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Evidence for the stochastic aspect of prompt γ emission in spontaneous fission

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The prompt γ-ray energy and multiplicity distributions in the spontaneous fission of ^{252}Cf have been measured using a highly segmented 4π γ-ray calorimeter. Corrections were made for both distributions according to the detector response, which is simulated numerically using a model validated with the γ-ray calibration sources. A comparison of the total γ-ray energy distribution was made between the measurement and a simulation performed by random sampling of the corrected γ-ray energy and multiplicity distributions and then transporting those γ rays through the response of the detector array. The agreement between the measurement and simulation for the mean energy and width is markedly improved at higher multiplicities compared to the lower ones, illustrating the diminishing correlation between the γ-ray and multiplicity energy and the stochastic aspect of the prompt γ emission in spontaneous fission at higher multiplicities.

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Understanding the fission process is important for the design of nuclear reactors and the safety of their operation. The prompt energy released in fission is derived from three components: fission fragments, their prompt neutrons, and γ emission. Significant amount of work has been done for the measurement and modeling of the mass and kinetic energy distributions of fission fragments [1] as well as the energy distribution and the average multiplicity of emitted neutrons [2,3]. However, there are only limited measurements and modeling attempts of the energy and multiplicity distribution for emitted γ rays [4].

In fission, the excited fission fragments deexcite by the neutron evaporation first until the excitation energy falls below the neutron separation energy then followed by the γ emission. The latter is the main path in producing the prompt γ rays in fission. Thus, the γ-ray spectrum can be very complex because of the number of nuclear species with various deformations involved and the broad distribution of excitation energies and angular momentum of their initial states populated after neutrons evaporated. With the advance of modern highly segmented 4π γ-ray detector arrays such as the Heidelberg-Darmstadt Crystal Ball [5] and the Detector for Advanced Neutron Capture Experiments (DANCE) [6] at the Los Alamos Neutron Science Center (LANSCE), precision measurements of the prompt γ-ray multiplicity (M_γ) and γ-ray energy (E_γ) distribution in fission become feasible. In fact, E_γ and M_γ together with the neutron multiplicity from ^{252}Cf spontaneous fission as a function of fission-fragment mass were measured using the Crystal Ball [7,8]. For the current work, our goal is to improve the understanding of the prompt γ-ray emission process in fission by evaluating the correlations between E_γ and M_γ. This requires not only the precision measurement of those quantities but also the possibility to correct for the detector response through an unfolding procedure. The unique features of DANCE, such as the nearly γ-ray energy independence of the multiplicity response, fulfill the requirement and offer the opportunity to achieve this goal.

Such a study has been done for the spontaneous fission of ^{252}Cf. The prompt γ rays have been measured by the DANCE array in coincidence with the detection of fission fragments by a parallel-plate avalanche counter (PPAC). The measured individual E_γ and M_γ distributions were corrected for the detector response ("unfolded") to produce the true distributions from fission. The unfolding was done using both the iterative Bayesian [9] and singular value decomposition [10] methods using a Monte Carlo detector response matrix as described later.

A ^{252}Cf source with a strength $\sim$0.15 μCi was prepared by stippling the material on a 3 μm thick titanium foil, and then covered by a 1.4 μm thick aluminized mylar to serve as a cathode. The two anodes, made of the same thickness aluminized mylar sheet, were placed on both sides of the cathode at a distance of 3 mm and electrically connected. The PPAC was operated with isobutane gas at the $\sim$4.00 torr pressure stabilized by a feedback loop of constant gas flow. It has efficiency of $\sim$82% for the detection of fission fragments with a bias of 375 V applied on the anodes and provides a fast and clean fission trigger for the DANCE array to record the coincident γ rays from fission fragments within a time window of $\sim$30 ns. Details of the design and fabrication of this PPAC are described in Ref. [11].

DANCE is a 4π γ-ray calorimeter made of 160 BaF_2 crystals; each crystal has equal solid-angle coverage. It was designed to study neutron-capture reactions on small quantities of radioactive and rare stable nuclei. For capture studies, neutron-capture events are recognized by the measured total γ-ray energy and the summed photopeak energy is equivalent to the reaction Q value plus the kinetic energy of the incident neutron. Besides its use as a calorimeter for the study of the neutron-capture reaction, DANCE can be used for the precision measurement of the E_γ and M_γ distributions as well as the total γ-ray energy (E_{tot}) distribution in fission as long as the measurement is accompanied by a charged-particle detector, such as the PPAC mentioned above, for detecting fission fragments.

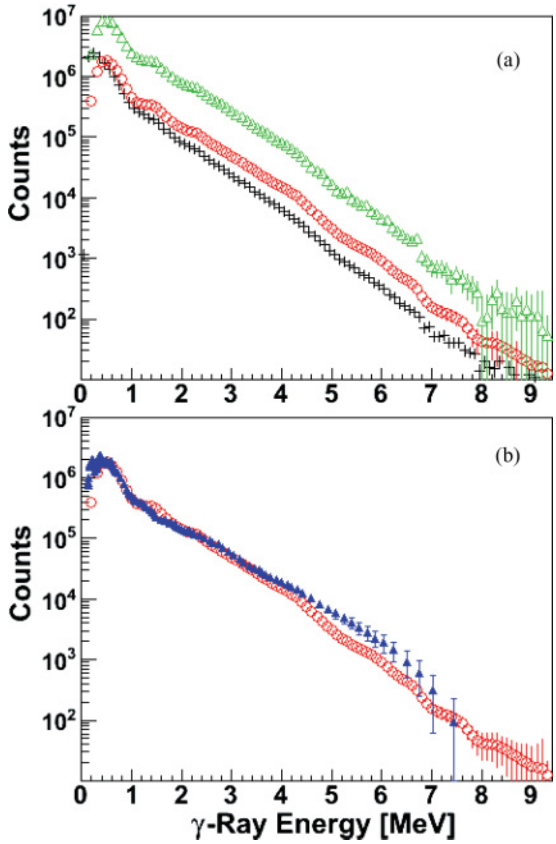

FIG. 1. (Color) Prompt γ-ray spectra of the ^{252}Cf spontaneous fission. (a) The measured one in black crosses and the unfolded ones in red circles and green triangles derived by using Bayesian and SVD methods, respectively. The distribution derived from the SVD method is scaled up by a factor of 5 for clarity. (b) Shown in blue triangles are Verbinski's previous results (Ref. [20]).

The experiment was fielded at LANSCE over a period of 3 d. A total of $\sim 3 \times 10^6$ fission events was selected for the current study by placing a time window of $\sim$10 ns on the coincident time spectrum between DANCE and the PPAC. A time resolution of $\sim$2 ns is achieved between the two detectors. This time window is set to restrict the contribution from prompt fission neutrons to less than 1%. The pulse height and timing information for both DANCE and the PPAC were derived from the recorded waveforms by 500 Megasample/s, 300 MHz bandwidth Acqiris digitizers. The coincident E_γ and M_γ distributions were determined from the DANCE array for the fission events. To reduce the summing effect on the E_γ distribution, the spectrum, plotted in Fig. 1(a), was obtained by requiring that the γ ray detected by the BaF_2 crystal register without any adjacent crystals being triggered. The sensitivity of DANCE for γ-ray spectroscopy studies is clearly demonstrated, where the strength of γ ray falling monotonically as a function of energy over a range of five decades until tapering off $\sim$8 MeV is observed.

The E_γ and M_γ responses for the DANCE array have been studied extensively [12–14]. A geometric model of DANCE together with the surrounding material was created to study the response numerically and the validation was carried out by the comparison between the measurement and simulation using GEANT4 [15] with three γ-ray calibration sources, ^{22}Na, ^{60}Co, and ^{88}Y. Many unique features of DANCE are quantified, such as the detection efficiency, the peak-to-total ratio, and the detected multiplicity distribution of a single γ ray are all nearly independent of E_γ ranging from 150 keV (detector threshold) to 10 MeV. For instance, the detection efficiency varies between 84 to 88 %, the peak-to-total is nearly constant $\sim$55%, and the average multiplicity varies no more than 16% [12,16]. Note that the values of E_γ and M_γ were derived by combining adjacent crystals if triggered. Introducing the geometric model of PPAC into the simulation reduces the efficiency by 4% and 1% for E_γ of 200 keV and 1 MeV, respectively. For the current work, however, the E_γ spectrum, plotted in Fig. 1(a), is derived from the requirement that no adjacent crystals are triggered.

As was mentioned earlier, the least known quantity for the prompt energy released in the fission process is the γ emission. The current work yields a precision measurement of the prompt E_γ and M_γ distributions for the spontaneous fission of ^{252}Cf and allows us to evaluate the correlations between E_γ and M_γ. However, before such a study can be carried out, the unfolding procedure is needed to correct those measured distributions according to the detector response.

The relationship between the measured distribution $\mathbf{M_{obs}}$ and the actual one $\mathbf{M_{phys}}$ can be expressed as a matrix equation $\mathbf{M_{obs}} = \mathbf{R} \cdot \mathbf{M_{phys}}$, where $\mathbf{R}$ is the response matrix for a given detector system. In principle, one can derive the actual distribution from the measured one by inverting the response matrix, but in reality, this technique fails to produce sensible solutions in most cases. An extensive discussion on this subject and the techniques developed to achieve better solutions can be found in Ref. [17]. In our current work, the unfolding of measured E_γ and M_γ distributions was carried out using both the iterative Bayesian and SVD methods implemented in the ROOT add-on software package [18]. The results derived from both methods are essentially identical. More on the unfolding of measured energy and multiplicity by DANCE can be found in Ref. [19].

The response matrices of DANCE for both E_γ and M_γ were simulated numerically using GEANT4 according to the model validated by the γ-ray calibration sources discussed earlier. For the energy response, the simulation is restricted by the same experimental condition, which requires the γ ray detected by the BaF_2 crystal without any adjacent crystals being triggered. This reduces the detection efficiency to 67–28 % for E_γ = 150 keV–10 MeV but the peak-to-total ratio remains the same $\sim$55%. Shown in Fig. 1(a) is the unfolded E_γ distribution together with the measured one. The average E_γ energies for both Bayesian and SVD unfolded spectra are 0.94 and 0.98 MeV, respectively, and the accuracy better than 5% is achieved. These values are slightly higher than 0.87(2), the weighted average of previous measurements quoted in Ref. [4]. The average γ-ray energy in the current work is overestimated since the strength of the γ-ray energy below 200 keV is poorly determined due to the limited energy resolution of $\sim$14% together with the 150-keV energy threshold for the DANCE array. Shown in Fig. 1(b) is the comparison with the previous measurement by Verbinski *et al*. [20] and the agreement is

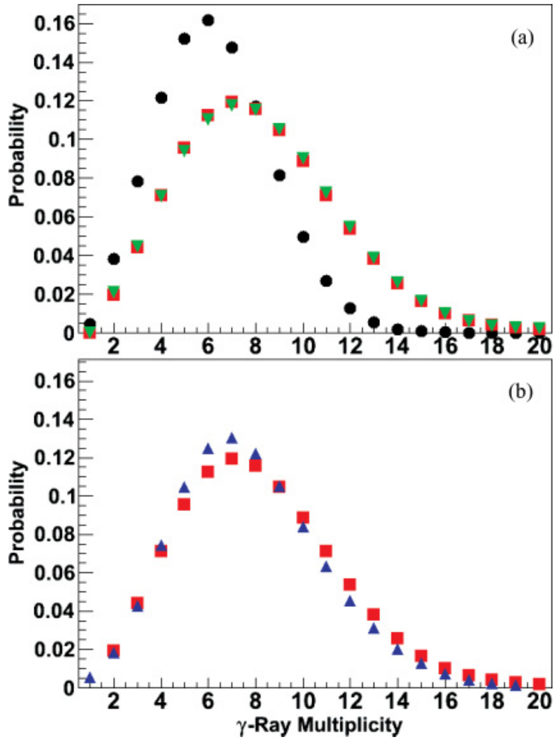

FIG. 2. (Color) Prompt M_γ distribution for the ^{252}Cf spontaneous fission. (a) The measured one shown in black circles and the unfolded ones shown in red square and green triangles derived by using Bayesian and SVD methods, respectively. (b) Shown in blue triangles are the previous results from Ref. [21].

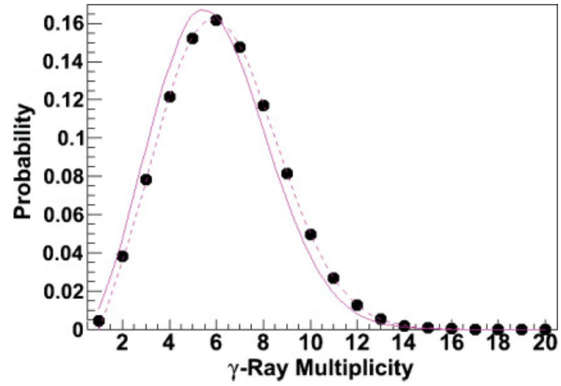

FIG. 3. (Color) The comparison of the γ-ray multiplicity distribution between the measured (black circles) and two simulated ones including the DANCE response in both solid and dashed lines in magenta. See the text for details.

reasonable in general until ∼4 MeV where the current data drops steeper toward higher γ-ray energy. This discrepancy cannot be attributed to the enhancement of the γ-ray energy in the 4–8 MeV region observed by Hotzel *et al.* [8] in their gated spectrum with fragment masses centering at 132, since the yield of this mass region is a very small fraction of the total mass distribution.

For the multiplicity response, the matrix was created by the simulations weighted by the unfolded E_γ distribution to minimize the contribution from the residual energy dependence of the multiplicity response. Shown in Fig. 2(a) is the unfolded γ-ray multiplicity distribution together with the measured one. The average γ-ray multiplicities are 8.16 and 8.14 for the unfolded distributions by Bayesian and SVD methods, respectively, and a consistency better than 1% is achieved between these two methods. These values are consistent with 7.98(40), the weighted average of previous measurements quoted in Ref. [4]. Shown in Fig. 2(b) is the comparison with the semiempirical formulated distribution by Brunson [21], where the average multiplicity is 7.79. The agreement is good in general even though the current data shifts toward higher γ-ray multiplicity. Note that this is the first time that the actual γ-ray multiplicity in spontaneous fission is derived experimentally.

To evaluate the correlations between E_γ and M_γ for the prompt γ emission from spontaneous fission, a comparison of the $E_{\rm tot}$ distribution was made between the measurement and a Monte Carlo simulation. The simulated $E_{\rm tot}$ spectrum was made by, first, random sampling the unfolded multiplicity spectrum to obtain a sample number of γ rays per event N_γ. Next, the unfolded individual E_γ spectrum, Fig. 1, was sampled N_γ times. The resulting event of N_γ γ rays was processed through the GEANT4 simulation of the DANCE detector. Ten million events were sampled. Shown in Fig. 3 is a comparison of the M_γ between the measurement and simulation. The simulated M_γ, plotted as the solid line, has the average value of 5.91 versus measured one of 6.29. This 0.38 discrepancy is introduced by the uncertainty in the unfolded γ-ray multiplicity distribution, modulated through the DANCE response in the simulation. A significant improvement in reproducing the multiplicity distribution, plotted as the dashed line in Fig. 3, can be achieved by correcting the unfolded multiplicity distribution by an amount of 0.4 unit to a higher value in the simulation. Therefore, we believe an accuracy of 0.4 out of the average value of 8.14 is reached for the unfolded γ-ray multiplicity.

Shown in Fig. 4(a) and (b) are the comparisons of the mean value and the FWHM, respectively, for $E_{\rm tot}$ between the measurement and simulation. The improvement in the agreement is evident for both the mean value and the FWHM for events with multiplicity 7 or higher. For example, the differences in the mean energy drop from ∼30% to ∼8% from the lower to higher multiplicity, while the deviations in the FWHM decrease from ∼40% to ∼6%. The agreement is not accidental since the simulation with the single γ-ray energy at 0.94 MeV for all multiplicities underestimates FWHM by ∼30% or more despite the mean energy being well reproduced.

For events with the multiplicity 11 and higher, the simulation overestimates both the mean energy and width compared to the measurement. This may not be a surprising result since there is no restriction placed on the total γ-ray energy when the individual γ rays are chosen randomly according to the unfolded energy distribution. However, their width-to-mean ratios, shown in Fig. 4(c), are in excellent agreement between the measurement and simulation. This indicates that the importance of correlations between E_γ and M_γ diminishes for the higher multiplicity events. One plausible

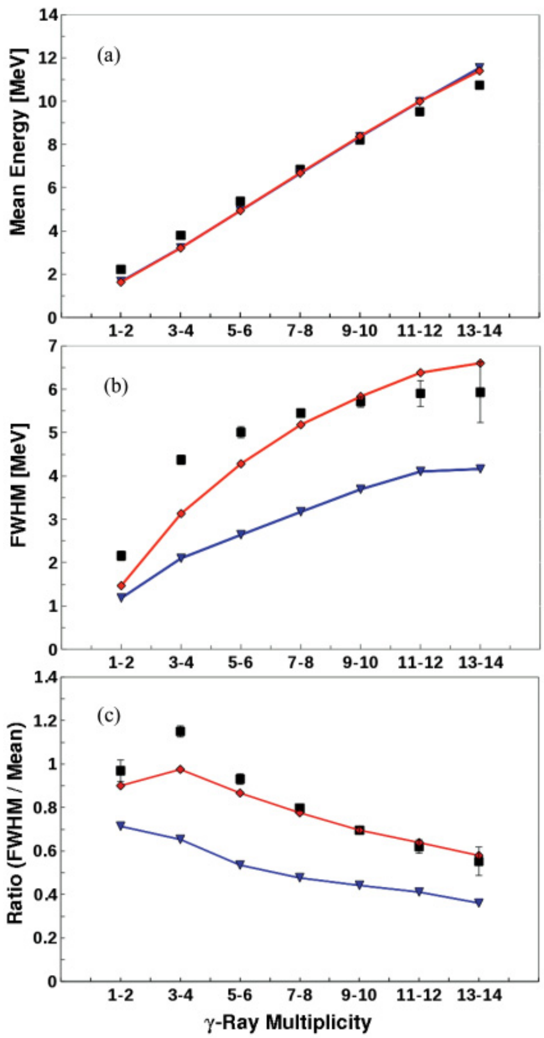

FIG. 4. (Color) The comparison of the mean (a) and FWHM (b) of E_{tot} between the measured (in black) and simulated (in red) including the DANCE response spectra. The error bars are not visible if they are smaller than the marker size. The simulation with the fixed E_γ = 0.94 MeV (in blue) is nearly identical to the red curve in (a). The ratio FWHM-to-mean is plotted in (c).

explanation for this observation is that such events are most likely originated from the initial states with higher excitation energy and angular momentum. Under this circumstance, the higher level density together with a significant level mixing results in an ensemble of γ rays that can be treated statistically, which manifests the stochastic aspect of the γ emission in spontaneous fission of ^{252}Cf. Obviously, their correlations cannot be ignored and the detailed nuclear structure of fission fragments has to be considered to fully account for these discrepancies. Nevertheless, this is an important finding to advance the modeling of the prompt γ emission in fission.

The prompt E_γ and M_γ distributions in spontaneous fission of ^{252}Cf were measured using the DANCE array. Both distributions were unfolded using the iterative Bayesian and SVD methods and consistent results were obtained. A comparison of the E_{tot} energy distributions was made between the measurement and simulation by random sampling of both unfolded distributions and going through the detector response. For events with higher multiplicity, a rapid improvement in the agreement between the measurement and simulation is observed, which implies diminished correlations between the E_γ and M_γ and manifests the stochastic aspect of the prompt γ emission in the fission process. This is important for developing a predictive model of fission. Evidently, other aspects, such as details of nuclear structure in fission fragments, need to be considered in order to fully explain the prompt γ emission in spontaneous fission. We plan to extend current study to the neutron-induced fission in order to further improve our understanding of the prompt γ emission process in fission.

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