Laser photodetachment of radioactive ions: towards the determination of the electronegativity of astatine

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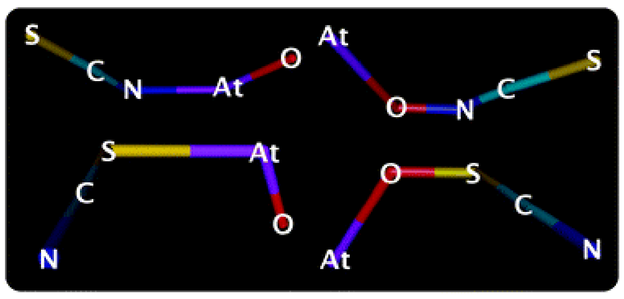
Motivation: astatine chemistry

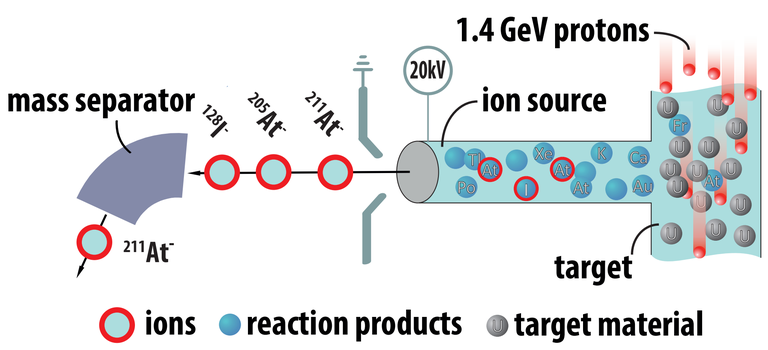
The radioactive element astatine is the rarest naturally occouring element on Earth, with an abundance of 70 mg in the crust [1]. Artificial production of long lived isotopes (e.g. ^{211}At, $t_{1/2}$=7.22 h) enables to conduct only limited physico-chemical studies.



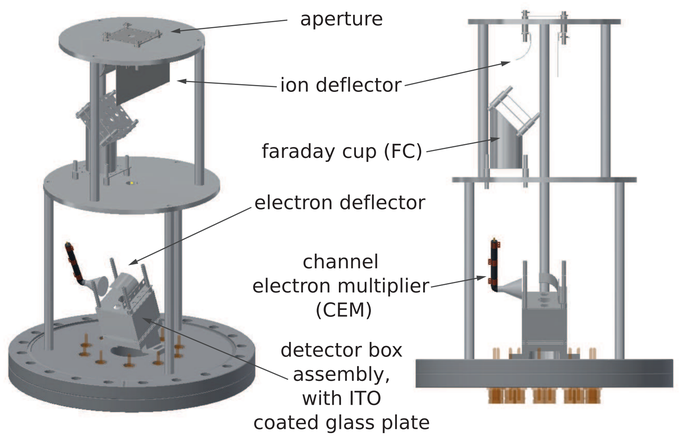
The chemical behaviour of astatine in compounds can be predicted through in-silico calculations [2] which require experimental values for fundamental properties such as the ionization potential (IP) and the electron affinity (EA). Following the successful measurement of IP(At) [3] we present here the ongoing programme to measure the EA(At) to be finally able to determine the electronegativity of astatine via equation 1.

Isotope production

The isotope separator on-line method is used at ISOLDE [4], a radioactive ion beam facility located at the CERN accelarator complex.



The detector



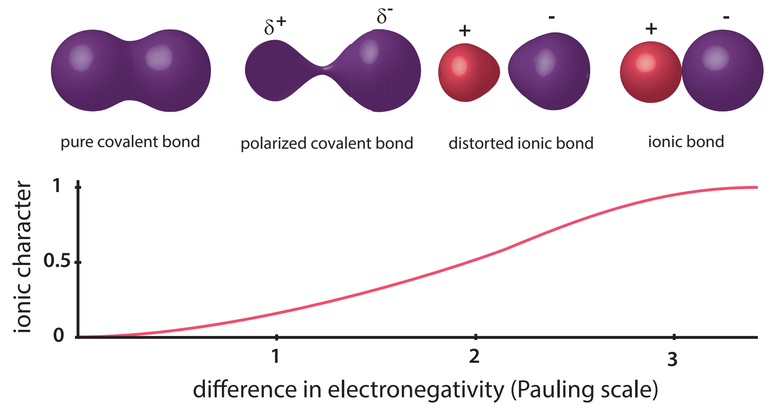
Electronegativity

The Mulliken electronegativity scale is defined by:

$$X^{\mathrm{M}} = \frac{IP + EA}{2} \qquad (1)$$

and can be connected to the established empirical Pauling scale [5].

- EA = electron binding energy in negative ion
- IP = atomic electron binding energy



Laser photodetachment

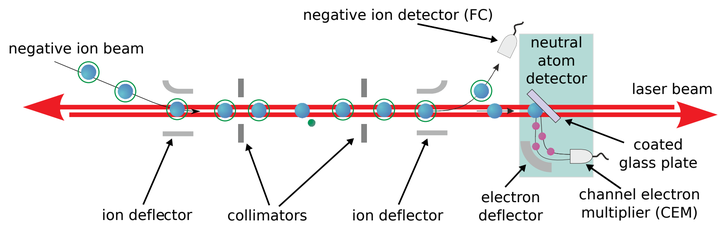
The EA can be measured using the laser photodetachment spectroscopy method [6]. The loosely bound extra electron in a negative ion can be detached due to the absorption of a photon according to the process

$$\gamma + A^- \rightarrow A + e^-. \qquad (2)$$

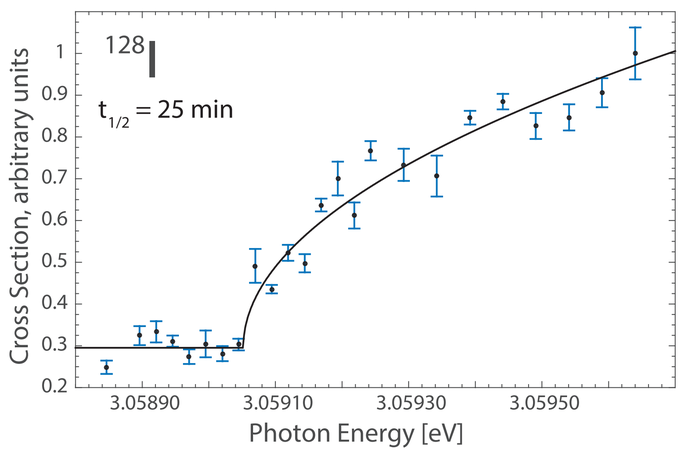
The probability for photodetachment to take place is given by the Wigner law

$$\sigma_{\mathrm{PD}} \propto E_{\mathrm{e}}^{l+1/2} = (\hbar\omega - E_{\mathrm{Th}})^{l+1/2}, \qquad (3)$$

where E_{e} is the energy and l is the angular momentum of the detached electron [7]. E_{Th} is the threshold energy needed for the electron to detach and $\hbar\omega$ is the incoming photon energy.



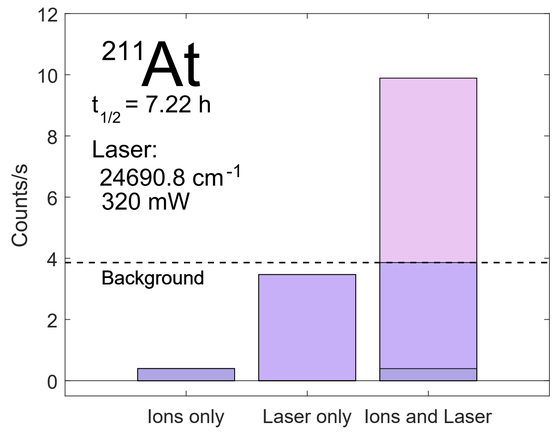
Electron affinity of ^{128}I



- Ion rate (^{128}I): 35000 s^{-1}
- Measurement time: 39 min
- Fit eqn.3: EA(^{128}I)=3.059 052(38) eV [8]

This result marks the first measurement of the electron affinity of a radioisotope. The laser bandwidth of 10 GHz did not allow to resolve the difference in EA between stable ^{127}I and ^{128}I. Isotope shift measurements of lighter elements are subject to future experiments.

Photodetachment of ^{211}At



- Ion rate (^{211}At): 6200 s^{-1}
- Measurement time: 1 h
- EA(At) < 3.06 eV

The first laser photodetachment signal of astatine marks a milestone towards the measurement of the photodetachment threshold and therefore the EA.

Outlook

- Reduce background: second CEM for tuning
- Add photoelectron suppression
- Optimize the neagtive ion source
- ISOLDE beam time request submitted

References

[1] Asimov, I. The natural occurrence of short-lived radioisotopes. *J. Chem. Educ.* **30**, 616 (1953).

[2] Champion, J. *et al.* Assessment of an effective quasirelativistic methodology designed to study astatine chemistry in aqueous solution. *Phys. Chem. Chem. Phys.* **13**, 14984 (2011).

[3] Rothe, S. *et al.* Measurement of the first ionization potential of astatine by laser ionization spectroscopy. *Nature Communications* **4**, 1835 (2013).

[4] Kugler, E. The isolde facility. *Hyperfine Interactions* **129**, 23–42 (2000).

[5] Herrick, D. R. Connecting pauling and mulliken electronegativities. *Journal of Chemical Theory and Computation* **1**, 255–260 (2005). PMID: 26641296.

[6] Rienstra-Kiracofe, J. C. *et al.* Atomic and Molecular Electron Affinities: Photoelectron Experiments and Theoretical Computations. *Chemical Reviews* **102**, 231–282 (2002).

[7] Wigner, E. P. On the Behavior of Cross Sections Near Thresholds. *Phys. Rev.* **73**, 1002–1009 (1948).

[8] Rothe, S. *et al.* Laser photodetachment of radioactive 128-iodine. *J.Phys.G, accepted* .

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