Fissility of Actinide Nuclei by 60-130 MeV Photons

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Nuclear fissilities obtained from recent photofission reaction cross section measurements carried out at Saskatchewan Accelerator Laboratory (Saskatoon, Canada) in the energy range 60-130 MeV for ^{232}Th, ^{233}U, ^{235}U, ^{238}U, and ^{237}Np nuclei have been analysed in a systematic way. To this aim, a semiempirical approach has been developed based on the quasi-deuteron nuclear photoabsorption model followed by the process of competition between neutron evaporation and fission for the excited nucleus. The study reproduces satisfactorily well the increasing trend of nuclear fissility with parameter Z^2/A.

Keywords: Photofission reaction; Nuclear fissility

I. INTRODUCTION

The availability of high-quality monochromatic (or quasi-monochromatic) photon beams generated by different techniques (tagged photons [1,6], and Compton backscattered photons [7,8]) has opened new possibilities for experimental investigation of photonuclear reactions in the energy range $0.02 \lesssim E_\gamma \lesssim 4.0$ GeV. In particular, the development of high-performance parallel-plate avalanche detectors (PPAD) for fission fragments [2, 5, 9, 10] has allowed researchers to obtain photofission cross section data for actinide target nuclei (Th, U, Np) within $\sim$ 5% of uncertainty, and for pre-actinide (Pb, Bi) within $\sim$ 12%, covering a large incident photon energy-range from about $\sim$30 MeV on.

The experimental data from such photonuclear reactions (cross section measurements and fission probability) have been generally interpreted on the basis of a current nuclear photoreaction model for intermediate- ($30 \lesssim E_\gamma \lesssim 140$ MeV) and high- (E$\gtrsim$ 140 MeV) energy photons [2, 11-14]. In brief, during the first reaction step, the incoming photon interacts with a neutron-proton pair ($E_\gamma \lesssim 500$ MeV) and/or individual nucleons ($E_\gamma \gtrsim 140$ MeV), where pions and baryon resonances, and recoiling nucleons initiate a rapid ($\sim 10^{-23}$s) intranuclear cascade process which produces a residual, excited nucleus. After thermodynamic equilibrium was reached, a second, slow process takes place where fission may occur as a result of a mechanism of competition between particle evaporation (neutron, proton, deuteron, triton and alpha particle) and fission experienced by the excited cascade residual.

Nuclear fissility, f, is the quantity which represents the total fission probability of a target nucleus (Z,A) at an incident photon energy, E_γ, and it is defined as the ratio of photofission cross section,σ_f, to photoabsorption cross section, σ_a^T, both quantities being measured at the same energy,i.e.,

$$f(Z,A,E_\gamma) = \frac{\sigma_f(Z,A,E_\gamma)}{\sigma_a^T(Z,A,E_\gamma)} \ . \qquad (1)$$

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Five years ago, a group of researchers reported on results of high-quality photofission cross section measurements for ^{232}Th, ^{233}U, ^{235}U, ^{238}U and ^{237}Np nuclei carried out at Saskatchewan Accelerator Laboratory (Saskatoon, Canada) [5], covering the energy range of 68-264 MeV. The aim of the present work is to perform an analysis of the nuclear fissility data obtained from such measurements in a semiempirical and systematic way by making use of the two-step photoreaction model [12, 14]. The study will cover the range 60-130 MeV, where nuclear photoabsorption has been described by the interaction of the incident photon with neutron-proton pairs (quasi-deuterons), as described for the first time by Levinger [15,16].

II. QUASI-DEUTERON NUCLEAR PHOTOABSORPTION

In the incident photon energy range here considered, the incoming photon is assumed to be absorbed by a neutron-proton pair as described by Levinger's original and modified quasi-deuteron model [15, 16]. Accordingly, the total nuclear photoabsorption cross section has been evaluated by

$$\sigma_a^T = LZ\left(1 - \frac{Z}{A}\right)\ \sigma_d(E_\gamma) f_B(E_\gamma) \ . \qquad (2)$$

In this expression, $\sigma_d(E_\gamma)$ is the total photodisintegration cross section of the free deuteron, the values of which have been taken from the data analysis by Rossi *et al.* [17] (Fig.1-a). L is the so-called Levinger's constant, and it represents the relative probability of the two nucleons being near each other inside the nucleus as compared to a free deuteron. An evaluation of Levinger's constant for nuclei throughout the Periodic Table by Tavares and Terranova [18] gives $L = 6.5$ for actinide nuclei. Finally, $f_B(E_\gamma)$ is the Pauli block function, which can be established in a semiempirical way, if we observe that, among the actinide nuclei which have been investigated , ^{237}Np is the one which has a low fission barrier (4.63 MeV, therefore, a good chance for fission) and, at the same time, the greater neutron separation energy (6.58 MeV). Thus, we can assume that the photofission cross section of ^{237}Np represents its total photoabsorption cross section, i.e.,

$$\sigma_a^T(E_\gamma)_{Np} = \sigma_f(E_\gamma)_{Np} \ . \qquad (3)$$

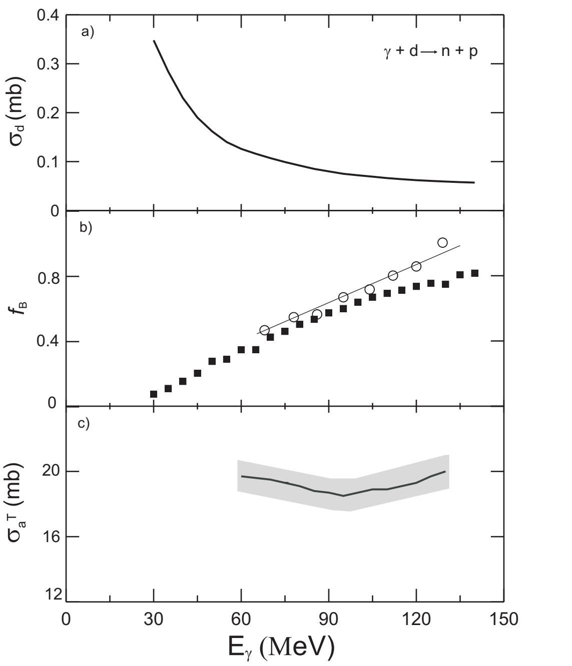

FIG. 1: a) Total photodisintegration cross section for the "free" deuteron, σ_d, versus energy[17]. b) Pauli blocking function, $f_B(E_\gamma)$: open circles represent semiempirical values obtained as described in the text; the straight line is the trend of f_B in the range 60-130 MeV (Eq.(4)); full squares are results from a Monte Carlo calculation [19]; c) total nuclear photoabsorption cross section for actinide nuclei as given by Eq.(15); the shaded area indicates the uncertainties (estimated to ± 5%).

This is the same as to assume fissility constant and equal to unity for ^{237}Np. Hence, from Eqs. (2,3), and by making use of the experimental data of σ_f for ^{237}Np (Table 1, 10^{th} column), the Pauli block function in the incident photon energy range 60-130 MeV, can be expressed as

$$f_B(E_\gamma) = KE_\gamma \ , \ K = 0.00718 \ \text{MeV}^{-1} \ . \qquad (4)$$

Table 1: Photofission cross section[a] (σ_f) and nuclear fissility[b] ($\bar{f}$) for actinide nuclei in the quasi-deuteron photoabsorption energy region.

E_γ	^{232}Th		^{233}U		^{235}U		^{238}U		^{237}Np	
(MeV)	σ_f	f	σ_f	f	σ_f	f	σ_f	f	σ_f	f
68	8.8	0.45	15.9	0.82	16.7	0.86	15.6	0.80	19.0	0.98
78	8.8	0.46	15.3	0.80	16.3	0.85	14.4	0.76	19.0	~ 1
86	8.8	0.46	15.9	0.84	16.7	0.88	14.4	0.76	17.7	0.93
95	9.5	0.51	16.4	0.88	16.9	0.91	14.8	0.80	18.5	~ 1
104	9.2	0.49	16.4	0.87	18.8	0.99	15.2	0.80	18.5	0.98
112	9.5	0.50	16.9	0.89	17.3	0.91	15.0	0.79	19.3	~ 1
120	10.6	0.55	17.4	0.90	18.8	0.97	17.0	0.88	19.6	~ 1
129	10.8	0.54	19.6	0.98	19.4	0.97	18.0	0.90	22.0	~ 1

[a] Cross sections (expressed in mb) are experimental data obtained at Saskatchewan Accelerator Laboratory (Saskatoon, Canada) by Sanabria *et al.* [5];

[b] Nuclear fissilities are deduced values from $f = \sigma_f/\sigma_a^T$, where σ_a^T is total nuclear photoabsorption cross section as explained in the text (section 2).

This result is shown in Fig.1-b (straight line) together with the semiempirical values of f_B (circles). For a comparison, the results of a Monte Carlo calculation (full squares in Fig.1-b) obtained by de Pina *et al.* [19] is also shown. Except for 129 MeV, the above results differ from each other by 8% on the average. Finally, if one adopts for $f_B(E_\gamma)$ the result expressed in (4), the total cross section for nuclear photoabsorption in the range 60-130 MeV for actinide nuclei can be calculated as

$$\sigma_a^T(E_\gamma) = CE_\gamma\sigma_d(E_\gamma) \ , \ \ C = LZ\left(1-\frac{Z}{A}\right)K \ . \qquad (5)$$

Since the constant C does not vary significantly for the actinide nuclei ($\bar{C} = 2.60 \pm 0.03$ MeV^{-1}), we can assume the same curve $\sigma_a^T(E_\gamma)$ as depicted in Fig.1-c valid for all actinide here analysed. Therefore, the nuclear fissilities, as defined in (1), can be obtained in this way, and results are listed in Table 1.

III. NUCLEAR EXCITATION AND FISSION PROBABILITY

As a consequence of the quasi-deuteron primary photointeraction, $\gamma + (\mathrm{n+p}) \rightarrow \mathrm{n}^* + \mathrm{p}^*$, different residual nuclei can be formed according to one of the three possible modes: 1) The neutron escapes from the nucleus, at the same time that the proton remains retained; 2) the proton escapes and the neutron is retained; 3) both neutron and proton are retained inside the nucleus. A fourth possibility there exists clearly of obtaining residual nuclei, namely, escaping of both the neutron and proton simultaneously from the nucleus. In this case, however, no excitation energy is left to the residual nucleus, with the consequence of null chance for fission from this residual [14]. Let τ_n and τ_p denote, respectively, the probabilities of escaping for neutron and proton from the nucleus (without suffering for any secondary interaction), i.e., the nuclear transparencies to neutron (τ_n) and proton (τ_p). Complete retention (or reabsorption, or non-escaping) of nucleons means $\tau_p = \tau_n = 0$. The probabilities for neutron and proton retention are, respectively, $(1-\tau_n)$ and $(1-\tau_p)$. Therefore, the probabilities of residual nuclei formation (excited or not) following one of the four routes mentioned above are given by $p_1 = \tau_n(1-\tau_p)$, $p_2 = \tau_p(1-\tau_n)$, $p_3 = (1-\tau_p)(1-\tau_n)$ and $p_4 = \tau_p\tau_n$.

Nuclear transparencies depend essentially upon neutron and proton kinetic energies in their final states [12,14]. For photons in the energy range 60-130 MeV interacting with actinides, it is found that for almost $\sim$ 80% of cases excited residual nuclei are formed ($\overline{E}_1^* \approx \overline{E}_2^* \approx E_\gamma/2$, hence, highly fissionable residuals), and the third mode of formation is the predominant one.

The total fission probability, i.e, the fissility of the target nucleus is thus given by

$$f(E_\gamma) = \sum_{i=1}^{4} p_i P_{fi}(E_i^*) \ , \qquad \sum_{i=1}^{4} p_i = 1 \ , \qquad (6)$$

where P_{f_i} represents the fission probability for the residual nucleus formed according to the mode i of formation of residuals. Since $E_4^* = 0$, it follows that $P_{f_4} = 0$, and, therefore

$$f(E_\gamma) = p_1 P_{f_1}(Z, A-1, \overline{E}_1^*) + p_2 P_{f_2}(Z-1, A-1, \overline{E}_2^*) + p_3 P_{f_3}(Z, A, \overline{E}_3^*) \ . \quad (7)$$

The different residual nuclei formed are actinide (like the target ones) and, in this case, the fission barriers are, in general, lower than the respective neutron separation energies ($B_{f_0} < S_n$). Therefore, we can say that the fission probability of residual nuclei formed following the formation modes $i = 1, 2$, and 3 is given approximately by their first-chance fission probability, f_{1_i}, i.e,

$$P_{f_1} \approx f_{1_1}(Z, A-1, \overline{E}_1^*); \ P_{f_2} \approx f_{1_2}(Z-1, A-1, \overline{E}_1^*); \ P_{f_3} \approx f_{1_3}(Z, A, E_\gamma) \ . \quad (8)$$

On the other hand, the first-chance fission probabilities, $f_{1_i}(i = 1, 2, 3)$, should not differ significantly for adjacent actinide nuclei, because all of them do exhibit low fission barrier values ($B_f \lesssim 5$ MeV), at the same time that f_{1_i} should not vary significantly with energy too (see Table 1). Calculations of nuclear transparencies for actinides [14] show that $(0.20 \lesssim \tau_n \lesssim 0.50)$ and $0.10 \lesssim \tau_p \lesssim 0.15$ for neutron and proton energies considered here, in such a way that the mode $i = 3$ is the predominant one, at the same time that $p_4 \lesssim 8\%$. Thus, to a good approximation, we can write

$$f(Z, A, E_\gamma) \approx f_1(Z, A, E^* = E_\gamma) \ , \ \ 60 \lesssim E_\gamma \lesssim 130 \text{ MeV} \ . \quad (9)$$

This result means that, for actinide nuclei, nuclear photofissility can be described by the first-chance fission probability of the target nucleus with an excitation energy equal to the incident photon energy.

IV. COMPETITION BETWEEN NEUTRON EVAPORATION AND FISSION

The most probable residual nucleus (Z, A, E_γ) formed after the primary quasi-deuteron photoabsorption de-excites by a mechanism of competition between nucleon evaporation and fission. For actinide residuals of low excitation energy ($E^* \approx E_\gamma \lesssim 130$ MeV) the main modes of nuclear de-excitation are neutron evaporation and fission (charged particles, such as proton and alpha particle, have the additional difficult of a coulomb barrier which hinders the emission of these particles). The quantitative description of the process [12,14] is based on the drop model for nuclear fission proposed by Bohr and Wheeler [20], and the statistical theory of nuclear evaporation developed by Weisskopf [21]. Accordingly, the fission probability relative to neutron emission is given by Vandenbosch-Huizenga's equation [22], which reads

$$\frac{\Gamma_f}{\Gamma_n} = F = \frac{15}{4} \frac{(\sqrt{4ra_n(E^* - B_f)} - 1)}{rA^{\frac{2}{3}}(E^* - S_n)} \times \exp\{2\sqrt{a_n}[\sqrt{r}\sqrt{E^* - B_f} - \sqrt{E^* - S_n}]\} \ . \quad (10)$$

Thus, the first-chance fission probability, i.e., the nuclear photofissility for the nuclei here considered has been calculated as

$$f \approx f_1 = \frac{F}{1 + F} \ . \quad (11)$$

In expression (10), S_n is neutron separation energy [23], and B_f is the fission barrier height correct for the nuclear excitation,

$$B_f = B_{f_0}\left(1 - \frac{E^*}{B}\right) \ , \quad (12)$$

where B_{f_0} is the ground state fission barrier [24], and B is the total nuclear binding energy [23]. For the level density parameter of residual nucleus after neutron evaporation, a_n, we use a modern expression,

$$a_n = \tilde{a}\left\{1 + [1 - \exp(-0.051E^*)] \ \frac{\Delta M}{E^*}\right\} \ , \quad (13)$$

which has been proposed by Iljinov *et al.*[25]. Here, ΔM represents the shell model correction to the nuclear mass [24], and

$$\tilde{a} = 0.114A + 0.098A^{\frac{2}{3}} \qquad \text{MeV}^{-1} \quad (14)$$

is the asymptotical value of a_n (a small correction on E^*-values due pairing effects was neglected in (13)). The constants which appear in (13) and (14) are adjustable parameters resulting from the systematic study on level density carried out with hundreds of excited nuclei (for details see [25]).

The quantity $r = a_f/a_n$ (ratio of the level density parameter at the fission saddle point to a_n) is unknown, and it has been determined in a semiempirical way. The experimental fissility data from Table 1 are used together with equations (10-14) to obtain the ratio $r = a_f/a_n$ for each reaction case. Once the r-values were obtained as described above they could be fitted to a general expression of the type

$$r = 1 + a\left(\frac{Z^2}{A} - b\right) \ , \quad (15)$$

where a and b are constants to be determined by least-squares analysis. Finally, we insert back into Eq. (11) the semiempirical r-values given by (15) to obtain the nuclear fissility for each target nucleus and the different incident photon energies.

V. RESULTS AND DISCUSSION

For the various target nuclei considered in the present nuclear photofissility study we observe, firstly, that the photofission cross sections do not vary remarkably in the range 60-130 MeV ($\sim 23\%$ for ^{232}Th and ^{233}U, and $\sim 16\%$ for ^{235}U, ^{238}U and ^{237}Np, as it follows from data of Table 1). The same occurs with the total nuclear photoabsorption cross section (a variation not greater than 8%, or practically constant, within the uncertainties, as we can see in Fig. 1-c). Therefore, nuclear fissility f from the measurements results practically constant in the range 60-130 MeV for each target nucleus (Table 1), thus it is sufficient to consider the average value, $\bar{f}$, for each case. However, we remark on an increasing trend of $\bar{f}$ with parameter Z^2/A (Fig.2-a, full circles) except for the ^{233}U isotope. Since this nuclide exhibits the lowest fission barrier height as calculated from the droplet model (4.60 MeV [24]), one should expect for a value of nuclear fissility greater than that for ^{235}U, which has a higher fission barrier (4.77 MeV). On the contrary, this was not the case observed experimentally (Fig. 2-a).

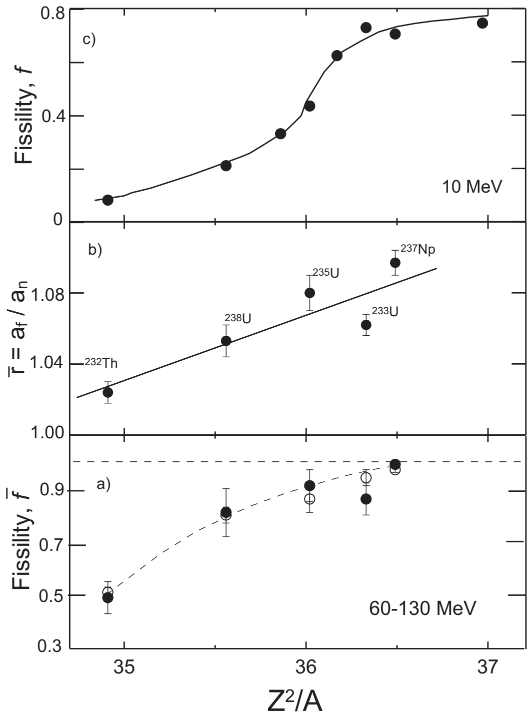

FIG. 2: a) Average nuclear fissility, $\bar{f}$, versus parameter Z^2/A: full circles are average values from experimental data listed in Table 1; open circles are calculated values from the present analysis (see text); the curve is drawn by eye. b) ratio $r = a_f/a_n$ versus Z^2/A: points represent average semiempirical values as obtained in this work; the straight line is a least-squares fit to the points (cf. Eq.(15)). c) Experimental nuclear fissility-values at 10-MeV photon energy for various actinides [27,28]; the curve is drawn by eye.

The values for ratio $r = a_f/a_n$ determined semiempiracally for each target nucleus (section 4) result rather independent of energy, and their average value, $\bar{r}$, calculated in the range 60-130 MeV is plotted versus Z^2/A, where the value for ^{233}U isotope appears displaced somewhat from the general linear trend. By assuming the parametrization defined by Eq.(15) as proposed in [26], which is valid only for actinides, we found for the constants a and b the values $a = 0.0389$ and $b = 34.24$. Finally, the $\bar{r}$-values determined in this way are inserted into (10) to obtain the calculated nuclear fissilities. The average values are shown in Fig. 2-a (open circles), where we can see that the relative position of the average measured and calculated fissilities for each nucleus is similar to that shown in Fig.2-b for the quantity $\bar{r}$, between the points and the straight line fitted to them.

Finally, let us consider the unexpected case of $\bar{r}$- and $\bar{f}$-values for ^{233}U target nucleus being slightly lower than the predicted ones ($\sim$ 10% lower in the case of $\bar{f}$ and $\sim$ 3% in the case of $\bar{r}$, as shown in Fig.2-a,b). Such a result contradicts experimental fissility data at low energies [27,28] (Fig. 2-c), as well as the prediction from droplet model for fission [24]. However, serious difficulties there exist when evaluating small (but important) shell effects which surely contribute to the final value of the fission barrier. This latter value, in turn, change the semiempirical $\bar{r}$-values. In addition, systematic uncertainties associated with σ_f-measurements of isotopes very close to each other, such as ^{235}U and ^{233}U can make difficult a better determination of the experimental σ_f-values for these isotopes (for details see Ref. [5]).

VI. CONCLUSION

A semiempirical approach has been developed based on the current two step-model (quasi-deuteron nuclear photoabsorption followed by a process of competition between neutron evaporation and fission) for photonuclear reactions and used to analyse available experimental data on photofission cross section of actinide nuclei (^{232}Th, ^{233}U, ^{235}U, ^{238}U and ^{237}Np) in the range 60-130 MeV [5]. The adjustable parameter $r = a_f/a_n$ resulted constant for each target nucleus in the incident photon energy range considered, but the average value $\bar{r}$ shows an increasing linear behavior with parameter Z^2/A (Fig. 2-b). This result is valid only for actinide nuclei ($Z^2/A \gtrsim 34.25$), which is in quite good agreement with previous conclusions by Martins *et al* [26]. The average nuclear fissilities have been quite well reproduced by the present analyses, and have shown also an increasing trend with Z^2/A (Fig. 2-a), although differences not yet completely resolved there exist in the case of ^{233}U isotope.

Acknowledgments

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