Direct k-space photoemission imaging of the Fermi surface of Cu

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A technique has been used to accurately measure the bulk Fermi surface of Cu. Photoemission images $I(\mathbf{k},\hbar\omega)_{E_F}$ have been measured of the Fermi surface of Cu along two high symmetry k-space planes using constant initial-state spectroscopy. Many transitions from the Fermi surface were found to be adequately represented by free-electron final states over the entire photon energy range (10 – 100 eV) measured. The shape of the Fermi surface is preserved in the photoemission images measured and could be transformed directly back into k space, revealing an accurate picture of the corresponding Fermi-surface cross sections. Good agreement was found between our results and previously reported de Haas–van Alphen measurements and a linear muffin-tin orbital calculation.

The electronic band structure spanning the Fermi level of metals intimately determines the physical and chemical properties of bulk and surface regions of such materials.[1] The linear dimensions of the Fermi surface are, in particular, very sensitive to the crystal potential providing characteristic quantities of the electronic structure. de Haas–van Alphen (dHvA) measurements provide very high k-space resolution, yielding linear dimensions of the Fermi surface with better accuracy than calculations.[2–4] Photoemission spectroscopy has a poorer k-space resolution than dHvA spectroscopy [better than 0.2% of the measured values in the case of dHvA (Ref. 5) compared to $\sim 4-10$ % for photoemission spectroscopy [6] (PES)]. For metallic compounds however, dHvA measurements are difficult to perform and to interpret due to more than one type of elemental scattering center and the need for very-long-range sample periodic order [e.g., see dHvA results for Cu_3Au (Refs. 7 and 8)]. As a result, relatively few such compound measurements have been reported.[9] Positron annhilation is well suited to Fermi-surface measurements of metallic compounds. The accuracy of this technique (2–5 % of measured values) is comparable to PES. Determining the Fermi-surface radii, however, from these measurements is nontrivial: for example, in the case of Cu, corrections must be applied to separate the contribution of the $3d$ core electrons from that of the conduction electrons in the angular correlation data.[10] It is also necessary to correct for finite angular resolution and enhancement effects due to positron motion.[10,11] Photoemission spectroscopy on the other hand, does not suffer from such drawbacks since the initial k-space position of the electrons photoemitted from the Fermi surface may be directly measured. In addition to the methods outlined above there are a number of techniques that may be employed to measure the Fermi surface of metals but not metallic compounds. Magnetoacoustic geometric resonance (MAGR) and radio frequency size effect (RFSE) measurements yield dimensions with good resolution (MAGR better than 5% and RFSE 1%). The experimental conditions are similar to dHvA measurements (low temperature, high field), however, the determination of Fermi-surface dimensions is not as simple as for dHvA.[12–14] Other techniques include ultrasonic, anomalous skin effect and cyclotron resonance measurements. These are not as widely used and are less direct methods of acquiring information about the Fermi-surface dimensions.[15]

To date, little work has been reported on mapping the bulk Fermi surface of metals using photoemission spectroscopy. Early work using display type analyzers showed that it was indeed possible to obtain a photoemission image $I(\mathbf{k})_{E_F,\hbar\omega}$ at a fixed photon energy of the Fermi surface given that data covering sufficient emission angles at the appropriate kinetic energy were obtained.[16] Other workers[17–23] later improved the energy and k-space resolution by using energy dispersing analyzers.[24] One drawback of such a technique for determining the shape of the three-dimensional surface is that the photoemission image does not originate from a plane in k space but rather originates from a curved surface that intersects the Fermi sphere and that disperses in three dimensions.[25] Accurate measurements of spanning distances (neck or belly radii, for example) are difficult, or in general not attainable, using this technique.

One way to correctly map the shape of a three-dimensional Fermi surface is to measure the photoemission image $I(\mathbf{k})_{E_F}$ as a function of the excitation energy $\hbar\omega$, from the Fermi level within the appropriate high-symmetry planes. That is, to perform constant initial-state (CIS) spectroscopy using electrons from the Fermi surface for an appropriate range of polar emission angles θ, and photon energies that span the bulk Brillouin zone cross section.

We have measured such photoemission images $I(\mathbf{k},\hbar\omega)_{E_F}$ at the Fermi level for the $\{110\}[001]$ and $\{110\}[\bar{1}10]$ planes of Cu. A stack plot $I(k^{\parallel})$ of the photocur-

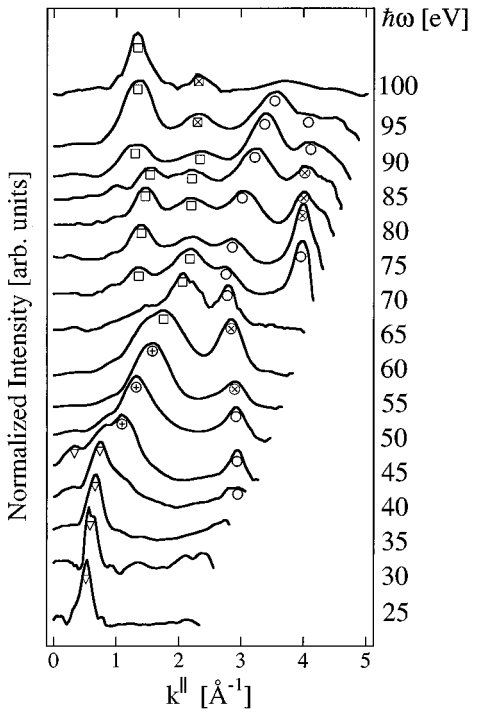

FIG. 1. Stack plot of the Cu{110} [001] constant initial-state spectra. Open symbols indicate transitions that fall onto free-electron primary cone final states; circles enclosing plusses indicate surface umklapp transitions; symbols enclosing crosses indicate transitions that do not fall onto free-electron primary cone final states.

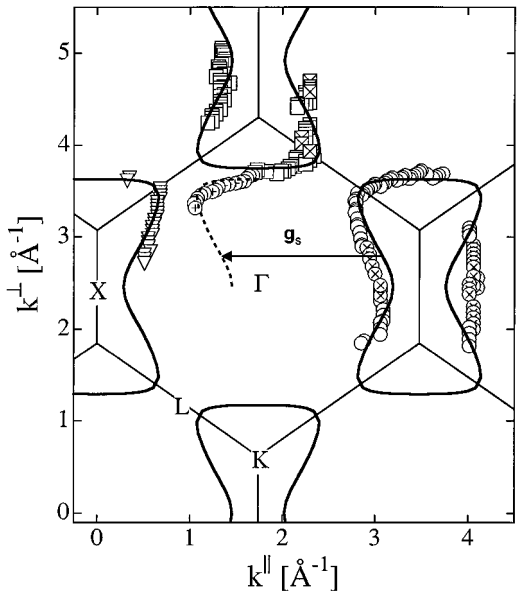

FIG. 2. The Fermi cross sections (thick solid lines) for the {110} [001] plane plotted in the extended Brillouin zone. Experimental positions are shown by triangles, squares, and circles. Thick dotted lines indicate the position of the Fermi slice allowing for surface umklapp.

rent taken at the Fermi level for a range of photon energies is shown in Fig. 1. Spectra were obtained by measuring a large number of polar angles ($\pm 90°$ in steps of $1°$) for the appropriate photon and kinetic energy combinations such that emission from only the Fermi level was measured. Experiments were performed at the Berlin synchrotron using a toroidal electron energy analyzer[26] and a toroidal grating monochromator.[27,28] The combined energy resolution of the spectrometer and monochromator was determined by measuring the width of the Fermi edge at each photon energy taken. The energy resolution varied from $\Delta E = 0.2$ to 0.4 eV for photon energies between 10 and 100 eV. The polar angle resolution of the analyzer was determined to be $\sim \pm 2°$ (Ref. 29); the azimuthal resolution was $\pm 1°$ as determined from the geometric dimensions of the analyzer.

The parallel component of the wave vector $k^{\parallel}$, given along the horizontal axis in Fig. 1, was determined using the usual equation $k^{\parallel} = \sqrt{(2m/\hbar^2)E_K}\sin\theta$. The background from the analyzer and the monochromator have been subtracted from each of the spectra shown in Fig. 1 using CIS measurements taken a few eV above the Fermi level (in the noise region) for the full range of angles and photon energies used. After background normalization the angle spectra were further normalized so that the intensity of the highest peak observed for each photon energy is the same. Finally, the spectra taken at negative and positive polar angles were summed to give averaged spectra. All spectra were acquired with *p*-polarized light at normal incidence to the surface: as a result spectra measured at equivalent negative and positive polar angles had nearly the same intensity distributions. Surface cleanliness was monitored by the identification of various known surface states in the energy spectra.[30]

Restricting measurements to transitions originating from the Fermi surface in a particular *k*-space plane allowed only those transitions from the selected Fermi cross section to be observed (to within a surface umklapp vector, $\mathbf{g}_s$). All major peaks in Fig. 1 have been identified as transitions from the Fermi cross section of the {110}[001] plane. Symbols mark the position of peaks that we have identified as direct transitions. Figure 2 shows the extended Brillouin zone for this plane; the *k*-space position of peaks identified in Fig. 1 are also shown. The Fermi surface cross section (shown as thick solid lines) was calculated using a self-consistent linear muffin-tin orbital (LMTO) band structure program provided by N. E. Christensen. A one panel calculation using $l = s$, p, d, and f atomic orbitals was performed to give the scalar-relativistic band structure. The shape of the calculated Fermi surface is clearly mimicked by the dispersion of the peaks as a function of photon energy in Fig. 1. These points have been mapped back onto the *k*-space plane of Fig. 2 assuming that transitions involved free electron final-state bands. The best fit between the experimentally mapped transitions and the calculation for both planes was obtained when an inner potential V_0, of 4.0 eV (with respect to the Fermi level), was used: final-state bands were defined by the free-electron formula $(E_K + V_0) = (\hbar^2/2m)\mathbf{k}^2$ where $E_K = \hbar\omega$ when $E_F = 0$, $k^{\perp}$ being the only unknown.

Transitions that are denoted by triangles, circles, and squares can be identified in Figs. 1 and 2 as originating from three locations on the Fermi surface. A large proportion of the mapped points (open symbols) are found to lie within experimental error ($\sim \pm 0.1$ Å^{-1}) of the calculated curves in Fig. 2. Not all mapped points, however, fall onto the Fermi

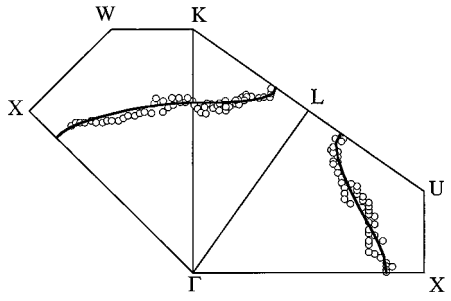

FIG. 3. The experimental Fermi surface of Cu as determined by photoemission. Experimental points (open circles) mapped back onto the irreducible Brillouin zone. Thick lines indicate the calculated position of the Fermi surface.

surface within experimental error. Some of these points (circles containing plusses) can be mapped back to the Fermi surface if the surface umklapp vector $\mathbf{g}_s=(2\pi/a)(110)$ is included so that $\mathbf{k}^{\parallel}=\mathbf{k}^{\parallel}-\mathbf{g}_s$. Thick dashed lines show the possible positions of such umklapped points. The experimental points falling on these lines come from very intense transitions located at $3.0\leqslant k^{\parallel}\leqslant 3.75$ Å^{-1}, $75\leqslant\hbar\omega\leqslant 95$ eV.[31] Peaks appearing around $k^{\parallel}=4.0$ Å^{-1}, $2.0\leqslant k^{\perp}\leqslant 3.0$ Å^{-1}, $75\leqslant\hbar\omega\leqslant 85$ eV in Figs. 1 and 2 are caused by a very intense transition located 0.5 eV below the Fermi level around $k^{\parallel}=4$ Å^{-1}, $k^{\perp}=2.2$ Å^{-1}, $\hbar\omega=70$ eV. This feature has been identified as a transition from the bulk sp band with a high joint density of states.[32] Broadening in energy and k allows this transition to be observed in the CIS spectra over a wide range of photon energies. All other remaining points not falling onto the calculation (symbols with crosses) can be mapped back onto the calculated curves using an absolute k-space coincident plane triangulation method:[32] these transitions are in general from the Fermi level to bands other than less free-electron-like final bands.

Figure 3 gives a summary of our results for the two planes measured. Primary cone transitions have been mapped back onto the irreducible Brillouin zone cross sections. The error in k space for all points shown is $\sim\pm 0.1$ Å^{-1}. The ability to accurately position the sample exactly at the center of the analyzer focus is of the order of a degree in polar and azimuthal angle. The measured k-space positions of such transitions therefore may be in error by as much as 0.2 Å^{-1}, especially near normal incidence and at high photon energies.

Table I lists several linear distances of the Fermi surface of Cu along several high-symmetry lines. For comparison, the corresponding calculated values and dHvA values are also given. Our results are within experimental error of the previously reported values. The largest error occurs around the neck region (i.e., the crossing point between the L-U and L-K symmetry lines). This discrepancy may be due to a temperature related broadening of the spectra: our data were acquired at room temperature while dHvA measurements require low temperatures. Recently the strong temperature dependence of the {111} surface state of Cu around the neck region was observed[33] and attributed to the similar temperature dependence reported for the bulk bands.[34]

Improvements to the accuracy of the technique reported here are essentially limited by the processes of the photoemission phenomena itself. Photoemission is a many-body phenomenon involving quasiparticles with finite lifetimes:[35–37] as a result the simple energy conservation relation for direct transitions given above has to be modified to include the real part of the self-energy (a many-body term) and importantly, it is relaxed due to the possibility of nonconserving k-space transitions. The latter is a result of the generally small mean free paths of ultraviolet photoelectrons[38] in the range of $10\leqslant\hbar\omega\leqslant 100$ eV. The eigenstates of photoelectrons from noble metals are smeared out in k space by $\Delta k\sim 0.1$ Å^{-1} for $\hbar\omega<20$ eV and $\Delta k\sim 0.2$ Å^{-1} for $\hbar\omega<100$ eV.[38] Our assumption that the final states are free-electron-like is valid in this energy range because the error in determining $k^{\perp}$ using this model is smaller than or equal to the intrinsic k-space error: The only parameter that was adjusted in our analysis to get the best fit between experiment and calculation was the inner potential V_0. If our value of 4.0 eV was in error by as much as 3–4 eV for $\hbar\omega\geqslant 30$ eV and 1–2 eV for $\hbar\omega<30$ eV this would give $\Delta k^{\perp}\sim 0.1$ Å^{-1}. An additional complication that limits k-space resolution is the problem of creating and maintaining perfect surfaces:[35,39] the elimination of impurity-defect smearing of $k^{\parallel}$ is a difficult but possible exercise.[33]

The photoemission k-space resolution appears then to be limited to around $\Delta k\sim 0.1-0.2$ Å^{-1} at present. Using very low or high photon energies and perfect surfaces may, however, reduce this value. Below $\sim$ 10 eV and at several hundred eV the mean free path of photoelectrons from noble metals increases to about 40 Å;[38] giving an error of $\Delta k\sim 0.03$ Å^{-1}. At these energies possible surface interference effects due to mixing between the surface and bulk excitations are also expected to be reduced[36,37] and hence distortion of the bulk spectra should be minimal.

In summary, we have presented the results of a photoemission study that has successfully mapped the bulk Fermi surface of Cu. More complicated Fermi surfaces should also yield to this technique. The intrinsic k-space resolution of the present technique has been discussed and it has been suggested that it may be reduced to obtain more accurate results, by acquiring spectra at either very low (<10 eV) or high (>100 eV) photon energies.

TABLE I. Cu Fermi-surface linear dimensions (in units of $2\pi/a$).

Direction	Experiment [a]	dHvA [b]	LMTO [c]
$\Gamma\Delta X$	0.84	0.8268	0.839
		0.8279	
ΓKX	0.74	0.7429	0.742
		0.7431	
$L-U$	0.20	0.14737 [d]	0.160
$L-K$	0.18	0.14737[d]	0.160
$\Gamma-W$	0.73		0.753
$\Gamma-U$	0.81		0.777

[a]Value derived using free-electron final states.
[b]Values taken from Coleridge and Templeton (Ref. 5) (first value) and Halse (Ref. 41) (second value).
[c]For details see text.
[d]Neck radius (mean value).

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