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ANGULAR RESOLVED U.V. PHOTOEMISSION FROM (100) NICKEL SURFACE

T. T. A. NGUYEN, R. C. CINTI

C.N.R.S. Groupe des Transitions de Phases, B.P. 166, 38042 Grenoble Cedex, France

and

S. S. CHOI

Applied Optics Laboratory, Korea Institute of Science and Technology
P.O. BOX 131, Dong Dae Moon, Seoul, Korea

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Résumé. — On présente des résultats préliminaires de l'étude de la face (100) du nickel paramagnétique par photoémission en U.V. lointain. Les spectres obtenus sur la surface pure par un système de mesure angulairement résolu sont discutés en fonction de la structure de bande calculée et des résultats parallèles obtenus sur le nickel polycristallin.

Abstract. — Initial results obtained on a (100) paramagnetic nickel surface by angular-resolved U.V. photoemission are presented. The spectra measured for different electron emission angles are discussed in terms of the calculated band structures and are compared with similar results previously obtained on polycrystalline samples.

U.V. photoemission spectroscopy has been increasingly used during the past few years to investigate both the bulk [1, 3] and surface [4, 5] electronic properties of metals. The technique has been refined and recent work [6, 7] using angular resolved systems explores the anisotropy of photoemission from single crystals. Results analysed with the bulk three step model [1] or the surface emission model [4, 8] are often in good agreement with the calculated band structures assuming specular refraction conditions for photoelectrons escaping through the surfaces [9]. They are also often more detailed than those obtained by angular integrated U.P.S. which tends to smear out some details of the spectra.

In this letter we present our initial results obtained on a (100) nickel surface with an angular-resolved, windowless, ultra high vacuum spectrometer described elsewhere [10, 11]. We compare the results with those obtained on polycrystalline samples by the angular-integrated method [12, 13].

The single crystal used in the experiment was cut along the (100) crystallographic plane to within 0.5°. After a careful mechanical polishing and electropolishing it was cleaned *in situ* by successive ion-bombardments and annealings until there was no contamination, especially by sulfur, as detected by Auger analysis.

During the experiment contamination by carbon

monoxide was avoided by periodic heating to 450 °C where Co desorbed without segregation of sulfur from the bulk. Spectra were measured when the crystal was cooling, in the temperature range between 450° and 360° where the nickel is paramagnetic.

Figure 1 shows the clean surface spectra for the four incident photon energies used : 10.2, 11.8, 13.5, and 16.8 eV. The bottom scale refers to the initial state energy of electrons before excitation relative to the Fermi level. No correction for the variation of analyser resolution and transmission with energy was applied to the curves and the intensity scales were arbitrarily normalized for presentation. In this set of experiments the incident angle of the unpolarized light was about 48° to the surface normal and only electrons emitted in a cone of 5° semi-angle directed along the [100] direction were collected to form the spectra.

A common feature seen on these curves is the well-defined peak -0.2 eV below the Fermi level and stable in initial energy when $\hbar\omega$ varies. With this structure, for $\hbar\omega = 11.8$ eV a small shoulder also emerges which moves to lower energies when the excitation energy increases. Because of the present lack of reliable band structure data extending to sufficiently high energies for nickel, it is difficult to determine the transitions involved in these spectra exactly. We can only propose possible interpretations and

FIG. 1. — Experimental spectra of photoelectrons emitted normal to the (100) surface for excitation energies between 10.2 and 16.8 eV.

discuss our spectra in relation to the corresponding ones obtained on polycrystalline samples.

The first important point is the large stationary peak situated -0.2 eV below E_F also present in evaporated thin films over a very large excitation energy range (5 to 40 eV). Smith and Traum [12] have shown its similarity to the large structure found at the upper limit of the « d » band of many of the F.C.C. transition metals. The stability of this peak when $\hbar\omega$ is varied gives it *initial state character* and shows that it is associated with the occupied « d » band. However this stability does not unambiguously determine the type of photoemission mechanism operating here as calculations show the upper « d » sub-band, which must certainly participate in this emission, to be very flat [14].

Conversely the second structure observed definitely has the character of a direct bulk transition between the upper « d » sub-band along the ΓX direction and the quasi-parabolic empty band situated above the Fermi level in the same direction of the Brillouin zone.

The second important point is the general difference

between our spectra and the angular-integrated ones. Confirming the directional properties of photoemission they are less rich in structure : for example, for the same excitation energy of 16.8 eV two important peaks located at about -1.7 eV and -2.2 eV below E_F observed by Eastman [13] are not present on our curves. One can note also the good fit (within the order of the experimental error) between the locations of the first peak found in our experiments on paramagnetic nickel and in the work on the ferromagnetic metal. This confirms the previous observations of Spicer and Pierce [15] who found no important differences between the photoemission properties of the two magnetic phases.

To complement these first results we have measured some spectra from the same surface with different analysis angles θ with respect to the crystal normal, arcs θ projecting onto the [001] direction in the surface plane. As in our experimental system both the photon incident direction and the analyser axis were fixed, this was done by rotating the crystal around an axis perpendicular to the two first and contained in the surface plane (Fig. 2).

FIG. 2. — Schematic view of the experimental arrangement. Axis of analyser and incident photon flux direction are fixed and lie in a plane containing the surface normal. The crystal rotates about an axis perpendicular to this plane.

For the first time the influence of the angle of photon flux incidence was checked by measuring for the same excitation energies, pairs of spectra obtained for two angles θ and $-\theta$ symmetric with respect to the surface normal. As theoretically expected in the dipole approximation only small variations in the relative amplitudes of these spectra were observed. Figure 3 shows a set of curves measured for $\hbar\omega = 16.8$ eV when θ is varied between 0° and 55° , the total emission current being kept constant and equal for each measurement. The intensity of spectra decreases clearly when θ is increasing and a second peak grows up from the initial shoulder to become larger than the first. This second structure moves in initial energy with the collection angle giving some confirmation for its being a bulk direct transition

FIG. 3. — $\hbar\omega = 16.8$ eV. Experimental spectra measured for different collection angles. θ is defined in figure 2.

as proposed above. Conversely the position of the first peak remains stationary as in figure 4 where spectra were measured for $\hbar\omega = 10.2$ eV in the same total emission current conditions. Surprisingly no significant variation of intensity with θ is observed for this photon energy, furthermore no supplementary structure appears. This last fact suggest a possible interpretation in which the absence of final empty states available for $\hbar\omega = 10.2$ eV is due to the large

FIG. 4. — $\hbar\omega = 10.2$ eV. Experimental spectra measured for different collection angles. θ is defined in figure 2.

band gap situated between the symmetry points X'_4 and X_1 above the Fermi level, in the ΓX direction of the Brillouin zone [16]. The first peak should originate then by a surface emission process in the sense of Feuerbacher and Christensen [4].

A more complete report with results for the (110) and (111) surfaces is in preparation.

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