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5-2006

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### Recommended Citation

Valla, T.; Kidd, T. E.; Rameau, J. D.; Noh, H.-J.; Gu, G. D.; Johnson, P. D.; Yang, H.-B.; and Ding, H., "Fine Details of the Nodal Electronic Excitations in Bi2Sr2CaCusO8+delta" (2006). Faculty Publications. 13. [https://scholarworks.uni.edu/phy\\_facpub/13](https://scholarworks.uni.edu/phy_facpub/13?utm_source=scholarworks.uni.edu%2Fphy_facpub%2F13&utm_medium=PDF&utm_campaign=PDFCoverPages) 

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### Fine details of the nodal electronic excitations in  $Bi_2Sr_2CaCu_2O_{8+\delta}$

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(Received 28 December 2005; published 17 May 2006)

Very high energy resolution photoemission experiments on high quality samples of optimally doped  $Bi_2Sr_2CaCu_2O_{8+\delta}$  show new features in the low-energy electronic excitations. A marked change in the binding energy and temperature dependence of the near-nodal scattering rates is observed near the superconducting transition temperature, *TC*. The temperature slope of the scattering rate measured at low energy shows a discontinuity at *TC*. In the superconducting state, coherent excitations are found with the scattering rates showing a cubic dependence on frequency and temperature. The superconducting gap has a *d*-wave magnitude with negligible contribution from higher harmonics. Further, the bilayer splitting has been found to be finite at the nodal point.

DOI: [10.1103/PhysRevB.73.184518](http://dx.doi.org/10.1103/PhysRevB.73.184518)

PACS number(s): 74.25.Jb, 74.72.Hs, 79.60.Bm

High temperature superconductivity continues to present some of the biggest challenges in materials science today. What is the nature of the low energy excitations and what is the pairing mechanism that leads to the superconductivity? In attempting to answer these questions, angle-resolved photoemission spectroscopy (ARPES), with high resolution in both energy and momentum, has emerged as one of the leading techniques for the study of strongly correlated materials, including the high  $T_c$  superconductors. Indeed, the demonstration that the *k*-dependence of the amplitude of the superconducting gap in these materials is consistent with *d*-wave symmetry represents an important contribution of the technique to the field.1 More recently, it has been shown that ARPES is an excellent probe of the single particle selfenergy a quantity that reflects the quasiparticle (QP) interactions within the system and manifests itself in a renormalization of the single-particle dispersion and structure in the spectral width. $2,3$  The discovery of the mass renormalization or "kink" in the dispersion in  $Bi_2Sr_2CaCu_2O_{8+\delta}$  has lead to renewed speculation about the origin of high temperature superconductivity and the possibility that the observed renormalization reflects coupling to some boson involved in the pairing.4 Indeed, the "kink" has become one of the central issues in subsequent ARPES work, with considerable controversy regarding its source.<sup>1,5–8</sup> Is it related to the presence of spin excitations or does it reflect an interaction with phonons or indeed any other collective mode? Note that this is not an easy issue to resolve as the various energy scales are nearly identical. Our earlier studies of the doping and momentum dependence pointed to spin fluctuations, $\frac{7}{1}$  while some papers favor phonons<sup>1,6</sup> as the source of mass enhancement.

A second important question that remains open is the detailed *k*-dependence of the superconducting gap  $\Delta(\mathbf{k})$ . Although the momentum dependence of the gap amplitude was one of the first ARPES contributions to the field, $<sup>1</sup>$  the</sup> *k*-dependence from the near-nodal region has not previously been measured with sufficient precision. Early work showed that the contribution of higher *d*-wave harmonics grew as the doping was reduced.9 Some recent papers suggest a significant contribution of higher harmonics in the cos  $k_x$ -cos  $k_y$ distribution, even for samples relatively close to optimal doping.10 However, low-temperature thermal transport measurements suggest that the near nodal gap scales with the maximal gap  $\Delta_0$  as measured in ARPES<sup>1</sup> or tunnelling,<sup>11</sup> even in severely underdoped samples.12 A detailed *k*-dependence of the gap is crucial as it determines the range of pairing interactions in real space. Further if the gap is a superposition of two competing orders, the *k*-dependence of the gap could provide insights into the relationship between them.

Finally, even though it is generally believed that the normal state of the cuprate superconductors is not a Fermi liquid, the appearance of well-defined QPs in the superconducting state has been well documented in other experimental techniques such as thermal conductivity, $12$  microwave, $13$  and scanning tunnelling spectroscopy.<sup>14</sup> However, there are conflicting ARPES results on the existence of nodal QPs.<sup>4,15</sup> In particular, one of our earlier studies reported that the spectral width in the nodal region was almost completely insensitive to the superconducting transition.<sup>4</sup> Theoretical studies indicate that in the superconducting state, phase space restrictions should result in a cubic dependence on *T* and  $\omega$ .<sup>16</sup>

In the present letter, we focus on these issues again and show that the sample quality and experimental resolution are crucial factors in resolving "fine details" of the electronic structure near the nodal point. We show that with sufficient resolution and high-quality samples, the ARPES experiments detect a marked change in the nature of the near-nodal excitation at the superconducting transition. Coherent states are found in the superconducting state, with the QP width being less than its energy. Below  $T_c$  the scattering rate displays a cubic dependence on *T* and  $\omega$  up to  $\sim$  75 meV. In the normal state, the scattering rate varies as  $\omega$  for  $\omega > T$ , and as *T*, for  $T \leq \omega$ . Further, we have found that the gap function near the node may be correlated with the amount of elastic scattering. In the highest quality samples the superconducting gap amplitude has a typical "V" profile, but with increasing elastic scattering the near-nodal gap becomes reduced and convex.



FIG. 1. (Color online) (a) High-resolution photoemission spectrum of optimally doped  $Bi_2Sr_2CaCu_2O_{8+\delta}$  at 8 K, along the dashed line in the Brillouin zone, as indicated in (c). (b) MDCs at  $\omega=0$  (circles) and at  $\omega=-10$  meV (triangles). (c) Fermi surface reconstructed from a series of spectra along momentum lines parallel to the dashed line. (d) EDC at a fixed momentum  $(k-k_F=-0.015 \text{ Å}^{-1})$ . The solid line shows a Lorentzian fit to the near-peak and the high-energy side of the bonding state peak.

We have also found a finite bilayer splitting at the nodal point.

The experiments were carried out on a Scienta SES-2002 electron spectrometer at beam line U13UB of the National Synchrotron Light Source. The combined instrumental energy resolution was set either to  $\sim$  5 meV or to 10 meV as indicated in the text. The angular resolution was better than  $\pm 0.003$  Å<sup>-1</sup> at the 16.7 eV photon energy used in the present study. Optimally doped samples  $(T_C=91 \text{ K})$ , grown by the travelling solvent floating zone method, were mounted on a liquid He cryostat and cleaved *in situ* in the UHV chamber with a base pressure  $6\times10^{-9}$  Pa. The temperature was measured using a calibrated silicon sensor mounted near the sample. The photoemission spectra were analyzed using both energy distribution curves (EDC) and momentum distribution curves (MDC).

Figure 1 shows the photoemission spectrum recorded with 5 meV resolution in the superconducting state along the momentum line as indicated in the inset. A bilayer splitting that is finite and measurable is obvious in both the MDCs and EDCs. Several lines were scanned in momentum space parallel to this line  $(30^{\circ}$  relative to  $\Gamma$ Y) allowing the Fermi surface in the near-nodal region to be reconstructed. Note that the bonding state dominates, while the antibonding state results in a less bright shoulder in the contour plot. At the nodal point the splitting is visible only with the energy resolution of 10 meV or better. Measured with 5 meV resolution, we determine by fitting with two Lorentzian peaks that the width of the bonding component is  $\Delta k_B \approx 0.009 \text{ Å}^{-1}$  and the splitting between the two components is  $k_B - k_A \approx 0.008 \text{ Å}^{-1}$ 



FIG. 2. (Color online) (a) Momentum width of the Fermi surface as a function of angle  $\phi$  for two optimally doped samples (A and **B**). The momentum splitting between bonding and antibonding states for the sample with sharper states (A) is also plotted. Components normal to the Fermi surface are shown. (b) Superconducting gap for samples **A**, **B**.

at the nodal point. These values represent the components perpendicular to the Fermi surface. Similar values for the widths and splitting have been reported elsewhere.<sup>18</sup>

In Fig.  $2(a)$  we show the *k*-dependences of momentum width of the bonding state at the Fermi surface and of the momentum splitting between the bonding and antibonding states. Again, the components perpendicular to the Fermi surface are shown, this time as measured with 10 meV resolution. As anticipated, the splitting between the states becomes larger as one moves away from the nodal point. As an indication of the quality of the present sample, **A**, we also show the near nodal width from another optimally doped sample, **B**, measured at the same temperature and with the same experimental resolution. The larger widths reflect a higher level of impurity or defect scattering. In Fig. 2(b) we show the amplitudes of the superconducting gaps,  $\Delta(\mathbf{k})$ , as measured using the leading edge of a spectrum integrated over a finite momentum range, typically  $\pm 0.05$  Å<sup>-1</sup> around  $k_F$  along the measured momentum lines. We have also used different methods of extracting the gap amplitude, $19$  resulting in the same *k*-dependence. The gap function of the sample with sharp nodal states (A) clearly has a V-like profile, while the sample with broader states (B) shows a convex gap function (flatter gap) near the node. The function  $\Delta_0[B\cos(2\phi)]$ +(1−*B*)cos(6 $\phi$ )] has been used to describe the anisotropy of the gap with  $\Delta_0$  the maximum gap amplitude.<sup>9</sup> The V-shape is characteristic of a pure  $cos(2\phi)$  with little or no contribution from higher harmonics and points to the dominance of nearest neighbor interactions in the pairing in sample **A**. However, with an increasing contribution from impurity scattering, the gap amplitude near the node gets reduced, states become broader, and any fine detail of the type discussed below becomes unresolved.

Figure  $3(a)$  shows the effects of the superconducting transition on the scattering rates for sample **A**. A normal state spectrum was recorded with 10 meV resolution and fitted with a single Lorentzian line shape.<sup>20</sup> Two spectra in the



FIG. 3. (Color online) (a) Width of the MDC peak in the nodal region in the normal and superconducting state recorded with 5 meV and 10 meV energy resolution, as indicated. (b) The 5 meV resolution data from (a), fitted with a cubic fit (solid line). (c) EDC width as a function of binding energy at 8 K. The straight line indicates the "QP" boundary,  $\Gamma(\omega) = |\omega|$ .

superconducting state were recorded consecutively with 10 meV and 5 meV resolution and fitted with a single Lorentzian peak and with two Lorentzian peaks, respectively. It is clear that most of the change between the normal and superconducting states occurs in the low binding energy range, below  $\sim$  75 meV. If taken with high resolution, the superconducting spectrum shows a smooth variation of the MDC width in that region, with no indication of any prominent structure up to  $\sim$ 75 meV, where the functional form changes into a nearly linear dependence. In the latter regime, the scattering rate is almost unaffected by the superconducting transition.<sup>21</sup> The low energy region recorded with high resolution may be fitted with an  $\omega^3$  dependence [Fig. 3(b)], characteristic of electron-electron scattering in the presence of a *d*-wave gap.16 It is clear that the fit does not catch the lower frequencies accurately. In this region a linear dependence seems more appropriate. The total scattering rate or inverse lifetime reflects both elastic and inelastic contributions, the former from impurities, and the latter from electronic and bosonic scattering processes. As such, the finite scattering rate at the lowest temperatures at  $\omega = 0$  reflects the elastic scattering from impurities or defects. Furthermore it has been suggested elsewhere<sup>16,17,22</sup> that the lowest frequency region is dominated by the quasilinear dependence of weak elastic scattering from out-of-plane impurities, consistent with our observations.

We also show in Fig.  $3(c)$  the width of the lower (bonding) state as determined from EDCs recorded with 5 meV resolution and fitted with two Lorentzian peaks. Fitting the EDCs with Lorentzians works well because  $\text{Im } \Sigma$  is a slowly varying function, and its value is small at low energies ( $|\omega|$ )  $\leq 50$  meV). As indicated in the figure, it is clear that over a finite range of energies near the Fermi level, the width of the state is less than its energy, in agreement with recent laser induced ARPES studies, $^{23}$  an indication that nodal QPs exist in the superconducting state. However, this clear evidence of QPs appears to be true only at the low temperatures. In the vicinity of  $T_c$ , the criterion that the width of the state is less



FIG. 4. (Color online) (a) The MDC-derived dispersion along the line indicated in Fig. 1(c) at different temperatures. (b) The corresponding MDC peak widths. (c) The *T*-dependence of MDC width at the Fermi level from two data sets indicated by open and solid circles. The solid line is a cubic fit to all data points below  $T_C$ .

than its energy is satisfied only marginally and over a very narrow range of energies even after subtracting the contribution from impurity scattering and energy resolution. This is markedly different from the observations on metallic systems such as our study of molybdenum.<sup>2</sup>

More detailed temperature dependent studies of the electron self-energies for the same momentum line (from Fig. 1) are shown in Fig. 4. In Fig. 4(a) the MDC deduced dispersions are shown for several temperatures, while Fig.  $4(b)$ shows the corresponding energy dependence of the momentum widths. The *T*-dependence of the width at the Fermi level derived from two overlapping data sets is shown in Fig. 4(c). For this analysis all the spectra were taken with 10 meV total resolution and the MDCs were fitted with a single Lorentzian.20 As such the widths at low *T* are larger than the equivalent width in Fig.  $3(b)$ . Figure  $3(a)$  and Figs. 4(b) and 4(c) allow us to compare the scattering rates more directly above and below  $T_c$ . It is clear that the opening of the *d*-wave superconducting gap modifies the functional form of the nodal scattering rate, leading to the cubic dependence on binding energy at low frequencies as discussed above. Also, the MDC width at the Fermi level  $(\omega=0)$  as a function of *T* shows a discontinuity in slope at, or slightly above  $T_c$ . The discontinuity and a rapid narrowing below a certain temperature suggest the formation of coherent quasiparticles in the low-*T* state, consistent with our observations discussed earlier. Below  $T_c$  the spectral points in Fig. 4(c) can be fitted with a cubic dependence as predicted for QPs scattering off spin fluctuations in a superconducting state with  $d$ -wave gap.<sup>16,22</sup>

Elsewhere it has been suggested that the kink or mass renormalization observed in the nodal direction in the cuprates reflects coupling to some optical phonon mode.<sup>1,6</sup> Let us consider this suggestion. In the usual Migdal-Eliashberg approximation, the scattering rate or imaginary component of the self-energy is given by

Im 
$$
\Sigma(\omega) \propto \int_0^{\omega} d\nu \frac{N(\nu)}{N_0} \alpha^2 F(\omega - \nu),
$$
 (1)

where in the superconducting state with the order parameter having *d*-wave symmetry, the density of states (DOS) may

be approximated by  $N(\omega)/N_0 \approx \omega/\Delta_0$  at low frequencies. Here  $\Delta_0$  again represents the maximal superconducting gap energy and  $N_0$  represents the normal state DOS. Examination of Eq. (1) shows that if the phonon mode energy (corresponding to the kink energy) is greater than  $\Delta_0$ , the scattering rate as a function of binding energy at energies below the mode will be insensitive to the superconducting transition. This is clearly inconsistent with Figs.  $3(a)$  and  $4(a) - 4(c)$  and *essentially rules out the possibility that the kink reflects coupling to such a single Einstein mode*. The linear term and a finite zero-energy offset indicate, as already noted, that the lowest energy range is still dominated by impurity scattering. In that case, the low energy scattering rate would be affected by the transition. However, the change in the elastic scattering would be limited to energies inside the maximal superconducting gap  $\Delta_0$ , as it would reflect the redistribution of density of states upon transition. Here, the observation that there are changes at frequencies below the mode, but outside the gap energy  $\Delta_0$ , rules out the scattering from a single phonon mode. One simple explanation would be that the bosonic spectrum itself changes at transition. A redistribution of bosonic states at low energies, while conserving the total density (e.g., by opening of a gap at low energies) would strongly affect the electronic scattering rates at low energies, with minimal impact on those at high energies. We note in passing that any changes in the scattering rate or Im  $\Sigma$  for the nodal excitations must be accompanied by equivalent changes in Re  $\Sigma$  through causality. This is clearly visible in Fig. 4(a) where the changes in Re  $\Sigma$  affect the measured dispersions, in accordance with our previous study.7

In summary we have demonstrated a marked change in both the energy and temperature dependence of the scattering rates for the low energy excitations in the nodal region on entering the superconducting state. As we have reported before<sup>4</sup> and as found in optical conductivity studies<sup>24</sup> the states at higher binding energies show the same linear dependence on binding energy independent of whether the system is in the normal or superconducting state. This observation points to strong scattering in the electron-electron channel that reflects the entire band. The states at low frequencies have lifetimes that suggest they represent coherent Fermionic excitations. Further they appear sensitive to the presence of the superconducting gap and indeed the scattering rates have the anticipated cubic dependence on binding energy and temperature that reflect the restricted phase space in the *d*-wave gap. In addition, the observation that the scattering rate as a function of binding energy is modified on entering the superconducting state also provides evidence that the interaction responsible for this is electronic in origin. A high frequency phonon at  $\omega_0$  would not affect the relaxation rate until *T* or  $\omega$ exceeds  $\omega_0$ .

#### **ACKNOWLEDGMENTS**

The authors acknowledge useful discussions with Andrey Chubukov, Steve Kivelson, Maurice Rice, Doug Scalapino, John Tranquada, Alexei Tsvelik, and Ziqiang Wang. The research work described in this paper was supported by the Department of Energy under Contract No. DE-AC02- 98CH10886, and NSF Grant No. DMR-0353108.

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- $20$  Due to a large asymmetry in intensity of bilayer split states, the errors caused by fitting the doublet with a single Lorentzian peak are generally small and do not affect our conclusions.
- <sup>21</sup> In the linear range, the imaginary component of the self-energy, Im  $\Sigma = (\Delta k v_0)/2$ , varies as  $\sim 0.79\omega$ , if the bare velocity is set to  $\sim$ 3 eV/Å.
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