Selective Population of Ground Terms in ^{14}N Atoms after Ion-Beam–Surface Interaction at Grazing Incidence

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The orientation of angular momenta in the ground terms of ^{14}N atoms after the interaction of 350-keV $^{14}N^+$ ions with a solid surface at grazing incidence is investigated by a Zeeman quantum-beat technique. After the ion-solid interaction, a term-selective and highly polarized fast beam of nitrogen atoms is observed. The phenomenon is interpreted in terms of a Pauli-principle–induced selective population.

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The ion-beam–surface interaction at grazing incidence (IBSIGI) yields a large orientation in the distribution of the orbital angular momenta of the reflected atoms and ions.[1–6] The large degree of orientation after the surface interaction results in a considerable transfer of anisotropy into the electronic and nuclear spin systems via the fine and hyperfine (hf) interactions, so that high-resolution atomic spectroscopy by the quantum-beat technique is feasible[7–9] and a vector polarization of nuclear spins up to $P \approx 20\%$ is obtained after surface scattering.[7,10] In order to interpret such high degrees of nuclear spin polarization, a dominant polarization and population of the ground terms has to be assumed in the grazing incidence, but no direct measurements are available which support this concept.

We report here on the first investigation of the product of orientation times population of ground terms in ^{14}N atoms and ions after IBSIGI, for which we have developed a new technique. Figure 1 displays the setup of our experiment. 350-keV $^{14}N^+$ ions are deflected by electric field plates onto a polycrystalline copper target (surface roughness $\lesssim 100$ nm) at a grazing angle of about 0.4°. The target is positioned in a UHV chamber at a pressure of about 10^{-8} mbar, with differential pumping at both ends to isolate it from the vacuum of the beam line (5×10^{-6} mbar) and the region of detection (5×10^{-7} mbar). By the ion-surface interaction, ground and excited terms of the ^{14}N atoms and ions are anisotropically populated, but only the orientation of excited terms is directly accessible to observation by the polarization of the fluorescence light emitted in transitions from those levels. In order to obtain access to ground terms, we detect the nuclear orientation of the atoms after IBSIGI by an optical method[7] and induce selectivity with respect to different terms by a static magnetic field H_z.

For the further discussion of the experiment, we choose the orbital angular momentum fractional orientation ${}^L\mathscr{O}_{\gamma LS}$ of a (γLS) term [γ denotes the additional quantum numbers needed to specify the (LS) term] and write a time-averaged nuclear fractional orientation ${}^I\mathscr{O}_{\gamma LS}$ (the x axis is the axis of quantization)[7]:

$$ {}^I\mathscr{O}_{\gamma LS} = \frac{\sum_{JF} C^1}{\sum_{JF} C^0} {}^L\mathscr{O}_{\gamma LS} = \sum_{JF} C_{JF}\, {}^L\mathscr{O}_{\gamma LS}, \tag{1} $$

with

$$ C^k = (-1)^{J+L+S+k} \frac{(2J+1)(2F+1)^2}{(2S+1)(2I+1)} \begin{Bmatrix} J & J & k \\ L & L & S \end{Bmatrix} \begin{Bmatrix} F & F & k \\ J & J & I \end{Bmatrix} \begin{Bmatrix} F & F & k \\ I & I & J \end{Bmatrix}. \tag{2} $$

In the experiment all populated and oriented terms contribute to the total nuclear orientation:

$$ {}^I\mathscr{O} = \sum_{\gamma LS} \alpha_{\gamma LS}\, {}^I\mathscr{O}_{\gamma LS} = \sum_{\gamma LS} \alpha_{\gamma LS}\, {}^L\mathscr{O}_{\gamma LS} \sum_{JF} C_{JF}, \tag{3} $$

where $\alpha_{\gamma LS}$ are normalized weighting factors depending on the corresponding populations of the different (γLS) terms.

To detect the nuclear orientation ${}^I\mathscr{O}$ we use the optical method introduced by Andrä and co-workers.[7] The polarized ions enter a thin foil (surface normal $\hat{n} \parallel \hat{z}$) where a new configuration of the electronic shell is formed at the rear surface of the foil. Since for this interaction process axial symmetry holds, no orientation of angular momenta can occur.[11] The orientation of the nuclei is practically unaffected by the short foil in-

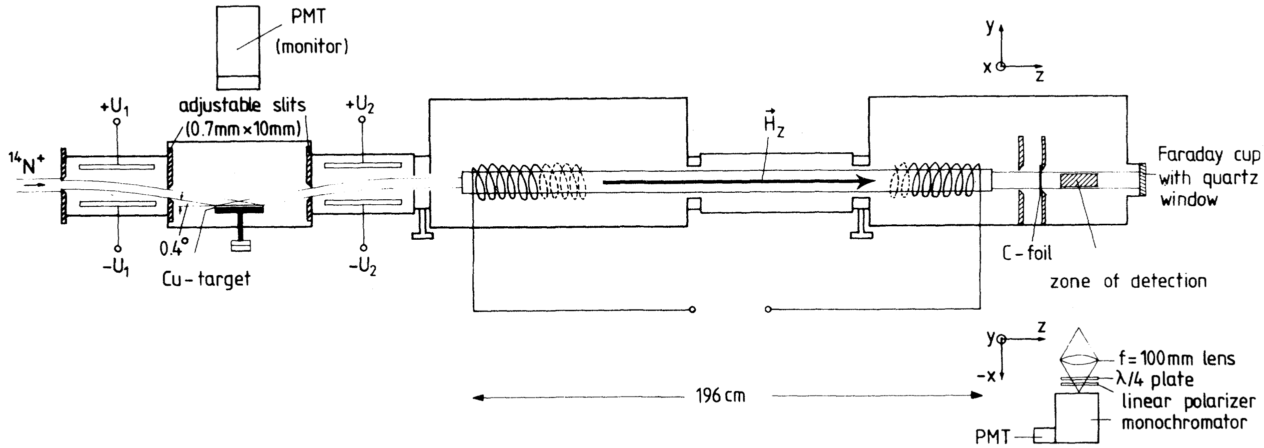

FIG. 1. Experimental setup.

teraction (10^{-14} s) and is afterwards partially transferred back to the "new" electronic shell so that the transferred orientation to a spectrally selected term of the "new" atom is a direct measure of ${}^{I}\mathcal{O}$.

In our experiment we observe the circular polarization fraction S/I of the light emitted along x of the N II $2p3s\ {}^3P-2p3p\ {}^3D$ $\lambda = 568$ nm transition with a cooled RCA 31034A multiplier through a quarter-wave plate, a linear polarizer, and a Jobin Yvon HRS2 spectrometer. For this multiplet transition one deduces[6]

$$S/I = 0.34\,{}^{I}\mathcal{O}. \tag{4}$$

By combining Eqs. (3) and (4) we also have access to the orientation of the ground terms ${}^{L}\mathcal{O}_{\gamma LS}$, but the information on a single (γLS) term is masked by the superposition of different terms.

In order to introduce selectivity with respect to (γLS) terms in the detection scheme outlined so far, we apply the longitudinal static magnetic field H_z between the solid target and the foil over a distance of 196 cm. In this field the angular momentum of a level $|\gamma(LS)JIF\rangle$ precesses, because of its associate magnetic moment $\vec{\mu} = g_F \vec{\mu}_B$ ($\vec{\mu}_B$ is the Bohr magneton), around the H_z direction, so that

$$S/I = 0.34 \sum_{\gamma LS} \alpha_{\gamma LS}\,{}^{L}\mathcal{O}_{\gamma LS} \sum_{JF} C_{JF} \cos[g_F(\mu_B/\hbar)\Delta t H_z], \tag{5}$$

where Δt is the time of flight of the atom in the magnetic field H_z. Because of experimental reasons we keep Δt constant, and by scanning H_z S/I oscillates with frequencies determined by the g_F factors. In Fig. 2 a result corresponding to the detection of the singly ionized component of ^{14}N is displayed

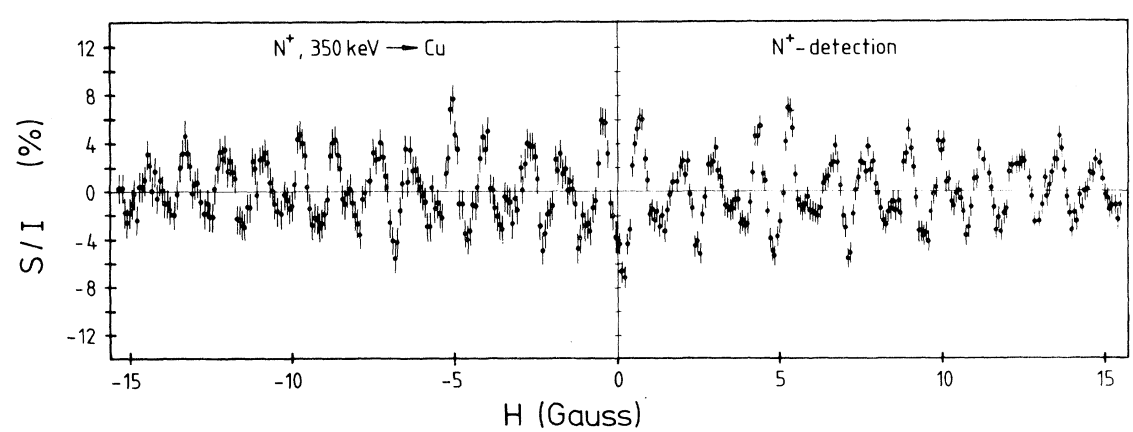

FIG. 2. Circular polarization fraction S/I of the N II $2p3s\ {}^3P-2p3p\ {}^3D$ $\lambda = 568$ nm transition as a function of the longitudinal magnetic field H_z after IBSIGI of 350-keV N^+ ions. The detection is performed with the reflected N^+ ions which are separated by electric field plates behind the IBSIGI target.

which represents the first "Zeeman quantum-beat" measurement[12] on atomic or ionic ground terms in a fast beam.

In Fig. 3 we show the Fourier transform of the data in Fig. 2 and refer the observed frequencies to a g_F-factor scale. In the spectrum of the experimental g_F factors, the two relevant N^+ ground terms $2p^2\,{}^1D$ and $2p^2\,{}^3P$ are clearly identified and we compare this result with the theoretical spectrum normalized with respect to a common $\alpha_{\gamma LS}{}^L\mathcal{O}_{\gamma LS}$ for both terms. The solid lines represent the ${}^1D$ term and the broken ones the ${}^3P$ term. We find a satisfactory agreement especially within the ${}^1D$ terms.

Figure 4 shows the spectrum of g_F factors of the neutral ^{14}N component where we expect contributions from the $2p^3\,{}^2P$ and $2p^3\,{}^2D$ terms. The surprising result is, however, that the g_F spectrum of the ${}^2D$ term is nicely reproduced whereas components of ${}^2P$ are comparable with the noise of the transform; i.e., the neutral component of the reflected beam only consists of atoms in the ${}^2D$ term. Since the N I $2p^3\,{}^4S$ and N II $2p^2\,{}^1S$ ground terms are not oriented after the surface interaction, they cannot be detected by this technique. We conclude, however, from the large transferred nuclear orientation that these terms are poorly populated.

In order to interpret this surprising result we propose a model of a selective capture of an oriented electron by an already oriented ion core. In a first step the N II $2p^2$ core becomes oriented by the surface interaction with respect to the x axis, and in a second step a further $2p$ electron is captured with a pronounced orientation of its angular momentum also along x.

The main features of our model are illustrated with the help of Fig. 5. The $2p^2\,{}^1D$ and $2p^2\,{}^3P$ N II ground terms serve as parent terms which are assumed to be oriented by a collision with the surface. Then in the second step a third $2p$ electron is captured with a large degree of orientation of its angular momentum. As outlined in Fig. 5 the capture of a $2p$ electron with oriented angular momentum predominantly results in an N I $2p^3\,{}^2D$ term with N II $2p^2\,{}^3P$ as the parent term, whereas the formation of a neutral term from the N II $2p^2\,{}^1D$ term is forbidden by the Pauli principle. Thus the absence of the ${}^2P$ components in the N I spectrum of the g_F factors can be well understood.

The model for the interpretation of our data essentially contains the features of the two-step model for the production of polarization by grazing-incidence surface interaction given by Schröder,[13] which was developed to describe the polarization of the emission from excited terms with $L=0$. To account for a finite orientation of such excited S terms ($L=0!$) in Ar II and Kr II,[4] Schröder had to assume a considerable orientation of the ion core. The existence of such a high core polarization could not unambiguously be shown by data obtained in former studies on excited states. Our experiments support this concept of Schröder and demonstrate that even the ion core has to be highly polarized in the grazing-incidence surface interaction. Furthermore, our data imply that excited terms are poorly populated in comparison to the ground terms; this is consistent with the recent results of Winter *et al.*[14] where evidence for resonant electron transfer processes in IBSIGI were found.

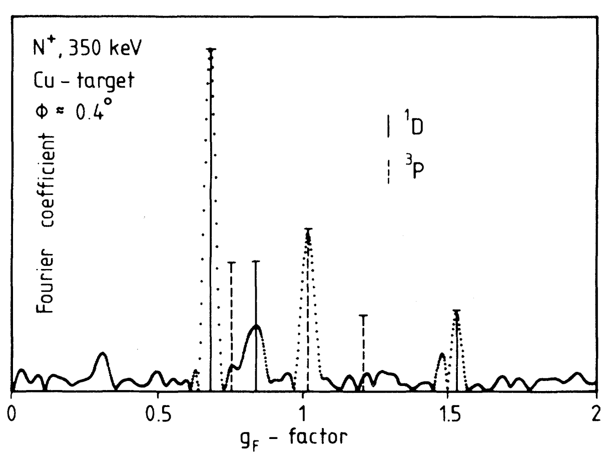

FIG. 3. Specturm of the g_F factors of the N^+ component after IBSIGI.

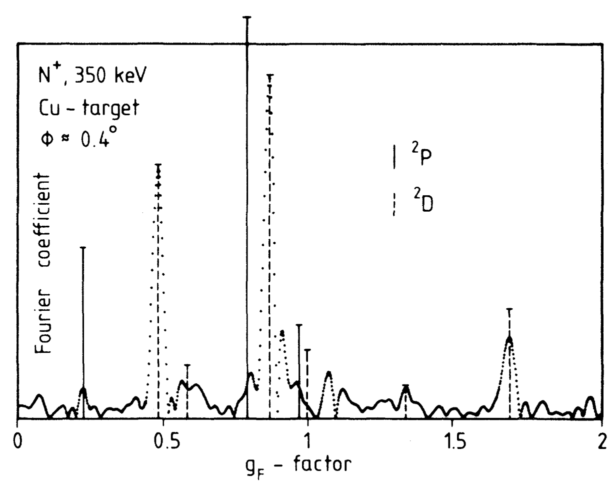

FIG. 4. Spectrum of the g_F factors of the N^0 component after IBSIGI.

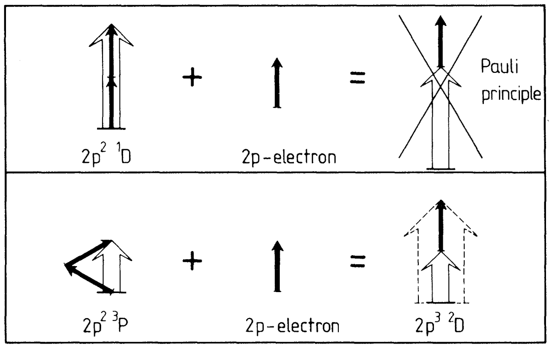

FIG. 5. Sketch of the two-step model of the coupling of an oriented $2p$ electron to an oriented ion core.

The shift of the minimum of S/I from $H_z=0$ in Fig. 1 is due to Earth's magnetic field which was not shielded in the experiments. For the minimum of S/I in the neutral component we find $S/I=-0.095$ and from Eqs. (3) and (4) we deduce ${}^L\mathcal{O}_{^2D}=0.84$. Thus after IBSIGI a highly oriented and term-selective beam of fast neutral nitrogen atoms is available which can be applied for the study of collision processes with polarized beams.

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