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Strong interaction between electrons and collective excitations in the multiband superconductor MgB_2

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We use a tunable laser angle-resolved photoemission spectroscopy to study the electronic properties of the prototypical multiband BCS superconductor MgB_2. Our data reveal a strong renormalization of the dispersion (kink) at ~65 meV, which is caused by the coupling of electrons to the E_{2g} phonon mode. In contrast to cuprates, the 65 meV kink in MgB_2 does not change significantly across T_c. More interestingly, we observe strong coupling to a second, lower energy collective mode at a binding energy of 10 meV. This excitation vanishes above T_c and is likely a signature of the elusive Leggett mode.

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In conventional superconductors, the pairing is mediated by phonons and favored by strong electron-phonon coupling, as described by Bardeen-Cooper-Schrieffer (BCS) theory [1,2]. This strong electron-phonon coupling in general gives rise to a renormalization of the band dispersion called a "kink" and an abrupt change of quasiparticle lifetime at an energy related to the phonon frequency Ω [2]. This idea has been extended to unconventional superconductors, where the mechanism of pairing is unknown, and the coupling of electrons to several collective excitations was reported [3–9]. Their origin and relation to pairing is still debated. For example the "70 meV" kink in the $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212) cuprate is strongest at the antinode and vanishes above T_c [8], a behavior that resembles the magnetic resonance mode reported by inelastic neutron scattering [10]; however, the electron-phonon interaction may have similar characteristics [11,12]. On the other hand, the kinks in the dispersion along the nodal direction in single layer $Bi_2Sr_2CuO_{(6+\delta)}$ (Bi2201) do not seem to change significantly with temperature [13]. Surprisingly, there is little data on dispersion renormalization effects in conventional superconductors; several low energy kinks have been reported in $NbSe_2$ [14], but the situation is complicated by the presence of a charge density wave phase coexisting with superconductivity, and the measurements were carried out only at low temperatures. In other materials, difficulties in clearly observing three-dimensional (3D) band dispersions and low transition temperatures are limiting factors. MgB_2 [15–20] is a notable exception: It is a layered material with multigapped, phonon-mediated superconductivity at $T_c =$ 39 K with some quasi-two-dimensional (2D) bands, making it an ideal candidate to study the temperature dependence of the dispersion renormalization due to electron-phonon coupling. Local density approximation (LDA) calculations [17] predict four bands crossing the Fermi level in MgB_2: two quasi-2D σ bands from the p_x, p_y orbitals around Γ and two 3D π bands from the p_z orbital [21–23]. The superconductivity is believed to be caused by the E_{2g} phonon mode at 75 meV that couples strongly to the 2D σ bands, but more weakly to the π bands [19], leading to two different gaps, $\Delta_\sigma = 6.5$ meV and $\Delta_\pi = 1.5$ meV, as revealed by previous tunneling [24] and angle-resolved photoemission spectroscopy (ARPES) studies [21]. Inelastic neutron scattering studies have reported optical phonon modes at ~35, 55, 75, 85, and 100 meV [25], while Raman scattering reports a single, broad asymmetric peak at 75 meV, attributed to the E_{2g} mode, as well as a sharp peak at $2\Delta_\sigma = 12$ meV due to a pair breaking excitation [26]. As a bonus, due to its multigap nature, MgB_2 also contains another exotic collective mode that can couple to the electrons: the Leggett mode [27–31]. This mode is a longitudinal fluctuation corresponding to equal and opposite displacements of the two condensates. As it is a neutral excitation, it is not pushed up to the plasma frequency like the Bogoliubov-Anderson mode, however, it is predicted to have a mass in between $2\Delta_\pi$ and $2\Delta_\sigma$ for MgB_2, where it will be partially damped by decaying into π quasiparticles [32]. As the superconductivity is believed to be phonon mediated and due to intraband pairing, the two gaps are expected to have the same relative sign, giving rise to s_{++} pairing, in contrast to the iron-based superconductors, where interband repulsive pairing likely leads to $s_\pm$ pairing and a Leggett mode is not expected.

High resolution band dispersion data demonstrating the coupling of the conduction electrons to collective excitations such as phonons or Leggett modes are not available in the published literature. One of the main reasons is that high quality single crystals need to be synthesized under high pressure, resulting in rather small $\leqslant 500$ μm crystals, which are difficult to measure in traditional ARPES setups. We use tunable laser ARPES to study the electronic properties of MgB_2 multiband superconductivity. The use of low photon energy increases significantly the bulk sensitivity due to an increased escape depth and momentum resolution due to the increased $Å^{-1}$/deg ratio. The ability to focus the laser beam down to ~30 μm enables the measurement of very small single crystals and also helps to improve the momentum resolution. We find evidence for the strong coupling of conduction electrons to a 75 meV acoustic phonon with λ estimated at ~1.3 that persists above T_c, unlike those in the cuprates. In fact, we observe no significant changes with temperature up to 65 K, more than 50% above T_c. Furthermore, we

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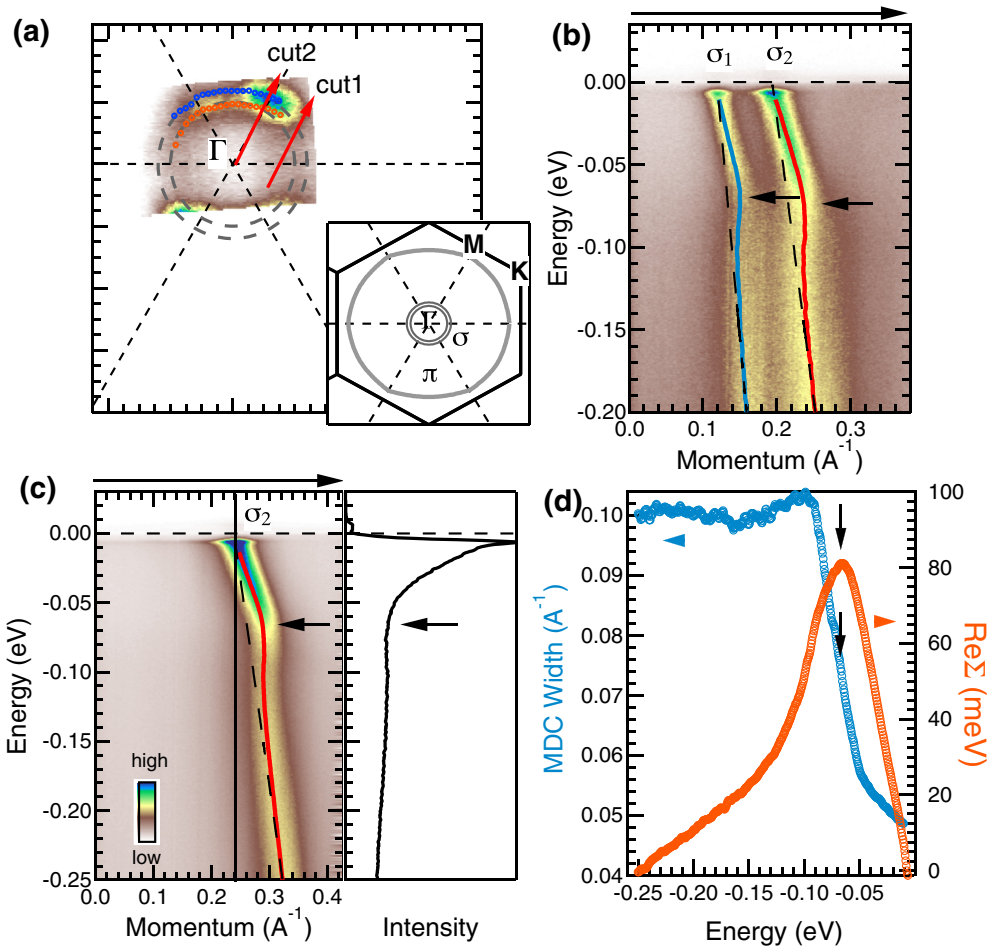


FIG. 1. (Color online) Renormalization effects (kink) in a σ band. (a) Measured FS at 40 K. Blue and red circles mark the Fermi momentum extracted from the MDC peak position at E_F. Two red arrows mark the locations of measured cuts in the momentum space. The inset is a schematic diagram of the FS and Brillouin zone. (b) ARPES intensity along cut No. 1 at 15 K. Solid lines are dispersions obtained by MDC fits for the two bands. Black dashed lines signify a bare band dispersion extrapolated from higher binding energies. (c) Left: Same as (b) but along cut No. 2; right: EDC at k_F for the outer σ band. (d) Energy dependence of the MDC width (blue solid line) and effective real part of self-energy (red dotted line) for data in (c). Arrows mark the energy location of the kink and associated features.

observe a very strong renormalization of the dispersion in the superconducting state at ~10 meV. Instead of the expected Bogoliubov-like back bending of the dispersion, a sharp, nondispersive quasiparticle peak centered at the gap energy of 6.5 meV is present and is separated by a dip from the high energy spectral weight. All these features vanish above T_c, as expected for a superfluid excitation, and are likely due to the interaction of electrons with a Leggett mode.

MgB_2 single crystals with $T_c = 39$ K and a typical size of $0.5 \times 0.5 \times 0.3$ mm^3 were grown in the Ames Laboratory by a high pressure synthesis technique similar to that outlined in Ref. [33] using a pure ^{11}B isotope. Optimally doped (OP) $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212) single crystals with $T_c = 93$ K (OP 93 K) were grown by the conventional floating-zone (FZ) technique. Sample were cleaved *in situ* at a base pressure of lower than 8×10^{-11} Torr. ARPES measurements were carried out using a laboratory-based system consisting of a Scienta R8000 electron analyzer and a tunable VUV laser light source [34]. All data were acquired using a photon energy of 6.7 eV. The energy resolution of the analyzer was set at 1 meV and the angular resolution was 0.13° and ~0.5° along and perpendicular to the direction of the analyzer slits, respectively. Samples were cooled using a closed cycle He refrigerator and temperature was measured using a silicon-diode sensor mounted on the sample holder. The energy corresponding to the chemical potential was determined from the Fermi edge of a polycrystalline Au reference in electrical contact with the sample. The aging effect was checked by recycle measurements. The consistency of the data was confirmed by measuring several samples.

The Fermi surface (FS) data are shown in Fig. 1(a), with a schematic plot of the Brillouin zone inset. We used the peak position of momentum distribution curves (MDCs) at E_F to quantitatively extract k_F. Results are superposed as red points on the image data. Both σ FS sheets are round with $|k_F| \sim 0.2$ and ~0.25 Å^{-1}, respectively. If we ignore the small warping of these two σ sheets along k_z, the contribution to the carrier concentration would be 0.069 holes for the inner FS sheet and 0.108 holes for the outer one. The measured area of the outer FS is consistent with previous quantum oscillation results, while the inner one is slightly larger [35,36]. Figures 1(b) and 1(c) show ARPES intensity plots at two

different cuts in the Brillouin zone. In the data measured along cut No. 1 [Fig. 1(b)] the Fermi crossings for both σ sheets are clearly visible, whereas in cut No. 2, along the symmetry axis [Fig. 1(c)], the intensity of the inner σ band is strongly suppressed due to the matrix element effect. In order to quantitatively analyze the renormalization effects due to the collective modes, we fit the MDCs of each data set with Lorentzians and plot such extracted dispersions as blue and red lines in Figs. 1(b) and 1(c). It should be noted that MDC peaks do not reflect the dispersion at very low energies in the presence of the superconducting gap, therefore the fitting is carried out only for binding energies larger than $\sim 2\Delta$. We estimate bare dispersion by extrapolation from higher binding energies and plot them as dashed lines. In all three data sets, a very pronounced kink structure is clearly visible (indicated by arrows), where the renormalized dispersion deviates from the bare estimate. In this case, the renormalization of the dispersion (kink) is the fingerprint of coupling the conduction electrons to phonon mode(s). This coupling is rather strong and the energy distribution curves (EDCs) at k_F develop a dip [right side of Fig. 1(c)]. The kink in dispersion and dip in EDC occurs at $\sim -(70$ to $75)$ meV. By comparison with Raman data [26], we can conclude that it is due to the E_{2g} optical phonon responsible for pairing in MgB_2 [19]. We subtract an estimated bare dispersion from the measured one to obtain the approximate $\Sigma'(\omega)$, and use the widths of the Lorentzian MDC fits to obtain $\Sigma''(\omega)$. These quantities are plotted in Fig. 1(d). Not surprisingly, $\Sigma'(\omega)$ has a peak at -70 meV, which is very close to the phonon energy. $\Sigma''(\omega)$ rises rapidly with binding energy and has a midpoint of the step roughly located at the same energy as the peak in $\Sigma'(\omega)$. The line shape of both curves is consistent with them being related by Kramers-Kronig. This is further verified by simulations presented in the Supplemental Material [37].

The dispersion data extracted by fitting the positions of the MDC peaks for several temperatures both below and above T_c are shown in Fig. 2(a). There are no significant changes to the kink structure across T_c, as is evident from the overlay of the high temperature curve (dashed red line) onto the lowest temperature one. The only noticeable changes occur at low energies due to the opening of the superconducting gap. Following the procedure outlined above, we extract $\Sigma'(\omega)$ for each temperature, and plot these in Fig. 2(b). The peak position does move to lower energies as the temperature decreases, starting at T_c, consistent with the expectation that the peak frequency increases from $\Omega \rightarrow \Omega + \Delta$. However, the magnitude of the shift, 3 meV, is significantly smaller than the expected $\Delta_\sigma = 6.5$ meV. In fact, this shift should be even larger as the screening electrons are gapped out and the E_{2g} phonon hardens below T_c. This hardening was predicted to be $\approx 10\%$ or ≈ 7 meV [19], but Raman measurements find that it only shifts by $\Delta\Omega \approx 2.5$ meV [38]. So the overall shift below T_c is naively expected to be $\Delta_\sigma + \Delta\Omega \approx 9$ meV. However, this analysis neglects the multigap nature, which may account for shifts as small as 3 meV by allowing scattering into the π band.

In addition to extracting the energy of the collective mode, we can also estimate the electron-phonon coupling $\lambda = v_0/v_F - 1$, where v_0 and v_F are the bare and renormalized Fermi velocities, respectively. v_0 is estimated from

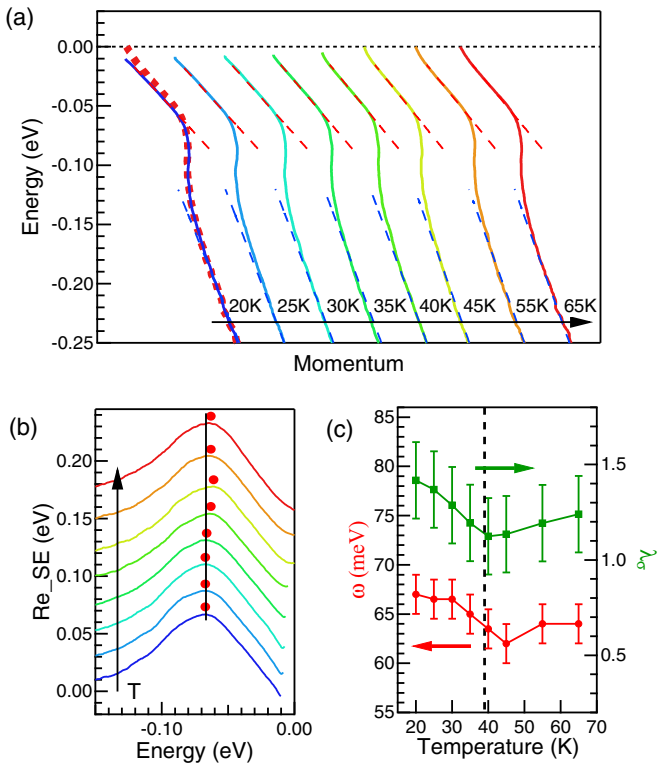


FIG. 2. (Color online) Temperature dependence of the renormalization effect. (a) Band dispersions obtained from MDC fits along cut No. 2 [see Fig. 1(a)] measured at different temperatures. Data are shifted horizontally for clarity. The band dispersion at 25 K (blue solid line) is superimposed with 65 K data (thick red dotted line) for a direct comparison of the effect below and above T_c. Blue dotted and red dashed lines illustrate the dispersion used to extract velocity at low and high binding energies for estimating the coupling constant. (b) Extracted effective real part of self-energy obtained from (a). Data are shifted vertically with 25 meV intervals for each temperature. Red dots mark the peak position. (c) Temperature dependence of the kink energy (red squares) and the coupling constant (green circles).

the dispersion at high energy and v_F is obtained from the dispersion above the kink [as illustrated in Fig. 2(a) by red dashed and blue dotted lines]. For cut No. 2 in Fig. 1(c), $v_0 \sim 3.77$ eV Å and $v_F \sim 1.56$ eV Å, which implies $\lambda_\sigma = 1.42$. The electron-phonon coupling can be calculated numerically, where it results from scattering between the σ and π bands, $\lambda_{\sigma\pi} = 0.23$ or from intra-σ-band scattering, $\lambda_{\sigma\sigma} = 0.96$, for a total of $\lambda_\sigma = 1.19$ [19,39], which is slightly smaller, but within the error bars of our results, given the uncertainty of estimating v_0.

We now turn to describe the second, low energy excitation present only below the superconducting transition. In Figs. 3(a)–3(c) we plot the dispersion of the σ_1 band in close proximity to E_F below and above T_c. At low temperature, well below T_c, the shape of the dispersion is rather unusual for a superconductor. Instead of back bending of the band as it reaches the energy of the superconducting (SC) gap, a sharp streak of intensity is present at the gap energy on either side of k_F. At 29 K [Fig. 3(b)], this feature is also above E_F due to increased thermal excitation. All of these features vanish just above T_c [Fig. 3(c)], where the ordinary

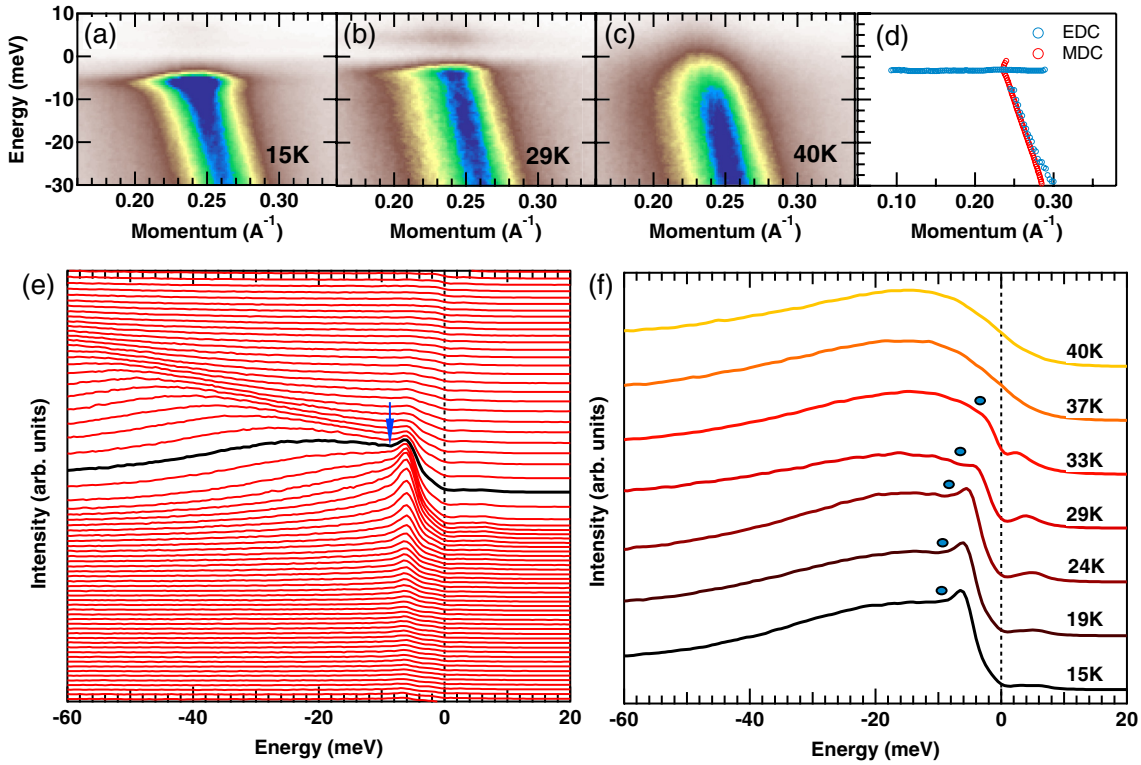


FIG. 3. (Color online) Low energy band dispersion and kink. (a)–(c) ARPES intensity measured at 15, 29, and 40 K along cut No. 2 [see Fig. 1(a)]. (d) Dispersion at low temperature extracted using MDC and EDC fits to data in (a). (e) EDCs measured at 15 K: The arrow marks the location of the dip which roughly corresponds to the binding energy at which the Leggett mode is observed. The energy of the Leggett mode itself is the difference between this binding energy and value of the superconducting gap as explained in the text. (f) Temperature dependence of the EDCs. The clear dip in the spectrum due to the interaction with the Leggett mode is marked by an oval.

conduction band is present. In Fig. 3(d) we plot the dispersion of the low temperature features extracted using EDC and MDC fits. The sharp peak of intensity is almost dispersionless and persists over δk of 0.2 Å^{-1}, unlike what is expected for Bogolubov quasiparticles [40,41]. The EDCs for low temperature data are plotted in Fig. 3(e). Here again we observe a sharp, dispersionless quasiparticle peak centered at the energy of the SC gap and separated from the rest of the spectral weight by a dip at ∼10 meV (marked by an arrow). The temperature dependences of the EDCs slightly off k_F are shown in Fig. 3(f). A clear dip in the spectrum is observed, which vanishes as the temperature approaches T_c. All of these features are due to abrupt changes in the self-energy (i.e., onset of resonant scattering) and are very characteristic of an interaction between the electrons and a collective excitation. It should be noted that opening of the SC gap can lead to suppression of scattering within an energy of $3\Delta_\sigma = 19.5$ meV for intraband and $\Delta_\sigma + 2\Delta_\pi = 9.5$ meV for interband electron channels. We do not observe signatures of suppression of intraband scattering (i.e., line shape features at 19.5 meV) which are deemed to be stronger than the interband ones [42]. Furthermore, strong suppression of scattering and a reduction of $\Sigma''(\omega)$ below a certain energy alone cannot produce a spectral dip and nondispersive peaks. That requires a resonant process, such as an interaction with a collective mode that causes a peak in $\Sigma''(\omega)$. An illustration of this in the form of simulations of the spectral function for various scenarios of $\Sigma''(\omega)$ are presented in the Supplemental Material [37].

The lowest energy of an optical phonon in MgB_2 is 35 meV and there are no other obvious low energy excitations that could couple strongly to the electrons other than the Leggett mode. Whereas Raman spectroscopy has found the Leggett mode at 9.4 meV [31], the similarity of this value with the dip energy of 10 meV is coincidental, as the dip occurs at the frequency where electrons above the gap may scatter into the collective mode plus quasiparticles above the superconducting gap. Therefore, we extract the value of the mode energy from the ARPES data of 3.5 meV, which is the difference between the energy location of the sharp peak and dip in the spectrum. We stress that this is an estimate as the dip location is slightly affected by the functional form of the self-energy. The vertex corrections for Raman spectroscopy [31,32] and ARPES are almost certainly different and can potentially explain the difference between the Leggett mode energy measured by the two techniques. Previous electron spectroscopy studies reported a value of the Leggett mode energy of 3.9–4.0 meV, consistent with our results. While the bare mode was calculated to have a frequency of 5.1–6.2 meV [31,32], if the experimentally measured $\Delta_\pi = 1.5$ meV is used, the calculated frequencies decrease to 3.8–4.5 meV. More theoretical efforts will be required to fully understand the origin of the difference.

We also note that spectral characteristics reported in Fig. 3 (a nondispersive sharp peak and the presence of a dip) closely resemble features observed at the antinode in the cuprates that are attributed to the strong coupling of electrons to a

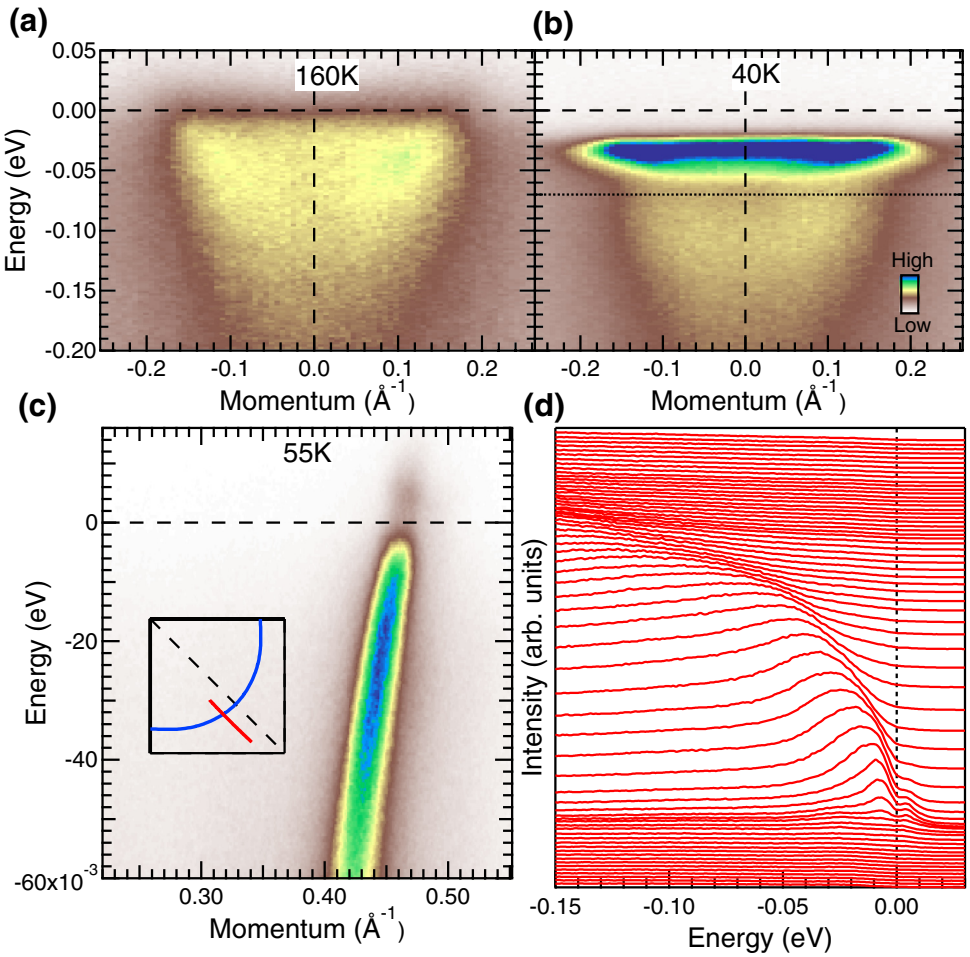


FIG. 4. (Color online) ARPES data from OP Bi2212 for comparison. The intensity plot at the antinode in (a) normal and (b) superconducting states measured using a helium discharge lamp and 21.2 eV photons. The dashed line in (b) marks the location of the dip (local minimum of intensity) due to the interaction with a 40 meV collective mode. This feature is absent in the normal state (a). (c) Intensity plot and (d) EDCs in the SC state along a cut slightly off the node (marked in the inset), where the SC gap magnitude is ∼8 meV measured using a laser source and 6.7 eV photons. The intensity peak above E_F is due to electrons thermally excited above 2Δ.

collective mode. The single order parameter of the cuprates does not support a Leggett mode, but these properties are characteristic of any collective mode developing only below the superconducing transition. In Fig. 4 we show data from optimally doped Bi2212 for comparison. Figures 4(a) and 4(b) show data at the antinode where electrons couple strongly to a collective mode below T_c. In the SC state, a very sharp, weakly dispersing peak emerges at the energy of the SC gap, followed by a dip of intensity. These two features—the lack of substantial dispersion of the sharp quasiparticle peak and the presence of a dip—are key signatures of the interaction with a collective mode. The data close to the node ($\Delta = 8$ meV) where the coupling to low energy modes in Bi2212 is very weak [9] in the SC state display a Bogoliubov-like dispersion and sharp, nondispersive peaks are absent. This is in stark contrast to the antinodal direction and the present data from MgB_2.

Our results demonstrate that in a prototypical, conventional superconductor MgB_2, the coupling of the conduction electrons to the phonon mode is not significantly affected by the SC transition. This is in contrast to the behavior of the collective mode at the antinode in cuprates, which disappears upon the transition to the normal state. We also discovered a signature of a second collective mode in MgB_2 with energy $\Omega \sim 3.5$ meV that exists only below T_c. All the characteristics of this mode are consistent with the Leggett mode which arises due to the relative oscillation of the phases of two superconducting condensates present in this material.

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