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The effect of colonies of aligned grains on critical current in high-temperature superconductors

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Abstract

X-ray microdiffraction has been used to map the orientational distribution of grains in a 4×5 mm^2 polycrystalline film of $Tl_{1-y}Ba_2Cu_2Cu_3O_x$ (Tl-1223) grown on yttria-stabilized zirconia (YSZ). The film consists of "colonies", each containing ~ 10^4 grains with their *a*-axes aligned; the *c*-axes of all grains are aligned normal to the film, but there is no overall *a*-axis alignment. While the grain boundaries within a colony are small angle, colonies are separated by large-angle tilt boundaries. The typical colony size is 0.4 mm. Measured critical currents are compared with simulations based on observed distributions of grain alignment; we find that variations in grain alignment account for the observed variations in the critical current density. Simulations over longer distances indicate that a colony structure can substantially increase the critical current, especially in magnetic fields.

High-temperature superconductors can exhibit excellent intrinsic transport properties, supporting high critical currents (J_c), even in magnetic fields. Tl-1223, for example can support an intragranular J_c of 10^5 A/cm^2 in a 1 T field at a temperature of 77 K and 10^7 A/cm^2 in 1 T at 4.2 K [1]. Thus far, however, these properties have been realized only in highly aligned materials such as epitaxial thin films. It remains a challenge to produce aligned materials which will carry current over the distances required for practical applications (typically ~ 1 m to 1 km).

A high J_c, both in and out of a magnetic field, has been reported in Tl-1223 films deposited on polycrystalline YSZ substrates [2,3]. The high values of J_c – over 10^5 A/cm^2 in zero magnetic field at a temperature of 77 K and 10^4 A/cm^2 in a 2 T field at 60 K – are of particular interest because the films are made using techniques which are amenable to the production of useful quantities of superconducting wire: the substrates are polycrystalline, and the films are spray-deposited. The high J_c's are not, however, obtained consistently; J_c varies by as much as a factor of 35 between different 1×0.2 mm^2 segments of a 4×0.2 mm^2 film. While the films have an overall *c*-axis alignment normal to the surface, segments of high J_c have been shown to coincide with colonies of grains, up to 1 mm in extent, with aligned *a*-axes; over larger distances the *a*-axes are aligned randomly in the plane of the film [4,5].

The colony microstructure observed in Tl-1223 films clearly leads to a high *intracolony* J_c. It has been suggested that this microstructure can produce a high *intercolony* J_c as well [4,5]. Because each colony contains grains with a distribution of orientations, even relatively high-angle colony boundaries will contain a fraction of low-angle grain boundaries. These low-

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angle boundaries may provide a percolative high-current path.

In this paper we determine under what conditions a colony microstructure can significantly enhance J_c over that of a microstructure with random in-plane alignment. X-ray microdiffraction is used to map the distribution of grain orientation over a 4×5 mm^2 area, providing a statistical measure of the colony size and of the mosaic of orientations within each colony. Simulations, based on this orientational map and on measurements of J_c as a function of misorientation at the grain boundaries [6], show that grain alignment can account for a great deal of the variation observed in J_c for short segments. Simulations of the critical current of a randomly generated lattice of colonies show that in zero magnetic field this colony structure can increase J_c by a factor of 2.7 over a film with no in-plane alignment. Simulations of J_c in a magnetic field indicate a 15-fold increase for an optimal colony microstructure.

Measurements were made of a Tl-1223 film, 3 μm thick, deposited on polycrystalline YSZ. The deposition process is described in Refs. [2] and [3]. The films are composed of c-aligned grains which are typically 1 μm in thickness and 5 μm in lateral extent. A monochromatic synchrotron X-ray beam 0.1 mm in diameter was used to map the grain orientation as described in Ref. [4]. Briefly, the sample was mounted in a symmetric geometry (incident and diffracted beams at equal angles to the film normal), at a scattering angle and inclination set for diffraction from the {1 1 18} planes of c-aligned grains. A rotation of the sample about the film normal brings each grain into alignment for diffraction at an angle ϕ corresponding to its in-plane a-axis orientation. The distribution of the grain orientations was measured at each point on a 5×4 mm^2 square grid with 0.2 mm spacing. The crosses in Fig. 1 indicate the most common orientation at each measured point. The colony structure is reflected in the similar orientation of neighboring points.

The colony microstructure is described quantitatively by an orientational correlation function, shown in Fig. 2:

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

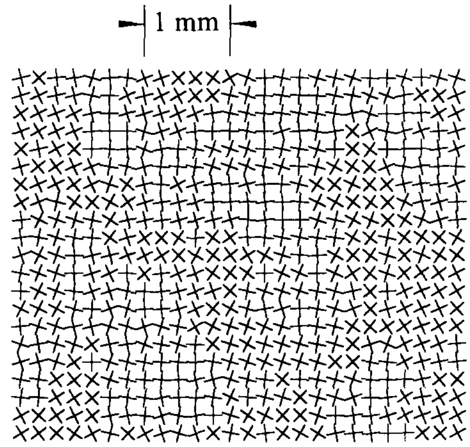

Fig. 1. Map of grain orientations. Each cross indicates the orientation of the a-axes for the most common orientation in a region 0.1 mm in diameter.

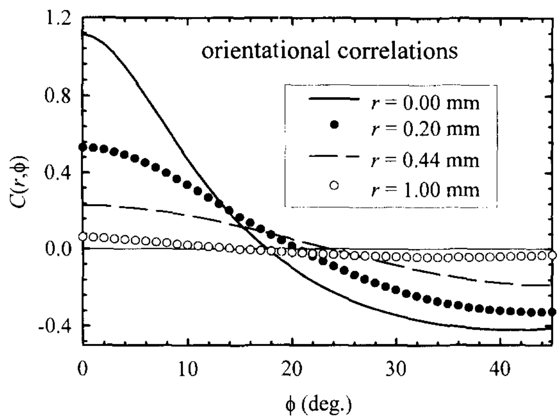

Fig. 2. Orientational correlation function, illustrating the decay of correlations with distance.

$I(r_0, \phi_0)$ is the diffracted intensity from position r_0 and orientation ϕ_0, $\bar{I}$ is the average measured intensity, and the brackets $\langle\,\rangle_{r,\phi}$ indicate an average over all pairs of measurements separated by a displacement r and misorientation ϕ. To better understand what the correlation function measures, let us consider a distribution of orientations $I(r_0, \phi_0)$ which is Gaussian for each position r_0. The zero-displacement correlation function $C(0, \phi)$ will be Gaussian as well, but with a width $2^{1/2}$ as great. Thus, we use $2^{-1/2}$ times the FWHM of $C(0, \phi)$ as a measure of the FWHM of the colony mosaic. For circular colonies of diameter D, $C(0.5D, 0)=C(0, 0)/2$; we use this equation to determine a statistical colony size D. In this way we find that the film consists of colonies with typical size $D = 0.4$ mm and mosaic 16° FWHM.

In order to test whether this colony microstructure causes the observed [2,3] variations in J_c, we have calculated J_c from the observed map of orientational distributions. For all near-neighbor points i and j in the array shown in Fig. 1, we compute the critical current

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

where the average $\langle\ \rangle$ is over all pairs of angles ϕ_i and ϕ_j. The grain boundary critical current i_c, based on measurements of Tl-1223 films on $SrTiO_3$ bicrystals at 77 K in zero magnetic field [6], is 1×10^5 A/cm^2 for grains aligned to within 12° and 3×10^3 A/cm^2 otherwise. The limiting-path model is used to sum all current paths and find J_c. We have applied the transfer matrix method without including returning paths; this has been shown to be an accurate approximation in this context [7]. In Table 1 we compare J_c measured for 1 and 4 mm long segments in a superconducting film to J_c calculated for all regions of the same dimensions within the area shown in Fig. 1. Calculations based on the observed colony microstructure give an excellent prediction of the average values of J_c, and account for most of the variation in J_c for these highly homogeneous films.

It has been shown [7] that J_c for a lattice of randomly oriented grains approaches, for large lattices, the lowest J_c (i.e. that of a large-angle boundary). A long-range texture increases J_c above this minimal value. A colony microstructure is an intermediate case, random overall but locally textured. To see what effect a colony microstructure will have on long wires, we have simulated J_c for square arrays of colonies, each with random orientation. The grain orientation within each colony follows a Gaussian distribution, with the same FWHM for each colony. The intercolony J_c is calculated using the limiting path model [7], as described above. For large FWHM within a colony, the current transport is limited by the intracolony J_c. This occurs for poor intracolony alignment because while a small fraction of the grains at colony boundaries are well aligned and can carry large currents across the boundary, there is no continuous high-current path across each colony.

The intracolony J_c is simulated by calculating J_c for a 100×100 square array of grains. Fig. 3 shows the intracolony, intercolony, and total J_c for a 100×100 square array of colonies. To see the qualitative effect of a colony microstructure on the in-field transport, we consider a weak magnetic field in which $i_c = 10^5$ A/cm^2 for grains aligned to within 12° and $i_c = 300$ A/cm^2 for larger angle grain boundaries. Fig. 4 shows the total J_c for square arrays of varying size in zero field.

These simulations suggest that J_c of short wires is increased more effectively by a colony microstructure than J_c of long wires. When a large variability is observed in J_c for short, narrow wires, as in Table 1, J_c for long, wide wires will correspond to the lowest value observed for shorter wires. In zero magnetic field, the colony microstructure produces only a modest variation in J_c for long wires, from 3×10^3 A/cm^2 in the limit of either very well or very poorly aligned colonies to 8×10^3 A/cm^2 for an optimum colony FWHM of 29°. In a magnetic field, a more dramatic increase is observed, from 3×10^2 to 5×10^3 A/cm^2.

In several respects, the models applied here are

Table 1
Mean and standard deviation of J_c for bridges at 77 K and zero magnetic field. The measured J_c's are for the most homogeneous films (schedule #3, static reactor; see Ref. [3]). The calculated J_c's use the measured map of grain orientations for 1 and 4 mm (Fig. 1) and a simulated array of colony with the same FWHM for 40 mm long bridges

Length × 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.
1×0.2	4.0	1.3	3.7	0.9
4×0.2	2.7	0.7	2.6	0.4
40×40	–	–	0.40	0.01

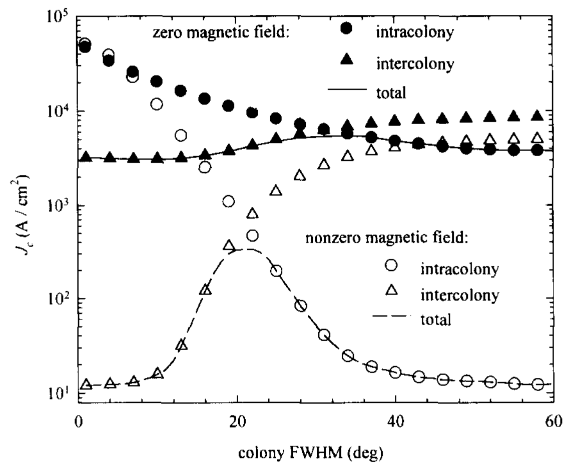

Fig. 3. Simulated critical currents for 100 × 100 arrays of colonies as a function of grain alignment within colonies. J_c is limited by intercolony transport for low FWHM and by intracolony transport for high FWHM.

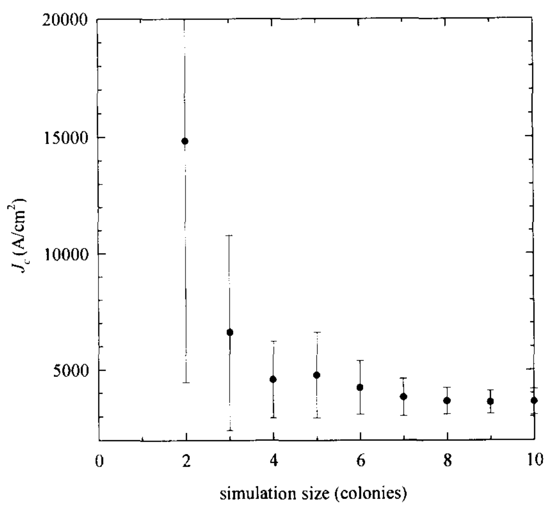

Fig. 4. Simulated critical currents for square colony arrays of varying size, in zero magnetic field. Error bars indicate the standard deviation for random selection of colony orientation.

conservative in their estimation of the enhancement of J_c of a colony microstructure over that of randomly oriented *c*-axis textured grains. These models assume that grains at colony boundaries follow the overall distribution of the grain orientation within the colony. If the distribution of grain orientation shifts continuously from one colony to the next, J_c would be much larger. “Special” large-angle grain boundaries (e.g. near-coincidence site lattice boundaries) with a high J_c may be another means of accommodating large colony misfits. Electron-backscatter measurements of grain orientation with high spatial resolution are in progress to determine whether continuous shifts in orientation and special boundaries are prevalent in the actual distribution of grain misorientations at colony boundaries [8]. Another method for increasing J_c is to produce an overall, rather than local, grain alignment [7]. This may occur more readily in a system, such as Tl-1223, which has a propensity for local alignment. A further increase may be effected by increasing the connectivity of the colony microstructure. In fact, simulations based on the observed colony structure (Table 1) produce a J_c twice as great as those based on square arrays of colonies (Fig. 4). We attribute this difference to two factors. Each colony in a square array has just four neighbors, which is unrealistically low: each colony in a hexagonal lattice, for example, would have six near neighbors, yielding a higher J_c. More importantly, the meandering colony boundaries provide a much larger colony boundary length than the straight boundaries of the square array.

Finally, note that we have considered strictly two-dimensional models. Three-dimensional current flow would increase the paths available for percolation [9,10]. Thicker films and films whose *c*-axes are not so well-aligned may have increased three-dimensional current flow. A key question is how the colony microstructure evolves with increasing film thickness.

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