2 and 5.
[10]R. Kelly and N. Q. Lam, Radiat. Eff. 10, 247 (1971).
[11]S. L. Ruby, B. J. Zabransky, and J. G. Stevens, J. Chem. Phys. 54, 4559 (1971).

Electron Spin Polarization in Field Emission from EuS-Coated Tungsten Tips

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Field-emitted electrons from EuS-coated W tips cooled to below 21 K show spin polarization of up to $(89 \pm 7)\%$. The preferential direction of the magnetic moments of the electrons is parallel to the magnetization of the emitter. The high spin polarization is explained in terms of emission from spin-polarized $4f^7$ states of ferromagnetic EuS or in terms of emission assisted by interaction with these states.

An ensemble of electrons is said to have electron spin polarization (ESP) when they show a preferential spin direction.[1] ESP is described by the vector $\vec{P}$ in the preferential direction, whose magnitude is $P = (N_+ - N_-)/(N_+ + N_-)$, where N_+ and N_- are the numbers of electrons with expectation value of the spin parallel or antiparallel to the preferential direction. In electron emission, ESP is to be expected when states with a certain spin direction have a preference in the emitter and the emission ensues from these states or by interaction with them. As with the photoelectric effect,[2] in field emission (FE) ESP affords information on the emission process or on the electronic states in the emitter. Proposals,[1,3,4] calculations,[4,5] and experiments[1,6,7] dealing with FE of spin-polarized electrons always utilize the emission from spin-polarized states in the region of the Fermi edge. Experiments hitherto only conducted on ferromagnetic metals show P below 15%.[6,7]

Tunneling mechanisms involving interactions with vacant or occupied spin-preferred states in a barrier have so far not been discussed with respect to polarization effects. Here we report the first results of a FE experiment utilizing a W tip coated with a thin film of insulating, ferromagnetic EuS.

Figure 1 shows the experimental setup. The W tip is mounted on a He-cooled finger in the center of a magnetic field coil. The cold finger works on the contact fluid principle[8] with liquid H_2 permitting attainment of temperatures between 14 and 21 K. The magnetic field $\vec{H}$ defines the preferential spin direction, i.e., $\vec{P}$, antiparallel to the resulting magnetization in the emitter. The coil is cooled with liquid N_2 and pulsed (pulse length, 250 μsec; repetition rate, 0.1–0.5 Hz; $H_{max} = 20$ kOe). The tip and the coil can each be aligned. The W tip is coated *in situ* with EuS from a removable oven heated by electron bombardment. The FE patterns are projected by the extraction optics on a screen, which can be viewed in the mirror. For detection of the ESP the Mott scattering technique[9] is used. The principle of the arrangement is similar to that applied in photoemission experiments.[2] However, we use a four-detector arrangement.[9,10] Two detectors set at scattering angles of 120° are sensitive to ESP and the other two, set at 45°, serve

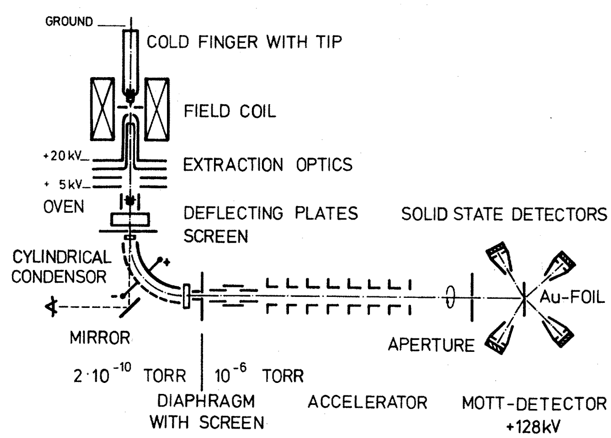

FIG. 1. Experimental setup.

to control the beam alignment. The entire Mott scattering arrangement can be rotated about the beam axis to eliminate instrumental asymmetries.

Film preparation.—The first step requires a W tip with a clean W FE pattern. Then the tip is coated with 500–2000 Å of EuS at a rate of 1–3 Å/sec, while the tip is either cooled or slightly heated. The residual gas pressure during evaporation is kept below 10^{-8} Torr. The FE pattern of a freshly coated tip does not show any ordering.[11] Annealing at about 800°C causes the EuS to migrate towards the tip shank. The film thickness thereby decreases and the EuS assumes an epitaxial structure. At this time *emission type I occurs*. In the FE pattern individual regions of high emission primarily in the ⟨112⟩ directions of W are observed. The emission potential[12]—always referred to the vacuum level of EuS—is about -1.8 eV at 14 K. Further annealing produces *emission type II,* with a likewise twofold symmetric but otherwise completely different FE pattern. About the [110] direction there is a rosette from which four beam-shaped punctiform arrangements issue. The emission potentials is -4.1 ± 0.3 eV. Further annealing at temperatures of up to 1200°C results in *emission type III.* As in type I, the FE here is in the ⟨112⟩ directions[11] and the emission potential is -3.3 ± 0.2 eV.

ESP measurements.—In emission type II an ESP of up to $(89 \pm 7)\%$ was found. Figure 2 shows our measurements on this type. The scale error is due to the uncertainty in calibration.[13] The positive sign of $P_\parallel$ and the ratio $P_\parallel/P_\perp$ proves $\vec{P}$ to be nearly antiparallel to $\vec{H}$. In the investigated temperature and magnetic field ranges of 14–21 K and 2.3–18.5 kOe, respectively, $\vec{P}$ varies weakly, although the Curie point of well-annealed EuS films is about $T_C' = 16.6$ K and the saturation magnetization is about 14 kG.[14]

Few measurements were made on emission type I. At 14 K the ESP is about 50% and decreases with increasing temperature to about 20% at 21 K. What is important is the temperature dependence in the region of the normal Curie point T_C of EuS. In emission type III, as well as in the case of a clean W tip,[15] no ESP could be detected (detection limit 5%).

The results are interpreted by means of the model shown in Fig. 3. For the EuS we use the electronic structure found by optical measurements.[2,16–19] The characteristic features of the EuS are the $4f^7$ levels lying in the band gap and the ferromagnetically ordered spins below T_C

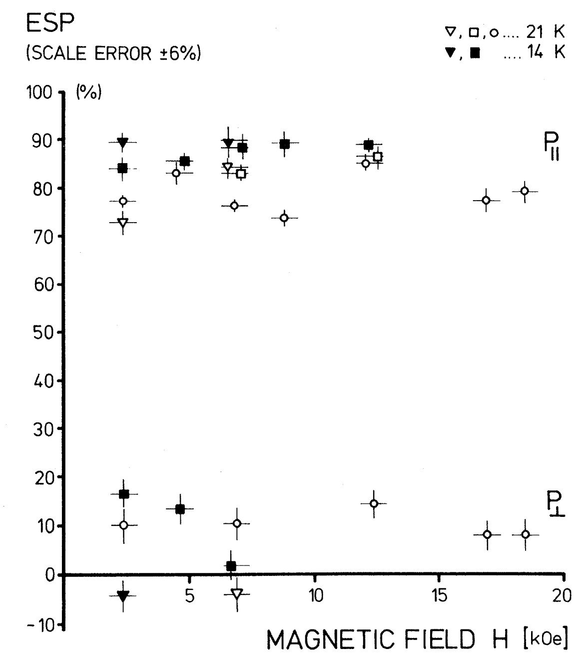

FIG. 2. Electron spin polarization of field-emitted electrons from EuS-coated W tips as a function of the magnetic field for two temperatures around the normal T_C of EuS (emission from or after interaction with $4f^7$ states). $P_\parallel$ is the component of the polarization vector $\vec{P}$ parallel to the external magnetic field $\vec{H}$. $P_\perp$ is one component of $\vec{P}$ perpendicular to $\vec{H}$. The lower points (open circles) are measured under less favorable vacuum conditions.

$= 16.5$ K. As a result of interaction with the $4f^7$ states the conduction bands are magnetically split. The lower edges of these bands have d character, and so as a result of spin-orbit coupling they do not contain pure spin states only.[20,17] The $4f^7$ states are assumed to be localized. Their energy levels are located to correspond to transitions from the $4f^7\ {}^8S_{7/2}$ state to a state at the vacuum level, leaving behind a ${}^7F_J$ multiplet state ($J = 0, 1, \ldots, 6$) of the excited $4f^6$ configuration.[16] If the lowest multiplet state ${}^7F_0$ is involved, the transition energy is equal to $\chi = \sim 4$ eV.[16,2] Since χ is smaller than $\varphi_W = 4.5$ eV, and the vacuum level is continuous at the W-EuS interface, $4f^7 \to 4f^6$ transitions are possible in the boundary layer. The resulting excess charges cause a direct matching of the Fermi levels of W and EuS by a potential drop in the boundary layer[21] ($\varphi_{EuS} = 3.3 \pm 0.3$ eV).[16] The emission field F of typically 2.5×10^7 V/cm is weakened in the EuS film to $F' = F/\epsilon_{stat}$ ($\epsilon_{stat} = 10.2$).[22] F' lowers the potential of the electronic states in the EuS linearly with the distance x from W. This model

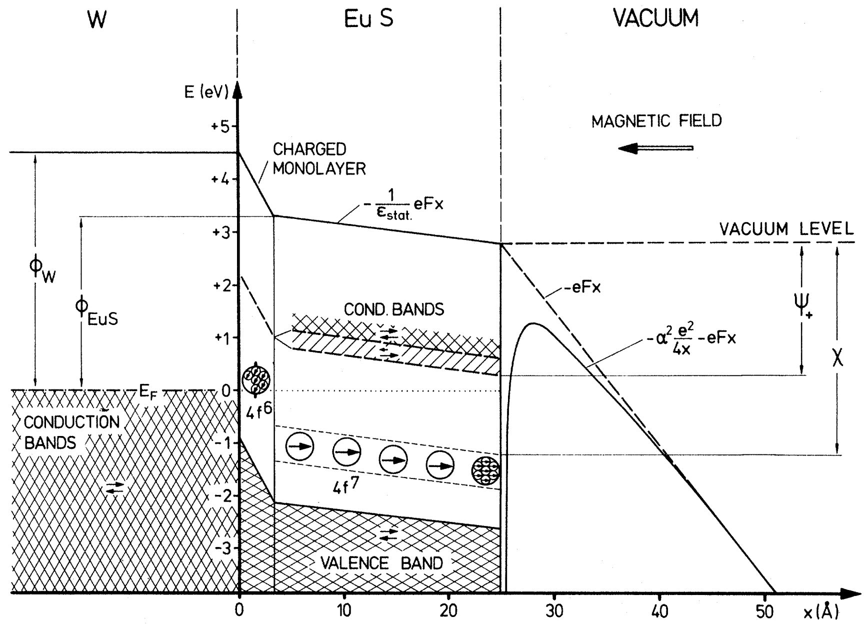

FIG. 3. Field-emission model for EuS on W for a typical field $F = 2.5\times10^7$ V/cm. In the image force potential α is $(\epsilon_{opt}-1)/(\epsilon_{opt}+1)$ with $\epsilon_{opt}=5.2$ (Ref. 22). ψ_+ is the electron affinity of the lower split conduction band, χ the photothreshold, and φ_W and φ_{EuS} the work functions of W and EuS.

gives the following essential FE processes:

(1) Tunneling through conduction-band states[23]: With a sufficiently thick film, EuS conduction-band states are lowered below the Fermi level E_F of W. Electrons from E_F can tunnel into these states and hence into the vacuum. The emission potential should be higher than $-\psi_+ = -2.5$ eV. Because of the d character of the used states the ESP of the emitted electrons will be well below 100%.

(2) Tunneling from $4f^7$ states or tunneling assisted by interaction with $4f^7$ states: In these cases the emission potential should be about $-\chi$.[23,24] With ferromagnetic ordering an ESP of close to 100% is to be expected. In thick films process (2) is at a marked disadvantage compared with (1); it will be only observed if the film is too thin for (1) (Fig. 3).

(3) Direct tunneling of W electrons from the region of E_F[23]: As with (2) exclusion of (1) is necessary for the existence of this process (Fig. 3). The emission potential should be -3.3 eV, corresponding to φ_W lowered to φ_{EuS} by the charged layer. If the $4f$ spins are ordered the conduction bands are spin split, and consequently the height of the EuS barrier is spin dependent.[25,26] The tunneling electrons will be polarized to a similar degree as in (1). No ESP is to be expected for a very thin film comprising only the charged boundary layer with its nonmagnetic $4f^6$ states. Process (3) will always occur in competition with (2). With thick films (2) is preferred and with very thin films it is (3) that prevails.[24]

We relate our emission types I, II, and III to the FE processes (1), (2), and (3), respectively. The succession of the three emission types during the annealing process, the emission potentials, and the observed ESP agree with the proposed model. The slight magnetic field dependence in the measurements (Fig. 2) is explained by the fact that the EuS has moved down the tip shank and as a cylindrical film is easily magnetized axially. Therefore, the area near the pole of the tip experiences nearly total magnetization in or against the direction of the tip even for small external fields. The existence of $P_\perp$ (Fig. 2) is probably caused by asymmetries in the film, by misalignments of the axis of tip and coil, and by radial components of the magnetic field.[27]

The saturation behavior of $P(T,H)$ at emission type II (Fig. 2) indicates a large increase of T_c. This may be due to an additional exchange interaction via W electron states and/or lattice compression at the EuS-W interface.

We believe that the experiment described leads the way to further interesting FE effects and to a source of high brightness for highly spin-polarized electron beams. In static operation a current of 10^{-6} Å seems possible.

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[1]For a detailed review see P. S. Farago, Advan. Electron Phys. 21, 1 (1966).

[2]G. Busch, M. Campagna, and H. C. Siegmann, J. Appl. Phys. 41, 1044 (1970), and 42, 1781 (1971).

[3]N. Müller, H. C. Siegmann, and G. Obermair, Phys. Lett. 24A, 733 (1967).

[4]G. Obermair, Z. Phys. 217, 91 (1968).

[5]B. A. Politzer and P. H. Cutler, Phys. Rev. Lett. 28, 1330 (1972).

[6]M. Hofmann, G. Regenfus, O. Schärpf, and P. J. Kennedy, Phys. Lett. 25A, 270 (1967); G. Chrobok, M. Hofmann, and G. Regenfus, Phys. Lett. 26A, 551 (1968).

[7]W. Gleich, G. Regenfus, and R. Sizmann, Phys. Rev. Lett. 27, 1066 (1971).

[8]G. Klipping and R. Vanselow, Z. Phys. Chem. (Frankfurt am Main) 53, 195 (1967).

[9]Review in W. Eckstein, Garching Internal Report No. IPP 7/1, 1970 (unpublished); N. Müller, Dipolmarbeit Universität München, 1966 (unpublished).

[10]J. van Klinken, Nucl. Phys. 75, 161 (1966)

[11]W. Eckstein, K. F. Georg, W. Heiland, J. Kirschner, and N. Müller, Z. Naturforsch. 25a, 1981 (1970).

[12]The emission potential is the highest potential from which electrons are tunneling to the vacuum. It is determined by comparing the slope of Fowler-Nordheim plots for the clean W tip and for the covered tip. The Nordheim function $v(y)$ is taken into account [see R. H. Good, Jr., and E. W. Müller, in *Handbuch der Physik*, edited by S. Flügge (Springer, Berlin, 1968), Vol. XXI, p. 186]. For type I the method may be poor because of geometric effects.

[13]For calibration the ESP-dependent scattering asymmetry $A(d)$ of gold foils of thickness $d = 50$, 100, and 150 $\mu g/cm^2$ was extrapolated to $d = 0$. For $A(0)$ the value $S = 0.41$ was used [see S. Lin, Phys. Rev. 133, A965 (1964), and Ref. 9].

[14]E. Bayer and W. Zinn, Z. Angew. Phys. 32, 83 (1971).

[15]In contrast, G. Regenfus *et al.* [in Proceedings of the Eighteenth Field Emission Symposium, Eindhoven, The Netherlands, 1971 (unpublished)] reported an ESP of up to 18% in FE from W. Owing to spin rotation discussed in Ref. 7, there is a small chance that an actual ESP $\neq 0$ was obscured by a 5% ripple in our our magnetic pulse plateau. But no rotation of $\vec{P}$ was found in any of our EuS measurements.

[16]D. E. Eastman, F. Holtzberg, and S. Methfessel, Phys. Rev. Lett. 23, 226 (1969).

[17]G. Güntherodt, P. Wachter, and D. M. Imboden, Phys. Kondens. Mater. 12, 292 (1971).

[18]D. E. Eastman and M. Kuznietz, Phys. Rev. Lett. 26, 846 (1971).

[19]Impurity states are ignored on the assumption that their influence on FE is negligible owing to their relatively low density.

[20]J. O. Dimmock, J. Hanus, and J. Feinleib, J. Appl. Phys. 41, 1088 (1970).

[21]The transition $4f^7 \rightarrow 4f^6$ in the boundary layer possibly accounts for the inconsistency in the height of the tunneling barriers of a Au-EuSe contact as compared to a Al-EuSe contact found in L. Esaki, P. J. Stiles, and S. von Molnar, Phys. Rev. Lett. 19, 852 (1967).

[22]P. Wachter, Phys. Kondens. Mater. 8, 80 (1968).

[23]H. Neumann, Ann. Phys. (Leipzig) 19, 270 (1967).

[24]Possible interactions are resonance scattering and exchange scattering (with magnon creation). In both interactions electrons having nearly the same potentials as the $4f^7$ states are favored.

[25]The influence of conduction-band splitting on tunneling is to be seen in Esaki, Stiles, and von Molnar, Ref. 21, and W. A. Thompson, F. Holtzberg, T. R. McGuire, and G. Petrich, in *Magnetism and Magnetic Materials –1971*, AIP Conference Proceedings No. 5 (American Institute of Physics, New York, 1972), Pt. 2, p. 827.

[26]Esaki, Stitles, and von Molnar, Ref. 21.

[27]W. Eckstein and N. Müller, to be published.