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# *c*-axis penetration depth and interlayer conductivity in the thallium-based cuprate superconductors

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The $c$-axis Josephson plasmons in optimally doped single-layer and bilayer high-$T_c$ cuprates $Tl_2Ba_2CuO_6$ and $Tl_2Ba_2CaCu_2O_8$ have been investigated using infrared spectroscopy. We observed the plasma frequencies for these two compounds at 27.8 and 25.6 $cm^{-1}$ respectively, which we interpret as Josephson resonances across the TlO blocking layers. No maximum in the temperature dependence of the $c$-axis conductivity was observed below $T_c$, indicating that even in the superconducting state a coherent quasiparticle contribution to the $c$-axis conductivity is absent or very weak. [S0163-1829(99)51346-5]

Studies of the $c$-axis properties in high-$T_c$ superconductors are of considerable importance. Although the materials are highly anisotropic, with much of the physics being of a two-dimensional nature, it is clear that an understanding of both the $c$-axis and $ab$-plane behavior is relevant to the quest for the mechanism of high-$T_c$ superconductivity. It is already well established that the normal-state $c$-axis transport in these materials is strongly incoherent. A number of theories treat this problem, and generally they can be divided into two categories, i.e., Fermi-liquid and non-Fermi-liquid approaches. The Fermi-liquid approaches rely on special properties of the quasiparticle scattering, e.g., a strong momentum dependence along the quasi-two-dimensional Fermi surface of the quasiparticles.[1] Within the non-Fermi-liquid approaches, the transport processes involve particles carrying unconventional spin and charge quantum numbers. One of the most radical approaches, based on spin-charge separation in the copper-oxide planes, has resulted in the notion of ''confinement'' of single charge carriers to the copper-oxide planes in the normal state.[2] Within the latter class of models the formation of Cooper pairs is accompanied by the deconfinement of those pairs, resulting in a center-of-mass kinetic-energy gain. In principle this also provides a mechanism for superconductivity,[3] and a fundamental experimental test of this hypothesis was proposed by Anderson.[4] The experimental results indicated that interlayer tunneling of pairs is *not* the main mechanism for superconductivity, at least in $Tl_2Ba_2CuO_6$.[5–7] This immediately raises the question of whether the incoherent $c$-axis transport arises from the two-dimensional confinement of single charge carriers to the copper-oxide planes, and whether or not the confinement persists in the superconducting state.

From an analysis of the infrared reflectivity spectra using the two-fluid model, Tamasaku *et al.*[8] have concluded that for $T<T_c$ the $c$-axis quasiparticle scattering rate of $La_{2-x}Sr_xCuO_4$ (LSCO) for $T<T_c$ decreases strongly with temperature, and ''the $T$ dependence looks very similar to that for the quasiparticle scattering rate in the $ab$ plane.'' This result indicated that coherent quasiparticle transport is recovered simultaneously with the occurrence of a finite critical current along the $c$ direction. A different result was later obtained by Kim *et al.*[9] Experimentally the $c$-axis transport was monitored via the temperature dependence of the real part of the conductivity, $\sigma_c(\omega)$, which exhibited a sharp drop below $T_c$. The drop in $\sigma_c(\omega)$ was attributed ''exclusively to the opening of a gap, i.e., *without* a change of the electronic scattering rate'' of the carriers along the $c$ direction.[9] Later, a similar behavior of $\sigma_c(\omega)$ versus temperature was reported for $Bi_2Sr_2CaCu_2O_{8+\delta}$,[10,11] and $YBa_2Cu_3O_{7-\delta}$ (YBCO).[12,13]

In this paper we address the issue of ''confinement'' experimentally. We study the degree of coherence of $c$-axis quasiparticle transport in the superconducting state, the temperature dependence of the $c$-axis plasma frequency, and the role of intrabilayer splitting by comparing the single-layer and bilayer materials within the same family of Tl-based cuprates. Here we report data on epitaxially grown $Tl_2Ba_2CuO_6$ and $Tl_2Ba_2CaCu_2O_8$ thin films, with superconducting transition temperatures of 80 and 98 K, respectively, established by susceptibility measurements. Details of the sample preparation and growth methods have been reported elsewhere.[14–16] Dimensions of the films in the $ab$ plane are typically 50–100 $mm^2$. We measured grazing incidence reflectivity in the far-infrared (FIR) region ($17-700$ $cm^{-1}$) using a Fourier transform spectrometer. A grazing angle of 80° was chosen in order to predominantly probe the $c$-axis response for $p$ polarization of the light. Absolute reflectivi-

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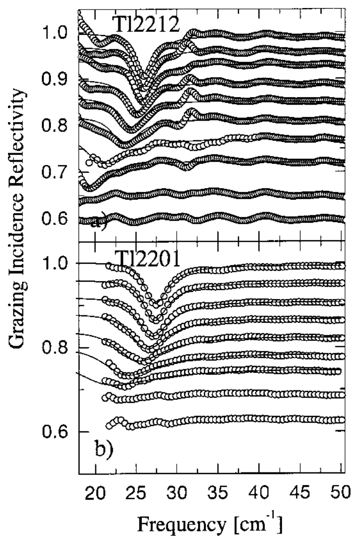


FIG. 1. (a) Grazing incidence reflectivity (open circles) of Tl2212. From top to bottom: 4, 10, 20, 30, 40, 50, 60, 70, 80, and 110 K. The curves have been offset for clarity. The solid curves are fits to the data. (b) Grazing incidence reflectivity (open circles) of Tl2201, at 4, 10, 20, 30, 40, 50, 60, 75, and 90 K. The solid curves are fits to the data.

ties were obtained by calibrating the reflectivities against the reflectivity of a gold film deposited *in situ* on the sample without moving or rotating the sample holder.

In Fig. 1 we display the grazing incidence reflectivity of Tl2201 [Fig. 1(b)] and Tl2212 [Fig. 1(a)] in the frequency region from 17–50 $cm^{-1}$ for various temperatures below $T_c$. The spectra above 50 $cm^{-1}$ are dominated by the $c$-axis phonons, and a detailed analysis is presented elsewhere.[17] The only absorption below 50 $cm^{-1}$ corresponds to the Josephson plasmon, a collective oscillation of the Cooper pairs along the $c$ axis. The observed resonant frequencies ($\omega_J$) at 4 K are 27.8 $cm^{-1}$ in Tl2201 and 25.6 $cm^{-1}$ in Tl2212, and they redshift as the temperature approaches $T_c$ from below. These values correspond to $c$-axis penetration depths of $\lambda_c = 17.0 \pm 0.3$ $\mu$m ($\varepsilon_S = 11.3 \pm 0.5$) and $\lambda_c = 20.6 \pm 0.6$ $\mu$m ($\varepsilon_S = 9.1 \pm 0.7$), respectively.[17] In Fig. 2 we present $p$-polarized reflectivity for various temperatures below $T_c$, normalized to the 110 K spectrum (which is essentially featureless). A magnetic field of 0.35 T was sufficient to shift the resonance out of our spectral window. The strong redshift upon applying an external magnetic field confirms that the absorption corresponds to the Josephson plasmon. Let us first address the question of why $\omega_J$ is almost the same in Tl2201 and Tl2212: In a model of superconducting planes weakly coupled by Josephson coupling, a separate resonance frequency is associated with each link.[18] As the only structural difference between the two materials is the insertion of an extra $CuO_2$ layer and a layer of $Ba^{2+}$ ions separating the two $CuO_2$ layers, the same resonance frequency should appear for the link across the thallium oxide blocking layer. Extrinsic variations in the Josephson coupling could arise as a result of stacking faults or extrinsic variations of the stoichiometry, and result in an inhomogeneous broadening of the plasma resonance.[18] Hence the main plasma resonance in our data is not affected by spurious weak links in the system.

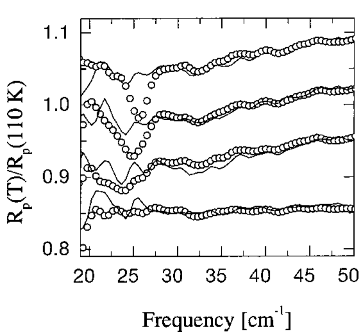


FIG. 2. $p$-polarized reflectivity normalized to the spectrum at 110 with magnetic field (solid curve) and without magnetic field (open circles). From top to bottom: 4, 30, 60, and 90 K.

We analyze the spectra using the Fresnel formula for uniaxial crystals.[5] The resonance is due to the zero crossing of $\varepsilon_c(\omega)$, for which we adopt the standard two-fluid expression appropriate for the superconducting state

$$\varepsilon_c(\omega) = \varepsilon_S(\omega) - \frac{c^2}{\lambda_c(T)^2 \omega^2} + \frac{4\pi i \sigma_{qp}(\omega)}{\omega}, \tag{1}$$

where $\lambda_c$ is the $c$-axis penetration depth. Assuming the Josephson model to be applicable to this case, the pole strength of the second (London) term, coming from the condensate, provides directly the critical current in the $c$ direction. Hence $1/\lambda_c(T)^2$ represents the superfluid density appropriate for the $c$ direction. Oscillations of the condensate are electromagnetically coupled to interband transitions and lattice vibrations, which are represented by the dielectric function $\varepsilon_S$. The third term arises from currents due to thermally activated quasiparticles. The condition for the propagation of longitudinal modes in the medium along the $c$ direction in the long-wavelength limit, is that $\varepsilon_c(\omega) = 0$, which occurs at the Josephson plasma frequency $\omega_J = c/\{\lambda_c \sqrt{\mathrm{Re}\, \varepsilon_S(\omega_J)}\}$. This is also the frequency of long-wavelenth plasma polaritons propagating along the planes, which one observes in optical experiments. The $c$-axis optical conductivity at the screened plasma frequency $\sigma_{qp}(\omega_J)$ determines the width of the plasma resonance in our spectra.[5] This follows from the fact that the resonance line shape is given by

$$R_p(\omega) \approx 1 - \mathrm{Im}\, e^{i\phi} \sqrt{1 - \frac{\sin^2\theta/\mathrm{Re}\,\varepsilon_S}{\omega(\omega + i\Gamma) - \omega_J(T)^2}}, \tag{2}$$

where $\theta$ is the angle of incidence, and $\phi = (\pi - \arg \varepsilon_{ab})/2$ is a weakly frequency dependent phase factor, ranging from $\phi = 0$ (ideal superconducting response) to $\phi = \pi/4$ (metallic response), and $\Gamma = 4\pi\, \mathrm{Re}\, \sigma_{qp}(\omega_J)/\mathrm{Re}\, \varepsilon_S(\omega_J)$ determines the resonance linewidth. In this expression the imaginary part of $\sigma_{qp}$ has to be included in $\mathrm{Re}\, \varepsilon_S$, and vice versa. We see now that the linewidth of the $c$-axis plasma resonance is directly proportional to $\mathrm{Re}\, \sigma_{qp}$ at the resonance position. This does *not* imply a direct proportionality of the plasma resonance linewidth to the quasiparticle scattering rate. In fact assuming here a Drude line shape of the quasiparticle term, $\mathrm{Re}\, \sigma_{qp} = ne^2 m_\perp^{-1} \tau/(1 + \omega^2\tau^2)$, the plasma resonance linewidth is proportional to $n_{qp}/\tau$ if $\omega_J\tau \gg 1$ and to $n_{qp}\tau$ if $\omega_J\tau \ll 1$. In LSCO,[8,9] YBCO,[19] and recently Tl2201,[20] where crystals were available suitable for reflection spectroscopy on the ac-crystal face, it can be seen directly from the con-

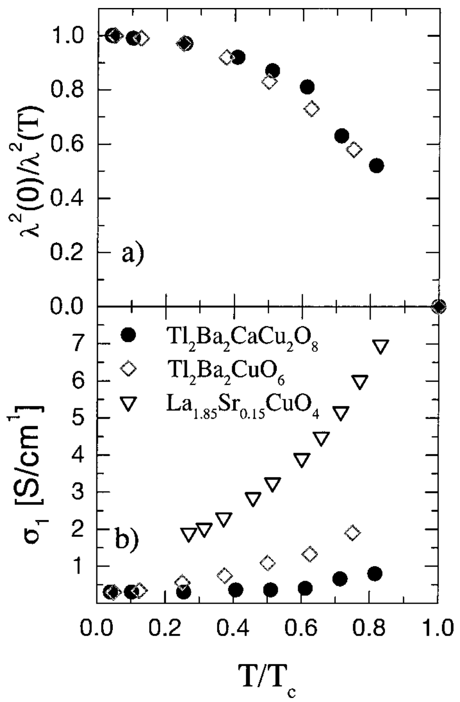


FIG. 3. ( a) Temperature dependence of the $c$-axis penetration depth of Tl2201 (open diamonds) and Tl2212 (solid circles). (b) The temperature dependence of the $c$-axis optical conductivity of Tl2201, Tl2212, and LSCO.

ductivity spectra that $\sigma_c(\omega)$ is essentially independent of frequency in the superconducting state for frequencies below $\simeq 100\ \mathrm{cm}^{-1}$. This corresponds to the limit where $\mathrm{Re}\,\sigma_{qp} \propto n_{qp}\tau$. Hence the drop of the plasma-resonance linewidth below the phase transition represents the reduction of quasiparticle density at the Fermi energy due to the opening of the superconducting gap.[8] By fitting Eq. (1) to the measured spectra, using the full Fresnel expression for the grazing reflectivity, we obtain accurate values of the $c$-axis superfluid density, $\lambda^2(0)/\lambda^2(T)$, and the optical conductivity at the resonance frequency, $\sigma_1(\omega_J)$, as a function of temperature.

In Fig. 3 we present the temperature dependence of the $c$-axis superfluid density, $\lambda^2(0)/\lambda^2(T)$, and the $c$-axis superfluid density of LSCO of Ref. 9. Fitting the $c$-axis superfluid density to a power law $\lambda^2(0)/\lambda^2(T) = 1-(T/T_0)^\eta$ for temperatures below $0.5T_c$, we obtain an exponent $\eta = 2.1 \pm 0.1$ for Tl2201 and $\eta = 2.4 \pm 0.1$ for Tl2212. Hence our results show that there is also no linear temperature dependence for the $c$-axis superfluid density in Tl2201 and Tl2212, as has been observed in several other high-$T_c$ materials.[11–13] The behavior in both Tl-based compounds is close to quadratic.

In Fig. 3(b) we present the $c$-axis optical conductivity versus temperature for Tl2201, Tl2212, and LSCO. In all compounds $\sigma_c$ exhibits a smooth drop below $T_c$. The drop in $\sigma_c(T)$ has also been observed in YBCO and Bi2212, and is very different from the temperature dependence of the $ab$-plane conductivity, where $\sigma_{ab}(T)$ shows a large peak below $T_c$, attributed to a rapid increase in the quasiparticle lifetime. The absence of a subsequent rise of $\sigma_c$ at lower temperature indicates that effectively we observe no enhancement of the quasiparticle lifetime for this transport direction.

Recently Xiang and Wheatley[21] calculated the $c$-axis response assuming $d$-wave pairing, and assuming a model for the $c$-axis transport where momentum parallel to the planes, $k_\parallel$, is conserved. Motivated by the work of Chakravarty *et al.*,[22] they argued that for high-temperature superconductors (HTSC's) with a simple (non-body-centered) tetragonal structure, $t_\perp \propto [\cos(k_x a)-\cos(k_y a)]^2$ (see also Ref. 23), leading to an exponent $\eta = 5$ at low temperature. Allowing some scrambling of $k_\parallel$ due to impurity scattering during the interplane hopping reduces this to $\eta = 2$ at very low temperatures, crossing over to the $T^5$ above a temperature which depends on the impurity scattering rate.

Within the same line of thinking, Ioffe and Millis[1] recently proposed that the scattering lifetime has a strong dependence on $k_\parallel$ along the two-dimensional Fermi surface, namely

$$\Gamma(k_\parallel) = \frac{\Gamma_0}{4}\sin^2(2\Theta) + \frac{1}{\tau}, \qquad (3)$$

where $\Theta$ is the angle of $k_\parallel$ relative to the diagonal (''nodal'') direction. For the in-plane response this leads to an expression for the optical conductivity which fits the in-plane experimental spectra rather well, and has the prototypical property of the optimally doped cuprates that the effective scattering rate $1/\tau^* \equiv \omega\,\mathrm{Re}\,\sigma/\mathrm{Im}\,\sigma$ has a linear frequency dependence if $\Gamma_0$ is assumed to be temperature independent, while $1/\tau \propto T^2$.

For the $c$ direction, we must take into account, that $t_\perp \propto [\cos(k_x a)-\cos(k_y a)]^2$, as has been done by Xiang and Wheatley.[21] The $c$-axis transport arises in this picture from regions at the Fermi surface well removed from the nodes of the gap. Due to the strong suppression of $t_\perp^2$ along the nodal directions, $\sigma_c(\omega)$ probes only regions away from the nodes, closer to the regions of maximum gap value. [In those regions of $k$ space $\Gamma(k_\parallel)$ is large, of the order of 1 eV.] This places the $c$-axis optical conductivity well inside the dirty limit,[24] and it provides a simple microscopic argument as to why an analysis of the $c$-axis optical conductivity in the superconducting state, using the Mattis-Bardeen expressions for $s$-wave superconductors, compares so well to the experimental data.[9]

The question of how to understand the large difference in scattering properties and gap observed along two different optical axes, has thus been shifted to a strong dependence of the scattering rate on the position along the Fermi surface (which is a *phenomenological* assumption) and a strong dependence of the hopping parameter on $k_\parallel$ (which is a straightforward result of band theory). The ''confinement'' in the sense of an anomalous renormalization of the transport properties due to many-body effects, is in this picture associated with the antinodal regions in $k$ space. Near the nodal regions the confinement is dominated by a straightforward single-particle effect on $t_\perp(k_\parallel)$, which follows directly from band theory. However, the anticorrelation between these two kinds of confinement seems hardly a coincidence, and certainly deserves further attention. This leaves open the question of the microscopic implications of this phenomenological model. The correspondence between $\Delta(k_\parallel)$ and $t_\perp$ has been previously attributed to the interlayer tunneling mechanism.[3] Likewise the correspondence between $\Delta(k_\parallel)$ and $\Gamma(k_\parallel)$ may contain important clues regarding the pairing mechanism leading to superconductivity.

In conclusion, we have observed the *c*-axis Josephson plasma resonance in Tl2201 and Tl2212. The values of the resonant frequencies in the two compounds are quite close, which indicates that the weakest link is the intercell link. In a magnetic field of 0.35 T the resonance is completely pushed out of our spectral window, which is the expected behavior for the Josephson collective exitation. From our data, we were able to extract the *c*-axis superfluid density, and conductivity $\sigma_c(T)$. The temperature dependence of both quantities indicates the two-dimensional confinement of the charge carriers.

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