EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

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

Feasibility studies towards the systematic investigation of the β-delayed fission in the neutron-rich actinides. Part I: ^{230,232,234}Fr and ^{230,232,234}Ac

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Abstract: The β-delayed fission (βDF) can provide wealth of information on low-energy fission of exotic isotopes and has an important impact on production of elements via the astrophysical r-process on the neutron-rich side of the Nuclidic Chart. At present, the βDF was only reported for five neutron-rich isotopes ^{228,230}Ac, ^{236,238}Pa and ^{256m}Es. All these measurements suffered from the impossibility to create a pure sample, which made specific A and Z assignments difficult, and caused complex background conditions.

The availability of mass-separated and laser-ionized beams of elements Ac, Pa and Np and surface-ionized Fr provides a crucial opportunity for the βDF investigations at ISOLDE. This LoI aims to initiate a unique programme of βDF and nuclear spectroscopic studies in this region, not paralleled by any facilities worldwide. A multi-step approach is considered within this programme, which requires several technical developments, including possible use of the Pa and Np molecular beams foreseen by the LISA (Laser Ionisation and Spectroscopy of the Actinides) Marie Sklodowska-Curie Innovative Training Network.

The project will run over several years and will involve a number of separate experiments. As a first step in this programme we wish to proceed with the feasibility βDF studies of the most neutron-rich Fr and Ac isotopes. The requested beam time will be used to test the yields, beam purity, possible use of molecular beams, background conditions and to perform the first identification of βDF in ^{230,232}Fr and in ^{232,234}Ac. Upon a successful completion of the first step, a dedicated proposal for detailed studies in this region will follow.

Requested shifts: If performed as a standalone experiment: 5 shifts with UC$_x$ target, in a single run: 1 shift for Ac laser tuning, 3 shifts for ^{230,232,234}Ac, and 1 shift for ^{230,232,234}Fr measurements. The experiment will be carried out at the ISOLDE Decay Station (IDS).

1 Motivation

With this LoI, we propose to initiate an extended programme to study β-delayed fission and nuclear spectroscopic properties of very neutron-rich Fr-Ac-Pa-Np nuclei, up to the present limit of their knowledge for some isotopic chains.

An overarching goal of this programme is to provide low-energy fission data for very exotic nuclei, whose fission studies are difficult or impossible by other presently-available techniques. Such data include e.g. fission probabilities, branching ratios, fission fragments mass distributions (FFMD) and charge distributions, γ-ray and neutron multiplicities.

In the last decade, our collaboration has performed a series of successful β^+/EC-delayed fission (ECDF) studies at ISOLDE for an extended region of the odd-odd neutron-deficient isotopes[1] of Tl, Bi, At and Fr [1, 2, 3, 4, 5, 6, 7, 8], see also reviews [9, 10]. Combined with the data in the heavy neutron-deficient actinides, a consistent picture of β^+/ECDF starts to emerge for the *neutron-deficient* nuclei (in total 24 cases) as summarized in [10].

In contrast to this, the fission knowledge is very sparse on the neutron-rich side, which is relevant to the fission termination and re-cycling in the r-process [11, 12, 13, 14]. This limitation is due to the experimental difficulties to access the nuclei in this region.

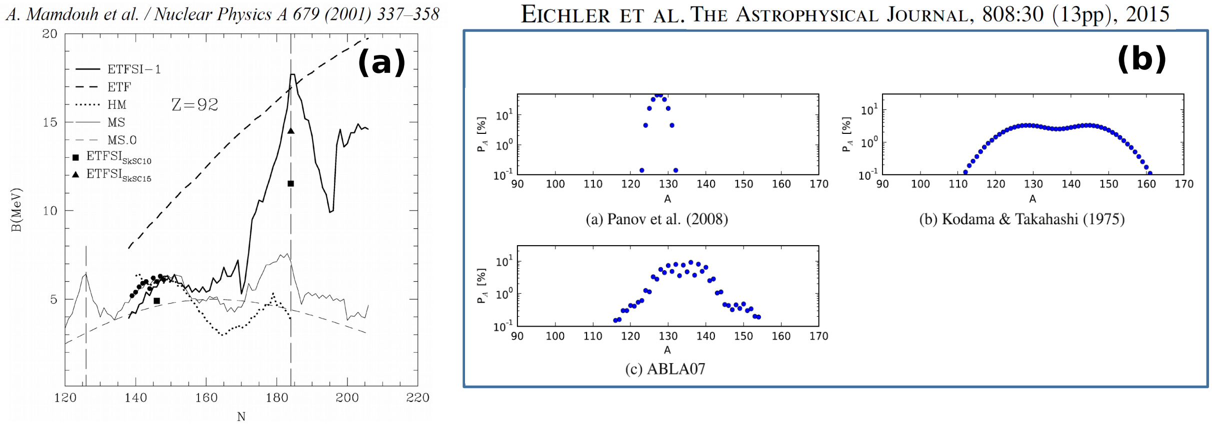
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Figure 1: a) Primary fission barriers for the uranium isotopic chain, calculated with the various models [16]. Neutron closed shells at $N = 126$, 184 are indicated by vertical dashed lines, experimental values by solid circles. b) Examples of three different theoretical FFMDs assumed for fission of ^{274}Pu [12].

The need for new data in this region is highlighted by Fig. 1. It presents the two main difficulties of the modern theoretical fission calculations, to predict the fission properties of nuclides with the N/Z ratios very different from the classical region of fission in e.g. uranium nuclei. Fig. 1a) shows that different calculations reproduce quite well the experimentally-deduced fission barrier heights for e.g. $^{232-238}$U ($N = 140$–146) isotopes in the vicinity of the β-stability line. The latter is because the parameters of such models are typically fitted to the known properties of nuclei in this region. However, the predictions strongly disagree for the very neutron-rich isotopes around e.g. ^{276}U ($N = 184$), with a spread of calculated values of up to 10 MeV between the models, which shows their

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[1] We remind that due to the energy considerations, only odd-odd isotopes are expected to have a measurable βDF mode, which is the case for all observed cases so far, see details in [9]. The Ra precursors, mentioned in the text, cannot have βDF, due to unfavourable energy balance.

deficiencies in respect of the isospin dependence. Such a difference in the fission barriers will directly influence the outcome of the fission termination in the r-process, whereby the probabilities of all three contributing fission mechanisms (spontaneous, βDF and neutron-induced) will be either strongly enhanced or suppressed [11, 12, 13, 14]. Fig. 1b) presents the typical theoretical FFMDs assumed by several commonly-used astrophysical network calculations for the case of very neutron-rich ^{274}Pu, which leads to quite different final elemental abundances via the fission re-cycling, see details in [12]. Since recently, the importance of fission data for the final r-process outcome is strongly emphasized in the literature [12, 13, 14], in addition to the need for the improved knowledge of other three experimental quantities which are typically discussed by the community: masses, half-lives and neutron-capture cross sections.

In particular, the βDF can provide wealth of information on low-energy fission of exotic isotopes [9] and has an important impact on production of elements in the nucleosynthesis [11, 17]. Both the βDF probability ($P_{\beta\mathrm{DF}}$) and the FFMDs play an important role in final abundances of isotopes via fission recycling in the r-process [12, 13, 14].

Importantly, one can expect specific differences between the β^+/ECDF and β^--delayed fission on the neutron-deficient and neutron-rich sides, respectively. Indeed, the two processes can have a different energy balance, due to the differences in e.g. neutron separation energies, fission barrier heights, β-decay strength functions. The structure of the fission barrier itself is known to be very different in the two regions, with a common occurrence of a complex double-humped barrier in the heavy actinides. A recent attempt to account for the latter was presented in a phenomenological βDF study [18]. The measurements of the $P_{\beta\mathrm{DF}}$ values, which depend exponentially on the difference Q_β(Parent)-B_f(Daughter), allows to estimate the height of the fission barriers, see e.g. [2] and references therein, thus to judge on the validity of different theoretical models.

2 Goals of this LoI

The main goal of this LoI is to initiate the systematic studies of the most neutron-rich isotopes ^{230,232}Fr and ^{230,232,234}Ac, which are expected to exhibit βDF, and which are relatively easily accessible at ISOLDE. Specific tasks include: the confirmation of the βDF in ^{230}Ac and its identification in ^{232,234}Ac and in ^{230,232}Fr; the investigation of production yields, beam purity and background conditions; tests of the most optimal detection system for the measurements of low βDF branching ratios.

As a by-product of this program, not requiring additional beam time, we will obtain extensive β-decay data for some of these isotopes, some of which were only studied in a single experiment [19, 20]. This will include the use of fast-timing technique, to get first life-time data on a large number of excited states in the daughter Ra and Th isotopes.

2.1 Earlier data for βDF of ^{228,230,232}Ac [21, 22, 23]

So far, the only evidence of βDF in actinium isotopes was claimed for ^{228,230}Ac [21, 22]. This was achieved via a complex chemical separation of an ^{230}Ra precursor (half-life of ≈1.5 h) from the transfer products in the ^{18}O+^{232}Th reaction [21], or from the products

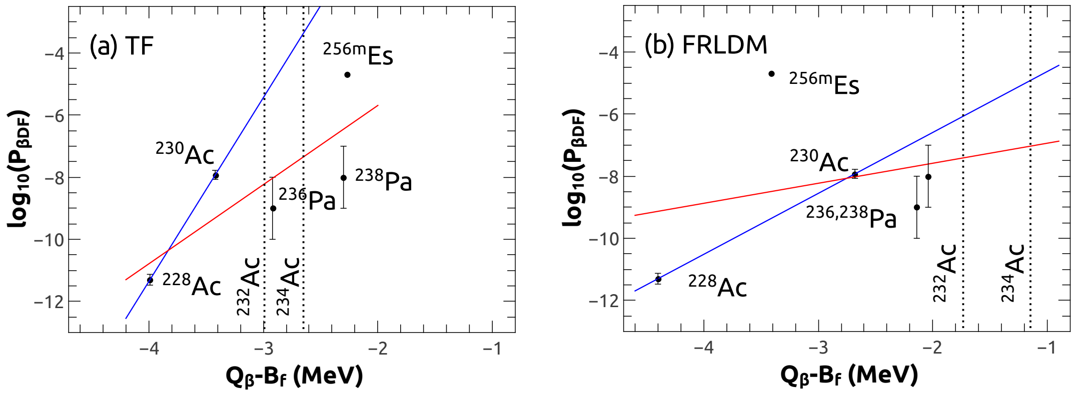

Figure 2: The known $P_{\beta\mathrm{DF}}$ values for the neutron-rich isotopes ^{228,230}Ac, ^{236,238}Pa and ^{256m}Es as a function of $Q_\beta(Parent) - B_f(\text{Daughter})$. The fission barrier heights, B_f, are taken from a) TF model [27] and b) FRLDM [28]. Experimental Q_β values are from [29]. Solid blue lines show a linear extrapolation from the $P_{\beta DF}$ values for ^{228,230}Ac only, the red lines are the fit to all five known $P_{\beta DF}$ values with equal weights to all points, see text for details. The disparity in the gradients for the extrapolations and fits, and thus the extracted $P_{\beta\mathrm{DF}}(^{232,234}\mathrm{Ac})$ values, is due to a difference in the predicted TF and FRLDM fission barriers. The dashed vertical lines show $Q_\beta - B_f$ values for ^{232,234}Ac within the respective models.

of the decay chain of ^{232}Th for the case of long-lived ($T_{1/2}$ ≈6 years) ^{228}Ra [22]. The fission observation was performed with the mica detectors, which are only sensitive to fission event as such, but cannot provide energy or time information. Furthermore, the chemical separation used could not provide clean sources, neither it removed some of the reaction or decay products, which could contribute to the fission background.

For ^{228}Ac, 17 fission events were reported during 720 days of measurements (thus a very low fission rate of 1 fission event in ≈ 40 days), resulting in a value of $P_{\beta DF}(^{228}\mathrm{Ac})=5(2)\times10^{-12}$ [22], being the lowest ever reported βDF probability. For, ^{230}Ac, based on 2 observed fission events, a value of $P_{\beta DF}(^{230}\mathrm{Ac})=1.19(40)\times10^{-8}$ was tentatively deduced [21]. In our opinion, due to the absence of A, Z or half-life identification in these experiments, doubts may be raised with respect to the validity of these results and their assignment to a specific isotope, thus it is important to check these data.

No βDF was observed for ^{232}Ac [23], with an upper limit of $P_{\beta DF}(^{232}\mathrm{Ac})<10^{-6}$ reported.

2.2 Fission yield estimation for ^{230,232,234}Ac

At ISOLDE, the βDF of mass-separated isotopes ^{230,232,234}Ac can be studied following their direct production with laser ionization, see also the concurrent proposal aimed at the Ac charge radii measurements by R. Heinke *et al.* [24]. These isotopes will also be produced via the β decay of the surface-ionized isobaric Ra precursors. In our short test in 2018 of the ^{213}Ra^{19}F molecular beam, we obtained a yield of ^{232}Ra as ≈2×10^4 ions/μC, which is comparable to 5.6×10^4 ions/μC quoted at the ISOLDE web-page [15], albeit for the SC-based experiment. The expected rate for the yet unmeasured ^{234}Ra, which will contribute to ^{234}Ac, was extrapolated from the lighter Ra isotopes based on their relative production cross-sections from the ABRABLA calculations and a conservative estimate of the ratio of decay losses of ^{232,234}Ra due to the difference in their half lives. The expected Ac decay rates are shown in Table 1. One of the aims of this LoI is to check these yields.

Table 1: Expected Ac decay rates, recalculated from the Ac proposal [24] and including the Ac production via the surface-ionised Ra precursors for 1 μA proton beam intensity. The 3rd column: the βDF probabilities for ^{232,234}Ac extrapolated from the data for ^{228,230}Ac for the TF and FRLDM approaches, see Fig. 2. The last column: the respective numbers of observed fission events. For ^{230}Ac, a measured $P_{\beta DF}$ is used, taken from [21]. The fission rate estimates include the ≈40% singles fission detection efficiency of IDS.

Isotope, $T_{1/2}$	Ac decays/s	$P_{\beta\mathrm{DF}}(^{232,234}\mathrm{Ac})$		FF/shift	
		TF	FRLDM	TF	FRLDM
^{232}Ac, 119(5) s	2×10^4	4.2×10^{-6}	8.6×10^{-7}	1000	200
^{234}Ac 44(7) s	2×10^2	4.3×10^{-4}	1.2×10^{-5}	1000	30
	Ac decays/s	$P_{\beta\mathrm{DF}}(^{230}\mathrm{Ac})$ [21]		FF/shift	
^{230}Ac, 122(3) s	1×10^5	$1.19(40) \times 10^{-8}$		14	

^{230}Ac fission yield estimate. Based on the ^{230}Ac decay rate shown in Table 1 (which includes its direct production via laser-ionization and indirect production via surface-ionized precursors ^{230}Fr,Ra) and by using the published $P_{\beta DF}$ value for ^{230}Ac from [21], an observed βDF rate of ≈14 fission/shift can be estimated. A rather small number of fission events makes this part of the experiment quite difficult, but on the other hand it constitutes a crucial check of the previous results from [21]. **We request one full shift for this important measurement for ^{230}Ac.**

$P_{\beta DF}$ and fission rate estimates for ^{232,234}Ac were made based on two approaches: either via a direct extrapolation from the measured $P_{\beta DF}$ data for ^{228,230}Ac from [21, 22] or by also including the known data for ^{236,238}Pa [25] and ^{256m}Es [26]. To check the sensitivity of the results, two widely-used fission models were exploited: the TF [27] and FRLDM [28], which predict quite different fission barrier heights.

In the first approach, by linearly-extrapolating the $P_{\beta DF}$ values from ^{228,230}Ac to ^{232,234}Ac, as shown by the blue lines in Fig. 2, the $P_{\beta DF}(^{232,234}$Ac) values were obtained for the TF and FRLDM models, and respective expected numbers of fission events were calculated (see Table 1). Expected statistics for ^{232,234}Ac (several tens to a thousand of fissions per shift within either TF or FRLDM approaches) should be enough for the first identification of βDF occurrence in these isotopes[2].

In the second approach, we used a weighted fit to the five known $P_{\beta DF}$ values, see the red lines in Fig. 2. However, we stress that only a handful of fission events was observed in a complex βDF evaluation for ^{236,238}Pa, with an order of magnitude uncertainty [25]. Only two βDF events were tentatively attributed to ^{256m}Es, with no uncertainty quoted [26]. This emphasized the difficulties of the previous experiments and the need for the re-investigation of these isotopes[3].

Based on a highly-tentative fit, the $P_{\beta DF}(^{232,234}$Ac) become up to two orders of magnitude lower, leading to respective fission yield reductions. However, even with such lower

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[2] For a reference, the observed fission rate in our βDF study of ^{180}Tl was ≈ 10 fissions/h. [1].

[3] We mention that ^{236,238}Pa and some other Pa isotopes could become available for the future βDF studies at ISOLDE following the technical developments within the EU-LISA network.

fission rates, we expect to get at least a few fission events for these isotopes, to establish the presence of their βDF. Furthermore, based on the difference in the predicted fission yields between the TF and FRLDM approaches, a conclusion on their validity for this case could also be made.

A large uncertainty between the fission rate estimations by the two approaches prohibits us from proceeding with a Proposal, that is why we first wish to perform a test measurement within this LoI. **Two shifts are asked for the measurements for ^{232,234}Ac.**

3 Tests for βDF in ^{230,232}Fr and for the first production of ^{234}Fr

The surface-ionized isotopes ^{230,232}Fr (half-lives $\approx$5–20 s) should also posses a βDF branch. Due to absence of reliable βDF systematics in this region (see the discussion of Fig. 2), any $P_{\beta DF}$ estimates for these isotopes would be too tentative, thus we need to perform a test before we consider a full proposal. Due to their shorter half-life values in comparison with the isobaric Ac's and Ra's, Fr decay can be studied in clean conditions with a properly selected faster implantation-decay tape cycling of IDS. The published yields of ^{230,232}Fr are $\approx 10^5$ and $\approx 3\times10^3$ ions/μC [20], and $\approx 2(1)\times10^3$ ions/s for ^{233}Fr [30]. Based on these yields, we can reach a level of $P_{\beta DF}\approx10^{-9}$ and $\approx10^{-7}$ in a 1-hour run for ^{230,232}Fr, respectively. These values are lower by at least an order of magnitude than the rather high upper limits reported in [23]. **We request one shift in total for ^{230,232}Fr and for a production test of the yet unknown ^{234}Fr.**

4 A future extension towards Pa and Np beams

This LoI will contribute towards a possible follow-up extended programme to investigate the βDF and nuclear spectroscopy properties of the scarcely-studied neutron-rich Pa and Np isotopes. As mentioned earlier, βDF was claimed for ^{236,238}Pa in the 1980s [25], but the data are very uncertain and needs to be re-investigated. The βDF of ^{228}Np was identified by Dubna and Berkeley experiments, respectively, see overview [9]. Thus, the extension of the studies to the Pa and Np species at ISOLDE would be of a great interest. This goal, however, requires a dedicated beam development program for such beams, which is foreseen by the activities within the LISA network. Molecular Pa and Np beams, as well as their laser ionization will be investigated.

5 Detection setup

The βDF measurements will be performed with the IDS setup in its standard configuration, which includes a tape station and 4 HPGe clover detectors. An annular silicon detector, or an array of several solar cell detectors, which are less sensitive to the radiation damage, will be installed in front of the implantation tape for measurement of singles α particles and fission fragments with geometric efficiencies of $\approx$ 20% and of $\approx$ 40%, respectively, in a configuration reminiscent of our previous work with the Windmill. A twice higher detection efficiency for fission fragments is because each fission event produces two fission fragments. A plastic scintillator detector will be placed behind the tape to register β particles and allow β-fission and β-γ coincidences The fast-timing $LaBr_3$ detectors will be also used for concurrent lifetimes measurements for excited states populated in β decays of Fr and Ac isotopes and their daughter nuclides as a by-product during the βDF runs (no extra beam time required).

Summary of requested shifts: If performed as a standalone experiment: **5 shifts** with UC_x target, in a single run: **1 shift for Ac laser tuning**, **3 shifts for ^{230,232,234}Ac, and 1 shift for ^{230,232,234}Fr measurements**. Otherwise, we request 4 shifts, if the Ac experiment [24] is accepted, and both experiments are combined together.

Upon the completion of the aforementioned tests, we plan to submit a proposal on the isotopes for which a complete investigation, including the measurement of the fission fragment mass and energy distribution with coincidence measurements, could be performed.

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Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT

The experimental setup comprises: (*name the fixed-ISOLDE installations, as well as flexible elements of the experiment*)

Part of the	Availability	Design and manufacturing
IDS	☒ Existing	☒ To be modified: Addition of annular Si detector or of an array of solar cells for α and fission fragment detection, and $LaBr_3$ detectors for fast-timing measurement

HAZARDS GENERATED BY THE EXPERIMENT (if using fixed installation:) Hazards named in the document relevant for the fixed IDS installation.

Additional hazards:

Hazards	α/fission-decay setup	usual usage	
Thermodynamic and fluidic			
Pressure	[pressure][Bar], [volume][l]		
Vacuum	Standard ISOLDE vacuum		
Temperature	[temperature] [K]		
Heat transfer			
Thermal properties of materials			
Cryogenic fluid	[fluid], [pressure][Bar], [volume][l]		
Electrical and electromagnetic			
Electricity	[voltage] [V], [current][A]		
Static electricity			
Magnetic field	[magnetic field] [T]		
Batteries	☐		
Capacitors	☐		
Ionizing radiation			

IDS tape	The tape where the radioactive samples are implanted could be damaged or stuck sometimes. Should it break and the IDS chamber needs to be opened, usual radioprotection procedures need to be followed.		
Beam particle type (e, p, ions, etc)	Fr, Ac beams		
Beam intensity	10^3–10^5 ions/s		
Beam energy	30-60 keV		
Cooling liquids	[liquid]		
Gases	[gas]		
Calibration sources:	☒		
• Open source	☒		
• Sealed source	☐ [ISO standard]		
• Isotope	^{241}Am		
• Activity	50 Bq		
Use of activated material:			
• Description	☐		
• Dose rate on contact and in 10 cm distance	[dose][mSV]		
• Isotope			
• Activity			
Non-ionizing radiation			
Laser			
UV light			
Microwaves (300MHz-30 GHz)			
Radiofrequency (1-300 MHz)			
Chemical			
Toxic	[chemical agent], [quantity]		
Harmful	[chem. agent], [quant.]		
CMR (carcinogens, mutagens and substances toxic to reproduction)	[chem. agent], [quant.]		
Corrosive	[chem. agent], [quant.]		
Irritant	[chem. agent], [quant.]		
Flammable	[chem. agent], [quant.]		

Oxidizing	[chem. agent], [quant.]		
Explosiveness	[chem. agent], [quant.]		
Asphyxiant	[chem. agent], [quant.]		
Dangerous for the environment	[chem. agent], [quant.]		
Mechanical			
Physical impact or mechanical energy (moving parts)	usual IDS chamber, surrounded by Clovers		
Mechanical properties (Sharp, rough, slippery)	[location]		
Vibration	[location]		
Vehicles and Means of Transport	[location]		
Noise			
Frequency	[frequency],[Hz]		
Intensity			
Physical			
Confined spaces	[location]		
High workplaces	[location]		
Access to high workplaces	[location]		
Obstructions in passageways	[location]		
Manual handling	[location]		
Poor ergonomics	[location]		

Hazard identification:

Average electrical power requirements (excluding fixed ISOLDE-installation mentioned above): negligible