Anisotropic Pressure Dependence of T_c in $YBa_2Cu_3O_{7-\delta}$: Evidence for the Tunneling-Unit Model

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The superconducting transition temperature T_c of $YBa_2Cu_3O_{7-\delta}$ shows a decrease with uniaxial pressure in the **a** direction and an approximately equal increase with pressure in the **b** direction near $T_c \approx 91.5$ K. The jump in the thermal expansivity near T_c is also approximately equal and of opposite sign in the **a** and **b** directions. We show that the recently derived tunneling-unit model for high T_c superconductivity explains the anisotropic pressure dependence of T_c and the resulting jump in the thermal expansivity near T_c in $YBa_2Cu_3O_{7-\delta}$. For $\delta \approx 0$ our results are in close quantitative agreement with measurements. We suggest further experiments to clarify the nature of high T_c superconductivity in $YBa_2Cu_3O_{7-\delta}$ for $0 \leq \delta \leq 0.5$. [S0031-9007(97)03160-8]

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The pressure dependence of the transition temperature in high T_c superconductors (HTSC's) is of a good deal of interest, for it gives information on the microscopic origin of the superconducting state. And perhaps more importantly, a strong pressure p dependence of T_c indicates that the material is capable of reaching higher values of T_c, for example, by doping in order to change the "chemical" pressure in the material [1]. While the effects of hydrostatic (isotropic) pressure in $YBa_2Cu_3O_{7-\delta}$ are interesting and have been studied extensively [1,2], a great deal of additional information can be obtained from the anisotropic behavior of T_c as a function of the uniaxial pressure. The results yield considerable insight. For example, in an untwinned single crystal of $YBa_2Cu_3O_{7-\delta}$ near "optimal" doping ($T_c \approx 91.5$ K) uniaxial pressure dependence measurements [3,4] revealed the following pressure derivatives: $dT_c/dp_a = -2.0 \pm 0.2$ K/GPa, $dT_c/dp_b = +1.9 \pm 0.2$ K/GPa; for the **c**-axis compression $dT_c/dp_c = -0.3 \pm 0.1$ K/GPa, where the subscripts ($i = a, b, c$) denote the corresponding crystallographic directions. Note that while the **c**-axis pressure derivative is small, the **a** and **b** axis derivatives are large and of opposite sign. Not only does this explain why the hydrostatic pressure dependence [1,2] of T_c in $YBa_2Cu_3O_{7-\delta}$ is small (as a result of the cancellation of the **a**-axis and **b**-axis pressure dependences [3]), but also shows that the anisotropy along the **a** and **b** directions is more dramatic than the small lattice constant anisotropy would indicate, suggesting a strong role played by the Cu-O chains in these materials. Similar anisotropic measurements were also found in the thermal expansivity [4] $\alpha \equiv d \ln L(T)/dT$ [$L(T)$ is the sample length]. The jump in α, $\Delta\alpha$, near T_c as the material goes from normal to superconducting state in a near-optimally doped $YBa_2Cu_3O_{7-\delta}$ sample has approximately the same value along the **a** axis as along the **b** axis, except that they are of opposite sign. Along the **c** axis $\Delta\alpha$ is small.

While these experimental results seem to yield important insight into $YBa_2Cu_3O_{7-\delta}$ materials, there is no microscopic theory (of which we are aware) that can explain them. Indeed, a great deal of effort to understand high T_c superconductivity has still not resulted in a generally accepted theory [5]. Yet it is clear that a successful microscopic theory of high T_c superconductivity should explain these results in addition to other properties.

In this Letter we show how a recently derived *microscopic* model [6,7], the tunneling-unit (TU) model, can explain these results and is therefore the first theoretical *microscopic* model to do so. We also propose new experiments which could separate the pressure induced charge transfer effects from the pressure dependent tunneling-unit effects for less than "optimally" oxygenated samples, thus further elucidating the cause of the HTSC in $YBa_2Cu_3O_{7-\delta}$ for $0 \leq \delta \leq 0.5$.

Our paper is organized as follows. First we give a brief review of the TU model. Then we show how the change in the microscopic width a of the TU affects T_c, and we relate this change Δa to the strain in the solid. The pressure derivatives of T_c are then expressed in terms of the pressure derivatives of the strain, which can be calculated from the elastic constants. We find that the values of dT_c/dp_i calculated in this manner are surprisingly close to those obtained experimentally. Finally we suggest experiments which could elucidate the pressure dependences in underdoped $YBa_2Cu_3O_{7-\delta}$.

In the TU model the conduction electrons are scattered by localized tunneling units (double-well or multiwell potentials for certain atoms in the crystal lattice). $YBa_2Cu_3O_{7-\delta}$ has Cu-O chains along the **b** axis. The oxygen atoms in the chains are located 0.08 Å off the chain axis and constitute two-level tunneling units [6]. Experiments show [8] that in $YBa_2Cu_3O_{7-\delta}$ the tunneling units are directed (defined by a vector from one well to the other in a two-level system) in the **a** direction [9]. The TU model gives an analytic derivation of the gap function $\Delta(\mathbf{k})$ and finds that $\Delta(\mathbf{k})$ is a linear combination of s-wave and d-wave components. Furthermore, the model gives an isotope effect consistent with experimental observations and

may also explain the s-state Josephson tunneling along the **c** axis [10] in spite of the predominantly d-wave character of $\Delta(\mathbf{k})$. The derivation of the uniaxial pressure dependence of T_c and the jump in thermal expansivity near $T_c \approx 91$ K in agreement with experiment gives additional evidence for the validity of the TU model for HTSC.

The expression for the attractive electron-electron interaction Hamiltonian, $H_{el,el}$, in the TU model is [6]

$$H_{el,el} = \sum_{\mathbf{k},\mathbf{q}} N(\mathbf{q})N(-\mathbf{q}) \frac{J_{\text{ave}}}{(\epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k}+\mathbf{q}})^2 - J_{\text{ave}}^2} \times c^*_{\mathbf{k}+\mathbf{q}} c^*_{-\mathbf{k}-\mathbf{q}} c_{-\mathbf{k}} c_{\mathbf{k}}\,, \quad (1)$$

where J_{ave} is the average strain interaction at a tunneling-unit site, $c^*_{\mathbf{k}}$ and $c_{\mathbf{k}}$ are electron creation and annihilation operators with wave vector **k** (with spin index suppressed), $\epsilon_{\mathbf{k}}$ is the energy of the conduction electron, and $N(\mathbf{q}) = N(\mathbf{k} - \mathbf{k}')$ is the coupling between electrons and tunneling units [6].

Let $\Psi_G(\mathbf{R})$ and $\Psi_E(\mathbf{R})$ be the ground-state and first-excited-state wave functions for a tunneling unit at site **R**. Then [6],

$$N_{\mathbf{q}} = \frac{V_0(\mathbf{q})}{2} \int e^{i\mathbf{q}\cdot\mathbf{R}} [\Psi^\dagger_G(\mathbf{R})\Psi_E(\mathbf{R}) + \text{H.c.}]\, d^3\mathbf{R}\,, \quad (2)$$

where $V_0(\mathbf{q})$ is the Coulomb interaction potential between electron and the tunneling atom and H.c. stands for Hermitian conjugate. The major contribution to the integral $N_{\mathbf{q}}$ comes from the oxygen on the Cu-O chain tunneling from one well to the other well on the same site [9], since the integral in Eq. (2) becomes exponentially small for wells at two different sites. The tunneling units are oriented in the **a** direction, thus the overlap of the wave functions is appreciable only in the x direction (i.e., the **a** direction of the TU's) and we use the approximation

$$\Psi^\dagger_G(\mathbf{R})\Psi_E(\mathbf{R}) = \Psi^\dagger_G(x)\Psi_E(x)\delta(y)\delta(z)\,, \quad (3)$$

where $\delta(y)$ and $\delta(z)$ are delta functions.

We simulate the tunneling unit by the potential shown in Fig. 1. As seen from Fig. 1, the states $\Psi_G(x)$ and $\Psi_E(x)$ are primarily localized in the right and left wells of the double-well potential, respectively. Let the width of the double-well potential be $2a(p)$, a function of pressure p. Let $\Psi(x; a(p))$ be the wave function corresponding to pressure p. Let $N_q(p)$ be the value of N_q under pressure p. Using Eq. (3) in Eq. (2) gives

$$N_q(p) = \frac{V_0}{2} \int_{-a(p)}^{a(p)} e^{iqx(\cos\theta - \cos\theta')} \times [\Psi^\dagger_G(x; a(p))\Psi_E(x; a(p)) + \text{H.c.}]\, dx\,, \quad (4)$$

where $\cos\theta$ and $\cos\theta'$ are the angles the **k** and **k**′ vectors make with the x axis and V_0 is the effective Coulomb potential, assumed to be a constant.

We have explicitly indicated in Eq. (4) that the wave functions and the overlap integral depend on the width of the tunneling unit $a(p)$. We assume that the wave functions scale according to the width of the double-well potential while maintaining the normalization,

$$\Psi(x; a(p)) = \left(\frac{a(0)}{a(p)}\right)^{1/2} \Psi\left(x\,\frac{a(0)}{a(p)}; a(0)\right). \quad (5)$$

We substitute Eq. (5) into Eq. (4), use $x' \equiv xa(0)/a(p)$, and finally drop the prime to find that

$$N_q(p) = \frac{V_0}{2} \int_{-a(0)}^{a(0)} \exp\left(iq\,\frac{a(p)}{a(0)}\,x(\cos\theta - \cos\theta')\right) \times [\Psi^\dagger_G(x; a(0))\Psi_E(x; a(0)) + \text{H.c.}]\, dx = N_{q'}(0), \quad (6)$$

where $q' = qa(p)/a(0)$.

We note that for $|q| = 0$, N_q is identically zero because $\Psi_G(x)$ and $\Psi_E(x)$ are orthogonal. We calculated the expression for $N_q(0)$ and we found that with a high degree of accuracy $N_q(0) \propto q(\cos\theta - \cos\theta')$, therefore

$$N_q(p) = N_{q'}(0) = \frac{a(p)}{a(0)}\, N_q(0)\,. \quad (7)$$

We next derive the pressure dependence of T_c along the different crystal axes. Let $a(p) = a(0) + \Delta a$, where Δa is the change in the width of the double-well potential as a function of pressure [11]. The transition temperature for the TU model is [6]

$$k_B T_c \approx J_{\text{ave}}\, e^{-1/N_0 V_{\text{eff}}}, \quad (8)$$

where V_{eff} is an effective potential which depends on the attractive electron-electron interaction V_2 and the Coulomb repulsion between the electrons V_1,

$$V_{\text{eff}} = V_2\left(\frac{1}{2} + \frac{V_1 + 0.75V_2}{-V_1 + \sqrt{(V_1 + V_2)^2 + 0.5V_2^2}}\right). \quad (9)$$

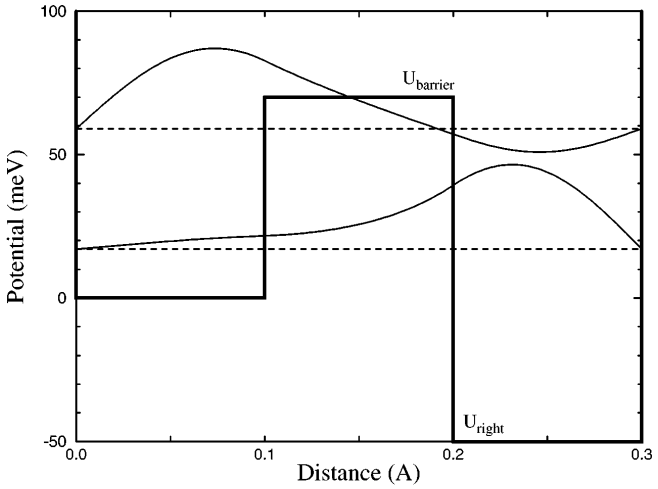

FIG. 1. A model potential for calculating the overlap integral given by Eq. (2). The figure also shows the wave functions for the two lowest energies of the tunneling unit. U_{barrier} and U_{right} are measured from the bottom of the left well, taken to be zero.

In Eq. (9) V_1 is negative, i.e., a repulsive interaction. Using Eqs. (1) and (7) we obtain that $V_2 \propto a(p)^2$. Substituting the expression for V_{eff} in Eq. (8) we obtain

$$T_c(p) \approx T_c(0)\left[1 + \frac{2}{N_0 V_{\text{eff}}}\frac{\partial \ln V_{\text{eff}}}{\partial \ln V_2}\frac{\Delta a}{a}\right] \equiv T_c(0)\left[1 + K\frac{\Delta a}{a}\right] = T_c(0) + \Delta T_c\,, \quad (10)$$

$$\Delta T_c = T_c(0)K(\Delta a/a)\,, \quad (11)$$

where $T_c(0)$ is the transition temperature at ambient pressure. We thus have $\Delta T_c \propto \Delta a/a$. Equation (11) is the central result of this paper.

We next calculate dT_c/dp_a for the uniaxial pressure in the **a** direction from the TU model using the elastic constants C_{ij} and compare the results with experiment. We assume that $\Delta a/a \equiv \epsilon_a$, the macroscopic (bulk) strain in the **a** direction. Differentiating Eq. (11) with respect to p_a gives

$$\frac{\partial T_c}{\partial p_a} = T_c(0)K\frac{\partial \epsilon_a}{\partial p_a}\,. \quad (12)$$

$(\partial \epsilon_a/\partial p_a)$ is the compliance coefficient $s_{11} \equiv (\hat{C}^{-1})_{11}$, where $\hat{C}$ is the elastic tensor. We calculate $K \approx 4.5$ from the TU model for a uniaxial pressure in the **a** direction for the weak coupling case ($J_{\text{ave}}/T_c \approx 3.2$) and use the complete set of values for C_{ij} from Zoubolis *et al.* [12] to obtain $\partial T_c/\partial p_a \approx -1.8$ K/GPa. If we use another complete set of C_{ij} reported by Lei *et al.* [13] we obtain $\partial T_c/\partial p_a \approx -2.5$ K/GPa. These results compare favorably with the experimental value of $\partial T_c/\partial p_a = -2.0$ K/GPa. Such a close agreement is somewhat surprising because of the uncertainty in the experimental values of the elastic constants (see Ref. [12] for a review of experimental data).

We next consider a uniaxial pressure along the **b** direction. From the thermal expansion measurements [4] it is found that the two-dimensional volume in the **a**-**b** plane is approximately conserved for the $T_c \approx 91$ K sample. Thus for a uniaxial pressure in the **b** direction the distances in the **b** direction decrease while those in the **a** direction increase, leading to the positive Δa and increasing T_c. Two-dimensional volume conservation can be expressed as $d\epsilon_a = -d\epsilon_b$. The uniaxial pressure dependence in the **b** direction then becomes

$$\frac{\partial T_c}{\partial p_b} = T_c(0)K\frac{\partial \epsilon_a}{\partial p_b} = -T_c(0)K\frac{\partial \epsilon_b}{\partial p_b} = -T_c(0)Ks_{22}\,. \quad (13)$$

Using the same two sets of elastic constants [12,13] we obtain $(\partial T_c/\partial p_b) \approx +1.4$ K/GPa and $(\partial T_c/\partial p_b) \approx +2.3$ K/GPa. Considering the fact that the elastic constants are not accurately known [12] and that the tunneling units may have effective elastic constants different from the bulk solid, the discrepancy between the calculated and the measured values is small. Thus our tunneling-unit model gives not only the qualitative, but quite close quantitative agreement with the experimental anisotropic pressure dependence of T_c.

The values of K in Eq. (11) calculated from the TU model increase with increasing values of $(N_0 V_{\text{eff}})^{-1}$. Detailed calculations of K as a function of $N_0 V_{\text{eff}}$ will be presented elsewhere [14].

We next discuss the changes in the thermal expansivity α near T_c. The pressure derivatives $\partial T_c/\partial p_i$ are related to changes in the thermal expansivities [4] $\Delta\alpha$ at T_c. Let $\Delta\alpha_i = \alpha_{Si} - \alpha_{Ni}$, where α_{Si} and α_{Ni} are the superconducting and normal values of α_i at T_c. Let ΔC_p be the jump in the specific heat at T_c. Using the Ehrenfest relations [15] one has [4]

$$\Delta\alpha_i = \frac{\Delta C_p}{V_m T_c}\left(\frac{\partial T_c}{\partial p_i}\right), \quad (14)$$

where V_m is the volume of the material. It is important to point out that using the Ehrenfest relations and the value of ΔC_p, one directly relates the anisotropic pressure dependence $\partial T_c/\partial p_i$ to $\Delta\alpha_i$. Thus following the calculations of Meingast *et al.* [4], which are not repeated here, the TU model gives both the uniaxial pressure dependence of T_c as well as the jump in $\Delta\alpha_i$ near T_c.

So far in this paper we dealt with $YBa_2Cu_3O_{7-\delta}$ close to optimal oxygenation, $T_c \approx 91$ K, and $\partial T_c/\partial p_c \approx 0$. We next consider pressure dependences for underdoped $YBa_2Cu_3O_{7-\delta}$.

The variation of T_c with hydrostatic pressure can be described by the pressure induced charge transfer (PICT) mechanism. In the PICT mechanism the charge carrier density n_h in the CuO_2 planes is one of the key parameters controlling the value of T_c [16,17]. A graph for the values of T_c vs n_h is a bell shaped curve. From the PICT model n_h and hence T_c change with pressure and dT_c/dp depends strongly on the position of T_c on the bell shaped curve [17]. dT_c/dp is expected to be positive for underdoped, negative for overdoped, and zero for the "optimally" doped cuprates. Charge transfer is expected to occur primarily by changes in the **c**-axis bond length [1,18], hence uniaxial pressure in the **c** direction could investigate the PICT model [19]. Thus for $YBa_2Cu_3O_{7-\delta}$ close to $\delta \approx 0$ and $T_c \approx 91$ K, dT_c/dp_c is small, because T_c is near the top of the bell shaped curve for n_h, showing no important PICT contribution. Furthermore, in the PICT model it is not expected that the charges transferred are strongly influenced by uniaxial pressure in the **a** and **b** directions. If so, the PICT model could not explain the **a** and **b** axis anisotropies.

When oxygen is removed from the system T_c drops and $\partial T_c/\partial p_c$ increases [20]; the uniaxial pressure dependences in the **a** and **b** directions become unequal but still opposite in sign for six different values of δ ranging from 0.05 to 0.46, indicating that the tunneling units still make an important contribution [20–23]. There is a large

increase in $\partial T_c/\partial p_c$ with increasing δ, suggesting that PICT becomes more important with increasing δ. Thus our derivation suggests that the uniaxial pressure dependences have (at least) two contributions, one arising from the charge transfer, and the other from changing the width of the wells of the tunneling units.

We propose experiments to separate these two effects. The distribution of the tunneling units in the material and their orientation with respect to the crystal axes strongly affect the response of the superconductor to uniaxial pressure in the three directions. The distribution of the TU's could be obtained from neutron diffraction studies [8]. Thus when the oxygen concentration in the material is changed one could separate the changes in the uniaxial pressure dependences in the three directions into two components; (i) arising from the change in the number and in the orientation of the TU's and in the pressure dependent width of the tunneling unit and (ii) pressure dependent changes in the **c** direction, which cause charges to transfer from the CuO_2 planes. Therefore a good deal of physics could be learned from measuring the uniaxial pressure dependences of T_c as a function of the oxygen concentration, coupled with neutron diffraction experiments to obtain information on the number, width, and direction of the tunneling units.

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[1] J. S. Shilling and S. Klotz, in *Physical Properties of High Temperature Superconductors*, edited by D. Ginsberg (World Scientific, Singapore, 1992), Vol. III, p. 59.

[2] R. J. Wijngaarden and R. Griessen, in *Studies of High Temperature Superconductors*, edited by A. V. Narliker (Nova, New York, 1990), Vol. 2, p. 29.

[3] U. Welp, M. Grimsditch, S. Fleshler, W. Nessler, J. Downey, G. W. Crabtree, and J. Guimpel, Phys. Rev. Lett. **69**, 2130 (1992).

[4] C. Meingast, O. Kraut, T. Wolf, H. Wuhl, A. Erb, and G. Miller-Vogt, Phys. Rev. Lett. **67**, 1634 (1991).

[5] For a general review of high T_c superconductivity, see *Physical Properties of High Temperature Superconductors*, edited by D. Ginsberg (World Scientific, Singapore, 1992–1995), Vols. III–V.

[6] S. B. Simanovsky and M. W. Klein, Phys. Rev. B **54**, 7430 (1996).

[7] S. B. Simanovsky and M. W. Klein, Solid State Commun. **99**, 729 (1996).

[8] A. W. Hewat *et al.*, IBM J. Res. Dev. **33**, 220 (1989).

[9] Tunneling units not in the chain but still oriented in the **a** direction will give the results derived in this paper, and it matters little whether the TU is an oxygen atom or a copper atom.

[10] R. Dynes *et al.*, Phys. Rev. Lett. **72**, 2267 (1994).

[11] In Ref. [7] we denote the distance between the wells by x, however for our purposes it is more convenient to use x as the variable, and let $2a$ be the width of the tunneling unit and Δa be the change in distance resulting from the uniaxial pressure.

[12] E. Zoubolis *et al.*, Physica (Amsterdam) **190C**, 329 (1992); $C_{11} = 248$, $C_{12} = 36.7$, $C_{13} = 61.3$, $C_{22} = 301$, $C_{23} = 5.88$, and $C_{33} = 183$ (all in GPa).

[13] M. Lei *et al.*, Bull. Am. Phys. Soc. **37**, 649 (1992); $C_{11} = 231$, $C_{12} = 132$, $C_{13} = 71$, $C_{22} = 268$, $C_{23} = 95$, and $C_{33} = 186$ (all in GPa).

[14] S. B. Simanovsky and M. W. Klein (to be published).

[15] M. Lang *et al.*, Z. Phys. B **69**, 451 (1988); K. Kadowaki *et al.*, Physica (Amsterdam) **153-155C**, 1028 (1988).

[16] O. Kraut *et al.*, Physica (Amsterdam) **205C**, 139 (1993), and references therein.

[17] J. B. Torrence *et al.*, Physica (Amsterdam) **162-164C**, 291 (1981); Y. Tokura *et al.*, Phys. Rev. B **38**, 7156 (1988); Y. J. Uemura *et al.*, Phys. Rev. Lett. **62**, 2317 (1989).

[18] G. Baumggartel *et al.*, Phys. Rev. B **45**, 308 (1992).

[19] A. J. Millis and K. M. Rabe, Phys. Rev. B **38**, 8908 (1988).

[20] O. Kraut *et al.*, Physica (Amsterdam) **205C**, 139 (1993); J. D. Jorgensen, Physica (Amsterdam) **171C**, 93 (1990); Y. Yamada *et al.*, Physica (Amsterdam) **173C**, 185 (1991); M. Meingast *et al.*, Physica (Amsterdam) **235-240C**, 1313 (1994); H. Wuhl *et al.*, Physica (Amsterdam) **185-189C**, 755 (1991); H. Claus *et al.*, Physica (Amsterdam) **198C**, 42 (1992).

[21] An exception to this is $YBa_2Cu_3O_{7-\delta}$ for $\delta = 0.27$, for which the pressure dependence is positive in both the **a** and **b** directions. We have no explanation for this result.

[22] R. Benishke *et al.*, Physica (Amsterdam) **203C**, 293 (1992).

[23] C. Meingast *et al.*, Physica (Amsterdam) **209C**, 591 (1993).