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New Approach to Nuclear Photofission Reactions above 0.15 GeV

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A simple approach to evaluate nuclear photofissilities at energies above the pion photoproduction threshold has been developed. It is based on the current, two-step model for intermediate-energy photonuclear reactions, i.e. a photon-induced intranuclear cascade followed by a fission-evaporation competition process for the excited, post-cascade residual nucleus. The calculation method (semiempirical by nature) shows that fissility (i.e., total fission probability) is governed by two basic quantities, namely, the first-chance fission probability for the average cascade residual, and a parameter which defines an evaporative sequence of residuals in which the average, equivalent chance-fission probabilities of nuclides belonging to the same generation are located. The ^{nat}Pb photofissility data measured recently in the range $\sim 0.2 - 3.8$ GeV at the Thomas Jefferson Laboratory could be explained very satisfactorily by the present approach.

The simultaneous photofission cross section measurements carried out very recently at the Thomas Jefferson Laboratory on a number of actinide target nuclei and natural lead using tagged photons in the range $\sim 0.2-3.8$ GeV and PPAD-detectors for detection of fission fragments [1,2] made possible to extract important conclusions about the nuclear photoabsorption and fissility of these nuclei. Among others, i) the ²³⁷Np photofissility is very close to unity in the whole energy interval, thus indicating that its photofission cross section is almost completely equal to the total photoabsorption cross section, and ii) the photofissility for all other actinide nuclei (uranium isotopes and thorium) is less than unity, therefore their photofission cross section does not represent the photoabsorption cross section for these nuclei. Very recently, results of a detailed and refined description of photofission reactions in heavy nuclei covering a large photon energy range ($\sim 0.07-4.0$ GeV) became also available [3]. Since photofissility data for actinide targets have been already analysed to some detail [2-4], we decided in the present work to focus attention on the newest photofissility data of ^{nat}Pb reported in [1,2]. Photofissility data will be here analysed in the framework of a phenomenological, semi-empirical way, aiming to obtain average calculated fissility values from an approach which has been developed for the first time in describing intermediate-energy photofission reactions in the entire energy range of $\sim 0.2-3.8$ GeV covered by the measured photofission cross section data. Monte Carlo calculations are, at present, the main tool to describe quantitatively both the cascade and fission-evaporation competition processes, as well as to obtain the total fission probabilities (i.e., fissility values) for a number of photofission reaction cases. However, for cases where the target nucleus is expected to have very low fissility-values (pre-actinide, intermediate-mass, and less massive nuclei), the available codes may reveal themselves very time-consuming in ob-

taining a calculated fissility-curve of acceptable uncertainty over a large energy-interval such as $\sim 0.2-4.0$ GeV. This fact led us to develop an alternative method to evaluate nuclear photofissilities when they are expected to be not greater than $\sim 10\%$, as it is the case for nuclei of $Z < 84$ and $A < 210$.

An excited, residual nucleus is always produced at the end of the rapid intranuclear cascade process as a result of escaping (or not) of a small number of nucleons and/or pions. For an incident photon energy E_γ on a target nucleus (Z, A) two extreme situations can be envisaged: i) no proton, neutron, and/or pion are ejected at all during the cascade, i.e., $N_p^c = N_n^c = N_\pi^c = 0$, and, in this case, the maximum value of the excitation energy left to the cascade residual is given by

$$E_{max}^* \approx \begin{cases} E_\gamma, & E_\gamma < B \\ B, & E_\gamma \geq B, \end{cases} \quad (1)$$

where B is the total binding energy for (Z, A) ; ii) nearly all the incident energy E_γ is used to eject nucleons in the rapid cascade stage of the reaction and, in this case, the maximum number of protons and neutrons emitted are, respectively,

$$N_{pmax}^c \approx \frac{Z}{A} \frac{E_\gamma}{E_c^p}, \quad \text{and} \quad N_{nmax}^c \approx \left(1 - \frac{Z}{A}\right) \frac{E_\gamma}{E_c^n}. \quad (2)$$

In this second extreme situation the minimum of excitation energy left to the produced residual nucleus is that energy with which an additional nucleon cannot be emitted during the cascade. The value for this energy is approximately given by the average value of the neutron and proton cut-off energies, i.e.,

$$E_{min}^* \approx \frac{1}{2}(E_c^n + E_c^p). \quad (3)$$

Assuming that the average characteristics of the post-cascade residual nucleus (\bar{Z}^* , \bar{A}^* , \bar{E}^*) are defined by the simple mean between the extreme values of the quantities mentioned above, we can write

$$\begin{aligned}\bar{Z}^* &\approx Z - \frac{E_\gamma}{2} \frac{Z}{A} \frac{1}{E_c^p}, \\ \bar{A}^* &\approx A - \frac{E_\gamma}{2} \left[\frac{Z}{A} \frac{1}{E_c^p} + \left(1 - \frac{Z}{A}\right) \frac{1}{E_c^n} \right],\end{aligned}\quad (4)$$

and

$$\bar{E}^* \approx \begin{cases} \frac{E_\gamma}{2} + \frac{1}{4}(E_c^n + E_c^p), & E_\gamma < B \\ \frac{B}{2} + \frac{1}{4}(E_c^n + E_c^p), & E_\gamma \geq B. \end{cases}\quad (5)$$

The above quantities represent the average atomic and mass number (4), and excitation energy (5) of the post-cascade residual nuclei, and they are thought as the substitutes of their respective distribution functions in the sense that the average cascade residual is produced with probability equal to unity.

The second stage of the photofission reaction is described by a fission-evaporation competition process starting from the average initial, excited residual nucleus (\bar{Z}^* , \bar{A}^* , \bar{E}^*). Neutron, proton, and alpha particle emissions are here considered the modes of de-excitation which may compete more significantly with each other and with the fission mode for all subsequent residuals formed at each step along the evaporative sequence. Fissionable evaporation residuals can be thought as being formed in generations. Let n be the order of a generation of residuals: $n = 1$ corresponds to the cascade residual, i.e., the first residual (\bar{Z}^* , \bar{A}^* , \bar{E}^*), and the partial fission probability is simply the first chance-fission probability, $P_1^p = f_1$. For $n = 2$, formation of three evaporation residuals may occur, and the partial fission probability due to the chance-fission of the residuals in the second generation is

$$P_2^p = n_1 f_{2n} + p_1 f_{2p} + \alpha_1 f_{2\alpha}, \quad (6)$$

where n_1 , p_1 , and α_1 represent the probability for neutron, proton, and alpha particle emissions, respectively, and the f_2 's are the second chance-fission probabilities. Similar expressions can be written for higher-order generations of residuals. Each term in (6) represents the chance-fission probability of the respective residual nucleus formed. The number of fissionable residuals which may be formed in the generation of order n is 3^{n-1} , and the total fission probability of the cascade residual is, therefore, given by

$$P_f^t(\bar{Z}^*, \bar{A}^*, \bar{E}^*) = \sum_{n=1}^{n_g} P_n^p. \quad (7)$$

The maximum number of generations of residuals is estimated as $n_g \approx \bar{E}^*/\bar{E}_{ev}$, where \bar{E}_{ev} represents the average total energy removed from the system per particle evaporated.

The routine calculation of the probability-values for the neutron emission (n), proton emission (p), alpha-particle

emission (α), and fission (f) modes has been already detailed in [5-7]. For the level-density parameter of the residual nucleus after neutron evaporation, a_n , we adopted the expression

$$a_n = \tilde{a} \left\{ 1 + [1 - \exp(-0.051E^*)] \frac{\Delta M}{E^*} \right\} \text{ MeV}^{-1} \quad (8)$$

proposed by Iljinov *et al* [8], in which ΔM is the shell correction in the calculated nuclear mass as tabulated in [9], and

$$\tilde{a} = 0.114A + 0.098A^{2/3} \text{ MeV}^{-1} \quad (9)$$

is the asymptotic value of a_n (a small correction on E^* due to pairing energy effects has been neglected in (11)) (for details see [8]). Finally, the values for parameter $r = a_f/a_n$ (ratio of the level density parameter at the fission saddle point to a_n) have been obtained from a semiempirical determination of r -values which resulted from a systematic study of fissility on a number of experimental photofission cross section data measured in the quasi-deuteron energy region of photonuclear absorption on twelve target nuclei ranging from Sm to Bi [10]. An analysis of all such data allowed us to parametrize the r -values according to

$$\begin{aligned}r &= 1 + \frac{\xi}{E^{*\eta}}, \quad \xi = \exp[0.150(222 - A)], \\ \eta &= 0.0352(235 - A),\end{aligned}\quad (10)$$

which expressions are valid for $150 < A < 210$ and excitation energies $E^* \gtrsim 40$ MeV.

A chance-fission probability, q_{ni} , is a quantity defined by the product of the formation probability of residual i in the generation n times the fission probability of this residual, f_{ni} . Certainly, a given evaporation residual may be formed through many different evaporation paths, and it may have different formation probabilities, thus leading to different chance-fission probabilities for this residual. We have used the routine calculation outlined above to evaluate the chance-fission probabilities for the *most* and *least* probable evaporation paths starting from the initial, average cascade ^{196}Pt excited to 533 MeV, produced by the interaction of 1.02-GeV photons with $^{\text{nat}}\text{Pb}$ target (Eqs.(4-5)). Figure 1 shows the q_{ni} values so obtained (points), where values for the second (3 points) and third (9 points) generations are made evident. Surprisingly, the q_{ni} -results lie (in a log-scale) very approximately on straight lines, one for the most likely evaporation path, i.e., for the greatest chance-fission probabilities in each generation of residuals, and another one for the least probable chance-fission of residuals eventually formed. This means that the chance-fission probability for any other evaporation residual formed in a given generation should be a value between these two linear limiting trends (Fig. 1). Calculations have been also performed at other (0.21-, 0.54-, 1.53-, and 3.78-GeV) incident photon energies on $^{\text{nat}}\text{Pb}$ target, therefore producing different average excited cascade residuals (^{205}Tl , ^{201}Hg , ^{191}Os , and ^{166}Ho , respectively). We remark that the same pattern like the one exhibited in Fig. 1 was apparent in all these cases studied. The very interesting results reported above suggest

that the chance-fission probabilities can be parametrized by an equation of the form $q_{ni} = f_1 e^{-(n-1)s_i}$, in which s_i denotes (in ln-scale) the slope of the straight lines (Fig. 1).

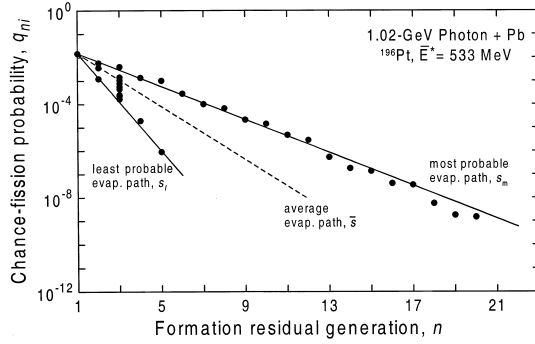


Figure 1. Chance-fission probability, q_{ni} , plotted against the order of generation of evaporation residuals, n , for 1.02-GeV incident photon on ^{nat}Pb . The extreme evaporation paths are indicated by the full straight lines of slopes s_m and s_l , and the evaporation path on which the equivalent, average residuals of each generation are located is represented by the dashed line of slope \bar{s} . Points represent calculated q_{ni} -values as explained in the text.

For a given generation of residuals (n fixed) the partial fission probability is the summation

$$P_n^p = \sum_{i=1}^N q_{ni}, \quad N = 3^{n-1}, \quad q_{ni} = f_1 e^{-(n-1)s_i}. \quad (11)$$

We can obtain an estimation of the P_n^p 's by taking simply the product of the number of residuals which may be formed in generation n times a certain average chance-fission probability, \bar{q}_n , i.e., $P_n^p \approx \bar{q}_n \times 3^{n-1}$. The \bar{q}_n -values are, in turn, obtained from a certain average slope-value, \bar{s} ($s_m < \bar{s} < s_l$), such that $\bar{q}_n = f_1 e^{-(n-1)\bar{s}}$. Parameter \bar{s} defines the slope of an average sequence of evaporation residuals which lies between the most and least probable sequences of residuals (dashed line in Fig. 1). In other words, the sum of the 3^{n-1} chance-fission probabilities as given by Eq.(11) represents, for each generation of residuals, the chance-fission probability of an average, equivalent evaporation residual located on the \bar{s} -sequence.

Parameter \bar{s} is introduced here to overcome the difficulty of calculating the $\sum_{n=1}^{n_g} 3^{n-1} \approx 3^{n_g}/2$ chance-fission probabilities which may appear during the de-excitation process of the cascade residual (in a 2-GeV photointeraction with a ^{nat}Pb target, for instance, we would have to calculate $\sim 3 \times 10^{23}$ chance-fission probabilities!). One should remark that parameter \bar{s} is, in a sense, model dependent, since one could take into account, for instance, not only neutron, proton, and alpha particle as the competitors with fission, but also deuteron, triton, ^3He , and other more complex clusters competing with fission. Finally, the average nuclear fissility is calculated as

$$\bar{f}_c(E_\gamma) = P_f^t(\bar{Z}^*, \bar{A}^*, \bar{E}^*) = \sum_n P_n^p = \bar{f}_1 \sum_n 3^{n-1} e^{-(n-1)\bar{s}} \quad (12)$$

which gives

$$\bar{f}_c = \frac{\bar{f}_1}{1 - 3e^{-\bar{s}}}. \quad (13)$$

In this way, for each incident photon energy on a target nucleus (i.e., an average cascade residual), fissility can be easily calculated provided the values of \bar{f}_1 and \bar{s} are known.

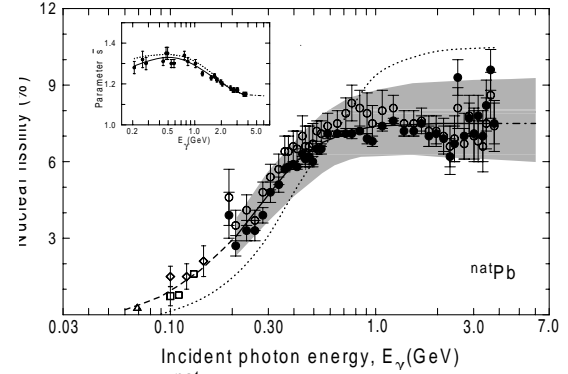


Figure 2. Fissility of ^{nat}Pb versus photon energy. Full circles represent the fissility-values relative to ^{237}Np measured by Cetina *et al.* [2]; open circles are the absolute fissility-values obtained from the total photoabsorption cross section for ^{nat}Pb as explained in the text. The full line is the smooth, semiempirical fissility-curve as obtained in the present work in the range ~ 0.2 – 3.8 GeV, and the dash-dotted line is the prediction up to 6 GeV (the shaded area indicates the associated uncertainty). The dotted line is the result using the RELDIS code (variant B) as reported in [3]. Experimental fissility data at lower energies are taken from [11,12] (squares), [13] (triangle), and [14,15] (diamonds), and the dashed line is a trend drawn by eye through these points. In the inset, points represent the semiempirical \bar{s} -values obtained from the average experimental fissilities (\bar{f}_e -values); the full line is a least-squares fit to these points, and the dotted line is the trend of \bar{s} when only the relative fissilities are considered.

The average first-chance fission probability, \bar{f}_1 , can be evaluated, for instance, from the three first-chance fission probabilities which define the most probable evaporation path (q_{11}, q_{21}, q_{31}), and from those which define the least probable one (q_{11}, q_{23}, q_{39}) (see Fig. 1). By least-squares analysis one obtains

$$\bar{f}_1 = \left[\frac{q_{11}^5 \times q_{21} \times q_{23}}{(q_{31} \times q_{39})^{1/2}} \right]^{1/6}. \quad (14)$$

The values of \bar{s} are, in turn, determined by taking the experimental fissility-values corresponding to the incident photon energies which produce average cascade residuals of both \bar{Z}^* and \bar{A}^* integer. The final values of \bar{s} are then extracted from a smooth trend of \bar{s} versus E_γ , and inserted into back Eq. (13) to obtain the \bar{f}_c 's.

We have applied the present phenomenological, semiempirical, photofission approach to analyse the photofission experimental data for ^{nat}Pb recently obtained at the Thomas Jefferson Laboratory by Cetina *et al.* [2]. Results are summarized in Fig. 2. Both the relative and absolute photofission data have been considered to define the average experimental photofission-values, $\bar{f}_e = (f_r + f_a)/2$. The f_r -values are obtained directly from Table V of Ref.

[2] by taking the ratio of the photofission cross section measurements listed in column 7 to those in column 2. These are showed as full circles in our Fig. 2. The absolute photofissility, f_a , is defined as the ratio of the photofission cross section to total photoabsorption cross section, and the f_a -values for ^{nat}Pb have been obtained here as the quotient of the entries listed in column 7 referred above to the corresponding (in energy) values from the smooth curve fitted to the existing photoabsorption cross section data for ^{nat}Pb (this curve is that one reported in Fig. 12-a of [2]). The f_a -values so obtained are represented as open circles in our Fig. 2. Some of the \bar{f}_e -data are interpolated values corresponding to E_γ -values previously chosen in such a way as to give both \bar{Z}^* and \bar{A}^* integer. Fig. 2 also shows a few experimental points obtained at different laboratories in the quasi-deuteron region of photoabsorption [11-15]. A free, dashed line shows the trend of fissility in this low energy region.

The semiempirical \bar{s} -values which result when the average experimental photofissilities (\bar{f}_e) are used into Eq. (13) are represented by points (with error bars) plotted in the inset within Fig. 2. The smooth trend (full line) through these points gives the final values of parameter \bar{s} to be used back in Eq. (13) in order to obtain the calculated photofissility-values. Differences between \bar{f}_e and \bar{f}_c along the interval $\sim 0.2\text{--}4.0$ GeV have shown indeed small (less than $\sim 11\%$) if one considers the uncertainties of both \bar{f}_e and \bar{f}_c . This means that the quantity \bar{s} can be considered a good parameter for the present method of analysis of photofission reactions. A very similar trend for \bar{s} (dotted line in the inset graph) can be appreciated when the experimental photofissility of ^{nat}Pb relative to ^{237}Np target (i.e., the f_r -values) is used into Eq. (13) instead of the \bar{f}_e -values. For the sake of comparison, the calculated, smooth fissility-curve is depicted in Fig. 2 as the full line, where the shaded area indicates the error band associated with the \bar{f}_c -curve. The agreement between experimental (both f_r and f_a) and calculated fissility values can be considered very satisfactory. Calculated results of total fission probability for ^{nat}Pb following photoabsorption in the range $\sim 0.07\text{--}3.8$ GeV obtained with the RELDIS Monte-Carlo code by Pshenichnov *et al* [3] are represented in Fig. 2 as the dotted line (variant B in their notation). Inspection on Fig. 2 shows that this calculated fissility-curve reveals a trend which does not differ greatly from the experimental one, but it is slightly underestimated at energies below 400 MeV, and overestimated above ~ 1 GeV. The general behavior of ^{nat}Pb fissility with E_γ shows a monotonous increase of f from the lower energies up to ~ 600 MeV, and then a tendency to saturate around 7.5% at energies up to at least ~ 4 GeV. Besides, this saturation seems to be valid towards higher energies as indicated by the predictions of fissility carried out up to 6 GeV with the present approach (dash-dotted line in Fig. 2).

Parameter \bar{s} allows one to extract some information about the *point of fission*, i.e. the location on the evaporation-fission competition sequence where fission takes place. Inspection on Eq.(12) shows that fissility is reached with the cumulative, partial fission probability which increases with the order of generation of residuals,

n , but at a rate dictated by the \bar{s} -value. For example, in the case of 470 MeV photons interacting with the ^{nat}Pb target (~ 260 MeV of average excitation for the cascade residual) \bar{s} equals to 1.33 (see Fig. 2), and a simple calculation indicates that only 6 or 7 generations of residuals are needed to reach, for instance, $\sim 80\%$ of fissility. Since in this example it is possible to have nearly 22 generations of residuals, this means that fission is more likely to occur in the first third part of the evaporative sequence. At higher energies, however, \bar{s} may amount to only 1.15, and in these cases a higher number of generations of residuals is needed to obtain almost the total fission probability (~ 25 generations for 3.8 GeV photons), at the same time that more generations become possible of being formed (~ 50 in the case of 3.8-GeV photons), therefore making the *point of fission* uncertain to some extent. In other words, more energy available, more fission chances opened, therefore less defined becomes the point-of-fission (note that Eq.(13) imposes the limiting condition of $\bar{s} > \ln 3$).

In summary, nuclear photofissilities at intermediate-energies have been evaluated semiempirically using a new, simple approach according to which the distributions of atomic number, mass number, and excitation energy of the cascade residuals which would result from Monte Carlo calculations are replaced here by their respective average values (\bar{Z}^* , \bar{A}^* , \bar{E}^*). These have been defined as functions of the incident photon energy by means of simple expressions (Eqs. (4-5)) in which the neutron and proton cut-off energies play a fundamental role. Next, the de-excitation of the average cascade residuals has been described by the usual way, where neutron, proton, and alpha particle emissions and fission are considered as the main de-excitation channels. A direct calculation of the total fission probability for the cascade residual (i.e., the target nucleus fissility) has been performed by taking into account all intermediate chance-fission probabilities of residuals eventually formed throughout the evaporation chain. This calculation has been simplified in view of the remarkable pattern exhibited by the chance-fission probability values, according to which the chances for fission are shown to lie between two rather linear (in log-scale) trends, one for the most probable, and another one for the least probable sequences of fissionable residuals (Fig. 1). Finally, an adjustable parameter, \bar{s} , has been introduced, and its value (found semiempirically) defines an evaporative sequence in which the average, equivalent chance-fission probabilities of residuals belonging to the same generation are located. In this way, fissility-values can be easily calculated provided the first-chance fission probability (\bar{f}_1) and the \bar{s} -values are known (Eq. (13)). The experimental fissility data for ^{nat}Pb target have been reproduced by the present scheme very satisfactorily (Fig. 2), at the same time that parameter \bar{s} do exhibit a rather monotonous trend in the entire photon energy range investigated here (inset within Fig. 2).

To conclude, the present approach can certainly be applied to other photofission reaction cases, such as those for heavy actinide, pre-actinide, and intermediate-mass target nuclei (in the case of actinides it is necessary first to search for an appropriate expression for the ratio $r = a_f/a_n$

(Eq.10)). An upper limiting value for the incident photon energy, however, there exists (about 9 GeV in the case of heavy target nuclei), therefore the basic, simplified assumptions made to define the average characteristics of the post-cascade nucleus (eqs. 4-5) should be reformulated to attain the very-high photon energy region.

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