Actinide symmetric/asymmetric nucleon-induced fission up to 200 MeV

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Abstract

The fission cross sections of the symmetric SL-mode and the asymmetric lumped (S1 + S2)-mode of the $^{235}$U(n, F), $^{237}$Np(n, F) and $^{238}$U(p, F) reactions are calculated up to $E_n = 200$ MeV within a statistical model. For each fissioning nuclide, emerging in (n, xnf) reactions, a separate triaxial outer fission barrier is assumed for the SL-mode. To reproduce the measured branching ratio of symmetric and asymmetric fission events, strong contribution of fission from neutron-deficient nuclei is assumed. Damping of the contribution of triaxial collective modes to the level density at the SL-mode outer saddle seems to be essential for the description of the branching ratio. The sensitivity of the calculated branching ratio to the fissility of the target nuclide and the incident particle is investigated.

1. Introduction

In neutron-induced fission of actinides, the contribution of the symmetric or superlong (SL) fission mode [1,2] increases with the excitation energy [3–7]. At incident neutron energies $E_n \gtrsim 6$ MeV, pre-fission neutrons might be emitted, before the fissioning nuclide reaches the outer saddle point. We will call them pre-saddle. This peculiarity considerably complicates the interpretation of fission observables. A number of nuclides might contribute to the fission observables, about $\sim 20$ for the $^{238}$U(n, xnf) fission reaction at $E_n \sim 200$ MeV [8]. In other words, an ensemble of nuclides, which emerge after emission of $x$ pre-saddle neutrons, contributes to the symmetric and asymmetric fission. The branching ratio of symmetric fission events to the total observed fission events $r_{\text{sym}} = \sigma_{\text{fSL}} / (\sigma_{\text{fSL}} + \sigma_{\text{f(S1+S2)}})$ was obtained for $^{238}$U(n, F) by Zoller et al. [9] at $E_n$ up to $\sim 500$ MeV. The values of $r_{\text{sym}}$ were obtained from the deduced yield distributions of primary fission fragments and their average total kinetic energies (TKE) as a function of mass. Description of the branching ratios $r_{\text{sym}}$ by Zoller et al. [9] up to $E_n \sim 200$ MeV favors the major contribution to the observed fission cross section of fission chances with a larger number of pre-saddle neutrons [8]. With increasing fissility of the target nuclide this number might be somewhat lower. We will apply the same approach, which was previously used for the description of the cross section and the branching ratio of symmetric/asymmetric fission of the observed $^{238}$U(n, F) reaction, to which either first chance $^{238}$U(n, f) and emission
sive fission $^{238}\text{U}(n,xnF)$ reactions contribute, to describe measured fission cross sections in the reactions $^{235}\text{U}(n,F)$, $^{237}\text{Np}(n,F)$ and $^{238}\text{U}(p,F)$. In particular, we will predict the $^{235}\text{U}(n,F)^{\text{sym}}$, $^{237}\text{Np}(n,F)^{\text{sym}}$ and $^{238}\text{U}(n,F)^{\text{sym}}$, $^{237}\text{Np}(n,F)^{\text{sym}}$ fission cross sections. Simultaneous analysis of $^{238}\text{U}(n,F)$ and $^{238}\text{U}(p,F)$ reaction cross sections would help to investigate the dependence of observed fission cross sections on the projectile.

2. Statistical model

We assume that the fission fragments are emitted from a chain of U(Np) nuclei after pre-equilibrium (PE) emission and evaporation of neutrons [10]. We do not take into account charged-particle emission, for justification see the discussion in [8]. A coupled-channels model, fitting the $^{238}\text{U}$ total fission cross sections [11] up to $E_{\text{ex}} \approx 200$ MeV is employed. The contribution of the SL-mode $\sigma_{\text{SL}}$ to the observed fission cross section, originating from $(n,nF)$ fission reactions, was calculated using the fission probability $r_{\text{SL}}^{\pi}(U)$ for symmetric fission of the fissioning $x$th nucleus,

$$
\sigma_{\text{SL}}(E_n) = \sigma_{\text{SL}}(E_n) + \sum_{x=1}^{X} \int_{0}^{U_{\text{max}}} W^{\pi}(x+1)(U) r_{\text{SL}}^{\pi}(x+1)(U) dU,
$$

where $K_{\text{rot}}(U,J)$ and $K_{\text{vb}}(U)$ are the factors of rotational and vibrational enhancement. At saddle and ground-state deformations $K_{\text{rot}}(U)$ is defined by the symmetry class, adopted from shell-model calculations [2,14,15]. At inner saddle we assume axial symmetry for neutron numbers $N \leq 144$ and triaxial shape for $N > 144$. At outer saddle triaxiality is assumed for mass symmetric mode, while axial shape is assumed for the mass asymmetric mode. For more extensive discussions see [2,7,8] and references therein. At excitations $U \geq U_{\text{cr}}$, damping of rotational modes was anticipated [16]. Damping of rotational modes contribution to the nuclear level density $\rho(U,J,\pi)$ might be different for axially symmetric and triaxial nuclei [17], i.e.,

$$
K_{\text{rot}}^{\text{sym}}(U) = (\sigma_1^2 - 1)F(U) + 1, \quad (3)
$$

$$
K_{\text{rot}}^{\text{asym}}(U) = K_{\text{rot}}^{\text{sym}}(U)((2\sqrt{n}\sigma_1 - 1)F(U) + 1), \quad (4)
$$

$$
F(U) = (1 + \exp(U - U_{\text{cr}})/\delta W_{\text{cr}})^{-1}. \quad (5)
$$

Here, $\sigma_1^2$ and $\sigma_2^2$ are the spin-distribution parameters. The mass asymmetry for the S1(S2)-modes at the outer saddles doubles the rotational enhancement factors as defined by Eqs. (3), (4). Shell effects in the level density are modeled with the dependence of the $\alpha$-parameter on the shell correction $\delta W$ as recommended by Ignatyuk et al. [13]:

$$
\alpha(U) = \bar{\alpha}(1 + \frac{\delta W(U)}{\delta W_{\text{cr}}}).
$$

The value of the asymptotic $\bar{\alpha}$-parameter $\bar{\alpha}$ is defined by fitting to the neutron-resonance spacing $(D_{\text{obs}})$ or by systematics. We assume $\bar{\alpha} = \bar{\alpha}_{\text{cr}}$, then the $\alpha_1/\alpha_2$ ratio depends on values of $\delta W_{\text{cr}}$, taken from [18] $(\delta W_{\text{cr}})$ and [19] $(\delta W_{\text{cr}})$. The total fission cross section $\sigma_{\text{nf}} = \sigma_{\text{SL}} + \sigma_{\text{SF}(S1+S2)}$ depends on the contributions of both terms. These contributions strongly depend on the asymptotic value $\bar{\alpha}(A)$ of the $\alpha$ parameter of the fissioning nuclei, while the branching ratio $r^{\text{sym}}$ depends both on the contributions of fission chances and the damping of the contribution of the triaxial collective modes to the nuclear level density (see Eqs. (4), (5) for the SL fission mode [8]. The contributions of fission chances to the observed fission cross section $\sigma_{\text{nf}}$ are affected by decreasing the asymptotic value of the $\alpha_1$ parameter.

3. Analysis

The total fission cross section $\sigma_{\text{nf}} = \sigma_{\text{SL}} + \sigma_{\text{SF}(S1+S2)}$ depends on the contributions of both terms. These contributions strongly depend on the asymptotic value $\bar{\alpha}(A)$ of the $\alpha$ parameter of the fissioning nuclei, while the branching ratio $r^{\text{sym}}$ depends both on the contributions of fission chances and the damping of the contribution of the triaxial collective modes to the nuclear level density (see Eqs. (4), (5) for the SL fission mode [8]. The contributions of fission chances to the observed fission cross section $\sigma_{\text{nf}}$ are affected by decreasing the asymptotic value of the $\alpha_1$ parameter.
with energy as

\[ \tilde{\alpha}_f(U, A) = \tilde{\alpha}_f(A) \left( 1 - 0.1 \left( \frac{U - 20}{U} \right)^{1/4} \right). \]  

(6)

This expression was obtained by consistent description of observed Th, U and Pu fission cross sections [20] as well as branching ratio \( r^{\text{sym}} \) for \(^{238}\text{U}(n, F)\) reaction [8]. The heights of the outer fission barrier \( B \) of the SL mode for \(^{239}\text{U}\) and \(^{236}\text{U}\) fissioning nuclides were derived to be higher than those of the asymmetric modes, i.e., \( (E_{\text{BSL}} - E_{\text{BSL(S2)}}) \sim 3.5 \text{ MeV} \), while \( h\omega_{\text{BSL}} = 2.25 \text{ MeV} \) [7]. The contributions of lighter U nuclides via \((n, x_{\text{nf}})\) reactions to the observed symmetric fission might be obtained assuming for each of them the same difference of the outer barriers for the symmetric SL and the asymmetric fission \( S1(S2) \) modes, the shell-correction values being defined as \( (\delta W_{\text{BSL}} - \delta W_{\text{BSL(S2)}}) \sim 3.5 \text{ MeV} \), assuming \( \delta W_{\text{BSL(S2)}} \sim 0.6 \text{ MeV} \) [19]. The assumption that the difference of mass-symmetric and mass-asymmetric fission barriers do not vary with the neutron number might seem too crude (see Schmidt et al. [21], however, we believe that weak isotopic dependence of \( (E_{\text{BSL}} - E_{\text{BSL(S2)}}) \) could be compensated by slight variation of asymptotic value of the \( \tilde{\alpha}_f \) parameter (see Eq. (6)). The inner and outer fission-barrier parameters of uranium for the double-humped fission-barrier model [22], relevant for the saddle asymmetries, predicted by SCM calculations [14] for asymmetric fission, were defined in [23,24]. This simple systematics of level-density and fission-barrier parameters for U nuclides allowed to reproduce the observed \(^{235}\text{U}(n,F)\) fission cross section [25]. The same approach works in the case of neutron-induced fission of \(^{237}\text{Np} \) target nuclides.

It might be anticipated that further sophistication of the model, i.e., inclusion of the temperature and angular momentum dependence of fission barriers and shell corrections, influence of neutron shell \( N = 126 \) on collective enhancement in neutron channel [17], or use of calculated with SCM method [2] fission barriers for symmetric and asymmetric fission modes of U nuclei, would not change pattern of emissive fission contributions. Lowering of asymptotic value \( \tilde{\alpha}_f \) of \( \alpha_f \) parameter at saddle deformations might be considered as reproducing lumped effect of all these factors. We anticipate also that because of strong emissive fission nature of \(^{238}\text{U}(n, f)\) reaction for \( E_n \lesssim 200 \text{ MeV} \), vanishing of the distinction of symmetric and asymmetric valleys at high excitation of fissioning nuclei [26] would not be much pronounced.

Fig. 1 shows the calculated symmetric \(^{235}\text{U}(n,F)^{\text{sym}}\), asymmetric \(^{235}\text{U}(n,F)^{\text{asym}}\) and symmetric + asymmetric \(^{235}\text{U}(n,F)\) fission cross sections for the energy-dependent asymptotic \( \tilde{\alpha}_f(U, A) \) of \( \alpha_f \) parameter (Eq. (6)). The set of solid curves shows the \(^{235}\text{U}(n,F)^{\text{sym}}\) and \(^{235}\text{U}(n,x_{\text{nf}})^{\text{sym}}\) fission cross sections, while the dashed lines show those for the asymmetric neutron-induced fission of \(^{235}\text{U}\) target nuclide.

Fig. 2 shows the symmetric \(^{237}\text{Np}(n,F)^{\text{sym}}\), asymmetric \(^{237}\text{Np}(n,F)^{\text{asym}}\) and symmetric + asymmetric \(^{237}\text{Np}(n,F)\) fission cross sections. The lines on Fig. 2 have the same meanings as on Fig. 1. The sum of the calculated \(^{237}\text{Np}(n,F)^{\text{sym}}\) and \(^{237}\text{Np}(n,F)^{\text{asym}}\) reaction cross sections (dash-dotted line) is quite compatible with the observed fission cross section [27] up to \( E_n \sim 200 \text{ MeV} \). The data by Hambsch et al. [3] for the symmetric fission yield are also reproduced.

In case of the proton-induced fission reaction \(^{238}\text{U}(p,F)\), the observed fission reaction cross section could be calculated using the fission-barrier and level-density parameters of Np nuclei, obtained by fitting
the $^{237}$Np(n, $\gamma$) fission cross section. It might be anticipated that $^{238}$U(p, x$\gamma$) reactions give the dominant contribution to the observed fission cross section. The sum of $^{238}$U(p, $\gamma$)$_{\text{sym}}$ and $^{238}$U(p, $\gamma$)$_{\text{asym}}$ cross sections is compared with the measured data on Fig. 3.

The relative contributions of $\sigma_{\text{sym}}$ and $\sigma_{\text{asym}}$ to the observed fission cross section $\sigma_{\gamma}$ could be controlled by comparing the calculated branching ratio $r_{\text{sym}}$ with the measured data by Zoller et al. [9] (see Fig. 4) for the $^{238}$U(n, $\gamma$) reaction and with the measured data below the fission threshold by Hambsch [5] for $^{238}$U(n, $\gamma$), Vives et al. [4] for $^{235}$U(n, $\gamma$) and Hambsch et al. [3] for $^{237}$Np(n, $\gamma$). The relative contributions of fission chances with low and high number of pre-fission neutrons have a major influence on the energy dependence of $r_{\text{sym}}$ at $E_{\gamma} \gtrsim 25$ MeV. When the energy-dependent asymptotic $\tilde{a}(A)$ of $a_{\gamma}$ parameter is employed (see Eq. (6)), higher chances make a predominant contribution to the observed fission cross section, and damping of triaxial collective modes contribution (see Eqs. (4), (5)) at outer symmetric fission saddle allows to reproduce the measured data by Zoller et al. [9] for $^{238}$U(n, $\gamma$) at $E_{\gamma} \gtrsim 10$ MeV. Below the fission threshold there is a systematic difference of branching-ratio data by Hambsch [5] for $^{238}$U(n, $\gamma$) and Vives et al. [4] for $^{235}$U(n, $\gamma$).

The calculated branching ratios $r_{\text{sym}}$ for $^{235}$U(n, $\gamma$) and $^{238}$U(n, $\gamma$) reactions are different around the
235,238\textsuperscript{U}(n,\textit{nf}) reaction thresholds. A strong dip is observed for the \textit{238}\textsuperscript{U}(n,F) reaction, a similar dip of lower amplitude is predicted for the \textit{235}\textsuperscript{U}(n,F) reaction. These peculiarities are due to different contributions of (n,F)\textit{ asym} and (n,F)\textit{ sym} reactions because of higher fission thresholds of the symmetric fission modes as compared with those of asymmetric fission. At incident neutron energies $E_n \geq 25$ MeV, symmetric fission makes the higher contribution to the observed fission cross section in the case of the \textit{235}\textsuperscript{U}(n,F) reaction, but the lowest in the case of \textit{238}\textsuperscript{U}(n,F). This might be explained by different emissive fission contributions to the (n,F)\textit{ sym} and (n,F)\textit{ asym} reactions. The branching ratio for the \textit{238}\textsuperscript{U}(p,F) reaction is not much different from that of the \textit{238}\textsuperscript{U}(n,F) reaction at incident nucleon energies $E_{n(p)} \geq 50$ MeV. At lower energies, i.e., $15 \leq E_{n(p)} \leq 50$ MeV, the difference is due to the different contributions of emissive fission reactions to the observed proton- and neutron-induced fission cross sections. In summary, different shapes of branching ratios $r^{\textit{sym}}$ for the \textit{237}\textsuperscript{Np}(n,F), \textit{238}\textsuperscript{U}(n,F), \textit{238}\textsuperscript{U}(n,F) and \textit{238}\textsuperscript{U}(p,F) reactions for $E_n \lesssim 90$ MeV might be attributed to the higher contribution of lower fission chances in case of higher target nuclide fissility, for higher energies $E_n \gtrsim 90$ MeV they do not differ much.

4. Conclusions

The damping of the contribution of axial collective modes to the level density at the inner and the outer saddles and at equilibrium deformations as well as triaxial damping at the outer saddle deformations of the SL-mode, produce symmetric/asymmetric emissive fission partitioning of \textit{238}\textsuperscript{U}(n,F) based on the consistent description of the observed fission cross section and the symmetric/asymmetric fission branching ratio. The estimate of asymptotic level density parameter $\tilde{a}_f(A)$ of the fissioning nuclei is equivalent to the more fission events at lower intrinsic excitation energies (which is due to pre-fission neutron emission), or more fission events from neutron-deficient U nuclei, i.e., from higher fission chances. Triaxial damping at the outer saddle of the SL mode is equivalent to less symmetric fission events at higher excitation energy and more symmetric fission from neutron-deficient isotopes, as compared with the “no triaxial damping” case. The dependence of the symmetric fission branching ratio on the fissility of the target nuclide as well as on the incident particle is demonstrated. It would be of much interest to estimate the probability of symmetric fission in the \textit{235}\textsuperscript{U}(n,F) reaction \cite{28} as a function of internal excitation energy of the fissioning nuclei in the emissive fission domain. For this we should calculate the exclusive \textit{238}\textsuperscript{U}(n,\textit{xn}f) reaction neutron spectra for $x \sim 20$. This will be done as an extension of the present work.

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References


