

### Letter of clarification for addendum INTC-P-550-ADD-1

“Nuclear moments of excited states in neutron rich Sn isotopes studied by on-line PAC”  
(For the TinPAC collaboration: H. Haas)

Before answering the specific questions of the INTC a short description of the present status of the TinPAC project (IS673) is required. As made obvious in Fig 1 practically all on-line PAC measurements possible with the present setup have been successfully completed. Within the technical, financial, and manpower limitations the present collaboration cannot afford the necessary modifications. For the off-line results (Fig 2), on the other hand, precision and reliability were limited by three difficulties that we now can overcome. This has led to the present proposal.

I understand that the present addendum asking in total for 10 separate measurements using 5 different matrices is difficult to follow without on-site experience with PAC. In addition to the knowledge of the nuclear physics techniques it requires familiarity with various technical aspects of the PAC method and sample handling. It is thus of central importance for the completion of our project with the present technical possibilities that the three experienced senior investigators (HH, JGC, JR) are available for successfully conducting the complex measurement program. As this will presumably not be the case after the coming long shutdown of the ISOLDE operation at the end of this year, the scheduling in 2025 of the 2 days for the planned experiments is of vital importance.

As appendix I have added the listing of the suggested experiments with the code used in the submitted proposal and my communication with INTC member Dr J.A. Lay that also touches some of the questions.

**Question 1: The original proposal allocated 4 shifts to measure the 5- isomer in  $^{130}\text{Sn}$ . During the presentation, it was mentioned that this was not measured due to the lack of narrow-band laser ionization. Please explain why this measurement is no longer included in the current proposal as the narrow-band laser mode could be requested?**

As stated above, the prime purpose of the present addendum is to obtain as precise as possible nuclear moments for the 5- states in  $^{116}, ^{118}, ^{120}\text{Sn}$  by a new off-line run using Sb isotope sources. A project to measure the 5- state in  $^{130}\text{Sn}$  would require a  $^{130}\text{In}$  beam from ISOLDE, favorably with narrow-band laser ionization. As mentioned in the enclosed slide 6 of my oral presentation, the difficulties encountered with the extremely short-lived isotope  $^{130}\text{mIn}$ , 0.29s, would require a complete rebuilding of our now existing system, not realizable under the present conditions. An attempt at a similarly difficult case, the magnetic moment of the 6+ isomer in  $^{132}\text{Sn}$ , is being considered as a test using the ISOLDE Decay Station this year.



Figure 1: Slide 6 of presentation

**On-line run in June 2023**  
using fully digital PAC spectrometer (LaBr<sub>3</sub>)

**What we have measured:**

<sup>119</sup> In(2.4m) to <sup>119</sup> Sn(3/2 <sup>+</sup> , 18ns) in Zn	undamped spectrum at RT, $\nu_Q$ agrees with MS
<sup>122</sup> In(10.8s) to <sup>122</sup> Sn (5 <sup>-</sup> , 7.9ns) in Graphite	acceptable data for QI
<sup>122</sup> In(10.8s) to <sup>122</sup> Sn (5 <sup>-</sup> , 7.9ns) in Fe	approximate value for $\mu$
<sup>124</sup> In(3.7s) to <sup>124</sup> Sn (5 <sup>-</sup> , 520ns) in Zn	good data! Q determined
<sup>124</sup> In(3.7s) to <sup>124</sup> Sn (5 <sup>-</sup> , 520ns) in Cd	acceptable data, efg ratio Zn/Cd determined!
<sup>124</sup> In(3.7s) to <sup>124</sup> Sn (5 <sup>-</sup> , 520ns) in Fe	very good data for $\mu$ , damping similar to <sup>116</sup> Sn
<sup>126</sup> In(1.6s) to <sup>126</sup> Sn (5 <sup>-</sup> , 10.8ns) in Graphite	preliminary data for QI

**Difficulties encountered:**

For T<sub>1/2</sub><10s isotopes time-structure of ISOLDE beam makes measurement inefficient!

Suggested solutions: Including time in data system, replace photomultipliers with semiconductors, use staggered ISOLDE beam, use state-selective laser ionization, use mechanical system to reduce long-lived contaminants

Better interpretation of data in Graphite call for a (technically demanding) reference measurement with <sup>118</sup>In implantation

For states with T<sub>1/2</sub> below 50ns Gd matrix (cooled) needed for accurate  $\mu$

Question 2: During the presentation, it was stated that the quadrupole moments were determined with high precision already. What additional value will further measurements bring? Specifically, why is it necessary to measure 24 samples for <sup>116</sup>Sn and two samples for <sup>118</sup>Sn (see table p. 6 of proposal)?

This question requires a somewhat complex answer, since it touches the process used to obtain precision (meaning precision within the possibilities of the present experimental approach) nuclear quadrupole moments for the 5- states in <sup>116</sup>, <sup>118</sup>, <sup>120</sup> Sn. The values for Q come from the value for the state in <sup>116</sup>Sn as taken from [2], recalculated using the newer and more precise BE2 value [4], via the ratios of the corresponding quadrupole interaction frequencies  $\nu_Q$ . As stated in Fig 2, the slide 5 of the presentation, the only obtained “precision” ratio value is the Q ratio between <sup>120</sup> and <sup>118</sup> Sn from the measurements in graphite. The proposed measurements a) and c) both serve the purpose of determining the ratio between <sup>116</sup> and <sup>118</sup> with precision, while b) is intended to obtain a “precision”  $\nu_Q$  for <sup>116</sup>Sn(5-) in Cd, as the 2 available values from PAD experiments [8] [9] differ by almost 5%.

Figure 2: Slide 5 of presentation

**Off-line run in October 2021  
using 3 different PAC spectrometers**

**What we have measured:**

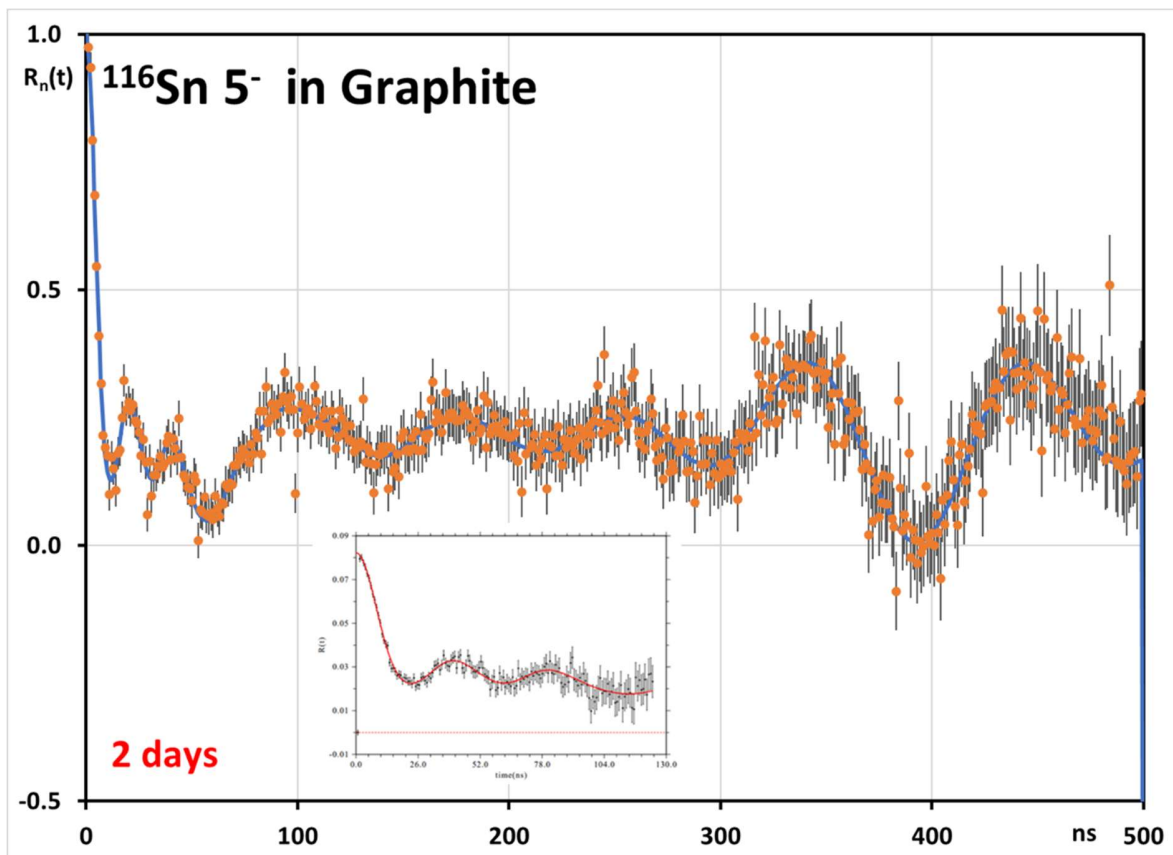
- |  |  |
|--|--|
| $^{116m}\text{Sb}(1\text{hr})$ to $^{116}\text{Sn}$ (5 $^-$ , 350ns) in Fe       | accurate data, $\mu$ agrees with PAD                   |
| $^{116m}\text{Sb}(1\text{hr})$ to $^{116}\text{Sn}$ (5 $^-$ , 350ns) in Zn       | expected spectrum not seen, failed annealing!          |
| $^{118m}\text{Sb}(5\text{hr})$ to $^{118}\text{Sn}$ (5 $^-$ , 22ns) in Fe        | good data for polarized Fe foil give $\mu$             |
| $^{118m}\text{Sb}(5\text{hr})$ to $^{118}\text{Sn}$ (5 $^-$ , 22ns) in Zn        | annealing results in strong damping! Hope!             |
| $^{118m}\text{Sb}(5\text{hr})$ to $^{118}\text{Sn}$ (5 $^-$ , 22ns) in Graphite  | good data for QI, reference?, annealing studied        |
| $^{120}\text{Sb}(5.8\text{d})$ to $^{120}\text{Sn}$ (5 $^-$ , 5.6ns) in Graphite | precision spectra give Q relative to $^{118}\text{Sn}$ |

**Difficulties encountered:**

- Annealing of Sb implanted in Zn at 200C apparently resulted in segregation at surface  
Spectra for  $^{118}\text{Sn}$  confirm this, preliminary data without annealing acceptable!  
For  $^{118,120}\text{Sn}$  measurement in Gd or high external field needed for precision  $\mu$

This leaves to explain the number of samples required for each of these measurements:

Figure 3: Simulation for experiment a)

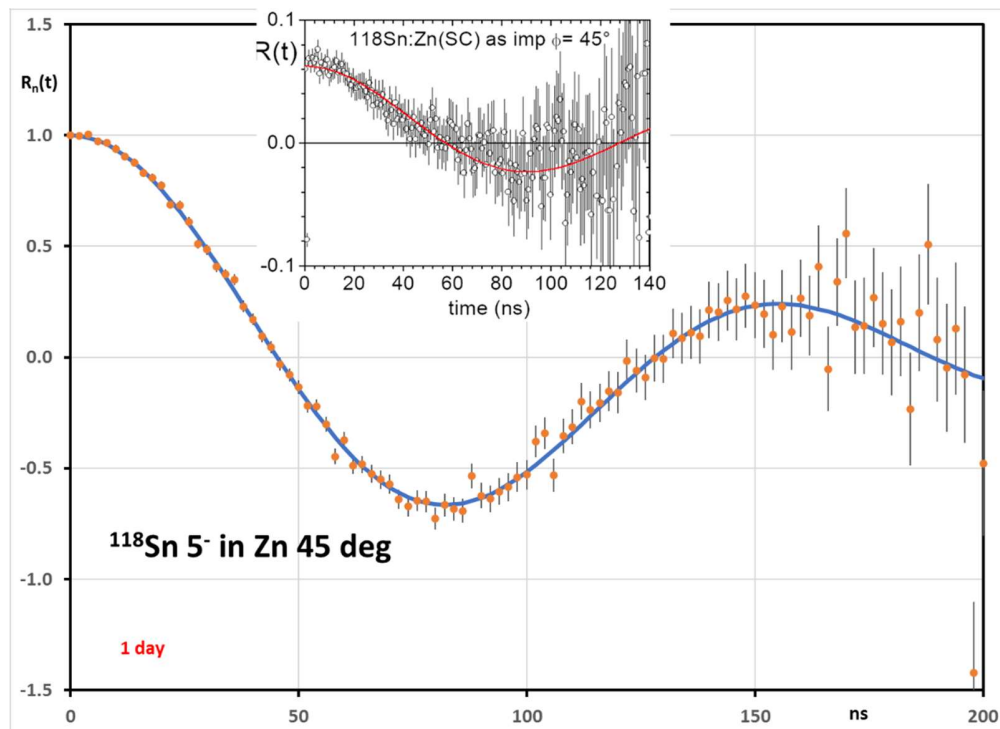


a) Figure 3 shows the expected perturbation function for the  $^{116}\text{Sb}$  source in graphite following annealing simulated using the corresponding result for 118 (insert) assuming 2 times larger  $Q$ . Realistic error bars are obtained from measurements for  $^{116}\text{Sn}$  during the first off-line run. For the determination of the frequency ratio only the first two peaks in the 0 to 60 ns range are relevant. The long running time required is due to the small time binning necessary for the expected high frequency, the small active amplitude and the unused data after 60 ns. With the short source half-life of 1hr this translates to the required 16 samples to reach statistical significance. The peak at 400 ns in the simulation has its origin in the low-frequency component necessary to fit the mass 118 data. In case it really appears in the final data it could be related to nuclei in the graphite interlayer space.

b) Since the most efficient PAC spectrometer will be all the time occupied by the a) measurement, another system, somewhat less efficient, will have to be used for the Cd matrix experiment. In principle only 3 to 4 samples should be sufficient, but possible annealing failures could require more.

c) The much easier measurements with the  $^{118}\text{Sb}$  source, half-life 5 hrs, normally need just one sample. The two foreseen in this case could, however, also become necessary for an annealing test. The precision for the 116 to 118  $Q$  ratio unfortunately depends on this uncertainty. The simulated perturbation function is shown in Figure 4 together with the result of the short measurement during the first off-line run as insert.

Figure 3: Simulation for experiment c)



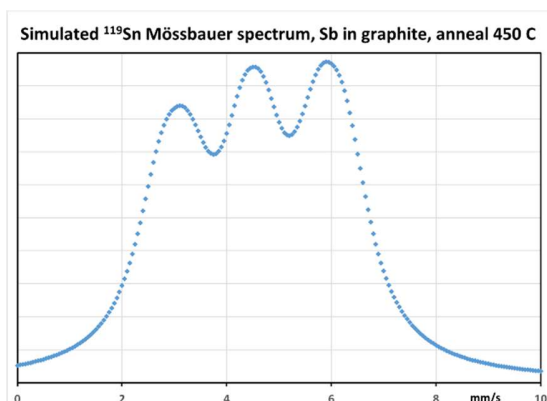
Question 3: What can be gained by measuring the same properties of  $^{119}\text{Sn}(3/2^+)$  from both  $^{119}\text{Sb}$  offline and  $^{119}\text{mIn}$  online implantation? The addendum suggests that comparing these with existing measurements for  $^{116},^{118}\text{Sn}$  will result in high precision measurements, but this needs further justification.

The additional information gained by these suggested Moessbauer spectroscopy measurements needs for each case to be described separately. It should be considered, however, that Moessbauer spectra, in addition to the nuclear quadrupole and magnetic interaction, also depend on the isomer shift difference of the nuclear sites somewhat, as well as the measuring temperature. Thus, their interpretation is a little more complicated. We are fortunate that the two leading specialists of the ISOLDE MES collaboration (HM and HPG) have joined our project.

g) Time allowing, we will first take a ME spectrum of the unannealed graphite sample. Comparison with the proposed measurement i) will check the at present necessary assumption of using the corresponding PAC nuQ distribution in graphite from the off-line data with Sb sources also in analyzing the on-line PAC spectra taken with In sources.

The expected spectrum following annealing, simulated in Fig 5, will serve as a check of the correctness of the analysis of the corresponding  $^{118}\text{Sn}$  PAC spectrum (insert in Fig 3). Since the expected quadrupole splitting of the dominant contribution is much larger than the one we had measured in Zn and can be directly compared with the PAC result, an independent relation of the presently obtained values for Q for the 5- states to the one of the  $3/2^+$  Moessbauer state in  $^{119}\text{Sn}$  will be obtained.

Figure 4: Simulation for experiment g)



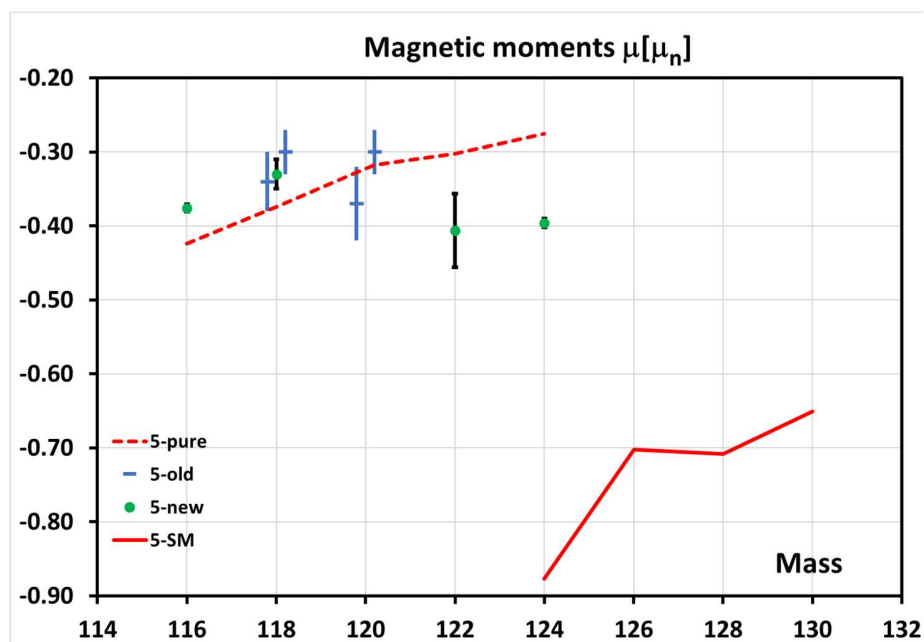
h) This measurement will serve as a check of the magnetic field distribution function as used in fitting the PAC data for  $^{116}\text{Sn}(5^-)$  obtained in the first off-line run in a correspondingly annealed Fe sample.

i) This on-line MS measurement will serve as a check of the magnetic field distribution function as used in fitting the PAC data for  $^{124}\text{Sn}(5^-)$  obtained in the first on-line PAC run in Fe foil not annealed during the experiment. We do not expect an inconsistency, however, that would significantly change the unexpected  $\mu$ .

j) See text in the addendum

**Question 4: It is argued that the magnetic moments of the 5- isomeric states would benefit from additional measurements. But what are the expected theoretical values, what do the shell model calculations predict for example?**

The precision determination of the magnetic moments for the 5- states of 118 and 120 Sn are clearly the most important measurements planned in the present addendum. Here the accuracy possible with the now available techniques, implantation into Gd (hyperfine field about a factor 4 larger than in Fe) and higher external magnetic field available with the MULTIPAC facility, coupled with more efficient PAC spectrometers, will reduce the errors relative to the earlier data (from the 1960s!) by at least a factor of 4. As far as the theoretical understanding goes, the enclosed Fig 6 shows the present situation. For mass 116 to 124 the available experimental values of the neighboring odd Sn isotopes have been used (with some extrapolation) assuming pure  $(1h_{11/2} \times 3s_{1/2})$  configuration and for mass 124 to 130 shell model calculations by the Shanghai group (Y.Y. Cheng et al) are shown. One clearly notes the failure of both approaches. This is still more the case for the nuclear quadrupole moments, where the shell model calculations even predict a wrong sign for Q at mass 124. I am in contact with the Chinese group concerning the possible expansion of their work.



**Question 5: Given the significant overhead to set up RILIS for just one shift of indium, please clarify if surface ionisation (yielding around 5 pA of beam) is a feasible alternative.**

As has been stated on various other occasions, the measurements i) and j), taking a total of one shift from ISOLDE, will only be performed as part of a eMS beamtime with  $^{119}\text{Sn}$ . A specific laser setting for our measurements will therefore not be required. In case such a run will not be scheduled in 2025, we will have to postpone these to a much later time, making the interpretation of our present data, to be published soon, in some respects somewhat uncertain.

**Question 6: Provide more details on how measurements in liquids are performed to allow for a proper risk assessment from a radiological protection (RP) standpoint.**

As stated in the Addendum, we at the moment consider two different possibilities in making the liquid samples required. The actual operations will certainly have to be discussed in detail with the CERN RP professionals. We, however, have extensive experiments in both. The implantation into ice has been used about 50 times in the very successful recent project IS640 and implantation into gallium was in fact used in our first PAC project at ISOLDE (about 1976).

**Question 7: If  $^{119}\text{In}$  requires laser ionization, specify whether a narrow or broad-band laser is needed (in the light of point 1).**

See answer to question 5.

Appendix 1: Summary of the proposed measurements with the code used in manuscript

ISOLDE Beam, UC/RILIS						State of interest			Experiment				
	$I^\pi$	$t_{1/2}$	Int [at/ $\mu\text{C}$ ]	Req [at/samp]	$t_{\text{coll}}$ [min]		$I^\pi$	$t_{1/2}$ [ns]	meas	host	Nr	sa	code
$^{116\text{m}}\text{Sb}$	$8^-$	1 h	$5 \cdot 10^7$	$1 \cdot 10^{10}$	6	$^{116}\text{Sn}$	$5^-$	320	$v_Q$	Gra	16		a)
									$v_Q$	Cd	8		b)
$^{118\text{m}}\text{Sb}$	$8^-$	5.1 h	$1 \cdot 10^8$	$4 \cdot 10^{10}$	30	$^{118}\text{Sn}$	$5^-$	22	$v_Q$	Zn	2		c)
									$v_L$	Ga?	3		d)
									$v_L$	Gd	3		e)
$^{120}\text{Sb}$	$8^-$	5.8 d	$2 \cdot 10^8$	$2 \cdot 10^{11}$	120	$^{120}\text{Sn}$	$5^-$	8	$v_L$	Gd	1		f)
$^{119}\text{Sb}$	$3/2^+$	38 h	$2 \cdot 10^8$	$1 \cdot 10^{11}$	60	$^{119}\text{Sn}$	$3/2^+$	18	$v_Q$	Gra	2		g)
									$v_L$	Fe	2		h)
$^{119\text{m}}\text{In}$	$1/2^-$	18 min	$2 \cdot 10^8$		on line	$^{119}\text{Sn}$	$3/2^+$	18	$v_L$	Fe	4hrs		i)
									$v_Q$	Gra	4hrs		j)



Appendix 2: Correspondence with José Antonio Lay, INTC member

JOSE ANTONIO LAY VALERA [lay@us.es](mailto:lay@us.es) 11/7/2024  
to Heinz Haas <Heinz.Haas@cern.ch>; Georgi Georgiev  
<georgi.georgiev@csnsm.in2p3.fr>; Juliana Schell <juliana.schell@cern.ch>

Dear spokespersons of proposal 550-ADD-1,

I am José Antonio Lay, member of the INTC. I would like to pose you some questions regarding this proposal.

It is a little bit hard to follow so many different options and measurements, and how they complement to what was done. For example, checking with what was approved I find missing in your summary of experiments done the following:

- $^{130}\text{Sn}$  in Zn (heated) and Fe (RT)
- $^{116}\text{Sn}$  in In and Ni
- $^{118}\text{Sn}$  and  $^{120}\text{Sn}$  in Gd
- $^{119}\text{Sn}$  in Graphite, S, Se, and Fe

What prevented at the time to perform such experiments? Why the decision to measure  $^{119}\text{Sn}$  only on Zn was taken? One has to consider that some of these cases are now proposed to be measured: those for  $^{118}, ^{119}, ^{120}\text{Sn}$ . So, why it is expected a better result now?

Regarding Figure 4, one would say that previous experiment has done quite a great job with the Quadrupole moments. The measurements are quite in good agreement with the estimations, maybe  $^{116}\text{Sn}$  is slightly deviated. So, the question here is why to revisit this quadrupole moment measurements? Looks like the effort should be put into the magnetic moment measurements.

My last question would be why to measure  $^{119}\text{Sn}$  twice: from  $^{119}\text{In}$  and  $^{119}\text{Sb}$ . What are the complementarities of the two measurements? For the case of  $^{119}\text{In}$  it is argued that will help with  $^{124}\text{Sn}(5-)$  disagreement with theoretical expectations, but does not seem to be such disagreement from figure 4. You mention that it will help with the interpretation of some data of previous experiment in general but I should have lost which and why. This is quite important because also the TAC is not sure about the  $^{119}\text{In}$  rate according to its comments.

Looking forward to hearing from you.

Yours sincerely,

José A. Lay.



Heinz Haas [Heinz.Haas@cern.ch](mailto:Heinz.Haas@cern.ch) 11/7/2024

To Jose Antonio Lay Valera [lay@us.es](mailto:lay@us.es)

Cc Georgi Georgiev <[georgi.georgiev@ijclab.in2p3.fr](mailto:georgi.georgiev@ijclab.in2p3.fr); Juliana Schell <[juliana.schell@cern.ch](mailto:juliana.schell@cern.ch)>

Dear colleague

Thank you for your advance questions on our addendum 550-ADD-1. It is my pleasure to answer in detail.

- **130Sn in Zn (heated) and Fe (RT):** As specified in the original proposal, narrow-band laser ionization would be required for measurements with  $^{130}\text{In}$  sources. In our first on-line PAC run we had only the broad-band version available. Actually, data have been taken on this mass setting in Fe for a few hours, but the student who tried to make an analysis did not even arrive at consistent half-life spectra. In this context it should be noted that this for the other cases quite successful experiment was the first on-line PAC run for sub-second sources. I am only aware of two other on-line PAC experiments ever performed at ISOLDE, both with much longer source lifetimes and quite simple decay schemes.
- **116Sn in In and Ni:** These rather complex measurements had only been proposed as a basis for experiments with much longer state half-lives. These, however, are clearly not possible with the much reduced shift allocation by the INTC.
- **118Sn and 120Sn in Gd:** At the time of our first off-line measurements we did not have access to a high-quality chemical glove box for the very sensitive target matrix, in particular for the unfortunately quite low implantation energy at the present ISOLDE. As this technical limit will not exist any more soon, the measurements are proposed in the addendum now submitted.
- **119Sn in Graphite, S, Se, and Fe:** Due to the very low stop-gamma energy of the cascade the measurements were much more complex than foreseen with the spectrometer in normal setting. Following several modifications, the spectrum in Zn, for normalization most important, could be obtained. The cases of S and Se have lost their interest as possible matrix. For Graphite and Fe corresponding Moessbauer measurements, a completely different technique, are included in the present addendum.
- **those for 118,119,120Sn:** Apparently there is a misunderstanding of our project. In graphite we have very clear PAC spectra for 118 and 120Sn 5- states concerning quadrupole interaction from our first off-line run. No further data are needed. For these isotopes the proposed new measurements will be to obtain much improved magnetic moments only. For  $^{119}\text{Sn}$  only Moessbauer experiments are proposed. Their purpose is to understand better the effects of the structural damage on the extracted magnetic (for Fe) and quadrupole (for graphite) moments. There is in addition a chance to get an improved ratio of the present cases with the quadrupole moment of the  $^{119}\text{Sn}$   $3/2^+$  state.
- **Regarding Figure 4:** I completely disagree with your impression that the earlier determined quadrupole moments are quite good. They deviate from our values, accurate to the 2-3% level, by almost a factor of two! Clearly your comment on the

magnetic moment discrepancy is justified, however. This actually was the main point to request the new measurements described in the present addendum.

- **measure  $^{119}\text{Sn}$  twice:** This question touches on a central problem of PAC measurements without annealing, clearly the only ones possible for very short-lived source isotopes as in the present pioneering project. In principle the different chemical nature of two implanted atoms might lead to a different defect structure in the environment possibly creating a different hyperfine field distribution. For the case of a dense matrix like Fe one does not expect a large difference, and the two such cases shown in Fig 3 confirm this assumption (the different perturbation amplitude and sign having a nuclear decay origin). For a molecular-type structure like graphite this might well not be the case and requires study, thus two measurements. The comment of the TAC comes from a misunderstanding of our request for  $^{119}\text{In}$ . In the addendum it is clearly stated that the two  $^{119}\text{In}$  measurements (one shift) will be made during a standard ISOLDE Moessbauer run with the same isotope.

I hope to have answered the posed questions to your satisfaction. I am clearly available to give more details if needed.

Sincerely,  
Heinz Haas