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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

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In our recent experiment (March 2007) at the velocity filter SHIP(GSI) we observed the electron-capture delayed fission of the odd-odd isotope ^{194}At. This is the first unambiguous identification of this phenomenon in the very neutron-deficient nuclei in the vicinity of the proton shell closure at Z=82. In addition, the total kinetic energy (TKE) for the daughter nuclide ^{194}Po was measured, despite the fact that this isotope does not decay via spontaneous fission. Semi-empirical analysis of the electron-capture Q_{EC} values and fission barriers B_f shows that a relatively broad island of ECDF must exist in this region of the Nuclide Chart, with some of the nuclei having unusually high ECDF probabilities.

Therefore, this Proposal is intended to initiate the systematic identification and study of beta delayed fission at ISOLDE in the very neutron-deficient lead region. Our aim is to provide unique ***low-energy fission data*** *(e.g. probabilities, TKE release, fission barriers and their isospin dependence, mass/charge distribution of fragments, neutron and gamma multiplicities) for the region of the nuclei, which do not decay by spontaneous fission.* More generally, the beta-delayed fission is believed to play an important role in the r-process (e.g. production of heavy elements, termination of the r-process, “fission recycling”.

As a first step in the program we propose to study ^{178,180}Tl and ^{200,202}Fr isotopes, for which ISOLDE can provide pure beams with the intensities not accessible anywhere else.

I. General features of the electron-capture delayed fission (ECDF).

Beta-delayed fission, and in particular, electron-capture delayed fission, discovered in 1966 in Dubna for the isotopes ^{232,234}Am [1,2], is a rare nuclear decay process in which a parent nucleus first undergoes beta decay, populating excited states in the daughter nucleus, which then may fission with some probability (Fig.1). Such beta-delayed fission 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 (in practice – unmeasurable) spontaneous fission branch. The beta-delayed fission is also believed to play an important role in the r-process [3,4] (e.g. production of heavy elements, termination of the r-process, “fission recycling”).

ECDF is expected to occur with a detectable probability when the total 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 EC decay can populate relatively high-lying excited states in the daughter nucleus which possess large fission widths (fission actually happens in competition with the gamma decay) [5]. It is important to stress that the excitation energy of the fissioning daughter nucleus is typically a few MeV only, therefore ECDF provides unique fission data at low excitation energy, in which the shell effects might play a very important role.

The probability of the ECDF, P_{ECDF} is defined [2,6-8] as the ratio of the number of EC events resulting in fission, N_{ECDF}, to the total number of EC decays, N_{EC}:

$$P_{ECDF}=\frac{N_{ECDF}}{N_{EC}}=\frac{\int_0^{Q_{EC}} f(Q_{EC}-E)S_\beta(E)\frac{\Gamma_f}{\Gamma_f+\Gamma_\gamma}(E)dE}{\int_0^{Q_{EC}} f(Q_{EC}-E)S_\beta(E)dE}$$

, where the product of the integrated Fermi function $f(Q_{EC}-E)$ and the beta-strength function $S_\beta(E)$ accounts for the population of excited states in the daughter nucleus, while the ratio $\Gamma_f/(\Gamma_f+\Gamma_\gamma)$ describes the competition between γ-cascades leading to the ground state (gamma width Γ_γ) and fission (fission width Γ_f). The very strong energy dependence of P_{ECDF} (see below) stems from the exponential variation of the fission width Γ_f for the sub-barrier fission, which allows to deduce the fission barrier B_f provided the P_{ECDF} value is measured [6-8].

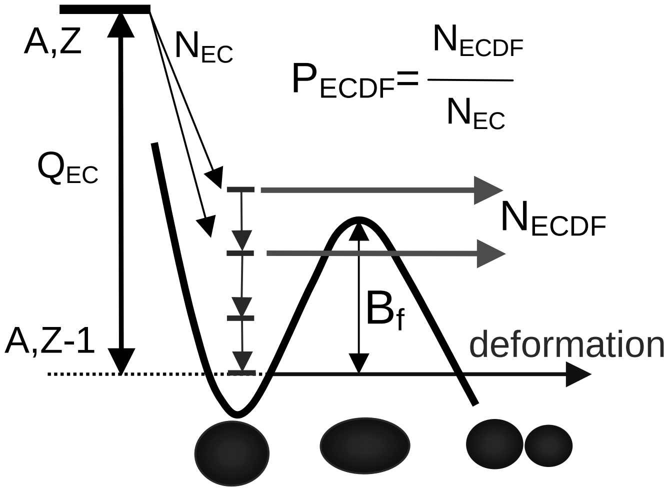

Fig.1(in color). Schematic diagram of potential energy versus deformation for the ECDF process. The parent nucleus (A,Z) undergoes EC decay and populates excited states in the daughter nucleus, which might fission (in a competition with γ-transitions toward the ground state).

So far, most of the experimentally know fission data (in particular, B_f values) are for nuclei in the vicinity of the beta-stability line. In this respect, the fission data for very exotic nuclei (both neutron-deficient and neutron-rich) are of crucial importance for the long-standing discussion of the isospin dependence of the fission barriers and the observed large deviation of the calculated and experimentally deduced fission barriers by moving away from the valley of stability (see, e.g. [9,10]). Indeed, as was shown by a large number of experimental studies (including our work at SHIP for Bi-Po isotopes [9]), a decrease of the calculated B_f values by up to 30-40% is required in order to reproduce the measured fusion-evaporation cross-section values in a broad region of neutron-deficient nuclei with A>150. ***Our experiments will provide unique low-energy fission data for the very neutron-deficient nuclei in the lead region, which will allow investigation of the isospin dependence of the fission barriers in more details.***

Another important issue in the ECDF studies is the mass/charge distribution of the fission fragments from very exotic (in terms of the N/Z ratio) fissioning nuclei. As an example, the typical N/Z ratio for nuclei in the vicinity of the beta-stability valley is ~1.59 (^{238}U) or ~1.54 (^{208}Pb), while for ^{194}At studied in our SHIP experiment the ratio is N/Z=1.31. The ratio is even lower (N/Z=1.25) for ^{180}Tl being one of the goals of this Proposal. ***Clearly, such unusual ratio might lead to completely unexpected mass/charge distributions in comparison to fission of the less exotic nuclei.*** It is indeed well known that fission of the common heavier nuclei is mostly dominated by asymmetric fission, in which the Z=50 and N=82 shell closures play an important role (leading to heavy fragments in the vicinity of ^{132}Sn). For the lighter nuclei which fission via ECDF on the other hand, we expect symmetric fission to prevail. In the case of the ECDF of ^{178,180}Tl e.g., leading to low-energy fission of the daughters ^{178}Hg(Z=80,N=98) and ^{180}Hg(Z=80,N=100), we expect symmetric fission dominated by the N=50 shell closure resulting in fission fragments in the vicinity of (N=50,Z=40), hence conserving the N/Z-ratio of the fissioning nucleus. Due to the low excitation energy of these fissioning systems, shell effects will indeed not be washed out, and we wonder if a double magic asymmetric fission component formed by very neutron-deficient ^{100}Sn(Z=50) and very neutron-rich ^{78}Ni(Z=28) will also be observed.

Since the discovery of ECDF, a number of experiments has been performed (see recent references in e.g. [2,11]), but so far only 11 cases of ECDF are known, all of them, with the exception of the yet unconfirmed ^{180}Tl [5], being in the trans-uranium region. Fig.2 summarizes the measured data by showing the P_{ECDF} data as a function of the difference Q_{EC}-B_f (Fig.2b). The experimental P_{ECDF} values are typically quite low (~10^{-7}-10^{-2}), which suggests the sub-barrier nature of the process in the studied actinide nuclei. Furthermore, the P_{ECDF} value exponentially increases with the increase of the Q_{EC}-B_f difference (with the notable deviation from this rule in ^{180}Tl, see below). This strong sensitivity of P_{ECDF} to the Q_{EC}-B_f value is the reason why the ECDF studies are considered as a very promising method to deduce fission barriers of very exotic nuclei [6-8], which so far are un-accessible for fission studies by other presently available methods. As an example, by using the ECDF of ^{232}Am, the fission barrier of its daughter nuclide ^{232}Pu was deduced with the precision of ~7.5% (B_f=5.3(4) MeV) [8], which is well comparable to the precision of traditional 'direct' methods for the determination of the fission barriers.

We also note that the relatively low-energy (E*~11 MeV) electromagnetically-induced fission of the secondary fragments from the projectile fragmentation of ^{238}U is another promising method to study the fission in this region [10]. However, the presently available secondary ion beam intensities do not allow the studies of extreme neutron-deficient nuclei as achieved by ECDF.

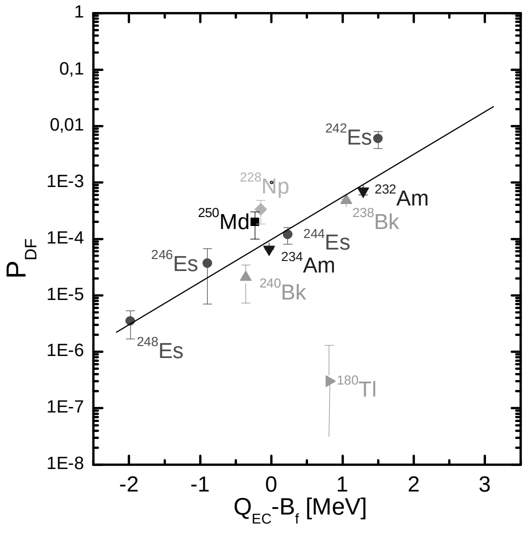

Fig.2 (in color). ECDF probability [1,2,8,11] as a function of the Q_{EC}-B_f value (B_f is the fission barrier of the daughter). The data are marked by the parent odd-odd nuclei. The only reported (unconfirmed) data in the lead region is $P_{ECDF}(^{180}\mathrm{Tl})=3\times10^{-(7\pm1)}$ [5,12]. The Q_{EC} and B_f values are taken from calculations [13] and [14], respectively, see also Fig.3.

Another observation, which is useful for the following discussion and for the whole proposed program is that in all these cases the odd-odd (thus, even-A) parent nuclide decays to the even-even daughter. This is readily understood due to two main reasons. First of all, due to the odd-even effect in the masses, an odd-odd parent nuclide possesses a higher Q_{EC} value in comparison with its odd-A neighbors (cf. Fig.3 for the Pb region). Secondly, due to the so-called 'specialization' energy, an even-even daughter nuclide has usually a lower fission barrier in comparison with its odd-A neighbors. Both these factors substantially increase the P_{ECDF} value for the odd-odd parent nuclei and so far no data for the odd-A nuclei have been measured.

II. ECDF in the lead region: earlier results

The ECDF phenomenon is also expected in the lead region, see Fig.3, which shows calculated data on the Q_{EC} [13] and B_f [14] for the parent-daughter pairs Tl-Hg, Bi-Pb, At-Po and Fr-Rn. Based on the above-mentioned arguments we are interested in the lightest odd-odd parent nuclides of Tl, Bi, At and Fr for which $Q_{EC}(A,Z)-B_f(A,Z-1)>0$ and the $b(_{\beta}+/_{EC})>1\%$.

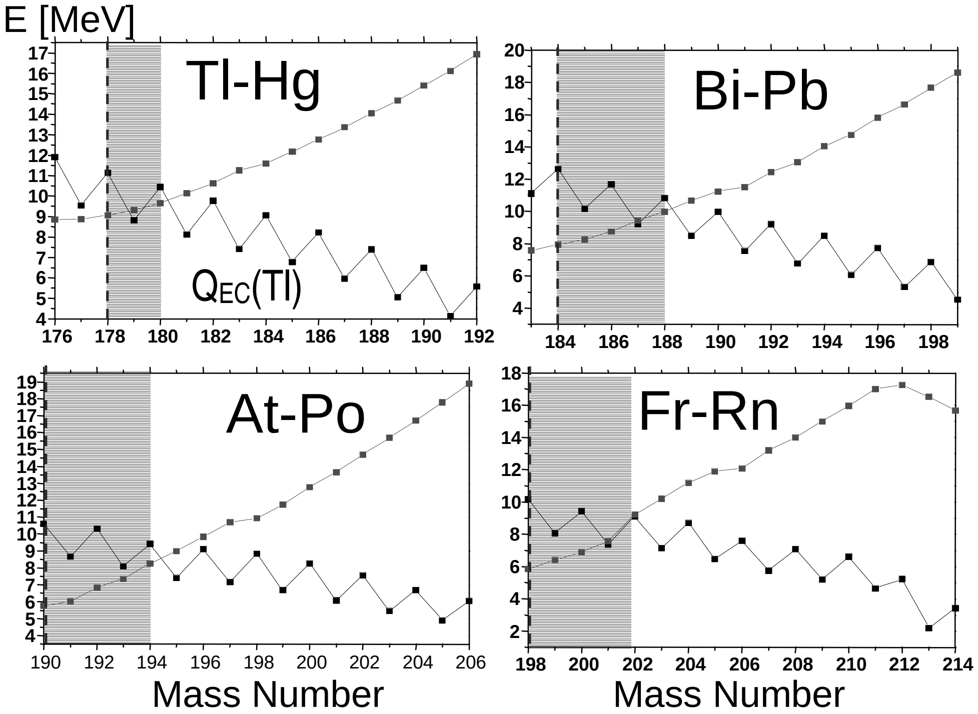

Fig. 3 (in color). Calculated $Q_{EC}(A,Z)$ (black points, [13]) and $B_f(A,Z-1)$ (red points, [14]). The regions of our interest are shown in blue color. The left border of these regions is determined by $b_{EC}<1\%$. The following nuclei are expected to have measurable P_{ECDF} values: ^{178,180}Tl, $^{184-188}$Bi, $^{192-194,196}$At and $^{200-202}$Fr.

Actually, experiments in the lead region have already been performed in Dubna two decades ago and in total three ECDF candidates have been suggested: ^{180}Tl, ^{196}At and ^{188}Bi [5,12]. Unfortunately, an unselective method used in the experiments did not allow an unambiguous identification of A and Z of the produced nuclei. Therefore, only the *most probable* fissioning nucleus was suggested based on the measured half-life along with the large number of cross-irradiations with different projectile-target combinations. As a result, ^{180}Tl was suggested as the most probable candidate for the ECDF activity observed in the $^{40}Ca+^{144}Sm\rightarrow^{180}Tl+p3n$ reaction, and the lowest ever value of $P_{ECDF}(^{180}Tl)=3\times10^{-(7\pm1)}$ was deduced [12], see Fig.2. The value was deduced by comparing the number of observed fission-like events in the mica detectors to the *estimated* (see below) number of ^{180}Tl nuclei produced in the thick (in terms of the excitation function) ^{144}Sm target, which was also not pure (88% enrichment). The measured P_{ECDF} value does not fit to the P_{ECDF} systematics in Fig.2, but two possible reasons have been suggested in [5], such as different shape of the fission barrier in the actinides compared to the lead region and the possible presence of the beta-delayed proton and alpha emission in the very neutron-deficient isotopes in the lead region.

However, we believe that the quoted value $P_{ECDF}(^{180}Tl)$ is strongly underestimated. This conclusion is based on the following arguments. The absolute production cross-section of ^{180}Tl in the $^{40}Ca+^{144}Sm\rightarrow^{180}Tl+p3n$ reaction was not known 20 years ago and the authors of [5,12] ***assumed*** a value of $\sigma(^{180}Tl)=0.1-1$ mb. However, since then reliable experimental data became available for this region of nuclei and now we estimate the cross-section for this reaction as $\sigma(^{180}Tl)=2$ μb, thus a factor 50-500 lower than assumed in [5,12]. Based on this, the recalculated value should be $P_{ECDF}(^{180}Tl)= 1.5\times10^{-5}-1.5\times10^{-4}$. This value would also fit perfectly to the systematics of P_{ECDF} versus $Q_{EC}-B_f$ value in Fig. 2(right panel). The higher P_{ECDF} value and a relatively high yield of ^{180}Tl at ISOLDE will allow us to collect at least a few thousands decays (see below), which should be enough for detailed studies such as mass distribution and identification of fission fragments, TKE dependence on the mass split, fission-gamma rays (and X rays) coincidences.

To conclude this section we mention that for two other ECDF candidates - ^{188}Bi and ^{196}At no P_{ECDF} values have been deduced in [5,12]. Unfortunately, none of these data, including ECDF of ^{180}Tl, have ever been confirmed.

III. ECDF in the lead region: unambiguous identification of ECDF of ^{194}At at SHIP [15].

In view of the weakness of the previously reported Dubna results on one hand and a wealth of data which can be achieved on the other hand, we decided to initiate a dedicated experimental program at SHIP (GSI) with the aim to provide unambiguous identification and detailed studies of the ECDF in the At nuclei. The At nuclei are presently not easily accessible at ISOLDE as a dedicated negative ion source is required (we will ask for its development).

In our short pilot experiment (in total 2.5 days, including excitation function measurements) in March 2007 at SHIP, we have chosen the $^{56}Fe+^{141}Pr\rightarrow^{194}At+3n$ reaction because ^{194}At is expected to have a quite large positive difference $Q_{EC}-B_f=1.5$ MeV, which should lead to a high P_{ECDF} value. Furthermore, ^{141}Pr has a natural 100% abundance, which greatly facilitates the reaction channel identification. Finally, by using a recoil separator, a clearer reaction channel selection can be achieved. Fig. 4(left, top panel) shows the total spectrum in the PSSD, in which we observed $\sim86\times10^3$ nuclei of ^{194}At. Fig 4(left, bottom panel) shows the same spectrum, but collected within 15 ms of the ‘beam off’ interval (the beam time structure was: 5 ms “beam on”/15 ms “beam off”). The 66 events in the energy range of 90-160 MeV were attributed to fission events resulting from the ECDF of ^{194}At [15].

This is based on a) the excitation function for these events, which follows very well the one for alpha decays of ^{194}At; b) the half-life behavior: the fission half-life value of $T_{1/2,f}=300(60)$ ms is in a good agreement with $T_{1/2,\alpha}(^{194}At)=280(20)$ ms; c) approximately half of these events are double-fold events, with the coincident signal measured in the BOX detector, which is consistent with the expected efficiency of this detector; d) most of these events are in coincidence with gammas in the Clover detectors, as it is expected for the fission fragments due to the high gamma-multiplicity in fission [16].

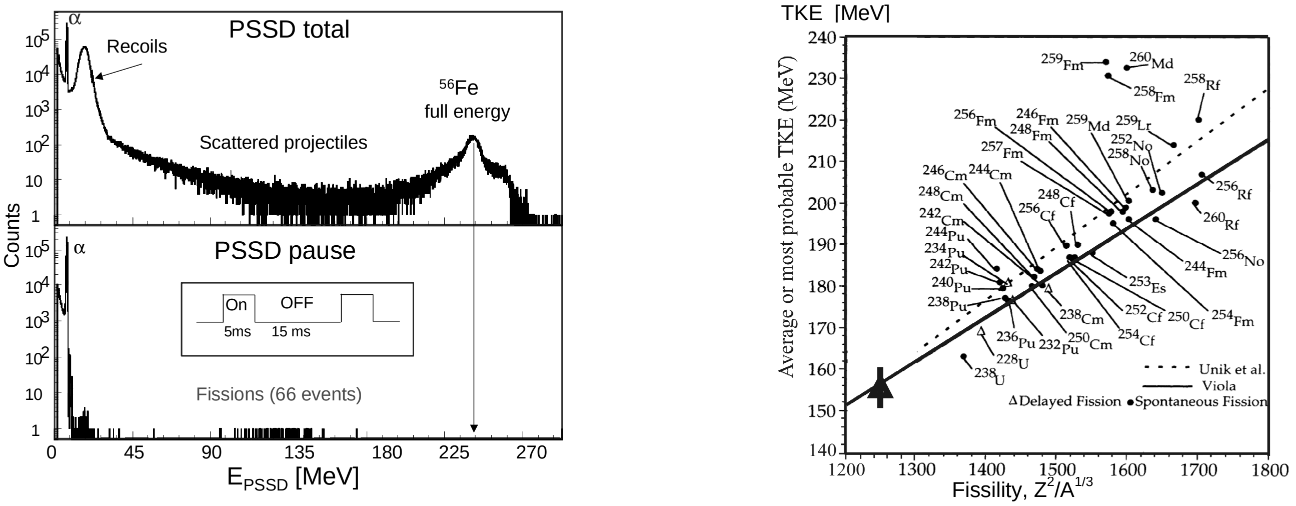

Fig. 4 Identification of ECDF in ^{194}At. Left panel: top) Total spectrum in the PSSD. Shown are (from right to left): low-intensity "leaky" full-energy ^{56}Fe beam particles, lower-energy scattered ^{56}Fe ions, recoils from the reaction (E_{rec}~20 MeV) and α decays of the reaction products. Bottom part: The same spectrum, measured during the "beam off" interval. Only decay particles are expected in the spectrum. 66 fission events with the energy of ~90-160 MeV are shown in red color. Right panel: Viola-plot for TKE values. Our value of TKE(^{194}Po)=157(15) MeV is shown by the blue triangle.

Taken together, all these data provide unambiguous identification of these events as proceeding from the ECDF of ^{194}At. Furthermore, by adding up the energy measured in the PSSD and in the BOX detectors (for coincident events only), we performed the first determination of the total kinetic energy for ^{194}Po: TKE(^{194}Po)=157(15) MeV. This value fits well to the Viola-systematics, see right panel of Fig.4.

These results are consistent with our expectations for the occurrence of the ECDF nuclei based on Fig. 3, which gives us the confidence that the ECDF indeed exists in the nuclei suggested for studies in this Proposal.

IV. Identification of new ECDF cases and their systematical studies

From the simple analysis of the Q_{EC}-B_f differences in Fig. 3 (see also Table 1), we expect that the following nuclides should have a measurable P_{ECDF} branching: $^{178\text{-}180}$Tl, $^{184\text{-}188}$Bi, $^{192\text{-}194,196}$At and $^{200\text{-}202}$Fr. Therefore, given our convincing data for ^{194}At and preliminary data for the ECDF candidate nuclei ^{196}At and ^{186}Bi, we propose to start a systematical identification and study of ECDF in the above-mentioned nuclei and to perform a detailed studies of their properties. This will include the following data for each nucleus:

a) P_{ECDF} value, from which the fission barrier B_f will be deduced.
b) Individual energies of the fission fragments (thus, also TKE energy), from which the mass distribution will be deduced.
c) Gamma and neutron multiplicities
d) In most cases we will be able to deduce the EC branch, b_{EC}, directly from our data (the b_{EC} branch is required for the P_{ECDF} determination). This can be done, e.g. in case of ^{180}Tl, by using the parent-daughter relationship and comparing α-decay rates of ^{180}Tl and its daughter (after α decay) ^{176}Au (after corrections for the known b_α(^{176}Au) and recoil effects from the implantation foil). Alternatively, we can compare α-decays of ^{180}Tl and its

daughter ^{180}Hg (after the EC decay). The ^{180}Hg nucleus will not be extracted from the RILIS, thus the EC decay of ^{180}Tl is the only source for ^{180}Hg production.

All this will allow us to address the above-mentioned problems related to the low-energy fission of the very exotic nuclei: a) the height and the isospin dependence of the fission barriers; b) the mass/charge distribution; c) the influence of the shell effect on fission in this region of nuclei.

V. ISOLDE beam time estimate

The proposed experimental program at ISOLDE can be divided in 3 parts, according to the studied element (Tl, Fr or At):

- ^{178,180}Tl (6 days in total) – can be studied now with the RILIS. The RILIS ionization scheme for Tl is known and was tested in the earlier experiments [17,18]
- ^{200,202}Fr (7 days in total) – can be studied now with the usual surface ionisation ion source
- $^{194.196}$At (possibly, ^{192}At) – needs a negative ion source, which presently is not available. For this part of the program we request for the development of such a source.

The experimental set-up is shown in Fig. 5. We will use the modified version of the 'wind-mill' system, which was successfully used in our experiments IS387 and IS456 (Pb and Po charge radii measurements). The expected yields of fission fragments are given in Table and were calculated as $N_{fission}=N_{produced}\times b_{EC}\times P_{ECDF}$. The measured yield of the nuclei (except for ^{178}Tl) were taken from the ISOLDE yield data base [17,18]. The P_{ECDF} values necessary for the fission rate calculations were estimated based on the systematics from Fig.2 (right panel) and using the Q_{EC}-B_f values from Table.

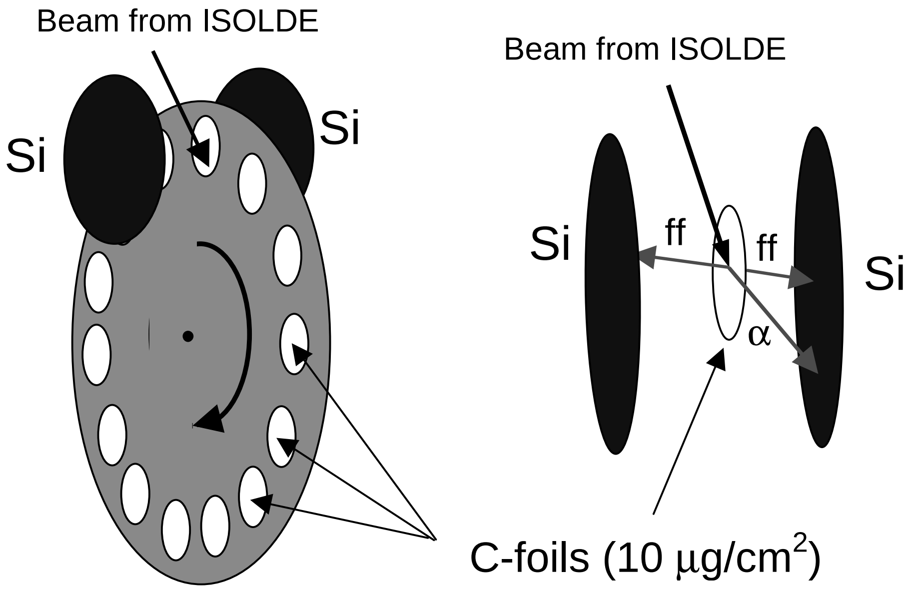

Fig. 5 The "wind-mill" system for the ECDF measurements with up to 20 thin (~10 μg/cm^2) carbon foils mounted (general view and blow-up of the foil-Si detectors arrangement). The ISOLDE beam is implanted in the carbon foil, which is surrounded by 2 Si detectors for α and fission decay measurements. The 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 the "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. We also consider the use of a neutron detector to measure the neutron multiplicities.

The expected yield for ^{178}Tl (no measurements were performed so far), was extrapolated from the measured yields of the heavier $^{179\text{-}184}$Tl isotopes [17]. We note that $T_{1/2}(^{178}\mathrm{Tl})$=250 ms is longer than $T_{1/2}(^{179}\mathrm{Tl})$=160 ms, thus the yield losses due to the relatively short half-life of ^{178}Tl will be less than for ^{179}Tl.

The expected yield for ^{202}Fr was recalculated from the measured SC ISOLDE data [17]. In the SC experiments with 600 MeV proton beam, an intensity of 70 ions/μC was achieved with the 13 g/cm^2 UC$_x$ target and W surface ionization source. We hope to obtain at least an order of magnitude higher intensity by using the PSB beam and a thicker target.

Table. Expected fission rates for nuclei of interest. The measured yields are from ISOLDE yield data base. The b_{EC} values were estimated by comparing the experimental half-lives to the beta-decay QRPA half-lives calculations from [19]. The P_{ECFD} values were estimated based on Fig.2 (right panel) and assuming a linear dependence of P_{ECDF} on the Q_{EC}-B_f value.

ECDF Parent ($T_{1/2}$)	Measured yield [ions/μC] source, target [16]	Q_{EC}-B_f [MeV]	b_{EC} [%] [19]	Estimated P_{ECDF}	$N_{fission}$ /day
^{180}Tl (0.7s)	610, RILIS(Nb) UC$_x$,50 g/cm^2	0.8	88-98 (SHIP)	$1.5\times(10^{-5}\text{-}10^{-4})$	500-5000
^{178}Tl (0.25s)	0.1 (estimated)	2.5	50 [17]	~10^{-2}	~25
^{200}Fr (0.05s)	0.2, Nb surface, ThC$_x$ 51g/cm^2	2.5	1.6 [17]	~10^{-2}	~2
^{202}Fr (0.34s)	700, surface	0	3 [17]	~4×10^{-5}	~40

To summarize, the whole program requires the following ISOLDE beams:

- 3 days for study of ^{180}Tl. This is a unique nucleus in the whole program, for which we expect to collect a few thousands of fission events. Apart from the precise P_{ECDf} measurement, this will allow us to study in detail the separate energy distributions of the fission fragments (thus, the mass distribution can be deduced), gamma spectra for fission fragments and the TKE value.
- 3 days for ^{178}Tl. The expected number of events will be enough for a quite precise (~10%) P_{ECDF} and TKE measurements.
- 7 days for ^{200,202}Fr (4 and 3 days for each of the isotope, respectively). The expected number of events will be enough for P_{ECDF} and TKE measurements.

Therefore, we request 2 separate runs:

- **6 days for ^{178,180}Tl with the RILIS**
- **7 days for ^{200,202}Fr with the surface ionization ion source.**

Additionally, we request a development of the At negative ion source for our program aimed at the ECDF studies of the $^{192\text{-}196}$At.

References

[1] V.I. Kuznetzov and N.K. Skobelev, Yadernaya Fizika, vol.4, 279 (1966); idem, vol.5, p.271 (1967) and p.1136 (1967)

[2] V.I. Kuznetzov and N.K. Skobelev, Fiz. Elementarnux chastiz", vol.30 ,p.1514 (1999)
[3] E. Ye. Berlovich and Yu.N. Novikov, Phys. Lett. B29, 155 (1969)
[4] I. V. Panov et al., Nucl. Phys.A747, 633(2005)
[5] Yu. A. Lazarev et al., Europhys. Lett. 4(8), 893 (1987)
[6] A. Staudt et al., Phys. Rev. Lett. 65, p.1543 (1990)
[7] H.V. Klapdor et al., Z. Phys. A, 292, p.249, (1979)
[8] D. Habs et al., Z. Phys. A285,p.53(1978)
[9] A. Andreyev et al., Phys. Rev. C., vol. 72, 014612 (2005)
[10] A. Grewe et al., Nucl. Phys., A614, 400 (1997)
[11] D.A. Shaughnessy et al., Phys. Rev. C65, 024612; idem, vol. C61, 044609
[12] Yu. A. Lazarev et al.,In Proc. 6th Int Conf. On Nuclei Far From Stability, Bernkastel-Kues, (1992).
[13] P. Moller et al. At. Data and Nucl. Data table 59, 185 (1995)
[14] W.D. Myers, W. Swiatecki Phys. Rev. C60, 014606 (1999)
[15] A.Andreyev et al., paper in preparation (2007)
[16] S. Hofmann et al., Eur. Phys. J., A32, 251 (2007)
[17] Isolde yield web page at www.cern.ch/isolde
[18] U. Koster, private communication
[19] M.Hirsch et al., ADNDT, 53, 165 (1993)