## Unusual Strong-Coupling Effects in the Tunneling Spectroscopy of Optimally Doped and Overdoped Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8+δ</sub>

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(Received 7 July 1997)

Tunneling spectroscopy measurements are reported on single crystals of  $Bi_2Sr_2CaCu_2O_{8+\delta}$  using vacuum tunneling and point-contact methods. A reproducible dip feature in the tunneling conductance is found near  $|eV| = 2\Delta$ , observed for *both* voltage polarities in the best resolved spectra. With overdoping the position of the dip continues to scale with  $\Delta$ , and its magnitude decreases as  $\Delta$  decreases. These results indicate that the dip feature arises from a strong-coupling effect whereby the quasiparticle lifetime is decreased at a characteristic energy of  $\sim 2\Delta$ , consistent with an electron-electron pairing interaction. [S0031-9007(97)04908-9]

PACS numbers: 74.50.+r, 74.62.Dh, 74.72.Hs

A provocative feature that has commonly been observed in the superconductor-insulator-normal (SIN) metal tunneling conductances of  $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212) is a dip at  $|eV| \sim 2\Delta$  that is very large for voltage polarities which correspond to the removal of quasiparticles from the superconductor [1-3]. This feature has generated much interest due to its similarity to a dip found in the angle-resolved photoemission (ARPES) [4,5] spectra of Bi2212 and to strong-coupling effects in general [6]. But in contrast to the tunneling phonon structures observed in strong-coupled, low- $T_c$  superconductors [6], the dip is often highly asymmetric with bias voltage. For voltages corresponding to electron injection, the dip has appeared as a shoulder in early point-contact tunneling (PCT) measurements [1] and is scarcely observable in some scanning tunneling microscope (STM) measurements [2]. We report here that the dip feature is indeed observed for both bias voltage polarities in the best resolved SIN spectra obtained from both STM and PCT methods. This points toward a strong-coupling interpretation. As  $T_c$  and  $\Delta$  are reduced by overdoping (the latter from 37 to 15 meV in this study), the dip location continues to scale with  $\Delta$  and its magnitude is reduced. The coupling to  $\Delta$  is quite unusual and suggests that the dip arises from a pairing interaction that is purely electronic so that the superconducting gap feeds back into the excitations which mediate the pairing. This is also consistent with recent interpretations of the dip in ARPES spectra as arising from the coupling of quasiparticles to collective excitations [7,8].

SIN tunneling spectroscopy is a unique probe of high temperature superconductors (HTS) in that it can, in principle, reveal the quasiparticle excitation spectrum directly with an energy resolution better than 1 meV. In conventional, *s*-wave superconductors, strong-coupling effects due to quasiparticle emission of bosons of frequency  $\omega$ 

produce a dip feature [6] in the tunneling conductance, dI/dV, near a voltage,  $eV = \Delta + \hbar \omega$ . More precisely, it is the maximum negative slope if dI/dV vs V that pinpoints the boson frequency. In d-wave superconductors, which have gap nodes, quasiparticle decay processes which turn on at some threshold energy (e.g.,  $2\Delta$ ) display a dip in the density of states (DOS) near that energy [9], unshifted by  $\Delta$ .

There have been numerous SIN tunneling studies of Bi2212 including STM [10] and PCT [1,3] which displayed a spectral feature (dip or shoulder) at a voltage approximately twice that of the conductance peak ( $eV \sim$  $2\Delta$ ). An enhanced, symmetric dip feature in the conductance is found at approximately  $3\Delta$  in superconductorinsulator-superconductor (SIS) junctions [11], which can be probed with our PCT techniques [3]. This is consistent with a dip at  $\sim 2\Delta$ , in a *d*-wave DOS [9] because features are shifted by an additional factor of  $\Delta$  in the SIS configuration. The enhanced size of the dip in SIS junctions as well as the symmetry are consequences of the convolution of the quasiparticle density of states with itself [3,9]. In a recent STM study [2] of Bi2212 it was argued that the dip feature was absent for voltage polarities corresponding to electron injection. Under such an assumption, the dip and a higher energy hump could be explained by ordinary band structure effects in the background (normal state) tunneling conductance. Our results indicate that the dip is much more significant in that it is revealing information about the underlying pairing interaction. The interpretation of the dip in ARPES spectra [4,5] has been an ongoing debate [7,8,12,13], but recently a case has been made that this feature arises from the coupling of quasiparticles to collective excitations of some type centered near the  $(\pi, \pi)$  point of the Brillouin zone [7,8,12]

Given the inherent complexity of HTS cuprates, their surfaces, and the tunneling process in general,

experimental reproducibility becomes the key in determining the magnitude of the energy gap and in focusing on particular spectral features. We note, for example, that phonon structures have been reported in the tunneling conductance of some Bi2212 junctions [14], but these are much less common than the dip discussed here. In order to establish reproducibility in our experiment, we have analyzed a large number of samples and junctions and have utilized two distinct tunneling instruments, STM and PCT. This study is also unique in that Bi2212 crystals of unusually high quality have been measured.

The single crystals were grown using a floating zone process in a double-mirror IR image furnace NEC SC-M15HD modified with an external home-built control for very slow growth  $\sim 0.1 \text{ mm/h}$ . Growth was carried out without solvent from high-density, carbonate-free, near-stochiometric single-phase rods in flowing 20% oxygen [15]. Absence of impurities, stoichiometry, and high cationic homogeneity yielded  $T_c = 95$  K at optimal doping, the highest yet obtained in the pure Bi2212 with a transition width of 1 K. An example of the dynamic conductance measurements, dI/dV vs V, at 4.2 K on cleaved crystals using a PCT method with a Au tip are shown in Fig. 1. Details of the PCT techniques have been described elsewhere [1]. Here the bias voltage is that of the sample with respect to the tip so that negative (positive) values correspond to quasiparticle extraction (injection). The data display the high-bias decreasing background conductance often seen in Bi2212 tunneling [1,2,16] but by extending the measurement out to higher bias voltages we find that this anomalous feature ends at about  $\pm 400$  mV, whereupon the conductance flattens out and begins to increase slowly. Sharp conductance peaks are found at  $V_p = \pm 40 \text{ mV}$  and a strong dip feature is seen at -80 mV. The origin of the decreasing background is



FIG. 1. A representative SIN normalized PCT conductance (solid line) for optimally doped Bi2212, at 4.2 K and smeared BCS fit (dashed line) with parameter,  $\Delta = 37 \text{ meV}$ ,  $\Gamma = 4 \text{ meV}$ . The uppermost curve shows the reduced conductance which is the normalized conductance divided by the smeared BCS density of states.

not known; however, a possible source is the normal state electronic density of states which, according to ARPES measurement [17], is peaked near the Fermi energy, but no evidence of a sharp van Hove singularity is observed.

Given the uncertainty about the origin of the decreasing background conductance we normalize the data by a constant and compare it to a smeared BCS density of states,  $N_{\text{BCS}}(E) = \text{Re}E - i\Gamma/[(E - i\Gamma)^2 - \Delta^2]^{1/2}$ . This simple analysis led to a reasonable fit of the gap region of the tunneling conductance and gave a gap value of  $\Delta =$  $37 \pm 1 \text{ meV}$  that was reproduced in more than 20 junctions. This indicates that these optimally doped crystals are quite homogeneous. The strong-coupling parameter in this case is  $2\Delta/kT_c = 9.3$ . Also shown in Fig. 1 is the reduced conductance, which is the normalized conductance divided by  $N_{BCS}(E)$ . The reduced conductance is often used to indicate phonon structures in conventional strong-coupled superconductors [6]. The reduced conductance shows that at V = +80 mV there is a distinct feature which mimics the stronger dip found at -80 mV. Furthermore, the dip at negative bias appears to be part of a larger spectral feature that includes a pronounced hump at V = -150 mV. At V = +150 mV there also appears to be a hump, although again weaker than seen at negative voltages. Although the dip-hump feature is asymmetric in the SIN junctions obtained by PCT they clearly indicate that it is observed for both junction polarities. This result is found also with more sophisticated analyses of these and other PCT data (ongoing) using models which incorporate the band structure as well as a *d*-wave gap.

The STM measurements were performed with a home-built low temperature STM operating in a helium exchange gas at 4.2 K. To obtain clean atomically flat surfaces the samples, mounted in front of a Pt-Ir tip, were cleaved under helium atmosphere just before cooling down the STM. We investigated a  $T_c = 92$  K sample (slightly overdoped) and a 72 K sample (overdoped). Vacuum tunneling conditions were verified by checking the reproducibility of scanned images and of the spectra recorded at various tunneling resistances ranging between 1 to 5 G  $\Omega$ . Figure 2 shows a series of typical dI/dV vs V spectra (raw data) recorded at various locations on the 92 K sample. All the spectra show consistently the same features: a very low zero bias conductance of the order of 7%, sharp conductance peaks at  $V_p \sim \pm 35$  mV, and a dip and a hump structure.

Similar to that found in PCT the dip occurs at a voltage  $V \approx 2V_p$  and the hump is found in a range,  $V = \pm (90-110)$  mV. These features are superimposed on an asymmetrical, weakly decreasing background which is consistent with other STM measurements on Bi2212 [2,18]. The important novelty in the spectra of Fig. 2 is that the dip-hump structure is clearly observed at positive bias in the best resolved spectra. The STM background conductance is relatively flatter than found with PCT, and therefore we normalize the spectra by



FIG. 2. Experimental dI/dV vs V spectra for SIN junctions recorded with an STM (Pt-Ir tip) at 4.2 K at various locations on a 92 K Bi2212 sample. For clarity, the upper spectra have arbitrarily been shifted with respect to the lower spectrum. The vacuum tunnel resistance  $R = 1.6 \text{ G}\Omega$ . The dashed line in the bottom curve represents the polynomial fit used in the normalization procedure.

a smoothly varying polynomial fit. Care is taken to make sure the conservation of states rule is verified. An example of this procedure is presented for one spectrum in Fig. 2 (dashed line). The result of the normalization is shown in Fig. 3 for both the 92 and the 72 K sample. The normalized conductances are very consistent from junction to junction and the dip feature is clearly seen for both junction polarities. Note that in the 72 K sample the data exhibit qualitatively the same features, although



FIG. 3. Normalized STM dI/dV spectra corresponding to SIN junctions recorded at 4.2 K on a 72 and a 92 K Bi2212 sample. Four spectra are superimposed for each sample. An offset of 1.5 has been added to the curves of the 72 K sample.

the dip-hump is at lower energies and is, on average, less pronounced than in the 92 K sample. To estimate the gap value we fit the data with a broadened BCS density of states as before which leads to a value  $\Delta \approx 28-32$  meV and  $\Delta \approx 20-24$  meV for the 92 and the 72 K samples, respectively. This corresponds to a strong-coupling ratio  $2\Delta/kT_c \approx 7.1-8.0$  and 6.4–7.5, respectively. Fits to a *d*-wave DOS lead to identical gap values.

The PCT method has produced an unexpected but valuable result, namely, reproducible SIS junctions between two pieces of the Bi2212 crystal. This occurs after the Au tip is pushed with sufficient force to perforate any surface barrier layer and which leads to a low resistance contact (~1  $\Omega$ ), far less than the typical junction resistances of  $\sim 10 \text{ k}\Omega$ . With an additional increase and subsequent relief of the tip pressure, SIS junctions of high quality are observed. Evidence that these are SIS junctions include peaks at  $|eV| = 2\Delta$ , very symmetric conductance data, and well-defined, hysteretic Josephson currents. Examples of three such SIS junctions on Bi2212 crystals of various doping and  $T_c$  are shown in Fig. 4. All of the conductance data have been normalized by a constant value given by the conductance at V = -200 mV. Note that in the case of the optimally doped sample A, the conductance peak is located at 76 mV, almost exactly twice the  $\Delta$  value found in the SIN junction of Fig. 1. In the inset of Fig. 4 is shown a typical current-voltage (I-V)measurement near zero bias for an overdoped crystal with a  $T_c = 62$  K. The vertical current rise at zero bias is clearly seen in the data and the hysteretic behavior points



FIG. 4. Comparison of normalized SIS tunnel junctions at 4.2 K on Bi2212 for different oxygen concentration. *A*, *B*, and *C* represent optimally doped Bi2212 ( $T_c = 94$  K), moderately overdoped one ( $T_c = 82$  K), and highly overdoped one ( $T_c = 62$  K), respectively. Each curve has been normalized by its corresponding value at -200 mV, and the zero bias conductance peak from each data set which comes from Josephson current has been removed. The inset shows typical hysteretic current-voltage characteristics near zero bias for an overdoped Bi2212 with a  $T_c = 62$  K. Using peak voltage position as a superconducting gap value,  $2\Delta_p(A) = 76$  meV,  $2\Delta_p(B) = 51$  meV, and  $2\Delta_p(C) = 39$  meV.

to its origin as a Josephson effect. More details of these SIS junctions will be published elsewhere.

The SIS junctions offer a unique perspective on the dip feature. First, the conductance peak is very close to  $2\Delta$ which calibrates the energy scale of the dip. Second, the enhancement of the dip feature in the SIS configuration allows a better measure of its magnitude, especially in heavily overdoped crystals. In sample A, which is optimally doped, the dip is very pronounced and its center is at  $3\Delta$ . This is in agreement with previous SIS junctions [3,19] on the Bi2212 system. As the  $T_c$  is reduced by overdoping, the conductance peak moves to lower voltages and the dip location continues to scale with  $\Delta$ , down to the smallest gaps observed ( $\Delta = 15 \text{ meV}$ ) for  $T_c = 62 \text{ K}$ . Furthermore, there is a clear reduction in the magnitude of the dip feature in overdoped samples, an effect which was observed in the STM data of Fig. 3, but with less clarity. This decrease in magnitude correlates with the decrease of the strong-coupling ratio from 9.3 to (5.6–6.7) for  $T_c = 94.5$ and 62 K, respectively, and gives further support to the identification of the dip feature as a strong-coupling effect.

By strong-coupling we mean simply that a quasiparticle decay process turns on at a characteristic energy, producing a feature at  $\sim 2\Delta$  in the *d*-wave DOS [9]. The size of the dip (>10% deviation in the normalized conductance of Fig. 3) suggests that it might arise from the frequency dependence of the underlying pairing interaction. Feedback effects in electron-electron theories have been shown to produce such  $2\Delta$  peaks in the pairing function [the analog of  $\alpha^2 F(\omega)$  for phonons] near the  $(\pi, \pi)$  point of the Brillouin zone [20]. Related to these effects, it has recently been suggested that long-lived, collective excitations exist at  $(\pi, \pi)$  which couple to the quasiparticles in an analogous fashion to electron-phonon coupling in conventional superconductors [7,8]. In such scenarios it appears that the tunneling conductance is providing potentially detailed information on the microscopic pairing interaction.

This work was partially supported by the U.S. Department of Energy, Division of Basic Energy Sciences-Material Sciences under Contract No. W-31-109-ENG-38 (K.G., D.H.) and the National Science Foundation, Office of Science and Technology Centers under Contract No. DMR 91-20000 (Y. D. W., N. M., P. G., M. I., J. Z.) and the INFM (M. I.). L. O. acknowledges support from Izmir Institute of Technology, Turkey.

 Q. Huang, J.F. Zasadzinski, K.E. Gray, J.Z. Liu, and H. Claus, Phys. Rev. B 40, 9366 (1989).

- [2] Ch. Renner and O. Fischer, Phys. Rev. B 51, 9208 (1995).
- [3] J.F. Zasadzinski, N. Tralshawala, P. Romano, Q. Huang, J. Chen, and K.E. Gray, J. Phys. Chem. Solids 53, 1635 (1992).
- [4] H. Ding, A.F. Bellman, J.C. Campuzano, M. Randeria, M.R. Norman, T. Yokoya, T. Takahashi, H. Katayama-Yoshida, T. Mochiku, K. Kadowaki, G. Jennings, and G.P. Brivio, Phys. Rev. Lett. **76**, 1533 (1996).
- [5] D. S. Dessau, B. O. Wells, Z.-X. Shen, W. E. Spicer, A. J. Arko, R. S. List, D. B. Mitzi, and A. Kapitulnik, Phys. Rev. Lett. **66**, 2160 (1991); D. S. Dessau, Z.-X. Shen, B. O. Wells, D. M. King, W. E. Spicer, A. J. Arko, L. W. Lombardo, D. B. Mitzi, and A. Kapitulnik, Phys. Rev. B **45**, 5095 (1992).
- [6] E. L. Wolf, Principles of Electron Tunneling Spectroscopy (Oxford University Press, New York, 1985).
- [7] Z.-X. Shen and J. R. Schrieffer, Phys. Rev. Lett. 78, 1771 (1997).
- [8] M. R. Norman, H. Ding, J. C. Campuzano, T. Takeuchi, M. Randeria, Y. Yokoya, T. Takahashi, T. Mochiku, and K. Kadowaki (to be published).
- [9] D. Coffey and L. Coffey, Phys. Rev. Lett. 70, 1529 (1993).
- [10] A. Chang, Zhao Y. Rong, Yu. M. Ivanchenko, Farun Lu, and E. L. Wolf, Phys. Rev. B 46, 5692 (1992).
- [11] D. Mandrus, J. Hartge, C. Kendziora, L. Mihaly, and L. Forro, Europhys. Lett. 22, 199 (1993); D. Mandrus, L. Forro, D. Koller, and L. Mihaly, Nature (London) 351, 460 (1991).
- [12] L. Coffey et al., Phys. Rev. B 56, 5590 (1997).
- [13] G.B. Arnold, F.M. Mueller, and J.C. Swihart, Phys. Rev. Lett. 67, 2569 (1991).
- [14] N. Miyakawa *et al.*, in Proceedings of the 5th International Conference on Materials and Mechanisms of Superconductivity [Physica C (Amsterdam) (to be published)].
- [15] P. Guptasarma and D. G. Hinks, Bull. Am. Phys. Soc. 42, 713 (1997); 62 K overdoped crystals are from a process described by C. Kendziora *et al.*, Physica (Amsterdam) 257C, 74 (1996).
- [16] T. Hasegawa, H. Ikuta, and K. Kitazawa, in *Physical Properties of High Temperature Superconductors III*, edited by D. M. Ginsberg (World Scientific Pub., Singapore, 1993).
- [17] M.R. Norman, M. Randeria, H. Ding, and J.C. Campuzano, Phys. Rev. B 52, 615 (1995).
- [18] Ch. Renner, B. Revaz, J.-Y. Genoud, and O. Fischer, J. Low Temp. Phys. **105**, 1083 (1996).
- [19] J. F. Zasadzinski, L. Ozyuzer, Z. Yusof, J. Chen, K. E. Gray, M. Mogilevsky, D. G. Hinks, J. L. Cobb, and J. T. Markert, in *Spectroscopic Studies of High T<sub>c</sub> Cuprates*, edited by I. Bozovic and D. van der Marel, SPIE Proceedings Vol. 2696 (SPIE, Bellingham, 1996), p. 338.
- [20] P. Monthoux and D.J. Scalapino, Phys. Rev. Lett. 72, 1874 (1994).