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$^{234}\text{U}(n,f)$ model description of sub-barrier fission cross-section resonances and calculation of prompt neutron emission data

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Abstract

Neutron induced cross-sections of $^{234}\text{U}$ are calculated, focusing on the description of sub-barrier resonances in the fission cross-section. The correlation between the resonant behaviour of fission cross-section of fertile actinides (characterizing the pre-scission stage) and the visible fluctuations of their fission fragment and prompt neutron data (characterizing the post-scission stage), already discussed for the case of $^{238}\text{U}(n,f)$, is outlined and quantitatively supported in this case, too.

The recently measured fission fragment distributions of $^{234}\text{U}(n,f)$ at incident energies covering the range 0.2-5 MeV, allow the averaging of multi-parametric matrices provided by the Point-by-Point (PbP) model, leading to interesting behaviours of prompt neutron data. For the first time the average prompt neutron multiplicity $\langle\nu\rangle$ as a function of TKE is calculated at many incident energies (En) revealing interesting facts: the slope $d\text{TKE}/d\langle\nu\rangle$ does not vary with En and the flattening of $\langle\nu\rangle$ at low TKE values is more pronounced at low En. Average model parameters (like energy release, neutron separation energy from fragments, level density of fragments) as a function of TKE exhibit nice and regular behaviours that can be fitted very well. The obtained average parameter dependences on TKE allow the use of the most probable fragmentation approach (Los Alamos model with subsequent improvements) having as advantages a very short computing time compared to the PbP and Monte Carlo treatments and the possibility to extend the lower and upper limits of the TKE range.

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1. Pre-scission stage, calculation of neutron induced cross-sections

Taking into account that the main purpose of the neutron induced cross-sections calculation for $^{234}$U is the model description of sub-barrier resonances in the fission cross-section, we focus on the incident energy (En) range where only the main compound nucleus is involved. In this En range the reaction takes place through the direct interaction (DI) and compound nucleus (CN) mechanisms. For actinides being permanent deformed nuclei the DI is treated by the coupled channel method. Calculations are performed by the code ECIS using a deformed optical model parameterization (based on microscopic calculation with dispersion taken into account) recently proposed by (Soukhovitskii et al, 2005) in (JENDL4, 2010) and (JENDL/Ac, 2008). The CN mechanism is treated by a statistical model (in the frame of the extended HRTW method) using the refined statistical model for fission with sub-barrier effects of Vladuca et al. A description of this model developed for double or triple humped barriers regarding the so-called optical model for fission (direct, indirect and isomeric components, fission probabilities and so on), contained in the code STATIS and EMPIRE and under development in the code TALYS, can be found in many references (see for instance (Vladuca et al, 2004, 2006) and references therein).

Effective level density parameters of $^{234,235}$U are obtained from recent reported s-wave resonance data $<D_0>$ (Bn) (Mughabghab, 2006; RIPL3, 2011). The obtained level densities at equilibrium deformation describe very well the experimental cumulative number of levels. In the treatment of gamma-channel the spectral factor for E1 is calculated as an incoherent sum of two generalized Lorenzian terms as proposed by Kopecky-Uhl. For normalization a recent experimental s-wave $\gamma$-width is used $<\Gamma_{\gamma}>(Bn)=26$ meV (Mughabghab, 2006), leading to an excellent agreement of the calculated capture cross-section with experimental data (EXFOR, 2012).

Level densities on the fission path contain enhancement factors that are defined relative to the ground state deformation and take into account the nuclear shape symmetries at the inner and outer barrier deformations (details can be found in (Vladuca et al, 2006; Tudora et al, 2012) and reference therein). According to theoretical calculations (as reported in Capote et al, 2009 and references therein) a double humped barrier is assumed for the fissioning nucleus $^{235}$U. The values of the inner and outer barrier curvatures are very close to values of recent systematics of RIPL3 (Capote et al, 2009 and RIPL3, 2009). The present barrier heights are lower than those of RIPL3 systematic because they were obtained in the frame of a much more refined fission model compared to the systematic of RIPL3 (that is based on a classical Hauser-Feshbach treatment with complete damping and humps completely decoupled).

A very good description of experimental fission cross-section data (EXFOR, 2012) is obtained over the entire En range of interest in this work, as it can be seen in Fig. 1. Compared to recent evaluations (such as (ENDF/B-VII, 2006) plotted with green dashed line and (JENDL4, 2010), plotted with cyan dash-dotted line) that are adjusted to agree with experimental data, the present pure calculation (red solid line) without any adjustment succeeded to describe for the first time the sub-barrier resonant behaviour of experimental data (visible in both lower and upper part of Fig. 1 as well as in the lower part of Fig. 4).

2. Post-scission stage, prompt neutron emission calculation

The deterministic Point-by-Point (PbP) model provides as primary results the so-called multi-parametric matrices regarding both fission fragment and prompt emission quantities as a function of fragment (Z and A) and of total kinetic energy (TKE), such as prompt neutron multiplicity $v(Z,A,TKE)$, prompt neutron energy in center-of-mass system $\varepsilon(Z,A,TKE)$, prompt neutron spectrum $N(Z,A,TKE)$ and so on. It also provides model parameters like energy release ($E_r$) corresponding to each pair of fragments, average neutron separation energy $S_n(Z,A,TKE)$, level density parameter $a(Z,A,TKE)$. Basic features of the PbP model including the
manner to obtain the fragmentation range, the excitation energy partition between complementary fragments, the residual temperature distribution of fragments, the manner to calculate the level density parameters of fragments and so on are summarized in Ref. (Tudora, 2013) and references therein.

Fig.1: Present calculation of the fission cross-section (red solid line) in comparison with experimental data taken from EXFOR (different symbols) and recent evaluations ENDF/B-VII (green dashed line) and JENDL4 (cyan dash-dotted line). The low En range is focused in the upper part, the plateau region as well as the large resonance at 0.8 MeV are visible in the lower part.

The fission fragment distributions were measured at IRMM (Al-Adili, 2012; Al-Adili et al, 2012; Al-Adili, 2013) at 14 incident energies covering the range 0.2-5 MeV. Those input data allow the averaging of PbP multi-parametric matrices in order to obtain quantities characterizing fragments and prompt neutron emission as a function of fragment mass number (such as $\gamma(A)$, $g(A)$), as a function of TKE (such as $\langle\gamma\rangle(TKE)$, $g(TKE)$, $\langle\alpha\rangle(TKE)$, $\langle\text{Sn}\rangle(TKE)$) and total average quantities at the En values where the fragment distributions were measured.

2.1. Effects of sub-barrier resonance fission cross-section on fission fragment and prompt neutron emission data

The non statistical fluctuations of fragment properties around the incident energies where the fission cross-sections of fertile actinides exhibit sub-barrier resonances was firstly observed in the case of experimental $\langle\text{TKE}\rangle$ of three fissioning systems $^{238}\text{U(n,f)}$ (Tudora et al, 2012), $^{234}\text{U(n,f)}$ (Al-Adili et al, 2012) and $^{232}\text{Th(n,f)}$ (not yet reported). This correlated behaviour makes the link between the two stages of fission, pre- and –post scission, usually treated by two different classes of models. In the pre-scission stage only one nucleus is involved, the compound one, with the evolution on the fission path, changing the shape from equilibrium deformation, passing trough different stages of deformation up to the rupture point. In this stage the quantity of interest is the fission cross-section obtained concomitantly with other neutron induced cross-
sections (of elastic, inelastic and gamma capture channels). In the post-scission stage many nuclei are involved (the fission fragments (FF) resulted from many possibilities of CN fragmentation) emitting prompt neutrons and gammas according to its structure properties and excitation energy partition. This stage of prompt fission is characterized by quantities referring to both FF and prompt neutrons and gammas as a function of En. The correlation between the sub-barrier fission cross-section (in the pre-scission stage where only one nucleus is involved) and the post-scission stage involving many nuclei can be analyzed in a coherent and consistent manner by taking into account the behaviour of average prompt fission quantities (obtained by averaging the multi-parametric matrices over the FF distributions). The sub-barrier resonances of the fission cross-section, reflected by an increase of the fission channel population in the pre-scission stage, lead in the post-scission stage to an increase of FF distributions around the En values of resonances. This fact is reflected in the non-statistical fluctuations of fragment and prompt neutron data. As it can be seen in Fig. 2, total average quantities like average heavy fragment mass $<A_H>$ (upper part), energy release (middle part) and average separation energy of the first neutron from FF $<S_{n1}>$ (lower part) plotted with full circles exhibit visible variations around En values of resonances. $<E_r>$ and $<S_{n1}>$ are obtained by averaging Q-values of the FF pairs and $<S_{n1}>$ of fragments (forming the FF range taken into account in the PbP treatment) over the charge distribution $P(Z)$ taken as a narrow Gaussian (Tudora, 2013) and the experimental $Y(A,TKE)$ distributions (Al-Adili, 2012, 2013). Q-values, $S_{n1}$ and $P(Z)$ do not change with En, consequently the $<E_r>$ and $<S_{n1}>$ dependences on En are given exclusively by $Y(A,TKE)$.

![Fig. 2: $<A_H>$ (upper part), $<E_r>$ (middle) and $<S_{n1}>$ (lower part) as a function of En. Non-statistical fluctuations around the energy of the highest sub-barrier resonance are indicated by arrows. Appropriate fits are plotted with dashed lines.](image)

Other average quantities like the level density parameter $<a>$ (given traditionally as $<C>=A_0^2<a>$ with $A_0$ the mass number of the fissioning nucleus) obtained by averaging the level density parameter of fragments calculated in the frame of the generalized super-fluid model (see (Tudora, 2013) and reference therein) over the FF distributions, as well as the average total excitation energy $<TXE>$ at full acceleration exhibit non statistical fluctuations around the En of resonances, too, see Fig. 3.
The most interesting result is obtained for the total average prompt neutron multiplicity (obtained by averaging $v(Z,A,TKE)$ over $P(Z)$ and experimental $Y(A,TKE)$). As it can be seen in the upper part of Fig.4, it reveals visible variations (indicated by arrows) around the $En$ values where the fission cross-section (plotted in the lower part of the figure) exhibits sub-barrier resonances.

Fig.3: PbP results of the parameter $<C> = A_0/\langle a \rangle$ (upper part) and $<TXE>$ (lower part) as a function of $En$.

Fig.4: Upper part: total average prompt neutron multiplicity: PbP calculation (red circles) and most probable fragmentation result (solid line) in comparison with experimental data (full black squares) and their linear fit (dashed line) focusing the $En$ range of sub-barrier resonances. Lower part the fission cross-section (experimental data with different symbols and present calculation with red solid line). The variations of the PbP multiplicity result around the incident energies of fission cross-section resonances are indicated by arrows.
Even if the present total average multiplicity result reveals a different increasing slope compared with other evaluations (ENDF/B-VII, 2006; JENDL4, 2010) (that are almost linear fits of the four experimental points of Mather et al., given in EXFOR, 2012) it remains in agreement with these data.

The sub-barrier resonances in the fission cross-section of $^{234}$U are more pronounced (especially the large resonance at around 0.8 MeV) than in the case of $^{238}$U, having as consequence more pronounced variations of fragment and prompt neutron emission data around the $E_n$ of resonances compared to the previous studied case of $^{238}$U (Tudora et al, 2012).

2.2. PbP calculation of average prompt neutron quantities as a function of TKE

Taking into account the deterministic manner of generating the fragmentation range in the frame of the PbP model (as described in (Tudora, 2013)) for the computational point of view it is more convenient to average the primary multi-parametric matrices (such as $v(Z,A,TKE)$, $s(Z,A,TKE)$) over $P(Z)$ and the double distributions $Y(A,TKE)$ reconstructed from the experimental single ones $Y(A)$, $TKE(A)$ and $\sigma_{TKE}(A)$ according to:

$$Y(A,TKE) = Y(A) \frac{1}{\sqrt{2\pi\sigma_{TKE}(A)}} \exp\left(\frac{(TKE - TKE(A))^2}{2(\sigma_{TKE}(A))^2}\right)$$

(1)

The reconstructed double distributions are verified by comparing the TKE distributions calculated as:

$$Y(TKE) = \sum_A Y(A,TKE) / \sum_A Y(A)$$

(2)


$\langle v \rangle$(TKE) and $\langle s \rangle$(TKE) are calculated at 14 incident energies where FF distributions were measured. Examples of such calculations at three $E_n$ values are given in Figs. 5 and 6 (PbP results being plotted with different symbols). As it can be seen in Fig.5 the behaviour of $\langle v \rangle$(TKE) as a function of $E_n$ reveals two interesting facts: the slope $dTKE/dv$ does practically not vary with $E_n$ and the flattening of $\langle v \rangle$ at low TKE values is more pronounced at low incident energies and it is diminishing with increasing $E_n$.

2.3. Dependence on TKE of average model parameters issue from the PbP treatment

Average model parameters as a function of TKE can be easily obtained by averaging the corresponding multi-parametric matrices as following:

$$< \text{param} > (TKE) = \sum_A \text{param}(A,TKE)Y(A,TKE) / \sum_A Y(A,TKE)$$

(3)

where $<\text{param}>$ means $<E_r>$, $<S_n>$ and $<a>$. They exhibit nice and regular behaviours as a function of TKE that can be fitted well. An example for $E_n = 0.5$ MeV is given in Fig. 7 where the PbP results according to Eq.(3) are plotted with full squares and the corresponding polynomial fits with red lines.

The obtained average parameter dependences on TKE allow the use of the most probable fragmentation approach (meaning the Los Alamos (LA) model with subsequent improvements) giving the $\langle v \rangle$(TKE) and $\langle s \rangle$(TKE) results plotted with solid lines in Figs. 5 and 6. This possibility to use the LA model has as advantages a very short calculation time compared to the PbP model and the Monte-Carlo treatment and the
possibility to extend the TXE range (especially at lower TKE values compared to the PbP calculations).

![Graph](image_url)

Fig. 5: PbP calculations of $\langle v(TKE) \rangle$ at $E_n = 0.5$ MeV (red squares), $3$ MeV (blue circles) and $5$ MeV (green triangles). Results of the most probable fragmentation approach using average parameters dependences on TKE from the PbP treatment are plotted with solid lines in the same colours as the corresponding PbP results.

![Graph](image_url)

Fig. 6: PbP calculations of $\langle v(TKE) \rangle$ at $E_n = 0.5$ MeV, $3$ MeV and $5$ MeV (with the same symbols as in Fig. 5). Results of the most probable fragmentation approach are plotted with solid lines in the same colours as the corresponding PbP results.
2.4. Consistency of PbP model calculations

The consistency of the present calculations can be proven by comparing the total average parameters and total average prompt emission quantities obtained by averaging the respective parameters and quantities given as a function of $A$ over $Y(A)$ with quantities given as a function of TKE averaged over $Y$(TKE) as following:

$$< \text{param} > = \sum_{TKE} \frac{< \text{param} > (TKE) \cdot Y(TKE)}{\sum_{TKE} Y(TKE)}$$

(4.1)

compared to:

$$< \text{param} > = \sum_A \frac{\text{param}(A) \cdot Y(A)}{\sum_A Y(A)}$$

(4.2)

and

$$< \nu_p > = \sum_{TKE} \frac{< \nu > (TKE) \cdot Y(TKE)}{\sum_{TKE} Y(TKE)}$$

(5.1)

compared to

$$< \nu_p > = \sum_A \frac{\nu(A) \cdot Y(A)}{\sum_A Y(A)}$$

(5.2)
where the notation \( \text{param} \) means \( E_r, TKE, Sn \). In the case of \( \langle C \rangle = A_0/\langle a \rangle \), \( \langle \text{param} \rangle \) means the average level density parameter \( \langle a \rangle \) obtained by averaging \( a(A) \) over \( Y(A) \) and respectively \( a(TKE) \) over \( Y(TKE) \).

In Table 1 an example of such a comparison is given for calculations performed at \( E_n = 3 \) MeV.

Table 1. Total average model parameters and prompt neutron multiplicity obtained by averaging over \( Y(A) \) and over \( Y(TKE) \)

<table>
<thead>
<tr>
<th>Total average parameter and multiplicity</th>
<th>Aver. over ( Y(A) )</th>
<th>Aver. Over ( Y(TKE) )</th>
<th>Deviation (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \langle E_r \rangle ) (MeV)</td>
<td>188.210</td>
<td>188.2096</td>
<td>0.0002</td>
</tr>
<tr>
<td>( \langle TKE \rangle ) (MeV)</td>
<td>170.181</td>
<td>170.167</td>
<td>0.10</td>
</tr>
<tr>
<td>( \langle Sn \rangle ) (MeV)</td>
<td>5.6706</td>
<td>5.7856</td>
<td>2.03</td>
</tr>
<tr>
<td>( \langle C \rangle = A_0/\langle a \rangle ) (MeV)</td>
<td>11.2526</td>
<td>11.1417</td>
<td>0.99</td>
</tr>
<tr>
<td>( \langle \nu \rangle )</td>
<td>2.7492</td>
<td>2.7756</td>
<td>0.96</td>
</tr>
</tbody>
</table>

As it can be seen (deviations being given in the last column of Table 1) the values of total average parameters obtained by averaging over \( Y(TKE) \) are very close to the values obtained by averaging over \( Y(A) \). Also very close values of total average multiplicity are obtained by averaging \( \nu(A) \) over \( Y(A) \) and \( \langle \nu \rangle (TKE) \) over \( Y(TKE) \) (see the last line of Table 1).

3. Conclusions

Model calculations of important quantities characterizing both pre- and post-scission stages of the fission process are performed.

In the pre-scission stage neutron induced cross-sections are calculated using the refined statistical model for fission as well as a recent deformed optical model parameterization and new average resonance data. For the first time the calculated fission cross-section describes very well the pronounced sub-barrier resonances of experimental data.

The correlation between the sub-barrier resonances of the fission cross-section and the non-statistical fluctuations of quantities characterizing both the fission fragments and the prompt neutron emission (already discussed for \( ^{238}\text{U}(n,f) \)) are revealed in the case of \( ^{234}\text{U}(n,f) \), too.

In the post-scission stage Point-by-Point model calculations of all prompt neutron data are performed. The primary multi-parametric matrices provided by the PbP model were averaged over experimental fragment distributions recently measured at IRMM at incident energies covering the range 0.2 – 5 MeV. The \( \langle \nu \rangle (TKE) \) calculation, for the first time made at many incident energies, has revealed two interesting facts: the slope \( dTKE/d\nu \) practically does not vary with the incident energy and the flattening of \( \langle \nu \rangle \) at low TKE values is more pronounced at low incident energies.

Average model parameters (energy release, average neutron separation energy and level density parameter) as a function of TKE, resulted from the PbP treatment, exhibit nice and regular behaviours that can be fitted well. The resulted dependences on TKE allow the use of the most probable fragmentation approach (Los Alamos model with subsequent improvements) having as advantages a very short computational time compared with the PbP and Monte-Carlo treatments and also the possibility to extend the TKE range especially at lower TKE values compared to the PbP calculations.
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