Surface Magnetism of Ni(100) near the Critical Region by Spin-Polarized Electron Scattering

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It is shown that scattering of polarized low-energy electrons can be used to study magnetic *critical* behavior of well-characterized, free surfaces. For the topmost layers of the Ni(100) surface of a single-crystal magnetic circuit it is found that, in the temperature range $0.008 \leqslant 1-T/T_C \leqslant 0.1$, the magnetization decreases with the critical exponent $\beta_1 = 0.825^{+0.025}_{-0.040}$, possibly indicating XY coupling at the Ni surface.

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Critical behavior of semi-infinite systems has been and still is the subject of great current theoretical interest.[1-6] In contrast, experimental data on free, well-characterized, clean surfaces have been rather scarce, primarily, we believe, because of the lack of adequate, easily accessible probes. The first pioneering steps in this direction have been taken by Palmberg, DeWames, and Vredevoe,[7] with the low-energy electron-diffraction (LEED) technique, and about ten years later by Celotta *et al.*[8] with spin-polarized LEED (SPLEED). In this Letter, we report results of an experimental effort towards the systematic investigation of the critical behavior of magnetic surfaces. It is shown that by scattering of spin-polarized low-energy electrons, information on the surface *critical behavior* of itinerant-electron ferromagnets can be made available. As a test for the feasibility of the experiment we have chosen the model itinerant-electron ferromagnet Ni.

In Fig. 1 we show a scheme of the scattering geometry involving a very special target geometry of the Ni crystal so as to reduce the magnetic stray fields.[9] The magnetic single-crystal circuit is magnetized by a current pulse through a self-supporting Cu coil wound around one leg of the frame. An electron beam of transversal spin polarization $\vec{P}_0$ and kinetic energy E_k, obtained from an $Al_xGa_{1-x}As$ spin-polarized source, is scattered from the front surface of the picture-frame single crystal which is magnetized along the [110] direction parallel or antiparallel to $\vec{P}_0$. In the present experimental arrangement the direction of the magnetization $\vec{M}_{110}$ is parallel to the normal of the scattering plane defined by $\vec{n} = (\vec{k}_1 \times \vec{k}_2)/|\vec{k}_1 \times \vec{k}_2|$ (see Fig. 1). The intensity of the specularly reflected elastic electron beam is measured for $\vec{P}_0$ parallel and antiparallel to the magnetization $\vec{M}_{110}$ as a function of the temperature. In addition the experimental determination of the asymmetry in the scattered intensity is done for $\vec{M}_{110}$ in both directions, parallel and antiparallel to the scattering plane, so as to determine its true zero.

The intensity of the scattered electron beam is determined by the Hamiltonian[10,11]

$$H = V + V_{so} + \sum_j J(\vec{r} - \vec{R}_j)\vec{s} \cdot \vec{S}_j, \qquad (1)$$

where V describes the spin-independent part of the scattering potential, V_{so} is the spin-orbit interaction, and the last term describes the exchange interaction between the spin $\vec{s}$ of the incident electron and the spin $\vec{S}_j$ of the magnetic ion located at $\vec{R}_j$; $J(\vec{r} - \vec{R}_j)$ is the exchange coupling constant. The exchange scattering asymmetry A_{ex} is defined as the normalized difference of the scattered intensity for parallel ($I_{\uparrow\uparrow}$) and antiparallel ($I_{\uparrow\downarrow}$) spins:

$$A_{ex} = (1/|\vec{P}_0|)(I_{\uparrow\uparrow} - I_{\uparrow\downarrow})/(I_{\uparrow\uparrow} + I_{\uparrow\downarrow}).$$

The apparatus used for the present work is an ultrahigh-vacuum (UHV) multiple-chamber system which allows the growth of p-doped (with Be) $Al_xGa_{1-x}As$ by molecular-beam epitaxy (MBE) and its use *in situ* as a source of spin-polarized electrons.[12] The source used in these experiments is an $Al_{0.37}Ga_{0.63}As(100)$ layer grown on a GaAs(100) substrate. This compound was chosen to achieve high polarization, $P_0 = 44.1 \pm 1.3\%$, at the He-Ne laser line ($h\nu = 1.96$ eV). The MBE layer can be transferred after growth from the MBE chamber into the preparation chamber for Cs and O_2 deposition to achieve negative electron affinity. After activation the photocathode is transferred to the scattering chamber where it is irradiated with circularly polarized light from a He-Ne laser. As a $\lambda/4$ retarder a Pockels cell driven by a special 3-kV power supply[13] is used, so that the electron polarization can be switched between left and right circular polarization. The (longitudinal) spin polarization of the photoemitted electrons is therefore switched synchronously between $\pm\vec{P}_0$ with respect to $\vec{n}$. The photoelectrons are deflected electrostatically by 90° in order to

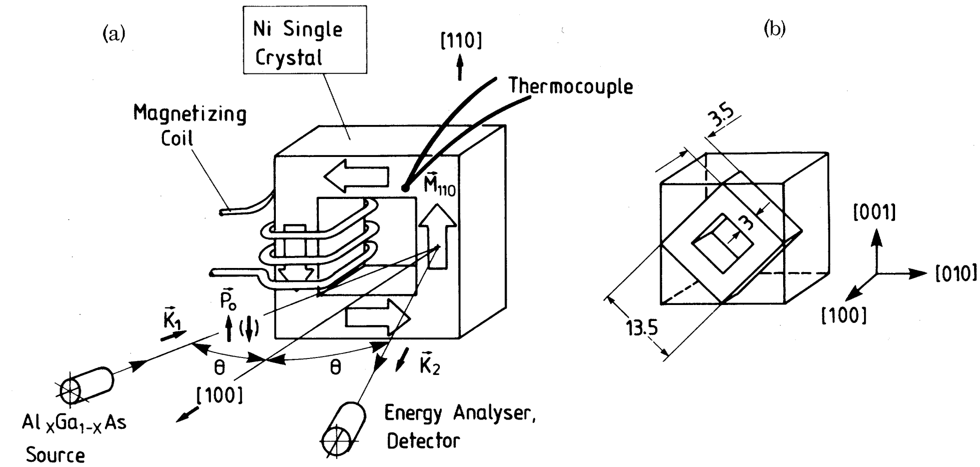

FIG. 1. (a) Scattering geometry of the Ni single-crystal window-frame target cut from a large single crystal. (b) Orientation of the picture-frame crystal with respect to the main crystallographic directions: dimensions in millimeters.

obtain a transversally polarized beam at the sample for the SPLEED experiment. The primary beam can be accelerated into a UHV chamber at 100 keV where the polarization is measured by Mott scattering from an Au foil. The surface of the Ni picture-frame single crystal is cleaned by prolonged Ar-ion bombardment at 550 °C in the Mumetal scattering chamber until no contamination can be detected by Auger spectroscopy performed with a hemispherical energy analyzer in the same chamber. The temperature of the Ni sample is stabilized by indirect pulse heating. The single crystal is magnetized after each heating pulse to account for demagnetization induced by the heating-current pulse. The scattered intensity is detected with the hemispherical energy analyzer (resolution ≤ 0.22 eV full width at half maximum). A desk computer, which also immediately calculates the scattering asymmetry, controls the measurement. The data collection is switched off during the heating and magnetizing current pulses.[14] The bulk Curie temperature T_{CB} of the Ni target is determined *in situ* by a method developed in our laboratory and based on the magneto-optical Kerr effect,[13] where the magnetic field is modulated. The laser employed is also used for optical alignment of the experimental setup. We have found that T_{CB} is equal to the surface Curie temperature T_{CS} (determined by electron scattering) to within ±4 K.

Experimentally we determine the normalized asymmetry

$$A^\sigma = (1/|\vec{P}_0|)(I_\uparrow^\sigma - I_\downarrow^\sigma)/(I_\uparrow^\sigma + I_\downarrow^\sigma),$$

where $I_\uparrow^\sigma$ ($I_\downarrow^\sigma$) is the scattered intensity measured for incident electron spin parallel (antiparallel) to the normal of the scattering plane and for the surface magnetized parallel ($\sigma = +$) or antiparallel ($\sigma = -$) to $\vec{n}$. It can be shown[11] that by using the relations $\tilde{A}_{ex} = (A^+ - A^-)/2 = A_{ex}$ and $\tilde{A}_{so} = (A^+ + A^-)/2 = A_{so}$ the magnetic and spin-orbit asymmetries can be determined separately. The equality $\tilde{A}_{ex} = A_{ex}$ (or $\tilde{A}_{so} = A_{so}$, respectively) is estimated to be exact to within δA_{so} (δA_{ex}) with δ of the order of $A_{ex}A_{so}$. δ can also be estimated experimentally and is generally found to be $|\delta| \ll 1\%$ at room temperature, so that $\delta A_{so} < 0.01 A_{ex}$ at all temperatures reported in this work. Fig. 2(a) shows experimental data of A_{ex} vs T/T_C. Here we are interested only in $\tilde{A}_{ex} = (A^+ - A^-)/2 = A_{ex}$, which is directly related to the magnetic scattering term. Recently Feder and Pleyer[11] have discussed the temperature range of validity of the relationship $A_{ex} \propto M(T)$ in the frame of dynamical LEED theory suggesting that the *linear* relationship between A_{ex} and the magnetization $M(T)$ is valid near the critical temperature. Since near the Curie temperature the magnetic coherence length $\xi_\perp$ in a direction perpendicular to the Ni(100) surface is much larger than the depth probed in this experiment the fast decrease of A_{ex} is attributed to the decrease of the magnetization at the topmost Ni atomic layers. This is why information about the critical exponent β_1 can be extracted by fitting the data of Fig. 2(a) by the function

$$A_{ex} \propto M_S(T) \propto (1 - T/T_{CS})^{\beta_1}.$$

This has been performed through a log-log plot [Fig. 2(b)], from which we obtain $\beta_1 = 0.825^{+0.025}_{-0.040}$ in the range $0.008 \leq 1 - T/T_C \leq 0.1$. The uncertainty in the determination of T_C from a given $A_{ex}(T)$ measurement is typically ± 1 K or better. We estimate correspondingly that the error in β_1 is about $+0.025$ and -0.040, respectively. From measurements performed at a different kinetic energy ($E_K = 37$ eV) we find $\beta_1 = 0.805^{+0.045}_{-0.085}$.

We compare in Table I the SPLEED result with results of various recent calculations. We notice that the experimental results are in the range of the best Monte Carlo results for semi-infinite systems.[1] At present it seems that the experiment favors results of the XY model, although such theoretical results might not be applicable in a simple way to the Ni(100) surface. In addition it is also not clear to what extent the Ni surface can be described near T_{CS} by a Heisenberg model. On the other hand in order to provide data allowing a more stringent test of current theories it might be necessary to do measurements even closer to the critical point T_{CS}—an experiment feasible in the near future—since in the present range of temperatures the reported values of β_1 might indeed be slightly reduced.[16] The experiment is therefore likely to yield a lower limit for the real β_1. Despite the current uncertainties in the comparison between theory and experiment it is surprising that the critical behavior of the magnetization of the top layers of Ni(100) agrees with results for the XY model. An XY behavior could, however, be understood by noticing that the magnetic dipole interaction between surface and bulk atoms is expected to be dependent on the orientation of their corresponding spins relative to the surface plane. This situation tends in any case to perturb the isotropy of the bulk (Heisenberg) interaction giving rise to

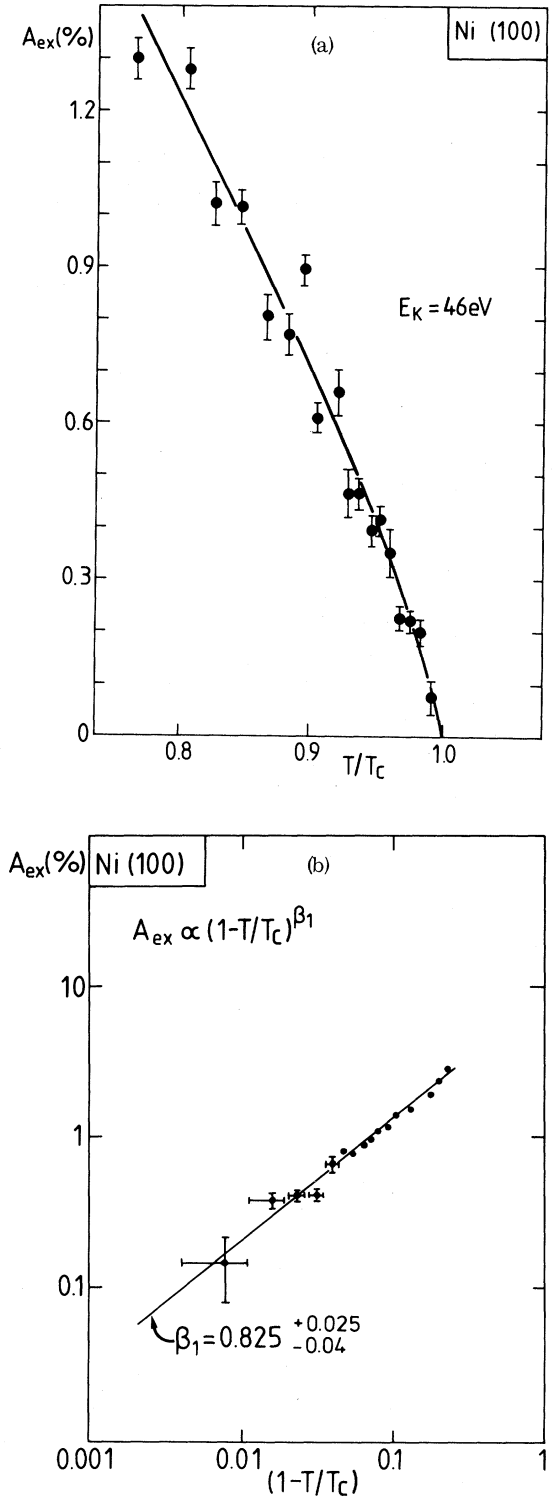

FIG. 2. (a) Exchange scattering asymmetry A_{ex} vs reduced temperature T/T_C. Angle of incidence of the electron beam: $\theta = 15°$ with respect to the surface normal. (b) Log-log plot of the exchange-scattering asymmetry vs reduced temperature.

TABLE I. Critical exponent β_1 for Ni(100) as obtained from SPLEED and compared to theoretical results determined using the scaling relation $\beta_1 + \gamma_1 = \beta + \gamma$ with the values of γ_1 given in Refs. 4 and 5. The values of β and γ used are those calculated by Le Guillou *et al.* (Ref. 15).

Experiment SPLEED	Theory Ising	Theory XY	Theory Heisenberg
$0.825^{+0.025}_{-0.04}$	0.78 [a]	0.8365 [b]	0.8782 [b]

[a] $\gamma_1 = 0.78$ (Ref. 5).
[b] γ_1 given by Eq. (6) of Ref. 4.

the XY anisotropy.[16] Other mechanisms which would also induce XY anisotropy at the surface are surface lattice relaxation, estimated to be less than 3% on Ni(100), coupled with changes in the interatomic exchange coupling strength, or crystal-field effects on the magnetic shells.

This work should also stimulate theoretical research with (mixed) systems where the bulk and the surface are allowed to exhibit different anisotropies, e.g., Heisenberg bulk and XY surface behavior.

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