

EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Letter of Intent to the ISOLDE and Neutron Time-of-Flight Committee

Development of new solid-state total-energy detectors for neutron-capture measurements at CERN n_TOF

April 18, 2023

O. Aberle¹, V. Alcaÿne², M. Bacak¹, J. Balibrea-Correa³, N. Colonna⁴, D. Cano-Ott²,
A. Casanovas⁵, C. Domingo-Pardo³, O. Fjeld¹, F. Gunsing⁶, J. Lerendegui-Marco³,
C. Lederer-Woods⁷, C. Massimi^{8,9}, E. Mendoza², A. Mengoni^{4,10}, A. Manna^{9,10},
A. Musumarra^{11,12}, N. Patronis¹, M.G. Pellegriti¹²

¹*European Organization for Nuclear Research (CERN), Switzerland*

²*Centro de Investigaciones Energéticas Medioambientales y Tecnológicas (CIEMAT), Spain*

³*Instituto de Física Corpuscular, CSIC - Universidad de Valencia, Spain*

⁴*Istituto Nazionale di Fisica Nucleare, Sezione di Bari, Italy*

⁵*Universitat Politècnica de Catalunya, Spain*

⁶*CEA Irfu, Université Paris-Saclay, F-91191 Gif-sur-Yvette, France*

⁷*School of Physics and Astronomy, University of Edinburgh, United Kingdom*

⁸*Istituto Nazionale di Fisica Nucleare, Sezione di Pavia, Italy*

⁹*Dipartimento di Fisica e Astronomia, Università di Bologna, Italy*

¹⁰*Istituto Nazionale di Fisica Nucleare, Sezione di Bologna, Italy*

¹¹*Dipartimento di Fisica e Astronomia, Università di Catania, Italy*

¹²*Istituto Nazionale di Fisica Nucleare, Sezione di Catania, Italy*

Spokespersons:

Javier Balibrea-Correa javier.balibrea@ific.uv.es

Agatino Musumarra musumarra@lns.infn.it

Technical coordinator: O. Aberle Oliver.Aberle@cern.ch



Abstract: Liquid C_6D_6 has been the detector of choice for performing most of the neutron-capture cross section measurements at CERN n_TOF over the last 20 years. In this LoI we propose a series of measurements at CERN n_TOF EAR2 in order to characterize the performance of new solid-state scintillators for time-of-flight neutron-capture experiments. In particular, we plan to benchmark the performance of regular and deuterated stilbene (stilbene- d_{12}) scintillation crystals with respect to state-of-the-art C_6D_6 liquid scintillators and segmented Total Energy Detectors (sTED). If a similar performance can be confirmed, we plan to design and develop a new generation of total-energy detectors with enhanced detection sensitivity and improved safety and maintenance conditions.

Requested protons: 6.0×10^{17} protons on target
Experimental Area: EAR2

1 Introduction

Liquid scintillation C_6D_6 detectors have been used since the very first measurements at CERN n_TOF for the determination of neutron-capture cross sections by means of the total energy detection and pulse-height weighting technique (PHWT) [1, 2]. In the first phase of measurements at CERN n_TOF, a big effort was made in order to optimize the C_6D_6 detectors in terms of neutron sensitivity and neutron-induced backgrounds in the detector material itself [3]. This work turned out to be of importance in order to address some remarkable inconsistencies in previous cross-section data, which suffered from significantly large uncertainties due to the aforementioned neutron-sensitivity effect. One of such example was the 2.3 keV resonance in $^{209}Bi(n,\gamma)$, which has a three orders of magnitude larger neutron scattering width Γ_n than neutron capture width Γ_γ (see e.g. Fig.3 in [4], and [5]). Later efforts at n_TOF focused on further improving this type of detector, not only in terms of neutron sensitivity, but also in terms of safety [6]. Improved encapsulations allowed to avoid potential leaks of scintillation liquid and other safety issues, thus increasing their safety and stability during the experiments. However, it is worth recalling that the sensitive detector material used in this type of detector over the last 20 years, C_6D_6 , still corresponds to Cat.2 in terms of flammable liquid, skin corrosion/irritation and serious eye-damage hazard. Further, C_6D_6 belongs to Cat.1 hazard in terms of germ cell mutagenicity (1B), carcinogenicity (1A) and toxicity for the hematopoietic system and aspiration. With this situation in mind, in this LoI we aim at exploring the performance and suitability of the trans-stilbene organic scintillators (stilbene), and the deuterated version (stilbene- d_{12}) [7], as a potential replacement of conventional C_6D_6 liquid scintillator. The chemical composition of stilbene/stilbene- d_{12} is $C_{14}H(D)_{12}$, 16% larger C to H(D) ratio compared to conventional C_6D_6 . Thus, it is expected that, in terms of neutron-sensitivity, these detectors will perform similar, if not better in the case of stilbene- d_{12} , than conventional C_6D_6 because of the reduction of structural material compared to the active volume. The solid organic crystal has a density of about 30% larger than the commonly used C_6D_6 , which also represents an advantage in terms of intrinsic detection efficiency and enables compact configurations with larger solid angle coverage. Finally, being stilbene a solid-state material without any chemical hazard makes it a very attractive material in terms of detector of choice for future experiments at CERN n_TOF. Also, solid scintillators allow to avoid the quartz-crystal window commonly required with liquid C_6D_6 , which is one of the main contributors to the neutron sensitivity of the detector itself [3].

Finally, another aspect to explore in this proposal is the possible replacement of the bulky Photomultiplier tube by a lightweight and compact array of silicon photomultiplier (SiPM) sensors. This feature would reduce further the intrinsic neutron sensitivity while offering the possibility of even more compact setups and the use of low-voltage (~ 30 V) power supplies [8].

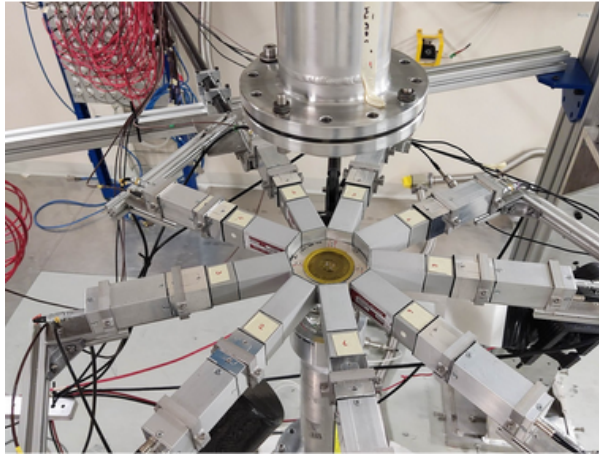


Figure 1: Compact configuration of 9 small-volume sTED C_6D_6 liquid scintillators set-up in the EAR2 station.

2 State of the art

Several different types of C_6D_6 detectors have been developed and utilized since two decades at CERN n_TOF, most of them designed and assembled within the collaboration. Initially, rather large cell volumes of about 1 l of C_6D_6 [3] were used, which were significantly larger than commercial Bicorn C_6D_6 detectors (500 mL). This approach was intended to maximize detection efficiency and thus to optimize the total measuring time for every experiment. Subsequent improvements focused on keeping large detection volumes and high efficiency, while improving encapsulation stability and overall detector safety [6].

With the development of the EAR2 station and the correspondingly large neutron luminosity, detector design had to be adapted to the new demanding conditions of very high instantaneous counting rates. This situation led to the development of small volume C_6D_6 cells, so-called sTED detectors [9]. With small-volume detectors it became clear that detection sensitivity can be significantly improved with respect to previous set-ups, thanks to much shorter sample-detector distances and more compact configurations while keeping at bay systematic errors such as dead-time corrections. Fig. 1 shows a picture of the new compact configuration of sTED C_6D_6 liquid scintillation detectors.

Thanks to the latter developments, in 2022 we were able to measure for the first time neutron capture cross sections of very challenging radioactive samples, available in very small quantities, such as $^{79}Se(n,\gamma)$ [10] and $^{94}Nb(n,\gamma)$ [11]. Until the current date, except for an exceptional measurement made in 2017, it was not possible [12].

The recent development of solid-state organic trans-stilbene scintillators has opened a new line of research for a replacement of the current liquid organic scintillation detectors for multiple applications. In particular, stilbene- d_{12} crystals have intrinsically low neutron sensitivity [7], opening up the window to a new generation of total-energy detectors with an even larger detection sensitivity and remarkably improved safety and stability characteristics.

A performance comparison between PMT and SiPM sensors is something to be carefully

investigated. If, on one hand, SiPM are expected to improve the neutron sensitivity reducing the overall material around the scintillator, array of SiPM need a more complex electronic front-end in order to maintain comparable time response, linearity and dynamic range. SiPM intrinsic recovery time is also quite large (50-100 ns) making n-gamma Pulse Shape Discrimination (PSD) not trivial and in principle worse than standard PM. Anyhow, it is also worthwhile to stress that SiPM are insensitive to magnetic fields and operate at low voltages (30-60 V). In order to shed light on this crucial point, a preliminary characterization was performed at NCSR (Athens) coupling a LLNL Stilbene-d₁₂ crystal to HAMAMATSU and AdvanSiD SiPM and optimizing the PSD analysis by using three different signal processing techniques which analysis is undergoing.

Experimental setups based on TEDs have always aimed at lowering the intrinsic neutron sensitivity [3, 6]. This experimental effect, originating from neutrons scattered in the sample and captured directly in the detector, leads to an overestimation of the resonance area and is especially sizable for resonances with large neutron scattering to capture ratio ($\Gamma_n \gg \Gamma_\gamma$). The aim of this campaign will be to experimentally characterize the neutron sensitivity of the different detection devices using as a reference the state-of-the-art Legnaro-C₆D₆ [6].

Consequently, in this LoI we propose a series of measurements in order to characterize and validate the performance of stilbene crystals and neutron sensitivity for all detection devices in the context of (n,γ) experiments while exploring the use of SiPM and PM at EAR2 n_TOF.

3 Working plan

A series of detectors are available for this proposal, which include three sTED detectors [11, 10], one state-of-the-art C₆D₆ detector [6], two stilbene-d₁₂ crystals with a size of 25×25×13 mm³ developed by the group of N. Zaitseva at Lawrence-Livermore National Laboratory [7] (LLNL), and four units of cylindrical stilbene scintillators, 25.4 mm in diameter, 25.4 mm long (1 inch x 1 inch) produced by Inrad Optics US (Scintinel) according to the LLNL trans-stilbene growing process. It is foreseen that one of the stilbene detectors, regular or deuterated, will be coupled to a SiPM during these test measurements. The rest of detectors will use conventional PMTs.

The main aspects to be investigated in these tests are the following:

- *Gamma-flash response:* The quick recovery of the detector after the strong gamma-flash determines the highest neutron-energy that can be measured with each type of detector. This aspect will be investigated and benchmarked against state-of-the-art small-volume C₆D₆ detectors (Configuration 1 reported in table 1).
- *Gain stability along the time-of-flight:* Check of the gain stability as a function of the counting rate conditions, this is important for a reliable assessment of the cross section over the entire neutron-energy interval (Configuration 1 and 6).
- *Neutron sensitivity:* The neutron-induced (prompt) background in the detectors will be investigated by means of a measurement involving ²⁰⁹Bi sample and a ⁵⁶Fe

sample. These two isotopes have resonances at 2.3 keV and 25 keV, respectively, with G_n/G_γ ratios in excess of 1000. They are therefore ideal cases to characterize the neutron sensitivity of the detectors, and benchmark it against the C-fibre based C_6D_6 scintillators [6]. The capture cross-section of these two resonances is relatively small and, therefore, at least 3 days of measuring time will be required for this study (Configuration 3 and 4).

- *Counting rate capability:* The time-response of the detector is an important aspect to study, particularly for measurements at the EAR2 area, where instantaneous count-rates become very large for some specific cross sections and considering the slow tail of SiPM. It is also important in measurements involving highly radioactive samples. The count rate capability directly impacts the final sensitivity of the detection set-up, because it allows to place the detector closer to the sample enhancing the detection sensitivity versus other detectors setup, e.g. large C_6D_6 , which can be placed, at most, at 15-20 cm from the sample. The count-rate capability can be quickly addressed by means of a thick gold-sample measurement, which shows a large number of resonances in the keV energy range (Configuration 6).
- *Overall detector performance:* The full detector performances will be evaluated, as usual, with the measurement of Au, C, ^{209}Bi , ^{56}Fe samples and Empty frame. Although some of these measurements are not expected to vary a lot, from one type of detector to another, it is important to address the different background levels in order to design a future array optimized for the different background contributions (Configurations 1-6).

In Tab. 1 are described all the configurations involved in the aforementioned aspects to be investigated and proton requested to accomplish each task.

Configuration	Target	protons $\times 10^{17}$
1	Thin ^{197}Au	0.3
2	Empty	0.4
3	Fe	2
4	^{209}Bi	2.5
5	C	0.5
6	Thick ^{197}Au	0.3

Table 1: Summary of individual configurations required for the proposal, targets and protons devoted.

It is foreseen to calibrate all the detectors at the beginning of the run by means of standard γ -ray calibration sources ^{137}Cs , ^{88}Y , ^{207}Bi , Am/Be and Pu/C. An additional detector gain verification will be performed at the end of the experiment with one or more calibration sources mentioned before.

Summary of requested protons: 6.0×10^{17} protons on target .

References

- [1] U. Abbondanno et al. New experimental validation of the pulse height weighting technique for capture cross-section measurements. *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, 521(2):454–467, 2004.
- [2] U. Abbondanno et al. Neutron capture cross section measurement of ^{151}Sm at the cern neutron time of flight facility (n_tof). *Phys. Rev. Lett.*, 93:161103, Oct 2004.
- [3] R. Plag et al. An optimized c_6d_6 detector for studies of resonance-dominated (n,γ) cross-sections. *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, 496(2):425–436, 2003.
- [4] F. Käppeler et al. The s process: Nuclear physics, stellar models, and observations. *Rev. Mod. Phys.*, 83:157–193, Apr 2011.
- [5] C. Domingo-Pardo et al. New measurement of neutron capture resonances in ^{209}Bi . *Phys. Rev. C*, 74:025807, Aug 2006.
- [6] P. Mastinu and t. n. Collaboration. New c_6d_6 detectors: reduced neutron sensitivity and improved safety, 2013.
- [7] F. Becchetti et al. Deuterated stilbene (stilbene-d12): An improved detector for fast neutrons. *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, 908:376–382, 2018.
- [8] J. Balibrea-Correa et al. A first prototype of c_6d_6 total-energy detector with sipm readout for neutron capture time-of-flight experiments. *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, 985:164709, 2021.
- [9] V. Alcayne and t. n. Collaboration. A segmented total energy detector (sted) for (n,γ) cross section measurements at n_tof ear2 (nd2022), 2022.
- [10] J. Leredegui and t. n. Collaboration. New detection systems for an enhanced sensitivity in key stellar(n ,) measurements, 2023.
- [11] J. Balibrea-Correa et al. First measurement of the $^{94}\text{Nb}(n,)$ cross section at the cern n_tof facility, 2023.
- [12] Alcayne, V. et al. Measurement of the 244cm capture cross sections at both cern n_tof experimental areas. *EPJ Web Conf.*, 239:01034, 2020.

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 <u>fixed</u> installation you will be using [Name fixed/present n_TOF installation: 3 sTED and 1 C ₆ D ₆ detectors]	<input checked="" type="checkbox"/> To be used without any modification <input type="checkbox"/> To be modified
If relevant, describe here the name of the <u>flexible/transported</u> equipment you will bring to CERN from your Institute Instituto de Física Corpuscular (IFIC): <ul style="list-style-type: none"> • 25×25×13 mm³ stilbene-d₁₂ • Standard hamamatsu photo-multiplier, Hamamatsu SiPM and specific pcb read-out 	Standard equipment supplied by a manufacturer <input type="checkbox"/> CERN/collaboration responsible for the design and/or manufacturing
INFN Sezione di Catania : <ul style="list-style-type: none"> • 25×25×13 mm³ stilbene-d₁₂ crystal • four cylindrical (25.4 mm diameter × 25.4 mm length) standard stilbene crystals • Standard hamamatsu photo-multipliers with DC-DC HV converters 	Standard equipment supplied by a manufacturer <input type="checkbox"/> CERN/collaboration responsible for the design and/or manufacturing

HAZARDS GENERATED BY THE EXPERIMENT

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

Domain	Hazards/Hazardous Activities	Description
Mechanical Safety	Pressure	<input type="checkbox"/> [pressure] [bar], [volume][l]
	Vacuum	<input type="checkbox"/>
	Machine tools	<input type="checkbox"/>
	Mechanical energy (moving parts)	<input type="checkbox"/>

	Hot/Cold surfaces	<input type="checkbox"/>	
Cryogenic Safety	Cryogenic fluid	<input type="checkbox"/>	[fluid] [m3]
Electrical Safety	Electrical equipment and installations	<input type="checkbox"/>	[voltage] [V], [current] [A]
	High Voltage equipment	<input type="checkbox"/>	[voltage] [V]
Chemical Safety	CMR (carcinogens, mutagens and toxic to reproduction)	<input type="checkbox"/>	[fluid], [quantity]
	Toxic/Irritant	<input type="checkbox"/>	[fluid], [quantity]
	Corrosive	<input type="checkbox"/>	[fluid], [quantity]
	Oxidizing	<input type="checkbox"/>	[fluid], [quantity]
	Flammable/Potentially explosive atmospheres	<input type="checkbox"/>	[fluid], [quantity]
	Dangerous for the environment	<input type="checkbox"/>	[fluid], [quantity]
Non-ionizing radiation Safety	Laser	<input type="checkbox"/>	[laser], [class]
	UV light	<input type="checkbox"/>	
	Magnetic field	<input type="checkbox"/>	[magnetic field] [T]
Workplace	Excessive noise	<input type="checkbox"/>	
	Working outside normal working hours	<input type="checkbox"/>	
	Working at height (climbing platforms, etc.)	<input type="checkbox"/>	
	Outdoor activities	<input type="checkbox"/>	
Fire Safety	Ignition sources	<input type="checkbox"/>	
	Combustible Materials	<input type="checkbox"/>	
	Hot Work (e.g. welding, grinding)	<input type="checkbox"/>	
Other hazards			