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Improved values for the characteristics of prompt-fission γ-ray spectra from the reaction ^{235}U($n_{\rm th}$, f)

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In this paper we present results from measurements of prompt γ rays from the thermal neutron induced fission of ^{235}U. Photons were measured in coincidence with fission fragments with cerium-doped LaCl$_3$ and LaBr$_3$ as well as CeBr$_3$ scintillation detectors, which offer an intriguing combination of excellent timing resolution and good resolving power. The spectra measured with all employed detectors are in excellent agreement with respect to their shapes. Characteristic parameters were extracted for a γ-energy range from 0.1 to 6.0 MeV and the results obtained with several detectors were averaged. From that, the average emission yield of prompt-fission γ rays was determined to be $\overline{\nu}_\gamma = (8.19 \pm 0.11)$ per fission, the average energy per photon to be $\epsilon_\gamma = (0.85 \pm 0.02)$ MeV, and the total energy to be $E_{\gamma,\rm tot} = (6.92 \pm 0.09)$ MeV. The uncertainties are much lower than the 7.5% requested for the modeling of advanced nuclear reactor cores. Estimating the influence of γ rays with energies between 6 and 10 MeV on the values determined in this work revealed a negligible deviation of the order of the found uncertainties.

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Introduction. In recent years the measurement of prompt-fission γ-ray spectra (PFGS) has gained renewed interest [1–3], after about forty years since the first comprehensive studies of the reactions ^{235}U($n_{\rm th}$, f) [4–6] and ^{252}Cf(sf) [4]. These new experimental efforts were motivated by requests for new values especially for γ-ray multiplicity and mean photon energy release per fission in the thermal neutron induced fission of ^{235}U and ^{239}Pu [7]. Both isotopes are considered the most important ones with respect to the modeling of innovative cores required for the fast Generation-IV reactors [8–10]. As a consequence of recent instrumental advancements like the development of new detectors as well as digital data acquisition systems, the determination of new and improved PFGS characteristics became possible with high precision. An example of that is a recent study on the PFGS from the spontaneous fission of ^{252}Cf (see Ref. [1] and references therein).

At the same time, new and advanced computer codes were developed both at CEA Cadarache [11,12] and by a Los Alamos–New York collaboration [13,14], which simulate prompt neutron and γ-ray emission from primary fission fragments by a Monte Carlo approach together with a full Hauser-Feshbach calculation. Both theoretical models not only describe the spectral shape of the continuous prompt γ-ray spectra well, but also reproduce the discrete γ peaks observed experimentally below 1 MeV photon energy (e.g. Ref. [1]). These very detailed features allow a serious comparison of the deduced PFGS characteristics such as photon multiplicity and the average and total photon energy per fission with experimentally found values, if experiments provide the same amount of detailed information as given by the calculations mentioned above. The recent work on ^{252}Cf showed that this is possible indeed [1]. Based on the experiences there, we performed such high precision measurements of thermal neutron induced prompt-fission γ rays from ^{235}U, which are reported below.

Experiments and data treatment. Two experiments were performed at the 10 MW research reactor of KFKI, Budapest, Hungary, in order to measure prompt-fission γ rays from the reaction $n_{\rm th}$+^{235}U. In both, a uranium sample was exposed to a cold neutron beam, and γ rays were measured in coincidence with fission fragments, although different instrumentation was used, as described below in more detail.

In the first measurement campaign that took place during ten days of beam time in February and March 2010, photons were detected with a 3 in. × 3 in. coaxial LaCl$_3$:Ce scintillation detector, located at a distance of about 30 cm from a very thin ^{235}U target with an effective mass of 113 μg, mounted on a 34 μg/cm^2 thick polyimide backing. The sample itself was placed in the fission fragment spectrometer VERDI [15,16]. The fast fission trigger was provided by a polycrystalline chemical vapor deposited (pcCVD) diamond detector of size 1 cm × 1 cm, placed directly on top of the uranium sample (distance approximately 1 mm). More information on artificial diamond detectors may be found in Ref. [17].

The second experiment was carried out in June 2012 with about six days of actual beam time. An ultra-thin spectroscopic

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uranium sample (thickness 91.5 $\mu g/cm^2$) of effective mass 640 μg was placed inside a twin Frisch-grid ionization chamber, which delivered the fission trigger. The coincident measurement of photons was accomplished with two coaxial $LaBr_3$:Ce as well as two coaxial $CeBr_3$ scintillation detectors simultaneously, whose size (diameter × length) is 2 × 2 and 1 × 2 in., respectively. The properties of the used lanthanide halide detectors are described in detail in Refs. [1,18–21].

In both measurements, pulse height and time of flight of the coincident events in each scintillation detector were recorded and stored in list mode. The first one gives, after proper calibration with different γ sources, the energy deposited in the detector, while the second enables us to discard all events that arrived later in time than expected from a prompt γ ray, e.g., due to neutron-induced reactions. For deducing the emitted prompt-fission γ-ray spectrum, the measured spectra have to be corrected with the response function of each detector. These response functions were determined by means of Monte Carlo simulations with the PENELOPE2011 computer code [22], folded with the energy resolution of the corresponding detectors. In Ref. [1], this as well as the actual extraction of the emission spectrum is described in detail, which is the reason why we restrict ourselves here to present only the resulting emission spectra and their characteristics.

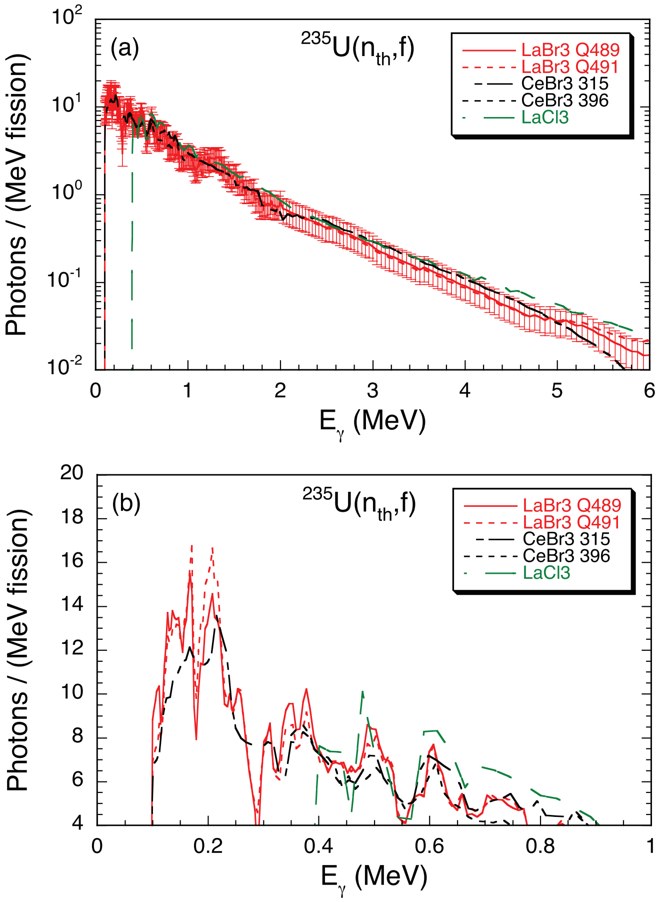

FIG. 1. (Color online) (a) Unfolded prompt-fission γ-ray emission spectrum from the reaction $n_{th}+^{235}U$, taken with five different detectors (see text for details). (b) Enhanced view on the low energy part, exhibiting a distinct peak structure in very good agreement between the results obtained with all detectors used in this work.

Results and discussion. According to the procedure sketched above, prompt-fission γ-ray spectra were obtained from the data taken with five different detectors. They are depicted in the upper part of Fig. 1, where the labels indicate both respective scintillator crystal and detector number. All energy distributions exhibit very good agreement with respect to both shape and absolute value over the entire energy region up to 6 MeV, as indicated by the error bars, which for the sake of clarity are shown only for one detector (Q489). However, it has to be mentioned that the spectrum for the $LaCl_3$:Ce detector from the first experiment was scaled down by a factor of 1.22 in height relative to the first $LaBr_3$:Ce detector (Q489), because there the precise assessment of the solid angle obviously was quite difficult due to the short distance of about 1 mm

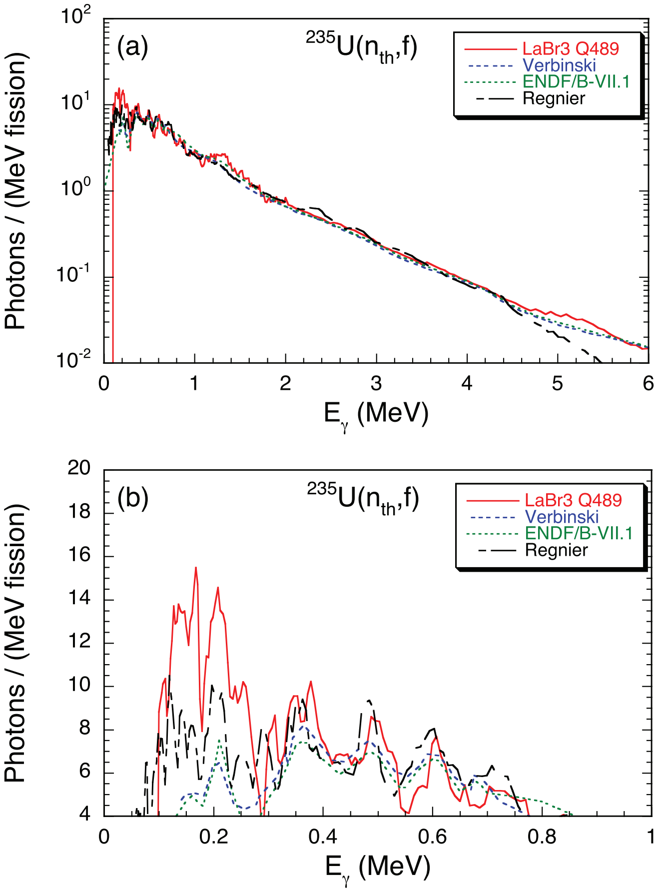

FIG. 2. (Color online) (a) The prompt-fission γ-ray emission spectrum from this work taken with one of the 2 × 2 in. $LaBr_3$:Ce detectors (full red line) is shown together with experimental data from Ref. [4] (dashed blue line) and data from the evaluated library ENDF/B-VII.1 [23] (dotted green line). Recent results from model calculations [11] are shown as well (dashed-dotted black line). In the high energy range, all spectra agree very well with each other. (b) The low energy part of the spectrum corroborates this impression, since the historical data from Ref. [4] and from the recent ENDF/B-VII.1 evaluation [23] are well reproduced by our data. The best description, however, with respect to the yields at very low energies is given by the calculations [11,24].

(see above) between the sample and the diamond detector. As a consequence, data from this detector were not taken into account when determining averaged values for the mean photon multiplicity as presented below. Figure 1(b) shows an enhanced view on the low energy part of the spectra above, exhibiting a distinct structure of discrete γ peaks. According to, e.g., Ref. [12], these peaks are due to deexcitations of secondary fission fragments along the yrast bands, taking place after the statistical part of dipole transitions, which leads to a continuous γ spectrum. This peak structure appears in all five spectra, despite different thresholds and even though the different energy resolutions of the detectors are reflected in the widths of the respective peaks. The agreement of the peak positions is excellent, hence confirming that the energy calibration had been performed correctly for all detectors.

In the following, we compare our results to other available data for the same fissioning system. For that we pick the spectrum obtained with one of the $LaBr_3$:Ce detectors (Q489) and display it in Fig. 2 together with the measured one from 1973 (the values were digitized from Ref. [4]). Also shown there is the evaluated data from ENDF/B-VII.1 [23] and the recent result [24] of calculations according the model described in Ref. [11]. The general impression is that the other data agree well with our measured spectrum in both shape and height, as visible in Fig. 2(a). Looking again in more detail at the region below 1 MeV, as shown in Fig. 2(b), this impression is corroborated. The historical data from Verbinski *et al.* [4] are in good agreement with ours in describing the observed discrete peak structure, taking the poorer energy resolution of the NaI detector used in that work into account. The values from the evaluated data tables agree also well with both our results and the ones in Ref. [4], the latter being not really surprising, since the evaluation most probably was made according to hitherto available experimental data, like the one in just that reference. However, the intensities for photons with energies below 300 keV are underestimated compared to our experimentally obtained spectra. The theoretical calculations from Ref. [24] succeed too in describing well the observed peak structure and they reproduce better the observed low energy yields, although they are also somewhat low compared to the experimental data. The individual peak positions seem to be shifted by about 10 keV, which could indicate that some fine-tuning of the model might be advisable. Nevertheless, from our point of view, the model presented in Ref. [11] seems to be a very promising tool for the prediction of PFGS.

Characteristic parameters for prompt-fission γ-ray emission, such as the average number of photons per fission as well as the mean and total energy, were obtained by integrating over the measured photon distributions. This was done for all detectors used in this work up to energies of 6 MeV. However, since different thresholds on the pulse height had been applied during the experiment to the individual detectors, different lower integration limits had to be used in some cases.

This threshold had been well below 100 keV for three of the detectors used in this work; hence, for them the integration was performed for the energy range 0.1–6.0 MeV. The characteristic parameters were determined and average values, weighted with the uncertainties, were calculated. One of the $CeBr_3$ detectors (396) had a threshold corresponding to below 300 keV, while it was around 450 keV for the $LaCl_3$:Ce detector. For those, the lower integration limits were chosen to be 300 and 500 keV, respectively. To be able to compare the results from these detectors with those from the other three, new integrations were performed for them starting from 300 and 500 keV and new average values were calculated. The ratios of the mean values for 100 keV to the ones from the higher lower limits were then applied to the corresponding detector. Hence, their covered energy ranges were extrapolated to the same as for the other detectors. Here, uncertainties for the averaged values were taken into account, explaining the somewhat larger error budget. We believe that this procedure is justified by the very similar shape of the spectra, independent of the particular detector with which the

TABLE I. Summary of prompt γ-ray characteristics for the neutron-induced fission of ^{235}U. Experimental results from this work for the average γ-ray multiplicity $\overline{\nu}_\gamma$, the average energy ϵ_γ, and the total energy $E_{\gamma,\mathrm{tot}}$, obtained with all five detectors employed in this work, are given and the covered energy range is indicated. Averaged values for the first three detectors are presented as well and compared to previously measured ones from Refs. [4–6] as well as corresponding numbers from the evaluated nuclear data files in ENDF/B-VII.1 [23] and from calculations from Refs. [14,24]. The results denoted by *) are calculated on the basis of the average values (see text for details).

Results	Detector	Diameter × length (in.)	$\overline{\nu}_\gamma$ (per fission)	ϵ_γ (MeV)	$E_{\gamma,\mathrm{tot}}$ (MeV)	Energy range (MeV)	
This work	$LaBr_3$:Ce (Q489)	2 × 2	8.25 ± 0.17	0.84 ± 0.02	6.94 ± 0.14	0.1–6.0	
This work	$LaBr_3$:Ce (Q491)	2 × 2	8.09 ± 0.23	0.84 ± 0.03	6.79 ± 0.18	0.1–6.0	
This work	$CeBr_3$ (315)	1 × 2	8.16 ± 0.22	0.86 ± 0.03	7.03 ± 0.19	0.1–6.0	
This work	$CeBr_3$ (396)	1 × 2	7.94 ± 0.78	0.86 ± 0.09	6.81 ± 0.99	0.1–6.0	*)
This work	$LaCl_3$:Ce	3 × 3	8.21 ± 0.41	0.85 ± 0.05	6.99 ± 0.35	0.1–6.0	*)
This work	Averaged values		8.19 ± 0.11	0.85 ± 0.02	6.92 ± 0.09	0.1–6.0	
Verbinski *et al.* [4]	NaI	2.3 × 6	6.70 ± 0.30	0.97 ± 0.05	6.51 ± 0.30	0.14–10.0	
Pleasonton *et al.* [5]	NaI	5 × 4	6.51 ± 0.30	0.99 ± 0.07	6.43 ± 0.30	0.09–10.0	
Peelle *et al.* [6]	NaI	1.75 × 1	7.45 ± 0.35	0.96	7.18 ± 0.26	0.14–10.0	
ENDF/B-VII.1 [23]	Evaluation		6.86	0.96	6.58	0.1–10.0	
Becker *et al.* [14]	Calculation		8.05	0.88	7.06	0.14–10.0	
Regnier *et al.* [24]	Calculation		7.900 ± 0.005	0.840 ± 0.001	6.64 ± 0.012	0–13.5	

spectrum was taken (see Fig. 1). All results are summarized in Table I, where the extrapolated values are indicated by an asterisk. They are compared to experimental results from the early 1970s [4–6] as well as to results from recent Monte Carlo Hauser-Feshbach calculations [14,24] and data from the evaluated library ENDF/B-VII.1 [23]. The results are also shown in Fig. 3, where the symbols denote the individual values, while mean values and their uncertainties are indicated by full drawn and dashed lines, respectively.

We notice that the results from all detectors employed in this work are in excellent agreement with each other and very well reproduced by the recent Monte Carlo Hauser-Feshbach calculations from Ref. [14]. The results from similar calculations, based on Ref. [11] and provided by Ref. [24], are a bit differing, although they describe well the measured PFGS (see Fig. 2). This is most probably explained by the fact that there also γ rays below 100 keV energy were taken into account, which is not the case for the other results. In contrast, the experimental results from the early 1970s [4–6] exhibit deviations that are larger than the uncertainties from this work. For those three experiments, the reported γ-ray multiplicities are consistently too low, while the mean energy per photon is too high. The total γ-ray energies are not all in agreement within their error bars and scattered around our mean value. The same discrepancies are observed for the present data in ENDF/B-VII.1 [23], which may be explained by the fact that the corresponding evaluation is based upon the historical data mentioned above.

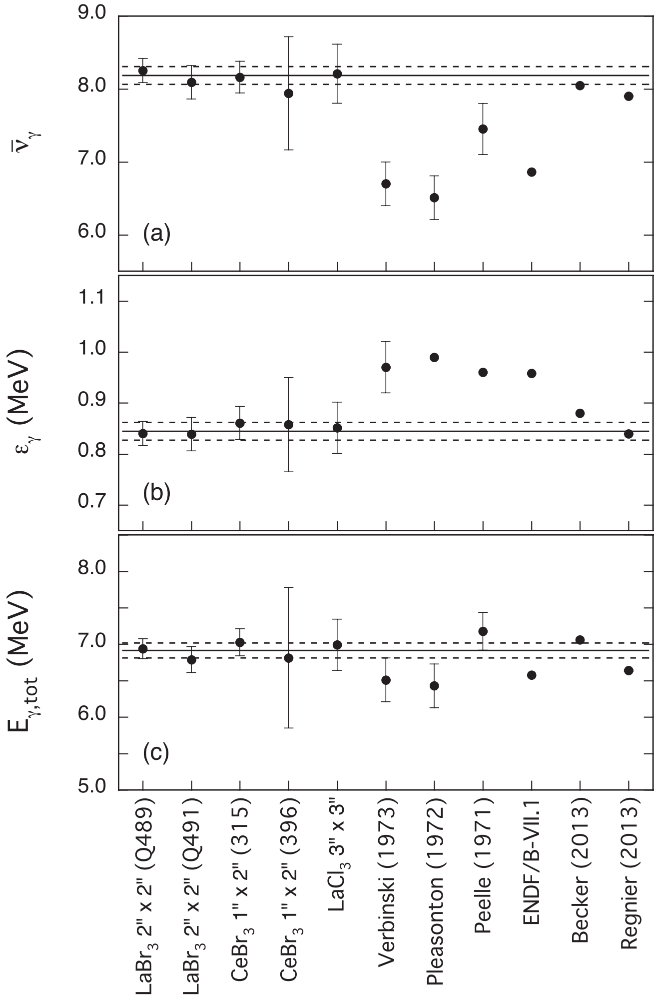

FIG. 3. Overview of results for the measurement of prompt γ-ray emission for the neutron-induced fission of ^{235}U: (a) Average photon multiplicity, (b) mean photon energy per fission, and (c) total released photon energy from this work are compared to experimental results from the early 1970s by Verbinski (1973) [4], Pleasonton (1972) [5], and Peelle (1971) [6], respectively, and values from ENDF/B-VII.1 [23] as well as results from recent Monte Carlo Hauser-Feshbach calculations by Becker (2013) [14] and Regnier (2013) [24]. Values averaged over the results obtained with the first three detectors and their uncertainties are displayed as full drawn and dashed lines, respectively.

The fact that the maximum photon energy considered in this work is 6 MeV and thus lower than the 10 MeV from previous studies cannot account for these differences, since this would cause corrections in the opposite direction. Nevertheless, we estimated anyway the impact of photons with energies between 6 and 10 MeV on our results presented above. For that purpose we fitted the emission spectrum taken with one of the LaBr$_3$:Ce detectors (Q489) between 3 and 6 MeV with an exponential function. The fit result was then integrated analytically within the limits 6 and 10 MeV, leading to a number of additional γ rays per fission of 1.24×10^{-2} and a surplus amount in total energy of 8.58×10^{-2} MeV. This difference is obviously of the order of the uncertainty of the total energy determined in this work and may therefore practically be disregarded. Hence, we are convinced that the comparison of our results with the others as done in Fig. 3 and Table I makes sense indeed.

Conclusions. In this work we have reported on the measurement of prompt fission γ-ray spectra (PFGS) from the reaction $n_{\mathrm{th}}+^{235}$U. Different lanthanide halide scintillation detectors were employed and have proven again (cf. Ref. [1]) that they constitute a well-suited choice of instrumentation for this kind of investigation. PFGS characteristics were determined with high precision, which meets by far the request for an uncertainty of 7.5% at most with respect to γ heating in advanced nuclear reactor core simulations [8–10]. This achievement was possible due to the excellent agreement between the emission spectra obtained with each of our detectors, which allowed averaging the individual results in order to reduce the uncertainties. In an upcoming experiment, dedicated to the measurement of PFGS from ^{241}Pu(n_{th}, f), we plan to use again several different lanthanide halide detectors simultaneously, like the ones reported about in this work. Although we have demonstrated that the influence of photons with energies above 6 MeV is practically negligible on the PFGS characteristics, we intend to employ then in addition a 4×4 in. bismuth germanate (BGO) scintillation detector in order to extend the energy range of detectable γ rays.

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