

## Upper critical fields and anisotropy limits of high- $T_c$ superconductors $R_1\text{Ba}_2\text{Cu}_3\text{O}_{7-y}$ , where $R = \text{Nd, Eu, Gd, Dy, Ho, Er, and Tm}$ , and $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$

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The upper critical fields  $B_{c2}(T)$  were measured up to 28 T for  $R_1\text{Ba}_2\text{Cu}_3\text{O}_{7-y}$ , where  $R = \text{Nd, Eu, Gd, Dy, Ho, Er, and Tm}$ , and for  $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$ . The midpoints  $(dB_{c2}/dT)_{T=T_c}$  cluster about  $(2.8 \pm 0.2)$  T/K, the  $T_c$ 's  $= (93 \pm 2)$  K show a slight systematic decrease from Nd to Tm, and the extrapolated values of  $B_{c2}(0)$  cluster about  $(160 \pm 20)$  T. The wide range of normal-state resistivities indicates that they are not intrinsic to the superconductor. The anisotropy  $A = B_{c2\parallel}/B_{c2\perp}$  is estimated to be less than 12 based on the data for the resistive transition. Criteria for the midpoint resistive transition are also reexamined.

In this Rapid Communication we present high-field data with applied fields up to 28 T on the high- $T_c$  superconductors  $R_1\text{Ba}_2\text{Cu}_3\text{O}_{7-y}$ , with  $R = \text{Nd, Eu, Gd, Dy, Ho, Er, and Tm}$  and  $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$ . The  $T_c$ 's are clustered within a few degrees about 92 K, the slopes of the midpoint critical fields  $(dB_{c2}/dT)_{T=T_c}$  cluster about 2.8 T/K, but the resistivities near  $T_c$  for the rare-earth compounds range from 200 to 2000  $\mu\Omega\text{cm}$ . The large variation in resistivity is not reflected in the superconducting properties. The rapid and nearly linear temperature dependence of the normal-state resistivity and the very broad nature of the resistive transition in a magnetic field present difficulties in defining  $B_{c2}(T)$ . Our present results define the resistive transitions using extrapolations of the resistivities from the linear region well above  $T_c$ . The resulting midpoint  $(dB_{c2}/dT)_{T=T_c}$  is more consistent and much higher than that obtained earlier by us<sup>1</sup> and others. The anisotropy of  $B_{c2}$  in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$  at 4.2 K is estimated to be less than 12 based on these high-field data.

These materials were prepared at Bell Communications Research as described in Ref. 2 and are the same samples used in Ref. 2 or sections cut from the same batch. All the compounds measured are single phase, except for Nd which has a few percent of a second unidentified phase. Although compounds with  $R = \text{Sm, Yb, and Lu}$  were also prepared, the upper critical fields were not measured in these because good electrical contacts were not obtained, although mechanical removal of the surface layer did not alleviate the problem. We assume this problem is due to a surface oxide.

The upper critical fields  $B_{c2}(T)$  were measured resistively with a standard dc four-probe technique in transverse dc magnetic fields in water-cooled magnets up to 20 T or in the 30 T Hybrid magnet at the Francis Bitter National Magnet Laboratory facility. The samples were

small ( $0.5 \times 3 \times 8 \text{ mm}^3$ ) rectangular polycrystalline slabs with low porosity. The leads were attached with Ag paint and a low dc current density ( $< 300 \mu\text{A}/\text{cm}^2$ ) was applied. A general-purpose temperature-controlled cryostat furnished by the facility was used for measurements to 20 T. Measurements were made in the same manner described earlier for Y-Ba-Cu-O.<sup>1</sup> The midpoint of the resistive transition was obtained at constant temperature and swept field.

The large temperature dependence of the resistance of these high- $T_c$  materials makes it difficult to define the midpoint of the transitions which are broadened radically in applied fields. If measurements are taken at temperatures near  $T_c$ , as has been done by us and others for Y-Ba-Cu-O, then one would define  $\rho_N(T)$  below  $T_c$  as the dashed line (labeled as  $l$ ) in Fig. 1, and the resulting resistivity as  $\rho_{Nl}(T)$ . However, if the resistivity is extrapolated from temperatures well above  $T_c$  (above about 115 to 120 K) then one would define  $\rho_N(T)$  below  $T_c$  as the dash-dotted line (labeled as  $h$ ), determined from the extrapolation as shown in the expanded  $T$  scale of the inset of Fig. 1. We have chosen what we believe to be the more reasonable definition as the  $h$  extrapolation with the midpoint  $0.5 \rho_{Nh}(T)$ . [If the  $l$  extrapolation were chosen,  $\rho_{Nl}(T)$  would become negative as  $T$  approached zero; clearly an inappropriate procedure.] Figure 1 shows the results for the  $\text{ErBa}_2\text{Cu}_3\text{O}_{7-y}$  specimen. However, the general features are reflected in all the  $R\text{Ba}_2\text{Cu}_3\text{O}_{7-y}$  data. The corresponding midpoints are indicated in the figures as  $0.5R_{Nh}$  and  $0.5R_{Nl}$ . Because the resistive transitions broaden rapidly in an applied magnetic field, the criterion  $0.5R_{Nl}(T)$  gives a lower  $B_{c2}(T)$  and a lower  $(dB_{c2}/dT)_{T=T_c}$  than  $0.5R_{Nh}(T)$ .

Figure 2 shows the resultant  $B_{c2}(T)$  data for the  $\text{NdBa}_2\text{Cu}_3\text{O}_{7-x}$  specimen using the  $l$  and  $h$  extrapola-

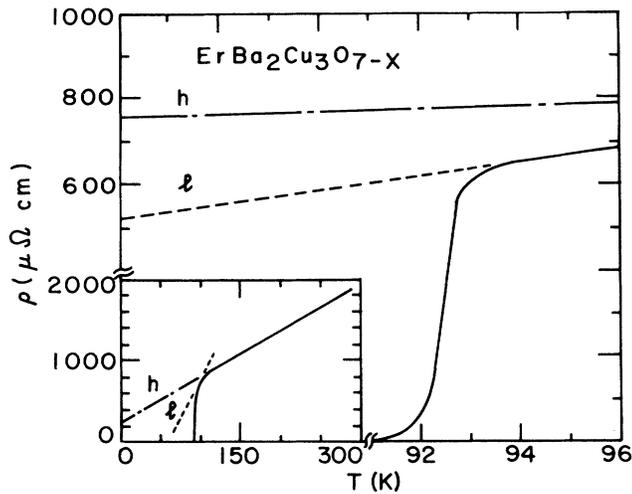


FIG. 1. Comparison of extrapolations of linear variation of normal-state resistance vs temperature  $T$  in  $R\text{Ba}_2\text{Cu}_3\text{O}_{7-x}$  with  $R=\text{Er}$  as an example. The upper dash-dotted line ( $h$ ) indicates the linear extrapolation from above about 115 K, whereas the lower dash ( $l$ ) line is obtained close to  $T_c$  where previous work was examined. The inset shows the comparison over a large  $T$  range.

tions. Both methods give a linear  $B_{c2}(T)$  which extrapolates to nearly the same value of  $T_c$  at  $B_0=0$ . It is clear in Fig. 2 that the  $(dB_{c2}/dT)_{T=T_c}$  is substantially different for the  $l$  and  $h$  extrapolation procedure. We also show the  $0.9R_{Nl}$  data which give a higher slope and an increased extrapolated value of  $T_c$ . The corresponding  $0.9R_{Nh}$

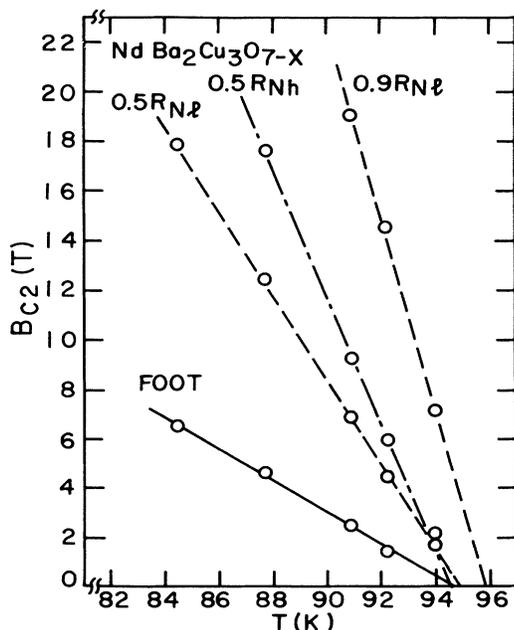


FIG. 2. Extrapolated data for upper critical field  $B_{c2}$  vs  $T$  for  $\text{NdBa}_2\text{Cu}_3\text{O}_{7-y}$  using the midpoint for the  $h$  and  $l$  extrapolation  $0.5R_{Nh}$  and  $0.5R_{Nl}$  and for  $0.9R_{Nl}$ .

not plotted because it is difficult to define [and would result in very high values of  $(dB_{c2}/dT)_{T=T_c}$  and  $T_c$ ]. The question here is whether the upper portion of the resistive transition reflects a small superconducting fraction well above the rapid drop in resistance. The change in resistance of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$  was less than 1% at 116 K for fields up to 20 T, suggesting that at this temperature any superconducting contribution is quite small.

Another characteristic feature of the broad resistive transition is illustrated in Fig. 2. A knee is observed near the bottom of the transition. The field at which the knee occurs, where the resistance starts to increase rapidly (labeled FOOT), also shows a linear variation with  $T$  (about 0.7 T/K). The combination of the various linear dependences shows that there is a large broadening of the transition as a function of field and that all the various features have approximately linear variations with  $T$  near  $T_c$ . Thus there is a range of criteria which can be used to discuss these materials, and extremely high fields are needed to examine the full transition below  $T_c$ . For this paper we choose to use the  $h$  extrapolation and the midpoint data defined as  $0.5R_{Nh}$ .

A comparison of  $T_c$ ,  $(dB_{c2}/dT)_{T=T_c}$ , and  $B_{c2}(0)$  is shown in Fig. 3 for the rare-earth series. Here  $B_{c2}(0)$  is an extrapolation to zero temperature assuming the  $0.5R_{Nh}$  extrapolation and the standard WHH theory with no Pauli-spin pair breaking.  $T_c$  is the value obtained by extrapolating  $0.5R_{Nh}$  vs  $B_0$  to  $B_0=0$ . As shown in Fig. 2, this value of  $T_c$  is quite close ( $\leq 0.5$  K) to that obtained by extrapolation of  $0.5R_{Nl}$ . Figure 3 shows that the compounds  $R\text{Ba}_2\text{Cu}_3\text{O}_{7-x}$  are characterized by a nearly constant  $T_c=93\pm 2$  K and a nearly constant  $(dB_{c2}/dT)_{T=T_c}\approx 2.8\pm 0.2$  T/K. Consequently the  $B_{c2}(T)$  data, along with the previously reported  $T_c$ , susceptibility, and oxygen-doping<sup>2</sup> data suggest that the magnetic rare-earth

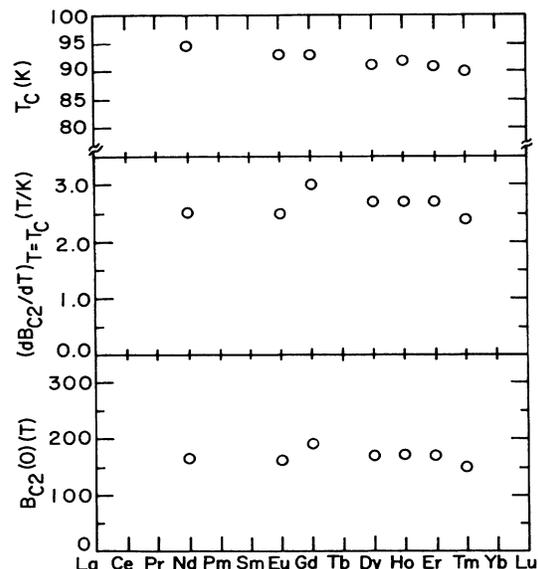


FIG. 3. Comparison of results for  $R\text{Ba}_2\text{Cu}_3\text{O}_{7-y}$  specimens vs  $R$ . Top: critical temperature  $T_c$ ; center:  $(dB_{c2}/dT)_{T=T_c}$  for  $0.5R_{Nh}$ ; bottom: calculated  $B_{c2}(0)$  based on data above.

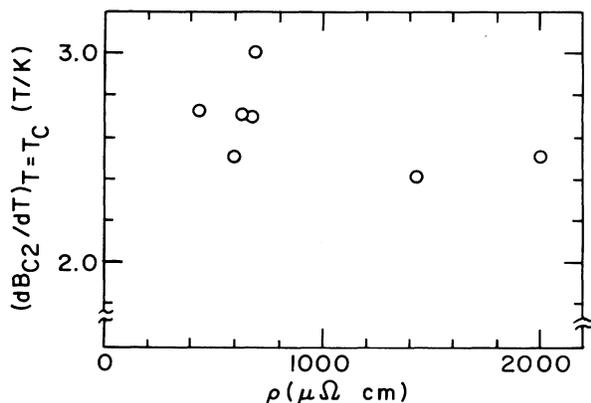


FIG. 4. Midpoint slope  $(dB_{c2}/dT)_{T=T_c}$  vs resistivity  $\rho$  for different  $RBa_2Cu_3O_{7-y}$  specimens.

ions do not interact with the conduction electrons in the compounds  $RBa_2Cu_3O_{7-x}$ . It has been suggested that the insensitivity of superconducting properties to the presence of magnetic rare-earth ions is a result of the large distance between the magnetic ions and the superconducting electrons, which is possible if the electrons in Cu-O chains are responsible for the superconductivity. The systematic small decrease of  $T_c$  versus rare-earth atom correlates with the lattice parameter change.<sup>2</sup>

Despite the insensitivity of  $B_{c2}(0)$  and  $T_c$  to the rare-earth ion in the  $RBa_2Cu_3O_{7-y}$  series, there is a large variation in the resistivity as indicated in Ref. 2. Figure 4 shows a plot of  $(dB_{c2}/dT)_{T=T_c}$  vs  $\rho$  at  $T_c$ . There is a cluster of the data near 400 to 600  $\mu\Omega$  cm, but the Sm and Nd compound have  $\rho$  values of  $\sim 1400$  and  $2000$   $\mu\Omega$  cm. Using 200  $\mu\Omega$  cm as an upper limit of resistivity for the  $YBa_2Cu_3O_{7-y}$  would result in a correspondingly higher value of  $\gamma$  than that in Ref. 1. (The high resistivity material,  $R=Nd$ , has a second phase present which may have contributed to the very high  $\rho$ .) Examination of Fig. 4 indicates that  $\rho$  is not directly correlated with magnetic scattering.

From the standard theory for  $B_{c2}(T)$  we would expect that the slope would increase linearly with increasing resistivity if all other parameters which affect the superconductivity (such as  $T_c$ , the density of states, the electron carrier density, etc.) remained constant throughout the series  $RBa_2Cu_3O_{7-x}$ . In the dirty limit, the change in resistivity from 200 to 2000  $\mu\Omega$  cm would cause the slope to increase tenfold. However, if we demand that the change from 200 to 2000  $\mu\Omega$  cm in the resistivity is to have less than a 10% effect on the slope (as is observed), then the material would have to have an electron carrier density of less than  $10^{19}$   $cm^{-3}$ , a value which is two orders of magnitude too small. Alternatively, for the slope to remain nearly constant, the material would have to be sufficiently in the clean limit. Consequently, the slope is not straightforwardly related to the measured resistivity.

One possibility is that the resistivity is dominated by the intergranular conduction, whereas the superconductivity properties are determined by the intragranular resistivity. Recent studies suggest that the relatively small critical current density in polycrystalline Sr-Ba-Cu-O (Refs. 3 and 4) and Y-Ba-Cu-O (Ref. 5) may result from such intergranular conduction. Based on our data, we suggest that the upper limit of  $\rho$  in  $RBa_2Cu_3O_{7-y}$  is 200  $\mu\Omega$  cm. Single-crystal data would provide a definitive measure of the intrinsic  $\rho$ .

We have also examined single-phase  $YBa_2Cu_3O_{7-x}$  material and find  $(dB_{c2}/dT)_{T=T_c} = 2.9$  T/K for the  $h$  midpoint,  $T_c = 91.4$  K, and the extrapolated  $B_{c2}(0) = 180$  T. The  $0.9R_{Nh}$  values yield  $(dB_{c2}/dT)_{T=T_c} = 5.3$  T/K,  $T_c = 93.1$  K, and  $B_{c2}(0) = 340$  T. These values are higher than presented earlier by us for a multiphase specimen,<sup>1</sup> mainly because we are using the  $h$  criterion. If we use the range  $(dB_{c2}/dT)_{T=T_c}$  from 2.2 to 6 T we obtain  $B_{c2}(0)$  from 140 to 380 T. In an attempt to outline the possible range of the anisotropy of  $B_{c2}$  we examined the onset of resistance at 4.2 K and high field. The assumption is that the crystallites with layers parallel to  $B_0$  establish the upper limit of  $B_{c2||}$  and that those crystallites with the layer planes perpendicular to  $B_0$  will become normal (and resistive) at a lower field  $B_{c2\perp}$ . The onset of resistance gives a lower bound estimate of  $B_{c2\perp}$  because other mechanisms can also lead to an early onset of the resistive state. We found zero resistance ( $< 5 \times 10^{-2}$   $\mu\Omega$  cm at  $< 0.1$ - $\mu$ V level) up to 28 T. Using  $B_{c2\perp} = 28$  T for the lowest limit we conclude that the anisotropy  $A = B_{c2||}/B_{c2\perp}$  is less than  $\frac{340}{28} \approx 12$ . If we choose to argue that  $B_{c2||} = 2[B_{c2||}(\text{midpoint}) - B_{c2\perp}]/B_{c2\perp}$  then  $A \leq 2[140 - 28]/28 \approx 8$ . Several of the  $RBa_2Cu_3O_{7-y}$  specimens were also examined at 4.2 K and all showed zero resistance up to 20 T setting a conservative upper limit of  $A < \frac{340}{20} = 17$  for this class. Data of Ekin *et al.*<sup>6</sup> gave zero values of resistance for  $YBa_2Cu_3O_{7-y}$  up to  $\approx 4$  T at 77 K. If this is used as a lower limit of  $B_{c2}$  for our data then  $A < 13$ , using  $B_{c2||}$  at  $0.9R_{Nh}$ . We should note that the anisotropy of  $B_{c2}$  in a single crystal on  $Nb_3Sn$  and  $V_3Si$  (Ref. 7) and the layered  $NbSe_2$  (Ref. 8) is independent of temperature. The present estimates suggest that this may also be the case for  $YBa_2Cu_3O_{7-y}$  and that the anisotropy in  $B_{c2}$  is  $\leq 13$  over the range 77 to 4.2 K. Recently, Santhanam *et al.* reported<sup>9</sup> an anisotropy of 30 in the critical current density  $J_c$  of single-crystal  $YBa_2Cu_3O_{7-y}$  which is above our present estimates of the  $B_{c2}$  anisotropy. Although  $J_c$  is affected by  $B_{c2}$ , other materials properties contribute to the pinning mechanisms in addition to the  $B_{c2}$ .

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