Importance of matrix elements in the ARPES spectra of BISCO

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We have carried out extensive first-principles angle-resolved photointensity (ARPES) simulations in Bi\(_{2}\)\(_{2}\)\(_{1}\)\(_{2}\) wherein the photoemission process is modelled realistically by taking into account the full crystal wavefunctions of the initial and final states in the presence of the surface. The spectral weight of the ARPES feature associated with the CuO\(_2\) plane bands is found to undergo large and systematic variations with \(k_\parallel\) as well as the energy and polarization of the incident photons. These theoretical predictions are in good accord with the corresponding measurements, indicating that the remarkable observed changes in the spectral weights in Bi\(_{2}\)\(_{2}\)\(_{1}\)\(_{2}\) are essentially a matrix element effect and that the importance of matrix elements should be kept in mind in analyzing the ARPES spectra in the high-Tc’s.

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Angle-resolved photoemission spectroscopy (ARPES) has contributed significantly towards an understanding of the nature of the normal as well as the superconducting state of the cuprates \([1,2]\). Much of the data on the cuprates, however, has been analyzed by assuming that the ARPES essentially measures the one-particle spectral function of the initial states. While this simple approach yields insights into the underlying physics, a satisfactory function of the initial states. A simple approach yields insights into the underlying physics, a satisfactory function of the initial states. While this simple approach yields insights into the underlying physics, a satisfactory function of the initial states. While this simple approach yields insights into the underlying physics, a satisfactory function of the initial states. While this simple approach yields insights into the underlying physics, a satisfactory function of the initial states.

This article presents first-principles simulations of ARPES spectra in Bi\(_{2}\)\(_{2}\)\(_{1}\)\(_{2}\) (BISCO) using the one-step model of photoemission which incorporates the aforementioned effects realistically \([3,4]\). We focus on the ARPES signature of CuO\(_2\) plane bands which are widely believed to be the key to the mechanism of superconductivity in the cuprates. The spectral weight of the ARPES peak associated with the CuO\(_2\) plane bands is found to undergo large variations with \(k_\parallel\) as well as the energy and polarization of the incident photons. These theoretical predictions are in remarkable accord with the corresponding measurements \([1,4]\) on BISCO, and show clearly the importance of “matrix element effects” \([2]\) in the ARPES spectra. Notably, a substantial increase in the ARPES spectral weight in BISCO in going from \(\Gamma\) to \(M\) was noted early by Anderson \([3]\), who speculated that this puzzling behavior may be the hallmark of spin charge separation.

The physical and formal underpinnings of our approach may be exposed by starting with the following Golden rule-based expression for photointensity from initial states at energy \(E\) with photons of energy \(\hbar\omega\): \([3]\)

\[
I(E, \hbar\omega) = \frac{2\pi e^2}{\hbar} \sum_{i,f} \left| \langle \Psi_f | \Delta | \Psi_i \rangle \right|^2 \delta(E_f - E_i - \hbar\omega) \tag{1}
\]

\(\Psi_f(\Psi_i)\) are the initial (final) states of the semi-infinite solid, and \(\Delta = \epsilon_0/2mc(p \cdot A + A \cdot p)\) is the interaction Hamiltonian with the electron momentum operator \(p\) and the vector potential \(A\) of the photon field.

In the one-step model used in the present computations, Eq. 1 is manipulated into the form \([3]\):

\[
I(k_\parallel, E, \hbar\omega) = \frac{1}{\pi} I_m \left| k_\parallel | G^\parallel_2(E + \hbar\omega) \Delta G^\parallel_1(E) \Delta^\dagger \right| G^\parallel_2(E + \hbar\omega) | k_\parallel > \tag{2}
\]

where the matrix element involves the free electron final state of momentum \(k_\parallel\). \(G_2\) and \(G_1\) denote the retarded (+) or advanced (-) one-electron Green functions at appropriate energies.

Notably, the so-called three-step model of photoemission approximates the matrix element in Eq. 1 via the \textit{bulk} Bloch wavefunctions yielding for the photointensity within the solid \([3]\):

\[
P(E, \omega) = \sum_{f,i} \int d^3k \left| \langle \Psi_f^{\text{bulk}} | \Delta | \Psi_i^{\text{bulk}} \rangle \right|^2 \times A_f(E + \hbar\omega) A_i(E) \tag{3}
\]

which is cast in terms of the one-particle spectral functions of the initial and final states \((A_i\) and \(A_f\), and the processes of transport and emission are to be considered separately. Assuming that: (i) The system is strictly 2D, and (ii) the final states form a structureless continuum, Eq. 3 reduces to:

\[
P(E, \omega) \sim \sum_i \left| \langle \Psi_f^{\text{bulk}} | \Delta | \Psi_i^{\text{bulk}} \rangle \right|^2 A_i(E) \tag{4}
\]

in terms of just the \(A_i(E)\)’s. Further, for a single band solid, one obtains:

\[
P(E, \omega) \sim \left| \langle \Psi_f^{\text{bulk}} | \Delta | \Psi_i^{\text{bulk}} \rangle \right|^2 A_i(E) \tag{5}
\]
where the Fermi function on the right side is suppressed. Brief comments on forms 1-5 are appropriate in order to highlight the underlying approximations. Since forms 3-5 ignore the presence of the surface, it is very difficult to include effects of different surface terminations, and the associated distortions of bulk wavefunction which can be quite severe. In form 4, even near an ARPES peak from a specific initial state, other states will in general contribute a background upon being broadened due to their finite lifetimes. Forms 4 and 5 ignore the structure in the final state spectrum which will be seen below to be quite significant. We emphasize that the distinctions between the processes of excitation, transport and emission through the surface invoked by the three-step model (Eqs. 3-5) are artificial since the more satisfactory one-step formula (Eq. 2) does not admit such a decomposition.

The relevant technical details of our computations are as follows. In order to keep the problem manageable, the modulation of the lattice is neglected and the crystal structure of BISCO is assumed to be perfectly tetragonal; this still involves 30 atoms per unit cell, and a substantial extension of the earlier work on relatively simpler lattices. The crystal potential was first obtained self-consistently within the KKR scheme, and essentially yielded the well-known LDA-based band structure and Fermi surface of Bi2212; however, the actual initial spectra. The surface is assumed to terminate in the (k_x, k_y) plane for two different polarizations of the incident light where the initial state is held fixed at E_f. The color plots give computed intensities over a dense grid of k_||-values and display the two CuO_2 plane band sheets A and B in the band structure; these plots are representative of what could be measured in a suitably arranged constant-initial-energy angle-scanned (CIE-AS) ARPES measurement. 

Incidently, Figs. 3a-2 are not symmetric about a horizontal or vertical line through the center. In Figs. 3b and 3c, the light is incident along the M direction and the figure therefore is symmetric only about this diagonal line. In Fig. 3a, on the other hand, the light is polarized horizontally, and the intensities are symmetric around the M line; this symmetry becomes visible only when the figure is extended in the vertical direction to include a larger range of momenta.

The intensity associated with the outer plane band sheet A as one goes around the Fermi surface is shown in quantiative detail in Figs. 3a-3a. Since A is generally more intense than the inner plane band B, A is presumably more relevant in connection with the experimental data near E_f. For light polarized along the horizontal direction (Fig. 3a), we see that the intensity is large around α = 0° and decreases rapidly beyond α ≈ 20°. A 45° rotation of the polarization vector (Figs. 3b and 3c) induces substantial changes in the shape and magnitude of the intensity, and the appearance of a minimum at β or γ = 45°. The experimental points in Fig. 3 are in good overall accord with the measurements, some discrepancies around α and β ≈ 0° notwithstanding. Thus quite large observed variations (nearly an order of magnitude) in the emission intensity from different parts of the Fermi surface are mainly the consequence of matrix element effects.

Finally, Fig. 4 considers the photon energy dependence of the emission intensity of the spectral feature around M. The theoretical curve displays a prominent peak around 22 eV and indicates clearly that the final states in BISCO possess considerable structure which is neglected in approximations of Eqs. 5 and 6, especially when the matrix element in these equations is further replaced by a constant. The experimental points which show the presence of a broad peak centered around 21 eV are in good overall agreement when we keep in mind that errors of the order of a few eV’s in locating the final states are generally expected in the first principles band structures.

We emphasize that the agreement seen in Figs. 2-4 is robust to uncertainties inherent in such a comparison on
the theoretical as well as the experimental side. The experimental weights depend on the specific energy window used in their definition, and on whether or not a suitable background is subtracted. In this vein, the computed weights depend also of course on the specific values of the initial and final state damping parameters $\Sigma_i^{\prime\prime}$ and $\Sigma_f^{\prime\prime}$. In order to assess these effects, we have carried out extensive simulations using a variety of different values and energy-dependencies of $\Sigma_i^{\prime\prime}$ and $\Sigma_f^{\prime\prime}$, in addition to varying the real part of the initial state self-energy (in order to mimic correlation effects, even though the LDA framework underlies our computations), and find that the main features of the results of Figs. 2-4 are insensitive to such variations.

In summary, we have carried out extensive first-principles one-step ARPES simulations in Bi2212 wherein the photoemission process is modelled realistically by taking into account the nature of the initial and final state crystal wavefunctions as well as the multiple scattering effects in the presence of a specific surface termination. We focus on the nature of the ARPES feature arising from CuO2 plane bands, and consider in particular its spectral weight as a function of $k_{\parallel}$ as well as the energy and polarization of the incident photons. Large variations in the spectral weights predicted theoretically along three different high symmetry directions are in good accord with the corresponding measurements. A good agreement between theory and experiment is also seen with regard to changes in spectral weights with photon energy around the $k_{\parallel}$-point, as well as along the Fermi surface contours in the $(k_x, k_y)$ plane for two different polarizations. This study shows clearly that the remarkable observed changes in the ARPES spectral weights in Bi2212 are essentially a matrix element effect and that the importance of matrix elements should be kept in mind in analyzing the ARPES spectra in the high-Tc’s. Another notable implication of this work is that the integral (over energy) of the ARPES intensity does not yield the momentum density of the electron gas.

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a perfect tetragonal lattice.

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Figure captions

Figure 1: (a): Simulated ARPES intensity in BISCO for $k_\parallel$ varying between $\Gamma$ and $M$ (bottom to top) at $h\nu = 22\text{eV}$. (b) and (c) show computed variations in spectral weights (areas under peaks) of the two CuO$_2$ plane band features A and B with photon energy for different $k_\parallel$-values, based on a crystal potential without Bi-O hole pockets at $M$. A small value of the initial and final state damping parameters is used to highlight spectral features.

Figure 2: Theoretical weights obtained by integrating the $h\nu = 22\text{eV}$ ARPES spectra over the binding energy range of 0-500 meV are compared with the corresponding experimental results 11. Curves for the three symmetry lines (Brillouin zone in inset) are offset vertically for clarity; $k_\parallel$ is given in relative units such that the distance from $\Gamma$ to X, Y or M is defined to be unity for each direction. Theoretical values are normalized to match the experimental value at the maximum around $k_\parallel \approx 0.8$ in the $\Gamma M$ curve.

Figure 3: The color plots give simulated ARPES intensities for emission from $E_f$ at $h\nu = 22$ eV for two different polarizations (white arrows) of the incident light. The CuO$_2$ plane band sheets are marked A and B. The intensity (area under peak) of the outer plane band A is shown in (a1)-(c1) as a function of the angles $\alpha$, $\beta$ and $\gamma$. Experimental data after Ref. 11. Theory normalized around $\alpha \approx 0$ as shown in (a1).

Figure 4: Spectral weight (over a 500 meV window) of the feature at $M$-point is compared with the corresponding experimental 11 results as a function of the photon energy. Theory normalized to experiment around 21 eV.
Peak intensity along Fermi Surface (FS) [arb. units]

(a1) FS angle $\alpha$ [degrees]

(b1) FS angle $\beta$ [degrees]

(c1) FS angle $\gamma$ [degrees]

(a2) Experiment

(b2) Theory

(c2) Experiment

(a2) $\Gamma - M$

(b2) $\Gamma - M - M - X - Y$

(c2) $\Gamma$

$\alpha$, $\beta$, $\gamma$
Spectral weight [arb. units] vs. photon energy [eV].

- **Theory**
- **Experiment**