Laser ionization scheme development for in-gas-jet spectroscopy studies of Th^+

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ABSTRACT

The in-gas laser ionization and spectroscopy (IGLIS) technique has been a cornerstone in the study of heavy elements. The addition of a convergent-divergent (de Laval) nozzle to perform laser ionization in a cold hypersonic gas jet greatly improves the achievable resolution. Recent efforts have focused on preparing the in-gas-jet method for the study of the low-lying ^{229}Th isomer. Tailor-made recoil sources of ^{233}U are installed inside a fast extraction gas cell to provide the (isomeric) thorium ions. A level search above the second ionization potential (IP) uncovered several auto-ionizing states, greatly improving the laser ionization efficiency of singly charged thorium ions.

1. Introduction

Laser spectroscopy studies play a key role in the study of actinides. They provide nuclear model-independent measurements of observables such as nuclear spins, changes in mean-square charge radii and electromagnetic moments [1, 2] needed to benchmark nuclear theory in the heavy element region. Moreover, these studies test state of the art atomic theory in a region where electron correlations, relativity and QED effects play an important role in the electronic structure [2]. Experimental data in this part of the nuclear chart are, however, sparse. Many isotopes are short-lived and only available in limited, often only minute quantities which complicates laser spectroscopy studies. The in-gas laser ionization and spectroscopy (IGLIS) [3] technique uses high-purity argon gas to stop, thermalize and neutralize products of reactions in a gas cell. Combining this with ion or characteristic decay radiation detection allows for efficient, low-background in-gas-cell laser spectroscopy measurements with a typical resolution in the GHz range.

The spectral resolution limit in in-gas-cell sepctroscopy is dictated by the Doppler and pressure broadening mechanisms and can be overcome with the use of a convergent-divergent (de Laval) nozzle to create a collimated hypersonic gas jet. The in-gas-jet laser ionization and spectroscopy technique allows for high-resolution ($\sim$ 200 MHz) hyperfine structure measurements. The low temperature and pressure environment as well as good uniformity of flow of hypersonic gas jets provide an ideal spectroscopy environment without sacrificing efficiency [4].

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The isotope ^{229}Th has recently garnered interest as a candidate for use in a new type of optical clock, which will utilize a nuclear transition rather than one in the electronic shells. Very recent measurements have pinpointed the excitation energy of its low-lying isomeric state and gave a first indication of the lifetime [5, 6]. The laser-accessible energy, the isomer's long radiative half-life [7] and the small nuclear moments involved have led to the concept of a so-called nuclear optical clock [8]. This clock is predicted to be highly sensitive to beyond standard model physics due to the strong and electromagnetic interactions in the nuclear environment by usage of clock-comparison experiments [9].

Of note is the fact that the isomer in ^{229}Th has not yet been observed in a singly-charged state. Detections have been made in higher charge states [5, 7, 10, 11], where internal conversion (IC) is energetically forbidden, and in the neutral atom, where IC is the dominant decay path [7, 10]. Based on the experimental conditions of these measurements, it is suggested that the isomer has a half-life of less than 10 ms in the 1+ charge state [10] even though IC is also forbidden in this case. Different depopulation mechanisms might be at play such as electronic bridge processes, bound internal conversion or gas quenching [10]. Moreover, laser spectroscopy studies performed on 2+ and 3+ charge states [11, 12] and improved theoretical predictions show a mismatch in the dipole magnetic moment of the thorium ground state while having good agreement with the isomeric state [13].

Preparatory work for in-gas-jet spectroscopy of $^{229(m)}Th^+$ has been initiated with the goal of observing the signature hyperfine structure of the isomer in the 1+ charge state, opening the possibility for the production of pure isomeric

beams for further use in VUV spectroscopy studies [6] as well as remeasuring ground and isomeric state properties. We report on the production of the isomeric thorium in a dedicated fast-gas cell and the laser ionization scheme developments.

2. Isomer production

Populating the isomeric state has to date been achieved via two avenues: the alpha decay of ^{233}U and the beta decay of ^{229}Ac [5]. The first route is available offline and is used in these studies. To increase the survival chance of the isomeric state, a dedicated fast-gas cell was designed and commissioned [14]. The gas cell is loaded with two cylindrical ^{233}U recoil sources such that the stopping volume containing the recoiled species is evacuated in approximately 1 ms, according to simulations performed using the COMSOL software [14].

Two recoil sources were produced via molecular plating. Each source consists of several foils that were placed in the two cylindrical holders. Source 1 consists of ^{233}U plated on gold-coated tantalum (activity in 2π solid angle: 7.6 kBq, recoil efficiency: 3.8%) from TU Vienna and was used to initially characterize and optimize the transport parameters for thorium in the IGLIS mass separator. Based on the known transport efficiency (66(6)%) of the separator [15], the gas cell extraction efficiency for thorium was estimated to be 30.5(35)% providing 53(3) cps of Th^+ and 14(2) cps Th^{2+} [14].

Source 2 was commissioned with higher activity at JGU Mainz. The source consists of two titanium foils plated with ^{233}U, (activity in 2π solid angle: 30.8 kBq and 9.2 kBq, recoil efficiency: 3.2% and 6.9% respectively). The isotopic composition the uranium is 87.87% ^{233}U, 11.21% ^{238}U, 0.83% ^{234}U and $<$ 0.1% for each isotope of ^{232,235,236}U. Source 2 provides a mass spectrum containing about 680(20) cps of singly-charged thorium ions and 210(10) cps of doubly-charged ions. This would indicate an extraction efficiency of 85(9)%. A mass scan of both sources is shown in Fig. 1.

3. Laser scheme development

The low branching ratio of the alpha decay of ^{233}U to the isomer (2%) necessitates an efficient laser ionization scheme to perform laser spectroscopy on $^{229m}Th^+$. The high second ionization potential (IP) of thorium (between 11.9 – 12.3 eV [16]) forces the use of three laser steps to perform resonance ionization studies. Laser ionization of the singly-charged thorium ions was initially achieved by employing a 2-colour, 3-step scheme as shown in Fig. 2. The scheme was taken from [16]. Here, the third transition step promotes an electron to the continuum using the same laser frequency as the second step. The attained laser ionization efficiency in-gas-cell ($< 0.5\%$) was too low to perform high resolution in-gas-jet spectroscopy. While hundreds of levels are known in Th^+ [17], no auto-ionizing (AI) state is reported on in the literature. Based on a known ionization scheme reported on in [16], a level search above the second IP was conducted.

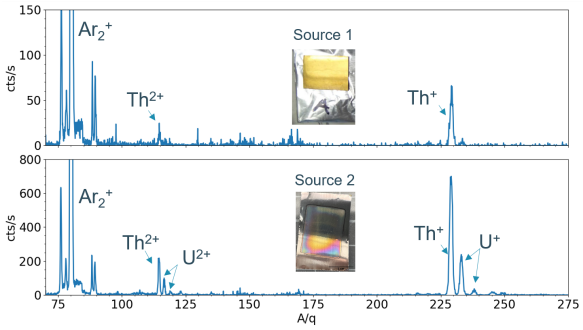

Figure 1: Mass spectra of the two recoil sources. The argon stagnation pressure was 25 mbar and a free expanding jet (exit hole diameter: 1.5 mm) was used. Besides the thorium recoils, sputtered uranium ions are seen both from ^{233}U and an isotopic contaminant ^{238}U.

3.1. Ablation source and lasers

A natural (^{232}Th) metallic thorium target foil (25mm x 25mm, 64 microns, Goodfellows) was installed in an online-type gas cell (see [18]) and ablated using a 20 Hz pulse repetition rate Nd:YAG laser (Quanta-Ray, Spectra Physics Lasers) at 532 nm. In order to have a few thousand ablated ions per second, the Nd:YAG laser beam with an average energy of 5.3 mJ/pulse and pulse length of 10ns was focused on an approximately 1 mm^2 surface. A low stagnation pressure of 25 mbar was used together with a 1.5 mm hole diameter exit aperture to maximise the number of ablation ions.

In order to develop the ionization scheme and perform laser spectroscopy of ^{229m}Th a new laser system was commissioned in the IGLIS laboratory. The setup comprises three high-power, high-repetition rate dye lasers (Credo, Sirah Lasertechnik), each pumped by a separate Nd:YAG laser (Innoslab, Edgewave) with a maximum repetition rate of 15 kHz and 150 W and 75 W laser power at 532 nm and 355 nm, respectively. All three excitation lasers (Fig. 2) were overlapped in transverse geometry through view ports located in the exit channel of the gas cell.

3.2. Results

A range of 1800 cm^{-1} was scanned in the course of several days, yielding the level structure depicted in Fig. 2. Laser powers of the first and second step were kept steady at 900 mW and 10 mW, respectively at a repetition rate of 7 kHz. Lower laser power in the second step ensured the suppression of the 2-colour 3-step photo-ionization that was previously observed and reduced the laser dye degradation over time. A step-size of 0.5 cm^{-1} was used and the recorded signal was integrated over 10 s to account for fluctuations of rate of ablation ions. Counts were normalized with respect to the non-resonant ion background. The bandwidth of the scanning laser was 3.4 GHz.

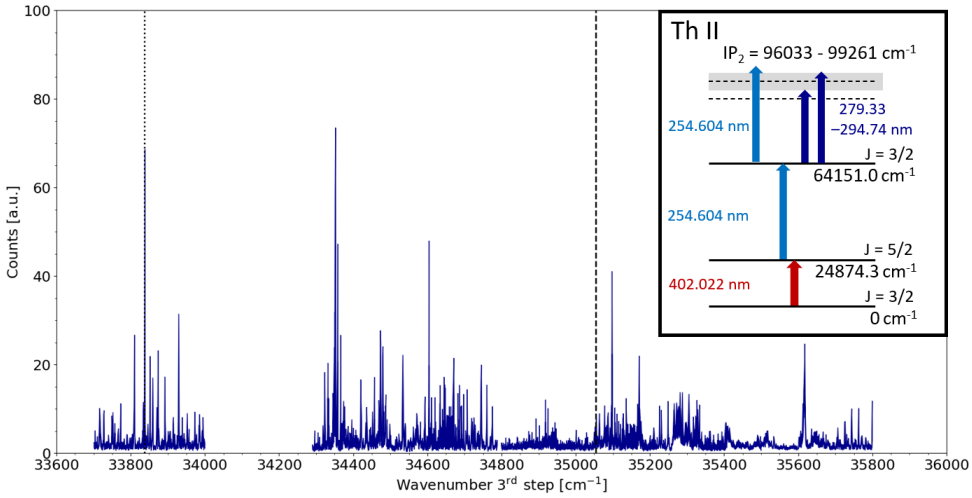

Figure 2: Third step scan range. The dotted line shows the highest intensity AI state. The dashed line shows the upper bound for the second IP at 12.3 eV. Inset: the laser scheme that was used. Left: 2-colour, 3-step scheme described in [16].Right: region covered by the scan.

To cover the selected region, two different dye solutions were used: Rhodamin 6G and Pyrromethene 597, both in ethanol. The lower energy part ($33700 - 34000\,cm^{-1}$) was scanned using a lower power (300 mW) than in the other region where laser power was in the range of $800-1100$ mW due to pump laser issues. Several auto-ionizing states were found. The strongest with an ionization efficiency of 11(5)% was found at 33837.28(2) cm^{-1} (3^{rd} step wavenumber) and is indicated by the dotted line in Fig. 2. In the region of $34300 - 34400\,cm^{-1}$, several strong transitions were observed that originated from the third step alone (1-colour, 3-step). Regular checks were performed to verify whether the photo-ions were produced by a 3-colour, 3-step laser resonant process. Thus an upper bound for the second ionization potential of thorium at 12.14 eV was obtained. Further investigations are planned, specifically the full uncertainty range of the ionization potential will be scanned in order to determine its value with greater precision than is currently available in literature.

4. Outlook

We reported on the progress towards applying in-gas-jet laser ionization spectroscopy on $^{229}Th^+$. With the use of laser ablation, several AI states were identified with the strongest having an efficiency of 11(5)%. A future campaign is planned to determine the second IP experimentally via a scan of the Rydberg series in Th^+. The fast and efficient extraction of recoiling thorium ions from the gas cell makes it possible to perform in-gas-jet spectroscopy of the $^{229}Th^+$ nuclear ground state and a search for the isomer in the "missing" singly-charged state. Besides research on ^{229m}Th, these developments would allow the study of neutron deficient thorium isotopes in the vicinity of the $N = 126$ shell closure and the proton drip line planned at S^3-LEB.

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