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Exploratory Study of Fission Product Yield Determination from Photo-Fission of ^{239}Pu at 11 MeV with Monoenergetic Photons

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Measurements of fission product yields play an important role for the understanding of fundamental aspects of the fission process. Recently, neutron-induced fission product-yield data of ^{239}Pu at energies below 4 MeV revealed an unexpected energy dependence of certain fission fragments. In order to investigate whether this observation is prerogative to neutron-induced fission, a program has been initiated to measure fission product yields in photo-induced fission. Here we report on the first ever photo-fission product yield measurement with monoenergetic photons produced by Compton back-scattering of FEL photons. The experiment was performed at the High-Intensity Gamma-ray Source at Triangle Universities Nuclear Laboratory on ^{239}Pu at E_γ=11 MeV. In this exploratory study the yield of 8 fission products ranging from ^{91}Sr to ^{143}Ce has been obtained.

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I. INTRODUCTION

Using continuous-energy neutrons produced in critical assemblies and fast reactors, the fission product yield (FPY) of ^{239}Pu has been reported by Selby *et al.* [1] and Maeck *et al.* [2]. The data presented for the high-yield fission fragment ^{147}Nd in the neutron energy range between 0.2 and 1.9 MeV were evaluated by Chadwick *et al.* [3] and Thompson *et al.* [4]. They found an energy dependence for the ^{147}Nd FPY with positive slope of 3.7%/MeV and 3.2%/MeV, respectively. Very recently, this finding was confirmed by Gooden *et al.* [5] using mono-energetic neutron beams, as opposed to the white neutrons of Refs. [1–3]. The data of Gooden *et al.* [5] extended the positive slope to at least 5 MeV neutron energy. In addition, in Ref. [5], largely unexplained positive slopes were also observed for a number of other high-yield fission products, in the A=95-143 mass range for neutron-induced fission of ^{239}Pu, while this was in general not the case for ^{235}U and ^{238}U. Here, the notation FPY refers to the cumulative yield, i.e., the yield of a particular nuclide after all prompt and delayed decays have occurred. In order to complement the neutron-induced FPY studies, FPY measurements with photons as a probe may shed light on this newly observed phenomenon.

In photo-fission the compound nucleus is the same as the target nucleus A, and its excitation energy is equal to the incident photon energy, in contrast to neutron-induced fission with its compound nucleus A+1 and its excitation energy given by the kinetic energy of the incident neutron plus its binding energy. Furthermore, because the photon carries spin 1, only a restricted number of compound nuclear states are available from which fission can take place, even if the excitation energy is matched to that obtained in neutron-induced fission, where MeV neutrons can transfer a range of angular momenta to the compound nucleus.

Because of the lack of photo-fission experiments with monoenergetic photons, and the observation of an energy dependence of important high-yield fission products obtained from neutron-induced fission of ^{239}Pu at low energies [5], a program was initiated to study photo-fission with monoenergetic photon beams. The first step consists of measurements performed in the same excitation energy range of the compound nuclear systems, ^{240}Pu for ^{239}Pu(n,f) and ^{239}Pu for ^{239}Pu(γ,f). The ultimate goal is, however, to compare the reactions ^{239}Pu(n,f) and ^{240}Pu(γ,f) at the same excitation energy

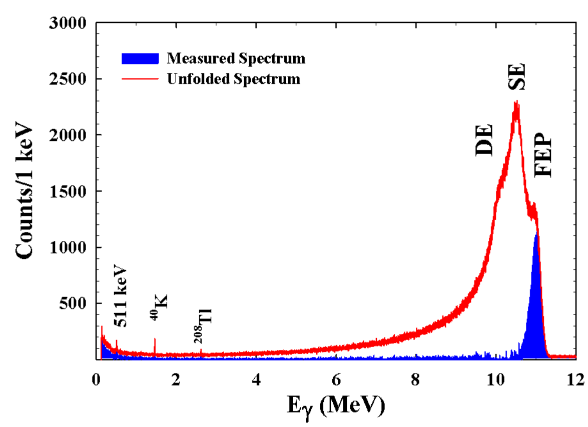

FIG. 1: (Color online)Photon spectrum (in red) measured with a 123% HPGe detector positioned at 0° relative to the incident photon beam of 11.0 MeV. The full-energy peak (FEP), single escape (SE), and double escape (DE) are labeled. In the low-energy part of the spectrum, room background lines are clearly seen. An unfolded spectrum (in blue) of the FEP is indicated by the shaded area.

of the ^{240}Pu compound nucleus. This will provide important test data for fission codes like FREYA [6], GEF [7], and CGMF [8] and fission theory in general. Most

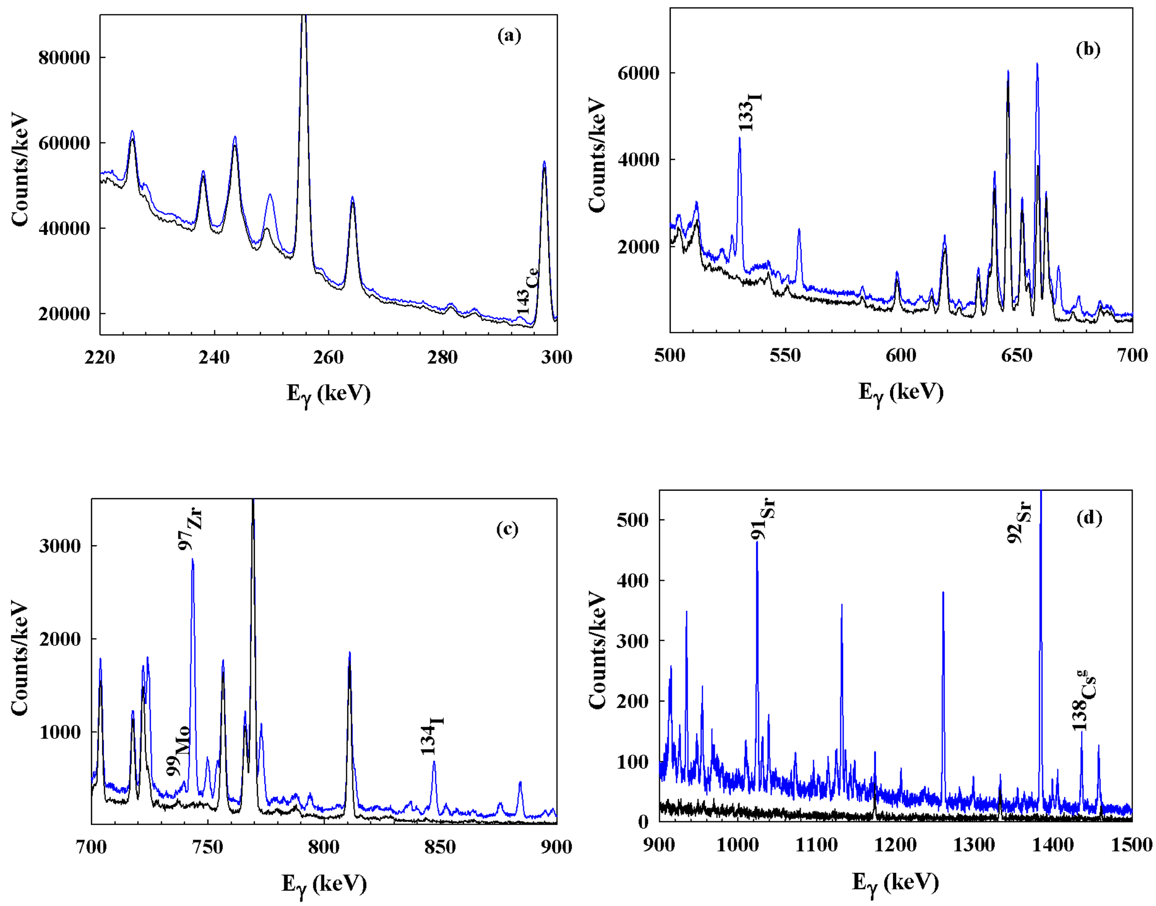

FIG. 2: Gamma-ray energy spectra of interest for ^{239}Pu prior to (black) and after irradiation (blue) with 11 MeV photons. The transitions corresponding to the fission products identified in the present work are labeled. Both spectra are normalized to the same live-time.

likely, the positive slope issue has a nuclear nuclear structure component, caused by the competition between pairing and shell effects. Our measurements will also test the influence of the probe - electromagnetic versus hadronic - on the fission product yields (FPYs). Even if the excitation energy is matched, the compound nucleus is not the same, unless two different target nuclei are used, for example ^{239}Pu and ^{240}Pu, as stated already above. Of course, spin and parity differences of the compound nucleus may still be an issue. All the questions could be answered by precise neutron-and photon-induced FPY experiments.

Here we report on the photo-fission of ^{239}Pu at 11 MeV. The incident photon energy was chosen to compare to the neutron-induced fission data of Gooden *et al.* at 4.49±0.25 MeV, therefore providing the same excitation energy of the compound nucleus ^{239}Pu and ^{240}Pu, respectively.

II. EXPERIMENTAL PROCEDURE, ANALYSIS AND RESULTS

The photo-fission of ^{239}Pu was conducted using TUNL's High Intensity Gamma-Ray Source (HIγS). Details about this free-electron laser based Compton-back scattering facility are given in Ref. [9]. The previously unirradiated ^{239}Pu target was provided by Los Alamos National Laboratory. This target of 1.24 cm diameter and mass of 0.2337 g was sandwiched between Au foils of the same diameter and thickness of 0.01 cm. Other details of the ^{239}Pu target are given in Ref. [5]. The Au foils served for photon fluence determination based on the ^{197}Au(γ,n)^{196}Au reaction of known cross section [10]. The target assembly was irradiated for about 9 hours with approximately 10^7 $\gamma/(\mathrm{cm}^2\mathrm{s})$. The energy distribution of the attenuated photon beam was measured with a 123% HPGe detector at the 0° position (Fig. 1). The energy spread of the 11.0 MeV photon beam was approximately 140 keV (FWHM). A 1.27 cm diameter lead collimator of 15.24 cm length was positioned 2.5 m

TABLE I: Relevant nuclear decay data for identified fission products [12].

Fission Fragment	$T_{1/2}$	E_γ (keV)	I_γ (%)
^{91}Sr	9.63(5)h	1024.3(1)	33.50(1.10)
^{92}Sr	2.611(10)h	1383.93(5)	90(4)
^{97}Zr	16.749(8)h	743.36(3)	93.09(16)
^{99}Mo	65.976(24)h	739.500(17)	12.26(22)
^{133}I	20.83(8)h	529.872(3)	87.0(23)
^{134}I	52.5(2)m	847.025(25)	96(3)
^{138}Cs^g	33.41(18)m	1435.86(9)	76.3(16)
^{143}Ce	33.039(6)h	293.266(2)	42.8(4)

TABLE II: Fission product yield results obtained from photo-fission of ^{239}Pu at E_γ=11 MeV, neutron-induced fission at E_n=4.49 MeV [5] and photo-fission with bremsstrahlung beam of maximum energy $E_{\gamma max}$=28 MeV [14].

Fission Fragment	present data at E_γ = 11 MeV (%)	Ref. [5] E_n = 4.49 MeV (%)	Ref. [14] $E_{\gamma max}$ = 28 MeV (%)
^{91}Sr	4.15±0.51	3.08±0.14	2.89±0.23
^{92}Sr	4.21±0.49	3.25±0.19	-
^{97}Zr	6.63±0.76	5.77±0.18	4.63±0.17
^{99}Mo	7.90±0.94	6.73±0.29	5.76±0.22
^{133}I	7.56±0.89	6.60±0.27	5.43±0.21
^{134}I	7.39±0.89	-	-
^{138}Cs^g	6.18±0.75	-	-
^{143}Ce	4.41±0.51	4.34±0.22	3.26±0.13

upstream the ^{239}Pu target.

After irradiation, the ^{239}Pu target was γ-ray counted over a period of 2 months for a number of different counting cycles ranging from hours (at the very beginning) to days to measure the decay of the induced activity of the fission products. HPGe detectors of 60% efficiency (relative to a 7.62 cm × 7.62 cm NaI detector) in TUNL's low-background counting facility were used for the γ-ray counting of the ^{239}Pu target and gold monitor foils. Both ^{239}Pu and Au foils were counted in acrylic holders at 5 cm distance from the front faces of the detectors. To reduce the high dead time from the inherent radioactivity of the Pu sample, 1.1 mm of Cd was placed inside of the acrylic holder facing the HPGe detector. The cadmium helped to reduce the rate of low-energy γ rays while leaving the lines of interest of higher energy basically unaffected. We have identified the eight fission fragments ^{91}Sr, ^{92}Sr, ^{97}Zr, ^{99}Mo, ^{133}I, ^{134}I, ^{138}Cs^g and ^{143}Ce in this exploratory experiment. Table I provides the relevant spectroscopic information about these isotopes. The cumulative FPYs have been determined, closely following the procedure described in detail in Ref. [5]. The 11 MeV ^{239}Pu(γ,f) and ^{197}Au(γ,n)^{196}Au cross-section values were taken from Refs. [13] and [10], respectively. The results are given in column two of Table II. For comparison, column three of Table II gives the FPYs of Ref. [5] obtained from neutron-induced fission of ^{239}Pu at 4.49 MeV.

For some of the γ-ray transitions shown in Fig. 2 reliable fission product yields could not be obtained due to the lack of statistical accuracy and contamination from γ-ray lines other than those of interest in the present work.

TABLE III: Uncertainty budget.

Source of Uncertainty	Magnitude (%)
Photo-peak area	0.6-2.5
Detector efficiency	1.8-4
γ-ray emission probability	0.1-4.4
Half-life	< 0.6
γ-ray absorption	0.81
Target mass	0.1
Irradiation time	$<< 1$
Decay time	$<< 1$
Counting time	$<< 1$
^{239}Pu(γ,f) cross section	5
^{197}Au(γ,n)^{196}Au cross section	10

Figs. 2(a)-(d) show pulse-height spectra containing γ-ray transitions from the fission products analyzed in the present work. Here, the lower spectrum was obtained before irradiation and the upper spectrum after irradiation, both normalized to the same counting live time. The γ-ray spectra were processed using the peak fitting program TV [11]. Extensive background measurements were performed prior to irradiation in identical geometrical conditions as the counting of the activated target to check for interferences in the region of interest.

III. CONCLUSION

In general, the neutron-induced FPYs agree fairly well with those obtained from photo-fission of ^{239}Pu. The total uncertainty of our data is governed by uncertainties associated with the ^{239}Pu(γ,f) and ^{197}Au(γ,n)^{196}Au cross sections, as can be seen from the error budget of Table III. Finally, for comparison with our mono-energetic FPY data, column four of Table II shows FPYs of ^{239}Pu obtained with bremsstrahlung beam of endpoint energy $E_{\gamma max}$ = 28 MeV [14], corresponding to a mean photon energy of approximately 15 MeV. These higher energy data are lower than our data, consistent with an energy dependence of the FPYs.

In the near future data will be obtained at lower and higher photon energy with considerably improved statistical accuracy. Irradiation times of 100 hours are planned. Compared to the 9 hours used in the present work, this will increase the induced activity over the natural activity of ^{239}Pu considerably. Only then will it be possible to measure the FPY of ^{147}Nd and other fission products. These measurements will employ the ^{239}Pu dual fission chamber of Ref. [5]. Using this approach, the knowledge of photo-fission cross section and incident photon fluence is not required, thus reducing significantly the uncertainty of the FPY results.

IV. ACKNOWLEDGMENTS

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