Magnetic ordering of ultrathin Mn films on Fe(100) studied via capture of polarized electrons by fast ions

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The magnetic ordering of the topmost surface layer of Mn films grown on Fe(100) at different temperatures is investigated via the polarization of fluorescence light emitted after capture of spin-polarized electrons into excited atomic terms during scattering of 25-keV He^+ ions. For coverages up to about 0.5 ML the observed spin polarization is in accordance with in-plane ferromagnetic order of Mn moments with antiparallel coupling to moments of the Fe substrate. For 1–8-ML films the detected spin polarization vanishes, indicating in-plane antiferromagnetic order of the topmost layer. [S0163-1829(98)03730-8]

Considerable attention has been devoted to films of nonferromagnetic metals on a ferromagnetic substrate owing to their importance in understanding exchange-coupling phenomena in sandwich and multilayer systems.[1] Cases of current interest are Cr and Mn films on Fe(100), where the film material shows a tendency to antiferromagnetic ordering. Whereas the understanding of growth and structure of Mn films on Fe(100) has improved,[2,3] the magnetic ordering of the films is a matter of dispute, both theoretically[4–8] as well as experimentally.[9–14]

Various configurations of the magnetic moments of Mn atoms have been discussed, comprising parallel and antiparallel coupling *within* as well as *between* the film layers. Most experimental techniques, sensitive to a magnetization **M** at a surface, average over a macroscopic region *within* the surface and a number of layers *beneath* the surface. Though it is possible to investigate a ferromagnetic ordering, one cannot distinguish between different antiferromagnetic configurations, since magnetic moments average to $\mathbf{M}=0$.

Here we report on experimental studies where we have analyzed magnetic ordering during growth of a thin metal film via capture of spin-polarized electrons into excited atomic states of fast atoms during scattering from the film surface. The spin polarization is deduced from the polarized light emitted in the subsequent decay of those states.[15–18] This method can be considered as an attractive alternative to the technique to deduce the spin polarization from a complicated analysis of a nuclear reaction.[19]

Similar to other methods to probe surface magnetism, a quantitative relation between surface magnetization **M** and spin polarization of captured electrons P_S has not been established so far. A theoretical treatment for the formation of excited terms during grazing scattering from a target with a realistic spin-resolved band structure has still to be worked out. However, from simple concepts of charge exchange[20–23] some important conclusions for the application of our method can be drawn.

(1) Since electronic orbitals of excited atomic states have typical mean radii >10 a.u., these states can only survive from collisions where the atom core has reached a distance from the surface of about those radii on the outgoing trajectory. From this feature we conclude a sensitivity of our method to a region above the topmost layer of surface atoms. This extreme surface sensitivity makes electron capture a powerful technique to study magnetic ordering in thin films.[24]

(2) Some qualitative statements on the relation between magnetic moments and P_S can be given: P_S reflects the magnetization owing to an orientation of spin momenta, because the spin is assumed to be conserved in the capture process. The spins are collinear with **M**. P_S changes sign with **M**. As a consequence, it should be possible for thin films to discriminate between a parallel and antiparallel stacking of layers. Moreover, a distinction of parallel and antiparallel alignments *within* the surface plane becomes feasible.

The concepts and analysis of experiments on polarized light emission after electron capture are described in detail in Ref. 18. We refer to a geometry where the scattering plane is the xy plane, and light is emitted along the positive z direction. During interaction with the film surface, the ions capture electrons into excited atomic $|LSM_LM_S\rangle$ states (LS coupling). For orbital angular momenta $L\neq 0$ the broken cylindrical symmetry in the excitation geometry may lead to anisotropic populations of states with magnetic sublevels M_L, resulting in an anisotropy of orbital angular momenta $\langle L_z\rangle \neq 0$. In case the film surface shows a long-range magnetic order with magnetization $M_z\neq 0$, electrons with a net spin polarization are captured, and the electronic atomic states have $\langle S_z\rangle \neq 0$. In order to obtain the spin polarization $P_S=\langle S_z\rangle/S$ and the polarization of orbital angular momenta $P_L=\langle L_z\rangle/L$, two independent measurements of the polarization of fluorescence light are needed. This is accomplished by inverting the sign of P_S by a reversal of the magnetization,[15,18] which does not affect P_L.

In addition to the light emission, the intensity of specularly reflected ions is also measured. For layer-by-layer growth, the intensity varies with the coverage in an oscillatory way, similar to oscillations in reflection high-energy electron diffraction. This allows one to continuously monitor coverage and morphology of the film.[25–27]

In our experiments a well-collimated beam of 25-keV He^+ ions is directed onto the target surface at a polar incidence angle $\Phi\approx 1°-2°$ with respect to the surface plane. As substrate we use an Fe(100) single-crystal disk, instead of making use of a thin epitaxial Fe film, in order to prepare a

smooth substrate surface largely free of defects and to avoid complications owing to segregation of foreign atoms, which has been assumed to explain contradictory experimental results.[12,11] *In situ* preparation of the Fe surface is performed by cycles of grazing Ar^+ sputtering and subsequent annealing. Mn is deposited from high-purity rods by electron-beam heating in a commercially available evaporator (EFM3, Omicron) with integrated flux monitor, using typical growth rates of some 10^{-3} ML s^{-1}. Growth is monitored by recording the intensity of specularly reflected ions by means of a channeltron detector. Before and after deposition, Auger electron spectroscopy, spot-profile analysis of low-energy electron diffraction, and analysis of polar angular distributions of scattered He projectiles[28] show a clean and well-ordered surface.

The Fe crystal is mounted into the gap of a toroidal yoke with a coil of 50 windings. Magnetization of the crystal is performed along an easy axis of magnetization [001] or [001] in the (100) plane by current pulses. Via these pulses a reliable remanent magnetization is achieved, as checked by the longitudinal magneto-optic Kerr effect. A zero net magnetization is obtained by a gradual decrease of an ac current through the coil.

The light of the He I $1s2s\ ^3S$-$1s3p\ ^3P$ transition at $\lambda = 3889$ Å, which is in the UV spectral range, so that the detection is hardly affected by blackbody radiation from the filament for heating the crystal, is imaged 1:1 onto the cathode of a UV photomultiplier. The circular polarization fraction of the light is measured by means of a quarter-wave plate, a linear polarizer, and a narrow bandwidth interference filter via a rotation of the quarter-wave plate in intervals of 90°. We also replaced the linear polarizer by a polarizing cube beam splitter and measured with two photomultipiers the light intensities for opposite helicities simultaneously. The latter setup enhances count rate and stability of data, yet reduces the absolute accuracy of P_S and P_L owing to different efficiencies of both detection channels.

The overall quantum efficiency of our optical setup amounts to about 3×10^{-4} for the single-channel and 6×10^{-4} for the two-channel system. Typical count rates amount to 1500 and 4000 s^{-1}, respectively, for a current density of 20 nA mm^{-2}, which is low enough to avoid ion-beam-induced sputtering or damage of the film, as has been checked from Mn/Fe Auger signal ratios and angular distributions of scattered He projectiles.

In recent studies we found that growth of Mn on Fe(100) starts in a layer-by-layer mode[3] with a favorable growth temperature of about 560 K. Lower temperatures lead to kinetic roughening. After four layers, the growth mode changes from layer to island growth for temperatures above about 420 K. Below this temperature a (metastable) layer-by-layer growth is observed. In accordance with growth conditions used in published studies on Mn/Fe(100), we first show results for Mn films grown at 300 K (Fig. 1). The intensity of specularly reflected ions oscillates with coverage, which is the signature of layer-by-layer growth [Fig. 1(a), open squares]. The intensity of emitted light (sum of the light intensities for opposite helicities) shows the opposite behavior [Fig. 1(a), solid circles]. It increases first and oscillates at a higher level. The oscillations show a phase shift of 0.5 ML, compared to the intensity oscillations of reflected ions, i.e.,

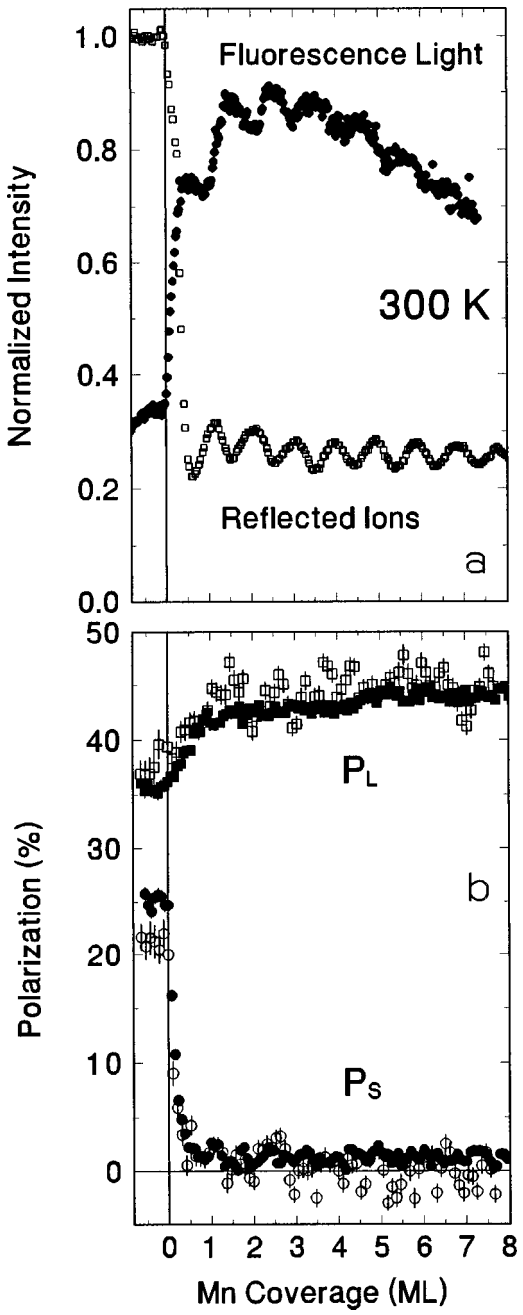

FIG. 1. (a) Normalized intensity of fluorescence light emitted in the He I $1s2s\ ^3S$-$1s3p\ ^3P$, $\lambda = 3889$ Å transition (solid circles) after grazing scattering of 25-keV He^+ ions from Mn films grown on Fe(100) and normalized intensity of specularly reflected 25-keV H^+ ions (open squares) [we note that there is no fundamental difference between scattering of He^+ and H^+ ions (Ref. 27)]. Incidence angle $\Phi = 1.75°$, growth temperature $T = 300$ K. (b) Spin polarization P_S (circles) and polarization of orbital angular momenta P_L (squares) obtained from the fraction of circular polarization in the fluorescence light measured with the single-channel (open symbols) and the two-channel (solid symbols) system, respectively. Each data point represents the average of five single measurements for sequentially reversed magnetizations. The total accumulation time per data point is 30 s, corresponding to about 1.2×10^5 counts. Note that the data have been accumulated continuously without interrupting the Mn flux. The left vertical line indicates the opening of the shutter.

the light intensity *increases* with the surface roughness and has maxima whenever the surface layer is about half filled.

The spin polarization P_S and the polarization of orbital angular momenta P_L are shown in Fig. 1(b). The open and solid symbols represent data measured with the single- and two-channel system, respectively. First, we note that P_L slightly *increases* upon deposition of Mn. This increase is strongest for submonolayer coverages and weakens after completion of the first layer. Upon deposition of Mn the work function should decrease from 4.7 eV for Fe(100) to about 4.1 eV for Mn, with the strongest change occurring in the submonolayer range. Calculations on polarized electron capture from a jellium metal indicate that P_L increases for lower work functions and might explain our findings.[29]

The spin polarization P_S shows a pronounced *decrease* from the clean Fe surface value (about 23%) to a small, almost constant value for Mn coverages beyond 0.5 ML. This behavior has been observed in all of our measurements with a scattering of data for different runs by $\pm 2\%$ around a mean value of about 2%. The initial decrease of P_S is in accordance with an opposite orientation of the Mn moments with respect to the Fe substrate. Assuming a proportionality between P_S and the surface magnetization M_z and comparable moments of Mn and Fe surface layer atoms,[4,30] a coverage of 0.5 ML should be sufficient to cancel P_S, in accordance with our observation. A parallel alignment of the Mn moments with an opposite orientation to the Fe moments is expected from calculations for the submonolayer regime[4] and in accordance with recent experiments on spin-resolved photoelectron spectroscopy[11] and magnetic circular dichroism.[13,14]

Upon further deposition of Mn P_S saturates to a small value, i.e., the net in-plane magnetization M_z is almost zero, indicating a loss of in-plane ferromagnetic order or the occurrence of antiferromagnetic or ferrimagnetic order. This is in accordance with calculations for a 1-ML film,[4,7] because the (antiparallel) Mn-Mn coupling prevails in the monolayer regime compared to the (antiparallel) Mn-Fe coupling dominant in the submonolayer regime. The absolute values of the moments of adjacent Mn atoms are similar (3.2–$3.4\mu_B$),[4,6–8] resulting in a ferrimagnetic in-plane configuration with very small net magnetic moment. An almost vanishing net moment for 1–3-ML films has been also reported in Refs. 11 and 14 and explained by intralayer or interlayer antiferromagnetic configurations.

In Fig. 2 we show data for growth at 560 K. The transition in growth mode from layer to island growth at 4 ML (Ref. 3) leads to a decrease for the intensity of reflected ions [Fig. 2(a)]. A similar behavior appears for the observed polarization of orbital momenta P_L, which abruptly decreases at 4 ML [Fig. 2(b), squares]. We ascribe this decrease to an increased surface roughness, which lowers P_L as has been found for poorly polished surfaces.

Note that P_L for the clean surface does not change with temperature within the experimental error. This is expected, since the geometry of the capture process does not vary with the target temperature. In contrast, the spin polarization P_S decreases with temperature (16% at 560 K [Fig. 2(b), circles]). This is typical for surface magnetism, where the magnetization shows a pronounced decay with increasing temperature.

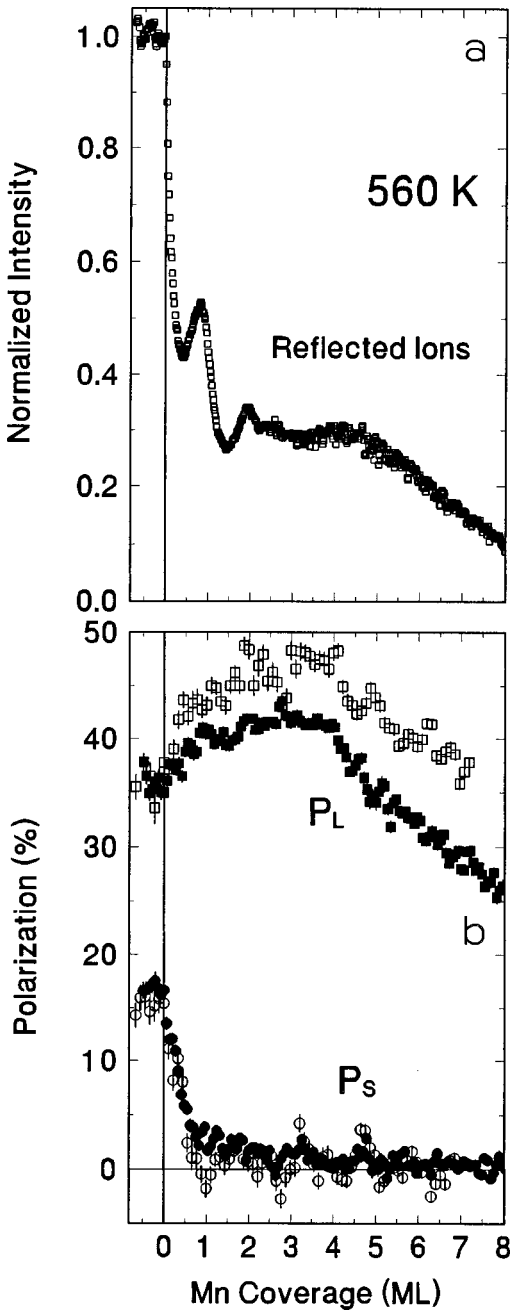

FIG. 2. Same as Fig. 1, but for growth at 560 K.

The dependence of P_S on the Mn coverage is similar for 560 K and room-temperature growth, although, for a coverage <1 ML, the decrease seems to be weaker for the higher temperature. An intermixing between Mn and Fe atoms during growth of the first layer, as observed by us recently,[31] may explain this finding.

In summary, we have presented experiments on the emission of polarized light after grazing scattering of fast ions from ultrathin magnetic films. The growth process as well as the magnetic properties of the topmost film layer can be studied simultaneously and in real time. A sensitivity to fractions of a monolayer is achieved. The spin polarization observed for electrons captured into the 3^3P state of He atoms after scattering from ultrathin Mn films grown on Fe(100) at different temperatures indicates an in-plane ferromagnetic order of Mn moments with antiparallel Mn-Fe coupling in

the submonolayer regime. The nearly vanishing spin polarization for the monolayer film is in accordance with an in-plane antiferromagnetic or ferrimagnetic order in agreement with recent theoretical predictions. Similar experiments on ultrathin Cr films on Fe(100) are in progress.

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