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Letter of Intent to the ISOLDE and N-ToF Experiments Committee (INTC)

Development of astatine ion beams with RILIS

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Abstract

A number of physics groups showed strong interest in the development and use of isobarically pure astatine beams, both at the neutron-deficient and neutron-rich sides. Elemental selective ionization with the RILIS laser ion source promises strong suppression of Pb, Bi, Po and Rn isobars for the At isotope mass range of interest. For this purpose laser ion source development for astatine is proposed, implying preparatory atomic spectroscopic work distributed over several slots of proton beam time.

1. Introduction

Spallation production of astatine isotopes requires an actinide target, i.e. a thorium or uranium compound. At present only ISOLDE at CERN provides routine use of actinide targets in combination with high energy proton beams with intensities >1 μA and a resonance ionization laser ion source.

Astatine beams were produced previously at ISOLDE-SC with the non-selective "hot plasma" ion source (MK5) and a UC_x/graphite target. Astatine isotopes down to ^{196}At (0.3 s) have been observed [1]. More neutron-deficient astatine isotopes are also produced, but could not be measured due to the overwhelming isobaric background of mainly polonium and bismuth isotopes. With the RILIS such background is eliminated nearly completely and only some thermally ionized thallium remains.

In the mass range of interest (A = 194-198) the thallium isotopes are relatively long-lived and decay mainly by EC decay. In recent spectroscopy experiments with laser-ionized neutron-deficient polonium and bismuth isotopes at ISOLDE [2, 3] the isobaric background of thallium isotopes was not significantly restrictive and we expect the same to be the case for astatine.

The isotopes $^{204\text{-}210}$At were also observed as beams from a Pb/Bi target with medium temperature transfer line and plasma ion source at ISOLDE [4]. They were produced in direct ^{209}Bi(p,π^-xn)$^{210-x}$At reactions as well as in secondary ^{209}Bi(^{3}He,xn)$^{212-x}$At and ^{209}Bi(^{4}He,xn)$^{213-x}$At reactions with spallation-produced ^{3}He and ^{4}He respectively. Such a target is not suitable for production of very neutron-deficient astatine isotopes but the release measurements show that already at 600° C a significant proportion of the astatine isotopes was released. Hence, as is the case for its lighter homologues bromine and iodine, astatine is relatively rapidly released and ISOL beams of very short-lived astatine isotopes can be produced.

Isobarically selective ionization of astatine at ISOLDE can be performed with two ionization techniques: negative surface ionization [5] and resonance laser ionization [6].

Due to the uncertainty in the reproducibility and lifetime of the specialized negative surface ionizer elements, these are not yet suitable for routine use at ISOLDE. Exploring the capability of the RILIS for production of astatine beams is therefore required.

Unfortunately, the optical spectrum and ionization potential of astatine so far are unknown. Only two optical lines of astatine have been experimentally observed in the absorption spectrum of a 70 ng astatine sample [7]. Therefore, methodical investigations to identify optical excitation and ionization schemes with reasonable efficiency are mandatory. The scope for off-line development work is limited due to the lack of stable astatine isotopes as well as long term apparatus contamination, so all laser-spectroscopic preparatory work must be performed on-line, e.g. by using in-source spectroscopy. Optical excitation and ionization can be developed on any astatine isotope so the favored isotope for these studies may be freely chosen based on factors such as available yield, absence of isobaric interference or preferred detection technique, e.g. via its emitted radiation. The methodology and experimental set-up will be similar to the successful investigation of the laser ionization schemes of polonium [2].

Such steps towards the identification of a RILIS ionization scheme for astatine will provide valuable information for the laser ion-source TRILIS at TRIUMF, Vancouver, Canada where an ambitious program involving astatine beam production is underway [8]. Thus a close scientific collaboration with joint development steps and direct exchange of data and information is foreseen. The collaboration with TRIUMF is strictly limited to laser spectroscopy and RILIS development.

2. Physics Cases

So far, a number of physics groups showed strong interest in the development and the use of the astatine beams, both at the neutron-deficient and neutron-rich sides.

Five main areas of interest could be identified at this moment (in no particular order):

a) Studies of the beta-delayed fission (βDF) in the lead region (Paisley-Leuven-Bratislava-Geneva-Gent-Liverpool-Los Alamos-Tokai- collaboration)

b) HFS, IS and charge radii measurements within the long chain of At isotopes, from the very neutron-deficient side, across the N=126 neutron closure, up to the most neutron-rich isotopes (Paisley-Leuven-Bratislava-Liverpool - collaboration)

c) Shape coexistence in the lightest Po isotopes (β^+/EC -decay products of At), in particular the search for coexisting oblate, prolate and spherical 0^+ band-heads and corresponding excitations in the odd-A Po isotopes (Paisley-Leuven-Bratislava-Liverpool-JYFL-Warsaw - collaboration)

d) Search for octupole collectivity in the neutron-rich Rn isotopes (beta-decay products of At) (Madrid, Oslo, Uppsala, Warsaw - collaboration)

e) Few-nucleon transfer reactions of At isotopes to study single-particle around N=126 and multi-particle multi-hole structures in the neutron-deficient and neutron-rich isotopes. This would need beam energies from HIE ISOLDE.

Due to the space limitation of this LOI, only a brief overview of each planned campaign is provided below. Provided the astatine beams become available in the near future, a dedicated proposal will be written at a later stage for each of the proposed experiments.

a) Studies of the Beta-delayed fission in the lead region (Paisley-Leuven-Bratislava-Geneva-Gent-Liverpool-Los Alamos-Tokai - collaboration)

In July 2008, a very exotic phenomenon – the low-energy beta-delayed fission of ^{180}Tl ($T_{1/2}$~1 s) was successfully studied by our collaboration using the RILIS tuned to thallium. In this two-step nuclear process a parent nucleus first undergoes β^+/EC decay, populating only states up to the electron-capture Q value Q_{EC} in the daughter nucleus which then may fission. A beta-decaying parent nucleus with a Q_{EC} value comparable to the fission barrier height (B_f) of the daughter nucleus can exhibit βDF. The investigation of the beta-delayed fission in the very neutron-deficient lead region is of special interest because it allows studying the low-energy fission properties of exotic nuclei, possessing

unusual neutron to proton ratios (e.g. N/Z=1.25 for ^{180}Hg studied in this work, as contrasted to a typical ratio of N/Z=1.55-1.59 in the uranium region). Due to extreme neutron deficiency of the nucleus undergoing fission, a few unexpected phenomena can be expected in this region of nuclei in comparison with the "classical" fission in the actinide and trans-actinide regions.

The power and uniqueness of ISOLDE for this study was that the combination of RILIS+ISOLDE provided an extremely pure ^{180}Tl beam with an intensity of ~150 ions/s (which presently is not accessible anywhere else). Both the purity and the intensity were crucial for our measurements, as the βDF probability, deduced by us, is very low: $P_{\beta DF}(^{180}Tl)=3.6(7) \times 10^{-3}$ %. Furthermore, the newly determined bDF branching ratio was two orders of magnitude larger compared to the previous value and agreed with the systematic trend of branching ratios as determined for the actinide isotopes. (For comparison, we mention that in our ISOLDE experiment, the fission counting rate was ~20 fissions/h, while in a complementary experiment at the state-of-art velocity filter SHIP (GSI), the fission counting rate was only 2 fissions/day).

The most surprising result however was that in contrast to a widely-expected symmetrical mass split of ^{180}Hg (the daughter of ^{180}Tl after the beta decay) in two semi-magic ^{90}Zr fragments, an asymmetric mass-split was observed as shown in Fig.1.

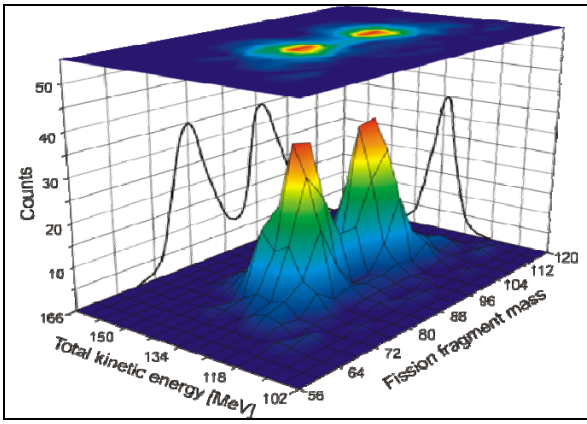

Figure 1. The derived fission-fragment distribution of ^{180}Hg as a function of the fragment mass and the total kinetic energy.

Meanwhile, the analysis of this experiment was completed and a manuscript is prepared for a submission to Nature. In this paper, we demonstrate that this ***is a new type of asymmetric fission***, ***not governed by large shell effects related to doubly-magic fragment structures***.

For ^{180}Tl nuclide and possibly other nuclei in the very neutron-deficient lead region, observation and further study of this effect is so far only feasible through beta-delayed fission measurements.

That is why we proposed an extensive program to study a number of other nuclei in the very neutron-deficient lead region. A proposal to study ^{178,182}Tl was already approved by INTC in 2008 (we expect the experiment to be run in 2010).

As a further extension of this program, we want to move to the very neutron-deficient ^{192,194,196,198}At (half-life range $T_{1/2}$~90 ms- a few seconds), for which the beta-delayed fission is also expected.

Earlier experiments aimed at the βDF studies of ^{192,194,196}At used complete fusion reactions with heavy ions as a production method and suffered from the relatively low yield (e.g., only 2 fissions/h for ^{194}At at SHIP, GSI), the impossibility to measure the fission fragment distribution and, importantly, from the contaminant nuclei, produced in different evaporation channels of the studied reactions.

We expect, that similar to the case of ^{180}Tl, the use of RILIS+ISOLDE will be provide the unique pure and intense beams of ^{192,194,196,198}At, which will allow to perform their dedicated βDF studies.

b) HFS and IS (charge radii) measurements within the long chain of astatine isotopes, from the very neutron-deficient side, through the N=126 neutron closure, up to the most neutron-rich isotopes (Paisley-Leuven-Bratislava-Liverpool - collaboration). Shape coexistence in the lightest At isotopes.

Recently, our group performed a series of HFS and IS (charge radii) measurements for the long isotopic chains of lead [9] and polonium isotopes [10], see Fig.2. A few important phenomena, such as the confirmation of the sphericity of the lightest lead isotopes in the vicinity of the neutron mid-shell at N=104 and an unexpectedly early onset of strong deformation in the neutron-deficient polonium isotopes (red curve in the plot), were clearly demonstrated by these data. Furthermore, for polonium isotopes, a kink in the charge radii when crossing the N=126 shell closure was found in the first time.

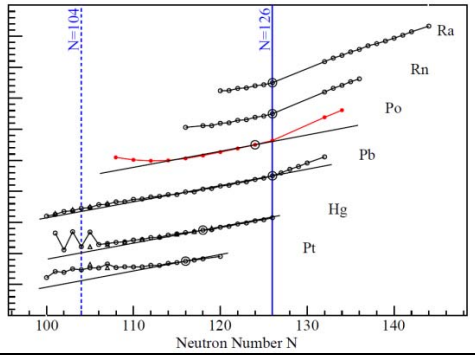

Figure 2. The systematic of the nuclear charge radii for the even-Z elements from Pt up to Ra.

Related to above, the recent in-beam and α-decay studies at the RITU gas-filled separator at JYFL and at the velocity filter SHIP (GSI) provided the first evidence on the onset of the At ground state deformation by approaching the mid-shell at N=104, in the isotopes $^{192\text{-}197}$At [11, 12, 13]. The studies of odd-Z nuclei are especially important for tracking the development of shape coexistence and deformation, as they allow to directly investigate the deformation-driving effect of a single valence nucleon [14, 15].

We expect that these isotopes should become accessible for the HFS/IS measurements with RILIS+ISOLDE, which will provide direct confirmation of the shape coexistence

phenomenon in the At nuclei and essential information complementary to the in-beam and beta decay information on the spin and parity (through HFS measurements) of the ground- and isomeric states.

c) **Shape coexistence in the lightest polonium isotopes (β^+/EC -decay products of astatine), in particular the search for coexisting oblate and prolate 0^+ band-heads and corresponding excitations in the odd-A polonium isotopes (Paisley-Leuven-Bratislava-Liverpool-JYFL-Warsaw - collaboration)**

Shape coexistence is a well-established phenomenon in the lightest polonium isotopes (see, e.g. [14, 15]), in which the coexistence at low energy of spherical, oblate and prolate configurations was established by several complementary methods (e.g. particle and in-beam decay spectroscopy, isomer spectroscopy, charge radii measurements).

One of the well-established methods to study excited coexisting configurations in the polonium isotopes is the beta decay of the parent astatine nuclei, see e.g. the study [16]. In this work, performed at the LISOL mass separator, a detailed study of the excited states in ^{200,202}Po has been done, whereby the evidence for the presence of π(4p-2h) intruder configurations was obtained.

With the development of the neutron-deficient astatine beams at ISOLDE, we will be able to extend such studies to even more neutron-deficient polonium isotopes. As a particular example, via the beta decay of ^{194}At we expect to identify the long-searched excited 0^+ state in ^{194}Po, being the band-head of the known oblate band in this nucleus. Furthermore, the scarcely-known excited states (including the intruder band-heads) in the odd-A isotopes $^{193\text{-}197}$Po will be accessible for studies.

d) **Search for octupole collectivity in the neutron-rich Rn isotopes (beta-decay products of At) (Madrid, Oslo, Uppsala, Warsaw - collaboration)**

The extensive experimental studies of isotonic and isotopic sequences of nuclei around A=225, performed at ISOLDE facility, were focused on the signatures of reflection asymmetric octupole shape, such as the rotational bands of alternating parity states, connected by fast E1 transitions [17]. The aim of the IS322 and IS386 experiments was to investigate the limits of the "island" of the octupole instability in the actinide region. The study of transitional nuclei in the upper border of this region is of relevance to understand the interplay of octupole and quadrupole collectivities. These experiments have provided the first information on the absolute values of B(E1) in this octupole transitional region. As an example, some particularly fast B(E1) transition rates observed in ^{231}Ac suggest that the octupole effects, although weak, are still present in this exotic nucleus [18].

Recently, high-spin octupole yrast levels in ^{216}Rn were studied and obtained data show that it is clearly a transitional nucleus [19]. The lightest nucleus beyond ^{208}Pb showing evidence of octupole collectivity at low spins is still ^{216}Fr [20].

We intend to search for octupole correlations in the neutron-rich Rn isotopes (beta-decay products of astatine). Studies on heavy radon nuclei are strongly complementary to the IS322 and IS386 projects. The obtained data will contribute to understanding the onset of octupole collectivity in the light actinide region.

e) **Few-nucleon transfer reactions of astatine isotopes to study single-particle around N=126 and multi-particle multi-hole structures in the neutron-deficient and neutron-rich isotopes.** This would need beam energies from HIE ISOLDE.

With the higher energies from HIE-ISOLDE, few-nucleon transfer reactions will become possible. These studies can probe the single particle character of the Po, At and Rn isotopes near the N=126 closed neutron shell, as well as e.g. multi-particle – multi-hole configurations in neutron-deficient bismuth isotopes using two-proton removal reactions.

3. Laser ionization of astatine

The task of identification and characterization of suitable ionization schemes can be divided into three stages, one for each individual excitation step of a three step ionization ladder typically used for multi-resonant excitation schemes.

Stage 1: **Verification of the only two transitions known for At**.

For astatine only two optical resonance lines are reported, lying in the UV spectral region and tentatively assigned to transitions starting from the ground state. The wavelengths were determined by optical absorption spectroscopy as 216.225 nm and 224.401 nm [7]. Corresponding laser radiation can be produced either with the existing ISOLDE RILIS dye laser system by frequency tripling or alternatively by frequency quadrupling of radiation from the ISOLDE Ti:Sapphire laser which is currently under construction. The achievable laser powers will be comparable. From the excited state, atoms will be ionized non-resonantly by a frequency doubled dye laser at ~280 nm or by a frequency quadrupled Nd:YAG laser at 266 nm, which presumably will provide sufficient energy to exceed the first ionization potential of astatine. This value has not yet been measured, but there are several estimates and ab-initio calculations published: 10.4 eV [21], 9.2±0.4 eV [22], 9.86 eV [23]. The corresponding simple two step ionization scheme is shown in Fig. 3. To verify the energy positions of the two excited levels, wavelength dependent ion signals with typical time structure for photoionization will be recorded and shape and widths of resonances will be analyzed. Once laser ions are successfully detected, saturation measurements will be carried out, providing information on the achievable ionization efficiency of the schemes. Since the two UV lines are close, measurements can be performed rapidly with both dye lasers and Ti:Sapphire lasers.

At this stage, following successful localization of the excited astatine levels, a wavelength scan of the frequency doubled dye laser used for ionization of excited astatine atoms can be performed. An abrupt decrease in the ionization efficiency above certain wavelength will indicate for the first time the position of the astatine ionization potential.

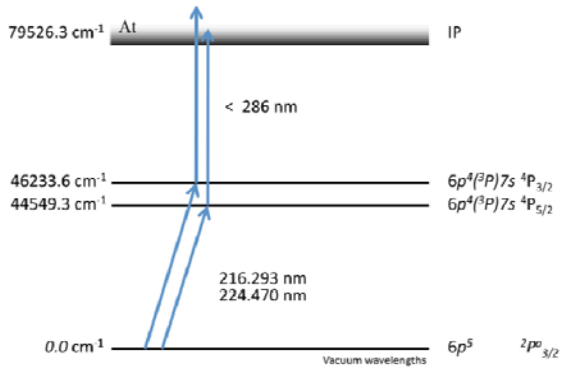

Figure 3: Sketch of the simple two step excitation for level verification within Stage 1.

Stage 2: **Development of refined RILIS schemes.**

The next steps towards useful astatine laser ion beams will strongly depend on the success of the test of the two reported lines as performed in Stage 1.

2a. Search for second step transitions.

Successful confirmation of the first two resonant steps given in literature through detection of resonant laser ions would enable a further investigation for the development of a three step ionization scheme to be started. Such a scheme is desirable since, for a highly excited atom, an ionization step using laser radiation in the green to infra-red range, for which a high laser powers are achievable, can be used. For this purpose resonant second excitation steps which can be excited efficiently must be identified. For the final ionization step fundamental laser light with high power from a dye laser, a Ti:Sapphire laser or a synchronized pump laser can be used. The most simple approach would be non-resonant ionization using the 355 nm or 532 nm output of the Nd:YAG pump laser. With no spectroscopic information to indicate the location of excited states of astatine in the range of higher excitation energy, a broad wavelength scan from the blue to infrared region has to be performed on top of one of the first excitation steps, as indicated in the scheme of Fig. 4.

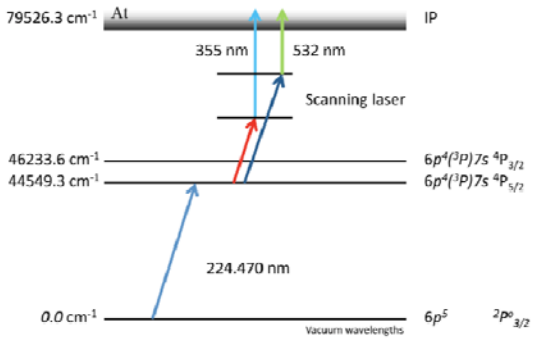

Figure 4: Sketch of the search for a three step ionization scheme.

2b. Search for first step transitions.

If the first resonant steps could not be verified during Stage 1, an extensive search for excited atomic levels in the range around 43000-47000 cm^{-1} must be performed to localize suitable first excitation steps. This would require a scan in the UV wavelength range. This could be performed using the ISOLDE dye lasers pumped by the 355 nm output of the Nd:YAG laser to generate a fundamental laser frequency in the blue range which could then be frequency doubled. This approach has not yet been tested but the specified dye laser performance for this mode of operation indicates that this is feasible. Once a first step is indicated by this scanning the development of the final ionization scheme will further proceed with the search for higher lying energy levels according to step 2a.

Stage 3: **Search for third step transitions and determination of the ionization potential of astatine**

As discussed above the basic quantity of fundamental relevance for the astatine atom, its first ionization potential (IP), has not been measured so far. Members of the collaboration have the expertise of the determination of this parameter. Based on an efficient LIS scheme involving two consecutive excitation steps through bound atomic states in At I, which have been identified and characterized within Stages 1 and 2, scanning of the third step laser across the energy range around the expectation value of the IP, should deliver high-lying Rydberg levels as well as auto-ionizing resonances. First of all, the laser excitation into such resonances will significantly enhance the ionization probability. Hence the overall ionization efficiency of the RILIS for astatine is most positively affected. On the other hand, the precise determination of the energetic positions for individual members of a Rydberg series, which converges towards the first IP according to the simple Rydberg-Ritz formula, will allow for a first time precision determination of the first IP for astatine. Apart from its relevance as fundamental parameter, this value will contribute for a better understanding and interpretation of the so far unknown atomic structure of the atomic spectrum of this heaviest halogen element known.

4. Conclusion

In order to carry out the atomic spectroscopy study of astatine needed to collect data for development of efficient RILIS ionization scheme, we require 36 shifts of on-line beam time distributed over two or three runs. The required time depends on the success of the very first and relatively short run, which will be devoted to confirmation of the existing literature data on first step astatine transitions. The beam time request is shown in following table.

Run #	Beam	Target material	Ion Source	Shifts
1 - stage 1	At	UCx	RILIS	6
2 – stage 2	At	UCx	RILIS	12
3 – stage 3	At	UCx	RILIS	18

For detection and identification of astatine isotopes we intend to install the α detection system that was used for in-source spectroscopy (IS456) of polonium, preferably at LA1 beam line. In the absence of surface ionized isobars the laser ions will be detected with the MCP-detector which is already installed at the RA0 beam line.

Since the wavelength of the one of expected first step transitions is very close to that used for zinc excitation, it could be reasonable to attach the first astatine run to a planned zinc run with the use of RILIS.

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