Influence of the third dimension of quasi-two-dimensional cuprate superconductors on angle-resolved photoemission spectra

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Angle-resolved photoemission spectroscopy (ARPES) presents significant simplifications in analyzing strictly two-dimensional (2D) materials, but even the most anisotropic physical systems display some residual three-dimensionality. Here we demonstrate how this third dimension manifests itself in ARPES spectra of quasi-2D materials by considering the example of the cuprate Bi$_2$Sr$_2$CaCu$_2$O$_8$ (Bi2212). The intercell, interlayer hopping, which is responsible for $k_z$, dispersion of the bands, is found to induce an irreducible broadening to the ARPES line shapes with a characteristic dependence on the in-plane momentum $k_i$. Our study suggests that ARPES line shapes can provide a direct spectroscopic window for establishing the existence of coherent $c$-axis conductivity in a material via the detection of this broadening mechanism, and bears on the understanding of 2D to 3D crossover and pseudogap and stripe physics in novel materials through ARPES experiments.

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Angle-resolved photoemission spectroscopy (ARPES) has been applied extensively in the recent years for investigating the electronic and quasiparticle properties of high-temperature superconductors. Much of the existing ARPES work on the cuprates and other quasi-two-dimensional (2D) materials implicitly assumes perfect two-dimensionality, ignoring the effects of dispersion in the third dimension. As energy resolutions of the order of a few meV’s have now become possible in the state-of-the-art ARPES instrumentation, it is natural to ask the question: How will $k_z$ dispersion play out in the ARPES spectra of the quasi-2D materials? An obvious answer is that spectral peaks will undergo shifts with photon energy—albeit small—much like in the 3D case. Our analysis, however, reveals a very different scenario in that insofar as ARPES response to $k_z$ dispersion is concerned, quasi-2D systems differ fundamentally from their 3D counterparts. We find that in the presence of typical final state dampedings of the order of eV’s, initial state dispersions over much smaller energy scales will not appear as energy shifts with $h\nu$, but will instead induce an irreducible linewidth in the spectral peaks with a characteristic $k_i$ dependence.

Our results bear on a variety of important issues in cuprate physics and give insight into a number of puzzling features of ARPES in the cuprates. Since our broadening mechanism does not have its origin in a scattering process, it explains why the lifetimes derived from ARPES spectra are generally found to be shorter than those obtained from other experiments. Moreover, Fermi surface (FS) maps obtained via ARPES often display broad patches of spectral weight rather than well-defined FS imprints, particularly near the antinodal point; such patches occur naturally in our calculations. Indeed, much of the pseudogap phenomena—in particular the lack of well-defined quasiparticles near $k=(\pi,0)$—will need to be reevaluated in light of the present findings. In this vein, these results also impact the analysis of stripe physics through ARPES experiments. Finally, detection of the $k_z$-related linewidths in the ARPES spectra offers a unique spectroscopic window for establishing the existence of coherent $c$-axis conductivity and intercell coupling in a system; this important intrinsic property is not accessible directly through other techniques.

Specifically, we focus in this article on the tetragonal body-centered Bi$_2$Sr$_2$CaCu$_2$O$_8$ (Bi2212) compound, which has been a workhorse of ARPES studies. Bi2212 is a nearly 2D material with two CuO$_2$ layers in the primitive unit cell. The *intracell* interaction between the two CuO$_2$ layers, spaced a relatively short distance of $\sim$3.2 Å apart, results in the well-known bilayer splitting. On the other hand, the *intercell* coupling between the bilayer slabs in different unit cells is expected to be smaller due to the larger intercell Cu-Cu distance of $\sim$12 Å resulting in weak but nonvanishing $k_z$ dispersion. The intercell coupling will be even more pronounced in other high-$T_c$ compounds since Bi2212 presents one of the longest $c$ axes in the cuprate family.

Concerning computational details, the band structure for Bi2212 was obtained within the local-density approximation (LDA) by using the well-established Green’s function methodology. The crystal potential used is the same as that employed in our previous studies of Bi2212 and involves 30 atoms per conventional unit cell, yielding good agreement with the experimentally observed Fermi surface (FS). ARPES intensities have been computed within the one-step photoemission formalism; see Refs. 6 and 7 for details.

Figure 1(a), which considers the familiar antibonding ($A$) and bonding ($B$) bands in Bi2212, shows that the $k_i$ dispersion depends strongly on $k_z$, as the associated bands wander over the two sets of shaded areas. The $k_z$ dispersion (i.e., the vertical width of the shaded areas) displays a striking dependence on $k_z$ and nearly vanishes at the antinodal point $k_i=0$. A clear bilayer splitting between the $A$ and $B$ bands is seen at all $k$-values, except at $k_i=k^*_i=0.2(2\pi/a)$ for $k_z=0$ (solid lines), where $A$ and $B$ bands touch. This level crossing leads to the anomalous $k_z$ dispersion shown in Fig. 1(b) for the $B$ band: The shape changes from being described approximately by the form $\sin^3(k_{i,c}/4)$ for $k_i=0$, to...
increased from a very small value in order to conserve energy. Note that the total shift in the peak imaginary part of the final state self-energy, $\Sigma_f$, provided by the simulations of Fig. 2, which consider the structure of Fig. 3. The complex behavior of the bilayer dispersion leads to an irreducible linewidth in the ARPES spectra, which cannot be resolved by changing photon frequency. This effect will also

Continuing to the intermediate case of $\Sigma_f=0.1$ eV in Fig. 2(b), the shift in the peak position from the bottom to the top of the initial state band is once again evident, but the line shapes are quite different, even though the initial state damping $\Sigma_i$ in (b) is identical to that in (a). It is striking that some spectral intensity appears in (b) at all energies encompassed by the initial state band at every $h\nu$. This remarkable effect comes about because the energy uncertainty permitted by the width of the final state allows the photoelectron to couple with initial states off the energy shell. The line shape thus develops a new component with an $h\nu$-independent width equal to the initial state bandwidth, which rides on top of the energy conserving peak in (a). Notice also how the changes in the peak position could allow observing different FS's as one maps different values of $k_z$. Finally, for the realistic final state width of $\Sigma_f=1$ eV in (c), the initial state bandwidth component dominates the lineshape. The line shape of the EDC curve is now virtually $h\nu$ independent. Despite the large final-state broadening, the energy spread of the EDC remains equal to the initial state bandwidth in $k_z$ direction because outside of this interval, there are no initial state electrons capable of absorbing the photon.

Figure 3 elaborates on these points by considering some of the results of Fig. 2 over a much broader range of photon energies. Figure 3(a) presents the spectra of Fig. 2(b) for $\Sigma_f=0.1$ eV for $h\nu=19–27$ eV. Different colored bands here represent the same initial states being excited to different final states. Comparing Figs. 3(a) and 3(b), we see that the peaks of Fig. 3(a) more or less follow the final state band structure of Fig. 3(b) (the agreement is not expected to be perfect of course, since the initial state dispersion is not entirely negligible). In Fig. 3(c), when a more realistic final state width of $\Sigma_f=1$ eV is assumed, much of the structure is lost. The large variations in colors (or intensities) observed in Figs. 3(a) and 3(c) are the consequence of the well-known $k_z$ and $h\nu$ dependency of the ARPES matrix element.

The preceding discussion of Figs. 2 and 3 makes it obvious that in a quasi-2D system, $k_z$ dispersion leads to an irreducible linewidth in the ARPES spectra, which cannot be resolved by changing photon frequency. This effect will also

FIG. 1. (a) Calculated first-principles $k_z$ dispersion in Bi2212 along the [100] direction for the $B$ and $A$ bands at three different $k_z$ values (in units of $2\pi/a$): $k_z=0$ (solid lines); $k_z=0.5$ (dashed), and $k_z=1$ (dash-dotted). Shading denotes the regions over which the bands wander as a function of $k_z$. (b) $k_z$ dispersion of the $B$ band at five different $k_z$ values ranging from 0 to $\pi/a$. (c) Same as (a), except that these results are based on a tight binding formalism.

FIG. 2. Simulated ARPES line shapes (EDC’s) in Bi2212 for a series of photon energies ($h\nu=25–25.7$ eV) at a fixed $k_z=(0.34,0.09)2\pi/a$ point using three different values of the final state broadening given by the indicated imaginary parts of the self-energy, $\Sigma_f$. In order to highlight the influence of $k_z$ dispersion, the initial state broadening is chosen to be very small, $\Sigma_i=0.2$ meV (see Ref. 17).
perimental resolution on the results. Note that even with the window of ±30 meV in (d), the broadening effect of $k_z$ dispersion is not washed out. In fact, the broadening is somewhat enhanced, especially near the antinodal point due to the contribution of the flat bands related to the van Hove singularity (VHS).

It is also interesting to consider changes in the linewidth in momentum, $\Delta k$, as one moves away from the Fermi level. In general, our simulations indicate that $\Delta k$ increases with increasing binding energy (BE), due mainly to the flattening of bands and the concomitant reduction in the band velocity. In any event, the broadening as a function of BE is neither simply quadratic nor exponential. Furthermore, considering the effect of varying $\Sigma''_i$, at a general $k_i$ point, we have found that as the initial state damping due to intrinsic scattering mechanisms (simulated via the value of $\Sigma''_i$) decreases, the linewidth $\Delta E$ becomes increasingly dominated by the irreducible width associated with $k_z$ dispersion. Along the antinodal direction this dispersion is negligible, and $\Delta E$ and $\Sigma''_i$ are related linearly.

We return now to comment briefly on the bands of Figs. 1(a) and 1(b). Insight into how the complex $k_z$ and $k_c$ dependencies of these bands reflect intercell as well as intracell hopping effects can be gained by modeling these bands within the TB framework. In the absence of intercell coupling, the conventional bilayer splitting possesses the form $t_{bi}=t_{i}(c_{s}-c_{i})^{2}$, with $c_{i}=\cos(k_{ia})$, $i=x,y$. The intercell coupling in the cuprates may be included in the one-band model with dispersion

\[
\epsilon_{i} = -2(t_{i}(c_{s}+c_{i})-4t'_{i}c_{i}T_{z}(k_{i},r_{i})[c_{i} - c_{s}]^{2}/4+a_{0}],
\]

where $s_{i} = \sin(k_{i}/4)$ and $a_{0}=0.6$ corrects for the finite bilayer splitting found at $k_{ij}=0$. The form of $T_{z}$ depends on the particular cuprate considered. While Ref. 22 lists seven inequivalent interlayer hopping parameters, we find that we can describe the dispersion reasonably by including only the dominant contribution associated with hopping between Cu 4$s$ levels by introducing $T_{z} = \pm \sqrt{(t_{s}-t'_{s})^{2} + 4t_{s}t'_{s}s_{z}^{2}}$, where plus (minus) sign refers to the bonding (antibonding) solution. $t_{s}$ is a constant associated with intracell interlayer hoppings, and $t'_{s}$ = $4t'_{s0}\cos(k_{ia}/2)\cos(k_{ia}/2)$ accounts for intercell hopping.

The extra angular dependence in this term arises because for intercell hopping the two CuO$_2$ planes are offset, so that one Cu atom sits above an empty site. Note that for $t'_{s}=0$ one obtains the simple bilayer splitting $t_{bi}$ with no $k_z$ dispersion. TB bands of Fig. 1(c) assume (in eV): $t=0.42$, $t'_{s}=-0.12$, $t_{s} = 0.125$, and $t'_{s0}=0.04$. A comparison of the first principles and TB bands in Figs. 1(a) and 1(c), respectively, shows that our TB model reproduces the bilayer splitting, the collapse of the $k_z$ dispersion around $(\pi/a,0)$, and the anomalous dispersions associated with level crossing to a reasonable degree.

We emphasize that $t'_{s}$, which provides intercell coupling, is essential for obtaining coherent $c$-axis conductivity. $t'_{s}$ thus controls the intrinsic resistivity anisotropy and gives a measure for discriminating between coherent and incoherent $c$-axis hopping. The lifetimes in the cuprates extracted from ARPES are considerably smaller than optical lifetimes or those deduced from transport or tunneling measurements. A part of this difference may be explained to be the result of an unresolved bilayer splitting. The re-
remaining anomalous broadening is often interpreted\textsuperscript{26} in terms of small-angle scattering, which does not contribute to transport, but its origin remains obscure.\textsuperscript{27} The present analysis indicates that part of the broadening of ARPES lines has its origin in the $k_z$ dispersion, unrelated to any scattering mechanism.

Our predicted effects of $k_z$ dispersion on the linewidths in Bi2212 should be resolvable with the power of currently available high resolution ARPES instrumentation.\textsuperscript{28} The size of our novel line broadening mechanism will be larger in transport, but its origin remains obscure.\textsuperscript{27} The present analysis indicates that part of the broadening of ARPES lines has its origin in the $k_z$ dispersion, unrelated to any scattering mechanism.

In conclusion, we have demonstrated that residual $k_z$ dispersion in quasi-2D materials will induce an irreducible linewidth in ARPES spectra. This intrinsic linewidth offers a new spectroscopic window for understanding in- as well as out-of-plane scattering mechanisms and the nature of the 2D to 3D crossover in the cuprates. This highly anisotropic line broadening mechanism, which has not been recognized previously and is unrelated to 2D physics, indicates that the existing analysis of stripe and pseudogap physics based on ARPES spectra should be reexamined. Our study shows how ARPES can be extended to unravel the hidden third dimension of energy bands and Fermi surfaces in quasi-2D systems with wide-ranging consequences for understanding the nature of electronic states in many novel materials.

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2. Throughout this article the, $z$ axis is defined to be normal to the Cu-O planes.
12. The well-known “Bi pockets” in the vicinity of the $M$ point have been lifted above the Fermi energy by modifying the Bi and O potentials, consistent with experimental data.
13. It should be kept in mind that LDA calculations systematically overestimate band dispersions, typically by a factor of about 2–4.
17. The spectra have not been convoluted with the Fermi function throughout this article.
18. The imaginary part of the self-energy $\Sigma$ is proportional to the inverse of the lifetime of the (quasi)particle in that state.
20. The ±30 meV window used would simulate an effective experimental resolution of about ±8 meV (see Ref. 13).
21. The situation at the nodal point is more complicated, due to a small residual bilayer splitting. See also A. A. Kordyuk et al., cond-mat/0311137, Phys. Rev. B (to be published).
23. Of course, in Bi2212, the anomalous temperature dependence of $\rho_x$ strongly hints that it is incoherent.
28. Note from Fig. 1 that a typical broadening is a significant fraction of the full bilayer splitting, which is experimentally found to be about 100 meV (recall Ref. 13).