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# SELECTIVE LASER IONIZATION OF N $\geq$ 82 INDIUM ISOTOPES: THE NEW r-PROCESS NUCLIDE $^{135}$In

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## ABSTRACT

Production yields and beta-decay half-lives of very neutron-rich indium isotopes were determined at CERN/ISOLDE using isobaric selectivity of a resonance-ionization laser ion-source. Beta-delayed neutron multiscaling measurements have yielded improved half-lives for 206(6) ms $^{132}$In, 165(3) ms $^{133}$In and 141(5) ms $^{134}$In. With 92(10) ms $^{135}$In, a new r-process nuclide has been identified which acts as an important 'waiting-point' in the In isotopic chain for neutron densities in the range $n_n \simeq 10^{24}$–$10^{26}$ n/cm$^3$, where the r-matter flow has already passed the A $\simeq$ 130 abundance-peak region.

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# 1 INTRODUCTION

The region around the neutron-rich, double-magic isotope $^{132}$Sn has been the subject of intensive experimental investigations in recent years. The reason for this interest is in principle twofold. First, the evolution of the single-particle structures over nearly 40 mass units between nuclides with N/Z $\simeq$ 1.0 and isotopes with N/Z $\geq$ 1.6 provides an opportunity to develop a unique microscopic approach to nuclear structure with predictive power towards nuclei at the particle drip lines which are not currently accessible for experimental study. The second motivation is related to the rapid neutron-capture (r-process) nucleosynthesis of elements beyond iron. The solar-system r-abundances ($N_{r,\odot}$) in the A $\simeq$ 130 peak region reflect one of the three neutron-magic numbers (N = 50, 82 and 126), where the r-matter flow is delayed by climbing up the staircases at the shell closure from presumably $^{125}$Tc (Z = 43) to $^{130}$Cd (Z = 48) which act with their relatively 'long' $\beta$-decay half-lives as classical waiting-point nuclei. Having reached the top of the $N_{r,\odot}$ peak, the r-process is able to escape the A $\simeq$ 130, N = 82 'bottle-neck' area in the In (Z = 49) isotopic chain and speeds up again towards the rare-earth region. Hence, the odd-mass, even-N indium nuclei $^{131}$In, $^{133}$In and $^{135}$In are the most important waiting-point isotopes in the high-mass wing of the $N_{r,\odot}$ peak. For recent reviews about the nuclear structure and astrophysical importance of the $^{132}$Sn region, see for example [1, 2, 3].

In this paper, we report first results of a study of the decay of the N $\geq$ 82 isotopes $^{132-135}$In obtained with considerably enhanced ionization efficiency by laser ionization at CERN/ISOLDE. Beta-delayed neutron measurements were used to determine In production yields and half-lives.

# 2 PRODUCTION

## 2.1 Converter target

Neutron-rich medium-mass nuclei are normally produced at ISOLDE by high-energy (1 or 1.4 GeV) proton-induced fission of $^{238}$U. However, with this production mechanism also neutron-deficient isobars will be formed by spallation and high-energy fission. The situation is particularly complicated in the mass regions A $\simeq$ 80 and 130, where the weakly produced exotic neutron-rich nuclei of interest (e.g. the isotopes around $^{78}$Ni and $^{132}$Sn) are covered by many orders of magnitude higher background of the surface-ionized and therefore difficult to suppress proton-rich isobars of rubidium and caesium, respectively. Fission induced by low- to medium-energy neutrons, however, does not create this problem. Such neutrons can be produced efficiently by high-energy proton spallation of heavy target materials, e.g. tantalum or tungsten. The use of a 'mini-spallation-neutron-source' (called 'converter') surrounded by a concentric ISOL fission target was first proposed by Nolen et al. [4]. While the realization of a concentric target requires sophisticated engineering, it is relatively easy to build a reduced version by just installing a heavy-metal rod close to a standard ISOLDE fission target. In this experiment, a 10 mm diameter tantalum rod of 150 mm length was mounted parallel to the $^{238}$U target at a distance of 21 mm axis-to-axis [5]. Only part of the neutrons produced in the converter will impinge on the ISOL target, but the primary beam and most of the high-energy secondary particles are located in a forward cone not hitting the target.

For this experiment a standard ISOLDE UC$_x$/graphite target [6] with 50 g/cm$^2$ $^{238}$U and about 10 g/cm$^2$ carbon was used. The target was kept at 2150 °C and the niobium ionizer line at 1850 °C. Pulses of 1.4 GeV protons (5.3 $\mu$C each) were hitting the tantalum converter every 2.4 s.

### 2.2 Ionization

Indium (with an ionization potential of 5.79 eV) is already surface ionized. But, to further enhance the ionization efficiency, the ISOLDE Resonance Ionization Laser Ion Source (RILIS) [7, 8, 9] was tuned to excite In in two steps: with a frequency-doubled dye-laser beam of 303.9 nm from the ${}^2P^o_{1/2}$ atomic ground state to the ${}^2D^o_{3/2}$ excited state, and then with the green and yellow copper vapour laser beams (510.6 nm and 578.2 nm) non-resonantly to the continuum. This RILIS excitation scheme had been developed at Troitsk [10] and was already used at ISOLDE to ionize the neutron-deficient isotopes ${}^{100-114}$In [11].

The laser beams were focused into the ion-source cavity, which was made from a niobium tube of 30 mm length and 3 mm internal diameter. The average power of the laser beams delivered to the ion source were 50 mW and 4 W, respectively, for the first and second steps. The line-width of the dye-laser radiation was rather broad, about 10 GHz, allowing the operation of RILIS with the same laser wavelengths for different isotopes without any correction for the hyperfine structure or the isotopic shifts.

With this setup, the use of the RILIS allowed the In yields to be increased by about a factor seven compared to pure surface ionization. The absolute ionization efficiency could not be determined in this experiment, but it is expected to be several per cent. Since the target was not equipped with a stable indium 'mass marker', the RILIS had to be tuned 'blindly', therefore not necessarily guaranteeing optimum efficiency. Under standard operation conditions (more copper vapour laser power focused to the ionizer, and simultaneous excitation of the thermally populated ${}^2P^o_{3/2}$ state with a second dye-laser [11]), the ionization efficiency could probably be further enhanced by at least another factor three.

### 2.3 Yields

Table 1 shows the observed yields of the most neutron-rich indium isotopes deduced from the measured beta (${}^{130,132}$In) and beta-delayed neutron ($\beta$dn) (${}^{133-135}$In) activities, respectively. For ${}^{133}$In the branching ratio for $\beta$dn emission ($P_n = 85\%$) was taken from Ref. [12] and for ${}^{134,135}$In we used the theoretical $P_n$ values of 93% and 95%, respectively, obtained from our QRPA calculations (see below). All quoted yields were integrated over the complete release curve and are normalized to 1 $\mu$C of primary protons as defined in Ref. [13].

Table 1: Yields of heavy indium isotopes from a 50 g/cm$^2$ UC$_x$/graphite target with the RILIS tuned for indium ionization. The yield $Y$ is given in ions per $\mu$C of primary proton beam [13] hitting the tantalum 'converter' rod.

| Isotope | ${}^{130}$In | ${}^{132}$In | ${}^{133}$In | ${}^{134}$In | ${}^{135}$In |
|---|---|---|---|---|---|
| $Y$ ($\mu$C$^{-1}$) | $> 3.5 \cdot 10^5$ | 8000 | 900 | $\approx 95$ | $\approx 2.4$ |

## 3 RESULTS AND DISCUSSION

### 3.1 Half-lives

After laser ionization and mass separation, the isotopically clean beams of In nuclides were transported to a beam line equipped with a moving tape system where $\beta$dn measurements could be performed. Counting took place directly at the point of deposit,

Table 2: Comparison of experimental beta-decay half-lives for $^{132-135}$In with literature values and QRPA predictions for Gamow–Teller decay [18, 19]. For further discussion, see text.

| Mass | Half-life [ms] | | | | |
|---|---|---|---|---|---|
| | This work | Lit. | Ref. | QRPA | cQRPA |
| 132 | 206(6) | 201(13) | [15] | 96 | 212 |
| 133 | 165(3) | 180(20) | [15] | 141 | 245 |
| 134 | 141(5) | 138(8 ) | [21] | 99 | 190 |
| 135 | 92(10) | – | – | 90 | 251 |

and the tape system was used to remove the activities of the longer-lived daughter nuclides. Because the In half-lives being sought are in the 100-ms range, data acquisition in the system was initiated by the CERN-PSB proton pulses, separated by a multiple of 2.4 s, and continued for 1.6 s for each cycle.

Beta-delayed neutron data were collected by multiscaling measurements using the high-efficiency Mainz $4\pi$ $^3$He neutron long-counter. This detector was equipped with 64 $^3$He proportional counters arranged in three concentric rings. The $\beta$dn multiscaling data were analysed with a multi-component least-squares fit to obtain the In half-lives by taking into account the known $\beta$dn contributions from the Sn daughter products at masses A and (A-1) [14, 15, 16] and a small constant neutron background. The resulting net $\beta$dn decay curves for $^{133-135}$In are shown in Figure 1. The half-life ($T_{1/2}$) values are summarized in Table 2, and are compared to literature values and predictions from two Quasi-Particle Random-Phase Approximation (QRPA) models. The first one is in principle a 'deformed' QRPA [17] for Gamow–Teller (GT) decay, which takes the ground-state shape of the beta-decay daughter isotopes as predicted by the Finite Range Droplet Model (FRDM) [18]. This QRPA version uses experimental nuclear masses as far as they are available [14] and otherwise FRDM predictions. The second GT strength-function model is the so-called continuum-QRPA of Borzov et al. (see, for example, Ref. [19]) which is limited to spherical nuclei. It uses nuclear mass predictions from the extended Thomas-Fermi plus Strutinski integral model (ETFSI) [20].

The comparison of the experimental In half-lives listed in Table 2 with the predictions derived from the QRPA calculations of GT strength functions [18, 19] indicates a satisfactory agreement within about a factor two. The differences between the QRPA and cQRPA predictions mainly reflect the effects from the different pairing models used (Lipkin–Nogami in Ref. [17], and BCS in Ref. [20]) and the different $Q_\beta$ values. To give an example, for $^{132}$In Audi et al. [14] give an experimental $Q_\beta$ value of 14.13 MeV, which is roughly 1 MeV higher than the predictions of both mass models, FRDM [18] and ETFSI [20]. Using this experimental $Q_\beta$ value, our QRPA calculates a $T_{1/2}(GT) = 96$ ms, as listed in Table 2. When using the theoretical $Q_\beta$ values from FRDM or ETFSI instead, with our QRPA model we would obtain about a factor two longer $T_{1/2}(GT)$ of 181 ms and 164 ms, respectively. Actually, it is this latter half-life from the lower $Q_\beta$ value of ETFSI (13.23 MeV) that should be compared with the cQRPA prediction of 212 ms, which is in better agreement with the experimental $T_{1/2} = 206$ ms. This example shows, however, that 'better agreement with experiment' may turn out to be fortuitous and does not necessarily mean that the underlying theoretical model is better.

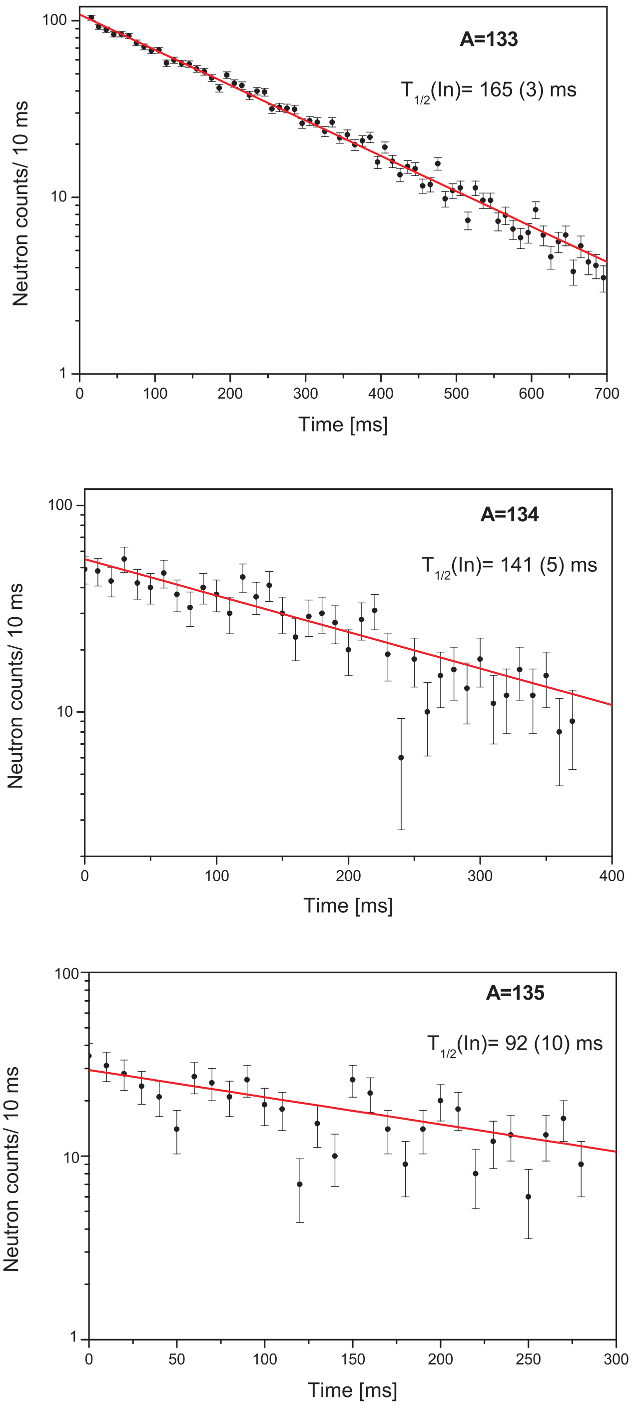

Figure 1: Beta-delayed neutron ($\beta$dn) decay curves for the neutron-rich isotopes $^{133-135}$In, after correction for $\beta$dn daughter activities and neutron background.

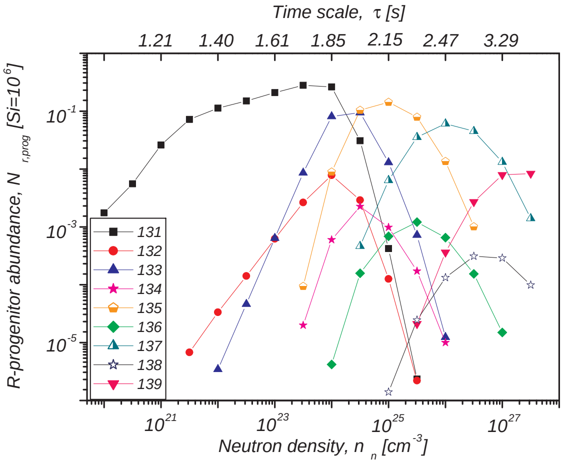

Figure 2: R-process progenitor abundances of N $\geq$ 82 indium isotopes at freeze-out conditions ($T_9 = 1.35$) as a function of neutron density. For details, see text.

### 3.2 R-process abundances

With respect to rapid neutron-capture nucleosynthesis (the r-process), among nuclear data beta-decay properties ($T_{1/2}$ and $P_n$ values) and masses (in particular neutron separation energies, $S_n$ values) are the most important input parameters for astrophysical calculations [22, 23]. As is shown in Fig. 2, the isotopes $^{131}In_{82}$, $^{133}In_{84}$, $^{135}In_{86}$, etc. are the major waiting-point nuclides in the region just beyond the A $\simeq$ 130 $N_{r,\odot}$ peak, carrying the main progenitor abundances prior to beta-decay. It is furthermore seen from this figure that the odd-neutron In isotopes (with ‘low’ $S_n$ values) only build up small progenitor abundances. This is because as soon as they have been formed in rapid neutron capture, the odd-neutron isotopes do not have time to beta-decay before they either capture an additional neutron to become an (A+1) even-neutron nuclide or photodisintegrate back to an (A-1) even-neutron In isotope (both with ‘high’ $S_n$ values) for which now beta-decay is the fastest reaction. As a consequence, the $T_{1/2}$ of these even-neutron isotopes are of major importance in the waiting-point concept, which implies the historical $N_{r,\odot}(Z) \times \lambda(Z) \simeq$ const. correlation.

In Fig. 2, we show the absolute r-abundances (normalized to Si $= 10^6$) of the $82 \leq N \leq 90$ In progenitors obtained from our standard multi-component, time-dependent calculations (see, for example, [24, 1]) for a freeze-out temperature of $T_9 = 1.35$ and assuming Fe as the seed element. Depending on the neutron density ($n_n$) and the corresponding process duration ($\tau$), different even-neutron In isotopes act as main waiting-points. For modest neutron densities of $n_n \simeq 10^{20}$–$10^{24}$ n/cm$^3$, representative for overpassing the A $\simeq$ 80 $N_{r\odot}$ peak and forming the second peak at A $\simeq$ 130, neutron-magic $^{131}$In is the waiting-point. At about $n_n = 10^{24}$ n/cm$^3$ and a process duration of 2.15 s, all major N = 82 r-progenitors below $^{132}$Sn, from $^{126}$Ru to $^{131}$In, have reached their approximate maximum abundances, thus reflecting the ‘bottle-neck’ behaviour of the neutron shell closure for the r-matter flow. When using an A $\simeq$ 90 seed composition, as suggested by the neutrino wind SN II scenario [24], the r-process avoids the delay of the matter flow by the N = 50 waiting-point isotopes $^{78}$Ni to $^{81}$Ga, and the above flow time reduces to about 350 ms. Our new N = 86 isotope $^{135}$In becomes the main waiting-point in the

In chain at neutron densities around $n_n \simeq 10^{25}$ n/cm$^3$, where the r-process has already succeeded to break through the N = 82 shell. Because of the expected high $P_n$ value of $^{135}$In, after freeze-out and $\beta$-decay back to stability, most of the A = 135 progenitor abundance will show up at (A-1), i.e. as $^{134}$Xe in the solar r-abundance curve. As is also seen from Fig. 2, for $n_n \geq 3 \times 10^{25}$ n/cm$^3$, the even-N isotopes $^{137}$In and $^{139}$In take over as waiting-points, although with decreasing abundances. Again, because of their predicted high $P_n$ values they will be the main progenitors of the (A-1) stable r-isotopes $^{136}$Xe and $^{138}$Ba, respectively.

## 4 SUMMARY

In this paper, we have presented production yields and beta-decay half-lives of very neutron-rich In isotopes, including the new r-process waiting-point nucleus $^{135}$In. These data, together with our recent results on neutron-rich Ag, Cd and Sn isotopes around doubly-magic $^{132}$Sn (see, for example, Refs. [1, 3, 16, 25, 26, 27]), have considerably improved our nuclear-structure knowledge in this interesting region. However, more detailed spectroscopic information is still needed; and in fact such experiments are underway by our collaboration at CERN/ISOLDE. The study of the beta-decay properties of the main N $\simeq$ 82 r-process waiting-point isotopes has undoubtedly led to a better understanding of the astrophysical conditions for the formation of the A $\simeq$ 130 $N_{r,\odot}$ peak [28]. Nevertheless, measurements of nuclear masses are still needed below and beyond $^{132}$Sn in order to obtain a completely experimental nuclear-data input in this important 'bottle-neck' region of the r-process matter flow.

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