Role of site selectivity, dimensionality, and strong correlations in angle-resolved photoemission from cuprate superconductors

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New Journal of Physics 7 (2005) 140
Received 2 March 2005
Published 7 June 2005
Online at http://www.njp.org/
doi:10.1088/1367-2630/7/1/140

Abstract. We have carried out extensive first-principles computations of angle-resolved photoemission (ARPES) spectra from the cuprate superconductors within the general framework of the local density approximation (LDA). Selected results on Bi\textsubscript{2}Sr\textsubscript{2}CaCu\textsubscript{2}O\textsubscript{8} (Bi\textsubscript{2212}), La\textsubscript{2-x}Sr\textsubscript{x}CuO\textsubscript{4} (LSCO) and Nd\textsubscript{2-x}Ce\textsubscript{x}CuO\textsubscript{4} (NCCO) are presented and discussed. Our focus is on understanding how the underlying electronic structure is mapped via the complex process of photoexcitation into the observed ARPES intensities. Effects of the ARPES matrix element and its remarkable selectivity properties with respect to the energy and polarization of the incident photons in exciting a specific state and/or electrons from a particular site in the lattice are clarified. The importance of deviations from perfect two-dimensionality and the associated interlayer couplings in shaping the ARPES spectra of the cuprates is delineated. Our computations explain many salient features of the experimental spectra. Surprisingly, this agreement extends in some cases to the underdoped regime where strong electron correlations are obviously important. We discuss how the LDA-inspired tight-binding parameters can serve as a useful starting point for the treatment of strong coupling effects in the cuprates.
1. Introduction

The seminal insight into the phenomenon of photoexcitation of electrons by light—the photoelectric effect—provided by Einstein in 1905 [1], has led 100 years later to the development of a sophisticated technique for investigating electronic structure of solid state systems in the form of angle-resolved photoemission spectroscopy (ARPES), as attested by the collection of papers in this volume commemorating the 100th anniversary of Einstein’s discovery.3 In this paper, we discuss issues connected with the state-of-the-art modelling of the ARPES spectra from complex materials where the full Bloch character of the initial and final states with finite lifetimes in the crystal is treated in the presence of a specific surface termination. Such first-principles computations of the photointensity provide fundamental insight into the nature of the photoemission process and, from a more practical viewpoint, into the complexities associated with how the underlying electronic spectrum is mapped in the ARPES spectra. Our focus is on understanding the physics of the cuprates, and accordingly, we present and discuss selected results from the hole-doped high-$T_c$ compounds Bi$_2$Sr$_2$CaCu$_2$O$_8$ (Bi2212) and La$_{2-x}$Sr$_x$CuO$_4$ (LSCO), and the electron-doped compound Nd$_{2-x}$Ce$_x$CuO$_4$ (NCCO).

The key specific aspects of the ARPES spectra from the cuprates discussed in this paper are: (i) effects of the ARPES matrix element; (ii) selectivity properties of the ARPES matrix element with respect to the energy and polarization of the incident photons; (iii) effects of the dispersion of states as a function of $k_z$ due to interlayer (intercell) couplings; and (iv) modelling of strong correlation effects. It is appropriate that we comment briefly on these points by way of providing some background and context to the reader.

In analysing the ARPES spectra from the cuprates, one often makes the simplifying assumption that the photointensity measured in an ARPES experiment can be represented by the one-particle initial-state spectral function of the system. The complex process involved in the excitation of the initial state via the dipole matrix element into the outgoing photoemitted electron and the modifications associated with the transmission of the electron through the surface are thus severely approximated. First-principles computations, in fact, show clearly that the photointensity arising from a given initial state, in general, depends strongly on the energy

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3 See, e.g. the collected volumes of [2]–[8].
and polarization of the incident light as this state connects to different final states via the ARPES matrix element [9]. In particular, this matrix element displays remarkable energy/momentum ‘selectivity’, in that by tuning the photon energy and/or polarization one can selectively resolve one or another member of a closely spaced group of states [10, 11]. Bilayer splitting in Bi2212 presents an interesting case [9], [12]–[14]. It is likely that if the nature of the ARPES matrix element had been well-understood, the presence of bilayer splitting in Bi2212 could have been adduced even from the lower resolution ARPES experiments and the controversy that plagued the field for several years in this connection perhaps avoided. Furthermore, the ARPES matrix element is also ‘site-selective’ in that it can be dominated by transitions from a specific lattice site over a range of photon energies [15, 16]. For example, in Bi2212, emission from CuO2 related initial states for low-photon energies of up to about 25 eV from the Fermi level is mainly due to excitations in the $O_{p\rightarrow d}$ channel even though the wavefunctions of these states involve a substantial Cu admixture.

The analysis of ARPES spectra from a strictly two-dimensional (2D) material presents significant simplifications. However, even the most anisotropic cuprate systems possess some residual three-dimensionality, and the associated coupling between the CuO2 layers in different unit cells yields dispersion of states with $k_z$. In a 3D material, at a fixed value of $k$, $k_z$-dispersion leads to shifts in the position of the ARPES peak with photon energy ($h\nu$) [17]–[19]. The behaviour in a quasi-2D system is fundamentally different in that the spectral peaks will generally not shift with $h\nu$, but will instead display an irreducible linewidth which is a function of the in-plane momentum $k_\parallel$ [20, 21]. This reflects the loss of $k_z$-selectivity in the underlying photoexcitation process when the final-state damping, which is typically of the order of eVs, becomes much larger than the energy scale of the initial-state dispersion. This line broadening is induced by $k_z$-dispersion and does not have its origin in any scattering mechanism. This effect also manifests itself in the Fermi surface (FS) maps obtained via ARPES in that such maps display $k_z$-dispersion related broadenings, which can be quite substantial especially near the anti-nodal point $(\pi, 0)$. It is clear that effects of $k_z$-dispersion must be kept in mind in deducing scattering lifetimes from observed ARPES linewidths and in identifying signatures of stripes, pseudogaps, and other strong correlation phenomena in the ARPES spectra.

It should be noted that our first-principles computations implicitly assume the presence of large FSs and an electronic spectrum of the form given by the local density approximation (LDA), albeit with some renormalization, and are limited therefore generally to the description of the optimally doped and overdoped cuprates. The situation becomes more complicated with underdoping, especially in LSCO and NCCO, as a lower Hubbard band with an increasing spectral weight develops, which is not accounted for by the LDA. However, the ‘uncorrelated’ LDA-based spectrum provides the traditional starting point for investigating the effects of strong correlations and is thus of considerable value in exploring the physics of the cuprates more generally. We discuss this aspect with some results on NCCO.

This paper is organized as follows. The introductory remarks are followed in section 2 by an overview of the formalism employed in our one-step ARPES computations. Section 3 gives the relevant technical details. Section 4 delineates the strong effects of the ARPES matrix element on the spectral intensities. Section 5 takes up the issue of selectivity properties of the ARPES matrix element. Section 6 considers how residual three-dimensionality plays out in the ARPES spectra. Section 7 briefly discusses results on LSCO and NCCO and the treatment of strong correlations effects using LDA-inspired tight-binding parameters. Finally, some concluding remarks are made in section 8.
2. Formal considerations

In order to highlight some of the formal issues involved, we begin by recalling the expression for photointensity from initial states at energy $E$ excited by photons of energy $\hbar \omega$ based on the golden rule [22]

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

(1)

where $\Psi_i$ and $\Psi_f$ denote the wavefunctions of the initial and final states of the semi-infinite solid, respectively, and $\Delta = e \hbar / 2mc(\mathbf{p} \cdot \mathbf{A} + \mathbf{A} \cdot \mathbf{p})$ is the interaction Hamiltonian in terms of the electron momentum operator $\mathbf{p}$ and the vector potential $\mathbf{A}$ of the photon field.

In the one-step model used in the present computations, equation (1) is manipulated into the form [23]–[25]

$$I(k_\parallel, E, \hbar \omega) = -\frac{1}{\pi} \text{Im} \langle k_\parallel | G_2^+ (E + \hbar \omega) \Delta G_1^+ (E) \Delta^\dagger G_2^- (E + \hbar \omega) | k_\parallel \rangle.$$  

(2)

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

We note that in the so-called three-step model of photoemission, the first step approximates the matrix element in equation (1) by replacing the wavefunctions $\Psi_i$ and $\Psi_f$ for the semi-infinite solid by the corresponding bulk Bloch wavefunctions. This yields the photointensity within the solid [26]

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

(3)

which we have cast in terms of the one-particle spectral functions of the initial and final states ($A_i$ and $A_f$). The processes of transport of the photoexcited electron to the surface (step two), and the emission of the electron from the solid across the surface (step three) are then considered separately. Further simplification is obtained by assuming that the system is strictly 2D and that the final states form a structureless continuum. Under these conditions equation (3) reduces to

$$P(E, \omega) \sim \sum_i |\langle \Psi_f^\text{bulk} | \Delta | \Psi_i^\text{bulk} \rangle|^2 A_i (E)$$

(4)

in terms of just the spectral functions $A_i (E)$’s of the initial states. Furthermore, for a single-band solid, one obtains

$$P(E, \omega) \sim |\langle \Psi_f^\text{bulk} | \Delta | \Psi_i^\text{bulk} \rangle|^2 A_i (E),$$

(5)

where the Fermi function on the right side is suppressed.

It is important to understand the nature of the photointensity described by equations (1)–(5) and the underlying approximations. Forms (3)–(5) ignore the presence of the surface. This makes it very difficult to include effects of different surface terminations [27] and the associated distortions of the bulk wavefunctions, which can severely modify the spectra in a
surface-sensitive probe such as the ARPES. 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 (equations (3)–(5)) are artificial since the more satisfactory one-step formula (equation (2)) does not admit such a decomposition. It will be seen that the right-hand side of equation (5), which is proportional to the one-particle bulk spectral density of a single initial-state band, involves significant approximations and assumptions. The extent to which such a description differs from the spectral intensity given for example by equation (2), may be referred to as the ‘matrix element effect’ in the ARPES spectrum. Some insight into the nature of the ARPES spectra can be gained through the analysis of the dipole matrix element $\langle \Psi_{i}^{\text{bulk}} | A | \Psi_{i}^{\text{bulk}} \rangle$ for bulk transitions, and such an approach is invoked in the discussion of section 5 below.

3. Computational details

The relevant technical details of the computations may be noted as follows. As a prelude to the ARPES intensity computations, the fully self-consistent electronic structures of the undoped parent compounds La$_2$CuO$_4$ (LCO), Nd$_2$CuO$_4$ (NCO), and Bi2212 were obtained within the LDA by using the well-established all electron Kohn–Korringa–Rostoker (KKR) Green function methodology [28, 29]; our band structures and FSs are in reasonable accordance with the published data [9, 21, 30]. Following common practice, the metallic state obtained by doping is assumed to be described by these LDA generated metallic band structures. The effect of O doping or of La/Sr or Nd/Ce substitution is mainly to adjust the electron concentration and, therefore, we have modelled the doped compounds at any given doping level $x$ by implicitly invoking a rigid band filling of the band structure with the appropriate number of electrons per unit cell. The crystal potentials used are the same as those employed in our previous studies of Bi2212, LSCO and NCCO [9, 15, 21, 30].

The ARPES intensities were computed within the one-step photoemission formalism, assuming a BiO or a LaO layer terminated [31] surface for Bi2212 and LSCO, respectively; see, e.g., [9, 15, 20, 21] for details. The results on NCCO are for the CuO$_2$–Nd–O$_2$–Nd terminated surface, which generally appears to be the most representative of the experimental spectra [32]. The Nd-terminated surfaces in NCCO show the presence of surface-induced states, which have so far not been observed. In this vein, the actual potential used in computations on Bi2212 was modified slightly in that the Bi–O pockets around the $\overline{M}$-point are lifted above the Fermi energy to account for their absence in the experimental spectra.\textsuperscript{4} Although the LDA-based band theory framework is implicit, appropriate energy-dependent self-energy corrections can be included straightforwardly in our computations, providing at least some handle on correlation effects in the spectra. The calculations properly model the photoemission process and include the crystal wave functions to describe the initial and final states in the presence of the surface, and take account of the associated dipole matrix element and its dependences on the energy and polarization of the photon. The finite lifetimes of states can be simulated by giving an imaginary part to the

\textsuperscript{4} Such an ad hoc adjustment of the LDA potential is of course not very satisfactory and a more robust understanding of how the Bi-O pockets predicted by the LDA get lifted above the Fermi energy in the Bi-compounds needs to be developed.
self-energies of the initial ($G_1$) and final ($G_2$) state propagators in equation (2), which are denoted by $\Sigma_i''$ and $\Sigma_f''$, where the subscript $i$ ($f$) refers to the initial (final) state. The LDA band structures also provide a starting point for developing the tight-binding model Hamiltonians for treating strong correlation effects in the cuprates as discussed in section 7 below with the examples of LSCO and NCCO.

4. Matrix element effects

Figure 1 illustrates how the ARPES matrix element modifies the imprint of the FS observed via photoemission. Here, the computed photointensity for emission from the Fermi energy is shown over a fairly large region in the $(k_x, k_y)$ plane in Bi2212, LSCO and NCCO at a photon energy of $h\nu = 21$ eV for two different polarizations of the incident light. If the ARPES matrix element were a constant, the FS maps of figure 1 would display a uniform intensity along the FS contours. This however is far from being the case, indicating the strong energy and polarization dependences of the matrix element. For example, in Bi2212 in (a), the anti-bonding FS sheet (contour centred around $\Gamma$) is quite intense around the anti-nodal points $(\pi, 0)$ and $(-\pi, 0)$, but possesses little intensity along the nodal direction. When the polarization vector of light is rotated by 45° in (b), this sheet develops considerably increased intensity at the other two M-points $(0, \pi)$ and $(0, -\pi)$. Other differences between the behaviours of the bonding

Figure 1. Computed FS maps for emission from the Fermi energy in Bi2212, LSCO and NCCO at $h\nu = 21$ eV for two different polarizations of light. White arrows give the direction of the incident light which is p-polarized. White squares mark boundaries of the 3D Brillouin zone in the $k_z = 0$ and $k_z = 2\pi/c$ planes.
and anti-bonding FS sheets for either polarization or between the two different polarizations of (a) and (b) can be observed by the reader. Similar remarks are also applicable to the results in (c)–(f) for LSCO and NCCO, where the FS is of course single-sheeted. Differences in the sizes and shapes of the FSs in Bi2212, LSCO and NCCO reflect those in the underlying band structures. The spectra are further influenced by the extent to which the band structure deviates from being perfectly 2D—a point to which we return in section 6 below.

The remarkable degree to which the theoretically predicted matrix element effects are in accordance with ARPES measurements is shown by the spectra of figures 2 and 3 on Bi2212, a system on which much data has been collected by various groups across the world. Computations in figures 2 and 3 match the experimental conditions of photon energy, polarization and resolution. The computed spectra of figure 2(b) were in fact published [9] well before the high-resolution data [33] in figure 2(a) from a Pb-doped sample (to suppress the superlattice modulation) became available. The bilayer split bonding and anti-bonding FS sheets are seen clearly in the data. Moreover, the detailed \( k_\parallel \)-dependences of the associated intensity variations are in substantial agreement with the first-principles calculations. In particular, the anti-bonding FS sheet, which was the subject of great controversy for a number of years is seen to give a broad patch of intensity near the \( M(\pi, 0) \)-point, making it difficult to resolve its presence in the earlier lower resolution experiments.

Figure 3 considers ARPES spectra taken at two different photon energies over a wide region of the \((k_x, k_y)\) plane from a Bi2212 sample without Pd-doping, so that the superlattice modulation is not suppressed [14, 34]. Here, the effects of the superlattice modulation and the orthorhombic distortion of the lattice are simulated in the calculations by taking the computed spectrum for the pristine system and superposing on it the same spectrum with appropriate weights after it has been suitably translated along various reciprocal lattice vectors. First-principles calculations on the modulated Bi2212 lattice would of course be more satisfactory, but such computations will be quite demanding given the very large unit cell involved. At \( h\nu = 21 \text{ eV} \) in (c) and (d), the primary spectral features arising from the underlying tetragonal lattice are quite strong compared to those in (a) and (b).
to the secondary features associated with the lattice modulations. In contrast, at $h\nu = 32$ eV in (a) and (b), the relative intensities of the primary and secondary features are more similar. As a result, the FS map at 32 eV looks very different from that at 21 eV, even though both sets of computed maps are based on identical underlying band structures and FSs. Figure 3 emphasizes how the photon energy-dependence of the matrix element can highlight various physical effects in the ARPES spectrum under different experimental conditions.

5. Selectivity properties of the matrix element

The ARPES matrix element displays strong selectivity in its ability to couple to states of different symmetries through its energy dependence. It can also show ‘site selectivity’ properties in the sense that in a given photon energy range it can couple, preferentially, to excitations from a specific site in the complex unit cell. We discuss these aspects now with reference to figures 4 and 5.

Figure 4 compares the computed photointensity from the bonding and the anti-bonding portions of the FS in Bi2212 as a function of the photon energy at three different values of $k_\parallel$. Large variations in the intensity are seen for all $k_\parallel$-values over the entire range of photon energies extending to 80 eV in the figure. For example, along the nodal direction in (a), at 60 eV the bonding state (blue dashed line) possesses larger cross-section than the anti-bonding state (red solid line). At 47 eV, on the other hand, the intensity of the anti-bonding band is much larger than that of the bonding band. This predicted amplification of the anti-bonding signal at 47 eV persists...
Figure 4. Computed ARPES intensity from the bonding (blue dashed lines) and anti-bonding (red solid lines) portions of the FS of Bi2212 for photon energies extending to $h\nu = 80$ eV. Three different $k_{||}$-points on the FS are considered. (a) Along the nodal direction, (b) along the direction at an angle of $\theta = 22.5^\circ$ with respect to the $\Gamma - X$ line, and (c) along the anti-nodal direction. Light is assumed to be unpolarized.

as a function of $k_{||}$ in (a)–(c) and has been exploited in [10] to study the anti-bonding states and the doping dependence of the bilayer splitting in Bi2212. By comparing (a)–(c), it will be seen that, in general, at any given photon energy, the intensity of the bonding or the anti-bonding state varies substantially with $k_{||}$. Nevertheless, one can identify photon energies where the $k_{||}$- and/or the binding energy dependence is weak. Such special photon energy windows will be best suited for extracting self-energy information from the ARPES spectra in proposals like those of [35] aimed at obtaining the spectral function of the bosonic glue mediating superconductivity in the cuprates [11].

Turning to the issue of site selectivity of the ARPES matrix element, insight in this regard can be obtained by considering the momentum matrix element $\langle \Psi^\text{bulk}_f | \mathbf{p} | \Psi^\text{bulk}_i \rangle$ in equation (3), which connects the bulk initial and final states $\Psi^\text{bulk}_i$ and $\Psi^\text{bulk}_f$ and the associated intensity

$$I = |\mathbf{A} \cdot \langle \Psi^\text{bulk}_f | \mathbf{p} | \Psi^\text{bulk}_i \rangle|^2.$$  (6)
As discussed in [15, 16, 36], equation (6) possesses the advantage that it can be decomposed straightforwardly into contributions from various atomic sites in the unit cell. Although form (6) does not account properly for the processes of transport and ejection of the photoelectron or for the effect of the distortion of the bulk wavefunctions due to the presence of the surface, it nevertheless provides a handle on the intrinsic strength of the photoexcitation process.

Figure 5 presents the intensity \( \tilde{I} \) based on equation (6) in LSCO for emission from three different points on the FS as a function of the photon energy, together with contributions to the total intensity from the Cu \( d_{x^2-y^2} \) and O\(_{\text{Cu}}\) \( p_x/p_y \) orbitals in the CuO\(_2\) planes. Contributions from other sites (not shown) are not always negligible as seen by comparing the total intensity curves with the sum of the two other curves. Incidentally, the O-contribution at photon energies below \( \sim 20 \text{ eV} \) in (a) is slightly higher than the total intensity—this reflects interference effects that come into play between contributions from various sites when the absolute value of the matrix element is taken in equation (6). Figure 5 shows that at low-photon energies (\( h\nu \lesssim 25 \text{ eV} \)) the Cu contribution to the photointensity is quite small and that the emissions from just the O\(_{\text{p}}\) \( p_x/p_y \) orbitals dominate the spectrum, despite the fact that the initial states involved possess a substantial Cu character. At photon energies above 35 eV, the Cu contribution is generally larger than that of the O sites. The relative Cu/O intensity varies substantially with \( k_{\|} \) and \( h\nu \). Notably, in Bi2212, [16] has shown that at low-photon energies, the emission from the bonding and anti-bonding states in the vicinity of the Fermi energy is also dominated by the O\(_{\text{p}}\) \( p_x/p_y \) orbitals throughout the Brillouin zone.

Our prediction that ARPES can preferentially sample Cu or O states by tuning the photon energy suggests novel possibilities for exploiting energy-dependent ARPES spectra for probing initial-state characters. Thus, even though ARPES cannot see the upper Hubbard band (and hence cannot measure \( U \) directly), it should be possible to adduce \( U \) by measuring the relative Cu/O character of states along the lower Hubbard band, and comparing the results to predictions of appropriate model computations. A determination of the doping dependence of \( U \) would help to check recent theoretical results and predictions [30], and give insight into the Zhang–Rice mechanism [37] in the cuprates.

6. Quasi-3D effects

We discuss now how \( k_z \)-dispersion will manifest itself in the ARPES spectrum of a quasi-2D material with the simulations [20] of figure 6 on Bi2212, which show the evolution of the lineshape

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5 Our discussion in terms of Cu and O orbital contributions is based on the fact that equation (6) formally admits decomposition into contributions from different sites and angular momentum channels. However, we should keep in mind that in a physical sense the assignment of electrons as belonging to different sites is not as clear in a solid as it is in an atomic system because tails of wavefunctions on a given site in a solid will in general overlap other nearby sites. Despite this subtlety, we expect our conclusion that O contribution dominates the cross-section at low photon energies to be robust. The reason is that excitations from Cu \( d \) states on the Cu sites are already small at low photon energies and therefore one would not expect the tail of this small term to show up stronger on neighbouring sites.

6 Note that atomic cross-sections are of course used routinely by spectroscopists to tune experiments to specific atoms. The novelty of our analysis concerns the exploitation of more subtle solid state effects, which can allow one to zoom in for example on the antibonding portion of the Fermi surface of Bi2212 using 47 eV photons as pointed out in connection with figure 4.
Figure 5. Computed intensity $\tilde{I}$ based on the strength of the dipole matrix element (see equation (6)) from the FS of LSCO for photon energies extending to $h\nu = 70$ eV (solid lines). Individual contributions from Cu $d_{x^2-y^2}$ (dashed lines) and $O_{Cu} p_x/p_y$ orbitals (dotted lines) in the CuO$_2$ planes in the lattice are shown. The three different $k_\parallel$-points considered in (a)–(c) are as in the caption of figure 4.

Light is polarized along the [110]-direction.

of the energy distribution curves (EDCs) as the broadening of the final state introduced via the imaginary part the self-energy, $\Sigma''$, is increased from a very small value in (a) to a realistic value in (c). 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 the transmission of the electron across the surface and the initial and final states can then 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 $\Sigma'' = 100$ meV in (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
Figure 6. Simulated ARPES EDCs in Bi2212 for a series of photon energies ($h\nu = 25$ to $25.7$ eV) at a fixed $k_{||} = (0.34, 0.09)2\pi/a$-point using three different values of the final-state broadening given by the indicated imaginary part of the self-energy $\Sigma''_f$ (after [20]). 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). Spectra have not been convoluted with the Fermi function.

The 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). 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 the $k_z$-direction because outside of this interval there are no initial-state electrons capable of absorbing the photon.

We emphasize that the irreducible linewidth of the ARPES peaks induced via $k_z$-dispersion does not have its origin in any scattering mechanism. It is reflected also in the FS maps obtained by the ARPES. Figure 7 delineates this aspect of the ARPES spectra with results on LSCO and Bi2212. Figure 7(a) shows the computed cross-section of the standard hole-like FS of optimally doped LSCO in the $k_z = 0$ plane. This cross-section changes with $k_z$ due to the residual three-dimensionality of the bands. Figure 7(b) collects such cross-sections for $k_z$ values varying over the $0$ to $2\pi/c$ range. In a strictly 2D system, various cross-sections in (b) will be identical and collapse into a single FS contour. Here, however, there are substantial variations, especially in the anti-nodal regions. The nodal region on the other hand, where the $k_z$-dispersion is small, undergoes little change. The projection of the FS on to the $(001)$-plane in (b) gives the region of allowed transitions in ARPES [20, 21]. In other words, the FS map measured in ARPES is intrinsically
Figure 7. Computed cross-sections of the FS of LSCO at $x = 0.15$ (top row) and Bi2212 (bottom row). (a) FS cross-section in LSCO in the $k_z = 0$ plane; (b) cross-sections of the 3D FS for various values of $k_z$ projected on to the (0 0 1)-plane; (c) Effect of integrating the constant energy maps like those of (b) over a binding energy range of ±15 meV around the $E_F$; (d)–(f) same as (a)–(c), except that these panels refer to Bi2212; B(A) in (d) refers to the bonding(anti-bonding) FS.

broadened due to the effect of $k_z$-dispersion so as to be contained within the boundaries set by the projection of (b), aside from the effects of resolution and the finite lifetimes of the initial and final states. Note that the FS maps in typical ARPES experiments on the cuprates are obtained by integrating the data over a binding energy range of 5–30 meV around the $E_F$. The effect of the integration process is simulated in (c) by integrating the spectra over a binding energy range of ±15 meV for the present illustrative purposes. We see that the integration has little effect on the FS map in LSCO.

Figures 7(d)–(f) consider the case of Bi2212. The effect of $k_z$-dispersion in Bi2212 is smaller than in LSCO since the $c$-axis in Bi2212 is longer and the relatively empty regions separating the Bi–O layers contribute to a reduced intercell hopping between the CuO$_2$ bilayers. In Bi2212, the intercell hopping between the CuO$_2$ bilayers, which is responsible for the $k_z$-dispersion, should be distinguished from the intracell hopping which splits the CuO$_2$ bands into the bonding and anti-bonding pair of bands. Figure 7(e) indicates that the bonding FS sheet suffers little effective broadening due to $k_z$-dispersion, while the effect on the anti-bonding FS is larger and possesses a characteristic $k_{∥}$ dependence. The process of integrating the computed spectra over the ±15 meV binding energy range has little effect on the bonding FS, but the anti-bonding FS is broadened.

The integration range in band computations appropriate for modelling this effect will typically be a factor of 2–3 larger than the experimental range since the band dispersions given by the LDA are observed to be renormalized (reduced) by a factor of 2–3 in the cuprates.
substantially as seen in (f). These differences between LSCO and the two FS sheets of Bi2212 are due in part to differences in the nature and placement of the Van Hove singularity (VHS) in the band structure around the \((\pi, 0)\) point in relation to the \(E_F\) in the two compounds.

7. Strong correlation effects

Strong electron correlation effects not accounted for by the LDA play an important role in the physics of the cuprates, especially in the underdoped regime. For example, in the lightly doped NCCO and LSCO, there is evidence for split Hubbard bands [38], while in LSCO mid-gap states appear with an increasing spectral weight on doping [39, 40]. Thus our analysis so far which implicitly assumes the presence of large FSs and LDA-type conventional electronic spectrum, even if with some renormalization, is generally limited to the description of the optimally doped and overdoped systems. Nevertheless, the LDA-based ‘uncorrelated’ bands often serve as the traditional starting point for addressing strong coupling effects via appropriately parametrized tight-binding model Hamiltonians [41]. This section discusses some results on LSCO and NCCO where LDA computations provide insight and/or parameters useful for understanding the behaviour of the correlated system.

8 We thank X J Zhou, T Yoshida, A Fujimori, Z Hussain and Z X Shen, for providing us with the original experimental data of [42] so that the data could be shown in the colour scheme of figure 8.
Figure 8 compares the calculated\(^9\) and experimental [42] FS maps for an underdoped LSCO sample (\(x = 0.06\)) close to the metal–insulator transition. The theoretical spectrum in (a) includes the effect of the ARPES matrix element and properly simulates the experimental conditions of polarization and energy of the incident light. The pink lines mark the regions of allowed transitions due to the effect of \(k_z\)-dispersion. Note that the experimental intensities in (b) are confined reasonably well within the limits of these pink lines, suggesting that much of the observed broadening in the experimental FS map may be tied to \(k_z\)-dispersion. On comparing theoretical and experimental intensities, good accord is seen around the anti-nodal \(M(\pi, 0)\) region, where the two sheets of the hole-like FS are well-separated in both computations and measurements. Similarly, when going further away from the \(M\)-point to the higher BZs, the two branches of the FS, above and below the \(k_z\)-axis, are correctly reproduced in the computation, i.e. the FS branch below the \(k_z\)-axis shows high intensity, whereas the branch above the \(k_z\)-axis is less intense. Notably, the computed intensity along the nodal direction (\(\Gamma - X\)) in the first BZ is strong in (b), whereas in the experimental data this intensity is absent. This discrepancy could indicate strong correlation physics, although this point requires further analysis. Interestingly, in the case of Bi2212, the experimental and theoretical intensities—including the nodal direction—are generally in good agreement (see, e.g., [9, 14, 33]). The good agreement between the experimental and theoretical FS maps in figure 8 is remarkable since, as already noted, strong coupling effects beyond the LDA are well known to be present in this case. Therefore, it is most sensible to view the FS map of figure 8 to be merely the residue of the mid-gap band near \(E_f\). Its close correspondence to the theoretical LDA-based calculation in figure 8 suggests that some sort of a generalized Luttinger theorem continues to apply even in the presence of strong coupling effects [21, 43].

The evolution of electronic states from a Mott insulator into the superconductor appears to follow different routes depending on electron versus hole doping. In the hole-doped LSCO, as noted above, prominent mid-gap states with an increasing spectral weight appear on doping. In contrast, in NCCO the doped electrons seem to go directly into the upper Hubbard band, and the doping leads to a gradual collapse of the Mott gap. Figure 9 considers the evolution of the FS of NCCO over a wide range of doping levels. The experimental FS maps in the bottom row are from the ARPES measurements of [38]. The theoretical FS maps in the top row are based on a four-band TB model Hamiltonian.

In an earlier publication, we demonstrated that the evolution of the Mott gap with doping in NCCO could be described by a one-band TB model using a doping-dependent Hubbard \(U\) [30]. However, the appropriateness of a one-band model can be questioned since the lower Hubbard band in the physical system is in fact a charge transfer band of predominantly O 2p character. Accordingly, we have reanalysed the data using a four-band TB model based on the Cu s, Cu \(d_{x^2−y^2}\) and the two O p\(_x/p_y\) orbitals in the CuO\(_2\) plane [44]. The resulting Hamiltonian is

\[
H = \sum_j \Delta_s s^\dagger_j s_j + \sum_j \Delta_d d^\dagger_j d_j + \sum_{\langle i,j \rangle} t_{sp} [s^\dagger_j p_i + (\text{c.c.})] + \sum_{\langle i,j \rangle} t_{dp} [d^\dagger_j p_i + (\text{c.c.})] + \sum_j U n_{d_{\uparrow} j} n_{d_{\downarrow} j} + \sum_i U_p n_{p_{\uparrow} i} n_{p_{\downarrow} i},
\]

where the energy zero is implicitly set at the on-site energy of the O p orbital. \(\Delta_s\) and \(\Delta_d\) give on-site energies of the Cu s and d orbitals, respectively, in relation to that of the O p orbital. \(t_{sp}\) and

\(^9\) Following the experimental procedure of integrating over a narrow energy window, the projected FS in figure 8 is integrated over \(E_f = \pm 5\) meV.
Figure 9. Top row: theoretical FS maps in NCCO for different doping levels \( x \) based on the four-band Hubbard model discussed in the text. Colour scheme is the same as in figure 8. Bottom row: corresponding experimental maps after [38].

\( t_{dp} \) are the copper-oxygen hopping parameters, which are limited to the nearest neighbour terms; \( U \) and \( U_p \) are the Hubbard parameters for Cu d and O p orbitals; and \( n_{dj} = d_j^\dagger d_j \) and \( n_{pi} = p_i^\dagger p_i \) are the corresponding number operators. We treat the model self-consistently at the mean-field level where we fit the experimentally observed doping dependent band dispersions near the Fermi energy. The final parameter values are found to be: \( t_{sp} = 1.4 \), \( t_{dp} = 0.9 \), \( \Delta_s = 6 \), \( \Delta_d = 0 \), \( U_p = 1.5 \) and \( U = 3.85 \) eV at zero doping (\( U_p \) is doping independent). Our values of \( t_{sp} \) and \( t_{dp} \) are renormalized (smaller) roughly by a factor of \( Z = 0.6 \) compared to those of [44]. Notably, the doping dependence of \( U \) as well as the staggered magnetization, \( m_Q = (n_{d\uparrow} - n_{d\downarrow})/2 \), obtained in the present four-band model is similar to the earlier results based on the one-band model [30].

Figure 9 compares the FS maps as a function of doping computed within the four-band model with the corresponding experimental results. The agreement between theory and experiment is seen to be good. For the lightly doped insulator at \( x = 0.04 \), both the theoretical and the experimental FS maps show the appearance of small electron pockets centred on the \((\pi, 0)\) and \((0, \pi)\) symmetry points. As the doping level increases to \( x = 0.10 \), these pockets begin to look squarish in shape with half of the square displaying relatively little intensity; also, weak intensity starts to appear around the magnetic zone boundary defined by the diagonal line joining the \((\pi, 0)\) and \((0, \pi)\) points. With further doping at \( x = 0.15 \), a new pocket around \((\pi/2, \pi/2)\) is clearly formed and the three pieces of the FS together begin to resemble a single large hole sheet centred at the \((\pi, \pi)\) point, much like the prediction of the LDA theory.

8. Conclusions

We have shown with reference to selected results on LSCO, NCCO and Bi2212 that in order to properly analyse and interpret ARPES spectra from the cuprates, effects of the ARPES matrix element and its strong selectivity properties with respect to the energy and polarization of
the incident light must be taken into account. Moreover, interlayer hopping and the resulting dispersion of states with $k_z$, which has been neglected in much of the existing literature on the cuprates, plays a key role in shaping the ARPES spectra and induces $k_\parallel$-dependent linewidths in spectral peaks, which do not have their origin in a scattering mechanism. The relatively simple LDA-based picture is shown to capture many salient features of the ARPES spectra in the cuprates. Surprisingly, this agreement extends into the underdoped regime in LSCO, suggesting that some sort of a generalized Luttinger theorem continues to hold even in the presence of strong coupling effects beyond the LDA. We conclude that with attention to the nature of the photoemission process and how the characteristics of the initial states are mapped in the observed intensities, it should be possible to significantly extend the reach of ARPES as a spectroscopic window on the electronic structure and self-energies in wide classes of complex materials.

Acknowledgments

We acknowledge technical assistance of Mr Hsin Lin. This work is supported by the US Department of Energy contract DE-AC03-76SF00098, and benefited from the allocation of supercomputer time at NERSC, Northeastern University’s Advanced Scientific Computation Center (ASCC), and the Institute of Advanced Computing (IAC), Tampere.

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