On deuteron interactions within surrogate reactions and nuclear level density studies

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Abstract. The consequences which the suitable consideration of direct interaction, namely the breakup, stripping and pick-up processes, may have for the analysis of the deuteron surrogate reactions as well as for studies of nuclear-level density models on the basis of nucleon spectra analysis in deuteron-induced reactions are discussed. An increased attention is paid to the deuteron breakup which is the dominant mechanism for the interaction processes at low incident energies around Coulomb barrier, particularly on actinides.

1. Introduction

The description of deuteron-nucleus interaction represents an important test for both the appropriateness of reaction mechanism models and evaluation of nuclear data requested especially by the ITER [1], IFMIF [2], and SPIRAL-2 [3] research programmes. The present work continues the series of recent deuteron-induced reaction analyses [4, 5, 6, 7, 8] pointing out the major effects of the direct interaction (DI) contributions responsible for the contradictory results reported so far in testing various approaches of deuteron surrogate reactions and the nuclear level density.

2. Neutron capture analysis by deuteron surrogate reactions

The surrogate reaction method is an indirect measurement technique used to overcome the difficult problems of preparing and handling the highly radioactive targets required for cross-section measurements. Its main point concerns the study the outgoing channel of interest by means of an alternative "surrogate" reaction that involves a projectile-target combination more accessible experimentally (e.g., [9, 10, 11, 12, 13, 14, 15]). A particular feature is that, since last decade, this method has been involved mainly in the investigation of the neutron-induced \((n, \gamma)\) and \((n, f)\) reaction cross sections, by using an appropriate stable beam and target combination ([9] and Refs. therein).

The "desired" \((n, \gamma)\) cross section for a target nucleus \(A\) is given within 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 \(\gamma\)-ray decay, by [10]:

\[
\sigma_{n\gamma}(E_{n}) = \sum J, \pi \sigma_{n}^{CN}(E_{ex}, J, \pi)G_{\gamma}^{CN}(E_{ex}, J, \pi),
\]

(1)

where \(E_{n}\) is the neutron incident energy, and \(J, \pi\) are the spin and parity of the excited state \(E_{ex}\) of the decaying compound nucleus \((A+1)\).
CN formation cross section is usually obtained on the basis of a neutron optical model potential, while the \( G^{CN} \) branching ratio requires accurate information on the Hauser-Feshbach model ingredients of the all competing decay channels. 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 \( \gamma \) channel.

The probability for CN 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 the \( \gamma \) channel is [10]:

\[
P_{d,p\gamma}(E_{ex}) = \Sigma_{J,\pi} F^{CN}_{d,p}(E_{ex}, J, \pi) G^{CN}_{\gamma}(E_{ex}, J, \pi),
\]

where \( F^{CN}_{d,p}(E_{ex}, J, \pi) \) is the probability for the formation of this excited surrogate CN \((A+1)\).

A specific point of the surrogate method is the experimental determination of \( P_{d,p\gamma}(E_{ex}) \), by measuring the total number of the \((d, p)\) processes, and the number of \( p - \gamma \) coincidences:

\[
P^{exp}_{d,p\gamma}(E_{ex}) = \frac{N_{\text{coincidences}}(E_{ex})}{N_{\text{surrogate events}}(E_{ex})}.
\]

The use of the measured \( P_{d,p\gamma}(E_{ex}) \), together with the calculated \( F^{CN}_{d,p}(E_{ex}, J, \pi) \), determines the branching ratios \( G^{CN}_{\gamma}(E_{ex}, J, \pi) \), leading to the determination of the desired cross section given by Eq. (1). Actually this is the theoretical frame of the surrogate reactions, before to apply approximations which simplify the analysis.

First, an approximation of the surrogate method considers similar \( J - \pi \) distributions in both desired and surrogate reactions [9]:

\[
F^{CN}_{d,p}(E_{ex}, J, \pi) \sim F^{CN}_{n}(E_{ex}, J, \pi) \equiv \frac{\sigma^{CN}_{n}(E_{ex}, J, \pi)}{\Sigma_{J',\pi',\pi}^{E^{CN}_{\gamma}}(E_{ex}, J', \pi')},
\]

where \( F^{CN}_{n}(E_{ex}, J, \pi) \) is the probability for the formation of the CN in the desired reaction.

Second, another approximation is related to the Weisskopf-Ewing (WE) limit of the Hauser-Feshbach formalism and considers the decay probabilities to be independent of \( J - \pi \):

\[
G^{CN}_{\gamma}(E_{ex}, J, \pi) = G^{CN}_{\gamma}(E_{ex}),
\]

so that the desired cross section becomes finally:

\[
\sigma^{WE}_{n\gamma}(E_{n}) = \sigma^{CN}_{n}(E_{n}) P^{exp}_{d,p\gamma}(E_{ex}).
\]

3. Effects of the deuteron surrogate reaction approximations

The validation of the deuteron surrogate method has got a great importance due to the possibility of using \((d, p\gamma)\) as a surrogate reaction for neutron capture while the importance of \((n, \gamma)\) reaction for basic and applied nuclear physics is well known. On the other hand, large discrepancies have already been pointed out by the validation tests comparing well known \((n, \gamma)\) cross sections with those provided by deuteron surrogate reaction \((d, p\gamma)\) [11, 12, 13, 14, 15] so that strong question mark concerns the suitability of the associated theoretical frame.

In this respect Allmond et al. [13] reported a 23% deviation between the known ratio \( ^{235}\text{U}(n, \gamma)/^{235}\text{U}(n,\text{f}) \) and the measured surrogate ratio \( ^{235}\text{U}(d, p\gamma)/^{235}\text{U}(d,\text{pf}) \). This large discrepancy has revealed ”breakdown of the Bohr CN and Weisskopf-Ewing approximation” [13], requesting an improved reaction model for the \((d, p)\) surrogate reaction. A similar request for an improved reaction model of the \((d, p)\) surrogate process has followed the validation test performed by Hatarik et al. [14] for the \(^{171,173}\text{Yb}(d, p\gamma)\) surrogate reactions, by comparison with known neutron capture cross sections. The large discrepancy between evaluated \(^{92}\text{Mo}(n, \gamma)\)
Figure 1. Left-side: Comparison of experimental [16] total proton–emission breakup fraction and the corresponding parameterization of Refs. [7, 8] (solid curves), and [17] (dashed curves) for deuteron interactions with target nuclei from $^{27}$Al to $^{232}$Th. Right-side: The energy dependence of the total breakup cross sections given by Avrigeanu et al. [7, 8] (solid thin curve) and Kalbach [17] (dashed curve) parametrizations for deuteron interactions with $^{27}$Al, $^{63}$Cu, $^{93}$Nb, $^{119}$Sn, $^{181}$Ta, and $^{231}$Pa, while $\sigma_R$ is shown by solid thick curves.

reaction cross sections and the corresponding $^{92}$Mo($d,p\gamma$) surrogate cross sections found by Goldblum et al. [15] points out the failure of the modeling the deuteron surrogate reactions through the Weisskopf-Ewing approximation. Wilson et al. [11] performed a ”stringent test of the applicability” of the deuteron surrogate method for the well known $^{232}$Th($n,\gamma$) cross sections. A large overestimation of the ($n,\gamma$) reaction cross sections by the ($d,p\gamma$) surrogate reaction results for the low neutron energy range has thus been reported. In addition 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. [12] as another source of the validation failure in the case of $^{238}$U($d,p\gamma$) reaction.

Actually, the apparent discrepancies evidenced by validation tests [11, 12, 13, 14, 15] are obviously the results of weak points corresponding to the approximations of the ($d,p$) interaction process analysis in the theoretical frame of the surrogate method. One approximation, being even a contradiction in the terms of the surrogate reaction method, concerns the fact that the direct nucleon-transfer ($d,p$) reaction forms an excited CN [11, 12, 13, 14, 15]. Therefore, an advanced assessment 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 [4, 5, 8]. As a matter of fact, BU has the strongest effects in the case of the deuteron surrogate reactions at low incident energies, for heavy targets nuclei (actinides), as it has been pointed out for the low-energy deuteron interaction with $^{231}$Pa target nucleus [6].

The deuteron breakup in the Coulomb and nuclear fields of the target nucleus involves 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. Actually, there are overall two opposite effects of BU on the deuteron activation cross sections that should be considered. First, the deuteron total-reaction cross section $\sigma_R$, that is shared among different outgoing channels, is reduced by the value of the total breakup cross section $\sigma_{BU}$. At the same time, 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 [4, 5, 6, 7, 8].

A detailed discussion of the work of Wilson et al. [11], using the surrogate reaction $^{232}\text{Th}(d, p\gamma)^{233}\text{Th}$ for an indirect measurement of the well-known $^{232}\text{Th}(n, \gamma)^{233}\text{Th}$ reaction cross sections for incident-neutron energies between 0 and around 1 MeV, is involved hereafter to underline the breakup effects which are present in the deuteron surrogate experiments. Wilson et al. reported a good agreement between indirect and direct $(n, \gamma)$ cross-section measurements only in the range 500 keV–1 MeV while large discrepancies have been found outside this range.

Firstly, this measurement [11] involved 12 MeV deuterons incident on $^{232}\text{Th}$ target nucleus, while the decay probabilities $P_{d,p\gamma}(E_{ex})$ of the excited nucleus $^{233}\text{Th}$ 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 which correspond to this excitation have had energies between $\sim$8.7 and 9.7 MeV while their maximum energy has been around 14.5 MeV. At the same time, 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 $^{233}\text{Th}$ below $S_n$ and were considered to prove the lack of any BU effect (Fig. 6 of Ref. [11]).

Secondly, the BF protons with energies between $\sim$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. Moreover, these BF neutrons interact with $^{232}\text{Th}$ target nucleus, populating the same analyzed the $^{233}\text{Th}$ CN, at the same excitation energies of interest. The $\gamma$-ray decay of the $^{233}\text{Th}$ compound nuclei populated through the BF enhancement contributes thus, together with the companion BF protons, to the measured $p-\gamma$ coincidence events.

Thirdly, in addition to the BF, stripping, and PE contributions to the population of the excited nucleus $^{233}\text{Th}$, one has to take into account the considerable amount of incident deuterons leakage through these processes [4, 5, 6, 7, 8], which strongly diminishes the probability $F_{d,p}^{CN}(E_{ex}, J, \pi)$ for forming the compound nucleus $^{233}\text{Th}$, given by Eq. (2). The importance of the total (EB+BF) proton-emission breakup fractions $\sigma_{BU}/\sigma_R$ is shown in the left-side of Fig. 1 by means of the comparison of experimental systematics [16], measured for target nuclei from Al to Th, and the calculated predictions of the empirical Kalbach’s [17] and Avrigeanu et al. [7, 8] parametrizations.

The total breakup cross sections predicted by Kalbach [17] and Avrigeanu et al. [7, 8] for deuterons interaction with target nuclei from Al to Pa and the total deuteron cross section are compared in the right-side of Fig. 1. Except the differences for incident energies lower than $\sim$10 MeV, where Kalbach’s parametrization [17] predicts too high values for the breakup cross sections, both parametrizations predict an increasing role of BU 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 for the target nucleus $^{231}\text{Pa}$ [6]. This conclusion is obviously in line with the experimental total proton-emission BU fraction data for deuterons on $^{232}\text{Th}$ [16] shown on the left-side of Fig. 1. Therefore, the dominance of BU for the actinides nuclei at energy around Coulomb barrier should be particularly considered in the case of $(d, x)$ surrogate reactions analysis [8]. Actually, it is obvious that the neglect of the breakup mechanism (Fig. 1) strongly affects the validation test, being the main reason of its
failure.

Moreover, 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 [12, 18]. 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 [11, 12, 13, 14, 15]. It is also even the use of the Hauser-Feshbach formalism alone, within deuteron-induced reactions analysis, which can not lead to expected good results in the absence of the unitary account of the involvement of all BU+DR+PE+CN reaction mechanisms [8].

4. Nuclear level density study with deuteron-induced reactions

The results rather contradictory which are obtained from the investigation of the level density models in deuteron induced reactions at low energies, around the Coulomb barrier, stand for another example of an approach of the deuteron interaction process which is too simplified.

Thus, we would like to comment the assumption of lower noncompound components of the neutron spectra measured at backward angles with 5–9 MeV deuterons on 27Al, 63,65Cu, 89Y [19], and 54,56,58Fe [20] as well as the experimental CN fractions of σR obtained by too simplified analysis of the neutron angular distributions resulted from the deuteron interactions with the same target nuclei. Hence, there have been obtained contradictory results as (i) finding that more than 70% of σR are determined by the CN mechanism, and (ii) failure to describe the proton, and alpha-emission spectra.

Conversely, we compared the experimental CN fractions from the above-mentioned studies [19, 20] with the results of the unitary analysis of deuteron interactions with the same target nuclei 27Al, 63,65Cu, 89Y, and 54,56,58Fe [4, 5, 7] and found apparent discrepancies in both the numerical values and the slope of their energy dependence [21]. While Refs. [4, 5, 7] include a detailed description of the model assumptions and consistent parameters sets we would mention here only the main lines of these analyses concerning the treatment of nuclear reaction mechanisms involved within the deuteron interaction process. Thus, in order to obtain the CN fractions of σR it has been taken into account the decrease of the deuteron total-reaction cross section, that is subsequently shared among different outgoing PE and CN channels, by DI through appropriate treatment of each one. The BU cross sections have been obtained from the empirical parametrization [7, 8] of both the EB, and the total BU including also the BF, while the contributions of (d, p) and (d, n) stripping and (d, t) and (d, α) pick-up direct reactions were calculated using the distorted-wave Born approximation (DWBA) method [4, 5, 7].

The unitary and consistent consideration of the BU, DR, PE, and CN processes has been proved successful by the agreement of the calculated and all available deuteron activation data for 27Al, 54,56–58, nat Fe, 63,65,nat Cu, and 93Nb target nuclei [4, 5, 7]. The calculated CN fractions have been thus validated.

In what follows, we enlarge our previous analysis [21] by comparison of the ratios of CN cross sections to σR with the similar values obtained by analysis of measured neutron angular distributions in deuteron-induced reactions on 54,56,58Fe [20] (upper left-side of Fig. 2). The similar fractions for the DI and PE components are also included in this figure in order to point out the need of their consideration. Moreover, the corresponding excitation functions for each DI process along a wider deuteron energy range are also shown in Fig. 2 for a better understanding of their importance [5]. There are two main features pointed out, on the whole, by the comparison shown in the left-side of Fig. 2. First, there is a significant difference between the larger CN contributions obtained from the measured neutron angular distributions [20], and the calculated values following the unitary analysis of every deuteron-interaction mechanism. The lower values of the calculated CN fractions are mainly due to the significant BU component,
which has been not taken into account by any of the analyses performed within Refs. [19, 20].

Second, a particular note should concern the opposite energy dependence of the experimental and calculated \(CN\) fractions for low deuteron incident energies. Contrary to the expectation of Ramirez et al. [19, 20] of a decreasing \(CN\) contribution with the deuteron-energy increase, this contribution is slightly increasing with energy since both the major \(BU\) and especially the \(DR\) component have a slower increase with energy than \(\sigma_R\). The energy dependence of the \(DI\) fractions of \(\sigma_R\) for \(^{54,56,58}\)Fe target nuclei is better understood by analyzing the behavior of the excitation functions for competing \(BU\) and \(DR\) processes through an enlarged incident energy domain, as shown in the lower left-side of Fig. 2. Most significant in this respect are the maxima of the \((d,p)\) and \((d,n)\) stripping excitation functions around 6–8 MeV, after which lead to the statistical emission increase. The energy dependence of the \(DI\) fractions of \(\sigma_R\) found for Fe isotopes [5] is also confirmed by the similar analysis of the nuclear reaction mechanisms involved in the deuteron interaction with Ni isotopes shown in the right-side of Fig. 2.
5. Conclusions

The present work has concerned a deeper analysis of the key role of the direct interaction, particularly of the breakup mechanism, in deuteron-induced reactions, whose neglect led to above-mentioned contradictory results. The opposite effects of the breakup mechanism, namely the enhancement of the counted protons $\gamma$ coincidences as well as the decrease of the CN cross section due to initial deuteron-flux leakage through breakup but also DR and PE processes may explain [7, 8, 21] the failure of the validation tests of the surrogate method using deuteron-induced reactions.

Similarly, there are significant differences between the CN components established by analysis of measured neutron angular distributions, in deuteron-induced reactions around the Coulomb barrier [19, 20], and the results following the unitary analysis of the deuteron interaction mechanisms [4, 5, 7, 21]. Actually, the latter may account for the failure to describe the neutron spectra from 7.5 MeV deuteron interaction with $^{63,65}$Cu, or proton spectra from 6 MeV deuterons on $^{89}$Y [19] as well as protons and alpha spectra for 9 MeV deuteron on $^{56,58}$Fe [20].

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