Paramagnetic Sheet at the Surface of the Heisenberg Ferromagnet EuO

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Measurements of the spin polarization of photoelectrons from (100) surfaces of "pure" and doped EuO monocrystals reveal a paramagnetic surface sheet existing at temperatures far below the Curie temperature of the bulk. The extra electrons that are introduced on doping produce an increase of the molecular field in the surface. $Eu_{1-x}La_xO$ crystals exhibit the highest degree of spin polarization (~80%) observed in a photoemission experiment so far.

Most theoretical papers dealing with magnetism in the surface have confined their attention to materials described by the Heisenberg model,[1] whereas the experimental work has been done with thin films of metallic itinerant ferromagnets,[2] for which molecular field theory is hardly applicable. It is the purpose of this Letter to report on a new experiment that allows one to measure the magnetization in a region very near the surface and to describe first results obtained with pure and doped single crystals of the model Heisenberg ferromagnet EuO.

EuO has a Curie temperature T_C of 68°K and crystallizes in the NaCl structure. The localized spin-only moment with $S=\frac{7}{2}$ arises from the half-filled $4f$ shell of Eu^{++}. Doping with trivalent cations introduces extra electrons, giving rise to several unique changes of magnetic, electric, optical, and photoelectric properties.[3]

The experiment employs photoemission of electrons including a measurement of the spin polarization of the photoelectrons. The degree of spin polarization along a direction parallel to the magnetization $\vec{M}$ of the crystal is given by $P=\text{const} \times M(H,T)/M_0$ for pure spin-state ferromagnets, where $M(H,T)$ is the magnetization at a magnetic field H and a temperature T and M_0 is the saturation magnetization. The escape depth of the photoelectrons determines the thickness of the region for which the relative magnetization is measured.

Cubic monocrystals with linear dimensions around 5 mm were pressed firmly with spring clamps against flat surfaces of stainless-steel holders. 22 such stainless-steel holders with crystals were stored in ultrahigh vacuum on a wheel. For a measurement, the wheel is turned so that the holder of the crystal under investigation is positioned underneath gripper number one which grips the holder and moves it to the anvil and knife. A slice of $\frac{1}{2}$–$\frac{1}{4}$ mm is cleaved from the crystal to obtain a fresh (100) surface. The holder with the crystal is put back into the wheel. The wheel is then turned so that gripper number two, which is cooled to liquid-He temperature, can grip the holder and pull it up into the 30-mm bore of a superconducting coil. Here, the light impinges on the freshly cleaved crystal surface and electrons are photoemitted. The photoelectrons are extracted from the magnetic field region, and an electron beam is formed. P is measured as described previously[4] by Mott scattering.[5] The pressure in the apparatus was between 2×10^{-10} and 4×10^{-10} Torr, rising to higher values only for very short periods of time during the movements and the cleaving of the crystal. The temperature T of a crystal during the measurement is determined by the thermal contact between the crystal and the holder and between the holder and the gripper and it can be somewhat higher than the temperature of gripper number two, $T \geq 4.2$°K. P did not depend on the light intensity, which indicates that the impinging light does not heat up the crystals.

The dependence of P on photon energy $h\nu$ at constant H is shown in Fig. 1 for two representative crystals. Nonintentionally doped EuO shows a low P for $h\nu$ near the photothreshold Φ which was found to be ~2 eV. The threshold value of P was different for each crystal surface and must therefore arise from uncontrolled impurity states. For $h\nu \geq 4$ eV, P becomes high, indicating that emission of electrons from the intrinsic $4f^7$ states is now dominant. The doped EuO sample shows a higher P for $h\nu$ near Φ, which was found to be ~1.7 eV. P reaches a flat maximum before it levels off to the constant value determined by the spin polarization of the $4f^7$ states. Uncontrolled impurities with low P must be present also in the doped sample, and P at low $h\nu$ is a su-

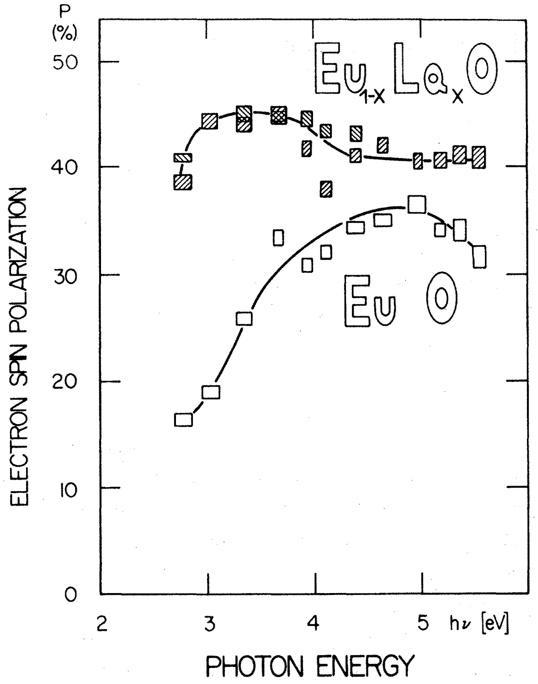

FIG. 1. The dependence of photoelectron spin polarization P on photon energy $h\nu$ at constant magnetic field strength $H = 6$ kG, for "pure" EuO and $E_{1-x}\mathrm{La}_x\mathrm{O}$, where $x \sim 1$ at.%. The horizontal width of the points gives the resolution of the monochromator, the vertical width the statistical uncertainty in the measurement of P. The lower points in the spectrum of $\mathrm{Eu}_{1-x}\mathrm{La}_x\mathrm{O}$ are taken 3 h after cleaving, the higher points 5 h after cleaving.

perposition of the polarization of the impurities and the extra electrons introduced on doping. It is surprising that P of the extra electrons alone is higher than P of the magnetic $4f$ electrons at $H=6$ kG.

The two spectra of doped EuO were taken 3 and 5 h after cleaving. The close similarity, with the exception of one unexplained point at $h\nu = 4.1$ eV, shows that the surface contamination has no major influence on the results with the present vacuum conditions. Figure 2 shows the dependence of P on the applied field H_C for the same crystal surfaces as Fig. 1. The energy distribution of the light was chosen such that the dominant contribution to the photocurrent came from $4f$ electrons. The main features of the photoelectric magnetization curves (PMC's) are the following: (i) There is no magnetic saturation; (ii) a kink occurs at 8–10 kG; (iii) doped EuO shows higher values for P compared to "pure" EuO. The PMC of Ni[6] exhibiting magnetic saturation is also shown for comparison.

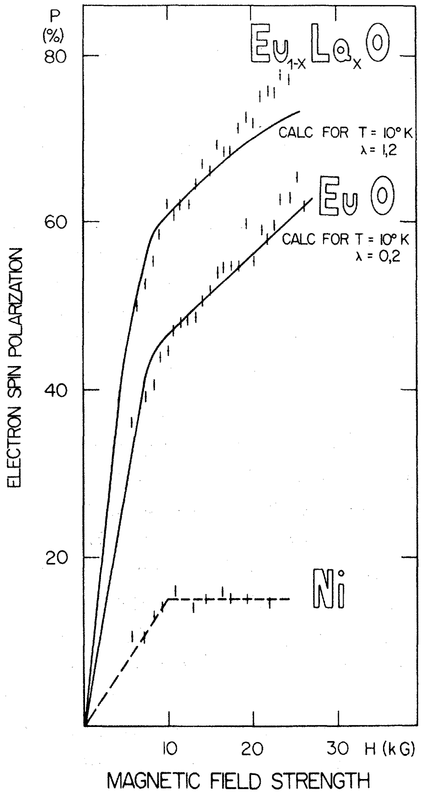

FIG. 2. The dependence of P on the magnetic field strength H for the same crystal surfaces as Fig. 1, and, for comparison, for Ni, after Ref. 6. The full lines are calculated assuming $T = 10$°K, and the best fit was obtained with the molecular field constants λ given in the graph.

Let us assume that a paramagnetic sheet is present at the surface. The magnetic field H_{eff} acting on a $4f$ spin in the surface is given by

$$H_{eff} = H_C + M_B - M_S + \lambda M_S, \qquad (1)$$

where H_C is the field generated by the coil, M_B is generated by the bulk magnetization, $-M_s$ is the demagnetizing field generated by the magnetization M_s of the sheet, and λM_s is the molecular field in the sheet.

First, we examine the kink in the PMC: It occurs when the bulk saturates. The demagnetizing factor of the crystals is $\sim\frac{1}{3}$ and $M_0 = 24$ kG, hence the bulk saturates at $\sim\frac{1}{3} \times 24$ kG.

It is possible to calculate PMC's from Brillouin functions for $S = \frac{7}{2}$ if one assumes that P is conserved in the process of photoemission and that the saturation magnetization of the sheet is equal to the bulk value. The only two parameters are T and the molecular field constant λ. For each choice of T there exists a λ for which the fit between calculated and measured PMC is best. The change $\Delta\lambda$ of the molecular field constant on doping proves to be independent of the assumed value of T in the range $5 \leqslant T \leqslant 15$°K. We obtained $\Delta\lambda = 1.0$ for the present crystal surfaces. The extra electrons introduced on doping are known to have a certain mobility,[3] and this is why they are an effective link between bulk and surface. That the mobile electrons produce the extra molecular field in the surface is directly seen from their high P in Fig. 1.

The paramagnetic sheet is an effective barrier for photoelectrons from inside the bulk as long as M_s is low, because it is known that spin-disorder scattering is very effective in EuO. When M_s builds up in an external magnetic field, the sheet becomes transparent. This corroborates the deviation of calculated and measured PMC's at higher H shown in Fig. 2. We believe that the thickness of the sheet observed at low M_s can be as small as a few atomic layers. The existence of the paramagnetic sheet at an estimated $T \sim 10°$ K, far below T_C, is surprising. Apart from the reasons already discussed in the literature,[1] an increase of the lattice constant in the surface may be important, which is known to reduce the ferromagnetic part of the exchange in the europium chalcogenides.[7] Further experiments should include the measurement of the T dependences, which will allow a complete molecular field analysis.

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Multiple-Reaction Correction to the Capture Cross Section*

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The multiple-reaction correction of Halpern, Drake, *et al.* to neutron radiative cross-section measurements is shown to be important also below 14 MeV, with the consequence that previously measured uncorrected (or unrestricted to first-chance capture only) (n,γ) cross sections above the first level are in error, being from about 1% to a factor of more than 10 too high.

Some time ago Drake, Whetstone, and Halpern[1] found their 14-MeV (p,γ) photon-spectrum–derived cross sections to be below 1 mb in contrast to previously measured (n,γ) cross sections which ran from 1 mb near magic to as much as 17 mb away from magic above $A = 40$. Cvelbar, Hudoklin, and Potokar[2] reported that their 14-MeV (n,γ) measurements, made by integrating the γ spectrum, up to $A = 138$, gave cross sections from 0.3 to 1.2 mb, again factors of 1.5 to 12 lower than expected. Drake, Bergqvist, and McDaniels[3] continued the (n,γ) cross-section measurements to higher A by measuring direct transitions to bound states with a $^{208}\mathrm{Pb}(n,\gamma)$ standard and found 14-MeV cross sections to be nearly constant at 0.9 to 1 mb, again very low.