

## Critical-magnetic-field anisotropy in single-crystal $\text{YBa}_2\text{Cu}_3\text{O}_7$

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We have measured the critical magnetic fields  $H_{c2\parallel}$  (parallel to  $a$ - $b$  plane) and  $H_{c2\perp}$  (perpendicular to  $a$ - $b$  plane) of single crystals of  $\text{YBa}_2\text{Cu}_3\text{O}_7$  as a function of temperature in fields up to 19 T. The slope  $-dH_{c2}/dT$  is greater by about a factor of 4 when the field is applied parallel to the  $a$ - $b$  plane than when it is applied parallel to the  $c$  axis.

### I. INTRODUCTION

One important aspect of the high- $T_c$  oxide superconductors<sup>1</sup> is the degree of anisotropy of their electronic properties arising from their layered crystal structure. Measurements of the critical magnetic field of polycrystalline samples gave hints of anisotropy,<sup>2</sup> however, recent experiments on single-crystal specimens have begun to show directly the extent of this anisotropy.<sup>3-6</sup> We present here critical-field measurements as a function of field direction on single crystals of  $\text{YBa}_2\text{Cu}_3\text{O}_7$  grown in our laboratory.

### II. EXPERIMENTAL TECHNIQUE

$\text{YBa}_2\text{Cu}_3\text{O}_7$  single crystals were grown by two different methods. In the first method, during the formation of the Y-Ba-Cu-O compound for fabricating sintered pellet samples, crystals formed on the surface of the reacted powder. These crystals,  $2 \times 2 \times 0.3$  to  $\sim 1 \times 1 \times 0.1$  mm<sup>3</sup> in size, were carefully removed and annealed in  $\text{O}_2$ . X-ray transmission Laue patterns showed them to be single crystals but with severe twinning. The second method of crystal growth was similar to that developed by Dinger, Worthington, Gallagher, and Sandstrom<sup>7</sup> and yielded many extremely flat, small crystals (area  $< 1$  mm<sup>2</sup> and thickness  $\lesssim 10$   $\mu\text{m}$ ), which were subsequently  $\text{O}_2$  annealed. Laue patterns of these samples proved them to be good single crystals with the  $c$  axis perpendicular to the flat face and with little twinning. Diagnostic susceptibility and resistivity measurements of these types of crystals showed them to be superconducting, with transition temperatures  $92.5 \pm 2$  K for the twinned Y-Ba-Cu-O crystals (which we designate type *B*) and  $89.7 \pm 2.4$  K for the less twinned crystals (which we designate type *A*). We present here critical field data for both types of crystal.

Gold wires were attached with silver paint to copper contacts evaporated onto the sample for four-terminal resistance measurement. The upper critical fields were determined by the resistive transition. Our earlier attempts with silver epoxy contacts invariably failed at low temperature, but with evaporated Cu contact pads, the leads remained good. For resistivity versus magnetic field measurements at various temperatures, the samples were mounted on a rotatable copper plate with a Pt resistance

thermometer adjacent to the sample. A small wire-wound heater supplied heat to the whole copper plate, thereby giving good stability in sample temperature. Helium exchange gas provided contact to the liquid- $\text{N}_2$  bath. A superconducting magnet with a room-temperature bore provided a magnetic field up to 15 T, and higher fields were obtained in a water-cooled Bitter magnet. The sample temperature was swept through the transition at various fixed fields. Critical field data were taken for both orientations  $H_{c2\parallel}$  and  $H_{c2\perp}$  (parallel and perpendicular to the  $a$ - $b$  plane, respectively) in the same run by changing the sample orientation.

### III. EXPERIMENTAL RESULTS

A resistivity measurement done in the  $a$ - $b$  plane of a type-*A* crystal in zero applied field is plotted against temperature in Fig. 1. At 280 K,  $\rho$  was estimated to be about  $400 \mu\Omega \text{cm}$ . The shape of the crystal was not ideal for determining the size of the conducting path, leading to a large uncertainty. The resistivity decreases linearly as  $T$  decreases with a slope of  $1.46 \mu\Omega \text{cm/K}$ , reaching a value of about  $140 \mu\Omega \text{cm}$  at 100 K. A line drawn through the data points extrapolates close to the origin. For comparison, the resistivity variation with temperature for a high-quality sintered pellet of Y-Ba-Cu-O is also shown in this

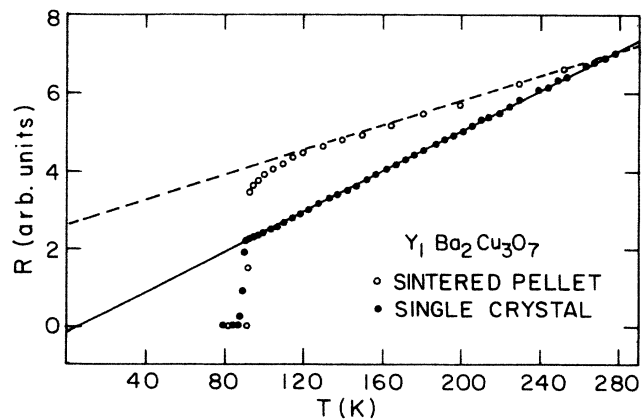


FIG. 1. Resistance vs temperature of a type-*A* single crystal and polycrystalline sample of  $\text{YBa}_2\text{Cu}_3\text{O}_7$ .

figure. As in sintered pellets,  $\rho$  decreases linearly with  $T$  to about 120 K. However, below this temperature there is a deviation from the linear behavior in the pellet, which has been attributed<sup>8</sup> to superconducting fluctuations and which is not visible in the single-crystal data. The slope at  $d\rho/dT$  is different in the two cases, which is not surprising since in the sintered pellet, grain boundaries play a significant role in the conduction process.

The measured upper critical fields of the type-*A* crystal as a function of temperature are shown in Figs. 2 and 3. The transition temperature  $T_c(H)$  was taken to be that temperature at which the resistance dropped to half of its value in the normal state at a measuring current of 200  $\mu\text{A}$  ( $\approx 10 \text{ A/cm}^2$ ). It was found that the  $T_c$  did not change with measuring currents from 100  $\mu\text{A}$  to 500  $\mu\text{A}$ , apparently eliminating the possibility of critical current or heating effects being significant. The temperatures have been corrected for the magnetoresistance of the Pt thermometer;<sup>9</sup> the corrections are as large as 1.2% at 15 T but decrease rapidly at lower fields. The transition temperature in zero-field  $T_{c0}=89.7 \text{ K}$  with a transition width of 2.4 K. As the magnetic field increased, the transition width increased, too. The midpoint as well as the 10% and 90% points of the resistive transition are shown in Figs. 2 and 3. The resistance for the normal state at any temperature and field was taken to be the extrapolation of the normal state shown in Fig. 1 to lower temperatures. As the temperature is lowered below  $T_c$ ,  $H_{c2\parallel}$  rises sharply, with a positive curvature close to  $T_c$ . The slope  $dH_{c2\parallel}/dT$

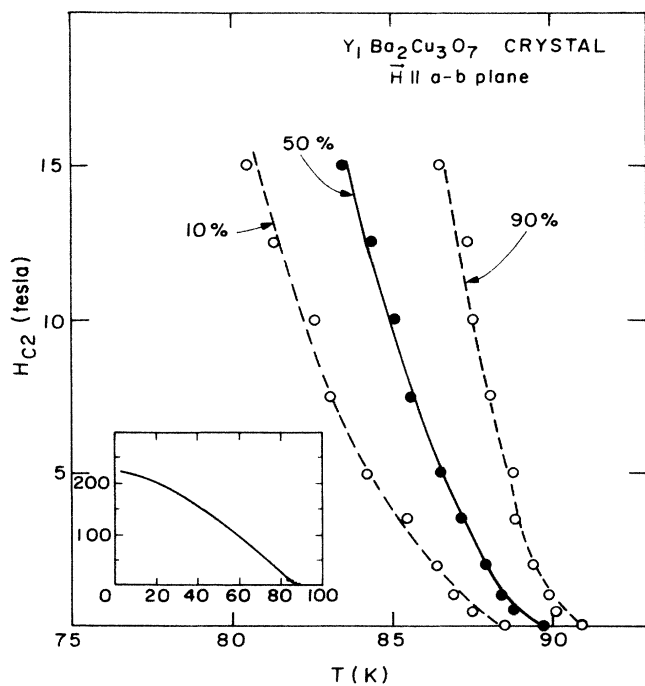


FIG. 2. Critical magnetic field vs temperature of a type-*A* crystal with the field in the *a-b* plane. The temperature at which 10%, 50%, and 90% of the normal-state resistance are restored are indicated for each field value. The inset shows the range of field and temperature covered by the measurements compared to the entire estimated phase boundary.

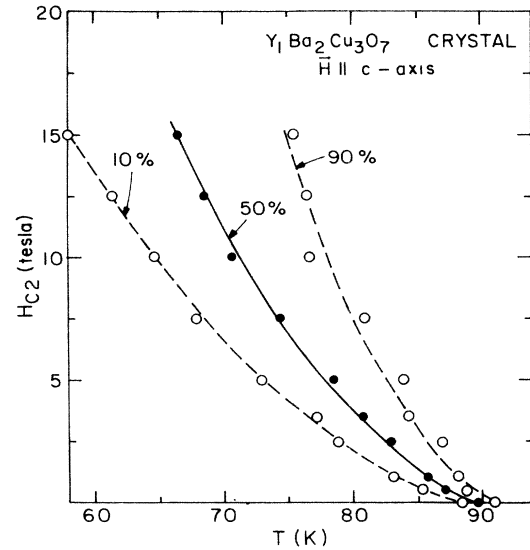


FIG. 3. Critical magnetic field vs temperature of a type-*A* crystal with the field perpendicular to the *a-b* plane. The temperatures at which 10%, 50%, and 90% of the normal-state resistance are restored are indicated for each field value.

near  $T_{c0}$  is  $-0.7 \text{ T/K}$ , whereas at about 85 K the slope is about  $-3.6 \text{ T/K}$ . For  $H_{c2\perp}$  the curvature persists down to the lowest temperature at which  $H_{c2\perp}$  could be measured. A slope of  $-0.2 \text{ T/K}$  was found very close to  $T_{c0}$  and  $-1.1 \text{ T/K}$  near 67 K. To estimate  $H_{c2\perp}(0)$ , we use the formula  $H_{c2\perp}(0)=0.69 [dH_{c2\perp}(T)/dT]T'$ . Here the slope is chosen to be the tangent to the curve near the highest field (15 T) and  $T'$  is at the intersection of this tangent with the temperature axis. From this  $H_{c2\perp}(0)=61 \text{ T}$ , giving a calculated coherence distance in the *a-b* plane  $\xi_{ab}(0)=23 \text{ \AA}$ . In a similar manner,  $H_{c2\parallel}(0)$  is estimated to be 222 T. However, the small temperature range, combined with the arbitrary use of a resistance equal to one-half of the normal-state resistance as the criterion to determine  $T_c(H)$ , makes the uncertainty of  $H_{c2\parallel}(0)$  as large as 25% even if the simple theory is valid for this material. With these reservations, and assuming there is strong Josephson coupling between layers and that  $H_{c\perp}(0)/H_{c\parallel}(0) = \xi_c(0)/\xi_{ab}(0)$ , the coherence distance perpendicular to the *a-b* plane  $\xi_c(0)$  is probably not more than 6.3  $\text{\AA}$ . The transition width at  $H_{c2\perp}$  becomes much wider than that at  $H_{c2\parallel}$  over the field range available, but it should be noted that as a function of temperature the transition widths broaden at about the same rate below  $T_{c0}$ . The high anisotropy of these crystals is shown by the ratios  $H_{c2\parallel}(0)/H_{c2\perp}(0) = \xi_{ab}(0)/\xi_c(0)=3.6$ .

The critical field measurements for type-*B* crystals are shown in Fig. 4. In this case, two crystals from the same batch were mounted with their *c* axis perpendicular to each other, so that the two curves representing the two crystalline directions are for different but similar crystals. The behavior of  $H_{c2}$  is more conventional in appearance in this case, with  $H_{c2\parallel}$  being nearly linear in  $T$  with a slope of  $-4 \text{ T/K}$ , and  $H_{c2\perp}$  having negative curvature with a slope of  $-0.96 \text{ T/K}$  near  $T_{c0}$ . The conventional dirty-limit

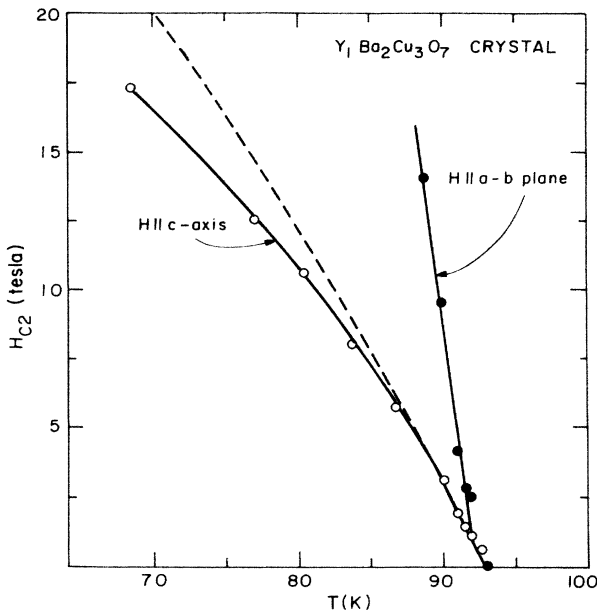


FIG. 4. Critical magnetic field vs temperature of a type-B crystal for the field in the  $a$ - $b$  plane (●) and perpendicular to the  $a$ - $b$  plane (○). The dashed line is the calculated critical field using the slope near  $T_{c0}$  of the perpendicular data.

theory<sup>10</sup> with this slope is shown for comparison by the dashed line, although the dirty-limit theory may not apply to this material because of the short coherence length. The extrapolated values of  $H_{c2\parallel}(0)$  and  $H_{c2\perp}(0)$  are 256 and 62 T, respectively.

#### IV. DISCUSSION

As we have seen, the two types of crystal have values of  $dH_{c2}/dT$  and  $\xi$  for the respective directions which agree within about 10% in spite of the details of the temperature dependence near  $T_{c0}$ . Further work is needed to determine what part of the differences in the two kinds of crystal is fundamental and what part is materials related. The more nearly perfect crystal, type A, was presumably somewhat deficient in oxygen, judging from the slightly reduced  $T_{c0}$ . Inhomogeneity might lead to the positive curvature we observed. The inset in Fig. 2 makes an important point, however, indicating the range of field and temperature explored in this experiment. Many superconductors show nonconventional critical-field behavior this

close to  $T_{c0}$ .

A number of other groups have made measurements of  $H_{c2}$  on single crystals. Results by Iye, Tamegai, Takeya, and Takei<sup>4</sup> in fields up to 9 T are qualitatively very similar, showing the strong anisotropy and curvature near  $T_{c0}$  of both  $H_{c2\parallel}(T)$  and  $H_{c2\perp}(T)$ . Analyzed in the same manner as our results, the data of Iye *et al.* give  $H_{c\perp}(0)=61$  T and  $\xi_{ab}(0)=23$  Å exactly as we obtained, whereas there is only 5% difference in the  $H_{c2\parallel}$  values. [Iye *et al.* took  $R=0$  to define  $T_c(H, T)$ , so their analysis is somewhat different.] Hidaka *et al.*<sup>3</sup> have also measured the critical fields of a single crystal of Y-Ba-Cu-O using the dc resistance method. They give a value of anisotropy factor of 5.5 in the critical fields; these data analyzed in the same way as ours appear to give a value of  $H_{c\parallel}(0)/H_{c\perp}(0)\approx 7$ . Both of these dc measurements were qualitatively similar to our results. There was strong anisotropy as well as strong curvature in both  $H_{c\parallel}$  and  $H_{c\perp}$  near  $T_{c0}$ . Also, there was an increased transition width as  $T$  was lowered below  $T_c$ . Worthington, Gallagher, and Dinger<sup>5</sup> have measured the critical fields of Y-Ba-Cu-O crystals by defining  $T_c(H)$  as the temperature of the most rapid change in reflected power from a 0.70 MHz resonant coil surrounding the crystal. They obtain a value of  $H_{c2\perp}(0)=29$  T which implies  $\xi_{ab}(0)=34$  Å and  $H_{c2\parallel}(0)=140$  T, giving a value of  $\xi_c(0)=7$  Å. The critical fields show no curvature but  $H_{c\perp}$  does not have a change of slope at 77 K. Noel *et al.*<sup>6</sup> have measured the anisotropy in the critical field to be about 8 for a single crystal of TmBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> by a resistive technique.

In summary, the various experiments give a value between 4 and 8 for the anisotropy in the critical field. As better characterized and more homogeneous samples become available, such matters as the details of the temperature dependence of  $H_{c2}$  can be cleared up. In addition, measurements to much higher fields are desirable as these would indicate the validity of the conventional theory<sup>10</sup> of the critical field at lower temperatures.

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