

PHOTOEMISSION WITH HIGH ANGULAR AND ENERGY RESOLUTION: TEMPERATURE DEPENDENT EXCHANGE SPLITTING IN NICKEL

T.J. Kreuz, P. Aebi*, J. Osterwalder

Physik-Institut, Universität Zürich-Irchel, CH - 8057 Zürich, Switzerland

*Institut de Physique, Université de Fribourg, CH - 1700 Fribourg, Switzerland

Ultraviolet photoemission spectroscopy data from a Ni(110) crystal taken with high angular and energy resolution at room temperature (RT) and near the ferromagnetic Curie temperature are presented. For a polar scan in the (001) plane we recorded energy spectra within 1.5 eV from the Fermi energy (E_F) in 1° steps. Distinct bands crossing E_F can be well separated and, by comparing the RT data with a recent band structure calculation [H. Eckardt, L. Fritsche, *J. Phys. F* 17, 925 (1987)], the bands can be assigned to a minority d -band and two exchange split sp -bands. While their exchange splitting can hardly be extracted from the energy spectra, the peaks are well separated in k -space, i.e., in polar angle. We determine $\Delta E_{ex} = 204 \pm 8$ meV for the sp -band in the examined k -region and find that the angular line width of the majority sp -band is significantly smaller than that of the minority sp -band. Upon approaching T_C the exchange splitting of the sp -band vanishes and the d -band becomes very broad.

Keywords: A. magnetically ordered materials, A. metals, D. electronic band structure, D. phase transitions, E. photoelectron spectroscopies

Angle-resolved ultraviolet photoemission spectroscopy (ARUPS) provides the most direct information on the occupied electronic states of metals¹. It is often applied to probe the bandstructure at discrete points in k -space by measuring energy distribution curves (EDCs) and by assigning peaks to direct transitions from occupied to unoccupied electronic bands. This mode of measurement, which we here term $E(k)$, has been used to experimentally determine the magnetic exchange splitting ΔE_{ex} of energy bands in itinerant ferromagnets, and in particular in nickel²⁻⁷. Below its Curie temperature ($T_C = 631$ K) Ni is ferromagnetic, which is reflected in the different number of electrons with their spins aligned parallel (majority electrons; "spin up") or antiparallel (minority electrons; "spin down") to the magnetization direction. Accordingly, there are two subsets of electronic states that are energetically separated by the exchange splitting ΔE_{ex} , which can be energy, wave vector and temperature dependent⁸.

Despite the large amount of experimental and theoretical work performed on Ni, its electronic structure near and above T_C is far from being well understood (for reviews see Ref. 8 and 9). ΔE_{ex} , and in particular its temperature dependence are good quantities for gauging the various theories, but unfortunately experimental data are relatively scarce. The exchange splitting is typically of the order of 200 meV²⁻⁷, which is close to the intrinsic linewidth of transitions observed in Ni. The values of ΔE_{ex} therefore depend strongly on the line-shape analysis in the EDCs and are thus not unambiguous. Spin-resolved photoemission experiments give here much

clearer results^{6,7}, but at the cost of very low photoelectron intensities, which is prohibitive for an extended study over a large portion of the Brillouin zone.

A rather new way for obtaining band structure information from ARUPS is to measure constant-energy surfaces in k -space, particularly near the Fermi energy, by using a display-type analyzer¹⁰ or by extended angular scanning¹¹. Two-dimensional cuts through the bulk Fermi surface (FS) can be obtained. We here term this mode of data acquisition $k(E)$, since for one energy many points in k -space are measured.

The experimental resolution in energy as well as in angle is of great importance in ARUPS, since a lack of resolution in either of the two will lead to a peak broadening in both quantities to an amount which depends on the probed band structure^{12,13}. In this paper we combine $E(k)$ and $k(E)$ by measuring angular scanned EDCs in one-degree steps in the $\Gamma X W K$ -plane of Ni at room temperature (RT; $T/T_C \approx 0.46$) and at elevated temperature ($T/T_C = 1$). We show that for a given energy and angular resolution the $k(E)$ mode separates the exchange-split sp -bands very clearly, whereas the related peaks overlap strongly in the EDCs. ΔE_{ex} for this case is found to vanish for $T \approx T_C$.

The experiments were performed in a modified VG ESCALAB Mark II with a base pressure of 2×10^{-11} mbar, which rose to 3×10^{-9} mbar during operation of the differentially pumped He-discharge lamp. Ar⁺-sputtering and annealing were applied to prepare a clean and ordered surface, which was checked by X-ray photoemission spectroscopy and low energy electron diffraction before

and after the measurements. No sulfur, less than 1% oxygen and less than 5% argon and carbon could be detected. The unmagnetized sample was excited with unpolarized He I radiation ($h\nu = 21.2$ eV). In 1° steps we measured EDCs spanning 1.5 eV and including E_F for a polar scan ranging from the surface normal ($\theta_m = 0^\circ$) to a polar angle of $\theta_m = 70^\circ$ along the $[\bar{1}10]$ -azimuth ((001) plane) of a Ni(110) single crystal. An energy resolution of approximately 30 meV and an angular resolution of better than $\pm 1/2^\circ$ were used. Measurements at elevated temperature were performed in a mode with alternating heating and measuring cycles to avoid disturbing electric and magnetic fields due to the indirect resistive ac sample heating.

Fig. 1 shows a section parallel to the (001) plane through k -space for face-centered cubic Ni (lattice constant $a = 3.52$ Å) with some bulk Brillouin zones and high-symmetry lines and points indicated. The measured polar scan lies in this plane. Assuming a free electron final state after the photoexcitation process and constant initial state energy (e.g. E_F), the emitted electrons lie on a sphere in k -space. The cut through this sphere for initial states at the Fermi energy is indicated in Fig. 1. Circles for other measured energies in our 1.5 eV range have only slightly different radii and are not drawn. During the refraction of the electrons at the solid-vacuum interface, the parallel component k_{\parallel} of the wave vector k and the energy are conserved. Assuming a work function of 4.7 eV and an inner potential of 10.7 eV⁴, one can also reconstruct the perpendicular component k_{\perp} and hence the initial electron states by intersecting a sphere of corresponding radius with the bulk band structure¹¹.

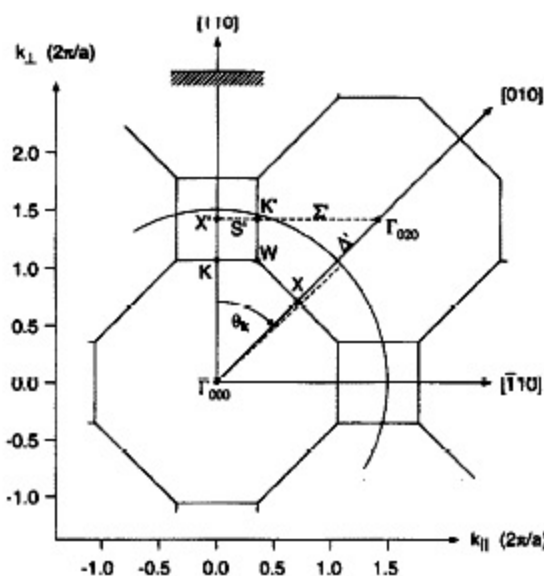


Fig. 1. Cut through reciprocal space perpendicular to the $[001]$ -direction in the extended zone scheme. The solid polygons indicate Brillouin-zone boundaries for face-centered cubic Nickel. Some high-symmetry points and lines are indicated. The final state free electron wave vectors for electrons from the Fermi edge lies on the circle.

Due to the refraction the measured polar angle θ_m in vacuum differs from the polar angle θ_k in k -space inside the solid. In our case the maximum angle of $\theta_m = 70^\circ$ corresponds to $\theta_k = 47^\circ$, which is indicated in Fig. 1. Thus the polar scans start close to X' , pass closely by K' , cross Σ' and finish 2° beyond Δ' . Consulting a recent bandstructure and FS calculation by Eckardt and Fritsche¹⁴, we expect to see five bands crossing E_F in our polar scan: first, close to normal emission the two minority bands from X_2^+ and X_2^- , and then at about $\theta_k = 16^\circ$, close to crossing the Σ -line, the minority d -band from K_2^+ , which we here name " Σ_2^+ ". Finally at $\theta_k = 36^\circ$ and $\theta_k = 40^\circ$ we expect the two exchange-split sp -bands between Σ_1^+ and Δ_1^+ and between Σ_1^- and Δ_1^- , respectively, which we will call " Σ_1^+ " and " Σ_1^- ".

The raw experimental data are shown in Fig. 2 in a linear gray scale with minimum intensity as black and maximum intensity as white. The intensity drop at E_F is clearly visible. In the RT data we can well distinguish two closely spaced bands crossing E_F at about 49.5° and 54.5° in θ_m (36.5° and 39.5° in θ_k) and another band crossing E_F at about $\theta_m = 27^\circ$ ($\theta_k = 21^\circ$). Near 0° we find intensity from at least two bands, which appear to mix strongly and which will not be analyzed further within this paper. Comparing these results to the above mentioned FS calculation¹⁴, we can clearly assign the bands at $\theta_k = 39.5^\circ$, $\theta_k = 36.5^\circ$ and $\theta_k = 21^\circ$ to the sp -bands " Σ_1^+ ", " Σ_1^- " and the d -band " Σ_2^+ ". The two-dimensional gray scale representation of the data (Fig. 2) is particularly instructive as it shows not only the positions of Fermi level crossings very clearly, but also how the bands disperse along the polar scan and how the relative intensities and linewidths depend on energy and angle.

The effects of temperature on these spectra, some of which are related to the magnetic phase transition, are quite dramatic. The Fermi edge for $T/T_C = 1$ can be seen to be less steep than in the RT measurement. More importantly, we see that all bands change strongly. There is only one band left in the $\theta_k = 38^\circ$ -region, which means that the two formerly exchange-split sp -bands have merged to one single " Σ_1 "-band. Looking at the " Σ_2^+ "-band, we find that it has broadened considerably, unlike the " Σ_1 ", which shows a comparable width in θ_m as at RT. All these changes happen continuously with increasing temperature¹⁵.

Fig. 3 shows EDCs extracted from the RT data in the angular range where the two sp -bands cross E_F . " Σ_1^+ " appears near E_F at $\theta_m = 53^\circ$ ($\theta_k = 38.5^\circ$) and " Σ_1^- " practically vanishes at $\theta_m = 59^\circ$ ($\theta_k = 42^\circ$); only at $\theta_m = 56^\circ$ or 57° both peaks are visible simultaneously in the EDCs, but still riding on a large background. Here the need for a very high angular resolution becomes evident: With an angular resolution of, e.g., 3° , the two peaks in the EDCs would be considerably broadened because of the dispersion and would strongly overlap, and even spin-resolved measurements could hardly separate them clearly. An accurate determination of the peak positions and of the exchange splitting would thus be nearly impossible.

Fig. 4 shows the polar intensity scans at the Fermi energy extracted from the data of Fig. 2 at RT and $T = T_C$. Obviously the exchange-split sp -bands are very clearly separated here and riding on a low and smooth back-

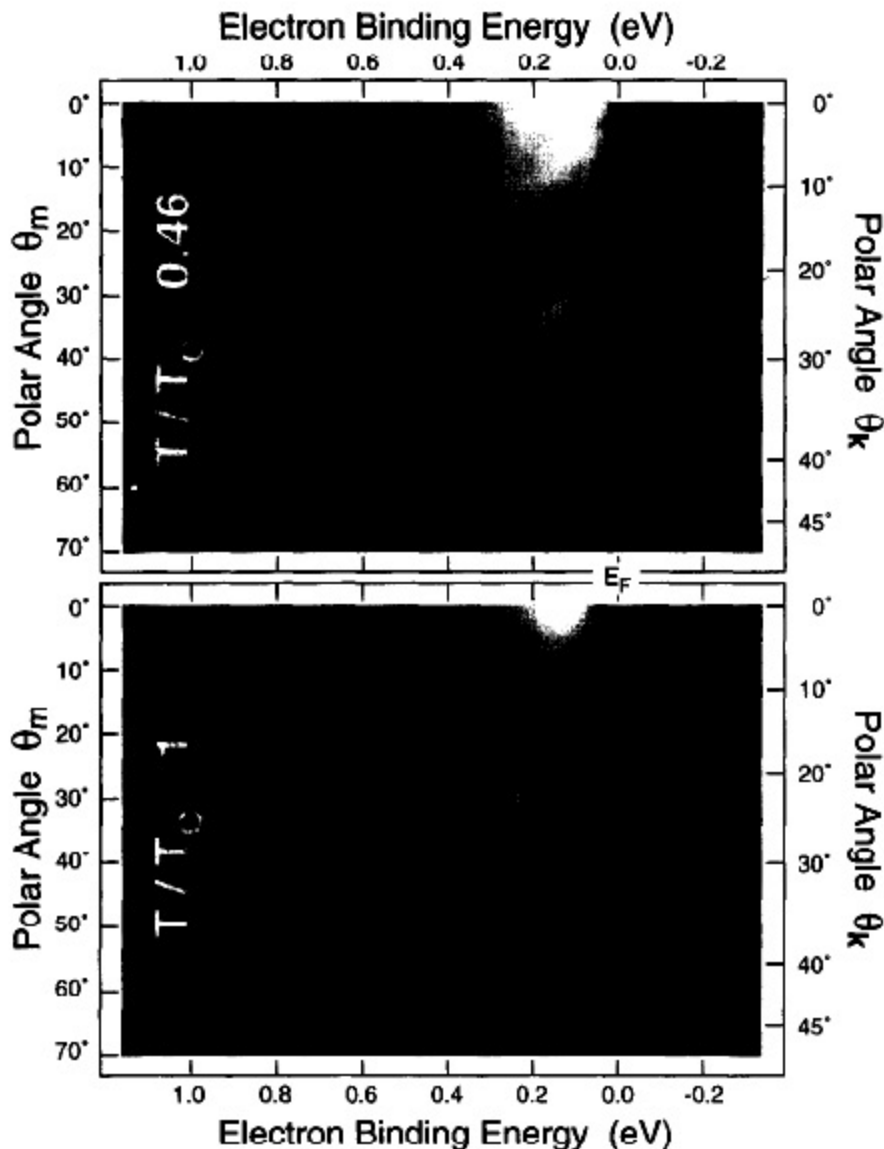


Fig. 2. Linear gray scale representation of the raw data with white as maximum intensity. The upper panel shows the angular scanned EDCs measured at RT, the lower panel the $T = T_C$ data.

ground, unlike the situation in the EDCs (see Fig. 3). The bands turn out to be much better separable in k -space than in energy. In the high temperature polar scan we see a single sharp peak at the " Σ_1 "-position, which shows unambiguously that the exchange splitting has vanished. Without the need of fitting one can, just from the raw data, determine the angular position of the " Σ_2 "- and the two sp -bands with an accuracy of better than $1/2^\circ$, by just locating the peak maxima. This works below E_F as well as up to some 100 meV above E_F . Doing so we find practically identical results as we get from a computer-aided fitting procedure, where we

fit the three peaks with Lorentzians riding on a quadratic background. In addition the fitting delivers peak widths (FWHM) and intensities. Fig. 5 shows results of fits from $\theta_m = 18^\circ$ ($\theta_k = 14^\circ$) to $\theta_m = 70^\circ$ ($\theta_k = 47^\circ$), which omits the bands near the surface normal. In the upper half (a) the intensity data¹⁶ of the polar scan at RT at E_F (crosses) and the fit curve (solid line) are presented. The fit is obviously very good. Below, in Fig. 5 (b), the fitted peak positions for the Σ -bands are presented as extracted from many polar scans at both temperatures. The results of the fitting are summarized in Table 1. At E_F the exchange-split sp -bands are separated by $3.0^\circ \pm 0.2^\circ$ in θ_k or by

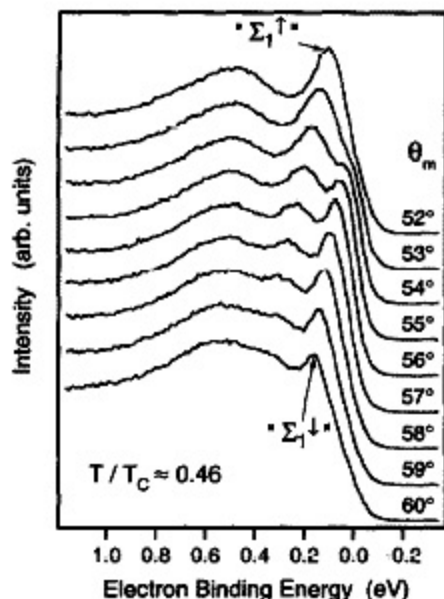


Fig. 3. Energy distribution curves measured at RT for different polar angles θ_m . The exchange-split sp -bands are hardly separable in energy.

(0.123 ± 0.005) a.u. $^{-1}$ in k along the measured polar scan. Making use of the very linear slope of the peak position with energy we can calculate the exchange splitting as $\Delta E_{ex} = (204 \pm 8)$ meV at E_F . We want to emphasize here that this fitting procedure of intensities as a function of angles, done for various energies, takes optimum advantage of the two-dimensional nature of these data sets: The found peak positions in angle can be combined to give very accurate dispersion curves as a function of

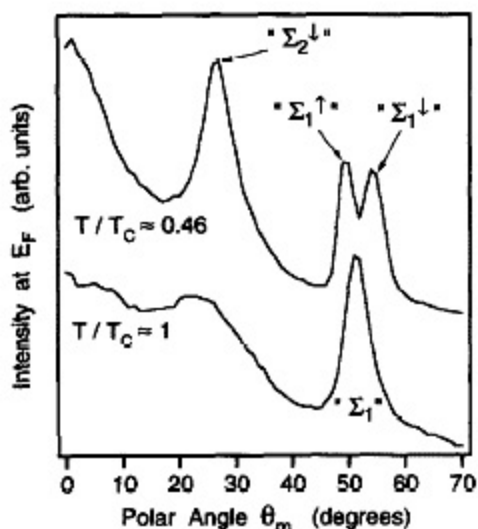


Fig. 4. Polar intensity scans at E_F measured at RT and for $T/T_C = 1$. The two exchange-split sp -bands " Σ_1^{\uparrow} " and " Σ_1^{\downarrow} " are well separated at RT, and near T_C only one sharp peak remains.

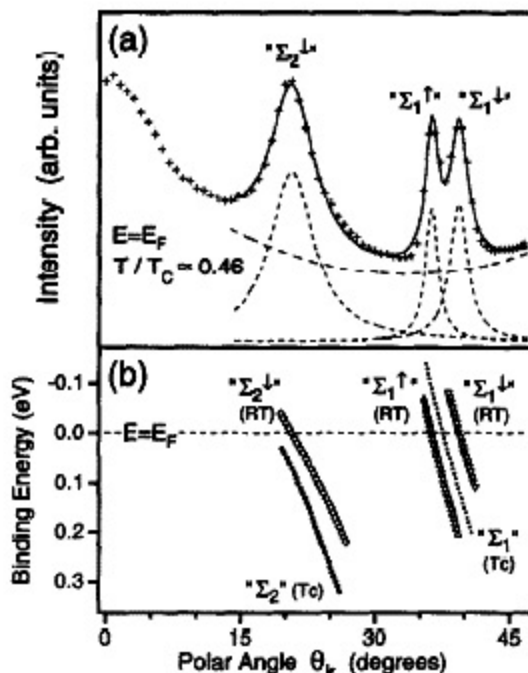


Fig. 5. Fitting the polar intensity scans (see text). In the upper half we present the fit (solid line) for the RT polar scan at E_F . The crosses are the experimental data¹⁶, the broken lines the components of the fit. The lower half shows the peak positions extracted from various polar scans at RT and at T_C .

energy (Fig. 5), from which the exchange splitting can be extracted very precisely.

Another remarkable observation is that the FWHM of the majority sp -band (" Σ_1^{\uparrow} ") is consistently smaller by about 0.3° than that of the " Σ_1^{\downarrow} " peak (not shown). It appears that this cannot be explained by the band structure (dispersion and FS-topology) in conjunction with the finite experimental resolution. And there are also no differences in linewidths due to electron-phonon interactions to be expected, since we detect minority and majority electrons at the same energy. Therefore this issue needs to be discussed under other aspects, like perhaps different final state lifetimes for majority and minority electrons^{5,17}, but this shall not be done in the present paper.

In conclusion we have measured a very complete set of ARUPS energy spectra near the Fermi energy from Ni(110) representing a polar scan with 1° angular steps

Table 1. Results of fitting the polar intensity scans.

Band	Position at E_F θ_k (degrees)	Dispersion in θ_k (meV/degree)	FWHM at E_F (degrees)
" Σ_2^{\downarrow} " (RT)	20.75 ± 0.2	34 ± 4	3.0 ± 0.2
" Σ_2^{\downarrow} " (T_C)	19.2 ± 1.0	45 ± 10	6 ± 2
" Σ_1^{\uparrow} " (RT)	36.4 ± 0.1	71 ± 6	0.9 ± 0.1
" Σ_1^{\downarrow} " (RT)	39.4 ± 0.1	64 ± 6	1.2 ± 0.1
" Σ_1^{\uparrow} " (T_C)	37.65 ± 0.15	66 ± 7	1.45 ± 0.2

along the $[\bar{1}10]$ -azimuth. Comparing the RT-data to a recent FS-calculation¹⁴ a minority d -band and two exchange-split sp -bands in the ΓXWK -plane can be unequivocally assigned. For this assignment, a free electron final state is assumed and appears to be sufficiently accurate. In the energy distribution curves the two sp -bands are poorly separable, but in the polar scans, i.e., in k -space, they clearly show a splitting of $3.0 \pm 0.2^\circ$ in θ_k (0.123 ± 0.005 a.u.⁻¹ in k). Fitting the peak positions in angle allows also to find the exchange splitting $\Delta E_{ex} = 204 \pm 8$ meV at E_F at the measured points in k -space. Furthermore we observe a distinctly larger line width

(in polar angle) for the minority than for the majority sp -electrons. Upon raising the temperature towards T_C the formerly exchange-split bands merge to one band, and the minority d -band broadens considerably. We want to emphasize that all these findings are only possible with a very high angular resolution.

Acknowledgement – Skilful technical support was provided by E. Moser, O. Raetz, H. Tschopp and F. Bourqui, and we want to thank T. Greber for fruitful discussions. This work has been supported by the Schweizerischer Nationalfonds.

REFERENCES

1. S.D. Kevan (ed.), *Angle Resolved Photoemission*. Elsevier, Amsterdam (1992).
2. D.E. Eastman, F.J. Himpsel, J.A. Knapp, *Phys. Rev. Lett.* **40**, 1514 (1978).
3. F.J. Himpsel, J.A. Knapp, D.E. Eastman, *Phys. Rev. B* **19**, 2919 (1979).
4. W. Eberhardt, E.W. Plummer, *Phys. Rev. B* **21**, 3245 (1980).
5. P. Heimann, F.J. Himpsel, D.E. Eastman, *Solid State Commun.* **39**, 219 (1981).
6. R. Raue, H. Hopster, R. Clauberg, *Phys. Rev. Lett.* **20**, 1623 (1983).
7. R. Raue, H. Hopster, R. Clauberg, *Z. Phys. B* **54**, 121 (1984).
8. M. Donath, *Surf. Sci. Rep.* **20**, 251 (1994).
9. H. Capellmann, *J. Magn. Magn. Mat.* **28**, 250 (1982).
10. A. Santoni, L.J. Terminello, F.J. Himpsel, T. Takahashi *Appl. Phys. A* **52**, 229 (1991).
11. P. Aebi, J. Osterwalder, R. Fasel, D. Naumović, L. Schlapbach, *Surf. Sci.* **307-309**, 917 (1994).
12. P. Aebi, J. Osterwalder, D. Naumović, R. Fasel, L. Schlapbach, *Solid State Commun.* **88**, 19 (1993).
13. R. Matzdorf, A. Goldmann, J. Braun, G. Borstel, *Solid State Commun.* **91**, 163 (1994).
14. I. Eckardt, L. Fritsche, *J. Phys. F* **17**, 925 (1987)
15. T.J. Kreuz *et al.*, publ. in prep.
16. Since the electron flux is thinned out for higher polar angles due to the refraction, we corrected the data for this before fitting [D. Naumović, A. Stuck, T. Greber, J. Osterwalder, L. Schlapbach, *Phys. Rev. B* **47**, 7462 (1993)]. However, this does practically not alter the fitting parameters. Corrections for the geometry (light spot, electron detection spot) were not made, because they nearly compensate each other. Lorentzians fitted the data better than Gaussians.
17. H.C. Siegmann, *Surf. Sci.* **307-309**, 1076 (1994)