In-Beam Polarization of Light Radioactive Nuclei Using Grazing Surface Scattering

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A 40-keV mass-separated radioactive beam of $^{13}N^{1+}$ ions has been polarized using the technique of grazing surface scattering (GSS) at a Si single crystal. A nuclear polarization of 11% was obtained. It was detected by measuring the beta asymmetry after implantation of the beam in a Pt foil at a temperature of 20 mK. This result demonstrates the feasibility of GSS as a general method for the production of polarized radioactive beams. The technique can be applied to low-intensity beams and used for investigations ranging from symmetry tests in weak interactions to solid-state studies.

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We report the first direct observation of a nuclear polarization for a radioactive nucleus, induced with the grazing surface scattering method (GSS) [1]. The nuclear polarization is detected radioactively, thus allowing one to use the method with low intensity beams ($\leq$1 pA), contrary to the optical detection method that was used previously [1]. The technique offers new possibilities for the study of fundamental symmetries and weak processes in nuclear beta decay, as well as for solid-state studies with radioactive beams.

Over the years, a number of different techniques to polarize low energy beams of radioactive nuclei have been developed. All rely on the hyperfine coupling in free atoms for the transfer of an initially induced atomic polarization to the nucleus: e.g., laser-based methods [2,3], electron capture in polarized alkali vapors [4], tilted foil polarization (e.g., Refs. [5] and [6]), etc. Most of these techniques are, however, applicable only to a limited number of elements. This is not the case for the tilted foil (TF) technique, but it has the advantage of producing relatively low degrees of polarization (mostly of the order of 1% and seldom exceeding 5%) unless stacks of foils are used [6]. As a consequence, the TF technique is best suited for beams with relatively high energy (i.e., MeV/amu order of magnitude) (e.g., Ref. [7]). The method we use here, i.e., grazing surface scattering [1], is based on the same physical principle as TF: the atomic polarization results from the breaking of the axial symmetry during the beam-surface interaction [8]. However, in the case of GSS the beams are polarized by specular reflection (at incidence angles of the order of 1°) from very flat surfaces. The first experiments in which beams were scattered from surfaces at grazing incidence angles were carried out by Rau [9] with a 150-keV D^+ beam and a magnetized Ni surface. The GSS method as we use it here, with nonmagnetic surfaces and heavy ion beams, was pioneered by Andrä [10] and Berry *et al.* [11,12] and was further developed and generalized by Winter and co-workers [1,13–15]. The phenomenological model of Schröder and Kupfer [16] accounts quite well for the observed effects. In this model the electronic polarization results from an asymmetric Coulomb interaction between the beam ions and the surface electrons. This causes a transfer of angular momentum, the direction of which is determined by the cross product of the electron density gradient at the surface and the beam velocity. More detailed experiments showed that resonant electron transfer processes together with the specific geometry of the beam-surface interaction were responsible for the generation of the atomic polarization, as was explained by more recent theories as well [17,18].

Although it has been shown with stable beams that GSS is more powerful than TF to induce the highest atomic and nuclear polarizations for a large variety of elements [6,19], up to now it was never incorporated at an on-line isotope separator for applications with radioactive beams. Also, the nuclear polarization induced with GSS was always measured indirectly (see, e.g., Ref. [1]). This method, however, is subject to uncertainties in the transfer process of the polarization from the nucleus to the atomic shell if the atomic transitions are not known. In our case the polarization is observed directly by measuring the beta asymmetry.

Our experimental setup has been described before [20–22]. All measurements reported here were performed with 40-keV beams at the central beam line of the LISOL (Leuven Isotope Separator On Line) at the CYCLONE cyclotron in Louvain-la-Neuve. This beam line is equipped with electrostatic lenses and several sets of deflection plates for beam focusing and steering, ensuring maximal transmission efficiency. This is especially important for radioactive beams, since these have intensities of the order of pA to nA only. The scattering crystal is a Si(111) single crystal with dimensions of $1 \times 10 \times 50$ mm^3 and a surface granulation smaller than 20 nm. It is placed in the center of a UHV chamber where a vacuum of 4×10^{-10} Torr can be maintained in on-line conditions by differential pumping. In this chamber the beam ions are deflected electrostatically toward the crystal. Angles of incidence of the order of 1° are required to assure both a high polarization and a large specularly reflected beam component. The profile of the incoming beam can

be monitored with a two-dimensional grid in front of the crystal. The scattered beam profile is monitored with a movable needle placed at a few centimeters behind the crystal. The reflected beam is implanted in a Pt foil that is kept at low temperature (between 20 mK and 1 K) by a ^{3}He-^{4}He dilution refrigerator. This refrigerator is connected to the UHV chamber by a 10 cm long cooled side-access containing four diaphragms (diameter 5–6 mm), which reduces the heat input in the refrigerator and at the same time prevents nonspecularly reflected ions to reach the Pt implantation foil. The distance between the scattering surface and the Pt foil is about 35 cm. The cooling of this foil is of utmost importance for measurements with isotopes with half-lifes longer than several seconds, in order to preserve the nuclear polarization during their lifetime. We chose a Pt foil to reduce any magnetic hyperfine or quadrupolar interaction, which would speed up the relaxation of the polarized nuclei. Furthermore, a magnetic field could be applied in order to decouple any residual interactions after implantation. Finally, two Si-PIP (planar implanted passivated) detectors with an active area of 300 mm^2 and a thickness of 1 mm were mounted inside the 4 K shield of the refrigerator, at 0° and 180° with respect to the direction of the (horizontal) magnetic field, for measuring the beta asymmetry of the implanted polarized nuclei. The solid angle for each detector was about 1.9% of 4π.

Prior to the experiments with a radioactive beam, a large number of measurements were carried out with stable beams in order to test and characterize the setup. In these measurements the atomic polarization was determined by measuring the relative Stokes parameter $S/I = [I(\sigma^-) - I(\sigma^+)]/[I(\sigma^-) + I(\sigma^+)]$, where $I(\sigma^\pm)$ is the intensity of light with positive and negative helicity, which characterizes the circular polarization of the de-excitation light emitted by the surface-excited beam ions. Most of the tests were carried out with a stable $^{14}N^{1+}$ beam, for two reasons. Firstly, in recent years Winter and Andrä have acquired a wealth of data with this beam (see, e.g., Refs. [1,14,15]), so their results served as a reference. Secondly, as S/I is related to the nuclear polarization $P = \langle I_z \rangle / I$, knowledge of the atomic polarization for ^{14}N allows us to estimate the amount of nuclear polarization that can be expected for the radioactive ^{13}N (taking into account the different nuclear spin values). An optical detection system, consisting of a set of lenses, a $\lambda/4$ plate, a linear polarizer, and an interference filter for wavelength selection [22], was used to observe the circular polarization of the light emitted from the region immediately behind the scattering crystal. Table I lists the S/I values that were deduced for a number of singly and doubly charge beams. As can be seen, electronic polarizations of several tens of percents are quite generally obtained with the GSS method, irrespective of the type of element used. The large value of $S/I = 70\%$ that was obtained for a (80 keV) $^{16}O^{2+}$ beam (Fig. 1), without any wavelength selection, demonstrates

TABLE I. The values for the circular polarization S/I obtained for different stable beams with both the grazing surface scattering (GSS) and the tilted foil (TF) methods. The 567 nm wavelength corresponds to the N II $2s^2\,2p3s\,^3P$-$2s^2\,2p3p\,^3D$ optical transition, and the 500 nm wavelength to the transition N II $2s^2\,2p3p\,^3P$-$2s^2\,2p3d\,^3F^0$. The beam energy was 40 keV, except for the $^{16}O^{2+}$ beam where it was 80 keV.

Method	Beam	S/I (%)	Wavelength
GSS	$^{14}N^{1+}$	22(1) – 30(1)	567 nm
GSS	$^{12}C^{1+}$	11.5(5)	White light
GSS	$^{16}O^{1+}$	10.0(5)	White light
GSS	$^{16}O^{2+}$	70(3)	White light
GSS	$^{40}Ar^{1+}$	18(1)	White light
TF	$^{14}N^{1+}$	10.2(19)	500 nm
TF	$^{14}N^{1+}$	6.3(4)	567 nm

the possibilities of this setup. The values of 22% to 30% that we observed for the 567 nm line of $^{14}N^{1+}$ can be compared to the results of Freier and Winter [15]. They obtained $S/I \cong 50\%$ for a $^{14}N^{1+}$ beam of 40 keV. This larger value is most probably caused by the *in situ* cleaning of the scattering surface, since with an uncleaned surface Winter and Andrä also observed much smaller S/I values [23]. Moreover, it was only after sputtering the crystal for several hours under grazing incidence conditions with a 40-keV Ar^{1+} beam that we observed the value of $S/I = 30\%$, which again points to the importance of the preparation of the scattering surface. Finally, from the comparison of the S/I values we observed with the tilted foil method with those obtained with GSS (Table I), it is clear that (much) higher polarizations can be obtained with the grazing surface scattering method.

The formalism relating the observed circular polarization S/I of the fluorescent light to the nuclear polarization P that can be expected has been discussed before [16,24]. It turns out that the nuclear polarization is directly proportional to the S/I circular polarization: $P = CS/I$, with

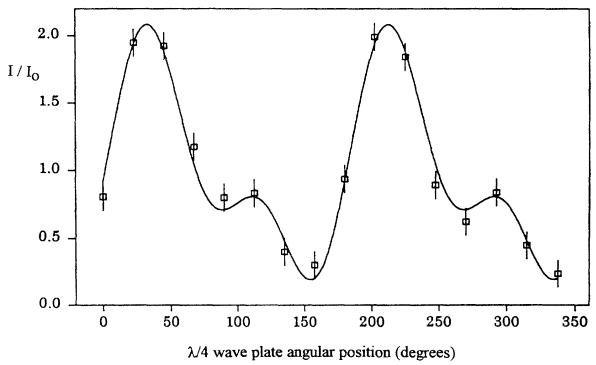

FIG. 1. Result of the optical measurement for the $^{16}O^{2+}$ beam. Shown is the normalized light intensity versus the position of the quarter wave plate. One clearly sees a main π periodicity with a small $\pi/2$ periodicity superimposed on it, corresponding [12] to the contributions of the circular (linear) polarization component in the de-excitation light.

the transfer coefficient C depending on the nuclear spin and on the spins of the atomic states involved. On the basis of this formalism, the S/I values of 22% to 30% we observed for ^{14}N correspond to an expected nuclear polarization of 7% to 10% in the case of ^{13}N. We chose this latter isotope for the first application of GSS to produce a nuclear polarized radioactive beam since it is abundantly produced at the radioactive ion beam facility that has recently become available in Louvain-la-Neuve [25]. Moreover, nitrogen is one of the elements for which no appreciable nuclear polarization can be induced with other polarization methods (e.g., $\ll 1\%$ with the classical low-temperature nuclear orientation method). The ^{13}N activity was produced by bombarding a ^{13}C target with a 30-MeV proton beam of 100 μA from the CYCLONE-30 cyclotron. It was transported in molecular form (i.e., as N_2) to an ECR-type ion source on the LISOL mass separator [26], through a 30 m long and 8 cm diameter stainless steel transfer tube. After being ionized, the ^{13}N ions were extracted from the ion source, accelerated, mass separated, and then transported to the surface interaction region in the central beam line. After optimization of the transport efficiency, a specularly reflected beam of up to 10^5 atoms/s was obtained, which was implanted continuously in the Pt foil in the refrigerator.

The directional distribution for allowed beta decay can be written as

$$W(\theta) = N(\theta)_{\mathrm{pol}}/N(\theta)_{\mathrm{unpol}} = 1 + (v/c)\,PAQ\cos\theta\,, \tag{1}$$

with $N(\theta)$ the count rate in the detector at angle θ with respect to the polarization axis, v/c (= 0.91) the velocity of the observed positrons with respect to the speed of light, P the nuclear polarization, A (= −0.333) the asymmetry parameter for the observed beta transition, and Q (= 0.98) a solid angle correction factor. Experimentally we determined the double ratio

$$W(0°)/W(180°) = [N(0°)/N(180°)]_{\mathrm{pol}}/[N(0°)/N(180°)]_{\mathrm{unpol}} = [1 + (v/c)\,PAQ]/[1 - (v/c)\,PAQ] \tag{2}$$

in order to correct for possible variations in the intensity of the implanted beam. To preserve the polarization of the ^{13}N nuclei for a sufficiently long time (compared to the ^{13}N half-life of 10 min), the temperature of the Pt implantation foil was kept as low as possible. Because of the heat load of a stable ^{13}C contaminant in the ^{13}N beam, this implantation temperature was 20 mK. From the systematics of the relaxation time for impurities in a cubic metal host lattice [27], one expects for nitrogen $g^2T_1T \approx 30$ s K, with g the nuclear g factor, T_1 the relaxation time, and T the temperature. For ^{13}N, $g = 0.64$, yielding $T_1 \approx 60$ min at 20 mK. Since this is 4 times longer than the ^{13}N lifetime, a sizable fraction of the nuclear polarization will be preserved during the lifetime of the nuclei. In order to have an unpolarized source, the implantation foil was warmed to about 0.7 K, at which temperature the relaxation time $T_1 \approx 2$ min, so that most of the nuclei had lost their polarization before decaying. This normalization method has the important advantage that one does not need to interfere with the beam transport, thereby assuring that any nonzero beta asymmetry is not caused by changes in the position of the implantation spot. Measurements were carried out with and without a magnetic holding field in the direction of the nuclear polarization. This holding field of 70 G was produced once with a superconducting split-coil magnet inside the refrigerator and once with an externally placed horseshoe type magnet, in order to check whether the magnetic field configuration had any influence on the nuclear polarization. The results are summarized in Table II. As can be seen, no significant polarization was observed in the absence of the holding field. However, since at 0.7 K the ratio $T_1/T_{1/2}$ is still about 0.2, it is not impossible that the polarization is not yet fully destroyed, even at this temperature, as might be suggested by the observed value of (1.9 ± 1.7)%. With a field of 70 G, an asymmetry of 6.5%, implying a nuclear polarization of 11%, was obtained with both magnets. Finally, although we measured the time evolution of the beta asymmetry, statistics unfortunately did not allow an accurate determination of the relaxation time of N in Pt. We could, however, set the following limits: $7 < g^2T_1T < 178$ s K. These are in good agreement with the value of 30 s K mentioned above and also with the experimental result for B in Pt [i.e., $g^2T_1T = 268(10)$ s K, from a measurement with ^{12}B in Pt [28]], for which element systematics predict the value to be about two times larger than for N.

The measurements presented here complete the first part of the project aiming at the realization of a dynamical polarization setup at the radioactive ion beam facility in Louvain-la-Neuve. Although, as was discussed, the experimental conditions of the measurements reported here can still be improved, we have clearly shown the usefulness of the GSS method for polarizing radioactive beams. Indeed, as was shown with ^{13}N, the GSS method

TABLE II. The beta asymmetry $[W(0°)/W(180°)] - 1$ and nuclear polarization P obtained for a 40-keV ^{13}N beam using the GSS method.

Holding field (G)	$[W(0°)/W(180°)] - 1$	P (%)
0	−0.011(10)	+1.9(17)
70[a]	+0.068(10)	−11.1(16)
70[b]	+0.065(11)	−10.6(17)

[a]With superconducting split-coil magnet.
[b]With externally placed horseshoe type magnet.

allows us to obtain substantial polarizations for nuclei for which no or only a small polarization can be obtained with other techniques. Moreover, our value of 11% is much higher than the results that were previously obtained with the tilted foil method at the Rochester recoil separator (with ^{33}Cl [29]) and at the Daresbury and ISOLDE mass separators (with ^{35}Ar and ^{23}Mg, respectively [30]). For ^{33}Cl a beta asymmetry of 0.2%–0.3% was observed, while for ^{35}Ar and ^{23}Mg 0°/180° beta anisotropies up to about 0.8% were obtained, all corresponding to nuclear polarizations of less than 1%.

Applications of this new development are, on the short term, mainly situated in the study of fundamental symmetries in nuclear beta decay, in succession of the recently reported experiments [31,32]. Furthermore, the universality of GSS makes it a very efficient tool for studies with polarized beams in various other fields of physics as well, especially when it can be installed at a facility where large amounts of short-lived nuclei are readily available. In this respect, its installation at the ISOLDE isotope separator facility in CERN, which is being planned now, would allow an optimal exploitation of this technique.

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