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Prompt γ -ray characteristics from ²³⁵U(n, f) at $\overline{E}_n = 1.7$ MeV

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Abstract In response to nuclear data requests by OECD NEA, we present here the first results with respect to promptfission γ -ray spectra characteristics for fast neutrons on ²³⁵U. For that we have analyzed previously taken prompt-fission γ -ray data from the reaction ²³⁵U(n, f) induced by fast neutrons of average energy $\overline{E}_n = 1.7$ MeV and spectral properties were deduced. It was expected that the values would be somewhat higher than for thermal-neutron induced fission of the same system, which at a first glance turned out to be not the case. However, taking the different prompt time windows relative to the coincidence timing resolution between both measurements into account, this discrepancy could be resolved. The results are an average multiplicity $\overline{M}_{\gamma} = 7.45$ \pm 0.48 γ rays per fission and an average total γ -ray energy release in fission $\overline{E}_{\gamma,tot} = (6.03 \pm 0.47)$ MeV, which includes an additional relative uncertainty of 4% from the time window correction. The corresponding average energy per γ ray is $\bar{\epsilon}_{\gamma} = (0.81 \pm 0.04)$ MeV.

1 Introduction

For future nuclear applications, such as fast reactors of the Generation IV type, precise knowledge about prompt fission γ -ray emission is important for the assessment of the decay heat released in the reactor core. While this information, based on experimental data, is available to quite some extent for many actinide nuclei and fission processes, this is surprisingly not the case for ²³⁵U, except for thermal-neutron induced fission.

In the very first experiment ever performed at the directional fast-neutron source LICORNE of IPN Orsay [1] some years ago, prompt-fission γ -ray spectra (PFGS) were measured with 14 hexagonal BaF₂ and three coaxial LaBr₃:Ce scintillation detectors, which were calibrated in energy with different sources over an energy range between 121 keV and 4.4 MeV [2]. The γ rays were produced in fission reactions induced by neutrons of average energy $\overline{E}_n = 1.7$ MeV hitting ²³⁵U and ²³⁸U samples of 10 mg approximate mass each, placed back-to-back at the central cathode position inside a cylindrical twin Frisch-grid ionization chamber. Until quite recently, only γ rays detected with the BaF₂ scintillators had been analyzed and first results were already published [2]. As stated there, the low counting statistics did not allow to treat the LaBr₃:Ce spectra back then due to difficulties in unfolding the detector responses. Concerning the BaF2 spectra, the detector responses were properly unfolded after a detailed MCNP5 [3] simulation of the experimental setup and verified by comparison with the measured calibration spectra. However, the thresholds were rather high, approximately 500 keV. Moreover, during this experiment we encountered problems associated with dead time, which might have resulted in uncertainties related to a proper normalization of the γ -ray spectra with respect to the total number of occurred fission events. For the reasons mentioned above, no absolute values for PFGS characteristics (i.e. average multiplicity, average and total γ -ray energy) could have been deduced. Instead, only ratios for ²³⁵U and ²³⁸U spectral properties were given [2]. However, subsequent experiments have provided us now with the means for analyzing also the spectra taken with the LaBr3:Ce detectors and determining the first prompt-fission γ -ray spectral characteristics for fast-neutron induced fission of ²³⁶U*.

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2 Normalization based on ²³⁸U(n, f)

One of the prerequisites mentioned above was the recent measurement of prompt γ rays from the reaction 238 U(n, f) at \overline{E}_n = 1.9 MeV [4]. Taking into account that the average promptfission γ -ray multiplicities for this reaction agree well within 1% for incident neutrons of 1.7 and 1.9 MeV kinetic energy, which may be inferred from the calculations in Ref. [5], the emission spectrum at 1.9 MeV can be used to obtain the correct normalization for the spectrum at 1.7 MeV by adjusting the latter to the first one. This was done for the γ -ray energy region 0.5 MeV $< E_{\gamma} < 6$ MeV to reproduce the same integrated multiplicity, and the result is shown in Fig. 1. There, another emission spectrum is shown that was taken with a bismuth germanate (BGO) detector for incident neutrons of 1.6 MeV kinetic energy [6]. It serves as verification of the assumed similarity of the first two spectra concerning both shape and absolute height.

Since the spectra detected with the BaF₂ cluster for both $n + {}^{235}U$ and $n + {}^{238}U$, respectively, were measured simultaneously, the scaling factor obtained above for $n + {}^{238}U$ may be applied to the PFGS for $n + {}^{235}U$. This in turn allows also to fix the absolute height of the ${}^{235}U(n, f)$ PFGS taken with the LaBr₃:Ce detectors to the one observed with the BaF₂ detectors. Prior to that, the spectra taken with the LaBr₃:Ce detector response. Henceforth we may refer to the spectra taken with the different detectors as "LaBr₃:Ce" and "BaF₂ spectra", respectively.

3 Data analysis and results

As mentioned above, counting statistics was rather low for the LaBr₃:Ce detectors (just below 1500 events), in contrast

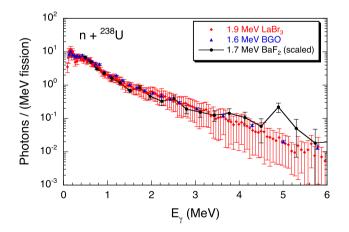


Fig. 1 Comparison of the unfolded prompt-fission γ -ray spectra for n + ²³⁸U at $\overline{E}_n = 1.9$ MeV [4] and 1.7 MeV [2], of which the latter was adjusted to the first one (see text for details). In addition, a spectrum is shown that was taken with a BGO detector at $\overline{E}_n = 1.6$ MeV for the same fissioning system [6]

to the spectra obtained with the massive BaF_2 clusters (about 70,000 events). For that reason a reliable unfolding of the detector response according to

$$\begin{pmatrix} a_1 \\ a_2 \\ \vdots \\ a_m \end{pmatrix} = \begin{pmatrix} b_{1,1} & b_{1,2} & \dots & b_{1,n} \\ b_{2,1} & b_{2,2} & \dots & b_{2,n} \\ \vdots & \vdots & \ddots & \vdots \\ b_{m,1} & b_{m,2} & \dots & b_{m,n} \end{pmatrix}^{-1} \begin{pmatrix} c_1 \\ c_2 \\ \vdots \\ c_n \end{pmatrix}$$
(1)

was not possible for the recorded LaBr₃:Ce spectra. This was discussed in Ref. [4], depending on the chosen unfolding technique and the number of recorded fission events. In Eq. 1, a_i and c_i denote the yields of the emission and the measured spectrum, respectively, at energy E_i . The values for $b_{i,j}$ represent the elements of the detector response matrix, which may be obtained by Monte Carlo simulation of the experimental setup with e.g. GEANT4 [7]. For a square matrix, i.e. m = n, this response matrix may be replaced by a diagonal matrix whose elements are given by

$$b_{i,j} = \begin{cases} c_i/a_i, & \text{if } i = j\\ 0, & \text{if } i \neq j \end{cases}$$
(2)

These matrix elements can be determined for an experiment for which measured and emitted spectra are known, e.g. after proper unfolding of the simulated detector response, and applied to a measured spectrum from another experiment to deduce the corresponding emission spectrum, as long as the experimental setup is identical and the shapes of both measured spectra a very similar. This procedure was already applied previously to PFGS from ^{240,242}Pu(sf) in Ref. [8]. Since the PFGS measurements from the reactions n + ²³⁸U and n + ²³⁵U at $\overline{E}_n = 1.7$ MeV were carried out simultaneously with the targets mounted back-to-back, the first condition was fulfilled. Moreover, we observed that both measured spectra indeed exhibit a very similar shape (see Fig. 2).

Hence, the second requirement is met too. Accordingly, the matrix elements $b_{i,i}$ were obtained for the system n + ²³⁸U, however by using the measured LaBr₃:Ce spectrum at $\overline{E}_n = 1.7$ MeV and the previously determined emission spectrum at $\overline{E}_n = 1.9$ MeV. Here we apply the same argument as above that the emission spectra at 1.7 and 1.9 MeV are practically identical, which is corroborated by Fig. 1, at least for γ -ray energies below 3 MeV. However, this does by no means imply a restriction, because the low statistics for the 1.7 MeV measurement does not allow to consider LaBr₃:Ce spectra above that energy anyway. Instead, this energy region is covered by the spectrum taken with the BaF₂ detectors.

As a result, we obtain emission spectra for 235 U(n, f) at $E_n = 1.7$ MeV, as shown in Fig. 3, one from the LaBr₃:Ce detectors and the other from the BaF₂ clusters, both depicted as symbols. The BaF₂ spectrum here has been scaled according to the BaF₂ spectrum from the 238 U(n, f) measurement

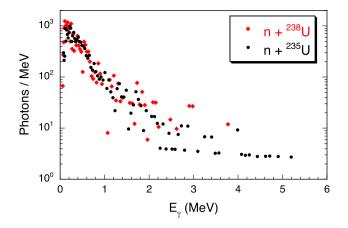


Fig. 2 Measured prompt fission γ -ray spectra from n + ²³⁸U and ²³⁵U depicted as red diamonds and black circles, respectively. Both spectra were taken simultaneously at $\overline{E}_n = 1.7$ MeV with LaBr₃:Ce detectors. Error bars are omitted because of legibility reasons

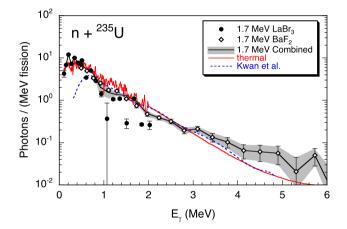


Fig. 3 Prompt-fission γ -ray emission spectra for the reaction n + ²³⁵U at $\overline{E}_n = 1.7$ MeV obtained with the LaBr₃:Ce detectors (this work) and the BaF₂ clusters [2], both depicted as symbols. In the overlap region between 0.5 and 2.0 MeV is shown as full drawn line. A recently measured emission spectrum from thermal neutrons on ²³⁵U [9] as well as a scaled spectrum from Ref. [10], the latter for neutrons of energies between 1 and 2 MeV, are shown too for comparison

Table 1 Summary of PFGS characteristics from the reaction ²³⁵U(n, f) from this work for the range 0.1 MeV $< E_{\gamma} < 6.0$ MeV. Average multiplicity \overline{M}_{γ} , average total γ -ray energy $\overline{E}_{\gamma,\text{tot}}$, and average energy per γ ray $\overline{\epsilon}_{\gamma}$ are given for two different prompt time windows. In addition, results from a recent measurement with thermal neutrons are given for comparison

Reference	This work		[9]
Detector	LaBr ₃ :Ce/BaF ₂		LaBr3:Ce
Prompt window	$\pm 2 \times \Delta t$	\pm 2.5 × Δ t	\pm 2.5 \times Δ t
\overline{E}_n (MeV)	1.7	1.7	thermal
\overline{M}_{γ}	7.16 ± 0.17	7.45 ± 0.48	7.38 ± 0.18
$\overline{E}_{\gamma,tot}$ (MeV)	5.79 ± 0.22	6.03 ± 0.47	6.37 ± 0.20
$\overline{\epsilon}_{\gamma}$ (MeV)	0.81 ± 0.04	0.81 ± 0.04	0.86 ± 0.03

(cf. Fig. 1), hence both have the same height relative to each other just like it is shown in Ref. [2]. The LaBr₃:Ce spectrum was adjusted to the BaF₂ spectrum by requiring the same integral, i.e. multiplicity, within the overlap region $0.5 \text{ MeV} < E_{\gamma} < 2.0 \text{ MeV}$. A black line with grey-shaded uncertainty band denotes a combined spectrum, which consists of the spectra taken with the LaBr₃:Ce and the BaF₂ detectors below and above the overlap region, respectively. and in between of an averaged spectrum of both-re-binned to the same energies. For the sake of clarity, it should be mentioned again that both original spectra, taken with the BaF₂ and the LaBr₃:Ce detectors, respectively, have different bin sizes and positions. It may appear that the combined spectrum in the overlap region does not always correspond to averaged values, but this is not the case and explained by statistical fluctuations of the LaBr3:Ce spectrum.

From this combined spectrum, PFGS characteristics for the fast-neutron induced fission of ²³⁵U were deduced like described e.g. in Ref. [4]. The results for the average multiplicity \overline{M}_{γ} , the average total γ -ray energy $\overline{E}_{\gamma,\text{tot}}$, and the average energy per γ ray $\overline{\epsilon}_{\gamma}$ are given in the first column of Table 1, together with corresponding values in column three from another recent experiment on the same fissioning system, but induced by thermal neutrons [9]. For the fast-neutron induced PFGS results, the uncertainties include statistical ones as well as those from the adjustments made and the transformation function. The spectrum from Ref. [9] is also depicted in Fig. 3 for comparison. For the sake of completeness, another spectrum, taken some years ago for incident neutrons of energies between 1 and 2 MeV [10], is shown as well. However, it has to be noted that the latter was not normalized to the number of fission events, but scaled here to guide the eye. Hence, no PFGS characteristics could be determined. Nevertheless, the slope above 1 MeV γ -ray energy agrees well with the others.

Apparently, the combined spectrum for fast-neutron induced fission agrees rather well with the one from thermal fission, both in shape and absolute height, as the numbers do in Table 1. Still, all values are somewhat lower for fast neutrons than for thermal ones, which is certainly opposite to our expectations, see Ref. [5] for $n + {}^{238}U$. As a matter of fact, this was already observed for the system $n + {}^{239}Pu$ (cf. Ref. [11] for thermal neutrons, Refs. [12, 13] for $\overline{E}_n =$ 1.8 MeV). However, one should take into account that the width of the chosen prompt window in the thermal-neutron induced fission on ²³⁵U [9] and the fast-neutron induced fission on 238 U [4] (used for the normalization) is five and four times the coincidence timing resolution, respectively. The influence of a different coverage of the prompt peak was assessed recently [14], indicating that the multiplicity (and the average total γ -ray energy, see Ref. [14]) at $\overline{E}_n = 1.7 \text{ MeV}$ is underestimated by a factor of 0.96 relative to the thermalneutron induced values. Applying this factor on the results

in column one of Table 1, the new values become $\overline{M}_{\gamma} = 7.45 \pm 0.48$ and $\overline{E}_{\gamma,\text{tot}} = (6.03 \pm 0.47)$ MeV, which contain an additional relative uncertainty of 4% from the time window correction. The average energy per γ ray and its uncertainty $\overline{\epsilon}_{\gamma} = (0.81 \pm 0.04)$ MeV remains unchanged. These final results are listed in column two of Table 1 and will be discussed below.

4 Discussion and conclusion

For the system n + 235 U we have presented the very first results for fast-neutron induced prompt fission γ -ray characteristics, which was achieved by combining results from a previously performed comparative measurement [1] and a recent study on n + 238 U [4] at almost identical incident neutron energy.

Previously reported preliminary values [15] had to be corrected—in two respects. The average incident neutron energy of 1.5 MeV is replaced by 1.7 MeV, after careful analysis of the distribution of neutrons, produced in an inverse $p(^{7}Li, ^{7}Be)n$ reaction with LICORNE, as stated in Ref. [1]. Moreover, the average multiplicity was estimated back then based on systematics [5] and on results from a previous experiment with thermal neutrons [16]. However, with the recent thermal-neutron induced measurement by Ref. [9] it turned out that earlier the amount of low-energy γ rays, i.e. E_{γ} below 1 MeV, was overestimated by an underestimation of the backscattering component in the detector response simulation [9]. As a consequence, the average multiplicity as well as the average total γ -ray energy released in fission determined back then [16] and the corresponding estimated values for fast neutrons [15] were too high. Such kind of estimates were not necessary anymore with the preceding PFGS measurement from $n + {}^{238}U$ [4]. We only assumed that a difference in incident neutron energy between 1.7 and 1.9 has a negligible impact on PFGS characteristics, which was inferred from semi-empirical model calculations [5] and corroborated by both a previous measurement at 1.6 MeV incident neutron energy [6] and a recent evaluation [17].

The final PFGS characteristics deduced in this work, as given in column two of Table 1, were obtained for a prompt timing window that initially had a width of four times Δt , with a coincidence timing resolution $\Delta t \approx 4$ ns. This allows certainly some γ decay of isomers to be included in the PFGS, but this contribution is much smaller than "real" prompt fission γ rays and does not really affect the spectral characteristics, as assessed elsewhere [18]. The results are in good agreement with the latest measurement with thermal neutrons [9]—within the uncertainties. It may be noted that the results from the recent PFGS measurement with thermal neutrons on 235 U were provided to the evaluators of ENDF/B-VIII.0 (cf. Ref. [17]). The observed average prompt-fission γ -ray

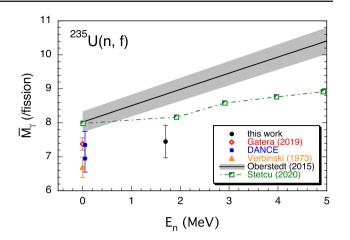


Fig. 4 Average prompt-fission γ -ray multiplicity for the reaction n + ²³⁵U as a function of incident neutron energy. Different experimental data points are depicted as symbols, while evaluations are shown as lines (see text for details)

multiplicity as function of incident neutron energy is shown in Fig. 4. The result from this work as well as a selection of experimental data from Refs. [9,19–21] for thermal and slow neutrons are shown as full circles, open diamonds, full squares and triangles, respectively, while evaluations from Refs. [5,17] are shown as lines. Accordingly, Fig. 5 shows the average total prompt-fission γ -ray energy released in fission as a function of incident neutron energy for data from the same references as in Fig. 4 (more data is included in Ref. [17]).

Apparently, all experimental multiplicity values agree rather well with each other, but both evaluations overestimate the average multiplicity regardless of incident neutron energy, although an increase with energy is weakly supported by the experimental value from this work. In contrast, the average total energy is well reproduced by the evaluations, but a suggested weak increase with energy cannot be con-

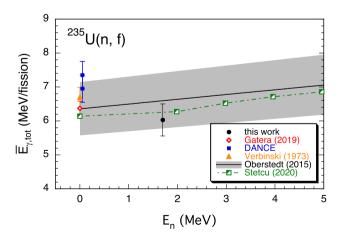


Fig. 5 Same as in Fig. 4, but average total prompt-fission γ -ray energy released in fission for the reaction $n + {}^{235}U$ as a function of incident neutron energy

firmed by our result. All experimental values for the average total γ -ray energy, except for those from DANCE, are in decent agreement – within overlapping error bars. This discrepancy is related to the different ways of determining $\overline{E}_{\gamma,\text{tot}}$ and the fact that DANCE PFGS suffer from a severe underestimation of γ rays with energies below 500 keV, which is reflected by higher mean energies per photon. This was explained in Ref. [22] for ²⁵²Cf(sf). The average energy per photon from this work is observed to be somewhat lower than for thermal fission [9], but still overlapping within the uncertainties (cf. Table 1). It is interesting to note that the recent CGMF calculations for ²³⁵U(n, f) in this incident neutron energy region and considering the same low-energy threshold of 100 keV, gave $\overline{\epsilon}_{\gamma} \approx 0.87$ MeV [17], which is in rather good agreement with our result.

It has to be repeated that the experiment back then suffered from rather low statistics, but still, it led to the first results with respect to PFGS characteristics for fast neutrons on ²³⁵U. Moreover, in times in which it is difficult to carry out new experiments, we believe that analyzing already taken data is worthwhile to obtain-at least-a pretty good idea about the results. Nevertheless, a new measurement with better statistics is highly recommended. The results are important from both applied and basic nuclear physics point of view. As far as applications are concerned, e.g. fast reactors of Generation IV type, PFGS characteristics for $n + {}^{235}U$ up to $E_n = 10$ MeV are requested by OECD NEA [23]. Apart from that, this information is important for the fundamental understanding of the fission process, e.g. an expected correlation between the de-excitation of fission fragments by neutron and γ -ray emission, respectively (see Ref. [13] and references therein). New experimental findings, in particular for fast-neutron induced fission, may then be compared to theoretical predictions in order to improve fission models.

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Data Availibility Statement This manuscript has associated data in a data repository. [Authors' comment: The datasets generated during and/or analysed during the current study are available from the corresponding author on reasonable request.]

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