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Magnetotransport mechanisms in strongly underdoped $YBa_2Cu_3O_x$ single crystals

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We report magnetoresistivity measurements on strongly underdoped $YBa_2Cu_3O_x$ ($x=6.25$ and 6.36) single crystals in applied magnetic fields $H\parallel c$ axis. We identify two different contributions to both in-plane $\Delta\rho_{ab}/\rho_{ab}$ and out-of-plane $\Delta\rho_c/\rho_c$ magnetoresistivities. The first contribution has the same sign as the temperature coefficient of the resistivity $\partial\ln\rho_i/\partial T$ ($i=\{c,ab\}$). This contribution reflects the incoherent nature of the out-of-plane transport. The second contribution is positive, quadratic in field, with an onset temperature that correlates to the antiferromagnetic ordering.

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Investigation of magnetoresistance of layered cuprates with different levels of doping has revealed a number of effects that are difficult to reconcile with the properties of conventional metals. One striking feature of the magnetoresistivity (MR) tensor is the opposite signs of the in-plane and out-of-plane MR. Specifically, for a certain range of doping, and within an extensive temperature range, different types of cuprates exhibit the same phenomenon: the in-plane magnetoresistivity $\Delta\rho_{ab}/\rho_{ab}$ is positive, while the out-of-plane magnetoresistivity $\Delta\rho_c/\rho_c$ is negative (see, for example, Refs. 1–4). These opposite signs of MR seem to correlate with the contrasting temperature dependence of the respective resistivities, namely, metallic ($\partial\rho/\partial T>0$) in-plane resistivity ρ_{ab} and nonmetallic ($\partial\rho/\partial T<0$) out-of-plane resistivity ρ_c.[5]

Another important aspect of the physics of the cuprates is the interplay between the charge and spin subsystems located in the CuO_2 planes. One way to probe the charge-spin interaction across the phase diagram is through magnetoresistance measurements. The majority of the investigations were limited, however, to the optimally doped or moderately underdoped cuprates. An investigation of the magnetoeffects of compositions located in the vicinity of the superconducting to antiferromagnetic (AF) phase transition could provide important information about the role played by the spin degrees of freedom on the nucleation of the superconducting state.

In this paper, we address these issues through magnetoresistivity measurements on strongly underdoped $YBa_2Cu_3O_x$ single crystals ($x=6.25$ and 6.36) with the magnetic field H applied parallel to the c axis. Our main result is that the magnetoresistivities of these cuprates reflect the superposition of two contributions:

(i) One contribution has the same sign as the corresponding temperature coefficient of the resistivity (TCR) $\partial\ln\rho_i/\partial T$ ($i=\{c,ab\}$). Thus, if the in-plane resistivity is metallic ($\partial\ln\rho_{ab}/\partial T>0$) and the out-of-plane resistivity is nonmetallic ($\partial\ln\rho_c/\partial T<0$), as is the case in underdoped crystals over an extended temperature range, the in-plane magnetoresistivity is positive while the out-of-plane magnetoresistivity is negative. The incoherence of the out-of-plane transport would lead to such a correlation, of the form: $\Delta\rho_i/\rho_i = Q\ \partial\ln\rho_i/\partial T$, where either $Q=\zeta^{orb}H^2>0$, reflecting the conventional orbital contribution to MR in the weak-field regime, or $Q\propto\ln(H/H_0)>0$, reflecting the two-dimensional (2D) quantum interference.

(ii) The other contribution to MR is positive irrespective of the sign of TCR, correlates with the onset of AF ordering, and has a $\gamma_i^{AF}H^2$ dependence.

Magnetoresistivity measurements were carried out on two strongly underdoped $YBa_2Cu_3O_x$ ($x=6.25$ and 6.36) single crystals, by keeping the temperature T constant while sweeping the magnetic field H up to 14 T, applied parallel to the c axis. Both components of the resistivity tensor, $\rho_{c,ab}$, as well as both in-plane $\Delta\rho_{ab}/\rho_{ab}$ and out-of-plane $\Delta\rho_c/\rho_c$ magnetoresistivities were measured by a multiterminal method[6] on the *same single crystal* as described in Ref. 7. This allowed us to carry out a *quantitative* comparison between $\Delta\rho_{ab}/\rho_{ab}$ and $\Delta\rho_c/\rho_c$ as a function of T and H. We note that special care was taken to maintain a constant temperature during the magnetic-field sweep and to eliminate the Hall-effect contribution to the measured magnetovoltages.

The puzzling coexistence of nonmetallic $\rho_c(T)$ and metallic $\rho_{ab}(T)$, characteristic to underdoped cuprates, is present in both concentrations. The out-of-plane resistivity is nonmetallic at all measured T for both oxygen concentrations. The in-plane resistivity remains metallic down to $T_{min}\approx 50$ K for $x=6.36$ and down to $T_{min}\approx 200$ K for $x=6.25$, where it turns insulating as well. The long-range AF ordering gives rise to an increase in zero field $\rho_c(T)$ upon cooling through T_N, while it has no noticeable effect on $\rho_{ab}(T)$.[8] The $x=6.36$ single crystal has a Néel transition temperature $T_N\approx 40$ K [determined from $\rho_c(T)$] while the $x=6.25$ single crystal is AF at all $T\leqslant 300$ K.

We had recently shown that the sign of the out-of-plane magnetoresistivity $\Delta\rho_c/\rho_c$ of the $x=6.36$ single crystal measured in a magnetic field of 14 T is, for $T\geqslant 150$ K, the same as the sign of the corresponding temperature coefficient of the resistivity $\partial\ln\rho_c/\partial T$.[9] This is a direct consequence of incoherent charge transport along the c axis. However, $\Delta\rho_c/\rho_c$ in 14 T becomes positive on approaching the antiferromagnetic AF phase (for $T\leqslant 125$ K), increasing strongly

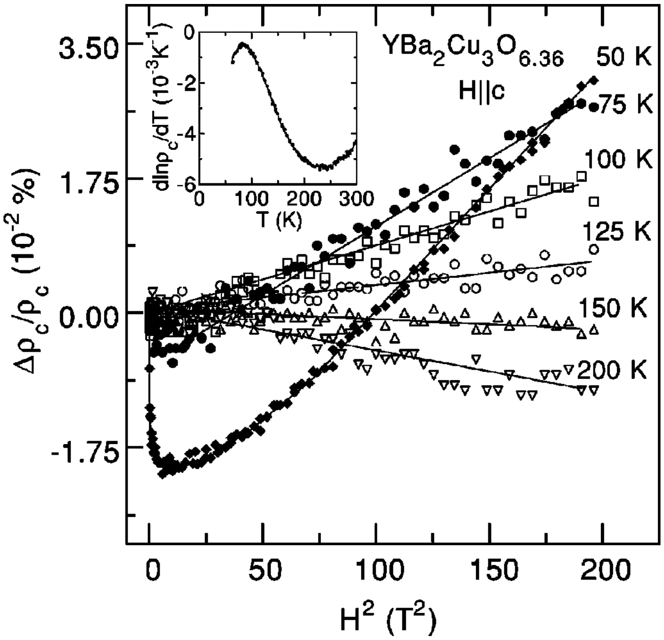

FIG. 1. Magnetic field H dependence of the out-of-plane magnetoresistivity $\Delta\rho_c/\rho_c$ of $YBa_2Cu_3O_{6.36}$ single crystal. Inset: T dependence of the corresponding temperature coefficient of the resistivity $\partial \ln \rho_c/\partial T$ measured in $H=0$ T.

with decreasing T, while $\partial \ln \rho_c/\partial T$ remains negative.[9] A recent report has shown that $\Delta\rho_c/\rho_c(T)$ of strongly underdoped $YBa_2Cu_3O_x$ single crystals measured in high magnetic fields $H \| c$ axis becomes positive well above T_N and increases sharply with decreasing T through T_N.[10] This positive contribution to $\Delta\rho_c/\rho_c$, which is quadratic in H was attributed to AF correlations.[10] Since $T_N \approx 40$ K for the $x=6.36$ single crystal, we associate the positive term that increases with decreasing T for $T \leqslant 125$ K, to increasing AF correlations with decreasing T.

In showing that the correlation between MR and the corresponding TCR is a signature of incoherent c axis charge transport in these samples, we start with the understanding that a fundamental property of incoherent c axis conduction is that the out-of-plane phase coherence length $l_{\varphi,c}$ (the average-distance electrons travel between dephasing inelastic collisions) does not change with temperature or magnetic field.[9] (The out-of-plane incoherent transport of underdoped $YBa_2Cu_3O_x$ was confirmed experimentally.[11–13] Therefore, the only length scale that determines the dissipation and can change with temperature or applied magnetic field is the in-plane phase coherence length $l_{\varphi,ab}$. Under these conditions, both conductivities depend only on the variable $l_{\varphi,ab}$ [$\sigma_{ab}(l_{\varphi,ab})$ and $\sigma_c(l_{\varphi,ab})$], so that their temperature and field dependences come from that of $l_{\varphi,ab}$. Hence:

$$\frac{\partial \rho_{c,ab}}{\partial H}=Q\frac{\partial \rho_{c,ab}}{\partial T}; \quad Q \equiv \frac{\partial l_{\varphi,ab}/\partial H}{\partial l_{\varphi,ab}/\partial T}. \tag{1}$$

The immediate consequence is that the sign of each component of magnetoresistivity is given by the sign of the corresponding TCR since $\partial l_{\varphi,ab}/\partial H<0$ (Refs. 14,15) and $\partial l_{\varphi,ab}/\partial T<0$.[14,16]

The H dependence of $\Delta\rho_c/\rho_c$ is shown in Fig. 1 for $50\text{ K} \leqslant T \leqslant 200$ K. Its inset shows the T dependence of the temperature coefficient of the resistivity $\partial \ln \rho_c/\partial T$ measured in zero field. (The fact that $\partial \ln \rho_c/\partial T<0$ over the whole T range reflects the semiconducting nature of the c axis conduction for all measured T.) For $T \geqslant 150$ K, the direct correlation between the signs of MR (negative) and the corresponding TCR (negative, see inset), i.e., Eq. (1), holds for all the values of the applied magnetic fields. Hence, $\Delta\rho_c/\rho_c$ is given by

$$\frac{\Delta\rho_c}{\rho_c}(H,T)=Q(H)\frac{\partial \ln \rho_c}{\partial T}(T), \tag{2}$$

with $Q \propto H^2>0$. According to Eq. (1), the H dependence of Q, hence, of both magnetoresistivities is given by the H dependence of $l_{\varphi,ab}$. Thus, this H^2 dependence of $\Delta\rho_c/\rho_c$ observed at $T \geqslant 150$ K, indicative of weak-field regime, is a result of the conventional orbital change of $l_{\varphi,ab}$ due to an applied magnetic field $H \| c$; i.e., $Q=\zeta^{orb}H^2$.

For temperatures $100\text{ K} \leqslant T<150$ K, $\Delta\rho_c/\rho_c$ becomes positive, while $\partial \ln \rho_c/\partial T$ is still negative (see inset to Fig. 1). The H dependence is, however, still quadratic. Presumably, there are two contributions to MR in this T range: a *negative* $\zeta^{orb}(\partial \ln \rho_c/\partial T)H^2$ contribution, which is a result of conventional orbital contribution to MR, described above, and a *positive* $\gamma_c^{AF}H^2$ contribution attributed to spin-spin correlations. This latter contribution dominates $\Delta\rho_c/\rho_c$ at these temperatures [$\gamma_c^{AF}>\zeta^{orb}(\partial \ln \rho_c/\partial T)$] since $\Delta\rho_c/\rho_c$ is positive.

At even lower temperatures ($T=50$ and 75 K), a negative component in the H dependence of $\Delta\rho_c/\rho_c$ is present at low fields, superimposed on the positive and quadratic in H term. This negative sign of $\Delta\rho_c/\rho_c$ is the same as the sign of $\partial \ln \rho_c/\partial T$. Therefore, at low H, $\Delta\rho_c/\rho_c$ is also given by Eq. (2), with the H dependence of the negative contribution compatible to $\ln(H/H_0)$ (H_0 is a small characteristic field). At low H, this $\ln(H/H_0)$ contribution to $\Delta\rho_c/\rho_c$ dominates the quadratic in H contributions (orbital and antiferromagnetic contributions).[7] At $H \geqslant 3$ T, the $\ln(H/H_0)$ contribution saturates while the antiferromagnetic contribution $\gamma^{AF}H^2>0$ takes over and changes the sign of $\Delta\rho_c/\rho_c$ to positive.

We had recently shown that the H dependence of $l_{\varphi,ab}$, hence, Q of crystals with 2D phase-coherent paths is given at low enough T and H by[7]

$$Q \propto \frac{\Delta l_{\varphi,ab}}{l_{\varphi,ab}} \approx \begin{cases} -\eta\dfrac{\ln(H/H_0)}{\ln(H_1/H_0)} & H_0<H<H_1 \\ -\eta & H>H_1. \end{cases} \tag{3}$$

Here η is a positive constant, $H_0 \sim \phi_0/l_{\varphi,ab}^2$, and $H_1 \sim \phi_0/l_{el}^2$, where ϕ_0 is the magnetic-flux quantum and l_{el} is the characteristic elastic length. Therefore, the $\ln(H/H_0)$ dependence of $\Delta\rho_c/\rho_c$ observed in Fig. 1 at low T and relatively small H indicates 2D quantum interference, thus, reflects again the incoherent c-axis conduction.

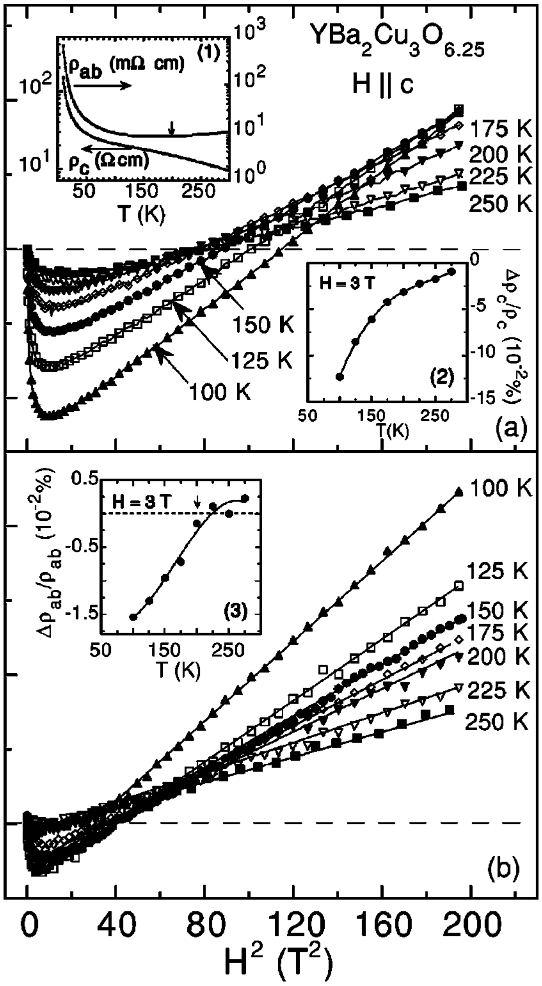

FIG. 2. Magnetic field H dependence of the (a) out-of-plane $\Delta\rho_c/\rho_c$ and (b) in-plane $\Delta\rho_{ab}/\rho_{ab}$ magnetoresistivities of $YBa_2Cu_3O_{6.25}$ single crystal measured at different temperatures T. Inset: (1) Temperature T dependence of in-plane ρ_{ab} and out-of-plane ρ_c resistivities and T dependence of (2) $\Delta\rho_c/\rho_c$ and (3) $\Delta\rho_{ab}/\rho_{ab}$ measured at 3 T.

Hence, for $T<150$ K, there are two contributions to MR: a contribution that has the same sign as the corresponding TCR and a positive contribution attributed to AF correlations, i.e.,

$$\frac{\Delta\rho_c}{\rho_c}(H,T)=Q(H)\frac{\partial\ln\rho_c}{\partial T}(T)+\gamma_c^{AF}(T)H^2, \tag{4}$$

where $Q=\zeta^{orb}H^2>0$ for $T>75$ K and is given by Eq. (3) for $T=50$ and 75 K, and $\gamma_c^{AF}>0$.

The H dependence of $\Delta\rho_{ab}/\rho_{ab}$ (data not shown) is similar with the H dependence of $\Delta\rho_c/\rho_c$ shown in Fig. 1. This reflects the fact that the H dependence of both $\Delta\rho_c/\rho_c$ and $\Delta\rho_{ab}/\rho_{ab}$ is given by $l_{\varphi,ab}(H)$. Specifically, $\Delta\rho_{ab}/\rho_{ab}$ is positive and quadratic in H over the whole field range for $T>75$ K. At $T=50$ and 75 K, $\Delta\rho_{ab}/\rho_{ab}$ is *negative* for $H<3$ T and increases quadratically for $H>3$ T. The negative value of $\Delta\rho_{ab}/\rho_{ab}$ at low fields correlates with the upturn in $\rho_{ab}(T)$ around 50 K and, hence, with the change of sign of $\partial\ln\rho_{ab}/\partial T$ from positive to negative.

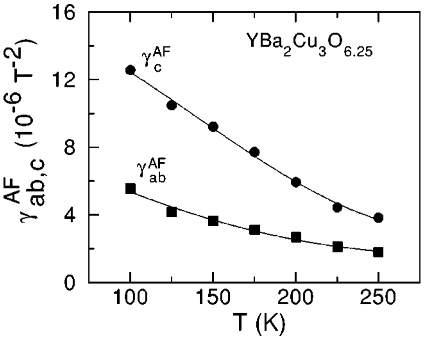

FIG. 3. Temperature T dependence of the coefficient $\gamma_{c,ab}^{AF}$ of the quadratic magnetic-field H dependence of the magnetoresistivities of $YBa_2Cu_3O_{6.25}$ single crystal.

To study the transport in the AF state, we also measured the magnetoresistivity tensor of another single crystal with a lower oxygen content ($x=6.25$) with $T_N>300$ K. Figure 2(a) shows the H dependence of $\Delta\rho_c/\rho_c$ for the $x=6.25$ single crystal measured at several temperatures. For all temperatures 100 K$\leq T\leq$250 K, these curves have the same H dependence as the ones for the $x=6.36$ sample measured close to T_N ($T=50$ and 75 K); i.e., $\Delta\rho_c/\rho_c$ has a nonmonotonic field dependence consistent with $\ln H/H_0>0$ ($H_0\approx0.3$ T at 100 K) at low H, which saturates to a certain negative value ϵ_c (for example, $\epsilon_c\approx-0.14\%$ at 100 K) for $H>3$ T, and a positive contribution quadratic in H that takes over at $H>3$ T and changes the sign of $\Delta\rho_c/\rho_c$ to positive at higher fields.

The H dependence of $\Delta\rho_{ab}/\rho_{ab}$ [Fig. 2(b)] is similar with the H dependence of $\Delta\rho_c/\rho_c$ [Fig. 2(a)]. However, the negative term in $\Delta\rho_{ab}/\rho_{ab}$ is about seven times smaller than the negative term in $\Delta\rho_c/\rho_c$.

The T profiles of $\Delta\rho_c/\rho_c$ and $\Delta\rho_{ab}/\rho_{ab}$ at $H=3$ T, where minima in the magnetoresistivities occur, are plotted in inset 2 and 3, respectively, of Figs. 2. Note that the sign of both $\Delta\rho_c/\rho_c$ and $\Delta\rho_{ab}/\rho_{ab}$ at this low H is the same as the sign of the corresponding TCR [see inset 1 to Fig. 2(a), which gives $\rho_c(T)$ and $\rho_{ab}(T)$]. Indeed, for all temperatures 100 K$\leq T\leq$275 K, $\Delta\rho_c/\rho_c$ at $H=3$ T is negative and increases in magnitude with decreasing temperature. On the other hand, $\Delta\rho_{ab}/\rho_{ab}$ at $H=3$ T is positive for $T>200$ K and negative for $T<200$ K. Therefore, both magnetoresistivities of this sample are given by Eq. (4) over the whole measured T range with Q given by Eq. (3).

The coefficients γ_c^{AF} and γ_{ab}^{AF}, determined from the fit of the quadratic dependence of $\Delta\rho_{c,ab}/\rho_{c,ab}$ at high H [see Eq. (4)] for the $x=6.25$ sample, scale for 100 K$\leq T\leq$250 K with $\gamma_c^{AF}/\gamma_{ab}^{AF}\approx2$. We found the same proportionality between the two coefficients for the $x=6.36$ single crystal, but only for the lowest measured temperature of $T=50$ K, presumably because the AF correlations are strong enough at this T so that the AF contribution dominates the orbital one. This scaling of γ_c^{AF} and γ_{ab}^{AF} is a strong indication that the same mechanism is responsible for the positive, quadratic contributions ($\gamma_{c,ab}^{AF}H^2$) to $\Delta\rho_{c,ab}/\rho_{c,ab}$ even though, as discussed above, this effect is noticeably weaker on the in-plane transport than on the out-of-plane transport.

The coefficients γ_c^{AF} and γ_{ab}^{AF}, for the $x=6.25$ sample are shown in Fig. 3. Both coefficients increase with decreasing temperature for 100 K$\leqslant T\leqslant$250 K. Previous work showed that, immediately below the Néel temperature, γ_c^{AF} decreases with decreasing T.[10] Hence, our data indicate that the temperature behavior of γ_c^{AF} far into the AF regime is different from the one in the vicinity of T_N.

In summary, all the results presented indicate that both magnetoresistivities of underdoped $YBa_2Cu_3O_x$ ($x=6.36$ and 6.25) are described by Eq. (2) above T_N, and by Eq. (4) around and below T_N. Therefore, both in-plane and out-of-plane magnetoresistivities of strongly underdoped single crystals of $YBa_2Cu_3O_x$ ($x=6.25$ and 6.36) are a result of two contributions: one that correlates in sign and temperature dependence with the corresponding temperature coefficient of the resistivity $\partial \ln \rho/\partial T$ and has either a $\zeta^{orb}H^2$ or an $\ln(H/H_0)$ dependence, and another one, which is positive, has a $\gamma_{c,ab}^{AF}H^2$ dependence, and dominates at high magnetic fields ($H>3T$). The first contribution is a fingerprint of the incoherent nature of the out-of-plane charge transport. The second contribution reflects the AF correlations.

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