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Layer-dependent magnetization at the surface of a band ferromagnet

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The temperature dependence of the magnetization near the surface of a band ferromagnet is measured with monolayer resolution. The simultaneous application of highly surface-sensitive techniques enables one to deduce the layer-dependent magnetization curves at a Fe(100) surface. Analysis of data is based on a simple mean-field approach. Implications for modern theories of itinerant-electron ferromagnetism are discussed.

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A ferromagnetic material is characterized by a spontaneous magnetization m which decreases with increasing temperature T until the paramagnetic state with $m=0$ is reached at the Curie point T_c. For very low temperatures the form of the magnetization curve $m(T)$ is governed by spin-wave excitations according to Bloch's law. For temperatures very close to T_c critical fluctuations result in a power-law dependence $m(T) \propto (T_c - T)^{\beta}$ with a critical exponent β. In the wide range of intermediate temperatures the form of $m(T)$ depends on the specific system. In case of a band-ferromagnetic material, such as Fe as a prototype, the detailed form of $m(T)$ in this intermediate regime must be explained from the underlying electronic structure.[1,2]

Density-functional theory within the local-spin-density approximation is known to give a quantitatively accurate description of several ground-state properties.[3] For finite temperatures, however, there is no satisfying implementation available. A microscopic theory must account for the existence of local magnetic moments above T_c in particular.[4] This requires one to deal with correlations among itinerant valence electrons as, for example, within the framework of an orbitally degenerate Hubbard-type model with realistic parameters.[5] The long history of itinerant-electron ferromagnetism shows that this is a demanding task.[2] On the other hand, comparatively simple mean-field approaches based on spin models are known to provide a successful phenomenological description in many cases (see, e.g., Ref. 6). Remarkably, while the Weiss mean-field theory fails to reproduce the known $T \to 0$ and $T \to T_c$ limits and substantially overestimates T_c, the form of the Fe magnetization curve $m(T)$ at intermediate reduced temperatures T/T_c is reasonably well described: For spin-quantum number $S=1/2$ there are deviations from the measured bulk magnetization curve of Fe within a few percent only.[7,8]

At the *surface* of a band ferromagnet the magnetization may be different for different layers α parallel to the surface because of the reduced translational symmetry. Hence, a key quantity that characterizes the surface magnetic structure is the layer-dependent magnetization curve $m_\alpha(T)$. Within the framework of classical spin models, the lowered surface coordination number implies that certain exchange interactions are missing. This directly leads to a reduced magnetic stability at the surface:[9] The top-layer ($\alpha=1$) magnetization is substantially reduced as compared with the bulk. However, significant deviations from the bulk magnetization curve are confined to the first few layers in the intermediate temperature range. This is confirmed qualitatively by calculations within Hubbard-type models of correlated itinerant electrons using slave-boson, decoupling and alloy-analogy approaches.[10] Roughly, the results are similar to those for Ising or Heisenberg systems in the range of intermediate temperatures. Yet, the precise form of $m_\alpha(T)$ for a band-ferromagnetic surface must still be considered as largely unknown.

On the experimental side, determination of the layer-dependent magnetization curve at a surface is a demanding task as well, which has not been achieved so far. In order to measure $m_\alpha(T)$, experimental techniques sensitive to surface magnetism are required with a magnetic probing depth tunable with monolayer (ML) resolution. Common surface-sensitive techniques like spin-resolved secondary-electron emission[11] or (inverse) photoemission[12] average over several layers beneath the surface resulting in a (nearly) bulklike behavior of $m(T)$. Nevertheless, in a number of sophisticated experiments a roughly linear temperature trend of $m(T)$ has been observed and attributed to the surface magnetization (see Refs. 13–15 for Fe surfaces and Ref. 16 for experimental techniques).

Here we report on an experiment to determine $m_\alpha(T)$ at the (100) surface of bcc Fe. The crucial feature of our experiment is the simultaneous application of different *in situ* techniques which are highly sensitive to the magnetization near the surface, but slightly differ in their magnetic probing depths.

Ultimate surface sensitivity (magnetic probing depth $\lambda = 0$ ML) is achieved by spin-polarized electron capture.[17,18] 25 keV He^+ ions are grazingly scattered (incidence angle to the surface plane 1–2°) off a magnetized Fe(100) surface. The ions are reflected and capture target electrons into excited atomic states. The spin polarization of captured electrons is deduced from the observed degree of circular polarization of emitted fluorescence light. Excited atomic states can only survive collisions for impact parameters exceeding the mean radius of the corresponding electronic orbital. Thus the final formation of atomic states takes place on the outgoing part of the trajectories, resulting in a sensitivity of electron capture to a region at or above the top surface layer.

An established technique to study magnetism near a surface is spin-polarized secondary-electron emission, induced by keV electrons at normal or oblique incidence.[19] Based on a mean-field study, Abraham and Hopster[11] infer from their observed temperature-dependence of the spin-polarization of secondary electrons from Ni(110) a magnetic probing depth of $\lambda = 3-4$ ML (with an upper limit of 7 ML) for electrons

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of about 0 or 10 eV kinetic energy. A compilation of electron scattering cross sections by Schönhense and Siegmann[20] suggests a similar value for Fe ($\lambda = 4.2$ ML), in accordance with a recent overlayer experiment by Pfandzelter *et al.*[21]

The probing depth in secondary-electron emission can be considerably reduced by using energetic ions instead of electrons as primary particles.[22,21] Grazingly incident ions are reflected from the top surface layer and do not penetrate into the bulk (''surface channeling''). In practice, structural imperfections like surface steps mediate penetration of some projectiles, leading to a contribution of excited electrons from layers beneath the surface. From computer simulations emulating ion trajectories[23] and an overlayer experiment,[21] we infer for scattering of 25 keV protons from our Fe(100) surface a probing depth of $\lambda = (0.5 \pm 0.2)$ ML for electrons of 10–20 eV kinetic energy. We note that λ seems to increase for lower electron energies owing to cascade multiplication governed by electron-electron scattering.[21] Hence, energy resolution is mandatory if maximum surface sensitivity is aspired.

Electron capture and electron emission yield information on the spin part of the magnetization. Although a general quantitative relationship between experimental observable (electron spin polarization) and magnetization has not been worked out so far, it is generally assumed that one can derive the (normalized) temperature dependence of the magnetization. This assumption appears to be justified in view of the, at least for the conditions of our experiment, weak selectivity of capture and emission processes in **k** space. Considering the small, well-defined, but different information depths of the techniques, a simultaneous application at the same surface thus should enable one to deduce the layer-dependent magnetization curves near the surface.

In our experiment, a (100) Fe single crystal disk is mounted to close the gap of a magnetic yoke with a coil. For the measurements the crystal is magnetized by current pulses through the coil along an easy axis of magnetization $[001]$ or $[00\bar{1}]$ in the (100) surface plane. This reproducibly yields a full remanent magnetization near the center of the crystal as checked by the magneto-optic Kerr effect. The (100) surface is prepared by cycles of grazing Ar^+ sputtering and annealing, until the surface is clean, atomically flat, and well ordered, as inferred from Auger electron spectroscopy, grazing ion scattering, and LEED. The target temperature is controlled by a thermocouple attached directly near the crystal. Systematic differences between the thermocouple reading and the crystal temperature are calibrated by Kerr effect (Curie temperature T_c) and pyrometer measurements and corrected.

Electrons are emitted by 25 keV protons at grazing incidence (1.2°) or by 4 keV electrons at oblique incidence (33°) and enter an electrostatic energy analyzer (cylindrical sector field) in a direction of about 10° from normal. Spin analysis is performed for electrons with 10–20 eV kinetic energy in a subsequent LEED spin polarization detector.[21] Each polarization spectrum is obtained from two identical measurements with reversed magnetizations to eliminate instrumental asymmetries.

Electron capture into the excited He I $1s3p^3P$ term

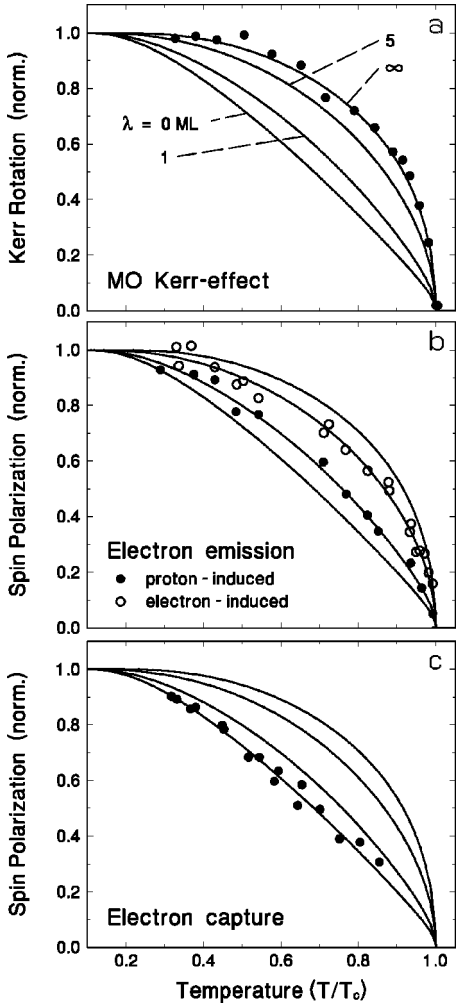

FIG. 1. Temperature dependence of (a) Kerr rotation, (b) spin polarization of secondary electrons, induced by electrons and grazingly scattered protons (open and solid circles, respectively), and (c) spin polarization of electrons captured into atomic states of He^+ ions, grazingly scattered off Fe(100). The curves represent mean-field calculations according to Eqs. (1) and (2) with different probing depths λ as indicated.

formed during grazing scattering of 25 keV He^+ ions is studied via fluorescence light emitted in the $2s^3S$-$3p^3P$, $\lambda = 388.9$ nm transition. The circular polarization fraction of the light is measured by means of a rotatable quarter-wave plate, a linear polarizer, a narrow bandwidth interference filter, and a cooled photomultiplier. The transition being in the UV spectral range, detection is affected on a tolerable level by stray light from the filaments for heating the crystal up to temperatures below about 900 K.

Experimental results are depicted in Fig. 1 as a function of the reduced temperature T/T_c, $T_c = 1043$ K. The data are collected from several quick heating and cooling runs. Short measurement times turned out to be important in order to avoid significant segregation of C at intermediate temperatures and S at temperatures close to T_c.[15] No systematic differences exceeding the statistical error (about ± 0.03 for the normalized polarization) were observed for heating and cooling runs, respectively. In Fig. 1(a) we show for compari-

son the rotation of the polarization axis associated with the longitudinal magneto-optic Kerr effect (solid circles). The temperature dependence is in agreement with previous Kerr-rotation measurements at Fe(100) by Sirotti *et al.*[15] and reflects the bulk magnetization, because of the large penetration depth of visible He-Ne-laser light [typically 20 nm (Ref. 24)].

Clearly different temperature-dependences are observed with the surface-sensitive techniques electron-induced electron emission [Fig. 1(b), open circles], proton-induced electron emission [Fig. 1(b), solid circles], and electron capture [Fig. 1(c), solid circles]. The curvatures gradually decrease, until, for electron capture, an almost linear behavior is observed. Remarkably, for electron-induced electron emission, a prominent technique to study surface magnetism, the data closely resemble the data from the Kerr effect.

Information on the layer-dependent magnetization cannot be extracted from the data directly, as these have to be interpreted as exponentially weighted averages over a number of layers corresponding to the probing depth. We use an indirect way by comparing with results of a simple mean-field calculation which is known to reproduce the bulk magnetization curve fairly well, provided that reduced quantities $m(T)/m(0)$ and T/T_c are used.

Accordingly, the spin-S Heisenberg model for the (100) surface of a bcc lattice with layer-independent nearest-neighbor exchange J is considered: $H=-J\Sigma_{\langle i\alpha,j\beta\rangle}\mathbf{S}_{i\alpha}\mathbf{S}_{j\beta}$. Here i labels the sites within a layer parallel to the surface and $\alpha=1,\ldots,\infty$ the different layers. The mean-field free energy is $F_{\mathrm{MF}}=-k_{\mathrm{B}}T\ln\mathrm{tr}\exp(-H_{\mathrm{MF}}/k_{\mathrm{B}}T)$ where H_{MF} is obtained from H by the usual decoupling $\mathbf{S}_{i\alpha}\mathbf{S}_{j\beta}\mapsto\langle\mathbf{S}_{i\alpha}\rangle\mathbf{S}_{j\beta}+\mathbf{S}_{i\alpha}\langle\mathbf{S}_{j\beta}\rangle-\langle\mathbf{S}_{i\alpha}\mathbf{S}_{j\beta}\rangle$. Assuming collinear ferromagnetic order, $\mathbf{m}_\alpha=m_\alpha\mathbf{e}_z$, and minimizing F_{MF} with respect to the order parameter, $m_\alpha=g\mu_{\mathrm{B}}\langle S^z_{i\alpha}\rangle$ (g: Landé factor; μ_{B}: Bohr magneton), yields a coupled set of Weiss self-consistency equations

$$m_\alpha=m_\alpha(0)B_S(Sb_\alpha/k_{\mathrm{B}}T), \tag{1}$$

with $m_\alpha(0)=g\mu_{\mathrm{B}}S$, the Brillouin function B_S (Ref. 25) and the layer-dependent Weiss field $b_\alpha=(2J/g\mu_{\mathrm{B}})(z_\parallel m_\alpha+z_\perp m_{\alpha-1}+z_\perp m_{\alpha+1})$. $z_\parallel=0$ and $z_\perp=4$ are the intra- and inter-layer coordination numbers for the (100) surface. The total coordination number is $z=z_\parallel+2z_\perp=8$. The equations (1) are easily solved numerically for a film of finite but sufficiently large thickness. For the actual calculations we have taken $S=1$. Assuming the orbital contribution to the magnetic moment to be quenched completely ($g=2$), this appears to be the proper choice in the case of Fe since the $T=0$ spin moment is $2.13\,\mu_{\mathrm{B}}$ per atom.[26]

To compare with the experiment we assume that each layer α gives a contribution proportional to m_α but weighted by an exponential factor $\exp(-\alpha/\lambda)$ where λ is the probing depth characteristic for the experimental technique applied. From the layer-dependent magnetization curves $m_\alpha(T)$, we thus calculate a quantity $P(T)$ as

$$P(T)\propto\sum_{\alpha=1}^{\infty}e^{-\alpha/\lambda}m_\alpha(T). \tag{2}$$

The results are shown in Fig. 1 (curves). The mean-field $P(T)$ nicely reproduces the temperature trend of the (properly normalized) measured data for the respective information depth λ. A value $\lambda=5$ ML is consistent with the estimates for the probing depth in electron-induced electron emission ($\lambda=4$–5 ML). We have also checked against the choice $S=1/2$. This does not change the temperature trend of $m_\alpha/m_\alpha(0)$ as a function of T/T_c significantly. Surprisingly, considering an enhancement of the $T=0$ top-layer magnetic moment (see Ref. 27) does not lead to a significant change of the temperature trend either. Following Ref. 28 one may expect a different exchange between the top- and the subsurface-layer moments: $J_{12}\neq J$. Within the experimental error, we find that the measured data are reproduced by calculations for a modified surface exchange in the range from $J_{12}/J=0.8$ to $J_{12}/J=1.1$.

We conclude that the mean-field calculation gives a rather accurate description of the layer-dependent magnetization at the Fe(100) surface at intermediate temperatures. Clearly, mean-field theory must be considered as a poor starting point to explain surface magnetism. Nevertheless the result is interesting as any theoretical approach that conceptually improves upon the Weiss theory should give the same results (within our experimental error).

In summary, this study gives detailed information on the layer-dependent magnetization at the surface of a prototypical band ferromagnet. We report on an experiment to measure temperature-dependent magnetization curves near the (100) surface of bcc Fe. We simultaneously apply different techniques, two of which are based on grazing scattering of energetic ions, resulting in an ultimate surface sensitivity. The magnetic information depths of the techniques being well defined but slightly different enables one to achieve a near monolayer resolution. The form of the layer-dependent magnetization curve is an important key quantity of surface magnetism which, for intermediate temperatures, represents a benchmark to discriminate between different microscopic theoretical approaches to explain surface magnetism from the underlying temperature-dependent electronic structure.

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