Structure information on the *r*-process nucleus ¹³⁵Sn

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The β^- decay of ¹³⁵Sn was studied for the first time yielding a half-life of 0.6(1) s and a partial level scheme for the β^- decay to ¹³⁵Sb. The ¹³⁵Sn activity was produced at the mass separator OSIRIS via the fast neutron-induced fission of a ²³⁸U target inside a specially constructed ion source. ¹³⁵Sn is the heaviest Sn isotope, for which spectroscopic results are presently determined. The new results are in disagreement with some of the theoretical predictions, which are used for modeling of the astrophysical *r* process.

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The astrophysical modeling of the processes of synthesis of elements and their abundance in nature relies heavily on nuclear structure data and predictions from nuclear models. In the rapid neutron capture process (r process) [1], about half of the nuclei heavier than iron are produced in sequences of neutron capture and β decays. The key nuclear input parameters controlling the process flow are the masses or separation energies and the β -decay half-lives. If the neutron capture is much more probable than β decay then a new nucleus with the same proton number Z, and an increased neutron number N+1, is produced. Along the path, the neutron separation energy decreases and the capture of the next neutron is stopped when the rates for the neutron capture and photodesintegration processes become equal. At such "waiting-point" nucleus, further flow proceeds by β decay. This occurs 15 to 20 neutrons away from the valley of stability, where the separation energy is of the order of 2 MeV. The β decay then drives the path of the r process along the constant mass number A, towards the valley of stability until the neutron capture dominates again.

The *r* process modeling, besides purely astrophysical parameters like temperature and neutron densities, requires nuclear structure parameters on hundreds of exotic nuclei. At present, most of the relevant nuclei in the A > 70 region are not amenable to experimental investigation. Many *r* process nuclei have been identified for the first time at the FRS/GSI facility [2] using the projectile fission of uranium at relativistic energies but their low production intensity excluded detailed spectroscopy. The *r* process path comes, however, very close to the doubly magic nuclei of ⁷⁸Ni and ¹³²Sn, where it *can* be verified experimentally. In the vicinity of ¹³²Sn, the *r* process path initially follows the N=82 line until it reaches proton number Z=48. Then it turns outward and crosses the Z=50 line around ¹³⁵Sn [3].

In order to access experimentally the exotic nuclei at 132 Sn new advanced techniques of production, separation, or identification of exotic nuclei have been developed at OSIRIS at Studsvik [4], at ISOLDE/CERN [5,6], and in prompt fission [7]. The excited states in 133 Sn were obtained from the βn -gated γ -ray studies in the decay of 134 In at ISOLDE [5] while those in 134 Sn were obtained from prompt fission studies [7–9]. Moreover, detailed β -decay studies of

¹³³Sn and ¹³⁴Sn were performed at OSIRIS [4,10] using more efficient detection systems or innovative techniques.

The new data are needed to replace the less reliable model predictions with precise experimental values, in order to improve *r* process modeling. Of particular importance is the evolution of collectivity and the empirical strength of the first-forbidden β transitions, which are notoriously difficult to model, and yet they relate to the configuration of the β -decaying state and its lifetime. For the *r* process modeling it is also important to establish any weakening of major shell gaps in the nuclei with a large excess of neutrons. In this respect little is known about the strength of the *Z*=50 shell closure at *N*>82.

Up to now, ¹³⁴Sn was the heaviest Sn isotope where spectroscopic information has been obtained [8,9], while the upturn of the *r* process path is expected for heavier Sn isotopes. In this paper, we report on the first study of the β^- decay of ¹³⁵Sn where a novel production technique has been applied. Preliminary results were listed in [10].

The experiment was performed at the OSIRIS online fission-product mass separator [11] located at the R2-0 reactor in Studsvik. A special ion source, containing ²³⁸U isotope as a target, was prepared to enhance the production of very neutron-rich nuclei via the fast-neutron induced fission of ²³⁸U. Moreover, special arrangements were made in the R2-0 reactor pool and reactor channel to optimize the yield of fast neutrons. Although the overall production of the fission products was lower with the ²³⁸U target, the three extra neutrons in the compound state, as compared to the thermal fission of ²³⁵U, increase the relative yield of the neutron-rich ¹³⁵Sn by a factor of 15–20 relative to the less exotic A = 135 isobars, which act as impurities. The mass separated beam of A=135 was collected on an aluminized Mylar tape. The γ -ray singles spectra and γ - γ coincidences were measured using two large and one small low-energy Ge detectors, in coincidence with β rays recorded in a thin plastic scintillator. The β^- -decay chain of neutron-rich A = 135 isobars included the decays of ${}^{135}Sn \rightarrow {}^{135}Sb$ with the expected half-life of a fraction of a second, ${}^{135}Sb \rightarrow {}^{135}Te$ $(T_{1/2}=1.7 \text{ s})$, ${}^{135}Te$ $\rightarrow {}^{135}I$ $(T_{1/2}=18.6 \text{ s})$, ${}^{135}I \rightarrow {}^{135}Xe$ $(T_{1/2}=6.6 \text{ h})$ and fureven longer ther two decays with half-lives



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FIG. 1. γ -ray singles spectra following the β^- decay of ¹³⁵Sn and its daughter nuclei. See text for details.

[12]. There should be no detectable production of 135 In. The half-life of ¹³⁵Sn was identified from measurements of γ -ray intensity as a function of time in cycles of 3.2 s, each divided into eight time intervals of 0.4 s. During the first 1.2 s the radioactive A = 135 beam was continuously accumulated on the tape, then the beam was deflected and the sample was left to decay out over the next 2.0 s. At the end of the cycle, the old sample was moved on the tape to a shielded position and a new cycle began. Figure 1(a) shows γ -ray singles spectrum measured throughout the entire cycle. The spectrum is dominated by γ lines from the ¹³⁵Sb \rightarrow ¹³⁵Te and ¹³⁵Te \rightarrow ¹³⁵I decays, while lines from the ¹³⁵I \rightarrow ¹³⁵Xe and longer-lived decays are not observed. One may further enhance lines corresponding to the short half-lives. The spectrum shown in Fig. 1(b) was measured within time intervals 1 to 4. In addition, long half-life components were eliminated in this spectrum by subtracting a spectrum measured in the eighth time interval, which was normalized to match intensities of γ lines in ¹³⁵I in the 1 to 4 spectrum. Consequently there are no lines from the ¹³⁵Te \rightarrow ¹³⁵I decay in Fig. 1(b), while lines from the decay of 135 Sb are suppressed by a factor of 2.

Most of the lines observed in the spectrum Fig. 1(b) are identified as the known γ transitions from β decay of ¹³⁵Sb [13], except for a pronounced line at 281.7(1) keV. Figure 2 illustrates the intensity of the 281.7 keV line measured as a function of time, along the analogous plots for γ lines corresponding to the known [12] β^- decays: the 1127.0 keV transition in ¹³⁵Te and the 603.5 keV transition in ¹³⁵I. Each decay curve consists of five points, corresponding to time intervals 4 to 8, when no beam was collected. The half-life of 1.6(2) s obtained for the β^- decay of ¹³⁵Sb fits well the adopted value of 1.68(2) s [12]. The least square fit to the 281.7 keV line data gives a half-life $T_{1/2}=0.6(1)$ s, which is faster than other known decays in this isobaric chain [12]. Since this line is not due to mass cross contamination, molecular beam impurity, or a new isomer in a lower A = 135

isobar, it is thus identified as γ ray in ¹³⁵Sb due to the β decay of ¹³⁵Sn.

In the γ spectrum of Fig. 1 there is a line at 317.8(3) keV $[I_{\gamma}=18(6)]$, thus exactly at the same energy as the 2⁻ \rightarrow 1⁻ transition in ¹³⁴Sb [14,10]. The observation of this line, which we assign to the β -delayed neutron emission of ¹³⁵Sn, provides further evidence for the presence of the β decay of ¹³⁵Sn.

The $\gamma\gamma$ coincidence results reveal that the 281.7 keV line is in cascade with two other transitions. The upper panel of Fig. 3 shows a γ spectrum gated on the 281.7 keV line, where the 732.4(2) keV and 923.4(2) keV lines are clearly seen. A spectrum gated on the 732.4 keV line, displayed in



FIG. 2. Intensity of γ rays in ¹³⁵I (603.5 keV), ¹³⁵Te (1127.0 keV), and ¹³⁵Sb (281.7 keV) as a function of time after the beam was deflected and the source was left to decay. Solid lines represent the exponential decay law fit to the data.



FIG. 3. Coincidence $\gamma\gamma$ spectra gated on the 281.7 and 732.4 keV lines in ¹³⁵Sb, as measured in the present work.

the lower panel, shows that the 732.4 keV and 923.4 keV lines are not in one cascade. A partial decay scheme of ${}^{135}Sn \rightarrow {}^{135}Sb$ is given in Fig. 4.

In ¹³⁵Sb the valence proton occupies the $g_{7/2}$ orbital and determines the ground-state spin and parity $I^{\pi} = 7/2^+$ [15]. For the odd-Z, N = 84 isotones the first excited state has spin and parity $5/2^+$ as discussed in Ref. [16] and illustrated in Fig. 5. The 281.7 keV level in ¹³⁵Sb fits smoothly this systematics, and thus we tentatively assign spin $5/2^+$ to the 281.7 keV state in ¹³⁵Sb.

The ground state of ¹³⁵Sn results from the coupling of three valence neutrons in the $f_{7/2}$ orbital. This coupling can produce a multiplet of states with spins ranging from 3/2 to 15/2 (there are no spins 1/2 and 13/2 in this configuration due to the Pauli principle). Figure 6 shows the systematics of



FIG. 4. The level scheme of ¹³⁵Sb obtained in the present work. The relative γ intensities, determined from γ -ray singles spectra, are given in parentheses.

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FIG. 5. Level systematics of odd-Z, N = 84 isotones. Data are from this work and Refs. [17–19].

such excitations observed in the N=85 isotones with even Z (the spin assignments are partially based on model calculations). The ground states in these nuclei correspond to one of the three lowest-lying members of the multiplet having spins $3/2^-$, $5/2^-$, and $7/2^-$. Though the $3/2^-$ and $5/2^-$ members of the multiplet are not yet identified in ¹³⁷Te, clear trends seen in Fig. 6 indicate that the ground states in both ¹³⁷Te and ¹³⁵Sn have spin and parity $7/2^-$ while the $3/2^-$ and $5/2^-$ levels are placed higher in these two nuclei.

The β feeding of the 281.7 keV state is of considerable interest since our shell model calculations (see, e.g., [21]) indicate that the strength of this β transition is very sensitive to the purity of the configurations involved, giving large log ft values when the configuration mixing is increased. A relatively fast β transition would imply a very modest amount of configuration mixing which is possible only if the Z=50 shell gap is well developed at ¹³⁵Sn. Our experiments did not give a direct measure of absolute transition strengths. Some guidance regarding intensities can be given by a comparison of γ transitions in ¹³⁵Sb and ¹³⁵Te and estimates of the relative fission yields of the precursor nuclei. The Z_p model by Wahl [22] predicts the yield of ¹³⁵Sb to be about 20-120 times higher than that of ¹³⁵Sn. Making use of the known [23] separation efficiencies and diffusion properties of these elements, a direct comparison of the 296.9 and 281.7



FIG. 6. Low-spin excitations in the N=85 isotones, from Ref. [20]. Dashed lines are drawn to guide the eye.

keV γ rays in our spectra [see Fig. 1(a)] suggests that the latter has an intensity of about 2–12 % per decay of ¹³⁵Sn. The upper range of this prediction is implied from a comparison of the decay properties of ¹³³Sn and ¹³⁵Sn discussed next.

The ground-state-to-ground-state β transition from the three-valence neutron nucleus of ¹³⁵Sn cannot be faster than the corresponding favored first forbidden transition from ¹³³Sn, which having one valence neutron, already shows an exceptionally fast $\log ft = 5.44$. An immediate consequence is that the β branching to the ground state of ¹³⁵Sb must be below 60%. This value is deduced using the tables of Gove et al. [24], the half-life of 0.6 s, the decay energy of 8.9 MeV [3], and the limit of log $ft \ge 5.44$. Thus about 40% or more of the β intensity populates the excited states of ¹³⁵Sb, leading to γ ray and delayed neutron emission. The β strength distributions in this region [13,25] do not favor a strong population of unbound states, however, since the only allowed Gamow-Teller strength resides in the core transitions (with a combined strength corresponding to a $\log ft$ of 4.0) to states positioned of the order of 2-3 MeV below the Q value. For energetic reasons, these transitions have modest intensities. A modest delayed neutron emission probability (P_n) is also indicated by the fact that only one γ ray (317.8 keV) from the known low-lying states of ¹³⁴Sb was seen in our spectra, with an intensity of about one-fifth of the 281.7 keV line. As a comparison, the decay of ¹³⁵Sb, having a similar gross decay pattern as ¹³⁵Sn, leads to about 21% neutron emission [26], and a P_n of 16% has been calculated [27] for ¹³⁵Sn. We thus take 20% as a representatative P_n value, which implies that β feeding of the lower lying γ decaying states must be $\sim 20\%$ or higher. However, we expect that the unobserved γ rays directly feeding the ground state represent less than 50% of total γ intensity. This is a conservative estimate in view of the low-level density in ¹³⁵Sb and the corresponding concentration of intensity in a small number of observed γ transitions. Consequently, the strong 281.7 keV γ ray from the first excited state is predicted to carry 10% or more of the total β decay intensity.

The coincidence spectra show no trace of γ ray population of the first excited state in addition to the 732.4 and 923.4 keV lines, suggesting that about 30% of the population of the 281.7 keV state is due to β feeding. In absolute units, this corresponds to 3% per decay or more, implying a fast log *ft* of less than 6.7 for this first-forbidden β transition between the 7/2⁻ and 5/2⁺ states.

Such an estimated value, significantly below 7, points towards the involvement of rather pure configurations and thus against any quenching of the Z=50 shell gap up to neutron number N=85. Despite large uncertainties, the estimated log *ft* 6.7 seems to be lower than the log *ft*=7.2 [28] measured for the corresponding β transition (9/2⁺ \rightarrow 7/2⁻) in the decay of ²¹¹Pb to ²¹¹Bi where the Z=82 shell closure is known to be strong. This finding is also supported by our shell model estimates which indicate that mixing in the 7/2⁻ state of ²¹¹Bi is stronger than in the 5/2⁺ state of ¹³⁵Sb, which we interpret as a consequence of a larger shell gap at ¹³⁵Sn than at ²¹¹Pb.

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FIG. 7. Half-lives of ¹³³Sn, ¹³⁴Sn, and the new experimental value for ¹³⁵Sn. Note a regular dependence of half-lives in Sn isotopes with the increasing neutron number. The dashed line is drawn to guide the eye.

The newly measured half-life of ¹³⁵Sn, $T_{1/2}=0.6(1)$ s, is compared in Fig. 7 to the theoretical calculations. A very good agreement is found between the experimental value and the results of $T_{1/2}=0.5$ s calculated within the so-called gross theory [29] and also using the finite Fermi system model [30]. On the other hand a value of $T_{1/2}=3.0$ s calculated on the basis of the QRPA model [3] differs significantly from our experimental result. The latter calculations, which are important for the *r* process modeling, consider only the Gamow-Teller β strength functions, and thus exclude the first-forbidden transitions, which clearly play an important, if not dominant, role in the β decay of ¹³⁵Sn. Moreover, they predicted the spin/parity of $1/2^-$ for the ground state of ¹³⁵Sn, quite inconsistent with our new results.

In summary, excited levels in ¹³⁵Sb, populated in β^- decay of the very exotic ¹³⁵Sn nucleus, were studied. The halflife of ¹³⁵Sn, measured as $T_{1/2}=0.6(1)$ s, agrees with the gross theory calculations but differs significantly from the prediction of the QRPA theory. The properties of the β decay of ¹³⁵Sn imply that the first-forbidden transitions to the ground state and the first few excited states are fast, which means that calculations based only on the Gamow-Teller β strength functions would strongly overestimate the half-life of ¹³⁵Sn.

Note added. There was a more recent study of the β decay of ¹³⁵Sn performed at ISOLDE using the resonanceionization laser ion-source (RILIS). Their preliminary results [31] are consistent with our findings.

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