

EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

ISOLDE and Neutron Time-of-Flight Committee

A Proposal on “Beta-delayed fission, laser spectroscopy and shape-coexistence studies with radioactive At beams”

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Introduction

This Proposal aims at the studies of β -delayed fission (β DF), laser spectroscopy and shape coexistence phenomena in the At ($Z=85$) isotopes, and of shape coexistence in their daughter Po ($Z=84$) isotopes (produced after β decay of At), and Bi ($Z=83$) isotopes (produced after α -decay of At).

The proposal is based on a recent successful development of the radioactive At beams performed by joint efforts of RILIS@ISOLDE and TRILIS@TRIUMF teams, following the LoI-I-086 submitted in January 2010 [1]. The LoI identified five projects of high interest, both at the neutron-deficient and neutron-rich sides, and was strongly endorsed by the INTC. Following this support, several collaboration test runs were performed at ISOLDE and at TRIUMF, which led to the development of the efficient ionisation scheme for the element At and which culminated in the first ever measurement of the At's ionisation potential via the observation of the Rydberg states [2]. Based on this, a full-fledged nuclear and atomic spectroscopy program with the At beams, which was proposed in LoI-I-086, can now be initiated at ISOLDE, first steps of which (beta-delayed fission and shape coexistence) are described in this Proposal.

Beta-delayed fission is an elegant way to study low-energy fission. The recently discovered asymmetric mass distribution after the fission of ^{180}Hg (populated in the β decay of ^{180}Tl ref. [3,4]) and the substantial beta-delayed fission branching ratios, give rise to a renewed interest in the fission process itself. In these experiments fission is observed in a different (lead) region of the nuclear chart, with a typical N/Z value of ~ 1.25 compared to the 'standard' region of trans-uranium nuclei with $N/Z \sim 1.55$. Several recent theoretical developments try to understand our observation and produce predictions that can be checked with the data we plan to obtain with the present proposal (see below).

The exact origin of shape coexistence in the very neutron-deficient lead region is still a matter of debate and the authors of the recent review [5] stated in their conclusion that "...there is much to be learned about many-nucleon systems and their separate independent-particle and correlated-particle behaviours, revealed through shape coexistence". However, despite the long history of such studies, experimental data on nuclei with $Z > 82$ is still lacking. Charge radii and quadrupole moments are needed to understand the collective behaviour, while magnetic moments provide crucial information on deformation driving single-particle orbitals.

In the past several years, our collaboration performed extensive and successful laser and nuclear-spectroscopic studies of long series of Pb ($Z=82$) [6,7] and Po ($Z=84$) isotopes [8,9]. Another recent highlight of this program includes identification of a long-sought low-lying 0^+ state in ^{180}Hg [10]. In the '2011 campaign, beta-delayed fission studies of ^{178}Tl and of $^{200,202}\text{Fr}$ were successfully performed [3], along with laser spectroscopic studies of the lightest Tl isotopes, up to ^{179}Tl [11]. Very useful information on the shape coexistence in $^{182,184}\text{Hg}$ (populated via β decay of the parent Tl isotopes) was also obtained [12], which is crucial for interpretation of the data from the complementary Coulex and life-time measurements.

The present Proposal aims at the extension of this program to At isotopes. The proposal consists of two main parts (Tasks 1-2 below, detailed in Section 1), partially linked to each other by the necessity to use, in some case, the isotopically and isomerically pure beams of At isotopes, and through the use of the same detection setup(s).

- **Task 1.** Fission fragments (ff's) mass distribution in the β DF of $^{194,196}\text{At}$. In our At RILIS development runs [1,2] we have already proven that $^{194,196}\text{At}$ isotopes have the fission rates sufficient for ff's mass distribution measurements of the respective daughter isotopes (after β decay) $^{194,196}\text{Po}$. **(10 shifts requested, lasers in broadband mode, see Section 1).**

We also request **4 shifts** for a dedicated development of the narrowband mode for the At laser operation, which is a prerequisite for Task 2. (see Section 2)

- **Task 2.** First ever measurements of isotope shifts, HFS and charge radii for a long series of isotopes $^{193-212,217-223}\text{At}$; and βDF of isomerically-pure beams of $^{194m1,m2}\text{At}$ (**14 shifts requested, lasers in narrowband mode**).
- **Task 3.** A “by-product” of Tasks 1&2, **which does not require extra beam time**: Extensive nuclear spectroscopic/shape coexistence studies, e.g. important branching ratios measurements and a search for the low-lying (non-yrast) intruder bandheads and states on top of them in the daughter isotopes $^{193-204}\text{Po}$, populated by β decay of $^{193-204}\text{At}$. Simultaneously, extensive α - γ coincidence data will be collected for the α decay of At isotopes, which will allow the study of excited states in the daughter Bi isotopes.

Total requested shifts: 28 shifts of At beam, split in two separate runs as explained below.

Section 1. Physics motivation for the Proposal

Task 1. Fission fragment mass distribution in the βDF studies of $^{194,196}\text{At}$ (10 shifts requested, broadband lasers)

As a detailed description of βDF was given in the IS466 Proposal and respective addenda [3], 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, $P_{\beta\text{DF}}$. 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, unmeasurable) spontaneous fission branch in their ground state. In βDF , the excitation energy of the fissioning daughter nucleus is limited by the available Q_{EC} value of the parent β -decaying nucleus, therefore βDF provides unique fission data at low excitation energy (excitation energy E^* is comparable to or lower than the fission barrier, B_f). E.g. in βDF of ^{180}Tl , $E^*_{\text{max}}(^{180}\text{Hg})=10.44$ MeV, limited by the Q_{EC} -value of ^{180}Tl . In such cases, the topology of the energy surface around the fission barrier and shell effects plays a very important role, see [4].

Relevant to the present Proposal, in our recent βDF experiments at ISOLDE [3], we measured fission fragment mass distributions of $^{178,180}\text{Hg}$ (daughters of $^{178,180}\text{Tl}$ after β decay) and of ^{202}Rn (daughter of ^{202}Fr after β decay), see Fig. 1. While in total ~ 330 coincident and 1111 singles fission fragments were observed for ^{180}Hg [4], only a randomly-selected subset of ~ 45 coincident ff's is shown in Fig.1a for this isotope. This is made solely for the sake of comparison with the case of ^{202}Rn , for which only 43 coincident ff's were observed in our '2011 experiment, see Fig.1c. This is also relevant for the justification of the beam request for $^{194,196}\text{At}$.

As seen in Fig. 1, an asymmetric fission fragment mass split of ^{180}Hg and symmetric mass split of ^{202}Rn were observed (see also a comment to Fig.1). In passing, we mention that despite only 8 coincident ff's were observed in case of ^{178}Hg [3], their asymmetric energy distribution clearly indicates an *asymmetric* mass split, similar to the case of ^{180}Hg .

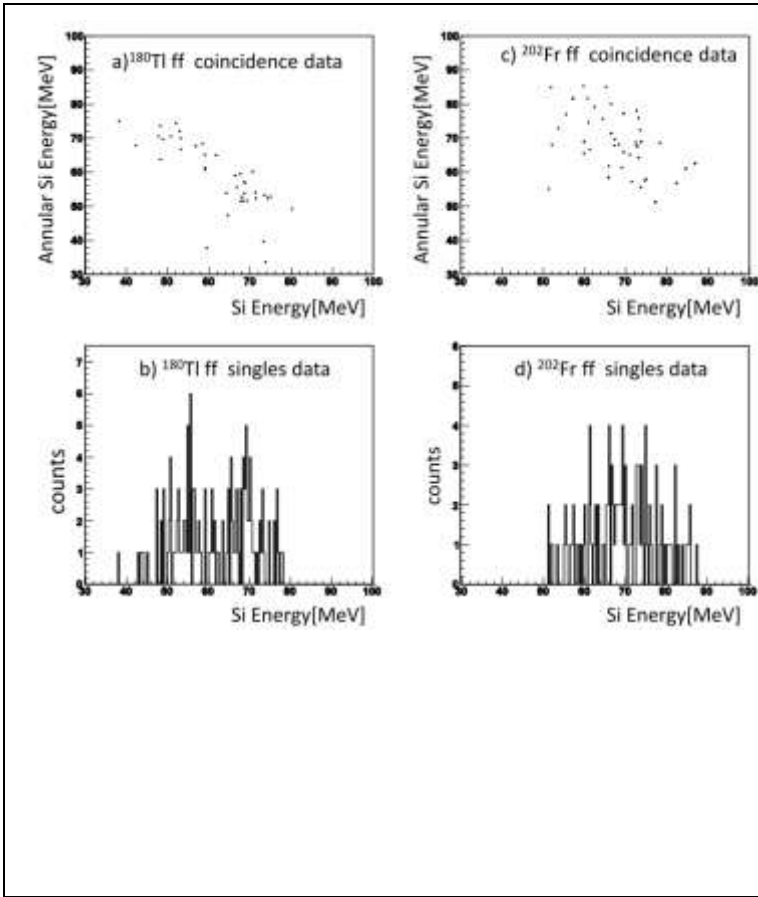


Fig.1. *Asymmetric* energy distributions of fission fragments from ^{180}Hg [4] and *symmetric* energy distribution for the case of ^{202}Rn measured by means of βDF of ^{180}Tl and ^{202}Fr [3], respectively. The top panels show the 2D coincidence ff's energy spectra observed in two silicon detectors of the Windmill (see Section 3), while the bottom panels show the singles ff's energy spectra measured in any of the two Si detectors. Only a randomly-selected subset of data is shown for ^{180}Tl , see text. As an asymmetric (symmetric) *energy* distribution signifies an asymmetric(symmetrical) *mass* distribution, in the text we will refer to the ff's mass distributions. We note that as ^{202}Fr has two isomeric states, it is not clear yet if the βDF happens in one or in both of them. Therefore, the observed events for βDF of ^{202}Fr could originate from two isomers. A CRIS-type experiment is needed to clarify this issue.

Thus, the $^{178,180}\text{Hg}$ data establishes the new region of asymmetric low-energy fission ($E^* < \sim 11\text{-}12$ MeV), in addition to the previously-known region in the heavy actinides, see Fig. 2. On the other hand, our ^{202}Rn data extends to even more neutron-deficient nuclei the previously-known region of *symmetric* fission, established in the experiments at FRS@GSI [13]. In the GSI experiments, the symmetric mass distribution in the chain of the lightest Rn isotopes was measured up to ^{204}Rn , with the excitation energy of ~ 11 MeV, which is comparable to that of ^{202}Rn , obtained via the βDF of ^{202}Fr .

Therefore, one expects a transition from the asymmetric to symmetric fission to happen in the region between $^{178,180}\text{Hg}$ and ^{202}Rn , shown by the thick blue arrow in Fig.2. This region comprises isotopes $^{192,194,196}\text{At}$, for which the βDF is expected to occur and was indeed observed in our experiments at SHIP(GSI) for $^{192,194}\text{At}$ [14] and at ISOLDE for $^{194,196}\text{At}$ [1,2]. We note that due to specific experimental conditions at SHIP, no ff's energy/mass distribution measurements is possible at SHIP.

Table 1. Measured rates of some At isotopes *during the At RILIS development runs* in '2010-2011 (proton beam intensity of $\sim 1.5 \mu\text{A}$). $^{193,194,196,217}\text{At}$ were measured with the Windmill system, by means of their α decays. $^{204,205}\text{At}$ were measured with a Faraday Cup (FC).

Isotop e	^{193}At (~ 25 ms, $b_\alpha \sim ?$) Windmill	$^{194m1,m2}\text{At}$ (~ 200 ms, $b_\alpha \sim ?$) Windmill	^{196}At (~ 400 ms, $b_\alpha \sim ?$) Windmill	$^{204,205}\text{At}$ FC	^{217}At Windmill
Rates	0.004 atoms/s	4 atoms/s ~ 5 ff/h	~ 220 atoms/s ~ 5 ff/h	20-50 pA	190 atoms/s

During the At ionisation scheme developments at ISOLDE, described in Section 2, the measured fission rate in singles (in any of two Si detector of the Windmill, see Section 3) was ~ 5 ff/h for β DF of both $^{194,196}\text{At}$, see Table 1. Thus, a coincidence fission rate for two fission fragments of ~ 1.6 ff/h is expected, based on known Si detector efficiencies from our previous experiments (a ratio of $\sim 1:3$ was deduced based on 330 coincidence and 1111 single fission events for ^{180}Hg , see above. Therefore, during 5 shifts (40 hours) for each isotope, ~ 64 coincident fission fragments (and ~ 200 singles ff's) should be observed for each of $^{194,196}\text{Po}$. This should be enough to draw a conclusion on the asymmetry (or symmetry) of their respective ff's mass distributions. This statement is confirmed by data in Fig.1, which clearly demonstrates that even with ~ 40 -50 coincident ff's events (or ~ 120 singles), a reliable discrimination between asymmetric fission of ^{180}Hg and symmetric fission of ^{204}Rn could be performed. **Therefore, we request 10 shifts for β DF studies of $^{194,196}\text{At}$ (lasers in the broadband mode).**

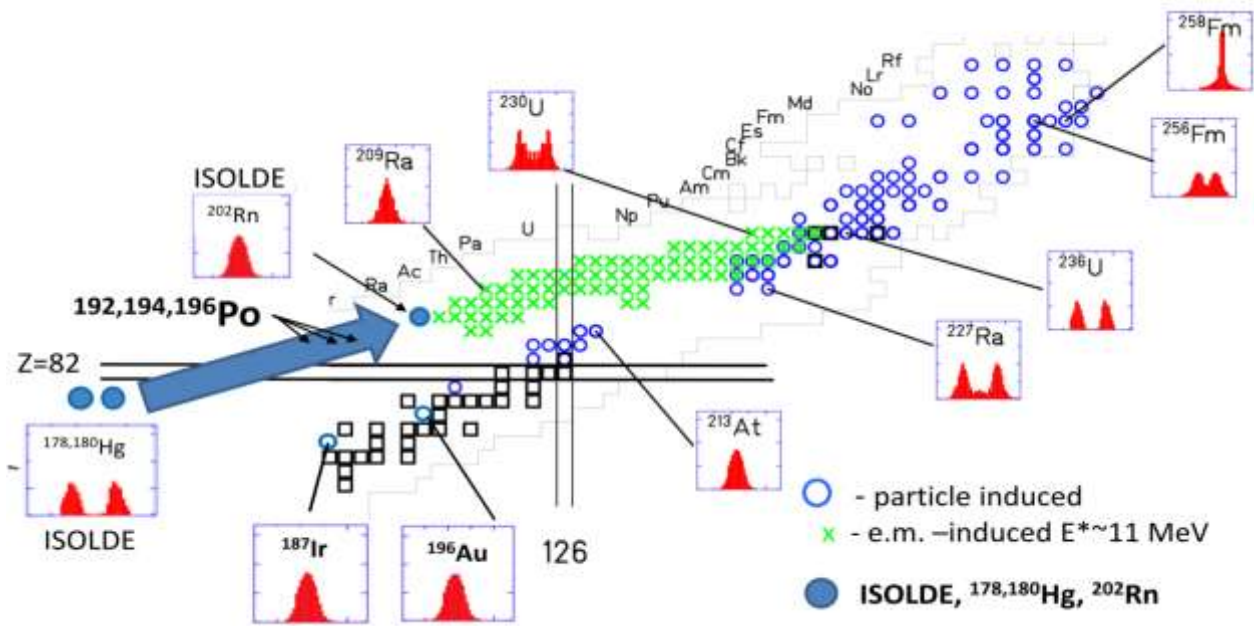


Fig. 2. (based on the plot by K.H.-Schmidt et al.). Nuclei with known ff's mass and/or charge distribution studied by low-energy fission. Two previously-known regions of predominantly *asymmetric* fission (heavy actinides, e.g. ^{227}Ra and trans-uranium nuclei, e.g. $^{230,236}\text{U}$, ^{256}Fm) and of *symmetric* fission (light actinides, e.g. ^{209}Ra and in vicinity of ^{208}Pb , e.g. ^{213}At) are shown by open symbols and crosses. Our recent beta-delayed fission data for *asymmetric* fission of $^{178,180}\text{Hg}$ and of *symmetric* fission of ^{202}Rn are shown by blue closed circles. A transition from *asymmetric* fission of $^{178,180}\text{Hg}$ to *symmetric* fission in ^{202}Rn is expected along the region marked by the blue thick arrow, which also includes $^{192,194,196}\text{Po}$ isotopes intended for fission studies in our program. In passing, we note that in the future we plan to study β DF of $^{186,188}\text{Bi}$, which also lie within the thick arrow.

The determination of the β DF probabilities for $^{194,196}\text{At}$ will also be performed with the precision of better than 10%, which is considered as a good result in the β DF studies. These data will be used to deduce the fission barriers of the daughter $^{194,196}\text{Po}$ isotopes, following the well-established procedure [15]. As ^{194}At has two isomeric states, we will also need a dedicated measurement with the narrowband laser for this isotope, 2 shifts are asked for this in Task 2.

Importantly, apart from providing otherwise inaccessible low-energy fission data, our measurements will also allow to check the predictions and applicability of recent fission approaches, which are being presently developed by different groups of theoreticians and which were motivated by our new fission data in this region of nuclei with very exotic ratio of $N/Z \sim 1.25$. We are already aware about several recent theoretical studies (submitted or under preparation) to calculate fission properties along the long chain of $^{176-198}\text{Hg}$ isotopes, some of them

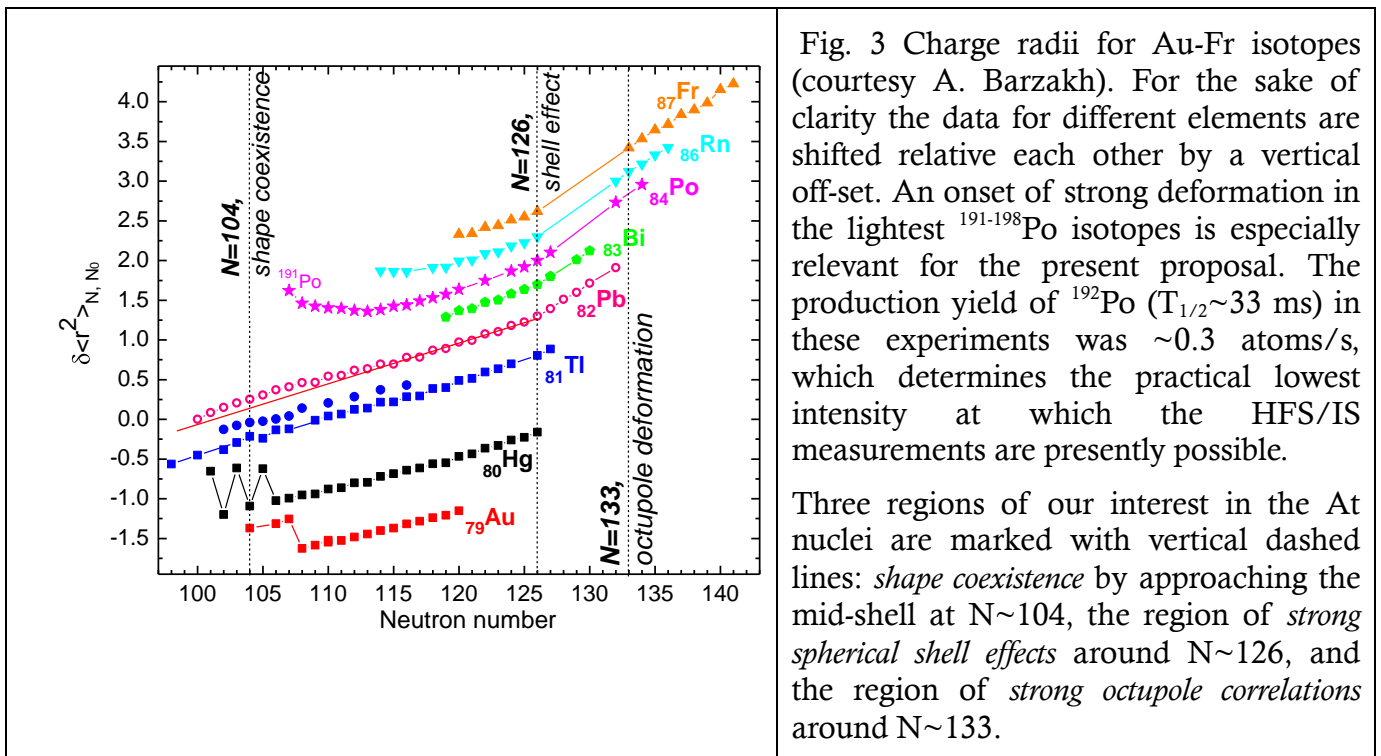
are mentioned in [16]. We are now in touch with these groups with a request to perform calculations for the broader region of nuclei, to include e.g. fission of ^{202}Rn , studied by us at ISOLDE [3], and also to provide predictions for $^{194,196}\text{Po}$ which will be studied in this Proposal.

Finally, we want to mention that two isomeric states exist in ^{194}At [17] and so far it is not clear which one (or both?) decay via βDF (a situation similar to ^{202}Fr). Therefore, provided the HFS splitting for two isomers in ^{194}At is sufficient, **2 shifts of ff's** measurements will then be also performed at two different RILIS settings in the narrowband mode, corresponding to two isomers. This isomer selection will uniquely determine whether only one or both isomers in ^{194}At decay via βDF . These two shifts are requested within Task 2 below, as this step can only be performed after the narrowband mode for At isotopes is developed.

Task 2. Isotope shifts, HFS and charge radii measurements for a long series of isotopes $^{193-212,217-223}\text{At}$ (12 shifts) and 2 shifts for βDF of ^{194}At (narrowband mode)

The region of very neutron-deficient nuclei in the vicinity of the proton shell gap at $Z=82$ is well-known for a prolific interplay between single particle and collective nuclear structure effects, see e.g. the latest review [5] and refs therein. A few examples, specifically relevant to the present Proposal, are shape coexistence, shape staggering and related phenomena in the broad region of the Pt-Fr isotopes, and the persistence of high- and low- spin isomeric states.

As mentioned in the introduction, our collaboration has recently performed successful laser spectroscopic experiments for long series of Tl ($Z=81$) [11], Pb ($Z=82$) [6,7] and Po ($Z=84$) [8,9] isotopes. The charge radii data resulted from these studies are shown in Fig. 3, together with the previously known results for some neighbouring elements. Specifically relevant for the present proposal is the observation of the strong onset of deformation in the lightest $^{191-198}\text{Po}$ isotopes, which actually happens earlier than it was proposed based on the previous in-beam and decay studies, see Fig. 3 and [8].

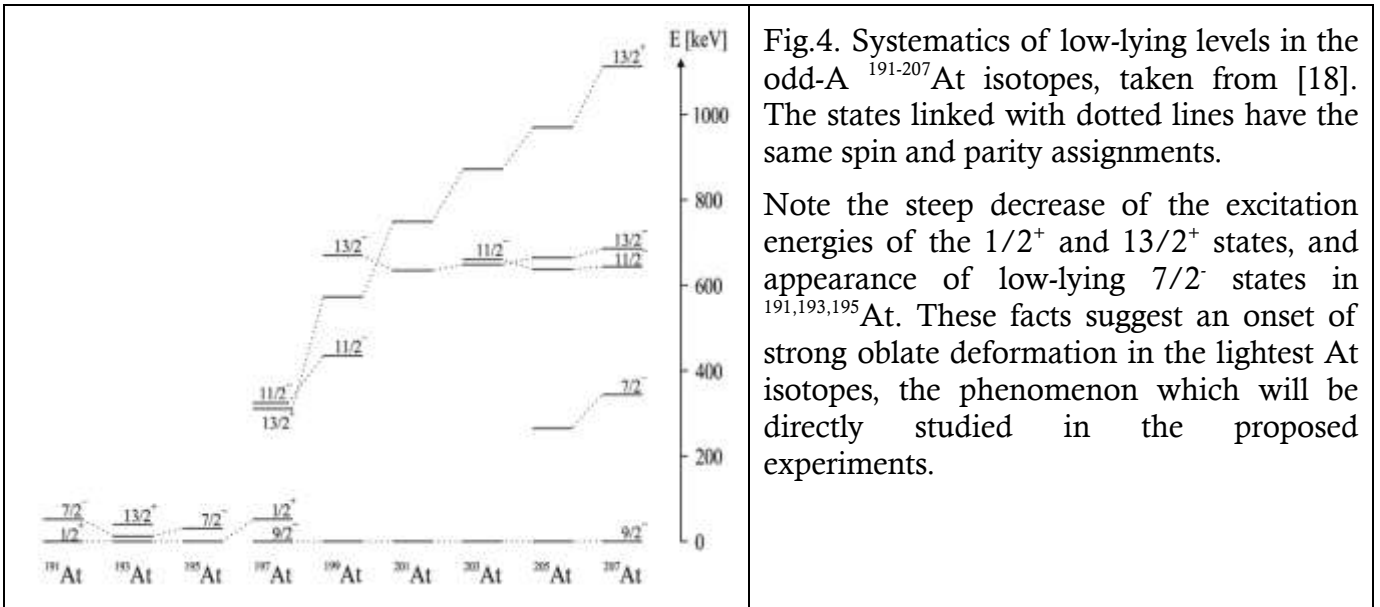


Furthermore, as also shown in Fig. 3, above Z=82 shell closure, only limited information exists for the odd-Z elements, with a few points only measured for relatively heavy Bi (Z=83) and Fr (Z=87) isotopes close to and above the neutron shell closure at N=126.

As an extension of our studies, we propose to perform laser spectroscopic measurements for a long series of $^{193-212,217-223}\text{At}$ isotopes covering the broad region of N=108-138 (isotopes $^{213-216}\text{At}$ cannot be studied by our technique due to their sub-millisecond half-lives).

In the first approximation, the At nuclei can be considered within an approach when a valence proton is coupled to the underlying Po core, thus an onset of deformation would also be expected in the At isotopes with masses $A(\text{At}) \leq 199$. This expectation is indeed confirmed by the recent in-beam and α -decay spectroscopy studies, see e.g. [17,18,19] and refs. therein, which suggested an onset of deformation in At isotopes starting around ^{198}At , see Fig. 4.

The heavier odd-A $^{197-207}\text{At}$ isotopes have a nearly spherical ground state with a (tentative) spin-parity assignment of $I=(9/2^-)$ due to the $(\pi 1h_{9/2})^3$ configuration, expected from the spherical shell model considerations. However, in $^{191,193,195}\text{At}$, a presumably oblate-deformed $1/2^+$ state (or the $\pi(4p-1h)$ configuration in the spherical shell model approach) becomes the ground state, with no evidence for the spherical $9/2^-$ state observed so far in these isotopes [18]. Instead, a low-lying $7/2^-$ state, based on presumably $7/2[514]$ Nilsson state, originated predominantly from the $\pi h_{9/2}$ orbital at sphericity and having a mixed $\pi 2f_{7/2}/\pi 1h_{9/2}$ character at oblate deformation was observed only slightly above ($E^* \sim 5-50$ keV) than the $1/2^+$ state. In ^{193}At , the energy difference $\Delta E(7/2^- - 1/2^+) = 5(10)$ keV [18], thus it cannot be presently excluded that actually the $7/2^-$ state becomes a ground state in this nucleus (see further discussion on ^{193}At below in the text). A steep decrease of the excitation energy of the $13/2^+$ state in odd-A $^{193,197-207}\text{At}$, see Fig.4, also hints to the onset of oblate deformation in the lightest At isotopes. These phenomena should also be seen in the neighbouring odd-odd At isotopes, and indeed the presence of the oblate-deformed $\pi 2f_{7/2} \otimes \nu 1i_{13/2}$ configuration was proposed in ^{192}At [19].



Apart from ^{191}At and $^{213-216}\text{At}$ having too short half-lives to be studied at ISOLDE (less than a couple of ms), most of the $^{192-212}\text{At}$ and $A(\text{At}) > 217$ isotopes should be accessible for our experiments.

We specifically note that in ^{193}At the $1/2^+$, $7/2^-$ and $13/2^+$ isomeric states were observed within the energy range of $E^* = 39(7)$ keV [18]. The half-life values of the three states are ~ 25 ms, which is comparable to the ~ 33 ms half-life of ^{192}Po , produced with the rate of ~ 0.3 atoms/s (see Fig.3). The latter brings us to a very intriguing observation which was made in our At

development run in '2011. Namely, the yield dropped by a factor of ~ 50 between $^{194,196}\text{At}$ (see Table 1), but a much larger drop by a factor of 1000 was observed by moving to ^{193}At , with the observed yield of only 0.004 atoms/s. A trivial explanation for so sharp a drop would be the relatively short half-life of ^{193}At , but even in this case we would not expect such a substantial drop as seen experimentally. A more intriguing explanation here would be that ^{193}At becomes very strongly deformed and has so huge an isotope shift that we missed ^{193}At even with our lasers in the broadband mode, used for the test yield measurements. If this idea is correct, then we would speak here about a large shape staggering, probably similar to the well-known effect in odd-A $^{181,183,185}\text{Hg}$ isotopes, see the Hg data in Fig. 3. This is certainly a very important issue, which requires a detailed study.

The physics quantities to be deduced in the laser studies include magnetic and quadrupole (except for $1/2^+$ states) moments, spin assignments, charge radii and deformations. To our knowledge, so far the magnetic moment for At isotopes was measured only for ^{217}At ($\mu = 3.81(18)$ n.m), which is close to the value expected for the $9/2^-$ proton, as discussed in [20]. The measurement was made by the low temperature nuclear orientation technique (NICOLE@ISOLDE) and relied on the known hyperfine field of At in a Fe host (see [20] and refs therein). These data are of importance for us as they will provide a stringent test for our atomic calculations, at least for the magnetic moments and hyperfine constant A (see also below).

Another interesting goal of this program is related to the fact that some heavy nuclei in the region of $N=132-138$ possess stable reflection-asymmetric octupole shape/deformation. It was earlier shown that the fingerprint of the octupole deformation is the inverse odd-even staggering effect in the charge radii trend. This was found in Fr, Ra, Rn and (possibly) Th chains. The heavy isotopes $^{217-223}\text{At}$ ($N=132-138$), to be studied in our program, cover exactly this region of possible octupole deformation, thus it would be important to measure their charge radii.

Furthermore, the isomer selectivity provided by the HFS technique will help to solve certain issues related to βDF studies, such as isomer separation in ^{194}At , mentioned above.

From a technical point of view, our collaboration has a large experience of laser spectroscopic studies in this specific region of the Nuclidic Chart, having performed the measurements for Tl, Pb and Po isotopes, both of the neutron-deficient and neutron-rich sides. Based on this experience, and also on the At development runs and yields measured in 2010-2011, we have a solid understanding on how to perform similar measurements for the At beams.

Namely, for the lightest and predominantly α -decaying $^{193-200}\text{At}$ isotopes, having relatively short half-lives (less than 1 minute) we will use the well-understood and well-performing Windmill system (see Section 5). This method was already used in our At development runs, when the fission rates of ~ 5 ff/h, quoted in Table 1 for $^{194,196}\text{At}$, were measured.

The heavier $^{204-212}\text{At}$ isotopes are produced with much higher yields which allows the use of the Faraday Cup (FC) technique, similar to what was used for e.g. stable $^{203,205}\text{Tl}$ isotopes and also for our Po measurements, see [8] for details. For example, the currents of $\sim 10-50$ pA were measured on the FC for $^{204,205}\text{At}$ and some of the neighbouring At isotopes. At some masses, the contamination from the surface-ionised Tl and Fr might become a problem. However, a number of different techniques were already applied by us in e.g. Po measurements to successfully overcome most of these problems. Regular reference measurements of the abundant ^{205}At isotope should be performed to ensure consistency during the measurement period and improve the accuracy of the relative isotope shift measurements.

The details on the laser operation for the above measurements are provided in Section 2.

The extraction of the charge radii from the measured isotope shift involves the use of atomic parameters, namely the atomic factor F and the specific mass shift constant SMS . The extraction of magnetic and quadrupole moments also requires the knowledge of atomic magnetic field strength and a gradient of atomic electric field at the nucleus. Those parameters are specific to the atomic transition and specific level and have therefore to be determined for each atomic system. The common technique relies on the comparison of the isotope shifts to the evolution of the charge radius measured with a different technique (e^- scattering, muonic decays, ...) over at least 3 isotopes and on the independent magnetic and quadrupole moments measurements by other techniques. Such experiments can however not be performed without a substantial amount of target material, that is why no data is available on the astatine isotopes yet (apart of ^{217}At , see above and [20]). Therefore, we have to rely on large-scale atomic calculations as in the case of the polonium isotopes, performed by S. Fritzsche [8]. Our systematic studies in the case of the polonium isotopes have shown that the uncertainty arising from these calculations are of the same order as the statistic uncertainty. The premise of such calculations have already been done in the frame of the search for new atomic levels in astatine [1,2]. Furthermore, as mentioned above, we will also use the known magnetic moment of ^{217}At for testing such atomic calculations. In addition, the ‘‘King’’ formalism, which gives the experimental ratio of the electronic factors F for different transitions, can be used for such a testing as well.

To conclude this section, we are confident of being able to perform reliable HFS/IS measurements for a long series of At isotopes and extraction of large amount of nuclear structure information from these measurements.

Task 3. Detailed α/β -decay studies of $^{193-204,217-223}\text{At}$ (no additional shifts requested)

As a ‘by-product’ of the βDF and IS/HFS studies, large statistics for α and β decays of $^{193-204,217-223}\text{At}$ will be collected, up to several orders of magnitude higher than in any previous experiments. Furthermore, both due to the high purity of the $^{193-204}\text{At}$ beams and the complementary use of the Wind-mill setup and/or Leuven tape station, which allow efficient measurements of α, β , conversion electron and γ decays, rich decay data will be obtained both for the parent At isotopes, and also for their daughter $^{189-200}\text{Bi}$ (after α decay) and $^{193-204}\text{Po}$ (after β decay) products.

The use of the HFS/isomer separation technique with RILIS will further improve the data quality for some cases, where the isomer separation is required. The latter is especially important for the determination of beta-branching ratios of $^{194\text{m}1, \text{m}2}\text{At}$, as these values are necessary to deduce the βDF probabilities for these isotopes.

Furthermore, the search for the low-lying 0^+ intruder bandheads (and respective bandheads in the odd-A neighbouring isotopes) and the members of the corresponding bands built on top of them will be possible for $^{193-204}\text{Po}$, which is important for the research on shape coexistence phenomena in this region of nuclei. In particular, these data will also provide important complementary information for the interpretation of recent Coulex experiments on Po performed and planned with REX-ISOLDE [21].

Our recent paper on the first identification of the long-sought excited 0^+ bandhead in ^{180}Hg [10] and new data on the mixing at low energy in $^{182,184}\text{Hg}$ [12] provide examples of the usefulness of such an approach.

Section 2. Development of the At narrowband mode and in-source At laser spectroscopy with RILIS

As mentioned above, the availability of pure astatine beams at ISOLDE owes itself to a series of successful resonance ionisation spectroscopy studies, carried out at ISOLDE and TRIUMF, [1,2] during which various efficient three-step ionisation schemes were developed for the RILIS, see Fig.5. During the scheme development RILIS lasers were operated in *broadband* mode to maximize the ionisation efficiency. Previously, RILIS has been used for a number of higher resolution in-source resonance ionisation spectroscopy for the study of ground state and isomer properties of exotic isotopes in the lead region with extremely low production yields and short lifetimes [6,7,8,9,11], where the scanning laser was operated in *narrowband* mode. This technique is particularly well suited to the heavier elements for which the Doppler broadening of the atomic lines due to the high temperature ionisation environment is lower.

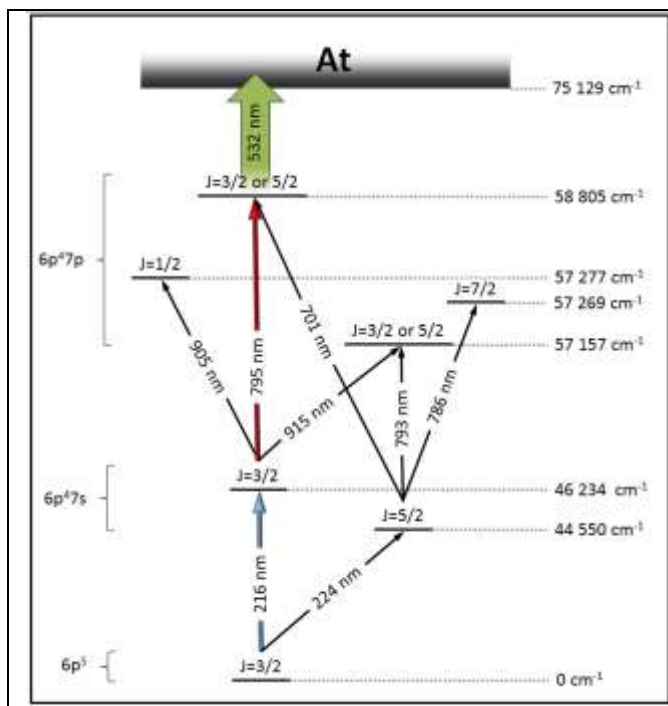


Fig.5 Developed ionisation schemes for astatine. The six different ionisation schemes were developed and investigated during I-086 [1,2]. One candidate for in-source laser spectroscopy is the highlighted path: 216-795-532.

In the proposal, we request 4 shifts specifically for characterization of the possibilities for scanning in the narrowband mode by using the 216 nm and/or 795 nm transitions and for the test of the possibilities of the isomers separation in the needed cases.

For the purpose of this proposal, astatine isotopes will be resonantly ionised with the RILIS inside the hot ioniser cavity of the ISOLDE target using the 3-step ionisation scheme 216-795-532 highlighted in Fig. 5. The laser used to excite the spectroscopic transition will operate in a reduced linewidth mode. In previous experimental campaigns, the laser linewidth was reduced by the addition of an etalon to the cavity of the scanning dye laser. Together with a reduction of the laser power to avoid saturation broadening of the atomic transition, the spectral resolution of 1 GHz matches that of the Doppler broadened atomic linewidth inside the hot cavity. A promising development for improving the scanning process and spectral resolution by pulsed dye amplification of a tunable CW diode laser has been successfully tested at LISOL, Leuven [22]. This technique is under investigation at ISOLDE. For this experiment, the preferred method for linewidth reduction will depend upon the choice of the spectroscopic transition and the outcome of these laser developments. During the ionisation scheme development both the 216 nm and the 795 nm transitions were scanned in broadband mode for various astatine isotopes from ¹⁹⁶At to ²¹⁷At. For both transitions the isotope shift was measurable, even with the scanning laser operating in broadband mode, see Fig.6, indicating the suitability of either transition for the proposed in-source spectroscopy study. For 216 nm the effect is approximately two times larger; however, since frequency tripling is required to generate laser radiation at 216 nm, the accuracy of

the transition frequency measurements reduces and the laser setup and maintenance is more complicated than for the generation of 795 nm.

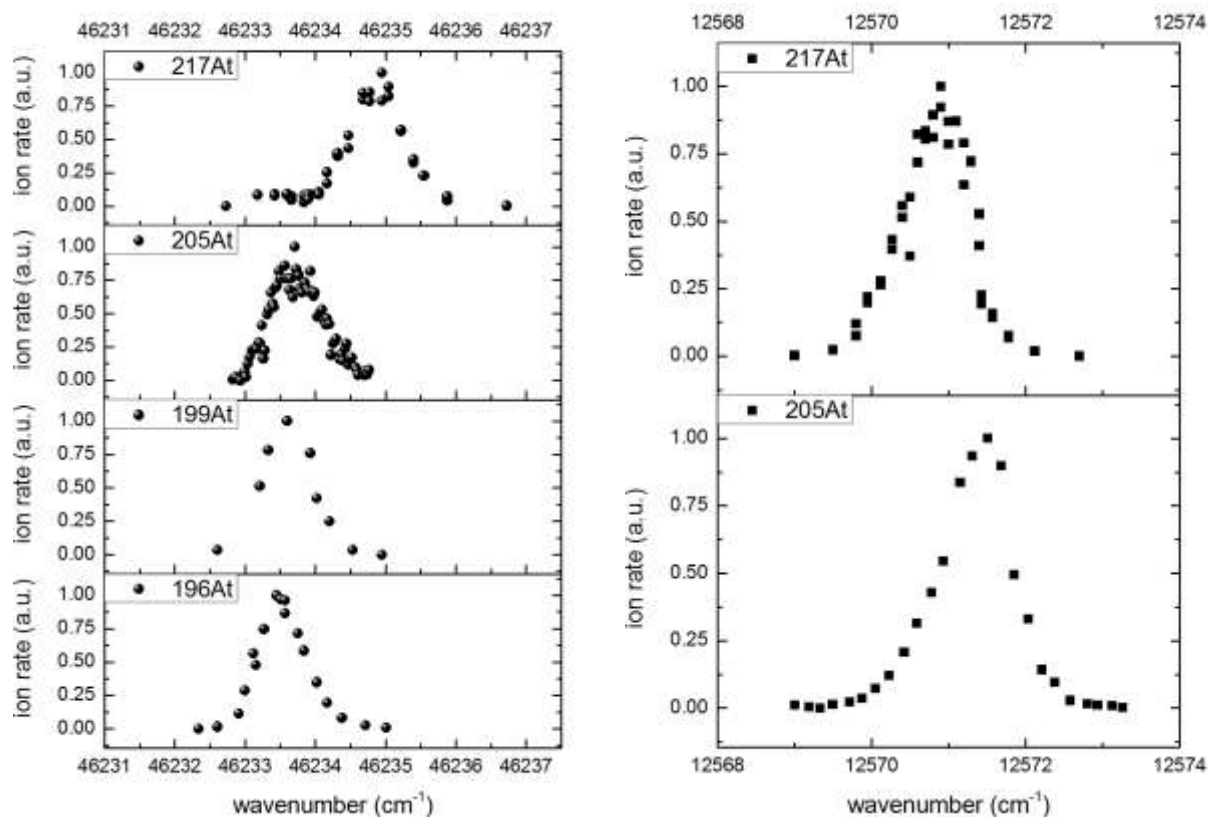


Fig. 6: Isotope shift measurements performed during initial spectroscopic studies within the LoI I-086. (a) Laser scans of the first step transition (216 nm) on astatine isotopes $^{196,199,205,217}\text{At}$. (b) Laser scans of the second step transition (795 nm) on astatine isotopes $^{205,217}\text{At}$.

4 shifts are therefore required to characterize the scanning performance across both of these transitions and for some preliminary isotope shift measurements. This will enable a reliable comparison of the behaviour of each transition for spectroscopy. The availability of data on isotope shifts for two transitions at least for several isotopes will allow for the application of the King formalism for proper extraction of mean square charge radii from the measured optical shifts. Besides, in these preliminary experiments we can test the possibilities of the isomer separation, which are needed for e.g. $^{194m1,m2}\text{At}$.

Ideally these measurements for several abundantly produced isotopes would be taken during the first part of a split two-part experimental campaign, with the laser spectroscopy study of all observable astatine isotopes to be performed in the second experiment. This would allow time for any necessary laser improvements to be made.

Section 3. Detection setup

The experimental set-up to be used in the proposed study is shown in Fig. 7 (taken from [4]). We will exploit the same ‘Windmill’ system, which was successfully used in IS466 (also in IS387,

IS407, IS456, I-086) and which allows the coincident fission fragments to be measured [4]. 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 Windmill rotates and a “fresh” foil is introduced for the implantation. The whole setup will be surrounded by the Ge-detectors to allow measurements of fission fragments (FF) in coincidence with gammas.

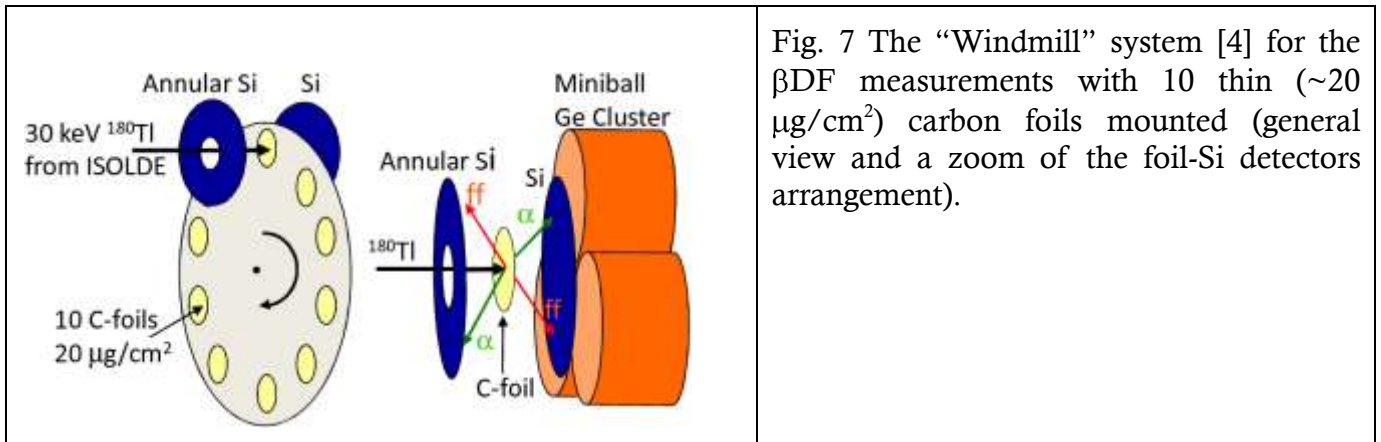


Fig. 7 The “Windmill” system [4] for the β DF measurements with 10 thin ($\sim 20 \mu\text{g}/\text{cm}^2$) carbon foils mounted (general view and a zoom of the foil-Si detectors arrangement).

For some isotopes, suitable for beta-gamma spectroscopy, we intend to use the Leuven tape station equipped with beta and Ge detectors. It could be installed at another beam line, so that we could send different isotopes to either WM or tape station setup, depending on the decay mode and production rate of each specific isotope.

Section 4. Summary of requested shifts

In total, we request 28 shifts of ISOLDE beam time, to be split in 2 periods.

1st period (14 shifts):

- 10 shifts for β DF of $^{194,196}\text{At}$ in broadband mode (ff mass yield measurements),
- 4 shifts for dedicated development of the narrowband mode for 2 transitions.

2st period (14 shifts):

- 12 shifts for laser spectroscopic studies of $^{193-212,217-223}\text{At}$ isotopes,
- 2 shifts for β DF measurements for ^{194}At in narrowband mode.

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Appendix

DESCRIPTION OF THE PROPOSED EXPERIMENT

The experimental setup comprises: a *Windmill* 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,IS511 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			
	Windmill	[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			
ionising 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	^{239}Pu , ^{241}Am , ^{244}Cm		
• Activity	1 kBq each		
Use of activated material:	-		
• Description			
• Dose rate on contact and in 10 cm distance	-		
• Isotope	-		
• Activity	-		

Non-ionising radiation			
Laser	Usual RILIS operation		
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. Hazard identification

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