Superfluid Anisotropy in YBCO: Evidence for Pair Tunneling Superconductivity

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(Received 11 August 1995)

Proximity effect and pair tunneling models are applied as alternative scenarios to explain the recently measured *ab*-plane and *c*-axis components of the superfluid tensor in copper-oxide superconductors which contain chains, such as $YBa_2Cu_3O_{6.95}$. It is argued that conventional proximity effect models, which couple chains and planes via single electron tunneling only, are incompatible with the experimental observations. On the other hand, several surprising features of the experimental data are readily explained by the presence of a microscopic pair tunneling process.

PACS numbers: 74.50.+r, 74.20.Mn

Recent measurements of the electromagnetic response of clean untwinned single crystals of $YBa_2Cu_3O_{7-\delta}$ and $YBa_2Cu_4O_8$ reveal a large *ab*-plane anisotropy [1,2]. The ratio of the superfluid density in *b* and *a* directions (CuO chains are along the *b* axis) at zero temperature is about ≈ 2.4 for the one-chain compound $YBa_2Cu_3O_{6.95}$ and rather larger, ≈ 6, for the two-chain compound $YBa_2Cu_4O_8$ [1]. Anisotropies of similar magnitudes are observed in the normal state resistivity [3]. Early work on Knight shift and the NMR relaxation time T_1 [4] in $YBa_2Cu_3O_{6.95}$ indicates that there is an appreciable gap on the chains below T_c. Taken together, these experiments unequivocally show that normal electrons on the CuO chains are coupled into the superconducting state below T_c. However, an unexpected feature of the recent data is that the temperature dependences of the superfluid densities in *a* and *b* directions are similar; both are clearly linear at low temperature [1], and roughly obey $\rho_a^{(s)}(T)/\rho_a^{(s)}(0) \simeq \rho_b^{(s)}(T)/\rho_b^{(s)}(0)$ up to T_c, where $\rho_{a(b)}^{(s)}$ is the superfluid density in the a (b) direction. Along the *c* axis, $\rho_c^{(s)}(T)$ approaches its zero-temperature value with a power higher than linear. The implications of these facts for the microscopic nature of *c*-axis coupling in copper oxides is the subject of this Letter.

Two distinct models of plane-chain coupling are considered here; first, a proximity model where intrinsically superconducting plane and chain layers are coupled through *single electron tunneling only* [5] and, second, an interlayer pair tunneling model where chains and planes are coupled through a Josephson-like *pair tunneling* process. In both cases the superfluid response is assumed to be dominated by coherent Fermi excitations about quasi-2D chain and plane Fermi surface sheets [6]. We mimic clean YBCO-type electronic structures by stacking planar "CuO_2" and chain "CuO" layers alternately along the *c* axis, and employ a simple tight-binding description. We show that the penetration depth results are incompatible with the proximity model, but find a natural explanation within the pair tunneling model.

The proximity model is defined by the mean-field Hamiltonian

$$H = H_0 + H_1 , \tag{1}$$

$$H_0 = \sum_k \left[\sum_n \varepsilon_{nk} c_{nk\sigma}^{\dagger} c_{nk\sigma} + \varepsilon_{\perp k} (c_{1k\sigma}^{\dagger} c_{2k\sigma} + \text{H.c.}) \right], \tag{2}$$

$$H_1 = \sum_{nk} \Delta_n \gamma_{nk} (c_{nk\uparrow}^{\dagger} c_{n-k\downarrow}^{\dagger} + \text{H.c.}) , \tag{3}$$

where $c_{1k\sigma}^{\dagger}$ and $c_{2k\sigma}^{\dagger}$ are the creation operators of electrons in the plane and chain bands, respectively, $\varepsilon_{1k} = -2t(\cos k_a + \cos k_b) - \mu$, and $\varepsilon_{2k} = -2t_c \cos k_b - \mu + \varepsilon_c$. Here μ is the chemical potential and ε_c the relative energy between the chains and the planes. It is sufficient to assume the simplest nearest-neighbor tight-binding dispersions for electrons on both chain and plane layers; the qualitative results are independent of the detailed forms of ε_{1k} and ε_{2k}. $\Delta_n \gamma_{nk}$ are the gap functions with the pairing symmetries γ_{nk} on the plane $(n = 1)$ and chain $(n = 2)$ bands. They are determined by self-consistent gap equations $(\lambda_n/V) \sum_k \gamma_{nk} \langle c_{nk\uparrow} c_{n-k\downarrow} \rangle = \Delta_n$, where λ_n $(n = 1, 2)$ are the strengths of the pairing potential on the plane and chain layers. When $\lambda_2 = 0$, the chain layer is not intrinsically superconducting, $\Delta_2 = 0$, and the superconducting correlation in the chain layer is generated purely by the proximity effect. The coupling between the chains and planes in Eq. (2) is via single electron interlayer hopping with matrix element $\varepsilon_{\perp k} = -2t_{\perp} \cos(k_c/2)$. This coupling vanishes at the zone boundary $k_c = \pi$. In the physically relevant regime the interlayer hopping constant $t_{\perp}$ is much smaller than the plane and chain hopping constants t and t_c.

The mean-field Hamiltonian (1) has four branches of quasiparticle excitations. Their energy dispersions are given by $\pm E_{\pm,k}$ with $E_{\pm,k} = \sqrt{b_k \pm \sqrt{b_k^2 - c_k}}$, where $b_k = \varepsilon_{\perp k}^2 + \sum_n (\Delta_n^2 \gamma_{nk}^2 + \varepsilon_{nk}^2)/2$ and $c_k = (\varepsilon_{1k} \Delta_2 \gamma_{2k} + \varepsilon_{2k} \Delta_1 \gamma_{1k})^2 + (\varepsilon_{\perp k}^2 - \varepsilon_{1k} \varepsilon_{2k} + \Delta_1 \Delta_2 \gamma_{1k} \gamma_{2k})^2$. While the $E_{+,k}$ quasiparticle excitation branch has a gap for all k, zero energy excitations may be present in the $E_{-,k}$ branch. The locations of such energy gap nodes are given by the condition $c_k = 0$, i.e., by the solutions of the simultaneous equations $\varepsilon_{1k} \Delta_2 \gamma_{2k} + \varepsilon_{2k} \Delta_1 \gamma_{1k} = 0$

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and $\varepsilon_{\perp k}^2 - \varepsilon_{1k}\varepsilon_{2k} + \Delta_1\Delta_2\gamma_{1k}\gamma_{2k} = 0$. Such solutions depend strongly on the pairing symmetries of electrons on both layers. If the planar electrons have d-wave pairing symmetry, i.e., $\gamma_{1k} = (\cos k_b - \cos k_a)/2$, line gap nodes always exist. However, if both γ_{1k} and γ_{2k} have isotropic s-wave symmetry (e.g., $\gamma_{1k} = \gamma_{2k} = 1$), nodal lines exist only when the pairing functions on the plane and chain layers have opposite signs, $\Delta_1\Delta_2 \le 0$, and, in addition, $4t_\perp^2 > |\Delta_1||\Delta_2|$. In the somewhat artificial case where the pair interaction on the chain layer vanishes, $\Delta_2 = 0$ and the "proximity" nodal line on the chain Fermi surface sheet ($\varepsilon_{2k} = 0$) lies along $k_c = \pi$, independent of the pairing symmetry of the plane layer. More generally, proximity induced nodal line loops are present on the chain Fermi surface sheet.

The superfluid tensor is related to the kinetic energy and the current-current correlation function [7–9]:

$$\rho_\mu^{(s)} = \langle -\alpha_\mu^2 K_\mu \rangle - \frac{i}{V}\int_{-\infty}^{0} dt\, e^{-i\omega t}\langle [J_\mu(0), J_\mu(t)]\rangle, \tag{4}$$

where $\langle A\rangle = \mathrm{Tr}(Ae^{-\beta H})/\mathrm{Tr}e^{-\beta H}$ and $\alpha_\mu = (a, b, c/2)$ for $\mu = (a, b, c)$ with (a, b, c) lattice constants. $\langle K_\mu\rangle$ and J_μ are the kinetic energy and the current operator along the μ direction, respectively. $J_a = \sum_{k\sigma} 2at \sin k_a c_{1k\sigma}^\dagger c_{1k\sigma}$, $J_b = \sum_{k\sigma} 2b \sin k_b (t c_{1k\sigma}^\dagger c_{1k\sigma} + t_c c_{2k\sigma}^\dagger c_{2k\sigma})$, and $J_c = ct_\perp \sin(k_c/2)(c_{1k\sigma}^\dagger c_{2k\sigma} + c_{2k\sigma}^\dagger c_{1k\sigma})$. For the proximity model Eq. (1), we find that the modification of the superfluid density along the nonchain a axis ($\rho_a^{(s)}$) due to chain-plane hybridization is very small, and its temperature dependence is almost the same as for a pure 2D system with the chains and planes decoupled. If the planar band has d-wave pairing symmetry, $\rho_a^{(s)}$ will be linear in T at low temperature. Along the b axis, however, the contribution of the chain band is important. Close to the transition temperature, $\rho_b^{(s)}$ is dominated by the contribution of the plane band and the difference between $\rho_a^{(s)}$ and $\rho_b^{(s)}$ is small. At low temperature, the leading temperature dependence of $\rho_b^{(s)}$ is determined by that of the low-energy density of states $\rho(\omega) = (1/V)\sum_k[\delta(\omega - E_{-,k}) + \delta(\omega - E_{+,k})]$. In the absence of nodes, for example, when $\gamma_{1k} = \gamma_{2k} = 1$ and $\Delta_1\Delta_2 > 0$, $\rho_b^{(s)}$ approaches its zero-temperature value exponentially as the temperature decreases. Since this is clearly inconsistent with experiments, we consider only (nodeful) gapless cases. When $\omega < \min(E_{+,k})$, only $E_{-,k}$ quasiparticles have contributions to $\rho(\omega)$. Since $E_{-,k} = \sqrt{c_k}/E_{+,k}$ and $E_{+,k}$ is weakly k dependent around the gap nodes in general, $\rho(\omega)$ at low energy is therefore determined mainly by the structure of c_k. If $\Delta_2 = 0$, the quasiparticles around the nodal line $k_c = \pi$ and $\varepsilon_{2k} = 0$ dominate the low energy excitations, and it can be shown that $\rho(\omega) \sim \sqrt{\omega}$ for small ω. This is a consequence of the extreme flatness of the quasiparticle dispersion in a direction normal to the nodal line $E_{-,k} \sim (k_c - \pi)^2$ near $k_c = \pi$. Correspondingly, the low temperature superfluid density along the b direction has pronounced upward curvature, increasing rapidly as $\sqrt{T}$ as zero temperature is approached [9]. This behavior is clearly inconsistent with the linear temperature dependence of the superfluid density in both planar directions in $YBa_2Cu_3O_{7-\delta}$ and $YBa_2Cu_4O_8$. On the other hand, in the realistic situation where both Δ_1 and Δ_2 are finite, it can be shown that $\rho(\omega) \sim \omega$ when $\omega \ll (|\Delta_1|, |\Delta_2|)$, and hence $\rho_b^{(s)} \sim T$ when $T \ll (|\Delta_1|, |\Delta_2|)$. In an intermediate temperature regime $|\Delta_2| \ll T \ll |\Delta_1|$, the strong upward curvature in $\rho_b^{(s)}$ persists. Moreover, in the regime of physically relevant parameters self-consistent solutions of the gap equations give $|\Delta_2| \ll |\Delta_1|$. Along the c axis, $\rho_c^{(s)}$ shows also a positive curvature. But the low temperature behavior of $\rho_c^{(s)}$ is more peculiar. If $\Delta_2 = 0$, it can be shown that at low temperature $\rho_c^{(s)} \sim T$ rather than $\sqrt{T}$ as a consequence of vanishing coherence factors on proximity nodal lines. If, however, Δ_2 is finite and $T \ll \Delta_2$, we find that $\rho_c^{(s)} \sim T^2$. Figure 1 shows the typical temperature dependence of the three components of the superfluid densities in the proximity model on a normalized plot.

The presence of a positive curvature in $\rho_b^{(s)}$ is not limited to the "pure" proximity effect limit discussed above but is in fact a general feature in weakly coupled two-gap systems. Consider the case where the chain and plane bands are completely decoupled with transition temperatures $T_c^{\mathrm{chain}} < T_c^{\mathrm{plane}}$ and superfluid densities $\rho_{\mathrm{chain}}^{(s)}$ and $\rho_{\mathrm{plane}}^{(s)}$, respectively. In this case, $\rho_b^{(s)} = \rho_{\mathrm{plane}}^{(s)} + \rho_{\mathrm{chain}}^{(s)}$

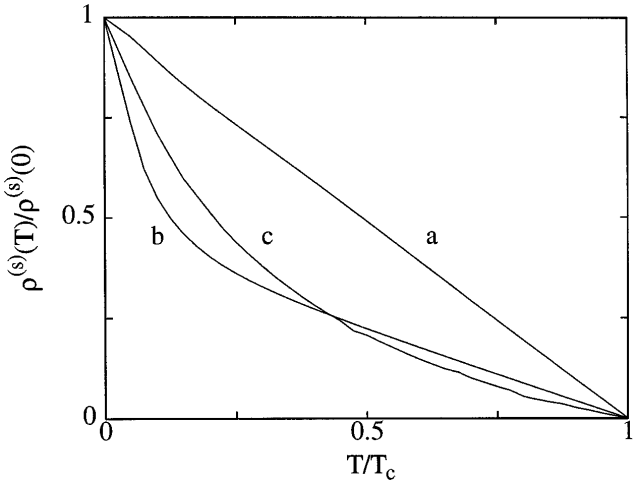

FIG. 1. Normalized superfluid densities $\rho^{(s)}(T)/\rho^{(s)}(0)$ as functions of T/T_c along a, b, and c directions in the proximity model. $\gamma_{1k} = (\cos k_a - \cos k_b)/2$ and $\gamma_{2k} = \cos k_b$. $t = 1$, $t_c = 0.85$, $t_\perp = 0.2$, $\varepsilon_c = 0.5$, $\lambda_1 = \lambda_2 = 1.5$ ($T_c \sim 0.23$), and the electron concentration on the CuO planes is about 0.85. $2\Delta_1(T = 0)/T_c \sim 4.6$, which is nearly the same as for a purely d-wave superconductor on square lattices at the filling 0.85. $\rho_b^{(s)}(0)/\rho_a^{(s)}(0) \sim 2.2$ and $\rho_b^{(s)}(0)/\rho_c^{(s)}(0) \sim 70$.

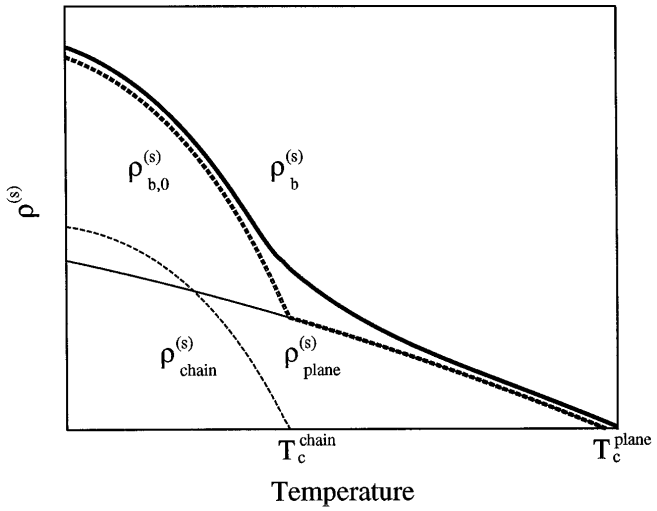

FIG. 2. Schematic representation of the superfluid density in the proximity model. $\rho^{(s)}_{\rm chain}$ and $\rho^{(s)}_{\rm plane}$ are the superfluid densities of the chains and planes when the interlayer coupling is zero. $\rho^{(s)}_{b,0} = \rho^{(s)}_{\rm plane} + \rho^{(s)}_{\rm chain}$. When chains and planes are weakly coupled by single electron tunneling, the chain-direction superfluid response develops a positive curvature near $T_c^{\rm chain}$.

will have a sudden change at $T_c^{\rm chain}$ as shown in Fig. 2. Switching on a weak interlayer coupling $t_\perp$ leads to single transition temperature $\simeq T_c^{\rm plane}$ but leaves a smooth upturn in $\rho_b^{(s)}$ in the vicinity of $T_c^{\rm chain}$.

While a $d_{x^2-y^2}$ paired state can account for the linear temperature behavior of $\rho_a^{(s)}$, we conclude that the proximity model cannot give a satisfactory explanation of the observed temperature dependences of $\rho_b^{(s)}$ or $\rho_c^{(s)}$. However, the above analysis does highlight paradoxical features of the experimental results, namely, the CuO chain layers must be intrinsically superconducting, yet there must be *a node of the energy gap on the chain Fermi surface sheet*. Otherwise, $(d\rho_b^{(s)}/dT)|_{T=0} \simeq (d\rho_a^{(s)}/dT)|_{T=0}$ as the chain band gives no contribution to the linear temperature term of $\rho_b^{(s)}$ at low temperature, in contradiction to experiment. As a quasi-1D system has an energy gap under ordinary circumstances, the presence of nodes suggests that the gap function of the chain band $\Delta_2\gamma_{2k}$ may have 2D character; i.e., γ_{2k} is not simply a function of k_b only. We are thus led to the surprising conclusion that superconducting coherence extends across CuO chains in the chain layer.

The failure of the single particle tunneling model leads us to introduce a pair tunneling model of the type originally proposed by Wheatley, Hsu, and Anderson [10]. The model consists of a kinetic energy term H_0, which is the same as defined in Eq. (2), and a local singlet pair tunneling potential term H_I:

$$H_I = -\frac{\lambda}{4} \sum_{r,\delta=\hat{a},\hat{b}} (\hat{\Delta}^\dagger_{1,r,\delta}\hat{\Delta}_{2,r+\hat{c}/2,\delta} + \hat{\Delta}^\dagger_{1,r,\delta}\hat{\Delta}_{2,r-\hat{c}/2,\delta} + \text{H.c.}), \quad (5)$$

where $\hat{\Delta}_{n,r,\delta} \equiv c_{nr\uparrow}c_{nr+\delta\downarrow} - c_{nr\downarrow}c_{nr+\delta\uparrow}$ is the singlet pair operator. This model is compatible with the two experimental requirements specified above. (1) The pairing functions in the chain and plane layers must have the same symmetry, e.g., both s or both d, if the admixture between the s- and d-wave components is weak. Otherwise, the only self-consistent solution to the mean-field Hamiltonian (6) (below) is the trivial one $\tilde{\Delta}_n = 0$. This model accounts for the presence of interchain pairing discussed above. (2) The magnitudes of the gap parameters on chains and planes are tied together; plane and chain layers are not independently superconducting.

Taking the BCS mean-field approximation, each term in (5) can be decoupled, for example, for the first term in (5), as $\hat{\Delta}^\dagger_{1,r,\delta}\hat{\Delta}_{2,r+\hat{c}/2,\delta} \simeq \langle\hat{\Delta}^\dagger_{1,r,\delta}\rangle\hat{\Delta}_{2,r+\hat{c}/2,\delta} + \langle\hat{\Delta}_{2,r+\hat{c}/2,\delta}\rangle\hat{\Delta}^\dagger_{1,r,\delta} - \langle\hat{\Delta}^\dagger_{1,r,\delta}\rangle\langle\hat{\Delta}_{2,r+\hat{c}/2,\delta}\rangle$, where $\langle\hat{\Delta}\rangle$ is the thermal average of the operator $\hat{\Delta}$. As the system is translationally invariant, $\langle\hat{\Delta}^\dagger_{n,r,\delta}\rangle$ should be r independent. If we further assume the amplitude of $\langle\hat{\Delta}^\dagger_{n,r,\delta}\rangle$ is δ independent, then the gap function $\lambda\langle\hat{\Delta}^\dagger_{n,r,\delta}\rangle$ can be written as $\lambda\langle\hat{\Delta}^\dagger_{n,r,\delta}\rangle = \tilde{\Delta}_n\alpha_{n\delta}$, here $\tilde{\Delta}_n$ and $\alpha_{n\delta}$ are, respectively, the amplitude and the phase factor of $\lambda\langle\hat{\Delta}^\dagger_{n,r,\delta}\rangle$. $\alpha_{n\delta}$ are determined by the pairing symmetries of the chain and plane bands. With $\alpha_{1\delta} = \alpha_{2\delta} = \alpha_\delta$, the mean-field decoupled pairing potential H_I can be written as

$$H_I' = \sum_k \Big[(\tilde{\Delta}_2\gamma_k c^\dagger_{1k\uparrow}c^\dagger_{1-k\downarrow} + \tilde{\Delta}_1\gamma_k c^\dagger_{2k\uparrow}c^\dagger_{2-k\downarrow} + \text{H.c.}) + \frac{2}{\lambda}\tilde{\Delta}_1\tilde{\Delta}_2 \Big], \quad (6)$$

where $\gamma_k = \sum_\delta \alpha_\delta \cos k_\delta$. On a square lattice, the nearest-neighbor singlet paired state can have either extended s-wave symmetry, $\gamma_k = (\cos k_a + \cos k_b)/2$, or d-wave symmetry, $\gamma_k = (\cos k_a - \cos k_b)/2$. As only the d-wave pairing state can have gap nodes on the planar Fermi surface, we consider only this case.

At the mean-field level the proximity and pair tunneling models H_I' and H_I have the same form; the difference lies only in how the self-consistent procedure for the gap function has been applied. Thus the mean-field quasiparticle energy spectra are identical except that $\Delta_1\gamma_{1k}$ and $\Delta_2\gamma_{2k}$ in the proximity model spectrum should now be replaced by $\tilde{\Delta}_2\gamma_k$ and $\tilde{\Delta}_1\gamma_k$, respectively.

Figure 3 shows the components of the normalized superfluid tensor as functions of T/T_c computed in the pair tunneling model using a typical parameter set. The ratio $\rho_b^{(s)}/\rho_a^{(s)}$ at zero temperature is mainly determined by t_c/t, while $\rho_b^{(s)}/\rho_c^{(s)}$ is mainly determined by $t_\perp/t$. For the case shown in Fig. 3, the parameters have been chosen so that the ratios $\rho_b^{(s)}/\rho_a^{(s)}$ and $\rho_b^{(s)}/\rho_c^{(s)}$ at zero temperature are approximately 2.4 and 100, which gives a qualitative fit to the experimental values for $YBa_2Cu_3O_{6.95}$. The overall temperature dependences of $\rho_b^{(s)}(T)/\rho_b^{(s)}(0)$

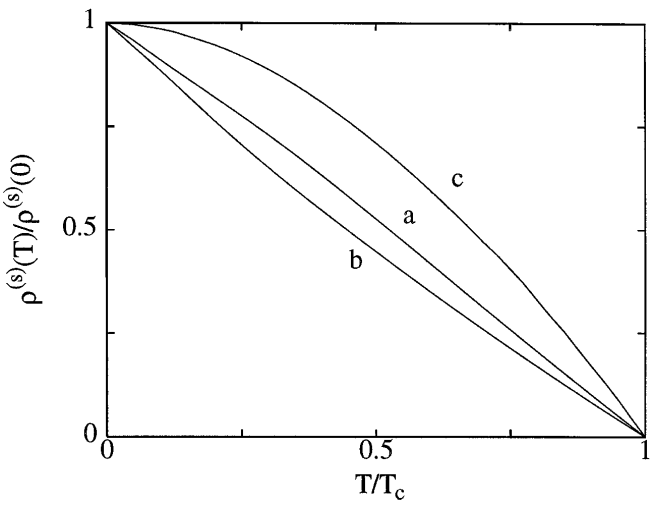

FIG. 3. Normalized superfluid densities $\rho^{(s)}(T)/\rho^{(s)}(0)$ as functions of T/T_c along a, b, and c directions in the interlayer pair tunneling model with d-wave pairing symmetry. $t = 1$, $t_c = 0.85$, $t_\perp = 0.17$, $\varepsilon_c = 0.5$, $\lambda = 1.5$ ($T_c \sim 0.17$), and the electron concentration on the CuO planes is about 0.85. At $T = 0$, the ratio between the maximum gap and T_c on the planar Fermi sheet is $2\tilde{\Delta}_2/T_c \sim 4.6$.

and $\rho_a^{(s)}(T)/\rho_a^{(s)}(0)$ are quite similar. At low temperature $\rho_a^{(s)}(T) \sim T$ because of the planar d-wave state. As both $\tilde{\Delta}_1$ and $\tilde{\Delta}_2$ are finite in this case, from the previous discussion $\rho(\omega) \sim \omega$ at low energy and $\rho_b^{(s)} \sim T$ at low temperature. The temperature dependence of $\rho_c^{(s)}(T)/\rho_c^{(s)}(0)$ is quite different from $\rho_a^{(s)}(T)/\rho_a^{(s)}(0)$, especially at low temperature where the numerical results indicate $\rho_c^{(s)} \sim T^2$. All of these results agree very well with the experimental measurements [1]. Increasing ε_c so that the effective electron concentration in the chain band decreases, we find that the difference between $\rho_a^{(s)}(T)/\rho_a^{(s)}(0)$ and $\rho_b^{(s)}(T)/\rho_b^{(s)}(0)$ becomes smaller, but the difference between $\rho_c^{(s)}(T)/\rho_c^{(s)}(0)$ and $\rho_a^{(s)}(T)/\rho_a^{(s)}(0)$ becomes even larger. These changes of $\rho_a^{(s)}(T)/\rho_a^{(s)}(0)$, $\rho_b^{(s)}(T)/\rho_b^{(s)}(0)$, and $\rho_c^{(s)}(T)/\rho_c^{(s)}(0)$ with the electron concentration in the chain band agree qualitatively with the experimental results for $YBa_2Cu_3O_{6.6}$ and $YBa_2Cu_3O_{6.95}$ [1].

In the above discussion we have assumed that the amplitude of the order parameter $\langle\hat{\Delta}_{n,\delta}\rangle$ is the same in a and b directions. A more complete self-consistent solution of the gap equations should allow admixture of an s-wave component to form a $d + s$ wave state [11]. The gap nodes survive for weak admixture, but the positions of the nodes shift away from the diagonals of the Brillouin zone. Admixture of a small s component is consistent with the c-axis Josephson tunneling experiments on $YBa_2Cu_3O_{7-\delta}$ [12]. While full self-consistent treatment for the gap equations including an s component is complicated technically in our model, preliminary calculations suggest that this improves the agreement between the pair tunneling model and experiments. Another refinement of the model would include a finite plane layer pairing interaction.

In conclusion, we have pointed out that the planar anisotropy of electromagnetic response of clean YBCO-type structures forms a useful probe of the microscopic state of copper-oxide superconductors. We have argued that the data of Hardy and co-workers [1] imply that the CuO chains layers are intrinsically superconducting but that there is a node on the chain Fermi surface sheet. The experimentally observed temperature dependences of superfluid densities are incompatible with conventional d-wave proximity models in the clean limit without an implausible fine tuning of parameters. The data are, however, readily compatible with a model of d-wave pair tunneling superconductivity.

We thank J. Waldram for bringing our attention to the significance of Ref. [1].

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