EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Letter of Intent to the ISOLDE and Neutron Time-of-Flight Committee

Fission properties probed via measurements of transfer-induced fission with actinide beams in inverse kinematics using the ISOLDE Solenoidal Spectrometer

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Abstract: The ISOLDE Solenoidal Spectrometer (ISS) will be used to measure various fission observables using a range of actinide beams via the (d, pF) reaction. These include fission barrier heights and fission fragment atomic charge (Z) and mass (A) yields. Neutron-induced fission cross sections will be measured by studying the variation of fission probability with excitation energy. The effect of angular momentum on the fission probability will also be studied. By extending both the range of isotopes and excitation energies beyond the capabilities of fixed target experiments, these measurements will have implications for nuclear models, astrophysics and applications including energy production.

Installation: ISS

1 Motivation

Studying the process of nuclear fission offers fundamental information required to both validate and further develop reliable nuclear models, whilst also providing crucial data for a wide range of fields including nuclear astrophysics, basic research, and applications such as nuclear energy, security, safeguards and isotope production [1]. Despite fission being discovered over 80 years ago, this complex process still presents challenges for both experimentalists, studying fissioning systems, and theorists alike, aiming to produce robust and predictable descriptions of the fission process. An opportunity exists to extend the current scope of study of fission by combining the potential availability of actinide beams at HIE-ISOLDE with the implementation of a fission detection setup with the ISS.

Measurements of fission cross sections, fission-fragment yield distributions and barrier heights collectively provide fundamental information on the fission process [2]. These observables are also inputs into state-of-the-art nucleosynthesis calculations for the r-process and the related fission recycling processes [3, 4]. Furthermore, these data are critical in the design of advanced nuclear reactors and waste transmutation in sub-critical reactors [2]. In particular, whilst direct measurements of fission barriers and angular momentum distributions are experimentally challenging, they are important components in understanding the fission process [5, 6, 7, 8].

The study of neutron-induced fission by direct neutron irradiation is possible only in a limited subset of nuclei close to stability. For example, many fission cross section measurements have been performed at the n_TOF facility [9] from thermal up to GeV incident neutron energies. However, experiments are limited by target properties and availability; aside from access to enriched material, both the natural radioactivity and diminishing lifetimes of ever more exotic desired isotopes make fixed target experiments unfeasible. Additionally, direct measurement of the fission barrier height in fissile isotopes is not possible using neutron induced reactions. Charged particle and photon-induced fission have been studied [5, 6, 7] but these measurements exist only for isolated stable or long-lived isotopes [5, 6]. Measurements have generally focused on extracting one experimental parameter rather than using one data set to determine several — it is advantageous to determine fission parameters simultaneously using multi-modal detection and under selectable experimental conditions. Moreover, the range of available excitation energies of the compound nucleus prior to fission is often restricted [6] and often not well defined in a particular experiment. Recent measurements with radioactive ion beams have used beta-delayed fission to measure barrier heights; these results exhibit discrepancies between 15% and 40% with respect to theoretical calculations [8]. To overcome these issues, experimental techniques that directly probe the aforementioned fission properties are required.

Using radioactive ion beams in inverse kinematics offers a way of achieving multi-modal detection of fission observables in exotic fissioning systems with a number of advantages. By passing the radioactive ion beam through a deuterated CD_2 target and using the single-neutron-adding (d,p) reaction (see for example [10]), compound nuclear states across a wide range of excitation energy can be populated, both above and below the fission barrier and neutron separation energy, S_N. Direct measurement of this excitation energy through the kinematics of the ejected proton and observation of the fission fragments gives the fission probability as a function of excitation energy, in the regions above and below both the fission barrier and S_N simultaneously, in a way not currently possible with other techniques. This technique therefore provides a direct measurement of the fission barrier height.

The use of inverse kinematics provides further advantages. Light-mass target contamination can limit (d, pF) measurements in normal kinematics to low energies [2, 11, 12]; using an actinide beam in inverse kinematics can address this problem. The solenoidal spectrometer technique allows the (d, pF) reaction to be well separated from other potential transfer and fusion evaporation reaction channels involving the carbon in the target — at the beam energies of >7.5 MeV/u required for the transfer reaction, reaction products from these channels are predominantly forwards focused, whilst the protons from the (d, p) reaction are emitted into the backwards hemisphere. The heavy compound system is strongly forward focused in the laboratory frame and can fission 'in-flight' allowing for the detection of fission fragments over a relatively small solid angle ($\theta_{LAB} \lesssim 20°$ - dependent on beam energy). Gas ionisation detectors placed downstream of ISS will allow the measurement of both the fission-fragment energy and specific energy loss ($\mathrm{d}E/\mathrm{d}x$) along the full fragment trajectory via off-line pulse shape analysis, allowing the measurement of both mass and charge yields via kinematic reconstruction and Bragg spectrosopy respectively. Finally, recent theoretical work has demonstrated a sensitivity of the fission probability to the angular momentum of the fissioning state [13, 14]; it may be possible to shed light on the role of angular momentum in the fission process by measuring the average angular distribution of the ejected protons in the centre-of-mass frame, which is related to the angular momentum distribution of the states that subsequently lead to fission.

A measurement is planned in early 2021 using the setup described in the following section to test and verify the technique with HELIOS at Argonne National Laboratory. The test-case will be the $d(^{238}\mathrm{U}, pF)$ reaction, for which properties such as the fission barrier height, yields and neutron induced cross section are well known. This experiment will offer valuable insights into the details of the experimental and analysis procedures allowing new measurements to be performed at ISS with confidence.

2 Experimental details

2.1 Experimental setup

The experimental setup will be composed of the existing ISS spectrometer in its standard (d,p) configuration and an array of four Fission Fragment Identification (FIFI) Bragg detectors placed on the downstream door of ISS. An illustration of the set-up is shown in Figure 1. An event will be defined by the detection of a proton or a fission fragment. The fission channel will be selected by gating on the detection of fission fragments in the FIFIs in coincidence with a proton. The solid angle that the FIFI detectors subtend is planned to increase with a future design, compared to the initial implementation at HELIOS, using an upgraded detector geometry in which they can be placed closer to the deuterated target. For the purpose of this LoI however, details of the existing apparatus are given here. A simulation of the intensity distribution for fission fragments arriving at the plane of the downstream door is shown in Figure 2 in which the openings to the four FIFI detectors are indicated, positioned at radii of 18 cm on the downstream door. Opposing pairs of detectors are orientated at angles of 10° and 15° to the beam axis, aimed at detecting heavy and light fragments respectively. For the case of $d(^{238}\mathrm{U}, pF)$ with a beam energy of 10 MeV/u, the centre-of-mass angular coverage for fission fragments is 62-147° for light fragments ($A \sim 100$) and 72-132° for heavy fragments

($A \sim 140$) with the target positioned 0.65 m from the downstream door. For the current purposes, the mean of the equilibrium charge state distribution, following passage through a carbon charge-reset foil, has been used to calculate the effect of the magnetic field on the fragments — but trajectories do not vary much across the charge state distribution compared to the size of the detector opening. The combined solid-angle coverage of the FIFI detectors is 1.94 and 1.37 sr for light and heavy fragments, respectively. Each detector will have a position-sensitive MWPC detector at the entrance to provide a fast timing reference signal and to allow reconstruction of the fragment trajectories.

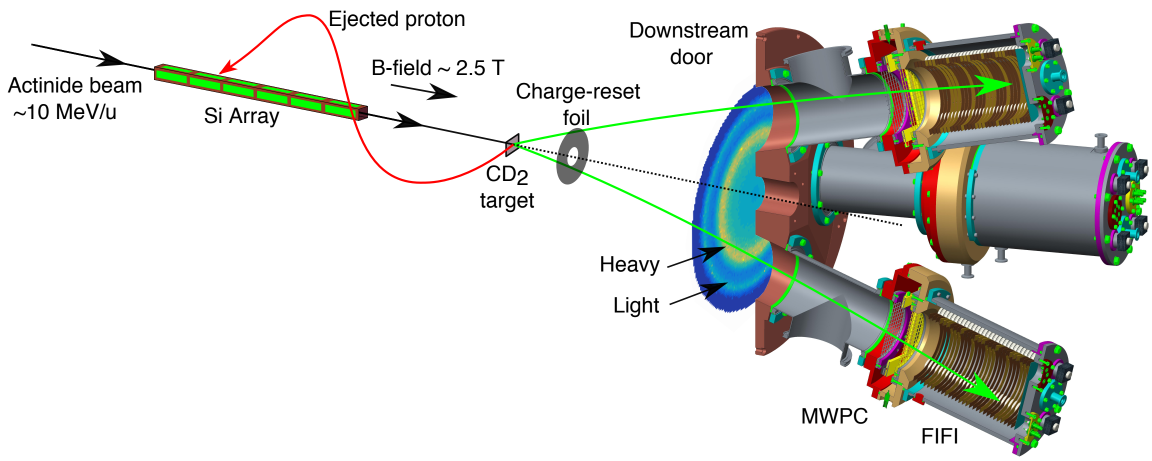

Figure 1: Illustration of the experimental arrangement within the ISS magnet. Trajectories of the proton and fission fragments from a single (d, pF) event are shown. A cut-through of the FIFI detectors mounted on the rear door of the ISS is shown, with three out of four detectors visible. The simulated distribution of fission fragments is also shown.

Absolute transfer cross sections will be extracted by monitoring the target thickness via elastically-scattered deuterons detected in an on-axis silicon monitor detector combined with a measurement of the integrated beam dose on a Faraday cup at zero degrees, both positioned downstream of the target.

2.2 Background reactions

The most problematic possible background reactions would be fusion-evaporation events on the carbon in the target that could lead to a fissioning nucleus with the emission of an evaporated proton — this would lead to false 'proton + fission' coincidences. Simulations show, however, that only $\sim 1\%$ of the total number of 'proton + fission' reactions are composed of fusion-evaporation-fission events, in which the emission of the evaporated proton is directed into the backwards hemisphere. To investigate the contribution from such a background channel, measurements with a C target can be made. This background can also be removed by putting a constraint on the sum of the Z and masses of the two fragments measured in the FIFI detectors.

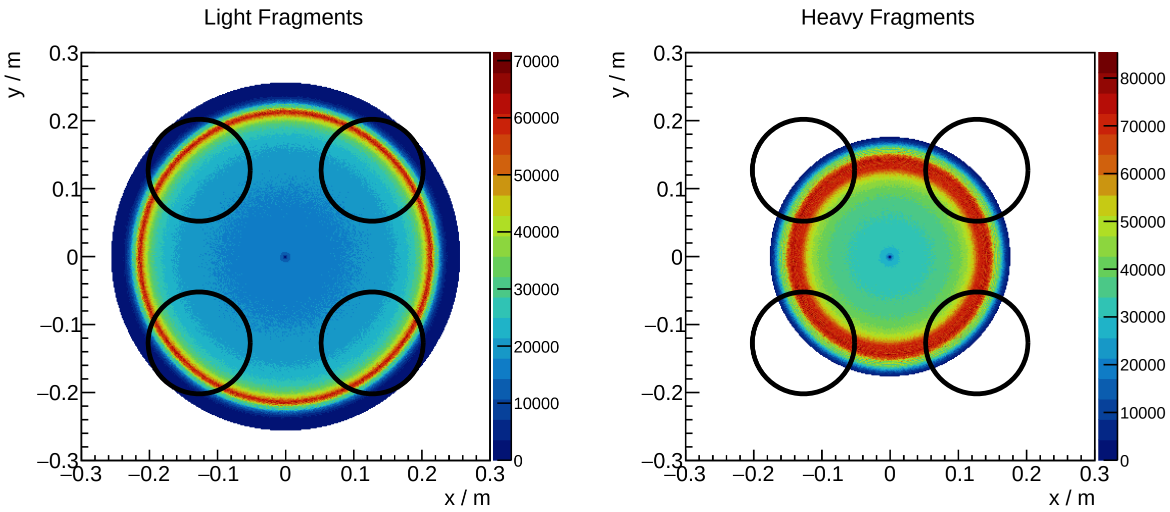

Figure 2: Fission-fragment intensity distributions at a distance of 0.65 m from the target simulated for the light (left) and heavy (right) fragment peaks for the case of the $d(^{238}\mathrm{U}, pF)$ reaction with a beam energy of 10 MeV/u. Positions of the entrance apertures to the FIFI detectors are shown as black circles. The fission-fragment mass distributions are taken from Ref. [15] with an isotropic angular distribution assumed in the centre-of-mass frame. The magnetic field mapping is included in the simulation.

2.3 Measurable fission properties

Fission-barrier height: The fission barrier height and shape can be extracted by measuring the yield of fission-fragments detected in the FIFI detectors coincident with protons from the (d, p) reaction in the position-sensitive silicon array with respect to the total (d, p) yield as a function of excitation energy in the residual compound nucleus — accounting for the counting efficiencies, this quantity defines the fission probability. For excitation energies at and above the fission barrier, a sharp increase in the fission probability will occur allowing the magnitude of the fission barrier to be determined.

Neutron induced fission cross section: The fission probability differential in excitation energy, determined experimentally as previously described, can be used to constrain Hauser-Feshbach calculations of neutron induced fission cross sections using an approach that is now well established [2], and proven to be accurate to within $\sim 10\%$. Studies of this nature therefore provide a source of key nuclear data in those cases not feasible in fixed target experiments.

Fission-fragment charge distribution: At low fragment energies, such as those in normal-kinematics experiments, obtaining a good atomic number identification capability is challenging as fragments are produced with energies below that of the Bragg peak (above which conventional Bragg-curve spectroscopy may be employed). Due to the kinematic boost of the fragments in inverse kinematics, from which the fragments have increased velocities in the lab frame ($\sim 0.1 - 0.2\, c$) compared to those in normal kinematics, a measurement of the full Bragg peak is possible. The Z distribution of fission fragments will be determined using Bragg spectroscopy; Bragg curve spectrometers typically give Z resolutions of $\sim$1% and energy resolutions $\sim$0.5% [16].

Fission-fragment mass distribution: Measurement of the laboratory frame emission angles and kinetic energies, using the MWPC and Bragg detectors respectively, of the light and heavy fragments allows their masses to be determined; reaction kinematics combined with accurate knowledge of the beam energy and measurements of the ratio between both laboratory emission angles and ratio of the kinetic energies of the two fragments uniquely defines the fragment masses. The effect of the magnetic field on the measured laboratory emission angles of the two fragments (of similar mass-to-charge after passing through a charge-reset foil) almost completely cancels in the ratio. Given the uncertainties associated with the required measurements, we expect a mass resolution of $\sim$5 u.

Angular momentum of participating states: The centre-of-mass angular distribution of the protons detected with the silicon array will allow identification of the angular momentum of the states leading to fission, although this is complicated by the doorway states through which the nucleus channels before fission. Whilst the states populated are potentially unbound and so no isolated states will be identified, it may be possible to extract an average distribution for small ranges in excitation energy. This aspect of the technique will be much more incisive where the fission barrier resides below S_N.

Fission γ-rays and neutrons: The proposed setup could in addition be augmented with γ-ray detectors and liquid scintillators to perform prompt and delayed spectroscopy. These further developments would also allow efforts to study the competition between fission and other decay modes. Although not currently part of the experimental setup, this is an envisaged upgrade.

3 Beam requirements

Despite the previously noted numerous benefits over fixed target experiments, the limiting factor with this technique is not the target life-time, radioactivity or availability but rather the beam production rate. The novel aspect of this experimental technique means that there exists a plethora of suitable beam candidates, and this LoI proposes an extensive future experimental campaign involving many actinide beams. This will require the development of new actinide beams, already a focus of the LISA project at ISOLDE. The feasibility of an individual measurement is multifaceted, including many beam requirements. In general, the beam must have a minimum energy of 7.5 MeV/u in order to maximise the (d, p) cross section and allow a suitable range of excitation energies and thus fissioning states in the compound nucleus to be populated. The beam intensity required depends on the individual case and the probability of fission, P_f, once the compound nucleus has formed as well as the experimental efficiency; a minimum requirement of 10^5 pps is estimated. The beam must be relatively isotopically pure; contaminants from other elements are less problematic due to the accurate Z measurement of the fissioning system allowed by this technique.

For nuclear energy applications, actinides found in reactor fuel (thorium, uranium and transuranics) will be of particular interest for future measurements and therefore effort to develop these beams is welcomed. For other fields and applications, particularly nuclear astrophysics, measurements of the neutron rich actinides in general are ideal candidates to provide new experimental data for nuclear models. Protactinium isotopes provide the most logical starting point for an online experimental campaign, given the fission probability and expected production rates (see Fig 3). It is envisaged more challenging measurements will be performed in the future as beam production and experimental apparatus are refined and developed. Figure 3 summarises possible actinide beam candidates for a potential experimental campaign — the significant overlap of fission probability with in-target production rates indicates the many possibilities for future experimental fission campaigns with ISS. Finally, although this LoI concentrates on actinide beams, the application of this technique can also be used to measure other fissionable isotopes such as those of Rn and Ra which will be of future interest to this collaboration.

In order to test the delivery of beams at this mass through HIE-ISOLDE, it is envisaged a beam such as ^{238}U would be used, which could simultaneously commission the experimental setup. Furthermore, offline measurements over the winter periods of longer-lived actinides, using a previously irradiated target, could be performed in collaboration with the LISA project, dependent on the outcome of yield measurements planned for early 2021.

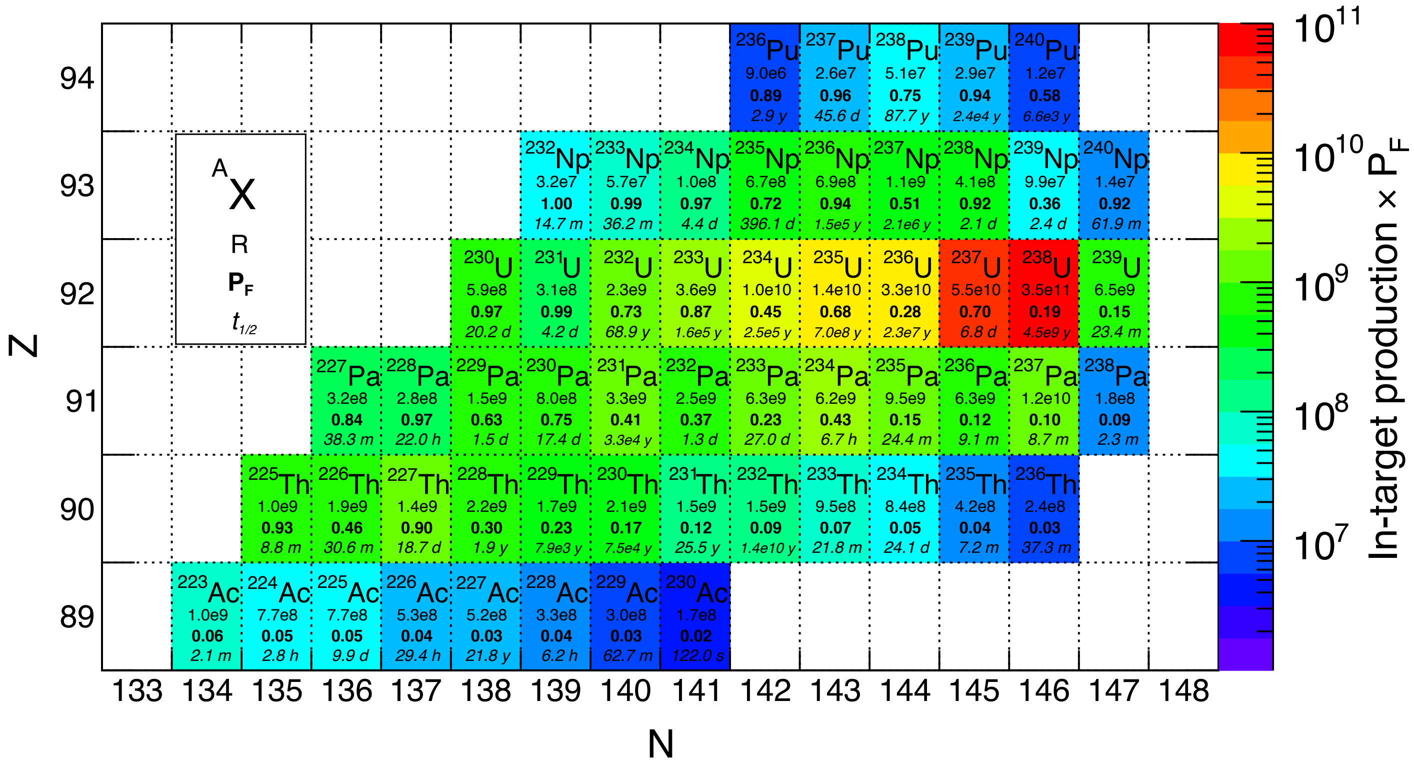

Figure 3: Fission probabilities at the peak of the first chance fission threshold (calculated with GEF), alongside estimated FLUKA in-target production rates (μC^{-1}) (R) in a standard UC material (taken from [18]) given with their half-lives. Neglecting extraction losses, beam contaminants and small differences in (d, p) cross sections, overlap between production rate and fission probability is indicative of possible actinide beam candidates for a (d, pF) measurement campaign with the ISS and the product of R and P_F is shown in the colour scale.

4 Summary

The motivation for studying and measuring fission observables across a wide range of isotopes is clear and shared across many fields and applications. An opportunity exists to perform a comprehensive experimental campaign studying the nuclear fission process using novel methods. The proposed experimental setup offers many advantages over previous efforts and if combined with the development of actinide beams at ISOLDE shall lead to the first measurements of fission observables for certain isotopes. Although the methods are novel, the underlying experimental processes and concepts are well understood and the scheduled ^{238}U(d, pF) test measurement with HELIOS will aid refinements and help optimise future measurements with the ISS. The experimental setup can be commissioned with a ^{238}U beam before benefitting from a possible winter running period, measuring longer-lived actinides. This will be in collaboration with the LISA project, aiding actinide beam development at ISOLDE in order to utilise this opportunity to its maximum.

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