

***ab*-plane tunneling spectroscopy of underdoped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_y$** 

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We have measured the tunneling spectra of underdoped and slightly overdoped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_y$  single crystals in the *ab* plane at different temperatures, ranging from 4.2 K to above  $T_c$ . Gaplike spectra have been observed at low temperatures in both underdoped and overdoped compounds. However, overdoped samples do not display any gaplike features at temperatures above  $T_c$ . On the contrary, the underdoped samples do display gaplike features above  $T_c$ . The tunneling spectra in the pseudogap state are broader, giving rise to a larger gap value as compared to the superconducting state gap. [S0163-1829(98)51038-7]

The observation of an energy gap above the critical temperature (the so-called ‘‘pseudogap’’) in underdoped high- $T_c$  cuprate superconductors has given a totally new direction to the field of high- $T_c$  superconductivity research. There are several different experiments confirming the presence of a pseudogap in different high- $T_c$  materials,<sup>1-7</sup> including tunneling.<sup>8</sup> There are various theoretical models explaining the origin of pseudogap type behavior. One possible explanation is the existence of Cooper pairs above  $T_c$ . As we have observed in the present experiment and from other groups’ results,<sup>8,9</sup> the critical temperature of cuprates decreases while the energy gap increases, as the oxygen concentration (or hole concentration) is reduced. This is quite unexpected from the viewpoint of BCS theory. Moreover, from resistivity and tunneling data, the low temperature gap seems to scale with the ‘‘pseudogap phase’’ onset temperature  $T^*$ .<sup>9</sup> This may imply that the coupling parameter (effective  $T_c$ , i.e.,  $T^*$ ) increases with the oxygen underdoping. However, at higher temperatures the phase fluctuations give rise to pair breaking scattering which destroys the long-range superconducting order well below  $T^*$ .<sup>10</sup> As a result the Cooper pairs exist above  $T_c$  without any bulk superconductivity. On the other hand, from neutron scattering<sup>11</sup> and NMR measurements<sup>12</sup> it is difficult to rule out the magnetic origin of the pseudogap.<sup>13</sup> There are other possible models, including a Van-Hove singularity at the Fermi level,<sup>14</sup> a strong electron-phonon interaction giving rise to a polaron-bipolaron type gap,<sup>15</sup> an SO(5) model<sup>16</sup> or a spin-charge separation mechanism.<sup>17</sup> Though the exact mechanism of the pseudogap is not fully determined, it is clear that the normal state of these cuprates does not follow a simple Fermi liquid type behavior.

Tunneling spectroscopy is one of the most direct and well established methods for measuring the energy gap in superconductors. There have been numerous tunneling measurements on conventional<sup>18</sup> as well as the high- $T_c$  superconductors.<sup>19</sup> It provides better energy resolution than most other measurements, hence enabling researchers to study the change in the magnitude of gap with different controlled parameters. Scanning tunneling microscope (STM) provides a versatile method of forming a tunnel junction in which the tunneling barrier can be readily changed, although it is sensitive to mechanical noise and temperature variation. So far, tunneling measurements on the pseudogap have been

in the *c*-axis, which is expected to be different from the *ab*-plane gap, given the layered structure of these compounds. In this paper, we report on the results from a series of experiments in which we have studied the temperature dependence of the *ab*-plane tunneling spectra of the junctions formed between a platinum foil and single crystals of underdoped and slightly overdoped  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_y$  ( $\text{Bi}2212$ ).

$\text{Bi}2212$  is one of the most common samples studied by STM because it has a relatively high  $T_c$  and good quality single crystals can be easily grown. Furthermore the oxygen level at the surface is more stable than some of the other cuprates like  $\text{YBa}_2\text{Cu}_3\text{O}_y$ . The single crystals we used in this experiment were grown in air by methods reported by Mitzi *et al.*<sup>20</sup> These are slightly overdoped with a  $T_c$  of 85 K. Samples from the same batch have been fully characterized and used previously in other experiments.<sup>21</sup> The underdoped crystals were obtained by annealing these slightly overdoped crystals under 10 mTorr vacuum at 750 °C for a few hours and quenching into liquid nitrogen to avoid oxygen inhomogeneity. The critical temperature  $T_c$  of the underdoped sample was measured to be 70 K by four point resistivity measurement and it was found to be very sensitive to the annealing atmosphere and temperature. The same  $T_c$  was also determined independently by magnetization measurements with a superconducting quantum interference device (SQUID) magnetometer in an ac magnetic field. The real and imaginary parts of the magnetization are shown in Fig. 1. A consistent  $T_c$  and sharp transition establish the reproducibility and homogeneity of our samples. The quality of the crystals was further established by single crystal x-ray diffraction (XRD). The lattice parameters from XRD were found to be very close to those for the nonannealed crystals, within less than a percent. The superstructure modulation peaks were observed and used to distinguish between *a* and *b* axes, since the modulation exists only along *b* direction. In all the crystals, no superstructure peaks showed fractional indices along two directions, ruling out the twinning of the crystals. The XRD was also used to determine the crystal orientation.

The tunneling junction was formed using a low temperature STM. A similar method has been used by Kane *et al.*<sup>21</sup> in earlier studies on gap anisotropy. The quality of the tunneling edge was established by scanning electron microscopy (SEM) imaging. A nichrome wire was used as heater and the

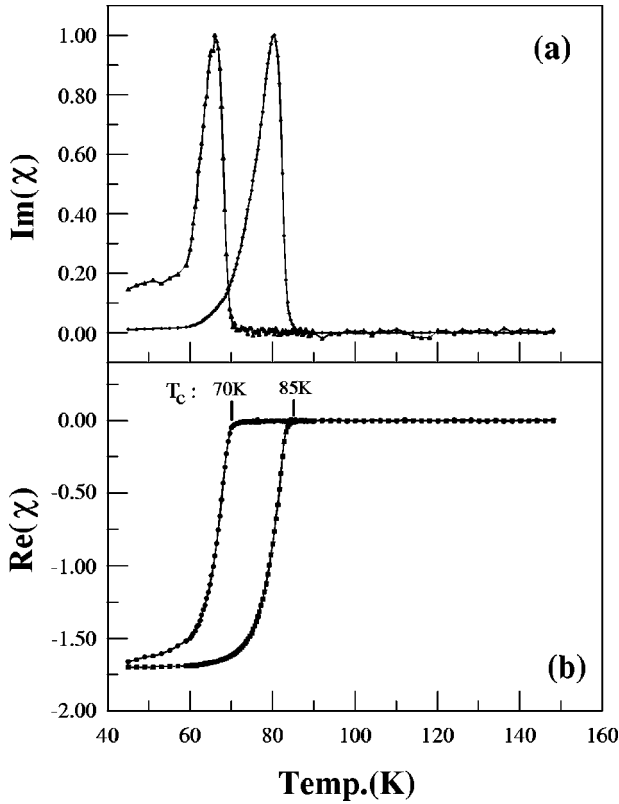


FIG. 1. (a) The imaginary and (b) real parts of the magnetic susceptibility of slightly overdoped ( $T_c = 85$  K) and underdoped ( $T_c = 70$  K)  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_y$  measured by SQUID magnetometer in an ac-magnetic field with 10 Hz frequency and 2.0 G amplitude.

temperature was stabilized with a temperature controller.<sup>22</sup> The temperature was stable within 10 mK, ensuring the stability of the junction. The junction was formed between a platinum foil and freshly cut single crystals. Before each change in temperature the sample was retracted to avoid crash between the sample and the Pt foil resulting from differential thermal expansion of the STM. As a result most of the tunneling curves presented here correspond to different junctions. The tunneling was performed on the edge of the crystal, along the maximum gap direction determined by XRD as described in Ref. 21. From photoemission studies, the underdoped Bi2212 has the same gap anisotropy in the superconducting state as well as in the pseudogap state and maximum pseudogap occurs in the same direction as the superconducting gap maximum.<sup>3</sup> We performed our tunneling experiments near the maximum gap direction so as to minimize the thermal effects at high temperatures.

Typical tunneling conductance curves for the overdoped compound at different temperatures are shown in Fig. 2. As we have pointed out before, each curve corresponds to a different junction. The tunneling curves have been normalized with respect to the conductance at  $-100$  mV and the higher temperature curves have been shifted up for clarity. The junction resistance was kept between 100 k $\Omega$  and 10 M $\Omega$ . It is slightly smaller than the junction resistance of most microscopy work, because of the larger tunneling area in our experimental configuration. The spectra show the quasiparticle peaks at  $\pm\Delta_p$ , where  $\Delta_p$  is defined as half the peak to peak separation ( $\Delta_{p-p}$ ) in our tunneling spectra. There is no exact method of extracting the energy gap from the tunneling

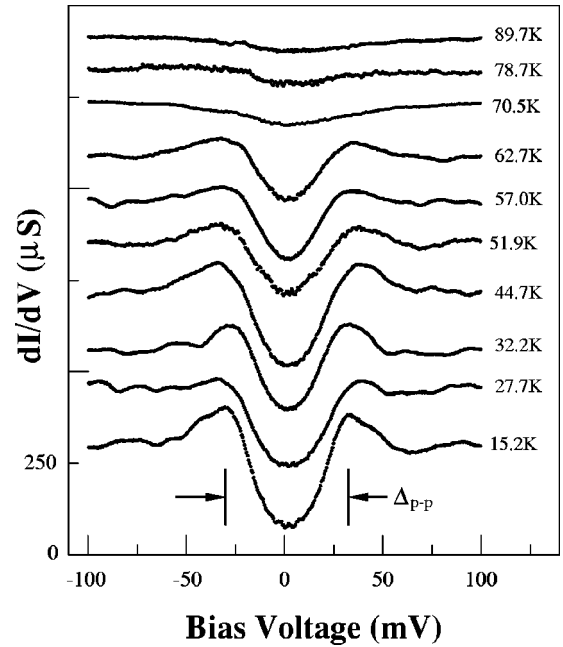


FIG. 2. Tunneling spectra of slightly overdoped Bi2212 at various temperatures. The curves are normalized with respect to the conductance at  $-100$  mV and the higher temperature curves have been shifted up for clearance. The vertical scale corresponds to the lowermost curve.

spectra, since the exact functional form of the density of states for high- $T_c$  superconductors is not known, therefore we use this  $\Delta_p$  as a measure of the gap. This  $\Delta_p$  can be larger than the actual superconducting gap,  $\Delta$ , because of thermal, or other smearing effects. The smearing also gives rise to a finite zero bias conductance (ZBC) which increases with smearing and, therefore, with temperature. The zero bias conductance for low temperature spectra was found to be less than 15% of the background conductance, which is larger than most of the  $c$ -axis measurements on the same compound. This may arise from processes other than elastic tunneling; however, in  $ab$ -plane tunneling, it can be attributed to the tunneling contributions from different directions in a narrow angular cone centered at the maximum gap angle.<sup>21</sup> This cannot be the case for  $c$ -axis tunneling, since there is a single gap contributing to the spectra in this case. The dip feature, seen at the positive bias of the low temperature curve, is actually consistent with the dip feature seen by other tunneling measurements.<sup>23</sup> In those measurements the dip feature occurred at the negative side because the tip was kept at zero potential as opposed to our measurements, where the sample was kept at zero potential.

The gap value,  $\Delta_p$ , at low temperatures for the slightly overdoped compound was found to be 30 meV ( $\pm 2$  meV), and with a  $T_c$  of 85 K,  $2\Delta_p/kT_c = 8.2(\pm 0.5)$ . This value is consistent with other measurements.<sup>8,21,23</sup> The tunneling direction for this set of data determined by single crystal x-ray diffraction was along the  $a$  axis. From our previous studies the maximum gap occurs along  $a$  direction, which is  $45^\circ$  off as compared to the photoemission measurements. On reap-proach or on changing the  $x$ - $y$  voltage of the piezoelectric transducer,  $\Delta_{p-p}$  varied slightly for different junctions (within  $\pm 2$  mV). This may be attributed to the fact that different junctions may occur in different regions and at slightly

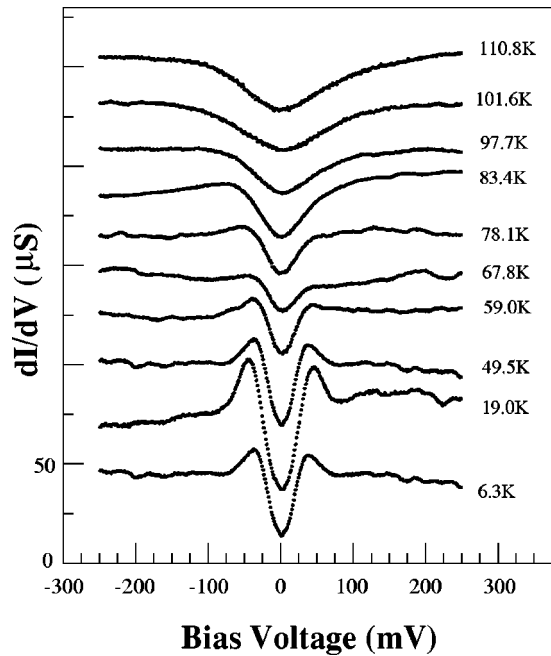


FIG. 3. Tunneling spectra of underdoped Bi2212 at various temperatures. The curves are normalized with respect to the conductance at  $-250$  mV and the higher temperature curves have been shifted up for clearance. The vertical scale corresponds to the lowest curve. Note the different bias voltage scale as compared to Fig. 2.

different tunneling angles, as this compound has some inhomogeneities and the gap is anisotropic.<sup>21,24</sup> The peak to peak separation does not change within the error as the temperature is raised. Above 85 K gaplike features are essentially absent. However a slightly parabolic background persists which extends to fairly high bias voltages. This parabolic background above  $T_c$  has been studied by other groups,<sup>25</sup> and should not be considered as a gaplike feature in the tunneling conductance curves.

Typical tunneling spectra for the underdoped material are shown in Fig. 3. The low temperature gap value as measured from the peak to peak separation,  $\Delta_p$ , is 38 meV, larger than the gap value for the optimally doped compound. From this the  $2\Delta_p/kT_c$  is 12.6, which is large partly because of the reduction in  $T_c$ . In Fig. 3 each curve corresponds to a different junction and the curves have been normalized with respect to the conductance at  $-250$  mV and shifted upward for clarity (note the different voltage scale as compared to Fig. 2). The features below  $T_c$  are similar to those of slightly overdoped material. The peak to peak separation remains the same within error as the temperature is increased. However unlike the overdoped compound, the gaplike features persist far above  $T_c$  as opposed to the overdoped compound, where the gap feature becomes insignificant within a few K above  $T_c$ . There are some noticeable differences between the spectra of underdoped and slightly overdoped compound and between the curves below  $T_c$  and the ones at high temperatures.

One feature for underdoped compound is the asymmetry of the spectra with respect to the sign of the bias voltage. This asymmetry exists at all temperatures, below as well as above  $T_c$ ; while in the overdoped case there is no asymmetry in the spectra. The shoulder (or peak) at positive bias

(i.e., below the sample's Fermi level) is more rounded as compared to that at negative bias. This asymmetry can play a significant role in explaining the pseudogap. It can be explained by density of states effects such as modeled by a two band picture<sup>26</sup> or it has also been found in the tunneling spectra calculated from some of the recent pseudogap models.<sup>27,28</sup> On the other hand, such asymmetric features have been seen in several other tunneling experiments<sup>29-31</sup> with nonsuperconducting electrodes. For instance, the asymmetric linear background<sup>30</sup> and sharp asymmetric features<sup>31</sup> have been observed in tunneling measurements and have been attributed to the inelastic tunneling due to the excitations in the barrier. The asymmetry arises because of the asymmetric location of these excitations in the barrier. The surface of high- $T_c$  compounds is not very well known chemically. So it is possible that the surface layer of underdoped material has a continuous band of excitation modes providing extra channels for tunneling. The tunneling probability depends on the electron energy. When an electron tunnels from a normal metal to the superconducting side it gets scattered inelastically by the surface only after it has tunneled through the barrier. However, when it tunnels from the superconducting side it gets scattered inelastically, losing some energy, before tunneling to the normal side and therefore reducing the tunneling probability. This will give rise to a reduction in tunneling current, and therefore reduction in conductivity, when the electron tunnels from superconductor to the metal electrode. This corresponds to a positive bias on the metal electrode, consistent with the present observation.

Another noticeable feature is the smearing of the spectra above  $T_c$ . The curves are much more smeared in pseudogap state as compared to the sharply featured spectra in superconducting state. As a result of this, the conductance ratio at zero bias to flat background above  $T_c$  is much larger and the quasiparticle peaklike features are not observed. Instead, we see a wide dip at zero bias with two broad shoulders. This gives the effective gap value ( $\Delta_p \sim 100$  meV from shoulder to shoulder separation) much larger in the pseudogap state as compared to the superconducting state. The peak to peak separation for the numerically generated tunneling spectra using a BCS density of states (DOS) increases as  $\Delta_{p-p} = 2\Delta_0 + 4kT$ , with  $\Delta_0$ , the gap parameter in the DOS, is taken to be independent of temperature.  $4kT$  increases only 21 meV between 50 K to 110 K; however, the peak to peak separation in the pseudogap state (of the order of 200 meV at 110 K) is much larger than that of the superconducting state (76 meV). Therefore, thermal effects alone cannot explain the broadening.

Norman *et al.* have fitted the photoemission spectra for Bi2212 for two different underdoped compounds with critical temperatures 83 and 77 K, using a phenomenological model involving three parameters  $\Gamma_1$ ,  $\Gamma_0$  and  $\Delta$  representing single particle scattering rate, inverse lifetime of pairs, and energy gap, respectively.<sup>32</sup> From the fitting they found that  $\Gamma_0$  has a sharp increase at  $T_c$  for both the samples and  $\Delta$  stays roughly constant for 83 K compound across  $T_c$ ; however,  $\Delta$  increases for the 77 K compound above  $T_c$  without any sharp transition in it at  $T_c$ . The underdoped sample in our experiment is heavily underdoped with a  $T_c$  of 70 K, therefore it is possible that this compound has a large gap in pseudogap state. A large  $\Gamma_1$  above  $T_c$ , a finite  $\Gamma_0$  and the

thermal smearing are also responsible for increasing the separation between shoulders at high temperatures. This kind of phenomenological model is based on the existence of Cooper pairs in the pseudogap state, and, has been explained more rigorously, by Franz *et al.*, on the basis of phase fluctuations giving rise to finite lifetime of Cooper pairs above  $T_c$ .<sup>33</sup> Though the existence of Cooper pairs above  $T_c$  is not proved and requires further investigation, this phenomenological form of self-energy describes the photoemission as well as tunneling data reasonably well.<sup>32,33</sup>

In summary, our *ab*-plane tunneling studies of Bi2212 show that a slightly overdoped compound ( $T_c=85$  K) does not show the gaplike features above  $T_c$ ; however, the underdoped compound ( $T_c=70$  K) does show gaplike features in the spectra. The spectra above  $T_c$ , for underdoped com-

pound, show a larger peak to peak separation as compared to the superconducting state. This can be attributed mainly to a larger gap value in the pseudogap state as compared to the superconducting gap, and also to other smearing effects.

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