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Prompt fission neutron multiplicity and spectrum evaluation for $^{235}\text{U}(n, f)$ in the frame of the multi-modal fission model

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Abstract

The multi-modal fission approach, in the frame of an improved version of the Los Alamos model, is used for the evaluation of the prompt fission neutron multiplicity and spectra for the neutron-induced fission of ^{235}U .

The three most dominant fission modes (standard I, standard II and superlong) are taken into account. The multi-modal parameters entering the multiplicity and spectrum model are determined on the basis of experimental data measured at IRMM.

The total prompt fission neutron multiplicity and spectra, both obtained as superposition of the modal contributions are in good agreement with the experimental data. Also, the average prompt gamma-ray energy for each fission mode is obtained and the total prompt gamma-ray energy is in good agreement with the existing experimental data, too.

The agreement of the prompt neutron spectrum with the experimental data is improved especially in the lower fission neutron energy region of the spectrum when the neutron emission anisotropy effect is taken into account.

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1. Introduction

The prompt fission neutron multiplicity and energy spectra of actinides are nuclear data of crucial importance for energy and non-energy nuclear applications. For this reason, new evaluations of these quantities with a higher degree of accuracy are required. Especially, for ^{235}U , being the standard nucleus for evaluated nuclear data files, an improved evaluation of the prompt fission neutron multiplicity and spectra, based on recent nuclear models, which incorporate more physical effects in a consistent way, is welcome.

In this work, an attempt to improve the evaluation of the prompt fission neutron multiplicity and spectra for $^{235}\text{U}(n, f)$ is made, including for the first time the concept of multi-modality of the fission process.

So far, for the prompt fission neutron spectrum and multiplicity evaluations of $n + ^{235}\text{U}$, the Madland–Nix model, usually named *Los Alamos (LA) model* [1,2], has been used. This model takes into account the following physical effects: (a) the distribution of the fission fragment (FF) excitation energy, (b) the energy dependence of the inverse process compound nucleus (CN) formation cross-section, (c) the center-of-mass motion of the fission fragments (FF), and (d) the multiple fission chances at high incident neutron energies. In the original model, applied first for $^{235}\text{U}(n, f)$, only the most probable fragmentation {Sr-96, Xe-140} was taken into account. The model parameters (e.g., the average energy released in fission, the average FF neutron separation energy, etc.) were calculated using the “7-point approximation” and the level density parameter was taken into account as $a = A_{\text{CN}}/C$, with $C = 11 \text{ MeV}^{-1}$ (where A_{CN} is the mass number of the CN undergoing fission). The same approach was used by D. Madland to obtain the $(n + ^{235}\text{U})$ fission neutron spectrum, included in the latest version of the ENDF/B-VI library [3].

In the present work, the multi-modality of the fission process is incorporated into an improved version of the LA model and applied for the prompt fission neutron spectra and multiplicity evaluation of $^{235}\text{U}(n, f)$. This is a continuation of the already successful application of the model to the evaluation of the prompt fission neutron spectra and multiplicities for $^{238}\text{U}(n, f)$ and $^{237}\text{Np}(n, f)$ up to the second chance fission threshold [4]. The three most important fission modes, two asymmetric modes named Standard I (S1) and Standard II (S2), and one symmetric mode named superlong (SL), are taken into account.

We would like to stress, however, that only the idea of multi-modal fission is taken into account. The modal parameters entering the calculation are based purely on experimental results and do not depend on which kind of theoretical model being used for the potential energy calculation and determination of the fission modes.

2. Basic features of the model

In the frame of the multi-modal fission approach, the prompt fission neutron spectrum $N_{\text{tot}}(E)$ and multiplicity ν_p at a given incident neutron energy E_n are calculated as superposition of the spectra and multiplicities associated with a particular fission mode [4]:

$$N_{\text{tot}}(E) = \frac{\sum_m w_m \langle v \rangle_m N_m(E)}{\sum_m w_m \langle v \rangle_m}, \quad (1)$$

$$\langle v \rangle_{\text{tot}} = \frac{\sum_m w_m \langle v \rangle_m}{\sum_m w_m}, \quad (2)$$

with each sum running over all fission modes involved.

In Eqs. (1) and (2), w_m is the branching ratio of mode m .

The quantities $N_m(E)$ (with E the energy of the emitted prompt neutron) and $\langle v \rangle_m$ are, respectively, the prompt fission neutron spectrum and multiplicity corresponding to the mode m :

$$N_m(E) = \frac{1}{2} (N_m^L(E, E_f^L, \sigma_{c,L}) + N_m^H(E, E_f^H, \sigma_{c,H})) \quad (3)$$

where $N_m^L(E, E_f^L, \sigma_{c,L})$ and $N_m^H(E, E_f^H, \sigma_{c,H})$ are the individual light (LF) and heavy fragment (HF) spectra of a mode m , respectively. Because the number of prompt neutrons emitted by the LF and HF is only experimentally known in the case of $^{235}\text{U}(\text{n}_{\text{th}}, \text{f})$ we adopted the usual assumption that an equal number of neutrons is emitted by the LF and HF. The individual FF spectra are calculated in the frame of the LA model with the energy dependence of the CN cross-section formation as described in Refs. [1,4,5].

So far, the assumption that neutron emission is only possible from fully accelerated fragments has been used. In recent years, the question whether neutron evaporation from accelerating fragments is possible or not has been raised again (see, e.g., Ref. [6] and references therein). This has, of course, consequences on the angular distribution of the emitted neutrons. Neutron emission during fragment acceleration would result in a non-isotropic neutron spectrum in the center-of-mass system (CMS).

If the anisotropy of the prompt fission neutron emission is taken into account, the expression, giving the spectrum of an individual FF, is modified as follows:

According to Terrel [7], it can be assumed that the anisotropy of the neutron emission, if present, is symmetrical about 90° and the FF prompt neutron spectrum in the CMS could be described by the following equation:

$$\Phi(\varepsilon, \theta_{\text{cm}}) = \Phi(\varepsilon) \frac{1 + b \cos^2 \theta_{\text{cm}}}{1 + b/3}, \quad (4.1)$$

where $\Phi(\varepsilon)$ is the center-of-mass fission neutron spectrum from Refs. [1,4,5] and b is the anisotropy parameter. Consequently, the relation giving the prompt fission neutron spectrum, in the laboratory system (LS), for a given fission fragment f , becomes the following:

$$\begin{aligned} N(E, E_f, \sigma_{\text{cf}}) &= \frac{1}{2T_m^2 \sqrt{E_f}} \\ &\times \int_{(\sqrt{E}-\sqrt{E_f})^2}^{(\sqrt{E}+\sqrt{E_f})^2} \sigma_{\text{cf}}(\varepsilon) \sqrt{\varepsilon} \left(\frac{1}{1+b/3} + \frac{b(E-\varepsilon-E_f)^2}{4\varepsilon E_f(1+b/3)} \right) I(\varepsilon) d\varepsilon \end{aligned} \quad (4.2)$$

with

$$I(\varepsilon) = \int_0^{T_m} k_f(T) T \exp(-\varepsilon/T) dT \quad \text{and} \quad k_f(T) = \left[\int_0^\infty \sigma_{cf}(\varepsilon) \varepsilon \exp(-\varepsilon/T) d\varepsilon \right]^{-1},$$

where E is the prompt fission neutron energy in the LS, E_f is the FF average kinetic energy per nucleon, σ_{cf} is the CN cross-section of the inverse process, and T_m is the maximum of the triangular FF residual nuclear temperature distribution.

The average prompt neutron multiplicity of each fission mode m is calculated using the following relation (obtained from energy conservation):

$$\langle \nu \rangle_m = \frac{\langle E_r \rangle_m + E_n + B_n - \langle TKE \rangle_m - \langle E_\gamma \rangle_m}{\langle S_n \rangle_m + \langle \varepsilon \rangle_m}, \quad (5)$$

where B_n is the neutron binding energy in the CN undergoing fission, $\langle E_r \rangle_m$ and $\langle TKE \rangle_m$ are the average energy released in fission and the average FF total kinetic energy for the mode m , respectively. $\langle S_n \rangle_m$ is the average neutron separation energy of the fission fragments for a given mode m , and $\langle \varepsilon \rangle_m$ is the first order momentum of the CMS spectrum of each fission mode calculated as in Ref. [4].

According to Ref. [8], a linear dependence of the average prompt gamma ray energy as a function of prompt neutron multiplicity (with the same slope and intercept for all fission modes) is taken into account:

$$\langle E_\gamma \rangle_m = p \langle \nu \rangle_m + q. \quad (6)$$

Consequently, Eq. (5), giving the prompt neutron multiplicity of mode m , can be expressed as:

$$\langle \nu \rangle_m = \frac{\langle E_r \rangle_m + E_n + B_n - \langle TKE \rangle_m - q}{\langle S_n \rangle_m + \langle \varepsilon \rangle_m + p}. \quad (7)$$

3. Multi-modal parameters entering the multiplicity and spectrum model

The multi-modal parameters entering the multiplicity and spectrum model were calculated for $^{235}\text{U}(n, f)$ at incident neutron energies from thermal up to 5.5 MeV (the upper limit in energy, where only the first fission chance is involved).

For the determination of the input model parameters the following data are needed:

- Experimental data for the two-dimensional mass yield versus total kinetic energy (TKE) distribution, $Y(A, TKE)$ measured at IRMM [9].
- Multi-modal representation of the former data taking into account the most important fission modes mentioned above: two asymmetric modes S1 and S2, and one symmetric mode SL, concerning the following quantities:
 - the multi-modal branching ratios w_m ,
 - the average total kinetic energy of each mode $\langle TKE \rangle_m$,
 - the average FF mass of each mode $\langle A \rangle_m$,
 - the standard deviation of the FF mass distribution of each mode σ_{Am} .

The behavior of the modal branching ratios versus E_n is given in Fig. 1.

The average FF masses and their standard deviations for the S1 and S2 fission modes are given in Table 1. Since the contribution of the SL mode is very small, the average FF mass was fixed as $\langle A \rangle_{SL} = 118$ as well as the standard deviation of the FF mass distribution as $\sigma_{A,SL} = 15$ at all incident neutron energies where measurements were made.

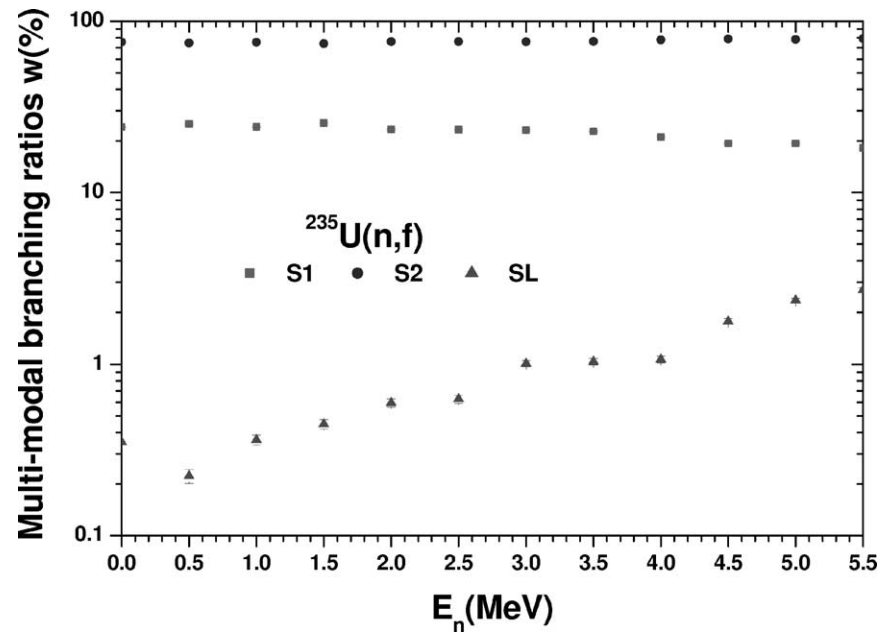


Fig. 1. Multi-modal branching ratios as a function of incident neutron energy E_n .

Table 1
 $^{235}\text{U}(n, f)$ IRMM modal data-set

E_n (MeV)	Average HF mass of the standard modes, $\langle A_H \rangle_m$		$\sigma_{A,m}$	
	S1	S2	S1	S2
0.0	134.4186 ± 0.0033	140.8853 ± 0.0031	3.1326	5.1109
0.5	134.5550 ± 0.0268	141.0413 ± 0.0260	3.0806	5.1471
1.0	134.5531 ± 0.0250	141.0610 ± 0.0237	3.0849	5.1658
1.5	134.5709 ± 0.0267	141.1831 ± 0.0252	3.1999	5.1522
2.0	134.6107 ± 0.0274	140.8607 ± 0.0256	3.1834	5.3786
2.5	134.6282 ± 0.0288	140.9112 ± 0.0269	3.2127	5.4115
3.0	134.5564 ± 0.0305	140.8807 ± 0.0281	3.2964	5.4849
3.5	134.5176 ± 0.0300	140.7175 ± 0.0274	3.3163	5.5571
4.0	134.3321 ± 0.0323	140.4829 ± 0.0293	3.2400	5.6553
4.5	134.4695 ± 0.0370	140.4049 ± 0.0323	3.3124	5.8294
5.0	134.3608 ± 0.0373	140.4066 ± 0.0309	3.6438	6.0584
5.5	134.4411 ± 0.0390	140.2956 ± 0.0324	3.4739	5.9995

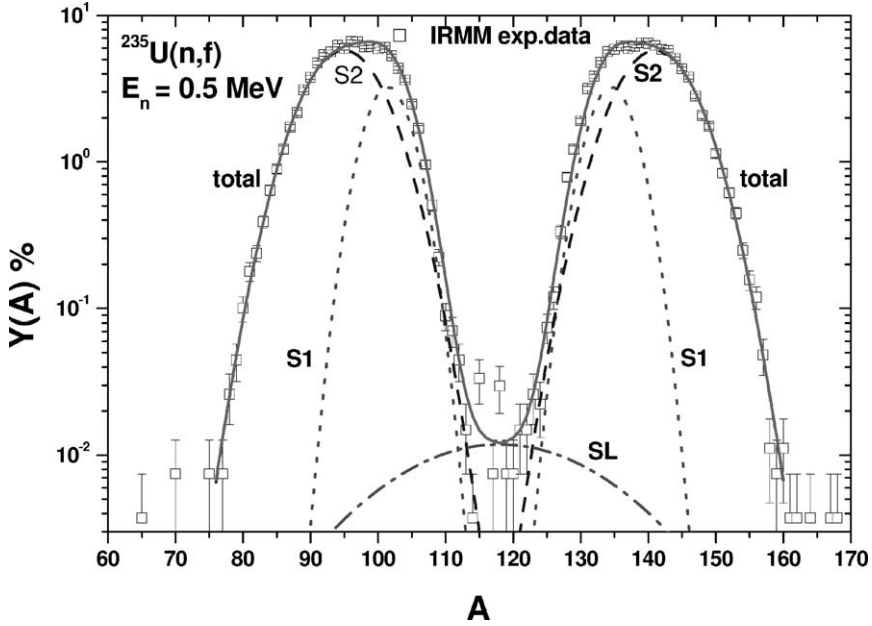


Fig. 2. Multi-modal and total FF mass distribution at $E_n = 0.5$ MeV in comparison with the experimental data.

3.1. FF mass distribution of each mode

The fragment mass distribution, for each fission mode m , is the first quantity necessary to determine the model parameters. As usual, it is described by a superposition of Gaussian functions:

$$Y_m(A) = \frac{w_m}{\sigma_{A,m}\sqrt{2\pi}} \exp\left(-\frac{(A - \bar{A}_m)^2}{2\sigma_{A,m}^2}\right). \quad (8)$$

The sum over all fission modes has to reproduce the experimental FF mass distribution:

$$Y(A) = \sum_m Y_m(A). \quad (9)$$

Fig. 2 shows, for example, the fitted FF mass distribution of each mode and the sum in comparison with the experimental data at $E_n = 0.5$ MeV.

3.2. Range of FF pairs

In the present work, the entire experimental FF mass range ($A_L \in [76, 118]$ and $A_H \in [118, 160]$) is taken into account, totalling 43 mass pairs.

For each FF mass pair $\{A_{L,i}, A_{H,i}\}$ two isobars per mass are taken into account with values of the nuclear charge Z that are the nearest integer values above and below the most probable charge Z_p . Consequently, 86 FF pairs are taken into account in the present calculations.

3.3. Average energy released for each fission mode

The average energy released for each fission mode m is calculated using the following relation:

$$\langle E_r \rangle_m = \frac{\sum_i Y_{m,i} p z_i E_{ri}}{\sum_i Y_{m,i} p z_i}, \quad (10)$$

with m the index of the mode (here S1, S2 and SL) and i the range of FF pairs. $Y_{m,i}$ is the FF mass yield for mode m and FF pair i . $p z_i$ is the charge distribution corresponding to the FF pair i , approximated by a Gaussian function [2,4,10]:

$$p z_i = \frac{1}{\sqrt{\pi} c} \exp\left(-\frac{(Z_i - Z_{p,i})^2}{c}\right) \quad \text{with } c = 2\left(\sigma^2 + \frac{1}{12}\right), \quad (11)$$

with Z_i the charge number of the heavy (or light) fragment of the i th FF pair, and the most probable charge $Z_{p,i}$ obtained using Z_{UCD} (unchanged charge distribution) and the charge polarization value 0.25.

The energy released of each binary fission considered is given by:

$$E_{r,i} = \Delta(Z_{CN}, A_{CN}) - \Delta(Z_{L,i}, A_{L,i}) - \Delta(Z_{H,i}, A_{H,i}) \quad (12)$$

where Δ is the mass excess expressed in MeV (taken from the Audi nuclear data library [11]).

The behavior of $\langle E_r \rangle_m$ versus E_n is shown in Fig. 3(a).

3.4. Average level density parameter of each fission mode

The average value of the level density parameter for each fission mode m is calculated using the following expression:

$$\langle a \rangle_m = \frac{\sum_i Y_{m,i} p z_i a_i}{\sum_i Y_{m,i} p z_i}, \quad (13)$$

with the sums running over all FF pairs. According to the LA model, the input model parameter is:

$$\langle C \rangle_m = \frac{A_{CN}}{\langle a \rangle_m} \quad (14)$$

where A_{CN} is the mass number of the CN undergoing fission. For each FF pair $\{Z_{L,i}, A_{L,i}, Z_{H,i}, A_{H,i}\}$ the level density parameter

$$a_i = a_{L,i} + a_{H,i} \quad (15)$$

is calculated using the Ignatiuk super-fluid model [4,12] with the shell corrections, entering the level-density parameter formulae, taken from the Möller and Nix nuclear data library [13].

The average level-density parameters for the three fission modes involved are given in Fig. 3(b).

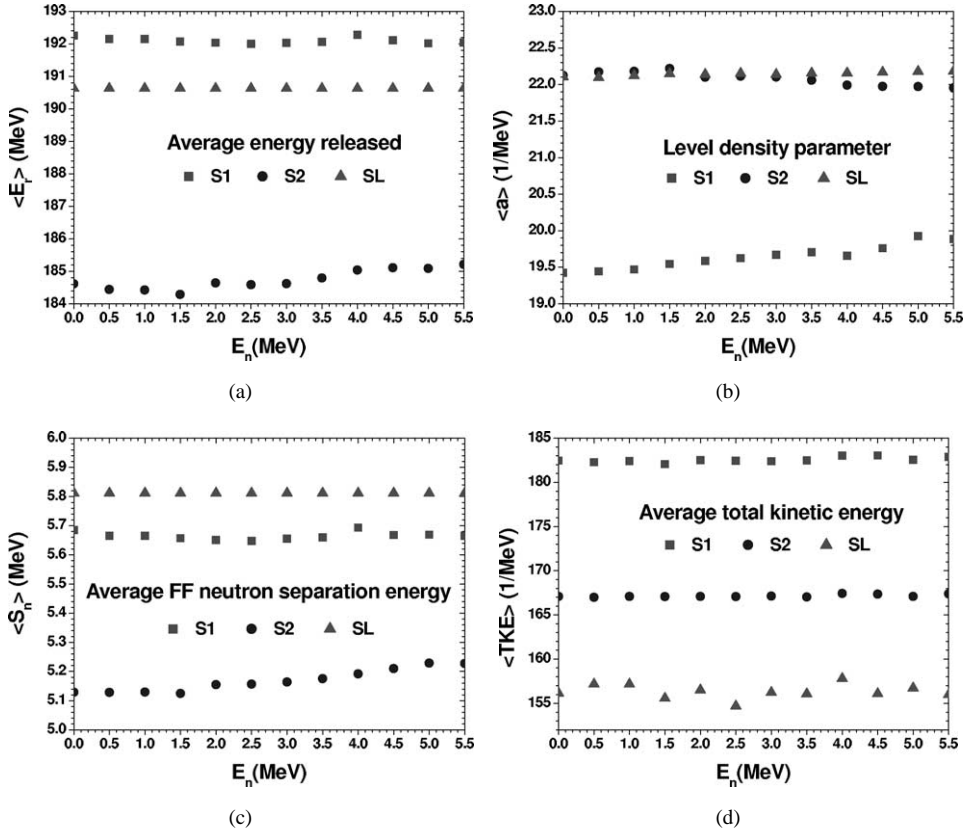


Fig. 3. (a) The average energies released in fission for each fission mode, (b) the average level density parameter of each fission mode, (c) the FF multi-modal average neutron separation energies, and (d) the FF multi-modal average total kinetic energies.

3.5. Average FF neutron separation energy for each fission mode

The average FF neutron separation energy for each fission mode m is calculated according to the following equation [4]:

$$\langle S_n \rangle_m = \frac{\sum_i Y_{m,i} p z_i \overline{S_{n,i}}}{\sum_i Y_{m,i} p z_i}, \quad (16)$$

where the average neutron separation energy of each FF pair i is given by:

$$\overline{S_{n,i}} = \frac{1}{2} (\overline{S_{n,i}^L} + \overline{S_{n,i}^H}), \quad \overline{S_{n,i}^{L,H}} = \frac{1}{2} \left(S_{n,i}^{L,H} + \frac{S_{2n,i}^{L,H}}{2} \right), \quad (17)$$

and S_n and S_{2n} are calculated using the Audi nuclear data library [11]. The inclusion of S_{2n} is done to remove any pairing effects on the value of $\langle S_n \rangle_m$.

The behavior of $\langle S_n \rangle_m$ as function of E_n is shown in Fig. 3(c).

3.6. Average prompt gamma-ray energy for each mode

According to Refs. [4,14] the prompt γ -ray energy parameterization versus the prompt neutron multiplicity depends only on Z_{CN} and A_{CN} :

$$p = 6.71 - 0.156 \frac{Z_{\text{CN}}^2}{A_{\text{CN}}}, \quad q = 0.750 + 0.088 \frac{Z_{\text{CN}}^2}{A_{\text{CN}}}. \quad (18)$$

3.7. Multi-modal average total kinetic energy of the FF

The FF average total kinetic energies of S1, S2 and SL fission modes, obtained by fitting the experimental $Y(A, TKE)$ distribution are given in Fig. 3(d) as a function of E_n .

3.8. Most probable fragmentation of each fission mode

The most probable fragmentation of each fission mode is the following:

Standard I	Zr-102	Te-134,
Standard II	Sr-95	Xe-141,
Super-long	Pd-118	Pd-118.

For these nuclei optical model calculations of the CN cross-section of the inverse process are made.

4. Equivalent single-mode input model parameters

The parameters entering the prompt fission neutron multiplicity and spectrum model can be reduced to single-mode values, according to the following relations:

$$\begin{aligned} \langle E_r \rangle &= \frac{\sum_m w_m \langle E_r \rangle_m}{\sum_m w_m}, & \langle TKE \rangle &= \frac{\sum_m w_m \langle TKE \rangle_m}{\sum_m w_m}, \\ \langle a \rangle &= \frac{\sum_m w_m \langle a \rangle_m}{\sum_m w_m}, & \langle S_n \rangle &= \frac{\sum_m w_m \langle S_n \rangle_m}{\sum_m w_m}. \end{aligned} \quad (19)$$

The E_n -dependence of the equivalent single-mode average values of the above model parameters is shown in Figs. 4(a)–(d). Using the same procedure the equivalent single-mode most probable fragmentation in the case of the $^{235}\text{U}(n, f)$ process is found to be {Sr-97, Xe-139}.

5. Results and discussions

The calculations presented in this work were performed with the following computer codes:

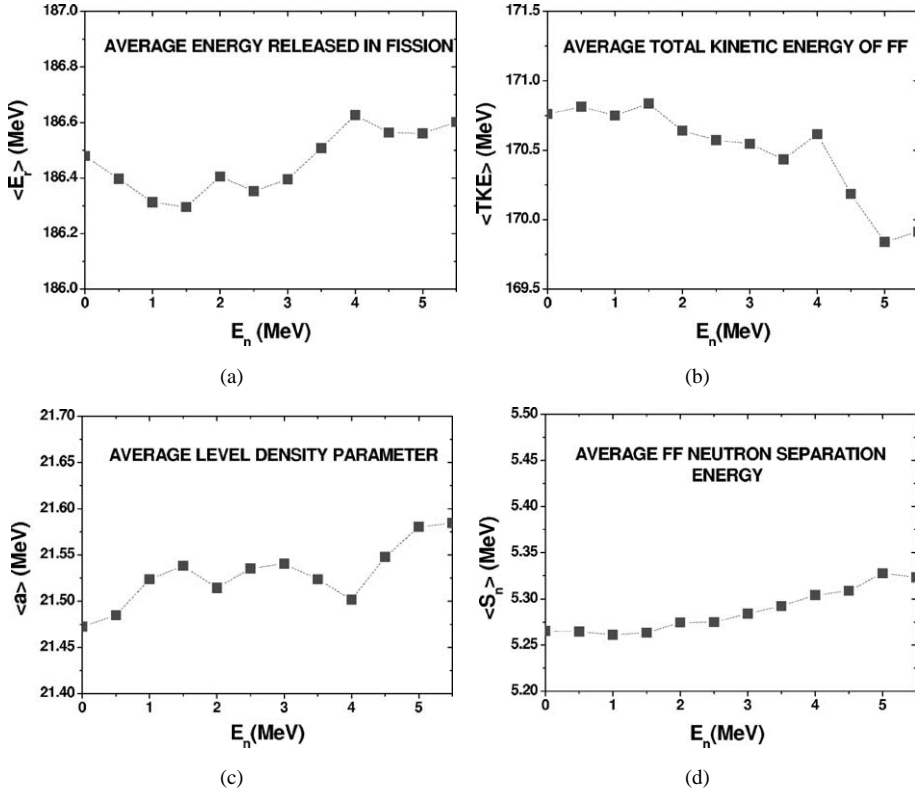
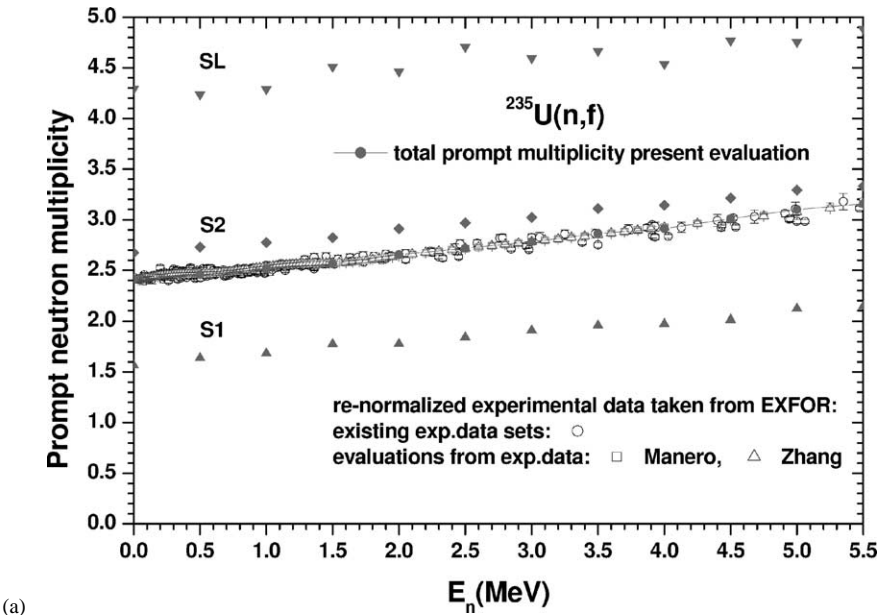


Fig. 4. Equivalent single-mode input model parameters versus E_n : (a) the average energy released in fission, (b) the average FF total kinetic energy, (c) the average level density parameter, (d) the average neutron separation energy.

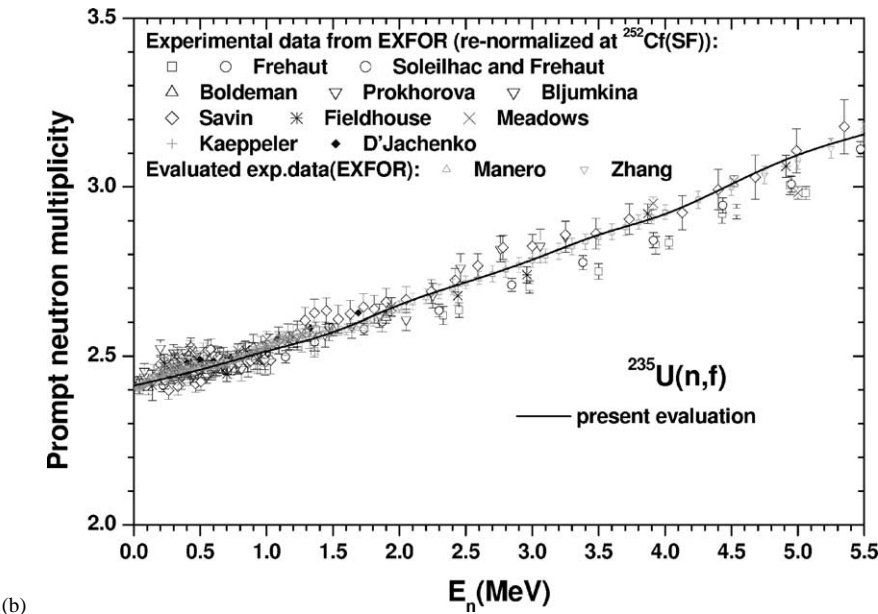
- The **SPECMOD code** was used for prompt fission neutron multiplicity and spectra calculation. It is an improved version of the code SPECTRUM [5] containing the model as described in Refs. [1,2,4] and in the second section of this work;
- The **MODPAR code** was used for multi-modal input model parameter calculation. This code is working with a part of the nuclear data bases contained in the RIPL nuclear data library [11,13] and it uses as main input the experimental TKE(A)- and Y(A)-data as well as the modal values for w_m , $\langle A \rangle_m$, and $\sigma_{A,m}$;
- The **SCAT2 optical model code** [15] with the Becchetti–Greenless optical potential was used for the CN cross-section of the inverse process calculation.

The results obtained in this work can be summarized in the following items:

- (1) The total average prompt fission neutron multiplicity is obtained in good agreement with the existing experimental data taken from the EXFOR nuclear data library [16] and renormalized to the $^{252}\text{Cf}(\text{SF})$ prompt neutron multiplicity standard value (used as a reference). This is illustrated in Fig. 5(a), where the contribution of the S1, S2 and



(a)



(b)

Fig. 5. (a) Multi-modal, and (b) total average prompt fission neutron multiplicity calculations in comparison with experimental data (taken from the EXFOR library and renormalised to the standard value of $^{252}\text{Cf}(\text{SF})$ used as a reference).

- SL modal multiplicities is plotted and in Fig. 5(b), where a zoomed view of the total prompt multiplicity is given.
- (2) The total average prompt gamma-ray energy is also obtained in good agreement with the experimental data of Fréhaut [8], as can be seen in Fig. 6 (where the prompt gamma-ray contribution of each fission mode is plotted as well).
 - (3) The prompt fission neutron energy spectra are obtained in good agreement with the experimental data (taken from the EXFOR nuclear data library [17]) for the thermal incident neutron energy (Fig. 7(a)) and for $E_n = 0.5$ MeV (Fig. 8(a)). In order to emphasize this agreement Figs. 7(b) and 8(b) present the calculated spectra and experimental data as ratio to the ENDF/B-VI evaluation [3] and, respectively, to a single-mode calculation (taken from Ref. [14]).
 - (4) The prompt fission neutron spectrum calculations at other incident neutron energies, where experimental data exist (in the EXFOR library [17]), show a reasonable agreement, as can be seen in Figs. 9(a)–(c).
 - (5) The present prompt neutron spectra calculations are close to the ENDF/B-VI (MF = 5, MT = 18) evaluation, as can be seen in Fig. 7(b).
 - (6) Especially, in the lower part of the fission neutron energy range, the available experimental data tend to be higher than the evaluations made using models based on the assumption that the fission neutrons are emitted by evaporation from the fully accelerated FF without anisotropy taken into account. A possibility to improve the agreement of the evaluation with the experimental data is to take into account the anisotropy effect [7,18–22].

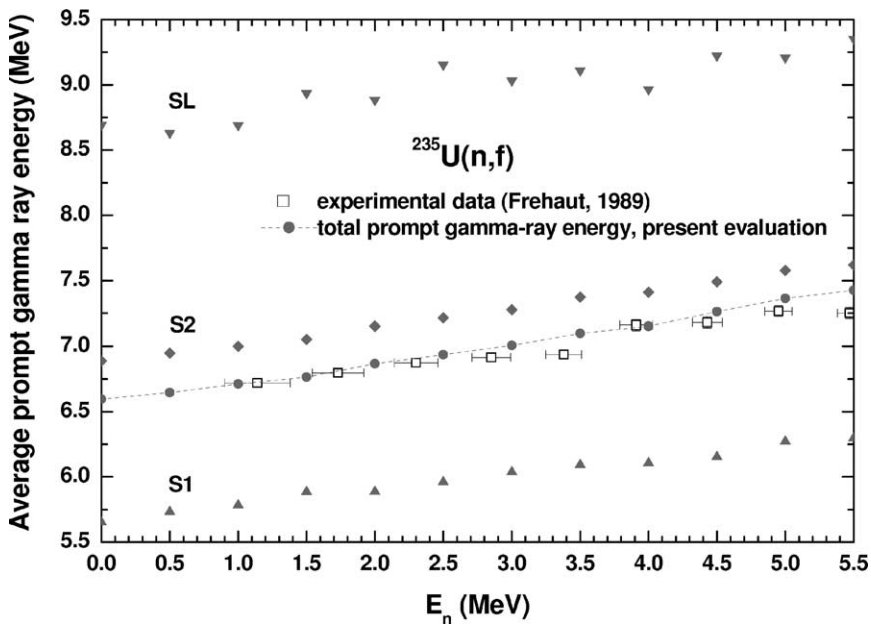


Fig. 6. Multi-modal and total average prompt gamma-ray energies in comparison with the experimental data of Fréhaut [8].

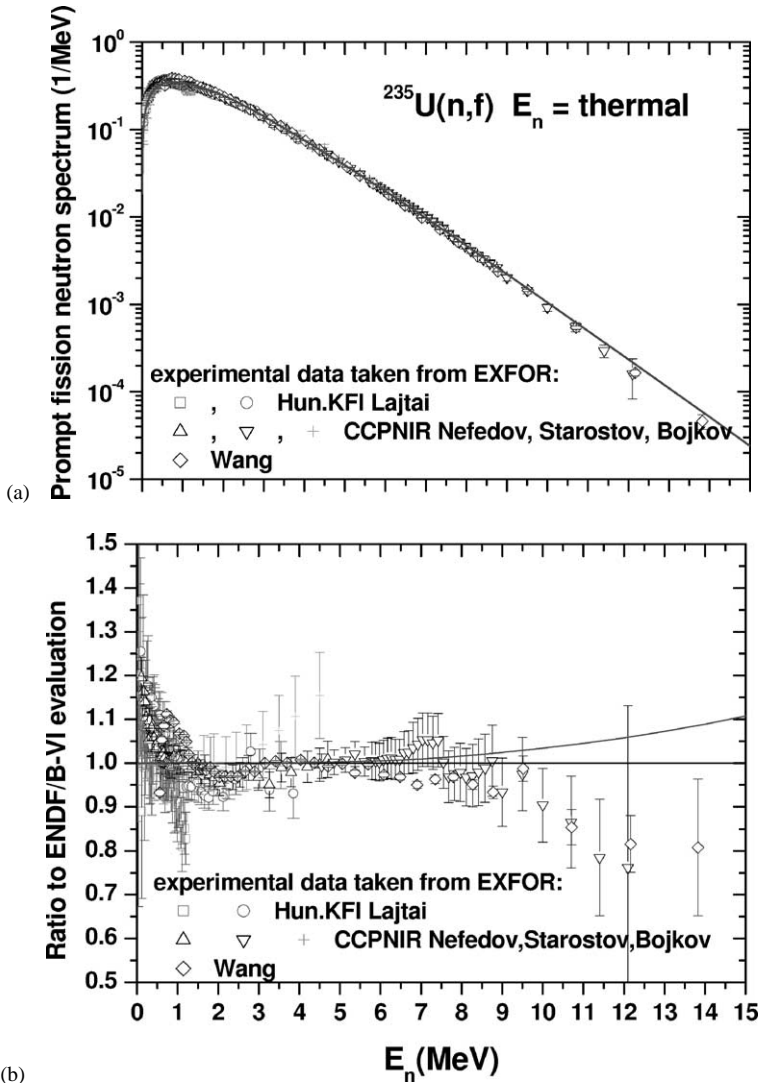


Fig. 7. Multi-modal prompt fission neutron spectrum calculations at thermal incident neutron energy in comparison with the experimental data (taken from EXFOR library): (a) usual presentation, and (b) presented as ratios to the ENDF/B-VI evaluation.

In order to do so, equivalent single-mode calculations are done using the model parameter values from Eq. (19), since we do not know the anisotropy parameter of each mode. The spectrum calculations, considering an anisotropy effect, led to the following results:

As can be seen in Fig. 10 at thermal incident neutron energy, the agreement with the experimental data is considerably increased, especially, in the lower fission neutron energy

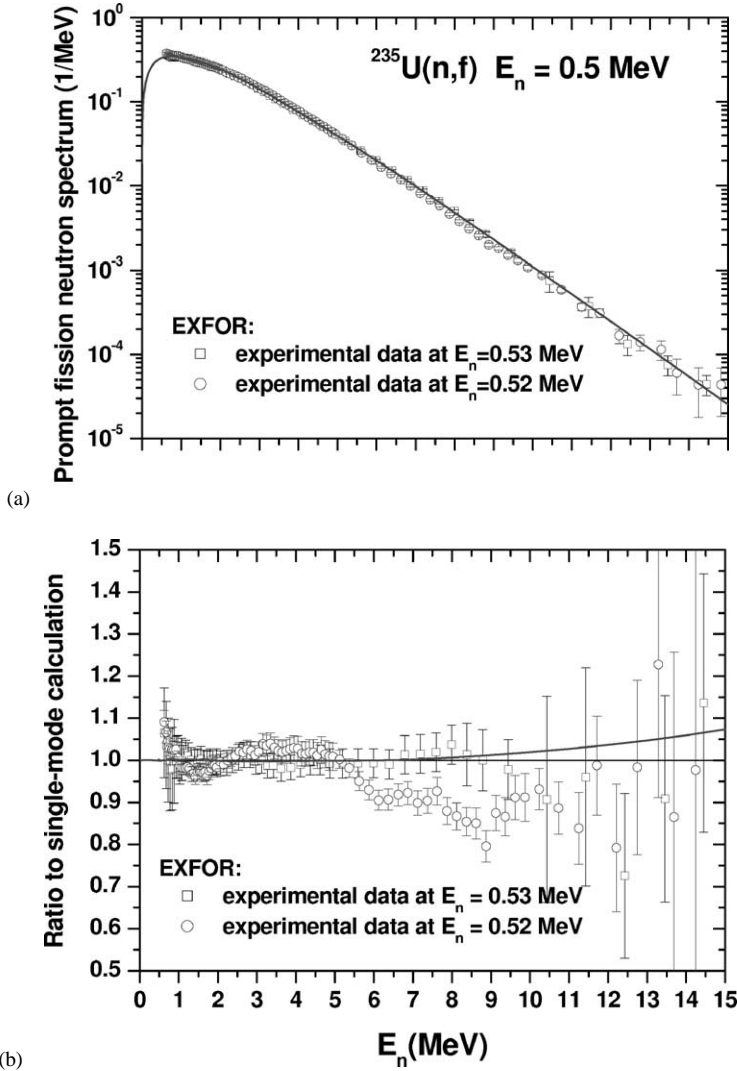


Fig. 8. Multi-modal prompt fission neutron spectrum calculations at $E_n = 0.5$ MeV in comparison with the experimental data (taken from EXFOR library): (a) usual presentation, and (b) presented as ratios to the single-mode calculation from Ref. [14].

region of the spectrum using an anisotropy parameter $b = 0.2$. The same considerably improved agreement with experimental data is obtained at $E_n = 0.5$ MeV (see Fig. 11).

The value of the anisotropy parameter b was actually determined by a fit of the experimental neutron spectra at thermal and 0.5 MeV incident neutron energy.

To our knowledge no prompt fission neutron spectrum evaluations in the frame of the LA model taking into account the anisotropy effect are done for $^{235}\text{U}(n,f)$. Some results considering the fission neutron anisotropy are given by Terrel [7] who used a simplified

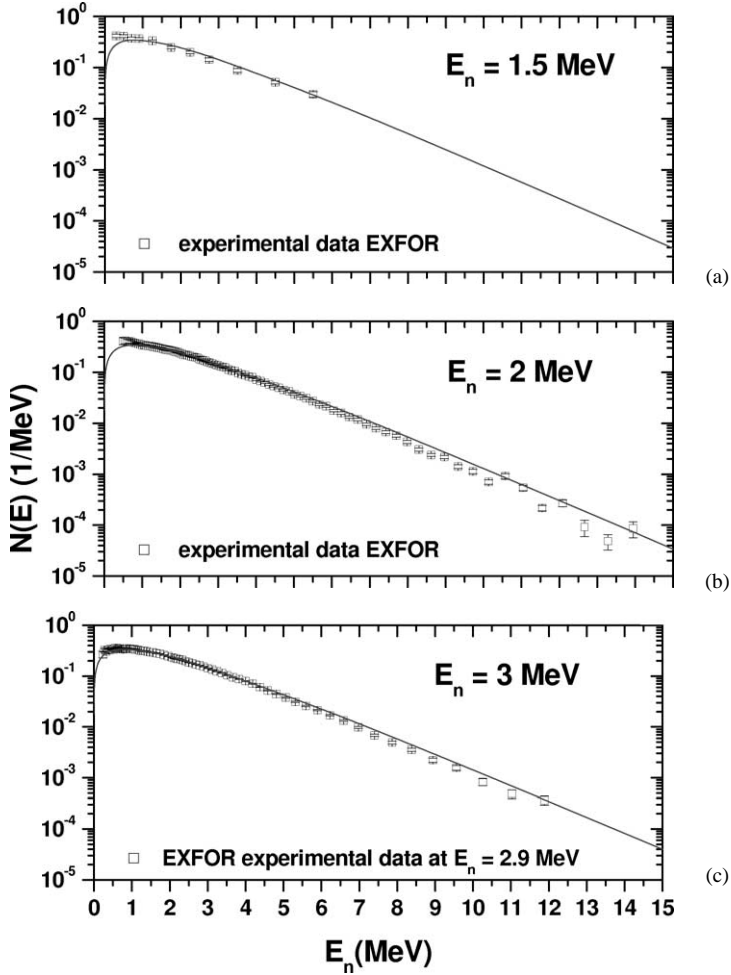


Fig. 9. Multi-modal prompt neutron spectrum calculation at: (a) $E_n = 1.5 \text{ MeV}$, (b) $E_n = 2 \text{ MeV}$, (c) $E_n = 3 \text{ MeV}$, in comparison with the existing experimental data (taken from the EXFOR library).

prompt fission neutron model (also based on neutron evaporation from the FF) before the elaboration of the LA model [1].

6. Conclusions

- For the first time the multi-modal approach (with modal parameters experimentally determined at IRMM) has been applied to the prompt fission neutron energy spectra and multiplicity evaluation of $^{235}\text{U}(n, f)$ using an improved version of the LA model and a dedicated procedure for the determination of the input model parameters ($\langle E_r \rangle_m$, $\langle a \rangle_m$, $\langle S_n \rangle_m$ and $\langle E_\gamma \rangle_m$).

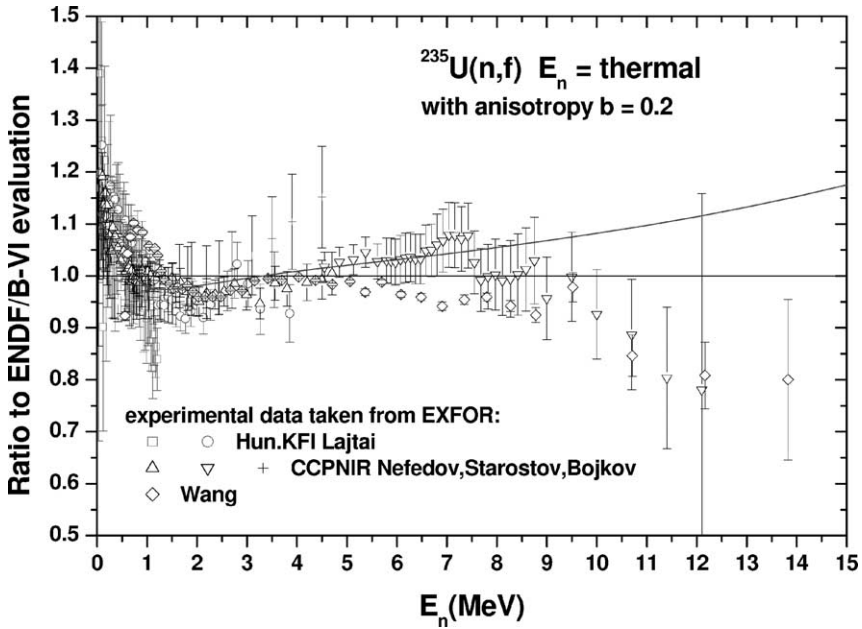


Fig. 10. Prompt fission neutron spectrum with the anisotropy effect taken into account at thermal incident neutron energy and presented as ratio to the ENDF/B-VI evaluation, in comparison with the experimental data (taken from the EXFOR library).

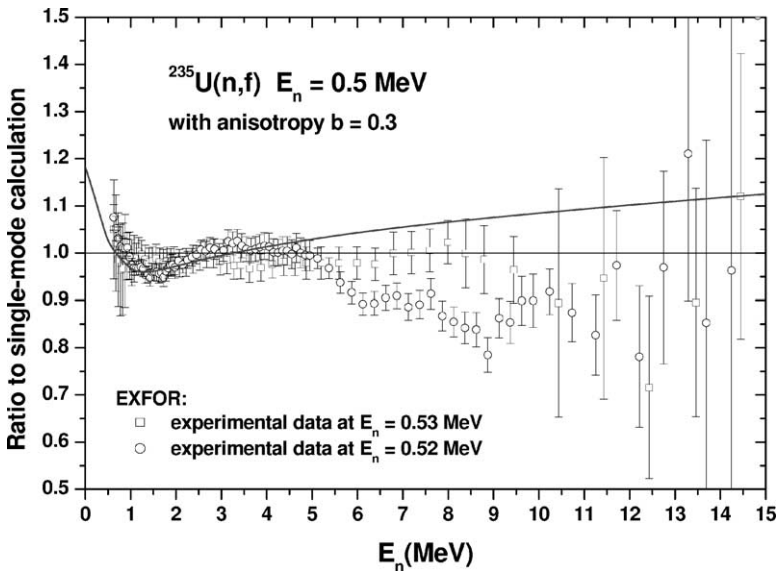


Fig. 11. Prompt fission neutron spectrum with the anisotropy effect taken into account at $E_n = 0.5$ MeV and presented as ratio to the single-mode calculation (from Ref. [14]), in comparison with the experimental data (taken from the EXFOR library).

- The present prompt neutron multiplicity evaluation is obtained in good agreement with the experimental data.
- The overall agreement of the prompt fission neutron spectra with the existing experimental data is also good, although, especially at thermal incident neutron energy, the different experimental data sets are quite scattered.
- The total average prompt fission γ -ray energy is in good agreement with the experimental data as well.
- The inclusion of the anisotropy effect leads to a better agreement with the experimental data in the lower fission neutron energy region of the spectrum.

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