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First simultaneous measurement of fission and gamma probabilities of ^{237}U and ^{239}Np via surrogate reactions

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Abstract. Fission and gamma decay probabilities of ^{237}U and ^{239}Np have been measured, for the first time simultaneously in dedicated experiments, via the surrogate reactions ^{238}U(^{3}He,^{4}He) 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 relevant for fundamental nuclear physics, as well as for astrophysics and nuclear energy applications. Indeed these data play an important role in the understanding of the nucleosynthesis r- and s- processes, and for nuclear waste transmutation via fast neutrons. However the lifetimes of many relevant isotopes are often too short

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for producing and/or handling a target, making the direct measurement of these cross sections very difficult or impossible.

The surrogate reaction method was proposed for the first time in the 70's by Cramer and Britt [1]. It 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 Bohr hypothesis: the compound nucleus decay probability in a given channel is independent of the formation channel. Therefore the same compound nucleus A* formed in a neutron-induced reaction (n+(A-1)→A*) can be formed in a transfer reaction on a slightly different (but more accessible) target nucleus (b+Y→A*+c). In this case, by identifying the ejectile c one can 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 method has the main advantages of allowing one to access short-lived nuclei, not otherwise accessible via direct measurements, and to simultaneously investigate several transfer channels 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 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 hypotheses, 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 justified by the high nuclear level density in the excitation energy region close to and above the neutron separation energy, which is the region of interest. 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. [2]), 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.[3].

Several measurements (e.g. Ref.[4]) showed a very good agreement of the fission cross sections obtained with the surrogate and direct methods for actinides. However, in recent experiments [5, 6] 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 hindering of the neutron emission channel and therefore in the increase of the gamma emission probability [6]. This effect is expected to be reduced when studying actinides, whose level densities are 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 two experiments where this simultaneous measurement was first performed with the aim of further investigating 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 Experiments

The first measurement was performed in 2012 at the Oslo cyclotron. A deuteron and a ^{3}He beams at 15 and 24 MeV energies, respectively, were impinged on a 99.5% isotopically pure ^{238}U target of 260 μg/cm^2 thickness, deposited on a 40 μg/cm^2 C layer. The experimental setup coupled the CACTUS [7] NaI(Tl) array for gamma detection, the NIFF PPAC [8] for fission fragment detection and the SiRi silicon telescope array [9] for the ejectile detection and identification. A detailed description of the setup and of the data analysis can be found in [10–12].

A new dedicated experiment was performed in April 2015 at the Orsay TANDEM accelerator, with the aim of obtaining increased statistics, an increased precision in the excitation energy and in the fission and gamma emission probabilities. A 24 MeV ^{3}He beam was impinged on a 99.5% isotopically pure ^{238}U target of 260μg/cm^2 thickness, deposited on a 39μg/cm^2 C baking. The targets were produced at GSI and extreme attention was payed to reduce their oxidation.

The detection setup coupled dedicated detectors for the detection of gamma rays, fission fragments and light ejectiles. Gamma rays were detected by 4 C_6D_6 scintillators, placed at 90° polar angle around the target, assuring the detection over a broad energy range (up to 10 MeV). Their efficiency was determined with the EXtrapolated Efficiency Method [13]. For more details see Ref. [10, 14]. Six Ge detectors were also placed around the target to perform precise intensity measurements of selected gamma transitions. The fission fragments detector was constituted by 8 segmented photovoltaic cells. Five cells were arranged in a pentagone and covered the polar angles from 19° to 75° and the whole azimutal angle. The other three cells were placed upstream of the target and cover from 105° to 150° polar angle. The segmentation of the photovoltaic cells allows one to experimentally measure the fission fragments anisotropy, which affects the estimation of the fission detection efficiency. This will allow us to reduce the uncertainty on both the gamma decay and fission probabilities with respect to the previous experiment. Finally the ejectiles were detected by two double-side stripped ΔE(300μm)-E(2mm) silicon detectors, covering the polar angles from 116° to 159°. The silicon telescopes allow one to unambiguously identify the ejectile and measure its angle and kinetic energy. This information, combined with the reaction Q-value of the studied reaction, allows one to determine the excitation energy of the formed compound nucleus with a resolution of $50 - 80$ keV. The experimental decay probability of the nucleus A* in the channel j (fission or gamma emission) can be obtained as:

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

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 mentionned, 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}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 (^{3}He,^{4}He) reaction channel (i.e. the decay of the excited ^{237}U), for which good quality neutron-induced cross section values are available.

In Fig.1a we present the preliminary fission probability obtained for this nucleus. The experimental data (crosses) are compared to results obtained in the Oslo experiment (squares) and to the evaluated neutron-induced data (full line) given by JENDL 4.0. The shown error bars account for the moment only for statistical uncertainties. The fission threshold is located around 6.1 MeV ^{237}U excitation energy. A very good agreement is found between the data obtained in the two experiments and the much higher statistics and excitation energy resolution of the Orsay experiment can be observed. An agreement between our experimental 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 slighty less statistics. In Fig. 1b the experimental gamma emission probability P_γ (full circles) of ^{237}U is shown. As expected the P_γ is equal to 1 below the neutron emission threshold of 5.1 MeV (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. Once more a good agreement with the data of the Oslo experiment is found. Our data are then compared to neutron-induced data (JENDL 4.0 - full line) and discrepancies a factor 3 at 6.4MeV 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 10 for the gamma emission probability. This seems to indicate that, while the fission probability 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 probability, which is strongly enhanced by the neutron emission hindering. However, calculations based on the statistical 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 [6] we extracted direct information on the populated J^π distribution from the experimental gamma decay probabilities, using the TALYS code [15]. Assuming a Gaussian angular momentum distribution, with no dependence on the excitation energy, the experimental gamma emission probability can be written as:

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

where $G_\gamma(E^*,J^\pi)$ are the TALYS gamma decay probability. The unkonwn $\bar{J}$ 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 $6\hbar$ 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 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 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 to the compound nucleus angular momentum. Therefore, our experimental observations are not currently explained within a statistical model with standard ingredients.

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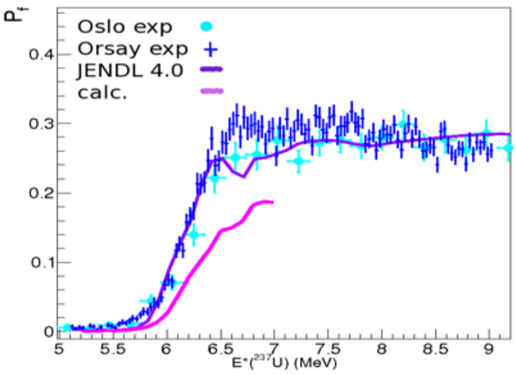
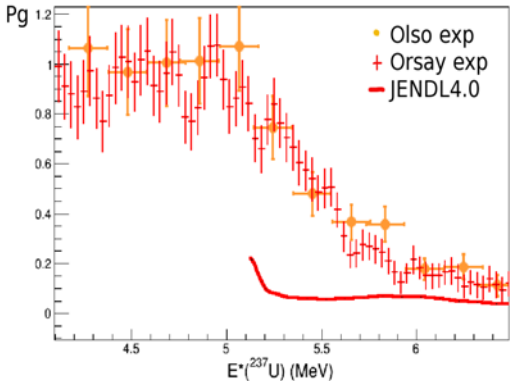


(a)



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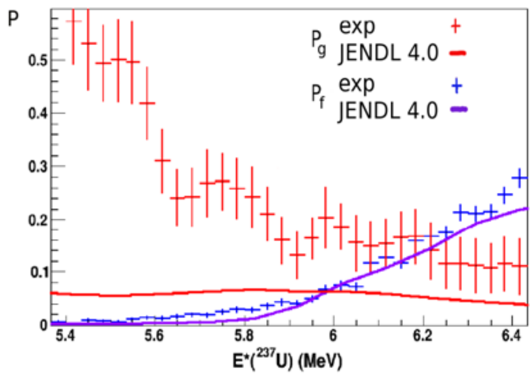

(c)

Figure 1: ^{237}U fission P$_f$ (*a*) and gamma emission P$_\gamma$ (*b*) probabilities as a function of the compound nucleus excitation energy (E*) obtained in the ^{238}U(^{3}He,^{4}He) 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.

Similar results are obtained when studying the other transfer channels. In fig.2 we present the fission and gamma probabilities of ^{239}Np compound nucleus, obtained in the ^{238}U(^{3}He,d) transfer reaction. This is an interesting nucleus since the fission barrier is lower than the neutron separation energy, S$_n$. Therefore both fission and gamma decay channels are open below S$_n$ and the sum of the corresponding decay probabilities should be 1. Below the fission barrier $P_\gamma = 1$ and it decreases around 5.5 MeV where the fission sets in. The fission probability increases up to an excitation energy of about 6.2 MeV, the S$_n$, and then it drops, due to the competition with the neutron emission. In fig 2 we also plot $P_f + P_\gamma$, whose good agreement with 1 below S$_n$ gives us confidence in the procedure used in the analysis.

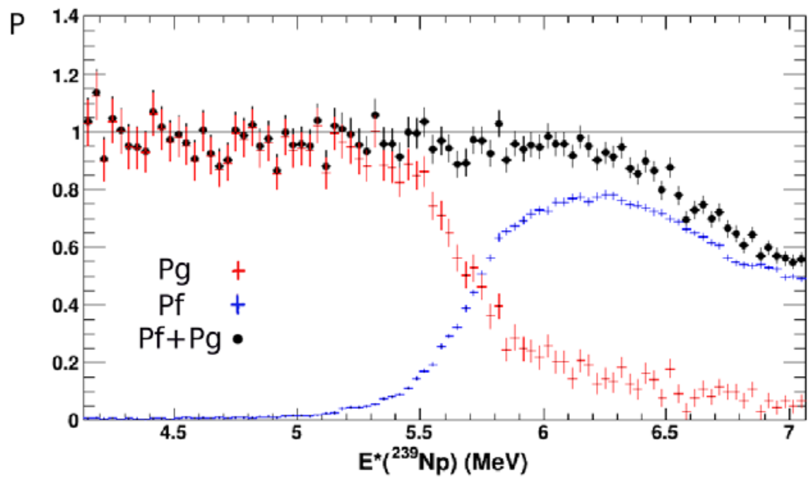

Figure 2: ^{239}Np P_f and P_γ probabilities as a function of the compound nucleus excitation energy (E^*) obtained in the ^{238}U(^{3}He,d) reaction and sum of P_f and P_γ.

4 Conclusions

In summary, we have performed two dedicated experiments 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 probability 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 [6]. 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. Therefore our observations are nowadays not explained within a statistical model picture with standard ingredients. 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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