Spin-Polarized Electrons in Collisions of Multicharged Nitrogen Ions with a Magnetized Fe(001) Surface

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We report on the first spin-resolved energy spectra for the emission of electrons during grazing scattering of 150 keV multicharged nitrogen ions from a magnetized Fe(001) surface. A substantial spin polarization for *KLL* Auger electrons emitted in the final stage of the neutralization sequence during the interaction of multicharged ions with a metal surface is observed. We conclude from our data that the projectile *L* shell is dominantly populated by electrons from the conduction band of the target. For low energy electrons we find an increase of their spin polarization with an increase of the projectile charge.

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Over the last decade considerable progress has been achieved in the understanding of the complex electronic interaction phenomena occurring during collisions of slow multicharged ions with solid surfaces [1,2]. In brief, the neutralization and deexcitation sequence of a multicharged ion approaching a metal surface is described in two steps. Electron transfer between projectile and solid sets in at a distance where, in a classical picture, the electronic potential barrier between a metal surface and an ion is lowered to the Fermi level [3–5]. In this regime, via resonant electron capture and reionization, electronic levels of the projectile with binding energies comparable to the target work function are predominantly populated (i.e., highly excited Rydberg levels), whereas inner shell vacancies in the projectile ions survive so far. As a consequence, a basically neutral atom is formed with a dominant occupation of outer electronic shells, a so called "hollow atom" [1,2,6]. The formation sequence of those atoms is well described by a classical overbarrier approach [5,7,8], as tested by effects of the image charge on projectile trajectories [9–12]. These processes take place in an interval of distances starting from some 10 a.u. (a.u. = atomic unit) down to some a.u., where the considerable potential energy (typically keV) stored in the inner shell vacancies of the highly excited projectile is still available.

The subsequent evolution of the hollow atom, especially the filling of inner shell vacancies, proceeds in close contact with the solid during penetration into the bulk or backscattering from the surface plane. In this part of the neutralization sequence, Auger electron and radiative transitions, electron capture, ionization, and electron promotion processes play a dominant role. From intra-atomic Auger electron [1,13–16] and x-ray transitions [17] basic interaction mechanisms have been revealed; however, the contributions of the mechanisms related to the filling of projectile inner shells are still under debate (see below).

In the experiments reported here, we have scattered 150 keV N^{q+} ions ($q \leq 6$) under a grazing angle of incidence $\Phi_{in} = 1.5°$ from a bcc Fe(001) surface. For these scattering conditions electron spectra imply an efficient and fast filling of the projectile *L* shell [18–20]. Furthermore, the relatively high projectile energy of $E =$ 150 keV warrants sufficiently large currents of N^{6+} ions from our 10 GHz electron cyclotron resonance ion source. For grazing scattering, the ions approach the surface plane with an energy of only $E_z = E \sin^2\Phi_{in} \approx 100$ eV. From computer simulations on projectile trajectories we conclude that the majority of projectiles (more than 95%) is reflected from the topmost surface layer ("planar surface channeling") [21].

The target was mounted on a toroidal soft-magnetic yoke and kept in a remanent, single domain state of magnetization parallel to the surface plane ("in-plane" magnetization) as checked by the magneto-optical Kerr effect. The base pressure in the UHV chamber was some 10^{-11} mbar, and the target surface was prepared by cycles of grazing sputtering with 25 keV Ar^+ ions and subsequent annealing to about 700 °C. Electrons emitted normal to the surface plane were detected by means of a cylindrical sector analyzer (Focus CSA300) with an acceptance angle of 30° for the detection cone and a spin-polarized LEED (SPLEED) detector [22] mounted behind the exit slit of the analyzer. In this detector electrons are scattered at a constant energy of 104.5 eV from a W(001) surface and the intensities of the $\langle 2, 0 \rangle$-LEED spots are recorded by means of four channeltrons. From the asymmetries *A* of the in-plane and "out-of-plane" signals we deduced the electronic spin polarization $P = A/S$, where $S \approx 0.2$ is the effective Sherman function [22,23]. In order to eliminate instrumental asymmetries, measurements were performed for reversed settings of the magnetization of the target and checked against data for scattering from a paramagnetic Ta sheet translated to the position of the target.

Figure 1 shows a spectrum for electron energies ranging from about 100 to 450 eV. In this spectral range one observes electrons emitted during the filling of the *K*-shell vacancy of N^{6+} ions via a *KLL* Auger transition. Depending on the occupation of the *L* and *M* shells the corresponding electron energies lie between about 350 and 380 eV [13,18]. The open circles in the figure represent intensities

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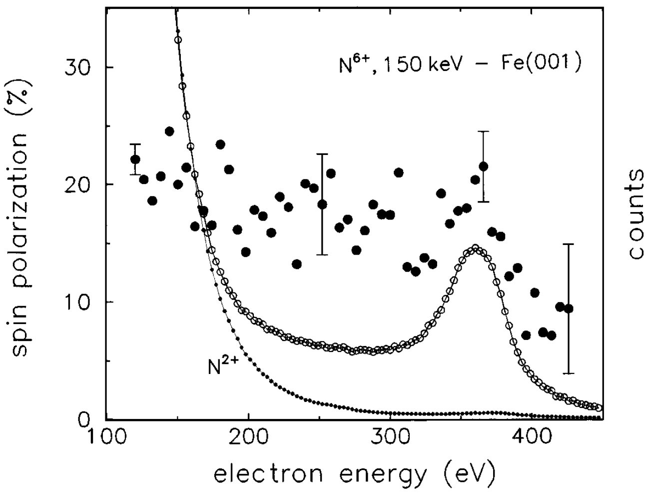

FIG. 1. Electron spectra for 150 keV N^{6+} ions scattered from a magnetized Fe(001) surface under $\Phi_{in} = 1.5°$. Open circles: intensity (about 20 000 counts were accumulated at the *KLL* Auger peak; for comparison, the small full circles represent data for 150 keV N^{2+} ions normalized to the same intensity at 150 eV). Full circles: spin polarization.

of the electron signal (sum of the counts in the four detectors) for spectral scans recorded during bombardment of the surface with N^{6+} ions. These data show a pronounced peak at the expected *KLL* Auger transition energies with a substantial low energy tail. Comparison with a spectrum obtained with N^{2+} ions at otherwise the same conditions (small full circles) indicates that the spectral tail in the N^{6+} data results mainly from inelastically scattered Auger electrons.

The energy resolution in our experiments of about 13 eV (FWHM) is not sufficient to resolve $1s\,2s^n\,2p^m$ ($n + m \leq 5$) satellites in the Auger transitions as demonstrated in previous studies with N^{6+} ions and other sorts of multicharged ions [1,13–16,18,19]. The (poor) spectral resolution in our work has been sacrificed in favor of highest possible count rates, since the signals for energy analyzed electrons are reduced owing to the small efficiency of the SPLEED device by more than 3 orders of magnitude. With a high pass energy (320 eV) and a large width of entrance and exit slits of the electron analyzer, i.e., settings resulting in a low energy resolution, we obtained count rates of up to 50 counts per second for *KLL* electrons at a current of some 10 nA for the incident N^{6+} beam.

For the measurements of asymmetries, i.e., normalized differences in intensities, long times of accumulation of detector counts are needed, in order to achieve an adequate statistical spread of the data. The full circles in Fig. 1 represent the spin polarization P of emitted electrons. The data were accumulated over a total time of about 50 h, where actual measurements had to be suspended after about 1 h for preparation cycles of the crystal surfaces in the scattering and SPLEED chamber lasting for about 30 min. Irrespective of the still considerable statistical uncertainties of the polarization data at higher electron energies, the data reveal a substantial spin polarization of about 20% for the in-plane component of the electron spin. In passing we note that the out-of-plane components (not shown here), i.e., an orientation of spins normal to the surface plane, are consistent with a vanishing spin polarization. At the high energy tail of the Auger peak the polarization seems to drop, but owing to large statistical uncertainties this conclusion is vague.

The finding of a substantial spin polarization for direct and inelastically scattered *KLL* Auger electrons emitted during the deexcitation of multicharged ions at a magnetized surface is new and allows one to obtain important information on the filling of inner shell vacancies in collisions of multicharged ions with surfaces. In order to illustrate this issue, we display in Fig. 2 a sketch of an energy diagram showing a typical (metal) surface-ion potential and some electronic levels of target and projectile ion. The full circles represent occupied atomic levels, the open circles inner shell vacancies, and the vertical arrows a *KLL* Auger transition in the projectile. Close to the surface atomic levels might be shifted via dielectric response, screening, and promotion effects.

Different mechanisms have been discussed for the population of inner shells: (1) cascading from higher levels via intra-atomic Auger (AI) or radiative transitions, (2) Auger neutralization (AN), (3) Auger deexcitation (AD), and (4) direct vacancy transfer (VT) with inner shells of target atoms (for a discussion, see, e.g., Refs. [1,18,25–28]). For the conditions of our experiments ($E = 150$ keV,

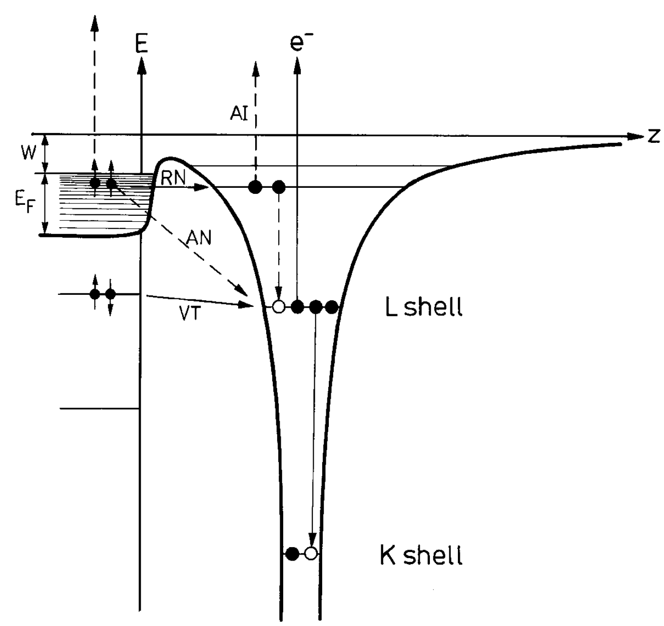

FIG. 2. Sketch of an energy diagram showing charge transfer and electronic transitions for a multicharged ion in front of a metal surface. RN = resonant neutralization; AN = Auger neutralization; VT = vacancy transfer between inner shells; AI = autoionization. Note that electronic levels might be shifted via screening and promotion effects.

$\Phi_{in} = 1.5°$) we expect from high resolution studies by Limburg *et al.* [13,18] [$E = 60$ keV, $\Phi_{in} = 2.5°$, Al(110) target] a filling of the projectile L shell with higher transition rates than the KLL Auger decay. For light ions, two mechanisms are assumed to compete in the fast filling of the L shell ("side feeding" [24]): Auger transitions and capture from target atoms [25]. The direct capture of target core electrons in close collisions with target atoms is attributed to a level crossing (Landau-Zener) or orbital promotion (Fano-Lichten) type of charge transfer [1,13,18,19,24–29]. Other filling mechanisms are considered as being too slow in view of the KLL Auger decay rates.

It is beyond the scope of this paper to present a detailed model describing the polarization data. However, a first qualitative interpretation of data can be performed in view of the fact that conduction electrons of bcc Fe have a mean spin polarization of about 27% [30]. For the direct capture to nitrogen levels of the L shell with binding energies ranging from about 80 eV ($1s\,31^6$) to 20 eV ($1s\,21^5\,31$) [18] $3p$ electrons of Fe atoms with binding energies of 52.7 eV will dominate. Since the $3p$ shells of Fe are fully occupied and show small exchange splittings, the dynamic inner shell transfer will lead to a negligible effective spin polarization of electrons captured into inner shells of the projectiles. Thus a dominant fraction of electrons transferred to inner shells during the ion impact with the surface has to stem from the valence band. As a consequence, direct filling of the L shell from target inner core levels in grazing collisions of N^{6+} ions with a Fe(001) target might play a less important role than assumed so far in published model calculations. Rather it seems that the population of inner shells proceeds via electronic transitions from the conduction band assisted by dynamical and screening effects [25,31]. In this respect the spin polarization of Auger electrons emitted in inner shell transitions during multicharged ion-surface impact provides important complementary information in addition to standard spectroscopic work.

We also investigated the low energy part of the electron spectrum which is dominated by secondary electrons showing a peak at about 2 eV. In order to reduce effects of small stray fields, the emitted electrons were accelerated by a bias voltage of -10 V applied to the target. We checked that this procedure led to an intended constant shift in electron energies but did not affect asymmetries (see, e.g., Ref. [32]). The different symbols in Fig. 3 represent spin polarizations of electrons emitted for the impact of isotachic N^{2+}, N^{5+}, and N^{6+} ions. For comparison we have also plotted results obtained with our setup for the bombardment of the target surface with 2 keV electrons (dashed curve). The spin polarization for electron impact shows the established behavior of values close to the mean polarization of conduction electrons at higher energies (about 20 to 30 eV here) [33–35] and of an increase to lower energies, caused by the so called "spin-filter effect" (spin dependent scattering length for inelastically scattered electrons) [36]. The polarization data for ion impact reveal a similar structure but have somewhat larger polarizations. The striking new feature found with ions is an enhancement of the spin polarization with the projectile charge. This effect is particularly pronounced for the data obtained with N^{6+} ions, with polarizations up to 70%.

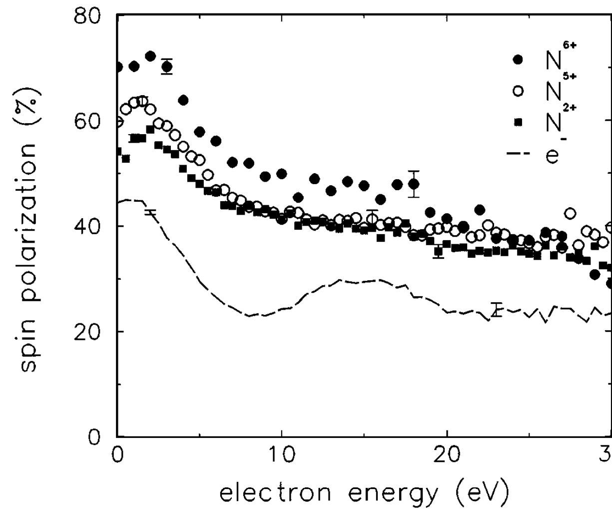

FIG. 3. Spin polarization of low energy electrons emitted during scattering of 150 keV N^{2+} (full squares), N^{5+} (open circles), and N^{6+} ions (full circles) from a Fe(001) surface. The dashed curve represents data for bombardment of the target with 2 keV electrons.

An explanation for this finding might be related to the fact that the low energy electron spectrum comprises information on the initial neutralization sequence of multicharged ions, i.e., capture and loss of electrons in front of the surface resulting in the formation of hollow atoms. Owing to effects caused by screening, promotion, etc., the majority of those weakly bound electrons is "peeled off" from the projectile and results in the emission of predominantly low energy electrons [37]. The data can possibly be interpreted by the (surface state) enhancement of the magnetic moment at the surface as found from ground state calculations [38]. The number of electrons captured by the ions from the conduction band well in front of the surface rises with the charge of the incident ion and may explain the enhanced spin polarizations for increasing projectile charge owing to enhanced contributions of those electrons to the yields of low energy electrons. This is in full accord with established concepts describing the initial interaction sequence of multicharged ions in front of metal surfaces.

In conclusion, we present the first data on the emission of spin-polarized electrons after the impact of multicharged ions upon a magnetized surface. In our spin and energy resolved spectra we observe a substantial spin polarization, which we attribute to a predominant capture of electrons from the conduction band of the magnetized sample. From our data for KLL Auger electrons we conclude that a direct transfer of inner shell electrons between target and projectile will play a minor role for the population of inner shells for the system studied here. We hope that our new data will stimulate new theoretical work on the filling dynamics of projectile inner shells during the interaction of multicharged ions with metal surfaces, where the electronic spin—measured in this type of work for the first time—serves as a "label" for the origin of the electrons.

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