A resonant ionization laser ion source at ORNL

Y. Liu, D.W. Stracener

Physics Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA

Abstract

Multi-step resonance laser ionization has become an essential tool for the production of isobarically pure radioactive ion beams at the isotope separator on-line (ISOL) facilities around the world. A resonant ionization laser ion source (RILIS) has been developed for the former Holifield Radioactive Ion Beam Facility (HRIBF) of Oak Ridge National Laboratory. The RILIS employs a hot-cavity ion source and a laser system featuring three grating-tuned and individually pumped Ti:Sapphire lasers, especially designed for stable and simple operation. The RILIS has been installed at the second ISOL production platform of former HRIBF and has successfully provided beams of exotic neutron-rich Ga isotopes for beta decay studies. This paper reports the features, advantages, limitations, and on-line and off-line performance of the RILIS.

This manuscript has been authored by UT-Battelle, LLC, under Contract No. DE-AC05-00OR22725 with the U.S. Department of Energy. The United States Government retains and the publisher, by accepting the article for publication, acknowledges that the United States Government retains a non-exclusive, paid-up, irrevocable, world-wide license to publish or reproduce the published form of this manuscript, or allow others to do so, for United States Government purposes.

1. Introduction

The former Holifield Radioactive Ion Beam Facility (HRIBF) [1] at Oak Ridge National Laboratory (ORNL) was a DOE User Facility with a mission to deliver high-quality radioactive ion beams (RIBs) produced by the isotope separator on-line (ISOL) technique for nuclear research. At HRIBF, nuclei far from stability were produced by bombarding a thick target with a beam of light ions (proton, deuteron, or alpha) from the Oak Ridge Isochronous Cyclotron (ORIC)[2] driver accelerator. The radioactive atoms that diffused from the target material were ionized, accelerated, and mass selected by a high resolution magnet system. The desired RIB was then delivered to experiments or transported to the 25-MV tandem accelerator for further acceleration. In many cases, the isotopes of interest were produced at much lower yields than many neighboring isobars and the resolving power of the magnetic separators was insufficient to provide the desired suppression of the isobaric contamination. To address this problem, various beam purification techniques have been used or investigated at HRIBF to improve the elemental selectivity of the ion source or to add an isobar suppression step in the ISOL process [3–6].

Although HRIBF has not operated as a National User Facility since April 2012, some of its infrastructure and expertise has been maintained and could be used to benefit other RIB facilities in the future, including the resonant ionization laser ion source (RILIS) developed [7–10] in collaboration with the Laser Resonance Ionization Spectroscopy for Selective trace Analysis (LARISSA) group at the University of Mainz. In the RILIS, atomic species are selectively ionized by laser radiation via stepwise atomic resonant excitations followed by ionization in the final transition. This ionization process is highly selective for the targeted atoms. The RILIS is particularly useful for purifying RIBs that have large isobaric contamination but no effective ion selection method. The second ISOL production platform at the former HRIBF, Injector for

Radioactive Ion Species 2 (IRIS2) [1,11], was designed to enable the use and development of various beam purification techniques such as RILIS, ion beam cooling [3], and selective photodetachment [5]. The RILIS is based on a hot-cavity ion source and all-solid-state Ti:Sapphire lasers. In off-line studies with stable isotopes, it has been used to develop resonant ionization schemes for 14 elements and to measure ionization efficiencies for 8 of these elements with values ranging from 1% to 40% [10]. It is installed at IRIS2 and was commissioned in 2012 to deliver pure beams of short-lived, neutron-rich Ga isotopes for beta decay studies [12]. In this paper we report the features, advantages, limitations, and on-line performance of the RILIS as well as the recent off-line results of efficient ionization of uranium and thorium.

2. Description of the RILIS

The RILIS consists of a hot-cavity ion source and three all-solid-state, pulsed Ti:Sapphire lasers to provide up to three-step, three-photon ionizations. The hot-cavity ion source is mounted on the target and ion source (TIS) platform of the IRIS2, while the Ti:Sapphire lasers are installed in a dedicated laser room adjacent to the IRIS2 target and ion source room. The laser beams are collimated and combined in the laser room, transported into the IRIS2 TIS room, and then focused into the hot-cavity ion source through the viewport of the mass analysis magnet, as illustrated in Fig. 1. The total laser beam path is about 18 meters.

Fig. 2 shows the layout of IRIS2 and the location of the laser room and laser beam path. The TIS assembly is installed on a 60- kV platform. Ions accelerated from the TIS were mass selected by the first-stage isotope-separator magnet system with a nominal resolution of $\Delta m/m = 1/1000$ and then transported to the isobar separator magnets. The resulting isobar-separated RIBs were either

post-accelerated by the 25-MV tandem or delivered to the Low-energy Radioactive Ion Beam Spectroscopy Station (LeRIBSS).

2.1. Hot-cavity ion source

A schematic view of the hot-cavity ion source assembly is shown in Fig. 3. The cavity is a 30-mm long Ta tube of 3-mm inner diameter and 1-mm thick wall, which is connected to a target reservoir via a Ta transfer tube of 8.5-mm inner diameter. The Ta cavity and the transfer tube are resistively heated by passing an electrical current through them in series and can be heated to temperatures exceeding 2000 °C. The target reservoir can be independently heated to temperatures exceeding 2000 °C by a separate Ta heater surrounding the reservoir. Atomic species diffusing from the target material and effusing through the transfer tube into the hot cavity are selectively ionized by laser beams entering the cavity through the extraction electrode. The ions are subsequently extracted from the cavity and transported to the mass separators. A more detailed description of the hot-cavity ion source can be found in Ref. [13].

It is noted that the Ta transfer tube between the cavity and the target reservoir is typically on the order of 100 mm in length. A longer transfer tube could provide a longer laser–atom interaction region and thus could potentially result in higher photoionization probabilities for the atoms. On the other hand, it also means longer effusion lengths and larger adsorption surfaces, which is not desired for short-lived or chemically-reactive isotopes.

2.2. Ti:Sapphire laser system

The laser system includes three pulsed Ti:Sapphire (Ti:Sa) lasers operating at 10 kHz pulse repetition rate, all manufactured by Photonics Industries International, Inc. in Ronkonkoma, New

York. The Ti:Sa lasers are typically tunable between 720 nm and 960 nm, with about 2.5W peak power near 810 nm. Shorter wavelengths in the blue and ultraviolet regions are obtained by frequency doubling, tripling, and quadrupling of the fundamental laser outputs. Two of the three Ti:Sa lasers are combined with units for second-harmonic generation (SHG), third-harmonic generation (THG), and fourth-harmonic generation (FHG). In combination, the lasers can provide two frequency-doubled SHG outputs with 800 mW peak power, one frequency-tripled THG output with 120 mW peak power, and one frequency-quadrupled FHG output with peak power up to 30 mW at 215 nm. A more detailed description of the laser system can be found in Ref. [10].

The Ti:Sa laser system has some unique features. First, each Ti:Sapphire laser is individually pumped by a Q-switched Nd: YAG laser at 532 nm with an average output power of 19 W at 10 kHz pulse repetition rate. The Ti:Sa output pulses are typically 25–30 ns wide. The advantage of using individual pump lasers is that the three Ti:Sapphire laser outputs can be easily synchronized in time by synchronizing the pump lasers with external triggers. Thus, intra-cavity Pockels cells are no longer needed in the Ti:Sapphire lasers for synchronization purpose. Consequently, higher Ti:Sapphire laser output power can be obtained. The disadvantage is that multiple pump lasers are more expensive and less compact than a single pump laser of similar total power.

The second feature is that all three Ti:Sapphire lasers use a diffraction grating for wavelength selection so that they are continuously tunable over the available spectral range. Moreover, only one mirror set is used for the entire tuning range. Hence, wavelength changing and scanning across the full tuning range is easy and simple with these lasers. A major limitation of the grating-tuned Ti:Sa lasers is their output power: the maximum output power is about 2.5 W,

limited by the thermal effect and damage threshold of the gratings. Another limitation is on the tuning ranges of the Ti:Sa lasers, which are between 720 and 960 nm. We continue to improve the spatial and spectral qualities of the Ti:Sa lasers. The Ti:Sa lasers often have more than one transverse modes, especially under high pump power conditions. As a result, small wavelength jumps or mode hopping, on the order of up to one wavenumber in cm^{-1}, have been observed. In order to eliminate high-order modes and mode hopping, the resonator cavities of two Ti:Sa lasers have recently been modified with an intracavity telescope placed between the Ti:Sa crystal and the grating to better match the beam size of the fundamental mode with the pump beam size in the crystal. The outcome is that the two laser output beams are improved with good spatial uniformity and near TEM_{00} mode under high pump power conditions. The peak laser output power is slightly lower, about 10% less than that before the modification, but no mode-hopping has been observed in subsequent experiments. To obtain true TEM_{00} mode, larger modifications of the laser cavity may be required, which cannot be implemented due to limited space. It is also found that the modifications improved the tuning range of these two lasers to 700–960 nm. However, wavelengths below 700 nm are still not accessible.

Finally, the Ti:Sa lasers are compact. Each Ti:Sa laser and its pump laser are mounted together as one unit. The Ti:Sa laser has a dimension of 22” × 7.5” × 3.75” (Length × Width × Height). The harmonic generation stages are located immediately after the Ti:Sa cavity, in the same enclosure. The pump laser has a similar dimension of 22”(L) × 7”(W) × 3.75”(H). However, the disadvantage is that the lasers do not have space for large adjustments. Any improvements that require significant modifications, such as changing the rotation stage and stepping motor system for finer laser wavelength scan and redesigning the laser cavity to obtain true TEM_{00} transverse mode, cannot be easily implemented.

3. RILIS performance

3.1. On-line operation

The hot-cavity RILIS was commissioned in 2012 to provide beams of neutron-rich Ga isotopes for beta decay studies at LeRIBSS [14]. The radioactive Ga isotopes were produced via fission in a uranium carbide (UC_x) target bombarded by 50-MeV proton beams from ORIC. The Ga isotopes diffused out of the target material, effused along the transfer tube to the cavity region, and were selectively ionized by the Ti:Sa lasers in a two-step ionization scheme. The off-line ionization efficiency for stable Ga isotopes was about 9%. In combination with the high-resolution isobar separator magnets, beams of isotopically pure ^{78,83,85,86}Ga isotopes were delivered to LeRIBSS. The beam of particular interest was the exotic ^{86}Ga. The A = 86 beam was overwhelmingly dominated by ^{86}Br when an electron beam plasma ion source was used. With the RILIS, the isobar contamination observed for ^{86}Ga was surface-ionized ^{86}Rb, which, due to the relatively large mass difference, was easily removed by the isobar separator magnets. The excellent purity of the ^{86}Ga beam from the RILIS enabled the first measurement of the beta decay properties of ^{86}Ga [12]. With up to 15 μA of 50 MeV proton beam bombarding the UC_x target, the best RIB rates delivered to LeRIBSS were approximately 12,000 ions/s, 100 ions/s, and 0.3 ions/s for ^{83}Ga [$T_{1/2}$ = 308.1(10) ms], ^{85}Ga [$T_{1/2}$ = 93(7) ms], and ^{86}Ga [$T_{1/2}$ = 43(+21, −15) ms] [12], respectively.

For the on-line operation, the laser system was continuously operated for 17 days, including the time for laser alignment and initial tests with stable Sn and Ga isotopes. The stability of the lasers was monitored during the operation. Although only two lasers were needed for the two-step ionization scheme, three Ti:Sa lasers were used: one laser with THG output at 287.508 nm

for the first excitation step and two lasers with the fundamental outputs at 841.782 nm to provide a total of ~1.6 W into the ion source for the final ionization step. The two fundamental laser beams were very stable throughout the entire operation – adjustment in their power, wavelength, and timing was rarely needed. The THG output was very stable during the first week, but later slowly decreased in power and delayed in pulse time. This was found to be caused by a slow decrease in the pump laser power. The decrease in THG power did not significantly affect the yields of Ga ions since the saturation power of the first excitation step was on the order of 1.6 mW in the hot-cavity [14]. As the pump power dropped, the timing of the THG laser pulses needed to be adjusted about every 4–6 h during the last 4–5 days, but the laser wavelength was relatively stable, as shown in Fig. 4. After the online operation, a damaged laser mirror was found in that pump laser and repaired. The spatial alignment of all three laser beams to the hot-cavity ion source on the high-voltage platform of IRIS2 was very stable – no adjustment was needed during the run for Ga RIBs. There was no air conditioning and no temperature control in the laser room. The temperature in the laser room fluctuated about 1–3 degrees between day and night. No correlation between any changes in the laser wavelengths and the laser room temperature was observed.

3.2. Off-line operation

Currently, the RILIS is being used to study efficient ionization schemes for uranium and thorium. This work is motivated by the demand for more efficient ion sources for ultra-trace analysis of unwanted impurities in the materials used for the construction of the detectors for ultra-low background experiments, such as the Majorana Demonstrator (MJD) experiment [15] designed to search for neutrinoless double-beta-decay of the isotope ^{76}Ge. The MJD is a 40-kg detector array made from enriched ^{76}Ge and natural germanium. A specific goal of MJD is to

achieve the ultralow backgrounds, about one count per ton-year in the region of interest, required for the ton-scale experiment. Monte Carlo simulations indicate that uranium (^{238}U) and thorium (^{232}Th) impurities in the copper materials used for the detector assemblies are expected to be the dominant sources of background and must be at or below concentrations of a few 10^{-14} by weight. To detect such ultralow level impurities using the RILIS, highly sensitive techniques that are capable of direct counting atoms are considered, such as accelerator mass spectrometry (AMS) and resonance ionization mass spectrometry (RIMS). For conventional AMS, Cssputter negative ion sources are typically used and their ionization efficiencies for U are reported to be 0.06–0.1% [16,17]. The RILIS is much more efficient than Cs-sputter sources and, in addition, highly selective. It could be an alternative ion source for AMS, in combination with a charge-exchange-cell or directly used for positive ion AMS. Although the feasibilities remain to be demonstrated, for both AMS and RIMS, the efficiency of the ion source is essential for high sensitivity.

Extensive spectroscopy studies on resonant ionization schemes for actinide elements, including U and Th, using Ti:Sapphire [18–20] and diode [21] lasers have been carried out by the LARISSA group at the University of Mainz. Their studies revealed numerous atomic energy levels in U and Th that were previously not known and identified various excitation and ionization schemes optimized for Ti:Sapphire or diode lasers. Based on their prior work, we selected several three-step, three-photon ionization schemes and then conducted further spectroscopic studies to exploit new schemes and search for the most efficient scheme for U and Th. Detailed spectroscopic results will be reported in separate publications. Having all grating-tuned Ti:Sa lasers is of great advantage for spectroscopy and ionization scheme development. We can scan the fundamental wavelength of the fundamental Ti:Sa laser radiation used for the

second and third transitions over a wide range and quickly compare different schemes when the neutral atom density in the ion source is relatively stable.

The efficiency of the selected schemes has been measured with calibrated samples containing 5.06×10^{16} U atoms or 5.19×10^{16} Th atoms. The samples were made from 1000 ppm U or Th atomic absorption (AA) standard solutions (from Inorganic Ventures), which contained U and Th in a nitric acid matrix in the form of uranyl nitrate and thorium nitrate, respectively. To make a sample, 20 μL of the selected AA solution was dried on a thin Ti or Zr foil (0.0005 or 0.001 inch in thickness and about 5 × 6 mm in size) and then wrapped in the foil. For efficiency measurements, the target reservoir (Fig. 3) was removed and the 100 mm transfer tube served as the sample reservoir. The sample was placed near the end of the transfer tube. The detailed procedure of the efficiency measurement has been reported in Ref. [7].

For Th, a scheme shown in Fig. 5a was developed by Raeder et al. [20] at Mainz University with an efficiency of 0.6% reported. We conducted two efficiency measurements with our RILIS using the same scheme and obtained an overall efficiency of about 14% and 32% (Fig. 5b), respectively. The first measurement gave a much lower efficiency. This may be attributed to a brand new ion source which was not cleaned or passivated under high temperatures and could have much more reactions with and adsorption of the sample material. After the first use, the overall efficiency became higher and reproducible. Such behavior of the ion source has been observed repeatedly. A new ionization scheme was found to be about 15% more efficient. One measurement with the new scheme was then conducted using the same source. An overall efficiency of about 40% was obtained, in good agreement with the expectation. The detailed studies on Th ionization schemes will be reported in a separate publication. For U, overall ionization efficiencies of 9% have been measured. The preliminary work has been reported in

Ref. [22]. Again, our result is significantly higher than that of 0.04% reported by the LARISSA group at Mainz [19].Work is in progress to further improve the ionization efficiency for U.

The overall ionization efficiency depends not only on the ionization scheme but also on many other factors such as laser-neutral overlapping, ion transportation and detection, and sample release, etc. The difference between the overall efficiencies of our RILIS and the LARISSA setup, for the same ionization scheme, could be caused by the different ion source materials and geometries and the different ion optics. For example, the 0.6% Th efficiency of the LARISSA setup was reported to be limited by the transmission of the spectrometer ion optics, which was estimated to be on the order of 10% [20]. In our measurements, nearly 100% of the U and Th ions extracted from the hot-cavity were transported to the detector and detected. Moreover, the long transfer tube in our RILIS has also been attributed to the considerably higher ionization efficiencies for Sn than other RILISs using the same ionization scheme [7]. The overall high efficiency of our RILIS may be attributed to the long transfer tube, thus long laser–atom interaction region, in our hot-cavity ion source and the high efficiencies of beam extraction and transportation of IRIS2.

4. Summary

A RILIS has been developed at ORNL for the generation of pure beams of short-lived nuclei for experimental nuclear research. It consists of a hot-cavity ion source and three pulsed Ti:Sa lasers with 2nd, 3rd, and 4th harmonic generation capability. The unique features of the laser system include individual pumping of each Ti:Sa laser and grating-tuning of all three Ti:Sa lasers. The RILIS was successfully used to produce pure beams of neutron-rich Ga isotopes and enabled the beta decay properties of the exotic 86Ga to be studied for the first time. Recently, the RILIS has

been used to study efficient ionization schemes for U and Th. Overall ionization efficiencies of up to 40% for Th and 9% for U have been obtained.

Acknowledgments

This material is based upon work supported by the U.S. Department of Energy, Office of Science, Office of Nuclear Physics. This research used resources of the Holifield Radioactive Ion Beam Facility of Oak Ridge National Laboratory, which was a DOE Office of Science User Facility.

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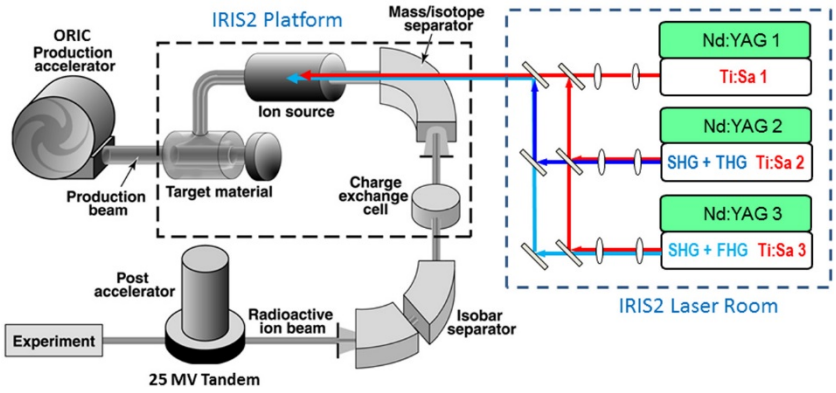

Figure 1. RILIS implementation at HRIBF.

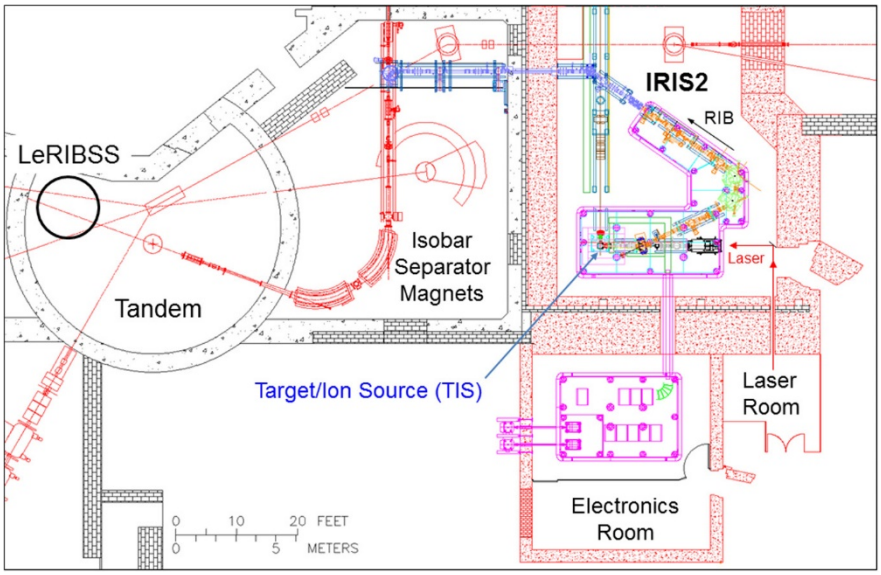

Figure 2. Layout of the IRIS2, laser room, tandem accelerator, and LeRIBSS areas.

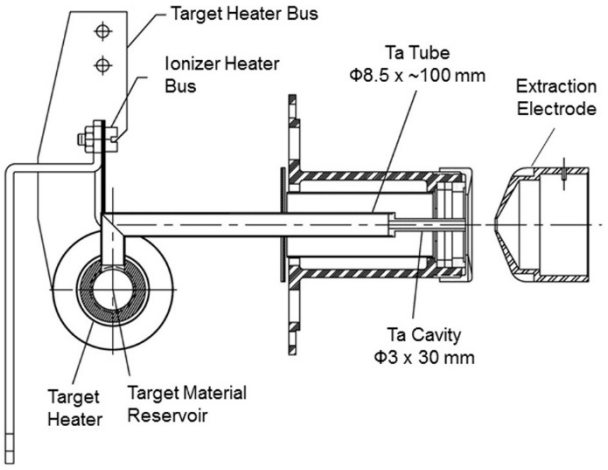

Figure 3. Schematic drawing of the hot-cavity ion source and target reservoir assembly.

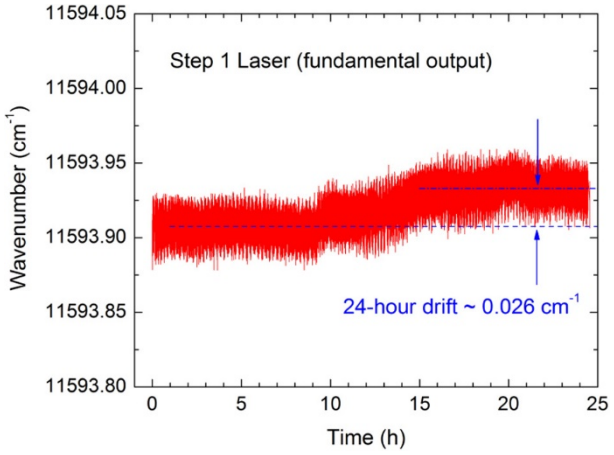

Figure 4. Stability of the laser wavelength for the first excitation step of Ga, monitored during the second week of the on-line operation. The 24-h drift of 0.026 cm^{-1} in the fundamental wavelength corresponded to about 0.078 cm^{-1} or 2.34 GHz shift in the THG output, which was much smaller than the linewidth of the resonance.

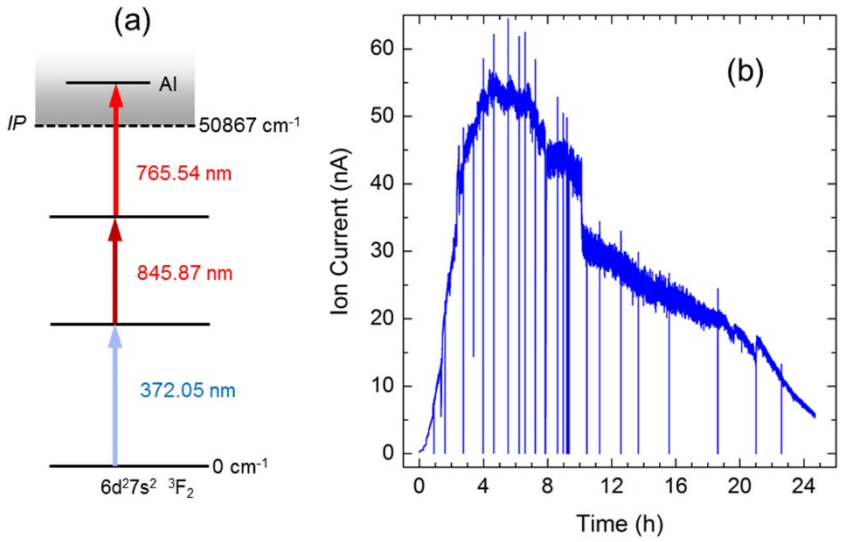

Figure 5. (a) Three-step ionization scheme for Th from Raeder et al. [20]. Th atoms are excited to an autoionizing (AI) state in the final step. (b) Th ion currents measured at ORNL with this scheme and a calibrated sample of 5.19×10^{16} Th atoms. The data yielded an overall ionization efficiency of about 32%.