

Resonance neutron induced fission of ^{239}Pu

F.-J. Hamsch¹⁾, I. Ruskov^{1,2)}, L. Dematté³⁾

- 1) European Commission, Joint Research Centre, Institute for Reference Materials and Measurements, Retieseweg 111, 2440 Geel, Belgium
 - 2) Bulgarian Academy of Sciences, Institute for Nuclear Research and Nuclear Energy, Tzarigradsko chaussee 72, 1784 Sofia, Bulgaria
 - 3) CINECA, I-40033 Casalecchio di Remo, Italy
- Franz-josef.hamsch@ec.europa.eu

Abstract: A measurement of the resonance neutron induced $^{239}\text{Pu}(n, f)$ fission fragment kinetic energy and mass distributions has been performed using the twin Frisch grid ionization chamber technique at the GELINA white spectrum pulsed neutron source. Special emphasis was devoted to cope with the strong α -activity of the ^{239}Pu target, taking advantage of an improved pulse pile-up rejection system. For incident neutron energies up to about 200eV all resonances could be resolved and fission fragment mass and total kinetic energy distributions deduced. Compared to a similar experiment on $^{235}\text{U}(n, f)$, in the same resonance region, less pronounced fluctuations of the fission fragment mass and total kinetic energy have been observed in the case of $^{239}\text{Pu}(n, f)$. From a physical point of view such fluctuations have been expected, because the only possible low-energy spin states ($J^\pi = 0^+, 1^+$) belong to well separated (about 1.25MeV) compound system transition state bands. A small spin dependence of about 70keV has been found for the fission fragment mean total kinetic energy in the neutron energy range above 1eV. This means that viscosity effects could take place during the fission of ^{240}Pu . A recently developed theoretical approach has given a possible explanation of the absence of pronounced fluctuations of $^{239}\text{Pu}(n, f)$ fission properties. The experimental two-dimensional mass-total kinetic energy distributions have been interpreted within the theoretical multi-modal fission model of Brosa et al.

Introduction

The investigation of the neutron induced fission of ^{239}Pu is still of primary interest both from fundamental and applied physics point of view. The capture of a s-wave neutron (spin $1/2^+$) with thermal or resonance energy by ^{239}Pu ($I^\pi = 1/2^+$) forms the ^{240}Pu compound nucleus (CN) mainly in 2 states [1] with spins $J^\pi = 1^+$ and $J^\pi = 0^+$. They belong to two well separated ($\sim 1.25\text{MeV}$) transition state bands with $K^\pi = 1^+$ and $K^\pi = 0^+$ [2]. If the coupling between the collective and single-particle degrees of freedom is weak (the system is adiabatic, not viscous) then the energy difference between the bands should appear after scission in the mean total kinetic energy $\langle \text{TKE} \rangle$ of the primary (before neutron emission) fission fragments (FF) from resonances of both spin groups [3].

In case of resonance neutron induced fission of ^{235}U [4, 5] quite pronounced fluctuations of the FF mass (A) and total kinetic energy (TKE) from resonance-to-resonance were observed. They were interpreted within the frame of the multi-modal random-neck-rupture (MM-RNR) model of fission [6]. The transition state spectrum in a ^{236}U CN above the fission barrier results from the mixing of $J^\pi = 3^-$ and 4^- for $K^\pi = 1^-$

and $K^\pi=2^-$ bands.

Experimental data on $^{239}\text{Pu}(n,f)$ are very important for the design of nuclear facilities and in view of nuclear waste management. Knowledge about the average prompt neutron emission $\langle v_p \rangle$ from this reaction becomes important since ^{239}Pu is used in mixed oxide (MOX) fuel elements. In the resonance neutron energy region quite large fluctuations of $\langle v_p \rangle$ have been observed [7]. These fluctuations have a significant impact on the reactivity coefficient of advanced water reactors [8], but their origin still is not quite clear. Are they correlated with $Y(A, \text{TKE})$ -distribution fluctuations, as it was found for $^{235}\text{U}(n,f)$ [4], or are they a result of the competition of the direct fission $^{239}\text{Pu}(n,f)$ with $^{239}\text{Pu}(n, \gamma f)$ -reaction [9]? The influence of the $(n, \gamma f)$ -reaction has been observed in the neutron $\langle v_p \rangle$ and gamma $\langle v_\gamma \rangle$ multiplicities, as well as in the average gamma energy $\langle E_\gamma \rangle$ [10-12] and in the FF independent yields [13]. Results of two measurements of FF characteristics at neutron resonance energies have been reported in the past [3, 14] with apparently controversial outcome.

Therefore, new measurements of $^{239}\text{Pu}(n,f)$ FF $Y(A, \text{TKE})$ -distributions in the resolved resonance region have been performed at Geel Electron LINear Accelerator (GELINA) "white" spectrum neutron source time-of-flight (TOF) spectrometer of IRMM in Geel, Belgium.

Data acquisition, analysis and discussion

The experimental setup and some preliminary results have already been published elsewhere [15-17]. A twin ionization chamber (IC) with Frisch grids was used as charged particle (α , FF) spectrometer. As detecting gas pure CH_4 was used at a pressure of $\approx 1.1 \times 10^5 \text{ Pa}$ (electron drift velocity $\sim 10 \text{ cm}/\mu\text{s}$) in a continuous gas flow rate of about 0.1 l/min, securing nearly constant FF pulse amplitudes during the duration of the experiment. The electron collecting time was $\sim 300 \text{ ns}$.

Table 1. ^{239}Pu sample characteristics

Support		
Backing	Material	Polyimide
	Thickness	$\approx 36 \mu\text{g}/\text{cm}^2$
Coating	Material	Au
	Prep. method	Evaporation
	Thickness	$\approx 79 \mu\text{g}/\text{cm}^2$
Pu target		
Chemical form		PuF_3
Enrichment, ^{239}Pu		$99.9774 \pm 0.0027 \%$
Preparation method		Evaporation
Layer	Diameter	$\approx 45 \text{ mm}$
	Thickness	$\approx 32 \mu\text{g}/\text{cm}^2$
Total mass of Pu		$\approx 514 \mu\text{g}$
Specific α -activity		$\approx 1 \text{ MBq}$

The main characteristics of the target are summarized in Table 1. The IC was installed at a distance of $\approx 9.4 \text{ m}$ from the GELINA neutron producing target. This way all the resonances were measured simultaneously in the same experimental conditions.

Five parameters were recorded in list-mode (event-by-event): the neutron TOF, 2 anode amplitudes containing FF kinetic energy information and 2 cathode-grid electron drift times, from which the FF emission angles were determined. The latter were used also for calculating the energy losses of FF in the sample and backing [4, 15-19].

The data acquisition was performed with two different GELINA parameter sets, corresponding to two different neutron energy ranges (Table 2).

Table 2. GELINA set of parameters

f [Hz]	100	800
$\langle I_e \rangle$ [μ A]	≈ 40	≈ 75
L [m]	≈ 9.4	≈ 9.4
Δt [ns]	≈ 2000	≈ 1
E_n [eV]	≥ 0.01	≥ 0.3
δE_n ($E_n=10$ eV)	≈ 0.02	≈ 0.006
fission events	$\sim 4 \cdot 10^6$	$\sim 5 \cdot 10^6$
In-beam filter	Cd	BC ₄
ΔE_n , eV	0.008–1	0.3–200

Because of strong pile-up between the pulses from α -particles and FF, a special pile-up rejection system [16] was applied leaving only $\sim 25\%$ of all collected fission events for the analysis. The angular cone of accepted events was restricted to $\cos(\theta) \geq 0.3$ to avoid events with too much degraded kinetic energies.

For 27 incident neutron kinetic energy intervals from 0.008eV up to 1eV, 22 single

isolated resonances with $J^\pi=1^+$ and 9 resonances with $J^\pi=0^+$ from 1eV up to 200eV, as well as for 6 intervals between the resonances, two-dimensional $Y_{L,H}(A,KE)$ distributions for light (L) and heavy (H) fragments were obtained for the first time. Because primary $Y(A,TKE)$ -distributions, after applying all the corrections, should be symmetrical with respect to mass $A=120$, only heavy fragment (HF) $Y(A,TKE)$ -distributions were used in the further analysis, particularly when a model was fitted to them.

The calculated $^{239}\text{Pu}(n_{th},f)$ FF $\langle A \rangle_{th}$ and $\langle TKE \rangle_{th}$ and their standard deviations were in agreement with the available literature data in the limits of their experimental uncertainties [16, 20, 21-25].

$^{239}\text{Pu}(n_{th},f)$ reaction data, obtained at 100Hz, served as a reference for the TOF-spectra calibration and for comparison to the $J^\pi=1^+$ and 0^+ resonance FF mass-energy distributions. One should keep in mind that to the thermal neutron induced FF yield contribute a broad resonance at $E_n < 0$ with $J^\pi=0^+$ [26] ($\sim 63\%$) and resonances at $E_n > 0$ (mainly $J^\pi=1^+$ resonance at $E_n < 0.296$ eV) ($\sim 37\%$) [27, 28].

Comparison of fission fragment mass yield and kinetic energy distributions

Qualitatively, the experimentally obtained resonance neutron FF (mass, kinetic energy)-yields $Y_{res}(A, KE)$ and their projections $Y_{res}(A)$ and $Y_{res}(KE)$ were compared to the thermal neutron ones using the Kolmogorov-Smirnov test on the corresponding distributions.

Quantitatively, the comparison was done by calculating the difference and ratio between the corresponding one-dimensional distributions, as follows $\Delta Y_{res}(A) = Y_{res}(A) - Y_{th}(A)$ and $R_{res}(A) = Y_{res}(A) / Y_{th}(A)$, $\Delta Y_{res}(KE) = Y_{res}(KE) - Y_{th}(KE)$ and $R_{res}(KE) = Y_{res}(KE) / Y_{th}(KE)$

The results for the 0.296eV (in short 0.3eV) 1^+ resonance are shown in Figure 1a and Figure 1b, the ratio on top of the difference. There is a pattern structure seen in the asymmetric part relative to the $Y_{th}(A)$ distribution. A definite decrease in the symmetric yield for the 0.296eV resonance compared to thermal is visible, which reaches about 30%. In an early radiochemical experiment [29] the ratio of the yield of ^{99}Mo to that of ^{115}Cd from the 0.296eV resonance neutron induced fission $R(99/115)^{0.3eV}$ was found to be 3 times larger than that from fission with thermal neutrons, $R(99/115)_{th}$. The value $R(99/115)^{0.3eV}_{th} = 3.00 \pm 0.28$ from Ref. [29] is ~ 2.5 times higher than the value of $R(99/115)^{0.3eV}_{th} = 1.15 \pm 0.06$, which can be deduced from Figure 1a. This significant difference is due, probably, to the different experimental techniques which have been adopted.

A similar pattern as observed in Figure 1 is seen in the difference between the yield from all $J^\pi=1^+$ resonances $Y_{1^+}(A)$ relative to $Y_{th}(A)$. Here $R(99/115)^{1^+}_{th}=1.19\pm 0.07$, which is of the same order of magnitude as that for the 0.296eV resonance, which can be expected, since this resonance has the same $J^\pi=1$.

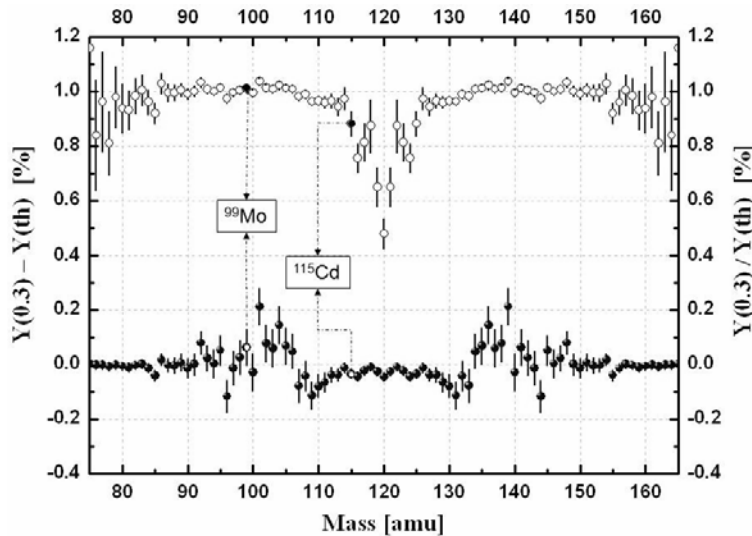


Figure 1a. $Y(0.3)$ mass yields relative to thermal $Y(th)$ ones.

The ratio and the difference between the mass-yields from the resonances of both spin groups $J^\pi=1^+$ and 0^+ show similar patterns in mass-yield difference, as those in Figure 1a, but here the fluctuations of the individual mass-yields are stronger.

Because of $R(99/115)^{0^+}_{th}=1.00\pm 0.11$, one can expect the mass-yield ratio

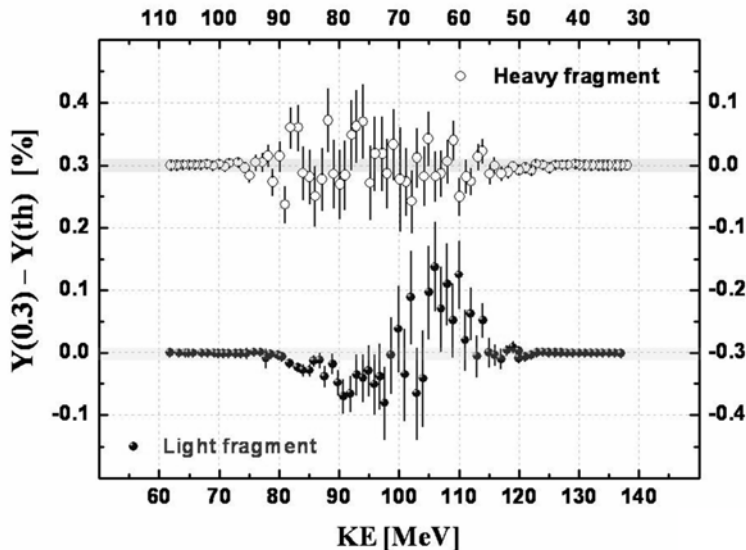


Figure 1b. $Y(0.3)$ KE yields relative to thermal $Y(th)$ ones.

The thermal mass distribution peak-to-valley (P/V) value has been determined to be $(P/V)_{th} = 76 \pm 4$. It is less than the value of $(P/V)_{th} = 114 \pm 2$, reported in [21]. The difference is coming from different characteristics of the sample and experimental setup used, and, probably, from not fully suppressed pile-up between the FF and α

The fluctuations of the ratio and the difference between the mass yield $Y_{0^+}(A)$ from the sum of all resonances with $J^\pi=0^+$, compared to thermal neutron FF yield $Y_{th}(A)$, are small and here $R(99/115)^{0^+}_{th}=1.00\pm 0.11$ (it can be coincidence!). It can be explained if the assumption of the authors of Ref. [30], that $^{239}\text{Pu}(n_{th},f)$ reaction is characterized as following the $(J^\pi, K)=(0^+, 0)$ state at the saddle point in the limit of FF mass formation, is correct.

$R(99/115)^{1^+}_{0^+} = 1.19 \pm 0.11$ not to differ from that of $R(99/115)^{1^+}_{th}$ in their experimental uncertainty limits.

The resonances from both spin groups are forming the FF mass-yield distributions at the neutron energy ranges between them, $Y_{ir}(A)$. The thermal neutron induced FF mass-distribution originate, also from mixed 1^+ and 0^+ states, so, significant differences between $Y_{ir}(A)$ and $Y_{th}(A)$ were neither expected nor found.

pulses. The relative value of the 0.296eV resonance $(P/V)_{0.3}$ to thermal was found to be $(P/V)_{0.3}/(P/V)_{th}=1.74 \pm 0.11$. Despite of the relatively large errors bars, the P/V-ratio changes, as was suggested by Wheeler [31], in the case of resonance neutron induced fission of ^{239}Pu , fluctuate up to a factor of 2-3 for certain resonances. Such a behavior is completely different from that of the fast neutron induced fission, where the P/V-value and $\langle\text{TKE}\rangle$ decrease with increasing E_n .

Variation of fission fragment $\langle\text{TKE}\rangle$ with incident neutron kinetic energy

The energy interval below 1eV was divided into 27 E_n -bins (ΔE_n). Relative to $\langle\text{TKE}\rangle_{th}$, the $\langle\text{TKE}\rangle$ in the intervals was increasing towards the 0.296eV resonance. A similar increase in $\langle\text{TKE}\rangle$ was found by Walsh *et al.* [3].

From the obtained ${}^{exp}\langle\text{TKE}\rangle_{th} = 177.83 \pm 0.02\text{MeV}$ and ${}^{exp}\langle\text{TKE}\rangle_{0.3\text{eV}} = 177.96 \pm 0.01\text{MeV}$, the difference between the experimental $\langle\text{TKE}\rangle$ of the FF from thermal and 0.296eV resonance neutron induced fission was found to be $\Delta{}^{exp}\langle\text{TKE}\rangle_{0.3\text{eV}-th} = 130 \pm 22\text{keV}$. This value is of the same order of magnitude as the measure value of $\Delta{}^{exp}\langle\text{TKE}\rangle_{0.3\text{eV}-0.03\text{eV}} = 185 \pm 75\text{keV}$ [3] and the calculated value of $\Delta{}^{cal}\langle\text{TKE}\rangle_{0.3\text{eV}-th} = 160 \pm 80\text{keV}$ [32].

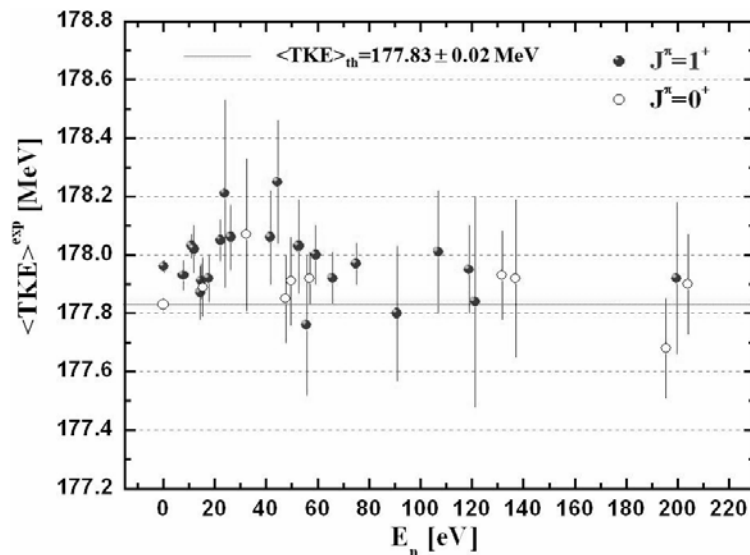


Figure 2. Variation of $\langle\text{TKE}\rangle^{exp}$ at neutron resonance energies.

One can see, from Figure 2, that the fluctuations of FF $\langle\text{TKE}\rangle$ for $J^\pi=1^+$ resonances are stronger around the $E_n \sim 15\text{eV}$ and 40eV resonance clusters. For $J^\pi=0^+$ resonances the variation of $\langle\text{TKE}\rangle$ is around the thermal value $\langle\text{TKE}\rangle_{th} = 177.83 \pm 0.02\text{MeV}$ and in the range of their experimental uncertainties. This behavior of $\langle\text{TKE}\rangle$ is similar to the one observed in the resonance neutron-induced fission of ^{235}U [4] at $E_n \sim 15, 35, 55\text{eV}$, etc., but

less pronounced.

The difference $\Delta\langle\langle\text{TKE}\rangle\rangle_{1-0}$ between $\langle\text{TKE}\rangle_{1+}$ and $\langle\text{TKE}\rangle_{0+}$ for different resonance intervals were found to be $\Delta\langle\langle\text{TKE}\rangle\rangle_{1-0} (E_n = 7-85\text{eV}) = 78 \pm 27\text{keV}$ and $\Delta\langle\langle\text{TKE}\rangle\rangle_{1-0} (E_n = 7-200\text{eV}) = 68 \pm 54\text{keV}$. These values are of the same magnitude as the difference $\Delta\langle\langle\text{TKE}\rangle\rangle_{1-0} (E_n = 7-85\text{eV}) = 50 \pm 90\text{keV}$ of Ref. [14]. The maximum value of the difference $\langle\text{TKE}\rangle_{res} - \langle\text{TKE}\rangle_{th}$ is of the order of $\sim 300-400\text{keV}$, which is of the same order of magnitude as for resonance neutron induced fission of ^{235}U . On the other side, it is only about 30-40% of the $(E_{1+^*} - E_{0+^*}) \sim 1.25\text{MeV}$, available at the 1st saddle point of the CN. This means [3] that either ^{240}Pu is a quite viscous system or somewhere along the fission path some mixing between $K^\pi=0^+$ and $K^\pi=1^+$ fission channels takes place.

Influence of the modes of fission

A quantitative description of the fission process became possible in the frame of the theoretical approach of Brosa et al. [6], combining the multi-modal fission [33] with the random neck-rupture (MM-RNR) model [34].

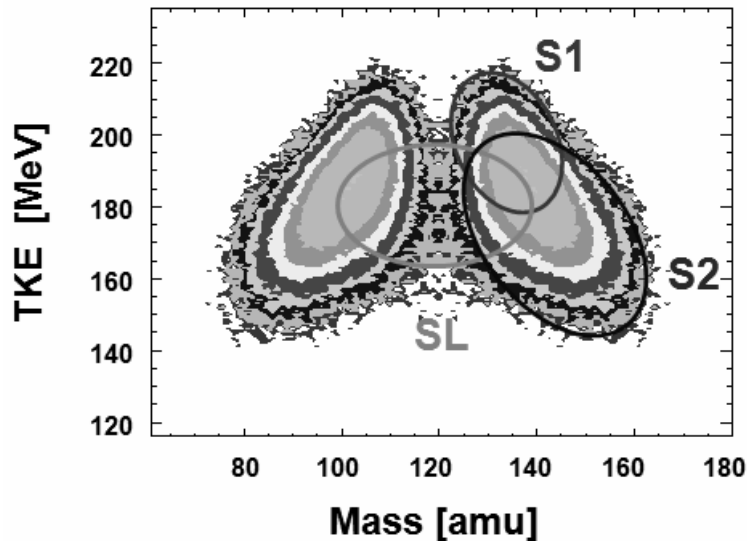


Figure 3. Contour plot of $FF Y(\text{Mass}, \text{TKE})$ from fission with thermal neutrons.

showed that their values from resonance-to-resonance fluctuate slightly and do not differ very much from those of the thermal neutron $Y(A, \text{TKE})$ -distribution. That is why all the distributions were fitted once more, but with all the parameters, except the fission yields, fixed to the thermal values.

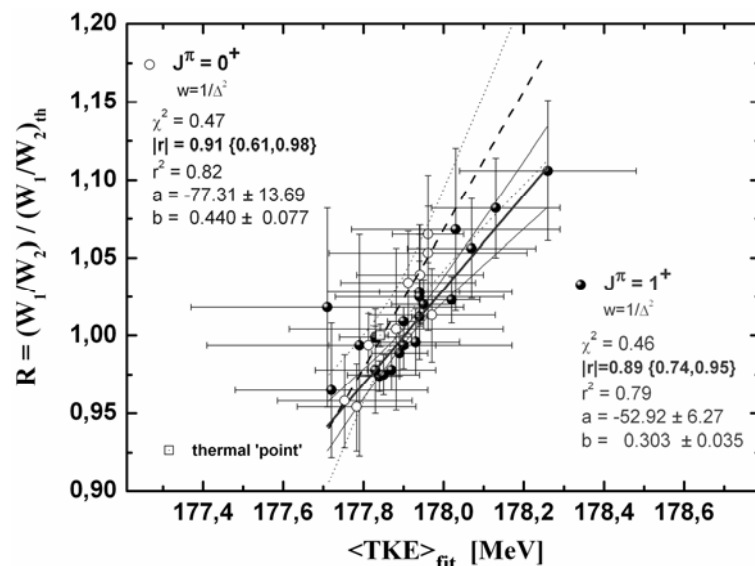


Figure 4. Correlation between the relative main fission mode branching ratios and $FF \langle \text{TKE} \rangle_{\text{fit}}$.

The R -values of all the measured resonances relative to the thermal $R_{\text{th}} = (W_1/W_2)_{\text{th}}$ branching ratio show fluctuation similar to those of

Because $^{239}\text{Pu}(n, f)$ pre-neutron $Y(A, \text{TKE})$ -distributions and their projections $Y(A)$ and $Y(\text{TKE})$ are near identical for the light and heavy fragment peaks, the model was fitted only to the heavy fragment $Y(A, \text{TKE})$ -distributions. The three most important modes were considered - two asymmetric (standard I-S1, standard II-S2) and one symmetric (super-long, SL) (Figure 3).

Plots of the model parameters as a function of the resonance energy E_n

Not only $Y(A)$ and $\text{TKE}(A)$ distributions were compared, but also higher moments, namely, the dispersion $\sigma(\text{TKE})$ and the skewness (dissymmetry) of TKE distributions as a function of A .

The model $\langle \text{TKE} \rangle_{\text{fit}}(E_n)$ values show fluctuations similar to those of the experimental ones $\langle \text{TKE} \rangle_{\text{exp}}(E_n)$. From the two main asymmetric mode areas (probabilities), the “absolute” branching ratio $R_{\text{res}} = (W_1/W_2)_{\text{res}}$ as a function of resonance

$\langle \text{TKE} \rangle_{\text{fit}}(E_n)$. Both, R and $\langle \text{TKE} \rangle$ -fluctuations, amount to about $\sim 10\%$ with some kind of bump-like structure at $E_n \sim 30\text{-}40\text{eV}$ like in Figure 2.

The level of dependence between the obtained FF mass-energy characteristics was determined by calculating Pearson's product moment coefficient $|r|$.

From the similarity between the changes in $\langle \text{TKE} \rangle$ and R from resonance-to-resonance, one can expect that changes in R will invoke corresponding changes in the $\langle \text{TKE} \rangle$. The correlation plot is given in Figure 4. The correlation coefficient is high and the correlation is significant. It means that $\sim 70\text{-}80\%$ of the fluctuations in $\langle \text{TKE} \rangle$ are due to fluctuations in the branching ratio R . A change in the relative branching ratio $dR \sim 10\text{-}15\%$ leads to a change in the mean total kinetic energy $d\langle \text{TKE} \rangle \sim 0.4\text{-}0.5\text{MeV}$.

Influence of the $(n, \gamma f)$ -reaction

Predicted by Lynn [9] the $(n, \gamma f)$ -reaction, as a possible concurrent of the direct fission, was found in the 1^+ resonance neutron induced fission of ^{239}Pu and investigated in detail [10-12]. When it occurs it will cool-down the CN and as a result the mean prompt neutron emission from FF $\langle \nu_p \rangle$ decreases while the γ -ray yield and its multiplicity should, not so remarkable, increases.

The existence of a strong linear dependence of the $\langle \nu_p \rangle$ on $1/\Gamma_f$ is used as a 'test' for the possible existing of the $(n, \gamma f)$ -reaction. An anti-correlation between these two quantities was found to be moderate and significant. The correlations between $\langle \text{TKE} \rangle$ and $1/\Gamma_f$ and $(P/V\text{-ratio and } 1/\Gamma_f)$ were found to be moderate, but insignificant, because of the small number of resonances with relatively small fission widths in this neutron energy region.

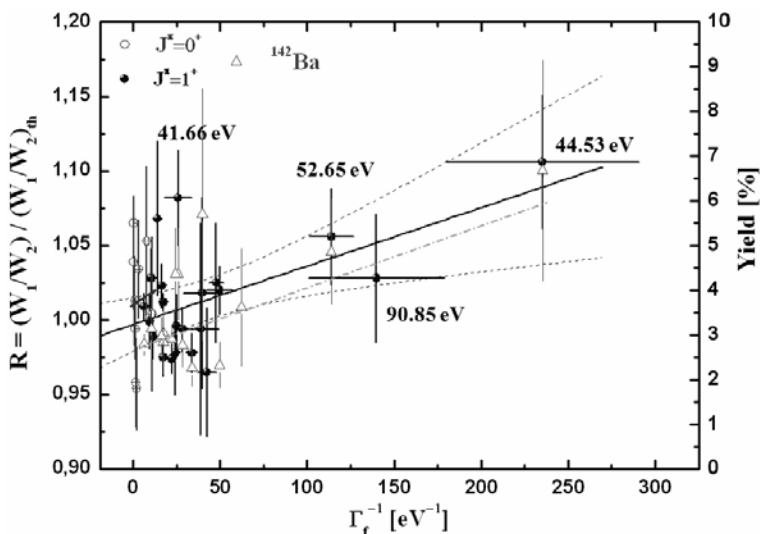


Figure 5. Correlation between the relative main fission mode branching ratios and resonance reverse fission widths.

The same holds for the correlation between the branching ratios R for resonances with $J^\pi=1^+$ and $1/\Gamma_f$. It can be seen in Figure 5, where the relative branching ratio R is plotted together with the independent yields of ^{142}Ba [13, 35]. According the authors of these papers such a behaviour can results from the occurrence of the $(n, \gamma f)$ -reaction. For resonances with $J^\pi=0^+$ there no significant correlation was observed.

Conclusions

In comparison to the resonance neutron induced fission of ^{235}U [4], less pronounced fluctuations in $\langle A \rangle$ and $\langle \text{TKE} \rangle$ distributions for the $^{239}\text{Pu}(n, f)$ reaction were observed. A possible explanation could be, that in the case of resonance neutron

induced fission of ^{239}Pu , for each spin state J^π of the ^{240}Pu CN, only one possible K-channel above the outer barrier is open, whereas for fission of the ^{236}U CN, a mixture of two to three K-channels can take place [36]. If the quantum number K is considered to be a “good” quantum number, i.e. if it is conserved from second saddle to scission, the scission configuration should have the same K. This way, the fission fragment properties, for a given fission mode and K quantum number should be “fixed”. Hence, the superposition of different transition states with different K-quantum number and thus, different fission fragment property distributions can result in fluctuations from resonance-to-resonance. In case of ^{240}Pu CN, with only one single transition state, such fluctuations should be absent or be less pronounced, as it was observed.

The influence of Bohr’s channel spin ($J^\pi=1^+$ and $J^\pi=0^+$) on $\langle\text{TKE}\rangle$ was found to be small: $\Delta\langle\text{TKE}\rangle = \langle\text{TKE}\rangle_{1^+} - \langle\text{TKE}\rangle_{0^+} = 0.068 \pm 0.054\text{MeV}$, which is of the same order of magnitude as given in Ref. [14].

The correlation of $\langle\text{TKE}\rangle$ and R with $1/\Gamma_f$ and anti-correlation with $\langle\nu_p\rangle$ were found to be moderate or low, but insignificant, because they are based on 2-3 resonances with relatively small fission widths and large experimental uncertainties.

By the occurrence of an (n, γ f)-reaction in the 1st minimum of the fission barrier one can explain the relatively small fluctuations in the primary FF characteristics from resonances with $J^\pi=1^+$ and absence of pronounced fluctuations for $J^\pi=0^+$.

The existence of a β -vibration state [30] at $\sim 3\text{MeV}$ below the outer saddle, as well as the decaying of the shape isomer through a ($J^\pi K=0^+0$) fission channel, pick-up the question about the existence of the (n, γ f)-reaction in the IInd well of the double-humped fission barrier, too. Such a hypothesis can be indirectly supported by the existence of two energy groups of γ -rays accompanying the resonance neutron induced fission of ^{239}Pu [10,11].

Despite of not so significant correlations, the understanding of the fluctuations in the FF characteristics from the resonance neutron induced fission of ^{239}Pu are of great importance for evaluations, especially those of the prompt neutron multiplicity $\langle\nu_p\rangle$ and/or the delayed neutron (DN) yields [37]. The latter is supposed to fluctuate from resonance-to-resonance, because the precursors of the DN are lying in the range where some interplay between fission modes can take place.

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