Monte Carlo simulation of γ and fission transfer reactions using extended $\mathcal{R}$-matrix theory

Olivier Bouland[1],*

[1]CEA, DES, IRESNE, DER, SPRC, Physics Studies Laboratory, Cadarache, F-13108 Saint-Paul-lez-Durance, France

Abstract. This paper comes back on the accuracy of the *surrogate-reaction method* (SRM) historically used for neutron-induced average partial cross sections inference from measured surrogate-reaction probabilities. The SRM level of performance is examined in relation to a reasonably accurate reference calculation performed with the $\mathcal{AVXSF}$-$\mathcal{LNG}$ code [1] through a challenging test case : the ^{240}Pu* compound system. This paper argues on some ingredients of the reference calculation [2] and returns some hints about the failure now well-known of the neutron-induced γ average cross section inference. It shows also that in some special cases, the SRM can be poorly accurate also in terms of neutron-induced fission average cross section inference.

1 Introduction

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 [3]. Over the years, a variety of surrogate reactions have been used as stripping (d,p) and pickup (p,d) reactions, (^{3}He,p), (^{3}He,d) and (^{3}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 surrogate-reaction method (SRM) [4]. Early promising comparisons [3] made between neutron-inferred fission cross sections and fission cross sections directly measured by neutron spectroscopy led to agreement within 10% to 20% at neutron energy above the two quasi-particle excitations energy gap although exhibiting larger deviations at lower energies. Major limitations in neutron fission cross section inference from surrogate-reaction data were promptly noticed [3, 5] with the difficulty to estimate a) the compound nucleus formation cross section by neutron absorption, b) the possible influence of the differences between the angular momentum distributions populated by neutron capture and direct reactions and c) the validity of the Weisskopf-Ewing (WE) hypothesis on reaction decay probability spin-parity independence [6].

Nearly two decades ago, surrogate reactions received renewed interest in terms either of simulation [7, 8] or experimental investigation (beginning by the study [9]; reviewed in Ref. [10]) to infer in addition to fission, neutron-induced radiative-capture cross sections. Newly measured γ-ray emission probabilities were readily analyzed within the SRM that proved to be very poor in terms of inferred neutron-induced capture cross sections [4]. Indeed the SRM 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 circumvented even if strong difficulties remain in the modeling of direct reactions. Keep using the SRM might suggest that surrogate-reaction data are inappropriate to help although surrogate-reaction spectroscopy is definitively of great help for target material with unsuitable lifetimes (less than several days) or with high radiotoxicity.

In a previous paper [1], 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 (MC) 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 [11]. We must enlighten that we were also able to achieve confident prediction by inclusion of fission probability data induced by (d,p), (t,p), (^{3}He,t) or even (^{3}He,d) direct reactions. The surrogate-fission data analysis of our study is being documented [2, 12]. Across those papers we do not cope with probabilities measured according to γ-decay because of historical 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. [4]. A series of new surrogate-reaction data dealing simultaneously with γ-emission and fission probabilities induced by transfer and inelastic scattering reactions of light projectile nuclei impinging on heavy target nuclei, is soon to be re-

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[*] e-mail: olivier.bouland@cea.fr

leased following the recent development of a dedicated experimental set-up [13]. This will give us the opportunity to quantify the reliability of present MC $\mathcal{R}$-matrix technique for confident inference of neutron-induced capture cross sections below the energy range of second chance fission.

2 Recalls on the SRM hypotheses

The starting point and appropriate formalism for describing compound nucleus (CN) reactions is pure Hauser-Feshbach (HF) statistical theory [14] together with the familiar (energy-dependent) in-out-going channel width fluctuation correction factor (WFCF), $W_{c,c'}$. Applied to neutron-induced reactions at the energy E_n, this can be written [2] concisely for the partial average cross section,

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

with $\sigma_n^{CN}(E_n)$, the neutron-induced total compound nucleus formation cross section. $\mathcal{F}_n^{CN}(E_n, J, \pi) = \sigma_n^{CN}(E_n, J, \pi)/\sigma_n^{CN}(E_n)$ is the fraction of CN excited states formed with given J^π (spin/parity). $\mathcal{B}_{c'}^{J^\pi}(E_{c'})$ is the individual decay probability (or branching ratio) into channel c' from a (J, π) state. Similarly the surrogate-reaction probability can be formulated as,

$$\mathcal{P}_{surr,c'}^{A^*}(E_x) = \sum_{J^\pi} \mathcal{F}_{surr}^{A^*}(E_x, J, \pi) \times \mathcal{B}_{c'}^{J^\pi}(E_x) \times W_{surr,c'}^{J^\pi} \tag{2}$$

The historical SRM is based on the following hypotheses:

A) the absence of WFCF in the cross section formulation. We know however that it plays in low-energy neutron-induced reactions ($E_n < 2$ MeV) a major role in averaging over partial width distributions when calculating average cross sections. This was well illustrated in the neutron-induced WFCF model comparisons by Hilaire *et al.* [15],

B) the WE hypothesis [6] of reaction decay probability spin-parity independence applied to the $\mathcal{B}_{c'}^{J^\pi}(E_{c'})$ term,

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

3 In-house Monte Carlo reaction decay probabilities algorithm

In the spirit of the extended $\mathcal{R}$-matrix MC simulations made according to the low-energy neutron-induced fission cross sections for the isotopes of the Pu family [1], a counterpart of the analytical Eq.(2) was programmed in the $\mathcal{AVXSF}$-$\mathcal{LNG}$ (*AVerage CROSS Section F*ission - *L*ynn and *N*ext *G*eneration) code to be able to calculate both neutron-induced cross sections and surrogate-reaction probabilities with an unique set of input nuclear structure parameters. On this secure footing the master equation dedicated to surrogate-reactions is

$$\mathcal{P}_{surr,c'}^{A^*}(E_x) \;=\; \sum_{J^\pi} \left[\mathcal{F}_{surr}^{A^*}(E_x, J, \pi) \times \mathcal{B}_{c',MC-surr}^{J^\pi}(E_x) \right] \tag{3}$$

Eq.(3) above carries the advantage of not decoupling the various terms involved in $\mathcal{B}_{c',MC-surr}^{J^\pi}(E_x)$ such as $\mathcal{B}_{c'}^{J^\pi}(E_x)$ and $W_{surr,c'}^{J^\pi}$. Both terms are merged in one single that is calculated using an efficient MC algorithm described in much details in [1, 2]. This MC approach is definitively sensitive in terms of fission channels whose treatment involves 1) an additional WFCF, the W_{II}, dedicated to the correlation between coupling and fission widths of the class-II states lying in the second well of the double-humped fission barrier and 2) a correction of the associated number of degrees of freedom [16]. Present formulation (Eq.(3)) also does not consider the excited nucleus prior to decay (A^*) necessarily as a *compound nucleus*, meaning as a nucleus in a complete thermal equilibrium. A fraction, possibly large, of the observed decay can be simply induced by the first states formed in the chain of nucleon-nucleon collisions after the two-body interaction. In this picture, the profile of the quantity $\mathcal{F}_{surr}^{A^*}(E_x, J, \pi)$ witnesses the various components of the reaction forming A^* at the time of the decay. This statement can be made because of the reactions used in surrogate-reaction spectroscopy for which the ingoing charged-particles energy has to be higher than the Coulomb barrier. This is illustrated by the ^{238}U(^{3}He,^{4}He)^{237}U* measurement [17] that used a 24 MeV ^{3}He beam. At such high energy, the excited nucleus is not well equilibrated and this question must be taken into consideration when formulating the WFCF.

3.1 Modified WFCF definition according to surrogate reactions (WFCF → SWFCF)

In view of the considerations above, 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-reaction measurements) rather than of compound nucleus state nature (case of neutron spectroscopy). This is equivalent to ask if the formulation of $W_{surr,c'}^{J^\pi}$ must be identical to the standard $W_{n,c'}^{J^\pi}$ factor [15]. The answer is most likely not. The outcome is a surrogate-reaction-dedicated formulation of $W_{c,c'}$, so called SWFCF for the calculation of Eq.(2) or equivalently in the MC algorithm (Eq.(3)). Details about its formulation are given in Ref. [2]. Application to the fissile ^{240}Pu* compound system returns the SWFCF shapes of Fig.(1). From this, we first recover the customary high-energy pattern since each partial reaction SWFCF curve tends to unity when the total number of reaction channels involved becomes very large; in practice at minimum above $(S_n + 1.0)$ MeV. The absence of elastic channel correlation prevents any classic elastic enhancement and we observe that both radiative and fission decays can now endorse the role of the enhanced channel with maximum impact on the γ decay channel; +20% of enhancement above

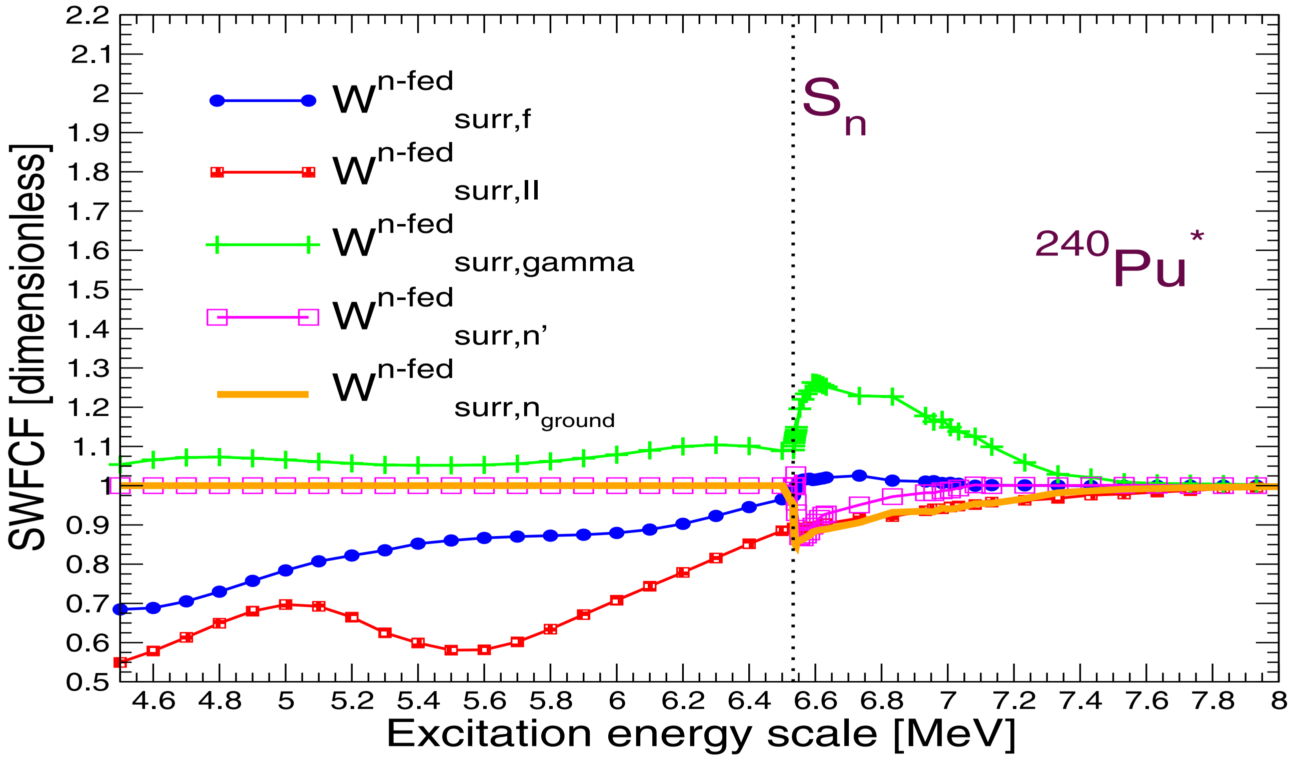

Figure 1. (Color online) Comparison of SWFCF curves associated to fission ($W_{surr,f}$), γ decay ($W_{surr,\gamma}$) and neutron emission with residual nucleus in ground state ($W_{surr,n_{ground}}$) or in excited states ($W_{surr,n'}$) according to surrogate reactions for the ^{240}Pu* over the whole excitation energy range. $W_{surr,II}$ is a specific correction for the statistical fluctuations of the widths of the class-II states lying in the second well of a double-humped fission barrier. The various SWFCF displayed must be seen as global coefficients related to each partial reaction but integrated over all J^π compound system excited states. For easier visualization, they are calculated with the neutron-incident excited-state population; (n-fed) superscript.

(S_n + 100) keV in present fissile nucleus case. By reciprocity, neutron emission channels are depreciated accordingly to the total amount of reaction rate redistributed. We notice the new role of $W_{surr,n_{ground}}$, the width fluctuation correction factor related to neutron emission with residual nucleus in ground state since this channel represents above S_n the largest flux contributor to the capture channel. Below S_n the flux is redistributed from the fission channel ($W_{surr,f}$) to the γ channel ($W_{surr,\gamma}$). It shrinks dramatically as excitation energy decreases and so, the enhancement on radiative decay remains moderate and constant (~ 10%). In Fig.(1), one observes in terms of $W_{surr,II}$ limited impact over the range S_n to (S_n+1 MeV) but increasing negative correction as decreases the excitation energy (larger than 30% below 5.65 MeV that is the height [1] of the highest fundamental hump of the double-humped fission barrier for the ^{240}Pu*).

4 In-house simulated surrogate-reaction probabilities vs SRM inference

The extended $\mathcal{R}$-matrix MC approach carried in this work gives us the opportunity to compare, once and for all, the SRM-based reaction probabilities to those obtained from a *reasonably accurate* reference (this work). The application will be here below made on the most tricky case that is the ^{240}Pu* compound system. First, any (even Z - even N) character fissioning nucleus includes a sparse discrete transition state sequence [1] on top of fundamental barrier heights and as possible consequence is the failure of the WE hypothesis of fission decay probability spin-parity independence [6]. Beyond this, the ^{240}Pu* is very specific in the way that according to neutron-incident s waves, two J^π states can be excited : 0^+ and 1^+. Low-neutron-energy fission-decay magnitude is then correlated to the accessibility of J^π=0^+ and 1^+ transition states. Unfortunately only few J^π=0^+ transition states can contribute to the fission and in addition, the fission across 1^+ transition states plays little role [2]. This definitively invalid the WE hypothesis. Figure (2) shows the comparison between the SRM-based surrogate-reaction probabilities with the present $\mathcal{LNG}$ code results according to the ^{240}Pu(α,α'c')^{240}Pu* reactions with c'= γ or f. The $\mathcal{F}^{A^*}_{\alpha'}(E_x, J, \pi)$ distribution as input to the $\mathcal{LNG}$ code is supplied easily by the TALYS code [18] that triggers an ECIS06 optical model DWBA calculation. The SRM-based reaction probabilities are simply the corresponding neutron reaction cross sections reconstructed from the JEFF-3.3 evaluation [19] divided by the neutron-induced total compound nucleus formation cross section $\sigma_n^{CN}(E_n)$ of the Eq.(1) according to the ^{240}Pu* excited nucleus. This latter quantity is also supplied by the TALYS calculation.

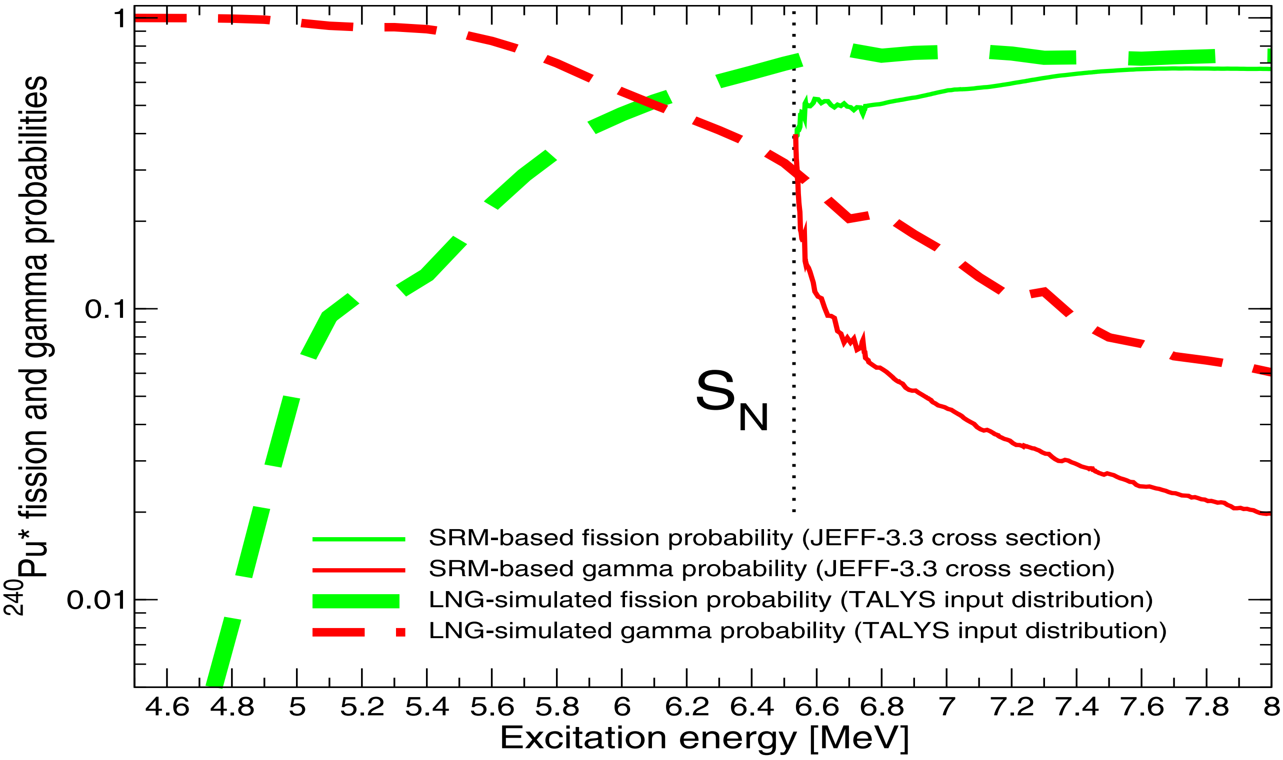

Figure 2. (Color online) Fission and γ probabilities according to the ^{240}Pu$(\alpha,\alpha' f)^{240}$Pu* and ^{240}Pu$(\alpha,\alpha'\gamma)^{240}$Pu* reactions as a function of excitation energy. The neutron emission threshold (S_n) quoted provides the neutron-scaled baseline (dotted vertical line). SRM-based estimates are represented by thin solid curves to be compared to our reference simulation (present work; thick dashed line).

5 Conclusion

Figure (2), as itself, well sums up the level of performance that can be reached by the SRM. It demonstrates again that the SRM fails in terms of neutron-induced capture cross section inference but also shows that it can be poor also in terms of neutron-induced fission cross section inference in some cases, especially for the peculiar ^{240}Pu* compound system [12]. It recalls that for heavy excited nuclei, any hope in accurate capture cross section inference relies above all, on an accurate fission channel treatment. The shape of $W_{surr,\gamma}$ that contrasts strongly with the standard $W_{n,\gamma}$ profile is also part of the explanation [2] of the systematic failure of the SRM in terms of neutron-induced radiative-capture cross section inference in the non-statistical energy range. As a conclusion, I would like to emphasize that nowadays there are no more arguments to carry the approximations brought by the *surrogate-reaction method*.

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