1 Photofission cross-section ratio measurement of
2 $^{235}\text{U}/^{238}\text{U}$ using monoenergetic photons in the energy
3 range of 9.0 to 16.6 MeV

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10 **Abstract**

Photofission cross-section ratios of ^{235}U and ^{238}U have been measured using monoenergetic photon beams at the HI γ S facility of TUNL. These measurements have been performed in small energy steps between 9.0 and 16.6 MeV using a dual-fission ionization chamber. Measured cross-section ratios are compared with the previous experimental data as well as with the recent evaluated nuclear data library ENDF.

11 *Keywords:* Photon beam, Photofission, Dual-fission chamber

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13 **1. Introduction**

14 Nuclear fission is a complex process in which the collective motion of the
15 nucleons results in a strong change in the shape of the nucleus, in the formation
16 of nucleon configurations, and in the multiple redistribution of internal energy
17 between the different degrees of freedom. Of special interest in fission studies
18 are measurements of the fission cross section, which is the main observable
19 used to test models of the reaction dynamics and associated nuclear structure.
20 The use of an electromagnetic probe in fission studies, such as real photons,
21 simplifies the interpretation of data by offering the advantage of a well known
22 interaction with strict angular-momentum selection rules. Photon absorption

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23 at low energies (below about 100 MeV) is predominantly E1 absorption and to
 24 a smaller extent M1 and E2 absorptions, thus restricting the J^π value of the
 25 initiating excited states in an even-even compound nucleus to 1^- , 1^+ , and 2^+ ,
 26 respectively. Another special feature of photon probes, as opposed to hadronic
 27 probes, is the absence of binding energy of the projectile with the target nucleus
 28 to form the compound nucleus system that fissions. That is, the excitation
 29 energy of the precursor compound nucleus to the fission corresponds to the
 30 energy of the incident photon. In addition, because the photon is electrically
 31 neutral, there is no Coulomb barrier in the entrance channel. These features
 32 give rise to the unique possibility of studying nuclear fission at any excitation
 33 energy above or below the fission barrier, e.g., sub-barrier fission.

34 Precise photon-induced nuclear-reaction measurements are important in a
 35 variety of research and development areas, e.g., basic nuclear physics, activa-
 36 tion analysis of materials, remote detection of nuclear materials, nuclear foren-
 37 sics, production of medical isotopes, physics and technology of fission reactors,
 38 plasma diagnostics in fusion reactors, and characterization and transmutation
 39 of nuclear waste [1–6].

40 The majority of photonuclear cross-section data have been obtained using
 41 bremsstrahlung and quasi-monoenergetic photons from annihilation in flight of
 42 relativistic positrons at the Lawrence Livermore National Laboratory (LLNL) in
 43 the USA and the Centre d'études Nucléaires de Saclay in France [6–9]. Detailed
 44 studies of the photon-induced fission cross section on actinides have been car-
 45 ried out at LLNL by groups from LLNL and Los Alamos National Laboratory
 46 (LANL) [8, 9]. In 1964, Bowman *et al.* [10] measured the photodisintegration
 47 cross sections of ^{235}U using photons obtained from the annihilation in flight of
 48 fast positrons at LLNL. Later, in 1980, Caldwell *et al.* [8] measured the photofis-
 49 sion cross sections for ^{235}U and ^{238}U (along with some other actinides) in the
 50 Giant Dipole Resonance (GDR) region. These experiments were also performed
 51 using photon beams produced by the annihilation in flight of fast positrons at
 52 LLNL. In 1973, Anderl *et al.* [11] reported photofission cross-section measure-
 53 ments on ^{235}U and ^{238}U at $E_\gamma = 5.0 - 8.0$ MeV. The photons were obtained

by Compton scattering of γ rays produced by neutron capture on nickel. Using quasi-monoenergetic photons from e^+ annihilation and direct fission fragment detection at the Giessen positron annihilation facility, Ries *et al.* [12] measured the absolute photofission cross section for ^{235}U and ^{238}U in the energy range from 11.5 to 30.0 MeV. In addition, Soldatov *et al.* [13] measured the photofission cross sections of ^{235}U and ^{238}U using bremsstrahlung.

The main objective of these studies has been to obtain nuclear fission information at excitation energies near the fission threshold and in the GDR region. The data is plagued by clear disagreements in both shape (*i.e.*, energy dependency) and magnitude of the cross section. Furthermore, these disagreements exist not only between data obtained using different methods, but data obtained using the same method. These differences were larger than the reported statistical uncertainties. The majority of these photonuclear reaction cross-section data have been obtained some time ago and are included in the evaluated databases [14]. Therefore, a modern data set is needed to improve the accuracy and reliability of the cross sections.

Using bremsstrahlung or even annihilation in flight of fast positrons imposes some limitations and difficulties for the measurement of photonuclear cross sections. For example, using bremsstrahlung, broad-energy beams one had to step through the bremsstrahlung end-point energy in small increment, and then differentiate it to determine the excitation function [13]. This requires a great stability in the accelerator parameters, enormous counting statistics, and knowledge of the bremsstrahlung flux and spectrum, especially near the end point. The last one is hard to measure or to calculate reliably. In case of annihilation in flight of fast positrons, the effect of the positron bremsstrahlung contaminant has to be subtracted by repeating the same measurements with electrons. As pointed out by several authors [12], reliable data on photofission cross sections can only be obtained using monoenergetic photon sources with good resolution and sufficient intensity. In the present work, we report on the technical development to measure photofission cross section ratios using tunable monoenergetic photon beams provided by the High Intensity γ -ray Source (HI γ S) facility at the

Triangle Universities Nuclear Laboratory (TUNL), that eliminates all of these difficulties using bremsstrahlung beams and those associated with positron in flight annihilation. This technique is illustrated here in the photofission measurements using ^{235}U and ^{238}U .

2. Experimental technique and dual-fission chamber

HI γ S, located on the campus of Duke University, generated circularly polarized and monoenergetic photon beams for the present experiment. Details on the HI γ S facility can be found in Ref. [15] and are briefly summarized here. The HI γ S facility produces a quasi-monoenergetic Compton backscattered photon beam with a wide and tunable energy range and either linear or circular polarization. A high-flux photon beam is created by colliding relativistic electron beam bunches circulating in a storage ring with photon pulses inside the high-power optical cavity of a free-electron laser (FEL). For this experiment 450 and 540 nm FEL wavelength settings and 400 to 650 MeV electron energy settings were used to provide high-energy photon beams between 8.0 and 17.0 MeV. The peak energy of the photon beam was selected by an Al collimator of length 44.5 cm with a cylindrical hole of 1.905 cm diameter and positioned 55 meters downstream from the FEL photon-electron collision point in the storage ring. This collimator diameter results in an energy spread of $\sim 2\text{-}3\%$ (FWHM) of the photon beam. Aluminum was chosen as the collimator material to minimize the number of neutrons produced via (γ, n) reaction because of its high neutron separation energy of 13.6 MeV. Figure 1 shows a schematic diagram of the experimental setup in the Upstream Target Room (UTR) experimental area of the HI γ S facility.

The principal task in obtaining accurate fission cross-section data is the precise determination of the number of fissions which occur during the measurements. To accomplish this, a specially fabricated dual-fission ionization chamber (DFC) [16] was used, as seen in the layout of the present experimental setup (Fig. 1). In its original form, this DFC contains a thick activation

Figure 1: (Color online) Schematic diagram (not to scale) of the experimental setup using a dual-fission chamber and a 123% efficient (relative) HPGe detector. In the present photofission measurements, the thick activation target was not used (shown by the dashed-line at the center of the fission chamber).

114 target at its center, sandwiched by two thin reference foils of the same isotope
 115 in the front chamber and back chamber of the DFC. Such chambers have been
 116 extensively employed at TUNL to perform accurate and systematic investiga-
 117 tions of fission product yields using monoenergetic neutron beams [16, 17]. In
 118 the present measurements, the thick activation target was not used. Reference
 119 foils of ^{235}U and ^{238}U were used in the downstream (FC1) and upstream (FC2)
 120 fission chamber, respectively. By taking the ratio of the number of fissions in
 121 each chamber, one can remove the necessity of knowing the photon flux, as the
 122 same collimated photon flux is seen by both of the chambers. In addition, the
 123 nearly ideal parallel photon beam produced at the target position, eliminates
 124 the need for a source-target angular-correlation correction. This is due to the
 125 long distance (~ 60 m) between the FEL collision point and the DFC.

126 Details of a dual-fission chamber can be found in Refs. [16, 17] and they are
 127 briefly summarized here. The dual-fission chamber was adopted from the design
 128 developed by Gilliam *et al.* at NIST [18]. However, several design changes were

Table 1: Information on the uranium reference foils used in the individual FCs. Diameter of each reference foil is 1.27 cm.

Foil	Chamber	Mass(μ g)	Isotopic abundance(%)
^{235}U	FC1	160.36(1.60)	99.835(0.002)
^{238}U	FC2	121.90(1.22)	99.954(0.002)

made. The gas connections to the individual FCs are made of stainless steel tubing and the fission chambers were made leak tight. The signals from each of the FCs are sent to the manifold via a UT-085 semi-rigid coaxial cable that runs inside the gas line. P-10 (90% argon and 10% methane) was used as the DFC fill gas. The reference foils are electroplated on 0.013 cm thick Ti disks. The active area of the deposit is 1.27 cm in diameter, which is about 7.0 mm smaller than the gamma beam diameter at the target, thereby allowing an alignment tolerance of ± 3.5 mm, which is well within our capabilities. In addition, photon beam imaging was performed at the DFC position using an imaging system [19, 20]. This imaging system has sub-mm spatial resolution (better than 0.5 mm) and a high contrast sensitivity (better than 5%). Table 1 gives detailed information about the reference foils used in the present work. The mass of the reference foils was determined by alpha spectroscopy using well calibrated ionization chambers at LANL, and the mass measurement of each foil is given the conservative uncertainty of $\pm 1\%$.

Signals from the FCs are input to Ortec 142-PC preamplifiers. The preamplified signals are then amplified by a Canberra 2026 spectroscopic amplifier and then fed to a Canberra Multiport II multichannel analyzer, supported by the GENIE2000 software [21].

3. Data analysis and results

For the present measurements circularly polarized photon beams of energies ranging from 9.0 to 16.6 MeV, in 200 keV increments, were used. A 123% effi-

151 cient (for 1332 keV γ rays relative to a standard 7.62 cm x 7.62 cm NaI detector)
 152 coaxial HPGe detector was placed upstream of the fission chamber position to
 153 measure the beam-energy distribution. During beam-energy measurements, the
 154 photon beam was attenuated by a series of copper attenuators mounted up-
 155 stream. Figure 2 shows a typical energy distribution of the incident beam. The
 156 FWHM resolution of the beam at 9.0 MeV is $\sim 2.0\%$ (~ 180 keV) compared
 157 to the peak energy of 9.014 MeV. The uncertainty in its centroid energy is less
 158 than ± 10 keV. After beam-energy measurement, the HPGe detector and atten-
 159 uators were moved to an off-axis position during the actual cross section ratio
 measurements.

Figure 2: (Color online) γ -ray spectrum (in red) measured with a 123% HPGe detector positioned at 0° relative to the incident photon beam of 9.0 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 (indicated by the asterisk) are clearly seen. An unfolded spectrum of FEP is shown by the blue line.

160

161 Typical pulse-height fission spectra produced by the DFC during photon
 162 irradiation are shown in Fig. 3. They were each integrated to give the total
 163 number of fissions measured in the individual chamber for a given photon en-
 164 ergy. From these numbers, the fission cross-section ratio was deduced using the

Figure 3: Fission chamber pulse-height spectra for the downstream (FC1) and upstream (FC2) chambers are shown for 13.60 MeV incident photon beam. These chambers were operated with ^{235}U and ^{238}U , respectively.

formula:

$$\left(\frac{^{235}\text{U}}{^{238}\text{U}}\right)_{CS} = \frac{(N)_{^{235}\text{U}} \times (M)_{^{235}\text{U}}}{(N)_{^{238}\text{U}} \times (M)_{^{238}\text{U}}} \times \frac{(m)_{^{238}\text{U}} \times (t)_{^{238}\text{U}}}{(m)_{^{235}\text{U}} \times (t)_{^{235}\text{U}}}, \quad (1)$$

where N is the number of fission counts, M is the molar mass, m is the areal mass density of the foil, and t is the live time of the data-acquisition system. As can be derived from Eq. 1, the main uncertainty in the $(^{235}\text{U}/^{238}\text{U})$ cross-section ratio measurement is due to the relative uncertainties in the number of counts collected from each chamber and in the mass density of the DFC foils. It should be noted that the attenuation of the photon beam from FC2 to FC1 is practically zero at these incident energies.

Our measured $^{235}\text{U}/^{238}\text{U}$ fission cross-section ratios at 27 photon energies are tabulated in Table 2 and plotted in Fig. 4 (as upside-down triangles). Also shown in Fig. 4 are the ratios computed from the ^{235}U cross-section data of Caldwell *et al.* [8] and Soldatov *et al.* [13], and the ^{238}U cross-section data of Caldwell *et al.* [8], Soldatov *et al.* [13], Ries *et al.* [12], Bergere *et al.* [22],

Figure 4: (Color online) Experimental fission cross-section ratios for $^{235}\text{U}/^{238}\text{U}$ are compared with previous measurements [7, 8, 12, 13, 22–24] and the ENDF/B-VII.1 evaluation [14].

Neto *et al.* [23], and Katz *et al.* [24]. The previous data for ^{235}U and ^{238}U were interpolated/extrapolated to correspond to the incident photon energies used in the present work. The prediction based on the evaluation ENDF/B-VII.1 [14] is also shown in Fig. 4.

As can be seen from Fig. 4, the present measured cross-section ratio values show, more or less, a smoothly increasing trend until the maximum value of 2.992 (0.096) at $E_\gamma = 12.4$ MeV. Above this energy the cross-section ratios drop, again smoothly, with a small bump at $E_\gamma \sim 15$ MeV. A similar trend is seen in the previous measurements of Caldwell *et al.* [8] except for the data at $E_\gamma \sim 12.5$ MeV (see Fig. 5). We have not observed such a fluctuation in our data in this energy range.

The ENDF/B-VII.1 evaluation [14], although on the average lower than our

Figure 5: (Color online) Experimental fission cross-section ratios for $^{235}\text{U}/^{238}\text{U}$ are compared with the previous measurement [8] and the ENDF/B-VII.1 evaluation [14]. Also shown (in the lower panel) are the differences (in %) between the ENDF evaluation and our present data. The relative difference in our data and the ENDF/B-VII.1 evaluation range from 0% to about 12% with our data constantly higher than the evaluation values.

190 data is in reasonably good agreement with the present measurements except
 191 for the region close to $E_\gamma \sim 12.0 - 13.5$ MeV, where the measured ratios are
 192 considerably higher than the evaluation.

193 4. Conclusion

194 Photofission cross-section ratio measurements were carried out for ^{235}U and
 195 ^{238}U targets in the energy range from 9.0 to 16.6 MeV. Monoenergetic photon
 196 beams were provided by the HI γ S facility at TUNL. A dual-fission ionization
 197 chamber, containing well known ^{238}U and ^{235}U monitor foils in the upstream

(FC2) and downstream (FC1) fission chambers, respectively, was employed to determine the total fission events from these actinide samples. Using this approach most systematic uncertainties cancel, in contrast to previously published experimental or evaluated ratios. Our cross-section ratio data are compared with the ratios computed from the published cross-section data and were found to be more or less in good agreement with the Caldwell *et al.* [8] data except for the region at $E_\gamma \sim 12.0$ and 15.0 MeV. The ENDF/B-VII.1 evaluation, although on the average lower than our data, the trend with energy is in reasonably good agreement with the present measurements except in the $E_\gamma \sim 12.0 - 13.5$ MeV energy region.

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Table 2: Photofission cross-section ratio data for $^{235}\text{U}/^{238}\text{U}$ obtained in the present experiment.

E_γ (MeV)	$\sigma(^{235}\text{U}(\gamma,f)/^{238}\text{U}(\gamma,f))$	(+/-) uncertainty
8.98	1.713	0.074
9.17	1.765	0.072
9.38	1.924	0.077
9.54	1.788	0.072
9.80	1.918	0.071
9.97	1.832	0.068
10.18	1.951	0.071
10.36	1.829	0.067
10.58	1.905	0.070
10.78	1.956	0.070
10.96	1.972	0.072
11.18	2.178	0.075
11.38	2.105	0.074
11.57	2.433	0.087
11.79	2.508	0.084
11.95	2.846	0.095
12.17	2.786	0.090
12.36	2.992	0.096
12.58	2.737	0.083
12.77	2.774	0.079
12.95	2.610	0.083
13.17	2.420	0.078
13.38	2.370	0.075
13.56	2.251	0.053
13.77	2.145	0.066
13.94	2.118	0.066
14.18	2.024	0.061
14.37	2.02914	0.061
14.58	1.922	0.059
14.77	2.039	0.062
14.98	1.977	0.060
15.18	1.979	0.061
15.57	1.856	0.060