Compton imaging for enhanced sensitivity (n,γ) cross section TOF experiments: Status and prospects

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Abstract. Radiative neutron-capture cross sections are of pivotal importance in many fields such as nucleosynthesis studies or innovative reactor technologies. A large number of isotopes have been measured with high accuracy, but there are still a large number of relevant isotopes whose cross sections could not be experimentally determined yet, at least with sufficient accuracy and completeness, owing to limitations in detection techniques, sample production methods or in the facilities themselves.

In the context of the HYMNS (High-sensitivitY Measurements of key stellar Nucleo-Synthesis reactions) project over the last six years we have developed a novel detection technique aimed at background suppression in radiative neutron-capture time-of-flight measurements. This new technique utilizes a complex detection

set-up based on position-sensitive radiation-detectors deployed in a Compton-camera array configuration. The latter enables to implement gamma-ray imaging techniques, which help to disentangle true capture events arising from the sample under study and contaminant background events from the surroundings. A summary on the main developments is given in this contribution together with an update on recent experiments at CERN n_TOF and an outlook on future steps.

Introduction

A large number of neutron-capture reactions of astrophysical interest have been measured utilizing the time-of-flight technique in combination with total-energy detection systems [1, 2]. The former Moxon-Rae detectors [1] were soon replaced with organic scintillators in combination with the pulse-height weighting technique aiming at higher detection sensitivity and better accuracy [3]. Nowadays, organic C_6D_6-scintillators represent the state-of-the-art in this field because they provide the fast time-response required for high-resolution TOF measurements and they show also an intrinsically small sensitivity to backgrounds induced in the detector itself [2, 4, 5].

However, for measurements of astrophysical interest in the keV neutron-energy range the signal-to-background ratio in these detectors can be significantly deteriorated by contaminant neutron-capture reactions in the walls and surroundings of the set-up, as it was found in the Monte Carlo study reported in [6, 7]. This effect is particularly prominent when there is a large neutron-scattering in the sample under study itself. As a consequence, the high background level can hinder the measurement of capture levels of interest, already beyond a few keV of neutron energy [8–10]. This situation was already noticed in the first measurements in the early 60's, where massive lead shielding was used, as it can be seen in Fig. 2 of [1] at Harwell (UK), in Fig. 1 of [2] at ORNL (USA) and in Fig. 3 of [11] at FZK (Germany). See also [12] for more details on these aspects. Indeed, contaminant neutron reactions in the lead shielding itself made the situation worse and, as a consequence, total-energy detection systems evolved later towards lightweight set-ups, with as less surrounding materials as possible, as shown in Fig.2 of [13] at CERN n_TOF and in Fig.3 of [14] at IRMM-Geel. The following section summarizes recent efforts to reduce the aforementioned neutron-scattering γ-ray backgrounds from the surroundings of the set-up by means of a new detector development for TOF experiments.

Compton-imaging aided total-energy detection systems

An alternative approach to reduce the effect of neutron-scattered backgrounds without using a massive lead shielding is gamma-ray imaging based on the Compton-scattering law, also referred to as electronic collimation [15]. This option was proposed and studied in detail on the basis of MC simulations in [16]. A schematic figure of the proposed total-energy detection system with imaging, i-TED, is shown in Fig. 1 (a). The main underlying idea is to gain information on the spatial origin (or incoming direction) of the detected radiation, which can then be
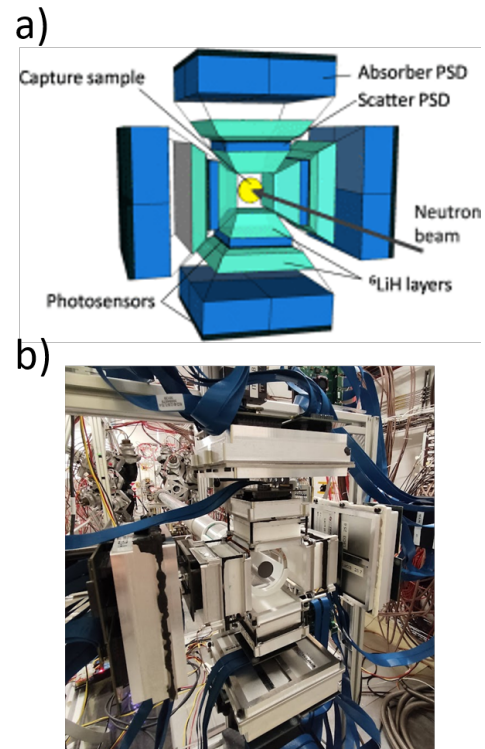

Figure 1. Original design of i-TED as proposed in 2016 [16] (a) and implementation at CERN n_TOF EAR1 for the measurement of ^{79}Se(n, γ) in 2022[17] (b).

utilized to reject gamma-rays that are not arising from the sample under study. Compared to previous attempts based on pin-hole gamma-cameras [18], electronic collimation has the advantage that no massive mechanical collimator is required. However, Compton cameras were never used in the field of neutron-capture time-of-flight experiments before and, therefore, several technical challenges had to be overcome, which are summarized in table 1 and discussed in the following.

Among the main requirements, high energy- and position-resolution are required in Compton imaging to attain a high angular resolution which, in turn, enables the background rejection [16]. Neither energy- nor position-resolution can be satisfactorily achieved with organic scintillators. The only alternative that still provided a sufficiently fast time-response for TOF experiments was inorganic lanthanum-halide crystals. In particular, $LaCl_3$(Ce) was chosen due to the smaller resonance-capture cross-section for Cl than for Br. 3D-position sensitivity was achieved by using pixelated SiPMs optically coupled to the crystals [19]. The energy resolution was optimized by using crystals with reflector around the walls and base

Table 1. Main challenges for the implementation of Compton imaging in neutron-capture TOF experiments and solutions implemented to overcome them. See text and quoted references for details.

Challenge	Solution
High position resolution	pixelated SiPM (1280 pixels) [19], ML-algorithms [19, 20]
High energy resolution	lanthanum-halide crystals with reflector [21]
Low intrinsic neutron sensitivity	$LaCl_3(Ce)$, ^{6}Li-HDPE-absorbers [22, 23]
High (intrinsic) efficiency	Large monolithic crystals, ML-algorithms [20, 24]
High (geometric) efficiency	4 imagers, scalable ASIC-elecronics (1280 channels) [22]
Fast time response	$LaCl_3(Ce)$, SiPMs, TOFPET2 ASIC-readout [19, 25]
Bunch $t_\circ$ time-stamp reference for TOF	Customized electronics with external CERN PS-trigger [22]

which, however challenged the position reconstruction because of the severe pin-cushion effects [21]. Therefore, new algorithms had to be developed for the reconstruction of the γ-ray hit position in the scintillation volume itself. The use of ML-techniques enabled position resolutions of 1-3 mm FWHM in the three space coordinates of the crystal volume with useful field-of-views of 70%-80% of the full crystal surface (50×50 mm^2) [19, 20]. The final system comprised an array of four Compton imagers, with a total of 1280 pixels and readout channels. A picture of the final system installed at CERN n_TOF EAR1 is shown in Fig. 1 (b). Because of the large number of channels, the acquisition-system of n_TOF, based on digitizer modules, could not be used. i-TED was therefore instrumented with latest generation of ASIC-based TOFPET2 front-end readout electronics, which were originally developed for high-resolution medical PET imaging [19, 25]. In terms of Compton imaging, the performance of the resulting i-TED apparatus was very satisfactory but, for its implementation in TOF experiments further developments were required for time-stamping the $t_\circ$-value of each proton bunch and also for reducing the intrinsic neutron sensitivity. The latter was accomplished by means of customized ^{6}Li-enriched high-density polyethylene absorbers (^{6}Li-HDPE). More details can be found in [22]. Technical and proof-of-concept validation measurements were carried out with one i-TED module in 2018 at CERN n_TOF EAR1 using the ^{56}Fe(n, γ) reaction as reference, which was also measured with C_6D_6 detectors as reference. As reported in [22], the results allowed to validate the technical developments implemented in i-TED and the proposed methodology for active background rejection via γ-ray imaging. The main limitation found was related to the analytical algorithm for background rejection which, however, could be overcome also with more advanced methods based on ML-techniques, as reported in [22].

Status and outlook

In 2022 the first physics measurement with the full i-TED array was carried out at CERN n_TOF EAR1 (see Fig. 1). The system was used to measure, for the first time, ^{79}Se(n, γ) with high resolution over a broad neutron-energy range [17]. An eutectic lead-selenide (^{208}Pb^{78,79}Se) alloy sample was produced for this experiment, as described in [17, 26]. The TOF-measurement of this sample was particularly challenging because of two reasons.
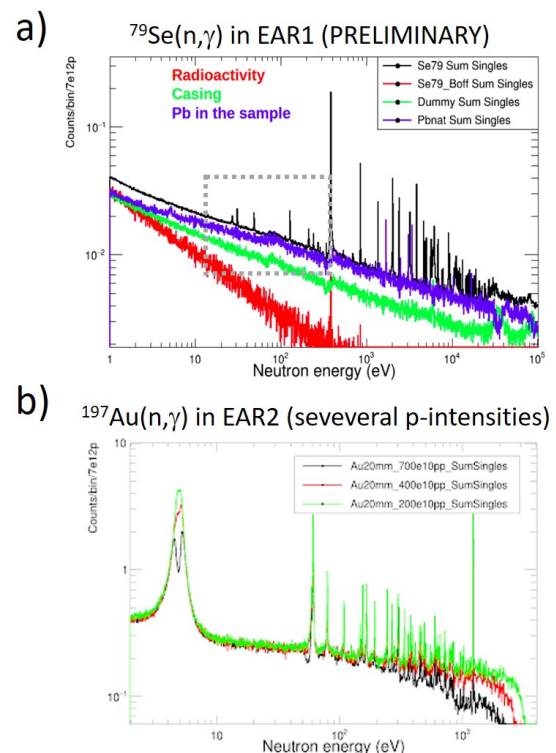

Figure 2. Count-rate vs. neutron-energy spectrum of ^{79}Se(n, γ) measured with i-TED in EAR1 (a) [27]. Spectrum of ^{197}Au(n, γ) measured with one i-TED prototype in EAR2 for proton-beam intensities between 2×10^{12} and 7×10^{12} protons/bunch (b).

Firstly, the large ^{208}Pb content in the sample induced a correspondingly large neutron-scattering γ-ray background in the surroundings. Secondly, a large γ-ray activity was also present in the sample due to activated impurities of ^{75}Se- and ^{60}Co, leading to activities of 5 MBq and 1.4 MBq, respectively. Fig. 2 shows first preliminary data measured with i-TED for the ^{79}Se(n, γ) at EAR1 with i-TED, still without any use of γ-imaging [27]. The sample-radioactivity (red line) has been significantly suppressed by means of a relatively high-energy threshold in the data. The main remaining background contribution is due to neutron-scattering in the lead- and sample-encapsulation, as shown by the blue and green lines, respectively. Next steps in the data analysis will therefore focus on implementing the ML-based imaging-algorithm developed in [22] to reduce this remaining background component. The

outcome will be crucial for understanding to which extent the imaging-technique can help in this type of experiments. At present, the main limitation of i-TED is related to its count-rate capability, which is constrained to about 500 kEvents/s per detector. This feature presently prevents its application with very high-peak neutron-fluxes, as it is the case of EAR2. This statement is illustrated in Fig. 2 (b), which shows the $^{197}Au(n,\gamma)$ count-rate per bunch as a function of the neutron energy measured with one i-TED module placed at 36 cm from the neutron beam line using a 0.1 mm thick sample. Several runs with different proton-beam intensities were made and it was found that a reduction by a factor of three was required in the nominal intensity to avoid severe dead-time effects in the i-TED data. In the future, this limitation could be somewhat mitigated with the use of pixelated inorganic crystals, although they are more expensive and less efficient. However, the main factor limiting the i-TED count-rate capability is the ASIC-based readout electronics, which show an intrinsic count-rate constrain of <600 kHz per readout channel. Considering the swift advance of microelectronics, new generations of readout chips based on 65 nm CMOS ASIC technology [28, 29] may help in the near future to overcome this performance limitation. Finally, an aspect which was found in [22] to have a remarkable impact in the attainable signal-to-background ratio of i-TED is the coincidence-time resolution or CTR. Efforts are being made with i-TED to bring this parameter down to a few 100 ps, which would allow one to maximize its performance in terms of background rejection. In this respect, future developments achieved within the 10 ps CTR Challenge task-force, proposed by the Crystal Clear Collaboration [30], could help also to push i-TED performances one step further.

Acknowledgements

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