intensity, and the brackets $\langle \rangle$ indicate an average over all intensity pairs at a given angular and spatial separation. $C(0, \phi)$ is just the average autocorrelation function of the in-plane orientation. Assuming a Gaussian distribution, $C(0, \phi)$ will have a width equal to $\sqrt{2}$ times the colony mosaic. The length over which $C(r, 0)$ falls to zero gives a measure of colony size. The orientational correlation function for the scans of Fig. 6 is shown in Fig. 7. The colonies have a typical size of 0.4 mm and a mosaic of 16° FWHM.¹⁵

3.4 Electron Backscatter

Electron backscatter diffraction patterns (EBSP's) are used to determine the orientation of individual grains. Samples were mounted in a scanning electron microscope. The electron beam current was 5 nA, the spot size 100 nm, the energy 10 to 15 keV. The beam was incident at a 19° angle to the sample surface normal. Backscattered electrons were imaged on a phosphor screen 40 mm from the sample. EBSP's are the Kikuchi patterns formed by Bragg diffraction of inelastically scattered electrons. A typical EBSP is shown in Fig. 8. The four-fold $[001]$ pole is evident near the bottom of the figure; pattern-matching software is used to determine the precise orientation of the grain. The sample is rastered, and an EBSP is acquired each time a shift in the pattern is evident; such a shift marks a grain boundary.

Fig. 6. Map over a $6 \times 8 \text{ mm}^2$ area of TI-1223 of most common in-plane orientation.

Fig. 7. Orientational correlation function for TI-1223 as a function of angle at a variety of distances.

Fig. 8. Typical EBSD of Tl-1223

Typical maps of grain orientation are shown in Figs. 9 and 10. Fig. 9 illustrates a colony boundary which consists of a line of large-angle grain boundaries marking an abrupt boundary between two colonies. This is the basis for the model we use to calculate J_c . Fig. 10 illustrates a more complex region in which the boundary between colonies consists of a series of smaller-angle grain boundaries: the shift in orientation from one colony to the next is more gradual, and J_c will be higher than calculated in the models used in this paper.

4. CONDUCTION MODELS

Comparison of the J_c values of Table 2 to the orientation maps of Fig. 5 shows that colonies of good alignment correspond to high J_c . The highest J_c are in segments CD and DE of sample SS-467, in which in-plane orientation varies continuously, with no large-angle grain boundaries. Other segments include colony boundaries, in which orientation abruptly shifts at large-angle grain boundaries, and J_c is correspondingly lower.

The map of orientational distributions obtained for the sample shown in Fig. 6 provides sufficient data to quantitatively compare predicted and measured J_c for the colony microstructure. Calculated values are based on measurements by Nabatame of J_c on Tl-1223 films grown epitaxially on SrTiO_3 bicrystals at 77 K in zero magnetic field which show that the grain boundary critical current i_c is $1 \times 10^5 \text{ A/cm}^2$ for grains aligned to within 12° and 3×10^3 for less well-aligned grains¹⁶. We make the simple assumption that the distribution of grain boundaries at the boundary between colonies is composed of grains randomly selected from the distribution of orientations within each grain: there is an abrupt shift from one orientational distribution to the other at the intercolony boundary. This assumption gives a lower J_c than the alternative of a gradual shift in orientation between colonies.

Fig. 9. Pole figure and arrow map of a colony boundary composed of large-angle grain boundaries.

Fig. 10. Pole figure and arrow map of a colony boundary composed primarily of small-angle grain boundaries.

The calculated J_c between neighboring points i and j in the lattice in Fig. 6 is

$$J_c^{ij} = \frac{\langle I_i(\phi_i) I_j(\phi_j) i_c(\phi_i - \phi_j) \rangle}{\bar{I}_i \bar{I}_j}, \quad (2)$$

where $I_i(\phi_i)$ is the intensity of an x-ray scan at point i and $\bar{I}_i$ is the average intensity. The limiting-path model is used to calculate the total J_c for the lattice.¹⁷ In one dimension, the critical current of a chain is that of the weakest link. The limiting path model is generalization to two dimensions, providing an efficient algorithm for finding the line of weakest links which limits J_c .

In Table 1, the mean and standard deviations of J_c measured for patterned traces are compared with J_c values calculated from the unpatterned 6 x 8 mm² sample using this model. The 546 scans on the sample include 441 segments 1 mm long and 189 segments 4 mm long. J_c is calculated for each of these segments; the mean and standard deviation are in excellent agreement with the observed values. Thus we have shown that the colony microstructure determines J_c for these samples.

The mean colony size and the mosaic of in-plane orientation within each colony may be used to predict J_c for larger samples. This calculation requires simulation of J_c both within and between colonies, since if each colony has a large mosaic of in-plane orientations, J_c will be limited by intracolony transport. The fact that intercolony transport can be better than intracolony transport can be understood by considering a simple model where a grain boundary is either conducting or nonconducting. Intracolony J_c will remain zero until the conducting fraction reaches the percolation threshold (e.g., 0.5 for a square lattice), while intergranular J_c will be proportional to the fraction of conducting intercolony boundaries — greater than zero for any non-zero conducting fraction. Intracolony J_c is simulated by assigning orientation to each grain on a 100 x 100 square lattice following a Gaussian distribution, calculating grain boundary J_c as above based on Nabatame's results¹⁶, and applying the limiting path model. Intercolony J_c is simulated by assigning each colony a Gaussian distribution of orientations with a random mean orientation, using Eq. (2) to find intercolony J_c , and applying the limiting path model. The final result applies the limiting path model to the lattice of colonies in which each intercolony boundary is assigned the lowest of the intercolony J_c and the intracolony J_c for the two neighboring colonies.

As shown in Fig. 11, J_c simulated for square colony arrays is much lower than the observed J_c and the J_c calculated from actual distributions of grain orientations. For example, a 4 mm segment is 10 colonies long and has a mean measured J_c of 27,000 A/cm² (Table 1), a value which is in good agreement with that calculated from actual grain distributions, while J_c calculated from simulated grain distributions is only 3,000 A/cm² (Fig. 11). This is due in part to the choice of a square lattice, in which a colony has only four neighbors; a hexagonal lattice with six neighbors would yield higher J_c . More importantly, the measured colony boundaries are not straight lines. The actual winding boundaries provide a longer boundary than does the simulation. Even so, the result is sobering: as the

Fig. 11. Simulated J_c for square arrays of colonies as a function of array size.

sample size increases, J_c falls to the minimum value for intracolony transport.

Can this result be improved by optimizing the colony mosaic? As shown in Fig. 12, J_c is only weakly affected by colony mosaic for zero magnetic field (solid line). No matter what the mosaic, either intracolony or intercolony transport limits J_c to near its minimum value. Mosaic can make a more dramatic difference in a magnetic field, when i_c becomes more sensitive to magnetic field. Based again on Nabatame's data,¹⁶ we take i_c in a magnetic field of 0.1 T to be 10^5 A/cm² for grain boundaries of 12° or less and 300 A/cm² for greater misorientations. As shown in Fig. 12 (dashed line), an optimum colony FWHM of 29° can increase J_c by a factor of 15 above its minimum value.

5. FUTURE DIRECTIONS

Using x-ray diffraction with a 0.1 mm probe and electron diffraction with a $0.1 \mu\text{m}$ probe, we are developing a comprehensive model of conductivity in Tl-1223 superconductors. One limitation of present techniques is the surface sensitivity of electron probes. We have been able to obtain high spatial resolution only for exposed surfaces which are flat and clean. Can x-ray microdiffraction be used to measure grain-by-grain orientation of buried layers and of films with ill-conditioned surfaces?

As discussed elsewhere in this Proceedings, rapid progress is being made in the production of x-ray microbeams, and we can soon expect high-flux beams with sub-micron diameters. Such a microbeam can be used in several ways. The technique described in this paper is to illuminate the sample with a monochromatic beam and rotate about the sample surface normal. Because a monochromatic beam is used, the scattering from the superconducting film is separated from the substrate scattering by setting the detector to the correct Bragg angle. This approach is limited by the requirement that the beam illuminate the same spot on the sample as the sample rotates: the best four-circle goniometers have only $\sim 10 \mu\text{m}$ precision. Furthermore, the need to rotate the sample through a large angle at each point limits the speed of data collection.

Another data collection scheme is to illuminate the sample with a white microbeam and measure the diffraction pattern as the sample is rastered. No rotation is required because a white beam is used; each grain will diffract in any orientation. This is a rapid way to acquire data with high spatial resolution. The challenge will be to separate scattering of the substrate from that of the

Fig. 12. Effect of colony mosaic on simulated J_c for 100×100 square arrays of colonies.

Fig. 13. A monochromatic beam with the proper energy will produce Bragg reflections from an aligned film.

superconducting film. This scheme may prove useful for films on amorphous substrates, which produce a diffuse scattering background which is readily distinguished from the Bragg diffraction of the crystalline film, or on single crystals, in which the substrate diffracts at known angles. For films on polycrystalline substrates, the substrate will produce a random array of diffracted beams that will make analysis of the film difficult.

A third scheme takes advantage of the alignment of the film. Each set of planes (hkl) with spacing d will be tilted at a particular angle χ to the surface normal. For example, the (1118) planes of Tl-1223 are tilted 17° from the [001] surface normal. When the sample is illuminated by x rays of wavelength $\lambda = 2d\cos(\chi)$ at normal incidence, all these planes will diffract simultaneously; as shown in Fig. 13. The in-plane alignment can then be measured by observing the diffraction pattern. Because diffraction from the film will occur along a ring of fixed Bragg angle, it can be readily distinguished from substrate scattering. A small variation in the film alignment may be accommodated by the convergence of the incident beam. Large variations can be accommodated by increasing the bandwidth of the "monochromatic" incident beam: a bandpass $\Delta\lambda$ will lead to a spread $\Delta\chi = 0.5 \cot \chi \Delta\lambda/\lambda$ in tilt angles.

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7. REFERENCES

1. T.R. Dinger, T.K. Worthington, W.J. Gallagher, and R.L. Sandstrom, "Direct observation of electronic anisotropy in single-crystal $Y_1Ba_2Cu_3O_{7-x}$ ", *Phys. Rev. Lett.* **58**, 2687-2690 (1987).
2. D.P. Hampshire and S.-W. Chan, "The critical current density in high fields in epitaxial thin films of $Y_1Ba_2Cu_3O_{y-\delta}$: Flux pinning and pair breaking", *J. Appl. Phys.* **72**, 4220-4226 (1992).
3. Y. Yamada, N. Fukushima, S. Nakayama, H. Yoshino, and S. Murase, "Critical current density of wire type Y-Ba-Cu oxide superconductor", *Jpn. J. Appl. Phys.* **26**, L865-L866 (1987).
4. P. Majewski, "Processing of high-temperature superconducting tapes", *Adv. Mater.* **6**, 593-594 (1994).
5. D.J. Miller, *et al.*, "Brick-wall structure in polycrystalline $TlBa_2Ca_2Cu_3O_x$ thick films with high critical currents", *Appl. Phys. Lett.* **63**, 556-558 (1993).
6. L.N. Bulaevskii, "Limits to the critical current in high- T_c superconducting tapes", *Phys. Rev. B* **48**, 13798-13816 (1993).
7. B. Hensel, J.-C. Grivel, A. Jeremie, A. Perin, A. Pollini, and R. Flükiger, "A model for the critical current in $(Bi,Pb)_2Sr_2Ca_2Cu_3O_x$ silver-sheathed tape: The role of small-angle c-axis grain boundaries and of the texture", *Physica C* **205**, 329-337 (1993).
8. D.M. Kroeger and A. Goyal, "Models for long-range current flow in bulk oxide superconductors", *J. Min. Mat. Soc.* **46**(12), 14-19 (1994).
9. A. Goyal, E.D. Specht, D.M. Kroeger, T.A. Mason, D.J. Dingley, G.N. Riley, and M.W. Rupich, "Grain boundary misorientations and percolative current paths in high- J_c powder-in-tube Bi-2223", *Appl. Phys. Lett.* **66**, 2903-2905 (1995).
10. J.A. DeLuca, P.L. Karas, J.E. Tkaczyk, P.J. Bednarczyk, M.F. Garbauskas, C.L. Briant, and D.B. Sorensen, "The preparation of "1223" Tl-Ca-Ba-Cu-oxide superconducting films via the reaction of silver-containing spray deposited Ca-Ba-Cu-oxide with thallium oxide vapor", *Physica C* **205**, 21-31 (1993).

11. A. Goyal, E.D. Specht, D.M. Kroeger, J.E. Tkaczky, C.L. Briant, and J.A. DeLuca. "Formation of colonies of locally aligned grains during thallination of spray-pyrolyzed $\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ thick films", to be published in IEEE Transactions.
12. A. Habenschuss, G.E. Ice, C.J. Sparks, and R.A. Neiser, "The ORNL beamline at the National Synchrotron Light Source", *Nucl. Instrum. and Meth. A* **266**, 215 (1988).
13. W.R. Busing and H.A. Levy, "Angle calculations for 3- and 4- circle x-ray and neutron diffractometers", *Acta Cryst.* **22**, 457 (1966).
14. E.D. Specht, A. Goyal, D.M. Kroeger, J.A. DeLuca, J.E. Tkaczky, C.L. Briant, and J.A. Sutliff, "X-ray microdiffraction analysis of mm-scale orientational correlations in Tl-1223 high-critical-current, high-temperature superconducting films", *Physica C* **226**, 76-84 (1994).
15. E.D. Specht, A. Goyal, D.M. Kroeger, J.A. DeLuca, J.E. Tkaczky, C.L. Briant, and J.A. Sutliff, "The effect of colonies of aligned grains on critical current in high temperature superconductors", *Physica C* **242**, 164-168 (1995).
16. T. Nabatame, S. Koike, O.B. Hyun, I. Hirabayashi, H. Suhara, and K. Nakamura, "Transport superconducting properties of grain boundaries in $\text{TlBa}_2\text{Ca}_2\text{Cu}_3\text{O}_x$ thin films", *Appl. Phys. Lett.* **65**, 776-778 (1994).
17. J. Rhyner and G. Blatter, "Limiting-path model of the critical current in a textured $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ film", *Phys. Rev. B* **40**, 829-832 (1989).

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