Eur. Phys. J. A (2011) **47**: 138
DOI 10.1140/epja/i2011-11138-8

THE EUROPEAN
PHYSICAL JOURNAL A

Letter

Spin-polarization of radioactive $^{123}In_{g.s.}$ by the tilted-foil method

Y. Hirayama[1,a], M. Mihara[2], Y.X. Watanabe[1], S.C. Jeong[1], N. Imai[1], K. Matsuta[2], H. Miyatake[1], T. Hashimoto[3], H. Ishiyama[1], S. Ichikawa[3], T. Ishii[3], T. Izumikawa[4], I. Katayama[1], H. Kawakami[1], I. Nishinaka[3], K. Nishio[3], H. Makii[3], S. Mitsuoka[3], S. Momota[5], A. Osa[3], Y. Otokawa[3], T.K. Sato[3], and Y. Wakabayashi[3]

[1] Institute of Particle and Nuclear Studies, High Energy Accelerator Research Organization (KEK), Ibaraki 305-0801, Japan
[2] Department of Physics, Osaka University, Osaka 560-0043, Japan
[3] Japan Atomic Energy Agency (JAEA), Ibaraki 319-1195, Japan
[4] Radioisotope Center, Niigata University, Niigata 951-8510, Japan
[5] Kochi University of Technology, Kochi 782-8502, Japan

Received: 31 August 2011 / Revised: 13 October 2011
Published online: 17 November 2011

Communicated by B.R. Fulton

Abstract. A spin-polarized radioactive ^{123}In ($I^{\pi} = 9/2^{+}$, $g = 1.220(2)$, $T_{1/2} = 5.97(5)$ s) beam has been successfully generated by the tilted-foil method. This nuclide is the heaviest ever polarized in its ground state by this method. Using the ISOL-based re-acceleration–type facility TRIAC, an $^{123}In_{g.s.}$ beam of 305 keV/nucleon went through a stack of 15 carbon foils with a tilt angle of 70°, and an asymmetry of $0.76 \pm 0.25\%$ of β-decays was observed by the β-NMR technique. The asymmetry shows that the tilted-foil method combined with a re-acceleration facility is useful for producing spin-polarized beams for applications such as nuclear physics and materials science.

Spin-polarized radioactive isotope beams (RIBs) provide an opportunity to explore nuclear structure through β-decay spectroscopy as well as probing the electromagnetic properties of materials using the β-NMR technique. The tilted-foil (TF) method [1] is one of the most useful techniques for producing spin-polarized RIBs, because the method can be applied in principle to any element of short-lived RIB generated at isotope separation on-line (ISOL)-based re-acceleration–type facilities, except for the nuclei with $I = 0$ and/or a lifetime shorter than a few nanoseconds.

The process of nuclear polarization by the TF method is considered as follows: First, atomic spin-polarization is introduced through anisotropic atomic collisions of incident ions with electrons at the exit surface of the foil. Then, the atomic spin-polarization is transferred to nuclear spin-polarization via the hyperfine interaction during flight in vacuum. From the successful polarization of ^{147}Gd nuclei in isomeric states with a half-lifetime of 26.8 ns [2], the time scale for the production of nuclear polarization by the TF method can be inferred to be fast (the order of nanoseconds). RIBs of a few hundred keV/nucleon are considered to be most suitable for the effective capture of electrons, which is essential for the primary atomic polarization [2,3].

[a] e-mail: yoshikazu.hirayama@kek.jp

The induced nuclear polarization by using multi-tilted foils increases with increase of the number of foils and saturates according to a classical model equation [4] as follows:

$$P'_I(N) = P'_I(\infty)\left\{1 - (1 - \frac{P'_I(1)}{P'_I(\infty)})^N\right\}, \quad (1)$$

$$P'_I(1) = P'_J P(I, J),$$

$$P(I, J) = \frac{1}{4\lambda^2}\left\{2\lambda + (\lambda^2 - 1)\ln(\frac{1+\lambda}{1-\lambda})\right\}, \quad \lambda = \frac{2IJ}{I^2 + J^2},$$

$$P'_I(\infty) = \frac{P'_J}{P'_J + (1 - P'_J)J/I}.$$

Here N is the number of foils, I and J are nuclear and averaged atomic spins, respectively. The atomic and nuclear polarizations are conveniently expressed by $P'_S = P_S\sqrt{(S+1)/S}$ ($S = I, J$) using the usual polarization P_S in these equations. Nuclear polarization could be estimated by using the eq. (1) with a reasonable averaged atomic spin J and the atomic polarization P_J. The degree of polarization also increases with increasing the tilt angle ϕ between the beam axis ($\overrightarrow{v}$) and the axis normal ($\overrightarrow{n}$) to the foil surface [5]. The direction of polarization, which is well defined by $\overrightarrow{n} \times \overrightarrow{v}$, is easily reversed by reversing the normal axis of the foil surface [5]. Thus, the nuclear polarization produced by the TF method is controllable and the most suitable conditions for maximizing the polarization of each RIB can be achieved.

The TF method has been applied in various experiments. For the application to isomeric states with short lifetime $T_{1/2} \approx 10$ μs, reviewed in ref. [2], the signs of nuclear quadrupole moments in high-spin isomers such as ^{54}Fe ($T_{1/2} = 357$ ns) and ^{147}Gd ($T_{1/2} = 22.2, 26.8$ and 510 ns) were determined by the time differential perturbed angular distribution (TDPAD) method. As reported in ref. [3], the TF method was also extended to the study of the electromagnetic moments of radioactive light nuclei in ground states with $T_{1/2} < 10$ s in combination with the β-NMR technique. The polarization of radioactive heavy nuclei in ground states, however, has not yet been reported. This is because high-quality RIBs accelerated up to a few hundred keV/nucleon, which is essential to achieve higher nuclear polarization by using multiple foils, have only become available recently.

We report here the production of a spin-polarized ^{123}In beam by the multi-TF method at the ISOL-based re-acceleration–type facility TRIAC [6].

The TF method for producing spin-polarized RIBs has been developed at the TRIAC for the study of nuclear structure through the measurement of electromagnetic moments and the determinations of spins and parities in the ground and excited states [7] around the doubly magic nucleus ^{132}Sn. As the first step in the development of the TF method, we successfully produced a 7.3 ± 0.5% polarized ^{8}Li ($T_{1/2} = 838$ ms) beam [8]. Investigating the nuclear polarization of ^{8}Li as a function of the number of foils, incident energies and tilt angles, we confirmed the above-mentioned features of TF method experimentally and found that lower beam energy is desirable for the production of ^{8}Li nuclear polarization, where, after passing through more than 15 foils, ^{8}Li can be implanted into appropriate depth of the catcher material for the β-NMR technique. To establish the TF method for heavy nuclei, as the next step, the ^{123}In$_{\text{g.s.}}$ ($I^{\pi} = 9/2^{+}$, $g = 1.220(2)$, $T_{1/2} = 5.97(5)$ s, $Q_{\beta} = 4.391$ MeV) [9–11] nucleus was chosen for the following reasons. First, nuclear polarization of ^{123}In ($Z = 49$) could be expected to be about 9% using eq. (1) and referring to the experimental result from the production of polarized excited state of Pd ($Z = 46$) nuclei [12] by the TF method. Second, the g-factor of ^{123}In$_{\text{g.s.}}$ has been measured by laser spectroscopy [10] and hence the RF frequency of β-NMR is known. Third, the catcher material (semiconducting indium) to preserve the nuclear polarization during the lifetime was investigated previously by β-NMR of ^{116}In [13]. Therefore, we could focus our attention on the nuclear polarization by the TF method.

In the present experiment, a stack of 15 carbon foils was used for polarizing the ^{123}In nuclei with energy 305 keV/nucleon (velocity $v/c = 0.026$). This is because the expected nuclear polarization of ^{123}In ($Z = 49$) nuclei is as high as about 9%, as shown in fig. 1, using an atomic polarization 3% of Pd nuclei ($Z = 46$) with velocity $v/c = 0.029$ [12] and an averaged atomic spin $J = 5/2$, which is reasonable approximation in these mass

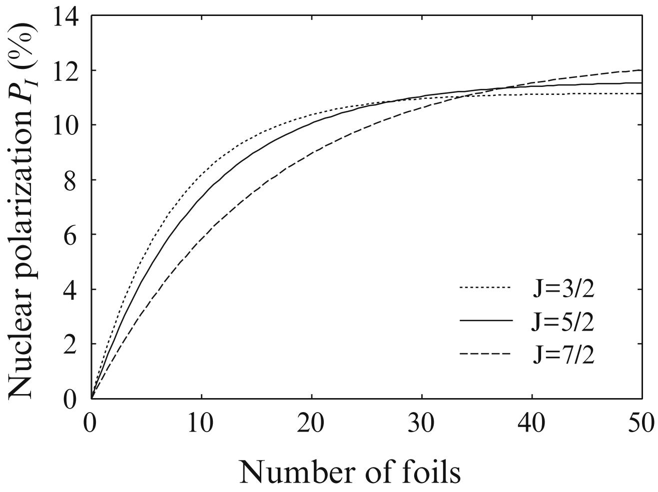

Fig. 1. Calculated nuclear polarization as a function of the number of the foils by using eq. (1) with the atomic polarization of 3%. Here, the dotted, solid and dashed lines were calculated using the averaged atomic spin $J = 3/2$, 5/2 and 7/2, respectively.

region [12]. Depending on the averaged atomic spin J, the induced nuclear polarization is varied at the same number of foils. The number of foils to achieve saturated polarization also depends on the atomic spin J. In the present case, several percent polarization of ^{123}In with 15 foils would be expected, even though the averaged atomic spin J is less than the approximated value of $J = 5/2$. In addition, the outgoing beam energy through the 15 carbon foils was adjusted to be about 175 keV/nucleon with the straggling of 20 keV/nucleon, and the implantation depth in the InAs crystal was estimated by the SRIM-2008 code [14] to be about 5 μm, which would be sufficient to preserve nuclear polarization.

The ^{123}In nuclei were produced by proton-induced fissions of a $^{\text{nat}}$UC$_{\text{x}}$ target (0.6 ∼ 0.8 g/cm^{2}), which was bombarded by a proton beam with an energy of 34 MeV. The extracted ^{123}In^{+} was mass-separated and, after being charge-bred to 16^{+}, accelerated to 305 keV/nucleon by the linac complex of TRIAC. Typical beam intensities of ^{123}In at the entrance of a stacked tilted multi-foils were about 2×10^{3} particles per second. The beam purity was about 90% and the dominant contamination was a β-decaying isomer of ^{123}In ($E_{\text{x}} = 327.21$ keV, $I^{\pi} = 1/2^{-}$, $g = 0.800(8)$, $T_{1/2} = 47.8(5)$ s) [9–11] identified by β-delayed γ-rays measured by a germanium detector.

A schematic side view of the experimental setup and top view of the stacked tilted foils are presented in fig. 2(a) and (b), respectively. The accelerated ^{123}In beam was polarized after passing through the stacked tilted foils and was implanted into the catcher material cooled by a refrigerator for longer spin-lattice relaxation time, where a magnetic field B_0 was applied to preserve the nuclear polarization. To apply the β-NMR technique, the nuclear polarization was manipulated by a radio frequency (RF) magnetic field B_1 induced by RF coils and was measured by detecting β-rays in β-ray telescopes.

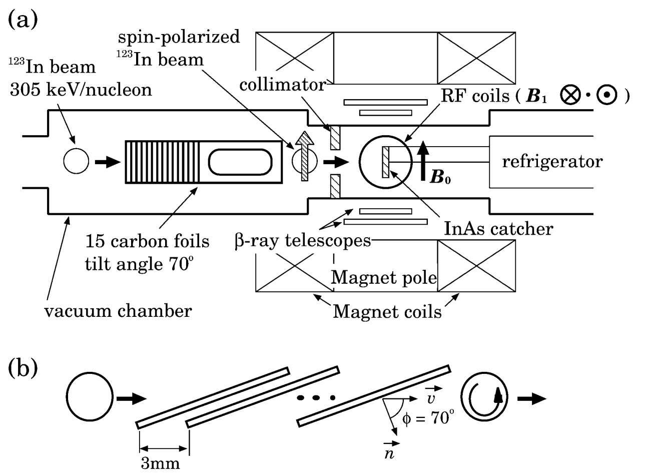

Fig. 2. Schematic side view (a) of the experimental setup and top view (b) of the stacked tilted foils. See the text for more details.

A stack of 15 carbon foils was placed in a vacuum chamber and the exit of the stacked foils was located 145 mm upstream from the catcher material. Each of the foils on the non-magnetic copper frame was $10\,\mu$g/cm^2 thick and was tilted at an angle of 70°. The direction of nuclear polarization, which is perpendicular to the beam direction as shown in fig. 2(a), could be easily changed to be up ($+P$) or down ($-P$) by reversing the surface direction of the stacked foils (by using a stepping motor outside the vacuum chamber). The distance between each foil was 3 mm along the beam axis, as shown in fig. 2(b). The area of each carbon foil was $45\,\mathrm{mm}\times 15\,\mathrm{mm}$, which corresponds to $15\,\mathrm{mm} \times 15\,\mathrm{mm}$ at the tilt angle of 70° as viewed from the beam direction. The area is about twice as large as the beam spot, about 8 mm in diameter, at the entrance of the stacked foils, and is designed to achieve more than 90% transmission through 15 carbon foils under the effect of multiple scattering in each carbon foil. The beam spot on the catcher material was calculated to be about 18 mm in diameter considering the effect of the multiple scattering [14] and the collimation by the thick stainless-steel collimator (opening area $15\,\mathrm{mm} \times 15\,\mathrm{mm}$) placed 100 mm upstream from the catcher material. β-rays emitted from the ^{123}In nuclei stopped on the collimator were prevented from hitting the β-telescope.

The spin-polarized ^{123}In beam was implanted into a cubic semiconductor crystal of InAs (N-type) ($20\,\mathrm{mm} \times 20\,\mathrm{mm} \times 0.45\,\mathrm{mm^t}$) at a temperature of about 18 K to reduce the spin-lattice relaxation in the InAs crystal, where a magnetic field $B_0 \sim 0.5\,\mathrm{T}$ was applied to preserve the nuclear polarization. The expected spin-lattice relaxation time T_1 of ^{123}In at $T \sim 20\,\mathrm{K}$ is about 27 s from the ^{116}In case [13], which is much longer than the lifetime of the radioactive isotope ^{123}In.

The nuclear polarization was measured by the depolarization method [15] of the β-NMR technique. A RF magnetic field B_1 for NMR, the direction of which was perpendicular to B_0 and the beam direction, was induced for the depolarization by an RF coil fixed around the InAs crystal in a vacuum chamber, as shown in fig. 2(a). In the experiment, the defect-induced broadening of the NMR linewidth is considered to be wide due to the large Q-moment of 0.757(9) b [10] and difficulties in terms of the B_1 strength in reversing the nuclear polarization by the adiabatic fast passage (AFP) technique of NMR [15]. Therefore, the nuclear polarization was destroyed by applying a weak B_1 of amplitude $\sim 0.3\,\mathrm{mT}$. The applied RF frequency $\nu_0 \pm \Delta\nu_Q = 4649 \pm 200\,\mathrm{kHz}$ corresponding to the Larmor frequency ν_0 and the resonance width $2\Delta\nu_Q$ were estimated from the NMR linewidth of the ^{116}In in InAs crystal [13] and the ratio of the Q-moments of $Q[^{123}\mathrm{In_{g.s.}}]/Q[^{116}\mathrm{In_{g.s.}}] \sim 84$. The duration of the RF sweep from $\nu_0 - \Delta\nu_Q$ to $\nu_0 + \Delta\nu_Q$ was 20 ms. The probability for reversing the nuclear polarization was calculated to be less than 85% in a single RF sweep [15]. Therefore, a set of 24 sweeps in a total of 480 ms would reduce the initial nuclear polarization by a factor $< 1/50$ and thus destroy the nuclear polarization.

Double-layered plastic scintillator telescopes in the atmosphere, placed above and below the InAs crystal, were used for the detection of the emitted β-rays, and the thickness of each scintillator was 1 mm. The β-rays with $Q_\beta = 3.2\,\mathrm{MeV}$ had a mean energy of 1.1 MeV after they passed through the 0.3 mm thick stainless-steel vacuum window and were detected by β-telescopes. The efficiency of the β-ray detection calculated by a Monte Carlo simulation with the GEANT3 code [16] considering the experimental setup was 9% for the ^{123}In nucleus.

β-rays emitted from a polarized nucleus have an angular distribution of $\mathrm{W}(\theta) \sim 1 + AP\cos\theta$. Here, θ, A and P are the emission angle with respect to the nuclear polarization axis, the asymmetry parameter of β-transition and the nuclear polarization of the parent nucleus, respectively. For the $^{123}\mathrm{In_{g.s.}}$ nucleus, an asymmetry parameter of $A \sim -0.96$ was obtained from the weighted average of dominant β-decay transitions reported in ref. [17]. The asymmetry $AP = (R'/R - 1)/(R'/R + 1)$ was obtained from the up/down ratio $R = W(0°)/W(180°)$ of the β-ray counting rates with the RF field ($P = 0$: depolarization) and R' without the RF field (P), which are expressed as

$$R \sim \frac{f_{\mathrm{up}}}{f_{\mathrm{down}}}, \qquad (2)$$

$$R' \sim \frac{f_{\mathrm{up}}(1 + AP)}{f_{\mathrm{down}}(1 - AP)}, \qquad (3)$$

where $f_{\mathrm{up}}/f_{\mathrm{down}}$ is a constant factor derived from the solid angles and efficiencies of the telescopes.

One measurement cycle consisted of two consecutive beam cycles (beam on and off) for the R and R' measurements, respectively, as shown in fig. 3(a). In each cycle, the beam was chopped at JAEA-ISOL with beam-on and -off periods of 8 s and 10.48 s. Typical time spectrum obtained from β-ray events with the beam cycle is shown in fig. 3(b). The spectrum was fitted using exponential function for the ground states of ^{123}In and a constant background including the β-decay of the isomer state. The obtained half-lives for the ground state was $T_{1/2} = 5.8 \pm 0.2\,\mathrm{s}$ which was

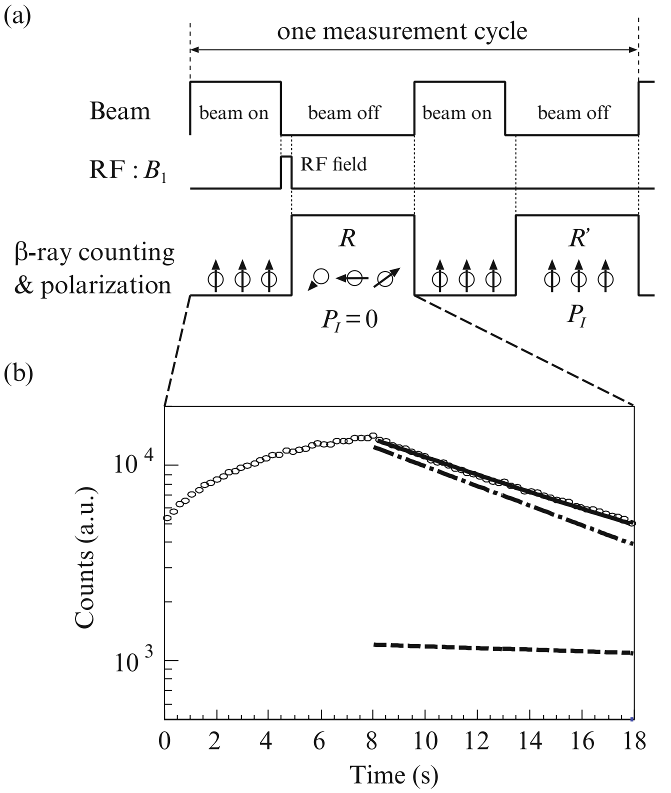

Fig. 3. (a) Time sequence for measuring the nuclear polarization by the depolarization method of the β-NMR technique. (b) Typical time spectrum of β-ray events with the beam cycle. Open circles show the counting rates of the β-ray and the statistical error bars are smaller than the open circles. The solid line shows the fitted result with the β-decay of the ^{123}In ground state (dash-dotted line) and the background (dashed line) including the β-decay of the ^{123}In isomer state.

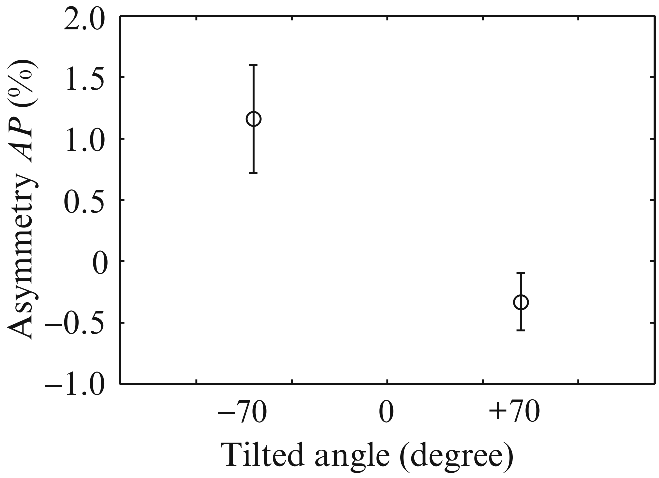

Fig. 4. Asymmetries of emitted β-rays from ^{123}In measured at tilt angles of $\pm 70°$, reversing the stacked tilted foils.

good agreement with the literature value [11]. In the case of the R measurement cycle, 24 sweeps of the frequency modulated RF field were applied during 0.48 s from the beginning of the beam-off period to destroy the nuclear polarization completely. Then, the β-rays were counted during the following 10 s and the R value was obtained. In the same way, the R' value was measured without the RF field. The set of beam cycles was repeated many times to obtain sufficient counting statistics.

The asymmetries AP of emitted β-rays from ^{123}In were measured to be $-0.33\pm0.23\%$ and $+1.16\pm0.44\%$ at tilt angles of $\pm 70°$, reversing the stacked tilted foils, respectively, as shown in fig. 4. Although the measured AP values may appear asymmetric, these are in agreement within the statistics errors. We are confident in this assertion because the much smaller asymmetries AP of ^{8}Li using the same experimental setup and procedure, but with much better statistics, were nicely in agreement: $-0.464 \pm 0.037\%$ and $+0.455 \pm 0.038\%$ at tilt angles of $\pm 70°$ [8]. This precise result gives confidence that the ^{123}In nuclei were indeed polarized by the TF method. From the ratio between the values of $(R'/R)_{+70°}$ and $(R'/R)_{-70°}$, the asymmetry $|AP| = 0.76 \pm 0.25\%$ with the RF field was deduced with an accuracy of 3.0 times the standard deviation (σ).

The degree of nuclear polarization $|P| = 1.63 \pm 0.54\%$ was evaluated considering the correction factors ($f_{\rm solid} \times f_{\rm relax} \times f_{\rm bg} \times A = -0.47$) for the measured β-ray asymmetry AP: the measured polarization is attenuated by these correction factors $f_{\rm solid}$, $f_{\rm relax}$ and $f_{\rm bg}$ originating from the finite solid angles of the telescopes ($f_{\rm solid} = 0.80$), relaxation of polarization during the measurement time ($f_{\rm relax} = 0.76$) and the background β-rays from daughter nuclei and isomer contaminations ($f_{\rm bg} = 0.80$), respectively. The factor $f_{\rm solid}$ was estimated by a Monte Carlo simulation with the GEANT3 code [16] considering the experimental setup. The factor $f_{\rm relax}$ can be expressed as $1/(1 + T_{1/2}/(T_1 \ln 2))$ in the case of $T_1 \gg T_{1/2}$. The factor $f_{\rm bg}$ was evaluated from the ratio of β-ray counting rates of the $^{123}{\rm In}_{\rm g.s.}$ nuclei to the background rate, which was obtained from fitting the decay curve (fig. 3(b)) during the polarization measurement.

The degree of polarization $1.63 \pm 0.54\%$ was about a factor of 5 smaller than the value of about 9% expected using eq. (1). The reason for this could be due to the spin-relaxation phenomena in the catcher materials for the β-NMR technique. The measured nuclear polarization in ionic crystals is often smaller than that in metallic catcher materials, where the induced nuclear polarization is precisely measured in many cases. For example, in the case of ^{20}Na nuclei, the degree of polarization induced by laser optical pumping were measured in an ionic crystal MgF_2, and the measured polarization was about six times smaller than that in a platinum catcher [18]. In the present experiment, the nuclear polarization primarily induced by the TF method could be as large as the expected values of 9%. However, the measured nuclear polarization in ionic InAs crystal, which was reported as a good catcher material to preserve the nuclear polarization of indium isotopes [13], would be about a factor of 5 smaller than the primarily induced polarization. The same spin-relaxation may occur in the implantation into the InAs crystal, but the mechanism has not been fully resolved so far. In addition, the relaxation time of the polarization in the InAs crystal might not be as long as the value in the present case suggested by ref. [13] because of the different implantation sites in the InAs crystal from the experiment in ref. [13]. In the experiment of ref. [13], the polarized ^{116}In nuclei were produced from the capture of polarized thermal neutrons by ^{115}In nuclei in an InAs crystal, so that the sites occupied by the ^{116}In nuclei were expected to be the original cubic sites (substitutional sites in the case of ^{123}In implantation). In the present case, many energetic ^{123}In nuclei might be implanted into interstitial sites in the InAs crystal, where the observed polarization is about 30% smaller than that of substitutional sites due to spin-relaxation and/or splitting of the resonant RF frequency caused by a local electric-field gradient [19]. Even though the energetic beam used in the TF method could reduce the degree of polarization, it is not a dominant factor for the polarization presently measured to be smaller than anticipated. A study of catcher materials would be essential for further applications with much higher degrees of polarization.

In summary, we have produced spin-polarized radioactive ^{123}In in the ground state for the first time by the tilted-foils method at the ISOL-based re-acceleration–type facility TRIAC. The obtained polarization by the TF method was about 1%, which can be used for the study of nuclear structure and the electromagnetic properties of materials. Even though searching for catcher materials with spin-relaxation time much longer than the lifetime of nuclei of interest is essential for further applications, we have successfully demonstrated the usefulness of the TF method combined with ISOL-based re-acceleration–type facilities, even for polarizing heavy elements.

The authors acknowledge the staff of the JAEA tandem accelerator for their support.

References

1. H.G. Berry, M. Hass, Annu. Rev. Nucl. Part. Sci. **32**, 1 (1982) and references therein.
2. G. Goldring, Hyperfine Interact. **33**, 19 (1987).
3. Y. Nojiri, Hyperfine Interact. **100**, 23 (1996).
4. G. Gorldring, Y. Niv, Hyperfine Interact. **21**, 209 (1985).
5. N. Tolk *et al.*, Phys. Rev. Lett. **47**, 487 (1981).
6. Y.X. Watanabe *et al.*, Eur. Phys. J. ST **150**, 259 (2007).
7. Y. Hirayama *et al.*, Phys. Lett. B **611**, 239 (2005).
8. Y. Hirayama *et al.*, submitted to Eur. Phys. J. A.
9. C.V. Weiffenbach, R. Tickle, Phys. Rev. C **3**, 1668 (1971).
10. J. Eberz *et al.*, Nucl. Phys. A **464**, 9 (1987).
11. B. Grappengigsser *et al.*, *Proceedings of the International Conference on the Properties of Nuclei far from the Region of Beta Stability (Leysin, Switzerland)* CERN-report 70-30, 1093 (1970).
12. E. Dafni *et al.*, Phys. Rev. C **25**, 1525 (1982).
13. A. Winnacker *et al.*, Z. Phys. **244**, 289 (1971).
14. J.F. Ziegler, `http://www.srim.org/`.
15. A. Abragam, *The Principles of Nuclear Magnetism* (Clarendon, Oxford, 1961).
16. CERN program library.
17. B. Fogelberg *et al.*, Z. Phys. A **276**, 381 (1976).
18. K. Minamisono *et al.*, Nucl. Instrum. Methods A **616**, 45 (2010).
19. G.D. Morris *et al.*, Phys. Rev. Lett. **93**, 157601 (2004).