Electric quadrupole giant resonance in the photofission of ^{238}U

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(Received 2 March 1978)

The ^{238}U nucleus was studied measuring the electrofission yield and angular distributions of fission fragments, in the energy range of 5.5 to 28.3 MeV, using a new method of analysis. An $E2$ isoscalar giant resonance was found in the photofission cross section of ^{238}U. This resonance exhausts $(71\pm7)\%$ of the energy-weighted sum rule and is located at 9.9 ± 0.2 MeV with a width of 6.8 ± 0.4 MeV. The position of this resonance is in reasonable agreement with the Bohr and Mottelson prediction ($58A^{-1/3}$ MeV). The width of 6.8 ± 0.4 MeV is compatible with a possible triple splitting of the resonance. From the angular distributions of photofission fragments and yield measurements of multipoles other than $E1$, evidence of an $M1$ mixture in the energy region 6–7 MeV was found.

NUCLEAR REACTIONS, FISSION Electrofission of ^{238}U $(e, e'f)$, $E_0=5.5$ to 28.3 MeV, measured electrofission yield and angular distributions of fission fragments. Deduced $E2$ cross section and $E2$ isoscalar giant resonance parameters.

I. INTRODUCTION

In the last six years many data have been accumulated about "new giant resonances" and, in particular, about one localized just below the well-known electric dipole giant resonance (GDR) observed in photonuclear reactions. This new resonance has been related to a probable electric quadrupole oscillation of the nucleus under study. In light nuclei the excitation energy is about $60A^{-1/3}$ MeV and in heavy nuclei about $65A^{-1/3}$ MeV.[1]

Isoscalar and isovector electric quadrupole giant resonances (GQR), in the photoabsorption cross section of spherical nuclei, have been predicted by Bohr and Mottelson at energies approximately equal to $58A^{-1/3}$ and $135A^{-1/3}$ MeV, respectively.[2]

Some evidence of an electric quadrupole component ($E2$) in the photofission of ^{238}U has been pointed out in the literature in the past years, mainly through the study of the angular distribution of the photofission fragments.[3] These experiments, performed utilizing "bremsstrahlung", neutron capture γ rays, or annihilation photons, have provided the coefficients of the angular distribution function: $W(\theta)=a+b\ \sin^2\theta+c\ \sin^2(2\theta)$, in which the a and b contain both dipole and quadrupole contributions, and the c only quadrupole contribution. This coefficient has been used to estimate the $E2$ contribution. Nevertheless, this is only a lower limit for the total quadrupole cross section.

More recently some experiments have shown evidence of a GQR in the photoabsorption cross section of ^{238}U. Lewis and Horen[4] found a structure in the ^{238}U(p,p') spectra, between 10 and 13 MeV, tentatively attributed to an $E2$ giant resonance exhausting about 85% of the energy weighted sum rule (EWSR). Wolynec *et al.*[5] have shown by the study of the reaction ^{238}U$(e, e'\alpha)^{234}$Th, in the energy range 9–24 MeV, that their experimental data could be explained by a (γ, α) cross section with a Breit-Wigner shape, with a width of 3.7 MeV, localized at 8.9 MeV, and exhausting 50% of the EWSR. Arruda Neto *et al.*,[6] investigating the electrofission channel, have shown the existence of a concentration of components different from $E1$, in the energy region 6–9 MeV. The inelastic scattering of electrons was utilized by Houk *et al.*[7] in order to investigate giant resonances in ^{238}U. An $E2$ isoscalar resonance was evidenced by these authors at 9.9 MeV, with a width of 2.5 MeV, identified by the variation of the form factor with the momentum transfer and exhausting 40% of the EWSR.

The reaction channels most utilized in the study of the GQR have been (p,p') and (e,e'); the main limitations of these processes are the uncertainty associated with background subtraction and the dependence on nuclear models for the identification of the resonance multipolarity.

In this work, we report an experiment in which a GQR in the photofission cross section of ^{238}U was found. This resonance has been detected previously[8] but the experimental data were insufficient to provide a definite conclusion. In the present experiment the total yield for the reaction ^{238}U$(e, e'f)$ was carefully measured in the energy range 5.5–28.3 MeV. The reason for the choice of this reaction channel is that virtual photon spectra are more intense for $E2$ transitions

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than for $E1$.[9] As was indicated by Nascimento *et al.*,[10] this property of the virtual photon spectra makes the electroexcitation process a useful probe to investigate $E2$ transitions. The $E2$ component in the virtual photon spectra is more intense than the $E1$ component, which is not the case for the real photon spectra which contain all multipolar components in equal amounts. This fact enhances the ratio of $E2$ to $E1$ cross section in electroexcitation as compared with photoexcitation. The method for the extraction of the $E2$ component based on the virtual photon formalism is explained in Sec. III.

II. EXPERIMENTAL PROCEDURE

The electron and bremsstrahlung induced fission yields and some angular distributions of electrofission fragments have been measured by irradiating natural uranium targets with the electron beam of the University of São Paulo linear accelerator. The fission fragments were detected using mica foils placed at different angles with respect to the incident beam direction. The accelerator, scattering chamber, beam monitoring devices, and the fission fragment detection technique, as well as some other experimental details, have been described in a previous paper.[6]

Two targets, with thicknesses 310 $\mu g/cm^2$ and 434 $\mu g/cm^2$ have been used, and were prepared by molecular plating of $UO_2(NO_3)_2 \cdot 6H_2O$ on 7 μm aluminum backings.[11]

The electrofission yield was measured from 5.5 to 12.0 MeV, in steps of 0.25 MeV, and from 12.0 to 28.3 MeV, in steps of 0.5 MeV. The bremsstrahlung induced fission yields were measured at the energies 10.0, 12.0, 14.0, 16.0, and 18.0 MeV, by placing an aluminum radiator with a thickness of 1.04×10^{-2} radiation length before the target. This measurement was performed to determine the normalization constant K, as explained in Sec. III.

III. METHOD OF ANALYSIS

The method used to obtain the $E2$ component is based on the measurement of the electrofission yield $Y_{e,e'f}(E_0)$, and on the knowledge of the total cross section for photofission $\sigma_{\gamma,f}(E)$.

This method is general, in the sense that it may be extended to other reaction channels.

For the yield of the reaction $(e,e'f)$, using the virtual photon formalism, we may write the following:

$$Y_{e,e'f}(E_0)=\sum_{\lambda L}K\int_0^{E_0}\sigma_{\gamma,f}^{\lambda L}(E)N_v^{\lambda L}(E,E_0)\frac{dE}{E}\ , \tag{1}$$

where λ identifies the electric or magnetic character of the transition and L its multipolarity; $N_v^{\lambda L}(E,E_0)$ is the virtual photon spectrum calculated in the distorted-wave Born approximation (DWBA)[9]; E_0 is the electron incident energy and E the virtual or real photon energy; K is a normalization constant.

Making the assumption that, besides the $E1$ component, only two multipole components, $E2$ and $M1$, are present, we have

$$\begin{aligned}Y_{e,e'f}(E_0)=&K\int_0^{E_0}\sigma_{\gamma,f}^{E1}(E)N_v^{E1}(E,E_0)\frac{dE}{E}\\&+K\int_0^{E_0}\sigma_{\gamma,f}^{E2}(E)N_v^{E2}(E,E_0)\frac{dE}{E}\\&+K\int_0^{E_0}\sigma_{\gamma,f}^{M1}(E)N_v^{M1}(E,E_0)\frac{dE}{E}\ ,\end{aligned} \tag{2}$$

where

$$\begin{aligned}\sigma_{\gamma,f}(E)&=\sum_{\lambda L}\sigma_{\gamma,f}^{\lambda L}(E)\\&=\sigma_{\gamma,f}^{E1}(E)+\sigma_{\gamma,f}^{E2}(E)+\sigma_{\gamma,f}^{M1}(E)\end{aligned} \tag{3}$$

is the total photofission cross section.

In order to use Eqs. (2) and (3) to obtain $\sigma_{\gamma,f}^{E2}$, we need to make some assumptions in relation to $\sigma_{\gamma,f}^{M1}$. Evidence of $M1$ components was obtained mainly in light nuclei, and the results suggest their localization in the energy range $(30-45)A^{-1/3}$ MeV.[12] The virtual photon spectrum for $M1$ can be written

$$N_v^{M1}(E,E_0)=F(E,E_0)N_v^{E2}(E,E_0)\ ,$$

and in a range of a few MeV near the position of the peak of the $M1$ resonance (6–7 MeV for uranium) we may take $F(E,E_0)=F(E)$, and the average value of $F(E)$ in this energy region is $\langle F(E)\rangle\simeq3$, using the $M1$ spectrum from Ref. 9.

Eliminating $\sigma_{\gamma,f}^{E1}$ from (2) and (3), and grouping, we obtain

$$Y_{e,e'f}(E_0)-Y^*_{e,e'f}(E_0)=\Delta Y(E_0)=K\int_0^{E_0}\sigma_{\gamma,f}^{\rm add}(E)[N_v^{E2}(E,E_0)-N_v^{E1}(E,E_0)]\frac{dE}{E}\ , \tag{4}$$

where

$$Y^*_{e,e'f}(E_0)=K\int_0^{E_0}\sigma_{\gamma,f}(E)N_v^{E1}(E,E_0)\frac{dE}{E}\ . \tag{5}$$

and

$$\sigma_{\gamma,f}^{\rm add}(E)=\sigma_{\gamma,f}^{E2}(E)+F(E)\sigma_{\gamma,f}^{M1}(E)\ . \tag{6}$$

Equation (4) was obtained by making the approximation

$$N_v^{E2}(E,E_0)-\frac{N_v^{E1}(E,E_0)}{F(E)}\simeq N_v^{E2}(E,E_0)-N_v^{E1}(E,E_0)\,,$$

considering that $N_v^{E2}\simeq 5N_v^{E1}$ and $\langle F(E)\rangle\simeq 3$, in the region 6–7 MeV; the error is less than 10%. Above this energy region the error is even smaller owing to the enhancement of the $E2$ virtual spectra in relation to the $E1$ spectra.

The solution of the integral equation (4), with the indicated kernel, gives the cross section $\sigma_{\gamma,f}^{\text{add}}$, which represents the contributions of additional multipoles, other than $E1$. To obtain this solution, it is necessary to know the electrofission yield, as well as the value of $Y_{e,e'f}^{*}(E_0)$, defined by Eq. (5). The electrofission yield $Y_{e,e'f}(E_0)$ and the normalization constant K may be determined experimentally. The integral of Eq. (5) may be calculated numerically, since $\sigma_{\gamma,f}(E)$ can be found from photofission experiments.

The difference represented by Eq. (4) is significant at energies below the GDR, as N_v^{E2} or $N_v^{M1} > N_v^{E1}$. This enhances the additional multipoles ($E2$ and $M1$) as compared to $E1$, and makes $\Delta Y(E_0)$ an experimentally detectable quantity.

The normalization constant K may be determined by measuring the bremsstrahlung induced fission yield $Y_{B,f}(E_0)$ and by calculating the integral $\int_0^{E_0}\sigma_{\gamma,f}(E)N_B(E,E_0)dE$, where $N_B(E,E_0)$ is the bremsstrahlung spectrum, and $\sigma_{\gamma,f}(E)$ are values of the photofission cross section known from the literature, resulting in

$$K=\frac{Y_{B,f}(E_0)}{\int_0^{E_0}\sigma_{\gamma,f}(E)N_B(E,E_0)dE}\,. \tag{7}$$

IV. RESULTS AND DATA PROCESSING

Figure 1 shows some electrofission fragment angular distributions, in energies near the fission barrier; the curves represent the best fits of the function $W(\theta)=a+b\sin^2\theta+c\sin^2(2\theta)$ to the experimental data. The distributions show maxima shifted to 45°, indicating a significant contribution of the quadrupole component in this energy region. This is also evidenced by the relatively high values of the c/b ratio. The experimental results for the electrofission yield $Y_{e,e'f}(E_0)$ are presented in Fig. 2. The continuous curve represents $Y_{e,e'f}^{*}(E_0)$ defined by Eq. (5).

The virtual photon spectra were calculated by an analytical expression, which is a function of E_0, E, and Z, as explained in Ref. 10. Recently,

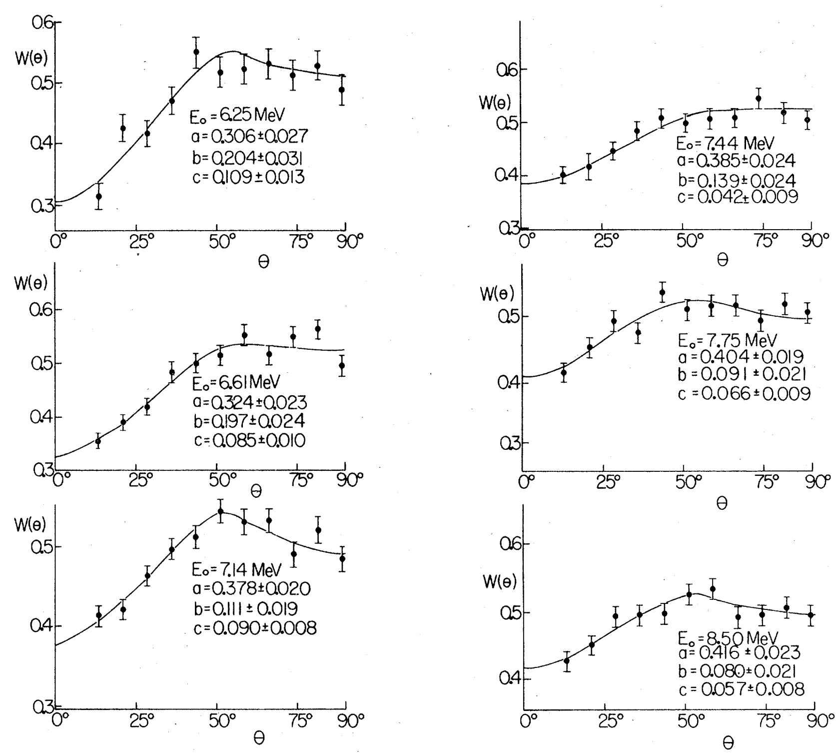

FIG. 1. ^{238}U electrofission fragment angular distributions at several energies, near the fission barrier. The curves are least square fits of the function $W(\theta)=a+b\sin^2\theta+c\sin^2(2\theta)$ to the experimental points.

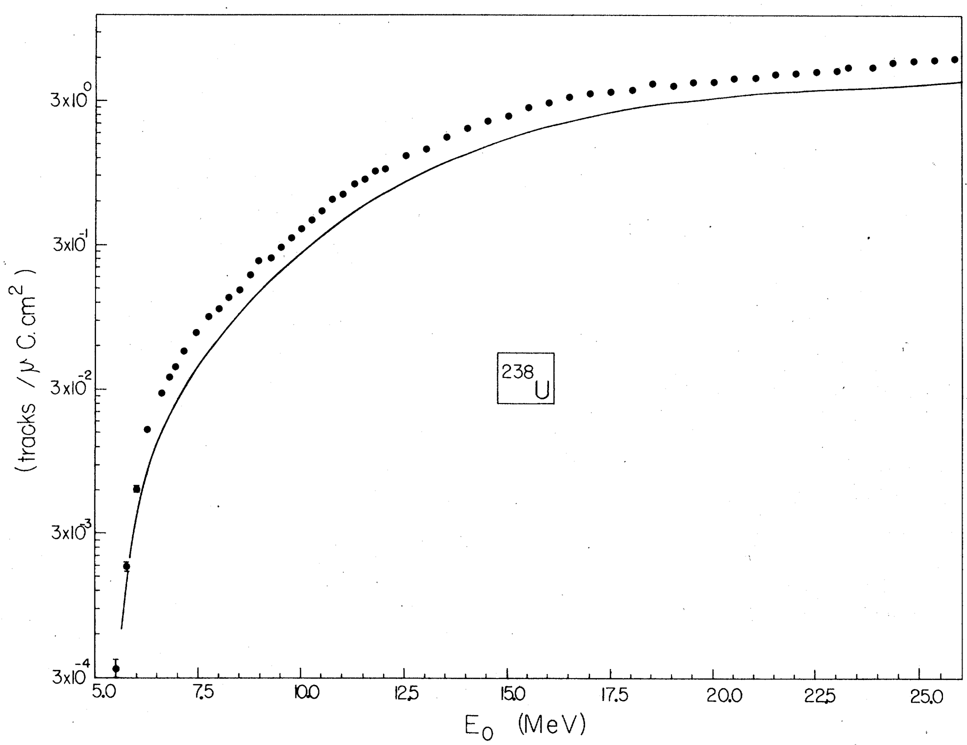

FIG. 2. Experimental electrofission yield for ^{238}U, $Y_{e,e'f}(E_0)$. The continuous curve represents $Y^*_{e,e'f}(E_0)$, defined by Eq. (5).

Soto Vargas, Onley, and Wright[13] presented a new computational technique for calculating the virtual photon spectra in DWBA, and observed a discrepancy between their new calculation and the ones obtained using the analytical expression; however, we have verified that this discrepancy does not affect our results since it is more important above 20 MeV, that is, above the energy region where the measured $E2$ is concentrated.

The values for $\sigma_{\gamma,f}(E)$ were obtained from Dickey and Axel[14] in the range 5.5 to 8.0 MeV, from Veyssière *et al.*[15] in the range 8.0 to 18.0 MeV, and from Arruda Neto *et al.*[6] in the range 18.0 to 30.0 MeV.

The normalization constant K was determined by means of Eq. (7). For this we measured the bremsstrahlung induced fission yield in the same energy range as the electrofission measurements and calculated numerically the integral of the denominator using the $\sigma_{\gamma,f}(E)$ cross section referred to above. The thin target bremsstrahlung spectra were corrected for the finite thickness of the aluminum radiator.[16]

Figure 3 presents a plot of $\Delta Y/K$ as a function of the incident electron energy E_0. The circles with error bars represent the difference

$$[Y_{e,e'f}(E_0) - Y^*_{e,e'f}(E_0)]/K$$

and the continuous curve is the result of the folding back of $\sigma^{add}_{\gamma,f}(E)$, shown in Fig. 4, with the kernel

$$[N_v^{E2}(E,E_0) - N_v^{E1}(E,E_0)]/E.$$

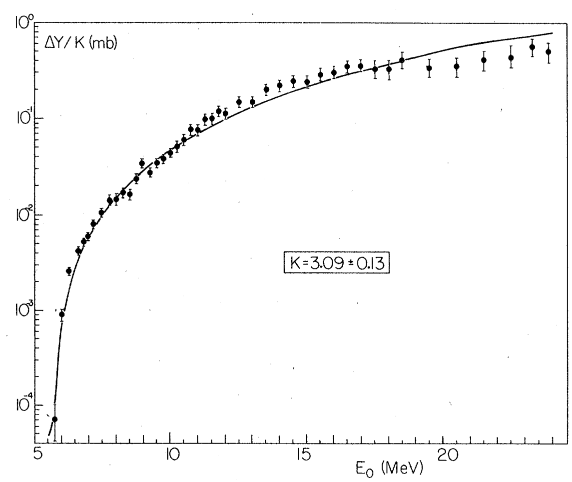

FIG. 3. $\Delta Y/K$ as a function of the incident electron energy E_0. The circles with error bars represent the difference $[Y_{e,e'f}(E_0) - Y^*_{e,e'f}(E_0)]/K$, and the continuous curve is the result of the folding back of $\sigma^{add}_{\gamma,f}(E)$ with the kernel $[N_v^{E2}(E,E_0) - N_v^{E1}(E,E_0)]/E$.

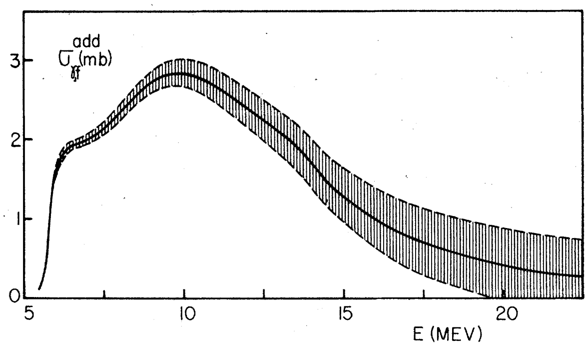

FIG. 4. The cross section $\sigma_{\gamma,f}^{add}$ (mb), defined by Eq. (6), as a function of the photon energy. The error bars define an uncertainty band for the values of $\sigma_{\gamma,f}^{add}$ (E).

The cross section $\sigma_{\gamma,f}^{add}(E)$, as a function of the photon energy E (MeV), is presented in Fig. 4. This cross section was obtained by solving the integral equation (4), using the least structure unfolding method developed by Cook.[17] The error bars define an uncertainty band for the values of $\sigma_{\gamma,f}^{add}(E)$.

V. DISCUSSION AND CONCLUSIONS

The cross section $\sigma_{\gamma,f}^{add}(E)$ shown in Fig. 4 is a mixture of $\sigma_{\gamma,f}^{E2}(E)$ and $\sigma_{\gamma,f}^{M1}(E)$, mainly between 6 and 7 MeV. In order to estimate the $M1$ contribution we can use the angular distribution of photofission fragments represented by

$$W(\theta)=a_\gamma+b_\gamma\sin^2\theta+c_\gamma\sin^2(2\theta).$$

Considering only contributions from the channels $(2^+,0)$, $(1^-,0)$, and $(1^+,1)$ of the transition nucleus, which is a reasonable assumption near the barrier, we can write[18]

$$\frac{4}{5}\frac{b_\gamma}{c_\gamma}=\frac{\sigma_{\gamma,f}^{E1}-\sigma_{\gamma,f}^{M1}}{\sigma_{\gamma,f}^{E2}}=r. \tag{8}$$

Defining $f(E)$ as

$$f(E)=\frac{F(E)\sigma_{\gamma,f}^{M1}(E)}{\sigma_{\gamma,f}^{add}(E)}, \tag{8a}$$

using Eq. (6) and the fact that

$$\sigma_{\gamma,f}(E)=\sigma_{\gamma,f}^{E1}(E)+\sigma_{\gamma,f}^{E2}(E)+\sigma_{\gamma,f}^{M1}(E), \tag{8b}$$

we can obtain from Eq. (8) the value of $f(E)$ as a function of r, $\sigma_{\gamma,f}(E)$, and $\sigma_{\gamma,f}^{add}(E)$:

$$f(E)\simeq 1-\frac{1}{r}\frac{\sigma_{\gamma,f}(E)}{\sigma_{\gamma,f}^{add}(E)}. \tag{9}$$

Taking the results of Dowdy and Krysinski[3(d)] and Rabotnov *et al.*[3(c)] in the energy range 6 to 7 MeV we have $r\simeq 30$ for the ratio $\frac{4}{5}b_\gamma/c_\gamma$ for ^{238}U. Using our results for $\sigma_{\gamma,f}^{add}(E)$ and the results of Dickey and Axel for $\sigma_{\gamma,f}(E)$, we obtain

$$\left\langle\frac{\sigma_{\gamma,f}(E)}{\sigma_{\gamma,f}^{add}(E)}\right\rangle=4.5\pm0.5 \text{ between 6 and 7 MeV}.$$

This gives for $f(E)$ the average value of 0.86 ± 0.13, and considering that $\langle F(E)\rangle=3.1\pm0.2$ in this energy region we have

$$\left\langle\sigma_{\gamma,f}^{M1}\right\rangle=(28\pm5)\% \text{ of } \left\langle\sigma_{\gamma,f}^{add}\right\rangle. \tag{10}$$

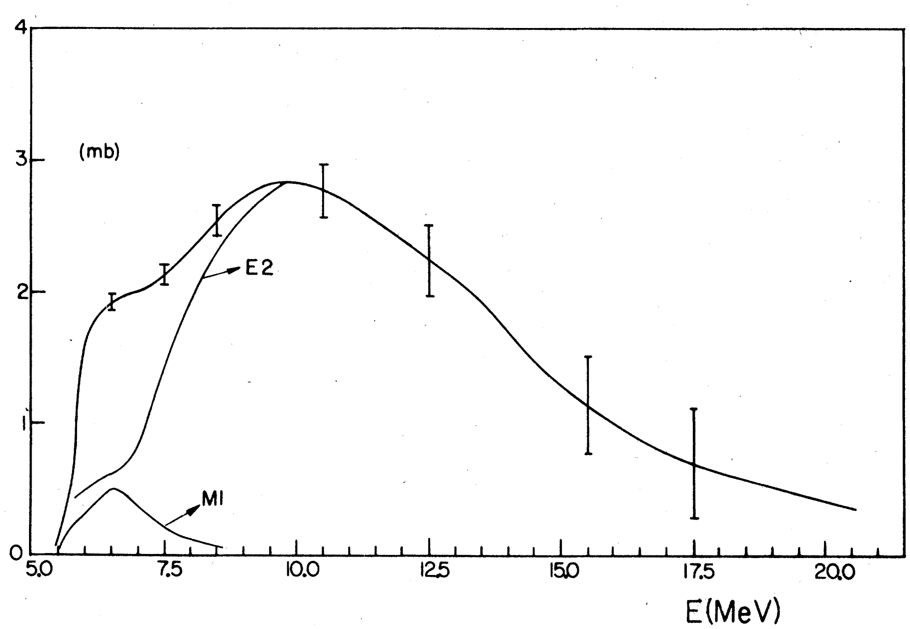

FIG. 5. The $E2$ and $M1$ components of $\sigma_{\gamma,f}^{add}$ (mb), as a function of the photon energy. The $M1$ cross section, $\sigma_{\gamma,f}^{M1}(E)$, is represented by a Breit-Wigner curve, with a cutoff at the low-energy tail due to the decrease in the fission probability.

These estimates show the presence of a non-negligible $M1$ component in the photofission of ^{238}U, in the energy range 6–7 MeV. In Fig. 5, the $M1$ cross section is represented by a Breit-Wigner curve, with peak at 6.5 MeV, width of 1.5 MeV, and a peak value of 0.5 mb (~30% of $\langle\sigma_{\gamma,f}^{add}\rangle$). On the other hand, the $M1$ component contributes to $\sigma_{\gamma,f}^{add}$ amplified by the factor $\langle F(E)\rangle$ [Eq. (8a)], owing to the fact that $N_v^{M1} > N_v^{E2}$ in the energy range considered, which makes its detection more sensitive. In this way, the $E2$ component will be given by

$$\sigma_{\gamma,f}^{E2}(E) \simeq \sigma_{\gamma,f}^{add}(E) - \langle F(E)\rangle\, \sigma_{\gamma,f}^{M1}(E)\,, \quad \text{for } E \sim 6\text{–}7 \text{ MeV}$$

and

$$\sigma_{\gamma,f}^{E2}(E) \simeq \sigma_{\gamma,f}^{add}(E)\,, \quad \text{for } E > 7 \text{ MeV}\,.$$

The $E2$ component is plotted in Fig. 5, evidencing a resonant character for the process with the following parameters: peak of (2.8 ± 0.2) mb at (9.9 ± 0.2) MeV and width of (6.8 ± 0.4) MeV. This cross section exhausts $(71 \pm 7)\%$ of the EWSR, calculated using the expression given by O'Connel.[19]

The following remarks can be made about these results:

(a) The peak at (9.9 ± 0.2) MeV corresponds to a value between the Bohr and Mottelson theoretical prediction of $58A^{-1/3}$ MeV (9.4 MeV for $A = 238$), and the experimental systematics for heavy nuclei, $65A^{-1/3}$ MeV (10.5 MeV for $A = 238$).

(b) The value $(71 \pm 7)\%$ of the EWSR indicates a preference for the fission channel in the decay of the isoscalar GQR.

(c) The width of (6.8 ± 0.4) MeV is a value greater than the ones obtained for heavy nuclei (3 to 4 MeV); however, GQR observed in permanently deformed nuclei have suggested the occurrence of a splitting similar to that observed in GDR,[20] with the difference that, for the $E2$ resonance, the splitting should be triple, corresponding to the modes $\mu = 0$, $\mu = \pm 1$, and $\mu = \pm 2$ (where μ are substates of $\lambda = 2$, the angular momentum of the phonon associated with vibrations of the nucleus). In Fig. 6, the observed GQR is, tentatively, split into three modes; nevertheless, this must be understood as a mere speculation, and the curves have been shown without the intention of rigorously reproducing well-known forms of resonant curves, such as Breit-Wigner or Lorentzian curves. The following values were obtained, for the energies $E(\lambda, \mu)$ of the component peaks of an isoscalar GQR, for ^{238}U: $E(2,0) = 8.6 \pm 0.5$ MeV, $E(2, \pm 1) = 10.5 \pm 0.5$ MeV, and $E(2, \pm 2) = 13.0 \pm 0.5$ MeV.

Recently, Suzuki and Rowe[21] obtained the following theoretical expression for the energies $E(\lambda, \mu)$, as a function of the deformation parameter δ:

$$E(2,0) = E_0(1 - \tfrac{1}{3}\delta)\,,$$
$$E(2, \pm 1) = E_0(1 - \tfrac{1}{6}\delta)\,, \qquad (11)$$
$$E(2, \pm 2) = E_0(1 + \tfrac{1}{3}\delta)\,.$$

Table I compares energy ratios predicted from

TABLE I. Ratios of $E(\lambda, \mu)$ for the ^{238}U isoscalar GQR.

	Suzuki and Rowe (Ref. 21)	Present work
$E(2,0)/E(2,\pm 2)$	0.8	0.7 ± 0.1
$E(2,0)/E(2,\pm 1)$	0.9	0.8 ± 0.1
$E(2,\pm 2)/E(2,\pm 1)$	1.2	1.2 ± 0.1

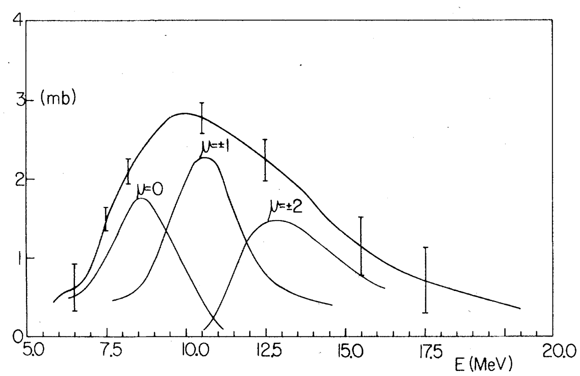

FIG. 6. The observed $E2$ component (GQR) in ^{238}U, tentatively split into three modes.

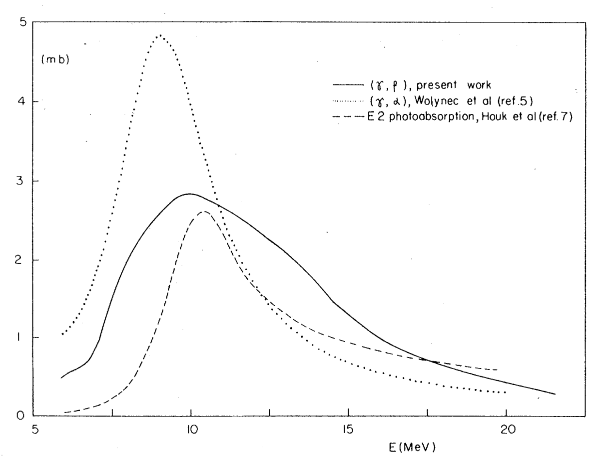

FIG. 7. Results for $E2$ isoscalar giant resonance in ^{238}U from different authors.

TABLE II. Isoscalar giant quadrupole resonance in ^{238}U.

Peak (MeV)	Width (MeV)	%EWSR	Reaction	Reference
10–13	...	85	(p,p')	Lewis and Horen (Ref. 4)
8.9 ± 0.3	3.7 ± 1.2	80(50)[a]	$(e,e'\alpha)$	Wolynec *et al.* (Ref. 5)
10.5 ± 0.2	$3.9^{+1.1}_{-0.5}$	40	(e,e')	Houk *et al.*[b]
9.9 ± 0.2	6.8 ± 0.4	71	$(e,e'f)$	Present work

[a]The value of 80% was calculated by using expression (84) of Ref. 19 and the rms value of 5.730 fm (Ref. 7) for the ^{238}U nuclear radius. The published value is 50%.
[b]These values are for the parameters of the cross section. The values in Ref. 7 are for the reduced transition probability.

Eqs. (11) using $\delta = 0.33$,[15] with ratios estimated from the present work. These show reasonable agreement.

Figure 7 and Table II show the results of the present work compared with some other results from the literature.

The most serious discrepancy occurs in the percentage of the EWSR exhausted, mainly between the result of this work and the results of Wolynec *et al.*,[5] and Houk *et al.*[7] The former work concerns the decay of the GQR through the α-emission channel, whose strength, when added to the one corresponding to the fission channel (present work), gives about 150% of the EWSR. As may be seen in Fig. 7, the (γ,α) peak is unreasonably high, especially when compared to the one obtained from (e,e') which represents the total $E2$ photoabsorption. The position and intensity of the peak obtained by Houk *et al.*[7] agree with the results of the present work, but the strength (40% of the EWSR) is lower. This is probably due to the subtraction of the radiation tail and continuous spectrum due to bremsstrahlung, or even to the ghost peak at 6.5 MeV as suggested by the authors.

Finally, we should state that the method described in the present work, which is based on a reasonably precise knowledge of the virtual photon spectra, allows the investigation of multipole contributions, other than $E1$, to the photoabsorption cross section of nuclei. The strength and some details related to the shape of the resonances are obtained in a model-independent way.

ACKNOWLEDGMENTS

The authors wish to thank Alvaro Vannucci, Rosana Hermann, and Lucia Setiuko Tengan for their assistance in preparing the mica detectors and in data handling.

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