Spin Polarization of Field-Emitted Electrons from Monocrystalline Nickel

W. Gleich, G. Regenfus, and R. Sizmann
Sektion Physik der Universität München, München, Germany
(Received 4 August 1971)

Field-emitted electrons from magnetized monocrystalline Ni tips at 80 K have been investigated for spin polarization. A maximum polarization of 13% was observed. The predominant direction of the spins was parallel to the external magnetic field for electrons emitted around ⟨100⟩, ⟨110⟩, and ⟨137⟩ and antiparallel for electrons emitted around ⟨111⟩. Direction and degree of spin polarization is discussed in the context of band-structure theory and recent model calculations of field emission from 3*d* transition elements.

Band-structure calculations of ferromagnetic Ni indicate[1,2] that the spin-split electron states are predominantly of the 3*d* minority type at the Fermi surface. Since in field emission the electrons tunnel from the states at and near the Fermi edge,[3] Ni is expected to emit electrons with pronounced spin polarization. This we were able to confirm in previous experiments with polycrystalline Ni tips.[4]

Here we report on measurements of the spin polarization of electrons emitted from various crystallographic axes of 99.99%-grade nickel. The tips were etched electrolytically from ⟨100⟩-, ⟨110⟩-, ⟨111⟩-, and ⟨137⟩-oriented Ni rods.

Figure 1 shows the experimental layout. The apparatus consisted of (1) the field emission part, located in a low-temperature cryostat; (2) an accelerator to accelerate the electrons to an energy of 100 keV for Mott scattering; and (3) the polarization detector ("Mott detector"). The field-emission needle which was centered in the solenoid was kept at the temperature of liquid N_2. The first electrode maintained an electric field of about 10^7 V/cm at the tip, sufficient for a field-emission current of ≈ 10 nA. The pulsed magnetic field pointed parallel or antiparallel to the direction of electron acceleration (the $+z$ axis).

After acceleration the electron linear momentum is turned through 107° in a spherical condensor, thus converting the longitudinal polarization into transverse. Then the degree of polarization was analyzed using a standard technique: The electrons were scattered alternately by thin gold and aluminum foils, and the asymmetry of the left- and right-scattered intensities was determined with two surface-barrier counters which were located symmetrically opposite to each other in a plane perpendicular to the beam (the detection plane, cf. Fig. 1).

The scattering angle of the electrons was 120°. The two counters could be rotated around the beam direction. This allowed the projection $\vec{P}_D$ of the polarization vector onto the detection plane

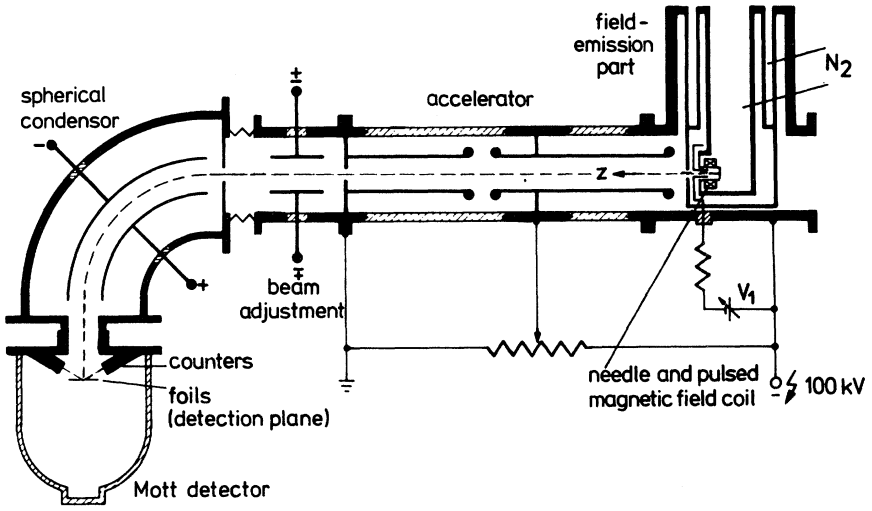

FIG. 1. Layout of the apparatus (not to scale). Extraction voltage $V_1 \approx 2$ kV. Vacuum at the emitter tip $\approx 10^{-8}$ Torr.

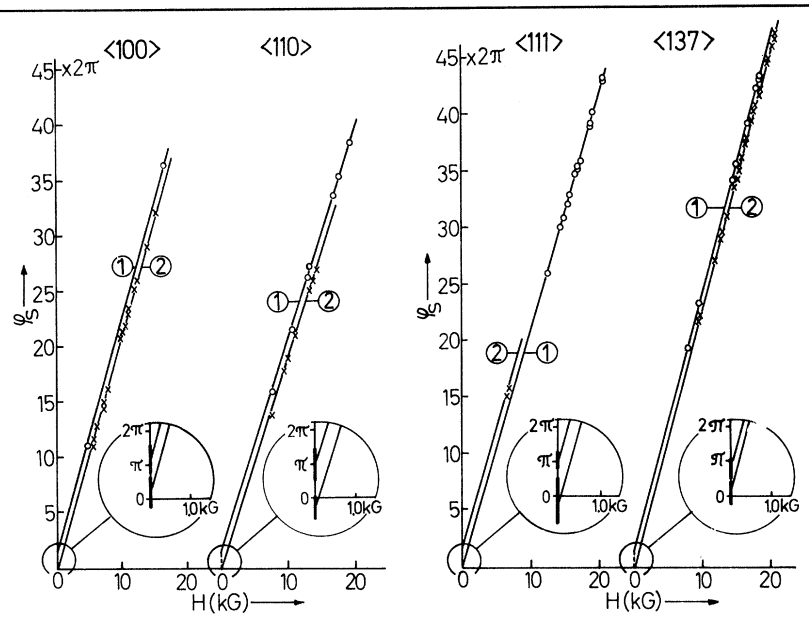

FIG. 2. Rotation of spin polarization vector $\vec{P}_D$ in the detection plane. φ_s is the azimuthal angle of $\vec{P}_D$ (in units of 2π), versus external magnetic field (in kilogauss) at the field-emission tip. The results are plotted for the ⟨100⟩, ⟨110⟩, ⟨111⟩, and ⟨137⟩ directions of emission from monocrystalline Ni tips at 80 K. The magnetic field H points in $-z$ for line 1 and in $+z$ for line 2. The enlarged inserts show the ordinate intercepts of lines for $H=0$ and the error bars (standard deviations). The slopes of the lines are [(2.03 ± 0.02), (1.82 ± 0.02), (2.06 ± 0.02), and (2.21 ± 0.01)]2π (kG)$^{-1}$ for ⟨100⟩, ⟨110⟩, ⟨111⟩, and ⟨138⟩, respectively. For scaling reasons the lines 1 and 2 are plotted with a phase difference of 3π instead of π.

to be measured. It was found that $\vec{P}_D$ is in general not at $\varphi = 0$ or π but at some angle φ_s (cf. Ref. 4), where the sense of φ is taken from the $+z$ direction.

The determination of $\vec{P}_D$ indicated a precession of the spin polarization in the detection plane and a change in absolute value for a stepwise-increasing magnetic field. With respect to the direction opposite to the electron beam, the sense of rotation of $\vec{P}_D$ was anticlockwise for an increasing magnetic field pointing in the $+z$ direction, H_{+z}, and clockwise for an increasing magnetic field pointing in the $-z$ direction, H_{-z}.

Figure 2 shows the linear behavior of φ_s with variation of magnetic field strength H_{+z} for the various crystal orientations under investigation. The ordinate scale is in units of full spin rotations, $2\pi n$. The slopes of the lines measured for each tip at H_{+z} or H_{-z} are identical within the error limits. The $H_{\pm z}$ pairs of lines exhibit a phase difference in φ_s varying little from π. This corresponds to the reversal of the spin polarization with reversal of the external magnetic field.

It will be seen from Fig. 2 that either the ordinate intercept of the line measured with H_{+z} or the intercept measured with inverted field H_{-z} is zero. This proves to be the decisive point of the experiments. In fact, there are two cases in Fig. 2: For ⟨100⟩, ⟨110⟩, and ⟨137⟩ it is the H_{+z} line which passes through the origin, but for ⟨111⟩ this occurs for the H_{-z} line. Since $\varphi_s = 0$ in the limit of external $H_{+z} \to 0$ ($H_{-z} \to 0$) means that the tunneling process delivers electrons whose spins are preferentially parallel (antiparallel) to the field direction, we conclude that the electrons emitted along ⟨111⟩ and those emitted along ⟨100⟩, ⟨110⟩, and ⟨137⟩ have opposite overall spin polarization.

In Table I a summary is given of the experimental results. It should be remarked that in consecutive runs the P_D values show large fluctuations, whereas the φ_s value at a given external magnetic field strength is always found to be the same to within ± 40°.

The observed spin rotation presents an intriguing problem. The cyclotron rotation angle φ_c as determined on a luminescent screen placed in the detection plane (cf. Fig. 1) agrees in sign and magnitude with the well-known relationship $(e/m) \times \int v^{-1} B(z)\,dz$, when computed along the magnetic field configuration beginning at the tip position. In addition, the experimental dependence of φ_c on extraction voltage V_1 and tip position in the solenoid corresponds well with this relationship. On

TABLE I. Summary of experimental values of direction and magnitude of the spin polarization of field electrons from nickel.

Direction of emission around $\langle hkl \rangle$	Spin polarization relative to external magnetic field H	Maximum polarization measured P_D^{max} (%)
$\langle 100 \rangle$	Parallel	10.0 ± 2
$\langle 110 \rangle$	Parallel	7.8 ± 2
$\langle 137 \rangle$	Parallel	9.5 ± 2
$\langle 111 \rangle$	Antiparallel	7.5 ± 2
$\langle hkl \rangle$[a]	Parallel	13.0 ± 2

[a]Measurements on seven different tips of polycrystalline material with unknown crystal orientation but of the same preferential cold working texture.

the other hand, we notice that φ_s is 4 to 7 times φ_c and that the slopes of φ_s vs H (cf. Fig. 2) are almost independent of V_1 and the tip position. A computation of φ_s according to the relations for spin rotation in electromagnetic fields given by Bargmann, Michel, and Telegdi[5] yields, in the present field configuration, only a few tenths of a percent of the observed value φ_s. From this we conclude that those equations have no application to the strongly inhomogeneous field in question, or that the spin rotation already takes place during the emission process.

For an interpretation of the measured spin polarization we refer to recent theoretical work. Band-structure calculations by Hodges, Ehrenreich, and Lang[1] and also by Connolly[2] indicate that around the axes $\langle 100 \rangle$, $\langle 110 \rangle$, and $\langle 137 \rangle$ the main fraction of the electrons may tunnel from the minority β bands, whereas the emission around $\langle 111 \rangle$ may stem predominantly from the majority 6α electrons. This is in accord with the experimentally determined directions of the spin polarization (cf. Table I, column 2).

To estimate the magnitude of the spin polarization we elaborate the reasoning of Gadzuk[6] about the partition of the total field-emission current I_{tot} between contributions from the $4s$ and $3d$ bands, $I_{tot} = I_{4s} + I_{3d}$. In line with this we assume the spin polarization to be

$$P = (I_{4s\uparrow} + I_{3d\uparrow} - I_{4s\downarrow} - I_{3d\downarrow})/I_{tot}, \quad (1)$$

where $I_{4s\uparrow}$, $I_{4s\downarrow}$, $I_{3d\uparrow}$, and $I_{3d\downarrow}$ are the partial currents from the spin-split bands. In Ni and Fe the $4s$ electrons are hardly split by the spin.[7] This reduces Eq. (1) to

$$P \approx (I_{3d\uparrow} - I_{3d\downarrow})/I_{tot}. \quad (2)$$

Now, for Fe the density of states in $3d\uparrow$ and $3d\downarrow$ is nearly equal at the Fermi surface.[8] In fact, the spin polarization measured with Fe tips is found to be smaller than $\approx 6\%$.[9] In certain crystallographic directions of Ni, however, one of the two partial $3d$ currents in Eq. (2) dominates. For a rough estimate of the magnitude of P we may proceed as follows: The ratio of the tunneling probabilities of d and s electrons, T_d/T_s, from Ni in $\langle 100 \rangle$ is calculated by Politzer and Cutler[10] to be about 10^{-2}. The density of states in the $3d$ band is about 10 times higher than in the $4s$ band. Then, the product of the two quantities, ≈ 0.1, is a measure of the polarization of field-emitted electrons. The experimental value, 0.1 ± 0.02 (cf. Table I), is in accord with this estimate.

Field emission probes a depth of ≈ 0.1 eV at the Fermi level. In contrast, by photoemission, electrons are expelled from variable depths, e.g., from 0.4 and 0.8 eV in the measurements of Bänninger *et al.*[11] on polarized photoelectrons from polycrystalline Ni. Differences in degree and even in direction of the polarization found with both methods are conceivable (cf. the density of states in Ni as measured by Eastman and Krolikowsky[12] and discussed by Zornberg[7]).

In conclusion we may note that the investigation of the spin polarization of electrons emitted around particular crystallographic axes appears as a valuable new tool for studying local properties of the Fermi surface.

The authors would like to thank G. Chrobok, M. Hofmann, and R. Helwig for valuable discussions, and Dr. D. Krause for supplying single crystals. The work was supported by the Bundesministerium für Bildung und Wissenschaft.

[1]L. Hodges, H. Ehrenreich, and N. D. Lang, Phys. Rev. 152, 505 (1966).

[2]H. W. D. Connolly, Phys. Rev. 159, 415 (1967).

[3]R. H. Good and E. W. Müller, in *Handbuch der Physik*, edited by S. Flügge (Springer, Berlin, 1956), Vol.

XXI, p. 189.
[4]G. Regenfus and R. Helwig, Verh. Deut. Phys. Ges. 5, 181 (1970).
[5]V. Bargmann, L. Michel, and V. L. Telegdi, Phys. Rev. Lett. 2, 435 (1959).
[6]J. W. Gadzuk, Phys. Rev. 182, 416 (1969).
[7]E. I. Zornberg, Phys. Rev. B 1, 244 (1970).
[8]S. Wakoh and J. Yamashita, J. Phys. Soc. Jap. 21, 1712 (1966).
[9]G. Regenfus, to be published.
[10]B. A. Politzer and P. H. Cutler, Surface Sci. 22, 277 (1970).
[11]U. Bänninger, G. Busch, M. Campagna, and H. Chr. Siegmann, Phys. Rev. Lett. 25, 585 (1970).
[12]D. E. Eastman and W. F. Krolikowsky, Phys. Rev. Lett. 21, 623 (1968).

Theorem for Energy-Weighted Averages of Spectroscopic Factors*

C. M. Shakin† and J. Da Providência
Laboratório de Física, Universidade de Coimbra, Coimbra, Portugal
(Received 23 July 1971)

It is shown that for a system with strong correlations, the centroid of the spectroscopic amplitudes for particle removal is given by the single-particle energy (defined appropriately) times the occupation probability of the orbit.

If a particle is removed from a system very rapidly, or if the distorted-wave Born approximation is expected to apply to the description of the removal, then the states of the residual system are expected to be excited with a strength proportional to the spectroscopic factors $|\langle\Psi_n|a_i \times|\Psi_0\rangle|^2$. Here $|\Psi_0\rangle$ is the ground state of the target, $|\Psi_n\rangle$ is the state of the residual system, and a_i is the destruction operator for a particle in the orbit i. The energy-weighted average of the spectroscopic factors is a quantity of current theoretical interest,[1] particularly with respect to the interpretation of $(p,2p)$ and $(e,e'p)$ experiments at high energies.

We assume we have a system whose dynamics is governed by a Hamiltonian H containing two-body forces. (We will use the notation where $i,j,k,\cdots$ refer to occupied states, $m,n,p,\cdots$ to unoccupied states, and $\alpha,\beta,\gamma,\cdots$ to either.)

Consider the operator defined through the relation[1]

$$W_i=\{a_i^\dagger,[H,a_i]\}, \tag{1}$$

where the curly brackets mean an anticommutator. It is easily seen that

$$\langle\Psi_0|W_i|\Psi_0\rangle=\sum_n(E_n-E_0)|\langle\Psi_n|a_i|\Psi_0\rangle|^2 -\sum_n(E_n-E_0)|\langle\Psi_n|a_i^\dagger|\Psi_0\rangle|^2, \tag{2}$$

where $H|\Psi_0\rangle=E_0|\Psi_0\rangle$, $H|\Psi_n\rangle=E_n|\Psi_n\rangle$. It is well known that in the Hartree-Fock approximation $\langle\Psi_0|W_i|\Psi_0\rangle=\epsilon_i$, where ϵ_i is the Hartree-Fock single-particle energy. In general the operator W_i is given by

$$W_i=\langle i|t|i\rangle+\sum_{\alpha\beta}\langle i\alpha|v|i\beta\rangle_A a_\alpha^\dagger a_\beta, \tag{3}$$

where t is a kinetic-energy operator and

$$\langle i\alpha|v|i\beta\rangle_A=\langle i\alpha|v|i\beta\rangle-\langle i\alpha|v|\beta i\rangle.$$

It is clear that in a system with singular interactions the ground-state expectation value of W_i is infinite and Eq. (2) is not useful. We can trace this difficulty to the use of the anticommutator. If we denote the correlated ground state by $|\Psi_0\rangle$, we note that the quantity $\langle\Psi_0|[H,a_i]a_i^\dagger \times|\Psi_0\rangle$ is the source of the divergence of $\langle\Psi_0|W_i \times|\Psi_0\rangle$. This is because we are introducing a particle in orbit i into the system in such a manner that its correlations (due to the hard core) with the other particles are absent. Thus we are led to the consideration of the operator

$$\Omega_i=a_i^\dagger[H,a_i], \tag{4}$$

where, as before, we have

$$C_i\equiv\langle\Psi_0|\Omega_i|\Psi_0\rangle=\sum_n(E_n-E_0)|\langle\Psi_n|a_i|\Psi_0\rangle|^2. \tag{5}$$

We may write, in general,

$$|\Psi_0\rangle=F|\Phi_0\rangle/\langle\Phi_0|F^\dagger F|\Phi_0\rangle^{1/2}, \tag{6}$$

$$F=1+F^{(2)}+F^{(3)}+\cdots,$$

where $F^{(n)}$ is an n-body operator. (We may also write $F=e^S$, with $S=S^{(2)}+S^{(3)}+\cdots$. The neglect of $S^{(n)}$, $n>2$, is equivalent to the neglect of three-