## Large anisotropic critical magnetization currents in single-crystal YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub>

G. W. Crabtree, J. Z. Liu, A. Umezawa,\* W. K. Kwok,<sup>†</sup> C. H. Sowers, S. K. Malik,<sup>‡</sup>

B. W. Veal, D. J. Lam, M. B. Brodsky, and J. W. Downey

Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439

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Magnetization measurements on single crystals of  $YBa_2Cu_3O_{7-\delta}$  show strong anisotropic flux pinning with critical magnetization current densities of  $1.4 \times 10^6$  A/cm<sup>2</sup>. The critical magnetization currents fall sharply with temperature in both single-crystal and polycrystalline samples. The high critical magnetization current densities can be reduced by nearly two orders of magnitude by annealing.

The discovery<sup>1,2</sup> of the 90-K superconductor YBa<sub>2</sub>- $Cu_3O_{7-\delta}$  has led to an intensive effort to characterize this material for use in technological applications. The most important impact of the new superconductor probably will be the fabrication of wires possessing extremely high critical fields and critical currents. However, the measured<sup>3</sup> critical currents of this compound were found to be very low compared to good A15 superconductors. The low critical currents are often attributed to the poor connective nature of superconductivity due to granularity of the sintered polycrystalline samples of this compound. In this paper, we characterize the inherent behavior of bulk  $YBa_2Cu_3O_{7-\delta}$  using magnetization measurements on single-crystal samples. We find strong anisotropy of the flux pinning and the magnetization critical currents due to the presence of defects in the crystals. These defects can be controlled by annealing, leading to large changes in the magnetization critical current. The single crystals have magnetization critical currents two orders of magnitude greater than those found in polycrystalline samples.

Measurements were made on several single crystals with typical dimensions of  $0.3 \times 0.3 \times 0.3$  mm<sup>3</sup> grown by a technique described elsewhere.<sup>4</sup> One of the crystals was annealed at 500 °C in oxygen for 24 h before it was measured. X-ray measurements on several crystals from the same batch showed an orthorhombic unit cell with cell parameters a = 3.82 Å, b = 3.88 Å, and c = 11.67 Å. Laue x-ray patterns showed a clear distinction between the a and **b** directions. No evidence of "double reflections" associated with twinning was observed, demonstrating that the crystals were relatively untwinned. However, this method cannot quantitatively measure the degree of twinning and we expect from experience that some twinning defects are present in the crystals. dc susceptibility measurements on individual single crystals used in this work showed a sharp break at 91.7 K, essentially the same temperature as observed for a collection of single crystals prepared by the same technique and reported earlier.<sup>4</sup> In contrast to work reported elsewhere,<sup>5</sup> our crystals did not require any additional annealing to obtain the high transition temperatures. The polycrystalline samples used for comparison measurements were prepared by mixing powders of  $Y_2O_3$ , CuO, and BaCO<sub>3</sub> in the proper ratios, pressing the powder into pellets, reacting the pellets in flowing O<sub>2</sub> at 950°C for 24 h and finally annealing in

flowing  $O_2$  at 700 °C for 24 h, followed by a furnace cool to room temperature.<sup>6</sup>

Magnetization measurements were carried out using an SHE SOUID magnetometer in fields up to 10 kG and temperatures between 5 K and  $T_c$ . The magnetization at 5 K in the **a**, **b**, and **c** directions of an unannealed crystal is shown in Fig. 1. The strong irreversibility in the magnetization curve in all three directions obscures the intrinsic superconducting behavior. The initial magnetization in the **c** direction is linear only up to about 1 kG, giving an upper limit for  $H_{c1}$ . This upper limit is considerably smaller than the value of 5 to 8 kG reported elsewhere<sup>5</sup> for a single crystal with considerably more structural defects. Above 1 kG the magnetization curve continues to rise with a diamagnetic slope up to 9 kG, as is typical of samples with strong flux pinning. At high fields, the return curve has nearly the same slope as the initial magnetization curve, indicating that essentially all the flux which entered the sample at high fields is trapped. At lower fields, the slope of the return curve decreases as some of the trapped flux escapes, leaving a remnant flux equivalent to 55% of the flux which entered the sample at high field.



FIG. 1. Magnetization curves showing anisotropy of flux pinning and critical magnetization current. The derived values of  $J_c$  are  $1.4 \times 10^6$  A/cm<sup>2</sup> in the plane perpendicular to **c** and a factor of 10-50 less in the planes perpendicular to **a** and **b**, depending on applied field.

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In contrast to the c direction, the a and b directions show much less flux pinning. The remnant flux in these directions corresponds to about 5% of that which entered the sample at high field. Although the magnetization curves along the a and b directions are similar in shape, the magnitude in the a direction is consistently larger than that in the b direction.

The magnetization current density  $J_c$  can be obtained from these curves using the Bean model<sup>7,8</sup> of the critical state. The critical magnetization current for the field in the **c** direction (i.e., for current in the **a-b** plane) is  $1.4 \times 10^6$  A/cm<sup>2</sup>, independent of field up to 10 kG. This critical current is similar in magnitude to that derived from magnetization measurements in thin films<sup>9</sup> of YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub>. In the **a** and **b** directions the critical magnetization currents are much smaller and are strongly field dependent. For fields less than 1 kG,  $J_c$  for the **a** and **b** directions is a factor of 10 smaller than for the **c** direction, falling to a factor of 50 smaller at 10 kG.

The flux trapping and hysteresis for fields along the c direction decreases dramatically with increasing temperature, as shown in Fig. 2. The maximum values of the magnetization and the remnant flux are both a factor of 10 smaller at 45 K and a factor of 100 smaller at 77 K than at 5 K. The strong decrease in flux trapping at 45 K cannot be due to a degradation of the superconducting behavior since at  $T_c/2$  the upper critical field, the superconducting coherence length, and the magnetic penetration depth have nearly achieved their limiting low-temperature values. At 77 K, the magnetization curve is nearly reversible, there being less than 1 G difference between the forward and reverse curves above 5 kG. The critical magnetization current at 45 K is nearly field independent up to 5 kG, while at 77 K there is a strong decrease with field, as shown in Table I.

Magnetization measurements on polycrystals at 5, 45, and 77 K were carried out in order to compare the derived



FIG. 2. Magnetization curves for H parallel to **c** at 45 and 77 K.

critical magnetization current with that of the single crystals. The magnetization curve for the polycrystalline sample at 5 K resembles that for the c direction of the single crystal but with a factor of 10 reduction in magnitude. However, because the polycrystalline sample is a factor of 10 larger in dimension than the single crystal, the derived critical magnetization current is two orders of magnitude smaller than that of the single crystal. This shows the possibility of obtaining extremely high critical current density in bulk single crystals where no granularity problem exists.

At higher temperatures, the magnetization curves for the polycrystal become more reversible and the critical magnetization currents decrease significantly, as shown in Table I. At 77 K, the magnetization curve of the polycrystal is nearly identical to that of the single crystal with only 1 G separating the forward and reverse magnetiza-

T (K)	H (kG)	$J_c(A/cm^2)$			
		a	Ъ	с	Polycrystal
5	0-10			$1.4 \times 10^{6}$	
5	1.0	$1.2 \times 10^{5}$	$1.1 \times 10^{5}$		
	4.0	$7.3 \times 10^{4}$	$6.0 \times 10^{4}$		$1.4 \times 10^{4}$
	6.0	$5.2 \times 10^{4}$	$4.5 \times 10^{4}$		
	10.0	$3.2 \times 10^{4}$	$2.8 \times 10^{4}$		$1.3 \times 10^{4}$
45	1.0			1.1×10 <sup>5</sup>	
	2.0				$6.6 \times 10^{2}$
	4.0			$9.4 \times 10^{4}$	
	8.0				$6.2 \times 10^{2}$
	10.0			$6.9 \times 10^{4}$	
77	1.0			$1.1 \times 10^{4}$	
	2.0				$2.3 \times 10^{2}$
	4.0			$5.7 \times 10^{3}$	
	6.0				$1.2 \times 10^{2}$
	10.0			$4.3 \times 10^{3}$	

TABLE I. Temperature and field dependence of critical magnetization current on single and polycrystalline samples.

tion curves. The derived critical magnetization currents for the polycrystal sample shown in Table I are in good agreement with those reported elsewhere.<sup>10,11</sup>

The anisotropy in the flux pinning implies a pinning center whose spatial extent or spatial distribution is anisotropic. In order to obtain effective pinning, the distance lbetween the pinning centers must be larger than the coherence length  $\xi_0$ . A three-dimensional distribution of point defects cannot produce anisotropic pinning unless the ratio  $l/\xi_0$  varies from larger than one to less than one with direction. In  $YBa_2Cu_3O_{7-\delta}$ , the coherence length is large along the **a** and **b** direction and small along the **c** direction. Thus, in order to achieve the observed strong pinning anisotropy, the spacing between the pinning centers must be larger along **a** and **b** than along **c**. The oxygen vacancies known to occur in the one-dimensional chains in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub> are spaced farther along the c direction than along a and b. Therefore the inherent oxygen vacancies<sup>12</sup> in the structure cannot cause the anisotropic pinning patterns observed. A more likely candidate for pinning centers are (110) twin boundaries which occur on cooling the crystal through the tetragonal to orthorhombic phase transition. These plane defects will pin vortex lines along the c direction (e.g.,  $H \parallel c$ ) as they traverse the twinning plane. However, vortex lines along the **a** or **b** directions always intersect the twinning plane as they move through the crystal. Because their interaction energy with the twinning plane is independent of their position, they will not pin. Moreover, the twin boundary planes serve as strong pinning centers for vortices along the c direction because the flux line interacts with the twinning plane along its entire length. The origin of the pinning force could be the disruption of the onedimensional Cu-O chains which occurs at the twinning plane.

The flux pinning and the critical magnetization current can be significantly reduced by annealing. Figure 3 shows two measurements of the magnetization along the c direction on an annealed crystal. The initial magnetization curve resembles the data of the unannealed crystal shown in Fig. 1. However, at 7 kG, there is a sharp drop in the magnetization of more than a factor of two and the flux trapping on the return curve is reduced by approximately the same amount. The discontinuity in the magnetization reflects a flux jump in the sample, indicating that the pinning forces after annealing have been weakened. A second magnetization curve taken immediately after the first one shows almost no pinning with critical magnetization current of  $2 \times 10^4$  A/cm<sup>2</sup> compared with  $1.4 \times 10^6$ A/cm<sup>2</sup> for the unannealed crystal at the same tempera-



FIG. 3. Magnetization curves for H parallel to c at 5 K for an annealed single crystal. The first run shows a flux jump at 7 kG and the second run shows greatly reduced flux pinning. The solid line is a guide to the eye.

ture and field. A third magnetization curve resembled the first curve except that the flux jump occurred at 4 kG. This history-dependent behavior is expected for nonequilibrium systems where two states are very close in energy. The dependence of the magnetization curve on annealing demonstrates explicitly that the critical current can be controlled by sample-preparation techniques.

We have shown that critical magnetization currents achievable in bulk material far exceed those of sintered polycrystalline material. The current densities are sufficiently high for applications at low temperatures. However, the sharp decrease in the critical magnetization current with increasing temperature seems to be an inherent problem which is common to both polycrystalline samples and single crystals. Assuming this thermal degradation is unavoidable, our work suggests that a low-temperature critical current of order  $10^8 \text{ A/cm}^2$  is needed in order to achieve a working critical current of  $10^6 \text{ A/cm}^2$  at 77 K.

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- <sup>•</sup>Joint appointment, Department of Physics, University of Alberta, Edmonton, Canada.
- <sup>†</sup>Joint appointment, Department of Physics, Purdue University, West Lafayette, IN 47907.
- <sup>‡</sup>Permanent address: Tata Institute of Fundamental Research, Bombay, India.
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