Assessing the role of the $(n, \gamma f)$ process in the low-energy fission of actinides

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Abstract. We review the role of the $(n, \gamma f)$ process in the low-energy neutron-induced fission reaction of ^{239}Pu. Recent measurements of the average total γ-ray energy released in this reaction were performed with the Detector for Advanced Neutron Capture Experiments (DANCE) at Los Alamos. Significant fluctuations of this quantity in the resonance region below 100 eV can be interpreted by invoking the presence of the indirect $(n, \gamma f)$ process. Modern calculations of the probability for such an event to occur are presented.

1 Introduction

The possibility for a heavy compound nucleus formed through low-energy neutron-induced reactions to emit a γ ray before fissioning was postulated theoretically long ago by Stavinsky and Shaker [1] and Lynn [2]. In this scenario, a compound nucleus is first formed with excitation energy $E^* = \epsilon_n + B_n$, angular momentum $J = I \pm \frac{1}{2}$, and parity π. ϵ_n is the kinetic energy of the incident neutron, B_n the neutron binding energy for the target, and I the spin of the target in its ground-state. Since we are considering low-energy reactions, only s-wave ($l = 0$) neutrons are taken into account. In the case of n+^{239}Pu reaction, and considering the target nucleus in its ground-state with $I^\pi = \frac{1}{2}^+$, only 0^+ and 1^+ compound states can be populated. At those energies, only the neutron elastic, fission and capture channels are open. Because the excitation energy E^* is higher than the fission barrier height by $\sim$1.5 MeV, γ rays can be emitted leaving the nucleus with enough residual excitation energy to overcome the fission barrier, leading to an additional $(n, \gamma f)$ channel.

Beyond the rather esoteric interest for this particular fission decay, there are practical implications to the existence of this process. The emitted γ ray carries with it an excitation energy of about 1 MeV, which then represents a deficit for the total excitation energy available for the emission of prompt neutrons. Because the average neutron multiplicity $\overline{\nu}$ plays such an important role in nuclear technologies, as for instance in the very accurate simulation of critical assembly and/or nuclear reactor benchmarks, a small change in $\overline{\nu}$ has important practical consequences, as noted in [3, 4].

Such fluctuations of $\overline{\nu}$ have been observed already, as shown in Fig. 1 for incident energies up to 100 eV in the neutron-induced fission of ^{239}Pu. To a good approximation, the neutron multiplicity reflects how much excitation energy is available in the fission fragments shortly after scission. In turn, this energy depends on the total kinetic energy of the complementary fragments and on the Q value of

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the fission reaction. The latter is a function of the specific fragments (A, Z) produced in the reaction. Besides the $(n, \gamma f)$ process, fluctuations in the particular fragment yields in mass and charge could also lead to fluctuations in the number of emitted neutrons [5]. However, such an interpretation would not explain fluctuations in the observed total γ-ray energy, as will be explained below.

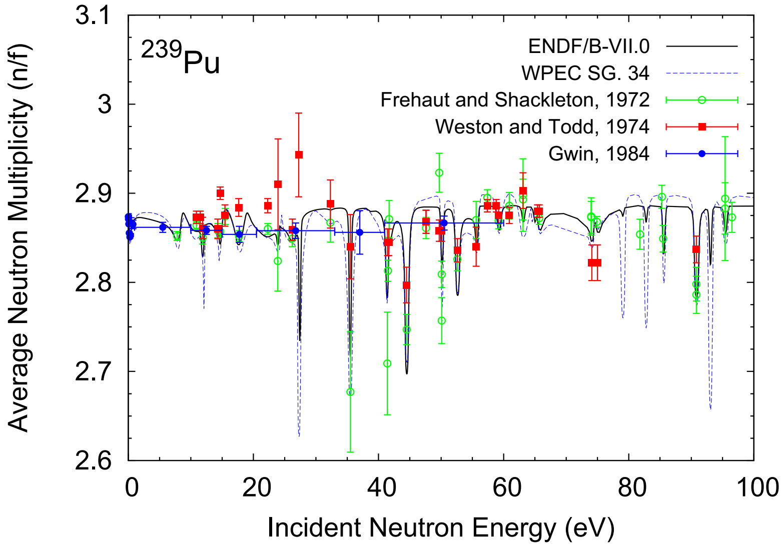

Figure 1. Fluctuations in the average prompt fission neutron multiplicity $\overline{\nu}_n$ for the ^{239}Pu (n, f) reaction.

2 Theoretical Framework

Here, we briefly review the theoretical framework used to compute the pre-fission γ-ray emission probability $\Gamma_{\gamma f}$ and spectrum. This probability can be written as follows

$$\Gamma_{\gamma f}(E^*, J^\pi) = \sum_{Xl} \sum_{J'=|J-l|}^{J+l} \int_0^{E^*} d\epsilon_\gamma \rho(E^* - \epsilon_\gamma, J', (-)^{Xl}\pi) \Gamma_{\gamma,Xl}(\epsilon_\gamma) P_f(E^* - \epsilon_\gamma, J', (-)^{Xl}\pi), \quad (1)$$

where $\rho(E, J, \pi)$ is the level density in energy, spin and parity; $\Gamma_{\gamma,Xl}$ is the γ-ray strength function, and $P_f(E, J, \pi)$ is the fission probability for a given energy, spin and parity. The level density is calculated at the residual excitation energy after γ-ray emission, $E^* - \epsilon_\gamma$. The residual spin and parity depend on the particular electromagnetic transition– denoted by Xl, leading to the final configuration. We calculate the compound nucleus level density following the combinatorial model described in [6]. Single-particle states are combined with vibrational and rotational enhancements to infer the total level density. The neutron and proton pairing gap parameters are adjusted to reproduce the known average resonance spacing for ^{240}Pu.

Only $E1$ and $M1$ electromagnetic transitions are considered in this work, with higher-order transitions assumed to be negligible. The γ-ray strength functions are obtained using the giant dipole resonance formalism. The Kopecky-Uhl generalized Lorentzian expression is used to compute $E1$ transitions, while a standard Lorentzian is used for $M1$ transitions.

Finally, the fission probabilities entering in Eq. 1 are calculated in the cross-section modeling formalism discussed in [6, 7]. This model is especially suited to the study of fission reactions near or below the fission barrier, as coupling between the two wells along the fission path lead to significant corrections to the simpler statistical model [6]. Since ^{239}Pu is a fissile nucleus, its fission barrier height lies below its neutron separation energy, and the direct estimation of its barrier height and width using neutron-induced reactions is not possible. Instead, surrogate reactions such as ^{238}Pu (t, pf) transfer-induced fission reactions [8] were used to infer the needed input parameters for the fission channel.

More details on the model and each of these quantities will be provided in a longer publication, in preparation.

3 DANCE Experiment and Simulations

The Detector for Advanced Neutron Capture Experiments (DANCE) at the Los Alamos Neutron Science Center (LANSCE) is a 4π-calorimeter that consists of 160 BaF_2 crystals, each 734 cm^3 in volume with faces located 19 cm from the beam center. Such an array has a very high efficiency and is very well suited to measure the total energy and multiplicity released by prompt γ rays in a fission event. Located on the Lujan flight path, incident neutrons are produced by spallation on a tungsten target and moderated to energies ranging from thermal up to a few hundred keV. Of particular interest to the present study is the region below 100 eV where well-resolved neutron resonances appear. Those resonances are also well characterized in energy and spin, providing an ideal test ground for our calculations.

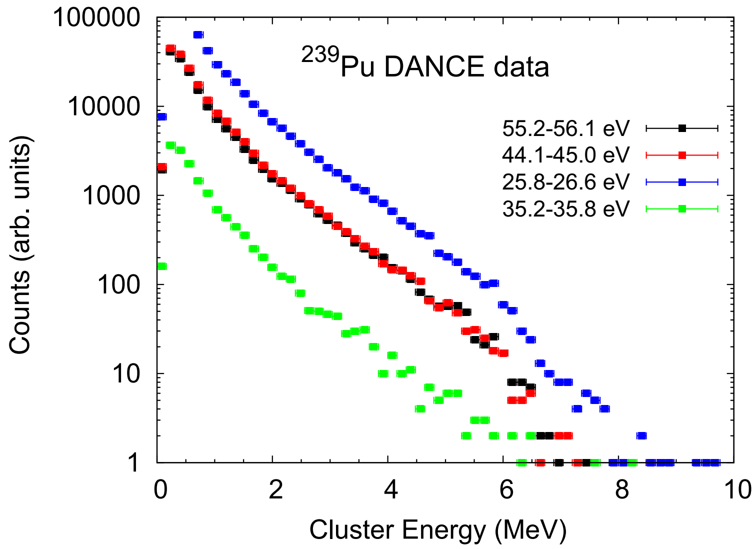

Figure 2. Total γ-ray energy measured with the DANCE detector for different energy bins.

Measurements of the total γ-ray energy and total γ-ray multiplicity for different neutron resonances are shown in Figs. 2 and 3, respectively. Due to the complexity of the DANCE detector, the unfolding of the raw signal into a useful and physical γ-ray spectrum is no easy task. Instead, we chose to forward propagate simulated γ rays into a transport simulation of the DANCE detector

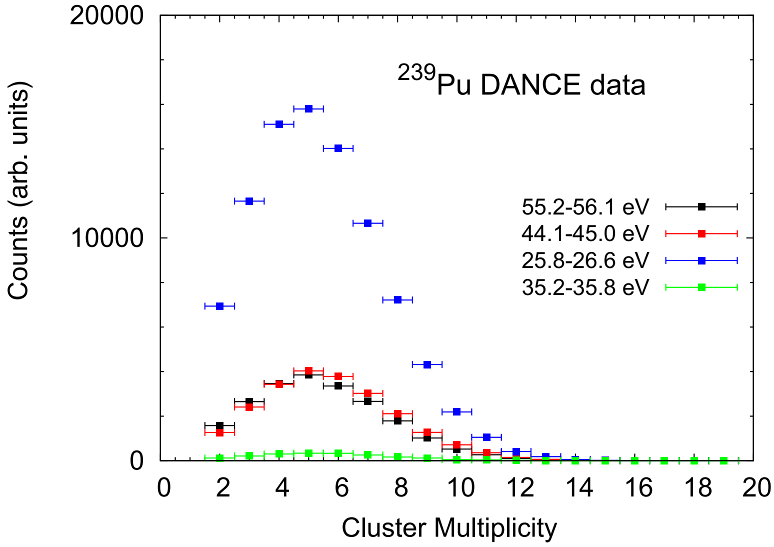

Figure 3. Total γ-ray multiplicity measured for the same energy bins as in Fig. 2.

response. The GEANT4 transport code was used, leveraging an already built model of the complete DANCE array. As mentioned above, we are not able to differentiate between γ rays emitted right before fission from those emitted right after, so we have to calculate the contribution from the fission fragments as well. The prompt fission γ rays evaporated from the fission fragments are calculated using our CGMF code [9], which computes the de-excitation of the primary fragments through neutron and γ emissions in a Monte Carlo implementation of the Hauser-Feshbach statistical nuclear reaction theory. The calculated prompt fission γ-ray spectrum, excluding the contribution from the $(n, \gamma f)$ events, for the thermal neutron-induced fission of ^{239}Pu is shown in Fig. 4. The agreement between the CGMF calculations and the measurement by Chyzh [10] and by Verbinski [11] is quite good.

The total γ-ray spectrum is obtained by combining the CGMF-calculated spectrum of γ rays evaporated from the fragments with the γ-ray spectrum from the $(n, \gamma f)$ process, weighted by the ratio $\Gamma_{\gamma f}/\Gamma_f$

$$N_\gamma^{tot}(\epsilon_\gamma) = \frac{\Gamma_{\gamma f}}{\Gamma_f} N_{\gamma f}(\epsilon_\gamma) + N_{FF}(\epsilon_\gamma), \tag{2}$$

where Γ_f is the sum of the width for the “direct" fission process and $\Gamma_{\gamma f}$. To obtain this equation, we are making the reasonable assumption that the spectrum of prompt γ rays emitted from the fragments is mostly insensitive to a modest change in the initial excitation energy of the fragments.

We then perform random samplings of the total spectrum N_γ^{tot} to produce random fission events that are used as input in the GEANT4 transport simulations. The final propagated results can then be compared to raw experimental data from DANCE.

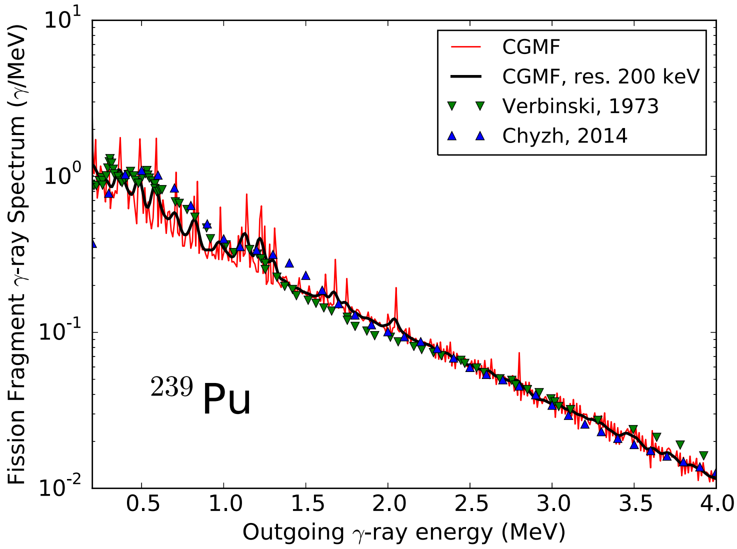

Figure 4. Prompt fission γ-ray spectrum calculated with the CGMF code for the thermal neutron-induced fission of ^{239}Pu. Experimental data sets from Chyzh *et al.* [10] and Verbinski *et al.* [11] are plotted for comparison.

4 Results and Discussion

Using the ^{238}Pu (t, pf) transfer-induced fission reactions [8], the characteristics in height and width of the double-hump fission barrier for ^{240}Pu* were obtained and are in good agreement with the values given in [7]. Of course, those values are somewhat model-dependent and not directly observable.

The fission probabilities calculated for ^{240}Pu* are shown in Fig. 5 for several spin and parity states. At low excitation energy, the spectrum of an even-even nucleus such as ^{240}Pu is dominated by the K=0 ground-state band (0^+, 2^+, ...) and at slightly higher energy, the mass asymmetry band (1^-, 3^-, ...). With s-wave neutrons ($l = 0$) impinging on ^{239}Pu in its ground-state ($1/2^+$), only 0^+ and 1^+ compound nuclear states can be formed. Only $M1$ transitions can then lead to 0^+ and 2^+ fission transition states, hence explaining the stronger than expected contribution of $M1$ relative to $E1$ transitions in the $(n, \gamma f)$ process.

In Eq. 1, it is the convolution of the fission probabilities P_f with the widths $\Gamma_{\gamma,Xl}$ that determines the magnitude of the $(n, \gamma f)$ process. Contrary to what is commonly observed in capture reactions, the $M1$ component is strongly enhanced in this case, due to the higher values of the fission probabilities for the 0^+ and 2^+ states, which again, can only be accessed through $M1$ transitions.

As noted earlier, disentangling γ rays emitted in the $(n, \gamma f)$ process from all prompt γ rays emitted in coincidence with a fission event is not possible. Due to the large number of compound nucleus states available, the width $\Gamma_{\gamma f}$ can be assumed to be rather constant across resonances. Due to the large fluctuations of the total fission widths Γ_f, the impact of the $(n, \gamma f)$ process can only be revealed for resonances with very small total fission width. In the n+^{239}Pu reaction, only a few resonances satisfy this criterion. Below 100 eV, of particular interest are the 1^+ resonances at energies 27.24, 35.50, 41.42, 44.48, 50.08, 52.60, 90.75, and 92.97 eV. The most promising candidate is the 41.42

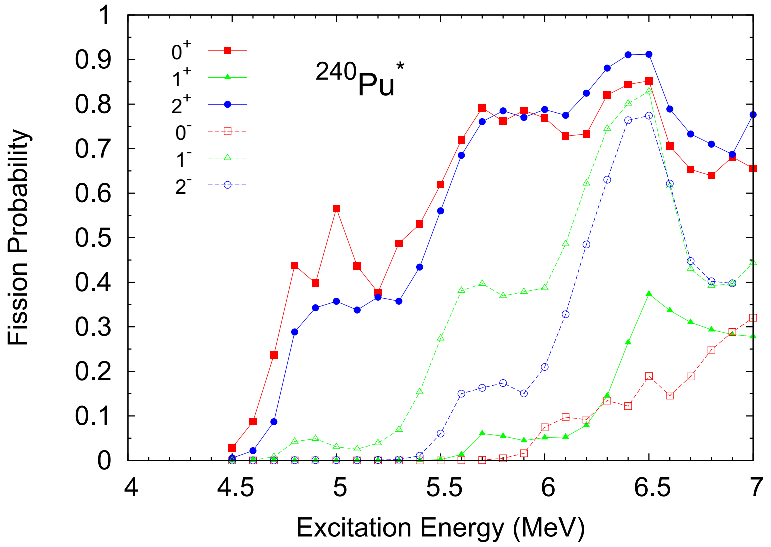

Figure 5. Spin and parity-dependent fission probabilities calculated for ^{240}Pu*. Positive-parity states are depicted with full symbols and solid lines, while negative-parity states are shown with empty symbols and dashed lines.

eV resonance with a total fission width of only 6.4 meV [12]. All 0^+ states have much larger fission widths.

One way to reveal the contribution of the pre-fission γ rays to the total fission γ-ray spectrum is to study the ratio of γ-ray spectra obtained for two different resonances, with two very different total fission widths. As an illustrative example, we shown in Fig. 6 the results obtained for the 35.5 eV resonance (Γ_f=3.5 meV) vs. the 14.6 eV resonance (Γ_f=30 meV). The observed (black) and simulated (red) γ-ray spectra clearly show a “bump" in the 1-1.5 MeV range, where the contribution from the $(n, \gamma f)$ process is expected to be strongest. The agreement between the simulated and experimental data is quite good in this ratio plot. Note that the addition of a $M1$ “scissors" mode contribution worsens the agreement by predicting too many pre-fission γ rays. However, other parameterizations of this process could lead to a different conclusion. This work is in progress.

Finally, to address the question of the role of the fluctuations of the mass and the total kinetic energy yields of the fission fragments in the resonance region as observed by Hambsch *et al.* [5], we performed `CGMF` calculations of the decay of the fission fragments by modifying artificially the average total kinetic energy of the fragments and study the sensitivity of the calculated neutron and γ-ray multiplicities on TKE. The result is shown in Fig. 7. As can be seen, the average neutron multiplicity (red) is extremely sensitive to TKE. On the other hand, the average γ-ray multiplicity is almost constant over a wide range, ±2 MeV, of TKE values. Such a result confirms that only the $(n, \gamma f)$ mechanism can explain the type of fluctuations observed with DANCE and in previous experiments. This is not to say that the yield fluctuations in mass and TKE do not play a role in the fluctuation of $\overline{\nu}$ observed in the resonance region (see Fig. 1). The exact contributions of the two phenomena for each resonance would be difficult to assess though.

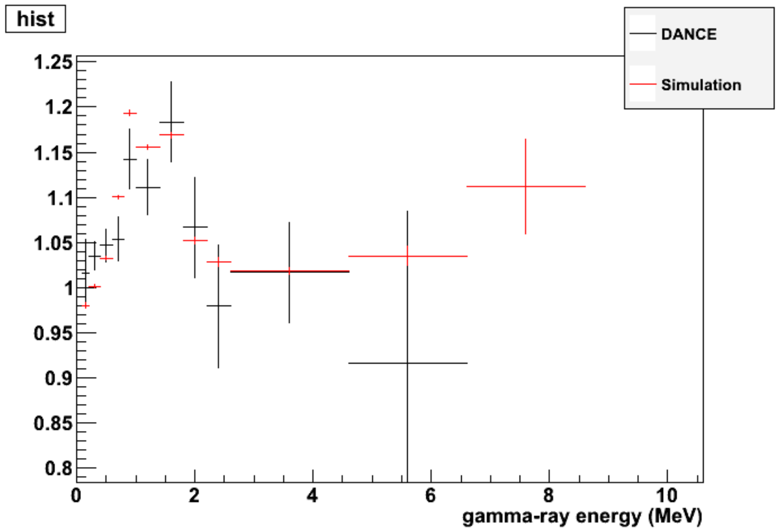

Figure 6. Ratio of γ-ray energy spectrum for two resonances, 35.5 eV (Γ_f=5 meV) and 14.6 eV (Γ_f=33 meV), as observed (black) and simulated (red) in the DANCE detector. A clear "bump" appears in the 1-1.5 MeV range, corresponding to the predicted average γ-ray energy emitted in the $(n,\gamma f)$ process.

5 Conclusion

In this contribution, we reviewed an old problem with fresh eyes, fresh model calculations, and fresh experimental data. While the possibility for the emission of a γ ray prior to fission was postulated theoretically 50 years ago, rather conclusive experimental evidence for such a process came only in the seventies. Very little information on the pre-fission γ-ray spectrum is available however, even though it is important to infer the particular nature of the γ transitions preceding fission. We have applied a modern theoretical framework to compute this spectrum, along with the spectrum of γ rays from the fission fragment themselves. Propagating these results into a GEANT4 model of the DANCE detector array, we were able to directly compare the calculated results with recent DANCE experimental data. Because of the intrinsic difficulties in disentangling pre- and post-fission γ rays, we studied the ratios of various spectra for different resonances. The contribution of the $(n,\gamma f)$ process is best observed for resonances with very small total fission widths. In the case of ^{239}Pu (n,f), we were able to show and explain the important role of $M1$ transitions in the pre-fission γ-ray spectrum. More work is in progress to complete calculations and experimental data analysis for neutron-induced fission reactions on both ^{239}Pu and ^{235}U.

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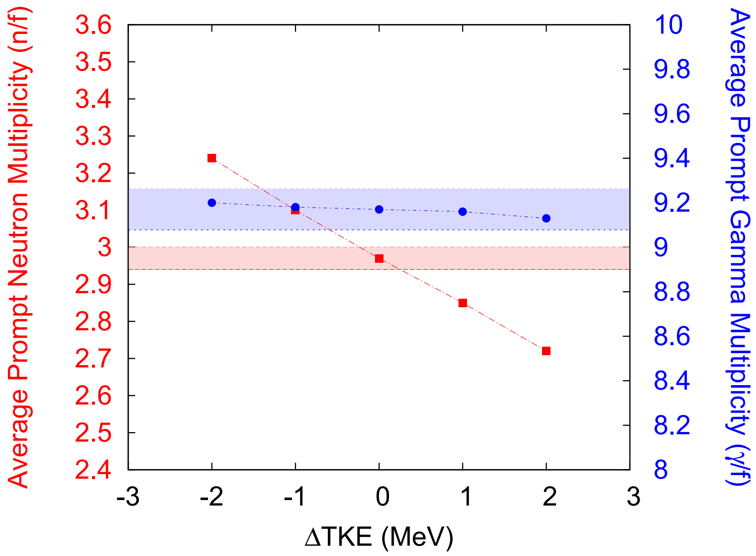

Figure 7. Sensitivity of the calculated average prompt neutron and γ-ray multiplicities on a change in the total kinetic energy TKE of the fission fragments.

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