Role of the direct mechanisms in the deuteron-induced surrogate reactions

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Abstract

An extended analysis of the key role of direct interactions, i.e., breakup, stripping and pick-up processes, for the deuteron-induced surrogate reactions is presented. Particular comments concern the deuteron breakup which is dominant in the case of the $(d, p\gamma)$ surrogate reactions on actinides target nuclei, around the Coulomb barrier.

1. Introduction

The surrogate reaction method is an indirect measurement technique proposed by Cramer and Brit [1] to overcome the difficult problems of preparing and handling the highly radioactive targets required for cross-section measurements. Therefore, the outgoing channel of interest is studied via an alternative "surrogate" reaction that involves a projectile-target combination more accessible experimentally. The use of surrogate reaction method (e.g., [2, 3, 4, 5, 6, 7, 8, 9]) to provide indirect informations on cross sections that can not be measured directly or calculated accurately is steadily increasing. Since last decade this method has been involved mainly in the investigation of the neutron-induced (n, γ) , and (n, f) reaction cross sections, by means of an appropriate stable beam and target combination ([2, 3] and Refs. therein).

2. Deuteron surrogate reactions for neutron capture

The "desired" (n, γ) cross section for a target nucleus *A* is given in the Hauser-Feshbach formalism, in terms of compound nucleus (CN) formation cross section $\sigma_n^{CN}(E_{ex}, J, \pi)$ and the branching ratio $G_{\gamma}^{CN}(E_{ex}, J, \pi)$ toward the desired outgoing channel of γ -ray decay, by [4]:

$$\sigma_{n\gamma}(E_n) = \Sigma_{J,\pi} \,\sigma_n^{CN}(E_{ex}, J, \pi) G_{\gamma}^{CN}(E_{ex}, J, \pi), \tag{1}$$

where J, π are the spin and parity of the excited state E_{ex} of the decaying compound nucleus:

$$E_{ex} = \frac{A}{A+1}E_n + S_n,\tag{2}$$

 E_n being the neutron incident energy, and S_n the binding energy of the neutron in the compound nucleus.

Usually the compound nucleus formation cross section is obtained from a neutron optical model potential, while the G_{γ}^{CN} branching ratio requires accurate information on the Hauser-Feshbach model ingredients of the all competing decay channels, e.g., optical potentials, level densities, strengths functions, etc. Such difficulties should be avoided by using alternative surrogate reactions. Among them, the deuteron surrogate reaction $(d, p\gamma)$ produces the same excited nucleus (A + 1), decaying through the desired γ channel.

The probability for the compound nucleus formed in the (d, p) surrogate reaction, with the same specific excitation energy, spin, and parity values as in the desired reaction, to decay through γ channel is [4]:

$$P_{d,p\gamma}(E_{ex}) = \sum_{J,\pi} F_{d,p}^{CN}(E_{ex}, J, \pi) G_{\gamma}^{CN}(E_{ex}, J, \pi),$$
(3)

where $F_{d,p}^{CN}(E_{ex}, J, \pi)$ is the probability for the formation of this excited surrogate compound nucleus.

The specific feature of the surrogate method is the experimental determination of $P_{d,p\gamma}(E_{ex})$, by measuring the total number of the surrogate events, e.g. number of (d, p) processes, and the number of coincidences surrogate ejectile–CN decay channel, e.g. number of $p - \gamma$ coincidences:

$$P_{d,p\gamma}^{exp}(E_{ex}) = \frac{N_{p,\gamma}^{coincidences}(E_{ex})}{N_{d,p}^{surrogateevents}(E_{ex})}$$
(4)

corrected for the detector efficiency.

Further, the use of measured $P_{d,p\gamma}(E_{ex})$, together with the calculated $F_{d,p}^{CN}(E_{ex},J,\pi)$, to determine the branching ratios $G_{\gamma}^{CN}(E_{ex},J,\pi)$, leads to the determination of the desired cross section, Eq. (1). This is the theoretical frame of the surrogate reactions, before approximations which simplify the analysis.

A first approximation of the surrogate method considers similar $J - \pi$ distributions in both desired and surrogate reactions [2, 3]:

$$F_{d,p}^{CN}(E_{ex},J,\pi) \approx F_n^{CN}(E_{ex},J,\pi) \equiv \frac{\sigma_n^{CN}(E_{ex},J,\pi)}{\sum_{J',\pi'} \sigma_n^{CN}(E_{ex},J',\pi')},$$
(5)

where $F_n^{CN}(E_{ex}, J, \pi)$ is the probability for the formation of this excited compound nucleus in the desired reaction.

Next approximation, within the Weisskopf-Ewing (WE) limit of the Hauser-Feshbach formalism, considers the decay probabilities $G_{\gamma}(E_{ex}, J, \pi)$ to be independent of $J - \pi$:

$$G_{\gamma}(E_{ex}, J, \pi) = G_{\gamma}(E_{ex}), \tag{6}$$

the desired cross section becoming finally:

$$\sigma_{n\gamma}^{WE}(E_n) = \sigma_n^{CN}(E_n) P_{d,p\gamma}^{exp}(E_{ex}).$$
⁽⁷⁾

3. Tests of deuteron surrogate reaction approximations

Given the importance of (n, γ) reaction for basic and applied nuclear physics and the possibility of using $(d, p\gamma)$ as a surrogate reaction for neutron capture, the validation of the deuteron surrogate method has got a great importance. The validation test comparing already well known (n, γ) cross sections with those provided by deuteron surrogate reaction $(d, p\gamma)$ stressed out large discrepancies [5, 6, 7, 8, 9] which rise a strong question mark concerning the suitability of the associated theoretical frame.

Thus, Allmond *et al.* [7] reported a 23% deviation between the known ratio ${}^{235}\text{U}(n,\gamma)/{}^{235}\text{U}(n,f)$ [11] and the measured surrogate ratio ${}^{235}\text{U}(d,p\gamma)/{}^{235}\text{U}(d,pf)$. Such large discrepancy reveals "breakdown of the Bohr compound nucleus and Weisskopf-Ewing approximation" [7], requesting an improved reaction model for the (d, p) surrogate reaction. The same request of an improved reaction model for the (d, p) surrogate process results from Hatarik *et al.* [8] validation test for the ${}^{171,173}\text{Yb}(d,p\gamma)$ surrogate reactions by comparison with known neutron capture cross sections [10]. The large discrepancy between ENDF/B-VII.0 [11] evaluated ${}^{92}\text{Mo}(n,\gamma)$ reaction cross sections and the corresponding ${}^{92}\text{Mo}(d,p\gamma)$ surrogate cross sections found by Goldblum *et al.* [9] points out the failure of the modeling the deuteron surrogate reactions through the Weisskopf-Ewing approximation. Wilson *et al.* [5] directed a "stringent test of the applicability" of the deuteron surrogate method in the actinides region, for the ${}^{232}\text{Th}$ target nucleus, for well known neutron-capture cross sections [12]. Large overestimation of the (n, γ) reaction cross sections by the $(d, p\gamma)$ surrogate reaction results for the low neutron energy



Fig. 1: Comparison of experimental [17] total proton–emission breakup fraction and the corresponding parametrization of Refs. [15, 19, 20] (solid curves), and [18] (dashed curves) for deuteron interactions with target nuclei from 27 Al to 232 Th.

range $E_n < 1$ MeV has thus been reported. Supplementary to the criticism of the Weisskopf-Ewing approximation used in the surrogate formalism, the effect of the breakup process is mentioned by Ducasse *et al.* [6] as another source of validation failure in the case of deuteron surrogate reaction $^{238}U(d, p\gamma)$.

It is obvious that the apparent discrepancies evidenced by validation tests [5, 6, 7, 8, 9] are the results of weak points, actually the approximations of the (d, p) interaction process analysis in the theoretical frame of surrogate method.

One approximation, appearing even as a contradiction in the terms of the surrogate reaction method is that the direct nucleon-transfer (d, p) reaction forms an excited compound nucleus [5, 6, 7, 8, 9]. Therefore, a reconsideration of the reaction mechanisms involved in deuteron surrogate reactions, populating a highly excited nucleus, should include the direct reactions (DR, e.g., stripping), statistical processes, e.g., pre-equilibrium emission (PE), and CN processes, as well as the deuteron breakup (BU) particularly for deuteron interaction processes [13, 14, 15]. Actually, in the case of the deuteron surrogate reactions at low incident energies, for heavy targets nuclei (actinides) the deuteron breakup has the strongest effects, as it has been pointed out for the low-energy deuteron interaction with ²³¹Pa target nucleus [16].

The physical picture of the deuteron breakup in the Coulomb and nuclear fields of the target nucleus considers two distinct processes, namely the elastic breakup (EB) in which the target nucleus remains in its ground state and none of the deuteron constituents interacts with it, and the inelastic breakup or breakup fusion (BF), where one of these deuteron constituents interacts with the target nucleus while the remaining one is detected. Overall, there are actually two opposite effects of the deuteron breakup on the deuteron activation cross sections that should be considered. Firstly, the total-reaction cross section, that is shared among different outgoing channels, is reduced by the value of the total breakup cross section σ_{BU} . On the other hand, the BF component, where one of deuteron constituents interacts with the target nucleus leading to a secondary composite nucleus, brings contributions to different reaction channels [13, 14, 15, 16]. Thus, the absorbed proton or neutron following the deuteron breakup, contributes to the enhancement of the corresponding (d, xn) or (d, xp) reaction cross sections, respec-



Fig. 2: The energy dependence of the total breakup cross sections given by Avrigeanu *et al.* [15, 19, 20] (solid curves) and Kalbach [18] (dashed curves) parametrizations for deuteron interactions with ²⁷Al, ⁶³Cu, ⁹³Nb, ¹¹⁹Sn, ¹⁸¹Ta, and ²³¹Pa, while σ_R is shown by dotted curves.

tively. The breakup effects which are present in the deuteron surrogate experiments will be stressed out in the following through a detailed examination of the work of Wilson *et al.* [5] using the surrogate reaction 232 Th $(d, p\gamma)^{233}$ Th for an indirect measurement of the well-known 232 Th $(n, \gamma)^{233}$ Th reaction cross sections [11, 12] for incident-neutron energies between 0 and around 1 MeV. A good agreement between indirect and direct (n, γ) cross-section measurements was found only in the range 500 keV–1 MeV while large discrepancies have been present outside this range.

First, the measurement of Wilson *et al.* [5] has involved 12 MeV deuterons incident on ²³²Th target nucleus, while the decay probabilities $P_{d,p\gamma}(E_{ex})$ of the excited nucleus ²³³Th have been measured at excitation energies between the corresponding neutron binding energy S_n =4.786 MeV and 1 MeV above it. The protons from the $(d, p\gamma)$ reaction corresponding to this excitation have had energies between ~8.7 and 9.7 MeV while their maximum energy has been around 14.5 MeV. On the other hand, the BF protons have had a maximum energy of 9.673 MeV in the center-of-mass system, with a twofold outcome for these BF protons: they match the proton emission involved in the surrogate-reaction analysis, but have energies lower than the protons which populate the excited nucleus ²³³Th below S_n and were considered to prove the lack of any BU effect (Fig. 6 of Ref. [5]).

Second, the BF protons with energies between ~8.7 and 9.7 MeV correspond to BF neutrons with energies between around 1 MeV and 0, respectively, i.e. very much alike to the desired neutron capture process. Furthermore, these BF neutrons interact with ²³²Th target nucleus, populating the same analyzed ²³³Th compound nucleus, at the same excitation energies of interest. The γ -ray decay of ²³³Th compound nuclei populated through the BF enhancement contribute thus, together with the companion BF protons, to the measured $p - \gamma$ coincidence events.

Third, in addition to the BF, stripping, and PE contributions to the population of the excited nucleus ²³³Th, one has to take into account the considerable amount of incident deuterons leakage through the above mentioned processes [13, 14, 15, 16], strongly diminishing the probability $F_{d,p}^{CN}(E_{ex}, J, \pi)$ for forming the compound nucleus ²³³Th, Eq. (3).

The importance of the total (EB+BF) proton-emission breakup fractions σ_{RU}^p/σ_R , where σ_R is

the deuteron reaction cross section, is given in Fig. 1 by means of the comparison with the experimental systematics [17], measured for target nuclei from Al to Th. The calculated curves represent the predictions of the empirical Kalbach's [18] and Avrigeanu *et al.* [15, 19, 20] parametrizations.

A comparison of the *total* breakup cross sections predicted by Kalbach [18], and Avrigeanu *et al.* [15, 19, 20] for deuterons interaction with target nuclei from Al to Pa and the total deuteron cross section is presented in Fig. 2. Regardless of the differences for incident energy lower than ~10 MeV, where Kalbach's parametrization [18] predicts too high values for the breakup cross sections, both parametrizations predict an increasing role of deuteron breakup with increased target-nucleus mass/charge, pointing out the dominance of the breakup mechanism at the deuteron incident energies below and around the Coulomb barrier of 231 Pa [16]. Actually, this conclusion is in line with the experimental total proton-emission BU fraction data for deuterons on 232 Th [17], Fig. 1. Particularly, the dominance of the breakup mechanism for the actinides nuclei at energy around Coulomb barrier should be considered in the case of (*d*, *x*) surrogate reactions analysis.

From Figs. 1 and 2 it is obvious that the neglection of the breakup mechanism strongly affects the validation test, being the main reason of its failure.

The other assumption concerning the equality of the branching ratios for the deuteron surrogate and the neutron-induced reactions should be considered with increased caution in the analysis due to the population and decay differences between the excited and compound nuclei, respectively, formed in surrogate and desired reactions [21, 6].

Finally, one should be more careful in assuming that the failure of the surrogate-method validation tests follows the use of the too weak Weisskopf-Ewing approximation [5, 6, 7, 8, 9]. Even the use of the Hauser-Feshbach formalism alone within deuteron-induced reactions analysis can not lead to expected good results in the absence of the unitary account of BU+DR+PE+CN reaction mechanisms involvement.

4. Conclusions

The present work has concerned a deeper analysis of the key role of the direct interaction, particularly the breakup mechanism in the deuteron surrogate reactions. The opposite effects of the breakup mechanism, namely the enhancement of the counted *protons* – γ coincidences as well as the decrease of the compound nucleus cross section due to initial deuteron flux leakage through breakup but also stripping and pre-equilibrium processes should explain the failure of the validation tests of the deuteron surrogate method particularly at low incident energies around the Coulomb barrier and on actinides target nuclei.

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