c Axis Superfluid Response of Copper Oxide Superconductors

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A novel interplay between d-wave superconducting order parameter symmetry and the underlying Cu $3d$ orbital based electronic structure of copper oxides leads to a striking anisotropy in the superfluid response of these systems. In clean tetragonal materials the c axis penetration depth increases as T^5 at low temperature, in contrast to linear T behavior in the ab plane. Disorder is a relevant perturbation which causes all components of the superfluid response to depend quadratically on temperature at low temperature. However, the crossover temperature scale from the intrinsic d-wave behavior to the disorder dominated behavior for the in-plane response may be different from that for the out-plane response. [S0031-9007(96)01768-1]

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A variety of experimental data on copper oxide superconductors are compatible with a pairing state of $d_{x^2-y^2}$ symmetry in CuO_2 planes. The existence of energy gap nodes in this state has profound consequences for the low temperature electromagnetic response. The in-plane penetration depth increases linearly with T at low T in contrast to the activated behavior of conventional superconductors. The linear-T result holds in the regime where the response is governed by coherently propagating fermion quasiparticle excitations, i.e., at temperatures $\Gamma \ll T \ll T_c$ where Γ is the superconducting quasiparticle scattering rate. Linear-T behavior in the ab plane has now been established in both chain and nonchain copper oxides [1–4]. The c-axis penetration depth $\lambda_c(T)$ in $La_{2-x}Sr_xCuO_4$ [5], $YBa_2Cu_3O_{7-\delta}$ [1], $Bi_2Sr_2CaCu_2O_{8+x}$ [2,6], and $HgBa_2Ca_2Cu_3O_{8+x}$ [4] has now been measured. However, as it is very difficult to determine accurately the low T dependence of λ_c due to the high anisotropy of high-T_c oxides, the published data of λ_c still lack consistency except all have shown that the T dependence of λ_c is much weaker than that observed in the ab plane. In $La_{2-x}Sr_xCuO_4$, $\lambda_c(T)$ drops exponentially with T at low temperature; while in other compounds, $\lambda_c(T)$ approaches its zero temperature value in a power law. Straightforward extension of the d-wave model to 3D leads to line gap nodes on a warped cylindrical Fermi surface. This gives a linear-T c-axis response in apparent contradiction to experiment [7].

In this paper we propose that weak T dependence of the c-axis superfluid density ρ_s^c is a general feature of nonchain cuprates with crystal tetragonal symmetry, whose origin lies in the simple but unusual electronic structure of these materials. Our analysis is based on the assumption that coherent Bloch bands along the c axis are present at low temperatures; i.e., k_z is a good quantum number. As discussed below, an essential feature of the electronic structure is that the c-axis hopping integral $t_\perp(k_\parallel)$ is a function of the in-plane momentum $k_\parallel$ and is vanishingly small when $k_\parallel$ lies along the zone diagonals of the 2D Brillouin zone [8,9]. The nodal lines of the $d_{x^2-y^2}$-wave gap $\Delta(k_\parallel) = \Delta(\cos k_x - \cos k_y)$ and the zeros of $t_\perp(k_\parallel)$ therefore coincide. This weakens the d-wave node contribution along the c axis relative to the ab plane and leads to a weak T dependence of the c-axis superfluid response ρ_s^c. At sufficiently low temperatures disorder effects are always relevant, and tend to enhance the T dependence of ρ_s^c.

Let us first consider the electron structure in high-T_c compounds with tetragonal symmetry. For simplicity we consider a tetragonal monolayer system, for example $HgBa_2Cu\,O_{4+\delta}$ [10]. Low-density approximation (LDA) band structure calculations reveal that a minimal tight-binding model for high-T_c compounds involves nearest-neighbor hopping between Cu $3d_{x^2-y^2}$ and $4s$ (with some $3d_{3z^2-1}$ characters) and O p_x and p_y orbitals [8,9]. Thus a model Hamiltonian for $HgBa_2CuO_{4+\delta}$, expressed in terms of bonding, $\alpha_k = 2[\cos(k_x/2)p_{xk} + \cos(k_y/2)p_{yk}]/\omega_k$ where $\omega_k = 2\sqrt{\cos^2(k_x/2) + \cos^2(k_y/2)}$, and nonbonding, $\beta_k = 2[\cos(k_y/2)p_{xk} - \cos(k_x/2)p_{yk}]/\omega_k$, representation of orthogonal oxygen Wannier orbitals with respect to the Cu $d_{x^2-y^2}$ orbital [11], is given by

$$H = \sum_k C_k^\dagger \begin{pmatrix} -E_d & 0 & t_{pd}\omega_k & 0 \\ 0 & E_s + 2t_{ss}d\cos k_z & t_{ps}\mu_k & t_{ps}z_k \\ t_{pd}\omega_k & t_{ps}\mu_k & 0 & 0 \\ 0 & t_{ps}z_k & 0 & 0 \end{pmatrix} C_k + \sum_i U n_{di}^2 , \quad (1)$$

where $C_k = (d_k, s_k, \alpha_k, \beta_k)$, (E_d, E_s) are the energy of $(3d, 4s)$ orbital with respect to the O orbital, and (t_{pd}, t_{ps}, t_{ss}) are wave function overlap integrals among d, s, and p orbitals. The symmetry functions appearing here are $\mu_k = \omega_k(\cos k_x - \cos k_y)/2$ and $z_k = 2\omega_k \cos(k_x/2)\cos(k_y/2)$. The Hamiltonian (1) differs from that in Ref. [9] purely by retention of a strong Coulomb interaction U in the Cu d orbital. This produces a charge transfer insulator at half filling, a feature absent in LDA calculations.

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In the strong coupling limit, the four band model (1) can be simplified as a one-band model by eliminating the high-lying Cu $4s$ orbitals and the high-lying $d-p$ spin triplets. This projection procedure can be carried out in two steps: first, by elimination of the high-lying $4s$ orbital; and second, by solving the correlation problem within the unit cell and treating intercell hopping as a degeneracy lifting perturbation following Refs. [12,13]. This yields the well-known t-J model with both intralayer and interlayer hoppings and exchanges. We assume the nonbonding orbitals are sufficiently low lying that they are filled and inert. We notice that the dominant c-axis overlap is via large radius s orbitals [9]. Thus terms to second order in E_s^{-1} must be retained to describe effective c-axis hopping via O bonding orbitals. The lowest energy states within the CuO_2 unit cell are spin $1/2$ doublets $|1\sigma\rangle$ with one hole per unit cell and Zhang-Rice singlets $|2\rangle$ with two holes per unit cell. The ab plane hopping matrix elements for Zhang-Rice singlets involve both d-α and effective α-α overlap. However, along the c axis, the only channel for hopping is via the bonding O orbitals. It can be shown that the projected c-axis hopping integral has the form $t_\perp(k_\parallel) = t_\perp^0 \mu_k^2$, where $t_\perp^0 = t_{ss}(t_{pd}t_{ps}M_0/E_dE_s)^2$ and $M_0 = \langle 1\sigma|\alpha_\sigma^\dagger|2\rangle$ is the overlap of the state produced by eliminating the hole in the O bonding orbital and the ground state doublet. Thus $t_\perp(k_\parallel)$ contains a $k_\parallel$ dependent factor μ_k^2, which vanishes when $k_x = \pm k_y$ [14]. This is a robust feature of realistic strong correlation models of nonchain tetragonal materials and directly reflects the matrix element between Cu $4s$ and bonding O α orbitals. It is worth pointing out that ab plane anisotropy observed in c-axis magnetotransport experiments may be dominated by precisely this effect [15].

Now let us consider the effect of this $k_\parallel$ dependent $t_\perp$ on the c-axis superfluid response. We take the BCS weak coupling approximation and assume the gap order parameter has the $d_{x^2-y^2}$ symmetry. For a clean system the electromagnetic response is governed by coherent 3D fermion quasiparticle excitations at low temperature. In this case the superfluid density (or inverse square of penetration depth) is given by [16]

$$\rho_s^\mu = \frac{1}{\Omega}\sum_k \left[2\left(\frac{\partial \varepsilon_k}{\partial k_\mu}\right)^2 \frac{\partial f(\lambda_k)}{\partial \lambda_k} - \frac{\partial^2 \varepsilon_k}{\partial k_\mu^2}\frac{\varepsilon_k}{\lambda_k}\tanh\frac{\beta\lambda_k}{2}\right], \tag{2}$$

where ε_k is the energy dispersion of electrons, $\lambda_k = \sqrt{\varepsilon_k^2 + \Delta_k^2}$, and $f(\lambda_k)$ is the Fermi function. When $t_\perp(k_\parallel) = t_\perp^0 \mu_k^2$, it is straightforward to show that at low temperature the c-axis superfluid response is

$$\rho_s^c(T) \sim \frac{3}{4}N(0)(t_\perp^0)^2\left[1 - \frac{5}{12}\left(\frac{\Delta(T)}{\Omega}\right)^2 - 450\left(\frac{T}{\Delta_0}\right)^5 + o(T^5)\right], \tag{3}$$

where $N(0)$ is the normal density of states on CuO planes, $\Delta(T)$ is the energy gap, $\Delta_0 = \Delta(0)$, and Ω is the energy cutoff which is much larger than Δ. The T^5 term is from the first term in (2): one T is from the contribution of the linear density of states of the d-wave state, and the other T^4 is due to the $(\cos k_x - \cos k_y)^4$ factor in $t_\perp^2(k_\parallel)$. The second term in (3) comes from the second term in (2); at low temperature it contributes a positive T^3 term to ρ_s^c since the d-wave gap $\Delta(T)/\Delta_0 \sim 1 - (T/\Delta_0)^3$. However, the coefficient of this T^3 term is proportional to $(\Delta_0/\Omega)^2$, which is extremely small. Therefore it is likely that the dominant temperature dependence of ρ_s^c should be T^5 at low temperature. This T^5 behavior in ρ_s^c is clearly very different from the $t_\perp(k_\parallel) = \text{const}$ case where ρ_s^c varies linearly with T at low temperature. In Fig. 1, we compare the temperature dependence of $\rho_s^c(T)/\rho_s^c(0)$ in the whole temperature regime for the above two cases. The temperature dependence of $\rho_s(T)/\rho_s(0)$ along a axis is also shown in Fig. 1 for comparison.

There are several effects which may lead to a finite hopping integral along the c axis in the vicinity of the gap nodes in clean systems and thus a stronger T dependence of ρ_s^c. Direct interplane hoppings which are not assisted by Cu $4s$ orbitals, for example, can make the c-axis dispersion finite around the gap nodes and eliminate the zeros in $t_\perp(k_\parallel)$ altogether. In this case, we may model the c-axis electronic structure by $t_\perp(k_\parallel) = t_\perp^0 \mu_k^2 + t_\perp^{\text{node}}$, where $t_\perp^{\text{node}}$ denotes the interlayer hopping integral at the gap nodes contributed from all possible interlayer hopping channels not assisted by Cu $4s$ states. $t_\perp^{\text{node}}$ is expected to be small in high-T_c compounds, but it generates a

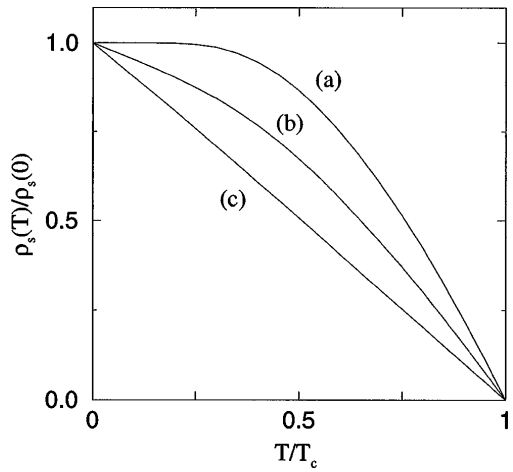

FIG. 1. Normalized superfluid density $\rho_s(T)/\rho_s(0)$ vs T/T_c. The energy dispersion of electrons $\varepsilon_k = -2t \times (\cos k_x + \cos k_y) - 4t' \cos k_x \cos k_y - 2t_\perp(k_\parallel)\cos k_z$ is used in the calculation. Curve (a) is for the model with $t_\perp(k_\parallel) = t_\perp^0(\cos k_x - \cos k_y)^2/4$ along the c axis. Curves (b) and (c) are for the model with $t_\perp(k_\parallel) = t_\perp^0$ along the c and the a axes, respectively. $t = 1$, $t' = -0.25$, $t_\perp^0 = 0.1$, $T_c = 0.14$, and the filling factor is 0.4. (The results are not sensitive to the change of parameters.)

small linear term in ρ_s^c, with a slope proportional to $(t_\perp^{\rm node})^2$, which dominates the c-axis superfluid response when $T \ll t_\perp^{\rm node}$.

Sufficiently close to a nodal line the c-axis hopping rate always becomes small in comparison with the impurity scattering rate, and thus disorder is always a relevant perturbation. Disorder introduces a finite quasiparticle lifetime and consequently a finite density of states at the Fermi level. At low temperature the correction to the superfluid density caused by finite quasiparticle scattering rates is approximately given by

$$\delta\rho_s^\mu \sim -\frac{2}{\pi}\sum_k\left(\frac{\partial\varepsilon_k}{\partial k_\mu}\right)^2 \times\int_{-\infty}^{\infty} d\omega f(\omega)\ {\rm Im}\,\frac{(\omega+i\Gamma)^2+\lambda_k^2}{[(\omega+i\Gamma)^2-\lambda_k^2]^2}. \quad (4)$$

It can be shown that both the ab plane and the c-axis superfluid responses behave as T^2 when T is much smaller than the quasiparticle scattering rate Γ [17]. However, the coefficients of the T^2 terms are very different for the ab plane and the c-axis responses. On the ab plane, $\rho_s^{ab}(T)/\rho_s^{ab}(0) \sim -(\Delta_0/\Gamma)\,(T/\Delta_0)^2$, while along the c axis, $\rho_s^c(T)/\rho_s^c(0) \sim -(\Gamma/\Delta_0)\,(T/\Delta_0)^2$. Thus the T^2 behavior of the c-axis response is by a factor $(\Gamma/\Delta_0)^2$ weaker than that on the ab plane (generally $\Gamma \ll \Delta_0$) due to the μ_k^2 factor in $t_\perp(k_\parallel)$. Notice that disorder has opposite effects in ab and c axis; it weakens the T dependence of the in-plane response and strengthens the T dependence of the out of plane response.

Another important aspect of disorder is that it may disrupt the symmetry of bonding orbitals about the copper site. The selection rule preventing hopping for momentum along $k_x = \pm k_y$ is then relaxed. For example, consider the effect of interlayer defects on O site. This leads to a random component of $t_\perp$ due to random variation in the matrix element M_0 for a Zhang-Rice singlet discussed earlier. Near the gap nodes this fluctuating component in $t_\perp$ dominates c-axis hopping, and thus electrons in the vicinity of nodes are well described by an impurity assisted hopping model which has been introduced in the literature on phenomenological grounds [7]. Impurity assisted hopping gives a new conduction channel and has a direct contribution to the c-axis superfluid density $\rho_{s,\rm imp}^c$,

$$\rho_{s,\rm imp}^c \sim \frac{4}{\beta}\sum_{\omega_n,k,k'}\langle\delta t_\perp(k)\delta t_\perp(-k)\rangle_{\rm imp} \times{\rm Tr}\Big[\sin^2\frac{k_z}{2}\,G(k+k',\omega_n)G(k',\omega_n) - \cos^2\frac{k_z}{2}\,G(k+k',\omega_n)\tau_3 G(k',\omega_n)\tau_3\Big]. \quad (5)$$

Thus the superfluid response induced by the impurity assisted hopping $\rho_{s,\rm imp}^c$ depends on the impurity average of the assisted hopping matrix element $\langle\delta t_\perp(q)\delta t_\perp(-q)\rangle_{\rm imp}$. If the impurity scattering is isotropic in space, i.e., $\langle\delta t_\perp(q)\delta t_\perp(-q)\rangle_{\rm imp}$ is independent on q, then the assisted hopping contribution to ρ_s^c in a d-wave state is zero, resulting from the vanishing average of the d-wave gap on Fermi surface. If, however, the scattering is anisotropic, for example, if $\langle\delta t_\perp(q)\delta t_\perp(-q)\rangle_{\rm imp}$ has the Lorentzian form $\langle\delta t_\perp(q)\delta t_\perp(-q)\rangle_{\rm imp} = v_0^2 k_F\delta k[(\mathbf{k}-\mathbf{k}')^2+(\delta k)^2]$, [7] then in the strong forward scattering limit ($\delta k \to 0$) $\rho_{s,\rm imp}^c$ behaves as T^2 at low temperature,

$$\rho_{s,\rm imp}^c \propto \Gamma_\perp\Delta N(0)\left[1-8\ln 2\left(\frac{T}{\Delta}\right)^2+o(T^2)\right], \quad (6)$$

where $\Gamma_\perp = 2\pi v_0^2 N(0)$. For a small but finite δk, $\rho_{s,\rm imp}^c$ behaves similarly to the $\delta k \to 0$ case, but the overall amplitude of $\rho_{s,\rm imp}^c$ decreases with increasing δk.

The above discussion indicates that both the lifetime effect and the impurity assisted hopping contribute a T^2 term to ρ_s^c at low temperature. The T^5 behavior of ρ_s^c from the coherent tunneling of electrons is therefore observable only when $T_c \gg T \gg \max(\Gamma, T^*)$, where $T^* \sim \frac{1}{2}[\Gamma_\perp\Delta_0/(t_\perp^0)^2]^{1/3}T_c$ is a characteristic temperature above which the contribution from the coherent hopping of electrons to ρ_s^c is larger than that from the impurity assisted hopping. For a highly anisotropic and clean high-T_c superconductor, generally $\Gamma_\perp \ll t_\perp^0$ and $t_\perp^0$ is smaller than or of the same order as Δ_0. However, because of the $1/3$ exponent in T^*, T^* is a rather large temperature scale compared with even Γ. Thus assisted hopping may affect the c-axis superfluid response in a larger temperature region than the scattering lifetime effect.

Summarizing the discussion above, we may draw a qualitative phase diagram for the temperature dependence of ρ_s^c and ρ_s^{ab} at low temperature: When $T \ll \Gamma$, the lifetime effect is important; both ρ^{ab} and ρ_s^c vary quadratically with T. When $T^* \gg T \gg \Gamma$, the impurity assisted hopping becomes more important to the c-axis superfluid response. In this case, ρ_s^c varies still quadratically with T, but the coefficient of the T^2 term in $\rho_s^c(T)/\rho_s^c(0)$ is proportional to $\Gamma_\perp/(t_\perp^0)^2\Delta$, which is generally smaller than that in the $T \ll \Gamma$ case where the coefficient of T^2 term in $\rho_s^c(T)/\rho_s^c(0)$ is proportional to Γ/Δ^3. When $T_c \gg T \gg \Gamma$ for ρ_s^{ab} or $T_c \gg T \gg T^*$ for ρ_s^c, the disorder effect is not so important and the temperature dependences of ρ_s^c and ρ_s^{ab} in a clean system should be recovered. Thus there is a crossover from T^2 to T when $T \sim \Gamma$ for ρ_s^{ab} and from T^2 to T^5 when $T \sim T^*$ for ρ_s^c as T increases at low temperature [18]. A simple formula which gives a qualitative measure for the crossover of ρ_s^c from the intrinsic T^5 behavior to the disorder T^2 behavior is $\rho_s^c(T) \approx \rho_s^c(0) + \alpha T^2[1+(T/T^*)^3]$ with α a scattering potential dependent constant. The above picture may be also valid for the case of substitutional impurities such as Zn, but we cannot rule out a possible formation of an "impurity band" which may have a stronger effect on the low temperature behavior of ρ_s^c than the disorder effects previously discussed.

In comparing the predictions of this model to experiment for a given system, it is essential to assess whether coherent Bloch bands are present at all. The answer to this question in the normal state of highly anisotropic copper oxides, such as BSCCO, appears to be no. The c-axis mean free path is generally very short compared with the c-axis lattice constant. Microwave [19] and thermal Hall measurements [20], however, reveal that the mean free path grows very rapidly in the superconducting state. The superconducting quasiparticle scattering rate is therefore much lower than the extrapolated normal state scattering rate. Thus it is our belief that the available experimental data favor coherent Bloch band formation along c axis at low temperature below T_c. For BSCCO, $t_\perp^0$ is small and ρ_s^c might be dominated by the impurity assisted hopping [7]. However, for other compounds with lattice tetragonal symmetry and relatively lower anisotropy, such as LaSrCuO, TlBaCaCuO, and HgBaCuO, we believe that the coherent band should have substantial contribution to ρ_s^c. Experimentally, a crucial test for this can be done by carefully measuring and analyzing ρ_s^c data in these compounds to see if the predicted T^5 behavior exists.

YBCO compounds (for both $YBa_2Cu_3O_{7-\delta}$ and $YBa_2Cu_4O_8$) contain both CuO_2 planes and CuO chains. In these materials, the c-axis hopping integral between two CuO_2 layers within a cell as well as that between a CuO chain layer and a CuO_2 layer deviates noticeably from the behavior $t_\perp \propto (\cos k_x - \cos k_y)^2$ and is finite around the $d_{x^2-y^2}$-wave gap nodes due to the hybridization between CuO chains and CuO_2 planes and the one-dimensionality of CuO chains [9]. Thus the T^5 behavior in ρ_s^c is not applicable to YBCO. The impurity assisted hopping may have a substantial contribution to ρ_s^c in $YBa_2Cu_4O_8$, but it may not be so important in the fully oxygenated $YBa_2Cu_3O_{7-\delta}$ To understand the properties of the c-axis superfluid response in YBCO, a comprehensive study for the electronic structures of CuO_2 planes as well as CuO chains is needed [21].

In conclusion, we have demonstrated that there exists a remarkable interplay between underlying electronic structure and orbital parameter symmetry in copper oxides. The c-axis electromagnetic response reflects not only the symmetry of the order parameter but also the relative symmetry of the copper orbitals involved in c-axis and ab-plane conduction. At low temperature the disorder effect dominates and both ρ_s^c and ρ_s^{ab} have T^2 dependence. Above some characteristic temperatures, which may be different for ρ_s^c and ρ_s^{ab}, the temperature dependences in a clean system, linear T for ρ_s^{ab} and T^5 for ρ_s^c, are expected.

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