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Unusual $1/T^3$ temperature dependence of the Hall conductivity in $YBa_2Cu_3O_{7-\delta}$

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Measurements of the Hall coefficient R_H and resistivity ρ_a of $YBa_2Cu_3O_7$ disclose anomalies in their temperature (T) dependences. The $1/R_H$ vs T curve has a negative curvature, while $d\rho_a/dT$ displays a previously unreported step near 290 K. The Hall conductivity σ_H follows an unusual $1/T^3$ dependence from 100 to 360 K. We contrast the results with predictions of conventional Bloch theory, and compare them with recent calculations based on large-U models. Implications for cyclotron damping at low T are discussed.

The normal-state Hall coefficient R_H of the 90 K superconductor $YBa_2Cu_3O_7$ (Y-Ba-Cu-O) shows an anomalous temperature dependence which remains unexplained.[1] An R_H that is positive and increases monotonically with decreasing temperature T appears to characterize all the cuprate superconductors. The T dependence in Y-Ba-Cu-O (Refs. 2–7) is most pronounced, but there is increasing evidence that R_H behaves similarly in the other cuprates as well. Although early measurements in ceramic samples of the cuprates $La_{2-x}Sr_xCuO_4$ (2:1:4), $Nd_{2-x}Ce_xCuO_{4-\delta}$, and $Bi_2Sr_2CaCu_2O_8$ (Bi 2:2:1:2) showed a weak T dependence in R_H, more reliable results are now available from samples that have been optimized systematically for superconductivity. Recent results show that, in each compound, the T dependence of R_H increases with T_c. Suzuki and Takagi *et al.* found that R_H is strongly T dependent in 2:1:4 when x is within the superconducting window.[8] Doping studies by Clayhold *et al.*[9] showed that when superconductivity is destroyed in Y-Ba-Cu-O or 2:1:4 by doping with Co or Ni, the slope dn_H/dT shows a corresponding decrease ($n_H \equiv 1/eR_H$). More recently Kubo *et al.*[10] reported that in the Tl-based cuprates, oxygen annealing has a pronounced effect on T_c and R_H. In their data, the slope dn_H/dT shows a strong positive correlation with T_c. In $Nd_{2-x}Ce_xCuO_{4-\delta}$ single crystals that are superconducting, R_H becomes positive below 50 K, and increases monotonically as T decreases to 2 K.[11] These studies suggest that the strong T dependence of R_H is an intrinsic property of the charge carriers in the CuO_2 layer. The T dependence of ρ_a and R_H have recently been calculated in large-U models that incorporate a gauge force.[12,13] We compare below our results with the predictions of these models.

In Y-Ba-Cu-O, approximate descriptions of the T dependence of the Hall coefficient and the in-plane resistivity ρ_a are often described by the equations[1]

$$1/R_H \approx aT+b, \ (a,b>0)\,, \tag{1}$$

$$\rho_a(T)=\rho_a(0)+\alpha T \ (\alpha>0)\,. \tag{2}$$

How reliable are these equations? Because Hall data on Y-Ba-Cu-O are usually sparse, Eq. (1) has not been tested to high precision. Moreover, simultaneous measurements of ρ_a are often not available, so that neither the Hall angle θ_H nor the Hall conductivity $\sigma_H=\sigma_{xy}$ (our interest here) can be computed. Equation (2) has only been tested (at the 1% level) for temperatures below 300 K in Y-Ba-Cu-O. Measurements on crystals to this level of accuracy are rare above 300 K.

To address these issues, we have carried out detailed measurements of R_H and ρ_a to high temperatures in single-crystal Y-Ba-Cu-O that have T_c's above 90 K. To attain the requisite precision and data density, we have automated the Hall measurements. At each temperature, the sample is rotated through 180°, in a fixed field **B** of 8 T, by a computer-controlled stepper motor. At each orientation of **B** normal to the a-b plane, the Hall voltage V_H and the resistance were simultaneously measured for the two ($\pm$) current directions. We have checked that R_H at 100 K remains linear in B between +8 and −8 T. By careful shielding of leads, we attain a resolution for V_H approaching 20 nV. The high-temperature resistance measurements[14] were performed with the sample either in a vacuum, in a high oxygen pressure (~3 atm), or in flowing oxygen. By comparing data from the cooling and warming runs (rate 100 K/h), we checked that there is no measurable hysteresis in ρ_a in each case, and that loss of oxygen is not a serious factor below 600 K.

Figure 1 displays the temperature dependence of the reciprocal of the Hall coefficient $1/R_H$ and the in-plane resistivity ρ_a in samples 1–3. Although sample 3 has a resistivity (and R_H) larger than in samples 1 and 2, all samples show the same qualitative behavior. We first discuss the R_H curves. For samples 1 and 2, the value of the Hall density n_H equals 6.2×10^{21} cm^{-3}, or 1.1 carriers per unit cell at 100 K. In contrast to the linear-T description in Eq. (1), the $1/R_H$ curve shows a negative curvature. The data trend suggests that R_H may approach a constant value at T above 400 K. Close examination of the resistivity data (solid lines) also shows a slight positive curvature in ρ_a vs T in all 3 samples. This feature becomes quite apparent when we look at the T dependence up to 600 K in Fig. 2 (samples 4 and 5). By sighting along the curves, we see that the bend is actually caused by a change in slope that occurs between 280 and 300 K. The derivative $d\rho_a/dT$ shows a distinct step in this temperature interval, followed by a slow increase with T above 300 K. Thus, ρ_a is approximately linear up to ~290 K where it undergoes a change in slope. Above 300 K, its increase with T is slightly faster than linear (the data fit well to T^n where $n=1.3\pm0.02$). These results demonstrate that subtle changes in ρ_a occur near room tempera-

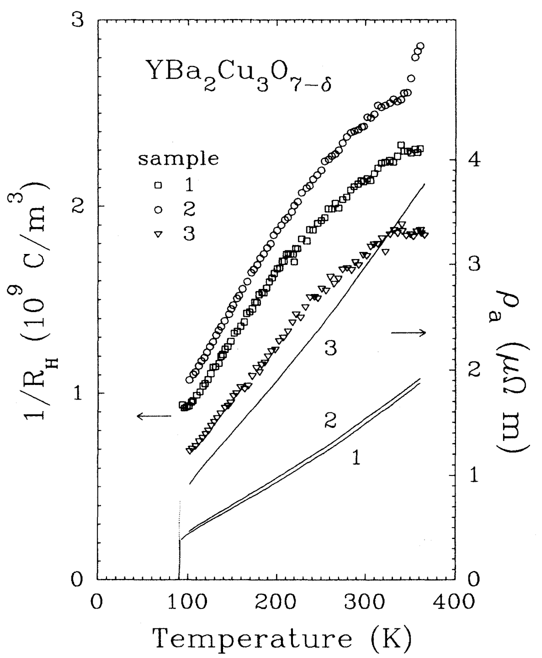

FIG. 1. The temperature dependence of $1/R_H$ (symbols) and the in-plane resistivity ρ_a (solid lines) in single-crystal $YBa_2Cu_3O_7$. R_H and ρ_a are measured in a field of 8 T normal to the a-b plane. (Dotted line near 90 K shows the zero-field transition in the three samples.)

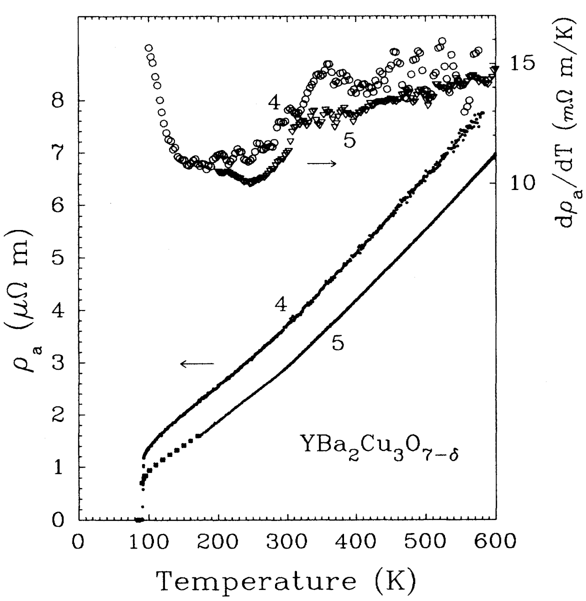

FIG. 2. The T dependence of ρ_a in two crystals of Y-Ba-Cu-O (lower curves) measured by the van der Pauw technique with current reversal. The data for sample 4 were taken while warming and cooling in a vacuum in the sequence 295 → 700 → 77 K. For sample 5, the sample was in 3 atm of oxygen when warmed from 295 to 700 K. The cool down from 700 to 170 K was performed in 1-atm-flowing O_2. (Condensation of the oxygen disrupted the measurement below 170 K.) A separate run below 170 K was performed to obtain the solid square symbols in sample 5. The numerical derivatives of both data sets are shown as open symbols.

ture and above. Since the oxygen ions remain mobile at these temperatures, it is possible that oxygen reordering causes a slight change in the scattering rate $1/\tau_{tr}$. A positive curvature is discernible in two previous high-T measurements on ceramic[15] and thin-film[16] Y-Ba-Cu-O samples. Interestingly, Martin *et al.*[16] observe a change in slope in $YBa_2Cu_3O_{8+\delta}$ (1:2:4) between 250–300 K [but not in Y-Ba-Cu-O (1:2:3)].

Our results show that neither Eqs. (1) nor (2) provide accurate descriptions of the T dependences. It is of interest to examine, instead, the T dependence of the simpler quantities, $\tan\theta_H$ and the Hall conductivity σ_H. Plotted on a log scale (Fig. 3), the data for $\tan\theta_H \equiv \sigma_H \rho_a = R_H B/\rho_a$ approximate the power-law behavior,

$$\tan\theta_H = D/T^2 , \tag{3}$$

over the whole temperature range, with $D \approx 150$. However, the data deviate noticeably from Eq. (3) (by as much as 25% in samples 1 and 2). By contrast, we find that the Hall conductivity $\sigma_H = (\tan\theta_H)/\rho_a$ more closely follows the power law

$$\sigma_H = C/T^3 . \tag{4}$$

To emphasize deviations of the data on σ_H from Eq. (4), we have plotted the product $T^3\sigma_H$ against T in Fig. 4. Because $1/T^3$ is a steep function of T, the relatively flat variation of the curves (under 7%) in Fig. 4 provides persuasive evidence that the power-law dependence in Eq. (4) is closely adhered to, at least below $\sim$360 K. (Below 120 K, field suppression of superconducting fluctuations leads to an observable magnetoresistance of the order of $\sim 10^{-3}$. This causes the slight upturn in Fig. 4.) Of the three quantities R_H, $\tan\theta_H$, and σ_H, the last fits most closely to a power-law dependence. Since σ_H is a simpler

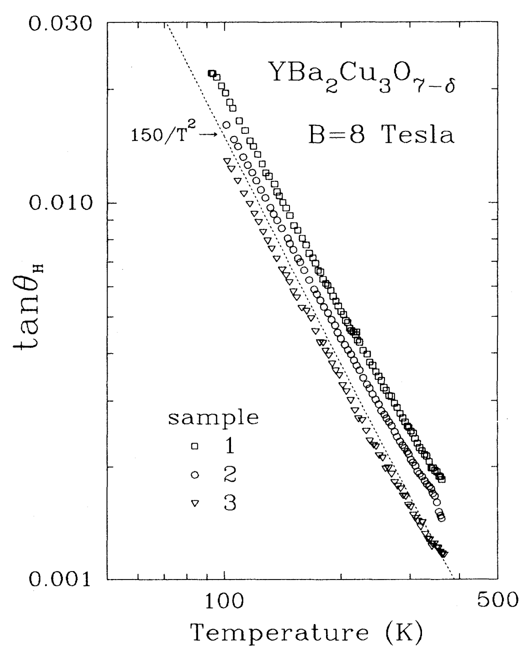

FIG. 3. Variation with temperature of the $\tan\theta_H$ in Y-Ba-Cu-O measured at 8 T.

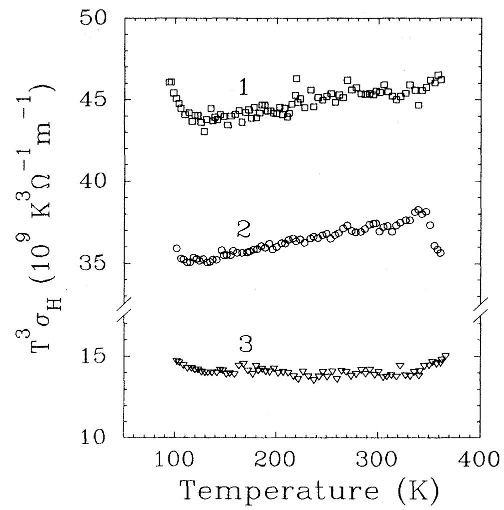

FIG. 4. Plot of the product $T^3\sigma_H$ vs T in Y-Ba-Cu-O. ($\sigma_H = R_H B/\rho_a^2$ is calculated from the data in Fig. 1. $B = 8$ T.)

quantity than $R_H = \sigma_H \rho_a^2/B$, its observed $1/T^3$ dependence is also fundamentally more interesting than the temperature dependence of R_H.

Equation (4) is anomalous when analyzed in terms of conventional transport theory. In the single-band Drude model, R_H is temperature independent if the *anisotropy* of the scattering rate $1/\tau(\mathbf{k})$ is independent of T. If the average of $\tau(\mathbf{k})$ over the Fermi surface (FS), τ_{tr}, is linear in $1/T$ (as required by the observed ρ_a), we must have $\tan\theta_H \sim 1/T$ and $\sigma_H \sim 1/T^2$. All three predictions are in disagreement with the data here. Next, we relax the constraint on the anisotropy of $1/\tau(\mathbf{k})$. It is well known that in conventional metals R_H is often strongly temperature dependent because the anisotropy in $1/\tau(\mathbf{k})$ changes with temperature.[17] As T decreases, phonon scattering becomes weighted towards regions of the FS that have small caliper, so that their contribution to the Hall current is suppressed. The nonmagnetic metals, Cu, Ag, Cd, W, and Mg, have FS's of varying degrees of complexity. Although the T dependence of R_H in these metals is pronounced at low temperatures, it becomes very weak above a characteristic temperature $T_H \equiv s\Theta_D$ in each case (Θ_D is the Debye temperature). The parameter s has the approximate value 0.29 (Cu), 0.44 (Ag), 0.24 (Cd), 0.20 (W), and 0.30 (Mg).[18] A calculation[19] shows that, in Cu, phonon scattering leads to a $\tau(\mathbf{k})$ that is isotropic at $T = \Theta_D$ and $0.5\Theta_D$, but strongly anisotropic at $0.2\Theta_D$. Thus, electron-phonon scattering leaves a characteristic imprint on the R_H vs T profile: Above T_H, R_H rapidly approaches a constant value as $\tau(\mathbf{k})$ becomes isotropic. In comparison with these metals, the variation of R_H with T in Y-Ba-Cu-O is striking both in its magnitude and in its persistence to 360 K. Moreover, the power-law variation of σ_H up to 360 K implies the absence of any characteristic temperature scale. (A phonon-scattering mechanism would require an effective Debye temperature greater than 900 K in Y-Ba-Cu-O, if we estimate $T_H \approx 400$ K and $s \approx 0.3$–0.4. Heat-capacity estimates of Θ_D in Y-Ba-Cu-O range from 400 to 440 K.) Barring the existence of such energetic phonons, we conclude that it is highly unlikely that the temperature dependence of R_H is caused by anisotropy of the electron-phonon scattering rate (within the a-b plane).

A related explanation is the two-band model in which the Hall current of the holes and electrons partially cancel. R_H may be strongly T dependent if the respective mean free paths have different temperature dependences. If electron-phonon scattering dominates at the temperatures of interest (>100 K), the analysis of the preceding paragraph applies, so that R_H should be a constant above T_H. Independent of this argument, Eq. (4) imposes additional difficulties for the two-band model. Since Hall currents are additive, σ_H is the sum of the Hall conductivities of the hole and electron band, σ_{H1} and σ_{H2}, respectively. ($\sigma_{H2} < 0$. We assume that both σ_{H1} and $\sigma_{H2} \sim 1/T^2$.) To get the $1/T^3$ variation in Eq. (4), we must have exact cancellation of the leading terms of σ_{Hi} in the series expansion $\sigma_{Hi} = A_i/T^2 + B_i/T^3 + \cdots$ (A_i and B_i are constants, $i = 1,2$). This cancellation[20] requires σ_{H1} and σ_{H2} to be very close in magnitude over the whole temperature range 100–360 K. Such a delicate balance in Y-Ba-Cu-O seems to us to be highly implausible. Moreover, as previously noted, it is contradicted by Hall measurements[9] on Y-Ba-Cu-O doped with Co or oxygen vacancies, and by high-pressure measurements[7] of R_H.

These difficulties, combined with doping results that demonstrates the unusual sensitivity of dn_H/dT to Ni and Co impurities,[9] suggest that it is futile to seek explanations within conventional Bloch-Boltzmann theory. Measurements of the field dependence[8] of R_H at low T (up to 20 T at 2 K in $Nd_{2-x}Ce_xCuO_{4-\delta}$)[11] also rule out conventional magnetic skew scattering as the source of the anomalous Hall current. Instead, we adopt the premise that only holes are present in Y-Ba-Cu-O (single band), and the anomalous σ_H reflects an *intrinsic* property of the unusual charge carriers in the CuO_2 planes. The appearance of a similar R_H vs T profile in the other superconducting cuprates (discussed above) suggests that the anomalous Hall current may be universal in these systems.

Two recent papers[12,13] have discussed the temperature dependence of ρ_a and R_H in large-U Hubbard models in which the constraint of single-site occupancy is locally enforced by coupling to a gauge field. A feature of the gauge models is that the electrical resistivity is the sum $\sigma_B^{-1} + \sigma_F^{-1}$, where σ_B (σ_F) are the conductivities of the boson (fermion) excitations.[21] In both papers,[12,13] ρ_a is linear in T since $\sigma_B \ll \sigma_F$. The linear-T behavior is consistent with our data on Y-Ba-Cu-O only up to ~ 280 K. The existence of the step in $d\rho_a/dT$, unfortunately, precludes comparison with data at higher T.[22] For the Hall effect, Nagaosa and Lee[12] obtain an R_H that increases with T ($R_H \sim \text{const} - c/T$), in disagreement with experiment. Ioffe, Kalmeyer, and Wiegmann[13] compute an n_H that increases monotonically with T but with pronounced negative curvature, in qualitative agreement with Fig. 1. The data on σ_H in Fig. 4 should provide a more direct, quantitative comparison with their calculations. Anderson[23] has proposed a third scenario, in the class of large-U models, that distinguishes the relaxation time for cyclotron motion τ_c from the transport relaxation time τ_{tr}. The former involves displacement of the wave vector **k** transverse to the velocity, whereas the latter involves longitudinal processes. If damping of the cyclotron motion proceeds via spin excitations alone, $\tan\theta_H = \omega_c\tau_c$ will vary like $1/T^2$ as in Eq. (3) (ω_c is a cyclotron frequency). The persistence of this behavior to low T implies that $\omega_c\tau_c \gg 1$ at 4.2 K. It may be possible to observe directly such long-lived cyclotron orbits despite screening by the Meissner current. [Anticipating that the trend in Fig. 3 saturates near 450 K, we may equate $\hbar/\tau_c$ to $\eta k_B T$ at that temperature ($\eta \sim 2$–3). This determines the cyclotron mass to be $\sim 28/\eta$ times the free mass.]

In summary, we have shown that, underlying the strong T dependence of the Hall effect in Y-Ba-Cu-O, is an unusual variation of σ_H with T. We discussed in detail why the observed $1/T^3$ behavior and its persistence to high temperatures are incompatible with conventional Bloch-Boltzmann theory. Neither in-plane anisotropy of the electron-phonon scattering rate nor a two-band Drude model works. We compared the T dependence of R_H with two recent calculations.[12,13] The implication of our results for damping of cyclotron orbits was briefly discussed. In addition, measurements of ρ_a to 600 K show that ρ_a develops a hitherto unsuspected change in slope near 290 K which we tentatively ascribe to oxygen reordering.

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