Predominantly Superconducting Origin of Large Energy Gaps in Underdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$ from Tunneling Spectroscopy

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New tunneling data are reported in $Bi_2Sr_2CaCu_2O_{8+\delta}$ which show quasiparticle excitation gaps, Δ, reaching values as high as 60 meV for underdoped crystals with $T_c = 70$ K. These energy gaps are nearly 3 times larger than those of overdoped crystals with similar T_c. Despite the large differences in gap magnitude, the tunneling spectra display qualitatively similar characteristics over the entire doping range. Detailed examination of the spectra, including the Josephson I_cR_n product measured in break junctions, indicates 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 dependencies of superconducting and normal state properties. In particular, underdoped HTS compounds have exhibited pseudogap phenomena in both spin and charge excitations [1] below a temperature T^* that is above T_c. Recently, tunneling [2] studies on $Bi_2Sr_2CaCu_2O_{8+\delta}$ (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. This energy gap follows the trend of T^* with doping [3], and thus if the gap has a superconducting origin it strongly suggests that the pseudogap state is due to some type of precursor superconductivity [1,4,5]. However, the smooth dependence on doping may also originate from a quasiparticle gap that evolves from superconducting character in the overdoped phase to another type (e.g., charge density wave) in the underdoped phase [6]. In support of this picture are some measurements that suggest a superconducting order parameter (or coherence gap) that scales with T_c [7–9]. Thus, a critical question is whether the large energy gap found in tunneling 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 more heavily underdoped Bi2212. Energy gaps, Δ, of 51 ± 2, 54 ± 2, and 58 ± 2 meV are observed for three underdoped crystals with $T_c = 77$, 74, and 70 K, respectively, extending the previously reported trend [2] 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.

Superconductor-insulator-superconductor (SIS) junctions provide an accurate measure of the gap from the peaks in tunneling conductance (at a bias voltage $|eV_p| = 2\Delta$) which are only weakly affected by thermal smearing or quasiparticle scattering [2,10]. 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 their physical origin. Most theoretical models of HTS stress the importance of electronic correlations which lead to spin density waves [11,12], or charge density waves in the underdoped phase [6,13]. These correlations give rise to gaps (or pseudogaps) in the electronic excitation spectrum, $\Delta_c(\mathbf{k})$, that are distinct from those arising from superconductivity, $\Delta_s(\mathbf{k})$. Since these other correlation gaps are used to explain pseudogap phenomena above T_c, our investigation here has a direct bearing on this issue. The experimental goal is to determine whether the single-particle excitation gap as measured in tunneling has contributions from both Δ_s and Δ_c. We argue that if two distinct gaps exist, (i) they should have different magnitudes as well as different doping and temperature dependencies; and (ii) it is unlikely that $\Delta_s(\mathbf{k})$ and $\Delta_c(\mathbf{k})$ will have identical momentum dependencies. Thus, the quasiparticle density of states (DOS) should exhibit *distinct* features corresponding to each gap. We observe that the shape of the gap region spectrum smoothly evolves with doping, with characteristic features (peak, dip, hump) changing mainly in energy scale. The peaks in tunneling conductance, which indicate a purely superconducting gap in the overdoped region, show no evidence that the origin of this gap has changed in the underdoped region.

We also examine a property of the SIS junction that depends solely on superconductivity, the Josephson current [2]. A statistical summary of the Josephson I_cR_n products of over 40 SIS junctions is presented, and it is shown that there are three I_cR_n values that are considerably larger than all of the others and these occur in underdoped crystals where the excitation gap is also 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, despite its unusual doping dependence, appears to be due predominantly to superconductivity. The implications of this result are discussed in more detail later in this Letter.

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. For this study, the range extends from overdoped ($T_c = 62$ K) to underdoped ($T_c = 70$ K). The 70 K underdoped crystal was prepared by a different procedure (see Ref. [14]), and there is good agreement among the differently processed samples. Both SIS break junctions and SIN ($N =$ normal metal) junctions were prepared on freshly cleaved surfaces by a point contact technique with Au tip [2,15,16], and the gap values from both types are consistent. Representative SIN tunneling conductances are shown in Fig. 1 over the entire doping range. A clear energy gap feature is indicated by the strong conductance peaks which are at energies, $|eV| \sim \Delta$. Values of Δ are obtained from the SIN conductances by fitting them to a d-wave density of states, and in the case of the most overdoped samples $\Delta(T)$ goes to zero at T_c as expected for a superconducting gap [17]. As the doping decreases from the overdoped region, the energy gap increases monotonically even in the underdoped region where T_c decreases. The inset of Fig. 1 shows the gap values obtained from both SIN and SIS junctions vs. hole concentration. The inset shows that, while the gap parameter for the overdoped region approaches the mean-field prediction for a d-wave superconductor [2], the most underdoped crystal has a Δ value near 60 meV, about 4 times the mean-field value. The Δ values more closely follow the measured T^* values [3] as indicated by the dashed line in the inset.

The spectra of Fig. 1 display a characteristic dip feature [2,15] at $|eV| = 2\Delta$ and a higher energy hump feature, most clearly seen at negative bias which corresponds to electron removal from the Bi2212. We find that, in addition to a general increase in broadening of the conductance peaks in the underdoped regime, the hump feature broadens and becomes quite prominent while also moving out to higher energies with decreased doping. The entire spectrum, including dip and hump, seems to scale with Δ. This scaling behavior is also seen in the SIS tunneling conductances as displayed in Fig. 2, where we show representative spectra over the same doping range. Josephson currents, which produce a large conductance peak at zero bias [2,18], have been removed for clarity. To demonstrate the scaling of the SIS spectra, we rescale the voltage axis by $V_p/2$ so that it is approximately in units of Δ/e. The bottom three curves of Fig. 2 are new SIS results of this study, and for the three crystals ($T_c = 70, 74, 77$ K) the curves are representative of many different junctions formed on each crystal. A notable feature in the SIS junctions of Fig. 2 is a more pronounced dip structure [2,15] which remains at $eV \sim 3\Delta$ over the entire range of this study; i.e., $\Delta = 15$–60 meV. This is the same dip feature found in the SIN data at $eV \sim 2\Delta$. What is most striking is that the gap region spectrum has nearly the same shape over the entire doping range, and

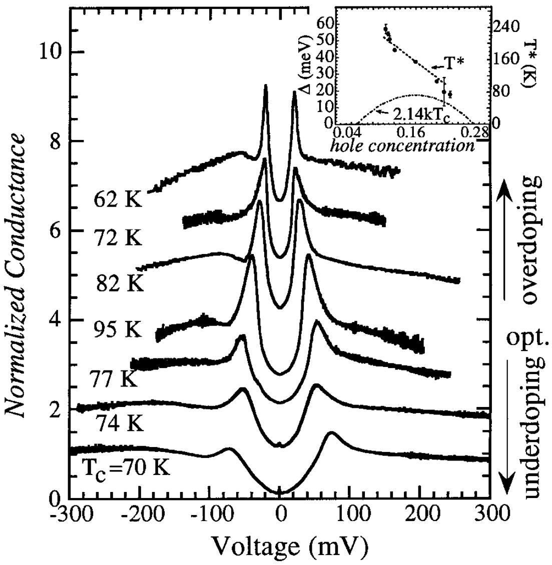

FIG. 1. Representative SIN tunneling conductances for various hole doping levels from underdoped to overdoped. Data are normalized by a constant and offset for clarity. The inset shows energy gap versus hole doping from SIN and SIS junctions (dots) along with a linear fit (dashed line) to the T^* values in Ref. [3].

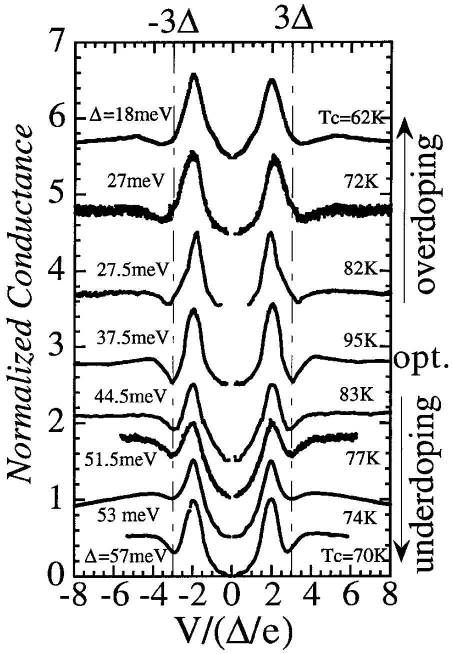

FIG. 2. Normalized SIS tunneling conductances of Bi2212 for various doping levels from underdoped to overdoped. The voltage axis has been rescaled in units of Δ/e.

TABLE I. Doping dependence of I_cR_n values and superconducting gap, Δ.

	Average I_cR_n (mV) (No. of junctions)	Max. I_cR_n (mV)	Δ (meV)
Overdoped ($T_c = 62$ K)	2.4 (10)	7.0	15–20
Near optimally doped ($T_c = 92–95$ K)	2.9 (14)	7.8	30–40
Underdoped ($T_c = 70–83$ K)	4.1 (18)	25, 14, 9.1	44–57

all that is changing is the energy scale of the spectral features. While our understanding of the dip feature is still incomplete, it is generally accepted to be associated with the opening of a superconducting gap, perhaps a strong coupling effect similar to the phonon structures found in junctions on conventional superconductors [2,10,15]. It therefore serves as an additional indicator that the gap value given by the conductance peaks in both SIN and SIS data is of superconducting origin.

Table I shows the average and maximum I_cR_n values for over 40 SIS junctions on Bi2212 for a variety of doping levels. Here, R_n is estimated from the high bias conductance which is relatively constant as shown in Fig. 2. 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. Large values of I_cR_n (15–25 mV) were previously reported for an 83 K underdoped sample [2], and as the table shows such values are significantly larger than typically found in overdoped crystals. 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 an SIS junction on the $T_c = 77$ K underdoped crystal. Here, the Josephson peak at zero bias is left in. As clearly seen in this figure, the superconducting gap peak at 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 the Fermi level start filling in. The quasiparticle peak coming from the superconducting gap and the zero-bias peak coming from Josephson current continuously disappear near the bulk T_c. The decrease of the gap magnitude with temperature is also seen directly in the raw SIS data for an 83 K underdoped and a 95 K optimal doped crystal [2] and is consistent with other SIS break junctions on Bi2212 [19].

To attempt a more quantitative analysis, the gap parameter, $\Delta(T)$, and quasiparticle scattering rate, $\Gamma(T)$, as a function of temperature have been estimated by fitting the data in Fig. 3 to a model for SIS junctions [2,10] which uses a smeared BCS DOS to describe the Bi2212. This analysis leads to a slightly smaller estimate in the magnitude of the gap when compared to a d-wave model [2], but is used for simplicity. 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. If this large gap had been due to an electronic effect other than superconductivity, e.g., a charge density wave, it would be very difficult to understand the rapid reduction of magnitude near T_c. For $T > 72$ K, the conductance data are so smeared out that no accurate values for Δ and Γ can be obtained. The quasiparticle gap feature has essentially disappeared and a 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 this weak depression is tentatively identified as the pseudogap [20]; however, we have not taken data up to much higher temperatures to verify this. The indications are that for underdoped crystals the superconducting gap has evolved into a pseudogap above T_c.

In summary, all of the experimental data, including the spectral shapes, the Josephson I_cR_n products, and

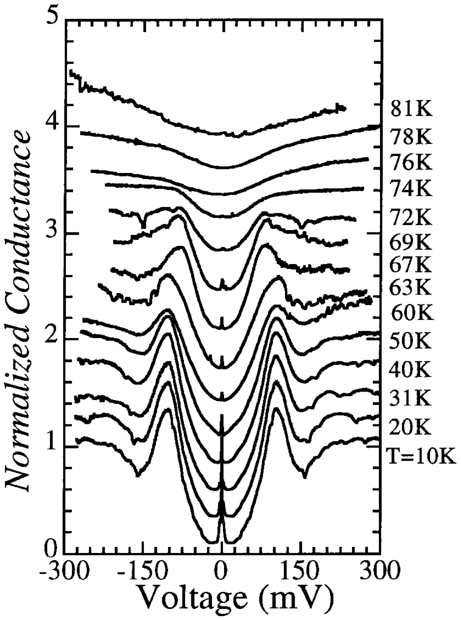

FIG. 3. Temperature dependence of SIS tunneling conductance on an underdoped Bi2212 ($T_c = 77$ K) break junction. For clarity, each conductance has been normalized by its value at 200 mV and (except for the 10 K curve) is offset vertically.

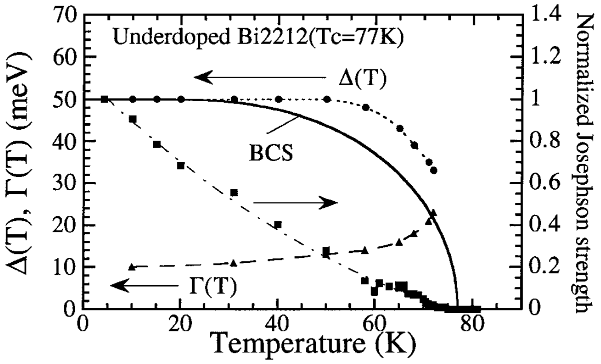

FIG. 4. Temperature dependence of superconducting gap $\Delta(T)$ (circle), quasiparticle scattering rate $\Gamma(T)$ (triangle), and Josephson strength, $I_c^* R_n$ (square) normalized by $I_c^* R_n$ (4.2 K). Here, the Josephson current I_c^* is estimated from the peak in conductance at zero bias. The full curve represents the BCS superconducting gap $\Delta(T)$.

the T dependence of the gap, indicate that, despite the anomalously large magnitude of Δ, it nevertheless appears to be due to superconductivity. Since the doping dependence of Δ and T^* follow the same trends [3] in Bi2212, this suggests that the pseudogap state between T_c and T^* is characterized by some form of precursor superconductivity and that T_c is the temperature where long-range phase coherence sets in [1,4,5]. However, the rapid drop of Δ near T_c argues more toward a picture of superconducting fluctuations [5] and not tightly bound preformed pairs as discussed in Ref. [1]. The results of this study are in direct contradiction with experiments which imply that the superconducting gap follows T_c [7–9]. One explanation is that the other experiments are sensitive to the nodal regions of the d-wave gap [7] along the (π, π) momentum direction, and either this gives an inaccurate value of the maximum gap or it is possible that this region indeed scales with T_c. The conductance peaks in our tunneling data are revealing the maximum superconducting gap near the $(\pi, 0)$ region of the Brillouin zone [2,15], and this gap follows T^*.

Focusing on the 70 K underdoped crystal in Fig. 2, the SIS dip feature is at ~150 meV. This is a very high energy scale compared to kT_c but one which is nevertheless associated with superconductivity. This issue has been addressed recently by Rubhausen *et al.* [21] with regard to changes in Raman spectra in Bi2212 below T_c which occur at frequencies which are much higher than expected based on a mean-field estimate of the gap. Our results emphasize that T_c is a phase coherence temperature and is a poor indicator of the true superconducting energy scale in the underdoped region. The hump feature, which appears to be a unique spectral entity, will be the focus of a future report. Here, we note that the energy of this feature and its doping dependence are identical to that found in angle-resolved photoemission spectroscopy (ARPES) measurements [22]. Preliminary indications are that the momentum dispersion of this hump as measured in ARPES are consistent with it being associated with an incipient spin density wave gap [12,22]. If so, then tunneling data are indeed revealing a second, distinct gap, but at an energy scale 3 times higher than the superconducting gap.

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