Comments

Comments are short papers which criticize or correct papers of other authors previously published in the **Physical Review**. *Each Comment should state clearly to which paper it refers and must be accompanied by a brief abstract. The same publication schedule as for regular articles is followed, and page proofs are sent to authors.*

Lifetime broadening in bulk photoemission spectroscopy

H. I. Starnberg, H. E. Brauer, and P. O. Nilsson
Department of Physics, Chalmers University of Technology, S-412 96 Göteborg, Sweden
(Received 13 July 1992)

We establish, theoretically and experimentally, that the "anomalous" broadening of features in photoemission spectra close to the Fermi energy, reported by Hwu *et al.* [Phys. Rev. B **45**, 5438 (1992)] is readily understood, by standard theory, as due to final-state broadening. Although we agree with the authors' statement that "anomalous" broadening is not sufficient as evidence for non-Fermi-liquid behavior, we do not endorse the claim that their results are qualitatively similar to previous results for surface bands.

In a recent paper, Hwu *et al.*[1] discussed whether the spectral width of photoemission features near the Fermi edge is relevant as a measure of the lifetime broading. This issue has attracted considerable interest because of recent suggestions that photoemission linewidths may provide vital information about the nature of high-T_c superconductors. Whereas conventional Fermi-liquid theory predicts a lifetime broadening depending quadratically on the binding energy, Olson *et al.*[2] have observed a linear dependence for $Bi_2Sr_2CaCu_2O_8$ (in its normal state). A possible interpretation could therefore be that this compound is not a conventional Fermi liquid. As pointed out by Hwu *et al.*,[1] such a conclusion seems rather farfetched as the quadratic dependence has never been observed in photoemission linewidths close to the Fermi edge, even for systems that are recognized as proper Fermi liquids.[3–8]

In their recent paper, Hwu *et al.*[1] referred to the results of Kevan *et al.*,[4–7] which convincingly demonstrated, for Cu and W surfaces, that lifetime broadening is not the dominant contribution to surface-state photoemission linewidths. In an attempt to generalize these results to bulk states of metals and quasi-two-dimensional materials, Hwu *et al.*[1] presented bulk photoemission data from Ag(100) and 2H-$NbSe_2$. In both cases they found increasing photoemission linewidths as the Fermi level E_F was approached, but they were not able to explain this "anomalous" broadening.

We want to point out that the "anomalous" broadening reported by Hwu *et al.*[1] is readily explained by standard photoemission theory as due to final-state broadening, and their results are therefore not comparable to those of Kevan *et al.*[4–7]

In Fig. 1, we illustrate our point of view by showing data obtained from the Cu(100) surface, in the ΓLUX symmetry plane, using Ne I radiation ($h\nu=16.85$ eV). These spectra correspond closely to those obtained by Hwu *et al.*[1] from Ag(100). We have, however, extended the measurements to larger emission angles θ, so that one can see the peak crossing the Fermi level a second time, moving down below E_F again. Just as for the case of Ag(100), the peak is rather broad (FWHM$\approx$0.7 eV) up to the first Fermi-level crossing. It is very striking, however, that the peak is significantly sharper (FWHM$\approx$0.4 eV) as it reappears at larger emission angles. We will

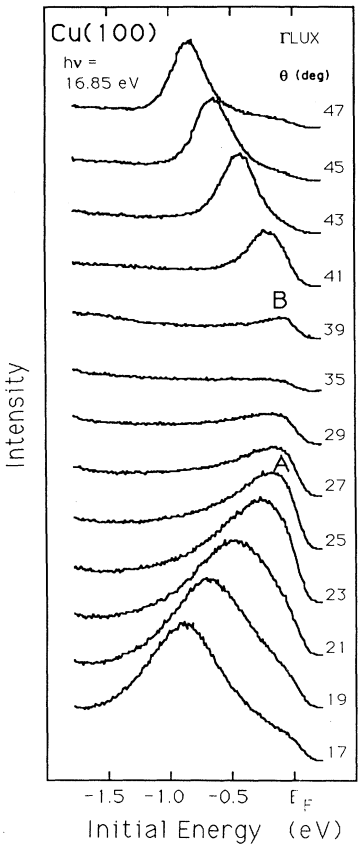

FIG. 1. Experimental energy distribution curves measured from Cu(100) for different emission angles in the ΓLUX plane. The initial-state energy is referred to the Fermi edge. The Fermi-level crossings are labeled *A* and *B*.

0163-1829/93/48(1)/621(3)/$06.00 ©1993 The American Physical Society

now show that this is precisely what can be expected if the lifetime broadening of the final-state band is accounted for.

The lifetime broadening of the final-state bands affects the photoemission peak widths by, in terms of the three-step model, relaxing the conservation of $k_\perp$, the wave-vector component perpendicular to the surface. The energy and the wave-vector component parallel to the surface are strictly conserved in the photoemission process, however. In terms of the three-step model, allowed direct transitions must satisfy the equations

$$E_i(\mathbf{k})+h\nu=E_f(\mathbf{k}) , \tag{1}$$

$$(k_\parallel)^2=(2m/\hbar^2)[E_i(\mathbf{k})+h\nu-\Phi]\sin^2\theta , \tag{2}$$

where $E_i(\mathbf{k})$ and $E_f(\mathbf{k})$ are the initial and final bands, $h\nu$ the photon energy, Φ the work function, and θ the polar emission angle. The energies are given relative to the Fermi level, and the repeated and extended zone schemes are used for initial and final states, respectively. Equations (1) and (2) can be used to calculate the angular dispersion of photoemission peaks from calculated or parametrized band structures.[9] For a certain emission angle θ, the allowed peak positions E_p (on the initial energy scale) are just those values of $E_i(\mathbf{k})$ which satisfy Eqs. (1) and (2). The effect of final-state broadening (width W) can be simulated by relaxing Eq. (1), so that transitions are allowed if the left- and right-hand sides do not differ by more than $W/2$. Figure 2 shows the result of such a calculation in correspondence with the experimental results in Fig. 1. The final-state band was approximated by a free-electron parabola (inner potential $V_0=-6.0$ eV) of width $W=4$ eV. The initial-state bands were obtained from a combined interpolation scheme as described by Smith and Mattheis.[10] It is clearly seen how the peak corresponding to the uppermost occupied band approaches and cross the Fermi level at A. As θ increases further, the peak crosses the Fermi level once again (at B) and moves down below E_F. The calculation reproduces the dispersion of the peak fairly well, and it correctly predicts that the peak width should be very different for the two branches of the band. While the final-state lifetime broadening of the peak is large for $\theta<30°$ it becomes almost negligible for the branch at larger angles. That the narrowing of the experimental peak when going to larger emission angles is less dramatic in the experimental results (see Fig. 1) than suggested by the calculation is readily understood as due to causes of broadening which were not included in the calculation. We are not analyzing these causes in detail, but expect the most important to be limited energy resolution of the analyzer (≈ 0.1 eV), in combination with the doublet nature of Ne I radiation ($\Delta h\nu=0.18$ eV), and the finite angular resolution ($\pm 2°$) together with the large angular dispersion of the peak. Inherent $k_\parallel$ broadening, as suggested by Tersoff and Kevan,[4,5] and remaining final-state broadening probably limits the narrowing that can be achieved by experimental refinements.

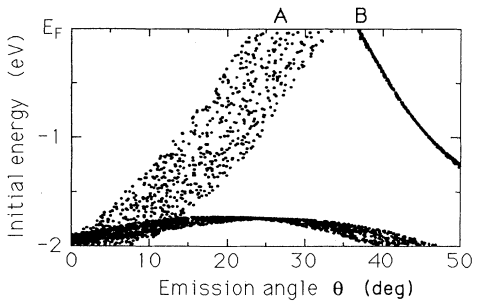

FIG. 2. Structure plot calculated in correspondence with the experimental data in Fig. 1, showing the peak dispersion as a function of θ. The calculation included a 4-eV final-state broadening, which resulted in varying broadening of the resulting structures. The Fermi level crossings are labeled as in Fig. 1.

It is instructive to examine in more detail why the final-state broadening affects the resulting peak widths so differently for different branches of the same initial-state band. In particular, it is striking that the calculated peak is very narrow beyond point B in Fig. 2, despite the assumed 4-eV width of the final-state band. We approach this question by looking at the peak positions E_p that can be obtained from solving Eqs. (1) and (2), and in particular how these peak positions change if a small energy shift ξ is applied to the final-state band. After introducing such a shift in Eq. (1) one may differentiate Eqs. (1) and (2) to evaluate the derivative

$$\left[\frac{\partial E_p}{\partial \xi}\right]_\theta = \frac{\partial E_i/\partial k_\perp}{\partial E_i/\partial k_\perp-\partial E_f/\partial k_\perp+(m\sin^2\theta/\hbar^2 k_\parallel)[(\partial E_f/\partial k_\perp)(\partial E_i/\partial k_\parallel)-(\partial E_f/\partial k_\parallel)(\partial E_i/\partial k_\perp)]} . \tag{3}$$

This derivative has previously been evaluated for the special case of a free-electron-like final band.[9] Since the lifetime-broadened final-state band can locally be expressed as a distribution of sharp bands with varying shifts ξ away from its centroid, this derivative becomes the coefficient by which the final-state width should be multiplied to obtain the final-state lifetime contribution to the resulting photoemission peak width. Equation (3) can be recognized as a component of the expression for the combined effect of initial- and final-state lifetime broadening which was derived by Chiang *et al.*[11] The single most important part of Eq. (3) is the numerator $\partial E_i/\partial k_\perp$, because if this is close to zero, as frequently is the case, the final-state contribution to the peak width becomes very small. Figure 3 shows approximately the path taken in $\mathbf{k}$ space as θ is increased during the measurement of the spectra in Fig. 1. In particular, the intersection with the Fermi surface at A and B is indicated. At A the Fermi surface is roughly parallel to the (100) surface, resulting in a large value of $\partial E_i/\partial k_\perp$ and, consequently, a large final-state contribution to the peak width. In contrast, at B the Fermi surface is almost perpendicu-

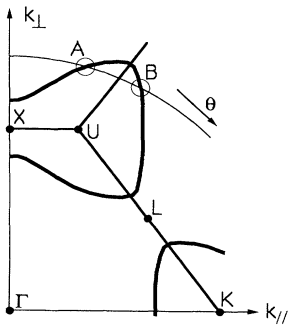

FIG. 3. Cross section of the Cu Fermi surface in the ΓLUX plane. The free-electron energy surface $E_f = 16.85$ eV (relative to E_F) and its intersections (A and B) with the Fermi surface are indicated, together with an arrow showing the movement in **k** space of direct transitions as θ is increased.

lar to the (100) surface, $\partial E_i/\partial k_\perp \approx 0$, and the resulting width, for this branch of the photoemission peak, is therefore dominated by other contributions, e.g., experimental energy resolution and angular acceptance. The **k**-space geometry of Fig. 3 was also considered by Tersoff and Kevan,[5] as they studied final-state broadening, using the angular variation method of Grepstad *et al.*[3]

The final-state broadening should be important also in the case of 2H-$NbSe_2$, since the band structure of this and similar compounds, although strongly anisotropic, is not truly two dimensional. In fact, band calculations[12] for 2H-$NbSe_2$ suggest that for several bands, including the ones close to the Fermi level, the dispersion perpendicular to the layers is quite comparable to the parallel dispersion.

In conclusion, Hwu *et al.*[1] correctly pointed out that the quadratic dependence of initial-state lifetimes has never been observed in photoemission, but they did not recognize the importance of the final-state width in bulk photoemission, and their results therefore do not represent a generalization of earlier surface-state results.

This work was supported by the Swedish Natural Science Research Council.

[1]Y. Hwu, L. Lozzi, S. La Rosa, M. Onellion, P. Alméras, F. Gozzo, F. Lévy, H. Berger, and G. Margaritondo, Phys. Rev. B **45**, 5438 (1992).
[2]C. G. Olson, R. Liu, D. W. Lynch, R. S. List, A. J. Arko, B. W. Veal, Y. C. Chang, P. Z. Jiang, and A. P. Paulikas, Phys. Rev. B **42**, 381 (1990).
[3]J. K. Grepstad, B. J. Slagsvold, and I. Bartos, J. Phys. F **12**, 1679 (1982).
[4]S. D. Kevan, Phys. Rev. Lett. **50**, 526 (1983).
[5]J. Tersoff and S. D. Kevan, Phys. Rev. B **28**, 4267 (1983).
[6]S. Kevan, Phys. Rev. B **28**, 4822 (1983).
[7]S. Dhar and S. D. Kevan, Phys. Rev. B **41**, 8516 (1990).
[8]A. Goldmann, W. Altmann, and V. Dose, Solid State Commun. **79**, 511 (1991).
[9]H. Mårtensson and P. O. Nilsson, Phys. Rev. B **30**, 3047 (1984).
[10]N. V. Smith and L. F. Mattheiss, Phys. Rev. B **9**, 1341 (1974).
[11]T. C. Chiang, J. A. Knapp, M. Aono, and D. E. Eastman, Phys. Rev. B **21**, 3513 (1980).
[12]G. Wexler and A. M. Woolley, J. Phys. C **9**, 1185 (1976).