

Monte Carlo simulation of γ and fission transfer-induced probabilities using extended \mathcal{R} -matrix theory: Application to the $^{237}\text{U}^*$ system

Olivier Bouland^{1,a} and Beatriz Jurado²

¹ CEA, DEN, DER, SPRC, Physics Studies Laboratory, Cadarache, 13108 Saint-Paul-lez-Durance, France

² CENBG, CNRS/IN2P3-Université de Bordeaux, Chemin du Solarium, BP. 120, 33175 Gradignan, France

Abstract. This paper deals with simultaneous neutron-induced average partial cross sections and surrogate-like probability simulations over several excitation and de-excitation channels of the compound nucleus. Present calculations, based on one-dimensional fission barrier extended \mathcal{R} -matrix theory using Monte Carlo samplings of both first and second well resonance parameters, avoid the surrogate-reaction method historically taken for surrogate data analyses that proved to be very poor in terms of extrapolated neutron-induced capture cross sections. Present theoretical approach is portrayed and subsequent results can be compared for the first time with experimental γ -decay probabilities; thanks to brand new simultaneous $^{238}\text{U}({}^3\text{He}, {}^4\text{He}\gamma)$ and $^{238}\text{U}({}^3\text{He}, {}^4\text{He}f)$ surrogate measurements. Future integration of our strategy in standard neutron cross section data evaluation remains tied to the developments made in terms of direct reaction population probability calculations.

1. Context and objectives

The idea to supplement neutron-induced cross-section data for actinides and higher transuranic nuclides with particle-transfer-induced reactions has been raised a long time ago [1]. Over the years, a variety of surrogate vectors have been used as stripping (d,p) and pickup (p,d) reactions, (${}^3\text{He},p$), (${}^3\text{He},d$) and (${}^3\text{He},t$) charge-exchange reactions or even two-neutron transfer reactions as (t,p) and (p,t) reactions. Analytical calculations of these measured direct-reaction fission probabilities were performed under several simplifications contained in the so-called surrogate-reaction method [2]. Early promising comparisons [1] made between extrapolated neutron fission cross sections and cross sections directly measured by neutron spectroscopy led to agreement within 10% to 20% at neutron energy above the nucleus pairing energy although exhibiting larger deviations at lower energies. Major limitations in surrogate extrapolation were promptly noticed [1,3] with the difficulty to estimate the compound nucleus formation cross section by neutron absorption, the possible influence of angular momentum differences between neutron capture and direct reactions and the validity of the Weisskopf-Ewing (WE) hypothesis related to fission decay probability spin-parity independence [4].

A decade ago, surrogate reactions received renewed interest in terms either of simulation [5,6] or experimental investigation (initiated by study [7]; reviewed by Ref. [8]) to infer in addition to fission, neutron-induced capture cross sections. Newly measured decay capture probabilities were readily analyzed within the surrogate-reaction method that proved to be very poor in terms of extrapolated neutron-induced capture cross sections [2].

Indeed the surrogate-reaction method was understandable in the seventies because of computer limitations, lack of precise information on nuclear level densities across the deformation and difficulties for achieving confident optical model calculations over a large range of nuclides. Nowadays the bulk of those approximations can be avoided even if tremendous difficulties remain in the modeling of direct reactions. Keep using the surrogate-reaction method might suggest that surrogate data are inappropriate to help although surrogate spectroscopy can be of great help for target material with unsuitable lifetimes (less than several days) or with high radio-toxicity.

In a previous paper [9], we have enlightened the actual possibility to carry one-dimensional fission barrier extended \mathcal{R} -matrix simulations accurate enough to make predictions of low-energy neutron-induced fission cross sections for the isotopes of the Pu family for which no neutron spectroscopy measurements exist. This has been accomplished thanks to Monte Carlo samplings of both first and second well resonance parameters (reaction widths and energies) of the actinide double-humped fission barrier and to model input parameters in part obtained from macroscopic-microscopic nuclear structure calculations [10]. We enlighten that we were able to achieve confident prediction by inclusion of fission probability data induced by (d,p), (t,p), (${}^3\text{He},t$) or even (${}^3\text{He},d$) direct reactions. The 'surrogate' part of the study is being documented [11]. Obviously across the latter paper we do not cope with probabilities measured according to γ -decay because of lack of experimental data with respect to the Pu isotope family and this is precisely the most challenging part of the work as underlined in Ref. [2]. Brand new surrogate data [12] dealing simultaneously with γ and fission decay probabilities

^a e-mail: olivier.bouland@cea.fr

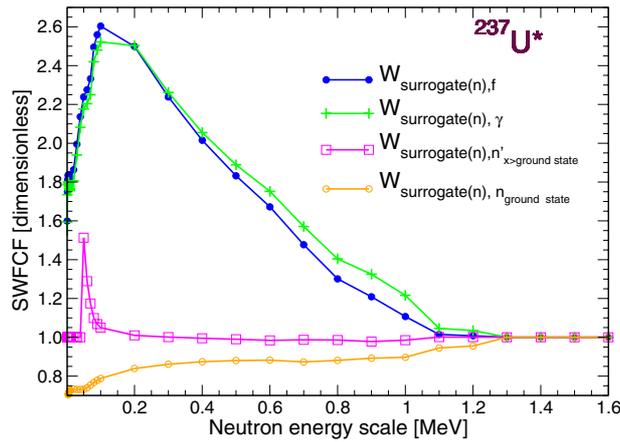


Figure 1. Comparison of SWFCF curves associated to fission (f), γ decay and neutron emission with residual nucleus in ground state (n) or in excited states (n') according to surrogate reactions for the $^{237}\text{U}^*$. Note: for better comparison with WFCF as involved in neutron cross sections [14], present transfer calculations are fed with the entrance population obtained from the corresponding neutron-incident reaction.

related the $^{238}\text{U}(^3\text{He},^4\text{He})^{237}\text{U}^*$ supply the opportunity to discuss the validity of present Monte Carlo \mathcal{R} -matrix technique for more confident inference of neutron-induced capture cross sections below the energy range of second chance fission.

2. Hypotheses carried by the surrogate- reaction method

The starting point and appropriate formalism for describing compound nucleus (CN) reactions is Hauser-Feshbach (HF) statistical theory [13] with the familiar (energy-dependent) in-out-going channel width fluctuation correction factor (WFCF), $W_{c,c'}$. Applied to neutron-induced reactions at neutron energy E_n , this can be written [2] concisely for the average partial cross section,

$$\sigma_{n,c'}(E_n) = \sigma_n^{CN}(E_n) \sum_{J\pi} \left[\mathcal{F}_n^{CN}(E_n, J, \pi) \times \mathcal{P}_{c'}^{J\pi}(E_{c'}) \times W_{n,c'}^{J\pi} \right] \quad (1)$$

with $\mathcal{F}_n^{CN}(E_n, J, \pi) = \sigma_n^{CN}(E_n, J, \pi) / \sigma_n^{CN}(E_n)$; the fraction of CN excited states formed with (J, π) spin/parity. $\mathcal{P}_{c'}^{J\pi}(E_{c'})$ is the individual decay probability into channel c' from given (J, π) state. Similarly the surrogate probability becomes,

$$\mathfrak{P}_{\text{sur},c'}(E_x) = \sum_{J\pi} \left[\mathcal{F}_{\text{sur}}^{CN}(E_x, J, \pi) \times \mathcal{P}_{c'}^{J\pi}(E_{c'}) \times W_{\text{sur},c'}^{J\pi} \right] \quad (2)$$

The historical surrogate-reaction method is based on:

[Hyp. A)] the absence of WFCF in the cross section formulation. We know however that it plays in low energy neutron-induced reactions ($E_n < 2\text{ MeV}$) a major role in averaging over partial width distributions to calculate average cross sections. This was well illustrated in the neutron-induced WFCF model comparisons by Hilaire et al. [14],

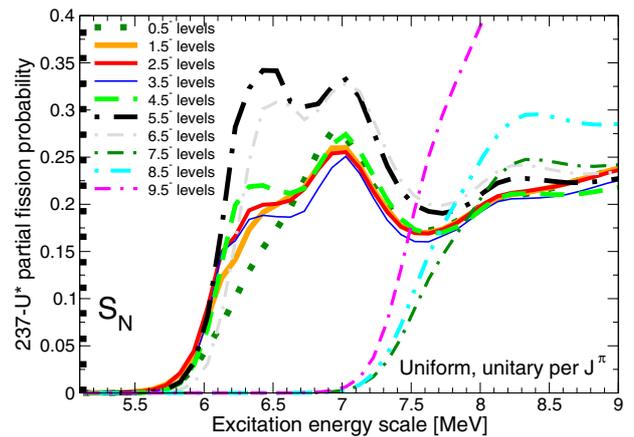


Figure 2. Monte Carlo \mathcal{R} -matrix double barrier surrogate-like fission probabilities of $^{237}\text{U}^*$ as a function of resonance spin for negative parities and excitation energy up to a neutron kinetic energy of 4 MeV. The vertical-dashed bar at 5.13 MeV materializes neutron emission threshold. Note: for unbiased analysis, those transfer-like calculations are fed with an entrance CN population both uniform and unitary per J^π .

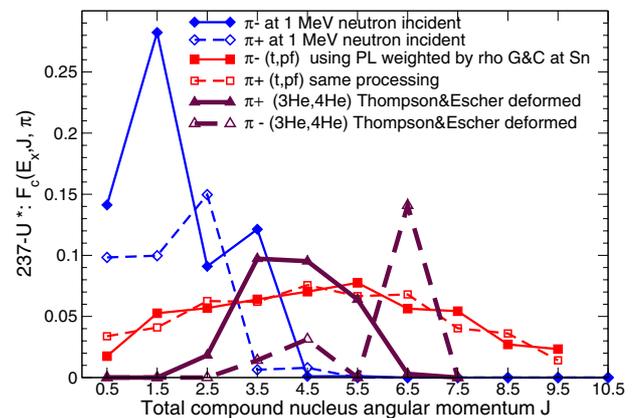


Figure 3. Population distribution of total angular momenta corresponding to the $^{237}\text{U}^*$ resulting from either a neutron-induced reaction ($n+^{236}\text{U}$) at 1 MeV neutron energy or derived from (t,p) DWBA calculations, early made by Back et al. [17]. $^{238}\text{U}(^3\text{He},^4\text{He})^{237}\text{U}^*$ calculations from Thompson and Escher [18], to be used with caution since preliminary, supply a plausible picture of that direct entrance reaction. Solid lines connecting the symbols and addressing negative parity (respectively dashed lines for positive π) are drawn to guide the eye.

[Hyp. B)] the WE hypothesis of reaction decay probability spin-parity independence [4],

[Hyp. C)] the idealized matching between neutron-induced and surrogate spin-parity entrance distributions; reading $\mathcal{F}_{\text{sur}}^{CN}(E_x, J, \pi) \equiv \mathcal{F}_n^{CN}(E_n, J, \pi)$. The impact is similar to that of hypothesis B.

2.1. Modified WFCF definition according to surrogate reactions (WFCF \rightarrow SWFCF)

At this stage, it is worthwhile to think about conceptual differences between surrogate and neutron physics spectroscopy (NPS) measurements. As far as NPS are concerned and neutron reactor applications, the total cross

section over reactor spectrum ($0 < E_n < 20$ MeV) is well characterized by the only sum of shape elastic and CN cross sections. It simply reflects the fact that the interaction time is long enough to wash out any prior history making sufficient the use of CN theory. On the opposite because of the high energy of the incident charged particle requested to overcome the coulomb barrier (e.g; 24 MeV selected for the ^3He beam of present investigated $^{238}\text{U}(^3\text{He}, ^4\text{He})^{237}\text{U}^*$ data), any final excited nucleus (prior to decay) preserves in term of its spin-parity state population the characteristics of the first individual nucleon excitations (leading to a ‘CN’) making direct reactions (other than elastic) and pre-equilibrium reactions sizable and associated modeling critical. Among reactions other than compound and ranked in decreasing order of importance for present application, are direct pickup, pre-equilibrium and direct inelastic contributions. Thus we can debate on the strength of the correlation between entrance and exit channels when the entrance channel width is of single-particle state character (case of surrogate measurements) rather than of compound nucleus state nature (case of NPS). The regular $W_{c,c'}$ definition has already been challenged in Ref. [15]. The outcome was a surrogate-dedicated formulation of $W_{c,c'}$ which applied to the fissile $^{240}\text{Pu}^*$ and fertile $^{241}\text{Pu}^*$ compound nuclides demonstrated special behavior that we are not accustomed to deal with. In particular it enlightens that both radiative and fission decays can now endorse the role of the enhanced channel, no more acted by the elastic channel. The enhancement factor observed on both fission and γ -decay probabilities is especially strong for fertile heavy isotopes as the $^{237}\text{U}^*$. Surrogate-dedicated width fluctuation correction factor (SWFCF) values according to this isotope show an increase by up to a factor of 2.6 (Fig. 1 - fission and γ -decays) in the energy range above neutron ‘inelastic’ emission threshold ($S_N + 0.1$ MeV) although the actual consequence of wrong (or the absence of) correction on small fission decay probability (magnitude < 0.1) for fertile nuclides remains limited. On the opposite, this clearly has some importance whenever accurate extrapolation of neutron-induced capture cross section from surrogate data is foreseen.

2.2. Monte Carlo reaction decay probabilities

Real situation commands to avoid the decoupling hypotheses between reaction decay probabilities ($\mathcal{P}_{c'}^{J^\pi}$ of Eqs. (1) or (2)) and WFCF (or SWFCF) since statistical properties of normal and fission isomer deformation reaction level widths are obviously correlated [9]. We also realize that an exact solution for the HF equation relies on the possible derivation of an analytical expression pertaining to the actual coupling strength situation between both resonance wells of the fission barrier. A powerful alternative to analytical formulae is our Monte Carlo-type (MC) method [9] which presents the advantage of providing either average cross sections or surrogate-like decay probabilities taking full account of the various parameter statistical fluctuations under relevant coupling conditions. The MC approach leads to a compact formulation of the surrogate-dedicated master equation

(Eq. (2)) meaning,

$$\mathfrak{P}_{surr,c'}(E_x) = \sum_{J^\pi} \left[\mathcal{F}_{surr}^{CN}(E_x, J, \pi) \times \mathcal{P}^{\mathcal{MC}J^\pi}_{surr,c'}(E_{c'}) \right]. \quad (3)$$

The $^{237}\text{U}^*$ is characterized by its odd-N character and subsequent low energy combination of single-quasineutron state with vibrational states. We then expect fewer Bohr transition spectrum specificities than for even-even nuclides because of the dissimilar nature of quasiparticles as well as denser level density right above Fermi energy. Figure 2, plotting the surrogate-like MC partial fission probabilities for transition states of negative parities, returns some hints about possible impact of the WE hypothesis of reaction decay probability spin-parity independence. We observe three groups of shapes corresponding respectively to total level spin sequences less than $\frac{11}{2}\hbar$, equal to $\frac{11}{2}\hbar$ and $\frac{13}{2}\hbar$, and finally larger than $\frac{13}{2}\hbar$. The latter group corresponds to fission across continuum states located at least 1 MeV above the two fission barrier humps which spin-parity is not expected within the discrete transition state energy range. We underline that present MC simulation was carried out assuming full second well β -vibrational resonance damping [11] and so, no giant resonance patterns can be observed on Fig. 2. Present strong damping strength choice is based on little evidence of wide fluctuations both in (t,p) and (d,p) fission decay measurements [16] as well as in the presently analyzed $^{238}\text{U}(^3\text{He}, ^4\text{He})^{237}\text{U}^*$ measurement. We emphasize that incompletely damped vibrational approach [11] is frequently requested for even-even fissioning nuclides whose giant resonance patterns are mainly correlated to a sparser discrete transition state sequence on barrier tops.

2.3. Spin-parity population as function of entrance vector for surrogate-like MC simulation

CN state population distribution of total angular momenta corresponding to the neutron-induced reaction (the $\mathcal{F}_n^{CN}(E_n, J, \pi)$ factor of Eq. (1)) is extracted directly from appropriate optical model potential or using the \mathcal{AVXSF} code [9] on the ground of Moldauer neutron transmission coefficient prescription. As far as surrogate data related to $^{237}\text{U}^*$ are concerned, present $\mathcal{F}_{surr}^{CN}(E_x, J, \pi)$ are based on distorted-wave Born approximation (DWBA) calculations early performed by Back et al. [17] for the (t,p) direct interaction. Present $^{238}\text{U}(^3\text{He}, ^4\text{He})^{237}\text{U}^*$ simulations have been fed with recent direct calculations from Thompson and Escher [18]. The latter must be considered with caution since being preliminary [19]. However it brings a plausible direct reaction pattern for the present simulation. Figure 3 displays the assumed entrance distributions.

3. \mathcal{AVXSF} surrogate-like γ and fission probability simulations applied to $^{237}\text{U}^*$

On the ground of a more secure approach using Monte Carlo-based \mathcal{R} -matrix decay probabilities [9] and suitable prior to decay CN state population distribution, neutron-induced average partial cross sections and surrogate-like probability simulations according to

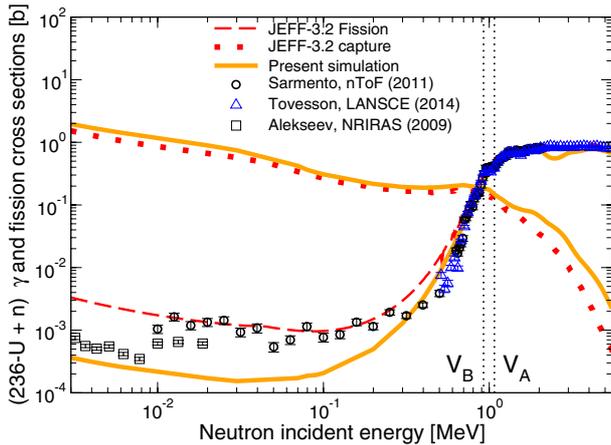


Figure 4. ^{236}U neutron-induced fission and capture cross sections (orange-solid curves) computed with \mathcal{AVXSF} and compared with the European evaluated data file JEFF-3.2 and recent NPS measurements. Best estimates of inner and outer barrier heights are represented by vertical dashes; noted $V_{X=A,B}$ relatively to S_N on above Figure.

$^{237}\text{U}^*$ have been performed simultaneously. Preliminary results are displayed on Figs. 4 and 5 regarding the γ and fission channels. Present \mathcal{AVXSF} neutron-induced fission cross section undercuts under-threshold fission barrier experimental data although the theoretical curve merges with A. Alekseev et al. data (*EXFOR 41503*) at low energy. Above the fission threshold, $\sigma_{n,f}$ exhibits an abnormal cusp (at about 2.5 MeV) that requires more attention. In terms of surrogate-like probability simulations (Fig. 5), present preliminary results contrast with the historical surrogate-reaction method reconstruction [2] since the agreement with the experimental data in terms of γ -channel is now reached although the treatment of the entrance direct pattern remains approximative [19]. This confirms that the surrogate-reaction method is inappropriate in terms of capture cross section reconstruction since it is pertaining that hypothesis **B**) does not hold in terms of γ channel (see Fig. 6 of Ref. [2]) and that the capture right above S_n is deeply impacted by the absence of SWFCF corrections (Fig. 1).

4. Summary and perspectives

Promising results off the surrogate-reaction method were obtained consistently in terms of cross sections and surrogate probabilities according to both fission and γ channels; thanks to the recent simultaneous fission and γ channels $^{238}\text{U}(^3\text{He},^4\text{He})^{237}\text{U}^*$ surrogate measurements [12]. However, spin-parity entrance population distribution calculations remain a genuine challenge that needs special attention in future developments of

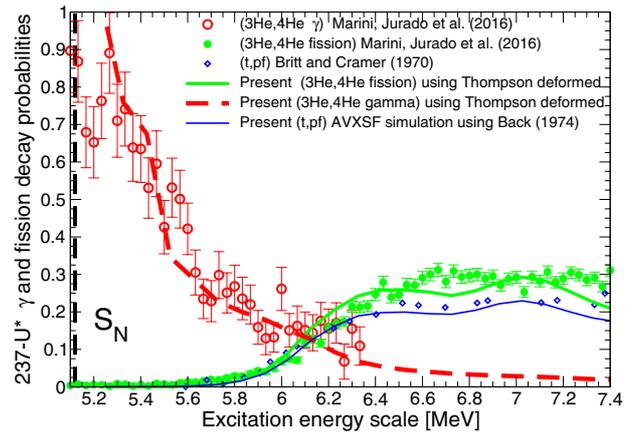


Figure 5. $^{237}\text{U}^*$ fission (solid curves) and γ (dashed curve) surrogate-like probabilities computed with \mathcal{AVXSF} and compared with preliminary $^{238}\text{U}(^3\text{He},^4\text{He})^{237}\text{U}^*$ surrogate data (circles) from P. Marini, B. Jurado et al. [12] and (t,pf) data (diamonds) from Britt and Cramer [16].

present approach. Extended experimental database of γ -decay surrogate probabilities is welcome for actinides to test further present strategy and that especially on the isotopes of the Pu family recently investigated [9, 11]. Forthcoming integration of our approach is expected in a near future at CEA cadarache to improve standard neutron cross section data evaluation method.

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