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OBSERVATION OF M1 STRENGTH IN THE PHOTOFISSION OF ACTINIDE NUCLEI

by

J.D.T. Arruda-Neto, S.B. Herdade, and
B.L. Berman

Instituto de Física, Universidade de São Paulo

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OBSERVATION OF M1 STRENGTH IN THE PHOTOFISSION OF ACTINIDE NUCLEI*

J.D.T. Arruda-Neto, S.B. Herdade, and B.L. Berman[†]
Physics Institute, University of São Paulo, São Paulo, Brazil

ABSTRACT

Electrofission-fragment angular distributions for ^{234}U, ^{236}U, and ^{238}U, in the energy range from 5 to 7 MeV, are analyzed in the framework of virtual-photon formalism, which allows the separation of the multipolar components in the fission channel. Strong evidence for a substantial concentration of M1 strength in the fission decay of these nuclei is found.

Keyword abstract:

NUCLEAR REACTION $^{234,236,238}U(e,f)$; E_e = 5-7 MeV. Measured $\sigma(\theta)$. Deduced M1-component of the photofission process.

Classification Numbers:
2520 – 2585 – 2790

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[†] Permanent address: Lawrence Livermore National Laboratory, University of California, Livermore, CA 94550.

I. INTRODUCTION

Magnetic excitations have attracted attention with respect to the existence of magnetic multipole giant resonances, spin-spin and spin-isospin forces, quenching of the magnetic moment, core polarization, and mesonic effects (Knüpfer et al 1978, Sagawa et al 1979, Richter 1981). The question of the existence of a giant magnetic dipole resonance (GMDR) has been the subject of much experimental and theoretical work (in this regard, we refer the reader to the excellent survey of ground-state magnetic dipole strength by Horen (1979)). However, the experimental evidence supporting the existence of the GMDR for heavy nuclei is not compelling when compared to the giant dipole resonance (GDR), the giant quadrupole resonance (GQR), and the giant monopole resonance (GMR). For example, the subject of the 1^+ states in ^{208}Pb is a long standing problem. Several states in ^{208}Pb have been proposed to be 1^+, but at present the only ones which remain unchallenged are some weak states which were located by neutron-induced reactions at a few hundred keV above the neutron threshold (Horen 1979, Horen et al 1977, Laszewski et al 1977). The fact that substantial M1 strength was not observed in heavy nuclei has been a puzzle for a long time. However, recent experiments (Richter 1981, Horen et al 1980, Meuer et al 1980, Anantaraman et al 1981) have revealed both the existence of giant magnetic excitations and the quenching phenomenon to be common in heavier nuclei. In any case, it is believed that the GMDR is located in the energy range between $\sim 35\ A^{-1/3}$ MeV (for light nuclei) and $\sim 45\ A^{1/3}$ MeV; that the sum-rule strength is nearly exhausted in a number of light nuclei; and that the width is $\Gamma_x \cong 0.2\omega_x$ (where ω_x is the excitation energy) (Hanna 1969, 1977).

Recently, experimental evidence for some concentration of M1 strength in ^{238}U and ^{236}U was obtained (Arruda-Neto et al 1978, 1980a, 1980b, 1981) by the analysis of electro-fission-fragment angular distributions. More recently, we performed detailed measurements of electrofission angular distributions for ^{234}U (Arruda-Neto et al 1982), using an electron beam from the University of São Paulo Linear Accelerator (details in Arruda-Neto et al (1980b) and Arruda-Neto et al (1982)). In the present work we analyze these angular-distribution data for ^{234}U, using the virtual-photon formalism (Arruda-Neto et al 1978), in an attempt to find a concentration of M1 strength in the energy region just above the fission barrier. In so doing, we had in mind two motivations: (1) to establish a systematics for the occurrence of M1 strength in the fission decay of actinide nuclei, because of the controversial nature of the results reported to date related to the detection of M1 strength in heavy nuclei; and (2) as a stringent test of our technique and of our previous results for ^{236}U and ^{238}U.

As pointed out recently by Hicks et al (1982), it is very difficult to obtain reliable information on M1 transitions in heavy nuclei from single-arm (e,e') experiments, even at backward scattering angles. According to the predicted q-dependence of the M1 form factor in an (e,e') experiment, the current measurements barely reach (from the high-q side) the first diffraction maximum, and their interpretation is sensitive to model uncertainties.

On the other hand, an inclusive (e,x) cross section, as for the (e,f) reaction investigated in the present work, is domintated by small-momentum-transfer (low-q) events, for which the longitudinal component is negligible with respect to the transverse one (Arruda-Neto et al 1980b, 1982). Thus, electrofission measurements correspond nearly to measurements at the photon point ($q=\omega$). An (e,e') measurement samples the strength function at $q>\omega$, and the extrapolation of the form factor back to the photon point is a tedious and uncertain enterprise. Also, the interpretation of (e,x) data is model-independent, and this advantage can compensate for the drawback of dealing with integral cross sections. Finally, the angular distribution of fission fragments induced by virtual photons has proved to be eminently suitable for the study of the low-lying levels in the fission spectrum (at the saddle point) (Arruda-Neto et al 1980a, 1982). As shown in Fig. 1, the collective band structures for an even-even transition nucleus (with a stable quadrupole deformation) (Albertson and Forkman 1965, Vandenbosch and Huizenga 1973) exhibit an $I^{\pi}=1^{+}$ level at the mass asymmetry/bending band, located approximately 0.5 MeV above the fission barrier. Such a level can be populated by M1 photoabsorption; therefore, the study of near-barrier electrofission angular distributions offers a unique opportunity to sample the M1 strength, as described at length in Arruda-Neto et al (1980a) and Arruda-Neto et al (1980b).

II. METHOD OF ANALYSIS

The technique used here (and described in Arruda-Neto et al (1980b)) for obtaining the M1 component in the photofission channel requires detailed electrofission angular-distribution data near the fission barrier as well as accurate photofission cross sections measured with real photons. The

combined analysis of electrofission and photofission cross sections, using the virtual-photon formalism (Arruda-Neto et al 1978, 1980b), allows the determination of the cross section σ^{Add} (which represents the contributions of the "additional multipoles," i.e., those other than E1):

$$\sigma^{Add}(\omega) = \sigma^{E2}(\omega) + \left\langle \frac{N^{M1}(\omega,E_e)}{N^{E2}(\omega,E_e)} \right\rangle \sigma^{M1}(\omega) \qquad (1)$$

where the $\sigma^{\lambda L}$ represent the partial photofission cross sections for the transitions λL (λ identifies the electric or magnetic character of the transition and L its multipolarity), and $N^{\lambda L}$ is the virtual-photon spectrum calculated in DWBA (Soto Vargas et al 1977). The neglect of E0 and E3 contributions to the photofission process is justified in Arruda-Neto et al (1980b). The disentanglement of the E2 and M1 components in σ^{Add} is accomplished by the use of the electrofission-fragment angular distributions. The electrofission differential cross section for L=1 and 2 (λ=E and/or M) is given by (Arruda-Neto et al (1980a))

$$\frac{d\sigma_e(E_e,\theta_f)}{d\Omega_f} = A(E_e) + B(E_e)\sin^2\theta_f + C(E_e)\sin^2 2\theta_f \qquad (2)$$

where E_e is the incident electron energy and θ_f is the angle of the fission fragment with respect to that of the incident electron beam. The coefficient C contains only E2 contributions; for excitation energies near the fission barrier we get (Arruda-Neto et al (1980a))

$$C(E_e) = \frac{15}{32\pi} \int_0^{E_e} \sigma^{E2}(\omega)\, N^{E2}(\omega, E_e)\, \frac{d\omega}{\omega} \quad . \tag{3}$$

The cross sections $\sigma^{Add}(\omega)$ for the even-even uranium isotopes as well as the coefficients C were determined at this Laboratory from electrofission experiments (Arruda-Neto et al 1978, 1980a, 1980b, 1981, 1982). Only for ^{236}U have we performed a detailed search for a concentration of M1 strength (Arruda-Neto et al 1980b). The method of data analysis for the delineation of the E2/M1 components near the fission barrier is: (1) we combine eqns. (1) and (3) to get

$$C(E_e) = \frac{15}{32\pi} \int_0^{E_e} \left[\sigma^{Add}(\omega) - \left\langle \frac{N^{M1}(\omega, E_e)}{N^{E2}(\omega, E_e)} \right\rangle \sigma^{M1}(\omega) \right] N^{E2}(\omega, E_e)\, \frac{d\omega}{\omega} \quad ; \tag{4}$$

(2) we represent $\sigma^{M1}(\omega)$ by a Breit-Wigner curve, and its parameters are obtained from a least-squares fit of the expression in eqn. (4) to the experimentally determined coefficients C (see section IV). (We note that theory says nothing about the way in which the M1 strength might spread into the threshold region).

Some important points about the ingredients of this method of analysis should be stressed here:

(1) The coefficient C is a well established experimental quantity, especially for ^{238}U; the electrofission angular distributions for ^{238}U measured by the Giessen group (Aschenbach et al 1979) agree very well with our own results (see, e.g., Fig. 2 of Aschenbach et al (1979)).

(2) DWBA calculations for the virtual-photon spectra at low excitation energies (5-7 MeV), where nuclear finite-size effects are negligible, were tested (at the 20% level) for high-Z nuclei and E2-spectra (Arruda-Neto et al 1980c). Other tests for E1 virtual-photon calculations (at the 10% level) can be found in the literature (Nascimento et al 1975, Dodge et al 1983). However, no experimental test for calculations of M1 virtual-photon spectra has been performed; and in the light of these limitations of the calculated spectra, the absolute uncertainties given below are large. Their relative values, on the other hand, are determined much more precisely.

(3) Finally, the cross section σ^{Add} also plays an important role in the delineation of the M1 component for actinide nuclei. To obtain σ^{Add} we need accurate electrofission cross sections. For ^{238}U there is disagreement among the absolute electrofission cross-section results from several laboratories (see, for example, Arruda-Neto and Berman (1980)). However, below ~10 MeV (the energy region under investigation in the present work) the existing data agree reasonably well (Arruda-Neto et al 1978, Aschenbach et al 1979, Ströher et al 1981).

III. RELEVANT THEORY

The cross section for the absorption of photons by a single isolated level $(\omega_n; I_n)$, where ω_n is the level energy and I_n its spin, is (Hayward 1964)

$$\sigma_n(\omega) = 2\pi \lambda\!\!\!^{-}{}_n^2\, g_n\, \frac{\Upsilon_n}{\Gamma_n}\, \frac{(\omega\, \Gamma_n)^2}{(\omega_n^2 - \omega^2)^2 + (\omega\, \Gamma_n)^2} \tag{5}$$

where γ_n is the ground-state radiation width; Γ_n is the total width of the state $(\omega_n; I_n)$, that is, the sum of the partial widths associated with its decay to all lower states; ω is the incident photon energy; $g_n = \frac{2I_n+1}{2I_o+1}$ is the statistical factor; and $\lambda\!\!\!^{-}_n = \frac{\hbar c}{\omega_n}$.

The cross section integrated over the absorption line is given by

$$\int_{\text{resonance}} \sigma_n(\omega)\,d\omega = 2\pi \lambda\!\!\!^{-2}_n\, g_n \frac{\gamma_n}{\Gamma_n} \left(\frac{1}{2}\pi\Gamma_n\right) = (\pi\lambda\!\!\!^{-}_n)^2\, g_n \gamma_n \quad . \qquad (6)$$

The physical quantity of interest which is a property of the nuclear levels involved in the transition, to be extracted from the experiment, is the well known <u>reduced transition probability</u> $B(\lambda L;\omega_n)$ or, rigorously speaking, $B\!\uparrow(\lambda L;\omega_n)$, because in our case we have a photoabsorption process. In the long-wave length limit, $B\!\uparrow(\lambda L;\omega_n)$ is simply related to the integrated cross section (Eisenberg and Greiner 1970, Bartholomew <u>et al</u> 1973, Überall 1971):

$$\int_{\text{resonance}} \sigma_n(\omega)\,d\omega = (2\pi)^3 \alpha \frac{(L+1)}{L[(2L+1)!!]^2}\, \omega_n^{2L-1}\, B\!\uparrow(\lambda L;\omega_n) \quad . \qquad (7)$$

For M1 transitions in particular, we have

$$\int_{\text{resonance}} \sigma_n(\omega)\,d\omega = 4.45 \times 10^{-2}\, \omega_n \left[\frac{B\!\uparrow(M1;\omega_n)}{\mu_N^2}\right] \text{(MeV mb)} \qquad (8)$$

where μ_N is the nuclear magneton, and (Shapiro and Emery 1969)

$$\frac{B\!\uparrow(M1;\omega_n)}{\mu_N^2} = \frac{3}{4}\pi \sum_{M_n,\kappa} |\langle I_n M_n | M(M1,\kappa) | I_o M_o \rangle|^2$$

where $M(M1,\kappa) = \sum_{j=1}^{A} [g_\ell(j)\ell_\kappa(j) + g_s(j)s_\kappa(j)]$.

The M1 strength is often expressed in terms of the so called "reduced width" defined as (Shapiro and Emery 1969)

$$k_{M1}(\omega) = \frac{\gamma_n(M1)}{\omega^3 D} \qquad (9)$$

where D and ω are in MeV and γ_n is in eV; D is the average spacing in the region of the final state of levels of the same spin and parity. The terminology appearing in the literature is somewhat confusing. For example, if we define γ_n in MeV in eqn. (9), the "new" k_{M1} is now called the "strength function" f_{M1}, that is (Bartholomew <u>et al</u> 1973)

$$f_{M1}(\omega) = 10^{-6}\, k_{M1}(\omega) = \frac{2.6 \times 10^{-7}\, \sigma_n^{M1}(\text{mb})}{g_n\, \omega} \text{(MeV}^{-3}) \quad . \qquad (10)$$

After some algebra it is straightforward to show that

$$\frac{B\!\uparrow(M1;\omega_n)}{\mu_N^2} = 86.5\, g_n \int_{\text{resonance}} k_{M1}(\omega)\,d\omega \quad . \qquad (11)$$

In the present study, σ_n represents the photo-absorption cross section for the excitation of an even-even nucleus from its ground state $(I_o^\pi = 0^+)$ to a state $I^\pi = 1^+$ in the continuum. However, what we observe is the fission decay mode near the barrier. In this sense, the low-lying saddle-point levels act like a filter; that is, the fission decay proceeds only through the level $(I^{\pi},K) = (1^+,1)$ of the K=1 rotational band (Albertson and Forkman 1965, Vandenbosch and Huizenga 1973) of the transition nucleus (saddle-point). Therefore, the cross section $\sigma^{M1}(\omega)$ appearing in eqn. (4) can be written as

$$\sigma^{M1}(\omega) = \sigma_n(\omega)\,\frac{\Gamma_f}{\Gamma}\,(1^+;\omega) \qquad (12)$$

where $\frac{\Gamma_f}{\Gamma}(1^+;\omega)$ is the branching ratio for the (γ,f) reaction proceeding through the low-lying fission level 1^+. [Because of the fact that $\Gamma_f/\Gamma \leq 1$, the M1 strength deduced from the photofission cross section $\sigma^{M1}(\omega)$ represents only a lower limit.]

IV. RESULTS

Figures 2(a), (b), and (c) show the experimentally determined coefficient C of the electrofission angular distributions for ^{234}U, ^{236}U, and ^{238}U, respectively (Arruda-Neto et al 1980a, 1980b, 1982). The experimental details and data-handling procedures are described at length in Arruda-Neto et al (1980b) and Arruda-Neto et al (1982). The dashed curves in Fig. 2 were obtained by the numerical integration of the experimentally determined $\sigma^{Add}(\omega)$ (Arruda-Neto et al 1978, 1980b, 1981) in the kernel of the integral defined by eqn. (4), assuming no M1 strength [that is, $\sigma^{M1}(\omega) = 0$]. Assuming a Breit-Wigner shape for $\sigma^{M1}(\omega)$, as discussed before, and repeating the integration (eqn. (4)) in the energy range of 5-7 MeV we obtain the solid curves in Fig. 2 which are in good agreement with the experimental points in this energy range. The best Breit-Wigner parameters obtained from this fitting procedure are given in Table 1. All of the solid curves agree with the data up to ~ 7 MeV; at this energy the opening of the K=1 fission channel acts to reduce C (Arruda-Neto et al 1980a), so that the fact that the data points shown in Fig. 2 fall below the solid curves above this energy is expected. We note here that in the present study we analyzed the ^{234}U angular-distribution data using an improved version of the analytical expression for the M1 virtual-photon spectrum, obtained from the routine VIRFO (Soto Vargas et al 1977, Soto Vargas 1979). We reanalyzed the ^{236}U and ^{238}U data as well; for the ^{236}U we deduced slightly different M1 parameters from those obtained previously (Arruda-Neto et al 1980b).

The M1 reduced transition probability (in units of μ_N^2) concentrated in the fission channel $\frac{B(M1)}{\mu_N^2}\cdot\frac{\Gamma_f(1^+)}{\Gamma}$ were calculated from the area of $\sigma^{M1}(\omega)$ using the definitions presented in section III. [We note again that the lack of any experimental test of the calculated M1 virtual-photon spectra makes it impossible to place a quantitative overall systematic uncertainty on these values of B(M1) or γ_n.] In this analysis, we assumed the excitation of a single 1^+ state of the transition nucleus. It is worth remembering that at energies above ~ 7 MeV (above the pairing gap), the level densities in ^{238}U are high enough that one would expect more than one 1^+ state to be excited. However, below 7 MeV (the energy region under investigation here), M1 photoexcitation occurs through the overlap between the virtual-photon energy and the 1^+ level energy, and thus the uncertainty introduced into the derived M1 strength by the assumption of a Breit-Wigner lineshape is minimized.

Also given in Table 1 is the theoretical result for $B(M1)/\mu_N^2$ that we derived from magnetic dipole gamma-ray strength function (more specifically, the average reduced width) computed by Shapiro and Emery (1969) for the reaction $^{238}U(n,\gamma)^{239}U$, on the basis of the Nilsson model, pairing, and a residual spin-spin interaction. They found that the M1 strength functions for some strongly deformed heavy nuclei show peaks (see Fig. 2 of Shapiro and Emergy (1969)). The reason to present these theoretical results, which refer to the M1 gamma-ray decay for ^{239}U, together with those of the present work, is to provide a yardstick for our own results. However, when the ground-state I^{π} is not 0^+, then even in lowest order we do not expect the M1 strength to be concentrated in one excited state but rather to be distributed over several states, such as is permitted by angular-momentum coupling. The total transition strength to these final states, however, should be approximately equal to the strength of the $0^+ \rightarrow 1^+$ transition.

Our results for the M1 strength should be interpreted as lower limits, because $\Gamma_f(1^+)/\Gamma \leq 1$. For ^{234}U and ^{236}U the M1 strength peaks below B_n; therefore, it is reasonable to consider an extreme assumption for the fission probability, namely $\Gamma_f(1^+)/\Gamma \simeq 1$. On the other hand, the M1 fission decay of ^{238}U competes with neutron emission; therefore, for this nucleus it is reasonable to assume that $\Gamma_f(1^+)/\Gamma \simeq 0.25$, the value for the fission decay of the GDR (Caldwell et al 1980). The above assumptions for $\Gamma_f(1^+)/\Gamma$ are somewhat speculative; however, even the lower limit shows that the concentration of M1 strength is substantial, especially for ^{238}U. Such a concentration of M1 strength does not indicate, necessarily, the proximity of a giant M1 resonance, but our results suggest this possibility. In this regard, we would like to call attention to two mean quantities we derived from our results (with the dispersion from the mean): $\langle\omega_x A^{1/3}\rangle \simeq 38 \pm 2$ MeV, and $\langle\Gamma_x/\omega_x\rangle = 0.23 \pm 0.01$, to be compared with the values given by Hanna (1977) for the GMDR (see section I) and with the theoretical results from Shapiro and Emery (1969) (see Table 1).

As a by-product of our determination of the M1 photofission cross sections we obtain the E2 photofission cross sections for $^{234,236,238}U$ (see eqn. (1)). In Table 2 the E2 results are given, expressed in terms of the reduced transition probability B(E2), in the energy region 5-7 MeV, plus those deduced from the photofission angular-distribution experiments performed at Lund (Lindgren et al 1978). There is reasonable agreement, within the experimental uncertainties, between the two sets of values, and good agreement between their average values. This comparison with the photofission angular distributions is a stringent test of our technique and results.

V. CONCLUSION

Prior to the present measurements, no M1 states had been reported for actinide nuclei, either from hadron or from electron experiments. The evidence presented here on the M1 strength for the even-even uranium isotopes shows a substantial concentration in the fission channel just above the fission barrier. However, the details of the experimental situation for these heavy nuclei need clarification, both as to the question of the existence of a general giant M1 resonance, and to its quenching.

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TABLE 1

M1 strength parameters.

Nucleus	B_n (a) (MeV)	Peak (MeV)	Width (MeV)	$\frac{B\uparrow(M1)}{\mu_N^2}\cdot\frac{\Gamma_f(1^+)}{\Gamma}$	$\left\langle\frac{\Gamma_f(1^+)}{\Gamma}\right\rangle$ (b)	$\frac{B\uparrow(M1)}{\mu_N^2}$
^{234}U	6.8	6.4 ± 0.3	1.4 ± 0.2	5.8 ± 1.8 (d)	≤ 1	> 6
^{236}U	6.5	5.8 ± 0.2	1.3 ± 0.2	4.0 ± 1.2 (d)	≤ 1	> 4
^{238}U	6.15	6.5 ± 0.3	1.5 ± 0.2	4.1 ± 1.2 (d)	~ 0.25	~ 16
^{239}U (c)	–	6.2	2.0	–	–	18.5

(a) Neutron binding energy (from Caldwell et al (1980)).

(b) See discussion in the text.

(c) From theory (Shapiro and Emery (1969)).

(d) The uncertainties quoted here include no systematic uncertainty in the calculated M1 virtual-photon spectra.

TABLE 2

E2 reduced transition probability, in the energy region 5-7 MeV, for the $(2^+,0)$ fission channel.

Nucleus	$B(E2)\cdot\frac{\Gamma_f(2^+)}{\Gamma}$ (fm^4)	
	(a)	(b)
^{234}U	1200 ± 240	1920 ± 360
^{236}U	1440 ± 220	890 ± 220
^{238}U	710 ± 120	830 ± 120

(a) Present work.

(b) Derived from the cross sections published in Lindgren et al (1978).

FIGURE CAPTIONS

Fig. 1 - Schematic diagram of some possible collective band structures for an even-even transition nucleus with a stable quadrupole deformation (adapted from Vandenbosch and Huizenga (1973)).

Fig. 2 - Absolute values for the coefficient of the $\sin^2 2\theta_f$ term in the electrofission differential cross section $C(E_e)$ (eqn. (2)), as a function of the incident electron energy (data points) obtained from the measured angular distributions for (a) ^{234}U, (b) ^{236}U, and (c) ^{238}U. The solid curves represent this coefficient after M1 subtraction (for other details, see text).

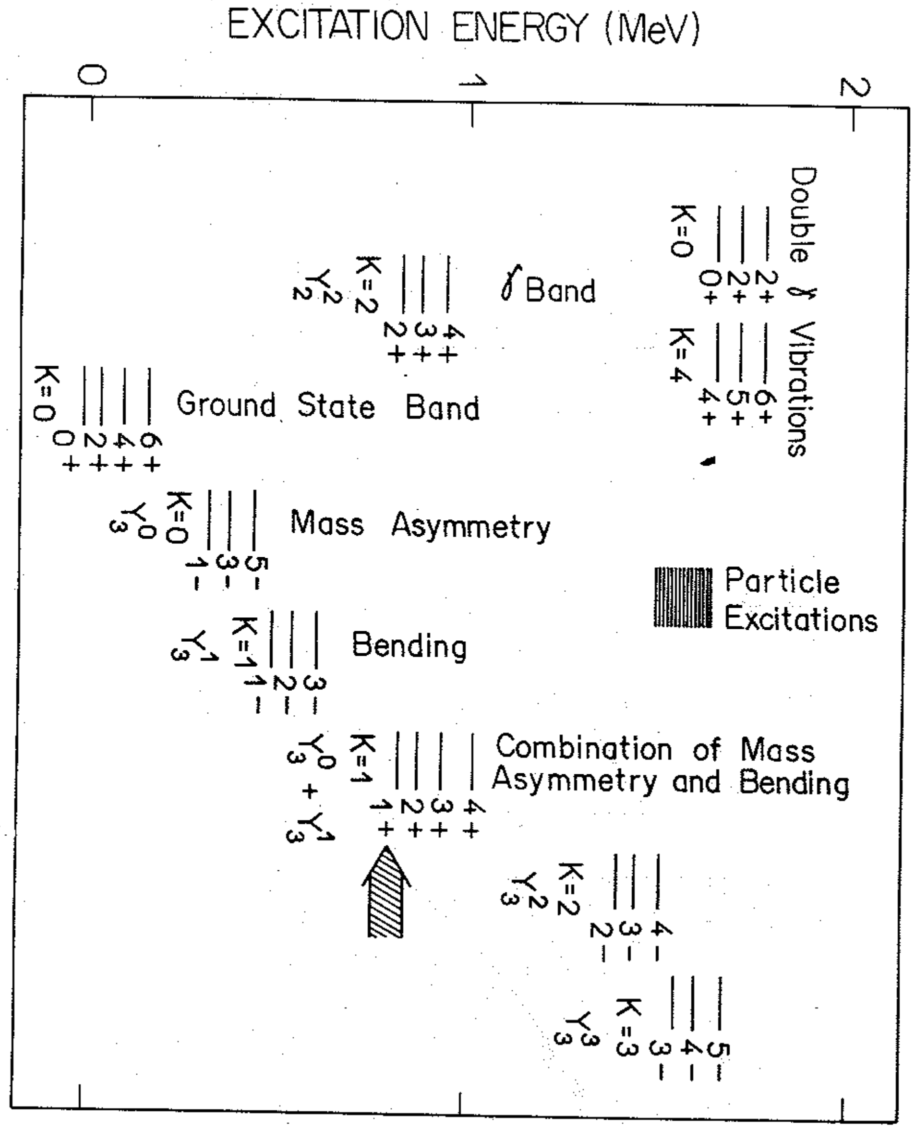

Fig. 1

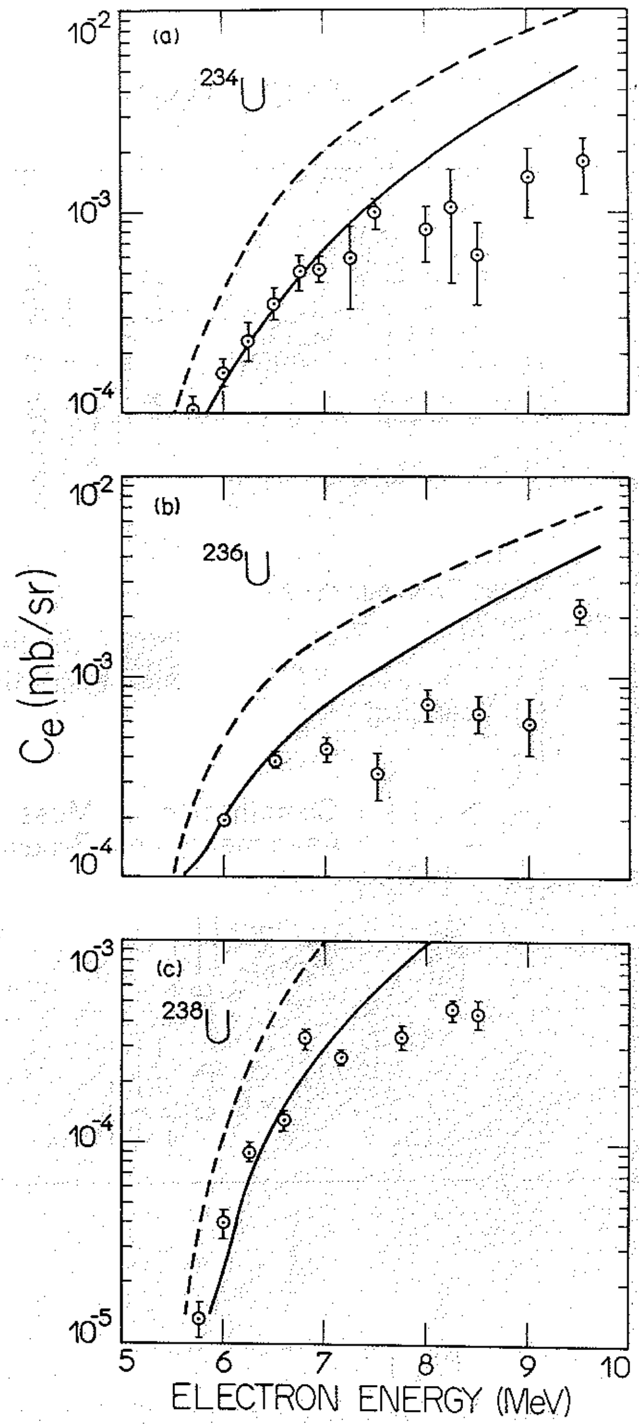
(a)
^{234}U
(b)
^{236}U
(c)
^{238}U
C_e (mb/sr)
10^{-2}
10^{-3}
10^{-4}
10^{-5}
5
6
7
8
9
10
ELECTRON ENERGY (MeV)

Fig. 2