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# New prompt fission $\gamma$-ray spectral data and its implication on present evaluated nuclear data files

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**Abstract**

In this paper we report on new spectral prompt $\gamma$-ray measurements from the spontaneous fission of $^{252}$Cf and thermal neutron-induced fission of $^{236}$U$^*$. In both experiments, $\gamma$-ray multiplicities, average and total $\gamma$-energies were extracted. Apart from one recent measurement on $^{252}$Cf, about four decades have passed since the last dedicated experiments were reported in literature. Hence, there was a need for a revision, not only with respect to high priority nuclear data requests by the Nuclear Energy Agency (NEA). In the first mentioned experiment we have measured prompt fission $\gamma$-rays with both cerium-doped $LaBr_3$ and $CeBr_3$ scintillation detectors, which both exhibit excellent timing and good energy resolution. The results from both detectors are in excellent agreement with each other and confirm the historical data. In the experiment on $^{235}$U(n$_{th}$,f) we employed cerium-doped $LaCl_3$ detectors, together with the lanthanum bromide detectors mentioned above. Even here the first results indicate a good agreement with data from the early 1970's. They are also in accordance with data in evaluated libraries like ENDF/B-VII.0, while this is not the case for $^{252}$Cf(SF). Hence, here an update is strongly recommended.


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*Keywords:* Prompt fission $\gamma$-rays; total $\gamma$-ray energy; prompt $\gamma$-ray multiplicity; Generation IV; nuclear energy; lanthanum bromide; cerium bromide; lanthanum chloride; scintillation detector; $^{252}$Cf(SF); $^{235}$U(n$_{th}$,f)
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## 1. Introduction

With the potential of more advanced nuclear reactors in the near future, a better understanding of the entire fission process is needed. Since four out of six of the impending Gen-IV reactors that have been selected by the Generation-IV International Forum (GIF) are fast reactors, an innovative core design is required to be able to handle the excessive heat deposit from the fission process. In order to model these cores, a better understanding of the released heat from

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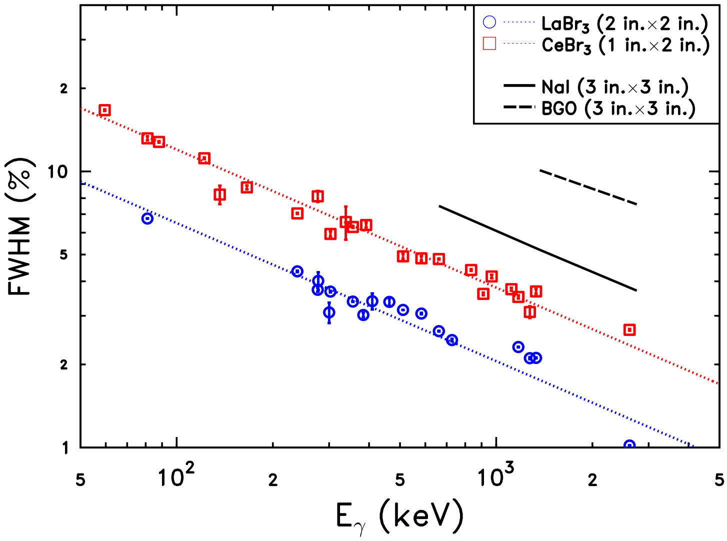


Fig. 1. Energy resolution (FWHM) in percent as a function of $\gamma$-ray energy for a 2 in. × 2 in. $LaBr_3$:Ce (circles) and a 1 in. × 2 in. $CeBr_3$ (squares) detector; our data is compared to a standard NaI detector as well as a BGO detector (Knoll, 2000).

the common reactor isotopes is crucial. Present knowledge regarding this heat deposit implies that approximately ten percent of the total energy released in fission is due to $\gamma$-rays, of which around 40 percent of the heat originates from prompt fission $\gamma$-rays (Krane, 1988). According to (Rimpault, 2006; Rimpault et al., 2006) it is necessary to achieve an uncertainty of at most 7.5% in regard of the $\gamma$-heating in order to adequately model these cores. However, with evaluated data the $\gamma$-heating is underestimated with up to 28% for the neutron-induced fission of the main reactor isotopes $^{235}$U and $^{239}$Pu. Therefore, these two isotopes have been included in NEA's high priority list for prompt fission $\gamma$-rays data, in particular new values for $\gamma$-multiplicity and mean energy are requested (NEA, 2006). The data in the evaluated data tables for both isotopes relies on results that were measured in the early 1970's (Verbinski et al., 1973; Pleasonton et al., 1972), and was recently confirmed (Kwan et al., 2012). Hence, it might be more likely that the underestimation comes from the reactions $^{238}$U(n,f) and $^{241}$Pu(n,f), involving isotopes that are always produced in a reactor (Sérot, 2011). The evaluated data for those two isotopes exhibit exactly the same structure, with an individual scaling factor; and the same formula is also used for $^{252}$Cf. Accordingly, it seems that no experimental data has been used to evaluate neither of these three isotopes. Since the 1970's a lot has happened in regard of detector development, especially with the release of new lanthanide-halide scintillation detectors (Billnert et al., 2011, 2012; Oberstedt A. et al., 2012, 2013) and references therein, as well as of data acquisition and signal processing techniques (Al-Adili et al., 2010, 2012). Consequently, we wanted to take advantage of these advancements towards high-quality measurements of the $\gamma$-decay heat from the fission process. In order to make an independent verification of the historical data, we performed an experiment on measuring prompt $\gamma$-rays from the neutron-induced fission of $^{236}$U*, and we also plan an identical measurement on $^{241}$Pu, in order to investigate, whether this isotope is the source of the underestimation mentioned above. In order to be able to accurately measure these isotopes, we needed to be certain of the quality of our experimental setup as well as the technique to extract results. Therefore, we started with studying the spontaneous fission of $^{252}$Cf. Since this reaction was measured in the early 1970's (Verbinski et al., 1973; Pleasonton et al., 1972) as well as very recently at LLNL (Chyzh et al., 2012), it serves as excellent proof of principle. After having obtained this confirmation, we carried on to analyze the measured prompt $\gamma$-ray spectra from the reaction $^{235}$U($n_{th}$,f). The results from the recent spectral data measurements on both systems are presented in this work.

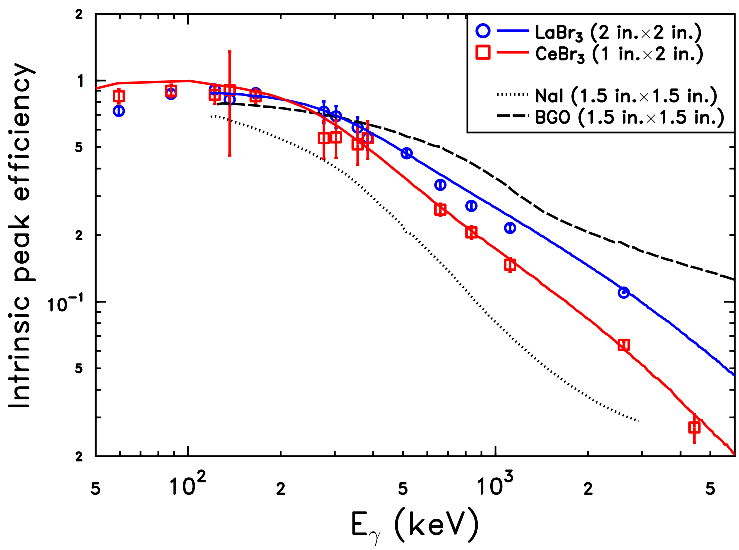


Fig. 2. Intrinsic peak efficiency as a function of $\gamma$-ray energy for a 2 in. × 2 in. large $LaBr_3$:Ce (circles) and a 1 in. × 2 in. $CeBr_3$ (squares) detector together with the corresponding results from Monte-Carlo simulations (full lines). For comparison the efficiency of both a NaI detector (dotted line) and a BGO detector (dashed line), both of size 1.5 in. × 1.5 in., are shown as well (Knoll, 2000).

## 2. The lanthanide halide scintillation detectors

In order to minimize the uncertainty in determining $\gamma$-ray multiplicity and mean energy, three important detector characteristics ought to be considered: (1) energy resolution, in order to be able to determine the structure of the emission spectra with good precision, (2) intrinsic full peak efficiency, in order to decrease the uncertainty of the response function, and (3) timing resolution, in order to efficiently separate prompt fission $\gamma$-rays from prompt fast neutrons by means of time-of-flight. The new lanthanide-halide scintillation detectors promise to provide a good compromise of these three properties. Therefore, at IRMM three different scintillation detector types were purchased, based on cerium-doped lanthanum-chloride ($LaCl_3$:Ce), cerium-doped lanthanum-bromide ($LaBr_3$:Ce) and cerium-bromide ($CeBr_3$) crystals, respectively. After their extensive characterization, we decided to use lanthanum-bromide, because of its superior timing as well as energy resolution (Billnert et al., 2012) and cerium-bromide, for the absence of intrinsic activity (Billnert et al., 2011; Lutter et al., 2013), for the experiment on $^{252}$Cf. In previous measurements on prompt fission $\gamma$-rays sodium-iodine (NaI) detectors were used to investigate the $\gamma$-multiplicity and mean energy (Verbinski et al., 1973; Pleasonton et al., 1972). Therefore it was important for us to know how our detectors compare to a typical sodium-iodine detector. As shown in Fig. 1, the energy resolution, defined as FWHM in percent, of a 1 in. × 2 in. $CeBr_3$ and a 2 in. × 2 in. $LaBr_3$:Ce detector is around 2/3 and 2/5, respectively, of the one of a 3 in. × 3 in. NaI detector. For comparison, data for a 3 in. × 3 in. bismuth germanium oxide (BGO) detector is depicted, too. With regard of intrinsic peak efficiency (Fig. 2), both lanthanide-bromide detectors are about twice as good as a 1.5 in. × 1.5 in. NaI detector, but only around 2/3 of a 1.5 in. × 1.5 in. BGO detector. The last property we needed to consider is timing resolution (Fig. 3). Here we wanted to investigate the timing resolution relative to the energy of the incoming particle, so we measured with either a $^{22}$Na or a $^{60}$Co source and applied different energy thresholds. The measurements were carried out in coincidence with a previously characterized $LaCl_3$ detector (Oberstedt A. et al., 2012). Even though we do not have any comparable data for the other detectors regarding different thresholds, we can still see that both the $CeBr_3$ and the $LaBr_3$:Ce detector are much faster than both NaI and BGO detectors. Although the properties of $CeBr_3$ and $LaBr_3$:Ce detectors are slightly better, we used also a 3 in. × 3 in. $LaCl_3$:Ce detector for the $^{235}$U($n_{th}$,f) experiment, simply because of its larger volume and, hence, higher intrinsic peak efficiency.

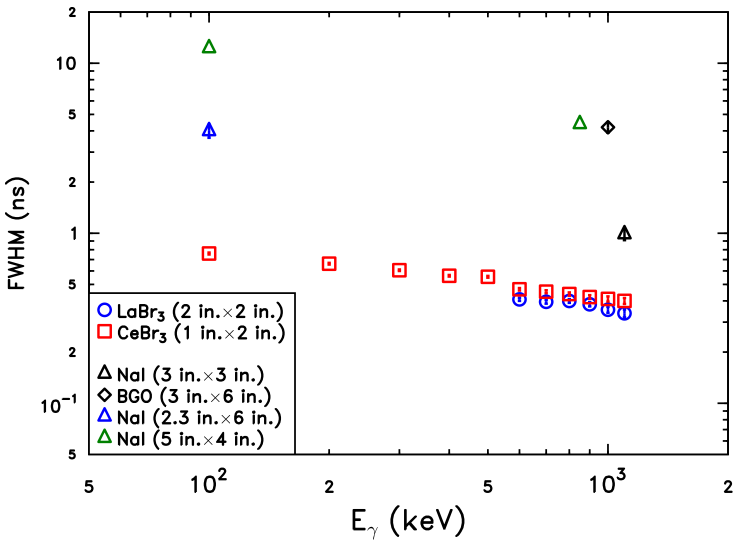


Fig. 3. Timing resolution (FWHM) as a function of $\gamma$-ray energy threshold for a 2 in. × 2 in. $LaBr_3$:Ce (circles) and a 1 in. × 2 in. $CeBr_3$ (squares) detector; see text for details. For comparison the timing resolution for different NaI and BGO detectors (Knoll, 2000), used in previous studies for spectral measurements (Verbinski et al., 1973; Pleasonton et al., 1972), is shown as well.

## 3. The $^{252}$Cf experiment

In order to separate prompt fission $\gamma$-rays from neutron induced $\gamma$-rays, we employed the time-of-flight method. This technique requires a fission trigger, which was provided by a simplified ionization chamber with a $^{252}$Cf source mounted inside, giving a signal for every fission event. This signal was fed into a slot of an Ortec 935 Quad Constant Fraction Discriminator (CFD) via a Timing Filter Amplifier (TFA) of type Ortec 474, and then with a proper delay into the stop input of a Ortec 567 Time-to-Amplitude Converter (TAC). For the start input we used the scintillation detector. The direct signals from both the scintillation detector and the fission trigger chamber were also fed via Ortec 460 Delay Line Amplifiers (DLA) into Canberra 8715 Analog-to-Digital Converters (ADC). This setup gave us the opportunity to select time windows, in order to eliminate all events that arrived later than what we expected from a prompt fission $\gamma$-ray. The distance between the detector and the source was determined with respect to the specific timing resolution of each detector, to make sure that there was a sufficient time difference between the prompt $\gamma$-peak and the fast neutron interactions.

Once we had measured the prompt fission $\gamma$-ray spectra, we needed to unfold them with the corresponding detector's response function in order to determine the emission spectra from the source. These integral response functions were obtained by simulating the response of 220 different energies using a Monte Carlo code (PENELOPE, 2011) and folded with the experimentally found energy resolution (cf. Fig. 1). These individual energy spectra were then fitted to the measured prompt fission $\gamma$-ray spectra, starting with the highest energy and moving towards the lower ones. From the factor needed to adjust the first simulated energy peak to the measured spectra, we deduced the amount of photons of that particular energy, which the source emits in $4\pi$. The fully simulated distribution, including Compton continuum and escape peaks (for energies above 1.022 MeV), was then subtracted from the measured spectrum, and the next lower energy was fitted, and so on. The simulated integral response function as well as the corresponding prompt $\gamma$-ray spectrum from the measurement with the $LaBr_3$ detector can be seen in the upper part of Fig. 4. The middle part of the same figure shows the residuals, i.e. the relative deviations between simulated and measured spectra. The overall agreement is obviously good. The unfolded emission spectra from both the $LaBr_3$:Ce and the $CeBr_3$ detector are depicted in the lower part of Fig. 4. Here we notice that the results obtained with both detectors agree very well with each other, which is impressively obvious also by the low-energy structure shown in the inset. In Fig.

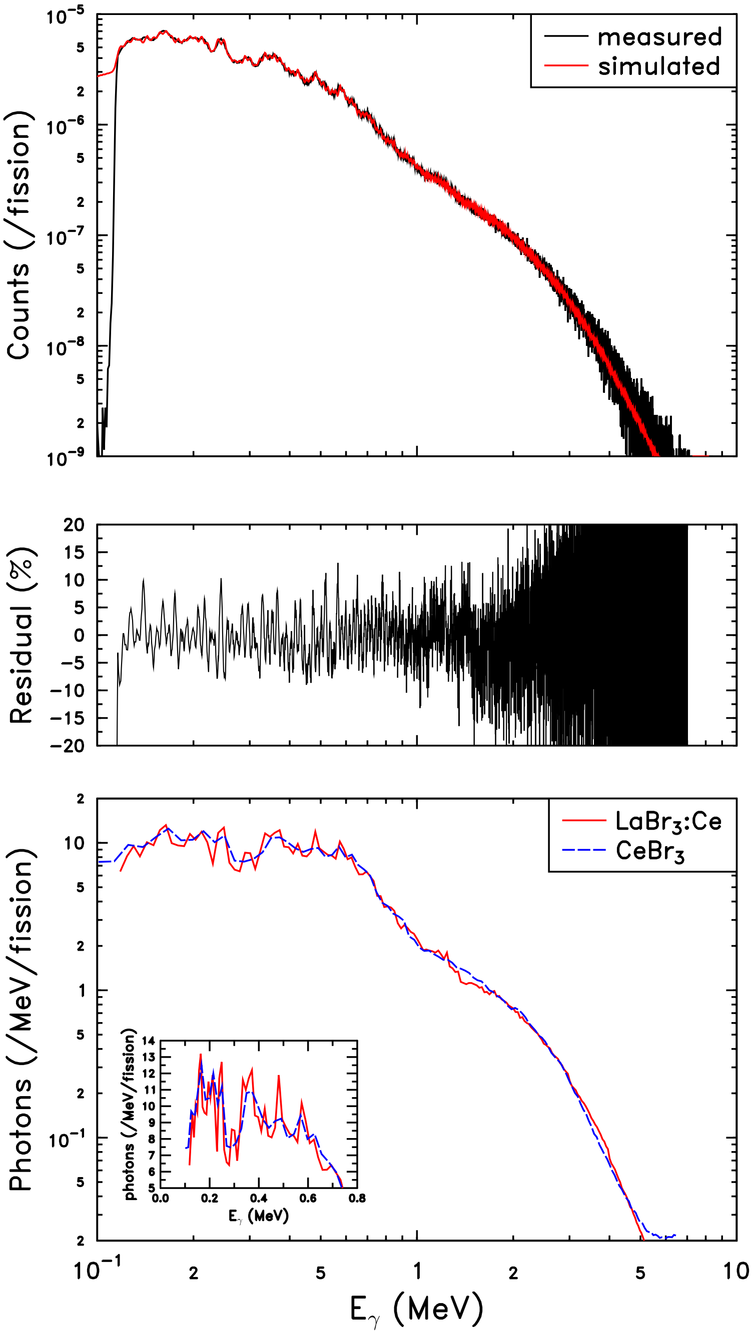


Fig. 4. The upper part shows the integral simulated spectrum (red line) adjusted to the experimental one (black line) from the measurement with a 2 in. × 2 in. $LaBr_3$:Ce detector, while the relative difference between measured spectrum and simulation is shown in the middle part. The lower part shows the unfolded prompt fission $\gamma$-ray emission spectrum taken with a 2 in. × 2 in. $LaBr_3$:Ce (full red line) and a 1 in. × 2 in. $CeBr_3$ detector (dashed blue line). The inset focusses on $\gamma$-ray energies below 1 MeV and demonstrates the very good agreement between the results obtained with both detectors used in this work.

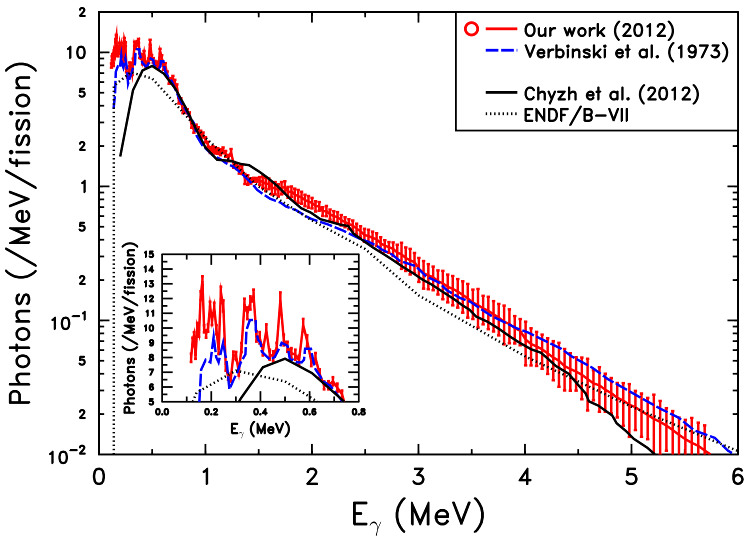


Fig. 5. The prompt fission $\gamma$-ray emission spectrum of $^{252}$Cf(SF) from this work taken with a 2 in. × 2 in. $LaBr_3$:Ce detector (full red line) is shown together with data from (Verbinski et al., 1973; Chyzh et al., 2012) as well as from ENDF/B-VII.0 for comparison. In the high energy range, all spectra agree rather well with each other, but the data from both ENDF/B-VII.0 and (Chyzh et al., 2012) lacks structure in the low energy range. This is even more obvious in the inset, which focusses on $\gamma$-ray energies below 1 MeV, and demonstrates on the other hand the very good reproduction of the historical data from (Verbinski et al., 1973) (see text for details).

5 we compare our results obtained with the $LaBr_3$:Ce detector with those from (Verbinski et al., 1973; Chyzh et al., 2012), as well as with the data from the evaluated data tables, ENDF/B-VII.0. As can be seen, our data is consistent with the previously measured spectra, but the evaluated data does not describe either of these experimental datasets. The explanation for this discrepancy is probably that the evaluated data for $^{252}$Cf might not be based on any experimental data. In the low energy region we notice again the previously mentioned structure in the emission spectra, which is similar for all three of the experimental data, but since the $LaBr_3$ detector has a superior energy resolution, the structure is more pronounced in the spectrum taken with this detector (cf. also Fig. 4). For integral $\gamma$-multiplicity and total energy, this energy resolution is not important. But, if we want to further correlate prompt $\gamma$-emission with certain fission product characteristics, e.g. mass, it is important to have an energy resolution as good as possible. It should be noted that the error bars for our data contain both statistical uncertainties as well as uncertainties from the determination of the response function. The results that are most relevant for nuclear applications are summarized in Table 1.

Table 1. Overview of results for the spontaneous fission of $^{252}$Cf. The experimental results from this work for the prompt fission $\gamma$-ray multiplicity $\nu_\gamma$, the average energy $\epsilon_\gamma$ and the total energy $E_{\gamma,tot}$, obtained with both detectors employed here, are compared to previously measured values from (Verbinski et al., 1973; Pleasonton et al., 1972; Chyzh et al., 2012) as well as corresponding numbers from the evaluated nuclear data files in ENDF/B-VII.0.

| Results: | $\nu_\gamma$ (per fission) | $\epsilon_\gamma$ (MeV) | $E_{\gamma,tot}$ (MeV) |
|---|---|---|---|
| This work ($LaBr_3$:Ce) | 8.30 ± 0.54 | 0.80 ± 0.01 | 6.60 ± 0.50 |
| This work ($CeBr_3$) | 8.39 ± 0.76 | 0.80 ± 0.02 | 6.69 ± 0.62 |
| Verbinski et al. (1973) | 7.80 ± 0.30 | 0.88 ± 0.04 | 6.84 ± 0.30 |
| Pleasonton et al. (1972) | 8.32 ± 0.40 | 0.85 ± 0.06 | 7.06 ± 0.35 |
| Chyzh et al. (2012) | 8.14 ± 0.40 | 0.94 ± 0.05 | 7.65 ± 0.55 |
| ENDF/B-VII.0 | 7.23 | 0.81 | 5.84 |

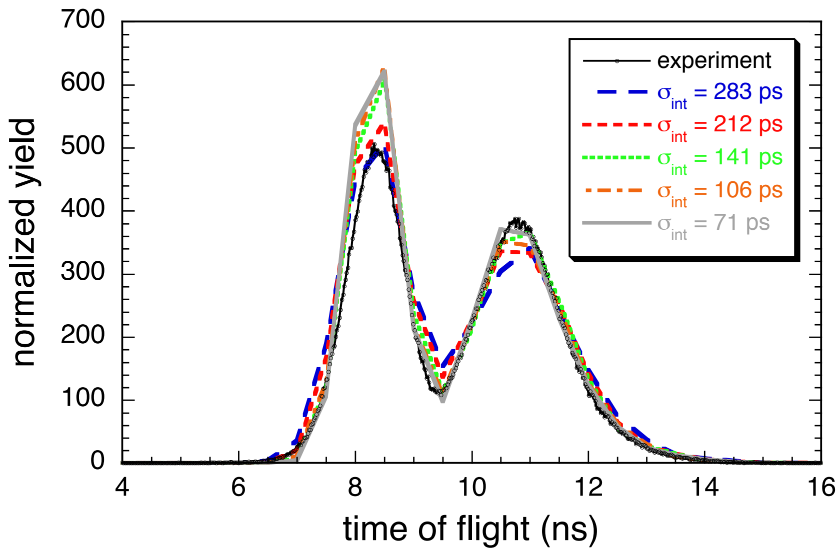


Fig. 6. Time-of-flight spectrum of fission fragments from $^{252}$Cf taken for a pair of 180 $\mu$m thick and 4-fold segmented polycrystalline CVD diamond detectors of total area $1 \times 1$ cm$^2$ (black line and symbols) at a distance of 10.5 cm. The experimentally obtained distribution is compared to the results of Monte Carlo simulations assuming different intrinsic timing resolutions.

## 4. The $^{235}$U(n$_{th}$,f) experiment

This experiment was performed at the 10 MW research reactor of KFKI Budapest. The $\gamma$-rays were measured in coincidence with fission fragments, which were detected by using the fission fragment spectrometer VERDI (Oberstedt S. et al., 2010). It contained a $^{235}$U sample of mass 113 $\mu$g, mounted on a 34 $\mu$g/cm$^2$ thick polyimide backing, a polycrystalline chemical vapour deposited (pCVD) 4-fold segmented diamond detector, which provided the fast fission trigger. In a preceding study the intrinsic timing resolution was determined by Monte Carlo simulations reproducing the measured time-of-flight spectrum of fission fragments from $^{252}$Cf (cf. Fig. 6) to $\sigma$ = (106 ± 21) ps. More information on the diamond detectors may be found elsewhere (Oberstedt S. et al., 2013). The reactor delivered a cold neutron beam with a flux of some $10^7$ cm$^{-2}$ s$^{-1}$, causing a fission rate above $10^4$ s$^{-1}$. Four scintillation detectors (one 3 in. × 3 in. and two 1.5 in. × 1.5 in. $LaCl_3$:Ce detectors as well as one 2 in. × 2 in. $LaBr_3$:Ce detector) were placed outside the time-of-flight spectrometer VERDI at a distance of about 30 cm. Blankets containing $^6$Li and lead blocks were applied as shielding against scattered thermal neutrons and $\gamma$-rays, respectively. The signals from the four $\gamma$-detectors were fed into an Ortec 935 Quad Constant Fraction Discriminator and via amplifiers further into the same Ortec 567 Time-to-Amplitude Converter of range 1 $\mu$s. They were giving the start signal, while the signals from the diamond detector, with an appropriate delay, provided the stop signal of the coincidence for the time-of-flight measurements. For all four scintillation detectors the pulse height was digitized by Canberra 8715 Analog-to-Digital Converters and stored in listmode, together with three pulse shape discrimination signals for the $LaCl_3$:Ce detectors only, as well as the TAC signal.

So far only the data taken with the 3 in. × 3 in. $LaCl_3$:Ce detector has been processed. The unfolding procedure of its response function was performed in the same way as described above for the $^{252}$Cf experiment (cf. Sect. 3). The resulting emission spectrum of prompt fission $\gamma$-rays is depicted in Fig. 7, together with experimental data of (Verbinski et al., 1973) and a recent evaluation in ENDF/B-VII.0. The agreement between all three spectra is obviously very good. However, it has to be noted that the threshold in our measurement was quite high, i.e. around 450 keV, which was necessary in order to reduce the dead time because of low-energy noise due to the high sensitivity of the $LaCl_3$:Ce detector. Extracted prompt fission $\gamma$-ray multiplicity, average energy and total energy are summarized in Table 2.

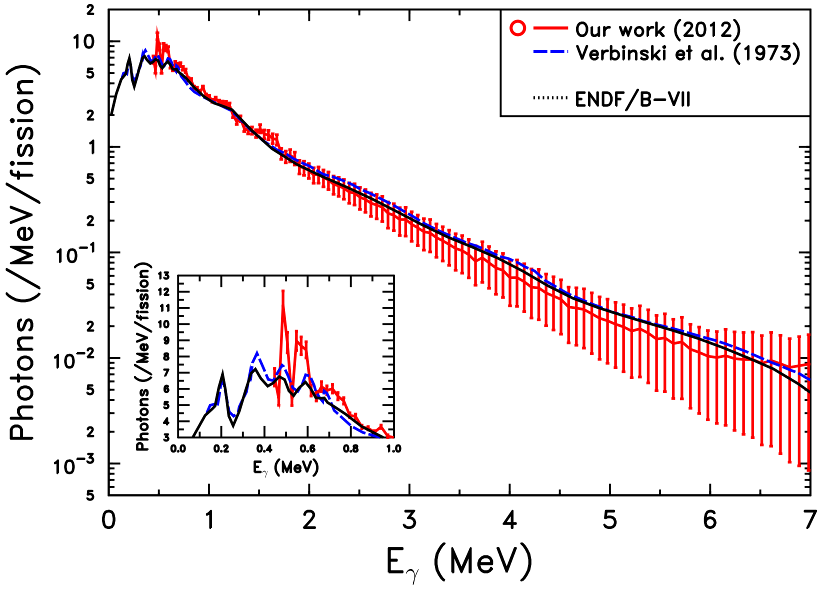


Fig. 7. The prompt fission $\gamma$-ray emission spectrum of $^{235}$U($n_{th}$,f) from this work taken with a 3 in. × 3 in. $LaCl_3$:Ce detector (full red line) is shown together with experimental data from (Verbinski et al., 1973) as well as from ENDF/B-VII.0 for comparison. The agreement between all spectra is very good, even the structure at low energies is reproduced (see inset). However, it has to be noted that the threshold in our measurement is quite high (around 450 keV), which was necessary in order to suppress the low-energy noise (see text for details).

## 5. Conclusions and outlook

In this work we have presented the results from the prompt fission $\gamma$-ray spectral measurements of the reaction $^{252}$Cf(SF). From that, the emission yield was determined to $\nu_\gamma$ = (8.30 ± 0.54)/fission and $\nu_\gamma$ = (8.39 ± 0.76)/fission, the average energy to $\epsilon_\gamma$ = (0.80 ± 0.01) MeV and $\epsilon_\gamma$ = (0.80 ± 0.02) MeV, and the total energy to $E_{\gamma,tot}$ = (6.6 ± 0.5) MeV and $E_{\gamma,tot}$ = (6.7 ± 0.6) MeV, with the $LaBr_3$ and $CeBr_3$ detector, respectively. The shapes of the measured spectra from two different detectors agree very well with each other and with previously published ones from the early 1970's (Verbinski et al., 1973; Pleasonton et al., 1972). The same is true, at least for high $\gamma$-energies, for recent results of (Chyzh et al., 2012). The characteristic parameters as listed in Table 1 are also nicely reproduced, except for the average energy of a $\gamma$-ray as well as the total $\gamma$-energy given in (Chyzh et al., 2012). This discrepancy, however, is understandable, since their detectors are not able to efficiently measure the low energy region due to absorption effects. Hence, both mean and total energy are overestimated. All this gives us the confidence that both our method of measuring prompt fission $\gamma$-rays and the determination of the integral response function of both employed detectors are accurate. Since the data in ENDF/B-VII.0 does match neither our data nor the experimental data from (Verbinski et al., 1973; Pleasonton et al., 1972), we strongly suggest that the evaluated data tables should be updated as soon as

Table 2. Overview of results for the thermal neutron-induced fission of $^{235}$U. The first experimental results from this work for the prompt fission $\gamma$-ray multiplicity $\nu_\gamma$, the average energy $\epsilon_\gamma$ and the total energy $E_{\gamma,tot}$, obtained with a 3 in. × 3 in. $LaCl_3$:Ce detector, are compared to previously measured values from (Verbinski et al., 1973) as well as corresponding numbers from the evaluated nuclear data files in ENDF/B-VII.0.

| Results: | $\nu_\gamma$ (per fission) | $\epsilon_\gamma$ (MeV) | $E_{\gamma,tot}$ (MeV) |
|---|---|---|---|
| This work ($LaCl_3$:Ce) | 4.7 ± 0.6 | 1.20 ± 0.04 | 5.63 ± 0.70 |
| Verbinski et al. (1973) | 4.55 | 1.28 ± 0.05 | 5.8 ± 0.3 |
| ENDF/B-VII.0 | 5.1 | 1.22 | 5.4 |

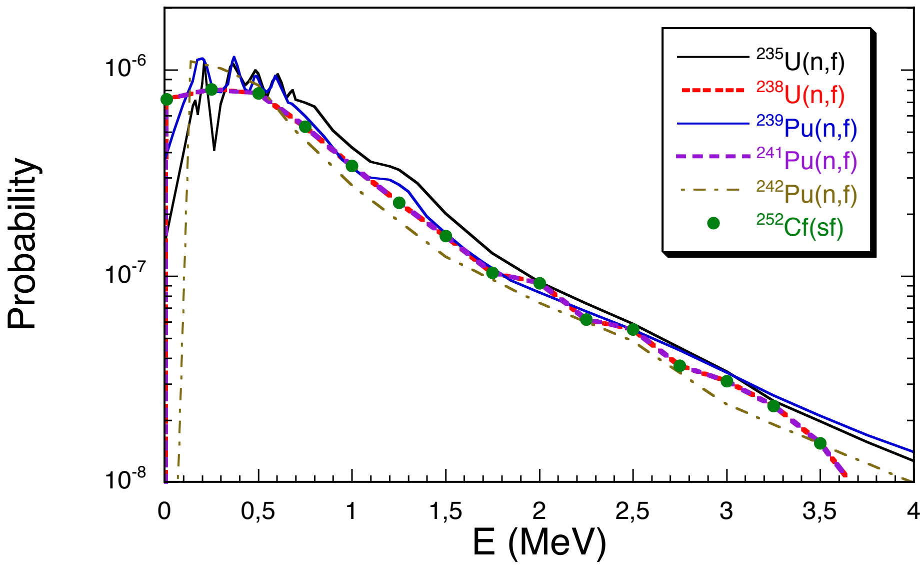


Fig. 8. Evaluated prompt fission $\gamma$-ray emission spectra of different neutron-induced and spontaneous fission reactions as provided by ENDF/B-VII.0. The similarity between the spectra for $^{238}$U(n, f), $^{241}$Pu(n, f) and $^{252}$Cf(SF) is striking as well as for $^{235}$U(n, f) and $^{239}$Pu(n, f). Whether this is really true requires experimental confirmation.

possible. In the meantime, this part of our work has been published elsewhere in more detail (Billnert et al., 2013).

We have also reported on our first measurement of the prompt fission $\gamma$-ray spectrum from the reaction $^{235}$U(n$_{th}$,f). Here we have employed yet another scintillation detector compared to the previous experiment. The obtained results so far are an emission yield of $\nu_\gamma = (4.7 \pm 0.6)$/fission, an average photon energy of $\epsilon_\gamma = (1.20 \pm 0.04)$ MeV and a total $\gamma$-ray energy of $E_{\gamma,tot} = (5.63 \pm 0.70)$ MeV. Again, the agreement of the spectral shape with the historical results from (Verbinski et al., 1973) is good, but here this also true also for the data in ENDF/B-VII.0. Recently another measurement was performed, this time employing two 2 in. × 2 in. $LaBr_3$ and three 1 in. × 2 in. $CeBr_3$ detectors. The data analysis is not completed yet, still, preliminary results for the data taken with one of the $LaBr_3$ detectors indicate already excellent agreement with the results from the experiment reported about in this work.

We also noticed that the data in the evaluated tables for $^{238}$U and $^{241}$Pu seems to have been obtained by simply applying a scaling factor to the evaluation for $^{252}$Cf (cf. Fig. 8). Since we have shown here that the evaluated data for $^{252}$Cf are in conflict with existing experimental results, it might be reasonable to assume that at least some of the underestimation mentioned by (Rimpault, 2006; Rimpault et al., 2006) is due to an unrealistic evaluation of prompt fission $\gamma$-ray data from $^{238}$U and $^{241}$Pu. Therefore, an experiment on $^{241}$Pu(n$_{th}$, f) is planned for next year in Budapest. A corresponding application for financial support within the frame work of ERINDA was submitted and has recently been granted. In the meantime we will continue the analysis of the remaining data from our $^{235}$U(n$_{th}$, f) measurements, in order to further investigate correlations between certain $\gamma$-energies with different fission fragment characteristics for both $^{235}$U(n$_{th}$, f) and $^{252}$Cf(SF).


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