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Anisotropic pressure dependence of the resistivity in $YBa_2Cu_4O_8$

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The anisotropic pressure dependence of the resistivity of $YBa_2Cu_4O_8$ has been determined for the two plane directions a and b. The resistivity in a direction, ρ_a, reflecting CuO_2 planes, revealed a strong pressure dependence $d\ \ln\rho_a/dp=13\times10^{-3}$ kbar^{-1}. In the stiffer b direction with two conduction channels, along CuO chains and CuO_2 planes, a smaller decrease upon pressure but the strongest change with respect to the lattice contraction was found. The pressure dependence observed can be explained by a charge transfer from chains to planes.

The discovery of the ceramic high-temperature superconductors (HTSC) by Bednorz and Müller[1] has not only enhanced the critical temperature T_c than also revealed unusual normal-state properties such as a linear resistivity in function of temperature, an opening of a spin gap above T_c in underdoped substances, etc. Another amazing feature of the HTSC's is the strong pressure dependence[2] of T_c. Conventional BCS superconductors have a pressure dependence of T_c in the order of $dT_c/dp=$ 0.01 K/kbar.[3] In contrast we have found $dT_c/dp=0.56$ K/kbar in $YBa_2Cu_4O_8$ (Ref. 4) together with the high $T_c=80$ K. Also the resistivity of the HTSC's shows a strong pressure dependence. For metals, relative pressure derivatives $d\ln\rho/dp$ in the order of 0.001 kbar^{-1} have been reported.[5] As we show below HTSC's reveal a value of $d\ln\rho/dp\approx$ -0.013 kbar^{-1}. In this paper we address the nature of the strong pressure dependence of the resistivity and study its correlation with the increase of T_c upon applied pressure.

$YBa_2Cu_4O_8$ (1:2:4) has the same structure as $YBa_2Cu_3O_7$ (1:2:3) except the single CuO chain along the b direction which is replaced by a double CuO chain. Oxygen atoms in the double CuO chain are higher coordinated giving rise to an enhanced stability of the oxygen stoichiometry. In fact, it is hardly possible to extract any oxygen out of 1:2:4 below the melting point of the substance.[6] Because of that 1:2:4 is genuinely untwinned as it never undergoes the tetragonal orthorhombic phase transition which is responsible for the twinning in 1:2:3. Untwinned 1:2:3 which could be prepared by annealing under uniaxial stress is only metastable, preventing the study of the anisotropic components under pressure.

As the mechanism responsible for superconductivity is concealed by the excess carrier density in highly doped HTSC's, modestly doped samples should be preferred for unraveling the basic physics. A fingerprint of underdoped HTSC's is an opening of a spin gap above T_c and a magnetic susceptibility increasing with temperature. Both signatures have been found in 1:2:4 by means of the spin Knight shift,[7] the spin-lattice relaxation time T_1,[8] and the resistivity.[9] Unfortunately, with the exception of 1:2:4, underdoped samples are nonstoichiometric compounds and, therefore, contaminated with vacancies, lattice distortions, etc.

The response of the double CuO_2 planes contains the basic information about superconductivity. Any influence of the other block layers (e.g., BiO or HgO sheets) on the experiment should be discriminated. The crystal structure of 1:2:4 does not contain block layers. Anisotropic measurements along the a direction of 1:2:4 (perpendicular to CuO chain) reveal the response of the superconducting double CuO_2 planes alone. Hence 1:2:4 makes it possible to study a simultaneously stoichiometric, underdoped, and genuinely untwinned HTSC at applied pressure.

We present the anisotropic pressure dependence of the resistivity components along the two plane directions a and b of $YBa_2Cu_4O_8$. The a direction shows a strong decrease of ρ_a under pressure. For the b direction we have found a smaller decrease of the absolute value of the resistivity but the strongest change with respect to the compression of the lattice constant.

Single crystalline $YBa_2Cu_4O_8$ was grown at 900 bars O_2 and 1050 °C, using a BaO-CuO-rich flux. The dimensions of the untwinned single crystals were about $0.5\times0.5\times0.1$ mm^3. Gold pads had been sputter deposited using photolithographically produced masks. After an annealing at 500 °C for two hours, gold wires were attached by gold epoxy. The contact resistance was in the order of 1 Ω.

For the high-pressure experiments at room temperature, a piston cylinder equipment was used. The sample was inserted into a Teflon cell filled with the pressure transmitting medium n-pentane – isoamylalcohol. Pieces of pyrophylite were used as a gasket material. The pressure was determined *in situ* by means of a manganin coil.

In Fig. 1, we show the normalized, anisotropic pressure dependence of the resistivity of the two plane directions a and b at 300 K. A change of the measured sample resistance upon pressure can be assigned principally to two different effects. First, the length and area of the sample are compressed changing the geometry of the sample. The second effect is the intrinsic change of the resistivity due to, e.g., a changed scattering rate or an enhanced carrier density.[10] As we are interested in the intrinsic change of the resistivity, $\rho(p)$, of 1:2:4 upon applied pressure we have to compensate the influence of the volume compression on the sample resistance. A first-order correction for an orthorhombic crystal structure is given by

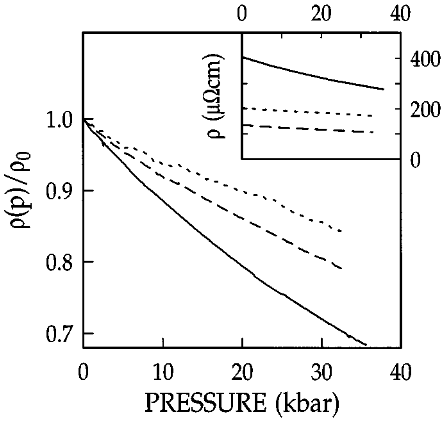

FIG. 1. Pressure dependence of the normalized resistivity of $YBa_2Cu_4O_8$ at 300 K for the two plane directions: ρ_a (solid line); ρ_b (dashed line). The resistivities are volume corrected due to Eq. (1). Also depicted is the calculated [Eq. (2)] resistivity of the CuO chain alone (dotted line). The inset shows the absolute change of resistivity.

$$\frac{\rho(p)}{\rho_0}=\frac{R(p)}{R_0}[1+(\beta_1^{\perp}+\beta_2^{\perp}-\beta^{\parallel})p] , \tag{1}$$

where $\beta_i^{\perp}$ and $\beta^{\parallel}$ are the coefficients of linear compressibility for the directions perpendicular and parallel to the current, respectively. $R(p)$ is the experimental resistance as a function of pressure. R_0 and ρ_0 are the resistance and resistivity at zero pressure, respectively. The data in Fig. 1 have been corrected using the linear compressibitities listed in Table I.

The change of resistance is not linear with applied pressure. In the b direction two parallel conduction channels are present: CuO_2 planes and the CuO chains. Exploiting the model of two shunted conduction channels for the current I $\parallel$ b axis, one can extract the pressure dependence of the resistivity of CuO chains alone

$$\frac{1}{\rho_{\text{chain}}}=\frac{1}{\rho_b}-\frac{1}{\rho_{\text{plane}}} \tag{2}$$

TABLE I. Relative changes of anisotropic resistivity ρ and lattice constants l under hydrostatic pressure of $YBa_2Cu_4O_8$.

	$\frac{d\ln\rho^{a}}{dp}$ (10^{-3} kbar^{-1})	β^{b} (10^{-4} kbar^{-1})	$\frac{d\ln\rho}{d\ln l}$
a direction	−13	−2.8	4.6
b direction	−10	-1.0^{c}	10
c direction		−4.8	
Chain	−7.5	−1.0	7.5

[a] Derivative at the pressure p=0 kbar.

[b] The linear compressibility β is defined as $d\ln l/dp$. Data from Ref. 11.

[c] The linear compressibility represents a combined effect of the stiffness of the CuO chain plus CuO_2 plane.

making use of the relation $\rho_{\text{plane}}=\rho_a$. Within this model the *temperature dependence* of $\rho_{\text{chain}}(T)$ at ambient pressure has been shown[12] to be quadratic, i.e., $\rho_{\text{chain}}=AT^2$ with the constant $A=2.3\times10^{-3}\mu\Omega$ cm/K^2. The calculated *pressure dependence* of ρ_{chain} shows a nearly linear behavior in function of pressure, implying a rate of decrease for the constant A of $dA/dp=-1.1\times10^{-5}\mu\Omega$cm/K^2 kbar. The relative change of $\rho(p)$ with respect to the reduced lattice constant is a factor of 2 higher for the b than for the a direction (see Table I). In the inset to Fig. 1 we plotted the absolute change of the resistivities.

There are several approaches to explain the change of resistivities under pressure. Assuming that electron-phonon scattering does prevail, the resistivity decreases upon high pressure because the Debye temperature becomes enhanced with the hardening of the phonon frequencies. Near and above the Debye temperature Θ_D one finds $\rho\propto T/\Theta_D^2$. The change of the resistivity under pressure can then be expressed as

$$\frac{d\ln\rho}{d\ln V}=-2\frac{d\ln\Theta_D}{d\ln V}=-2\gamma^{p} \tag{3}$$

with γ^{p} the Grüneisen constant. With a change of the average resistivity of $d\ln\rho/d\ln V\approx$ 10 one finds $\gamma^{p}\approx5$. An independent determination of γ is possible on the basis of elastic and thermodynamic quantities. Thermodynamics defines $\gamma^{\text{th}}=\alpha_V B V_m/C_p$ with α_V the thermal expansion coefficient, C_p the specific heat, B the bulk modulus, and V_m the molar volume. With the data for $YBa_2Cu_4O_8$,[13] the Grüneisen constant γ^{th} takes on the value 1.65. The discrepancy between the thermodynamic Grüneisen constant ($\gamma^{\text{th}}=1.65$) and the value deduced from the high-pressure experiment ($\gamma^{p}\approx5$) seems significant.

Another approach starts with the idea that the carrier density or doping level of CuO_2 planes changes with applied pressure. For the resistivity we set

$$\rho\propto\frac{m^*}{n\tau} \tag{4}$$

with m^* the effective mass, n the carrier density, and τ the relaxation time. The pressure dependence of the plane resistivity ρ_a is then given as

$$\frac{d\ln\rho_a}{dp}=\frac{d\ln m^*}{dp}-\frac{d\ln n}{dp}-\frac{d\ln\tau}{dp}. \tag{5}$$

Zimmermann *et al.*[14] determined experimentally the absolute charge transfer from CuO chains to CuO_2 planes by measuring the shift of the Cu nuclear quadrupole resonance in 1:2:4 under pressure. They extracted $d\ln n/dp\approx10\times10^{-3}$ kbar^{-1}. The density of free carriers at zero pressure was set as 0.1 carrier per plane Cu(2).[15] Scholtz *et al.*[16] deduced the value 9×10^{-3} kbar^{-1} from their measurements of critical temperature T_c and upper critical field H_{c2} under pressure.

In Eq. 5, the change of the carrier density alone accounts for $d\ln\rho_a/dp\approx-10\times10^{-3}$ kbar^{-1}[Eq. (5)] of the experimental value -13×10^{-3} kbar^{-1}. The incorporation of an enhanced relaxation time may explain the small difference: the relaxation time is mainly of a magnetic nature[9] and it is reasonable to assume that the increased carrier density screens more effectively the spin fluctuations.

One can test if the model of charge transfer also explains the pressure dependence of T_c. It is well known that the critical temperature T_c of the HTSC's is governed by the concentration of doped carriers in CuO_2 planes. The functional relation of T_c and doping level can roughly be approximated by the parabolic equation

$$T_c(x,p) = T_{co} - \alpha[n(x,p) - n_0]^2 \tag{6}$$

and

$$\frac{dT_c}{dp} = -2\alpha[n(x,p) - n_0]\frac{dn(x,p)}{dp}, \tag{7}$$

where $n(x,p)$ is the number of carrier density per CuO_2 plane, n_0 is the doping level with the maximum T_c, x a chemical stoichiometry parameter, and p the external pressure. For 1:2:4 one can extract[17] the parameters $n_0 = 0.17$, $T_{co} = 92$ K, and $\alpha = 2300$. Comparing to equivalently underdoped 1:2:3, the carrier concentration of 1:2:4 can be assigned to $n = 0.1$. The resistivity data above suggest the charge transfer $dn/dp \approx 0.001$ carrier/kbar. Using Eq. (7) the calculated pressure dependence, dT_c/dp, gives 0.32 K/kbar. The experimental value $dT_c/dp = 0.56$ K/kbar is a factor 1.75 higher.

The concept of a charge transfer or better the absence of a charge transfer is also appropriate for the electron-doped substances. $R_{2-x}Ce_xCuO_{4-y}$ (R = Pr, Nd, and Sm) has no apical oxygen over which the charge transfer is expected to take place.[18] The in-plane resistivity shows indeed only a modest pressure dependence[19] as does the pressure dependence of T_c. However Tozer *et al.*[20] have shown that more subtle effects than the charge transfer become crucial in the YBaCuO family on extending the pressure range to several tens of kbar.

In summary we presented anisotropic measurements of the resistivity tensor of the two plane directions of $YBa_2Cu_4O_8$ upon pressure. The superconducting CuO_2 plane, i.e., ρ_a revealed a strong decrease of the resistivity as pressure is increased. The change of resistivity is an order of magnitude higher than for a conventional metal. For the b direction the absolute decrease is smaller but the relative change with respect to the compression of the lattice constant is the strongest one.

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[3] S.V. Vonsovsky, Yu.A. Izyumov, and E.Z. Kurmaev, in *Superconductivity in Transition Metals*, edited by M. Cardona, Springer Series in Solid State Sciences, Vol. 27 (Springer, Heidelberg, 1982). Lanthanum has been regarded as an exotic substance because of the high $dT_c/dp = 0.1$ K/kbar.

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[8] H. Zimmermann *et al.*, Physica C **159**, 681 (1989).

[9] B. Bucher *et al.*, Phys. Rev. Lett. **70**, 2012 (1993).

[10] In semiconductors, the decrease of the resistivity under pressure is mainly due to an exponential enhancement of the carrier density as the gap closes linearly. In conventional metals, the Debye temperature Θ_D, i.e., the average phonon frequency, increases under pressure and, therefore, a reduced scattering results since the phonons are less thermally populated.

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[13] The bulk modulus is 1120 kbar (Ref. 11); the thermal expansitivities α are taken from Alexandrov *et al.*, Physica C **172**, 491 (1991); the specific heat at 300 K is 22 J/g at. K [Junod *et al.*, Physica C **168**, 47 (1990)].

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[15] A carrier density of 0.1 reflects the underdoped regime of 1:2:4. It is comparable to the value of an underdoped 1:2:3 which shows similar physical characteristics due to the low doping level.

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[18] The carrier concentration can be adjusted only by the stoichiometry, i.e., x and y.

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