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C. Plettner

H. Ai

C. W. Beausang
University of Richmond, cbeausan@richmond.edu

L. A. Bernstein

L. Ahle

See next page for additional authors

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Authors C. Plettner, H. Ai, C. W. Beausang, L. A. Bernstein, L. Ahle, H. Amro, M. Babilon, J. T. Burke, J. A. Caggiano, R. F. Casten, J. A. Church, J. R. Cooper, B. Crider, G. Gurdal, A. Heinz, E. A. McCutchan, K. Moody, J. A. Punyon, J. Qian, J. J. Ressler, A. Schiller, E. Williams, and W. Younes

Measuring reaction probability ratios to simulate neutron-induced cross-sections of short-lived nuclei

C Plettner¹, H Ai¹, C W Beausang^{1,2}, L A Bernstein³, L Ahle³, H Amro¹, M Babilon^{1,4}, J T Burke³, J A Caggiano¹, R F Casten¹, J A Church³, J R Cooper³, B Crider², G Gürdal^{1,5}, A Heinz¹, E A McCutchan¹, K Moody³, J A Punyon³, J Qian¹, J J Ressler¹, A Schiller⁶, E Williams¹ and W Younes³

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Abstract

Measuring the neutron-induced fission cross-sections of short-lived nuclei represents an experimental challenge due to target activity and the low intensity of neutron beams. One way to alleviate the problems inherent in the direct measurement is to use the surrogate method, where one measures the decay probability of the same compound nucleus formed using a charged beam and a stable target. The decay probability of the compound nucleus is then used to estimate the neutron-induced cross-section. As an extension to the surrogate method, we introduce a new method of reporting the fission probabilities of two compound nuclei as a ratio, which has the advantage of removing most of the systematic uncertainties. The ratio method was checked in a known case, the 236 U(n, f)/ 238 U(n, f) cross-section ratio, which turned out to be the same as the probability ratio of $P(^{236}$ U(d, pf))/ $P(^{238}$ U(d, pf)). As an application, the 237 U(n, f)/ 235 U(n, f) cross-section ratio was inferred, on the basis of the measured $P(^{238}$ U(d, d'f))/ $P(^{236}$ U(d, d'f)) probability ratio.

The neutron-induced fission cross-sections on various actinide nuclei have been deduced in the past by measuring the fission probabilities following the compound nucleus formation using a (t, pf) reaction [1–4] (see [5] for a re-interpretation). The fundamental idea behind this method is that both (n, f) and (t, pf) reactions can be separated into two sequential and independent steps: (i) a formation process, leading to the same compound nucleus, which equilibrates and (ii) a decay process, e.g. fission. Quantitatively, the (t, pf) reaction probability was measured to estimate the (n, f) reaction cross-section in [4], by

$$\sigma_{(n,f)}(E_n) = \sigma_{CN}(E_n) P_{(t,pf)}(E_x), \tag{1}$$

¹ Wright Nuclear Structure Laboratory, Yale University, New Haven, CT 06520, USA

² Department of Physics, Richmond University, Richmond, VA 23173, USA

³ Lawrence Livermore National Laboratory, Livermore, CA 94550, USA

⁴ Institut für Kernphysik, Technische Universität Darmstadt, D-64289, Germany

⁵ Clark University, Worcester, MA 01610, USA

⁶ MSU/NSCL, 1 Cyclotron Road, East Lansing, MI 48824, USA

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where $\sigma_{\rm CN}(E_n)$ is the compound nucleus formation cross-section as a function of incident neutron energy E_n , and $P_{(t,pf)}(E_x)$ is the fission probability following the (t,p) compound nucleus formation at an excitation energy $E_x = E_n + B_n$, with B_n the neutron binding energy. $\sigma_{\rm CN}(E_n)$ is obtained from an optical model calculation, while $P_{(t,pf)}(E_x)$ is the measured quantity. Equation (1) assumes high compound excitation energies $(E_x \sim 8 \text{ MeV})$, where many states overlap and the formation cross-section dependence on J^{π} disappears. It was shown in [5] that equation (1) cannot reproduce the data from the direct measurements at small excitation energies, unless a specific normalization prescription is applied and the angular momenta transferred by the (t,p) and neutron-induced reaction are taken into account.

The surrogate method is of great use for short-lived target nuclei, e.g. 237 U with a half-life of 6.7 days, where the direct measurement is difficult. However, the limitation of this approach is that the absolute fission probabilities must be determined, which relies on an accurate measurement of the number of the (t, p) events: $(P_{(t,pf)} = \frac{N_{(t,pf)}}{N_{(t,p)}})$. Due to target contamination, $N_{(t,p)}$ turns out to be the source of largest uncertainty. Instead, we propose to measure the fission probabilities for two similar compound nuclei in the same experiment, and report the results as a ratio where the $N_{(t,p)}$'s cancel out.

The experiment was performed by using deuterium beams of 24 and 32 MeV energy delivered by the ESTU tandem accelerator at Yale. The 236,238 U targets were prepared as nitrates of approximately $300~\mu g$ cm⁻² thickness, 'stippled' on a $200~\mu g$ cm⁻² carbon backing. The detection of charged particles emitted in the reaction was achieved by using the STARS detector (silicon telescope array for reaction studies) [6], which was configured as a $\Delta E-E$ telescope. The telescope consisted of three 'S2' type annular double-sided silicon (Si) detectors purchased from micron semiconductor. The thickness of the front detector (ΔE) was 140 μ m. The back detector (E) consisted of two Si detectors, E_1 and E_2 , of $1000~\mu$ m thickness each. The thickness of the back detectors was chosen such that the deuterons were fully stopped at a beam energy of 24 MeV. Each Si detector was segmented on one side into 48 rings and on the other side into 16 wedge-shaped segments. However, adjacent rings and sectors were electrically connected, so that for each detector a total of 24 rings and 8 sectors were recorded. The master trigger was generated whenever a sector or a ring in the back detector (E_1 or E_2) fired. The master trigger rate was typically 40 kHz, while the total rate in the ΔE detector was 20~kHz.

The (d, d'f) or (d, pf) events of interest are characterized by coincident deuteron (proton) and fission fragments. Flight path of the scattered particle through the telescope is traced back to the target to further remove random coincidences. The correct ring-ring and sector-sector correlation was determined from the known detector geometry and confirmed using the experimental data and Monte Carlo simulations of energy loss of protons and deuterons in Si detectors. $\Delta E - E$ matrices were created for each ring in the front detector which lies within the angular coverage of the back detector. An example of such a matrix is shown in figure 1. The various contributions of different charged particles are clearly visible on this plot. While the most energetic peak in the deuteron distribution is due to elastic scattering U(d, d'), the other peaks are due to reactions on light-ion target contaminants, such as carbon, nitrogen and oxygen. By setting polygonal gates around the various particle distributions, events associated with proton or deuteron emission could be selected.

Since the ΔE detector served for both light charged particle and fission fragment detection, the candidate fission events are identified by making use of the hit analysis and energy deposition in the ΔE detector. Hence, the selection of the (d,d'f) or (d,pf) subset of events was made from those events with (i) two distinguishable hits in the front detector (one hit corresponding to the detection of the light charged particle and the other one corresponding to

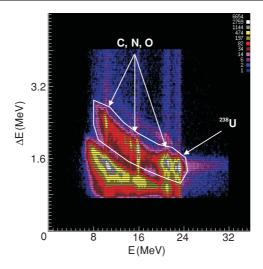


Figure 1. Particle identification ΔE –E spectrum for 24 MeV deuterons incident on a 238 U target with counts plotted on a logarithmic scale. The spectrum was recorded by ring no 3 in the ΔE detector, which is located at 37.4° with respect to the beam. The distribution inside the polygon corresponds to deuterons and the distribution below corresponds to protons.

(This figure is in colour only in the electronic version)

the detection of one of the fission fragments), and (ii) one hit in the back detector corresponding to the light charged particle. Spurious fission events originating from reactions on light contaminants in the targets were estimated by applying the same sort procedure to a target of ammonium nitrate (no fissionable nuclei), and they were consequently identified as particles with energy less than 14 MeV.

Once the number $N_{(d,pf)}$ ($N_{(d,d'f)}$) was established for both ^{236,238}U, a ratio of probabilities could be formed:

$$\frac{P_{(d,pf)}(^{238}\mathrm{U})}{P_{(d,pf)}(^{236}\mathrm{U})} = \frac{N_{(d,pf)}(^{238}\mathrm{U}) \times N_R(^{236}\mathrm{U}) \times W(^{236}\mathrm{U})(\theta)}{N_{(d,pf)}(^{236}\mathrm{U}) \times N_R(^{238}\mathrm{U}) \times W(^{238}\mathrm{U})(\theta)}.$$
 (2)

Here N_R is the number of Rutherford scattered deuterons which accounts for differences in target thickness and beam intensities for the $^{236}\mathrm{U}$ and $^{238}\mathrm{U}$ experiments, while $W(\theta)$ is a correction factor for the angular correlation between the outgoing proton and the fission fragment, which has been found to be 96% identical for $^{236}\mathrm{U}$ and $^{238}\mathrm{U}$ [7]. We note that, generally, the method of reporting ratios eliminates most of the systematic uncertainties, since the $N_{(d,p)}$ values are very similar for $^{236}\mathrm{U}$ and $^{238}\mathrm{U}$ and cancel out in the ratio.

We now consider the relationship of this ratio of probabilities to the corresponding neutron-induced fission cross-sections ratio. Based on equation (1), the neutron-induced cross-section can be expressed as follows:

$$\frac{\sigma_{(n,f)}(^{238}\mathrm{U})(E_x)}{\sigma_{(n,f)}(^{236}\mathrm{U})(E_x)} = \frac{P_{(d,pf)}(^{238}\mathrm{U})(E_x)}{P_{(d,pf)}(^{236}\mathrm{U})(E_x)},\tag{3}$$

because the compound nucleus formation cross-sections $\sigma_{\rm CN}$ are very similar for ²³⁸U and ²³⁶U, and cancel out in the ratio. (These compound nuclei have similar structure and the compound nucleus formation cross-sections scale as $A^{2/3}$ [8].) The ²³⁶U(d, pf) and ²³⁸U(d, pf) reactions serve as surrogates for the well-known ²³⁶U(n, f) and ²³⁸U(n, f), respectively, thus the corresponding ratio can be compared to direct measurements. In figure 2 the (d, pf) probability ratio is illustrated and compared to the direct (n, f) measurement data.

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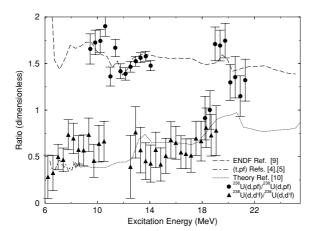


Figure 2. Fission probability ratios for the (d, pf) and (d, d'f) reactions on ²³⁸U and ²³⁶U targets, as a function of the excitation energy of the compound nucleus. The existing data and theoretical estimates are also indicated.

The surrogate ratio follows closely the reported direct measurement [9], giving confidence in the measurement and in the technique.

The $P_{(d,d'f)}$ ratio serves as a surrogate for the $^{237}\mathrm{U}(n,f)/^{235}\mathrm{U}(n,f)$ cross-section ratio and since the $^{235}\mathrm{U}(n,f)$ neutron-induced cross-section is known, the ratio can be used to deduce the unknown $^{237}\mathrm{U}(n,f)$ cross-section. In figure 2 these ratios are presented. The previous low energy surrogate results originating from the (t,pf) reaction [4, 5], also plotted in figure 2, show good agreement with the STARS data. Our new measurement extends the $^{237}\mathrm{U}(n,f)$ cross-section to much higher energies (\sim 14 MeV equivalent neutron energy). A theoretical estimate [10] based on an extrapolation of the surrogate data obtained previously is also indicated on the graph and the data match this prediction within 1σ accuracy. We note that this is the first measurement of the neutron-induced fission probability on $^{237}\mathrm{U}$ over this energy range.

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