Observation of Spin-Split Electronic States in Solids by Energy-, Angle-, and Spin-Resolved Photoemission

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The combination of high energy and angle resolution (100 meV, ± 3°) with spin analysis in photoemission spectroscopy is shown to allow the direct observation of magnetically spin-split electronic states. This is demonstrated for normal emission from Ni(110) with polarized Ne I radiation ($h\nu = 16.85$ eV). It is found that $\Delta_{ex} = 0.18$ eV for the exchange splitting of the S_4 band at the X point in good agreement with a band-structure and photoemission calculation that considers the self-energy correction explicitly.

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Angle-resolved photoemission spectroscopy (ARPES) has proved to be a powerful tool in determining the electronic structure of solids. For the itinerant-electron ferromagnet Ni the exchange splitting as well as the d-band width was shown[1,2] to be smaller than in self-consistent band-structure calculations.[3] Better agreement is achieved by taking the self-energy correction into account.[4] ARPES, however, cannot directly identify magnetically spin-split bands. For this, a comparison with band-structure calculations is necessary, as pointed out by Plummer.[5] *A direct experimental observation* of spin-split electronic states and a detailed line-shape analysis can be obtained by adding spin analysis to energy and angle resolution. The feasibility of energy- and spin-resolved photoemission has recently been demonstrated by measuring the spin polarization of the Ni 6-eV satellite.[6]

We report here on the first spin-polarized photoemission experiments on ferromagnetic samples with an energy and angle resolution (100 meV, ± 3°) comparable to that of conventional ARPES. To demonstrate the capability of this technique we have measured the angle- and spin-resolved energy distribution curves for Ni(110) in normal emission with Ne I photoexcitation ($h\nu = 16.85$ eV) at room temperature. We find an exchange splitting of $\Delta_{ex} = 0.18 \pm 0.02$ eV for the S_4 band at the X point of the Brillouin zone.

The experiments reported here were carried out in a specially designed ultrahigh-vacuum apparatus for spin-polarized electron spectroscopies with high energy and angle resolution. The main components are a specially designed electron optics including a hemispherical energy analyzer (180°) coupled to a high-efficiency Mott detector for spin analysis (see Fig. 1). For the present experiment a newly designed high-intensity resonance lamp providing polarized light ($P = 70\%$) in the vacuum-uv region was used.[7] The angle of incidence of the light beam was 30° with respect to the surface normal of the sample which was aligned for normal emission within ± 0.5° with respect to the electron-optical axis of the energy analyzer. The sample was a (110)-oriented Ni single crystal having a so-called "picture-frame" shape with its legs oriented in [110] directions (see inset in Fig. 1). A coil was wound around one of its legs. This geometry allows the measurements to be performed without an external magnetic field (i.e., "in remanence") with minimal magnetic stray fields. Easy reversal of the magnetization is achieved by a current pulse through the coil. The Ni crystal was spark cut and aligned to approximately 1° using standard

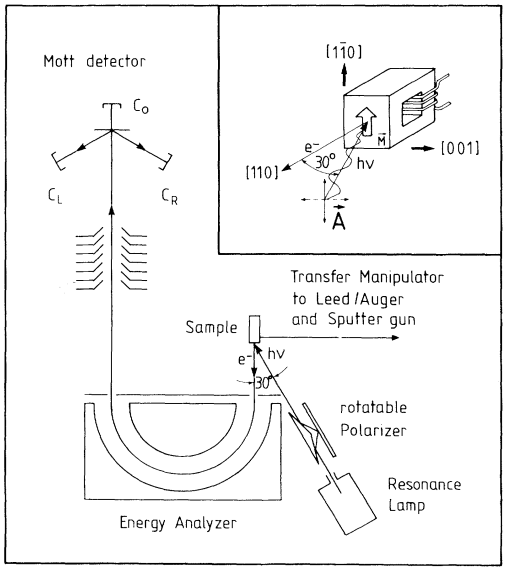

FIG. 1. Schematic of the experimental geometry and the picture-frame Ni single crystal (inset).

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Laue techniques. It was cleaned *in situ* by Ne-ion sputtering and heating cycles with subsequent flashing. The orientation of the surface and the cleanliness were checked with low-energy electron diffraction and Auger-electron spectroscopy. The base pressure in the system was 2×10^{-10} Torr which increased to 1×10^{-8} Torr during lamp operation. With a quadrupole mass spectrometer we ensured that the pressure increase was due only to Ne. We observed no detectable influence on the surface cleanliness.

In Fig. 2 we show the angle-resolved energy distribution curves (EDC's) $I_0 = I_\uparrow + I_\downarrow$, along with the *simultaneously measured* spin polarization $P = (I_\uparrow - I_\downarrow)/(I_\uparrow + I_\downarrow)$ for the electric field vector parallel to the $[1\bar{1}0]$ and [001] directions, respectively. The displayed spectra have been corrected for the Ne I doublet and the incomplete light polarization.[8] The spin-polarization data have been smoothed by averaging over three points. This procedure was not necessary for the EDC's which were measured in forward scattering in the Mott detector (counter C_0 in Fig. 1) allowing better statistics because of the higher intensity.

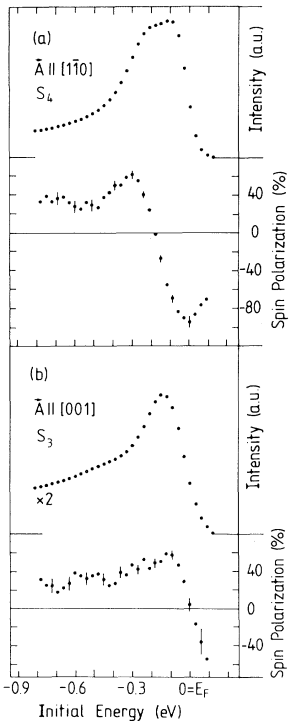

FIG. 2. Angle-resolved EDC's and corresponding spin-polarization curves for normal emission from Ni(110) for the electric field vector (a) in the $[1\bar{1}0]$ direction and (b) mainly in the [001] direction (see text).

Figure 2(a) shows the EDC and the corresponding spin-polarization curve for the electric field vector in the $[1\bar{1}0]$ direction. According to the optical dipole selection rules,[9] only bands with S_4 symmetry can be excited. The EDC shows a slight dip near 0.16 eV below E_F indicating the existence of two peaks. The spin-polarization curve, on the other hand, exhibits large variations reaching from almost -100% near E_F to approximately $+60\%$ near 0.3 eV below E_F. This is a *direct proof* that the two peaks in the EDC correspond to magnetically split bands of S_4 symmetry.

Figure 2(b) shows the EDC and the corresponding spin-polarization curve for the electric field vector parallel to the [001] direction {apart from a minor component in the [110] direction due to the angle of incidence of 30° (see inset in Fig. 1)}. The main intensity originates from bands with S_3 symmetry, but weak emission from bands with S_1 symmetry is possible.[10] The EDC exhibits a broad peak centered near 0.1 eV below E_F. The corresponding spin polarization is positive below E_F and exhibits a maximum near -0.1 eV indicating that the main intensity originates from the S_3 majority-spin band.

Figure 3 displays the spin-resolved contributions to the EDC's $I_\uparrow = I_0(1+P)/2$ and $I_\downarrow = I_0(1-P)/2$, obtained therefore without further assumptions from the experimental data of Fig. 2. To determine the actual peak positions we have analyzed the line shape. Following Jepsen, Himpsel, and Eastman[11] and Clauberg[10] we have used an asymmetric line shape [Doniach-Sunjic (DS)], as was also used by Maetz *et al.*[12] The DS line was multiplied with the Fermi function and convoluted with the resolution function of the apparatus, which we assumed to be a Gaussian of 0.1 eV full width at half maximum. A least-squares fitting routine yielded the peak positions which are indicated by tickmarks in Fig. 3.

Figure 3(a) shows the spin-up (majority) and the spin-down (minority) EDC's for the electric field vector in the $[1\bar{1}0]$ direction (only bands with S_4 symmetry contribute to the spectra). We have determined the peak positions as 0.06 and 0.24 eV below E_F for the S_4 minority- and majority-spin bands, respectively. The main intensity in the spectra arises from direct transitions occurring at the same k point into a band gap (see below). The difference in the peak positions thus directly reflects the exchange splitting $\Delta_{ex} = 0.18$ eV in the case of the S_4 bands.

Figure 3(b) shows the spin-resolved EDC's for

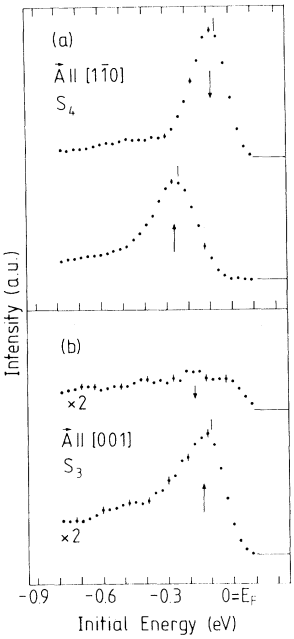

FIG. 3. Angle- and spin-resolved EDC's for normal emission from Ni(110) (a) for the electric field vector in the [1$\bar{1}$0] direction showing directly the exchange splitting of the S_4 band at the X point, and (b) for the electric field vector mainly in the [001] direction showing the energy position of the S_3 majority-spin band at the X point. Tickmarks indicate peak positions obtained by a least-squares fitting procedure (see text).

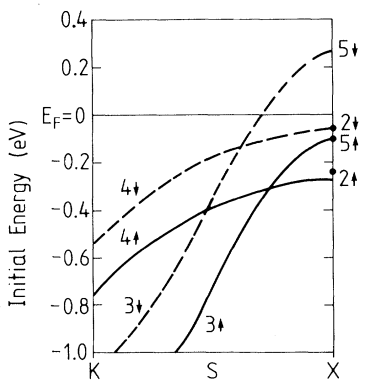

FIG. 4. One-dimensional ferromagnetic band structure along the $K(S)X$ direction including self-energy corrections (see text). Only bands which are allowed initial states for normal emission are shown. Dots indicate experimental values.

the electric field vector in the [001] direction. The spin-up EDC exhibits one peak originating from emission of the S_3 majority-spin band located at about 0.1 eV below E_F. The spin-down EDC shows only a weak broad structure. No peak position can really be determined. This part of Fig. 3 will be discussed in detail elsewhere.[10]

According to band-structure and photoemission calculations of Clauberg,[10] which take into account the self-energy correction as calculated by Liebsch,[4] in normal emission from Ni(110) with photon energies of $h\nu = 16.85$ eV the photoemission intensity is mainly due to direct transitions from S_4 and S_3 bands at the X point into the band gap between the $X_5^\uparrow$ and the X_3 point. Identifying our experimentally determined peak positions with the energy positions of critical points we find $X_2^\downarrow = -0.06$ eV, $X_2^\uparrow = -0.24$ eV, and $X_5^\uparrow = -0.10$ eV. For a more quantitative comparison with the calculation, e.g., one critical point of the band structure has to be adjusted, because only relative band positions can be calculated at present. In this case the $X_5^\uparrow$ point (−0.10 eV) was taken from the experiment. For the spin-split X_2 point the calculation then yields $X_2^\downarrow = -0.06$ eV and $X_2^\uparrow = -0.27$ eV in good agreement with experimental values (see Fig. 4).

Our results are in agreement with recent ARPES data of Heimann *et al.*[13] ($X_2^\downarrow = -0.04$ eV, $X_2^\uparrow = -0.24$ eV, and $X_5^\uparrow = -0.11$ eV), but contradict those of Eberhardt and Plummer,[2] who quote a value of -0.85 ± 0.1 eV for the spin-unresolved X_2 point.

In a previous study on the spin polarization of the secondary electrons[14] it was shown that the mean free path of the electrons can be spin dependent leading to an energy-dependent enhancement of the spin polarization. The present data suggest that spin-dependent electron scattering is negligible in the present energy range in agreement with the data of Ref. 14 which show no enhanced spin polarization between 16 and 17 eV above E_F.

In summary, we have demonstrated the feasibility of spin-polarized photoemission with high energy and angle resolution, allowing the experimental determination of ferromagnetic electronic structures. The experimentally observed exchange splitting for e_g states of 0.18 eV in the case of Ni as well as the energy position of the bands at the X point between E_F and −0.8 eV is in good agreement with a band-structure and photoemission calculation taking the self-energy correction explicitly into account. The technique presented here, which can be viewed as the "complete" photoemission experiment for the study of ferromagnets, will allow solution of problems such as the unambiguous identification of magnetic

surface states[15,16] and especially the current controversy concerning the temperature dependence of the exchange splitting,[1,12] yielding thereby new insights into the ferromagnetic-paramagnetic transition and related changes in the electronic structure.

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[8]For the correction with respect to the Ne I doublet ($h\nu$ = 16.85 and 16.67 eV) we used a procedure suggested by H. Neddermeyer making the reasonable assumption that two identical spectra are superimposed and shifted by 0.18 eV with respect to each other. The relative line intensities of the doublet were determined with the aid of the Fermi edge of a polycrystalline Au film. This correction was first performed on the spin-resolved EDC's which have been calculated from the uncorrected EDC and spin polarization P: $I_{\uparrow} = I_0(1 + P)/2$, $I_{\downarrow} = I_0(1 - P)/2$. The corrected EDC and spin polarization have then been recalculated. The correction for the incomplete light polarization was based on the fact that the measured spectra are a superposition of the "true" spectra weighted according to the light polarization.

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