

Correlation of Tunneling Spectra in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ with the Resonance Spin Excitation

J. F. Zasadzinski,^{1,2} L. Ozyuzer,^{2,3} N. Miyakawa,⁴ K. E. Gray,² D. G. Hinks,² and C. Kendziora⁵

¹Physics Division, Illinois Institute of Technology, Chicago, Illinois 60616

²Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439

³Department of Physics, Izmir Institute of Technology, TR-35437 Izmir, Turkey

⁴Department of Applied Physics, Science University of Tokyo, Tokyo, Japan

⁵Naval Research Laboratory, Washington, D.C. 20375

(Received 16 October 2000; published 23 July 2001)

New break-junction tunneling data are reported in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ over a wide range of hole concentration from underdoped ($T_c = 74$ K) to optimal doped ($T_c = 95$ K) to overdoped ($T_c = 48$ K). The conductances exhibit sharp dips at a voltage, Ω/e , measured with respect to the superconducting gap. Clear trends are found such that the dip strength is maximum at optimal doping and that Ω scales as $4.9kT_c$ over the entire doping range. These features link the dip to the resonance spin excitation and suggest quasiparticle interactions with this mode are important for superconductivity.

DOI: 10.1103/PhysRevLett.87.067005

PACS numbers: 74.50.+r, 74.25.Dw, 74.62.Dh, 74.72.Hs

Acceptance of the phonon mechanism for electron pairing in conventional superconductors began with the isotope effect, showing T_c scaling with a characteristic phonon energy, Ω_{ph} , and culminated with detailed, quantitative agreement between features in tunneling spectroscopy and the phonon spectrum measured by neutron scattering [1]. For high T_c cuprates, however, tunneling and other spectroscopies have not led to a consensus on the pairing mechanism and, in fact, are currently being interpreted within radically different theoretical frameworks. The doping dependence of spectral features can be a key in clarifying their origin. Thus we present superconductor-insulator-superconductor (SIS) tunneling data on $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ over a very wide doping range from underdoped ($T_c = 74$ K) to optimal doped ($T_c = 95$ K) to overdoped ($T_c = 48$ K). From zero bias up to the superconducting gap voltage, $2\Delta/e$, the measured conductances are close to that expected from the density of states (DOS) found in mean-field models of a d -wave order parameter. However, the conductances also reveal sharp dips at a voltage, Ω/e , beyond the gap edge. These dip features are similar to structures ascribed to phonons in conventional superconductors and suggest that electrons are coupled to some type of collective excitation of energy $\sim\Omega$, measurable by the dip minimum. Most importantly, it is found that Ω scales as $4.9kT_c$ over the entire doping range which is close to that of the resonance spin excitation energy, Ω_{res} , found in neutron scattering [2,3]. Thus the tunneling and neutron measurements together present a strong case that spin excitations play an important role in the pairing mechanism of high T_c superconductors.

An unusual spectral dip feature in the tunneling data of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ ($\text{Bi}2212$) was pointed out as early as 1989 in superconductor-insulator-normal metal (SIN) junctions [4] where the dynamic conductance, $\sigma(V)$, is expected to be proportional to the electronic DOS, $N(E)$, with $E = eV$. This feature was repeatedly observed in many subsequent tunneling studies of $\text{Bi}2212$, e.g.,

scanning tunneling spectroscopy (STS) [5,6], break junctions [5,7], and recently in intrinsic c -axis junctions [8] of $\text{Bi}2212$ crystals intercalated with HgBr_2 . These consistent observations (both magnitude and location) of the dip in such a variety of junction types tend to rule out extrinsic, surface related phenomena as being responsible. An STM study of $\text{Bi}2212$ showed that the dip strength in normalized data was nearly symmetric in bias voltage and represented a suppression of the density of states [5]. It was therefore argued to be some form of strong coupling effect analogous to the dip features from the electron-phonon interaction in conventional superconductors [5]. In conjunction with other tunneling measurements [5,9,10] on $\text{Bi}2212$, it was suggested that the dip voltage scaled with the maximum d -wave gap, Δ , (e.g., $eV_{\text{dip}} \sim 3\Delta$ in SIS junctions). However, we will demonstrate here that the sharp dips observed in these new SIS junctions reveal a trend with doping whereby the minimum is at $2\Delta + \Omega$, and Ω is proportional to T_c .

The linking of the tunneling dip feature to the resonance spin excitation has been made possible due to the confluence of a number of experimental and theoretical developments. Neutron scattering has shown that the resonance mode is generic to bilayer high T_c cuprates reaching a maximum energy of 41–43 meV at optimal doping and tracking T_c with underdoping [3] or slight overdoping [2]. Other spectroscopies including angle resolved photoemission (ARPES) [11,12] and optical conductivity [13] have exhibited spectral features that can be explained by assuming the conduction electrons are coupled to the resonance mode. Theoretical spin-fermion-type models [11,14] which have been invoked to explain the ARPES data have also been shown to produce dip features in the quasiparticle density of states and SIS spectra [15,16] that resemble those reported here. Unique to this tunneling work, however, is that we have measured the relations between Ω , T_c , and Δ where the latter varies by a factor of 6 over this doping range. Such a

systematic study allows trends to be observed and thereby makes a more convincing case that the tunneling dip is due to the resonance mode and not some other excitations such as phonons. In addition, we demonstrate that the tunneling Ω is always less than 2Δ which provides the first experimental evidence that the resonance mode is a magnetic exciton. This is discussed later.

The break junctions were obtained by a point contact technique (described elsewhere [5,10]) on Bi2212 crystals oxygen doped over a wide range of hole concentration. We stress that these break junctions are formed under high vacuum, cryogenic conditions and they occur deep in the Bi2212 crystal [17], thereby minimizing surface contamination. In Fig. 1 the dynamic conductance spectra at 4.2 K are shown for three SIS break junctions on Bi2212: (a) an overdoped, (b) an optimally doped, and (c) an underdoped crystal. These SIS spectra are a compendium of a much larger data set and they capture the principal features of interest. The main conductance peaks reveal the energy gap at $|eV_p| = 2\Delta$, which increases in the underdoped region even as T_c decreases [10]. For $|eV|$ beyond 2Δ there is a pronounced dip feature that is strongest at optimal doping. The new optimal doped data of Fig. 1 are quite similar to those of Ref. [10], but exhibit a somewhat stronger dip with a negative conductance at the minimum. For SIS

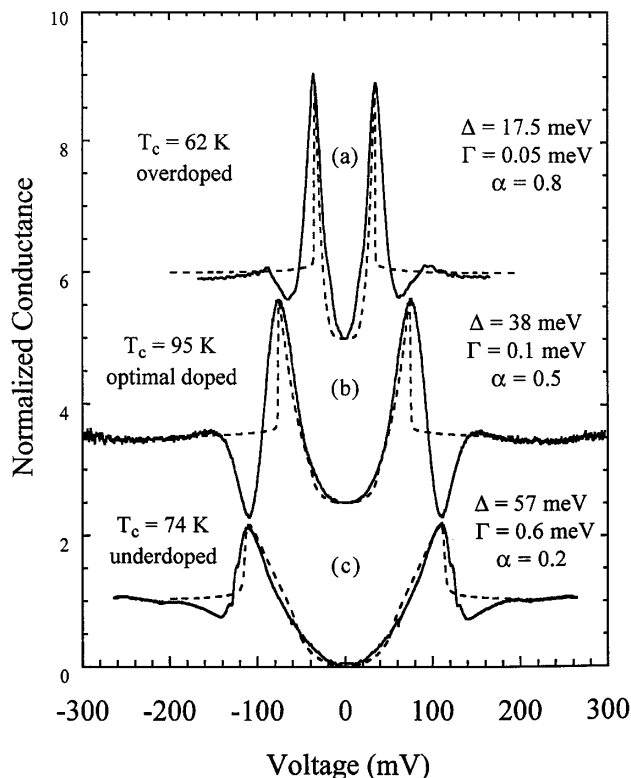


FIG. 1. SIS tunneling conductances for (a) overdoped, (b) optimally doped, and (c) underdoped Bi2212. Data have been normalized by a smooth background and shifted for clarity. Dashed lines are BCS d -wave fits using $\Delta(\phi) = \Delta \cos 2\phi$, a scattering rate, Γ , and a weighting function, $f(\phi) = 1 + \alpha \cos 4\phi$ as described in Ref. [17]. The zero of conductance for each curve is given by the measured and fitted curves at zero bias.

junctions this negative dI/dV is not unphysical but merely reflects a strong dip in $N(E)$ (see, for example, [15]). The spectra are compared with a weak coupling, d -wave fit [17] which includes a quasiparticle scattering rate, Γ . Considering the simplicity of the BCS, d -wave model, the agreement in the subgap region (up to $eV = 2\Delta$) is surprisingly good. This is also found for SIN junctions [17], where the d -wave DOS provides a very good fit in the subgap region, capturing the cusp feature measured at zero bias.

For $|eV| > 2\Delta$ there is an immediate positive deviation from the fit, followed by the strong negative deviation (dip) and finally a recovery toward a hump feature. Broadening the peaks in the d -wave model by increasing the scattering rate, Γ , leads to severe reduction of the peak heights which is clearly incompatible with the data. The excess width of the experimental conductance peaks appears to be intrinsic, reflecting a pileup of states which compensates for the depletion at the dip. We emphasize that the identical behavior is found throughout the doping range and that these deviations from the d -wave fit resemble the strong coupling effects from the electron-phonon interaction in conventional superconductors [1]. This suggests that the dip features are due to some type of bosonic collective excitation (or a relatively narrow spectrum of excitations).

A larger set of spectra over the entire range of doping is plotted in Fig. 2 on a voltage axis which is scaled by $\Delta = eV_p/2$. In addition to the dip strength being maximum at the highest T_c , another trend is evident. On this plot it can be seen that while the dip minimum is very close to 3Δ at optimal doping (as noted in previous work [9,10]) it is significantly greater than (less than) 3Δ with overdoping (underdoping). While a more subtle version of these trends can be found in an earlier paper [10], they are much easier to see here because the newer SIS spectra all have a peak height to background ratio >2 and exhibit sharper dip features. Both of these trends clearly link the dip to the superconductivity, but rule out trivial connections such as proximity effects [18]. Also, the shift of Ω inferred from the dip minima in Fig. 2 is consistent with theoretical predictions [14] for the resonance mode energy Ω_{res} within the gap 2Δ in the spin excitation spectrum. Thus Figs. 1 and 2 make a strong case that (1) the dip feature arises from quasiparticles coupled to some type of collective excitation of energy, Ω , and (2) the doping dependence of Ω bears a qualitative resemblance to the resonance spin excitation in experiment [2,3] and theory [14].

So far we have implied that the dip minimum might provide a quantitative measure of Ω , but some justification for this is required, especially in the absence of a full microscopic theory. We first note that SIN data, which should directly reflect $N(E)$, show reproducibly that the dip minimum is about 35–40 meV beyond the gap edge in optimal doped Bi2212 [5,6,10], close to the resonance mode energy. Considering conventional phonon structures in s -wave superconductors [1], phonon modes with energy Ω_{ph} would produce tunneling dip features near $eV = 2\Delta_s + \Omega_{\text{ph}}$ in SIS junctions where Δ_s is the

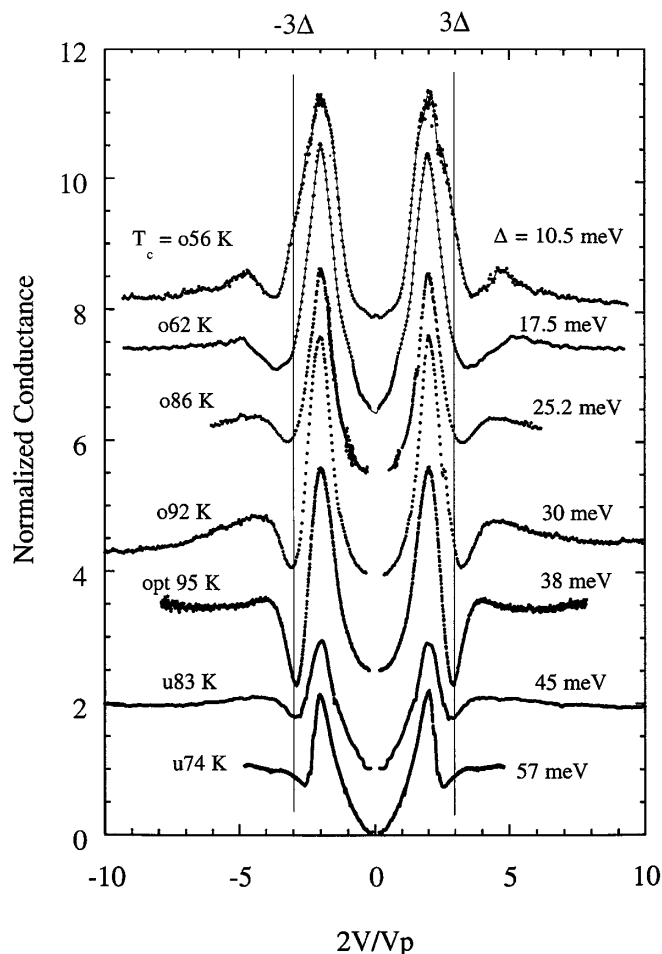


FIG. 2. SIS tunneling conductances of Bi2212 for various hole dopings. Notation is o = overdoped, opt = optimal doped, and u = underdoped. Voltage axis has been rescaled in units of Δ . Each curve has been rescaled and shifted for clarity. The Josephson current has been removed from each curve, and the inferred conductance at zero bias for each curve is close to zero.

s-wave superconducting gap, but the dip minima would overestimate the mode energy. However the cuprates are *d*-wave superconductors, and the presence of gap nodes can affect the location of strong coupling features. This result comes from the analysis of ARPES data in Bi2212 [11,12] which show a similar dip/hump feature for electrons near the $(\pi, 0)$ point, i.e., maximum gap region. The ARPES data can be analyzed within a model whereby the electrons near $(\pi, 0)$ are interacting with a collective mode [11]. This model has been extended to calculations of the SIS tunneling spectrum [15] by considering a *d*-wave gap, and the result is that the dip minimum is very close to $2\Delta + \Omega$. It has also been demonstrated using Eliashberg formalism for *d*-wave superconductors that the SIS tunneling dip minimum provides a good estimate for the energy of a generic, single-peak boson spectrum [19].

Considering the above discussion, it seems reasonable to extract Ω by assuming the dip minimum is at $2\Delta + \Omega$. In Fig. 3 are plotted the *measured* values of Ω and T_c vs doping obtained on 17 SIS junctions over the full dop-

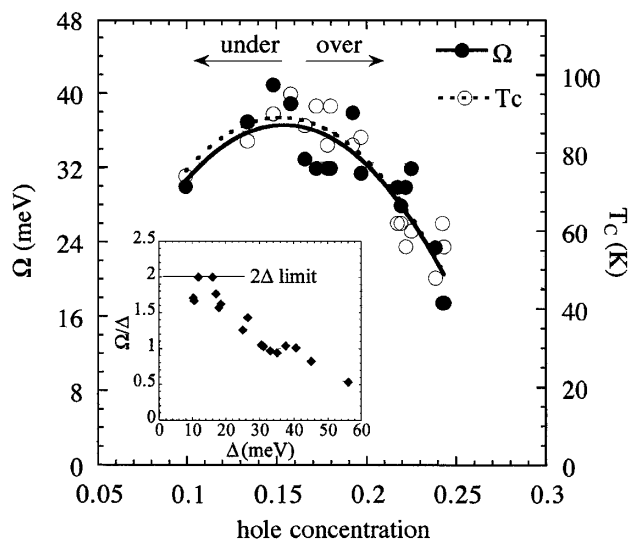


FIG. 3. Measured resonance mode energy Ω and bulk T_c value vs hole doping level obtained from 17 junctions over a wide doping range from u 74 K to o 48 K. Solid and dashed lines are quadratic fits of Ω and T_c vs doping. Inset shows Ω/Δ vs the measured Δ .

ing range. The doping level is obtained from the measured gap, Δ , which is nearly linear in hole concentration [10]. It is found that the maximum measured value of $\Omega \sim 42$ meV occurs near optimal doping and agrees with the maximum resonance mode energy obtained in neutron scattering [2,3]. The scatter in the data of Fig. 3 arises mainly from a general uncertainty in determining the mode energy, Ω , directly from the dip minimum, and also from the narrow spread of gap values obtained from different junctions on the same crystal (with the same bulk T_c). Despite the scatter in the data there is an unmistakable correlation between Ω and T_c . This is seen with simple quadratic fits to Ω and T_c vs doping (solid and dashed lines, respectively, in Fig. 3) which are nearly congruent and lead to the relation $\Omega/kT_c \sim 4.9$ which we note is in good quantitative agreement with neutron results [2,3] for $\Omega_{\text{res}}/kT_c \sim 5.1-5.5$. This is reminiscent of the conventional isotope effect in superconductors where T_c scales with Ω_{ph} . We note that the tunneling Ω scales with T_c far into the overdoped region, well beyond what has been measured so far for the neutron resonance.

In the plot of Ω/Δ vs Δ in the inset of Fig. 3 the relation of Ω to Δ is more clearly seen, providing important information on the nature of the excitation probed by tunneling. For the most overdoped region, the mode energy approaches but never exceeds 2Δ , and Ω/Δ monotonically decreases as doping decreases and the superconducting gap grows [20]. Thus the excitation exhibits excitonic character, always lying within the superconducting gap, 2Δ , which varies by a factor of 6. This argues against phonons and instead suggests an electronic excitation. The behavior is consistent with general ideas about collective modes of the conduction electrons in superconductors [21] (i.e., that mode energies $>2\Delta$ are heavily damped), as well as

with specific models of the resonance spin excitation [14]. In the latter, the overdoped region is considered to have weaker coupling of electrons to spin excitations and therefore the resonance mode is close to the gap edge, 2Δ , in the spin excitation spectrum. As doping decreases and the coupling gets stronger, the superconducting gap gets larger but the mode energy moves deeper into the gap and thus Ω/Δ decreases. In this picture the neutron resonance is a magnetic (or spin) exciton [14,15], and we believe the tunneling data provide the first clearcut evidence for such a viewpoint [22].

To summarize, the doping dependence of the tunneling dip feature in Bi2212 has revealed trends which link it to the resonance spin excitation. Other interpretations of the dip which suggest it is a background effect (e.g., the van Hove singularity) are difficult to reconcile with the various properties outlined here (and also the T dependence [8,10]) which clearly point toward it being a superconducting property. Since the dip resembles a strong coupling effect, the tunneling data also provide evidence that spin excitations are playing a crucial role in the superconductivity. It should be noted that a similar dip feature has been observed in the superconducting tunneling spectra of a heavy fermion superconductor [23] which has also been linked to a peak that develops in the spin excitation spectrum. Thus a spin fluctuation mechanism may have a more general relevance to superconductors beyond the high T_c cuprates. It is important to contrast this interaction with phonon mediated pairing. Phonons are collective excitations of the lattice and therefore exist in the normal state, whereas this collective mode seems to develop only below T_c (except for heavily underdoped materials). Thus this scenario displays a remarkable feedback effect: Superconductivity hollows out a gap in the spectrum of damped spin excitations, allowing a propagating collective mode to exist, but then that mode is at least partly responsible for the pairing mechanism. If, in addition, there are phonon contributions to the pairing, the phonon structures are not showing up in a clear and reproducible manner.

The selective coupling of electrons near $(\pi, 0)$ to spin excitations near (π, π) as suggested in ARPES [11,12,14,15] also explains the mysterious asymmetry of the dip strength with voltage polarity commonly observed in SIN tunneling [4–6]. In this picture, the quasiparticles at $(\pi, 0)$ are the most strongly coupled, and since this is an occupied state in Bi2212 [11,12] the strongest dip features should be found for bias voltages which remove electrons, as is observed in SIN experiments and model calculations [15]. A proof of the mechanism suggested here would require that the tunneling spectra quantitatively reproduce the measured Δ and T_c , as found with phonon structures [1]. This will require a full microscopic model, which does not exist at present. As a final comment, we note that similar dip features have been observed in the tunneling spectra of $Tl_2Ba_2CuO_6$ indicating that the

neutron resonance ought to be observed in a cuprate with a single Cu-O layer per unit cell [24].

The authors benefited considerably from discussions with A. Chubukov, B. Janko, M. Norman, and L. Coffey. This work was partially supported by the U.S.-DOE, BES-MS under Contract No. W-31-109-ENG-38 (K.E.G., D.G.H., J.F.Z.) and by a Grant-in-aid for Encouragement of Young Scientists from the Ministry of Education, Science and Culture, Japan (N.M.).

-
- [1] E. L. Wolf, *Principals of Electron Tunneling Spectroscopy* (Oxford University Press, New York, 1985), Chaps. 2–5.
 - [2] H. F. Fong, B. Keimer, D. L. Milius, and I. A. Aksay, *Phys. Rev. Lett.* **78**, 713 (1997); H. He *et al.*, cond-mat/0002013.
 - [3] Pengcheng Dai *et al.*, *Science* **284**, 1344 (1999).
 - [4] Q. Huang *et al.*, *Phys. Rev. B* **40**, 9366 (1989).
 - [5] Y. DeWilde *et al.*, *Phys. Rev. Lett.* **80**, 153 (1998).
 - [6] Ch. Renner and O. Fischer, *Phys. Rev. B* **51**, 9208 (1995).
 - [7] D. Mandrus *et al.*, *Nature (London)* **351**, 460 (1991).
 - [8] A. Yurgens *et al.*, *Int. J. Mod. Phys. B* **29–31**, 3758 (1999).
 - [9] J. F. Zasadzinski *et al.*, *J. Phys. Chem. Solids* **53**, 1635 (1992); D. Coffey and L. Coffey, *Phys. Rev. Lett.* **70**, 1529 (1993).
 - [10] N. Miyakawa *et al.*, *Phys. Rev. Lett.* **83**, 1018 (1999).
 - [11] J. C. Campuzano *et al.*, *Phys. Rev. Lett.* **83**, 3709 (1999); M. R. Norman and H. Ding, *Phys. Rev. B* **57**, 11089 (1998).
 - [12] Z. X. Shen and J. R. Schrieffer, *Phys. Rev. Lett.* **78**, 1771 (1997).
 - [13] J. P. Carbotte *et al.*, *Nature (London)* **401**, 354 (1999).
 - [14] Ar. Abanov and Andrey V. Chubukov, *Phys. Rev. Lett.* **83**, 1652 (1999).
 - [15] M. Eschrig and M. R. Norman, *Phys. Rev. Lett.* **85**, 3261 (2000).
 - [16] Ar. Abanov and Andrey V. Chubukov, *Phys. Rev. B* **61**, R9241 (2000).
 - [17] L. Ozyuzer *et al.*, *Phys. Rev. B* **61**, 3629 (2000).
 - [18] Proximity effect models (as described in Ref. [1]) cannot account for the trends observed. Furthermore they ascribe the dip/hump feature to the bulk superconducting gap which overestimates the gap by a factor of 2 or 3.
 - [19] L. Coffey, cond-mat/0103518.
 - [20] A similar shift of the tunneling dip with overdoping was seen in STM data in Bi2212. See Ch. Renner *et al.*, *J. Low Temp. Phys.* **105**, 1083 (1996).
 - [21] Paul C. Martin, in *Superconductivity*, edited by R. D. Parks (Marcel Dekker Inc., New York, 1961).
 - [22] These arguments hinge on our interpretation that the measured gap, Δ , is due solely to superconductivity, a point argued in Ref. [10], but see, for example, the Comment by R. S. Markiewicz and C. Kusko, *Phys. Rev. Lett.* **84**, 5674 (2000), and Reply by J. F. Zasadzinski and N. Miyakawa, *Phys. Rev. Lett.* **84**, 5675 (2000).
 - [23] M. Jourdan and A. H. Huth, *Nature (London)* **398**, 47 (1999).
 - [24] J. F. Zasadzinski *et al.*, *Physica (Amsterdam)* **341C–348C**, 867 (2000).