Summary of PhD thesis "Gas-phase chemistry, recoil source characterization and in-gas-cell resonance laser ionization of actinides at IGISOL"

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March 19, 2021

The production of radioactive isotopes at radioactive ion beam (RIB) facilities around the world has been essential in the study of fundamental properties of atomic nuclei and in many applications of nuclear decay. RIB facilities are increasingly relying on the thermalization and stopping of radioisotopes in noble gas-filled stopping cells to produce low-energy radioactive ion beams in a chemically unselective manner. In this thesis, several different approaches are studied aimed towards the production of actinide ion beams for high-resolution collinear laser spectroscopy used for the study of fundamental nuclear structure properties of radioactive isotopes at the IGISOL facility [1, 2] of the University of Jyväskylä. The production of actinide beams, namely isotopes of thorium and plutonium, was realized with the gas-cell technique while combining it with laser resonance ionization, alpha-recoil sources and accelerator-based production.

Firstly, the thesis focuses on the most critical aspect in operation of such a gas cell, the noble gas purification and relevant gas-phase ion chemistry which critically affects the efficiency of the gas-cell technique and formation of contaminant ion beams. The construction of the new IGISOL buffer gas purification system [3] accomplished sub-parts-per-billion level of impurity as demonstrated with mass spectroscopic measurements. This enabled subsequent gas cell developments and realization of ion beams of the highly chemically active elements of plutonium and thorium.

One of the main production techniques investigated was evaporations of long-lived isotopes of actinides from filament dispensers in a dedicated gas cell (Fig. 1), while performing the highly efficient and inherently element selective resonance laser ionization [4]. This resulted in the successful creation of intense plutonium ion beams such that optical spectroscopy of several long-lived isotopes of plutonium has been performed. Consequently, plutonium now represents the heaviest element yet measured with the high-resolution collinear laser spectroscopy technique [5].

Thorium, which is of great interest due to its exceptionally low-lying nuclear isomeric state in ^{229}Th of only 8.28 eV [6], was also a focus of this thesis. Similar to plutonium, the filament-based dispenser for thorium was successful in the creation of a thorium ion beam, however, the required filament temperature resulted in significant molecular contaminant beams. This prompted the study [7] of several different types of filament dispenser in collaboration with the Institute of Atomic and Subatomic Physics of TU Wien. These highlighted the importance of manufacturing filament dispensers with good cleanliness and with sufficient reduction capability while maintaining a good structural integrity of the filaments.

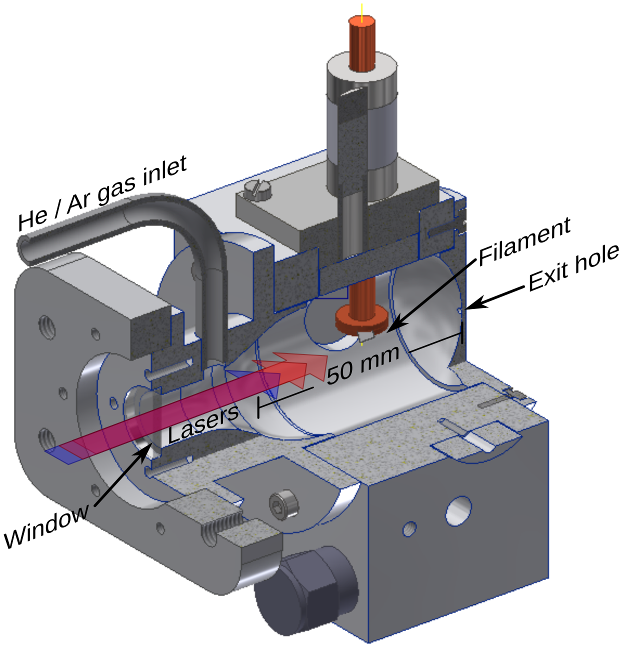

Figure 1: Cross-sectional view of the gas cell used in this thesis work.

During the development of the plutonium and thorium laser ion source, a collisional quenching phenomenon was discovered to have a significant effect on the ionization process. The resonance excitation, which had been efficiently realized in earlier hot-cavity studies performed under vacuum, showed little response or unexpected behavior in the gaseous environment of the gas cell. This led to a fruitful collaboration with Nagoya University, Japan, and joint investigations of the plutonium ionization scheme with a frequency-doubled grating-based Ti:sapphire laser. This resulted in a dramatically expanded set of atomic levels and a clearer understanding of the behavior of the ionization scheme (Fig. 2), which was lacking prior to this

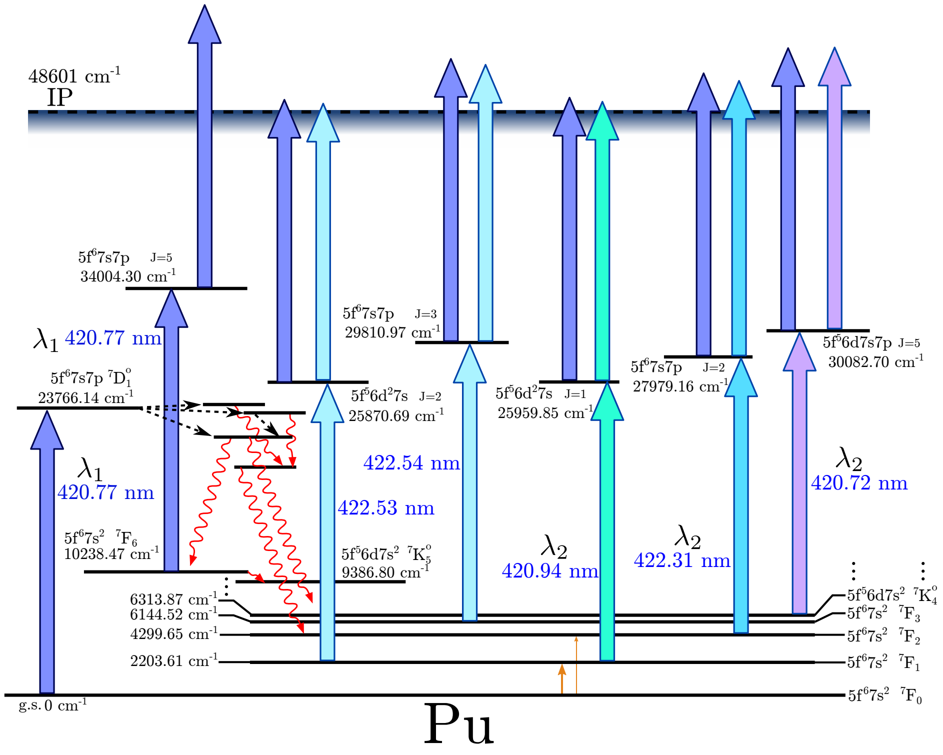

Figure 2: The level scheme constructed from the interpretation of the results obtained from the plutonium studies with the grating-based laser. Thick arrows correspond to detected laser transitions each with its own color. Dashed arrows show collisional transitions and the wavy lines radiative transitions. The orange arrows from the ground state depict thermal excitation. The level positions are to scale if energy and configuration is given.

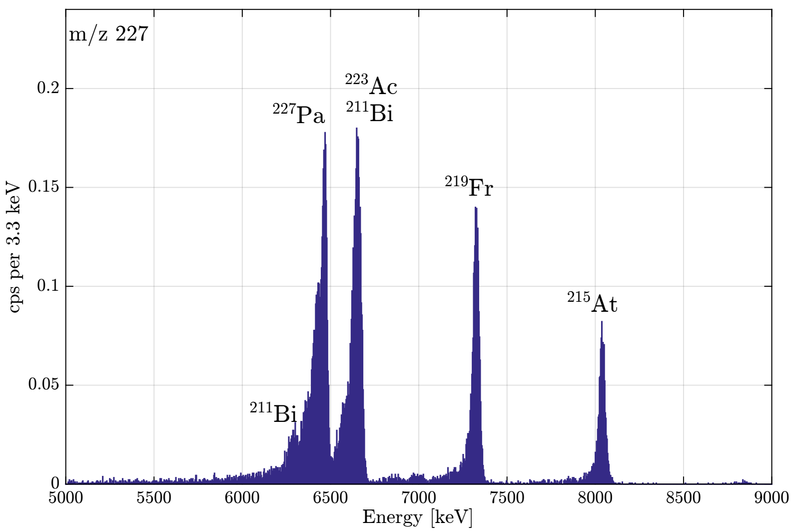

Figure 3: The alpha spectrum recorded from a Si detector onto which the mass-separated ion beam (A/q=227) was implanted. The alpha lines were identified to belong to isotopes in the decay chain of ^{227}Pa.

work.

The second investigated approach for the production of a thorium ion beam took advantage of the long-lived alpha active isotope of ^{233}U. By stopping ^{229}Th alpha decay recoils from ^{233}U in a helium-filled gas cell, a ^{229}Th ground and isomeric state ion beam can be produced. By mounting a ^{233}U source into the gas cell, beams of ^{229}Th^{3+} were detected and charge state manipulation to ^{229}Th^{2+} was demonstrated following the addition of a trace amount of Xe to the helium buffer gas. The recoil efficiency determination of two ^{233}U sources using direct and implantation foil gamma- and alpha-ray spectroscopy as well as surface characterization by Rutherford back scattering (RBS) measurements have shown the importance of good source quality [8]. The development of a new gas cell to house several such recoil sources is also presented, emphasizing the interplay between gas pressure, size of the gas cell, and diffusion losses during ion extraction.

Finally, a first on-line experiment to study the creation of singly-charged ^{229}Th^{+} ion beams through fusion-evaporation by bombarding a ^{232}Th target with high-energy protons was also realized. This experiment highlighted, through the measurement of alpha-decay lines (Fig. 3) of the fusion-evaporation products, that several heavy actinide isotopes can be successfully produced and extracted as ion beams from the IGISOL gas cell. This has not only given confidence to the realization of a high on-line yield of ^{229m}Th, but also promises access to elements and radioisotopes previously not studied using optical techniques, where fundamental ground-state nuclear structure including spins, electromagnetic moments and changes in mean-square charge radii may be accessed for the first time. This would complement efforts underway at other facilities utilizing heavy-ion fusion-evaporation reactions and in-flight separators. The development of new, more durable, targets also from long-lived radioisotopes in the actinide region, could expand the accessible isotopes for spectroscopic studies. Furthermore, the chemical non-selectively of the IGISOL method can be a great advantage if the reaction cross sections, especially relative changes in them, are to be experimentally determined.

The thorium ion source studies in this thesis are performed within the nuClock project [9], part of the Horizon 2020 EU framework program, which aims to develop a novel type of optical clock based on an extremely low-lying nuclear transition in ^{229}Th. The nuclear approach to a frequency standard can potentially reach about an order of magnitude higher precision than the current atomic clocks.

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