Violation of Kohler's Rule in the Normal-State Magnetoresistance of $YBa_2Cu_3O_{7-\delta}$ and $La_2Sr_xCuO_4$

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The normal-state magnetoresistance (MR) $\Delta\rho/\rho$ of 90-K and 60-K $YBa_2Cu_3O_{7-\delta}$ and $La_{2-x}Sr_xCuO_4$ ($x = 0.17$) has been measured in the longitudinal (field **H** parallel to current **J**) and transverse (**H** $\parallel$ **c**) geometries, with **J** in-plane. In all cases, the orbital component of the MR displays a temperature dependence that strongly violates Kohler's rule. We show that the classical orbital MR measures the variance of a local Hall angle $\theta(s)$ around the Fermi surface. The anomalous transverse MR in these cuprates is closely related to the temperature dependence of the Hall angle.

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Recently, progress has been achieved in understanding the unusual temperature dependence of the normal-state Hall effect of the high-temperature superconductors [1,2]. In all hole-type cuprates near optimal doping, the Hall resistivity ρ_{xy} displays a strong temperature dependence that persists to temperatures (T) as high as 500 K [2–10]. An important clue to the origin of this anomaly was obtained by analyzing the Hall angle θ_H rather than ρ_{xy} or the Hall conductivity σ_{xy} [1]. An investigation by Chien, Wang, and Ong [2] of how θ_H varies with T and c_{Zn} (concentration of Zn Impurities) in $YBa_2Cu_{3-x}Zn_xO_{7-\delta}$ revealed that the complicated dependences of ρ_{xy} on T and c_{Zn} simplify to the relationship $\cot\theta_H = \alpha T^2 + \beta c_{Zn}$ predicted in Ref. [1]. In addition to the Zn-doping study, several groups have since investigated this T^2 dependence in $YBa_2Cu_3O_{7-\delta}$ (YBCO) vs the oxygen deficit δ [3,4], as well as concentration of the impurities Pr, Fe [5], and Co [6]. The effect of impurities on $\cot\theta_H$ in $Bi_2Sr_2CaCu_2O_{8+\delta}$ has also been reported [7]. The Hall angle has been studied in $La_{2-x}Sr_xCuO_4$ doped with $3d$ elements [8], as well as in the two-chain superconductor $YBa_2Cu_4O_8$ [9]. These studies confirm that the $\cot\theta_H$ vs T^2 relationship is valid (up to 500 K in some cases). In some cases, deviations from the T^2 behavior become significant in the underdoped or overdoped regimes [10]. In the new superconductor HgBaCaCuO, $\cot\theta_H$ is also observed to vary as T^2 up to 400 K [11].

The T^2 behavior of $\cot\theta_H$ was inferred from the proposed compositeness of the charge carriers in the normal state and the assumption that the lifetime τ_H associated with scattering processes parallel to the Fermi surface (FS) has a $1/T^2$ dependence, distinct from the $1/T$ behavior of the transport lifetime τ_{tr}. To go further, we ask if the T^2 dependence in τ_H has consequences observable in other transport quantities. The classical orbital magnetoresistance (MR) is especially appealing since it involves the same scattering processes as the Hall current. It is well known that the classical MR is zero in an isotropic metal. Thus, the MR probes deviations of the FS from sphericity [12]. In a two-dimensional (2D) system, we show that the classical MR measures the *variance* of the local Hall angle around the FS. Thus, it provides unique information on how the electronic mean free path varies around the FS. We report here high-resolution measurements of the MR in 90-K and 60-K YBCO, and $La_{2-x}Sr_xCuO_4$ (LaSrCuO).

The twinned YBCO crystals, with $T_c = 92$ and 63 K, were annealed in flowing oxygen for ten days at 500 and 650 °C, respectively (the 60-K crystal was quenched from 650 °C into liquid nitrogen). The LaSrCuO crystal with $T_c = 38$ K was cut from a large boule grown by a traveling-solvent floating-zone technique. While MR measurements at a resolution of 100 ppm (parts per million) are routinely performed with capacitance thermometers as the regulating sensor, measurements at the level of 10 ppm are precluded because of dielectric aging effects (drifts may be observed 24 h after temperature stabilization) [13]. For our measurements, we used a resistive cernox sensor (Lakeshore 1050) that was carefully calibrated in a field as follows. We let the sample chamber equilibrate at a selected T for 3 h under open-loop control. The field is ramped up to 14 T and back to zero in 40 min. If the trace of the sensor reading versus field has a hysteresis less than 20 ppm, it is accepted as a calibration curve (this shows that ΔT is less than 5 mK). Calibration of the sensor allows the intrinsic MR signal of the sample to be extracted from the observed signal by compensating for the effect of field on the sensor.

The field dependence of the in-plane resistivity ρ is shown in Fig. 1 for 90-K YBCO in the transverse geometry ($\mathbf{B}\|\mathbf{c}, \mathbf{J}\perp\mathbf{c}$). At all temperatures, ρ increase as B^2 (positive MR) with a curvature that changes rapidly with T. By fitting with the form $[\Delta\rho(B)/\rho(0)]_\perp = a_\perp B^2$, we may extract the MR coefficient $a_\perp(T)$ at each temperature (subscript $\perp$ indicates that $\mathbf{B}\perp\mathbf{J}$). The fits are shown as smooth lines in Fig. 1. Classically, the transverse MR arises from "bending" of the electron trajec-

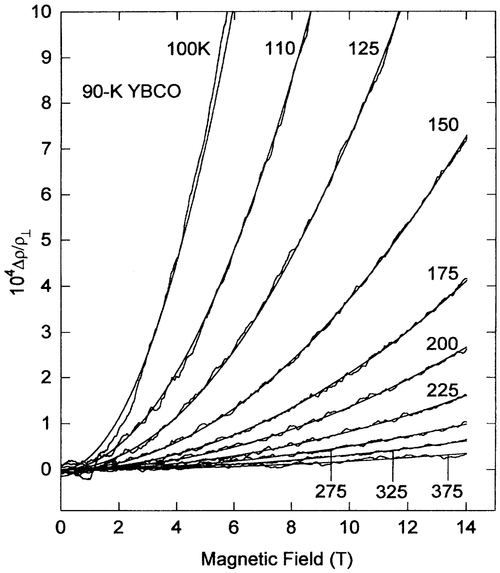

FIG. 1. Variation of the resistivity with field for a twinned 90-K $YBa_2Cu_3O_{7-\delta}$ crystal in the transverse geometry (**B**$\|$**c**, **J**$\perp$**c**), at selected temperatures. The curves with fluctuations at the level of 10–20 ppm represent actual data recording (each curve is comprised of ~1500 data points). Smooth lines are fits by $[\Delta\rho/\rho]_\perp = a_\perp B^2$. The data curves for 60-K YBCO and LaSrCuO have similar fluctuation levels (10–20 ppm).

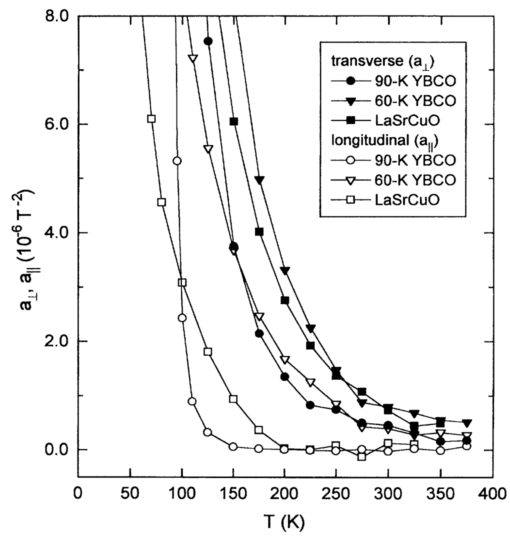

FIG. 2. The T dependence of the MR coefficients $a_\perp$ and $a_\|$ in 90-K and 60-K $YBa_2Cu_3O_{7-\delta}$, and LaSrCuO, in the transverse (solid symbols) and longitudinal geometry (open), where $a_\perp \equiv [\Delta\rho/\rho]_\perp B^{-2}$ and $a_\| \equiv [\Delta\rho/\rho]_\| B^{-2}$, where subscripts denote orientation of **B** relative to **J**. Lines are guides to the eye.

tory by the Lorentz force. We also made MR measurements with the field parallel to **J** (with **J**$\perp$**c**) to obtain the *longitudinal* MR coefficient $a_\| \equiv [\Delta\rho(B)/\rho(0)]_\| B^{-2}$ (the Lorentz force is absent in this geometry). Figure 2 summarizes the T dependences of $a_\perp$ (solid) and $a_\|$ (open symbols). In the three systems, the transverse MR is positive, strongly T dependent, and resolvable up to our highest temperature 375 K [13]. The longitudinal coefficient in 90-K YBCO, however, falls below our resolution, $a_\| < 0.2$ ppm, above 150 K (the steep increase below 150 K arises from paraconductivity effects). In contrast, $a_\|$ in 60-K YBCO grows rapidly below ~270 K [a similar positive increase below ~200 K is seen in LaSrCuO (open squares)]. Because these signals appear at temperatures too high above T_c to be caused by paraconducting fluctuations, we identify them with an isotropic spin-dependent term that is intrinsic to the normal state. We note that the spin-dependent contribution in 60-K YBCO lifts its transverse MR signal significantly above that of 90-K YBCO (compare the solid triangles and circles in Fig. 2). Hence, to extract the orbital part of the MR $a_{\rm orb}$, we systematically subtract, in all samples, the longitudinal from the transverse coefficient at each temperature, viz. $a_{\rm orb}(T) \equiv a_\perp(T) - a_\|(T)$. Figure 3 (main panel) displays the temperature dependence of $a_{\rm orb}$ in the three samples in log-log scale. In the YBCO samples between 100 and 375 K, $a_{\rm orb}$ follows a power law T^{-n}, with $n = 3.5$ and 3.9 in the 90-K and 60-K crystals, respectively. The additional MR caused by field suppression of superconducting fluctuations is apparent in both crystals as a very steep increase (starting near 100 K in the 90-K crystal). We will not discuss these well-studied paraconductivity MR effects, aside from noting that previous analyses may have unwittingly included the normal-state T^{-n} MR as part of the paraconductivity MR [14].

Unlike in YBCO, $a_{\rm orb}$ in LaSrCuO does not follow a power-law dependence (squares). Instead, the data above ~70 K fit well by the expression $[m/(bT^2 + c)]^2$. We trace the difference to the Hall angle θ_H. Whereas $\cot\theta_H$ in the two YBCO crystals is proportional to T^2, in LaSrCuO $\cot\theta_H$ varies as $b_H T^2 + c_H$ with b_H and c_H quite close in value to b and c, respectively (see inset). We interpret the finite intercept c_H as indicative of a large impurity scattering term in LaSrCuO arising from Sr disorder. Thus, in clean YBCO, as well as in LaSrCuO (where impurity scattering is significant), the T dependence of the orbital term tracks that of the Hall angle, viz. $a_{\rm orb} \sim \tan^2\theta_H$. The unusual T dependences and the correlation between θ_H and $a_{\rm orb}$ are our central results.

It is customary to analyze the classical orbital MR using the Kohler plot [15]. The motivation is that, in conventional metals, the coefficient of the B^2 term is proportional to the transport scattering time $\tau_{\rm tr}(T)$. Since ρ is proportional to $1/\tau_{\rm tr}$, a plot of $\Delta\rho/\rho$ vs

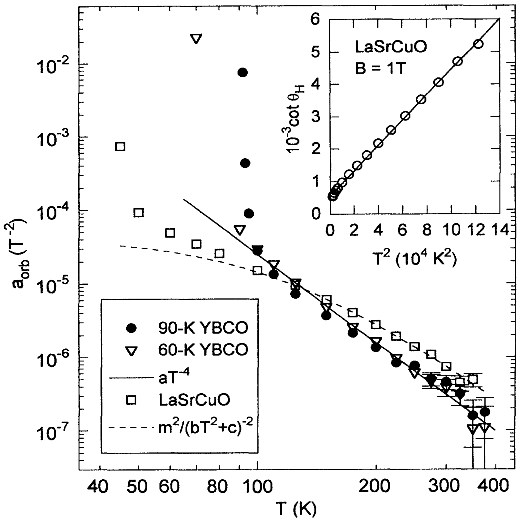

FIG. 3. (Main panel) The temperature dependence of the orbital part of the MR coefficient $a_{\rm orb}$ in 90-K YBCO (circles), 60-K YBCO (triangles), and LaSrCuO (squares) ($a_{\rm orb} \equiv a_\perp - a_\parallel$). The solid line has the form $a_{\rm orb} = CT^{-4}$ with $C = 2510$ K^4. The LaSrCuO data are fitted by $[m/(bT^2 + c)]^2$ with $m = 3.6$, $b = 0.0414$, $c = 554$ (broken line). The paraconductivity contributions are observed as steep deviations from power law in the YBCO data. The inset shows $\cot\theta_H$ (at 1 T) plotted against T^2 in LaSrCuO. The solid line is $\cot\theta_H = b_H T^2 + c_H$ with $b_H = 0.0391$ and $c_H = 554$.

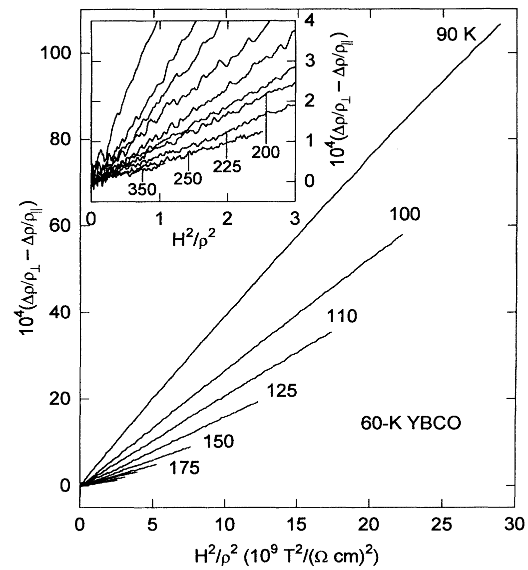

FIG. 4. Kohler plot for the 60-K YBCO crystal at temperatures between 90 and 175 K (main panel). The vertical axis represents the difference of the fractional changes in ρ in the two field geometries. The inset shows the same plot magnified by a factor of ~ 10 to show the curves between 200 and 350 K. The axes labels in the main panel also apply to the inset. All curves displayed represent actual (unsmoothed) data recordings. If Kohler's rule is valid, all the curves should coincide.

$(H/\rho)^2$ should fall on a straight line with a slope that is independent of T. In Fig. 4, we have replotted the data from the 60-K sample. It is clear that Kohler's rule is strongly violated. Instead of a single curve, we obtain a series of lines whose slope steeply decreases with increasing T. A similar violation of Kohler's rule is observed in 90-K YBCO and in LaSrCuO [16]. We argue below that this strong violation is related to the unusual temperature dependence of the Hall effect.

The classical MR derives from the bending of the electron trajectory. For a conventional 2D metal with an arbitrary FS, the correction to the in-plane conductivity $\Delta\sigma$ may be calculated from the Boltzmann equation to give [17,18] $\Delta\sigma = -(e^2/2\pi^2)(eB/\hbar)^2 \int ds\, l(s)[d/ds(l(s)\cos\phi(s))]^2$, where s is the arclength along the FS, $l(s)$ is the mean free path, and $\phi(s)$ is the angle between the velocity $\mathbf{v}(s)$ and the electric field $\mathbf{E}$. For a FS that is fourfold symmetric, we may replace $\cos\phi(s)$ with $\sin\phi(s)$. The fractional change in σ then assumes the simple form

$$\Delta\sigma/\sigma = -\int ds\, \Sigma(s)\theta(s)^2, \qquad (1)$$

where the conductivity weight $\Sigma(s)$, satisfying $\int ds\, \Sigma(s) = 1$, is equal to $(e^2/2\pi^2)l(s)\cos^2\phi(s)/\sigma$. The local Hall angle $\theta(s)$ on the patch ds is defined as the ratio $\Delta j_\perp/j_\parallel$, where $\Delta j_\perp$ is the change in the Hall current when the state shifts by $eBl/\hbar$ on the FS, and $j_\parallel \sim l\cos\phi$. Hence, we have

$$\theta(s) = (eB/\hbar\cos\phi)\, d(l\sin\phi)/ds. \qquad (2)$$

Combining Eq. (2) with the Hall conductivity expression [18], $\sigma_H = [e^3B/(2\pi\hbar)^2]\times \int ds\, l\cos\phi\, d(l\sin\phi)/ds$, we verify that the observed Hall angle is just the weighted average of $\theta(s)$ over the FS, i.e., $\theta_H = \int ds\, \Sigma(s)\theta(s)$. In general, the MR (up to terms in B^2) may be written as $\Delta\rho/\rho = -\Delta\sigma/\sigma - \theta_H^2$ (note $\rho \equiv \rho_{XX}$, $\sigma \equiv \sigma_{XX}$). Finally, using Eq. (1) for $\Delta\sigma/\sigma$, we obtain

$$\Delta\rho/\rho = \langle\theta(s)^2\rangle - \langle\theta(s)\rangle^2, \qquad (3)$$

where the averaging $\langle\cdots\rangle$ means $\int\Sigma(s)\cdots ds$. Equation (3), valid for a 2D FS that is fourfold symmetric but otherwise arbitrary, states that the classical, orbital MR measures the variance of the Hall angle over the FS. [In the elementary case of a circular FS, for which the variance vanishes, Eq. (3) reduces to the familiar result $\Delta\rho = 0$].

The observed Hall angle θ_H and the MR provide, respectively, information on the mean of the local Hall angle $\theta(s)$ and its variance at each temperature. Hall measurements show that $\theta_H^2 = D/T^4$, with $D = 1510$ and 1630 K^4 in the 60-K and 90-K YBCO crystals, respectively. There are two interesting extremes. For Kohler's rule to be observed ($\Delta\rho/\rho \sim T^{-2}$), we need the first term in Eq. (3), $\langle\theta(s)^2\rangle$, to vary as T^{-2} and to be dominant over the second term. In the other extreme, all three terms in Eq. (3) share the same T dependence, and Kohler's rule is strongly violated. As the variance has the same T dependence as the mean squared, $\theta(s)$ must change *uniformly* with T, i.e., its relative variation with position around the FS is the same at each temperature. Since the observed ratio $[\Delta\rho/\rho]/\theta_H^2$ is either T independent (in 60-K YBCO and LaSrCuO) or almost so (90-K YBCO), we are much closer to the second extreme in all three samples. The discussion on the behavior for $\theta(s)$ then implies that the mean free path $\mathbf{l}(s,T)$ has the form $\mathbf{v}(s)\tau_H(T)$, with τ_H independent of s [$\mathbf{v}(s)$ is the velocity and τ_H the relaxation time]. This conclusion seems incompatible with proposed models [6,7] in which $\mathbf{l}(s)$ is assumed to vary as $1/T$ and $1/T^2$ on different parts of the FS.

The numerical value of the ratio $[\Delta\rho/\rho]/\theta_H^2$ provides further information. In 60-K YBCO, the ratio equals 1.7, while in 90-K YBCO, it changes weakly with T from 1.5 to 1.7. The ratio in LaSrCuO, however, is much larger (13.6). Such a large value implies that, even though the observed Hall signal is positive in optimally doped LaSrCuO, the local Hall angle $\theta(s)$ is negative in sign over a significant segment of the FS. This is probably a precursor to the change of sign observed in θ_H when x increases past 0.25, towards the overdoped regime.

The previous analysis of the Hall angle [1] may be extended to treat the MR. The charge carriers are assumed to be decomposed into particles carrying charge and spin degrees. We propose that different "memory times" $\tau_{\rm tr}$ and τ_H are associated with the response to the E field and the Lorentz force, respectively. Heuristically, the distribution function in the kinetic equation may be written as $g_{\mathbf{k}} = \tau_{\rm tr} e\mathbf{E}\cdot\mathbf{v_k}(-\partial f_0/\partial\varepsilon) - \tau_H e\mathbf{v_k}\times\mathbf{B}\cdot(\partial g_{\mathbf{k}}/\partial\mathbf{k})$. In the zero magnetic field, $\tau_{\rm tr}$ alone enters into the resistivity. For the weak-field MR, $\tau_{\rm tr}$ cancels out in the weight Σ in the integrand of Eq. (1), so that $\Delta\sigma/\sigma \sim \tau_H^2$ (similarly, $\sigma_H \sim \tau_{\rm tr}\tau_H$, and $\theta_H \sim \tau_H$). If $\tau_{\rm tr}$ and τ_H vary as T^{-1} and T^{-2}, respectively, we obtain directly the observed power-law temperature dependences of the three transport quantities, σ, θ_H, and $\Delta\sigma/\sigma$. The violation of Kohler's rule in the classical MR of both 90-K and 60-K YBCO and in LaSrCuO uncovers yet another anomalous aspect of normal-state transport in the cuprates near optimal doping. In high-purity systems, the in-plane conductivity, the Hall conductivity, and the weak-field MR vary as T^{-n} with exponents (n = 1, 3, and 4, respectively). Impurity scattering adds a constant shift to the transport and Hall scattering rates. The latter change appears sufficient to account for the unusual MR behavior in LaSrCuO. It seems that these anomalous power laws reflect a fundamental property of the charge carriers and that the identification of two distinct lifetimes provides that most natural way to understand these experiments.

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