Effect of Uniaxial Stress on the Superconducting Transition in $YBa_2Cu_3O_7$

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We present the first direct measurements of T_c as a function of uniaxial stress p_i ($i=a,b,c$) along the crystal axes for an untwinned $YBa_2Cu_3O_{7-\delta}$ single crystal. T_c is linear in p_i with slopes $dT_c/dp_a = -2.0 \pm 0.2$ K/GPa, $dT_c/dp_b = +1.9 \pm 0.2$ K/GPa, and $dT_c/dp_c = -0.3 \pm 0.1$ K/GPa. These results confirm predictions based on thermal expansion measurements [C. Meingast *et al.*, Phys. Rev. Lett. **67**, 1634 (1991)] and show that the small hydrostatic pressure dependence of T_c in $YBa_2Cu_3O_{7-\delta}$ is due to a cancellation of large and opposite effects in the **a**-**b** plane.

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The pressure dependence of the superconducting transition temperature T_c is an important quantity for optimization of the material as well as for investigation of the nature of the superconducting state. A strong pressure dependence indicates that the material is capable of reaching higher values of T_c at ambient pressure, for example by doping in order to change the electronic structure or to invoke "chemical" pressure [1]. Pressure is also a "clean" and controlled means to change the lattice parameters of the system and check theoretical predictions.

Pressure effects in the $YBa_2Cu_3O_{7-\delta}$ superconductors have been studied extensively [1,2]. Most of the experiments as well as theoretical calculations are based on hydrostatic compressions. However, because of the large intrinsic anisotropy of the material, uniaxial deformations are required [3] in order to investigate the importance of charge transfer into the CuO_2 planes, which is sensitive to **c**-axis compression, and properties of the superconducting CuO_2 planes, most sensitive to **a**,**b**-axis compression. We note that in the case of heavy-fermion systems the coupling to uniaxial deformations has been important in revealing the symmetry of the superconducting order parameter [4].

Most previous reports [5-8] have obtained information on the effects of uniaxial stress in indirect ways: from discontinuities in high-resolution thermal expansion [5], from hydrostatic pressure measurements on supported films [6] grown with different orientations, from changes produced by deforming the substrate [7], and from measurements on oriented crystallites in an epoxy medium [8]. Although these experiments clearly show the importance of the uniaxial components of the stress, quantitative evaluation of the results usually involves some assumptions. Crommie *et al.* [9] have presented the pressure dependence of T_c for compression along the **c** axis from resistive transitions.

Here we report the first direct measurements of the changes in T_c induced by uniaxial stresses along the **a**, **b**, and **c** axes of an untwinned single crystal of $YBa_2Cu_3O_{7-\delta}$. We find that the pressure derivatives dT_c/dp_i ($i=a,b,c$) are large and opposite in sign for compression in the **a**-**b** plane: $dT_c/dp_a = -2.0 \pm 0.2$ K/GPa, $dT_c/dp_b = +1.9 \pm 0.2$ K/GPa, whereas for **c**-axis compression $dT_c/dp_c = -0.3 \pm 0.1$ K/GPa. These results show that the small hydrostatic pressure dependence of T_c is caused by a cancellation of large and opposite effects and that the "optimal" T_c for $YBa_2Cu_3O_{7-\delta}$ has *not* yet been reached.

All our measurements were performed on the same crystal, thereby eliminating any effects due to variations in oxygen content or other sample characteristics. The crystal was grown in a gold crucible using a self-flux method. This growth technique is known to introduce gold impurities predominantly on the chain copper sites [10]. The crystal for this study was detwinned as described in Ref. [11] and had $T_c = 91.5$ K and a transition width of about 0.5 K. The sample was quite close to being a perfect parallelepiped with dimensions $L_a \times L_b \times L_c = 0.76 \times 0.78 \times 0.30$ mm^3, but some small chips along the edges led to uncertainties in the cross-sectional areas of the order of 6%. T_c was detected by ac susceptibility measured at a frequency of 103 Hz. The sample and pickup coils were placed between a fixed and a mobile steel piston 5 mm in diameter. Force on the mobile piston was applied by means of a bellows pressurized by He gas (see inset of Fig. 1). This setup allows pressure changes while the sample is at low temperatures, thus reducing the probability of pressure-induced oxygen diffusion.

Figure 1 shows the temperature dependence of the in- and out-of-phase signals, χ' and χ'', respectively, at zero pressure. We observe the familiar step in χ' and the peak in χ'' near T_c. The large background signal is due to eddy currents in the metal pistons. The sharpest feature in these scans is the peak in the out-of-phase signal and we have used it to follow the changes in T_c as a function of pressure.

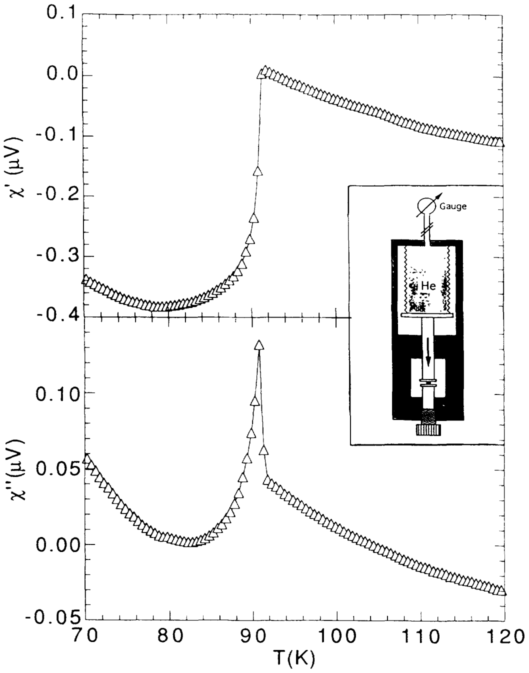

FIG. 1. In-phase (top) and out-of-phase (bottom) signal of the ac susceptibility of an untwinned $YBa_2Cu_3O_{7-\delta}$ single crystal at zero pressure. The temperature-dependent background is due to the steel pistons. Inset: Schematic of the uniaxial pressure cell.

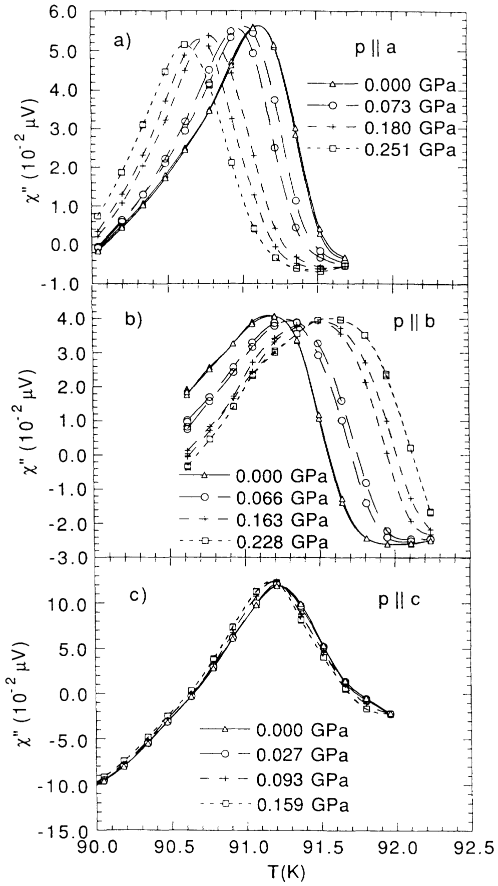

FIG. 2. Temperature and pressure dependence of the out-of-phase signal. Parts (a), (b), and (c) correspond to pressure along the **a**, **b**, and **c** axes, respectively. The lines are cubic-spline fits to the data points and the two curves for each pressure represent the data while increasing and decreasing the pressure.

Although plots like those in Fig. 1 appear to be ideal for determining the pressure dependence of T_c, we observed thermal hysteresis of up to 0.25 K between warming and cooling runs and between several runs at the same pressure. Therefore, we chose to stabilize the temperature and measure χ' and χ'' as a function of increasing and decreasing pressure at each temperature point. The χ'' results of these runs for pressures along the three principal axes are shown in Fig. 2. The different absolute values of χ'' for the three orientations are due to the different aspect ratios of the sample. The superconducting transition width does not increase significantly with increasing pressure, indicating that the pressure is approximately homogeneous in the sample. This observation also shows that our final results for dT_c/dp_i $(i=a,b,c)$ are independent of the definition of T_c. There is some hysteresis between the results for increasing and decreasing pressure, the largest value of about 75 mK occurring at 0.18 GPa, along the **a** axis. This induces a small uncertainty of about ±38 mK (in the worst case) into the determination of $T_c(p_i)$.

Our results are summarized in Fig. 3, which shows the measured changes in T_c as a function of pressure for the **a**, **b**, and **c** axes. The values of $T_c(p)$ shown in the figure were obtained by averaging the measurements for increasing and decreasing pressure. Within the error bars, given by the height of the symbols, the pressure dependencies are linear. The slopes for pressure along the **a**, **b**, and **c** axes are -2.0 ± 0.2, $+1.9\pm0.2$, and -0.3 ± 0.1 K/GPa, respectively, where the quoted errors include the uncertainty in the sample cross section. For p along the **a** axis, the results of three runs, one after remounting the sample, are shown, indicating the good reproducibility of the results. We note that the value at 0.18 GPa corresponds in two runs to the highest achieved pressure, whereas it corresponds in the last run to an intermediate pressure accompanied by the pressure hysteresis [see Fig. 2(a)]. The good agreement between the results shows that the pressure hysteresis does not falsify our estimates of dT_c/dp_i.

From the uniaxial pressure dependencies of T_c the hydrostatic pressure dependence dT_c/dp can be calculated according to $dT_c/dp=\sum_i dT_c/dp_i$, yielding $dT_c/dp=-0.4$ K/GPa. The hydrostatic pressure dependence of T_c has been studied extensively [1,2,12] and is found to be very sensitive to the oxygen content of the sample. For fully oxygenated samples dT_c/dp is small and negative, whereas a slight oxygen deficiency induces a sign change

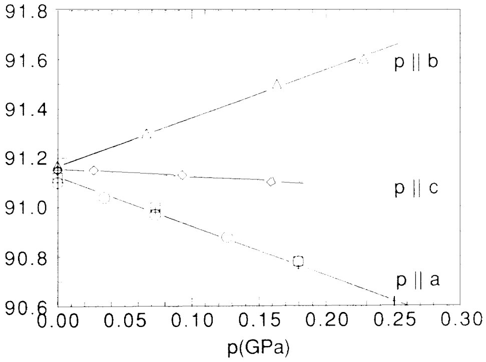

FIG. 3. Change in T_c as a function of pressure along the three axes. For p along the **a** axis the different symbols correspond to three different runs.

to positive values of dT_c/dp [13,14]. This indicates that our sample is "fully" oxygenated.

For a second-order phase transition the pressure dependence of the transition temperature is related to the anomalies in the specific heat and thermal expansion through the Ehrenfest relation [15]: $dT_c/dp_i = VT_c[(\alpha_{i,s} - \alpha_{i,n})/(C_s - C_n)]$, where $\alpha_i = (1/L_i)\,dL_i/dT$ are the expansivities along $i = a,b,c$, $C_s - C_n$ is the difference of the normal-state and superconducting-state heat capacities at T_c, and $V = L_a \times L_b \times L_c$ is the volume of the sample. At a second-order phase transition which is characterized by strong fluctuation effects, the specific heat and the thermal expansion are continuous at T_c and higher-order derivatives have to be employed in order to determine dT_c/dp_i. Therefore, recent high-resolution thermal expansion measurements [5] have been analyzed by extracting the mean-field part of the anomalies, yielding for the pressure derivatives -1.9, $+2.2$, and 0 K/GPa along **a**, **b**, and **c**, respectively. These values are in very good agreement with our determinations, indicating that the underlying mean-field contributions are well known. The slight discrepancy for the **c**-axis dependence, 0 K/GPa instead of -0.3 K/GPa, can be explained (see above) by a lower oxygen content of the crystal used for thermal expansion. This is also consistent with the lower value of T_c in their sample.

As a result of the Poisson effect (represented by the off-diagonal elastic moduli c_{12}, c_{13}, c_{23}), in a uniaxial pressure experiment a mixture of compression along the pressure direction and dilatation along the two perpendicular directions is generated. The pure strain (deformation) dependence $dT_c/d\varepsilon_i$ of T_c can be calculated from the uniaxial pressure dependence and the elastic moduli c_{ji} according to $dT_c/d\varepsilon_i = -\sum_j c_{ji} dT_c/dp_j$, where $\varepsilon_1 = \varepsilon_{aa}$, $\varepsilon_2 = \varepsilon_{bb}$, and $\varepsilon_3 = \varepsilon_{cc}$ denote deformations along **a**, **b**, and **c**, respectively (note that pressure corresponds to a negative stress). Using recent determinations [16] of the elastic moduli of untwinned $YBa_2Cu_3O_{7-\delta}$ we obtain $dT_c/d\varepsilon_1 = +230$ K, $dT_c/d\varepsilon_2 = -220$ K, and $dT_c/d\varepsilon_3 = +18$ K. Therefore, also for the pure deformations, we obtain the intriguing result that the effects in the **a**-**b** plane on T_c are large and opposite in sign, whereas for **c**-axis deformation T_c decreases only slightly. Thus, contrary to the trends observed upon oxygen removal, the pressure dependence of T_c shows that tetragonality enhances superconductivity. This has already been inferred from the decrease of the orthorhombicity at T_c as seen in thermal expansion [5]. The difference between our results for the strain derivatives and those obtained in Ref. [5] can be traced to their use of elastic constants appropriate for a twinned or tetragonal structure.

It is commonly accepted that for a given class of materials, i.e., $YBa_2Cu_3O_x$ or $La_{2-x}Sr_xCuO_4$, the value of T_c is determined by the number n_h of charge carriers in the CuO_2 planes. Through a charge-transfer mechanism n_h (and therefore T_c) can be changed by doping the structural elements neighboring the CuO_2 planes or by changing the atomic positions by applying pressure. There is an optimal value of n_h corresponding to the highest value of T_c. A further increase of the doping level leads to a rapid decrease in T_c, either due to "overdoping" [17] or chemical instability and phase segregation [18]. This T_c variation has been studied extensively for $La_{2-x}Sr_xCuO_4$ and recent experiments [19] on $YBa_2Cu_3O_{7-\delta}$ also indicate that T_c goes through a maximum as δ approaches zero.

Whereas oxygen-deficient $YBa_2Cu_3O_{7-\delta}$ ($T_c \approx 60$ K) shows large positive hydrostatic pressure derivatives, the 90-K compound is characterized by a small hydrostatic pressure dependence of T_c. High-resolution neutron scattering experiments [20] on 60- and 90-K material together with a bond valence sum analysis indicated that the pressure-induced charge transfer for both materials is the same and that T_c for the 90-K material does not increase any further with the application of pressure because the optimal charge transfer has already occurred at ambient pressure. The results presented here, however, show that there are strong, competing pressure effects in the 90-K material which almost cancel each other in a hydrostatic experiment. This implies that T_c in the 90-K compound is *not* yet at its optimal value. In fact, if epitaxial films can be grown on substrates with compressive and tensile lattice mismatch for **a** and **b**, respectively, T_c values around 100 K seem possible at zero applied pressure. For oxygen-deficient $YBa_2Cu_3O_{7-\delta}$, which shows a large hydrostatic pressure coefficient, T_c values (determined from the resistive onset) around 100 K under large hydrostatic pressures have been reported [21].

It has been argued that the position of the apical oxygen atoms relative to the CuO_2 planes is important for the charge distribution in the unit cell [22]. However, if this were the dominant effect it seems hard to explain that the pure **a**-axis and **b**-axis compressions (distances along the **c** axis do not change with these deformations) yield large and opposite pressure effects, whereas the pure **c**-axis compression influences T_c only slightly. Electronic structure calculations [23] indicate that T_c is almost pressure independent because there is essentially no net charge transfer in a hydrostatic pressure experiment. Further calculations as well as structure determinations under uniaxial compression are needed to determine how the atomic positions and the electronic bands rearrange in order to yield the large and opposite effects for uniaxial compression along **a** and **b**, and whether they are just a result of an accidental combination of plane- and chain-derived effects.

In the above discussion the deformation dependence of the charge transfer between the CuO_2 planes and the CuO chain layers is assumed to be the important mechanism. However, it has been suggested [3] that the result $|dT_c/d\varepsilon_1| \approx |dT_c/d\varepsilon_2| \gg |dT_c/d\varepsilon_3|$ indicates that the pressure dependence of T_c is a property of the CuO_2 planes and that the relative sign for the **a**- and **b**-axis compression may reflect the symmetry of the superconducting order parameter itself. An alternative explanation for these results may be the recently proposed [24] coupling of superconductivity to a shear deformation along ⟨110⟩. This coupling could be related to the correlated motion of the O(2)-O(3)-O(4) pyramids around $[1\bar{1}0]$ as inferred from recent inelastic neutron scattering experiments [25]. Thermal expansion measurements [26] indicate that the **a**-**b** anisotropy of the pressure derivatives persists in oxygen-deficient $YBa_2Cu_3O_{7-\delta}$ crystals, whereas the hydrostatic pressure dependence is dominated by the large positive **c**-axis component. Uniaxial pressure experiments on $La_{2-x}Sr_xCuO_4$, which does not show complications due to the CuO chains, are under way in order to further clarify these questions.

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