### **EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH**

#### **Addendum to the ISOLDE and Neutron Time-of-Flight Committee**

## **Nuclear moments of excited states in neutron-rich Sn isotopes studied by on-line PAC (IS673)**

[Submission date]

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**Abstract:** In the project IS673 we employ the Perturbed Angular Correlation (PAC) technique for measuring nuclear moments of excited states in neutron-rich Sn nuclei. In the two runs performed to date we have concentrated on the 5 $\cdot$  states in  $116$ Sn to  $120$ Sn by off-line PAC from decay of long-lived Sb sources and 122Sn to 128Sn from short-lived In isotopes by on-line PAC.

Though a large amount of new information has been obtained, analysis of the existing data has shown that a new series of off-line measurements could much improve the precision of the magnetic and quadrupolar moments. We therefore propose a further run using the Sb isotopes, partly with improved experimental facilities

In addition, we plan to apply the Mössbauer technique using 119Sb to augment our data. This will also lead to a more reliable nuclear quadrupole moment for the 3/2<sup>+</sup> state of <sup>119</sup>Sn and solid-state information on the technologically important system Sb in graphite.

**Summary of requested shifts:** 6+1 shifts

### **Motivation**

The overall motivation for the ongoing project IS673 has been detailed in the original project proposal [1] and shall not be reproduced here. Instead, we concentrate on the results obtained to date and the possibility to improve the results for the nuclear moments, both quadrupolar and magnetic, of the 5- states in 116,118,120Sn that form the basis for the studies of the more neutron-rich Sn isotopes with the on-line PAC technique. A short summary of our existing experimental results from the first two experimental runs concentrating mostly on the 5-states of predominantly ( $v h_{11/2} \otimes v_{1/2}$ ) configuration will be presented first. The first, off-line run, October 2021, was performed with Sn nuclei in decay of 116,118,120Sb isotopes with reasonably long halflives. For the study of the heavier Sn isotopes in a second run, June 2023, the short halflife of the necessary 122,124,119In isotopes required the use of an on-line PAC setup, practically forbidding sample annealing preceding the measurement.

The still lacking possible full precision for the nuclear moments obtained will here be documented and a series of experiments proposed to get still more precise and fully reliable values.



**Summary of experiments performed** (source and state half-life, T<sub>m</sub>, T<sub>ann</sub> and result)

Measurements are done from decay of parent isotopes Sb or In on Sn excited state "i" RT= Room Temperature, ap = applied external magnetic field (polarization)

### **Quadrupole interaction results**

The quadrupole moment (Q) of the 5 isomeric state has been determined in a series of Perturbed Angular Distribution (PAD) measurements of various high spin states in Sn isotopes [2] using Cd at high temperature as matrix, applying the then known B(E2)

value [3]. With the later somewhat changed B(E2) value [4] one calculates Q=.304(8) b. This number will serve as reference for all the other states. For this purpose, the quadrupole interaction frequencies  $(v_0)$  must be measured in the identical matrix (and temperature). Zn being known to be the simple metal matrix with the highest  $v_0$ , this was our original choice. A direct measurement, unfortunately, was not successful, partially from run time limitations and the also the difficulty in proper annealing of Sb. The necessary  $v_0$  may be obtained, however, from the electric field gradient (efg) ratio between Zn and Cd determined from on-line PAC of the 5- state in 124Sn following In implantation, see Fig 1.





 $118\text{Sn}(5)$ : The PAC measurements for implanted  $118\text{mSb}$  in Zn single crystals all show a similar G(t) decrease in the time range of 10 to 50ns as expected. The actual shape, however, depends somewhat on the annealing conditions used. The fitted perturbation function for the most trustworthy case has been analysed and a preliminary value of  $v_0$ obtained. For implantation into graphite, where the highest, but not unique,  $v_0$  was observed, the PAC spectra are presented in Fig 2. Clearly, the annealing at 450C has produced a practically unique site with a high interaction frequency. This observation had not really been expected and primarily triggered the present proposed new off-line experiment series.

 $120$ Sn(5<sup>-</sup>): After implantation into graphite, and also following the identical annealing, only the initial decrease of the perturbation function can be observed. Since, however, the exact functional form of G(t) is known, a relatively precise ratio of O relative to <sup>118</sup>Sn can be obtained.

 $122$ Sn(5 $\cdot$ ): For technical reasons, the experiments had to be performed at room temperature in graphite and obviously no annealing following the In implantation was possible. Not having a better possibility at present, the observed perturbation was fitted with the one observed for <sup>118</sup>Sn following Sb implantation, leading to a considerably smaller Q. A more reliable quantitative description would require that the functional form to be expected under these conditions. This would first have to be studied following In implantation for a case where Q is known. The corresponding experiment using 118Sn produced from In has been suggested in our original proposal. It is, however, technically quite demanding and could not be included in the first on-line PAC run.

 $124$ Sn(5 $\cdot$ ): For this isomeric state the on-line PAC technique has resulted in very precise results, also partly because the observed state half-life is practically twice as large as the one given in the literature. The perturbations measured for RT implantation of 124In in Zn and Cd, shown in Fig 3, demonstrate that no attenuation is present and very precise interaction frequencies have been obtained. From the ratio of the two one obtains a reliable  $v_0$  value for the case of  $116Sn$  in Zn at RT, practically only limited by the corresponding number in Cd.

 $119\text{Sn}(3/2)$ : In order to demonstrate the use of on-line PAC for implantations into Zn at RT, a measurement of  $v_0$  was also performed following implantation of  $119mIn$ . The fitted frequency is in full agreement with the previously obtained value from ISOLDE Mössbauer data [5].

### **Magnetic interaction results**

<sup>116</sup>Sn(5-): On implantation into iron foil and annealing the observed R(t) had the expected shape of a magnetic interaction experienced by about 70 % of the nuclei (see Fig 3) in the known magnetic hyperfine field [6] obtained from 119Sn Mössbauer spectroscopy. As the extracted magnetic moment agrees with the one from PAD experiments in an external magnet [3], one has to conclude that between 116Sn(5-) and  $119Sn(1/2<sup>+</sup>)$  no appreciable hyperfine anomaly exists.



Fig 3: Results for <sup>116</sup>SbFeann and 124InFe

 $118Sn(5)$ : Since measurements in a nonpolarized iron sample did not allow a precision magnetic interaction due to the relatively short nuclear state half-life, an experiment in a polarizing magnet was performed using a 5-detector PAC setup. A clear pattern was observed with this configuration that shows only the expected precession at two  $\omega$ <sub>L</sub>.

 $122Sn(5)$ : Though the quite short state half-life limits the obtainable accuracy, a fit to the functional form obtained for RT implantation of 124In into iron (Fig 3) can be considered as reliable.

 $124$ Sn(5<sup>-</sup>): The on-line PAC measurement after  $124$ In implantation into iron, shown in Fig 3, allows a precision determination of the magnetic moment, assuming that the effective hyperfine field is identical to the one acting in an annealed foil after Sb implantation, as observed in the off-line measurement for 116Sn(5-).

### **Summary of nuclear moment results**

The values for the nuclear moments obtained from the two runs of IS673 are presented in Fig 4, where they are also compared to previous results. Tentative values applying additivity relations with the corresponding nuclear moments of the neighbouring odd-A Sn isotopes [7] can qualitatively explain the observed positive trend (red lines) for the quadrupole moments. Only a minor discrepancy is noted for the moment ratio between mass 116 and 118. The tentatively seen negative trend in the data for the magnetic moments, however, goes against an equivalent treatment.





The major problems of the present data are accurate numbers for the 116Sn to 118Sn quadrupole moment ratio and the RT value of the  $116Sn$  n<sub>0</sub>, both being related to our failure in obtaining precision data from implantations into Zn. The absence of accurate values for the 118Sn and 120Sn magnetic moments and the missing understanding of the unpredicted trend from mass 116 to124 is also troubling.

In order to address these problems and improve the accuracy of several cases, we here propose a series of new off-line PAC measurements and new off-line Mössbauer experiments using implantation of 4 different Sb isotopes. These would be performed employing a variety of experimental facilities existing at ISOLDE in a run lasting 2 days. In addition, we plan two measurements as participants of a run of the ISOLDE on-line Mössbauer project using In beam, taking about 8 hours.

## **Proposed new off-line PAC and Mössbauer measurements**

These experiments with Sb isotopes implanted into the solids will require annealing of the samples prior to the PAC measurement. As the necessary procedures are now mostly known from our previous run, we here propose 8 new measurements with:

a) Measuring  $v_0$  for  $116Sn(5)$  in graphite

We have a precise value of  $v_0$  for mass 118 measured in graphite. A spectrum for  $116Sb$ , annealed in a way identical to the one found for 118Sb, will fix the somewhat imprecise ratio  $Q(118)$  to  $Q(116)$  mentioned above. Unfortunately, this experiment is the most difficult of the ones suggested here. It could require the use of up to 16 samples to obtain the necessary statistical reliability.

b) Measuring  $v_0$  for  $116Sn(5)$  in Cd

Since all nuclear quadrupole moments determined in project IS673 at present depend on the available PAD data for the quadrupole frequencies for 116Sn(5-) in Cd at higher temperatures, extrapolated to RT. A direct measure of this quantity would remove the inherent inaccuracy [8][9]. For this reason, we propose to measure the corresponding frequency directly after implantation of 116Sb followed by a moderate annealing procedure. Though a direct proof of this is still missing, from general experience a successful annealing should be much more likely in Cd than in Zn.

c) Remeasuring  $v_0$  for  $118Sn(5)$  in Zn

In our initial project the use of Zn, where the highest value for simple metals was expected optimal for a consistent determination of the  $Q(118)$  to  $Q(116)$  ratio was expected. Since somewhat inconsistent results were obtained, we now propose to determine this value in Cd, assuming the Cd implantations in experiment b) (above) are successful. Even without a positive output from experiment a) a more consistent value for this number would be important. The samples and annealing procedures would obviously have to be fully identical to these used in b).

d) Measuring an accurate magnetic moment for  $118Sn(5)$  in liquid

In principle the measurement of a magnetic moment in a high magnetic field externally applied is the most reliable PAC method, in particular when liquid sources can be used. At ISOLDE a facility has been constructed with a field of 8.5 T, practically identical to the one acting in our measurement of this moment in polarized iron, where a spectrum of high statistical accuracy could be obtained. We now propose such a measurement in liquid Ga (or water) using the MULTIPAC [10] facility.

e) Studying  $v<sub>L</sub>$  for  $118Sn(5<sup>-</sup>)$  in Gd

Observing the magnetic precession in the known [11] strong hyperfine field for Sn in Gd should be another approach to determine a precise value for m, assuming a successful annealing procedure can be found.

f) Measuring the magnetic moment for 120Sn(5-) in Gd

A precise value for  $\mu$  of 5 $\cdot$  in  $^{120}$ Sn can only be hoped for if the previous tests e) with <sup>118</sup>Sb implantation into Gd have been successful, even though such a PAC measurement can be performed with unprecedented statistical relevance. A measurement as d) would be a possible alternative, but necessarily less precise due to the much smaller half-life to magnetic field ratio.

g) Mössbauer measurement for  $119Sn(3/2<sup>+</sup>)$  in graphite after Sb implantation

The Mössbauer spectrum for 119Sb implanted graphite, annealed in the same way as the corresponding mass 116 (or 118) PAC spectrum, (Simulation below) will directly deliver the ratio of the corresponding quadrupole moments to the one of  $119Sn(3/2<sup>+</sup>)$ , with an accuracy considerably greater than the one available to date. Thus, Q for the Mössbauer state will be determined in a completely different way. Our previous approach along a similar path in the analysis of a very early ISOLDE experiment [5] had determined Q = .109 b, much different from the one accepted at present [12], .132(1) b [13]. In addition, a study of the annealing behaviour of the graphite-Sb system, of possible technological

interest for the efficiency of Li batteries [14], could easily be done within such a Mössbauer study.

h) Mössbauer spectrum for  $119Sn(3/2<sup>+</sup>)$  in iron after Sb implantation

A precise measurement of the effective hyperfine field distribution as function of annealing procedure for Sb implanted into Fe is proposed, a system that has already been roughly studied with Mössbauer spectroscopy earlier [15]. The results will complement our existing 116Sn PAC measurement and its interpretation.

## **Proposed on-line Mössbauer measurements**

i) Mössbauer spectrum for  $119Sn(3/2<sup>+</sup>)$  in iron after In implantation

This measurement will directly allow a study of the magnetic hyperfine field distribution acting at Sn following implantation as In at room temperature, which might be different from the one known to act in annealed samples, used in our present analysis. This might possibly explain the inconsistency of the magnetic moment for 124Sn(5-) determined from our on-line PAC measurement with theoretical expectations.

j) Mössbauer spectrum for  $119Sn(3/2<sup>+</sup>)$  in graphite after In implantation

The measurement of the efg distribution following on-line implantation of 119In, to be studied by Mössbauer spectroscopy, will help to interpret existing data of the first online PAC experiment of IS673, but also in preparing future such studies. Due to the presence of isomer shift effects in the data, however, it cannot fully replace the difficult measurement of this effect using on-line PAC in 118In decay as suggested in the original proposal.



### **Summary of requested implantations and proposed measurements**

## **References:**

[1] Haas, H., Georgiev, G., et al.: Nuclear moments of excited states in neutron rich Sn isotopes studied by on-line PAC. CERN-INTC-2020-022/INTC-P-550

[2] Bertschat, H., Echt, O., *et al*.: Quadrupole moments of high spin isomeric states in Sn isotopes. Hyp. Int. **2**, 326 (1976)

<https://doi.org/10.1007/BF01021160>

[3] Ishihara, M., Broda, R., Herskind, B.: Isomeric states in the Sn isotopes. Proc. Int. Conf. Nucl. Phys. **1**, 256 (1973)

[4] Broda, R., Mayer, R. H., et al.: Yrast Isomers in Tin Nuclei from Heavy Ion Collisions and the *h*11/2 Subshell Filling. Phys. Rev. Lett. **68**, 1671 (1992) <https://doi.org/10.1103/PhysRevLett.68.1671>

[5] Haas, H., Menningen, M., *et al*.: EFG sign for Sn in Zn, Cd, and Sb. Hyp. Int. **15**, 215 (1983)

<https://doi.org/10.1007/BF02159741>

[6] Price, D. C.: The temperature dependence of the hyperfine interactions at 119Sn in iron. J. Phys. F **4**, 639 (1974) <https://iopscience.iop.org/article/10.1088/0305-4608/4/4/017/pdf>

[7] Yordanov, D. T., Rodriguez, L. V., *et al.*: Structural trends in atomic nuclei from laser spectroscopy of tin. Phys. Comm. **3**, 107 (2020) <https://doi.org/10.1038/s42005-020-0348-9>

[8] Echt, O., Haas, H. *et al*.: Temperature dependence of the quadrupole interaction of tin in cadmium. Hyp. Int. 2, 273 (1976) https://www.osti.gov/etdeweb/biblio/7223279

[9] Ionescu-Bujor, M., Iordachescu, A., *et al*.: Lattice damage at recoil tin impurity in metallic cadmium. Hyp. Int. 11, 171 (1981)

[10] H-Schell, J., Lupascu, D. C., et al.: MULTIPAC-Setup for Perturbed Angular Correlation Experiments in Multiferroic (and Magnetic) Materials: proof-of-concept. CERN-INTC-2023-012/INTC-I-249

[11] Gotthardt, V., Möller, H. S., Mössbauer, R. L.: Magnetic field at 119Sn in gadolinium. Phys. Lett. A **28**, 480 (1968)

[12] Pykkö, P.: Year-2017 nuclear quadrupole moments. Mol. Phys. **116**, 1328 (2018) [https://doi.org/10.1080/00268976.2018.1426131](https://doi.org/10.1080/00268976.2018.14261319)

[11] Barone, G., Mastalerz, R, *et al*.: Nuclear quadrupole moment of 119Sn. J. Phys. Chem. A **112**, 1666 (2008) <https://doi.org/10.1021/jp710388t>

[14] Liu, Q., Fan, L., et al.: Antimony-Graphite Composites for a High-Performance Potassium-Ion Battery. Energy Technol. **7**, 1900634 (2019

[15] de Waard, H., Hafemeister, D. W., *et al*.: Large increase of magnetic hyperfine fields of 5 sp-shell impurities in ferromagnets after vacancy trapping. Phys. Rev. B **24**, 1274 (1981)

<https://doi.org/10.1103/PhysRevB.24.1274>

# **Details for the Technical Advisory Committee**

# **3.1 General information**

Describe the setup which will be used for the measurement. If necessary, copy the list for each setup used.

⊠ Permanent ISOLDE setup: *Moessbauer and digital PAC spectrometers*

⊠ To be used without any modification

☐ To be modified: *Short description of required modifications.*

☐ Travelling setup (*Contact the ISOLDE physics coordinator with details.*)

☐ Existing setup, used previously at ISOLDE: *Specify name and IS-number(s)*

☐ Existing setup, not yet used at ISOLDE: *Short description*

☐ New setup: *Short description*

# **3.2 Beam production**

For any inquiries related to this matter, reach out to the target team and/or RILIS (please do not wait until the last minute!). For Letters of Intent focusing on element (or isotope) specific beam development, this section can be filled in more loosely.

• Requested beams:



- Full reference of vield information: IS673 and standard Mössbauer runs
- Target ion source combination: UC/RILIS
- RILIS? *yes*

☐ Special requirements: (*isomer selectivity, LIST, PI-LIST, laser scanning, laser shutter access, etc.*)

• Additional features?

☐ Neutron converter: (*for isotopes 1, 2 but not for isotope 3.*)

☐ Other: (*quartz transfer line, gas leak for molecular beams, prototype target, etc.*)

- Expected contaminants: *Isotopes and yields*
- Acceptable level of contaminants: (*Not much sensitive to stable contaminants* )
- Can the experiment accept molecular beams?
- Are there any potential synergies (same element/isotope) with other proposals and LOIs that you are aware of? All Mössbauer projects

# **3.3 HIE-ISOLDE**

For any inquiries related to this matter, reach out to the ISOLDE machine supervisors (please do not wait until the last minute!).

• HIE ISOLDE Energy: *(MeV/u); (exact energy or acceptable energy range)*

⊠ Precise energy determination required

☐ Requires stable beam from REX-EBIS for calibration/setup? *Isotope?*

• REX-EBIS timing

⊠ Slow extraction

- $\Box$  Other timing requests
- Which beam diagnostics are available in the setup?
- What is the vacuum level achievable in your setup?

# **3.4 Shift breakdown**

The beam request only includes the shifts requiring radioactive beam, but, for practical purposes, an overview of all the shifts is requested here. Don't forget to include:

• Isotopes/isomers for which the yield need to be determined: None

• Shifts requiring stable beam (indicate which isotopes, if important) for setup, calibration, etc. Also include if stable beam from the REX-EBIS is required. No

An example can be found below, please adapt to your needs. Copy the table if the beam time request is split over several runs.

## **Summary of requested shifts:**



# **3.5 Health, Safety and Environmental aspects**

## **3.5.1 Radiation Protection**

• If radioactive sources are required:

- **–** Standard sealed gamma test samples
- **–** Na22, 133Ba
- **–** Activity?
- **–** Sealed
- For collections:
	- **–** Number of samples: up to 36
	- **–** Activity/atoms implanted per sample: typically 10<sup>10</sup> at/sample
	- **–** Post-collection activities? (*annealing in noble gas or vacuum.*)

### **3.5.2 Only for traveling setups**

- Design and manufacturing
	- ⊠ Consists of standard equipment supplied by a manufacturer

☐ CERN/collaboration responsible for the design and/or manufacturing



# • Describe the hazards generated by the experiment:



