PHOTOELECTRON SPIN POLARIZATION AND FERROMAGNETISM OF CRYSTALLINE AND AMORPHOUS NICKEL

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(Received 30 March 1970)

We observe electron spin polarization in photoemission from nickel, undetected in earlier experimental investigations. The saturation electron spin polarization for films prepared on hot substrates is higher than for films on cold substrates and the preferential direction of the magnetic moment is parallel to the magnetization even for electrons excited from energy states near the Fermi level.

The polarization of electrons emitted from ferromagnets is a striking illustration of the fact, well known from the experiment of Einstein and de Haas, that ferromagnetism is generated by spontaneous spin polarization of the electrons. This was demonstrated for field- and photo-emitted electrons from Gd,[1,2] and for photoemitted electrons from Dy[3] and the ferromagnetic insulators EuO, EuS, and EuSe,[4] but there remained the anomaly that the electrons emitted from the 3*d* ferromagnets Fe, Ni, and Co appeared not to be polarized.[5-7] We believe that this problem is now resolved and, as described below, we find that with pure material and with clean conditions Ni exhibits the largest electron spin polarization (ESP) of all the ferromagnetic metals so far investigated.

We examine first why earlier experiments did not detect this. The sample must have a macroscopic magnetic moment near the saturation value. This was achieved by applying an external magnetic field or by relying on the remanence. In both cases, stray magnetic fields $\vec{B}_s$ are generated outside the sample. If $\vec{E}$ is perpendicular to $\vec{B}_s$, where $\vec{E}$ is the electric field applied for acceleration, the photoelectrons will drift away in the direction $\vec{E}\times\vec{B}_s$; if $\vec{E}$ is parallel to $\vec{B}_s$, the electrons follow $\vec{E}$ and $\vec{B}_s$ can have some focusing effect. We have $\vec{E}$ essentially parallel to $\vec{B}_s$,[2,4] whereas $\vec{E}$ is perpendicular to $\vec{B}_s$ in Refs. 5 to 7. In the latter case it is difficult to see where the electrons that enter the detection system come from. Possibly from points for which $\vec{E}$ is parallel to $\vec{B}_s$; that is, from microscopic protrusions or other irregularities of the surface. The vacuum conditions are not very critical with Ni, but the samples cannot be exposed to pressure above 10^{-7} Torr. A further difference, mentioned in Ref. 2, is that our cylindrical condenser does not transmit photoelectrons from the first acceleration stages.

Experimental. – The Ni films were prepared from 99.999% pure metal and evaporated, by electron bombardment, onto a stainless-steel substrate kept at 4.2, 78, or 380°K. Mass-spectrometric analysis of a film evaporated under similar conditions onto a gold foil of known purity indicates that no contaminants are present.

In Fig. 1 we show the ESP measured at 4.2°K for a film prepared at (a) 383°K and (b) 4.2°K. During film preparation, the pressure was $\sim 10^{-8}$ Torr and within a few hours reached its ultimate value of 3×10^{-10}. A respective saturation value of $P_s = 15.15\,\%$ and $P_s = 9.44\,\%$ was attained for (a) and (b). Measurements on eleven different films showed similar results. The films prepared on a cold substrate showed considerably lower ESP when the starting material was not properly outgassed or was not pure (as in the case of Ref. 4, where Ni powder was used). Under clean conditions there was no difference between the results for films prepared on substrates at temperatures of 4.2 and 78°K. The

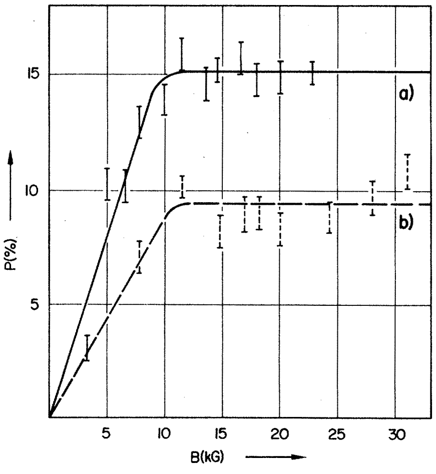

FIG. 1. Dependence of photo-ESP for Ni films on the magnetic field strength, measured at liquid-He temperature. (a) Film prepared on substrate at 383°K; (b) film prepared on substrate at 4.2°K. The preferential direction of the magnetic moment of the electrons is parallel to the magnetization. The full spectrum of a Hg-Xe high pressure arc was used.

films could be kept at room temperature in vacuo ~10^{-9} Torr for several days without change of ESP.

From the results tabulated below for a Ni film prepared at 380°K, we see that the ESP appears not to depend significantly on the sample temperature during measurement, up to room temperature.

Temp. T (°K)	Photo-ESP P(%)
4.2	15.84 ± 0.7
78	16.37 ± 0.5
300	15.2 ± 1

Discussion.—It has been suggested recently by DeWames and Vredevoe[8] that the measured ESP is determined not only by the spin-polarized band structure P_0, but also by effects of inelastic electron-magnon scattering. These effects occur in a path of length s between the point of excitation and the surface. If this point is at a depth x, on the average $s = cx$, where c is a constant >1. Starting from $dn_\uparrow = \sigma^+ n_\downarrow v dt - \sigma^- n_\uparrow v dt$ and $n_\uparrow + n_\downarrow = \text{const}$, we obtain

$$P = P_0(a + 1/x_s)/(c\sigma + a + 1/x_s) + c\sigma\eta/(c\sigma + a + 1/x_s)\cdots, \qquad (1)$$

where a is the optical absorption coefficient, x_s the escape depth, v the velocity, $n_\uparrow$ and $n_\downarrow$ the densities of the two kinds of electron, and dt a time interval. $\eta = (\sigma^+ - \sigma^-)/(\sigma^+ + \sigma^-)$ and $\sigma = \sigma^+ + \sigma^-$, where σ^+ and σ^- are the inverse mean free paths for magnon creation and annihilation, respectively. We have assumed that in magnon or phonon scattering the energy loss is so small that v does not appreciably change, and that in electron-electron scattering the energy loss is sufficient to prevent evasion of the surface-barrier potentials. For metals $a \ll 1/x_s$, $c \approx 1$, and $\sigma \ll 1/x_s$, and at $T = 0$, $\sigma^- = 0$. It follows that $P = P_0 + \sigma^+ x_s$. $\sigma^+(T = 0) < 500$ Å^{-1} for Ni after DeWames and Vredevoe,[8] and even 4 times smaller according to Baltensperger.[9] With an estimated $x_s = 30$ Å (see Eastman and Krolikowsky),[10] we obtain $\sigma^+ x_s < 6\,\%$. We therefore believe that the effects of inelastic electron-magnon scattering can be neglected. This result is confirmed by the observation that the ESP maintains a high value up to room temperature. If effects of electron-magnon scattering predominated in the manner suggested in Ref. 8 and in DeWames and Vredevoe,[11] ESP should, in our temperature range with essentially constant saturation magnetization, decrease with the Debye-Waller factor.

Films of type (a) in Fig. 1 had work functions between 4.7 and 5 eV. With our light source we can excite electrons within 0.8 eV from the Fermi level E_F. In the conduction band the average ESP is $P_m \sim 5\,\%$. The observed ESP of 15 % indicates that magnetic states with ESP higher than P_m must lie very near to E_F. In this respect, our result is consistent with the general shape of the density-of-states curve detected experimentally in recent photoemission studies,[10] and calculated, for example, by Hodges and Ehrenreich[12] and by Connolly[13]: The density of states has a sharp maximum near E_F that arises from states with $3d$ character. According to Stoner and Wohlfarth ferromagnetism in Ni is explained by assuming pure spin states and shifting the density of states for both spins relative to each other, as shown in Fig. 2. In this model, a reversed ESP arises near E_F, and the ESP varies strongly with the optical excitation depth ϵ. ϵ was reduced from an average of ~0.8 eV to about 0.4 eV by means of a filter, and we found not even a decrease of the ESP within ±2%. This appears to conflict with the Stoner-Wohlfarth model.

The ESP for films of type (b) in Fig. 1 cannot be explained by contamination nor by the reduced density that might exist in a film evaporated on cold substrate because the ESP is independent of intensity. With our electron optics, we can also exclude effects due to alterations in surface roughness. This is confirmed by the following observation: Gd films prepared on hot and cold substrates yielded $P = (5.70 \pm 0.21)\%$ and $P = (5.27 \pm 0.15)\%$. We must suppose that with Gd the surface roughness also changes with preparation conditions, but that one does not obtain films of type (b) with this material under our conditions; therefore the essentially constant ESP shows

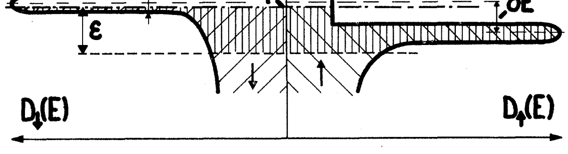

FIG. 2. Essential features of the density of states for Ni near the Fermi level E_F, and the splitting into the two spin sub-bands following the Stoner-Wohlfarth model. ϵ is the energy depth of optical excitation, δE the exchange shift of the order of 0.35 eV, and $B \gtrsim 0.2$ eV.

that there is no major influence of surface conditions. The results for Ni films can most reasonably be interpreted by assuming that the microstructures of the two types of film are different. Strict periodicity is not a requirement for ferromagnetism; so we can assume that in films of type (b) the exchange energy δE and possibly also the density of states $D(E)$ is altered due to a more random distribution of the atoms. An exact definition of the concept amorphous has not been given for pure ferromagnets to date. It is usually related to x-ray and electron diffraction patterns and to electrical conductivity measurements.[14] Since the exchange interactions are short-range interactions, ferromagnetism is a sharp tool for detecting and defining the amorphous state.

Tamura and Endo[15] used x-ray diffraction and magnetic measurements to detect the amorphous state of Ni. They measured the number of Bohr magnetons n_B per atom for Ni films of types (a) and (b) and found that $n_\mathrm{B}(\mathrm{b})/n_\mathrm{B}(\mathrm{a}) = 0.60$. If P_m is the average ESP of the conduction band, we obtain $P_m = n_\mathrm{B}/n$, with n the number of electrons per atom. From Fig. 1 we have $P_\epsilon(\mathrm{b})/P_\epsilon(\mathrm{a}) = 0.62$. The agreement of the two different kinds of measurements indicates that the change of the spin-polarized band structure, as the Ni transforms from the crystalline to the amorphous state, is to be observed in our narrow energy range, $\epsilon \simeq 0.8$ eV, at the Fermi surface.

The authors are indebted to many of their colleagues for helpful comments and suggestions, to Dr. J. T. Muheim for mass-spectrometric analysis, and to Dr. D. J. Fabian for assistance with the manuscript. Financial support was received from the Schweizerischer Nationalfonds zur Förderung der wissenschaftlichen Forschung and the Verein zur Förderung der Festkörperphysik an der ETH.

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