

Comparing fission-product yields from photon-induced fission of ^{240}Pu and neutron-induced fission of ^{239}Pu as a test of the Bohr hypothesis in nuclear fission

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Abstract. Fission product yields (FPYs) are a uniquely sensitive probe of the fission process, with well established dependence on the species of nucleus undergoing fission, its excitation energy and spin. Thus FPYs are well suited for testing Bohr's hypothesis in the context of nuclear fission, which states that the decay of a compound nucleus with a given excitation energy, spin and parity is independent of its formation. Using FPYs, we have performed a new high-precision test of the combined effects of the entrance channel, spin and parity on the fission process from two of the most commonly used particles to induce fission - neutrons and photons. The $^{239}\text{Pu}(n,f)$ reaction at $E_n = 4.6$ MeV and the $^{240}\text{Pu}(\gamma,f)$ reaction at $E_\gamma = 11.2$ MeV were used to produce a $^{240}\text{Pu}^*$ compound nucleus with the same excitation energy. The FPYs from these two reactions were measured using quasimonoenergetic neutron beams from the TUNL's FN tandem Van de Graaff accelerator and quasimonoenergetic photon beams from the High Intensity γ -ray Source (HI γ S) facility. The FPYs from these two reactions are compared quantitatively for the first time.

1 Introduction

Since the discovery of nuclear fission in 1938, there has been extensive efforts to develop a theoretical description of the fission mechanism. The compound reaction-channel formalism of Bohr and Wheeler, first proposed in 1939 [1] is remarkably successful at describing the fission process. An essential assumption of this theory is that the nucleus proceeds through the fission process in a well defined quantum state. Thus the state of the nucleus influences the fission process, while the formation mechanism of the compound nucleus does not. Consequently all fission theory relies on the "Bohr hypothesis", which states that the decay of a compound nucleus for a given excitation energy, spin, and parity is independent of its formation [2]. The Bohr hypothesis is a particularly reasonable assumption for nuclear fission, where the timescale for the formation of the compound nucleus ($\sim 10^{-16}$ seconds) is long compared to the time needed to reach an equilibrium. However, despite the Bohr hypothesis

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underlying nearly every modern theoretical fission model, it has yet to be tested experimentally to a high degree of precision. Currently, the Bohr hypothesis is verified to about 20% accuracy in fission using nuclear reactions such as (t,pf), (^3He ,df), and (^3He ,tf) [3–7]. In this work we propose to test Bohr’s hypothesis in the context of nuclear fission by comparing the fission product yields (FPYs) from a compound $^{240}\text{Pu}^*$ nucleus produced via two different nuclear reactions: $^{239}\text{Pu}(n,f)$ and $^{240}\text{Pu}(\gamma,f)$.

2 Experiment

In this work we performed the first-ever experiment to identify the practical consequences of the Bohr hypothesis in the context of induced nuclear fission. We measured the FPYs from the $^{240}\text{Pu}(\gamma,f)$ reaction at the HI γ S facility at Triangle Universities Nuclear Laboratory (TUNL) [8] and compared the results to the FPYs obtained from the $^{239}\text{Pu}(n,f)$ reaction that our group recently measured at the TUNL Tandem Van de Graaff accelerator [9–12]. Both reactions produced a ^{240}Pu compound nucleus with the same excitation energy since the energies of the incident neutron and photon beams were purposely chosen to account for the 6.5 MeV difference in binding energy between ^{239}Pu and ^{240}Pu . The only difference was the spin-parity distributions from $n + ^{239}\text{Pu}$ and $J^\pi = 1^-$ from $\gamma + ^{240}\text{Pu}$, which is widely believed to be inconsequential at high excitation energies.

2.1 Dual-fission chambers

One of the most significant components in measurements of fission product yields is determining the number of fission reactions that occurred in the target. In the present work the fission reactions are counted using a Dual-fission ionization Chamber (DFC) [9], based on the design by Grundl *et al.* [13] for FPY measurements in reactors and critical assemblies [14].

The irradiation for the $^{239}\text{Pu}(n,f)$ measurements in Gooden *et al.* [11] were performed with the thick ^{239}Pu activation target mounted in the center of the DFC, as is the standard procedure used by our group [9–11, 15]. The two FCs then measure the effective neutron flux just before and after the target, and the effects of the beam divergence can be accounted for with Monte Carlo calculations [9]. The previously published data for $^{239}\text{Pu}(n,f)$ at $E_n = 4.6$ MeV were only sensitive to FPYs with half lives of \sim days or longer, so the data were augmented with a new recent measurement using the same experimental procedure and target but with only 2 hours of irradiation, in order to measure the yields of FPYs with half lives of a few minutes to a few hours.

In the case of the ^{240}Pu photofission measurements, the thick ^{240}Pu activation targets were positioned just outside of the DFC, with the long-activation target upstream and the short-activation target downstream with respect to the γ -ray beam. Unlike the neutron beams at TUNL, the HI γ S γ -ray beam is highly parallel with negligible divergence over a few cm distance. Thus it is not necessary to account for the spread of the γ -ray beam between the FC and the targets. This assumption was confirmed by comparing the relative beam flux measured in each chamber in the DFC. When a DFC is mounted in the neutron beam at TUNL, the beam divergence causes the upstream FC to typically see $\sim 25\%$ greater flux than the downstream FC, after accounting for the masses of the reference foils in each chamber. In the HI γ S γ -ray beam, the ^{240}Pu DFC upstream to downstream FC ratio of 1.02 ± 0.01 was reasonably consistent with negligible γ -ray beam divergence. In addition, the self-absorption and scattering of the 11.2 MeV γ -ray beam from the thick foils was less than 1%, as calculated by MCNP simulations.

2.2 Activation targets & reference foils

The ^{240}Pu targets used in this measurement both consist of $^{240}\text{PuO}_2$ powder which is compressed into a 1.27 cm diameter Al cylinder, the same diameter as the ^{239}Pu target that was used in Ref. [11]. The ^{240}Pu mass of each target was 73.01 ± 0.04 mg and 89.11 ± 0.05 mg, with both targets enriched to 99.873% ^{240}Pu . The Al holders have a mass of ~ 19 mg, with a 1.91 cm outer diameter and a 0.191 cm thickness. The Al material is relatively transparent to the γ -rays emitted by the β -decay of fission products and also is not activated by exposure to the 11.2 MeV γ -ray beam.

The reference foils for the DFC consist of ^{240}Pu electroplated onto a 1.27 cm diameter area on a Ti backing with ^{240}Pu masses of 25.97 ± 0.18 μg and 23.60 ± 0.12 μg and an isotopic enrichment of 99.873%. The spatial uniformity of the deposition was verified by scanning the surface of the foils with a 1 mm aperture α -detector. Like the ^{239}Pu reference foils, the ^{240}Pu deposits were sufficiently thin that fission fragments could easily escape the electroplated material and deposit their energy in the gas of the ionization chambers with nearly 100% efficiency.

2.3 Monoenergetic photon beams from the HI γ S facility

The HI γ S facility produces intense, quasimonoenergetic γ -ray beams via intra-cavity Compton backscattering of free electron laser photons and relativistic electrons [8]. Photofission measurements require intense γ -ray beams such as the ones provided by the HI γ S facility since the (γ, f) reaction is typically a factor of ~ 5 – 10 smaller than the (n, f) reaction for a given target nucleus.

For this work, the HI γ S facility was operated using 540 nm free electron laser photons to produce linearly-polarized, 11.2 MeV γ -ray beams with a resolution of approximately 360 keV (FWHM) and an average flux on target of 1.95×10^7 γ -rays/(cm^2 s). The γ -ray beam was collimated with a 1.27 cm diameter, 15.24 cm long Pb collimator placed ~ 2.5 m upstream of the target assembly. The energy spectrum of the γ -ray beam was measured with a 123% high-purity germanium (HPGe) detector positioned in the beam axis, while copper attenuators were inserted into the γ -ray beam far upstream of the collimator to reduce the flux.

One ^{240}Pu target (#1) was irradiated with $E_\gamma = 11.2$ MeV γ -ray beam for 2 hours to measure the short-lived FPYs (minutes to hours), the other (#2) for 92.6 hours to build up activity of long-lived FPYs (days). The 2 hour irradiation induced a total of $8.25 \pm 0.15 \times 10^6$ fissions in the short-lived FPY target, while the 92.6 hour irradiation induced a total of $4.38 \pm 0.07 \times 10^8$ fission events.

2.4 Detection of γ -rays from the β -decay of fission products

After irradiation by the HI γ S γ -ray beam, the activated ^{240}Pu targets were monitored by a 60% relative efficiency HPGe. The first target with 2 hours of irradiation was counted for ~ 90 hours while the second, long-lived FPY target was being irradiated. After the long irradiation was completed, the second ^{240}Pu target replaced the first one in the same HPGe counting station. Thus both ^{240}Pu targets were monitored by the same HPGe at the same distance. Since this HPGe detector is the same one that was used in the $^{239}\text{Pu}(n, f)$ measurements of Gooden *et al.* [11] (and in the supplemental $^{239}\text{Pu}(n, f)$ measurements made recently), all of the FPY measurements are on the same systematic footing. Consequently this work possesses a unique sensitivity for comparing FPY distributions from the $^{239}\text{Pu}(n, f)$ and $^{240}\text{Pu}(\gamma, f)$ reactions. The measured fission product γ -ray spectra from the $^{239}\text{Pu}(n, f)$ and $^{240}\text{Pu}(\gamma, f)$ reactions are compared in Fig. 1.

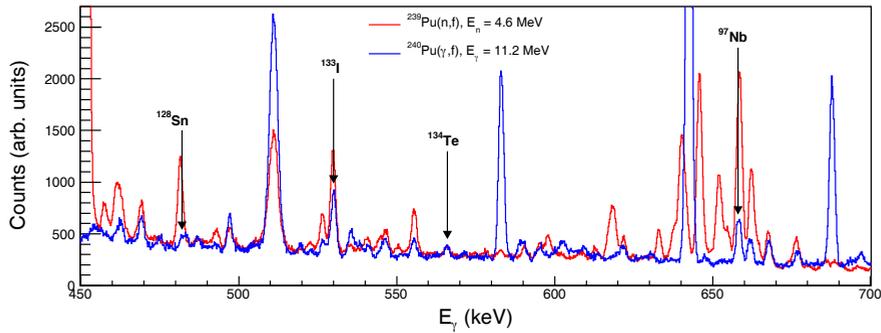


Figure 1. γ -ray spectra from fission products from $^{240}\text{Pu}(\gamma, f)$ at $E_\gamma = 11.2$ MeV and $^{239}\text{Pu}(n, f)$ at $E_n = 4.6$ MeV, measured by a 60% relative efficiency HPGe. Both spectra come from targets which were irradiated for 2 hours.

2.5 Analysis & data reduction

Yields of specific fission products are determined by counting γ -rays emitted by the decay of the corresponding fission products. Choosing the counting window in time is complicated by the interplay of the activity of the γ -ray of interest, interfering background peaks and the smooth continuum background. Because all of these features are time dependent, it can be difficult to predict a priori what the ideal integral time window is to fit the net photopeak area. In this work, a procedure was developed to automatically determine the integration time in a way which minimizes the uncertainty in the net photopeak counts, which is often one of the largest sources of uncertainty in the short-lived FPY measurements. Multiple spectra were generated with each successive spectrum having a longer counting time. The photopeak of interest was fit in each spectrum with a Gaussian function, including a linear background and other Gaussian peaks. The uncertainty of the net photopeak counts from the fit was then plotted as a function of counting time. In addition to minimizing the uncertainty in the photopeak counts, this method removes the choice of the counting period as a potential source of bias.

Using the same formalism as in Ref. [11], the FPYs can be defined as:

$$\text{FPY}_i = \frac{\lambda_i N_i}{F_T n_a I_{\gamma i} \epsilon_i} \frac{1}{1 - e^{-\lambda_i t_e}} \frac{1}{e^{-\lambda_i t_d}} \cdot \frac{1}{1 - e^{-\lambda_i t_m}} \prod_k C_{ki}, \quad (1)$$

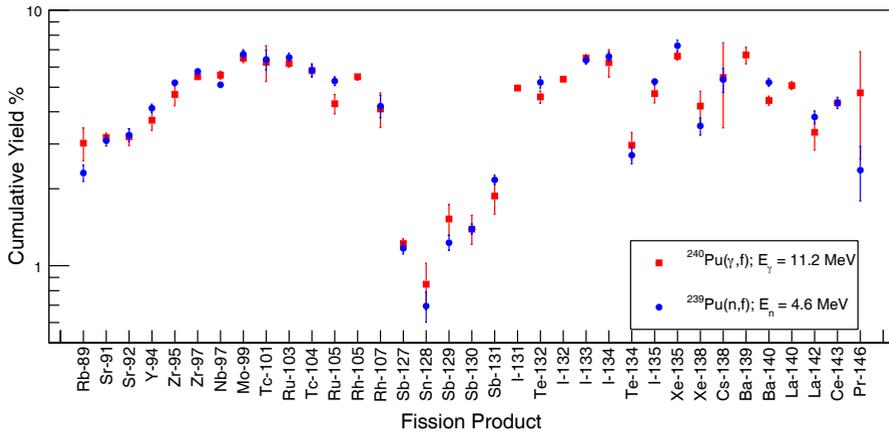


Figure 2. Preliminary FPYs from $^{240}\text{Pu}(\gamma, f)$ at $E_\gamma = 11.2$ MeV and $^{239}\text{Pu}(n, f)$ at $E_n = 4.6$ MeV. The $^{239}\text{Pu}(n, f)$ FPYs include new measurements of short-lived FPYs in combination with 13 long-lived FPYs from Gooden *et al.* [11]

where

$$\begin{aligned}
 \lambda_i &= \text{Decay constant} \\
 N_i &= \text{Number of counts in the photopeak area} \\
 n_a &= \text{Number of target nuclei} \\
 I_{\gamma i} &= \text{Branching ratio} \\
 \epsilon_i &= \text{Photopeak efficiency of HPGe detector} \\
 F_T &= \text{Fission rate in target determined from the DFC} \\
 t_e &= \text{Time of beam exposure} \\
 t_d &= \text{Decay time from end of activation} \\
 t_m &= \text{Target measurement time} \\
 C_{ki} &= \text{Correction factors}
 \end{aligned} \tag{2}$$

The correction factors include effects due to beam fluctuations, γ -ray attenuation and summing in the HPGe.

3 Results and discussion

The 35 unique FPYs measured from $^{240}\text{Pu}(\gamma, f)$ at $E_\gamma = 11.2$ MeV are plotted in Fig. 2 along with the corresponding yields from $^{239}\text{Pu}(n, f)$ at $E_n = 4.6$ MeV, if available. There were 29 unique FPYs which were measured for both sets of data. The FPYs are generally in good agreement, though there are some notable differences where the yields differ by multiple standard deviations. Significantly, the light mass peak in the FPY distribution doesn't show a systematic shift between the two distributions. Such a shift is expected in the case where the fissioning compound nucleus has a different mass, as the heavy fragment tends to remain constant due to shell closures, and additional nucleons are added to the light fragment. The lack of a systematic difference between the two FPY distributions is consistent with Bohr's hypothesis.

In order to quantify the similarity between the two sets of FPYs, a goodness of fit value may be defined as

$$\chi^2 = \sum_i \frac{\left(Y_{^{240}\text{Pu}(\gamma, f)}(^A Z_i) - Y_{^{239}\text{Pu}(n, f)}(^A Z_i) \right)^2}{\sigma_{^{240}\text{Pu}(\gamma, f)}^2 + \sigma_{^{239}\text{Pu}(n, f)}^2}, \quad (3)$$

where $Y_{^{240}\text{Pu}(\gamma, f)}(^A Z_i)$ and $Y_{^{239}\text{Pu}(n, f)}(^A Z_i)$, are the yields of the fission product $^A Z_i$ from $^{240}\text{Pu}(\gamma, f)$ and $^{239}\text{Pu}(n, f)$, respectively; and $\sigma_{^{240}\text{Pu}(\gamma, f)}$ and $\sigma_{^{239}\text{Pu}(n, f)}$ are the uncertainty in the $^{240}\text{Pu}(\gamma, f)$ and $^{239}\text{Pu}(n, f)$ FPYs, respectively. The number of degrees of freedom is equal to the number of pairs of FPYs. A histogram of the χ^2 for each individual FPY pair is shown in Fig. 3.

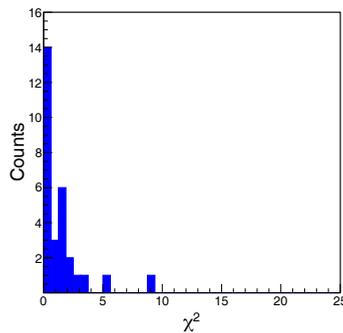


Figure 3. Preliminary χ^2 distribution for the comparison of FPYs from $^{240}\text{Pu}(\gamma, f)$ at $E_\gamma = 11.2$ MeV and $^{239}\text{Pu}(n, f)$ at $E_n = 4.6$ MeV.

The total χ^2 per degree of freedom for this work is 1.36, reasonably consistent with the expected value of 1 if the FPY distributions from $^{240}\text{Pu}(\gamma, f)$ and $^{239}\text{Pu}(n, f)$ at the same compound nuclear excitation energy are the same. Thus, these preliminary results are consistent with Bohr's hypothesis.

4 Conclusion & Future Work

This work represents the first quantitative comparison of FPYs from the $^{240}\text{Pu}(\gamma, f)$ and $^{239}\text{Pu}(n, f)$ reactions at the same compound nuclear excitation energy. The primary goal of the present experiments was to obtain an accurate comparison of the $^{240}\text{Pu}(\gamma, f)$ and $^{239}\text{Pu}(n, f)$ FPY measurements. The required accuracy was obtained by using the same experimental techniques and exact same HPGe detectors for both measurements, eliminating systematic errors which would otherwise make a direct comparison of the data sets difficult.

In order to improve the validation of Bohr's hypothesis in the context of fission, we have performed additional measurements of the FPYs from the $^{240}\text{Pu}(\gamma, f)$ reaction at $E_\gamma = 8.0$ and 11.2 MeV. The 11.2 MeV measurement was performed with significantly greater γ -ray beam flux, which will increase the number of measurable yields and decrease the statistical uncertainties on the FPYs presented in this work. The 8.0 MeV data set can be compared to existing $^{239}\text{Pu}(n, f)$ data at $E_n = 1.5$ MeV [11], allowing us to test if the energy evolution of the FPYs from $^{240}\text{Pu}(\gamma, f)$ and $^{239}\text{Pu}(n, f)$ are consistent. The $E_\gamma = 8.0$ MeV data, in conjunction with the $E_n = 1.5$ MeV data, should also be more sensitive to the pairing and shell effects which are more pronounced at these relatively low excitation energies. In this case the small mismatch of the angular momentum in the entrance channel will be further investigated.

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