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First experimental prompt γ -ray spectra in fast-neutron-induced fission of ^{238}U

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The knowledge of prompt fission γ -ray emission has been of major interest in reactor physics for a few years. Only few experimental spectra were published until now for fast-neutron-induced fission, and measurements would be also valuable in order to improve our understanding of the fission process. A simple experimental method was used to measure the first prompt fission γ -ray spectra up to 10 MeV. In this approach, the γ rays are measured with a bismuth germanate (BGO) detector which offers two significant advantages with respect to other γ -ray detectors: a high peak-to-total ratio and a high efficiency. The prompt fission neutrons are rejected by the time-of-flight technique between the BGO detector and a fission trigger given by a fission chamber. Prompt fission γ -ray spectra were measured for 1.6 ± 0.1 , 5.1 ± 0.2 and 15.0 ± 0.6 MeV neutron-induced fission on ^{238}U at the CEA, DAM, DIF Van de Graaff accelerator; average multiplicity and mean photon energy per fission were deduced from the spectra.

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I. INTRODUCTION

Prompt fission γ rays were first experimentally studied in the 1960s and the 1970s [1,2]. In spontaneous, thermal, and resonance neutron-induced fission, some measurements of the energy spectrum [3,4], of the mean multiplicity and the mean total energy (see for example [3–7]) were performed. Forty years later, a renewed interest is growing both on the experimental side [8–11] as well as on the theoretical one (see, e.g., Ref. [12]).

Prompt fission γ rays are a source of information on fission dynamics. If around 75% of the fragment's excitation energy is dissipated by prompt neutrons, most of their angular momentum is evacuated by prompt γ rays. Now it is known that the fission fragments are produced in a relatively high angular momentum state [13], according to a mechanism that remains to be understood. When most of the fragment excitation energy has been dissipated, only the emission of γ rays is possible. For excitation energies above the neutron binding energy, the daughter fragment angular momentum may favor the emission of a γ rays. This leads to the so-called neutron- γ competition measured and discussed by Nifenecker *et al.* [14] and Fréhaut *et al.* [15]. Hence the study of prompt γ rays can shed light on the angular momentum generation and set constraints in models on prompt particle emission.

On the other hand a better knowledge of the prompt fission γ -ray emission is needed for simulating the γ heating in reactor cores. Accurate data are required on the energy spectrum shape up to 10 MeV, as well as on the average multiplicity and mean photon energy per fission, both in thermal and fast-neutron-induced fission [16,17]. In fast-neutron-induced

fission, the only experimental work before the year 2010 was performed by Fréhaut, who obtained the mean total energy, relative to the one of ^{252}Cf , released in the fission of ^{235}U , ^{237}Np , ^{232}Th induced neutrons with energies between 2 and 15 MeV [18] and in the fission of ^{241}Pu [15]. Madland *et al.* [19] have used that data to perform an evaluation of the prompt γ -ray mean total energy for incident neutrons up to 15 MeV in the fission of ^{235}U . They have also proposed an evaluation in the case of fast fission on ^{238}U and ^{239}Pu . The case of fast-neutron-induced fission is particularly interesting to understand how extra excitation energy in the fissioning system converts in the exit channel. First measurements of the prompt fission γ -ray spectra (PFGS) in fast-neutron-induced fission of actinides were recently performed on ^{235}U by Kwan *et al.* [20] and on ^{238}U by us [21,22].

In this paper we extensively report on our PFGS measurements at incident neutron energies of 1.6, 5.1, and 15.0 MeV. After a presentation of the measurement method, the experiments and the data treatment are described, whereupon the results are given and discussed.

II. EXPERIMENT

The experimental method consisted in irradiating a fission chamber with a monoenergetic neutron beam. A γ -ray detector was placed 1 m away from the fission chamber chosen in a way that the time-of-flight (TOF) technique allows separating prompt fission γ rays from prompt fission neutrons. The fission chamber signal was used to build a pulse-height distribution in order to discriminate the fission fragments from α particles. The fission chamber signal was used as the event trigger in the experiment. The measured photon spectra were

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FIG. 1. Peak to total ratio P/T for different kinds of cylindrical γ -ray detectors (2 in. $\times$ 2 in.) as a function of the incident γ -ray energy (GEANT 3.21 calculation). The density ρ is 3.67 for NaI, 4.88 for BaF₂, 5.08 for LaBr₃, 5.23 for Ge, and 7.13 for BGO. The effective atomic number Z_{eff} is 32 for Ge, 40.5 for LaBr₃, 50 for NaI, 52 for NaI, and 75 for BGO. The P/T for the 3 in. $\times$ 6 in. BGO detector used in the experiments is also plotted.

then used to obtain the prompt fission γ -ray emission spectra, after unfolding the detector response.

According to Ref. [13], page 501, more than 90% of the prompt γ rays are emitted within 1 ns. Hoffman and Hoffman [1] asserted that about 95% of the prompt γ rays are emitted within less than 10 ns. Moreover, Skarsvag [23] showed for the spontaneous fission of ^{252}Cf that, at a given emission time, the proportion of released energy is larger than the proportion of the number of emitted γ rays. So about 95% of the number of γ rays and more than 95% of the total energy should be measured within a time window of 10 ns.

As no fission axis is preferred inside the active target, the prompt γ -ray measurement was not sensitive to the anisotropy and the γ -ray detector could be placed at any angle from the axis of the neutron beam impinging on the fission target.

The neutrons were produced at the CEA, DAM, DIF Van de Graaff accelerator using the following neutron production reactions: $^3\text{H} (p, n) ^3\text{He}$ for 0.5–3-MeV neutrons, $^2\text{H} (^2\text{H}, n) ^3\text{He}$ for 3–7-MeV neutrons, and $^3\text{H} (^2\text{H}, n) ^4\text{He}$ for 15–20-MeV neutrons. The neutrons fluxes ranged from 10^6 to $10^7 \text{ s}^{-1} \text{ sr}^{-1}$.

The choice of the detector was mainly done with respect to its peak-to-total ratio (P/T), i.e., the ratio of the counts in the peak normalized to the counts in the entire spectrum. By choosing the highest one, the efficiency at high energy was privileged, together with the minimization of distortions of the spectra. These two aspects are crucial to minimize uncertainties in the unfolded spectra. Several γ -ray detectors were compared by performing simulations with the Geant 3.21 Monte Carlo code [24]. BGO, HPGe, NaI, BaF₂, LaBr₃ of the same dimensions (2 in. $\times$ 2 in.) were considered and the result of the simulation is plotted in Fig. 1. The highest P/T ratio is obtained with a bismuth germanate detector (BGO = $\text{Bi}_4\text{Ge}_3\text{O}_{12}$) which offers a value twice as high as that of a LaBr₃ detector at 1 MeV and even five times higher at 10 MeV. The high values of P/T for BGO is due to the high effective atomic number ($Z_{\text{eff}} = 75$), and due to its

high density ($\rho = 7.13 \text{ g/cm}^3$). To our knowledge, only one detector offers a comparable P/T ratio to that of the BGO detector: a PbWO_4 -based scintillation detector, which was developed for high-energy physics experiments. However, its very low light yield prevents using it in the measurement of γ -ray energies lower than several MeV. Moreover, compared to BaF₂ and LaBr₃ of the same dimensions, the BGO detector offers a full-energy peak efficiency more than twice and five times higher at 1 and 10 MeV, respectively.

A $\Phi 3 \text{ in.} \times \text{L6 in.}$ BGO detector ($\Phi 7.6 \text{ cm}$, L15.2 cm) was bought from Scionix Holland BV, equipped with a Photonic XP 3330 photomultiplier. As shown in Fig. 1, the P/T ratio reaches 83% at 1 MeV and 36% at 10 MeV. The energy resolution was measured to be close to 11% at 0.662 MeV. In the range of interest, the pulse height response was found to be remarkably linear with energy. The energy resolution calibration curve was found to be compatible, within 68% C.L., with the expected energy square root variation: the relative full width at half maximum is $\text{FWHM} (\%) = a \times E^{-b}$ with $a = 346 \pm 29$, $b = 0.517 \pm 0.017$, and E in keV. During the experiment, the neutron background induces $^{73}\text{Ge} (n, \gamma)$ radiative capture reactions taking place inside the BGO crystal. This leads in the background spectrum to a peak at 10.19 MeV, corresponding to the ^{74}Ge neutron binding energy emitted by all capture γ rays. This peak allowed us to monitor online the overall gain, in addition to dedicated radioactive source runs.

To deliver an absolute emission spectrum of the prompt fission γ rays, the full-energy peak efficiency must be known in the whole energy range. From 0.06 up to 1.8 MeV, standard radioactive sources are commercially available. Above 2 MeV, (α, n) radioactive sources were used. The AmBe source emits 4.44-MeV γ rays whose fluence is related to the known neutron one by a factor close to 0.6 measured by several authors (see for example Refs. [25] or [26]). The Pu- ^{13}C source emits a 6.13-MeV γ ray whose fluence was given in the calibration certificate. To measure the full-energy peak efficiency up to 10 MeV, an experiment was carried out to take advantage of the $^{27}\text{Al} (p, \gamma) ^{28}\text{Si}$ reaction: the proton resonance at 992 keV proton energy leads to a γ cascade with emission of 1.778- and 10.763-MeV γ rays whose emission probability ratio is known with an accuracy of 1.2% [25]. This ratio then allows deducing the efficiency at 10.763 MeV from that at 1.778 MeV. Doing so, a full-energy peak efficiency curve was derived up to 10.76 MeV, along with uncertainties coming from the measured ones and the source's activity ones given in the manufacturer's data sheets.

It is of relevance to observe that, thanks to the high density and effective atomic number of the BGO crystal, the efficiency at 10 MeV is only 2.5 times less than that at 1 MeV. As shown in Fig. 2, a good agreement was found with the Geant 3.21 calculation. We also used the PENELOPE code [27], which provided efficiency values very close to the one obtained with Geant 3.21, with typically discrepancies smaller than 1%.

III. SETUP AND EXPERIMENTAL CONDITIONS

Two experiments were carried out: during the first experiment, one measurement was performed with neutrons

FIG. 2. 3 in. $\times$ 6 in. (diameter $\times$ length) BGO detector full-energy peak efficiency at a distance of 90 cm.

produced by shooting 2.5-MeV protons on a tritiated titanium target (p,T reaction) and another one by shooting 0.4-MeV deuterons on another tritiated titanium target (d,T reaction). In the second experiment, with a slightly different setup, the measurement with the p,T reaction was performed again. A second measurement was performed with neutrons produced by shooting 2-MeV deuterons on a deuterated titanium target (d,D reaction).

In the thickness of the production target, the ions slowed down and this caused a dispersion in the emitted neutron energy. On the other hand, the neutrons impinging the fission chamber were emitted at an energy that depended, to a certain extent, on the emission angle. Taking into account both phenomena leads to the following values of the mean energy and standard deviation of the incident neutrons: 1.6 ± 0.1 MeV for the (p,T), 5.1 ± 0.2 MeV for the (d,D), and 15.0 ± 0.6 MeV for the (d,T) reaction, respectively.

In both experiments, the revolution axis of the BGO detector was aligned perpendicular to the fission chamber axis. The detector front face was placed respectively at 72.5 and 77 cm from the fission chamber axis. Since the incident neutrons coming from the production target were emitted in a 4π solid angle, the BGO detector had to be shielded. These background neutrons undergo inelastic scattering in the BGO crystal, and in particular on ^{209}Bi leading to a prominent γ peak at 0.896 MeV. A shielding made of iron, polyethylene and lead were placed between the BGO detector and the production target. Moreover, a boron-loaded plastic material was used to reduce this neutron capture background due to room return. The $^{10}\text{B}(n,\alpha)^7\text{Li}$ reaction is efficient to sharply reduce the flux of low-energy neutrons and thus the capture rate inside the BGO. The counterpart of the (n,α) reaction is the emission of a 477.6-keV γ ray that added to the low-energy background, in an energy range however where the signal to background ratio is very good.

The fission chamber, containing 14 g of ^{238}U , was placed in front of the neutron production target and the recorded fission rate was about 100 s^{-1} . In the fission chamber the inox plates were covered with 1.7 mg cm^{-2} of ^{238}U over a diameter of 7 cm, and separated by a 1-mm gap. The gas used was a mixing of Ar (80%) and CH_4 (20%) under a pressure of 5 bars. The plates were electrically assembled by

FIG. 3. Time-of-flight spectrum between the fission chamber and the BGO detector for 1.6 ± 0.1 MeV incident neutrons in the second experiment, for γ -ray energies greater than 1 MeV. Typical TOF cuts applied to extract the (signal + background) and the background γ -ray events are shown by vertical lines.

groups of 20, each delivering a signal read by a preamplifier. In the first experiment an Ortec 142C charge-sensitive preamplifier was used in conjunction with analog electronics (Constant fraction discriminator - CFD, pulse shaping) to read out each group of plates. In the second experiment, preamplifiers developed in our laboratory and adapted to high capacitance detectors were used. The output signals were processed with a digital acquisition system based on 12-bit 500-MHz flash analog-to-digital converters [28] and field programmable gate array (FPGA) signal processors. Pulse shaping and constant fraction discrimination were implemented into these FPGAs. In both experiments the energy deposited in the fission chamber allowed us to separate the fission fragments from the α particles. The overall timing resolution measured with the γ peak in the time-of-flight (TOF) spectrum was 17 ns at FWHM in the first experiment and was improved in the second one to 10 ns (FWHM). An example of a TOF spectrum taken at a low-energy threshold for γ rays at 1 MeV is depicted in Fig. 3.

IV. DATA ANALYSIS

The data analysis consisted first in building the experimental γ -ray spectra and second in unfolding the experimental spectra from the setup response in order to deduce the emission spectra, i.e., the PFGS. In order to unfold the spectra, the detector response was simulated at different energies with the PENELOPE Monte Carlo code.

The prompt fission γ -ray events were selected thanks to the coincidence with the fission events and with the γ peak in the TOF spectrum. That selection gave the (signal + background) spectrum. The background due to uncorrelated neutrons was obtained by constructing the BGO spectra in coincidence with events arriving just before the γ peak (see Fig. 3). The time window has the same width as the one used to select the γ peak. The resulting background-suppressed γ -ray spectrum was mainly composed of prompt fission γ rays, but also contained a proportion of events coming from high-energy prompt fission neutrons (detected via inelastic scattering).

TABLE I. Width of the prompt γ peak in the TOF spectrum at FWHM for different γ -ray energy regions: For both experiments, the fraction of prompt neutron rejection is given.

ΔE (MeV)	First experiment			Second experiment		
	FWHM (ns)	Cut (ns)	Neutron rejection	FWHM (ns)	Cut (ns)	Neutron rejection
<0.2	27	25	91.4%	20	20	99.3%
0.2–0.3	18	20	98.6%	17	18	99.8%
0.3–0.4	17	19	99.2%	13	16	99.98%
0.4–0.5	14	19	99.2%	10	16	99.98%
>0.5	12	18	99.6%	8	14	>99.99%

However this contribution is very small as we will see later. It could also include a proportion of γ rays coming from the deexcitation of isomeric states in the fragments as well as γ rays from inelastic scattering of prompt fission neutrons on the fission chamber materials; both kinds of events are detected later than prompt fission γ rays hitting the BGO detector. Without those events the γ peak shape is expected to be Gaussian and symmetric; however if there would be a significant number of those γ rays arriving later, a tail should enlarge the peak on the higher times side. However, there is no asymmetry observed for all γ -ray energy bins except for the 0.1–0.2-MeV region. This bin contributes to around 15% in the whole spectrum and the extra events in the tail are estimated not to increase the number of events by more than 10–20% compared to a symmetric-shaped peak; thus the contribution of γ rays due to isomeric decays and inelastic neutron scattering should be less than a few percent.

It is well known that the sensitivity of the BGO crystal to fast neutrons is much lower than to γ rays and this feature is enhanced with BGO compared, for example, to NaI [29]. For γ -ray energies above 1 MeV, the prompt fission neutron bump appeared clearly as can be seen in Fig. 3. Considering a Watt spectrum with $A = 1.093 \text{ MeV}^{-1}$ and $B = 2.3075 \text{ MeV}^{-1}$ for the prompt fission neutron spectrum [30], a 99.5% rejection of prompt fission neutrons implies that all neutrons with energy less than 8.3 MeV have to be rejected. For a flight path d , the corresponding TOF in nanoseconds is $25.1 \times d(\text{m})$, that is 18 and 19 ns for the first and the second experiment, respectively. With such a criterion, the prompt γ -ray events in the γ peak had to be selected with a cut around the centroid of ± 18 ns for the first experiment and ± 19 ns for the second one.

However the γ peak has not the same width for all energies. It is broader for lower γ -ray energy, because the number of photoelectrons is poor at low energy and the signal shape deteriorates, leading to an enhanced jitter appearing in the time pickoff. Table I summarizes the FWHM of the prompt γ peak as a function of the γ -ray energy region. Thus, it is clear that in a range ± 18 or 19 ns not all low-energy γ rays can be included. This is why the width of each cut was defined as a function of the γ -ray energy, so that all prompt fission γ rays could be integrated. The chosen time cuts are given in Table I along with the consecutive proportion of rejected prompt fission neutrons. In order to test a possible contamination of prompt fission neutrons in the spectra, we also extracted the spectra of the second experiment with the first one's larger cuts: both spectra agreed with each other within statistical uncertainties. So, it could not be deduced that

the cuts chosen for the first experiment include more prompt fission neutrons than those chosen for the second one.

The Watt parameters used for the estimation of the neutron rejection were extracted from prompt neutron spectra measured in the 0.5-MeV neutron-induced fission of ^{235}U [30]. A recent measurement of prompt neutron spectra [31] has allowed extracting the Watt parameters in fast-neutron-induced fission of ^{238}U : $A = 1.306 \pm 0.007 \text{ MeV}^{-1}$ and $B = 5.45 \pm 0.04 \text{ MeV}^{-1}$. With these new parameters, the neutron rejection is still better, so we can consider that the values given in Table I are conservative. Indeed, with the new parameters, in the first (respectively second) experiment, it amounts to 93.2% (respectively 99.65%) for $\Delta E < 0.2 \text{ MeV}$ and >99.99% for $\Delta E > 0.4 \text{ MeV}$ (respectively for $\Delta E > 0.3 \text{ MeV}$).

The second step of the data analysis consisted in unfolding the experimental spectra from the setup response. We used the PENELOPE Monte Carlo code [27] to calculate the detector response for different γ -ray energy bins for the two experiments. Compared to the P/T of the nude BGO detector, we found that the P/T was deteriorated by 30% by adding the fission chamber, and by 40% when taking the complete geometry of both setups with all shielding materials surrounding the BGO crystal and the ones used to support the detector.

The two spectra taken at $E_n = 1.6 \pm 0.1$ and $5.1 \pm 0.2 \text{ MeV}$ were distributed into 37 bins, and the one taken at $E_n = 15.0 \pm 0.6 \text{ MeV}$ into 38 bins. The width of each bin follows the energy resolution of the BGO detector. The response of the BGO crystal for a γ ray at given energy was calculated for each of the 37 and 38 incident energy bins. The unfolding procedure consisted in subtracting every event with partial energy deposit, starting from the highest energy, down to the lowest one. This was done by weighting the simulated deposited energy spectra, so that the bin at full energy has the same content as in the experimental spectrum. All uncertainties from the statistics in the simulated spectra as well as the ones in the experimental spectrum were propagated. The most important increase of the uncertainty is found in the lowest bins in which contributions from all higher energy bins has to be accounted for. The effective low-energy threshold is 0.11 and 0.19 MeV for the first and the second experiment, respectively.

Figure 4 shows the unfolded spectra along with the experimental folded spectra for the four measurements. Above 0.7 MeV the amplitude of the four unfolded spectra has decreased by 20–30% compared to the folded one; this decrease was expected from calculations we made earlier [21]. Below

FIG. 4. Folded (triangles) and unfolded (squares) spectra for the four measurements: (a) 1.6 ± 0.1 MeV and (d) 15.0 ± 0.6 MeV incident neutron for the first experiment, (b) 1.6 ± 0.1 MeV, and (c) 5.1 ± 0.2 MeV for the second experiment.

0.7 MeV, the correction increases when the energy lowers. This is explained by the fact that all incident energies can contribute to the lowest energy bins. Since the highest points could not be affected by the unfolding procedure, we applied an *ad hoc* correction. Taken into account that the discrepancy is of 20–30% in all the energy range above 0.7 MeV, it was decided to lower the yield at highest energies by 20%. After unfolding the spectra for both experiments, each unfolded spectrum was divided by the full-energy peak efficiency of the setup and normalized to the number of fissions, leading to the experimental PFGS, shown in Figs. 5 and 6. Both PFGS for 1.6-MeV incident neutrons agree within 1σ for most of the points, within 1.5σ for all points. Weighted by the statistical uncertainties, they were combined to give a higher statistical significance to the spectral characteristics.

FIG. 5. Experimental PFGS in the fission of ^{238}U induced by 1.6- and 5.1-MeV incident neutrons (dots). Comparison with the one of Verbinski *et al.* [4] in thermal fission of ^{235}U (line).

The 68% C.L. statistical uncertainty varies from 2% to 20% below a γ -ray energy of 5 MeV, and from 20% up to 86% up to 10 MeV. Systematic uncertainties result basically from the simulation of the detector response and the efficiency calibration (10–13%, depending on the incident energy), and from the number of fission (3%). For each energy bin, both statistical and systematic uncertainties are given in Table II, in superscript and subscript, respectively, of the number of photons per MeV and per fission.

V. RESULTS

At 1.6 MeV incident neutron energy, 1.2×10^7 fissions and 5.3×10^4 prompt fission γ rays were acquired in the first experiment. In the second experiment, 1.1×10^7 fissions and

FIG. 6. Experimental PFGS in the fission of ^{238}U induced by 15.0-MeV incident neutrons, compared to the one obtained with 1.6-MeV incident neutrons. In addition, PFGS from Ref. [20] for the system $n + ^{235}\text{U}$ for similar incident neutron energies are shown for comparison.

TABLE II. PFGS data (in $\text{MeV}^{-1} \text{fission}^{-1}$) in ^{238}U fission induced by 1.6-, 5.1-, and 15.0-MeV neutrons. The center and the width of the energy bins are given. Given at 68% C.L., the statistical uncertainty appears in superscript, the systematic one in subscript.

E_γ (keV)	ΔE (keV)	Incident neutron energy (MeV)		
		1.6 $\pm$ 0.1	5.1 $\pm$ 0.2	15.0 $\pm$ 0.6
128	35.5	8.6 $^{0.7}_{1.5}$		10.6 $^{1.0}_{1.6}$
166	40.4	10.4 $^{0.6}_{1.6}$		12.1 $^{0.8}_{1.8}$
209	45.4	8.2 $^{0.4}_{1.3}$	8.7 $^{0.9}_{1.3}$	8.1 $^{0.6}_{1.2}$
256	50.3	7.5 $^{0.3}_{1.0}$	8.8 $^{0.5}_{1.2}$	5.6 $^{0.3}_{0.6}$
309	55.3	7.7 $^{0.2}_{1.0}$	8.4 $^{0.4}_{1.0}$	6.0 $^{0.4}_{0.7}$
367	60.2	7.2 $^{0.2}_{0.8}$	7.9 $^{0.3}_{0.9}$	5.6 $^{0.3}_{0.6}$
430	65.2	8.0 $^{0.2}_{0.8}$	8.1 $^{0.3}_{0.8}$	7.0 $^{0.3}_{0.7}$
497	70.1	6.6 $^{0.2}_{0.7}$	6.9 $^{0.2}_{0.7}$	6.4 $^{0.3}_{0.7}$
570	75.1	5.8 $^{0.1}_{0.6}$	6.3 $^{0.2}_{0.7}$	5.8 $^{0.2}_{0.6}$
647	80.0	5.2 $^{0.1}_{0.5}$	5.1 $^{0.5}_{0.1}$	5.7 $^{0.2}_{0.6}$
730	85.0	4.4 $^{0.1}_{0.5}$	4.7 $^{0.1}_{0.5}$	5.1 $^{0.2}_{0.5}$
817	85.0	4.3 $^{0.1}_{0.4}$	4.0 $^{0.1}_{0.4}$	4.8 $^{0.2}_{0.5}$
925	128	2.5 $^{0.1}_{0.3}$	2.5 $^{0.1}_{0.3}$	3.6 $^{0.2}_{0.4}$
1057	136	1.94 $^{0.05}_{0.20}$	2.1 $^{0.1}_{0.2}$	2.5 $^{0.1}_{0.3}$
1198	145	1.96 $^{0.05}_{0.20}$	1.91 $^{0.07}_{0.20}$	2.3 $^{0.1}_{0.2}$
1348	154	1.42 $^{0.04}_{0.15}$	1.35 $^{0.04}_{0.14}$	1.9 $^{0.1}_{0.2}$
1506	163	1.04 $^{0.04}_{0.11}$	1.04 $^{0.04}_{0.11}$	1.6 $^{0.1}_{0.2}$
1673	172	0.85 $^{0.03}_{0.09}$	0.83 $^{0.04}_{0.09}$	1.31 $^{0.08}_{0.14}$
1849	180	0.69 $^{0.03}_{0.07}$	0.67 $^{0.03}_{0.07}$	0.96 $^{0.08}_{0.10}$
2034	189	0.59 $^{0.03}_{0.06}$	0.55 $^{0.03}_{0.06}$	0.77 $^{0.06}_{0.08}$
2228	198	0.49 $^{0.02}_{0.05}$	0.48 $^{0.03}_{0.05}$	0.72 $^{0.07}_{0.07}$
2430	207	0.38 $^{0.04}_{0.02}$	0.35 $^{0.02}_{0.04}$	0.62 $^{0.06}_{0.07}$
2641	216	0.30 $^{0.02}_{0.03}$	0.31 $^{0.02}_{0.03}$	0.53 $^{0.06}_{0.06}$
3032	578	0.20 $^{0.01}_{0.02}$	0.18 $^{0.01}_{0.02}$	0.33 $^{0.03}_{0.03}$
3637	633	0.122 $^{0.007}_{0.013}$	0.093 $^{0.006}_{0.010}$	0.26 $^{0.026}_{0.027}$
4297	688	0.062 $^{0.005}_{0.006}$	0.047 $^{0.004}_{0.005}$	0.14 $^{0.023}_{0.014}$
5012	743	0.020 $^{0.004}_{0.002}$	0.018 $^{0.002}_{0.002}$	0.105 $^{0.020}_{0.011}$
5782	798	0.013 $^{0.003}_{0.001}$	0.012 $^{0.002}_{0.001}$	0.065 $^{0.018}_{0.007}$
7040	1760	0.0067 $^{0.0028}_{0.0007}$	0.0026 $^{0.0006}_{0.0003}$	
9412	3053	0.00098 $^{0.00085}_{0.00010}$		
6836	1330			0.045 $^{0.012}_{0.005}$
8288	1579			0.019 $^{0.008}_{0.002}$
10026	1904			0.0082 $^{0.0045}_{0.0009}$

4.3×10^4 prompt fission γ rays were recorded. The combined PFGS data are given in Table II and illustrated in Fig. 5. Since no PFGS data were ever measured in fission on ^{238}U before, we compare our spectrum here with the one measured by Verbinski *et al.* in thermal-neutron induced fission on ^{235}U [4] that used to be considered as PFGS reference and was recently confirmed and improved by Oberstedt *et al.* [11]. It can be observed that, despite the fact that the fissioning

systems are different, the spectra are remarkably similar in shape and amplitude up to 7–8 MeV.

At 5.1-MeV incident neutron energy, 1.4×10^7 fissions and 6.0×10^4 prompt fission γ rays were measured. As shown in Table II and Fig. 5, the spectrum compares very well with the one at 1.6 MeV, both in shape as well as in amplitude: in most cases, they agree within the 68% C.L. statistical uncertainties. In particular, the high-energy parts of the two spectra match perfectly, and this indicates that the mean temperature of the fission fragments emitting the γ rays should be rather similar for both incident energies. This observation coincides well with the usual understanding that most γ rays are emitted at the very end of the deexcitation of the excited primary fragments.

At $E_n = 15.0$ MeV finally, 8.3×10^6 fissions and 4.3×10^4 prompt fission γ rays were acquired. Up to 0.8 MeV the spectrum is compatible, within 68% C.L., with the one obtained at 1.6-MeV incident neutrons, but it becomes significantly harder above 1.5 MeV, up to one order of magnitude (see Table II and Fig. 6).

At this incident energy also second and third chance fission occurs. For first chance fission, the compound nucleus has an extra excitation energy of 13.9 MeV compared to the 1.6-MeV neutron-induced fission. Since the neutron binding energy is 4.8 MeV in ^{239}U , and 6.1 MeV in ^{238}U , the excitation energy in the fissioning system is still 10.8 MeV in second chance fission and 4.7 MeV in third chance fission. According to the GEF code [32] (release of January 2015, version 1.1), the probabilities are 33% for the first, 49% for the second, and 17% for the third chance fission.

The deexcitation of the fragments is first made by the emission of statistical prompt neutrons, then $E1$ γ rays. When the excitation energy approaches the Yrast line, $E2$ transitions take place. This known deexcitation scheme tells that the high-energy part of the PFGS is made of statistical γ rays whereas the low-energy part is dominated by discrete ones which evacuate most of the fragment angular momentum. If we compare fission induced by 15.0 with that induced by 1.6-MeV neutrons, the extra excitation energy will be mostly evacuated during the statistical decay of the fragments, i.e., that means that the observed increase in amplitude in the high-energy range of the spectrum might be expected, even if the excitation energy is mostly evacuated by prompt neutrons. On the other hand, between 1.6 and 15.0 MeV, the incident neutrons carry an extra angular momentum of no more than 2–3 $\hbar$, split over both fragments, which are produced each with an angular momentum of about 6 $\hbar$ [13]. So the angular momentum state is not very different and we should not expect a significant increase of discrete prompt γ rays. This confirms the observed comparable amplitude in the low-energy range of both spectra.

The PFGS are compared in Fig. 7 to predictions made in the framework of the GEF code (release of January 2015, version 1.1). For 1.6-MeV incident neutrons, the agreement is quite good up to 6 MeV. Above 6 MeV, the predicted spectrum falls much more rapidly than the experimental one does in agreement with the Verbinski one (cf. Fig. 5). The predictions for 1.6- and 5.1-MeV incident neutron energy are very similar and the experimental spectra agree within

FIG. 7. Comparison of the experimental PFGS (dots) to predictions (lines) from the GEF code (release of January 2015, version 1.1) [32] and the FIFRELIN one [34].

the uncertainties. The spectra predicted by the code for 1.6- and 15.0-MeV incident neutron energy do not differ as much as the experimental spectra do in the high-energy range. So, for 15.0-MeV incident neutrons we observe an agreement between the predicted and the experimental spectra up to 4 MeV.

In Fig. 6 our spectra are compared with corresponding ones whose neutron energy range matches our incident neutron energies, however for the similar system $n + {}^{235}\text{U}$. These values were digitized from Fig. 8(d) in Ref. [20], normalized and adjusted to our spectra. Obviously, the overall agreement between both datasets is not really good, matching each other only for γ -ray energies above 0.6 MeV. This deviation at low energies has been explained and quantified in Ref. [33]. At γ -ray energies above some MeV, both spectra from Ref. [20] agree rather well with our spectrum taken at $E_n = 1.6$ MeV, while our spectrum taken at $E_n = 15.0$ MeV appears much harder. Hence, the difference in hardness of the spectra for $n + {}^{238}\text{U}$ with respect to both incident energies, as explained above, is not observed for $n + {}^{235}\text{U}$.

In Fig. 7 we also show a spectrum recently calculated with the FIFRELIN code [34] for 1.8-MeV incident neutrons: the shape and amplitude are rather similar to the GEF spectrum in the whole energy range as well as the experimental ones up to 6 MeV.

From the spectra we were able to deduce the mean photon energy and the average multiplicity. They are given in Table III. Experimental values are given for a low-energy threshold of 0.110 MeV for the measurement at 1.6- and 15.0-MeV incident neutron energy and for a corresponding threshold at 0.186 MeV for the measurement at 5.1-MeV incident neutron energy: The values are $7.05 \pm 0.10^{\text{stat}} \pm 0.72^{\text{syst}}$, $8.12 \pm 12^{\text{stat}} \pm 0.83^{\text{syst}}$, and $6.50 \pm 0.07^{\text{stat}} \pm 0.70^{\text{syst}}$, respectively, for the average multiplicity, and $0.84 \pm 0.01^{\text{stat}} \pm 0.03^{\text{syst}}$, $1.12 \pm 0.04^{\text{stat}} \pm 0.03^{\text{syst}}$, and $0.86 \pm 0.02^{\text{stat}} \pm 0.03^{\text{syst}}$, respectively, for the mean photon energy per fission. If we apply a threshold of 0.186 MeV, the multiplicity is $6.32 \pm 0.04^{\text{stat}} \pm 0.64^{\text{syst}}$, in 1.6-MeV neutron-induced fission, in quite good agreement with the value for 5.1-MeV incident neutrons. With this threshold, the mean photon energy is $0.92 \pm 0.01^{\text{stat}} \pm 0.03^{\text{syst}}$, which agrees well with the value for 5.1-MeV incident neutrons, too.

TABLE III. Mean values of multiplicity and photon energy of prompt fission γ rays in the fission of ${}^{238}\text{U}$ induced by 1.6-, 5.1-, and 15.0-MeV neutrons. The values calculated with the GEF code are also given. Given at 68% C.L., the statistical uncertainty appears in superscript, the systematic one in subscript.

E_n (MeV)	Threshold (MeV)	Multiplicity		Mean energy	
		This work	GEF	This work	GEF
1.6 ± 0.1	none		8.06		0.784
	0.11	$7.05^{0.10}_{0.72}$	7.55	$0.84^{0.01}_{0.03}$	0.821
	0.19	$6.32^{0.04}_{0.64}$	6.78	$0.92^{0.01}_{0.03}$	0.843
5.1 ± 0.2	none		8.59		0.806
	0.19	$6.50^{0.07}_{0.70}$	7.32	$0.86^{0.02}_{0.03}$	0.855
15.0 ± 0.6	none		9.76		0.855
	0.11	$8.12^{0.12}_{0.83}$	9.25	$1.12^{0.04}_{0.03}$	0.896
	0.19	$7.26^{0.12}_{0.74}$	8.50	$1.23^{0.05}_{0.04}$	0.913

The GEF code allows calculating the PFGS mean values with and without setting a low-energy threshold. The code gives spectra beginning around 50 keV, and the predicted multiplicities calculated without any threshold are systematically larger than the experimental values. With the experimental thresholds, they are about 10% larger than the experimental values. Without any threshold, the mean energies are systematically lower than the experimental values. By applying the experimental thresholds, the calculated multiplicities agree very well for 1.6- and 5.1-MeV neutron-induced fission; for 15.0-MeV incident neutrons, the predicted value is 20% lower than the experimental one, which reflects that the increase in amplitude in the high-energy range of the experimental spectrum is not well reproduced in the calculated one.

VI. CONCLUSION

For the first time prompt γ -ray emission was investigated experimentally in fast-neutron-induced fission of an actinide. The spectra were measured in the fission of ${}^{238}\text{U}$ induced by incident neutrons of 1.6 ± 0.1 , 5.1 ± 0.2 , and 15.0 ± 0.6 MeV. Mean values of energy and multiplicity were derived.

At 1.6- and 5.1-MeV incident neutron, the spectra agree well within 68% C.L. The spectrum measured at 15.0 MeV exhibits a much smoother slope above 2 MeV, such that the amplitude is higher by one order of magnitude around a prompt γ -ray energy of 7–8 MeV. For the first time, a significant difference is observed in the spectrum as a function of the incident neutron energy. Previous measurements for the system $n + {}^{235}\text{U}$ do not corroborate our findings.

Theoretical spectra calculated with the GEF and FIFRELIN codes agree rather well with the experimental data up to 5–6-MeV γ -ray energy. Above, the disagreement is substantial, up to one order of magnitude. The mean values obtained with GEF agree well with the experimental ones for 1.6- and 5.1-MeV incident neutrons. A small discrepancy for 15.0-MeV incident neutrons may be observed, which is consistent with the fact

that the GEF code does not reproduce the increased γ -ray yield above 2 MeV.

The BGO detector turned out to be an appropriate choice for such measurements, especially for the high-energy part of

the spectra. The unfolding of the experimental spectra from the detector response and the impact of the setup geometry may be considered above 0.7 MeV as a correction as low as 20–30%.

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