

Scientific Workshop on Nuclear Fission dynamics and the Emission of Prompt Neutrons and Gamma Rays, THEORY-3

Prompt fission γ -ray spectra characteristics - systematics and predictions

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Abstract

Revised systematics for prompt fission γ -ray spectra (PFGS) characteristics as function of both atomic and mass number of the compound system, derived from recent experiments on thermal-neutron induced and spontaneous fission, is presented and applied to fission induced by fast neutrons. Results from these calculations for $^{238}\text{U}(n, f)$ and $^{235}\text{U}(n, f)$ for incident neutron energies from 0 to 20 MeV are compared to new experimental results, exhibiting nice agreement. Very recent PFGS measurements for $^{240}\text{Pu}(sf)$ and $^{242}\text{Pu}(sf)$ have been evaluated and the determined PFGS characteristics are shown to also fit well with the systematics. From this we conclude that the obtained systematics, although purely empirical, is indeed a useful tool for the prediction of average total γ -ray energy released in prompt fission, mean energy per photon as well as average photon multiplicity for fissioning systems, which are difficult or even impossible to study experimentally.

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Peer-review under responsibility of the European Commission, Joint Research Centre – Institute for Reference Materials and Measurements

Keywords: Prompt fission γ -rays; total γ -ray energy; prompt γ -ray multiplicity; systematics; A and Z dependence; energy dependence; $^{235}\text{U}(n, f)$; $^{238}\text{U}(n, f)$; $^{240}\text{Pu}(sf)$; $^{242}\text{Pu}(sf)$

1. Introduction

In recent years the measurement of prompt-fission γ -ray spectra (PFGS) has gained renewed interest. After about forty years since the first (and at the same time last) comprehensive studies on this topic, the development of lanthanide halide scintillation detectors as well as new data acquisition and signal-processing techniques provided appropriate tools to determine PFGS characteristics, i.e. average total γ -ray energy released in prompt fission, mean energy per photon as well as average photon multiplicity, with unprecedented accuracy. These new experimental efforts were motivated by OECD/NEA requests for new values especially for gamma-ray multiplicities and mean photon energies,

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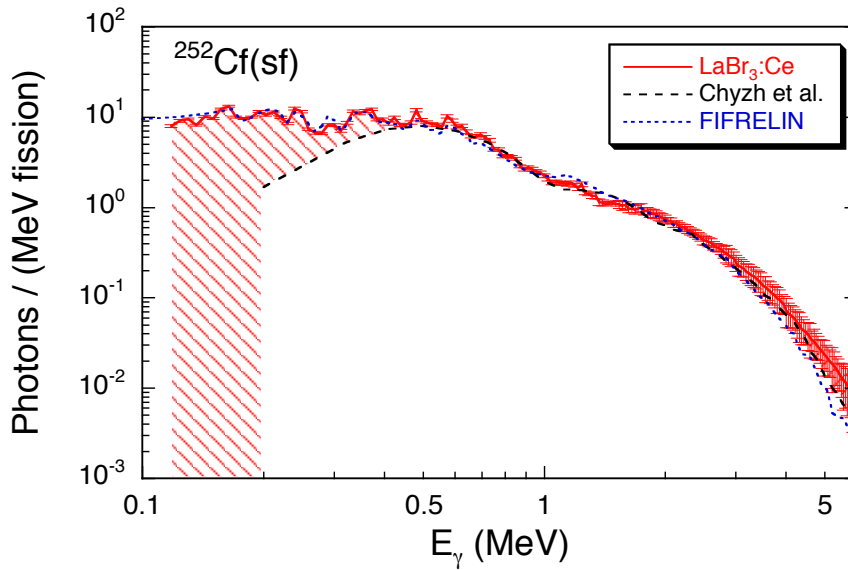


Fig. 1. (Color online) Comparison of measured PFGS from $^{252}\text{Cf}(\text{sf})$ from Billnert et al. (2013) and Chyzh et al. (2012), using the DANCE detector system (Heil et al., 2001), depicted as full (red) and dashed (black) line, respectively, together with the result of calculations with the Hauser-Feshbach Monte Carlo code FIFRELIN (Regnier et al., 2013), provided by Regnier (2013) (blue dotted line). The (red) hatched areas below 500 keV and above 4 MeV indicate the missing photon yield in the DANCE spectrum of the amount of 30%.

in particular for $^{235}\text{U}(\text{n}, \text{f})$ and $^{239}\text{Pu}(\text{n}, \text{f})$ (NEA, 2006). Both target isotopes are considered the most important ones with respect to the modeling of innovative cores for fast Generation-IV reactors (Rimpault et al., 2012).

Based on recent experimental results from the reactions $^{235}\text{U}(\text{n}_{\text{th}}, \text{f})$ (Oberstedt A. et al., 2013) and $^{241}\text{Pu}(\text{n}_{\text{th}}, \text{f})$ (Oberstedt S. et al., 2014) as well as from the spontaneous fission of ^{252}Cf (Billnert et al., 2013; Oberstedt A. et al., 2015), we have recently presented a revised version of systematics for PFGS characteristics as function of both atomic and mass number of the compound systems (Oberstedt A. et al., 2014a), originally established already in 2001 (Valentine, 2001). Although corresponding PFGS measurements were carried out also with the DANCE detector system at Los Alamos National Laboratory (Heil et al., 2001) during the last couple of years, those results are not taken into account for the systematics, because of grave underestimations of especially low-energy photons, as depicted in Fig. 1. This leads obviously to severe differences when determining PFGS characteristics, which is discussed in more detail by Oberstedt A. et al. (2015). Nevertheless, this systematics allows estimating gamma-ray multiplicity, mean and total photon energy in cases, where target nuclei are not available or accessible experimentally. While this has been done before for thermal-neutron induced and spontaneous fission, we show in this work how PFGS characteristics may even be predicted for fission induced by fast neutrons. Below we give examples for $^{238}\text{U}(\text{n}, \text{f})$ and $^{235}\text{U}(\text{n}, \text{f})$ and compare predicted PFGS characteristics with values obtained in both recent experiments and model calculations. Moreover, preliminary results for $^{240}\text{Pu}(\text{sf})$ and $^{242}\text{Pu}(\text{sf})$ are presented and compared to the systematics too.

2. $^{238}\text{U}(\text{n}, \text{f})$ PFGS characteristics – experiments and predictions

From the revised systematics presented by Oberstedt A. et al. (2014), PFGS properties were inferred for the fissioning system $\text{n} + ^{238}\text{U}$ in the incident neutron energy range $E_n = 0$ to 20 MeV. The results are denoted as prediction and depicted in Fig. 2. The upper part shows the predicted average total γ -ray energy released in fission as function of incident neutron energy together with a linear fit to an empirical approach from Madland (2006). Part of this has recently been presented by Oberstedt A. et al. (2014a), where also the result of a FIFRELIN (Regnier et al., 2013) calculation at $E_n = 1.8$ MeV was shown (Litaize et al., 2014a), here indicated by a (blue) open circle. The (green) squares and triangles represent results from calculations by Tudora (2013) in the framework of the Point-by-Point

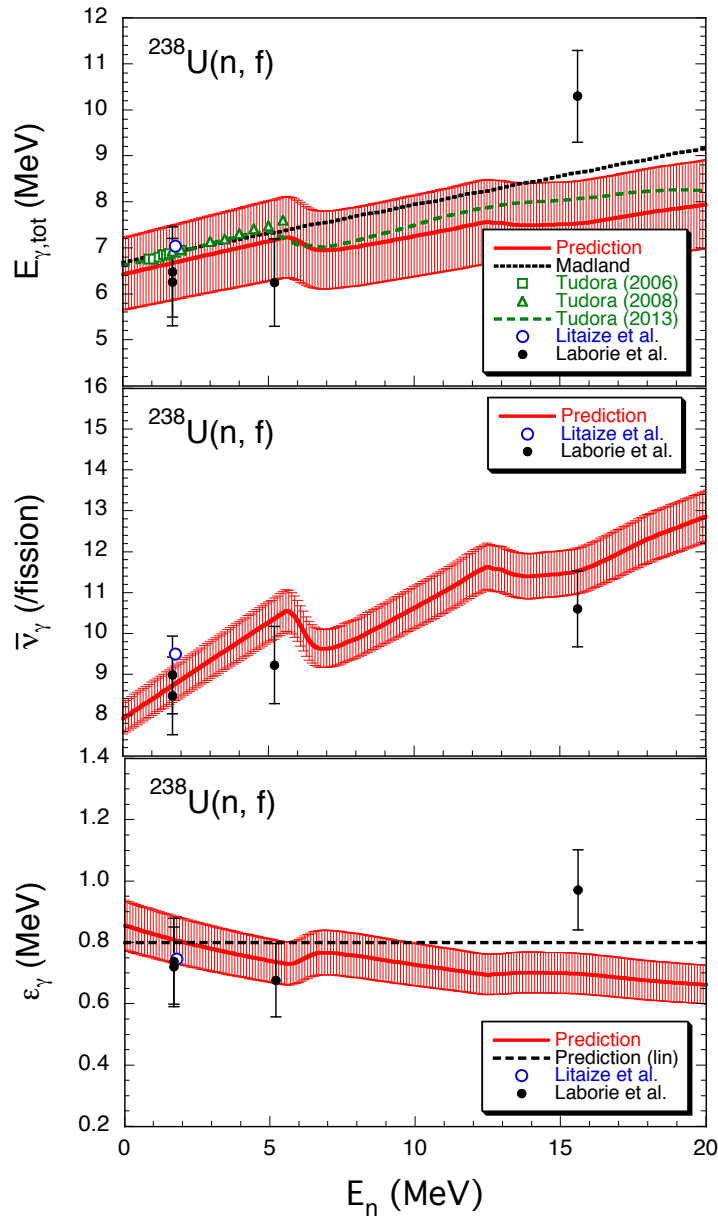


Fig. 2. (Color online) Prediction of PFGR characteristics, i.e. average total γ -ray energy (upper part), average photon multiplicity (middle part) as well as mean energy per photon (lower part), for the reaction $^{238}\text{U}(n, f)$ as function of incident neutron energy (full red lines with error bars) compared to corresponding experimental results (symbols) and results from different model calculations (lines); see text for details.

model (for details see e.g. in Tudora (2013a) and references therein). The (green) dashed line indicates results from the most recent calculations based on the same model (Tudora, 2013), however with model parameters used by Tudora et al. (2012). Experimental results are scarce for this fissioning system, but two measurements were reported at $E_n = 1.7$ and 15.6 MeV by Laborie et al. (2012). In the meantime two more experiments were performed at $E_n = 1.7$ and 5.2 MeV and the data analysis is in progress. Preliminary results were provided by Laborie (2014), which are also shown in Fig. 2 as full (black) circles. The middle part of Fig. 2 shows our prediction for the average prompt fission γ -ray multiplicity as function of incident neutron energy. One theoretical value is available (Litaize et al., 2014),

again depicted as (blue) open circle, and the preliminary experimental results from Laborie (2014) (full black circles) could be shown as well for comparison. The lower part of Fig. 2 finally contains our predictions for the average γ -ray energy per photon as function of incident neutron energy, obtained by dividing the predicted values depicted in the upper part with those in the middle part. The model predicts a constant value (Oberstedt A. et al., 2014), which is indicated by the (black) dashed line. Again a theoretical result from Litaize et al. (2014) and preliminary experimental results from Laborie (2014) are shown as open (blue) and full (black) circles, respectively. They too were obtained by dividing the values in the upper part of Fig. 2 with the corresponding ones in the middle.

Obviously, the agreement between our predicted PFGS characteristics and the results from the model calculations is excellent, in particular with respect to the kinks apparent at the thresholds for second and third chance fission. These are a consequence of taking into account – and subtract – pre-fission neutron emission in order to properly assess the energy dependence of prompt fission particle emission, with the prompt fission neutron multiplicities $\bar{\nu}_n(E)$ taken from the evaluated library ENDF/B-VII.1 (2011). A detailed explanation is given by Oberstedt A. et al. (2014) and will be the subject of an upcoming paper (Oberstedt A. et al., 2015a). In contrast, this effect has not been considered in the approximation by Madland (2006), hence the linear energy dependence. The few experimental values, although very preliminary according to Laborie (2014), are in reasonable agreement with our predictions. Still, a correct judgement will have to wait until the data analysis has been finished. This applies as well to data from a recent experiment performed at the novel directional neutron source LICORNE (Lithium Inverse Cinematiques ORsay NEutron source) recently installed at IPN Orsay (Lebois et al., 2014), which will be described below in more detail.

3. $^{235}\text{U}(n, f)$ PFGS characteristics – experiments and predictions

In July 2013 a first experiment with LICORNE was conducted over a period of two weeks, split into two parts with about 100 h of beam time each. The aim was to measure PFGS from the target nuclei ^{232}Th , ^{235}U and ^{238}U . In the first part thin targets of ^{235}U and ^{238}U of 10 mg approximate mass each were placed back-to-back at the central cathode position inside a cylindrical twin Frisch-grid ionization chamber. The counting gas was P10 (90% argon, 10% methane), providing a detection efficiency for fission fragments of almost 100%. In coincidence with the fission fragments, γ -rays were measured with 14 hexagonal BaF_2 scintillation detectors of 62 kg scintillator material in total, configured into two independent clusters of seven detectors, as well as with three coaxial $\text{LaBr}_3\text{:Ce}$ scintillation detectors of size 5.08 cm \times 5.08 cm. The second part of the experiment involved the same two BaF_2 clusters in a close packed geometry around thick samples of ^{238}U (38 g) and ^{232}Th (50 g), forming a calorimeter with a geometric efficiency of approximately 70%. The ionization chamber had been removed. Neutron beams were pulsed at 2.5 MHz rate, corresponding to 400 ns between bunches, and a bunch width of around 2 ns. This allowed timing information from the beam buncher to be used as a reference with which to measure event detection times relative to the bunch. Fission events can be discriminated from background by looking for high sum-energy and multiplicity events in the calorimeter that occurred within a short time window. More technical information about this experiment is given by Lebois et al. (2014a) and Wilson et al. (2015). From there it is obvious that the neutrons, produced in an inverse $p(^7\text{Li}, ^7\text{Be})n$ reaction, had an average energy $\bar{E}_n = 1.5$ MeV. Below we focus on the PFGS measurement from $^{235}\text{U}(n, f)$, performed during the first part of the experiment, and first data taken with the $\text{LaBr}_3\text{:Ce}$ detectors.

In order to extract an emission spectrum from the measured one, the response function of the detector(s) must be determined and unfolded. However, the usual procedure by means of Monte Carlo simulation of mono-energetic γ -rays, taking into account the geometrical efficiency and the experimental setup and adjusting the resulting spectra to the measured spectrum (details are given by Billnert et al. (2013)), is not possible here due to poor statistics. Instead we followed a different approach, which is motivated by the fact that the detectors in use were of exactly the same type as those employed before in a similar geometry in the PFGS measurement from $^{235}\text{U}(n_{th}, f)$, and because the measured spectra from that experiment (Oberstedt A. et al., 2013) and this work exhibit the same shape. From that experiment a properly unfolded PFGS had been extracted previously (Oberstedt A. et al., 2013). This spectrum may also be deduced from the measured one by taking into consideration the geometrical efficiency ϵ_{geom} and the number of fission events $N_{fission}$ according to

$$ES(E_{\gamma,i}) = TF(E_{\gamma,i}) \times MS(E_{\gamma,i}) / \epsilon_{geom} / N_{fission} \quad (1)$$

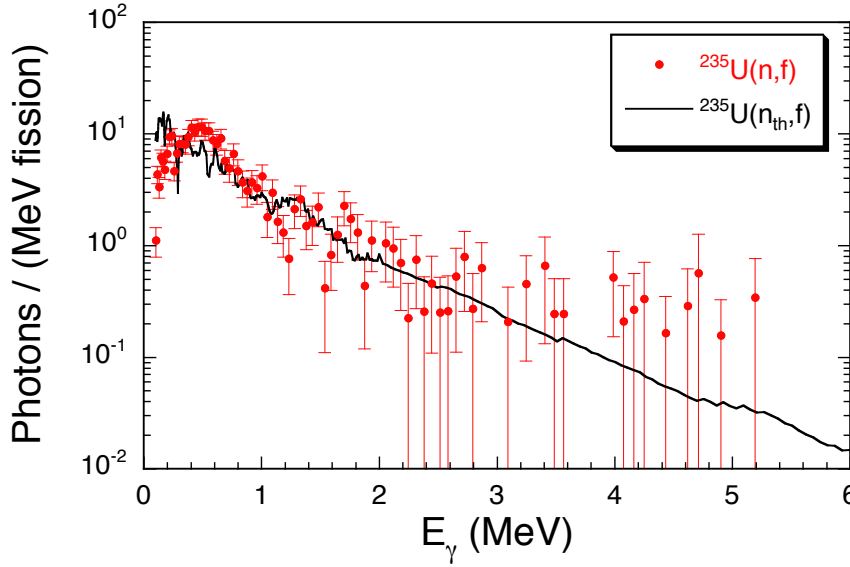


Fig. 3. (Color online) Prompt γ -ray spectrum for $^{235}\text{U}(n, f)$ at $\bar{E}_n = 1.5$ MeV depicted as full (red) circles. It is compared to a corresponding spectrum for $^{235}\text{U}(n_{th}, f)$ from Oberstedt A. et al. (2013), shown as thin (black) line.

Here, $ES(E_{\gamma,i})$ denotes the emission spectrum, $MS(E_{\gamma,i})$ the measured one and $TF(E_{\gamma,i})$ a transformation function. The latter was determined for the experiment on $^{235}\text{U}(n_{th}, f)$ and eventually applied to the measured spectrum taken at LICORNE. Finally, we obtain an emission spectrum as depicted in Fig. 3. However, the absolute values had to be adjusted due to loss of photon events during the measurement. But from recent systematics from Oberstedt A. et al. (2014), we know how the average prompt fission γ -ray multiplicity $\bar{\nu}_\gamma$ should depend on the prompt fission neutron multiplicity $\bar{\nu}_n$ and, hence, on the incident neutron energy. Hence, from the recently published multiplicity for thermal-neutron induced fission of the same system, $\bar{\nu}_\gamma(therm) = 8.19 \pm 0.11$ (Oberstedt A. et al., 2013), we deduce

$$\bar{\nu}_\gamma(1.5\text{MeV}) = \bar{\nu}_\gamma(therm) + [(16.6 \pm 0.5) - (11.0 \pm 0.4) \times 10^{-2} \times Z^{5/3} A^{-1/2}] \times [\bar{\nu}_n(1.5\text{MeV}) - \bar{\nu}_n(therm)] \quad (2)$$

$$= 8.69 \pm 0.43$$

where $Z = 92$ and $A = 236$ for the fissioning system and $\bar{\nu}_n(therm) = 2.421$ as well as $\bar{\nu}_n(1.5\text{MeV}) = 2.578 \pm 0.018$ (ENDF/B-VII.1, 2011) was used. The latter is the mean value of the tabulated multiplicities for 1.4 and 1.6 MeV and its uncertainty represents the standard deviation. The emission spectrum, adjusted as outlined above, is shown as full (red) circles in Fig. 3. The error bars include the statistical uncertainties from the measured spectrum, uncertainties in the transformation function as well as the uncertainty of the assumed multiplicity from Eq. 2. For comparison the PFGS for the thermal-neutron induced fission of the same compound system is shown as black line. We observe an excellent agreement, at least above 300 keV, which is no surprise rather than a confirmation that using the transformation function in this case works well. The reason for this discrepancy will be explained elsewhere. For now we restrict ourselves to stating that this had been corrected for, leading to an average energy per photon $\bar{\epsilon}_\gamma = (0.85 \pm 0.07)$ MeV. Together with an average multiplicity $\bar{\nu}_\gamma = 8.7 \pm 0.4$, derived from the systematics according to Eq. 2, we obtain an average total γ -ray energy release per fission $\bar{E}_{\gamma,tot} = (7.4 \pm 0.7)$ MeV. Although only part of all experimental data could have been considered so far, the deduced preliminary results on PFGS characteristics are compared to other ones, from both experiments and calculations. For this fissioning system we also predicted PFGS characteristics for fast-neutron induced fission, based on the same systematics as used in the previous section.

Figure 4 contains the predictions of PFGS properties for $n + ^{235}\text{U}$ in the incident neutron energy range $E_n = 0$ to 20 MeV. As shown in the previous section for $^{238}\text{U}(n, f)$, the upper part shows the average total γ -ray energy released in fission, the middle part the average photon multiplicity and the lower part the mean energy per photon. Our recent

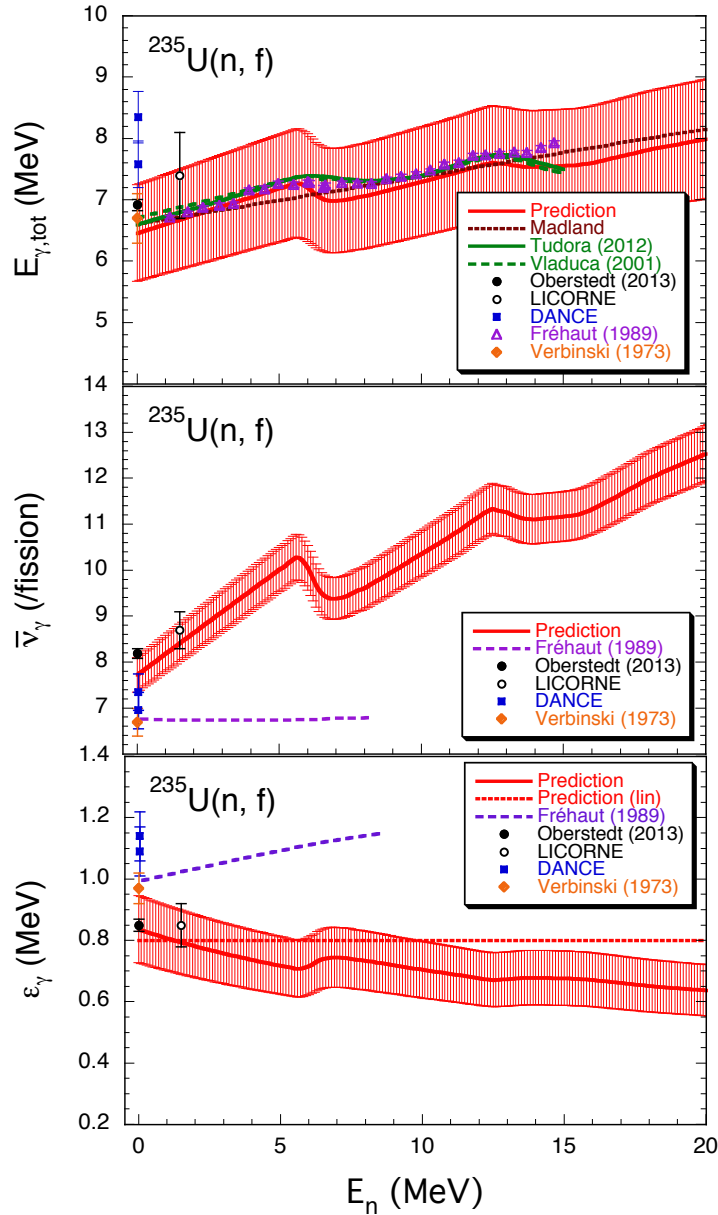


Fig. 4. (Color online) Prediction of PFSG characteristics, i.e. average total γ -ray energy (upper part), average photon multiplicity (middle part) as well as mean energy per photon (lower part), for the reaction $^{235}\text{U}(n, f)$ as function of incident neutron energy (full red lines with error bars) compared to corresponding experimental results (symbols) and results from different model calculations (lines); see text for details.

experimental result at $\bar{E}_n = 1.5$ MeV is depicted as open black circle, other results on fast-neutron induced fission from Fréhaut (1989) are indicated as open (purple) triangles (upper part only). The long dashed (purple) lines in the middle and lower part correspond to evaluations from the same author. The upper part of Fig. 4 shows also results from several model calculations for the average total γ -ray energy. The values from Vladuca et al. (2001) and Tudora et al. (2012a) are denoted as long dashed and full drawn (green) lines, while the linear empirical approach from Madland (2006) is indicated by a short dashed (brown) line. Again, our predictions agree very well with the cited calculations, apart from the fact that the approach from Madland (2006) does not take into account higher chance fission and, hence,

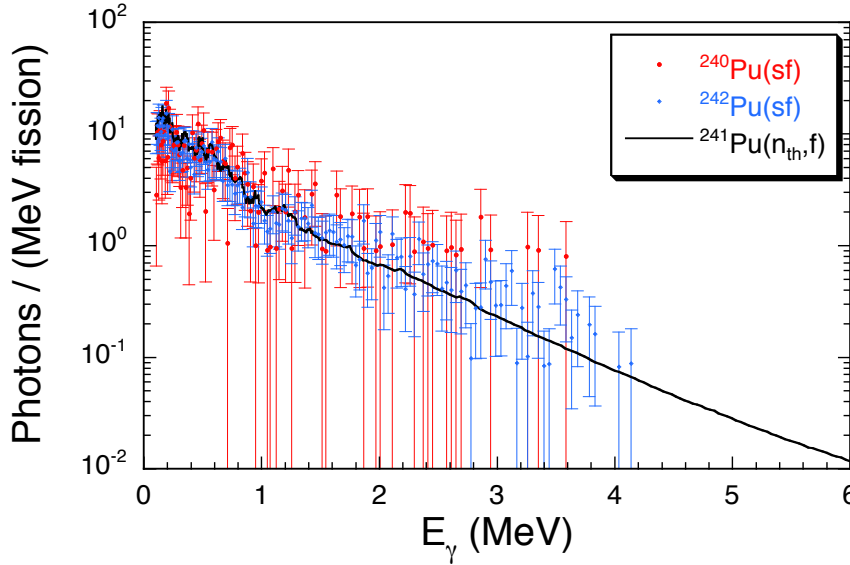


Fig. 5. (Color online) Prompt γ -ray spectra for $^{240}\text{Pu}(\text{sf})$ and $^{242}\text{Pu}(\text{sf})$, depicted as red and blue dots. It is compared to a corresponding spectrum for $^{241}\text{Pu}(n_{\text{th}}, \text{f})$ from Oberstedt S. et al. (2014), shown as thin (black) line.

does not exhibit the thresholds for second and third chance fission. The experimental results from both LICORNE (this work) and Fréhaud (1989) are also reproduced well. For comparison, data from thermal-neutron induced fission from Oberstedt A. et al. (2013), Chyzh et al. (2013, 2014) and Verbinski et al. (1973) are shown as well, as full (black) circles, (blue) squares and (orange) diamonds, respectively. The agreement is good, except for the results obtained with DANCE (Chyzh et al., 2013, 2014). However, we remind that these deviations have already been addressed and explained with the lack of low-energy photons in the detected PFGS (cf. Fig. 1 in Sect. 1).

4. Experimental results from $^{240,242}\text{Pu}(\text{sf})$

Recently, PFGS were measured from the spontaneous fission of both $^{240}\text{Pu}(\text{sf})$ and $^{242}\text{Pu}(\text{sf})$. The experimental setup was identical to the one used in a previous experiment on $^{241}\text{Pu}(n_{\text{th}}, \text{f})$, as described by Oberstedt S. et al. (2014), i.e. photons were measured with a coaxial $\text{LaBr}_3:\text{Ce}$ scintillation detector, this time of size $7.62 \text{ cm} \times 7.62 \text{ cm}$, in coincidence with fission fragments. The fission trigger was provided by a cylindrical twin Frisch-grid ionization chamber. The $\text{Pu}(\text{OH})_4$ samples of mass $92.9 \mu\text{g}$ ^{240}Pu and $671 \mu\text{g}$ ^{242}Pu , respectively (Salvador-Castiñeira et al., 2013), were placed on each side of the central cathode, which allowed the simultaneous detection of fission fragments from both isotopes. Prompt photons were selected within a time-of-flight range of $\pm 5.25 \text{ ns}$ with respect to the prompt peak, which ensured a good suppression of photons created in other reactions than fission. To date, only part of this data has been analyzed, corresponding to about one week of measurement. As a consequence of this, together with the long half-lives of both isotopes for spontaneous fission (Salvador-Castiñeira et al., 2013), the measured spectra suffer from rather low statistics. Therefore, emission spectra were deduced according to the method applied before on the data from $^{235}\text{U}(n, \text{f})$ at $\bar{E}_n = 1.5 \text{ MeV}$ (cf. Sect. 3), however with a transformation function determined from the measurement on $^{241}\text{Pu}(n_{\text{th}}, \text{f})$ (Oberstedt S. et al., 2014). The resulting PFGS are shown in Fig. 5, with the red and blue dots denoting ^{240}Pu and ^{242}Pu , respectively. For comparison, the PFGS from $^{241}\text{Pu}(n_{\text{th}}, \text{f})$ is depicted as black line. The deduced characteristics are

- for $^{240}\text{Pu}(\text{sf})$: $\bar{E}_{\gamma, \text{tot}} = (6.9 \pm 0.7) \text{ MeV}$, $\bar{\nu}_{\gamma} = 7.7 \pm 0.5$, $\bar{\epsilon}_{\gamma} = (0.9 \pm 0.1) \text{ MeV}$
- for $^{242}\text{Pu}(\text{sf})$: $\bar{E}_{\gamma, \text{tot}} = (6.9 \pm 0.3) \text{ MeV}$, $\bar{\nu}_{\gamma} = 7.7 \pm 0.4$, $\bar{\epsilon}_{\gamma} = (0.89 \pm 0.06) \text{ MeV}$

Below these results are compared with the previously mentioned systematics for PFGS characteristics.

5. Summary and conclusions

Above we have presented predictions for PFGS characteristics from fast-neutron induced fission, based on empirically found systematics as function of both atomic and mass number of the fissioning system. The systematics in turn is based on recently obtained results from measurements on $^{235}\text{U}(\text{n}_{\text{th}}, \text{f})$ (Oberstedt A. et al., 2013), $^{241}\text{Pu}(\text{n}_{\text{th}}, \text{f})$ (Oberstedt S. et al., 2014) and $^{252}\text{Cf}(\text{sf})$ (Billnert et al., 2013; Oberstedt A. et al., 2015). Figure 6 gives an overview of all experimental results for $E_{\gamma, \text{tot}}(\bar{\nu}_n, Z, A)$, $\bar{\nu}_\gamma(\bar{\nu}_n, Z, A)$ and $\epsilon_\gamma(Z, A)$ in the upper, middle and lower part, respectively. The depicted presentations were chosen according to the work of Valentine (2001). The full drawn (black) lines correspond to his evaluation, based on experimental results that were reported until 1973, denoted by full drawn (black) circles (the complete list of references is given by Oberstedt A. et al. (2014)). The (blue) open squares indicate the recent results obtained by using the DANCE detector system (Chyzh et al., 2012, 2013; Ullman et al., 2013). Our results, also recently published by Billnert et al. (2013); Oberstedt A. et al. (2013, 2015); Oberstedt S. et al. (2014), are shown as (red) open circles. The values for $\bar{\nu}_n$ were taken as given in Valentine (2001). Due to obvious discrepancies between the historical and the recently obtained experimental data, a new evaluation seems to be reasonable on the basis of these new results. However, even those exhibit considerable differences as mentioned above, depending on by which experimental group they were obtained. An explanation has been given in Sect. 1. Hence, only values from our previous work were included in a new evaluation, whose result is depicted by (red) dashed lines in Fig. 6. They were obtained by least-squares fits to our experimental results mentioned above, weighted with the uncertainties, which leads to the following description of the average total γ -ray energy released in fission in MeV

$$E_{\gamma, \text{tot}}(\bar{\nu}_n, Z, A) = [(3.02 \pm 0.21) - (1.54 \pm 0.15) \times 10^{-5} \times Z^2 A^{1/2}] \times \bar{\nu}_n + 4.0 \quad , \quad (3)$$

and the average energy per photon in MeV

$$\epsilon_\gamma(Z, A) = (0.80 \pm 0.41) - (0.00 \pm 0.22) \times 10^2 \times Z^{1/3} A^{-1} \quad , \quad (4)$$

while the average prompt fission γ -ray multiplicity may be approximated by

$$\bar{\nu}_\gamma(\bar{\nu}_n, Z, A) = [(16.60 \pm 0.52) - (10.98 \pm 0.40) \times 10^{-2} \times Z^{5/3} A^{-1/2}] \times \bar{\nu}_n \quad . \quad (5)$$

Although the fit parameters are afflicted with considerable uncertainties, basically due to the fact that only few experimental results could have been considered for the new evaluation, the differences compared to the work of Valentine (2001) are quite obvious. In addition, the recent preliminary results from the fast-neutron induced fission of ^{235}U and ^{238}U from the previous sections are shown as well in Fig. 6 as full (orange) diamonds. The corresponding values of $\bar{\nu}_n$ at a given neutron energy were taken from the evaluated library ENDF/B-VII.1 (2011). Within the certainly still considerable uncertainties one may observe a reasonable agreement with the revised evaluation. This is also true for the new preliminary results from the spontaneous fission of ^{240}Pu and ^{242}Pu , represented by open (orange) diamonds.

From this we conclude that the new systematics, which was originally found for thermal-neutron induced and spontaneous fission (Valentine, 2001) and revised with up-to-date experimental results, may as well be applied to fission induced by fast neutrons as long as the corresponding prompt fission neutron multiplicities are known and correctly used. Hence, we consider it a useful tool for the prediction of average total γ -ray energy released in prompt fission, mean energy per photon as well as average photon multiplicity for fissioning systems, which are difficult or even impossible to investigate experimentally.

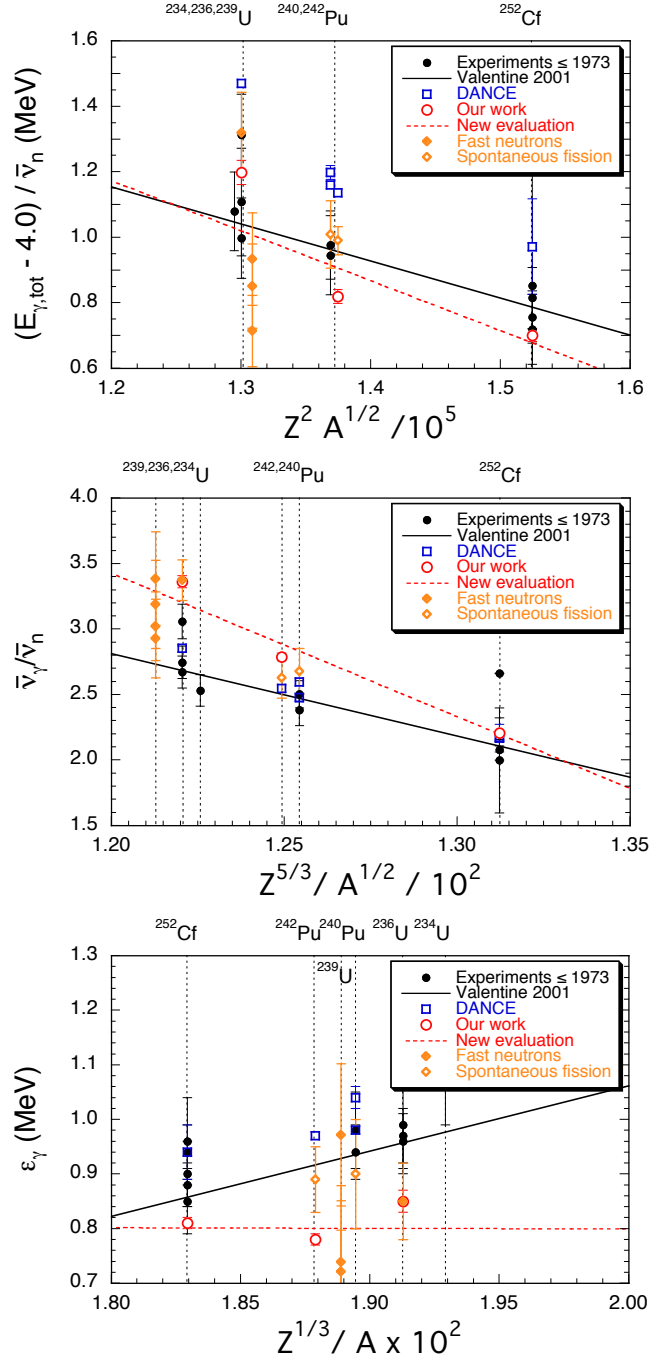


Fig. 6. (Color online) Overview of experimental results for the average total γ -ray energy released in fission (upper part), average prompt fission γ -ray multiplicity (middle part) and mean energy per photon (lower part) as function of A and Z for different fissioning systems. Full (black) circles denote historical results, open (blue) squares indicate results obtained with DANCE and open (red) circles represent results from our previous work (see Oberstedt A. et al. (2014) for detailed list of references). Also shown are results from evaluations by Valentine (2001) (solid black line) and from our work (dashed red line), based on the historical data and our previous results, respectively. In addition, recent results from fast-neutron induced (full (orange) diamonds) and spontaneous (open (orange) diamonds) fission mentioned in this work are depicted as well. For the sake of clarity, the corresponding fissioning systems are given, too.

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