EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Exploring new frontiers of neutron inelastic cross section measurements at n_TOF: testing the performances of a mixed array of HPGe and $\mathrm{LaBr_3(Ce)}$ detectors in beam

September 26, 2023

G. Alpar[1], S. Amaducci[2], M. Bacak[3], M. Boromiza[4], A. Casanovas[3,5,6], N. Colonna[7], L. Cosentino[2], M. Diakaki[8], P. Finocchiaro[2], A. Gandhi[4], M. Kokkoris[8],G. Lorusso[9], P. Mastinu[10], C. Neacsu[4], A. Negret[4], T. O'Donnell[1], A. Olacel[4], N. Patronis[3,11], C. Petrone[4], I. Porras[12], E. Stamati[3,11], G. Tagliente[7], P. Torres-Sánchez[12], R. Vlastou[8], T. Wright[13] and the n_TOF Collaboration[14]

[1] *University of Dallas, USA*
[2] *INFN Laboratori Nazionali del Sud, Catania, Italy*
[3] *European Organization for Nuclear Research (CERN), Switzerland*
[4] *Horia Hulubei National Institute for R&D in Physics and Nuclear Engineering, Romania*
[5] *Institut de Tecniques Energetiques (INTE) - Universitat Politecnica de Catalunya, Spain*
[6] *Instituto de Fısica Corpuscular, CSIC - Universidad de Valencia, Spain*
[7] *Instituto Nazionale di Fisica Nucleare, Sezione di Bari, Italy*
[8] *National Technical University of Athens, Greece*
[9] *National Physical Laboratory, UK*
[10] *INFN Laboratori Nazionali di Legnaro, Legnaro, Italy*
[11] *University of Ioannina, Greece*
[12] *University of Granada, Spain*
[13] *University of Manchester, United Kingdom*
[14] *www.cern.ch/ntof*

Spokesperson: C. Petrone (cristina.petrone@nipne.ro), M. Bacak (michael.bacak@cern.ch)
Technical coordinator: O. Aberle (oliver.aberle@cern.ch)

Abstract: Accurate knowledge of a wide variety of nuclear data is needed for a complete understanding of the fundamental processes occurring in nature and represent the building blocks for the next generation of nuclear technologies. In particular, the cross section of fast neutron induced reactions are important for the design of the Gen IV nuclear reactors: four out of the six prototypes considered are fast reactor types. Fusion reactors are another type of application that require high precision neutron inelastic cross section data. Usually, low uncertainties are obtained for neutron inelastic scattering cross-sections when studied through high-energy resolution γ spectroscopy. However, this method can be applied only for energy ranges for which the decay information of the populated states is known completely. Further improvements could be achieved by combining the high energy resolution of HPGe detectors with the excellent timing response and large efficiency of $\mathrm{LaBr_3(Ce)}$ crystals. Owing to its excellent neutron beam characteristics, n_TOF is the ideal facility for testing the feasibility of measuring the neutron induced inelastic channel with a mixed array of HPGe and $\mathrm{LaBr_3(Ce)}$ detectors.

Requested protons: $1.4 \cdot 10^{18}$ protons on target, (split into 2 runs over 1 year)
Experimental Area: EAR1

1 Physics Motivation

New developments of nuclear applications, such as the next generation of reactors (Gen IV) or accelerator driven systems (ADS), require precise knowledge of fast neutron induced reaction cross-sections for specific materials. Fast neutrons mostly interact by elastic and inelastic scattering, while at high energies other threshold reactions (e.g. n,xn) become also important. Consequently, the design of these facilities requires a very precise knowledge of the neutron induced reaction cross sections for a wide range of incident energies. Information such as cross sections and neutron angular distributions determine the final energy spectrum and spatial diffusion of neutrons inside the reactor core and therefore represent crucial parameters for evaluating the reaction rates [1]. Neutron inelastic scattering reactions are also responsible for neutron damage, and their cross sections are used to estimate the lifetime of structural materials, an important aspect for the development of fusion reactors [2]. Usually for these specific applications, low uncertainties in the range of 5 to 20% are required [3]. Considering the characteristics of the n_TOF pulsed neutron source, the nuclear data community could substantially benefit from inelastic measurements performed at CERN. In particular, the energy distribution of its neutron beam [4], shown in Figure 1, reaches maximum values in the energy range of the evaporation peak, between 100 keV and 10 MeV, an energy region of interest for neutron inelastic scattering measurements. Moreover, time structure of the PS pulse (with a FWHM of only few ns), results in a narrow resolution function of n_TOF neutron beam, an essential characteristic for resolving the compound nucleus resonances. Moreover, the presence of a high energy component in the neutron spectrum, extending up to hundreds of MeV, would allow to collect data well above the current limits, allowing to refine theoretical models.

Neutron inelastic scattering reactions can be studied experimentally by detecting either the scattered neutrons or the γ rays emitted during the de-excitation of the target nucleus. The advantages of the γ-spectroscopy method are the superior sensitivity and higher resolution [5]. Up to now, numerous such high-resolution measurements using γ spectroscopy that employed arrays of high purity germanium (HPGe) detectors have been performed at various facilities, providing data with low uncertainties [6–8]. At n_TOF, similarly to other neutron spallation facilities, although the neutron flux characteristics are excellent for performing such measurements, the implementation of HPGe based detection system was not possible due to high sensitivity of this type of detector to the so-called γ-flash, and the long recovery time, typically of the order of hundreds of μs. Only recently, following the development of a dedicated HPGe detector prototype equipped with a specially designed preamplifier [9], inelastic cross section on ^{56}Fe and ^{7}Li could be investigated for the first time [10]. Recent in-beam tests, showed that neutron energies in the range of few MeV can be achieved with this HPGe prototype [11].

Although it has its advantages, the γ-spectroscopy method can only be applied for studying neutron inelastic process for the sequence of levels starting from the first excited level up to the highest excited level for which decay information is known completely and for which levels with lower excitation energy were not omitted [1]. Incomplete information or missed transitions for higher levels will result in an underestimated total inelastic cross-section. Such situations can

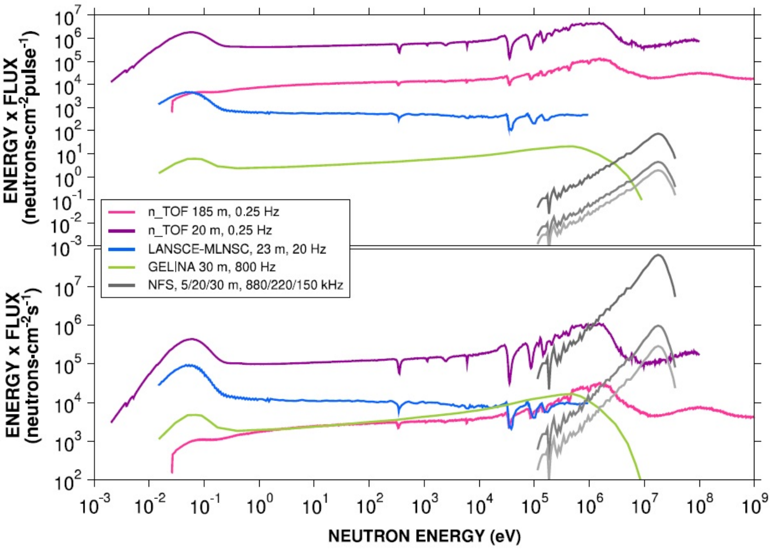

Figure 1: The energy distribution of neutrons for several TOF facilities, expressed as neutrons per logarithmic energy bin (unit of lethargy), either per second (average flux- bottom image) or per time-of-flight pulse (instantaneous flux - top image), adopted from [4].

be improved if we couple the HPGe detectors, known for their excellent energy resolution, with high efficiency detectors such as LaBr$_3$(Ce) crystals. In the last two decades, this new type of inorganic scintillator has found many applications in γ-ray spectroscopy mostly due to the high detection efficiency that comes along with the relatively good energy resolution. In comparison with HPGe detectors, higher intrinsic efficiencies can be obtained because of the larger density and effective atomic number [12]. Another very important advantage of LaBr$_3$(Ce) crystals, relative to HPGe detectors, is their fast timing response, which minimizes the signal pile-up problem and, in principle, should allow to reach neutron energies up to 100 MeV. When it comes to neutron sensitivity, the major asset of LaBr$_3$(Ce) crystals derives from the fact that the elastic scattering process generates signals in the scintillation crystal through the energy loss of recoiling nuclei and since the crystal contains large masses of the nuclei, the recoil energies are smaller and therefore produce a negligible light, contrary to the semiconductor detector behaviour. Nevertheless, due to their limited energy resolution there are only few physics cases (i.e. nuclei with energy levels well-spaced apart) for which the neutron inelastic cross section could be studied exclusively with LaBr$_3$(Ce) scintillators. Consequently, we propose, as a first attempt to measure neutron inelastic cross section with LaBr$_3$(Ce) crystals, to adopt a conservative approach by coupling them with the recently tested HPGe prototype. In this way, the presence of possible contaminant peaks in the energy range of interest can be checked using the HPGe. At the same time, the excellent time response of LaBr$_3$(Ce) detectors combined with the minimal time-width of the n_TOF neutron pulsed beam, will allow us to determine with unprecedented precision the energy of the neutrons with values ranging from 1 keV at 1 MeV up to 4 keV at 10 MeV. Moreover, by taking advantage of the LaBr$_3$(Ce) high efficiency, γ-γ coincidence analysis can be performed to extract information about the feeding pattern of different states and extend the available information about the level cross section.

One physics case that complies with the requirements discussed above for testing the feasibility of performing neutron inelastic cross section measurements with a mixed array of HPGe and LaBr$_3$(Ce) crystals, is ^{24}Mg. The neutron inelastic cross section on ^{24}Mg is known with very good precision from a previous measurement performed at GELINA [8] by a team partially involved in the current proposal. The level scheme of the states populated in this reaction is shown in Figure 2, the transitions observed in the GELINA experiment and for which the production cross sections were measured, are marked with continuous arrows. In total, six transitions could be observed with GAINS spectrometer, the closest in energy being the 2754 keV ($4_1^+ \rightarrow 2_1^+$ transition) and 2869 keV ($2_2^+ \rightarrow 2_1^+$ transition). GEANT4 simulations [13] were performed to check if the energy resolution of the LaBr$_3$(Ce) allows a clear separation of the two peaks, the resulting spectrum is displayed in Figure 3. The energy resolution included in the simulation is based on the experimental value obtained with a ^{56}Co radioactive source.

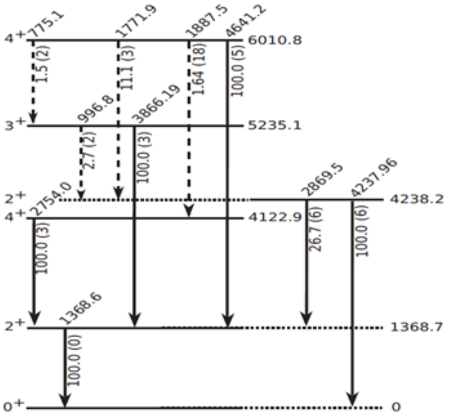

Figure 2: Excited states of ^{24}Mg observed in the neutron inelastic cross section measurement performed at GELINA [8].

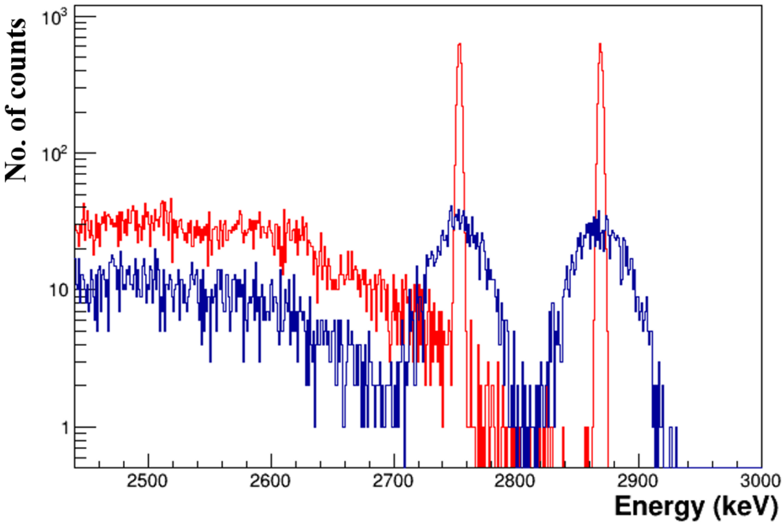

Figure 3: Simulated energy spectra of HPGe and LaBr$_3$(Ce) detectors for the 2754 and 2869 keV γ-rays produced by inelastically scattered neutrons on ^{24}Mg sample.

Apart from being an excellent physics case to be studied with LaBr$_3$(Ce) detectors (i.e. low level density, well-known level scheme and high energy γ-rays), measuring neutron inelastic cross section on ^{24}Mg is also relevant for the design of different nuclear applications. Case in point: the European Facility for Industrial Transmutation (EFIT), which is an accelerator driven system (ADS) with a subcritical core loaded with dedicated fuels containing minor actinides (MA) and cooled by liquid lead. Magnesium is an important component of the primary candidates considered as possible fuels for this application: CERamic-CERamic (CERCER) and CERamic-Metallic (CERMET) inert matrix fuels (IMF) consisting of (Pu,MA)O_2-x particle dispersed in MgO and Mo inert matrices, respectively [14]. Neutron-induced reactions cross sections on ^{24}Mg are also relevant for the design of the generation IV reactors [15]: small quantities of ^{24}Mg can accumulate inside the sodium-cooled fast reactor (SFR, a Gen IV reactor prototype) following the capture of neutrons by ^{23}Na, that forms ^{24}Na ($T_{1/2}$ ≈15 h). Moreover, in medical applications, for the design of neutron beams in boron neutron capture therapy, magnesium is proposed as one of the main moderator components of beam shaping assemblies (e.g. within MgF_2 or in combination with other metal halides) [16].

In summary, the aim of this LoI is to test the abilities of the in beam γ-ray spectroscopy at n_TOF for (n,n') and (n,xn) reaction studies using a mixed detection setup made of the HPGe detector (known for their high energy resolution) and $LaBr_3$(Ce) detectors (that have a fast response and higher intrinsic efficiency). If proven to be feasible, performing in the future measurements at CERN for the neutron inelastic channel using $LaBr_3$(Ce) coupled with HPGe detectors will add redundancy and reliability for the data already measured at GELINA and other facilities. Furthermore, the significantly higher n_TOF neutron flux above 10 MeV as compared to the other facilities (see Figure 1), should improve the statistical uncertainty of the previous datasets and even extend the (n,xn) data to higher neutron energies.

2 Experimental set-up and beam time request

As discussed in the previous section, due to the severe γ-flash effect, specific to the n_TOF environment, performing inelastic cross section measurements was not possible until recently at this facility. Given its high efficiency for detecting γ-rays, one can expect for the $LaBr_3$(Ce) crystal a similar behaviour to the HPGe detector. However, the recovery time should be significantly shorter because of the fast response of the scintillators. Indeed, short tests performed in parasitic mode, have showed that depending on the detector-target configuration, recovery times after the γ-flash as short as 1 μs can be reached with $LaBr_3$(Ce) crystals. Nevertheless, to assure a complete understating of this detector behaviour in beam conditions, we propose to test two different readout systems for the light output: one based on the classical photomultiplier tube (PMT) and a second one, based on a gated silicon photomultiplier (SiPM). Each one of these systems have their advantages and weak points. Depending on the crystal size and sensitivity to the γ-flash, a gating circuit might be needed to prevent the saturation of the readout electronics. From this point of view, the design of a gating circuit is easier to be implemented for SiPMs due to their low operation bias. On the other side, the timing characteristics of the SiPM signals are slower compared with the PMT's and this might result in a larger probability of having pile-up signals at high count rates, causing a deterioration of the energy resolution. High count rates are known to affect also the gain of PMT, resulting also in a worse energy resolution or even dead time effects. We propose to investigate all these possible issues in a two-step test experiment with dedicated beam time in EAR1. This approach will allow us to determine the in-beam performances of the two readout systems independently.

For each test run the detection system will consist of two $LaBr_3$(Ce) crystals coupled to either PMTs or SiPMs, placed at 110° and 150° with respect to the beam direction, and the HPGe detector placed on the opposite side of the target at 125°, as shown Figure 4. We prefer to mount the detectors at backward angles to avoid the excessive background energy deposition caused by the γ-flash. Furthermore, the selected angles will allow the angle integration of the differential cross section data, following the procedure described in [8].

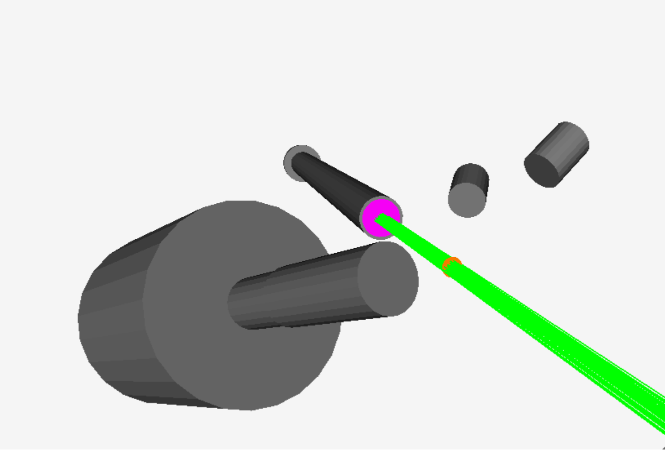

Figure 4: Proposed experimental setup for measuring the neutron inelastic cross section with two $LaBr_3(Ce)$ crystals and the HPGe prototype in EAR1.

Photopeak efficiency for one $LaBr_3(Ce)$ crystal of 2 inch at 1368 keV (the first excited state in ^{24}Mg), was evaluated to be ~0.12% from tests with ^{152}Eu, ^{60}Co and ^{137}Cs radioactive sources placed at 10 cm from the detector. The detector response as a function of count rates will be adjusted during the experiment by modifying the detector-sample distance or by using one or two natural metallic samples of Mg (78.99% of ^{24}Mg), each one having a thickness of 0.25 mm. In Figure 5 the estimated count rates for a run of $7 \cdot 10^{17}$ protons ($\sim$ 1 week beam time), considering the photopeak efficiency at 1368 keV and the associated γ production cross section, is presented.

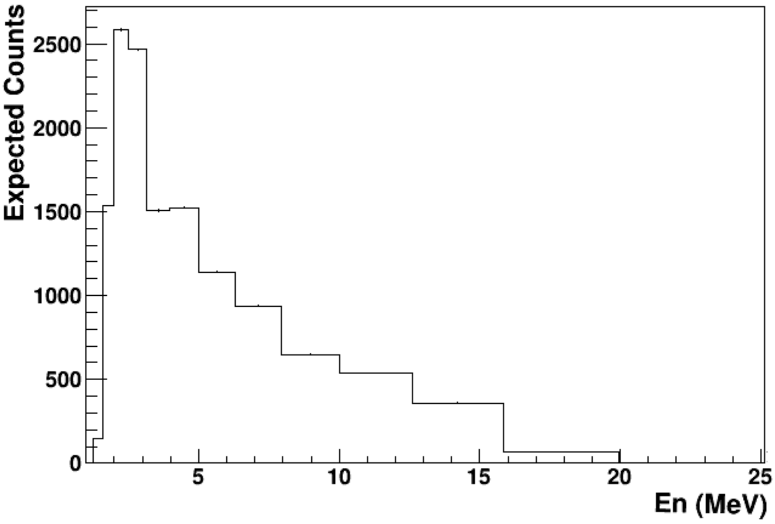

Figure 5: Expected number of counts for 10 bpd for $7 \cdot 10^{17}$ protons (~1 week beam time) for the $LaBr_3(Ce)$ detector at 10 cm distance with respect to the ^{24}Mg sample.

Summary of requested protons: $1.4 \cdot 10^{18}$ protons split into 2 runs.

References

[1] L.C. Mihailescu, L. Olah, C. Borcea, A.J.M. Plompen, *A new HPGe setup at Gelina for measurement of gamma-ray production cross-sections from inelastic neutron scattering*, Nucl. Instr. Phys. A 531, 375-391, 2004

[2] S. J. Zinkle and J.T. Busby, *Structural Materials for Fission & Fusion Energy*, Mater Today 12, Issue 11, 12–19, 2009.

[3] G. Aliberti et al., *Impact of Nuclear Data Uncertainties on Transmutation of Actinides in Accelerator-Driven Assemblies*, Nuclear Science and Engineering 146, 13-50, 2004

[4] N.Colonna et al., *Neutron physics with accelerators*, Progress in Particle and Nuclear Physics 101,177–203, 2018

[5] D. A. Lind, *Studies of Gamma Rays from Neutron inelastic scattering*, Annals of Physics 12, 485-532, 1961

[6] R. O Nelson, N. Fotiades, M. Devlin, J. A. Becker, P. E. Garrett, and W. Younes, *Cross Section Standards for Neutron Induced Gamma-Ray Production in the MeV Energy Range*, AIP Conf. Proc. 769, 838, 2005

[7] A. Negret et al., *Cross-section measurements for the* $^{56}Fe(n, xn)$ *reactions*, Phys. Rev. C 90, 034602, 2014

[8] A. Olacel et al., *Neutron inelastic cross-section measurements for* ^{24}Mg, Phys. Rev. C 90, 034603, 2014

[9] S. Urlass et al., *Gating of charge sensitive preamplifiers for the use at pulsed radiation sources*, Nucl. Instr. Phys. A 1002, 165297, 2021

[10] https://cds.cern.ch/record/2765698?ln=en

[11] M. Bacak et al., *First results from (n,n') XS measurements with a gated HPGE at EAR1*, n_TOF Data Analysis Meeting, May 2023, CERN

[12] E.V.D. van Loef et al., *Scintillation properties of* $LaBr_3{:}Ce^{3+}$ *crystals: fast, efficient and high-energy-resolution scintillators*, Nuclear Instruments and Methods in Physics Research Section A 486, 254, 2002

[13] S. Agostinelli et al., *Geant4 - a simulation toolkit*, Nucl. Instr. Phys. A 531, 506, 250-303, 2003

[14] F. Gabrielli et al., *Fuel for ADS: State-of-the-art, requirements, current and future programmes*, NEA-NSC-DOC–2015-7, Nuclear Energy Agency of the OECD (NEA), 2015

[15] https://www.gen-4.org

[16] INTERNATIONAL ATOMIC ENERGY AGENCY, Advances in Boron Neutron Capture Therapy, Non-serial Publications , IAEA, Vienna (2023)

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
HPGe and 2 $LaBr_3$(Ce) crystals	☒ To be used without any modification
Two stable natural metallic samples of Mg (0.25mm thickness)	☒ Standard equipment supplied by a manufacturer

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			