Nuclear Instruments and Methods in Physics Research B 376 (2016) 39–45

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Blurring the boundaries between ion sources: The application of the RILIS inside a FEBIAD type ion source at ISOLDE

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ARTICLE INFO

Article history:
Received 2 September 2015
Received in revised form 15 February 2016
Accepted 3 March 2016
Available online 11 April 2016

Keywords:
RILIS
VADLIS
Resonance laser ionization
Ion source
ISOLDE

ABSTRACT

For the first time, the laser resonance photo-ionization technique has been applied inside a FEBIAD-type ion source at an ISOL facility. This was achieved by combining the ISOLDE RILIS with the ISOLDE variant of the FEBIAD ion source (the VADIS) in a series of off-line and on-line tests at CERN. The immediate applications of these developments include the coupling of the RILIS with molten targets at ISOLDE and the introduction of two new modes of FEBIAD operation: an element selective RILIS mode and a RILIS + VADIS mode for increased efficiency compared to VADIS mode operation alone. This functionality has been demonstrated off-line for gallium and barium and on-line for mercury and cadmium. Following this work, the RILIS mode of operation was successfully applied on-line for the study of nuclear ground state and isomer properties of mercury isotopes by in-source resonance ionization laser spectroscopy. The results from the first studies of the new operational modes, of what has been termed the Versatile Arc Discharge and Laser Ion Source (VADLIS), are presented and possible directions for future developments are outlined.

1. Introduction

The Resonance Ionization Laser Ion Source (RILIS) has become the principal ion source of the ISOLDE radioactive ion beam facility [1]. At ISOLDE, resonance laser ionization is typically applied inside a hot cavity surface ion source: a metal tube 34 mm in length with an inner diameter of 3 mm, made from tungsten or tantalum with a standard operational temperature in the region of 2000 °C. The increasing ubiquity of the RILIS stems from its combination of element selective ionization with efficiencies in the region of 10%. There are two avenues by which to expand the RILIS capabilities. The first is laser ionization scheme development, enabling new or more efficiently produced laser ionized ion beams. The second avenue for development is the exploration of alternative laser/atom interaction and ion extraction conditions, of which the results reported here are an example.

The recent studies presented here demonstrate the possibility to blur the boundaries between ion sources at ISOLDE. The RILIS [2] has been combined with the Versatile Arc Discharge Ion Source (VADIS) [3], ISOLDE's variant of the Forced Electron Beam Induced Arc Discharge (FEBIAD) type ion source [4], enabling laser resonance ionization to be achieved inside the VADIS anode cavity. The combination, termed the Versatile Arc Discharge and Laser Ion Source (VADLIS), offers both immediate applications and promising directions for further development to meet the laser ion source requirements of the future.

The original motivation for coupling the RILIS and the VADIS was as a step towards the production of refractory metal ion beams at ISOLDE: extracting refractory elements as molecules [5], dissociation to atomic form using the VADIS, then selective ionization using the RILIS lasers. While this work continues, the initial

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http://dx.doi.org/10.1016/j.nimb.2016.03.005
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promising results obtained for resonance laser ionization of non-refractory elements has led to both immediate applications and additional opportunities. Here we introduce the VADLIS as a new ion source option for ISOLDE and provide a summary of the modes of operation that have been characterized thus far. The immediate applications and the progress towards future opportunities are also discussed.

2. The ISOLDE RILIS and VADIS

At ISOLDE, reaction products created inside the target effuse via a transfer line to an ion source for ionization. The principles of FEBIAD type ion sources, described in detail by Kirchner and Roeckl [4], are schematically outlined in Fig. 1. Electrons are emitted by a cathode, typically heated to over 2000 °C. The anode is a cylindrical electrode held at 100–150 V with respect to the cathode. The face of the anode cylinder directly opposite the cathode is a grid, allowing the passage of accelerated electrons into the anode cavity. A portion of the reaction products inside the anode volume are ionized in the arc discharge resulting from electrons accelerated from the cathode [4,6]. Ions are extracted from the anode cavity through a 1.5 mm aperture at the opposite end to the grid.

The anode geometries of all ISOLDE VADIS type ion sources are identical to that of the MK5 FEBIAD [7,8], though with molybdenum rather than graphite components. The material modification was implemented because of the apparent sensitivity of the FEBIAD to the outgassing of CO [3,7]. As such, the results presented here are expected to be equally applicable to FEBIAD variants used at radioactive beam facilities worldwide.

The ISOLDE RILIS, described by Rothe et al. in these proceedings [9], uses tunable lasers to target a progressive series of resonant atomic transitions, before a final ionizing transition, either to an autoionizing state or a non-resonant transition to the ionization continuum. The lasers are pulsed with a repetition rate of 10 kHz, which is well suited to the mean effusion time of 100 µs for atoms along the length of the hot cavity. The hot cavity walls confine the atoms, providing a complete overlap of the RILIS lasers with the reaction products. If the RILIS lasers are instead transmitted through the 1.5 mm VADIS exit aperture, the geometry of the VADIS anode cavity limits the laser-atom overlap region to just 7% of the anode volume. However, assuming saturation of the directed into the anode volume, the efficiency of the laser ionization within the cavity is independent of the area of the extraction aperture [2,10]. Neglecting wall sticking times, the mean atom residence time within this volume is expected to be of the order of 10 ms [4].

3. Experimental set-up

Here we describe the experimental set-up used for the first VADLIS tests, which were conducted at the ISOLDE off-line separator. This is representative of the set-up used for the subsequent tests performed at the ISOLDE GPS separator (off-line and on-line) [11]. Significant differences in the experimental set-ups are highlighted individually. The off-line facility is a replica of the ISOLDE on-line front-end, attached to a dipole mass separator magnet. For simplicity, the ionization of gallium in the VADIS anode cavity was investigated during all tests at the off-line separator. Gallium ionization in both the hot cavity (surface and laser ionization) [12] and the Laser Ion Source Trap (LIST) [13] has been well characterized before. The ionization potential of 6 eV ensures that gallium is surface ionized at a typical hot cavity operating temperature of 2000 °C, but with low efficiency (<1%), thereby providing a benchmark by which to judge the relative efficiencies of other ionization methods. The boiling point and the natural isotope abundance signature of gallium make it a reliable and easily identifiable ion beam for ion source development work. Furthermore, the two-step laser ionization scheme $\{\lambda_1 \mid \lambda_2\} = \{294\,\text{nm} \mid 532\,\text{nm}\}$ [1] requires only one tunable RILIS laser. The experimental set-up is summarized in Fig. 2.

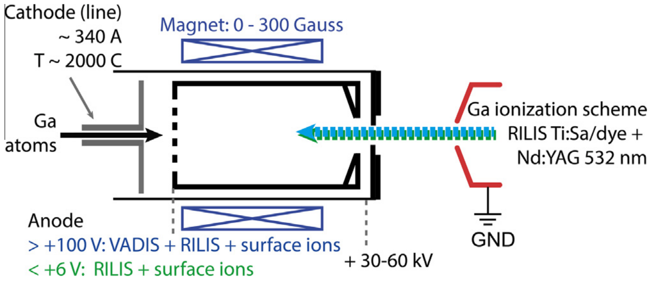

Fig. 1. A schematic of the VADLIS ion source.

Light from a titanium-doped sapphire (Ti:Sa) laser was frequency tripled to produce the first step of 294 nm. The non-resonant final step transition of 532 nm was provided by a frequency doubled Nd:YAG laser (Edgewave). The wavelength of the Ti:Sa laser was measured using a HighFinnesse/Angstrom WS6 wavelength meter. The laser beams were directed through a window in the dipole mass separator magnet, into the anode cavity of a VADIS via the exit aperture as depicted in Fig. 1. A sample of stable gallium was evaporated from a resistively heated refractory metal capillary (mass-marker) attached at the rear of the ion source. The laser parameters (beam position, average power, wavelength, timing) were maintained constant whilst the VADIS parameters (anode voltage, cathode heating, magnetic field strength) were varied to determine the optimal conditions for the extraction of laser ions. At the off-line separator depicted in Fig. 2, the target and ion source construction was held at +32 kV so that the grounded extraction electrode, located downstream of the anode aperture, accelerated the positive ions, creating a 32 keV ion beam.

A LeCroy Waverunner 104MXi oscilloscope was synchronized with the 10 kHz laser trigger and used to record a time structure histogram of the ion signal, measured with a microchannel plate (MCP) detector installed in the focal plane of the mass separator. The ion current incident on the MCP detector was limited to below 1 pA. For ion currents in excess of 1 pA, a Faraday cup was used as a more robust measurement device, but without the time resolution capabilities of the MCP detector.

4. Off-line testing and characterization

With the presence of the RILIS laser light in the VADIS anode cavity, gallium atoms may be ionized by three distinct processes: arc discharge ionization, surface ionization and resonance laser ionization. At a constant cathode temperature, adjusting the anode voltage influences both the total ion quantity and the relative contribution from each ionization process. Operation in the 1–5 V region offers a selective RILIS mode of operation, where only laser and surface ionization is observed. A sufficient positive voltage on the anode grid provides active suppression of surface ions ionized outside of the anode cavity, leaving only the internal anode walls, radiatively heated by the cathode, as a point of origin for surface ions extracted from the source. Arc discharge ionization can occur once the electrons emitted from the cathode are accelerated with a sufficient anode voltage. The effect of increasing the anode voltage on the arc discharge related ion current typically saturates between 100 V and 160 V, this is the standard VADIS mode of operation. The introduction of the RILIS lasers while in VADIS mode has been demonstrated to increase the overall ion current and is referred to hereafter as the RILIS + VADIS mode of operation.

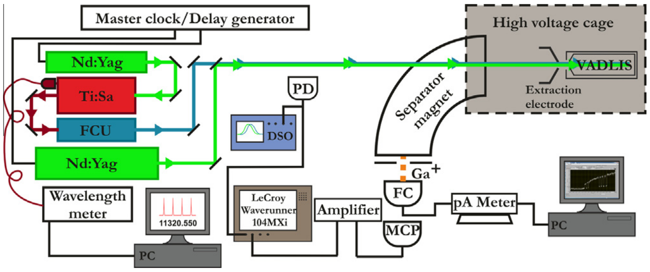

Fig. 2. Experimental set-up used at the ISOLDE off-line separator. DSO: digital storage oscilloscope, FC: Faraday cup, FCU: frequency conversion unit, MCP: microchannel plate, PD: photo diode.

The distinct modes of operation are observable in the measurement of the ^{71}Ga ion rate during a scan of the anode voltage with the cathode heated with 340 A (Fig. 3a). Other than the anode voltage, all VADIS parameters were maintained constant during the scan. At each anode voltage step, measurements were made with the 294 nm resonant first step transition both blocked and unblocked to enable a determination of the laser ionized fraction (red curve) and the VADIS ionized fraction (black curve) from the total ion current (dashed blue curve).

As depicted in Fig. 3a, the maximum ion current of ^{71}Ga was observed with the lasers directed into the ion source and an anode voltage of 2 V. With increasing anode voltage, resonance laser ionization was observed to dominate the ionization mechanisms until 60 V was reached, at which point arc discharge ionization became the dominant process for ionization. Both the RILIS related and arc discharge related ion currents dropped in the step from 23 V to 24 V, the RILIS related ion current from 176 pA to 100 pA and the arc discharge related ion current from 34 pA to 20 pA. Similarly, following the step from 55 V to 60 V, both the RILIS related and arc discharge related ion currents increased: the RILIS related ion current increased from 36 pA to 80 pA and the arc discharge related ion current increased from 55 pA to 330 pA. The coincidence of the decrease and increase in both currents, indicates a change in the extraction efficiency of the ions at these voltage steps, rather than a particular increase or decrease of the ionization efficiency of either types of ionization mechanism.

The cathode heating current was then increased to 360 A and the anode scan was repeated. The result, shown in Fig. 3b, demonstrates a partial loss of the RILIS mode of operation. Here, we observe the onset of an increase in the efficiency of laser ion extraction, coincident with an improved extraction or ionization rate of arc discharge ionized gallium between 10 V and 15 V. An increase in the maximum achievable VADIS mode ion current of 485 pA was also observed following the increase in the cathode heating current to 360 A, however, this ion current did not exceed the maximum recorded RILIS mode ion current of 530 pA achieved with a cathode heating current of 340 A.

To some extent, the features that appear in Fig. 3a can be interpreted by an analysis of the time structure of the ion beam, relative to the laser pulse arrival time. The time structure measurements required an MCP and were therefore performed with ion currents three orders of magnitude below those recorded in the anode scans presented in Fig. 3a and b. The time structures recorded at the maxima and minima in the laser ion signal (0 V, 2 V, 5 V and 15 V), observed in the anode voltage scan with the 340 A cathode heating, are presented in Fig. 4. The histograms have been normalized to 35,000 counts over the 100 μs time window.

Similarly to laser ionization in a hot cavity, the first peak in the time structures shown in Fig. 4 is understood to correspond to laser ions created at the exit aperture of the cavity, in a region where penetration of the extraction field is possible [14]. The second peak observed in the time structure measurements recorded at 5 V and 15 V and the slow release component, observed as a continuous background of ~17 counts, are features believed to correspond to the potential distribution along the axis of the anode cavity.

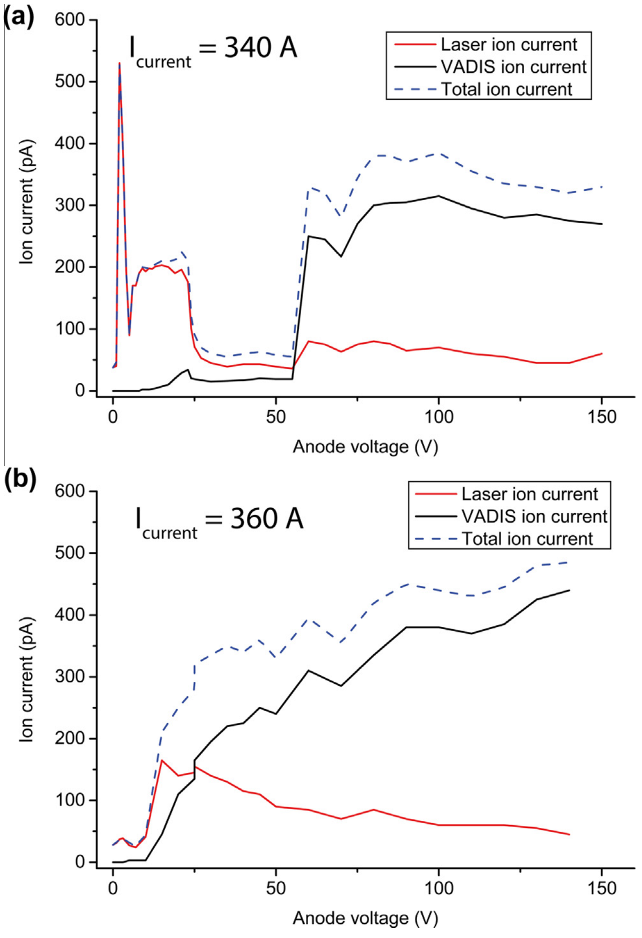

Fig. 3. Anode voltage vs ^{71}Ga ion current for an applied cathode heating current of 340 A (a) and 360 A (b). The laser ion current was determined by measuring the reduction in the total ion current following the blocking of the 294 nm laser beam.

Ion survival in a hot cavity is understood to be strongly dependent on electron emission from the inner walls creating a negative potential well along the central axis, with respect to the positively charged walls, that prevents the positive ions from colliding with the walls [2,15]. VADIS simulations indicate a similar, but completely enclosed, potential well at the center of the anode cavity when the VADIS is operated with 150 V applied to the anode [3]. It was concluded that only the ions created in the region outside of this central potential well could be extracted efficiently.

A qualitative understanding of the features observed in the time structures presented in Fig. 4, can be formulated by assuming the existence of such a potential well when lower voltages are applied to the anode cavity. Under conditions where laser ions can escape the potential well, the average laser ion survival is enhanced, though the extraction of these ions is retarded by the electric field distribution, resulting in a DC component of the laser ion current, observable as the ~17 count background in the time structures taken at 2 V and 15 V. In conditions where the potential well is too deep and laser ions are unable to escape, the total laser ion rate is reduced due to the poor extraction efficiency and the prompt release component is dominant. This can be observed in the time structures taken at 0 V and 5 V, anode voltages that correspond to minima in the ion current. The second peak, visible in the time

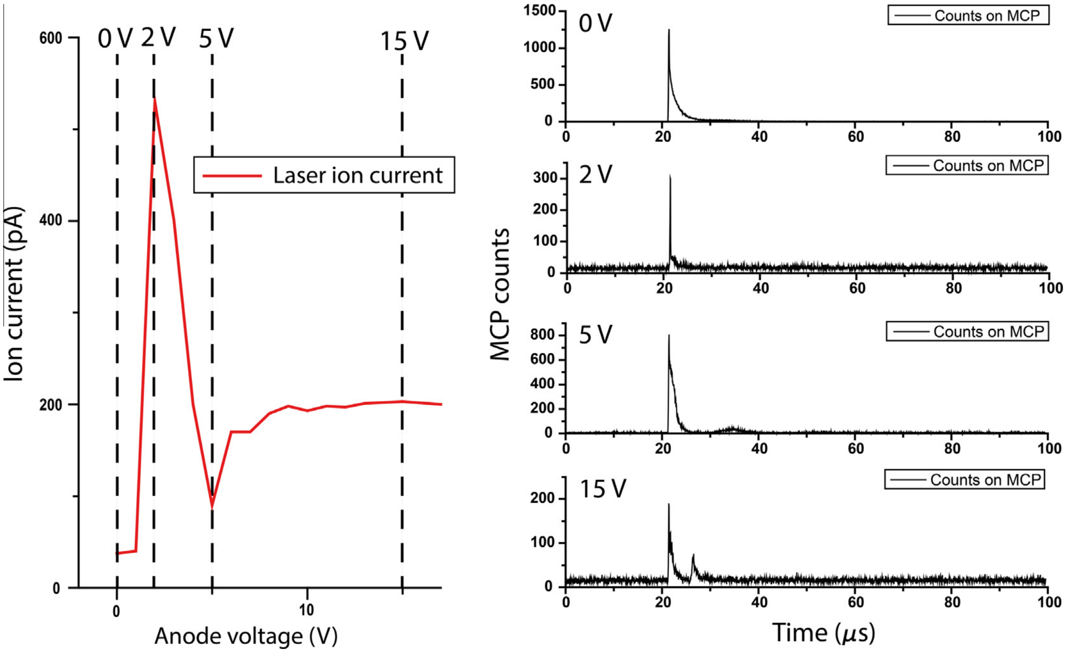

Fig. 4. Time structure measurements of the ^{71}Ga laser ionized beam, cathode 340 A. The distinct voltages at which the time structures were obtained are highlighted in the anode voltage scan (detailed view from Fig. 3a). Details are given in the text.

structures measured at 5 V and 15 V, is understood to correspond to laser ions created at the rear of the anode cavity, outside of the central potential well.

The differing ion current dependence on the anode voltage between Fig. 3a and b could therefore be understood as the result of a variation in the electron density within the anode cavity. The increased electron density in the anode cavity due to an increase in the cathode heating current to 360 A, resulted in a potential well too deep for the laser ions to escape until ions of other elements, produced after the onset of arc discharge ionization began to occupy the central potential well, thereby balancing the influence of the additional electrons.

To illustrate the dependence of RILIS mode efficiency on extraction from the assumed potential well at the center of the anode cavity, the laser ion signal is plotted with the percentage of the MCP counts relating to the slow release DC component in Fig. 5a and b, for operation at 340 A and 360 A respectively.

The time structure measurements corresponding to the dashed (blue) line in Fig. 5a and b were recorded with ion currents three orders of magnitude below that of the Faraday cup measurements, due to the limits of the MCP; however, between 0 V and 15 V there is still a clear correlation between the fraction of the total counts coming from the slow release DC component of the time structure and the ^{71}Ga ion rate measured on a Faraday cup. This indicates a requirement for the extraction of what are understood to be ions from the center of the cavity for the most efficient RILIS mode operation. In Fig. 5a, this can be seen in the minima at 0 V and after 5 V, and the maxima at 2 V and 10 V in both the ion current measured with a Faraday cup and the contribution of the DC background to the total count rate recorded in the time structure measurements. Similarly, in Fig. 5b it can be seen in the increase after 10 V is applied to the anode. The sensitivity of the laser ion extraction to the electron distribution within the anode cavity was confirmed by an observed influence of the source magnet on the extracted laser ion current by up to a factor of two or more. Electron trajectories are significantly affected by the weak magnetic field but any direct influence on ion dynamics is expected

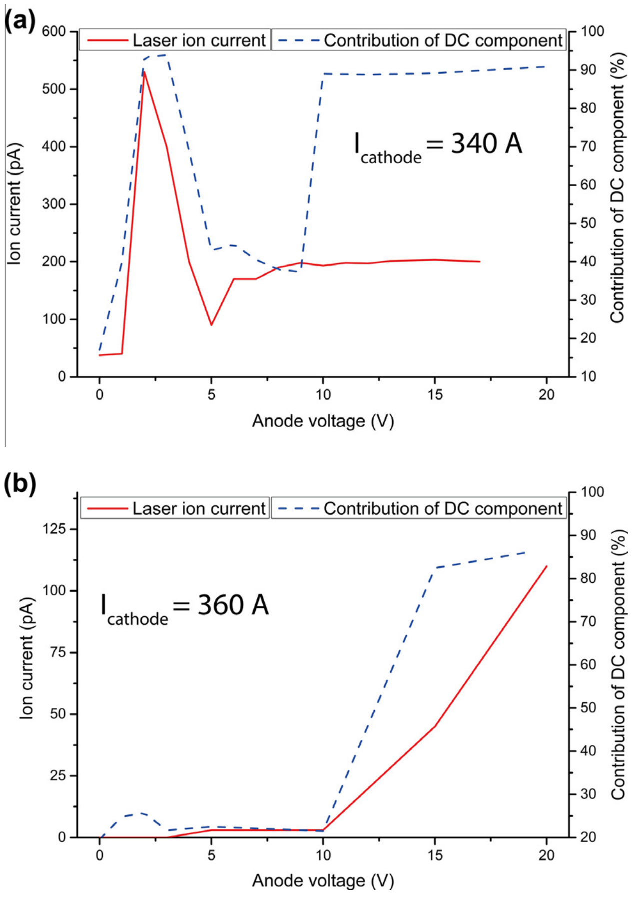

Fig. 5. Correspondence of the RILIS related ^{71}Ga ion current with the DC slow release component observed in time structure measurements, cathode heated with 340 A (a) and 360 A (b).

to be negligible. The understanding and optimization of the RILIS mode of operation will be the subject of further investigation.

In addition to the standard VADIS mode of operation, two new operating modes have therefore been established: the predominantly element selective RILIS mode, with the anode voltage set below the threshold for electron impact ionization to occur and a RILIS + VADIS mode, with the RILIS laser light whilst a nominal VADIS mode anode voltage is applied. The RILIS + VADIS mode was observed to result in the highest ion current during the on-line operation, as discussed in Section 6. Surface ionization can also contribute to the total ion production however, this is strongly dependent on the ionization potential of the element.

5. VADLIS for selective RILIS operation

The VADLIS modes of operation were investigated off-line at ISOLDE with barium. The RILIS lasers were tuned to ionize barium with a two-step $\{\lambda_1 \mid \lambda_2\} = \{350\,\text{nm} \mid 532\,\text{nm}\}$ scheme [16] and the laser beams were directed into the anode cavity of a VADIS mounted on the ISOLDE HRS front end [11]. The results are presented in Fig. 6.

At 2.5 V, corresponding to the maximum recorded ion current, the application of the RILIS lasers enhanced the barium related signal by a factor of 8. By comparison, off-line tests of barium laser ionization in a hot cavity using the same scheme resulted in a laser enhancement of the barium ion current of less than 1.5. The enhanced selectivity of the VADLIS is thus far understood to be the consequence of a reduced efficiency for surface ionization compared to that of a standard hot cavity surface ion source, combined with surface ion suppression by the anode grid. The signal reduction when the lasers were blocked, with 150 V applied to the anode, was below 10%, indicating that the majority of the ions extracted at this voltage were ionized by arc discharge ionization.

The dominance of a RILIS mode of operation for barium is apparent from the results depicted in Fig. 6. Reduced FEBIAD efficiencies for alkali metals, following the application of a VADIS mode anode voltage, was reported by Burkard et al. [17]. It would appear that this behavior is consistent for barium which also has a comparatively low ionization potential of 5.2 eV. The repelling action of the anode voltage was also observed during the same VADIS test for surface-ionized ^{133}Cs where the largest ion current was observed at 0 V. The mechanism behind these effects is under investigation.

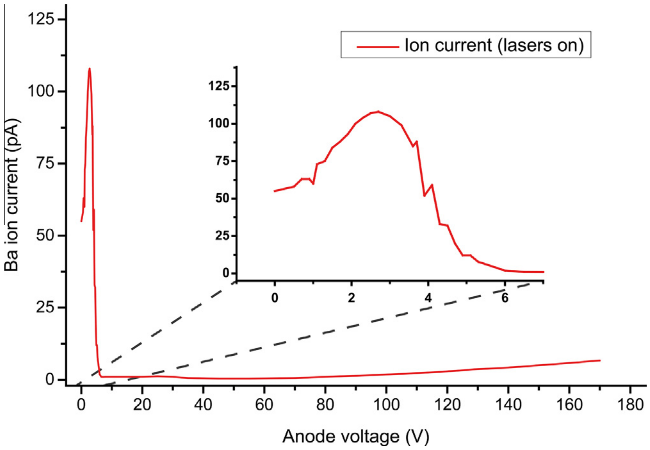

Fig. 6. ^{138}Ba ion current during an anode voltage scan with RILIS lasers directed into the anode cavity. The total signal was recorded without distinction between laser ionization and other ionization mechanisms.

The use of the VADLIS as a laser ion source, has the potential to both increase selectivity compared to hot cavity RILIS operation and expand the applicability of the laser resonance ionization technique to elements currently surface ionized at ISOLDE. We intend to investigate the performance of the VADLIS with an inverted-polarity transfer line cathode, combined with a low work function material for the VADLIS anode cavity such as thoriated or lanthanated tungsten.

6. On-line testing and verification

Following successful off-line testing, an immediate application of the VADLIS developments is the possibility to couple the RILIS with molten metal targets at ISOLDE. The RILIS mode of VADLIS operation was demonstrated on-line at ISOLDE on three occasions during 2015: yield checks of mercury and the study of mercury isotopes by in-source resonance ionization laser spectroscopy using a molten lead target [18] and a test of the VADLIS modes of operation with radiogenic ^{114}Cd from a molten tin target. Fig. 7 is a comparison of the VADLIS modes of operation, observed for the two different target units during mercury yield checks and cadmium ionization tests. The ^{178}Hg rate was measured by α-decay spectroscopy in the CRIS DSS 2.0 set-up [19], while the ^{114}Cd signal was measured using a Faraday cup located after the ISOLDE GPS dipole magnet.

The on-line tests described here took place in-between scheduled ISOLDE experiments. The VADIS parameters were therefore maintained at the optimized values for on-line VADIS mode operation. All RILIS mode operation has thus far been validated by comparison to VADIS mode ion currents, for which the absolute efficiency has not yet been determined. Fig. 7 demonstrates a comparable efficiency for RILIS mode and VADIS mode operation under on-line conditions. A mercury ionization efficiency of 60% was reported at ISOLDE with the MK6 FEBIAD variant [8]. The MK6 however, differs in anode design and component material compared to the VD3 VADIS variant, currently used to ionize mercury at ISOLDE. An efficiency measurement of the RILIS ionization scheme for mercury in a hot cavity gave a lower limit efficiency of 6%. Fig. 7 demonstrates the comparable on-line ionization efficiencies of RILIS mode and VADIS mode operation for mercury and cadmium, with the highest ion currents observed in the RILIS + VADIS mode. Fig. 7 also includes the VADIS + Blaze effect of directing the 40 W laser into the VADIS anode, the increased

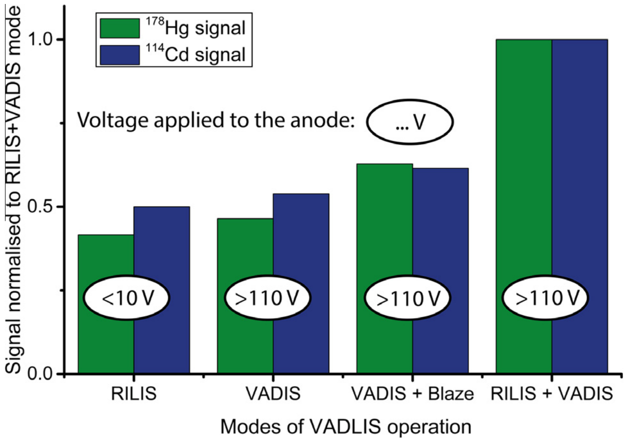

Fig. 7. Comparison of the VADLIS modes of operation. The Blaze is a 40 W frequency doubled Nd:YVO$_4$ laser, typically directed into a cavity for a non-resonant final step transition to the ionization continuum.

VADIS mode signal is thought to be due the incident laser beam increasing the temperature of the cathode surface.

7. On-line application and further development

The coupling of the RILIS with molten targets, combined with what is essentially a new type of laser ion source cavity, has already enabled the study of nuclear ground state and isomer properties of mercury isotopes between ^{177}Hg and ^{208}Hg. The possibility of switching between RILIS and VADIS modes of operation also has a wider significance for ISOLDE operation. Element selective and in some cases isomer selective ionization is now available from molten metal targets at ISOLDE, offering the possibility to significantly reduce isobaric and isomeric contamination in extracted ion beams.

The application of the RILIS requested for signal identification as part of a Coulomb excitation experiment using a VADIS ion source [20]. Using the VADLIS, combined with the new RILIS ionization scheme for tellurium [21], enables this to be accomplished using a single target/ion source assembly, in one continuous experimental run.

An enlarged anode aperture of 2.5 mm was trialed during the initial round of testing (not reported in this proceedings) in order to better match the focusing limitations of the RILIS lasers at a distance of ~20 m but at the expense of mass resolving power [4]. Following these successful initial tests, in order to maximize the flexibility of the ion source and enable immediate on-line application, the focus of the investigations moved towards optimizing modes of operation compatible with current VADIS anode geometries, the results of which have been reported here. It is therefore important to note that so far, only the optimal operating conditions for the existing VADIS design that have been investigated. There are numerous areas for further ion source development and testing:

- Selective ionization of refractory metal beams at ISOLDE: extraction from the target as a volatile molecule and dissociation in the VADIS before chemically selective resonance laser ionization.
- Selective RILIS: a low work function anode cavity combined with an inverted polarity cathode transfer line. The enhanced selectivity could potentially widen the range of RILIS applicable elements to those that are typically surface ionized at ISOLDE.
- Efficiency measurements: a series of RILIS mode and VADIS mode efficiency measurements are required to enable the absolute efficiencies of the hot cavity RILIS and VADLIS operation to be compared.
- Optimization of the anode cavity geometry and electrical configuration for RILIS and VADIS mode operation: the results reported here all took place using unmodified VADIS units, it is therefore unlikely that the existing design is optimal for RILIS mode operation. Simulations of the RILIS mode of operation would significantly aid understanding and streamline the design process. Resonance laser ionization could also be used to probe the electric field distribution inside the VADIS anode during VADIS mode operation, offering a minimally invasive method of study.
- Two-photon spectroscopy: it has been demonstrated that polished molybdenum is moderately reflective ($\geqslant 50\%$) across a broad spectral range, including at temperatures exceeding 1500 °C (private communication Maxim Seliverstov). The VADIS anode grid is a perforated molybdenum disc with 37 holes. Omitting the central hole, on-axis with the laser beam path, and polishing the molybdenum surface may enable “Doppler free” two-photon spectroscopy inside the anode cavity volume.

8. Conclusion

The ISOLDE RILIS has been successfully combined with the VADIS, ISOLDE’s variant of the FEBIAD type ion source. The combination, termed the VADLIS has been tested off-line with gallium and barium isotopes and on-line with mercury and cadmium isotopes. Three distinct modes of operation are now available: an element selective RILIS mode, the standard VADIS mode and a combined RILIS + VADIS mode for enhanced on-line efficiencies. The RILIS mode of operation has been investigated, the efficiency appears to depend strongly on the possibility of extracting laser ions from the full anode cavity. This is related to the cathode temperature, anode voltage and source magnet current. The selectivity of the RILIS mode has been demonstrated, potentially widening the range of RILIS applicable elements to those that are typically surface ionized at ISOLDE. The possibility to further suppress surface ions will be investigated. The results presented here mark the beginning of a series of further VADLIS developments and optimization.

Acknowledgments

This project has received funding through the European Union’s Seventh Framework Programme for Research and Technological Development under Grant Agreements: 262010 (ENSAR), 267194 (COFUND), and 289191 (LA^{3}NET). K.M.L. and L.P.G. acknowledges FWO-Vlaanderen (Belgium) as FWO Pegasus Marie Curie Fellows. S.S acknowledges a Ph.D. grant of the Agency for Innovation by Science and Technology (IWT).

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