Strengths and limitations of the surrogate reaction method to access neutron-induced cross sections of actinides

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Abstract

Gamma decay and fission probabilities of ^{237,239}U and ^{238,239}Np have been measured, for the first time simultaneously, via the surrogate reactions ^{238}U(^{3}He,^{4}He), ^{238}U(d,p), ^{238}U(^{3}He,t) and ^{238}U(^{3}He,d), respectively. While a good agreement between our data and neutron-induced data is found for fission probabilities, gamma decay probabilities are several times higher than the corresponding neutron-induced data for each studied nucleus. We study the role of the different spin distributions populated in the surrogate and neutron-induced reactions. The compound nucleus spin distribution populated in the surrogate reaction is extracted from the measured gamma-decay probabilities, and used as input parameter in the statistical model to predict fission probabilities to be compared to our data. A strong disagreement between our data and the prediction is obtained. Preliminary results from an additional dedicated experiment confirm the observed discrepancies, indicating the need of a better understanding of the formation and decay processes of the compound nucleus.

1. Introduction

Neutron-induced reaction cross sections of short-lived nuclei are crucial for fundamental nuclear physics, as well as for astrophysics and nuclear energy applications. In particular these data are important for understanding the nucleosynthesis r- and s- processes, and for nuclear waste transmutation via fast neutrons. However many of the key isotopes have often lives-time too short for producing and handling a target, making the direct measurement of these cross sections very difficult.

The surrogate reaction method, proposed for the first time in the 70's [3], is an indirect method which aims at determining compound nucleus reaction cross sections involving short lived and/or difficult-to-produce targets. The method is based on the assumption of the independence of the compound nucleus decay probability in a given channel on the formation channel (Bohr hypothesis): the same compound nucleus A* formed in a neutron-induced reaction (n+(A-1)$\rightarrow$A*) is now formed in a transfer reaction on a slightly different (but more accessible) target nucleus (b+Y$\rightarrow$A*+c). In this case the identification of the ejectile c allows one to determine the charge and mass (Z,A) of the decaying nucleus A, and the ejectile kinetic energy and emission angle provide its excitation energy E^*. The nucleus A can decay through different exit channels: fission, gamma emission, neutron emission, etc.

Therefore the measurement of the number of coincidences between the ejectile and the decay products of interest, normalized to the total number of detected ejectiles (i.e. to the total number of nuclei A produced) allows one to extract the decay probability P^{A}_{decay} for the corresponding decay channel. The neutron-induced cross section for the nucleus A-1 can then be obtained as

$$\sigma^{A-1}_{decay} \simeq \sigma^{A}_{CN}(E_n) P^{A}_{decay}(E^*) \tag{1}$$

where $\sigma^{A}_{CN}(E_n)$ is the compound nucleus formation cross section via the (A-1)+n reaction and it is typically obtained by optical model calculations.

The main advantages of this method are that it allows to access short-living nuclei, not otherwise accessible via direct measurements, and that several transfer channels can be simultaneously investigated on a broad excitation energy range. In addition this kind of measurements is performed with charged particles, whose beam intensity can be few orders of magnitude higher than the nowadays available neutron beams, and that the measurement is performed in a neutron-free environment, eliminating the issues related to neutron scattering typically associated to direct measurements. However, the equivalence of neutron-induced and surrogate reaction measurements relies on two hypothesis, which need to be tested. First, the formation of a compound nucleus must take place both in the neutron-induced and in the transfer reactions. This means that the formed nucleus loses memory of the entrance channel (except for the conserved quantities, i.e. energy and J^{π}) and its decay is independent of its formation. This assumption is reasonable in the excitation energy region close to and above the neutron separation energy, where the nuclear level density is high. The second one is that the decay probabilities of the compound nucleus are independent of its angular momentum and parity distributions -J^{π} - (the so-called Weisskopf-Ewing limit, see Ref. [4]), or that the J^{π} distributions populated in neutron-induced and transfer reactions are the same. Further details on the method and on the underlying assumptions can be found in Ref.[5].

Several measurements (e.g. Ref.[6]) showed a very good agreement of the fission cross sections obtained with the surrogate and direct methods for actinides. However, in recent experiments [7, 8] radiative capture cross sections on rare earths obtained in surrogate reactions were found to be higher up to a factor of 10 than the corresponding neutron-induced reaction data. These important discrepancies were attributed to the large differences in the angular momentum between the mother and the daughter nuclei around the neutron separation energy, which results in the suppression of the neutron emission channel and therefore in the increase of the gamma emission probability [8]. This effect is expected to be reduced when studying actinides, whose level density is much higher than the rare earth one even at low excitation energies. However, a simultaneous measurement of fission and gamma emission probabilities of actinides was not performed up to now. In this work we report the results of this first-time experiment with the aim of further investigate the validity of the assumption of the surrogate reaction method and therefore to pin down to which extent it can be applied to infer neutron-induced cross sections.

2. Experiment

The measurement was performed at the Oslo cyclotron. A deuteron and a ^{3}He beams at 15 and 24MeV energy, respectively, were impinged on a 99.5% isotopically pure ^{238}U target of 260μg/cm^2 thickness. The target, deposited on a 40μg/cm^2 C layer, was produced at GSI and extreme attention was payed to reduce its oxidation.

The experimental setup coupled the CACTUS [9] NaI(Tl) array for gamma detection, the NIFF PPAC [10] for fission fragment detection and the SiRi silicon telescope array [11] for the ejectile detection and identification. The CACTUS array is constituted by 27 NaI(Tl) scintillators located 22 cm around the target. Its efficiency was determined with the Extrapolated Efficiency Method [12] and the

Pulse Height Weighting function Technique [13], which give results in good agreement up to several hundred of keV above the neutron separation energy. For more details see Ref. [14]. The NIFF array is constituted by 4 PPAC, filled with 5 mbar C_4H_{10} gas, covering from 12° to 63° polar angle around the target, with a geometrical detection efficiency of 40% of 2π [14]. Finally, the SiRi telescope array is constituted by two segmented ΔE(130μm)-E(1500μm) silicon detectors, covering from 126° to 140° polar angle around the beam direction. The silicon telescopes allow one to unambiguously identify the ejectile and measure its angle and kinetic energy. This information, combined to the reaction Q-value of the studied reaction, allows one to determine the excitation energy of the formed compound nucleus. The experimental decay probability of the nucleus A* in the channel j (fission or gamma emission) can be obtained as:

$$P_j(E^*) = \frac{N^j_{coinc}(E^*)}{N_{singles}(E^*)\varepsilon^j(E^*)} \tag{2}$$

where $N^j_{coinc}(E^*)$ is the number of ejectiles detected in coincidence with each decay channel product and $N_{singles}(E^*)$ is the total number of detected ejectiles, $\varepsilon^j(E^*)$ is the detection efficiency of the decay product.

3. Results

As mentioned, several nuclei can be accessed simultaneously during surrogate reaction measurements. In particular, in this experiment we measured the gamma decay and fission probabilities of ^{239}U via the ^{238}U(d,p) tranfer reaction and of ^{239}Np, ^{237}U and ^{238}Np via the ^{238}U(^{3}He,d) , ^{238}U(^{3}He,^{4}He) and ^{238}U(^{3}He,t) reactions, respectively. In a first moment we will focus on the (d,p) reaction channel (i.e. the decay of the excited ^{239}U), for which we have the highest statistics. However, the study of this reaction requires to account for the deuteron breakup, which is known since the 70's to modify the values of the measured fission cross section of about 50% [15]. Despite the difficulties associated to this correction, the (d,p) reaction channel is very relevant for the surrogate reaction method because it is the closest reaction to a neutron induced reaction.

In Fig.1a we present the fission probability obtained for this nucleus. The experimental data (full squares) are corrected for the deuteron breakup (empty square) [16] and compared to the evaluated neutron-induced data (full line) given by JENDL 4.0. For more details on elastic and inelastic deuteron breakup correction see Ref.[14]. The shown error bars account for both statistical and systematic uncertainties. The fission threshold is located around 6.2 MeV ^{239}U excitation energy. An agreement between the corrected data and the neutron-induced data is observed for the fission threshold and the cross section values above the threshold. Similar agreements were found when analysing the other transfer reactions, although with less statistics, for which the deuteron breakup correction was not necessary. In Fig. 1b the experimental gamma emission probability P_γ (full circles) of ^{239}U is shown. As expected the P_γ is equal to 1 below the neutron emission threshold of 4.8MeV (we remind that the gamma emission is the only open channel below S_n since the nucleus is not fissile, and the proton separation energy is bigger than the neutron separation energy) and it significantly drops above this energy due to the competition with the neutron emission. Our data are then compared to neutron-induced data (JENDL 4.0 - full line) and discrepancies up to a factor 10 are observed. In Fig. 1c we plot both the fission and gamma emission probabilities shown in Figs. 1a and 1b in the region where both decay channels are open simultaneously, and we compare the experimental data to the evaluated neutron induced data. Also in this excitation energy region we observe a good agreement with the neutron-induced data for the fission probability and a discrepancy of up to a factor 3 for the gamma emission probability. This seems to indicate that, while the fission process is independent of the neutron emission hindering, and therefore independent of the compound nucleus populated J^π distribution, it is not the case for the gamma emission, which is strongly enhanced by the neutron emission hindering. However, calculations based on the statistical

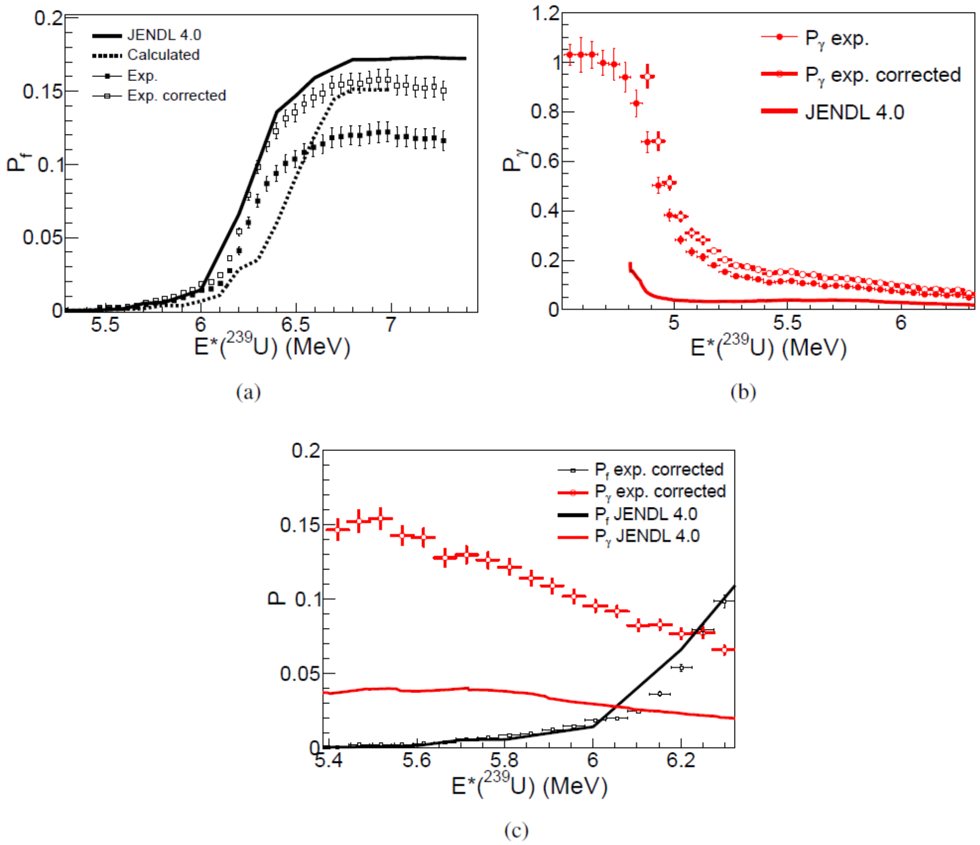

Fig. 1: ^{239}U fission P_f (*a*) and gamma emission P_γ (*b*) probabilities as a function of the compound nucleus excitation energy (E^*) obtained in the ^{238}U(d,p) reaction. Fig.*c* is a zoom of Figs. *a* and *b*, in the E^* region where the fission and gamma emission channels are both open.

model with standard ingredients show a strong dependence of the fission probability on the spin.

To further investigate it, we compare the measured fission probability to the one calculated by the statistical model. Following the procedure described in [8] we extracted direct information on the populated J^π distribution from the experimental gamma decay probabilities, using the TALYS code [17]. Assuming a Gaussian angular momentum distribution, with no dependence on the excitation energy, the experimental gamma emission probability can be written as:

$$P_\gamma(E^*) = \sum_{J^\pi} [\frac{1}{2\sigma\sqrt{2\pi}} e^{-\frac{(J-\bar{J})^2}{2\sigma^2}}] G_\gamma(E^*, J^\pi) \tag{3}$$

where $G_\gamma(E^*,J^\pi)$ are the TALYS gamma decay probability. The unknown $\bar{J}$ and σ parameters, which correspond to the average and width of the spin distribution, are obtained by fitting the experimental data with Eq.3 in the compound-nucleus excitation energy region around 6MeV. The mean value of the surrogate spin distribution is around $5\hbar$ and the width is $2\hbar$. These values are higher than those obtained for the neutron-induced spin distribution, which is centered around $1\hbar$ with a width of about $0.5\hbar$. The surrogate spin distribution is now used as input parameter to the statistical model TALYS to determine the fission probability. The so-calculated fission probability is plotted in Fig.1a as dashed line and compared to the experimental data. The calculated fission probability does reproduce neither the values nor the fission threshold obtained experimentally. In particular, we observe that the statistical model

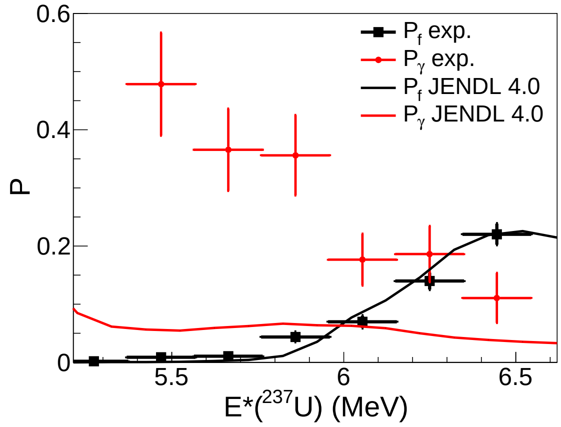

Fig. 2: ^{237}U fission P_f probability as a function of the compound nucleus excitation energy (E^*) obtained in the ^{238}U(^{3}He,^{4}He) reaction. (Oslo experiment)

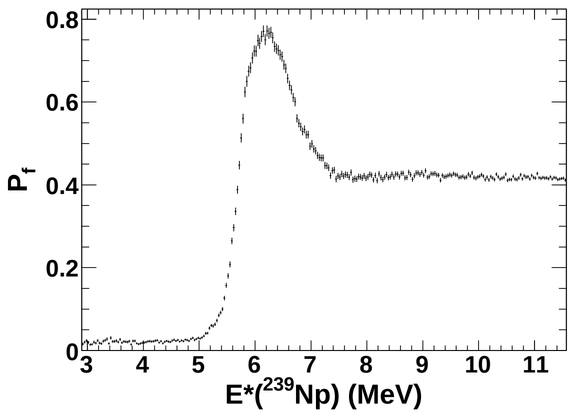

Fig. 3: ^{239}Np fission P_f probability as a function of the compound nucleus excitation energy (E^*) obtained in the ^{238U}U(^{3}He,d) reaction. (IPN Orsay experiment)

predicts a dependence of the fission threshold on the mean angular momentum of the compound nucleus, which increases as we increase the input mean spin of the compound nucleus. On the contrary, the agreement between the fission thresholds measured in surrogate and neutron-induced reactions observed experimentally indicates an independence of the fission probability of the compound nucleus angular momentum. Therefore, our experimental observations are not currently explained within a statistical model.

Similar results are obtained when studying the other transfer channels. Although the statistics in these channels is lower (and therefore the fission threshold can be determined with less precision), they are not affected by the projectile break-up and therefore a direct comparison between the surrogate and neutron-induced data is possible, free of theoretical corrections. The ^{238}U(^{3}He,^{4}He) reaction is of particular interest and the results obtained for this transfer channel are shown in Fig. 2. A new dedicated experiment was performed in April 2015 at the Orsay TANDEM accelerator, with the aim of studying this reaction (among others) with an increased statistics, an increased precision in the excitation energy and in the fission and gamma emission probabilities. The latter is obtained by segmenting the fission fragment detectors to have a measurement of the fission fragments anisotropy, which affects both the fission and gamma probability measurements (indeed fission gamma rays need to be subtracted from the gamma emission probability). The data analysis is currently ongoing. In fig.3 we present the fission probability of ^{239}Np compound nucleus, obtained in the ^{238}U(^{3}He,d) transfer reaction. Although the results are very preliminary, a good agreement between the results obtained in the two experiments below S_n and between surrogate and neutron-induced fission threshold value is found.

4. Conclusions

In summary, we have performed an experiment to study the validity of the surrogate reaction method to extract neutron-induced reaction cross sections. It is the first time that transfer-induced gamma emission and fission probabilities of actinides are simultaneously measured. The comparison of our experimental data to those obtained in neutron-induced reactions shows a good agreement for the fission probability and a strong disagreement for the gamma emission probability for the same compound nucleus and excitation energy. This indicates a strong sensitivity of the gamma emission to the compound nucleus populated spin distribution at excitation energies slightly above the neutron separation energy. Indeed it was previously shown that the spin distribution populated in surrogate reaction is centered at higher values and it is broader than the one populated in neutron-induced reactions [8]. On the contrary we do not observe a dependence of the fission probability on the populated angular momentum distribution of the compound nucleus. We have compared these observations to the statistical model predictions. We have determined the spin distribution from a fit to the measured gamma emission probabilities via a statistical model calculation performed with the TALYS code. The so-obtained spin distribution is used as input parameter to deduce the fission probability. Statistical model calculations predict an influence of the angular momentum on the fission threshold. Such a dependence is not observed in the experimental data. Preliminary results of a more dedicated experiment confirm the presented results. Therefore our observations are nowadays not explained within a statistical model picture. It is then crucial to better understand the formation and decay mechanisms of the compound nucleus in transfer reactions. Indeed, the surrogate reaction method allows one to access cross sections of short-lived nuclei, that cannot be directly measured.

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