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Report to the INT Committee

Decay study for the very neutron-rich Sn nuclides, $^{135-140}$Sn separated by selective laser ionization

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Abstract

Data have been taken on two separate occasions for this study of the decay of the very neutron-rich Sn nuclides using the Resonance Ionization Laser Ion Source, first in July 2000 and the second in August 2002. In the first experiment, time dependent gamma ray singles and coincidence spectra were taken for decay of ^{135}Sn as well as time dependent beta-delayed neutron spectra for ^{135}Sn, ^{136}Sn, and ^{137}Sn. The information that could be extracted from the gamma-ray spectra at A = 135 was limited owing to the large activity from isobaric 53-min ^{135}Cs^{m}. Clean neutron spectra were obtained at A = 135 and 136, but surface-ionized Te was present at A = 137 and dominat at A = 138. In a second experiment in August 2002, the neutron converter was used. Under these improved conditions, some of the previous measurements were partially repeated. In about 3 hours, better gamma-ray data were taken at A = 135 than in all of the 2000 experiment. Likewise, the background limitations at A = 137 for the delayed neutrons were reduced significantly and new data for beta-delayed neutron data were obtained for ^{138}Sn decay. In this addendum, we are requesting an 24-shifts, 18 for experiment using the neutron converter and the High-Resolution Separator with the goal of obtaining gamma-ray data at A = 137 and improved beta-delayed neutron data at A = 138. and 6 shifts for study of separation of SnS^{+} ions.

1. Introduction

These experiments, approved as IS-378, to use the Resonance Ionization Laser Ion Source (RILIS) to achieve high selectivity for the study the decay of very neutron-rich Sn nuclides were performed in July 2000 and August 2002. The goals of these experiments are to determine the half-lives and gamma-ray spectra for very neutron-rich Sn nuclides that lie directly in the path of r-process nucleosynthesis. The last several years have been very exciting times for studies of r-process nucleosynthesis, in part, because of the discovery of SEVERAL old halo stars which exhibit abundances for elements with Z > 56 that are quite close to the solar r-process distribution as shown in Figure 1. [1,2]

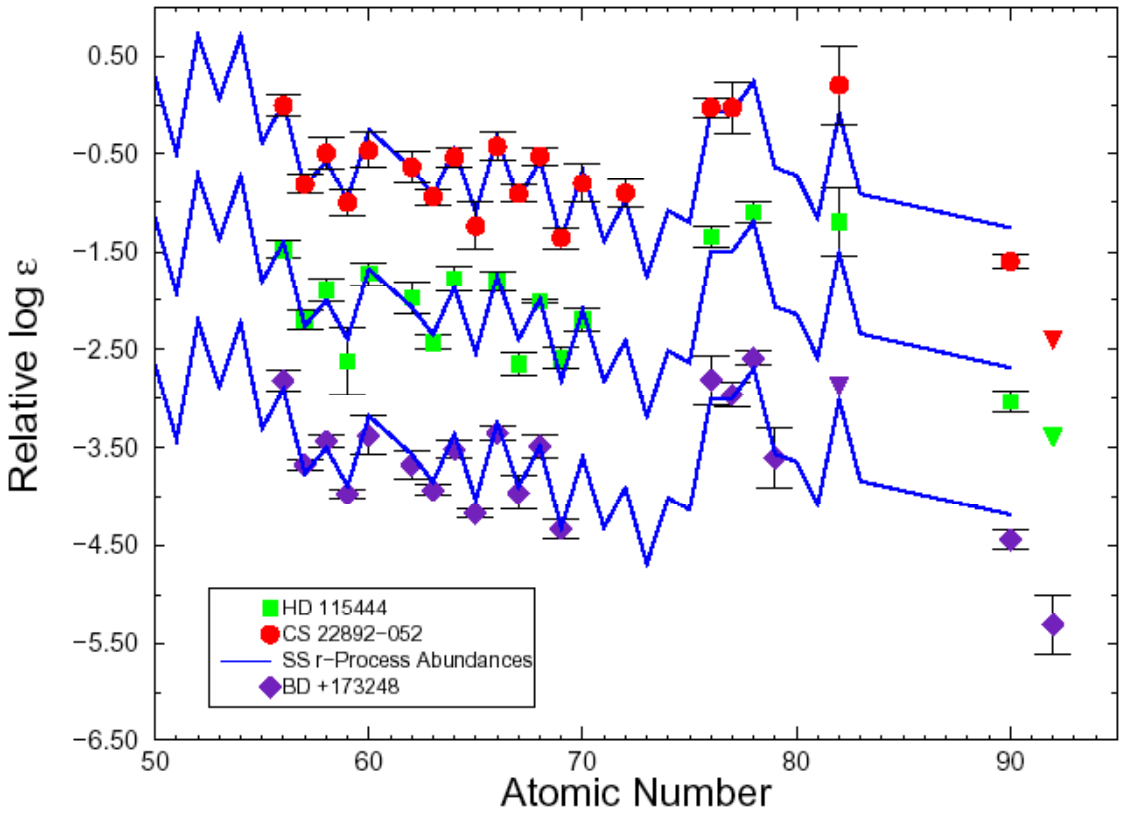

Figure 1. The heavy element abundance patters for two stars CS22892-052 and HD 115444 are compared with the solar system r-process abundance distribution (solid lines).

These and other data add fuel to the debate about the presence of a single site for r-process nucleosynthesis, or at least a single mechanism for elements above Z = 56 and possibly a second site/mechanism for elements below Z = 56. As the production of such elements with Z > 56 must pass the A = 130 “waiting-point” region, added significance is placed on the quality of the experimental data for the nuclides in the A = 130 region.

II. Results in 2000 and 2002

The data from the 2000 experiment hve been published. [3] In that paper, new structure for ^{135}Sb was reported that included an unexpectedly low energy for the first excited $d_{5/2}$ level. New data with low uncertainties were also reported for the half-life and Pn values of ^{135}Sn and ^{136}Sn. New data for the half-life and P_n values were also reported for ^{137}Sn decay, but, as no data existed for the decay of the daughter ^{137}Sb, the data had much larger uncertainties. Interferences from other isotopes, probably ^{138}Te completely covered up any ^{138}Sn that was produced. No gamma rays that went away when the laser was blocked were observed at either ^{136}Sn or ^{137}Sn owing to surface ionized Cs and Ba.

Our goals in 2002 were to improve the quality of the data for A = 137, both for gamma rays, and to obtain new data for ^{138}Sn decay. To accomplish these goals, the neutron converter was used with great success. Far better gamma spectra for ^{135}Sn decay were obtained in a few hours of data collection. This was because most of the interfering Cs isotope, ^{135}Cs^{m} is produced in spallation and was hence greatly reduced by the use of the converter. As the count rates in the delayed-neutron spectra at A = 137 and A = 138 with the laser off were dramatically lowered, it was possible to obtain better precision for the half-life and P_n values for ^{137}Sn decay and to observe neutrons from ^{138}Sn decay.

Using the new half-life data for ^{137}Sb decay of 330(90) ms from GSI data provided by Hendrick Schatz, [4] we now determine a half life for ^{137}Sn of 185(35) ms as compared with 190(60) from the 2000 data. As no useful P_n value emerged from the GSI study, we still deduce P_n values of over 50% for both ^{137}Sn and ^{137}Sb. The background reduction is shown in Figure 2 where the data were normalized to the laser-on spectrum.

Below in Figure 3 are shown the laser-on and laser-off data for ^{138}Sn decay. What are shown are the data with the laser on, laser off, the difference, and fits to the difference with a 200 ms half-life for ^{138}Sn, a 250-ms half-life for ^{138}Sb, and a 330-ms half-life for ^{137}Sb along with a 40% P_n value for ^{138}Sn. What is interesting is that the difference data can be fit to a single decay curve with a 450-ms half life, which is more or less the sum of the first generation ^{138}Sn decay and the second generation ^{138}Sb and ^{137}Sb decay. Thus, a shorter ^{138}Sn half life would require a longer ^{138}Sb half-life and visa versa. We estimate the uncertainties for the half-lives of ^{138}Sn and ^{138}Sb at about 75 ms at this point. Stated another way, it is difficult to fit the data with ^{138}Sn half-lives above and below than this range (125 ms to 275 ms) using realistic values for the P_n and half-life of ^{138}Sb.

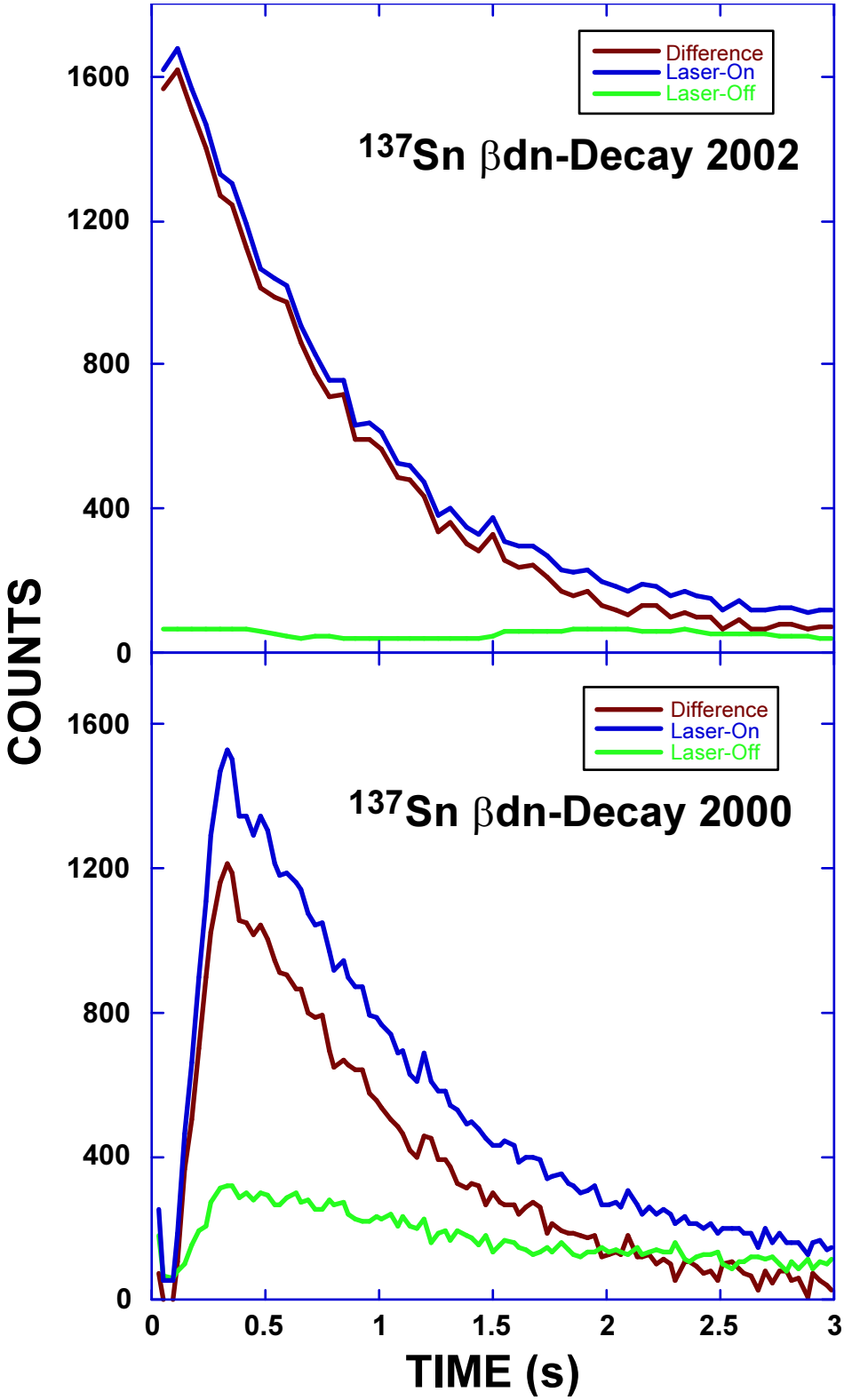

Figure 2. Beta-delayed neutron data as a function of time showing the lowered data rate with the laser off in 2002 where the neutron converter was used. Note that the counting in 2000 started with the PSB pulse hitting the target and in 2002, counting started when the beam gate was closed.

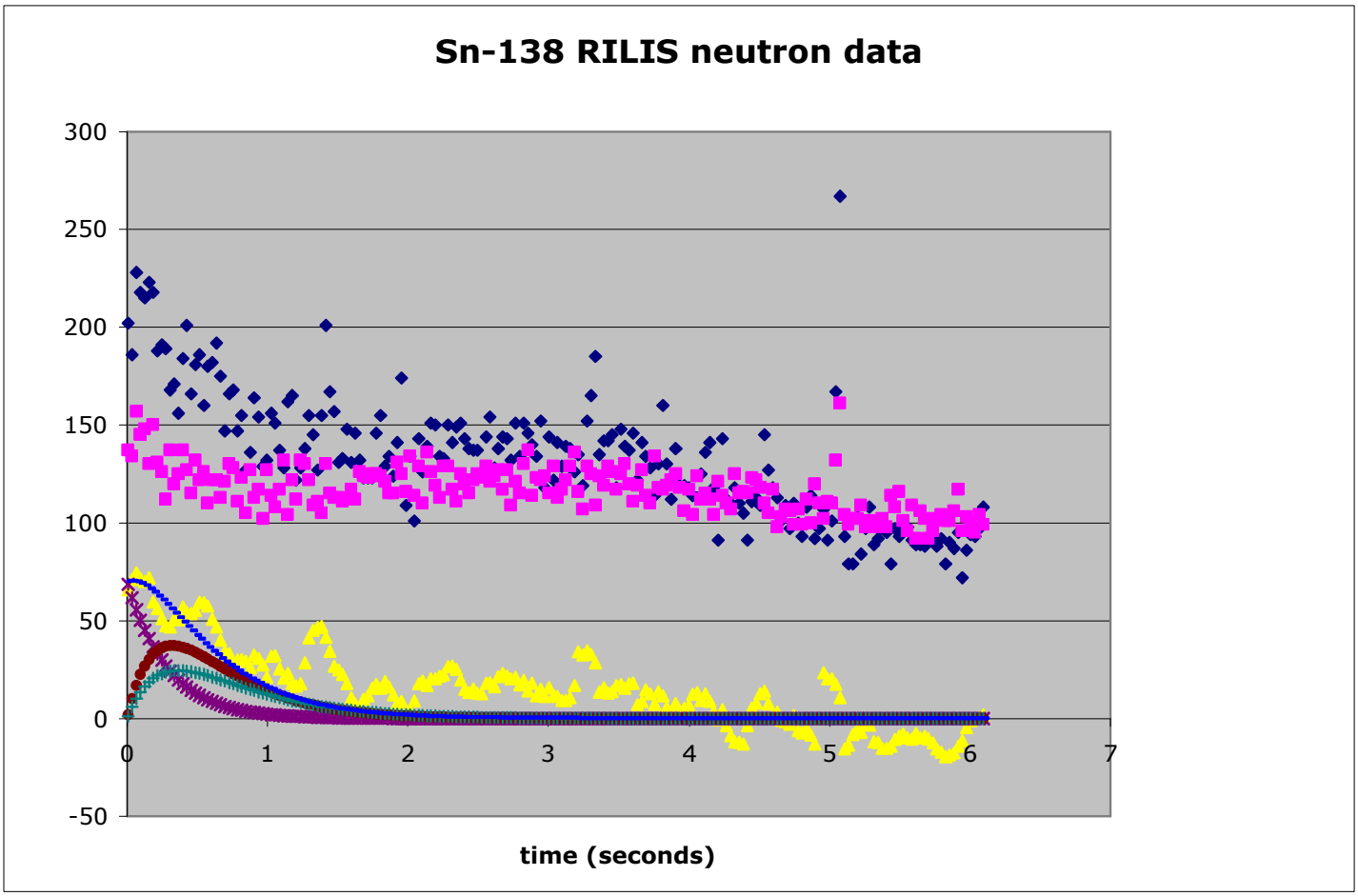

Figure 3. Beta-delayed neutron data at A = 138.

There are clearly some oscillations in the data that are probably related to new preamplifiers that had to be discarded in favor of the older preamplifiers later in the running cycle. Clearly these problems were quite small and did not affect the data at A = 137. But, the low count rates and relatively large background at A = 138 did reveal the problem, but only after the experiment was completed.

We quickly realized that any extended running for gamma rays at A = 136 would contaminate the tape with 13-day ^{136}Cs. Hence, we concentrated our gamma-ray efforts at A = 137, and again were not able to observe any gamma rays that disappeared with the laser light was blocked. The principle interferences came from 2-min ^{137}Ba^{m} andfrom ^{138}Cs. The ^{138}Cs activity was not a function of poor setup of the GPS, but rather from the very broad peak for the surface ionized Cs that comes from many different places inside of the front end. To check this, we took data at A = 130 where the gamma rays from both ^{130}In isomers and from ^{131}In isomers are well known and found no ^{131}In gamma rays at A = 130.

In Table I the new results for the very neutron-rich Sn nuclides are added into the table presented in the original proposal. The uncertainties in the half-lives and P_n values for both ^{137}Sn and ^{138}Sn will improve when new and better data are obtained for the half-lives and P_n values for the antimony daughters.

Table I. Measured and calculated half-lives for very neutron-rich Sn nuclides.

Values in blue are from 2000, green 2000, improved in 2002, and red from 2002.

A	133	134	135	136	137	138	139	140	
T meas (s)	1.44	1.12	530(20)	275(25)	185(35)	200(80)			
Pn(%)	2.9(2)	13(1)	21(3)	30(5)	50(20)	50(20)			
QM (MeV)	8.0	7.1	9.4	8.2	10.5	8.4	10.9	9.5	
QAudi	7.6	7.2	8.6	8.0	9.7	8.9	10.7	9.8	
TM(GT) (ms)	10.3 s	3.5 s	3000	950	800	480	390	120	
THilf (ms)			731	189	110	49	28	16	
TGroote (ms)				312	493	327	116	77	37
T ff+GT (ms)	[1996]		300	209	186	162	62	57	
T ff+GT (ms)	[this work]		400	600	120	200	80	100	

The lines QM and TM(GT) are the published values from Moeller, Nix and Kratz. [5]

The TM(GT) half lives include only for the Gamow-Teller branches.

The values labeled THilf and TGroote were taken from the compilation of Staudt et al. [6]

III. Plans for 2003

Our goals for the experiment proposed in this addendum are to improve the quality of the data for gamma rays from ^{137}Sn decay and for neutrons from ^{138}Sn decay by using the High-Resolution Separator and neutron converter to lower the backgrounds of surface-ionized Cs nuclides and whatever is observed at A = 138 in the neutron spectrum. The laser-off data can be fit by a combination of 1.5-second ^{138}Te and both growth and independently produced 6-second ^{138}I.

In Figure 4 are shown data taken with the RILIS Ag ion source as a function of HRS mass settings. These data were taken as a part of IS-333 also that was performed after IS-378 in August 2002. The data show that the FWHM of the HRS is 0.03 amu or 27 MeV for the very tightly focused ^{126}Ag beam that originates at the center of the ion source where the ionization is driven only by the laser. In contrast, both the In and Cs show a broader distribution as they originate on surfaces as well as in the center of the ion source.

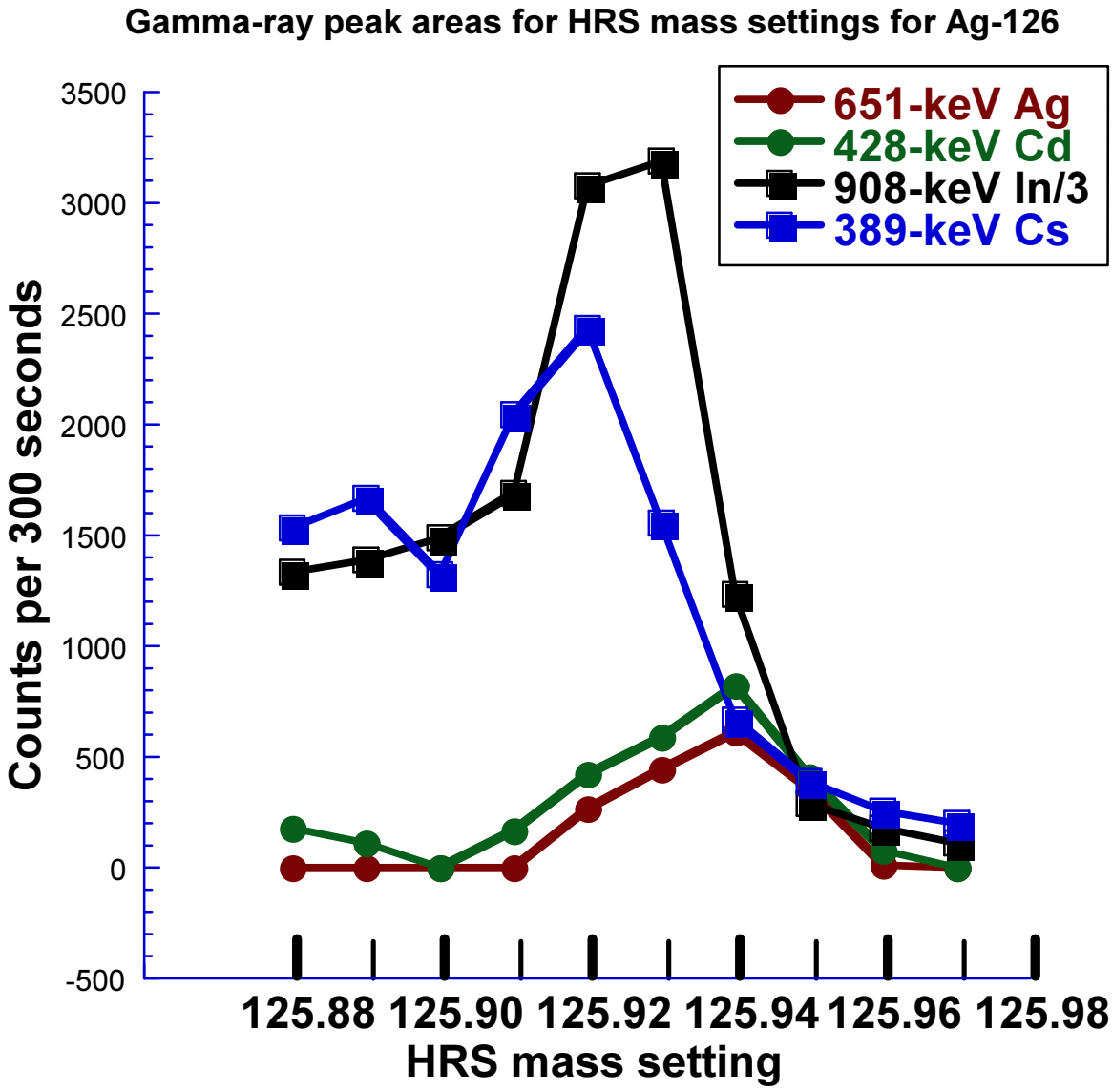

Figure 4. HRS data for A = 126 showing Ag, Cd, In, and Cs activities. The In data are scaled by a factor if 0.33.

Our plan for the run proposed in this addendum is to take advantage of the narrow focus for the laser-ionized Sn isotopes to use the HRS with carefully positioned slits that include most of the Sn, but exclude a significant fraction of the less-well focused Cs, I, and Te isotopes whose gamma rays and delayed neutrons interfere with our studies of Sn gamma rays and delayed neutrons.

We are most eager to find gamma rays from ^{137}Sn decay as well s from growth and decay of ^{137}Sb. Such data will greatly improve the precision of the half-life values and permit a much better determination of the P_n values for ^{137}Sn and ^{137}Sb decay. In addition, we are seeking information about the location of the first excited state in daughter ^{137}Sb to determine if the low energy for this level persists as N increases from 84 to 86. We are also eager to improve our data for decay of ^{138}Sn decay. In our previous paper, [3] we noted that this nuclide is most likely the waiting point for r-process nucleosynthesis at neutron densities needed to reach the A = 190 peak, whereas our new data now indicate that ^{136}Sn will be the waiting-point at moderate neutron densities (10^{23} – 10^{24} n/cm^3).

IV. Improved selectivity with an SnS^+ beam.

In our initial proposal, we had hoped to be able to reach ^{139,140}Sn. It now appears that under current operating conditions, this would not be possible owing to both isobaric contaminations, small that they may be, as well as the release time for isotopes in the ion source. Therefore, we would also like to move forward with additional tests of the use of molecular/ionic tin sulfide as a way to isolate much cleaner beams.

The study of exotic isotopes is mainly limited by two factors:
1. The release losses of short-lived isotopes.
2. The remaining isobaric background.
For tin it has been shown recently that the release can be accelerated significantly by using a chemical evaporation method.[7] Adding sulfur or a sulfur-containing compound (CS_2, H_2S, SO_2) to the target will lead to the creation of tin sulfide molecules (SnS) which are much more volatile than atomic tin. Thus the surface desorption time is reduced by a factor 100. The release is accelerated and, hence, the release efficiency of short-lived isotopes is significantly increased.

In the case of neutron-rich tin the most disturbing background is due to surface-ionized cesium and barium and in certain cases also due to Sb, Te, I or Xe ionized by another mechanism. Even a tiny ionization probability of < 10^{-4} will create disturbing background due to the much higher production cross-sections. Using the SnS molecule not only for the transport to the ion source, but also for the separation (after ionization to SnS^+) makes it possible to escape from most of the isobars which do not form stable mono-sulfide ions themselves. It has been shown that e.g. Sb is thus suppressed by at least a factor 200. [7] Up to 60% of the extracted tin ions appear in the molecular sideband.

At present the separation of SnS^+ molecules is very successfully used for the investigation of isotopes close to ^{100}Sn at the GSI-ISOL separator. [8] In 2002 a brief on-line test was made at ISOLDE using an old UC_x/graphite target, a MK5 ion source and addition of SO_2 via the standard gas leak. [9] Molecular sidebands of SnS^+ were observed, but the sulfur addition was by far too low to reach saturation of the SnS^+/Sn^+ ratio. Also some additional problems appearing on the neutron-rich side need to be studied more thoroughly: background from $^{A-2}Sn^{34}S^+$, from $^{A+32}Ln^+$, from $^{A+16}Ln^{16}O^+$, etc. The ultimate beam purity could be obtained after breaking the pre-separated SnS+ beam in a second stage: in the REXTRAP, the REX-EBIS, the charge-breeding-ECRIS, or in a gas cell (after slight acceleration with the HV platform) and re-separating now the atomic Snx+ beam. The first separation stage will remove the direct isobars and the second one the A + 32 isobars.

For a detailed exploration of this exciting new method we are asking for an additional 6 shifts with a UC_x/graphite target with sulfur addition and coupled to a MK5 ("hot plasma") source. These shifts will be used to carefully map the yields of the atomic and molecular ions present in the mass range 160 < A < 170 and to measure the release function of SnS+. Depending on the background at mass 168, it might even be possible to obtain some gamma-ray spectroscopy for ^{136}Sn decay (so far impossible when separated as atomic ion).

V. Beam-time request

At this point, 7 shifts remain for IS-378. We would like to request an additional 11 shifts for a total of 18 shifts with the HRS and neutron converter to be used as follows. We would expect to use 4 shifts for studies A = 134 and A = 135 where we can determine the shape of the Sn peak and a very close mass calibration for the HRS to be sure that we are at the right mass and slit settings for maximum yield and beam purity at A = 137 and 138. Then, we would use 6 shifts for the study of gamma rays at A = 137, 4 shifts with the laser on and 2 shifts with the laser blocked. And we would use 8 shifts at A = 138 to study beta-delayed neutrons, 4 with the laser on and 4 with the laser blocked. These beam time requests are based on the expectation of yields comparable to those observed in 2000 and 2002 which are summarized below.

We have had a number of discussions concerning the nature of the experimenter's responsibility for setup and calibration of the HRS. We have been encouraged to include such time in the beam-time request. This time includes release-time studies for the ion

source, optimization of the location of the proton beam on the converter, and setting of the beam gates as well as the setting of the mass calibration and the setting of the slits.

Not as much time with the laser blocked is required for gamma rays studies as the data are taken as a function of time and the time at the end of the counting period is usually low in the Sn activity and high in background and daughter activity.

For the beta-delayed neutron data, however, we observed only about 10% more counts with the laser on as with the laser blocked. As our interest is in the overall difference between these two spectra, approximately equal counting times provide the best statistical properties for the final result.

Below we summarize the beam time request:

mass	time (shifts)	nuclides/PSB pulse	
134/135	4	10,000	beam tuning and setup
137	6	~100	gamma spectroscopy
138	8	~2	neutron spectroscopy
135/136	6	?	search for tin-sulfur ions at A = 167/168

The total efficiency for the low energy gamma rays we seek is ~2%/detector or 8% for the summed spectra from four detectors. Based on the observation of 2 counts/sec in the 282-keV peak from ^{135}Sn decay and a loss of yield of a factor of 100, we would expect ~1000 counts in a peak near 200 keV for ^{137}Sn decay in each detector. The efficiency for the neutron detection is ~50% providing ~1 net count/PSB pulse, well within our range of observation. [For 429 PSB pulses, we observe ~4100 counts with the laser on and 3600 counts with the laser blocked.]

We wish to re-iterate that the gamma spectroscopy request at A = 137 is largely aimed at finding gamma rays belonging to decay of both ^{137}Sn and ^{137}Sb so that improved half-lives can be determined. We certainly expect to observe the first excited state of ^{137}Sb populated in the decay of ^{137}Sn and compare its position with the 282-keV first excited state in ^{135}Sb, but beyond that detailed level structure information is not a high priority.

The current rate for nuclides deposited on tape for ^{138}Sn is ~ 2 nuclides/PSB pulse added to a rate of ~16 nuclides/PSB pulse of other isobars. If we are able to reduce that background by a factor of 3 through the use of a narrower slit width, we should be able to obtain a significantly more precise values for the half-lives of ^{138}Sn and its daughters.

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