Hall-effect measurements of $HgBa_2CaCu_2O_{6+\delta}$

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The resistivity ρ and Hall coefficient R_H have been measured for samples of the two-plane superconductor $HgBa_2CaCu_2O_{6+\delta}$ ($T_c \sim 124$ K) with different annealing histories. The observed temperature dependence $R_H \sim 1/T$ (up to 470 K) matches the steep dependence previously seen in 90-K $YBa_2Cu_3O_{7-\delta}$ and indicates low-impurity scattering. We find T-linear behavior for ρ in the lowest-resistivity samples and confirm that $\cot\theta_H \sim T^2$. These findings add support to Anderson's proposal of distinct longitudinal and transverse scattering times.

I. INTRODUCTION

The discovery of the Hg-Ba-Ca-Cu-O family of high-T_c superconductors[1,2] has stimulated many experiments including studies of the crystal structure, susceptibility, pressure dependence of T_c,[3] resistivity, and thermopower,[4] but many important physical properties, including the Hall effect, remain unexplored. In the study of the normal state of cuprate superconductors, the Hall effect has assumed a significance far beyond that of simply measuring the carrier density because of its anomalous behavior.[5] The Hall coefficient R_H is strongly temperature dependent up to high temperatures for motion of the charge carriers in the CuO_2 planes. By contrast R_H is temperature independent and negative for the geometry in which the Hall electric field is directed along the c axis in the hole-type cuprates.[6] Although R_H can be temperature dependent at low temperatures for ordinary metals with dominant electron-phonon scattering, it saturates to a constant value at temperatures above a small fraction of Θ_D (0.2–0.4). The cuprates have $\Theta_D \approx 400$ K, so another interpretation for the in-plane R_H must be found.

Anderson has proposed that the Luttinger liquid theory provides a natural explanation for anomalous features of transport data in high-T_c superconductors.[7] Within this picture, spin-charge separation in the CuO_2 planes results in two different relaxation rates, a longitudinal or transport relaxation rate $1/\tau_{tr} \sim T$ and a transverse or Hall relaxation rate $1/\tau_H \sim T^2$ (with a temperature-independent term added to both rates when impurity scattering is present). The first gives the well-known linear-T resistivity, while the second gives the Hall angle dependence $\cot\theta_H = \alpha T^2$. The Hall coefficient R_H is a composite of the two more fundamental quantities and has a nominal $1/T$ dependence since $R_H = \rho_{xx}\tan\theta_H/B \propto \tau_H/\tau_{tr}$.

The relationship $\cot\theta_H = \alpha T^2 + \beta'$ (where β' is proportional to the impurity concentration) was initially found to hold for Zn-doped $YBa_2Cu_3O_{7-\delta}$ (YBCO) by Chien, Wang, and Ong.[8] It has subsequently been confirmed for a large number of systems including the following: optimally doped and underdoped YBCO (Ref. 6) and $La_{2-x}Sr_xCuO_4$ (LSCO) (Ref. 9) up to 500 K; Pr-, Co-, and Fe-doped YBCO;[10–12] untwinned YBCO;[13] Fe-, Co-, Ni-, Zn-, and Ga-doped LSCO;[14] Y-, Ni-, and Zn-doped $Bi_2Sr_2CaCu_2O_8$ (BSCCO);[15] and untwinned $YBa_2Cu_4O_8$ with double CuO chains (except below 160 K when **J** is parallel to the chain direction).[16] Systematic deviations have been reported in some materials. In the electron-doped superconductor $Nd_{2-x}Ce_xCuO_4$, the Hall angle can be fit to $\cot\theta_H = A + BT^2 + CT^4$.[17] Also, deviations have been noted for overdoped LSCO. On the whole, the T^2 dependence has been found to be as robust as the well-known linear-T resistivity and is a hallmark of the normal state of the cuprates.

In this paper we report measurements of ρ and R_H on the double CuO_2 layer compound $HgBa_2CaCu_2O_{6+\delta}$ (Hg-1212) as a function of temperature (up to 470 K) and oxygen annealing. We also test the $\cot\theta_H = \alpha T^2$ rule and compare our results with Hall-effect data from other cuprate superconductors.

II. EXPERIMENTAL DETAILS

Hg-1212 is a member of the homologous series $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$ with $n = 2$, indicating two tightly coupled CuO_2 planes per unit cell. The preparation of polycrystalline Hg-1212 samples has been described in detail by Meng *et al.*[18] The sample purity is 85% as determined by x-ray-diffraction, neutron-diffraction, and magnetization measurements. Impurities consist of $CaHgO_2$, $BaCuO_2$, and $Ba_2Cu_3O_{5.9}$. Several groups have studied the changes in lattice parameters and T_c with oxygen annealing.[19–21] Samples can be either under- or overdoped as determined by the amount of oxygen in the centered position of the Hg layers. We annealed our samples at 300 °C in one atmosphere of oxygen, a procedure that has been found to produce samples with close to optimal doping. The resulting T_c's ranged from 121 to 124 K at the midpoint with transition widths

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of 3–5 K. The samples were cut into bars 2 mm×1 mm×175 μm and then mechanically polished, taking care to minimize the exposure time to atmospheric moisture. Contact pads were made by sputtering a Ag film or applying Ag epoxy. Having a freshly polished surface was important to producing low-resistance contacts (0.5–5Ω) with low annealing temperatures (300 °C or less) and annealing times as short as 30 min.

Hall-effect and resistivity measurements were carried out in a 14 T superconducting magnet using two lock-in amplifiers synchronized at 16 Hz with $J \sim 0.5$ A/cm^2. The longitudinal and transverse voltages were found to be linear in current and magnetic field. We reversed the magnetic field at each temperature by rotating the sample 180° to cancel the lead misalignment. The sample was carefully isolated from the liquid-helium bath using 15 layers of aluminized mylar so temperatures to 550 K could be reached. The sample or contacts became slightly unstable at high temperatures, however, limiting reproducible measurements of resistivity to 420 K and Hall effect to 470 K. (The Hall-effect limit is slightly higher because the quick field reversal eliminates slow, uniform drifts.) Careful temperature control was essential since the measured Hall angles were as small as 3×10^{-5} rad. Each of the points above 300 K is the average of several readings.

III. RESULTS AND DISCUSSION

The magnitude and temperature dependence of the resistivity showed considerable variation from sample to sample and with different annealing conditions as shown in Fig. 1. The magnitude of the resistivity in these polycrystalline materials is, of course, increased by a considerable factor over its intrinsic value by random orientation of grains, grain boundaries, and impurities. Samples A and B were cut from the same piece of ceramic and annealed at 300 °C in oxygen for 14 and 1.5 h, respectively, while sample C was annealed for less than 1 h. Although oxygen annealing initially raises the T_c of as-grown samples, it appears that further annealing increases the resistivity and introduces curvature into the resistivity versus temperature curve. Meng *et al.* reported a similar degradation in resistivity curves upon repeated annealing.[22] They attributed this to an insulating phase forming in the grain boundaries during brief exposures of the samples to atmospheric humidity or to Hg loss during the 300 °C annealing. In light of these findings, we expect that the intrinsic in-plane resistivity is linear in temperature with zero intercept, behavior which sample C closely approximates.

The Hall coefficient R_H is positive (hole-like) and strongly temperature dependent up to the highest temperature measured (470 K). It can be fit to γ/T quite accurately in both samples A and B as shown in Fig. 2. R_H is essentially identical for the two samples, which is surprising since their resistivities differ by a factor of 2. This indicates that R_H is much less sensitive to grain boundaries, inhomogeneities, and impurities than ρ. Previous experience on high-T_c ceramics[5] and theoretical calculations[23] indicate that R_H should be within 10% of the in-plane R_H (**J**‖*ab*-plane and **H**‖**c**) of single crystals of the same material. The magnitude of R_H is similar to that of other cuprates, but the carrier density n_H cannot be extracted simply even in the context of Boltzmann theory since $n_H = 1/(eR_H)$ only for the simple case of isotropic scattering, parabolic bands and a circular Fermi surface.

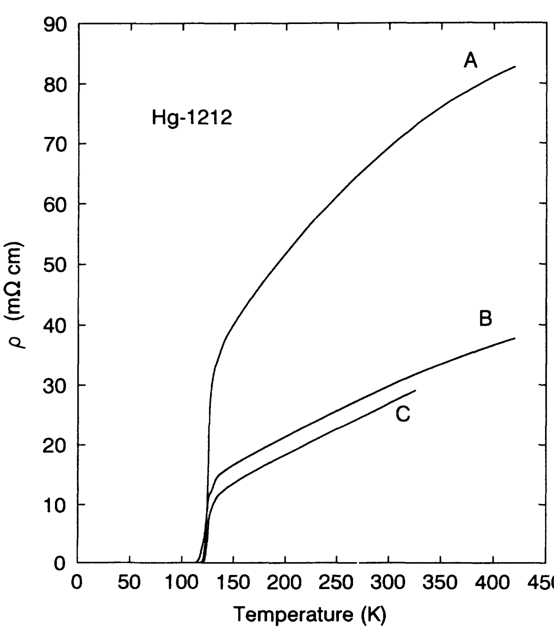

FIG. 1. The temperature dependence of the resistivity ρ for samples A, B, and C of polycrystalline Hg-1212. The samples were annealed in flowing oxygen at 300 °C for 14, 1.5, and <1 h, respectively.

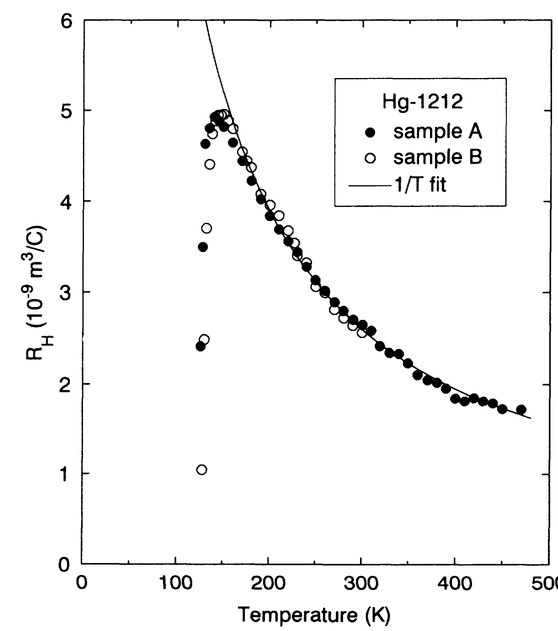

FIG. 2. The temperature dependence of the Hall coefficient R_H in Hg-1212 for samples A and B. The data can be fit accurately to γ/T as shown by the solid line. The magnitude and temperature dependence of R_H is the same for both samples in spite of their different resistivities (see Fig. 1).

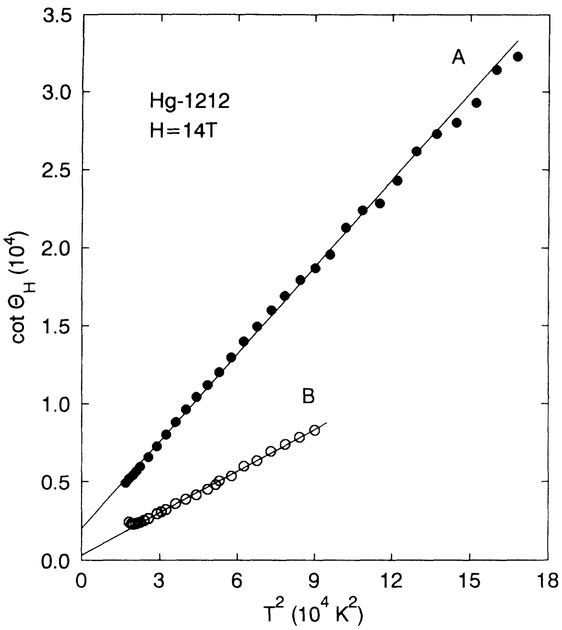

FIG. 3. The Hall angle data for samples A and B plotted as $\cot\theta_H$ ($=\rho_{xx}/BR_H$) vs T^2. The solid lines show fits of the data to $\cot\theta_H=\alpha T^2+\beta'$. The magnetic field is 14 T.

The observed $1/T$ temperature dependence is well known from 90 K YBCO, while LSCO and BSCCO approach this steep dependence for very pure samples. It has long been known that impurities suppress the temperature dependence of R_H in the cuprates. The suppression follows naturally from the form $\cot\theta_H=\alpha T^2+\beta'$ since $R_H=\rho/B\cot\theta_H$ becomes less temperature dependent as the impurity term β' increases. Thus, the steep temperature dependence of R_H in YBCO is related to β' being approximately zero. As we show next, Hg-1212 is also a clean system in the sense that the impurity scattering term β' is quite small.

The Hall-effect data are plotted in terms of the Hall angle θ_H in Fig. 3. Anderson's relation $\cot\theta_H=\alpha T^2+\beta'$ is found to hold, with small deviations in sample A resulting from ρ being slightly sublinear in T. As we noted above, deviations from $\rho\propto T$ seem to reflect slight sample degradation rather than an intrinsic effect. A least-squares fit gives the parameters for samples A and B as $\alpha=0.186$, $\beta'=2020$ and $\alpha=0.0891$, $\beta'=337$, respectively. The ratio of $\beta'/\alpha=3780$ for sample B is smaller than that of any cuprate superconductor other than YBCO. The small ratio indicates that the doping process of adding oxygen to the Hg layers introduces little disorder for electronic transport in the CuO_2 planes.

IV. CONCLUSIONS

Normal-state transport measurements of longitudinal resistivity and Hall coefficient were performed on polycrystalline Hg-1212 samples with different annealing times. These measurements strongly resemble results obtained on other high-T_c families, and are especially similar to optimally doped YBCO. Specifically, we observe the temperature dependences $\rho\sim T$, $R_H\sim 1/T$, and $\cot\theta_H\sim T^2$. The temperature dependence of R_H is as steep as any seen so far in the cuprates, and it is indicative of low-impurity scattering. The strikingly different temperature dependences of ρ and θ_H provide further evidence for non-Fermi-liquid transport and constitute a test for theories of the normal state.

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