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Dependence of the prompt fission γ-ray spectrum on the entrance channel of compound nucleus: Spontaneous vs. neutron-induced fission

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ABSTRACT

Prompt γ-ray spectra were measured for the spontaneous fission of ^{240,242}Pu and the neutron-induced fission of ^{239,241}Pu with incident neutron energies ranging from thermal to about 100 keV. Measurements were made using the Detector for Advanced Neutron Capture Experiments (DANCE) array in coincidence with the detection of fission fragments using a parallel-plate avalanche counter. The unfolded prompt fission γ-ray energy spectra can be reproduced reasonably well by Monte Carlo Hauser–Feshbach statistical model for the neutron-induced fission channel but not for the spontaneous fission channel. However, this entrance-channel dependence of the prompt fission γ-ray emission can be described qualitatively by the model due to the very different fission-fragment mass distributions and a lower average fragment spin for spontaneous fission. The description of measurements and the discussion of results under the framework of a Monte Carlo Hauser–Feshbach statistical approach are presented.

The prompt energy released in the nuclear fission is dominated by the kinetic energy of the fission fragments and then followed by the prompt neutron and γ-ray emission from the fully accelerated fission fragments. In the past, most model and experimental efforts were devoted to the kinematic energy of fission fragments and the neutron emission. Little attention was paid to the γ-ray emission until recently. A single γ-ray detector was used for most measurements made in 1970's and their results were summarized in Ref. [1]. Recent years have seen an increased interest in the prompt γ-ray emission in fission [2–14] because the data are important for fission modeling and applications in nuclear industries. For example, new prompt fission γ-ray data at thermal neutron energy and above for ^{235}U and ^{239}Pu, required for the precise modeling of γ-ray heating in reactor cores, were categorized as high-priority by the Nuclear Energy Agency under the Organization for Economic Cooperation and Development [15].

The majority of measurements for the prompt γ-ray emission in fission were made using one or a few γ-ray detectors for the neutron-induced fission of U and Pu isotopes as well as ^{252}Cf(sf) and ^{240,242}Pu(sf). More recently, a new class of fast scintillators, such as cerium-doped-LaBr$_3$, CeBr$_3$, and LaBr$_3$ detectors, was used by Billnert et al. [4], Oberstedt et al. [7,10,12,13], and Gatera et al. [14]. Lately, a new generation of measurements has emerged for the prompt γ-ray emission in fission that uses highly segmented 4π γ-ray calorimeters, such as the Heidelberg–Darmstadt Crystal Ball [16] and the Detector for Advanced Neutron Capture Experiments (DANCE) array [17,18].

Measurements of the prompt fission γ-ray emission for the cases mentioned above were made for either the neutron-induced fission at a given incident neutron energy or the spontaneous fission. No report was made for the impact of compound nucleus entrance channel on prompt fission γ-ray emission except for a recent study of ^{240,242}Pu*, where the spontaneous fission was measured for γ-ray energy up to 4 MeV [13]. The comparison with thermal neutron-induced fission ^{241}Pu(n_{th}, f) indicates no or lit-

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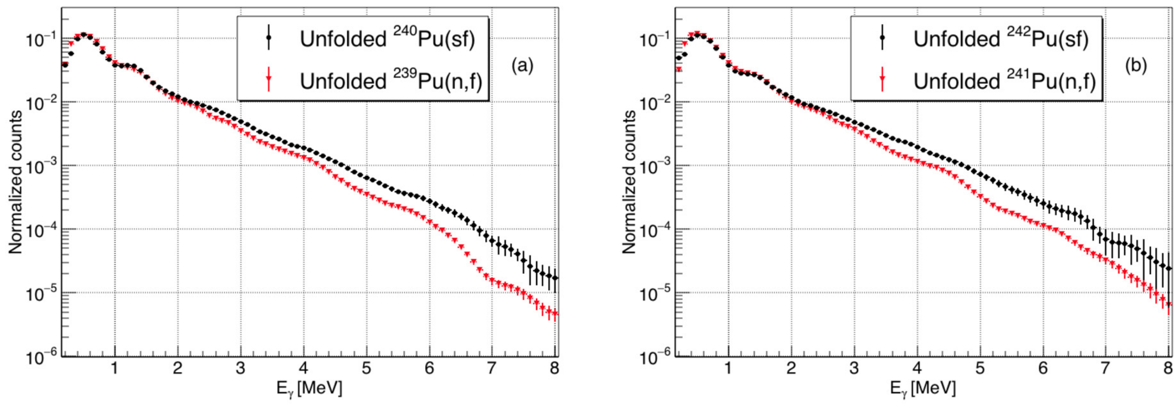

Fig. 1. Comparison of unfolded E_γ spectrum between ^{240}Pu(sf) (black) and ^{239}Pu(n, f) (red) is shown in (a) and between ^{242}Pu(sf) (black) and ^{241}Pu(n, f) (red) shown in (b). All spectra are self-normalized to one.

tle dependence on the entrance channel of the ^{242}Pu* compound nucleus. Furthermore, there are known cases where the prompt fission γ-ray spectra were measured using fast neutrons with energy up to 20 MeV for ^{235}U(n, f) and ^{238}U(n, f) [19,23] and no obvious dependence on the incident neutron energy was found. However, the measurement given in Refs. [20,21] showed that the prompt γ-ray spectrum for neutron-induced fission of ^{238}U after the third-chance fission is different from those with lower incident neutron energy that can be described adequately by model calculations [22].

In this work, we present a new study of the dependence of prompt fission γ-ray emission on the entrance channels of ^{240,242}Pu* compound nuclei. There are two distinct entrance channels for their fission. One is spontaneous fission and has an entrance channel of zero intrinsic excitation energy and spin of 0^+. The second channel is neutron-induced fission of ^{239,241}Pu with the incident neutron energy from thermal to 100 keV. They have the entrance channel of ≈ 6.3 MeV intrinsic excitation energy with spin of 0 or 1 for ^{239}Pu(n, f) and ≈ 6.5 MeV intrinsic excitation energy with spin of 2 or 3 for ^{241}Pu(n, f). The prompt fission γ-ray emission for both fission channels of both compound nuclei was measured using the DANCE array in coincidence with the detection of fission fragments by a compact parallel-plate avalanche counter (PPAC) [24], designed specifically for DANCE. The description of experiments and data analysis will be presented in the sections below. These experimental results on the dependence of prompt fission γ-ray spectrum on entrance channels of ^{240,242}Pu* compound nuclei is analyzed using a Monte Carlo implementation of the Hauser–Feshbach statistical theory, the CGMF code [8], with a focus on the primary mass yields and the fragment spin distribution.

Measurements of the prompt γ-ray spectrum of ^{239,241}Pu(n, f) and of ^{240,242}Pu(sf) were performed at the Lujan Neutron Scattering Center at LANL/LANSCE. For neutron-induced fission experiments, PPACs with either ^{239}Pu or ^{241}Pu targets were assembled at LLNL and bombarded by neutrons with energies from thermal up to several hundred keV. Neutrons were produced first by bombarding a tungsten target with an 800 MeV proton beam at a repetition rate of 20 Hz 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 PPACs. A total of over 10^6 fission events with at least one γ ray detected by DANCE were collected for both isotopes. These results were published earlier [5,9]. For the spontaneous fission, PPACs with a total mass of about 642 μg of ^{242}Pu enriched to 99.93% or about 769 μg of ^{240}Pu enriched to 98.86% were assembled at LLNL and used for the fission-fragment detection in coincidence with the detection of the prompt γ rays by DANCE. A total of $\approx 10^5$ fission events with at least one γ ray detected by DANCE were collected for both targets. The same ^{242}Pu PPAC was used for the absolute (n, γ) cross section measurement [25].

In the offline analysis using the code FARE [26], a valid fission event required a coincidence between a fission fragment detected by the PPAC and a γ ray detected by DANCE with an 8–10 ns time window on their time difference spectrum. A time resolution better than 2 ns was achieved for all fission reactions studied. Three physical quantities were inferred from the coincident γ rays detected by DANCE: (1) the total prompt fission γ-ray energy $E_{\gamma,\mathrm{tot}}$ spectrum defined as the sum of energy of all detected γ rays; (2) the total prompt fission γ-ray multiplicity M_γ determined according to the number of clusters, grouped adjacent detectors triggered; (note that this counting method for M_γ avoids double counting due to the Compton scattering, is largely independent of the γ-ray energies E_γ, and is closer to the simulated results using γ-ray calibration sources [3,27,28]) (3) the prompt fission γ-ray energy E_γ spectrum determined by excluding any γ ray with adjacent crystals triggered to avoid the summing effect. Details of this analysis have been described in our earlier publications [3,5,9]. In this letter, we focus on the discussion of the prompt fission γ-ray energy E_γ spectrum dependence on the entrance channel of the compound nuclei.

Corrections must be made to the measured E_γ spectra to obtain the true physical ones that can be compared to model calculations. This can be accomplished by unfolding the measured spectra using the detector response matrices. For unfolding one-dimensional spectra, the iterative Bayesian [29–31] and the singular-value decomposition (SVD) [32] methods are available to correct the E_γ spectrum. The detector response matrices are simulated using the GEANT4 [33] geometrical model including both DANCE and PPAC [3,5,9,34]. To make sure the simulated detector response matrices have sufficient coverage of the phase space beyond the measured one, we use the E_γ spectrum in the range 0.1–12 MeV for the response matrix in the unfolding of one-dimensional spectra.

The unfolded E_γ spectra obtained by using the iterative Bayesian method, for ^{240}Pu(sf) as well as ^{239}Pu(n, f) are shown in Fig. 1(a), and the spectra for ^{242}Pu(sf) and ^{241}Pu(n, f) are shown in Fig. 1(b). A very similar trend is observed for fission of both compound nuclei; that is the E_γ spectrum for the spontaneous fission is harder than that of the neutron-induced fission for γ-ray energies above 2 MeV. The difference in yield is nearly a factor of 2 for γ-ray energy near 6 MeV. In general, the systematic uncertainty is about 10% for the unfolding with simulated detector responses, which is an order of magnitude smaller than the observed difference in yield and has no impact on the conclusion.

We have used the CGMF code [8] to model the de-excitation of fission fragments through a Monte Carlo implementation of the statistical Hauser–Feshbach theory [35]. Both the prompt γ-ray observables as well as prompt neutron observables are calculated. These include the average prompt neutron multiplicity $\bar{\nu}$, its dependence on fragment mass $\bar{\nu}(A)$, and the distribution $P(\nu)$, as well as the average prompt γ-ray multiplicity $\overline{M}_\gamma$ and prompt fission γ-ray energy E_γ spectrum (PFGS). The prompt neutron observables for ^{239,241}Pu(n, f) and ^{240,242}Pu(sf) are used to constrain the CGMF calculations.

To conduct a CGMF calculation, one requires the initial yields of the pre-neutron emission fission fragments in mass A, charge Z, and total kinetic energy TKE: $Y(A, Z, \mathrm{TKE})$. Mass yields $Y(A)$ are taken from experimental data, the charge yields follow the Wahl systematics [36], and the average TKE dependence on fragment mass $\langle\mathrm{TKE}\rangle(A)$ is taken from experimental data. We allow the overall magnitude of $\langle\mathrm{TKE}\rangle(A)$ to vary by a parameter η in order to scale the total $\langle\mathrm{TKE}\rangle$ for the entire fission reaction

$$\langle\mathrm{TKE}\rangle = \eta \sum_{A_H} Y(A_H)\cdot\langle\mathrm{TKE}\rangle(A_H), \qquad (1)$$

where the sum is conducted only over the heavy fragment masses A_H. The spin J distribution of the fission fragments follows a Gaussian form [8,11]

$$P(J|A, Z) \propto (2J+1)\exp\left[\frac{-J(J+1)\hbar^2}{2\alpha T\mathcal{I}_0(A, Z)}\right], \qquad (2)$$

where we use a spin-scaling factor α to shift the average spin $\langle J\rangle$ of the fragments, the nuclear temperature T is determined from the excitation energy and level density parameter, and the moment of inertia $\mathcal{I}_0(A, Z)$ is for a rigid-rotor of the ground-state shape for a fragment with mass A and charge Z. A larger α, and thus a larger $\langle J\rangle$ via Eq. 2, increases the competition between γ-ray and neutron emission above the yrast line, which increases the γ-ray multiplicity while slightly decreasing the prompt neutron multiplicity. Increasing α also decreases the average γ-ray energy $\langle E_\gamma\rangle$ as described in Ref. [11]. Equal probability is assumed for positive and negative parities in the level densities.

For the results presented here, the $Y(A)$ come from experimental data for ^{239}Pu(n_{th}, f) [37], ^{240}Pu(sf) [37], ^{241}Pu(n_{th}, f) [38], ^{242}Pu(sf) [38]. The $\langle\mathrm{TKE}\rangle(A)$ come from experimental data for ^{239}Pu(n_{th}, f) [39], ^{240}Pu(sf) [37], ^{241}Pu(n_{th}, f) [40], ^{242}Pu(sf) [38], again utilizing similar experimental setups for each compound nucleus. We have verified that using other experimental data sources for $Y(A)$ and $\langle\mathrm{TKE}\rangle(A)$ has a negligible effect on our results. The total excitation energy between a sampled pair of fragments is determined via energy conservation $\mathrm{TXE} = Q - \mathrm{TKE}$, where Q is the energy available from fission into the pair of fragments. The TXE is shared between the fragments via a ratio of temperatures $R_T(A)$, which is determined by a fit to the $\bar{\nu}(A)$ data [41] for ^{239}Pu(n_{th}, f) or the predictions from Wahl's systematics [36] otherwise. This procedure has been described in previous works [42] and has produced reasonable agreement with a variety of experimental data for many fissioning systems [8,11,42]. Previous studies have shown that the detector threshold E_{thresh} and timing window Δt can have large effects on the prompt γ-ray multiplicity [43], so we have matched the DANCE detector with $E_{\mathrm{thresh}} = 150$ keV and $\Delta t = 10$ ns for our CGMF calculations.

First, we begin with the toy example, Fig. 2, to demonstrate the impact of specific fission properties, such as the excitation energy E^*, mass yields $Y(A)$, average total kinetic energy $\langle\mathrm{TKE}\rangle$,

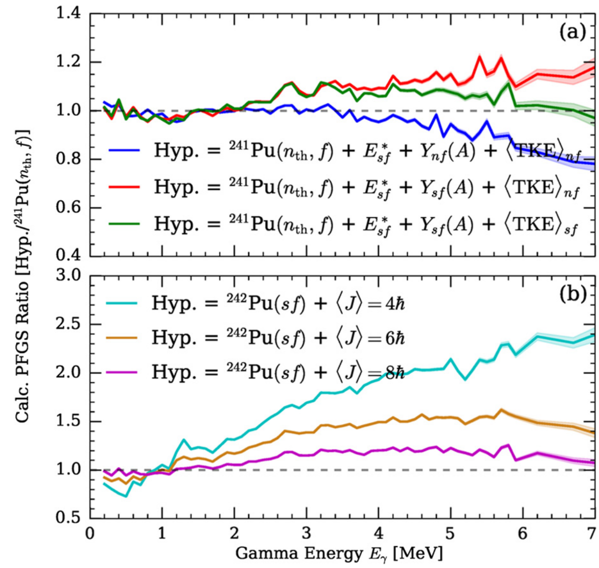

Fig. 2. The ratio of the calculated prompt fission γ-ray spectra (PFGS) for a hypothetical fission reaction (Hyp.) over our best-fit calculation of ^{241}Pu(n_{th}, f). The hypothetical reaction begins identical to ^{241}Pu(n_{th}, f), but we progressively change the excitation energy E^*, mass yields $Y(A)$, and average fragment total kinetic energy $\langle\mathrm{TKE}\rangle$ to reflect spontaneous fission (sf) of ^{242}Pu instead of thermal-neutron induced fission (n_{th}, f) of ^{241}Pu (a). In (b), the hypothetical reaction has all of the fission properties of ^{242}Pu(sf) and we vary the average fragment spin $\langle J\rangle$. When $\langle J\rangle$ is varied, we also vary $\langle\mathrm{TKE}\rangle$ in order to maintain $\bar{\nu} = 2.149$ neutrons/fission [44].

and average spin $\langle J\rangle$, on the resulting PFGS. We start with two identical CGMF calculations of the ^{241}Pu(n_{th}, f) PFGS, the nominal result seen in Fig. 3(c). We progressively change the E^*, $Y(A)$, and $\langle\mathrm{TKE}\rangle$ from those of ^{241}Pu(n_{th}, f) to those of ^{242}Pu(sf) and note the changes in the PFGS relative to the original ^{241}Pu(n_{th}, f) PFGS in Fig. 2(a). Lowering the excitation energy from $E^* \approx 6.5$ MeV, in the case of ^{241}Pu(n_{th}, f), to $E^* = 0$ for spontaneous fission (sf) produces a slightly softer PFGS for the new hypothetical fission reaction (blue curve). This is because the lower excitation energy results in less average energy for the prompt γ rays, even though the majority of the excitation energy is emitted as prompt neutrons [8]. In addition, the change in E^* affects the neutron emission resulting in a slight change of the intensities for some low-energy discrete γ-ray peaks. The red curve illustrates one of the main conclusions of this work; when we account for the dramatically different mass yields $Y(A)$ between the (n_{th}, f) and (sf) reactions, the PFGS of the spontaneous fission reaction is actually *harder* than that of thermal-neutron induced fission. This is because the $Y(A)$ for ^{240,242}Pu(sf) are peaked near the $N = 82$ neutron shell closure. Coincidentally, the average γ-ray energy also peaks in this $130 \le A_H \le 138$ mass region [2,45], which leads to the observed hardening. Finally, the green curve in Fig. 2(a) increases the $\langle\mathrm{TKE}\rangle$ to reflect ^{242}Pu(sf). This effect slightly softens the PFGS of the hypothetical reaction because the increase in $\langle\mathrm{TKE}\rangle$ leads to a decrease in $\langle\mathrm{TXE}\rangle$.

In Fig. 2(b), we use all of the fission properties of ^{242}Pu(sf) and adjust the average spin of the fission fragments $\langle J\rangle$. Changing $\langle J\rangle$ will slightly change $\bar{\nu}$, so we adjust η to keep $\bar{\nu} = 2.149$ neutrons/fission [44]. This results in $\langle\mathrm{TKE}\rangle = 181.5, 181.0, 180.5$ MeV via Eq. 1 for the $\langle J\rangle = 4, 6, 8\ \hbar$ calculations, respectively. The absolute values of the $\langle\mathrm{TKE}\rangle$ can have large systematic errors, on the order of 1–2 MeV [46], but some experimental groups have measured $\langle\mathrm{TKE}\rangle$ for both fission reactions in ^{240}Pu* [37,47] and ^{242}Pu* [48] with similar detector setups. This provides us with some degree of confidence on the $\langle\mathrm{TKE}\rangle$ difference $\Delta_{\mathrm{TKE}} = \langle\mathrm{TKE}\rangle_{(sf)} - \langle\mathrm{TKE}\rangle_{(n_{\mathrm{th}}, f)}$. The $\langle\mathrm{TKE}\rangle$ values used in Fig. 2(b) result

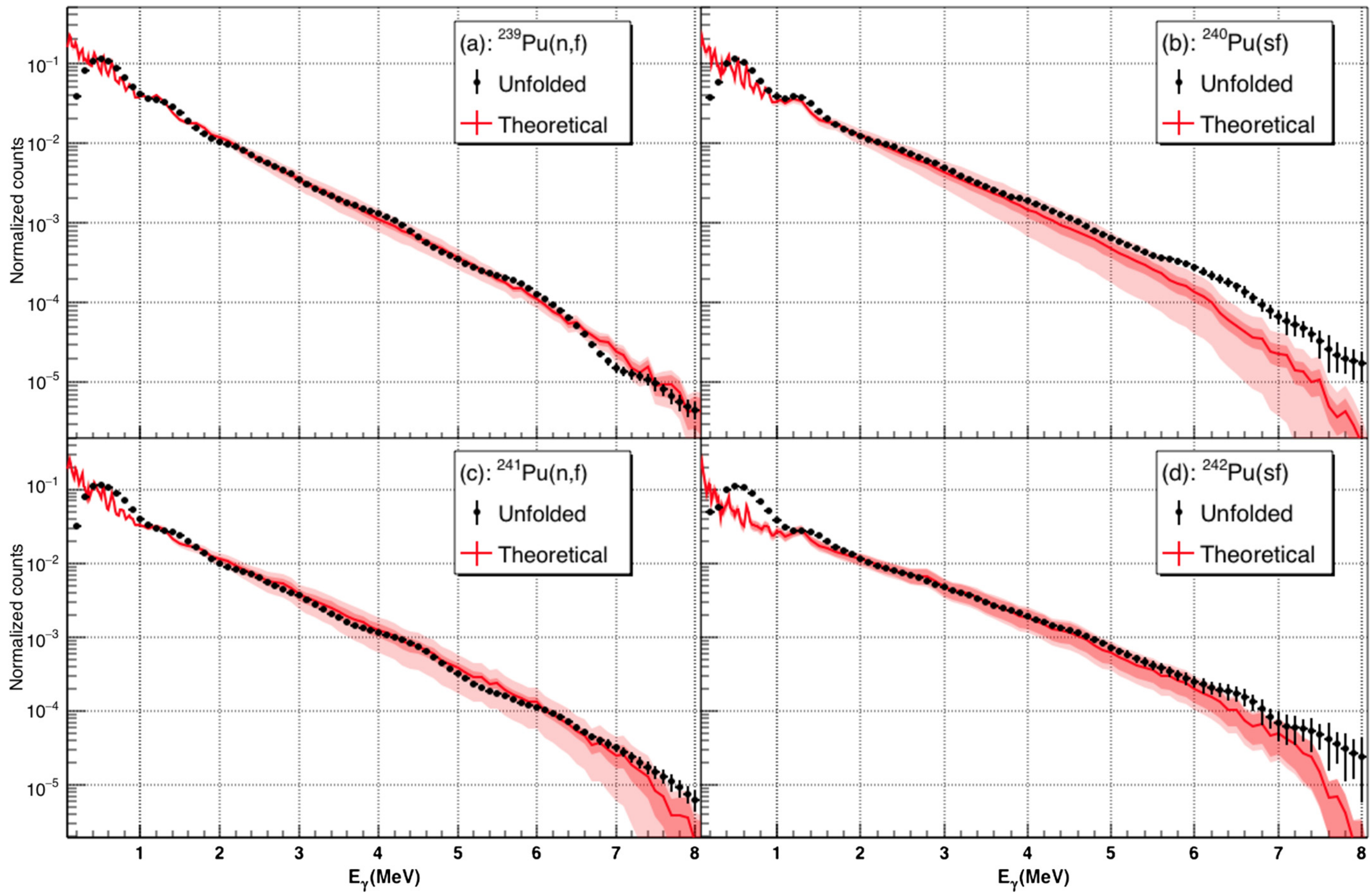

Fig. 3. Comparison of the unfolded (points) and calculated (lines and bands) prompt fission γ-ray spectrum (PFGS) for ^{239}Pu(n, f) (a), ^{240}Pu(sf) (b), ^{241}Pu(n, f) (c), and ^{242}Pu(sf) (d). The calculated central values (lines) use the nominal total kinetic energy of the fragments $\langle \text{TKE} \rangle_e$ given in the text and the dark (light) bands are the ± 0.5 MeV (± 1.0 MeV) uncertainties. Unfolded spectra are self-normalized to one. To account for a lack of experimental sensitivity below 1 MeV, calculated data were normalized to experimental data in the $1 \le E_\gamma \le 5$ MeV range.

in $\Delta_{\text{TKE}} = 2.5, 2.0, 1.5$ MeV for the $\langle J \rangle = 4, 6, 8\ \hbar$ calculations. For comparison, Ref. [48] reports $\Delta_{\text{TKE}} = 2.69$ MeV. Overall, Fig. 2 shows that the lower E^* and larger $\langle \text{TKE} \rangle$ for spontaneous fission softens the PFGS relative to thermal-neutron induced fission, but the spontaneous fission $Y(A)$ and lower $\langle J \rangle$ act oppositely and can dominate, depending on the spin. A similar result occurs for the ^{240}Pu* compound system.

For our best-fit CGMF calculations, we have utilized the nominal experimental values of $\langle \text{TKE} \rangle_e = 177.80,\ 178.85,\ 179.00,\ 181.50$ MeV, which are within the 1σ experimental uncertainties for ^{239}Pu(n_{th}, f) [39], ^{240}Pu(sf) [49], ^{241}Pu(n_{th}, f) [48], and ^{242}Pu(sf) [48]. The α values are then varied until $\bar{\nu} = 2.881, 2.157, 2.925, 2.149$ neutrons/fission in agreement with the experimental values for ^{239}Pu(n_{th}, f) [50], ^{242}Pu(sf) [44], ^{241}Pu(n_{th}, f) [44], and ^{242}Pu(sf) [44]. The variance of the $\langle \text{TKE} \rangle (A)$ is scaled in order to achieve reasonable agreement with the $P(\nu)$ from various experiments [44,50] or evaluations [51].

Plotted in Fig. 3 are the comparisons between the unfolded results from DANCE and the CGMF calculations. The darker and lighter bands indicate the calculated spectra when we vary $\langle \text{TKE} \rangle_e$ by ± 0.5 MeV and ± 1.0 MeV. Some of the fine structure in the calculations are not reproduced in the unfolded data as the energy resolution of DANCE is limited. The structure seen for $E_\gamma < 1$ MeV is due to discrete transitions, where peak locations are in good agreement with low-energy experimental data [10,13,14,52]. We can see that the measured ^{239}Pu(n, f) PFGS is reproduced nicely by the calculations, even up to $E_\gamma \sim 7$ MeV. The ^{240}Pu(sf) PFGS is reasonably well reproduced, but the slope is too steep. The ^{241}Pu(n, f) calculation is slightly harder than the measured result, indicating that a lower $\langle \text{TKE} \rangle$ than the used $\langle \text{TKE} \rangle_e$ could produce a better fit and a higher $\overline{M}_\gamma$ as well, in agreement with Ref. [9,10]. The ^{242}Pu(sf) PFGS can reproduce the unfolded data reasonably well, but the large $\langle \text{TKE} \rangle_e$ we have used generates a very small $\overline{M}_\gamma \sim 4.2\ \gamma$/fission, far below the values in Ref. [1,13]. For both ^{240}Pu* and ^{242}Pu*, neutron-induced fission required a larger $\langle J \rangle$ to achieve good agreement with $\bar{\nu}$. Overall, the neutron-induced fission reactions are in better agreement than spontaneous fission.

In summary, the prompt γ-ray spectra of ^{240,242}Pu(sf) and ^{239,241}Pu(n, f) with the incident neutron energy range from thermal to 100 keV were measured using the DANCE array in coincidence with the detection of fission fragments using a PPAC. This offers an opportunity to study the dependence of prompt fission γ-ray emission on the entrance channel for the formation of the compound nucleus. It was carried out by comparing the unfolded experimental spectra and the ones calculated using the CGMF code, a Monte Carlo Hauser–Feshbach statistical model. The experimental results with the DANCE detector observed a relative hardening in both the ^{240}Pu* and ^{242}Pu* compound systems. The observed differences in the E_γ spectrum between the spontaneous and neutron-induced fission were qualitatively confirmed by the model calculations and interpreted as due to the difference in the fission-fragment mass distributions and fragment spin distributions. The mass distributions for spontaneous fission peak near $A \sim 133$ and has a narrower variance, where the average γ-ray energies are known to increase. A portion of the observed hardening of the E_γ

spectrum relative to the neutron-induced reaction for the ^{242}Pu* and ^{240}Pu* compound system can be attributed to this change in mass distributions. A decrease in $\langle J \rangle$ for the spontaneous fission reactions could account for a majority of the observed differences in the prompt γ-ray spectra.

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