

A Novel Direct Method of Fermi Surface Determination Using Constant Initial Energy Angle-Scanned Photoemission Spectroscopy

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(Received 26 April 1996)

We show that a constant initial energy, angle-scanned (CIE-AS) photoemission spectrum for emission from the Fermi energy (E_F) contains Fermi surface (FS) signatures which originate from density of states type *indirect* transitions. Such previously unrecognized FS features in a CIE-AS spectrum would provide a robust and straightforward means of determining Fermi surfaces. Furthermore, the associated photointensity should yield a new window on k_{\perp} dispersion related issues in materials. Extensive simulations of CIE-AS spectra from low index faces of Cu are presented within the framework of the one-step photoemission model in order to delineate the nature of these new spectral features. [S0031-9007(96)01334-8]

PACS numbers: 71.18.+y, 79.60.Bm

The value of a constant initial energy type angle-scanned photoemission spectrum has been highlighted recently by the work of [1] which considers fermiology related issues in Bi2212 [2]. The idea is to arrange the parameters of the experiment in such a way that emission from the vicinity of the Fermi energy (E_F) is measured over a fairly dense grid of k_{\parallel} values. It is then argued that rapid variations in the observed intensity herald quasiparticle excitations dispersing across E_F as a function of k_{\parallel} , allowing insight into Fermi surfaces of materials and the mapping of 3D band structures. However, photoemission is intrinsically a complex process which involves energy and crystal momentum conservation in the presence of finite initial and final state lifetimes, and the matrix element for emission is, in general, influenced strongly by the final state wave function. It is important therefore to establish clearly the fundamental nature of features manifest in a constant initial energy, angle-scanned (CIE-AS) spectrum in terms of the underlying Fermi surface (FS).

With this motivation we have carried out an extensive theoretical study of the CIE-AS spectrum from low index faces of Cu as a test case. Here we focus on a novel aspect of our results. We find that the CIE-AS spectrum not only displays the usual FS features arising from direct transitions, but also contains FS signatures originating in density of states (DOS) type indirect transitions. The latter provide a surprisingly simple imprint of the underlying FS in sharp contrast to the direct transitions. Further, it turns out that the associated photointensity is proportional to the 1D DOS of initial states as a function of k_{\perp} and should thus provide a new window on issues related to k_{\perp} dispersion in materials more generally. To our knowledge, the importance of the aforementioned DOS type FS features in a CIE-AS spectrum has not been recognized previously in the literature.

All the computations reported in this article are based on the one-step model of photoemission wherein the multiple scattering effects on the initial as well as the

final electronic states are treated properly in the presence of a semi-infinite surface [3]. The crystal potential of Cu, taken from Ref. [4], is well known to yield a reasonable electronic spectrum of Cu. Finite lifetime effects are modeled by giving a suitable imaginary part to the self-energies of the initial and final state propagators. Much of the relevant methodology is well documented (see, e.g., Ref. [5]) and requires little elaboration.

The essence of our new results may be illustrated with reference to Fig. 1 which shows the computed CIE-AS spectra from Cu(001) for three different values of the imaginary part Σ_f'' of the final state energy. Σ_f'' determines the mean free path ℓ_f of the photoemitted electrons via the relation [6], $\ell_f = \sqrt{2E}/2\Sigma_f''$. $\Sigma_f'' = 2$ eV, Fig. 1(b), is representative of the physical situation. Hypothetical cases of much smaller and larger values of Σ_f'' are also considered as these results give insight into the origin of various spectral features. For $\Sigma_f'' = 16$ eV, Fig. 1(c), ℓ_f is very small, and the emitted electrons essentially emanate from the topmost layer, while $\Sigma_f'' = 0.1$ eV, Fig. 1(a), implies $\ell_f = 200$ Å and the photoelectrons sample ~ 110 layers. The initial state is assumed in all calculations to possess a small damping of 0.1 eV as this state is generally expected to suffer little scattering.

Figure 1(a) shows the presence of a number of features labeled A–D; A is seen to be the most intense, and the intensities of B–D are about 50 times smaller. Since ℓ_f in this case is rather large, the surface contribution is negligible, and all spectral features are expected to arise from *direct* transitions in the underlying band structure. We have verified that this is indeed the case by extensively plotting energy bands as a function of k_{\perp} for many different ranges of appropriately chosen k_{\parallel} values. For a direct transition, k_{\perp} must, in general, vary with k_{\parallel} in order to satisfy the energy conservation constraint, $E_{\text{final}} = E_{\text{initial}} + h\nu$. Therefore, features in Fig. 1(a) give FS dimensions along complicated contours in the momentum space, rather than any simple cuts through

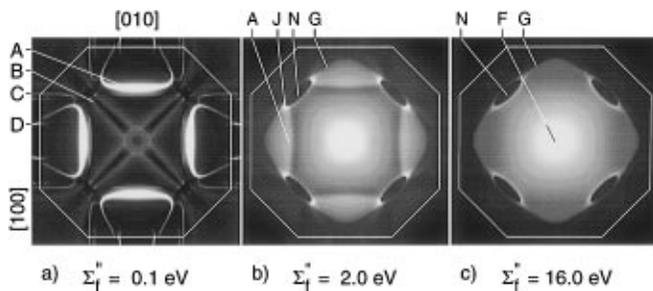


FIG. 1. Theoretical CIE-AS spectra from Cu(001) for 21.2 eV unpolarized light using three different values of the final state damping. (a) and (c) refer to artificially low (0.1 eV) and high (16 eV) values of damping, respectively; the damping (2 eV) in (b) is representative of the physical situation. Letters A, B, etc., denote various spectral features discussed in the text. The intersection of the 3D Brillouin zone with the (001) plane passing through the zone center is shown. Whites denote regions of high spectral intensity.

the FS. It is noteworthy that if the final states were assumed to be free electrons (within the crystal), and matrix elements were to be neglected, we will obtain only the feature A in Fig. 1(a) with a uniform intensity extending to the Brillouin zone boundary. Dramatic variations in the intensity of A as well as presence of features B–D are striking consequences of the fact that matrix elements are included in our computations, together with the effects of the crystal potential on the final states.

It is useful to consider Fig. 1(c) next which simulates the situation for a small value of ℓ_f . Figure 1(c) is obviously reminiscent of the FS of Cu, and displays striking changes compared to the spectrum of Fig. 1(a). Much of the sharp structure A–D of Fig. 1(a) is washed out; an essentially isotropic bump F emerges near the zone center, in addition to the appearance of the large closed contour G and the elliptical regions N.

It is clear that Fig. 1(c) differs intrinsically from Fig. 1(a). In order to gain a handle on the nature of these changes, we recall first that the spectral photointensity in the one-step model used here is given by [3]

$$I = -\frac{1}{\pi} \text{Im} \langle k_{\parallel} | G_f^+ \Delta G_i^+ \Delta^{\dagger} G_f^- | k_{\parallel} \rangle, \quad (1)$$

where G_f and G_i are the one-electron Green functions at the final state (E) and the initial state ($E - \hbar\omega$) energies, respectively; the superscript + (–) indicates forward (backward) propagation. Δ is the photon field operator. Equation (1) can be manipulated to obtain a form for the number of excited angle-resolved photoelectrons [7] $N(E, \hbar\omega)_{k_{\parallel}}$ more suitable for the present purpose as

$$N(E, \hbar\omega)_{k_{\parallel}} \sim \int dk_{\perp} |M_{if}|^2 A_i(E - \hbar\omega - E_i(k_{\perp})) \times A_f(E - E_f(k_{\perp})). \quad (2)$$

Here, $M_{i,f}$ denotes the matrix element between the initial and the final state. A_f (or A_i) is the spectral function for the final (or initial) state which may be modeled by a

Lorentzian for a state possessing a finite damping,

$$A_f(E - E_f(k_{\perp})) = \frac{\pi^{-1} \Sigma_f''}{[E - E_f(k_{\perp}) - \Sigma_f']^2 + (\Sigma_f'')^2}, \quad (3)$$

with a similar expression for the spectral function $A_i(E - \hbar\omega - E_i(k_{\perp}))$ [8].

If both the initial and the final states are assumed to be sharp (i.e., small values of Σ_i'' and Σ_f'') and the \mathbf{k} dependence of the matrix element is neglected, $N(E, \hbar\omega)_{k_{\parallel}}$ is seen from Eq. (2) to be proportional to the joint density of states (JDOS) for a fixed value of k_{\parallel} , as expected. We also see that in the limiting case of large Σ_f'' , the final state Lorentzian A_f will become broad and may be taken out of the integration in Eq. (2) yielding [9]

$$N(E, \hbar\omega)_{k_{\parallel}} \propto \int dk_{\perp} \delta(E - E_i(k_{\perp}) - \hbar\omega). \quad (4)$$

The right-hand side of Eq. (4) is the 1D density of initial states $\rho(k_{\parallel}, E)$ associated with the k_{\perp} band dispersion for a given k_{\parallel} . The final state dispersion no longer enters the energy dependence of the photointensity because these states now constitute a uniform continuum as a result of their large damping.

Equation (4) shows clearly that for a highly damped final state an angle-resolved experiment directly measures the 1D density $\rho(k_{\parallel}, E)$ of the initial states. Since $\rho(k_{\parallel}, E)$ is proportional to $1/[dE_i/dk_{\perp}]$, it is clear that extrema in the band structure as a function of k_{\perp} will induce features in the photoemission spectrum; these band extrema are usually expected to occur only at off-normal emission ($k_{\parallel} \neq 0$). In our case, $E_i = E_F$, therefore, whenever such an extremum crosses E_F , a feature representing a simple projection of the FS will in principle appear in the photointensity.

Figure 1(c) illustrates this effect. The outer closed contour G results from the minimum in the band at $k_{\perp} = 0$, and thus gives the cross section obtained when FS of Cu is cut by a (001) plane passing through the zone center. The elliptical regions N are similarly related to the projections of the (111) necks onto the (001) plane and involve the band extrema for nonzero k_{\perp} values [10]. G and N may be viewed as being DOS related features since $\rho(k_{\parallel}, E)$ enters crucially in making their appearance prominent in the CIE-AS spectrum. Of course, as Σ_f'' increases, the structure due to direct transitions becomes diffuse. The peak F in Fig. 1(c) is the broadened remnant of direct transitions [see Fig. 1(a)] around the zone center.

These results imply that as the electron mean free path decreases, and the number of layers sampled by the emitted electrons decreases concomitantly, the spectral signature of FS features such as G and N becomes relatively more intense. This may seem puzzling at first sight since we normally associate FS features with the long range coherence of Bloch states. There is no contradiction, however, so long as the initial states are sharply defined and coherent; it is just that the FS features in a CIE-AS

spectrum turn out to be remarkably simple when these states are probed via highly damped emitted electrons.

Bearing the preceding discussion in mind, we expect that the CIE-AS spectrum for an intermediate value of Σ_f'' will, in general, display features characteristic of both the small as well as the large Σ_f'' limits. This is seen to be the case in Fig. 1(b) for $\Sigma_f'' = 2$ eV which as noted above is representative of the physical situation. Figure 1(b) shows the presence of DOS features G and N of Fig. 1(c), on the one hand, and of broadened direct transitions such as A of Fig. 1(a), on the other hand. The complex k dependencies of the intensities of various features [note, e.g., prominent white spots J in Fig. 1(b)] result from subtle interplay between effects of matrix elements and the dispersion and damping of initial and final state bands.

Figures 2 and 3 consider the (110) and (111) surfaces, respectively. Much of the commentary of Fig. 1 above for the (100) surface is applicable with obvious changes and need not be repeated. With increasing Σ_f'' , the imprint of the direct transitions (e.g., A' or A'') becomes progressively more diffuse while the DOS related FS features (e.g., G' or G'') gain in prominence. For the (110) surface, the necks project into two dark spots N'_1 and N'_2 which are seen together with the intersection G' of the Cu FS with the (110) plane in Figs. 2(b) and 2(c). On the (111) surface, the white ring S near the center, present even when Σ_f'' is small, Fig. 3(a), is not a bulk transition but the well known Shockley state on Cu(111) in the bulk band gap on the hexagonal Brillouin zone face. The center of Figs. 3(b) and 3(c) also contains the neck N'' although the boundary of the associated dark spot is masked by the surface state S. In this connection, we have carried out simulations wherein the position of the surface barrier in the computations was placed in such a way that the (111) surface state was artificially removed with little change in the bulk bands; the resulting CIE-AS spectrum, Fig. 3(d), now shows clearly the dark spot N'' due to the neck around the center.

The question of how the present spectra are affected by various experimental parameters—especially the energy and polarization direction of the incident photons, and the energy and k resolution of the detector—deserves some comment. To this end, many additional CIE-AS spectra

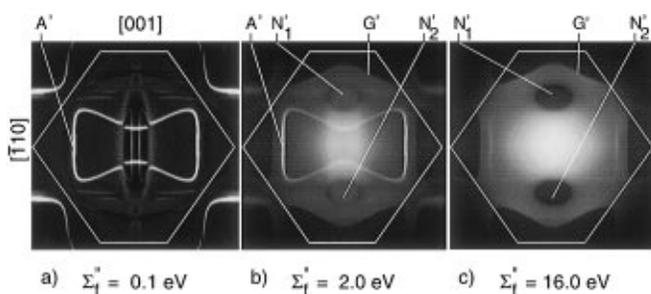


FIG. 2. Same as Fig. 1, except that this figure refers to the (110) surface.

for the three low index faces were computed. Figure 4 for the (100) surface using $\Sigma_f'' = 2$ eV is illustrative of the results. The spectra are influenced strongly by photon energy, as expected, because the direct transitions then involve a different final state. Figure 4(a) shows that for 26 eV photons many of the contours are quite different from those of Fig. 1(b), pointing to difficulties endemic to any attempt at deducing quantitative FS information in terms of direct transitions; in sharp contrast, the DOS related FS features in a CIE-AS spectrum are quite insensitive to $h\nu$ and provide a more straightforward measure of the FS dimensions [11]. The spectra possess a substantial polarization dependence which may in some cases help delineate spectral details. Differences between Figs. 4(b) and 1(b) indicate, for example, that the elliptical contours N involve several contributions which may be differentiable via polarized measurements. The C_4 symmetry is seen to break down in Fig. 4(b) for the p -polarized light considered. Finally, concerning the E - and k -resolution effects, a comparison of Figs. 4(c) and 1(b) shows that the energy resolution of even as much as 200 meV does not degrade FS features substantially. The spectra are, however, more sensitive to k resolution as seen from Fig. 4(d) where the spectrum for an approximately 2° analyzer acceptance window is simulated. The intrinsic accuracy with which the FS dimensions may be measured via the new DOS features would therefore be limited mainly by the k resolution of the experiment; this is also the case for FS determination using direct transitions.

Concerning relevant experiments, we are not aware of any literature where DOS type FS signatures in the CIE-AS spectra have been investigated along the lines

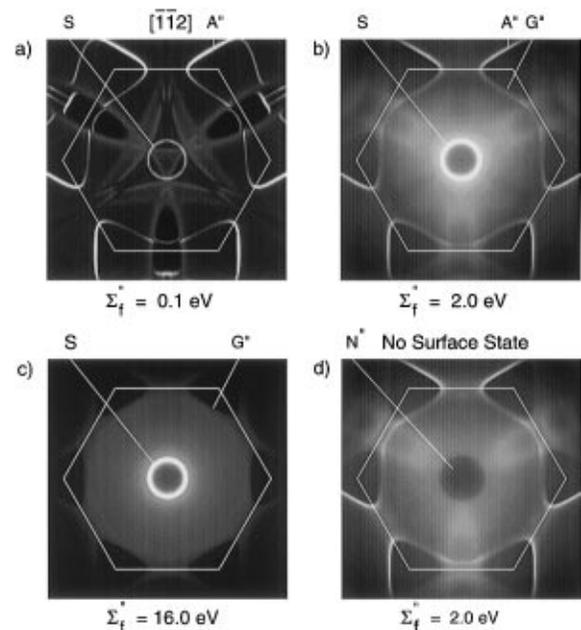


FIG. 3. Same as Fig. 1, except that this figure refers to the (111) surface. (d) The spectrum corresponding to (b) where the Shockley state around $p_{\parallel} = 0$ has been removed to expose the imprint of the (111) neck.

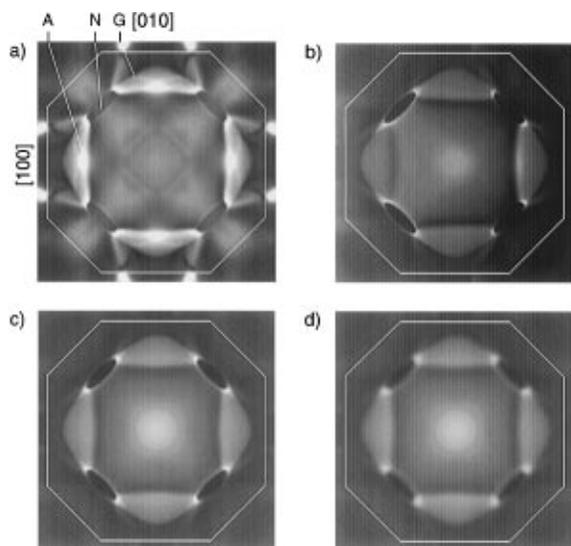


FIG. 4. Effects of various experimental parameters on the spectrum of Fig. 1(b). (a) is for 26 eV unpolarized light. (b) Spectrum for light polarized along the (100) direction. (c) The effect of 200 meV energy resolution, and (d) of k resolution assuming that the detector possesses an $\approx 2^\circ$ acceptance angle.

of this work. References [12] and [13] have, however, considered the FS of Cu in terms of direct transitions manifest in the CIE-AS spectra [14]. In this regard, we have compared our theoretical results in detail with those of Ref. [12] which reports CIE-AS data at the photon energy (21.22 eV) used in the computations of Figs. 1–3. The main direct FS features such as A in Figs. 1(b), 2(b), and 3(b) are found to be in reasonable accord with the corresponding measurements from the three low index faces of Cu (see Fig. 2 of Ref. [12]); some discrepancies between theory and experiment are present, but are not considered significant in view of uncertainties inherent in first principles computations, especially with respect to the final state. Interestingly, our analysis indicates that the weak feature observed around the \bar{Y} point from the (110) surface [see Fig. 2(c) of Ref. [12]] is the image of the Cu neck and not a surface state as suggested by Refs. [1,15,16].

In summary, we have shown that a CIE-AS spectrum for emission from the Fermi energy contains new FS signatures which have their origin in DOS type *indirect* transitions [17]. These DOS type FS features would provide a more robust and straightforward means of determining Fermi surfaces in comparison to the more conventional approach employing *direct* transitions. Since the associated photointensity is shown to be proportional to the 1D DOS of initial states as a function of k_\perp , these spectral features should also provide a new window for investigating k_\perp dispersion related issues in materials via the CIE-AS spectra.

This work is supported by the U.S. Department of Energy under Contract No. W-31-109-ENG-38, including a subcontract to Northeastern University, and the allocation

of supercomputer time at NERSC and the Pittsburgh Supercomputer Centers.

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- [1] P. Aebi, J. Osterwalder, P. Schwaller, L. Schlapbach, M. Shimoda, T. Mochiku, and K. Kadowaki, Phys. Rev. Lett. **72**, 2757 (1994).
 - [2] For a recent discussion of related issues see the conference proceedings, Special Issue on Spectroscopies in Novel Superconductors, edited by Z. X. Shen, D. H. Liebenberg, and A. Bansil [J. Phys. Chem. Solids **56**, No. 12 (1995)].
 - [3] J. B. Pendry, Surf. Sci. **57**, 679 (1976).
 - [4] G. A. Burdick, Phys. Rev. **129**, 138 (1963).
 - [5] J. F. L. Hopkinson, J. B. Pendry, and D. J. Titterington, Comput. Phys. Commun. **19**, 69 (1981).
 - [6] J. Pendry, in *Photoemission and the Electronic Properties of Surfaces*, edited by B. Feuerbacher, B. Fitton, and R. F. Willis (John Wiley and Sons, New York, 1978), p. 93.
 - [7] C. Kunz, in *Photoemission in Solids II*, edited by L. Ley and M. Cardona (Springer-Verlag, Berlin, 1979), p. 313.
 - [8] For completeness, the real part of final state self-energy Σ_f' is inserted into Eq. (3).
 - [9] Equation (4) is presented only to help gain insight into the new FS features. All computations reported in this paper employ the full one-step form (1). Equation (4) is reminiscent of the XPS spectra in that the JDOS is replaced by DOS; note, however, that the k resolution crucially important for inducing FS features in the present CIE-AS case is lost in XPS.
 - [10] Strictly, the necks will yield a dark spot corresponding to the k_\parallel region where no initial states are available. Therefore, care is necessary in deducing neck dimensions in terms of the size of the elliptical contour N in Fig. 1(c).
 - [11] The insensitivity of the DOS type FS features to $h\nu$ would allow these features to be distinguished from direct transitions experimentally. Also, the DOS type FS features would appear as cusps (rather than peaks) in the photoemission spectra.
 - [12] P. Aebi, J. Osterwalder, R. Fasel, D. Naumovic, and L. Schlapbach, Surf. Sci. **307–309**, 917 (1994).
 - [13] Z. Qu *et al.*, Surf. Sci. **324**, 133 (1995); A. P. J. Stampfl *et al.*, Surf. Sci. **331–333**, 1272 (1995).
 - [14] For recent work using a display-type angle-resolved photoemission setup in nonmetallic systems, see E. L. Shirley *et al.*, Phys. Rev. B **51**, 13 614 (1995); **53**, 10 296 (1996).
 - [15] P. Heimann, J. Hermanson, H. Miosga, and H. Neddermeyer, Surf. Sci. **85**, 263 (1979).
 - [16] We note, however, that the computational framework employed here only treats Shockley type surface states, and therefore the presence of some other type of surface state (e.g., Tamm type) around the \bar{Y} neck in the experimental spectrum cannot be ruled out.
 - [17] After this work was completed, a reexamination of the original CIE-AS Cu data of Ref. [12] has revealed clear hints of the presence of DOS type FS signatures G shown in Figs. 1(b), 2(b), and 3(b). These signatures are, of course, quite weak in the data as expected, and further work is required to pin down the details. We are very grateful to Dr. Aebi for important discussions in this connection.