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 $k_{\parallel}$. Our study suggests that ARPES line shapes can provide a direct 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\textsuperscript{3,4,5}.

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 ~ 3.2 Å apart, results in the well-known bilayer splitting\textsuperscript{6,7,8,9,10}. 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 ~ 12 Å resulting in weak but non-vanishing $k_z$-dispersion. The intercell coupling will be even more pronounced in other high-Tc’s 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\textsuperscript{11}. The crystal potential used is the same as that employed in our previous studies of Bi2212 and involves 30 atoms per conventional unit cell\textsuperscript{12,13,14,15,16}. ARPES intensities have been computed within the one-step photoemission formalism; see, Refs.\textsuperscript{12} and\textsuperscript{17} for details.

Figure 1(a), which considers the familiar antibonding (A) and bonding (B) bands in Bi2212, shows that the $k_z$-dispersion depends strongly on $k_{\parallel}$, 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 $k_{\parallel}$ and nearly vanishes at the antinodal point $k_{\parallel} = (\pi/a, 0)$. A clear bilayer splitting between the A and B bands is seen at all $k_z$ values, except at $k_{\parallel} = k_{\parallel}^c = 0.2(2\pi/a)$ for $k_z = 0$ (solid...
binding (TB) Hamiltonian as seen from Fig. 1(c). We be modeled reasonably well by a relatively simple tight-principles computations of Figs. 1(a) and 1(b) can approximately by the form $\sin(2k_zc/\pi)$ for $k_z = 0$ (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 $k_{\parallel}$ values ranging from 0 to $\pi/a$. (c): Same as (a), except that these results are based on a tight binding formalism.

FIG. 1: (a): Calculated first-principles $k_{\parallel}$-dispersion in Bi2212 along [100]-direction for the B and A bands at three different $k_z$ values (in units of $2\pi/c$): $k_z = 0$ (solid lines); $k_z = 0.5$ (dashed), and $k_z = 1$ (dash-dotted). The complex behavior of these results is based on a tight binding formalism.

From the displayed graph, one can observe the change in $k_z$-dispersion for different $k_{\parallel}$ values. The B band shows a peak at $k_{\parallel} = 0$ and disperses with increasing $k_{\parallel}$, while the A band remains flat.

FIG. 2: Simulated ARPES lineshapes (EDCs) in Bi2212 for a series of photon energies ($h\nu = 25 - 25.7$ eV) at a fixed $k_{\parallel} = (0.34, 0.09)2\pi/a$-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.

In (a), the position of the spectral peak undergoes the familiar shift as $h\nu$ varies. This shift results from the fact that in the photoexcitation process $k_{\parallel}$ must remain unchanged in transmission of the electron across the surface and the initial and final states can connect only at a specific value of $k_z$ in order to conserve energy. Note that the total shift in the peak position in the EDCs of (a) gives the size of the $k_z$-dispersion of the initial state. This should not be confused with the change in $h\nu$ needed to probe such a band. The change in $h\nu$ is controlled by the final state dispersion, which is generally much larger than that of the initial state.

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 lineshapes 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 lineshape 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 FSs 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 lineshape 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.

Fig. 3 elaborates on these points by considering some of the results of Fig. 2 over a much broader range of photon energies. Fig. 3(a) presents the spectra of Fig. 2(b)
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 appear in the FS maps observed by measuring emission from \( E_F \) in the \((k_z,k_y)\)-plane. Fig. 4 illustrates how this plays out. The standard bilayer split FS in the \( k_z = 0 \) plane, given in Fig. 4(a), is the FS usually thought to be measured in ARPES. In Fig. 4(b) the full 3D FS has been projected on to the (001)-plane by collecting individual FS cuts corresponding to different \( k_z \) values. The \( k_z \)-dispersion is now seen to introduce an effective “broadening” to the FS imprint. In a perfect 2D-system, the maps in (b) and (a) would be identical. The effect of using a finite energy window of \( \pm 30 \) meV around \( E_F \) in the computations is shown in Figs. 4(c) and 4(d). These simulations indicate the influence of a finite experimental resolution on the results.\(^{13,20}\) Note that even with the window of \( \pm 30 \) 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.\(^{21}\) In any event, the broadening as a function of BE is neither simply quadratic nor exponential. Furthermore, considering the the effect of \( \Sigma''_f \) at a general \( k_z \)-point, we have found that as the initial state damping due to intrinsic scattering mechanisms (simulated via the value of \( \Sigma''_f \)) 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 \( \Sigma''_f \) are related linearly.\(^{21}\)

We return now to comment briefly on the bands of Figs. 1(a) and 1(b). Insight into how the complex \( k_{\parallel} \)- and \( k_z \)-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 \( t_{bi} = t_z(c_x-c_y)^2 \), with \( c_i = \cos(k_i a) \), \( i = x,y \). The intercell coupling in the cuprates may be included in the one-band model with dispersion\(^{22}\)

\[
el_k = -2t(c_x + c_y) - 4t'c_x c_y - T_z(k_\parallel, s_z)[(c_x - c_y)^2/4 + a_0],
\]

where \( s_z = \sin(k_z c/4) \) and \( a_0 = 0.6 \) corrects for the finite bilayer splitting found at \( k_z = 0 \). 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 4s levels by introducing $T_z = \pm \sqrt{(t_z - t^2) + 4t_z s^2}$, where plus (minus) sign refers to the bonding (antibonding) solution. $t_z$ is a constant associated with intracell interlayer hoppings, and $t'_z = 4t_z \cos(k_x \alpha / 2) \cos(k_y \alpha / 2)$ 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 cell. Note that for $t'_z = 0$ one obtains the simple bilayer splitting $t_{ni}$ with no $k_z$-dispersion. TB bands of Fig. 2c assume (in eV): $t = 0.42$, $t' = -0.12$, $t_z = 0.125$, and $t''_z = 0.04$. 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 reasonably good degree, some minor discrepancies notwithstanding.

We emphasize that $t'_z$, which provides intercell coupling, is essential for obtaining coherent $c$-axis conductivity. $t'_z$ 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 remaining anomalous broadening is often interpreted in terms of small-angle scattering, which does not contribute to transport, but its origin remains obscure. 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. The size of our novel line broadening mechanism will be larger in materials with greater 3D character (e.g. YBCO, LSCO, NCCO, ruthenates, manganites, etc.), although we expect the results of Figs. 1-4 to provide at least a qualitative handle in quasi-2D materials more generally. Notably, coherent 3D coupling has recently been reported in a Tl cuprate. Additionally, a similar $k_z$-induced broadening may be expected to have an impact on other experimental probes, such as Raman scattering.

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-rangng consequences for understanding the nature of electronic states in many novel materials.

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