EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

The neutron capture cross section of ^{124}Sn and its impact on neutrinoless double β decay searches

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Abstract: The search for neutrinoless double β decay ($0\nu\beta\beta$) - a very exotic process that may clarify if the neutrino is a Dirac- or a Majorana-type fermion - is, like all rare-event experiments, extremely sensitive to background contributions (for instance the subsequent γ cascade following a neutron capture reaction). Several possible candidates were identified for this exotic process with ^{124}Sn being one of them. A precise study of the neutron capture channel on ^{124}Sn - including the capture yield but also spectroscopy of the prompt γ's de-exciting the capture state in ^{125}Sn, would provide valuable information related to the neutron interaction with this target nucleus: neutron capture probability and γ cascades. Such data can then be used in better assessing (and subtracting) the neutron-induced background in neutrinoless double β decay experiments. To that end, here we propose to measure the ^{124}Sn(n,γ) cross section from thermal up to the highest achievable neutron energy by sTED detectors in n_TOF's EAR2. For future work, a possible Addendum to this proposal would be a spectroscopy experiment aiming to characterise prompt γ cascades de-exciting the capture state in ^{125}Sn with HPGe and/or LaBr$_3$ detectors in EAR1.

Requested protons: 2.0x10^{18}
Experimental Area: EAR2

Introduction and Scientific Motivation

Neutrinoless double β decay ($0\nu\beta\beta$) is an exotic decay mode never observed experimentally but energetically allowed for certain isotopes. If confirmed experimentally, this will clarify a long-standing open question in elementary particle physics related to the Majorana or Dirac nature of the neutrino, as the latter is the only fermion which can be a Majorana-type particle [1]. This will have important implications not only for the neutrino physics, and therefore the Standard Model, but also for connected fields like (cold) dark matter searches as Majorana fermions are one of the candidates considered for such searches [2]. Up to now, only 35 possible candidates have been identified for this very rare process [3]. The reaction rates of the double beta ($2\nu\beta\beta$) and $0\nu\beta\beta$ decay modes are proportional to Q^{11} and Q^5, respectively [4]. For this reason, only high-Q-value reactions (typically above 2 MeV) are currently worth studying for $0\nu\beta\beta$ decay searches [4]. This fact restricts the 35 possible candidates to only 11 feasible cases, one of them being ^{124}Sn [3, 4].

The present proposal aims at providing relevant neutron capture data for $0\nu\beta\beta$ decay searches on ^{124}Sn. In this type of experiments, ^{124}Sn is used as both the source and the detector medium to investigate the $0\nu\beta\beta$ decay of ^{124}Sn to ^{124}Te, ($T_{1/2} > 2.4 \times 10^{17}$ years) [3–6]. Historically, ^{124}Sn was the first nucleus employed to study the $2\nu\beta\beta$ decay process using Geiger counters [7]. More recently, an experiment searching for the $0\nu\beta\beta$ decay of ^{124}Sn was performed by M. J. Hwang et al. [5] in Korea where they employed an active-source detector technique using a Tin-loaded liquid scintillator. The experiment yielded inconclusive results. Also, efforts have been initiated to search for the $0\nu\beta\beta$ decay of this nucleus at the upcoming underground facility of the India-based Neutrino Observatory (INO). They plan to use an enriched ^{124}Sn cryogenic superconducting bolometer: TIN.TIN (The INdia-based TIN detector). This detector is currently under construction at the Tata Institute of Fundamental Research (TIFR) and its (expected) resolution should be $\sim 0.2\%$ (4 keV) at 2 MeV [6,8,9].

Over the years several experiments made great strides in the race to set limits on or potentially measure the $0\nu\beta\beta$ reaction. The overall sensitivity of this type of experiments depends on several factors like detector mass and resolution, the isotopic abundance of the isotope under study, level of background, etc. [4–18]. A common challenge to all $0\nu\beta\beta$ decay searches is the extremely high sensitivity to background. The background events in the energy region of interest (ROI) are defined by a narrow energy band, centered on the Q-value of the reaction: 2292.7(4) keV for ^{124}Sn [19]. The ROI is mainly determined by the resolution of the detection system [5, 9]. Getting a low background level requires not only extremely radio-pure targets and shielding materials to minimize internal radiation but also sophisticated measures to minimize external contributions. Especially the neutrons generated by deeply-penetrating cosmic rays and/or (α,n) reactions following α-decay events of ^{nat}U and ^{nat}Th from the rocks are an issue. These neutrons thermalize and can induce capture reactions inside the ^{124}Sn detector material. If such capture events produce γ rays in the ROI, they can *mimic* the $0\nu\beta\beta$ decay signals of interest.

Neutron capture by ^{124}Sn leads to ^{125}Sn which has a both beta-unstable ground state, with $T_{1/2} = 9.64(3)$ d, and also an isomeric first excited state with $T_{1/2} = 9.52(5)$ m. For ^{125m}Sn, there is no isomeric transition, so the spectroscopic study of the decay of both the isomeric and ground states can only be done through the detection of γ transitions in ^{125}Sb [$T_{1/2}$: 2.75856(25) y] populated following the β^- decay of ^{125}Sn. The former then further decays through another β emission to ^{125}Te (stable) [19]. It may be noted that the neutron separation energy in ^{125}Sn is 5733.50(20) keV [19].

The authors of Ref. [10] show that, after bombarding a ^{124}Sn sample with a (reactor-induced) neutron thermal flux and measuring the delayed γ rays following the β decay of ^{125}Sn (produced by neutron capture on ^{124}Sn) with a HPGe detector, a strong summing peak of 2288.2 keV is observed in the spectra (see Figure 7 of Ref. [10]). This peak is generated by summing several γ-ray energies, coming mostly from ^{125}Sb but also from ^{125}Te, and it has a very close energy to the Q-value of the $0\nu\beta\beta$ decay of ^{124}Sn: 2292.7(4) keV. Moreover, the authors also compare the experimentally-observed yield (counts per keV) coming from this summing peak with a simulation yield and get a consistent 30% difference between the two (see Table 4 of Ref. [10]). No tentative explanation is given as for the cause of this difference but it could come from a poorly known neutron capture cross section on ^{124}Sn used in the Monte Carlo simulations. We note that in such rare-event experiments the (known) background in the ROI is solely subtracted by means of simulations and, therefore, a (not understood) 30% difference is a very serious drawback. A new neutron capture measurement mapping the resolved resonance region (RRR) up to 50-100 keV, where the strongest compound nucleus resonances are, may help improve the agreement between the simulated and experimental yields discussed in Ref. [10].

We point out that the authors used activation techniques to measure the γ yield in the ROI (that is, they measured only the *delayed* γ rays mostly from the longer-lived daughter nucleus ^{125}Sb and from ^{125}Te). Obviously, this technique missed the prompt γ rays decaying from the capture state in ^{125}Sn. These transitions, or their summing, can (potentially) also induce background in the ROI. Consequently, we may consider a future Addendum proposal to the present one to measure these prompt γ's in EAR1 using HPGe and/or $LaBr_3$ detectors. To conclude, the capture channel on ^{124}Sn (both its probability and the prompt γ rays) is an issue to be clarified if one is to judge the feasibility of measuring the $0\nu\beta\beta$ decay of this nucleus.

In the review paper [20] several Monte Carlo codes (MCNP, OpenMC, Monteburns, etc.) are employed aiming to reproduce experimental benchmarks on nuclear fuel depletion. The results of the MC codes for the Computed Concentrations (C)/Experiment (E) ratios yield less than 15% accuracy for important actinides, including uranium and plutonium, but also for certain fission products. However, the authors point out that there are also some nuclides which are notoriously difficult to predict, one of them being ^{125}Sb. In the various studies discussed in this review paper, it was found that the computed concentrations (C) of Antimony (^{125}Sb) were initially off by more than 250% [21], then improved to 50% [22], compared to the experimentally-known (E) values. The source of such discrepancies was conjectured to be related to the multiple ^{125}Sb production pathways in the reactor that are inadequately simulated. For ^{125}Sb, the two common production routes are fission and (n,γ) on ^{124}Sn followed by the β decay of ^{125}Sn. Tin can usually be found in minute quantities in the fuel cladding and ^{125}Sb is present in the irradiated cladding or in moderator tubes fabricated of Zircaloy. The (still open) question is if whether or not the data libraries alone are responsible for the observed ^{125}Sb divergence.

Because of all the reasons discussed in previous paragraphs, improving the evaluation of the neutron capture channel on ^{124}Sn would be very helpful for all research activities relying on the quality of such nuclear data. We would like to support this effort with a new data set measured at n_TOF that might yield further progress in clarifying these issues.

Status of the ^{124}Sn(n,γ)^{125}Sn cross section data

A survey in EXFOR shows the data plotted in Figure 1, together with the evaluated values for this reaction (in green). It can be seen that several data points were measured in the thermal region, below the first resonance, and above 10 keV, mostly through activation techniques. These cross section points are reported with fairly large uncertainties (10-20%) [23–34].

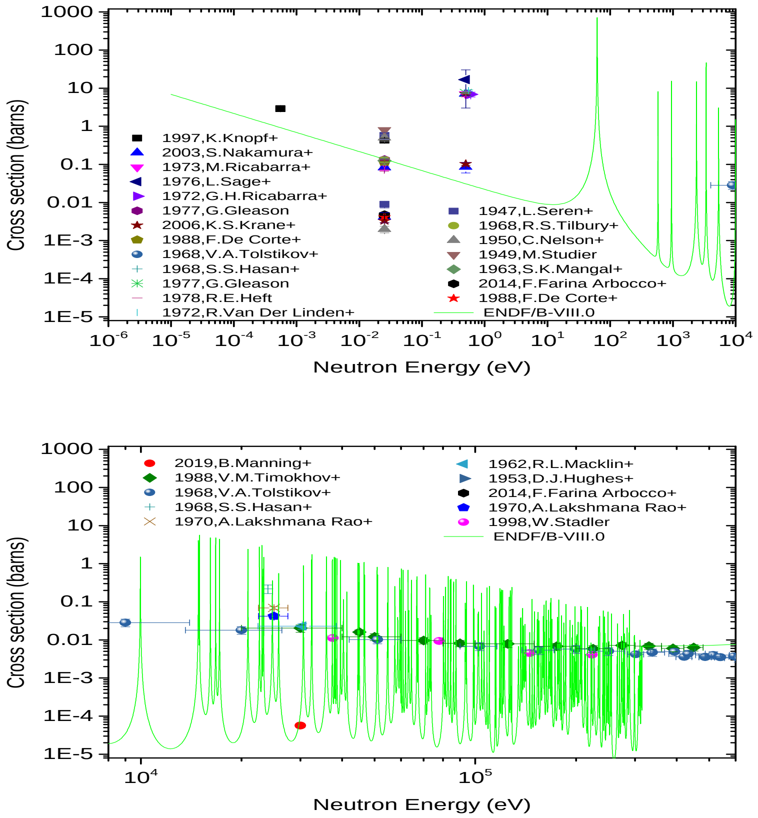

Figure 1: Comparison of the ENDF/B-VIII.0 evaluation with cross-section data available in the EXFOR database for the ^{124}Sn(n,γ)^{125}Sn reaction.

In the 10 to few hundreds keV region there are also a few time-of-flight measurements but they, unfortunately, employed only very short flight paths (a few meters). This translated into very poor incident energy resolution. It is important to highlight that in the RRR, especially below 10 keV, there is only one time-of-flight-based data set measured at MLF-J-PARC neutron facility by A. Kimura et al. [35]. The authors employed a 21.5-m flight path and HPGe detectors to construct the neutron capture cross section for several tin isotopes, including ^{124}Sn. Unfortunately, they only measured up to 4 keV and did not send the extracted data to EXFOR (only the conference proceedings referenced above is available). Moreover, the authors only report the detection of the first strong resonance in ^{124}Sn, at 62 eV, and not of the next two, at 579 and 950 eV. This is, indeed, very puzzling because these two resonances were actually observed in high-resolution transmission experiments (measuring the total cross section) reported by Fuketa [36] and Yu.V. Adamchuk [37] (we note however that the authors sent to EXFOR only the resonance parameters and not the actual total cross section points). There is also one transmission measurement above 10 keV performed at ORNL reported by R.F. Carlton et al. [38]. The S.F. Mughabghab et al. [39] compilation, and the ENDF.B-VIII.0 evaluation shown in Figure 1, are based on the resonance parameters extracted in these transmission experiments.

In summary, there are no high-resolution time-of-flight neutron capture data on ^{124}Sn in the RRR. Taking advantage of the very high neutron flux of EAR2, a new measurement of the (fairly weak) ^{124}Sn neutron capture channel - complementary to the transmission data mentioned above - will therefore both provide, for the first time, good resolution capture data able to map out the CN resonances in the RRR up to (at least) 15-20 keV and will also help clarify the existence of the two resonances, at 579 and 950 eV, seen thus far only in transmission.

Furthermore, as can be seen from Figure 2, there is a large disagreement between several evaluations associated to this reaction channel (even differences as high as 100% can be observed). Moreover, JENDL-4.0 and CENDL-3.2 show two resonances, at 6950 and 8720 eV, which are missing from the other evaluations. A new measurement at n_TOF may also help clarify these issues, at least partially.

Experimental details and difficulties

We plan to measure the neutron capture cross section of the stable ^{124}Sn isotope with an array of 9 sTED detectors at the n_TOF EAR2 (see Figure 3), exploiting the total energy technique based on the so-called Pulse Height Weighting Technique (PHWT). Such a setup will allow us to gather good statistics in a reasonable beam time while at the same time taking advantage of the well-known low neutron sensitivity of sTED detectors. An important difficulty of the proposed experiment is the (very) unfavorable elastic to capture cross section ratio [40] giving rise to large corrections for elastically-scattered neutrons which are then captured by surrounding materials and/or the detector itself. The natural abundance of ^{124}Sn, 5.79%, is fairly small as compared to the dominant isotopes (which all have comparable neutron capture cross sections). Therefore, the use of a highly enriched sample is of prime importance. We plan to use a ^{124}Sn sample enriched to 97.9%. This

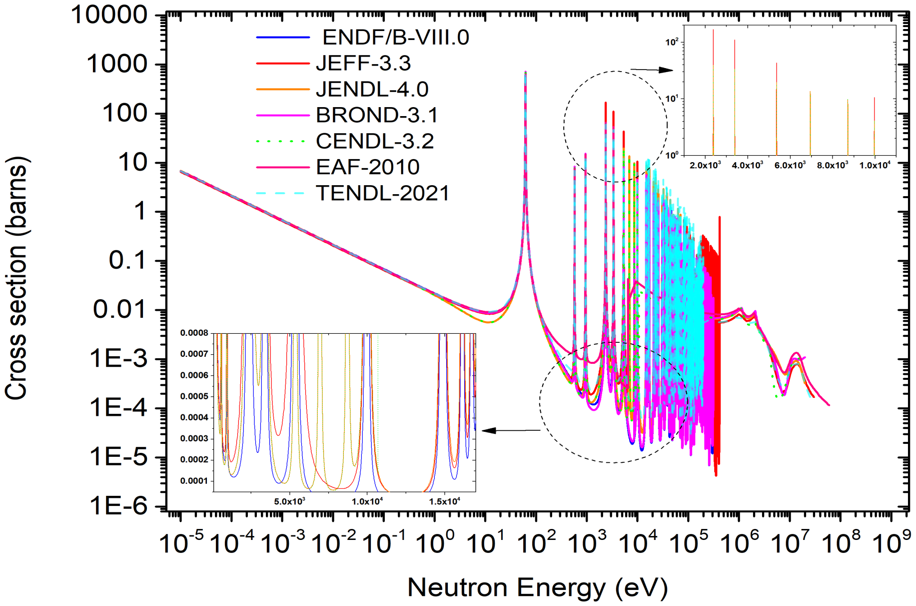

Figure 2: Evaluated cross sections associated to the ^{124}Sn(n,γ)^{125}Sn reaction.

enrichment should be high enough so that the polluting contributions coming from the resonances of other tin isotopes be negligible (see Figure 4). We will however keep this under control by also irradiating a ^{nat}Sn sample. This will also help us cross-check the enrichment claimed by the target manufacturer. We mention that both sample materials (3 g of enriched ^{124}Sn and 25 g of ^{nat}Sn) are already available.

Figure 5 showcases the broadening of the resonances due to both the resolution function and neutron multiple scattering (the latter quantified using the resonance analysis code SAMMY [41]) for a 1-g sample (0.00618 at/b). One can also notice that the first, most intense resonance at 62 eV is saturated by the 1-g sample. Hence, we can use it for self-normalization purposes and so to cross-check the standard ^{197}Au-based normalization routinely used at n_TOF. However, to extract clean parameters for this saturated resonance, we need a thin-thick sample approach. We therefore plan to also measure a 0.1-g sample (1.1x10^{17} of proton flux should be enough to get decent statistics for this very intense resonance). Unfortunately, due to the resolution function broadening, for neutron energies above 20 keV the count estimate (discussed below) on this isotope decreases severely (see Figure 6). To increase our chances of observing these weaker resonances, or at least to extract average cross sections corresponding to the 20-100 keV region while also keeping the beam time at reasonable values, we will also measure for 4.8x10^{17} proton flux a 3-g sample. This measurement may be even more relevant if we manage to substantially increase the sTED array efficiency (the collaboration is currently testing an upgrade to 27 sTED's [42]).

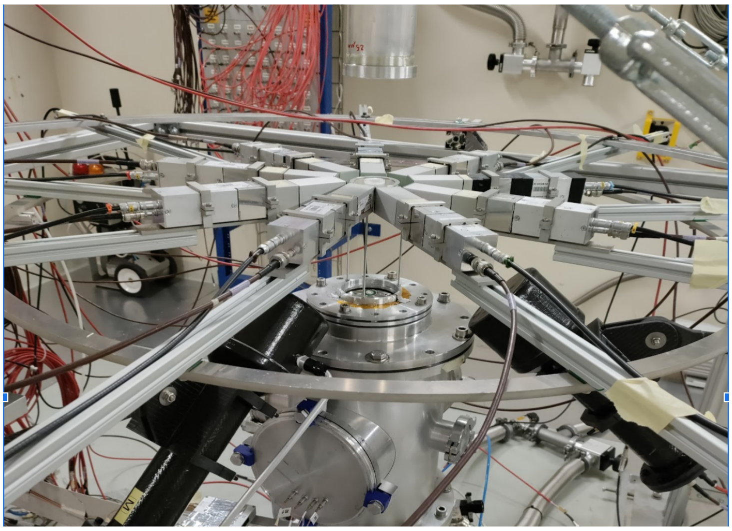

Figure 3: The 9 sTED detector array in EAR2 used during the 2024 ^{209}Bi and ^{146}Nd campaigns [43, 44]

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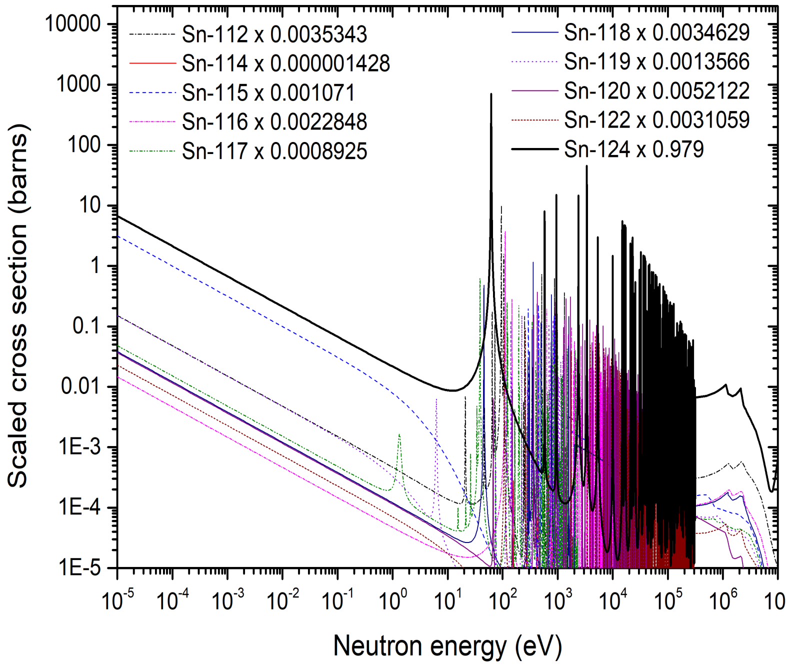

Figure 4: Scaled cross section associated to the other tin isotopes (scaled by the abundances of the ^{124}Sn enriched sample). One can see that a 97.9% enrichment (in black) is enough to make the polluting resonances coming from the other tin isotopes negligible.

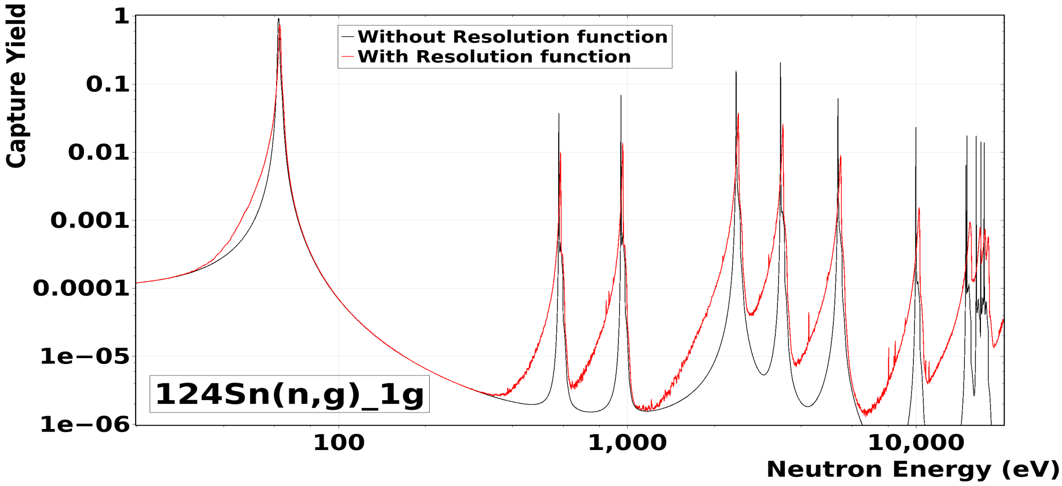

Figure 5: The impact of the resolution function and neutron multiple scattering (from SAMMY) for a 1-g ^{124}Sn sample in the EAR2 capture position.

Figure 6 shows the count rate we expect during the ^{124}Sn(n,γ) experimental campaign using 0.1 g and 1 g samples along with the main background components in EAR2 (elastic + in-beam γ rays + empty). The plot makes use of the evaluated capture cross section for this target nucleus given by JEFF-3.3 and of the most recent evaluation of the neutron flux in EAR2. We used an assumed 4.5% γ cascade efficiency for the proposed setup. The counts from Figure 6 translate into decent statistical uncertainties associated to the integral of the most intense resonances above the background level in the resolved resonance region up to 15-20 keV. However, one can clearly see that unfortunately above this incident energy the background starts dominating over the 1-g sample counts.

In summary, our proton budget request is divided as follows:

- **1.1x10^{17} protons** to extract clean parameters for the first resonance using a *thin* (0.1 g) sample,
- **6.5x10^{17} protons** to map out the RRR below 10-20 keV using a *thick* (1 g) sample,
- **4.8x10^{17} protons** to, at least, extract an average cross section for the 20-100 keV region using a 3-g sample,
- **1.1x10^{17} protons** for the assessment of possible contaminants coming from the other tin isotopes using a ^{nat}Sn sample,
- **5.4x10^{17}** for ancillary measurements dedicated to background subtraction, and finally
- **1.1x10^{17}** for normalization purposes using a ^{197}Au sample.

Therefore, we request a total of 2.0x10^{18} protons for the full experiment in n_TOF's EAR2.

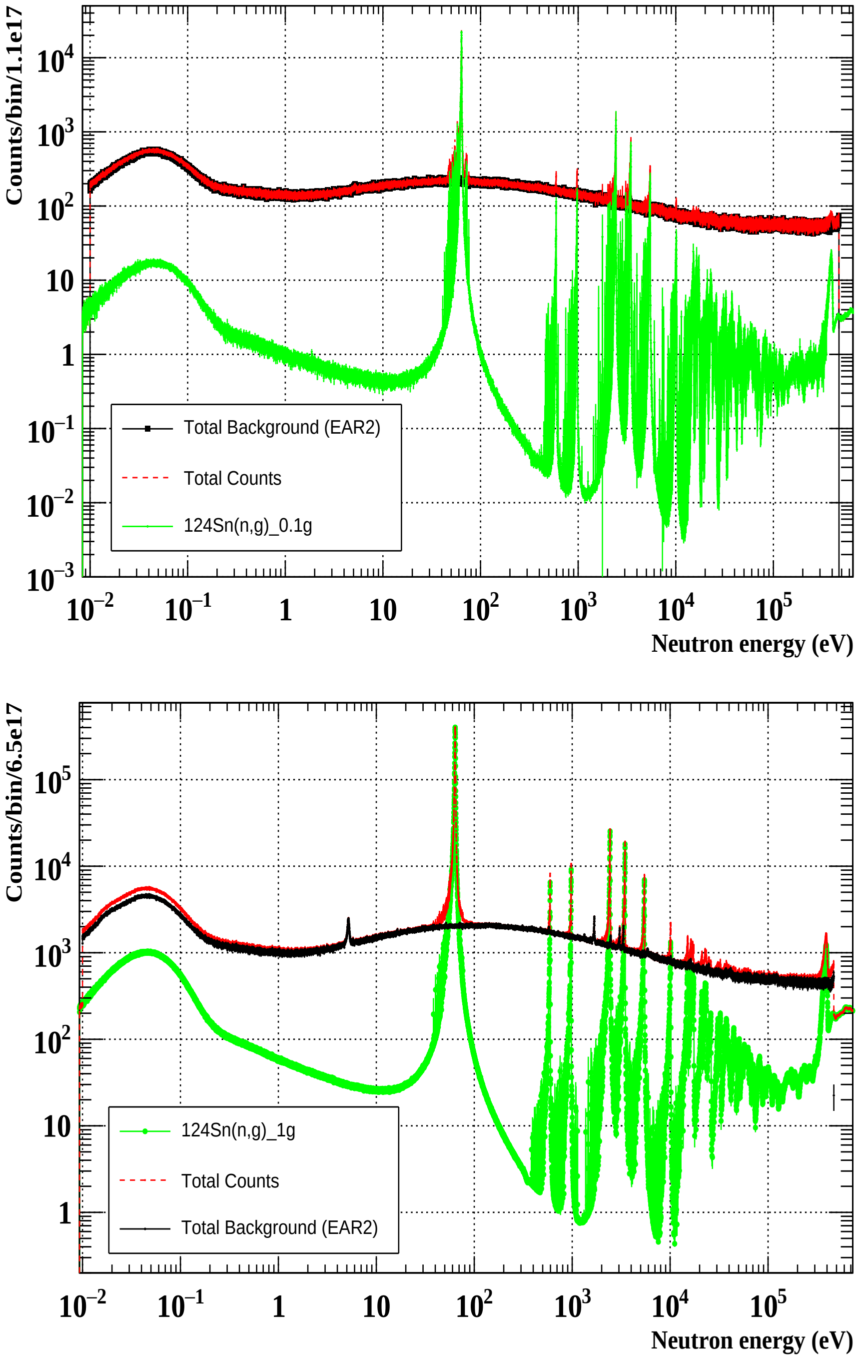

Figure 6: Expected counting rate for the ^{124}Sn(n,γ) experiment, in 2000 bpd and broadened by the resolution function together with the main background components in EAR2, corresponding to 0.1 g (top) and 1 g (bottom) sample mass. The calculations were performed for a ^{124}Sn target enriched to 97.9%.

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Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT

Please describe here below the main parts of your experimental set-up:

Part of the experiment	Design and manufacturing
If relevant, write here the name of the fixed installation you will be using sTED, SIMON present in n_TOF EAR2.	☒ To be used without any modification ☐ To be modified
If relevant, describe here the name of the flexible/transported equipment you will bring to CERN from your Institute [Part 1 of experiment/ equipment]	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing
[Part 2 of experiment/ equipment]	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing
[insert lines if needed]	

HAZARDS GENERATED BY THE EXPERIMENT

Additional hazard from flexible or transported equipment to the CERN site:

Domain	Hazards/Hazardous Activities		Description
Mechanical Safety	Pressure	☐	[pressure] [bar], [volume][l]
	Vacuum	☐	
	Machine tools	☐	
	Mechanical energy (moving parts)	☐	
	Hot/Cold surfaces	☐	
Cryogenic Safety	Cryogenic fluid	☐	[fluid] [m3]
Electrical Safety	Electrical equipment and installations	☐	[voltage] [V], [current] [A]
	High Voltage equipment	☐	[voltage] [V]
Chemical Safety	CMR (carcinogens, mutagens and toxic to reproduction)	☐	[fluid], [quantity]
	Toxic/Irritant	☐	[fluid], [quantity]
	Corrosive	☐	[fluid], [quantity]
	Oxidizing	☐	[fluid], [quantity]
	Flammable/Potentially explosive atmospheres	☐	[fluid], [quantity]
	Dangerous for the environment	☐	[fluid], [quantity]
Non-ionizing radiation Safety	Laser	☐	[laser], [class]
	UV light	☐	

	Magnetic field	☐	[magnetic field] [T]
Workplace	Excessive noise	☐	
	Working outside normal working hours	☐	
	Working at height (climbing platforms, etc.)	☐	
	Outdoor activities	☐	
Fire Safety	Ignition sources	☐	
	Combustible Materials	☐	
	Hot Work (e.g. welding, grinding)	☐	
Other hazards			