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Modelling Neutron-induced Reactions on $^{232-237}\text{U}$ from 10 keV up to 30 MeV

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Comprehensive calculations of cross sections for neutron-induced reactions on $^{232-237}\text{U}$ targets are performed in the 10 keV–30 MeV incident energy range with the code EMPIRE-3.2 Malta. The advanced modelling and consistent calculation scheme are aimed at improving our knowledge of the neutron scattering and emission cross sections, and to assess the consistency of available evaluated libraries for light uranium isotopes. The reaction model considers a dispersive optical potential (RIPL 2408) that couples from five (even targets) to nine (odd targets) levels of the ground-state rotational band, and a triple-humped fission barrier with absorption in the wells described within the optical model for fission. A modified Lorentzian model (MLO) of the radiative strength function and Enhanced Generalized Superfluid Model nuclear level densities are used in Hauser-Feshbach calculations of the compound-nuclear decay that include width fluctuation corrections. The starting values for the model parameters are retrieved from RIPL. Excellent agreement with available experimental data for neutron emission and fission is achieved, giving confidence that the quantities for which there is no experimental information are also accurately predicted. Deficiencies in existing evaluated libraries are highlighted.

CONTENTS

References

164

I. INTRODUCTION	138
II. REACTION MODELS AND PARAMETERS	139
A. Optical Model and Direct Excitation of Collective Levels	139
B. Statistical Models	140
1. Level Densities at the Equilibrium Deformation	141
2. Level Densities at the Saddle Points	141
3. Fission	142
III. RESULTS AND DISCUSSION	147
A. Total, Elastic and Nonelastic Cross Sections	147
B. Inelastic, $(n,2n)$ and $(n,3n)$ Cross Sections	150
C. Radiative Capture Cross Sections	154
D. Fission Cross Sections	154
E. Overview of Calculated Cross Sections for $n+^{232-237}\text{U}$ Reactions	163
IV. CONCLUSIONS	163
Acknowledgments	164

I. INTRODUCTION

Nuclear Energy Agency started a new international collaboration called CIELO (Collaborative International Evaluated Library Organisation) with the main goal to improve our understanding of neutron reactions on key isotopes that are important in nuclear applications [1–3]. Among the nuclei selected for the pilot CIELO project, a prominent role is played by the major actinides, $^{235,238}\text{U}$ and ^{239}Pu , for which fission is an important channel.

Recently, we published a formalism [4] to calculate fission cross sections based on the extension of the optical model for fission [5–11] and on the recursive method [12]. Developed formalism was used in calculations on $^{234,235,238}\text{U}$ and ^{239}Pu targets [4]. Obtained fission cross sections of Ref. [4] agree with the $^{235,238}\text{U}(n,f)$ Neutron Standards within 3% [13, 14]. Such good description allows a proper modelling of the main competing channels – neutron capture, elastic and inelastic scattering, and multiple neutron emission. However, studies of Ref. [4] focused only on calculations of neutron-induced fission cross sections. Further theoretical studies are needed for nuclear data evaluation of many actinides, and to assess the status of data in existing evaluated nuclear data libraries.

There is no unique set of models and parameters which

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can describe accurately the experimental reaction data, and especially the fission cross section. At a recent IAEA meeting on “Recommended Input Parameters for Fission Cross Section Calculations” [15] consultants pointed out: *... some problems have been identified since the latest RIPL-3 database release [16], in particular, RIPL-3 input parameters for fission have not been comprehensively validated against available experimental data, and therefore do not guarantee a reproducible and/or accurate calculation of fission cross sections ...*

While both differential and integral (benchmark) experimental data provide extremely important constraints for evaluated data, nuclear modelling is required to fill the gaps in experimental data and make evaluated data files both complete and physically consistent. Calculations of neutron-induced reactions on the uranium isotopic chain have been undertaken before, e.g., by Young *et al.* [17], Iwamoto *et al.* [18], Goriely *et al.* [19, 20], and Romain *et al.* [21]. Goriely *et al.* [19, 20] explored the predictive capability of microscopic models, but used a simplified fission formalism only valid above the fission barriers. Romain *et al.* [21] used triple-humped barriers to describe uranium isotopes, but with a completely different fission formalism. Other references studied the reaction modelling used to evaluate uranium data in existing libraries ENDF/B-VII [22, 23] (Young *et al.*), and JENDL-4 [24] (Iwamoto *et al.*), correspondingly. A consistent and reliable set of fission parameters as model independent as possible is the one which provides simultaneously a reasonable description of multiple fission chances induced by neutrons, photons, protons or direct transfer reactions leading to the same fissionable compound nucleus. A similar approach has been followed by CEA/DAM researchers for evaluations of neutron-induced reactions on actinide targets [21, 25], but they use different experimental constraints leading to different evaluated data [26].

Our goal is to undertake comprehensive and consistent model calculations of neutron-induced reactions on light uranium isotopes $^{232-237}\text{U}$ for incident energies from 10 keV up to 30 MeV using a validated regional optical model and the extended optical model for fission [4]. If the employed optical model gives a good description of the total and the direct component of neutron scattering cross sections, and the modelling reproduces accurately the measured fission cross section, then remaining cross sections could be predicted with higher accuracy. Calculated cross sections will also be used to assess the consistency of available evaluated libraries.

Systematic behaviour of neutron scattering and emission cross sections along the uranium isotopic chain is compared with selected evaluated data [22–24, 26, 27]. Comprehensive reaction modelling of neutron-induced reactions on ^{238}U target [28–30] is used as a guidance and for isotopic trend comparisons.

Nuclear reaction models and associated input parameters are described below in Sec. II and its subsections. In Sec. III and its subsections, calculations for each uranium isotope are described and compared with experi-

mental data from EXFOR [31] and selected previous evaluations [23, 24, 26]. Isotopic trends are also discussed. Finally, concluding remarks are given in Sec. IV.

II. REACTION MODELS AND PARAMETERS

Given the limited accuracy and availability of experimental data for the light uranium isotopes, nuclear reaction modelling plays a central role for nuclear data evaluations. Therefore, improvement and benchmarks of the modelling and of the model parameters are crucial for a better prediction of unknown cross sections. The present model calculations are based on a theoretical analysis with the nuclear modular system EMPIRE-3.2 [32, 33]. Nuclear reactions induced by neutrons on uranium targets in the studied energy range occur through the direct, pre-equilibrium and compound nucleus mechanisms. The optical and direct reaction models (see Sec. IIA), the pre-equilibrium exciton model, and the full featured Hauser-Feshbach statistical model [34] including width fluctuations as proposed by Hofmann-Richert-Tepel-Weidenmüller [35] (see Sec. IIB) are used to calculate the observables for neutron-induced reactions.

The starting values for other model parameters were taken from the International Atomic Energy Agency (IAEA) RIPL library [16], including the updated discrete level database. Some details of the employed theoretical modelling are discussed in the following section.

A. Optical Model and Direct Excitation of Collective Levels

The optical model is one of the fundamental theoretical tools which provides the basis of nuclear reaction modelling for nuclear reactions [36]. It is well known that an accurate calculation of the nucleon scattering from a well-deformed actinide nucleus using the optical model should include the coupling to the low-lying collective states using the coupled-channel (CC) method [37]. Tamura’s CC formalism [38] permits an exact solution of the CC equations for rotational nuclei.

The coupled-channel optical model code ECIS06 by Raynal [39] was used for all optical model calculations (within the EMPIRE-3.2 code system). The dispersion integrals were calculated externally using analytical solutions [40, 41]. Cross sections of the direct interaction on the ground state rotational band and the corresponding transmission coefficients for the incident neutron channel were calculated using a dispersive coupled-channel optical model potential of Capote *et al.* [42, 43] (RIPL 2408 [16]). This model is isospin dependent and approximately Lane consistent [44, 45] as demonstrated in Ref. [46]. Therefore, it can be used to describe nucleon induced reactions on all uranium isotopes.

Originally, the RIPL 2408 optical model coupled only five levels of the ground-state rotational band [43]. How-

ever, recent studies recommended to couple multiple bands for even-even targets [47–49], and to increase the number of coupled levels of the ground-state rotational band in odd targets [50]. We coupled up to nine levels of the ground state rotational band for odd- A uranium targets following Dietrich *et al.* [50]. However, for even uranium isotopes we kept the original number of coupled levels (five), and considered the important contribution of vibrational bands by Distorted Wave Born Approximation (DWBA) calculations using the ECIS06 optical model code [39]. For odd- A nuclei we have also used DWBA calculations on discrete collective levels from neighbor even-even nuclei to estimate the cross sections for fragmented vibrational states. These calculations were combined with the compound nucleus contribution to determine the inelastic scattering cross sections and corresponding angular distributions on low-lying discrete states.

Young *et al.* [17] pointed out that at higher excitation energies, where excited levels can not be identified, the measured neutron emission spectra for $n+^{238}\text{U}$ reactions by Baba *et al.* [51] are significantly higher than what can be accounted for by conventional compound-nucleus or preequilibrium calculations. Marcinkowski *et al.* [52] also observed a similar effect for measured neutron emission spectra from tungsten isotopes near 14 MeV of neutron incident energy, and proposed the existence of collective states in the continuum as a possible explanation. Later works by Dupuis *et al.* [53, 54] and Wienke *et al.* [55] suggested a microscopic origin of the additional collective strength in the continuum proposed by Marcinkowski *et al.* [52].

In this work we followed the empirical approach used by Young *et al.* (see Table VI in Ref. [17]), who postulated the existence of a series of collective 2^+ and 3^- states at excitation energies in the range $E_x=1\text{--}4$ MeV with dynamical deformations fitted to reproduce measured emission spectra at 14 MeV on ^{238}U target. Cross sections and angular distributions from these collective states were calculated by DWBA, and their strength was spread in the continuum using a Gaussian resolution function with $\sigma=70$ keV. The postulated collective levels for ^{238}U were used for all uranium isotopes. Those levels can be interpreted as collective levels in the continuum calculated by a QRPA method [54] or by a Tamura-Udagawa-Lenske quantum-mechanical preequilibrium model for deformed targets [55].

The assumed collective levels in the continuum lead to neutron emission spectra in good agreement with measured Kammerdiener *et al.* data on ^{235}U and ^{238}U targets at several incident energies [56], as well as to improved Monte Carlo simulations of time-of-flight spectra in LLNL pulsed-sphere experiments [57] for ^{235}U and ^{238}U targets. Additional collective strength in the continuum also proved to be important for the description of multiple neutron emissions.

B. Statistical Models

The statistical model used in the EMPIRE is an advanced implementation of the Hauser-Feshbach theory [34]. The exact angular momentum and parity coupling is observed. We considered the emission of neutrons and protons only along with the competing fission channel, as the emission of heavier charged particles is strongly suppressed owing to the high Coulomb barrier in actinides (about 20 MeV for protons). Fission transmission coefficients were calculated using the extended optical model for fission [4] as described in Sec. II B 3.

To account for correlations between incident and exit channels in elastic scattering (the width fluctuation corrections) the model proposed by Hofmann, Richert, Tepel and Weidenmüller (HRTW) [35] is used. The importance of considering the influence of direct reactions on calculated neutron inelastic scattering for well-deformed nuclei was recently highlighted [28, 58]. This effect increases the neutron inelastic scattering cross section on even-even uranium targets between 100 and 500 keV [28]. However, it has been shown in Ref. [58] that the impact on fission and capture cross sections is almost negligible, the increase in the inelastic cross section being compensated by a decrease in the elastic cross section. In this work we did not consider this effect. Therefore, calculated elastic (inelastic) cross sections may be slightly overestimated (underestimated) for neutron incident energies below 1 MeV.

Pre-equilibrium emission was taken into account by the EMPIRE module PCROSS featuring a one-component exciton model with gamma, nucleon and cluster emissions [59]. Default pre-equilibrium parameters were used in the EMPIRE calculations, except that the mean free path multiplier [59] (playing a role similar to the matrix element normalization constant in the exciton model) was taken as 1.8 for a better description of the high energy tail in $^{238}\text{U}(n,n')$ and $^{238}\text{U}(n,2n)$ reactions [28–30]. The nuclear single-particle state density was set to $A/13$ MeV $^{-1}$.

The full γ -cascade in the compound and residual nuclei was considered including both continuum and discrete transitions with a maximum multipolarity of 2 (i.e., only E1, M1 and E2 transitions were allowed). Gamma-ray transmission coefficients were obtained from γ -ray strength functions computed using the modified Lorentzian (MLO1) closed form [16]. Evaluated values of $2\pi\Gamma_\gamma/D_0$ determined from experimentally inferred values of Γ_γ and D_0 [16] were used to normalize the E1 gamma-ray strength function computed using the Modified Lorentzian model. The GDR parameters were retrieved from RIPL [16]; and M1 resonance parameters – from Ref. [60]. Giant Quadrupole Resonance E2 energy was taken after Ref. [61], and GQR width and peak cross sections – from Ref. [62].

Phenomenological level density functions were used to represent continuum of levels. The level densities, both at equilibrium deformation and at saddle points, are described with the Enhanced Generalized Superfluid Model (EGSM) [16, 32, 33] in the approximation $J \gg K$

which makes them similar to the level densities developed by Ignatyuk *et al.* in the Generalized Superfluid Model (GSM) [63, 64]. Note that in fission cross section calculations it is important to adopt the same level density formulation to represent the continuums of levels in the ground state and at the saddle point deformations, as the level density ratios between normal and deformed states is the truly relevant quantity. The adopted nuclear level densities have a very large impact on statistical model calculations, so we discuss a few related aspects in Secs. IIB 1 and IIB 2.

1. Level Densities at the Equilibrium Deformation

The starting values for the parameters of the EGSM level density functions were taken from the RIPL library [16]. The RIPL EGSM global systematics defines the level density parameter a at the neutron separation energy; its asymptotic value $\tilde{a}$ is tabulated in RIPL [16], and was derived by fitting the average s -wave neutron resonance spacings D_0 compiled in RIPL [16]. Fitted $\tilde{a}$ values are independent from the considered number of discrete levels. When level densities are calculated, a shift to the excitation energy is applied automatically to reproduce the cumulative number of levels at the energy corresponding to the highest level considered in the calculations. This shift is linearly decreased with increasing energy in such a way to reach a zero value at the neutron separation energy to preserve the level density value fitted to D_0 . Therefore, level densities below the neutron separation energy depend strongly on the number of discrete levels adopted.

Two types of level density optimization have been performed to describe the measured cross sections:

- a proper selection of the number of discrete levels based on the analysis of the cumulative level-density plots automatically generated by the EMPIRE code to avoid missing levels,
- the renormalization of the asymptotic values of the level density parameter $\tilde{a}$ defined by EGSM global systematics in the limit of a few percents.

The selection of the maximum number of discrete levels must be done carefully because experimental structure information needs to be preserved, but a right behaviour of the level density below the neutron separation energy is also needed. When the excitation energy increases the slope of the cumulative number of levels may decrease because of missing levels, with consequences on the level densities, and hence on the calculated cross sections. The level densities below the neutron separation energy have a strong impact on the radiative capture cross section and on the inelastic and (n,xn) cross sections below their maxima. The choice of the number of discrete levels must be also related to the number of collective levels involved in direct excitation (see Sec. IIA). To avoid matching

the level densities to discrete levels in a region where levels may be missing, the number of discrete levels used in our calculations is typically about half the number recommended in the RIPL library [16] (see Table I). A noticeable exception from this general rule was the ^{231}U nucleus for which we have considered six levels (instead of just the ground state as recommended in RIPL [16]).

The value of the asymptotic level density parameter $\tilde{a}$ of the target nucleus defines the maximum inelastic cross section, the $\tilde{a}$ value of the residual nucleus after a second neutron is emitted defines the maximum $(n,2n)$ cross section, and so on. Additionally, not only the neutron emission cross sections are affected, but also the corresponding fission chances through competition. Adjustments on the asymptotic values $\tilde{a}$ are sometimes needed to compensate deficiencies either in their definition, in other parameters, or to compensate reaction model defects.

TABLE I. Number of discrete levels N_{lev} and corresponding excitation energy E_{lev} used in level density calculations. Values in parentheses correspond to RIPL estimates [16]. $N_{\tilde{a}}$ represents the renormalization factor of the asymptotic level density parameter $\tilde{a}$ used for each nucleus.

Nucleus	N_{lev}	E_{lev}	$N_{\tilde{a}}$	Nucleus	N_{lev}	E_{lev}	$N_{\tilde{a}}$
^{231}U	6 (1)	0.482	1.10	^{232}U	17 (17)	0.967	0.96
^{233}U	20 (37)	0.425	0.97	^{234}U	22 (44)	1.09	0.99
^{235}U	30 (56)	0.474	1.00	^{236}U	21 (55)	1.06	1.00
^{237}U	40 (70)	0.690	1.00	^{238}U	38 (54)	1.27	1.00

2. Level Densities at the Saddle Points

There are no experimental information about the excited states at the large nuclear deformations corresponding to the fission-saddle points. Therefore, the transitional nuclear level densities (NLD) are significantly more uncertain than the level densities of the normal states. They can be obtained indirectly from the fit of the experimental fission cross sections in strong correlation with the parameters of the fission barriers.

The default values of the NLD parameters entering the EGSM formulation for the saddle points have been derived by modifying the corresponding parameters for the normal states together with fission barrier parameters to fit the experimental fission cross sections for a significant number of actinides. Where available, we have adopted the RIPL recommendations [16, 65] (e.g., for shell corrections and pairing energies). Where appropriate (e.g., for the moments of inertia) we replaced the equilibrium deformation with the deformations corresponding to the saddle points. The asymptotic value $\tilde{a}_f$ and the vibrational enhancement factor $K_v(f)$ of the transitional level density have been obtained by applying correction factors deduced from the fit to their correspondents for normal states: $\tilde{a}_f = 1.3\tilde{a}$, $K_v(f) = 4K_v(gs)$. To increase the level densities, especially at low energies, a default shift of 330 keV in the effective excitation energy has been introduced. The order of symmetry of the nuclear shape at saddles was taken

into account by multiplying the level density with a specific enhancement factor as defined in Refs. [66, 67], and considering the barrier symmetry defined in the RIPL database for the empirical fission barriers [16, 65].

Following this procedure, we established recommended values for the parameters involved, but they are not unique, being clearly model dependent. Therefore, we cannot claim that a general prescription for their calculation is provided. As for the density of the normal states, two types of optimization have been performed in this paper for the level density at each saddle-point to describe the measured cross sections: (i) shifts in the effective excitation energy; and (ii) changes of the asymptotic level density parameter $\tilde{a}_f$. The changes are within 7% of the default value described above, but there is not a systematic trend of these changes across the suite of uranium isotopes.

3. Fission

An accurate description of fission cross sections for studied nuclei is important both for nuclear data evaluation and reaction modelling. From the nuclear data (applications) perspective, a good description of the evaluated (based on measurements) fission cross section is a necessary condition for a good prediction of the cross sections for the competing channels. From the reaction modelling point of view it is important to describe accurately and consistently the measured fission cross sections of a chain of isotopes using the same model and keeping unchanged the set of parameters assigned to each isotope in the multi-chance fission calculation.

The fission formalism implemented in EMPIRE and employed for the present calculations is based on an extended version of the optical model for fission. The basic ideas of this model are: (i) the main fission mode is associated to the nuclear vibrational motion; (ii) fission occurs directly by resonant transmission across the multi-humped barrier at excitation energies corresponding to those of the vibrational states in the wells (seen as doorway states) and indirectly by re-emission into the fission channel after the coupling between the fission mode and the internal degrees of freedom in the well(s); (iii) this coupling, interpreted as a damping of the strength of the vibrational states in the well(s), is simulated by adding to the real part of the deformation potential an imaginary term in the region of the well(s) to absorb out flux from the fission mode.

The third shallow hyper-deformed minimum in the potential energy of light actinides predicted more than four decades ago by Möller *et al.* [68–70] was confirmed experimentally (e.g., see Refs. [71–77]). Recent theoretical calculations [78–82] found that light uranium and thorium isotopes (among even- Z nuclei) appear to be the best candidates for triple-humped fission barriers with shallow hyper-deformed minima. Our own reaction modelling of neutron-induced fission cross section on ^{232}Th

and ^{231}Pa [11, 83] and on ^{234}U and ^{235}U [4] also supported a shallow depth of the hyper-deformed minimum.

A comprehensive description of the extended optical model for fission has been recently published [4], therefore the description given here is limited to the elements specific to the neutron-induced fission cross section calculation for uranium isotopes.

Fission barriers. In its journey from the equilibrium deformation towards the scission point the nucleus can follow an infinite number of paths corresponding to different excited states. To each of these states, defined by the angular momentum J , parity π and angular momentum projection on the nuclear symmetry axis K , is associated a fission barrier. The multi-humped barriers employed in the optical model for fission can be either defined numerically or parameterized by parabolas smoothly joined as function of the quadrupole deformation (see Fig. 1). The barrier associated to the state in which the entire excitation energy of the nucleus is bound in deformation, so that the nucleus can be considered in its ground state, is called fundamental. In this paper a fundamental barrier with N_h humps and N_w wells is defined as in Ref. [4],

$$\begin{aligned} V_h(\beta) &= E_{fh} - \frac{1}{2}\mu(\omega_h)^2(\beta - \beta_h)^2, \\ V_w(\beta) &= E_{fw} + \frac{1}{2}\mu(\omega_w)^2(\beta - \beta_w)^2, \end{aligned} \quad (1)$$

where h is the barrier (hump) index that runs from 1 to N_h , and w is the well index that runs from 2 to N_w . Note that the first well, that corresponds to the equilibrium deformation, is not included in the parametrization (as the model assumes the full damping of the class I vibrational states). The energies E_{fh} (E_{fw}) correspond to the maxima (minima) of the potential, β_h (β_w) are the corresponding deformations, the harmonic oscillator frequencies ω_h (ω_w) define the curvature of the parabolas for humps and wells near the maxima. The inertial mass parameter μ is assumed to be independent of the deformation β and is calculated according to Refs. [84, 85] with the semi-empirical expression $\mu = 0.054A^{5/3}\hbar^2 \text{ MeV}^{-1}$, where A is the nuclear mass number and $\hbar$ is expressed in MeV·sec. In this approximation, the only role of μ is to scale the potential energy of deformation [85]. As shown below, the deformation dependence is considered by parameters involved in the definition of the additional barriers (e.g., the moments of inertia).

The same parametrization is used for the barriers corresponding to the discrete excited states of the nucleus. The discrete energy spectrum of the deformed nuclei consists of rotational levels built on vibrational or single-particle band-heads. The excited states with deformations corresponding to the maxima (humps) are often called transition states while the excited states with the deformations corresponding to the minima (wells) are called class I,II,III... states. The excitation energies of these levels characterized by a given set of quantum numbers ($KJ\pi$)

FIG. 1. (Color online) A triple-humped fission barrier and associated parameters.

are

$$E_i(KJ\pi) = V_i + \epsilon_i(K, \pi) + \frac{\hbar^2}{2\mathfrak{S}_i} [J(J+1) - K(K+1)], \quad (2)$$

where i runs over all humps and wells, $\epsilon_i(K, \pi)$ are the rotational band-head energies, and $\hbar^2/2\mathfrak{S}_i$ are the inertial parameters of the same band (the decoupling parameter for $K = 1/2$ bands was neglected).

The energy spectra of the transition states is discrete up to certain energies E_{c_h} above which they become continuous and are described by transitional level density functions. The level densities at the saddle points are discussed in Sec. II B 1.

The damping of the class II/III vibrational states within wells is simulated by introducing negative imaginary potentials, iW_w , in the corresponding deformation ranges, that causes absorption of the incoming flux in these wells [8, 9, 86–89]. The imaginary potential strengths W_w are assumed to be quadratic functions of the deformation, β , like the real part,

$$W_w(\beta) = -\alpha_w(E)[E - V_w(\beta)] \quad w = [2, N_w]. \quad (3)$$

The factors $\alpha_w(E)$ help control the strength of the imaginary parts of the fission potential. These factors are chosen to allow the fit of the resonances in sub-barrier fission cross section and to guarantee a proper asymptotic behaviour of transmission coefficients at higher excitation energies.

All the nuclei studied in this paper are considered to have triple-humped fission barrier, meaning that the number of humps and wells are $N_h = N_w = 3$. The first well ($w=1$) corresponds to the equilibrium deformation and accommodates the normal states, the second well ($w=2$) accommodates the super-deformed (SD) states, also known as class II states, while the third well ($w=3$) accommodates the hyper-deformed states also known as class-III states. The only exception is the ^{238}U nucleus for which a double-humped fission barrier is considered

($N_h = N_w = 2$). According to some theoretical predictions and to our previous calculations, the shape of the triple-humped barrier for light uranium isotopes is similar to the one sketched in Fig. 1: the first hump is lower and wider than the outer humps, the second well is rather deep while the depth of the third well is less than 1 MeV.

Fission transmission coefficients. It was mentioned that fission occurs directly by resonant transmission across the multi-humped barrier at excitation energies corresponding to those of the vibrational states in the wells, and indirectly by re-emission into the fission channel after partial absorption of the incoming flux into the wells. The optical model for fission uses direct $T_d^{(h,h')}$ and absorption coefficients $T_a^{(w,w')}$, which are associated to the direct and indirect transmission mechanisms across the fission barrier, correspondingly. The meaning of their notation is the following: $T_d^{(h,h')}$ describes the direct transmission through the humps h and h' and $T_a^{(w,w')}$ describes the absorption from the well w into the well w' . The general expressions of these coefficients for a multi-humped barrier specific to the optical model for fission have been obtained by an iterative procedure [12] adopting as starting point the transmission coefficients for a double-humped barrier from Ref. [90] derived in the first-order Jeffreys-Wentzel-Kramers-Brillouin (JWKB) approximation. The expressions can be found in Ref. [4], therefore they are not reproduced in detail in this paper. However, we reproduce the basic formulae from Ref. [90] just to remind how the fission barrier parameters enter the fission coefficient definition.

In the JWKB approximation the transmission coefficients are expressed in terms of momentum integrals for the humps and wells of the real barrier, as well as for the imaginary potential(s) in the well(s) [91, 92]. The transmission coefficient across one hump h is defined as

$$T_h = \frac{1}{1 + \exp(2K_h)}, \quad (4)$$

with the momentum integral given by

$$K_h = \pm \int_{a_h}^{b_h} [2\mu(E - V_h(\beta))/\hbar^2]^{1/2} d\beta, \quad (5)$$

where the plus sign is taken when the excitation energy is lower than the hump under consideration, and the minus when it is higher; a_h, b_h stand for the deformations corresponding to the classical turning points of the potential hump. The classical turning points are represented by the red dots in Fig. 1. For a triple-humped barrier the relations between the abscissae of the classical turning points for the humps (a_h, b_h) and for the wells (a_w, b_w) are $a_{h_j} = b_{w_j}, b_{h_{j-1}} = a_{w_j}$ with $j=2,3$.

The direct transmission coefficient across a double-humped fission barrier with an imaginary potential in the second well reads

$$T_d^{(1,2)} = \frac{T_1 T_2}{e^{2\delta_2} + 2A^{1/2} \cos(2\nu_2) + Ae^{-2\delta_2}}, \quad (6)$$

where $A = (1 - T_1)(1 - T_2)$. The momentum integral ν_2 depending on the real part of the potential is approximated as

$$\nu_2 = \int_{a_{w2}}^{b_{w2}} \left[\frac{2\mu}{\hbar^2} (E - V_2(\beta)) \right]^{1/2} d\beta. \quad (7)$$

The momentum integral δ_2 of the imaginary potential is defined as

$$\delta_2 = - \left(\frac{\mu}{2\hbar^2} \right)^{1/2} \int_{a_{w2}}^{b_{w2}} \frac{W_2(\beta)}{[E - V_2(\beta)]^{1/2}} d\beta. \quad (8)$$

The absorption coefficient associated to the shape transition from the first minimum ($w = 1$) to the second minimum ($w = 2$) reads

$$T_a^{(1,2)} = T_d^{(1,2)} \left[\frac{e^{2\delta_2} - [1 - T_2]e^{-2\delta_2} - T_2}{T_2} \right]. \quad (9)$$

For a triple-humped barrier, the expressions of the direct and absorption coefficients are a generalization of the above equations. In this paper the focus is not on the formal definition of these coefficients, but on their physical meaning and on how are they applied for the particular barrier shape described at the end of the previous paragraph. In this case, the total fission coefficient is the sum of the direct and two indirect coefficients,

$$T_f = T_d^{(1,3)} + R[T_i^{(2)} + T_i^{(3)}], \quad (10)$$

where $T_d^{(1,3)}$ represents the direct transmission coefficient through all three humps, $T_i^{(2),(3)}$ stand for the indirect transmission coefficients associated to the second and third well, and R is a normalization factor [12]. The indirect coefficient $T_i^{(2)}$ is the probability of transmission through the outer humps after absorption in the second well,

$$T_i^{(2)} = T_a^{(1,2)} \left[\frac{T_d^{(2,3)}}{\sum T(2)} + \frac{T_a^{(2,3)}}{\sum T(2)} \cdot \frac{T_3}{\sum T(3)} \right], \quad (11)$$

where the first term represents the probability of direct transmission through the second and third hump from the second well while the second term represents the probability of transmission through the third hump after a shape transition from the second to the third well.

The indirect coefficient $T_i^{(3)}$ is the probability of transmission through the outer hump after absorption in the third well,

$$T_i^{(3)} = T_a^{(1,3)} \left[\frac{T_3}{\sum T(3)} + \frac{T_a^{(3,2)}}{\sum T(3)} \cdot \frac{T_d^{(2,3)}}{\sum T(2)} \right], \quad (12)$$

where the first term represents the probability of transmission through the third hump from the third well while the second term represents the probability of direct transmission through the second and the third hump after a

shape transition from the third to the second well. The denominators represent the sum of the transmission coefficients for the competing channels specific to the second and third wells,

$$\sum T(2) = T_1 + T_d^{(2,3)} + T_a^{(2,3)} \quad (13)$$

$$\sum T(3) = T_d^{(2,1)} + T_3 + T_a^{(3,2)}. \quad (14)$$

γ -decay in both wells was neglected. Its contribution has to be considered when the transmission coefficients through the humps are small enough to become comparable with the coefficient for γ -decay, which happens at deep subbarrier excitation energies. This is not the case for neutron-induced fission and triple-humped barriers as those used in this paper. The first hump is usually comparable with the neutron separation energy. Therefore, γ -decay in the second well is negligible compared to the probability of a shape transition from the second to the first well. In other words the transmission coefficient through the first hump at excitation energies close or above its top is huge compared to the coefficient for γ -decay inside the well. The third well being shallow, the excitation energies in this well are close to the top of the second and especially the third hump, so that the shape transition from the third to the second well and mainly the transmission across the third hump toward fission are significantly more probable than γ -decay inside the well.

As demonstrated in Ref. [12], the normalization factor R which takes into account the effect of the infinite number of shape transitions and guarantees the flux conservation has the expression,

$$R = \left[1 - \frac{T_a^{(2,3)}}{\sum T(2)} \frac{T_a^{(3,2)}}{\sum T(3)} \right]^{-1}. \quad (15)$$

For a double-humped barrier the expression of the total fission coefficient becomes

$$\begin{aligned} T_f &= T_d^{(1,2)} + RT_i^{(2)} = \\ &= T_d^{(1,2)} + T_a^{(1,2)} \frac{T_2}{T_1 + T_2}, \quad R = 1. \end{aligned} \quad (16)$$

At excitation energies below the wells there is only direct transmission. This holds true also at low excitation energies in the wells where the class II/III vibrational states preserve their strength (decay probability is small so states are not damped). In the zero-damping limit no absorption occurs, the indirect transmission coefficients tend to zero ($T_i^{(2)} \rightarrow 0$, $T_i^{(3)} \rightarrow 0$) and the fission coefficient becomes equal to the direct transmission coefficient across all the humps,

$$T_f = T_d^{(1,N_h)}, \quad (17)$$

with $N_h = 2$ for the double- and $N_h = 3$ for the triple-humped barrier.

In the full-damping limit, which happens for excitation energies close to the top of the humps and above, the

direct transmission coefficients tend to zero ($T_d^{(1,3)} \rightarrow 0$, $T_d^{(1,2)} \rightarrow 0$, $T_d^{(2,3)} \rightarrow 0$, $T_d^{(3,2)} \rightarrow 0$). Hence, the absorption coefficients between wells separated by two humps tend to zero ($T_a^{(1,3)} \rightarrow 0$, therefore, $T_i^{(3)} \rightarrow 0$) and the absorption coefficients between wells separated by a single hump become equal to the transmission coefficient through that hump ($T_a^{(1,2)} \rightarrow T_1$, $T_a^{(2,3)} \rightarrow T_2$, $T_a^{(3,2)} \rightarrow T_2$). For a double-humped barrier the direct transmission coefficient tends to zero ($T_d^{(1,2)} \rightarrow 0$), and the absorption coefficient from the first to the second well becomes equal to the transmission coefficient through the first hump ($T_a^{(1,2)} \rightarrow T_1$). Making these replacements in Eqs. (10) and (17), one obtains the well known expression,

$$T_f = \left[\sum_{h=1}^{N_h} \frac{1}{T_h} \right]^{-1}. \quad (18)$$

The fission coefficients just presented correspond to transmission through only one barrier associated to a single fission path, while in reality there is an infinite number of fission paths corresponding to different excited states of the nucleus along the deformation process, hence there is an infinite number of barriers.

In the statistical models enter the fission coefficients corresponding to the transmission through all barriers having the same quantum numbers $J\pi$. The calculation of these effective fission coefficients are presented in detail in Ref. [4], therefore we retain here only the essential elements. In principle, all of them are the sum of two contributions corresponding to the discrete and continuous part of the transition state spectrum,

$$\begin{aligned} T_h(EJ\pi) &= T_{h,dis}(EJ\pi) + T_{h,cont}(EJ\pi), \\ T_{dir}^{(h,h')}(EJ\pi) &= T_{dir,dis}^{(h,h')}(EJ\pi) + T_{dir,cont}^{(h,h')}(EJ\pi), \\ T_{abs}^{(w,w')}(EJ\pi) &= T_{abs,dis}^{(w,w')}(EJ\pi) + T_{abs,cont}^{(w,w')}(EJ\pi). \end{aligned} \quad (19)$$

To explain the discrete contributions, we remind that the partial lift of degeneracy associated to the nuclear shape asymmetry is considered by enhancement factors specific to each saddle point which are applied to the transitional state densities (see Sec. IIB 1) and should be applied for consistency to the discrete transition states also. As explained in Ref. [4], for practical reasons, we adopted an approximation in which the asymmetry enhancements $d_{sym}^{(h)}$ are applied on the transmission coefficients and not on the discrete states. This approximation is based on the assumption that the total penetrability through n identical humps is equal to the penetrability through a single hump multiplied by n . Consequently, the transmission coefficient through the single-humped barriers $h = [1, N_h]$ associated to the discrete transition state spectrum reads

$$T_{h,dis}(EJ\pi) = \sum_{K \leq J} d_{sym}^{(h)} T_h(EKJ\pi). \quad (20)$$

Calculation of direct transmission coefficients in the optical model for fission requires a continuous fission path

along the deformation variable. Formally, this means that the number of transition states must be the same above all humps crossed. This does not happen because the asymmetry enhancement factors are different for different humps. To guarantee the continuity of the fission barriers and the possibility of direct transmission, the lowest enhancement factor (denoted d_{sym}^{min}) is applied on the direct transmission coefficients

$$T_{dir,dis}^{(h,h')}(EJ\pi) = \sum_{K \leq J} d_{sym}^{min} T_d^{(h,h')}(EKJ\pi). \quad (21)$$

In the “full K-mixing” approximation the excitation of the internal degrees of freedom in the second well makes it possible for the nucleus to change its K value during the time the energy is bound in internal motions. Formally, “full K-mixing” is described by adding the absorption from different transition states irrespective of the associated K value into a quantity preserving the spin and parity. The main consequence is that the absorption coefficient for a certain $J\pi$ is

$$T_{abs,dis}^{(w,w')}(EJ\pi) = \sum_{K \leq J} d_{sym}^{(h)} T_a^{(w,w')}(EKJ\pi). \quad (22)$$

The transmission coefficient through the single-humped barriers associated to the continuous part of the transition state spectrum reads

$$T_{h,cont}(EJ\pi) = \int_{E_{ch}}^{\infty} \frac{\rho_h(\varepsilon J\pi) d\varepsilon}{1 + \exp \left[-\frac{2\pi}{\omega_h} (E - V_h - \varepsilon) \right]}, \quad (23)$$

where $\rho_h(\varepsilon J\pi)$ is the level density at the saddle h , E_{ch} is the energy where continuum starts and ε represents the excitation energy with respect to the top of the fundamental hump V_h .

The contributions of the direct and indirect transmission across the barriers in continuum are evaluated based on the following physical arguments regarding the particularities of the triple barriers used in this paper. Direct transmission occurs via the vibrational states in the wells which play the role of the doorway states for fission. For a deep well, such as the second well, the class II vibrational states dissipate their strength becoming almost damped for excitation energies close to the top of the humps (and corresponding width of states are broad at those excitation energies). For most of the fissioning nuclei, excepting probably the odd-odd ones, the continuum starts at least at several hundreds of keV above the fundamental humps, meaning that the probability of crossing the barriers in the continuum becomes significant at excitation energies for which the class II vibrational states are completely damped. In this case, the direct transmission coefficient for continuum tends to zero, and the absorption in the second well becomes equal to the transmission across the

FIG. 2. (Color online) Evaluated and calculated total cross section for $n+^{232-237}\text{U}$ compared to experimental data from EXFOR [93–103].

first hump,

$$\begin{aligned}
 T_{dir,cont}^{(1,2)}(EJ\pi) &= 0; \quad T_{dir,cont}^{(3,2)}(EJ\pi) = 0, \\
 T_{abs,cont}^{(1,2)}(EJ\pi) &= T_{1,cont}(EJ\pi), \\
 T_{abs,cont}^{(3,2)}(EJ\pi) &= T_{2,cont}(EJ\pi).
 \end{aligned} \tag{24}$$

For a shallow well, such as the third well, the class III vibrational states owing to their low excitation energy, still preserve some of their strength at the energies where trans-

mission across the humps in continuum become significant. Formally, in a statistical model, where the level densities are $(EJ\pi)$ dependent, while the transitional states are $(EKJ\pi)$ dependent, an unitary treatment for both discrete and continuous channels within the optical model for fission is difficult. Therefore, we are using the surrogate optical model described in Ref. [4] which defines the total fission coefficient as an energy dependent weighted sum of the two asymptotic cases previously discussed: a direct

coefficient corresponding to the zero-damping limit, and an indirect coefficient corresponding to the full damping of the vibrational states in the well,

$$\begin{aligned} T_{dir,cont}^{(2,3)}(EJ\pi) &= (1 - p_3(E))T_{d(0),cont}^{(2,3)}(EJ\pi), \\ T_{abs,cont}^{(2,3)}(EJ\pi) &= p_3(E)T_{a(f),cont}^{(2,3)}(EJ\pi). \end{aligned} \quad (25)$$

The direct coefficient corresponding to the zero-damping limit $T_{d(0),cont}^{2,3}$ is given by

$$T_{d(0),cont}^{(2,3)}(EJ\pi) = \int_{E_c}^{\infty} \overline{T_{d(0)}^{(2,3)}}(E - \varepsilon J\pi) \rho_{min}(\varepsilon J\pi) d\varepsilon,$$

where

$$\overline{T_{d(0)}^{(2,3)}} = \frac{T_2 T_3}{1 + A}, \quad (26)$$

with $A = (1 - T_2)(1 - T_3)$, and $\rho_{min}(\varepsilon J\pi)$ is the minimum of the level density at the outer two saddles supposed to control the direct transmission. The indirect coefficient $T_{a(f),cont}^{(2,3)}(EJ\pi)$ in the full damping limit is defined by Eq. (18) with transmission coefficients given by Eq. (23). The weight $p_3(E)$ rises from 0 for excitation energies close to the bottom of the third well V_3 , to 1 for the excitation energy V_d where full damping is supposed to be reached above the barrier,

$$p_3(E) = \frac{(E^2 - V_3^2)}{(V_d^2 - V_3^2) \exp[-(E - V_d)/b_3]}, \quad (27)$$

with b_3 representing an input parameter which defines the width of the excitation energy region where weight is damped with increasing excitation energy.

III. RESULTS AND DISCUSSION

As emphasized before, the aim of this paper is not to undertake evaluations of neutron-induced reaction on uranium isotopes, but to define models and parameters which provide comprehensive and consistent results that describe available experimental data in all reaction channels up to $E_n = 30$ MeV. Therefore, we first will present for each reaction channel a comparison for all studied uranium isotopes (instead of discussing all reaction channels for each of the studied nuclei). This approach is intended to demonstrate the consistency of the calculations.

The total, elastic, nonelastic, inelastic, $(n,2n)$, $(n,3n)$, radiative capture and fission cross sections are compared with the evaluated data from selected libraries ENDF/B-VII.1 [23], JENDL-4.0 [24], and JEFF-3.2 [26], where independent evaluations exist. For the ^{232}U target, the ENDF/B-VII.1 library adopted the evaluation from JENDL-4.0 [24]. Therefore, we compared to the ENDF/B-VII.0 [22] library instead, so ENDF/B-VII.1 should be read as ENDF/B-VII.0 in the case of this nucleus. The experimental data used for comparison are retrieved from EXFOR [31].

A. Total, Elastic and Nonelastic Cross Sections

For all studied nuclei, the total cross section, the nonelastic cross section, the direct contribution to the elastic and inelastic cross sections, and the particle transmission coefficients used in the statistical models are calculated with a dispersive coupled channel optical model RIPL 2408 [42, 43]. The optical-model coupling scheme is described in Sec. II A. The isovector terms and the weak dependence of the optical-model geometrical parameters on A allowed to extend the derived potential parameters to neighboring actinide nuclei with great confidence [43].

Total cross sections. The optical model potential was already shown in Ref. [43] to provide an excellent description of experimental total cross sections for ^{233}U and ^{235}U targets, as can be seen in Fig. 2. The agreement of EMPIRE calculations with JENDL-4.0 evaluation is very good for all isotopes in the whole energy range (only minor differences are noted for ^{237}U probably owing to differences in the employed coupling scheme). Such agreement is explained as JENDL-4.0 evaluators [24] used a very similar to RIPL 2408 dispersive optical model potential of Ref. [104] derived for a ^{232}Th target, and adjusted the deformation parameters to extend the potential to other actinides. The agreement of calculations with JEFF-3.2 evaluation [26] is also good, with JEFF-3.2 total cross section slightly larger for ^{235}U and ^{237}U targets between 100 keV and 2 MeV, and slightly lower for ^{236}U target between 80 keV and 400 keV.

Overall, calculated total cross sections above 1 MeV are in fair agreement with evaluated cross sections for $^{232-236}\text{U}$ isotopes for all considered libraries, the agreement being remarkable above 5 MeV. There is some disagreement between different evaluations and EMPIRE calculations for ^{237}U target below 5 MeV. Differences seen at lower neutron energies could be related to structure effects and differences in the employed coupling scheme in different evaluations.

Evaluated data in the ENDF/B-VII.1 library [23] for ^{232}U , ^{236}U and ^{237}U look out of trend and do not follow the expected systematic behaviour, therefore it should be revised. There is an obvious discontinuity in the ENDF/B-VII.1 library [23] for ^{234}U at 100 keV that should be corrected. Such discontinuity will be seen in all cross sections calculated for ^{234}U target, and therefore invalidates the comparison with the ENDF/B-VII.1 evaluation [23] in this energy region.

Elastic cross sections. Calculated shape elastic cross sections are obtained directly from the RIPL 2408 optical model potential and are strongly correlated to calculated total cross sections. Similar correlations are expected, and indeed observed, in evaluated data. Calculations are compared with selected evaluations in Fig. 3. The ENDF/B-VII.1 library [23] shows the same deficiencies pointed out for total cross sections for ^{232}U , ^{234}U , and ^{236}U targets. However, the agreement of EMPIRE calculations with evaluations for well studied fissile targets ^{233}U and ^{235}U is very good, except that the ENDF/B-VII.1 library slightly

FIG. 3. (Color online) Evaluated and calculated elastic cross section for $n+^{232-237}\text{U}$ compared to experimental data from EXFOR [98, 105, 106].

underestimates both calculations and other evaluations from 800 keV up to 5 MeV. JEFF-3.2 evaluation agrees very well with calculations for all targets, except for ^{237}U , where a suspicious drop can be seen in Fig. 3 for incident neutron energies below 100 keV. It is worth noting that sometimes, mostly in the past, when the evaluators needed to change the nonelastic cross section in order to fit the experimental data, they used the elastic cross section to compensate for these changes and preserve the

total cross section. Authors feel that such practice should be discontinued owing to the large impact of elastic cross sections and angular distributions on integral quantities.

Nonelastic cross sections. Nonelastic cross sections were calculated by subtracting the total elastic (shape + compound elastic) cross section from the total cross section calculated with the RIPL 2408 optical model potential [42, 43] with the coupling as described in Sec. II A. Calculations are compared with selected evaluations in

FIG. 4. (Color online) Evaluated and calculated nonelastic cross section for $n+^{232-237}\text{U}$ compared to experimental data from EXFOR [107–114].

Fig. 4. For the ^{237}U target discrepancies below 1 MeV reach 2 barns, being the EMPIRE result between the highest JEFF-3.2 evaluation [26] and the lowest ENDF/B-VII.1 library [23]. For a very well studied ^{236}U target EMPIRE calculations are in good agreement with the JEFF-3.2 library [26] below 1 MeV, while the ENDF/B-VII.1 evaluation [23] lies higher from 50 keV up to 3 MeV in a region, which is critical for fast reactors.

The smooth behaviour of EMPIRE nonelastic cross sec-

tions in Fig. 4 is in stark contrast with fluctuations seen in the ENDF/B-VII.1 library [23] for ^{232}U , ^{233}U , ^{234}U , ^{235}U , and ^{236}U . Such fluctuations appear even for very well studied targets like ^{236}U (e.g., see the region from 5 to 10 MeV), and have an effect on undertaken compound nuclear decay calculations. JEFF-3.2 evaluations behave in a physically sound way, and observed differences may be explained by different optical model potentials and coupling schemes (except for the large non-elastic cross sec-

FIG. 5. (Color online) Evaluated and calculated (n,n') cross section for $n+^{232-237}\text{U}$ compared to experimental data from EXFOR [111, 115–117].

tion evaluated in JEFF-3.2 for ^{237}U target below 1 MeV).

B. Inelastic, $(n,2n)$ and $(n,3n)$ Cross Sections

Inelastic cross sections. EMPIRE calculations of inelastic cross sections are compared with selected evaluations in Fig. 5. The consistency of EMPIRE model calculations is reflected in the calculated structure of the

inelastic cross sections of N -even targets near the fission threshold, corresponding to the resonance structure of the fission cross section, especially for ^{232}U and ^{234}U targets. Relatively large differences between calculations and evaluations are observed for ^{232}U , ^{233}U , ^{235}U , ^{236}U and ^{237}U targets, a situation similar to what was discussed in 2011 at an IAEA Technical Meeting on “Inelastic Scattering and Capture Cross-section Data of Major Actinides in the Fast Neutron Region” [118].

FIG. 6. (Color online) Evaluated and calculated $(n,2n)$ cross section for $n + ^{232-237}\text{U}$ compared to experimental data from EXFOR [119–121].

For ^{232}U target the ENDF/B-VII.1 library [23] is significantly lower than EMPIRE calculations up to $E_n \approx 2$ MeV, and the evaluated cross section has an unphysical kink at $E_n \approx 14$ MeV, which is also seen in ^{234}U and ^{237}U targets. The JENDL-4.0 evaluation for ^{232}U target is much higher than calculations from 500 keV to 2 MeV, which can not be explained by different optical models. For ^{233}U target the ENDF/B-VII.1 library [23] is higher than current calculations up to 1 MeV, while the JENDL-4.0 evaluation is in very good agreement with calculations.

For this target the fission cross section is almost twice the inelastic cross section up to the $(n,2n)$ threshold, therefore uncertainties in fission cross section have a large impact on estimated inelastic cross section, which may explain observed discrepancies.

For ^{235}U target the ENDF/B-VII.1 library [23] is higher than current calculations in the maximum, while the JENDL-4.0 evaluation is in very good agreement with calculated inelastic cross section in the whole energy range. The JEFF-3.2 evaluation [26] is higher than current calcu-

FIG. 7. (Color online) Evaluated and calculated $(n,3n)$ cross section for $n+^{232-237}\text{U}$ compared to experimental data from EXFOR [121, 122].

lations below 2 MeV, a behaviour which is correlated with observed higher total and non-elastic cross sections for the JEFF-3.2 evaluation [26] in the same energy region. The fission and inelastic cross sections are comparable for this target nucleus, therefore, inelastic cross section has a dip around 800 keV that corresponds to the large vibrational fission resonance observed at that energy. Such effect in the inelastic cross section can not be described if fission modelling is based on independent fission barriers.

Significant differences can be seen between current calculations and evaluated files for ^{236}U target; evaluations tend to be lower than our calculations. Largest differences can be seen at the maximum, being JENDL-4.0 the lowest cross section; additional differences are seen below 700 keV being ENDF/B-VII.1 the largest cross section and JEFF-3.2 the smallest. For this nucleus, the inelastic cross sections is the dominant decay channel up to the $(n,2n)$ threshold, therefore it is difficult to see the impact

on the inelastic cross sections of sub-barrier vibrational fission resonances.

The largest differences between evaluated inelastic cross sections and current calculations are observed for ^{237}U target, which is probably a result of differences seen in non-elastic cross sections. For this nucleus, similar to ^{236}U target, the inelastic cross sections is by far the dominant decay channel up to the $(n,2n)$ threshold. Therefore, fission cross section calculation are determined by the predictive capability of the inelastic scattering cross sections. The JEFF-3.2 evaluation [26] is the only evaluation in agreement with EMPIRE calculated values below 800 keV, with the ENDF/B-VII.1 evaluation much lower and the JENDL-4.0 evaluation much higher. The shape of the ENDF/B-VII.1 library [23] looks unphysical for $E_n = 7 - 14$ MeV.

Cross sections of $(n,2n)$ and $(n,3n)$ reactions. EMPIRE calculations of $(n,2n)$ and $(n,3n)$ neutron emission cross sections are compared with selected evaluations in Figs. 6 and 7, respectively. Note that the shape of EMPIRE calculations is similar for all calculated uranium isotopes. These reactions channels are the main competition to fission above 7–8 MeV of neutron incident energy, increasing in importance proportionally to the increasing number of neutrons for the same element. Additionally, the preequilibrium emission plays an important role in multiple neutron emission calculations, and the high energy (HE) tail seen in all $(n,2n)$ evaluations and current calculations comes from the contribution of non-compound reactions (either from the direct inelastic scattering to discrete levels or continuum, or from the pre-equilibrium emission). The HE tail in $(n,2n)$ calculations above 15 MeV is higher than in evaluations for practically all targets owing to the inelastic scattering to states in the continuum (as discussed in Sec.II A).

Multiple neutron emission is very small for ^{232}U and ^{233}U targets compared to fission, which increases the uncertainty in calculated cross sections. Very large differences in $^{232}\text{U}(n,2n)$ cross sections are observed between EMPIRE calculations and existing evaluations, being JENDL-4.0 [24] higher, and ENDF/B-VII.1 [23] lower than calculations. A similar disagreement of calculations with JENDL-4.0 [24] and ENDF/B-VII.1 [23] evaluations can be seen for ^{233}U and ^{234}U targets.

The agreement between calculations and JENDL-4.0 evaluation is much better for heavier isotopes from ^{235}U to ^{238}U targets (the latter is not shown in the figure, but it has been calculated in Refs. [28–30]). The ENDF/B-VII.1 evaluation [23] also agrees very well with JENDL-4.0 and EMPIRE calculations for ^{235}U target, JEFF-3.2 [26] being the only discrepant evaluation with a non-physical shape for this important nucleus.

While there are also disagreements between evaluations and current calculations for ^{236}U and ^{237}U targets, the evaluated shape is similar. For these nuclei, multiple neutron emission is a very important channel (for ^{237}U target the $(n,2n)$ reactions is the dominant channel around 10 MeV). Differences in the maximum $(n,2n)$ cross sec-

FIG. 8. (Color online) EMPIRE calculated (n,n') , $(n,2n)$, and $(n,3n)$ cross sections for $n+^{232-238}\text{U}$.

tion can be related to small differences in the nuclear level density of the corresponding residual nucleus in different evaluations. Additional differences may arise as a consequence of previously discussed differences in inelastic cross sections.

Even larger differences are observed between EMPIRE calculations and evaluations for $(n,3n)$ reactions, especially for light isotopes (^{232}U , ^{233}U , ^{234}U), where multiple neutron emission is very small compared to fission and even to the inelastic channel, as previously pointed out. JENDL-4.0 evaluations are generally much higher than

calculations, while the ENDF/B-VII.1 library is lower than calculations for ^{232}U and ^{234}U targets, but is higher for ^{233}U and ^{236}U targets.

Experimental data exist only for the ^{235}U target and are well described by EMPIRE calculations, while evaluations overestimate data near the maximum of the excitation function at 20 MeV. For the ^{237}U target, where the multiple neutron emission is the largest of studied nuclei, we can clearly see differences from the $(n,3n)$ threshold up to 20 MeV between evaluations (JEFF-3.2 and ENDF/B-VII.1) and present calculations. Such differences are directly traceable to the higher HE tail of the calculated $(n,2n)$ cross sections compared to the evaluated values.

The isotopic dependence of EMPIRE calculated (n,n') , $(n,2n)$, and $(n,3n)$ cross sections are shown in Fig. 8. Results of calculations for ^{238}U target [28–30] are included for completeness. In inelastic scattering on even-even targets, there is much more structure in current calculations than in Young *et al.* evaluation (compare top panel in Fig. 8 with Fig. 19 of Ref. [17]). These structures in the inelastic channel are a result of near-barrier vibrational resonances in fission cross sections of fertile targets, that are propagated to inelastic cross sections through competition.

Odd-even target effects are clearly seen in $(n,2n)$ cross sections. Odd targets have lower $(n,2n)$ thresholds and their cross sections typically cross the corresponding even-even curves, except for the ^{237}U target. This systematic trend may indicate an overestimation of the calculated $^{237}\text{U}(n,2n)$ cross section compared to the well measured $^{238}\text{U}(n,2n)$ cross section [28–30], leading to corresponding underestimation of calculated $^{237}\text{U}(n,3n)$ compared to the evaluated $^{238}\text{U}(n,3n)$ cross section [28–30].

The effect of adding neutrons to the lightest ^{232}U core is clearly seen for all neutron emission cross sections as discussed by Young *et al.* [17]; the cross sections increase from light to heavier uranium isotopes; $^{238}\text{U}(n,2n)$ and $^{238}\text{U}(n,3n)$ cross sections are roughly one order of magnitude larger than $^{232}\text{U}(n,2n)$ cross sections as shown in Fig. 8.

C. Radiative Capture Cross Sections

The capture cross sections were calculated using updated in 2015 discrete level schemes retrieved from RIPL [16] up to the cut-off energies listed in Table I, and EGSM level densities as discussed in Sec. II B 1. The E1 gamma-ray strength function from section II B was computed using the MLO model [16], and renormalized to $2\pi\Gamma_\gamma/D_0$ determined from experimentally inferred values of Γ_γ and D_0 [16] for ^{233}U , ^{234}U , ^{235}U , and ^{236}U targets.

To obtain more accurate capture cross sections avoiding the renormalization, it is important to include additional low-lying M1 strength in the radiative strength function, as suggested in Ref. [123]. Such extension will be studied elsewhere. This will be particularly relevant for those cases where no measured radiative strength functions are

available.

The results of our calculations are compared in Fig. 9 with the most recent evaluations and experimental data from EXFOR [31]. The agreement between current calculations and evaluations is very good at energies from 10 keV up to 1 MeV for ^{232}U , ^{233}U , ^{234}U , ^{235}U , and ^{236}U targets, which is not surprising if we consider the chosen $2\pi\Gamma_\gamma/D_0$ renormalization based on available experimental data. However, Γ_γ value is not evaluated in RIPL [16] for the ^{237}U target, which may explain the larger discrepancies observed between different evaluations for the $^{237}\text{U}(n,\gamma)$ reaction. Overall, the agreement of our calculations with JENDL-4 evaluation [24] is very good. Differences in non-compound γ emission may explain the differences between current calculations and ENDF-B/VII.1 evaluations [23] above 5 MeV.

D. Fission Cross Sections

Present calculations intended to describe accurately and consistently the experimental fission cross sections (and to predict the competitive reaction channels) of the suite of light uranium isotopes on an extended energy range by using triple-humped barriers predicted by theoretical studies as Ref. [79].

The starting point for this work were the $^{234}\text{U}(n,f)$ and $^{235}\text{U}(n,f)$ cross section calculations undertaken in Ref. [4] as some of the test cases for the extended optical model for fission. The excellent results obtained with triple-humped fission barriers encouraged us to include them in the ongoing evaluation of neutron-induced reactions on ^{235}U within the CIELO project [3]. This was done by using for $^{235}\text{U}(n,nf)$ cross section, the fission barrier parameters derived from the analysis of $^{234}\text{U}(n,f)$ cross section. The same procedure was applied for $^{234}\text{U}(n,nf)$ and $^{235}\text{U}(n,2nf)$ cross sections which were calculated with the fission barrier parameters obtained from the description of well-measured $^{233}\text{U}(n,f)$ cross section.

Because for $n+^{232}\text{U}$ reaction the only reliable experimental information is a single set of experimental data for fission [141], for the fission barrier parameters of the compound nucleus ^{233}U we relied on those derived from the simultaneous fit of $^{233}\text{U}(n,nf)$, $^{234}\text{U}(n,2nf)$ and $^{235}\text{U}(n,3nf)$ cross sections. The procedure was extended for heavier isotopes by adjusting the parameters used for $^{234}\text{U}(n,f)$ to fit the $^{236}\text{U}(n,f)$ experimental cross section. For higher fission chances, the parameters already derived for the lighter isotopes have been employed.

There are discussions on whether ^{238}U fissioning nucleus has a double- or triple-humped barrier. Measured $^{237}\text{U}(n,f)$ and $^{238}\text{U}(\gamma,f)$ cross sections do not provide a clear indication on this issue, and can be described by both types of barriers [124]. Considering that in Ref. [79] the heaviest uranium isotope supposed to have a triple-humped barrier is ^{237}U , and that the empirical double-humped fission barrier parameters of Maslov in RIPL-3 [16] fit very well both $^{237}\text{U}(n,f)$ and $^{238}\text{U}(\gamma,f)$ cross sec-

FIG. 9. (Color online) Evaluated and calculated (n,γ) cross section for $n+^{232-237}\text{U}$ compared to experimental data from EXFOR [125–140].

tions, we used double-humped barrier with Maslov parameters. An additional argument is the fact that calculated triple-hump barriers in both thorium and uranium isotopes become shallower with the increase of mass number along the isotopic chain, eventually disappearing. Such transition is expected to happen at ^{237}U target.

The results obtained based on this consistent methodology for even- N and odd- N targets are presented in Figs. 10 and 11 compared to the most recent evaluations

and experimental data from EXFOR [31].

The fission parameters for the three humps and two wells of the triple-humped fission barrier of $^{233,234,235,236,237}\text{U}$ used in our calculations are given in Table II, together with the neutron separation energies. The parameters for $^{230,231,232}\text{U}$ are less reliable and have not been included in this systematics. The parameters of the double-humped fission barrier of ^{238}U are provided in the text. More details about the fission barrier parameters

FIG. 10. (Color online) Evaluated and calculated fission cross section for even-even uranium isotopes $n+^{232,234,236}\text{U}$ compared with experimental data from EXFOR [141–165]. Left column for $E_n < 5$ MeV in log-log scale; right column for $E_n > 5$ MeV in log-linear scale.

will be presented in next paragraphs, followed by some comments on results of fission cross section calculations.

The fission barrier parameters have been obtained from the analysis of the fission experimental data, using the theoretical predictions and the empirical systematics as guidance, by performing sensitivity studies, and making correlations with the parameters which define the competing channels, as described below. Because studied fissioning nuclei have different nuclear structure with well-known odd-even effects (leading to systematic differences in neutron separation energies and discrete excited states), the

even-odd and even-even fissioning uranium isotopes are treated separately.

Fission barrier parameters for even-odd nuclei. The neutron separation energy of odd- N nuclei is usually lower than the fundamental fission barrier, therefore the cross sections of the neutron-induced fission on even- N targets have a threshold. Fig. 10 shows that this is true only for $^{234}\text{U}(n,f)$ and $^{236}\text{U}(n,f)$ cross sections. $^{232}\text{U}(n,f)$ is a special case, because its cross section is large starting from thermal incident energies and also has a sort of threshold around 1 MeV. The large fission cross section at low

TABLE II. Derived fission barrier parameters in MeV: barrier heights V_{h_1} , V_{h_2} , V_{h_3} and corresponding widths ω_{h_1} , ω_{h_2} ($\omega_{h_3} = \omega_{h_2}$), the excitation energy of the bottom of the wells V_{w_2} and V_{w_3} (the well depth ΔV_2 and ΔV_3 – in parentheses), and neutron binding energy for fissioning triple-humped nuclei (CN). Note that the width of potential wells $\omega_{w_2} = \omega_{w_3}$ was fixed to 1 MeV. Estimated parameter uncertainties are: 10–15% for V_{h_1} and ω_{h_1} ; 5% for V_{h_2} , V_{h_3} , and V_{w_3} ; and 10% for widths ω_{h_2} and ω_{h_3} . V_{w_2} and ω_{w_2} are poorly constrained by neutron-induced fission experimental data, so the uncertainty could be larger than 30%.

CN	V_{h_1}	ω_{h_1}	V_{h_2}	ω_{h_2}	V_{h_3}	V_{w_2} (ΔV_2)	V_{w_3} (ΔV_3)	S_n
^{233}U	4.70	0.7	6.06	1.40	5.70	1.70 (3.00)	5.05 (0.65)	5.77
^{234}U	4.60	0.6	5.90	1.30	5.20	1.60 (3.00)	4.70 (0.50)	6.84
^{235}U	4.80	0.6	6.10	1.45	5.78	1.60 (3.20)	5.10 (0.68)	5.29
^{236}U	4.60	0.6	5.90	1.45	5.64	1.60 (3.00)	4.90 (0.74)	6.54
^{237}U	5.25	0.5	6.18	1.50	5.80	2.30 (2.95)	5.57 (0.23)	5.13

energies is not related to the relation between the neutron separation energy and the height of the barrier, but to the optical potential which predicts for $^{232}\text{U}(n,\text{non})$ cross section at 0.01 MeV a value 2.7-3 times higher than for $^{234,236}\text{U}(n,\text{non})$, as can be seen in Fig. 4. At this incident energy the only competing channel which contributes to the nonelastic cross section is the radiative capture. From Fig. 9 one can see that $^{234,236}\text{U}(n,\gamma)$ and $^{234,236}\text{U}(n,\text{non})$ cross sections are very similar, while $^{232}\text{U}(n,\gamma)$ cross section is significantly smaller than the corresponding nonelastic cross section, therefore, the large difference can only go into the fission channel.

The analysis of the experimental fission cross sections below the threshold shows no visible change of slope or broad (damped) resonances, meaning that the height of the first hump is lower than the neutron separation energies and that the class II vibrational states are completely damped. The calculations confirm that the fission cross section is completely insensitive to reasonable changes in the depth of the second well, in the energies of the class II states, or in the α_2 parameter from Eq. (3) which controls the strength of the imaginary potential in this well. Therefore none of these parameters can be predicted in neutron-induced reactions. The calculations also show a small sensitivity to the parameters of the first hump and to the spectrum of the transition states above it. A lack of sensitivity to the first hump and second well of the fundamental barrier, to the transition states at the first hump, and to the class II states is more evident in the case of the odd- N targets $^{233,235}\text{U}$. In these conditions, we estimated uncertainties of around 10–15% for V_{h_1} and ω_{h_1} parameters and a very large uncertainty that can not be estimated for V_{w_2} and ω_{w_2} parameters.

For the nuclei with odd- N and double-humped barriers the excitation energy corresponding to the fission threshold is very close to the height of the highest hump, so that very valuable information on this parameter can be extracted from the experimental data. For example, the $^{238}\text{U}(n,\text{f})$ cross section has the threshold around 1.6 MeV

incident energy and the neutron separation energy for ^{239}U is 4.8 MeV, so that the height of the highest hump should be around 6.4 MeV, which coincides with the value used in calculations to describe the experimental data [4]. In the case of a triple-humped barrier, as can be seen in Fig. 12, the experimental fission threshold is not so well defined, because the region which links the rising part and the plateau of the fission cross section (which will be considered the fission threshold) is in fact a sequence of resonances which are not determined exclusively by the height of the highest hump.

In this work, as suggested in Ref. [79], the highest hump of the fundamental triple-barrier is the second hump as listed in Table II. Comparing the $^{234}\text{U}(n,\text{f})$ and $^{236}\text{U}(n,\text{f})$ cross sections, one can notice similar structures around the threshold, but shifted by 400–500 keV incident neutron energy (equivalent to 250–350 KeV of excitation energy). For a double-humped barrier this difference would be very likely reflected in the difference of the highest hump heights. For a triple-humped barrier this difference is reflected to a smaller extent in the second hump heights, but mainly in a difference in the depth of the third wells (see Table II). The highest hump height has a largest impact on the calculated fission cross section in the plateau region, therefore its value is constrained by the fit of the experimental data above the threshold. This constraint also applies indirectly to the third well's depth, owing to the correlation of these two parameters needed to describe the experimental threshold. This is a serious argument for a shallow third well. A deeper well would shift the fission threshold towards lower energies; the higher second hump needed to restore the threshold would produce an underestimation of the experimental fission cross section in the plateau region, which is difficult to be compensated by increased transitional level densities (at saddles).

The height of the third hump has an impact on the absolute value of the fission cross section at all energies, but especially below the rising part and in the second half of the plateau region. The widths of the outer humps are influencing mainly the slope below the threshold. All these parameters are derived from the fit of the experimental data.

The parameters of the fundamental barrier are very important because: (i) the fission probability through the fundamental barrier is high and represents a significant (if not the most significant) contribution to the fission cross section below the threshold and several hundreds of keV above it; and (ii) these parameters are used together with the level density functions at saddles to calculate the contribution of the fission channels in continuum, which becomes dominant around 1 MeV above the threshold. However, a well defined fundamental barrier is not enough for an accurate description of the experimental fission cross section. Equally important are the parameters of the higher barriers above the fundamental one, which correspond to the discrete excited states of the nucleus. These are the excitation energies $\varepsilon_i(K, \pi)$ (see Eq. (2)) at the deformations corresponding to the humps and wells

FIG. 11. (Color online) Evaluated and calculated fission cross section for odd- A uranium isotopes $n + ^{233,235,237}\text{U}$ compared with experimental data from EXFOR [129, 131, 140, 142, 143, 145, 147, 153, 154, 166–193]. Left column for $E_n < 5$ MeV in log-log scale; right column for $E_n > 5$ MeV in log-linear scale.

of the rotational band heads with given spin projection K and parity π , the associated widths, and the deformation-dependent moments of inertia needed to construct the rotational bands.

For the even-odd nuclei there is not much information about the discrete spectrum at high deformations to rely on. Therefore, the transition states and the parameters of the associated barriers have been selected to fit the gross vibrational resonance structure of the experimental fission cross section usually visible above the threshold. The

position of the resonances are mainly determined by the energies of the class III vibrational states and the widths of the hyper-deformed wells, while the magnitude of the resonances are mainly determined by the energies of the transition states at the outer two humps, the strengths of the imaginary potential, and also by their spin projection and parity. The resonant transmission occurs for excitation energies lower than the barrier, therefore the resonances with high energies imply transmission through even higher barriers. On the other side, the fission coef-

FIG. 12. (Color online) Calculated fission cross sections near the fission barrier for even-even uranium isotopes with a triple-humped barrier. Authors of measurements omitted for clarity, all are listed and referenced in Fig. 10.

ficients get lower when increasing the barriers and this makes the description of the resonances at higher energies difficult. The $^{234}\text{U}(n,f)$ cross section for example, has a sequence of resonances up to around 1.5 MeV incident energy (around 6.8 MeV excitation energy) followed by a broad peak around 1.8 MeV incident energy. To fit the experimental data above 1.5 MeV incident energy we used 30 rotational band heads. The same band heads with minor adjustments have been used for $^{232}\text{U}(n,f)$ and $^{236}\text{U}(n,f)$ cross section calculations with very good results. Despite the good results, using such large number of discrete transitional states to fit the data is artificial. An alternative interpretation may assume that an observed dip around 2.4 MeV incident energy is due to the competition of inelastic scattering. Such a hypothesis is supported by the lack of structure in measured $^{235}\text{U}(\gamma,f)$ cross section at the corresponding energy [194]. Further studies are needed to elucidate this problem, as other well-known experimental fission cross sections (e.g., $^{238}\text{U}(n,f)$) show a dip in the middle of the fission plateau.

Fission barrier parameters for even-even nuclei. The even-even fissioning nuclei have neutron separation energies higher than the fundamental barrier, but lower than some of the barriers associated to their collective excited states which fill the pairing gap. These features explain the lack of threshold and of resonant structure of the experimental neutron-induced fission cross sections for fissile (odd- A) targets. The almost smooth behaviour of these fission cross sections has two consequences: (i) no practical information on the fission barrier can be extracted from the analysis of the experimental data; and (ii) double-humped barriers and the full-damping limit can be used (and had been successfully used) to describe the neutron-induced fission cross sections for fissile targets. Because our aim is to incorporate the results of the theoretical fission-path studies into the nuclear data evaluation practice, and because these studies show that ^{234}U and ^{236}U are good candidates for triple-humped barriers, we used for the outer humps and the third well of these nuclei the theoretical fission parameters suggested

FIG. 13. (Color online) Partial contributions to the calculated fission cross section $^{235}\text{U}(n,f)$ of the rotational bands built on 0^+ , 2^+ , 0^- and 1^- transitional band-heads, and of the fission channels in continuum.

in Ref. [79] adjusted and completed to fit the data. For the reasons already mentioned, in the case of ^{237}U we adopted a double-humped fission barrier defined by the empirical parameters from RIPL-3 [16]: $V_{h1}=6.30$ MeV, $\omega_{h1}=1.00$ MeV, $V_{h2}=5.50$ MeV, $\omega_{h2}=0.60$ MeV, to which we added for completeness (but without consequences) the parameters of the isomeric well $V_{w2}=1.60$ MeV, $\omega_{w2}=1.00$ MeV.

The discrete spectrum in the pairing gap of the even-even nuclei used in our calculations consists of four rotational bands built on vibrational states with $K^\pi = 0^+, 2^+, 0^-, 1^-$ with excitation energies at the humps in agreement with the corresponding nuclear shape asymmetries, and with excitation energies in the third well chosen to describe the experimental data. Up to around 1 MeV incident energy, the compound nuclei ^{234}U and ^{236}U are populated at excitation energies for which the damping of the class III vibrational states is high, but not complete (because of the relative large distance between class III states specific to the even-even nuclei). Consequently, in this energy range it is expected, beside the dominant indirect transmission, a small contribution of direct transmission for the fission channels in the discrete and the lower part of continuum spectrum which need to be treated within the extended optical model for fission [4]. The high but not complete degree of damping of the class III states is reflected in a decrease of the calculated fission cross section, but not in a resonance structure (as it was demonstrated in Ref. [4]).

Partial contributions of the rotational bands and of the fission channels in continuum to the calculated $^{235}\text{U}(n,f)$ cross section are shown in Fig. 13. Those contributions are determined not only by the associated barriers, but by the spin and parity distribution of the compound nucleus formation cross section. For ^{235}U target with ground state spin $7/2^-$, the negative parity states are favoured at low incident neutron energies. This explains the dominant contribution to the calculated fission cross section below 40 keV of negative parity rotational bands observed in Fig. 13. For both even-odd and even-even nuclei, the contribution of the fission channels in continuum has been cal-

culated with the parameters of the humps listed in Table II, and for the shifts in the effective excitation energy and the renormalization factor of the ratio $\tilde{a}_f/\tilde{a}$ mentioned in Sec. II B 2 adjusted in the limit of 7%.

EMPIRE calculated fission cross sections. Our calculations describe well the experimental data on the entire energy range and agree to a great extent with evaluations. $^{232}\text{U}(n,f)$ cross section is in good agreement with the experimental data and with evaluations up to around 7.4 MeV. Above this energy, the experimental constraint disappears and one can notice a discrepancy between our calculation and JENDL-4.0 on one side, and ENDF/B-VII.0 on the other side. This seems to be related to the observed discontinuity of the ENDF/B-VII.0 inelastic cross section which appears around the same energy (see top left panel of Fig. 4). As can be seen in Fig. 15, $(n,2n)$ and $(n,3n)$ cross sections are practically negligible, the only competition remains the inelastic scattering which at these high energies occurs mainly by direct interaction and preequilibrium emission. The fission cross section results as the difference between the nonelastic and the inelastic cross sections, and does not depend too much on the fission parameters of $^{230,231,232}\text{U}$ which have significant uncertainties.

The current calculations describe nicely the resonance structure of the experimental $^{234}\text{U}(n,f)$ cross section and are very close to the evaluations, especially to ENDF/B-VII.0 in the first fission chance region. The resonance around 300 keV was confirmed by the latest n_TOF data and it is described as a class III vibrational resonance. Above 12 MeV there are some discrepancies between EMPIRE calculations and the evaluations which might be explained by the decision to follow different sets of experimental data, by the high inelastic cross section in ENDF/B-VII.0, and mainly by the high uncertainties of the fission parameters of the lightest $^{230,231,232}\text{U}$ isotopes.

The $^{236}\text{U}(n,f)$ calculated cross section agrees with ENDF/B-VII.0 and JENDL-4.0 in the first fission-chance region and with latest experimental datasets, while JEFF-3.2 shows a different resonance structure. Above 6 MeV all data are very similar; but it is worth to mention that the n_TOF data became available after the release of all three latest evaluations.

The present $^{233}\text{U}(n,f)$ cross section agrees perfectly with ENDF/B-VII.0 and JENDL-4.0 in the first fission chance and slightly overestimates evaluations at higher energies where it follows Lisowski *et al.* [142] and newer experimental data by Tovesson *et al.* [143].

The calculated $^{235}\text{U}(n,f)$ cross section is compared only with the ENDF/B-VII.1 evaluation which coincides with IAEA 2006 Neutron Standard cross section [13, 14] above 150 keV. We emphasize the agreement within 3% between EMPIRE model calculation and a non-model fit of the experimental data in Standard evaluation.

In the case of $^{237}\text{U}(n,f)$ cross section, there are significant discrepancies between the EMPIRE calculation and evaluations. Above 5 MeV calculated EMPIRE cross section is very similar to that from JENDL-4.0.

Calculated partial fission cross section for each contributing fission chance (solid lines) are shown in Fig. 14 and compared with those from the ENDF/B-VII.1 library (dashed lines). The differences between calculated and evaluated partial cross sections are significantly larger than for the total fission cross section. This result was expected, considering the differences in the inelastic and (n,xn) cross sections, and of course, in the models and parameters used to treat the fission channel. For $^{233-236}\text{U}$ we noticed the large contribution of the calculated second chance fission at high energies. The main competing channel of (n,nf) is $(n,2n)$ which also has large values at those energies. The consistency of the calculations would suggest a too high nonelastic cross section at high energies that results in both fission and neutron emission channels to be overestimated.

As mentioned at the beginning of this Section, the intention was to describe accurately and consistently the experimental fission cross sections of light uranium isotopes by using triple-humped barriers. Figs. 10 and 11 demonstrate that we described the experimental data to a great extent. The fact that the fission parameters for each isotope remained unchanged in the limit of 3% irrespective of the fission chance that isotope was responsible for (e.g., the fission barrier parameters of ^{234}U derived from the fit of the first fission chance $^{233}\text{U}(n,f')$ have been changed within these limits to describe $^{234}\text{U}(n,nf)$, $^{235}\text{U}(n,2nf)$, $^{236}\text{U}(n,3nf)$ cross sections) is an argument for the consistency of the calculations. Usually these parameters are changed to compensate the reaction models' and other parameters' deficiencies. The relatively small changes needed might be a confirmation that these deficiencies are minor.

It was considered interesting to compare the isotopic trend of fitted barrier parameters from Table II with the findings of other authors. Despite the fact that several papers have studied this subject, no adequate information for a proper comparison is available. For example, the recent paper by Romain *et al.* [21] presents a fission model that uses triple-humped barriers but does not tabulate the barrier parameters. A theoretical study by Zhou (see Table I in Ref. [82]) recommends for $^{232,234,238}\text{U}$ isotopes barriers higher than 7 MeV, which would strongly underestimate the experimental fission cross sections. The Hartree-Fock-Bogoliubov calculation of the RIPL-3 fission barriers [195] did not predict triple-humped barriers for light Uranium isotopes. To check the trend and not the absolute values, in Fig. 16 we compare the derived maximum of the fission barrier with the corresponding maximum height from Maslov empirical barriers [16]. Maslov barriers describe the experimental neutron induced fission cross sections at least above 1 MeV of neutron incident energy. One can notice a similar trend with odd-even effects in both sets. The absolute differences are explained by the different structure (shape) of the barriers, but we see a reduction of the differences with increasing mass number A , that may be linked to the reduction (and eventual disappearance) of the depth of the third well around $A = 238$.

FIG. 14. (Color online) Cross sections for total fission and partial fission chances ($E_n > 5$ MeV) calculated (solid line) and retrieved from ENDF/B-VII.1 (dashed line); for ^{237}U there is no information on the fission chances in ENDF/B-VII.1.

To conclude, we found a combination of models and parameters that is applicable for all studied nuclei, and not that the absolute values of the parameters are the most appropriate. They still need to be validated in other types of reactions (e.g., in photon-induced reactions) to test the choice of the discrete transition states, of the class II and III states, and of the level density functions for equilibrium deformation and for the saddle points. Further studies are also needed to derive more accurately the widths of humps and wells, the corresponding deformations, shell

corrections and moments of inertia.

The use of the extended optical model for fission is shown to be critical for a proper description of the measured fission cross sections for $^{232-236}\text{U}(n,f)$ reactions in which the fissioning nuclei are considered to have triple-humped barriers.

FIG. 15. (Color online) EMPIRE-3.2 calculations for major reaction channels in neutron-induced reactions on $^{232-238}\text{U}$ targets.

FIG. 16. (Color online) Isotopic trend of the fitted maximum barrier height (see Table II) compared to the maximum of Maslov double-humped empirical barriers from RIPL-3 [16]. Derived barriers for $^{232-237}\text{U}$ are triple-humped, and for $^{238,239}\text{U}$ are double-humped. ^{239}U was added for completeness.

E. Overview of Calculated Cross Sections for $n+^{232-237}\text{U}$ Reactions

Calculated cross sections for major reaction channels on $^{232-238}\text{U}$ targets, including ^{238}U target results from Refs. [28–30], are shown in Fig. 15. Clear trends can be noticed,

- Fission cross sections decrease with increasing mass number A as the highest fission barrier increases; even-even fissioning isotopes have the highest (second) barrier lower than odd- A ones as can be seen in Table II (odd-even effect).
- All even-even targets but ^{232}U show a well defined fission threshold located around 0.5 to 1.5 MeV of incident neutron energy. Lack of threshold for the $^{232}\text{U}(n,f)$ reaction was discussed in the previous section. $^{232}\text{U}(n,f)$ cross section still increases by 50% near 1 MeV.
- All odd- A uranium targets show a fissile behaviour, but $^{237}\text{U}(n,f)$ cross section is relatively flat from 10 keV up to 5-MeV owing to the strong competing inelastic scattering. $^{233}\text{U}(n,f)$ and $^{235}\text{U}(n,f)$ cross sections show a typical fissile behaviour being much higher at 10 keV than at 1 MeV.
- Capture cross sections show a weak dependence on mass number A of the target and a very similar shape. Capture cross sections at 10 keV range from 0.6 barn for ^{233}U target to 1.1 barn for ^{235}U and ^{237}U targets. Then, capture cross sections decrease almost monotonically above 10 keV becoming smaller than competing channels at 200 keV, and negligible – around 1 MeV. Such behaviour means that differential capture data above 200 keV are expected to be a poor constraint for capture evaluations. However, criticality benchmarks are highly sensitive to capture, so we may consider using benchmarks to reduce the evaluated capture uncertainty.
- Inelastic cross sections increase with increasing mass number A for neutron richer isotopes owing to a larger number of available neutrons; the (n,n') scattering cross sections are dominant from 1 to 5 MeV of neutron incident energy for all isotopes heavier than ^{233}U target, and becomes larger than all other channels by a factor of three for ^{236}U to ^{238}U targets at those energies.
- Multiple neutron emission cross sections also increase with increasing mass number A for neutron richer isotopes. The $(n,2n)$ cross section only becomes higher than fission for the heaviest isotopes – ^{237}U and ^{238}U targets. The $(n,3n)$ cross sections reach half of the fission values only for the heaviest ^{238}U target. Therefore, we may conclude that $(n,3n)$ cross sections are not very important for reactor applications.

IV. CONCLUSIONS

Theoretical calculations of cross sections for neutron-induced reactions on $^{232-237}\text{U}$ targets were undertaken in the 10 keV–30 MeV incident energy range with the code EMPIRE-3.2 Malta. These calculations were complemented by those undertaken for ^{238}U target [28–30], which form the basis of a newly proposed IAEA evaluation within the CIELO project [1, 3].

A dispersive optical model potential RIPL 2408 [42, 43] with extended couplings (up to 9 levels for odd- A targets) provided a consistent and comprehensive basis for reaction modelling in the whole energy range of interest by predicting the total, elastic and reaction cross sections for neutron-induced reactions on all uranium isotopes.

A triple-humped fission barrier with absorption in the wells was used for $^{232-237}\text{U}$ fissioning nuclei, while a double-humped fission barrier was used for the ^{238}U fissioning nucleus as predicted by theoretical barrier calculations [78–82]. The extended optical model for fission of Ref. [4] allowed to describe measured fission cross section within 5%, with small tuning of asymptotic level density parameters and fission barrier parameters uniquely assigned to each isotope. Good description of the cross sections confirms the advantage of using the latest nuclear structure, and of following the trend obtained in theoretical fission studies. Model parameters can be further improved; this issue will be addressed in the forthcoming international effort RIPL-4 coordinated by the IAEA.

By studying a chain of uranium isotopes, which span fertile and fissile nuclei, we have shown that current modelling reproduces the measured cross sections for all reaction channels, including the most difficult fission channel. Such good agreement inspires confidence that the quantities for which there is not experimental information are also accurately predicted, and indicates that the used model grasps the most important features of studied nuclear reactions.

The advanced modelling and consistent calculation scheme also allowed to predict a systematic behaviour of neutron scattering and neutron emission cross sections, and to assess the consistency of available evaluated libraries for those reactions. Several data deficiencies were identified in existing libraries, in particular for ^{232}U , ^{234}U and ^{237}U targets.

All these aspects are of great interest for ongoing international projects on nuclear data evaluation.

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