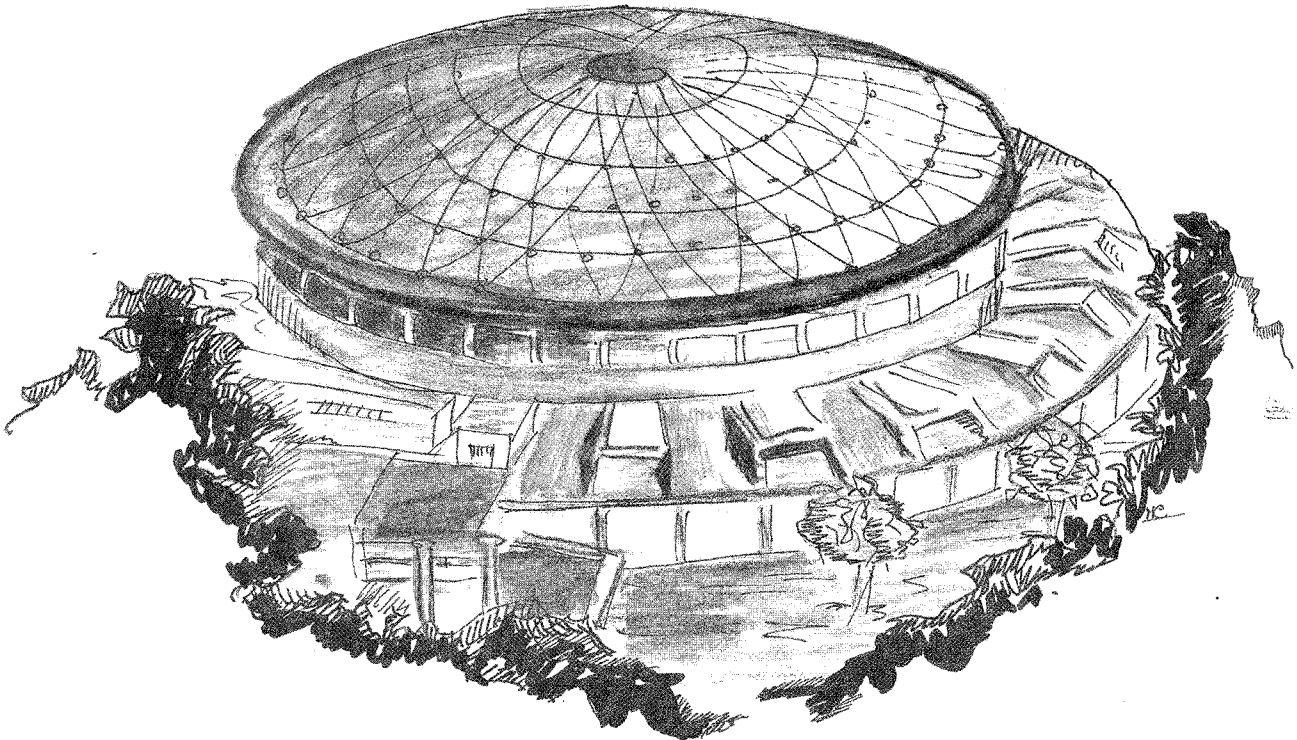# Laboratori Nazionali di Frascati

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V. Lucherini, C. Guaraldo, E. De Sanctis, P. Levi Sandri, E. Polli, A.R. Reolon,
A.S. Iljinov, S. Lo Nigro, S. Aiello, V. Bellini, V. Emma, C. Milone, G.S.
Pappalardo, M.V. Mebel:

Au PHOTOFISSION CROSS SECTION
BY QUASI-MONOCHROMATIC PHOTONS
IN THE INTERMEDIATE ENERGY REGION

Au photofission cross section by quasi-monochromatic photons in the intermediate energy region

V. Lucherini, C. Guaraldo, E. De Sanctis, P. Levi Sandri, E. Polli, A.R. Reolon,
and A.S. Iljinov[o]
Istituto Nazionale di Fisica Nucleare - Laboratori Nazionali di Frascati, I-00044 Frascati, Italy

S. Lo Nigro, S. Aiello, V. Bellini, V. Emma, C. Milone, and G.S. Pappalardo
*Dipartimento di Fisica, Università di Catania and Istituto Nazionale di Fisica Nucleare, Sezione di Catania I-95129
Catania, Italy*)*

M.V. Mebel
Institute for Nuclear Research of the Academy of Science of the USSR, Moscow, USSR

The photofission cross section of natural Au was determined in the energy range 100÷300 MeV by means of a quasi-monochromatic 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.

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. Such studies can thus give insight about how a local excitation is propagated in nuclear matter and transferred to a collective nuclear excitation.

In this context the photofission, in particular of preactinides, seems to be the more suitable tool to investigate the complex dynamics of the fission process at high excitation energies, since:

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a) the photon has a well known electromagnetic interaction with the nucleus, and transfers energy but comparatively small momentum and angular momentum to the struck nucleus, so giving the possibility to put in evidence 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, so being 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 composition 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;

d) moreover, for these nuclei, the role played by the fission barrier allows to perform very stringent tests of any semi-empirical 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. In fact, one usually 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: and the well known problems in performing such a transformation with a continuous photon beam[2] are expressed in the poor quality of the results. The use of electrofission to overcome these difficulties is faced with the further task of calculating a reliable virtual photon spectrum: a topic of still current investigation.[3]

We have therefore taken advantage of the LEALE (Laboratorio Esperienze Acceleratore Lineare Elettroni) photon beam facility at Frascati National Laboratories, which produces an intense quasi-monochromatic photon beam by in-flight annihilation of intermediate energy ($100\div300$ MeV) positrons;[4] the use of this beam gives remarkable advantages in studying the energy dependence of photofission processes.[5]

With this beam we recently studied the photofission of Bi,[6] and examined the role of the different photoexcitation mechanisms leading to fission: an item controversial until the last years.[7,8]

In this paper we report the results of photofission measurements of another preactinide, the Au nucleus, which, due to the lower Z^2/A, is characterized by a fissility which saturates at higher energy[9] and thus is sensitive to the excitation energy over a wider range. The absolute photofission cross sections of Au found in literature are difficult to use in order to draw a definite conclusion, since the results of different experiments are scarce and highly scattered, due to the use of continuous bremsstrahlung beams, the spectra of which were not measured during the data taking. In fact, the

experimental data[10÷17] of Au photofission cross sections display large discrepancies all over the explored photon energy range.

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 recently measured total photoabsorption cross section 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 stage the photon initiates an intranuclear cascade: as a result a highly excited residual nucleus is formed in which, after a certain time, thermodynamic equilibrium is established (*compound nucleus*); in the second, slow stage 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 in a remarkable agreement.

2. Experimental

I. The photon beam

The LEALE quasi-monochromatic photon beam facility used in this experiment was extensively described elsewhere,[4,6] so only its main characteristics are here summarized.

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 4μs) 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 continuously monitored by a non-intercepting 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 a dumping magnet. In order to increase the ratio of the annihilation monochromatic photons with respect to the unavoidable bremsstrahlung background, the photon were collected at ~0.8° with respect to the positron axis, giving a monochromatic flux of ~5 x 10^6 photons per second. The photon spectrum was on-line measured 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 to limit within only a few percent the uncertainty in the photon beam intensity. The collimated and cleaned photon beam had a circular spot (Φ ~4 cm) on the target position.

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

The fission fragments were detected by means of the glass sandwich technique.[20] The used target was natural Au with surface area 50 x 50 mm^2 and thickness 0.1 mm, sandwiched between two glass plates covering all the sample surface. It was struck at 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 negligibly affect the photon beam. In all measurements the same Au sample was irradiated. After irradiation, the glass plates were submitted to the usual procedure of chemical etching and microscope scanning.[20] Both plates of the sandwich were scanned in order to get also information on the fragments' forward-backward asymmetry. The asymmetry resulted weak (1.08÷1.14) and weakly energy dependent, in agreement with the results known from literature:[21] therefore an average of the counts of the two plates of each sandwich was taken in order to obtain results free from any dependence on forward-backward asymmetry. To check the effects of radiation damages in the glass plates and to estimate spurious events due to background contributions, also the glass surfaces not in contact with the target were scanned.

The fission fragments were measured at 20 different positron energies between 120 MeV and 300 MeV. The cross sections per equivalent quantum (shortly called "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), however, 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.

Having taken into account the efficiency of glass plates,[24] the overall error in the normalizing factor turned out to be ± 7%.

The so obtained experimental yields per equivalent quantum, $g(k_m)$, are shown in Fig.1 (full dots) as a function of the maximum photon energy, k_m; in the same figure the experimental data known from literature[10,11,13,15] in the 100÷300 MeV energy range are also shown, with the exception of the data of Vartapetyan et al.[12] since these authors, for photon energies below 300 MeV, used the yields measured by Ranyuk and Sorokin[11] and Mitrofanova et al.[15]

3. Photofission results.

I. Photofission cross section

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

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

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; 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. As fission threshold, we took $k_T = 70$ MeV, as the photofission cross section of Au is expected to be very low at photon energies less than 70 MeV:[12] the estimated systematic error due to the contribution of Au photofission values at photon energies less than 70 MeV was evaluated to be about 30% for the lowest energy point, about 3% for the 120 MeV point, and lower for the other points.

We evaluated the fission cross section for 11 photon energies at intervals of 20 MeV from 100 MeV to 300 MeV. The used unfolding method, applied to the experimental yields, gives a σ_f vector that represents an estimate of the photofission cross section averaged respect to the photon energy by a matrix R, whose meaning is that of an 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 $N(k,k_m)dk$, as well as on the value of a smoothing parameter γ, chosen to "regularize" the $\sigma_f(k)$ solution. The parameter γ was selected by applying a Bayesian method, suggested by Turchin et al.[26] This method allows to calculate the probability density $P(\gamma \mid g)$ of obtaining some γ-values for a fixed set of experimental yields $g(k_m)$. The $P(\gamma \mid g)$ function has a sufficient clear-cut maximum for a number of experimental yields larger than 15, as stated by Turchin et al.[26] In this case 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 in correspondence of which the $P(\gamma \mid g)$ probability assumes 10% of its maximum, which is reached at $\gamma = 4.5$. The rows of the energy resolution R-matrix obtained for $\gamma=4.5$ have similar shapes as those plotted in Fig.4 of Ref.6: they actually have the suitable form of

an energy resolution function with the 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 in using an annhilitation photon beam in order to study photofission of nuclei with high fission thresholds.

The photofission cross section values obtained from the above procedure are reported in Fig.3 (full dots). The errors were calculated by the usual propagation rule: they account for the experimental errors as well as for the auxiliary conditions imposed by the unfolding procedure. The above mentioned uncertainty of about 7% in the normalizing factor should be added. In the figure, all the relevant data for Au photofission measured in previous experiments in the photon energy range covered by this experiment are also drawn. 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 calculated 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 Andersson et al.[13] by using for the bremsstrahlung spectrum the Bethe-Maximon expression,[28] corrected for target and collimator effects.

As shown, all the 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 quasi-monochromatic photons, were derived using on-line measured photon spectra.

II. Fissility

From the measured photofission cross section $\sigma_f(k)$ it is possible to calculate the nuclear fissility P_f through the relation:

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

where $\sigma_{in}(k)$ is the total inelastic cross section. In our energy range, the cross section for photon elastic scattering is very low, and we can safely assume that it is $\sigma_{in}(k) \cong \sigma_T(k)$, 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 $\sigma_T(k)$, ranging from beryllium to uranium. It is worthwile to point out that the values of $\sigma_T(k)$ so obtained are compatible, within the errors, with the $\sigma_T(k)$ values deduced from a recent measurement of (γ,xn) reactions on Au.[30]

In Fig.4 the obtained P_f values of 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 done for the photofission cross section σ_f.

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

In Fig.5 the compound nucleus distributions over the excitation energy E^*, the mass and charge losses, ΔA and ΔZ respectively, are shown, 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 showed that compound nuclei produced by 300 MeV photons impinging on Au, Bi and U nuclei have the same average values of E^*, ΔA, ΔZ (see Tab.1), and very similar distributions.

In Table 2 and Fig.6, 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] For what said above, experimental fission cross sections σ_f and, hence, fissilities are the result of an average over the ensemble of the compound nuclei produced in the reaction.

In the same Table and Figure, 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.6 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 level

density parameters of the nucleus with equilibrium deformation and the nucleus which has a configuration corresponding to the fission saddle point was taken equal to 1.02. Moreover, we neglected shell effects, since they vanish at excitation energy $E^* > 30$ MeV.[34]

The agreement found between experimental results and theoretical calculations over the explored Z^2/A range is remarkable.

In principle, the analysis of data on nuclear fission by intermediate energy photons in the framework of the cascade-evaporation model allows to investigate effects such as: spread in composition and excitation energy of compound nuclei, thermal disappearance of shell effects, dependence of fission barrier heights on the excitation energy, proper choice of the liquid drop model parameters, and so on.[18] In order to drawn more definite conclusions one need 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 emitted particles. Moreover, one should expand the region of energies, probes, and Z^2/A explored. From the theoretical point of view, detailed calculation of still more sophisticated cascade-evaporation models are requested.

5. Conclusions

Here we summarize our main results and conclusions.

a) We determined the photofission cross section of natural Au in the energy range $100 \div 300$ MeV by taking advantage of a quasi-monochromatic photon beam (Fig.3).

b) The nuclear fissility P_f was calculated using the recently measured total photoabsorption cross sections (Fig.4).

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.5).

d) The fissilities from targets at the same excitation energy were compared with the intranuclear cascade calculation results (Fig.6) and a satisfactory agreement between the calculated and measured fissility values was found in the explored Z^2/A range.

Acknowledgements

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Tab.1 Characteristics of the compound nuclei produced in the interaction of 300 MeV photons with different target nuclei. The calculations have been performed with the Monte Carlo cascade-evaporation code.

Target	$<E^*>$ (MeV)	$<\Delta A>$	$<\Delta Z>$
^{97}Au	97	2.03	0.67
^{209}B	97	1.99	0.60
^{238}U	101	2.00	0.63

Tab.2 Fissilities 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 for 300 MeV photons.

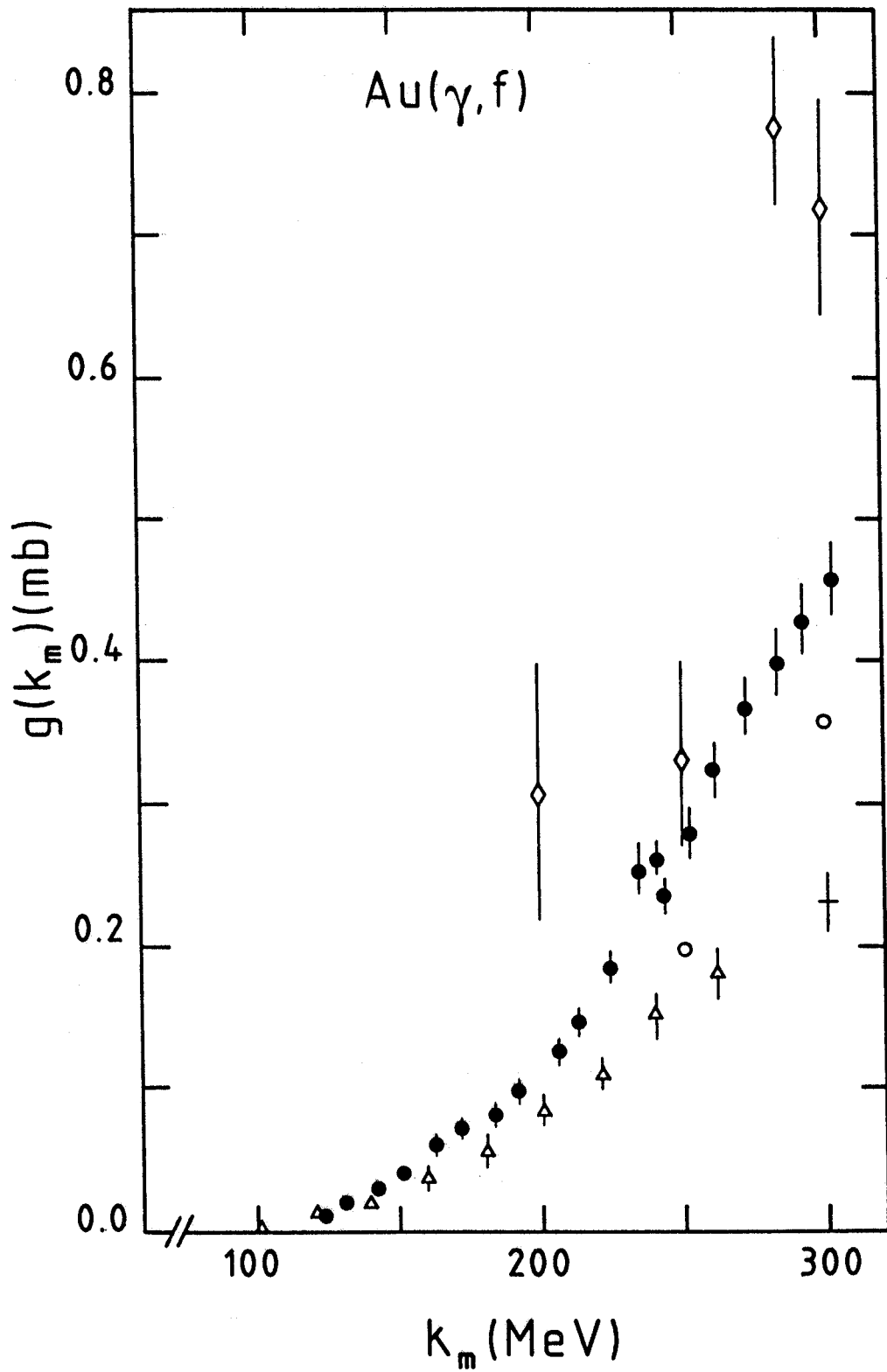| Target	P_f(exp.)	P_f(theory)
^{197}Au	$(1.85\pm0.35)\times10^{-2}$	1.4×10^{-2}
^{209}Bi	$(1.05\pm0.30)\times10^{-1}$	1.5×10^{-1}
^{238}U	$(8.70\pm1.30)\times10^{-1}$	7.8×10^{-1}

Fig.1 Au photofission yields per equivalent quantum vs. the maximum photon energy k_m. •, this experiment; ◊, Ref.10; Δ, Ref.11; o, Ref.13; +, Ref.15. Where not shown, the errors are not deducible from the original papers.
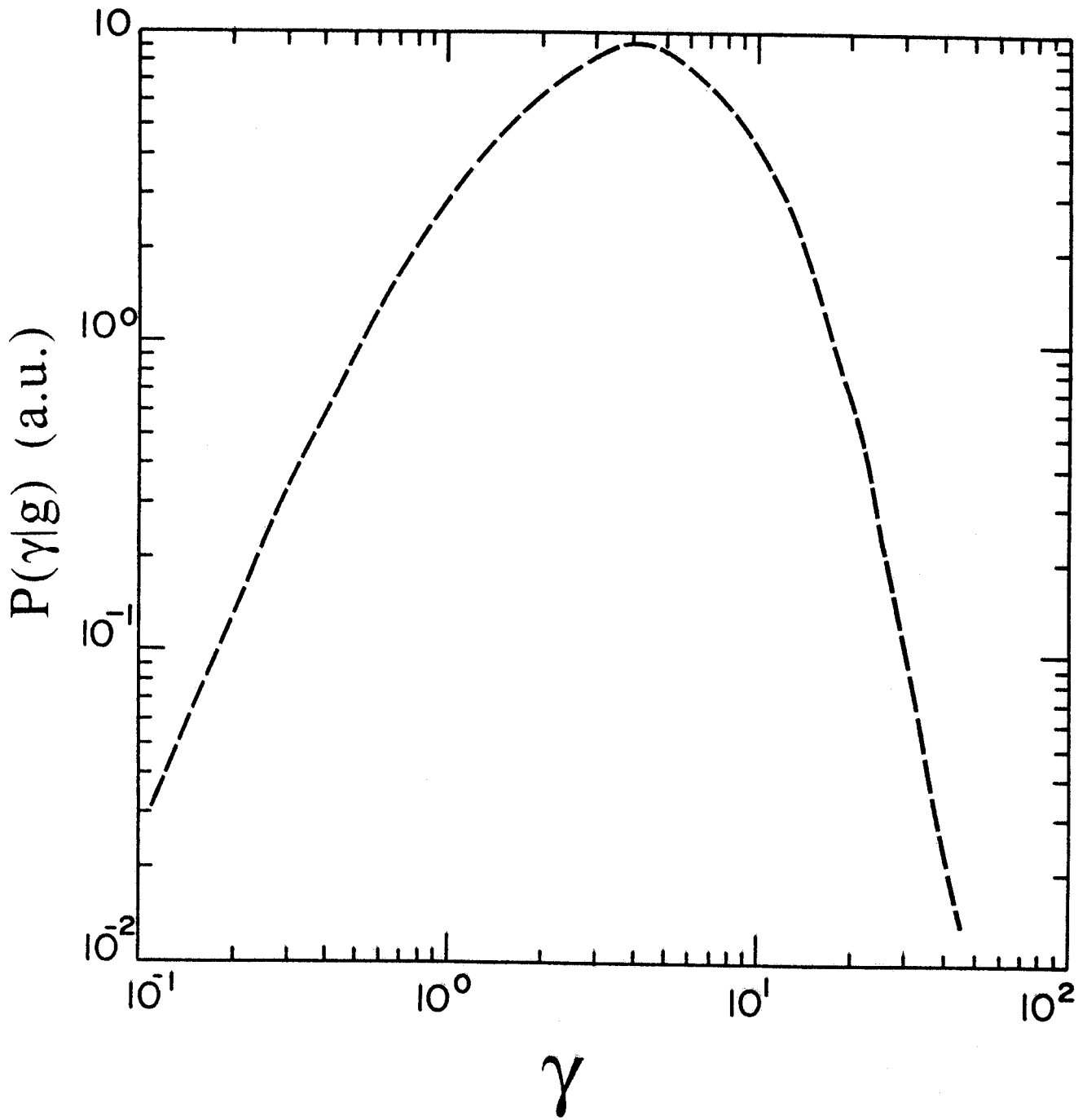
Fig.2 Probability density $P(\gamma\,|\,g)$ as a function of the smoothing parameter γ.
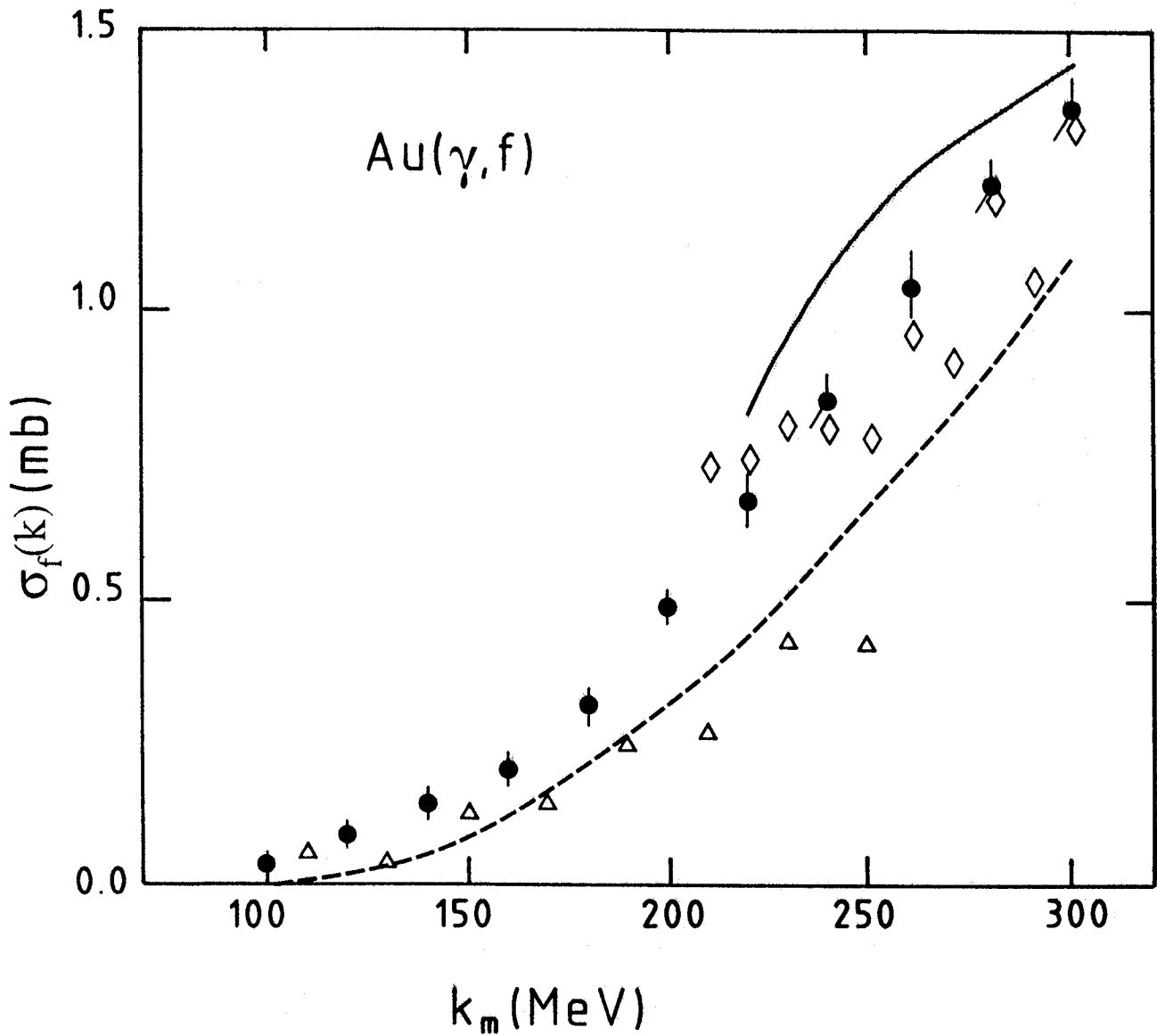
Fig.3 Photofission cross section f(k) of Au vs. photon energy k: •, this experiment; ◊, Ref.10; Δ, Ref.11; the dashed curve represents the results of Ref.12, with the low energy yields taken from Ref.11 and Ref.15; the solid line represents the data of Ref.13 (the shaded area gives an indication of the errors). Where not shown, the errors are not deducible from the original papers.
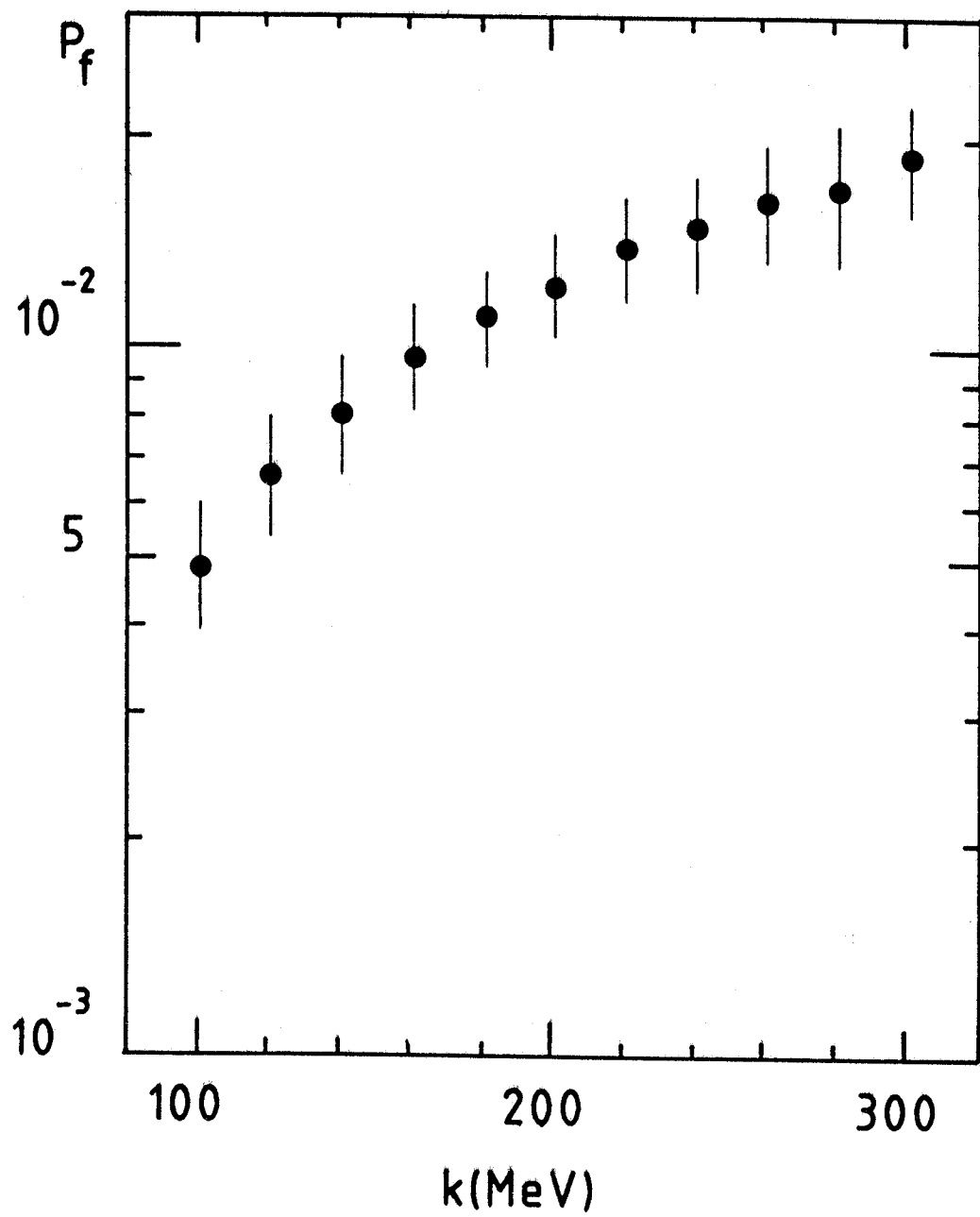
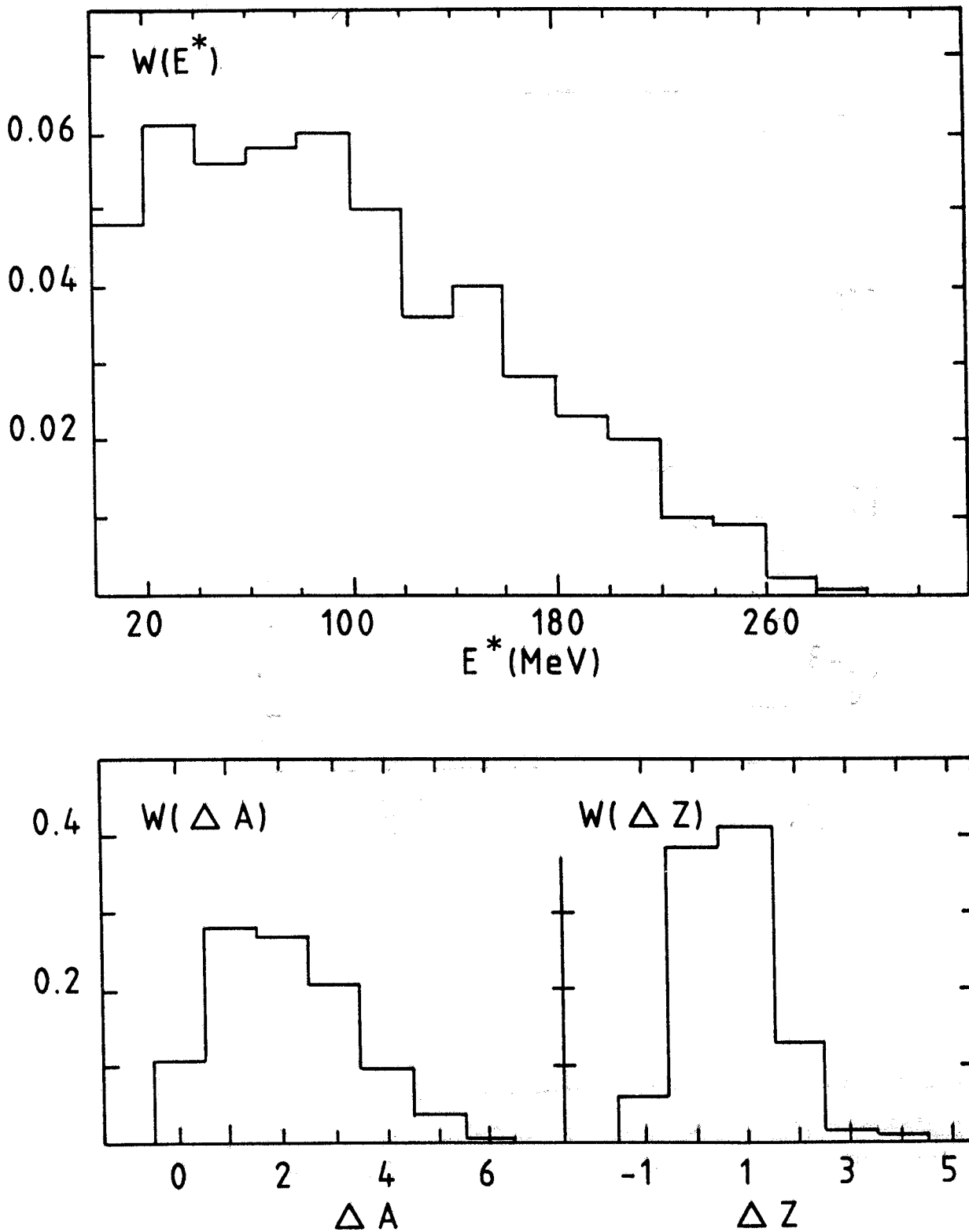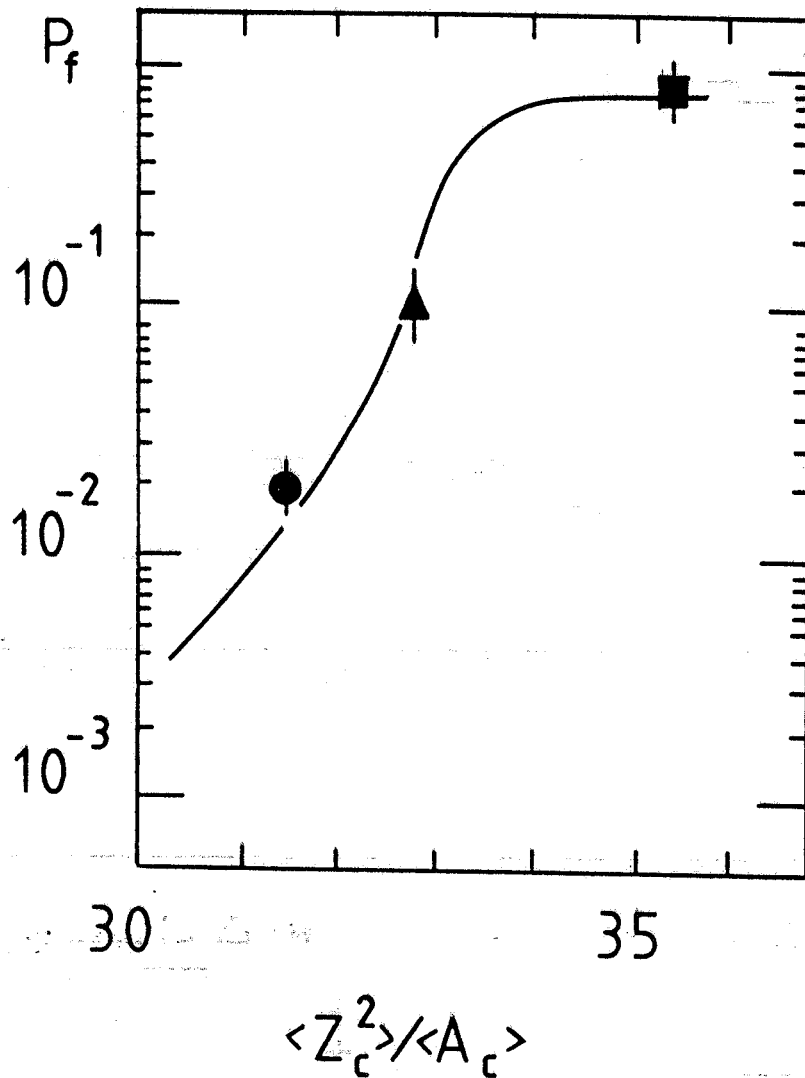Fig.4 Au nuclear fissility data from this experiment vs. photon energy k.

Fig.5 Distributions over the excitation energy E*, 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.

Fig.6 Fissilities values for ^{197}Au ($\bullet$, this experiment) and ^{209}Bi ($\blacktriangle$, Ref.6) excited by 300 MeV photons, and for ^{238}U ($\blacksquare$, Ref.32) excited by 280 MeV photons. The curve refers to the Monte Carlo cascade-evaporation calculation for 300 MeV photons.