Sub-barrier resonance fission and its effects on fission fragment properties

exemplified on ²³⁸U(n,f) and ²³⁴U(n,f)

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WONDER-2012, Aix-en-Provence

I. Correlation between the sub-barrier resonant behaviour of fission xs of non-fissile (fertile) actinides (*pre-scission stage*) and the non-statistical fluctuations of their FF and prompt neutron data (*post-scission stage*) around En of sub-barrier resonances

II. <u>Pre-scission stage</u>: calculation of neutron induced xs focusing the fission xs., in the frame of the *refined statistical model for fission with sub-barrier effects* (Vladuca et al, STATIS code). Applied in this work to $n+^{234,238}$ U; extended to take into account the multi-modal fission (exemplified here for $n+^{238}$ U).

III. <u>Post-scission stage</u>: the prompt neutron and γ -ray emission is treated in the frame of the *Point-by-Point (PbP)* model. Total FF and prompt neutron quantities as a function of En obtained by averaging the PbP results as a function of fragment over the FF distributions reveal variations around En of sub-barrier resonances of the fission cross-section.



The correlated behaviour of the fission xs and FF properties is better outlined in the cases ${}^{238}U(n,f)$ and ${}^{234}U(n,f)$ because these fertile nuclei benefit of experimental FF data measured at many En with a fine grid in the region of sub-barrier resonances (measurements performed at IRMM for both ${}^{234,238}U(n,f)$)

The correlated behaviour can be observed in the case of 232 Th(n,f) too, but unfortunately the existing experimental FF data are measured only at a few En without a fine grid around En of sub-barrier resonances of the fission cross-section as in the cases of 238,234 U(n,f).

The correlation makes the link between the two stages of fission PRE- and POST-SCISSION usually treated by 2 different classes of models

<u>Pre-scission</u>: <u>one single nucleus</u> \rightarrow the evolution of this CN on the fission path, with the change of shape from g.s. (equilibrium) deformation passing trough different stages of deformation up to the rupture point. In this stage the neutron induced fission is in competition with other channels (n,n), (n,n'), (n,γ) , treated by the modeling of nuclear reaction mechanisms. The main quantity of this stage is σ_{f} obtained concomitantly with σ_{el} , σ_{in} , σ_{γ} as a function of En. (here of interest only the En range where one CN in formed) Post-scission: many nuclei (FF resulted from many possibilities of CN fragmentation), each FF 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. These quantities can be as a function of fragment (TKE(A), v(A), $\varepsilon(A)$ etc. at a given En) or can be average quantities as a function of En (<TKE>, <Er>, < v_p >, spectra, <E γ > and so on)

The correlation between σ_f (pre-scission/one nucleus) and quantities of post-scission (involving many nuclei) can be quantitatively analyzed in a <u>consistent and coherent manner</u> by taking into account the behaviour of <u>average prompt fission quantities</u> (obtained by averaging the quantities as a function of fragment over the FF distributions)

Sub-barrier resonances of $\sigma_f \rightarrow$ reflected by an increase of the fission channel population in the pre-scission stage \rightarrow leading to an increase of FF distributions in the post scission stage at En values of resonances.

Variation of Y(A) exemplified for the FF range (PbP treatment) $\rightarrow \langle A_H \rangle$, $\langle Er \rangle$



In the En range where only the first fission chance is involved (that is of interest in this case) the total relative population of a given channel (such as fission, gamma capture, elastic and inelastic scattering by CN mechanism) can be given by the ratio of the respective channel cross section to the CN formation cross section $P_{i} = \langle P_{i} \rangle \langle P_{i} \rangle \langle P_{i} \rangle$

$$P_{n\alpha'}(E) = \sigma_{n\alpha'}(E) / \sigma_{CN}(E)$$



The variation of Y(A) around En of sub-barrier resonances (0.95 MeV, 1.25 MeV) is visibly reflected by the behaviour of <A_H> and <Er> as a function of En:
<Er> is obtained by averaging Q-values of FF pairs (forming the FF range of the PbP treatment) over Y(A) and P(Z) distributions. Q-values and P(Z) do not change with En
→ Consequently the <Er> dependence on En is given only by Y(A)

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²³⁴U(n,f)

Non-statistical fluctuations of quantities characterizing the FF are observed around the incident energies where the fission xs exhibits sub-barrier resonances (for instance visible variations are around 0.35 MeV and especially at around 0.8 MeV where the fission cross-section exhibits a high resonance)

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• <Sn1> dependence on En is given only by Y(A) (Sn1 of pairs do not change with En).

• <TXE> variations around 0.5 and 0.8 MeV still visible even if in the figure they seem to be less pronounced (really these variations are of the same order of magnitude as the variations of other quantities mentioned above)

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 $\langle TXE \rangle$ variations (at around 0.95 and 1.25 MeV) seem to be less pronounced compared to other quantities (like $\langle Er \rangle$, $\langle A_H \rangle$) because the difference between $\langle Er \rangle$ and $\langle TKE \rangle$ varies less than $\langle TKE \rangle$. Really the $\langle TXE \rangle$ variations are almost of the same order of magnitude as of other quantities mentioned above.

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The PbP result of total average prompt neutron multiplicity (obtained by averaging the PbP matrix v(Z,A,TKE) over the recent experimental Y(A,TKE) (IRMM) reveals visible non-statistical fluctuations around the En where the fission xs exhibits sub-barrier resonances.



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To synthesize:

In both cases ${}^{238, 234}$ U(n,f) the sub-barrier resonances of the fission xs, reflected by an increase of fission exit channel population at the respective En values, lead in the post-scission stage to an increase of FF distributions around the En of resonances. This fact was proven by the fragment observables and prompt neutron data obtained by averaging the quantities corresponding to fragment pairs over the fragment distributions Y(A), P(Z), such as <Er>, <Sn1> and so on and also by the experimental <TKE> data. All these quantities exhibiting visible variations around the En of resonances.

The sub-barrier fission xs <u>resonances of</u> ²³⁴U (placed at around 0.3 MeV, 0.5 MeV and 0.8 MeV) are <u>much more pronounced</u> (especially the resonance at 0.8 MeV is very high) compared to the <u>resonances of</u> ²³⁸U (placed at around 0.95 MeV and 1.25 MeV). Consequently <u>the effect in the properties of fragments and in the prompt</u> <u>neutron emission data is also more pronounced in the case of</u> ²³⁴U <u>compared to</u> ²³⁸U, as it is proved by the present results. The multi-modal fission concept also can give an explanation of the correlation between the sub-barrier resonances of the fission xs and the pronounced variation of experimental <TKE> and of other FF and prompt neutron data at almost the same En values.

In the frame of the multi-modal fission the <u>coherence</u> between the stages of pre-scission (<u>one</u> fissioning <u>nucleus</u>) and post-scission (<u>many nuclei</u>, FF) is assured by the behaviour of the <u>modal fission xs</u> (usually of S1, S2, SL modes) as a function of En directly correlated with the behaviour of <u>both modal</u> <u>prompt fission quantities</u> as a function of En: -the modal distributions ($Y_m(A)$, TKE_m(A), $\sigma_{TKEm}(A)$) and -the modal average quantities (such as $\langle TKE \rangle_m$, $\langle Er \rangle_m$ and so on) the index m means S1, S2, SL



- En (MeV) • Calculated fission mode xs exhibit resonances at around 0.95 and 1.25 MeV
- The experimental fission mode weights (branching ratios) exhibit pronounced variations around the fission xs resonances → an increase of the S1 weight and a respective decrease of the S2 weight

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The average TKE of S1 mode ($\langle TKE \rangle_{S1}$) has always the highest value (because of the split in almost spherical fragments in connection with the closed shells N=82 and Z=50 and the lowest distance between their charge centers) \rightarrow the increase of $\langle TKE \rangle_{S1}$ around the resonance energies determines the behaviour of the total $\langle TKE \rangle$.



This fact is illustrated in the following example where 2 consecutive closed En values (0.9 and 0.925 MeV) at which experimental <TKE> data exhibit a visible variation (increase) with En are taken.



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II Neutron induced xs calculation, with the fission channel treated

- \succ classically (without modes): $n+^{234, 238}U$
- ➢ multi-modal fission concept: n+²³⁸U

In the En range where only the first fission chance in involved

- **DI mechanism** → **CC** (ECIS) + deformed OM parameterizations (with dispersion)
- CN mechanism → statistical model with sub-barrier effects (STATIS code) including the extended model in the frame of the MM fission concept (5 channels in competition neutron scattering, capture and 3 fission channels S1, S2, SL)

After incident neutron absorption, the CN is populated in **class I states**. From these states it can decay by neutron emission, gamma transitions and "direct fission" or it can undergo a shape change by transition to a **class II state** (absorption in the isomeric well). The flux fraction absorbed in the second well is described by an **absorptive** (imaginary) potential in the deformation region of the second well. This absorbed fraction can decay i) by fission penetrating the outer barrier (the so-called "indirect fission"), ii) by radiative transition to the isomeric state followed by "isomeric fission" or iii) by another change of shape returning to a class I state after penetrating the inner fission barrier. The fission probability is given by the sum of <u>three components</u>: the direct, indirect and isomeric fission. If the fission-mode concept is taken into account, then the fission probability through each mode is also taken as a sum of the direct, indirect and isomeric fission components.

For each transition state a **double-or triple-humped barrier** is taken (parabolas). The **dumping of vibrational states** in the **isomeric well** is taken by **the imaginary part of the potential** in the region of the second well (also parabolic shape with respect to the deformation parameter).



Tudora et al., Nucl. Phys. A 890-891 (2012) 77















III. Prompt neutron emission calculation

PbP model provides as primary results the <u>multi-parametric</u> <u>matrixes</u> v(Z,A,TKE), N(Z,A,TKE), ε(Z,A,TKE), Εγ(Z,A,TKE) and so on.

FF range: the entire FF mass range covered by a mass distribution with a step of 1 mass unit. For each A two or four Z are taken as the nearest integer values above and below the most probable charge (taken as UCD corrected with a charge polarization)

Average quantities are obtained by averaging the corresponding multi-parametric matrix *quantity*(*Z*,*A*,*TKE*) over FF distributions: **P**(**Z**) (taken as a narrow gaussian)

Y(A, TKE) usually experimental data (or models TKE(A), simulations Y(A))

<u>Most probable fragmentation approach</u> (improved LA) – using average model parameter values (depending on En / E*) obtained from the PbP treatment used for the improvement and validation of the systematic of LA parameters, 2009)



The present PbP calculations as well as the most probable fragm. approach with average param. from the PbP treatment confirmed the following <u>predictions</u>: previous calculations reported in 2004 and model parameter values provided by the systematic (2009)

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TKE (MeV)



CONCLUSIONS

• The correlation between the sub-barrier resonant behaviour of $\sigma(n,f)$ of fertile actinides (pre-scission) and the visible fluctuations of their fragment and prompt neutron data (post-scission) around En of sub-barrier resonances is outlined and supported by quantitative results in the cases ²³⁸U(n,f) and ²³⁴U(n,f).

Through the PbP treatment of prompt emission and the multi-modal fission concept (also included in the statistical model with sub-barrier effects for nuclear reactions) we arrived at a quantitative explanation of the observed fluctuations.

• New calculations of neutron induced xs of ^{238,234}U using recent CC deform. optical model parameteriz., recent values of s-wave resonance data, the refined statistical model for fission. The consistency of present calc. is proven by all integral and differential xs in good agreement with experimental data

PbP model used with experimental Y(A,TKE) to provide average quantities characterizing the fission fragments and the prompt neutron emission, allow a) the quantitative support of the correlation mentioned above
 b) to validate the prediction of previous calculations and systematics in the case of ²³⁸U(n,f)