k-Resolved Inverse Photoemission from Cu(001) and Ni(001)

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Ultraviolet isochromat spectra ($\hbar\omega$= 9.7 eV) have been taken on the single-crystal surfaces Cu(001) and Ni(001) as a function of the angle of incidence of the incoming electrons. The energy dispersion and intensity variation of the observed peaks in the isochromat spectra are interpreted successfully in terms of direct transitions within the bulk band structure.

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Recent experimental progress[1-3] has aroused new hopes for ultraviolet bremsstrahlung isochromat spectroscopy as a technique for investigating the electronic structure of solids and their surfaces. The physical basis of the technique may be conveniently viewed as the inverse of the better known photoemission process.[4] An incoming electron couples into an unoccupied state of the sytem and then decays radiatively into another unoccupied state. Monitoring the emitted x-ray or ultraviolet photon flux as a function of incident electron energy permits one to map out features of the empty states. The important advantage of the technique is its ability to probe the states between the Fermi level and the vacuum level, a range which is inaccessible in conventional photoemission spectroscopy. The technique could therefore be particularly powerful in the investigation of magnetic materials and semiconductors, where the lowest-lying empty states are frequently the ones of most physical interest.

In this Letter, we announce the development of k-resolved inverse photoemission spectroscopy (KRIPES), and we report our first results on Cu(001) and Ni(001). The new feature of these experiments is that they are performed on single-crystal surfaces, and the direction of the incident electron beam is well defined. We therefore have knowledge of the parallel component $k_{\parallel}$ of the electron wave vector, analogous to the situation in angle-resolved photoemission. Our most significant new result is the observation of the energy dispersion relation $E(k_{\parallel})$ for peaks in the spectra. We have also performed model calculations based on direct transitions within the bulk band structure, and we find that they account satisfactorily for both the energy dispersion and the intensity behavior of the observed peaks.

The experimental KRIPES arrangement is similar to that described by Denninger, Dose, and Scheidt.[3] The photon detector is an iodine-filled Geiger-Müller counter with a CaF_2 window, having a band pass with full width at half maximum ~ 0.7 eV centered on the photon energy $\hbar\omega$ = 9.7 eV. The important difference in our arrangement is that we use for the electron source a commerical electron gun operated at a fixed energy of 40 eV. The incident electrons are then decelerated to the low energies required (~ 5–10 eV) by a parallel grid retardation stage placed between the gun and sample. The angle of electron incidence is varied by rotating the sample.

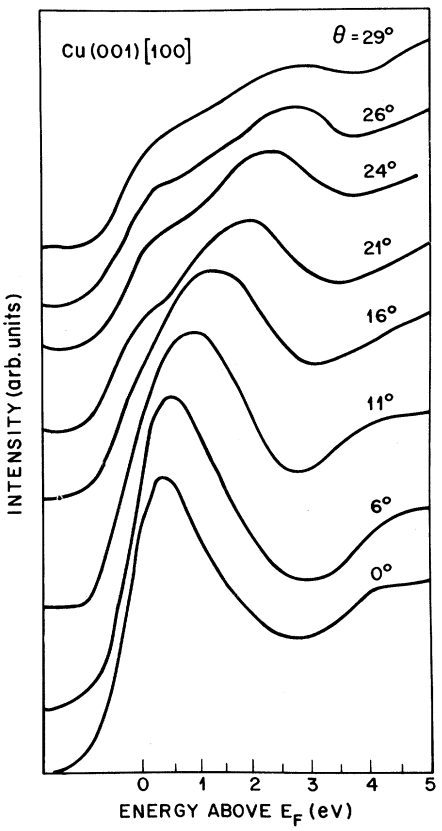

FIG. 1. Variation of the 9.7-eV isochromat spectra from Cu(001) with angle of electron incidence. Energies are referred to the Fermi edge.

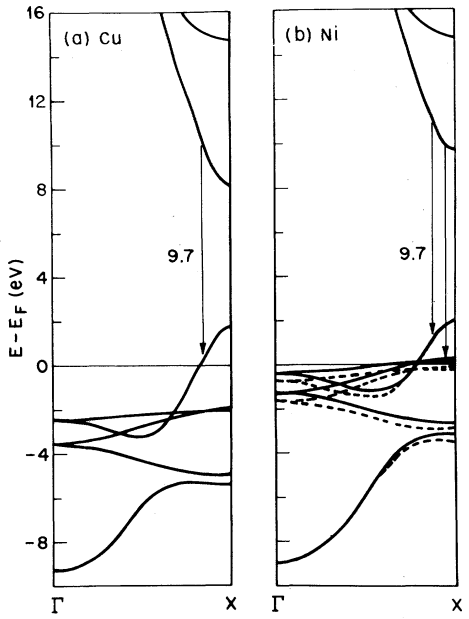

FIG. 2. Band structures along the ΓX direction appropriate to normal electron incidence on (a) Cu(001) and (b) Ni(001). In the case of Ni, full and dashed curves represent the minority and majority spins, respectively. Vertical arrows indicate the direct transitions possible at $\hbar\omega = 9.7$ eV.

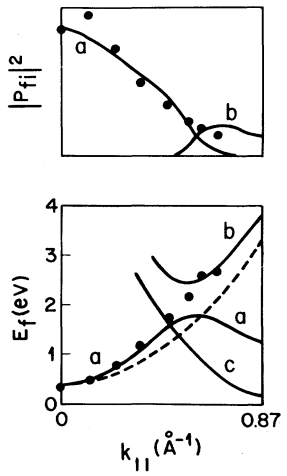

FIG. 3. Dispersion and intensity of the s,p peak in Cu(001). The lower panel shows the comparison between experimental peak positions (full circles) and the theoretical dispersion relations obtained from the combined interpolation scheme (full curves); the dashed curve is the prediction of a simpler two-band model. The upper panel shows the comparison between the measured peak intensities (full circles) and the calculated values for $|P_{fi}|^2$, the square of the momentum matrix element; the values are normalized to the same point at $k_{\parallel} = 0$.

This means that the photon collection angles are also varied. The experiment does not depend critically on having a well-defined direction of photon emission.[4] Moreover, the detector collects over a rather large solid angle (~ 10% of the total 2π sr) in order to enhance the signal level. Counting rates were typically several hundred per second for incident electron currents of ~6 μA. The samples were cleaned in the vacuum chamber (base pressure ~2×10^{-10} Torr) by the usual cycles of argon-ion bombardment and annealing.

The KRIPES data for Cu(001) are shown in Fig. 1 as a function of the electron incidence angle θ. At normal incidence a very prominent peak is seen just above the Fermi edge. On increase of θ, the peak is seen to disperse upwards in energy and to diminish in intensity. This behavior is readily understood in terms of direct transitions within the one-electron bulk band structure.

At normal incidence, $k_{\parallel} = 0$, we are confined to the bands along the ΓX direction in k space, which are shown for Cu in Fig. 2(a). A vertical transition is possible at 9.7 eV from band 7 to a state in band 6 lying 0.38 eV above the Fermi level, E_F. To treat the situation away from normal incidence, we have used a combined interpolation scheme[5] to calculate the final-state energies and momentum matrix elements for the 9.7-eV transitions. The results are compared with experiment in Fig. 3. The agreement for the upward dispersion for the final energy is seen to be good. The theoretical results display a number of branches (curves a, b, and c) corresponding to transitions from bands 7, 8, and 9 into band 6. From considerations of coupling to external plane waves,[5] we expect the experimental peak to follow the branches corresponding to initial states whose wave function is composed primarily of the plane wave $\exp[i(\vec{k}-\vec{G})\cdot\vec{r}]$ where $\vec{G}$ is the (0, 0, 2) reciprocal lattice vector. Indeed a two-band model leads to the following prediction for the off-normal energy dispersion[6]:

$$E(\vec{k}_{\parallel}) = \hbar^2 k_{\parallel}^2/2m + [(E_G - \hbar\omega)^2 - 4V_G^2]/4E_G, \quad (1)$$

where $E_G = \hbar^2 G^2/2m$ and V_G is the appropriate pseudopotential. Equation (1) is shown as the dashed curve in Fig. 3 where it follows the branches of strong external coupling and the experimental data. Our resolution is presently in-

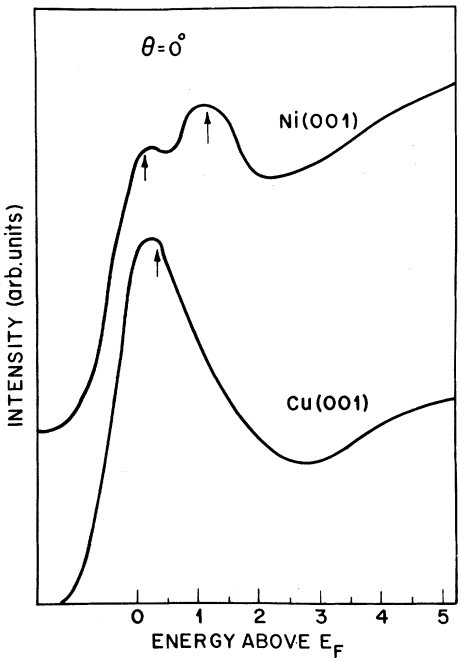

FIG. 4. Comparison of the normal incidence isochromat spectra for Ni(001) and Cu(001). The arrows indicate the final energies for possible direct transitions.

sufficient to detect the gap between branches a and b.

The momentum matrix elements $|P_{fi}|^2$ generated by the combined interpolation scheme are compared with the experimental intensities in Fig. 3. A crude background subtraction has been applied to the data by sketching in an extrapolation of the background adjacent in energy to the peak. The absolute intensities are likely to be influenced by the varying collection angles. Nevertheless, the agreement is quite reasonable. Note that the two-band model would definitely be inadequate here since it predicts constant momentum matrix elements.

Turning to Ni(001), we expect on the basis of the band structure [Fig. 2(b)] to see two features: one due to transitions into the s,p-like band 6 (Δ_1 symmetry) at an energy 1.16 eV above E_F; and one due to transitions into the unoccupied minority spin d bands (Δ_5 and Δ_2 symmetry) at 0.1–0.2 eV above E_F. (The band structure used here is an empirically adjusted one obtained by varying the parameters of the combined interpolation scheme to reproduce recent angle-resolved photoemission data.[7]) The normal-incidence KRIPES data for Ni(001 are shown in Fig. 4 where they are compared with those for Cu(001). The s,p peak is observed at the expected energy, but the d peak appears only weakly. This can be understood once again by examination of the momentum matrix elements. At the zone boundary transitions from the s-like X_1 level into the d-like X_5 and X_2 levels are symmetry forbidden, and they remain weak on moving into the zone. Transitions across the X_1-$X_{4'}$ s,p gap, on the other hand, are particularly strong.[5] Our calculations using the combined interpolation scheme predict that at $\hbar\omega = 9.7$ eV, the values of $|P_{fi}|^2$ for transitions into the d bands are less than 0.3% of those for transitions into the s,p band.

In summary, we have demonstrated the feasibility of KRIPES, and we have shown that the observed energy dispersion and relative peak intensities can be understood in terms of a rather simple model based on bulk band structure. Even with the present limitations of poor resolution and fixed photon energy, KRIPES promises to be fruitful. When these limitations are eventually removed, the technique should be quite powerful.

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