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PRESENT STATUS OF PHOTOFISSION OF ACTINIDES NEAR THRESHOLD

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ABSTRACT

Presently available data on photofission cross sections, photoneutron cross sections and on angular distribution of photofission fragments of actinides near threshold have been reviewed. Some suggestions for additional studies have been made with the aim towards a better understanding of "threshold fission" phenomenon in the framework of a double humped barrier in fission.

SUMÁRIO

Os dados presentemente disponíveis, referentes a secções de choque para fotofissão, secções de choque para produção de neutrons e distribuições angulares de fragmentos de fotofissão dos actinídeos perto do limiar são revistos. Algumas sugestões para estudos adicionais são feitas com a finalidade de proporcionar um melhor entendimento do fenômeno da fissão perto do "limiar", dentro do quadro de uma dupla barreira na fissão.

INTRODUCTION

Our purpose here is to review the available data from photofission and photoneutron mesaurements of actinides near threshold and to point out a rather unclear situation regarding the present understanding of the recently obtained data. The phenomenon of photofission near threshold has received serious attention in last few years in view of its relative importance in studying the shape of the double humped fission barrier suggested recently by Strutinsky[1] and also mainly because of better gamma ray facilities becoming available at several laboratories. It is our hope that this review report will be helpful in encouraging more work by experimentalists and theorists alike towards a better understanding of threshold fission phenomenon.

Threshold photofission is an excellent means of studying physics of low energy fission, especially due to the restricted angular momenta in the entrance channel corresponding mainly to the dipole and the quadrupole photoabsorption. Measurements of photofission cross sections and of the angular distributions permit the determination of the relative contributions of the various fission channels at different excitation energies. Photofission is particularly useful for studying the shape of the double humped fission barrier below the neutron threshold, which of course cannot be studied with neutron induced fission.

PHOTOFISSION CROSS SECTION MEASUREMENTS

Recently a number of photofission experiments in the actinide region have been performed near fission threshold using bremsstrahlung beams[2,3], monoenergetic gamma rays from neutron capture reactions[4,5] and Compton scattered capture gamma rays[6,7] from the reaction $^{58}Ni(n,\gamma)^{59}Ni$. A comparison of the results from these three diferent techniques for Th^{232} and U^{238} is shown in Figures (1) and (2) respectively. The continuous lines represent the results of Rabotnov et.al.[2,3] using bremsstrahlung beam to induce fission. This is a "gross resolution" measurement and suffers mainly in a poor knowledge of bremsstrahlung spectrum needed in unfolding the cross sections from the experimentally measured fission yields. The open circles represent the results of Mafra et.al.[5] using discrete neutron capture gamma rays from several elements. Neutron capture gamma rays have a high flux per energy interval and also allow photofission measurements at precise energies. However, the choice of energies is restricted and one cannot measure cross section at continuously varying gamma ray energies. Furthermore, these gamma rays have extremely narrow widths (of the order of few electron - volts) and are thus capable of exciting individual compound nuclear levels. One must therefore exercise caution in interpreting any resonance structure observed with this technique as it could also be due to structure in the entrance (photoabsorption) channel corresponding to the individual compound nuclear states.

This method can be classified into the category of a "fine resolution" experiment. The filled circles represent the results of Khan and Knowles(6) using Compton scattered neutron capture gamma rays. This method has an intermediate energy resolution and can be profitably used to complement the bremsstrahlung data. It should however be emphasized here that in view of the markedly different energy resolutions, any comparison of the results obtained by these different techniques is of limited value and useful only in a qualitative sense.

As seen in Figure 1, measurements using Compton scattered capture gamma rays reveal a resonance structure in photofission cross sections of Th^{232} at 5.5 and 6.4 MeV. Similarly, for U^{238}, as shown in Figure 2, the measurements indicate a resonance structure at 5.2, 5.7, 6.2, 7.1 and 7.8 MeV. It is difficult to understand the existence of such structure in subbarrier photofission cross sections in terms of simple liquid drop model. However, it can be understood in terms of vibrational states in the second well of a double humped fission barrier. Recently, a detailed calculation for U^{238} along these lines has been performed by Bhandari and Onley(8). A new expression for the penetrability through a double humped potential barrier has been obtained and a "spreading width" method has been introduced to account for the broadening of the pure transmission resonances. Halflives for ground state spontaneous fission and isomeric fission have also been calculated and a single set of parameters of the double humped barrier for U^{238} has been obtained which is reasonably consistent with experimentally available information on photofission cross sections and ground state spontaneous and isomeric fission halflives. This calculation(8), in addition to satisfactorily reproducing the already observed peaks near fission threshold also predicts several resonances in photofission cross sections at lower gamma energies. Since these low energy peaks are a direct consequence of the concept of a double humped fission barrier, they need to be confirmed to strengthen our faith in this concept. Recently some efforts(9) are being made to measure low energy photofission cross sections and we can only encourage such endeavours in view of the importance of low energy photofission in studying the shape of a double humped fission barrier.

FISSION BARRIER SHAPES FOR ACTINIDES

Recently, several groups(10,11,12) have used Strutinsky's prescription to amalgamate the shell effects and the collective potential energy furnished by liquid drop model to calculate the resulting potential energy surfaces for heavy and superheavy nuclei. Even if we do not rely upon the exact values of the barrier parameters obtained from such calculations as well as those obtained from the analysis of experimental results by Back et. al.(13) given in table I, some general features are worth mentioning. The general consensus in actinide region is that as one goes from Th^{232} to Pu^{240}, the height of the outer barrier decreases relative to that of the inner barrier. Thus for Th^{232}, the outer barrier is the higher one; for U^{238}, inner and outer barriers are approximately of the same heights while for Pu^{240}, the outer barrier is the lower one.

Further, the inner barrier has been found to be stable against the mass asymmetry degree of freedom but the outer barrier is found to be unstable. This has provided[14] at least a qualitative explanation of one of the oldest unresolved problem in fission theory, namely that of mass asymmetry. A complete explanation, however, must await a proper dynamical calculation.

Since in low energy photofission, the main contribution comes only from the dipole and the quadrupole channels, it is worth asking as to what happens to the barrier heights in these different excitation channels. For an even-even nucleus to which we are restricting our considerations here, the ground state has the angular momentum and parity given by 0^+. Dipole gamma ray absorption on an even-even nucleus leads to a compound nucleus with spin, parity 1^-. Similarly the absorption of a quadrupole photon on an even-even nucleus leads to a compound nucleus with spin and parity 2^+. It has been suggested[15] that the height of the outer barrier is the same for 1^- as well as 2^+ channels but that the inner barrier corresponding to 2^+ channel is significantly lower than that for 1^- channel. The latter characteristic follows from the energy level diagrams of an axially and reflection - symmetric deformed transition state nucleus. While discussing angular distributions of photofission fragments near threshold, we shall realize the importance of such barrier characteristics.

PHOTO - NEUTRON MEASUREMENTS

Below the fission threshold, photoneutron emission and gamma deexcitation are the only processes competing with fis - sion. Consequently, the measurement of the (γ,n) cross sections is important in understanding the intermediate structure obser ved in photofission cross sections. Recently, some measurements (5,16,17) have been reported with rather interesting results. Mafra et.al (5) first found that the photoneutron and photofis- sion cross sections in U^{238} and Th^{232} showed similar interme - diate structures. They have also recently (17) repeated the (γ,n) experiment on Th^{232} using activation analysis and results have confirmed their earlier findings. Their results are shown in Figures 3 and 4. However, their experiment was performed using neutron capture gamma rays with extremely narrow widths which are of the order of or even smaller than the level spa- cing of say 1^- levels in U^{238} near 6 MeV of energy. This promp- ted Huizenga (18) to suggest that the observed structure may be due to the structure in the entrance channel (photo- absorption) depending upon how such a narrow gamma line over- laps with the individual compond nuclear states. However, re - cently the same experiment has been repeated by Knowles and Mafra (16) for U^{238} using the intermediate resolution technique of Compton scattered capture gamma rays. They have also found the existence of similar intermediate structure in (γ,f) and (γ,n) processes although their detailed results are different from those obtained earlier by Mafra et. al (5) presumably due to markedly different energy resolutions employed in these two measurements. No such measurement on Th^{232} has been reported as yet from Chalk River.

In this technique (Compton Scattered Capture Gamma Rays) with intermediate resolution, contrary to the case with direct neutron capture gamma rays, the usual statistical situation of an average over many compound resonances is clearly fulfilled and therefore such a result is quite interesting in that it indicates the presence of a similar gross structure in photoabsorption cross sections as opposed to the fine resolution structure corresponding to individual compound nuclear levels. However it is worthmentioning here that such a conclusion does not imply that it is the structure in the photoabsorption channel which is also showing up in fission as well as in neutron emission channels. From $^{236}U(t,pf)U^{238}$ reaction studies[13], it has been independently established that there are peaks in fission cross sections of U^{238} at 5.15 and 5.80 MeV. Also the angular distribution measurements of photofission fragments of U^{238} have led Knowles[16] to conclude that 6.2 MeV peak is definitely a photofission resonance. Therefore it appears that this intermediate structure may exist in fission, neutron emission as well as in photoabsorption channels quite independently (experimentally) of each other. From a theoretical point of view, it will then be interesting to find an unifying explanation which could be consistent with these observed features. An explanation in terms of a doorway state mechanism may perhaps be in order. However, since the neutron multiplicity($\bar{\nu}$) enters in the extraction of (γ,n) cross sections from the measured total neutron production cross sections in the above measurements[5,16,17], it is important first to measure the energy dependence of the average number of neutrons emitted per fission more accurately.

No such detailed photofission or photoneutron cross-section measurements have been reported recently for Pu^{240}.

While discussing threshold photofission phenomenon, it is important to note here that experimental data on the cross sections for photoabsorption (dipole and quadrupole) at low gamma energies for actinides are not available and thus constitute a major drawback in analysing the data for photofission. For the energy dependence of the absorption cross section, one can only extrapolate from the data at higher energies. There is however an estimate given by Peter Axel[19] which yields satisfactory agreement[18] with the data on photoabsorption cross sections extrapolated to lower energies. However, it is necessary to have some direct measurements at low gamma energies to determine if any gross resonance structure is also present in photoabsorption cross sections.

ANGULAR DISTRIBUTION OF PHOTOFISSION FRAGMENTS

Besides the energy dependence of photofission cross sections, the other experimentally measured quantity which helps in determining the relative contribution of various fission channels at different excitation energies is the angular distribution of fission fragments. It is helpful here to briefly review the way different angular momentum dependences are extracted from the measured data on angular distribution of photofission fragments.

A.Bohr[20] pointed out in 1955 that despite the very high excitation energy of the fissioning nucleus, most of this energy is in the form of deformation energy. Hence the transition state nucleus (at the top of the single humped fission barrier, also called the "saddle point")is thermodynamically "cold" and is expected to have a spectrum of excited states analogous to those of a normal nucleus near its ground state. Bohr further pointed out that the case of photofission of even-even targets has some especially simple features because the electric dipole or quadrupole absorption involved can only excite states of low angular momentum, for example 1^- and 2^+ respectively. On the other hand, fission induced by charged particles or neutrons gives a large variety of momentum states. The relevant quantum numbers which determine the angular dependence of photofission fragments are $\vec{J}$, the total angular momentum; M, the component of $\vec{J}$ along the space fixed Z-axis also taken as the beam direction and K, the component of $\vec{J}$ along the nuclear symmetry axis. In general, the measured angular distribution of photofission fragments are fitted to the expression

$$W(\theta) = a + b\sin^2\theta + c\sin^2 2\theta \qquad (1)$$

where θ is the angle between the beam direction (space fixed Z-axis) and the nuclear symmetry axis along which the fragments are emitted. In this relation, the coefficients a and b are mixed coefficients coming from dipole as well as quadrupole fission while the coefficient c comes entirely from the quadrupole fission. A derivation of this relation and different K - distributions have been discussed in detail by Vandenbosch and Huizenga[21] in their recent text on nuclear fission.

Recently, several experimental investigations[2,22,23,24] have been reported on angular distributions of photofission fragments of actinides using again all the three types of photon sources as mentioned earlier. However no systematic investigation of all the actinides using the same gamma source with the aim of understanding the fission threshold phenomenon in the framework of a double humped barrier has yet been reported. such study for Th^{232}, U^{238} and Pu^{240} is in progress now at Instituto Energia Atomica, São Paulo using capture gamma rays. Since the energy widths of these capture gamma rays are narrow, if any interesting features are observed in this investigation, such a study should be repeated as for example with Compton scattered gamma rays or with the positron annihilation gamma rays which have an intermediate resolution. The importance of such an investigation stems from the fact that it is of interest to examine the effects of the recently suggested double humped barrier on the angular distribution of photofission fragments. For actinides, it is particularly interesting because as mentioned earlier, for Th^{232} the outer barrier is the higher one while for Pu^{240} the outer barrier should be the lower one. For U^{238}, the two barriers are approximately of the same heights. A systematic study of these actinides with the same gamma source as for example compton scattered capture gamma rays might yield useful information on "where" the angular distributions are determined in the course of passage through a double humped barrier and how these are affected by the relative heights of the two barriers and by the presence of the second well. This may help in understanding the complicated concept of a transition state nucleus in the framework of a double humped barrier in fission.

Before the introduction of the intermediate well, a set of "states" supposedly formed at the saddle point were identified as determining the necessary quantum-numbers which in turn determined the angular distributions.With the double barrier, there are, of course, two saddle points and according to Vandenbosch[15] and Huizenga[18], K-distribution of the outer rather than the inner barrier should determine the angular distribution irrespective of which is the higher barrier. However, as has been suggested recently by Bhandari and Onley[8], it is very tempting to discard the idea of a saddle point state when there is apparently a potential well sitting at the exit of the fission channel and inquire if the states formed at this point do not, more realistically determine the angular distribution of fission fragments.

From the measurements of angular distributions of fission fragments of actinides atleast one general feature seems to have been established. This is that for Pu^{240}, the quadrupole fission dominates in the low energy region; for U^{238} the quadrupole component is significant while for Th^{232}, there is very little quadrupole component in the low energy region. This is shown for these actinide nuclei in Figure 5 where the ratio (c/b) of the coefficients defined in equation (1) has been plotted as a function of energy. It should however be realized that the energy dependence of these coefficients extracted from bremsstrahlung induced fission data is different[32] from that obtained by using monoenergetic photons to induce fission. Therefore any comparison of the energy dependence of these coefficients obtained using different gamma ray sources is only of limited value. It is important to remember that equation (1) has been derived assuming absorption of monoenergetic photons of certain multipolarities.

Since the coefficient c comes entirely from quadrupole fission while b contains both dipole as well as quadrupole contributions, the ratio (c/b) is a measure of the relative quadrupole strength. We can see in Figure 5 that (c/b) becomes very large for Pu^{240} and U^{238} at low energies while no such increase is apparent in the case of Th^{232}. These observations seem to imply a strong dependence of the ratio of quadrupole to dipole absorption on the target nucleus. However Vandenbosch[15] has recently suggested another explanation in terms of the relative heights of the two barriers in the double humped fission barrier for these actinides. Taking the two extreme cases of Pu^{240} and Th^{232}, Vandenbosch argues that for plutonium, since the outer barrier is the lower one and about equally high for 1^- and 2^+ states, a lower inner barrier for 2^+ channel than that for 1^- channel compensates for the relatively small quadrupole to dipole photoabsorption ratio. This appears to be a reasonable explanation for Pu^{240} at least in a qualitative sense. However since the behaviour of penetrability through such barriers is now quite well known, it will be interesting to have some quantitative estimates of the relative enhancement due to a lower barrier and check if it does overcompensate the presently assumed small quadrupole to dipole absorption ratio[18].

However for Th^{232}, Vandenbosch argues that here since the outer barrier is the higher one and since it is nearly equally high for the 1^- and 2^+ states it therefore provides no enhancement for fission resulting from the quadrupole absorption . This does not seem to us to be fully justified because even here the inner barrier for 2^+ states is considerably lower than that for 1^- states and therefore as far as the penetration through the inner barrier is concerned, 2^+ state is definitely preferred. Once the inner barrier has been tunneled, the potential shape is the same for 1^- as well as 2^+ states. The argument presented by Vandenbosch will be valid for instance if the height of inner barrier is drastically lower than that of the outer barrier so that one will in effect be talking about excitations above the first barrier. However, this is not the case as is evident from the barrier shapes for Th^{232} and Pu^{240} used by Vandenbosch and reproduced here in Figure 6. These shapes are consistent with the barrier parameters obtained recently by Back et.al.[13] from direct reaction fission studies. However it is interesting to note that theoretical calculations[25] earlier had predicted significantly lower inner barrier and second minimum for Th^{232} which were not consistent with the experimentally determined values. This resulted in well known[25,26,27] "Thorium Anomaly" in the fission literature recently. Although the inner barrier for Th^{232} corresponding to 2^+ states is significantly lower than the outer barrier, the inner barrier for 1^- states is comparable[15] to or even higher than the outer barrier. The situation is further complicated by the recent results of Nix et.al.[11] suggesting that there might be a "third minimum" in the potential energy surface for Thorium. A study of penetrability through a three humped barrier is in progress and will be published elsewhere[28]. Also there is a low energy resonance in photofission cross section of Thorium at 5.6 MeV as reported by Rabotnov et.al[2] from their photofission data. It is not clear what effects such a resonance could have upon the relative qua drupole to dipole absorption ratio.

In view of evidences in favor of a potential landscape in which the height of the inner barrier (for Th^{232}) is not drastically lower than that of the outer barrier, the explanation suggested by Vandenbosch for Th^{232} is not very convincing. It could however be that at the lowest energies where we presently have the data available, due to comparatively large penetrability through the inner barrier corresponding to 2^+ states, the states formed by quadrupole absorption prefer to deexcite by gamma emission from the second well region back to the primary well. This could then provide an explanation for the fact that almost no quadrupole fission component has been observed in low energy photofission of Th^{232}. For dipole case, since the inner barrier is also much higher (at least comparable in height to the outer barrier) and the fact that gamma - deexcitation involves a two step process (first the penetration through the inner barrier and then the gamma deexcitation to the ground state) there is preferably the passage through the outer barrier along the fission path. It appears that results of photofission measurements at even lower excitaion energies than available now will help in clarifying the situation. If this explanation is correct, then one should not even expect to observe a fission isomer in Th^{232} because even if a shape isomer is formed, it would always preferably decay through gamma deexcitation and that too through quadrupole channel. The failure to observe a fission isomer in Th^{232} todate at least tends to support this hypothesis.

For U^{238}, the inner and outer barriers are approximately of the same height. In view of the above explanation, this will suggest that the greater strength of the decay of shape isomer of U^{238} should also lie in gamma deexcitation. This is supported by the recent results from Russo et al[29] although the question of multipolarity responsible for this deexcitation is still open. It would be very interesting to look into the gamma branch of decay of Th^{232} shape isomer and experiments on Th^{232} similar to the one reported recently by Russo et al[29] on U^{238} would yield useful information.

It has been shown recently[30,31] that the process of electroexcitation through a "virtual photon spectrum" is relatively more sensitive to the quadrupole and other higher multipoles than the usual process of photoexcitation involving "real photons". Since the quadrupole components involved in these threshold investigations are very small, it might be useful to exploit this important feature of the "virtual photon spectrum" for a systematic investigation of angular distributions of electron induced fission fragments of actinides near threshold to determine the relative contribution of the quadrupole and other multipole channels. Such investigations are in progress at this laboratory.

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FIGURE CAPTIONS

Figure 1 - Photofission cross-sections of Th^{232}. Solid line represents the resultsof Robotnov et al[2] using bremsstrahlung technique, filled circles represent the results of Khan and Knowles[6] using Compton scattered neutron capture gamma rays and open circles represent the results of Mafra et al[5] using direct neutron capture gamma rays obtained from various elements as labeled in the diagram.

Figure 2 - Photofission cross-sections of U^{238}. Solid line represents the results of Rabotnov et al[2] using bremsstrahlung technique, filled circles represent the results of Khan and Knowles[6] using Compton scattered neutron capture gamma rays and open circles represent the results of Mafra et al[5] using direct neutron capture gamma rays obtained from various elements as labeled in the diagram.

Figure 3 - Comparison of photofission and photoneutron cross-sections of Th^{232}. Filled circles represent photofission cross-section multiplied by ten (to show it clearly on the same plot as that of (γ,n)) and cross represent the photoneutron cross-sections. From O.Y.Mafra et al[17].

Figure 4 - Comparison of photofission and photoneutron cross-sections of U^{238}. Filled circles represent photofission cross-section multiplied by ten (to show it clearly on the same plot as that of (γ,n)) and cross represent the photo neutron cross-sections. From O.Y.Mafra et al[5].

Figure 5 - Ratio(c/b) for even-even actinides. Filled circles represent the results using bremsstrahlung technique[2] while open circles represent the results using monoenergetic gamma rays[22]. Taken from J.R. Huizenga and H.C.Britt, Proceedings[18] of International Conference on Photo-Nuclear Reactions and Applications, Asilomar, 1973.

Figure 6 - Fission barriers for Th^{232} and Pu^{240}. The dashed curve in the inner barrier region corresponds to 1^- channel while the solid curve represents 2^+ channel. From R.Vandenbosch[15].

TABLE CAPTION

I: Fission barrier parameters obtained by Back et al[13] from direct reaction induced fission for even-even actinides.

E_A, E_B, $\hbar\omega_A$, $\hbar\omega_B$ are the height of the inner barrier, height of the outer barrier, inner barrier curvature and the outer barrier curvature respectively.

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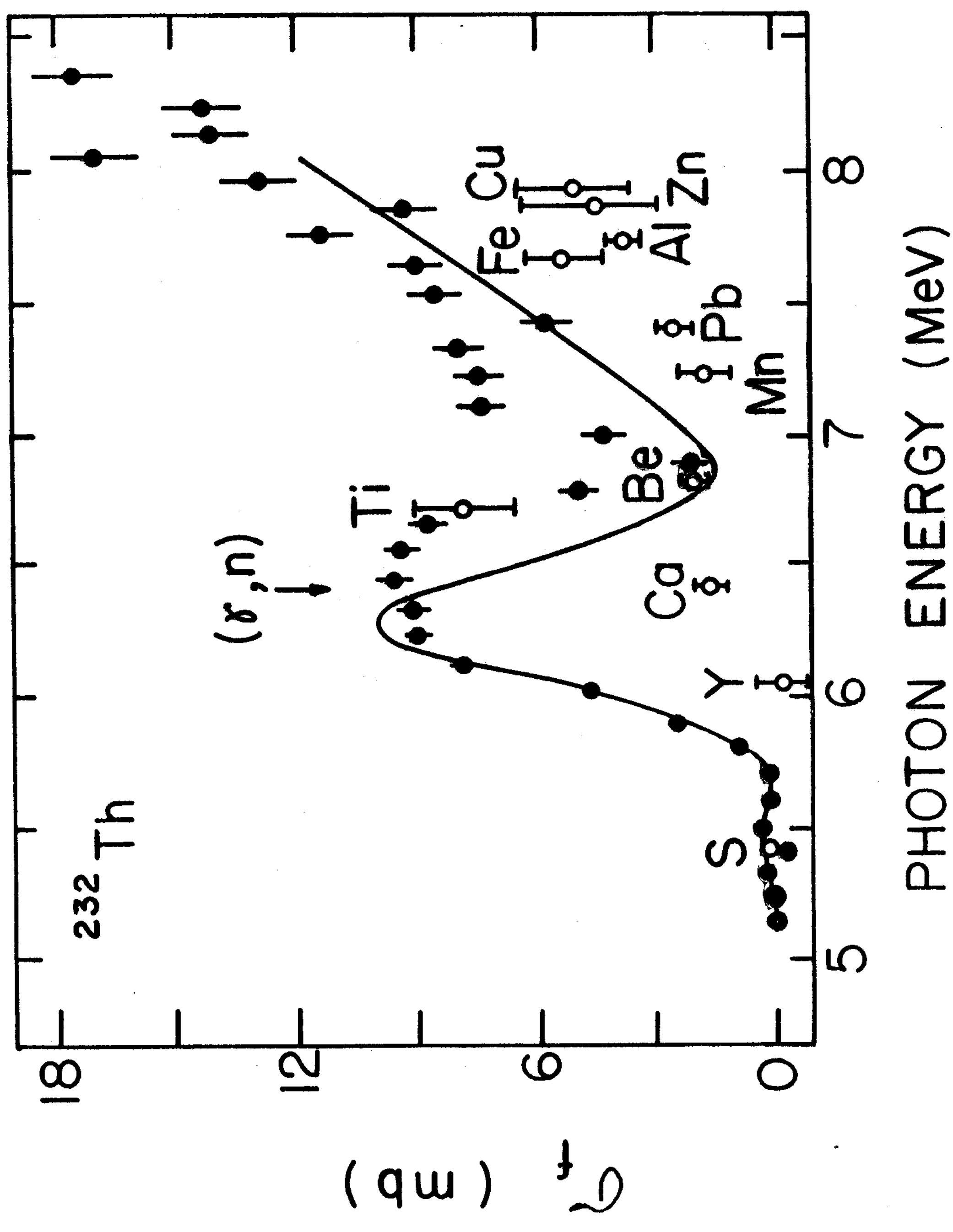
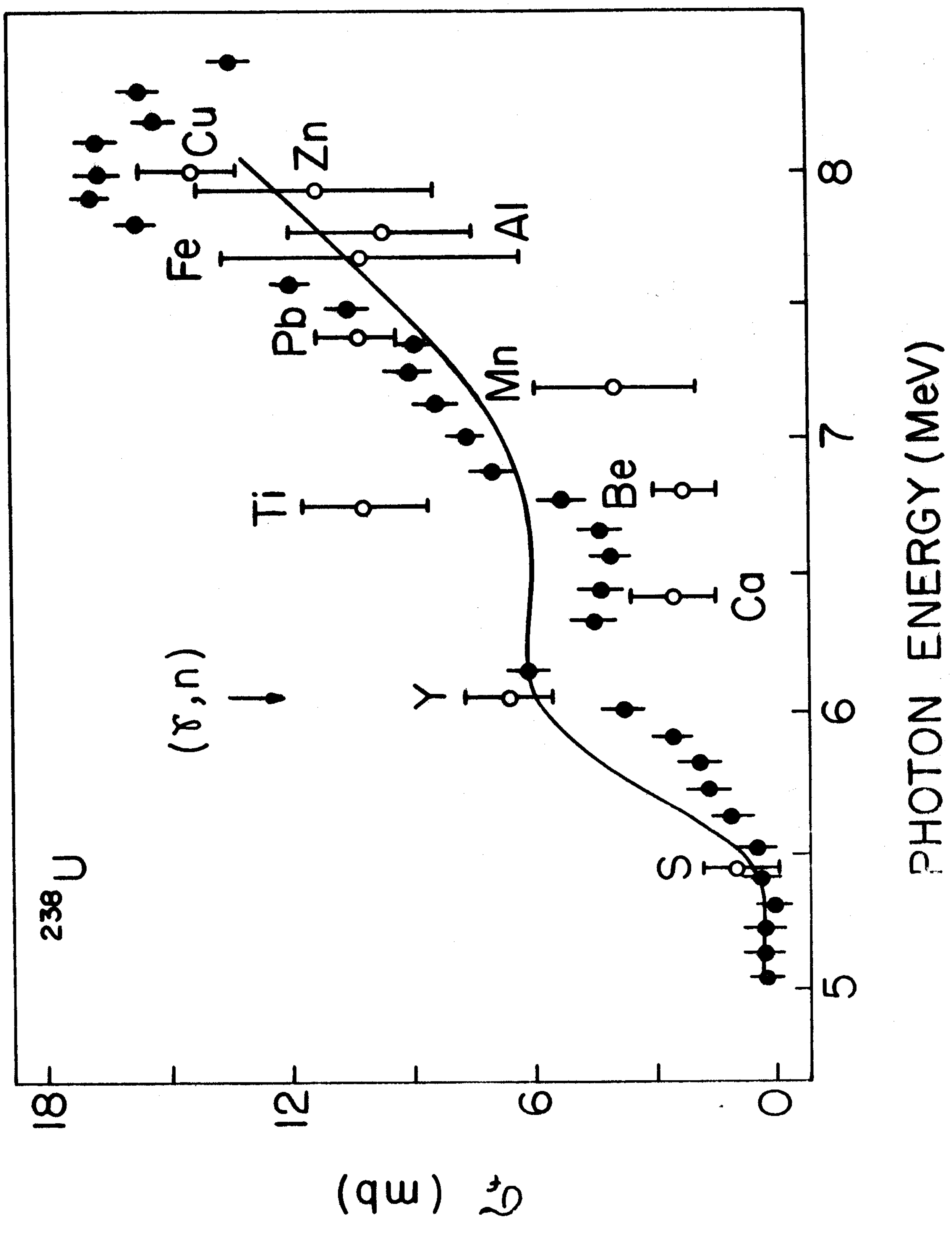


^{232}Th
(γ,n)
σ_f (mb)
PHOTON ENERGY (MeV)
S
Y
Ca
Ti
Be
Mn
Pb
Fe
Al
Cu
Zn

Fig. 1



^{238}U
(γ, n)
σ_f (mb)
0
6
12
18
5
6
7
8
PHOTON ENERGY (MeV)
S
Y
Ti
Ca
Be
Mn
Pb
Fe
Al
Zn
Cu

Figure 2

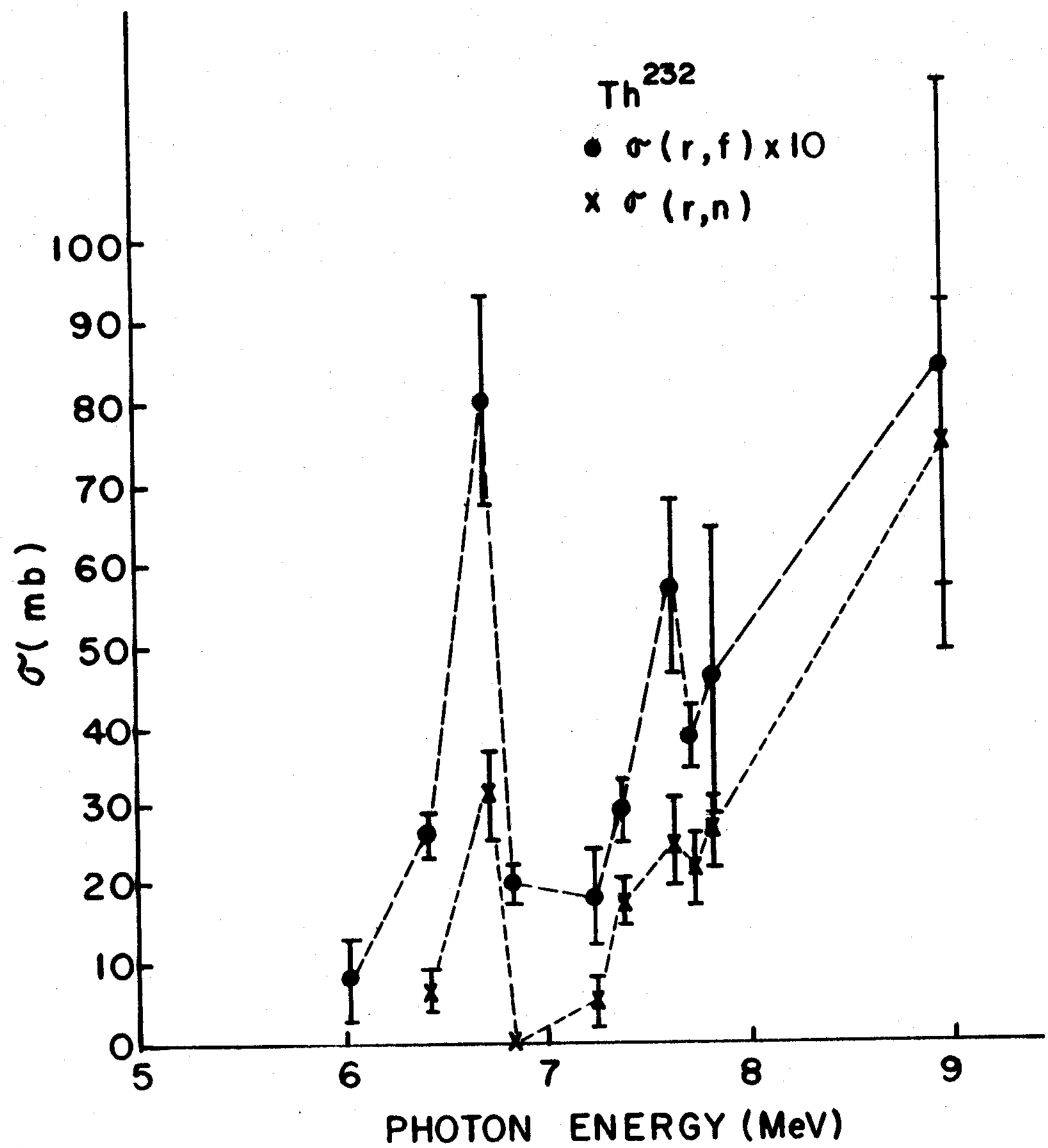

Th232
● σ (r,f) x 10
x σ (r,n)
100
90
80
70
60
50
40
30
20
10
0
σ (mb)
5
6
7
8
9
PHOTON ENERGY (MeV)

Figure 3

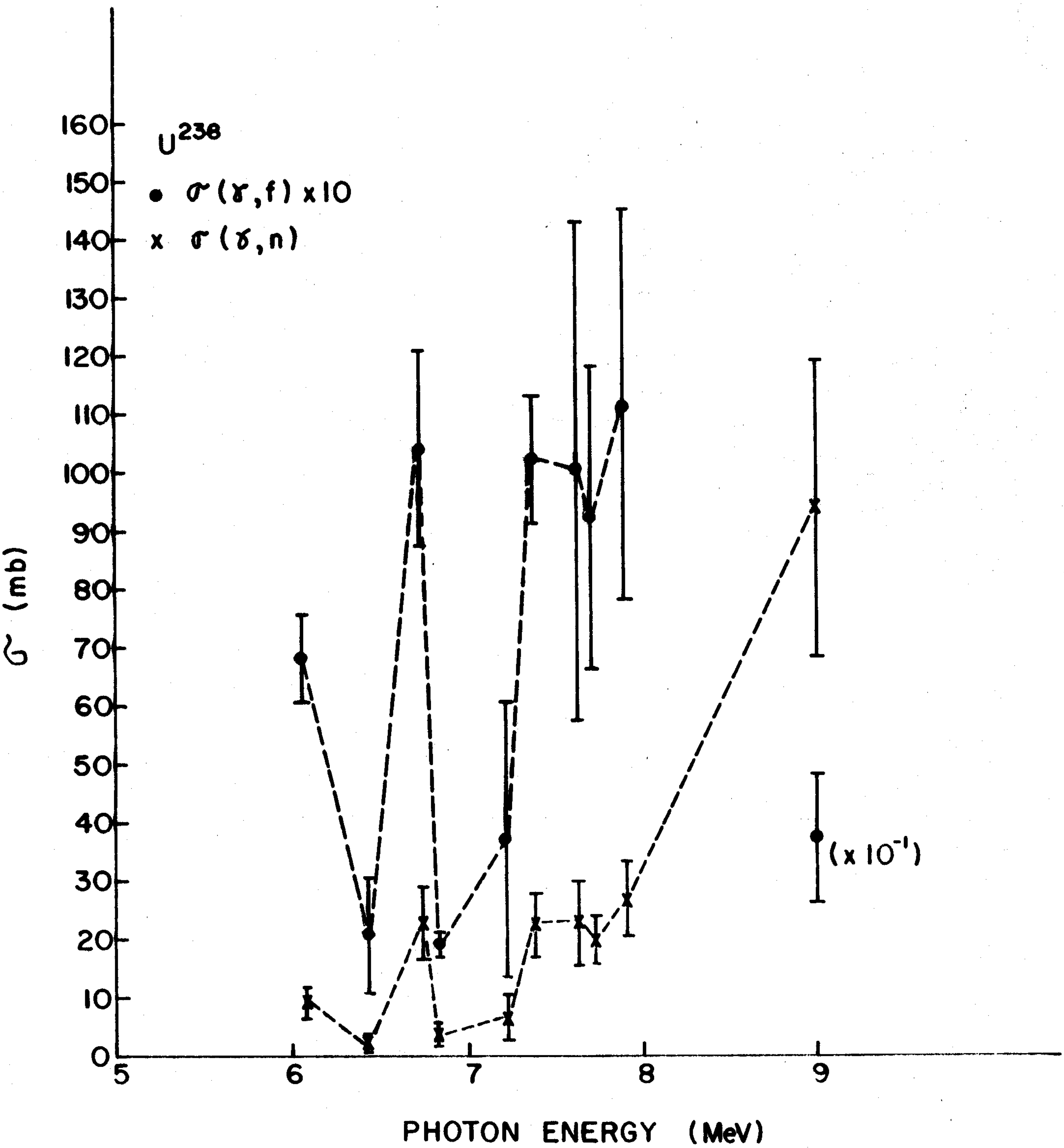

U^{238}
● $\sigma(\gamma,f) \times 10$
x $\sigma(\gamma,n)$
σ (mb)
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
0
5
6
7
8
9
$(\times 10^{-1})$
PHOTON ENERGY (MeV)

Fig. 4

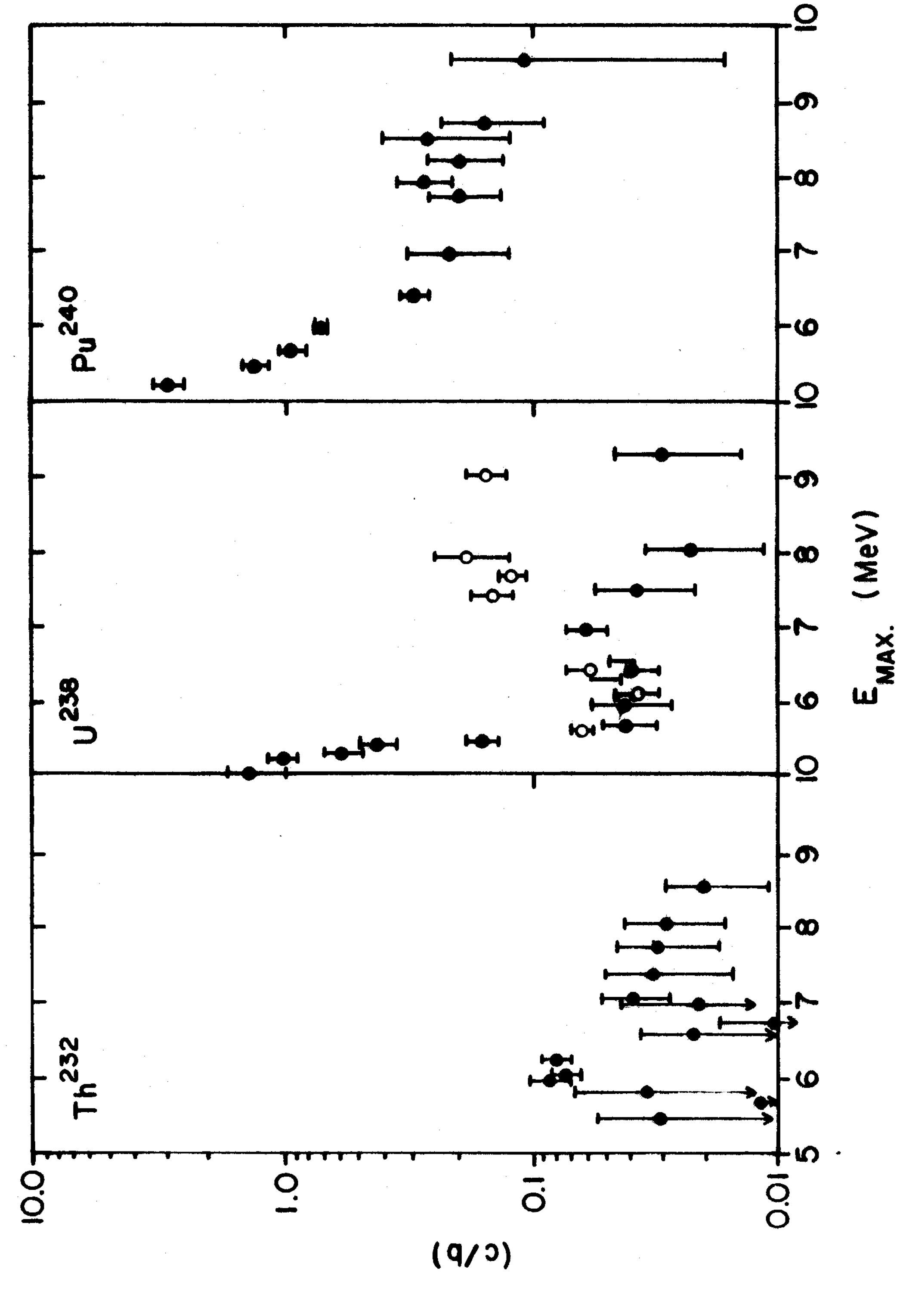


Fig.5

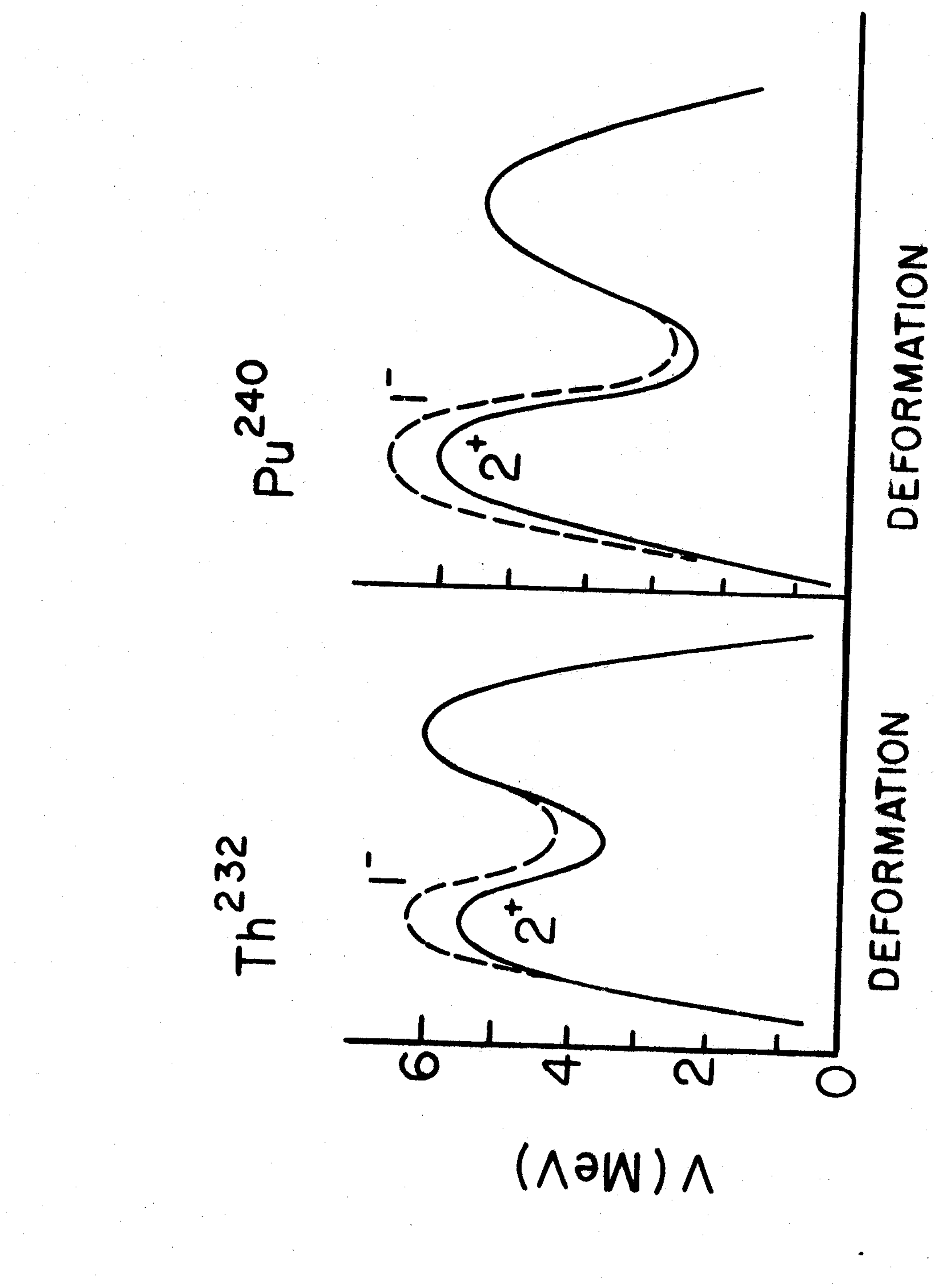


Fig. 6

TABLE I

NUCLEUS	E_A (MeV)	E_B (MeV)	$\hbar\omega_A$ (MeV)	$\hbar\omega_B$ (MeV)
^{230}Th		6.5 ± 0.3		
^{232}Th		6.15 ± 0.20		0.50 ± 0.10
^{234}Th	6.15 ± 0.20	6.52 ± 0.20	1.00 ± 0.10	0.75 ± 0.10
^{232}U	5.54 ± 0.20	5.45 ± 0.20	0.80 ± 0.10	0.55 ± 0.10
^{234}U	6.20 ± 0.25	5.95 ± 0.25	1.00 ± 0.10	0.65 ± 0.10
^{236}U	5.70 ± 0.20	5.68 ± 0.20	0.90 ± 0.10	0.50 ± 0.10
^{238}U	5.90 ± 0.20	6.12 ± 0.20	1.00 ± 0.10	0.62 ± 0.10
^{240}U	5.75 ± 0.20	5.95 ± 0.20	1.00 ± 0.10	0.70 ± 0.10
^{238}Pu	5.90 ± 0.20	5.20 ± 0.30	0.80 ± 0.10	0.55 ± 0.10
^{240}Pu	5.80 ± 0.20	5.45 ± 0.20	0.82 ± 0.10	0.60 ± 0.10
^{242}Pu	5.60 ± 0.20	5.63 ± 0.20	0.82 ± 0.10	0.59 ± 0.10
^{244}Pu	<5.6	5.35 ± 0.20		0.57 ± 0.10
^{244}Cm	6.12 ± 0.20	<4.9	0.90 ± 0.10	
^{248}Cm	6.15 ± 0.20	<4.6	0.90 ± 0.10	
^{250}Cm	5.15 ± 0.20	3.90 ± 0.30	0.72 ± 0.10	0.69 ± 0.10