# New prompt fission gamma-ray spectral data from $^{239}$Pu($n_{th}$, f) in response to a high priority request from OECD Nuclear Energy Agency

Angélique Gatera[1,7], Tamás Belgya[3], Wouter Geerts[1], Alf Göök[1], Franz-Josef Hambsch[1],Matthieu Lebois[4], Boglárka Maróti[3], Stephan Oberstedt[1,a], Andreas Oberstedt[2], Frederik Postelt[5], Liqiang Qi[4], Laszló Szentmiklósi[3], Marzio Vidali[1], and Fabio Zeiser[6]

[1] European Commission, Joint-Research Centre, Directorate G, Geel, Belgium
[2] Extreme Light Infrastructure - Nuclear Physics (ELI-NP) / Horia Hulubei National Institute for Physics and Nuclear Engineering (IFIN-HH), Bucharest-Magurele, Romania
[3] Centre for Energy Research, Hungarian Academy of Sciences, Nuclear Analysis and Radiography Department, Budapest, Hungary
[4] Institut de Physique Nucléaire Orsay (IPN-Orsay), Orsay, France
[5] Hamburg University, Department of Physics, Hamburg, Germany
[6] University of Oslo, Department of Physics, Oslo, Norway
[7] Ghent University, Department of Physics, Gent, Belgium

**Abstract.** Benchmark reactor calculations have revealed an underestimation of $\gamma$-heat following fission of up to 28%. To improve the modelling of new nuclear reactors, the OECD/NEA initiated a nuclear data High Priority Request List (HPRL) entry for the major isotopes ($^{235}$U, $^{239}$Pu). In response to that HPRL entry, we executed a dedicated measurement program on prompt fission $\gamma$-rays employing state-of-the-art lanthanum bromide ($LaBr_3$) detectors with superior timing and good energy resolution. Our new results from $^{252}$Cf(sf), $^{235}$U($n_{th}$,f) and $^{241}$Pu($n_{th}$,f) provide prompt fission $\gamma$-ray spectra characteristics : average number of photons per fission, average total energy per fission and mean photon energy; all within 2% of uncertainty. We present preliminary results on $^{239}$Pu($n_{th}$,f), recently measured at the Budapest Neutron Centre and supported by the CHANDA Trans-national Access Activity, as well as discussing our different published results in comparison to the historical data and what it says about the discrepancy observed in the benchmark calculations.

## 1. Introduction

Present knowledge on the released heat during fission states that around 10% of all energy released comes from $\gamma$ rays, and about 40% of these $\gamma$ rays are prompt [1]. Since currently evaluated data from the 1970s show a deviation from benchmark calculations of up to 28%, an urgent request for new prompt fission $\gamma$-ray spectra (PFGS) measurements was put high on the OECD/NEA's HPRL targeting at an uncertainty of 7.5% [2].

Prompt fission $\gamma$ rays are defined as all the $\gamma$ rays emitted prior to $\beta$-decay. In practice, most of the $\gamma$ rays are emitted within a few nanoseconds after fission. Time-of-flight techniques allow discriminating prompt fission $\gamma$ rays from secondary gammas due to prompt neutron inelastic scattering. To achieve this discrimination, we need detectors with sub-nanosecond timing resolution.

$LaBr_3$ detectors have a timing resolution of about 300 ps [3], which makes them very suited for the time-of-flight technique. They also have a better energy resolution than most widely used sodium iodide (NaI) or barium fluoride ($BaF_2$) detectors [4].

[a] e-mail: Stephan.oberstedt@ec.europa.eu

## 2. Experimental methods

Our experiment took place at the 10 MW research reactor of the Budapest neutron centre. We used a cold neutron beam whose flux was $5 \times 10^7$ neutrons/cm$^2$/s. The target had a layer of $^{239}$Pu (99.97% purity) weighting $(429.3 \pm 1.8)$ $\mu$g. The resulting fission rate was about 40.000 fissions/s and the $\alpha$-activity of the source was $(986 \pm 4)$ kBq.

The target was placed on the cathode of a Frisch Grid Ionisation Chamber (FGIC), full of Ar-10%CF4. Around the chamber (Fig. 1), 4 $LaBr_3$ detectors (5.08 cm $\times$ 5.08 cm were placed at a distance of 40 cm. The cathode signal was used as fission trigger and all $\gamma$-rays emitted in a 300 ns-wide time window after fission were digitized (14 Bit, 400 MSamples/s) and saved for later analysis.

## 3. Data analysis

To extract the average $\gamma$-ray multiplicity, mean and total energies per fission, we have to process both the fission and the $\gamma$-ray information. From fission fragment traces, we get the exact number of usable fissions and their timing. The cathode gives the fission trigger and the fission timing. The anode and grid signals are used to filter $\alpha$-particles and pile-ups in this case, or to analyse fission fragment masses

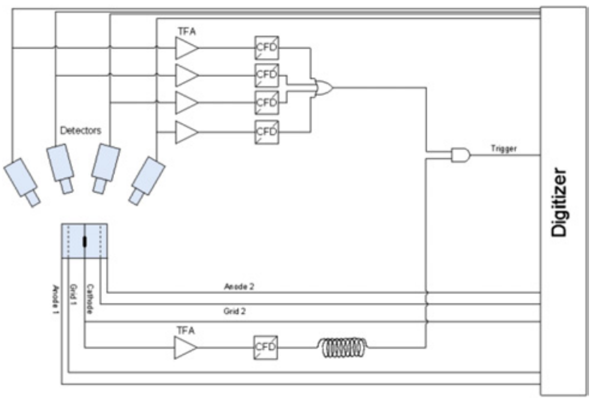


**Figure 1.** Detectors' setup: NIM modules such as Timing Filter Amplifier (TFA) and Constant Fraction Discriminator (CFD) are used to generate the coincidence conditions.

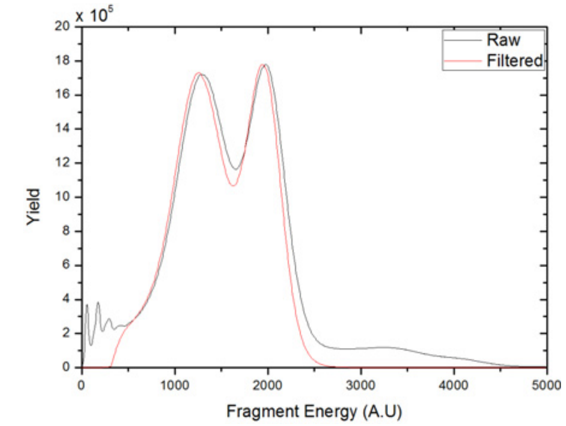


**Figure 2.** Energy as anode pulse height before and after applying the $\alpha-$ and pile-up rejection related cuts. The low energy peaks (below channel 500) in the raw spectrum are alphas and the high-energy bump (above channel 2500) contains the fission-fission pile-up events. Both have been cut out in the filtered spectrum. The filtered spectrum has been normalised to the raw spectrum's height.

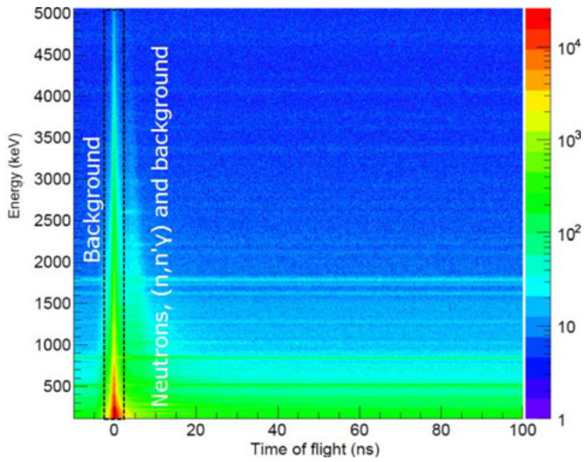


**Figure 3.** Photon time-of-flight as a function of their energies. Prompt $\gamma$-rays are the ones emitted at fission time, we can see them in the bright peak. The cut interval is defined as fission trigger, i.e. $\pm 2.5$ ns wide (black doted box).

and angular distributions. The $\gamma$-ray traces are processed to extract $\gamma$-ray energies and timing.

### 3.1. Fission selection

With a high fission rate and $\alpha-$activity, multiple pile-ups occur. The aim being to calculate the average gamma emissions per fission, we need to have a trustworthy fission count.

For instance, we need to make sure that all the fission triggers result from fission events only, and not $\alpha$-particles and, that we only have one fission in each event. For this we implemented an $\alpha$- and pile-up rejection algorithm, which is based on the cathode's signal length. When there is pile-up, the signal length is significantly increased due to the presence of more than one signal in the acquisition window.

The $\gamma$-rays emissions are only analysed for the filtered events. In our case they amount to $3 \times 10^8$ fission-gamma coincidences.

### 3.2. Prompt $\gamma$-rays selection

The prompt $\gamma$-rays are selected according to their time-of-flight. Thanks to the good timing resolution of our detectors, and a fast counting gas in the FGIC (Ar-10%$CF_4$), we achieve a coincidence timing resolution between 1.1 and 1.2 ns for the 4 detectors. A $\pm 2.5$ ns wide interval is defined as fission trigger (black doted box in Fig. 3) from which prompt fission $\gamma$-ray spectra were constructed. This allows a good separation of prompt $\gamma$ rays from prompt neutrons, and the $\gamma$ rays generated by inelastic scattering.

The background acquired before the fission trigger (see Fig. 3) is subtracted from the prompt window. The background-subtracted spectrum obtained is the measured prompt fission $\gamma$-ray (PFG) spectrum.

### 3.3. Spectrum unfolding

Simulations of our detectors' response functions were done for 200 energies between 80 keV and 12 MeV in variable steps correlated to the energy resolution [4,5]. A weighted sum of these simulated spectra is used to fit the measured spectrum (see Fig. 4) and hence to obtain an emission spectrum. The unfolded emitted spectrum is then used to calculate average $\gamma$-multiplicities, total and mean energies.

## 4. Method validation

### 4.1. $^{252}$Cf (sf)

A new measurement of the spontaneous fission of $^{252}$Cf was made to validate the experimental setup and data analysis. Our results (see Fig. 5) have a good overall agreement with published data. We can also see that we achieve a better definition of the spectrum at low energies.

This validation is a proof that our method works reliably and that we can confidently apply it to other fissioning systems.

### 4.2. Recent achievements

In recent years, the same protocol was used in our group for the measurements of $^{252}$Cf(sf), $^{235}$U($n_{th}$, f), and $^{241}$Pu($n_{th}$, f).

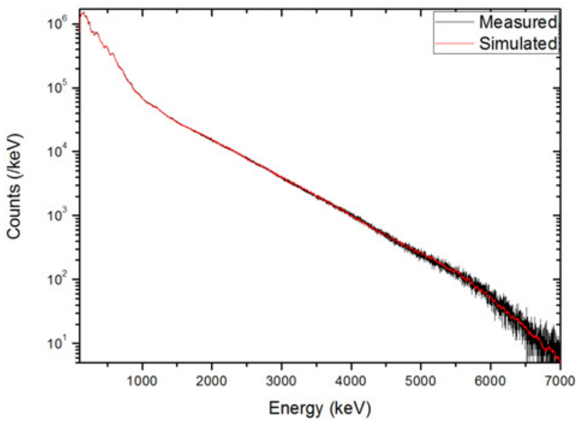


**Figure 4.** Measured spectrum fitted by a weighted sum of simulated spectra.

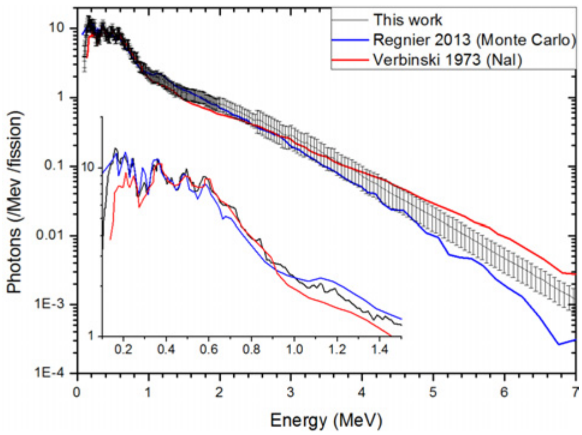


**Figure 5.** PFG spectra for the spontaneous fission of $^{252}$Cf. The black line is our data with error bars, blue line is Monte Carlo simulation from CEA [6] and the red line is reference data from Verbinski measured with NaI detectors in 1973 [7]. The inset shows a zoom on the low energy part exhibiting the low-energy threshold and a very good agreement with model calculations.

The results have been published and only a slight increase in average PFG energy of 5.2% and 3.6% for $^{235}$U(n$_{th}$, f) and $^{241}$Pu(n$_{th}$, f), respectively [8] was found.

## 5. Preliminary results for $^{239}$ Pu(n$_{th}$, f)

The data analysis for $^{239}$Pu(n$_{th}$, f) is still to be finalized. A few problems had to be investigated and the fixes are being implemented.

Our data agrees well with the other sets of data in Fig. 7 above $\sim$ 300 keV, but we exhibit a higher yield at low energies. After thorough investigation we found out that most of this yield is due to an underestimation of the low-energy scattered $\gamma$ rays in our setup's surroundings. We made major changes to our setup to accommodate a parallel neutron capture experiment and it increased consistently scattering into the low energy.

The fact that the low-energy scattering for high-energy $\gamma$ rays is not correctly unfolded, leads to an overestimation of the emitted spectrum at low energies. This means that all the response function simulations have to be redone implementing a more realistic geometry as close as possible to the experimental setup to finally getting rid of the overestimation at low energies.

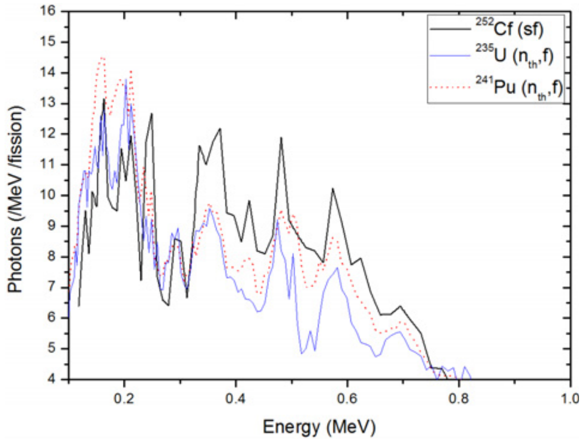


**Figure 6.** Low energy structures in the spectra from $^{252}$Cf(sf) [9], $^{235}$U(n$_{th}$, f) [10] and $^{241}$Pu(n$_{th}$, f) [11].

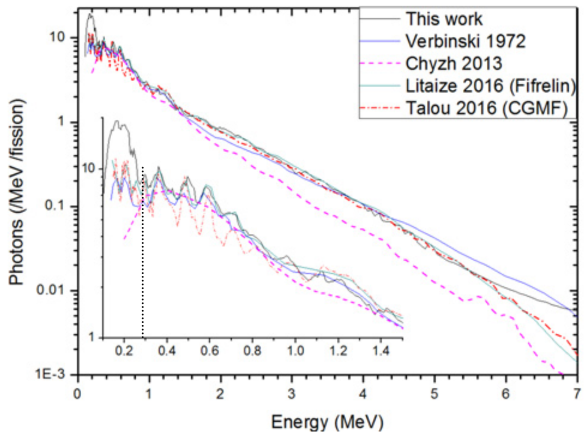


**Figure 7.** Our preliminary PFG spectrum for $^{239}$Pu(n$_{th}$, f) compared to historical data from Verbinski [7], Chyzh [12] and Monte Carlo calculations from Talou [13] and Litaize [14].

**Table 1.** Average values for the different set of data in Fig. 7 and evaluated data from ENDF/B-VII.1. $M_\gamma$ is the average $\gamma$-rays multiplicity per fission, $\varepsilon_\gamma$ is the average energy per photon and $E_{\gamma,\mathrm{tot}}$,tot is the average total $\gamma$-energy released per fission.

| Results | Detector | $M_\gamma$ (/ fission) | $\varepsilon_\gamma$ (MeV) | $E_{\gamma,tot}$ (MeV) | Energy range (MeV) |
|---|---|---|---|---|---|
| This work (extrapolated) | LaBr$_3$:Ce | 8,11 ± 0.41 | 0.87 ± 0.07 | 7.04 ± 0.37 | 0.1–7.0 |
| This work (extrapolated) | LaBr$_3$:Ce | 7.83 ± 0.40 | 0.89 ± 0.07 | 7.01 ± 0.37 | 0.14–7.0 |
| Verbinski | NaI:Tl | 7.23 | 0.94 ± 0.05 | 6.81 | 0.14–10.0 |
| Litaize | Calculation | 7.70 | 0.92 | 7.10 | 0.14–10.0 |
| Talou | Calculation | 7.22 | 0.91 | 6.55 | 0.14–10.0 |
| ENDF/B-VII.1 | Evaluation | 7.78 | 0.87 | 6.73 | 0.05–8.0 |

Since this iteration and sub-sequent re-doing of the response simulations fro the 200$\gamma$ energies will take time, we made, in the meantime, a first estimation on what our final results would be once the scattering issue corrected. To do so we calculated an extrapolation to low energies of our data based on Verbinski's data [7] and calculations from Ref. [14]. We calculated ratios in several energy windows between 300 and 1000 keV that were extrapolated to energies below 300 keV and then applied to our data. From that we were able to estimate the energy excess we measure compared to historical data.

Our estimate forecast an increase in average PFG energy below 5% with respect to evaluated data (see values in Table 1).

## 6. Conclusion and outlook

Presently, we are re-doing the response function simulations in order to finalize the results on $^{239}$Pu($n_{th}$, f). However, we have good reasons to believe already that the energy excess with respect to evaluated data will be of limited impact, as this was the case for the other measured isotopes.

Even though we achieved a higher precision in PFG data compared to historical data, we have found out that the small increase in average PFG energy we measured cannot explain the heat under-prediction from nuclear data in comparison to benchmark calculations.

It seems that the lack in nuclear data is most likely not from thermal neutron induced fission data, so other types of fission reactions have to be investigated. Our collaborators from IPN Orsay started to investigate fast neutron induced fission using the neutron source LICORNE [15].

Other possible sources of heat, that have not been studied yet, include photo-fission induced by high-energy capture $\gamma$-rays. This needs further consideration.

The authors are indebted to the CHANDA project for funding the experiments in Budapest. Special thanks go to O. Litaize and P. Talou for valuable discussions and their interest in this work. The European Commission is gratefully acknowledged for providing Ph.D. fellowship program support to A. Gatera.

## References

[1] K.S. Krane, Introductory Nuclear Physics, 489–493 (1988)
[2] http://www.oecd-nea.org/dbdata/hprl/hprlview.pl?ID=421 and references therein
[3] A. Oberstedt et al., Nucl. Instr. and Meth. A **708**, 7 (2013)
[4] http://www.oecd-nea.org/tools/abstract/detail/nea-1525
[5] S. Agostinelli et al., Nucl. Instr. and Meth. A **506**, 250 (2003)
[6] D. Regnier et al., Phys. Procedia **47**, 47 (2013)
[7] V.V. Verbinski et al., Phys. Rev. C **7**, 1173 (1973)
[8] S. Oberstedt et al., Phys. Procedia **64**, 83 (2016)
[9] R. Billnert et al., Phys. Rev. C **87**, 024601 (2013)
[10] A. Oberstedt et al., Phys. Rev. C **87**, 051602 (2013)
[11] S. Oberstedt et al., Phys. Rev. C **90**, 024618 (2014)
[12] A. Chyzh et al., Phys. Rev. C **87**, 034620 (2013)
[13] P. Talou et al., Phys. Rev. C (2016 to be published)
[14] O. Litaize, O. Serot, private communication (2016)
[15] M. Lebois et al., Phys. Rev. C **92**, 034618 (2015)