Photofission and photoneutron cross sections for $^{238}$U and $^{232}$Th

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Abstract. A specific objective of the recent IAEA Coordinated Research Project on Photonenuclear Data and Photon Strength Functions (Code F41032; Duration 2016-2019) has been to measure photonuclear cross-section data where needed, for unexplored nuclei and cases of discrepant existing data. A dedicated experimental campaign has been conducted at the laser Compton-scattering γ-ray source of the NewSUBARU synchrotron radiation facility of SPring8, Japan, where photoneutron reactions for 11 nuclei from $^9$Be to $^{209}$Bi have been investigated in the Giant Dipole Resonance energy region. The measurements followed the development of a flat-efficiency moderated neutron detection array and the associated neutron-multiplicity sorting techniques. The IAEA CRP campaign has been followed at NewSUBARU by new measurements of photofission and photoneutron reactions on $^{238}$U and $^{232}$Th in the energy range of 5.87 MeV – 20.14 MeV. The neutron-multiplicity sorting of high-multiplicity fission neutron coincidence events has been performed using a dedicated energy dependent, multiple firing statistical treatment. The photoneutron ($\gamma$, xn) and photofission ($\gamma$, fxn) reactions have been discriminated by considering a Gaussian distribution of prompt-fission-neutrons (PFN) multiplicities predicted by the evaporation theory. We here provide preliminary experimental ($\gamma$, n), ($\gamma$, 2n) and ($\gamma$, F) cross sections, average energies of PFN and of photoneutrons emitted in ($\gamma$, n) and ($\gamma$, 2n) reactions, as well as the mean number of PFN per fission act. The new $^{238}$U cross sections are compared with recent statistical-model calculations performed with the EMPIRE code on existing data.

1 Introduction

The recent IAEA Coordinated Research Project (CRP) on Photonenuclear Data and Photon Strength Functions (Code F41032; Duration 2016-2019) had as main goals to update the 1999 Photonuclear Data Library [1] and to generate a Reference Database for Photon Strength Functions [2], providing support to users from the fields of nuclear structure & reactions and nuclear matter and also from a wide range of applications. Following a specific objective of the IAEA CRP of measuring photonuclear cross-section data where needed, a dedicated experimental campaign has been conducted at laser Compton-scattering (LCS) γ-ray beam line at the NewSUBARU synchrotron radiation facility [3] of SPring8, Japan. Photoneutron reactions for 11 nuclei from $^9$Be to $^{209}$Bi have been investigated in the Giant Dipole Resonance (GDR) energy region between the neutron separation energy ($S_n$) and ~40 MeV excitation energy [4, 5], providing the basis for state-of-the-art calculations of E1 moments based on nonrelativistic mean field plus QRPA [6]. The measurements followed the development of a flat-efficiency moderated neutron detection array (FED) and associated neutron-multiplicity sorting techniques. Photoneutron emission and subsequent neutron detection have been treated with an energy dependent, multiple-firing statistical method [7] originally based on a direct-neutron-multiplicity sorting with constant detection efficiency [8].

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The IAEA CRP campaign has been continued at NewSUBARU by experimental investigations of photofission and photoneutron reactions on $^{238}\text{U}$ and $^{232}\text{Th}$ in the 5.87 MeV – 20.14 MeV energy range. The neutron-multiplicity sorting has been performed by extending the energy dependent, multiple-firing statistical treatment to high-multiplicity fission neutron emission. The photoneutron ($\gamma$, xn) with $x = 1$ and 2 channels have been discriminated against the photofission ($\gamma$, f$n$) reactions by considering a Gaussian distribution of prompt-fission-neutrons (PFN) multiplicities predicted by the theory of evaporation in sequential neutron emission from excited nuclei [9].

We here provide preliminary results for photoneutron and photofission cross sections and average neutron energies and compare the new $^{238}\text{U}$ cross sections with recent statistical model calculations [10].

2 Experimental technique and methodology

2.1 Laser Compton-scattered $\gamma$-ray beams

Quasi-monochromatic $\gamma$-ray beams are produced at NewSUBARU by head-on collisions between laser photons and relativistic electrons circulating in the storage ring. A system of two collimators is used to separate the high energy, backscattered photons and produce incident beam with typical energy resolution of 1 - 3 % in FWHM. The absolute energy of the $\gamma$-ray beams has been calibrated with an accuracy of $10^{-5}$ [11] by using low-energy LCS $\gamma$-ray beams produced with a grating-fixed CO$_2$.

The present experiment made use of $\gamma$-ray beams with maximum energies between 5.87 MeV and 20.14 MeV produced with a 1064 nm wavelength laser in combination with electron beam energies between 575 and 1072 MeV. The photon beam energy spectra were monitored with a $3.5''$ × $4''$ LaBr$_3$(Ce) detector. The experimental spectra for each irradiation energy have been reproduced using the dedicated e11LaBr LCS $\gamma$-ray source simulation code [12–14] implemented using the GEANT4 package [15]. The incident photon flux was monitored with a large volume $8''$ × $12''$ NaI(Tl) detector. The multiple-photon spectra characteristic to pulsed $\gamma$-ray beams of ~ tens of ns pulse width were processed with the Poisson fitting method [16].

2.2 Target materials and neutron detection

The LCS $\gamma$-ray beams irradiated nuclear fuel materials of 8.62 g ThO$_2$ and 4.06 g $\text{U}_3\text{O}_8$ shielded in pure-aluminum cylindrical containers of 8 mm inner diameters. From the natural abundances of $^{232}\text{Th}$, $^{235}\text{U}$, $^{238}\text{U}$, and $^{233}\text{U}$, we deduced the areal density of $^{232}\text{Th}$ and $^{238}\text{U}$ to be 17.15 and 8.08 g/cm$^2$, respectively. The actinide targets were placed in the center of a moderated high-and-flat efficiency neutron detection system [8] consisting of three concentric rings of 4, 9, and 18 $^{238}\text{U}$ targets embedded in a polyethylene moderator at 5.5, 13.0 and 16.0 cm from the $\gamma$-ray beam axis, respectively. Fig. 1(a) shows the total simulated neutron detection efficiency along with the inner ring one and the sum of the two outer rings. The simulations have been performed considering neutron evaporation spectra described by the Weisskopf-Ewing function [17] (black lines), which are typical for photoneutron emission, as well as Maxwell type neutron spectra characteristic for PFN. There is good agreement between the two curves, demonstrating a robust flatness of the neutron detection efficiency in a wide average neutron energy range between thermal region and 3 - 7 MeV. The ring-ratio curve employed for determining the average neutron energies is shown in Fig. 1(b) for the two types of neutron spectra. The two functions are in good agreement for average neutron energies up to 1 MeV and tend to disagree with the increase in the neutron energy.

2.3 Neutron multiplicity sorting

In order to measure multi-neutron coincidences, the LCS $\gamma$-ray beams was produced in a low frequency 1 kHz pulsed mode, where the 1 ms time interval between consecutive pulses was set based on the neutron die-away time inside the FED [4, 7, 8]. Neutrons recorded during the 1 ms events originate from both photoneutron and photofission reactions, depending on the incident photon energy. For example, for an incident photon energy between the 1-neutron separation threshold and the 2-neutrons separation threshold, the recorded neutron originate from the $\gamma$, n reaction and from the $\gamma$, 2n reaction. In low reaction rate conditions, we can disregard events in which multiple reactions are induced in the target by the same photon pulse. In such a single-firing approximation, neutrons recorded in higher than 1 coincidences have been emitted in photofission reactions only. However, as the mean number of photons per pulse is ~10 and the photon multiplicity is Poisson distributed, multiple-firing events
Figure 2. The present preliminary (a) (γ, n), (b) (γ, 2n) and (c) (γ, F) cross sections for $^{238}$U and $^{232}$Th compared with Livermore [18] (red), Saclay [21] (blue), bremsstrahlung [22–24], neutron capture γ-rays [25] and tagged photons [26] existing data. Note that all the $^{232}$Th(γ, F) cross sections are here scaled by a factor of 3 for better visualization in connection with the $^{238}$U(γ, F) results. The $^{238}$U cross section are compared with recent statistical model calculations [10] (black line) performed on the existing data. The average numbers of PFN per fission act are compared in (d) with Livermore [18] (red), HI$^S$ [27] (green) and bremsstrahlung data [28] (blue) and with Los Alamos model theoretical predictions [19] (lines). Present experimental average energies of neutrons emitted in (γ, n), (γ, 2n) and (γ, F) reactions are shown in (e).
can of course be reduced by limiting the incident photon flux, but they can not be completely eliminated. Following probabilities given by partial cross section, target material and number of incident photons per pulse, all combinations of energetically available reactions can be induced by each beam pulse. Thus, we used the energy dependent, multiple-firing statistical method [7] originally developed for photoneutron reactions only, where we have additionally implemented the photofission reaction channels.

The method models the multiple-firing of all available combinations of photoneutron ($\gamma$, $xn$) and photofission ($\gamma$, $f\alpha x$) reactions. Here the total fission cross section $\sigma(\gamma,F)$ is given by the sum cross section for the fission reaction with emission of $x$ neutrons, where $x$ takes values from 0 to $\sim$9. In order to limit the number of independent variables and to account for the limited statistics in registering high-multiplicity neutron events, we considered a Gaussian distribution of PFN multiplicities predicted by the theory of evaporation in sequential neutron emission from excited fission fragments [9]. Following the predictions of J. Terrell, the ($\gamma$, $f\alpha x$) channels are described by the PFN Gaussian distribution with three independent parameters: the total ($\gamma,F$) cross section, the mean number of PFN per fission act and a width parameter.

## 3 Preliminary results

The present preliminary results on the $^{238}$U and $^{232}$Th photoneutron and photofission in the GDR energy region are shown in Fig. 2. The ($\gamma$, $n$) and ($\gamma$, $2n$) reactions for $^{238}$U are in good agreement with the Saclay results [21]. The $^{232}$Th($\gamma$, $n$) cross section is systematically lower than both the Saclay and the Livermore results [18]. The $^{232}$Th($\gamma$, $2n$) cross section is in agreement with the Saclay results, but doesn’t confirm the higher energy hump present in the existing data. Both the $^{238}$U and $^{232}$Th photofission cross sections are generally in better agreement with the Saclay results, and systematically lower than the Livermore ones, except for the peak region of the higher energy hump, where they are in good agreement with the Livermore results. The recent $^{238}$U statistical model calculations of Ref. [10] overestimate the present photofission cross sections at energies higher than 18 MeV while underestimating the peak cross section on the second fission hump. Fig. 2(d) shows a good agreement between the present average numbers of PFN per fission act and previous experimental data. The average energies of ($\gamma$, $n$), ($\gamma$, $2n$) neutrons and PFN for $^{238}$U and $^{232}$Th are shown in Fig. 2(e). Both the $^{238}$U average PFN numbers and average PFN energies are well reproduced by theoretical calculations performed with the Los Alamos model using the most probable fragmentation approach with input parameters taken from systematics [19, 20].

## 4 Summary

New $^{238}$U and $^{232}$Th photoneutron and photofission cross section measurements in the GDR energy region have been performed at the NewSUBARU synchrotron facility using LCS $\gamma$-rays and a novel high-and-flat efficiency neutron detector. Neutron multiplicity sorting was performed using a statistical treatment of multiple-firing neutron coincidence events. The photoneutron and photofission channels were discriminated considering a Gaussian distribution of PFN multiplicities predicted by the evaporation theory. Present preliminary results are compared with existing experimental data and theoretical predictions.


**References**