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Total prompt γ-ray emission in fission

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Abstract. The total prompt γ-ray energy distributions for the neutron-induced fission of ^{235}U, ^{239,241}Pu at incident neutron energy of 0.025 eV – 100 keV, and the spontaneous fission of ^{252}Cf were measured using the Detector for Advanced Neutron Capture Experiments (DANCE) array in coincidence with the detection of fission fragments by a parallel-plate avalanche counter. DANCE is a highly segmented, highly efficient 4π γ-ray calorimeter. Corrections were made to the measured distribution by unfolding the two-dimension spectrum of total γ-ray energy vs multiplicity using a simulated DANCE response matrix. The mean values of the total prompt γ-ray energy, determined from the unfolded distributions, are ~ 20% higher than those derived from measurements using single γ-ray detector for all the fissile nuclei studied. This raises serious concern on the validity of the mean total prompt γ-ray energy obtained from the product of mean values for both prompt γ-ray energy and multiplicity.

1 Introduction

The total prompt γ-ray emission in fission accounts for about 40% of the total energy released by γ-ray emission that makes up about 10% of the total energy released in reactor core. The heating in nuclear reactors attributed to the total γ-ray emission in fission is underestimated up to 28% using the evaluated data for the main reaction channels, ^{235}U(n,f) and ^{239}Pu(n,f) [1]. This discrepancy is significantly greater than 7.5%, an upper bound of the uncertainty deemed necessary to adequately model the heat deposit in the fuel core [2,3]. Therefore, efforts are needed to improve the experimental data on the γ-ray emission in fission. As a matter of fact, the request for the new data on the prompt fission γ rays at thermal energy and above for those two isotopes has been categorized as the high-priority by the Nuclear Energy Agency under the Organization for Economic Co-operation and Development [4]. The majority of measurements made for the prompt γ-ray emission in fission always employed a single or a few γ-ray detectors. For example, a single NaI detector was used by Verbinski *et al.* [5] more than 40 years ago and the cerium-doped $LaBr_3$, $CeBr_3$, and $LaBr_3$ detectors were used recently by Billnert *et al.* [1] and Oberstedt *et al.* [6,7].

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Below we describe the analysis and results on the total γ-ray emission in fission measured by the DANCE array [8,9]. DANCE consists of 160 equal-volume, equal-solid-angle BaF_2 detectors, covering a 4π geometry space, and is located at the Los Alamos Neutron Science Center (LANSCE). Several unique features exhibited by DANCE are particularly attractive for those measurements, such as the nearly γ-ray energy independence for the detection efficiency, the multiplicity response, and the peak-to-total ratio, all of which are described in detail in Refs. [10-12]. For example, it enables one to measure the total γ-ray energy as a function of multiplicity. The only limitation is the energy resolution, which is about 14% for the measured total γ-ray energy. A series of measurements of the prompt γ rays in the neutron-induced fission of ^{235}U and ^{239,241}Pu, and the spontaneous fission of ^{252}Cf has been carried out recently using DANCE in coincidence with the detection of fission fragments by a compact parallel-plate avalanche counters (PPAC) [13]. The results on the measured and unfolded fission prompt γ-ray energy and multiplicity distributions for those isotopes have been published [12,14]. An independent analysis of the same data for ^{239}Pu, by assuming a general parameterized correlation between E_γ and M_γ, was presented in Ref. [15]. We also reported the total prompt γ-ray energy distributions for those isotopes, obtained by unfolding the measured two-dimensional spectrum of total γ-ray energy vs multiplicity [16]. This unfolding procedure and the implication on the γ heating in nuclear reactors are described.

2 Experiments and data analysis

The measurements of the prompt γ emission in the neutron-induced fission of ^{235}U and ^{239,241}Pu as well as the spontaneous fission in ^{252}Cf were performed at the Lujan Center of LANSCE. The experimental setup and the data analysis have been described in detail in our early publications [12,14-16]. A brief summary of the experiments is given here. For the neutron-induced fission experiment, neutrons with energies from thermal up to several hundred keV were produced first by bombarding an 800-MeV proton beam at a repetition rate of 20 Hz on a tungsten target then moderated by water. The prompt γ rays emitted in fission were detected by the DANCE array in coincidence with the detection of fission fragments by a compact PPAC [13]. More than 10^6 fission events with at least one γ ray detected by DANCE were collected for all isotopes studied. The threshold for detecting γ-ray energy by DANCE was set to 150 keV. The summed energy of all γ rays detected by DANCE within a time window of 40 ns was defined as the total prompt γ-ray energy ($E_{\gamma,tot}$) in fission for a given event. With this time window extended to 100 ns, little change was observed for the $E_{\gamma,tot}$ spectrum [15]. The possible background contribution to $E_{\gamma,tot}$ is due to capture of thermalized prompt fission neutrons by Ba isotopes, which is on the order of μs and too long in the time scale for prompt γ rays. Additional suppression of neutron contribution is made by placing a gate on the pulse height spectrum of PPAC in addition to the 8-ns gate on the time spectrum between PPAC and DANCE, show in Fig. 1. All the offline data analysis was carried out using the code, FARE [17]. Note that both DANCE and PPAC have a similar intrinsic time resolution of ~ 1.2 ns [13]. The total γ-ray multiplicity (M_γ) in fission is established not according to the number of detectors observing the γ ray, but instead according to the number of clusters by grouping adjacent detectors catching the γ ray in the same time window. This counting method for M_γ is closer to the simulated results using the γ-ray calibration sources [10-12]. In addition, the nearly γ-ray energy independence of the DANCE response to M_γ, indicated by the numerical simulations, enables one to unfold approximately the measured M_γ distribution in fission for the first time [12,14].

Corrections have to be made to the measured $E_{\gamma,tot}$ distribution to obtain the physical one, which would be useful for the applications. This can be accomplished by unfolding the two-dimensional spectrum of $E_{\gamma,tot}$ vs M_γ. The two-dimensional unfolding is necessary because of the strong dependence of $E_{\gamma,tot}$ on M_γ. It is numerically implemented by adopting the iterative Bayesian method [18-20]. The DANCE response matrix for $E_{\gamma,tot}$ vs M_γ is simulated using the GEANT4 [21] geometrical model of both DANCE and PPAC [12,14,22]. To make sure this two-dimensional response matrix has a sufficient coverage of the phase space beyond the measured one, the value of M_γ up to 25 and $E_{\gamma,tot}$ up to 40 MeV are included. The $E_{\gamma,tot}$ has a bin size of 200 keV and an energy threshold of 150 keV. So the response matrix has a size of 200 × 25.

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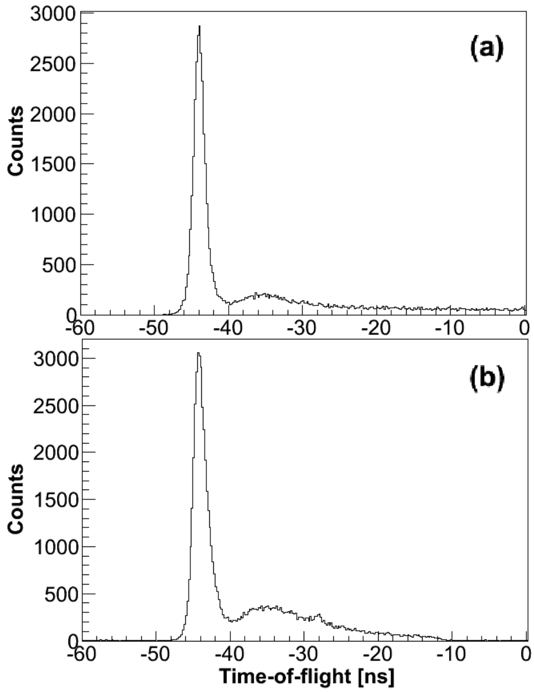

Figure 1. Time difference between γ rays detected by DANCE and fission fragments detected by PPAC for (a) ^{235}U and (b) ^{241}Pu experiments with an achieved time resolution of ~ 1.7 ns. The bump next to the peak is related to events with ambiguous correlation between DANCE and PPAC.

For any given grid point ($E_{\gamma,tot}$, M_γ) in the response matrix, a two-dimensional DANCE response matrix of a size of 200 × 25 is generated using GEANT4 with a given assembly of no more than 20,000 samples. Note that the DANCE response to the total prompt γ-ray is relatively insensitive to the content of γ rays for a given sample since the γ-ray detection efficiency (84 to 88%) and the peak-to-total ratio (~ 55%) remain nearly constant for the γ-ray energy ranging from 150 keV to 10 MeV [10-12]. Each sample has a matching number of γ rays to M_γ, selected randomly according to the unfolded γ-ray energy distributions [12,14] with the condition on the total γ-ray energy that is equal to $E_{\gamma,tot}$ ± 100 keV. This simulation is repeated for all the grid points within the lower and upper bound of $E_{\gamma,tot}$ for a given M_γ, established by this random sampling technique.

The resulting ($E_{\gamma,tot}$, M_γ) DANCE response matrix consists of ~ 3300 two-dimensional matrices with a size of 200 × 25 each. This numerically simulated DANCE response matrix is unique for each isotope studied, and was used to unfold the measured two-dimensional spectrum of $E_{\gamma,tot}$ vs M_γ into a physical one using the iterative Bayesian method. During the iteration stage, a single factor was applied to and varied for the response matrix at any given grid point.

3 Results and discussions

Typically it takes about 30 iterations to reach the convergence in the unfolding of the two-dimensional spectrum of $E_{\gamma,tot}$ vs M_γ using the Bayesian method. The results for the neutron-induced fission in ^{239}Pu are shown in Fig. 2 where the unfolded $E_{\gamma,tot}$ vs M_γ spectrum together with the measured one are given. In addition, the comparisons of the projected $E_{\gamma,tot}$ and M_γ distributions between the unfolded and measured ones are also given. The general trend of the results is that the mean value and the width of projected $E_{\gamma,tot}$ and M_γ distributions increases noticeably after the unfolding.

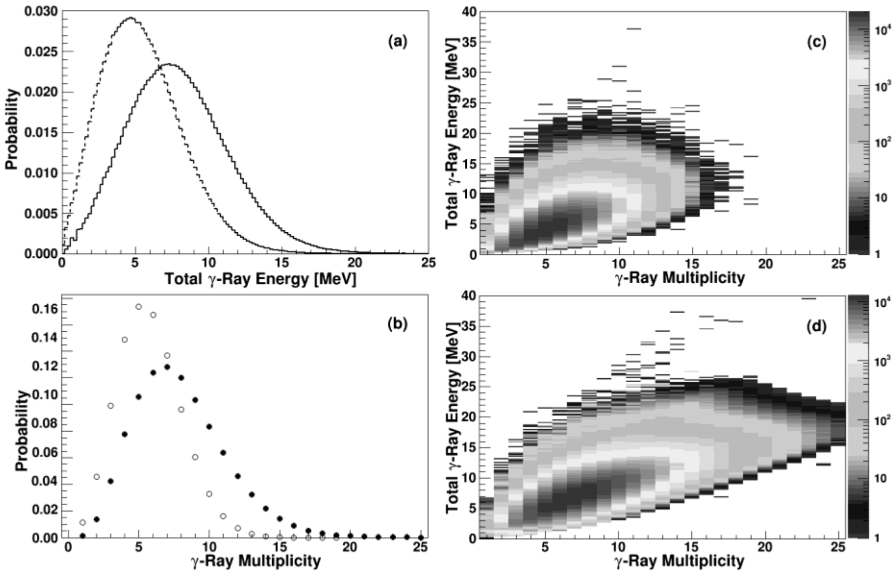

Figure 2. Shown in panel (c) and (d), respectively, are the measured and unfolded total prompt γ-ray energy vs. multiplicity distribution for the neutron-induced fission of ^{239}Pu. Comparison of the projected total γ-ray energy and multiplicity distributions between measured (dashed line, open circles) and unfolded one (solid line, filled circles) are given in panels (a) and (b), respectively.

Given in Table 1 is the comparison of $\langle M_\gamma \rangle$ derived from the unfolded M_γ distribution between the recent work (2-D) and the early one using the one-dimension unfolding technique [14] for all isotopes studied. For ^{235}U, the recent (2-D) mean value of 7.35 is 0.37 higher than 6.98 in the earlier 1-D work. However, the latter value is known to be underestimated by about 0.30. Since these values were derived from the same data set, this consistence in the derived mean M_γ from both the one- and two-dimensional unfolding techniques gives us a certain confidence in the validity of our work. This trend is the same for ^{239,241}Pu and ^{252}Cf. The comparison with other measurements and evaluations also is given in Table 1. Our measured $\langle M_\gamma \rangle$'s for all isotopes studied are consistently higher than the weighted-average of earlier measurements [23] by ~ 10% except for the most recent measurements [1,6,7], where their measured $\langle M_\gamma \rangle$ is ~ 11% greater than ours for ^{235}U but near in agreement with ours for both ^{241}Pu and ^{252}Cf. Moreover, ours are consistent with the evaluated data listed in ENDF/B-VII.1 [24]. The uncertainty for our derived $\langle M_\gamma \rangle$ has an upper bound of about 0.3-0.4 or ~ 5%.

The comparison of $\langle E_{\gamma,tot} \rangle$ between our measurements and previous ones is given in Table 2. For ^{235}U, the recent (2-D) derived mean $E_{\gamma,tot}$ of 8.35 MeV is higher than 6.53(20) MeV, the weighted average of previous measurements [23], and 6.60 MeV, the evaluated data listed in ENDF/B-VII.1. It also is higher than 6.92(9) MeV, the most recent measurement [6]. The same comparisons are also made for the neutron-induced fission in ^{239,241}Pu and the spontaneous fission in ^{252}Cf. Our measured $\langle E_{\gamma,tot} \rangle$ are consistently higher than the previous ones [1,6,7,23] by ~ 20% for all isotopes studied. The uncertainty for our derived $\langle E_{\gamma,tot} \rangle$ is dominated by the systematic error and roughly estimated to be better than 5%, assuming a similar uncertainty to that of the derived $\langle M_\gamma \rangle$.

Table 1. Comparison of the mean M_γ between our recent measurements and previous ones for the neutron-induced fission of ^{235}U and ^{239,241}Pu as well as the spontaneous fission of ^{252}Cf.

Isotope	2-D	1-D	Ref. 15	ENDF/B-VII.1	Ref. 23	Refs. 1, 6, 7
^{235}U	7.35	6.95		7.04	6.60(10)	8.19(11)
^{239}Pu	7.93	7.50	7.15	7.78	7.06(20)	
^{241}Pu	7.97	7.50		8.18		8.21(9)
^{252}Cf	8.75	8.16			7.98(40)	8.30(8)

An independent analysis of the same DANCE data for ^{239}Pu by assuming a very general parameterized correlation between E_γ and M_γ has been carried out by Ullmann et al. [15], which yields the $\langle E_{\gamma,tot} \rangle$ = 7.46 MeV and $\langle M_\gamma \rangle$ = 7.15. The $\langle E_{\gamma,tot} \rangle$, derived from the $E_{\gamma,tot}$ distribution, agrees within 6% of that obtained by using the 2-D unfolding technique. This agreement is significant and indicates the importance of the correlation between E_γ and M_γ to be considered in the determination of $\langle E_{\gamma,tot} \rangle$. It raises serious concern on the validity of the equation, $\langle E_{\gamma,tot} \rangle = \langle E_\gamma \rangle \times \langle M_\gamma \rangle$, which ignores the correlation between E_γ and M_γ exhibited in Fig. 2.

Table 2. Comparison of the mean $E_{\gamma,tot}$ (MeV) between our recent measurements and previous ones for the neutron-induced fission of ^{235}U and ^{239,241}Pu as well as the spontaneous fission of ^{252}Cf.

Isotope	2-D	Ref. 15	ENDF/B-VII.1	Ref. 23	Refs. 1, 6, 7
^{235}U	8.35		6.60	6.53(20)	6.92(9)
^{239}Pu	7.94	7.46	6.74	6.78(10)	
^{241}Pu	8.01		7.26		6.41(6)
^{252}Cf	8.52			6.95(30)	6.64(8)

4 Summary

A systematic study of the total prompt γ-ray emission in the neutron-induced fission of ^{235}U and ^{239,241}Pu as well as the spontaneous fission of ^{252}Cf has been carried out using the DANCE array together with a compact PPAC to select the fission event by detecting its fission fragments. The total γ-ray energy vs multiplicity spectrum for all fissile nuclei studied was constructed and unfolded using a two-dimensional unfolding technique, numerically implemented by adopting the iterative Bayesian method. The $\langle E_{\gamma,tot} \rangle$ derived from the projected $E_{\gamma,tot}$ distribution of the unfolded $E_{\gamma,tot}$ vs M_γ spectrum is about 20% higher than the previous measurements for all fissile nuclei studied. However, it agrees reasonably well with the result derived from the analysis by considering the correlation between E_γ and M_γ in a very general parameterization manner. In addition, the measured total prompt γ-ray energy vs multiplicity spectrum in fission enables one to evaluate the variance in addition to the average value of the energy deposited in a reactor core by the prompt fission γ rays. This may improve our understanding of the γ heating in many applications involving nuclear fission.

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