

NOTAS DE FÍSICA

VOLUME XII

Nº 7

PHOTONUCLEAR REACTIONS ABOVE THE MESONIC THRESHOLD
GAMMA NEUTRON REACTIONS IN IODINE AT ENERGIES BETWEEN
300 AND 1000 MeV

by

V. di Napoli, F. Dobici, F. Salvetti and H. G. de Carvalho

CENTRO BRASILEIRO DE PESQUISAS FÍSICAS

Av. Wenceslau Braz, 71

RIO DE JANEIRO

1966

PHOTONUCLEAR REACTIONS ABOVE THE MESONIC THRESHOLD
GAMMA-NEUTRON REACTIONS IN IODINE AT ENERGIES BETWEEN
300 AND 1000 MeV

V. di Napoli, F. Dobici, F. Salvetti
Laboratorio di Chimica delle Radiazioni
e Chimica Nucleare del C.N.E.N.
Istituto di Chimica Generale ed Inorganica
Universita, Roma

H. G. de Carvalho
Centro Brasileiro de Pesquisas Físicas
Rio de Janeiro

(Received April 28, 1966)

INTRODUCTION

The inelastic interactions between complex nuclei and nucleons with energies in excess of 300 MeV have been extensively studied and the details of these processes are by now well known.¹ However, knowledge of inelastic interactions between complex nuclei and high energy photons is another matter. This field has not yet been explored and the nuclear emulsion results presently available give only partial information. The present paper is part of a systematic work initiated to measure the inelastic interactions of high energy photons with complex nuclei, in detail. We intend

to study the magnitude, energy and mass number dependence of the photonuclear absorption cross-section at energies above the meson production threshold, since this is basic for understanding the interaction of high-energy quanta with complex nuclei.

In this paper we report the (γ, n) , $(\gamma, 2n)$ and $(\gamma, 3n)$ cross sections from iodine for photons of energies between 300 MeV and 1000 MeV. We found the (γ, n) cross-section value unexpectedly high; many times what was anticipated. In fact it exceeds in value the total inelastic cross-section calculated by the photo-meson optical model.

Phenomenologically, photonuclear reactions can be roughly grouped into three energy ranges. The "giant-resonance" process accounts for most of the (γ, n) and $(\gamma, 2n)$ reactions. In this energy range (5 to 25 MeV), the photons interact with the dipole moment of whole nuclei and the resulting nucleon emission can be described by means of a statistical or evaporation process. Due to the Coulomb barrier, neutron emission at these low energies is some 100 times more frequent than photon ejection and consequently the (γ, p) reaction in this energy range may be considered negligible. The "quasi-deuteron" process, in the energy range between the "giant-resonance" and the mesonic threshold, is such that the photons appear to interact mainly with sub-nuclear units, especially with the n-p pairs or "quasi-deuterons". This model was first proposed by Levinger². The predicted features of the "quasi-deuteron" model are well confirmed by experimental results. The so-called "mesonic-

region" is the energy region above 150 MeV; at these energies the photon interaction is believed to come about through a mesonic effect and usually it gives rise to a multineutron and multi-charge ejection process. The interaction takes place through a mechanism which can account for the multi-prong stars observed in nuclear emulsions. Miller ³, Kikuchi ⁴, George ⁵, Peterson and Roos ^{6,7} and Castagnoli et al. ⁸, observed stars in nuclear emulsions formed by bremsstrahlung of various energies up to 1125 MeV. Star-prong spectra for all emulsion elements, as a function of bremsstrahlung energy, were found. The yield of stars with two, three or more prongs, as a function of bremsstrahlung energy, rises sharply at the meson creation threshold. This suggests that above the meson threshold, a large proportion of stars with two, three, or more prongs are produced by a mesonic process. Kikuchi believed that for bremsstrahlung energies greater than 150 MeV most of the one and two-prong stars are produced by photons of energy less than 150 MeV.

The following mechanism of star formation was proposed by Ref ⁹ to explain the rapid rise in the yield of photostars with one or more prongs at photon energies above meson threshold. A photon interacts with a single nucleon in the nucleus to produce a real or virtual meson. The meson is absorbed by two nucleons of the same nucleus. The two nucleons which absorb the meson are ejected from the nucleus with great energy, leaving an excited nucleus. The nucleus is further excited if one or both of the nucleons which absorb the meson interact after absorption

with the other nucleons. The excited nucleus may then emit one or more particles.

Roos and Peterson have proposed an "optical model" similar to that of Reff along the following lines: the cross section for star formations is expressed by

$$\sigma_{\text{star}}(E) = A \sigma_{\pi}(E) P_a(A, E) \quad (1)$$

where $\sigma_{\pi}(E)$ is the total photomeson production cross-section per nucleon at the particular photon energy E , assuming that the photomeson cross-section from the neutron is very close to the total photomeson cross section from the proton. The $\sigma_{\pi}(E)$ per nucleon as obtained by assuming all the yields with appropriated weighing factors. The Fermi momentum of nucleons within complex nuclei can transform a high energy photon in laboratory system into a photon of very different energy in the rest system of the nucleon. Therefore it is necessary to average-over the spread of "effective photon energies" allowed by the Fermi motion of the nucleons. In the total cross-section curve, this straightens out considerably the bumps due to the effect of meson resonances, yielding a much smoother curve. The average value of σ_{π} between 300 MeV and 1000 MeV is 268 μbarns (See fig. 1).

Since high-energy photons have long free paths for interaction in nuclear matter, one assumes that all nucleons are equally probable sources of photomesons and therefore the $\sigma(\text{star})$ is proportional to the mass number A . Roos and Peterson's model⁷ visualized real pions emerging from random points within the nucleus; a situation quite different from pions produced within

complex nuclei by strongly interacting particles. In the optical model, $P_a(A, E)$ represents the probability of star production following the meson production and depends on the following: a) the scattering or absorption of a pion formed in the nucleus so as to eject at least two charged particles; b) the scattering of the recoil nucleon from the meson formation, giving rise to the ejection of a charged particle, or particles, together with the charged meson not absorbed and c) multiple photoproduction of charged-pions plus the possible interactions leading to charged particle ejection. This probability P_a may be expressed in the following way: $P_a = 1 - T_\pi T_n$, where T_π and T_n are the nuclear matter transparencies for pions and recoil nucleons. T_π and T_n , being functions of the energy of each particle and of the atomic mass A of the target nucleus and therefore functions of the photon energy E , may be obtained from the Monte Carlo calculations of Metropolis et al.,¹⁰ on pion-and-nucleon-initiated cascade. (See fig. 2.) Roos and Peterson have doubled the transparencies because in their model the photon-mesons originate uniformly within the nucleus.

Cross-section of gamma-neutron reaction produced by X-rays with energies between 300 MeV and 1000 MeV in Iodine

For comparison with nuclear emulsion results as well as for the sake of simplicity, one may divide the inelastic cross-section σ_i in three parts: σ_0 , the (γ, xn) part, corresponding to the zero-prong stars in nuclear emulsion, σ_1 , the (γ, p, xn)

corresponding to the one-prong star and the spallation cross-section σ_s , corresponding to σ (star).

$$\sigma_i = \sigma_o + \sigma_1 + \sigma_s$$

In 1953 Jones and Terwillinger measured the photoneutron production excitation function from 13.5 MeV to 320 MeV. Neutron yields from all reactions were detected at 90 degrees to the beam by a boron trifluoride long counter. This method has one inconvenience; it does not allow identification of the reaction from which neutrons have been emitted, therefore it is improper for the measurements of the (γ, n) reaction cross-sections but gives an upper limit for them. Jones and Terwillinger¹¹¹ explained the high cross-sections (per neutron) which they found, by assuming a quite high neutron-multiplicity per absorbed photon and by assuming that the photon interacts with the nucleus via meson production. The subsequent meson absorption by nuclear matter would lead to high excitation energies, the absorption by uranium of one 140 MeV photon being capable of causing an evaporation of 9 neutrons.

The (γ, n) reaction cross-section for photons of energies above the mesonic threshold has always been presumed to be very small. Consequently the great majority of the (γ, n) events produced by high energy bremsstrahlung beams has been attributed mainly to the giant resonance photons.

From the magnitude of the photofission cross-section of uranium and its fissionability at energies between 300 MeV and 1000 MeV, there is an indication that the inelastic cross-

section for photons of those energies is considerably larger than that calculated from the Roos and Peterson photomesonic optical model. A similar conclusion is reached if one assumes that the (γ, p) reactions in nuclear emulsion are not negligible, as believed. If one selects from the published data only those figures above 500 MeV and adds a lower limit value for the (γ, xn) reactions equal to a (γ, p) cross-section, one obtains an inelastic cross-section about three times larger than that calculated by the photomesonic model. From the above-mentioned facts it is clear that there must be a sizeable part of the inelastic cross-section associated with the emission of neutrons and in this paper we will show that the (γ, n) cross-section is much higher than is usually presumed.

The determination of the cross-section by the photon difference method necessitates accurate measurements of the cross-section per equivalent quantum σ_Q at several energies. The radiochemical method is entirely adequate for σ_K measurements of the (γ, n) cross-section reactions. Iodine was chosen because it is monoisotopic and yields, from (γ, xn) reactions, products convenient for the activity measurements regarding mean-lives and the nature of the radioactivity.

Dose Measurement

The bremsstrahlung beam was obtained by the collision of electrons, accelerated to the desired energy, with a thin tantalum target (0,05 cm). The dose measurements were always car-

ried out by means of a quantameter of the Wilson type ¹². The constant used was 4.79×10^{18} MeV/Coulomb, following Gomez et al. ¹³. The beam was "sharply" collimated so that it hit our target with a small cross-sectional diameter (1.8 cm). The possible ion-recombination within the quantameter (in the case of short pulses), was tested by comparing quantameter readings with the radioactivity simultaneously induced in copper disks at various pulse lengths. No detectable effect was found within 0.2%. Intercalibration was also made (for "long" pulses only) between the quantameter readings and those of a pair spectrometer: the comparison turned out to be satisfactory, a given dose being reproducible to better than 1%.

The same pair spectrometer was used for determining to what extent the gamma ray absorption by an interposed thickness of material would alter the quantameter readings. Quantameter and spectrometer data were compared by interposing various thicknesses of the iodine target (between zero and 2 mm). The maximum effect at maximum thickness was 2% at $E = 1000$ and 5% at $E = 300$ MeV. A correction was applied for our standard target thicknesses.

Energy Measurements

To select electron energy, the method of suddenly cutting out the acceleration radiofrequency at different times after the injection, was chosen. The short duration of the pulses did not lead to inconveniences from ion-recombination. Acceleration

time was accurately measured by displaying the ejection pattern given by a scintillator counter in an oscilloscope, and simultaneously triggering a calibrated oscillator whose pulses appeared superimposed on the same screen. The polaroid photograph of this image is the energy measuring device. The time resolution in such photography is about 100 μ sec., which corresponds to 1/200 of the total acceleration time at 1000 MeV. The energy measurement has an error of about 5 MeV at 1000 MeV.

Background Sources

Besides the high-energy photons, the following reaction sources have to be considered.

(i) Neutrons from "extended sources"

A general background of diffused relatively low-energy neutrons, as well as a neutron beam possibly generated in the tantalum electrosynchrotron target and in the collimator, have to be considered as possible sources of spurious (n, xn) events. This effect was investigated by means of uranium-loaded nuclear emulsions used in fission cross-section measurements¹⁴. The total angular aperture of the bremsstrahlung beam we used was 1×10^{-3} rad, so that the density of the fission tracks induced in the plate by the photons was expected to be a very "square" function of the distance from the spot center. Diffused low-energy neutrons, on the other hand, would give rise to a constant background throughout the plate, while neutrons from the collimator would give rise to a "penumbra" around the gam

ma-ray area. The beam profile, as determined by fission counting, did not show an appreciable contribution from such neutrons.

(ii) Secondaries from interactions within the iodine target.

This contribution was shown to be negligible both by a simple calculation and by comparing yields in targets of different thickness.

RESULTS AND DISCUSSION

a) Target preparation

The iodine targets were prepared with about 2g of Merck resublimated iodine located inside a perspex cylinder of 1.8 cm in diameter and 0.2 cm thick. The container was exposed at normal incidence to the bremsstrahlung beam produced by electrons of several energies in the range 0.3 GeV to 1 GeV. The radiochemical procedures and the activity measurements are described in a previous paper ¹⁵.

b) The results are summarized in Table I and in figures 3, 4 and 5, where the cross-sections per equivalent quantum σ_Q of the iodine are plotted as a function of the natural logarithm of the maximum bremsstrahlung energy. The slope of the solid lines through the experimental points represents σ_K . From the yields obtained and recorded in Table I, the calculated σ_K are shown in Table II. Within our experimental errors (15%), the (γ, n) cross-section appears to be constant in the energy range and the $(\gamma, 2n)$ and $(\gamma, 3n)$ cross-sections seem to increase slowly with the energy increase.

The (γ, n) reaction results were obtained by assuming

Table I

Energy in MeV	σ_Q Cross-section per equivalent quantum in 10^{-27} cm^2 as function of the bremsstrahlung peak energy		
	γ, n	$\gamma, 2n$	$\gamma, 3n$
300	195	1	0.21
500	235	5	0.23
700	267	11	0.36
800	270	12.5	0.45
900	280	14.5	0.53
1000	300	30	1.0

From the yields obtained and recorded in Table I, the σ_K calculated are shown in Table II.

Table II

Energy 300 MeV to 1000 MeV	σ_K - Average cross-sections in 10^{-27} cm^2		
	γ, n	$\gamma, 2n$	$\gamma, 3n$
	87.5 ± 8	20 ± 7	$.7 \pm .2$

"square" form for the bremsstrahlung spectrum. The actual "non square" form of the spectrum was taken in account for correction evaluation in the following manner: a numerical integration was made, assuming as correct the cross-section measurements made by Nathans and Halpern¹⁶ for iodine in the giant resonance region. These data were chosen because of their better agreement with the results of Ero and Keszthely¹⁷. The precision of (γ, n) cross-section measurements in the "quasi-deuteron" energy region is poor because of its very small values and for this reason is represented by a dotted line in figure 6. The values for the cross-sections in the "quasi-deuteron" region were taken from Jones and Terwilliger¹¹. The result of the numerical integral for 320 and 1000 MeV, together with the experimental points obtained using the "square approximation" are shown in fig. 6. In this case the actual form of the bremsstrahlung yields a small correction (+ 6 mb) to the cross-section. The numerical integral shows that the σ_K for (γ, n) reaction in the energy range is consistent with the $87.5 \times 10^{-27} \text{ cm}^2$, the corrected value found.

If the (γ, xn) high-energy photon reactions were due only to a meson reabsorption process, one would expect from an estimate of a reasonable neutron multiplicity in iodine derived from meson-capture events, that the $(\gamma, 3n)$ and $(\gamma, 2n)$ reactions cross-sections would be several times larger than the (γ, n) reaction cross-section. In the present work we found that the (γ, n) $(\gamma, 2n)$ and $(\gamma, 3n)$ average cross sections are proportional respectively to 125, 29 and 1. If we assume for the $(\gamma, 2n)$ and

$(\gamma, 3n)$ quantum equivalent cross-sections an energy dependence of the form $\sigma_Q = AE^{\alpha}$, (which fits well the experimental results), the σ_K is proportional to σ_Q . In this case, at 1000 MeV, σ_K for the $(\gamma, 2n)$ and $(\gamma, 3n)$ reactions are respectively 60 and 1 mb. It is therefore very unlikely that the photo neutrons are produced only by a meson reabsorption process; to the contrary, since the (γ, n) is much more abundant, it signifies that there is a frequent process in which the high energy photon gives only a low energy excitation (about 10 to 30 MeV) to the nucleus. The (γ, p) reaction observed in nuclear emulsion also supports this point of view since it shows that more than 90% of the one-prong stars observed in nuclear emulsion have kinetic energy of less than 20 MeV. Counter experiments by Odian et al ¹⁸, Levintal and Silverman ¹⁹ and Barton and Smith ²⁰ also confirm that most of the protons from the (γ, p) are low-energy protons. These results seem to indicate an "evaporative" secondary process. The nature of the mechanism leading to such high cross-section processes is unknown at the moment. The nature of the reaction will be better understood when new measurements, such as those of the (γ, p) cross-section as function of the mass number and of photon energy and also of cross-sections and neutron energy spectra from the (γ, n) reactions, become available.

From a preliminary measurement of the (γ, n) cross-section of carbon and the iodine results, we had our first indication of the mass number dependence of the (γ, n) cross-sections. For comparison, in fig. 7 we have plotted, from the work of Jones and

Terwilliger ¹¹, the total neutron yields for 320 MeV photons as function of the mass number (solid line) and the cross-sections found for carbon (~~2.1~~^{1.2} ± 0.2 mb) and iodine (solid circles). The (γ, n) reactions seem to depend on the mass number proportional to $A^{3/2}$. The measurements of (γ, n) cross-sections from elements with mass number between carbon and iodine and heavier than iodine are necessary for a more accurate determination of the mass number dependence.

The strong (γ, n) cross-section mass number dependence at energies between 300 and 1000 MeV reported in the present paper indicates a rather high (γ, n) cross-section for heavy elements. For uranium, for example, it predicts 220 mbarns. On the other hand, the cross-section for photo-fission of uranium measured in the same energy range by Carbonara ¹⁴ et al., is 74 mb and therefore a lower limit for the inelastic cross-section, obtained by adding only those two parts, is 294 mb. Thus the photofission-ability of uranium at such energies must be lower than 0.25. This result is three times smaller than the results previously obtained by Carbonara ¹⁴ et al.

* * *

Acknowledgements

The authors wish to thank the Machine Group of the Synchrotron Laboratory of the Laboratori Nazionali di Frascati for their assistance during irradiations. H.G. de Carvalho wish to thank the Brazilian C.N.Pq. and C.N.E.N. for help.

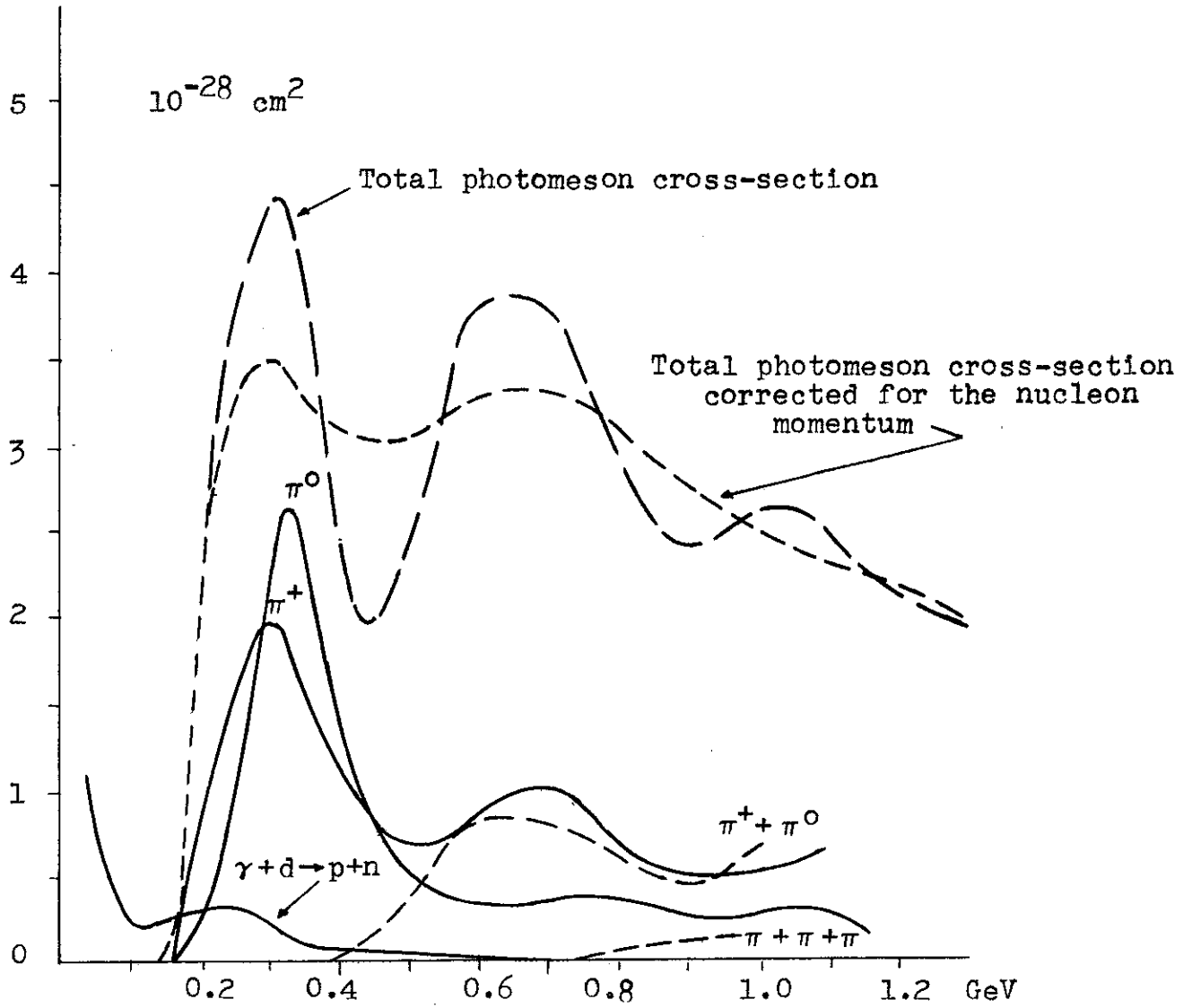


Fig. 1

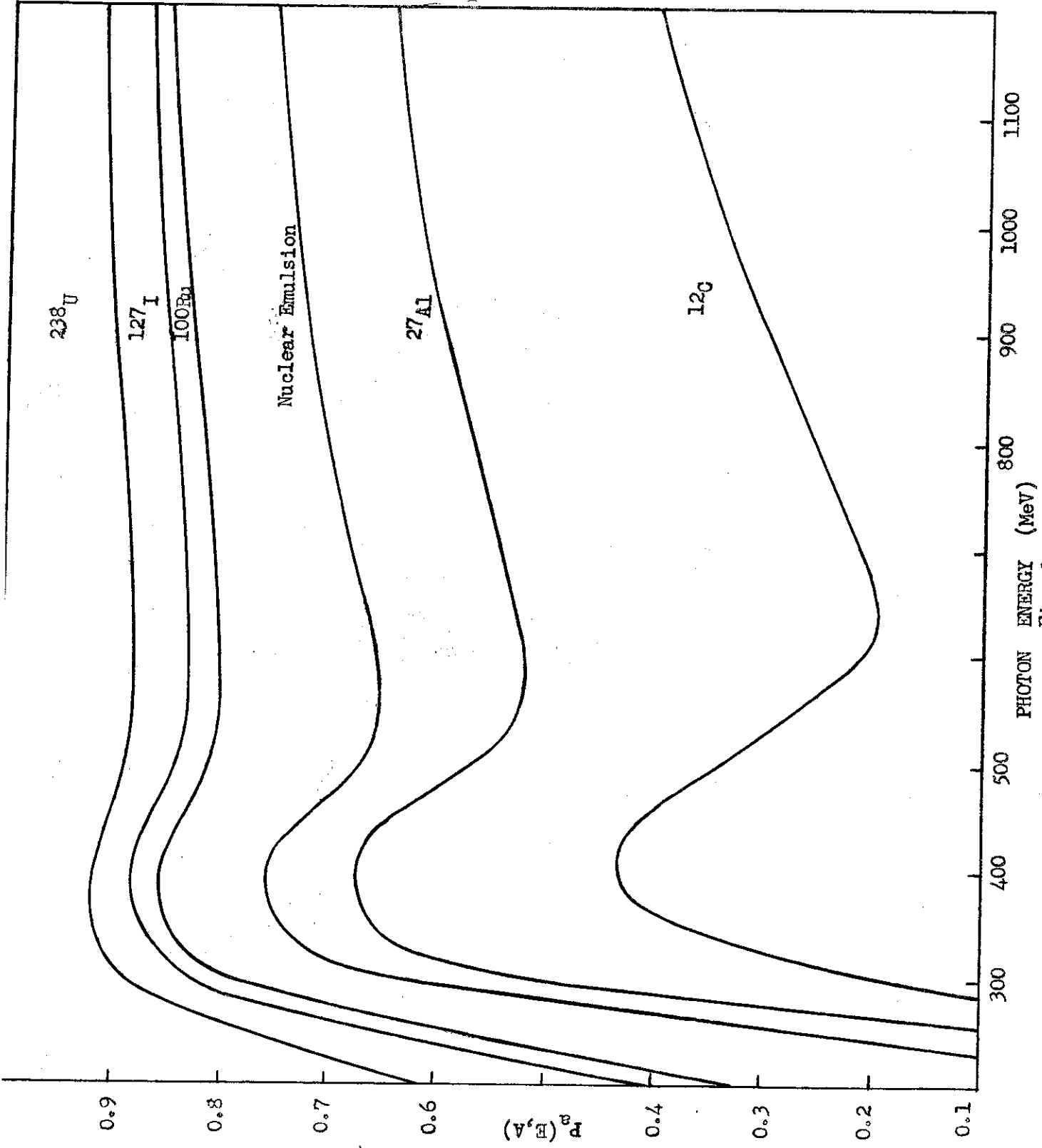


Fig. 2

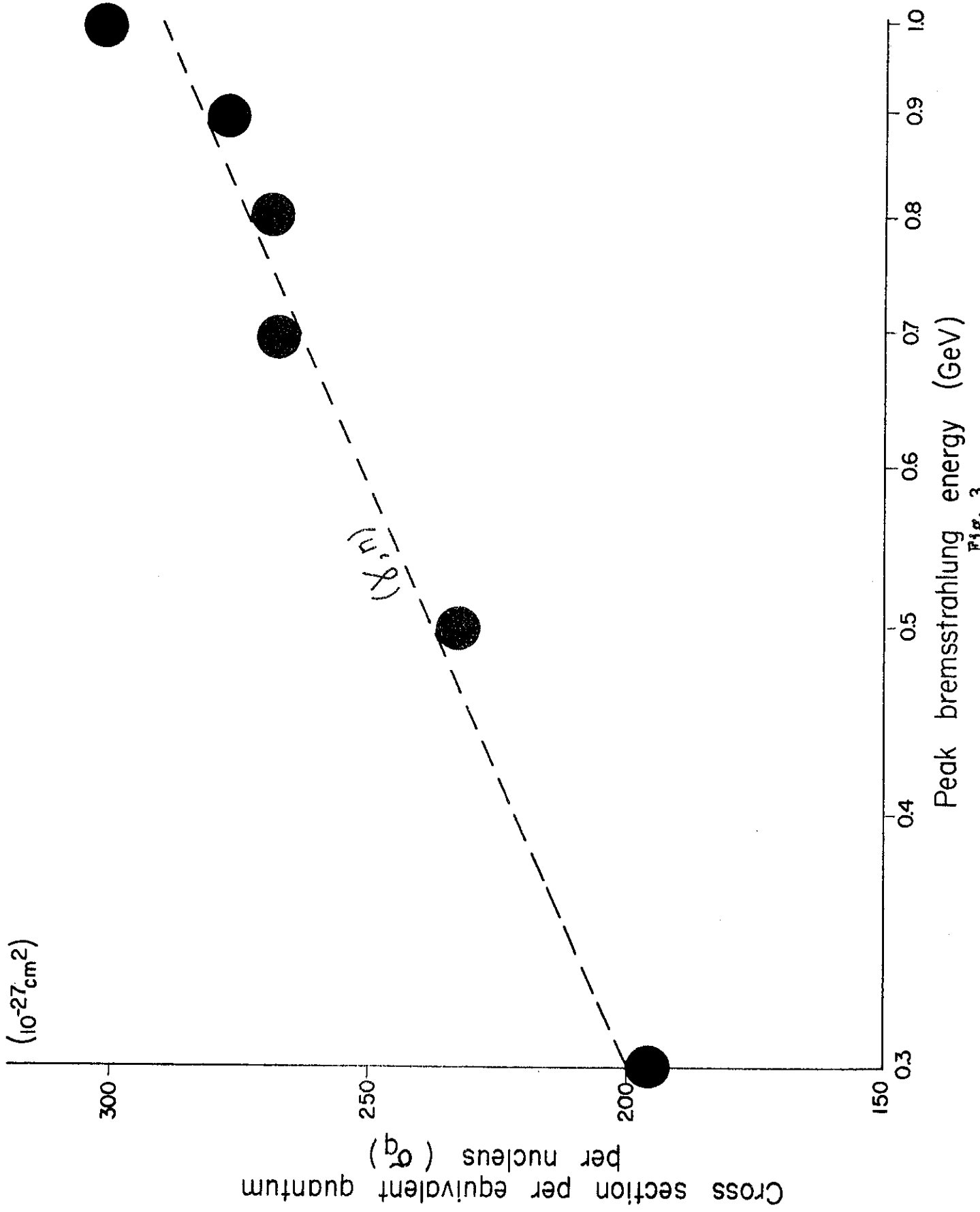
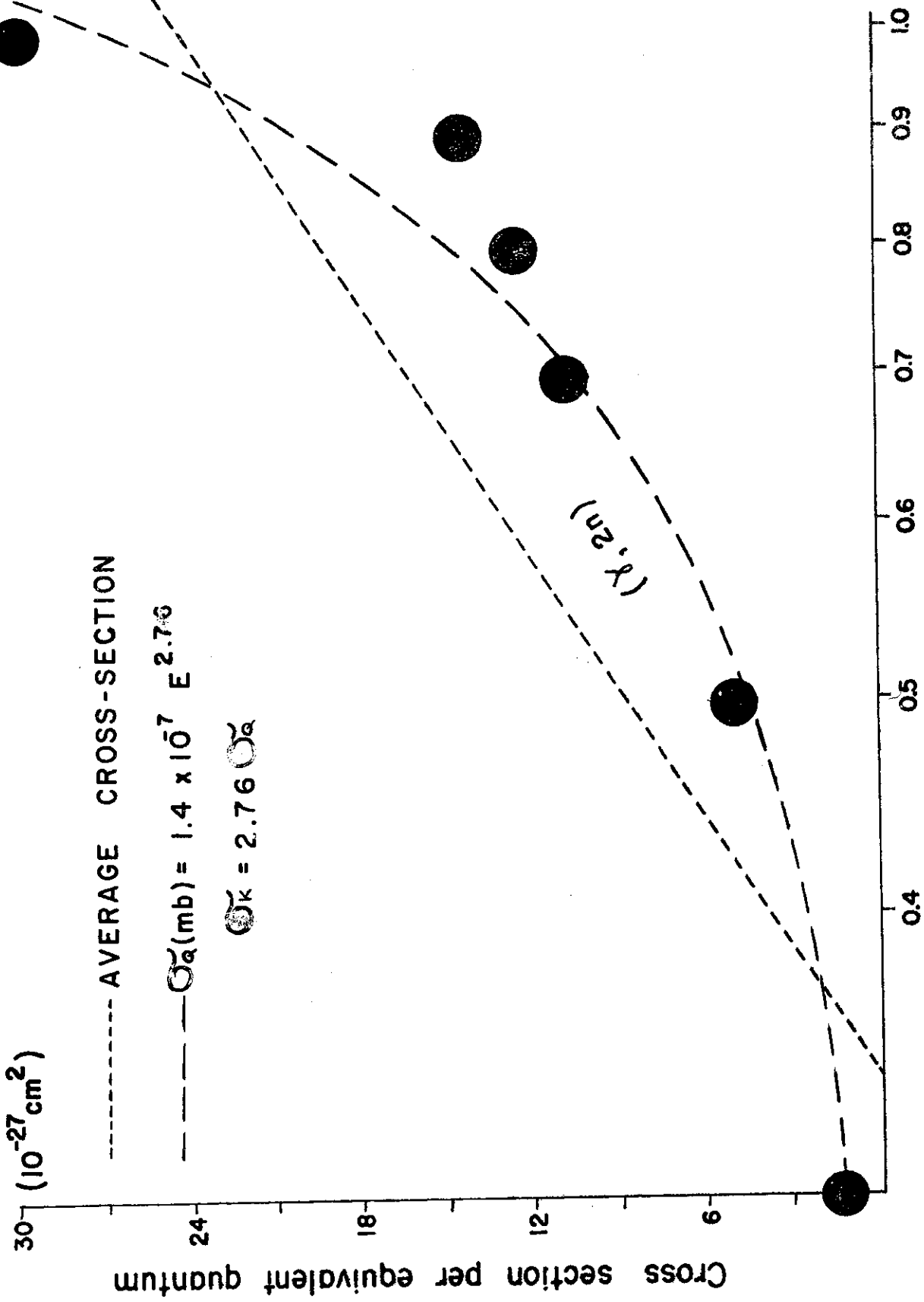


Fig. 3



Peak bremsstrahlung energy (GeV)

FIG. 4

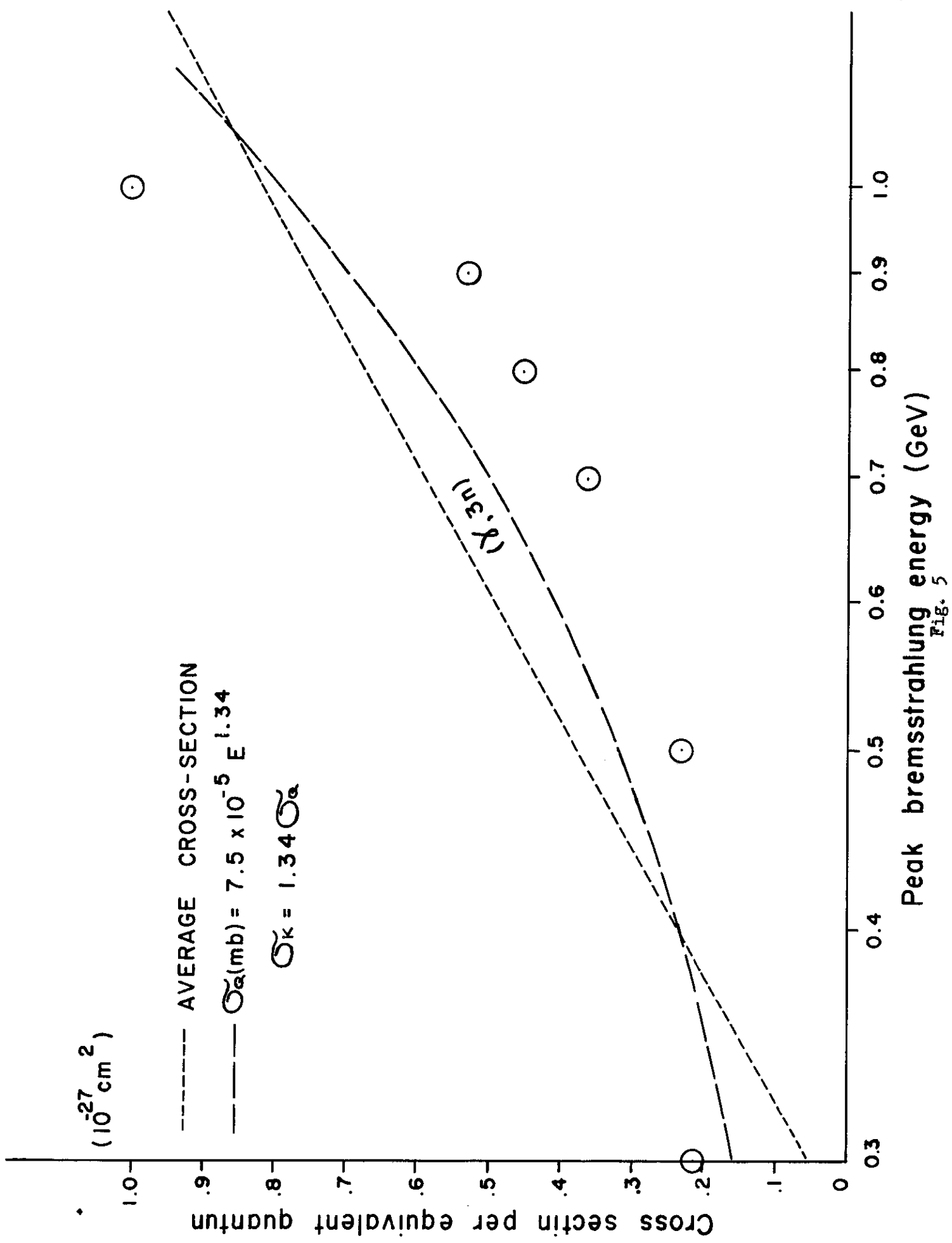


Fig. 5

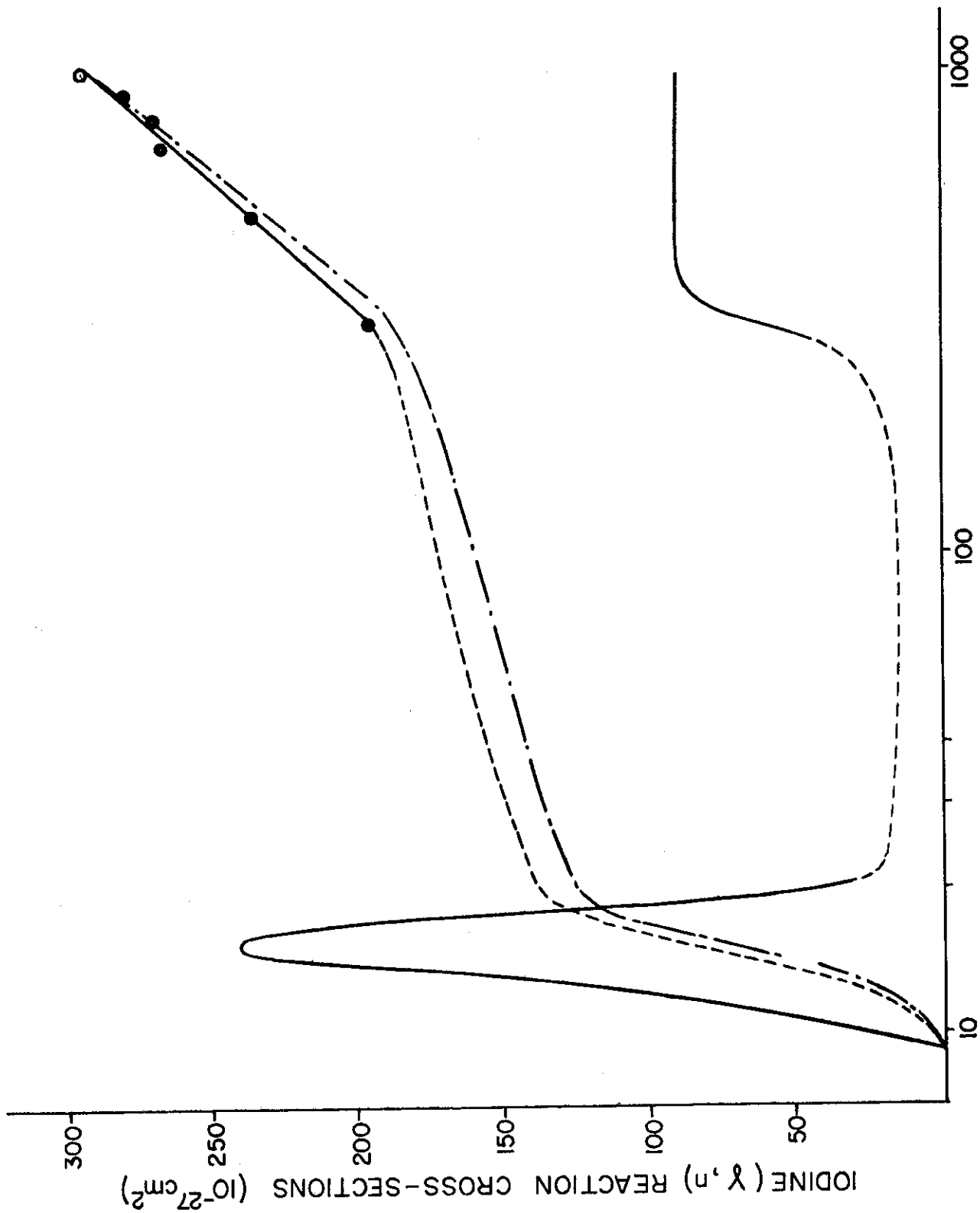


Fig. 6

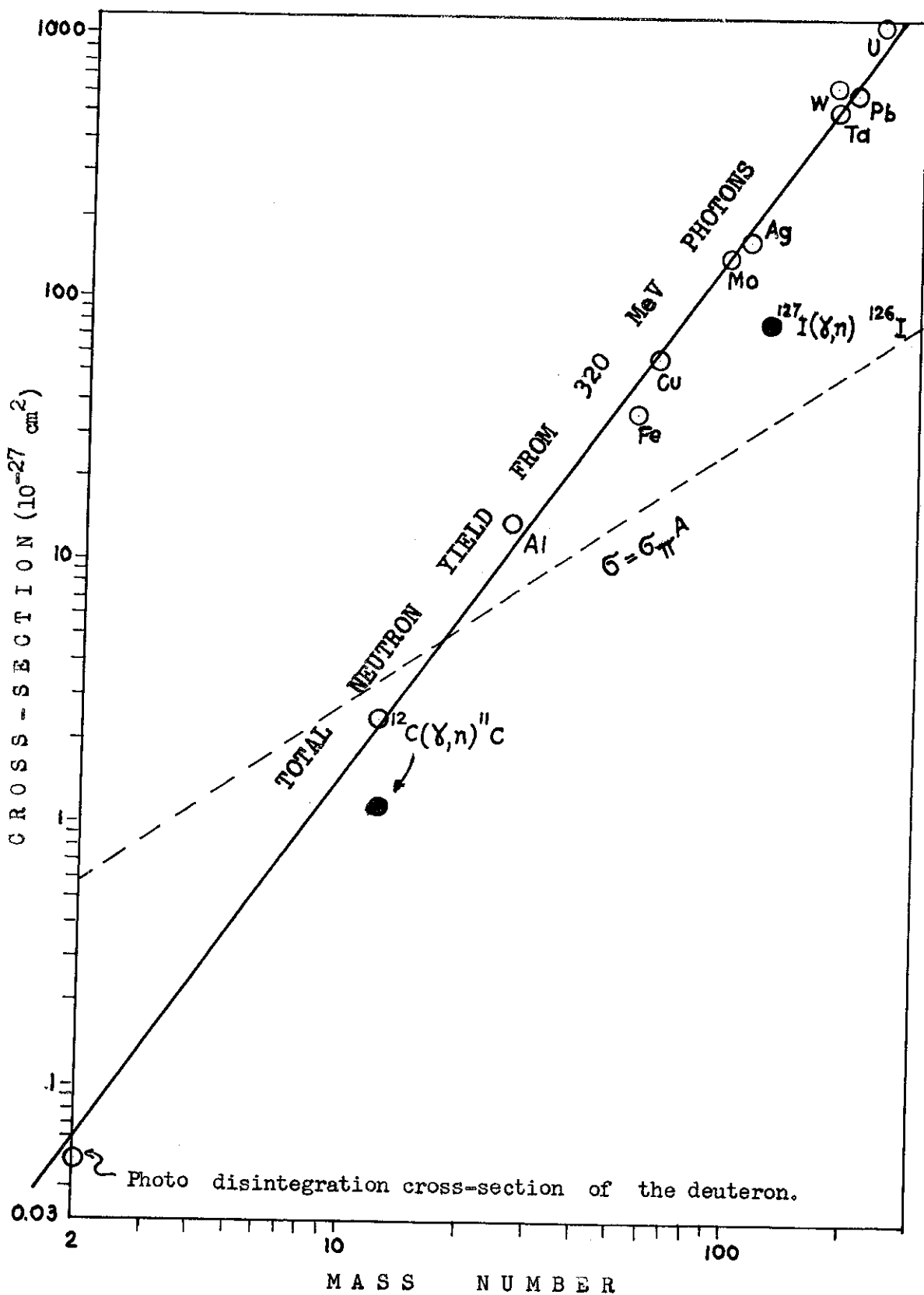


Fig. 7

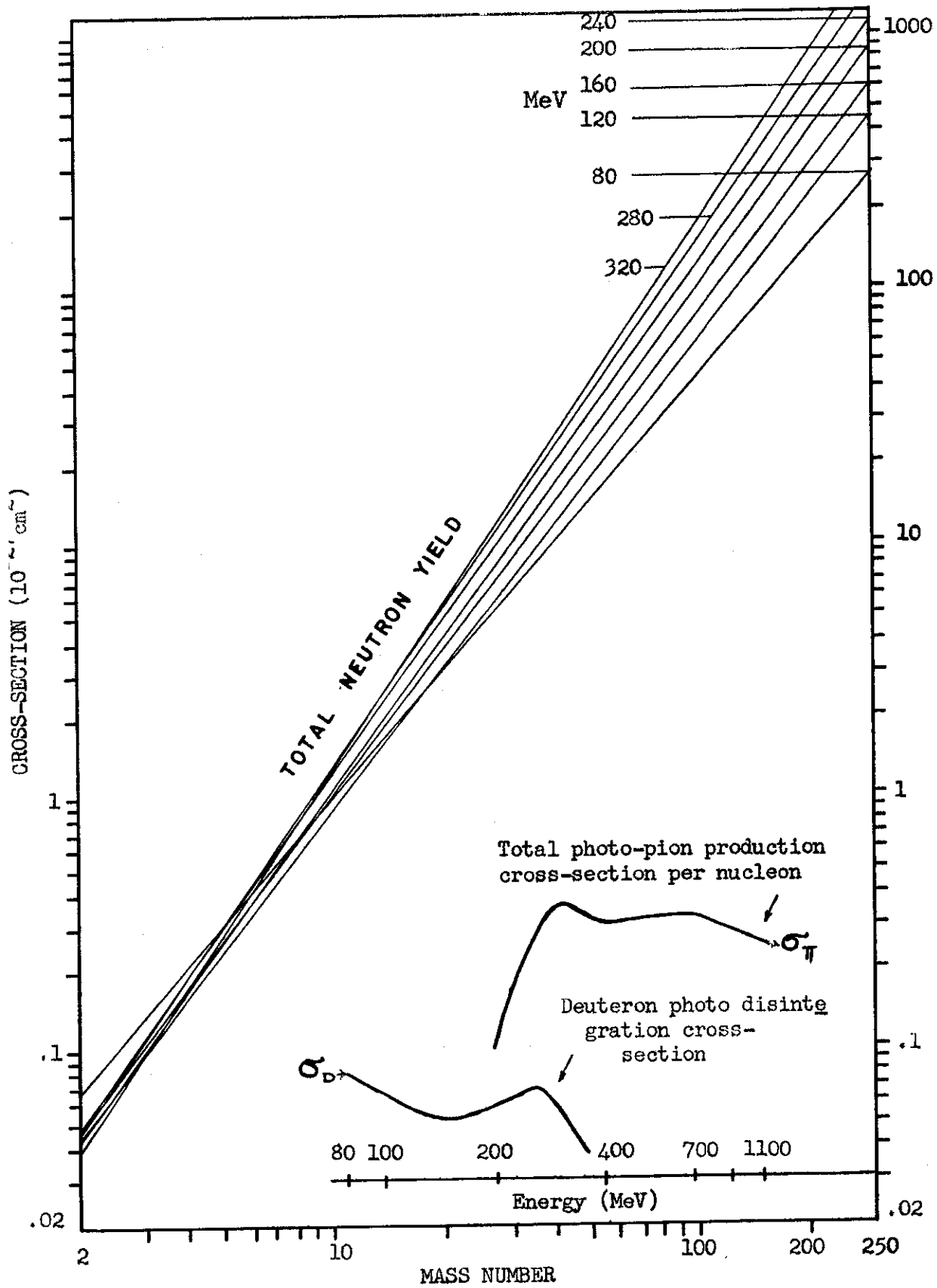


Fig. 8

FIGURE CAPTIONS

- Fig. 1 - Summation of all known high-energy meson photo-processes as functions of photon energy. No nuclear binding effect is included.
- Fig. 2 - Plot of $P(A, E)$ probability as function of the mass number A and photon energy E of nucleus excitation due to meson and/or recoil-nucleon interaction with nuclear matter, for U^{238} , I^{127} , Ru^{100} and nuclear emulsion.
- Fig. 3 - Experimental data for the (γ, n) iodine reaction cross-section "per equivalent quantum" are plotted versus natural log of the peak energy of the bremsstrahlung.
- Fig. 4 - Experimental data for the $(\gamma, 2n)$ iodine reaction cross-section "per equivalent quantum" are plotted versus the natural log of bremsstrahlung maximum energy.
- Fig. 5 - Experimental data for the $(\gamma, 3n)$ iodine reaction cross section "per equivalent quantum" are plotted versus the natural log of the peak energy of the bremsstrahlung.
- Fig. 6 - Iodine (γ, n) cross-sections. Giant resonance data from Nathans and Halpern. Points at 45 and 140 MeV in the "quasi-deuteron" region (dotted line) calculated from the Jones and Terwilliger¹¹ data and adjusted at other energies so as to fit our data in the 300 to 1000 MeV range. The numerical integration, taking into account the actual shape of the bremsstrahlung spectra at 320 and 1000 MeV, is shown to be in substantial agreement with the 87.5 mbarns cross-section found for the (γ, n) reaction in this energy range. Our experimental points are shown for comparison.
- Fig. 7 - Plotting of the total neutron yield from 320 MeV photons¹¹ and of the (γ, n) cross-sections from carbon and iodine (solid circles) as a function of mass number. The dotted line corresponds to the equation
- $$\sigma = \sigma_{\pi} A$$
- Fig. 8 - Plotting of the total neutron yield obtained by Jones and Terwilliger at different energies.