Predominantly Superconducting Origin of Large Energy Gaps in Underdoped Bi$_2$Sr$_2$CaCu$_2$O$_{8-\delta}$ from Tunneling Spectroscopy

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Abstract

New tunneling data are reported in underdoped Bi$_2$Sr$_2$CaCu$_2$O$_{8-\delta}$ using superconductor-insulator-superconductor break junctions. Energy gaps, $\Delta$, of 51±2, 54±2 and 57±3 meV are observed for three crystals with $T_c$=77, 74, and 70 K respectively. These energy gaps are nearly three times larger than for overdoped crystals with similar $T_c$. Detailed examination of tunneling spectra over a wide doping range from underdoped to overdoped, including the Josephson $I_cR_n$ product, indicate that these energy gaps are predominantly of superconducting origin.

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Efforts to understand the mechanism of pairing in high-$T_c$ superconducting (HTS) cuprates are currently focused on the unusual doping dependences of superconducting and normal state properties. In particular, underdoped HTS compounds have exhibited pseudogap phenomena above $T_c$ in both spin and charge excitations. Recently, tunneling studies on Bi$_2$Sr$_2$CaCu$_2$O$_{8-\delta}$ (Bi2212) in the superconducting state have shown a remarkable effect whereby the energy gap exhibits a strong, monotonic dependence on doping, increasing substantially in the underdoped phase even as $T_c$ decreases. It has been pointed out that the smooth dependence on doping may nevertheless originate from a quasiparticle gap that evolves from superconducting character in the overdoped phase to another type (e.g. charge density wave, spin density wave, etc.) in the underdoped phase. While the measured tunneling gap vs. doping is consistent with other probes including angle resolved photoemission (ARPES) and Raman, it is at odds with some measurements that support a superconducting order parameter scaling with $T_c$. Thus a critical question is whether this relatively large energy gap originates entirely from superconducting pairing or has a contribution from some other electronic effect. Here we address the nature of the gap measured by tunneling and report new data in very underdoped Bi2212 by superconductor-insulator-superconductor (SIS) break junctions. Energy gaps, $\Delta$, of $51\pm2$, $54\pm2$ and $57\pm3$ meV are observed for three underdoped crystals with $T_c=77$, 74, and 70 K respectively, extending the previously reported trend further into the underdoped regime. Detailed examination of the tunneling spectra over a wide doping range, including the Josephson $I_cR_n$ product, show that these energy gaps are predominantly of superconducting origin.

Historically, tunneling studies have been relied upon to examine the magnitude of the superconducting energy gap in both conventional and HTS. In particular, SIS junctions provide an accurate measure of $2\Delta$ from the peak in tunneling conductance which is only weakly affected by thermal smearing or quasiparticle scattering. However, the large magnitudes of energy gaps observed here lead to such extraordinarily large values of $2\Delta/kT_c$ (as high as 20) that it is necessary to examine carefully the entire tunneling spectrum to clarify the physical origin of these energy gaps. Most theoretical models of HTS stress
the importance of electronic correlations such as spin density waves \[1\] and its predecessors \[11\], or charge density waves in the underdoped phase \[3,12\] which might give rise to momentum-dependent quasiparticle excitation gaps, $\Delta_c(k)$ in addition to those arising from superconductivity, $\Delta_s(k)$. In these "two-gap" scenarios the energy gaps often add in quadrature \[3,12\] such that the total energy gap $\Delta = (\Delta_s^2 + \Delta_c^2)^{1/2}$. Since these other correlation gaps are often used to explain the pseudogap above $T_c$, our investigation here has a bearing on this issue as well. We argue first that if two distinct gaps exist, (1) they should have very different doping and temperature dependences; and (2) it is unlikely that $\Delta_s(k)$ and $\Delta_c(k)$ will have identical momentum dependences. Thus the quasiparticle density of states (DOS) should exhibit distinct features corresponding to each gap. We observe no evidence of a second gap feature and it will be shown that the shape of the gap region spectrum smoothly evolves with doping, with features changing mainly in energy scale.

We also examine a property of SIS junctions that depends solely on superconductivity, the Josephson current. A statistical summary of the Josephson $I_cR_n$ products of over 40 SIS junctions is presented and it is shown that the largest values (both average and maximum) are all found in underdoped crystals, where the measured quasiparticle gap is the largest. This links the measured quasiparticle gap to a purely superconducting energy scale, the Josephson strength. Thus we are forced to conclude that over the range of doping studied (from 70 K underdoped to 62 K overdoped) the measured quasiparticle gap appears to be due predominantly to superconductivity.

We grew high quality single crystals using a slightly modified floating-zone process as described elsewhere. \[2\] This yields an optimal $T_c$ onset of 95 K and the doping is varied through the oxygen concentration. The 70 K underdoped crystal was prepared by a different procedure (see ref. \[13\]) and there is good agreement among the differently processed samples. Both SIS break junctions and SIN (superconductor-insulator-normal metal) junctions were prepared on freshly cleaved surfaces by a point contact technique with Au tip. \[2,14,15\] Tunneling spectra and gap values in the SIN junctions \[16\] are consistent with those presented here but in this paper we focus on the SIS junctions.
In Fig. 1 is shown the $dI/dV$ vs. $V$ for an SIS junction on the most underdoped crystal with $T_c = 70$ K. The conductance data have been normalized by a constant which is the conductance at $V= 340$ mV. The shape of the conductance is similar to that found on optimally doped crystals [2] exhibiting sharp conductance peaks ($eV_p = 2\Delta$), subgap conductance and pronounced dip features at $eV= 3\Delta$. At zero bias there is a small Josephson current in the $I(V)$ curve (inset of Fig. 1) which shows up as a conductance peak. The dashed line of Fig. 1 is a fit using a weighted, momentum averaged $d$-wave density of states (DOS). [17] The fit is good except for the obvious discrepancies at the dips, and the weighting factor indicates that there is preferential tunneling along the $(\pi,0)$ point, the maximum of the $d$-wave gap. This analysis gives $\Delta = 60$ meV for the maximum $d$-wave gap and $\Gamma = 6$ meV where $\Gamma$ is a quasiparticle scattering rate. The data of Fig. 1 can also be adequately fit with a smeared BCS DOS leading to $\Delta=57$ meV, which is exactly half of conductance peak voltage, $V_p$. Thus far, reproducibility on this crystal is limited to four separate SIS junctions, but in each case a well-defined energy gap is found with $\Delta= 57\pm3$ meV. The large energy gaps found here extends the previously reported trend [2] further into the underdoped regime and leads to a value of $2\Delta/kT_c= 20$.

In Fig. 2 we show representative SIS tunneling spectra for a wide doping range from overdoped with a $T_c = 62$ K to underdoped with a $T_c = 70$ K. The Josephson peak has been removed for clarity. Four of the curves in Fig. 2 have been published previously [2,14] but are included to display the trend with doping. To compare the spectra, which have widely different gap values, we rescale the voltage axis by $V_p/2$. In this way the voltage scale is approximately in units of $\Delta/e$. The bottom three curves of Fig. 2 are new results of this study and in the cases of the other underdoped crystals ($T_c = 74$ K, 77 K) the curves are representative of many different junctions formed on each crystal. Well-defined gap structure was reproducibly observed with energy gaps, $\Delta$, of $51\pm2$, and $54\pm2$ for the crystals with $T_c=77$ and 74 K respectively. What is observed in Fig. 2 is a smooth evolution of the spectra from overdoped to underdoped, with a single gap feature that grows monotonically as the doping level is reduced. All of the curves exhibit subgap conductance, which as shown in
Fig. 1, can be attributed to a $d$-wave DOS. A notable feature in the SIS junctions of Fig. 2 is the pronounced dip structure which remains at $eV\sim 3\Delta$ over the entire range of this study, i.e. $\Delta = 15$ meV - 60 meV. In none of the curves is there seen any evidence of a second peak in the conductance which would be a clear indication of another gap in the quasiparticle spectrum. Rather, what is most striking is that the entire gap region spectrum has nearly the same shape over the entire doping range, and all that is changing is the energy scale of the spectral features. Thus we find no evidence that the nature of the energy gap is changing over the doping range.

The $I_cR_n$ value for the junction in Fig. 1 is about 2 mV as obtained from the I(V) curve, but values as large as 14 mV have been observed for one of the other junctions of this same crystal. Here $R_n$ is estimated from the high bias conductance which is relatively constant as shown in Fig. 2. Large values of $I_cR_n$ (15 mV-25 mV) were previously reported for an 83 K underdoped sample. Table I shows the average and maximum $I_cR_n$ values for over 40 SIS junctions on Bi2212 for a variety of doping levels. We find that the average $I_cR_n$ increases with decreased doping and that the three highest values among all the junctions are found in underdoped samples, consistent with the large quasiparticle gaps observed. Although the statistical distribution is still rough at present, the trend indicates that the quasiparticle gap is linked to the Josephson strength, $I_cR_n$, a purely superconducting energy scale.

The temperature dependence of tunneling conductance was measured in some cases and in Fig. 3 are shown the results for another junction on the $T_c = 77$ K underdoped crystal. The high bias junction resistance is $\sim 11$ kΩ and all of the data have been normalized by this constant value. Here the Josephson peak at zero bias is left in. As clearly seen in this figure, the superconducting gap peak, $V_p$, changes very little up to 50 K, but for $T > 60$ K the magnitude of the superconducting gap starts decreasing and states at Fermi level start filling in. The quasiparticle peak coming from superconducting gap and the zero-bias peak coming from Josephson current continuously disappear near the bulk $T_c$. This is important because it means that the local $T_c$ of the junction is essentially the same as the bulk $T_c$. The decrease of the gap magnitude with temperature is also seen directly in the raw data.
for an 83 K underdoped and a 95 K optimal doped crystal [2] and is consistent with other SIS break junctions on Bi2212. [18]

To attempt a more quantitative analysis, the superconducting gap, $\Delta(T)$, and quasiparticle scattering rate, $\Gamma(T)$, as a function of temperature have been estimated by fitting the data in Fig. 3 to a simple model for SIS junctions [2,9] which uses a smeared BCS DOS to describe the Bi2212. As discussed earlier in the examination of Fig. 1 this analysis might lead to a minor discrepancy in the magnitude of the gap when compared to a $d$-wave model, but the simpler smeared BCS function makes the analysis much easier. The results of this procedure are shown in Fig. 4. The principal result is that the gap magnitude decreases significantly as $T$ increases near $T_c$. For $T > 72$ K the conductance data are so smeared out that no accurate values for $\Delta$ and $\Gamma$ can be obtained. The decrease in gap magnitude near $T_c$ is in disagreement with the interpretation of recent STM experiments on underdoped Bi2212 where the raw data seem to suggest a $T$-independent gap. [19] We note, however, that $\Delta(T)$ cannot be inferred directly in SIN junctions due to importance of fermi factors in the tunneling conductance. We estimate the maximum Josephson current $I_{c}^*$ from the measured conductance peak at zero bias and use the junction resistance $R_n$ at high bias to plot the temperature dependence of the Josephson strength, $I_{c}^* R_n$, normalized to the value obtained at $T = 4.2$ K. The normalized Josephson strength plotted in Fig. 4 is approximate and it is used only to estimate the $T_c$ of the junction which can be seen is very close to the measured bulk $T_c$ of the crystal (77 K). We thus can say that the large energy gaps in our underdoped crystals correspond to the measured bulk $T_c$ and are not a consequence of some local deviation in stoichiometry.

Above the junction $T_c$ the quasiparticle gap feature has essentially disappeared and only a very weak depression in the conductance at zero bias remains. This behavior is consistent with our previous $T$-dependent SIS data [2] on an 83 K underdoped crystal and a 95 K optimally doped crystal as well as other SIS data in the literature. [18] We thus find no clear evidence of a pseudogap above $T_c$ in the SIS tunneling data, again in contrast to the strong gap feature observed in recent STM experiments. [19] One possible explanation is
that the T-dependent pseudogap is highly anisotropic in k-space as is indicated in ARPES. 

Since the SIS tunneling probes a momentum averaged DOS as indicated by the strong subgap conductance in Figs. 1 and 2, then the effect of a highly anisotropic pseudogap on these junctions may give just a weak depression in conductance as is found.

We now summarize the doping dependence of the tunneling data. The shape of the SIS quasiparticle spectra are qualitatively the same for all doping levels, exhibiting a single gap feature at $eV=2\Delta$ that evolves smoothly with doping. The full gap-region spectrum scales with $\Delta$ indicating that the character of the energy gap is the same over the doping range studied. Josephson currents are found in all the SIS junctions and in some cases the magnitudes of the $I_cR_n$ product are very large, nearly 25% of $\Delta/e$. Furthermore, the three highest $I_cR_n$ values among more than 40 SIS junctions were all found in underdoped samples where the quasiparticle gap is the largest. These results indicate that the quasiparticle gaps are predominantly due to superconductivity and we find no evidence of another gap, $\Delta_c(k)$, at low temperatures. The trend of $\Delta$ vs. doping closely follows the doping dependence of $T^*$, the pseudogap temperature for Bi2212. Thus the large gaps in the underdoped region suggests that significant superconducting pairing correlations exist at temperatures between $T_c$ and $T^*$ and that $T_c$ is the temperature where long-range phase coherence sets in.

The temperature dependence of the superconducting gap, $\Delta(T)$, in underdoped crystals may also be providing a subtle clue about the nature of the superconducting fluctuations above $T_c$. While $\Delta$ does not close at $T_c$, there is nevertheless a significant decrease in its magnitude that is seen even in the raw data. This argues against a picture of tightly bound, pre-formed bosons below $T^*$, which would be expected to have a T-independent gap near $T_c$. Rather the data are more consistent with intermediate coupling regime scenarios for the pairing fluctuations which persist up to $T^*$.

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Fig. 1 Differential conductance, dI/dV, for SIS break junction on an underdoped single crystal of Bi2212 with T_c=70 K. The inset shows a Josephson current at zero bias.

Fig. 2 Normalized SIS tunneling conductances of Bi2212 with various doping levels from underdoped to overdoped. The voltage axis has been rescaled in units of $\Delta/e$.

Fig. 3 Temperature dependence of SIS tunneling conductance on an underdoped Bi2212 ($T_c=77$ K) break junction. For clarity, each conductance has been normalized by its value at 200 mV and (except for the 5 K curve) is offset vertically.

Fig. 4 Temperature dependence of superconducting gap $\Delta(T)$ (circle), quasiparticle scattering rate $\Gamma(T)$ (triangle) and normalized Josephson strength, $I^*_cR_n$ (square) (see text in details), where the normalization of $I^*_cR_n(T)$ has been done by $I^*_cR_n(4.2$ K). The full curve represents the BCS curve of superconducting gap $\Delta(T)$.
Table I  Doping dependence of $I_{cRn}$ values and superconducting gap, $\Delta$

|                  | Average $I_{cRn}$ (mV) (# of junctions) | Max. $I_{cRn}$ (mV) | $\Delta$ (meV) |
|------------------|-----------------------------------------|----------------------|----------------|
| Overdoped (T$_c$=62K) | 2.4 (10)                               | 7.0                  | 15-20          |
| Optimally-doped (T$_c$=92-95K) | 2.9 (14)                               | 7.8                  | 30-40          |
| Underdoped (T$_c$=70-83K)       | 4.1 (18)                               | 25, 14, 9.1         | 44-57          |
Fig. 1, N. Miyakawa et al.
Normalized Conductance $V/(\Delta/e)$

$T_c=70K$

$T_c=62K$

$\Delta=18\text{meV}$

$\Delta=27\text{meV}$

$\Delta=27.5\text{meV}$

$\Delta=37.5\text{meV}$

$\Delta=44.5\text{meV}$

$\Delta=51.5\text{meV}$

$\Delta=57\text{meV}$

Fig. 2 N. Miyakawa et al.
Fig. 3, N. Miyakawa et al.
Fig. 4, N. Miyakawa et al.