Au photofission cross section by quasimonochromatic photons in the intermediate energy region

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The photofission cross section of Au was determined in the energy range 100–300 MeV by means of a quasimonochromatic photon beam. The nuclear fissility P_f was calculated using the recently measured total photoabsorption cross sections. The nuclear excitation energy E^*, charge and mass of compound nucleus were obtained by means of an intranuclear cascade Monte Carlo calculation. The fissility values determined for Au, Bi, and U were compared with the predictions of the cascade-evaporation model and remarkably fitted by the calculation.

I. INTRODUCTION

The interest in photofission studies in the intermediate energy region stems from the fact that the photofission process is strictly related to the absorption and excitation mechanisms of nuclei by photons and to the subsequent process of nuclear deexcitation. While the intermediate energy photon mainly interacts with single nucleons or nucleon clusters in the nucleus, fission must be regarded as a collective process of the nucleus. Thus photofission studies can give insights about how a local excitation is propagated in nuclear matter and transferred to a collective nuclear excitation.

In this context the photofission of preactinides seems to be the most suitable tool to investigate the complex dynamics of the fission process at high excitation energy, since (a) the photon has a well-known electromagnetic interaction with the nucleus, and transfers energy but comparatively little momentum and angular momentum to the struck nucleus, so giving the possibility to observe excitation energy effects; (b) the photon, in the intermediate energy region, "sees" all nucleons in the nucleus, due to its volume dependent absorption cross section, and is therefore very effective in "heating" the nucleus; (c) for preactinide nuclei, the fissility (i.e., the probability that a nucleus with a given Z and A will undergo fission) is a strong function of the excitation energy, due to the high fission threshold, and, consequently, it is more directly related to photoexcitation processes; and (d) for these nuclei the role played by the fission barrier provides stringent tests of any semiempirical mass formula, as expressed within the liquid drop model.[1]

The drawbacks are the lack of intense monochromatic photon beams with variable energy in this energy domain, and the low fission cross section of these nuclei. One wishes to measure photofission cross sections as a function of photon energy, while, when using a normal bremsstrahlung beam, one is forced to transform the experimental data, i.e., bremsstrahlung yield curves, to photon cross sections. The well-known problems in performing such a transformation with a bremsstrahlung photon beam[2] are evident in the poor quality of the results. If we use an alternative reaction, electrofission, we are faced with the further task of calculating a reliable virtual photon spectrum: a topic of current investigation.[3]

To avoid these difficulties we have taken advantage of the LEALE (Laboratorio Esperienze Acceleratore Lineare Elettroni) photon beam facility at Frascati National Laboratories, which produces an intense quasimonochromatic photon beam by in-flight annihilation of intermediate energy (100–300 MeV) positrons.[4] The use of this beam gives remarkable advantages for studying the energy dependence of photofission processes.[5]

With this beam we have studied the photofission of Bi,[6] and examined the role of the different photoexcitation mechanisms leading to fission; a controversial item until recently.[7,8] In this paper we report the results of photofission measurements of another preactinide, the Au nucleus, which, due to its lower Z^2/A, is characterized by a fissility that saturates at higher energy[9] and thus is sensitive to the excitation energy over a wider range. The absolute photofission cross sections of Au in the literature[10–17] display large discrepancies all over the explored photon energy range. This is probably due to the use of bremsstrahlung beams, the spectra of which, moreover, were not measured during the data taking. These inconsistent data cannot be used to draw conclusions about the photofission process in Au.

In this experiment the Au photofission cross sections were derived from the experimental yields and from the on-line measured photon spectra by solving the Volterra equation using an improved unfolding method.[6] The nuclear fissility was then deduced by taking advantage of the total photoabsorption cross section measured recently in the same energy range. The analysis has been performed on the basis of the two-step picture[18] of the process. In the first, fast step the photon initiates an intranuclear cascade by forming a highly excited residual nucleus in which, after a certain time, thermodynamic equilibrium is established (*compound nucleus*). In the second, slow step this highly excited nucleus evaporates particles or undergoes fission. To define the compound nucleus excitation energy and composition, a Monte Carlo calculation of the intranuclear cascade initiated by a photon was performed. The Au fissilities from this experiment and from our previous photofission measurements on Bi and U, at the same excitation energy, were then compared with the cascade-evaporation Monte Carlo calculations and found to be in a remarkable agreement

II. EXPERIMENTAL

A. The photon beam

The LEALE quasimonochromatic photon beam facility used in this experiment was extensively described elsewhere,[4,6] so only its main characteristics are summarized here. The annihilation photons were obtained by allowing the positron beam (energy 100–300 MeV, average current ~ 15 nA, repetition rate 150 Hz, beam burst width 4 μs) to impinge upon a liquid hydrogen target, 0.0118 radiation lengths thick, enclosed in a cell with 0.012 cm kapton windows. The intensity of the positron beam was monitored continuously by a nonintercepting ferrite toroid monitor set on the beam pipe immediately before the hydrogen target, and measured by a Faraday cup placed in the focal plane of the dumping magnet. In order to increase the ratio of the annihilation monochromatic photons with respect to the unavoidable positron bremsstrahlung background, the photons were collected at $\sim 0.8°$ with respect to the positron axis, giving a monochromatic flux of $\sim 5\times 10^6$ photons per second. The photon spectrum was measured on-line by a pair spectrometer[19] and the photon flux was monitored by a quantameter; the simultaneous measurement of both the total energy and the spectrum of the photon beam allowed us to establish the intensity of the photon beam with an uncertainty of only a few percent. The collimated and cleaned photon beam had a circular spot ($\Phi \sim 4$ cm) at the target position.

B. Target assembly, fission fragment detectors, and data collection

The fission fragments were detected by a means of the glass sandwich technique.[20] The target was a Au foil 50 mm$\times$50 mm having a thickness of 0.1 mm that was sandwiched between two glass plates covering the sample surface. It was struck at a right angle by the photon beam. We employed a thick target in order to get a sufficient number of fission events in a reasonable irradiation time; however, the sandwich was thin enough to have a negligibly effect on the photon beam. In all measurements the same Au sample was irradiated. After irradiation, the glass plates were treated by the usual procedure of chemical etching and microscope scanning.[20] Both plates of the sandwich were scanned in order to obtain information on the forward-backward asymmetry of the fragments. The observed asymmetries were weak (1.08–1.14) and were weakly energy dependent, in agreement with other measurements.[21] Therefore we averaged the counts of the two plates of each sandwich in order to obtain results free from any dependence on forward-backward asymmetry. To check the effects of radiation damage in the glass plates and to estimate spurious events due to background contributions, the glass surfaces not in contact with the target were also scanned.

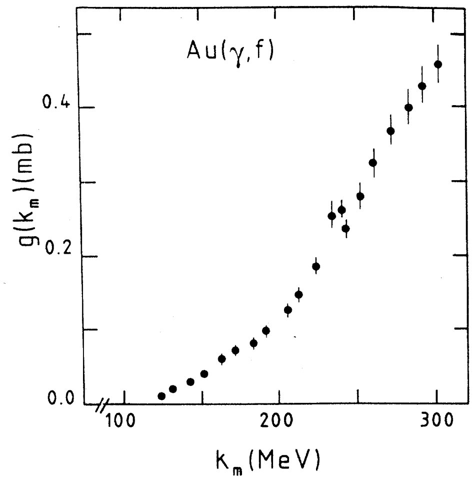

FIG. 1. Au photofission yields per equivalent quantum vs the maximum photon energy k_m.

The fission fragments were measured at 20 different positron energies between 120 and 300 MeV. The cross sections per equivalent quantum ("yields") were obtained from the number of fission tracks counted in the scanned surfaces while the exposure doses were taken from the quantameter readings. The values were obtained in arbitrary units, due to the use of a thick target. At three positron energies (150, 200, and 270 MeV) a thin Au target was also irradiated, in order to normalize the yields. The Au layer was deposited by thermal evaporation directly on the surface of one of the glass plates; the thickness and uniformity of the layer were measured by an optical interferometer[22] and through the back-scattering method.[23] The thickness resulted to be 3.80 ± 0.08 mg/cm^2. Taking the efficiency of glass plates[24] into account, the overall error in the normalizing factor was $\pm 7\%$. The experimental yields per equivalent quantum $g(k_m)$ are shown in Fig. 1 as a function of the maximum photon energy k_m.

III. PHOTOFISSION RESULTS

A. Photofission cross section

The experimental yields $g(k_m)$ are connected to the photofission cross section $\sigma_f(k)$ by the Volterra linear equation:

$$g(k_m)=\int_{k_T}^{k_m} N(k,k_m)\sigma_f(k)dk \ , \tag{1}$$

where $N(k,k_m)dk$ is the experimentally measured number of photons per equivalent quantum in the energy range $(k, k+dk)$, k_T is the fission energy threshold, and k_m is the maximum photon energy of each spectrum, assumed equal to the respective positron energy. In order to calculate the photofission cross section $\sigma_f(k)$ from the experimental yields, we have solved the integral equation (1) by a method similar to the numerical one proposed by Cook,[25] improving the accuracy in the representation of the $\sigma_f(k)$ solution by approximating it by a natural spline function instead of a stepwise one. We took $k_T=70$ MeV as the fission threshold, since the photofission cross section of Au is expected to be very low at photon energies less than 70 MeV;[12] the systematic error due to the contribution of Au photofission values at photon energies less than 70 MeV was estimated to be about 30% for the lowest energy point, about 3% for the 120 MeV point, and lower for the higher energy points.

We evaluated the fission cross section for 11 photon energies at intervals of 20 MeV from 100 to 300 MeV. The unfolding method used yields a σ_f vector that represents an estimate of the photofission cross section averaged with respect to the photon energy by a matrix **R**, that corresponds to the energy resolution function.[25] The shape of the **R**-matrix rows and, consequently, the cross-section values, depend on the accuracy of the experimental yields, on the kernel $N(k,k_m)dk$, and on the value of a smoothing parameter γ that is chosen to "regularize" the $\sigma_f(k)$ solution. The parameter γ was selected by applying a Bayesian method, suggested by Turchin and Turovceva.[26] This method allows the calculation of the probability density $P(\gamma|g)$ for obtaining particular γ values for a fixed set of experimental yields $g(k_m)$. As discussed by Turchin and Turovceva,[26] the $P(\gamma|g)$ function has a sufficiently clear-cut maximum for a number of experimental yields larger than 15. For the data reported here we had 20 experimental yields; that ensured a satisfactory estimate of the optimum γ parameter, as shown in Fig. 2. In any case we ascertained that there is not a significant change in the $\sigma_f(k)$ solution for γ values whose $P(\gamma|g)$ probability is $\geq 10\%$ of its maximum (at $\gamma=4.5$). Some typical rows of the energy resolution **R** matrix obtained for $\gamma=4.5$ are plotted in Fig. 3: They have the suitable form of an energy resolution function with maxima at the correct photon energies and relative widths of less then 20%. As stressed in our previous paper,[5] this result is a clear indication of the advantages of using an annihilation photon beam in order to study photofission in nuclei with high fission thresholds.

The photofission cross-section values obtained from the above procedure are shown in Fig. 4 (solid dots). The er-

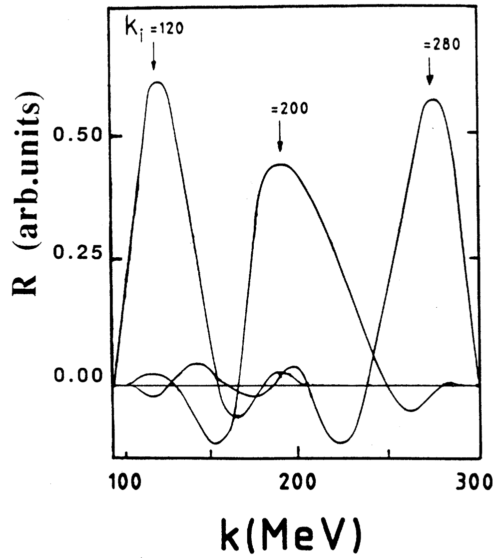

FIG. 3. Typical **R**-matrix rows for $\gamma=4.5$ at different photon energies.

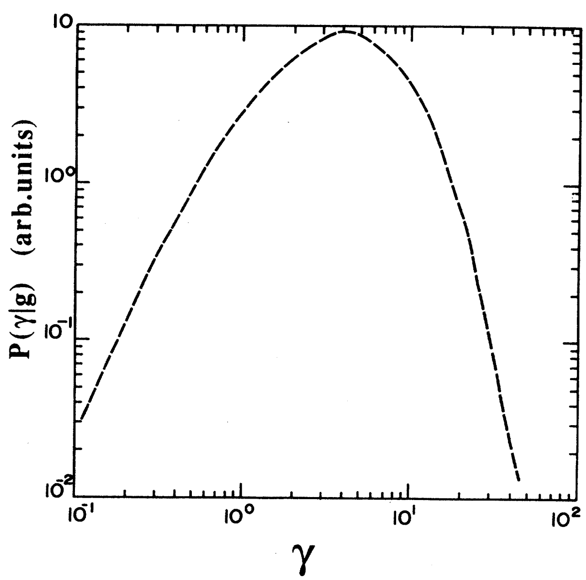

FIG. 2. Probability density $P(\gamma|g)$ as a function of the smoothing parameter γ.

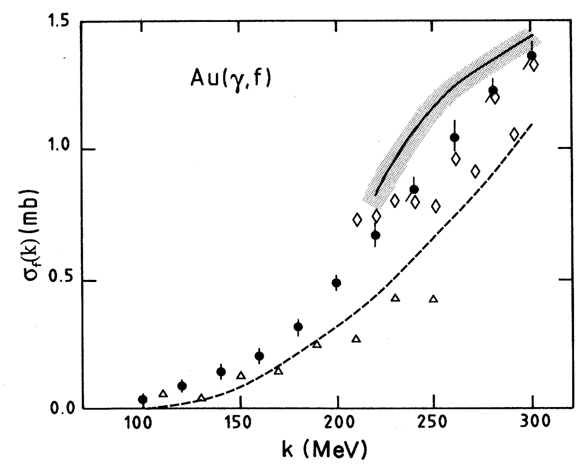

FIG. 4. The photofission cross section $\sigma_f(k)$ of Au vs photon energy k: ●, this experiment; ◇, Ref. 10; △, Ref. 11; the dashed curve represents the results of Ref. 12, with the low-energy yields taken from Refs. 11 and 15; the solid line represents the data of Ref. 13 (the shaded area gives an indication of the errors). Where not shown, the errors are not deducible from the original papers.

rors were calculated by the usual propagation rules; they account for the experimental errors as well as for the additional uncertainties resulting from the unfolding procedure. A further overall uncertainty of about 7% in the normalizing factor should be added. All the relevant data for Au photofission measured in previous experiments in the photon energy range covered by this experiment are also shown in the figure. The data of Jungerman and Steiner[10] (open diamonds) were deduced by using the photon difference method with the Shiff theoretical expression[27] for the bremsstrahlung spectrum. The data of Ranyuk and Sorokin[11] (open triangles) were obtained under the k^{-1} approximation of the bremsstrahlung spectrum. The dashed curve represents the photofission cross section inferred by Vartapetyan *et al.*[12] by fitting their experimental yields and the yields at lower energies of Refs. 11 and 15 with an assumed photoabsorption cross section, a constant fissility, and the Shiff bremsstrahlung spectrum.[27] The solid line represents the data deduced by Anderson *et al.*[13] using the Bethe-Maximon[28] expression for the bremsstrahlung spectrum, and correcting for target and collimator effects. These data present different absolute values, and rather different trends: In this respect we want to stress that our results, in addition to be the only ones obtained with quasimonochromatic photons, were derived using on-line measured photon spectra.

B. Fissility

It is possible to calculate the nuclear fissility P_f from the measured photofission cross section, $\sigma_f(k)$, through the relation

$$P_f = \frac{\sigma_f(k)}{\sigma_{\text{in}}(k)} , \qquad (2)$$

where $\sigma_{\text{in}}(k)$ is the total inelastic cross section. In our energy range, the cross section for elastic photon scattering is very low, so we can safely assume that $\sigma_{\text{in}}(k) \cong \sigma_T(k)$, where $\sigma_T(k)$ is the total photoabsorption cross section. For $\sigma_T(k)$ we used the experimental values of Carlos *et al*,[29] who measured $\sigma_T(k)$ for a different set of heavy nuclei, and whose findings strongly suggest, in our energy region, a linear dependence with A of $\sigma_T(k)$ for nuclei ranging from beryllium to uranium. It is worthwhile to point out that the values of $\sigma_T(k)$ obtained with this assumption are compatible, within errors, with the $\sigma_T(k)$ values deduced from a recent measurement of (γ,xn) reactions on Au.[30]

In Fig. 5 the P_f values obtained for Au are plotted as a function of the photon energy k. It must be pointed out that to deduce the fissilities the $\sigma_T(k)$ values were also averaged by the **R** matrix, as was done for the extraction of the photofission cross section $\sigma_f(k)$.

IV. DISCUSSION

The peculiarity of nuclear fission induced by intermediate energy particles is that the compound nuclei have wide distributions over the excitation energy E^* and mass and charge numbers A and Z. These distributions

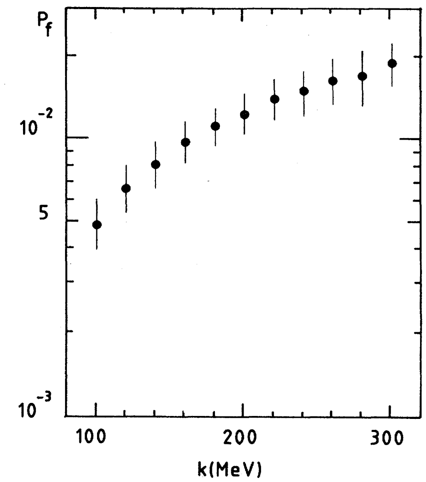

FIG. 5. Au nuclear fissility data from this experiment vs photon energy k.

are determined by the mechanisms of the nuclear absorption of the probe. In the case of photons and in the energy region investigated, two mechanisms play a major role: (i) photon absorption by a quasideuteron n-p pair, for $k < 140$ MeV and (ii) photon absorption via pion production on an intranuclear nucleon, for $k > 140$ MeV. The nucleus is "heated" mainly through the scattering of the nucleon pair produced in the quasideuteron absorption of photon or through absorption of the pion on a nucleon-nucleon pair.

In Fig. 6 the calculated compound nucleus distribu-

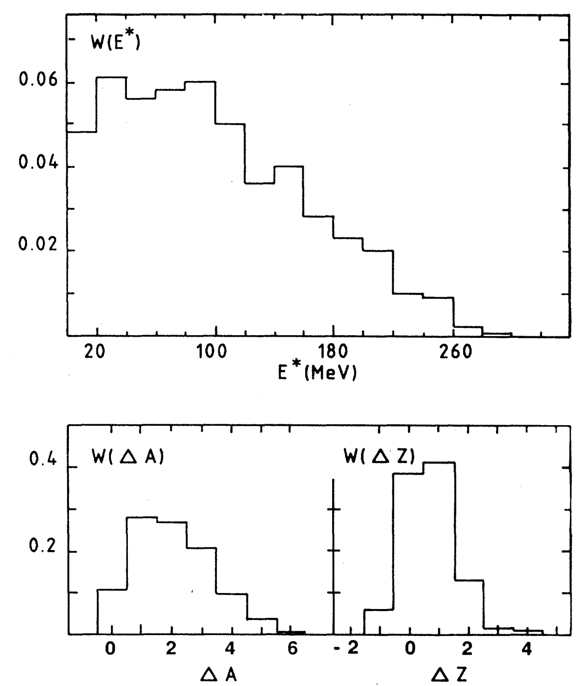

FIG. 6. Calculated distributions of the excitation energy E^*, and the mass and charge loss, ΔA and ΔZ, respectively, of the compound nuclei produced by 300 MeV photons impinging on Au. All curves refer to Monte Carlo intranuclear cascade calculations and are normalized to one inelastic interaction.

TABLE I. Characteristics of the compound nuclei produced in the interaction of 300 MeV photons with different target nuclei as calculated with a Monte Carlo cascade-evaporation code.

Target	$\langle E^* \rangle$ (MeV)	$\langle \Delta A \rangle$	$\langle \Delta Z \rangle$
^{197}Au	97	2.03	0.67
^{209}Bi	97	1.99	0.60
^{238}U	101	2.00	0.63

tions are shown as a function of the excitation energy E^*, and the mass and charge losses, ΔA and ΔZ, respectively, as an example, for the interaction of a 300 MeV photon with an Au nucleus. The distributions were calculated in the framework of the intranuclear cascade model,[31] with parameter values[18] chosen to reproduce the data on fissilities of nuclei by intermediate energy particles. The calculation also predicts that compound nuclei produced by 300 MeV photons impinging on Au, Bi, and U nuclei have the same average values of E^*, ΔA, and ΔZ (see Table I), and that the distribution of these variables were similar.

In Table II and Fig. 7, we report the experimental fissility values P_f derived at 300 MeV on Au from this experiment (solid dot), and from our previous photofission measurements on Bi (solid triangle),[6,7] and U (solid square).[32] Due to the considerations outlined above, the experimental fission cross sections σ_f and, hence, the fissilities are the result of an average over the ensemble of the compound nuclei produced in the reaction. Also in Table II and Fig. 7, the results of our intranuclear cascade calculations with the evaporation model of Ref. 18 are also reported for 300 MeV incident photons; the abscissa in Fig. 7 are the average values of the "fissility" parameter Z^2/A. In order to calculate the fission barrier heights of compound nuclei, we used the modified liquid drop model.[33] The a_f/a_n ratio between the level density parameters of the nucleus with equilibrium deformation and the nucleus which has a configuration corresponding to the fission saddle point was taken to be 1.02. Moreover, we neglected shell effects, since they vanish for excitation energy $E^* > 30$ MeV.[34] The agreement found between experimental results and theoretical calculations over the range of Z^2/A explored is remarkable.

In principle, the analysis of data on nuclear fission by intermediate energy photons in the framework of the cascade-evaporation model allows us to investigate effects such as the spread in composition and excitation energy of compound nuclei, the thermal disappearance of shell effects, the dependence of fission barrier heights on the excitation energy, and the proper choice of the liquid drop model parameters.[18] In order to draw more definite conclusions one needs to infer the actual excitation energy and nuclear composition of highly excited compound nuclei. Therefore, from the experimental point of view, one should perform exclusive experiments with monochromatic photons, measuring the characteristics of both the fission fragments and the emitted particles. Moreover, one should expand the region of energies, probes, and Z^2/A explored. From the theoretical point of view, detailed calculations using more sophisticated cascade-evaporation models are needed.

TABLE II. Fissility values for ^{197}Au (this experiment) and ^{209}Bi (Ref. 6) excited by 300 MeV photons, and for ^{238}U (Ref. 32) excited by 280 MeV photons. The calculations refer to the Monte Carlo cascade-evaporation code results for 300 MeV photons.

Target	P_f (expt.)	P_f (theory)
^{197}Au	$(1.85\pm0.35)\times10^{-2}$	1.4×10^{-2}
^{209}Bi	$(1.05\pm0.30)\times10^{-1}$	1.5×10^{-1}
^{238}U	$(8.70\pm1.30)\times10^{-1}$	7.8×10^{-1}

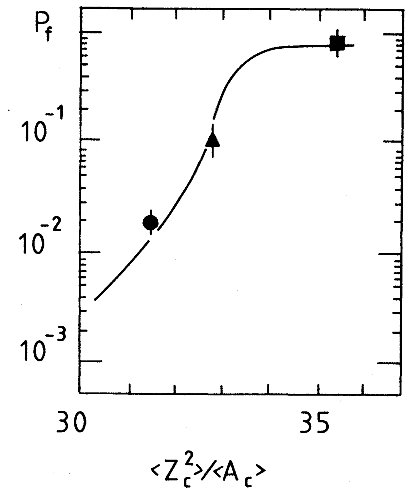

FIG. 7. Fissility values for ^{197}Au (●, this experiment) and ^{209}Bi (▲, Ref. 6) excited by 300 MeV photons, and for ^{238}U (■, Ref. 32) excited by 280 MeV photons. The curve refers to the Monte Carlo cascade-evaporation calculation for 300 MeV photons.

V. CONCLUSIONS

Here we summarize our main results and conclusions. (a) We determined the photofission cross section of Au in the energy range 100–300 MeV by taking advantage of a quasimonochromatic photon beam (Fig. 4). (b) The nuclear fissility P_f was calculated using the recently measured total photoabsorption cross sections (Fig. 5). (c) The excitation energy, atomic, and charge number distributions of compound nuclei following the absorption of intermediate energy photons were calculated by means of an intranuclear-evaporation Monte Carlo calculation (Fig. 6). (d) The fissilities from targets at the same excitation energy were compared with the intranuclear cascade calculation results (Fig. 7) and a satisfactory agreement between the calculated and measured fissility values were found in the explored Z^2/A range.

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