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TEMPERATURE DEPENDENCE OF FERROMAGNETISM AT Ni(111) SURFACES DETERMINED WITH ELECTRON CAPTURE SPECTROSCOPY (ECS) *

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Electron capture spectroscopy has been used to probe the electron spin polarization (ESP) at the topmost atomic layer at Ni(111) surfaces. Contrary to bulk behavior, the ESP-signal measuring the long-range surface magnetic order decreases from −43% at $T = 293$ K almost *linearly* to zero at the bulk Curie temperature $T_{Cb} = 631$ K. "Local" surface magnetic order is temperature independent and is detected far above T_{Cb}. Such a behavior is only found for the 3d transition metal Ni.

Considerable theoretical work recently has been concerned with surface magnetic phase transitions [1–3]. Thus, there is interest in any available experimental information on the temperature dependence of surface magnetic order (SMO). Measurements performed over a wide range of temperatures T above and below the critical temperature would be highly desirable, as would be measurements very close to the critical temperature.

One of the reasons for the lack of sufficient experimental data on the T-dependence of the surface magnetic order for ferromagnetic materials is that it is difficult to prepare clean surfaces, which will remain uncontaminated and well-defined at temperatures far below and above T_{Cb} during multiple heating and cooling cycles. Such surfaces are necessary in order to obtain reliable data. The other reason is that there are not many experimental techniques which enable reliable data to be obtained over a wide range of temperatures. Surface effects can only be detected if the experimental probing depth is of the order of the spin–spin correlation length which amounts to about a lattice constant for tempeatures below and above T_{Cb}, while at T_{Cb} it diverges causing surface effects to extend deep into the bulk.

In this paper we report on the T-dependence of the long-range and "local" surface magnetic order for Ni(111). The surface magnetic order is measured with electron capture spectroscopy (ECS) at the topmost atomic layer of atomically clean and flat Ni(111) surfaces between 293 and 940 K. It is found that – contrary to the T-dependence of the bulk magnetization – the long-range surface magnetic order decreases almost *linearly* with increasing T until it drops to zero at the bulk Curie temperature T_{Cb} and remains zero above T_{Cb}. Nonzero "local" surface magnetic order, however, is found to exist still at temperatures far above $T_{Cb} = 631$ K.

The basic process in electron capture spectroscopy is the capture of one or two spin-polarized electrons during specular surface reflection of fast deuterons from a ferromagnetic target [4]. For an angle of incidence of 0.2°, the distance of closest approach of the deuterons to a surface is about 1–2 Å. Thus, the deuterons probe the spin-polarized local electron densities of state at the topmost surface layer.

The experimental set-up used in ECS is shown schematically in fig. 1. A well-collimated deuteron beam is incident at grazing angle on the target, which is magnetized parallel to the surface plane, and perpendicular to the plane defined by the incident and reflected beams, by use of a C-shaped electromagnet. Specularly reflected particles then enter a transverse electric field which spatially separates remaining D^+ from D^0 atoms and D^- ions formed by one- and two-electron capture, respectively. The ion currents are measured using two Faraday cups. The D^0 atoms, however, impinge on a tritium-target where their nuclear polarization is determined by measuring the asymmetry in the angular distribution of α-particles emitted in the reaction $T(d, n)\alpha$. A weak magnetic field, parallel to the target magnetizing field, provides a well defined quantization axis.

Long-range surface magnetic order is detected by means of one-electron capture processes. If the surface is ferromagnetic, one-electron capture results in the formation of deuterium atoms having a net electron spin polarization (ESP). This electron spin polarization is partially converted to a nuclear polarization by the hyperfine interaction. Determination of the resultant nuclear polarization provides a measure of the net electron spin polarizatin of the captured electrons. Defining the polarization, P, along the target magnetizing field yields $P = (n^+ - n^-)/(n^+ + n^-)$ where n^+ and n^- represent fractional numbers of electrons with spin moment antiparallel (majority spin electrons) and parallel (minority spin electrons), respectively, to the sample magnetizing field. Because the electrons captured by different deuterons originate at widely separated points on the target surface, long-range surface magnetic order is detected with one-electron capture.

The situation in two-electron capture is significantly different. The only stable state of D^- is the $1s^2\ ^1S$ state. D^- can therefore only be formed by capture of electrons with opposite spins. The characteristic length within which two-electron capture occurs is small, ≈

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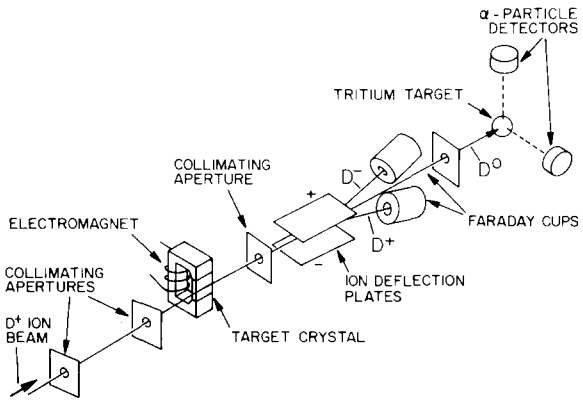

Fig. 1. Schematic diagram of the apparatus used in electron capture spectroscopy experiments.

10–20 Å. Thus the captured electrons must originate in the same "local" surface region, and two-electron capture will be strongly suppressed by the presence of "local" surface magnetic order. The reduction in the D^-/D^+ ratio in the reflected beam, relative to that for a nonmagnetic sample such as Cu, therefore, provides a measure of the "local" surface magnetic order. For further details we refer to refs. [4,5].

The experiments are performed under well defined conditions in the 10^{-10} mbar region, and numerous experimental difficulties were overcome in order to maintain clean surfaces during multiple heating and cooling cycles. At all temperatures C, S and O coverages are less than 0.02 monolayer using a cylindrical mirror analyzer for Auger analysis.

The upper part of fig. 2 gives ESP results indicating the long-range and "local" surface magnetic order at Ni(111) surfaces as function of the reduced temperature T/T_{Cb}. At 293 K ($T/T_{Cb} = 0.5$) the ESP amounts to -43% and decreases almost *linearly* with increasing T until it drops to zero at T_{Cb}. This behavior of the long-range surface magnetic order is drastically different from the magnetization of bulk Ni. The lower part of fig. 2 shows a plot of the T-dependence of the magnetization of layers parallel to the (111) surface of a fcc ferromagnet in the mean-field model [6–8]. The ESP-signal for the long-range surface magnetic order measured with electron capture spectroscopy at the topmost surface layer follows closely the T behavior of the magnetization for the topmost surface layer, as predicted by the mean-field theory, which is expected to be qualitatively correct [8] when the spin-spin correlation length is not large compared with the lattice spacing (valid for $T/T_{Cb} < 0.9$).

The T-dependence of the "local" surface magnetic order at Ni(111) surfaces shows significantly different behavior. The magnitude of the "local" ESP [5,10] (see (▲) in fig. 2) – being close to the long-range ESP measured at 293 K – amounts to about 47% and remains constant until the highest measured temperature of 940 K, which is far above T_{Cb}. This implies that exchange effects between minority and majority spin electrons from nearby atoms are temperature independent [5]. We remark that this behavior of the "local" surface magnetic order is only found for the 3d transition metal Ni. ECS studiese at surfaces of the 4f rare earth metal Tb reveal a somewhat different T-dependence of the "local" surface magnetic order, which decreases quite rapidly with temperature above the critical temperature and finally disappears at 80 K above T_{Cb} [11].

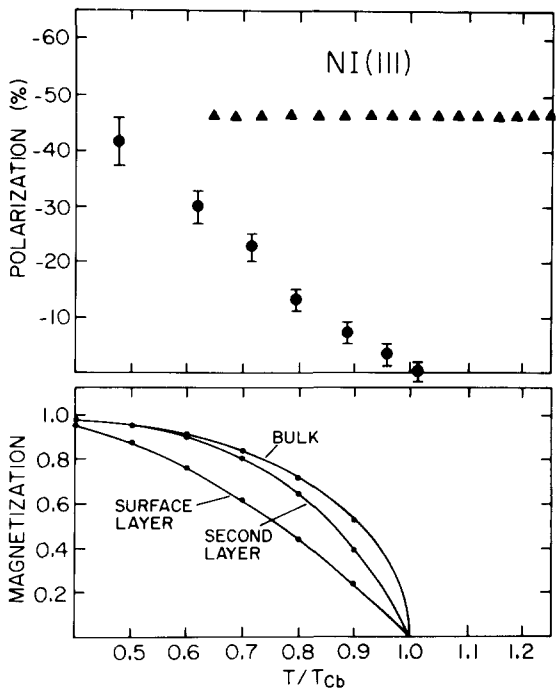

Fig. 2. Upper part: T-dependence of the ESP-signal originating from long-range (●) and from "local" (▲) surface magnetic order on Ni(111). Lower part: T-dependence of the magnetization of the first and second layer and the bulk in mean-field theory [6].

More refined electron capture spectroscopy measurements on Ni(111) surfaces, especially measurements very close to T_{Cb}, are underway to determine whether the surface Curie temperature is possibly slightly different from T_{Cb}, which is of great importance for the reliable evaluation of surface critical exponents. This includes investigations on the magnetic field dependence of the electron-spin polarization around T_{Cb} which is important for testing scaling laws in surface magnetism [3].

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