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Investigating radioactive negative ion production via double electron capture

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ABSTRACT

The relative cross sections for radioactive negative ion production via double electron capture have been measured for collisions between a 40 keV projectile beam of uranium-238 and potassium vapor. This was performed at the collinear resonance ionization spectroscopy (CRIS) experiment at CERN-ISOLDE and is a step towards measuring the electron affinities (EAs) of elements that cannot be efficiently produced in negative ion sources at radioactive ion beam (RIB) facilities. This includes short-lived radioactive isotopes that have low production quantities and heavy and superheavy elements that systematically have smaller EAs than work functions of available ion source materials. Negative ions are particularly sensitive to electron–electron correlation effects, which make such studies ideal for benchmarking atomic structure models that go beyond the independent particle model. While the EAs of most light elements have been measured, experimental investigations on heavier elements, namely the actinides, remain scarce due to their radioactive nature and production difficulty. By developing negative ion production by charge exchange, we aim to make these studies feasible at RIB facilities.

1. Introduction

Negative ions have a valence electron that is bound in a shallow induced dipole potential which arises from pronounced electron–electron correlation effects and are thus excellent systems to investigate these correlations [1]. Due to this weak potential, negative ions have binding energies that are usually an order of magnitude smaller than neutral atoms. This binding energy is called the electron affinity (EA), which is the amount of energy released when an electron binds to a neutral atom to make it negatively charged [2].

Electron affinity measurements are useful for benchmarking calculations of highly-correlated and relativistic systems [3], targeted alpha therapy using astatine-211 [4], uranium mine management [5], and future production of antiprotonic ions [6], to name a few. By measuring the isotopic shift (IS) in EAs of many-electron systems one can determine the specific mass shift (SMS). This quantity is sensitive to electron–electron correlation unlike the normal mass shift (NMS), which is a purely kinematic effect. The SMS cannot be readily calculated and otherwise prevents the extraction of the field shift (FS) from IS measurements. Therefore, measuring the SMS is relevant for nuclear physics where the FS is necessary for determining changes in mean-square charge radii and, in turn, nuclear charge radii [7,8].

Traditionally, negative ions have been produced in sputter and plasma ion sources which require large samples, and surface ion sources which work most effectively for elements with EAs larger than the work function of the material [9]. The first EA measurements for radioisotopes were iodine-128 (^{128}I) [10] and astatine-211 (^{211}At) [11] performed at CERN-ISOLDE. They were accomplished with negative ions produced from an LaB_6 surface ionization source ($\phi_{eff} \approx 2.6$–2.9 eV) [12–14]. However, when producing beams of radioactive negative ions that have relatively low EA values, the aforementioned

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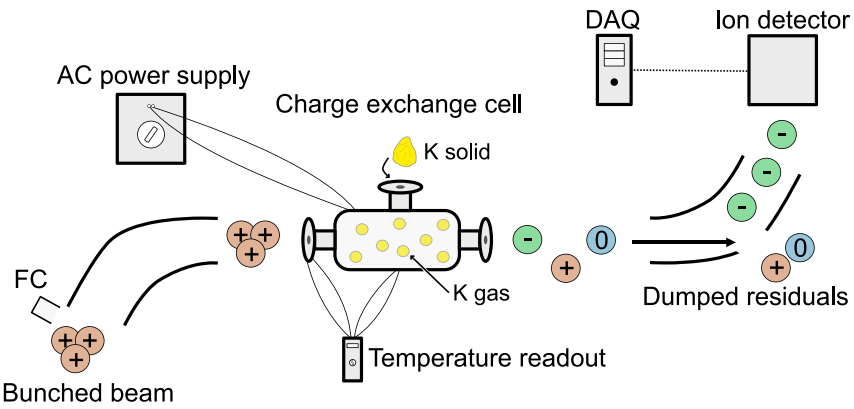

Fig. 1. The cooled and bunched positive ion beam from ISOLDE (red) was guided to the CRIS experimental setup and into the charge exchange cell (CEC) where solid potassium (K) was inserted from the top and heated until gas phase was reached. With increasing pressure, two separate electron capture reactions between the projectile beam and the K atoms in gas phase (yellow) occurred to produce negative ions (green). The remaining positive ions that did not capture any electrons and the neutral atoms (blue) that only underwent one electron capture reaction were dumped while the negative ions were electrostatically bent towards the ion detector, which was connected to the data acquisition system (DAQ). The CEC was heated by the AC power supply and the temperature was read with thermocouples from the CEC end and center, with the center temperature values recorded for the measurement. Finally, the transmission efficiency measurements were taken between the Faraday cup (FC) and the ion detector for positive ions.

methods will not suffice. An alternative to producing such beams is with a projectile beam of positive ions that collides with a target vapor and captures two electrons, also known as double charge transfer or exchange. This has been studied with metal vapor targets such as cesium [14,15], magnesium [16], and sodium [17], but has been limited to stable projectiles. While neutralization cross sections have been studied for elements up to Z = 89 [18], further investigations are required to understand radioactive negative ion production with charge exchange.

As we improve and develop production and experimental techniques, studying radioactive elements, such as the actinides, can become more attainable. For these negative ion studies, we need to rely on production via double electron capture reactions. Here, we demonstrate the feasibility of negative ion production in a charge exchange cell with uranium-238 (^{238}U) at the collinear resonance ionization spectroscopy (CRIS) experiment at ISOLDE [19,20].

2. Experimental method

2.1. Negative ion production

Negative ions can be produced via two sequential electron capture reactions from a projectile beam of positive ions and an electron donor target, most commonly gas phase alkali metal atoms. This process is known as double charge exchange and depends on the projectile beam energy, the EA of the projectile, the ionization potential (IP) of the charge exchange target vapor, and the vapor density [9,14]. The double electron capture process can be described by the following reactions:

$$X^+ + Y \rightarrow X + Y^+ + \Delta E_1 \quad (1)$$

$$X + Y \rightarrow X^- + Y^+ + \Delta E_2 \quad (2)$$

Here X is the projectile beam, Y is the target vapor, and ΔE is the energy defect for the reaction. In the first electron capture reaction (1), ΔE_1 is equal to $IP(X) - IP(Y)$, while in the second electron capture reaction (2), ΔE_2 is equal to $EA(X) - IP(Y)$. Negative ion yields are optimal when the energy defects for both reactions are as small as possible [9,14]. For ion beam neutralization, it is common to choose elements that have similar IPs to ensure a relatively large atomic ground state population. This cannot be done for the latter reaction because EAs are significantly smaller than IPs. Therefore, it is necessary to minimize the energy defect as it will always be negative.

2.2. Experimental parameters and setup

Radioactive isotopes are produced when protons from the PSBooster impinge on the ISOLDE target which are subsequently ionized and extracted as a positively charged ion beam [21]. The beam is then mass selected by the high-resolution separator and cooled and bunched in a gas-filled radio frequency quadrupole linear Paul trap (ISCOOL) [22]. For this experiment, a beam of $^{238}U^+$ with an energy of 40 keV was produced with a pre-irradiated uranium carbide target and the resonant ionization laser ion source (RILIS).

After the $^{238}U^+$ beam reached the CRIS experimental setup, the ions entered the potassium (K) filled charge exchange cell (CEC) where two electron capture reactions must occur for negative ion production, as previously explained. Potassium was chosen for the charge exchange target vapor due to its lower first IP compared to the other available option, sodium (Na) (IP(K) = 4.34 eV and IP(Na) = 5.14 eV [23]; EA(U) = 314.97(9) meV [24]). The CEC was resistively heated with cartridge heaters and a Variac variable transformer AC power supply (200 V) to a maximum relative temperature of ~140 °C. The recorded temperature of the cell was taken from the center with thermocouples. It took about one hour of consistent heating to collect all data points.

After production, the negative ions were electrostatically separated from the residual products and detected with a MagneToF single ion detector. This required the polarity of the electrostatic benders to be flipped. Lastly, there was a maximum transmission efficiency for positive ions of ~5% to the end of the CRIS beamline from ISOLDE. This was determined by Faraday cup measurements after ISCOOL and MagneToF detector measurements in single ion counting mode placed after the bend. The experimental setup is shown in Fig. 1.

3. Results and discussion

We successfully produced $^{238}U^-$ in a CEC from collisions between a 40 keV projectile beam of $^{238}U^+$ and gas phase K atoms. The results are shown in Fig. 2. We recorded negative ion production rates as a function of increasing temperature to determine optimal production conditions. The first electron capture reaction dominates the onset of the curve at lower temperatures. As the pressure in the cell increases, the cross section for negative ion production also increases. The second electron capture reaction is prevalent until the saturation point at around 134 °C. We then see a decrease in negative ion production. This can be explained by a third reaction where the negative ion is destroyed due to sufficiently high pressure within the cell. A similar temperature dependence curve was observed by Alton et al. for production of negative calcium ions with a lithium vapor target [25].

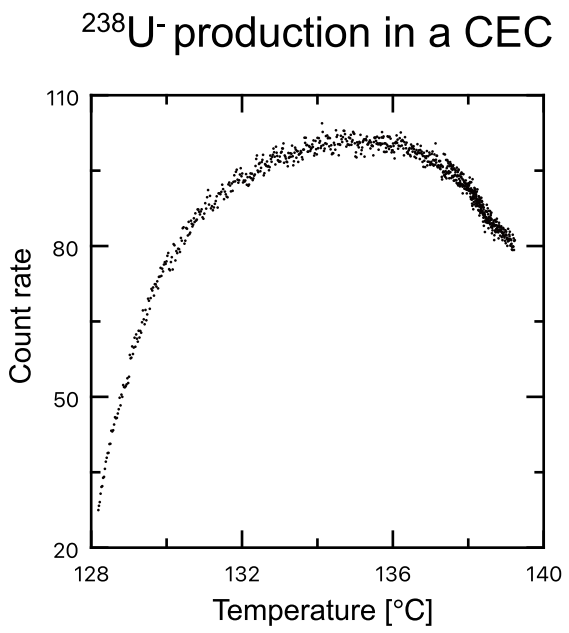

Fig. 2. Temperature dependence of negative uranium-238 ion production in a charge exchange cell as measured with a MagneToF detector. A 40 keV projectile beam of positive uranium-238 ions collides with potassium vapor to undergo a single electron capture reaction. As the temperature in the cell increases, so does the probability for a second electron capture reaction until the curve saturation point at ~134 °C. As temperature continues to increase, the cross section of a third collision also increases in which the negative ions are subsequently destroyed.

The cross section of negative ion production with charge exchange heavily depends on the target vapor density. However, we were unable to determine the K vapor density because the absolute temperature of the vapor was unknown due to the measurement method. From Heinemeier et al. we also know that beam energies between 10 and 30 keV typically result in higher efficiencies of negative ion production [16, 17]. From their results for similar EA elements, we could expect an efficiency for $^{238}U^{-}$ production at ~10%. For technical reasons, we did not have a choice in beam energy nor were we able to determine production efficiency because we did not have a way to detect all three charge states simultaneously.

4. Conclusion

We have shown that ^{238}U negative ions can be produced in a charge exchange cell. Future studies of interest include varying the projectile beam energy, projectile element, and charge exchange target. Upgrades to the pre-existing beamline and detectors are required to facilitate these studies. The aim is to open up possibilities of studying short-lived radioactive isotopes that cannot be produced in efficient quantities from sputter, plasma, or surface ion sources. We are most interested in studying heavy, radioactive negative ions, such as the actinides, because little is known about their atomic properties. However, there are still lighter radioactive elements, such as polonium and francium, which have EA values that are experimentally unknown and will be investigated. By measuring these elements first, we will be able to better understand the production mechanisms and develop more precise experimental techniques.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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