Correlations Between $\gamma$-Ray Multiplicity and Compound Nucleus Excitation Energy in $^{239}$Pu$(n,f)$

Nathan Giha, Stefano Marin, James A. Baker, Isabel E. Hernandez, Keegan J. Kelly, Matthew Devlin, John M. O’Donnell, Ramona Vogt, Jørgen Randrup, Patrick Talou, Ionel Stetcu, Amy E. Lovell, Olivier Serot, Olivier Litaize, Abdelhazize Chebboubi, Ching-Yen Wu, Shaun D. Clarke, and Sara A. Pozzi

1 Department of Nuclear Engineering and Radiological Sciences, University of Michigan, Ann Arbor, MI 48109, USA
2 Physics Division, Los Alamos National Laboratory, Los Alamos, NM 87545, USA
3 Nuclear and Chemical Sciences Division, Lawrence Livermore National Laboratory, Livermore, CA 94550, USA
4 Physics and Astronomy Department, University of California, Davis, CA 95616, USA
5 Nuclear Science Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA
6 Computational Physics Division, Los Alamos National Laboratory, Los Alamos, NM 87545, USA
7 Theoretical Physics Division, Los Alamos National Laboratory, Los Alamos, NM 87545, USA
8 CEA, DES, IRESNE, DER, SPRC, Physics Studies Laboratory, Cadarache, F-13108 Saint-Paul-lès-Durance, France
9 Department of Physics, University of Michigan, Ann Arbor, MI 48109, USA

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We study $\gamma$-ray emission following $^{239}$Pu$(n,f)$ over an incident neutron energy range of $2 < E_i < 40$ MeV. We present new experimental evidence for positive correlations between the total angular momentum generated in fission and the excitation energy of the compound nucleus prior to fission. The $\gamma$-ray multiplicity increases linearly with incident energy below the $2^+$-chance fission threshold with a slope of $0.085 \pm 0.010 \, \gamma/\text{MeV}$. This linear trend appears to hold for the excitation energy of the compound nucleus between $9 < \langle E_x \rangle < 19$ MeV. Most of the multiplicity increase comes from a resonance around a $\gamma$-ray energy of 0.7 MeV, which we interpret as stretched quadrupole $\gamma$ rays that indicate an increase in total fission-fragment angular momentum.

Nuclear fission was discovered over eighty years ago [1, 2] but the microscopic details of the process are still not fully understood. The importance of fission in the r-process of nucleosynthesis [3,7], synthesis of superheavy nuclei [8, 9], and developing Generation-IV fast-fission reactors [10] has motivated renewed interest in predictive fission models like CGMF [11], FIFRELIN [12], and FREYA [13]. One of the most prominent questions in contemporary fission physics is the nature of the mechanism by which two fragments, each with 6-8 $h\ell$ of angular momentum, emerge from a system with zero or near-zero angular momentum. Recently, there has been much discussion regarding angular momentum generation in fission [14, 15]. This discussion highlights the lack of definitive experimental evidence for any particular angular momentum generation mechanism. Experimentally-determined correlations between fission observables offer powerful tests of fission models and will be instrumental in discovering which mechanism is correct.

Emerging fission fragments quickly de-excite, rendering the intrinsic angular momenta of the fragments after scission inaccessible in the laboratory [20]. This information is encoded in the subsequent fragment de-excitation via neutron and $\gamma$-ray emission. Electric quadrupole ($E2$) transitions along yrast bands, in particular, remove much of the intrinsic angular momentum [15, 21]. Thus, correlations between these $\gamma$ rays and other fragment properties, such as excitation energy, are an experimentally-accessible signature of correlations between angular momentum and excitation energy in fission. Understanding the relationship between the excitation energy of the fissioning system—and consequently of the fragments—and their angular momenta is critical for constraining the possible mechanisms of angular momentum generation. For example, the popular statistical model posits that the high angular momenta with which fragments emerge are solely due to the higher density of high-angular momentum states at large excitation energy [22]. This model would result in a nonlinear dependence of angular momentum on excitation energy.

Experimental investigations on the dependence of $\gamma$-ray emission on the energy of the fissioning system are sparse. In most cases, the experiments investigated only a few different energies, or a limited energy range, and thus could not resolve any trends. Table I summarizes these experiments with the investigated reaction, energies, and whether or not they were able to resolve changes in the $\gamma$-ray multiplicity and spectrum. The ENDF/B-VIII.0 evaluation for $^{239}$Pu$(n,f)$ is also included. Note that only Gjostvang et al. identified a significant change in $\gamma$-ray multiplicity. Only Laborie et al. found changes in the $\gamma$-ray spectrum, but they observed these changes exclusively above 2 MeV in $\gamma$-ray energy, uncharacteristic of yrast transitions.

In this paper we analyze the $^{239}$Pu$(n,f)$ data from Kelly et al. [31], in which a broad range of excited states of $^{240}$Pu$^+$ were populated. We present clear experimen-
Since the target nucleus $^{239}\text{Pu}$ is unstable to $\alpha$ decay, the PPAC signal from pileup of multiple $\alpha$ events cannot always be separated from that produced by decelerating fission fragments. The bias associated with erroneous triggers from $^{239}\text{Pu}$ $\alpha$ decay is estimated by examining the measured PPAC activity and spectrum in the absence of beam. We quantify the chance coincidence $\gamma$-ray background by introducing a random coincidence signal in the analysis. Its contribution is small and we subtract it. While multiple $\gamma$ rays and neutrons are usually emitted in the same fission, pileup can be neglected due to the low absolute efficiency of the detector array.

The pulsed nature of the broad-spectrum neutron beam results in low-energy neutrons from a previous beam micropulse arriving at the target simultaneously with high-energy neutrons from the current micropulse. We estimate the amount of fission induced by these low-energy neutrons and subtract. This correction is negligible at low $E_\gamma$ and never exceeds 3.4% as $E_\gamma$ approaches 40 MeV.

We apply the following unfolding procedure to recover the emitted $\gamma$-ray spectrum at each $E_\gamma$: we first model the system response of the Chi-Nu liquid scintillator array using isotropic, monoenergetic photon sources in MCNPX-POLINI [34]. We then convolve the resulting response matrix with experimentally-determined detector resolution and then invert it via Tikhonov regularization [37] to correct the measured multiplicity for efficiency and unfold the emitted $E_\gamma$ spectrum from the measured $\gamma$-ray light output spectrum. We determine an energy acceptance window for the unfolded spectra of $0.4 < E_\gamma < 2.2$ MeV by comparing the unfolded $\gamma$-ray spectrum at our lowest data point, $2 < E_\gamma < 3$ MeV, with the ENDF/B-VIII.0 evaluated spectrum for $^{239}\text{Pu}(n_{\text{th}},f)$ [35]. The $\bar{N}_\gamma$ reported throughout this paper thus includes only gamma rays within this acceptance window, representing $\approx 60\%$ of the integrated $^{239}\text{Pu}(n_{\text{th}},f)$ $\gamma$-ray spectrum above 0.1 MeV. Almost all of the excluded $\gamma$ rays fall below the acceptance region. We estimate the unfolding uncertainty in $\bar{N}_\gamma$ by constructing a covariance matrix by varying the regularization parameter.

In Fig. 1, we present the relationship between $\bar{N}_\gamma$ and $E_\gamma$ for $2 < E_\gamma < 40$ MeV. Our data show a clear increase in $\bar{N}_\gamma$ across the entire $E_\gamma$ range. Uncertainties include variation across PPAC foils and unfolding. Statistical uncertainties are comparatively negligible. Also plotted in Fig. 1(a) are $\gamma$-ray multiplicities from the ENDF/B-VIII.0 evaluation [25] and data from Qi [26] and Laborie [27]. These data are scaled to match our $0.4 < E_\gamma < 2.2$ MeV acceptance region. We integrate the ENDF/B-VIII.0 $^{239}\text{Pu}(n,f)$ and $^{238}\text{U}(n,f)$ $\gamma$-ray spectra within our acceptance range, then again for a threshold $E_\gamma > 0.1$ MeV. Most of the experimental results are reported for a 0.1 MeV threshold and extend up to sufficiently high $E_\gamma$ that their upper limit does not significantly affect $\bar{N}_\gamma$. Thus, the evaluation and experimental data in Fig. 1 are scaled by the ratio of these two integrals for the appropriate reaction.

### Table I: Fission $\gamma$-ray measurements and whether they were able to discern changes in $\gamma$-ray multiplicity, $\Delta N_\gamma$, and changes in the $\gamma$-ray spectrum, $\Delta \Sigma_{\gamma}$. For neutron-induced reactions other than $^{239}\text{Pu}(n,f)$, $E_\gamma$ above the $2^{0}\text{MeV}$-chance fission threshold are omitted. Experiments by Fréhaut are frequently cited in discussions about the energy dependence of angular momentum in fission, but the conclusions in Refs. [23] and [24] are contradictory.

| Reference   | Reaction          | $E_{\gamma}$ | $E_{\gamma}$ | $\Delta N_\gamma$ | $\Delta \Sigma_{\gamma}$ |
|-------------|------------------|--------------|--------------|-------------------|-------------------------|
| This work   | $^{239}\text{Pu}(n,f)$ | 2-40         | 9-19         | ✓                 | ✓                       |
| ENDF/B-VIII.0 [25] | $^{239}\text{Pu}(n,f)$ | 0-20         | 6.53-19      | ✓                 | ✓                       |
| Fréhaut [23, 24] | $^{235}\text{U}(n,f)$ | 1.14-14.66   | 7.69-12.22   | N/A               | N/A                     |
| Qi [26]     | $^{239}\text{U}(n,f)$ | 1.90,4.90    | 6.71,9.61    | ✓                 | ✓                       |
| Laborie [27] | $^{239}\text{U}(n,f)$ | 1.6,5.1,15.0 | 6.41,9.91    | ✓                 | ✓                       |
| Oberstedt [28] | $^{239}\text{U}(n,f)$ | $E_\gamma = 1.7$ | $E_\gamma = 8.25$ | ✓ | ✓ |
| Rose [29]   | $^{233}\text{U}(d,p)$ | -            | 4.8-10       |                   |                         |
| Rose [29]   | $^{239}\text{Pu}(d,p)$ | -            | 4.5-8.8     |                   |                         |
| Gjestvang [30] | $^{240}\text{Pu}(d,p)$ | -            | 5.5-8.5     | ✓                 | ✓                       |
We note that \( \overline{N}_x \) varies linearly with \( E_i \) below the 2\(^{\text{nd}}\)-chance fission threshold with a slope of 0.085 ± 0.010 \( \gamma/\text{MeV} \) and that extrapolating this fit down to \( E_i \approx 0 \) yields good agreement with the well-studied multiplicity at thermal fission [39]. Uncertainty on the slope includes variation across PPAC foils, uncertainty from unfolding, and estimated variance of the fitted slope. We do not necessarily expect the Qi [26] and Laborie [27] data to agree with our data since they study a different reaction. The ENDF/B-VIII.0 points above thermal fission were inferred from total \( \gamma \)-ray production data, assuming a 20% uncertainty [38].

In Fig. 1(b), we compare our data to predictions of \( \overline{N}_x \) as a function of \( E_i \) from the release versions of CGMF and FREYA, integrated over the acceptance region. 0.4 < \( E_\gamma \) < 2.2 MeV. CGMF predicts a similar trend, although the discontinuities at the \( n^\text{th} \)-chance fission thresholds are overemphasized compared to experiment. FREYA predicts about 0.5 too few \( \gamma \) rays within the acceptance region. The model uncertainties are statistical.

The neutron separation energies, \( S_n \), of different fissioning isotopes can vary by several MeV so comparing \( \gamma \)-ray emission from different reactions at a given \( E_i \) is not always appropriate. It is instructive to instead look at the excitation energy of the fissioning nucleus, \( E_x \), which is independent of this variation. If we neglect the small kinetic energy imparted to the compound nucleus by the incident neutron, the excitation energy of the fission \( ^{240}\text{Pu}^* \) nucleus is

\[
E_x = E_i + S_n^{(240)},
\]

where \( E_i \) is the incident neutron energy and \( S_n^{(240)} = 6.53 \text{ MeV} \) is the neutron separation energy of the compound \( ^{240}\text{Pu}^* \) nucleus. However, the \( E_x \) — and in fact, the isotope—of the compound nucleus just before fission cannot be uniquely determined once the incident neutron energy exceeds the fission barrier height, \( B_i \), due to the presence of multi-chance fission and pre-equilibrium neutron emission. Thus, multiple \( E_x \) values are possible for a given \( E_i > B_i \) and the average excitation energy, \( \langle E_x \rangle \), of the fissioning nucleus is generally lower than what may be expected from Eq. (1). At a fixed \( E_i \), \( \langle E_x \rangle \) can be written

\[
\langle E_x \rangle = E_i + S_n^{(240)} - \sum_{j=1} \left[ S_n^{(240-j+1)} + \langle k_j \rangle \right] p_j \tag{2}
\]

where \( S_n^{(240-j+1)} \) is the separation energy of the \( j^\text{th} \) neutron, \( \langle k_j \rangle \equiv \langle k_j \rangle(E_i) \) is the average kinetic energy of a pre-fission neutron, and \( p_j = p_j(E_i) \) is the relative probability of emitting \( j \) neutrons prior to fission. Note that Pu isotopes lighter than \( ^{240}\text{Pu}^* \) contribute to the total observed fissions when pre-fission neutron emission occurs. For compound nuclei that are close in mass, correlations between \( \langle E_x \rangle \) and \( \gamma \) rays should be relatively independent of the isotope. \( \langle E_i \rangle \) and \( p_j \) are model dependent; \( \langle k_j \rangle \) was estimated using CGMF and \( p_j \) was calculated using the ENDF/B-VII.1 cross sections [41].

\( E_x \) becomes a better description for the state of the compound nucleus just before fission once \( E_i > B_i \). To investigate the relationship between \( \overline{N}_x \) and \( E_x \), in Fig. 2, we translate \( E_i \) to \( E_x \) using Eq. (2). This translation corrects for the effects introduced by pre-fission neutron emission and reveals the approximate linearity of \( \overline{N}_x \) with respect to \( \langle E_x \rangle \) for 9 < \( \langle E_x \rangle \) < 19 MeV. The model-dependent parameters \( p_j \) and \( \langle k_j \rangle \) in Eq. (2) bias the translation, so we assign 10\% uncertainties to \( p_j \) and \( \langle k_j \rangle \) which give rise to the horizontal uncertainties on our data. The models do not predict these values for \( E_i > 20 \text{ MeV} \), so the data above this limit are excluded from Fig. 2.

Also plotted in Fig. 2 are the ENDF/B-VIII.0 evaluation [29] and the Qi [26], Laborie [27], Rose [28], and Gjestvang [30] data. The energy transformation in Eq. (2) was also applied to the ENDF/B-VIII.0 evaluation. The incident energies of Qi and Laborie are shifted using Eq. (1). The \( E_i = 15.0 \text{ MeV} \) point from Laborie is omitted due to lack of nuclear data for determining \( p_j \) and \( \langle k_j \rangle \) for \( ^{238}\text{U}(n,f) \).

Our data agree well with other experiments in the limited range of overlap, although agreement with our extrapolation to lower \( E_x \) is mixed. We note in the cases...
of Rose [29] and Gjestvang [30] that some disagreement could arise from ion-induced fission populating different states of the compound nucleus [42, 43]. Recent theoretical work, however, concluded that the angular momentum of the compound nucleus has little effect on the angular momenta of the fragments [14].

In Fig. 2(b) we compare our data to predictions from CGMF, FREYA, and FIFRELIN, integrated over the acceptance region, $0.4 < E_\gamma < 2.2$ MeV. In CGMF and FREYA, simulated events were binned by compound nucleus excitation energy. The excitation energy of the compound nucleus was directly specified in FIFRELIN. CGMF predicts the $\bar{N}_{\gamma}$ well across the entire $\langle E_x \rangle$ range—with some deviation at high $\langle E_x \rangle$, where we expect the energy translation in Eq. (2) be more uncertain. FIFRELIN also predicts the trend well, but the absolute multiplicity within our acceptance window, although it still predicts positive correlations. The model uncertainties are statistical.

In Fig. 3(b), slopes from fits to models are shown for comparison. The model uncertainties are standard fit-parameter uncertainties. We observe good agreement with FIFRELIN, which correctly predicts the magnitude of the resonance around $E_\gamma = 0.7$ MeV. CGMF agrees somewhat around the resonance, but does not predict the dip around $E_\gamma = 0.5$ MeV that we observe in our data. FREYA does not predict the observed resonance around $E_\gamma = 0.7$ MeV. Most of the additional $\gamma$ rays that it predicts lie below our acceptance region, explaining the discrepancy between FREYA and our data in Figs. 1(b) and 2(b). We believe that FIFRELIN agrees well partially because it creates artificial levels in nuclei where compiled discrete level libraries like RIPL [40] are lacking.

We have presented the first direct measurement of $\gamma$-ray multiplicity, $\bar{N}_{\gamma}$, across a large incident neutron energy range, $2 < E_i < 40$ MeV, in the $^{239}$Pu(n,f) reaction. We observe a clear increase in $\bar{N}_{\gamma}$ over the entire range. We find an approximately linear relationship between $\bar{N}_{\gamma}$ and $E_i$ below the $2^{\text{nd}}$-chance fission threshold, with a slope of $0.085 \pm 0.010 \gamma/\text{MeV}$. This relationship is preserved upon translating incident neutron energy to compound nucleus excitation energy in the range $9 < \langle E_x \rangle < 19$ MeV. These extra $\gamma$ rays are found around energies characteristic of stretched electric quadrupole transitions, experimentally confirming positive correlations between the excitation energy of the compound nucleus and the total angular momenta of the fragments. In future experiments, we plan to probe lower $E_x$, which will be more sensitive to the functional form of the angular momentum dependence, by examining the relationship between $\gamma$-ray emission from $^{252}$Cf(sf) and fragment total kinetic energy. We also suggest induced-fission experiments with higher-resolution $\gamma$-ray detectors to resolve the low-energy region of the $E_\gamma$ spectrum, as well as unambiguously identify known $E2$ transitions on an event-by-event basis.

1 FIFRELIN calculations were done by adjusting the available 4 free parameters in order to reproduce the total prompt neutron multiplicity provided in the JEFF-3.3 library [44].
FIG. 3. Dependence of the slope, $\Delta N_\gamma / \Delta \langle E_x \rangle$, on $E_x$. In (a), $\gamma$-ray spectra from the experiment for $\langle E_x \rangle = 9, 12.1, 15$, and $17.5$ MeV are also shown on the right-hand side. The area outside the $E_\gamma$ acceptance region is shown as the grey shaded region. $E_\gamma$ bins are 0.1 MeV.

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