Polarized Light Emission after Grazing Ion-Surface Scattering Due to Capture of Spin-Polarized Electrons

H. Winter, H. Hagedorn, R. Zimny, and H. Nienhaus
Institut für Kernphysik der Universität Münster, Wilhelm-Klemm-Strasse 9, D-4400 Münster, Federal Republic of Germany

J. Kirschner[(a)]
Institut für Atom- und Festkörperphysik, Freie Universität Berlin, Arnimallee 14, D-1000 Berlin 33, Federal Republic of Germany

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We have observed the capture of polarized electrons into excited terms of atoms after the interaction of fast ions with a magnetized Fe(110) surface at grazing incidence. The spin polarization of captured electrons results in a modified circular polarization fraction of fluorescence light. This experiment has considerable potential as a new analytical tool for investigating surface magnetism with extreme surface sensitivity, as a method for detailed studies of ion-surface interaction, and as a means to produce nuclear spin-polarized beams.

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We report on first investigations of polarized light emission after scattering of fast ions from a magnetized surface at grazing incidence. The new technique allows us to detect the electron spin polarization at magnetized surfaces via electron capture into excited terms of scattered atoms. This is an important development in the studying of surface magnetism by ion scattering which was pioneered by Rau and Sizmann,[1] who scattered fast deuterons from magnetized targets and observed the capture of polarized electrons in the $1s$ ground term of deuterium via an asymmetry in the nuclear $T(d,n)^4\mathrm{He}$ reaction. Despite the fact that the mechanisms of the capture process are only vaguely understood, this technique has been used to study long- and short-range magnetic order at surfaces.[2]

The basic idea of the experiments reported here is that an ensemble of excited atomic terms with anisotropically distributed total angular momenta generally decays via the emission of polarized light. Atomic anisotropy (orientation) can be induced via excitation of atoms by grazing ion-surface scattering and/or by capture of polarized electrons. The orientation of atomic angular momenta can be probed simply by the fraction of circular polarization in the fluorescence light as given by the Stokes parameter[3]

$$S/I=[I(\sigma^-)-I(\sigma^+)]/[I(\sigma^-)+I(\sigma^+)]\,,$$

where $I(\sigma^-)$ and $I(\sigma^+)$ denote the intensities of light with negative and positive helicity, respectively. After grazing ion-surface scattering from nonmagnetic targets, large orientations resulting in polarized fluorescence light with S/I up to 95% have been observed.[4]

The time scale for the final-state formation in grazing ion-surface collisions ($<10^{-14}$ s) is generally short in comparison to typical fine- and hyperfine-interaction frequencies, so that the anisotropic formation of excited terms can be described in terms of orbital angular momentum L. After electron capture from the solid, coupling with electronic spin S and nuclear spin I via fine-structure and hyperfine interactions will take place. The capture of polarized electrons from a magnetized sample induces anisotropy into the electronic spin system. Thus, via fine-structure interaction the anisotropy of total angular momenta of atomic terms is modified relative to that resulting from the capture of unpolarized electrons.[5,6]

This Letter gives first clear experimental evidence that the interaction of fast ions with a magnetic surface results in a modification of the orientation of excited terms observed after grazing ion-surface scattering. We show that this modification is directly related to the spin polarization of captured electrons. Since the final electron capture process takes place at a well defined distance from the topmost layer,[7-10] this technique is extremely surface sensitive.

This capture of polarized electrons into excited atomic terms may stimulate a number of new developments which are relevant for different fields of physics, in particular, for surface studies:

(1) Although it is not complete, the current understanding of the mechanisms of the formation of excited terms after grazing ion-surface scattering implies a powerful new technique for detailed studies of surface magnetism with extreme surface sensitivity which, in addition, is rather simple in application.

(2) Capture of polarized electrons "marks" the target electron and thus enables detailed studies of the atom-surface interaction. We will deduce a first application of this method from our data presented here.

(3) Capture of polarized electrons at grazing ion-surface scattering will significantly enhance the atomic anisotropy so that the ion beams with high nuclear polarization may be obtained.[5,11,12]

In our experiments we scattered He^+, N^+, and Ar^+ ions with energies between 10 and 250 keV at grazing angles $\Phi < 1°$ off a clean, magnetized Fe(110) surface at

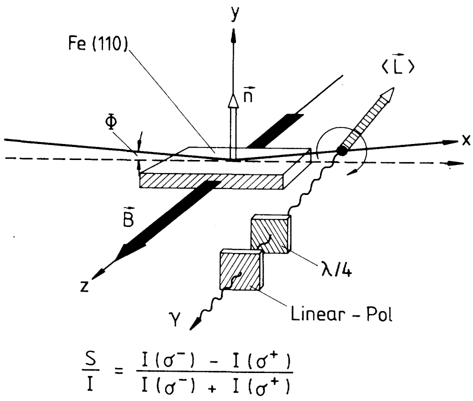

FIG. 1. Sketch of the experimental setup.

room temperature. Normal to the plane of scattering, we observed the polarized fluorescence light by an achromatic quarter-wave plate, a Glan Thompson polarizer, a McPherson Mod.218 spectrometer, and a cooled RCA 31034A photomultiplier (Fig. 1). The Fe(110) target was an elliptically shaped slice of about 13 mm diameter and 2 mm thick mounted on top of a magnet yoke of toroidal shape made of soft iron. The sample was oriented with the [001] direction of easy magnetization normal to the plane of scattering and positioned by a manipulator in an UHV chamber at a base pressure of 10^{-11} mbar. The cleanliness of the surface was inspected by Auger-electron spectroscopy. A current pulse of some amperes was applied through a coil around the yoke and the sample was kept in remanence by the magnetic short circuit. By this technique an electron spin-polarization parallel or antiparallel with respect to the collision-induced orientation is obtained. By a gradual reduction in ac current through the coil, an "unpolarized" target is approximated on a macroscopic scale by the formation of randomly oriented magnetic domains at the surface. In order to observe the effects reported here, a clean surface is crucial and was provided by our heating the target up to about 600 °C and sputtering by noble-gas ions.[13]

In Fig. 2 the circular polarization fraction S/I of the light emitted in the He I 2^3S-3^3P, $\lambda = 389$ nm transition is displayed for seven consecutive runs with 150-keV He^+ ions at a grazing angle of incidence $\Phi \approx 0.8°$. In the first run the sample is randomly magnetized so that the electronic spins captured from the solid are randomly oriented and the observed polarization is solely due to orbital angular momentum orientation $\langle L \rangle$. This anisotropy is partially reduced by the transfer of orientation into the (isotropic) electronic spin system via fine-structure interaction before the excited term has decayed.

In the second run a magnetic field is applied antiparallel to the orientation of orbital angular momenta $\langle L \rangle$. the pronounced increase in S/I indicates that electrons with a spin polarization along that axis are the ones that are predominantly captured. Reversal of the direction of the magnetic field consequently results in a reduction of the collision-induced orientation as demonstrated in the third run. Subsequent runs in which the magnetization of the target is changed and which differ in time by about 10 min clearly demonstrate the reproducibility and stability of our data.

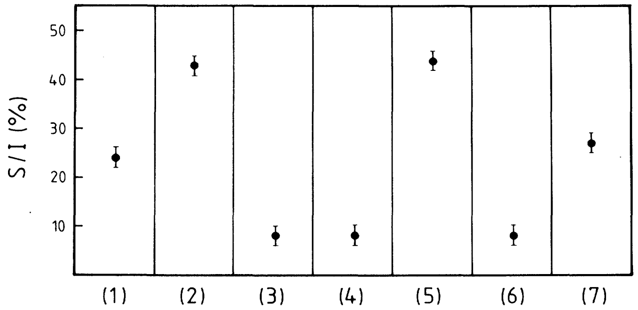

FIG. 2. S/I of light emitted in the He I 2^3S-2^3P, $\lambda = 389$ nm transition after the interaction of 150-keV He^+ ions with a Fe(110) surface in seven consecutive runs. Runs (1) and (7) are taken with a demagnetized target; (2) and (5) are taken with a magnetization antiparallel to the direction of orientation of orbital angular momentum ($-z$ direction); (3), (4), and (6) are taken with magnetization parallel to orbital angular momenta. Note the strong variation of circular polarization S/I depending on whether the captured majority electrons are oriented parallel or antiparallel with respect to orbital angular momenta.

In Table I we summarize some typical results of our experiments for a number of unresolved multiplet transitions in various atoms. For singlet transitions (electronic spin $S=0$) S/I independent of the spin polarization of the target. This observation is expected, since singlet terms have no electronic spin polarization. It shows that our data are not affected by stray magnetic fields. In all doublet transitions ($S=\frac{1}{2}$) and even more pronounced in the triplet transitions ($S=1$), S/I depends on the polarization of the target. The fact that S/I increases by induction of a magnetization opposite to the orientation of orbital angular momenta indicates that the spins of captured electrons are predominantly oriented in that direction, i.e., majority electrons are captured. These results already qualitatively demonstrate the potential of this technique to study surface magnetism.

In our experiments typical counting rates generally amount to some thousand per second so that—aside from its rather simple use—this technique provides short intervals for measurements. This clearly differs from the time-consuming method of Refs. 1 and 2.

For a quantitative analysis of our data, we have to establish the connection of the light polarization S/I with the orientation of orbital angular momenta $P_L = \langle L_Z \rangle / L$ and the polarization of electronic spins $P_S = \langle S_Z \rangle / S$. This problem is related to the coupling of ensembles with anisotropic distributions of angular momenta. In general, the evaluation for our problem yields somewhat lengthy expressions. For the present case of unresolved

TABLE I. Stokes parameters S/I of light emitted in some multiplet transitions in He I, Ni II, Na I, and Ar II for target electrons not polarized (0), and polarized parallel (↑) and antiparallel (↓) with respect to $\langle L \rangle$; the upper term of transition is given in the table. From the data we deduce the atomic orbital angular momentum and electron spin polarizations P_L and P_S, respectively. All polarizations are given in percent; the precision of the data is estimated to be about ±5% for S/I and about ±10% for P_L and P_S.

S	Upper term of transition	Energy (keV)	S/I (%)			P_L	P_S
			0	↑	↓		
0	He I $1s(^2S)3p\,^1P$	150	32.2	33.5	34.3	−22	
	$1s(^2S)3d\,^1D$	150	53.3	52.2	53.0	−35	
	N II $2p(^2P)3p\,^1D$	150	53.1	54.1	52.2	−44	
$\frac{1}{2}$	Na I $(^1S)3p\,^2P$	200	26.7	29.9	23.0	−49	15
		150	28.4	31.0	25.0	−52	13
	Na I $(^1S)4p\,^2P$	150	23.3	27.0	20.4	−43	14
	Ar II $3p^4(^1D)4p\,^2F$	230	73.6	75.5	70.9	−51	14
1	He I $1s(^2S)3p\,^3P$	25	⋯	27.6	−5.4	⋯	22
		150	25.7	43.3	8.1	−34	23
	He I $1s(^2S)3d\,^3D$	150	48.8	59.7	39.1	−41	22.5
	N II $2p(^2P)3p\,^3D$	150	55.9	62.5	43.9	−57	21
	$2p(^2P)3d\,^3F$	250	60.0	67.0	54.4	−51	20

(LS,JI,F)-$(L_0S,J'I,F')$ multiplet transitions and $P_LP_S \ll 1$, these experiments can be approximated by the particularly simple form

$$P_S=[S/I(\uparrow)-S/I(\downarrow)]/2A_S\,, \qquad (1)$$

$$P_L=[S/I(0)]/A_L=[S/I(\uparrow)+S/I(\downarrow)]/2A_L\,, \qquad (2)$$

where "↑" ("↓") denotes magnetization along z ($-z$) in Fig. 1 and "0" represents the "nonmagnetic" target. A_S and A_L can be considered as "analyzing powers" of our method to detect P_S and P_L. Both constants depend on the type of transition and structure of the atomic term chosen for detection and are discussed in detail elsewhere.[5,14] For the transition chosen in our investigations, we find A_S between 0.16 and 0.75.

According to Eqs. (1) and (2), P_S and P_L are deduced from polarization data obtained at opposite directions of magnetization of the target. As an independent check of consistency of the evaluation of data and of the validity of both equations, we verified the relation $S/I(0)=[S/I(\uparrow)+S/I(\downarrow)]/2$ in Eq. (2). P_L and P_S, obtained from the analysis of our data, are given in Table I. We note that P_S can also be deduced from relative spectral line intensities, in case fine-structure multiplet components are spectroscopically resolved.[14] However, optical resolution of fine-structure splittings in experiments with fast ions is limited to cases of sufficiently large fine-structure interaction.

Doublet transitions ($s=\frac{1}{2}$) between levels with a singlet parent term seems to be best suited to probe the electronic spin orientation. In this case the observed effect is due to the polarization of a single captured electron which reflects the state of spin polarization at the surface. At the projectile velocities used ($0.4 < v/v_0 < 1.2$, where v_0 is the Bohr velocity) the one-electron pickup proceeds via resonant charge exchange.[7–10,15] This electron transfer results in a well defined distance of formation y_s from the topmost layer of the solid where the final atomic term is formed and which can be approximated by $y_s \approx (2Z)^{1/2}|E_B|+3$ a.u. ($E_B=$ binding energy of atomic term in a.u., $Z=$ charge of ion core). Furthermore, electrons of the solid with energy $E \approx E_F$ are predominantly captured.[7] As a consequence, our technique is assumed to be sensitive to the electron spin polarization of Fermi electrons $P_S(E_F,\,y_s)$ at a distance y_s from the topmost layer. Variation of y_s by the selection of atomic terms with different binding energies E_B offers the new and unique possibility to study the dependence of P_S on the distance from the solid.

A corresponding analysis of our data with respect to doublet transitions is given in Table II. Note that we find no significant dependence of P_S on the distance y_s

TABLE II. Binding energies and distances of capture y_s (in atomic units) from the topmost atomic layer for some Na I and Ar II terms and P_S as deduced from our analysis of data. The D I term refers to data of Refs. 1 and 2. Since the mechanisms of interaction are only vaguely known in this case, no well defined y_s is given.

Upper term of transition	E_B (a.u.)	y_s (a.u.)	P_S (%)
Na I $4\,^2P$	0.05	31	14 ± 2
Na I $3\,^2P$	0.11	16	15 ± 2 13 ± 2
Ar II $4p\,^2F$	0.25	9	14 ± 2
D I $1\,^2S$	0.5	?	13 ± 2

within a range $10 < y_s < 30$ a.u.

In this context it appears remarkable that our data are in quantitative agreement with the analysis by Rau[2] for the same type of sample where $P_S = (13 \pm 2)\%$ is found for electrons captured in the $1s\,^2S$ ground term of the deuterium atom. A recent investigation of the neutralization of protons at grazing surface scattering indicates that at the velocities of the experiments of Ref. 2 ($v \approx 1.7 v_0$) effects in the very tail of the electron density distribution at the surface play a minor role.[16] Charge transfer seems to basically proceed via Auger-type processes[17] in close vicinity to the atomic surface plane ($y_s < 2a_0$). At present no detailed calculations are available which allow us to unravel the contributions of electrons within the conduction band of the solid with respect to symmetry and energetic locations. The good agreement with our data seems to show that the ground term of deuterium is preferentially populated by Fermi electrons[2] and that P_S for the Fe(110) surface has a weak variation with distance y from the surface plane.

Recent theoretical calculations of the electronic structure of Fe(110) yield $P_S(E_F) = +(53 \pm 4)\%$ from the bulk densities of states[18-20] and $P_S(E_F) = -(70 \pm 20)\%$ from the planar densities of states at the surface layer.[20] At present no information on the planar densities of states well above the topmost layer of Fe(110) is available. The negative value of P_S at the surface layer results from a localization of minority-spin electrons among the surface atoms as has been found for the Fe(001) surface.[21] However, above the topmost layer of the Fe(001) surface, the majority-spin electrons dominate again and the same may hold for the Fe(110) surface.

A second attractive feature of our new method is to gain deeper insights into the ion-surface interaction mechanisms, since the electrons of the solid are "marked" via their spin polarization. Interesting information in this respect is gained from the spin polarization observed in triplet transitions ($S = 1$) with a doublet parent term. The polarization for two unpaired electrons is significantly larger than that of a single electron in a doublet term. Assuming that the observed spin polarization results from the capture of a single polarized electron, whereas the second electron (to form the $S = 1$ term) is not polarized, one may calculate the degree of polarization P_S to obtain the experimental result. We find $P_S \approx 32\%$, which is much larger than the polarization obtained from our experiments at doublet transitions. However, assuming that both electrons of a triplet term are captured with the same spin polarization, one finds $P_S = (16 \pm 2)\%$ which agrees fairly well with the experimental data for the capture of a single polarized electron. We suppose at the present state of investigation that the formation of excited terms in ion-surface scattering is not a pure one-electron capture process.

In conclusion, we have outlined a new method which has considerable potential to investigate surface magnetism and electronic ion-solid interaction. Since for Ni(110) $P_S = -(96 \pm 3)\%$ is reported,[2] one may expect large effects in using this surface instead of Fe(110). This will imply the feasibility of our producing highly nuclear polarized beams by this technique.[11,12]

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[a] On leave from Institut für Grenzflächenforschung und Vakuumphysik, Kernforschungsanlage Jülich, Postfach 1913, D-5170 Jülich, Federal Republic of Germany.

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