

## ARTICLE

### Development of two-color resonance ionization scheme for Th using an automated wide-range tunable Ti:sapphire laser system

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Two-color resonance ionization schemes of Th were investigated by an automated wide-range tunable, grating-assisted Ti:Sa laser system with intracavity SHG option. A two-color ionization scheme via autoionizing state (1<sup>st</sup> step: 372.049 nm and 2<sup>nd</sup> step: 401.031 nm) was developed and its relative efficiency was lower by factor of three compared to a known three color scheme.

**Keywords:** resonance ionization; Ti:sapphire laser; thorium, isotope analysis

#### 1. Introduction

Resonance ionization of atoms by laser light is a well-established efficient and selective technique to produce ions of the element of interest. For ultra-trace analysis of pure alpha/beta emitting and/or long-lived radioactive isotopes of actinide and few other elements, resonance ionization mass spectrometry has been specifically developed up to routine application [1]. In addition, for the investigation of atomic and nuclear properties of exotic nuclei including heavy elements, resonance ionization laser ion sources are widely applied at several on-line radioactive beam facilities worldwide [2-6]. Pulsed high-repetition rate tunable Ti:Sapphire (Ti:Sa) lasers are particularly well suited for resonance ionization due to their reliable and maintenance-free long term operation. Conventional Ti:Sa lasers have an output power of up to several Watts in the fundamental mode at 10 kHz operation and offer a wide spectral tuning range via exchange of laser cavity mirrors [7]. However, this laser design does not allow for “mode-hop free” wide range tuning, which is required for development of highly efficient ionization schemes and to rapidly switch between different ionization schemes or even between different elements. Therefore, a dedicated automated wide-range tunable, grating-assisted Ti:Sa laser system was developed. Its applicability was demonstrated by development of ionization schemes for the actinide element, Th.

#### 2. Automated wide-range tunable Ti:sapphire laser system

A grating-assisted Ti:Sa laser has been developed and used for resonance ionization spectroscopy of Rydberg and autoionizing (A.I.) states [8,9]. We extended automated wavelength tuning of the laser into its second harmonic generation (SHG). **Figure 1** shows a setup of the grating Ti:Sa laser system with intracavity SHG option. To keep the critical phase matching condition in the nonlinear crystal (BBO) during wavelength tuning, its angle is automatically tracked by a motorized mount. **Figure 2** shows tuning ranges of the grating Ti:Sa laser; red and blue lines indicate the output power in the fundamental mode and in the second harmonic, respectively. Mode-hop free tuning was successfully demonstrated in the fundamental mode as well as in the second harmonic spectral range, controlled via a graphical user interface. Detailed specifications and computer-controlled operation of the laser system will be described in a separate paper planned to be submitted.

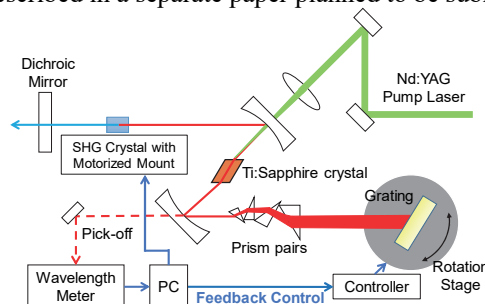


Figure 1. Automated, mode-hop free wide-range tunable, grating Ti:Sa laser system with an intracavity SHG option.

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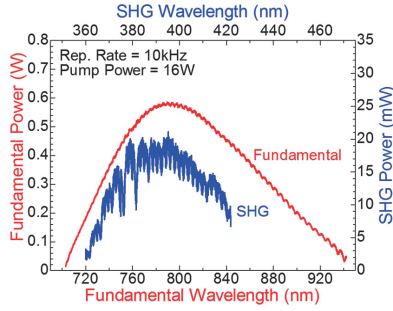


Figure 2. Tuning ranges of the grating Ti:Sa laser and its intracavity SHG.

### 3. Development of two color resonance ionization scheme for Th

#### 3.1. Experimental setup

The most efficient known laser ionization scheme [10] uses three colors for ionization of Th, however, a two laser setup would be preferable for lower installation and operation cost. To show capability of the grating-assisted Ti:Sa laser for resonance ionization of radioisotopes and ionization scheme development, a two color ionization scheme of Th was developed. **Figure 3** shows the experimental setup for investigation of two-color schemes. Th atoms were generated by ohmic heating of a thorium oxide (thoria) - coated tungsten filament next to an acceleration electrode in a small time-of-flight mass spectrometer. **Figure 4** shows resonance ionization schemes of Th used in this work. Conventional Z-cavity Ti:Sa laser, named “standard” Ti:Sa laser #1, was tuned at 372.049 nm for transition to the excited state at 26878.16  $\text{cm}^{-1}$ . Ti:Sa laser #1 was also used for non-resonant ionization. Pulses of the standard Ti:Sa and the grating Ti:Sa laser were overlapped and synchronized and then sent to the laser interaction region between the electrodes. The wavelength of the grating-assisted Ti:Sa laser was scanned from 380 to 430 nm to investigate efficient ionization via Rydberg states or A.I. states. Count rate in region of mass 232 region in each time of flight spectrum was recorded.

During the wavelength scan, the output power of the grating-assisted Ti:Sa laser was measured. In order to monitor temporal variation of atomic source yield, ions generated via time-delayed one-color ionization by an additional laser (a standard Ti:Sa laser #2) at 382.947 nm were counted in shifted region.

#### 3.2. Efficient two-color scheme for ionization of Th

With Ti:Sa#1 at the fixed 1<sup>st</sup> step wavelength of 372.049 nm, the grating Ti:Sa laser wavelength was scanned from 380–430 nm. **Figure 5** shows relative intensity of ion count rate in within this wavelength range (red curve). To confirm that each peak in Figure 5 was obtained via a proper two-color process, a separate spectrum was obtained with detuned 1<sup>st</sup> step wavelength (green curve).

Assuming that each 2<sup>nd</sup> step transition corresponding

to a peak in Figure 5 was not saturated, the detected ion count rate was normalized by the SHG output power of the grating Ti:Sa laser. One of the most efficient 2<sup>nd</sup> step transition was at 401.031 nm to 51813.9  $\text{cm}^{-1}$ . **Figure 6** (a) and (b) show detailed resonance ionization spectra for 1<sup>st</sup> step transition at 372.049 nm, and 2<sup>nd</sup> step transition at 401.031 nm, respectively. Saturation curves of (a) 1<sup>st</sup> step and (b) 2<sup>nd</sup> step transitions are shown in **Figure 7**.

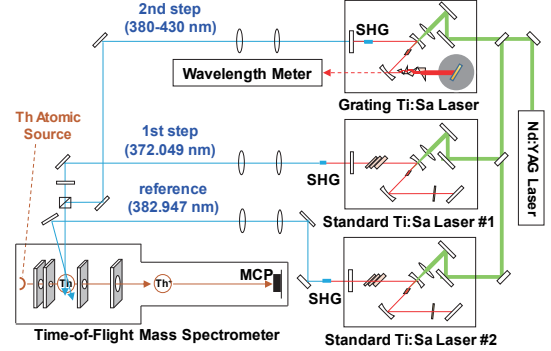


Figure 3. Experimental setup for investigation of two-color scheme for ionization of Th.

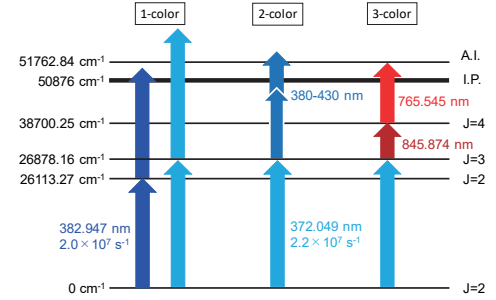


Figure 4. Resonance ionization schemes for Th used in this work. The number indicated below the wavelength is Einstein A coefficient of the transition.

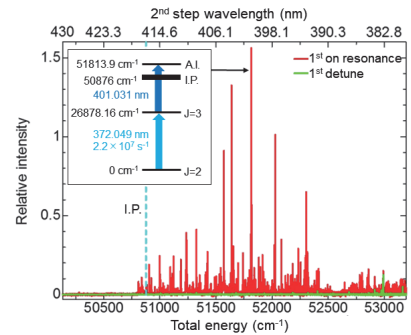


Figure 5. Results of wide wavelength scan in the second harmonic spectral range from 380 to 430 nm.

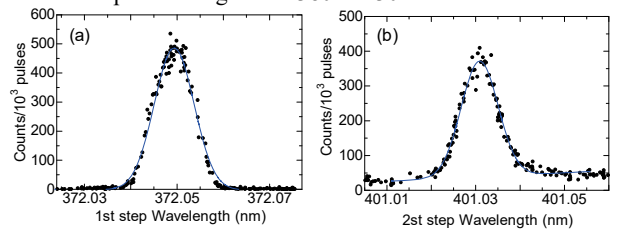


Figure 6. Resonance ionization spectra (a) 1<sup>st</sup> step transition at 372.049 nm, (b) 2<sup>nd</sup> step transition at 401.031 nm.

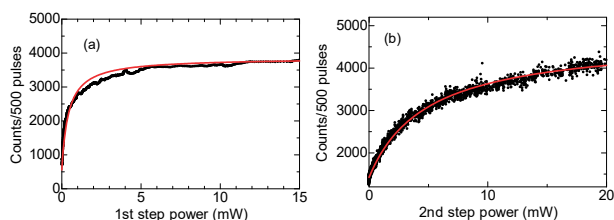


Figure 7. Saturation curves of (a) 1<sup>st</sup> step transition at 372.049 nm, and (b) 2<sup>nd</sup> step transition at 401.031 nm in two-color scheme.

In the three-color scheme, three standard Ti:Sa lasers were used. For efficiency comparison between the two- and the three- color schemes, relative atomic source yield during measurement in the three color scheme was possible to be estimated by detected count rate via one-color scheme (372.049 nm) by temporarily blocking 2<sup>nd</sup> step and 3<sup>rd</sup> step laser pulses. After measurements of saturation curves of each step in the three-color scheme, relative efficiency was evaluated. Efficiency of the two-color scheme (1<sup>st</sup> step: 372.049 nm and 2<sup>nd</sup> step: 401.031 nm) was estimated to be lower by factor of three compared to the three-color scheme.

#### 4. Conclusion

A grating-assisted Ti:Sa laser with computer-control for wavelength tuning and automatically tracking intra-cavity second harmonic generation was developed. Mode-hop free tuning was successfully demonstrated in the fundamental mode as well as in the second harmonic spectral range. A two-color ionization scheme of Th via autoionizing state was developed and its relative efficiency was compared with the known three-color scheme.

Presently we prepare to install the Ti:Sa laser system at a secondary neutral mass spectrometry system for multicolor post resonance ionization for direct and spatially resolved micro-imaging of actinides and other radionuclides in aerosol particles and on surfaces[11].

Furthermore, the laser is suitable for spectroscopic investigations of ionization schemes for gas cell applications such as at the Accelerator Laboratory IGISOL of the University of Jyväskylä, Finland. In a gas-filled environment, conventional schemes developed in high vacuum conditions might turn out to be inefficient and unsuitable due to strong quenching of excited states by collisions with buffer gas atoms.

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