# Prompt fission neutron emission from <sup>235</sup>U(n,f): thermal and resonance region

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#### Abstract

For nuclear modelling and improved evaluation of nuclear data, knowledge of fluctuations of the prompt neutron multiplicity as a function of incident neutron energy is requested for the major actinides <sup>235</sup>U and <sup>239</sup>Pu. Experimental investigations of the prompt fission neutron emission in resonance-neutron induced fission on <sup>235</sup>U are taking place at the GELINA facility of the IRMM. The experiment employs an array of scintillation detectors (SCINTIA) in conjunction with a newly designed 3D position-sensitive twin Frisch-grid ionization chamber.

In addition, the mass-dependent prompt neutron multiplicity, v(A), has attracted particular attention. Recent, sophisticated nuclear fission models predict that the additional excitation energy, brought into the fission system at higher incident neutron energies, leads to an increased neutron multiplicity only for heavy fragments, as observed in the  $^{237}$ Np(n,f) reaction. A first feasibility study has been performed at the JRC-IRMM VdG accelerator to measure v(A) for  $^{235}$ U(n,f).

#### 1 Introduction

The energy spectrum as well as the multiplicity of prompt neutrons emitted in nuclear fission plays an important role in many applications. In particular, accurate predictions of nuclear criticality using neutron transport codes are dependent on the underlying nuclear data, especially the prompt neutron multiplicity and the prompt fission neutron spectrum. Both observables have received recently a lot of attention. On the one hand for the prompt fission neutron spectrum an IAEA Coordinated Research Project has been launched, and the final report is being published [1]. On the other hand recent OECD-Nuclear Energy Agency (NEA), Working Party of Evaluation Cooperation (WPEC) documents [2, 3] state the lack of inclusion of prompt neutron multiplicity fluctuations for the major isotopes <sup>235</sup>U and <sup>239</sup>Pu, in the resonance region relevant for fission cross section evaluations. In Ref. [2] it has been clearly stated that both reliable measurements and modelling of the shape of the prompt neutron multiplicity (nubar) in the resonances are still lacking. In addition, at higher incident neutron energy, recent theoretical modelling [4, 5] has confirmed the increase of prompt neutron multiplicity mainly for heavy fragment masses, if the incident neutron energy is increased. This is so far not considered in the correction for the energy dependence of prompt neutron multiplicity as a function of mass (see e.g. Fig. 8 in Ref. [6]). For pre-neutron mass determination from 2E experiments this may have important consequences in the post-neutron yield distributions [7].

The nuclear data facilities at the JRC-IRMM are predestined to tackle the above mentioned problems. With the white neutron source GELINA and using the neutron time-of-flight technique the resonance region is accessible. The Van de Graaff (VdG) accelerator gives high neutron flux at

incident neutron energies in the MeV region to study the change of the prompt neutron multiplicity as a function of fragment mass.

The experimental technique used to study prompt neutron emission both in resonance-neutron induced fission at the GELINA facility and at higher incident neutron energy at the VdG accelerator is based on techniques pioneered by Bowman et al. [8]. The method involves extracting fission fragment masses by measuring either velocity (2v) or energy (2E) of the two fragments, and coincident measurements of fission neutron time-of-flight at known angles with respect to the fission axis. In an experiment on prompt fission neutrons in <sup>252</sup>Cf(SF) Budtz-Jørgensen and Knitter exploited the combination of a twin Frisch-grid ionization chamber for fission fragment properties (2E technique) and a liquid scintillator for neutron detection [9]. Their result serves as the basis for our experimental setup for studying prompt neutron emission. The ionization chamber has a large solid angle, which not only facilitates the fragment-neutron coincidence rate, but also introduces a less biased selection of coincident events. The ionization chamber allows determining the fission fragment emission angle relative to the chamber axis. By placing the neutron detector along the chamber axis this angle coincides with the angle relative to the momentum of detected neutrons.

In the following sections the different experiments will be highlighted and first results discussed.

# 2 **Prompt fission neutron multiplicity**

## 2.1 Introduction

The total prompt fission-neutron multiplicity is an important quantity for nuclear applications and needs to be known to very high accuracy in the 0.25% range [1, 10] for major actinides. For the analysis of fission yield data also the information about the prompt fission neutron multiplicity as a function of fission fragment mass, v(A), is needed. In recent years the focus was put towards the behaviour of this quantity v(A) as a function of incident neutron energy. Improved model calculations predict that the additional excitation energy, brought into the fission system at higher incident neutron energies, leads to an increased neutron multiplicity only for heavy fragments [4, 5], like observed in the <sup>237</sup>Np(n,f) reaction [11]. In Ref. [7] an analysis of the impact of the prompt fission neutron multiplicity correction on the fission fragment yield of <sup>234</sup>U(n,f) was performed. The result showed an effect in the order of up to 30% on the most abundant fission fragments whether the standard correction (e.g. as done in Ref. [6]) with an upscale of the neutron multiplicity as a function of mass or a solely higher heavy fragment neutron multiplicity was used.

An experiment campaign has been started to verify the theoretical predictions in collaboration with Uppsala University within the frame of the EUFRAT project [12].

# 2.2 Experiment

The data presented in this paper were measured at the 7 MV Van de Graaff accelerator of the IRMM in Geel, Belgium. Neutrons of 0.5 MeV were produced via the reaction <sup>7</sup>Li(p,n). A twin Frisch-Grid ionization chamber (FGIC) was used to detect the fission fragments. The chamber was operated with P-10 (90% Ar and 10%CH<sub>4</sub>) as counting gas, and the gas pressure was set to  $1.05 \times 10^5$  Pa with a gas flow of 0.1 l/min. A thin, transparent <sup>235</sup>U sample was mounted at the common cathode. The prompt fission neutrons from the <sup>235</sup>U(n,f) reaction were measured with two NE213 equivalent scintillation detectors. The experimental setup is shown in Fig. 1. The beam was thermalized with the help of a thick layer of paraffin. The thermal neutron induced fission of <sup>235</sup>U was studied in this experiment. In future experiments the energy range will be extended towards higher incident neutron energies.

MCNP and FLUKA simulations have been performed to see the influence of the direct beam and the moderated beam (using paraffin of 7.5 cm thickness) on the neutron detectors. Using a direct beam of only 0.5 MeV neutron energy helped in this experiment a lot as the sensitivity of the used scintillation neutron detectors is very low at this neutron energy. In addition the neutron detectors were shielded towards the direct beam by an additional 10 cm of paraffin, 3 cm of lead and 5 mm of copper.

The data from the ionisation chamber and the neutron detectors were acquired using waveform digitizers of 250 MHz and 12 bit pulse height resolution. Data analysis was done using the ROOT software package [13] and digital signal processing routines developed at JRC-IRMM.



Fig. 1: Upstream view of the experimental setup for the neutron multiplicity measurement at the VdG. The FGIC is seen in the centre with the two NE213 on either side. The shielding arrangement is also visible.

### 2.3 Preliminary results

The analysis of the fission fragment signals taken without coincidences with the neutron detectors can be used as quality check of the set-up. In Fig. 2a the pulse height distributions for forward (red) and backward (blue) emission of the fragments are shown. Both distributions are in very good agreement showing that all the corrections needed to be applied to the raw signals are well under control. Fig. 2b shows the cosine of the angular distribution of the fission fragments again for forward (red) and backward (blue) direction of the fragment emission. Also the cosine of the angle from both emission directions of the fragments is in good agreement.

Fig. 3 shows the pulse shape information from the neutron scintillation detectors versus the time-of-flight information. Despite the large  $\gamma$ -ray background the neutrons of interest could be identified in the encircled region. Further analysis is still in progress.



**Fig. 2:** a) Pulse height distributions for forward (red) and backward (blue) emission of the fragments. b) Cosine of the emission angle for forward (red) and backward (blue) emission of the fragments.



Fig. 3: Pulse shape information versus time-of-flight of the neutron detector signals. Among a large  $\gamma$ -ray background the neutrons of interest are found in the encircled area.

# **3** Neutron-fission fragment correlations in the resolved resonance region

### 3.1 Introduction

For nuclear modelling and improved evaluation of nuclear data the knowledge about fluctuations in the prompt neutron multiplicity as a function of incident neutron energy is requested for the major actinides <sup>235</sup>U and <sup>239</sup>Pu [2, 3]. Fluctuations in fission fragment mass and total kinetic energy (TKE) in both isotopes have been observed in resonance neutron induced fission [14, 15]. Independently, fluctuations in the number of emitted neutrons have also been observed [16]. In view of the fact that both neutron number and fission fragment properties have been found to vary it is necessary to study the correlations of prompt neutron multiplicity and fission fragments properties in the resonance region [17]. Furthermore, the knowledge of the prompt neutron multiplicity as a function of mass and TKE is needed when determining post-neutron emission fission fragment mass distributions experimentally via the double kinetic energy or double velocity techniques. Experimental investigations of correlations between the prompt fission neutron multiplicity with fragment properties

in resonance-neutron induced fission on <sup>235</sup>U and <sup>239</sup>Pu are taking place at the GELINA facility of the JRC-IRMM.

#### 3.2 Experiment

The experimental setup for investigating correlations of prompt neutrons with fission fragments in resonance neutron induced fission on <sup>235</sup>U and <sup>239</sup>Pu is illustrated in Fig. 4. The fission target is placed inside the ionization chamber, about 9.2 m away from the neutron production target of the GELINA facility. An array of neutron detectors (SCINTIA) is employed in order to achieve a reasonable fission-neutron coincidence count rate. The SCINTIA array consists of 7 NE213 equivalent liquid scintillators (Scionix LS-301) and 5 para-therphenyl crystalline scintillation detectors.





Fig. 4: Schematic drawing (left) and photograph (right) of the experimental arrangement at the GELINA facility.



**Fig. 5**: Time-of-flight spectrum with resonance of <sup>235</sup>U(n,f).

When employing an array of neutron detectors, each detector forms an axis of symmetry around which the orientation of the fission axis needs to be known. Hence, the traditional ionization chamber is no longer sufficient to reconstruct the kinematics in the fragment rest frame. In order to solve this problem, the ionization chambers anode plate is replaced by a position sensitive readout structure.

This allows determination of all three space components of the fission fragments direction of travel. A 22 channel, fully digital data acquisition is used.

The acquisition is triggered by the current signal from the ionization chamber cathode, giving the instant of a fission event in time with a resolution better than 1 ns FWHM. For each fission event the digital waveforms of all channels, sampled at 400 MS/s with 14-bit resolution, are stored on disk for off-line treatment together with the incident neutron time-of-flight for each event. In Fig. 5 a time-of-flight spectrum of the on-going  $^{235}$ U(n,f) experiment is shown.

#### 3.3 Preliminary results

The top panel of Fig. 6 shows the average total kinetic energy (TKE) as a function of incident neutron



**Fig. 6** Top panel: average total kinetic energy (TKE) as a function of incident neutron energy in the range up to 45 eV. Lower panel: ratio of the neutron multiplicity for a given resonance or resonance group relative to the mean neutron multiplicity. The red points highlight strong resonances in the fission cross section of <sup>235</sup>U. The dashed line refers to the thermal value.

energy in the range up to 45 eV. Each point corresponds to either a single resonance or resonance group. The red points highlight the strongest resonances in the fission cross section of <sup>235</sup>U. A clear fluctuation exceeding the experimental uncertainty is observed. This was already pointed out in Ref. [14]. However, the statistical significance has very much improved in the present work leading to a clearer picture of the energy dependent changes compared to the earlier work.

In the lower panel of Fig. 6 the ratio of the neutron multiplicity for a given resonance or resonance group relative to the mean neutron multiplicity is shown. Again, the strongest resonances are highlighted in red. At the current moment of data collection, an anti-correlation, i.e., high TKE corresponds to lower neutron multiplicity, is observed when all points in the given incident neutron energy interval are plotted. This is expected from energy conservation. Of course, also possible changes in the mass distribution need to be accounted for. Nevertheless, there seem to be no sizable fluctuations in the neutron multiplicity in the strong resonances.

### Conclusions

In conclusion, new experiments have been started to measure prompt neutron multiplicity in neutroninduced fission of <sup>235</sup>U. Presently both the thermal and the resonance region are covered. Measurements at higher incident neutron energies are planned for the near future. We are confident that the new data will shed light in the understanding of the observed neutron multiplicity fluctuations in the resonance region and the behaviour of the neutron multiplicity as a function of mass and incident neutron energy.

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