Laser ion source tests at the HRIBF on stable Sn, Ge and Ni isotopes

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Abstract

As one step in the ion source development for the Rare Isotope Accelerator, a hot-cavity laser ion source using an all-solid-state titanium–sapphire laser system has been tested at the Holifield Radioactive Ion Beam Facility. Resonance ionization of stable isotopes of Sn, Ge and Ni has been studied in a Ta hot cavity. Efficient three step resonant ionization schemes applying frequency tripling for the first excitation step and using auto-ionizing or atomic Rydberg states in the ionizing step have been identified for all three elements, resulting in laser ion beams of typically around 100 nA. By saturating most of the optical excitation steps involved, ionization efficiencies of 22%, 3.3% and 2.7% have been measured for Sn, Ge and Ni, respectively.

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1. Introduction

For studies of nuclei far from stability, it is often necessary to have intense, mass-separated radioactive ion beams (RIBs) uncontaminated by isobars from neighboring elements. Resonant laser ionization has proven to be an universal method, applicable to nearly 80% of the elements in the periodic table, for selective ion production. Ions of a selected element are produced via stepwise atomic resonant excitations using one or two laser beams tuned to the transition energies specific to the element of interest, followed by either resonant or non-resonant ionization in the last transition. Because each element has its own unique atomic energy levels, the resonant photo-ionization process can, in principle, provide elemental selectivity of nearly 100%.

Dedicated laser ion sources (LIS) based on resonant photo-ionization were suggested and successfully tested more than 20 years ago [1–3]. Currently, they are in use at numerous isotope separator on-line (ISOL) facilities to provide efficient and selective production of isobarically pure radioactive ion beams [4–7]. For example, more than 20 radioactive ion beams can now be routinely provided at CERN-ISOLDE using copper vapor laser-pumped dye lasers with ionization efficiencies ranging from a few percent to near 30% [8].

Non-selective surface ionization is the main limitation on the purity of beams from existing hot-cavity LIS systems. Improvements in the ion source and laser technologies can further enhance the selectivity and efficiency of resonant laser ionization. Specifically, advanced laser ion sources will be needed to generate RIBs of high purity and intensity required by ground-breaking research on far-from-stability nuclei at the future Rare Isotope Accelerator (RIA) facility [9].
We have initiated a research effort to enhance the performance of present laser ion sources, with the ultimate objective of developing a prototype LIS capable of the required selectivity and efficiency for research with RIBs at RIA. Copper vapor laser (CVL) pumped dye laser systems have been used for most of the existing LISs. Meanwhile, tunable Titanium:Sapphire (Ti:Sa) lasers, which have emerged as an attractive all-solid-state alternative to CVL-dye laser combinations, have shown their feasibility for LIS in first demonstration experiments at an off-line test stand of the ISAC facility at TRIUMF [10,11]. Very recently this development has made it possible to selectively produce and investigate short-lived neutron-deficient Ga isotopes, in particular $^{62}$Ga, at ISAC [12]. In comparison, Nd:YAG pumped Ti:Sa lasers are in principle more rugged, more compact, and easier to operate and maintain than CVL-dye laser combinations, making them ideal for reliable and successful RIA applications. We therefore investigated tunable Ti:Sa lasers as the principal laser system for the prototype LIS. A hot-cavity LIS system with three Ti:Sa lasers has been set up and operated at the off-line Ion Source Test Facility (ISTF-2) of the Holifield Radioactive Ion Beam Facility (HRIBF) at Oak Ridge National Laboratory (ORNL), in collaboration with the University of Mainz, Germany. Initial laser ion source experiments, designed to demonstrate the use of Ti:Sa lasers in LIS applications, have been performed. Three-step resonant ionization on stable isotopes of Sn, Ge and Ni was achieved with Ti:Sa lasers and excellent ionization efficiencies of these elements have been measured on calibrated samples. Details of the experimental setup and results of these initial LIS experiments, along with our future plans, are presented herein.

2. Basics of resonance ionization and laser ion sources

Resonant laser ionization is effected through successive resonant photo-excitation of a valence electron from its ground state in the atom of interest to bound excited states followed by photo-ionization of the excited atoms. The ionization of an excited atom can be done via a non-resonant transition to the continuum or resonant excitation into an auto-ionizing state or to a Rydberg state with subsequent field ionization. For maximum ionization yield, simultaneous saturation of all the successive atomic transitions by the laser radiation is a prerequisite. The cross section for resonant transitions between discrete atomic levels is of the order of $10^{-10}$ to $10^{-14}$ cm$^2$ for the ideal situation of an atom at rest, while the ionization cross section for a non-resonant transition directly into the continuum is only of the order of $10^{-17}$ to $10^{-19}$ cm$^2$. Pulsed lasers can, at present, deliver the high photon flux needed for saturating all the participating atomic transitions, including the critical last ionization step. However, even at high repetition rate in the 10 kHz range, pulsed lasers have very small duty-factors ($\sim 10^{-4}$). The overall ionization efficiency is therefore normally intrinsically small due to poor temporal overlap between the pulsed laser beams and the continuously released atomic species. To overcome this limitation, the standard technique of the hot-cavity LIS at ISOL-facilities usually employs photo-ionization taking place inside an extended high temperature cavity with only a small exit aperture [13–15]. Radioactive reaction products are thermally transported out of the production targets and into the hot cavity. The laser beams are focused into the cavity for resonant excitation and ionization. The photo-ionized species then drift and are extracted out of the cavity by the penetration of the electric acceleration field of a subsequent extraction electrode. Neutral atoms can be confined in such cavities for considerably long time, allowing them to interact with multiple successive laser pulses. As a result, the ionization efficiency is greatly improved.

3. Description of laser ion source and laser system

3.1. Hot-cavity ion source

A hot-cavity LIS (Fig. 1) was designed and constructed at the HRIBF. The source has the basic structure of the standard high temperature RIB ion sources employed for on-line operation at the HRIBF. The ionizer cavity is a 30 mm length Ta tube having a 3 mm inner diameter and 1 mm wall thickness, resistively heated to near 2000°C. Target materials are vaporized in the heated target material reservoir. Atomic species of interest effuse through the transport tube (6 mm diameter and approximately 100 mm length) into the ionizer cavity where they are ionized by resonant absorption of photons. Elements with low ionization potentials could also be ionized by surface ionization on the hot tantalum surface. The ions produced are extracted from the cavity and accelerated to 20 keV energy. Laser beams are sent into the cavity from the extraction electrode side, in the direction opposite to the ion beams. A repeller electrode with a 6 mm diameter aperture is located at 1 mm from the exit face of the hot cavity. A positive bias voltage of up to 300 V can be applied to the repeller electrode to reject both laser-ionized and surface-ionized ions generated inside the tubular cavity.

3.1.1. Operating temperatures

In order to reduce the delay times associated with the effusion and adsorption processes, it is necessary to operate the source at high temperatures. The ionizer cavity and the transport tube are resistively heated by passing a dc electrical current through the tubular structure. The direction of the electrical current flow is such that the resistive heating also produces an axial electric field in the ionizer cavity which pushes the ions towards the extraction electrode. The target reservoir is heated radiatively by an independent cylindrical Ta heater that co-axially surrounds the reservoir. Although the target reservoir and the transport tube are mechanically attached, their temperatures can be separately controlled with limited coupling. Fig. 2(a) shows the temperatures of the ionizer cavity and the vapor transport.
the heating dc electrical current. As shown, the ionizer cavity can be heated to near 2000 °C. During the LIS experiments, the temperature of the target reservoir was monitored with an optical pyrometer looking through a view port on the vacuum enclosure of the ion source. Shown in Fig. 2(b) are the target temperatures as a function of the heating electrical current. The target reservoir could be heated to temperatures exceeding 1500 °C. During this experiment, up to 1300 °C was sufficient for sample vaporization.

3.1.2. Ion optics

The computer code PBGUNS [16] has been used to simulate the ion optics of the hot-cavity LIS. Fig. 3 is a PBGUNS model of the ion optics for extracting 1 µA positive Cs ions from the ionizer cavity, showing the ion trajectories and equipotential contours. The PBGUNS simulations assume a thermal plasma inside the ionizer cavity: ions are extracted from the plasma and accelerated to 20 keV by the extraction electrode. The plasma conditions are set by input parameters such as ion beam current which determines the plasma density of the extracted particles, temperature of the background electrons and initial drift energy of the ions. The plasma-beam interface is then determined automatically by PBGUNS. The values of the parameters used in the simulation studies were: Cs ion beam current 100 nA to 10 µA, initial ion energy 0.5–3 eV and electron temperature 0.25–2 eV. It is found that the location and shape of the plasma-beam boundary is most sensitive to the ion beam current and extraction electrode position, and relatively insensitive to the electron temperature and initial ion energy. Emittance values for this hot-cavity ion source, operating as a surface ionization source without the repeller electrode, have been measured experimentally to be ~10 r.m.m rad at the 90% contour level for 20 keV Cs+ ion beams [17].

3.1.3. Neutral residence time in the hot cavity

The dimensions of the Ta ionizer cavity, 3 mm inner diameter and 30 mm length, were chosen based on the
typical cavity geometries used for existing hot-cavity LISs, e.g. at CERN-ISOLDE. In this way comparison of data should be feasible. In general, tubular cavities having an inner diameter, \( d \), that matches the laser beam size, and a length \( l \gg d \) are expected to give high ionization efficiencies [15] because this geometry provides good spatial overlap and improves the temporal overlap between the neutrals and the laser beams. Neutrals entering such a cavity undergo many collisions with the wall of the cavity. As a result, their retention time residing in the cavity can be made long enough such that each neutral is irradiated at least once by the laser pulses before leaving the cavity. Consequently, the efficiency of photo-ionization is enhanced. The neutral residence time in the cavity has been estimated using a simple model [13,15,18]: the mean number of wall collisions, \( N \), that a neutral particle may undergo while passing through a cylindrical tube is equal to the ratio of the inner surface to the cross section area of the exit aperture. Thus, for a 3 mm inner diameter and 30 mm long tube, \( N \sim 40 \) and the corresponding mean residence time at 2000 °C would be approximately 100 μs for a particle of mass 30. Therefore, pulsed lasers with a repetition rate of 10 kHz or above are suited for particles of mass \( \geq 30 \).

For a more detailed and precise description, a Monte-Carlo code has been developed at HRIBF to simulate effusive processes of neutral particles through complex ISOL vapor-transport systems [19]. The code uses the particle detector simulation toolkit GEANT4 [20] to perform particle tracking in 3D geometries, and introduces thermal motion and particle–surface interaction for effusion processes. We have used the Monte-Carlo code to calculate the neutral residence time in the 3 mm diameter and 30 mm long ionizer cavity. Listed in Table 1 are the average number of wall collisions and the average distance traveled inside the tube, as calculated for different assumed angular distributions of particle’s re-emission from the surface after each collision: cosine distribution, isotropic distribution and specular reflection about the surface normal. Each result is averaged over 1000 individual particles. As noted, the Monte-Carlo calculation with specular reflection agrees well with the above mentioned simple model. However, it is expected that neutrals impinging on a solid surface are not specularly reflected, but leave the wall according to Knudsen’s cosine law. In the latter case, the number of wall collisions, and thus the corresponding average particle residence time, is more than 4 times larger.

### 3.2. Ti:Sa laser system

A tunable all-solid-state laser system consisting of three Ti:Sa lasers pumped by a high repetition rate Q-switched Nd:YAG laser has been developed at the University of Mainz, primarily dedicated for application in laser based ultra-trace analysis and atomic spectroscopy studies [21]. This laser system is also well suited for three-step resonance photo-ionization in a LIS. It provides wide tunability, reliable handling and good performance even in rough operation conditions and is compact for easy transport. The Ti:Sa lasers and all optical components required for beam monitoring, optimization and transport into the LIS are provided by the Mainz group for this study. The pump laser is a frequency-doubled Nd:YAG laser from Photonics Industries International, which operates at 10 kHz repetition rate with a maximum of 60 W average output power at 532 nm. With second and third harmonic generation capabilities, the Ti:Sa laser system can generate narrowband (~3 GHz) tunable laser radiation in the 720–925 nm (fundamental), 350–463 nm (frequency doubling) and 233–308 nm (frequency tripling) spectral regions. Each Ti:Sa laser is computer controlled for automatic spectral scanning and fine tuning of the fundamental wavelength. Temporal synchronization of the three lasers is better than 5 ns because each laser is actively Q-switched using internal...
3.3. Overall experimental setup

The laser ion source experiments documented here were performed at ISTF-2 of the HRIBF. The hot-cavity ion source was mounted on a high voltage platform located inside a fenced cage, with interlocks to the high voltage power supplies. For resonant ionization studies, solid sample materials, including tin metal foil, germanium metal shots and nickel metal wires, were placed in a graphite target reservoir, which was heated to desired temperatures for sample evaporation and release. Atoms of the species of interest effused from the target reservoir to the ionizer cavity where they were ionized by the laser radiation. The ions were extracted from the ionizer cavity with 20 keV energy and focused by an Einzel lens into a 90° dipole magnet for mass selection. Two Faraday cups with secondary electron suppression were used to measure the total beam intensity from the ion source before the analyzing magnet and the beam intensity after mass separation, respectively. The extraction electrode position was remotely adjustable with a computer via optical fiber linked RS232 communications.

The entire laser system and optical components were installed on a 4’ × 8’ optical table located after the mass analyzing magnet. Laser beams from the three Ti:Sa lasers were merged into one beam and sent into the vacuum through a 45° window on the analyzing magnet chamber. The laser beam was directed collinearly and in the opposite direction to the ion beam through the mass analyzing magnet and focused over a distance of about 4 m into the ionizer cavity.

4. Results and discussion

Resonant ionization of Sn, Ge and Ni was studied using the Ti:Sa laser based hot-cavity LIS. All three elements are among those radioactive ion beams of high importance for studies in nuclear structure and nuclear astrophysics. Sn was chosen as the primary benchmark species for testing the efficiency of the Ti:Sa laser-based LIS system because it had already been studied using copper vapor laser-pumped dye lasers as well as Ti:Sa lasers at ISOLDE. An efficient three-photon ionization scheme for Sn that leads to an auto-ionization state in the last step had been identified earlier [8] with dye lasers, and was recently demonstrated with Ti:Sa lasers [22]. To the best of our knowledge, no previous LIS studies have been reported for Ge. While Ni excitation schemes are known for the dye laser wavelength regime, the excitation ladders appropriate for Ti:Sa lasers had to be developed for this study.

As a preparatory investigation, the excitation schemes for Ni which are accessible for Ti:Sa lasers were examined at Mainz University. For all three elements, the first excitation step required ultraviolet (UV) radiation which was obtained by frequency tripling of the fundamental laser beam, while the second and third steps directly used the fundamental output of the Ti:Sa lasers. In addition, preliminary two-step resonant ionization of Ga was also conducted, but not finally concluded in this study. An overview of the parameters of the different excitation schemes for the three elements and the individual ionization efficiencies for three-step resonant ionization are compiled in Table 2. Note that for both Ge and Ni two alternative first excitation steps were explored with rather comparable experimental conditions.
4.1. Ionization of tin

The ionization scheme used for Sn is shown in Fig. 5(a). The Sn atoms were first excited from the \(5p^2 \, ^3P_0\) ground state to the \(5p^6s \, ^3P_1\) state (\(\lambda_1 = 286.42\) nm), and then promoted with an infrared photon (\(\lambda_2 = 811.62\) nm) to the \(5p^6p \, ^3P_2\) state. Ionization was carried out in the last step leading to the \(5p^9s \, ^3P_2\) auto-ionization state at \(59\, 375.9\) cm\(^{-1}\) by another infrared photon, \(\lambda_3 = 823.67\) nm. The mass spectrum displayed in Fig. 5(b) shows the Sn ions observed with the laser beams introduced into the ionizer cavity. Ion currents above 100 nA were easily obtained when the target reservoir was heated to about 1100°C. Also seen in the mass spectrum are Cs ions. When the laser beams were blocked, all Sn ions disappeared completely while the Cs ion current remained unchanged, as shown in Fig. 5(c). This indicates that all the Sn ions were produced by photo-ionization, while the Cs ions were surface ionized in the hot-cavity. If any one or two of the three laser beams were blocked, no Sn ion currents were detected, suggesting that the photo-ionization process was completely a three-color, three-photon process. It was not surprising that no surface ionized Sn ions were observed, as in this measurement the ion source was heated with a DC current of 340 A, which according to Fig. 2(a), meant that the Ta ionizer cavity temperature reached about 1700°C. Due to its high ionization potential of 7.344 eV the probability of surface ionization of Sn is negligible at

### Table 2

<table>
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<tr>
<th>Element</th>
<th>Initial state, energy</th>
<th>First excited state, energy</th>
<th>Second excited state, energy</th>
<th>Third excited state, energy</th>
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<tr>
<td>Sn</td>
<td>(5p^2 , ^3P_0, 0) cm(^{-1})</td>
<td>(5p^6s , ^3P_1, 34, 914.3) cm(^{-1})</td>
<td>(5p^6p , ^3P_2, 47, 235.2) cm(^{-1})</td>
<td>(5p^9s , ^3P_2, 59, 375.9) cm(^{-1})</td>
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<tr>
<td>Ge</td>
<td>(4p^2 , ^3P_0, 0) cm(^{-1})</td>
<td>(4p^5s , 1P_1, 40, 020.5) cm(^{-1})</td>
<td>(4p^5p , 1S_0, 51, 011.4) cm(^{-1})</td>
<td>Not assigned, 63, 828.3 cm(^{-1})</td>
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<tr>
<td>Ni</td>
<td>(3d^4s , ^3D_3, 204.8) cm(^{-1})</td>
<td>(3d^4s , ^1D_2, 879.8) cm(^{-1})</td>
<td>(3d^4d , J = 3, 49, 313.8) cm(^{-1})</td>
<td>Not assigned, 61, 479.7 cm(^{-1})</td>
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<table>
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<th>Ionization mechanism</th>
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<td>Field ionization</td>
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<tr>
<td>Auto-ionization</td>
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<th>First transition wavelength</th>
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<th>Ge</th>
<th>Ni</th>
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<tr>
<td>(286.42) nm</td>
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<td>(811.62) nm</td>
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<td>(823.67) nm</td>
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<th>Average efficiency</th>
<th>Sn</th>
<th>Ge</th>
<th>Ni</th>
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<tr>
<td>22%</td>
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<tr>
<td>3.3%</td>
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<tr>
<td>2.7%</td>
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this temperature. The Cs ions came from residual Cs left in the ion source from previous tests of the source in surface ionization mode using an external Cs oven.

The laser powers measured before the analyzing magnet were typically 10 mW of the UV beam for the first excitation step, and 1.5 W of the fundamental beams for the second and third excitations, respectively. As shown in Fig. 6, the laser powers available for this study are sufficient to saturate all the three excitation transitions. By fitting the measured data, the saturation laser power densities are estimated to be 40 mW/cm² and 3 W/cm² for the first and second transitions, respectively. It is not possible to extract the saturation power value for the last excitation step from the data due to the laser beam steering effects that occurred during laser attenuation. The saturation values are obtained assuming a homogeneous illumination on the whole ionizer tube area of ~10 mm², which agreed with visual inspection. No beam profiles were available for more precise determination of the laser beam size and spatial power density distribution.

4.2. Ionization of germanium

Resonant three-step ionization of Ge was successfully obtained for the first time. As shown in Fig. 7(a), Ge atoms were excited from the 4p ² ³P 1 (thermal population = 42% at 1700 °C) ground state level to the 4p5s ¹P 1 excited state (λ₁ = 253.39 nm), and then to the 4p5p ¹S 0 state (λ₂ = 909.85 nm). For the last transition, a search for auto-ionization or Rydberg states was conducted by scanning the third laser wavelength between 755 nm and 845 nm. A rich spectrum of Rydberg states and several auto-ionization states were successfully observed. They will be discussed in a later publication. The largest ionization yield was obtained using the auto-ionization state at 63 828.26 cm⁻¹. The mass scan data are also plotted in Fig. 7, showing Ge ions produced by laser ionization [Fig. 7(b)], and Ga ions formed by surface ionization [Fig. 7(c)]. No surface ionized Ge ions were observed. Ga existed in the ion source because the samples used in this study were Ge metal shots doped with 0.1–0.5% Ga. When
all three lasers were off, no Ge ions were observed. However, if either the second or third, or both, lasers were blocked, the Ge ion current did not drop completely to zero; a very small fraction (<0.001) of Ge ions could still be detected, suggesting that there is a small probability of ionizing Ge atoms by the ultraviolet photons alone, via a non-resonant, one-color, two-photon ionization process. Fig. 8 shows the measured Ge⁺ current versus laser power for each excitation step. The data indicate that the first two transitions are saturated at laser power densities of \( \frac{25}{100} \text{mW/cm}^2 \) and \( \frac{24}{100} \text{mW/cm}^2 \) for first and second transition, respectively. The third transition was not fully saturated with the available laser power of 10 W/cm².

4.3. Ionization of nickel

Based on the preparatory spectroscopic studies at Mainz University, two promising excitation schemes for Ni were discovered [Fig. 9(a)]. Assuming a pure Boltzmann distribution within the hot cavity at 1700 °C, the thermal populations of Ni in the hot cavity at 1700 °C are 40%, 27% and 12% for the ground state \( 3d^94s^2 \, \text{^3}F_4 \) and the two states, \( \text{^3}D_1 \) and \( \text{^3}D_2 \), of the low lying configuration \( 3d^94s \), respectively. Due to favorable transition wavelengths, two different first excitation steps, starting from either the \( 3d^94s \text{^3}D_3 \) (\( \lambda_1 = 274.75 \text{ nm} \)) or the \( 3d^94s \text{^3}D_2 \) (\( \lambda_1 = 279.94 \text{ nm} \)) level, and both leading to the \( 3d^94s^4p \text{^1}D_2 \) excited state, were used. As expected from the thermal populations, slightly higher Ni ion current was obtained with the \( 3d^94s \text{^3}D_3 \rightarrow 3d^94s^4p \text{^1}D_2 \) transition. In the second transition, the Ni atoms were excited to the \( 3d^{10}4d \text{^3}[\frac{7}{2}]_3 \) state of even parity. Auto-ionization states located between 61 678 cm⁻¹ and 62 815 cm⁻¹ are known, but they have even parity. Thus, no reasonable transition probability from the \( 3d^{10}4d \text{^3}[\frac{7}{2}]_3 \) state to those auto-ionization states is expected. An extensive search was conducted to identify unknown auto-ionization states of odd parity. Only one auto-ionization state of odd parity located at 62 217.2 cm⁻¹ was found with a weak resonance enhancement of about a factor of 3 over non-resonant ionization. A variety of Rydberg states with significantly higher ionization yields were, however, detected at 61 070–61 599 cm⁻¹.

Fig. 9. (a) Three-photon excitation scheme used for Ni. The last ionization step involved a Rydberg state. Also shown are the mass spectra observed with the lasers turned on (b) and off (c).
or 0.068–0.003 eV below the continuum; a detailed analysis of their structure will be given in a later publication. The excited Ni atoms in these Rydberg states were subsequently ionized, probably via field ionization within the acceleration field. The strongest Ni ion signals were obtained from the Rydberg states near 61 479.7 cm$^{-1}$. The laser power density available for this study was typically 100 mW/cm$^2$ in the UV for the first transition, and about 10 W/cm$^2$ for the second and third steps, respectively, which was not sufficient to reach saturation in the first and the last transitions. Shown in Fig. 9 are the mass spectra observed with the lasers turned on [Fig. 9(b)] and off [Fig. 9(c)] and off (Fig. 9(b)). Ga surface ions stemming from the sample material served as an ideal mass marker, while similar to Sn and Ge, no surface ionized Ni ions were observed.

4.4. Ionization efficiency

The ionization efficiency for all three elements Sn, Ge and Ni was measured using calibrated liquid samples, which contained a quantified amount of material in the range of $10^{16}$–$10^{17}$ atoms per sample. For efficiency measurements, the target reservoir was removed and the samples were directly placed in the sealed end of the Ta vapor transport tube. In this way, the samples could be heated to higher temperatures similar to on-line conditions for complete evaporation and depletion. While the source was heated, the laser wavelengths were tuned to the most efficient transitions investigated above and the laser ion current was continuously recorded until the sample atoms completely evaporated out of the source. The ionization efficiency was then calculated as the ratio of the integrated total number of detected ions to the total number of sample atoms contained in the initial sample. Most attention was given to Sn, which serves as an inter-comparison element to the efficiency data from the LIS at CERN-ISOLDE. Thus, three independent efficiency measurements were repeated for Sn. Although one of the measurements was stopped in the middle of the collection after 5 hours, the two complete measurements were fully reproducible. Fig. 10 shows the recorded Sn$^+$ currents for the two complete measurements, from which the overall ionization efficiency for Sn was extracted to be 21% and 23%, respectively. Agreement of these results suggests that the operation of the laser system and the hot-cavity ion source was stable and reliable.

In a similar procedure, the ionization efficiencies for Ge and Ni were measured to be 3.3% and 2.7%, respectively, which both might still be subject to further optimization. Especially for Ni, one of the reasons for the low ionization efficiency is the fact that the first and the third excitation steps are not saturated in this study. In addition, a somewhat different temporal release of the sample species is experienced. The Ni$^+$ current recorded during the efficiency measurement is plotted in Fig. 11. The data depict the release of Ni atoms as a function of time while the heating current for the Ta cavity was increased. As noted, the Ni$^+$ current reaches a sharp maximum when the ionizer cavity is heated to about 1700 °C, and then decreases monotonically – further increasing the temperature of the ionizer cavity and Ta vapor transport tube does not increase the Ni$^+$ current. Thus no sustained high ion currents as in the case of Sn are obtained. This phenomenon in Ni, which has been observed before [23], suggests that a limitation on the ionization efficiency for Ni could be due to reactions with the transport tube material (Ta). Atomic Ni may diffuse into tantalum at sufficiently high temperatures, potentially undergoing chemical, eutectic or alloy trapping. As a result, the overall atomization, and correspondingly, the ionization efficiency are significantly lowered. The ionization efficiency results are summarized in Table 2.

4.5. Repeller electrode

When a positive bias voltage of about 20 V was applied to the repeller electrode, both laser and surface generated ions were suppressed immediately by a factor of 100 and
more. In general, with a repeller bias voltage above 20 V, any ion signal almost completely vanished. This means that all the ions, laser ions and surface ions, were generated exclusively inside the hot tubular cavity. Apparently almost no atoms were ionized by laser light after the repeller, unlike observations in other repeller LIS investigations at off-line mass separator facilities [10]. To suppress surface-ionized ions only, the repeller bias voltage should be switched off when the laser pulse is on so that laser ions can be extracted from the cavity. Further optimization of cavity and repeller geometry and implementation of pulsed repeller bias voltage is under way.

5. Conclusion

A hot-cavity type laser ion source has been set up and initial laser ionization experiments using Ti:Sa lasers have been successfully performed at the ORNL-HR1BF, using the ISTF-2 oﬀ-line test facility. Three-photon resonant ionization of Sn, Ge and Ni has been successfully carried out with the Ti:Sa laser system from Mainz University. Operation of the hot-cavity ion source and the laser system were found to be very stable and reliable during the 2 week experimental period.

Overall ionization efficiencies of 22%, 3.3% and 2.7% have been achieved for Sn, Ni and Ge, respectively. Tin is chosen as the primary benchmark species for the efficiency and selectivity of the Ti:Sa based hot-cavity laser ion source. The overall ionization efficiency of ~22% for Sn obtained in this study is considerably higher than the 10% efficiency achieved using CVL pumped dye lasers and the 6.5% efficiency obtained with Ti:Sa lasers at ISOLDE [8,20]. This fact may be attributable to better laser-neutral overlap in this work. A longer transport distance between the target reservoir and the ionizer cavity may also have contributed to the observed higher efficiency, as a reasonable fraction of the photo-ions could be generated in the transfer line and pushed towards the exit by the axial electric field. No surface ionized background ions of Sn, Ge and Ni were present, but strong contributions of Cs and Ga were observed. Our results present the first resonant ionization LIS work reported for Ge, and the first efficiency measurement reported for Ni using Ti:Sa lasers. During these studies a variety of Rydberg and auto-ionization states have also been identified in Ni and Ge, which will be of value in future work. Analysis of the atomic spectroscopic data obtained is in progress and the results will be published separately.

As the ionization process was not fully saturated for either Ge or Ni in this work, improvements in ionization efficiency for both elements are expected, i.e. with a more powerful Ti:Sa laser system in the future. We are presently preparing research and development on more efficient excitation and ionization schemes for Ni and Ge, as well as on new elements of interest. In addition, systematic spectroscopic studies on auto-ionization and Rydberg states to be employed in the last ionization step will be conducted. Improvements in the mechanical design and material properties of the hot-cavity ion source are also important in order to improve the release and rapid transport of atomic species towards the laser ionization region while reducing surface-ionized ions. As a first step in this direction, the temporal distribution of the laser ions and the effect of a pulsed repelling potential will be investigated in further studies.

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