Spin-polarized electron capture during ion impact on a ferromagnetic surface

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25-keV He^+ ions are scattered from a clean and Mn-covered (1 ML) Fe(100) surface. The spin polarization of electrons captured into excited $3\,^3P$ states of He atoms is probed via an analysis of polarized fluorescence light. For projectiles impinging from grazing to normal incidence we observe the same spin polarization of captured electrons. By a coverage of 1 ML of Mn atoms we demonstrate the surface sensitivity of the method and discuss its potential for spin-sensitive microscopy and sputter depth profiling. [S0163-1829(99)00406-3]

In recent years considerable progress has been achieved in the study of magnetic phenomena at surfaces by making use of electron, ion, and atom beam probes.[1–4] In electron capture spectroscopy (ECS) fast ions are scattered under a grazing angle of incidence from a magnetized surface and capture (spin-polarized) electrons into stable and excited atomic states. In early studies of Rau and Sizmann[5] with 150-keV D^+ ions, the spin polarization of electrons captured from the target into the ground state of deuterium atoms is deduced from the analysis of a subsequent nuclear reaction. This method has been applied in studies on surface magnetism for clean metal targets and thin films.[6,7] Since in grazing scattering geometry trajectories of projectiles and the final electron capture events are localized to a region well in front of the topmost layer of surface atoms,[8,9] ECS is a technique with high sensitivity to the topmost surface region.

In an equivalent manner, polarized electrons captured into excited atomic states can be studied by the decay via the emission of polarized light.[10–13] From circularly polarized fluorescence light, e.g., described by the Stokes parameter $S/I=[I(\sigma^-)-I(\sigma^+)]/[I(\sigma^-)+I(\sigma^+)]$ with intensities of light with negative/positive helicity $I(\sigma^-)$, $I(\sigma^+)$, the spin polarization $P_s=\langle S_z\rangle/S$ of captured electrons is deduced in a straightforward manner.[14] P_s can be related to a long-range magnetic order at the target surface. The advantage of this "optical" technique over conventional ECS with nuclear detection is its simpler setup, good signals, and a wider choice of projectiles and their energies. It has been applied so far to clean Fe(110) (Refs. 10–16) and Ni(110),[17] a thin film of Mn on Fe(100),[18] and an amorphous ferromagnetic $Fe_5Co_{75}B_{20}$ ribbon.[19]

Ion-scattering experiments under a glancing angle of incidence Φ_{in} (up to some degrees) run under conditions of planar "channeling,"[20] i.e., the projectile motion is characterized by a "slow" motion normal to the surface plane and a "fast" parallel motion. Then atomic projectiles are reflected from the topmost layer of surface atoms. Penetration into the bulk of a crystal occurs only at larger angles of incidence (above "critical angles") or is mediated by defects of the target surface. Combined with features of electron capture, ECS is characterized by a high sensitivity to the topmost layer of a surface. In this respect ECS clearly differs from most other methods of studying surface magnetism with probing depths of some atomic layers, as, e.g., techniques based on electron spectroscopy or on the "magneto-optical Kerr effect."

One property of ECS is the large target area probed by the projectile beam impinging under grazing incidence. Since the projection of the beam onto the target surface scales with $1/\sin\Phi_{in}$, i.e., for $\Phi_{in}\approx 1°$ with about 50, even a collimation of the incoming beams in the sub-mm domain results in a signal from an area of several mm in length from the target surface. It is obvious that the grazing incidence geometry is counteracting requirements for a microscopic spatial resolution.

In this paper we report on our finding that the high surface sensitivity of ECS is also obtained for larger angles of impact towards normal incidence. In our experiments, 25-keV He^+ ions are scattered under $1.5°\leqslant\Phi_{in}\leqslant 80°$ from a clean and flat Fe(100) target that is kept by current pulses through the coil of an attached yoke in a remanent state of magnetization at a pressure of about 10^{-10} mbar. The target surface is prepared by cycles of grazing sputtering with 25-keV Ar^+ ions and subsequent annealing. Polarized fluorescence light emitted from excited He atoms in the $2s\,^3S$-$3p\,^3P$, $\lambda=388.9$ nm transition is detected through a quartz window by means of a quarter-wave retarder plate, a narrow bandwidth interference filter, a linear polarizer (dichroic sheet or beam splitter cube), and a cooled photomultiplier. At current densities of some 10 nA/mm^2 for the incident ion beams typical count rates amount to several 10^3 counts/sec in pulse-counting mode.

Because of the broken symmetry in the atomic capture and excitation process at the surface, the $3p\,^3P$ term shows a polarization of atomic-orbital angular momenta $P_L=\langle L_z\rangle/L$,[21,22] independent of the electronic spin, leading also to an emission of circularly polarized light. For a magnetized target the spin polarization P_s and P_L of captured electrons can be deduced from two measurements of S/I for reversed settings of magnetization $[S/I(\uparrow),S/I(\downarrow)]$ (Refs. 10–19) where P_s scales with $[S/I(\uparrow)-S/I(\downarrow)]$ and P_L with $[S/I(\uparrow)+S/I(\downarrow)]$.

In Fig. 1 we show P_s and P_L for incident 25-keV He^+ ions as a function of the angle of incidence Φ_{in} referred to the surface plane. The Fe(100) target, a thin slice of 10 mm diameter in a soft magnetic yoke is at room temperature and the sectional area of the impinging ion beam is 0.3×1 mm^2 or 1×1 mm^2. The single crystal is magnetized along an easy direction ⟨001⟩. We observe that within the uncertainties P_s is constant, whereas P_L shows a pronounced decrease with

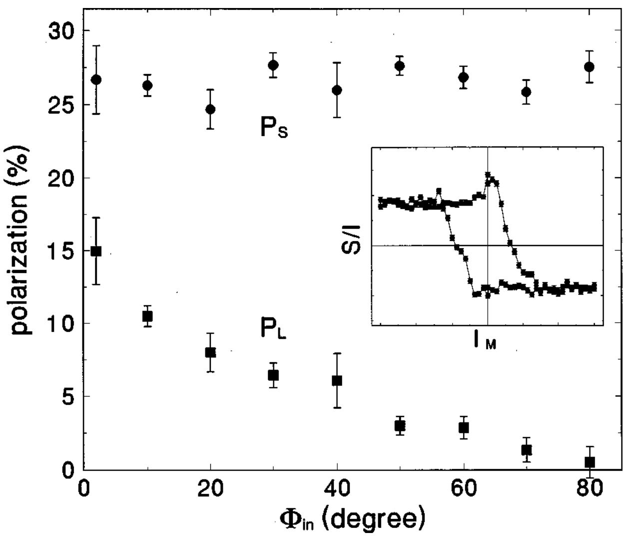

FIG. 1. Spin polarization P_s and polarization of orbital momenta P_L as function of the incidence angle Φ_{in} with respect to the surface for 25-keV He^+ ions scattered from a clean Fe(100) surface. $T = 300$ K. Inset: Hysteresis loop (Stokes parameter S/I vs current through the coil I_M) for $\Phi_{in} = 70°$.

increasing Φ_{in}. The different dependences of P_s and P_L have considerable consequences for the application of ECS to the study of surface magnetism. The observations can be interpreted qualitatively by the following simple arguments.

The decrease of P_L with Φ_{in} is understood in a similar manner as for the excitation of fast ions after transmission through thin solid foils (''beam-foil excitation'') where the surface normal is tilted with respect to the beam axis.[21] For projectiles leaving the solid along the surface normal (corresponds to $\Phi_{in} = 90°$ here) axial symmetry for the capture/excitation process holds and P_L vanishes.[22] With decreasing Φ_{in} the axial excitation symmetry is broken, giving rise to a nonzero P_L as observed.

Our finding of a constant P_s with Φ_{in} means that the spin polarization of captured electrons is the same despite clearly different excitation mechanisms for grazing and oblique/normal incidence collisions with the target surface. The projectile trajectories are described below critical angles Φ_{crit} (some degrees here) by concepts of channeling,[20] characterized by specular reflection and no penetration of projectiles into the bulk. For $\Phi_{in} > \Phi_{crit}$ an increasing portion of projectiles penetrate into the bulk, accompanied by an increased mean energy loss and a decreasing coefficient of reflection with increasing Φ_{in}. For the conditions of our experiments we estimate for normal incidence a coefficient of reflection of about 0.1.[23,24] In this respect it is interesting to note that the normalized intensity of emitted light from reflected projectiles decreases from grazing to normal incidence by only a factor of 2, i.e., excitation under normal impact is clearly more efficient than under grazing incidence.

Charge transfer and excitation under those conditions can be summarized by the admittedly oversimplyfying, but intuitive picture that (excited) atomic states can only survive the interaction with the solid, if the distance of the atomic core from the surface R is of the same size as the spatial extension of the relevant electronic orbits $\langle r \rangle$, i.e., $R \approx \langle r \rangle$.[24] For the $3p\,^3P$ term $\langle r \rangle$ amounts to about 1 nm. Then the final forma-

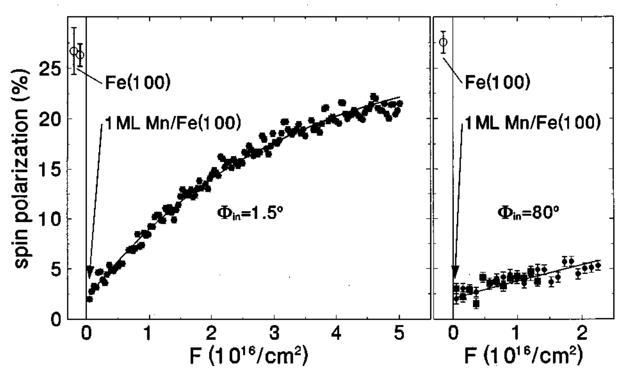

FIG. 2. Spin polarization P_s for 25-keV He^+ ions scattered from clean Fe(100) (open circles) and 1-ML Mn/Fe(100) (solid circles) under $\Phi_{in} = 1.5°$ (left panel) and $\Phi_{in} = 80°$ (right panel), respectively. The fluence is measured after the shutter of the evaporator is closed.

tion of atomic states that decay thereafter via the emission of photons with radiate lifetimes ($\tau \approx 90$ ns here) much longer than the collision times will take place on the outgoing part of the trajectories. This leads to the high surface sensitivity of electron capture even for impact under normal incidence.

In the inset of Fig. 1 we show a hysteresis loop where S/I is recorded as a function of the current through the coil I_M for $\Phi_{in} = 70°$. The constant saturation values for S/I at higher I_M demonstrate that the data are not affected by magnetic stray fields in the vicinity of the target. Those fields generally lead to a precession of angular momenta accompanied with a reduced S/I. A discussion of the curve is beyond the status of the present experiments and the scope of this paper.

The topmost layer sensitivity of electron capture is demonstrated here by epitaxial growth of a thin film of 1 ML Mn on the Fe(100) surface at room temperature. For this system an almost vanishing net magnetic moment of Mn atoms, but only a slight reduction of the net moment of Fe atoms in the interface layer has been observed.[25–28] In a recent study performed under grazing incidence[18] we have shown that the spin polarization of about 27% observed for the clean Fe(100) surface almost vanishes for this film (left side of left panel in Fig. 2). We observe the same feature for $\Phi_{in} = 80°$ (left side of right panel in Fig. 2) and conclude a similar topmost layer sensitivity for this case. A discussion of the physics of surface magnetism resulting from our observation is given elsewhere.[18]

In Fig. 2, we have plotted P_s during consecutive measurements of fluorescence light as a function of the fluence for continuous irradiation of the thin-film surface with 25-keV He^+ ions. This bombardment leads via sputtering to a gradual removal of atoms from the surface layer and to an increase of P_s. For grazing scattering (left panel of Fig. 2) a fluence of some 10^{16} ions/cm^2 leads to a substantial removal of Mn atoms and P_s approaches the value for the clean Fe(100) substrate. For large-angle impact we observe a similar effect (right panel of Fig. 2); however, we stopped the experiments at a smaller fluence in order to prevent the Fe(100) crystal from damage in the subsurface region due to ion implantation. In this respect it is important to note that the time for recording the data (left panel) amounts to about 2 h. Single measurements of P_s with sufficient statistics, however, can be performed within several seconds.

The solid curves through the data are fits using a model where the Mn coverage decreases at a rate proportional to the product of the ion fluence and the coverage. For a given coverage, the net spin polarization is obtained as weighted average of the polarization for clean Fe surface patches and for Mn covered areas.

In conclusion, we see interesting consequences for future work that directly follow from the experimental observations outlined above.

(1) So far ECS has been restricted to grazing scattering from atomically smooth surfaces. Our findings clearly show that this method can be also applied at larger angles of impact beyond the regime of channeling. This offers the possibility of studying magnetic properties of structured or rough surfaces, imperfect films, adsorbed clusters, etc.

(2) The combined effects of sputtering surface atoms and capture of spin-polarized electrons bears the potential of a magnetic sputter depth profiling with topmost layer resolution. Our method is similar to previous work[1,29] where the spin polarization of secondary electrons excited by noble-gas ion impact is analyzed. The advantage of the method proposed here is its simple setup and a low sensitivity of excited atoms to external electric fields. Then it should be particularly simple to perform in the same experiment an analysis of secondary ions, i.e., an element-specific secondary-ion mass spectroscopy profiling. Since the electron capture into excited atomic states is not limited to He projectiles, the use of heavier projectiles, e.g., Ne or Ar, would lead to more efficient sputtering yields.

(3) Scanning microscopy to study magnetic bit or domain patterns at surfaces appears to be feasible. For (near) normal-incidence impact of ions the probed area of the surface is given by the lateral width of the focused incident ion beam. With modern liquid-metal ion sources spot sizes of only a few tens of nanometers across can be achieved.[30] In order to achieve a sufficient signal, the photon counting rates can be enhanced by a larger solid angle and, in particular, a detection of atomic transitions over the whole optical spectral range (detection without interference filters). Then an analysis of local magnetic properties at the topmost layer of surfaces is possible. Though the striking feature of the technique is the ultimate surface sensitivity, a lateral resolution typically achieved with other state-of-the-art microscopies (10–100 nm) with magnetic contrast[31] is conceivable. Moreover, a combination with lithography, micromachining, and ion beam deposition could be performed.

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