

## Observation of $-100\%$ Spin-Polarized Photoelectrons from a Transversely Magnetized Ni(110) Single Crystal

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We have measured for the first time the electron spin polarization  $P$  of photoelectrons emitted from a transversely magnetized sample in the absence of an externally applied magnetic field. With this photoemission geometry, use has been made of optical selection rules for selectively exciting spin-polarized electron states of different symmetry as illustrated for emission from the  $\Sigma_3$  and  $\Sigma_4$  bands of Ni(110), where values of  $P$  up to  $-100\%$  have been observed.

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In this Letter we demonstrate for the first time that spin-polarized photoemission experiments from ferromagnets can successfully be performed with a transversely magnetized sample (i.e., *transverse* geometry) without applying an external magnetizing field  $\vec{H}_e$  during the measurements. The long-standing belief that a *longitudinal* geometry (i.e.,  $\vec{H}_e$  parallel to the electron optical axis) is a necessary condition in spin-polarized photoemission studies does therefore not hold true any more.<sup>1</sup> As a further consequence, many spin-polarized photoemission experiments (especially energy-resolved spin-polarized work) will be much more easily performed.

We use this new method for investigating Ni(110) and report for the first time effects on the spin-polarization spectra of a ferromagnetic metal by selective excitation of electronic energy bands on the same single-crystal surface.

Conventional angle-resolved photoemission spectroscopy (ARPES)<sup>2,3</sup> and spin-polarized photoemission studies<sup>4-6</sup> of single-crystal Ni recently yielded definite experimental evidence that the exchange splitting  $\Delta$  and the Stoner gap  $\delta$  (distance of the top of the majority-spin  $d$  band from the Fermi energy  $E_F$ ) are only about half of the value or smaller than that predicted by self-consistent band calculations. The observed negative spin polarization at photothreshold is already predicted by the simple Stoner-Wohlfahrt-Slater theory of ferromagnetism,<sup>7,8</sup> but the value of the crossover energy  $E_c$  from negative to positive spin polarization is a more difficult problem and has been the subject of several theoretical investigations.<sup>9-11</sup>

Recently, the first energy-resolved spin-polarization experiments on Ni(111) with synchrotron radiation from the ACO storage ring at Laboratoire pour l'Utilisation de Rayonnement Electromagnétique (LURE), Orsay, up to 40 eV<sup>12</sup>

showed that it is possible to detect spin-polarized bands in ferromagnetic metals at binding energies beyond 0.5 eV below the Fermi energy, a range where ARPES can no longer distinguish spin up and spin down, primarily because of lifetime effects.

After the failure of the first and other spin-polarized photoemission experiments performed with the so-called "transverse" geometry,<sup>13,14</sup> all the subsequent successful studies were performed with the so-called "longitudinal" geometry, as was the first successful one.<sup>15</sup> In all these studies the direction of emission of the photoelectrons (i.e., the angle information) is completely lost in the strong magnetic field<sup>1</sup> and a (differential) energy analysis is made very troublesome because of the occurrence of transverse velocity components when extracting the photoelectron beam out of the magnetic field.<sup>16,17</sup> Only by taking advantage of the fact that the electrons emitted with maximum kinetic energy are leaving the crystal along the surface normal, angle and energy analysis has been performed recently with use of the retarding-field energy analyzer.<sup>12</sup> With the transverse geometry (magnetization perpendicular to the electron optical axis) the above-mentioned electron-optical problems, which are specific for the longitudinal geometry, are not present.

In this Letter we show that, in contrast to common belief and to the findings of earlier experiments, the transverse geometry can be used successfully in spin-polarized photoemission experiments,<sup>18</sup> for electron kinetic energies down to the millielectronvolt region.

The apparatus used is a modification of the one described by Kisker *et al.*<sup>1</sup> A new electron-lens system, schematically shown in Fig. 1, was constructed in order to collimate the photoelectrons effectively since the (initially) focusing longitudi-

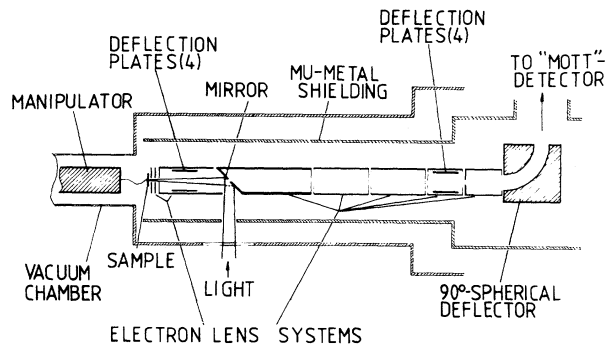


FIG. 1. Scheme of the extraction optics used with the *transverse* magnetization geometry, i.e., the magnetization is at  $90^\circ$  with respect to the electron-optical axis.

nal magnetic field is missing. The lens system also provides a rapid acceleration of the photoelectrons in order to reduce deflection and spin precession due to the magnetic stray fields.

The sample is shaped as a thin, long plate of nominal dimensions  $0.3 \times 3 \times 12 \text{ mm}^3$ , magnetized parallel to the longer side which, in our case, corresponds to the  $[1\bar{1}1]$  direction, which is an easy magnetization direction. The  $[110]$  direction was chosen to be perpendicular to the emitting surface and parallel to the light beam (see Fig. 2, inset). We found that it was not even necessary to use a closed ferromagnetic circuit, i.e., it was unnecessary, in the present experiment, to further minimize the transverse magnetic stray field because of the rapid electron acceleration. The stray field, as measured with a "Förster-Sonde" placed at the middle of the sample, was 1 Oe. Because of the magnetic anisotropy the magnetic domains are aligned in the surface (i.e., the hysteresis loop is nearly rectangular, as it was detected by magneto-optic Kerr effect). No external magnetic field was applied during the spin-polarization measurements. The sample is at room temperature during the measurements. It can be rotated around the  $[110]$  direction (surface normal) in order to adjust a specific azimuthal angle of the crystal orientation with respect to the electric vector of the linearly polarized light. With the Mott detector<sup>1</sup> we measure the vertical spin-polarization component which is parallel to the emitting surface and parallel to the electric field vector  $\vec{E}$  of the light (whose direction is vertical and fixed in space). When the magnetization direction (i.e., the  $[1\bar{1}1]$  of Fig. 2) is not parallel to the  $\vec{E}$  vector (not vertical) the measured spin polarization  $P$  of the corresponding crystallographic direction is reduced

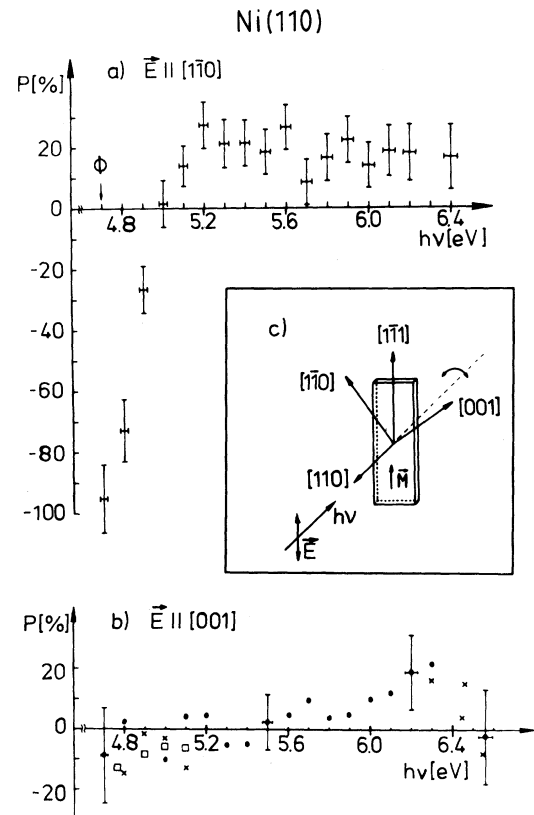


FIG. 2. The spin polarization of the total photoyield as a function of the photon energy for directions of the light electric vector  $\vec{E}$  parallel to the  $[1\bar{1}0]$  and to the  $[001]$  direction, respectively, corresponding to  $\Sigma_4$ - and  $\Sigma_3$ -band symmetry excitation. The statistical errors are shown as vertical error bars. The inset shows the sample geometry. The light is impinging normally to the  $(110)$  surface.

from the actual  $P_0$  value by  $\cos\theta$ . The angle  $\theta$  is between the  $\vec{E}$  vector (the vertical) and the  $[1\bar{1}1]$  direction.

In view of future differential energy analysis and in order to obtain a large optical angular aperture, the light path was decoupled from the electron-optical axis by means of a mirror placed close to the sample and tilted by  $45^\circ$  (see Fig. 1). In order to transmit the electron beam the mirror has an aperture of 1 mm diameter onto which the electron beam is focused with the help of an aperture-lens system of very short focal length. By means of a tube-lens system the electron beam is transported into the spherical condenser.<sup>19</sup> The experimentally determined efficiency of the beam transport from the photocathode to the Mott detector is about 0.1 and the loss in the

Mott scattering process is about  $10^{-4}$ . The magnetization direction can easily be reversed by bringing a small permanent magnet close to the sample in the desired direction with the sample retracted into the small tubing of the vacuum chamber (see Fig. 1). The count rate in the Mott detector is nearly the same as in the experiments with a longitudinal geometry. Notably, the optimal electron transmission is obtained in the transverse geometry for 30 eV pass energy of the spherical condenser, compared to 1000 eV in the experiment with the longitudinal magnetic field applied.<sup>4,5</sup> The differential energy resolution, obtained at this pass energy, is 0.8 eV full width at half maximum, with the Mott detector apertures acting as the analyzer exit slits.<sup>16</sup> As the light source we used a combination of a 500-W xenon arc lamp (Hanovia) and a grating monochromator (Jobin Yvon H20), yielding an optical bandwidth of 40 meV at 5 eV photon energy with 0.5 mm slit width. Because of the three reflections in the monochromator and the one at the mirror near the sample the light is more than 80% linearly polarized with the electric field vector parallel to the mirror planes.

The measured dependence of the spin polarization on photon energy is shown in Figs. 2(a) and 2(b) for the light polarization  $\vec{E}$  parallel to the  $[1\bar{1}0]$  and to the  $[001]$  direction, respectively. It follows from the optical selection rules<sup>20</sup> that, for normal incident light and normal emission, only initial states with  $\Sigma_4, S_4$  and  $\Sigma_3, S_3$  symmetry are allowed along the  $\Gamma-K-X$  direction (see Fig. 3). We ascribe the dramatic difference of the spectra of Figs. 2(a) and 2(b) to the selection rules for emission from the  $\Sigma_4$  and  $\Sigma_3$  bands, respectively.

Reliable empirical band-structure information for the  $\Gamma-K-X$  direction of the Ni Brillouin zone, which is sampled in the present experiment, does not yet exist. Correspondingly, no calculation of the spectral dependence of the spin polarization is available. The discrepancies between the theories, based on self-consistent band-structure calculations, and the experiments, are quite well known for Ni(100) and Ni(111).<sup>2,5,6</sup> Without any calculation, it can, however, be stated that our data on Ni(110) are also not compatible with the existing knowledge on the band structure: According to the estimate that  $E_c \sim 2\delta$ ,<sup>8</sup> it follows from Fig. 2(a) that  $X_2^\uparrow \sim 0.15$  eV below  $E_F$ . This value is much smaller than that obtained from self-consistent calculations ( $X_2^\uparrow \sim 0.47$  eV<sup>21</sup>), and even smaller than that obtained from the semi-empiri-

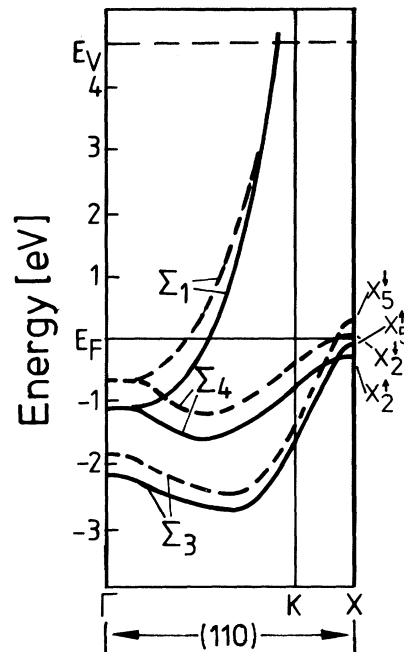


FIG. 3. Electronic structure of Ni along  $\Gamma-K-X$  direction in the Brillouin zone, as determined by ARPES (Ref. 2) and semiempirical calculations (Ref. 11). Only those bands are shown which are "dipole" allowed for  $s$  polarization and normal emission.

cal calculation of Smith and Chiang<sup>11</sup> ( $X_2^\uparrow \sim 0.3$  eV below  $E_F$ ).

From the value of the threshold spin polarization for  $\Sigma_3$  excitation [Fig. 2(b)] we obtain information on the position of  $X_5^\uparrow$ . Since  $P \sim -10\%$ , we excite, because of the limited optical resolution ( $\Delta E = 40$  meV), not only the minority-spin bands, but also majority bands. We therefore conclude that  $X_5^\uparrow \leq 40$  meV. This value is in agreement with the theoretical findings of Ref. 6, and specifies the experimental value of Ref. 2 ( $X_5^\uparrow = 0.1_{+0.05}^{-0.1}$ ).

The value of the exchange splitting can only be derived from spin-polarized photoemission data on the basis of a rigorous calculation. Especially the (110) face of Ni represents a stringent test of the photoemission calculations since, in contrast to Ni(100) and Ni(111), dipole-allowed final states exist at the vacuum level. For the (100) and (111) surfaces, band-gap emission was the only possible mechanism, so that a very simple model,<sup>7,8</sup> based on the density of states, was also able to describe the data in a reasonable way. A proper calculation for Ni(110) is therefore strongly needed, to provide a consistent description of this prototype ferromagnet.

We have shown for the first time that spin-po-

larized photoemission experiments from ferromagnetic materials can be performed with use of transversely magnetized samples. Since the stray magnetic field of the samples can be made negligibly small by using either very thin or large samples, or even ring-shaped ones as proposed by Saldaña and Helman,<sup>22</sup> energy- and angle-resolved spin-polarized photoemission experiments are now feasible. Since synchrotron radiation can also be used in spin-polarized photoemission studies, as was recently shown,<sup>12</sup> this opens the way to novel possibilities in photoemission spectroscopy.

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<sup>1</sup>For a recent review on this field, see E. Kisker, M. Campagna, W. Gudat, and E. Kuhlmann, in *Festkörperprobleme XIX* (Max-Planck-Institut für Festkörperforschung, Stuttgart, 1979), p. 259.

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<sup>18</sup>A transverse sample magnetization has been used in a spin-polarized low-energy electron-diffraction (LEED) experiment by R. J. Celotta, D. T. Pierce, G. L. Wang, S. O. Bader, and G. P. Felcher, *Phys. Rev. Lett.* **43**, 728 (1979). This experiment cannot directly be compared with the present one because of the very different kinetic-energy regimes involved. In spin-polarized photoemission near threshold the electrons are very slow ( $E_{\text{kin}} \geq 20$  meV) and spin precession and depolarization could occur. In the LEED experiment the electrons had kinetic energies larger than 20 eV.

<sup>19</sup>The aperture-type lens system was designed with the help of the electron-optical program of W. B. Hermannsfield, SLAC Report No. 166, 1973 (unpublished). The tube-lens system has been designed with the help of the fast-running ray-tracing program of J. Fink and E. Kisker, *Rev. Sci. Instrum.* **51**, 918 (1980).

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