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New developments of the in-source spectroscopy method at RILIS/ISOLDE

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ABSTRACT

At the CERN ISOLDE facility, long isotope chains of many elements are produced by proton-induced reactions in target materials such as uranium carbide. The Resonance Ionization Laser Ion Source (RILIS) is an efficient and selective means of ionizing the reaction products to produce an ion beam of a chosen isotope. Coupling the RILIS with modern ion detection techniques enables highly sensitive studies of nuclear properties (spins, electromagnetic moments and charge radii) along an isotope chain, provided that the isotope shifts and hyperfine structure splitting of the atomic transitions can be resolved. At ISOLDE the campaign to measure the systematics of isotopes in the lead region (Pb, Bi, Tl and Po) has been extended to include the gold and astatine isotope chains. Several developments were specifically required for the feasibility of the most recent measurements: new ionization schemes (Po, At); a remote controlled narrow line-width mode of operation for the RILIS Ti:sapphire laser (At, Au, Po); isobar free ionization using the Laser Ion Source Trap, LIST (Po); isobar selective particle identification using the multi-reflection time-of-flight mass separator (MR-ToF MS) of ISOLTRAP (Au, At). These are summarized as part of an overview of the current status of the in-source resonance ionization spectroscopy setup at ISOLDE.

1. Introduction

The study of nuclear ground state and isomer properties using the technique of laser spectroscopy of atoms and ions has existed

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for over 30 years [1]. The measurement observables: the *hyperfine structure* (*HFS*) and *isotope shift* (*IS*) of an electronic transition contain information about the nuclear properties such as spins, electromagnetic moments and nuclear charge radii. Recently published reviews [2,3] of the measurements in this field summarize the methods that have been employed at various facilities worldwide. Most of the work reported is sub-Doppler collinear laser fluorescence spectroscopy of fast atom or ion beams. Typically single mode continuous wave (CW) laser light is used, thereby achieving a spectral resolution that approaches the natural linewidth of the electron transition. Resonance Ionization Spectroscopy (RIS) is an alternative approach that relies on the production and detection of photo-ions during a laser scan, rather than the detection of the fluorescence photons. The higher efficiency of ion detection, particularly through the use of modern particle detection techniques based on characteristic decay radiation, makes this method highly sensitive and therefore preferable for the study of exotic isotopes with low production rates. The Resonance Ionization Laser Ion Source (RILIS) [4,5] of the ISOLDE facility at CERN can be adapted from its usual role as an efficient and element selective ion source, to become a powerful tool for performing these measurements [6]. The first application of this technique, which is known as In-Source Resonance Ionization Spectroscopy, is reported in [7].

At ISOLDE exotic isotopes are produced in nuclear reactions that occur when the 1–1.4 GeV proton beam (up to 2 μA) from the CERN PS-Booster interacts with the ISOLDE target. Through the Isotope Separator On-Line (ISOL) process [8], these reaction products are ionized, extracted and mass separated as an ion beam which can be immediately transported to an ion detector or decay spectroscopy station. The highest yield of the spallation products, Au, At and Po, are achieved using a UCx target, which typically contains 50 g/cm^2 of depleted uranium (99.6% of ^{238}U). The ion rate is proportional to the proton beam intensity and the ionization and extraction efficiencies.

Atoms with a low ionization potential can be efficiently ionized upon contact with the heated surfaces of the target, transfer line or surface ionizer cavity. When the RILIS is used, these surface ions are a potential source of isobaric contamination in the mass separated beam. Laser ionization takes place when the atomic vapor interacts with the laser beams, which are precisely wavelength tuned to the transition energies of a chosen ionization scheme. Only the atoms with an atomic energy level structure that can be resonantly excited by the RILIS lasers will be ionized. The ionization process is therefore completely element selective. The broad-band (BB) RILIS laser-linewidth of 10–20 GHz is often larger than IS or HFS effects. In this case all isotopes and isomers of the chosen element will be ionized with roughly equal efficiency.

For in-source resonance ionization spectroscopy one of the transitions of the ionization scheme is chosen for laser scanning in a 'high resolution'mode using a narrow-band (NB) (≈1 GHz) laser. The other laser beams are at a fixed frequency and provided by broad-band (BB) lasers, optimized for maximum ionization efficiency. In this case the precision of wavelength tuning is sufficiently high so that a degree of isotope or isomer selectivity is achieved, hence the suitability of the RILIS technique for the study of isotope shifts and hyperfine structures by performing a laser scan of the spectroscopic transition while recording the photo-ion production rate. Fig. 1 is an illustration of the in-source RIS technique showing the ion production process and the three detection systems that are discussed in this article.

The standard laser/atom interaction region of the RILIS is the hot (>2000 °C) cavity of the surface ion source. The spectral resolution is therefore limited by the Doppler broadening of atomic lines due to the thermal velocity distribution of the atoms in the high temperature environment. This is larger for the lighter elements but still exceeds 1 GHz for the heaviest isotopes that can be produced at ISOLDE. Despite this broad line-width limitation, sub-100 MHz accuracy has been demonstrated for HFS of Tl isotopes using the ISOLDE RILIS [9].

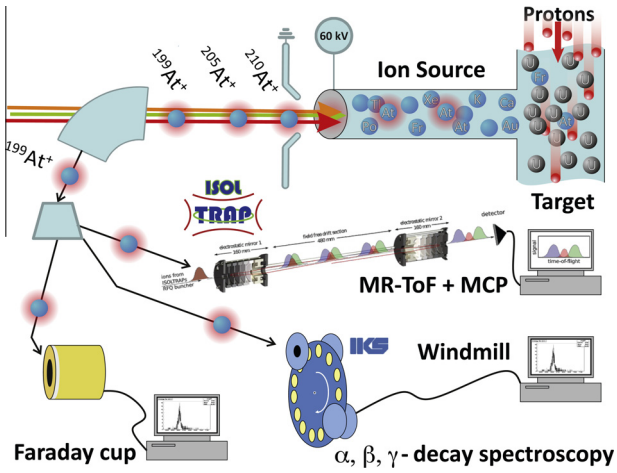

Fig. 1. An overview of the in-source RIS setup showing three photo-ion detection methods.

This technique has been successfully applied for the study of the nuclear spins and electromagnetic moments of Cu isotopes [10,11], the separation of Ag isomers, Be isotope shift measurements [12], and for the ongoing campaign to measure the systematics of the elements in the lead (Z = 82) region (Pb [13–15], Bi [16], Tl, Po [17,18], Au and At isotope chains). Several developments of the in-source RIS technique were required for the most recent of these measurements: New ionization schemes (Po, At); narrow-band mode for the RILIS Ti:Sapphire (Ti:Sa) laser (At, Au, Po); surface ion suppression using the Laser Ion Source Trap (LIST) (Po); isobar separated particle identification using the ISOLTRAP Multi-reflection Time-of-Flight Mass Separator (MR–ToF MS) (Au, At).

2. Ionization scheme developments

The study of the polonium and astatine isotope chains required dedicated atomic spectroscopy studies to determine suitable and efficient ionization schemes. The scheme development, described in [19] (Po) and [20] (At), was particularly challenging due to the absence of stable isotopes. For the same reason, the availability of atomic data was also limited, particularly in the case of astatine for which only two atomic lines were previously documented. Fig. 2 shows the polonium and astatine ionization schemes used for the in-source RIS measurements. The observation of four HFS components for the 795 nm transition of the $I = \frac{1}{2}$ isomer of ^{197}At (Fig. 3) enables an unambiguous assignment of $J = \frac{3}{2}$ as the atomic spin of the 58805 cm^{-1} level of At.

3. RILIS technical developments

Since 2007 the RILIS performance and capabilities have been gradually increased through a multi-stage upgrade of the laser system: replacement of the copper vapor laser with a Nd:YAG laser; dye laser upgrade; addition of complementary Nd:YAG pumped Ti:Sapphire laser system [21,22,5].

All users of RILIS ionized beams have benefitted from the resulting increase in laser ionization efficiency and improved reliability and stability. The aspects of the RILIS upgrade which are not described in the references above, and which were specifically aimed at the application of the RILIS in-source resonance ionization spectroscopy are presented here.

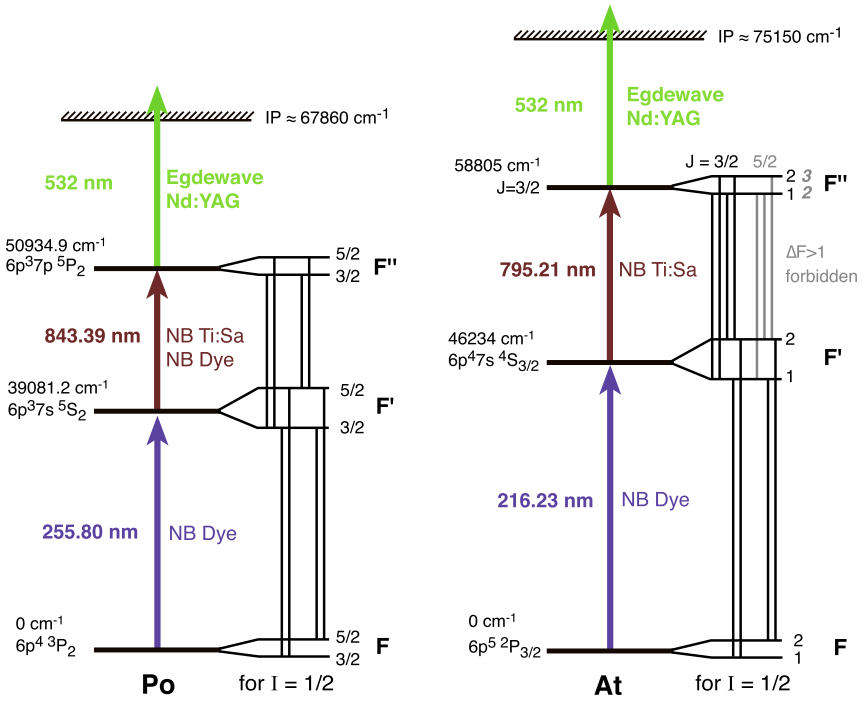

Fig. 2. The ionization schemes used for the study of polonium and astatine isotopes. The narrow-band (NB) scanning lasers used for the spectroscopic transitions are indicated. The HFS components (F values) and allowed transitions are shown for a nuclear spin $I = \frac{1}{2}$.

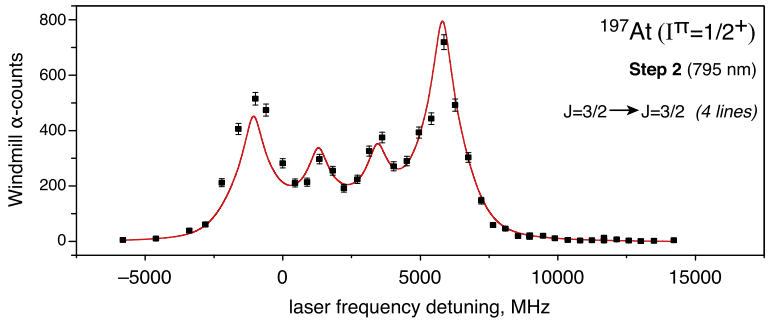

Fig. 3. A NB-TiSa scan of the second step transition for the ^{197}At, $I = \frac{1}{2}$ isomer using the Windmill α-detection system. Four partially resolved HFS components are seen, thereby excluding the possibility of a $J = \frac{5}{2}$ assignment for the atomic spin of the upper level of the transition (see Fig. 2).

3.1. Narrow line-width Ti:Sa laser

To approach the maximum measurement precision, equivalent to the ≈1 GHz Doppler broadened spectral line-width in the hot cavity, the line-width of the excitation laser should be similar to this value. The RILIS narrow-band dye laser was supplied by DMK Laser Microsystems and installed in 2010. It is a grating-tuned Littrow resonator with a laser line-width of 10–20 GHz. This is reduced to ≈1 GHz by placing an angle tunable ≈6 mm Fabry–Perot etalon inside the resonator cavity. This method for line-width reduction, which is first reported in [23] has been applied for all of the laser spectroscopy experiments using the RILIS dye lasers.

For the recent (2012) measurements of Po, At and Au isotopes, the NB mode of operation of the RILIS Ti:Sa laser was established by installing an angle-tuned intra-cavity thick etalon, in addition to the two existing frequency selective elements (birefringent tuner and thin etalon) of the Ti:Sa laser. The performance and characterization of the NB Ti:Sa is described in detail in [24]. The use of this laser for in-source RIS required motorization of the two etalons and a computer-controlled performance monitoring and optimization (power, line-width) and stabilization (wavelength). This is realized by the use of a new RILIS monitoring, control and DAQ system, as described in [25]. Fig. 4 compares the spectra obtained for ^{197}Au by scanning the third harmonic of the Ti:Sa across the resonance of the first step of ionization scheme in broad-band and narrow-band mode. The NB Ti:Sa was used for polonium (second step, 843 nm), gold (first step, third harmonic, 268 nm) and astatine (second step, 795 nm).

3.2. Laser ion source trap (LIST)

The study of the α-decaying ^{211m,212m2,213,217,219}Po isotopes by in-source RIS at ISOLDE is not possible if laser ionization takes place in the standard hot cavity environment. This is because of the abundance of surface ionized Fr isobars, which would saturate the α detectors and contaminate the detection setup. The isotopes ^{217,219}Po were studied for the first time in 2012 as a successful first demonstration of the application of the LIST for a physics experiment. The LIST device is based on a novel laser ion source concept first proposed by Blaum et al. [26]. It uses a surface ion repeller, positioned immediately downstream of the hot cavity, to create a surface ion free region in which the RILIS lasers can overlap with

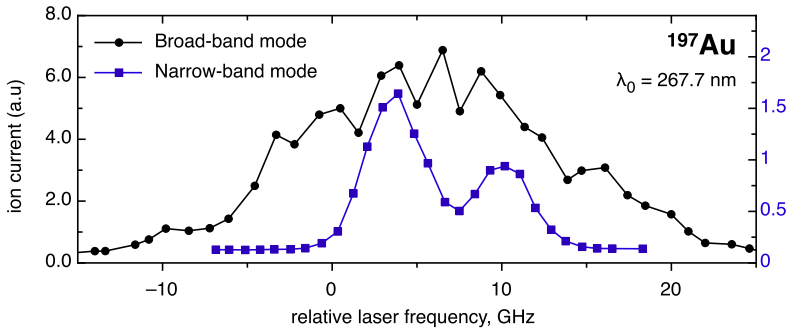

Fig. 4. A comparison between broad and narrow-band hyperfine structure scans of the ^{197}Au first step transition using the frequency tripled output of the Ti:Sa laser. In narrow-band mode the laser power is attenuated to avoid power (saturation) broadening of the atomic line, hence the reduced ion current.

the atom beam that exits the cavity. This region is surrounded by an RFQ ion guide structure and is shielded from the extraction electrode potential by an end electrode. Laser ions are therefore transversely confined as they drift towards the extraction electrode. The performance and characterization of the LIST during this experiment is described in detail in [27]. A surface ion suppression factor of approximately 10^3 was achieved along with an estimated 20× drop in laser ionization efficiency (due to the relatively poor spatial overlap of the laser beams with the atoms inside the LIST). The improved Po:Fr ion beam ratio, coupled with the high sensitivity and α-energy resolution of the α-detection system enabled laser scans of ^{216,217,218,219}Po to be recorded simultaneously.

Fig. 5 is a comparison of laser scans of the polonium second step transition (in narrow-band mode) for ionization inside the LIST or inside the hot ion source cavity.

4. Photo-ion registration

Due to the variation in production rates, decay properties and contamination ratios for ion beams of different members of an isotope chain, several complementary methods of photo-ion detection are required. The recent developments and suitability of three of these are described below.

4.1. ISOLDE Faraday cups

A Faraday cup (FC) is a standard beam diagnostics tool for ion current measurements. In 2010 an upgrade of the ISOLDE FC DAQ improved the sensitivity by an order-of-magnitude to ≈0.1 pA, corresponding to 6×10^5 ions/s. Prior to the upgrade all FC signals were connected to a common Keithley electrometer, multiplexed to read the largely varying signals from each FC in use. For laser scanning the analogue output of the picoammeter could be read by the RILIS control computer using a 12-bit ADC card. The new FC system uses a modular approach: a dedicated DAQ and pneumatic FC movement controller box is placed next to each FC to reduce electrical noise picked up by the signal cables. The device includes an electron repeller voltage supply and an amplifier using an AD549L and has an adjustable integration time (200 ms to 1 s). The specified dynamic range is 0.2 pA to 10 mA. Communication with the ISOLDE control system uses a Profibus interface. During the laser scan the FC is set to a fixed integration time, usually either 200 or 500 ms and the ion current is read from the ISOLDE Front-end Software Architecture (FESA) framework using the RILIS LabVIEW monitoring system [25]. For a continuous laser scan the FC integration time results in a systematic shift of the observed resonance which depends on the scanning speed of the laser. This effect has to be characterized so that the data are corrected accordingly. The use of discrete scanning steps with a hold time per step greater than the FC integration time avoids this issue. For radiogenic isotopes, coordinating scan steps with the proton pulse structures minimizes fluctuations in the total in-target isotope production for each laser wavelength position. The same principle is applied for laser scans using the other photo-ion detection methods. Due to the simplicity and speed the FC is the preferred detection method for intense beams of long-lived or stable isotopes with relatively high isobaric purity and for the frequent scans of designated reference isotopes during a measurement run.

4.2. The Windmill α-decay spectroscopy setup

For less intense beams of radioactive isotopes, their characteristic decay radiation can be used as a selective and efficient means of photo-ion detection. The ISOLDE tape station [28], a routinely used device for yield measurements at ISOLDE, is suitable for the measurement of β or γ active isotopes with half-lives above 100 ms. For shorter lived or α-active isotopes the KU Leuven Windmill α-decay spectroscopy set-up (Fig. 6) is used. This contains a rotating wheel of up to 10 carbon catcher disks (6–10 mm diameter) with a

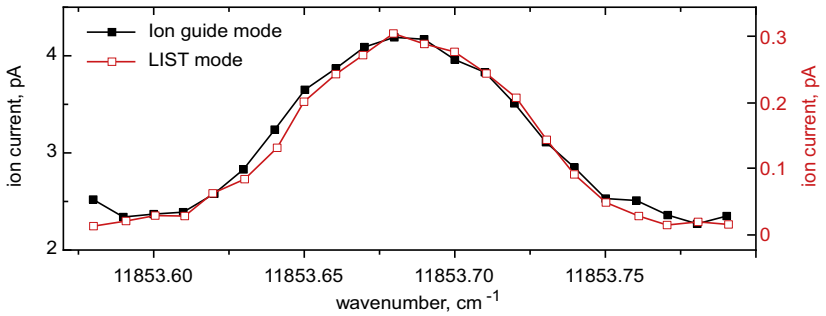

Fig. 5. A comparison of LIST and hot cavity RILIS scanning of the second step resonance for ^{208}Po.

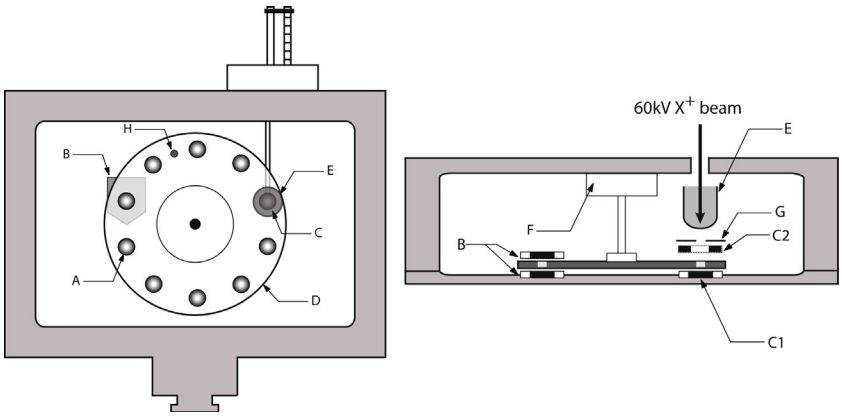

Fig. 6. The KU Leuven Windmill alpha detection device. Ten carbon disks (A) are mounted on a wheel (D) that can be rotated by a stepping motor (F). The incident beam current can either be measured with a movable Faraday cup (E) or can be sent through a collimator (G) onto the foil. A pair of silicon detectors (C1 and C2) measure the alpha spectrum during implantation on the foil, a second detector pair (B) can measure the activity of a previously implanted foil to verify its cleanliness before re-use. A 30–50 Bq ^{241}Am source (H) can be moved to the detector locations for calibration purposes.

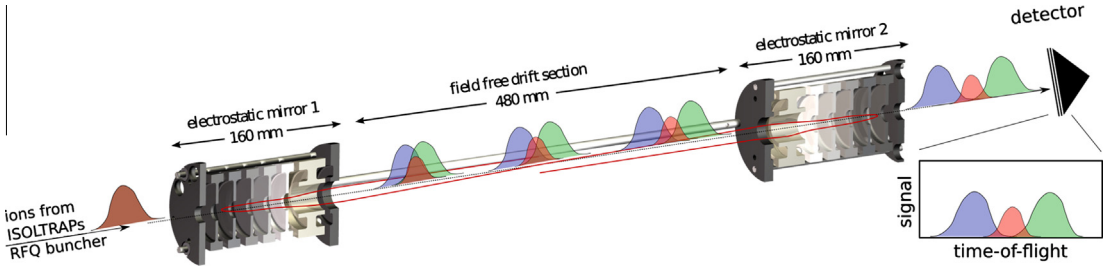

Fig. 7. An illustration of the ISOLTRAP MR–ToF MS, as presented in [32]. An ion bunch from the ISOLTRAP RFQ cooler-buncher oscillates between electrostatic ion-mirrors, thus extending the flight path to a length of several kilometers. Triggering the ion detector on the ejection time from the RFQ enables discrimination between different ion species (isobars) according to their mass-dependent flight time distribution.

thickness of 20 μg/cm^2, made at the GSI target laboratory [29]. At two of the 10 disk locations the wheel is surrounded by a pair of Si α-detectors, three of these are circular detectors with an active area of 300 mm^2, placed as close as possible to the catcher disk. The fourth, placed 4 mm in front of the implantation site is a larger diameter (24 mm) annular detector which allows passage of the ion beam, while maximizing the α detection solid angle (≈66%) for both detectors. Two germanium detectors can be placed outside the Windmill chamber for γ-ray detection: one at the position of the narrow-walled end flange and the other at an angle of 90°, to one side of the implantation site.

A transition from the original LynxOS and MBS DAQ [14] to the use of a COMET and Narval modules was made in 2007/8. During 2012 a XIA DGF-4C digital acquisition system, used for β-delayed fission studies [30], became the standard DAQ system for in-source spectroscopy. The efficiency of on-line data-analysis was improved through the transfer of the PAW based analysis routines to ROOT [31]. In 2012, the laser scanning process was streamlined through the use of a new laser scan control program that makes use of the LabVIEW based RILIS control infrastructure [25]. This enables synchronized exchange of RILIS and Windmill variables (laser wavelength, power, gated α count rate), the generation of a RILIS ready TTL signal and the plotting of live α-rate vs. wavelength spectra.

4.3. ISOLTRAP MR–ToF mass separator

The MR–ToF MS [32], shown schematically in Fig. 7 was recently added to the ISOLTRAP setup [33] for fast separation of ions of interest from isobaric contaminants. It accepts ion bunches from the ISOLTRAP linear RFQ cooler/buncher (Paul trap) and separates the isobar components according to the total flight time for their multiple oscillations within the electrostatic mirror system. The primary use of the device is the removal of contaminants in the ion bunch before injection into the dual Penning trap precision mass measurement system. This is achieved by time-of-flight separation of the isobars followed by fast gating (using a Bradbury Nielsen gate [34]). For in-source spectroscopy the physical isobar gating is not required and the ion bunch is sent to an ion detector (micro-channel plate or electron multiplier). If the DAQ is triggered on the ejection time of the ion bunch from the ISOLTRAP RFQ cooler, the isobar components of the ion beam are detected as discrete, isotopically pure ion packets, separated by their flight time to the detector. An example is shown in Fig. 8a). In this case the ion beam contains laser ionized ^{207}At (which is identified by its absence from the laser *OFF* TOF spectrum) and the surface ionized ^{207}Tl and ^{207}Fr components. The total flight time is some tens of ms and the ion beam components are distributed over several μs. The mass resolving power achieved is of the order of 10^5.

The MR–ToF MS is therefore a fast and selective device for obtaining background-free laser scans from low purity ion beams. Additionally, the use of an ion detector means that the detection efficiency is independent of the radioactive decay mode or half-life. Effective use of the MR–ToF MS with the RILIS scanning system required the implementation of remote control of the measurement cycle, initiated by the RILIS laser scan step 'ready' signal. The gated ion signal was made available to the RILIS control PC as an additional shared variable within the RILIS LabVIEW monitoring system. This enabled on-line plotting of the count rate during a laser scan. Fig. 8**b**) is an example of a MR–ToF scan of ^{207}At. In this

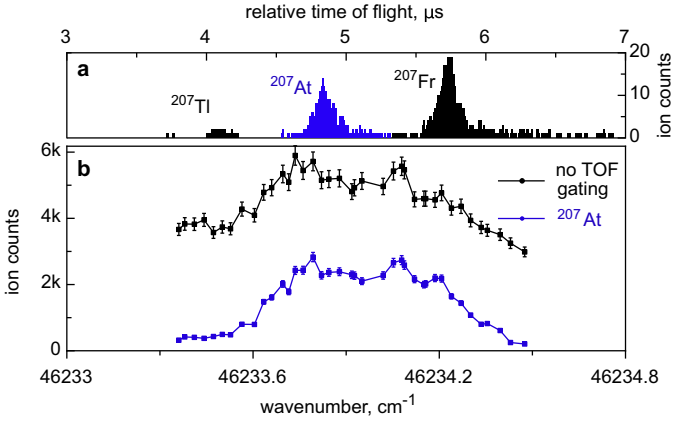

Fig. 8. (a) The MR–ToF spectrum for mass 207 with the RILIS optimized for the ionization of astatine. (b) An un-gated (black) and gated (blue) laser scan of the 216 nm first step transition for astatine. The TOF gate applied is indicated by the blue color in spectrum (a). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

case the astatine ions are a significant component of the un-gated ion signal so the resonance profile of the 216 nm astatine transition is clearly seen in both the gated and un-gated spectra. Nevertheless, the benefit of the improved signal-to-background ratio for the gated spectrum is reflected by the smaller statistical uncertainties for the data points. The availability of the surface ion production rate at each laser wavelength step improves the scan quality by providing a correction factor that accounts for non-laser related production rate fluctuations. The gated spectrum of Fig. 8 includes these correction factors.

5. Summary and outlook

The recent developments of the in-source RIS setup at ISOLDE has enabled isotope shift and hyperfine structure measurements of particularly challenging cases. Because of the flexibility offered by the new dual-RILIS laser installation, for the first time it has been possible to study two different atomic transitions during one experiment. This is particularly important for elements such as astatine for which little spectroscopic information is known, or for reducing the reliance on atomic calculations during the extraction of charge radii [35].

The successful application of the LIST has shown that this new ion source option for ISOLDE offers the potential to extend the laser spectroscopy measurements to isotopes which would otherwise be overwhelmed by isobaric contamination. With the planned future optimization of the LIST performance, a further increase in the selectivity is expected. At least an order-of-magnitude improvement of beam purity will be required for the study of ^{211,212} Po, due to the high production rate and relatively poor LIST suppression of ^{212}Fr.

The Windmill alpha-decay spectroscopy station offers unrivaled detection efficiency, coupled with sufficient energy resolution to obtain clean spectra for the most exotic isotopes produced at ISOLDE. In combination with the inherent isomer selective laser ionization during a laser scan, the decay data obtained with the Windmill contains an abundance of additional nuclear spectroscopic information such as decay schemes, spin and parity assignments, etc [36–38].

The ISOLTRAP MR–ToF MS was used for photo-ion detection for the first time. This method is essential to obtain background-free laser spectra for many ion beams containing surface ionized isobars, particularly for α-inactive, or long-lived isotopes.

The results obtained for astatine and gold isotopes and the new progress in the study of shape coexistence in the lead region will be reported in several pending articles.

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