Systematics of prompt γ -ray emission in fission

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The prompt γ -ray energy and multiplicity distributions were measured for the neutron-induced fission in ²³⁵U and ^{239,241}Pu by using a highly segmented $4\pi \gamma$ -ray calorimeter in coincidence with the detection of fission fragments by a gas-filled parallel-plate avalanche counter. Both distributions were unfolded according to the detector response, which was simulated numerically by using a model validated with the γ -ray calibration sources. The mean value and the width of the γ -ray multiplicity distribution show a systematic increase with increasing mass of fissile nucleus, whereas, the energy distribution shows the dependence of γ -ray energy above 5 MeV on the species of fissile nuclei. The correlations between γ -ray energy and multiplicity were studied by comparing the mean value and the width of the total γ -ray energy between measurement and simulation by using an assembly with elements selected by random sampling of their unfolded distributions. The detector response was taken into account in the simulation. These results together with the detailed description of the experiment and analysis are presented.

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I. INTRODUCTION

The prompt γ -ray emission is the least studied process among the main energy-release mechanisms in fission, such as fission fragments and neutron emission. Even among the existing data for emitted γ rays [1], only limited measurements and modeling of the energy and multiplicity distributions were attempted. These data are important for many applications in the nuclear energy industry among others. With the advent of modern highly segmented highly efficient $4\pi \gamma$ -ray detector arrays, such as the Heidelberg-Darmstadt Crystal Ball [2] and the Detector for Advanced Neutron Capture Experiments (DANCE) [3,4] at the Los Alamos Neutron Science Center (LANSCE), precision measurements become feasible for the prompt γ -ray energy (E_{γ}) and multiplicity (M_{γ}) distributions in fission. In fact, for spontaneous fission of 252 Cf, prompt E_{ν} and M_{ν} together with the neutron multiplicity as a function of fission-fragment mass were measured by using the Crystal Ball [5,6], and both E_{γ} and M_{γ} distributions as well as their correlations were studied by using the DANCE array [7].

For the prompt E_{γ} distribution in spontaneous fission of ²⁵²Cf measured by the Crystal Ball, the γ rays with energies between 3.5 and 8.0 MeV, gated on the fission-fragment mass near 130, were significantly enhanced over those gated on other mass regions [5,6]. This mass-dependent enhancement also was found in the compound-nucleus fission using the fusion-fission reactions with the Crystal Ball [8]. By lowering the level density in the mass region near 130, the observed enhancement can be reproduced [6] in the statistical model calculations by using the CASCADE code [9].

For experiments using the DANCE array, the prompt M_{γ} distribution in the spontaneous fission of ²⁵²Cf was unfolded for the first time by taking advantage of the unique feature of DANCE where the M_{γ} response was nearly γ -ray energy independent [7,10,11]. Together with the unfolded E_{γ} distribution, the study of correlations between E_{γ} and M_{γ} was realized by comparing the measured total γ -ray energy distribution

to a simulation performed by random sampling of their unfolded distributions. The detector response was taken into account in the simulation. From this comparison, the stochastic aspect of prompt γ emission in the spontaneous fission is illustrated [7]. In this paper, we report the measurements of the prompt γ -ray emission in the neutron-induced fission of ²³⁵U and ^{239,241}Pu by using DANCE together with a compact gas-filled parallel-plate avalanche counter (PPAC) [12] for the fission-fragment detection. The measured spectrum resulted from the integration over the incident neutron energy from thermal to ~ 100 keV except for ²⁴¹Pu where the integration was performed from $\sim 2 \text{ eV}$ to $\sim 100 \text{ keV}$ with the exclusion of the resonances of the ²³⁹Pu contaminant. A systematic comparison of both the unfolded E_{γ} and M_{γ} distributions among the neutron-induced fission in the U and Pu isotopes and those in the spontaneous fission of ²⁵²Cf are made. The study of the correlations between the E_{γ} and the M_{γ} is also presented.

II. EXPERIMENTAL SETUP AND MEASUREMENTS

The measurements of the prompt γ emission in the neutroninduced fission of ²³⁵U and ^{239,241}Pu were performed at LANSCE by using the DANCE array in conjunction with three PPACs for the fission-fragment detection. The schematic of the experimental setups is shown in Fig. 1. Incident neutrons with energies from thermal up to several hundred keV were produced by spallation induced by an 800-MeV proton beam at a repetition rate of 20 Hz, which was impinged on a tungsten target, then was moderated by water. DANCE is located at the Lujan Neutron Scattering Center of LANSCE at the end of a 20.23-m flight path. The incident neutron energy was determined from the linac by measuring the time difference between the beam pulse and the event detected by either the DANCE or the PPAC, both of which have similar intrinsic time resolutions of ~ 1.2 ns. For the neutron-flux monitoring, three detector systems were installed about 2 m downstream



FIG. 1. (Color) A schematic of the experimental setup for the DANCE array together with a gas-filled PPAC. The PPAC with an internal target is installed inside the beam pipe at the center of the DANCE array.

of DANCE; they are a Si detector for a rate measurement of the ⁶Li(n, α) reaction, the BF₃ gas detector for the rate measurement of the ¹⁰B(n, α) reaction, and the ²³⁵U ionization chamber for the rate measurement of ²³⁵U(n, f). Details of the detector systems together with the data processing used for these experiments are described below.

A. DANCE

The DANCE spectrometer is a 4π highly segmented highly efficient γ -ray calorimeter. It consists of 160 BaF₂ scintillation crystals, and each one has the same solid-angle coverage. DANCE was designed to study neutron capture reactions on small quantities of radioactive or rare stable nuclei. Additionally, it is an excellent 4π detector array useful for the study of the γ -ray spectroscopy for reactions other than capture, such as fission. It has many unique features, such as the nearly independent detection efficiency for all γ -ray energies, peak-to-total ratio, and the multiplicity response [10,11]. For example, the detection efficiency varies between 85% and 88% for γ -ray energy between 150 keV and 10 MeV, the peak-to-total ratio is about 55%, and the average multiplicity from the detector array response deviates no more than 16% at these energies. All these quantities are derived from the simulations using GEANT4 [13] with a geometric model of DANCE together with the surrounding materials. A model validation was carried out by comparison between the measurement and the simulation by using GEANT4 with the γ -ray calibration sources ²²Na, ⁶⁰Co, and ⁸⁸Y [10,11,14,15].

One of those unique features mentioned above is illustrated in Fig. 2 where the γ -ray multiplicity responses are shown for the five γ rays with the same incident energies of 0.5, 1.0, and 3.0 MeV. The mean values for the folded γ -ray multiplicity are 4.0, 4.16, and 4.22, respectively. The variation in the mean value is no more than 6%. This unique property enables us to approximately unfold the prompt γ -ray multiplicity



FIG. 2. (Color online) The DANCE response for events with $M_{\gamma} = 5$ and the individual γ energy at 0.5, 1.0, and 3.0 MeV, shown in black solid, blue dots, and red dashes, respectively.

distributions and gives an opportunity to study correlations between E_{γ} and M_{γ} in fission [7]. The energy and time of the detected γ ray are derived from the recorded digital waveforms. Besides the E_{γ} and M_{γ} distributions, DANCE can also measure the total γ -ray energy for a given reaction. An example is shown in Fig. 3 where the total γ -ray energy vs multiplicity is plotted for the neutron-induced fission of ²³⁵U.

B. PPAC and targets

DANCE is a γ -ray calorimeter and is designed to study the neutron capture reactions by summing all the detected γ rays. However, the origin of individual γ rays detected by DANCE cannot be established without additional information, in particular, for fissile nuclei where the fission channel competes favorably with capture. The uniqueness of the fission prompt γ rays can be established if a PPAC is used to tag the detected γ rays by detecting the fission fragments. To accomplish this, a compact gas-filled PPAC [12], designed specifically for DANCE, was fabricated at Lawrence Livermore National Laboratory (LLNL).

The PPAC consists of two electrically connected anodes and one cathode. The anodes are made of 1.4- μ m-thick aluminized mylar and are located 3 mm away from either side of the cathode. The cathode is made of 3- μ m-thick titanium foil with the target material electroplated on both sides then sandwiched



FIG. 3. (Color) The measured total γ -ray energy vs cluster multiplicity for the neutron-induced fission of ²³⁵U.



FIG. 4. (Color online) The pulse height spectrum for the chargedparticles detected by the PPAC in the ²³⁵U (a) and ²⁴¹Pu (b) experiment is shown in blue dashes. The one with an additional requirement of at least one γ ray being detected by DANCE is shown in red solid and the α peak at the channel ~22 drops significantly with this condition for ²⁴¹Pu. No significant change for ²³⁵U is observed.

between two mylar foils of the same thickness. The technique for the double-sided target fabrication is described in Ref. [16]. The PPAC was operated with a gas pressure at ~ 4 Torr, stabilized by a feedback system by regulating the flow of isobutane. The anode was biased at about +400 V, and the signal was processed by an amplifier with a gain of \sim 300 and a bandwidth of 500 MHz before it was sent to the digitizer of the same model used for the DANCE detector array. The pulse height of about several hundred millivolts and the average rise time of \sim 3 to 4 ns are derived for the detected fission fragments from the recorded digital waveforms. Since all the experiments were carried out in the inclusive mode, the PPAC detection efficiency can be estimated by comparing the events with the total γ -ray (energy, multiplicity) \geq (10 MeV, 8) to those with an additional requirement on the detection of fission fragments. An average PPAC detection efficiency \sim 70% was achieved. No attempt was made to resolve the fission-fragment mass by the current design of the PPAC.

For a typical PPAC, fission fragments can easily be distinguished from α 's because of the disparity of their energy loss in the counter gas. However, in the current PPAC design with the internal target, the separation between them is not sharp due to the strong angular dependence of flight paths of fission fragments and α 's. This can be observed in Fig. 4 where the fission fragments are approximately separated from α 's in the pulse-height spectrum for the experiment with the ²⁴¹Pu



FIG. 5. The time difference between γ rays detected by DANCE and charged particles detected in the PPAC for the ²³⁵U (a) and ²⁴¹Pu (b) experiment, where the time resolution ~1.7 ns is achieved with the peak at the channel ~-45. The bump next to the peak is related to events with an ambiguous correlation between DANCE and PPAC.

target. The ratio of fission fragments to α 's is further improved by nearly an order of magnitude by requiring, at least, one γ ray to be detected by DANCE.

For fission events, the identification of γ rays detected by DANCE is established with conditions set not only on the pulse-height spectrum, shown in Fig. 4, but also on the time spectrum between γ rays detected by DANCE and charged particles detected by the PPAC. Such a time spectrum for the ²³⁵U target is shown in Fig. 5 where the time resolution of about 1.7 ns was observed. With these constraints on both the pulse height and the time spectra, the fission prompt E_{γ} and M_{γ} distributions in addition to the total γ -ray energy vs multiplicity are derived. An example of the latter is shown in Fig. 3. The quantitative description of these constraints is given in the next section.

C. Data acquisition and processing

The data acquisition hardware for DANCE is based on the 8-bit Acquiris DC265 digitizers with a sample rate of 500 MHz and a bandwidth of 150 MHz. The front-end software was developed under the MIDAS framework [17]. The off-line analysis was performed by using the customized software [18]. The details are described in Refs. [19,20]. Valid events were taken when two or more γ rays above the 150-keV threshold were detected by DANCE within a 100-ns time coincidence window. For each BaF₂ scintillator of DANCE, two digitizer channels with different gains were used to record the waveforms to maximize the advantage in determining the time and pulse-height information from the fast and slow components, respectively. In addition, their ratios can be used to distinguish γ ray from α decay, which originates from the impurity of Ra isotopes in the BaF₂ crystal [21]. The time and pulse height of the detected charged particle by the PPAC were derived from the waveforms recorded by the same model of digitizer.

The fission prompt E_{γ} and M_{γ} distributions are derived by gating on the pulse-height spectrum of charged particles detected by the PPAC, shown in Fig. 4 as well as on both the coincidence time window for DANCE and the time spectrum between DANCE and PPAC, shown in Fig. 5. A time window of 40 ns is applied for the former and 8 ns (-48 to - 40 ns) for the latter. To avoid the summing effect for the E_{γ} spectrum, any γ ray detected by DANCE with adjacent crystals, which are triggered in the given 40-ns time window, was excluded. However, this condition is not applied in derivation of the total γ -ray energy spectrum.

For the M_{γ} spectrum, the multiplicity is not derived from the number of crystals that are triggered. Instead, the multiplicity for any γ ray detected by DANCE with adjacent crystals that are triggered in the given time window is counted as one. The advantage for this criterion is that (1) the overcounting of the multiplicity due to the Compton scattering can be minimized and (2) the M_{γ} response for DANCE is nearly γ -ray energy independent according to the numerical simulation that uses a geometric model of DANCE together with the surrounding materials [10,11], which leads to an opportunity to unfold the M_{γ} spectrum. Together with the unfolded E_{γ} spectrum, the correlation between the γ -ray energy and the multiplicity can be studied.

III. RESULTS AND DISCUSSIONS

In the following sections, the measured prompt E_{γ} and M_{γ} distributions together with their unfolded ones for the neutron-induced fission of ²³⁵U and ^{239,241}Pu are presented. The spectrum was derived with the incident neutron energy from thermal to about 100 keV. The unfolded distributions were determined by using both the iterative Bayesian [22] and the singular-value decomposition (SVD) [23] methods. Their numerical implementations are carried out by using the ROOT add-on software package [24] with the response matrices simulated numerically by using a geometric model of DANCE, validated with γ -ray calibration sources [10,11,14,15]. Details of this procedure have been described in Ref. [7]. Systematic comparisons are made for the unfolded prompt E_{γ} and M_{γ} distributions in the neutron-induced fission among the U and Pu isotopes together with those from the spontaneous fission of 252 Cf. Results from the study of correlations between E_{γ} and M_{ν} in fission also are presented.

A. Prompt γ emission in the neutron-induced fission of 235 U

The experiment was fielded by using the 235 U target fabricated at LLNL with an isotopic enrichment of 99.91% and a total mass of ~0.923 mg. A total of ~7.8 × 10⁶ valid fission events were collected over a period of ~16 days of beam on



FIG. 6. (Color online) The measured E_{γ} spectrum for the neutron-induced fission of ²³⁵U is shown by black crosses together with the unfolded ones, derived from the iterative Bayesian (red circles) and SVD (green triangles) methods. The results from SVD are scaled up by a factor of 5 for clarity.

target. The measured E_{γ} spectrum binned into 100 keV per channel together with the unfolded ones are shown in Fig. 6. Despite the spectrum monotonically falling as a function of energy for the γ -ray energy above 1 MeV, there are distinct bumplike structures observed at near 0.5, 1.5, 4.0, and 6.0 MeV with varied degrees of strength. A similar pattern was observed for the prompt E_{ν} spectrum for the spontaneous fission of ²⁵²Cf [7]. A discussion of physics for these structures is presented later together with those for other isotopes. The mean energy for γ rays between 0.15 and 9.50 MeV is 1.09 and 0.97 MeV for the unfolded E_{γ} distributions derived from the iterative Bayesian and SVD methods, respectively. The mean energy is consistent with 0.97(2) MeV [1], the weighted average of previous measurements. The comparison with the earlier measurement of Verbinski et al. [25] is shown in Fig. 7, and the agreement is reasonable.

We believe the iterative Bayesian method yields a morereliable unfolded spectrum since it can handle the space far beyond the truncated space observed in the measurement. This is a unique capability important for recovering the lost



FIG. 7. (Color online) The comparison of the unfolded E_{γ} distribution for ²³⁵U between the current measurement (red circles) and the early one (blue triangles) by Verbinski *et al.* [25]. The relative yields are normalized according to the intensity of the γ -ray energy between 1 and 4 MeV.



FIG. 8. (Color online) The measured M_{γ} distribution for the neutron-induced fission of ²³⁵U (filled black circles) together with the unfolded ones, derived from the iterative Bayesian (red squares) and SVD (green triangles) methods.

information for a given measurement. The difference in the derived physical value between the two unfolding approaches can be viewed as the upper bound for the uncertainty.

The measured M_{γ} distribution together with the unfolded ones are shown in Fig. 8. The mean multiplicity is 6.95 and 6.98 for the unfolded M_{γ} distributions derived from the iterative Bayesian and SVD methods, respectively. It is slightly higher than 6.60(10) [1], the weighted average of previous measurements. To verify the "goodness" of this unfolded spectrum and a rough estimate of the systematic uncertainty introduced by ignoring the slight energy dependence of the γ -ray multiplicity response of DANCE, a simulation was performed by using an assembly with elements selected from a random sampling of the unfolded E_{γ} and M_{γ} distributions and then folded with the DANCE response. Results from this simulation together with the measured one are shown in Fig. 9. By shifting the unfolded M_{γ} distribution to a higher multiplicity by 0.3, an excellent agreement between the measurement and the simulation is achieved. Thus, we



FIG. 9. (Color online) The comparison of the M_{γ} distribution between the measurement (shown by filled black circles) and two simulations for ²³⁵U (magenta solid and green dashed lines). The results from the simulation with the unfolded M_{γ} distribution are shown by the magenta solid line, and the one with a correction to the unfolded distribution by shifting to the higher multiplicity by 0.3 is shown by the green dashed line.



FIG. 10. (Color online) The measured E_{γ} spectrum for the neutron-induced fission of ²³⁹Pu is shown by crosses together with the unfolded ones, derived from the iterative Bayesian (circles) and SVD (triangles) methods. The results from SVD are scaled up by a factor of 5 for clarity.

believe the systematic uncertainty of this paper is on the order of 0.3 for the unfolded M_{γ} distribution in the neutron-induced fission of ²³⁵U.

B. Prompt γ emission in the neutron-induced fission of ²³⁹Pu

The experiment was fielded by using the ²³⁹Pu target fabricated at LLNL with an isotopic enrichment of 99.97% and a total mass of ~0.937 mg. A total of ~2.8 × 10⁶ valid fission events were collected in a period of ~17 days of beam on target. The measured spectrum for the fission prompt γ rays together with the unfolded ones are shown in Fig. 10. The bumplike structures, similar to those of ²³⁵U, also are observed in the E_{γ} spectrum. The mean value for the γ -ray energies between 0.15 and 9.5 MeV is 0.98 and 0.86 MeV for the unfolded E_{γ} distributions derived from the iterative Bayesian and SVD methods, respectively. It is consistent with 0.95(3) [1], the weighted average of the previous measurements. The comparison with the previous measurement of Verbinski *et al.* [25] is shown in Fig. 11, and the agreement



FIG. 11. (Color online) The comparison of the unfolded E_{γ} distribution for ²³⁹Pu between the current measurement (red circles) and the early one (blue triangles) by Verbinski *et al.* [25]. The relative yields are normalized according to the intensity of the γ -ray energy between 1 and 4 MeV.



FIG. 12. (Color online) The measured M_{γ} distribution for the neutron-induced fission of ²³⁹Pu (filled circles) together with the unfolded ones, derived from the iterative Bayesian (squares) and SVD (triangles) methods.

is reasonable up to 4 MeV, then our data show a steeper yield drop for energy above 4 MeV.

The measured M_{γ} distribution for the fission prompt γ rays in ²³⁹Pu together with the unfolded ones are shown in Fig. 12. The mean multiplicity is 7.50 and 7.52 for the unfolded M_{γ} distributions derived from the iterative Bayesian and SVD methods, respectively. These are slightly higher than the 7.06(10) [1], the weighted average of the previous measurements. A simulation, similar to the one for ²³⁵U, was performed to check the goodness of this unfolded M_{γ} distribution and to obtain a rough estimate of the systematic uncertainty due to the neglect of the slight energy dependence of the γ -ray multiplicity response of DANCE. Similar results as those for ²³⁵U are obtained, that is, the systematic uncertainty on the order of ~0.3 is achieved for the unfolded M_{γ} distribution.

C. Prompt γ emission in the neutron-induced fission of ²⁴¹Pu

The experiment was fielded by using the ²⁴¹Pu target fabricated at LLNL with an isotopic enrichment of 69.51% and a total mass of ~ 0.220 mg, which results in a total mass of ~ 0.147 mg for ²⁴¹Pu in the target. Since it has a low isotope abundance, the potential background contribution to the measurement from other contaminations is discussed. The daughter nucleus ²⁴¹Am was removed chemically before the target was fabricated. The major contribution to the background of this measurement is from ²³⁹Pu (4.21%) and not from the adjacent isotopes ²⁴⁰Pu (16.12%), ²⁴²Pu (10.09%), or ²⁴¹Am since they are threshold fissioners. To minimize the contribution from ²³⁹Pu, the spectrum was derived with the incident neutron energy integrated from $\sim\!2~\text{eV}$ to $\sim\!100~\text{keV}$ with the exclusion of the ²³⁹Pu resonances. An upper bound of $\sim 2\%$ was estimated for the contribution from ²³⁹Pu, derived from the observed fission spectrum as a function of the incident neutron energy. A total of $\sim 1.35 \times 10^6$ valid fission events was collected for a period of ~ 17 days of beam on target. The measured spectrum for the fission prompt γ rays together with the unfolded ones is shown in Fig. 13. Again, the bumplike structures, similar to those of ²³⁵U and ²³⁹Pu, are observed in the E_{γ} spectrum. The mean energy for the γ -ray energy between 0.15 and 9.50 MeV is 0.97 and 0.86 MeV,



FIG. 13. (Color online) The measured E_{γ} spectrum for the neutron-induced fission of ²⁴¹Pu is shown by crosses together with the unfolded ones, derived from the iterative Bayesian (circles) and SVD (triangles) methods. The results from SVD are scaled up by a factor of 5 for clarity.

respectively, for the unfolded E_{γ} distributions derived from the iterative Bayesian and SVD methods. The mean energies are nearly identical to those of ²³⁹Pu. However, the detailed comparison of the E_{γ} spectrum between ²³⁹Pu and ²⁴¹Pu, shown in Fig. 14(a), indicates that the yield of the γ -ray energy near 7 MeV is enhanced by nearly a factor of 2 for ²⁴¹Pu. The possible cause for this enhancement is discussed in the next section.



FIG. 14. (Color online) The comparison of the unfolded (a) E_{γ} and (b) M_{γ} distributions between ²³⁹Pu (blue squares) and ²⁴¹Pu (green diamonds).



FIG. 15. (Color online) The measured M_{γ} distribution for the neutron-induced fission of ²⁴¹Pu (filled circles) together with the unfolded ones, derived from the iterative Bayesian (squares) and SVD (triangles) methods.

The measured multiplicity distribution for the fission prompt γ rays together with the unfolded ones are shown in Fig. 15. The mean multiplicity is 7.50 and 7.51 for the unfolded M_{γ} distributions derived from the iterative Bayesian and SVD methods, respectively, which are essentially identical to those of ²³⁹Pu, shown in Fig. 14(b), despite them being different by two neutrons. By following the same procedure outlined in two previous sections to check the goodness of this unfolded distribution and to obtain a rough estimate of the systematic uncertainty, an accuracy on the order of ~0.3, the same as that for ²³⁵U and ²³⁹Pu, is achieved.

D. Comparison of the prompt γ emission in fission of ²³⁵U, ^{239,241}Pu, and ²⁵²Cf

The successful measurements of the prompt γ -ray emission following the neutron-induced fission of ²³⁵U and ^{239,241}Pu as well as the spontaneous fission of ²⁵²Cf by using the DANCE array coupled with a PPAC gives one an opportunity to study not only the systematics of the prompt E_{γ} and M_{γ} distributions in fission, but also the correlations between E_{γ} and M_{γ} . The availability of both unfolded E_{γ} and M_{γ} distributions makes the latter possible.

Shown in Fig. 16 is the comparison of the unfolded E_{γ} spectra for ²³⁵U, ²³⁹Pu, and ²⁵²Cf where their relative yields are normalized to each other according to the intensity of the γ -ray energy between 1 and 4 MeV. The comparison between ²⁴¹Pu and ²³⁹Pu is given in Fig. 14(a). Despite many similarities among their E_{γ} spectra, one distinct feature is readily observed, that is, the spectrum above 5 MeV shows a strong dependence on the species of fissile nuclei. This dependence could result from the differences in the yield of fission fragments with mass near 130 where the intensity of γ rays with energy between 3.5 and 8.0 MeV is significantly enhanced compared to those of other mass regions [5,6,8]. The observed enhancement for the spontaneous fission of ²⁵²Cf can be reproduced [6] by lowering the level density for fission fragments with mass near 130 in the statistical model calculations using the CASCADE code.





FIG. 16. (Color online) The comparison of the unfolded E_{γ} distribution among ²³⁵U (red circles), ²³⁹Pu (blue squares), and ²⁵²Cf (black triangles).

It is also possible that this enhancement results from the γ decay of pygmy dipole resonances, which are populated in the fission fragments with mass near 130. Two observations support this explanation. One is that the pygmy resonances located at ~10 MeV just below the giant dipole resonances have been observed in the neutron-rich Sn-Sb region with mass near 130 by using the Coulomb excitation of high-energy radioactive beams at the LAND-FRS facility of GSI [26,27], which roughly is the sum of the γ -ray bumps at ~4 and ~6 MeV. The other is the observation of the γ decay of giant dipole resonances that follow the spontaneous fission of ²⁵²Cf [28,29]. Therefore, the population of pygmy resonances in fission fragments with mass near 130 is plausible.

The bumplike structure, observed at near 0.5 MeV in the spectrum, most likely results from the yrast or near-yrast γ transitions of fission fragments as suggested in Ref. [8]. The same explanation might be applicable for the bumplike structure observed at 1.5 MeV.

Shown in Fig. 17 is the comparison of the unfolded M_{γ} distributions for ²³⁵U, ²³⁹Pu, and ²⁵²Cf. The comparison between ²⁴¹Pu and ²³⁹Pu is given in Fig. 14(b). Their distributions have a very similar Gaussian-like shape with a tail that extends to the higher multiplicity. The mean multiplicity



FIG. 17. (Color online) The comparison of the unfolded M_{γ} distribution among ²³⁵U (red circles) and ²³⁹Pu (blue squares), and ²⁵²Cf (black triangles).

is 6.95, 7.50, 7.50, and 8.16 for ²³⁵U, ²³⁹Pu, ²⁴¹Pu, and ²⁵²Cf, respectively, whereas, their corresponding widths (FWHM) are 6.98, 7.39, 7.55, and 8.08. One obvious feature is that the mean multiplicity is increased with increased mass of fissile nuclei, whereas, the width shows the same trend and is nearly the same as the mean value. It is quite clear from this study that the M_{γ} distribution, instead of its mean value, should be adopted in the numerical simulation of any application that involves the fission prompt γ emission.



The study of correlations between E_{γ} and M_{γ} in fission was carried out for ²³⁵U, ²³⁹Pu, and ²⁴¹Pu by using the same procedure outlined in Ref. [7] by comparing the total γ -ray energy between the measurement and a simulation that uses the random sampling technique. The latter was performed by first randomly selecting a number according to the unfolded M_{γ} distribution then by selecting a matching number of γ rays randomly selected according to the E_{γ} distribution. This selected set of γ rays was folded according to the DANCE response, and the total γ -energy was derived. Some 10 × 10⁶ samples were selected for each of those simulations. The results together with the measurements for ²³⁵U are shown in Fig. 18. For comparison, an additional simulation was



FIG. 19. (Color online) The same as Fig. 18, except for ²³⁹Pu.

performed with the assumption of a δ function for the E_{γ} distribution that peaks at 1 MeV, which is about the average γ -ray energy.

The mean value for the total γ -ray energy, derived from the simulation as a function of multiplicity and shown in Fig. 18(a), is underestimated for the lower-multiplicity events but is overestimated for the higher-multiplicity events compared to the measured ones. This discrepancy may be due to the fact that the constraint is not placed on the total γ -ray energy in the selection of individual γ rays for a given sample. However, the two simulations give nearly the same result, which indicates that the sampling procedure was correctly carried out. The width derived from the simulation, shown in Fig. 18(b), has the



FIG. 20. (Color online) The same as Fig. 18, except for ²⁴¹Pu.

same trend as those of the mean value in comparison with the measured ones. The simulation with the fixed individual γ -ray energy does not reproduce the data numerically. To partially compensate the deficit in the sample selection of individual γ rays without any constraint on the total γ -ray energy, the spectrum taken with the ratio of the width to the mean value is shown in Fig. 18(c). The numerical agreement between the measurement and the simulation for events with multiplicity 7 and higher provides evidence for the stochastic aspect of prompt γ emission in fission. These results are consistent with those from our early study on the prompt γ emission in the spontaneous fission of ²⁵²Cf [7]. Similar results are obtained for ²³⁹Pu and ²⁴¹Pu, shown in Figs. 19 and 20, respectively. Although the gross features of the mean energy vs multiplicity distributions are reasonably well reproduced by the simple stochastic picture, the residual deviations between model and data may suggest that the γ -ray energy distribution may have a dependence on the γ -ray multiplicity. This possibility is investigated more in Refs. [30,31].

IV. SUMMARY

The prompt γ emission in the neutron-induced fission of ²³⁵U, ²³⁹Pu, and ²⁴¹Pu has been successfully measured by using the DANCE array in coincidence with the detection of fission fragments by three PPACs. The E_{γ} and M_{γ} distributions with the energy range between 0.15 and 9.50 MeV in fission are measured and subsequently are unfolded according to the DANCE response, simulated numerically according to a geometrical model validated with the γ -ray calibration sources. These measurements together with those for the spontaneous fission of ²⁵²Cf give us a unique opportunity to study the system of the prompt γ emission in fission.

The unfolded E_{γ} distribution shows many interesting features, such as the bumplike structures observed at the energy near 0.5, 1.5, 4.0 and 6.0 MeV with various enhancements. The most unique feature is that the intensity of the γ -ray energy above 5 MeV is sensitive to the species of fissile nuclei, which is most likely due to the difference in the yield of fission fragments with mass near 130. The population of pygmy resonances in this mass region is a possible explanation for this observation.

The actual M_{γ} distribution is derived for the neutroninduced fission of ²³⁵U and ^{239,241}Pu through the unfolding of the measured one. Together with the unfolded E_{γ} distribution, the correlations between E_{γ} and M_{γ} in fission can be explored. By comparing the total γ -ray energy between the measurement and a simulation that uses an assembly with elements selected randomly according to the unfolded E_{γ} and M_{γ} distributions, the evidence of the stochastic aspect of the γ emission in fission is illustrated. Furthermore, the mean multiplicity increases with increased mass of fissile nuclei. The width shows the same trend and is numerically nearly the same as its mean value. This observation has important implications for the numerical simulations in many applications that involve the fission prompt γ emission, that is, the M_{γ} distribution should be used instead of the average multiplicity.

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