Electrofission and photofission of ^{238}U in the energy range 6–60 MeV†

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Experimental results are presented for the electrofission and photofission of ^{238}U in the energy range 6 to 60 MeV. The importance of the inclusion of the Coulomb corrections in the calculation of the virtual photon spectrum is emphasized and the relative $E1$ contribution to the electrofission process has been evaluated using the distorted-wave Born-approximation analysis of the experimental data.

[NUCLEAR REACTIONS, FISSION Measured electrofission and photofission cross sections of ^{238}U in the energy range 6–60 MeV. Deduced relative $E1$ contribution to the electrofission process using DWBA analysis of the experimental data.]

I. INTRODUCTION

The study of the photonuclear reactions, and of photofission in particular, has been carried out in several laboratories since the discovery of nuclear fission. Although electron-induced nuclear reactions have also been studied in the last two decades, there are no data available on the electron-induced fission cross sections of ^{238}U in the giant dipole resonance region and only a few investigations[1-3] have been reported for energies above this region.

The use of electrons and that of photons to induce nuclear reactions is equivalent in the sense that in both cases, the nuclei are excited by means of the electromagnetic interaction. In the case of electro-excitation, this interaction takes place through the virtual photon spectrum as compared to that through the real photons in the case of photoexcitation. The virtual photon spectrum differs from the real photon spectrum mainly in that: (1) it contains both longitudinal as well as the transverse photons, and (2) it depends strongly upon the multipolarity of the photons. The electroexcitation process can therefore, at least in principle, serve as an important tool for the investigation of the multipolarities of the transitions involved.

Using plane waves for the incoming as well as the outgoing electrons, Thie, Mullin, and Guth[4] obtained the virtual photon spectrum for the $E1$, $E2$, and $M1$ transitions in the plane-wave Born approximation (PWBA). However, Gargaro and Onley[5] have recently calculated the expressions for the virtual photon spectrum for all the multipole orders by using a distorted-wave treatment (DWBA). The electron induced fission experiments mentioned earlier were reported before this distorted-wave calculation of the virtual photon spectrum became available. Therefore the spectrum calculated in PWBA was used in those investigations to analyze the experimental data. Since these studies were made at very high electron energies (60–1000 MeV), the use of PWBA in the data analysis could be justified. However, in the energy region of the present investigation, it has been shown recently[5,6] that this procedure is subject to large errors depending on the electron bombarding energies and the target atomic numbers.

The interaction of photons with nuclei, in general, exhibits a typical characteristic: a resonance in the photonuclear cross sections at an energy roughly equal to $(80/A^{1/3})$ MeV, attributed to the electric dipole ($E1$) mode of excitation, and usually called the "giant resonance." For photofission of ^{238}U, this resonance is located at an energy of 14 MeV. Fission fragment angular distribution studies[7,8] of the heavier actinides, at energies near fission threshold, have indicated the presence of a significant electric quadrupole component. Also, Bohr and Mottelson[9,10] have recently predicted the possible existence of an isoscalar and of an isovector quadrupole giant resonance at energies roughly equal to $(60/A^{1/3})$ and $(135/A^{1/3})$ MeV, respectively. There have been several measurements recently which confirm the existence of such resonances.[11]

One of the important characteristics of the DWBA calculation of the virtual photon spectrum is that it predicts a significantly larger intensity for the electric quadrupole mode (and also for the magnetic dipole mode) as compared to that for the electric dipole mode. This is because the Coulomb correction expected for the electric quadrupole and magnetic dipole levels is much greater than that for the electric dipole. This useful property of the virtual photon spectrum then offers an opportunity to use the process of electroexcitation as an important tool for studying the otherwise weak multipole components, other than electric dipole, in the nuclear spectrum in more detail than is possible with real photons alone.

From the experimental point of view, the cross sections for the electroexcitation processes are roughly $(1/\alpha)$ times smaller than the corresponding cross sections for the photoexcitation. Here α is the fine structure constant. Thus the electroexcitation yields are much smaller than those for photoexcitation. However, it is much easier to obtain well focused intense electron beams than well collimated photon beams. It is also easier to measure electron beam intensities than photon beam intensities.

In the present paper, experimental results are presented for the photofission (20 to 60 MeV) and for the electrofission (6 to 60 MeV) of ^{238}U. Preliminary results of these measurements were reported earlier[12] but have been corrected in the present paper. The relative $E1$ contribution to the electrofission process has been evaluated using the DWBA analysis of the experimental data.

II. EXPERIMENT

The electron beam was provided by the University of São Paulo linear accelerator. It consists of two 3-m SLAC-type accelerating sections. The electron beam is supplied by a 100-kV pulsed electron gun at a repetition rate of 60 and 120 Hz. After acceleration, the beam is analyzed by two deflecting magnets and then focused on the target using two quadrupole magnetic lenses. The maximum current of the analyzed beam is 1 μA and the electron energy resolution is 1%.

An uranium target (natural UO_2) was placed in the center of a cylindrical vacuum chamber, making an angle of 45° with the incident beam direction. A simple illustration of the target geometry is shown in Fig. 1. The fission chamber used in the experiment was 21 cm high and had a diameter of 40 cm. The target was prepared by electrodeposition[13] on a 7-μm aluminum backing. A target thickness of 172 $\mu g/cm^2$ was measured by absolute α particle spectrometry using surface barrier solid state detector and the uniformity of the UO_2 deposit was checked by irradiating the target with a known neutron flux at the Instituto de Energia Atômica de São Paulo research reactor and measuring the (n,f) fission fragment spatial distribution with a mica foil.

The fission fragments were detected using mica foils placed at different angles with respect to the incident beam direction. The mica foils were preetched in 50% hydrofluoric acid for about 20 h to develop the fossil fission background and for about 10 h after the irradiations.[14] Fission tracks were counted using an optical projection microscope with a 100× magnification.

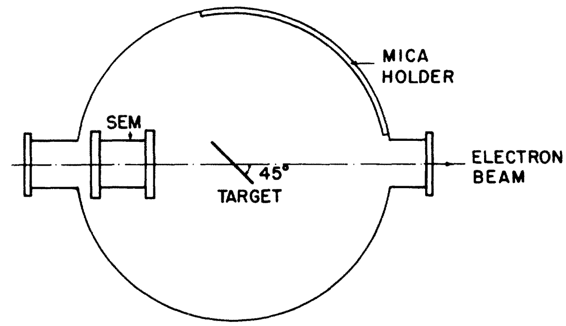

FIG. 1. A simple illustration of the target geometry used in the experiment.

For the production of bremsstrahlung, an aluminum radiator with a thickness equal to 2.08×10^{-2} radiation length was placed before the target. The electron beam was monitored by a Faraday cup for the electrofission measurements and by a secondary emission monitor (SEM),[15] placed before the radiator and the target, for the photofission experiment.

The possible contamination of the electron beam with bremsstrahlung and neutrons in the electrofission measurements was checked experimentally and was found to be negligible. The bremsstrahlung produced in the SEM aluminum foils during the photofission measurements contributed about 4% of the total bremsstrahlung induced fission yield and this was corrected for. Corrections for the finite thickness of the target and radiator were made using the method described by Barber.[16]

III. RELEVANT THEORY AND METHOD OF ANALYSIS

The bremsstrahlung induced fission cross section as a function of the end point photon energy, E_0, can be written as

$$\sigma_B(E_0)=\int_0^{E_0}\sigma_\gamma(E)K^B(E,E_0)dE\,, \tag{1}$$

where

$$\sigma_\gamma(E)=\sum_{\lambda L}\sigma_\gamma^{\lambda L}(E)$$

is the total photofission cross section; $\sigma_\gamma^{\lambda L}(E)$ represents the partial cross section for fission induced by λL photons where L defines the multipole order of the transition and λ its electric or magnetic character. $K^B(E,E_0)$ is the bremsstrahlung spectrum for a thin radiator.

The unfolding of Eq. (1) allows the determination of $\sigma_\gamma(E)$ from the experimentally measured $\sigma^B(E_0)$. However, as Rabotnov *et al.*[8] have pointed out, the determination of $\sigma_\gamma(E)$ from this equation is an improperly formulated problem and is subject to uncertainties due to the "swinging" of the solutions. This feature of the unfolding process then calls for caution in the interpretation of any irregular behavior of $\sigma_\gamma(E)$ at certain energies. The resolution in this method is determined mainly by the slopes of the bremsstrahlung spectrum and of the photofission yield curve. We have used the thin radiator bremsstrahlung spectrum for intermediate screening[17] and the photon difference method[18] for the unfolding of $\sigma_\gamma(E)$ from Eq. (1). The unfolding technique was satisfactory as is shown later in Sec. IV.

The electron induced fission cross section $\sigma_e(E_0)$ can be analyzed in terms of the photofission cross section by means of the virtual photon formalism. In this formalism[4] the virtual photon spectrum is defined as:

$$\frac{N^{\lambda L}(E,E_0)}{E}=\frac{1}{\sigma_\gamma^{\lambda L}(E)}\frac{d\sigma_e^{\lambda L}(E,E_0)}{dE}$$

which reduces to

$$\sigma_e^{\lambda L}(E_0)=\int_0^{E_0}\sigma_\gamma^{\lambda L}(E)N^{\lambda L}(E,E_0)\frac{dE}{E}. \tag{2}$$

In Eq. (2), E_0 is the incident electron energy, $\sigma_e^{\lambda L}(E_0)$ is the partial cross section for the fission induced by λL virtual photons, and $N^{\lambda L}(E,E_0)$ is the virtual photon spectrum. It is necessary here to use the virtual photon spectrum calculated in DWBA because in PWBA, it is underestimated.[5,6] As mentioned earlier, Gargaro and Onley have recently published a DWBA calculation for the virtual photon spectra for all multipole orders. Unfortunately, this calculation requires very long computer times and therefore analytical expressions have been obtained recently[6,19] by a best fit to a few points calculated for each spectrum using the original DWBA program.[5] We have used these expressions in the analysis of the experimental data in the present work. The total electrofission cross section is then given by

$$\sigma_e(E_0)=\sum_{\lambda L}\sigma_e^{\lambda L}(E_0). \tag{3}$$

Besides the electric dipole, the other modes of photoabsorption in the energy range of the present investigation for the doubly even actinide nuclei could be electric quadrupole and magnetic dipole:

$$\sigma_\gamma(E)=\sigma_\gamma^{E1}(E)+\sigma_\gamma^{\mathrm{ad}}(E). \tag{4}$$

Similarly, for the electroexcitation process:

$$\sigma_e(E_0)=\sigma_e^{E1}(E_0)+\sigma_e^{\mathrm{ad}}(E_0), \tag{5}$$

where $\sigma_\gamma^{\mathrm{ad}}$ and σ_e^{ad} represent the contributions due to multipoles other than the electric dipole.

There is evidence[20] for strong $M1$ transitions between 5 and 9 MeV in heavy nuclei up to ^{208}Pb but not as yet for doubly even actinides except for a possible explanation[20,21] in terms of such an excitation of the 6.2-MeV peak in photofission cross sections of ^{238}U. Some analyses[22-24] of the angular distribution data for the photofission of ^{238}U indicate a significant quadrupole component in the energy region 6–9 MeV. A preliminary analysis[25] of our electrofission angular distribution data also supports this conclusion. Since the electroexcitation process enhances the magnetic dipole mode as well as the electric quadrupole mode almost equally,[5] the presence of a significant $\sin^2 2\theta$ component in our angular distribution measurements suggests that the $M1$ component, even if present, is at least not dominant.

The relative contribution of the $E1$ excitation with respect to the total electroexcitation process can then be obtained from the ratio

$$R(E_0)=\frac{\sigma_e^{E1}(E_0)}{\sigma_e(E_0)}, \tag{6}$$

where $\sigma_e(E_0)$ is the experimentally determined electrofission yield $[\sigma_e^{\exp}(E_0)]$ and $\sigma_e^{E1}(E_0)$ can be calculated with a good approximation as

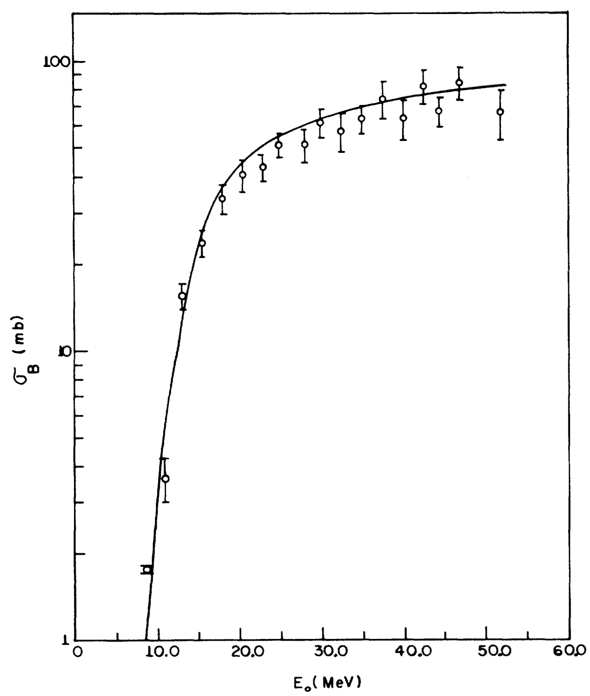

FIG. 2. Bremsstrahlung induced fission yield σ_B of ^{238}U as function of the maximum photon energy E_0. The solid line represents the yield curve obtained by folding in the photofission cross section of ^{238}U (as shown in Fig. 3) with the bremsstrahlung spectrum for a thin radiator (Ref. 17).

$$\sigma_e^{E1}(E_0) \approx \int_0^{E_0} \sigma_\gamma(E) N^{E1}(E, E_0) \frac{dE}{E}. \tag{7}$$

In Eq. (7), $\sigma_\gamma(E)$ is the photofission cross section obtained by unfolding Eq. (1) and $N^{E1}(E, E_0)$ is the virtual photon spectrum calculated in the DWBA. The value of $\sigma_e^{E1}(E_0)$ obtained in this approximation is overestimated by an amount equal to the integral

$$\int_0^{E_0} \sigma_\gamma^{\text{ad}}(E) N^{E1}(E, E_0) \frac{dE}{E}.$$

The value of this integral is of the order of 3% of σ_e^{E1} [as defined in Eq. (2)] at 8 MeV and of the order of 1% above 10 MeV.[26] These values are consistent with the photoabsorption estimates for the different multipole excitations as given by Axel[27] and by Blatt and Weisskopf,[28] respectively.

IV. RESULTS AND DISCUSSION

The measurement of the photofission yield of ^{238}U as a function of the maximum photon energy E_0 is shown in Fig. 2. Below 18 MeV, where only a few points have been measured, we have folded Veyssière *et al.*'s monochromatic photon cross sections[29] with the bremsstrahlung for a thin radiator[17] to obtain the yield curve, the solid line below 18 MeV in Fig. 2. This yield curve is in close agreement with the points. The yield curve is joined to our yield curve above 18 MeV and the total curve unfolded using the photon difference method. The resulting photofission cross sections, in the entire energy range from 6 to 60 MeV, are shown as a solid line in Fig. 3. The experimental points shown on the solid line are the original data of Veyssière *et al.*[29] and the close proximity of these points to the solid line is a check on the unfolding technique used.

The results obtained in the present experiment for the electrofission of ^{238}U, σ_e, as a function of the incident electron energy E_0 are shown in Fig. 4. This diagram also shows a comparison of our experimental results with the semitheoretical curves $E1$(DW), $E1$(PW), and QD (quasideuteron). The $E1$(DW) curve was obtained by evaluating the folding integral of Eq. (7) with $\sigma_\gamma(E)$ as obtained in the present work (Fig. 3) and the virtual photon spectrum as calculated in the DWBA. The $E1$(PW) curve has the same meaning except that the virtual photon spectrum used here was obtained in PWBA.

The curve labeled by QD also refers to σ_e^{E1} but with $\sigma_\gamma(E)$ calculated on the basis of the quasideuteron model[30,31] by the expression

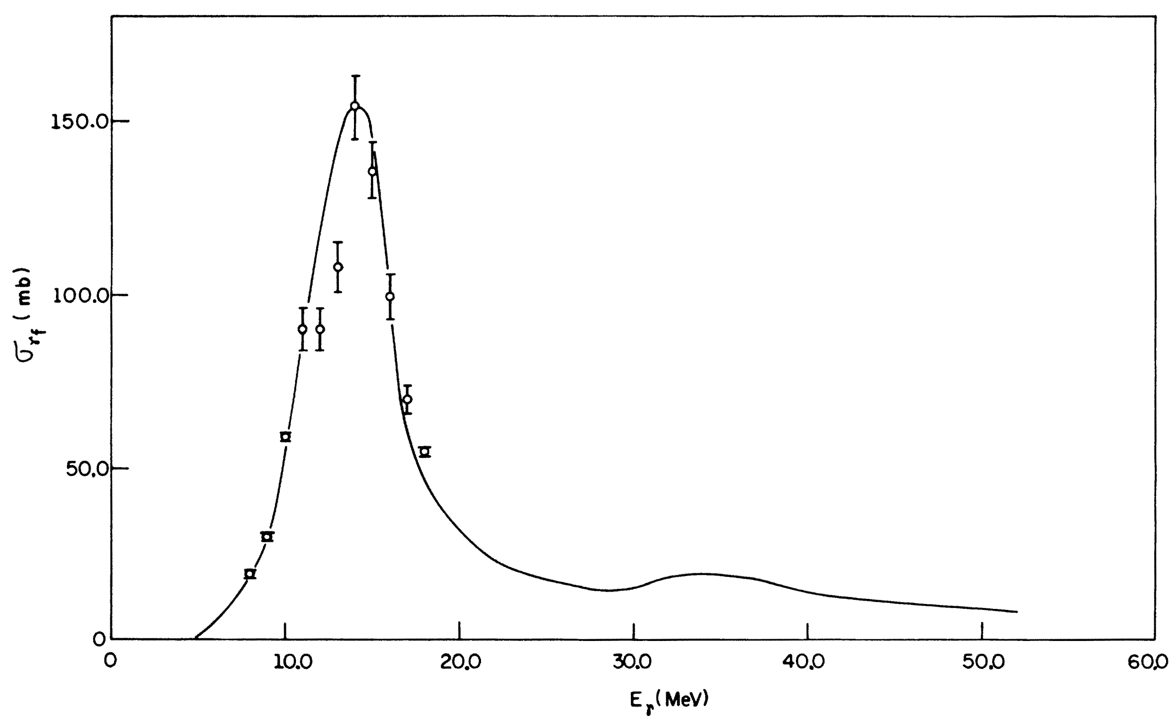

FIG. 3. Photofission cross section $\sigma_{\gamma f}$ of ^{238}U as a function of photon energy E_γ is shown by the solid curve obtained by the unfolding of the bremsstrahlung induced fission yields as described in the text. The experimental points shown are the monochromatic photon cross sections of Veyssière *et al.* (Ref. 29).

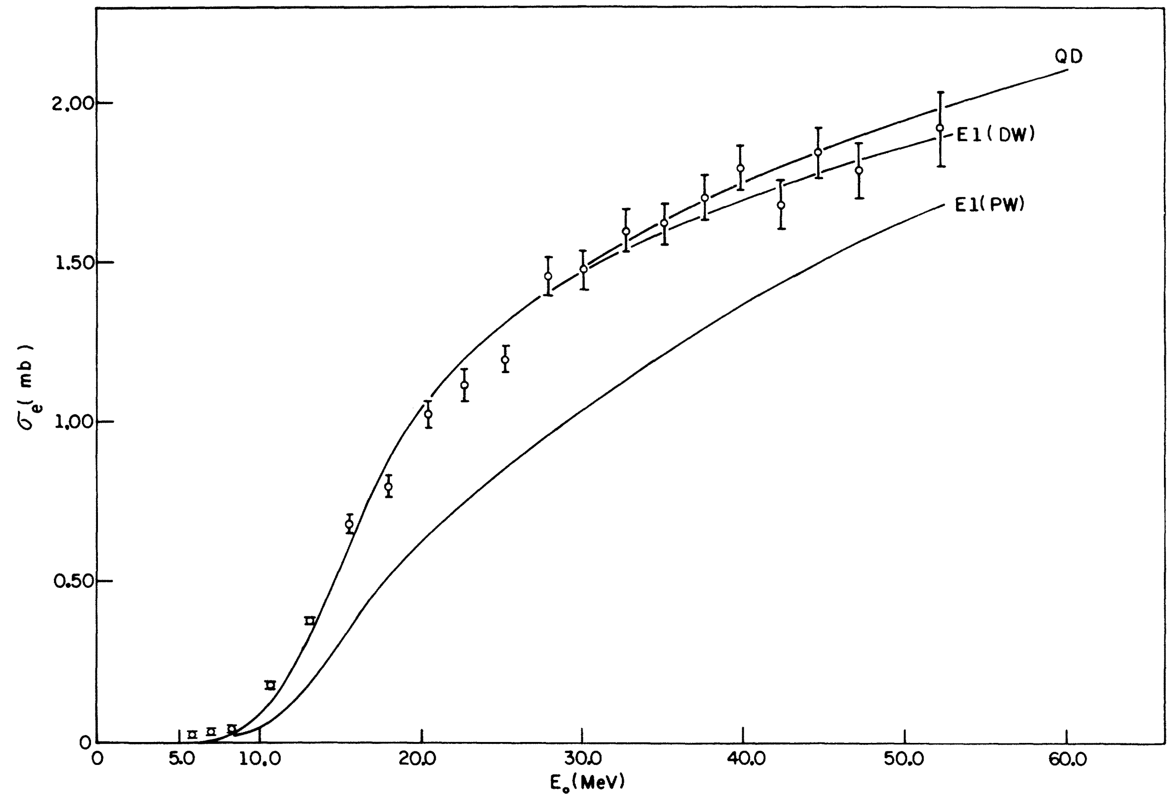

FIG. 4. Electron induced fission yield σ_e of ^{238}U as a function of the incident electron energy E_0. The solid lines labeled by $E1$(DW), $E1$(PW), and QD are the semitheoretical curves as explained in the text.

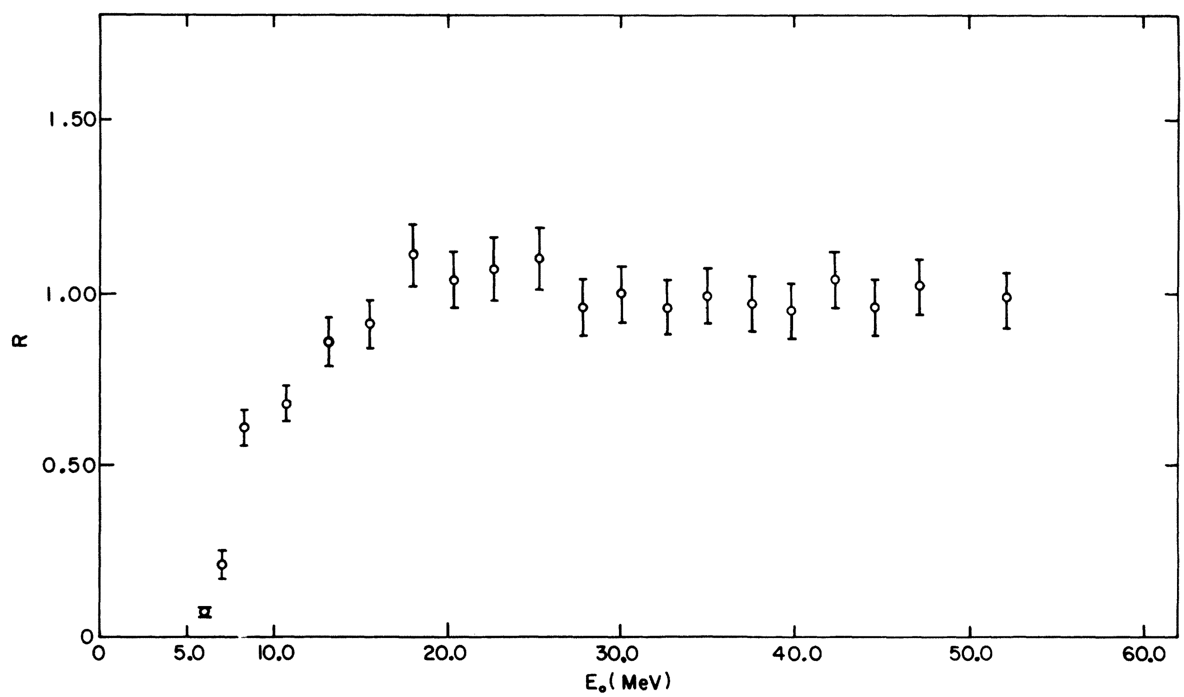

FIG. 5. The relative contribution of the $E1$ excitation with respect to the total electroexcitation process, as defined in Eq. (6) of the text by the ratio R, is plotted as a function of the incident electron energy E_0.

$$\sigma_\gamma(E) = \sigma_a^{\text{QD}}(E) P_f(E), \tag{8}$$

where $\sigma_a^{\text{QD}}(E)$ is the total photoabsorption cross section given by the quasideuteron model and $P_f(E)$ is the relative fission probability obtained from the theoretical expressions given by Nix and Sassi.[32] The results shown in Fig. 4 can be summarized as follows:

(a) The σ_e^{E1} curve obtained in PWBA underestimates by a large amount the electrofission yield, indicating thereby that PWBA calculation of the virtual photon spectrum is inadequate in analyzing the electrofission data in this energy region.

(b) The QD curve agrees reasonably with the experimental points above 30 MeV, indicating thereby that the quasideuteron process for the photoabsorption may be considered as a possible excitation mechanism to describe the behavior of the electrofission cross section above the giant dipole resonance. Since the fission barrier for ^{238}U is only 6 MeV, it is expected that the photofission cross section for this nucleus well above the fission barrier should essentially reflect the photon interaction cross section. Moretto *et al.*[3] have also found a reasonable agreement between their photofission measurements of ^{238}U and the quasideuteron model in the energy region 60–200 MeV.

(c) The $E1$(DW) curve agrees well with the experimental points in the entire energy region showing the dominance of the $E1$ giant resonance except at low energies, where for example at 8 MeV, the electric dipole excitation accounts for only about 60% of the total electrofission yield, leaving the rest to other excitation modes.

This behavior of the decreasing dominance of the electric dipole excitation mechanism in the low energy region can be better visualized in Fig. 5 where we have shown the ratio $R(E_0)$ as defined in Eq. (6), as a function of the incident electron energy E_0. The ratio fluctuates around unity in the energy region above 15 MeV but decreases rapidly in the low energy region. This provides quantitative evidence of a component, other than electric dipole, in the low energy photofission of ^{238}U. Although fission fragment angular distribution results of Rabotnov *et al.*[8] indicate a negligible amount of quadrupole component in the energy region 6–9 MeV, other measurements[22–24] do indicate the presence of a significant quadrupole component. A preliminary analysis[25] of our electrofission angular distribution data also suggests a large quadrupole component in this energy region. Whether this quadrupole component is distributed uniformly over a wide energy region or is concentrated in a specific energy interval to qualify as a quadrupole giant resonance remains to be seen. As mentioned earlier, Bohr and Mottelson[9,10] have predicted the existence of an isoscalar quadrupole giant resonance at an energy roughly equal to $(60/A^{1/3})$ MeV (~9.6 MeV for ^{238}U).

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