ELECTRON SPIN POLARIZATION (ESP) ON SURFACES AND THIN FILMS OF FERROMAGNETIC METALS *

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The ESP at surfaces of oligatomic epitaxial Ni(100)-layers prepared on Cu(100) and NaCl(100) substrate crystals is determined by electron capture spectroscopy (ESC). No evidence is found for the existence of "intrinsically magnetic dead layers" at Ni(100) surfaces. ECS measurements at surfaces of antiferromagnetic bulk Cr show that ferromagnetic order exists at 293 K ($T_N = 310$ K) at the topmost atomic layer of the Cr(100)c(2×2) surface.

1. Introduction

The surface magnetic structure of bulk and thin film ferro- and antiferromagnets is determined by the ratio of electron densities for spin-up and spin-down. These densities depend on the crystallographic directions in real space, a fact which directly suggests experiments where the surface orientation is well-known. Experiments of this kind certainly simplify the comparison with theoretical data on spin-polarized electron densities which in modern theories also exhibit a crystallographic dependence. In the past years few much experimental and theoretical work has been centered upon the electronic and magnetic properties of oligatomic films of ferromagnetic materials such as Ni, Co and Fe [1]. In particular experiments by Liebermann and co-workers [2] stimulated the discussions on magnetic phenomena in thin films. These authors measured at room temperature the magnetization of ultrathin electrolytically prepared polycrystalline Ni, Co and Fe films as function of film thickness and concluded on the basis of their experimental results that "intrinsically magnetic dead layers" exist at surfaces of 3d-transition metals. For Ni, e.g., they found that up to a film thickness of 4 atomic layers the magnetization at room temperature remains zero. With increasing layer thickness this "dead layer" effect did not disappear. Using different substrate material (Cu, Au) they could exclude the possibility that these dead layers could be introduced by diffusion of substrate atoms into the ferromagnetic films. Based on experimental facts [3-5] Gradmann [6] pointed out that this effect may be induced by H-chemisorption during the electrolytical preparation of these films and may be not present at clean surfaces of these metals. Therefore magnetic measurements performed under UHV conditions at atomically clean and well-defined (single-crystalline) surfaces are very desirable [6].

Here we report on electron spin polarization (ESP) measurements at oligatomic (2-64 layers) epitaxial Ni(100) films prepared by electron beam evaporation under UHV conditions on Cu(100) and NaCl(100) substrate crystals.

Further we report on ECS measurements on the surface magnetic structure existing at the topmost atomic layer of a bulk antiferromagnetic Cr(100) single crystal.

2. Experimental

The basic process in ECS is the capture of one [one-electron capture (OEC)] or two [two-electron capture (TEC)] spin-polarized electrons during small-angle scattering of 150 keV deuterons at single crystalline surfaces of magnetic crystals. The distance of closest approach of the fast ions during the grazing-angle surface reflection amounts to about 0.2 nm showing that the ions probe in real space only the exponential tail of the electronic wave functions at the surface, a fact which reveals the extreme surface sensitivity of ECS. With OEC long-range ferromagnetic order at surfaces can be detected. Further details of ECS are described in refs. [7–9]. For the measurements the single crystals are magnetized to saturation parallel to the surface plane. Defining the ESP along the magnetizing field we obtain

$$P = (n^{+} - n^{-})/(n^{+} + n^{-}), \qquad (1)$$

where n^+ and n^- represent fractional numbers of electrons with spin moment antiparallel and parallel, respectively, to the target magnetizing field which is applied to align randomly oriented Weiss domains thereby producing a macroscopic magnetization which defines a preferred direction in space along which the sign and magnitude of P can be measured. The measurements were performed under UHV conditions in a vacuum below 3×10^{-10} mbar. During the evaporation of the oligatomic Ni films the working pressure is kept in the 10^{-10} mbar region to avoid undesired contamination of the films. The substrate crystals are prepared with a

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surface orientation better than 0.01° . After several standard cleaning-annealing procedures the final C and O contaminations are less than 0.02 monolayer as monitored *in situ* with AES using a cylindrical mirror analyser. The monocrystalline state of the substrate surfaces is detected by LEED. In case of Cr(100) a $c(2 \times 2)$ surface structure is found.

Oligatomic (2-64 layers) Ni(100) films are prepared by electron beam evaporation on Cu(100) at room temperature and on NaCl(100) at 200°C. The absence of holes in these oligatomic films is carefully checked by measuring AES signals as function of layer thickness [10,11] and the monocrystalline state of the film surfaces is detected by LEED.

3. Results and discussion

Fig. 1 gives ECS results using OEC at surfaces of thin Ni(100) layers. The measurements are performed at room temperature. The ESP at the surface of two atomic layers Ni(100) on Cu(100) amounts to -19% clearly excluding the existence of so-called "intrinsic magnetic dead layers" at Ni(100) surfaces. Increasing the thickness of the Ni(100) layers on Cu(100) up to 64 layers increases the ESP up to -65% close to the ESP value (-64%; see fig. 1) measured at surfaces of bulk Ni(100). We remark that the decrease of the ESP with decreasing Ni-layer thickness cannot be attributed to a decrease of the bulk Curie temperature T_{Cb} because it is known from reliable UHV experiments [12,13] that T_{Cb} does not depend on layer thickness at least down to a layer thickness of 2 nm (6 atomic layers).

In a further experiment 7 atomic layers Ni(100) are evaporated on NaCl(100) substrate crystals. The ESP at these surfaces amount to -64% (see fig. 1) and is not reduced compared to the Ni(100)/Cu(100) experiment where, e.g., 8 monolayers Ni(100) on Cu(100) yield an ESP of -33%. This suggests that the observed reduction of the ESP at the free Ni(100) surface using Cu(100) substrate crystals might be caused by the influence of



Fig. 1. ESP of *n* monolayers Ni(100) on Cu(100) substrate (\Box) (*d* = Ni-film thickness in Å).

non-polarized Cu-substrate electrons.

Unfortunately, at present, there exist no detailed calculations on the ESP at surfaces of thin Ni(100) layers on Cu(100) and NaCl(100) substrate crystals as a function of Ni-layer thickness. We note, however, that a self-consistent calculation on the magnetic structure of a 9 layer thick *unsupported* Ni(100) film is available [14]. The ESP at the surface of this film amounts to -57% which is close to the ESP at the surface of 7 atomic layers Ni(100) on NaCl(100) which amounts to -64%. These authors note that for films of thickness less than 7 layers there is a dependence of the ESP on layer thickness also for unsupported films.

Quite recently Freeman and coworkers [15] investigated the surface magnetism of one Ni overlayer on a five layer Cu(100) substrate. They find a reduction of the surface-ESP of Fermi electrons caused by the Cusubstrate which is in agreement with the experimental ECS results.

In a further self-consistent calculation Jepsen et al. [16] investigated the spin-polarized electronic structure of unsupported thin Ni(100) films. Graphical evaluation of the ESP for Fermi electrons yields for the ESP at the surface of a 5-layer thick Ni(100) film an ESP of -75%. We note that also from these self-consistent calculations no evidence is found for magnetic dead surface layers at Ni(100) surfaces.

In case of Cr(100) the ESP is investigated at 2×10^{-11} mbar. Cr is antiferromagnetic below the bulk Néel temperature $T_{\rm Nb} = 310$ K and consists of two magnetically compensating ferromagnetic sublattices. In the (100)-direction the spin structure is sinusoidal and incommensurate with the lattice period. Therefore one might already expect ferromagnetic order at the unreconstructed Cr(100) surface below $T_{\rm Nb}$.

For applied magnetic fields up to 0.09 T an ESP up to -(18+2)% is found at T = 293 K which clearly reveals the existence of long-range ferromagnetic order at the topmost atomic layer of $Cr(100)c(2 \times 2)$ below T_{Nb} [17].

At present there is one calculation available where within a simple tight-binding approximation it is shown that already the nonreconstructed Cr(100) surface is ferromagnetic [18] which is in basic agreement with the ECS experiment.

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