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The photofission cross-section of ^{238}U was measured at sub-barrier energies as a function of the γ-ray energy using, for the first time, a monochromatic, high-brilliance, Compton-backscattered γ-ray beam. The experiment was performed at the High Intensity γ-ray Source (HIγS) facility at beam energies between E_γ=4.7 MeV and 6.0 MeV and with $\sim$ 3% energy resolution. Indications of transmission resonances have been observed at γ-ray beam energies of E_γ=5.1 MeV and 5.6 MeV with moderate amplitudes. The triple-humped fission barrier parameters of ^{238}U have been determined by fitting EMPIRE-3.1 nuclear reaction code calculations to the experimental photofission cross section.

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Photofission measurements enable selective investigation of extremely deformed nuclear states in the light actinides and can be utilized to better understand the landscape of the multiple-humped potential energy surface (PES) in these nuclei. The selectivity of these measurements originates from the low and reasonably well-defined amount of angular momentum transferred during the photoabsorption process. The present study is designed to investigate the PES of ^{238}U through observation of transmission resonances in the prompt photofission cross section. A transmission resonance appears when directly-populated excited states in the first potential minimum overlap energetically with states either in the superdeformed (SD) 2^{nd} or hyperdeformed (HD) 3^{rd} potential minima [1, 2]. The fission channel can thus be regarded as a tunneling process through the multiple-humped fission barrier as the gateway states in the first minimum decay through states in the other minima of the PES. So far, transmission resonances have been studied primarily in light-particle-induced nuclear reactions through charged-particle, conversion-electron or γ-ray spectroscopy. These studies do not benefit from the same selectivity found in photonuclear excitation and consequently they are complicated by statistical population of the states in the 2^{nd} and 3^{rd} minima with a probability of $10^{-4} - 10^{-5}$. This statistical population leads to a typical isomeric fission rate from the ground-state decay of the shape isomer in the 2^{nd} minimum of $\sim$1/sec. These measurements have also suffered from dominating prompt-fission background.

Until now, sub-barrier photofission experiments have been performed only with bremsstrahlung photons and have determined only the integrated fission yield. In these experiments, the fission cross section is convolved with the spectral intensity of the γ-ray beam, resulting in a typical effective γ-ray bandwidth $\Delta E/E$ between 4×10^{-2} and 6×10^{-2}. These experiments observe a plateau, referred to as the "isomeric shelf", in the fission cross section between E_γ=3.5 MeV and 4.5 MeV, resulting from competition between prompt and delayed photofission [3, 4]. Higher-resolution studies can be performed at tagged-photon facilities, though only with marginal statistics, due to the limited beam intensities realizable through tagging [5]. This beam intensity cannot be significantly improved beyond $\sim 10^4\ \gamma/(\mathrm{keV}\cdot\mathrm{s})$, since it is determined by the random coincidence contribution in the electron-tagging process. Thus, high statistics photofission experiments in the deep sub-barrier energy region, where cross sections are typically as low as σ=1 nb-10 μb, cannot be performed with tagged-photon beams. The relatively recent development of inverse-Compton scattering γ-ray sources, capable of producing tunable, high-flux, quasi-monoenergetic γ-ray beams by Compton-backscattering of eV-range photons off a relativistic electron beam, offers an opportunity to overcome previous limitations. The present study was carried out at such a facility: the High Intensity γ-ray Source (HIγS) located at TUNL. It should be emphasized that a measurement of the photofission cross section in the deep sub-barrier energy region will be a crucial step towards a reliable characterization of the PES, including unambiguous determination of the double- or triple-humped nature of the surface and precise evaluation of the barrier parameters. Next-generation Compton-backscattering γ-ray sources, such as MEGa-ray (Lawrence Livermore National Laboratory, California, US) [6] and ELI-NP (Bucharest, Romania) [7], are anticipated to provide beams with spectral fluxes of $\sim 10^6\gamma/(\mathrm{eV}\cdot\mathrm{s})$ and energy resolution of $\Delta E \approx$ 1keV, far superior to those currently available at HIγS. The capabilities of these next-generation sources allow one to aim at an identification of sub-barrier transmission resonances in the fission decay channel with integrated cross sections down to $\Gamma\sigma \approx 0.1$ eV $\cdot$ b, whereas the present study is only sensitive to resonances with $\Gamma\sigma \approx 10$ eV $\cdot$ b. The narrow energy bandwidth expected for the new γ-ray beam facilities will also allow for a significant reduction of the presently dominant background from non-resonant processes. Thus, next-generation γ-ray sources are expected to allow preferential population and identification of vibrational resonances in the photofission cross section and ultimately to enable observation of the fine structure in the isomeric shelf. This may open the perspective towards a new era of photofission studies.

Sub-barrier photofission of ^{238}U so far has only been studied with intense bremsstrahlung, however, without being able to resolve any resonances [8]. A previous ^{236}U(p,t) measurement showed pronounced resonance structures at excitation energies of $E^* = 5.6 - 5.8$ MeV and at $E^* = 5.15$ MeV, as well as a weaker resonance at $E^* = 4.9$ MeV [9]. A whole sequence of further transmission resonances at lower energies is expected to explain the isomeric shelf [4], but such resonances have not yet been observed. Furthermore, it has been found experimentally in several measurements on ^{234}U [10], on ^{236}U [11] and most recently on ^{232}U [12] in agreement with older theoretical predictions [13], that for the uranium isotopes the HD 3^{rd} potential minimum is in fact as deep as the SD 2^{nd} minimum. According to this experimental systematics, the existence of a HD 3^{rd} minimum is also predicted for ^{238}U, however, it has not yet been supported experimentally. On the other hand, recent calculations using a macroscopic-microscopic model do not predict the existence of a deep 3^{rd} minimum for the even-even uranium isotopes [14, 15]. This puzzle was more recently addressed within a self-consistent theoretical approach, where the conditions for the existence of HD potential minima were studied [16].

The aim of the present study was to measure the ^{238}U(γ,f) cross-section at deep sub-barrier energies and to search for transmission resonances. The experiment was performed at the HIγS facility with its Compton-backscattered γ-ray beam, having a bandwidth of ΔE=150-200 keV and a spectral flux of about $10^2\gamma/(\text{eV}\cdot\text{s})$. An array of parallel plate avalanche counters, consisting of 23 electrolytically-deposited $^{238}UO_2$ (2 mg/cm^2) targets [17], was used to measure the photofission cross section. Both fission fragments were detected in coincidence to suppress the α-particle background to an extremely low level, which is required by the particularly low counting rates (typically 0.1-1 Hz at E_γ=5 MeV). The total efficiency of the array was estimated to be 70% based on Ref. [17].

The present experimental photofission cross section of ^{238}U as a function of the γ-ray energy is shown in Fig. 1a, along with the experimental data of Ref. [8]. Near the top of the barrier the two data sets are in a good agreement. The present data are extended by about an order of magnitude in cross section to the deep sub-barrier region down to E_γ=4.7 MeV. A clear transmission resonance has been observed at E_γ=5.6 MeV, which is consistent with the observation of Ref. [8]. A slight deviation from the exponential slope of the cross section indicates the existence of a resonance at E_γ=5.1 MeV, however, with only a limited resonance signal contrast due to the moderate bandwidth of the γ-ray beam.

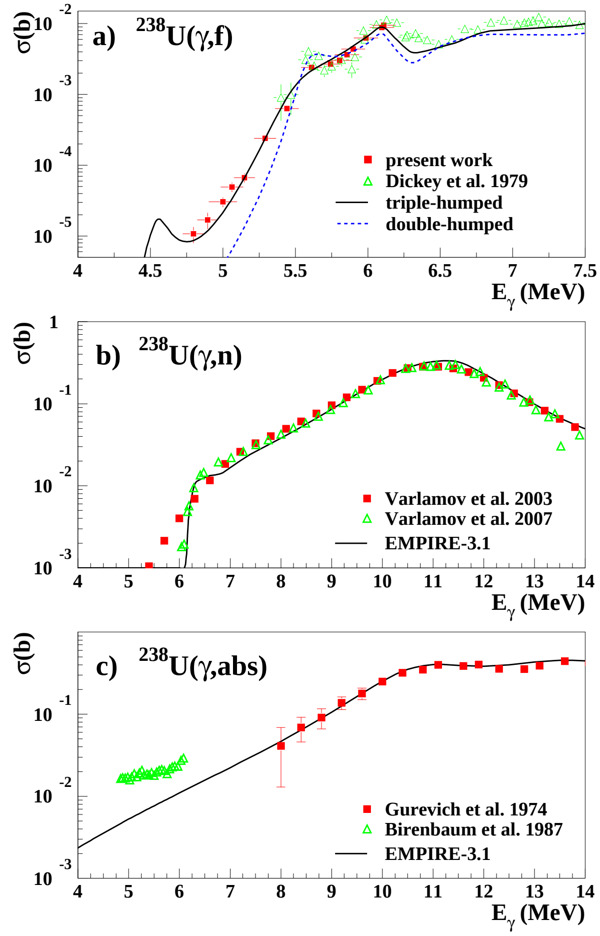

FIG. 1. (Color online) a) The measured photofission cross-section of ^{238}U in the γ-ray energy range of E_γ=4.7-6.0 MeV. The result of the present experiment and the experimental data of Ref. [8] are indicated by full squares and open triangles, respectively. b) Experimental ^{238}U(γ,n) cross-sections of Refs. [29] and [30] are indicated by full squares and open triangles, respectively. c) Total photo-absorption cross-section of ^{238}U as a function of the γ-ray energy. The experimental data from Ref. [18] and Ref. [19] are indicated by full squares and open triangles, respectively. In all panels, the cross sections calculated using EMPIRE-3.1, as discussed in the text, are shown as black lines; the calculations in panel b) and c) assume a triple-humped barrier structure, however, without influencing the resulting cross sections.

For the theoretical evaluation of the present ^{238}U photofission experimental data, we performed nuclear reaction code calculations using the EMPIRE-3.1 code [20].

TABLE I. Double-humped fission barrier parameters of ^{238}U (in MeV) used in the calculations. The resulting photofission cross section is indicated in Figure 1a by the dashed line.

E_A	E_{II}	E_B	$\hbar\omega_A$	$\hbar\omega_{II}$	$\hbar\omega_B$
6.3±0.2	2.0±0.2	5.65±0.20	1.1±0.1	1.0±0.1	0.6±0.1

Within the code, the fission transmission coefficients are calculated using the Hill-Wheeler formalism [21], followed by Hauser-Feshbach statistical model calculations [22], allowing the fission channel to compete with emission of particles and photons. The triple-humped fission barrier parameters of ^{238}U were extracted by tuning the inputs to these calculations and comparing the resulting predictions of the photofission cross section to the experimental data.

The reliability of the code was tested and the relevant model parameters were adjusted using calculations of the total photo-absorption cross section $\sigma_{\gamma,\text{abs}}$ and experimental (γ,n) cross section data. First, $\sigma_{\gamma,\text{abs}}$ had to be determined and checked against existing experimental data. In the present evaluation, the modified Lorentzian parameterization (MLO) was chosen for the γ-ray strength function. Although the experimental data of Ref. [18] are quite well reproduced (solid line in Fig. 1c), the experimental results of Ref. [19] are underestimated at lower energies. Yet, we have not attempted to tune the MLO parameters to reproduce the experimental data. The parameterization used is based on a global fit of experimental data over a wide range of isotopes and excitation energies. Attempts to reproduce this dataset would have a drastic impact on the competing reaction channels, leading (especially for fission) to unphysical parameters. The photo-absorption cross sections of Ref. [19] were inferred from the measured energy-averaged, angle-integrated photon elastic-scattering cross sections $\sigma_{\gamma\gamma}$, employing a complex analysis technique described in details in Ref. [23]. In such an analysis, the measured values are renormalized by an energy-dependent factor to obtain the corrected photo-absorption cross section $\sigma_{\gamma,\text{abs}}$. Our calculated values are located between the measured $\sigma_{\gamma\gamma}$ and the corrected $\sigma_{\gamma,\text{abs}}$ cross sections, perhaps indicating systematic uncertainties in the aforementioned analysis.

The transmission coefficients for the particle emission were determined using the global optical parameter set of Ref. [24]. The level density parameters were taken from the enhanced generalized super-fluid model [25], adjusted to fit the discrete level scheme of ^{238}U. Those were taken from the most recent reference input parameter library (RIPL3). In the code, the optical model for fission [26–28] is applied to calculate the fission transmission coefficients. For comparison, both triple- and double-humped fission barriers were used in the calculations.

The parameters of the double-humped fission barrier were taken from the RIPL3 library and were slightly adjusted to achieve a better description of the present data. In Figure 1a, the dashed line shows the calculated (γ,f) cross-section using the parameters listed in Table I. The triple-humped barrier parameters were adjusted to best describe the experimental photofission and (γ,n) cross sections over the entire energy range. In Figure 1a, the solid line represents the best description with the parameters of Table II used in the calculation. The calculated (γ,n) cross-sections are shown as the solid line in Fig. 1b together with the available experimental data [29, 30]. The calculated and the experimental values are in a fair agreement. The uncertainties of the barrier parameters were estimated to be 200 keV for the barrier heights and 100 keV for the curvature parameters.

The present model is capable of reproducing the sub-barrier fission resonances empirically, while at higher excitation energies it naturally provides the same results for the fission barrier penetration as the classical models. Since photofission is considered to occur only through the excitation of the giant dipole resonance and ^{238}U is an even-even nucleus, one can assume that only states with J^π=1$^-$ contribute to γ-ray induced fission. This is true at low energies, but when (γ,γ'f) processes become significant, states with different quantum numbers get involved. The discrete transition states are rotational levels characterized by a given set of quantum numbers (angular momentum J, parity π and angular momentum projection on the nuclear symmetry axis K) built on vibrational or non-collective band-heads. For an even-even nucleus the band-heads with K^π=0$^+$, 2$^+$, 0$^-$ and 1$^-$ are usually considered. In this case, J^π=1$^-$ rotational levels belong both to K^π=0$^-$ and 1$^-$. In Table III, the energies of the discrete transition band-heads (ϵ_A, ϵ_B and ϵ_C) are listed that were used in the calculations.

The damping of the vibrational states within the strongly deformed potential wells has been simulated by introducing a negative imaginary potential in the corresponding deformation ranges. These potentials are assumed to be quadratic functions of the deformation, like for the real part, and their strengths are chosen to fit the width of the resonances in the sub-barrier fission cross section and to be consistent with physical values for the transmission coefficients at higher energies. More details about the fission coefficients calculation for multi-humped barriers can be found in Ref. [28].

The experimental data of the present experiment could be reproduced dramatically better with a calculation assuming a triple-humped fission barrier than with a double-humped one. Because of the high damping of the SD vibrational states, the corresponding resonances are smeared out. Thus, the resonance at E_γ=5.55 MeV was attributed to the HD well. Due to the energy spacing of the vibrational states in the HD oscillator well, which is determined by the curvature parameter of the 3rd potential minimum ($\hbar\omega_{III}$=1.0 MeV), a further resonance should appear at E_γ=4.55 MeV. Experimental evidence for the existence of such a resonance would fully confirm our present theoretical interpretation. It is also evident that the existing (γ,f) and (γ,n) experimental data suffer

TABLE II. Triple-humped fission barrier parameters of ^{238}U (all in MeV) used in the calculation, represented by the solid line in Figure 1a.

E_A	E_{II}	E_B	E_{III}	E_C	$\hbar\omega_A$	$\hbar\omega_{II}$	$\hbar\omega_B$	$\hbar\omega_{III}$	$\hbar\omega_C$
4.3±0.2	2.05±0.20	5.6±0.2	3.6±0.2	5.6±0.2	0.4±0.1	1.0±0.1	0.7±0.1	1.0±0.1	0.7±0.1

TABLE III. The energies of the transition band-heads (in MeV) used for ^{238}U in the calculations.

K^π	ϵ_A	ϵ_B	ϵ_C
0^+	0.0	0.0	0.0
2^+	1.09	0.10	0.10
0^-	0.10	1.20	1.20
1^-	0.90	0.84	0.94

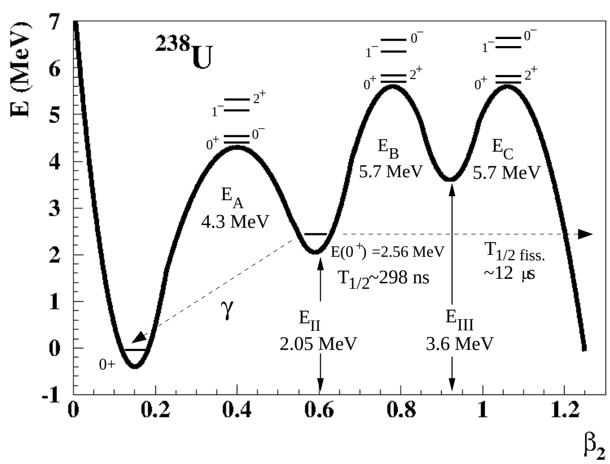

FIG. 2. The triple-humped fission barrier of ^{238}U as determined in the present study, using the parameters listed in Table II. The half-life of the isomeric ground state at E=2.56 MeV and the partial isomeric fission half-life are also indicated.

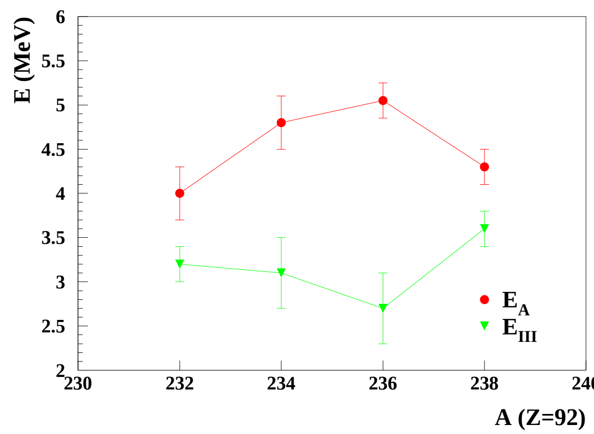

FIG. 3. (Color online) The height of the inner barrier $E_{\rm A}$ and the depth of the third minimum $E_{\rm III}$ for even-even uranium isotopes, shown as red circles and green triangles, respectively. The experimental data for ^{232}U, ^{234}U and ^{236}U were taken from Refs.[10–12].

from large uncertainties. It would be highly important to improve the γ-ray beam energy bandwidth and the energy density of data points (requiring a higher γ-ray beam intensity), in order to explore a full set of deep sub-barrier fission resonances.

The present results on the fission barrier parameters of ^{238}U supplement the previous findings on the systematics of the barrier parameters of the uranium isotopes [12]. Fig. 3 shows the present results on ^{238}U together with previous experimental results on ^{232}U [12], ^{234}U [10] and ^{236}U [11]. A reversal of the trends followed by the lighter uranium isotopes for the height of the inner barrier E_A and the depth of the third minimum (expressed by E_{III}), respectively, as a function of the neutron number, is observed. For ^{238}U, the data suggests a decreasing barrier height E_A and a decreased depth of the third minimum. Moreover, the particularly low values of the curvature parameters derived from the present data, especially the one for the inner barrier ($\hbar\omega_A$=0.4 MeV), may suggest a need for reconsideration of the well-accepted approximation of the fission barrier with a harmonic oscillator potential curve. An anharmonic, "tower-like" potential, originally suggested by Bowman *et al.* decades ago [31], would better approximate the potential landscape determined from the current data.

In summary, we measured the photofission cross-section of ^{238}U in the γ-ray energy region of E=4.7-6.0 MeV with the monochromatic, high-brilliance, Compton-backscattered γ-ray beam of the HIγS facility. With the significantly higher intensity of the beam, when comparing to a tagged-photon facility, the cross-section could be measured at deep sub-barrier energies. EMPIRE-3.1 reaction code calculations were performed to extract the fission barrier parameters of ^{238}U. Our present results on the fission barrier of ^{238}U support a deep 3^{rd} minimum with $E_{\rm III}$=3.6 MeV, a low inner barrier height $E_{\rm A}$=4.3 MeV and outer barrier heights of $E_{\rm B}$=5.7 MeV and $E_{\rm C}$=5.7 MeV. Though in line with the extensive body of experimental evidence for deep third potential minima in uranium isotopes acquired over the last 15 years, this result is in disagreement with recent calculations of Ref. [14], a puzzle that still needs to be resolved. Indications of predicted resonance structures have also been observed, however, with moderate amplitudes. The results indicate the need for further investigations at lower γ-ray energies and using smaller-bandwidth, higher-intensity γ-ray beams. ELI-NP, MEGa-ray, and other next-generation γ-ray sources will enable such measurements.

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