tem of copper clearly favors antiferromagnetism but the question whether a transition occurs above 50 nK must be decided by future investigations.

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Enhancement of the Electron Spin Polarization in the Photoyield of Ni(111) in the Vacuum Ultraviolet

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We have found that the spin polarization of the photoyield from single crystal Ni(111) at 16.8- and 21.2-eV photon energy is $(8.0\pm3.5)\%$ and $(8.1\pm2)\%$, respectively, which is significantly larger than the bulk electron-spin polarization. The results of a theoretical model calculation are in good agreement with the experiment. We interpret this as direct evidence for spin-dependent electron-electron scattering in an itinerant strong ferromagnet.

Almost ten years after the first observation of electron spin polarization (ESP) in photoemission from Gd thin films in the photon energy range $h\nu \leq 5.5$ eV,[1] we report the first observation of ESP in photoemission from a ferromagnetic material (Ni) using photon energies in the near vacuum ultraviolet (vuv), namely, at 16.8 and 21.2 eV. We find that the ESP of the photoyield at these photon energies is positive (i.e., magnetic moment parallel to the magnetization) and significantly larger ($\sim 8.1\%$ at 21.2 eV) than the bulk band contribution to the magnetization and giving a spin polarization $P=n_B/n \simeq 0.51/10 \simeq 5.1\%$ of bulk Ni at room temperature. These data are the encouraging results of initial steps of a major effort in our laboratory towards energy-resolved, photon-energy–dependent ESP measurements using tunable vuv radiation.

The results on Gd have been interpreted within the three-step model of the photoemission process and a direct correlation of the photoelectron-spin polarization with the spin polarization of the conduction electron has been suggested.[1] Later, De Wames and Vredevoe[2] pointed out that inelastic scattering from magnons during escape could significantly alter the initial ESP after photoexcitation. In subsequent experiments on Gd and Dy, Bänninger *et al.*[3] found evidence that effects of electron-magnon scattering were negligible. Since then most experimental results on ESP of photoelectrons have been interpreted without considering spin-flip processes,[4] especially because of the small photon-energy range used in the experiments done so far.

The first photoelectron-spin polarization data on single-crystal 3*d* ferromagnets were obtained

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on Ni(001) surfaces.[5] These experiments, which test primarily bulk properties of Ni because of the large escape depth of the photoelectrons near threshold, show a change of the ESP from about − 30% to + 40% with a crossover at about 50 meV above threshold. These, and more recent angle-resolved vuv photoemission studies[6] of single-crystal Ni cannot be directly related to the most complete *ab initio*, self-consistent band-structure calculations of Ni,[7] which predict, e.g., a magnetic exchange splitting twice as large as the observed one ($\Delta E_{\rm exch} \sim 0.35$ eV).[6] The observation of negative ESP near the Fermi energy E_F solves at least qualitatively a long-standing puzzle regarding the prediction of the photoelectron-spin polarization within the band model for a strong ferromagnet like Ni.[8,9]

A scheme of the source part of the novel apparatus is shown in Fig. 1. A detailed report on this novel setup optimized to meet the specific requirements for an electron-spin and -energy analysis will be presented elsewhere.[10] Photons from a rare-gas resonance-line light source impinge in normal direction onto the (111) face of the cylindrically shaped Ni sample. The 5*N*-purity (99.999% purity) Ni single crystal is spark cut to a diameter of 3 mm, electrochemically etched and cleaned by *in situ* successive argon-ion sputtering and electron-bombardement heating cycles at a base pressure of about 2×10^{-10} Torr. The cleanliness and the crystal structure of the surface have been examined by Auger spectroscopy and low-energy electron diffraction (LEED), respectively. In Fig. 1, the sample is shown in front of the electron-extraction optics, in the homogeneous part of the external field which is generated by an iron-shielded electromagnet. The field defines the quantization axis, i.e., it serves to align the Weiss domains. For a complete analysis of the photoelectron-spin polarization from ferromagnets, one has to know (1) the trajectories of the photoelectrons in the inhomogeneous magnetic and electric fields, (2) the magnitude of the electron-spin precession for the various trajectories, and (3) the electron-beam characteristics after extraction from the magnetic field. We will discuss these effects, which we have analyzed numerically for our design, separately.[10] Here, we only point out that the size of the illuminated spot on the sample is of crucial importance for an efficient extraction of the photoelectrons from the magnetic field region. This because the photoelectrons generated at a distance r_0 from the axis of the magnetic field have a transverse energy $E_\perp$ which is proportional to the square of the magnetic field B and to the fourth power of r_0. In the inhomogeneous magnetic field (basically the fringe field), the electron spins rotate and precess with a Larmor frequency proportional to the radial magnetic field component, thus giving rise to depolarization effects. It can be shown numerically,[10] and this is confirmed experimentally, that this effect is minimized by passing the fringe fields with sufficiently energetic electrons. Similar arguments hold also for the magnetic stray field of the cylindrically shaped Ni sample. After having changed the longitudinal polarization of the photoelectrons to a transverse one by means of a 90° spherical deflection analyser, the ESP is

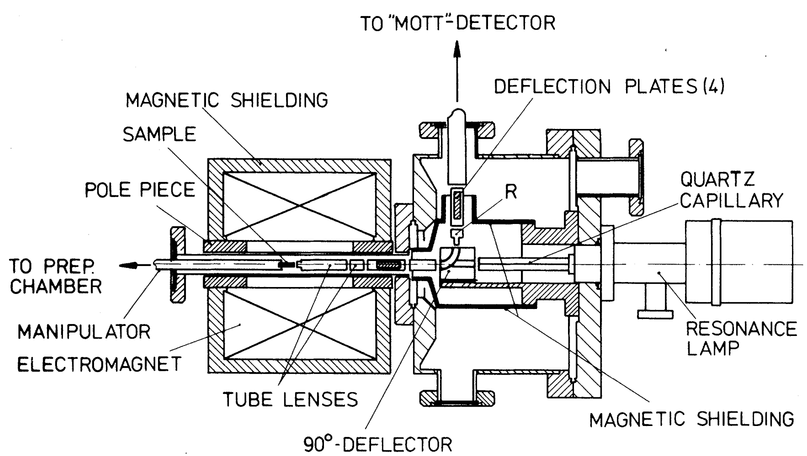

FIG. 1. A scheme of the source part of the novel apparatus optimized for electron-spin and -energy analysis is shown. For details, see text.

determined by Mott scattering.[1]

The photoelectron-spin polarization data for the Ni(111) surface at an applied magnetic field of 2 kOe are given in Table I. The ESP value at 21.2-eV photon energy as well as the one obtained at 16.8 eV is larger than the one expected by using the model of De Wames and Vredevoe,[2] which predicts that the conduction-band polarization could be measured directly by polarization measurement of high-energy photoelectrons. We have furthermore observed that at a magnetic field of 500 Oe the photoelectron-spin polarization is very small. At this field value, the Ni sample is known to be magnetically saturated in the bulk as it was deduced from magnetization measurements. This observation implies that the magnetization of the probed region is not yet saturated at 500 Oe even if the bulk has already reached saturation.

For the interpretation of the data of Table I, we have to note that at 16.8 and 21.2 eV a considerable amount of inelastically scattered electrons contributes to the total photocurrent. Estimates based on energy distribution curves show that about 70% at 21.2 eV and about 50% at 16.8 eV of the emitted yield is indeed due to scattered electrons. For excitation energies below the (bulk) plasmon threshold, the main loss mechanisms are electron-electron and electron-magnon scattering (electron-phonon scattering being disregarded as being weak). Electron-magnon scattering is clearly spin dependent, but there is evidence that the corresponding mean free path[4] at an electron energy of about 20 eV above E_F exceeds by at least an order of magnitude the mean free path for electron-electron scattering ($\lambda_{ee} \sim 5$ Å). The latter is thus the dominant interaction mechanism.[11] We show now that λ_{ee} is, contrary to common belief in earlier work,[4] a strongly spin-dependent quantity for low-energy electrons in an itinerant ferromagnet.

The scattering of an excited electron at an energy below about 50 eV by a conduction-band electron is (to a very good approximation) isotropic in the center-of-mass system.[12] Antisymmetry of the total wave function then implies

TABLE I. Experimental and theoretical spin polarization of the photoyield (for selected photon energies). Legend: P_0 is theoretical spin polarization of the elastically emitted (unscattered) electrons; P_1 is theoretical spin polarization of the photoelectrons which have undergone *one* scattering event; P_{tot} is theoretical spin polarization of the photoelectrons obtained after 0 and 1 scattering event; and P_{expt} is experimental spin polarization of the photoyield.

$h\nu$ (eV)	P_0 (%)	P_1 (%)	P_{tot} (%)	P_{expt} (%)
11.2	9.66	5.91	8.56	8.0±6[a]
16.8	10.48	5.92	7.97	8.0±3.5
21.2	10.49	5.94	7.73	8.1±2

[a]Ref. 5.

that the triplet-scattering amplitude vanishes, i.e., scattering occurs only between electrons of opposite spin orientation. In a ferromagnet, excited electrons with spin down are therefore scattered only by majority-spin electrons (which we label as with spin up.) As a consequence the mean free path $\lambda_{ee}{\downarrow}$ for spin-down electrons is shorter than $\lambda_{ee}{\uparrow}$ for spin-up electrons. Since a proper many-body calculation of $\lambda_{ee}{}^{\sigma}$ is yet not available, we make for the purpose of a model calculation the plausible assumptions:

$$\lambda_{ee}{\uparrow}/\lambda_{ee}{\downarrow} = n_b{\uparrow}/n_b{\downarrow}, \quad 1/\lambda_{ee}{\uparrow} + 1/\lambda_{ee}{\downarrow} = 2/\lambda_{ee}, \tag{1}$$

where $n_b{\uparrow}$ ($n_b{\downarrow}$) is the number of the spin-up (spin-down) electrons (per atom) and λ_{ee} is the mean free path for the paramagnetic state of the system. In order to study the spin polarization of the photoyield, we use the previously mentioned step model for the photoemission process and we distinguish between electrons that undergo 0,1,2, etc., scattering events with conduction electrons. For photon energies up to 21 eV, the dominant contribution to the secondary electron current stems from singly scattered electrons.[13] Denoting by N^{σ} the internal distribution of electrons excited by the photon energy $h\nu$, by $S_0{}^{\sigma}$ and $S_1{}^{\sigma}$ the external distribution of electrons unscattered and scattered one time, respectively, we have[14]

$$S_0{}^{\sigma}(E,h\nu) = T^{\sigma}(E) N_0{}^{\sigma}(E,h\nu), \quad T^{\sigma}(E) = \tfrac{1}{2}\{1 - [V_0/(V_0+E)]^{1/2}\}\alpha\lambda_{\sigma}, \tag{2}$$

$$S_1{}^{\sigma}(E,h\nu) = \tfrac{1}{2} T^{\sigma}(E) \sum_{\sigma} \int_E^{h\nu} dE' p(E,E') N_0{}^{\sigma'}(E',h\nu)\left[1 - \frac{\lambda_{\sigma}(E)}{\lambda_{\sigma'}(E')} \ln\left(1 + \frac{\lambda_{\sigma'}(E')}{\lambda_{\sigma}(E)}\right)\right], \tag{3}$$

where V_0 is the inner potential (13.5 eV), E the kinetic energy (referred to the vacuum level), and $p(E,E')$ the probability for scattering an electron from E' to E. We note that $p(E,E')$ corresponds to pair production and is therefore normalized to 2. The spin splitting of V_0 is negligible in the present context, since it is at most of the order of the exchange splitting. The corresponding spin polarization distributions P_0, P_1, and P_{tot} are then obtained as

$$P_i(E,h\nu)=[S_i^{\uparrow}(E,h\nu)-S_i^{\downarrow}(E,h\nu)]/[S_i^{\uparrow}(E,h\nu)+S_i^{\downarrow}(E,h\nu)],\quad i=0,1,$$
$$P_{tot}(E,h\nu)=P_0[\sum_\sigma S_0^\sigma/(\sum_\sigma S_0^\sigma+\sum_\sigma S_1^\sigma)]+P_1[\sum_\sigma S_1^\sigma/(\sum_\sigma S_0^\sigma+\sum_\sigma S_1^\sigma)]. \quad (4)$$

From Eqs. (1)–(4), it is apparent that as a consequence of the spin dependence of the mean free path, P_0 is strongly enhanced compared to the bulk spin polarization $P_b=(N_0^{\uparrow}-N_0^{\downarrow})/(N_0^{\uparrow}+N_0^{\downarrow})$, while P_1 should attain a value not too far from P_b. In order to obtain a quantitative estimate for P_{tot}, however, a numerical calculation is needed. The excitation function $N_0^\sigma(E,h\nu)$ depends on matrix elements and joint density of states for optical transitions. However, we are mainly interested in the influence of the transport process of the spin polarization of the outgoing electrons; therefore, we assume that the photon $h\nu$ simply shifts the initial density of states N_i in energy by $h\nu$: $N_0^\sigma(E,h\nu)\propto N_i(E-h\nu+\varphi)$. For N_i, we choose a simple analytical fit to the density of states given by Moruzzi, Janak, and Williams,[7] such that we have a total band width of 6 eV and an exchange splitting of 0.4 eV. For the work function, we use $\varphi=5$ eV. The spin average $\lambda(E)$ [Eq. (1)] was obtained by least-squares fit to compiled experimental data points.[15] Numerical results are shown in Table I.

As expected, our numerical calculations show a strongly enhanced P_0 contribution to the ESP (with respect to the bulk ground-state polarization P_b) and a P_1 contribution close to P_b, and a total ESP of the photoyield slightly enhanced. This is in encouraging agreement with the experimental fundings at 21.2 and 16.8 eV and it clearly demonstrates the importance of electron-electron scattering in determining the ESP of photoelectrons. In view of its strong effect on P_0, electron-electron scattering should be important also at lower photon energies. In fact, our calculation is in agreement with the experiment at 11.2-eV photon energy.[5]

Now that there is evidence for the spin dependence of electron-electron scattering, it is desirable to compare accurate experimental results over a wide spectral range with theoretical predictions. Obviously the latter should include appropriate joint-density-of-states and matrix-element effects, as well as contributions to (4) from higher-order scattering processes. Work along these lines is in progress and will provide detailed information on the spin dependence of the electron-electron scattering in ferromagnets and thereby (eventually) also on the magnetization in the surface region.

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