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Analysis of prompt fission neutron spectrum and multiplicity for ^{237}Np(n, f) in the frame of multi-modal Los Alamos model*

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The improved version of Los Alamos model with the multi-modal fission approach is used to analyse the prompt fission neutron spectrum and multiplicity for the neutron-induced fission of ^{237}Np. The spectra of neutrons emitted from fragments for the three most dominant fission modes (standard I, standard II and superlong) are calculated separately and the total spectrum is synthesized. The multi-modal parameters contained in the spectrum model are determined on the basis of experimental data of fission fragment mass distributions. The calculated total prompt fission neutron spectrum and multiplicity are better agreement with the experimental data than those obtained from the conventional treatment of the Los Alamos model.

Keywords: ^{237}Np(n, f), prompt neutron spectrum, neutron multiplicity, multi-modal model, Los Alamos model

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1. Introduction

The prompt fission neutron energy spectrum and multiplicity of actinides are crucial important nuclear data for nuclear application and theory, and a new calculation of these quantities with a higher accuracy is required. The early representations of the prompt fission neutron spectrum, in which many physical effects were neglected, are the Maxwellian and Watt spectrum representations with one or two parameters that are adjusted to reproduce the experimental spectrum.[1,2]

During the last two decades, several models for calculating the prompt fission neutron spectrum and multiplicity have been developed. The Dresden model[3] and the Hauser–Feshbach calculations[4] require too many input data though they probably yield more accurate calculations of the neutron spectrum. The widely-used Los Alamos (abbreviated to LA model hereafter) model[5] accounts for some physical effects such as the variety of fission fragments (FFs) with various excitation energies and has a good predictive power with using less input parameters.

In recent years the concept of the multi-modal random neck-rupture model[6,7] has been used to quantitatively predict the FF properties. If scission takes place in different fission modes (e.g. standard I, standard II and superlong modes) separately, the energy partitions and the nuclear properties of FFs before neutron emission are different; thus the prompt neutron spectrum and the multiplicity are dependent on fission mode. For the neutron induced fission of actinides, the multi-modal analysis of the prompt neutron spectrum and the multiplicity is necessary and has been applied to some calculations of prompt neutron spectrum of actinide nuclei isotopes.[8−14] The basic discrepancy among recent studies arises from the fact that the fission fragment distributions were analysed by using different methods.

The present paper gives the results obtained by using the multi-modal (MM) approach to calculate the prompt fission neutron spectrum and the multiplicity for neutron induced fission of ^{237}Np in an incident neutron energy range below the second chance fission threshold. A detailed analysis of the new empirical

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data of the fragment mass distributions is performed at the Institute for Reference Materials and Measurements (IRMM). The three main fission modes (S1, S2, and SL) are taken into account in order to determine the input model parameters. The partial spectrum and multiplicity for each mode are calculated separately by using the improved LA model,[2] and the total neutron spectrum and the multiplicity are synthesized. The calculated prompt neutron spectrum and multiplicity are in good agreement with the experimental data.

2. Method

In the frame of the multi-modal fission approach,[8,11] the prompt neutron spectrum $N_{\rm tot}(E)$ and multiplicity υ_p at incident energy E_n are calculated as the superposition of prompt neutron spectra and multiplicities associated with a particular fission mode from the following expressions:

$$N_{\rm tot}(E)=\frac{\sum_m \omega_m\langle\upsilon\rangle_m N_m(E)}{\sum_m \omega_m\langle\upsilon\rangle_m}, \tag{1}$$

and

$$\langle\upsilon\rangle_{\rm tot}=\frac{\sum_m \omega_m\langle\upsilon\rangle_m}{\sum_m \omega_m}, \tag{2}$$

where ω_m is the branching ration of mode m. $N_m(E)$ and $\langle\upsilon\rangle_m$ are, respectively, the prompt fission neutron spectrum and the multiplicity for the mode m:

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

where $N_m^L(E,E_f^L,\sigma_{c,L})$ and $N_m^H(E,E_f^H,\sigma_{c,H})$ are the spectra of individual light fragment (LF) and heavy fragment (HF) for mode m. In the present work, we assume the numbers of neutrons emitted from the LF and the HF to be identical. For each fission mode the prompt fission neutron spectrum $N^j(E,E_f^j,\sigma_{c,j})$ of the individual FF ($j=L$ or H) is calculated by using the LA model.[2,5]

The average prompt neutron multiplicity of mode m is obtained from the energy conservation and can be calculated by using the following relationship:

$$\langle\upsilon\rangle_m=\frac{\langle E_r\rangle_m+E_n+B_n-\langle{\rm TKE}\rangle_m-\langle E_\gamma\rangle_m}{\langle S_n\rangle_m+\langle\varepsilon\rangle_m}, \tag{4}$$

where E_n and B_n are the incident neutron energy and the neutron binding energy in the fissioning nucleus, respectively. $\langle E_r\rangle_m$ and $\langle S_n\rangle_m$ are, respectively, the average energy released from fission and the average neutron separation energy from the FFs for mode m, and $\langle{\rm TKE}\rangle_m$ and $\langle E_\gamma\rangle_m$ are the average total FF kinetic energy and the average prompt gamma ray energy, respectively. The first-order moment of the centre-of-mass system spectrum of mode m, $\langle\varepsilon\rangle_m$, is calculated as follows:[2,11]

$$\begin{aligned}\langle\varepsilon\rangle_m =&\frac{1}{T_{\rm max}^2}\int_0^\infty \varepsilon^2\sigma_{cL}(\varepsilon){\rm d}\varepsilon \\ &\times\int_0^{T_{\rm max}} k_L(T)T\exp(-\varepsilon/T){\rm d}T \\ &+\frac{1}{T_{\rm max}^2}\int_0^\infty \varepsilon^2\sigma_{cH}(\varepsilon){\rm d}\varepsilon \\ &\times\int_0^{T_{\rm max}} k_H(T)T\exp(-\varepsilon/T){\rm d}T,\end{aligned} \tag{5}$$

where

$$k_j(T)=\left[\int_0^\infty \sigma_{cj}(\varepsilon)\varepsilon\exp(-\varepsilon/T){\rm d}\varepsilon\right]^{-1}$$

with

$$j=L \text{ or } H, \tag{6}$$

where ε and $\sigma_{cj}(\varepsilon)$ are, respectively, the neutron energy in the centre-of-mass system (CMS) and the compound nucleus formation cross section in an inverse process, and they can be calculated by using an optical-model potential with explicit isospin dependence[2] to describe neutron-rich FF more correctly. All quantities involved in the terms on the right-hand side of expression (5) are related to fission mode m.

According to Ref.[15], there is a linear dependence of the average prompt gamma ray energy on prompt neutron multiplicity, where the slope and the intercept for all fission modes are the same and depend only on charge and mass number (Z_{CN} and A_{CN}) of the fissioning nucleus, the average prompt gamma ray energy $\langle E_\gamma\rangle_m$ is given by

$$\langle E_r\rangle_m=p\langle\upsilon\rangle_m+q \tag{7}$$

with

$$p=6.71-0.156\frac{Z_{CN}^2}{A_{CN}},$$

and

$$q=0.750+0.088\frac{Z_{CN}^2}{A_{CN}}. \tag{8}$$

3. Multi-modal parameters in LA model

The multi-modal parameters contained in the LA model are calculated for ^{237}Np(n, f) at incident neutron energies from 0.3 to 5.5 MeV, where only the first fission chance is involved. The experimental FF yield $Y(A)$ data are analysed to determine the following parameters: multi-modal branching ration ω_m, average FF mass of mode $\overline{A}_m$, and standard deviation of the FF mass distribution of mode σ_{Am}.

3.1. Fragment mass distribution

According to the Moriyama–Ohnishi systematics,[16] the fragment mass distribution can be described as

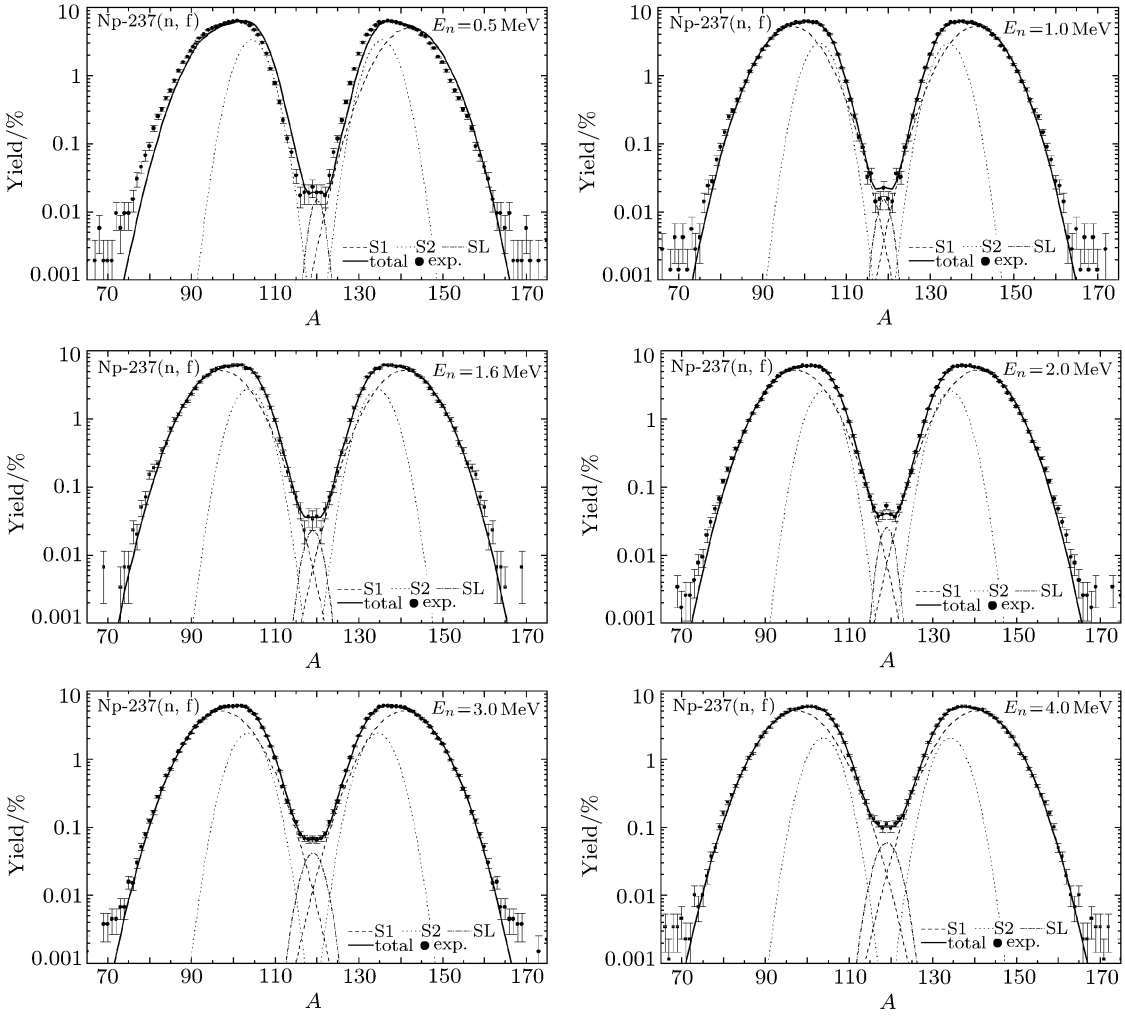
$$Y(A) = \sum_m Y_m(A), \tag{9}$$

where the mass distribution for fission mode m is expressed in the following form:

$$Y_m(A) = \frac{\varpi_m}{\sigma_{Am}\sqrt{2\pi}} \exp\left(- \frac{(A - \overline{A}_m)^2}{2\sigma_{Am}^2} \right). \tag{10}$$

The sum over all in the process must reproduce the experimental data.

In the present work, the Gauss–Newton nonlinear least square method is used to analyse the FF mass distribution. The comparisons between the fitted FF mass distributions and the experimental data for ^{237}Np(n, f) at E_n = 0.5, 1.0, 1.6, 2.0, 3.0, 4.0, 5.0, 5.5 MeV are shown in Fig.1.



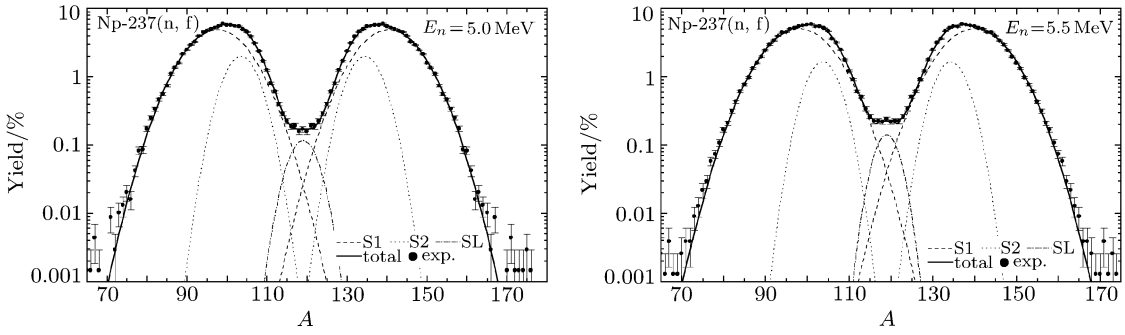


Fig.1. Mass distributions of fission fragments for ^{237}Np(n, f) for incident neutron energies from 0.5 to 5.5 MeV. The contributions of fission modes are shown with the dashed (for S2 mode), dotted (for S2 mode) and dash-dotted (for SL mode) lines, separately. The solid curves show the superposition results of the three modes. The experimental data are cited from Ref.[17].

The behaviours of the modal branching ratio versus incident neutron energy are shown in Fig.2.

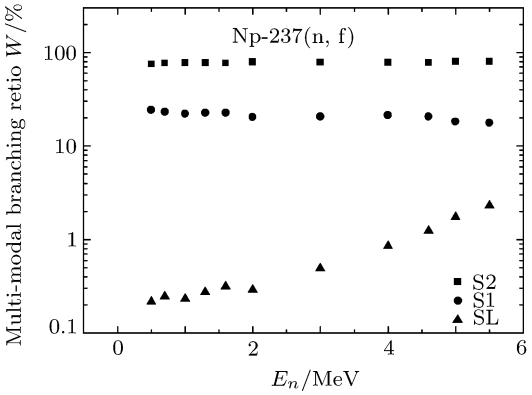


Fig.2. Multi-modal branching ratios as a function of incident neutron energy E_n.

In the present work, the experimental FF mass range ($A_L \in [76, 118]$ and $A_H \in [118, 160]$) is taken into account. For each FF mass pair, four isobars per mass are taken into account with values of nuclear charge Z which are the nearest integer values above and below the most probable charge.

3.2. Average energy released from each mode

The average energy released from fission mode m is calculated by using the following relation:

$$\langle E_r\rangle_m = \frac{\sum\limits_i Y_{mi} \sum\limits_{j=z_p-2}^{j=z_p+2} pz_{i,j} E_{ri,j}}{\sum\limits_i Y_{mi} \sum\limits_{j=z_p-2}^{j=z_p+2} pz_{i,j}}, \tag{11}$$

where m is the index of the mode (S1, S2, SL), i is the range of FF pairs, $Y_{m,i}$ is the FF mass yield for mode m and FF pair i, and pz_i is the charge distribution corresponding to the FF pair i, approximated by a Gaussian function.[11,18−20]

The dependence of $\langle E_r\rangle_m$ on incident neutron energy E_n is shown in Fig.3(a).

3.3. Average FF neutron separation energy for each fission mode

The average FF neutron separation energy for fission mode m is calculated by using the following expression:

$$\langle S_n\rangle_m = \frac{\sum\limits_i Y_{mi} \sum\limits_{j=z_p-2}^{j=z_p+2} pz_{i,j} \overline{S_{n,i,j}}}{\sum\limits_i Y_{mi} \sum\limits_{j=z_p-2}^{j=z_p+2} pz_{i,j} \overline{S_{n,i,j}}}, \tag{12}$$

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

$$\overline{S_{n,i}} = \frac{1}{2}(\overline{S^L_{n,i}} + \overline{S^H_{n,i}}),$$

and

$$\overline{S^{L,H}_{n,i}} = \frac{1}{2}\left(S^{L,H}_{n,i} + \frac{S^{L,H}_{2n,i}}{2}\right) \tag{13}$$

with

$$\begin{aligned} S_n &= [M(Z, A-1) + m_n - M(Z, A)]c^2 \\ &= \Delta(Z, A-1) + \Delta(n) - \Delta(Z, A), \end{aligned} \tag{14}$$

and

$$\begin{aligned} S_{2n} &= [M(Z, A-2) + 2m_n - M(Z, A)]c^2 \\ &= \Delta(Z, A-2) + 2\Delta(n) - \Delta(Z, A), \end{aligned} \tag{15}$$

where S_{2n} is used to remove pairing effects. S_n and S_{2n} are calculated by using the Reference Input Parameter Library (RIPL).[20]

The dependence of $\langle S_n\rangle_m$ on incident neutron energy E_n is shown in Fig.3(b).

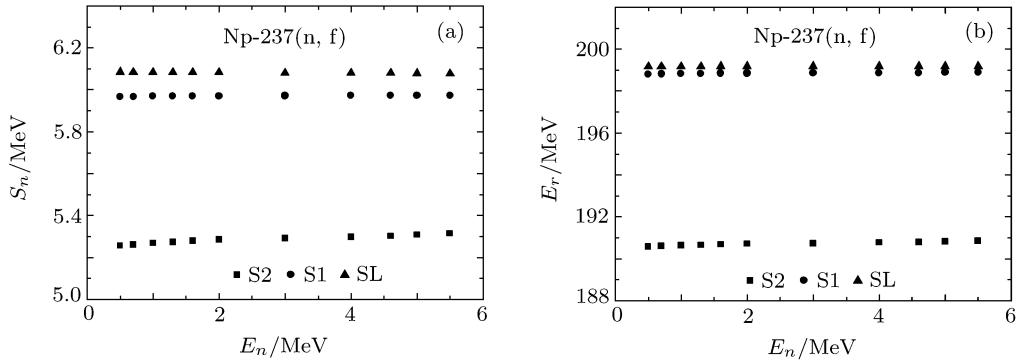


Fig.3. Average FF neutron separation energies (a) and average energies released from fission (b) each as a function of incident neutron energy for S2, S1 and SL modes separately.

3.4. Multi-modal average total kinetic energy of FF

The average total kinetic energy of the FF for fission mode m is calculated as

$$\langle \mathrm{TKE} \rangle_m = \frac{\sum\limits_i Y_{m,i} \overline{\mathrm{TKE}_i}}{\sum\limits_i Y_{m,i}}, \tag{16}$$

where $\overline{\mathrm{TKE}_i}$ is the average total kinetic energy of the i-th FF pair which can be written as

$$\overline{\mathrm{TKE}_i} = E_{\mathrm{Cou},i} + E_{\mathrm{nuc},i} + K_{s,i} \tag{17}$$

with $E_{\mathrm{Cou},i}$ and $E_{\mathrm{nuc},i}$ being the Coulomb repulsion and nuclear energy between the newborn fragments, respectively, and $K_{s,i}$ being the prescission kinetic energy.

In the present work, the formulae given by Brosa *et al*[6] are used to calculate $E_{\mathrm{Cou},i}$ and $E_{\mathrm{nuc},i}$. There are experimental indications that the values of $K_{s,i}$ are small, the upper limit being 8 MeV,[21] thus the upper limit is used in this work. The calculated values of average total kinetic energy $\overline{\mathrm{TKE}}(A)$ of the fragments from ^{237}Np(n, f) at $E_n = 4.0$ MeV are plotted versus the heavy fragment mass in Fig.4(a). The average total kinetic energies of the fragments for S2, S1 and SL modes each as a function of the incident neutron energy obtained from our calculations are shown in Fig.4(b).

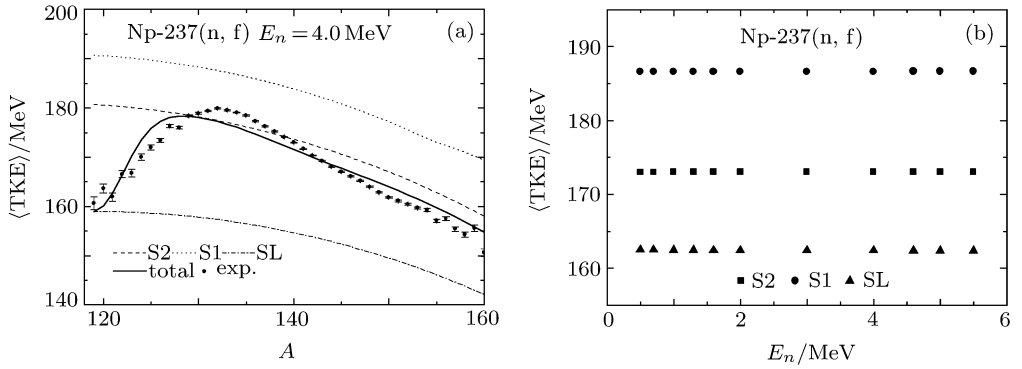


Fig.4. (a) Comparison of experimental data[22] (symbols) with corresponding fitting of the $\overline{\mathrm{TKE}}(A)$ for ^{237}Np(n, f) at $E_n = 4.0$ MeV, where dashed lines represent contributions from the single modes and solid line denotes the superposition result of the three modes; (b) calculated values of average total kinetic energy $\overline{\mathrm{TKE}}(A)$ of the fragments of ^{237}Np(n, f) for S2, S1 and SL modes each as a function of incident neutron energy.

4. Results

4.1. Analyses of partial spectrum of ^{237}Np(n, f)

In the present work, the neutron spectra of ^{237}Np(n, f) for neutron incident energies from thermal up to 5.0 MeV are calculated. Figure 5 shows the calculated partial spectra for three modes at $E_n = 0.5$, 2.0, 3.0 and 5.0 MeV separately. From Fig.5, it can be found that the S1-spectrum is the softest, and the partial spectra for S2 and SL modes are harder, i.e. the larger the deformation of compound nucleus

is, the harder its corresponding partial spectrum will be, which accords with the experimental observation and the calculated results presented by Ohsawa and Hambsch *et al.*[8,11]

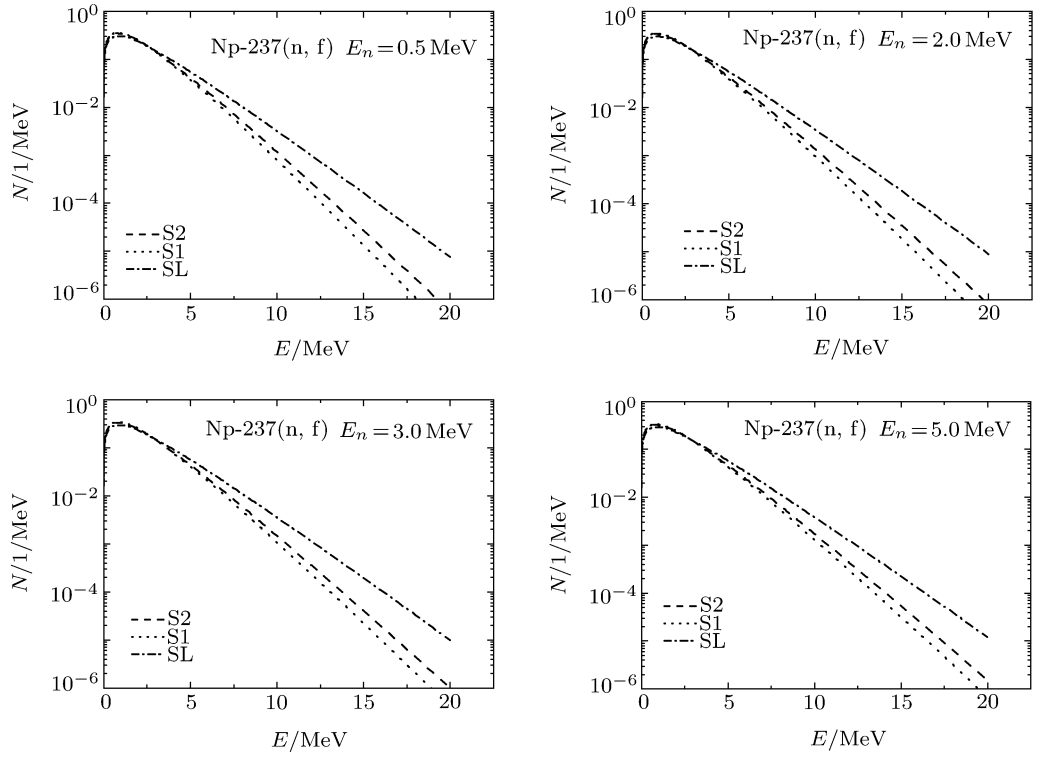


Fig.5. Calculated partial spectra for S2, S1, SL modes at $E_n = 0.5$, 2.0, 3.0 and 5.0 MeV separately.

4.2. Comparison of total spectrum with experimental data

The total prompt neutron spectra calculated from expressions (1) and (2) for $E_n = 0.52$ and 4.9 MeV are shown in Figs.6(a) and 6(b), respectively, together with experimental data given by Kornilov *et al* [23] and Trufanov *et al.*[24] The Maxwellian spectra are also shown in the figures. It can be seen that the multi-modal calculations reproduce the experimental data and are in better agreement with the experimental data than the Maxwellian spectrum.

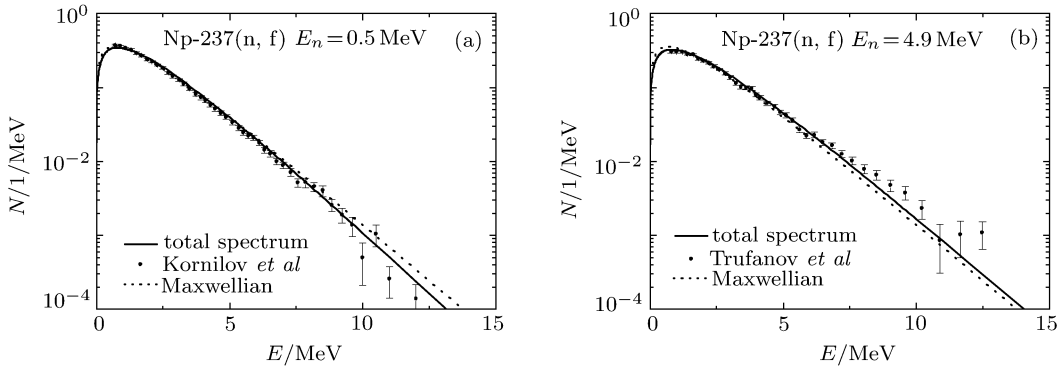


Fig.6. The total neutron spectra for $E_n = 0.52$ MeV (a) and 4.9 MeV (b) compared with experimental data[23,24] and Maxwellian spectrum.

4.3. Comparison between single modal and multi-modal calculations

It is interesting to compare the conventional single modal results with the multimodal LA model calculations. Figure 7 shows the calculation results obtained by using the two methods and experimental data[23] for $E_n = 0.52$ MeV. It is observed that although the experimental spectrum tends to be softer, the multimodal calculations are in better agreement with the experimental data.

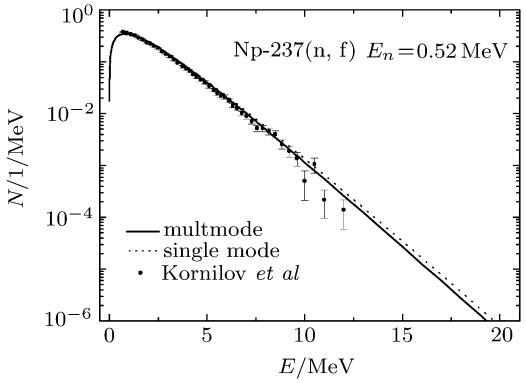


Fig.7. Total neutron spectra calculated by using conventional single modal and multi-modal methods with experimental data[23] at $E_n = 0.52$ MeV.

4.4. Comparison of total average prompt neutron multiplicity with experimental data

The total average prompt multiplicity calculated on the basis of the multi-modal multiplicities by using expressions (2) and (4) accords well with experimental data,[25–27] as can be seen in Fig.8.

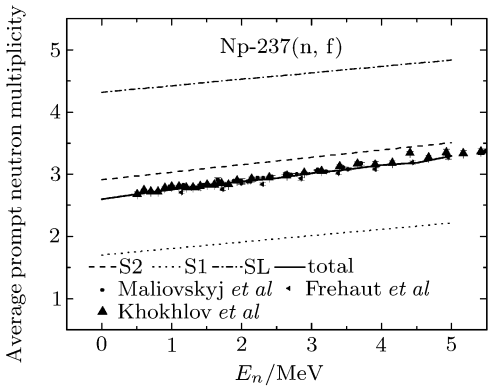


Fig.8. Average prompt fission neutron multiplicity (solid line) as a function of incident neutron energy, compared with the experiment data,[25–27] where contributions of three fission modes are also shown.

5. Conclusions

The improved version of Los Alamos model with the multi-modal fission approach is used to calculate the prompt neutron spectra and multiplicities of ^{237}Np(n, f) for incident neutron energies from 0.5 to 5.5 MeV. The total prompt neutron spectrum and multiplicity calculated in the present work accord well with the relevant experimental data. A detailed multimodal analysis of the new empirical data of the fragment mass distributions is performed by taking three dominant fission modes (S1, S2, and SL) into account. The calculations of the multi-modal average total kinetic energy of the fission fragments and the total prompt gamma ray energy are in good agreement with the experimental data.

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