Advanced fission models in nuclear data calculations

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Abstract. Transition states at the saddle points and superdeformed or hyperdeformed states in the secondary wells of multiple-humped potential barriers play an important role in low-energy fission processes. In the present work discrete collective spectra at large nuclear deformations are predicted by means of the dinuclear model and combined with the optical model for fission of the Empire-3 system of codes. The formalism is applied to the $^{233}$U$(n, f)$ reaction and the computed cross section compared with recent experimental results of the n_TOF Collaboration. Angular anisotropies of fission fragments are evaluated with an improved version of the scission-point model.

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1. Introduction

Accurate neutron cross sections are of great importance to the design of innovative systems for nuclear energy production and nuclear waste incineration, such as accelerator driven systems and Generation IV fast reactors, whence the need for high-precision measurements and evaluations of neutron data based on reliable reaction models and codes.

In order to perform accurate measurements of radiative capture and fission cross sections, an innovative neutron time-of-flight (n_TOF)¹ facility has been constructed at CERN, Geneva. Neutrons are produced by 20 GeV/c protons from the CERN Proton Synchrotron impinging on a lead block surrounded by a water layer acting as coolant and moderator of the neutron spectrum, which spans several orders of magnitude, from thermal energy to approximately 1 GeV.

As far as fission is concerned, during the first experimental campaign (2002-2004), the n_TOF Collaboration measured $(n, f)$ cross sections of nuclides relevant to the $Th – U$ fuel cycle ($^{232}$Th, $^{233,234,235}$U) and of minor actinides commonly appearing in the spent fuel of present-day reactors ($^{237}$Np, $^{241,243}$Am, $^{245}$Cm). In the second experimental campaign, started in 2008, measurements are planned or under way for fission cross sections of $^{240,242}$Pu and angular distributions of fission fragments ($^{232}$Th, $^{234}$U, $^{237}$Np).

The main purpose of the present project is to establish a set of advanced fission models and computer codes suited to the theoretical interpretation of the n_TOF measurements, and, as an useful by-product, to the evaluation of neutron cross sections of the mentioned nuclides in the energy range of interest to fission reactors. The preliminary calculations described in the
present work are focused on the $^{233}U(n,f)$ cross section, measured by the n_TOF Collaboration from thermal energy up to 20 MeV$^2$.$^3$.

2. Models, codes and results

The present section is dedicated to a concise description of the system of codes used in calculating fission cross sections, to the models adopted in determining part of the fission input and to the comparison of calculated results with experimental data.

2.1. The fission channel of the Empire-3 system

Empire-3, used in cross section calculations, is a modular system of nuclear reaction codes, based on advanced nuclear models and designed for calculations over a broad range of incident energies and incident particles (neutrons, protons, photons, light and heavy ions)$^4$.

The code accounts for the major nuclear reaction mechanisms, including direct, pre-equilibrium and compound nucleus ones, by means of a generalized optical model, multistep-direct and multistep-compound models, exciton model with cluster emission and statistical Hauser-Feshbach model.

With particular reference to the fission channel, Empire-3 can describe transmission through multi-humped barriers parametrized analytically or defined numerically. In the present work, use is made of the former option: the barrier as a function of a deformation parameter, $\beta$, along the fission path, is parametrized by $N$ smoothly joined parabolas

$$V_i(\beta) = E_{fi} + \frac{(-1)^i}{2} \mu (\hbar \omega_i)^2 (\beta - \beta_i)^2. \quad (i = 1, N)$$  (1)

Here, odd values of index $i$ refer to humps, even values to wells, $\mu \approx 0.054A^{5/3}$ MeV$^{-1}$ is a semiempirical inertial mass parameter, while heights $E_{fi}$ and curvatures $\hbar \omega_i$ can either be fitted on theoretical values or adjusted on experimental data. In principle, $N$ is an arbitrarily high odd number, but in the present work calculations are performed for $N = 3$ (two humps) and $N = 5$ (three humps).

On the humps and in the wells, the fissioning nucleus has a discrete and a continuous spectrum. The discrete part consists of rotational bands built on collective (vibrational) states or on non-collective (few quasi-particle) states, each state being labelled with its angular momentum, $J$, projection of $J$ on the nuclear symmetry axis, $K$ and parity $\pi$

$$E_n(J, K, \pi) = E_{fi} + \epsilon_n(K, \pi) + \frac{\hbar^2}{2I_n} \left[ J(J+1) - K^2 \right] . \quad (2)$$

The collective band-head energies, $\epsilon_n(K, \pi)$ and moments of inertia, $I_n$, are theoretically evaluated within the framework of the dinuclear model, as discussed in the next subsection.

Above the lowest-lying non-collective excitation, a two-quasiparticle state in the case of an even-even compound nucleus, like $^{234}U$, the discrete spectrum is replaced with a continuum, i.e. a non-collective level density computed in the frame of the generalized superfluid model, with an energy-dependent collective enhancement factor, as described in Ref.$^4$.

The intermediate potential wells may have an imaginary component, according to the optical model for fission, which simulates the damping of collective states in the wells themselves due to the coupling with non-collective states in the ground-state well. In this way, the transmittance through the multiple-humped barrier, computed in JWKB approximation as described in Ref.$^5$, will be the sum of a direct contribution, due to the probability flux that crosses the whole barrier, and an indirect contribution, due to the flux absorbed in an intermediate well and reemitted in the fission channel.
In the case of the $^{233}U(n,f)$ reaction, however, vibrational states inside the wells do not give a sizable contribution, because the excitation energy is always greater than the barrier. Therefore, the analysis will be focused on collective transition states on the humps.

2.2. Transition states in the dinuclear model

A theoretical estimate of the fission barrier parameters, such as heights and curvatures of the parabolic approximation given by formula (1), can be obtained by self-consistent microscopic methods (Hartree-Fock-Bogoliubov or relativistic mean field) or by macroscopic-microscopic methods (Strutinsky's shell and pairing corrections to liquid drop energies); after defining a set of collective coordinates suited to the description of nuclei at large deformations, the potential energy of the system as a function of convenient subsets of the above mentioned coordinates lends itself to the determination of fission paths.

In the present work we have used the macroscopic-microscopic method, with shell corrections evaluated by means of the two-centre shell model[6], suited to the description of a system of two clusters (prefragments) of masses $A_1$, $A_2$, charges $Z_1$, $Z_2$ in terms of mass asymmetry, $\eta = (A_1 - A_2)/(A_1 + A_2)$, charge asymmetry, $\eta_Z = (Z_1 - Z_2)/(Z_1 + Z_2)$, distance, $d$, of cluster centres (elongation) and cluster deformations, $\beta_i = c_i/a_i$, ($i = 1, 2$), where $c_i$ and $a_i$ are the major and minor semiaxis of the $i^{th}$ cluster, schematized as an ellipsoid.

The potential energy, $U$, of the system is the sum of the shell-corrected potential energies of the two clusters, their nuclear and Coulomb interaction and the rotational energy. Fission paths can be determined on the potential energy surface depending on mass asymmetry and elongation, once the other collective variables have been fixed so as to minimize $U$ for given $\eta$ and $d$.

In their equilibrium condition, the two ellipsoids are in a pole-to-pole configuration, with an elongation slightly greater than the sum $c_1 + c_2$ of the major semiaxes by a quantity of the order of 0.5\,\text{fm}, owing to the interplay of the attractive nuclear and repulsive Coulomb interactions.

The model predicts for the compound nucleus $^{234}U$ a three-humped fission barrier with peak heights decreasing with increasing elongation. At each point on the fission path, the nuclear wave function is a superposition of different dinuclear configurations; the dominant one is $^{230}Th + ^4He$ at normal deformation, $^{212}Po + ^{22}O$ at the first hump, $^{194}Os + ^{40}S$ at the second hump, $^{128}Sn + ^{106}Mo$ at the third hump and $^{132}Te + ^{102}Zr$ at the scission point. Charge and mass multipole moments and moments of inertia can be computed in the frame of the dinuclear model as described in Ref. [7].

In principle, the collective spectrum of a dinuclear system is obtained by diagonalizing a Hamiltonian depending on 15 collective coordinates, mass asymmetry, elongation, 3 Euler angles describing rotations of the system as a whole, 3 + 3 Euler angles describing rotations of the two clusters, 2 + 2 Bohr coordinates describing internal excitations of the two clusters, on the assumption that their deformations are quadrupolar. In practice, not all degrees of freedom have the same importance and several simplifying assumptions can be made.

If one is interested in the lowest-lying collective bands of both parities of a system with stable octupole deformation, like a compound nucleus at the saddle points and in the intermediate wells of a fission barrier, use can be made of the bending approximation[8], where the relevant collective motions are basically three: exchange of nucleons between the two clusters, rotations of the dinuclear system as a whole and oscillations of the heavy cluster around its equilibrium position inside the system. In fact, the frequencies of oscillation of the light deformed cluster around the pole-to-pole orientation of the system are much higher than those of the heavy cluster and give rise to collective states at higher energy than the first few-quasiparticle states. The bending Hamiltonian can be diagonalized analytically and the resulting spectrum consists of rotational bands of type (2), with band-head energies that do not depend on parity

$$
\epsilon_n(K, \pi) = \hbar \omega_b (2n + |K| + 1).
$$
In other words, when $K \neq 0$, states of the same angular momentum $J$ and opposite parity are degenerate. As an example, Fig. 1 shows the calculated collective spectrum of $^{234}$U at the second saddle point. Such a degeneracy does not appear in the spectrum at ground-state deformation, owing to the dominant contribution of the mononucleus configuration, which does not have a stable octupole configuration. There, the theoretical collective bands turn out to be in excellent agreement with the experimental ones.

Since we are interested in reproducing the measured fission cross section of $^{233}$U up to 20 MeV, collective spectra at the saddle points and in the intermediate wells of fission barriers are computed also for the fissioning nuclei $^{233}$U and $^{232}$U, in order to evaluate second- and third-chance fission ($(n, n'f)$ and $(n, 2nf)$, respectively). While the spectra of transition states are kept fixed, we take the freedom to adjust barrier heights and curvatures for a better fit of experimental data. Results are shown in Fig. 2 for three-humped and two-humped barrier calculations.

![Figure 1](image1.png)

**Figure 1.** Collective spectrum of $^{234}$U at the second saddle point.

![Figure 2](image2.png)

**Figure 2.** Calculated and experimental $^{233}$U($n, f$) cross sections. Experimental data are taken from Ref.[3]

### 2.3. Distributions of fission fragments in the scission-point model

The fission cross section alone is not enough to check the transition states, since it can be equally well reproduced with different sets of collective bands. Angular distributions of fission fragments are much more sensitive to the $(K, \pi)$ quantum numbers of the bands than the fission cross section itself.

In order to evaluate angular distributions in a static fission model, two extreme assumptions can be made: either angular distributions depend on the $(K, \pi)$ quantum numbers of states at the outer saddle point, approximately conserved in the descent from saddle to scission, or on the $(K, \pi)$ states at the prescission configuration. Testing the validity of the first assumption requires modifying the statistical model routines of the Empire code and is under study, testing the second assumption is possible within the framework of the scission-point model[9].

The model assumes the existence of a well defined scission configuration, up to which strong non-adiabatic effects maintain statistical equilibrium among various degrees of freedom and
beyond which there is no nuclear interaction of the fragments. It is thus possible to calculate the probabilities of different scission configurations and, consequently, to describe various fission characteristics, such as mass, charge and kinetic energy distributions of the fragments.

As for angular distributions, if the dinuclear system at the scission point has excitation energy \( E^* \), angular momentum \( J \), with projection \( M \) on the direction of the incident neutron beam and parity \( \pi \), the angular distribution of fission fragments reads

\[
W_{JM\pi} (A_1, A_2, Z_1, Z_2, E^*, \theta) = A(J) \left( \frac{2J + 1}{2} \right) \sum_{nK} \exp \left[ -\frac{E_{nJM\pi}}{T(E^*)} \right] |d_{MK}^J(\theta)|^2,
\]

where \( A(J) \) is a normalization constant, \( E_{nJM\pi} \) the eigenvalues of the dinuclear-model Hamiltonian, \( T(E^*) \) the nuclear temperature and \( d_{MK}^J(\theta) \) the Wigner reduced matrix elements. The angular anisotropy of fission fragments, \( W(\theta = 0)/W(\theta = \pi/2) \), evaluated with an improved version\(^{[10]} \) of the scission point model, as a function of incident neutron energy is compared with experimental data in Fig.3. The agreement between theory and experiments might be improved at low neutron energy by considering the contributions of bands built on quasiparticle states, not yet included in the model. In any case, further analysis is needed. As a perspective for future work, we intend to use the dinuclear model for a consistent evaluation of the collective enhancement factor of level densities at the saddle points and to extend the present analysis to the fission cross section of the adjacent nucleus \(^{234}U\), also recently measured by the \( n_\text{TOF} \) Collaboration\(^{[12]} \), where collective bands in the intermediate wells play a sizable role in the under-threshold region.

References

[1] Gunsing F et al (the \( n_\text{TOF} \) Collaboration) 2007 Nucl. Instr. Meth. B \textbf{261} 925 and references therein.
[2] Calviani M et al (the \( n_\text{TOF} \) Collaboration) 2009 Phys. Rev. C \textbf{80} 044604.
[3] Belloni F et al (the \( n_\text{TOF} \) Collaboration) 2011 Eur. Phys. J. A \textbf{47} 2.
[4] Herman M et al 2007 Nucl. Data Sheets \textbf{108} (2007) 2655.
[5] Sin M et al 2006 Phys. Rev. C \textbf{74} 044608.
[6] Maruhn J and Greiner W 1972 Z. Phys. \textbf{251} 431.
[7] Shneidman TM et al 2000 Nucl. Phys. A \textbf{671} 119.
[8] Adamian GG et al 2007 Phys. At. Nuclei \textbf{70} 1350.
[9] Wilkins BD, Steinberg EP and Chasman RR 1976 Phys. Rev. C \textbf{14} 1832.
[10] Andreev AV et al 2004 Eur. Phys. J. A \textbf{22} 51.
[11] Simmons JE and Henkel RL 1960 Phys. Rev. \textbf{120} 198; Leachman RB and Blumberg L 1965 Phys. Rev. \textbf{137} B814.
[12] Paradela C et al (the \( n_\text{TOF} \) Collaboration) 2010 Phys. Rev. C \textbf{82} 034601.