Resistive transitions under applied fields in oriented thin films of $Y_{0.6}Pr_{0.4}Ba_2Cu_3O_7$: Evidence for conventional three-dimensional behavior

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We report a detailed study of the resistive transitions in oriented films of $Y_{0.6}Pr_{0.4}Ba_2Cu_3O_7$ (PYBCO) under applied fields. Our analysis shows a more conventional three-dimensional behavior, with no giant flux-creep effects, and a considerable increase in the coherence length as compared to that of $Y_1Ba_2Cu_3O_7$ (YBCO), both in the in-plane and out-of-plane directions. This large increase cannot be explained by just the magnetic pair breaking supposedly responsible for the decrease in the critical temperature. It is proposed that the extended $4f$ wave functions of the Pr atoms lead to a strong coupling between CuO plane, thus removing the van Hove singularity responsible for the anomalously short coherence length in YBCO.

Six years after the discovery of the high-temperature superconductors by Bednorz and Müller, these materials are far from being understood. The unusual broadening of the resistive transition in the presence of a magnetic field led to the proposal of a new magnetic phase of the cuprates, namely the glassy state,[1] separated from the Abrikosov phase by a $H^*(T)$ line, related to the previously identified irreversibility line.[2,3] The extremely short coherence length ξ_c in the c direction, which suggests that the layered superconductors are two-dimensional in nature (ξ_c shorter than the lattice parameter c), is thought to be at the origin of this behavior.[4]

An elegant probe of the nature of the cuprate superconductors is the substitution of the Y atom by other members of the Lanthanides.[5] Among these atoms, only Ce (Ref. 6) an Pr (Refs. 7–10) alloys are not superconducting at concentration $x=1$, with $x_{cr}\cong 0.5$, although they are isomorphic to all others and with similar magnetic properties [all the other alloys containing rare-earth atoms have the same transition temperature as $Y_1Ba_2Cu_3O_7$ (YBCO)]. Numerous experiments have been made on the YPrBaCuO system, with different Pr concentrations, which include spectroscopic,[11] magnetic moment, thermal properties and structural analysis,[6] Hall effect,[9] magnetic penetration depth,[12] pressure dependence of T_c,[13] and magnetic[10] and resistive[14,15] measurements of the upper critical field. Yet, the mechanism that reduces T_c in this system is unclear, with the majority of the experiments interpreted as indicating magnetic pair breaking due to the Pr magnetic moment, or hole filling (or localization) mechanism due to the larger valence of Pr compared to Y. In the case of the upper critical field, the directional resistive measurements reported by Antogonazza *et al.*[14] have not been related to the coherence lengths ξ_{ab} and ξ_c in the $Y_{1-x}Pr_xBa_2Cu_3O_7$ system, and hence their values and the origin of the change as compared to YBCO have not been discussed. The data reported on Ref. 10 were taken on polycrystalline samples of $Y_{1-x}Pr_xBa_2Cu_3O_7$ with various x values and show an unusual bell-shaped $H(T)$ phase diagram. The work of Jia *et al.*,[15] which measured the resistive transitions of $Y_{1-x}Pr_xBa_2Cu_3O_7$ single crystals at various x values shows no sign of the unusual bell-shaped H-T phase diagram reported in Ref. 10. The authors of this paper focus their attention and interpretation on the question of whether the mechanism responsible for the suppression of superconductivity in the Pr-doped YBCO is hole filling or pair breaking, and reach the conclusion that the data is not consistent with a pair-breaking picture but rather with some change in the concentration of free holes. Another result reported in their work, which however they do not discuss, is *the pronounced increase in both* ξ_c *and* ξ_{ab} with increasing Pr concentration, particularly at $x\geq 0.4$.

In this paper, we report detailed measurements of the resistive transitions of oriented thin films of $Y_{0.6}Pr_{0.4}Ba_2Cu_3O_7$ (PYBCO) for $\mathbf{H}\|ab$ and $\mathbf{H}\|c$, for fields up to 7 T. We find that the broadening of the transition in the case of the field parallel to the c axis is much smaller than in YBCO, giving relatively close values of $\xi_{ab}\cong 55$ Å irrespective of the exact criterion used for the determination of H_{c2}, as for conventional three-dimensional low-temperature superconductors. The resistive transition with the field applied parallel to the ab planes is wider, but still less than the one measured for YBCO, with a lower bound for ξ_c that is three times larger than in the case of YBCO, 12 Å $\geq\xi_c\geq$ 6 Å. A detailed analysis of the transitions show that the giant flux-creep model proposed by Yeshurun and Malozemoff[3] and further developed by Tinkham,[16] does not apply to this alloy, which rather shows a conventional three-dimensional behavior.

The samples were grown *in situ* by off-axis rf magnetron sputtering on MgO substrates held at $T\cong 700$ °C, using a 2″ target of stoichiometric $Y_{0.6}Pr_{0.4}Ba_2Cu_3O_7$ in a method reported by Eom *et al.*[17] The films were c-axis oriented, with $c\cong 11.7$ Å, as indicated from x-ray-diffraction patterns. The resistive transitions in the presence of the field were measured with the standard four-probe method on unpatterned films, with the measuring current always perpendicular to the field, in fields up to 7 T. The temperature was measured with a calibrated

carbon-glass resistor. The data in the presence of the field was taken, while slowly heating the zero-field-cooled samples, at a rate of approximately 10 K h^{-1}, the samples being in good thermal contact to the sample holder block, in a He gas environment.

The resistive transition in zero field has $T_c^{\text{onset}} \cong 40$ K and a transition width of about 4 K, with a slight increase in the resistance for $T \leq 100$ K, characteristic for the samples of YPrBaCuO with large Pr concentrations.[8–10,15] The estimated value of the resistivity at room temperature is about 700 $\mu\Omega$ cm, smaller by a factor of about 2 than values reported on ceramics.[8–10,18] The low values of the resistivity are a good indication of the high quality of our samples.

Figure 1(a) shows the R-T curves with $\mathbf{H} \| c$, from $\mathbf{H}=0$ T to $\mathbf{H}=7$ T, from which it is evident that the shape of the transition in the field is qualitatively different from that for the same field orientation in YBCO. The transitions are narrower, and closer to conventional superconductors. We believe that this is an evidence of the different dimensionality of the $Y_{0.6}Pr_{0.4}Ba_2Cu_3O_7$ system, as will be discussed later. Fig. 1(b) gives the value of H_{c2} as a function of the temperature for three different choices of the resistance value (at $R/R_n = 0.12$, 0.5, 0.9). The symbols are the measured points, and solid lines are the linear fits to the data, with $(dH_{c2}/dT)|_{T_c} = -0.54$ TK^{-1} for $R/R_n = 0.5$ and -0.91 TK^{-1} for $R/R_n = 0.9$. These slopes, using the Werthamer, Helfand, and Hohenberg (WHH) formula

$$H_{c2}(0) = 0.7(dH_{c2}/dT)|_{T_c} T_c$$

result in $H_{c2}(0) = 13.7$ and 24.7 T, respectively. Putting these values into

$$H_{c2}(0) = \Phi_0/(2\pi(0.74)^2 \xi_{ab(0)}^2)$$

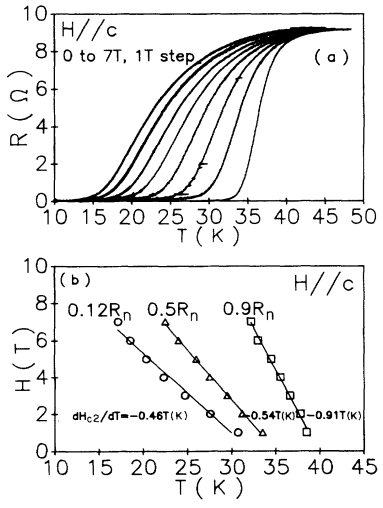

FIG. 1. (a) R-T curves in the presence of $\mathbf{H} \| c$. Note the small broadening in the resistive transition. The large shift in the low-field regime is probably due to some sample inhomogeneity, expressed in the large zero-field transition width, (b) The temperature dependence of the upper critical field for $\mathbf{H} \| c$ for three different R/R_n criteria. The points are data taken from (a) and the solid lines are the linear fits.

gives $\xi_{ab(0)} = 64$ and 48 Å for the two different choices. These two values differ only by 25%, while for YBCO the different choices result in about a factor of two in the ξ's, as reported in early measurements[19] and as recently measured by us.[20] This result, which is a reflection of the different shape of the resistive transition indicates that the choice of the R/R_n criterion is not very important, as for low T_c three-dimensional superconductors. The values for dH_{c2}/dT at $R/R_n = 0.5$ are consistent with slopes deduced from previous measurements,[10,14,15] and with the value of ξ_{ab} estimated from an all high-temperature superconductors (HTSC) proximity experiment recently reported by Polturak *et al.*[21] This agreement between the different $H_{c2}(T)$ measurements (on polycrystalline, single crystals and oriented films) and between the proximity effect experiment is very important, and will be discussed later.

The data taken for $\mathbf{H} \| ab$ planes is shown in Fig. 2(a). For this field orientation the R-T curves look more like those of YBCO, but with a smaller broadening. The field temperature dependence near T_c for three different criteria, $R/R_n = 0.12$, 0.5, and 0.8 are shown in Fig. 2(b), with slopes of -1.8 T/K, -2.91 T/K, and -6.9 T/K, respectively. Using again the WHH formula for the slopes extracted from $R/R = 0.5$ and 0.8, and putting the values of the zero-temperature critical fields in the three-dimensional anisotropic Ginzburg-Landau (G-L) formula

$$H_{c2}(0) = \Phi_0/(2\pi(0.74)^2 \xi_{ab(0)} \xi_{c(0)})$$

one gets 12 Å $\geq \xi_{c(0)} \geq 6$ Å, with the upper bound being essentially the c parameter of PYBCO. Here again, our result is very close to the one reported by Jia *et al.*[15] This result, combined with the estimated value for $\xi_{ab(0)}$ results in an anisotropy factor of 5–8, very close to the anisotropy ratio for YBCO and to other reported values.[15]

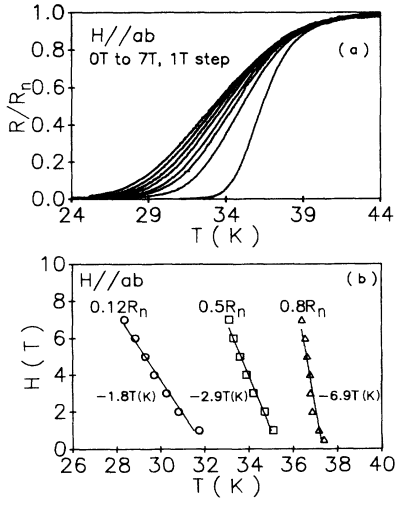

FIG. 2. (a) R-T curves in the presence of $\mathbf{H} \| ab$. The large shift in the low-field regime is probably due to some sample inhomogeneity, expressed in the large zero-field transition width, (b) The temperature dependence of the upper critical field for $\mathbf{H} \| ab$ for three different R/R_n criteria. The points are data taken from (a) and the solid lines are the linear fits.

Neither our in-plane or out-of-plane R-T curves fit to Tinkham's model[16] for the resistive transitions in applied magnetic field for HTSC. Our R-T curves cannot be fitted to

$$R/R_n=[I_0(\gamma_0/2)]^{-2}\ , \tag{1}$$

where I_0 is the tabulated modified Bessel function and

$$\gamma_0 \propto \frac{J_{c0}(1-t)^{3/2}}{T_c H} \quad \left[t \equiv \frac{T}{T_c} \right]$$

is the normalized barrier height for thermally activated flux motion, and J_{c0} is the pair-breaking current at $T=0$ and $H=0$. The fit for $\mathbf{H}\|c$ for various field values is shown in Fig. 3. While a fit is possible at low fields, the high-field data cannot by fitted by Eq. (1) (even when modifying the—in principle not adjustable—parameter J_{c0}). A similar disagreement is found for $\mathbf{H}\|ab$. We also checked the scaling consequences of Tinkham's theory: The resistively measured critical field H_{c2}^* should scale as $H_{c2}^* \propto (1-t)^{1.5}$ and that the width of the transition at any two arbitrary R/R_n levels should scale as $\Delta T \propto H^{2/3}$. For the field applied parallel to the c axis, the best fit for all the field range, no matter what resistance criterion used, is nearly linear $H_{c2}^* \propto (1-t)$, [Fig. 1(b)], and for ΔT taken between $R/R_n=0.9$ and 0.1 and between $R/R_n=0.9$ and 0.5 we get $\Delta T \propto H^{0.32}$. For the ab direction there is some curvature for low fields ($H<2$ T), which disappears for higher fields [Fig. 2(b)]. When a fit is done for all the field range, the best fit is $H_{c2} \propto (1-t)^{\alpha}$, where $\alpha \cong 2$. We do not believe, however, that this exponent reflects a fundamental property of the material, but rather its inhomogeneities (for $\mathbf{H}\|ab$ in our applied field range, the shift of T_c is smaller than the width of the transition at zero field). Taking ΔT for R/R_n between 0.8 to 0.1 and R/R_n between 0.5 and 0.1 results in $\Delta T \propto H^{0.2}$. Qualitatively these results mean that the broadening of the resistive transition in applied fields is much less than that predicted by Tinkham's theory. This disagreement between our data and the theory of Tinkham supports our identification of the H_{c2}^* as obtained from the resistive measurements with the thermodynamic H_{c2}, or at least close to it. Furthermore, the lack of any significant difference between the values of dH_{c2}/dT for single crystals and thin films may also suggest that in this system the role of flux flow is much less important, and hence, self consistently the relevance of the giant flux-creep and flux lattice melting models for this alloy should be questioned.

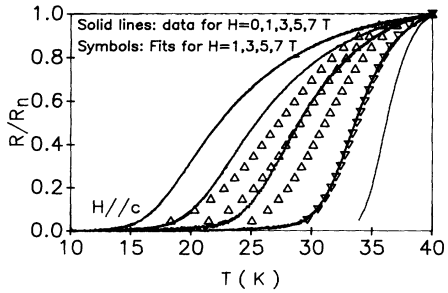

FIG. 3. Experimental vs fitted curves for $H=0,1,3,5,7$ T applied parallel to the c axis. At each level of R/R_n, the theoretical downshift (T_c-T) as predicted by Eq. (1) is plotted relative to the experimental $R(T)$ curve for $H=0$. The data for $H=1$ T could nicely be fitted with $J_{c0}(0)=2\times10^5$ A/cm^2 and $T_c=40$ K. The same parameters were kept for the fits to the upper field values, but now with total disagreement between the data and the fit.

We now discuss the origin of the depression of T_c. As previously proposed by other groups, the reduction of T_c with increasing Pr concentration can be understood in terms of magnetic pair breaking (PB), as given by

$$\ln(T_c/T_{c0})=\Psi(1/2)-\Psi(1/2+\hbar/4\pi\tau_i k_B T_c)\ . \tag{2}$$

Here T_{c0} is the transition temperature in the pure material, T_c is the transition temperature with the magnetic impurities, τ_i is the interaction characteristic time and Ψ is the tabulated di-gamma function. This analysis led to a good fit to the measured $T_c(x)$, with $X_{cr} \cong 0.6$ (i.e., full destruction of superconductivity).[8,10] Since Eq. (2) is an exact solution for T_c for the entire temperature range (i.e., not only in the G-L regime), one can try to use the same approach for the PYBCO film in applied fields by replacing $1/\tau_i$ with

$$1/\tau=1/\tau_i+1/\tau_H\ , \tag{3}$$

where τ_H is the characteristic pair-breaking time due to the applied field. Putting into Eq. (2) the expression for τ from Eq. (3) with a fixed value for τ_i, and letting the value of τ_H vary with the field as

$$1/\tau_H \propto H_{c2}D\ , \tag{4}$$

where D is the electrons diffusion constant (assumed to be equal for PYBCO and YBCO), we can estimate the ratio between the dH_{c2}/dT of pure YBCO (Refs. 19 and 20) and PYBCO. The result of this analysis is that the expected ratio is about 2/3, while the measured ratio is of the order of 1/3 to 1/4, in agreement with the result reported by Jia *et al.*[15] Correction due to the probably smaller diffusion constant of PYBCO will increase the discrepancy with the measured dH_{c2}/dT values. We believe that this analysis gives an indication that the mechanism responsible for the reduction of T_c is not PB as a sole mechanism, but we cannot exclude the hole-filling explanation, or a combination of both hole filling and PB as suggested by Neumeier *et al.*[22]

A major point in all cuprate high-temperature superconductors (HTSC) is the presence of the Cu-O planes, in which the conduction takes place. This makes these systems two-dimensional in nature, as also indicated by their extremely short coherence length in the c direction. As shown by van Hove, the two-dimensional structure imposes the presence of singularities in the density of states (DOS). If the Fermi level falls at or near the singularity in the DOS, the Fermi velocity vanishes at the singular points of the Fermi surface. Then, the *averaged* Fermi velocity over the Fermi surface taken a much lower value due to the weight of the singularity (high DOS). From the relation $\xi=\hbar\mathbf{V}_F/\pi\Delta$, it is clear that this results in a reduced ξ when $\mathbf{V}_F$ is properly averaged.[23] Indeed, the

value of $\mathbf{V}_F$ in YBCO as determined from H_{c2} (Ref. 24) (which is dependent on the averaged $\mathbf{V}_F$) is roughly a factor of 5 smaller than the directionally measured value of $\mathbf{V}_F \geq 7^{*}10^{7}$ cm*sec^{-1}.[25]

In contrast, for $Y_{0.6}Pr_{0.4}Ba_2Cu_3O_7$ the ξ_{ab} as determined from H_{c2} agrees with the value of ξ_{ab} derived from (directional) proximity effect measurements in $Y_1Ba_2Cu_3O_7/Y_{0.6}Pr_{0.4}Ba_2Cu_3O_7/Y_1Ba_2Cu_3O_7$ step edge junctions reported by Polturak *et al.*[21] In the proximity effect experiments the current flows in the *ab* planes across the junctions, which means that the deduced value of ξ_{ab} reflects the Fermi velocity in the direction of the current flow (i.e., in a *specific direction in* **k** *space*), while the $H_{c2}(T)\|c$ measurements, give a value for ξ that reflects the averaged fermi velocity over *all values of* **k** around the Fermi surface. The agreement between the two types of determination of ξ_{ab} in $Y_{0.6}Pr_{0.4}Ba_2Cu_3O_7$ suggests a removal of the two-dimensional singularity in this alloy. This is borne out by the increased value of ξ_c, which approaches c, by the agreement between the resistive and magnetic measurements of H_{c2}, and by the linear temperature dependence of H_{c2}. We suggest that the origin of the more three-dimensional-like behavior of $Y_{0.6}Pr_{0.4}Ba_2Cu_3O_7$ resides in the spatially extended $4f$ wave function of the Pr atom as compared to most other rare earths.

An excellent test for dimensionality effects are high-pressure measurements of T_c and of H_{c2}. It is reasonable to assume that, at high pressures the dominant effect will be an increase of the overlap between the wave functions of adjacent planes. In the case of the 248 system ($YBa_2Cu_4O_8$), a recent work by Scholtz *et al.*[26] on the pressure dependence of T_c and H_{c2} shows an initially positive slope of $T_c(P)$ with saturation followed by a decrease at higher pressures, with a striking decrease in H_{c2} and in dH_{c2}/dT (where the criterion used was the onset of the resistance, which means that there is a clear shift in the curves). This is consistent with the suggestion that the short coherence lengths measured in the undoped cuprates reflects their two-dimensional character. A more three-dimensional character can be induced by appropriate doping and/or high pressure, resulting in much increased coherence lengths.

In conclusion, we have reported a detailed study and analysis of resistive transitions under applied fields in *c*-axis oriented thin films of $Y_{0.6}Pr_{0.4}Ba_2Cu_3O_7$, yielding 64 Å $\geq \xi_{ab} \geq$ 48 Å and 12 Å $\geq \xi_c \geq$ 6 Å. These values, as well as the detailed shape of the resistive transitions, point out to the three-dimensional conventional behavior of this alloy, in contrast with YBCO. Our suggested interpretation is the removal of the van Hove singularity present in YBCO by the Pr impurities.

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