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ANALYSIS OF THE EXPERIMENTAL RESULTS FOR ν PROMPT OF 239 Pu, 241 Pu, 235 U IN THE RESOLVED RANGE - COMPLEMENTARITY OF THE INTEGRAL AND MICROSCOPIC INFORMATIONS

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ABSTRACT

The fluctuations observed in the resolved resonance range in the ν prompt experimental values have been analysed in terms of spin and (n,γf) reaction effects. A good consistency has been found between the microscopic data and an extensive set of integral data. These fluctuations have been proven to have an impact in application.

INTRODUCTION

The average number of neutrons emitted per fission event ν is an important nuclear constant. Because of the stringent value of the requested accuracy (0.25 %) the evaluation of this important parameter for application should be as accurate as possible and for that reason should include as much basic physics as possible.

For the important fissile nuclei numerous measurements / 1 /, / 2 /, / 3 /, / 4 / have shown, long ago, the presence of fluctuations of more or less large amplitude associated with the resonances.

There has been long discussions about their origin.

For ^{239}Pu the interpretation by SHACKLETON / 5 / is now widely accepted. In this interpretation the fluctuations result from the competition between the direct fission process and a (n,γf) process on one side and from a spin effect on the other.

We propose a formalism, directly derived from the relationships established by SHACKLETON, to calculate a continuous curve $\nu_p(E)$ and we will show that the same formalism can be used also for ^{241}Pu and ^{235}U. In addition we will show that the fluctuations have an impact in application at least for ^{239}Pu and, consequently, cannot be ignored.

CALCULATIONNAL FORMALISM

To express the competition between the immediate fission and the (n,γf) process we have chosen to treat this last effect in a corrective term, as already done by SHACKLETON.

$$\bar{\nu}_p = \nu_i - \Delta_{n,\gamma f} \qquad (1)$$

This author gives for $\bar{\nu}_p$ corresponding to a given resonance of spin J, the following relationship.

$$\nu_p^J = \nu_i^J + \frac{\partial \nu^J}{\partial E} E - \frac{\partial \nu^J}{\partial E} \times \frac{\bar{\Gamma}^J_{\gamma f} . \bar{E}^J_{\gamma f}}{\Gamma_f^J}$$

$$\cong \nu_i^J + \frac{\partial \nu^J}{\partial E} . E - \delta\nu^J_{n,\gamma f} \qquad (2)$$

where $\nu_i^J + \frac{\partial \nu^J}{\partial E} E$ stands for the well known linear energy dependence of $\bar{\nu}_p$ for the immediate fission. The negative term stands for the (n,γf) process which reduces the energy available for fission by a prior γ decay and hence reduces $\bar{\nu}_p$. It involves the width $\overline{\Gamma_{\gamma f}}$ and the average γ energy $\overline{E_{\gamma f}}$ emitted in the (n,γf) reaction while Γ_f represents the total fission width.

From (2) we derive the following formalism which expresses the average number of prompt neutrons emitted by fission event as the sum of all the spin states contributions weighted by their fission strengths :

$$\bar{\nu}_p(E) = \frac{\sum_J (\nu_i^J + \frac{\partial \nu^J}{\partial E} E - \delta\nu^J_{n,\gamma f})\ \sigma_f^J(E)}{\sigma_f(E)} \qquad (3)$$

Obviously, this formalism is valid everywhere in the full energy range. In the resolved energy range and in the lower part of the unresolved range the "s" waves are predominant and the term $\frac{\partial \nu}{\partial E}.E$ is negligible so that (3) reduces to :

$$\bar{\nu}_p(E) = \frac{\sum_J (\nu_i^J - \frac{C^J}{\Gamma_f})\ \sigma^J_f(E)}{\sigma_f(E)} \qquad (4)$$

$$\text{where } C^J = \frac{\partial \nu^J}{\partial E} \times \bar{\Gamma}^J_{\gamma f} . \bar{E}^J_{\gamma f}$$

In the lower part of the unresolved range where the "s" waves are still predominant one has :

$$\bar{\nu}_p(E) = \frac{\sum_J \nu^J \sigma_f^J(E)}{\sigma_f(E)} - \sum_J \delta\nu^J_{n,\gamma f}(E) \qquad (5)$$

It is easy to show that the total (n,γf) effect for a J spin family is :

$$\Delta\nu^J_{n,\gamma f}(E) = \pi\lambda\!\!\!^{-2} . 2\pi\ S^J\ E^{1/2}\ \frac{1}{\sigma_f(E)} \cdot \frac{C^J}{\Gamma^J(E)} \cdot F_W^J \qquad (6)$$

In the expression (6) S^J is the "S" wave neutron strength function for the spin J, Γ_f^J is the total width and F_W^J a fluctuation corrective factor which can be set up to unity if the neutron width Γn is small compared to Γ.

Analysis

The calculational method of $\nu_p(E)$ is based on the formalism of relationships (4) and (5) using for ν_i^J and C^J the values obtained from the analysis of the pointwise microscopic results in the resolved range. A preliminary calculated $\nu_p(E)$ curve is compared to the other experimental data which are not given in a pointwise form, like, for example, those of GWIN. At this stage a carefull analysis may lead to the conclusion of a first renormalization for the calculations in order that they represent the best fit to the experimental data. Afterwards, large and consistent sets of thermal and epithermal systems are calculated using the $\nu_p(E)$ curve and the JEF1 library as data base. A so called "tendency research" method / 6 / is used to quantify a possible discrepancy between the integral and microscopic data concerning the neutron multiplicity, the cross-sections, the resonance integrals and the migration areas. This kind of adjustment involving first order perturbations and least squares fit is consistent, but in order to judge on its quality on the point of view of physics the adjusted integral data are compared to the well known corresponding quantities at thermal energy.

^{239}Pu

The original data by FREHAUT / 4 / have been renormalized by a factor 1.0066 as a result of a "a posteriori" better estimation of the background. The constants ν_i^J and C^J for the spin families 0^+ and 1^+ have been deduced from a least squares fit to the data where the ν_p and $E_{\gamma f}$ values are plotted as functions of Γ_f^{-1}. The negative correlation between them, which is representative of the $(n,\gamma f)$ effect, is clearly shown.

For a better representation of the experimental data in the whole energy range, in particular below 7.8 eV the constants derived from FREHAUT's data have been modified within the error bars. This modification corresponds to about a doubling of the spin effect and to an increase of 10 % of the $(n,\gamma f)$ effect. The final values used in the calculations are as follows :

$$\nu^{0+} = 2.8845 \qquad C^{0+} = 0.7\ 10^{-3}\ eV$$

$$\nu^{1+} = 2.8606 \qquad C^{1+} = 0.6941\ 10^{-3}\ eV$$

The calculations have been performed by using the resonance parameters recently derived by DERRIEN et al / 7 / and the multichannel multilevel REICH-MOORE formalism. In the resolved range the corrective term $\Delta\nu_{n,\gamma f}^J$ is calculated by expressing the resonance interference contribution as :

$$\sum_K \sum_L \frac{\sigma_{int}^{KL}(E)\ C^J}{\sigma_f(E)\,(\Gamma_f^K\ \Gamma_f^L)^{1/2}}$$

where K and L represent the two interfering resonances. At higher energy where averaged values are calculated, the interference contribution is neglected on the argumentation that it is negligible because of the small value of $\frac{\Gamma}{D}$ when the average energy intervals are large enough.

As expected the continuous $\nu_p(E)$ curve exhibits dips, each corresponding to a resonance. The observed structures have 2 origins :

- one due to the direct fission contribution which fluctuates between the two limits ν^{0+} and ν^{1+} according to the fission strengths of 0^+ and 1^+ spin families.

The figure 1 shows the good agreement of the $\nu_p(E)$ curve with the experimental values.

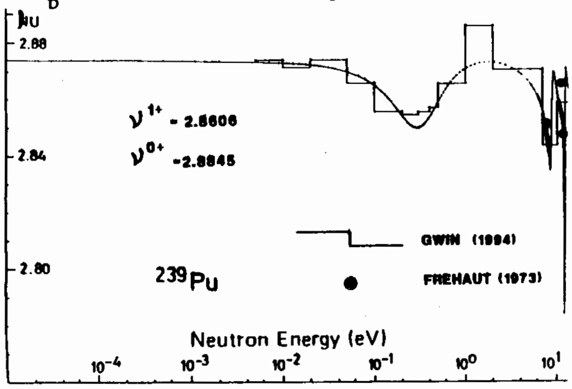

Fig. 1 - $\nu_p(E)$ for ^{239}Pu compared to the experimental data.

To qualify the $\nu_p(E)$ curve against integral data it is necessary to use a set of integral experiments as numerous as possible with different energy spectra. These conditions are perfectly fullfiled in the present case since have been used 26 plutonium fuelled criticals with three different moderators (light water, heavy water and graphite) and for each moderator several moderating ratios ranging from the one of the fully thermalized lattices to the one of epithermal tight pitch lattices. To summarize, the well thermalized media are mainly sensitive to the thermal value while the tight pitch lattices (also called low moderated systems) are strongly sensitive to the ν_p value at 0.296 eV resonance.

The set of integral data confirms the existence of the important dip observed by GWIN / 8 / around 0.3 eV. As a fact, if one uses for $\nu_p(E)$ a constant value equal to the thermal value, one obtains a least squares fit of medium quality since the χ^2 parameter has a value 35.2 instead of 26. The adjusted thermal ν_p value is 2.8594, significantly lower than the recommended value. The K_{eff} of the well thermalized media are underestimated (K_{eff} = 0.9975) while the ones of the epithermal systems are overestimated (K_{eff} = 1.00430). On the contrary if one uses the presented data, the thermal cross-sections and the K_{eff} are better estimated for both heavy water criticals (K_{eff} = 0.99883) and the epithermal systems (K_{eff} = 1.00350) and χ^2 is equal to 23.9. All these final informations are included in the tables I and II. To note that in the table I the thermal systems refer to all thermal systems (light water plus heavy water systems).

Table I

Comparison of $\langle K_{eff}\rangle - 1$ values (in pcm) calculated for all investigated thermal systems using a constant value and the present evaluation of ν_p.

$\bar{\nu}_p$	ν_p = cte	Present analysis
Original data	213 ± 481	11 ± 484
"Adjusted"	68 ± 464	77 ± 450
Adjusted $\bar{\nu}_p$ thermal	2.8608±0.007	2.8664±0.007

Table II

Comparison of $(\langle K_{eff}\rangle - 1)$ values (in pcm) calculated for epithermal systems using a constant value and the present evaluation of ν_p.

$\bar{\nu}_p$	$\bar{\nu}_p$ = cte	Present analysis
Original data	350 ± 368	22 ± 500
"Adjusted"	134 ± 378	66 ± 381
Adjusted $\bar{\nu}_p$ thermal	2.8602±0.007	2.8670±0.007

The ν_p(E) value at E = 0.0253 eV is 2.8726 to be compared with the data evaluated by AXTON (ν_t^{th} = 2.8736 $\pm$ / 9 / and by DIVADEENAM and STEHN (ν_t^{th} = 2.8768 $\pm$ 0.0057 / 10 /.

We have checked that the DOPPLER effect between 300°K and 1000°K is small.

^{241}Pu

Due to a lack of experimental informations the analysis is much more difficult for this nucleus. The data by FREHAUT / 11 / show a real but small spin effect :

ν^{2+} = 2.922 $\qquad$ ν^{3+} = 2.9082

They don't allow a firm conclusion about the existence of the (n,γf) effect. SIMON / 12 / theoretically investigated this problem. He calculated a $\Gamma_{\gamma f}$ value which is now in accordance with the observed total fission widths and gave indications that $(\Gamma_{\gamma f}.E_{\gamma f})^{2+}$ > $(\Gamma_{\gamma f}E_{\gamma f})^{3+}$. From these informations we conclude to the existence of a (n,γf) process.

Preliminary calculations have been performed using for C^{2+} and C^{3+} the values obtained by SIMON and for ν^{2+} and ν^{3+} the values deduced from FREHAUT's data. A renormalization factor of 1.0162 is needed to fit into GWIN'S data. Finally a renormalization factor of 1.01375 has been obtained leading to the following constants :

ν^{2+} = 2.9618 $\qquad$ C^{2+} = 1.155 10^{-4} eV

ν^{3+} = 2.9480 $\qquad$ C^{3+} = 0.9 10^{-4} eV

Among the 26 thermal and epithermal systems, only 7 were loaded with significant quantities of ^{241}Pu. This set is not large and diversified enough to undoubtly confirm the existence of a dip at the location of the first resonance, but the informations in the thermal range are reliable. The degree of consistency between microscopic and integral data is shown in the table III.

Table III

Comparison of $(\langle K_{eff}\rangle - 1)$ values in pcm calculated for thermal and epithermal systems

ν_p	ν_p = cte	Present analysis
Original data	- 100 ± 490	- 5 ± 480
"Adjusted"	69 ± 400	67 ± 380
Adjusted $\bar{\nu}_p$ thermal	2.942 ± 0.010	2.944 ± 0.010

The ν_p(E) value at E = 0.0253 eV is 2.942 ± 0.010 in good agreement with the values recommended by DIVADEENAM and STEHN (ν_t = 2.9369 $\pm$ 0.0074) and by AXTON / 14 / (ν_t = 2.9375 $\mp$ 0.007).

The figure 2 shows the energy dependence of $R(E) = \frac{^{241}\nu}{^{252}\nu}$ compared to the experimental data.

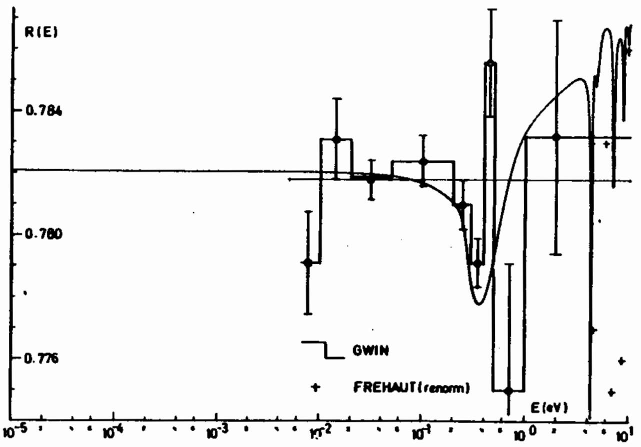

Fig. 2 - Energy dependence of $R(E) = \frac{^{241}\nu}{^{252}\nu}$ relative to ^{241}Pu.

^{235}U

The available experimental data for this nucleus are quite numerous. All show fluctuations which are located at the same energies prouving that these fluctuations have a real physical origin. The analysis has been performed in the same way as for the other investigated nuclei, but limited to the resonances with Γ_f less than 50 meV. Our results are similar to those of a previous study by SIMON / 12 / : the spin effect is not clearly evident from FREHAUT's data analysis and the ν_i values for 4^- and 3^- spin states are similar within the error bars (2.421 ± 0.009 for $J^\pi = 4^-$ and 2.419 ± 0.027 for $J^\pi = 3^-$). Since we expected from the Plutonium isotopes study $\nu_i^{4-} < \nu_i^{3-}$, we have adopted a small value for the spin effect $\Delta\nu = \nu_i^{3-} - \nu_i^{4-} = 0.0035$.

The evidence of the (n,γf) process is supported by the inequality $(\Gamma_{\gamma f}.E_{\gamma f})^{3-} > (\Gamma_{\gamma f}.E_{\gamma f})^{4-}$, although it has to be noted that for both spin states the product $\Gamma_{\gamma f}.E_{\gamma f}$ is determined with a large error bar.

The best fit of the microscopic data is obtained with the following parameters :

$\nu^{3-} = 2.423$ $\quad C^{3-} = 1.398 \times 10^{-4}$ eV

$\nu^{4-} = 2.4195$ $\quad C^{4-} = 1.860 \times 10^{-4}$ eV

The calculations have been performed with the resonance parameter of the Neutron Cross Sections Book Vol. I (1984) and have to be considered as preliminary.

The figure 3 compares the present calculations with the experimental data by GWIN / 8 /, FREHAUT / 11 /, REED / 13 / after a renormalization to the value adopted for the neutron multiplicity of the spontaneous fission of ^{252}Cf. The agreement is acceptable apart an observable energy shift. Nevertheless the experimental data by GWIN show a clear decrease of the average value above 1 eV compared to the one below. This same trend is indicated by the calculation and has to be analysed as the conjunction of a (n,γf) effect (dips in the $\nu_p(E)$ curve) and of a spin effect.

As a fact, in the energy interval 1 eV - 10 eV the 4^- resonances are more numerous and the contribution of their fission strength is major. Another interesting point is the small slope in the subthermal range, due to the bound state close to Bn (E = - 0.02 eV) with a small Γ_f (44 meV) as given by the Neutron Cross Sections Book. The existence of such a bound state has not been clearly demonstrated but is likely, being given the respective values of the average spacing (D = 0.44 eV) and the energy interval with the first observed resonance.

For the moment the positive energy dependence in the subthermal range is purely speculative but could be envisaged as a possible solution of the long standing problem of the η shape in the subthermal range.

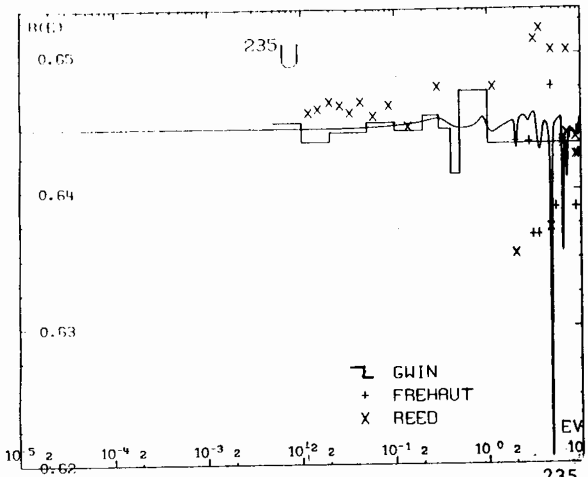

Fig. 3 - Energy dependence of $R(E) = \frac{^{235}\nu}{^{252}\nu}$ relative to ^{235}U

37 thermal systems loaded with ^{235}U have been used for the analysis checking. The comparison of the microscopic $\nu_p(E)$ data with the integral data is shown in the table IV.

Table IV

Comparison of (<K_{eff}>-1) values in pcm calculated for thermal systems

ν	ν_p = cte	This analysis
Original data	- 75 ± 612	- 57 ± 615
Adjusted	- 13 ± 434	- 15 ± 434
ν_t adjusted	2.430 ± 0.004	2.430 ± 0.004
ν_p adjusted thermal	2.414 ± 0.004	2.414 ± 0.004

The $\nu_p(E)$ value at thermal energy is 2.4 in excellent agreement with the data published in the literature.

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