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EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

ISOLDE and Neutron Time-of-Flight Committee

Addendum to the Proposal IS466: Identification and systematic studies of the β-Delayed Fission (βDF) in the lead region

IS466-3: Detailed βDF studies of ^{202}Fr and a search for βDF of ^{204}Fr

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Abstract

This Addendum is the continuation of our successful program to study exotic process of the β–delayed fission in the lead region, initiated by the proposal CERN-INTC-2008-001/P-235 [1], "Identification and systematical studies of the electron-capture delayed fission (ECDF) in the lead region - Part I: ECDF of ^{178,180}Tl and ^{200,202}Fr isotopes". The INTC-030 (Feb, 2008) wrote: "...Committee decided to recommend to the Research Board the approval of 18 shifts for the investigation of ^{178,180}Tl. ***For the neutron-deficient Fr nuclides of interest further yield checks should be performed and the Committee suggests that a new proposal is submitted for these cases once the experiments on the Tl isotopes have been completed***".

By now, two experiments (IS466-1 [1] and IS466-2 [2]) to identify and study βDF of ^{178,180,182}Tl were successfully performed (see, also [3]). Therefore, we wish to proceed to the βDF studies of Fr isotopes.

The goals of the present addendum are threefold.

- First of all, we will perform a search for the βDF of ^{204}Fr and a detailed β-delayed fission study of ^{202,204}Fr. We expect that for both isotopes the coincidence fission fragment measurements will be carried out, thus establishing the mass split of their daughter (after β decay) products ^{202,204}Rn. More generally, these studies will also supply unique near-barrier or even sub-barrier low-energy fission data for the region of nuclei, which do not decay by spontaneous fission. (12 shifts requested)
- Another important goal, which comes as a "by-product"(no extra beam time is required), is a search for the low-lying 0^+ intruder bandheads and states on top of them in the daughter isotopes ^{202,204}Rn, populated by β decay of ^{202,204}Fr. Simultaneously, extensive α–γ coincidence data will be collected for α decay of ^{202,204}Fr, which will allow studies of excited states in the daughter isotopes ^{198,200}At.
- Finally, the yield and background checks for the *most neutron-rich* Fr isotope presently known - ^{232}Fr and for the new isotope ^{234}Fr will be performed. These two isotopes are promising candidates for the βDF studies in the extremely *neutron-rich* nuclei, relevant for the r-process and its termination by fission (1 shift is requested)

The uniqueness of ISOLDE for this kind of studies is that it provides pure beams of both neutron-deficient and neutron-rich Fr isotopes with intensities not accessible anywhere else.

Requested shifts: 13 shifts of ISOLDE beam time are requested for the whole project.

1. General features of β-delayed fission.

As a detailed description of βDF was given in the IS466 [1] and IS466-2 [2] proposals, only a short reminder is provided here. βDF is a rare nuclear decay process in which a parent nucleus first undergoes β decay, populating excited states in the daughter nucleus, which then may fission with some probability (Fig.1). βDF is of special interest because it allows to study the fission properties (e.g. decay probability, fission barrier height, mass/charge distribution, total kinetic energy, gamma and neutron multiplicities) of exotic daughter nuclei which possess a very low (at present, un-measurable) spontaneous fission branch. The βDF is also believed to play an important role in the r-process [4] (e.g. production of heavy elements, termination of the r-process, "fission recycling").

βDF is expected to occur with a detectable probability when the Q_{EC}(A,Z) value of the *parent* nucleus is comparable with or greater than the fission barrier B_f(A,Z-1) of the *daughter* nuclide, see Fig.1. Then a certain branch of the parent β^+/EC decay can populate relatively high-lying excited states in the vicinity of the top of the fission barrier of the daughter nucleus which possess large fission widths (fission actually happens in competition with gamma decay). It is important to stress that the excitation energy of the fissioning daughter nucleus is limited by the available Q_{EC}(A,Z), therefore βDF provides unique fission data at low excitation energy, at which the topology of the energy surface around the fission barrier and shell effects might play a very important role.

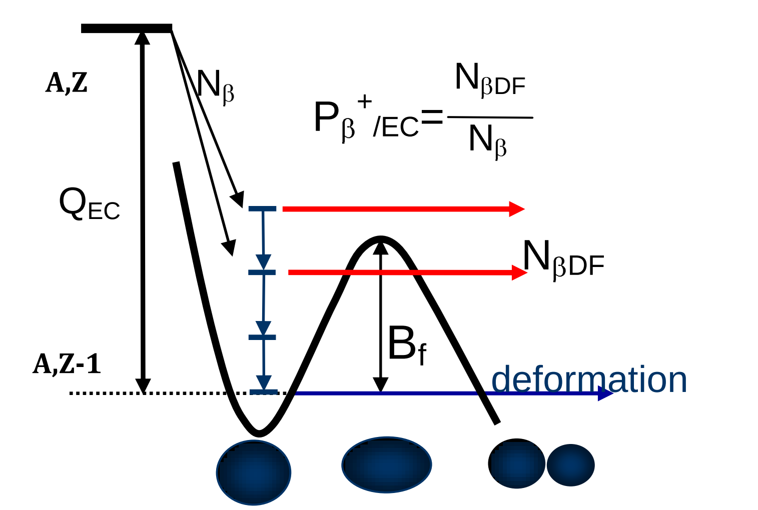

Fig.1. Schematic diagram of potential energy versus deformation for the βDF process. The parent nucleus (A,Z) undergoes β^+/EC decay and populates excited states in the daughter (A,Z-1) nucleus, which might fission.

In our first experiment IS466 (June, 2008 [1]), an unambiguous identification of the βDF of ^{180}Tl was achieved and a surprising asymmetric mass distribution of the fission fragments of the daughter ^{180}Hg was observed, see Fig.2 [3]. This is in contrast to the symmetrical split into two semi-magic ^{90}Zr nuclei, expected before the experiment.

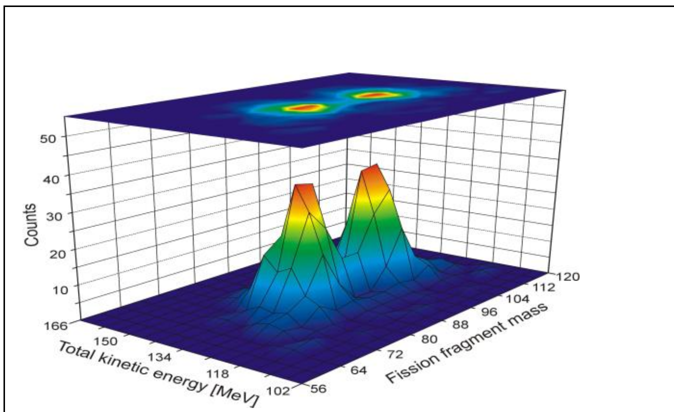

Fig.2 The derived fission-fragment distribution of ^{180}Hg as a function of the fragment mass and the total kinetic energy [3].

2. Selected results from the IS466-2 experiment at GPS, July 2010 [5,6]

The successful study of ^{180}Tl in the IS466 run was followed by the Addendum IS466-2 [2]. The goal of the IS466-2 experiment was a search for βDF of the isotopes ^{178,182}Tl; the experiment happened in July 2010. Below, some preliminary results are outlined [5,6].

a) Identification of βDF of ^{178}Tl ($T_{1/2}$~250 ms)

The βDF of ^{178}Tl was observed for the first time (7 events in singles, ~0.3 ff/h) and a (preliminary) value of the βDF probability $P_{\beta DF}(^{178}Tl)=1.4(7)\times10^{-4}$ was deduced [5]. (For comparison, $P_{\beta DF}(^{180}Tl)=3.6(7)\times10^{-5}$ [3]). With the limited statistics available, the observed apparent broadness of the fission fragments energy distribution *in singles* (see also below for ^{202}Fr) hints that the mass distribution is also asymmetrical as in the case of the βDF of ^{180}Tl.

b) Search for βDF of ^{182}Tl

A dedicated search for βDF of ^{182}Tl was also undertaken in this run. No fission fragments were observed, which results in a (preliminary) upper limit of $P_{\beta DF}(^{182}Tl)<1.4\times10^{-6}$ [4]. One should mention that according to our expectations, based on the extrapolation from ^{180}Tl and on known systematics of the $P_{\beta DF}$ values as a function of the difference Q_{EC}(Parent)-B_f(Daughter), a value of $P_{\beta DF}(^{182}Tl)\sim8\times10^{-6}$ was expected. One of the reasons for the non-observation of βDF of ^{182}Tl could be a dominant β-decay feeding towards low-lying states in ^{182}Hg, which could be significantly below the top of the (calculated) fission barrier of B_f=10.85 MeV [7]. This issue is presently addressed by a dedicated analysis of the complementary β-decay data of ^{182}Tl by the Leuven group [8], see also item d) below.

c) Identification of βDF of ^{202}Fr.

Following the recommendation of the INTC to investigate the feasibility to study the βDF of the francium isotopes, we collected a sample of ^{202}Fr for about 18 h in *parasitic* mode. During this short test, we were able to clearly identify the βDF of ^{202}Fr (34 fission fragments in singles, or ~1.9 ff per hour). The (very preliminary) value of $P_{\beta DF}(^{202}Fr)\sim2(1)\times10^{-4}$ fits quite well to the known systematics of the $P_{\beta DF}$ values as a function of the difference Q_{EC}(Parent)-B_f(Daughter), see Fig. 3.

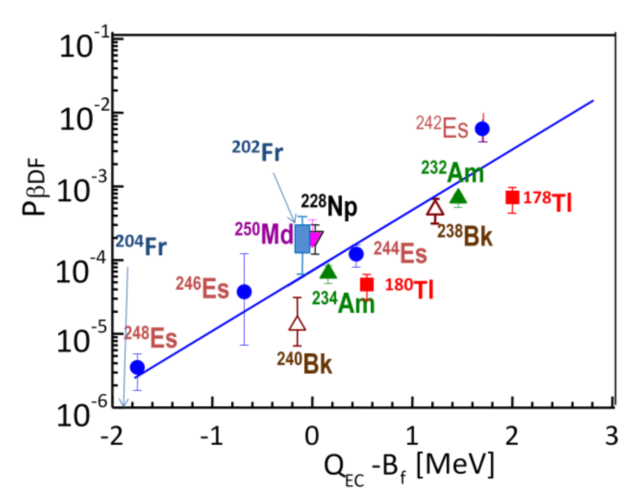

Fig.3 βDF probability $P_{\beta df}$ as a function of the difference Q_{EC}(Parent)-B_f(Daughter). 11 previously known cases in the U region are shown, along with our new data from ISOLDE measurements: ^{180}Tl [3] and (preliminary) data for ^{178}Tl and ^{202}Fr [5]. An estimate of $P_{\beta df}(^{204}Fr)\sim1\times10^{-6}$ can be deduced from this plot, which is approximately two orders of magnitude lower than for ^{202}Fr (to be used for the fission rate estimate for ^{204}Fr later in this proposal)

Therefore, one of the goals of the present proposal is to measure coincident fission fragments for βDF of ^{202}Fr.

d) Dedicated β-decay studies of ^{178,182}Tl.

Before our experiments, no β-decay studies have been done for ^{178,182}Tl. Due to high statistics, collected in our experiment, the detailed β-decay analysis for ^{178,182}Tl is now possible [5,6]. This allows us to search for low-lying coexisting configurations (including for presently unknown 0^+ intruder band-heads) in the daughter ^{178}Hg by using electron-γ coincidences between the electrons measured in the Si detector and gammas in the Ge detectors. The application of this method has been clearly demonstrated by a successful identification of the long-sought 0^+ intruder band-head in ^{180}Hg in our βDF study of ^{180}Tl in IS466 [8].

3. Summary of physics goals of the present Addendum

a) Coincident fission fragment measurements ^{202}Fr (8 shifts requested)

Based on the measured counting rate of ~1.9 ff/h for fission fragments in singles in IS466-2 run, we expect ~0.6 coincident fission fragment pairs per hour for ^{202}Fr (due to Si-Si coincidence efficiency, see [3] and Fig.5). Since, based on the data for ^{180}Tl from the IS466 run, observation of ~40 coincident fragments is sufficient to demonstrate the symmetry (or asymmetry) of the fission mass distribution, a measurement time of 64 hours (8 shifts) is required for ^{202}Fr. The measurements of the mass split in the βDF of this nucleus, apart from providing otherwise in-accessible low-energy fission data, will also allow to further check the predictions of the fission model [9], used in our work [3] to describe the observed mass asymmetry of ^{180}Hg.

b) Search for βDF of ^{204}Fr and determination of the fission fragment mass distribution of ^{204}Rn (4 shifts requested)

According to systematics of $P_{\beta DF}$ as a function of Q_{EC}(Parent)-B_f(Daughter) in Fig.3, the probability for the βDF of ^{204}Fr should be approximately two orders of magnitude lower than that for ^{202}Fr. On the other hand, the measured ISOLDE yield [10] for ^{204}Fr is at least three orders of magnitude higher than that for ^{202}Fr. Therefore, the expected fission rate of ^{204}Fr could be up to an order of magnitude higher than in case of ^{202}Fr. Thus, we expect not only to identify the βDF of ^{204}Fr, but also to measure the energy (thus, mass) distribution of fission fragments of its daughter ^{204}Rn.

For the particular case of ^{204}Rn fission, low-energy fission data are available from an earlier experiment at FRS(GSI) by using electromagnetically-excited fission of radioactive beams [11]. These data showed that, the charge split (thus, the mass split) of ^{204}Rn was *symmetric* (see Fig.20 of [11]). The excitation energy (E*(^{204}Rn)) in the FRS study showed a rather broad distrubtion, centered around ~11 MeV (Fig.15 of [11]); the distribution is overall much higher in energy than in our planned βDF study of ^{204}Fr. Indeed, the *maximal* excitation energy of ^{204}Rn in our study cannot exceed E*(^{204}Rn)=Q_{EC}(^{204}Fr)=8.72 MeV, which is limited by the available (calculated [12]) Q_{EC} value of the parent isotope ^{204}Fr. The comparison of these excitation energies with the calculated fission barrier of ^{204}Rn [7], shows that in the Coulex-excited fission of ^{204}Rn the fission is near-the barrier [11], while in our case it is sub-barrier. This difference could also be reflected in the resulting mass distributions of ^{204}Rn produced by two different methods.

In this respect, a valuable comparison with our recent study of βDF of ^{180}Tl [3] is in order here. As shown in [3], two fission valleys exist in ^{180}Hg (see Fig.4), with the entry to the asymmetric one being below by a few MeV than the entry to the symmetric one. It is suggested that due to this subtle effect, and despite a gain by ~14 MeV in the Q-value, which would happen if ^{180}Hg fissioned into two semi-magic ^{90}Zr fragments, we observed an asymmetric split in predominantly ^{100}Ru and ^{80}Kr [3]. A similar effect can also happen in case of ^{204}Rn.

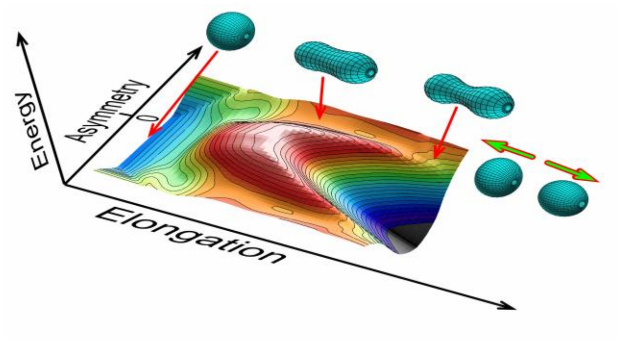

Fig.4 A schematic representation of the potential-energy surface for ^{180}Hg in two dimensions (elongation and asymmetry). The shapes shown, connected by arrows to their locations, are the ground state, the saddle point, and the point where the asymmetric valley disappears. Taken from [3].

The prediction of such subtle interplay between complex macro- and microscopic effects in such exotic nuclei is very difficult [7,9] and requires also experimental input, including the excitation energy dependence of all these effects. Exactly such kind of data can be provided by our studies, being complementary to those performed at FRS, but at a somewhat higher excitation energy.

c) Yield and background estimates for neutron-rich isotopes ^{232,234}Fr (1 shift is requested)

According to the modern approaches, see e.g. [4], the beta-delayed fission (along with the n-induced and spontaneous fission) might play an important role in the so-called r-process termination by fission. Unfortunately, presently, extremely neutron-rich heavy nuclei along the expected r-process path are not accessible for the experimental studies. Nevertheless, according to our semi-phenomenological estimates based on Fig.3, some of the presently accessible nuclei, e.g. the heaviest known Fr isotope - ^{232}Fr, can possess a measurable βDF branch. Earlier, a yield of ~3×10^3 ions/μC for ^{232}Fr was measured in the study [13]. To put our future program in the neutron-rich region on a solid footing, we would like both to confirm this value and to check background conditions before proceeding with a dedicated proposal for a βDF study of this isotope.
Furthermore, we also wish to perform similar tests for the new isotope ^{234}Fr, for which even higher βDF probability is expected.

These measurements are straightforward and will be done by using Ge-detectors of the WM system, or, possibly, by installing a simple tape system, including beta and germanium detectors.

d) Detailed α/β-decay studies of ^{202,204}Fr (no additional shifts requested)

As a ‘by-product’ of the βDF studies, large statistics for α and β decays of ^{202,204}Fr will be collected, several orders of magnitude higher than in any previous experiments. Furthermore, both due to the high purity of the ^{202,204}Fr beams and the use of the

Wind-mill setup, which allows efficient measurements of α,β, conversion electron and γ decays, rich decay data will be obtained both for the parent ^{202,204}Fr isotopes, and also for their daughter nuclei ^{198,200}At (after α decay) and ^{202,204}Rn (after β decay). In particular, the search for the low-lying 0^+ intruder bandheads and the members of the corresponding bands built on top of them will be possible, which is important for the research on shape coexistence phenomena in this region of nuclei. As such, our studies will also provide important complementary information for the recent Coulex experiments for ^{202,204}Rn performed with REX-ISOLDE.

4. Detection setup for the Present Proposal

The experimental set-up to be used in the proposed study of ^{202,204}Fr is shown in Fig. 5. We will exploit the 'Wind-mill' system, which was successfully used in IS466 (also in IS387, IS407, IS456, I-086) and which allows the coincident fission fragments to be measured [3]. The ISOLDE beam is implanted in the carbon foil, which is surrounded by 2 Si detectors for α and fission decay measurements. The fission fragments are measured both as single events and in coincidence to each other. The implantation and simultaneous measurement are performed in cycles of a few seconds in duration (depends on the half-life of the nuclide). After end of the cycle, the wind-mill rotates and a "fresh" foil is introduced for the implantation. The whole setup will be surrounded by the Ge-detectors of Miniball to allow measurements of fission fragments (FF) in coincidence with gammas.

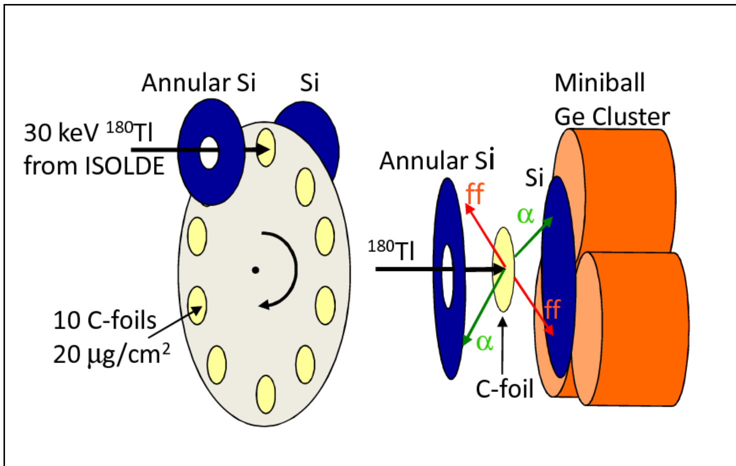

Fig. 5 The "wind-mill" system [3] for the βDF measurements with 10 thin (~20 μg/cm^2) carbon foils mounted (general view and a zoom of the foil-Si detectors arrangement). Note, that in the previous run (IS466-2), no annular Si detector was used, that is why no coincident fission fragments could be measured. In the present Proposal, we will use both Si detectors, so the coincident events will be measured.

We also foresee to calibrate prior to the ISOLDE run all our detectors with mass- and energy-separated beams from the fission fragment separator LOHENGRIN at ILL Grenoble to fully correct for dead layer and pulse height defect and deduce thus absolute fission fragment energies and TKE (total kinetic energy) respectively.

5. Summary of requested shifts:

In total, we request 13 shifts of ISOLDE beam time:

- **8 shifts for ^{202}Fr**
- **4 shifts for ^{204}Fr**
- **1 shift for the yield and background checks for the isotopes ^{232,234}Fr**

We also want to notice here, that the detection system and the DAQ used in this experiment are the same as necessary for the continuation of the At RILIS development (initiated by the LoI I-086, spokespersons: A. Andreyev and V. Fedosseev). In November 2010, our collaboration successfully completed the first phase of the At RILIS development (see a separate report to the INTC). This work is planned to be continued in 2011. Therefore, if the beam were granted for the Fr studies, it would be preferable to combine it with the At RILIS development program. In this case, one would start with the Fr run, while the lasers for At ionization are being prepared. After the finishing of the Fr part, the At RILIS development would follow by using the detection and DAQ systems from the Fr run.

References:

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Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT

The experimental setup comprises: *a Wind-Mill system with 2-4 Si detectors inside, and 1-2 Ge detectors outside. WM system was successfully used in the runs IS387, IS407, IS456, IS466 and I-086, therefore solid understanding of all possible hazards is available.*

Part of the Choose an item.	Availability	Design and manufacturing
Windmill	☒ Existing	☒ Used in several previous experiments, e.g. IS387,IS407, IS456, IS466, I-086 ☐ To be modified
	☐ New	☐ Standard equipment supplied by a manufacturer ☐ CERN/collaboration responsible for the design and/or manufacturing

HAZARDS GENERATED BY THE EXPERIMENT:

No 'special' hazards is expected (see also the table below)

Additional hazards:

Hazards	*Wind Mill*	*[Part 2 of the experiment/equipment]*	*[Part 3 of the experiment/equipment]*
Thermodynamic and fluidic			
Pressure	-		
Vacuum	Usual vacuum of ISOLDE		
Temperature	-		
Heat transfer	-		
Thermal properties of materials	-		
Cryogenic fluid	LN2 for Ge detectors (150 l)		
Electrical and electromagnetic			
Electricity	Usual power suppliers		
Static electricity	-		
Magnetic field	-		
Batteries	☐		
Capacitors	☐		
Ionizing radiation			
Target material	The C foils where the radioactive samples are implanted are very fragile. Should they break upon opening the Windmill, the pieces are so light that they would become airborne. Great care must be taken when opening the system and removing them (slow pumping/venting protective equipment: facial mask).		
Beam particle type (e, p, ions,	-		
Beam intensity	-		
Beam energy	-		

Cooling liquids	-		
Gases	-		
Calibration sources:	☒		
• Open source	☒		
• Sealed source	☐ [ISO standard]		
• Isotope	239Pu, 241Am, 244Cm		
• Activity	1 kBq each		
Use of activated material:	-		
• Description	☐		
• Dose rate on contact and in 10 cm distance	-		
• Isotope	-		
• Activity	-		
Non-ionizing radiation			
Laser	-		
UV light	-		
Microwaves (300MHz-30 GHz)	-		
Radiofrequency (1-300MHz)	-		
Chemical			
Toxic	Pb shielding (~20 bricks)		
Harmful	-		
CMR (carcinogens, mutagens and substances toxic to reproduction)	-		
Corrosive	-		
Irritant	-		
Flammable	-		
Oxidizing	-		
Explosiveness	-		
Asphyxiant	-		
Dangerous for the environment	-		
Mechanical			
Physical impact or mechanical energy (moving parts)	The chamber is heavy and needs to be handled with care during installation/ removing.		
Mechanical properties (Sharp, rough, slippery)	-		
Vibration	-		
Vehicles and Means of Transport	-		
Noise			
Frequency	-		
Intensity	-		
Physical			
Confined spaces	-		
High workplaces	-		
Access to high workplaces	-		
Obstructions in passageways	-		
Manual handling	-		
Poor ergonomics	-		

0.1 Hazard identification

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