Experimental and theoretical investigations of negative ions

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Outline

- Negative ion structure
- Experiments
 - Electron affinity measurements with partial cross-sections
 - Negative ion production with electron capture
 - Lifetime and isotope shift measurements with Sn
- Theory
 - Methods for calculating negative ion production cross-sections
- Summary and outlook





Introduction







Negative ion dynamics and structure

- The electron affinity (EA) is the binding energy of the additional electron to the neutral atom (~ few eVs)
- Electron-electron correlation is more prevalent due to the shallow potential of valence electron
- Usually, only one bound excited state where most transitions are optically forbidden



ΕA

A⁻

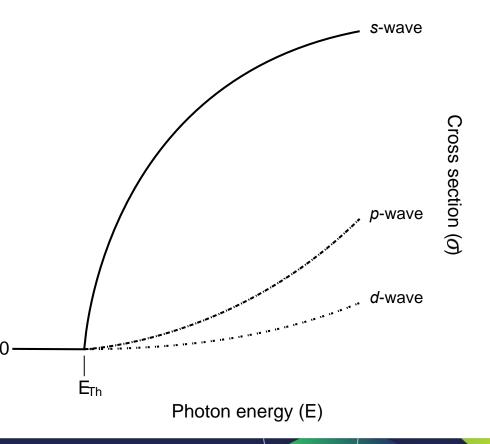
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Energy



Electron affinity measurements

• For the most part, to extract atomic data from negative ions they must be destroyed in a process called photodetachment





Experiments

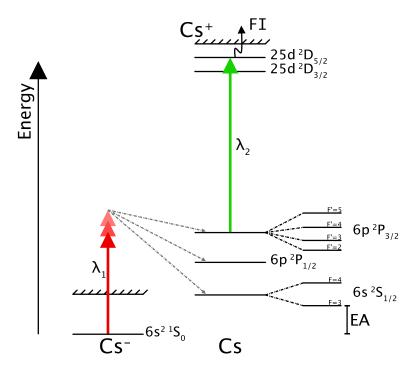


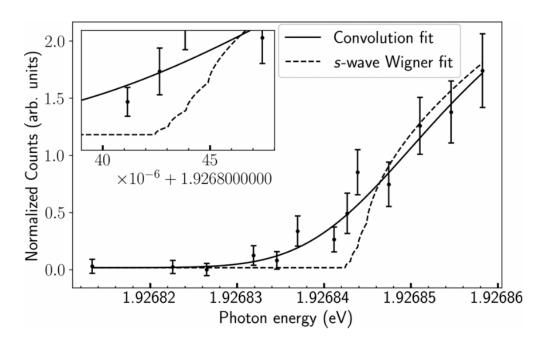




Electron affinity measurements of Cs and Rb

 Can achieve higher precision measurements by combining two spectroscopic techniques: laser photodetachment threshold spectroscopy (red arrows) and resonance ionization spectroscopy (green arrow) [1,2]





[1] Navarro-Navarrete *et al., PRA* **109** 022812 (2024)
[2] Ringvall-Moberg *et al., J. PHYS. B* **57** 155002 (2024)



Negative ion production

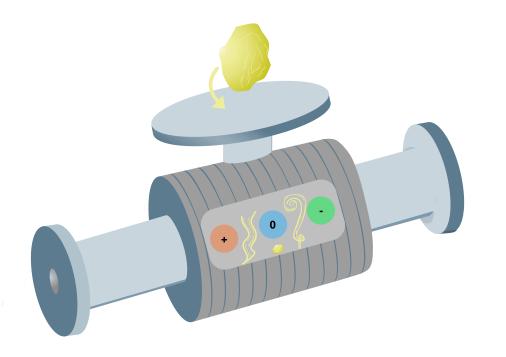
- Surface ionization
 - Requires $EA(x) < \phi(y)$
- Plasma & sputtering
 - Requires large samples

When it comes to production of small EA and low yield elements, such as the actinides, there must be a better way...



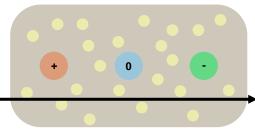


U⁻ production via charge exchange



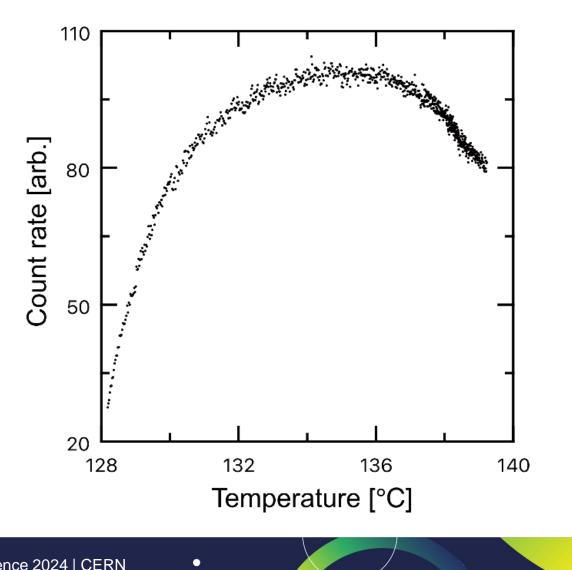
There are multiple reactions to consider:

$$U^{+} + K \rightarrow U + K^{+} + \Delta E$$
$$U^{-} + K \rightarrow U^{-} + K^{+} + \Delta E$$
$$U^{-} + K \rightarrow U + K + e^{-}$$



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Nichols et al., NIM B 541 264-267 (2023)



E1 allowed transitions in negative ions

- Most negative ions have *E1* forbidden transitions due to parity selection rules
- However, 5 elements do have E1 allowed transitions within the anion: Ce⁻, La⁻, Os⁻, Th⁻, and U⁻
 - Interesting for laser cooling but a nightmare to study
- If there are bound states, they are commonly long lived and decay via forbidden transitions
 - Lifetimes are measured instead of transitions

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Measuring radiative lifetimes

- E1 forbidden transitions lead to long lifetimes
- Photodetaching negative ions to study yield of neutrals over time
- Laser power is an important factor and must be as low as possible

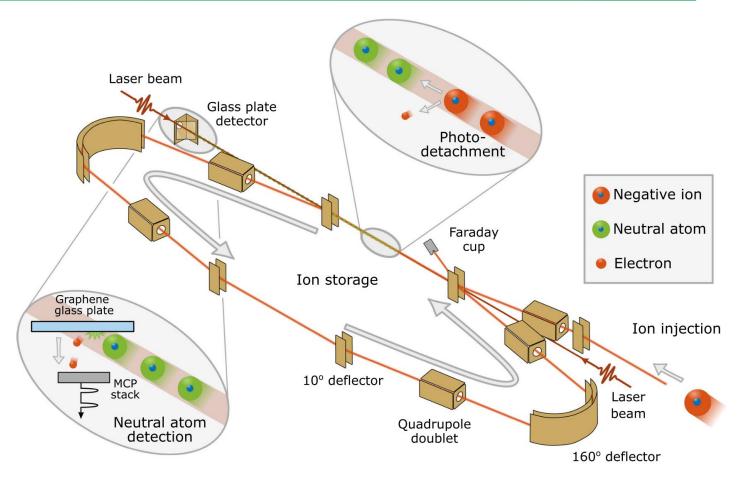


Image courtesy of M. K. Kristiansson et al., Nat. Commun. 13 5906 (2022)



DESIREE: The Double ElectroStatic Ion Ring ExpEriment

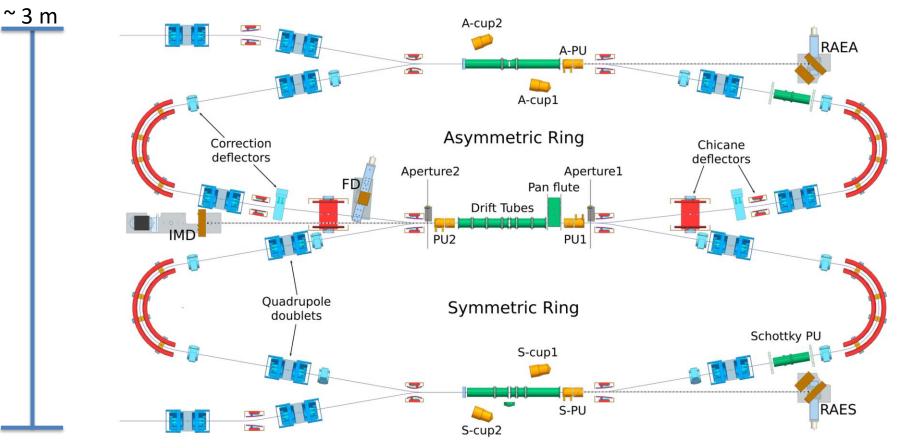


Image courtesy of https://www.desiree-infrastructure.com/experiments

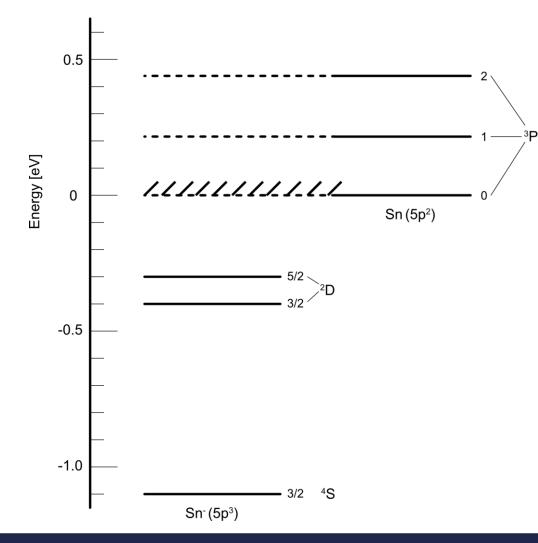
- C = 8.6 m / ring
- 10⁻¹⁴ mbar vacuum
- 13 K temperature
- Symmetric ring → up to 35 keV
- Asymmetric ring → up to 100 keV





The structure of carbon group anions

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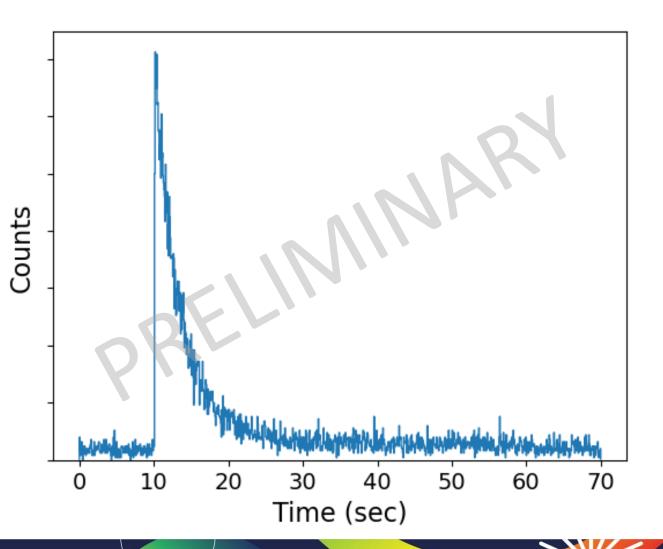


There's a ²D_J state that decays mainly through *M1*!



Lifetimes of the bound Sn⁻ states

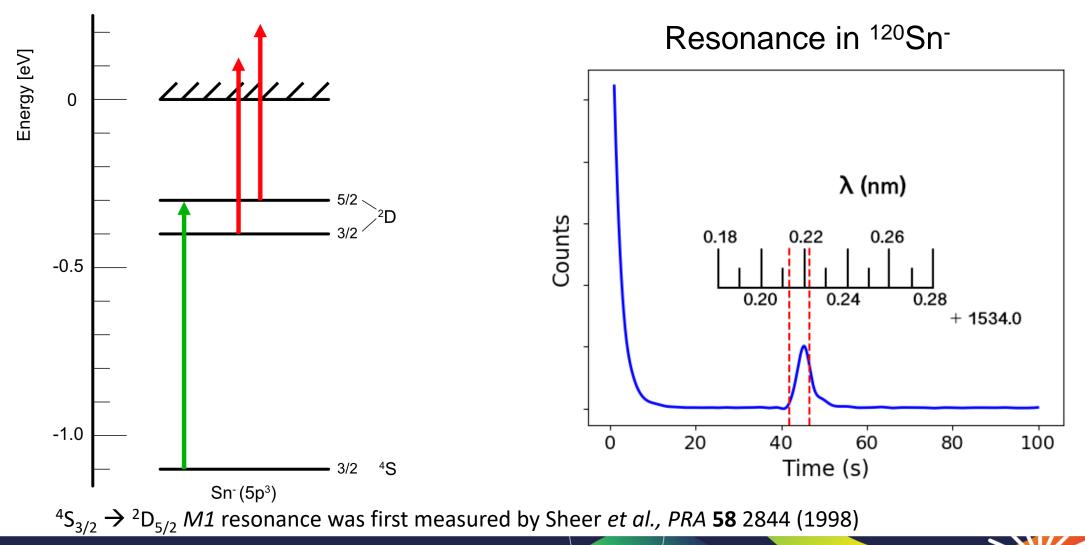
- Measure yield of neutrals
- Storage ~2950 s
- Lifetime of $D_{3/2}$ ~12s
- Lifetime of $D_{5/2}$ ~60s
- To distinguish states:
 - Deplete both states, then pump-probe a single state
 - Wait for shorter lived state to decay





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Do wave functions behave as we expect?



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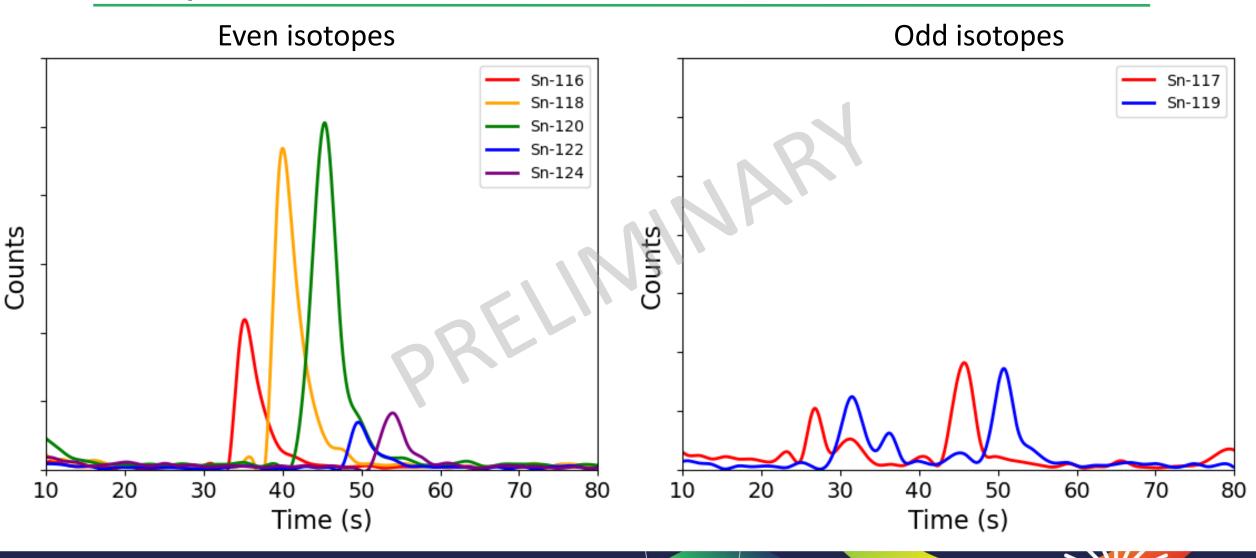
Laser-driven electronic state manipulation

- This system has allowed for spectroscopic investigation of forbidden transitions
 - How is it possible? Time!
 - Most linear experiments only probe for a few microseconds
 - At DESIREE, ions can be illuminated for hours
- With essentially infinite signal to background:
 - All bound transitions in negative ions can be studied, E1 forbidden or not
- Can allow for more clear lifetime measurements where two states are present
- This is also of interest for mutual neutralization studies





Isotope measurements



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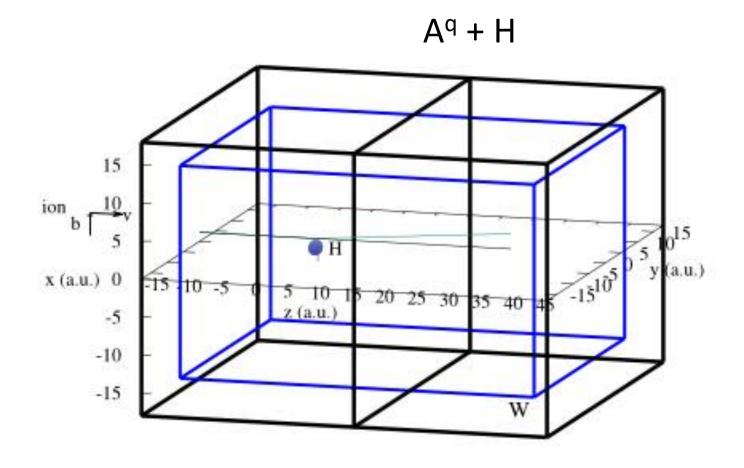






Collisions of bare ion projectiles, Z = 1 - 4

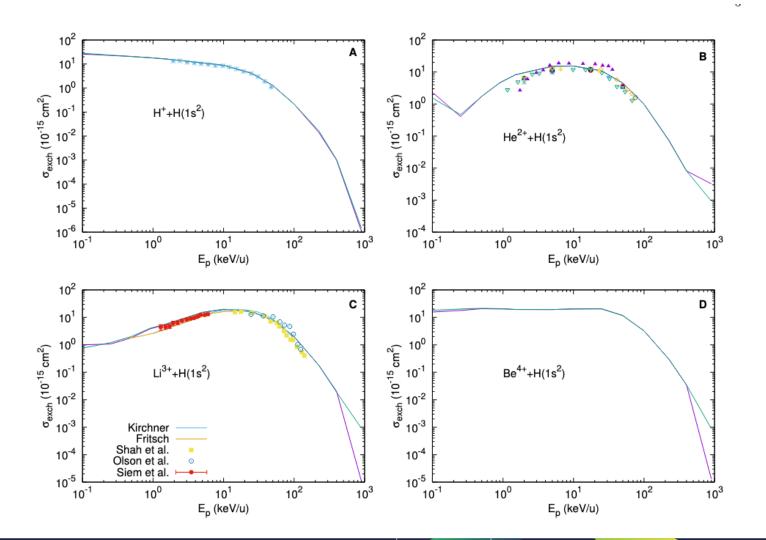
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- Investigating trajectory effects on charge exchange
- Solving TDSE with Crank-Nicholson approach within finite differences method
- Electro-nuclear dynamics solved with 4th order Runge-Kutta method



Electron capture cross-sections



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Conclusions







Summary and outlook

- Discussed how to study different aspects of negative ions:
 - State-selective EA measurements with alkali metals
 - Negative ion production for low EA and small sample elements
 - Laser-driven forbidden transitions and state manipulation for IS measurements in Sn
- Future work for my PhD and beyond:
 - Remeasure the IS of Sn for higher accuracy
 - Calculate electron capture cross sections for actinides using Thomas-Fermi-Amaldi
 - Method is set for EA of Fr measurement





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