c-Axis Electronic Raman Scattering in $Bi_2Sr_2CaCu_2O_{8+\delta}$

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We report on the c-axis-polarized electronic Raman scattering of $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystals with various oxygen concentrations. In the normal state, a resonant electronic continuum extends to 1.5 eV and gains significant intensity as the incoming photon energy increases. Below T_c, a superconductivity-induced 2Δ peak is observed for $\omega < 80$ meV and the $2\Delta/k_BT_c$ value increases with decreasing hole doping. In particular, this A_{1g} 2Δ peak energy, which is higher than that seen with in-plane polarizations for all doping levels studied, signifies distinctly different dynamics of quasiparticles created with out-of-plane polarization. [S0031-9007(99)08952-8]

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The peculiar aspects of the c-axis transport in the high-T_c cuprates continue to attract much attention [1]. Early on it was established experimentally that the normal-state in-plane resistivity typically varies linearly with temperature, whereas the out-of-plane resistivity almost universally displays semiconducting behavior [2]. The c-axis optical conductivity of the most cuprates [3,4] shows an electronic background with a very large scattering rate—that is, the mean free path appears to be less than the lattice spacing. These results suggest that there is no coherent electronic transport in the c direction: all motions are inelastic. A number of models have been proposed to explain the important mechanisms contributing to c-axis transport in high-T_c superconductors [1], although controversy remains [5].

Recent experiments have provided new insights about the c-axis electrodynamics of cuprates. Analysis of the interlayer infrared conductivity of $Tl_2Ba_2CuO_{6+\delta}$, $La_{2-x}Sr_xCuO_4$, and $YBa_2Cu_3O_{6.6}$ reveals an anomalously large energy scale extending up to midinfrared frequencies that can be attributed to the formation of the superconducting condensate [6]. New measurements of surface impedance in $YBa_2Cu_3O_{7-\delta}$ show that the c-axis penetration depth never has the linear temperature dependence seen in the ab plane [7]. Images of interlayer vortices in $Tl_2Ba_2CuO_{6+\delta}$ and $HgBa_2CuO_{4+\delta}$ give a value of c-axis penetration depth of a factor of 10–20 longer than that predicted by the interlayer tunneling model, indicating the condensation energy available through this mechanism is much smaller than is required for superconductivity [8].

The purpose of this study is to investigate, in the context of electronic Raman scattering spectroscopy, the role of c-axis polarizability in the $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi-2212) cuprates. Among all cuprate superconductors, the Bi-2212 system is special in many respects. Most notably, Bi-2212 has the extremely small energy scale set by the hopping interaction between the adjacent CuO_2 bilayers [1]. This two-dimensional character of Bi-2212 also manifests itself in the large c-axis penetration depth [9]. The large anisotropy between λ_{ab} and λ_c of Bi-2212 was shown to be best described within a picture of strongly superconducting CuO_2 layers weakly coupled by Josephson interaction along the c axis [10].

Raman scattering has been proved to be a valuable technique for understanding the quasiparticle dynamics on different regions of the Fermi surface in the cuprate systems by orienting incoming and outgoing photon polarizations [11]. The electronic Raman spectra polarized in the ab plane of Bi-2212 have been extensively studied [12]. In contrast, Raman data on the electronic scattering of Bi-2212 for photons polarized along the c axis are rare [13]. Our new results show that the zz-polarized electronic continuum of Bi-2212 extends to 1.5 eV and its intensity resonates towards near ultraviolet (UV) photon excitation. In the superconducting state, there is a low-energy redistribution of the electronic continuum and the presence of a 2Δ peaklike feature for all doping levels studied. Most importantly, the greater value of this A_{1g} 2Δ peak energy, relative to the in-plane cases, signifies that different quasiparticle dynamics are involved.

Single crystals of Bi-2212 were grown using a solvent-free floating zone process. We have repeatedly studied the same single crystal of dimensions $5 \times 1 \times 0.5$ mm^3 after successive annealing steps under controlled oxygen partial pressure. The superconducting transition determined by a dc magnetization measurement showed the sample to be successively overdoped (onset $T_c = 84$ K, $\Delta T_c = 4$ K), slightly overdoped ($T_c = 92$ K, $\Delta T_c = 2$ K), optimally doped ($T_c = 95$ K, $\Delta T_c = 1$ K), and underdoped ($T_c = 87$ K, $\Delta T_c = 4$ K). The Raman measurements were performed on two faces of the crystal. One face (labeled I in the inset of Fig. 1) contains both the c axis and either the a or b direction. The second face, face II, provided the

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a- and b-axis response. Throughout this study, x and y are indexed along the Bi-O bonds, rotated by 45° with respect to the Cu-O bonds. All symmetries refer to a tetragonal D_{4h} point group.

The low-frequency Raman spectra were taken in pseudobackscattering geometry with $\hbar\omega_i = 1.92$ eV photons from a Kr^+ laser. The laser excitation of less than 10 W/cm^2 was focused into a 50 μm diameter spot on the sample surface. The temperatures referred to in this paper are the nominal temperatures inside the cryostat. The spectra were analyzed by a triple grating spectrometer with a liquid-nitrogen cooled charge-coupled device detector. To investigate further the resonance property, we have used several excitation lines from Ar^+ and Kr^+ lasers ranging from visible red to near UV. All the Raman spectra were corrected for the spectral response of the spectrometer and detector, the optical absorption of the sample as well as the refraction at the sample-gas interface [14].

The inset of Fig. 1(a) shows the imaginary part of the c-axis-polarized Raman response function, obtained by dividing the original spectrum by the Bose-Einstein thermal factor, for the overdoped Bi-2212 crystal at 300 K. The spectrum is composed of a very weak electronic background and several $q \approx 0$ Raman allowed phonon modes, whose overall character is in good agreement with that reported previously [15–17]. In this paper, we will concentrate primarily on the resonant behavior and temperature dependence of the electronic Raman scattering response.

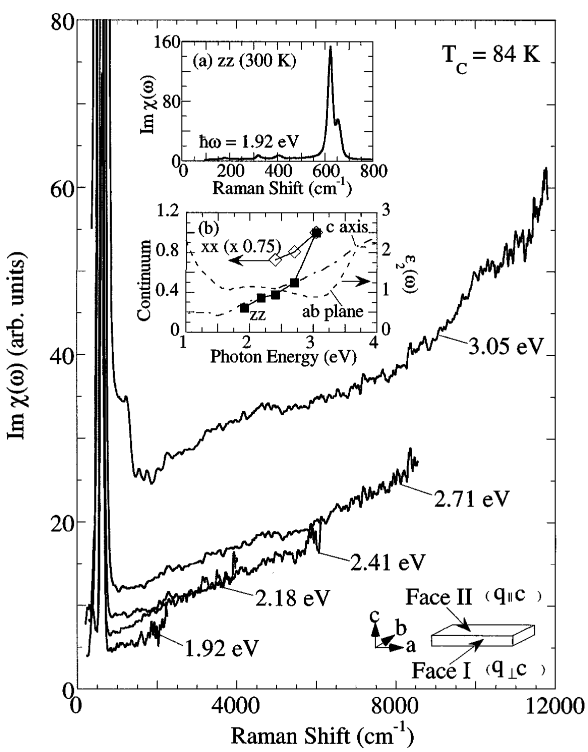

FIG. 1. Room-temperature c-axis-polarized Raman scattering for the overdoped sample with $T_c = 84$ K as a function of incident photon energy. The inset in the lower-right corner shows the sample faces that were used to measure Raman spectra. The inset (a) shows low-frequency Raman response functions in the zz polarization using red (1.92 eV) excitation at 300 K. The inset (b) illustrates the normalized resonance Raman excitation profile of the zz (filled square) and xx (face I) (open diamond) continua compared with the imaginary part of the c axis (dash-dotted line) and ab-plane dielectric constant (dashed line) [3].

In the main plot of Fig. 1 we show an expanded scale room-temperature c-axis Raman spectra excited with photon energies between 1.92 and 3.05 eV. For the latter incident energy, we follow the continuum to 1.5 eV Raman shift. The continuum intensity exhibits a dramatic increase when the excitation approaches the UV region. This is more clearly seen in the inset of Fig. 1(b) where we plot the resonance profile for the zz- and xx-polarized (face I) continua. The continuum intensity was measured at around 2000 cm^{-1} and normalized to its value at 3.05 eV. We note that the zz continuum gains intensity by a factor of $\sim$5 towards UV excitation and follows approximately the trend of the imaginary part of the c-axis dielectric constant [3], suggesting that light couples to the c-axis continuum via some intermediate excitation states with energy $>$3 eV [18]. In contrast, the change of the xx continuum intensity is rather modest. The substantially different resonance behavior of the zz and xx continua is in qualitative agreement with what one might expect from the strong anisotropy of the frequency-dependent Raman vertex [19], which can be interpreted as an inverse frequency-dependent "effective mass": If one were to extrapolate the resonance data to zero frequency, the value of the out-of-plane continuum scattering efficiency would be close to zero and significantly smaller than that of the in-plane symmetry. This is consistent with a great anisotropy in the carrier effective mass for Bi-2212, and supported by the transport and band structure studies [20,21]—that is, $m_{ab}^*(\omega = 0) \ll m_c^*(\omega = 0)$.

When the sample is cooled, the most interesting change below T_c occurs in the low-frequency portion of the electronic Raman response. As seen in the first panel of Fig. 2(a) for $T \ll T_c$, there is a decrease in the scattering strength of the c-axis continuum at low energies which redistributes into the weak and broad peak at higher frequencies, similar to the data (so-called coherent "2Δ peak") observed in the ab plane. These data were reproducibly observed at three different spots on the sample. We emphasize that the redistribution of the scattering intensity itself is not of phononic origin. The low-energy (red) excitation is used primarily to reduce the intensity of phononic scattering. Furthermore, a similar superconductivity-induced renormalization of the electronic continuum along the c direction was also observed in other less anisotropic members of the high-T_c superconductors, including $YBa_2Cu_3O_{7-\delta}$ [22] and $NdBa_2Cu_3O_{7-\delta}$ [23].

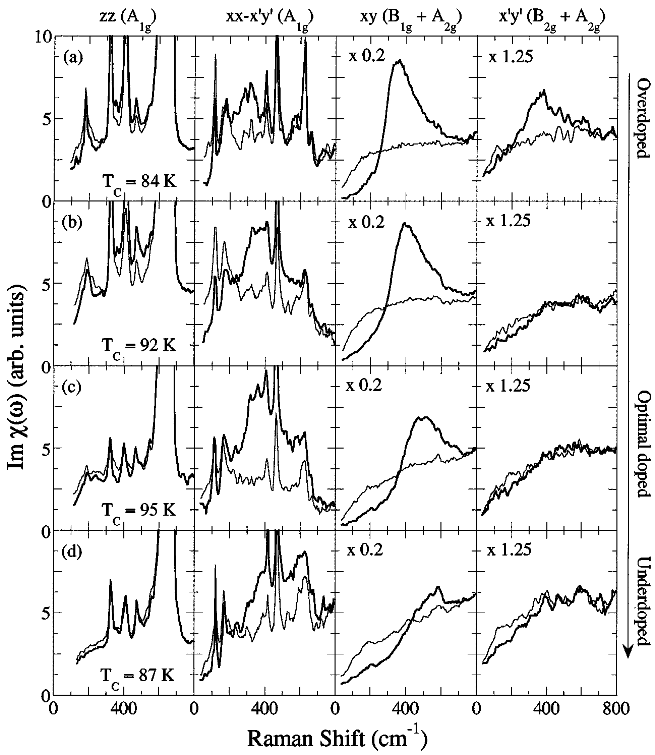

FIG. 2. The low-energy portion of the Raman scattering spectra taken with four different polarizations and using red (1.92 eV) excitation as a function of doping (a) for overdoped sample with $T_c = 84$ K, (b) for slightly overdoped sample with $T_c = 92$ K, (c) for optimal doped sample with $T_c = 95$ K, and (d) for underdoped sample with $T_c = 87$ K. The thick line denotes the spectra taken at 5 K and the thin line at 100 K.

It is instructive to compare the behavior of the low-frequency scattering of the out-of-plane A_{1g} symmetry component with those for the in-plane scattering configurations, which are shown in the remaining panels of Fig. 2(a). With behavior similar to that reported in prior work on overdoped Bi-2212 [12], the superconducting transition leads to the redistribution of the continuum into a broad peak for the in-plane A_{1g} polarization. The B_{1g} contribution is predominant in xy geometry, and gives an electronic continuum that is much stronger than that in any other polarization. Below T_c, the strong suppression of the continuum is observed, and the low-frequency intensity varies roughly as ω^3, while it is quite linear in $x'y'$ polarization. At the same time, the magnitude of the superconductivity-induced peak is much less intense in $B_{2g} + A_{2g}$ symmetry than that found in B_{1g} symmetry. Such ω dependences in both B_{1g} and B_{2g} continua are consistent with an order parameter of d-wave symmetry [$d(x^2 - y^2)$ when referred to Cu-O bonds] [24].

The effect of superconductivity on the c-axis electronic Raman scattering can be more carefully explored by studying the low-frequency part of the spectra as a function of doping for Bi-2212, shown in Figs. 2(b)–2(d). First, we notice that the characteristic loss of the c-axis electronic scattering intensity in the superconducting state appears at low frequencies for all doping levels studied. More interestingly, the frequency scale associated with the depletion of the spectral weight does change with doping. For the overdoped Bi-2212 with $T_c = 84$ K, the suppression of the electronic continuum occurs at $\omega < 300$ cm^{-1}. As the doping level is decreased towards the underdoped with $T_c = 87$ K, the onset frequency related to the changes of zz continuum is about 600 cm^{-1}. Second, we have estimated the c-axis 2Δ peak energy by representing the change between the normal and superconducting spectra in an enhanced manner (see the inset of Fig. 3), where the 5 K spectrum is normalized by (1) dividing by the 100 K spectrum (top curve), (2) subtracting the 100 K spectrum (middle curve), and (3) the difference of Raman spectra, as in (2), after first subtracting the phonon contributions (bottom curve). It is clear that in all cases the 2Δ peak position for the overdoped Bi-2212 crystal in zz polarization occurs near 400 cm^{-1} (50 meV).

Figure 3 plots the positions of the c-axis 2Δ peak versus hole doping p [25], with each T_c converted to p using the empirical relation $T_c/T_{c,\max} = 1 - 82.6(p - 0.16)^2$ [26]. For comparison, the results of Fig. 2 for the in-plane scattering symmetries are included. With decreasing doping, the c-axis A_{1g} peak shifts monotonically upward in energy despite a decrease in T_c. $2\Delta/k_BT_c$ changes from a value $\sim$7 for overdoped Bi-2212 with $T_c = 84$ K to a value approaching $\sim$10 in the underdoped region. A similar trend was also observed for both the ab plane A_{1g} and B_{1g} symmetries [12], but with smaller values of $2\Delta/k_BT_c$. Why is the energy scale of the 2Δ peak associated with the out-of-plane and the in-plane responses different? We can say with some certainty that the Raman scattering process in both cases creates two quasiparticles. With in-plane polarization they are most likely on the same CuO_2 plane, where with zz polarization they are most likely on adjacent planes. The two kinds of pairs could be expected to undergo different final state interactions and screening corrections.

Based on the conventional model of light scattering from a superconductor [27,28], the results of 2Δ features in Fig. 2 can be taken as a measure of the magnitude of the superconducting gap, i.e., a pair-breaking peak [11]. In Fig. 3, we note a resemblance of the doping dependence of 2Δ peak position to twice the value of the maximum superconducting gap obtained from the tunneling spectroscopy [29] as well as angle-resolved photoemission spectroscopy (ARPES) [30] measurements of the $(\pi, 0)$ gap (the d-wave maximum) for the same batch of Bi-2212 crystals. Importantly, we find that c-axis 2Δ is higher than the highest energy 2Δ peak seen with ab-plane polarizations, but still less than twice the value of Δ from single electron spectroscopies. We believe that the two quasiparticles created in the Raman scattering process out of the superconducting condensate continue

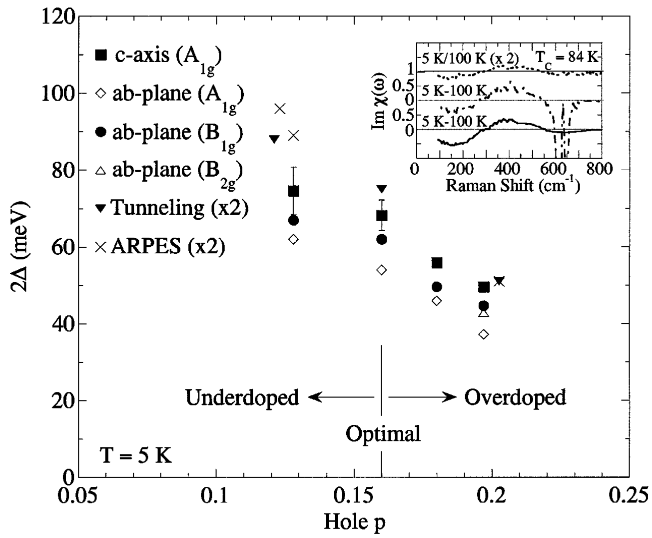

FIG. 3. Plot of the 2Δ peak energy (taken at 5 K) versus the hole concentration for four different scattering symmetries. For comparison, twice the value of Δ obtained from other experiments have also been plotted: Ref. [29] (filled triangle) from tunneling and Ref. [30] (cross) from ARPES. The inset shows the difference between 5-K and 100-K Raman spectra for the overdoped sample with $T_c = 84$ K, using the procedure described in the text.

to interact. Such final state interactions renormalize the magnitude of the superconducting gap. It costs more energy to create a quasiparticle pair in different planes than in the same plane.

In summary, c-axis electronic Raman scattering spectra have been investigated for Bi-2212 single crystals. We find that the c-axis electronic continuum extends up to very high energies and its intensity dramatically increases using UV photon excitation, suggesting that the scattering process in zz geometry is dominated by a resonance Raman vertex. Below T_c, there is a low-frequency redistribution of the electronic continuum and the formation of a 2Δ peak in the superconducting state. The monotonic increase of 2Δ and $2\Delta/k_B T_c$ with decreasing doping is unusual. Moreover, this 2Δ feature, which has A_{1g} symmetry, is found at higher frequencies than those seen in the planar symmetries for all doping levels studied. The data presented in this study pose a new challenge to those attempting to understand the important mechanisms contributing to c-axis excitations in high-T_c superconductors.

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