EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH ISOLDE and Neutron Time-of-Flight Committee

Addendum to the IS534 Proposal

Part I: "Beta-delayed fission, laser spectroscopy and shape-coexistence studies with astatine beams" (pp.1-6)
Part II: "Delineating the island of deformation in the light gold isotopes by means of laser spectroscopy" (pp. 6-10)

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Introduction

In 2012, two successful campaigns for astatine (IS534) and gold isotopes were performed by our collaboration. This Addendum summarizes the preliminary results of both studies and presents the new beam request.

To complete the yet outstanding goals of IS534 experiment we request 13 shifts of the astatine beams and 18 shifts of the gold beam.

Part I: Beta-delayed fission, laser spectroscopy and shape-coexistence studies with astatine beams

This part aims at the completion of the extensive program on the nuclear and laser spectroscopy of astatine isotopes, initiated by our letter of intent I-086 (2010, [2]) and by the IS534 proposal (2011, [1]).

The main goals of the IS534 proposal include:

- Identification of the β -delayed fission (β DF) of ^{194,196}At
- Laser spectroscopy to study shape coexistence phenomena in the long chain of astatine isotopes, from neutron-deficient to neutron-rich species.
- Shape coexistence in their daughter polonium (Z=84) isotopes (produced after β decay of light At's), and bismuth (Z=83) isotopes (produced after α-decay of At's). No beam time was requested for this part, the data are collected as a by-product of βDF and laser studies.

Following pioneering development of the ionization scheme for astatine in 2011-2012 [3], two successful experimental campaigns were performed in 2012: May 2012 – β DF studies of ^{194,196}At at GPS in broadband mode, and October 2012 – laser spectroscopy studies of ^{197,198,203,205,207,209,211,217}At at HRS. Section I below summarizes the main achievements from 2012 and provides the motivation for further studies of astatine isotopes, while Section II discusses the experimental techniques.

In total we request 13 shifts of astatine beams to complete the following outstanding goals:

- Task 1. The measurements of isotope shifts (IS) and hyperfine structure (HFS) for isotopes ^{193-196,199,201,202,204,206,218,219}At (to complete the measurements made in 2012 for ^{197,198,203,205,209,211,217}At) 10 shifts, narrowband HFS scanning.
- Task 2.βDF of isomerically-pure beams of ^{194m1,m2}At: 3 shifts, narrowband mode.

According to our estimate, approximately 4 shifts should still remain unused from IS534.

Section I. Results from the May's and October's 2012 campaigns and the goals of this Addendum

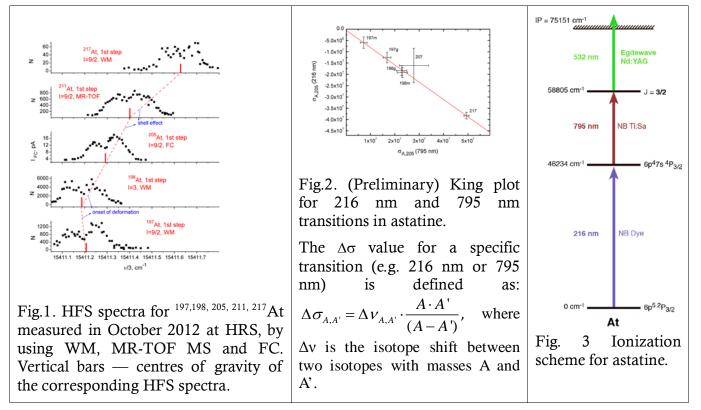
Task 1. The IS and HFS measurements for isotopes 193-196,199,201,202,204,206,218,219 At

In our HRS campaign in October 2012, successful HFS measurements for ^{197,198,203,205,207,209,211,217}At were performed. The Windmill (WM, detection of the decay radiation using silicon and germanium detectors for alpha's resp. gamma's), ISOLTRAP's multi-reflection time-of-flight mass separator (MR-TOF MS) and Faraday Cup (FC) techniques were used for different

isotopes, the choice made case by case, depending on the dominant decay mode (alpha/beta), halflife and contaminating background (e.g. francium or thallium isotopes). A comment on why only these isotopes were measured is provided below. To illustrate the quality of the results, a sub-set of astatine HFS spectra is shown in Fig.1.

It is important to mention that in October 2012, the measurements for some of the isotopes (197,198,207,205,217 At) were performed by scanning two alternative transitions, either the 1st step — 216 nm or the 2nd step — 795 nm (see Fig.3). The 1st step scanning is more suitable for the $\delta < r^2 >$ determination, while the 2nd step provides a broader and better resolved HFS structure, thus it is more suitable for μ and Q determination. To shorten the experiment and to restrict ourselves to the 2nd step scanning only, one should measure IS's for both transitions for a sufficiently large number of isotopes to apply the King-plot procedure to determine the electronic constants needed to extract $\delta < r^2 >$ from the isotope shifts (electronic constants for the 216 nm transition can be estimated from systematics due to the knowledge of the electronic configurations involved. As the electronic configuration of the upper level of the 2nd transition from the known constants for 216 nm transition).

Figure 2 shows the (preliminary) King-plot for 216 nm and 795 nm transitions, extracted from the data obtained in October 2012. This will allow us to use only the 795 nm transition for scanning the resonances in the next experimental campaign.



The relative charge radii extracted from the preliminary analysis are shown in Fig.4, in comparison with the charge radii of polonium isotopes, measured earlier by our collaboration [4]. The expected kink at N=126 is clearly seen, along with the similar onset of deformation in the lightest astatine isotopes, as in polonium. Note also the shape staggering in ¹⁹⁷At where the isomeric (intruder) state (I=1/2⁺) has markedly greater deformation than the ground state (I=9/2⁻). For confirming the ordering in energy of the two states in ¹⁹⁷At, the difference between their hyperfine structures was used to selectively ionize the two states and measure their masses in the Penning trap of the ISOLTRAP experiment.

This phenomenon of staggering was not observed in polonium. The observed trend, (together with available alpha spectroscopy and in-beam data [5,6]) suggests that even stronger deformation should be expected in the lightest isotopes ¹⁹³⁻¹⁹⁶At, which were not yet measured.

Therefore, the main goal of this part of the Addendum is to measure HFS spectra for alphadecaying isotopes ¹⁹³⁻¹⁹⁶At (with WM), to assess the evolution of charge radii and development of deformation by approaching the neutron mid-shell at N=104, where the influence of deformation effects should maximize.

We also need data for yet unmeasured isotopes ^{199-202,204,206,208,210,212,218,219}At. Altogether, the data for a long chain of isotopes will allow to fix global trend, to determine odd-even staggering and assess the possible influence of isomerism on charge radii. All of these isotopes are quite abundantly produced, therefore the measurements should be relatively easy with the choice of the appropriate technique (WM, MR-TOF MS, FC). The ^{218,219}At part of the measurements is possibly subject to a strong contamination from heavy francium isobars. In this case the measurements with the newly-build ISOLDE Decay Station (IDS) or using the tape station could be used, which should weaken the problem of contamination.

In passing, we note that due to yet unknown reason, the astatine yield in the October's 2012 HFS run was approximately a factor of 5-10 lower than in the May's 2012 βDF run (see next subsection). The available yields allowed us to measure the data for the more abundantly-produced nuclides, while we decided to leave the weaker-produced (e.g. the lightest isotopes) for a latter campaign, requested for by this Addendum. Important, however, is the fact that the yield suddenly became 'normal' (equal to that in the May's run) in the very last shift of the October's 2012 run. The reasons for this 'sudden' increase in yield are still under investigation in collaboration with the RILIS and ISOLDE Target Group. Possible issues with lasers and/or ISCOOL transmission are considered. Nevertheless, the fact that both in May and October 2012 we eventually had comparable and "good" rates, gives us confidence that the whole IS/HFS program requested by the IS534 proposal and this Addendum will be successfully completed within the requested 9 shifts.

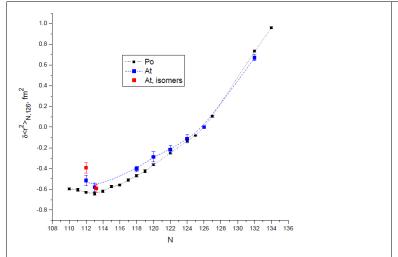


Fig.3. Comparison of deduced relative charge radii for astatine isotopes with the values for polonium. Data are normalized at N=126. Note the kink at N=126, and the onset of deformation in the light astatine isotopes similar to polonium. The two values shown for ¹⁹⁷At (N=112) and ¹⁹⁸At (N=113) correspond to two long-lived alpha-decaying nuclear states in each nucleus. Note the shape coexistence in ¹⁹⁷At where the isomer (intruder) state (I=1/2) has markedly greater deformation than the ground state (I=9/2).

Task 2. βDF of isomerically-pure beams of ^{194m1,m2}At (3 shifts, narrowband mode).

For the detailed motivation for β DF studies of ^{194,196}At we refer to to the IS534 Proposal [1]. In the May's 2012 campaign at GPS with lasers in the broadband mode, we performed successful β DF measurements for both isotopes, in which rates of ~40 ff/h and 8 ff/h fission events were observed, respectively. For ¹⁹⁶At, for which only one long-lived (ground) state is known, the β DF program is now considered as complete. An unexpected multi-modal energy (thus, also, mass)

distribution of fission fragments for βDF of ¹⁹⁶At was preliminary deduced, the data analysis is underway at York and KU Leuven.

Similar data were obtained for β DF of ¹⁹⁴At. However, this isotope has two long-lived predominantly alpha-decaying nuclear states [5], with yet unknown relative excitation energy (that is why we call them both as 'isomers' in the rest of the text). Furthermore, at this moment it is not yet clear if both isomers or only one of them undergoes β DF [7]. Several important issues will be addressed by these measurements, such as the influence of angular momentum on fission fragment mass distribution, the possible difference of β DF branching ratios. To resolve this issue and to deduce the fission fragments mass distribution for each isomer, we need a dedicated β DF measurement with the narrowband scanning for both isomers (this was included in our original IS534 proposal). With the narrowband laser operation for astatine isotopes developed in our October's 2012 campaign, this task becomes possible now. As only part of the excited HFS atomic levels will interact with the narrowband laser we expect that the fission rate will drop by a factor of 2-3 in comparison with the GPS experiment in May 2012.

Therefore, we request 3 shifts to complete the β DF study of ^{194m1,m2}At. This, first of all, requires the HFS scanning to find the laser frequencies at which the two isomers are sufficiently resolved. This can be done by monitoring the alpha and gamma decay of the two isomers [5]. This first task will be performed within the 9 shifts requested for the HFS measurements above. Afterwards, two dedicated β DF measurements will be performed at the respective laser frequencies, each 1 shift long to reach statistically significant number of fission events (or to prove non-observation of β DF for one of the isomers).

While studying the isomerism in neutron-deficient At isotopes by spectroscopy, the collected data might not be sufficient to clearly identify the states or the ordering. We thus propose to perform subsequent mass measurements using the ISOLTRAP Penning-trap setup. Using the alpha-energy information from the WM experiment, these direct mass measurements would also serve as anchor points for the evaluation of the masses along the alpha-decay chains in the region, valid also for the gold experimental program below. (A number of 3 shifts would be required for this, where we intend to use the remaining shifts of IS518, so no further shift request is needed at this point.)

Section II. Detection setup

The experimental set-up will be the same as in our previous HFS and βDF measurements. For shorter-lived, predominantly alpha-decaying isotopes, e.g. ^{193-196,199,200,201,218,219}At, the WM system will be used, while for some of the abundantly produced At isotopes without the Fr background, the FC will be exploited. For some isotopes (e.g. ^{202,204,206}At), for which the francium and/or thallium contamination will be present, the use of ISOLTRAP's MR-TOF MS is required. These measurements thus need to be performed at the ISOLTRAP setup in collaboration with the ISOLTRAP team. If the new ISOLDE Decay Station (IDS) is completed by the time of the experiment, we can use it for these measurements as well.

Section III. Summary of requested shifts for astatine

In total, we request 13 shifts of astatine beam time, in one running period.

- 9 shifts for IS and HFS measurements of ^{193-196,202,204,206,208,210,212,218,219}At
- 3 shifts for β DF of ^{194m1,m2}At in the narrowband mode.
- 1 shift in total for reference measurements with a suitable and easily measurable isotope for regular checks of the stability of the lasers, separator and counting system.

The requested beam time is based on our experience from May/October 2012 experiments.

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Part II: "Delineating the island of deformation in the light gold isotopes by means of laser spectroscopy".

Introduction

During the October 2012 "gold campaign" (4.5-shifts), first successful IS/HFS measurements for ^{177,178,179,180,181,182,191}Au were performed. Section I below provides the physics motivation for IS/HFS studies of the gold isotopic chain, while Section II summarizes the main achievements from the 2012 campaign and the arguments for the new beam request.

In total we request 18 shifts of gold beams to complete the whole program.

Section I. Physics goals

Fig. 1 shows the contrasting behaviour for charge radii for several isotopic chains in the lead region. The lead isotopes (Z=82) stay essentially spherical in their ground states, despite the presence of low-lying intruder configurations in the vicinity of the mid-shell at N=104 [1]. The mercury chain (Z=80) demonstrates the gradual increase of deformation by approaching N=104 with a sudden onset of deformation and shape staggering in the charge radii between ^{181,183,185}Hg on the one hand and ¹⁸⁷Hg on the other hand [2]. Finally, the chain of platinum isotopes (Z=78) also shows a gradual increase of deformation by approaching N=104 (without pronounced shape staggering), with what looks like a beginning of the trend to return to more spherical configuration in the lightest isotopes beyond the mid-shell [3].

Earlier IS/HFS measurements provided charge radii data for the neutron-deficient isotopes ¹⁸³⁻¹⁹⁹Au, see [4-6] and references therein. While the heavier isotopes ¹⁸⁷⁻¹⁹⁹Au were found to be nearly spherical (or weakly triaxial, as proposed by complementary in-beam studies, e.g. [7,8] and references therein), a sudden onset of deformation was observed between ¹⁸⁷Au and ¹⁸⁶Au [4], see Fig. 1. The latter effect is due to the influence of $1\pi h_{9/2}$ intruder orbital [4]. For the ground states of ¹⁸³⁻¹⁸⁶Au isotopes the deformation of $\beta_2 \sim 0.25$ was deduced, which is typical for strongly-deformed prolate configuration in the lead region. These observations raised an important question on how

the deformation will develop in the lightest gold isotopes, in particular by crossing and moving beyond the neutron mid-shell at N=104.

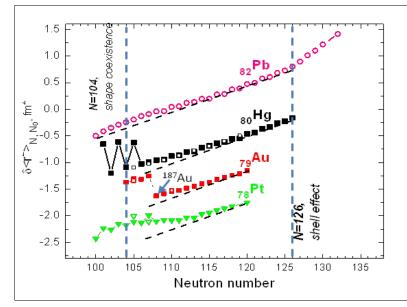


Fig.1. Charge radii for platinum, gold, mercury and lead isotopes. Isotopic series are arbitrarily shifted relative each other in the y-axis to avoid overlap of different chains. Sloped dashed lines show the predictions of the spherical droplet model. The sudden deformation jump between ¹⁸⁶Au and ¹⁸⁷Au is marked by the blue arrow.

Therefore, the main goal of our gold program is to extend the earlier IS/HFS measurements to the lighter isotopes close to and beyond N=104. Apart of charge radii, very important information on magnetic dipole moments will be extracted. (We note that the spins/magnetic moments of the ground states for isotopes with A(Au)>183 are known [9]). Magnetic moments will help to resolve the long-standing issue on the configuration of the low-spin ground states in the chain of ¹⁷¹⁻¹⁸³Au isotopes, see e.g. an extensive discussion of the possible spins and configurations in the in-beam study of ^{173,175,177}Au isotopes in [8]. Indeed, while the lightest known odd-A gold isotopes ^{171,173}Au were proposed to have spherical 1/2⁺ ground states (the conclusion derived solely based on spectroscopic factors and hindrance factors from proton and alpha-decay studies), the spin assignments for ¹⁷⁵⁻¹⁸³Au are far from being clear. This is due to possibility of triaxiality and/or strong deformation occurring in these isotopes.

Another important goal of this program is to search for isomeric states, which are expected in these nuclei, due to the close proximity of several proton configurations with low angular momentum $(3s_{1/2}, 2d_{3/2}, 2d_{5/2})$ and a high-j $1h_{11/2}$ orbital. As the $11/2^{-1}$ isomers are known in some of the odd-A Au isotopes, e.g. 171,173,175,177,189,191,193,195 Au (see e.g. [10]), the high-spin isomers are also expected (in some cases – known) in their odd-odd neighbours.

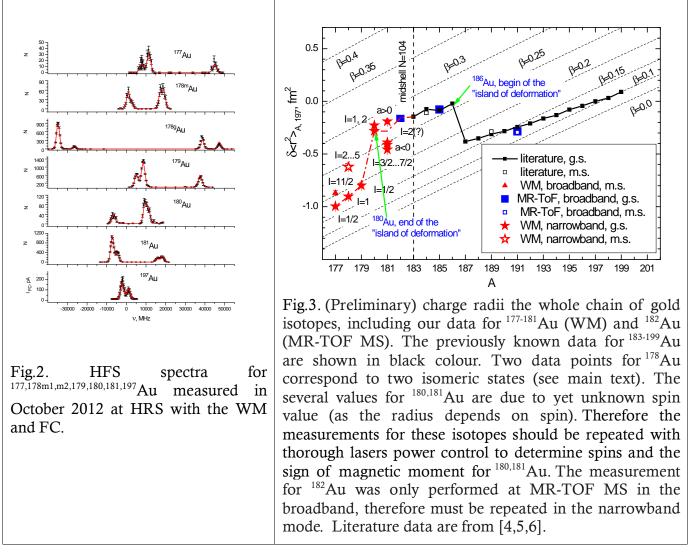
Section II. Results from the October 2012 campaign

In the IS534's October 2012 campaign, successful IS/HFS measurements for 177,178,179,180,181,182,191 Au were performed. These measurements were performed in the framework of development of a new, effective (from the point of view of charge radii and moment measurements) and efficient ionization scheme for the gold isotopes that was executed after the IS*534's* May 2012 experiment on β DF of astatine during spare nights shifts between day shifts of REX tuning.

Once the ionization scheme was established, we tested the applicability of the scheme for IS/HFS measurements on ^{177,178,179}Au with the WM system. Despite these tests were performed in the broadband laser mode (thus quite large uncertainty on the radii, and no information on magnetic moments/spins), they showed that the radii of ^{177,179}Au were considerably lower than those for strongly-deformed ¹⁸³⁻¹⁸⁶Au (after accounting for the trivial mass dependence). This result demonstrated the importance of and the necessity to extend these measurements and to put them on proper footing before the long ISOLDE shutdown in 2013-2014, to establish ISOLDE

leadership in the studies of this important phenomenon. Therefore, in consultation with the ISOLDE coordinator we used a number of shifts from the approved IS534 astatine program to proceed with the gold measurements in a timely manner. The physics output of the gold campaign could be equally good as of the astatine program. We performed the gold measurements after we completed an essential part of the astatine program, as described in the Part I of this Addendum.

The same techniques as for the astatine measurements were applied, including the measurements with the WM, FC and MR-TOF MS system, therefore we refer the reader to the description provided in Part 1. To illustrate the quality of the results, the measured HFS spectra are shown in Fig. 2, while the (preliminary) charge radii for ¹⁷⁷⁻¹⁸²Au, deduced from our data, are shown in Fig. 3. We note that with the help of HFS scanning with WM and gating on different alpha decays of ¹⁷⁸Au, we were able for the first time to establish the existence of two isomeric states in this nucleus. We will denote both states as 'isomers' in this document.



The mains (still preliminary) conclusions which can be drawn from Fig.3 are:

- ¹⁸⁰⁻¹⁸²Au seem to follow the trend of deformed ¹⁸³⁻¹⁸⁶Au (however, see a comment below). It is interesting to note that ¹⁸³Au (N=104) lies exactly in the middle of the 'island of deformation' in gold isotopes.
- A "reverse" transition from deformation to a less-deformed shape happens between ¹⁸⁰Au and ¹⁷⁹Au. ¹⁷⁹Au seem to have the deformation similar to that of ¹⁸⁷Au ($\beta_2 \sim 0.18$), however both isotopes might be influenced by triaxiality.

- ¹⁷⁷Au and one of the isomeric states in ¹⁷⁸Au continue the trend of reduced deformation established by ¹⁷⁹Au. To determine whether the state of reduced deformation is the ground state or the excited state in ¹⁷⁸Au, the two states were selectively ionized by exploiting the very different hyperfine splitting (Fig. 2) and separately sent to the Penning-trap system of the ISOLTRAP experiment, where their masses and ordering in energy were determined.
- The difference in charge radius between the two isomers in ¹⁷⁸Au indicates the shape staggering.

A few important points in respect of data in Fig.2 and 3 should be emphasized, which also explain our need for extra measurements of most of the isotopes measured in October 2012.

- It is obvious from Fig. 3, that the extension of the measurements towards ^{175,176}Au is indispensable. Both isotopes have two long-lived alpha-decaying isomeric states, thus it is important to evaluate the further development of the deformation trend for the lightest gold isotopes, to determine spins and magnetic moments (to ascribe configurations to the ground and isomeric states) and to check whether the shape staggering observed in ¹⁷⁸Au preserves in ^{175,176}Au. These measurements will constitute one of the main goals of the present beam request. The yields measured in October 2012 indicate that both isotopes should be accessible, but due to their lower production yields 3 shifts in total will be dedicated to these two isotopes. The preliminary broadband scans are needed to find approximate peak positions; wide HFS demands more time.
- There is only a single narrowband spectrum for each of ¹⁷⁷⁻¹⁸¹Au, taken with WM (for ¹⁷⁸Au high spin there are 2 spectra). As the uncertainties are determined mainly from the distribution of individual values, we need at least 3 spectra on each mass. From our experience in October 2012 we learned that each scan takes ~2 hours, thus on average, 6 hours of scanning is needed to complete one isotope.
- All MR-TOF MS measurements (e.g. ^{182,185,191}Au) were performed in the broadband mode with only a single scan for each isotope. Apart of a large resulting uncertainty for the charge radius and magnetic moment, the resolution of the HFS components is very poor and does not allow a conclusion on the spin to be made in ¹⁸²Au (spins and moments are known in ^{185,191}Au). Thus, ¹⁸²Au must be measured in the narrowband mode.
- For ¹⁸¹Au, depending on the assumption on the spin and the sign of magnetic moment, different charge radii can be obtained (see Fig.3), leading to very different conclusions on the deformation of this isotope. Therefore, extra HFS spectra must be measured for ¹⁸¹Au (in the narrow band, with thorough control of the laser power to determine spin and the sign of magnetic moment from the intensities ratio).
- There are long-lived high-spin isomers in ¹⁸⁷Au (probably 9/2⁻, presumed strongly deformed state, thus, a large isomer shift (shape coexistence) is expected) and in ^{191,193,195}Au (11/2⁻), with yet unmeasured HFS (thus no radii and moments). The single measurement with MR-TOF MS (^{191m}Au) was made in the broadband mode, with only a handful of frequency points, thus has a very high uncertainty. It seems natural to perform the HFS measurements for these isomers. Due to the long half-life of these isotopes and thallium contamination at these masses, these measurements must be performed with the MR-TOF MS or FC.

Section III. Summary of requested shifts for gold isotopes

In total, we request 18 shifts of gold beam time. All scanning must be performed in the narrowband mode.

• 9 shifts for HFS measurements of ¹⁷⁵⁻¹⁸¹Au with WM

- 2 shifts for HFS measurements of ^{182,183}Au with MR-TOF MS
- 3 shifts for HFS measurements of isomeric states in ^{187,191,193,195}Au using ISOLTRAP's MR-TOF MS
- 3 shifts for mass measurements of isomeric states discovered or observed in the HFS studies throughout the gold isotopic chain, having unknown excitation energies
- 1 shift in total for reference measurements with stable ¹⁹⁷Au and/or another suitable and easily measurable isotope for regular checks of the stability of the lasers, separator and counting system.

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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,
MR-ToF		IS456, IS466, I-086,IS511, IS534
	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).	
Beamparticle 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-ionising radiation		
Laser	Usual RILIS operation	
UV light	-	
Microwaves (300MHz-	-	

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20 CU-)		
30 GHz)		
Radiofrequency (1- 300MHz)	-	
Chemical		
Тохіс	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