# Experimental investigations of the $(n, \gamma f)$ reaction

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Experimental methods and results of investigations of the $(n, \gamma f)$ reaction are reviewed. Theoretical estimates of the probability of this reaction for some heavy nuclei are also given.

## INTRODUCTION

In 1965 Stavinskiĭ and Shaker,[1] and also Lynn[2] showed that for compound states formed by the interaction of slow neutrons with a heavy nucleus de-excitation could occur not only through scattering, radiative capture, and fission but also, as a theoretical possibility, through a two-step $(n, \gamma f)$ reaction, in which fission occurs after preliminary emission of one or several $\gamma$ rays. The scheme of such a reaction is shown in Fig. 1. An interesting feature of the $(n, \gamma f)$ reaction is that the intermediate compound state formed after the $\gamma$ transition may be above the fission threshold corresponding to it even in the case when the original compound state was below the fission threshold associated with it. In such a situation, fission could be observed in the resonance region of the energies for resonances for which the direct-fission width is effectively equal to zero.

Investigations of the $(n, \gamma f)$ reaction are of interest primarily for fission physics, since they can give information about the height and structure of the fission barriers and about the spectrum of above-barrier transition states and the degree of their damping, which characterizes the coupling of the collective mode of the nucleon motion in the nucleus and the internal degrees of freedom (excitation of quasiparticles and holes). Understanding of the $(n, \gamma f)$ reaction is also needed to solve the interesting problem in fission physics of the influence of the quantum numbers of the excited states of the fissioning nucleus on the properties of the fission products.

In addition investigations of the $(n, \gamma f)$ reaction represent one of the few ways of studying $\gamma$ transitions between highly excited states of heavy nuclei. The structure of such states, which include neutron resonances, is very complicated. As experimental data have accumulated, the quasiparticle–phonon nuclear model[3] has been successfully developed for their interpretation, alongside the statistical model traditionally used for that purpose. In the new model, the state wave functions are represented in the form of expansions with respect to the number of quasiparticles. At the present time there are practically no experimental data on the many-particle components of the wave functions of highly excited states. From this point of view investigations of prefission $\gamma$ transitions between highly excited states for which precisely the many-quasiparticle components make the main contribution to their wave functions could be an important source of information.

From the practical point of view, allowance for the $(n, \gamma f)$ reaction is needed to estimate nuclear-physics constants such as the cross sections for fission and radiative capture of a neutron, which are used in the design of nuclear reactors.

## 1. THEORETICAL ESTIMATES OF THE PROBABILITY OF THE $(n, \gamma f)$ REACTION

The first publication on the $(n, \gamma f)$ reaction was the theoretical paper of Stavinskiĭ and Shaker, who estimated the probability of this reaction for some fissioning nuclei under the following assumptions:

1) in the radiative transitions dipole emission is dominant, and the contributions of the $E1$ and $M1$ transitions are equal;

2) the matrix elements that determine the probabilities of the $\gamma$ transitions are constant;,

3) fission is possible only after emission of the first $\gamma$ ray and is improbable after subsequent $\gamma$ transitions;

4) the energy dependence of the level density is determined by a simple law with constant temperature:

$$\rho(E) \sim \exp(-E/T). \tag{1}$$

In addition, in accordance with the simplified scheme adopted in the paper for the excited states of even–even compound nuclei at the saddle point of a single-hump fission barrier, it was assumed that the lowest positive-parity levels were situated 1.6 MeV below the excitation energy, and that the negative-parity levels were 1 MeV lower.

The following estimates of the widths $\overline{\Gamma}_{\gamma f}$ were obtained:

$$\overline{\Gamma}_{\gamma f} \simeq \begin{cases} 0.5\overline{\Gamma}_\gamma, & ^{233}\text{U and } ^{239}\text{Pu}; \\ 0.3\overline{\Gamma}_\gamma, & ^{235}\text{U}. \end{cases}$$

Thus, the probability of the $(n, \gamma f)$ reaction was found to be comparable with the probability of radiative capture. This circumstance prompted Stavinskiĭ and Shaker[1] to the idea that fission of the compound nuclei $^{234}$U, $^{236}$U, and $^{240}$Pu proceeds through $3^+$, $4^-$, and $1^+$ states, respectively, only after prior emission of a $\gamma$ ray, whereas for resonances with other possible values of the spin ($2^+$, $3^-$, and $0^+$, respectively) direct fission is dominant. It follows from this that states with different $J^\pi$ must correspond to the two types of neutron resonances (with $\overline{\Gamma}_\gamma/\overline{\Gamma}_f \simeq 1$ and $\overline{\Gamma}_\gamma/\overline{\Gamma}_f \ll 1$) observed in the experiments. On this basis Stavinskiĭ and Shaker proposed a method for experimental verification of the existence of the $(n, \gamma f)$ reaction—measurement and comparison of the angular distributions of the fission fragments for resonances with different $J^\pi$. Unfortunately, such experiments have not so far been performed.

Lynn[2] also calculated the widths $\overline{\Gamma}_{\gamma f}$ for some fissioning nuclei ($^{233,235}$U and $^{239,241}$Pu). His values are more realistic estimates of the probability of the $(n, \gamma f)$ reaction, since:

1) the results of analysis of the $\gamma$-ray spectra of radiative capture of neutrons by heavy nuclei were used to calculate the matrix elements of the $\gamma$ transitions in accordance with a

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model of the giant dipole resonance (in Lorentz form):

$$M(E_\gamma) \sim \frac{\Gamma_G E_\gamma}{(E_\gamma^2 - E_G^2)^2 + (\Gamma_G E_\gamma)^2}; \qquad (2)$$

2) for the level density, Newton's model was used in place of the law with constant temperature.

Lynn obtained the following estimates for the widths $\overline{\Gamma}_{\gamma f}$:

$$^{235}\text{U}: \quad \overline{\Gamma}_{\gamma f} \simeq \begin{cases} 3\ \text{meV},\ 3^-\ \text{resonances} \\ 1.5\ \text{meV},\ 4^-\ \text{resonances} \end{cases}$$

$$^{239}\text{Pu}: \quad \overline{\Gamma}_{\gamma f} \simeq \begin{cases} 3\ \text{meV}(1^+) \\ 4 \div 7\ \text{meV}\ (0^+) \end{cases}$$

$$^{233}\text{U},\ ^{241}\text{Pu}: \quad \overline{\Gamma}_{\gamma f} \simeq 3\ \text{meV}$$

Only $E1$ transitions taken into account; the contribution of $M1$ transitions was assumed to be negligibly small.

It is worth noting here that the experimental data available when Refs. 1 and 2 were published supported Lynn's estimates, whereas those of Stavinskiĭ and Shaker appeared to be overestimated. Indeed, for a large number of $4^-$ resonances of $^{235}$U the minimal observed fission width was 2–3 meV, and for a $1^+$ resonance of $^{239}$Pu it was 4 meV. The experimentally observed fission width of resonances is a sum of two terms, $\Gamma_f = \Gamma_{\gamma f} + \Gamma_{fd}$, where $\Gamma_{fd}$ is a strongly fluctuating width corresponding to direct fission (i.e., without preliminary emission of a $\gamma$ ray). The width $\Gamma_{\gamma f}$ has a value that is constant (or fluctuates weakly) from resonance to resonance, since it is the sum of a large number of independent contributions (like $\Gamma_\gamma$, the radiative width). Thus, the experimentally observed minimal fission width satisfies $\Gamma_f(\min) \gtrsim \Gamma_{\gamma f}$, i.e., it gives an upper limit on the width of the $(n, \gamma f)$ reaction.

This circumstance gives a simple method of experimental estimation of the width $\overline{\Gamma}_{\gamma f}$, which, however, can be used effectively only in a restricted number of cases, since, first, it is precisely the resonances with the minimal fission widths that are measured with the greatest errors and may even be among the levels that are missed; second, the mean fission widths may be so great that the $(n, \gamma f)$ reaction will not make a significant contribution to the statistics of the fission widths (as is observed for the $3^-$ resonances of $^{235}$U and $0^+$ resonances of $^{239}$Pu, and also in the case of $^{233}$U and $^{241}$Pu).

The studies of Refs. 1 and 2 broke new ground and were the point of departure for the subsequent experimental and theoretical investigations of the $(n, \gamma f)$ reaction.

## 2. FIRST EXPERIMENTAL ESTIMATES OF THE PROBABILITY OF THE $(n,\gamma f)$ REACTION

After the publication of the theoretical studies of Refs. 1 and 2, several authors attempted to obtain an experimental confirmation of the existence of the $(n, \gamma f)$ reaction, using mainly analysis of the fission widths of resonances. The first claim of such experimental (indirect) detection of the $(n, \gamma f)$ reaction was the paper of Bowman *et al.*,[4] who considered the distribution of the fission widths of ten resonances of $^{238}$Pu in the region of neutron energies below 122 eV. To estimate the number of effective fission channels, they used the formula of Wilets:[5]

$$\nu_{\text{eff}} = \frac{2\langle \Gamma_f \rangle^2}{\langle \Gamma_f^2 \rangle - \langle \Gamma_f \rangle^2} \qquad (3)$$

Two forms of calculation ($\Gamma = \Gamma_n + \Gamma_\gamma$, since $\Gamma_f \ll \Gamma_\gamma$ and $\Gamma = \Gamma_n + \Gamma_\gamma + \Gamma_f$) gave $\nu_{\text{eff}} = 7 \pm 1.7$ and $\nu_{\text{eff}} = 6 \pm 1.5$,

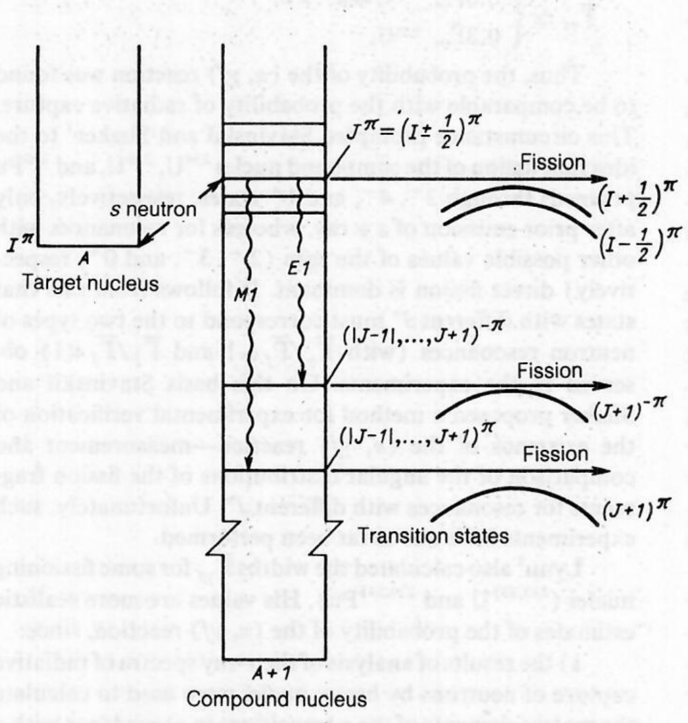


FIG. 1. Scheme of the $(n\,\gamma f)$ reaction.[2] Parabolas show the transition (lowest states) at the saddle point of the fission barrier.

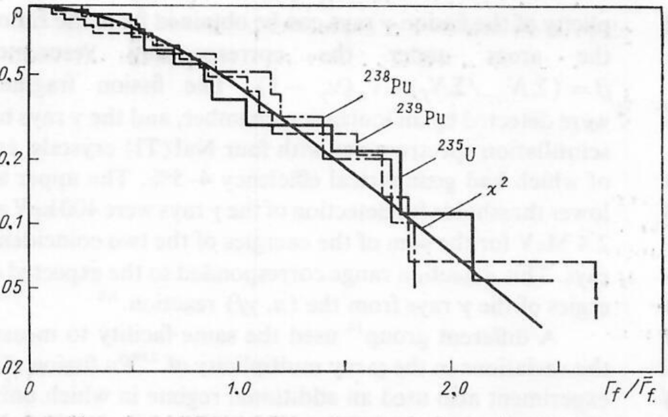


FIG. 2. Experimental and calculated distributions of fission widths for various isotopes.[6]

respectively. This large number of effective fission channels (i.e., the small variance of the distribution of the fission widths) for $^{238}$Pu was appreciably greater than the corresponding values calculated by the authors in accordance with (3) for $^{232,233,235}$U, $^{239,241}$Pu, and $^{241}$Am ($\nu_{\rm eff} \leqslant 2$). On the basis of this fact it was concluded in Ref. 4 that the fissioning of the $^{238}$Pu nucleus by slow neutrons proceeds through the $(n, \gamma f)$ reaction. They reached the same conclusion by considering the results of measurement of the cross sections $\sigma_f$ and $\sigma_\gamma$ for $^{240}$Pu for neutrons of a nuclear explosion, which revealed constancy of the ratio $\Gamma_f/\Gamma_\gamma$ in the region of neutron energies below 200 eV. Since the value of $\Gamma_\gamma$ is constant, the constancy of $\Gamma_f/\Gamma_\gamma$ indicates small fluctuations of $\Gamma_f$ and, therefore, a large $\nu_{\rm eff}$ and the presence of the $(n, \gamma f)$ reaction in the below-threshold fission of $^{240}$Pu. In announcing detection of the $(n, \gamma f)$ reaction on $^{238}$Pu, Bowman *et al.* could not fail to note that the results of their analysis depended strongly on the volume of the employed statistical material and its quality (experimental errors).

Vorotnikov and Otroshchenko[6] demonstrated that the method for detecting the $(n, \gamma f)$ reaction used by Bowman *et al.* was unreliable and that their conclusion in the case of $^{238}$Pu fission was incorrect. They compared the integrated distributions of $\Gamma_f/\bar{\Gamma}_f$ for $^{238}$Pu and for $^{239}$Pu (15 resonances with $J^\pi = 1^+$) and $^{235}$U (18 resonances with reduced yield of symmetric fragments). As can be seen in Fig. 2, all of the distributions are close to a $\chi^2$ distribution with $\nu = 5$, and the esimate in accordance with Wilet's formula gives $\nu_{\rm eff} = 6 \pm 1.5$ ($^{238}$Pu), $4.8 \pm 1.2$ ($^{235}$U), and $4.3 \pm 1.2$ ($^{239}$Pu). Thus, doubt was cast on the main result of the analysis of Bowman *et al.*—the anomalously narrow distribution of the fission widths of the $^{238}$Pu resonances—and therefore on the conclusion that the $(n, \gamma f)$ reaction on this nucleus exists. Before we consider the next study, we should note that the estimate in accordance with (3) of $\nu_{\rm eff}$ for $^{235}$U, $^{239}$Pu, and $^{238}$Pu using the data of the last publication of the Atlas BNL-325 (Ref. 7) gives the following values:

$^{238}$Pu: $\nu_{\rm eff} = 4.8 \pm 1.1$
(10 resonances, $E_n < 123$ eV),
$\nu_{\rm eff} = 0.8 \pm 0.1$ (49 resonances, $E_n < 500$ eV);
$^{239}$Pu: $\nu_{\rm eff} = 1.4 \pm 0.2$
(113 $1^+$ resonances, $E_n < 660$ eV),
$\nu_{\rm eff} = 1.4 \pm 0.2$ (47 $0^+$ resonances, $E_n < 660$ eV),
$^{235}$U: $\nu_{\rm eff} = 2.5 \pm 0.2$
(106 $4^-$ resonances, $E_n < 100$ eV),
$\nu_{\rm eff} = 2.8 \pm 0.2$ (88 $3^-$ resonances, $E_n < 100$ eV).

These values of $\nu_{\rm eff}$ also refute the results of the analysis of Bowman *et al.* and their conclusion concerning detection of the $(n, \gamma f)$ reaction in the fission of $^{238}$Pu by slow neutrons.

Vandenbosch[8] analyzed data on the anisotropy of $^{239}$Pu photofission and used the results to determine the position and curvature of the lowest fission barriers with $K^\pi = 1/2^-$ and $3/2^-$. Assuming that in the first $\gamma$ transitions only $E1$ emission is present (since it is such emission that leads to intermediate compound states with $K^\pi = 1/2$ and $3/2^-$), and taking the shape of the $\gamma$ spectrum in accordance with Lynn's study,[2] Vandenbosch obtained $\bar{\Gamma}_{\gamma f} \approx 1$ meV. His value of the width of the $(n, \gamma f)$ reaction is in good agreement with the minimal observed width $\Gamma_f = 1.2$ meV for the resonance 2.91 eV given by Bowman *et al.*, and Vandenbosch took this as a weighty argument in support of the correctness of his estimate of the width $\bar{\Gamma}_{\gamma f}$.

An interesting method of estimating the contribution of the $(n, \gamma f)$ reaction to the total fission width was proposed by Luk'yanov and Shaker.[9,10] The method is based on use of a generalized Porter–Thomas distribution[11] to analyze the distribution of fission widths, by means of which one can take into account arbitrary relative contributions of the various fission channels to the mean width. Making a statistical analysis of the experimental data[12] on the fission widths of $^{235}$U, they obtained[9] $\bar{\Gamma}_f(4^-) = 25$ meV, $\bar{\Gamma}_f(3^-) = 100$ meV, $\nu(4^-) \approx 6$, $\nu(3^-) \approx 14$. In arriving at this result, they assumed for the system of levels with $J^\pi = 4^-$ equally probable contributions of the various channels, while for the $J^\pi = 3^-$ levels 70% of the fission width was attributed to the two direct-fission channels, and the remaining 30% was ascribed to 12 equally probable channels corresponding to the $(n, \gamma f)$ reaction. At the same time, they noted that the results of their analysis for the number of fission channels and their relative contributions depended strongly on the structure of the experimental histogram giving the distribution of the fission widths. Confirmation was provided by the results of another study[10] of the same authors in which a different set of parameters was obtained for the same nucleus $^{235}$U but on the basis of different statistical material.

Thus, concluding our review of the early studies that obtained indirect evidence and estimates of the probability of the $(n, \gamma f)$ reaction on the basis of analysis of fission widths of resonances, we must say that these studies did not give an unambiguous answer to the question of the existence of the $(n, \gamma f)$ reaction, and the estimates obtained for its probability were unreliable, mainly because of the limited amount of experimental data used.

## 3. METHODS OF EXPERIMENTAL INVESTIGATION OF THE $(n, \gamma f)$ REACTION

The experimental methods that we now consider are based on the detection of prompt $\gamma$ rays and secondary fast fission neutrons.

If besides direct fission there does exist the $(n, \gamma f)$ reaction, then the experimentally observed fission $\gamma$ rays are a manifestation of the sum of these two mechanisms. Gamma rays are emitted from fission fragments mainly in the final stage of their de-excitation, after the emission of secondary neutrons. It may therefore be assumed that the excitation energy of the compound nucleus has a weak influence on fission properties such as the multiplicity and shape of the spectrum of $\gamma$ rays from fragments. And although this assumption itself requires experimental investigation, it does give a method for detecting and estimating the probability of the $(n, \gamma f)$ reaction by measurement of the variations of the multiplicity and total energy of the fission $\gamma$ rays.

As regards the secondary fission neutrons, the dependence of their multiplicity on the excitation energy is well known from measurements with fast neutrons. It can be assumed that the decrease in the energy of the compound nucleus after the emission of a prefission $\gamma$ ray leads to a decrease in the multiplicity of the secondary neutrons. Under such assumptions, we can, if the $(n, \gamma f)$ reaction does exist, write down the following relations for the multiplicity $\nu_\gamma$ of the fission $\gamma$ rays and their total energy $E'_\gamma$, and also for the multiplicity $\nu_n$ of the fission neutrons:

$$\nu_\gamma = \bar{\nu}_{\gamma 0} + \frac{\bar{\Gamma}_{\gamma f}}{\Gamma_f}\bar{\nu}_{\gamma f},\quad E'_\gamma = \bar{E}'_{\gamma 0} + \frac{\bar{\Gamma}_{\gamma f}}{\Gamma_f}\bar{E}_{\gamma f},$$
$$\nu_n = \bar{\nu}_{n0} - \frac{\bar{\Gamma}_{\gamma f}}{\Gamma_f}\left(\frac{\partial \nu_n}{\partial E^*}\right)\bar{E}_{\gamma f}, \qquad (4)$$

where $\bar{\nu}_{\gamma f}$ and $\bar{E}_{\gamma f}$ are the mean multiplicity and mean energy of the prefission $\gamma$ rays; $\bar{\nu}_{\gamma 0}$, $\bar{E}'_{\gamma 0}$, $\bar{\nu}_{n0}$ are, respectively, the mean multiplicity and total energy of the fission $\gamma$ rays and the multiplicity of the secondary fission neutrons in the absence of the $(n, \gamma f)$ reaction; and $\Gamma_f$ and $\bar{\Gamma}_{\gamma f}$ are the total fission width and the width of the $(n, \gamma f)$ reaction.

It is obviously sensible to measure the variations of $\nu_\gamma$, $E'_\gamma$, and $\nu_n$ for isolated neutron resonances with known spin and parity. In this case the quantities $\bar{\nu}_{\gamma 0}$, $\bar{E}'_{\gamma 0}$, and $\bar{\nu}_{n0}$ are averages over the set of measured resonances corresponding to direct fission. It can be seen from the relations (4) that if the $(n, \gamma f)$ reaction does exist, one must observe correlations between the values of $\nu_\gamma$, $E'_\gamma$, and $\nu_n$ and the reciprocal fission width $\Gamma_f^{-1}$ of the resonances, the values of $\nu_\gamma$ and $E'_\gamma$ being anticorrelated with $\nu_n$. It is precisely such correlations that are most frequently used to detect and investigate the $(n, \gamma f)$ reaction.

The first experimental investigation of the $(n, \gamma f)$ reaction by means of the multiplicity method was made by Borukhovich, Petrov, *et al.*,[13] who used the IBR-30 reactor at Dubna. They simultaneously measured the time-of-flight spectra of the yields of the $^{235}U$ fission fragments with and without coincidences with $\gamma$ rays, using the method of triple $f\gamma\gamma$ coincidences. In this case, information about the multiplicity of the fission $\gamma$ rays can be obtained from the ratio of the areas under the corresponding resonances: $\beta = (\Sigma N_{f\gamma\gamma}/\Sigma N_f) \sim \nu_\gamma(\nu_\gamma - 1)$. The fission fragments were detected by an ionization chamber, and the $\gamma$ rays by a scintillation spectrometer with four NaI(Tl) crystals, each of which had geometrical efficiency 4–5%. The upper and lower thresholds for detection of the $\gamma$ rays were 400 keV and 2.4 MeV for the sum of the energies of the two coincident $\gamma$ rays. This detection range corresponded to the expected energies of the $\gamma$ rays from the $(n, \gamma f)$ reaction.[1,2]

A different group[14] used the same facility to measure the variations in the $\gamma$-ray multiplicity of $^{239}Pu$ fission. This experiment also used an additional regime in which only $\gamma$ rays with total energy above 2.5 MeV were detected, i.e., there was a high reliability of detection of direct fission. In both experiments analysis of the experimental data enabled the authors to estimate the differences of the widths of the $(n, \gamma f)$ reaction for the two spin systems of the $^{235}U$ and $^{239}Pu$ resonances. A shortcoming of the employed method is that the contribution of the $(n, \gamma f)$ reaction cannot be detected when it is approximately the same for the two spin systems, even though relatively large.

The multiplicity method with detection of the $\gamma$ rays by NaI(Tl) detectors was used to investigate the $(n, \gamma f)$ reaction by a number of other authors. For example, Ryabov *et al.*[15,16] used the electron linac at Saclay to measure the variations of the $\gamma$-ray multiplicity of $^{239}Pu$ fission. The arrangement of this experiment is shown in Fig. 3.

Ryabov *et al.*[15] used the usual method of double $f\gamma$ coincidences, and fission events were detected with high efficiency by the secondary fission neutrons by means of the liquid scintillator NE-213. The $\gamma$ rays were detected by two NaI(Tl) detectors with a rather low detection efficiency $\varepsilon_\gamma \approx 2\%$ ($E_\gamma = 1$ MeV); this made it possible to avoid the effects of simultaneous detection of two or more $\gamma$ rays in the crystal.

For such a method of measurements, the ratio of the counting rates of fragment–$\gamma$-ray coincidences and of fission fragments is a number that is directly related to the multiplicity of the fission $\gamma$ rays:

$$\beta = \frac{N_{f\gamma}}{N_f} = \int_{E_{\gamma 1}}^{E_{\gamma 2}} \varepsilon_\gamma(E_\gamma)\,\nu_\gamma(E_\gamma)\,dE_\gamma, \qquad (5)$$

i.e., it corresponds to the true multiplicity $\nu_\gamma$ weighted by means of the energy dependence of the $\gamma$-detector efficiency. Therefore, the variations in the value of $\beta$ observed in the experiment reflect the variations of $\nu_\gamma$ or the $\gamma$-ray spectrum and, in principle both at once. Ryabov *et al.* found large variations of $\nu_\gamma$ from resonance to resonance, and these were correlated with the variations of $\nu_n$ and $E'_\gamma$, and also with the fission width $\Gamma_f$ of the resonances. From analysis of these correlations, they determined the widths $\Gamma_f$ for the $1^+$ resonances of $^{239}Pu$.

Using the neutron time-of-flight spectrometer GNEIS at Gatchina, Borukhovich *et al.*[17–19] measured the multiplicities of the $\gamma$ rays of $^{235}U$ and $^{239}Pu$ fission in the resonance region of neutron energies below 300 eV. The fission fragments were detected by means of multisection ionization chambers (Fig. 4), and the $\gamma$ rays were detected by two NaI(Tl) detectors, which had a lower detection threshold of 500 keV and an efficiency of not more than 2%. The back-

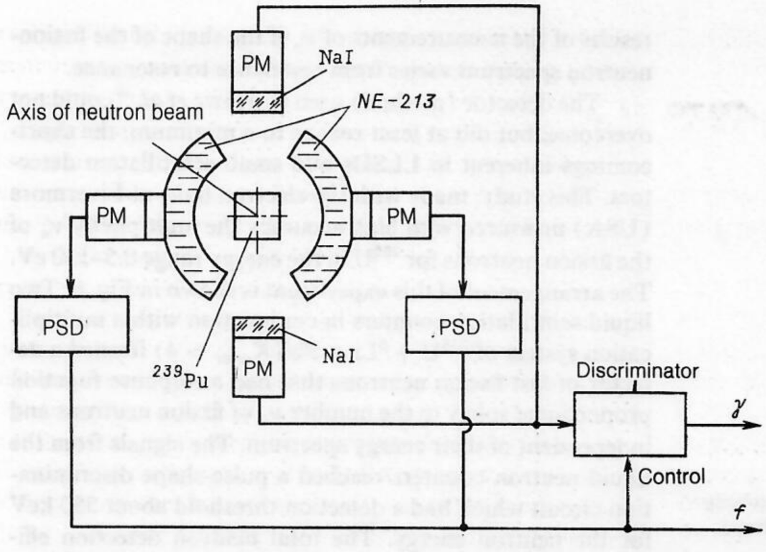


FIG. 3. Arrangement of the experimental facility at Saclay[15] for measuring the multiplicity of fission $\gamma$ rays (PSD: pulse-shape discriminator; PM: photomultiplier).

ground of random coincidences, which was determined mainly by radiative-capture $\gamma$ rays and was appreciable for resonances with small fission widths, was measured by means of the method of delayed coincidences. Analysis of the observed correlations between the multiplicity of the fission $\gamma$ rays and the reciprocal fission width $\Gamma_f^{-1}$ led to estimates of the widths $\Gamma_{\gamma f}$ for the $4^-$ resonances of $^{235}$U (Refs. 17 and 19) and the $1^+$ resonances of $^{239}$Pu (Refs. 18 and 19).

Besides measurements of the multiplicity, great interest attaches to measurements of the total energy of the fission $\gamma$ rays and of the variations of this energy in the resonance region of energies. The first experiment of this type was the study of Shackleton *et al.*[20] at Saclay. In this experiment the fission events were detected by means of an ionization chamber, and a large liquid scintillation detector (LLSD) of volume 520 liters (Fig. 5) was used to detect the $\gamma$ rays and measure their energies. The pulses from the $\gamma$ rays and the pulses associated with the secondary fission neutrons were separated by the delay time, since the neutrons were detected

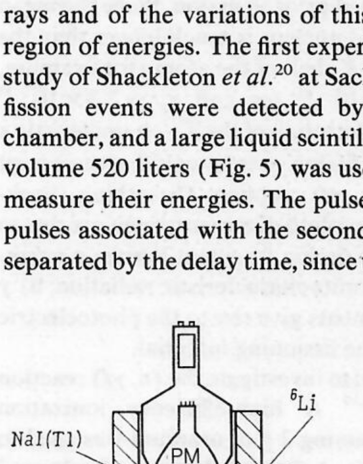


FIG. 4. Arrangement of experiment to measure the multiplicity of fission $\gamma$ rays with the GNEIS spectrometer[18,19] (PM: photomultiplier; IFC: ionization fission chamber).

by the LLSD mainly after they had been moderated by the protons of the scintillators. The moderated neutrons were mainly captured by Gd nuclei, which were present in the scintillator at the $\simeq 0.5\%$ level. A small correction for the effect of the detection of neutrons in the $\gamma$ channel (through fast recoil protons) was introduced in the process of analysis of the experimental data.

The results of measurements of the variations of $E'_\gamma$ for $^{239}$Pu resonances were given in Refs. 20–26. Data of analogous measurements for $^{235}$U are given in Refs. 22–26, and for $^{241}$Pu in Refs. 26–28. These experiments confirm the existence of a correlation of the total energy $E'_\gamma$ of the fission $\gamma$ rays with the reciprocal fission width $\Gamma_f^{-1}$ and with the $\gamma$-ray multiplicity $\nu_\gamma$, and also an anticorrelation of $E'_\gamma$ and the multiplicity $\nu_n$ of the fission neutrons.

Another method of measuring the variations of $E'_\gamma$ was used by Weston and Todd in a study[29] made with the ORELA accelerator at Oak Ridge. In this experiment the fission fragments were detected by an ionization chamber, and the $\gamma$ rays from the $^{239}$Pu fission were detected by two hydrogenless ($C_6F_6$) liquid scintillation detectors of small dimensions ($\varnothing = 100$ mm, $H = 50$ mm). To make the detection efficiency of $\gamma$ rays proportional to their energy, the detectors were operated in the regime of amplitude weighting.[30] The total efficiency of detection of a $\gamma$-ray cascade by such a detector is proportional to the total energy carried away by the cascade. The study of Weston and Todd also demonstrated the presence of a strong correlation between $E'_\gamma$ and $\Gamma_f^{-1}$, indicating manifestation of the $(n, \gamma f)$ reaction in the $^{239}$Pu resonances with low fission widths.

For the fissioning nuclei $^{235}$U and $^{239}$Pu, the most accurately measured out of the three quantities $\nu_\gamma$, $E'_\gamma$, and $\nu_n$ is the last, the multiplicity of the secondary fission neutrons. The first measurements of it for $^{239}$Pu (Refs. 31–34) in the resonance region of energies of the incident neutrons, made by different groups using different methods (mainly with the use of an LLSD), gave results that did not agree well with one another.

For example, the variations of $\nu_n$ as a function of the resonance spin measured at Dubna[31] and in the United States[32] were found to have opposite signs. The situation that had arisen at the beginning of the seventies, and also the

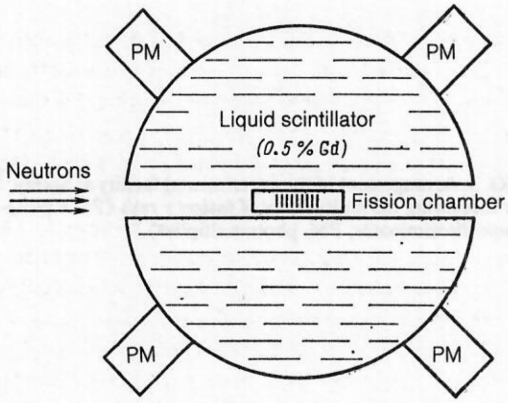


FIG. 5. Arrangement of experiment to measure the neutron multiplicity and total energy of fission $\gamma$ rays at Saclay[21,23] (PM: photomultipliers).

importance of such measurements for testing the basic propositions of A. Bohr's quantum theory of fission and for study of the $(n, \gamma f)$ reaction were a strong ground for the undertaking of new experiments.

In the investigations of Shackleton, Trochon, *et al.*[20–28] mentioned above, $\nu_n$ was measured by means of an LLSD. The efficiency of neutron detection by such a detector reached 70% (for neutrons from the spontaneous fission of $^{252}$Cf). In Ref. 35, Frehaut analyzed the various components of the background that exists for such measurements of $\nu_n$ and proposed a method for measuring them and taking them into account. Besides the $^{239}$Pu measurements,[20–26] the multiplicity $\nu_n$ of fission neutrons was measured at Saclay by means of the LLSD for the nuclei $^{235}$U (Refs. 22–26) and $^{241}$Pu (Refs. 26–28).

To measure $\nu_n$ for the fission of $^{239}$Pu by resonance neutrons, Trochon *et al.*[34] used a further method at Saclay, employing several small detectors. In this experiment the $^{239}$Pu sample was surrounded by four NE-213 liquid scintillation detectors ($\varnothing = 120$ mm, $H = 70$ mm). The employed scheme of discrimination based on the pulse shape had a $\gamma$-ray suppression coefficient of 1/1000 and neutron detection threshold at 1.2 MeV. Under these conditions, the efficiency of fission-neutron detection was $\lesssim 1\%$ per detector. The results showed that $\nu_n$ for the $^{239}$Pu resonances in the region 10–300 eV can be assumed to be constant to an accuracy of about 2%, i.e., the results of this study contradicted the data obtained by means of the LLSD.

When they measured the variations of $E'_\gamma$ for $^{239}$Pu, Weston and Todd[29] also measured the variations of the multiplicity of the fission neutrons for this nucleus. To detect the neutrons, they used two liquid scintillation detectors of the type NE-213 ($\varnothing = 100$ mm, $H = 50$ mm), the detection efficiency of which did not exceed 0.5% for one detector and for one fission neutron. The $\gamma$-ray discrimination scheme employed in this study in the neutron channel (based on the pulse shape) had suppression coefficient 1/600. Compared with the methods of measurement of $\nu_n$ in which LLSDs were used, the main advantage of the method used in Refs. 29 and 34 to detect the fission neutrons was the very low level of the background. A shortcoming of the method was the dependence of the neutron detection efficiency on the kinetic energy of the neutrons, which cannot fail to influence the results of the measurements of $\nu_n$ if the shape of the fission-neutron spectrum varies from resonance to resonance.

The detector (method) used by Howe *et al.*[36] could not overcome, but did at least reduce to a minimum, the shortcomings inherent in LLSDs and small scintillation detectors. This study, made with the electron linac at Livermore (USA) measured with high accuracy the multiplicity $\nu_n$ of the fission neutrons for $^{235}$U in the energy range 0.5–130 eV. The arrangement of this experiment is shown in Fig. 6. Two liquid scintillation counters in conjunction with a multiplication system of $^{235}\text{U} + {}^6\text{Li} + \text{Pb}$ ($K_{\text{mult}} = 4$) formed a detector of fast fission neutrons that had a response function proportional solely to the number $\nu_n$ of fission neutrons and independent of their energy spectrum. The signals from the liquid neutron counters reached a pulse-shape discrimination circuit which had a detection threshold about 550 keV for the neutron energy. The total neutron detection efficiency of the complete detecting system was about 45%. An ionization chamber was used in the experiment to detect the fission fragments. The results of Howe *et al.* demonstrated the presence of small but statistically significant variations of $\nu_n$ for $^{235}$U in the resonance region of energies. At the same time, no correlation between $\nu_n$ and the fission width $\Gamma_f$ was found.

One further method of investigating the $(n, \gamma f)$ reaction, first proposed by Bogdzel' *et al.*,[37] was used in experiments with the IBR-30 reactor at Dubna. The essence of this method consisted in detection of the characteristic x rays of the atoms of the fissioning material produced by internal conversion of $\gamma$ rays emitted prior to fission. Since the mean lifetime $\tau$ of the compound nucleus is much longer than the lifetime $\tau_K$ of holes in the $K$ shell of the atom (for uranium, for example, $\tau = 10^{-13}$–$10^{-15}$ sec and $\tau_K = 7.3\times10^{-18}$ sec), it was assumed that detection of the $K_\alpha$ characteristic x rays in coincidence with fission events would give correct information about the $(n, \gamma f)$ reaction. Disturbing (background) events in such a method of measurement are due to two processes, when: a) a fission fragment ionizes a neighboring atom, which then emits characteristic radiation; b) $\gamma$ rays from the fission fragments give rise to the photoelectric effect on other atoms of the fissioning material.

This method was used to investigate the $(n, \gamma f)$ reaction on the $^{235}$U nucleus.[38,39] A high-efficiency ionization chamber ($\varepsilon \simeq 50\%$) containing 2 g of uranium was used to detect the fission fragments. A Ge(Li) detector of volume 7 cm$^3$ was used to detect the $\gamma$ rays. On the basis of the data of this experiment it was concluded[38,39] that the $(n, \gamma f)$ reaction on the $^{235}$U nucleus does exist, and its probability was estimated.

In the study of the $(n, \gamma f)$ reaction, the most complicated experiments are those that measure the spectra of the prefission $\gamma$ rays, since:

1) the fraction of prefission $\gamma$ rays in the mean multiplicity of the fission $\gamma$ rays is small ($\lesssim 12\%$);

2) the high mean multiplicity ($\bar{\nu}_\gamma = 7$–$8$) of the fission $\gamma$ rays forces one to use detectors with low efficiency in order to avoid superposition effects when the $\gamma$ rays are detected;

3) the measured event counting rates are small precisely for the resonances with small fission widths, for which the contribution of the $(n, \gamma f)$ reaction to the fission is most appreciable;

4) in the measurements one observes a mixture of $\gamma$ rays from direct fission and from the $(n, \gamma f)$ reaction, and therefore the experiment to measure the spectrum of the prefission $\gamma$ rays must be of the difference type methodologically, and this also requires a high statistical accuracy on account of the smallness of the effect.

The information which can be obtained from measurements of the spectra of the prefission $\gamma$ rays is of great interest. First, it is necessary for the correct interpretation of the results of the measurements of $\nu_\gamma$, $E'_\gamma$, and $\nu_n$. Second, information about the spectra makes it possible to study the type (electric or magnetic), multipolarity, and other properties of the $\gamma$ transitions between highly excited states of the compound nucleus. Third, it can give information about the states of the fissioning nucleus at large deformations (in the second well of the two-hump fission barrier), which as yet can be obtained only in reactions of the $(d, pf)$ type.

In an experiment performed at the electron linac at Saclay, Trochon *et al.*[40,41] made an attempt to measure with high energy resolution the spectrum of prefission $\gamma$ rays from $^{239}$Pu. Fission events in the $^{239}$Pu sample were detected through the secondary fission neutrons by means of four liquid scintillation detectors. A Ge(Li) detector was used to measure the spectrum of the $\gamma$ rays. Two of the measured $^{239}$Pu resonances were chosen for comparison: 10.93 eV, a background resonance for which the $(n, \gamma f)$ reaction is masked by direct fission, and the resonance at 44.48 eV, for which the contribution of the $(n, \gamma f)$ reaction was, on the basis of the results of measurements of $\nu_\gamma$, $E'_\gamma$, and $\nu_n$, expected to be most significant. Trochon *et al.* succeeded in finding in the difference spectrum at least one level corresponding to a vibrational state in the second well, populated by $\gamma$ transitions from the $(n, \gamma f)$ reaction.

Among the experiments of this type we can also include that of Refs. 38 and 39, performed at Dubna, since in it not only the time-of-flight spectra but also the pulse-height spectra of the fission $\gamma$ rays were measured. Unfortunately, determination of the spectra of the prefission $\gamma$ rays for individual resonances was not one of the aims of this investigation.

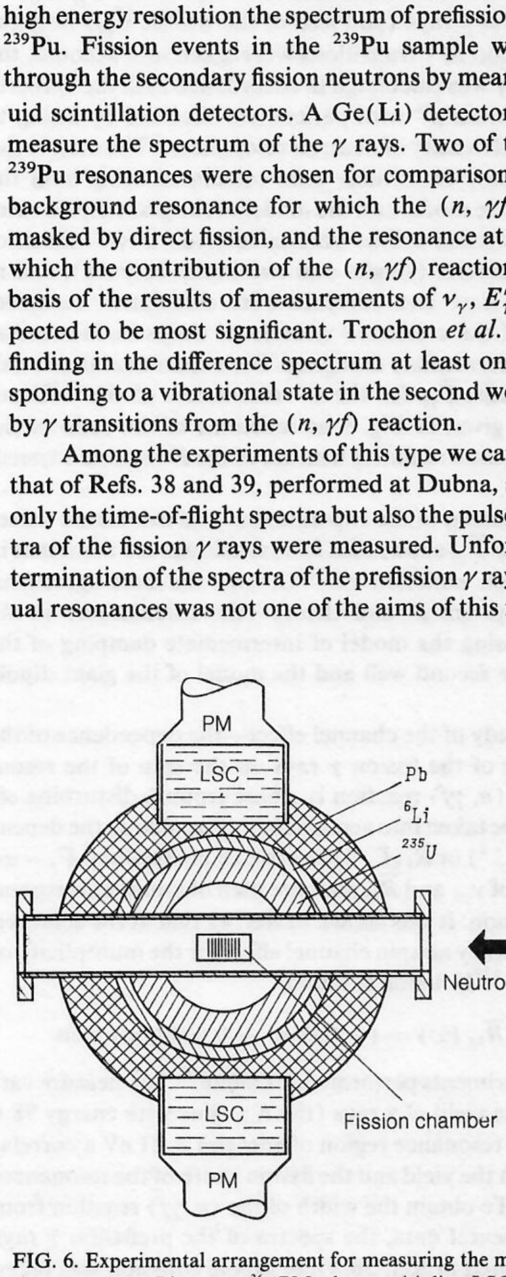


FIG. 6. Experimental arrangement for measuring the multiplicity of fission neutrons at Livermore[36] (PM: photomultiplier; LSC: liquid scintillation counter).

An experiment to measure the spectra of prefission $\gamma$ rays from $^{239}$Pu was performed with the GNEIS spectrometer at Gatchina.[19,42,43] The arrangement of the experiment and the detectors employed in it were the same as in the measurements of the multiplicity of the fission $\gamma$ rays (see Fig. 4). Seven $1^+$ resonances and one $0^+$ resonance of $^{239}$Pu in the region of energies below 100 eV were chosen. The measured amplitude spectra of the fission $\gamma$ rays were normalized to one fission event (using the areas of the corresponding resonances in the time-of-flight spectrum of the fragments). The results obtained in this study indicate that the spectra of the $\gamma$ rays from the fission fragments change little from resonance to resonance, while the shapes of the spectra for the $1^+$ resonances with large and small fission widths are different. The last circumstance can be explained by the presence of a component associated with the $(n, \gamma f)$ reaction.

Popeko *et al.*[44] used the VVR-M reactor at Gatchina to measure the spectra of conversion electrons and $\gamma$ rays from the fission of $^{239}$Pu by resonance and thermal neutrons. The resonance at 0.29 eV was separated by means of a samarium filter. The electron spectra were measured by means of a high-efficiency spectrometer with superconducting solenoids and by Si(Li) detectors, and the fission fragments were detected by two coaxial Si(Au) detectors. In the investigation of the $\gamma$-ray spectra, the fission fragments were detected by a gas scintillation chamber, and the $\gamma$ rays by means of an NaI(Tl) detector. In the opinion of the authors of the study, the measured difference spectra of the electrons and $\gamma$ rays of $^{239}$Pu fission could be satisfactorily explained by the existence of the $(n, \gamma f)$ reaction on this nucleus.

The results of the experiments whose methods have been considered in this section and a discussion of them are given below separately for each investigated nucleus.

## 4. RESULTS OF EXPERIMENTAL INVESTIGATIONS OF THE $(n, \gamma f)$ REACTION

### Results for $^{235}$U

The compound nucleus formed by capture of an *s* neutron in $^{235}$U has spin and parity $3^-$ or $4^-$. The spins of the $^{235}$U resonances in the region below 100 eV have been fairly reliably identified, mainly by the direct measurements of Moore *et al.*[45] with polarized neutrons on polarized nuclei. However, since the mean distance between the levels of the two spins ($\langle D \rangle \simeq 0.438$ eV) is comparable with the mean widths $[\langle \Gamma_f(3^-) \rangle \simeq 0.180$ eV and $\langle \Gamma_f(4^-) \rangle \simeq 0.091$ eV$]$ of the resonances,[45] the determination of any of the effects observed in the individual resonances involves the difficulty of taking into account the contribution from the neighboring resonances.

Borukhovich *et al.*[13] measured the multiplicity of the fission $\gamma$ rays for thermal neutrons and 13 resonances in the region of neutron energies below 35 eV. The results of the measurements (yields of $f\gamma\gamma$ coincidences) are shown in Fig. 7. This experiment did not reveal statistically significant deviations of the values of $\beta$ from the mean value, and the relative difference of the yields of $f\gamma\gamma$ coincidences was found to be $\eta_{\rm exp} = [\bar\beta(3^-) - \bar\beta(4^-)]/\bar\beta(3^-) = 0.014 \pm 0.008$. Comparison of $\eta_{\rm exp}$ with the theoretical estimates of this quantity showed that the best agreement with experiment is obtained for Lynn's estimate[2] ($\eta_{\rm teor} = 0.006$–$0.015$), whereas the estimates of Stavinskiĭ and Shaker[1]

($\eta_{teor} = 0.23$–$0.32$) and Luk'yanov and Shaker[10] ($\eta_{teor} = 0.23$–$0.26$) wee overestimated. The statistical accuracy achieved enabled the authors of the experiment to estimate the difference of the widths of the $(n, \gamma f)$ reaction for the two spin states: $|\bar{\Gamma}_{\gamma f}(3^-) - \bar{\Gamma}_{\gamma f}(4^-)| \leqslant 2$–$3$ meV. The possible existence of a correlation between the multiplicity of the fission $\gamma$ rays and the reciprocal fission width of the $^{235}$U resonances was not investigated in the experiment.

Figure 8 shows the results of measurements of the multiplicity of the $\gamma$ rays of $^{235}$U fission in the region 4–130 eV, made with the GNEIS neutron spectrometer.[17,19,42] The relative multiplicity was determined in this experiment for 21 isolated resonances and 30 energy intervals that included two or more resonances, the boundaries of the intervals being taken to be the same as in Ref. 36. The relative multiplicity for point $i$ (a resonance or interval) was defined as $R_i = \beta_i / \langle \beta \rangle$, where $\beta_i$ was calculated in accordance with the expression (5), and $\langle \beta \rangle$ is the mean value. The correspondence of the measured values of the multiplicity $R_i$ to a normal (Gaussian) distribution was tested by the calculation of

$$\chi^2 = \frac{1}{N-1} \sum_{i=1}^{N} \left( \frac{R_i - \langle R \rangle}{\Delta R_i} \right)^2, \tag{6}$$

where $N$ is the number of points.

For the complete set of measured values ($N = 51$) the value $\chi^2 = 11.28$ was obtained, indicating a definitely nonstatistical distribution of the $R_i$. Nonstatistical behavior was also inherent in the distribution of the $R_i$ within the groups corresponding to the two spin states: $\chi^2(3^-, N = 8) = 4.39$ and $\chi^2(4^-, N = 16) = 2.94$.

The study found a statistically significant correlation between the multiplicities $R_i$ and the reciprocal fission width $\Gamma_f^{-1}$ of the resonances for the $4^-$ resonances with $r(R_i, \Gamma_f^{-1}) = 0.50 \pm 0.20$ and for the $3^-$ resonances with $r(R_i, \Gamma_f^{-1}) = 0.63 \pm 0.24$. Such correlations, and also the correlations between $R_i$ and the yields $Y_\gamma$ of the characteristic x rays (data of Refs. 38 and 39) with $r(R_i, Y_\gamma) = 0.63 \pm 0.18$ and the anticorrelations between $R_i$ and the multiplicity $\nu_n$ of the fission neutrons (data of Howe *et al.*[36]) with $r(R_i, \nu_n) = -0.54 \pm 0.18$ for the $4^-$ resonances enabled the authors of the study to conclude that the $(n, \gamma f)$ reaction on $^{235}$U does exist.

The results of the measurements of the multiplicity of the fission $\gamma$ rays obtained with the GNEIS spectrometer were used to estimate the widths $\bar{\Gamma}_{\gamma f}$ for the $^{235}$U resonances:

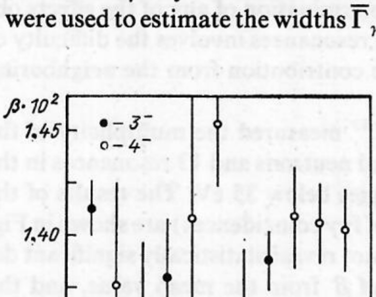


FIG. 7. Yields of triple $f\gamma\gamma$ coincidences for $^{235}$U resonances.[13]

$4^-$ resonances, $\bar{\Gamma}_{\gamma f} \leqslant 1.2$ meV (Refs. 17 and 19);

$4^-$ resonances, $\bar{\Gamma}_{\gamma f} = (0.32 \pm 0.13)$ meV; (Ref. 42)
$3^-$ resonances, $\bar{\Gamma}_{\gamma f} = (0.87 \pm 0.89)$ meV. (Ref. 42)

For the estimates of $\bar{\Gamma}_{\gamma f}$ the expressions (4) and (5), which can be reduced to the linear form $R_i = A\Gamma_f^{-1} + B$, where $A$ and $B$ are constants, were used. It was assumed that the width $\Gamma_{\gamma f}$ fluctuates weakly and that the spectrum and multiplicity of the $\gamma$ rays from the fission fragments do not change from resonance to resonance. The width $\bar{\Gamma}_{\gamma f}$ was calculated in accordance with the formula

$$\bar{\Gamma}_{\gamma f} = A \langle \beta \rangle \Big/ \int_0^\infty \nu_{\gamma f}(E_\gamma)\, \varepsilon_\gamma(E_\gamma)\, dE_\gamma. \tag{7}$$

The slope $A$ was calculated from the experimental data by the least-squares method, and the detection efficiency $\varepsilon_\gamma(E_\gamma)$ of the $\gamma$-ray detector was calculated by the Monte Carlo method. In the calculations of the spectra $\nu_{\gamma f}(E_\gamma)$ of the prefission $\gamma$ rays (and also of the widths $\bar{\Gamma}_{\gamma f}$) only the dipole $E1$ and $M1$ transitions were taken into account; the level density was calculated in accordance with the Gilbert–Cameron formula[46] with parameters obtained by fitting to the observed density of neutron resonances.[7] The matrix elements of the $\gamma$ transitions were calculated using both the single-particle model and the model of the giant dipole resonance. The fission widths were calculated for two models of the fission barrier (single- and two-hump barriers), and in the second case two variants were calculated: complete damping of the collective vibrational states in the second well and intermediate damping. The experimental and calculated widths $\bar{\Gamma}_{\gamma f}$ for the $4^-$ resonances of the $^{235}$U nucleus[42] are given in Fig. 9 as functions of the ratio of the intensities (total radiative widths) of the $E1$ and $M1$ transitions.

Comparison of the experimental and calculated values of the widths $\bar{\Gamma}_{\gamma f}$ shows that $M1$ transitions are dominant in the prefission emission of $^{236}$U, and the best agreement between experiment and theory (the calculation) is obtained by using the model of intermediate damping of the states in the second well and the model of the giant dipole resonance.

In a study of the channel effect—the dependence of the multiplicity of the fission $\gamma$ rays on the spin of the resonances—the $(n, \gamma f)$ reaction is a background, disturbing effect. It can be taken into account by extrapolating the dependence $\nu_\gamma(\Gamma_\gamma^{-1})$ or $\bar{R}_i(\Gamma_f^{-1})$ to zero, i.e., to the limit $\Gamma_f \to \infty$. The values of $\nu_{\gamma 0}$ and $\bar{R}_{i0}$ that are then obtained correspond to direct fission. It was shown in Ref. 42 that at the achieved level of accuracy no spin channel effect for the multiplicity of $\gamma$ rays from $^{235}$U fission is found:

$$\bar{R}_{i0}(3^-) - \bar{R}_{i0}(4^-) = (-0.0010 \pm 0.0040) \text{ rel. units}$$

In experiments performed at Dubna[38,39] to measure variations in the yield of x rays (the $K_{\alpha 1}$ line with energy 98.4 keV) in the resonance region of energies 2–21 eV a correlation between the yield and the fission width of the resonances was found. To obtain the width of the $(n, \gamma f)$ reaction from the experimental data, the spectra of the prefission $\gamma$ rays were calculated in Ref. 38. Only dipole emission was taken into account, the level density was calculated in accordance with the Gilbert–Cameron formula, and the matrix elements

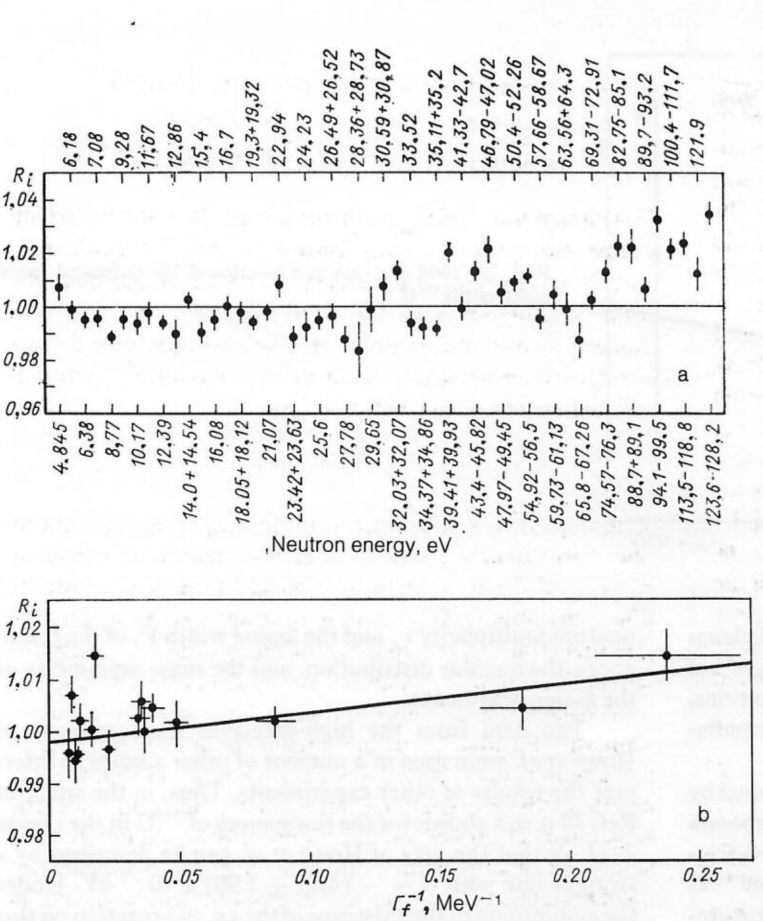


FIG. 8. Multiplicity of $\gamma$ rays from $^{235}U$ fission (Refs. 17, 19, and 42): a) variations of the multiplicity; b) correlations of the multiplicity and the reciprocal fission width for $4^-$ resonances.

of the $\gamma$ transitions were calculated both by the single-particle model of Weisskopf and by the model of a giant dipole resonance. The probability of transition through the fission barrier was calculated for two models of the barriers (single- and two-hump). Comparison of the different values of the widths $\overline{\Gamma}_{\gamma f}$ obtained from the experimental data for different types of calculation with the observed $\Gamma_f^{\min}(^{235}U)$ enabled the authors of Ref. 38 to conclude that, among the $\gamma$ transitions that precede fission, transitions of the type $M1$ must be predominant, and that $\overline{\Gamma}_{\gamma f} \simeq 4$ meV.

In a further series of measurements[39] which used the dependence of $R_i$ on $\Gamma_f$ in the form of a linear function of $(\Gamma_f^i)^{-1}$ (Fig. 10), the parameter values $A = (18.1 \pm 10.2) \times 10^{-5}$ meV and $B = (7.8 \pm 0.5) \times 10^{-5}$ were obtained by the least-squares method. From the slope $A$ of the line the relation

$$\left\langle \frac{\alpha_k}{1+\alpha_k} \right\rangle \langle \Gamma_{\gamma f} \rangle = (0{,}23 \pm 0.13) \text{ meV}, \tag{8}$$

where $\alpha_k$ is the coefficient of internal conversion of the prefission $\gamma$ rays, was obtained. Combining the relation (8) with another, also experimentally obtained, expression for the $4^-$ resonances of the $^{235}U$ nucleus,

$$\langle E_{\gamma f} \rangle \langle \Gamma_{\gamma f} \rangle = (1590 \pm 710) \text{ eV}^2, \tag{9}$$

the authors of Ref. 39 arrived at the equation

$$\langle E_{\gamma f} \rangle (0{,}145 \pm 0.107) = \left\langle \frac{\alpha_k}{1+\alpha_k} \right\rangle. \tag{10}$$

This analysis showed that Eq. (10) is satisfied best by the spectrum of prefission $\gamma$ rays calculated in accordance with the model of a giant dipole resonance for $M1$ transitions. The following estimates were obtained for the width of the $(n, \gamma f)$ reaction and for the mean energy of the spectrum of the prefission $\gamma$ rays:

$$\overline{\Gamma}_{\gamma f}(M1) = \left(2{,}1 \begin{array}{c} +1.5 \\ -1.7 \end{array}\right) \text{meV}, \quad \overline{E}_{\gamma f}(M1) = \left(750 \begin{array}{c} +400 \\ -130 \end{array}\right) \text{keV}.$$

According to an estimate of the authors, the maximal admixture of $E1$ transitions in the spectrum of the prefission $\gamma$ rays does not exceed 50%. Such an enhancement of the $M1$ tran-

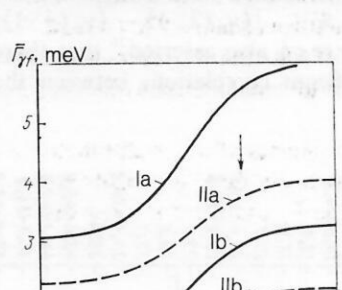


FIG. 9. Experimental and calculated widths $\overline{\Gamma}_{\gamma f}$ for $4^-$ resonances of the $^{235}U$ nucleus.[42] The arrow indicates the ratio of the intensities of the $E1$ and $M1$ transitions observed in the radiative capture of neutrons. The different calculations are as follows: I) single-hump barrier; II) two-hump barrier (complete damping); III) two-hump barrier (intermediate damping); a) Weisskopf model for probabilities of $\gamma$ transitions; b) and c) model of giant dipole resonance (probability of partial $\gamma$ transitions proportional to $\sim E_\gamma^4$ and $\sim E_\gamma^4$, respectively).

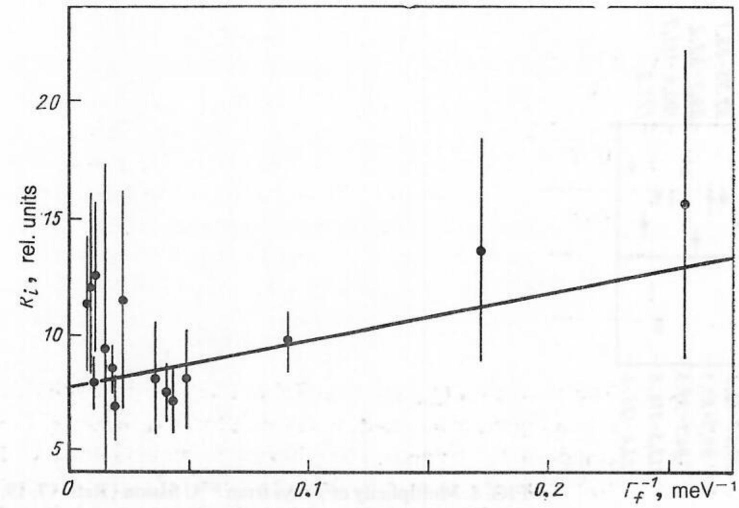


FIG. 10. Yield of x rays as a function of the reciprocal fission width of the $^{235}$U

sitions between the highly excited states requires an explanation itself, since it contradicts a well-known estimate[47] of the ratio of the intensities of $E1$ and $M1$ transitions, $\overline{\Gamma}_\gamma(M1)/\overline{\Gamma}_\gamma(E1) \simeq 0.2$, obtained from analysis of the radiative widths of heavy nuclei.

In the accuracy achieved, the experiment performed by Howe *et al.*[36] with the electron linac at Livermore surpasses all the other studies devoted to measurement of the variations of the multiplicity of the secondary neutrons of $^{235}$U fission in the resonance region. The results of these measurements for 33 isolated resonances and 23 resonance groups are given in Fig. 11. The data show that there is a definite structure in the energy dependence of $\nu_n$ within the measured range 0.5–125 eV. Thus, whereas for the ensemble of 56 points the value $\chi^2/55 = 1.25$ is obtained, the values grouped in 20 intervals (with the same statistical error of about 0.16%) give $\chi^2/19 = 3.45$, which is appreciably larger than one could expect for a random distribution of $\nu_n$ around a mean value.

For the isolated resonances no significant difference was found between the mean neutron multiplicities for the spin states $3^-$ and $4^-$: $|\langle\nu_n(3^-)\rangle - \langle\nu_n(4^-)\rangle| = 0.0010 \pm 0.0014$. Howe *et al.* also asserted[36] that there were no statistically significant correlations between the neutron multiplicity $\nu_n$ and the fission width $\Gamma_f$ of the resonances, the angular distribution, and the mass asymmetry of the fission fragments.

The data from the high-precision measurements of Howe *et al.* were used in a number of other studies to interpret the results of other experiments. Thus, in the study of Ref. 39 it was shown for the resonances of $^{235}$U in the region 2–21 eV that the data of Howe *et al.* can be described by a straight line with $A = -(4.67 \pm 2.90) \times 10^{-2}$ eV. Under the assumption of the existence of the $(n, \gamma f)$ reaction on this nucleus, the coefficient $A$ can be represented in the form

$$A = \frac{\overline{\Gamma}_{\gamma f}}{\langle\nu_n\rangle} \overline{E}_{\gamma f} \left(\frac{\partial\nu_n}{\partial E^*}\right). \qquad (11)$$

For the employed values $\langle\nu_n\rangle = 2.393 \pm 0.080$ and $\partial\nu_n/E^* = -0.1077$ neutron/MeV (Ref. 48) it was found in Ref. 39 that $\overline{\Gamma}_{\gamma f}\overline{E}_{\gamma f} = 1037 \pm 645$ eV, a value that to within the experimental errors agrees with the data of Trochon *et al.*[25] [see the expression (9)].

Results of experimental investigations of the $(n, \gamma f)$ reaction on $^{235}$U made at Saclay were given in Refs. 16 and 22–26. Measurements of the multiplicity of the fission $\gamma$ rays[16] showed that in the case of $^{235}$U its variations in the resonance region are small and do not enable one to establish whether

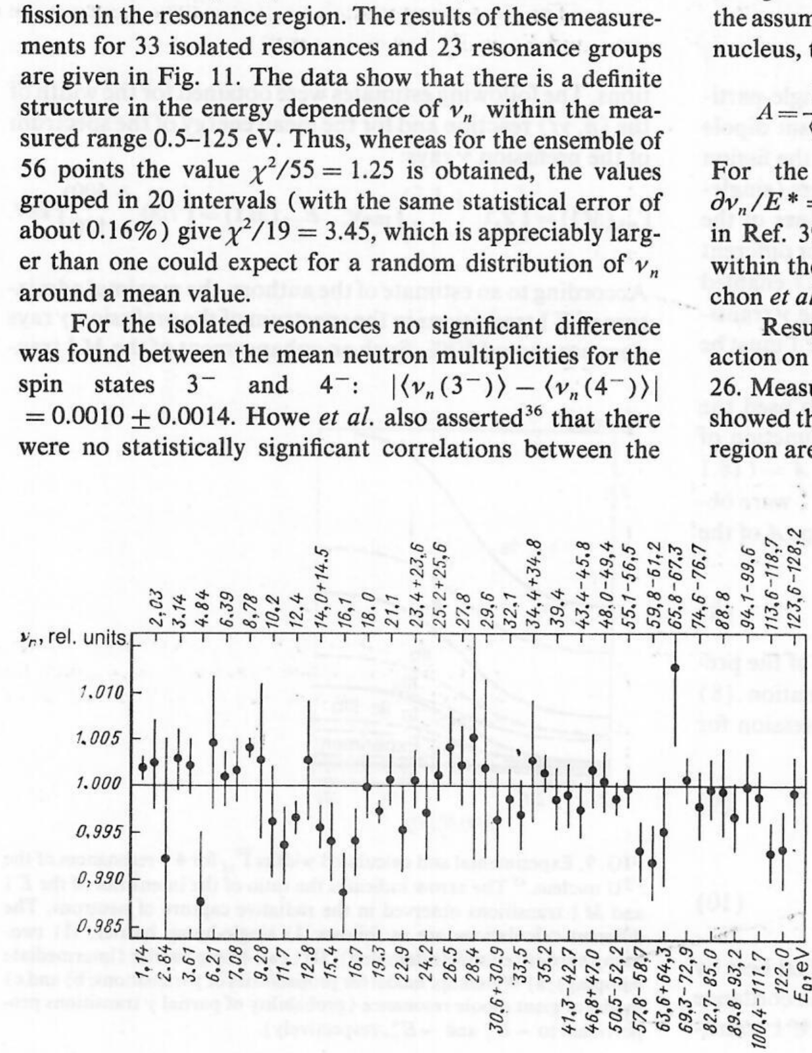


FIG. 11. Variations of the relative multiplicity of secondary fission neutrons of $^{235}$U in the reonance region of energies $E_n < 130$ eV (Ref. 36).

TABLE I. Results of statistical analysis of data for the $^{236}U$ nucleus.[23]

| $J^\pi$ | Number of resonances | $Q_{hx}(\nu_n)$ | $Q_{hx}(E'_\gamma)$ | $r(\nu_n, E'_\gamma)$ | $Q_r$ |
|---|---|---|---|---|---|
| $3^-$ | 11 | 0.99 | 0.006 | −0.55 | 0.08 |
| $4^-$ | 26 | 0.19 | 0.28 | −0.10 | 0.61 |
| unknown | 7 | 0.18 | <0.00005 | 0.06 | 0.89 |

or not the $(n, \gamma f)$ reaction exists on this nucleus. The results of measurements of $\nu_n$ and $E'_\gamma$ in the region 2–58 eV were discussed in Refs. 22, 24, and 25. Analysis of the correlations of $\nu_n$ and $E'_\gamma$ with the fission width $\Gamma_f$ yielded two concordant estimates of the product $\overline{\Gamma}_{\gamma f}\overline{E}_{\gamma f}$ for the $4^-$ resonances of $^{235}U$: $1590 \pm 710\ \text{eV}^2$ ($\nu_n$ data) and $1370 \pm 610\ \text{eV}^2$ ($E'_\gamma$ data).

The fullest accounts of the results and method of analyzing the experimental data are given in Ref. 23. Figure 12 shows data from this study. To explain the nature of the variations of $\nu_n$ and $E'_\gamma$ from resonance to resonance Shackleton used the so-called Birge test (criterion):[49]

$$hx = \sqrt{N}\left[\sqrt{\frac{1}{N}\sum_{i=1}^{N}\left(\frac{x_i - \langle x\rangle}{\sigma_i}\right)^2} - 1\right], \text{where} \langle x\rangle = \frac{\sum x_i/\sigma_i^2}{\sum 1/\sigma_i^2} \quad (12)$$

The quantities $Q_{hx}$ given in Table I are the probabilities of obtaining the calculated values of $hx$ for $N$ variables $x_i$ (with standard deviations $\sigma_i$) that have a normal (Gaussian) distribution. It can be seen from the data of Table I that the variations of $E'_\gamma$ for the $3^-$ resonances have a clearly nonstatistical nature, and this is also true for the resonances whose spin was assumed to be unknown. The variations of the other quantities have an entirely statistical origin. For the $3^-$ resonances there was found to be a fairly strong anticorrelation of $\nu_n$ and $E'_\gamma$, the existence of which is predicted by the theory of the $(n, \gamma f)$ reaction. For the correlation coefficient $r(\nu_n, E'_\gamma) = -0.55$ the quantity $Q_r = 0.08$ gives the significance level or probability of obtaining such a value of $r$ by chance under the condition that the "true" correlation coefficient is zero.

The least-squares analysis of the linear dependence of $\nu_n$ and $E'_\gamma$ on $\Gamma_f^{-1}$ gave the results

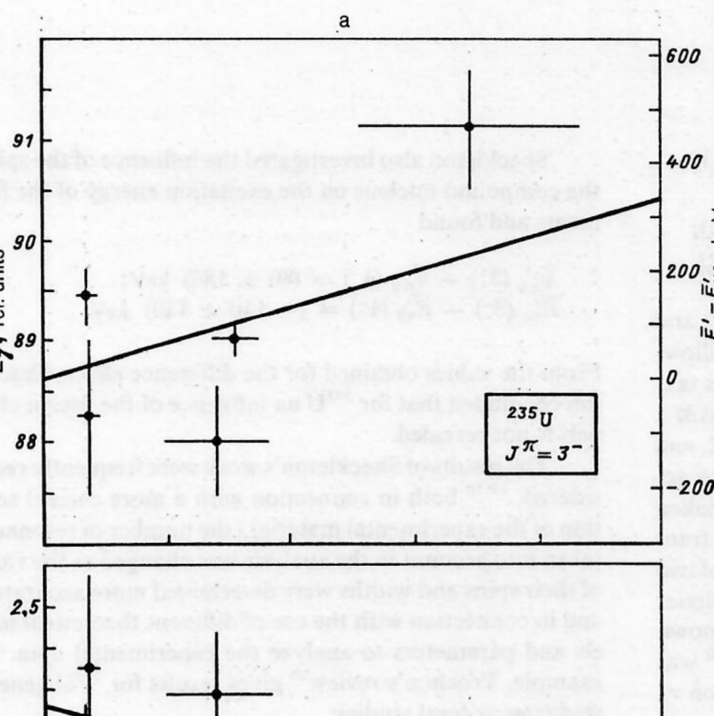


FIG. 12. Correlations of the neutron multiplicity $\nu_n$, the total energy $E'_\gamma$ of the fission $\gamma$ rays, and the reciprocal fission width of $^{235}U$ resonances (Ref. 23): a) $3^-$ resonances; b) $4^-$ resonances.

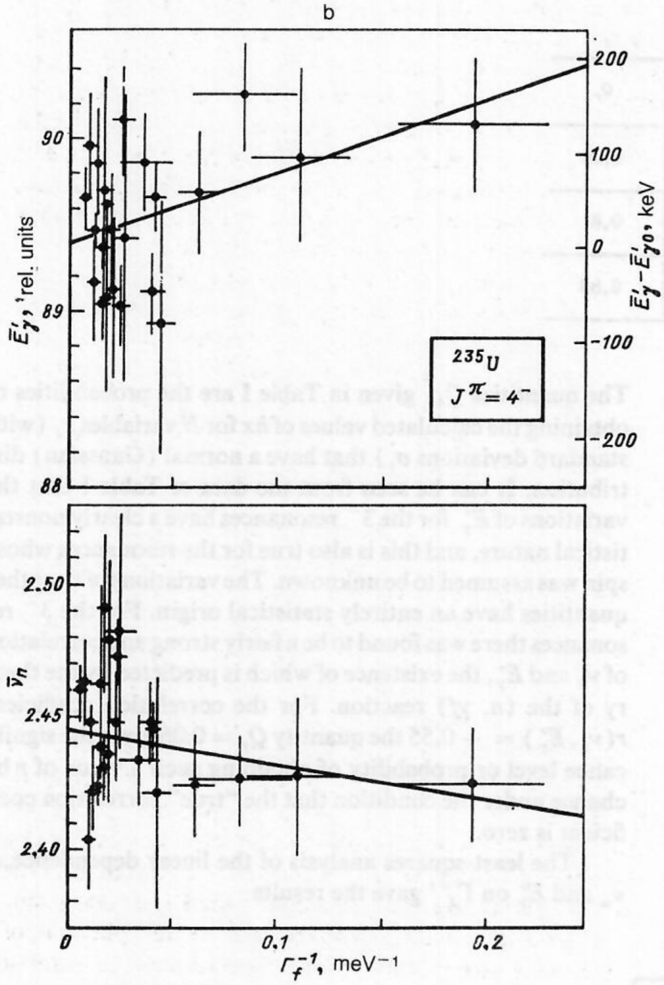


FIG. 12. *(Continued)*

$$4^- \text{ resonances: } \langle \overline{\Gamma}_{\gamma f}\overline{E}_{\gamma f}\rangle \begin{cases} = (1500 \pm 1400) \ \text{eV}^2 \ (\nu_n); \\ = (900 \pm 400) \ \text{eV}^2 \ (E'_\gamma); \end{cases}$$

$$3^- \text{ resonances: } \langle \overline{\Gamma}_{\gamma f}\overline{E}_{\gamma f}\rangle \begin{cases} = (7300 \pm 3300) \ \text{eV}^2 \ (\nu_n); \\ = (3000 \pm 4000) \ \text{eV}^2 \ (E'_\gamma). \end{cases}$$

The theoretical estimates of the product $\langle \overline{\Gamma}_{\gamma f}\overline{E}_{\gamma f}\rangle$ and of its factors were obtained by Shackleton under the following conditions: only the first prefission $\gamma$ transition was taken into account; the ratio of the intensities of the $E1$ and $M1$ transitions was taken equal to $\overline{\Gamma}_\gamma(M1)/\overline{\Gamma}_\gamma(E1) \simeq 0.2$, and transitions of higher multipolarities was not taken into account; the level density of the compound nucleus was taken in accordance with the Gilbert–Cameron formula; the transition matrix elements were taken in the form of sums of two Lorentz curves; and a model of a two-hump fission barrier with penetrability calculated by means of the well-known Hill–Wheeler approximation of a parabolic barrier[50] was used. Taking as the final experimental results the data on $\nu_n$ for the $3^-$ resonances and the data on $E'_\gamma$ for the $4^-$ resonances, Shackleton obtained the best agreement with experiment for the following calculated values:

$$4^- \text{ resonances: } \langle \overline{\Gamma}_{\gamma f}\overline{E}_{\gamma f}\rangle = 1270 \ \text{eV}^2,$$
$$\overline{\Gamma}_{\gamma f} = 1.18 \ \text{meV}, \ \overline{E}_{\gamma f} = 1.08 \ \text{meV};$$
$$3^- \text{ resonances: } \langle \overline{\Gamma}_{\gamma f}\overline{E}_{\gamma f}\rangle = 2260 \ \text{eV}^2,$$
$$\overline{\Gamma}_{\gamma f} = 2.26 \ \text{meV}, \ \overline{E}_{\gamma f} = 1.10 \ \text{meV}.$$

Shackleton also investigated the influence of the spin of the compound nucleus on the excitation energy of the fragments and found

$$\overline{\nu}_{n0}(3^-) - \overline{\nu}_{n0}(4^-) = (80 \pm 130) \ \text{keV};$$
$$\overline{E}'_{\gamma 0}(3^-) - \overline{E}'_{\gamma 0}(4^-) = (-140 \pm 140) \ \text{keV}.$$

From the values obtained for the difference effect, Shackleton concluded that for $^{235}U$ an influence of the fission channels is not revealed.

The results of Shackleton's work were frequently reconsidered,[24-26] both in connection with a more careful selection of the experimental material (the number of resonances taken into account in the analysis was changed as the values of their spins and widths were determined more accurately) and in connection with the use of different theoretical models and parameters to analyze the experimental data. For example, Trochon's review[26] gives results for $^{235}U$ generalized from several studies:

$$4^- \text{ resonances: } \langle \overline{\Gamma}_{\gamma f}\overline{E}_{\gamma f}\rangle = (1730 \pm 590) \ \text{eV}^2,$$
$$\overline{\Gamma}_{\gamma f} = (2{,}1 \pm 0{,}7) \ \text{meV},$$
$$\overline{E}_{\gamma f} = (800 \pm 50) \ \text{keV};$$
$$3^- \text{ resonances:}$$
$$\overline{\Gamma}_{\gamma f} = (4.7 \pm 2.3) \ \text{meV}, \ \overline{E}_{\gamma f} = (850 \pm 50) \ \text{keV}.$$

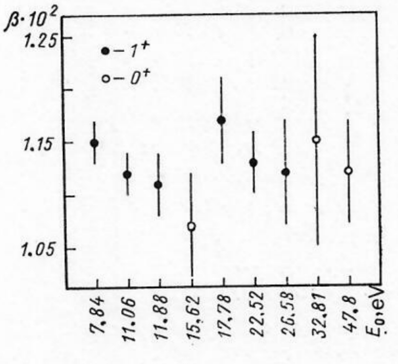


FIG. 13. Yields of triple $f\gamma\gamma$ coincidences for $^{239}$Pu resonances.[14]

### Results for $^{239}$Pu

For the interaction of $s$-wave neutrons with $^{239}$Pu nuclei, the resonances have spin and parity $1^+$ or $0^+$. Spin identification of levels, mainly by resonance scattering of neutrons, has been made for resonances in the energy region below 500 eV.[41] For the mean separation $\langle D \rangle \simeq 2.4$ eV between the levels of the two spins, it is as a rule fairly easy to measure the various effects at isolated $1^+$ resonances $[\langle \Gamma_f(1^-) \rangle \simeq 34$ meV]. The procedure is somewhat more complicated in the case of the $0^+$ resonances $[\langle \Gamma_f(0^+) \rangle \simeq 2.2$ eV], for which it is necessary to introduce corrections for the contributions from neighboring levels.

In the study of Ref. 14 made at Dubna by the same method as in Ref. 13, an attempt was made to detect experimentally the $(n, \gamma f)$ reaction in the case of $^{239}$Pu fission by resonance neutrons in the region 7–48 eV. The results of these measurements are shown in Fig. 13. For the two spin states, the relative difference of the yields of triple $f\gamma\gamma$ coincidences was found to be

$$\eta_{\rm exp} = \frac{\bar{\beta}(1^+) - \bar{\beta}(0^+)}{\bar{\beta}(0^+)} = (2{,}82 \pm 1{,}97) \cdot 10^{-2}.$$

Under the assumption that one $\gamma$ ray from the $(n, \gamma f)$ reaction was detected within the range 0.8–2.5 MeV of the experimental limits (thresholds) for $\gamma$-ray detection, the difference between the widths of this reaction for the two spin states was estimated from the values of $\eta_{\rm exp}$ to be

$$|\bar{\Gamma}_{\gamma f}(1^+) - \bar{\Gamma}_{\gamma f}(0^+)| < 4 \text{ meV}.$$

This estimate of the widths, as in the case of the $^{235}$U nucleus,[13] agrees with Lynn's theoretical estimates.[2] A very interesting and unexpected result is the fact that the experimental data obtained in this study in the different regime for which $f\gamma\gamma$ coincidences were detected at a total energy of the $\gamma$ rays above 2.5 MeV agreed well with the data of the first regime. On the basis of this, it was concluded by the authors of Ref. 14 that at the statistical accuracy which they achieved the $(n, \gamma f)$ reaction on $^{239}$Pu is not observed.

In the investigation of Refs. 15 and 16, Ryabov *et al.* used the electron linac at Saclay to measure the variations in the multiplicity of the $\gamma$ rays from $^{239}$Pu fission in the resonance region 0.2–200 eV. The results of these measurements are shown in Fig. 14. The most interesting feature of this experiment was the detection of appreciable variations of the multiplicity from resonance to resonance, which were especially large for the $1^+$ levels (reaching $14 \pm 3\%$ at $E_0 = 44.5$ eV). It was also found that these variations in the $\gamma$-ray multiplicity were strongly correlated with the variations in the neutron multiplicity $\nu_n$ and the total energy $E'_\gamma$ of the fission $\gamma$ rays measured by Shackleton *et al.*[20] and, in addition, that the values of $\nu_\gamma$, $\nu_n$, and $E'_\gamma$ were correlated with the reciprocal fission width $\Gamma_f^{-1}$ of the resonances. For 16 measured $1^+$ resonances, the correlation coefficient for the $\gamma$-ray multiplicity and $\Gamma_f^{-1}$ was $r \simeq 0.82$ at the significance level $Q_r < 0.001$.[23] The analysis made in Ref. 15 of the experimental data of Refs. 15 and 20 made it possible to conclude that the $(n, \gamma f)$ reaction is observed in the fission of $^{239}$Pu by resonance neutrons. In the case of the $0^+$ resonances, as for the $^{239}$U resonances, the reaction is masked by direct fission.

The experimental data on the neutron multiplicity $\nu_n$ of Ref. 20 were used in Refs. 15 and 16 to estimate the width of the $(n, \gamma f)$ reaction for $1^+$ resonances of $^{239}$Pu. For this, the mean energy $\bar{E}_{\gamma f}$ of the spectrum of prefission $\gamma$ rays was calculated in the approximation of a single-hump barrier. Only $E1$ transitions were taken into account in the calculation, and the level density was calculated in accordance with the Gilbert–Cameron formula. The results were[15]

$$\langle \bar{\Gamma}_{\gamma f} \bar{E}_{\gamma f} \rangle = (3230 \pm 700) \text{ eV}^2,$$

$$\bar{E}_{\gamma f} = (800 \pm 90) \text{ keV}, \quad \bar{\Gamma}_{\gamma f} = (4.1 \pm 0.9) \text{ meV}.$$

With allowance for only the resonances with large fission widths $\Gamma_f > 60$ meV [to eliminate the influence of the $(n, \gamma f)$

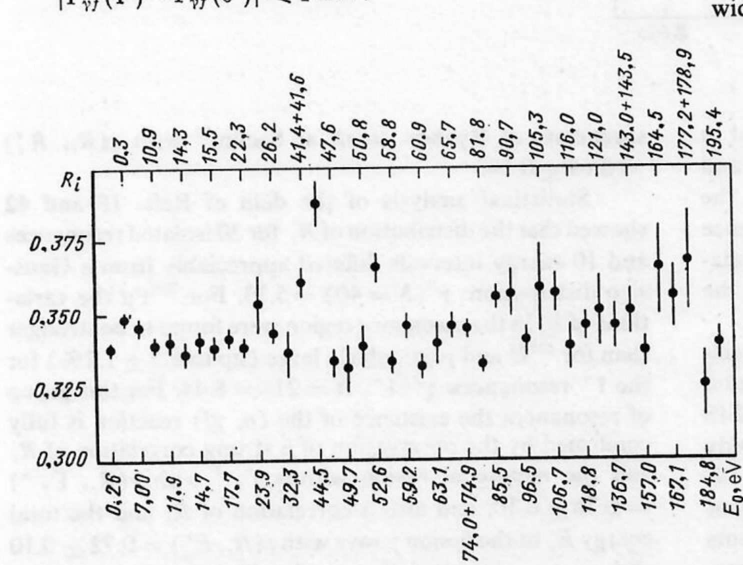


FIG. 14. Variations of the multiplicity of fission $\gamma$ rays of $^{239}$Pu (Ref. 15).

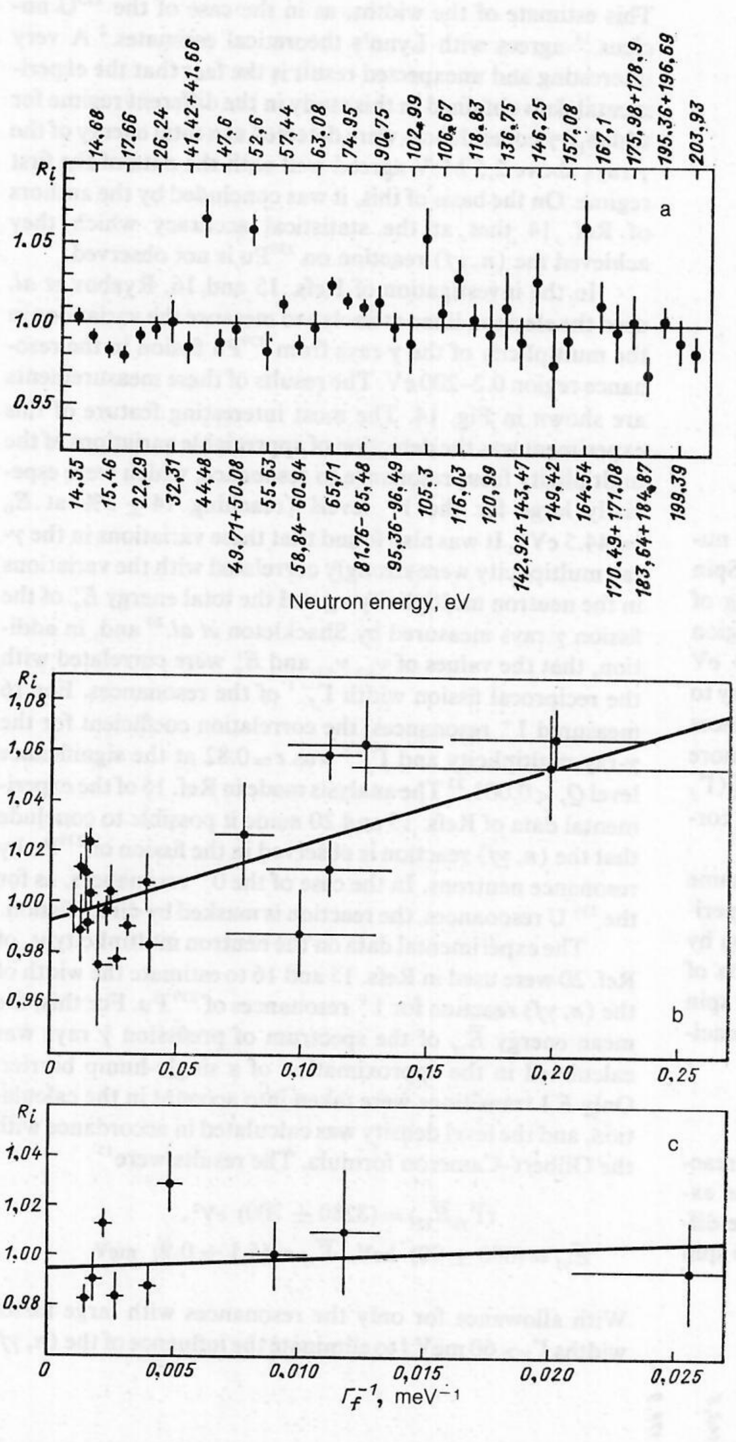


**FIG. 15. Multiplicity of fission $\gamma$ rays of $^{239}Pu$ (Refs. 18, 19, and 42): a) variations of the multiplicity; b) and c) correlations of the multiplicity and the reciprocal fission width, respectively, for $1^+$ and $0^+$ resonances.**

reaction] the mean $\gamma$-ray multiplicities were calculated in Ref. 15 for the two spin states: $\langle R_i(0^+)\rangle = 334.5 \pm 2.5$ and $\langle R_i(1^+)\rangle = 340.8 \pm 2.0$. The authors estimated that the probability of a purely statistical origin of such a difference was not more than 5%, indicating the presence of a correlation between the multiplicity of the fission $\gamma$ rays and the spin of the original compound nucleus (channel effect).

Figure 15 gives the results of measurements of the relative multiplicity $R_i$ of $\gamma$ rays from $^{239}Pu$ fissionin the region 14–206 eV (Refs. 18, 19, and 42), made using the GNEIS neutron spectrometer. Comparison of the data of this study with the results of other authors shows that despite the difference between the experimental methods good agreement between them exists. Thus, the results of the measurements of $R_i$ of this study agree with the data of the analogous measurements of Ryabov *et al.* at Saclay[15] with $r(R_i, R'_i) = 0.69 \pm 0.10$.

Statistical analysis of the data of Refs. 18 and 42 showed that the distribution of $R_i$ for 30 isolated resonances and 10 energy intervals differed appreciably from a Gaussian distribution: $\chi^2(N = 40) = 5.73$. For $^{239}Pu$ the variations of $R_i$ in the resonance region were found to be stronger than for $^{235}U$ and particularly large (up to $6.5 \pm 1.2\%$) for the $1^+$ resonances: $\chi^2(1^+, N = 21) = 8.44$. For this group of resonances the existence of the $(n, \gamma f)$ reaction is fully confirmed by the observation of a strong correlation of $R_i$ and the reciprocal fission widths $\Gamma_f^{-1}$ with $r(R_i, \Gamma_f^{-1}) = 0.76 \pm 0.10$, and also a correlation of $R_i$ and the total energy $E'_\gamma$ of the fission $\gamma$ rays with $r(R_i, E'_\gamma) = 0.72 \pm 0.10$ and an anticorrelation of $R_i$ and $\nu_n$ with

$r(R_i, \nu_n) = -0.60 \pm 0.14$ (data on $E'_\gamma$ and $\nu_n$ of Shackleton *et al.*[23]).

The dependence of $R_i$ on $\Gamma_f^{-1}$ was used in the study to estimate the width of the $(n, \gamma f)$ reaction for the $1^+$ and $0^+$ resonances of $^{239}$Pu (Ref. 42): $1^+$ resonances, $\overline{\Gamma}_{\gamma f} = 1.91 \pm 0.81$ meV; $0^+$ resonances, $\overline{\Gamma}_{\gamma f} = 2.8 \pm 9.2$ meV. Methods similar to those used for the $^{235}$U measurements were used to analyze the experimental data and make the theoretical calculations. The experimental and calculated values of the widths $\overline{\Gamma}_{\gamma f}$ for the $1^+$ resonances are given in Fig. 16. Comparison of the calculations and the experimental results showed that for the prefission $\gamma$ emission of $^{240}$Pu, in contrast to $^{236}$U, the $E$ 1 transitions are dominant, and the best agreement is obtained by using the model of a giant dipole resonance and the model of doorway states with intermediate damping of the states in the second well. It was also shown that there was no spin channel effect for the multiplicity of the $\gamma$ rays from $^{239}$Pu fission:

$$\overline{R}_{i0}\,(0^+) - \overline{R}_{i0}\,(1^+) = (0{,}0023 \pm 0{,}0090) \text{ rel. units}.$$

The results of measurements of the neutron multiplicity $\nu_n$ and the total energy $E'_\gamma$ of the fission $\gamma$ rays made by Weston and Todd[29] with the ORELA accelerator are shown in Fig. 17. Statistical analysis of the data for 29 isolated resonances and eight groups of resonances in the region 10–170 eV revealed a difference of the mean values $\langle E'_\gamma \rangle$ and $\langle \nu_n \rangle$ for the two spin states that was large for $E'_\gamma$ (2.5%) and slight for $\nu_n$ (0.5–0.8%). Such a difference agrees well with the estimate of it obtained from the balance of the excitation energy using the parameter $\partial \nu_n / \partial E^* = 0.128$ neutron/MeV.[51] The least-squares analysis of the dependence of $E'_\gamma$ and $\nu_n$ on $\Gamma_f^{-1}$ made in the study revealed the presence of a strong correlation of $E'_\gamma$ and $\Gamma_f^{-1}$, whereas such correlation was weaker for $\nu_n$. The observed correlations of $E'_\gamma$, $\nu_n$, and $\Gamma_f^{-1}$ were interpreted under the assumption of competition between the $(n, \gamma f)$ reaction and direct fission, the competition being most strongly manifested for resonances with small fission widths. In Ref. 29 the parameters of the $(n, \gamma f)$ reaction were not estimated.

The results of measurements of the neutron multiplicity $\nu_n$ and the total energy $E'_\gamma$ of the fission $\gamma$ rays, made with the electron linac at Saclay using a large liquid scintillation detector, were published in Refs. 20–26. In Ref. 20, variations

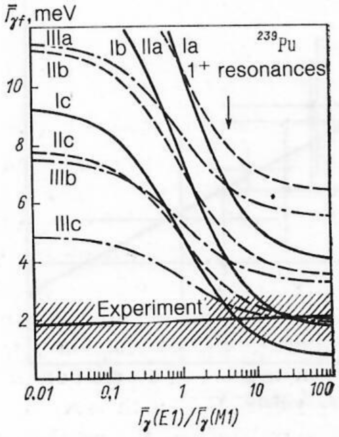


FIG. 16. Experimental and calculated widths $\overline{\Gamma}_{\gamma f}$ for $1^+$ resonances of the $^{239}$Pu nucleus.[42] The notation is the same as in Fig. 9.

of $E'_\gamma$ in the resonance region ($E_n < 110$ eV) were measured for the first time. Analysis of data for 29 resonances of $^{239}$Pu revealed the presence of a strong anticorrelation of the $\nu_n$ and $E'_\gamma$ variations. In accordance with the Birge criterion, the probability of a purely statistical origin of such variations of $\nu_n$ and $E'_\gamma$ for the $1^+$ resonances does not exceed 0.000 05, whereas for the $0^+$ resonances it is 3 and 64%, respectively. These results were interpreted in Ref. 20 under the assumption of the existence of the $(n, \gamma f)$ reaction on the $^{239}$Pu nucleus this being most strongly manifested in the case of $1^+$ resonances.

In Refs. 21, 22, 24, and 25, the results of measurements of $\nu_n$ and $E'_\gamma$ in an extended energy range ($E_n < 400$ eV) were discussed, and preliminary estimates of the product $\langle \overline{\Gamma}_{\gamma f} \overline{E}_{\gamma f} \rangle$ were also made. As for $^{235}$U, the results of the measurements of $\nu_n$ and $E'_\gamma$ for $^{239}$Pu are contained fully in Shackleton's thesis.[23] Figure 18 gives data of this study. Statistical analysis of the results of the measurements of $\nu_n$ and $E'_\gamma$ for the $^{239}$Pu resonances in the energy range $7 < E_n < 195$ eV confirmed the conclusions that had been drawn by the same author on the restricted statistical material in Ref. 20.

For the $1^+$ resonances, the measured values of $\nu_n$ and $E'_\gamma$ exhibit, in complete agreement with the hypothesis of competition between the $(n, \gamma f)$ reaction and direct fission, a strong dependence on the reciprocal fission width $\Gamma_f^{-1}$ of the resonances: $r(E'_\gamma, \Gamma_f^{-1}) = 0.94$ and $r(\nu_n, \Gamma_f^{+1}) = -0.87$. The least-squares analysis of the linear dependence of $\nu_n$ and $E'_\gamma$ on $\Gamma_f^{-1}$ made in Ref. 23 gave the results

$$1^+ \text{ resonances}: \langle \overline{\Gamma}_{\gamma f} \overline{E}_{\gamma f} \rangle = \begin{cases} (4800 \pm 530)\ \text{eV}^2\ (\nu_n); \\ (4490 \pm 400)\ \text{eV}^2\ (E'_\gamma); \end{cases}$$

$$0^+ \text{ resonances}: \langle \overline{\Gamma}_{\gamma f} \overline{E}_{\gamma f} \rangle = \begin{cases} (5000 \pm 7000)\ \text{eV}^2\ (\nu_n); \\ (8000 \pm 1900)\ \text{eV}^2\ (E'_\gamma). \end{cases}$$

Shackleton made theoretical estimates of the product $\langle \overline{\Gamma}_{\gamma f} \overline{E}_{\gamma f} \rangle$ and its factors for $^{239}$Pu under the same assumptions as for $^{235}$U. The best agreement with the experimental results was obtained for the following calculated values:

$$1^+ \text{ resonances}: \langle \overline{\Gamma}_{\gamma f} \overline{E}_{\gamma f} \rangle = 3760\ \text{eV}^2,$$

$$\overline{\Gamma}_{\gamma f} = 2.76\ \text{meV},\ \overline{E}_{\gamma f} = 1.36\ \text{MeV};$$

$$0^+ \text{ resonances}: \langle \overline{\Gamma}_{\gamma f} \overline{E}_{\gamma f} \rangle = 7900\ \text{eV}^2,$$

$$\overline{\Gamma}_{\gamma f} = 5.73\ \text{meV},\ \overline{E}_{\gamma f} = 1.38\ \text{MeV}.$$

For $^{239}$Pu, Shackleton[23] found a weak fission-channel effect: $\overline{\nu}_{n0}(0^+) - \nu_{n0}(1^+) = 99 \pm 42$ keV. $\overline{E}'_{\gamma 0}(0^+) - \overline{E}'_{\gamma 0}(1^+) = 10 \pm 10$ keV, from which $E_f^*(0^+) - E_f^*(1^+) = 109 \pm 43$ keV. If the "true" difference between the excitation energies $E_f^*$ of the fragments for the $0^+$ and $1^+$ resonances is zero, then in accordance with Shackleton's estimate the probability of chance observation of the observed difference does not exceed 1%.

In the review of Ref. 26 the experimental results of Ref. 23 for $^{239}$Pu were analyzed with allowance for the data on the fission barriers of Goldstone *et al.*[52] obtained from measurements of the cross section of the $^{239}$Pu$(d, pf)$ reaction. The estimates of the parameters of the $(n, \gamma f)$ reaction made in Ref. 26 differ somewhat from the estimates of Shackleton:

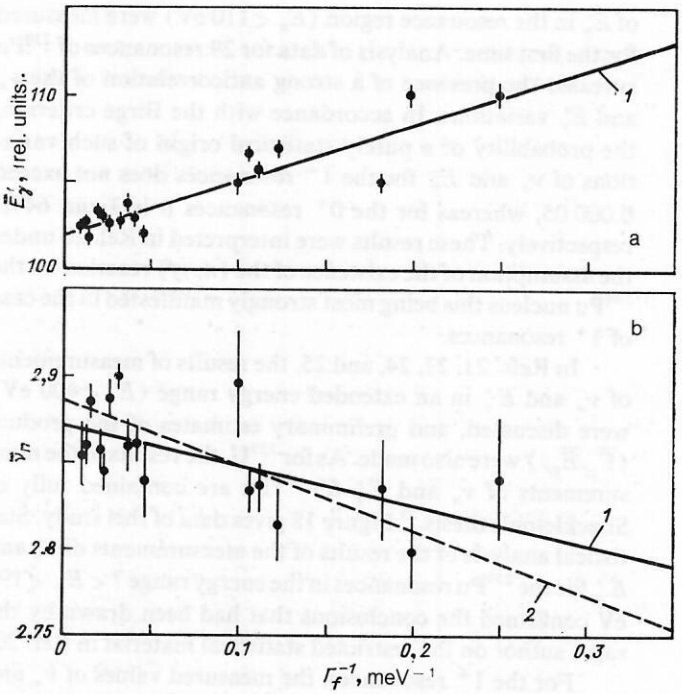


FIG. 17. Correlations of (a) the total energy $E'_\gamma$ of fission $\gamma$ rays and (b) the multiplicity $\nu_n$ of fission neutrons with the reciprocal fission widths of $1^+$ resonances of $^{239}$Pu (Ref. 29): 1) least-squares analysis; 2) least-squares analysis for weighted points.

1$^+$ resonances: $\langle \overline{\Gamma_{\gamma f} E_{\gamma f}} \rangle = (4600 \pm 300)$ eV$^2$,

$$\Gamma_{\gamma f} = (4.2 \pm 0.4) \text{ meV},$$

$$\overline{E}_{\gamma f} = (1080 \pm 50) \text{ keV};$$

The result of an experiment to measure the spectrum of prefission $\gamma$ rays from the $(n, \gamma f)$ reaction performed made by Trochon *et al.*[40,41] at Saclay is shown in Fig. 19. The spectrum shown here is the difference of the two instrumental spectra (normalized to one fission event) of the $\gamma$ rays for the $1^+$ resonances at 44.48 and 10.93 eV. The achieved statistical accuracy enabled Trochon *et al.* to assert that they had found a peak at an energy around 2 MeV (excitation energy 4.52 MeV). This peak corresponds to a vibrational state of

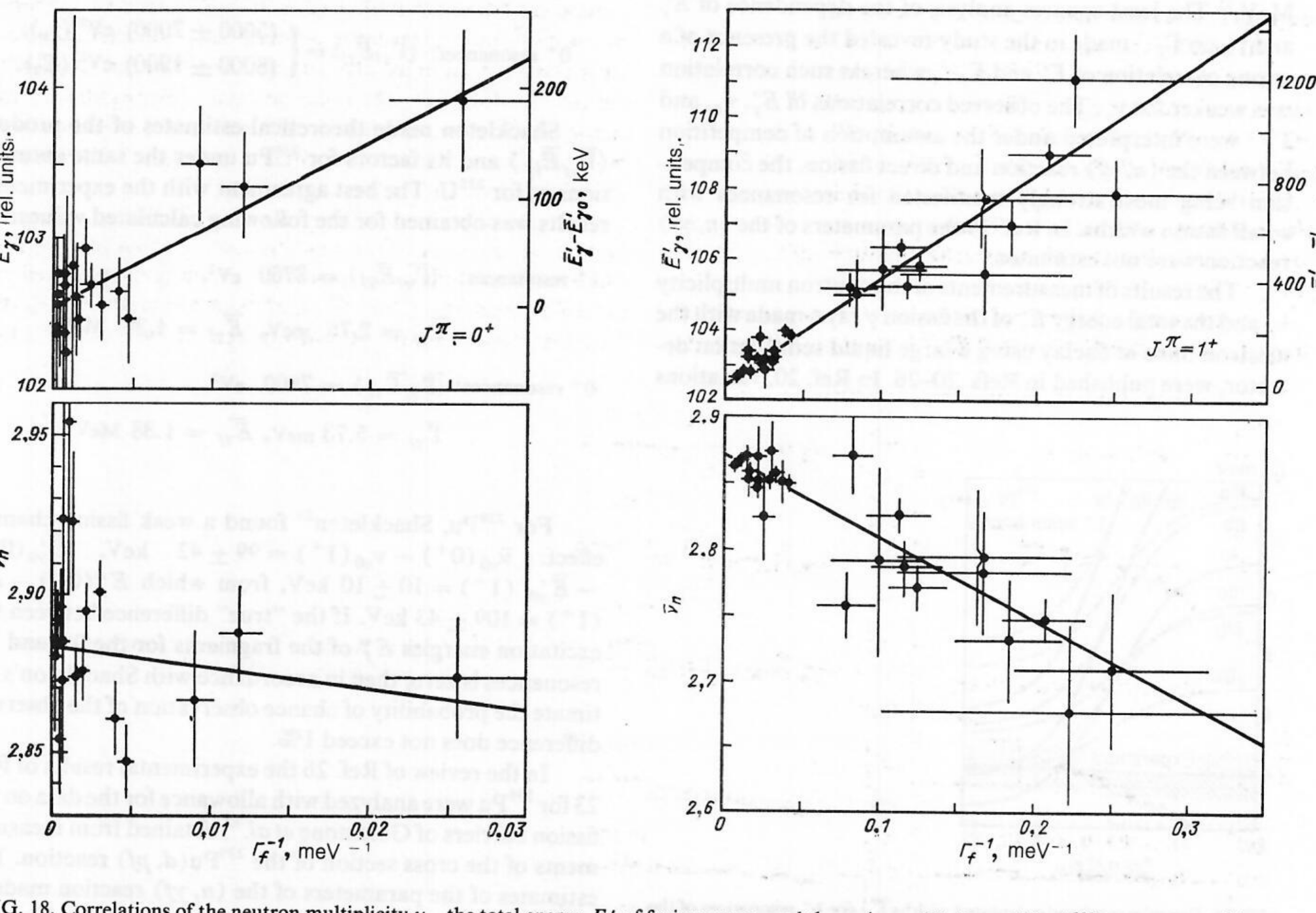


FIG. 18. Correlations of the neutron multiplicity $\nu_n$, the total energy $E'_\gamma$ of fission $\gamma$ rays, and the reciprocal fission width of $^{239}$Pu resonances.[23]

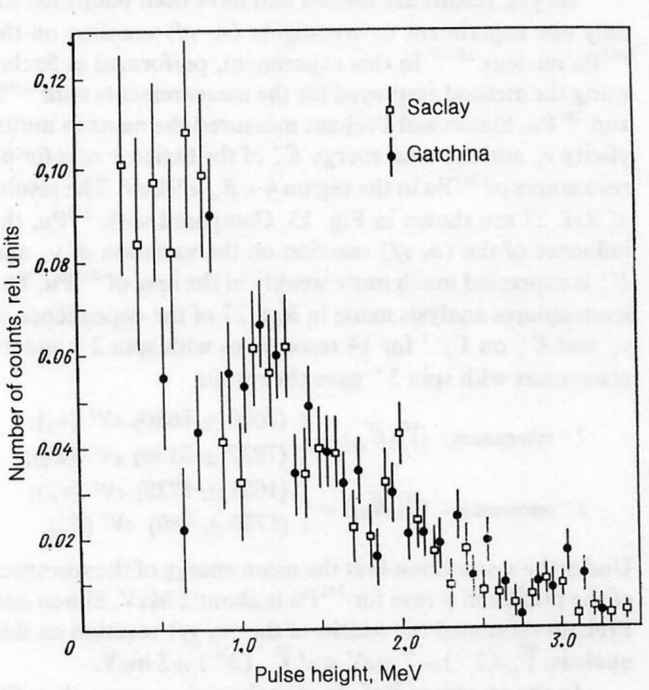


FIG. 19. Spectrum of prefission $\gamma$ rays of the $(n, \gamma f)$ reaction on $^{239}Pu$ according to data of measurements at Saclay[40,41] and with the GNEIS spectrometer at Gatchina.[42]

class II (in the second well of a two-hump fission barrier) with $K^{\pi}$ equal to $0^-$ or $1^-$. The observation of a resonance at the same excitation energy in the $(d, pf)$ reaction[52] can be regarded as a confirmation of this conclusion.

In a different experiment, performed with the GNEIS neutron spectrometer,[19,42,43] spectra of the fission $\gamma$ rays were measured for eight $^{239}Pu$ resonances: 90.75 eV ($R$ 1), 52.6 eV ($R$ 2), 47.6 eV ($R$ 3), 44.48 eV ($R$ 4), 41.66 eV + 41.42 eV ($R$ 5), 22.29 eV ($R$ 6), 17.66 eV ($R$ 7), and 10.93 eV ($R$ 8). Figure 20 shows the ratio of the $\gamma$-ray spectra for different combinations of these resonances. The ratios exhibit the following characteristic features of the pulse-height spectra of the $\gamma$ rays:

1) the spectra of the $\gamma$ rays from the $1^+$ resonances with large fission widths $\Gamma_f$ ($R$ 6, $R$ 7, $R$ 8) have similar shapes; the same can be said, admittedly with less statistical confidence, about the spectra from the $1^+$ resonances with small widths $\Gamma_f$ ($R$ 1, $R$ 2, $R$ 4, $R$ 5);

2) despite the difference of the spins, the spectrum of the $\gamma$ rays from the resonance $R\,3(0^+)$ is similar to the spectra of the group of weak $1^+$ resonances ($R\,1 + R\,2 + R\,4 + R\,5$):

3) on the average, the spectra of the weak $1^+$ resonances ($R$ 1, $R$ 2, $R$ 4, $R$ 5) differ appreciably in their shape from the spectra of the strong $1^+$ resonances ($R$ 6, $R$ 7, $R$ 8).

The first of these features of the spectra of the fission $\gamma$ rays supports the assumption used to analyze the data of experiments in which the multiplicity of the fission $\gamma$ rays was measured, namely, the spectrum of the $\gamma$ rays from the fragments varies little from resonance to resonance.

The difference pulse-height spectrum obtained in Ref. 42 for the $1^+$ resonances $R$ 4 (44.48 eV) and $R$ 8 (10.93 eV) is shown in Fig. 19. Comparison of the results of the two experiments shows that despite the difference between the response functions of the two detectors—NaI(Tl) and Ge(Li)—agreement of the data in the energy range $0.8 < E\gamma < 3.3$ MeV is observed. The results of the GNEIS measurements confirm the possible existence of the maximum at $E_\gamma \simeq 2$ MeV found at Saclay.

Comparison of the shapes of the measured and calculated spectra of the prefission $\gamma$ rays (Fig. 21) reveals the presence of a hard ($E_\gamma > 2$MeV) component, the origin of which can be explained by the manifestation in the $(n, \gamma f)$ reaction of transition states that were not detected in the $(d, pf)$ reaction, data on which were used in the calculations. For the resonance $R$ 4 and the other ($R$ 1, $R$ 2, $R$ 5) weak $1^+$ resonances of $^{239}Pu$ some structures found in the difference pulse-height spectra could be attributed to transitions to levels observed in the $(d, pf)$ reaction and corresponding to incompletely damped vibrational states in the second well of the $^{240}Pu$ fission barrier at excitation energies 1–3 MeV below the neutron binding energy.

In the experiment of Popeko *et al.*,[44] the spectrum of prefission $\gamma$ rays for $^{239}Pu$ was obtained by calculation from the measured difference spectrum of the conversion electrons for resonance (0.29 eV) and thermal neutrons. Figure 22 gives the spectra of $\gamma$ rays from the $(n, \gamma f)$ reaction (ob-

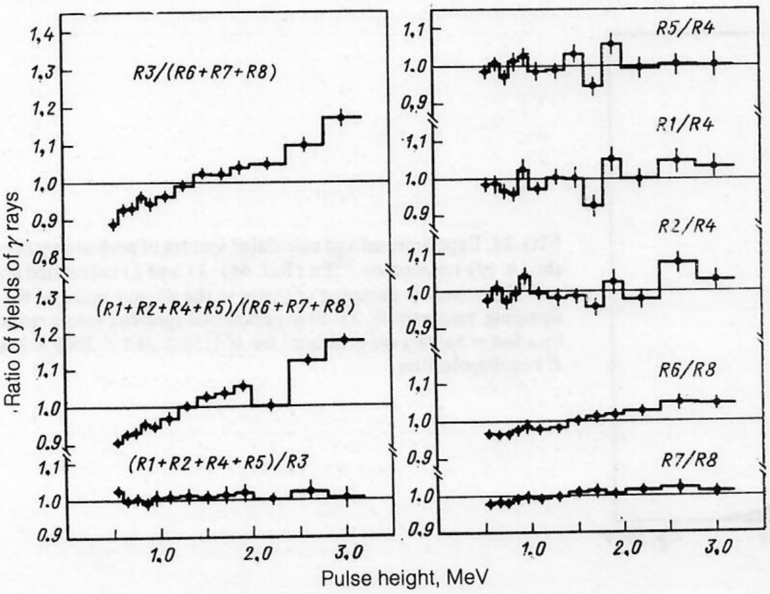


FIG. 20. Ratios of spectra of fission $\gamma$ rays for various combinations of $^{239}Pu$ resonances.[42,43]

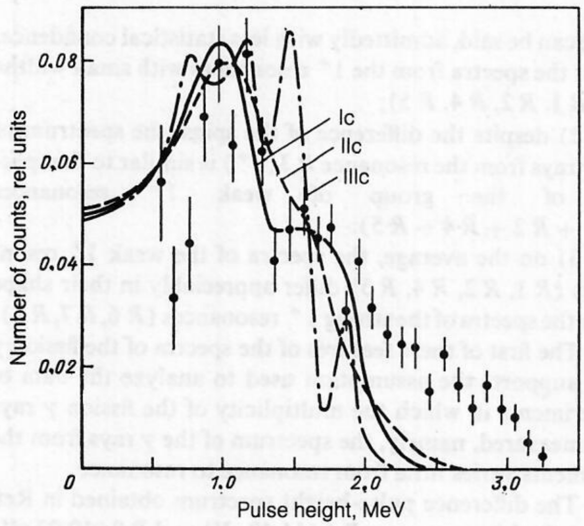


FIG. 21. Averaged difference ($R_{1,2,4,5} - R\,8$) spectrum of $\gamma$ rays, averaged over the $1^+$ resonances ($R\,1$, $R\,2$, $R\,4$, $R\,5$), and calculated pulse-height spectra of the prefission $\gamma$ rays of $^{239}$Pu (Refs. 42 and 43). The designation of the various calculations is the same as in Fig. 9.

tained under different assumptions about the multipolarity of the $\gamma$ transitions) and compares them with the theoretically calculated spectra of Ref. 40. It was found that the theoretical spectra are harder than the experimental spectra, and the best agreement is apparently obtained for $M\,1$ transitions or for transitions with an admixture of up to 50% of $E\,1$ transitions with intermediate damping of states in the second well.

### Results for $^{241}$Pu

The compound states formed when $s$-wave neutrons are captured by the $^{241}$Pu nucleus have spin and parity $2^+$ or $3^+$. The spin values of the resonances were obtained by a multilevel analysis of the data of measurements of the cross sections $\sigma_{tot}$ and $\sigma_f$ in the region of energies below 104 eV.[53,54] Since the mean separation between the levels for both spins is $\langle D\rangle \simeq 1.14$ eV and the fission widths are $\langle\Gamma_f(3^+)\rangle \simeq 87$ meV and $\langle\Gamma_f(2^+)\rangle \simeq 595$ meV, it is necessary to introduce corrections for the contribution from the neighboring levels for measurements made at isolated resonances.

As yet, results are known and have been published for only one experiment to investigate $(n, \gamma f)$ reaction on the $^{241}$Pu nucleus.[26-28] In this experiment, performed at Saclay using the method employed for the measurements with $^{235}$U and $^{239}$Pu, Simon and Frehaut measured the neutron multiplicity $\nu_n$ and the total energy $E'_\gamma$ of the fission $\gamma$ rays for 64 resonances of $^{241}$Pu in the region $4 < E_n < 91$ eV. The results of Ref. 27 are shown in Fig. 23. Compared with $^{239}$Pu, the influence of the $(n, \gamma f)$ reaction on the variation of $\nu_n$ and $E'_\gamma$ is expressed much more weakly in the case of $^{241}$Pu. The least-squares analysis made in Ref. 27 of the dependence of $\nu_n$ and $E'_\gamma$ on $\Gamma_f^{-1}$ for 14 resonances with spin $2^+$ and 19 resonances with spin $3^+$ gave the results

$$2^+ \text{ resonances: } \langle\overline{\Gamma}_{\gamma f}\overline{E}_{\gamma f}\rangle = \begin{cases} (5830 \pm 5680)\ \text{eV}^2\ (\nu_n); \\ (7932 \pm 5158)\ \text{eV}^2\ (E'_\gamma); \end{cases}$$

$$3^+ \text{ resonances: } \langle\overline{\Gamma}_{\gamma f}\overline{E}_{\gamma f}\rangle = \begin{cases} (1627 \pm 1229)\ \text{eV}^2\ (\nu_n); \\ (1750 \pm 789)\ \text{eV}^2\ (E'_\gamma). \end{cases}$$

Under the assumption that the mean energy of the spectrum of the prefission $\gamma$ rays for $^{241}$Pu is about 1 MeV, Simon and Frehaut estimated the widths of the $(n, \gamma f)$ reaction on this nucleus: $\overline{\Gamma}_{\gamma f}(2^+) \simeq 7$ meV and $\overline{\Gamma}_{\gamma f}(3^+) \simeq 2$ meV.

In the review of Ref. 26, Trochon gives somewhat different estimates of the parameters $\overline{\Gamma}_{\gamma f}$ and $\overline{E}_{\gamma f}$ for $^{241}$Pu, made on the basis of the experimental data of Ref. 27 and Goldstone's data[52] on the fission barriers:

$$2^+ \text{ resonances: } \overline{\Gamma}_{\gamma f} = (8.7 \pm 4.9)\ \text{meV},$$
$$\overline{E}_{\gamma f} = (800 \pm 100)\ \text{keV};$$
$$3^+ \text{ resonances: } \overline{\Gamma}_{\gamma f} = (2.1 \pm 0.8)\ \text{meV},$$
$$\overline{E}_{\gamma f} = (800 \pm 100)\ \text{keV}.$$

## 5. DEVELOPMENT OF METHODS TO ESTIMATE NUCLEAR DATA FOR THE $(n, \gamma f)$ REACTION

As follows from the foregoing review, experimental investigations of the $(n, \gamma f)$ reaction involve great methodological difficulties. This is the main reason for the rather limited amount of experimental data hitherto accumulated for this reaction (there are data for only the three fissioning nuclei $^{235}$U, $^{239}$Pu, and $^{241}$Pu). At the same time, as the data have appeared, the need to take into account the $(n, \gamma f)$ reaction to calculate and estimate the neutron cross sections

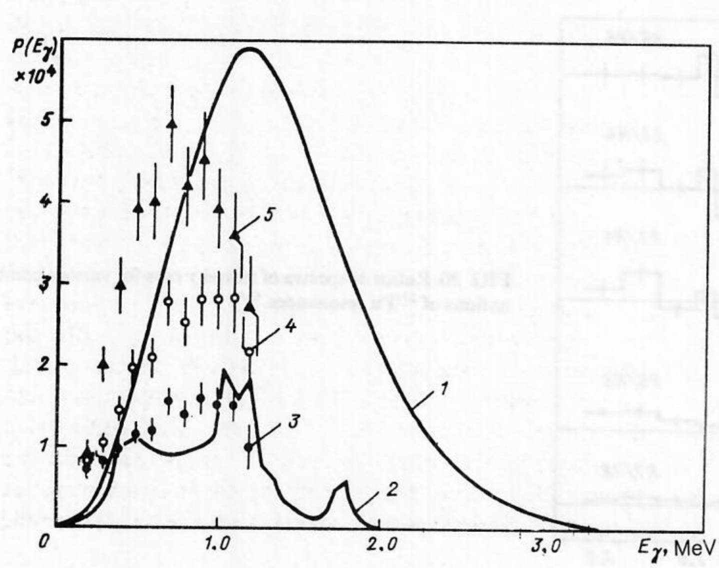


FIG. 22. Experimental and calculated spectra of prefission $\gamma$ rays of the $(n, \gamma f)$ reaction on $^{239}$Pu (Ref. 44): 1) and 2) calculated spectra[40] for complete damping of states in the second well and for no damping, respectively; 3)–5) experimental spectra (values reduced by a factor 5 along the ordinate) for $M\,1$, 50% $M\,1$ + 50% $E\,1$, and $E\,1$ multipolarities

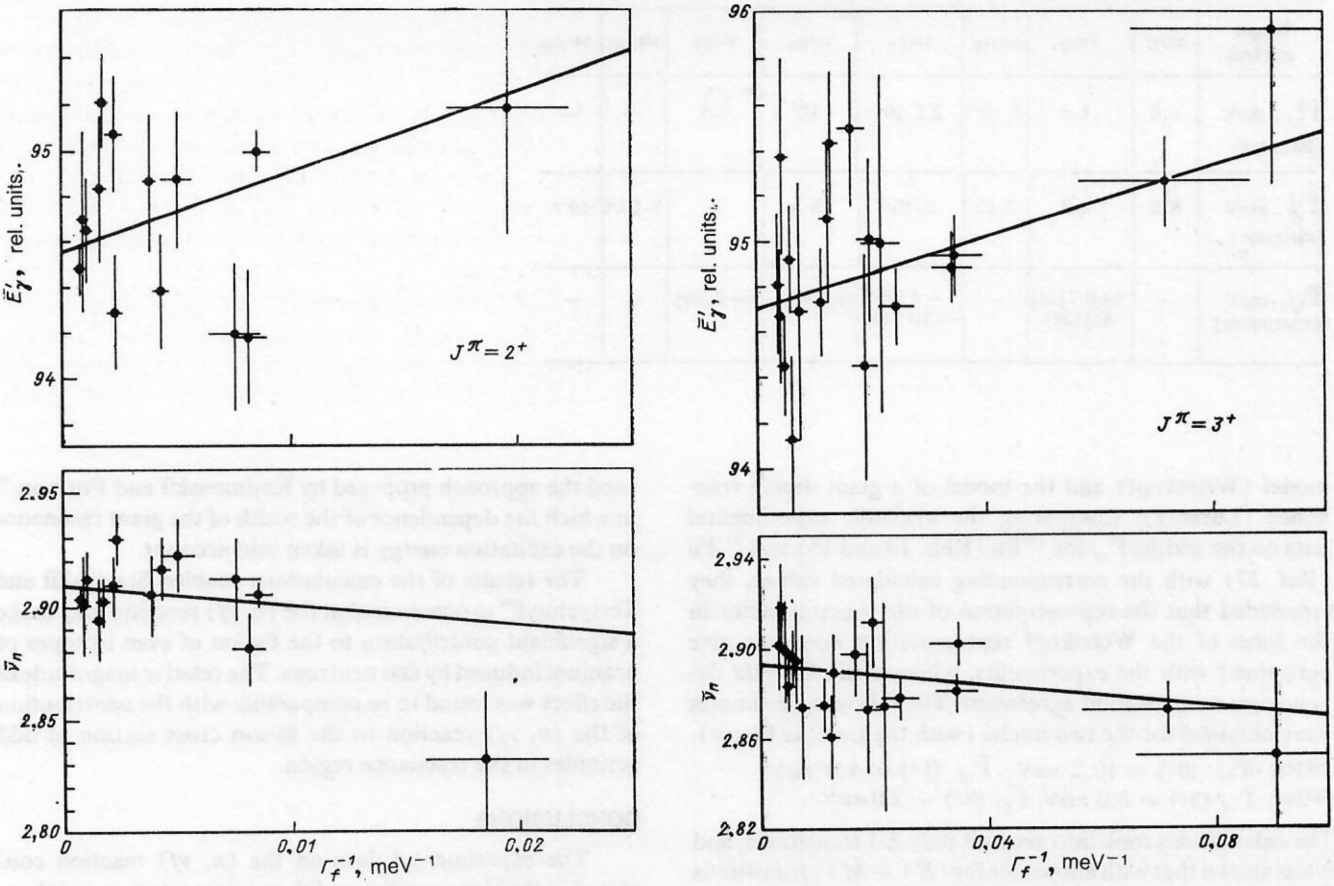


FIG. 23. Correlations of the neutron multiplicity $\nu_n$, the total energy $E'_\gamma$ of fission $\gamma$ rays, and the reciprocal fission width of $^{241}$Pu resonances.[27]

of fissioning nuclei in a wide range of energies of the incident neutrons has become quite obvious. For these reasons, and also as the methods of the theory of nuclear reactions have been improved, there have been a number of publications in which the experimental data have been analyzed and used to make new estimates and calculations of the parameters of the $(n, \gamma f)$ reaction for a number of fissioning nuclei.

A method for calculation of the widths $\overline{\Gamma}_{\gamma f}$ based on description of the mean probabilities of $\gamma$ transitions between highly excited compound states by means of the radiative strength function was developed by Vtyurin and Popov.[55,56] From the experimental data on the $(n, \gamma f)$ reaction they obtained radiative strength functions for $^{235}$U, $^{239}$Pu, and $^{241}$Pu under the assumption that $M1$ transitions are dominant among the soft ($E_\gamma < 2$ MeV) $\gamma$ transitions between the compound states. The following experimental facts provide the basis for this assumption:

1) analysis of the data on the $(n, \gamma f)$ reaction on the $^{235}$U nucleus[39] leads to the conclusion that the $M1$ transitions are dominant:

2) the shape of the spectrum of the $\alpha$ particles from the $(n, \gamma\alpha)$ reaction on nuclei with $A \sim 150$ can be best described under the assumption that the $M1$ transitions are dominant;

3) comparison of the widths $\Gamma_{\gamma\alpha}$ for the $^{143}$Nd resonances with different spins indicates a dominant contribution of the $M1$ transitions.

It was found that the strength function of the soft $\gamma$ transitions obtained from the experimental data on the $(n, \gamma f)$ and $(n, \gamma\alpha)$ reactions agreed, to within the errors with the radiative strength function of hard $M1$ transitions and does not exhibit a dependence on the atomic weight of the nucleus. On the basis of this fact and a strength function $S_\gamma(M1) = 2.4 \times 10^{-8}$, the widths $\sim\Gamma_{\gamma f}$ were calculated for a large number of fissioning nuclei (Table II). The calculations were made in the model of a two-hump fission barrier, with barrier parameters chosen from the well-known experimental data of Back *et al.*[57] on the $(d, pf)$ reaction. The values $\overline{\Gamma}^{\rm I}_{\gamma f}$ in Table II correspond to a "prompt" $(n, \gamma f)$ reaction, in which fission proceeds through both barriers after emission of a $\gamma$ ray, while the values $\overline{\Gamma}^{\rm II}_{\gamma f}$ correspond to a "hindered" reaction, in which the nucleus, being in one of the states of the second well of the fission barrier, fissions with the half-life of the corresponding isomer state. Of the experimental data given above, only the results of Refs. 38 and 39 can be included among measurements of a prompt $(n, \gamma f)$ reaction. Attention should be drawn to the satisfactory agreement of the estimates made in Refs. 55 and 56 with the experimental data at the disposal of the authors.

In Refs. 60–63, Kon'shin *et al.* analyzed the influence of the $(n, \gamma f)$ reaction in calculations of the mean widths and cross sections of radiative capture and fission for nuclei with a negative fission threshold. In particular, they showed that the results of calculations of the radiative width with allowance for the $(n, \gamma f)$ reaction differ appreciably for the two representations of the spectral factor (transition matrix elements) obtained in accordance with the single-particle

TABLE II. Widths of the $(n, \gamma f)$ reaction.[56] (References are given in square brackets.)

| Target nucleus | $^{233}$U | $^{235}$U | $^{237}$Np | $^{238}$Pu | $^{239}$Pu | $^{241}$Pu | $^{241}$Am | $^{243}$Am |
|---|---|---|---|---|---|---|---|---|
| $\overline{\Gamma}^{I}_{\gamma f}$, meV (estimate) | 4,6 | 1.6 | $2\cdot10^{-7}$ | $3.2\cdot10^{-3}$ | 12 | 1.4 | — | — |
| $\overline{\Gamma}^{II}_{\gamma f}$, meV (estimate) | 8.3 | 3.2 | 0,15 | $2\cdot10^{-2}$ | 16.5 | 2 | $5\cdot10^{-6}$ | $10^{-4}$ |
| $\overline{\Gamma}_{\gamma f}$, meV (experiment) | — | $2\pm0.7$ [24] $4^{+4}_{-2}$ [58] | — | $\sim 1$ [4] $< 10^{-1}$ [5] | $10\pm3$ [24] | $4\pm2$ [59] | — | — |

model (Weisskopf) and the model of a giant dipole resonance (Lorentz). Comparing the available experimental data on the widths $\overline{\Gamma}_{\gamma f}$ for $^{239}$Pu (Refs. 14 and 15) and $^{241}$Pu (Ref. 27) with the corresponding calculated values, they concluded that the representation of the spectral factor in the form of the Weisskopf representation does not give agreement with the experiments, whereas the Lorentz dependence ensures good agreement. The following estimates were obtained for the two nuclei (with the Lorentz factor):

$^{239}$Pu: $\overline{\Gamma}_{\gamma f}\ (0^+) = 10{,}2$ meV, $\overline{\Gamma}_{\gamma f}\ (1^+) = 4{,}9$ meV;
$^{241}$Pu: $\overline{\Gamma}_{\gamma f}\ (2^+) = 5{,}0$ meV, $\overline{\Gamma}_{\gamma f}\ (3^+) = 2{,}9$ meV.

The calculations took into account only $E\,1$ transitions, and it was shown that with allowance for $(E\,1 + M\,1)$ transitions ($E\,1/M\,1 \simeq 6.8$) the agreement with the experiments was somewhat poorer while an increase in the contribution of the $M\,1$ transitions ($E\,1/M\,1 \to 1$) only makes the agreement stil worse.

Allowance for the $(n, \gamma f)$ reaction, and also for the $(n, \gamma n')$ reaction, leads to a change in the energy dependence of the mean radiative widths and the cross section for radiative capture. Thus, for $^{239}$Pu (Fig. 24) and neutron energy $E_n \simeq 1$ MeV the results of calculation of $\sigma_\gamma$ with and without allowance for the $(n, \gamma f)$ reaction differ by almost a factor 2. At low energies, the $(n, \gamma f)$ reaction can make a significant contribution to the total fission cross section $\sigma_f = \sigma_{fd} + \sigma_{\gamma f}$. The calculation for $^{239}$Pu shows that at $E_n = 1$ keV the contribution of the $(n, \gamma f)$ reaction to $\sigma_f$ is about 10%. Allowance for this reaction also has an important influence on $\alpha = \sigma_\gamma / \sigma_f$. Thus, calculations of $\alpha(^{239}\text{Pu})$ with and without allowance for the $(n, \gamma f)$ reaction differ by 15% for $E_n \simeq 1$ keV, by 10% for 40 keV, by 20% for 300 keV, and by 50% for 700 keV.

In Ref. 64, Stavinskiĭ and Tertychnyĭ estimated the contribution of the $(n, \gamma f)$ reaction to the fission cross section of the even uranium isotopes with $A$ equal to 232, 234, 236, and 238 in the region of neutron energies $1.5 < E_n < 5.5$ MeV, which correspond to the first "plateau" of the fission cross section. They used a simple version of the statistical theory of nuclear reactions. At the same time, allowance was made for only the primary $E1$ transitions from the initial excited state of the nucleus, and Dilg's parametrization[65] was used to calculate the level density. The dependence of the results of the calculation on the form of the radiative strength function was investigated. Besides the widely used models of Weisskopf and of a giant dipole resonance, the authors also used the approach proposed by Kadmenskiĭ and Furman,[66] in which the dependence of the width of the giant resonance on the excitation energy is taken into account.

The results of the calculations enabled Stavinskiĭ and Tertychnyĭ[64] to conclude that the $(n, \gamma f)$ reaction may make a significant contribution to the fission of even isotopes of uranium induced by fast neutrons. The relative magnitude of the effect was found to be comparable with the contribution of the $(n, \gamma f)$ reaction to the fission cross section of odd actinides in the resonance region.

## CONCLUSIONS

The experimental data on the $(n, \gamma f)$ reaction contained in the above studies and their interpretation and theoretical description permit a number of conclusions to be formulated.

First the existence of the $(n, \gamma f)$ reaction can now be regarded as an experimentally established fact. For the three studied nuclei—$^{235}$U, $^{239}$Pu, and $^{241}$Pu—the volume and nature of the experimental information differ widely. However, a common feature is that the effects observed in the

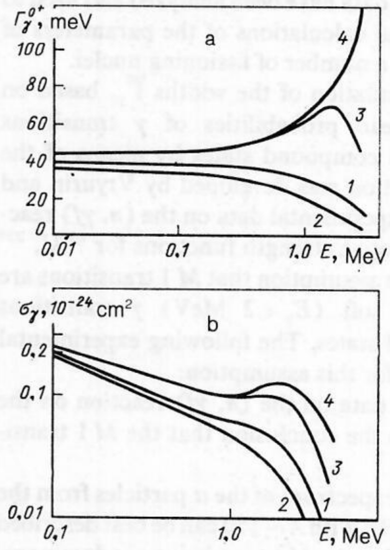


FIG. 24. Energy dependence of (a) the width $\overline{\Gamma}_\gamma(0^+)$ (b) the cross section $\sigma_\gamma$ for $^{239}$Pu with allowance for the $(n, \gamma f)$ and $(n, \gamma n')$ reactions (Ref. 62): 1) with allowance for $(n, \gamma f)$ and $(n, \gamma n')$, Lorentz; 2) with allowance for $(n, \gamma f)$ and $(n, \gamma n')$, Weisskopf; 3) with allowance for $(n, \gamma n')$, Lorentz; 4) without allowance for $(n, \gamma f)$ and $(n, \gamma n')$, Lorentz.

resonance region of energies that are associated with the multiplicity and total energy of the fission $\gamma$ rays, and also with the multiplicity of the secondary fission neutrons, can be interpreted from a unified point of view as a manifestation of the $(n, \gamma f)$ reaction in competition with direct fission. In this connection, it is important to make similar measurements for a larger number of nuclei, for example, for the uranium nuclei with $A$ equal to 233 and 236. As is evident from the estimates made in Ref. 56 (see Table II), the first of these must have a rather high probability for the $(n, \gamma f)$ reaction. With regard to $^{236}$U, it follows from measurements of the cross section for below-threshold fission of this nucleus in the region of energies below 415 eV (Ref. 67) that the fission width of the resonance fluctuates very weakly ($\nu_{\rm eff} \approx 18$) about the mean value 0.35 meV. One of the possible explanations[7] of this fact could be a dominant role of the $(n, \gamma f)$ reaction in the below-threshold fission of $^{236}$U.

Second, the widths $\Gamma_{\gamma f}$ obtained from the experimental data for the listed nuclei are known to an accuracy of 10–60%. The widths obtained by different authors using different methods may differ by up to several times. The errors in the widths $\overline{\Gamma}_{\gamma f}$ are largely determined by the experimental errors in the measured $\nu_\gamma$, $E'_\gamma$, and $\nu_n$, and also $\Gamma_f$. In addition, the spins of the resonances are not known sufficiently well. The errors $\Delta\overline{\Gamma}_{\gamma f}$ of this kind could in principle be reduced by increasing the number of measured resonances and improving the statistical accuracy. New experiments to identify the spins of the resonances like the measurements of Moore *et al.*[45] are also needed.

The results of measurements of the multiplicity of the fission neutrons are usually analyzed by means of the parameter $\partial\nu_n/\partial E^*$, obtained for the region of fast neutrons. Such linear extrapolation of the dependence $\nu_n(E^*)$ to the region of excitation energies below the neutron binding energy may also be a source of appreciable errors in the determination of $\overline{\Gamma}_{\gamma f}$. As the analysis of Ref. 68 showed, the region of excitation energies $4.5 < E^* < 5.5$ MeV, which contains the maximum of the spectrum of the prefission $\gamma$ rays, is a transitional region between two regions characterized by different fission types. For the superfluid region ($E^* \lesssim 4.5$ MeV) very weak damping is characteristic, and an increase in the excitation energy is expended almost entirely on an increase in the kinetic energy of the fission fragments. In the region with moderate and strong damping ($E^* \gtrsim 5.5$ MeV), the kinetic energy of the fragments decreases with increasing $E^*$, while the multiplicity of the fission neutrons increases. The behavior of the dependence $\nu_n(E^*)$ in the transition region (with a possible sharp growth with increase in $E^*$ and, accordingly, a larger value of $\partial\nu_n/\partial E^*$) is an uninvestigated phenomenon. Information about it would not only assist in solving the problem of the correct interpretation of the results of measurements of $\nu_n$ in the study of the $(n, \gamma f)$ reaction but would also make a significant contribution to our understanding of the fission mechanism itself.

A further source of error in $\overline{\Gamma}_{\gamma f}$ is the dependence of the results of the analysis of the experimental data on the adopted theoretical model. In this connection, there is a need for experimental information about the spectra of the prefission $\gamma$ rays, since from it one can obtain the mean energy $\overline{E}_{\gamma f}$ (or $\overline{\nu}_{\gamma f}$) and, hence,the value of $\overline{\Gamma}_{\gamma f}$ without recourse to any theoretical model. The first attempts at the experimental determination of the spectra of the prefission $\gamma$ rays demonstrated the extreme difficulty of such measurements due to the small observed effect, which is masked by the direct fission. For such measurements, detectors with a fairly high efficiency and moderate energy resolution, for example, NaI(Tl), would be most expedient. They could be used not only to determine the shape of the spectrum of the prefission $\gamma$ rays but also to investigate individiual $\gamma$ transitions to levels in the second well of the two-hump fission barrier.

Finally, as new data about the spins of the resonances, the probabilities of radiative transitions, fission barriers, and the other parameters used to analyze the experimental information become available it will be necessary to reexamine the results of the earlier investigations of the $(n, \gamma f)$ reaction.

It is a pleasant duty to thank G. A. Petrov for interesting discussions and constant interest in the work, Yu. P. Popov, who made a number of valuable comments about the data of the investigations of the $(n, \gamma f)$ reaction made by the present author, and also I. V. Maninen for assistance in the preparation of the publication.


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