Measurement of Anisotropic Resistivity and Hall Constant for Single-Crystal $YBa_2Cu_3O_{7-x}$

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The resistivity of single-crystal $YBa_2Cu_3O_{7-x}$ has been found to be anisotropic, with magnitude and temperature dependence similar to ceramic samples in directions parallel to the Cu-O planes, and with a 30 times larger room-temperature value and a much smaller temperature dependence in the orthogonal direction. The Hall coefficient, with a magnetic field applied parallel to the Cu-O planes, is negative (electronlike) and essentially temperature independent in these crystals, in direct contrast to the behavior of other types of samples.

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The report of superconductivity at unprecedented high temperatures in $La_{2-x}Ba_xCuO_4$ by Bednorz and Müller[1] led to the discovery of superconductivity at temperatures above 90 K in $YBa_2Cu_3O_{7-x}$ and numerous related compounds.[2] The layered perovskite structure[3] of these materials indicates that they should be quite anisotropic. Transport measurements on single crystals are thus an important step towards gaining an understanding of these materials. Anisotropic magnetic properties (critical fields, critical current density) have been reported in single crystals of both $La_{2-x}Ba_xCuO_4$ and $YBa_2Cu_3O_{7-x}$-type materials,[4-7] as has an anisotropy in resistivity in $La_{2-x}Sr_xCuO_4$ single crystals[8] and in oriented films[9] of $YBa_2Cu_3O_{7-x}$. In addition to being anisotropic, these new superconductors represent a state of matter significantly different from those which are presently understood. A fundamental question is whether or not the normal state from which the superconducting one condenses is a Fermi liquid, as is believed to be the case with all other known superconductors. One signature of the Fermi-liquid state is a quadratic dependence of electrical resistivity on temperature below the range in which phonon scattering dominates. The linear temperature dependence of the resistivity characteristic of these materials in ceramic form[3] appears, in this context, to be rather mysterious, implying a breakdown of the Fermi-liquid description. For example, Lee and Read[10] infer both nonphonon and *d*-wave pairing from this linear dependence.

In this paper we report measurements of the anisotropic resistivity and Hall coefficient in bulk single-crystal $YBa_2Cu_3O_{7-x}$. We find that the resistivity in the *a* and *b* directions (i.e., parallel to the Cu-O planes) is approximately 450 $\mu\Omega$ cm at room temperature, decreasing linearly with temperature above the superconducting transition with a slope of 1.3 $\mu\Omega$ cm/K. This is similar to the resistivity behavior of ceramic samples, for which slopes of 1.7–2.5 $\mu\Omega$ cm/K are typical.[3] Thus we can state that the relatively large magnitude and the linear temperature dependence of resistivity are bulk properties and not artifacts of the granular nature of the ceramic materials. In the *c* direction (i.e., orthogonal to the Cu-O planes), we find a room-temperature resistivity 30 times larger than in the in-plane direction, with only a weak temperature dependence, confirming the highly anisotropic nature of the material. The Hall coefficient, measured with a magnetic field applied parallel to the Cu-O planes, is electronlike and virtually temperature independent, in contrast to the holelike and temperature-dependent Hall constant observed in ceramic samples[11] and epitaxial thin films.[12]

The crystal-growth process, described elsewhere,[13] yields crystals in the shape of rectangular parallelpipeds. One typical form is thin platelets, as large as several millimeters in size in the basal (Cu-O) plane by a few tens of micrometers in the orthogonal (*c* axis) direction. The other common shape is euhedral, with dimensions of up to 0.5×0.5 mm^2 by 0.2 mm.

Low-resistance Ohmic contacts to these crystals proved to be difficult to form. Contact resistance values in the range 10^{-3} to 10 Ω cm^2 were obtained by ultrasonic bonding of Al or Au wires and by direct probing. The presence of a poorly conducting surface layer, possibly residual melt, often made even four-terminal measurements of the underlying superconducting crystal impossible in many cases. We developed a process, to be described elsewhere,[14] which results in Ohmic contacts with specific resistance values in the range 10^7 to 10^6 Ω cm^2. The crystal is masked with only the contact areas exposed during the contact process.

For measurement of the resistivity anisotropy, $\simeq 10$ μm$\times$10 μm^2 contacts were formed on the corners of a rectangular crystal face containing the *c* axis. The two directions in the face used in the measurement and the one orthogonal to it will be denoted by subscripts 1, 2 (along the *c* axis), and 3, respectively, with corresponding lengths of 275, 97, and 290 μm for the sample reported here. The standard Montgomery[15] and van der Pauw[16] techniques were used to extract the anisotropic resistivity and Hall coefficient.

Four-terminal ac measurements yielded two resistances, R_1 with the current terminals parallel to the *a*-*b* planes (perpendicular to the *c*-axis direction) and R_2 with the current terminals along the *c*-axis direction, as illustrated in Fig. 1. The two resistances have very different values and temperature dependences, as shown in Fig. 2. The small dots in Fig. 2 are data taken with an rms measurement current of 500 μA (peak current density 710 A/cm^2 at the contacts). In the low-resistance direction, R_1 falls rapidly with decreasing temperature. In the high-resistance (c axis) direction, R_2 is $\simeq 100$ times larger than R_1 at room temperature and increases with decreasing temperature, attaining a value about 60% above its room-temperature value just above the superconducting transition. The large solid resistance data points were obtained with an rms current of 20 mA (peak current density 2.8×10^4 A/cm^2 at the contacts) in a separate experiment. With the larger current, the superconducting transition was clearly seen in both R_1 and R_2. The good agreement with the lower-current data indicates that heating and critical current were not problems even with the larger current. There is a difference in T_c between the two runs. The transition temperature of this sample was originally 91 K with a 10%-90% width of less than 1 K. It dropped to a value of approximately 80 K, still retaining the narrow transition width, after being tested several times. The reason for this decrease is not known; however, the normal-state resistances R_1 and R_2 did not change. T_c was stable in other samples, which had contacts parallel to the a and b directions. T_c varied from $\simeq 75$ to 91 K from sample to sample, depending on the annealing conditions, without major differences in the normal-state resistivity.

The large difference between the resistances R_1 and R_2 indicates a substantial anisotropy between the in plane and c directions, since the higher resistance corresponds to the shorter sample dimension. To extract the components of the resistivity tensor, the method developed by Montgomery[15] was used. In keeping with his notation, the potentials and currents present in an anisotropic sample with dimensions l_1', l_2', and l_3' and (diagonal) resistivity tensor components ρ_1, ρ_2, and ρ_3, are mapped onto an equivalent isotropic sample with resistivity $\rho = (\rho_1\rho_2\rho_3)^{1/3}$ and dimensions l_1, l_2, and l_3. Given R_1 and R_2, the actual sample dimensions, and the assumption $\rho_1 = \rho_3 \neq \rho_2$, we obtain the components of resistivity. The magnitude and temperature dependence of ρ_1 which we obtain is in agreement with values obtained from measurements on other crystals involving contacts only in the a-b plane. No a-b plane anisotropy was observed, although a small anisotropy would be consistent with our measurements. These crystals exhibit twinning in the a-b plane,[13] which would tend to cause the in-plane resistivity to appear more isotropic. Thus the assumption that there are only two components of resistivity, ρ_{ab} $(= \rho_1 = \rho_3)$ and ρ_c $(= \rho_2)$, which we used in our analysis is a good one.

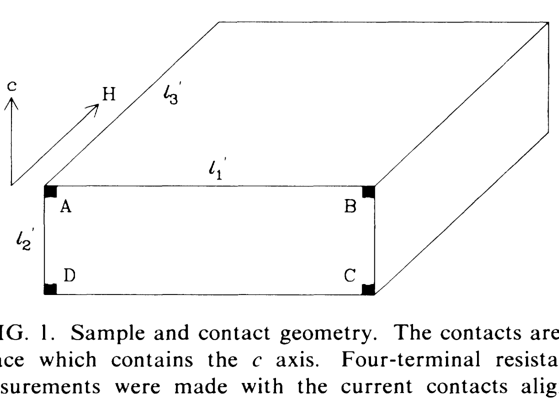

FIG. 1. Sample and contact geometry. The contacts are on a face which contains the c axis. Four-terminal resistance measurements were made with the current contacts aligned perpendicular to the c axis of the crystal ($R_1 = V_{DC}/I_{AB}$), and parallel to it ($R_2 = V_{BC}/I_{AD}$). For Hall measurements, V_{AC}/I_{DB} is measured with a magnetic field, **H**, applied perpendicular to the contacted face.

The rapid decrease in R_1 with decreasing temperature, which is essentially exponential, can be understood as being due to the increasing distance between the voltage leads and the current leads in the equivalent isotropic sample. As a result of the increasing value of the ratio R_2/R_1 with decreasing temperature, l_2/l_1 increases from 1.9 to 3.4 between 300 and 100 K ($l_2'/l_1' = 0.35$). l_2 grows from 300 to 440 μm while l_1 changes only slightly, from 160 to 130 μm. The resistivity ratio ρ_2/ρ_1 increases from 30 to $\simeq 80$ as temperature is lowered. In measurements on samples with all contacts in the a-b plane, the measured *resistances* are much larger because of the more favorable effective sample geometry.

The resistivities ρ_1 (or ρ_{ab}) and ρ_2 (or ρ_c) are plotted as functions of temperature in Fig. 3. ρ_1 is 450 $\mu\Omega$ cm at room temperature, 180 $\mu\Omega$ cm at T_c, and extrapolates to 75 $\mu\Omega$ cm at $T = 0$, with a slope of 1.3 $\mu\Omega$ cm/K. It

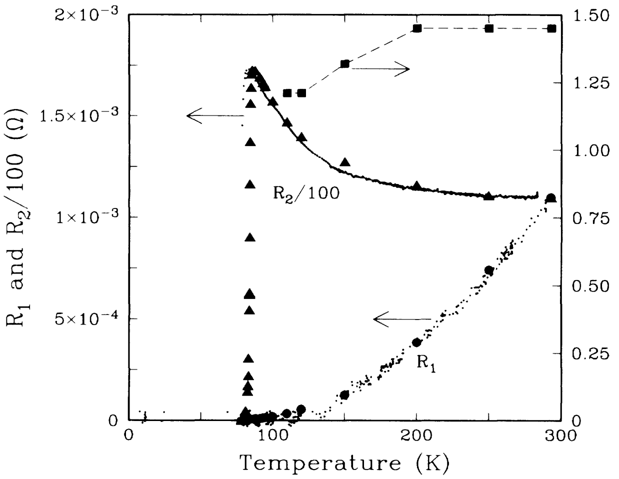

FIG. 2. Four-terminal resistances R_1 and R_2 as a function of temperature. Note the factor of 100 difference between R_1 and R_2 at room temperature. The small and large symbols represent data from different runs with currents of 500 μA and 20 mA, respectively. Also plotted is the Hall number (V_0/eR_H), corresponding to 1.2-1.5 electrons per unit cell.

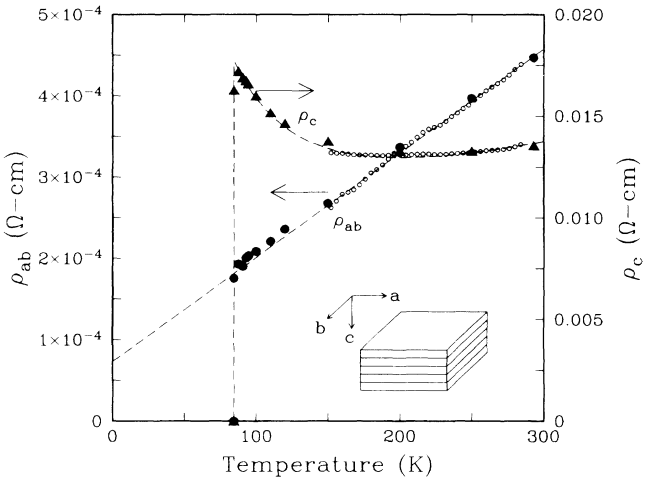

FIG. 3. Resistivity tensor components parallel to the Cu-O planes (ρ_{ab} or ρ_1) and perpendicular to them, i.e., along the c axis (ρ_c or ρ_2). The small open and large solid symbols are for two different measurements with currents of 500 μA and 20 mA, respectively. Inset: Schematically illustrates the crystal directions with respect to the Cu-O planes which dominate the conductivity.

is similar in magnitude and temperature dependence to the resistivity of ceramic samples. In contrast, ρ_2, the resistivity in the c direction, is much larger and increases somewhat with decreasing temperature. For a 500-μA current, the rapidly falling value of R_1 limits the temperature range of our analysis to above roughly 150 K. By assuming that ρ_1 is linear in temperature down to T_c, as observed in other samples with measurements along the a-b planes and in ceramic samples, we can extrapolate the behavior of ρ_2, which increases slightly as temperature is lowered, as shown by the dashed line in Fig. 2. For this we used an extrapolation of ρ_2/ρ_1, which varied smoothly with temperature. The resistance values R_1 and R_2 calculated using the extrapolated resistivities were in good agreement with the measured ones so that, given the linear $\rho_1(T)$, the determination of $\rho_2(T)$ is unique all the way to T_c. The extrapolation is consistent with the 20-mA data. The c-axis resistivity is much larger than the in-plane resistivity, increasing somewhat with decreasing temperature, with a value of 13–17 mΩ cm.

In earlier measurements[8] on $La_{2-x}Sr_xCuO_4$, a large resistivity anisotropy between the a and c directions was inferred with use of two different bar samples. Both exhibited broad transitions and similar temperature dependences, with resistivity increasing with decreasing temperature above the onset of superconductivity. Zero resistance was not attained until 3.8 K. We observed similar temperature dependences on $YBa_2Cu_3O_{7-x}$ samples with poor contacts and attributed our results to a nonsuperconducting surface layer rather than to poor sample quality. Recent measurements on oriented $YBa_2Cu_3O_{7-x}$ thin films[9] show a factor-of-20 anisotropy between in-plane and out-of-plane resistivities at room temperature, increasing to about 40 near T_c. The in-plane resistivity is higher by roughly a factor of 2 than that of our crystals, possibly as a result the imperfect stoichiometry of the films. The c-axis resistivity fell slightly with decreasing temperature.

Hall measurements were made with the same sample which we discussed above. A magnetic field of ±1 T was applied perpendicular to the contacted face of the crystal (i.e., parallel to the a-b planes). The Hall voltage was linear in H at 0.5 and 1 T. It is convenient to normalize the Hall constant, R_H, to the unit cell volume, $V_0 = 174$ Å^3, and the electronic charge so that, in the case of one isotropic parabolic band the Hall number, V_0/eR_H, is the number of carriers per unit cell. For this sample, the Hall constant is in the range -7.5×10^{-10} to -9×10^{-10} m^3/C, giving a Hall number, shown in Fig. 2, which is electronlike (negative) and nearly temperature independent, as expected for a metal, indicating 1.2–1.5 electrons per formula unit. The thickness of the equivalent isotropic sample, 150 μm, and not the actual thickness of 290 μm was used to obtain this value. This behavior is in sharp contrast to that observed in ceramic[11] and polycrystalline epitaxial[12] samples, in which cases the Hall number is holelike and proportional to T. In ceramic $La_{2-x}Sr_xCuO_4$, a p-type Hall coefficient which was nearly temperature independent was found.[11] The behavior was purely holelike for $x < 0.15$ but mixed (both holes and electrons) for $x > 0.15$.

Recently, Allen, Pickett, and Krakauer[17] used a band-theory approach to calculate the anisotropic transport properties, including the resistivity and Hall tensors, of doped La_2CuO_4-based materials. This analysis has been extended[18] to $YBa_2Cu_3O_{7-x}$. Both positive (for B parallel to the c axis) and negative Hall coefficients (for **B** in the a or b direction) are predicted for both of these materials, depending on the field direction (due to the nonparabolic bands, $R_H \neq -1/ne$). For in-plane magnetic fields orthogonal to and along the ordered Cu-O chains, they predict Hall constants of -3.5×10^{-10} and -11×10^{-10} m^3/C, in reasonable agreement with our single in-plane number of $\simeq -8\times10^{-10}$ m^3/C. They also predict zero-temperature resistivity anisotropies of 5.6 and 15 between the in-plane and c directions, somewhat smaller than we extrapolate from our measurements, and an anisotropy of 2.7 between the in-plane directions.

In summary, we have measured the anisotropic resistivity of single-crystal $YBa_2Cu_3O_{7-x}$. The in-plane behavior is like that of ceramic material, showing that the linear temperature dependence of resistivity is not an artifact of the granular materials. Along the c axis the resistivity is much larger and increases with decreasing temperature. The Hall coefficient, measured with the **B** field perpendicular to the c axis, is electronlike with a value corresponding to 1.3 carriers per cell.

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