Total prompt γ-ray emission in fission of ^{235}U, ^{239,241}Pu, and ^{252}Cf

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The total prompt γ-ray energy distributions for the neutron-induced fission of ^{235}U and ^{239,241}Pu in the neutron energy range of 0.025 eV – 100 keV and the spontaneous fission of ^{252}Cf were measured using the Detector for Advanced Neutron Capture Experiments (DANCE) 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 generated with a geometrical model of the detector arrays and validated with the γ-ray calibration sources. The mean values of the total prompt γ-ray energy, determined from the unfolded distributions, are $\sim$20% higher than those of early measurements for all the fissile nuclei studied. The implication for the γ heating in nuclear reactors is discussed.

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I. INTRODUCTION

The total prompt γ-ray emission in fission accounts for about 40% of the total energy released by γ-ray emission, which 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 total 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 high priority by the Nuclear Energy Agency under the Organization for Economic Co-operation and Development [4]. The majority of measurements made for the total prompt γ-ray emission always employed a single or a few γ-ray detectors. For example, a single NaI detector was used by Verbinski *et al.* [5] nearly 40 years ago. Cerium-doped $LaBr_3$, $CeBr_3$, and $LaBr_3$ detectors were used recently by Billnert *et al.* [1] and Oberstedt *et al.* [6].

Here we present results obtained using the DANCE array [7,8], which is an alternative to other techniques in measuring the prompt γ-ray emission in fission. DANCE consists of 160 equal-volume, equal-solid-angle BaF_2 detectors forming a 4π geometry coverage, 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 near 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. [9–11]. 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 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 compact parallel-plate avalanche counters (PPAC) [12]. The results on the measured and unfolded fission prompt γ-ray energy and multiplicity distributions for those isotopes have been published [11,13]. A model-dependent analysis of the same data for ^{239}Pu was presented in Ref. [14]. In this article, we analyzed the same data set [11,13] with a different technique and report 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. Details of this unfolding procedure and the implication on the γ heating in nuclear reactors are presented.

II. 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 in 2010 and 2011 at the Lujan Center of LANSCE. The experimental setup and the data analysis have been described in detail in our early publications [11,13,14]. 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 and 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. A minimum of 10^6 fission events with at least one γ ray detected by DANCE were collected for all isotopes studied. The hardware threshold for detecting γ-ray energy by DANCE was 150 keV.

The summed energy of all γ rays detected by DANCE within a time window of 40 ns and the 8-ns gate on the time spectrum between PPAC and DANCE was defined as the total prompt γ-ray energy ($E_{\gamma,\mathrm{tot}}$) in fission for a given event. The DANCE time window is used to suppress the

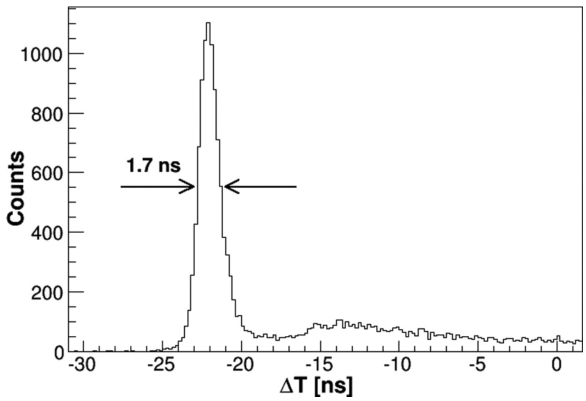

FIG. 1. The time spectrum between the detection of fission fragments by PPAC and the detection of γ rays by DANCE for the ^{239}Pu target. The time resolution of 1.7 ns is achieved. The broad bump next to the narrow peak results mostly from events with an ambiguous time correlation between PPAC and DANCE, because the ratio of neutron to γ is $\leqslant 2\%$ from a simulation of the DANCE response to fission.

slow neutrons captured by Ba isotopes, and when extended to 100 ns, little change was observed for the $E_{\gamma,\text{tot}}$ spectrum [14]. A typical time spectrum between PPAC and DANCE is given in Fig. 1, where the 8-ns window is applied to suppress fast neutrons. Some of the offline data analysis was carried out using the code FARE [15]. Note that both DANCE and PPAC have similar time resolutions of $\sim$1.2 ns [12]. 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 [9–11]. In addition, the near 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 [11,13].

Corrections have to be made to the measured $E_{\gamma,\text{tot}}$ distribution to obtain the physical one, which would be useful for the applications. This can be accomplished by unfolding the two-dimensional spectrum of $E_{\gamma,\text{tot}}$ vs M_γ. The two-dimensional unfolding is necessary because of the strong dependence of $E_{\gamma,\text{tot}}$ on M_γ. It is numerically implemented by adopting the iterative Bayesian method [16–18]. The DANCE response matrix for $E_{\gamma,\text{tot}}$ vs M_γ is simulated using the GEANT4 [19] geometrical model of both DANCE and PPAC [11,13,20]. 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 $E_{\gamma,\text{tot}}$ up to 40 MeV are included. The $E_{\gamma,\text{tot}}$ has a bin size of 200 keV and an energy threshold of 150 keV. So the response matrix has a size of 200×25.

For any given grid point ($E_{\gamma,\text{tot}}$, 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 20000 samples. Note that the DANCE response to the total prompt γ ray is relatively insensitive to the content of γ rays in a given sample since the γ-ray detection efficiency (84 to 88%) and the peak-to-total ratio ($\sim$55%) remain nearly constant for the γ-ray energy ranging from 150 keV to 10 MeV [9–11]. Each sample has a matching number of γ rays to M_γ, selected randomly according to the unfolded γ-ray energy distributions [11,13] with the condition

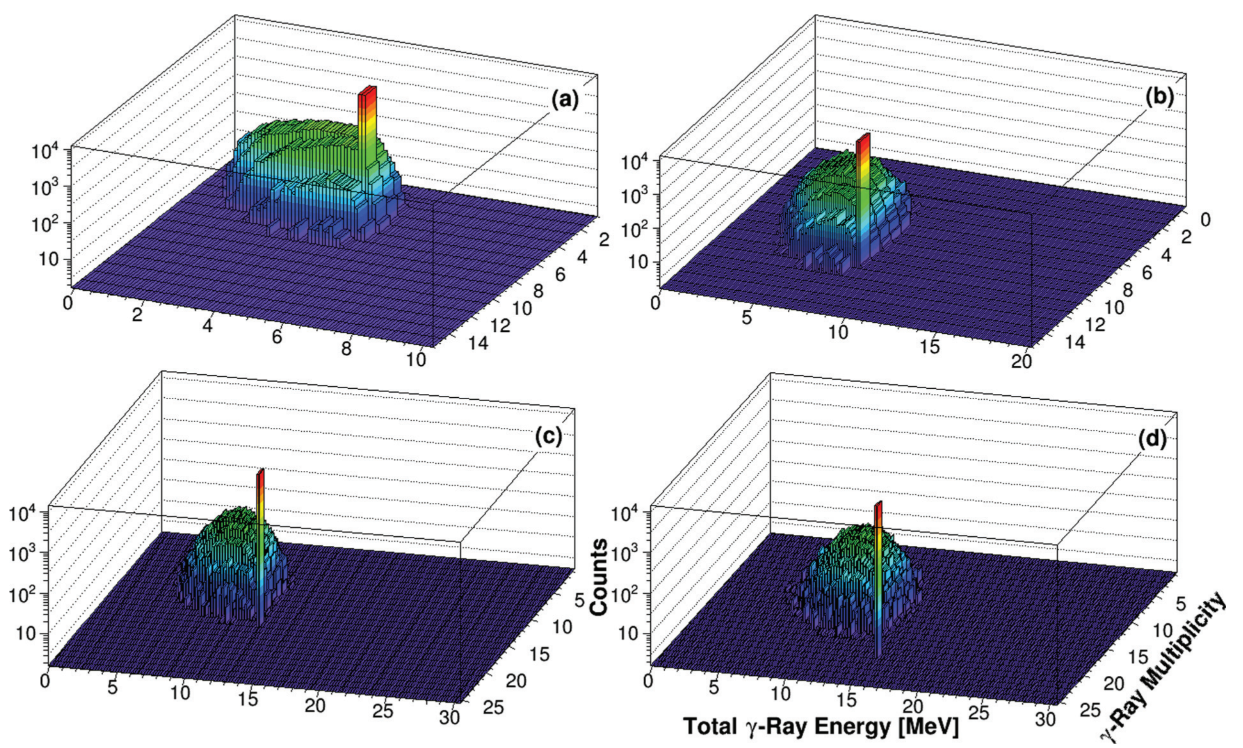

FIG. 2. (Color) The simulated DANCE response matrices for the total γ-ray energy vs multiplicity with the grid point ($E_{\gamma,\text{tot}}$, M_γ) at (5 Mev, 5), (8 MeV, 10), (12 MeV, 15), and (15 MeV, 20), shown in panels (a), (b), (c), and (d), respectively.

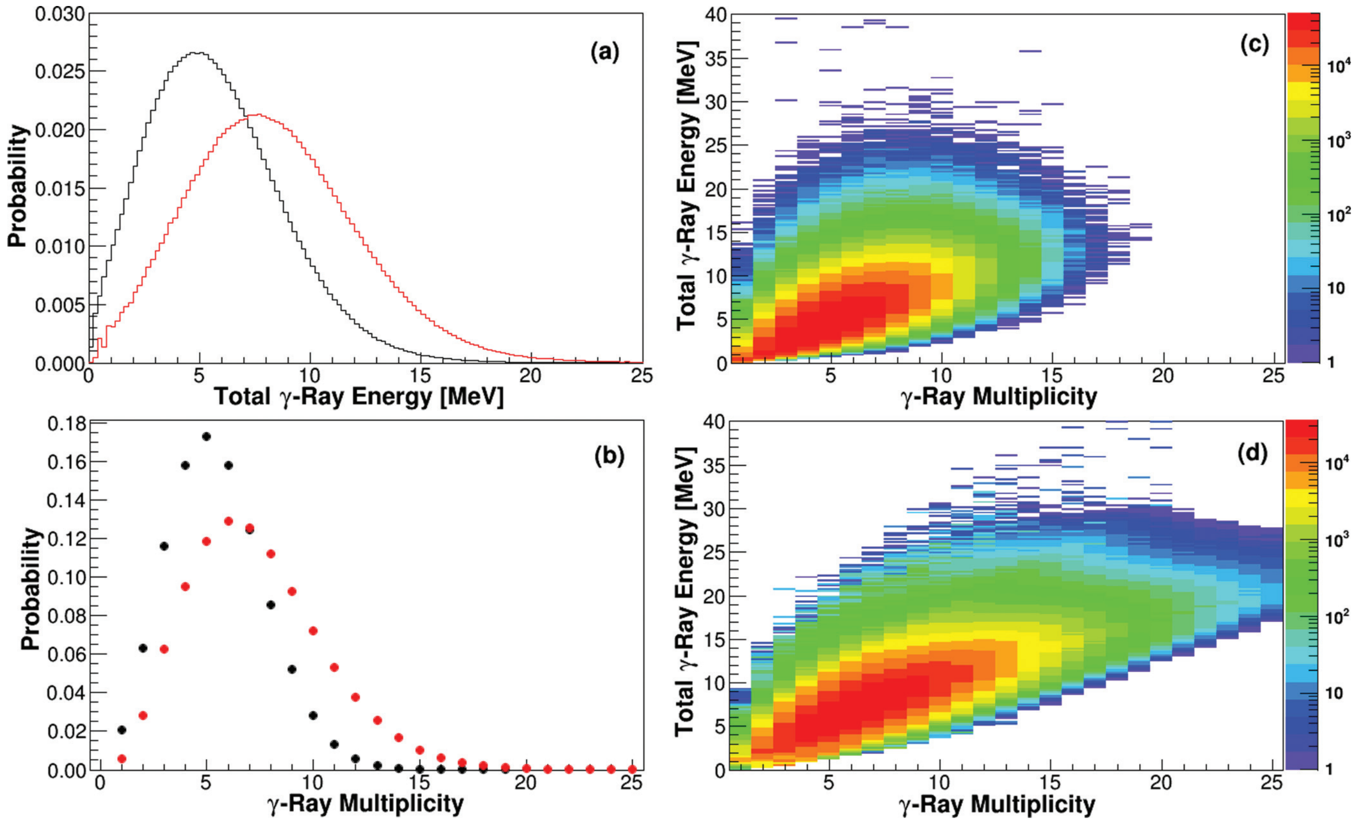

FIG. 3. (Color) Shown in panels (c) and (d), respectively, are the measured and unfolded total prompt γ-ray energy vs multiplicity distribution for the neutron-induced fission in ^{235}U. Comparisons of the projected total γ-ray energy and multiplicity distributions between measured (in black) and unfolded ones (in red [gray]) are given in panels (a) and (b), respectively.

on the total γ-ray energy that is equal to $E_{\gamma,\mathrm{tot}} \pm 100$ keV. This simulation is repeated for all the grid points within the lower and upper bounds of $E_{\gamma,\mathrm{tot}}$ for a given M_γ, established by this random sampling technique. Examples of the response matrices for a few selected pixels are shown in Fig. 2, where the DANCE response matrices for $(E_{\gamma,\mathrm{tot}}, M_\gamma) = (5$ MeV, 5), (8 MeV, 10), (12 MeV, 15), and (15 MeV, 20) are given.

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

III. RESULTS AND DISCUSSIONS

Typically it takes about 30 iterations to reach the convergence in the unfolding of the two-dimensional spectrum of $E_{\gamma,\mathrm{tot}}$ vs M_γ using the Bayesian method. The results for the neutron-induced fission in ^{235}U are shown in Fig. 3, where the unfolded $E_{\gamma,\mathrm{tot}}$ vs M_γ spectrum together with the measured one are given. In addition, the comparisons of the projected $E_{\gamma,\mathrm{tot}}$ 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 $E_{\gamma,\mathrm{tot}}$ and M_γ distributions increases noticeably after the unfolding.

Shown in Fig. 4 is the comparison of the unfolded M_γ distribution between the current work and the early one using the one-dimension unfolding technique [13] for all isotopes studied. For ^{235}U, the current mean value of 7.35 is 0.37 higher than 6.98 in the earlier work [13]. However, the latter value is known to be underestimated by about 0.3 [13]. 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 the current work. In addition, the agreement of M_γ distribution between the measurement and a simulation is much improved by using the current projected M_γ distribution compared to the one derived from the one-dimensional unfolding technique. Our mean M_γ is higher than 6.60(10), the weighted average of previous measurements [21], and lower than 8.19(11), the most recent measurement [6]. However, it is consistent with 7.04, the evaluated date listed in ENDF/B-VII.1 [22].

For ^{235}U, the current derived mean $E_{\gamma,\mathrm{tot}}$ of 8.35 MeV is higher than 6.53(20) MeV, the weighted average of previous measurements [21], 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]. Similar results are obtained for the neutron-induced fission in ^{239,241}Pu and the spontaneous fission in ^{252}Cf. Comparisons of their projected $E_{\gamma,\mathrm{tot}}$ and M_γ distributions for both measured and unfolded ones are given in Fig. 5. A model-dependent analysis of the same data for ^{239}Pu yields $\langle E_{\gamma,\mathrm{tot}}\rangle = 7.46$ MeV and $\langle M_\gamma\rangle = 7.15$ [14],

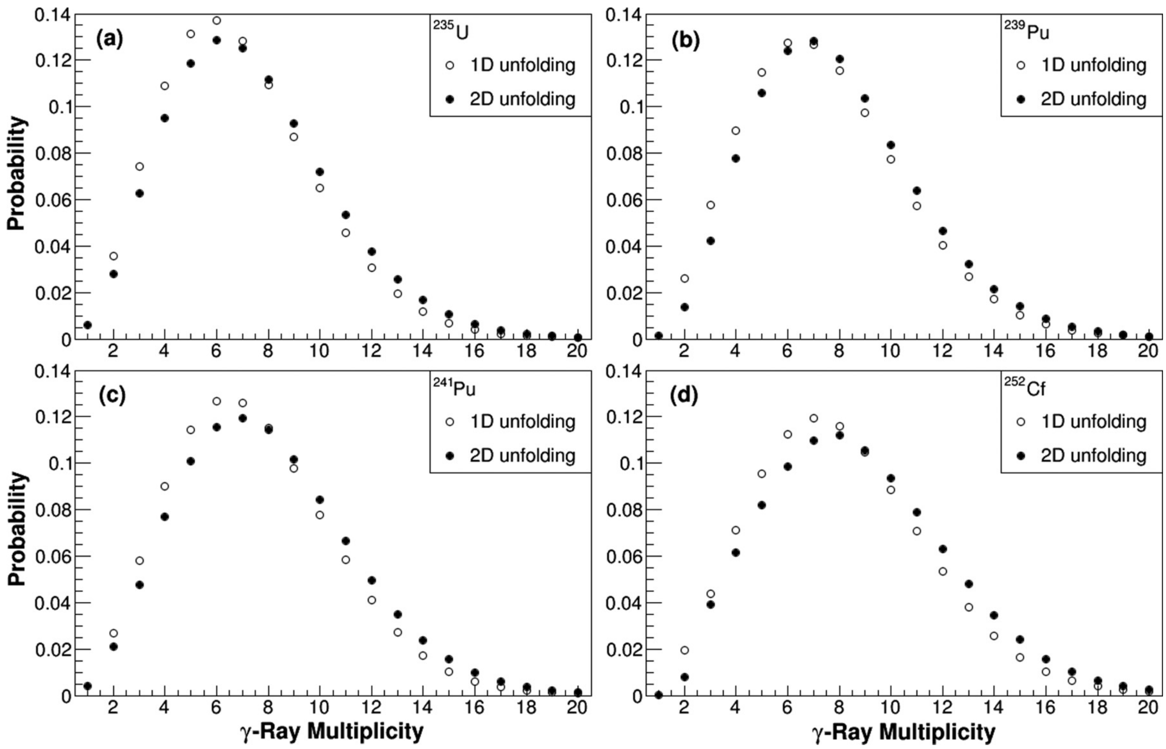

FIG. 4. Comparisons of the M_γ distribution for the neutron-induced fission in (a) ^{235}U, (b) ^{239}Pu, (c) and ^{241}Pu and the spontaneous fission in (d) ^{252}Cf using the one-dimensional (open circle) and two-dimensional (solid circle) unfolding techniques. The result derived from the latter is believed to be more precise.

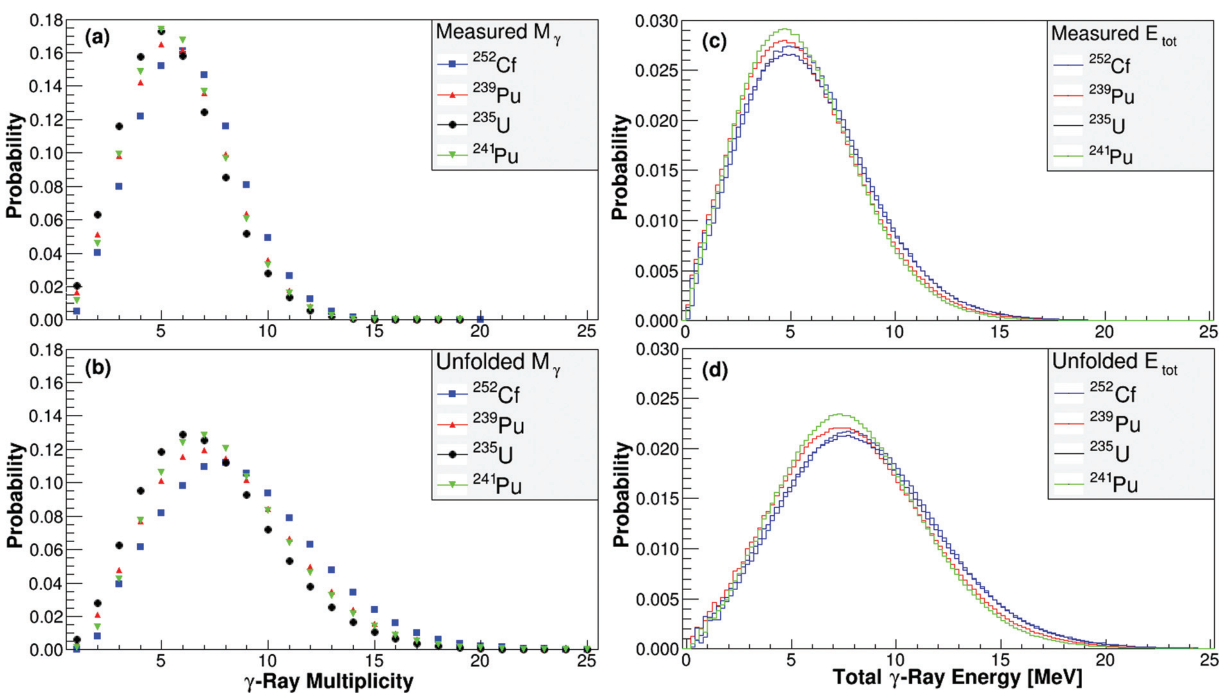

FIG. 5. (Color) Comparison of the projected γ-ray multiplicity distributions for the neutron-induced fission in ^{235}U and ^{239,241}Pu as well as the spontaneous fission in ^{252}Cf, for (a) the measured ones and (b) the unfolded ones. The same comparison for the total γ-ray energy is given for (c) the measured ones and (d) the unfolded ones.

TABLE I. Comparison of the mean $E_{\gamma,\mathrm{tot}}$ and M_γ between the current and previous measurements for the neutron-induced fission in ^{235}U and ^{239,241}Pu as well as the spontaneous fission in ^{252}Cf.

Isotope	$\langle E_{\gamma,\mathrm{tot}}\rangle$				$\langle M_\gamma\rangle$			
	Current	ENDF/B-VII.1	Ref. [21]	Ref. [6]	Current	ENDF/B-VII.1	Ref. [21]	Ref. [6]
^{235}U	8.35	6.60	6.53(20)	6.92(9)	7.35	7.04	6.60(10)	8.19(11)
^{239}Pu	7.94	6.74	6.78(10)		7.93	7.78	7.06(20)	
^{241}Pu	8.01	7.26			7.97	8.18		
^{252}Cf	8.52		6.95(30)	6.64(8)[a]	8.75		7.98(40)	8.30(8)[a]

[a]Reference [1].

compared to 7.94 MeV and 7.93, respectively, derived from the current analysis.

The means of $E_{\gamma,\mathrm{tot}}$ and M_γ derived from the projected distributions of the unfolded two-dimensional spectrum of the total γ-ray energy vs multiplicity are listed in Table I together with previous measurements and evaluated data. For $\langle E_{\gamma,\mathrm{tot}}\rangle$, our measurements are consistently higher than the previous ones [1,6,21] by $\sim$20% for all isotopes studied. The evaluated data for ^{241}Pu were derived from the systematics. The current work provides the first such a measurement. The uncertainty for our derived $\langle E_{\gamma,\mathrm{tot}}\rangle$ is dominated by the systematic error and roughly estimated to be better than 5%, assuming a similar uncertainty to that of the derived $\langle M_\gamma\rangle$.

For the $\langle M_\gamma\rangle$, our measurements are consistently higher than the previous ones [21] by $\sim$10% for all isotopes studied except for the most recent measurements [1,6], where their $\langle M_\gamma\rangle$ is $\sim$11% higher than ours in ^{235}U but $\sim$5% lower in ^{252}Cf. However, ours are consistent with the evaluated data listed in ENDF/B-VII.1. The uncertainty for our derived $\langle M_\gamma\rangle$ is estimated to be about 0.3–0.4.

IV. 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 $\langle E_{\gamma,\mathrm{tot}}\rangle$ derived from the projected $E_{\gamma,\mathrm{tot}}$ distribution of the unfolded $E_{\gamma,\mathrm{tot}}$ vs M_γ spectrum is about 20% higher than the previous measurements for all fissile nuclei studied. The current 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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[1] R. Billnert, F.-J. Hambsch, A. Oberstedt, and S. Oberstedt, Phys. Rev. C **87**, 024601 (2013).
[2] G. Rimpault, in *Nuclear Data Needs for Generation IV*, edited by P. Rullhusen (World Scientific, Antwerp, 2006), p. 46.
[3] G. Rimpault, A. Courcelle, and D. Blanchet, comment to "Nuclear Data High Priority Request List of the NEA": Req. ID H.3 and H.4.
[4] Nuclear Data High Priority Request List of the NEA (Req. ID: H.3, H4), http://www.oecd-nea.org/dbdata/hprl/hprlview.pl?ID=421 and http://www.oecd-nea.org/dbdata/hprl/hprlview.pl?ID=422
[5] V. V. Verbinski, H. Weber, and R. E. Sund, Phys. Rev. C **7**, 1173 (1973).
[6] A. Oberstedt *et al.*, Phys. Rev. C **87**, 051602(R) (2013).
[7] M. Heil, R. Reifarth, M. M. Fowler, R. C. Haight, F. Kappeler, R. S. Rundberg, E. H. Seabury, J. L. Ullmann, and K. Wisshak, Nucl. Instrum. Methods Phys. Res. A **459**, 229 (2001).
[8] R. Reifarth *et al.*, IEEE Trans. Nucl. Sci. **53**, 880 (2006).
[9] R. Reifarth *et al.*, Los Alamos National Laboratory Report No. LA-UR-01-4185, 2001 (unpublished).
[10] R. Reifarth *et al.*, Los Alamos National Laboratory Report No. LA-UR-03-5560, 2003 (unpublished).
[11] A. Chyzh *et al.*, Phys. Rev. C **85**, 021601(R) (2012).
[12] C. Y. Wu, A. Chyzh, E. Kwan, R. Henderson, J. Gostic, D. Carter, T. Bredeweg, A. Couture, M. Jandel, and J. Ullmann, Nucl. Instrum. Methods Phys. Res. A **694**, 78 (2012).
[13] A. Chyzh *et al.*, Phys. Rev. C **87**, 034620 (2013).
[14] J. L. Ullmann *et al.*, Phys. Rev. C **87**, 044607 (2013).
[15] M. Jandel, T. A. Bredeweg, A. Couture, J. M. O'Donnel, and J. L. Ullmann, Los Alamos National Laboratory Report No. LA-UR-12-21171, 2012 (unpublished).
[16] G. D'Agostini, Nucl. Instrum. Methods Phys. Res. A **362**, 487 (1995).

[17] G. D'Agostini, A&A **520**, A45 (2010).
[18] T. J. Adye, *Proceedings of the PHYSTAT 2011 Workshop, CERN, Geneva*.
[19] S. Agostinelli *et al.*, Nucl. Instrum. Methods Phys. Res. A **506**, 250 (2003).
[20] M. Jandel *et al.*, Nucl. Instrum. Methods Phys. Res. B **261**, 1117 (2007).
[21] T. E. Valentine, Ann. Nucl. Energy **28**, 191 (2001), and references therein.
[22] M. B. Chadwich *et al.*, Nucl. Data Sheets **112**, 2887 (2011).