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## GRAPhEME: performances, achievements (@EC-JRC/GELINA) and future (@GANIL/SPIRAL2/NFS)

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**Abstract.** GRAPhEME is a  $\gamma$ -spectrometer developed by CNRS/IPHC Strasbourg (France), in collaboration with EC-JRC Geel (Belgium) and IFIN-HH Bucharest (Romania). With its 6 High Purity Planar Germanium detectors and one fission chamber, GRAPhEME, installed at the EC-JRC GELINA facility, was optimized for measurements of accurate  $(n, xn\gamma)$  cross sections on actinides. The experimental methodology is based on the prompt  $\gamma$ -ray spectroscopy coupled to time-of-flight measurements. In this paper, we present an overview of fifteen years of experiments with GRAPhEME at EC-JRC GELINA facility, illustrated by main achievements to highlight the performances reached by our spectrometer. Beyond the experimental work, a close collaboration with theoreticians has emerged allowing the use of the data produced with GRAPhEME to test and constraint nuclear reaction codes like TALYS, CoH and EMPIRE. In a near future, GRAPhEME will be available to start measurement campaigns at the new neutron beam facility SPIRAL2/NFS. There, studies of  $(n, 2n)$  and  $(n, 3n)$  reactions will be possible and will complete the work done at EC-JRC GELINA on  $(n, n')$  reactions. Despite the amount of cross section data provided by GRAPhEME up to now, the prompt  $\gamma$ -ray spectroscopy method presents some weaknesses that our collaboration tries to overcome. This goes through new calculation schemes based on theoretical modeling constrained on experimental data to infer the total  $(n, xn)$  cross section, new instrument to measure conversion electrons but also by being proactive in dissemination activities to make the nuclear structure community aware of our needs about new accurate nuclear structure information on actinides.

### 1 Introduction

In the context of improvement of nuclear data bases for nuclear energy application, our collaboration IPHC-CNRS (Strasbourg, France), EC-JRC-Geel (Belgium) and IFIN-HH (Bucharest, Romania) has developed  $\gamma$ -spectrometers for the study of neutron inelastic scattering and  $(n, xn)$  reactions. These reactions are of importance in reactors as they partially drive the neutron population evolution and can also produce radioactive element that can cause radio-protection issues. In the nineties and after, several studies [1, 2] have shown the importance of improving the knowledge of neutron inelastic scattering and despite experimental and modeling efforts, there is still today room for improvements. Although the inelastic process can be experimentally studied by different methods like neutron detection, activation technique or prompt  $\gamma$ -ray spectroscopy,

the measurement is always a challenge and there is no ideal method. Each has strength and weaknesses. In the case of the prompt  $\gamma$ -ray spectroscopy method (PGRS), the reaction channel can be well identified due to high detector energy resolution (HPGe detector) contrary to the case of the neutron detection method. Time-of-flight measurements are possible with PGRS method contrary to the activation one where the measurement of the  $\gamma$  of interest is performed off-line. Finally, the PGRS method gives access to the exclusive  $(n, xn\gamma)$  cross sections that can be used in combination with nuclear structure information to infer the level production and  $(n, xn)$  total cross section. Fifty years ago, during the seventies, this method was used for the first time for  $(n, xn)$  reaction studies at the Karlsruhe cyclotron [3] and at the Oak Ridge electron linear accelerator ORELA [4] with Ge(Li) and NaI detectors respectively. Twenty years after, a step has been taken with the use of this method by the advent of the GEANIE high

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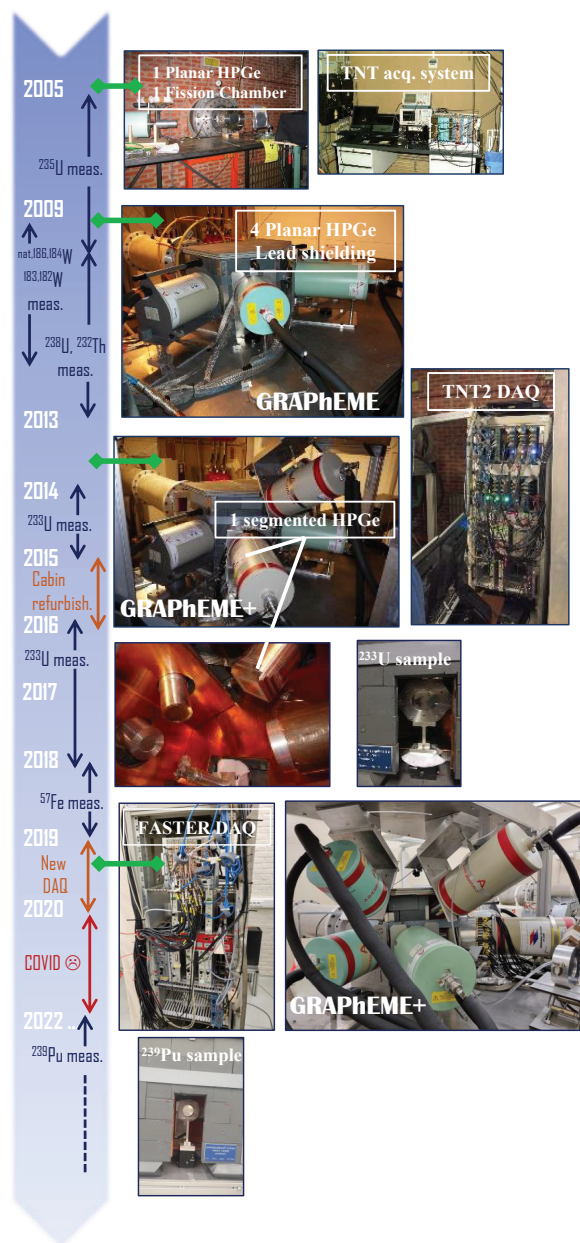
purity Germanium array at the WNR spallation time-of-flight facility of Los-Alamos. Then, since the middle of the nineties several experiments have been performed at WNR on a large variety of isotopes (Pb, Al, Pt, Mo, Ti, Sm but also U and Pu, etc) and for the neutron energy range from a few MeV up to 400 MeV (see [5] and references therein for examples). In the beginning of the 2000's, our collaboration took advantage of the multi-users facility GELINA, at EC-JRC-Geel in Belgium, to develop two  $\gamma$ -spectrometers dedicated to the experimental study of the neutron inelastic scattering and  $(n, xn)$  reactions by the prompt  $\gamma$ -ray spectroscopy coupled to time-of-flight measurement. While the GAINS (Gamma Array for Inelastic Neutron Scattering) setup, placed at a long flight path (100 m) [6], is mainly devoted to measurements on light and intermediate mass nuclei, GRAPhEME (GeRmanium array for Actinides PrEciSe MEasurements) is placed at 30 m from the neutron source and has been optimized for measurements on actinides. In this contribution, we propose an overview of the almost twenty years of  $(n, xn\gamma)$  measurements with GRAPhEME at EC-JRC GELINA facility (section 2) by highlighting the main conclusions obtained on each studied nucleus. In section 3, we present the project we aim to conduct at the recently commissioned GANIL/SPIRAL2/NFS facility. Finally, in the last section (4), we will address some shortcomings of the PGRS method and the strategy we develop to overcome them. It should be noted that this contribution is completed by five other papers published in this proceeding, which will present in more details some items mentioned in this general overview of the work performed by our collaboration with GRAPhEME.

## 2 GRAPhEME at EC-JRC GELINA

### 2.1 Story of GRAPhEME

The story of GRAPhEME has started in 2005 at the EC-JRC GELINA facility, with one High Purity Germanium detector and one Fission Chamber ( $^{235}\text{U}$  deposit) coupled to a digital TNT acquisition system developed at our laboratory (IPHC, Strasbourg). The setup is installed at 30 m from the GELINA neutron source, a distance which appeared as a good compromise between neutron energy resolution and available actinides quantities for sample making. After 4 years of setting up, GRAPhEME was mature, in 2009, to start measurement campaigns. In this configuration, it was composed of four HPGe detectors surrounded by a Pu-Cd-Cu shielding to protect the Germanium crystal from neutron and  $\gamma$  background. Between 2009 and 2014 several experimental campaigns have been performed on  $^{235}\text{U}$  [7],  $^{238}\text{U}$  [8],  $^{232}\text{Th}$  [9] and on stable isotopes of tungsten ( $^{nat,182,183,184,186}\text{W}$ ) [10]. The first upgrade of GRAPhEME was in 2014 with the addition of a segmented HPGe in view of measurements with very active targets. The digital acquisition system was also upgraded to accept more than 40 channels and measurement started on  $^{233}\text{U}$ . The second upgrade was made in 2019 and has concerned the replacement of the TNT acquisition system, become obsolete, by the new FASTER one

[11]. The COVID pandemic has delayed the starting of measurement on  $^{239}\text{Pu}$  during two years but it has finally started in June 2022. Figure 1 shows in a synthetic and illustrated way, the evolution of GRAPhEME over the years and the different measurement campaigns achieved up to now.



**Figure 1.** (color online) Timeline illustrating almost 20 years of GRAPhEME developments and associated measurements.

### 2.2 GRAPhEME today

Today, GRAPhEME is composed of one fission chamber and 6 HPGe detectors coupled to the FASTER digital acquisition system. The  $^{235}\text{U}$  ionization chamber, used for neutron flux determination, is positioned  $\approx 1$  meter upstream the sample. The deposit is made of  $^{235}\text{UF}_4$  with an areal density of  $0.324(2)$   $\text{mg}/\text{cm}^2$ . The six Germanium

detectors are planar types (with typical thickness around 2-3 cm and diameter around 5-6 cm). They are placed around the sample at  $110^\circ$  and  $150^\circ$  with regards to the neutron beam to allow the precise angular integration of the  $\gamma$ -ray production cross sections. One HPGe detector is segmented in 36 pixels. It allows to manage high counting rates without too high dead time and its very good energy resolution gives a precise  $\gamma$ -peak identification. These are real assets when performing measurement with highly active and fissile actinide samples. At a flight path of 30 m and with the time resolution of the Germanium detectors, the typical neutron energy resolution achieved is 10 keV at  $En = 1 \text{ MeV}$ . The current version of GRAPhEME will be used for the  $^{239}\text{Pu}$  measurements. In previous measurements, depending of the nucleus, the sample thickness ranged from 0.2 to 1.3 mm, the mass ranged from 8.3 to 48.4 g and the total beam time, for a measurement on one nucleus, varied between 527 hours and 4500 hours. Finally, GRAPhEME at GELINA was able to produce numerous  $(n, xn\gamma)$  cross sections ( $x = 1, 2, 3$ ) with total uncertainties ranging from 3% to 20% depending on the incident neutron energy. In the energy range of  $(n, n')$  reaction for which the GELINA neutron beam is very well adapted, the total uncertainty ranges from 3 to 5%. In the next section, an overview of the different campaigns made with GRAPhEME at GELINA is presented illustrated with a selection of important results for each studied nucleus.

## 2.3 GRAPhEME main achievements

### 2.3.1 Measurements on $^{238}\text{U}$ and $^{232}\text{Th}$

After a first measurement campaign on  $^{235}\text{U}$  which led to the determination of one  $^{235}\text{U}(n, n'\gamma)$  cross section and three  $^{235}\text{U}(n, 2n\gamma)$  cross sections [7], data collected on  $^{238}\text{U}$  [8] and  $^{232}\text{Th}$  [9] allow the determination of large sets of  $(n, xn\gamma)$  cross sections. For  $^{238}\text{U}$ , 36  $\gamma$  transitions have been analyzed. In most cases, the obtained cross sections compare well with existing data especially with the data from Fotiades *et al.* [5] obtained with the GEANIE spectrometer. Moreover, in the first published data set [8], we show 5  $\gamma$ -production cross sections never measured before. For  $^{232}\text{Th}$ , not less than 81  $\gamma$  transitions have been observed and led to cross section data in good agreement with the only available previous measurement performed by Dave *et al.* [12]. The cross section data set obtained with GRAPhEME is more comprehensive than the one from Dave *et al.* as we were able to measure 24  $\gamma$ -production cross sections never measured before especially for  $\gamma$ -energy transitions below  $\approx 600$  keV. The neutron energy range of our data starts from the threshold to  $\approx 20$  MeV while for Dave *et al.*, the maximum incident neutron energy is 2.1 MeV.

An important asset of both these sets of data on  $^{238}\text{U}$  and on  $^{232}\text{Th}$  is that we were able, for the first time, to measure the de-excitation of the first level which carries the most part of the total  $\gamma$  cascade and thus provides useful information on the total  $(n, n')$  cross section. This is illustrated in the contribution [13] in these proceedings.

With our new experimental data, using the  $\gamma$ -transitions intensity for transitions coming from a same

level, we were also able to test some branching ratio information coming from the ENSDF database [14]. It has been shown that, both for  $^{238}\text{U}$  [8] and  $^{232}\text{Th}$  [9], discrepancies exist between our results and the ENSDF database. This was the start of a more comprehensive study, based on sensitivity studies, of the role of nuclear structure knowledge and its impact in the prompt  $\gamma$ -ray spectroscopy method and in theoretical modeling. A conclusion, in the case of the  $^{238}\text{U}$  nucleus, was that, for some  $\gamma$  transitions, the sensitivity of  $(n, n'\gamma)$  cross section to branching ratio uncertainty can reach 0.4% per %. In ENSDF, the average branching ratio uncertainty is 8%, thus this information plays a significant role in the uncertainty estimation when inferring the  $(n, n')$  cross sections from  $(n, n'\gamma)$  ones. This kind of information can also be used to diagnose the reasons for differences between calculated and experimental data. More details on this study can be found in the contribution [15] in these proceedings.

Finally, our new cross section data for the  $^{238}\text{U}(n, n'\gamma)$  reaction have been used, in the frame of a close collaboration with theoreticians, to test modeling aspects especially relative to the description of compound nucleus and pre-equilibrium mechanisms. One important result of this work [16], is the validation of quantum mechanical microscopic approaches for the modeling of the pre-equilibrium process. Indeed, it has been demonstrated that these approaches (DWBA/1p-1h<sup>1</sup> [17] and QRPA<sup>2</sup> [18]), which explicitly calculate the spin-parity distribution, give more realistic distributions than the exciton model. These lead to a better calculation of  $(n, n'\gamma)$  cross sections for  $\gamma$  transitions at high spin in the ground state band as illustrated in Figure 2. Additionally to these results, it has been then possible to find new values of the spin cutoff parameter used in the prescription of Gruppelaar [19] which is included in the TALYS nuclear reaction code [20] to determine the spin-parity distribution in the frame of the exciton model. More details about the theoretical achievements relative to  $^{238}\text{U}(n, n'\gamma)$  cross section calculations can be found in [8].

### 2.3.2 Measurements on $^{nat,182,183,184,186}\text{W}$

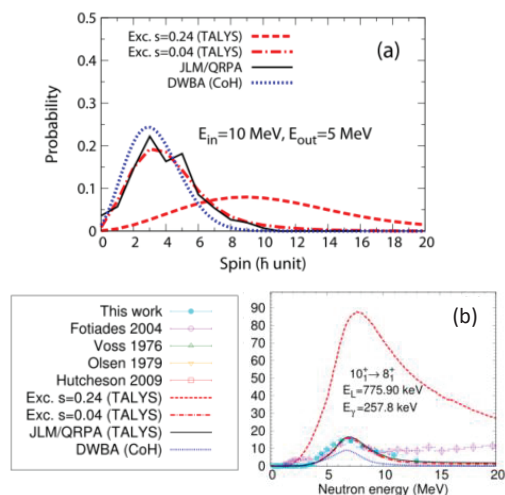
In addition to the measurement on a natural tungsten target, our collaboration had the opportunity to perform measurements on all stable isotopes of tungsten. The enriched samples were provided by the Oak Ridge National Laboratory (USA) with enrichment around 95% for isotopes 182, 184, 186 and around 85% for isotope 183. With GRAPhEME, we were then able to measure, for the first time, an average of twenty  $\gamma$ -production cross sections for each isotope, representing a comprehensive and unique set of data.

Contrary to the case of  $^{238}\text{U}$  and  $^{232}\text{Th}$  data analysis, for tungsten isotopes a Monte Carlo approach has been developed. It is detailed in [21]. This innovative methodology allows to propagate uncertainty in an easier way than in a total deterministic scheme but the major asset, in the

<sup>1</sup>Distorted Wave Born Approximation/one-particle one-hole excitation of the ground state

<sup>2</sup>Quasi-particle Random Phase Approximation





**Figure 2.** (color online) (a) Spin distribution for the incident energy 10 MeV and an excitation energy of 5 MeV in  $^{238}\text{U}$  calculated with different modelings. (b) Impact of these spin distribution on  $(n, n'\gamma)$  cross section for high spin level in the ground state band. Figures from [8].

case of time-of-flight measurements, is the possibility to take into account the uncertainty related to neutron time-of-flight resolution (which is very difficult in a classical calculation). Finally, the production of covariance and correlation matrices is also more straightforward.

As for  $^{238}\text{U}$ , a comprehensive work is in progress in collaboration with our theoretician colleagues using the three nuclear reaction codes TALYS, CoH3 [22] and EMPIRE [23], to test the quality of the cross sections modeling for those isotopes which are not fissionable but present a deformation like actinides nuclei. Preliminary results for  $^{nat,182,184,186}\text{W}$  can be found in [10], and for  $^{183}\text{W}$  a first presentation of preliminary data is given in this proceeding [24]. A first observation was that, including some discrete levels in the continuum (CoH3 code) gives a better  $(n, n'\gamma)$  cross section calculation for  $\gamma$  transition at high spin in the ground state band which is not the case for  $^{238}\text{U}$ . Secondly, it appears that the  $(n, n'\gamma)$  cross section for the  $2^+$  to  $0^+$   $\gamma$  transition in the ground state band predicted by the three codes presents a plateau, for neutron energy between 2 to 8 MeV, which is not observed in the measured data and this for the 3 even isotopes. Many experimental validity tests have been done to exclude any problems in the measured data, and today this difference remains a puzzle!

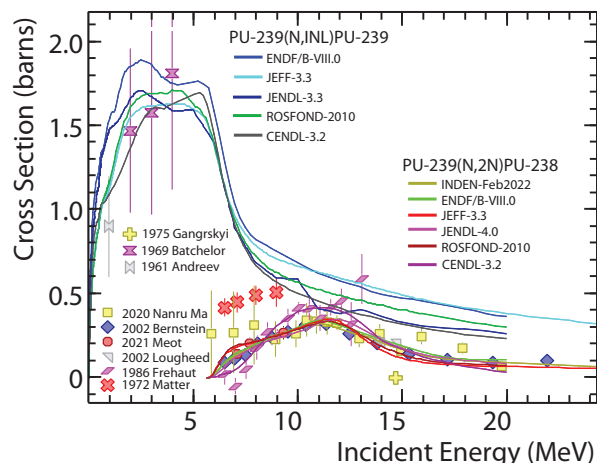
### 2.3.3 Measurements on $^{233}\text{U}$

At the very beginning, GRAPhEME was conceived to measure the  $^{233}\text{U}(n, 2n)$  cross section. With the experience of the first measurements performed on uranium isotopes (235 and 238), it was clear that, in its simple configuration and with monocrystal germanium detectors, GRAPhEME won't be able to reach the objective to measure  $(n, xn)$  reaction cross section for very active nuclei. The counting rate coming from the radioactivity of the sample will

induced a  $\gamma$  background but even more penalizing for accurate measurements, a huge dead time will be generated. The proposed solution was then to use a segmented germanium detector to reduce the counting rate on each active parts of the detector and reach an acceptable dead time. A new HPGe detector segmented in 36 pixels (surface and thickness of  $54 \times 54$  mm<sup>2</sup> and 20 mm respectively). The first tests performed in 2014 [25] have shown the very good capabilities of this new detector. The dead time has been reduced by a factor of 20 compared to the one observed in a classical HPGe detector to reach only a few percent. Moreover its very good energy resolution allows a precise peak identification which is crucial for data analysis in complex  $\gamma$  spectrum. After the refurbishment of the flight path cabin at 30 m and obtaining the sample (8.3 g of metallic  $^{233}\text{U}$  with a thickness of 0.64 mm for an activity of 3 GBq), the measurement started and finally 4500 hours of beam time were acquired. For the first time, after a difficult but meticulous data analysis,  $(n, n'\gamma)$  cross sections have been obtained for 12  $\gamma$  transitions in  $^{233}\text{U}$ . The contribution [26] in these proceedings presents the details of the data analysis and the obtained cross sections compared to TALYS calculations. As for other actinides measured with GRAPhEME, adjustments of nuclear models parameters will be required to improve the TALYS predictions with the specificity that, for  $^{233}\text{U}$ , no experimental data exist for neutron inelastic scattering process and nor for  $(n, 2n)$  reaction. Another important remark is that, at GELINA, due to the low neutron flux in the energy range of  $(n, 2n)$  reaction, the low cross section of this process and the high fission cross section (huge pollution in  $\gamma$  spectrum of peaks coming from fission product deexcitation), it was not possible to extract any  $^{233}\text{U}(n, 2n\gamma)$  cross section.

### 2.3.4 Measurements on $^{239}\text{Pu}$

Once the demonstration done on  $^{233}\text{U}$  that, with GRAPhEME, we are able to measure  $(n, xn\gamma)$  cross section on very active actinide, the second challenge has been launched. It concerns the measurement on  $^{239}\text{Pu}$  which is a key element in  $^{238}\text{U}/^{239}\text{Pu}$  fuel cycle and for which a lack of knowledge of  $(n, n')$  and  $(n, 2n)$  reactions is to be deplored. For  $(n, n')$  cross section data, only three sets of data are available in EXFOR, and for  $(n, 2n)$  reaction, more measurements exist but with important discrepancies especially just above the threshold of the reaction as illustrated in Figure 3. For this measurement, the first challenge was the preparation of the sample as free as possible from  $^{241}\text{Am}$ . From  $\text{PuO}_2$  powder provided by EC-JRC-Geel, SCK-CEN (Mol Belgium) managed to reduce by a factor of  $10^{-4}$  the activity ratio  $^{241}\text{Am}/^{239}\text{Pu}$  that reaches finally the value of  $\approx 1.5 \times 10^{-4}$ . With this material, EC-JRC-Geel has prepared a sample with a mass of 2.3 g, a diameter of  $\approx 5$  cm and a thickness around 0.1 cm. All this work has been performed in the frame of the workpackage 3 of the European project SANDA. After the upgraded of the data acquisition system with the new FASTER equipment, the measurement has started in fall 2022.

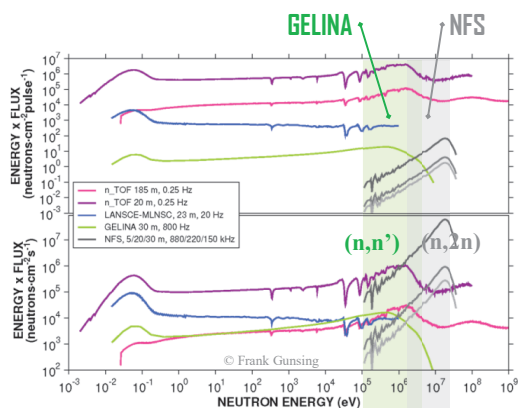


**Figure 3.** (color online) Evaluated and experimental cross section data for  $^{239}\text{Pu}(n, n')$  and  $(n, 2n)$  reactions (data have been obtained from EXFOR data library in September 2022).

### 3 GRAPHEME at GANIL/SPIRAL2/NFS

#### 3.1 Measurements at long flight path

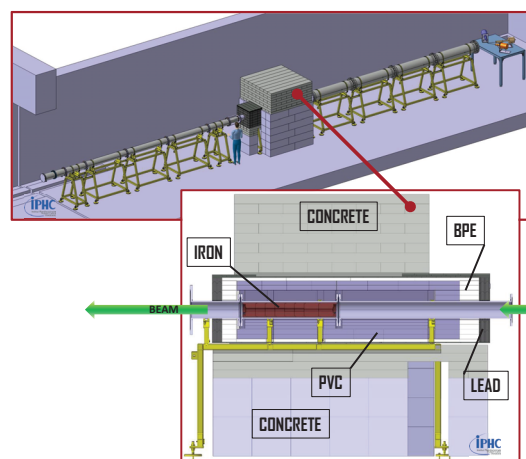
The arrival of the new neutron facility NFS (Neutrons for Science) at SPIRAL2/GANIL in Caen (France) [27] offers, among others, new opportunities to study  $(n, xn)$  reactions and especially  $(n, 2n)$  and  $(n, 3n)$ . This is shown on Figure 4 which compares neutron flux at different facilities and where neutron energy ranges for  $(n, n')$  and  $(n, 2n)$  reactions are highlighted. Our team (IPHC) has expressed



**Figure 4.** (color online) Neutron flux delivered by different time-of-flight facilities. Neutron energy ranges for  $(n, n')$  and  $(n, 2n)$  reactions are highlighted in green and grey respectively (Courtesy Frank Günsing).

from the origins of the NFS project its interest to study  $(n, xn)$  reactions by prompt  $\gamma$ -ray spectroscopy. We were strongly involved in the preparation of the experimental area at long flight path. This investment resulted in taking charge of the definition of the design, construction and installation of the neutron beam line and the second collimator and its shielding. Indeed, it was, first, very important to limit the scattering of neutrons in the air and second, to

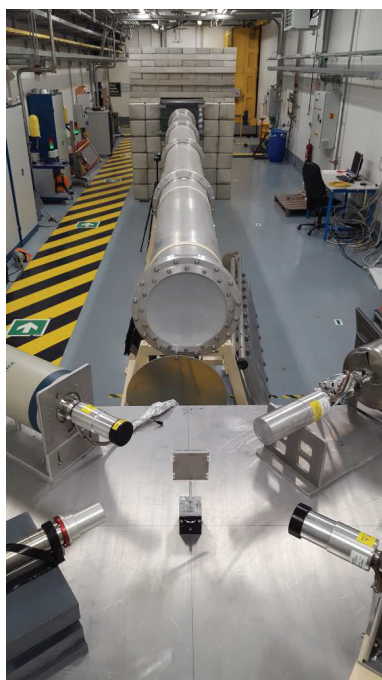
reduce the size of the beam at long flight path without increasing neutron and  $\gamma$  backgrounds. Without second collimation system, the neutron beam diameter increases up to almost 20 cm at 30 m from the neutron source. MCNPX simulations have been conducted in our team to design the size, the composition and the position of the second collimator in the flight path area. Then, the shielding of the second collimator has been studied too, in order to obtain a "clean" area at 30 m from the neutron source. As inputs for simulations, we have used the neutron beam produced by the interaction of a deuteron beam at 40 MeV on a thick Beryllium target. The maximum achievable intensity ( $50 \mu\text{A}$ ) was considered. The entire NFS hall (ground, walls), including the first collimator was entered in the simulations and the final objective was to obtain a neutron beam diameter around 6 cm at 28.5 m from the neutron production source. To satisfy this requirement, the second collimator, made in iron, has a length of 96 cm and an external diameter of 15 cm. The inner hole has a conical shape and inlet and outlet diameters of 3.4 cm and 4.2 cm respectively to minimize the halo effect. The shielding of the collimator has been designed to absorb the most of the secondary particles created by interactions of neutron on the collimator. Different shells of slowing down and absorbing materials have been assembled as shown in Figure 5. The total weight of the collimator and shielding is around 28 t distributed in  $\approx 23$  t of concrete, 4 t of lead, 800 kg of PVC, 500 kg of borated polyethylen (BPE) and 130 kg of iron. Another constraint that we considered in the mechanical design was the ability to remove the collimator from the shielding to allow the neutron beam to pass trough without interacting. This ensures that no additional background is induced for experiment placed at short flight path upstream of the shielding. A view of this apparatus and the neutron beam line is presented in Figure 6.



**Figure 5.** (color online) Schematic view of the TOF hall of NFS with the neutron beam line and the second collimator and its shielding. A cross section of the shielding shows the different layers used to catch the secondary particles emitted by the interaction of neutron with iron from collimator.

### 3.2 Preliminary results of tests

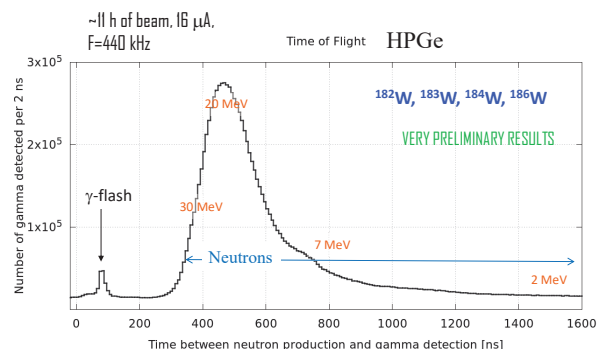
In fall 2021, our team made a first test of the use of the prompt  $\gamma$ -ray spectroscopy at the long flight path at SPIRAL2/NFS. The main goals were to verify that first, the neutron beam was collimated as expected at 30 m and second, the background conditions are favorable to use HPGe detectors. The experimental conditions for this test were the following. Neutrons were produced with the interaction of 40 MeV deuterons on a thick Beryllium target (8 mm) with an intensity of 16  $\mu$ A. As shown on Figure 6, three HPGe detectors, coupled to the FASTER acquisition system, were positioned on a table at 125° with respect to the neutron beam direction. A natural W sample (already used for measurements with GRAPhEME at GELINA) was placed in the beam. Unfortunately, some problems from the facility occurred during the test and only a dozen hours of beam time were available. Nevertheless, this was sufficient to verify several points. First, a photograph of the beam has been made at the position of the sample and it revealed that the second collimator was well aligned and the diameter of the neutron beam is as expected (around 5.5 cm at 28.9 m). The study of the background did not reveal any important contributions that could interfere with the measurement. Especially, no particle, coming from the beam dump placed just around 5 meters downstream the W sample position, has been observed.



**Figure 6.** (color online) Picture of the GRAPhEME setup installed at NFS during the first test in 2021.

The time-of-flight spectrum (Figure 7) shows well defined structures with a small  $\gamma$  flash at short times, followed by a bump corresponding to  $\gamma$  events induced by neutron reactions on tungsten. In the  $\gamma$ -energy spectra, several  $\gamma$  peaks have been identified which correspond to  $\gamma$  transitions in the different W isotopes produced by ( $n$ ,

$n'$ ), ( $n, 2n$ ), ( $n, 3n$ ) and ( $n, 4n$ ) reaction as shown in Figure 8.



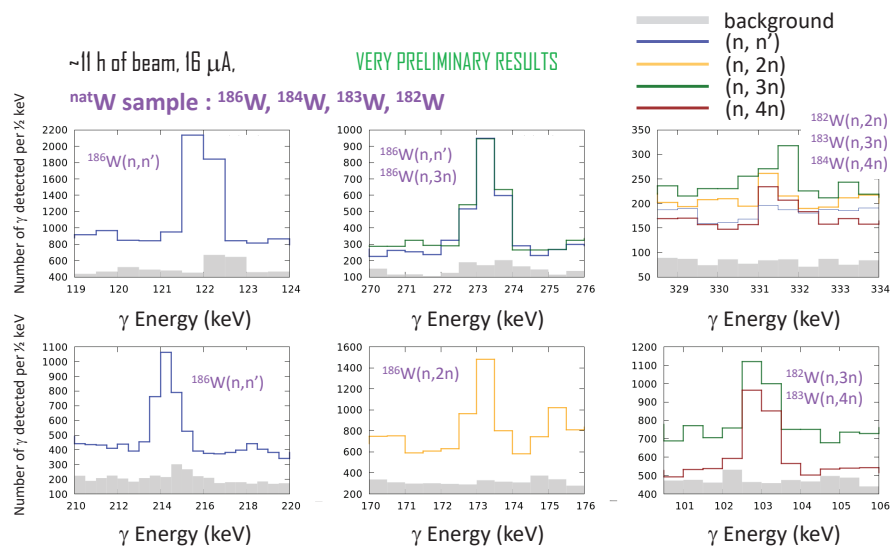
**Figure 7.** (color online) Time-of-flight spectrum obtained with one Germanium detector and natural W sample during the test in 2021.

With this first test, we were able to obtain qualitative information on the experimental conditions achieved at SPIRAL2/NFS in view of the use of the prompt  $\gamma$ -ray spectroscopy at long flight path. First conclusion is that this kind of measurement technique can be used under good conditions of neutron and  $\gamma$  backgrounds. A second test is planned during fall 2022 to continue the investigations and characterization of the experimental area. The use of a fission chamber with  $^{238}\text{U}$  deposit is planned to obtain the neutron flux measurement. All these quantitative information will be useful to prepare an experiment proposal which will be submitted to the GANIL PAC during fall 2022. In this proposal, we aim to ask beam for measurement campaigns on  $^{238}\text{U}$  and  $^{232}\text{Th}$ .

### 4 How to overcome weaknesses of the prompt $\gamma$ -ray spectroscopy method?

As already discussed, using the prompt  $\gamma$ -ray spectroscopy to infer ( $n, xn$ ) reaction cross section requires the knowledge of the nuclear structure (branching ratio and conversion electron coefficient) of the nucleus of interest [13]. In the case of actinides, we have shown [15] that this knowledge is not at the expected level to produce accurate ( $n, xn$ ) cross section from ( $n, xn\gamma$ ) ones. Moreover, in such nuclei the structure level is rather compact even at low energy. The planar germanium detectors of GRAPhEME are very sensitive to low energy  $\gamma$  transitions but its efficiency rapidly decreases for  $\gamma$ 's above 1 MeV. Thus we are not able to measure accurately  $\gamma$  transitions for energy above  $\approx 1.5$  MeV. A consequence is that the inferred ( $n, n'$ ) cross section is exact for neutron energy from reaction threshold to  $\approx 1.5$  MeV and a lower limit above this limit. A last penalizing point is that, in even-even actinides, the first excited level is at low energy (typically around 50 keV) and its deexcitation proceeds mainly by conversion electrons which are not detected by GRAPhEME. However, this transition collects more than 90% of the  $\gamma$  cascade. With GRAPhEME, for  $^{238}\text{U}$  and  $^{232}\text{Th}$ , we were able to determine the ( $n, xn\gamma$ ) cross section of the deexcitation of





**Figure 8.** (color online) Portions of  $\gamma$ -energy spectra gated on neutron-energy windows corresponding to  $(n, n')$  (blue),  $(n, 2n)$  (yellow),  $(n, 3n)$  (green) and  $(n, 4n)$  (red) process. Peaks relative to  $\gamma$  transitions in the different W isotopes are visible.

the first  $2^+$  level but with large uncertainty that thus dominates the inferred total inelastic cross section uncertainty. To overcome these weaknesses, our collaboration has developed several actions which are illustrated in Figure 9 and summarized in the following.

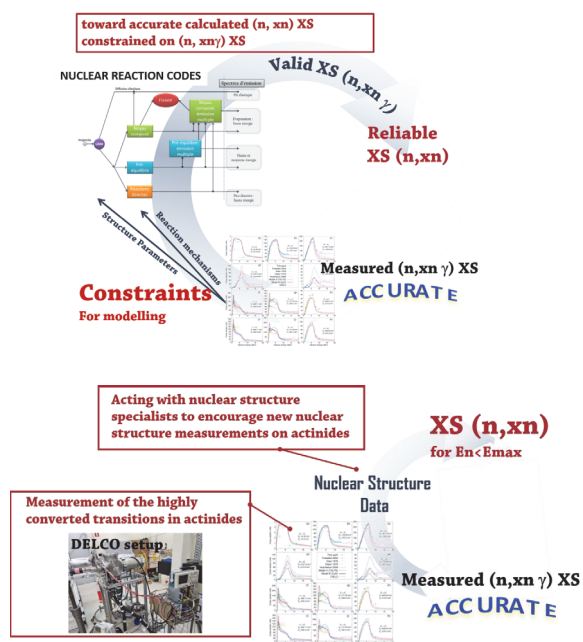
- In the frame of the close collaboration with theoreticians from CEA/DAM/DIF, LANL and IAEA, we work on the improvement of modeling of  $(n, n')$  and  $(n, xn)$  cross section reactions [28] based on the constraints given by the accurate  $(n, xn\gamma)$  measurements performed with GRAPhEME. For example, the progress made in the case of  $^{238}\text{U}$  described in [8] and reported in section 2, is a first and encouraging step toward more predictive models. Another goal is also to include in TALYS calculations, uncertainties estimation to facilitate and make more relevant the comparison between experimental and calculated data.
- As explained before, missing the major part of the deexcitation of the first excited level in even-even actinides is very penalizing in terms of final uncertainty on inferred total inelastic cross section. Our collaboration is thus working on a new instrument, named DELCO (Detection of ELectrons from CONversion) [29], which measures electrons. After several tests of different kinds of Silicon detectors, DELCO is now based on a cooled Si detector (surface and thickness are  $300\text{ mm}^2$  and  $5000\text{ }\mu\text{m}$  respectively). The last tests performed at GELINA, with a  $^{238}\text{U}$  sample from which the Si detector observed the radioactivity, showed that the achieved resolution is  $0.8\text{ keV}$  at  $10\text{ keV}$  and  $1\text{ keV}$  at  $100\text{ keV}$ . This good performance will allow the separation of conversion electrons of interest coming from neutron inelastic scattering. The next step of this instrumental project will be the irradiation of the  $^{238}\text{U}$  sample with neutrons from GELINA in view of validation of the detection concept.

- Finally, as explained in this contribution and more detailed in the contribution [15] in this proceeding, another weakness of the prompt  $\gamma$ -ray spectroscopy method is that it relies on nuclear structure data knowledge which has revealed to be not enough accurate for actinides. We have thus initiated an information action to the community of nuclear structure physicists to explain our needs about new accurate nuclear structure information for nuclei of interest in the context of nuclear energy applications. A first result was an invitation to collaborate to the  $\nu$ -ball2 experiment [30] performed in 2022 at the ALTO facility with the LICORNE neutron source [31]. By the coupling of the  $\nu$ -ball  $\gamma$ -ray spectrometer with the high flux neutron source LICORNE (neutron energy is around  $2\text{ MeV}$ ) this experiment allows a new measurement of the  $^{238}\text{U}$  nuclear structure. The data analysis is in progress but we hope to enrich the nuclear level scheme and precise some branching ratios up to excitation energy around  $2\text{ MeV}$ .

## 5 Conclusion

After almost twenty years of developments and measurements performed at EC-JRC-Geel, GRAPhEME is becoming a federative instrument to study, in a comprehensive way from experiments to modeling, the neutron inelastic scattering with the prompt  $\gamma$ -ray spectroscopy method. Numerous cross section data mostly on actinides but also on some intermediate mass nuclei have been measured. Beyond GRAPhEME, our collaboration is also working on several projects that can overcome the weaknesses of the prompt  $\gamma$ -ray spectroscopy method both from theoretical and instrumental points of view. And finally, a new horizon is emerging for GRAPhEME with the possibility of measuring  $(n, 2n)$  and  $(n, 3n)$  reaction cross sections at the new SPIRAL2/NFS facility.





**Figure 9.** (color online) Illustration of the three actions engaged by our collaboration to overcome weaknesses of the prompt  $\gamma$ -ray spectroscopy method used with actinides.

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