Emission and capture of spin-polarized electrons during ion reflection at magnetic surfaces: A new tool in surface magnetism

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Using electron-capture spectroscopy (ECS) and electron emission spectroscopy (EES), the electron-spin polarization (ESP) at the *topmost* surface layer of ferromagnetic materials is investigated. In ECS, the capture of spin-polarized electrons during grazing-angle surface reflection of fast deuterons is used to investigate the ESP due to long-ranged *and* short-ranged ferromagnetic order. In EES, the *ion-induced* emission of low-energy, spin-polarized electrons from magnetic surfaces is used to study long-ranged surface ferromagnetic order. The surfaces are well characterized using Auger electron spectroscopy, low-energy electron diffraction, reflection high-energy electron diffraction, and scanning tunneling microscopy. Nonzero ESP is found at surfaces of atomically clean, amorphous $Fe_{80}B_{20}$ and (110)-surface-oriented Ni picture-frame single crystals. We have studied the *energy-resolved* ESP of the emitted electrons at Ni(110) surfaces. The energy distribution of the ion-induced emitted electrons is *strongly* different from that of electron-induced secondary electrons and shows a marked spin dependence. The found energy and spin distributions of the electrons, originating from the topmost surface layer, bear a close resemblance to the electronic and magnetic properties of Ni(110) surfaces. The net average ESP of the emitted electrons amounts to $+5.5\%$, which is close to the total net Ni magnetization of $+5.6\%$.

I. INTRODUCTION

At present, magnetic phenomena at surfaces, interfaces, and in thin films receive great theoretical and experimental attention.[1,2] This arises from the fact that magnetic materials serve as nearly ideal systems to explore basic concepts in theoretical physics such as phase transitions and critical behavior of thermodynamic quantities in two or three dimensions.[2] On the other side, besides the experimental investigation of electronic and magnetic properties of surfaces, interfaces, and thin films, they are of pivotal importance in modern magnetic recording and memory devices.

In this paper, we show that the use of ion–surface interaction at small angles of incidence provides a powerful means to study the magnetic and electronic properties of the *topmost* atomic layer of magnetic materials. At *grazing* angles of incidence, ions cannot penetrate into a surface; they are specularly reflected. This simple fact reveals the extreme surface sensitivity of experimental methods where ion–surface interaction is used to retrieve data on the physical properties of surfaces with unprecedented sensitivity.[3]

We report on fundamental information on surface magnetic properties obtained by using capture of spin-polarized electrons and emission of spin-polarized electrons during grazing-angle reflection of fast ions at magnetic surfaces.

For our experiments, we use well-defined surfaces of Ni(110) picture-frame single crystals and surfaces of amorphous $Fe_{80}B_{20}$. It is well known that materials such as $Fe_{80}B_{20}$ are expected to possess unlimited potential for use in many technologically important applications.[4]

Using electron-capture spectroscopy (ECS),[3] we have studied the magnetic order, characterized by the short-ranged electron-spin polarization (ESP), at surfaces of the amorphous ferromagnet $Fe_{80}B_{20}$. We find that the surfaces order ferromagnetically at room temperature. There is no evidence found for so-called magnetically dead layers at atomically clean surfaces. At room temperature, the short-ranged ESP amounts to $\sim 55\%$.

Further, we use electron emission spectrosopy (EES) to study the emission of spin-polarized electrons at surfaces of thin films of amorphous $Fe_{80}B_{20}$. The spin polarization of the emitted electrons amounts to around 15%–20%, which clearly establishes the existence of long-ranged ferromagnetic order at these surfaces.

Using EES at surfaces of Ni(110) picture-frame single crystals, we have studied the *energy-resolved* ESP of the emitted electrons. The energy distribution of the electrons is *strongly* different from that of electron-induced secondary electrons and shows a marked spin dependence. The found energy and spin distributions of the electrons, originating from the *topmost* surface layer, bear a close resemblance to the electronic and magnetic properties of Ni(110) surfaces. Details about these new measuring techniques are discussed in Sec. II.

II. EXPERIMENTAL

The experimental results, presented in this paper, were obtained using ECS and a recent methodological advancement of ECS, EES. We use ECS to study the long-ranged or short-ranged ferromagnetic order at surfaces of magnetic materials.[3] The fundamental physical process in ECS is the capture of one or two spin-polarized electrons during small-angle surface reflection of fast deuterons D^+ or protons H^+.

For an angle of incidence of 0.2°, the distance of closest approach of the ions to the reflecting surface is about 0.8–1.5 Å. Therefore, the ions solely probe the spin-polarized local

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electron densities of state of the topmost surface layer of the sample.[3]

We briefly discuss the ECS technique as regards the investigation of two-electron capture processes. Details about the use of one-electron capture processes for the study of long-ranged ferromagnetic order are discussed in Ref. 3.

For the measurement of short-ranged ferromagnetic order, we use either protons or deuterons and study two-electron capture processes ($H^+ + 2e^- = H^-$ or $D^+ + 2e^- = D^-$). The only stable bound state of H^- or D^- is the $1s^2\ {}^1S$ state. Therefore, H^- or D^- can only be formed by capture of electrons with opposite spins. In the experiments, reported here, we use 20-keV hydrogen ions for the ESP measurements. For this energy range, the characteristic length within two electrons are captured by a single ion, amounts to ~ 5 Å. Thus, two-electron capture processes are sensitive to short-ranged ferromagnetic order existing within a range of two atomic neighbors. It is obvious that two-electron capture will be strongly suppressed by the presence of short-ranged ferromagnetic order where predominantly electrons with parallel oriented spins are available for capture by a single ion. The reduction in the H^-/H^+ ratio in the reflected beam, relative to that for a nonmagnetic target such as Cu, therefore, provides a direct measure of the short-ranged ESP at a magnetic surface.[6]

In EES, we use again grazing-angle surface reflection of 10 to 30 keV H^+ and study the emission of ion-induced, spin-polarized electrons. Using an einzellens, we record the emitted electrons emitted along the surface normal (emission cone angle: 11°) of a remanently magnetized target. For energy analysis of the electrons, we use an electrostatic energy analyzer.

For spin analysis, the electrons are accelerated to 150 eV and hit a "low-energy, diffuse scattering spin analyzer." This spin detector was recently pioneered by Pierce, Celotta, and Unguris and allows for a fast and efficient determination of the long-ranged ESP.[5] It is based on low-energy (150 eV) diffuse scattering of electrons from a high-Z target (preferably Au). For precise zero-ESP calibration, the Au target is replaced by an Al target. The spin–orbit interaction provides the physical basis of this spin analyzer. Using two channeltrons, positioned at equal angles to the right and the left of the direction of the incoming electron beam, we detect electrons elastically backscattered from the Au target at angles of 135°.

In our version of this detector, we focus the incoming electron beam on a Au target of 2 mm in diameter. This certifies that the measured count ratios of backscattered electrons in the channeltrons depend only on the counting statistics. The count ratios of the electrons, detected in the two channeltrons, provides a direct measure of the ESP of the electrons emitted from the magnetic surface. For further details on this simple and efficient ESP detector, we refer to Ref. 5.

A target magnetizing field {along the [111] direction in the (110) surface plane of the picture-frame single crystal} is applied to magnetically saturate the sample, by aligning otherwise randomly oriented Weiss domains, thereby producing a macroscopic magnetization along which sign and magnitude of the ESP are measured. With P, the ESP along the target magnetizing field, we have $P = (n^+ - n^-)/(n^+ + n^-)$, where n^+ and n^- are the fractional numbers of emitted electrons with spin moment antiparallel (majority spin electrons) and parallel (minority spin electrons), respectively, to the target magnetizing field. Emitted spin-polarized electrons originate at widely (2 mm) separated points at the target surface. Therefore, in EES measurements, long-ranged ESP is detected.

Numerous experimental difficulties must be overcome in order to maintain clean surfaces during the ESP measurements. The samples are prepared in a target preparation chamber at 1×10^{-10} mbar, and the residual surface contaminations C, S, and O were shown, using Auger electron spectroscopy, to be <0.01 monolayer (ML). Further details are given in Refs. 6–8. Then, the samples are transferred to a measurement chamber, and the cleaning procedures are repeated in this chamber. At present, in our low-energy experiments, the working pressure is at $\sim 8\times 10^{-10}$ mbar. Therefore, the experimental ESP data, resulting from these measurements, are considered to represent lowests limits of the original ESP at the surfaces investigated so far.

III. RESULTS AND DISCUSSION

At present, there is a lack in experimental results on magnetism at surfaces of bulk and thin-film materials.[9] One of the reasons for the lack of experimental data is, that there are not many experimental techniques of experimental probing depth of the order of, or less than, a lattice constant normal to the surface.

ECS and EES are sensitive to the magnetic order existing at the *topmost* layer of a surface thus enabling us to determine *intrinsic* surface magnetic properties. For the measurement of long-ranged ESP, a sample is magnetized to saturation in the surface plane, and the long-ranged ESP of the captured or emitted electrons is measured using EES or ECS. For the measurement of short-ranged ESP, using two-electron capture processes, the samples need not to be macroscopically magnetized because, even in demagnetized samples, the size of existing microscopic domains, which are always spontaneously magnetized, is larger than the overall distance within which two individual electrons are captured.[6]

In our first low-energy ECS experiments, we use small-angle reflection of 20-keV protons at surfaces of the amorphous ferromagnet $Fe_{80}B_{20}$. At room temperature, we find a short-ranged ESP of 55%. This value is very close to the spin polarization of photoelectrons emitted *near* photothreshold in photoemission experiments[10] at surfaces of $Fe_{83}B_{17}$.

For the spin polarization of ion-induced electrons, emitted from $Fe_{80}B_{20}$, we find polarization values between + 15% and + 20%. These values clearly show that these surfaces indeed are ferromagnetic. For these EES experiments, we used an angle of incidence of 3°. The obtained ESP data compare favorably with the value of the total average magnetization [11](+ 21%) and with the ESP of electron-induced secondary electrons emitted from surfaces of amorphous $Fe_{83}B_{17}$ amounting to + 10%.[12] We remark that for such large angles of incidence, the incident ions can penetrate into the solid and induce the emission of electrons also from layers located deeper than the topmost surface layer.

In order to obtain more refined data on spin-polarized EES, we performed EES experiments at (110) surfaces of Ni picture-frame single crystals. Reducing, for 25-keV incident protons, the angle of incidence from 3° to 1°, which prevents the penetration of the protons into the solid, results in a drastic change in the energy distribution $n(E)$ of the emitted electrons.

In Table I, results on $n(E)$ and on the electron-spin polarization $P(\%)$ as a function of the analyzer voltage are given for spin-polarized electrons emitted along the surface normal of Ni(110) at 300 K. Discussing our experimental data in terms of the kinetic energy of the emitted electrons and taking the work function of the analyzer and of the target into account, we refer to an energy scale of the electrons where the zero point of the electron kinetic energy is located at ~ 29 eV. Starting from very low values at ~ 1 eV, $n(E)$ increases steeply at ~ 3 eV and reaches a maximum at 4 eV, followed by a gradual decrease of $n(E)$ with increasing E. At 13 eV, $n(E)$ amounts to 0.1. Surprisingly, the ESP reaches a minimum (predominance of minority spin electrons) at an electron energy $E = 2.2$ eV and increases steeply to $+18\%$ at 2.5 eV followed by a steep decrease at 3 eV to $+7\%$ and reaches a constant value of 5.5% for energies above 9 eV. The value of 5.5% is close to the well-known value of 5.6% for the total average magnetization of electrons in Ni.[3]

At present, the electron emission processes are far from being well understood. However, from our experimental configuration, it is known that the protons cannot penetrate into the solid, and therefore can only excite electrons from the topmost surface layer. This simple fact already reveals the extreme surface sensitivity of this new method to study surface magnetism with extreme spatial resolution.

TABLE I. Energy distribution $n(E)$ and electron-spin polarization (ESP) of spin-polarized electrons emitted during grazing-angle surface reflection of 25-keV protons at surfaces of magnetized Ni(110) as a function of the analyzer voltage.

Analyzer voltage E (V)	Intensity $n(E)$ (arbitrary units)	ESP P (%)
28	0	···
29	0.1	0
30	1.2	0
30.5	1.9	− 8.7
31	2.6	− 12.8
31.2	2.8	− 13.2
31.5	2.2	+ 17.5
32	9.7	+ 7
32.8	15.1	+ 6.2
33	13.0	+ 6.0
33.5	10.2	+ 5.6
34	7.7	+ 5.1
34.5	4.9	+ 5.1
35	3.8	+ 5.3
36	2.0	+ 6.0
37	1.1	+ 5.8
38	0.5	+ 5.5
39	0.2	+ 5.5
40	0.15	+ 5.5
42	0.1	+ 5.5
45	0	···

In a series of publications, the ion-induced emission of electrons from solid is investigated. Great emphasis is given to calculate total yields for the electron emission.[13] To the best of our knowledge, there exists no theoretical investigation on the quantitative or qualitative relation between the energy distribution of ion-induced emitted electrons and the electronic density of state at surfaces of metals. Provided the availability of such a theory, one could directly link the measured electron energy and spin distribution to the spin-polarized electron densities at the topmost surface layer of ferromagnetic materials.

It is well known from literature data that, for nongrazing angles of incidence, the shape of ion- or electron-induced electron spectra is characterized by a steep increase from zero electron energy to a maximum located at very low energies (< 2 eV), being nearly independent of projectile type and energy, followed by a smooth decrease with increasing energy, reaching zero at around 40–70 eV.[14] In contrast to these findings, our experimental data, obtained using grazing-angle ion reflection at atomically flat surfaces, are characterized by a strong reduction of low-energy electron cascades which are caused by multiple scattering of secondary electrons originating from atomic layers located below the first few atomic layers.

A further interesting feature of the experimental results consists in the fact that the ESP of the emitted electrons exhibits, as function of electron energy, a drastic change in the sign of the ESP, a feature that is predicted by many theories on spin-polarized electron bands in Ni.

We believe that ion-induced emission of polarized electrons during grazing-angle ion–surface interaction can develop into a very important technique to study surface magnetism.

More experimental and theoretical work is needed to establish a close and direct relation between angle-, energy-, and spin-resolved experimental data from this unique and simple technique and theoretical values derived from spin-polarized surface band-structure calculations.

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