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THE $(n, \gamma f)$ REACTION INDUCED BY SLOW NEUTRONS IN ^{239}Pu AND ^{235}U[1]

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The number of prompt neutrons and also the number and total energy of γ-rays emitted per fission for the $^{239}\mathrm{Pu}(n, f)$ and $^{235}\mathrm{U}(n, f)$ reactions in the resonance energy region have been measured. The experiments have been carried out at Saclay, using the 60 MeV linac as a pulsed neutron source.

The data show fluctuations of these quantities from one resonance to another, strongly correlated to the fission width Γ_f. An explanation is given in terms of the $(n, \gamma f)$ process. It depends on the competition between the γ-ray emission and the fission in the decay of the compound nucleus. As confirmed by the experimental results, this interpretation leads to a constant value of the product $\Gamma_{\gamma f}\,\bar{\varepsilon}_{\gamma f}$, for each spin state of a given nucleus, where $\Gamma_{\gamma f}$ is the width of the $(n, \gamma f)$ reaction and $\bar{\varepsilon}_{\gamma f}$ the average energy value of the prefission γ-ray spectrum.

A set of parameters has been determined such that the experimental results for both nuclei can be calculated from the same model. These calculations are discussed in terms of a single and a double-humped fission barrier.

When a compound nucleus is formed by the capture of a slow neutron, the compound nucleus can decay in one of three ways: by the elastic scattering, or re-emission of a neutron back into the entrance channel; by emitting a capture γ-ray, or, if energetically possible, by fission. It has been pointed out by J. E. Lynn [1] that it is possible for fission to occur subsequent to a low energy γ-ray emission. The experimental evidence for such a process, called $(n, \gamma f)$, has been obtained at Sacley from two experiments, both by the fission of ^{239}Pu induced by slow neutrons in the resonance region [2—5]. The same experiments were performed on ^{235}U, but the phenomenon is not so clear there [5].

In the experiments the time-of-flight technique was used. The 60 MeV electron linac was used as a pulsed neutron source. All observations were made for resonances in the energy range from 7 to 195 eV for ^{239}Pu and from 2 eV up to 58 eV for ^{235}U. In the first experiment, the mean number $\bar{\nu}$ of neutrons and the mean energy of all emitted γ-rays were detected in coincidence with the fission fragments.

The second experiment was a measurement of the variation of the γ-ray multiplicity in the same energy range.

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The results of both experiments on ^{239}Pu are shown in Fig. 1. As we can see, correlations from resonance appear between the three quantities discussed above and the fission width Γ_f.

These results may be interpreted in terms of the (n, γ, f) process as a means of deexciting the compound nuclear states of ^{240}Pu (see Fig. 2). That is, first the compound nucleus emits a γ-ray of such an energy that further deexcitation by fission is still possible. But the number of neutrons emitted by the fragments is small, since the excitation energy of the compound nucleus is lower than usual when it fissions. In contrast, the number and energy of γ-rays detected in coincidence with the fission fragments are both large since the γ-rays emitted by them, which are almost independent of the excitation energy of the compound nucleus, are added to the γ-rays emitted before fission.

In Figure 2 a schematic shape of the prefission γ-ray spectrum is also given. For the low energy prefission γ-rays, emitted near and above the barrier tops, the spectrum is continuous. When the primary γ-ray leaves the nucleus in a state of the first well at the same energy as the vibrational state of the second well (class II state) with the same spin and parity, the fission probability is enhanced. The prefission γ-ray spectrum must contain some peaks corresponding to the damped class II states as observed by Spetch et al. in a high resolution (d, pf) reaction experiment (their resolution was 17 keV/channel [18]).

Experimentally we observe a mixture of $(n, \gamma f)$ and direct fission reactions. When the total fission width Γ_f is small enough to be comparable with the width $\Gamma_{\gamma f}$ of the $(n, \gamma f)$ reaction, the latter process contributes significantly to the observed resonance. This explains the correlations with the total fission width Γ_f. If one calls $E_{\gamma 0}$ the energy of the γ-ray emitted by the fragment and E_γ the measured γ-ray energy, the relation between these values and the partial widths is:

$$E_\gamma = E_{\gamma 0} + \frac{\Gamma_{\gamma f}}{\Gamma_f}\,\overline{e_{\gamma f}} \tag{1}$$

where $\overline{e_{\gamma f}}$ is the average energy value of the prefission γ-rays. The quantities $\Gamma_{\gamma f}$ and $\overline{e_{\gamma f}}$ are constant from one resonance to another because they are characteristic of the $(n, \gamma f)$ reaction, which is a process with a great number of exit channels.

In a similar fashion, one may relate the neutron multiplicity to the same parameters:

$$\bar{E_\nu} = \bar{E}_{\nu 0} - \frac{\Gamma_{\gamma f}}{\Gamma_f}\,\overline{e_{\gamma f}} \tag{2}$$

where $\bar{E_\nu}$ and $\bar{E}_{\nu 0}$ are deduced from $\bar{\nu}$ and $\bar{\nu}0$ with the experimental relation:

$$\mathrm{d}\nu/\mathrm{d}\bar{E_\nu} = 0.13 \text{ neutron/MeV of the excitation energy [19]} \tag{3}$$

$\bar{\nu}0$ is the number of neutrons emitted by a direct fission.

From these expressions, one deduces:

$$(\overline{E_\gamma} - \overline{E_{\gamma 0}}) = -\,(\bar{E_\nu} - \bar{E}_{\nu 0}) = \frac{\Gamma_{\gamma f}\overline{e_{\gamma f}}}{\Gamma_f} = \frac{constant}{\Gamma_f} \tag{4}$$

The last equation suggests plots of $\bar{\nu}$ and $\overline{E_\gamma}$ versus the quantity $1/\Gamma_f$. As the $(n, \gamma f)$ process may be different for each of the two initial spin states of the compound nucleus, the resonance have been separated into two spin families according to spin determinations from scattering measurements [6]. The plot for the 1+ family shows a linear dependence as expected (Fig. 3). The straight lines of Fig. 3 are obtained from least squares fits to the $\bar{\nu}$ and $\overline{E_\gamma}$ data sets. The resulting values are given in Table 1.

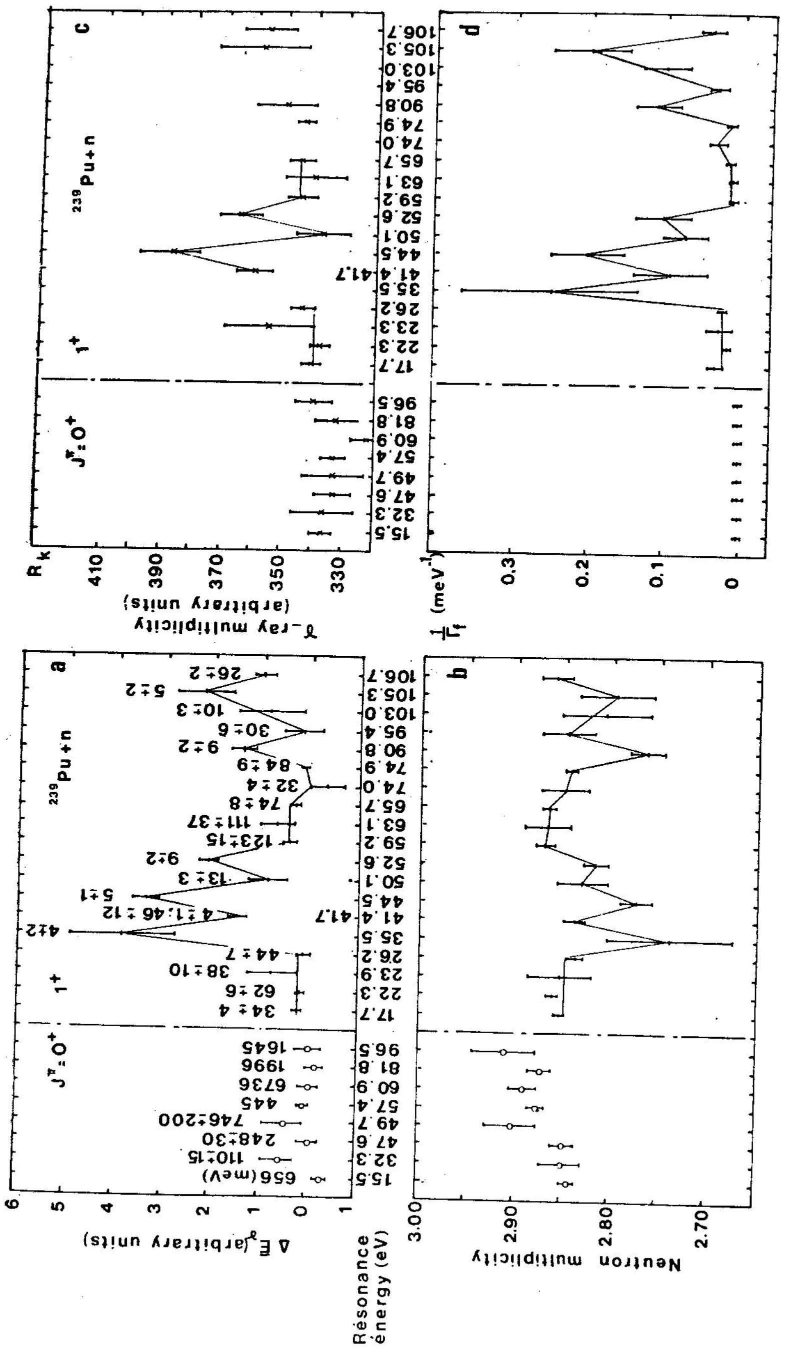


Fig. 1. $\bar{\nu}$, $\overline{E_\gamma}$, R_k (γ-ray multiplicity) and $1/\Gamma_f$ variations from one resonance to another [2, 3, 8]. For each resonance the fission width value is indicated near the $\overline{E_\gamma}$ experimental point. The straight lines between the experimental points have not physical significance. They only are to show the correlations.

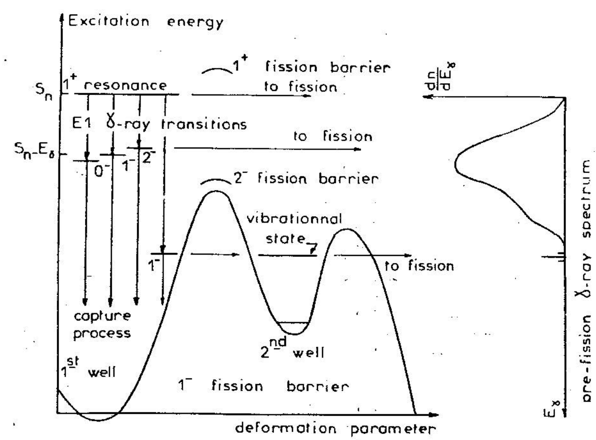


Fig. 2. Diagram of a 1^+ resonance deexcitation of the ^{240}Pu and the prefission γ-ray spectrum. Only the $E1$ γ-ray transitions are taken into account.

The same experiments have been performed on ^{235}U in the energy range of 2 to 58 eV. The fluctuations and correlations are much lower than in the ^{239}Pu data. The situation has been clarified by the recent spin assignements given by G. A. Keyworth from an experiment with a polarized neutron beam and a polarized target [8].

For 4^- resonances, the results given by D. Shackleton [9] are in Table 1. Unfortunately, the number of assigned 3^- resonances is too small to draw conclusions about this spin state.

An attempt has been performed to obtain the experimental values of the product $\Gamma_{\gamma f}\overline{e_{\gamma f}}$ by calculations using the fission barrier parameters deduced from other experiments.

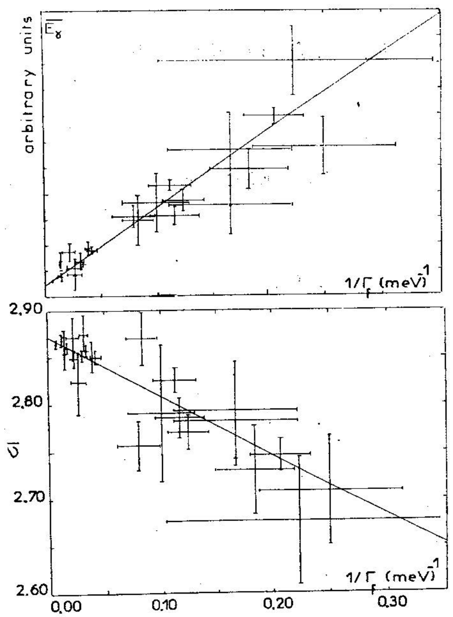
If after the emission of a γ-ray (energy E_γ) the compound nucleus is in a state of excitation energy $S_n - E_\gamma$ and spin and parity $J'\pi'$, the basic formula giving the width $\Gamma_{\gamma f}$ is:

$$\Gamma_{\gamma f} = K \sum_{J'} \int_0^U P(E_\gamma)\, \varrho\, (S_n - E_\gamma; J') \left\langle \frac{\Gamma_f(S_n - E_\gamma; J'\pi')}{\Gamma_f(S_n - E_\gamma; J'\pi') + \Gamma_\gamma(S_n - E_\gamma)} \right\rangle \mathrm{d}E_\gamma \qquad (5)$$

$P(E_\gamma)$ is the γ-ray emission probability, ϱ the level density, Γ_f the direct fission width and Γ_γ the capture width. K is a normalization coefficient. The integration is done up to the value of the $U = S_n$ − pairing gap energy. In fact the calculation includes the contribution of the electric and magnetic dipole transitions and the emission of one and two γ-rays before fission. The factor K is determined from the experimental value of the width Γ_γ at the neutron binding energy.

Since no experimental information was available, the calculations have been done with a probability of γ-ray emission proportional to E_γ^3, as determined by Blatt and Weisskopf [11] and also with one varying as a giant resonance shape [12] as proposed by Bollinger and Thomas [13]. We took $P(E_\gamma)E_1/P(E_\gamma)M_1 = 6.8$ as suggested by Bollinger [13]. The level density calculations were made with the Gilbert and Cameron formula.

Fig. 3. ^{240}Pu: $\bar{\nu}$ and $\overline{E_\gamma}$ variations versus $1/\Gamma_f$ for the 1^+ resonances. The Γ_f used values are given in Ref. [7].



The fission barrier transmission has been calculated, at first, with a single humped barrier as determined by Britt [14]. This determination preceded the Strutinski prescriptions. Other calculations with a double humped barrier have also been performed. The technique is the same as that used by Bondorf [15]. The damping in the second well is assumed to be increasing with the excitation energy, as proposed by Back [16]. The double humped fission barrier parameters are given by Back from (d, pf) measurements [17]. The results are summarized in Table 2.

The energies of the vibrational class II states are unknown for except maybe the 0^+ states, and the barrier penetrabilities depend sensitively on these locations, especially for the 1^- and 2^- transition states.

Table 1

Experimental values of the product $\Gamma_{\gamma f}\overline{e_{\gamma f}}$

		from $\bar{\nu}$ measurements	from E_γ measurements
^{240}Pu	1^+ resonances $\Gamma_{\gamma f} \cdot e_{\gamma f}$	4840 ± 630 eV^2	5170 ± 470 eV^2
	0^+ resonances $\Gamma_{\gamma f} \cdot \overline{e_{\gamma f}}$	5100 ± 7000 eV^2	9600 ± 2160 eV^2
^{236}U	4^- resonances $\Gamma_{\gamma f} \cdot \overline{e_{\gamma f}}$ ref. 8	1590 ± 710 eV^2	1370 ± 610 eV^2

Table 2

Calculated values of the product $\Gamma_{\gamma f}\overline{e_{\gamma f}}$ in eV. These results must be compared to the experimental values of Table 1

fission barrier type	^{240}Pu				^{236}U			
	E^3 law		giant res. law		E^3 law		giant res. law	
	0^+	1^+	0^+	1^+	3^-	4^-	3^-	4^-
1 hump ref. [14]	7368	5577	3675	3659	4383	1996	[illegible]	1004
2 humps ref. [17]	987	1334	390	730	1544	968	[illegible]	339
2 humps ref. [17] – 200 keV	2753	2747						
2 humps ref. [17] without damping	225	665						

In spite of these inaccuracies, the following conclusions can be drawn from these calculations. The $M1$ γ-ray contribution in the $(n, \gamma f)$ reaction from the 1^+ resonances of ^{240}Pu is important, if the ratio used for $P(E_\gamma)\ E1/P(E_\gamma)\ M1 = 6.8$ is correct [13]. The comparison of Tables 1 and 2 indicates that the double humped fission barrier parameters of ^{240}Pu as determined by B. Back do not allow sufficient penetrability, even though we reduce the barrier heights to their lower limits (lowered 200 keV) and in the most favourable case of the $P(E_\gamma)\alpha E^3$ law. Nevertheless, the $(n, \gamma f)$ reaction is a good probe to test the fission barrier because the number of fission exit channels which contributes strongly to the process is generally small and the initial spin and parity of the compound nucleus state can be exactly known. For instance, in the case of 0^+ resonances of ^{240}Pu, the fission after γ-ray emission occurs from 1^- exit channels. At these excitation energies the number of these channels is two.

An attempt to measure the prefission -ray spectrum of ^{240}Pu with a 5 keV resolution has been performed at Saclay under the same experimental conditions as the experiment above. The aim of this new experiment is to compare the experimental fission γ-ray energy spectra for resonances where the direct fission is the main deexcitation mechanism and for resonances where the $(n, \gamma f)$ process is dominant.

The experimental detector array consisted of four proton recoil liquid scintillators to detect fission events and a 60 cm^3 Ge-Li diode with a 5 keV resolution at 1 MeV to detect γ-rays. Only the γ-rays pulses in coincidence with the fission events are analyzed in the amplitude.

A complete analysis of these data is in progress. A prefission γ-ray spectrum has not yet been obtained, but no peaks seem to appear in the energy region corresponding to the class II states.

In conclusion, the first experiments have clearly shown the existence of the $(n, \gamma f)$ reaction in the deexcitation of the compound nuclei ^{240}Pu and ^{236}U. The experimental study of this process — when it is possible — appears to be a very sensitive probe to test fission barrier penetrabilities, especially if the prefission γ-ray spectrum can be determined.

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