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Spin polarization of field-emitted electrons from Fe, Co, Ni, and rare-earth metals

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The spin polarization of field-emitted electrons is investigated at 10-K emitter temperature and magnetic field strengths up to 6 T. Field emitters are electrolytically etched Fe, Co, and Ni tips of polycrystalline material and W tips coated by vapor deposition with Fe, Co, Ni, and rare earths. Uncoated W tips show no polarization. The maximum polarization obtained so far is + 80% and + 47% for Fe and Co, respectively (the "+" sign indicates predominant majority spins). Often the polarization is found to vary beyond control in magnitude and occasionally even in sign, depending upon, e.g., surface preparation, emission spot, tip crystallography, adsorption. Clean Ni tips show $(-3.5 \pm 1.5)\%$ in the vicinity of $\langle 111 \rangle$ and $(0 \pm 1.5)\%$ in the vicinity of $\langle 100 \rangle$. In both cases polarization increases with time up to + 15% by hydrogen adsorption. All ferromagnetic rare-earth metals give rather high electron spin polarization, clean Gd up to + 53%, EuO and EuS show up to + 90%. The temperature dependence of the polarization follows approximately the saturation magnetization, as checked with Ni, Gd, EuO.

I. INTRODUCTION

In 1930 Fues and Hellmann[1] proposed to extract polarized electrons by field emission (FE), photoemission, or secondary electron emission (see *note added in proof*) from ferromagnets. The early FE experiments with iron tips by von Issendorff and Fleischmann[2] and Pimbley and Mueller[3] remained unsuccessful. Polarized electrons were observed, however, in 1967 by Hofmann, Regenfus, Schärpf, and Kennedy[4] in FE from ferromagnetic gadolinium. Investigations with iron and nickel tips were again taken up and brought electron-spin polarization (ESP) values of 6% and 13% at maximum, respectively,[5] disregarding the (incorrect) assumption of a rotation of the polarization vector with magnetic field strength.

Almost complete polarization (89% at maximum) was found by Müller, Eckstein, Heiland, and Zinn[6] in FE from EuS, a ferromagnetic semiconductor, vapor deposited, and annealed on tungsten tips. Recently, Müller[7] and Campagna, Utsumi, and Buchanan[8] reported ESP measurements in particular directions of emission from single-crystalline Ni tips. Positive and negative values of about 3% were observed.

The history of attempts to produce polarized electrons by photoemission took an analogous course. The early experiments[9-11] were unsuccessful. ESP was found, however, in 1969 by Busch, Campagna, Cotti, and Siegmann in photoemission from thin vapor-deposited films of polycrystalline gadolinium[12] and later on from polycrystalline films of nickel[13] and iron and cobalt.[14,15] With iron, values as high as + 54% were found,[15] with pure EuO, values as high as 62% were found.[16] The experimental results on Ni and Co show predominantly majority spins, which is considered to be inconsistent with the Stoner-Wohlfarth band model.[17-19] Majority spins were also observed for Ni and Co in tunneling experiments by Tedrow and Meservey.[20-22] The controversy between experiment and theory is reviewed by Gutzwiller.[23] A recent development about the role of the surface-electron density of states in FE is pointed out by Penn.[24]

Two other novel techniques, ESP measurements via the capture of surface electrons by deuterons reflected at ferromagnetic single crystals (Rau and Sizmann[25-27]) and the application of the magneto-optic Kerr effect (MOKE)[28-31] recently furnished additional results for Fe, Co, Ni. In MOKE, the evaluation by Erskine and Stern[28] favors, in nickel, minority spins of the electrons near the Fermi level. The results derived from capturing surface electrons are more detailed than those of the other techniques, since the outcome varies with the chosen (*hkl*) of the crystallographic plane which reflects the deuteron beam. Predominant minority spins are found in nickel for (110), (100), and (111), whereas majority spins prevail in (120). Majority spins are also observed for Co in $(1\bar{1}00)$ and for Fe in (100).[27]

A probable explanation for the differences in ESP obtained is that the various measuring techniques address different kinds of electrons in the ferromagnets. Such problems are discussed for FE and photoemission in a series of recent papers.[32-35] In most measurements surface electrons are involved. Therefore, the application of band-structure calculations performed for bulk material[36-40] in interpreting such ESP results might not be justified. In several theoretical papers[41-43] the pecularities of the magnetic coupling in the uppermost lattice layers and of surface atom clusters to the bulk has been examined. Antiferromagnetic

coupling of the surface layer or "magnetically dead layers"[44] are discussed (see also Refs. 45–47).

On the other hand, particular care must be taken experimentally in preparing and handling the surfaces of the samples. In the ideal case they should not only be clean, but also atomically smooth and single crystalline of known crystallographic orientation.

In the following we report on measurements of electron-spin polarization of field-emitted electrons obtained with an advanced apparatus, which is described in Sec. II. Detailed investigations are now available for the 3*d* ferromagnets Fe, Co, and Ni, the rare-earth metal Gd, and compounds of Eu (Sec. III). In addition, first measurements are reported of the electron-spin polarization found in field emission with W tips coated with various other rare-earth metals.

Part of the following results were presented elsewhere.[48]

II. EXPERIMENTAL

A general view of the apparatus is seen in Fig. 1(a). The main parts are (i) the emission section with the FE source in the cryostat [a detailed layout of the FE source is displayed in Fig. 1(b)], (ii) the electron extracting system, (iii) the linear accelerator, (iv) an electrostatic steering unit for beam adjustment, (v) the spin precessor (Wien filter), (vi) the Mott scattering chamber, and (vii) the counting and processing electronics.

(i) The FE source is located at the center of a 30-mm-diam bore-hole in a 6-cm-long solenoid; the superconducting coil can produce a magnetic induction up to 7 T. The source head is adjustable for alignment with the solenoid axis. The FE tip is spot welded to a hairpin of 0.2-mm-diam tungsten wire which by dc resistance heating allows the temperature to be set between about 300 and 3000 K, measured with a W-Re thermocouple fixed near the FE tip. To vary the tip temperature between 10 and about 250 K, a small resistance heater, attached to the entire FE head is used. In this range the temperature is measured through the change in capacity of a ceramics multilayer condensor chip fixed to the FE head.

The FE source is surrounded by surfaces cooled with liquid helium. The coverage time of a clean W tip during a field-emission measurement is such that it corresponds to a vacuum better than 3×10^{-10} mbar, as found from the time dependence of the work function; the residual gas is mainly hydrogen.

The emission section accomodates a small evaporator which can be rotated in front of the FE tip, leaving a distance of 5 mm [see Fig. 1(b)]. It is either an oven made of a small Al_2O_3 ceramics tube, heated by electron bombardment, used in the first series of experiments for depositing Fe or it consists of a pin of the material to be evaporated (Fe, Co, Ni) by direct electron bombardment. The electron source is a fixed loop of tungsten wire, see Fig. 1(b); the voltage applied between the electrically heated loop and the pin is usually 500 V, allowing for a power output of about 10 W. A Ta tube which surrounds the pin is kept at ground potential and guarantees that only the very top of the pin is hit by the electrons and evaporated. Thus, the deposited film does not become contaminated with any oven material. Rare earths are evaporated from a tantalum container with which they do not react. The films deposited in

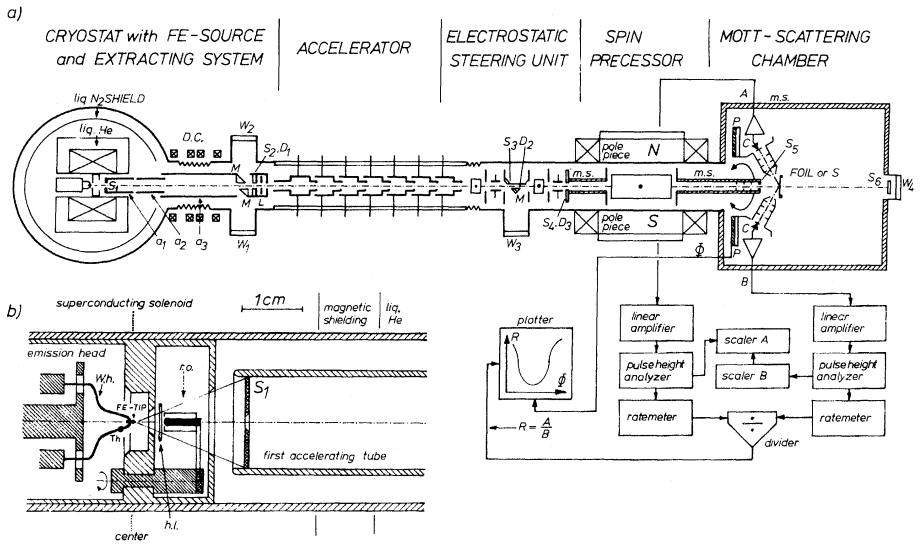

FIG. 1. (a) Layout of apparatus, top view (not to scale). Total length 3 m. (b) Emission system: $S_1 \cdots S_6$, fluorescent screens; $D_1 \cdots D_3$, diaphragms; M, mirror; $W_1 \cdots W_4$, viewing windows; L, electrostatic lens; D. C., two pairs of deflecting saddle coils; $a_1 \cdots a_3$, the first accelerating stages; m. s., magnetic shielding; P, Peltier elements; C, electron counters with preamplifiers; A, B, counting channels; W. h., tungsten heater; Th, W-Re thermoelement; r. o., rotating oven; h. l., resistance heater loop.

such a way at the FE tip usually have thicknesses of more than 100 nm; the evaporation rate is found hard to control. The revolving evaporator can be replaced by a thermal electron source. It is used to anneal and, if possible, to sharpen Fe or Ni tips *in situ* by electron bombardment.

In operation, an adjustable dc potential of up to −6 kV is applied between the tip and the solenoid case to produce the field-emitted electrons. At zero or low magnetic field the FE pattern can be observed on the small 15-mm-diam fluorescent screen S_1, see Fig. 1(b). It is located at a distance of 15 mm in front of the FE tip. The image can be viewed via a mirror through window W_1. The screen S_1 has a central hole, 2 mm in diameter. A magnetic field stronger than 0.1 T focuses the FE beams sufficiently so that the electrons leave the emission section through this hole, entering the next stage.

(ii) The extracting system consists of three successive potential stages which act as electrostatic lenses and raise the electron energy to 15 keV. The electrons produce an image at the fluorescent screen S_2 which can be observed via a mirror through window W_2.

The magnetic field of the long solenoid around the FE tip causes those electrons, which are emitted from various spots at the tip and, therefore, have different angles $\vartheta_{em} > 0$ towards the center axis to move on extended helices. The velocities of the electrons are almost independent of the emission angle ϑ_{em}. Consequently, the electrons with different ϑ_{em} have different focal lengths. If the original FE pattern contains several small emission spots, then at the screen S_2 the same number of luminous circles is produced; their diameters vary with ϑ_{em} and magnetic field strength (usually kept at 1.5 T). All the circles touch at S_2 the foot of the central axis with their local correlation and relative diameters independent of the magnetic field strength.

It is difficult to correlate the FE spots observed on S_1 at low or zero field with the circles observed at S_2 at high magnetic field. Such a correlation can be accomplished if the spots happen to differ distinctly in intensity. However, the FE pattern of a smooth and clean tip consists of electrons with a wide variety of emission angles ϑ_{em}. Then, at S_2 a central luminous patch is produced as the result of a superposition of numerous circles; their envelope is the border line of the patch. The various directions of emission are hardly separable in this case.

Central beams in the original FE pattern remain central at S_2. In the following, mainly such central parts are investigated. In principle, any part of the pattern at S_2 can be chosen for further polarization measurement. This is achieved by two pairs of crossed deflecting saddle coils: they render it possible to shift a particular part of the pattern towards the center hole of the screen whereby allowing it to pass through it and through the diaphragm D_1 behind S_2 thus allowing these electrons to enter the accelerator section.

(iii) The linear accelerator is an eightfold cascade designed in a ceramics-metal technique. The electrons leave this section with 100 keV (the FE source is at −100 kV below ground potential); such a high energy is required for the ESP measurement in Mott scattering.

(iv) The electrostatic steering unit of four deflecting condensors serves for aligning the electron beam in the axis of rotation of the succeeding Mott scattering chamber. The degree of alignment is controlled by means of the fluorescent screens and diaphragms S_3–S_6, observed through windows W_3 and W_4. The geomagnetic field is compensated by an air magnet coil framing the apparatus in total.

(v) The spin precessor is used to turn the electron-spin direction transverse to the beam direction as required for the ESP measurement in Mott scattering. Normally, the electrons are emitted with longitudinal direction of spin polarization. The 90° rotation (or any other angle) is achieved by a Wien filter, where the stationary electric field $\vec{E}$ and magnetic field $\vec{B}$ are mutually orthogonal and, in addition, perpendicular towards the velocity vector $\vec{v}$ of the electrons. The rotation angle θ of the spin of the electron in passing through the Wien filter of length L is given in units of the Système International by $\theta = egBL/(2m^2v/m_0)$ with e, g, m, and m_0 electron charge, g factor, relativistic, and rest mass, respectively.[49] The permalloy tubes on both ends of the filter [see Fig. 1(a)] secure a steep drop of the magnetic field which in the present case has an effective length of 20.3 cm. The separation of the electrostatic field plates widens at both ends to meet the condition $E/B = v = \text{const}$. The plates are slightly bent so that the filter is double focusing for 90° spin rotation (which is its normal case of operation).[50]

(vi) The Mott scattering chamber (40 cm in diameter and length) is completely magnetically shielded and at a vacuum of 2×10^{-6} mbar. The electron beam enters after passing through several collimator holes with a final aperture of 0.2 cm diam and hits in 2-cm distance the scattering foil at normal incidence. The foil is interchangeable by the rotating target holder. We used gold foils with 48.5, 99, and 211 $\mu g/cm^2$ and aluminum foils of 500 and 1120 $\mu g/cm^2$.

The Mott scattering system consists of two surface barrier detectors—located opposite to each

other—arranged so as to permit only electrons leaving the foil with $120° \pm 1.2°$ scattering angle to be detected. The effective aperture is 3 mm in diameter; the distance between foil and detector is 10 cm. The detector system can be revolved by 360°. Care is taken to align the axis of rotation with the beam axis. The rotation with the Wien filter switched on or off allows to measure the three space components of the ESP vector separately.

(vii) The preamplifiers are located within the scattering chamber directly fixed to the detectors and kept at $-5°C$ by Peltier cooling. Two independent sets of standard electronics (linear amplifier, pulse height analyzer, scaler, rate meter) process pulse rates up to one megacycle. At sufficient intensities the rate-meter outputs are used to compute on-line the ratio of the two count rates, $R = A/B$. An X-Y plotter records R versus the azimuthal position ϕ of the detector system.

In the following we give an account of checks and operation characteristics of the apparatus. Such checks are essential for the judgment on the reliability of the presently reported ESP values. In Fig. 2 an X-Y plot is shown which is recorded for field electrons extracted from an Fe-emitter tip and scattered with the 48.5-$\mu g/cm^2$ gold foil (marked R_{Au}) or the 500-$\mu g/cm^2$ aluminum foil (marked R_{Al}). The comparison between R_{Au} and R_{Al} is necessary for uncovering experimental asymmetries in the Mott detector system. The spin polarization P is calculated from

$$P = \frac{1}{S_{eff}} \frac{(R_{max}/R_{min})^{1/2} - 1}{(R_{max}/R_{min})^{1/2} + 1}, \tag{1}$$

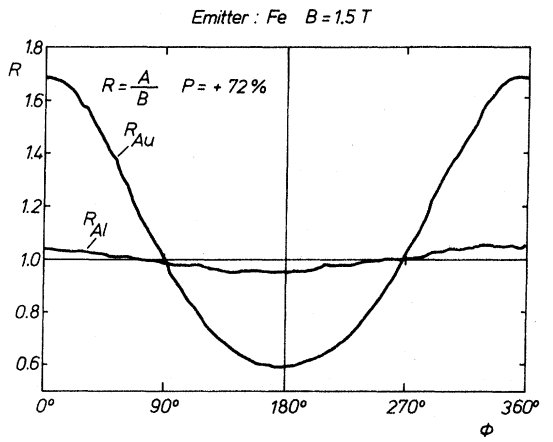

FIG. 2. Ratio $R = A/B$ of count rates of channels A and B vs rotation angle ϕ of the Mott scattering chamber, measured with Au foil 48.5 $\mu g/cm^2$ (evaporated on a Formvar foil of 20 $\mu g/cm^2$) and again with an Al foil 500 $\mu g/cm^2$ (without backing foil). The measurement is taken with an Fe tip at 10 K and 1.5 T magnetic field strength.

where R_{max} and R_{min} are the corrected maximum and minimum values of the ratio R_{Au}, respectively. The dependence of the Mott scattering on electron energy and target atomic number is accounted for by the Sherman function S.[51] In the present case the effective value S_{eff} for the thin Au foil (i.e., the value of S experimentally corrected for finite thickness of the scattering foil) is 0.35 ± 0.01. This yields $P = 72\%$ for the example given in Fig. 2.

We define the sign of the polarization P to be positive if majority spins are predominant (i.e., electrons with magnetic moment parallel to the direction of magnetization of the FE tip); a negative sign indicates that minority spins are present to a greater degree in the electron beam. We have chosen the direction of the magnetic field of the solenoid so that a maximum of R at the position $\phi = 0$ of the detector system corresponds to a positive polarization, whereas a minimum at $\phi = 0$ (or a maximum at $\phi = 180°$) is evidence of a negative polarization. Thus, Fig. 2 represents $P = +72\%$. A measurement (with a gold foil) where ϕ comes full circle takes about 1 min.

The analyzer is checked with clean tungsten FE tips, which definitely showed no polarization, $P = (0 \pm 1.5)\%$, and EuS-coated tungsten tips, which yielded $P = 90\%$, in agreement with Müller *et al.*[6] The absolute accuracy of the present ESP measurements is $\Delta P = \pm 1.5\%$, if the pair of electron counters in the Mott scattering chamber is rotated full circle with the Au and also with the Al scattering foil.

In case of high polarization values the correction for the finite thickness of the Au-scattering foil is particularly important and must be made carefully. The count rates are usually high so that statistical fluctuations are negligible.

A coordination of the measured electrons to a particular emitting area of the FE pattern of the tip cannot be controlled well enough with the rather poor electron optics of the present apparatus. Thus, if the ESP happens not to be constant over the emitting area in question, the observed polarization values may vary slightly from one measurement to the next even with the same tip.

Finally, we give a description of the tip preparation. They are made from wire material, a few millimeters in length, spot welded onto the center of a tungsten bow. The top of the wire is dipped into an electrolyte and slowly etched. The favored solutions are concentrated aqueous KOH for tungsten (7 V between tungsten and a platinum cathode); perchloric acid mixed with acetic acid 1:20 by weight for Fe tips (10 V); 10-wt.% HCl in water for Co tips (3 V); perchloric acid mixed with acetic acid 4:15 for Ni tips (15 V). First a constriction of the wire diameter is produced just above the

end of the wire. Such a reduction is formed automatically in case of W and Co, since the high specific gravity of the dissolved reaction products causes a downward flow, protecting the lower parts of the wire. In the case of Ni, the flow is upward, thereby forming the constriction just at the surface of the liquid. In the case of Fe, the top of the wire is covered with insulating lacquer; etching produces a reduction of the diameter. Then the lacquer is removed and etching is continued. In all cases the constriction finally breaks through the wire and the end piece drops off. This causes the etching current I to change rapidly with time. An electronic device sensitive to large dI/dt switches the current off; a sharp tip remains.

III. RESULTS

In this section we present detailed measurements on the transition metals Fe, Co, Ni, the rare-earth metal Gd, and compounds of Eu. Preliminary results are reported for Tb, Dy, Ho, Er, Tm. Field emission is observed at 10-K tip temperature and 1.5-T magnetic field strength unless stated otherwise.

A. Iron

The observed ESP from an iron tip is found to depend largely on the kind of pretreatment it received. Therefore, the following cases are considered separately.

(i) Tips made from polycrystalline iron wires by electrolytical etching, without any further treatment, yield at most 10% polarization in the field-emitted electrons.

(ii) The ESP obtained from such tips is found to improve considerably if heated *in situ*: After annealing at about 1300 °C during a few seconds up to +41% polarization is often observed. (The range of values found so far after such a treatment is −4% − +41%, or after annealing in H_2 up to 50%.)

(iii) The highest ESP value encountered with iron tips, +80%, is obtained after electron bombardment of the tip. Thereby, its top is raised above the melting temperature. The high ESP is found to remain usually stable over about 2 h. The same treatment sometimes produces a negative ESP occasionally as low as −13% which, however, is short lived.

(iv) By vapor depositing an iron film on an iron tip ESP values can be produced which range between (+41−−27)%, varying from one deposition and treatment to the other. The evaporation is done with the Al_2O_3 oven (see Sec. II). The negative values are found after additional annealing at 1300 °C during a few seconds only.

(v) Similar ESP values (+46−−21%) are observed with iron vapor deposited from the Al_2O_3 oven on tungsten field-emission tips.

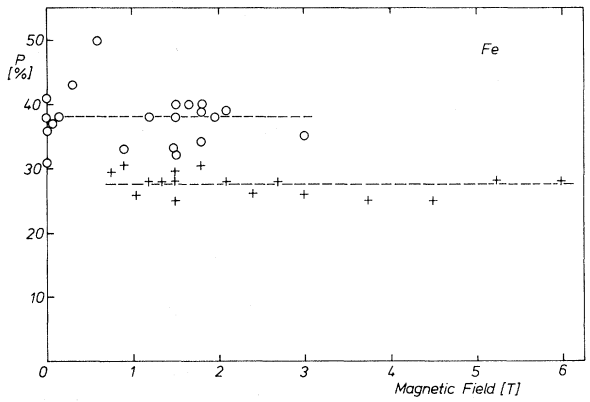

FIG. 3. Dependence of magnitude of ESP $|P|$ (in percent) upon magnetic field strength B (in tesla) for two different Fe tips (circles and crosses). The dashed lines are mean values.

(vi) The vapor pressure of Al_2O_3 at 1640 °C is about 0.1 mbar. Thus, it is likely that the deposited Fe films are contaminated. Consequently, in a series of experiments, the evaporation is accomplished by electron bombardment as explained in Sec. II. The ESP then ranges between +71%−−28%; the negative values are again obtained after an additional thermal treatment. If negative polarization is found, it appeared to be less stable than positive values which remain often steady over hours of operation.

Figure 3 shows results with variation of the external magnetic field around the tip. Two different bulk iron tips are used, both previously annealed at 1300 °C according to treatment (ii). The horizontal lines are the average values of the observed ESP, indicating an almost field-independent polarization. With tip 1 the external field is reduced to zero. The degree of polarization remains constant, the direction, however, changes from longitudinal (at field strengths > 0.1 T) to transversal. The scatter of the points at a given field strength is due to local variations of the electron-spin polarization in the field-emission patterns.

B. Cobalt

After several unsuccessful trials (except one, see below) with etched and annealed cobalt wires, we used tungsten tips covered with a cobalt film by vapor deposition. The evaporation is performed with electron bombardment (see Sec. II), thus avoiding contamination with foreign material. Shortly after evaporation the ESP is found to be negative; values as low as −36% are obtained. Annealing at 700 °C for a few seconds often changes

the ESP to positive values, up to +35%. Both polarization states are stable, e.g., in a particular experiment the ESP only rose from −27% to −26% within 20 min. Analogous to iron the polarization vector leaves its usual longitudinal alignment if the external magnetic field strength is sufficiently reduced. This happens with cobalt already below $B = 0.8$ T. At $B = 0$, the polarization vector is exactly transversal. The magnitude of the polarization, however, remains constant.

With one bulk Co tip, cleaned by field desorption, Ne bombardment and annealing in a hydrogen atmosphere at 800 °C up to +47% spin polarization is measured. The corresponding FE pattern, however, was still not perfect. The tip axis was close to ⟨111⟩, the structure fcc. We observed only positive ESP, stable over hours. Its dependence on magnetic field strength was similar to the behavior of the Co-coated tungsten tips as reported above.

C. Nickel

Neither with Fe nor with Co tips did we succeed in producing good field-electron-emission images: the tips brought about blurred pictures viewed on the fluorescent screen S_1, Fig. 1. The situation is different for bulk single-crystalline Ni tips with tip axis in ⟨100⟩ or ⟨111⟩ direction. (The nickel used is 99.995% pure.) They produce clear images if annealed in a 4×10^{-4}-mbar H_2 atmosphere at 1300 K, see Fig. 4(c). The formation of a clean surface during this treatment can be followed by intermittent viewing of the electron field-emission image at the given H_2 pressure. Then, by removing the hydrogen-atmosphere and annealing for 10 sec at 1300 K in a vacuum greater than 2×10^{-10} mbar, produces for the ⟨111⟩ tip orientation − 5% to − 2%, for the ⟨100⟩ tip orientation 0 to + 3% polarization, measured 1 min after the annealing. The effective emission directions of the tip surfaces are in fact the vicinal di-

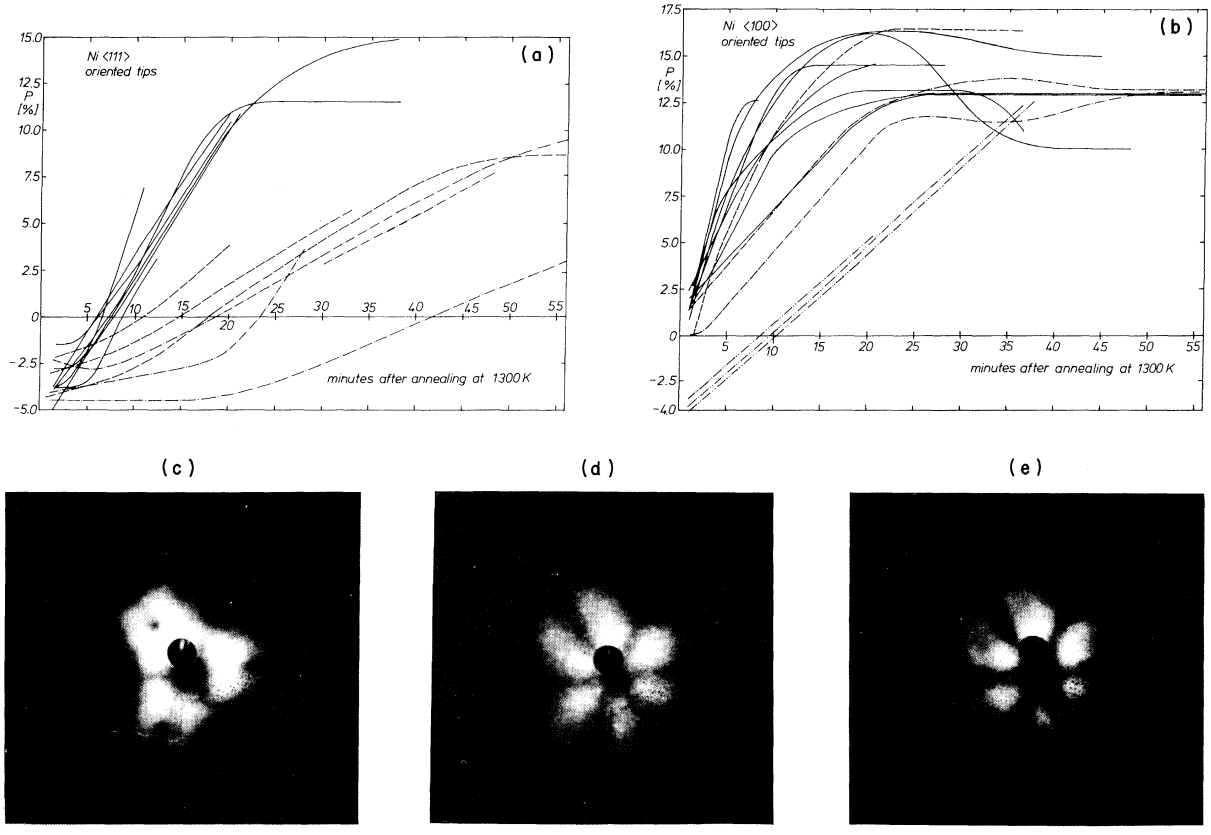

FIG. 4. (a) Time dependence of ESP for various ⟨111⟩-oriented Ni tips (annealed at 1300 K for 10 sec), as indicated by different lines. The given ESP values are uncertain by ±1. (b) Time dependence of ESP for various ⟨100⟩-oriented Ni tips analogs to (a). The dash-double-dotted line is valid for electrons from off-center parts of the FE pattern. Field emission pattern of a ⟨111⟩-oriented Ni tip, taken at 10 K. (c) Clean Ni pattern 10 sec after annealing at 1300 K; $P = (-3.5 \pm 1)\%$. (d) Same tip 100 min later, $P = (+11.3 \pm 1)\%$. (e) Same tip, completely covered with hydrogen (after admitting 10^{-6} mbar H_2), $P = (+5.4 \pm 1)\%$.

rections of the tip axis, ⟨111⟩ and ⟨100⟩, respectively.

In total we employed eight different tips, four of each orientation ⟨111⟩ and ⟨100⟩.

A remarkable feature of the single-crystalline Ni tips is that their degree of polarization increases continuously with time. These changes are different for ⟨100⟩- and ⟨111⟩-oriented tips, see Figs. 4(a) and 4(b). The final polarization, which is reached in about 2 h after annealing in vacuum is, for ⟨100⟩, +13% to +15%, and for ⟨111⟩ +10% to +13%. The change in time is independent of field emission or a magnetic field being switched on. There is evidence of attributing the increase in ESP to an increasing coverage of the tip surface with hydrogen: (i) Even at 10^{-10} mbar there is enough hydrogen for a sufficiently fast adsorption on the tip which is kept at about 10 K; (ii) By keeping the tip temperature between 400 and 450 K the polarization remains constant. This is found with the ⟨111⟩-oriented tip, where the −3.5% polarization stayed constant over 1 h of operation; (iii) The change is reversible: reannealing at ≥ 400 K restores the original polarization. Heating to 150 K has no effect in case of positive polarization. A negative polarization changes to a positive value of the same magnitude as measured at 15 K; (iv) ⟨111⟩ Ni tips completely covered with hydrogen display a unique field-emission pattern, see Fig. 4(e); (v) The change is not due to oxidation of the tip surface. Field-emission images of nickel tips covered with oxygen differ from the presently observed pattern in showing bright and narrow rings around the ⟨110⟩ or ⟨111⟩ direction. Such rings cannot be removed by reannealing in vacuum. It was found that oxidized tips do not exhibit any spin polarization of field-emitted electrons. Moreover, oxygen is practically absent in a cryovacuum.

The ESP of clean ⟨111⟩-oriented tips measured 10 sec after annealing at 1300 K is independent of the magnetic field strength: Between 0.14 and 1.5 T the spin polarization is always found to be between −3% and −4%. The ESP of ⟨100⟩- and ⟨111⟩-oriented tips covered with hydrogen is also independent of the field strength which in this case is varied between 0 and 3 T. The change from longitudinal to transversal polarization occurs at rather low fields, below 100 mT, for ⟨111⟩-oriented tips.

The temperature dependence of the ESP is investigated with the ⟨111⟩-oriented tips at 1.5 T. Measurements are listed in Table I. The field-emission pattern remains the same except the temperature exceeds about 500 K. The image then splits in six small spots around ⟨111⟩. The pattern changes again at still higher temperatures. The ESP is zero beyond T_c.

TABLE I. Variation of electron-spin polarization of a ⟨111⟩ nickel tip with temperature. Magnetic field strength 1.5 T.

T(K)	$P(T)$ (%)	T/T_c	$P(T)/P$(10K)
$10\{^{\pm 10}_{3}$	−3.5 ± 1	0.02	1
410 ± 20	−3.5 ± 1	0.65	≈1
530 ± 20	−2.1 ± 1	0.85	≈0.6
600 ± 20	−0.7 ± 1	0.96	≈0.2

In addition to the measurements with bulk Ni tips we used ⟨110⟩-oriented W tips coated with nickel by vapor deposition. No regular patterns could be obtained. The initial ESP values observed 1 to 2 min after deposition were between −7% and +8%, twice as many near zero as positive or negative values. For longer times only positive polarization up to 16% remains. This agrees with the results on bulk Ni tips except for the greater variations of the ESP. By varying the magnetizing field strength a behavior similar to the bulk Ni tips is observed.

D. Gadolinium

Electron-spin polarization is observed only with tips produced by vapor deposition of gadolinium on oriented tungsten. We used a tantalum oven to evaporate the gadolinium; the tungsten tip was kept at 600 to 700 K during deposition. In total, six ⟨110⟩- and one ⟨111⟩-oriented tungsten tips were used as substrates.

The ⟨110⟩ tips are annealed between 1000 and 1100 K for 10 sec. Thereby the (0001) plane of gadolinium becomes parallel to the (110) plane of tungsten. The field electron image consists of six symmetrical bright areas as noticed by Eckstein *et al.*[52] The ESP obtained is about +42%.

The ⟨111⟩ tip produces after annealing an image with a bright central area. The ESP is then about +53%. Annealing at 1000 K during field emission brings about nearly +50% for both orientations.

Initially, the polarization decreases slowly with time at a rate of (0.6 ± 0.2)% per minute. The residual pressure is less than 3×10^{-10} mbar and mostly due to hydrogen. After 12 h in a vacuum of 1×10^{-7} mbar the remaining polarization is between +7% and +20%; only a few lateral spots are left contributing to the field emission.

Negative ESP down to −10% is found occasionally, if a freshly filled oven for the evaporation is used, a small amount is deposited on the tungsten tip, followed by a short anneal. The field-emission pattern consists in this particular case of a few single spots only.

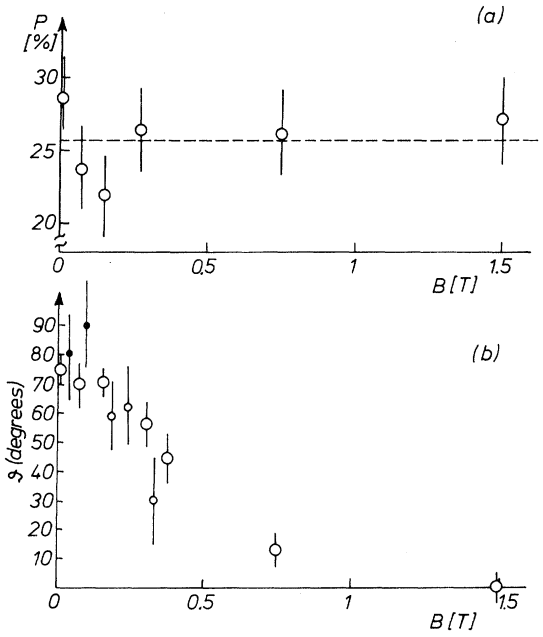

FIG. 5. (a) Dependence of magnitude of ESP, $|P|$ (in percent), upon magnetic field (in tesla) for Gd layers on W tips. To eliminate the gradual decrease of ESP with time, series of measurements with decreasing and increasing magnetic field strength are averaged. The duration of a measurement is often up to 1 h, which, therefore, gives a low mean value. (b) $\vartheta = \arctan P_x/P_z$, where ϑ is the angle of the spin vector $\vec{P}$ versus the positive z axis.

The dependence on magnetic field is shown in Fig. 5(a). (The tip temperature is 15 K.) Up to 4.5 T, the highest field applied in the present experiments, the ESP remains unaffected. Below 0.7 T the direction of the polarization starts turning transversal; at zero field it is entirely parallel to the emitting gadolinium layer, see Fig. 5(b). In these measurements we used the center part of the FE pattern.

Figure 6 illustrates the temperature dependence of ESP. We applied field strengths of 0.75, 1.5, 3.0, and 4.5 T, which, however, had no influence on ESP. The variations of ESP with temperature are reversible. The full line in Fig. 6 is the saturation magnetization of bulk gadolinium, taken from Ref. 53. The ESP values measured with the thin evaporated layers, $P(T)/P(15\text{ K})$, follow the magnetization curve $M(T)/M(4.2)$, although the points are lower.

E. Europium

Tungsten tips oriented along ⟨110⟩ were covered with europium by vaporizing an oxidized europium wire in the tantalum oven, as described in Sec. II.

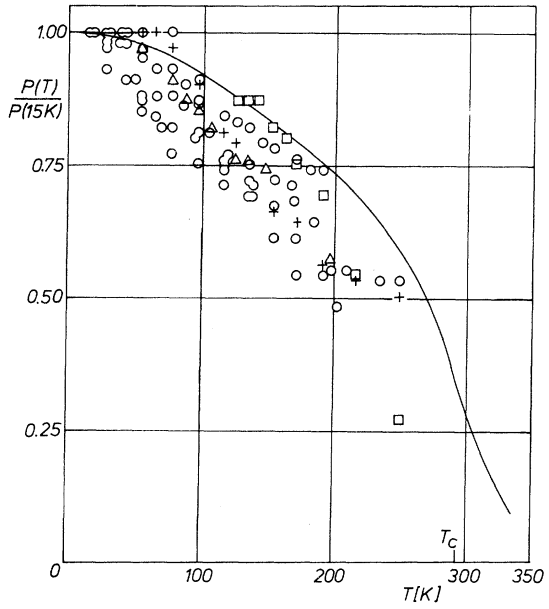

FIG. 6. Dependence of the reduced ESP $P(T)/P(15\text{ K})$ on temperature (in kelvin) for Gd layers on W tips for various magnetic field strengths: 0.75 T (Δ); 1.5 T (○); 3 T (+); 4.5 T (□). The solid line indicates for comparison the reduced spontaneous magnetization $\sigma_T/\sigma_{4.2\text{ K}} = M(T)/M(4.2\text{ K})$ at 1.7 T for the Gd c axis.

During evaporation the tip temperature was kept at 15 K; by heating the tip to 3000 K it can be cleaned again from europium. In such a series of evaporation-cleaning steps we observed that the ESP gradually decreased. This indicates that the

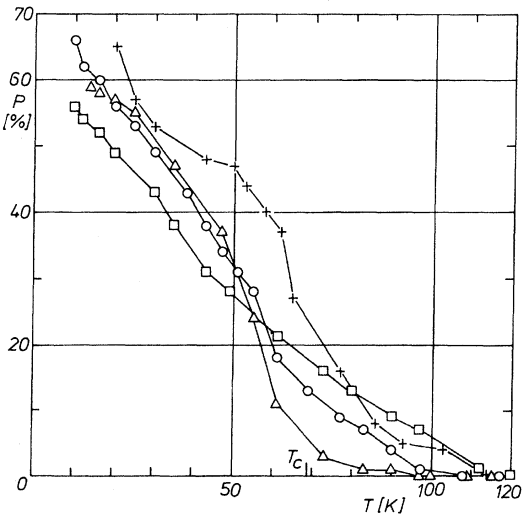

FIG. 7. Temperature dependence of the ESP of Eu layers, deposited on ⟨110⟩ W, oxidized in O_2 atmosphere, after annealing up to 1000 K for 10 sec, measured at 2×10^{-10} mbar. The parameter is the magnetic field strength B: 0.75 T (Δ); 1.5 T (○); 3 T (+); 4.5 T (□).

TABLE II. Compilation of results.

Material	Sample preparation	Annealing procedure	P observed (%)	Time dependence of P	Comments	Described in section
Fe	Massive tips	without annealing	Up to +10	Unstable		III A (i)
		1300 °C, 10 sec	− 4 to +41		$P(B)$ see Fig. 3	(ii)
		Electron bombardment	−13 to +80	Positive values stable over hours, negative values short lived	negative values only with freshly filled ovens	(iii)
	Films on Fe tips (Al_2O_3 oven)	1300 °C, 10 sec	−27 to +41			(iv)
	Films on W tips (Al_2O_3 oven)		−21 to +46			(v)
	Films on W tips (new oven)		−28 to +71			(vi)
Co	Films on ⟨110⟩ W tips	without annealing	Up to −36	Stable over hours		III B
	Films on ⟨110⟩ W tips	1000 °C, 10 sec	Up to +35			
	Tip, fcc, not clean	700 °C, 10 sec	+35 to +47			
Ni	Tips, ⟨111⟩ oriented	1000 K, 10 sec	−3.5 ± 1.5 clean	P increases due to hydrogen adsorption, see Fig. 4	$P(T)$, see Table I	III C
	Tips, ⟨100⟩ oriented		0 ± 1.5 clean			
	Films on ⟨110⟩ W tips		−7 to +8			
Gd	Films on ⟨110⟩ W tips	1000 K, 10 sec	+42 clean	P decreases with 0.6% min^{-1}	$P(B)$, see Fig. 5	III D
	Films on ⟨111⟩ W tips		+53 clean		$P(T)$, see Fig. 6	
EuS	Films on ⟨110⟩ W tips	1000 K, 10 sec	Up to +90	Stable over hours		III E
EuO	Films on ⟨110⟩ W tips	1000 K, 10 sec	Up to +89	Stable over hours	$P(T)$, see Fig. 7	
Tb	Films on ⟨110⟩ W tips	1000 K, 10 sec	−13 to +31	First results, positive values more frequent		III F (i)
Dy		without annealing	−35 to +45			(ii)
Ho		1000 K, 10 sec	−10 to +34			(iii)
Er		1000 K, 10 sec	− 8 to +66			(iv)
Tm		700 K, 10 sec	+13 at B=4.5 T, <+ 3 at B=1.5 T			(v)

TABLE III. Electron-spin measurements, polarization in percent.

	Moke (Ref. 28)	Tunneling (Refs. 20–22)	Photoemission (Refs. 11–16)	Electron capture (Refs. 24–26)	Field emission (recent investigations) Müller *et al.*, (Refs. 6 and 7)	Campagna *et al.*, (Ref. 8)	Present results
Fe		+44	+54	+38 max (100)			
Co		+34	+21	+33 max ($1\bar{1}00$)			$-27 \cdots +\underline{80}$ (layers and tips) $\underline{-36} \cdots +35$ (layers) +47 max (tip, fcc, not clean)
Ni	<0	+11	+15	−96 ⟨110⟩ −65 ⟨100⟩ −45 ⟨111⟩ +17 ⟨120⟩	−4.5±1 between ⟨110⟩ and ⟨111⟩ +3±1.2 ≈ ⟨120⟩	−2.9±0.6 ⟨112⟩, ⟨113⟩ +3.6±0.4 ⟨012⟩, ⟨013⟩	−3.5±1.5 clean around ⟨111⟩ 0±1.5 clean around ⟨100⟩ +42 ⋯ +53 clean
Gd		+4.3	+5.70±0.21				$-35 \cdots +\underline{45}$
Dy			+7.33±0.35				
EuS			+50		+89 max		+90 max
EuO			+62				+89 max

ESP is affected by contamination, presumably by europium oxide present on the Eu wire of the evaporation source. In total we used three different ⟨110⟩ tungsten tips, but obtained identical results.

After exposing such a tip covered with Eu to 0.5 mbar oxygen at room temperature for 5 min an ESP up to +89% is found in certain field-emission spots after annealing for 10 sec at 1000 K. At 10 K the ESP remains constant even if the magnetic field is varied between 0.15 and 4.5 T. The observed transversal component of the polarization vector is more likely to result from the angle of emission than from an insufficient strength of the longitudinal magnetic field.[54]

Raising the temperature decreases the ESP which reaches zero at 70 K, see Fig. 7. The temperature dependence is reversible. The Curie point of EuO is 69 K. It is likely that the high ESP originates rather from EuO than from Eu, which is antiferromagnetic below 90 K.

We investigated also EuS as an emitter of polarized electrons. The sulphide is evaporated onto tungsten tips and annealed to produce the field-emission pattern, described by Müller *et al.*[6] We found +90% polarization stable over hours of operation: Any transversal component of the polarization vector depends on the direction of emission rather than on magnetic field strength, as observed with Eu-EuO. At 100 K the ESP of EuS is still high, 28% to 33% of a EuS emitter showing 60% to 67% ESP at 10 K. The Curie temperature of bulk EuS is 16 K, see Ref. 55. However, in the experiments of Kisker *et al.*[56] with another technique of preparation, it is observed that the ESP vanishes above 16 K.

F. Other rare-earth metals

In the following we report first results on the ESP obtained with various other ferromagnetic rare-earth metals. In all cases the metal is vapor deposited on ⟨110⟩-oriented tungsten tips, using the tantalum oven. The ESP is measured at 15-K tip temperature in a field of 1.5 T. So far we have not succeeded in obtaining a regularly shaped FE pattern. The surfaces are likely to be contaminated, and, therefore, the ESP values in various evaporations of the same metal fluctuate widely.

(i) Terbium: After annealing at 1000 K for 10 sec ESP values between −13% and +31% are found.

(ii) Dysprosium: Without annealing after evaporation we obtain −35% to +45%. Any annealing reduces the ESP.

(iii) Holmium: After annealing at 1000 K for 10 sec we find −10% to +34% polarization.

(iv) Erbium: After annealing at 1000 K for 10 sec we observe −8% to +66% polarization.

(v) Thulium: Here we need a strong magnetic field, 4.5 T, to obtain +13% polarization. With 1.5 and 3 T the ESP is smaller than 3%. The tips were annealed at 700 K for 10 sec.

IV. CONCLUDING REMARKS

In general the reported results show that the ESP in field-emission experiments depends on emitter-surface conditions (preparation of the tip, annealing, adsorption), which so far are controllable only with Ni and Gd. ESP depends also on the crystallographic position of the electron-emitting spot at the tip surface. In high-magnetic fields longitudinal polarization is found. For small fields the polarization vector becomes transversal in the cases checked so far. A rotation of the spin-polarization vector with magnetic field strength as assumed in the analysis of data in Refs. 5, 57, and 58 cannot be justified anymore by the present experiments. The temperature dependence of the ESP is checked in the cases of nickel, gadolinium, and europium oxide emitters. It is found that the ESP follows roughly the saturation magnetization. The ESP disappears above the Curie temperature.

The increasing ESP of Ni emitters with hydrogen adsorption may be interpreted by resonance tunneling[59] or by change of the surface magnetization.[60]

The present results are compiled in Table II. The spread of the values for a given FE material is likely to depend on the uncertainties in surface conditions, as reported in Sec. III.

Finally, Table III lists for comparison ESP data obtained with those various other methods which are summarized in the Introduction.

Note added in proof. Electron spin polarization of secondary electrons ejected from magnetized europium oxide is demonstrated recently, see G. Chrobok and M. Hofmann, Phys. Lett. 57A, 257 (1976).

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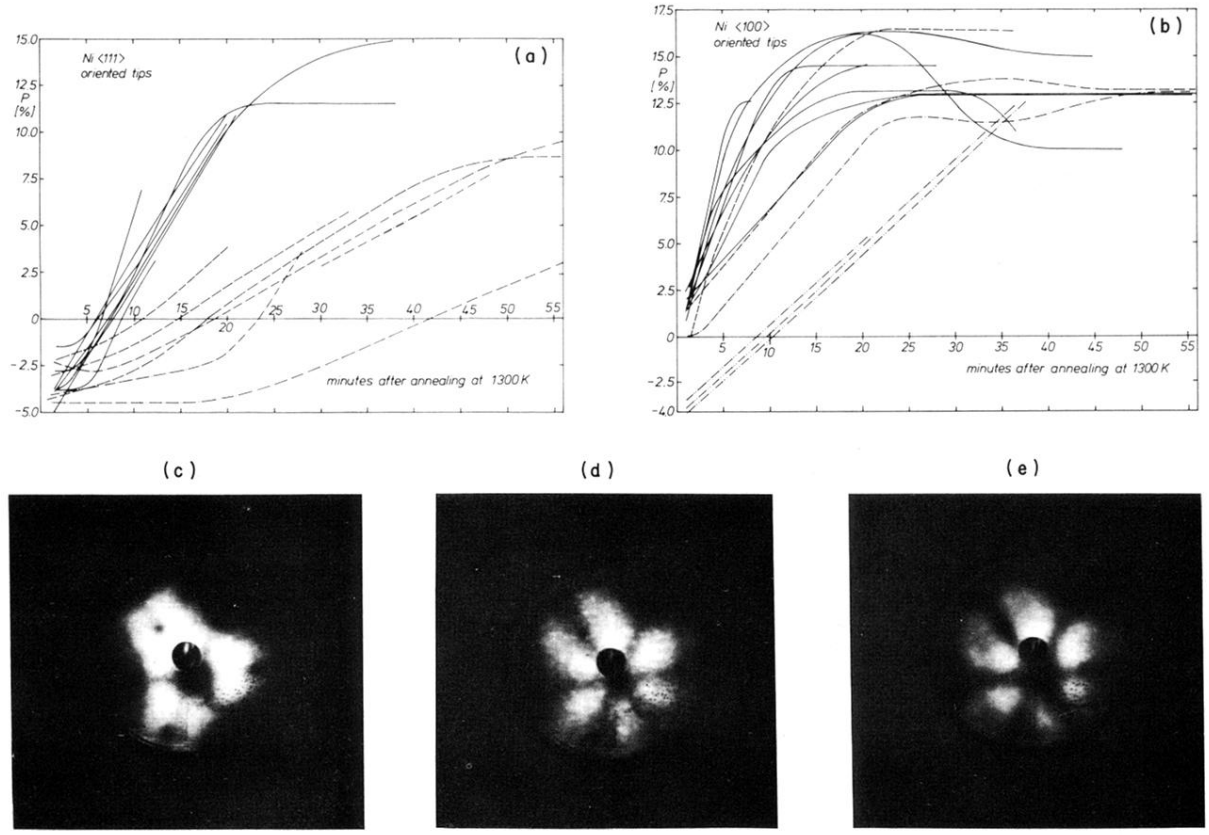

FIG. 4. (a) Time dependence of ESP for various ⟨111⟩-oriented Ni tips (annealed at 1300 K for 10 sec), as indicated by different lines. The given ESP values are uncertain by ±1. (b) Time dependence of ESP for various ⟨100⟩-oriented Ni tips analogs to (a). The dash-double-dotted line is valid for electrons from off-center parts of the FE pattern. Field emission pattern of a ⟨111⟩-oriented Ni tip, taken at 10 K. (c) Clean Ni pattern 10 sec after annealing at 1300 K; $P = (-3.5 \pm 1)\%$. (d) Same tip 100 min later, $P = (+11.3 \pm 1)\%$. (e) Same tip, completely covered with hydrogen (after admitting 10^{-6} mbar H_2), $P = (+5.4 \pm 1)\%$.