and which thus rapidly increases as the incommensurate-paraelectric transition $T_I$ is approached. In the vicinity of the defects the phase will be pinned, whereas in the region between the defects we shall have large phase fluctuations (i.e., the modulation wave will be rough). The radius of the "pinned" region (where the root mean square phase fluctuation amplitude is smaller than $\pi/2$) slowly decreases with increasing temperature, whereas the "floating" part (where the root mean square phase fluctuation amplitude exceeds $\pi/2$) increases with the mean distance between "pinning" defects. The fraction $\xi$ of the pinned nuclei giving rise to the broad NMR line thus decreases critically as the incommensurate-paraelectric transition temperature is approached:

$$\xi \propto (T_I - T)^{\beta(\gamma-1)}. \quad (13)$$

Thus NMR lines from the pinned and the floating regions should be observed below $T_I$ if the number of defects is not too large.

The experimental data show that $\gamma \approx 2$.

The appearance of the sharp NMR lines is thus due to a decrease in the strength of the phase pinning potential of impurities with increasing temperature and the corresponding increase in the mean distance between pinning defects. This increase in distance results in an increase in the mean square vibration amplitude of the modulation wave, which thus becomes "rough" and floating over an increasing part of the crystal. The observed phenomenon is thus similar to a roughening transition. Similar effects should be observable close to $T_I$ in other incommensurate systems provided the pinning defect concentration is not too large.

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[1]W. Finger and T. M. Rice, Phys. Rev. Lett. 49, 468 (1982); T. M. Rice, S. Whitehead, and P. Littlewood, Phys. Rev. B 24, 275 (1981).

[2]R. A. Cowley and A. D. Bruce, J. Phys. C 11, 3577 (1978).

[3]D. A. Bruce, J. Phys. C 13, 4615 (1980).

[4]V. J. Emery and J. D. Axe, Phys. Rev. Lett. 40, 1507 (1978).

[5]R. Blinc, Phys. Rep. 79, 333 (1981), and references therein.

[6]B. H. Suits, S. Couturie, and C. P. Slichter, Phys. Rev. Lett. 45, 194 (1980), and Phys. Rev. B 23, 5142 (1981).

[7]R. Blinc, S. Juznic, V. Rutar, J. Seliger, and S. Zumer, Phys. Rev. Lett. 44, 609 (1980).

[8]V. Rutar, F. Milia, B. Topic, R. Blinc, and Th. Rasing, Phys. Rev. B 25, 281 (1982).

[9]A. Abragam, *The Principles of Nuclear Magnetism* (Oxford Univ. Press, New York, 1961).

[10]S. Zumer and R. Blinc, J. Phys. C 14, 465 (1981).

## Evidence for Spin-Dependent Electron-Hole-Pair Excitations in Spin-Polarized Secondary-Electron Emission from Ni(110)

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The energy-resolved spin polarization $P$ of secondary electrons from Ni(110) has been measured with high energy resolution. At the lowest kinetic energy $P$ reaches a maximum of $(17 \pm 2)\%$ which is significantly higher than the mean conduction-band polarization (5.5%). Within about 3 eV towards higher kinetic energy the spin polarization decreases to about 8% and displays pronounced structures at higher energies. Spin-dependent electron-hole-pair excitations are suggested to be responsible for the observed spin polarization.

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Primary electrons with sufficient energy impinging on a solid generate low-energy secondary electrons via a cascade process. Within the cascade process, the number of hot electrons is multiplied due to scattering of the excited electrons primarily by conduction-band electrons.[1] In the absence of spin-dependent scattering processes it had been thought that the spin polarization of the secondaries directly reflected the net spin polarization of the conduction bands, $P = (n\uparrow$

$-n\downarrow)/(n\uparrow+n\downarrow)=n_B/n$, where $n_B$ is the number of Bohr magnetons/atom (e.g., $n_B=0.55$ for Ni). In two recent measurements, one on polycrystalline Fe films[2] and another one on amorphous $Fe_{81.5}B_{14.5}Si_4$,[3] it was found that the polarization of the secondaries with the lowest kinetic energy is highest and equal to the average magnetization of the conduction-band electrons and that it decreases to higher kinetic energies.

We report here energy-resolved spin polarization measurements with high resolution, performed on a clean Ni(110) single-crystal surface, which demonstrate that the scattering processes, which are connected with the secondary electron production, have a high spin asymmetry. The resulting spin polarization of the secondaries of lowest kinetic energy is larger by more than a factor of 3 than the average conduction-band polarization. In addition the measurements reveal structures in the energy-resolved spin polarization of the secondaries which have not been observed previously.

The measurements were carried out in a new UHV apparatus designed for energy- and angle-resolved spin-polarized-electron spectroscopies. Details of the apparatus will be published elsewhere.[4] Briefly (see Fig. 1), it consists of a hemispherical (180°) energy analyzer coupled to a high-efficiency Mott detector (100 kV) for spin analysis. The high transmission efficiency of the apparatus is enhanced by employing multichannel detection in the Mott detector, covering a range of scattering angles between 95° and 145°, together with an optimized electron lens system.[2] The calibration of the Mott detector is at present estimated to be correct to about 10% (relative accuracy), whereas relative polarizations can be measured within a few tenths of a percent, depending only on the counting statistics. The energy analyzer was used in the fixed pass-energy mode which was set to 4 eV for the present study yielding an energy resolution of 0.2 eV. A lens system is used to accelerate or retard the electrons to the appropriate pass energy. The angle of incidence of the primary electrons (400 eV) was 30° with respect to the surface normal. The sample was aligned with its surface normal parallel to the electron optical axis of the energy analyzer within 3°. A negative bias voltage (30 V) was applied to the sample resulting in an energy-dependent angle resolution, which vanishes for zero starting energy. The sample was a (110) oriented, so-called "picture-frame" shaped single crystal (see inset of Fig. 1) with a very small magnetic stray field. The crystal can be magnetized by a current pulse through a coil wound around one of its legs and the magnetization direction can easily be reversed. The sample was prepared in the usual way, consisting of Laue orientation (within 1°), spark cutting, and mechanical polishing. It was cleaned *in situ* by prolonged Ar-ion sputtering and heating cycles. The surface condition was checked by low-energy electron diffraction and Auger spectroscopy. Data accumulation time was typically 30–40 min for one spectrum.

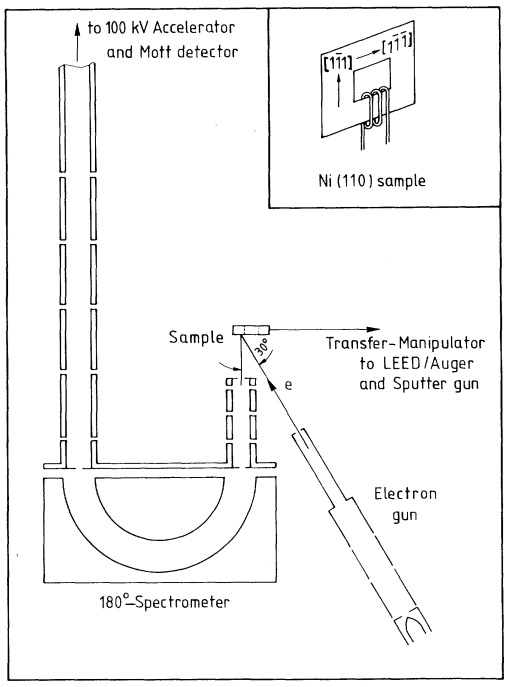


FIG. 1. Scheme of the experimental setup and picture frame Ni single crystal (inset).

The measured energy distribution of the secondaries $N(E)$ and the corresponding spin polarization $P(E)$ are shown in Fig. 2. We checked in different runs over smaller energy ranges with better statistics that the structures as emphasized by the solid lines are real. At the vacuum cutoff ($E=0$) we measure a spin polarization of $P=17\pm2\%$ which is significantly higher than the average conduction-band spin polarization $P_b=5.5\%$ of Ni. The spin polarization decreases to ~8% within about 3 eV above the vacuum cutoff. To enhance small structures in the $N(E)$ curve we show in Fig. 2 also the second derivative $d^2N(E)/dE^2$. We see that maxima in the spin polarization curve at 4, 8.5, and 15.5 eV correspond to structures in the energy distribution

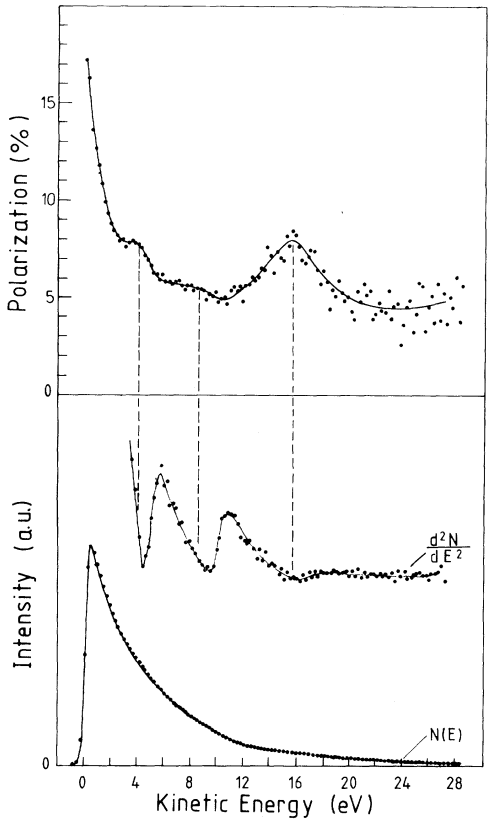


FIG. 2. Energy distribution $N(E)$ and spin polarization $P(E)$ of the secondary electrons as a function of kinetic energy (energy above $E_v$). The second derivative $d^2N(E)/dE^2$ emphasizes weak structures in the $N(E)$ curve.

curve. These energies compare well with the main peaks observed by Kato *et al.*[5] in a secondary-emission study on Ni(100) and have been attributed to maxima in the density of states (DOS) above the vacuum level $E_v$. Since the small structures in $N(E)$ are superimposed on a large rapidly varying background we have also determined the positions by subtraction of a smooth background function, e.g., an exponential, which essentially yields the same positions.

It is known that the cascade of the secondaries originates from inelastic scattering processes to which, for example, the following processes contribute: (i) radiative deexcitation ("inverse photoemission"), (ii) electron-electron scattering, involving electron-hole-pair creation, (iii) plasmon excitation and decay, and (iv) electron-magnon interaction. Process (i) is strongly spin asymmetric since there are only empty minority-spin states available within the high density of states of the $d$ bands. But the probability for this process is small.[6] Process (ii) is generally considered to be responsible for the cascade of secondary electrons and is spin dependent,[7-10] even for free electrons.[11] Processes (iii) and (iv) have been shown to be nearly spin independent.[12,13]

For small energy losses transitions within the $d$ bands are very likely. Since ferromagnetic Ni is characterized by its fully occupied majority-spin $d$-bands,[14] $d$-electron excitation is restricted to excitations within the minority-spin bands and to excitations from majority-spin bands to minority-spin ones (involving spin flip). Considering also the spin conservation rule, the following inelastic scattering processes are possible:

$$\uparrow(E_1)+\downarrow(E_0)\rightarrow\uparrow(E_1-\Delta E)+\downarrow(E_0+\Delta E), \quad (1)$$

$$\downarrow(E_1)+\downarrow(E_0)\rightarrow\downarrow(E_1-\Delta E)+\downarrow(E_0+\Delta E), \quad (2)$$

$$\downarrow(E_1)+\uparrow(E_0)\rightarrow\uparrow(E_1-\Delta E)+\downarrow(E_0+\Delta E). \quad (3)$$

$E_1$ is the energy of an excited electron before inelastic scattering, and $E_0$ is the energy of a $d$-band electron, and $\Delta E$ is the energy transfer in the scattering process. Excited minority-spin electrons, therefore, have a higher probability of being inelastically scattered than majority ones. Since, for electrons with energy less than $W$, where $W$ is the width of the $d$ bands, the chance of being trapped by the vacuum barrier after scattering increases with decreasing energy, an increase of the spin polarization to lower kinetic energies is expected. The probability of the spin exchange also increases with decreasing energy, and the number of spin-up electrons is therefore increased at the cost of the number of spin-down electrons. These two effects are considered to be the reason for the observed strong increase of the spin polarization towards zero kinetic energy. We note that these arguments apply also to Co, and, in a modified form, to Fe, where an increase of the spin polarization by about a factor of 2 at zero kinetic energy has recently been observed in photoemission experiments with synchrotron radiation.[15]

Current theories (Refs. 7–10 and 16) do not offer an explanation of the structures in the spin polarization curve at higher energies, especially the pronounced, symmetric peak at 15.5 eV.

We regard the structures in the spin polarization curve at higher energies also as originating from spin-flip processes. The correlation between the maxima in the spin polarization curve and the intensity curve suggests that they are of common origin. The intensity maxima occur near maxima in the DOS, which, for energies above

$E_v$, has been calculated by Szmulovicz and Pease.[17] When the DOS is large, the probability for a small energy loss is enhanced since the final state then falls in a region with a high DOS. Then the spin asymmetric processes (1)–(3) dominate over other energy-loss mechanisms which are not spin dependent, and a relative maximum is expected in the $P(E)$ curve due to the preferential spin-flip scattering of minority-spin electrons [process (3)]. In this context the present data strongly support the model of Yin and Tosatti,[10] who considered explicitly band-structure effects in electron-hole-pair excitation and showed that spin asymmetries can be very large at low energies.

We have noticed that the spin polarization is very surface sensitive. For example, an exposure to 0.5 L oxygen (1 L = $10^{-6}$ Torr sec) reduces the spin polarization at zero kinetic energy from 17% to ~12% although these low-energy electrons have a rather large mean free path (~30 Å). We suspect the reason for the surface sensitivity is that the electrons with low kinetic energy are predominantly emitted at grazing angles. This is consistent with the fact that we find in the angle-integrated energy distribution $N(E)$ the maximum intensity at zero kinetic energy whereas in the angle-resolved measurements the maximum is found between 5 and 20 eV.[1,18]

In conclusion we have shown that the spin polarization of electron-excited secondary electrons is influenced by spin-dependent inelastic scattering leading to a large increase of the spin polarization at low energies and to structures which are related to the DOS. These spin-dependent electron-hole-pair excitation processes are expected to be also important in the analysis of spin-polarized low-energy electron scattering from ferromagnetic surfaces.

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[1]R. F. Willis and N. E. Christensen, Phys. Rev. B 18, 5140 (1978).

[2]E. Kisker, R. Clauberg, and W. Gudat, Rev. Sci. Instrum. 53, 1137 (1982).

[3]J. Unguris, D. T. Pierce, A. Galejs, and R. J. Celotta, Phys. Rev. Lett. 49, 72 (1982).

[4]H. Hopster, E. Kisker, and R. Raue, to be published.

[5]H. Kato, Y. Sakisaka, M. Nishijima, and M. Onchi, Phys. Rev. B 22, 1709 (1980).

[6]J. B. Pendry, Phys. Rev. Lett. 45, 1356 (1980).

[7]A. Bringer, M. Campagna, R. Feder, W. Gudat, E. Kisker, and E. Kuhlmann, Phys. Rev. Lett. 42, 1705 (1979).

[8]R. W. Rendell and D. R. Penn, Phys. Rev. Lett. 45, 2057 (1980).

[9]J. A. D. Matthew, Phys. Rev. B 25, 3326 (1982).

[10]S. Yin and E. Tosatti, International Center for Theoretical Physics, Trieste, Italy, Report No. IC/81/129 (to be published).

[11]A. M. Bincer, Phys. Rev. B 107, 1434 (1957).

[12]J. S. Helman and W. Baltensperger, Phys. Rev. B 22, 1300 (1980).

[13]U. Bänninger, G. Busch, M. Campagna, and H. C. Siegmann, J. Phys. (Paris), Colloq. 32, C1-290 (1972).

[14]W. L. Moruzzi, J. F. Janak, and A. R. Williams, *Calculated Electronic Properties of Metals* (Pergamon, New York, 1978).

[15]E. Kisker, W. Gudat, and K. Schröder, Solid State Commun. 44, 591 (1982).

[16]Reference 9 would account for sharp spin polarization structures. It has been shown there that at "important" inelastic thresholds scattering takes place only in the singlet state, which would lead to a spin polarization enhancement. We cannot presently identify a certain inelastic threshold near 15.5 eV.

[17]F. Szmulowicz and D. M. Pease, Phys. Rev. B 17, 3341 (1978).

[18]J. Schäfer, R. Schoppe, J. Hölzl, and R. Feder, Surf. Sci. 107, 290 (1981).