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Electron tunneling study of the normal and superconducting states of $Bi_{1.7}Pb_{0.3}Sr_2CaCu_2O_x$

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We report point-contact tunneling into single crystals of $Bi_{1.7}Pb_{0.3}Sr_2CaCu_2O_x$. The current-voltage characteristics $I(V)$ are quite symmetrical and very reproducible, exhibit a decreasing conductance at high voltages, and can be fitted with a smeared BCS density of states (DOS). We are necessarily led to conclude that the normal-state conductance, in the absence of parabolic-tunneling-barrier effects, has a peak near the Fermi energy, and we *speculate* that it could be due to the normal-state DOS of $Bi_{1.7}Pb_{0.3}Sr_2CaCu_2O_x$.

Tunneling measurements[1] are an important probe of high-temperature superconductors (HTS), providing a direct measure of the superconducting energy-gap parameter, Δ. While surface problems plagued the early experiments on HTS, particularly in $YBa_2Cu_3O_7$, recently tunneling studies[2-9] in $Bi_2Sr_2CaCu_2O_x$ have resulted in more consistent estimates of Δ. We report point-contact tunneling from a Au tip onto freshly cleaved surfaces of a single crystal of $Bi_{1.7}Pb_{0.3}Sr_2CaCu_2O_x$ which has $T_c = 96$ K. Both the current-voltage characteristic $I(V)$ and dI/dV are quite symmetrical, displaying a well-defined BCS-like gap; however, the shape of the high-voltage conductance varies with junction resistance R_n, albeit in a systematic and reproducible way. For junctions with large R_n, the high-voltage conductance increases with increasing voltage, as is commonly found in HTS. However, for $R_n < 10$ kΩ the high-voltage conductance *decreases* with increasing voltage. We argue that this behavior is not due to short circuits or normal regions, but is consistent with the normal-state conductance being a convolution of the usual parabolic-tunneling conductance[10] and *a strongly energy-dependent normal-state density of states $N_n(E)$ which peaks near the Fermi energy.*

Our tunneling apparatus[11] has recently been used[12] on Nb to demonstrate that point-contact tunneling can accurately and quantitatively reproduce the phonon structure found in thin-film junctions.[10] Single crystals of Bi-Pb-Sr-Ca-Cu-O were grown by the flux method[13] and displayed sharp superconducting transitions starting at 96 K for both the Meissner and shielding effects, the former being more than half the latter at 60 K. These were cleaved prior to mounting and the cool down to 77 K took about 3 h. All low-temperature measurements were done with the apparatus cooled by exchange gas to liquid ^{4}He (2-5 K) or N_2 (77 K). After filling the appropriate cryogen, it was necessary to wait about 5 h for thermal stability before reproducible data could be taken. Raising the temperature above the cryogen boiling point with a heater was possible, but resulted in much poorer stability of the tunneling contact. As in previous cases,[11,12] an insulating surface prevented vacuum tunneling and the Au tip was used to mechanically scrape the surface before a measurable current was obtained. The resistance of resulting junctions could be varied by adjusting the transducer voltage and hence the force between the tip and sample. The data reported here is for three low-temperature runs, on two crystals, in one case after replacing the Au tip.

The $I(V)$ was monitored while the tip was maneuvered to obtain an acceptable junction. First and second derivatives were obtained using a bridge with the usual lock-in techniques. For stable junctions with large R_n, the high-voltage conductance increases with increasing voltage, as is commonly found in HTS, but the gap structure is obscured by it. However, for $R_n < 10$ kΩ, the high-voltage conductance *decreases* with increasing voltage and the $I(V)$ were always quite symmetrical, showing a well-defined BCS-like reduced DOS with conductance peaks at $\pm V_p$. These data exhibited a weak but reproducible, characteristic asymmetry in dI/dV which *always* ap-

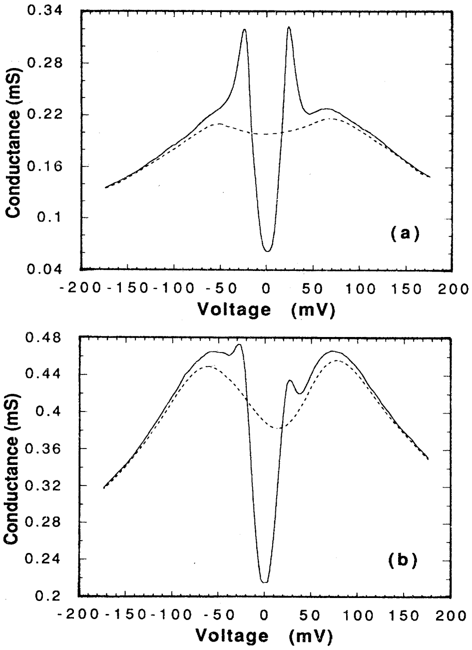

FIG. 1. Experimental conductances (solid lines) with the fitted normal-state conductance (dashed lines), each showing the characteristic small asymmetry with a dip above the peak at positive bias and a shoulder below the peak at negative bias. (a) No. 9 at $T = 4.2$ K. (b) No. 2 at $T = 77$ K.

peared as a dip (shoulder) in dI/dV just above $+V_p$ (below $-V_p$), i.e., for the Bi-Pb-Sr-Ca-Cu-O crystal at negative (positive) voltage compared to the Au tip. Examples of this at 4.2 and 77 K are shown in Fig. 1.

Although there is some scatter in V_p for these junctions (see Table I), the asymmetries in dI/dV indicate that they are likely the asymmetrical normal-metal–insulator–superconductor (NIS)-type junctions. A few contacts were extremely symmetrical, showed no dip or shoulder, indicated dc Josephson-type supercurrents, and exhibited peaks in dI/dV at considerably larger voltages (60 to 140 mV). All these effects are consistent with symmetrical superconductor-insulator-superconductor (SIS) junctions, either single or multiple, within the Bi-Pb-Sr-Ca-Cu-O crystal or between fractured crystal pieces. These were not used in further analysis, but the example shown in Fig. 2 also demonstrates the flatter conductance found for $R_n \sim 10$ kΩ. Considerable data were also collected at room temperature and a limited amount just above T_c by heating the sample above liquid N_2 temperature. These also showed structure and weak asymmetries which were consistent with the data below T_c. Virtually all contacts with $R_n < 10$ kΩ, whether symmetrical or not and whether above T_c or below, showed an unusual and unexpected *decrease in dI/dV at high voltages*, while in the normal state, a broad dip in dI/dV was found near zero bias.

We begin the analysis by noting that dI/dV for the asymmetrical NIS junctions at high voltages and/or $T > T_c$ should approximate the normal-state conductance. Although direct measurement of the normal-state conductance for each low-temperature junction is difficult due to thermal expansion of the tip assembly for temperatures near T_c, our results are consistent with the normal-state conductance being a convolution of the usual parabolic-tunneling conductance[10] and *a strongly energy-dependent normal-state density of states* $N_n(E)$ *which peaks near the Fermi energy.* Whenever the voltage becomes a considerable fraction of the tunnel barrier height, its effect on the $I(V)$ cannot be neglected, and one customarily finds an *increased* conductance at higher voltages which takes a parabolic form and is offset from zero bias for nonidentical electrode materials.[10] This parabola can explain the broad dip near zero bias in the normal state *and* the increasing high-voltage conductance for large R_n, but is *opposite* to the decreasing high-voltage conductance found experimentally for $R_n < 10$ kΩ. Therefore, the experimentally observed decrease in conductance must be due to something else. First, we point out that zero-bias anomalies[10] have a much smaller magnitude and occur at considerably lower voltages (~ 10 mV). We also rule out microscopic short circuits *in parallel* with the junction because the decrease in high-voltage conductance is greater than the zero-bias conductance for small R_n. Next, we consider localized normal regions *in series* with the junction which are caused by exceeding the critical current or temperature and expand in size with junction current. If the variation in R_n is due to changes in junction area, rather than barrier transmission, the current density would be constant so this effect would not depend on R_n. At the opposite extreme, we assume the junction areas are the same for each R_n. If, for junctions with $R_n \sim 5$ kΩ, we attribute the decrease in conductance at high voltage to such normal regions in series, then junctions with $R_n \sim 1$ kΩ should be similarly affected at lower voltages, i.e., in the gap region: However, there is no correlation of the gap structure or Δ with R_n (see Table I). Having exhausted other explanations, we propose that the decreased high-voltage conductance is due to $N_n(E)$, the normal-state DOS in Bi-Pb-Sr-Ca-Cu-O.

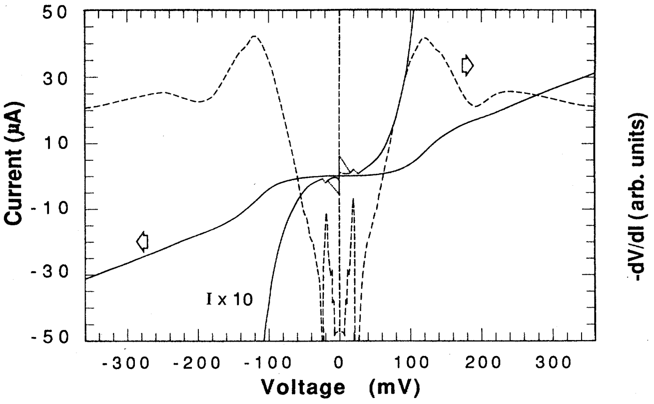

FIG. 2. Experimental current-voltage characteristics $I(V)$ and first-derivative spectra dI/dV (dashed line) for No. 3 taken at 4.2 K for a symmetrical contact exhibiting a dc Josephson-type supercurrent and conductance peaks at high voltages implying a multiple-SIS junction.

TABLE I. Parameters for fitting to a smeared BCS DOS: R_n, the normal-state junction resistance at zero bias; $Y(0)$, the conductance at zero bias in the superconducting state; V_p, the conductance peak voltage; Δ, the superconducting energy gap; and Γ, the smearing parameter. Data at 4.2 K except last one which is at 77 K.

R_n (kΩ)	$Y(0)R_n$	V_p (mV)	Δ (meV)	Γ (meV)
3.0	0.125	31.2	30	2
1.3	0.34	29.1	26.5	3
2.6	0.21	28.8	26.5	2.4
2.5	0.21	26.9	25.3	1.3
1.1	0.26	26.0	26.5	0.9
5.0	0.31	23.8	21.5	3.5
3.5	0.29	19.4	16	4.4
4.0	0.28	17.5	16.5	1.5
2.6 (77 K)	0.56	27.5	17.5	11

We have been able to fit all our measurements of dV/dI in the superconducting states at 2–5 and 77 K, together with the normal state at ~ 100 K and room temperature, to a *strongly energy-dependent* $N_n(E)$ *which peaks near the Fermi energy* and the usual parabolic-tunneling conductance, each of which varies slightly from junction to junction. To obtain these fits below T_c, we start with a high-order (7–9) polynomial fit of the data above 40 mV (dashed lines in Fig. 1) after a small superconducting correction based on the BCS theory is applied. A broad dip near zero bias is invariably found in these fits, and it is

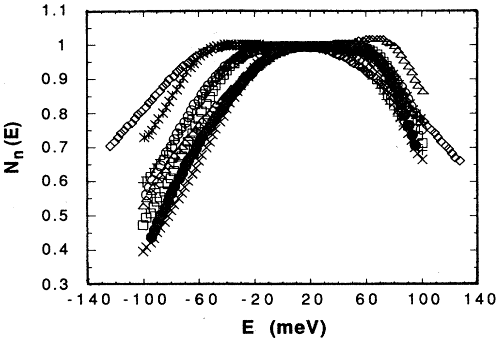

FIG. 3. Values of $N_n(E)$ obtained from fitting data taken at low temperatures: (open circles) No. 4; (open squares) No. 5; (open diamonds) No. 6; (crosses) No. 7; (plusses) No. 8; (open triangles) No. 9; (filled circles) No. 10; and (filled squares) No. 11.

used to define a tunneling parabola, $1+a(V-V_0)^2$, for each junction. After dividing by the tunneling parabola, a second fit, which forces the conductance peaks at $\pm V_p$ to have equal height, determines $N_n(E)$. These are summarized in Fig. 3 and show a sharply peaked (full width half maximum is 197–315 meV) $N_n(E)$ at the Fermi energy, $E=0$. Recent studies[14] of $Tl_2Ba_2CaCu_2O_x$ single crystals confirm the same systematic behavior of the background conductance on R_n.

Although it is not possible to unambiguously determine the correct tunneling parabola and $N_n(E)$ from our data, and the above decomposition is somewhat arbitrary, it emphasizes that the usual tunneling parabola cannot explain the decreased conductance at high voltages and explains the nonmonotonic variation of the normal-state conductance. For example, the flat portions of $N_n(E)$ shown in Fig. 3 are a direct result of our procedure and probably not real, and we could improve the consistency of $N_n(E)$ by choosing different tunneling parabolas [V_0 and the energy midway between the half-maxima of $N_n(E)$ have a very high correlation]. In addition, the propensity for the tunneling parabola to dominate over $N_n(E)$ and $N_s(E)$ as R_n increases explains the increasing high-voltage conductance and lack of gap structure in very-high-R_n junctions.

When the NIS data below T_c is divided by the normal-state conductance, we arrive at the *normalized superconducting DOS* $N_s(E)$, which can be compared to theory. We have used the smeared BCS DOS, first proposed by Dynes *et al.*[15] to account for lifetime effects, in which $N_s(E)=\mathrm{Re}\{(E-i\Gamma)/[(E-i\Gamma)^2-\Delta^2]^{1/2}\}$. Inelastic scattering may provide such lifetime effects. Such fits are shown in Fig. 4 and provide energy gaps Δ and smearing parameters Γ which are summarized in Table I. We have *not* accounted for the effect of thermal smearing in the 77-K data which, no doubt, contributes to the larger Γ. Note that *there are no correlations of any parameters with* R_n, which varies by a factor of 5. The dip and shoulder which remain in the reduced DOS in Fig. 4 may be features of the actual $N_n(E)$, but our procedure for obtaining $N_n(E)$ purposely avoided this voltage range where the superconducting effects are large.

Most of the recent tunneling studies[2–9] of HTS have concentrated on the magnitude of Δ to establish whether the materials are strong coupling. Our most important result is a set of $I(V)$ data, using Au point-contact tunneling into freshly cleaved single crystals of Bi-Pb-Sr-Ca-Cu-O, which is reproducible, exhibits a decreasing conductance at high voltages and which can be fit with a smeared BCS DOS. We are necessarily led to conclude that the normal-state conductance, in the absence of parabolic-tunneling barrier effects, has a peak near the Fermi energy, and we *speculate* that it could be due to the normal-

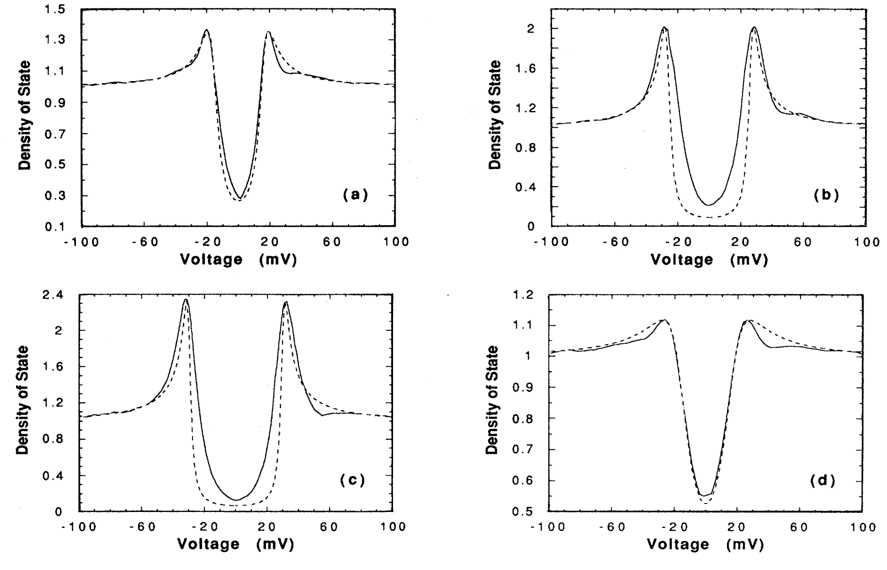

FIG. 4. Examples of fitting the reduced density of states to a smeared BCS DOS for several contacts. (a) No. 10 at 4.2 K, $\Delta=16$ meV, $\Gamma=4.4$ meV. (b) No. 6 at 4.2 K, $\Delta=26.5$ meV, $\Gamma=2.4$ meV. (c) No. 4 at 4.2 K, $\Delta=30$ meV, $\Gamma=2$ meV. (d) No. 2 at 77 K, $\Delta=17.5$ meV, $\Gamma=11$ meV.

state DOS of Bi-Pb-Sr-Ca-Cu-O. Structure in $N_n(E)$ is not usually observed in tunneling. Within the Wentzel-Kramers-Brillouin (WKB) approximation,[16] a cancellation occurs between the electron velocity and one-dimensional DOS. However, $N_n(E)$ may be seen in metals for which the Fermi wavelength is greater than the barrier width and the WKB approximation breaks down.[17] Examples are the low-carrier-density semimetals such as Bi and Sb. It is not unreasonable to expect that these conditions may apply in Bi-Pb-Sr-Ca-Cu-O also, because of its low carrier density. One previously reported tunneling study[2] of Bi-Pb-Sr-Ca-Cu-O found a decreasing conductance at high voltages, but this "single" crystal exhibited two low-T_c superconducting phases at 28 and 65 K. All other reports showed increasing[3-6] or flat[7-9] background conductances. Evidence for a $N_n(E)$ which peaks near the Fermi energy is also found in[14] Tl-Ba-Ca-Cu-O and in thin-film sandwichlike junctions[18] of $Ba_xK_{1-x}BiO_3$, another low-carrier-density superconductor.

For our asymmetrical junctions, Δ is between 16 and 30 meV at low temperatures, with most values between 25 and 27 meV. The origin of the spread in Δ is not known, but could result from anisotropy, local disorder and/or proximity effects, but is more likely different phases of Bi-Pb-Sr-Ca-Cu-O with different T_c. These values are not too different from Y-Ba-Cu-O implying similar values of $2\Delta/k_BT_c \sim 6.3$ (for $T_c = 96$ K and $\Delta = 26$ meV). Several reports of tunneling into Bi-Sr-Ca-Cu-O have employed this[3,7] and other[8] fitting schemes on measured conductance peaks and report values between 4.5 and 6.4. Additional reports[2,4,5] claim values between 6.7 and 8.7 *without* fitting. Finally, one report[9] shows a very large value of 12, but also a narrow conductance peak at zero bias, most likely indicating SIS behavior.

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