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Quasiparticle Tunneling Spectroscopy of High-T_c Cuprates*

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ABSTRACT

Superconductor-insulator-normal metal (SIN) and superconductor-insulator-superconductor (SIS) tunnel junctions provide important information on pairing state symmetry and mechanism. Measurements of such junctions on high T_c superconductors (HTS) are reported using mechanical point contacts, which generally display the optimum characteristics that can be obtained from HTS native-surface tunnel barriers. New tunneling data on the infinite-layer cuprate, $Sr_{1-x}Nd_xCuO_2$ are reported which show a remarkable similarity to another electron-doped cuprate, $Nd_{1.85}Ce_{0.85}CuO_4$. In particular, there is a strong, asymmetric linear background conductance that is indicative of inelastic tunneling from a continuum of states. A discussion is given of the anomalous "dip" feature found in the tunneling and photoemission data on BSCCO 2212. It is shown that a similar feature is found in many cuprate junctions and that this dip scales with the gap energy over a wide range. New data on the single-layer, tetragonal cuprate, $Tl_2Ba_2CuO_6$ (Tl2201) are presented and discussed in light of recent published results on the similar compound $HgBa_2CuO_4$ (Hg1201). The Hg1201 data display a low, flat sub-gap tunneling conductance which is consistent with a BCS density of states whereas the Tl2201 data display a cusp-like feature at zero bias which is more consistent with $d_{x^2-y^2}$ symmetry.

1 INTRODUCTION

Quasiparticle (single-electron) tunneling spectroscopy has long been viewed as a powerful probe of the superconducting state.[1] In conventional superconductors such as Pb and Nb, the gap in the density of states is seen in the tunneling conductance and the strong-coupling phonon structures are found as well. Inversion of the data allows the determination of the gap energy, the electron-phonon spectral function, $\alpha^2F(\omega)$, and the coulomb pseudopotential, μ^*, thereby providing a complete description of the superconductivity in conventional metals.[1] In the early studies of high Tc superconductors (HTS) the tunneling spectroscopy results were plagued by poor reproducibility, however, more recent studies on better characterized samples has led to a consensus on energy gap values for a number of HTS. Tunneling data on BSCCO 2212, for example, are highly reproducible and consistent results have been reported by a number of groups using various junction methods.[2] A good review of HTS tunneling is given by Hasegawa et al.[3]

We report here progress made in the development of SIN and SIS junctions on oxide superconductors using a mechanical, point-contact tunneling (PCT) approach. All of the PCT data presented are from the Argonne/IIT tunneling group. For our instrument, the term "point contact" is somewhat of a misnomer in that the contact area of the tip and sample can be large compared to atomic dimensions. This is by design as the intention is not to obtain atomic scale images as with an STM but rather to obtain stable contacts with relatively low resistances of typically 1 kΩ to 20 kΩ for improved signal-to-noise. Barrier height analysis of such junctions on Nb[4] using a Au tip indicated a contact diameter of ~2400 Å. Furthermore, the observation of the Nb phonon structures consistent with planar junctions[4] demonstrated that this method could be used for sensitive spectroscopy. This mechanical method has proven to be a reliable and versatile tool for making many quasiparticle junctions on a given sample. The tip can be used to scrape, clean and in some cases cleave the HTS surfaces at low temperatures, leaving a thin, native-barrier for elastic tunneling. The PCT method has generally provided the best quasiparticle junction characterisities of most HTS materials. For example, ideal, BCS quasiparticle characteristics were first discovered on $Ba_{1-x}K_xBiO_3$ (BKBO) using PCT.[5] Nine different HTS compounds have been examined by this technique and here we present a brief review of some earlier results along with new data on the infinite layer system $Sr_{1-x}Nd_xCuO_2$ and the single-layer, tetragonal compound, $Tl_2Ba_2CuO_6$.

2. TUNNELING DENSITY OF STATES

No discussion of tunneling data can proceed without a brief discussion of the density of states (dos) for cuprate superconductors. The tunnel current in an SIN or SIS junction can be written as,

$$I(V) = Ct^2\int_{-\infty}^{\infty}\rho_1(E)\rho_2(E+eV)[f(E)-f(E+eV)]dE\,. \qquad (1)$$

Here, $\rho_1(E)$ and $\rho_2(E)$ are the quasiparticle dos in the two electrodes and C is a constant which depends on (among other things) junction area. The Fermi functions, f(E) account for thermal population of quasiparticle states. Here, $\rho(E)=1$ for a normal metal which points out an often neglected fact that all band structure effects have mysteriously disappeared from the integral. The absence of the band structure dos in the tunneling data of conventional superconductors is experimentally established and a theoretical argument for this has been given by Harrison (see chap. 2 of ref 1). The tunneling matrix element, t^2, has an energy dependence which is assumed to be weak over the voltage range of interest (energy gap region) and has been taken out of the integral. In the limit, T=0 K, the tunneling conductance, σ_S =dI/dV, for an SIN junction becomes,

$$\sigma_S = Ct^2\rho(E) \qquad (2)$$

where we have now set E=eV. Eq. 2 simply states that a measurement of the tunneling conductance at low temperatures should reveal the quasiparticle dos, which for a BCS superconductor is given by $\rho(E)= E/(E^2-\Delta^2)^{1/2}$. An exact determination of $\rho(E)$ requires a measurement of the weak, voltage-dependent background (or normal state) conductances to divide out the prefactors of eq. 2, however such a measurement can be difficult due to the high critical fields and temperatures of HTS. Also, in some cases the background conductances are far from having a weak voltage dependence as we will show. In the absence of a normal state measurement, it is common to normalize the data by a constant taken at some arbitrary high bias voltage.

To examine the dos of HTS cuprates, we take a simple model identical to that described by Fedro and Koelling.[6] The two-dimensional Cu-O_2 planes are represented by a single, tight-binding band with nearest neighbor (NN) and second NN hopping described by t and t'. We choose t'=0 for simplicity and a hole concentration of 0.18 which shifts the van Hove singularity above the Fermi energy at E=0. Fig. 1 shows what the superconducting energy gap looks like in the dos.

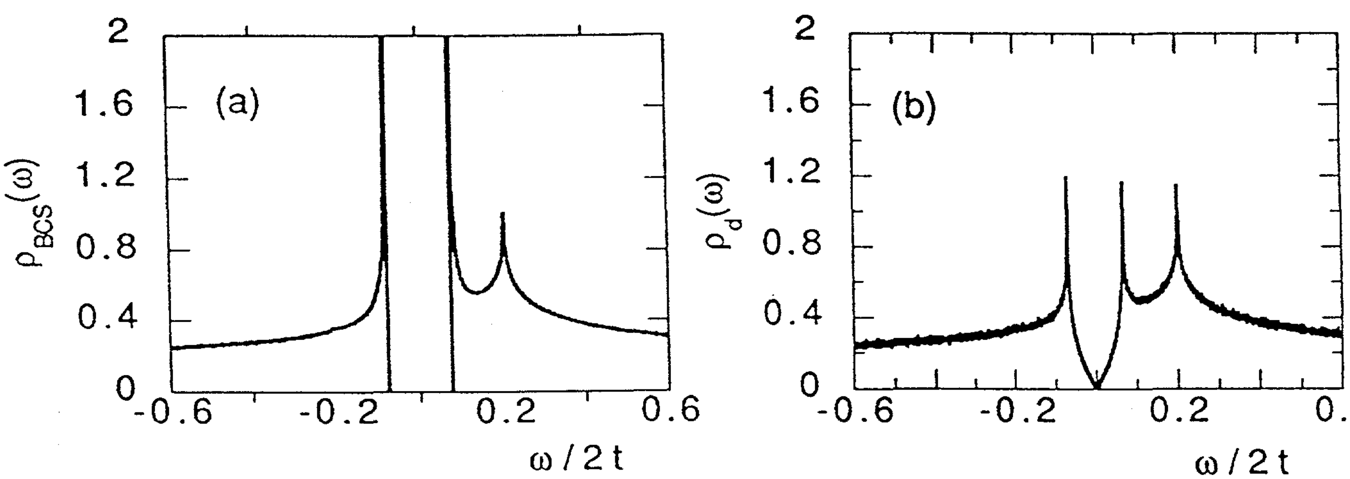

Figure 1 Two-dimensional, tight-binding band dos with (a) isotropic gap and (b) d-wave gap.

The energy scale ($E=\hbar\omega$) is normalized by 2t and the full band width is 8t which is approximately 1 eV for HTS cuprates. An isotropic, BCS gap parameter with Δ_0~20 meV would look Fig. 1(a). There is increasing evidence the gap parameter of certain HTS cuprates is of $d_{x^2-y^2}$ symmetry[7] and the dos in this case is shown in Fig. 1(b). In our simple model the d-wave gap parameter is characterized by $\Delta(k) = \Delta_0/2[\cos(k_x a)-\cos(k_y a)]$ where Δ_0 is a constant and is equal to the BCS gap parameter of Fig. 1(a). The d-wave gap parameter of Pines et al[8] which results from a fully self-consistent, Eliashberg-type treatment of spin fluctuation mediated pairing produces a dos similar to that of Fig. 1(b), however, the peak heights are somewhat smaller. The most obvious difference in the two curves of Fig. 1 is the cusp in the dos at the Fermi energy for the d-wave gap. As will be shown, the van Hove singularity, which is a striking feature of the dos is never seen in the tunneling data.

3. EXPERIMENTAL RESULTS

3.1 Electron-doped compounds

PCT data on $Nd_{1.85}Ce_{0.85}CuO_4$ (NCCO) with a $T_c = 22K$ were reported[5] in 1990. These were SIN junctions using a Au tip. The relatively low T_c allowed a direct measurement of the normal state conductance. These data showed well-defined gap features with zero-bias conductances that were ~15% of the normal state values. The gap region data could be fit reasonably well with eq. 1 and a BCS dos leading to a gap parameter Δ=3.6 $\pm$ 0.1 meV. The identical gap value was found on more than twenty junctions on three separate samples. This degree of reproducibility in the gap measurement is not typical of cuprate junctions but appears to be peculiar to NCCO. Other measurements of the NCCO gap from penetration depth measurements and Raman scattering give the same value[9] and taken together, the data overwhelmingly support a nearly isotropic, s-wave gap. Under these circumstances, the small but non-zero sub-gap conductances found in NCCO junctions would have to come from extrinsic sources, i.e., not due to the bulk dos. The sub-gap data do not display the cusp feature which is the signature of the d-wave state. One possibility is a slightly degraded surface which produces a proximity effect. Even $Ba_{1-x}K_xBiO_3$, which displays ideal BCS characteristics in PCT junctions[2,5] can display broadened gap features using, for example, thin film junction contacts.[2] Detailed tunneling spectroscopy of the phonon region in NCCO has been reported[2,5] and the resulting $\alpha^2F(\omega)$ has been shown to be in good agreement with the $F(\omega)$ from neutron scattering.[2,10] This suggests that not only is NCCO an isotropic, s-wave superconductor, but that the pairing mechanism is predominantly phonon mediated with a coupling constant λ ~1. Clearly, NCCO has a number of features which make it different from the higher T_c, hole-doped cuprates (see ref. 9 and references therein).

Another electron-doped system which is of great interest is the so called infinite layer class of HTS cuprates of which $Sr_{1-x}Nd_xCuO_2$ (SNCO) is an example. We performed PCT measurements on polycrystalline samples of SNCO[11] with T_c~ 35 K. A typical I-V and dI/dV vs. V characteristic is shown in Fig. 2 for an SIN junction on SNCO using a Au tip.

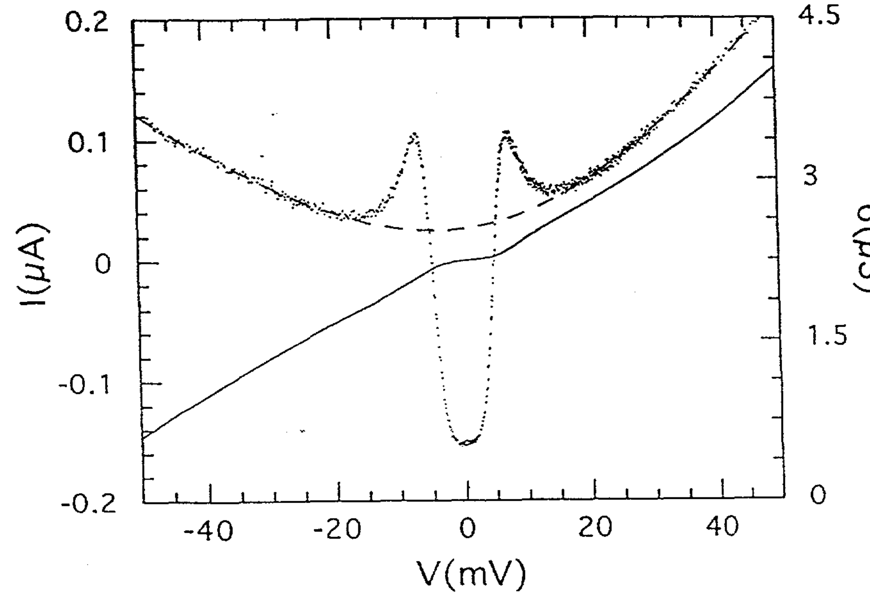

Figure 2. I(V) and dI/dV (dots) for SIN junction on SNCO. Dashed line is fitted normal state conductance.

The dI/dV curve displays a well-resolved gap feature, with nearly perfectly symmetric conductance peaks, but a non-zero value of the superconducting conductance at zero bias, $\sigma_S(0)$. Nevertheless, the conductance near zero bias is flat and does not show the cusp feature of the d-wave state. For these junctions, we estimated the normal state conductance by fitting a smooth curve through the high bias data (ignoring data for -20 mV<V<20mV) which should approach the normal state data. The normal state curve obtained in this way is shown as the dashed line in Fig. 2. The resulting normalized conductance curve was fit to a modified BCS density of states,

$$N(E) = \mathrm{Re}\, E-i\Gamma/[(E-i\Gamma)^2 - \Delta^2]^{1/2} \qquad (3)$$

first introduced by Dynes[12] et al to account for quasiparticle lifetime effects in superconductors. It should be noted that in our case the use of the expression in eq. 3 is one of convenience and the value of Γ may not have any physical significance for SNCO. The fit using eq. 3 leads to the parameters, Δ=6.0 meV and Γ=1.4 meV. Using the mid-point of the magnetic transition for T_c (35 K) we obtain $2\Delta/kTc=4.0$ which is the same as found on $Nd_{1.85}Ce_{0.15}CuO_4$ (NCCO) using the point contact method and indicates moderate coupling strength. Nearly identical gap values were found on 10 different junctions made on two different samples. It should be mentioned here that while our normalization procedure may be inaccurate, the magnitude of the gap was insensitive to the type of normal state curve used. For example, using a constant to normalize the data led to a poorer quality fit to eq. 3 but nevertheless the same gap value.

The tunneling data for the junction of Fig. 2 is shown out to a higher voltage range in Fig. 3(a) along with three other junctions made near the same spot on one SNCO sample. The four junctions differ in resistance by roughly an order of magnitude, from ~200 kΩ to ~2 MΩ at 50 mV bias and all show a sharp linear rise in conductance with bias voltage as has been observed in several cuprate superconductors.[13] Each junction has the identical gap value. The general behavior of the superconducting tunneling conductances of the SNCO is surprisingly similar to that of other n-doped cuprates. In Fig. 3(b) is shown the tunneling conductances out to high bias of a representative junction on $Nd_{1.85}Ce_{0.15}CuO_4$. The strong, linearly increasing backround with the weak asymmetry is the same as found on the infinite layer compounds. We have observed identical behavior in the $Pr_{1.85}Th_{0.15}CuO_4$ compound.[11]

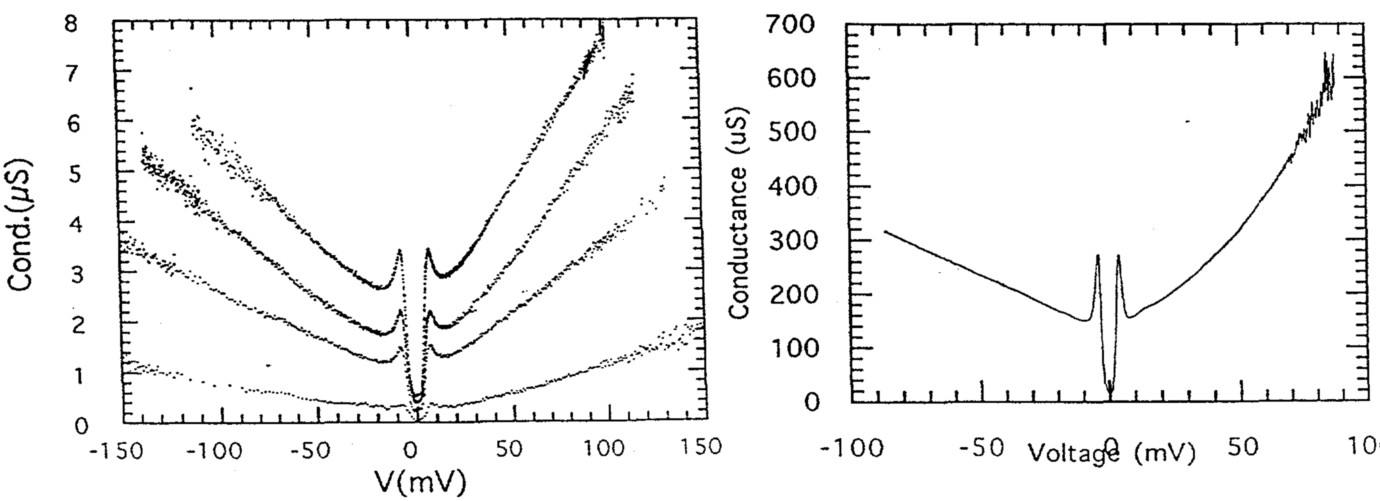

Figure 3. (a) superconducting dI/dV for four junctions on SNCO displaying the strong linear increase in background conductance, (b) superconducting dI/dV for NCCO. Voltage is that of the tip with respect to sample.

The general behavior of the three n-type cuprates we have studied by PCT[5,10,11] is as follows: for each compound highly reproducible gap values are found although the normalized conductance is generally broader than expected for a BCS dos and a strong, linearly increasing background conductance is found with an identical asymmetry. The reproducible gap values in NCCO are consistent with the apparent isotropic, s-wave symmetry[9] and this suggests that the infinite layer compound might have similar gap symmetry. The linear conductance background can be explained with an inelastic tunneling process from a broad continuum of states as suggested by Kirtley et al[13] but why are these background shapes the only ones observed? In PCT studies of hole-doped cuprates, a variety of background shapes are found including decreasing with bias[3,14] as will be shown in the next section.

3.1 The anomalous "dip" feature

PCT data on Pb-doped $Bi_2Sr_2CaCu_2O_x$ (BSCCO 2212) with a Tc=96 K were reported[14] in 1989. Two SIN tunneling conductance curves (Au tip) from that article are reproduced in Figs. 4(a) and 4(b) for the temperatures, 4.2 K and 77 K respectively. The background conductances decrease with applied voltage, an anomalous feature in itself but one which is typical for BSCCO 2212. In Fig. 4(a) the positive bias (Au tip is + with respect to the sample) is characterized by a sharp conductance peak at ~22 mV, a pronounced dip near 45 mV followed by another peak near 70 mV. For the negative bias direction, the dip feature is weaker (but nevertheless observable) and we originally described it as a shoulder. The observation of a dip feature near 2Δ in the SIN junctions on BSCCO and at 3Δ in SIS junctions has been found by many

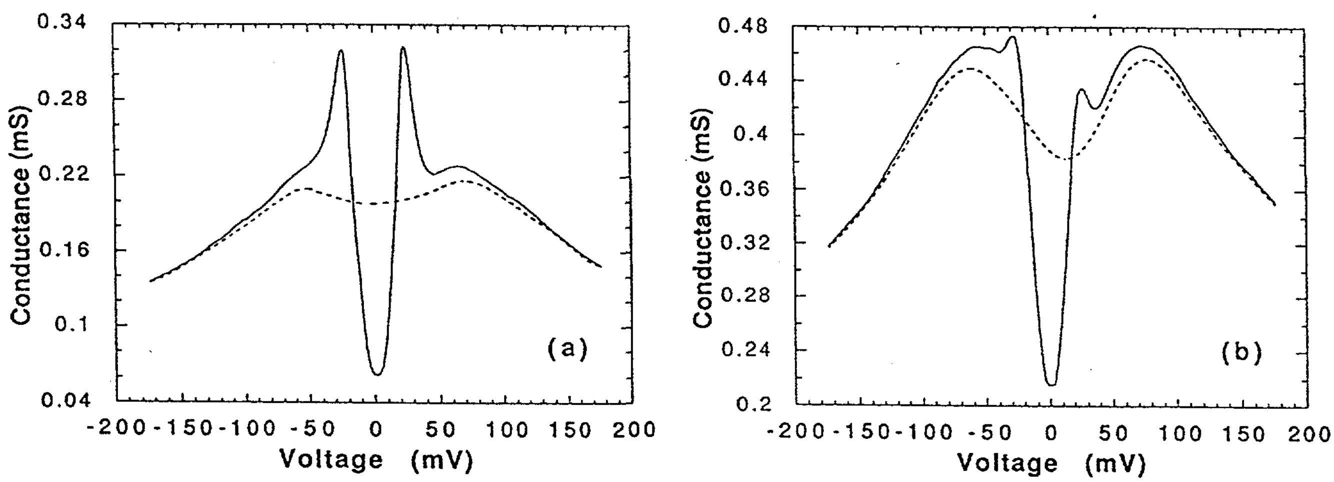

Figure 4. Superconducting dI/dV (solid lines) on Pb-doped BSCCO 2212 for (a) T = 4.2K and(b) T = 77K. Dashed lines are estimates of the normal state background as described in the text. Voltage is that of tip with respect to sample.

groups using various junction methods.[2] Note that features are shifted by an additonal factor of Δ in SIS junctions compared to SIN. The dip is clearly a reproducible feature and appears to be connected to a similar dip feature found in the spectral weight function measured in photoemission experiments.[15] To gain some insight into this feature we have constructed estimates of the normal state curves by fitting the data for $|V|>40$ mV to a high order polynomial. These curves are shown as dashed lines in Fig. 4. Considering the different conductance values for the two junctions and noting the different sensitivities in the ordinates, the inferred background shapes are quite similar. The reduced gap features of the 77 K junction put a greater focus on the peak near 70 mV and perhaps it is this feature to which attention should be paid. Similar dip features are found in other cuprate junctions including the electron-doped NCCO[2], although in that case it is a subtle feature exposed only because of the ability to measure the normal state conductance. SIN junctions on $Tl_2Ba_2CaCu_2O_x$ (TBCCO)[16] generally displayed pronounced dips for both bias values, again at a voltage nearly twice that of the conductance peak.

The dip features are symmetric and much more pronounced in SIS junctions[2] and for this reason we have chosen to plot the SIS characteristics of various cuprate junctions on a single plot and this is shown in Fig. 5. In the cases of NCCO and TBCCO we generated the SIS curves from SIN data using eq. 1 and the superconducting dos for each electrode. The T_c values of the cuprates range from 5.5 K for BSCCO 2201 to 100 K for TBCCO and to plot the data on a single graph we have normalized the voltage axis by $V_p/2$ where V_p is the voltage of the conductance peak. Using this normalization, the x-axis is in units of Δ. It is clear from Fig. 5 that the dip and subsequent peak features scale with the superconducting gap, which varies by a factor of 30 over the cuprates examined.

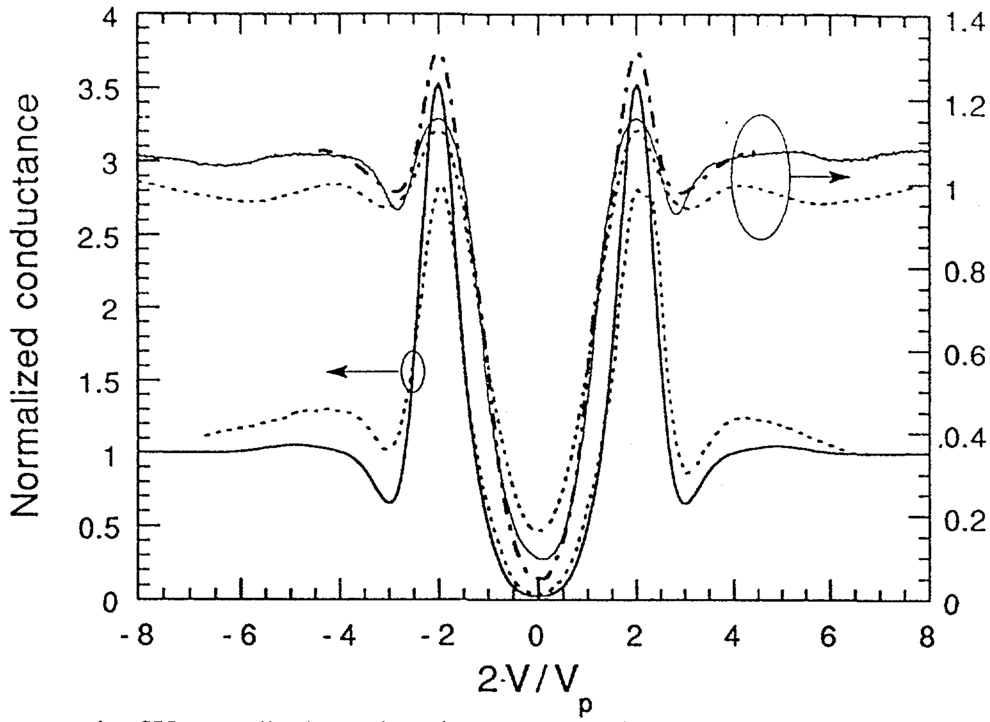

Figure 5. Representative SIS normalized tunnel conductances for various cuprate superconductors (T_c from 5.5 K to 100 K) plotted on a voltage axis renormalized in units which are ~ Δ. Left scale: NCCO (solid line), BSCCO 2212 (dashed line). Right scale: TBCCO (dashed-dot line), BSCCO 2201 (solid line), BSCCO film (dashed line).

Note that in the case of NCCO the dip is about 3.5 meV from the conductance peak, far below any phonon peak energies (typically 10 meV-70meV) and therefore does not interfere[2] with the determination of $\alpha^2F(\omega)$. Linking the dip feature to a superconducting energy scale is important to understanding its origin. D. Coffey has argued[17] that a natural explanation is an intrinsic, energy dependent quasiparticle decay mechanism, $\Gamma(\omega)$, put into the dos of eq.3, which turns on at a characteristic energy, 2Δ or 3Δ, for d-wave and s-wave respectively. He further argues that that the location of the dip at 3Δ in SIS in Fig. 5 (consequently 2Δ in the dos) is evidence for d-wave superconductivity. This is an attractive explanation however, the dip is found at the same location in NCCO which we have shown is most likely an s-wave superconductor. Perhaps the focus at the present time should not be on the precise location of the experimental dip as it might be affected by tunneling background shapes for example. An alternative explanation put forth recently by L. Coffey and K. Kouznetsov (CK)[18] is in terms of inelastic quasiparticle scattering processes off a strong spin fluctuation spectrum arizing from an oxygen deficient layer on the surface of the HTS. It is clear that the inelastic tunneling processes are present in many HTS junctions and by treating such processes using a realistic spin fluctuation spectrum, it is possible that the dip and peak features can be explained for the tunneling data and photoemission as well. If indeed, the anomalous dip is due to an inelastic tunneling process arising from a surface layer, then this might explain the observation by Shimada et al[19] of phonon structures in BSCCO 2212 using Schottky type junctions. This experiment is one of the few to not show the pronounced dip in BSCCO 2212. Perhaps in this case, the intimate contact of the semiconductor to BSCCO inhibits any oxygen-deficient, surface layer allowing predominantly elastic tunneling to be observed.

3.2 PCT junctions on $Tl_2Ba_2CuO_6$

It was recently reported by Chen et al[20,21] that PCT junctions on the single-layer Hg based cuprate, $HgBa_2CuO_4$ (Hg1201) exhibit BCS-like tunneling conductances for both SIN (Au tip) and SIS' (Nb tip) junctions. Given the choice of the two curves shown in Fig. 1, it was clear that the Hg1201 data were more compatible with an s-wave order parameter. Furthermore, the SIS' I(V) data exhibited sharp current onsets at the gap voltage making the Hg1201 material a potential candidate for quasiparticle based devices such as mixer-based photon detectors.[21] Typical gap parameters were Δ~13-16 meV, but one junction had Δ=24 meV. Considering that Hg1201 is tetragonal and has a single Cu-O layer per unit cell, we decided to examine a very similar compound, $Tl_2Ba_2CuO_6$ (Tl2201), which has the same structural properties as Hg1201 and a similar Tc (91 K-95 K). Single-crystals of Tl2201 approximately 0.5mm on edge were grown by Mogilevsky and

Hinks at Argonne. The PCT method offers distinct advantages for the study of such small crystals. Over 200 junctions have been made on 20 different crystals using both Au and Nb tips. The data are reproducible and we show a representative set of four junctions from two samples in Fig. 6. Note here the voltage is that of the sample relative to the tip.

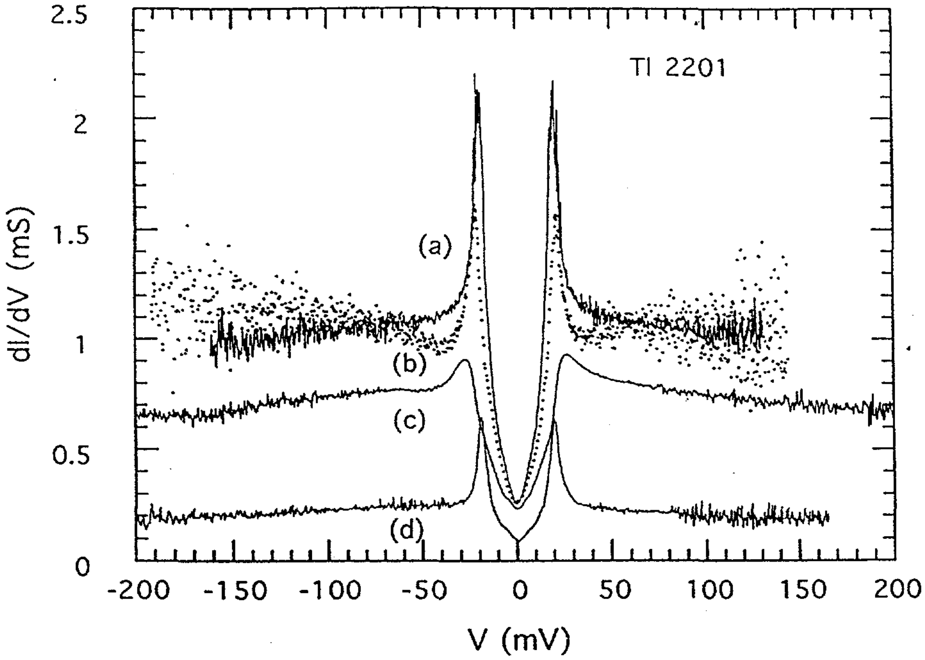

Fig. 6. Representative set of SIN (Au tip) junction conductances for two crystals of Tl2201. The curves are labelled (a) through (d) going from top to bottom. Junction (b) is represented by dots, all others by solid lines. The voltage, V, is that of the sample relative to the tip.

First note the background shape which is weakly decreasing with applied bias voltage similar to that typically found in BSCCO 2212. The junction conductances are characterized (in many cases) by very sharp conductance peaks located near 20 mV. Estimating Δ=20 meV and using 91 K for T_C one obtains $2\Delta/kTc=5.1$. We note that the ratio of peak conductance to the estimated normal state at $\pm$20 mV is often greater than 2 and in some cases is as high as 3.5, the latter value being one of the largest ratios we are aware of for any SIN cuprate junction.

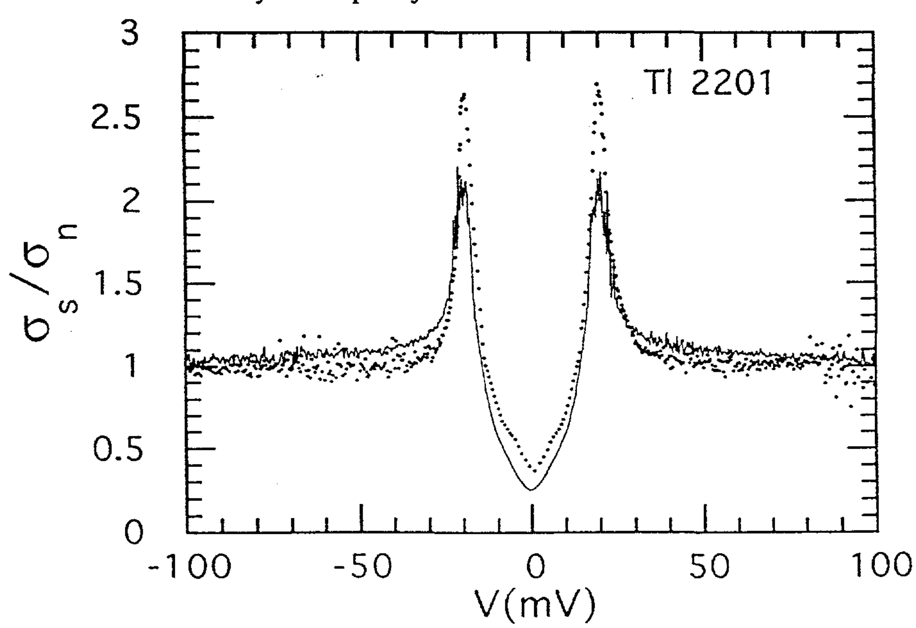

Figure 7. Normalized conductances of Tl2201 junctions (a) and (d) of Fi.g. 7.

Junction (c) of Fig. 6 is the most broadened of the four and the conductance peak voltage has been shifted to a slightly larger value. This shift of the conductance peak voltage to higher values for broadened junctions is a common effect in oxide superconductors.[2] We argue that those junctions with the sharpest conductance peaks are the most representative of the bulk dos of Tl2201 and thus junctions (a) and (d) from two different crystals are chosen for futher consideration. The curves were normalized by a constant and are shown in Fig. 7. Comparing the normalized conductances to the dos plots of Fig. 1, it appears that Tl2201 more closely resembles a superconductor with $d_{x^2-y^2}$ symmetry. This is evident from the pronounced cusp feature in the data at zero bias. This cusp can be seen in all of the junctions of Fig. 6 and is a general feature of most of the SIN junctions we have studied. Quantitative fits of the data using the d-wave model discussed above are currently underway. In comparing these data to those of Hg1201, an obvious question arises. Why would two such similar compounds display junction characteristics indicating different gap symmetries?

4. SUMMARY

Quasiparticle tunneling data on HTS cuprates can potentially give important information on pairing state symmetry and mechanism. However, it is clear that in many cases unusual background shapes occur in the tunneling conductances. The fact that these background shapes can vary from increasing with bias (often in a linear fashion) to decreasing with bias for the same cuprate suggests that they are arising from a conduction process which is not elastic tunneling. A strong likelihood is inelastic tunneling which has been shown to produce linearly increasing backgrounds when the spin fluctuation spectrum is flat. Perhaps a more rigorous treatment of inelastic tunneling, including a realistic spin fluctuation spectrum, a tight binding band structure and directional tunneling effects will result in an explanation of all of the background shapes. The background must be understood before a quantitative analysis of the elastic tunneling part can be undertaken. One way of minimizing the problem is to focus on those junctions which have a relatively weak voltage dependent background at least for one bias direction. This approach worked with NCCO where $\alpha^2F(\omega)$ spectra have been obtained.

The origin of the anomalous dip feature is still not understood. Our observation that the dip feature scales with the energy gap puts severe constraints on its interpretation. The possibility of an energy dependent decay rate, $\Gamma(\omega)$, is an attractive one and readily leads to the observed scaling behavior. However, there are still aspects of the dip feature which are not explained by this model, for example, the observed asymmetry of the effect with bias voltage as found in BSCCO 2212. There is also the possibility that the dip feature is associated with the same inelastic tunneling processes that affect the general background shape.

Quasiparticle tunneling cannot probe the sign of the order parameter and therefore cannot directly determine the pairing state symmetry. However, strong inferences can be made. For example, the observation of highly reproducible gap values in NCCO strongly supports an isotropic s-wave state and this is verified by a number of other experiments on NCCO including penetration depth and Raman scattering. The origin of the small sub gap conductance remains a puzzle. Other electron doped systems, including the infinite layer cuprate, $Sr_{1-x}Nd_xCuO_2$ display similar gap reproducibility which leads to the suggestion that they may be s-wave as well. Another way to probe the pairing state is to note that the s-wave and d-wave densities of states are quite different and should be reflected in the quasiparticle tunneling spectra. The observation of low, flat sub-gap conductances in junctions of Hg1201, for example, are difficult to reconcile with a d-wave pairing state. Strong directional tunneling effects must be invoked to explain the Hg1201 data within a d-wave scenario. In the case of the similar compound, Tl2201, the tunneling dos looks very much like the d-wave dos, including a pronounced cusp feature. In this case one must argue that there are no preferred tunneling directions and the total dos is being probed. For other cuprates, the presence of sub-gap conductance without any obvious cusp feature gives little information about pairing symmetry.

Finally, it should be mentioned that under no circumstances has the van Hove singularity feature ever been observed in PCT or to our knowledge, any other tunneling method. Such a distinctive feature as seen in Fig. 1 might be expected to show up in the tunneling data. Being strictly a band structure effect, its absence is probably linked to the common absence of band structure effects in the tunneling dos as found in conventional metals.

5. ACKNOWLEDGEMENTS

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