Importance of the residual nucleus level density in the calculation of the $^{239}$Pu($n$, $2n$)$^{238}$Pu excitation function

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Abstract. In parallel with the recent measurement by Méot et al. [1] of the $^{239}$Pu($n$, $2n$)$^{238}$Pu reaction cross section in the [7-10] MeV neutron-incident energy range, a modeling work was undertaken to support the new measurement. This theoretical framework is here dedicated to study the impact of the type of level density for the $^{239}$Pu residual nucleus (RN) on the final shape of the low-energy tail of the ($n$, $2n$) excitation function. For this purpose, the AVXSF-LNG program [4] has been upgraded to model second-chance reactions and coupled to the TALYS-ECIS06 nuclear reaction codes [3] that are used to provide the compound nucleus (CN) cross section to AVXSF-LNG as a function of the selected pre-equilibrium model. The spin-dependent population of the RN obtained after CN emission is commonly advertised when using an exciton-based model for calculating the total pre-equilibrium cross section. On the other hand a population based on a particle-hole state density is rather expected when using the MSD/MSC quantum mechanical description in place of the exciton-based model. As far as a spectrum of a few number of individual excitations is favored, the combinatorial Quasi-Particle-Vibrational-Rotational Level Density method [4] implemented in the AVXSF-LNG computer program is well suited to model the various alternatives. Several RN level densities have been tested in this work. It includes the testing of the Quasi-particle Random Phase Approximation-based result of Ref. [6]. As a guide line, the amount of pre-equilibrium flux in the total ($n$, $2n$) excitation function is being studied as a function of the type of level density selected for the residual nucleus.

1 Introduction

The importance of the residual nucleus (RN) level density (LD) in the calculation of the $^{239}$Pu($n$, $2n$)$^{238}$Pu reaction cross section has been raised up in the analysis of the recent measurement by Méot et al. [1] who used the recoil method for counting $^{238}$Pu nuclei. The latter experiment covers the [7-10] MeV neutron-incident energy range (Fig. 1; black-filled squares), right above the neutron-emission threshold ($E_{239}^{n}$ in Fig. 2) of the $^{239}$Pu RN. In this energy range, it is well-acknowledged [2] that pre-equilibrium reactions occur on a time scale comparable to the target-traversing-time by the projectile, after direct processes but well long before the statistical statistical equilibrium is reached (compound nucleus (CN) stage). Precise calculation of the pre-equilibrium contribution for the $^{239}$Pu($n$, $2n$) excitation function is strongly model-dependent. For instance, the TALYS-ECIS06 nuclear reaction system [3] used as an input to present study, provides two options (official releases) for calculating the total pre-equilibrium cross section:

- The first one involves a semi-classical two-component exciton model,
- The second is based on a quantum-mechanical multi-step direct (MSD) / multi-step compound (MSC) model.

Literature on semi-classical models reports a lack of accuracy to describe the angular distributions of the emitted particles, contrary to the MSD/MSC, since the MSD approach preserves some memory of the incident particle direction. Aside the use of the most performing model to calculate the total pre-equilibrium cross se-
tion, we must evaluate the sensitivity of the $^{239}\text{Pu}(n,2n)$ excitation function to the pre-equilibrium population (or level density) of the $^{238}\text{Pu}$ residual nucleus. Indeed, after entering the target nucleus, the interaction of the incident neutron with one nucleon progressively leads to more and more complicated states of excitation in the compound system (n + $^{239}\text{Pu}$). This is equivalent to say that the pre-equilibrium phase is expected to carry a spin-parity level-density distribution more singular than the one of a compound nucleus. In this paper, we show the impact of the type of level density selected for the non-equilibrated residual nucleus on the total (n,2n) cross section shape. We recall that the latter results from the sum of two processes: one of compound nucleus type and another of direct-like type.

Figure 2 shows a schematic drawing of the potential wells of the three nuclei involved in the $^{239}\text{Pu}(n,2n)^{238}\text{Pu}$ reaction below $E_n=12$ MeV neutron incident energy. This second-chance reaction occurs if, and only if, the inelastically-scattered neutrons ($n'$) reach a state of excitation in the $^{239}\text{Pu}$ residual nucleus above the neutron binding energy, $B_{239}$. Then a second neutron ($n''$) is possibly emitted leading to the $^{238}\text{Pu}$ second-chance residual nucleus. Both discrete and continuum states in the $^{239}\text{Pu}$ residual nucleus potential well encompass compound and direct-like mechanisms. The standard Hauser-Feshbach (HF) statistical theory of nuclear reactions [8] is widely used to calculate compound nucleus reaction cross sections. In this study, HF calculations modified to treat the fission deexcitation channel in the R-matrix formalism are carried out with the AVXSF-LNG [4] program, that is newly designed to model second-chance reaction cross sections using a dedicated reaction decay-probabilities module [5]. Direct elastic and inelastic scattering cross sections using a dedicated reaction decay-probabilities module [5]. Direct elastic and inelastic scattering cross sections as well as the total inelastic pre-equilibrium cross section corresponding to the n+$^{239}\text{Pu}$ entrance channel are computed with the TALYS-ECIS06 nuclear reaction system of codes [3]. The key elements of our modeling of the $^{239}\text{Pu}(n,2n)$ cross section are given in the next section.

2 Fraction of pre-equilibrium flux in the total (n,2n) cross section of the $^{239}\text{Pu}$

An overview of the importance of the pre-equilibrium (PE) mechanism for building the total (n,2n) cross section, $\sigma_{n,2n}^{\text{tot}}$, starts by estimating the fraction $\sigma_{n,2n}^{\text{PE}}/\sigma_{n,2n}^{\text{tot}}$. This quantity is obtained by coupling the TALYS-ECIS06 nuclear reaction system [3] with the AVXSF-LNG code [4]. The AVXSF-LNG program is preferred to the HF module of the TALYS code for the special treatment of the fission channel and the two different models for calculating channel width correction fluctuation factors (WFCF and SWFCF [5]). Latter definition sounds to be more appropriate to model second-chance decay-probability fluctuations. The present calculation is made in three steps:

1. The TALYS calculation provides $\sigma_{n,2n}^{\text{PE}}(E_n)$, the total pre-equilibrium cross section corresponding to the $^{239}\text{Pu}(n,n')$ direct-like channels.

2. Since $\sigma_{n,2n}^{\text{PE}}(E_n)$ regroups all inelastic direct-like transitions covering from $E_n$ to the lowest continuum energy bin of the residual nucleus, $\sigma_{n,2n}^{\text{PE}}(E_n)$ must be weighted by $\rho_{\text{RN}}(\Delta E_{\text{bin}};E_{\text{bin}})$, the relative spin-parity-dependent population of each bin $\Delta E_{\text{bin}}$, to assess $\sigma_{n,2n}^{\text{PE}}(E_n \rightarrow \Delta E_{\text{bin}};E_{\text{bin}})$, the partial pre-equilibrium cross section per bin,

3. $\sigma_{n,2n}^{\text{PE}}(E_n)$, the pre-equilibrium component of the total (n,2n) cross section, is finally calculated using $\mathcal{P}_{n''}$, the spin-parity-dependent second-chance neutron-emission probability that is computed by the AVXSF-LNG code.

The third step is equivalent to the following equation,

$$\sigma_{n,2n}^{\text{PE}}(E_n) = \sum_{k \in \Delta E_{\text{bin}}} \sum_{x \in \Delta E_{\text{bin}}} \sigma_{n,2n}^{\text{PE}}(E_n \times \mathcal{P}_{n''}, (1)$$

where $x$ is a given energy bin $\Delta E_{\text{bin}}$ of the continuum range $\Delta E'$ in the $^{239}\text{Pu}$ residual nucleus and $k$, a given energy bin $\delta E_{\text{bin}}$ (or a low-lying level) of the corresponding energy range $\Delta E''$ in the $^{238}\text{Pu}$ second-chance residual nucleus. Figure 3 shows the total $^{239}\text{Pu}(n,2n)$ cross section
over the [6-12] MeV incident neutron energy range and its compound nucleus component. The pre-equilibrium contribution is calculated as the difference between the total and CN curves. It represents at 9 MeV about 17% of $\sigma_{n2n}^{\text{pre}}$ and depends on the evaluation of two quantities: $\rho_{\text{RN}}(\Delta E_{\text{bin}})$ (as required by step 1) and $\rho_{\text{RN}}(\Delta E_{\text{bin}})$ (as required by step 2). For a long time, the former was modeled by the classical exciton model of pre-equilibrium neutron emission [10, 11], but nowadays quantum mechanical descriptions [6, 12] are favored. As mentioned in the introduction, TALYS [3] provides both classical and quantum-mechanical approaches.

Figure 4 shows, for the n$^+$$^{239}$Pu reaction below 12 MeV, a comparison in terms of magnitude and shape of the two different models for calculating the pre-equilibrium cross section with TALYS. The pre-equilibrium cross section is summed up with the CN inelastic cross section (computed by AVXSFLNG) and direct inelastic cross section (TALYS) to form the total inelastic cross section. The latter, reconstructed by AVXSFLNG (Fig. 4), is compared to the ENDF/B-VIII.0 evaluated curve for reference. Although the two distinct pre-equilibrium cross section profiles remain close right above the neutron-emission threshold, the two calculations (without renormalization) differ significantly above 4.6 MeV with a steeper ‘exciton-based’ cross section slope (and a cross section value 3 times larger at 20 MeV). In this work, we have chosen the MSD/MSC model of the TALYS code with an upward renormalization of 1.39. We are now ready to discuss in the next section the sensitivity of the total $^{239}$Pu(n,2n) cross section to $\rho_{\text{RN}}$, the second variable satisfying the step 2.

### 3 Dependence of the total $^{239}$Pu(n,2n) cross section on the RN LD

We now investigate the sensitivity of the total (n,2n) excitation function to the type of residual nucleus level density selected. As stated, the total (n,2n) cross section is constructed as the sum of CN and pre-equilibrium contributions through the $^{239}$Pu(n,n$^+$) reaction. For the exciton pre-equilibrium model, a residual nucleus level density determined as the LD after compound nucleus emission provides reliable results [3]. However the hypothesis for the RN of a level density pattern corresponding to a few particle-hole excitations seems to be more consistent with a non-equilibrated residual nucleus. As far as a spectrum corresponding to a few number of individual excitations is expected, the combinatorial Quasi-Particle-Vibrational-Rotational (QPVR) LD method implemented in AVXSFLNG [4] is appropriate to fulfill this goal. In contrast to the exciton approach provided in the TALYS code, our phenomenological model is based on the concept of excited quasi-particles for the individual excitations. We start from the formula for the quasi-particle energy, in terms of the independent particle states of a Nilsson-type spectrum in a deformed potential well with the pairing energy as parameter. The latter being taken not from the solution of the full pairing equations [13] but as a parameter value evaluated from experiments.

In this work, we have tested several types of level density for the $^{239}$Pu residual nucleus to quantify its impact on the calculated total (n,2n) cross section. Among those, Figure 5 shows the results corresponding to a level density after compound nucleus emission and to a pre-equilibrium phase restrained to the excited states formed by the breaking of one pair of neutron quasi-particles combined with one-neutron quasi-particle state excitations of the $^{239}$Pu ground state; ensemble of configurations noted {OP-1N},{OP-3N}. The extension of the present work includes
the test of the level density spin distribution resulting from the Quasi-particle Random Phase Approximation (QRPA) calculation of Ref. [6]. Since the QRPA results for the $^{239}$Pu residual nucleus were not available, we have selected the distribution obtained for a residual nucleus formed after the pre-equilibrium emission of one neutron in the neighboring $n+^{238}$U reaction. This distribution, so far limited to the contribution of the natural parity states, $\pi = (-1)^J$ [6], is expected to carry into our calculation the singular pattern commonly shown by the QRPA results. Figure 6 in Ref. [6] shows for the QRPA level spin distribution a weak excitation energy dependence and a sharp distribution centered around $J^P = 3^-$. Figure 5 shows large differences between the QRPA-based curve (green dots) and other LD options for the excitation function over the neutron-emission threshold energy region. At final, one observes good agreement among the presented calculations over a wide range, independently of the level density model, from 6.5 up to 12 MeV neutron incident energy.

4 Conclusion

The present study is part of a more extensive work motivated by the recent measurement performed by Méot et al. [1] of the $^{239}$Pu($n$,2$n$)$^{238}$Pu reaction cross section that is based on the recoil method for counting the $^{238}$Pu nuclei. Latter measurement aims to bring some clarification on the profile of the low-energy tail of the total (n,2n) excitation function. To benchmark the new data set with an up-to-date calculation, the TALYS-ECIS06 [3] system of codes was coupled with an upgraded version of the AVXSFLNG code [4] that is now designed to deal with second-chance reactions using a dedicated decay-reaction-probabilities module [5]. The present result for the total (n,2n) cross section confirms that at moderately-fast energies the CN component (treated with Hauser-Feshbach extended R-matrix theory) still represents the largest part of the total (n,2n) cross section for an actinide. This component clearly demonstrates a wavy shape feature that is amplified, for the total (n,2n) curve, by the presence of a singular pattern in the residual nucleus level density. This study enlightens a major difference in shape and magnitude between the two distinct ways provided by TALYS for calculating $\sigma_{n,n}^{PE}(E_n)$, the total pre-equilibrium cross section; either being based on the two-exciton semi-classical or the MSD/MSC quantum mechanical approach. The question of the most relevant level density for describing a non-equilibrated residual nucleus has been raised and several options tested. Among them, the QRPA-based LD that shows the most specific pattern, leads to significant differences with alternative level densities for the total (n,2n) excitation function over the energy region of the neutron-emission threshold.

References