Experimental and theoretical investigations of negative ions

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LISA
LASER IONISATION AND SPECTROSCOPY OF ACTINIDES

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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

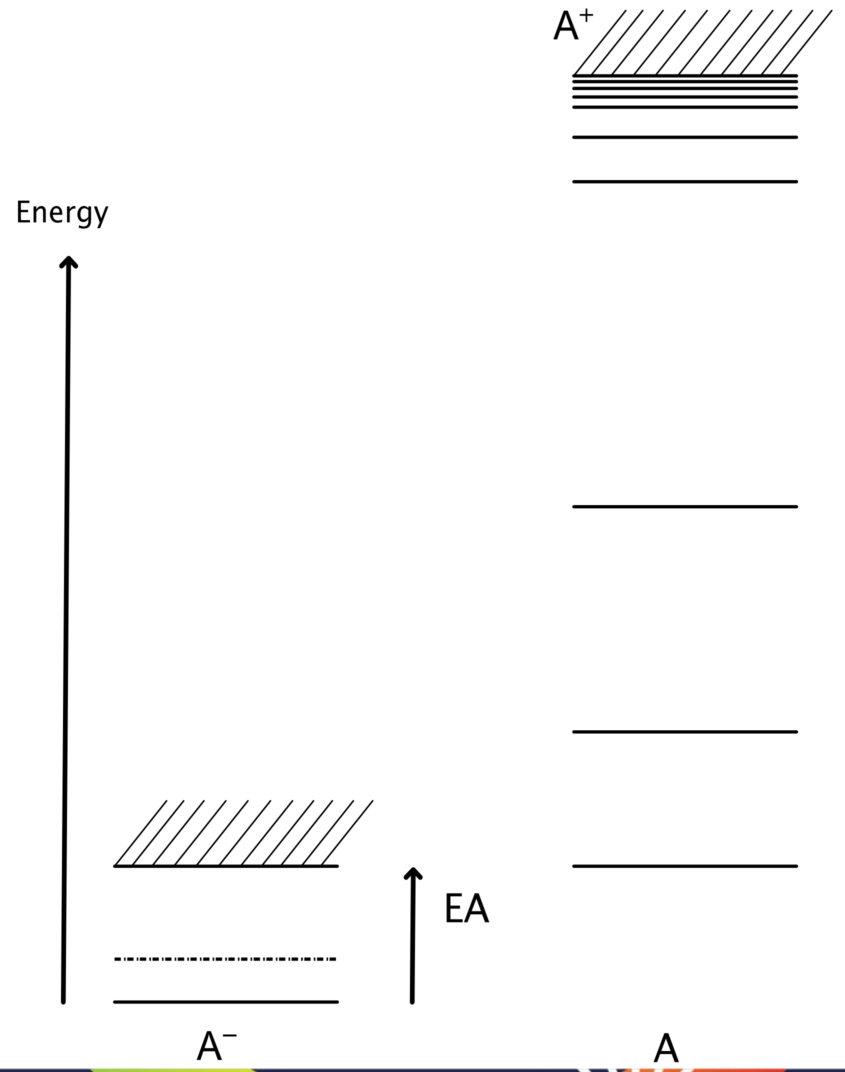
Introduction

Negative ion dynamics and structure

- The electron affinity (EA) is the binding energy of the additional electron to the neutral atom (~ few eVs)

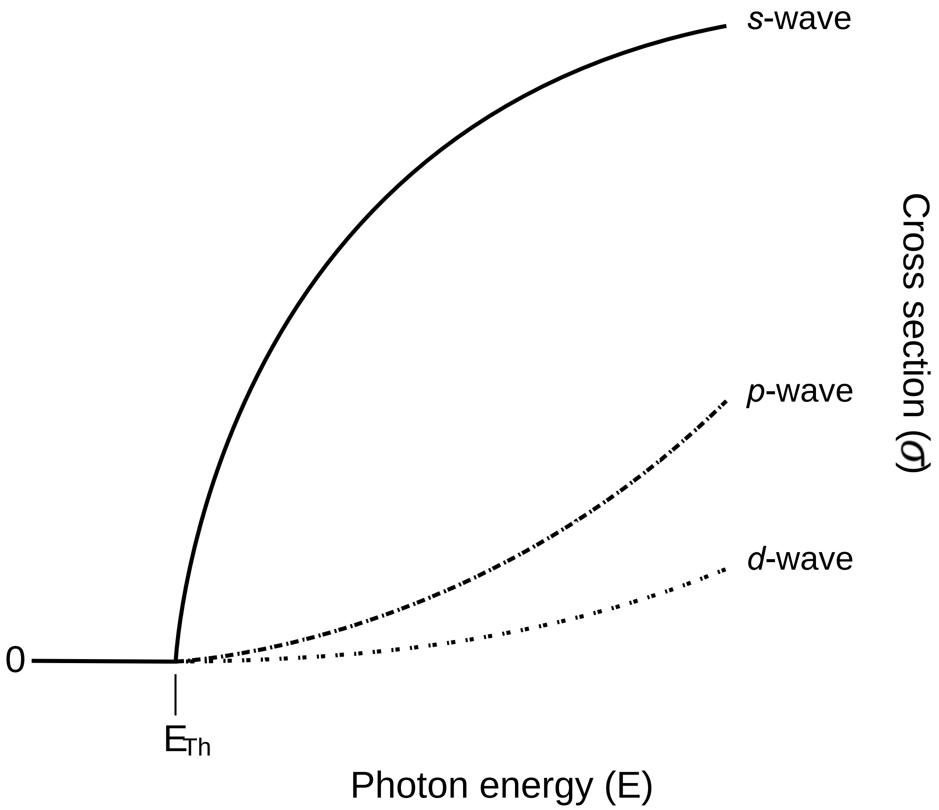
- Electron-electron correlation is more prevalent due to the shallow potential of valence electron

- Usually, only one bound excited state where most transitions are optically forbidden



Electron affinity measurements

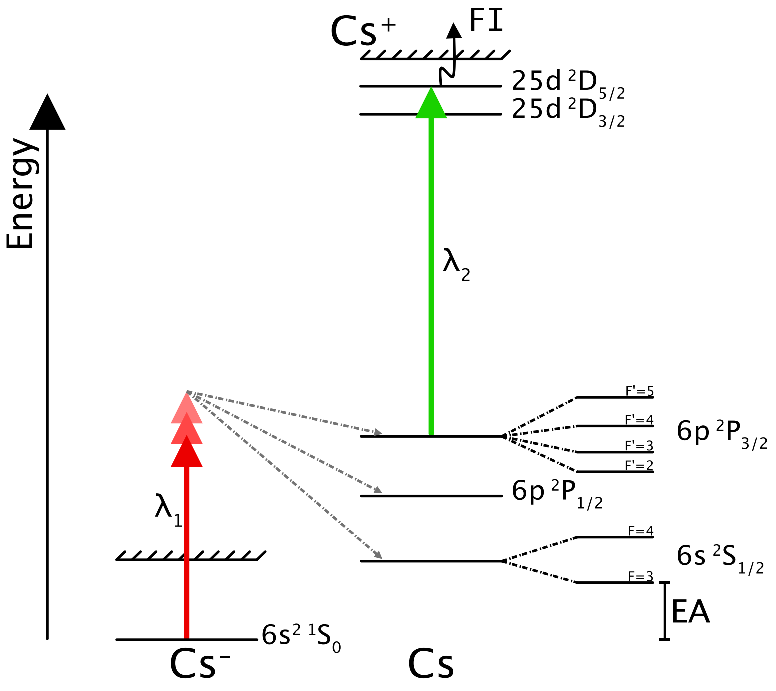
- For the most part, to extract atomic data from negative ions they must be destroyed in a process called photodetachment

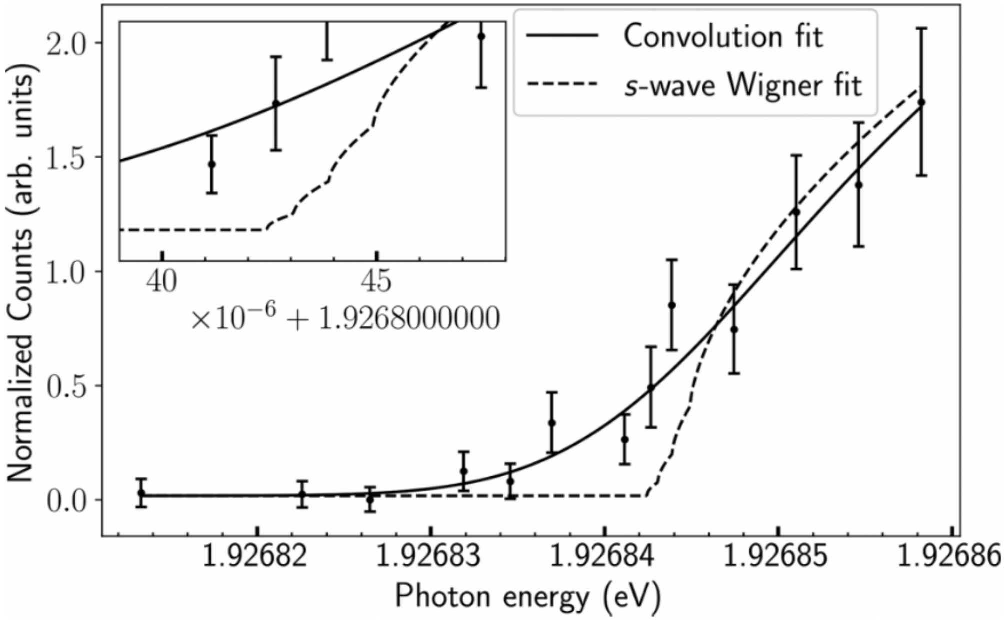


Experiments

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]





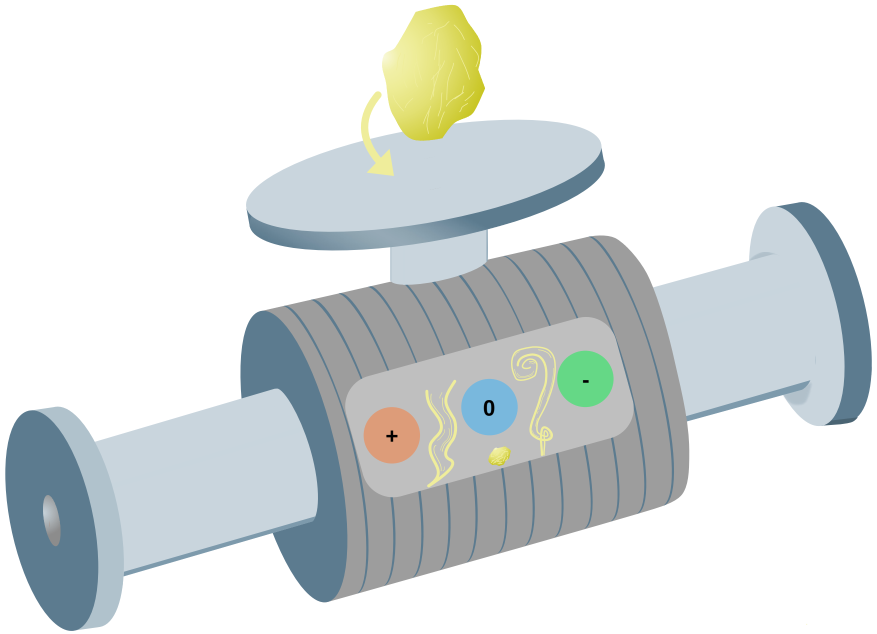
[1] Navarro-Navarrete *et al., PRA* **109** 022812 (2024)
[2] Ringvall-Moberg *et al., J. PHYS. B* **57** 155002 (2024)

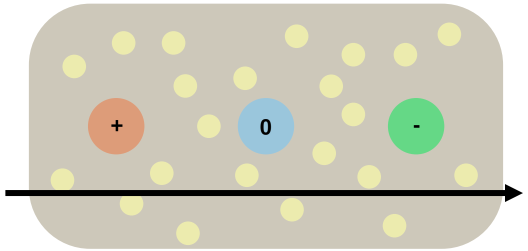
Negative ion production

- 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





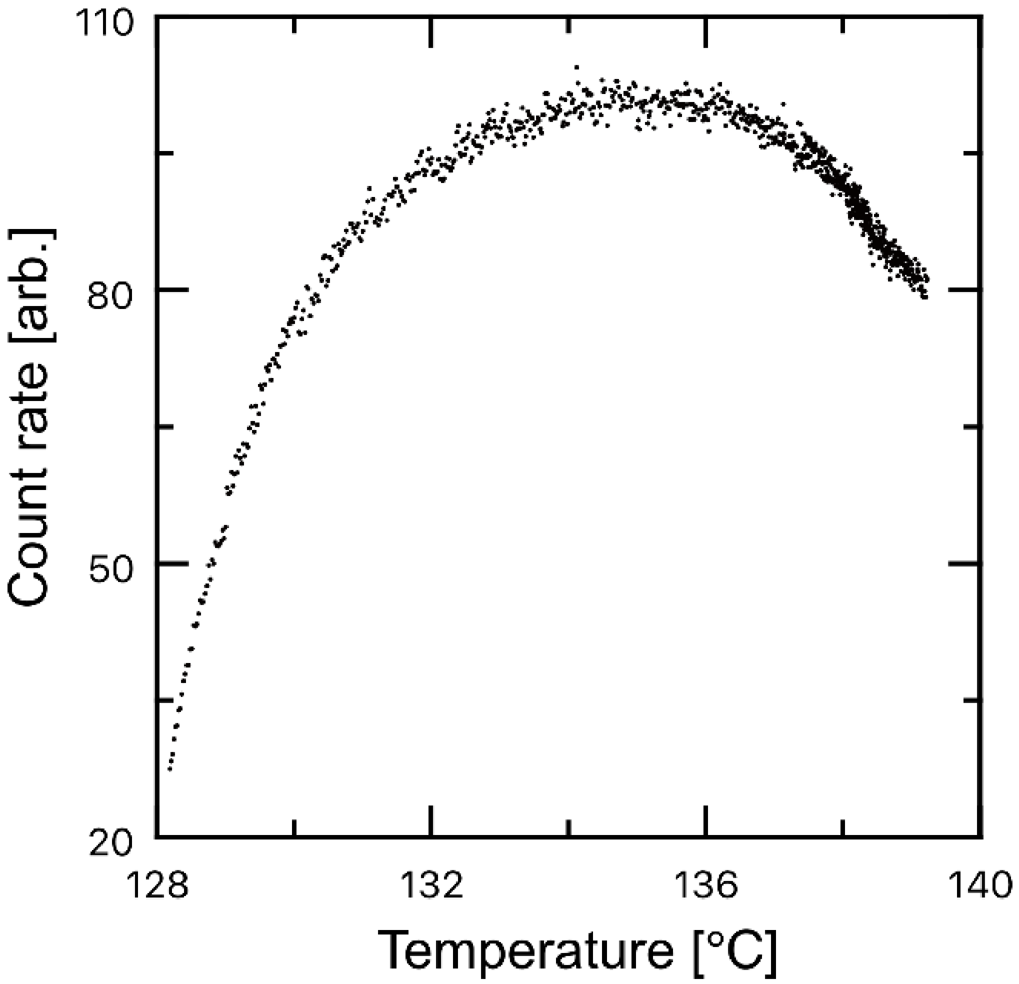
There are multiple reactions to consider:

$$U^+ + K \longrightarrow U + K^+ + \Delta E$$

$$U + K \longrightarrow U^- + K^+ + \Delta E$$

$$U^- + K \longrightarrow U + K + e^-$$

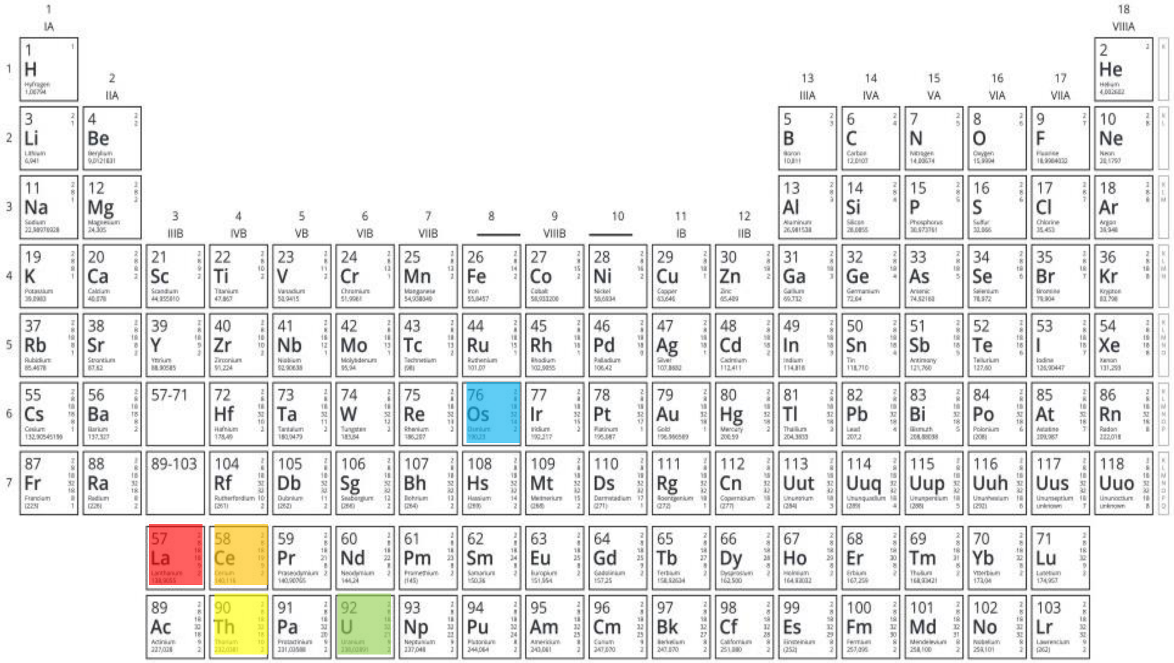
$^{238}U^{-}$ production



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



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

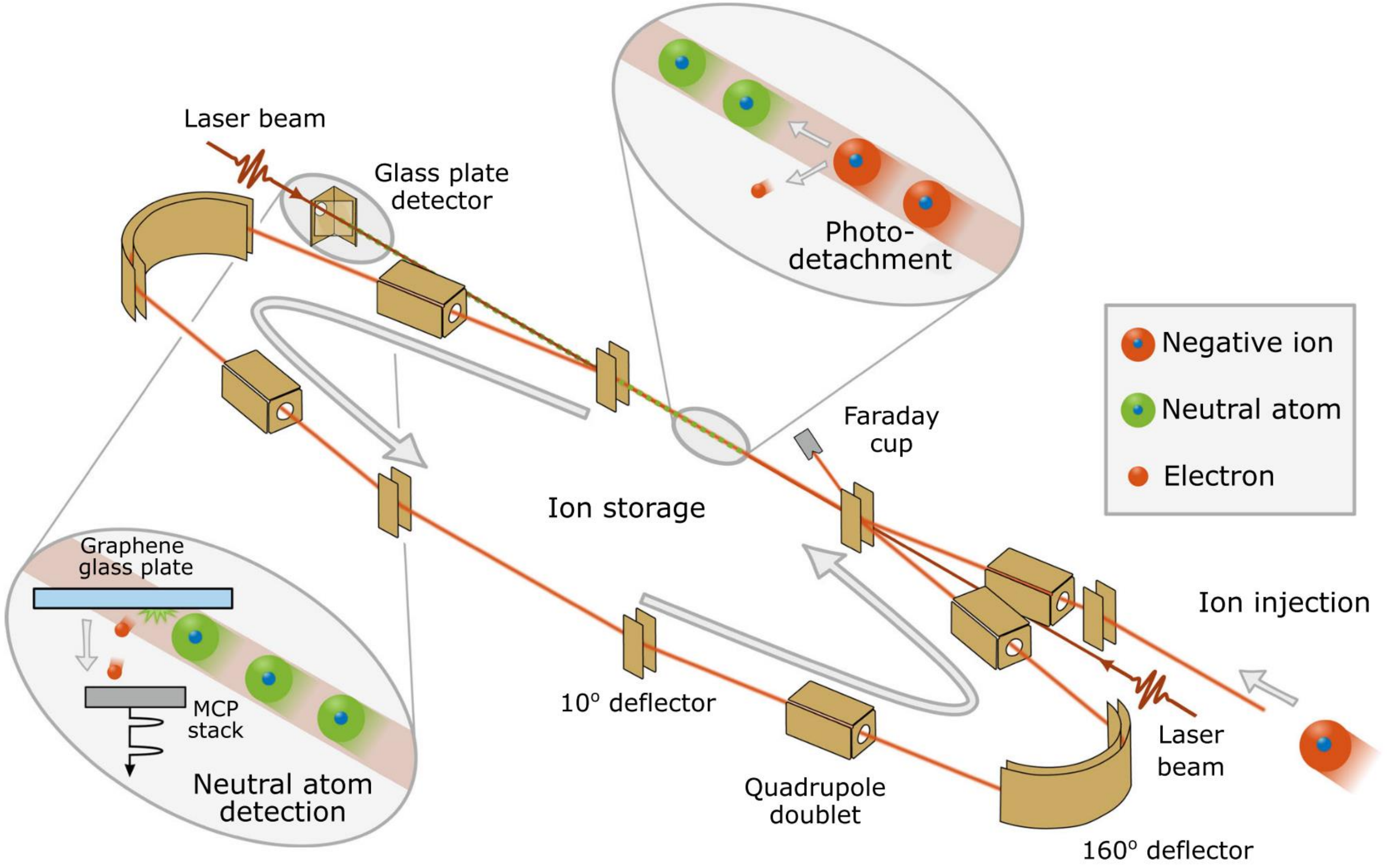


Image courtesy of M. K. Kristiansson *et al., Nat. Commun.* **13** 5906 (2022)

DESIREE: The Double ElectroStatic Ion Ring ExpEriment

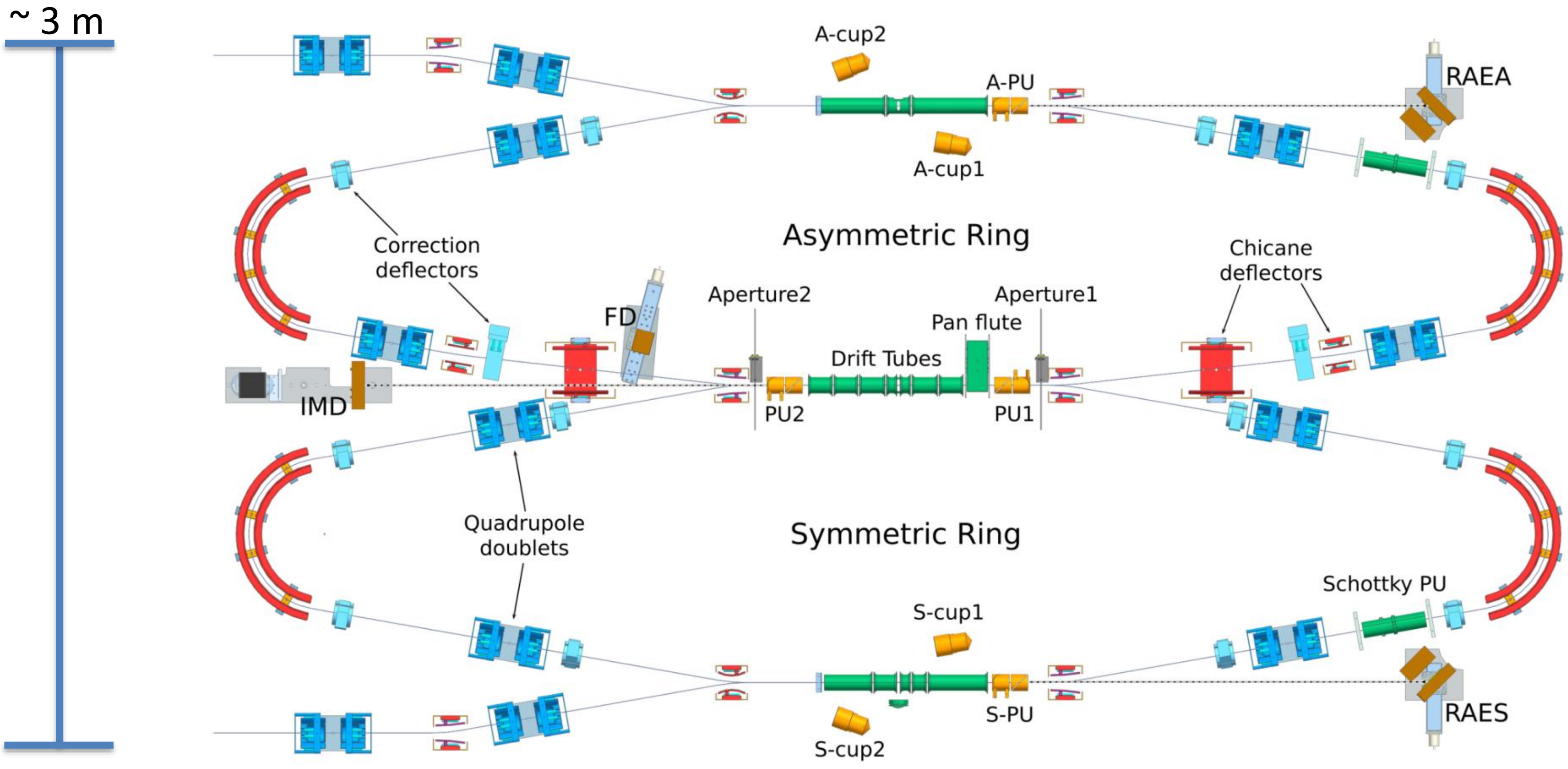
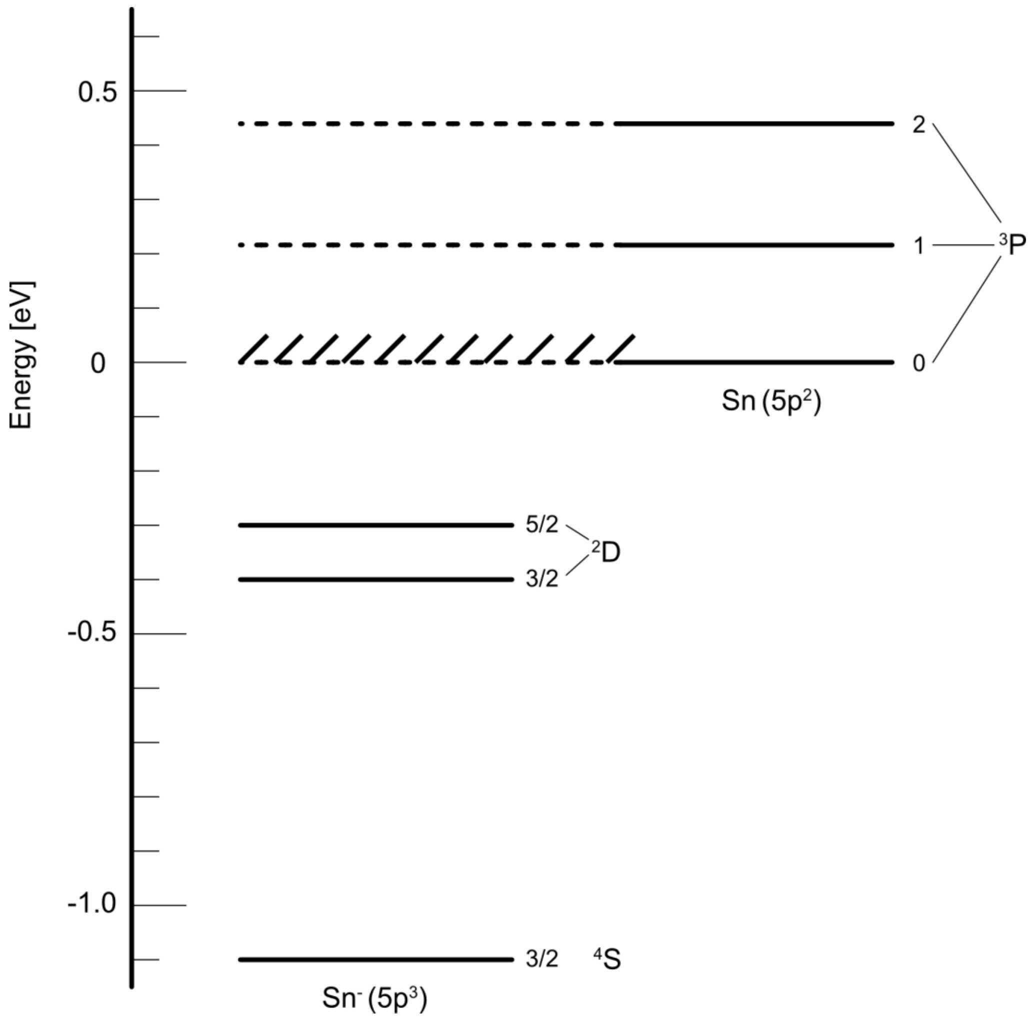


Image courtesy of https://www.desiree-infrastructure.com/experiments

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

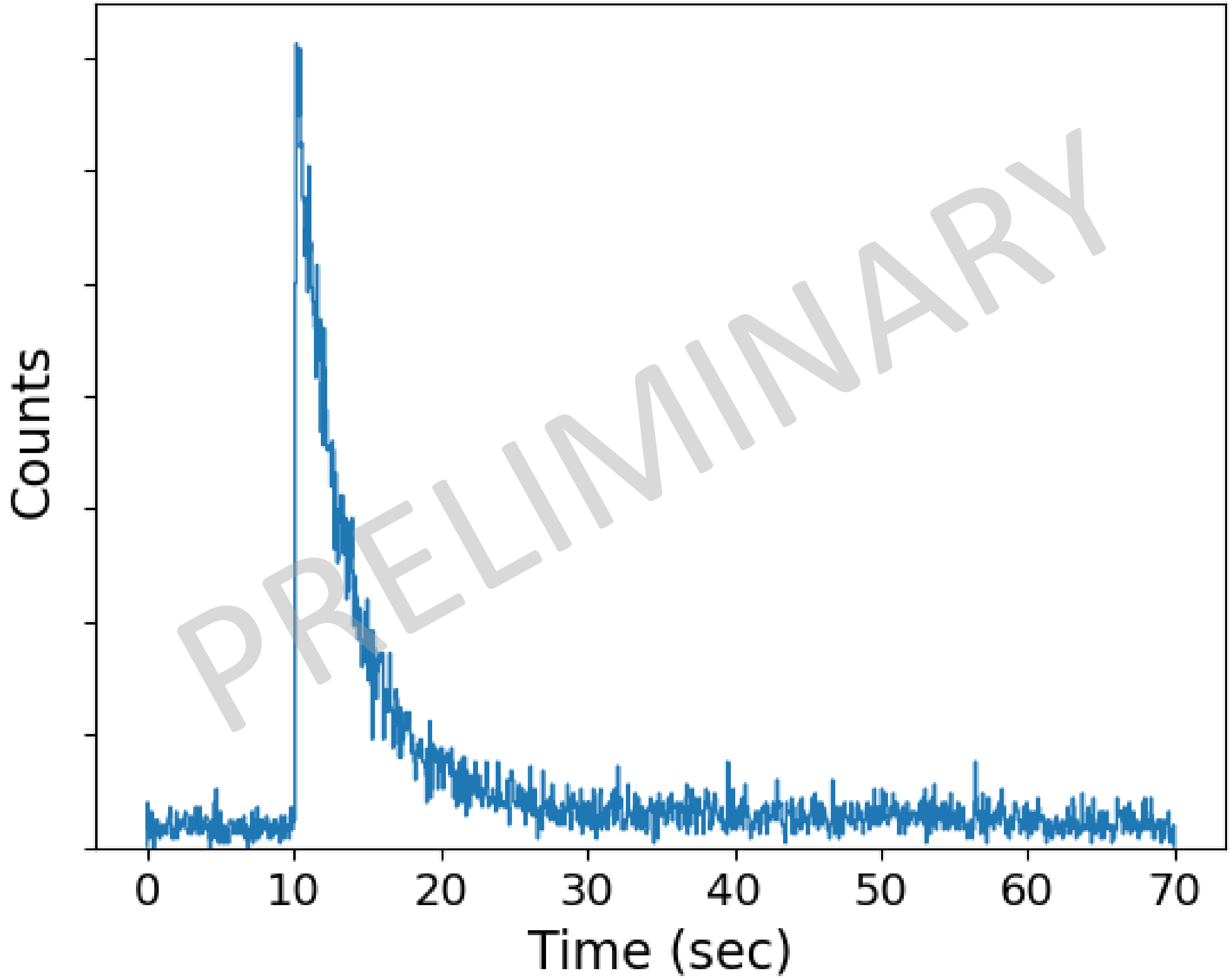
The structure of carbon group anions



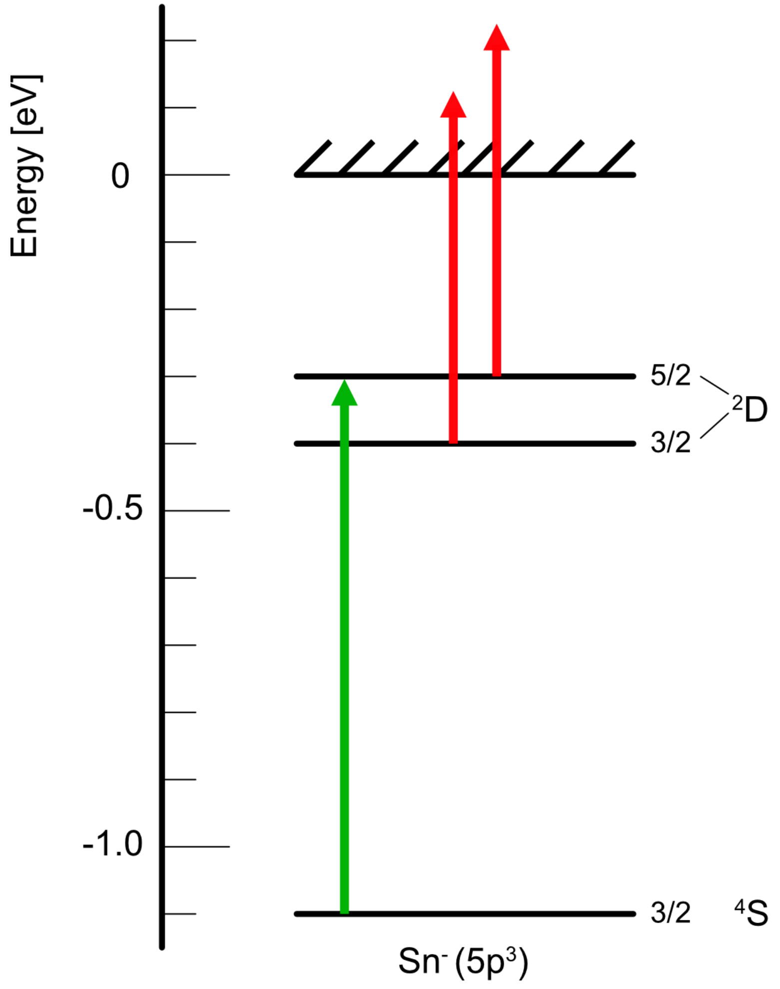
There's a ^2D_J state that decays mainly through *M1*!

Lifetimes of the bound Sn^- states

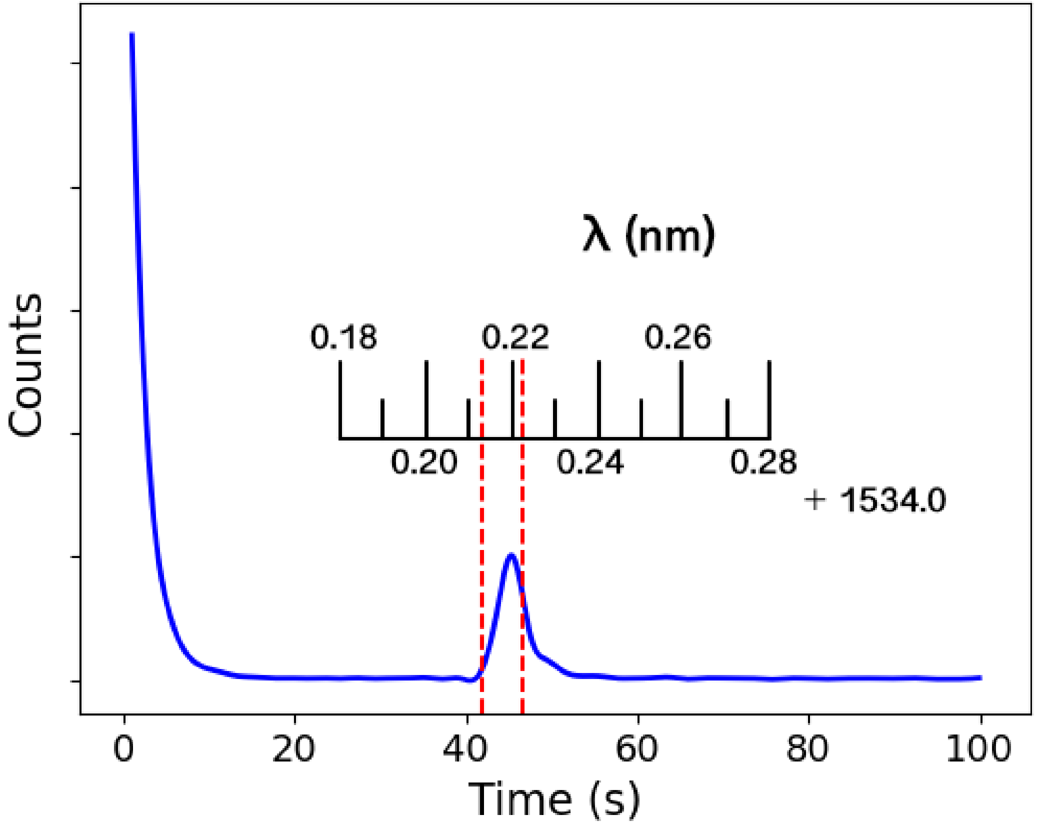
- Measure yield of neutrals
- Storage ~2950 s
- Lifetime of $D_{3/2}$ ~12s
- Lifetime of $D_{5/2}$ ~60s
- To distinguish states:
  - Deplete both states, then pump-probe a single state
  - Wait for shorter lived state to decay



Do wave functions behave as we expect?



Resonance in $^{120}Sn^-$

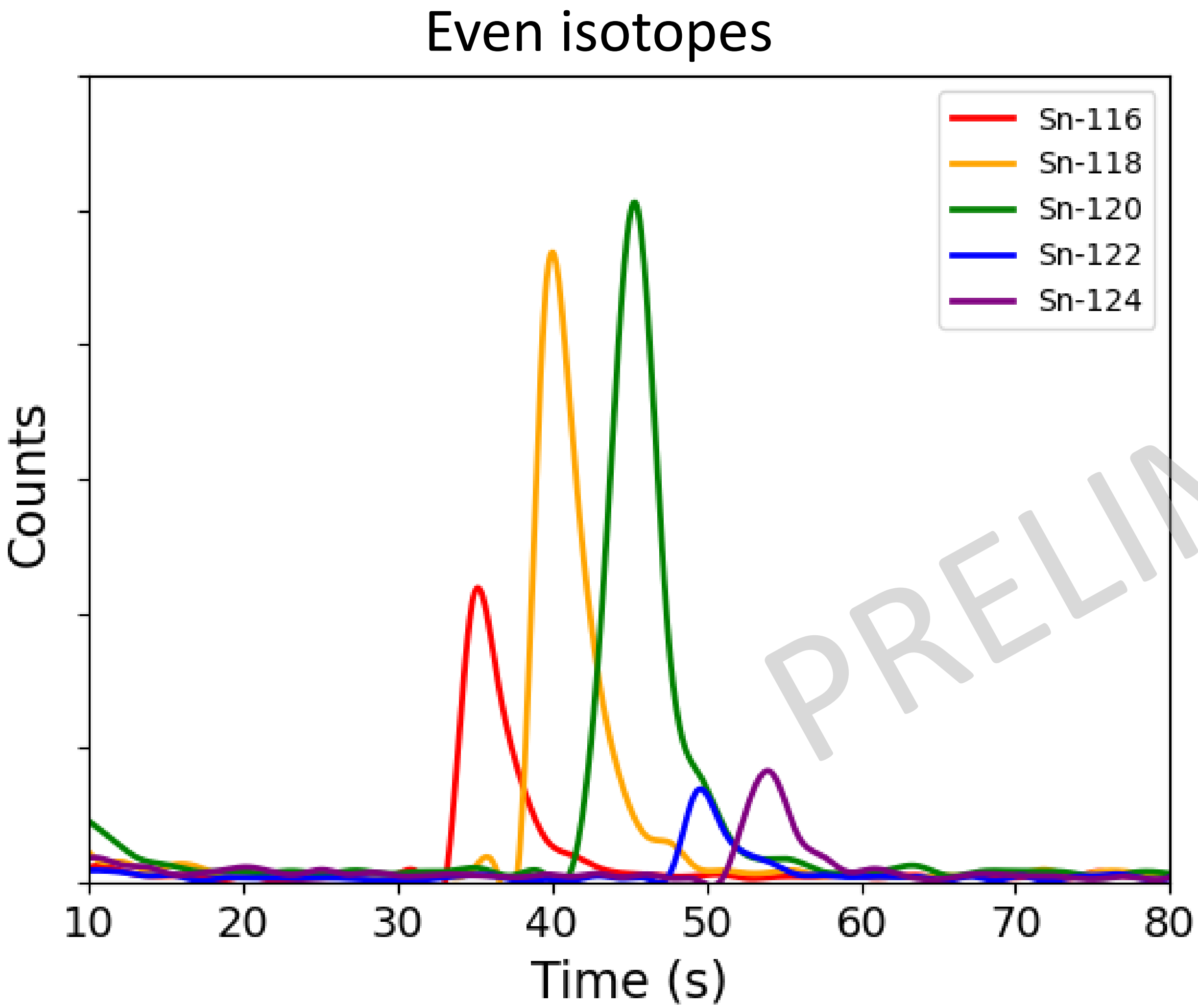


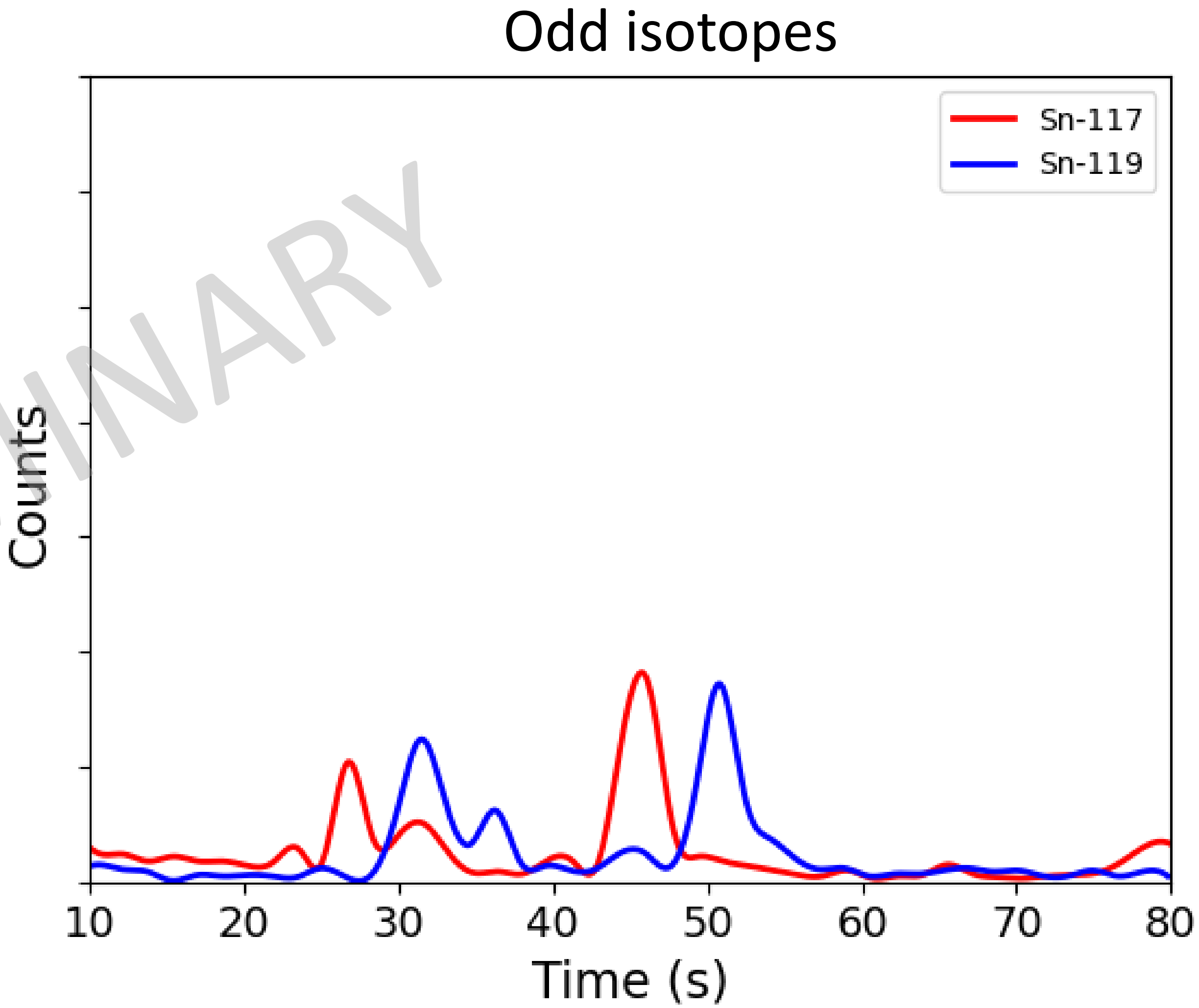
$^4S_{3/2} \rightarrow {}^2D_{5/2}$ *M1* resonance was first measured by Sheer *et al.*, *PRA* **58** 2844 (1998)

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

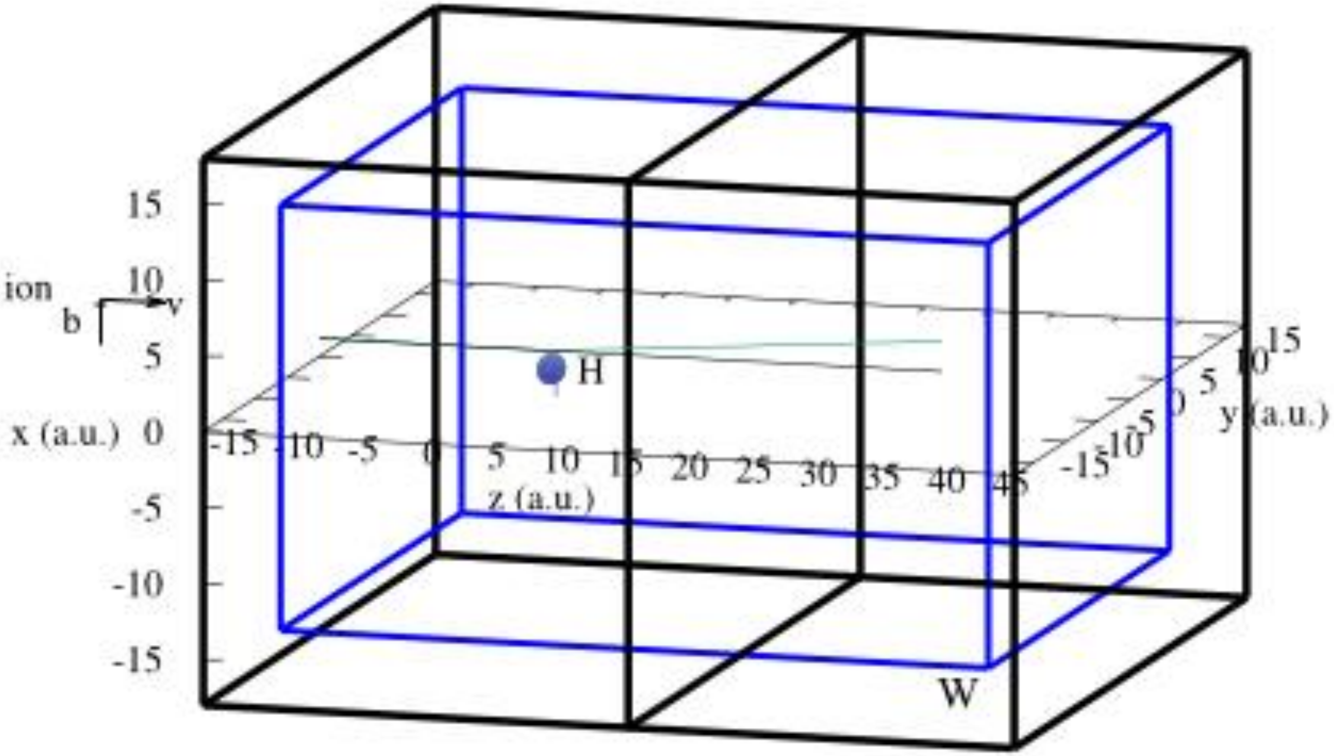




Theory

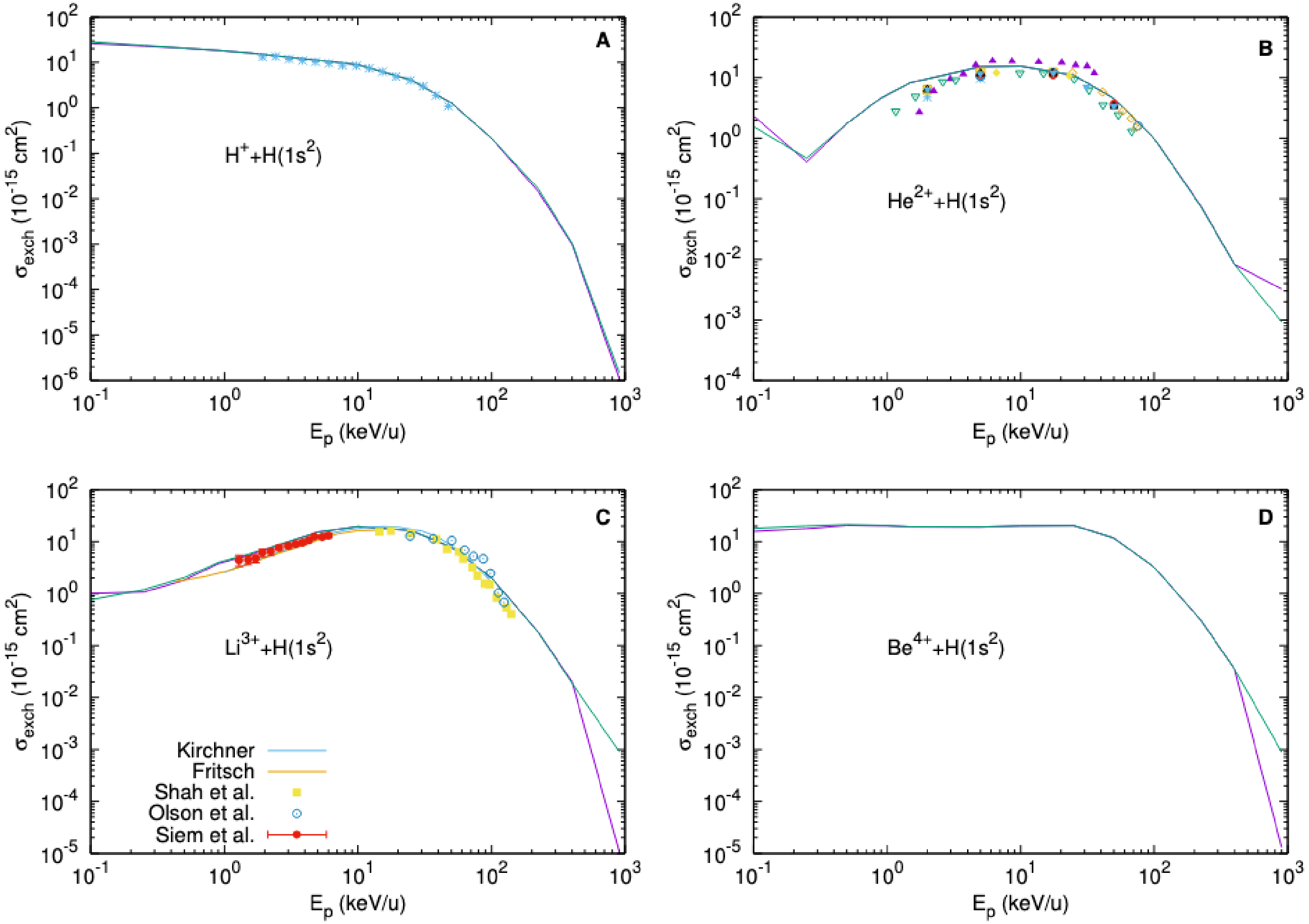
Collisions of bare ion projectiles, Z = 1 – 4

$A^q + H$



- 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



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

Acknowledgments

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M.Nichols[1], M. Athanasakis-Kaklamanakis[2,3], A. Borschevsky[5], T. E. Cocolios[3], R. Crosa-Rossa[5], R. P. de Groote[3], K. T. Flanagan[4], R. F. Garcia Ruiz[6], S. Geldhof[3], D. Hanstorp[1], Á. Koszorús[2], L. Lalanne[3], D. Leimbach[1], G. Neyens[3], J. Reilly[4], S. Rothe[2], S. G. Wilkins[6], X. F. Yang[9], and the ISOLDE technical staff **for the work on U^- production**

Annie Ringvall-Moberg[1], José E Navarro Navarrete[2], Uldis Bērziņš[3], Viola C D'mello[1], Julia Karls[1], Di Lu[1], Yazareth Peña Rodríguez[1,4], Rachel Poulose[1,5], Andrea Morales Rodríguez[1,4], Keerthana Ravi[1,5], Meera Ramachandran[1,5], Vitali Zhaunerchyk[1], Dag Hanstorp[1] and David Leimbach[1,6] **for the work on Rb**

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