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LLNL-JRNL-646377

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November 18, 2013

Physical Review C

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Total prompt γ-ray emission in fission

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The total prompt γ-ray energy distributions for the neutron-induced fission in ^{235}U, ^{239,241}Pu, and the spontaneous fission in ^{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.

PACS numbers: 25.85.Ec,29.30.Kv,29.40.Cs

I. INTRODUCTION

The total prompt γ-ray emission accounts for about 40% of the total γ-ray emission that makes up about 10% of the total energy released in fission [1]. 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) [2]. 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 [4, 5]. 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 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 [6]. In the past, 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 used by Verbinski *et al.* [7] nearly 40 years ago, or the recent efforts by Billnert *et al.* [2] and Oberstedt *et al.* [3] using the cerium-doped $LaBr_3$, $CeBr_3$, and $LaBr_3$ detectors.

A better approach for such a measurement is to use a γ-ray calorimeter such as the DANCE array [8, 9] which 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). 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 counter (PPAC) [10]. The results on the measured and unfolded fission prompt γ-ray energy and multiplicity distributions for those isotopes have been published [11–13]. In this article, we report the total prompt γ-ray energy distributions for those isotopes, obtained by unfolding the measured two-dimension 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.

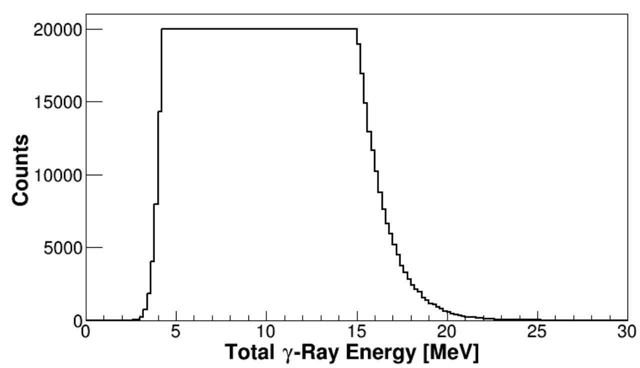

FIG. 1: Spectrum for the number of sample with a set of matching number of γ rays to $M_\gamma = 8$ as a function of $E_{\gamma,tot}$ for the neutron-induced fission in ^{235}U.

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 the year of 2010 and 2011 at the Lujan Center of LANSCE. The experimental setup and the data analysis has been described in details in our early publications [11–13]. 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 H^- 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. A minimum of 10^6 fission events with at least one γ ray detected by DANCE were collected for all isotopes studied.

The summed energy of all γ rays detected by DANCE within a time window of 40 ns is defined as the total prompt γ-ray energy ($E_{\gamma,tot}$) in fission for a given event. Note that both DANCE and PPAC have a similar time resolution of ∼1.2 ns [10]. The total γ-ray multiplicity (M_γ) in fission is established not according to the number of detectors detecting 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 [11, 14, 15]. 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 [11, 12].

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-dimension spectrum of $E_{\gamma,tot}$ vs. M_γ. The two-dimension 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 [16, 17]. The DANCE response matrix for $E_{\gamma,tot}$ vs. M_γ is simulated using the GEANT4 geometrical [18] model of both DANCE and PPAC [11, 12, 19]. To make sure this two-dimension 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.

For any given grid point ($E_{\gamma,tot}$, M_γ) in the response matrix, no more than 20,000 samples are assembled and each sample has a matching number of γ rays to M_γ, selected randomly according to the unfolded γ-ray energy distributions [11, 12] with the condition on the total γ-ray energy that is equal to $E_{\gamma,tot}\pm$ 100 keV. A two-dimension DANCE response matrix with a size of 200×25 for this grid point is generated using GEANT4 with the given assembly. With this random sampling technique, a natural lower and upper bound for $E_{\gamma,tot}$ is established for a given M_γ and shown in Fig. 1, where the number of samples with a set of matching number of γ rays to M_γ is plotted as a function of $E_{\gamma,tot}$ for the neutron-induced fission in ^{235}U. 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 (∼55%) remain nearly constant for the γ-ray energy ranging from 150 keV to 10 MeV [11, 14, 15].

This simulation is repeated for all the grid points within the bounds of any given M_γ as shown in Fig. 1. A DANCE response matrix is created and consists of ∼3,300 two-dimension matrices with a size of 200×25 each. Examples of the response matrices for a few selected grid points are shown in Fig. 2, where the DANCE response matrices for ($E_{\gamma,tot}$, M_γ) = (5 MeV, 5), (8 MeV, 10), (12 MeV, 15), and (15 MeV, 20) are given. This numerically simulated DANCE response matrix is generated for each isotope studied and used to unfold the measured two-dimension spectrum of $E_{\gamma,tot}$ vs. M_γ into a physical one using the iterative Bayesian method. During the iteration stage, the identity of the response matrix for any given grid point is kept and 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-dimension spectrum of $E_{\gamma,tot}$ 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 $E_{\gamma,tot}$ vs. M_γ spectrum together with the measured one is given. In addition, the comparison of the projected $E_{\gamma,tot}$ and M_γ distributions between the unfolded and measured ones is 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.

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 [12] for all isotopes studied. For ^{235}U, the current mean value of 7.35 is 0.37 higher than the early one of 6.98. However, the latter value is known to be underestimated by about 0.3 [12]. This consistence in the derived mean M_γ from both the one- and two-dimension 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-dimension unfolding technique. Our mean M_γ is higher than 6.60(10), the weighted average of previous measurements [20], and lower than 8.19(11), the most recent measurement [3].

For ^{235}U, the current derived mean $E_{\gamma,tot}$ of 8.35 MeV is higher than 6.53(20) MeV, the weighted average of previous measurements [20], and 6.92(9) MeV, the most recent measurement [3]. Our result is about 20% higher than those two values, which is very significant and almost makes up the deficit of the estimated γ heating in nuclear reactors using the evaluated data [2]. The two lower mean $E_{\gamma,tot}$ are closed to the evaluated data. Similar results are obtained for the neutron-induced fission in ^{239,241}Pu and the spontaneous fission in ^{252}Cf. Comparisons of their projected $E_{\gamma,tot}$ and M_γ distributions for both measured and unfolded ones are given in Fig. 5.

The mean of $E_{\gamma,tot}$ and M_γ derived from the projected distributions of the unfolded two-dimension spectrum of the total γ-ray energy vs. multiplicity are listed in Table I together with previous measurements. For the $<E_{\gamma,tot}>$, our measurements are consistently higher than the previous ones [2, 3, 20] by ∼20% for all isotopes studied. The uncertainty for our derived $<E_{\gamma,tot}>$ is estimated to be better than 5%. We believe the current measurements has a very significant impact on modeling the γ heating in the reactor fuel core, since there already is the evidence that the γ heating is underestimated by up to 28% [2].

For the $<M_\gamma>$, our measurements are consistently higher than the previous ones [20] by ∼10% for all isotopes studied but cannot be generalized in the comparison with the most recent measurements [2, 3]. The uncertainty for our derived $<M_\gamma>$ is estimated to be about 0.3-0.4. Note that the current $<E_{\gamma,tot}>$ and $<M_\gamma>$, derived from the projected distributions of the unfolded two-dimension spectrum of the total γ-ray energy vs. multiplicity, have the same magnitude of their respective FWHMs. This raises the question of the suitability in using the mean values and their uncertainties to quantify the uncertainty of the γ heating in reactors. It is our opinion that the unfolded or the physical two-dimension spectrum of the total γ-ray energy vs. multiplicity should be used for such applications.

IV. SUMMARY

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 the 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-dimension unfolding technique, numerically implemented by adopting the iterative Bayesian method. The $<E_{\gamma,tot}>$ 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. This is a very significant development since it almost accounts for the estimated deficit in the γ heating in reactors using the evaluated data. It is our opinion that the quantification of the uncertainty of the γ heating in reactors should be performed by using the total γ-ray energy vs. multiplicity spectrum since the mean value of both $E_{\gamma,tot}$ and M_γ has the same magnitude of their respective FWHMs.

This work benefited from the use of the LANSCE accelerator facility as was performed under the auspices of the US Department of Energy by Lawrence Livermore National Security, LLC under contract DE-AC52-07NA27344 and by Los Alamos National Security, LLC under contract DE-AC52-06NA25396. Partial funding is gratefully acknowledged from the U.S. DOE/NNSA Office of Defense Nuclear Nonproliferation Research and Development. All isotopes used in the measurements were obtained from Oak Ridge National Laboratory.

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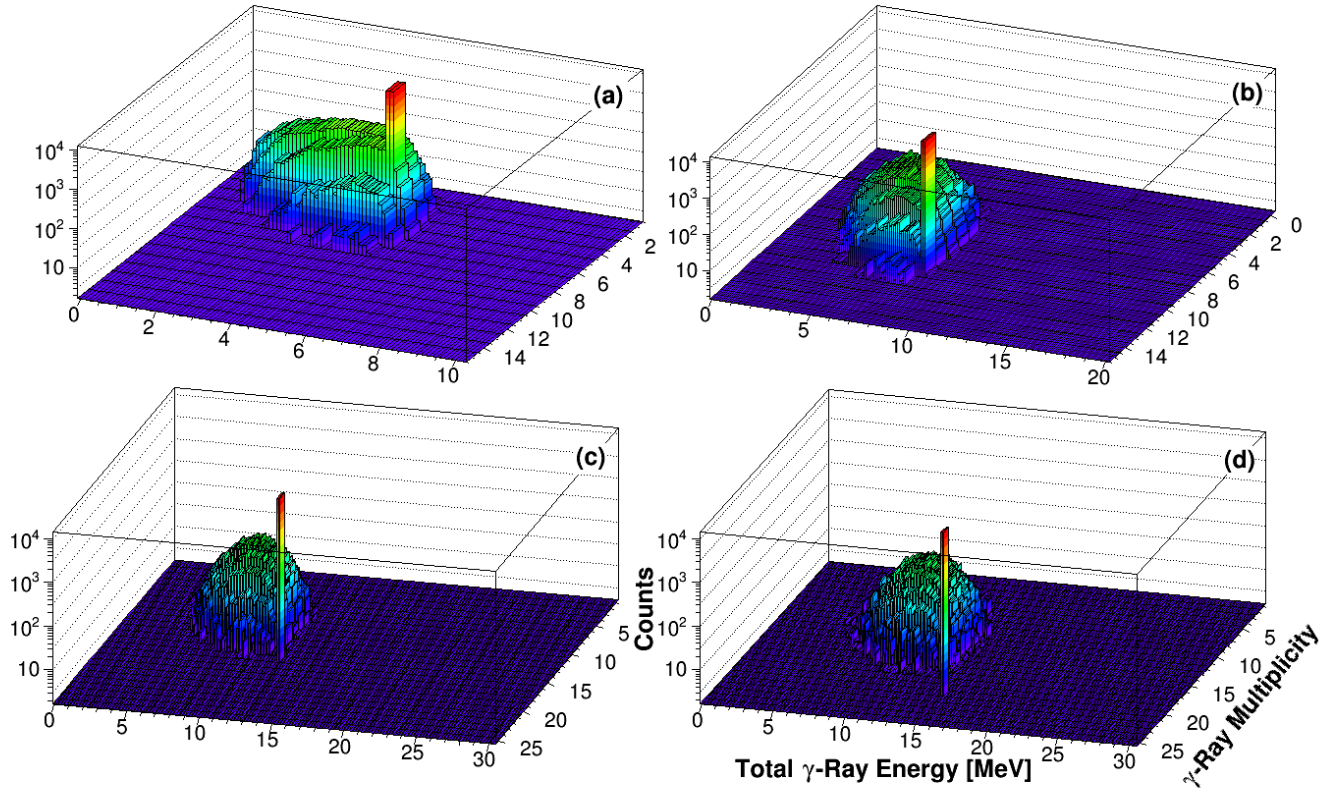

FIG. 2: The simulated DANCE response matrix for the total γ-ray energy vs. multiplicity with the grid point ($E_{\gamma,tot}$, M_γ) at (5 Mev,5), (8 MeV, 10), (12 MeV, 15), and (15 MeV, 20), shown in (a), (b), (c), and (d), respectively.

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

Isotope	$<E_{\gamma,tot}>$			$<M_\gamma>$		
	Current	Ref. [20]	Ref. [3]	Current	Ref. [20]	Ref. [3]
^{235}U	8.35	6.53(20)	6.92(9)	7.35	6.60(10)	8.19(11)
^{239}Pu	7.94	6.78(10)		7.93	7.06(20)	
^{241}Pu	8.01			7.97		
^{252}Cf	8.52	6.95(30)	6.64(8)*	8.75	7.98(40)	8.30(8)*

* Ref. [2]

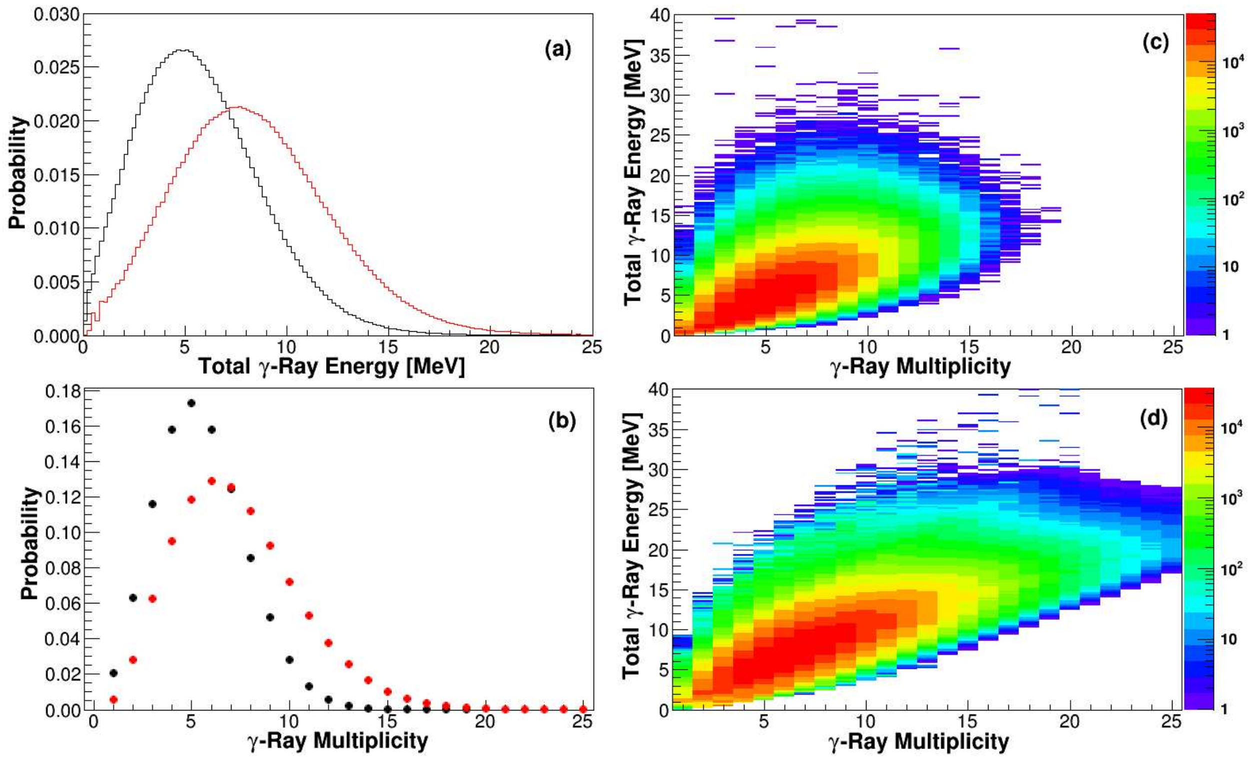

FIG. 3: Shown in (c) and (d), respectively, are the measured and unfolded total prompt γ-ray energy vs. multiplicity distribution for the neutron-induced fission in ^{235}U. Comparison of the projected total γ-ray energy and multiplicity distributions between measured and unfolded ones is given in (a) and (b), respectively.

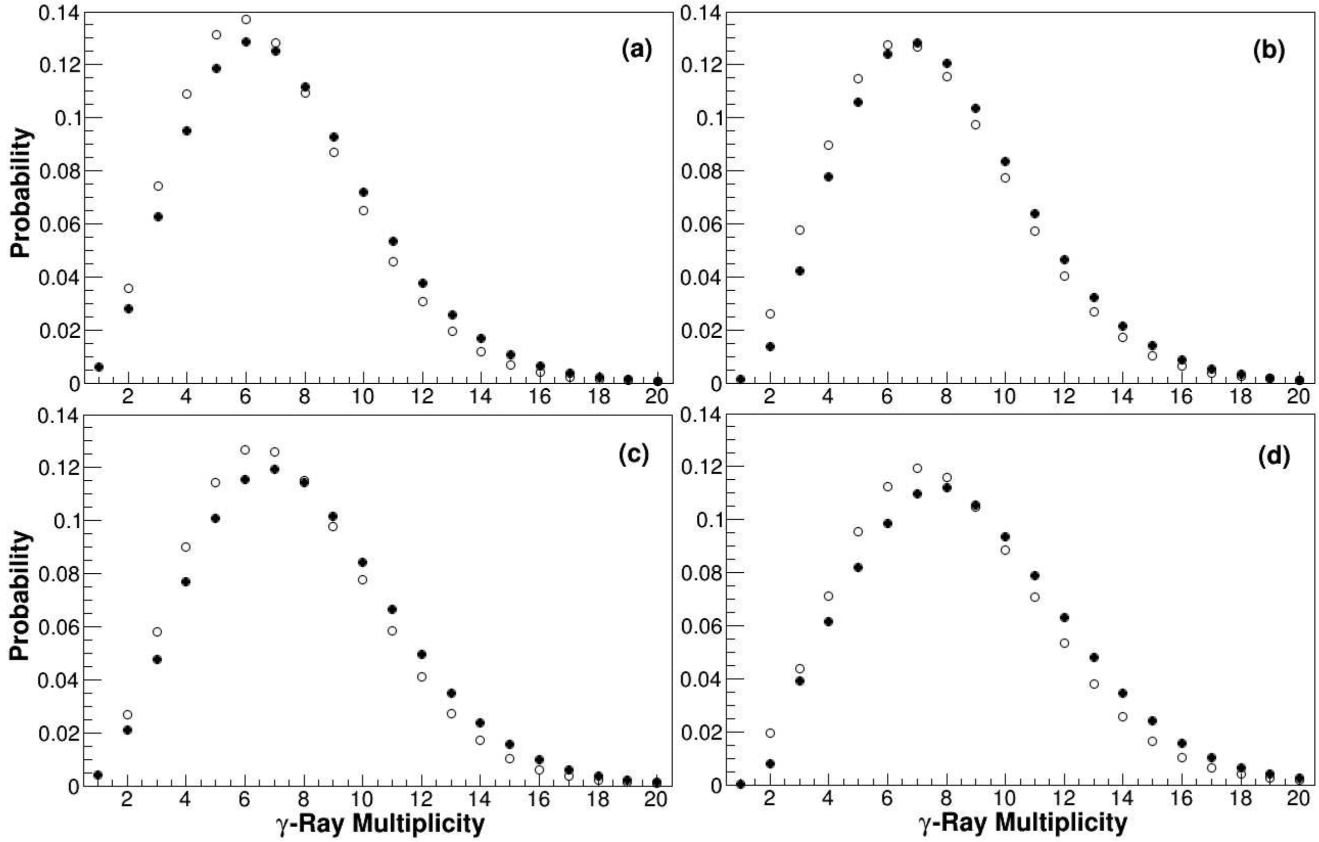

FIG. 4: Comparison of the M_γ distribution for the neutron-induced fission in (a) ^{235}U, (b) ^{239}Pu, (c) ^{241}Pu, and the spontaneous fission in (d) ^{252}Cf using one and two-dimension unfolding techniques. The result derived from the latter is believed to be more precise.

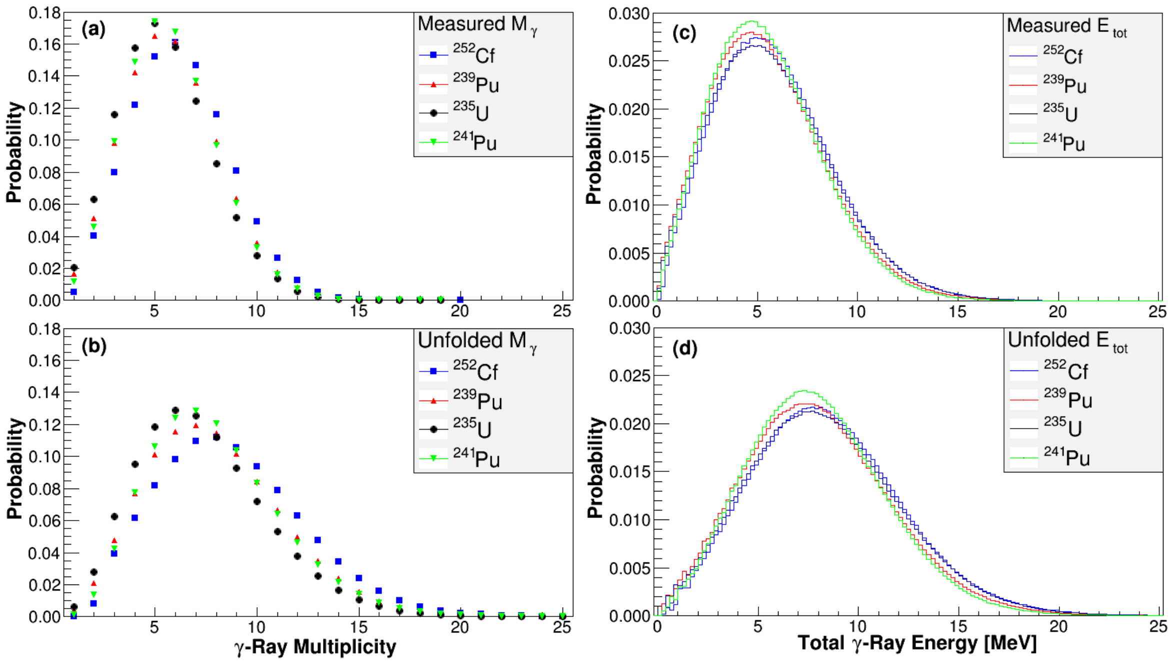

FIG. 5: Comparison of the projected γ-ray multiplicity distributions for the neutron-induced fission in ^{235}U and ^{239,241}Pu as well as the spontaneous fission in ^{252}Cf, is given in (a) for the measured ones and (b) for the unfolded ones. The same comparison for the total γ-ray energy is given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