c-axis stress dependence of normal and superconducting state properties of $YBa_2Cu_3O_7$

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We have applied pressure (i.e., stress) to single-crystal $YBa_2Cu_3O_7$ along the c axis to reduce the separation between the copper-oxygen planes. For the normal state, increasing pressures up to ~ 1 kbar causes the c-axis resistivity to decrease dramatically and tend toward a metalliclike temperature dependence. No change is observed in the ab-plane resistivity. The superconducting T_c increases with decreasing interplane separation with $dT_c/dP \approx 0.1$ K/kbar. Some implications of these findings for normal-state conduction mechanisms and models of high-T_c superconductivity for $YBa_2Cu_3O_7$ are discussed.

A feature common to high-T_c superconductors with transition temperature $T_c \gtrsim 40$ K is the presence of Cu-O planes which in turn lead to large anisotropy in the structural and electronic properties of these materials. In $YBa_2Cu_3O_7$, for example, normal-state resistivity,[1-3] Hall effect,[1] and thermoelectric power measurements[2] show dramatic anisotropies between the c axis and ab plane (Cu-O plane direction). Critical-field studies[4] show anisotropic behavior in the superconducting state as well. Although the superconductivity mechanism in high-T_c oxides is not yet understood, numerous theoretical approaches exploit the low-dimensional electronic structure of the Cu-O planes. Indeed, in the highest-T_c superconductors based on Bi and Tl,[5] there appears to be a correlation between T_c and the number and separation of Cu-O planes in the unit cell. On the other hand, the Ba-Bi-K-O oxide superconductors[6] have T_c's approaching 30 K but are relatively *isotropic* electronically.

In this article, we describe experiments where pressure (up to ≈ 1 kbar) is applied along the c axis of single-crystal $YBa_2Cu_3O_7$ which effectively squeezes the Cu-O planes closer together. Both the absolute value and temperature dependence of the normal-state c-axis resistivity ρ_c are dramatically influenced by the uniaxial stress. No effect on ab-plane resistivity is observed. As the Cu-O plane separation is decreased, T_c increases smoothly, suggesting that in $YBa_2Cu_3O_7$ the superconductivity is not strictly confined to the two-dimensional Cu-O planes, but depends sensitively on interplane coupling. We examine some implications of our results for normal-state conductivity and superconductivity mechanisms in $YBa_2Cu_3O_7$.

Single-crystal specimens were initially grown by slow cooling of a nonstoichiometric melt in a gold crucible as described elsewhere.[7] Ten crystals were used in this study. The ab-plane resistance was measured on six crystals, and the out-of-plane (c-axis) resistance was measured on four crystals. The onset temperature for superconductivity, T_{c0}, was between 88 and 91 K for all crystals, but transition widths varied from between 1 to over 10 K, with a typical value of ~ 2 K. The crystals were rectangular parallelopipeds with approximate dimensions of $0.7 \times 0.7 \times 0.1$ mm^3.

c-axis pressure was applied to each $YBa_2Cu_3O_7$ crystal by our sandwiching it between two small steel disks. A thin film of Stycast epoxy electrically insulated the ab planes of the crystal from the disks. During the measurement cycle, a constant force (generated by a compressed spring held at room temperature) was applied to the top disk which transferred pressure to the crystal along the c axis. Since no additional "gasket" was used between the steel disks, the apparatus applied only uniaxial stress, and no external pressure along the ab plane of the crystal. Temperature was measured with a calibrated silicon diode attached directly to the pressure cell with General Electric varnish. Electrical contacts to the crystal faces were made by gold wires silver painted to fired-on silver pads. The contacts were reliable and had resistances typically $\leq 0.25\ \Omega$ at all temperatures. Crystal resistivities for the ab plane and the c axis were determined by four-probe contact configurations to eliminate effects of contact resistances.

Figure 1 shows the c-axis resistance versus temperature for a particular crystal at four different c-axis pressures. All of the curves display the "semiconductorlike"

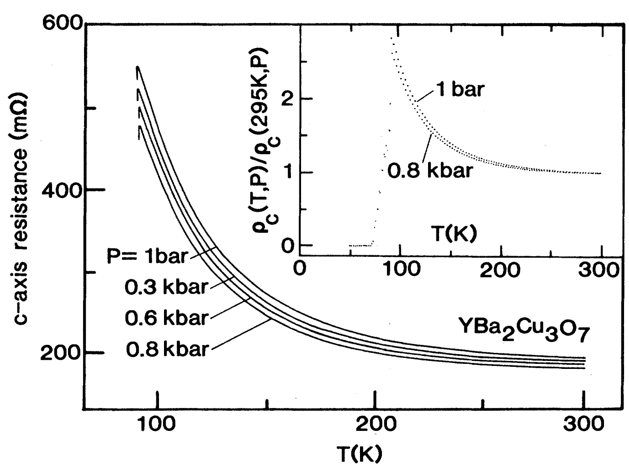

FIG. 1. c-axis resistance vs temperature for selected c-axis pressures in $YBa_2Cu_3O_7$. Inset: Normalized resistivity data for the two extreme pressures.

normal-state resistive upturn with decreasing temperature (before dropping to zero at T_c) first reported by Tozer *et al.*[1] The c-axis resistance is lowered monotonically with increasing pressure across the whole temperature range from room temperature to T_c. While the magnitude of the resistive upturn varies between different samples, the pressure dependence of the resistivity shows the same trends in all the samples. R vs T curves for different pressures in Fig. 1 are not simply related by a multiplicative factor. This is shown more clearly in the inset of Fig. 1, where the c-axis resistivities measured at two different pressures (ambient and $P=0.8$ kbar) are shown as functions of temperature. Both resistivities have been normalized to room-temperature values. From the inset to Fig. 1, it is apparent that an increase of the c-axis pressure depresses the resistive upturn in the R vs T curve, i.e., the c-axis resistivity tends toward a more metallic behavior.[8]

Figure 2(a) shows the detailed pressure dependence of ρ_c, for three selected temperatures. ρ_c is more pressure dependent at lower temperatures: at $T=295$ K, $(1/\rho_c)d\rho_c/dP \approx -0.075$/kbar; while at 95 K, $(1/\rho_c)d\rho_c/dP \approx -0.162$/kbar. While these values are somewhat sample dependent, the quoted numbers are representative.

Our contact configuration did not allow simultaneous measurement of c-axis and ab-plane resistivities on the same crystal. However, the ab-plane resistivity was measured on similar crystals under c-axis pressure. Between 295 K and T_c, no change was observed in ρ_{ab} up to $P=0.8$ kbar, the maximum applied pressure. We note that Borges *et al.*[9] have reported a large drop in the resistance of polycrystalline $YBa_2Cu_3O_7$ with the application of 6.3-kbar hydrostatic pressure. Although the polycrystalline resistance is most probably dominated by ab-plane resistivity, Borges *et al.* report that their measured effect is probably not an intrinsic crystal property, but is due rather to a change in the geometry of their sample with pressure. Another possibility is that polycrystalline resistance may change as grain boundaries are pushed together with the sample.

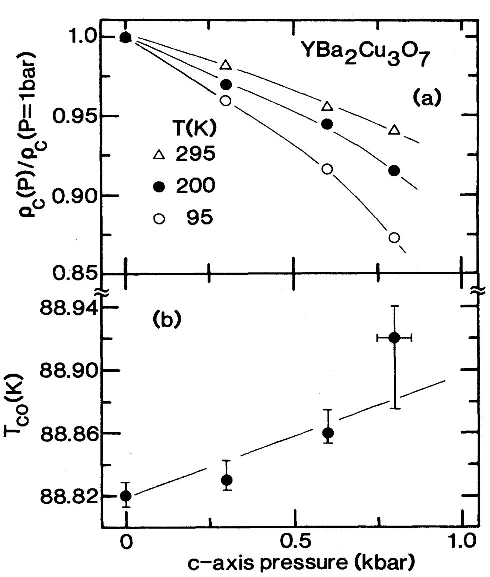

FIG. 2. (a) c-axis resistance vs pressure for selected temperatures in $YBa_2Cu_3O_7$. Solid lines are guides to the eye. (b) Superconducting onset temperature vs c-axis pressure in $YBa_2Cu_3O_7$.

The c-axis stress dependence of T_c was determined from the c-axis resistivity data (since T_c is a scalar, it makes no difference from which component of the resistivity tensor it is determined). Figure 2(b) shows the superconductivity onset temperature T_{c0} at selected pressures for the same crystal that was used for Figs. 1 and 2(a). T_{c0} was determined by the intersection of linear fits to the $R(T)$ curve just above and below the transition onset. Figure 2(b) shows that T_{c0} is sensitive to c-axis pressure, and hence interplane coupling. T_{c0} increases smoothly with increasing pressure; the solid line represents $dT_{c0}/dP=0.08$ K/kbar. Although there is evidence in Fig. 2(b) that dT_{c0}/dP is nonlinear and increases strongly near 1 kbar, experimental data at higher c-axis pressure is needed to confirm this possibility. Our data suggest that in the range 1 bar to 1 kbar, dT_{c0}/dP is between 0.03 and 0.1 K/kbar. Although no isotropic pressure studies have been reported for single-crystal $YBa_2Cu_3O_7$, polycrystalline measurements[9–11] indicate $dT_c/dP \approx 0.05$–0.09 K/kbar up to 170 kbar; this pressure dependence is of the same order of magnitude as observed in the present c-axis pressure studies.

The agreement between the polycrystalline results and the present single-crystal uniaxial stress results may be fortuitous because of filamentary superconductivity[12] and intergrain effects in polycrystalline samples. If these complications are ignored, then our findings would suggest that the c-axis pressure dependence is dominant in the polycrystalline results.

Since T_c changes when the distance between the planes is reduced, interplanar coupling is an important factor in the determination of T_c. Purely two-dimensional mechanisms[13] for superconductivity which ignore interactions between layers are likely to be inadequate. The role of interplanar coupling found here is consistent with recent observations on the Bi- and Tl-based[5] high-temperature superconductors, in which T_c is found to be a function of the number of Cu-O planes in the unit cell. Quantitatively, the measured pressure coefficient of T_c in $YBa_2Cu_3O_7$ (≈ 0.08 K/kbar) is quite small compared to that found in the La-based high-T_c materials. It is possible to account for this relatively small shift in T_c within the BCS theory without having to assume unreasonable values for unknown parameters. Indeed, Driessen *et al.*[11] have found that by using $\gamma \approx 2.5$ for the Gruneisen constant and $d\ln\lambda/d\ln V \approx 3$ within the BCS theory, they obtain the observed pressure coefficient of T_c in $YBa_2Cu_3O_7$.

The observed effect of pressure on the c-axis normal-state resistivity is dramatic. Using a Young's modulus of 2×10^3 kbar (Ref. 11), we estimate that the decrease in the separation between planes is only about 0.05%/kbar while the change in resistivity is measured to be over 16%/kbar at $T=95$ K. Since the details of the normal-state c-axis conduction mechanism are unclear, we have considered several aspects of transport theory. One simple interpretation of the pressure dependence of the c-axis resistivity is that an increase in the transfer integral between planes is responsible for the decrease in resistivity. From considerations of anisotropic transport,[14] the c-axis conductivity is roughly proportional to $|t_\perp|^2$, where $t_\perp$ is the matrix element for interplanar charge transfer. Assuming that the primary source for the measured pressure dependence of σ_c is the variation of $t_\perp$ under pressure, we estimate that $d\ln|t_\perp|/dP=\frac{1}{2}d\ln\sigma_c/dP=+0.08$/kbar at 95 K.

Both the qualitative temperature dependence and the dramatic volume dependence ($d\ln\rho_c/d\ln V\approx 150$ to 300) of ρ_c found in this study are suggestive of semiconducting behavior. However, the data are poorly fitted by the standard semiconductor model where $\rho(T)\sim e^{\varepsilon/kT}$ or $T^{-3/2}e^{\varepsilon/kT}$. Over the limited temperature range, 90 K $< T <$ 150 K, a fit to the exponential yields a small value of $\varepsilon\approx 14$ meV. The high temperature and pressure dependences of the resistivity do not follow typical intrinsic semiconductor behavior. This lends some uncertainty to the question of whether the standard band theory approach to transport properties is applicable to the c-axis conduction in $YBa_2Cu_3O_7$. Allen, Pickett, and Krakauer[15] have made theoretical predictions for the resistivity and thermoelectric power for $YBa_2Cu_3O_7$ using band theory and the Boltzmann transport equations. While some agreement is found with experiment, there are several discrepancies between the band theory predictions and experimental measurements,[2] suggesting the possibility of nonband transport mechanisms.

Phillips[16] has suggested that the defect structure found in the high-T_c materials plays an important role in determining their properties. By considering the effect of the defects, he is able to account for some of the superconducting and normal-state properties of these materials. If the defects such as oxygen vacancies are important, then it is not unreasonable to interpret the c-axis conduction in terms of hopping conduction as found in doped or amorphous semiconductors.[17] In fact, the small activation energies found in the c-axis resistivity are similar to those observed for amorphous semiconductors.

Using a model which involves a hybrid between amorphous semiconductor and metallic conductivity, we find a good fit of the normal-state c-axis resistivity to

$$\rho(T)\sim T^{\alpha}e^{\varepsilon/kT}, \quad (1)$$

where α is in the range of 0.5 to 1.0. The upper two curves in Fig. 3 show our c-axis resistance data taken at two different pressures plotted as $\ln(R/T^{\alpha})$ vs $1/T$ for $\alpha=0.7$. At both pressures, a good fit is obtained over the temperature range between T_c and room temperature. Also shown in Fig. 3 is the c-axis resistivity data of Tozer *et al.*[1] Equation (1) appears to describe their resistivity accurately as well with $\alpha=0.7$. The activation energy ε extracted from the plots of our data is about 22 meV at ambient pressures. This value is larger than the value found from our fitting the data to the standard semiconductor model, and it decreases with pressure according to $d\varepsilon/dP\approx -0.75$ meV/kbar. One physical interpretation of Eq. (1) is that the exponential term arises from activated behavior similar to the conductivity in amorphous semiconductors, while the T^{α} term comes from the temperature dependence of the mobility, and hence the scattering time τ. Possible sources of the temperature dependence of the mobility are the phonon occupation number and the average carrier velocity. The measured pressure dependence of the resistivity can be attributed to pressure dependences in both the activation energy and the mobility.

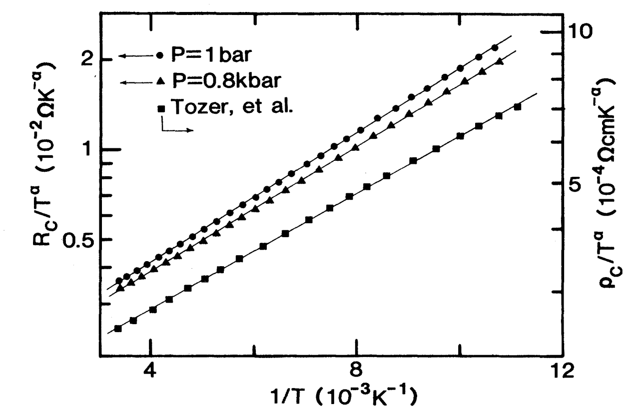

FIG. 3. Ratio of c-axis resistance to T^{α} ($\alpha=0.7$) vs $1/T$ at ambient pressure and at $P=0.8$ kbar (this work). Also shown are the c-axis resistivity data from Ref. 1.

We have also considered the possible role of localization of the carriers in the c-axis transport properties. However, the data do not fit the standard strong localization formula where $\ln\rho(T)\sim T^{-1/4}$.[17] We also note that no transverse magnetoresistance is observed in the c-axis conduction at $T=95$ K for fields up to 1 kG at ambient pressures.

One of the more novel proposals for the c-axis conduction in $YBa_2Cu_3O_7$ is given by the model of Anderson and Zou[18] in which hole solitons tunnel between layers in the resonating-valence-bond state. Although the $1/T$ dependence for the c-axis resistivity predicted by this model is consistent with some previous resistivity measurements over a limited temperature range,[1-3] we are unable to get a good fit to our data at temperatures below 150 K.

In conclusion, the c-axis stress and temperature dependence of the normal-state c-axis conduction in $YBa_2Cu_3O_7$ is inconsistent with band transport and other proposed mechanisms, but is well described by the empirical expression $\rho(T)\sim T^{\alpha}e^{\varepsilon/kT}$ with $\alpha\approx 0.7$ and $\varepsilon\approx 22$ meV. The c-axis stress dependence of T_c shows a strong sensitivity of T_c to interplane coupling, and rules out strictly two-dimensional superconductivity.

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