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STUDY OF FISSION γ-RAY YIELDS FROM LOW-ENERGY RESONANCES OF ^{237}Np

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Abstract. A study of fission γ-ray yields from ^{237}Np low-energy resonances has been performed at the Dubna IBR-30 pulsed reactor. A multiplate fission chamber with 1.5 g of high purity ^{237}Np for detection of fission events, and a large 6-section liquid scintillation detector for detection of 3 or more γ-quanta in coincidence with fission fragments were used for fission γ-ray yield measurements. A "$1/\Gamma_f$" dependence was found for the γ-ray yields in ^{237}Np resonances. This experimental result can be interpreted as an indication of a prefission γ-ray emission, i. e. of the existence of the $(n, \gamma f)$ process.

1. Introduction

An experimental study of low-energy neutron induced fission of ^{237}Np gives interesting and important information about the intermediate structure of the fission cross-section σ_f, allows some of the fission barrier parameters to be obtained, etc. [1, 2]. Unfortunately, due to the subbarrier fission of ^{237}Np, the average $\langle\sigma_f\rangle^{(7)}$ at neutron energies $E_n < 500$ eV is with almost 3 orders of magnitude less than the $\langle\sigma_f\rangle^{(5)}$ for ^{235}U [3]. The ranges of the measured fission widths $\Gamma_f^{(7)}$ for ^{237}Np and $\Gamma_f^{(5)}$ for ^{235}U are as follows: $10^{-6} \leq \Gamma_f^{(7)} \leq 10^{-3}$ eV and $10^{-3} \leq \Gamma_f^{(5)} \leq 1$ eV [4, 5].

To find whether the subbarrier fission of ^{237}Np is a good subject for researching the $(n, \gamma f)$ process [6, 7] one can make some simple estimations. If the resonance value of Γ_f is $\approx 10^{-6}$ eV, then the life-time of the resonance state that decay through the fission can be up to $\approx 10^{-9}$ s. The probability of a prefission γ-quantum being emitted could become significant in comparison with ^{235}U or ^{239}Pu resonances that have much larger Γ_f values. One can conclude that the existence of very weak ^{237}Np fission resonances with Γ_f values of $\approx 10^{-6}$ eV could be a serious advantage and, also, a reason for searching the $(n, \gamma f)$ process at the subbarrier fission of this nucleus.

To observe the $(n, \gamma f)$ process, we studied the variation of fission γ-ray yields from ^{237}Np resonances. It was expected that the yields should increase if an additional prefission γ-quantum was added to the fission γ-rays with a multiplicity of ν_γ.

As in previous papers, discussed in O. Shcherbakov's review [8], we used the method of coincidences between the fission fragment and the detected γ-quanta. Below we describe our experimental technique and the results obtained in these measurements. A possible connection between our results and the experiments with aligned ^{237}Np nuclei [9] is discussed.

2. Instrumentation and Results

The fission γ-ray yields from ^{237}Np low-energy resonances were obtained in time-of-flight (TOF) measurements at the Dubna IBR-30 pulsed neutron source. The repetition rate of the reactor neutron bursts is $100\,\mathrm{s}^{-1}$ and the half-width of the burst is $4\,\mu$s. The neutron flight-path was chosen to be 58.5 m. A multiplate ionization fission chamber (FC) loaded with 1.5 g of high purity ^{237}Np was used for detection of fission events. One of the sections of the FC contained small ^{235}U target, which was used for calibration purposes. The FC was inserted into the central hole of a large 6-section liquid scintillation detector (LSD) [10]. Having a low sensitivity to the prompt fission neutrons, the LSD detects fission γ-rays with an efficiency ε_γ of about 40 % at the γ-ray detection threshold of ≈ 0.2 MeV [10]. To provide more reliable detection of the fission γ-rays among the large LSD γ-ray background, due to the target radioactivity, scattered neutrons, etc., a fast coincidence between 3 or more γ-quanta was used. A slow "fission fragment $\geq 3\gamma$-quanta" coincidence identifies them as fission γ-rays. A computerized module was used for simultaneous recording of 4 TOF spectra: the coincidence and fission TOF spectra for ^{237}Np and similar spectra for ^{235}U. The layout of the FC and LSD at the IBR-30 neutron beam 3, together with the electronics, are shown in Fig. 1.

Special attention is drawn to the determination of the background in both ^{237}Np and ^{235}U TOF spectra. A method of resonance filters (Mn, Co, Rh, Cd) has been used to determine the background in ^{235}U TOF coincidence and fission spectra. Unfortunately, we could not apply this method to the ^{237}Np TOF spectra, because the interresonance σ_f is of about 10^{-2} barns [3] and, hence, resonance filter background measurements, similar to those as for ^{235}U, would have continued for an unreasonably long time. Also, due to the multiplication of neutrons in the reactor core, the IBR-30 emits delayed neutrons between neutron bursts. In the case of the subbarrier fission of ^{237}Np the fast neutron background in both the TOF fission and coincidence spectra becomes significant and cannot be avoided. For accurate determination of this background 15 intervals between the ^{237}Np fission resonances were chosen and a polynomial function has been used to draw the background curves.

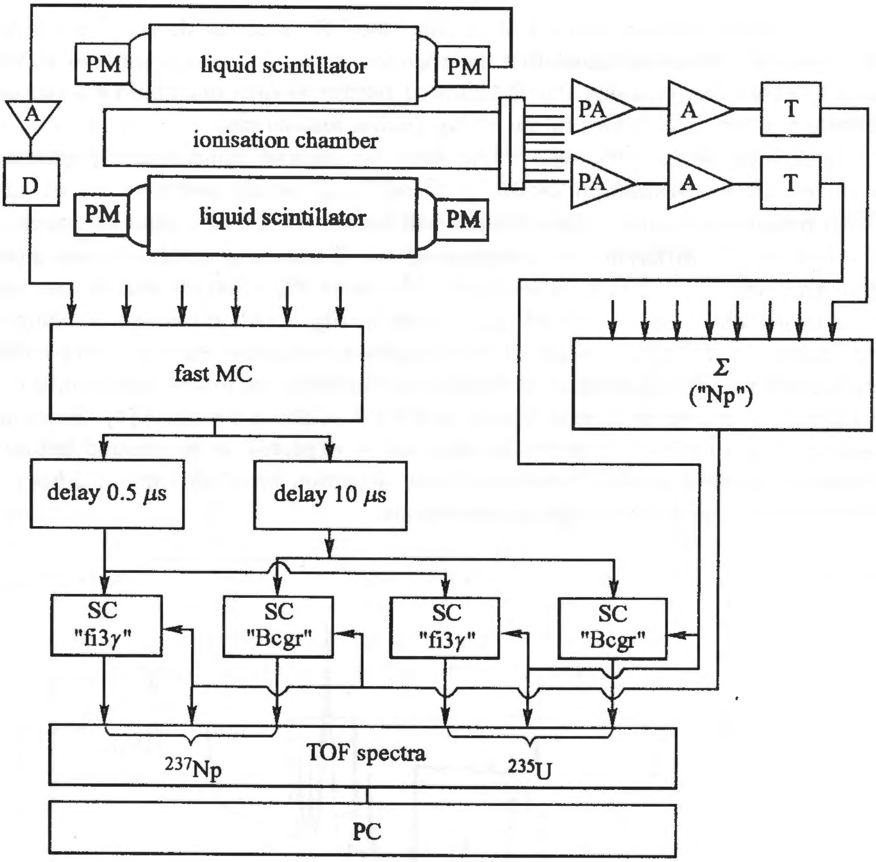

Fig. 1. Block-diagram of the detector electronics
PM — photomultipliers, PA — preamplifier, A — amplifier, T — threshold, D — discriminator, MC — majority coincedence, S — summator, SC — slow concedence, Bcgr — background

The fission γ-ray yields R from ^{237}Np resonances studied in these measurements are defined as a ratio of the total number of coincidences and fissions taken from both coincidence and fission TOF spectra at the corresponding resonances [11]

$$R = \frac{\sum_i N_c}{\sum_i N_f} \,. \tag{1}$$

One can also see that R values are actually the LSD detection probabilities of fission events with average γ-ray multiplicities $\langle \nu_\gamma \rangle$ and 3 or more detected fission γ-quanta. Since we need to know the $\langle \nu_\gamma \rangle$ values, one could extract them by using the calculation approaches developed by Theobald et al. [12] and by Muradyan et al. [13]. On the other hand, our measurements [11] of the averaged yields $\langle R \rangle$ of the fission γ-rays for ^{233}U, ^{235}U and ^{239}Pu isotopes, together with the $\langle \nu_\gamma \rangle$ measurements [14, 15] for the same isotopes, allow a shift from the "R" scale to the "ν_γ" scale to be made. A rough estimation shows that the change in $\Delta\nu_\gamma = 1$ corresponds to a ΔR of about 0.1. Therefore, studying the R yields of fission γ- rays one could also estimate the changes in the $\langle\nu_\gamma\rangle$ values in the ^{237}Np fission resonances.

An evaluation of the ^{235}U and ^{237}Np TOF fission and coincidence spectra taken with a γ-ray detection multiplicity of "≥ 2"and "≥ 3" would also give the ν_γ values at ^{237}Np fission resonances. These results will be the subject of a separate paper.

The results of 2 different measurements of the R values in resolved resonances at neutron energies $E_n \leq 10$ eV, in resonance cluster at $E_n = 40$ eV and in unresolved clusters at 100 eV $< E_n <$ 500 eV are shown in Fig. 2. All measured R values are normalized to the R(39.9 eV) value of the strongest resonance at the $E_n = 40$ eV fission resonance cluster. This resonance is expected to provide the lowest probability of the $(n, \gamma f)$ process due to its largest fission width Γ_f of about 6.4 MeV [4]. To estimate the energy $E_{\gamma f}$ of the prefission γ-quanta that is expected to be emitted before the fission of compound nuclei, 2 different γ-ray detection thresholds of $\approx$0.2 MeV and $\approx$0.6 MeV were chosen for these measurements.

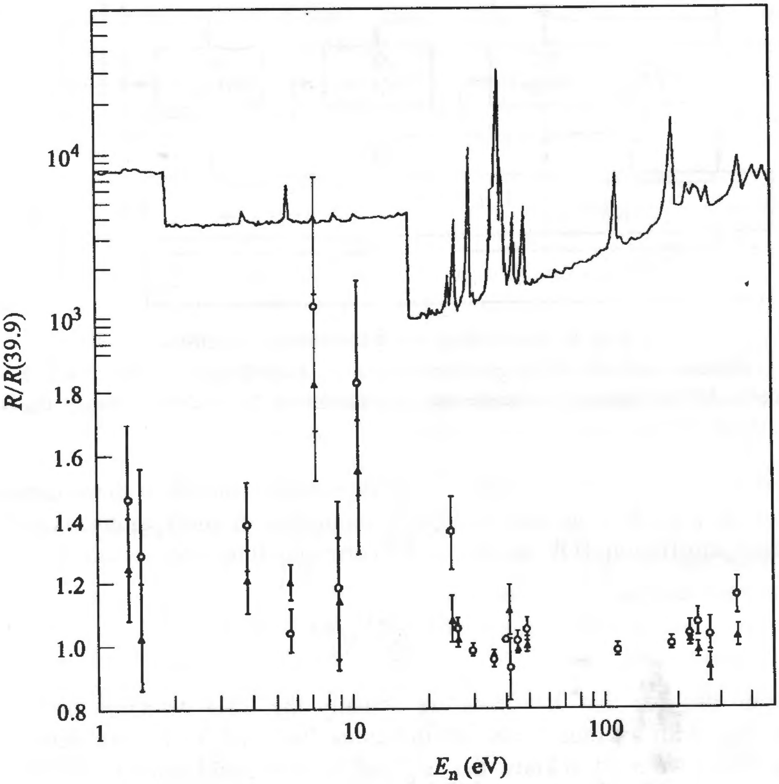

Fig. 2. Fisison γ-ray yields R in the neutron energy range $E_n = 1$-500 eV γ-ray detection thresholds: ▲ — 0.2 MeV; ○ — 0.6 MeV

In the neutron energy interval from 100 eV to 500 eV most of the points in Fig. 2 are close to $R = 1$, probably because they were averaged over many resonances with a wide range of Γ_f values. Unfortunately, we could not reach any satisfactory statistical accuracy in the R values below $E_n \leq 10$ eV, even though the main measurement conditions (fission fragment and γ-ray detection efficiencies, total amount of ^{237}Np in the FC, measurement time, etc.) are close to their reasonable limits. This problem is mainly because all measured resonances below 10 eV have extremely low fission widths of $\Gamma_f \leq 10$ μeV. Therefore, no conclusions about the individual R values of those week resonances can be made. More correct would be both the weighted and averaged values of $\langle R\rangle_w = 1.08 \pm 0.04$ and of $\langle R\rangle = 1.12 \pm 0.06$ for all resonances below 10 eV to be considered. Then, since $\langle R\rangle \approx 1.1$ one should expect that the prefission γ-quantum is emitted in almost all fission events. In principal, the value of $\langle R\rangle > 1.1$ could also mean that 2 prefission γ-quanta are emitted. However, due to the large errors in R values this hypothesis is not discussed below.

The most surprising result was obtained for the fission resonances that belong to the first cluster at $E_n = 40$ eV. As one can see from Fig. 2 and Fig. 3 both measurements with different γ-ray detection thresholds, E_γ, give similar results: the weaker the resonance (the less Γ_f values), the larger the value of R. The values of R increase at the "wings" of the resonance cluster with about 5 % relative to the value of R(39.9 eV). This observation could be interpreted as an enhancement of the "$1/\Gamma_f$" dependence of R values and, hence, as of the existence of the $(n, \gamma f)$ process.

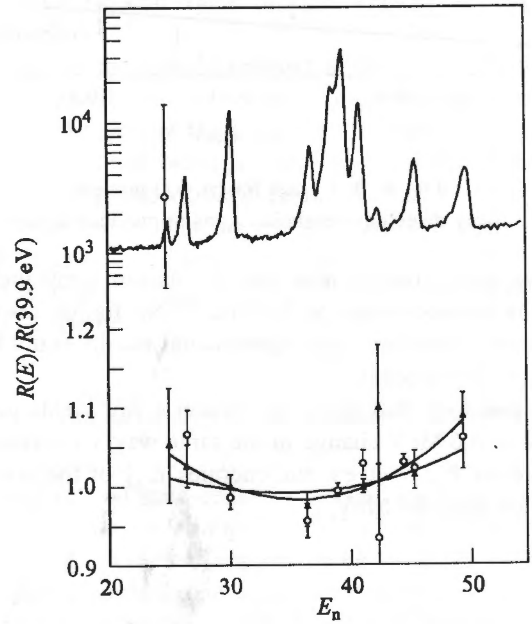

Fig. 3. Fission γ-ray yields R in the $E_n = 40$ eV cluster resonances γ-ray detection thresholds — as in previous figure

To obtain some additional arguments that should support this assumption we also plotted the resonance R value versus the reversed fission widths $1/\Gamma_f$ for each measurement with a certain γ-ray detection threshold, E_γ. For more objective check of that part of the "R–$1/\Gamma_f$" dependence, which corresponds to the week resonances and large $1/\Gamma_f$ values, we represent this function in linear scale. As shown in Fig. 4, both measurements with $E \approx 0.2$ MeV and ≈ 0.6 MeV satisfy the "$1/\Gamma_f$" test giving a correlation coefficients of 0.6 ± 0.2.

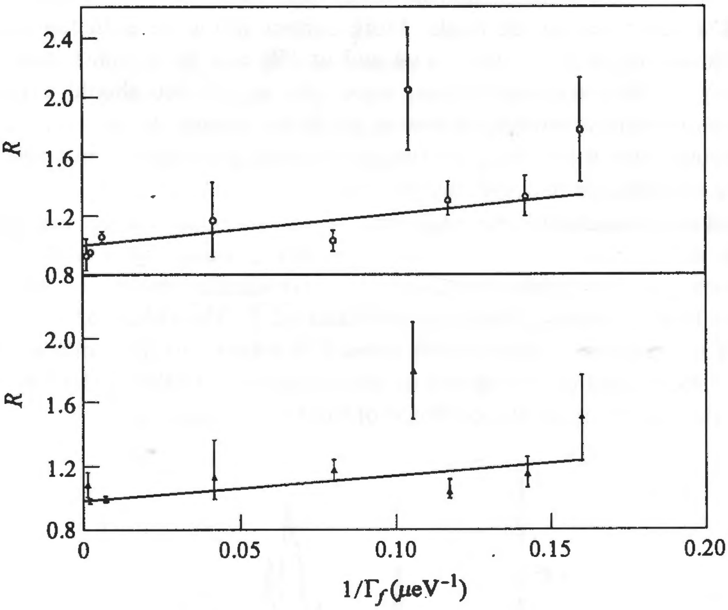

Fig. 4. $1/\Gamma_f$ test for $(n, \gamma f)$ process
γ-ray detection thresholds — as in previous figure

In conclusion we would like to note that the fission γ-ray yield measurements at $E_n \leq 10$ eV and the measurements at the first ^{237}Np fission resonances cluster give a positive "$1/\Gamma_f$" test. Therefore, our experimental results could be interpreted as an observation of the $(n, \gamma f)$ process.

One could also conclude that since the fission γ-ray yields measured at different E_γ values from 0.2 to 0.6 MeV change in the same way, i. e. arising at the "wings" of the resonance cluster at $E_n = 40$ eV, the energies $E_{\gamma f}$ of the prefission γ-quanta are estimated to be larger than 0.6 MeV.

3. Discussion

As a result of the $(n, \gamma f)$ process that might exist in the subthreshold fission of ^{237}Np, an emission of E1 or M1 prefission γ-quanta is expected. Then the initial spin and parity J^π of the ^{238}Np compound nucleus change to a new value of $(J^{\pm\pi})_{\gamma f}$. It would be interesting to compare the life-times $T_{\gamma f}$ of such compound nucleus excited states that decay through the emission of prefission γ-quantum with the life-times $T \sim \Gamma_f^{-1}$ of the ^{237}Np fission resonances. A rough estimation of the $T_{\gamma f}$ value based on the well known Weisscopf's $E_{\gamma f}^3$ dependence at $E_\gamma \approx 0.6$–1 MeV seems to be consistent with the value $T \sim \Gamma_f^{-1}$ at $\Gamma_f \approx 10^{-3}$–10^{-6} eV.

If the $(n, \gamma f)$ process exists, then it is natural that the fission γ-ray multiplicity $\nu_{\gamma f}$ is expressed in the following way [8]:

$$\nu_{\gamma f} = \nu_\gamma + \nu_{\gamma f}^0 \frac{\Gamma_{\gamma f}}{\Gamma_f} \, . \tag{2}$$

Here, $\nu_\gamma \approx 7$–8 [14, 15] is the fission γ-ray multiplicity with no $(n, \gamma f)$ process and $\nu_{\gamma f}^0$ is the multiplicity of prefission γ-quanta. Usually, one assumes that $\nu_{\gamma f}^0 = 1$. Since at the "wings" of the resonance cluster at $E_n = 40$ eV the R values increase with $\approx 5\,\%$ relative to the $R(39.9\text{ eV})$ the ratio $(\Gamma_{\gamma f}/\Gamma_f)$ is estimated to be about 0.35–0.40. This means that 35–40 % of the fission events in the cluster resonances are accompanied by emission of prefission γ-quanta.

Below the neutron energy of 10 eV, the weighted value of $\langle R \rangle_w$ is close to $R \approx 1.1$. One could assume that in the weakest resonances the case of $\Gamma_{\gamma f} \sim \Gamma_f$ is realized, i. e., the probability of the $(n, \gamma f)$ process is close to 100 %.

An estimation of $1\ \mu\text{eV} < \Gamma_{\gamma f} < 10\ \mu\text{eV}$ can be extracted from our experimental data below an E_n of 10 eV. Our value of $\Gamma_{\gamma f} \approx 1$–$10$ μeV exceeds the value of $\Gamma_{\gamma f} \approx 10^{-7}$ eV obtained in [16].

Since all resonances in the first Np cluster at $E_n = 40$ eV have $J^\pi = 3^+$ [17], the emission of the E1 prefission γ-quantum changes the value of this initial spin to $J^\pi = 2^-, 3^-$ and 4^-. The lowest fission barrier is expected to have new $(J_{\gamma f}, K)$-quantum numbers of $(3^-, 0)$ and belongs to the $K = 0$ mass asymmetry band.

Let us consider the disagreement between the results of ^{237}Np polarization experiments [17], which have shown that the $(3^+, 3)$ fission channel is predominant, and the fission fragment angular distribution study of ^{237}Np aligned nuclei [9]. The latter found the angular distribution coefficients A_2 to be almost half the expected value of $A_2(3^+, 3) = 3.35$ [17], where

$$A_2 = \frac{15}{4} \frac{I}{I+1} \left(\frac{3K^2}{J(J+1)} - 1 \right) \, . \tag{3}$$

If the $(n, \gamma f)$ process exists and a E1 prefission γ-quantum is emitted, then, as it is shown above, a new $(3^-, 0)$ state could be reached having a value of $A_2 = -2.68$. Assuming that such γ-quanta are emitted in 30 % of the fission events, we find that the value of the resulting coefficient reduces down to $A_2 = 1.54$. An emission of a M1 prefission γ-quantum could excite a new $(2^+, 0)$ state with a rather low fission barrier and a value of $A_2(2^+, 0) = -2.68$. In general, both possibilities considered could explain the A_2 value obtained by Kuiken at al. [9]. If the new $(J_{\gamma f}, K)$ states with $K = 0$, due to some reasons, are forbidden, then the states with $K = 1$ and $K = 2$ would also reduce the measured A_2 values.

An alternative explanation of Kuiken's results [9] has been proposed in [17]. Keyworth et al. [17] observed an admixture of $K = 3$ and $K = 2$ states that also could explain lower A_2 values measured in [9].

In conclusion, we would like to note that our results would be more reliable if the measurements could have been performed at better fast neutron background conditions, *i. e. by using other pulsed neutron sources with no multiplication of neutrons in the* neutron producing target. Only then one could totally solve this extremely interesting enigma.

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