NEUTRON GAMMA COMPETITION IN FAST FISSION

J. FREHAUT
Centre d'Etudes de Bruyères-le-Châtel
Service de Physique et Techniques Nucléaires
B.P. 12 - 91680 Bruyères-le-Châtel

INTRODUCTION

The early analyses of fission fragment desexcitation were based on the statistical evaporation model and assumed that neutron emission was taking place whenever energetically possible. The residual energy after neutron emission was considered to appear as prompt gamma-rays leading to an average gamma energy $\bar{E}_\gamma$ released per fission of the order of 4 to 5 MeV [1]

Since it was experimentally established that $\bar{E}_\gamma$ was rather of the order of 7 to 8 MeV, the competition between gamma-ray and neutron emission in the last stage of fragment desexcitation has been put forward to explain the difference.

More recent data reviewed by Nifenecker [2] showed that for individual fragments $\bar{E}_\gamma$ as well as $\bar{n}_\gamma$, the number of gamma quanta, present the same saw-tooth shape as $\bar{\nu}_p$ when plotted as a function of fragment mass. From these data a linear relation between $\bar{E}_\gamma$ and $\bar{\nu}_p$ was derived, in which the constant term was 4 MeV, i.e. the statistical prediction for $\bar{E}_\gamma$. The increase of $\bar{E}_\gamma$ and $\bar{n}$ with $\bar{\nu}_p$ was thus interpreted as an increase of the average fragment angular momentum with excitation energy [2].

In the present paper we analyse the data we have obtained on the distribution of the gamma-ray energy per fission, as well as on the average energy $\bar{E}_\gamma$, for the neutron induced fission of several isotopes, in the energy range up to 15 MeV. The data on $\bar{E}_\gamma$ have already been published [3].

EXCITATION ENERGY DEPENDENCE OF $\bar{E}_\gamma$

The measurement of $\bar{E}_\gamma$ was made at the same time as $\bar{\nu}_p$, using the large Gd-loaded liquid scintillator technique [4].

$\bar{E}_\gamma$ is proportional to the area of the prompt pulse detected in the scintillator in coincidence with a fission fragment. A small correction ($\lesssim$ 5 %) is to be applied to account for the contribution of the energy released by the slowing down of the emitted fission neutron. Each fission chamber used contained a ^{252}Cf deposit in order to calibrate the $\bar{\nu}_p$ and $\bar{E}_\gamma$ data. Furthermore the detector has a high detection efficiency and the statistics was large enough to obtain information on the total gamma-ray spectrum.

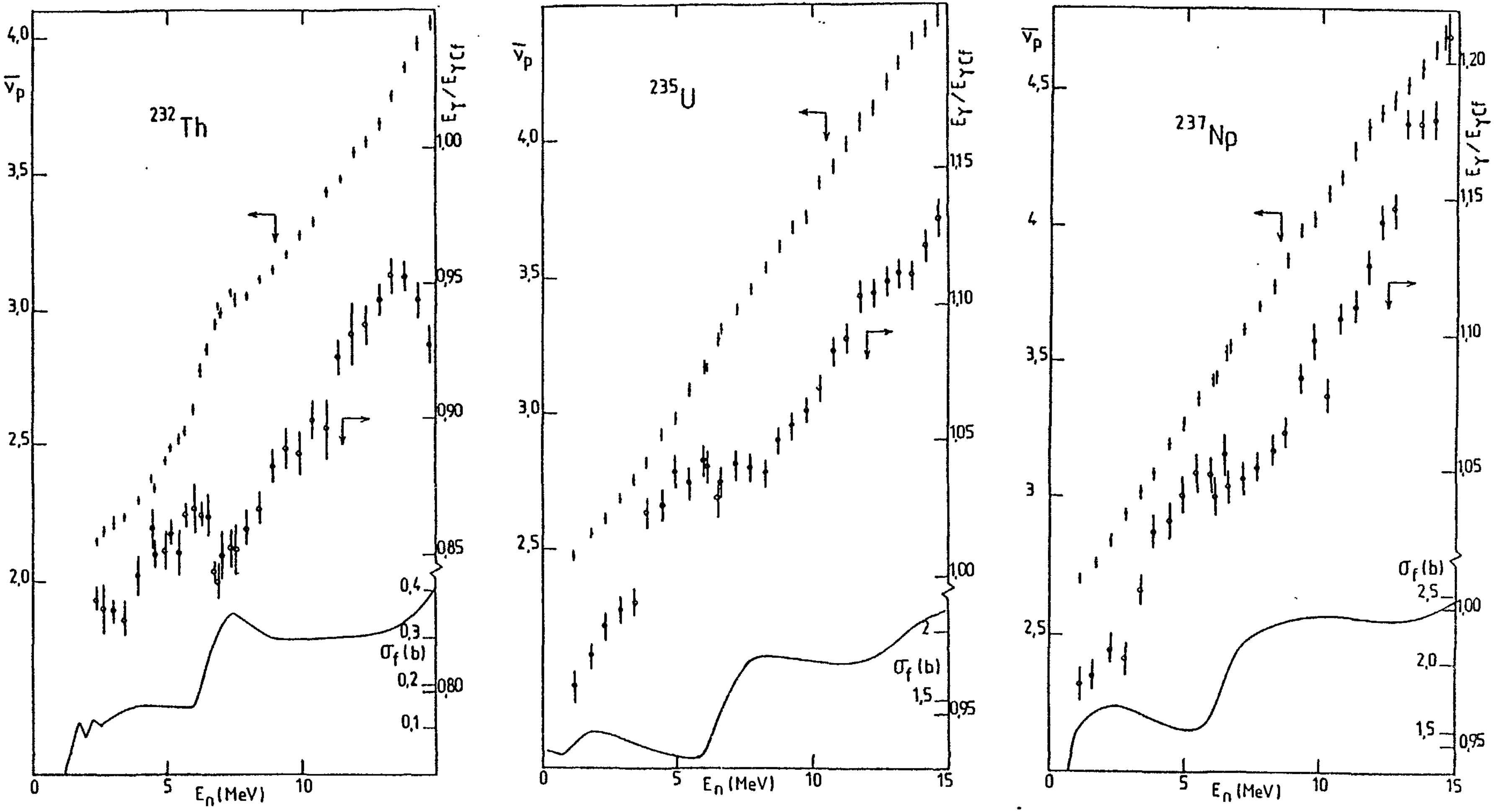

Fig. 1 : $\bar{\nu}_p$, $\bar{E}_\gamma$ and σ_f as a function of incident energy for the neutron induced fission of ^{232}Th, ^{235}U and ^{237}Np.
$\bar{E}_\gamma$ is given relative to the corresponding $\bar{E}_{\gamma Cf}$ value for the ^{252}Cf spontaneous fission.

The $\bar{E}_\gamma$ data obtained for ^{232}Th, ^{235}U and ^{237}Np [3] are plotted in fig.1 as a function of the incident neutron energy E_n. Below the second chance fission threshold $\bar{E}_\gamma$ is proportionnal to E_n. In the (n,n'f) reaction, a neutron is emitted prior to fission, without competition with gamma emission. The resulting nucleus will undergo fission with less excitation energy and therefore $\bar{E}_\gamma$ will be lower. The plateaus appearing in fig.1 above the second chance fission threshold can thus be qualitatively understood. A similar behaviour is observed just above the third chance fission threshold.

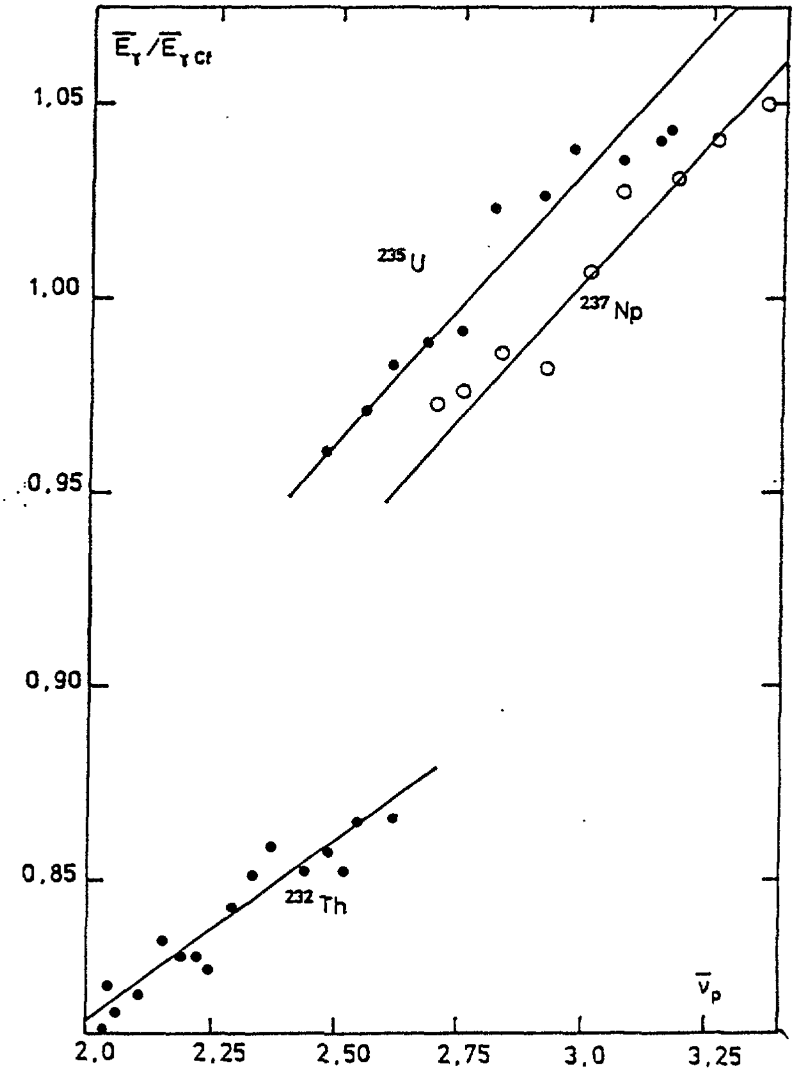

Fig. 2 : $\bar{E}_\gamma$ as a function of $\bar{\nu}_p$ for ^{232}Th, ^{235}U, ^{237}Np.
$\bar{E}_\gamma$ is given relative to $\bar{E}_{\gamma Cf}$, the gamma energy released in the ^{252}Cf spontaneous fission.

Since $\bar{\nu}_p$ is also a linear function of incident neutron energy below the second chance fission threshold, we can derive in this energy range a linear relation between $\bar{E}_\gamma$ and $\bar{\nu}_p$. The corresponding data are plotted in fig.2.

The fits to the data give :

$$^{235}\text{U} : \frac{\bar{E}_\gamma}{\bar{E}_{\gamma Cf}} = (0.139 \pm 0.010)\ \bar{\nu}_p + (0.619 \pm 0.030)$$

$$^{237}\text{Np} : \frac{\bar{E}_\gamma}{\bar{E}_{\gamma Cf}} = (0.1275 \pm 0.011)\ \bar{\nu}_p + (0.623 \pm 0.028)$$

$$^{232}\text{Th} : \frac{\bar{E}_\gamma}{\bar{E}_{\gamma Cf}} = (0.090 \pm 0.010)\ \bar{\nu}_p + (0.635 \pm 0.030)$$

The constant term is practically the same for the three nuclei and equal to 4.4 MeV if we assume a value of 7 MeV for $\bar{E}_{\gamma Cf}$. It is the value predicted by the statistical theory, assuming no neutron-gamma competition [1] . Here again, the linear dependence of $\bar{E}_\gamma$ with $\bar{\nu}_p$ can be interpreted in terms of a linear increase of the average spin of the fragments with energy. However, as we will see later, more information can be derived from the experimental data, leading to a different interpretation.

We have also obtained $\bar{E}_\gamma$ data for ^{241}Pu, but with a poor accuracy (fig.3). Assuming the constant term to be 0.63 we have derived :

$$\frac{\bar{E}_\gamma}{\bar{E}_{\gamma Cf}} = (0.112 \pm 0.012)\ \bar{\nu}_p + 0.63$$

From the experimental data on $\bar{\nu}_p$ and $\bar{E}_\gamma$ we can derive the relative contribution of first and second chance fission. For the first chance fission the measured quantities $\bar{E}_{\gamma 1}$ and $\bar{\nu}_1$ are related by the relation $\bar{E}_{\gamma 1} = a\ \bar{\nu}_1 + b$ as discussed previously. In the second chance fission the gamma ray energy is $\bar{E}_{\gamma 2}$ and the number of emitted neutrons is $\bar{\nu}_2 + 1$, $\bar{\nu}_2$ being the real number of fission neutrons. If α is the proportion of second chance fission, then the quantities measured are :

$$\bar{\nu}_m = (1 - \alpha)\ \bar{\nu}_1 + \alpha\ (1 + \bar{\nu}_2)$$

$$\bar{E}_{\gamma m} = (1 - \alpha)\ \bar{E}_{\gamma 1} + \alpha\ \bar{E}_{\gamma 2}$$

If we assume that the same relation holds for $\bar{E}_{\gamma 2}$ and $\bar{E}_{\gamma 1}$, i.e. that $\bar{E}_{\gamma 2} = a\ \bar{\nu}_2 + b$, then we obtain straightforwardly :

$$\alpha = \bar{\nu}_m - \frac{\bar{E}_{\gamma m} - b}{a}$$

Fig. 4 shows that realistic first and second chance fission cross sections can be derived below the third chance fission threshold for ^{235}U and ^{237}Np.

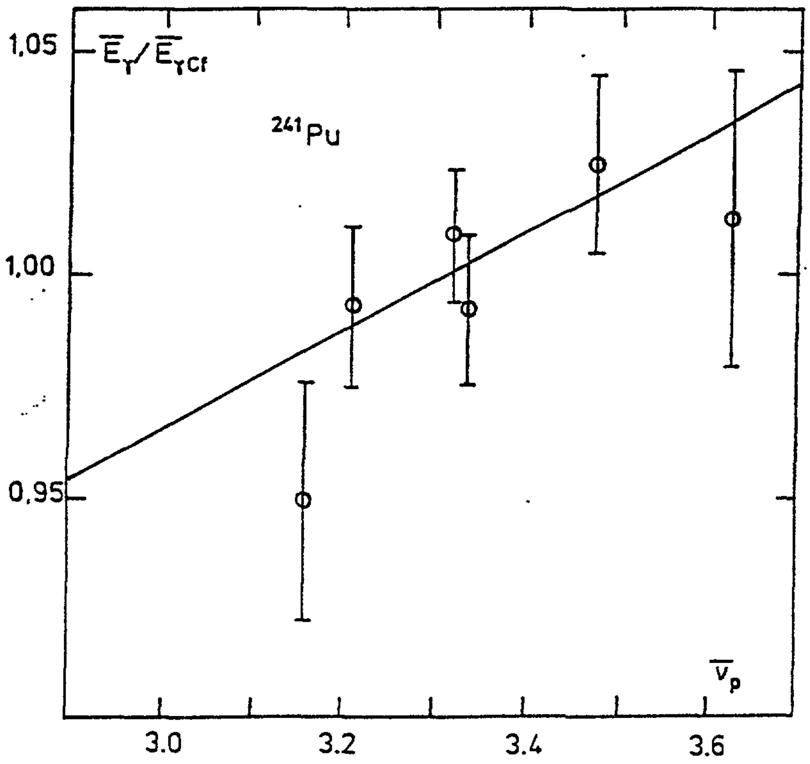

Fig. 3 : $\bar{E}_\gamma$, normalized to ^{252}Cf, as a function of $\bar{\nu}_p$ for ^{241}Pu.

In the case of ^{232}Th the situation is more complicated. The (n,2n) reaction is strongly competing with the (n,n'f) reaction. The emission of a second neutron is about 10 times more probable than fission. It seems that we observe an excess of gamma energy, which could result from the reaction (n,n'γf), with a cross section of the order of 100 mb.

FISSION GAMMA-RAY ENERGY SPECTRUM

The experimental spectra measured in the large liquid scintillator detector are quite broad (fig.5), but they were confirmed by a measurement of the total fission gamma-ray energy spectrum of ^{252}Cf made by J. Lachkar in our laboratory (1971, unpublished) using a large volume INa pit detector (fig.6).

Since the single quantum spectrum is also known, information about the distribution of the number of quanta can be obtained by comparing the two spectra.

The single quantum spectrum can be approximated by the relation :

$$P_1(E) = \frac{e^{-\frac{E}{\beta}}}{\beta}, \text{ where } \beta \text{ is the quantum average energy.}$$

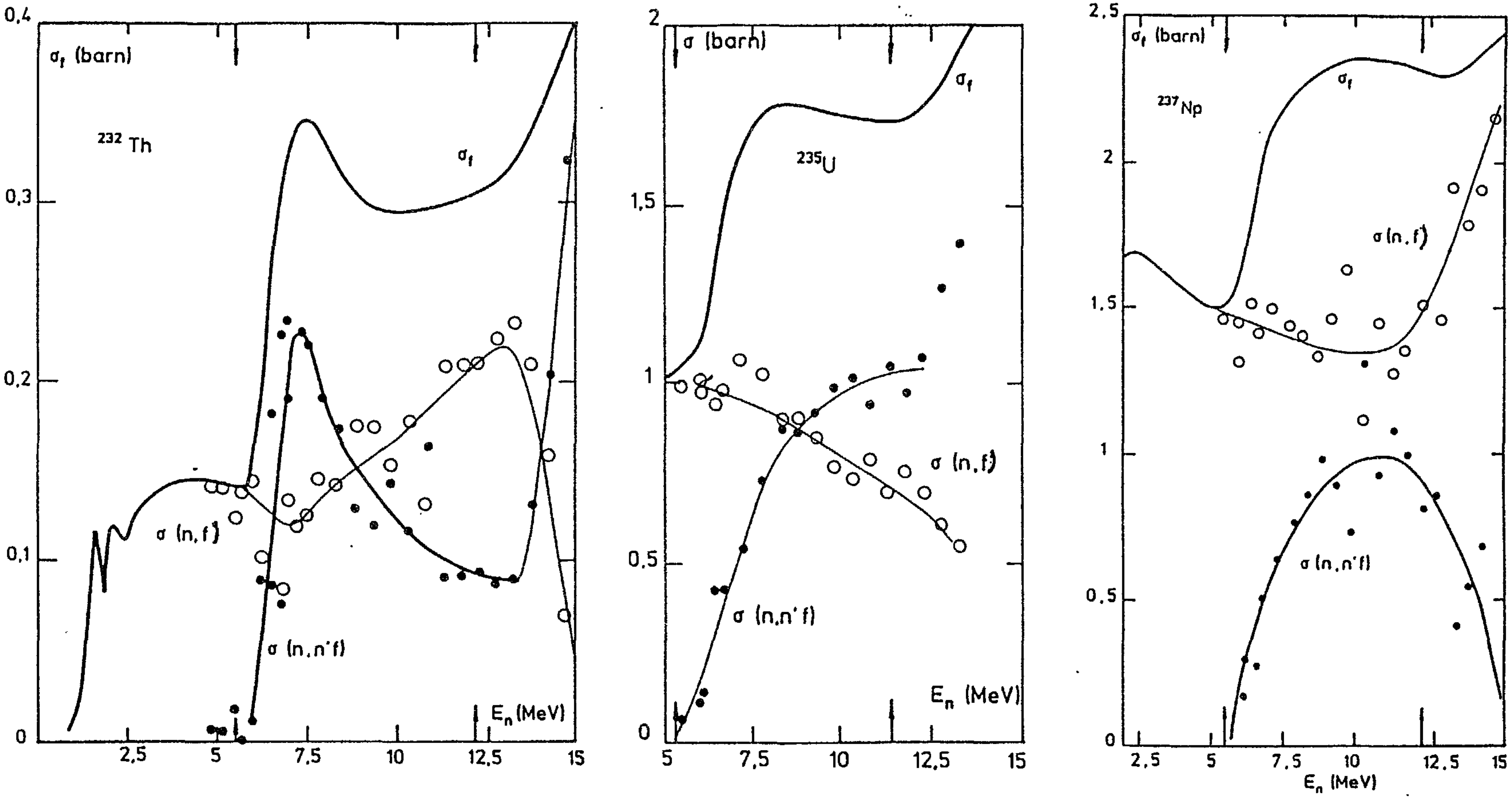

Fig. 4 : Relative contribution to σ_f of first and second chance fission deduced from $\bar{E}_\gamma$ and $\bar{\nu}_p$ measurements for ^{232}Th, ^{235}U and ^{237} Np. The arrows on energy scales indicate the second and third chance fission thresholds.

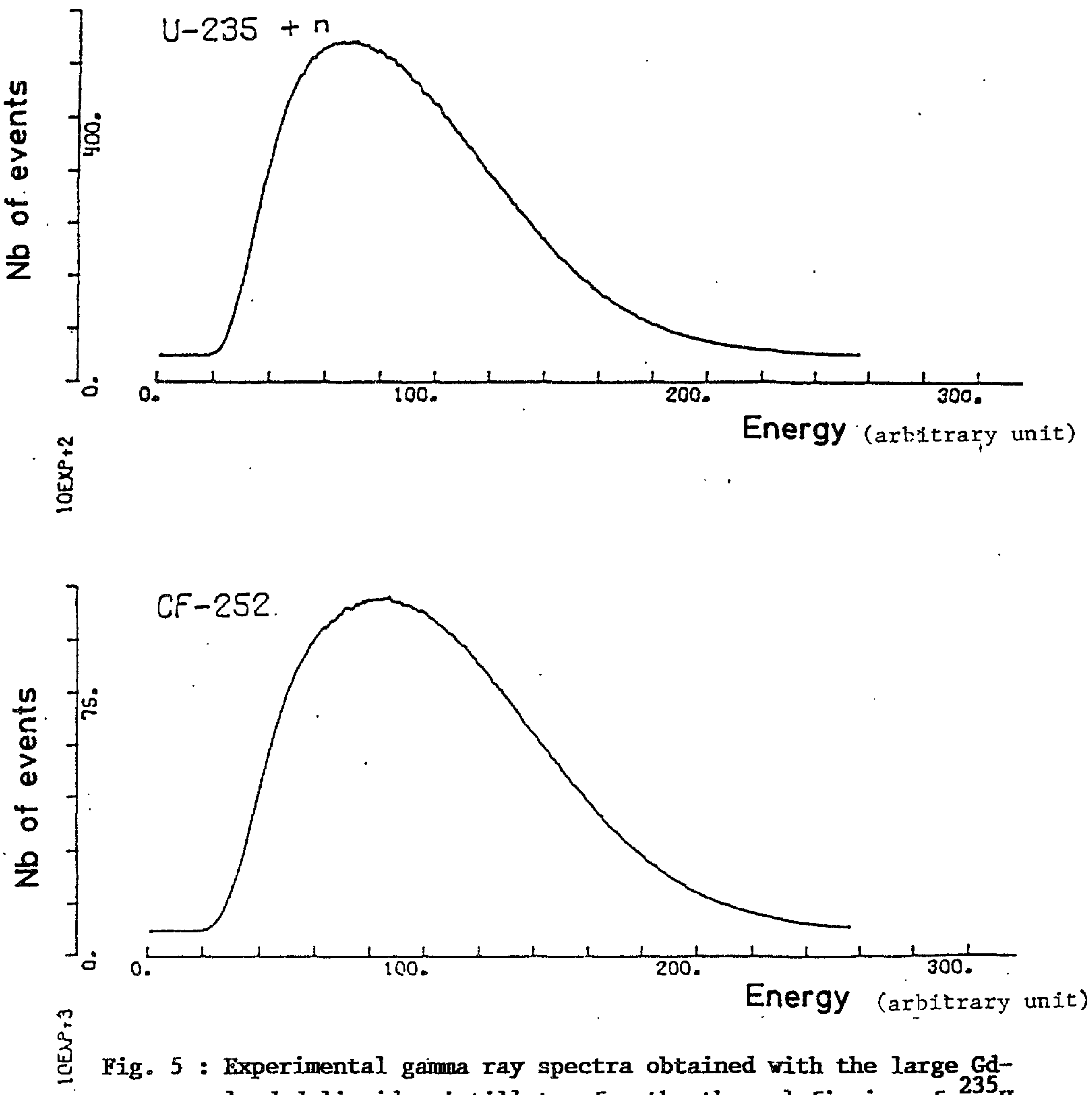

Fig. 5 : Experimental gamma ray spectra obtained with the large Gd-loaded liquid scintillator for the thermal fission of ^{235}U and the spontaneous fission of ^{252}Cf. The scales are relative and different for the two spectra.

When n quanta are emitted, the resulting spectrum is :

$$P_n(E) = \frac{E^{n-1}}{(n-1)!\ \beta^{n-1}} P_1(E)$$

with an average energy of $n\beta$.

For a distribution D(n) of the number of quanta the distribution is :

$$\mathcal{P}(E) = \sum_n D(n)\ P_n(E)$$

with an average number of quanta : $\bar{n} = \sum_{n} n\,D(n)$ and an average γ-ray energy per fission : $\bar{E}_\gamma = \bar{n}\beta$.

The experimental curve has been fitted assuming a gaussian shape for D(n) :

$$D(n) = \frac{1}{\sigma\sqrt{2\pi}}\, e^{-\frac{1}{2}\left(\frac{n-\bar{n}}{\sigma}\right)^2}$$

with $\bar{n}$, β and σ as free parameters.

The best fit (fig.6) is obtained for $\bar{n} = 7.4$, $\beta = 0.96$ and $\sigma = 2.6$, leading to $\bar{E}_\gamma = 7.1$ MeV, which compares very well with other experimental data [5].

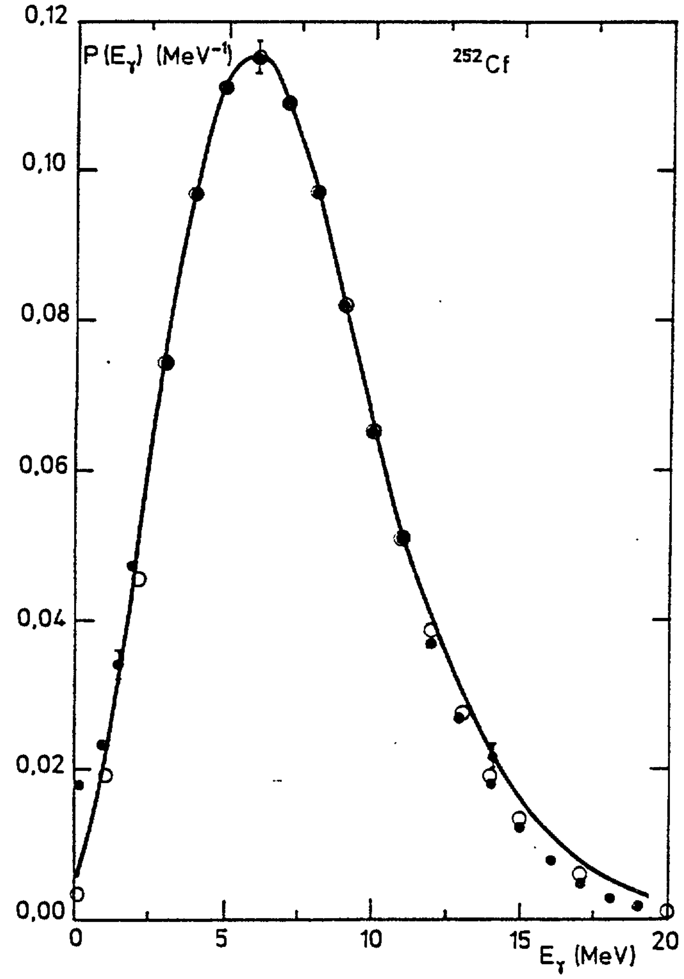

Fig. 6 : Experimental gamma ray spectrum for the spontaneous fission of ^{252}Cf using a NaI detector. Fits to the data (see text) :

● Gauss law for the distribution of the number of quanta.
O Poisson law for the distribution of the number of quanta.

Since the variance of D(n) is near the average value $\bar{n}$, we have also made a fit using a Poisson distribution :

$$D(n) = \frac{\bar{n}^{n}}{n!} e^{-\bar{n}}$$

with $\bar{n}$ and β as free parameters.

We obtain also a good fit, particularly at low energy (fig.6) with $\bar{n} = 7.7$ and $\beta = 0.92$ leading to $\bar{E}_\gamma = 7.08$ MeV.

With a Poisson distribution we can furthermore relate simply the average energy $\bar{E}_\gamma$ and the variance σ_γ^2 of the $\mathcal{P}(E)$ distribution to $\bar{n}$ and β :

$$\bar{E}_\gamma = \bar{n}\,\beta$$

$$\sigma_\gamma^2 = 2\,\beta\,\bar{E}_\gamma$$

so that from the experimental data $\bar{E}_\gamma$ and σ_γ^2 available from the liquid scintillator detector we can derive :

$$\beta = \frac{\sigma_\gamma^2}{2\,\bar{E}_\gamma} \quad \text{and} \quad \bar{n} = \frac{2\,\bar{E}_\gamma^2}{\sigma_\gamma^2}$$

as a function of excitation energy.

EXCITATION ENERGY DEPENDENCE OF β AND $\bar{n}$

The experimental data for σ_γ^2 below the second chance fission threshold are plotted in fig.7 as a function of $\bar{\nu}_p$ for ^{237}Np, ^{235}U and ^{232}Th. The data for ^{232}Th have been averaged to reduce the dispersion.

Within the uncertainties σ_γ^2 is a linear function of $\bar{\nu}_p$:

$$^{235}\text{U} : \frac{\sigma_\gamma^2}{\sigma_{\gamma\text{Cf}}^2} = 0.278\ \bar{\nu}_p + 0.394$$

$$^{237}\text{Np} : \frac{\sigma_\gamma^2}{\sigma_\gamma^2\text{Cf}} = 0.259\ \bar{\nu}_p + 0.291$$

$$^{232}\text{Th} : \frac{\sigma_\gamma^2}{\sigma_{\gamma\text{Cf}}^2} = 0.188\ \bar{\nu}_p + 0.470$$

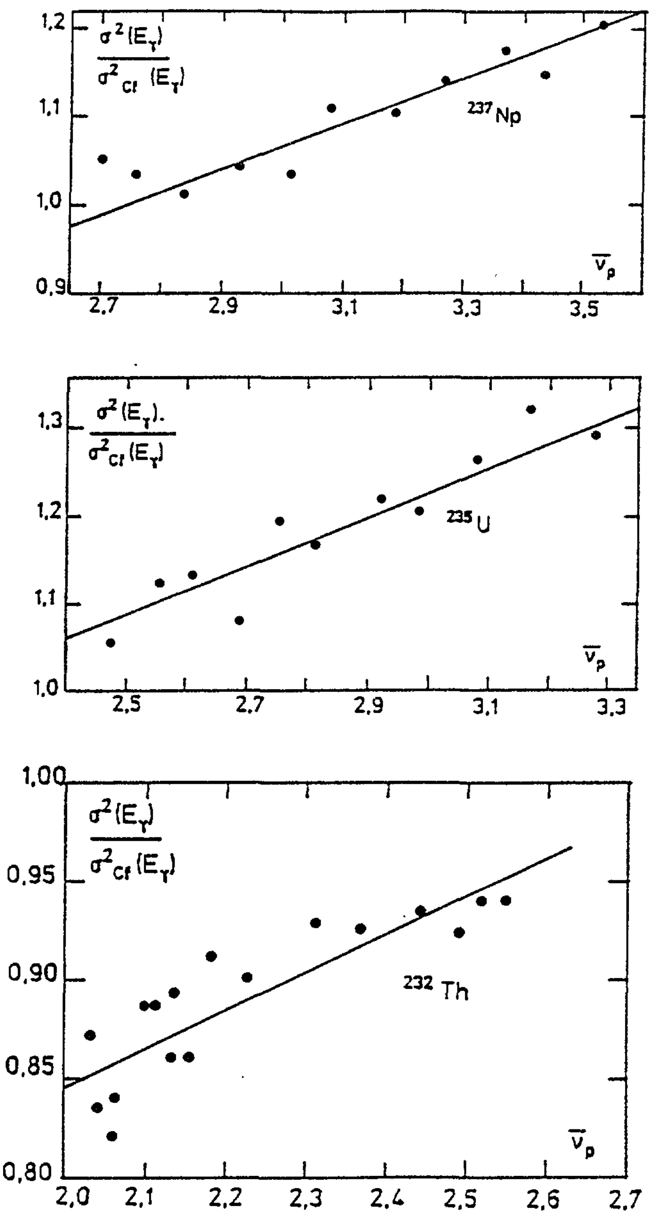

Fig. 7 : Variance of the fission gamma ray spectra as a function of $\bar{\nu}_p$ for ^{232}Th, ^{235}U and ^{237}Np (normalized to ^{252}Cf spontaneous fission).

From these fitted data and the corresponding fits to $\bar{E}_\gamma$ discussed previously we have derived β and $\bar{n}$ as a function of $\bar{\nu}_p$ using the values $\bar{E}_{\gamma Cf}$ = 7.08 MeV and $\sigma^2_{\gamma Cf}$ = 13 MeV^2 derived from our fit to the ^{252}Cf data of fig.6. The results are plotted in fig.8 et 9 for β and n, respectively.

We observe that the average number of quanta $\bar{n}$ remains practically constant, whereas β is increasing almost linearly with $\bar{\nu}_p$.

The relative values of β can be undestood, at least qualitatively, as level density effects if we remember that ^{236}U* is a even-even fissioning nucleus, ^{233}Th* is even-odd, and ^{238}Np* is odd-odd.

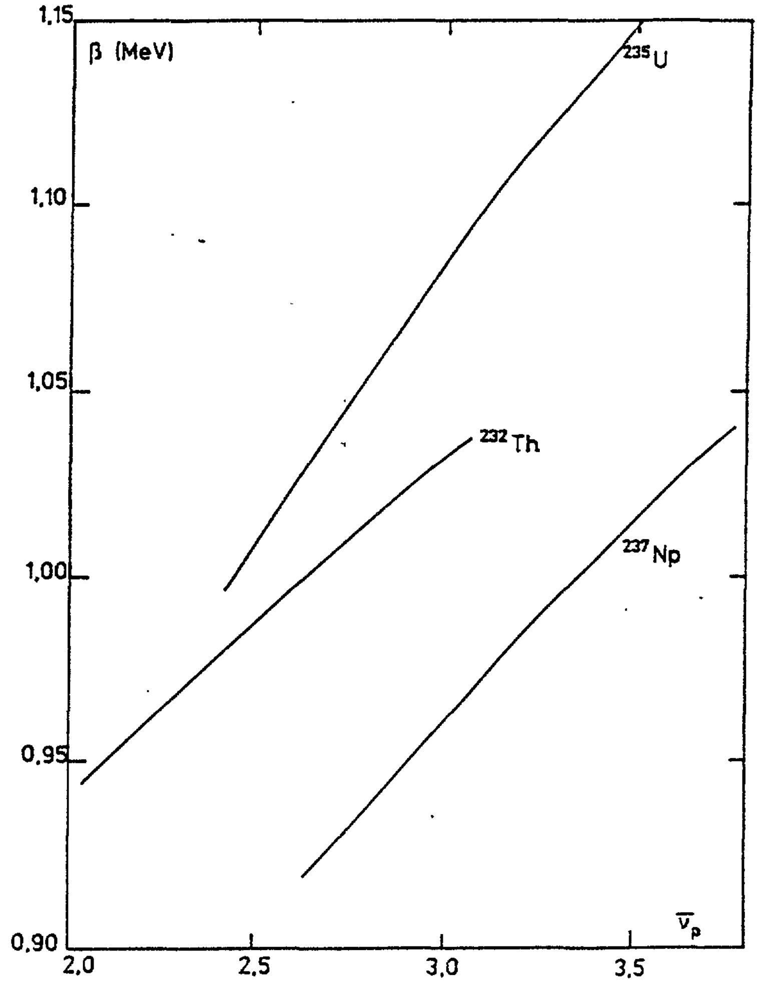

Fig. 8 : Average single fission gamma quantum energy as a function of $\bar{\nu}_p$ for ^{232}Th, ^{235}U and ^{237}Np.

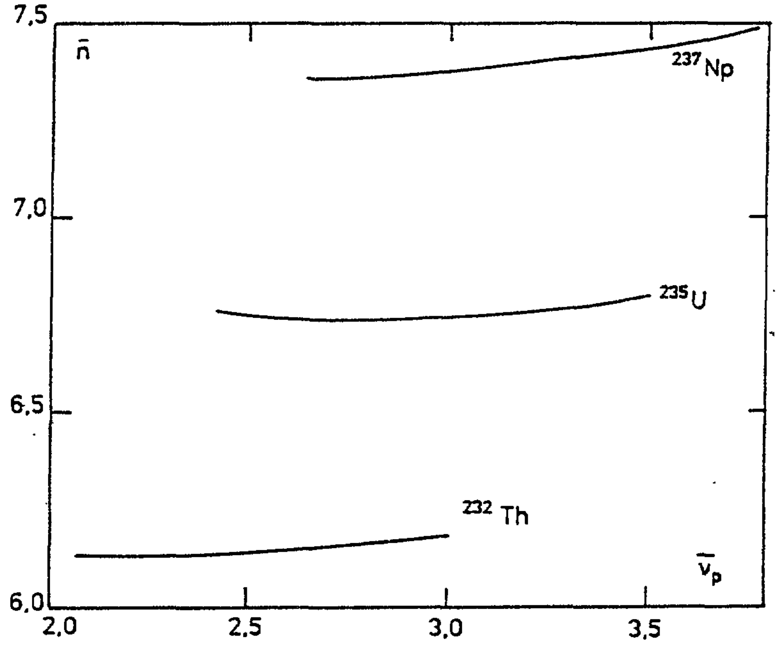

Fig. 9 : Average number of fission gamma quanta as a function of $\bar{\nu}_p$ for ^{232}Th, ^{235}U and ^{237}Np.

The increase of β with excitation energy can be understood in the frame of a larger available phase space allowing a larger number of relatively high energy gamma-rays to be emitted. In that case the quite constant value of $\bar{n}$ would mean that the average fragment angular momentum is not increasing with excitation energy. This interpretation is not in contradiction with the conclusion of an increase of fragment angular momentum with excitation energy derived from the study of the desexcitation of individual fragments. In that latter case, indeed, the average number of gamma quanta is also increasing with excitation energy.

Using the data of ref. [5] for ^{252}Cf we derive from our data for the thermal fission of ^{235}U :

$$\beta = 0.954 \text{ MeV}, \ \bar{n} = 6.92$$

to be compared to the evaluated data [5] :

$$\beta = 0.97 \text{ MeV and } \bar{n} = 6.89$$

EVALUATION OF β AND $\bar{n}$ FOR OTHER NUCLEI

- **^{233}U and ^{239}Pu**

For these nuclei $\bar{\nu}_p$, $\bar{E}_\gamma$ and $\bar{n}$ are experimentally known for the thermal fission :

	^{233}U	^{239}Pu	Ref
$\bar{\nu}_p$	2.495	2.882	[6]
$\bar{E}_\gamma$, MeV	6.69	6.77	[5]
$\bar{n}$	6.31	7.05	[5]

$$\text{Assuming :} \quad \frac{\bar{E}_\gamma}{\bar{E}_{\gamma Cf}} = a \ \bar{\nu}_p + 4.4 \text{ (MeV)}$$

$$\text{and } \bar{n} = \text{Cst, we can derive } \beta = \frac{\bar{E}_\gamma}{\bar{n}}$$

The results are :

$$^{233}\text{U} : \bar{E}_\gamma = 0.92 \ \bar{\nu}_p + 4.4 \quad \text{(MeV)}$$

$$\beta = 0.145 \ \bar{\nu}_p + 0.70 \text{ (MeV)}$$

$$^{239}\mathrm{Pu}: \quad \bar{E}_\gamma = 0.82\ \bar{\nu}_p + 4.4 \quad (\mathrm{MeV})$$

$$\beta = 0.117\ \bar{\nu}_p + 0.62\ (\mathrm{MeV})$$

- **Other nuclei**

In the absence of experimental data, we can use the same formalism, taking the low energy fission values of $\bar{E}_\gamma$ and $\bar{n}$ from the systematics of Hoffman and Hoffman [5] :

$$\bar{E}_\gamma = 0.028\ A + 0.09$$

$$\bar{n} = 0.112\ A - 19.94$$

CONCLUSION

We have taken advantage of the high efficiency to γ-rays of the large Gd-loaded liquid scintillator used in $\bar{\nu}_p$ measurements to derive information about the total prompt γ - ray spectrum in fast neutron induced fission. A careful analysis of the data led to the conclusion that the observed increase of the total average gamma energy $\bar{E}_\gamma$ with excitation energy is due to an increase of the average energy β of the single quanta, the number of quanta $\bar{n}$ remaining about constant. These findings do not support the idea of an increase of the average fragment angular momentum with excitation energy. These results do not contradict the idea of an increase of fragment angular momentum with excitation energy deduced from the study of the desexcitation of individual fragments (mass dependence of $\bar{\nu}_p$, $\bar{E}_\gamma$, $\bar{n}$), since in that case the number of gamma quanta is increasing with the mass correlated fragment excitation energy.

The decrease of $\bar{E}_\gamma$ observed for the second chance fission just results from a lower value of β, since the fissioning nucleus has in that case about 7 MeV less excitation energy than in first chance fission.

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