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Photon Induced Nuclear Reactions Above 1 GeV

IV. Mass Distribution in the 6.00 GeV Bremsstrahlung

Induced Fission of Natural Uranium

by

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ABSTRACT

The mass distribution in the 6.00 GeV bremsstrahlung induced fission of natural uranium has been analysed. The measured yields are shown to correspond to at least 90 per cent of the chain yields. The results are compared with other experiments. The mass distribution curve has been decomposed into two distributions, one symmetric and one asymmetric. Finally the peak-to-valley ratio obtained is compared with results at lower irradiation energies. Good agreement is found and from the results above 1 GeV it is deduced that the total fission cross section for ^{238}U decreases rapidly with energy.

1. Introduction

Numerous investigations have been devoted to the study of the mass distribution obtained in the bremsstrahlung-induced fission of ^{238}U in the energy region above the photomeson production threshold around 150 MeV [1-5]. Independent of the experimental technique employed, a two-humped distribution has been found. The peak-to-valley ratio, i.e. the yield at mass numbers 95-99 divided by the yield in the valley between the two humps, has been shown to depend upon the bremsstrahlung maximum energy E_{max} which has been interpreted as indicating increasing probability for fission events yielding symmetric products with increasing bombarding energy. Below E_{max} = 1 GeV, the agreement between different laboratories is satisfying, but above this energy the situation is less clear. It was felt important to clarify this situation by measuring the mass distribution at E_{max} = 6.00 GeV. In addition, this irradiation energy is the highest one so far for which mass distributions of photofission have been studied. The experiment was performed at DESY, and the experimental details and results are presented in part I of this paper [29]. Tables and notations referred to, but not given here are to be found in section 3.4 of part I.

2. Chain yield considerations

In order to investigate the extent to which the measured cumulative yields, given in Table I:IV and Fig.1, correspond to chain yields, the relevant quantities to use when mass distributions are discussed, some calculations were performed based on a postulated gaussian charge dispersion curve. This curve describes the probability of forming a fission product with charge Z in a given mass chain with the most probable charge denoted Z_p and the full width at half maximum height (FWHM) Γ_Z. The experimental information used is given in table IV:I showing, for some particular mass chains to which eq (I-1) is applicable, the ratio R between independent and cumulative yields. In fig.2 some measured yields are shown as a function of the parent factor in addition to the lines, fitted to the experimental values by a least squares method, used to solve eq. (I-1).

The ratio R may be expressed with the aid of cumulative gaussian distributions according to the following relation[6)]

$$R = \frac{\sigma_q^i(A,Z)}{\sigma_q^c(A,Z)} = \frac{\int_{Z-.5}^{Z+.5} \exp\left(-\frac{(Z-Z_p)^2}{2w^2}\right)dZ}{\int_{-\infty}^{Z+.5} \exp\left(-\frac{(Z-Z_p)^2}{2w^2}\right)dZ} \qquad \text{(IV-1)}$$

where Γ_Z and w are connected by $\Gamma_Z \approx 2(w^2+1/12)$ [7)]. From eq. (IV-1) the Z_p values of mass chains 92, 97, 112, 135 and 140 were calculated from the experimentally determined R values for different values of w corresponding to a variation in Γ_Z from 1 to 4 charge units. The values obtained for Z_p and for the fractional cumulative chain yields (FCY) for different Γ_Z values are given in table IV:I.

Cuningham et al [8)] found that a value of 3 Z units for the FWHM of a gaussian-shaped charge-dispersion curve gave the best fit to data obtained in the 23 MeV bremsstrahlung induced fission of ^{232}Th and ^{238}U. In this analysis a measured value at E_{max} = 300 MeV for A = 134 reported by Schmitt and Sugarman [1] from their study of uranium photofission has also been included. Meason and Kuroda [9] used $\Gamma_Z \approx 2$ charge units in the 17.5 MeV monoenergetic photofission of ^{238}U in order to reproduce their experimental observations by calculations in the narrow mass region A = 131-135. In the case of the proton-induced fission of ^{238}U, a variety of Γ_Z values have been reported

0.95 below E_p = 100 MeV in the entire fission products region [10], 2.8 at E_p = 170 MeV for A = 100-140 [11] and recently 2.90 for A = 100-120 and 3.21 for A = 125-129 at E_p = 450 MeV [12].

In the estimates made below, the value Γ_Z = 3 charge units has been used. The Z_p values needed to calculate the fractional cumulative chain yields for the nuclides given in table I:IV were obtained from a simple interpolation in the mass region 92-140 from the Z_p values given in table IV:I. In most cases $Z - Z_p$ for the nuclides used in table I:IV turned out to be larger than 1.0 charge unit which correspond to values of FCY around 90 per cent. For mass chains outside the region defined above, one would expect the conclusion to be valid except for A = 88 since, in this particular case, Kr is situated further from the stability line, as given by Coryell [13], than is the case for neighbouring nuclides. One thus concludes that in most cases the measured yields given in table I:IV correspond to more than 90 per cent of the cumulative chain yields and subsequently that the points plotted in fig.1 represent the mass distribution to the same degree.

As the estimates made above are only approximate, the measured yields have not been corrected and will be treated as chain yields in the following sections. One must observe that the analysis outline above does not answer the basic question about the justification of using a gaussian shaped charge disperson curve in σ_q space, a problem for which special experimental investigations remain to be performed in the photofission case.

3. Comparison with other experiments

The agreement between the mass distribution curve given in fig.1 and earlier results [5)] obtained with the same experimental method in the energy region E_{max} = 300-1100 MeV is in general good if the experimental errors are taken into consideration. The major disagreement appears at mass numbers 103 and 105 where the present yield values are approximately 20 per cent larger compared to those below E_{max} = 1.1 GeV. This disagreement may perhaps be explained by the fact that in the earlier measurements the activities were followed for only two weeks and that the gamma spectra were evaluated manually. In the measurements below 1.1 GeV catcherfoils were used and no measurements were performed on the irradiated uranium, a difference compared to the present experiment which is not considered to affect the comparison made. Within the errors, agreement is also obtained when comparing the present results to those of Williams et al [4)] at E_{max} = 1.5 and 3.0 GeV insofar as the general appearance of the mass distributions is concerned. Large deviations in detail exist, but the error limits in the experiment of Williams et al are too wide to allow any closer study of these differences. It might be interesting however to point out that Williams et al reported an abnormally large yield for ^{135}Xe which they attribute "to be due to the decay of ^{135}I for which the production probability is large because of its closed neutron shell". In the present study, an abnormally low yield is obtained for both ^{135}I and ^{135}Xe in agreement with recent measurements [14] in the energy region E_{max} = 115-760 MeV where the ratio between $\sigma_q^c(^{135}I)/\sigma_q^c(^{132}Te)$ was found to be close to 1.2. In this particular experiment the same experimental method was used as in the present study, but similar results have been obtained by radiochemical studies of iodine isotopes in the same energy region [15]. An abnormally low yield for the mass chain A = 135 was also reported by Meason and Kuroda [9], who discussed the fine structure around mass number 132 in some detail. At the present stage it seems to be difficult to understand the low yield of the mass chain discussed. One would expect a smooth variation in the mass distribution with mass number and the experimental fact is that the dip at A = 135 is very deep, about a factor of 2 lower in yield than expected. The survival of shell effects to energy regions well above the giant resonance energy region seems doubtful. On the other hand both the iodine and xenon yields were also found to be small, although the analysis in section 2 shows that at least 90 per cent of the chain yield has been reached at ^{135}Xe. Errors in the gamma branching ratios do not thus seem to be a reasonable solution to the problem.

4. Symmetric and asymmetric fission

The mass distribution given in fig.1 has been divided into two parts, one symmetric and one asymmetric, see fig. 3. In doing so, the FWHM of the mass-hump centered at a $A = 137$ was used to fit the other two curves in such a way that agreement was reached between the sum of these two distributions and the experimental yields for $A < 125$. Although the fit was made by hand, reasonably good agreement was obtained for the entire mass region. The widths at half maximum height were found to be $\Gamma_a = 13$ and $\Gamma_s = 21$ mass units for the asymmetric and symmetric humps respectively in agreement with earlier results below $E_{max} = 1.1$ GeV [5]. From the results of Schmitt and Sugarman [1] at $E_{max} =$ 300 MeV similar results may be deduced, in addition to the masses of the fissioning nuclei for asymmetric fission $A_{fa} = 235$ and symmetric fission $A_{fs} = 234$. In the energy range $E_{max} = 300\text{-}1100$ MeV, the values obtained are $A_{fa} = 234$ and $A_{fs} = 224\text{-}234$, the large uncertainty in the latter quantity being due to difficulties in the construction of the symmetric mass hump [5]. In the present experiment this hump is better defined and the values obtained are $A_{fa} = 232.5$ and $A_{fs} = 221$. At this stage it might be interesting to determine what fissioning nuclei the Z_p values determined for the nuclides in table IV:I correspond to. In this calculation, the usual hypotheses for the charge division were used, i.e. the ECD hypothesis (Equal-Charge-Displacement) of Glendenin et al [16] and the UCD rule (Unchanged-Charge-Distribution) of Goeckermann and Perlman [17]. The relation used for the ECD hypothesis is

$$Z_p = Z_A - \frac{1}{2}\,(Z_A + Z_{A*} - Z_f) \qquad \text{(IV-2)}$$

where Z_A is the most stable charge in the mass chain A and Z_{A*} the corresponding quantity for the complementary fission fragments, i.e. before the emission post-fission neutrons. The values of these most stable charges having been taken from Coryell [13], and Z_f is the charge number of the fissioning nucleus.

According to the UCD rule Z_p is given by the following equation:

$$Z_p = \frac{Z_f}{A_t - n}\,A \qquad \text{(IV-3)}$$

where n is the total number of particles emitted (in most cases neutrons) before fission, and A_t is 238. The results of such calculations are given in table IV:II with $Z_f = 91\text{-}92$. This choice seems plausible as the emission probability of protons after the scission point is reached is zero and the fission probability becomes very small compared to the probability of neutron evaporation if a proton is emitted in the cascade or in the evaporation prior to scission [18]. Recent Monte Carlo calculations at our laboratory [27], based on the computer code due to Gabriel and Alsmiller [28], give similar results for Z_f .

The values calculated with the ECD hypothesis are in good agreement with the values taken from fig.3 which means that the total number of neutrons amounts to 5-7 in the asymmetric case and to 13-17 in the symmetric. To estimate the number of neutrons prior to fission, the number of post-fission neutrons must be known. From mass distribution studies below E_{max} = 300 MeV [1,9], the total number of neutrons emitted was 3 close to the measured number of post-fission neutrons, 2.77 ± 0.13, for 7 MeV monoenergetic photons reported by Condé and Holmberg [19]. The number of post-fission neutrons are estimated to divide themselves between the two fission fragments in proportion to the fragment masses. One thus estimates the number of neutrons prior to fission, i.e. in the cascade-evaporation chain, to be 2-4 for asymmetric fission events and 10-14 for symmetric events. This implies large differences in deposition energy for post cascade nuclei leading to the two types of fission events.

To determine what the mass distribution without the contribution from energies below 30 MeV looks like, the yield curve at 31 MeV bremsstrahlung maximum energy given by Dahl and Pappas [20] normalized to 3.9 mb at ^{91}Sr from the known σ_q curve for this nuclide [14] was subtracted from the values given in fig.3. The result show a mass distribution (see fig.4) with larger contributions from symmetric fission than in fig.3, as one would expect. However, the asymmetric fission events contribute to a large extent, in agreement both with earlier measurements and with calculations based on those measurements [5]. A small error, which will not affect the conclusions drawn above, was introduced when deriving fig.4 which tends to favour asymmetric fission:

viz the contributions to σ_q above 31 MeV due to cross sections below this energy and the energy-dependent shape of the bremsstrahlung spectrum were not taken into account. It is interesting in connection with fig.3 to calculate the total fission cross section per equivalent quantum at 6 GeV. A value of 316 ± 32 mb is obtained in very good agreement with the measured values of Vartapetjan et al [21]. They report two somewhat different values at 5 GeV, in a table 274 ± 28 mb and in a figure 310 ± 30 mb.

Finally, the peak-to-valley ratio was found from fig.3 to be 1.36 from the ratio between the yields at mass numbers A = 95 and A = 112. This value has been plotted in fig.5 in addition to all the values known above 10 MeV. The agreement is satisfying over the whole energy range. As the peak-to-valley ratios are used to deduce the strengths of the two types of fission events, it is obvious that the value in the valley should not be taken in the deepest part of the valley but at the mass number corresponding to the peak value of the symmetric part of the mass distribution. The peak-to-valley ratios are found to level out above 1 GeV. Such behaviour would be expected if the fission cross section decreases rapidly above 1 GeV [22]. This has been found to be the case in two recent measurements by Methasiri [23] and Wakuta [24].

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T a b l e IV:I The ratio R between independent and cumulative yields. Values of Z_p and fractional cumulative yields (FCY) at different values of Γ_Z, the full width at half maximum height of the gaussian charge dispersion curve.

Nuclide	R	$\Gamma_Z = 1$		$\Gamma_Z = 2$		$\Gamma_Z = 3$		$\Gamma_Z = 4$	
		Z_p	FCY	Z_p	FCY	Z_p	FCY	Z_p	FCY
^{92}Y	.23	38.04	.99	37.91	.95	37.86	.90	37.86	.88
^{97}Nb	.11	39.71	.99	39.38	.99	39.17	.97	39.01	.96
^{112}Ag	.22	46.02	.99	45.87	.96	45.81	.92	45.79	.89
^{135}Xe	.22	53.02	.99	52.87	.96	52.81	.92	52.79	.89
^{140}La	.13	55.78	.99	55.49	.98	55.31	.96	55.19	.95

Table IV:II The fissioning nuclei for some fission products calculated from the Z_p values with the use of the ECD and UCD rules. The uncertainties in A_f given result from the variation in Z_f.

Nuclide	Z_p	A_f(ECD)	A_f(UCD)
^{92}Y	37.9	231 ± 2	224 ± 1
^{97}Nb	39.2	234 ± 1	229 ± 1
^{112}Ag	45.8	226 ± 1	226 ± 1
^{135}Xe	52.8	237 ± 1	237 ± 1
^{140}La	55.3	234 ± 1	235 ± 1

Figure captions

Figure 1 Measured yields versus mass number.

Figure 2 Measured yields versus the parent factor K for some fission products.

Figure 3 The mass distribution at 6.00 GeV decomposed into symmetric and asymmetric fission. The stars represent the experimental values.

Figure 4 The mass distribution resulting from photons above 31 MeV decomposed into symmetric and asymmetric fission.

Figure 5 Peak-to-valley ratios for ^{238}U in the bremsstrahlung peak energy range 10 MeV to 6.00 GeV [1-5,20,25,26].

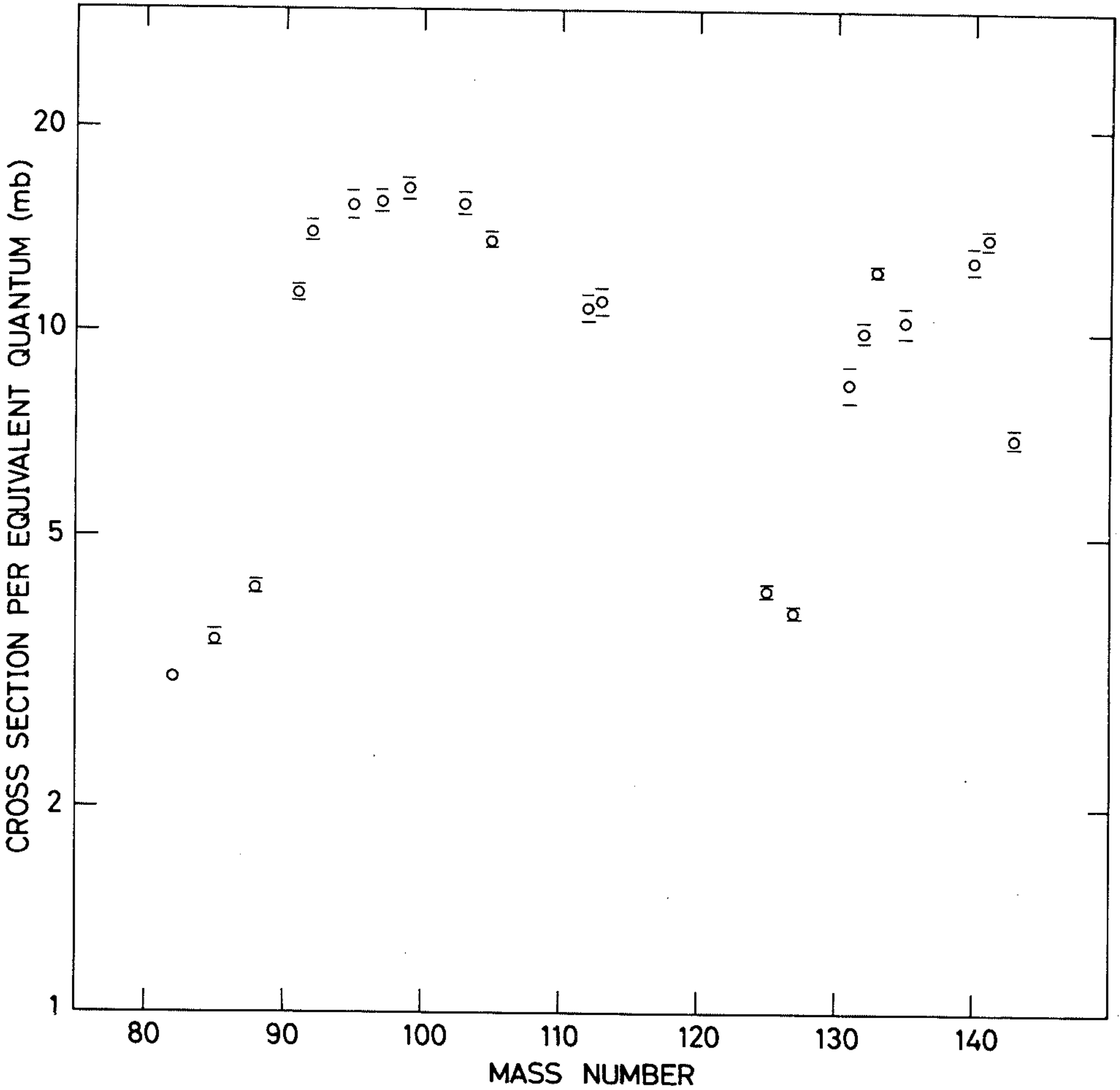

CROSS SECTION PER EQUIVALENT QUANTUM (mb)
20
10
5
2
1
80
90
100
110
120
130
140
MASS NUMBER

Figure 1

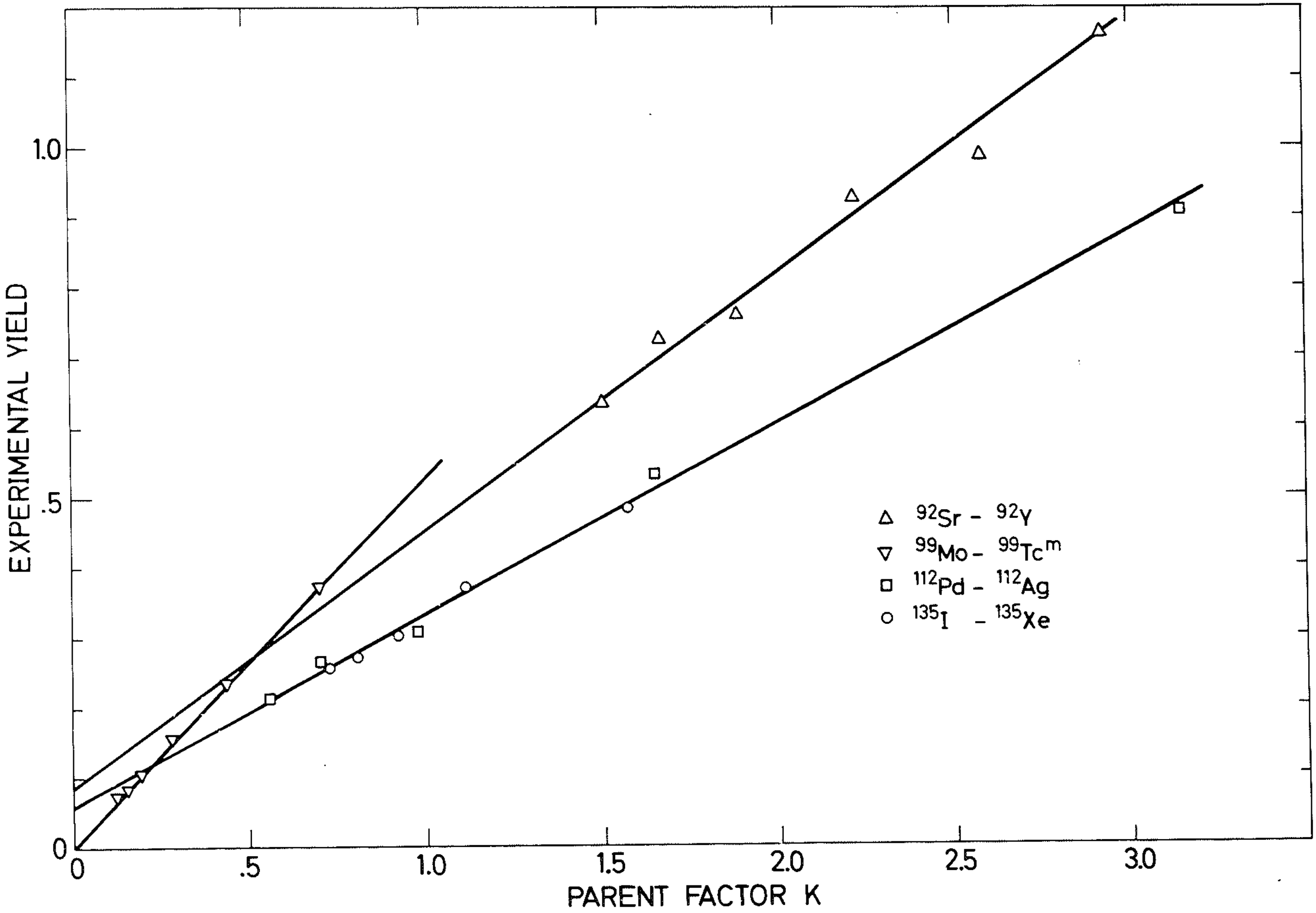
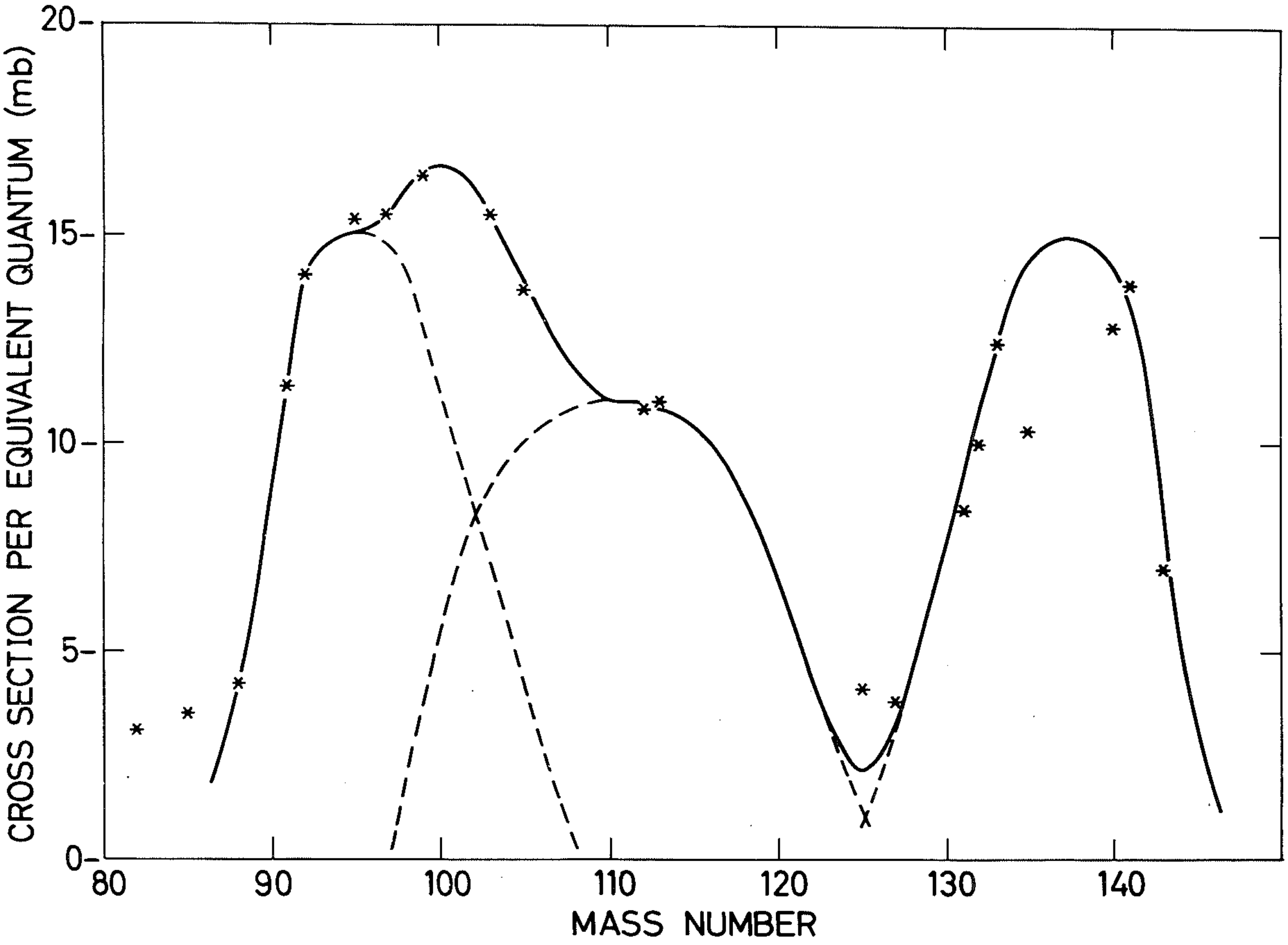

EXPERIMENTAL YIELD
PARENT FACTOR K
0
.5
1.0
1.5
2.0
2.5
3.0
^{92}Sr - ^{92}Y
^{99}Mo - $^{99}Tc^{m}$
^{112}Pd - ^{112}Ag
^{135}I - ^{135}Xe

Figure 2



CROSS SECTION PER EQUIVALENT QUANTUM (mb)
20
15
10
5
0
80
90
100
110
120
130
140
MASS NUMBER

Figure 3

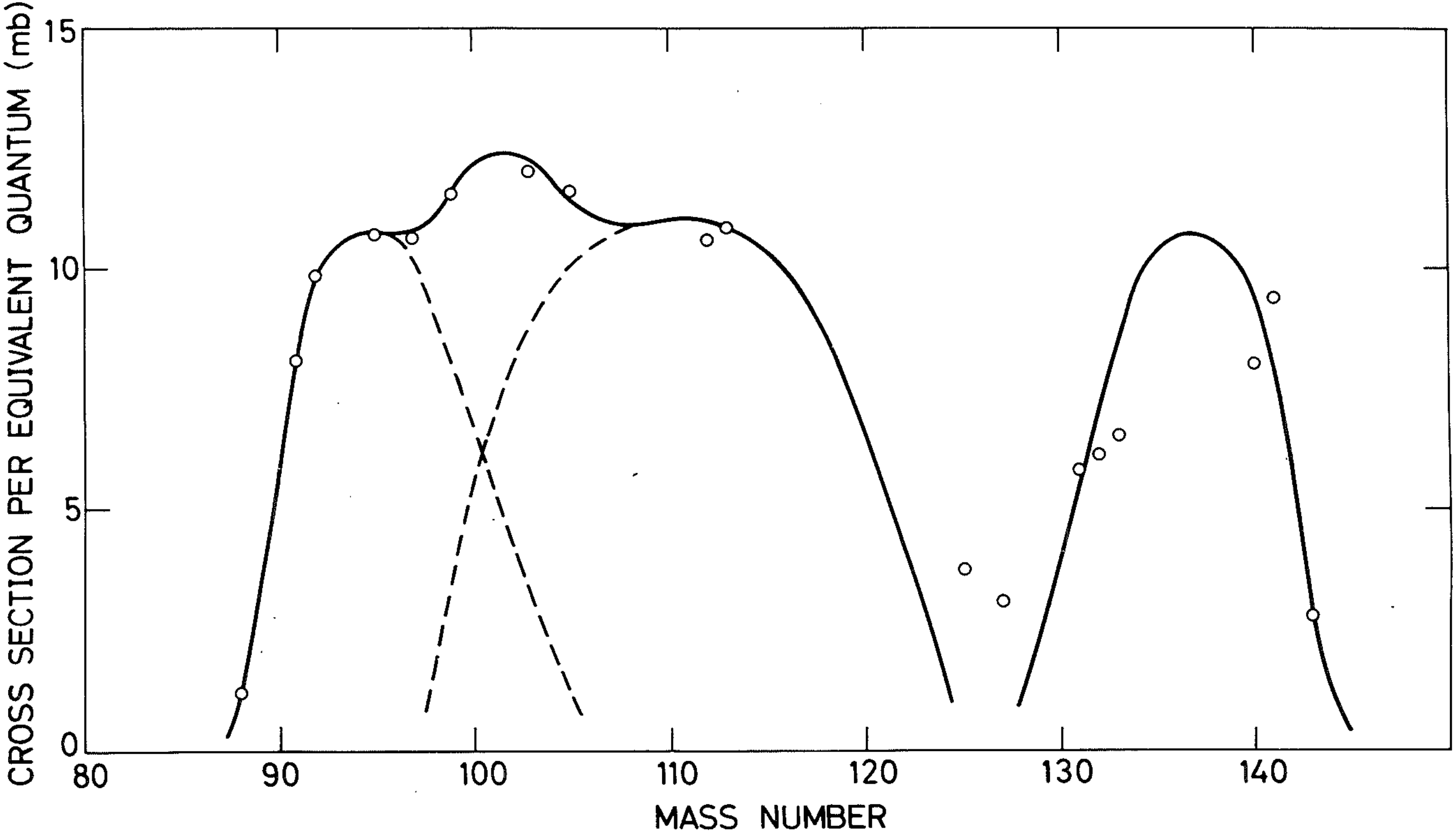

CROSS SECTION PER EQUIVALENT QUANTUM (mb)
0
5
10
15
80
90
100
110
120
130
140
MASS NUMBER

Figure 4

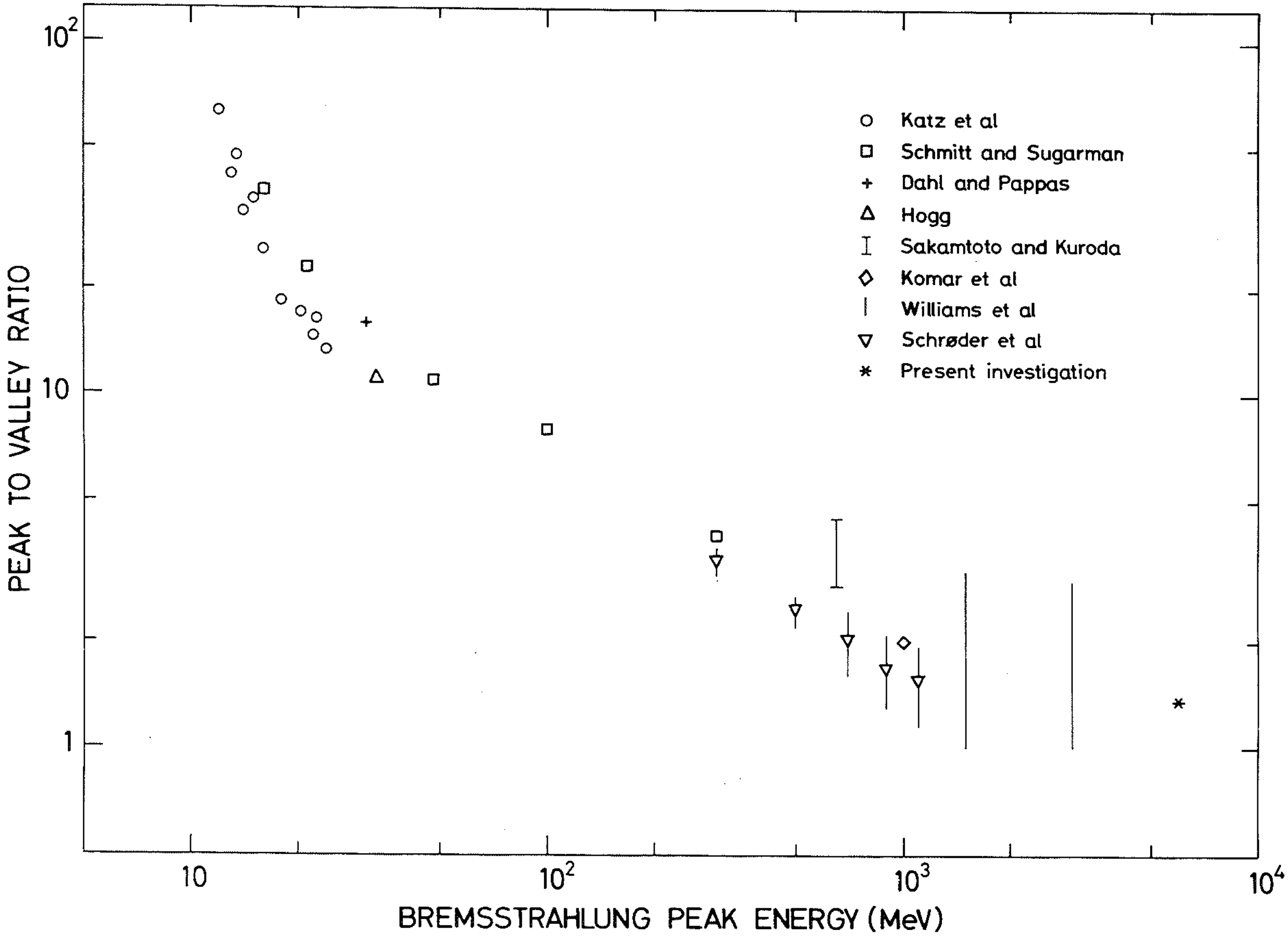

PEAK TO VALLEY RATIO
10^2
10
1
10
10^2
10^3
10^4
BREMSSTRAHLUNG PEAK ENERGY (MeV)
Katz et al
Schmitt and Sugarman
Dahl and Pappas
Hogg
Sakamtoto and Kuroda
Komar et al
Williams et al
Schrøder et al
Present investigation

Figure 5