Review and new concepts for neutron-capture measurements of astrophysical interest

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Abstract. The idea of slow-neutron capture nucleosynthesis formulated in 1957 triggered a tremendous experimental effort in different laboratories worldwide to measure the relevant nuclear physics input quantities, namely (n, γ) cross sections over the stellar temperature range (from few eV up to several hundred keV) for most of the isotopes involved from Fe up to Bi. A brief historical review focused on total energy detectors will be presented to illustrate how advances in instrumentation have led to the assessment of new aspects of s-process nucleosynthesis and to the progressive refinement of stellar models. A summary will be presented on current efforts to develop new detection concepts, such as the Total-Energy Detector with γ-ray imaging capability (i-TED). The latter is based on the simultaneous combination of Compton imaging with neutron time-of-flight (TOF) techniques, in order to achieve a superior level of sensitivity and selectivity in the measurement of stellar neutron capture rates.

1. Introduction

Observation, theory and experiment: this seems to be a persistent repetitive pattern of research in nuclear astrophysics since its origin. In 1952 Merrill observed for the first time absorption lines of the radioactive element Technecium in the stellar atmosphere of S-type stars [1], hence revealing recent or ongoing nucleosynthesis activity in the stars. Five years later, the seminal theoretical works of Burbidge et al. [2] and Cameron [3] (hereafter B2FHC) were published, thereby presenting a theory for the nucleosynthesis of heavy elements that essentially remains valid today [4]. Shortly after B2FHC a frenetic experimental activity followed in the nuclear physics laboratories. Firstly intended to demonstrate and secondly to probe and constrain the predictions of the models and the observations by the astronomers. This contribution focuses on some of those experimental efforts. Sec. 2 describes a few examples selected to illustrate how advances in instrumentation and new detection concepts have led to the progressive refinement of theoretical models and to a better understanding of the physical conditions in the corresponding stellar environments. Ongoing research to further enhance the sensitivity of TOF measurements and access more challenging measurements is presented in Sec. 3.

2. Instrumental developments and discoveries

There exist different techniques to determine neutron capture cross sections, which are the key nuclear physics input quantities for s-process model calculations. One of them is the activation method, which shows an extremely high sensitivity and selectivity (for recent examples see e.g. [5, 6]). However, the activation method is mostly applicable to nuclei that, upon capture, become radioactive and have a convenient half-life and decay pattern. In some cases, the use of Accelerator Mass Spectrometry has allowed to overcome this limitation [7, 8].

In this contribution we focus on the use of pulsed neutron beams in combination with the time-of-flight (TOF) technique to measure neutron capture rates of astrophysical interest. The TOF method is applicable to any nucleus, provided that a sufficiently large sample of material becomes available. Also, it offers the possibility to cover the full stellar energy range in a single measurement. By the time when the nucleosynthesis theory of B2FHC was published neutron capture TOF measurements were mostly based on the use of large scintillation tanks (1000 liters) [9] with a large γ-ray detection efficiency, as the one shown in Fig.1-a). However, the bulky setup showed a high sensitivity to neutron induced backgrounds, which strongly limited the detection sensitivity for the capture channel of interest. Indeed, measurements were restricted to only very large samples ($\sim$1 mole). Isotopically enriched samples of that size were very expensive and difficult to produce. Thus, the technique was mainly used to measure samples of elements with natural isotopic abundances [9]. As suggested by B2FHC, in order to quantitatively test the s-process theory it was necessary to know the abundances and the cross-sections of the individual involved isotopes, as the relative elemental abundances may be altered by uncertain physical or chemical fractionation processes.

The first breakthrough in the field came with the development and application of a radically new concept: radiation detectors that featured a detection efficiency proportional to the γ-ray energy, so-called Moxon-Rae detectors [16, 17], shown in Fig. 1-b). These detectors were based on three layers of graphite, bismuth and a plastic scintillator. The latter was read-out with a photomultiplier tube. The two front layers of C and Bi acted as electron converters for the incident radiation, thereby yielding a γ-ray detection efficiency roughly proportional to its initial energy. This proportionality condition between efficiency and γ-ray energy was of pivotal importance in order to avoid systematic errors in the cross section related to the multiplicity m of the capture cascade or to the particular resonance decay path. In short, by ensuring a sufficiently low detection efficiency $i)$ $\varepsilon_\gamma \ll 1$, and the proportionality condition $ii)$ $\varepsilon_\gamma = kE_{\gamma,j}$, the probability to detect a capture event ε_c (eq. 1) becomes proportional to the capture energy, αE_c, which is a constant well-defined value for each capture event at a given measured neutron energy ($E_c = S_n + E_n$).

$$\varepsilon_c = 1 - \prod_{j=1}^{m}(1-\varepsilon_{\gamma,j}) \overset{i)}{\simeq} \sum_{j=1}^{m} \varepsilon_{\gamma,j} \overset{ii)}{\simeq} \alpha \sum_{j=1}^{m} \varepsilon_{\gamma,j} = \alpha E_c. \tag{1}$$

Moxon-Rae detectors, also called Total Energy Detectors (TEDs), attained about one order of magnitude improvement in detection sensitivity. Thereby enabling measurements on samples ten times smaller ($\sim$0.1 mole) than those needed with the large scintillation tanks. Such sample quantities were better suited for isotopic enrichment and thus, the first measurement on isotopically enriched tin isotopes could be carried out using samples of “only” 30-35 g [11]. Fig. 2 schematically shows the evolution in time of the capture detection sensitivity using the TOF technique with low-volume detectors. Detection sensitivity is arbitrarily defined here as the inverse of the sample mass (mg) times the cross section (mb) at neutron energies of kT $\sim$30 keV. Thus, measurements of smaller sample quantities or lower cross sections indicate a higher capability of the detection apparatus to deliver results. Note that the examples shown in this contribution are not necessarily representative of the state-of-the-art at that time. Instead, they have been rather chosen in order to discuss the evolution of the methods in the field. It is also

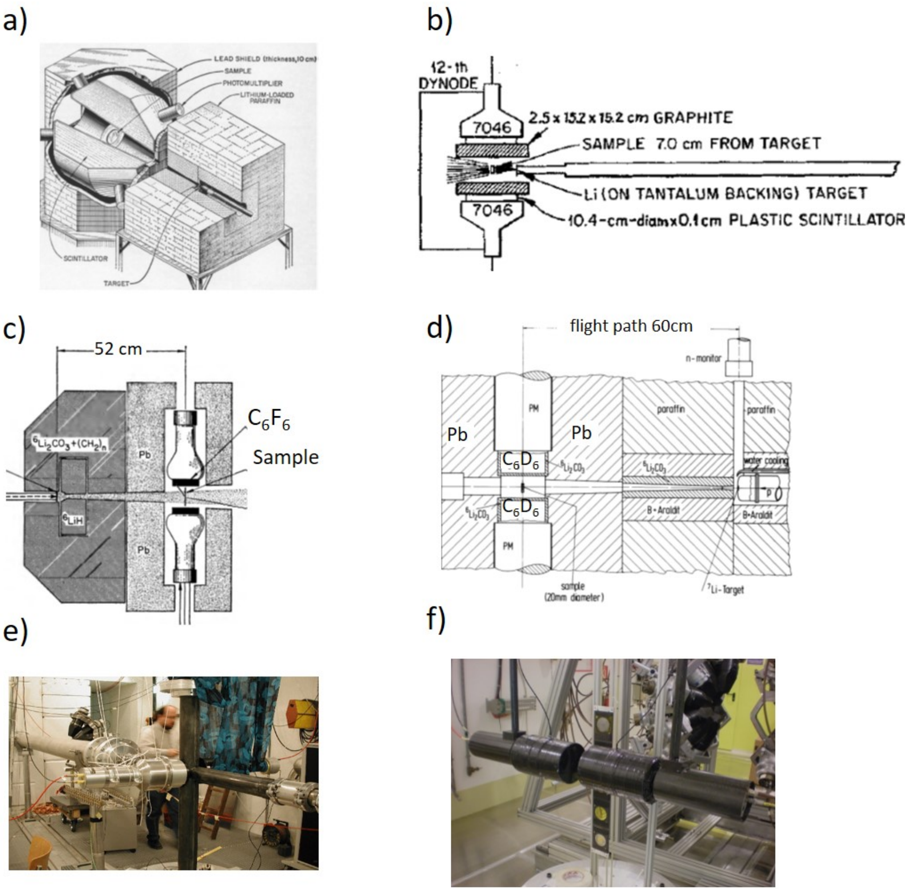

Figure 1. a) Scintillation tanks used for TOF experiments by the time when B2FHC was published [9]. b) Moxon-Rae detector developed for the first measurement of samples enriched on Sn and Sm isotopes (adapted from [10]), applied to the first experimental confirmation of the s-process theory [11]. c) C_6F_6 setup used with the PHWT to measure ^{99}Tc(n,γ) and to determine the AGB lifetime in the 3^{rd} dredge-up phase [12]. d) Neutron sensitivity improved set-up with C_6D_6 detectors [13]. e) Unshielded low-background C_6D_6 detectors used for the first ^{151}Sm(n,γ) TOF measurement [14]. f) Ultra-low neutron-sensitivity (LNS) C_6D_6 set-up similar as the one used for the ^{63}Ni(n,γ) TOF experiment to constrain pre-supernova Cu-content in massive stars [15].

worth to emphasize, that enhancements in detection sensitivity have been naturally due to concomitant improvements in accelerators, neutron-beams and sample purification and preparation techniques. However, these aspects will not be discussed further in this contribution.

The measurement of the tin nuclei [11] was shortly followed by another measurement on isotopically enriched Sm samples [10], both of them using Moxon-Rae TEDs. At that time, these results represented a first experimental validation of the s-process theory, because they demonstrated the inverse proportionality between the nuclear neutron capture cross section and the relative (s-process) isotopic abundances, as predicted by

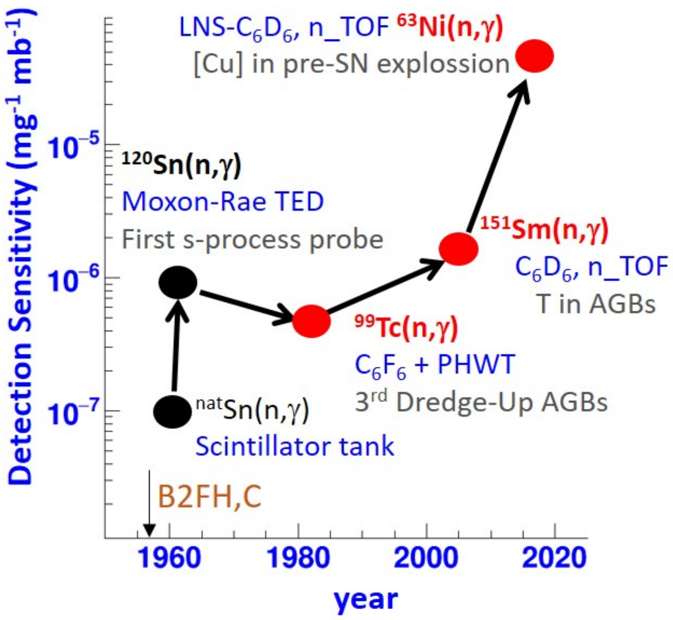

Figure 2. This figure shows some selected examples to illustrate how advances in the instrumentation helped first to validate, and afterwards assess and constrain many fascinating aspects of the stellar evolution. Sample radioactivity is not included in the sensitivity definition, but measurements with radioactive samples are shown with bold red circles. See text for details.

the s-process theory of B2FHC [11]. Quoting [10], *the results thus strongly confirm the s-process nucleosynthesis prediction.*

Despite their remarkable performance, the efficiency of Moxon-Rae detectors was so small, that very short flight-paths of only 5-10 cm could be utilized with the neutron beam intensities available at that time [17]. However, the efficiency-energy proportionality embedded within the Moxon-Rae detectors was a very powerful concept, that would be highly exploited in the following decades.

The next notable development was to realize, that the proportionality condition could be also achieved by using almost any type of fast radiation detector of low neutron sensitivity in combination with a pulse-height analyzer and a recording system. By storing the amplitude of the pulses during the measurement a convenient "weight" could be applied afterwards in order to recover the proportionality condition, without the need of (hardware) photo-electron converters. This was the origin of the so-called Pulse-Height Weighting Technique (PHWT) [18]. Large volumes (∼0.6 L) of non-hydrogenous scintillation liquid (C_6F_6) were readout with photomultiplier tubes [18], as shown in Fig. 1-c). The higher efficiency of this new technique allowed one to use longer flight-paths of about 50 cm, and also to tackle the measurement of more challenging and smaller samples, such as radioactive samples of ^{99}Tc [12] with a mass of only 2.3 g

($\sigma^{99Tc}_{30\ keV}$ = 933 mb see Fig. 2). The decrease of sensitivity in Fig. 2 for this example is only apparent, due to the simplified definition used here for detection sensitivity, which does not account for the experimental difficulty associated with the radioactivity of the sample itself. The cross section measurement of ^{99}Tc, in combination with Tc, Nb and Mo abundances observed in the stellar atmospheres of two S-type stars (R CMi and CY Cyg) allowed for the first determination of the lifetime spent by these stars in the 3^{rd} dredge-up phase of evolution [19, 12].

Further efforts to improve detection sensitivity focused on the reduction of the intrinsic neutron sensitivity in the detectors themselves, thereby replacing the fluorine in the liquid scintillator (C_6F_6) by deuterium (C_6D_6) [13, 20], as shown in Fig. 1-d). Later, it was found that the massive lead shielding around the scintillation detectors (Fig. 1-c)-d)) was, in most cases, amplifying the background rather than suppressing it. New lightweight and unshielded C_6D_6-based detection systems, as the one shown in Fig. 1-e), enabled many new astrophysically relevant results. One example is the first TOF measurement of the s-process branching nucleus ^{151}Sm(n,γ) using only ∼200 mg of sample mass [14] ($\sigma^{151Sm}_{30\ keV}$ = 3031 mb, see Fig. 2). Under the He-burning conditions of AGB stars the β-decay rate of ^{151}Sm ($t_{1/2}$=93 y) is significantly enhanced due to the thermal population of low-lying excited states. This effect can be used as a potential s-process thermometer. The measured cross section for ^{151}Sm(n,γ), in combination with stellar models and the solar system abundance of ^{152}Gd, allowed to constrain the temperature range of the He-shell flashes to 2.5-2.8$\times10^8$ K.

Presently, the state-of-the-art in TEDs is represented by C-fibre optimized detectors [21] and surrounding structural elements [22] (Fig. 1-f). The aim is to minimize the overall intrinsic neutron sensitivity of the detection set-up. Also, large detection volumes of ∼1 L C_6D_6 are possible without necessarily compromising the applicability and accuracy of the PHWT, despite of the ineluctable γ-ray summing in each detector. A methodology based on MC-simulations and the statistical nuclear model allows to reliably account for the enhanced γ-ray summing effect. This permitts an overall accuracy of better than 2% [23]. The capture detection sensitivity achieved with these low neutron sensitivity (LNS) C_6D_6 can be exemplified by the (n,γ) measurement of the unstable ^{63}Ni($t_{1/2}$=101.2(15) y) [15]. The latter, in conjunction with stellar models for 25 $M_\odot$ [24], could be used to constrain the Cu-composition of the s-process inventory in massive stars at their last evolutionary stage before exploding as supernovae [15].

3. New concepts: γ-ray vision for background discrimination

One of the limiting factors in current TOF experiments with state-of-the-art detectors is due to scattered neutrons in the capture-sample, and subsequently thermalized and captured in the surrounding walls of the experimental area. After capture in the walls, these stray neutrons emit radiation which eventually reaches the C_6D_6 detectors, thus enhancing the background level. This type of background is described in detail in Ref. [25] and actually limits the attainable peak-to-background ratio or detection sensitivity in many experiments already beyond few keV of neutron energy. One example is the measurement of ^{93}Zr(n,γ) [26, 27], where capture levels beyond $E_n \sim 8$ keV were already difficult to identify. The spectrum labeled as "Setup" in Fig. 1 of Ref. [26] shows the impact of this type of background, and reflects the limitation of existing systems to measure the cross section in the full energy range of stellar relevance.

One possibility to reduce this type of background would be to "focus" the C_6D_6 towards the sample and shield them from the surroundings, so that they can only "see" true capture γ-rays coming from the sample. This is not a new idea and, indeed, the first experimental setups using TEDs with the PHWT were already utilizing some sort of mechanical collimation, as it is demonstrated in Fig. 1-c) and d). However, as discussed in the previous section, the massive lead shielding is only effective for the suppression of γ-ray backgrounds, but not for the sample-scattered neutrons which produce further radiation in lead. This was also confirmed recently by using a γ-camera at CERN n_TOF, which consisted of a position-sensitive radiation detector coupled to a heavy pin-hole collimator made from lead [28].

Another possibility to "focus" the detectors towards the sample, without the need of mechanical collimation consists of using electronic collimation. Electronic collimation is based on the use of the Compton scattering law in order to infer a cone of possible incident directions [29]. For this to be implemented one needs a detection system, normally divided in two volumes, that can provide information on the position and the energy of the γ-ray interactions in the detector (see Fig. 3). For neutron capture TOF experiments the detection system must show a fast response and low sensitivity to neutrons. These are the main goals for a new detection system called i-TED (Total Energy Detector with γ-ray imaging capability) [30] that is being developed in the framework of the HYMNS project [31]. The proposed Compton imager is quite different from any previous detector used for (n,γ) measurements, a fact which exemplifies the versatility of the PHWT discussed in Sec. 2 where only the two conditions of low-efficiency and efficiency-energy proportionality are required to reliably measure a cross section. Low efficiency is guaranteed in i-TED because the time-coincidence between scatter- and absorber-planes already reduces the overall efficiency by a factor of $\sim$5 [30]. The applicability of the PHWT to achieve the proportionality condition has been also demonstrated on the basis of MC simulations [30].

In order to attain sufficient angular resolution ($< 15°$) for background rejection, high-resolution inorganic $LaCl_3$(Ce) scintillators are being implemented in i-TED [32]. High-resolution HPGe [33] or other semiconductor sensors are excluded due to their slow time-response, their higher neutron capture cross section and their lower intrinsic efficiency. Energy- and position-information from the $LaCl_3$(Ce) is gained by optically coupling them to pixelated silicon photomultipliers (SiPMs). In particular, we use $50 \times 50 \times 10$ mm^3 monolithic crystals for the i-TED scatter plane and 25 mm thick crystals for the absorber detection plane. Compared to other similar Compton cameras developed for medical applications [34, 35], i-TED uses significantly larger crystals which represents a challenge in terms of readout channels and overall performance. However,

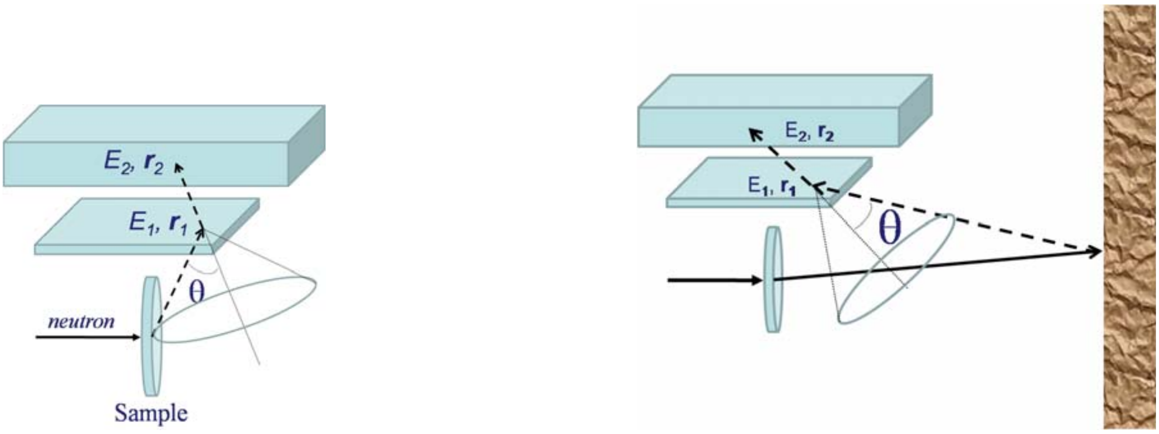

Figure 3. Schematic representation of the i-TED concept to discriminate true capture events (left) from contaminant radiation originating in the surroundings (right).

this is required for the intended TOF capture experiments in order to have a detection efficiency which is sufficient to perform a neutron capture experiment in a reasonable time frame. The energy resolution that has been achieved with this type of position-sensitive detectors (PSDs) using SiPMs is of about 4-5% FWHM at 662 keV [36]. The spatial resolution attainable with large monolithic crystals is also a technically difficult aspect. Thus far, we have investigated different approaches to reconstruct the 3D-coordinates of the γ-ray hit location in the $LaCl_3$(Ce) volume. Presently, the most suited method seems to be a least-squares fit on an event-by-event basis to the SiPM response using an analytic model [37], which accounts for the spatial propagation and reflection of the scintillation light produced by a point-like source in the crystal volume. By means of an accurate spatial calibration of our PSDs using a collimated γ-ray source in conjunction with an XY-positioning table, we have been able to obtain position resolutions of 1-2 mm FWHM at 511 keV for crystal thicknesses between 10 mm and 30 mm [38]. As shown in Ref. [30], the full i-TED system will comprise four large solid-angle Compton cameras arranged in a close geometry around the capture sample. In order to instrument the full array, about 1280 readout channels will be required. The SiPM signals are acquired using ASIC-based frontend readout electronics that have been developed by PETsys Electronics [39] for medical applications. The suitability of this readout approach for i-TED resides on its scalability and on the fact that it provides both energy (QDC) and time information with good resolution [36, 38]. In order to adapt the medical readout system to the neutron-TOF requirements, some customization of the electronics was needed. In particular an external trigger-signal from the CERN-PS was fed and time-stamped in one of the readout-channels in order to enable the possibility to build a TOF spectrum. A first i-TED prototype has been already tested at the CERN n_TOF EAR1 facility, and a count-rate versus neutron-energy spectrum is shown in Fig. 4 for ^{197}Au(n,γ). The latter represents a technical validation of the system in terms of TOF performance, as no degradation of the neutron-energy resolution is observed

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with the use of this new instrumentation.

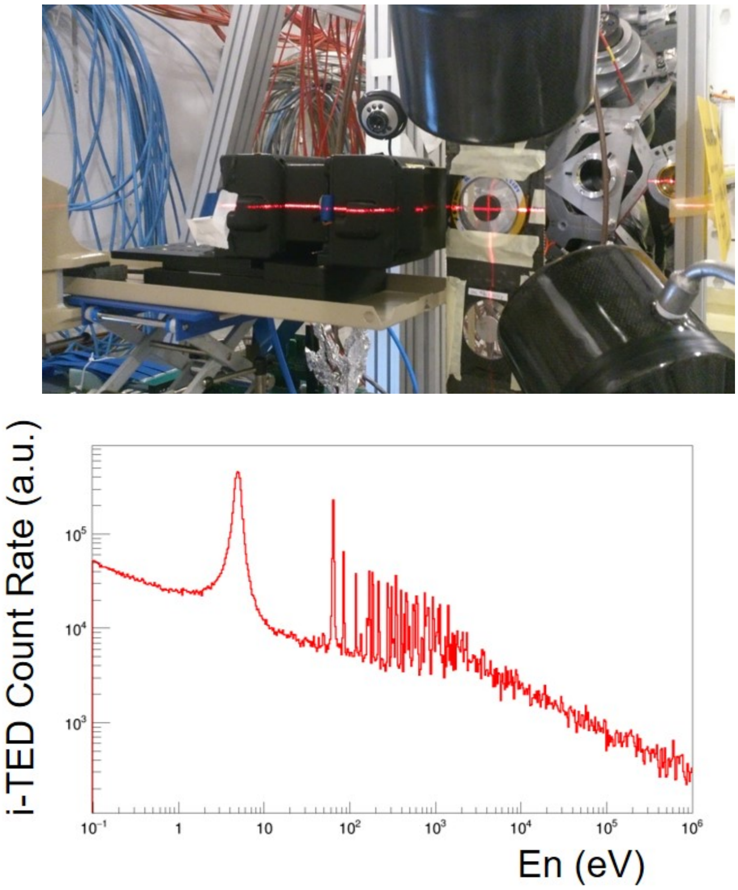

Figure 4. (Top) Picture of the experimental set-up at CERN n_TOF with an i-TED prototype being aligned at ~5 cm from the capture sample. (Bottom) Un-weighted count-rate histogram as a function of the neutron energy measured with i-TED for the gold capture sample.

Once the system was adapted for TOF experiments and technically validated, further capture tests were carried out at CERN n_TOF using an i-TED prototype more similar to the one described in Ref. [32]. The aim of these latter tests was to explore the applicability of the γ-ray imaging for background rejection. The data are being analyzed and results will be reported in future publications.

4. Summary and outlook

Several neutron capture experiments and the related instrumentation have been discussed in order to illustrate how progress in capture detection systems has helped, since the origin of the modern nucleosynthesis theory, to validate and constrain different astrophysical aspects. The emphasis here has been put on the working principle of the so-called Total Energy Detectors (TEDs) and their interesting evolution along the nearly

70 years of existence. Finally, a new type of TED based on Compton imaging has been presented as a possibility to further reduce neutron-induced gamma-ray backgrounds and thus enhance the signal-to-background ratio in this type of experiments.

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