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Gamma-ray Output Spectra from ^{239}Pu Fission

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Abstract. Gamma-ray multiplicities, individual gamma-ray energy spectra, and total gamma energy spectra following neutron-induced fission of ^{239}Pu were measured using the DANCE detector at Los Alamos. Corrections for detector response were made using a forward-modeling technique based on propagating sets of gamma rays generated from a paramaterized model through a GEANT model of the DANCE array and adjusting the parameters for best fit to the measured spectra. The results for the gamma-ray spectrum and multiplicity are in general agreement with previous results, but the measured total gamma-ray energy is about 10% higher. A dependence of the gamma-ray spectrum on the gamma-ray multplicity was also observed. Global model calculations of the multiplicity and gamma energy distributions are in good agreement with the data, but predict a slightly softer total-energy distribution.

1 Introduction

Measurements of the multiplicity and energy distributions of gamma rays emitted after neutron-induced fission provide insight into the physics of the fission process and also provide valuable information for power reactor design and global security applications. Gamma-ray emission is modelled as the decay of high-spin, high-excitation fission fragments after neutron decay has removed most of the excitation energy. Calculation of gamma spectra relies not only on fission parameters such as the spin and excitation of the fission fragments, but also on models for optical potentials, radiative strength functions, and level densites for unstable neutron-rich nuclei. Since these are usually poorly known, experimental measurements provide important constraints on the calculations. There are few previous measurements of the prompt gamma spectra from ^{239}Pu, and the two most widely cited measurements [1, 2], were both done at thermal neutron energies. The fission-product mass distributions change with incident neutron energy, resulting in different gamma-ray distributions, but little change is expected over the thermal to low keV region.

2 Experiment

The measurement was made using the Detector for Advanced Neutron Capture Experiments (DANCE) at the Los Alamos Neutron Science Center (LANSCE.) More details of the measurement and it's analysis have been published [3]. Briefly, DANCE is a nearly 4π array of 160 BaF_2 crystals, each 0.75 liter in volume. The inner radius of the array is 17 cm, and each crystal is 15 cm deep. The high granularity and large solid angle of the detector enables a direct measurement of the gamma-ray multiplicity and total energy distribution. The target was surrounded by a ^{6}LiH sphere to absorb low-energy scattered neutrons which can capture in the BaF_2, producing a background. Fission neutrons are not as efficiently absorbed, but after imposing a 20 ns coincidence window on the time between the fission tag from the PPAC (see below) and DANCE, the efficiency for their detection was only about 0.3%.

DANCE is located on Flight Path 14 at the Manuel J. Lujan Jr. Neutron Scattering Center, a spallation neutron source driven by 800 MeV protons. DANCE views a room temperature water moderator with a flight path of 20.25 m. For a proton beam current of 100 μA, the neutron flux was approximately $10^4/E^{-1.03}$ neutrons/(cm^2-eV-sec), where E is the neutron energy in eV, at the neutron monitor location 2 m downstream of the target.

The ^{239}Pu target consisted of a total of 937 μg of ^{239}Pu electroplated on both sides of a 3 μm Ti foil that served as the cathode of a fast-timing parallel-plate avalanche counter (PPAC) [4], which provided a fission-tagging signal. The deposit area was 0.385 cm^2, and the ^{239}Pu was enriched to 99.967%. The efficiency of the PPAC for detecting fissions was approximately 70%.

Even though the efficiency for detecting a full-energy gamma ray was about 85%, detector response corrections are large. This is immediately obvious by noting that the efficiency for detecting 7 fission gammas (the average number) is $(0.85)^7 = 0.32$! There are three ways to make the response correction. The first is the "spectrum stripping" method, which subtracts the calculated energy-dependent response bin-by-bin, starting at the highest gamma-ray energy. This can be very successfully applied to single-detector setups, as shown by Billnert, *et al.* [5]. The second approach is using an "Inverse Method," also

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called unfolding. This technique is usually expressed as a matrix equation **O** = **R•I**, where **O** is a vector of the observed output spectrum, **I** is the actual emitted spectrum, and **R** is a detector response matrix which can be determined by simulations or measurements. The emitted spectrum can be formally obtained as $\mathbf{I} = \mathbf{R}^{-1} \bullet \mathbf{O}$. This method relies on accurately determining the response matrix **R** and its inverse $\mathbf{R}^{-1}$. This method has been applied to DANCE data, including ^{239}Pu [6].

A third approach, applied in the analysis described here, uses a "Forward Method" which uses an analytic parameterization of the gamma-ray spectra to generate sets of gamma-rays, propagates them through a GEANT4 model to produce observed spectra, and compares the simulated spectra to the actual measured ones. The parameters of the input spectra are then varied to produce a best fit to the measurements. Ultimately, a real physics models could be used, but while the parameterization we will use is motivated by physics, it is **NOT** a real physics model. The six-parameter model is described in more detail by Jandel [7]. Briefly, the observed gamma-ray multiplicity is taken as the sum over two distributions:

$$M_\gamma = M_1 + M_2 \tag{1}$$

The gamma multiplicity is assumed to be the equal to the spin, an approximation roughly valid for E1 and M1 transitions, so the multiplicity probability distribution is taken to have the same functional form as the spin distribution

$$P(M_i) = (2M_i + 1)e^{-M_i(M_i+1)/B_i^2} \tag{2}$$

where B_i are fitted parameters. The gamma-ray energy distributions for the two multiplicities were taken as

$$P_1(\varepsilon) = \varepsilon^2 e^{-(a_1+M_\gamma b_1)\varepsilon} \tag{3}$$

$$P_2(\varepsilon) = \varepsilon^3 e^{-(a_2+M_\gamma b_2)\varepsilon} \tag{4}$$

where a_1, b_1, a_2, and b_2 are fitted parameters. Note that since the observed gamma-ray spectra are the sum over many fission products with different excitation energies, temperatures, and gamma multiplicities, this simple model can only represent an average parameterization.

Events consisting of M_γ gamma rays with random energies sampled from the above distributions were then generated and transported through a well-tested GEANT4 model of DANCE [8] to produce "experimental" spectra. Typically, 100,000 events were generated. The simulated spectra were compared to the measured ones, and all six parameters were varied using the "simulated annealing" method [9, 10] to produce a minimum χ^2. More details are provided in ref [3].

3 Results

Spectra generated using the best-fit parameters are shown below, and compared to previous measurements, the results using unfolding techniques, and theoretical calculations, as appropriate. The analysis used only the data from the 10.93 eV plus 11.89 eV 1^+ resonance complex; a comparison to 1^+ resonances at 7.82, 22.26, 75.0 eV and a 0^+ resonance at 32.33 eV showed little difference. The calculations were made using the Monte-Carlo Hauser Feshbach technique [11–13] with the parameter $\alpha = 1.5$. Figure 1 shows the response-corrected gamma-ray multiplicity from this work compared to the MCHF calculation and the result from one-dimensional unfolding [14]. The measured multiplicity is very sensitive to the detector threshold, which was 150 keV for this work.

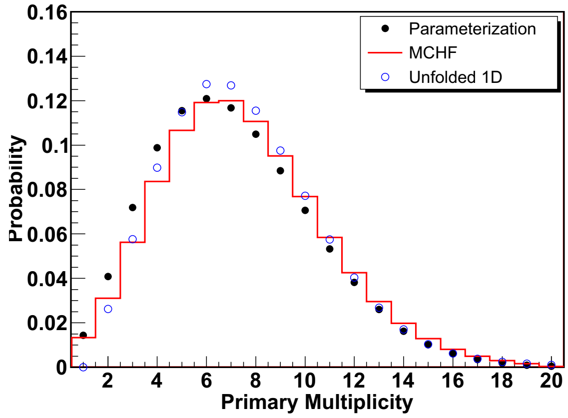

Figure 1. Response-corrected gamma-ray multiplicity from this measurement compared to the MCHF calculation with α=1.5 [11] and the results from one-dimensional unfolding [14]. The detector threshold was 150 keV.

Figure 2 shows the measured gamma-ray energy spectrum for several cluster multiplicities (Mcl), before correction for detector response. Since a gamma-ray interacting in DANCE can produce a signal in a cluster of several adjacent crystals due to pair production and Compton scattering, the cluster multiplicity is a signal proportional to the actual gamma-ray multiplicity. We note an obvious dependence of the shape of the spectra on the cluster multiplicity. This is perhaps expected, but has not been previously observed.

Figure 3 shows the response-corrected individual gamma-ray spectrum, summed over all multiplicities, compared to MCHF calculations, the 1D unfolding results [14], and ENDF/B-VII evaluation [15]. The DANCE data differs from the evaluation and calculation at low energies because of the DANCE resolution and gamma-energy threshold. Finally, Figure 4 shows the response-corrected total gamma ray energy, summed over all multiplicities, compared to the MCHF calculation with α=1.5.

Average multplicity and total gamma-ray energy can be determined from the spectra for comparison to other measurements. These results are shown in Table 3. In general, the average multiplicity determined in this work is in rough agreement with previous measurements and the MCHF calculations, but the average total gamma energy is about 10% higher.

It is interesting to note that while the gamma-ray multiplicity and total energy distributions for several resonances were very similar, the ENDF [15] evaluation shows that the fission neutron multiplicity has up to 10% dips for

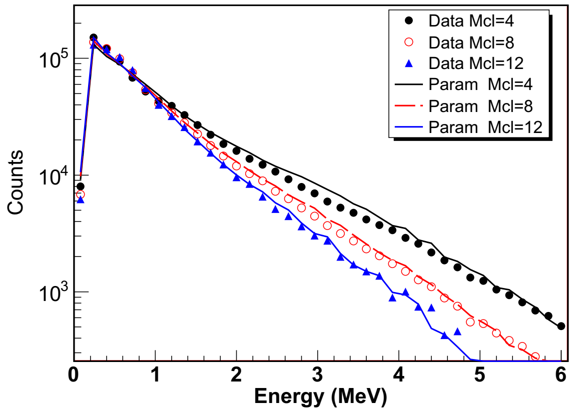

Figure 2. Measured cluster energy (before response correction) for cluster multiplicities 4, 8, and 12. For comparison, the spectra were all normalized to the Mcl=4 data over the 0.2 - 1.1 MeV energy range.

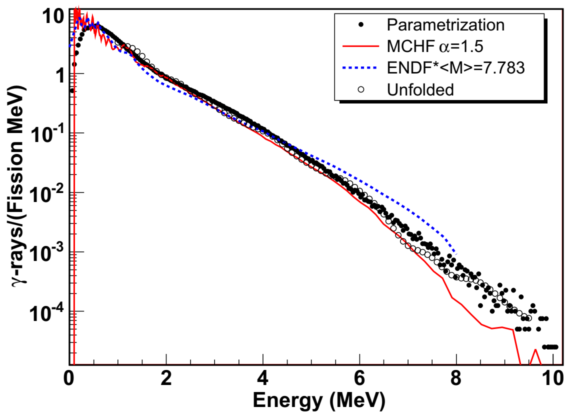

Figure 3. Response-corrected single gamma energy (all multiplicities) compared to ENDF/B-VII [15], the MCHF calculations [11], and results from unfolding [14].

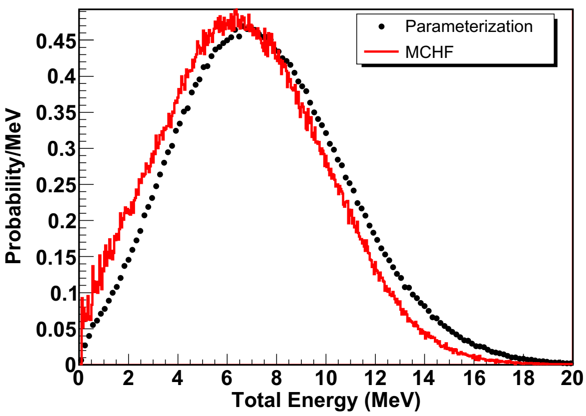

Figure 4. Response-corrected total gamma ray energy compared to the MCHF calculation with α=1.5 [11].

Table 1. Average Multiplicity and total gamma energy

	$\langle M \rangle$	$\langle E_{tot} \rangle$ (MeV)
This Work (10.93 eV)	7.15 ± 0.09	7.46 ± 0.06
Pleasonton [2]	6.88 ± 0.35	6.73 ± 0.35
Verbinski [1]	7.23	6.81
MCHF (α=1.5) [11]	7.05	6.74
ENDF/B-VII [15]	7.78	6.74
2D Unfolding [6]	7.93	7.94

several weak fission resonances. A preliminary look by Mosby at the average total gamma energy for these resonances, at 35.5, 41, 44.5, and 52.7 eV, showed a 5% to 10% increase, hinting at new fission physics to be investigated further.

4 Summary and Conclusions

This paper describes the measurement of the distribution of gamma-ray multiplicity, individual gamma energy, and total gamma energy from neutron-induced fission of ^{239}Pu. Corrections for detector response were made using "forward modeling" which involved generating sets of gamma rays from a parameterization of the data, propagating them through a GEANT model of the DANCE detector, and adjusting the parameters to obtain the best fit. This method requires having a highly-segmented, 4π detector. The multiplicity and individual gamma-ray energy distributions are in agreement with previous measurements, although the results below about 1.5 MeV are sensitive to the detection threshold and resolution of the BaF_2 array. A dependence of the shape of the individual gamma-ray energy distribution on the multiplicity of gamma rays was observed. The average E_{tot} is about 10% higher than previous values. An analysis of the same data set using unfolding techniques produced an even larger value. A theoretical model has been developed that does a good job of predicting these distributions for several actinides, but predicts a lower total energy distribution than observed in this work. The results reported here were obtained by analyzing spectra for the 10.93 eV + 11.89 eV 1^+ resonance complex, and similar results were seen for several other strong resonances. However, more detailed analysis of the resonance behavior by Mosby has indicated that spectra may be different for several weak fission resonances where dips in fission neutron multiplicity were reported. More work is required to understand these anomalies.

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References

[1] V.V. Verbinski, H. Weber, and R.E. Sund, Phys. Rev. C **7**,1173 (1973)

[2] F. Pleasonton, Nucl. Phys. **A213**, 413 (1973)

[3] J.L. Ullmann, E.M. Bond, T.A. Bredeweg, A. Couture, R.C. Haight, M. Jandel, T. Kawano, H.Y. Lee, J.M. O'Donnell, A.C. Hayes, I. Stetcu, T.N. Taddeucci, P. Talou, D.J. Vieira, J.B. Wilhelmy, J.A. Becker, A. Chyzh, J. Gostic, R. Henderson, E. Kwan, and C.Y. Wu,Phys. Rev. C **87**, 044607 (2013)

[4] C.Y. Wu, A. Chyzh, E. Kwan, R.A. Henderson, J.M. Gostic, D. Carter, T.A. Bredeweg, A. Couture, M. Jandel, and J.L. Ullmann, Nucl. Instrum. Methods A **694**, 78 (2012)

[5] R. Billnert, F.-J. Hambsch, A. Oberstedt, and S. Oberstedt, Phs. Rev. C **87**, 024601 (2013)

[6] A. Chyzh, C.Y. Wu, E. Kwan, R.A. Hendrson, T.A. Bredeweg, R.C. Haight, A.C. Hayes-Sterbenz, H.Y. Lee, J.M. O'Donnell, and J.L. Ullmann, Phys. Rev. C **90** 014602 (2014)

[7] M. Jandel, *et. al*, "Prompt gamma-ray emission in neutron-induced fission of ^{235}U," Los Alamos Report LA-UR-12-24975

[8] M. Jandel, T.A. Bredeweg, A. Couture, M.M. Fowler, E.M. Bond, M.B. Chadwick, R.R.C. Clement, E.-I. Esch, J.M. O'Donnell, R. Reifarth, R.S.Rundberg, J.L. Ullmann, D.J. Vieira, J.B. WIlhelmy, J.M. Wouters, R.A. Macri, C.Y. Wu, and J.A. Becker, Nucl. Instrum. Methods **261**, 1117 (2007)

[9] N. Metropolis, A.W. Rosenbluth, M.N. Rosenbluth, A.H. Teller, and E. Teller, J. Chem.Phys. **21**, 1087 (1953)

[10] S. Kirkpatrick, C.D. Gelatt, and M.P. Vecchi, Science **220**, 671 (1983)

[11] I. Stetcu, P. Talou, T. Kawano, and M. Jandel, Phys. Rev. C **90**, 024617 (2014)

[12] T. Kawano, P. Talou, M.B. Chadwick, and T. Watanabe, J. Nucl. Sci. Technol. **47**, 462 (2010)

[13] B. Becker, P. Talou, T. Kawano, Y. Danon, and I. Stetcu, Phys. Rev. C **87**, 014617 (2013)

[14] A. Chyzh, C.Y. Wu, E. Kwan, R.A. Henderson, J.M. Gostic, T.A. Bredeweg, A. Couture, R.C. Haight, A.C. Hayes-Sterbenz, M. Jandel, H.Y. Lee, J.M. O'Donnell, and J.L. Ullmann, Phys. Rev. C **87**, 034620 (2013)

[15] M.B. Chadwick, *et. al*, Nuclear Data Sheets **112**, 2887 (2011)