Comparing Coulomb explosion dynamics of multiply charged OCS after ionization by soft X-rays and few cycle femtosecond laser pulses

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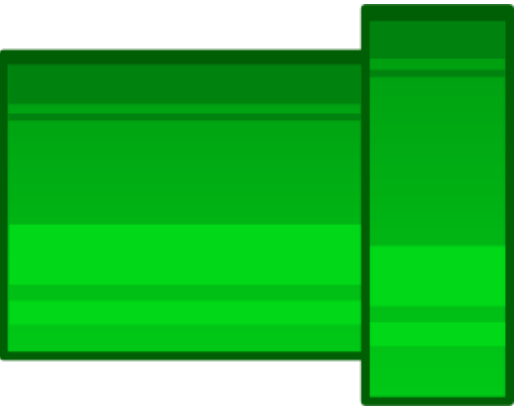
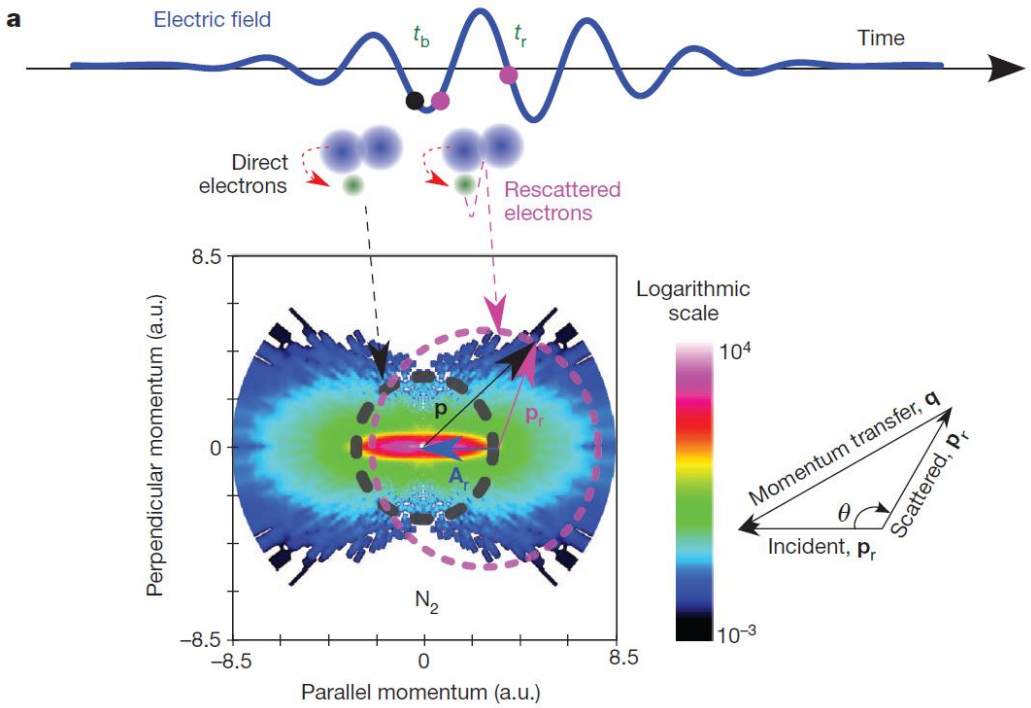


Time-resolved molecular imaging: **Laser vs. Synchrotron**

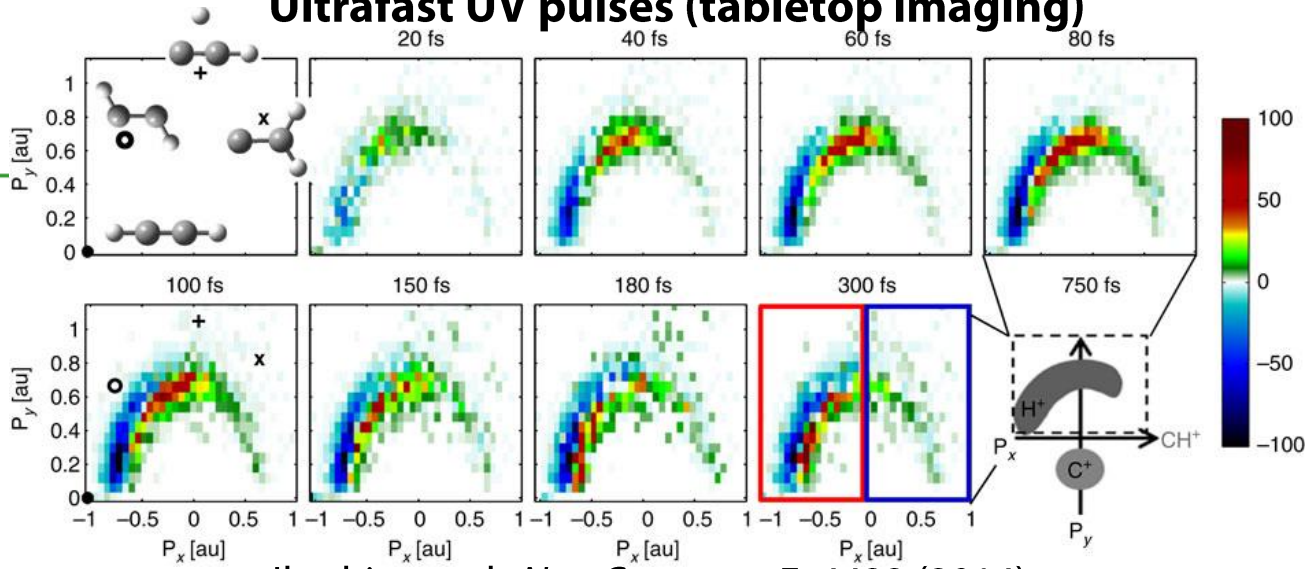
Introduction and motivation

Laser-induced electron diffraction (LIED)



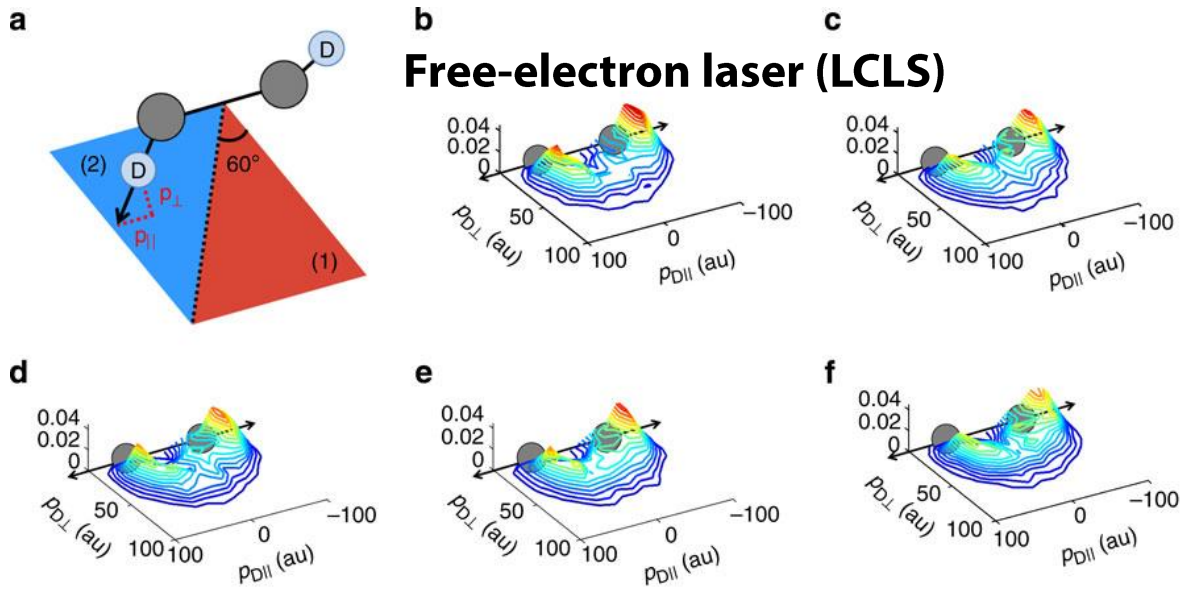
- Blaga et al., *Nature* **483**, 194 (2012)

Ultrafast UV pulses (tabletop imaging)



- Ibrahim et al., *Nat. Commun.* **5**, 4422 (2014)

Free-electron laser (LCLS)



- Liekhus-Schmaltz et al., *Nat. Commun.* **6**, 8199 (2015)

Experimental

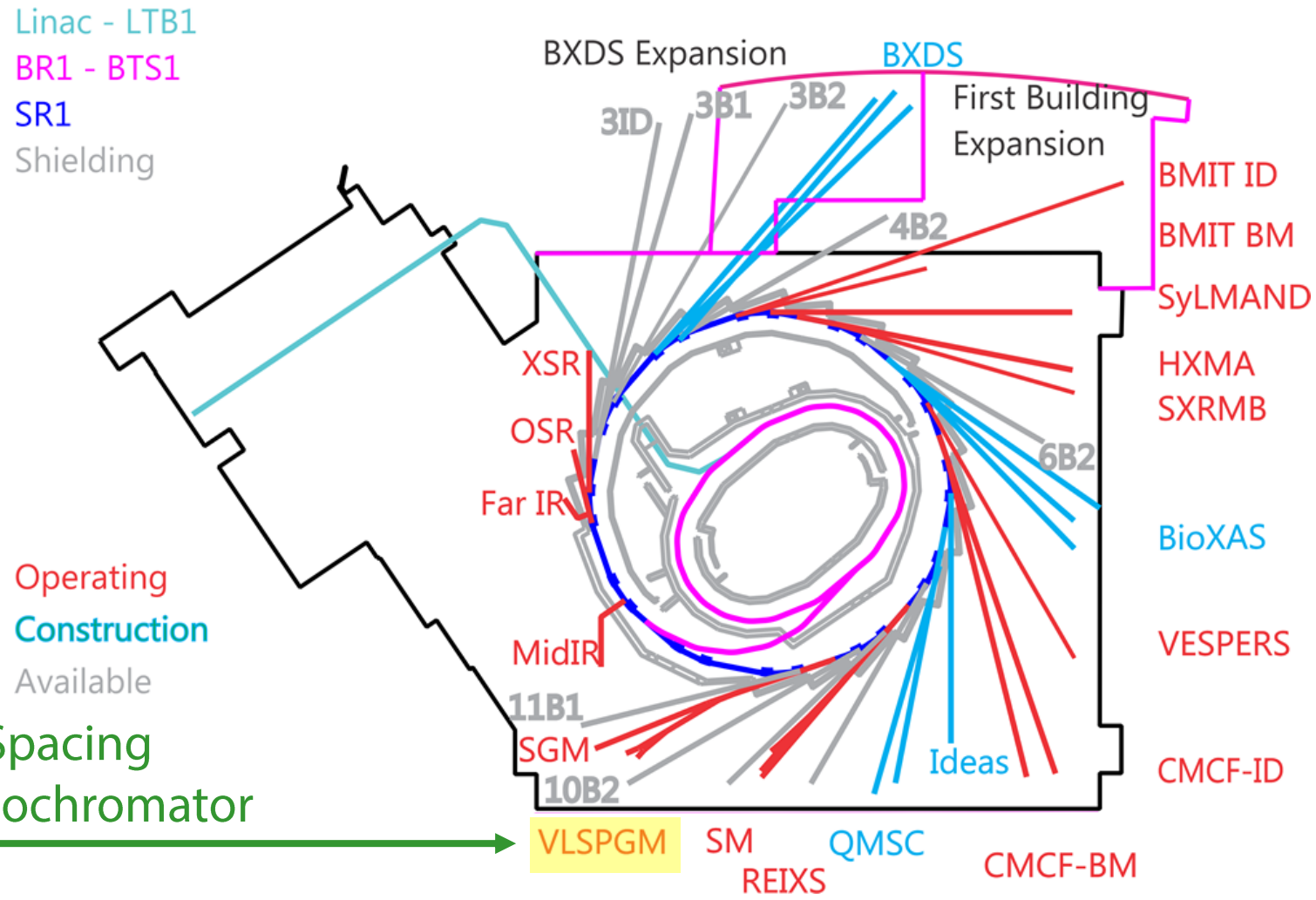
Apparently I had a strong sense of proper experimental practices but was still a bit short on skepticism...

Emily Levesque

Experimental: Canadian Light Source synchrotron

- Source: Google Maps

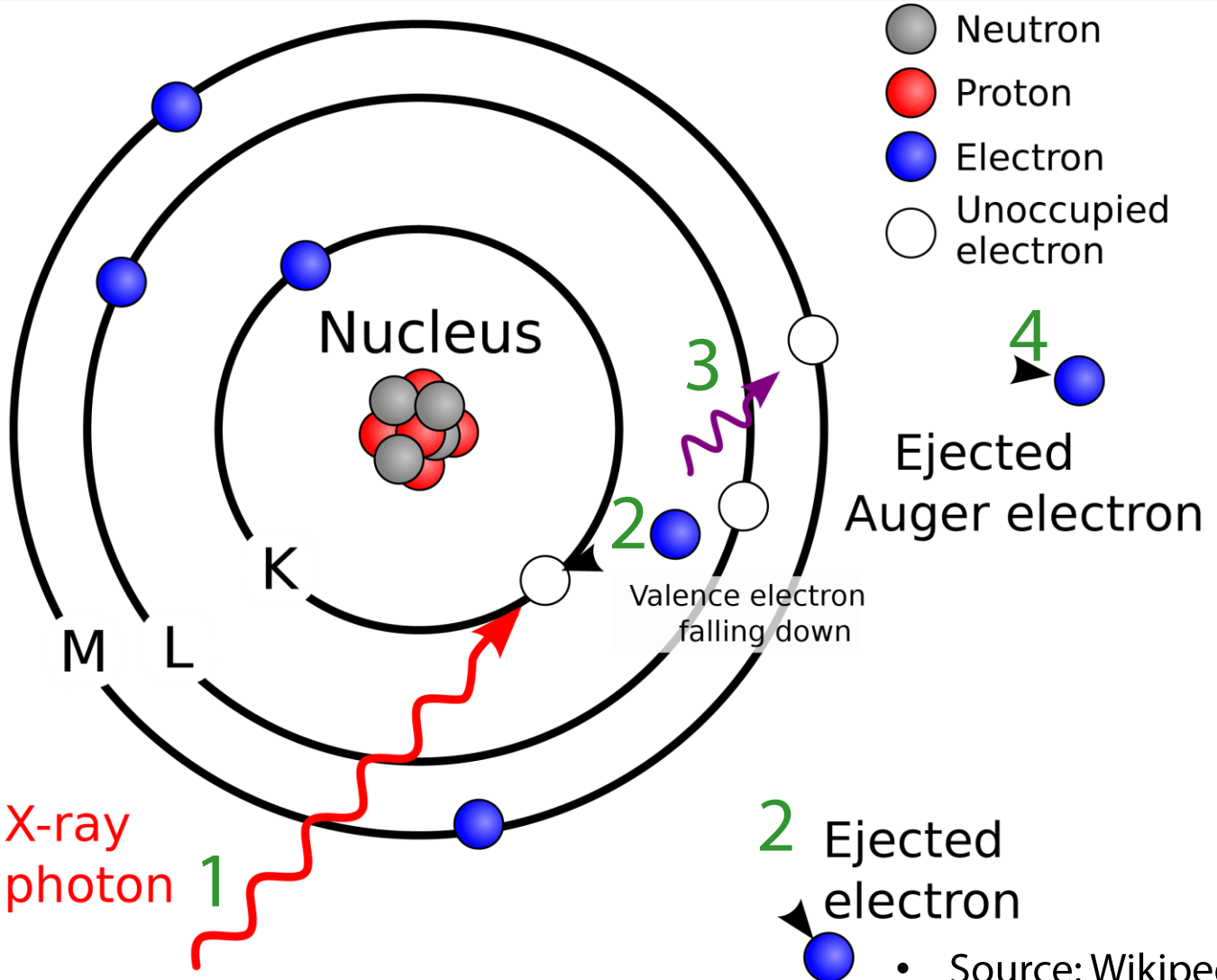
Experimental: Canadian Light Source synchrotron



Variable Line Spacing Plane-Grating Monochromator

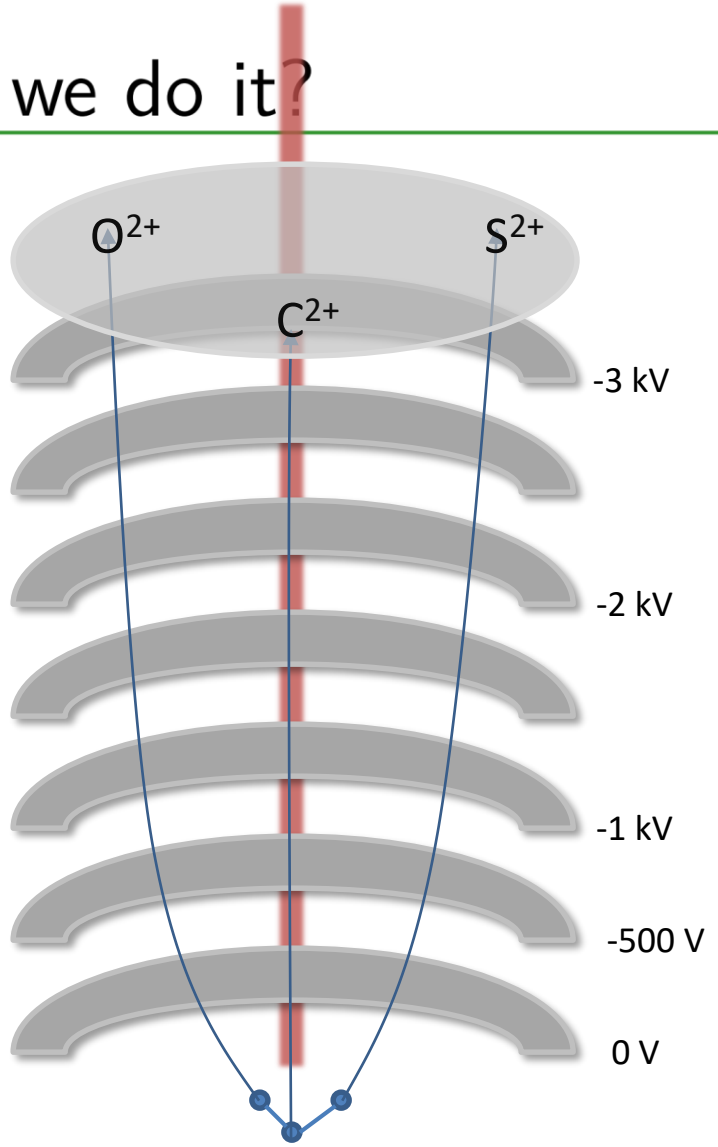
- Source: Canadian Light Source Inc.

Experimental: Auger effect

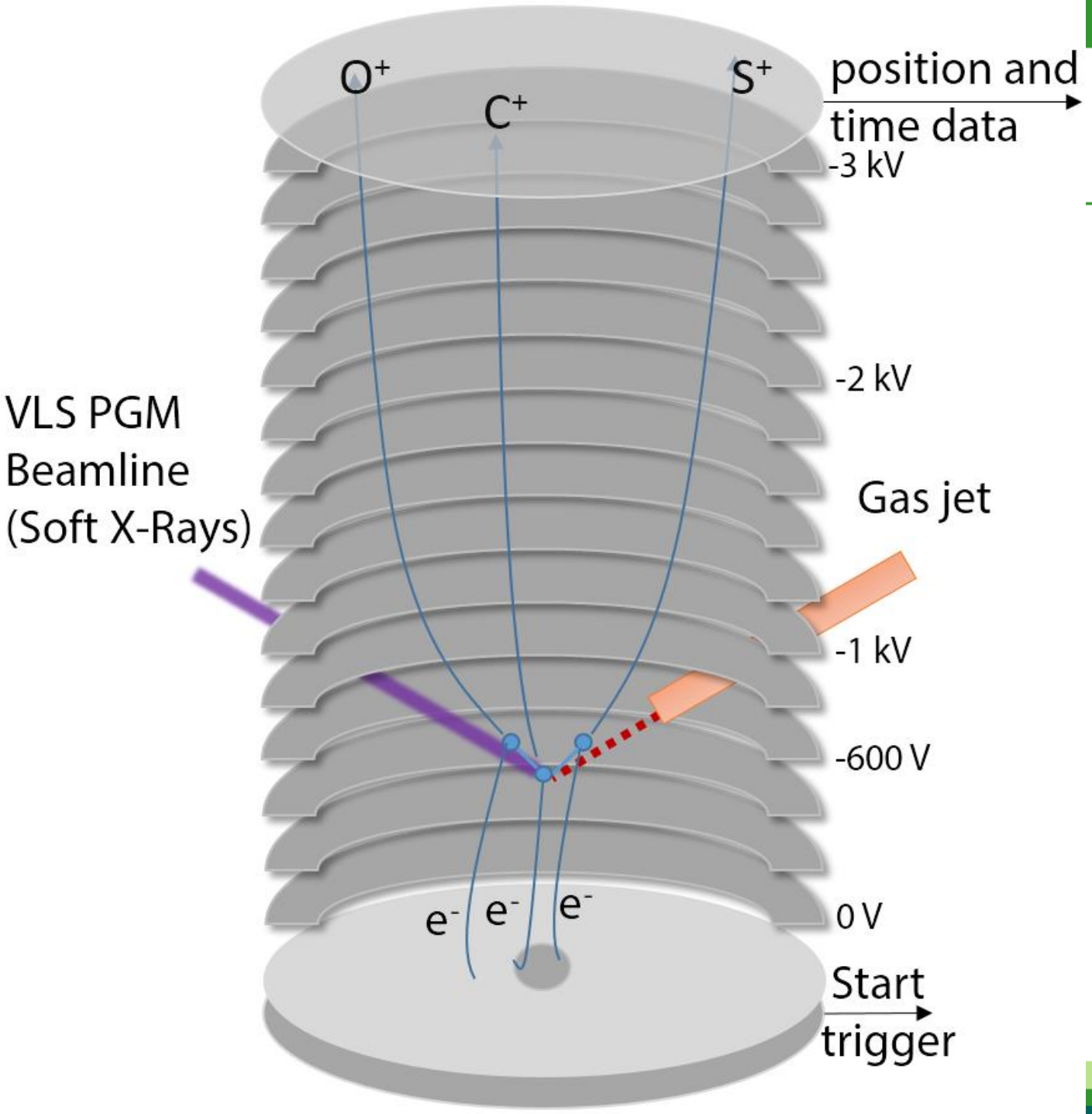


- Source: Wikipedia (modified)

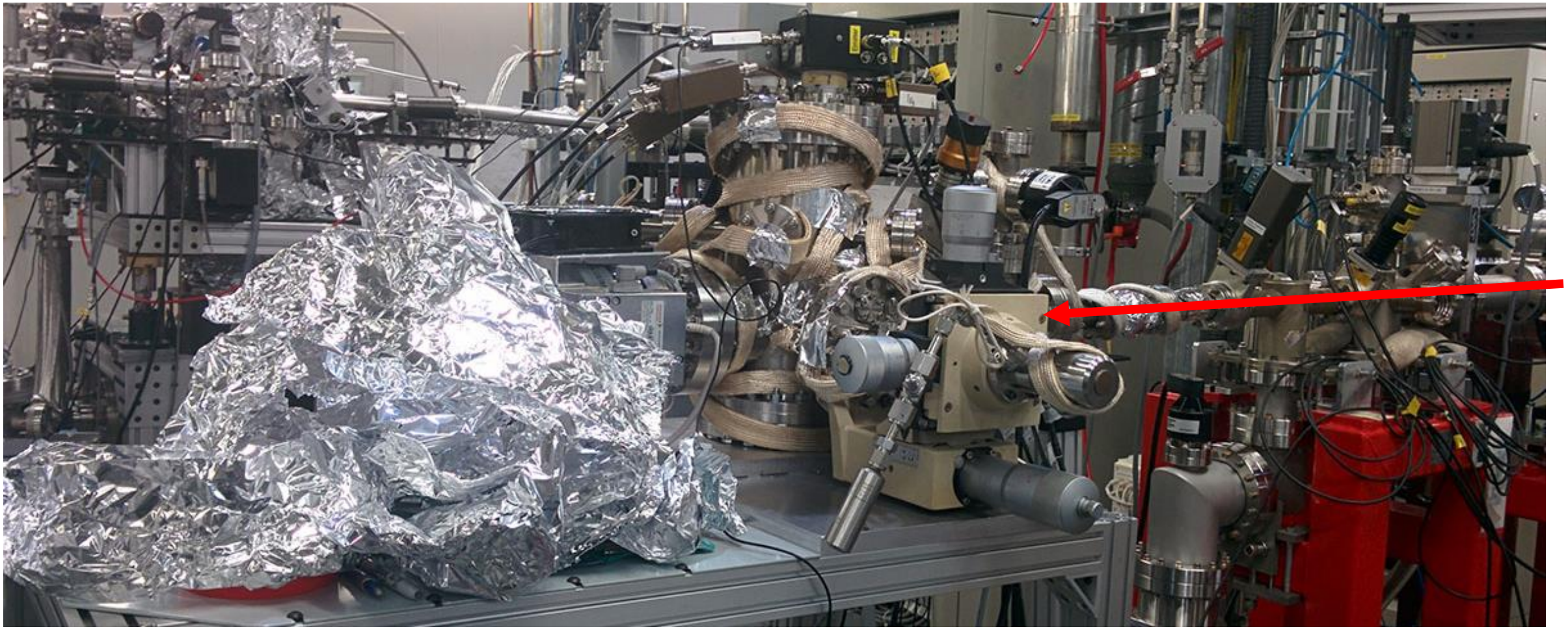
Experimental: How did we do it?



Experimental



The proof of the pudding is in the photos.



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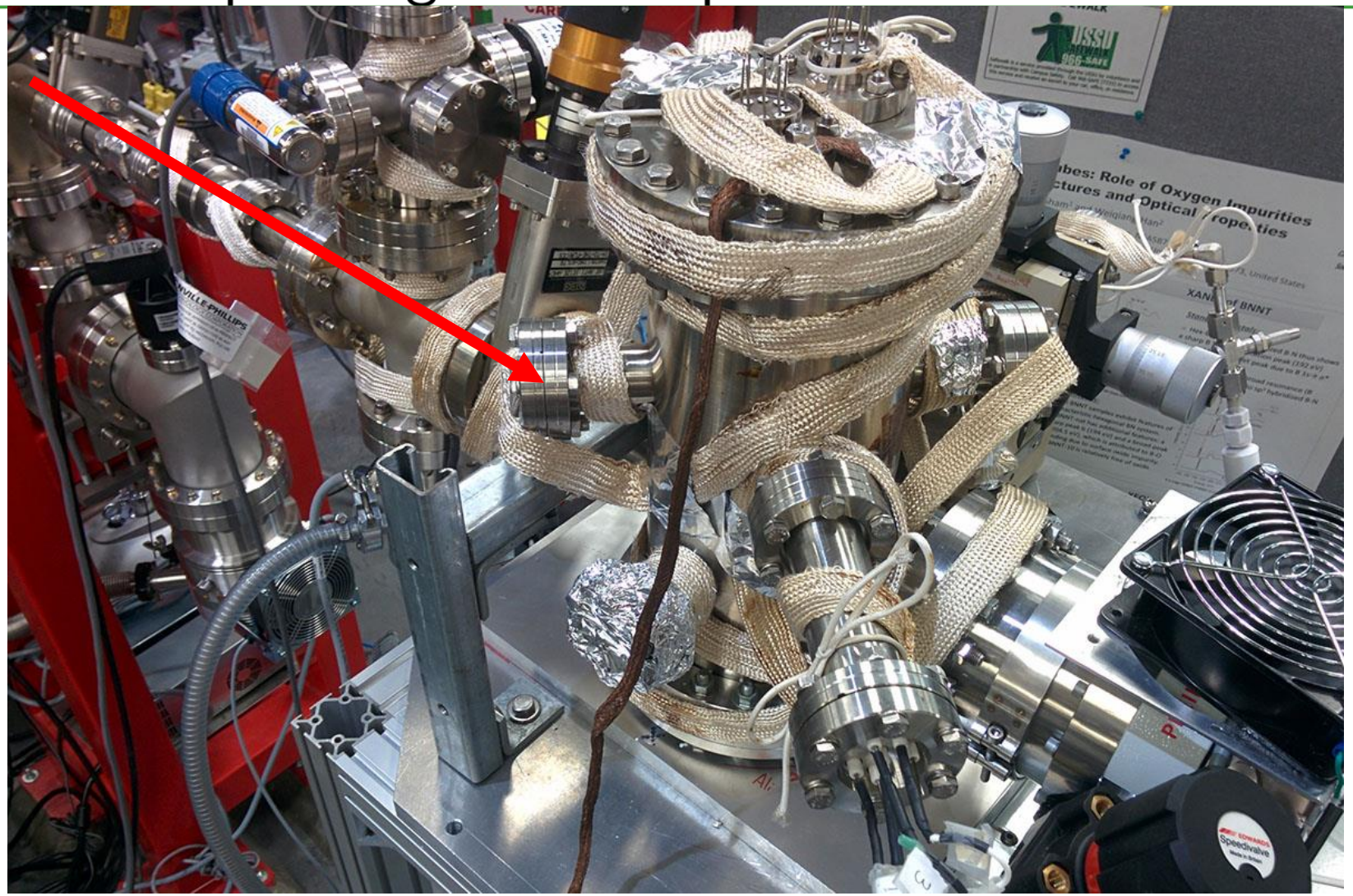
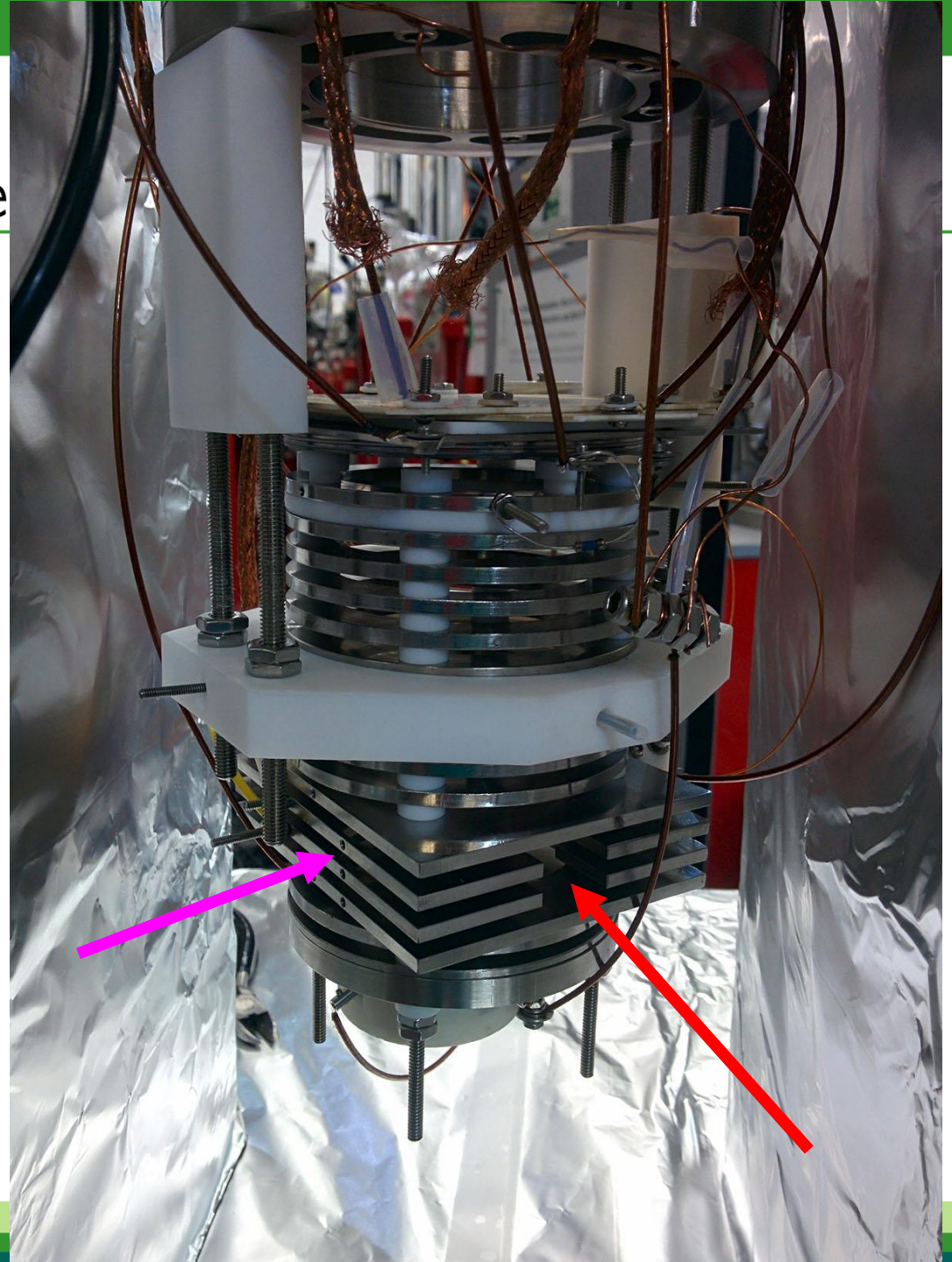


The proof of the



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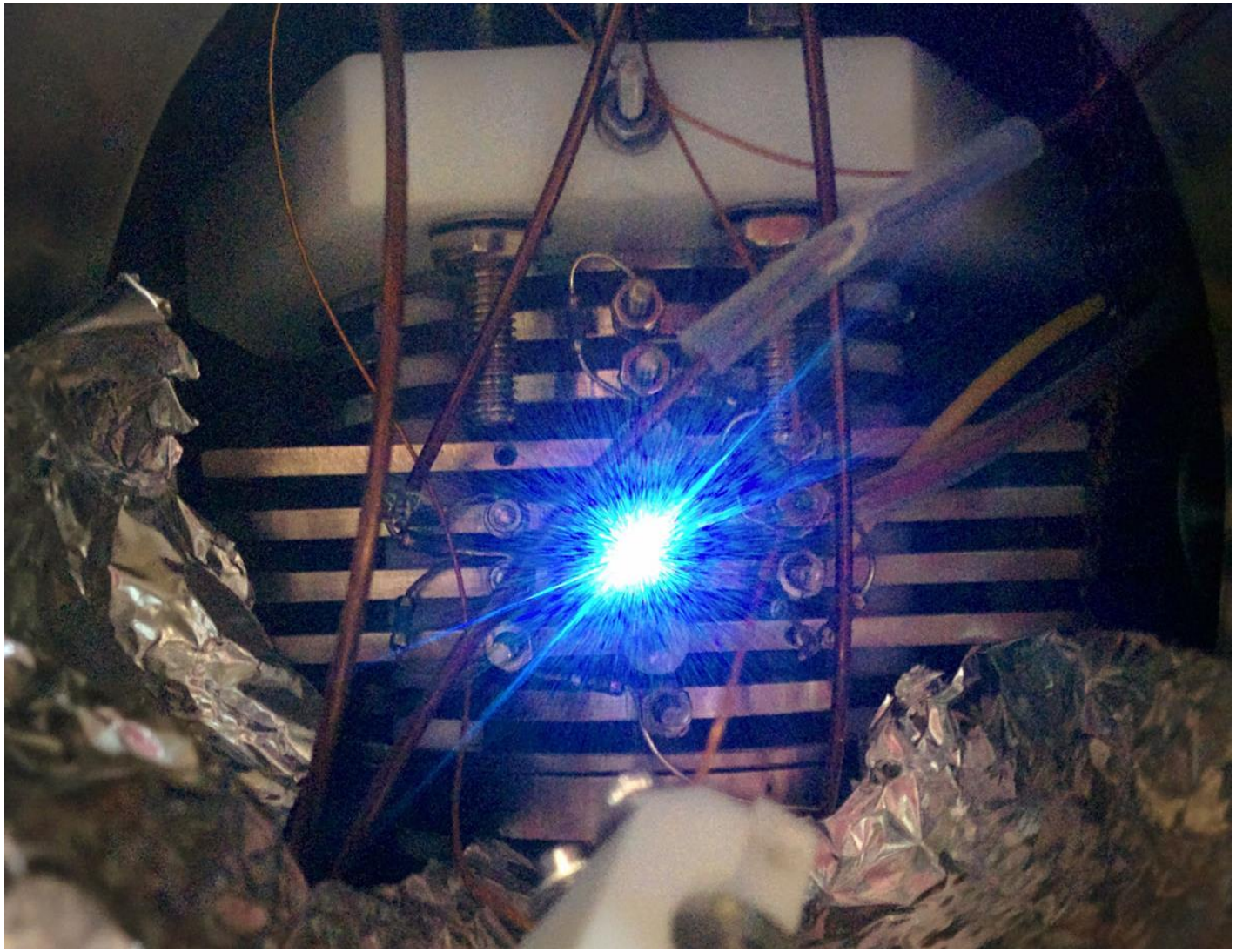
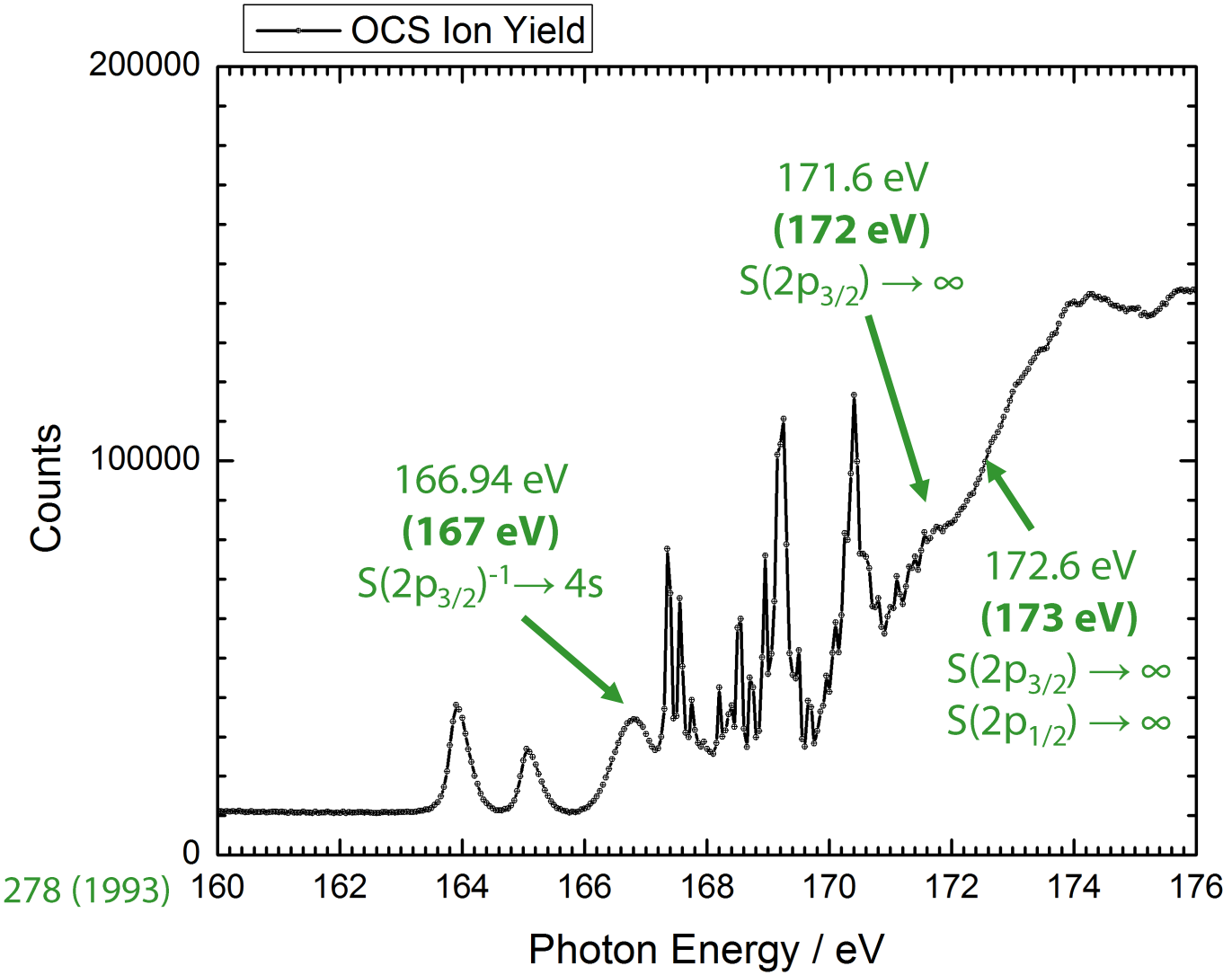


Results

It's not the result, but the journey.

Drake

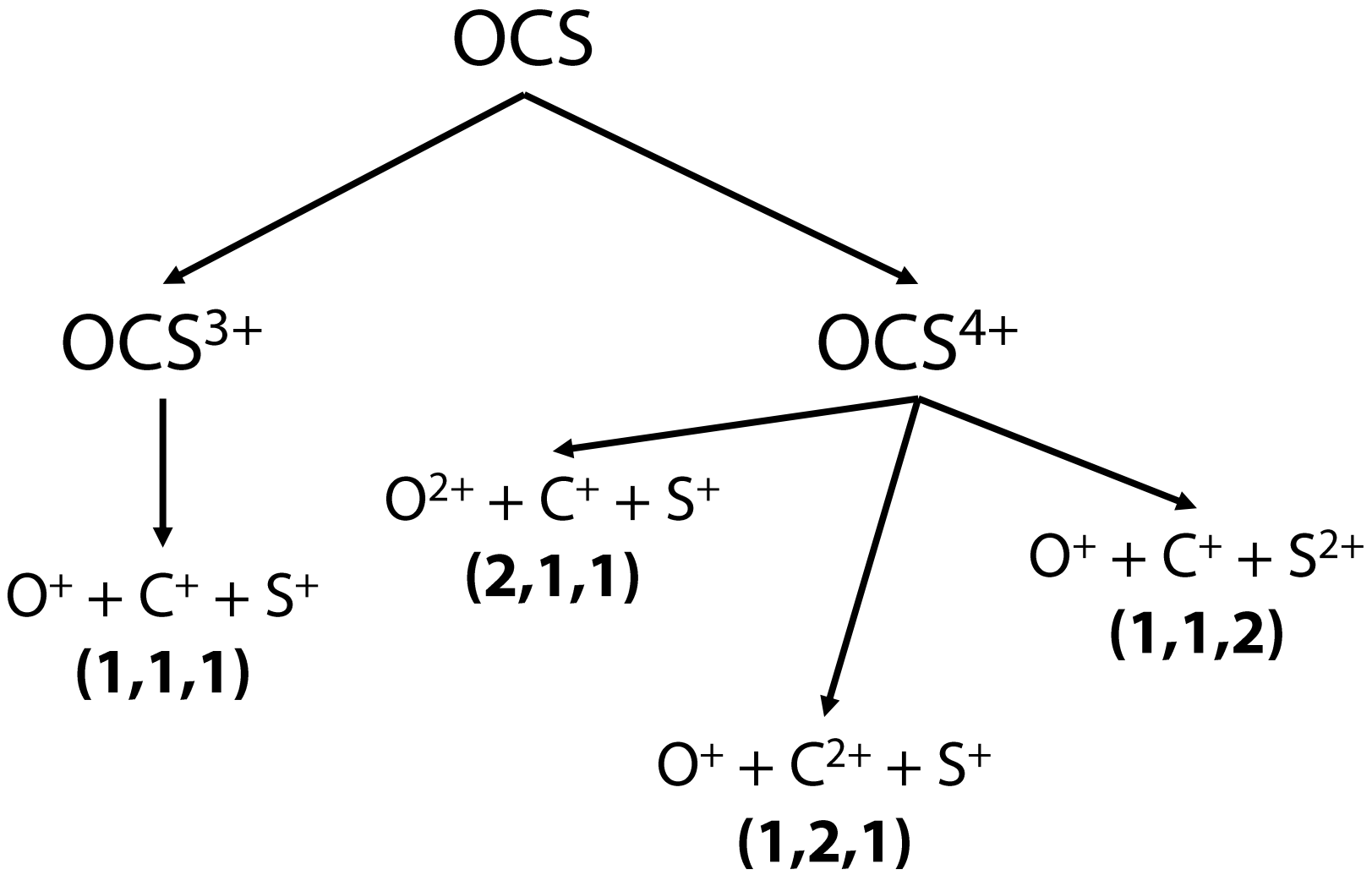
Photon energies used



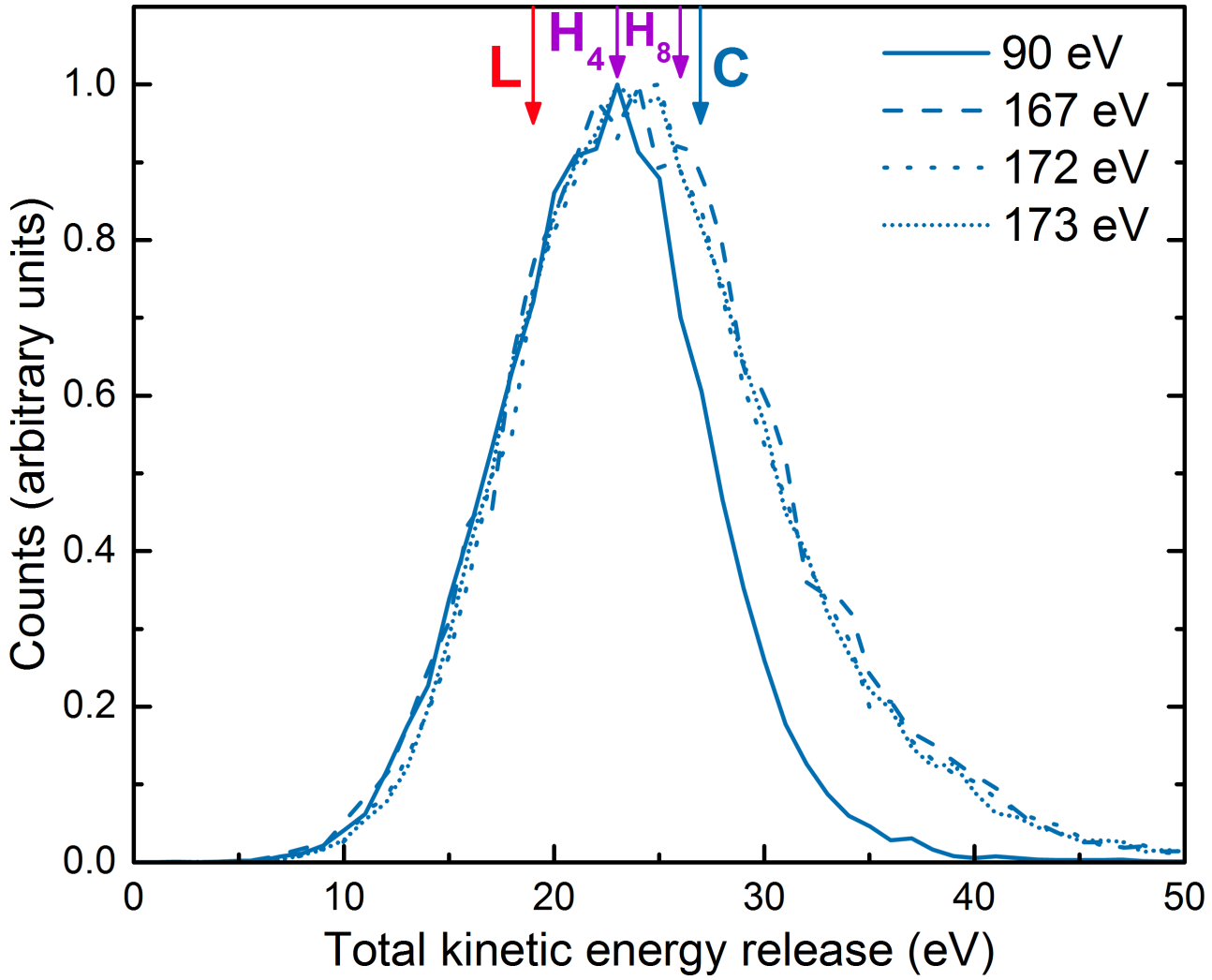
(90 eV)
Ground state at maximum ionization efficiency[1]

[1]Masuoka et al., *Phys. Rev. A* **47**, 278 (1993)

Fragmentation channels

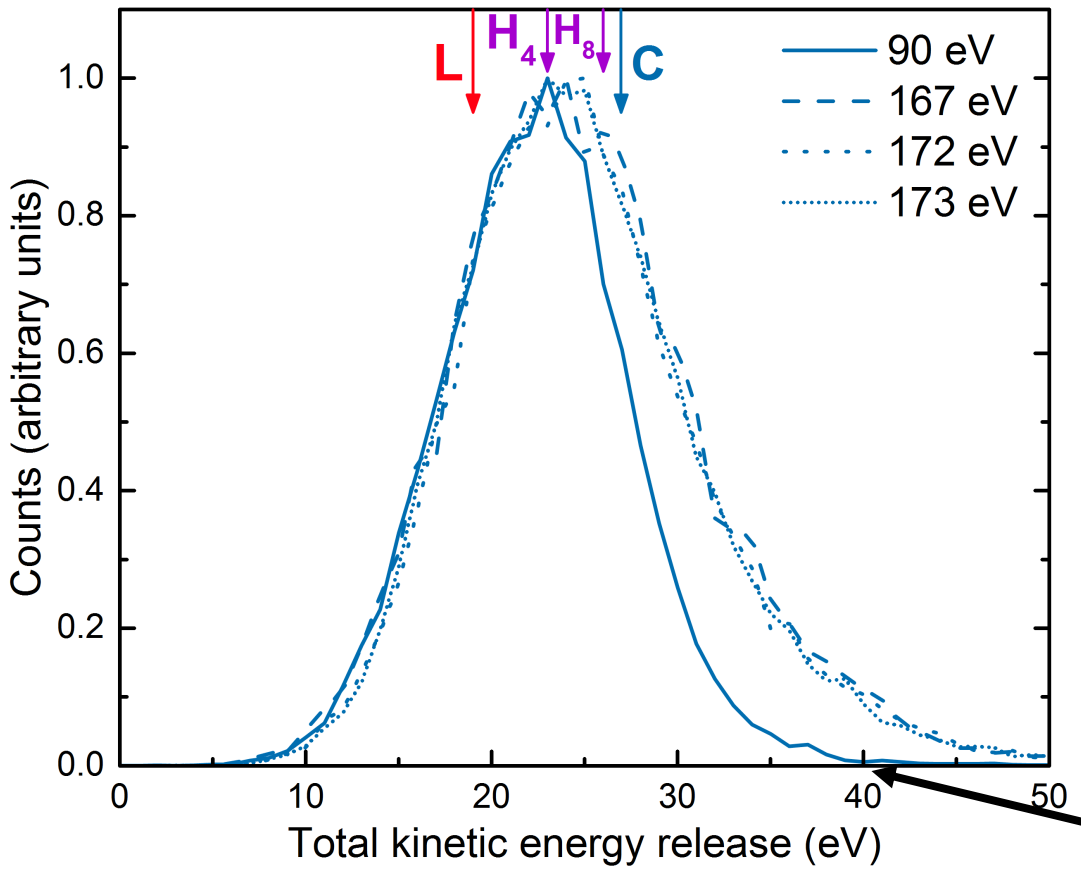


Results: kinetic energy release limited by photon energy



$$E = \frac{\mathbf{p}^2}{2m}$$

Results: kinetic energy release limited by photon energy



S—CO → S + CO	3.120 eV
C—O → C + O	11.162 eV
O → O+	13.618 eV
C → C+	11.260 eV
S → S+	10.360 eV

- Hohm et al., *J. Chem. Phys.* **101**, 6362 (1994)
- Sivakumar et al., *J. Chem. Phys.* **88**, 3692 (1988)

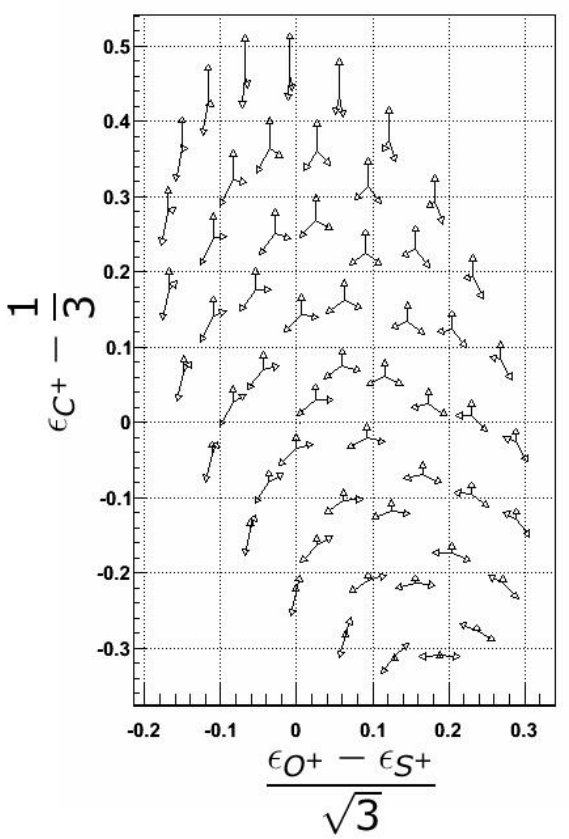
90 – 3.120 – 11.162 – 13.618 – 11.260 – 10.360 = 40.48 eV

Dalitz plots

- To categorize the possible break up channels we use the Dalitz plot method which lets us display the possible molecular dissociation geometries in a two dimensional histogram[1].

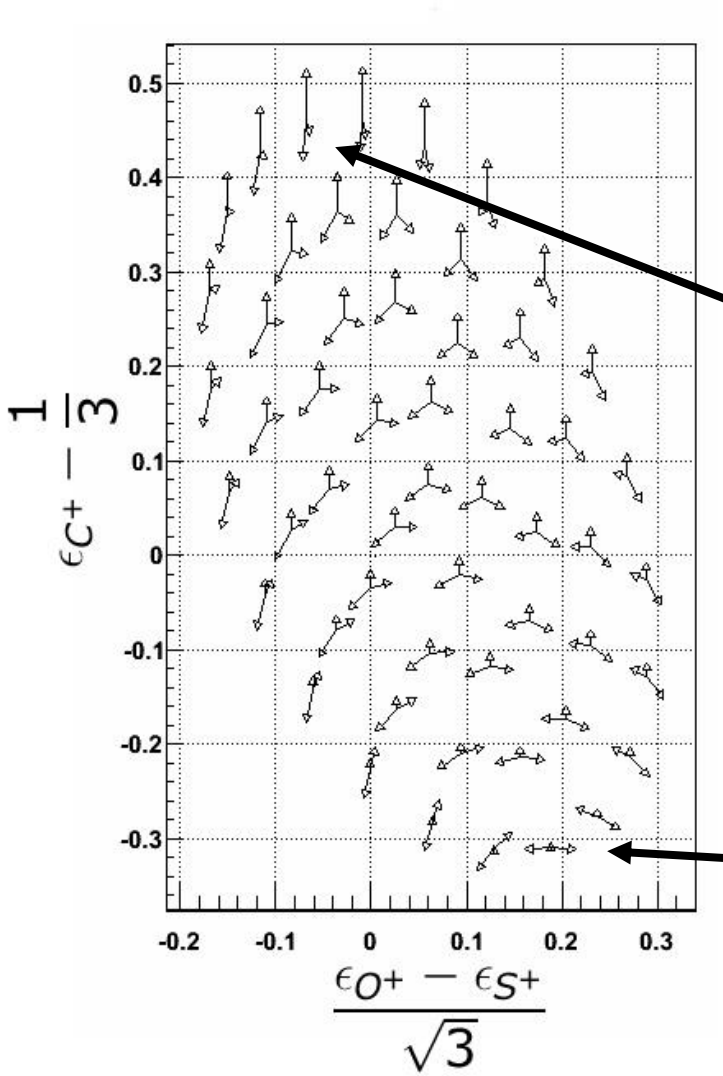
$$\epsilon_{O^+} = \frac{O^+ \text{ kinetic energy}}{\text{Total kinetic energy}}$$

$$\epsilon_{O^+} + \epsilon_{C^+} + \epsilon_{S^+} = 1$$



[1] R.H. Dalitz, *Phil. Mag.* **44**, 1068 (1953)

Dalitz plots



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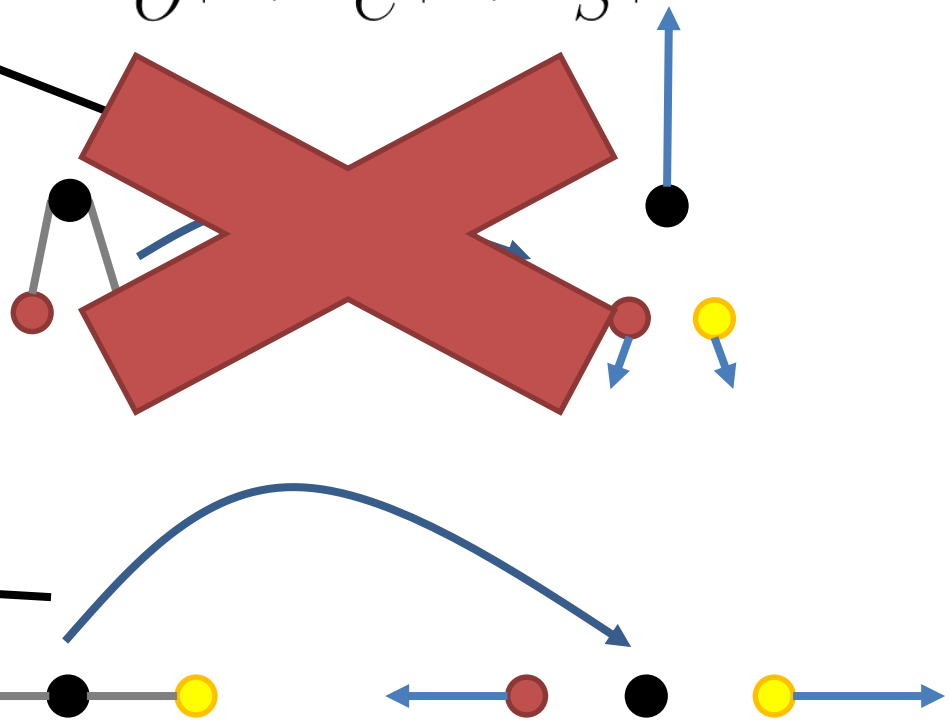
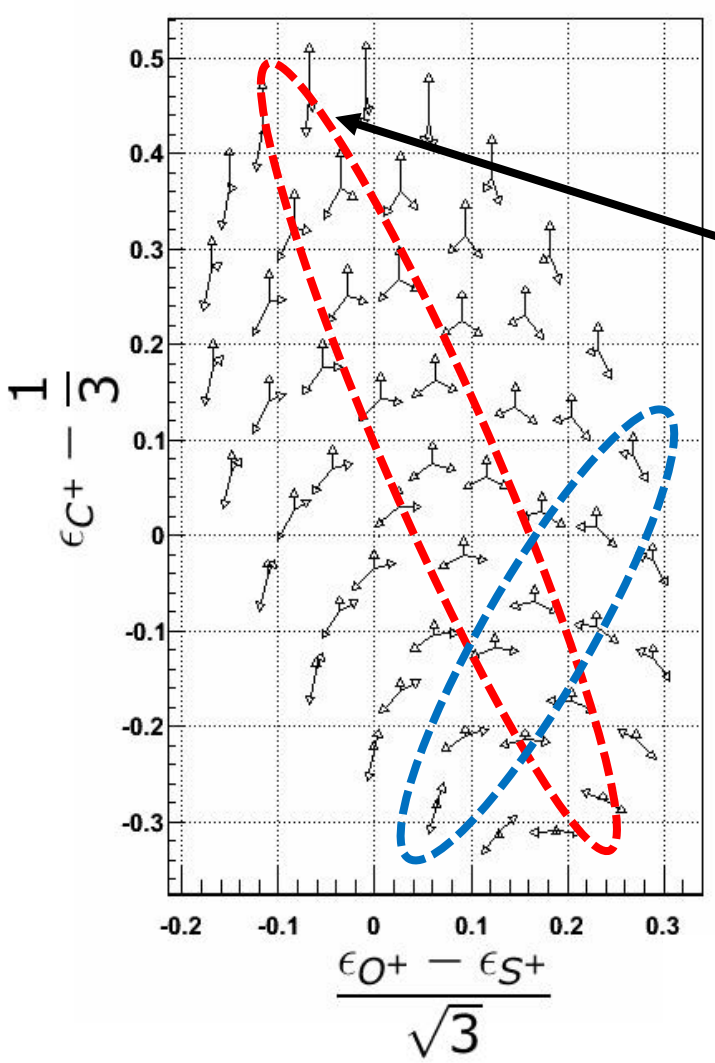


Dalitz plots



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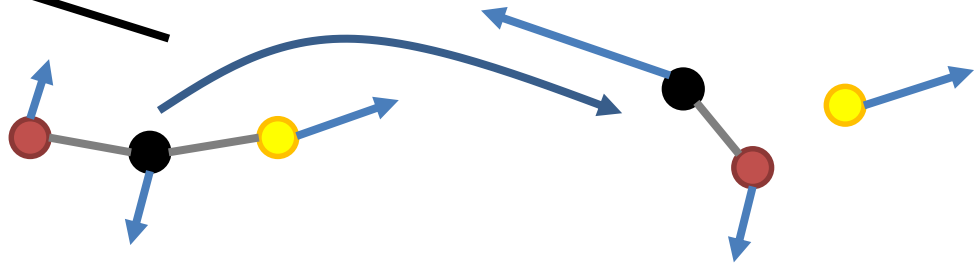
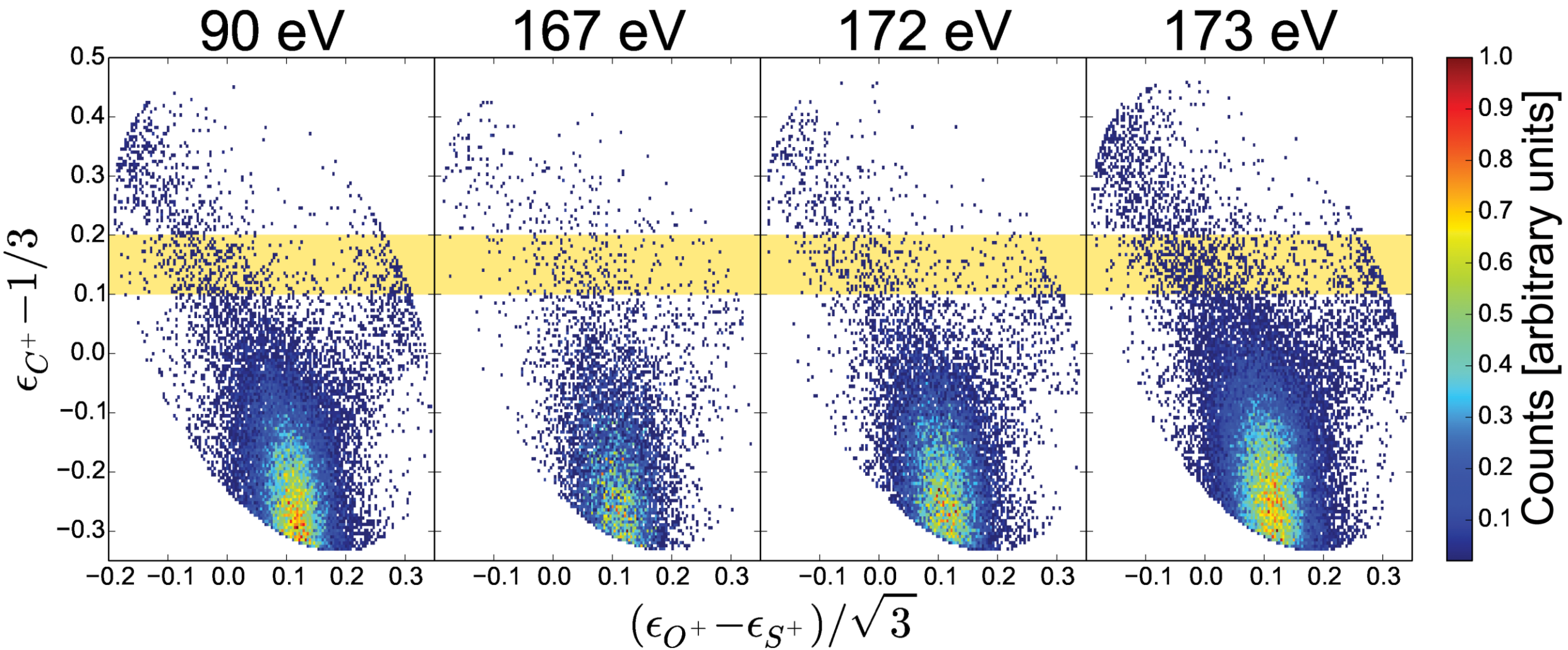


$OCS^{3+} \rightarrow OC^{2+} + S^+ \rightarrow O^+ + C^+ + S^+$

$OCS^{3+} \rightarrow O^+ + CS^{2+} \rightarrow O^+ + C^+ + S^+$

Results: timescale effects (synchrotrons are fast!)



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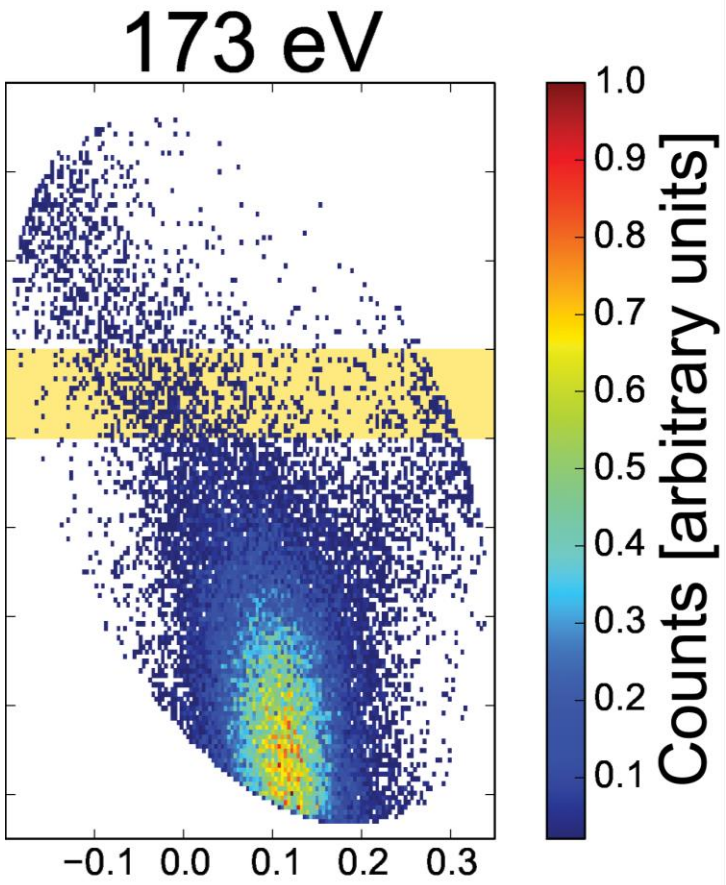
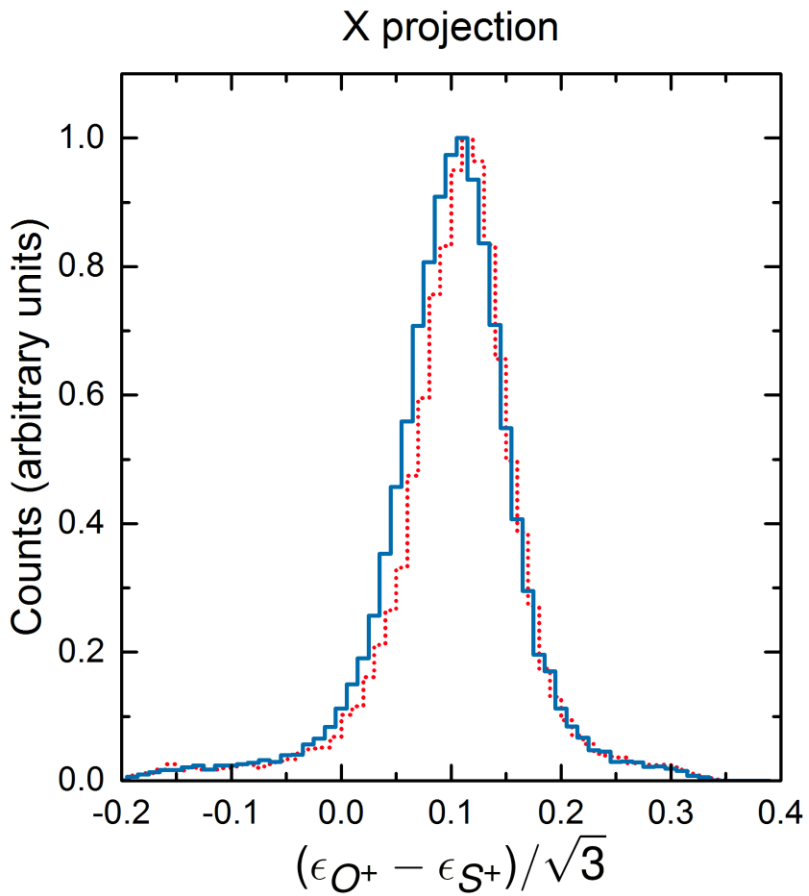
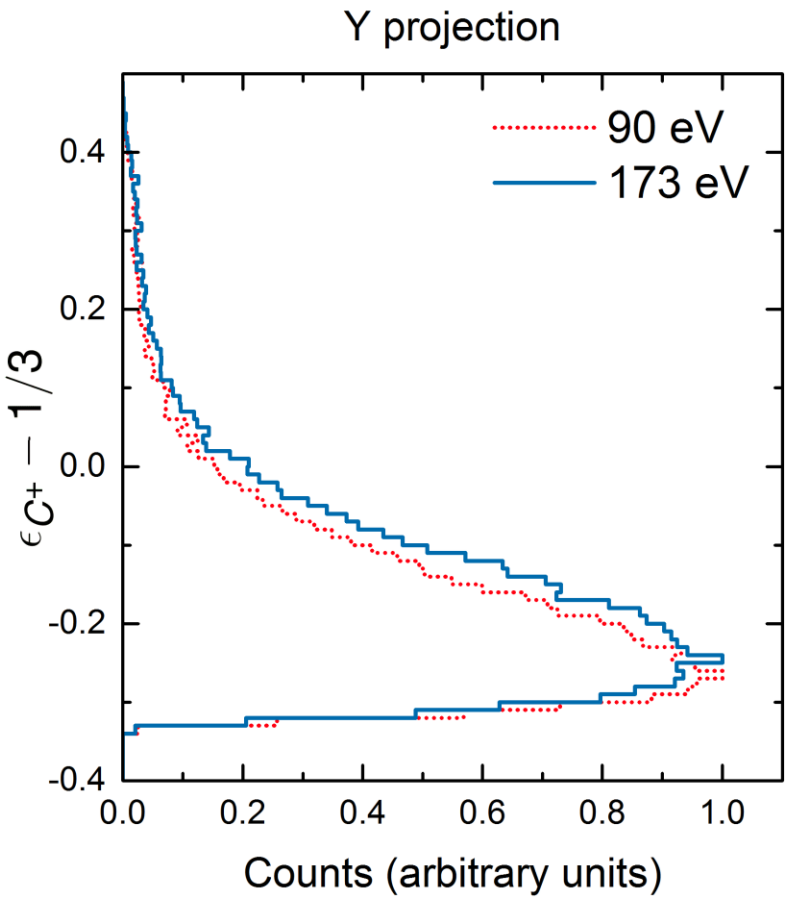
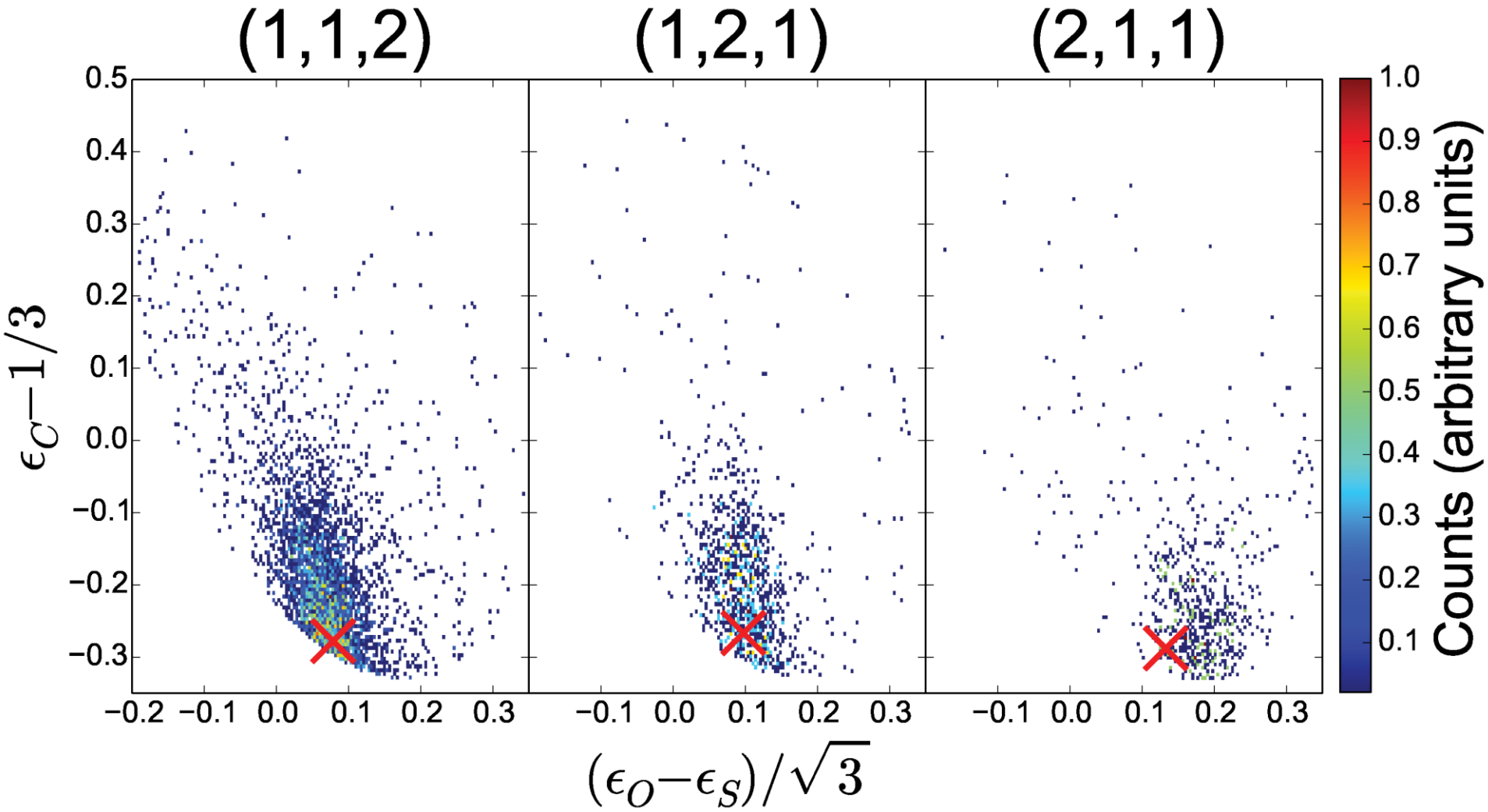






Results: OCS^{4+} oddities and non-equilibrium nuclear rearrangement



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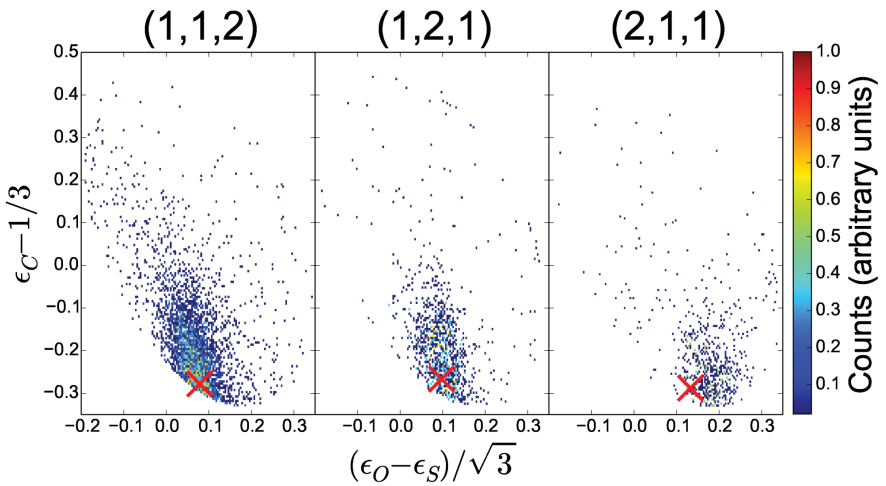
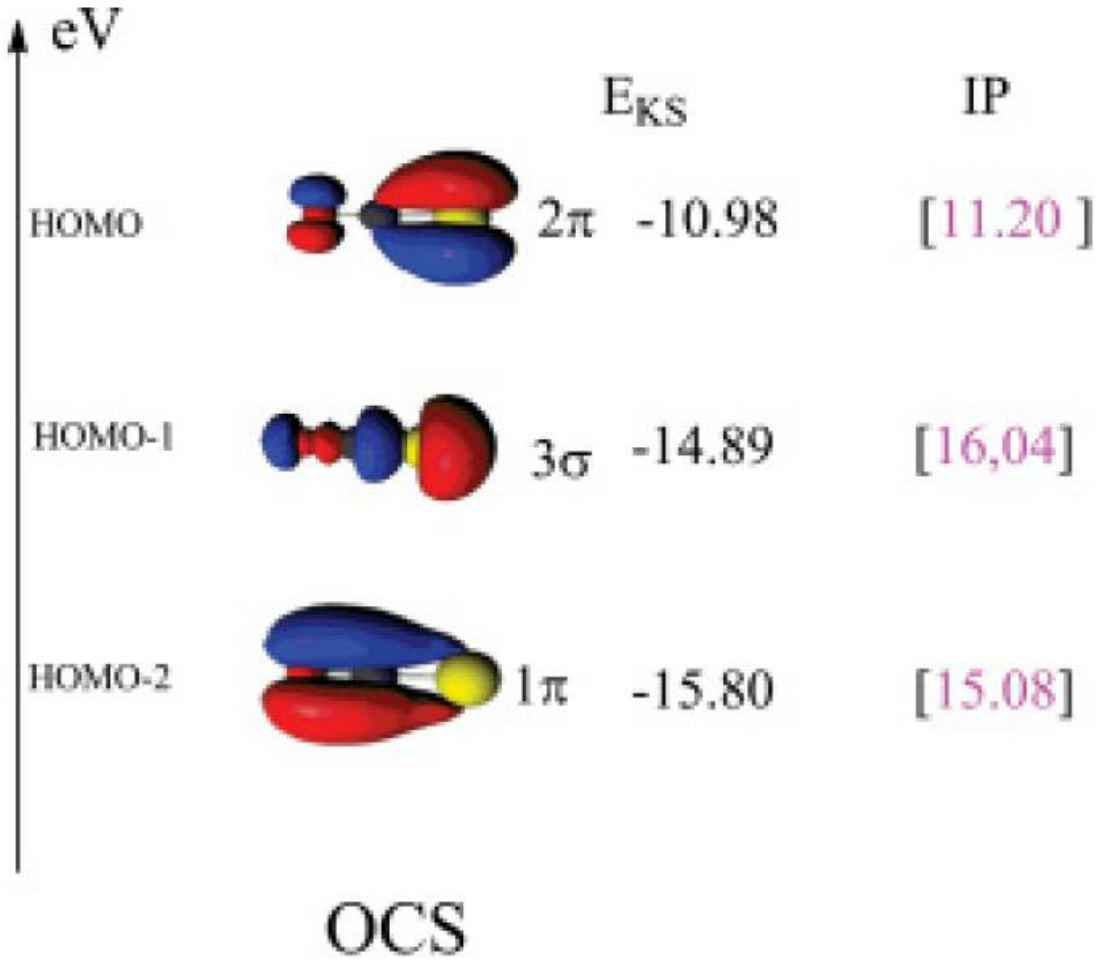


1. Removing electrons 1 and 2 shorten the CO bond.
2. Removing electrons 3 and 4 won't change bonds much.
3. HOMO-2 is extremely bonding w.r.t. CO, further shortening it.

4. Upon exploding, O^{2+} now has more energy than expected.



- Fowe et al., *Phys. Rev. A* **84**, 035402 (2011)

Conclusions (and wishful thinking)

In reality, experimenters
are cussed individuals.

Richard Henry Dalitz
*Nature **314**, 387 (1985)*

Conclusion and future experiments

- We can control the energy input. What can we control?
- Synchrotron ionization is faster than ultrafast laser and events may be resolved.
- Oddities and single processes may be resolved and "naively" explained with skepticism.
- X-ray beam polarization control will allow more molecules to be imaged.
- Move on to larger and more interesting molecules and chemical reactions.

Quotation is a serviceable substitute for wit.

Oscar Wilde

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Waterloo: Joe Sanderson, Benji Wales, Cathy Cai, Reza Karimi

CLS: Mike MacDonald, Lucia Zuin, Isabelle Gauthier

Canadian Centre canadien
Light de rayonnement
Source synchrotron

NSERC
CRSNG