

# EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

## Proposal to the ISOLDE and Neutron Time-of-Flight Committee

(Following HIE-ISOLDE Letter of Intent I-224)

### Fission of $^{230}\text{Ac}$

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**Abstract:** We propose to measure, for the first time, fission of the unstable nucleus  $^{230}\text{Ac}$ , using the ISOLDE Solenoidal Spectrometer. While a wealth of new data on fission of unstable nuclei on the neutron-deficient side of the valley of beta stability has been obtained in recent years, this is not the case for neutron-rich fissile nuclei. Fission studies in inverse kinematics, employing (d,pf) reactions, are a promising way to access such isotopes, which are interesting not only for increasing our understanding of nuclear fission, but also for investigating the role of fission in the r-process.

**Requested shifts:** 25 shifts, (split into 1 run over 1 year).



# 1 Physics case

In recent years, a series of experiments with radioactive beams at ISOLDE, GANIL, and GSI has more than doubled the number of nuclei whose low-energy fission properties have been investigated (see e.g. Ref. [1] and references therein). Breakthroughs in experimental techniques have allowed to study fission fragment mass or charge distributions as function of the proton and neutron numbers of the fissioning nucleus. These experiments were focused on neutron-deficient nuclei, while data on fission of neutron-rich nuclei are much more sparse. This proposal aims at the first confirmed observation of fission of  $^{230}\text{Ac}$ , a measurement of its fission barrier height, as well as collecting data on  $\gamma$ -rays emitted during fission. By employing the ISOLDE Solenoidal Spectrometer (ISS), the fission probability of  $^{230}\text{Ac}$  as function of the excitation energy can be studied in inverse kinematics using a  $(d,pf)$  reaction.

From a nuclear astrophysics point of view, fission ultimately limits the maximum mass of nuclei created during the *r-process*, when extremely neutron-rich isotopes formed by long chains of  $(n,\gamma)$  reactions and  $\beta$ -decays ultimately become so heavy that either  $(n,f)$ ,  $\beta$ -delayed fission, or spontaneous fission become possible [2]. The resulting fission fragments, again very neutron-rich, are expected to influence the elemental abundance as they can undergo further  $(n,\gamma)$  reactions and  $\beta$ -decays. The role of this so-called *fission recycling* in the *r-process* is still not clear. Data from a *kilonova*, the optical signature of a neutron star merger, suggest that the *r-process* takes place during such events [2]. This means that there is a need for data on fission of neutron-rich isotopes to test and to improve models employed in nuclear network calculations, simulating the *r-process* in neutron-star mergers [3].

Fission itself is a highly complex process, which constitutes a formidable challenge for experiment and theory alike. At low excitation energies, the interplay of microscopic nuclear structure effects, like pairing and shell closures, with macroscopic properties of cold nuclear matter, strongly influences observables such as the fission fragment mass and charge distributions. Another key observable reflecting the influence of nuclear structure is the fission barrier, indicating the energy threshold at which fission opens as a decay channel. Experimentally, the fission barrier is investigated by measuring the fission probability as function of the excitation energy. At facilities such as n\_ToF at CERN, such measurements can be performed with very high precision and allow studying, for example, the role of coupling between nuclear levels built on the ground state and on the minimum in a double-humped fission barrier, resulting in resonance structures in the measured cross-sections (see e.g. Refs. [4, 5]). Similar measurements for unstable neutron-rich nuclei are not possible, as suitably long-lived targets are not available. Experiments with radioactive beams in inverse kinematics, using the surrogate technique, allow us to overcome this limitation [4].

We propose to use the  $^{229}\text{Ac}(d,pf)$  reaction in inverse kinematics. The applicability of this approach has been demonstrated in a proof-of-principle experiment using a stable  $^{238}\text{U}$  beam at Argonne National Laboratory and the HELIOS spectrometer [6]. The obvious next step is to extend this approach towards unstable nuclei using the ISOLDE Solenoidal Spectrometer (ISS) [7]. This is challenging, as beam intensities, especially for neutron-rich fissile nuclei, are significantly lower than for stable beams. Therefore, the proposed setup

differs from that at HELIOS, insofar as it has been optimized for the detection efficiency of fission events at the cost of measurement of charge and mass distributions of the fission fragments. Given that predictions of fission fragment mass and charge distributions are quite consistent across microscopic models, measuring fission barriers is arguably more urgently needed (see e.g. Ref. [8]). Moreover, a measurement of  $\gamma$ -rays is foreseen to obtain data on the total energy and multiplicity of  $\gamma$ -rays emitted during fission. As a first physics case for this kind of experiment, fission of  $^{230}\text{Ac}$  has been chosen, based on available beam intensities, the expected fission probabilities, and the fact that there are no conclusive fission data at low excitation energies for this isotope available. We note that one experiment reported two inconclusive  $\beta$ -delayed fission events in mica foils [9], while a second focused on heavy-ion induced fission at high excitation energies [10]. Such a first step is also useful to pave the way for measurements using dedicated detectors for the measurement of fission fragment charges, which will be commissioned in an INTC-approved winter physics experiment [11].

Data on the energy and multiplicity of  $\gamma$ -rays emitted during fission are interesting for *r-process* simulations but also for a better understanding of angular momentum generation in fission [12].

The neutron separation energy of  $^{230}\text{Ac}$  is 4.923 MeV [13] and thus lower than the expected fission barrier height, which is 5.37 MeV according to Ref. [14], or 7.01 MeV according to the GEF model [15].

For fission measurements, beams of heavy protactinium or thorium isotopes would be very interesting due to their higher fission probabilities. Therefore, the development of beams of such isotopes is of great interest for future experiments of a similar kind. ISS is currently in a world-wide unique position to perform such experiments.

## 2 Experimental details

### 2.1 Setup

The proposed ISS experiment will use a number of detectors inside the ISS solenoid magnet, all in vacuum, for the detection of protons, elastically scattered deuterons, fission fragments, and  $\gamma$ -rays. A schematic figure of the detector arrangement is given in Fig. 1.

#### 2.1.1 Target

In order to allow for  $(d,pf)$  and  $(d,p\gamma)$  reactions, a deuterated plastic target,  $\text{CD}_2$ , with a thickness of  $1 \text{ mg/cm}^2$  will be used. The thickness limits the energy resolution but maximises the achievable luminosity. Also,  $0.5 \text{ mg/cm}^2$  targets will be mounted and used if the beam intensity on target is sufficiently high.

#### 2.1.2 Silicon array

Protons emitted in  $(d,pf)$  reactions can travel upstream in a helical trajectory, given by the solenoidal magnetic field. Those protons eventually hit the silicon array that surrounds the incoming beam-line [16] (Fig. 2). The excitation energy of the populated state as well

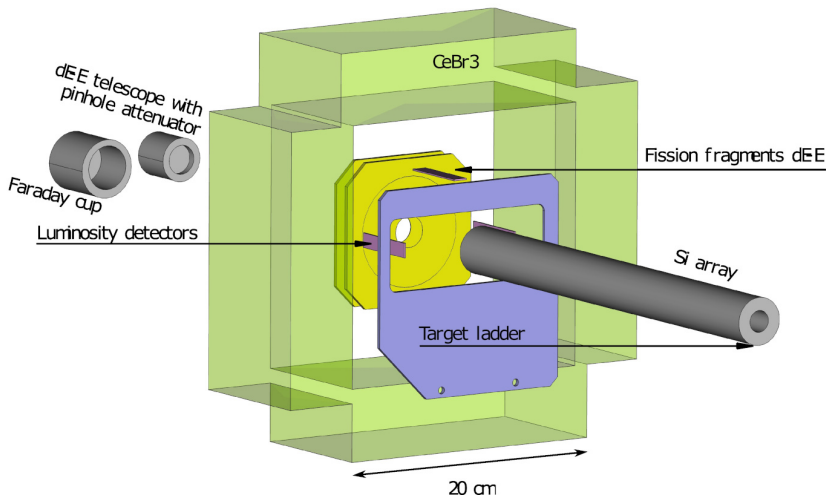


Figure 1: Schematic illustration of the detectors in the vicinity of the target, used inside the vacuum chamber of the ISS magnet. The Faraday cup and the  $\Delta E$ -E telescope with its attenuator are mounted on a movable support. Only one of those parts of the setup will be used at any given time. Except for those beam monitor detectors, all dimensions and positions are to scale with placements optimized by simulations. The hollow upstream silicon array is also indicated. Beam enters from the right.

as the emission angle of the proton in the centre-of-mass frame can be determined from the deposited energy and position along the array.

The main source of background are Multi-Nucleon Transfer (MNT) reactions with the carbon nuclei in the target, followed by fission [6, 17]. Requiring a coincidence between a proton in the silicon array and fission fragments, eliminates all reactions resulting in target-like ejectiles which move in forward direction in the laboratory frame. The remaining background channels consist predominantly of  $^4\text{He}$  or light charged ejectiles below  $A \approx 9$ . Fortunately, the cyclotron period  $T_{cyc}$  of those ejectiles is, with about 65 ns or longer, different from that of protons with about 32 ns and can be suppressed by measuring the time difference between a hit in the array and the fission fragment detectors with sufficient resolution. Inelastic scattering does not result in reaction products in backward direction and the probability for the emission of charged particles in a fusion-evaporation reaction is below 1%, according to a PACE4 calculation. Background measurements using a pure carbon target of equivalent thickness will be used to subtract any remaining background.

The data on protons without coincident fission fragments need to be corrected for deuteron breakup. While this affects the absolute value of the fission probability, the fission barrier height can still be extracted [6].

The strength of the magnetic field as well as the length and the positioning of the silicon array determine the excitation energy range covered by the measurement. By using a solenoid field of 2 T and covering a longitudinal range of 20 to 340 mm upstream from the target, excitation energies in the range of 6 to 9 MeV can be measured.

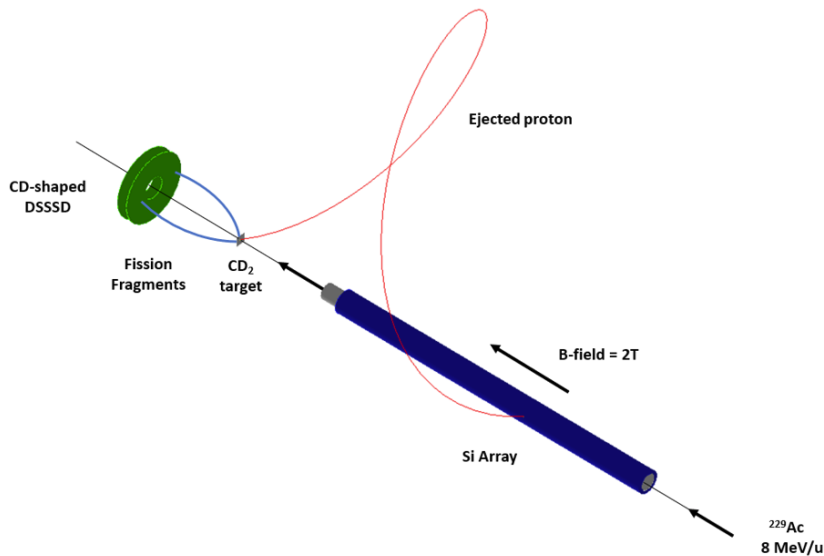


Figure 2: Schematic illustration of parts of the experimental setup. The accelerated beam enters from the right into the superconducting solenoid through the hollow position-sensitive silicon detector array, located in the upstream direction. Typical helical proton and fission fragment trajectories are indicated.

### 2.1.3 Fission fragments: $\Delta E$ -E CD DSSD

The two fission fragments (with approximately  $30 \leq Z \leq 60$  and  $80 \leq A \leq 150$ ) from a fission reaction in inverse kinematics both move in the forward direction inside a cone with an opening angle depending on the beam energy and the transverse momentum of the given fission fragment. Fission fragments can be detected with two annular CD-shaped double-sided silicon detectors, each of which is segmented in rings and sectors (Fig. 2). This allows to identify fission events by two coincident hits that differ in azimuthal angle by  $180^\circ$ . Using a  $\Delta E$  detector with a thickness of  $65 \mu\text{m}$  followed by a  $1000 \mu\text{m}$  detector allows performing a  $\Delta E$ -E measurement.

Although some of the fission fragments might punch through the  $\Delta E$  detector and give a signal in both detectors (the typical energy deposit will be about  $0.5 \text{ GeV}$  in each layer), the pulse-height defect of the energy loss signal of such heavy nuclei in silicon detectors limits the achievable energy resolution drastically. However, lighter nuclei, e.g. carbon or deuterium nuclei scattered out of the target, can be distinguished if they reach the detectors.

The geometric efficiency of this detector system, placed  $10 \text{ cm}$  downstream from the target, at a beam energy of  $8 \text{ MeV}/u$ , is  $87\%$  for detecting a single fission fragment, and  $82\%$  for detecting two fission fragments in coincidence. The annular design allows unreacted beam, as well as recoils from (d,p) events, to pass through the aperture in the centre of the detectors. Note that recoils from (d,p) reactions, which do not result in fission, are deflected only slightly from the beam direction due to the high mass of the projectile.

The  $\Delta E$ -E detectors have been purchased and are available. They differ only in thickness, segmentation and size from recoil detectors which are currently in use at ISS experiments.

#### 2.1.4 Luminosity monitors

The number of nuclei in the incoming beam is determined by using four off-axis silicon strip detectors. These detectors measure elastically scattered deuterons from the target, in the angular range of  $75^\circ - 85^\circ$  in the laboratory frame.

Four position-sensitive silicon detectors will be mounted parallel to the beam-line with the active surface oriented vertically or horizontally, respectively, see Fig. 1, allowing for a total detection efficiency of 0.8 %. This value assumes that the detectors are mounted 2 cm downstream from the target and at 3 cm off the beam axis. These four detectors are placed at positions differing by  $90^\circ$  in azimuthal angle, allowing to take an imperfect beam tune and a finite beam spot size on the target into account. This layout is motivated by experience at HELIOS/SOLARIS, where a similar configuration was used.

Procurement of the detectors as well as design of the mechanical support are in progress.

#### 2.1.5 Gamma-ray detection

Detecting  $\gamma$ -rays in coincidence with  $(d,pf)$  reactions requires the use of  $\gamma$ -ray detectors inside vacuum and a strong magnetic field. The Leuven group has demonstrated that  $\text{CeBr}_3$  detectors are an ideal solution [18]. We plan to construct a new mechanical support for the existing detectors of the Leuven group, allowing to position them in a box-like configuration with two rings, see Fig. 1. After simulating several geometric configurations, this arrangement optimizes the detection efficiency in terms of the total reconstructed  $\gamma$ -ray energy as well as the  $\gamma$ -ray multiplicity. The detectors will be protected by a 2 mm aluminium shield which prevents the implantation of charged particles in their active volume. The detection efficiency for  $\gamma$ -rays at 1 MeV is 7.5 %. During analysis, an unfolding procedure will be used to take the finite efficiency into account [19, 20]. Background from reactions with carbon nuclei in the target will be subtracted.

#### 2.1.6 Beam diagnostics

Downstream from the fission detectors, beam diagnostics for tuning and monitoring will be installed (Fig. 1). For beam tuning, a suppressed Faraday cup will be used, while the stability of the tune and intensity during the experiment will be monitored by a  $\Delta E$ -E silicon detector configuration with a pinhole attenuator. Both detectors are mounted on a motorized support. Either can be inserted into the beam-line remotely.

### 2.2 Beam and contaminants

A beam of  $^{229}\text{Ac}$  at  $\geq 8$  MeV/ $u$  is needed for the proposed experiment. Given by the kinematics of the reaction, the highest excitation energy of a fissioning nucleus that can be observed is limited by the position and length of the silicon array [6]. Fission of a nucleus at excitation energies higher than a few MeV above the beam energy results in downstream emission of protons, which is not covered by the silicon array.

For this proposal, a beam intensity of  $1.3 \times 10^5$  pps on the ISS target is required to measure the height of the fission barrier, see the discussion in Sec. 2.3. To avoid contamination from Ra or Fr, the  $^{229}\text{Ac}$  can be extracted as  $^{229}\text{AcFF}^+$  from a ThCx or UCx target with a VD5

ion source and a calibrated leak for CF<sub>4</sub> gas injection. Assuming a transmission efficiency of 1 % from the ISOLDE target to the ISS target, extracted rates of  $1.3 \times 10^7$  are required. Yields above  $10^7$  <sup>229</sup>AcFF<sup>+</sup> have been measured from a UCx-VD5 target and ion source combination at ISOLDE at high (>2000°C) target temperatures. Simulated in-target production using FLUKA [21] predicts higher production rates for ThCx ( $1.8 \times 10^{10}$  vs.  $3.3 \times 10^8$  for UCx). In both cases <sup>229</sup>Ac is additionally fed by the decay of <sup>229</sup>Ra. The beam composition of <sup>229</sup>AcFF<sup>+</sup> was studied using the ISOLTRAP MR-ToF MS [22], showing no notable contamination. Yields for <sup>229</sup>AcFF<sup>+</sup> from ThCx-VD5 were only measured at temperatures below 2000°C but even at low temperatures, rates are above  $10^5$ . This suggests that at high temperatures the needed rates are possible. Yields are discussed per  $\mu\text{C}$  of protons incident on the target.

Since the setup is optimized for the detection of fission events, contaminants with a higher fission probability and a lower fission barrier are the most severe. Calculations of the fission probabilities of isotopes with similar  $A/q$  with GEF [15] suggest that any contaminant with  $Z \geq 89$  would be problematic. On the other hand, contaminants with  $Z < 89$  would not cause any problems. Based on the information available to us, only insignificant levels of contaminants with  $Z \geq 89$  are expected.

### 2.3 Required statistics

The number of events necessary for the first conclusively identified fission of <sup>230</sup>Ac is estimated to be about 50 triple coincidences between the two fission fragments and the proton.

To successfully determine the fission barrier with a 0.2 MeV resolution, 100 counts of fission events above the barrier per 0.2 MeV excitation energy bin will be necessary, see Fig. 2 in Ref. [6]. That experiment used targets with a thickness of  $500 \mu\text{g}/\text{cm}^2$  and a beam intensity of  $\sim 10^6$  pps, with a total integrated beam dose of around  $5.5 \times 10^{11}$  ions. Based on this information, assuming that background due to  $\alpha$ -particles from reactions with carbon nuclei in the target can be suppressed with sufficient time resolution between the silicon array and the other detectors, we estimate that we can determine the fission barrier height with integrated statistics which is about a factor of four lower than in the HELIOS experiment. The CD-shaped fission fragment detectors improve the angular coverage by a factor of 8. Using also a target which is a factor of 2 thicker than that used in the HELIOS experiment, this means in total an improvement of a factor of 50.

Assuming that the prediction of the GEF model [15] for the fission probability of <sup>230</sup>Ac,  $P_f = 3 \%$ , is correct (compared to 20 % for <sup>239</sup>U), a beam intensity of  $1.3 \times 10^5$  pps on the ISS CD<sub>2</sub> target is needed for 18 shifts to obtain sufficient statistics to determine the fission barrier height.

In order to take background from reactions with carbon into account 6 shifts with a pure carbon target are needed. One more shift is requested for beam tuning and setting up electronics.

**Summary of requested shifts: 25**

## References

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## DESCRIPTION OF THE PROPOSED EXPERIMENT

Please describe here below the main parts of your experimental set-up:

Part of the experiment	Design and manufacturing
ISOLDE Solenoidal Spectrometer	<input type="checkbox"/> To be used without any modification <input checked="" type="checkbox"/> To be modified
Fission detectors	<input checked="" type="checkbox"/> Standard equipment supplied by a manufacturer <input type="checkbox"/> CERN/collaboration responsible for the design and/or manufacturing

## HAZARDS GENERATED BY THE EXPERIMENT

Additional hazard from flexible or transported equipment to the CERN site:

Domain	Hazards/Hazardous Activities	Description
Mechanical Safety	Pressure	<input type="checkbox"/> [pressure] [bar], [volume][l]
	Vacuum	<input checked="" type="checkbox"/>
	Machine tools	<input type="checkbox"/>
	Mechanical energy (moving parts)	<input type="checkbox"/>
	Hot/Cold surfaces	<input type="checkbox"/>
Cryogenic Safety	Cryogenic fluid	<input type="checkbox"/>
Electrical Safety	Electrical equipment and installations	<input type="checkbox"/>
	High Voltage equipment	<input checked="" type="checkbox"/> 1 kV
Chemical Safety	CMR (carcinogens, mutagens and toxic to reproduction)	<input type="checkbox"/>
	Toxic/Irritant	<input type="checkbox"/>
	Corrosive	<input type="checkbox"/>
	Oxidizing	<input type="checkbox"/>
	Flammable/Potentially explosive atmospheres	<input type="checkbox"/>
	Dangerous for the environment	<input type="checkbox"/>
Non-ionizing radiation Safety	Laser	<input type="checkbox"/>
	UV light	<input type="checkbox"/>
	Magnetic field	<input checked="" type="checkbox"/> 2.5 T
Workplace	Excessive noise	<input type="checkbox"/>
	Working outside normal working hours	<input type="checkbox"/>
	Working at height (climbing platforms, etc.)	<input type="checkbox"/>
	Outdoor activities	<input type="checkbox"/>
Fire Safety	Ignition sources	<input type="checkbox"/>
	Combustible Materials	<input type="checkbox"/>
	Hot Work (e.g. welding, grinding)	<input type="checkbox"/>

Ionising radiation	Calibration sources	<input checked="" type="checkbox"/>	$\alpha$ calibration source (open source)
			4236RP
			$^{148}\text{Gd}$ , $^{239}\text{Pu}$ , $^{241}\text{Am}$ , $^{244}\text{Cm}$
			1 kBq, 1 kBq, 1 kBq, 1 kBq
	Target materials	<input checked="" type="checkbox"/>	$\text{CD}_2$ (500, 1000 $\mu\text{g}/\text{cm}^2$ , deuterated polyethelene)
			$^{\text{nat}}\text{C}$ (500 $\mu\text{g}/\text{cm}^2$ )
	Beam ions	<input checked="" type="checkbox"/>	$^{229}\text{Ac}$
	Beam intensity	<input checked="" type="checkbox"/>	max. available ( $1.3 \times 10^5$ envisaged)
Beam energy	<input checked="" type="checkbox"/>	max. available (8 MeV/ $u$ envisaged)	