Magnetoresistance of Untwinned YBa₂Cu₃O_y Single Crystals in a Wide Range of Doping: Anomalous Hole-Doping Dependence of the Coherence Length

Yoichi Ando and Kouji Segawa

Central Research Institute of Electric Power Industry, Komae, Tokyo 201-8511, Japan (Received 2 July 2001; published 8 April 2002)

Magnetoresistance (MR) in the *a*-axis resistivity of untwinned YBa₂Cu₃O_y single crystals is measured for a wide range of doping (y = 6.45-7.0). The y dependence of the in-plane coherence length ξ_{ab} estimated from the fluctuation magnetoconductance indicates that the superconductivity is anomalously weakened in the 60-K phase; this observation, together with the Hall coefficient and the *a*-axis thermopower data which suggest the hole doping to be 12% for $y \approx 6.65$, gives evidence that the origin of the 60-K plateau is the 1/8 anomaly. At high temperatures, the normal-state MR data show signatures of the Zeeman effect on the pseudogap in underdoped samples.

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Magnetoresistance (MR) is a useful tool to study the electron transport in metals, though its origin can be complicated. One can infer, for example, the significance of the spin-dependent mechanisms through the longitudinal MR, and the fluctuation magnetoconductivity (FMC) observed above T_c of a superconductor can be used to estimate the coherence length. In high- T_c cuprates, the normal-state orbital MR is known to violate the Kohler's rule and to show an unusual temperature dependence, $\sim (aT^2 + b)^{-2}$ [1], which indicates that an unusual situation for the charge transport, possibly a scattering-time separation [2,3], is realized. Also, it has been discussed that a sizable FMC survives to much above T_c in cuprates [1,4], even twice as high as T_c in optimally doped $La_{2-x}Sr_xCuO_4$ (LSCO) [1]. Such unusual MR behavior naturally calls for detailed doping-dependence studies of the MR to better understand the charge transport in cuprates; however, there have been only a few reports on the hole-doping dependence of the MR behavior [1,4-6], and the key issues such as the evolution of the role of spins or the evolution of the fluctuation contributions are not really understood yet.

In this Letter, we report the MR measurements of untwinned YBa₂Cu₃O_v (YBCO) single crystals in a wide range of doping, from heavily underdoped ($T_c = 20 \text{ K}$) to slightly overdoped regions. Since YBCO contains Cu-O chains which can carry the electric current along the b axis, we measure the magnetic-field dependence of the *a*-axis resistivity ρ_a and pay particular attention to sorting out the genuine MR behavior of the CuO₂ planes with a careful analysis involving also the *b*-axis resistivity ρ_b and the Hall resistivity $\rho_{\rm H}$. Based on the magnetoconductivity of the CuO₂ planes obtained after the analysis, we discuss both the normal-state magnetoconductivity at high temperatures and the FMC at lower temperatures. Most notably, the FMC is found to show a nonmonotonic evolution with y, and its y dependence suggests that the superconductivity is anomalously weakened in the "60-K phase" [7] of YBCO; in combination with other in-plane transport properties which suggest that the hole doping is actually 1/8 at $y \approx 6.65$, the present data give evidence for the 1/8-anomaly origin of the 60-K plateau.

The YBCO single crystals are grown in Y₂O₃ crucibles by a conventional flux method [8], and are carefully detwinned and annealed (see Ref. [7] for details); the crystals reported here are in the range of y = 6.45-7.0 (the error in y is ± 0.02). The MR measurements are done using an ac four-probe method under sweeping magnetic fields of ± 10 or ± 14 T. Both the transverse (magnetic field *H* is perpendicular to the *ab* planes) and the longitudinal (*H* is parallel to the current *I*) MR are measured. The temperature is stabilized within ~ 1 mK during the MR measurements by a home-built temperature regulation system employing both the Cernox resistance sensor and a capacitance sensor. Other details of the measurements of ρ_a , ρ_b , and ρ_H are described in Ref. [7].

Figure 1(a) shows the $\rho_a(T)$ data for the series of our untwinned crystals. An unusual overlapping of the $\rho_a(T)$ data for the "60-K phase" samples, which is clear in Fig. 1(a), has been discussed in Ref. [7]. Figure 1(b) shows examples of the raw MR ($\Delta \rho_a$) data for y = 7.0. All the data plotted in Fig. 1(b) show the ordinary H^2 dependence, which becomes clear in the plot of $\Delta \rho_a$ vs H^2 [Fig. 1(c)]. In the underdoped samples, however, we observed that the MR starts to become concave downward (or tends to saturate) in the $\Delta \rho_a$ vs H^2 plot upon approaching T_c , as shown in Fig. 1(d) for y = 6.50 $(T_c = 35 \text{ K})$. Such H dependence is most likely caused by a decrease in the characteristic magnetic-field scale to suppress superconducting fluctuations, and some vortex fluctuations in the pseudogap state [9,10] are possibly involved in this anomaly. In any case, when this anomalous H dependence is observed near T_c , the MR data cannot be discussed on the same ground as those at higher temperatures, and therefore we do not include the MR data for temperatures very close to T_c in the discussions afterwards. However, as long as the temperature is not very close to T_c , the low-field part of $\Delta \rho_a$ can reasonably be fitted with H^2 [as shown for the 50-K data in Fig. 1(d)],



FIG. 1. (a) $\rho_a(T)$ data for all the y values studied. (b) Raw data of the transverse MR for y = 7.0 at selected temperatures. (c) $\Delta \rho_a$ vs H^2 plots of the data in panel (b). (d) Transverse MR for y = 6.50 at selected temperatures; the dashed line is the linear fit to the low-field part of the data.

and for such data we determine the "magnitude" of MR from the low-field slope of the $\Delta \rho_a$ vs H^2 plot.

Figure 2 shows the summary of the transverse and longitudinal MR ($[\Delta \rho_a / \rho_a]_{\perp}$ and $[\Delta \rho_a / \rho_a]_{\parallel}$, respectively) for the whole doping range. In Fig. 2, one may notice that the longitudinal MR rapidly diminishes with increasing temperature in highly doped samples (y = 6.95 and 7.0), while it remains noticeable up to 270 K in the underdoped samples. While the low-temperature growth of the longitudinal MR is likely to originate from the Zeeman term in the FMC, its behavior in the high-temperature region is expected to reflect the properties of the normal state.



FIG. 2. T-dependences of the transverse (•) and longitudinal (•) MR for most of the hole concentrations studied.

Figure 3 depicts the y dependence of $[\Delta \rho_a / \rho_a]_{\parallel}$ at high temperatures, where it is clear that there is a crossover near y = 6.8 above which the longitudinal MR is diminished.

Since the longitudinal MR in cuprates mostly measures the Zeeman effect on the spin terms, the observed crossover in $[\Delta \rho_a / \rho_a]_{\parallel}$ near y = 6.8 is likely to be a manifestation of the Zeeman effect on the pseudogap which opens in the normal state of underdoped samples. Remember that the pseudogap is accompanied by a spin gap [11], which reduces the magnetic scattering rate; since the magnetic scatterings are recovered when the spin gap is suppressed by the Zeeman effect, a measurable longitudinal MR is actually expected in the pseudogap state.

From the data of $[\Delta \rho_a / \rho_a]_{\perp}$ and $[\Delta \rho_a / \rho_a]_{\parallel}$, we can deduce the orbital magnetoconductivity of the CuO2 planes, although the correct procedure is not simple for an anisotropic system. For an isotropic system, the MR (up to terms in B^2) is written as $\Delta \rho / \rho = -\Delta \sigma / \sigma - (\sigma_{xy} / \sigma)^2$, where σ is the conductivity and σ_{xy} is the Hall conductivity [1]. This formula can be generalized to an anisotropic system as $\Delta \rho_a / \rho_a = -\Delta \sigma_a / \sigma_a - \sigma_{xy}^2 / (\sigma_a \sigma_b)$; note here that in an anisotropic system σ_{xy} and $\rho_{\rm H}$ are related (up to terms in B) by $\sigma_{xy} = \rho_{\rm H} / (\rho_a \rho_b)$ [7]. Therefore, to obtain the correct magnetoconductivity from the MR data of an anisotropic system, one should use the formula $\Delta\sigma_a/\sigma_a = -\Delta\rho_a/\rho_a - \rho_{\rm H}^2/(\rho_a\rho_b)$. This means one needs to measure not only the *a*-axis transport but also ρ_b and $\rho_{\rm H}$. To the best of our knowledge, this correct formula has never been used for the analyses of the magnetoconductivity in cuprates, even though $\Delta \rho_a / \rho_a$ and $\rho_{\rm H}^2/(\rho_a \rho_b)$ can become the same order at high temperatures. To obtain the orbital magnetoconductivity for our samples, we calculate both the transverse and the longitudinal magnetoconductivity by using the correct formula and take the difference. The data of ρ_b and ρ_H used in the analysis are shown elsewhere [7,12].

Figures 4(a)-4(c) show examples of the orbital magnetoconductivity data thus obtained. In the analysis of these data, we follow the recent trend [1,4,13] to interpret the



FIG. 3. *y* dependence of the longitudinal MR at high temperatures where fluctuation contribution is negligible.



FIG. 4. *T* dependences of the orbital magnetoconductivity for (a) y = 6.70, (b) 6.60, and (c) 6.50. The solid lines are the fits of the high-temperature region to the normal-state contribution $(aT^2 + b)^{-2}$. The parameters *a* and *b* are shown in panel (d). The dashed lines in (a)–(c) are the fits of the data to the additional Aslamazov-Larkin orbital term.

high-temperature part to come from the normal-state contribution [which is expressed as $(aT^2 + b)^{-2}$] rather than the Maki-Thompson orbital FMC [14]. The solid lines in Figs. 4(a)-4(c) are the fits of the data to the normal-state contribution. The data at high temperatures are actually very well fitted with the $(aT^2 + b)^{-2}$ dependence. The offset b is zero for y of down to 6.60, but becomes nonzero for smaller y, which corresponds well to the behavior of the $\rho_a(T)$ data where a noticeable residual resistivity becomes apparent for y < 6.60 [Fig. 1(a)]; this seems to give additional support to the validity of this analysis. The y dependence of the fitting parameters are summarized in Fig. 4(d). We note that the magnetoresistance is not as well fitted with $(aT^2 + b)^{-2}$ as the magnetoconductance, which implies the necessity of the correct calculation for $\Delta \sigma_a / \sigma_a$.

We can estimate the FMC by subtracting the normalstate contribution (determined from the high-temperature data) from the total magnetoconductivity. Figure 5(a) depicts the evolution of the FMC in the $(T - T_c)/T_c$ vs y plane using a color scale. It becomes clear in Fig. 5(a) that, in addition to the general growth of the FMC with decreasing y, there is an enhancement in the FMC near y = 6.7, which corresponds to the 60-K phase of YBCO; this means that the superconducting fluctuations are more easily suppressed with magnetic field, and thus the characteristic field scale for superconductivity is reduced near y = 6.7.

This situation can be more quantitatively analyzed by fitting the FMC data to the Aslamazov-Larkin orbital (ALO) term [15]; the dashed lines in Figs. 4(a)-4(c) are the fits of the lower-temperature data to the normal-state contribution plus the ALO term. In determining the dashed lines, there are two fitting parameters, the in-plane and *c*-axis



FIG. 5 (color). (a) Evolution of the fluctuation magnetoconductivity in the $(T - T_c)/T_c$ vs y plane. (b) y dependence of ξ_{ab} obtained from the ALO fits. (c) Mean-field H_{c2} calculated from ξ_{ab} .

coherence lengths, ξ_{ab} and ξ_c . (T_c is set to the zeroresistance T_c .) Since the fits become insensitive to ξ_c for y < 6.9 where $\xi_c \leq 1$ Å, for underdoped samples we fix ξ_c to be near 1 Å and only change ξ_{ab} in the fits. Although not many data points are available for fitting for each composition, we can determine ξ_{ab} with roughly 20% error from the ALO fits.

Figure 5(b) shows the y dependence of ξ_{ab} obtained from the ALO fits. The general trend is that ξ_{ab} increases with decreasing hole doping, which is natural because the mean-field upper critical field at zero temperature, $H_{c2}^{\rm MF}$ [= $\Phi_0/(2\pi\xi_{ab}^2)$], is expected to be reduced as T_c goes down. However, for $y \simeq 6.7$ there is a marked anomaly that ξ_{ab} deviates upwardly from the general trend, which corresponds to a suppression of H_{c2}^{MF} in the 60-K phase [Fig. 5(c)]. This is in accord with the trend already apparent in Fig. 5(a). We note that this implication on the upper critical field is actually corroborated by the behavior of the resistive transition in 16 T, where the 60-K-phase samples show a marked broadening compared to other compositions, as demonstrated in Fig. 6(a). Therefore, both ξ_{ab} and the resistive transition appear to indicate that the superconductivity is anomalously weak to the applied magnetic field in the region near y = 6.7.

It is worthwhile to note that the present data are useful for elucidating the origin of the 60-K plateau. Remember that the 60-K plateau is just a *plateau* and is not a *dip* in T_c , and there has been no clear evidence that the superconductivity is *weakened* in the plateau; it should therefore be recognized that our result for H_{c2} gives evidence that the superconductivity is actually weakened in the plateau. Since the weakening of the superconductivity is the



FIG. 6. (a) $\rho_a(T)$ data for y = 6.45, 6.70, and 7.00 in zero field (thin lines) and in 16 T (thick lines). (b),(c) Plots of $R_H(T)e/V_0$ and $S_a(T)$, respectively, for 60-K YBCO and LSCO at x = 0.12.

fundamental feature of the 1/8 anomaly in the La-based cuprates, the present result clearly speaks for the 1/8-anomaly origin of the 60-K plateau. Furthermore, comparisons of the Hall coefficient R_H and *a*-axis thermopower S_a of 60-K YBCO to those of LSCO at x = 0.12strongly suggest that the hole doping is in fact 1/8 for $y \approx 6.65$; Figs. 6(b) and 6(c) show such comparisons in $R_H(T)e/V_0$ (V₀ is the volume per Cu in the plane) and $S_a(T)$, which demonstrate that LSCO at x = 0.12 and YBCO at $y \approx 6.65$ show quite similar values near room temperature, and these parameters have been proposed to be good indicators of the hole doping in cuprates [16,17]. Therefore, it appears that in the 60-K plateau the hole doping is actually $\sim 1/8$ and the superconductivity is weakened, which together mean that the 60-K plateau is a manifestation of the 1/8 anomaly. It is useful to note that a reasonably good case for the connection between the plateau and the 1/8 anomaly was made previously in a complementary way, using Ca doping [18].

It should, however, be noted that static charge stripes (a sort of charge density wave that localizes the carriers) do not appear to be the fundamental "cause" of the weakening of the superconductivity in YBCO, since there is no evidence for static stripes in YBCO. It is of course possible that the impact of the stripes at the 1/8 doping varies depending on the level of dynamics of the stripes in various cuprate systems. A more interesting possibility is that a proximity to a quantum critical point (QCP) [19] is the more fundamental cause of the 1/8 anomaly: Recently, Aeppli et al. reported [20] that the magnetic response of the Nd-free LSCO (where there is also no evidence for static charge stripes) bears a signature of the quantum criticality. In our data, fluctuations seem to be enhanced near y = 6.7 while the superconductivity is suppressed, which might also be an indication of a proximity to a QCP; if so, this particular QCP suppresses superconductivity through enhanced fluctuations, rather than creates superconductivity.

Last, we note the implication of the overall evolution of ξ_{ab} . In cuprates, the pseudogap causes the energy gap Δ to *grow* upon underdoping [11], and thus there is no proportionality between Δ and T_c . It is therefore not obvious whether ξ_{ab} should follow the behavior of Δ or that of T_c . This is a rather fundamental problem in the pseudogap physics [21], but experimental data have been lacking. The complicated y dependence of ξ_{ab} indicates that neither Δ nor T_c are solely decisive and that an elaborate theory is necessary to describe ξ_{ab} .

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