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

Addendum to the IS534 Proposal

(based on the text submitted in May 2013)

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

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Introduction and explanatory note for the Addendum

In 2012, two successful campaigns for astatine (IS534) and gold isotopes were performed by our collaboration. The main results both for gold and astatine were summarized in the Addendum, submitted in to June's2013 INTC, in which we requested 13 extra shifts (plus 3.5 shifts remaining from 2012) for astatine to finish this study, along with an extra 18 shifts to study the Hyperfine Structure (HFS) of the gold isotopes (the latter part was approved in June 2013). The astatine part of the Addendum was, however, not approved by the INTC in June 2013 and some questions were raised, as shown in this extract from the INTC comments (shortened version, only the part relevant to astatine is retained):

"…It is proposed to continue the experiment IS534 by studying the properties of long chains of isotopes of astatine (Part I) and gold (Part II). The main physics goals include the identification of beta-delayed fission, laser spectroscopy, and the determination of deformation and shape coexistence. In Part I it is planned to measure isotope shifts and the hyperfine structure of astatine isotopes not covered in previous experiments as well as beta-delayed fission of isomerically pure beams of ^{194m1,m2}At. … The previous, similar experiments performed by the same group are considered successful and the motivation for the proposed extension is found convincing and sound. However, the ***justification for the number of required shifts was found to be insufficient***. In addition, ***some technical issues appeared concerning the yields of the lightest astatine isotopes, and the problems are still not resolved***. Therefore, the committee decided to recommend for approval only Part II of the addendum, devoted to gold isotopes".

First of all, we highly appreciate that the INTC acknowledges in their recommendations that the physics motivation itself is convincing and sound. Meanwhile, the data analysis is being performed by the KU Leuven and York groups, and first publications are in preparation. Based on the above, our Collaboration is strongly convinced that we need to complete the outstanding measurements as soon as possible to achieve full physics goals of the astatine project. This will maintain and strengthen the ISOLDE's leadership in these interesting subjects.

The present Addendum addresses the questions, raised by the INTC. Moreover, our better understanding of the statistics and precision needed, which we gathered from the on-going data analysis, leads us to increase the requested number of shifts by 3.5. The physics motivation and goals are identical to the previous Addendum. A dedicated section (Section IV) has been added to specifically comment on the INTC's questions.

To complete the yet outstanding goals of the IS534 experiment we request **16.5 shifts of the astatine beams**. In addition to the 3.5 shifts remaining from the 2012 campaign, this amounts to **20 shifts** to finish the study of astatine isotopes (in the May's 2013 Addendum, 13 shifts were requested, with further 3.5 shifts remaining from 2012, in total 16.5 shifts). The detailed explanation is now given in Section IV.

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 (βDF) 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 – βDF 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 now request **16.5 shifts (+3.5 shifts available, thus total amount, if approved, should be 20 shifts)** of astatine beams to complete the following outstanding goals:

- **Task 1.** The measurements of isotope shifts (IS) and hyperfine structure (HFS) for isotopes ^{193,194,195,196,199,201,202,204,206,218,219}At (to complete the measurements made in 2012 for $^{197,198.203,205,207,209,211,217}$At) – **17 shifts, narrowband HFS scanning.**
- **Task 2.**βDF of isomerically-pure beams of ^{194m1,m2}At: **3 shifts, narrowband mode.**

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 $^{193\text{-}196,199,201,202,204,206,218,219}$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), half-life 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 $\delta\langle r^2\rangle$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 $\delta\langle r^2\rangle$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 may be mixed, the King-plot procedure enables one to calculate these constants for 795 nm 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.

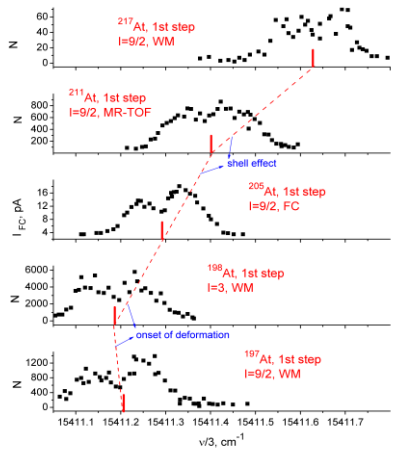


Fig.1. HFS spectra for ^{197,198,205,211,217}At measured in October 2012 at HRS, by using WM, MR-TOF MS and FC. Vertical bars — centres of gravity of the corresponding HFS spectra.

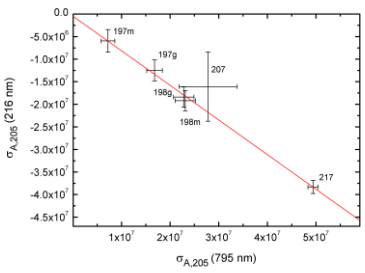


Fig.2. (Preliminary) King plot for 216 nm and 795 nm transitions in astatine.

The $\Delta\sigma$ value for a specific transition (e.g. 216 nm or 795 nm) is defined as: $\Delta\sigma_{A,A'} = \Delta\nu_{A,A'} \cdot \frac{A \cdot A'}{(A - A')}$, where $\Delta\nu$ is the isotope shift between two isotopes with masses A and A'.

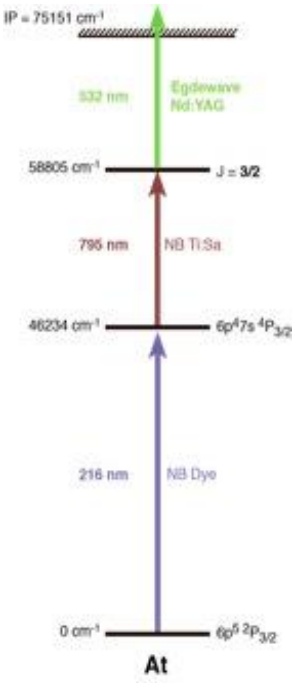


Fig. 3 Ionization scheme for astatine.

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 withthe similar onset of deformation in the lightest astatine isotopes, as in polonium. Note also the shape staggering in ^{197}At 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 ^{198}At, 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 $^{193-196}$At, which were not yet measured.

Therefore, the main goal of this part of the Addendum is to measure HFS spectra for alpha-decaying isotopes $^{193-196}$At (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 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 shifts.

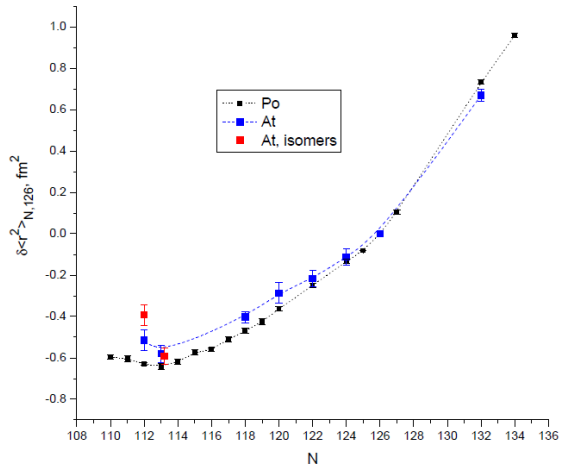


Fig.4. Comparison of deduced relative charge radii for astatine isotopes with the values for polonium. Data are normalized at N=126. Note the kink at N=126, and the onset of deformation in the light astatine isotopes similar to polonium. The two values shown for ^{197}At (N=112) and ^{198}At (N=113) correspond to two long-lived alpha-decaying nuclear states in each nucleus. Note the shape coexistence in ^{197}At 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 βDF studies of ^{194,196}At we refer to the IS534 Proposal [1]. In the May's 2012 campaign at GPS with lasers in the broadband mode, we performed successful βDF measurements for both isotopes, in which rates of ~40 ff/h and 8 ff/h fission events were observed, respectively. For ^{196}At, for which only one long-lived (ground) state is known, the βDF program is now considered as complete. An unexpected multi-modal energy (thus, also, mass) distribution of fission fragments for βDF of ^{196}At was preliminary deduced, the data analysis has been finalized at York and KU Leuven, two papers are in preparation.

Similar data were obtained for βDF of ^{194}At. 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 βDF [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 βDF branching ratios. To resolve this issue and to deduce the fission fragments mass distribution for each isomer, we need a dedicated βDF 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, thus ~15-20 ff/h for ^{194}At is expected now.

Therefore, we request 3 shifts to complete the βDF 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 17 shifts requested for the HFS measurements above. Afterwards, two dedicated βDF measurements will be performed at the respective laser frequencies,

each 1.5 shift long to reach statistically significant number of fission events **(>100),** or to prove non-observation of βDF for one of the isomers.

While studying the isomerism in neutron-deficient astatine 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. $^{193\text{-}196,199,200,201,218,219}$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 16.5 (+3.5 shifts already available) shifts of astatine beam time, in one running period. The requested beam time is based on our experience from May/October 2012 experiments, see Section IV.

We would also like to mention that, provided the astatine part is also approved, we could consider to run both astatine (20 shifts) and gold measurements (18 shifts approved) in one running period, using a single target unit. This would be a very effective solution and reduce the setting-up time for the ISOLDE/RILIS/target team as well as for the users teams.

Section IV. Specific comments on the issues raised by the INTC in June 2013.

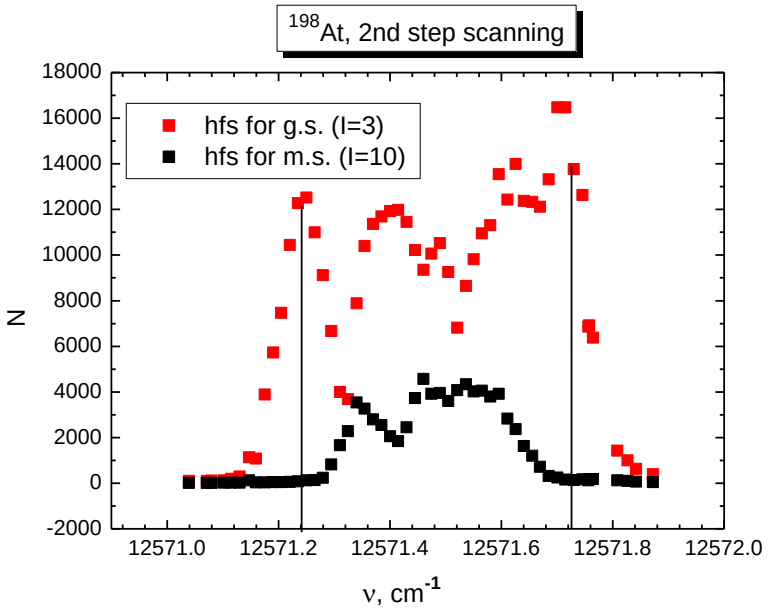
1. **"…**the justification for the number of required shifts was found to be insufficient."

   The number of requested shifts for HFS is based on our experience with the HFS measurements in the long chains of the gold, thallium, lead, polonium and astatine isotopes. Typically, one scan across the whole HFS spectrum for a single astatine isotope takes about 2 hours. To achieve the necessary level of precision, we need 3 scans per isotope (thus, 2x3=6 hours). Additionally, regular scans of the reference isotope ^{205}At are essential in order to account for the absolute frequency drift of the laser wavelength meter during the run. This allows to measure isotope shifts as the frequency difference between the reference isotope and the isotope in question. Last, but not least, changing from one mass to another at the HRS takes typically ~15-20 minutes, which across the whole run for all isotopes, plus references measurements, adds up to nearly 1 shift.

   Thus, on average, slightly less than 1 shift is required per isotopes. We wish to measure 15 isotopes not measured in the October 2012 run: ^{193,194,195,196,199,200,201,202,204,206,208,210,212,218,219}At, this leads to the necessity to use **14 shifts** for these HFS measurements. In addition we have to re-measure ^{211,209,203}At. These isotopes were measured in October 2012 run, but only with the first step scanning, therefore their HFS constants were determined with much larger

errors than those of ^{205,207,217}At. This is because the HFS of the first step transition proves to be substantially more narrow than that of the second step transition. At the same time, ^{209}At and ^{211}At should be used for μ and Q- momenta calibration as there are reliable literature data for electromagnetic moments of these isotopes obtained by the TDPAD method. Therefore the uncertainties of the HFS constants of ^{211,209}At will determine the final accuracy of μ and Q for all other isotopes. Moreover, these measurements will give additional points for the King-plot and as a consequence, decrease the uncertainties for the *F* and *M* electronic factors (now we have only 6 points on the King-plot see Fig. 2). Thus additional **3 shifts** are needed, giving us in total **17 shifts for the HFS measurements.**

As mentioned on p.6, we need **3 shifts** for the βDF measurements in the narrowband mode for ^{194}At. This enables accumulation of the necessary statistics and answer the very important question whether both or only one of the isomers undergo βDF.



It is worth to note that during the October 2012 run we have checked the isomer selectivity, needed in the βDF measurements for ^{194}At, by the exploring the HFS in $^{198.}$At. It is clearly seen from the Figure on the left, that e.g. at the 2nd step frequency of 12571.24 cm^{-1} or 12571.72 cm^{-1}, we can obtain a pure beam of the ^{198}At in the g.s., without high spin isomer admixture.

2. ***"…some technical issues appeared concerning the yields of the lightest astatine isotopes, and the problems are still not resolved"***

We believe we addressed this issue already in our 2012 Addendum (see p.5 of the present text, not changed since May 2012), but it seems our message was not clear enough, therefore we will re-iterate our arguments here.

In total, over 2011-2012, we had three astatine campaigns. In the 2011 and May 2012 runs, the consistently 'normal/expected' intensity was obtained. In contrast to this, in the latest (October 2012) campaign, the yield was ~5-10 times lower, than in the previous two campaigns. However, surprisingly, the yield became 'normal' (comparable to the first two campaigns) in the very last shifts of the October 2012 run.

At present, we still cannot determine the definite reasons for this, but *the fact itself, that finally the rate became normal shows, that our expected yields are correct, thus all the goals of the Addendum are fully achievable.*

Several possible ideas are, however, available, which might explain the lower intensity. Some issues were noted with the ISCOOL transmission in that particular run, which could be

responsible for a part of the observed intensity decrease. Due to the ISCOOL modifications, better control can be expected now.

At least partly the intensity drop may be explained if a not fully accurate tuning of the resonance transitions of the 3-step scheme happened. Namely, when the 2nd step frequency was scanned in the narrow bandmode, the 1st step broadband laser was used with the bandwidth of nearly 10 GHz (all values quoted in this paragraph are before the frequency tripling). At the same time IS (217-197) = 13GHz. Moreover, the HFS for the 1st transition is of the order of 5-8 GHz. So isotope-dependent tuning of the 1st (broadband) frequency is necessary to avoid an efficiency decrease and spectrum distortion. During the October 2012 run we did not know the exact optimal 1st step frequency values as these were the first ever HFS/IS measurements for the astatine isotopes. Now we know these frequencies with sufficient accuracy to guarantee a correct setting of the laser frequencies. Based on this experience, we will implement a more rigorous procedure for monitoring the laser tuning.

References:

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[6] H. Kettunen et al., "alpha -decay of the new isotopes ^{191,193}At", Eur. Phys. J., A17, 537(2003)
[7] A.Andreyev et al., "βDF of ^{192,194}At" Phys. Rev. C., 87, 014317 (2013)

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 MR-ToF	Existing	Used in several previous experiments, e.g. IS387,IS407, 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 andremoving 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	^{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