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FISSION OF COMPLEX NUCLEI INDUCED BY 52-MeV MONOCHROMATIC AND POLARIZED PHOTONS

by

O.A.P. TAVARES, M.L. TERRANOVA[1], L. CASANO[2],
A. D'ANGELO[2], D. MORICCIANI[2], C. SCHAERF[2],
D. BABUSCI[3], B. GIROLAMI[4], J.B. MARTINS,
E.L. MOREIRA and J.L. VIEIRA

Centro Brasileiro de Pesquisas Físicas - CBPF/CNPq
Rua Dr. Xavier Sigaud, 150
22290 - Rio de Janeiro, RJ - Brasil

[1]Dipartimento di Scienze e Tecnologie Chimiche,
II Universitá di Roma "Tor Vergata", Via E. Carnevale,
00173 Roma, Italy
Istituto Nazionale di Fisica Nucleare-INFN,
Sezione di Roma 2, Roma, Italy

[2]Dipartimento di Fisica, II Universitá di Roma "Tor Vergata",
Via E. Carnevale, 00173 Roma, Italy
Istituto Nazionale di Fisica Nucleare-INFN,
Sezione di Roma 2, Roma, Italy

[3]Istituto Nazionale di Fisica Nucleare-INFN, Laboratori
Nazionali di Frascati-LNF, Casella Postale 13,
00044 Frascati (Rm), Italy

[4]Laboratorio di Fisica dell'Istituto Superiore di Sanitá,
Viale Regina Elena 299, 00161 Roma, Italy
Istituto Nazionale di Fisica Nucleare-INFN, Sezione Sanitá,
Roma, Italy

ABSTRACT- Experimental fissility data from photofission reactions of ^{232}Th, ^{238}U, and ^{235}U nuclei at 52 MeV have been obtained using monochromatic and polarized LADON photon beams and dielectric fission-track detectors. These data, along with literature data for ^{237}Np,^{235,238}U, ^{232}Th, ^{209}Bi, ^{208}Pb and ^{174}Yb nuclei have been analysed within the framework of a simple two-step model for photofission reactions, i.e.,absorption of the incident photon by a neutron-proton pair followed by an evaporation-fission competition mechanism for the excited nucleus. Estimated fissility for nuclei in the Ta-Np region clearly evidentiates shell effects in the vicinity of ^{208}Pb. In the case of ^{238}U the effect of photon polarization on fission direction has been also studied, and isotropy was observed in the fragment azimuthal distribution.

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The influence of shell effects on fissility of nuclei in the vicinity of ^{208}Pb has been studied by us in a previous paper[1] which reports both experimental and calculated results on fission cross section and fissility for a number of nuclei for incident photons of 69-MeV mean energy. For target nuclei in the region of actinides, the photofission cross section has shown almost to exaust the total reaction channel,whereas for nuclei with $A \lesssim 210$ the total fission probability amounts to 10^{-3}-10^{-2} only , reaching values as low as 10^{-8} as one goes towards the lanthanides.

The experiment reported in Ref.1 has been performed by using the monochromatic and polarized LADON photon beams obtained by laser light backscattered against high-energy electrons circulating in the storage ring ADONE (Frascati National Laboratories)[2,3]. The data collected from this experiment as well as those obtained in other laboratories have been interpreted on the basis of a simplified model for the photofission reactions. Following this model the primary interaction is described according to Levinger's quasi-deuteron mechanism of nuclear photoabsorption and is followed by a process of evaporation-fission competition for the excited nucleus. The trend of fissility with parameter Z^2/A calculated for nuclei along the β-stability valley, from silver up to neptunium, has shown structures due essentially to shell effects ,mainly in the region of Z=82, N=126, where the data have been reproduced quite well. Since the model predicts that such structures should occur more clearly at lower nuclear excitation energies, we felt therefore worthwhile to extend our investigation of shell effects by a study at an incident photon energy of 52 MeV.

In the present paper we report the measured photofission cross section and fissility data for some actinide nuclei (^{232}Th, ^{238}U and ^{235}U) at 52-MeV photon energy, together with fissility values calculated according to the previous model for nuclei in the Ta-Np region. No attempts were made to measure photofission cross section for nuclei of mass number $A \lesssim 210$ in view of both the low total fission probability expected for such nuclei at 52 MeV (less than 10^{-3}) and the value of the total dose ($\sim 10^{10} \gamma$'s) which can be attained during the exposures.

The experimental procedure has been described in our previous paper[1], but some details of interest to the present measurements are given as follows. The target materials (0.21-0.30 mg/cm^2 thin films of $^{232}ThO_2$, $^{nat}UO_3$, and 93% - enriched $^{235}UO_3$ oxides prepared on mica sheets as supports) were contacted with a 100 µm-thick makrofol N (Bayer AG) which served as detectors for fission fragments. In the case of thorium the micas themselves were used as fission-track detectors . The total effective number of target nuclei (in units of 10^{18} cm^{-2}) for the different stacks was 3.4, 8.1, and 1.9, respectively, for ^{232}Th, ^{238}U, and ^{235}U. The packs were exposed perpendicularly to monochromatic (with an energy resolution of ~ 10% FWHM) and fully polarized LADON photon beams of maximum photon energy of 59.5 MeV and intensity of 10^5 γ/s. The integral photon doses taken by a large

NaI(Tl) crystal monitor were 3.7×10^9 for ^{232}Th and ^{235}U targets, and 3.4×10^9 for ^{238}U target.The energy spectra continuously taken by a magnetic pair spectrometer did not show significant deviations from the mean energy profile. An effective photon mean energy $\bar{k}$ = 52 MeV has been deduced. The background contribution due to continuous Bremsstrahlung over the entire energy range was estimated to less than 1%. The track-etch detectors were processed and analysed by the methods previously described^1. The data have been treated by considering the geometry of exposure, statistics, counting and registration efficiencies, and fission-track background from both spontaneous (in the case of ^{238}U) and low-energy ($k \lesssim 30$ MeV) photon-induced fission (for all cases studied). The final numbers of photofission tracks were 62±8, 268±19, and 112±11, respectively, for ^{232}Th,^{238}U and ^{235}U. By taking into account the associated mean total efficiencies (89-96%) we obtained the absolute fission cross sections at photon energy $\bar{k}$= 52 MeV .

Results have been reported in Table I (4th column) together with those obtained in other laboratories. The errors indicated represent a combination of statistical plus systematic errors associated with the different quantities directly related to the determination of the cross sections.

The effect of photon polarization on the direction of the fission fragments has been investigated in the case of ^{238}U nucleus by measuring of the azimuthal angle ϕ of the recorded fission fragment tracks. The ϕ -distribution obtained (Fig.1) did not show meaningful anisotropy in ^{238}U- fission under the conditions of the present experiment. This result may be an indication that for incident photon energy of 52 MeV, fission occurred rather after the primary photoabsorption stage of the reaction when the absorbed energy was already completely distributed into the whole nucleus.

Nuclear fissility (total fission probability) has been deduced for the nuclei listed in Table I by taking the ratio σ_f / σ_a^T of the photofission to the total nuclear photoabsorption cross sections. This latter quantity has been evaluated by using Levinger's modified quasi-deuteron model [11], according to which

$$\sigma_a^T(k,A) = L(NZ/A)\ \sigma_d(k)\exp(-D/k)\ , \qquad (1)$$

where σ_d is the total photodisintegration cross section of the free deuteron, NZ is the number of neutron-proton pairs in the nucleus, and D and L are parameters depending on mass number A. A systematic study of total nuclear photoabsorption cross section data by Terranova et al.[12] has shown that the parameter D can be evaluated by $D = 0.72\ A^{0.81}$ MeV . As far as the parameter L is concerned, a very recent re-evaluation by Tavares and Terranova[13] gives $L = 6.8 - 11.2\ A^{-2/3} + 5.7\ A^{-4/3}$. Taking σ_d=0.152 mb for the cross section of free deuteron photodisintegration induced by monochromatic photons of 52 MeV[14], we evaluated the σ_a^T-values and, accordingly, deduced the fissility values for the nuclei under study. These data are reported in Table I (3rd and 5th columns, respectively).

A comparison between each other fissility data shows that quite good agreement is found in the case of ^{235}U. The experimental results for ^{238}U are scattered within the range ~0.6-1.8, but the weighted mean value of 0.9±0.1 from all quoted results indicates that the photofission reaction almost exaust the total reaction channel. The large differences observed in the fissility data in the case of ^{238}U may be ascribed to rather different experimental procedures (photon sources,target preparation, and detection methods) used in such measurements as well as to some systematic errors, the origin of which is difficult to detect. For ^{232}Th, the result of the present work differs from the interpolated value of the data reported by Leprêtre et al.[5] by ~30% (a weighted mean fissility of 0.46±0.04 is found in this case). Some disagreement is noted also when the results for ^{209}Bi are compared with each other, but all measurements lead to fissility data of the same order of magnitude, giving a weighted mean value of $(1.06\pm0.08)\times10^{-3}$. Finally, the results quoted for ^{208}Pb show a difference of ~ 35-54% when compared with each other.

Experimental fissility data listed in Table I have been compared also with values calculated for incident photons of 52 MeV by applying the previously discussed two-step model for photofission reactions[1]. The calculation has been extended, in a systematic way, to nuclei located along the β-stability valley from Tantalum up to Neptunium. A detailed description of the model, also with the values of the different physical quantities involved in the calculations, has been presented in Ref.1. As remarked before, the present calculation does not apply to preactinide nuclei, since for these nuclei the lack of experimental data prevents one from evaluating the ratio a_f/a_n of the level density parameter at the fission saddle point to that of the residual nucleus after neutron evaporation.

When comparing calculated with experimental fissility values one observes that the calculated ones as a whole fit the experimental data within a factor of 2, which can be considered a satisfactory agreement (a exception is found in the case of ^{174}Yb nucleus, for which a discrepancy of about two orders of magnitude is noted). Substantial agreement is indeed found for various fission cases studied such as ^{237}Np, ^{235}U, ^{238}U (Refs.4,5), ^{232}Th (Ref.5), and ^{209}Bi (Ref.6). Deviations are noted, on the contrary, for ^{238}U (Refs.6,7 and this work), ^{232}Th (this work), ^{209}Bi (Refs.8,9), and the data quoted for ^{208}Pb.

Fig.2 reports all fissility data plotted as a function of Z^2/A. It is seen that at incident photon energy (excitation energy) of 52 MeV the calculated fissility extends over six orders of magnitude in the Ta-Np region. Calculated fissilities at 69 (previous work[1]) and 52 MeV (present work) rather coincide in the region of actinides, where for the heavier nuclei fissility is found close to unity, in agreement with experiments. As expected, remarked differences (one order of magnitude on the average) appear when comparing the trends of fissility obtained at 52 and 69 Mev for nuclei of $Z^2/A \lesssim 33.3$. For target nuclei of A< 210 both trends exhibit similar structures, particularly in the region of Z=82, N=126, where shell effects are clearly manifest. In addition, such effects are seen indeed more pronounced at the lower excitation energy.

In concluding this paper we would stress the fact that, in spite of a limited number of fissility data obtained from photofission experiments at 52 MeV, and of their uncertainties, results reported here clearly indicate the presence of shell effects in fission of nuclei in the region around the Z=82, N=126 shell closures. For actinide nuclei both experimental and calculated results give a total fission probability close to unity and which seems to be independent of excitation energy in the quasi-deuteron region of nuclear photoabsorption. The present results, together with those achieved in the previous experiment carried out at 69 MeV, seem to indicate that the proposed mechanism of photo-interaction is valid in describing photofission reactions in the energy range considered.

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FIGURE CAPTIONS

Fig. 1- Azimuthal angle distribution of ^{238}U fission fragments obtained from normal incidence of a polarized and monochromatic photon beam of 59.5-MeV maximum energy (photon beam energy of 52 MeV).

Fig. 2- Nuclear fissility plotted against parameter Z^2/A. Experimental results (points) obtained at 52-MeV photon mean energy are those as reported in Table I: ■, ^{237}Np and ^{238}U (Ref.4); ○, ^{235}U, ^{238}U, and ^{232}Th (Ref.5); △, ^{238}U, ^{209}Bi, and ^{208}Pb (Ref.6); ▼, ^{238}U (Ref.7); □, ^{209}Bi (Ref.8); ▽, ^{209}Bi (Ref.9); ▲, ^{209}Bi (D.Türck et al., quoted in Ref.9); ◊, ^{208}Pb (Ref.10); ●, ^{235}U, ^{238}U, and ^{232}Th (this work). The broken lines connect fissility values estimated at 52 (this work) and 69-MeV (Ref.1) excitation energy. The dashed lines are used for regions of Z^2/A where the ratio a_f/a_n is not known from experiment (for details see text).

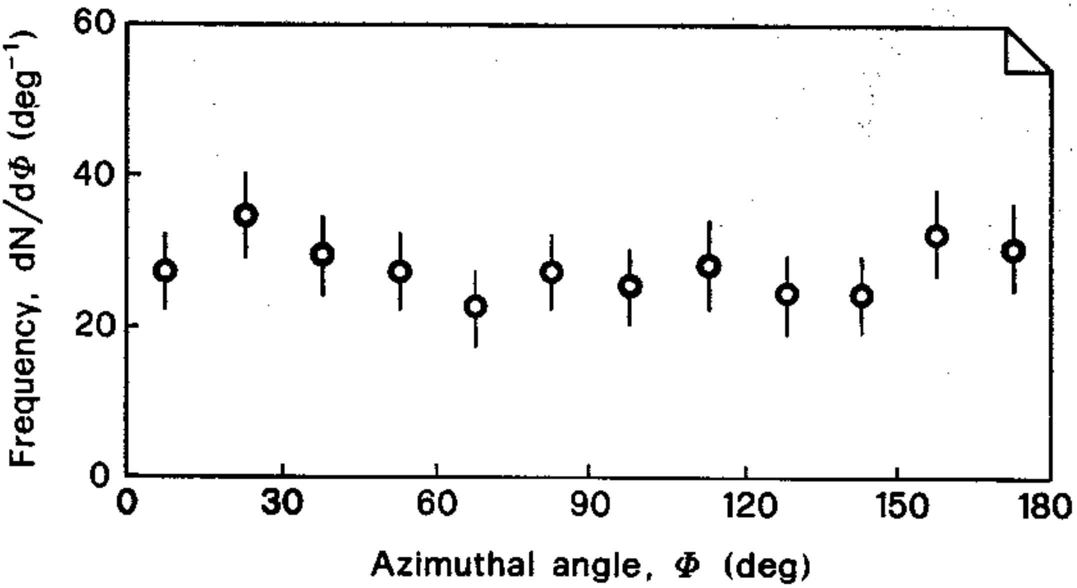
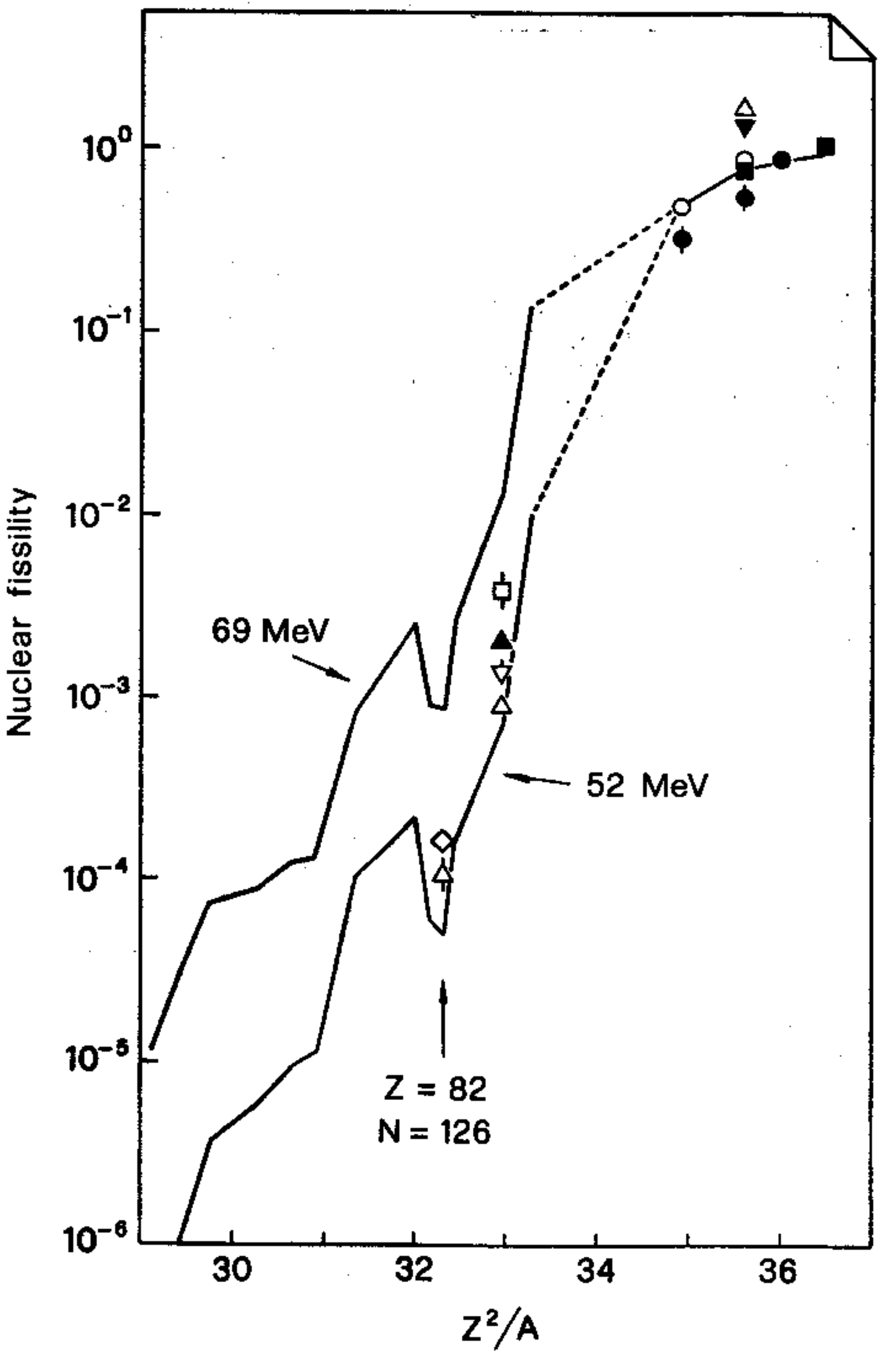

60
40
20
0
0
30
60
90
120
150
180
Frequency, dN/dΦ (deg^{-1})
Azimuthal angle, Φ (deg)

Fig. 1



Nuclear fissility
10⁰
10⁻¹
10⁻²
10⁻³
10⁻⁴
10⁻⁵
10⁻⁶
69 MeV
52 MeV
Z = 82
N = 126
30
32
34
36
Z²/A

Fig. 2

TABLE I - Absolute photofission cross section and fissility at 52-MeV photon mean energy.

Target nucleus	Z^2/A	Total nuclear photoabsorption cross section σ_a^T (mb)	Photofission cross section σ_f (mb)	Nuclear fissility	
				exp.	calc.
^{237}Np	36.49	17.6	20±2[a]	1.1±0.1	0.97±0.01
^{235}U	36.02	17.6	16±4[b]	0.91±0.23	0.85±0.04
			16±2[c]	0.91±0.11	
^{238}U	35.56	17.5	16±2[b]	0.91±0.11	0.81±0.05
			32±2[d]	1.8±0.1	
			14±2[a]	0.80±0.11	
			25±3[e]	1.4±0.2	
			10±1[c]	0.57±0.06	
^{232}Th	34.91	17.5	8.6±0.6[b]	0.49±0.03	0.55±0.09
			6±1[c]	0.34±0.06	
^{209}Bi	32.96	17.4	$(16\pm1)\cdot10^{-3}$[d]	$(0.92\pm0.06)\cdot10^{-3}$	$(0.72\pm0.21)\cdot10^{-3}$
			$(70\pm12)\cdot10^{-3}$[f]	$(4.0\pm0.7)\cdot10^{-3}$	
			$(24\pm3)\cdot10^{-3}$[g]	$(1.4\pm0.2)\cdot10^{-3}$	
			$36\cdot10^{-3}$[h]	$2.1\cdot10^{-3}$	
^{208}Pb	32.33	17.3	$(1.9\pm0.3)\cdot10^{-3}$[d]	$(1.1\pm0.2)\cdot10^{-4}$	$(0.50\pm0.14)\cdot10^{-4}$
			$3.0\cdot10^{-3}$[i]	$1.7\cdot10^{-4}$	
^{174}Yb	28.16	16.7	$(3.2\pm0.5)\cdot10^{-5}$[d]	$(1.9\pm0.3)\cdot10^{-6}$	$(2.3\pm0.7)\cdot10^{-8}$

[a] extrapolated value from data of Ref.4;

[b] interpolated value from data of Ref.5;

[c] this work;

[d] Ref.6;

[e] Ref.7;

[f] Ref.8;

[g] interpolated value from data of Ref.9;

[h] D. Türck et al., quoted in Ref.9;

[i] Ref.10;

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*Permanent address: Conselho Nacional de Desenvolvimento Cientifico e Tecnologico-CNPq, Centro Brasileiro de Pesquisas Fisicas-CBPF, Rua Dr.Xavier Sigaud 150, 22290 Rio de Janeiro-RJ,Brazil.

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