BERKELEY LAB
Bringing Science Solutions to the World

U.S. DEPARTMENT OF ENERGY
Office of Science

Precision Spectroscopy of Heavy and Superheavy Elements with AETHER

Erich Leistenschneider

Nuclear Science Division, Heavy Element Group

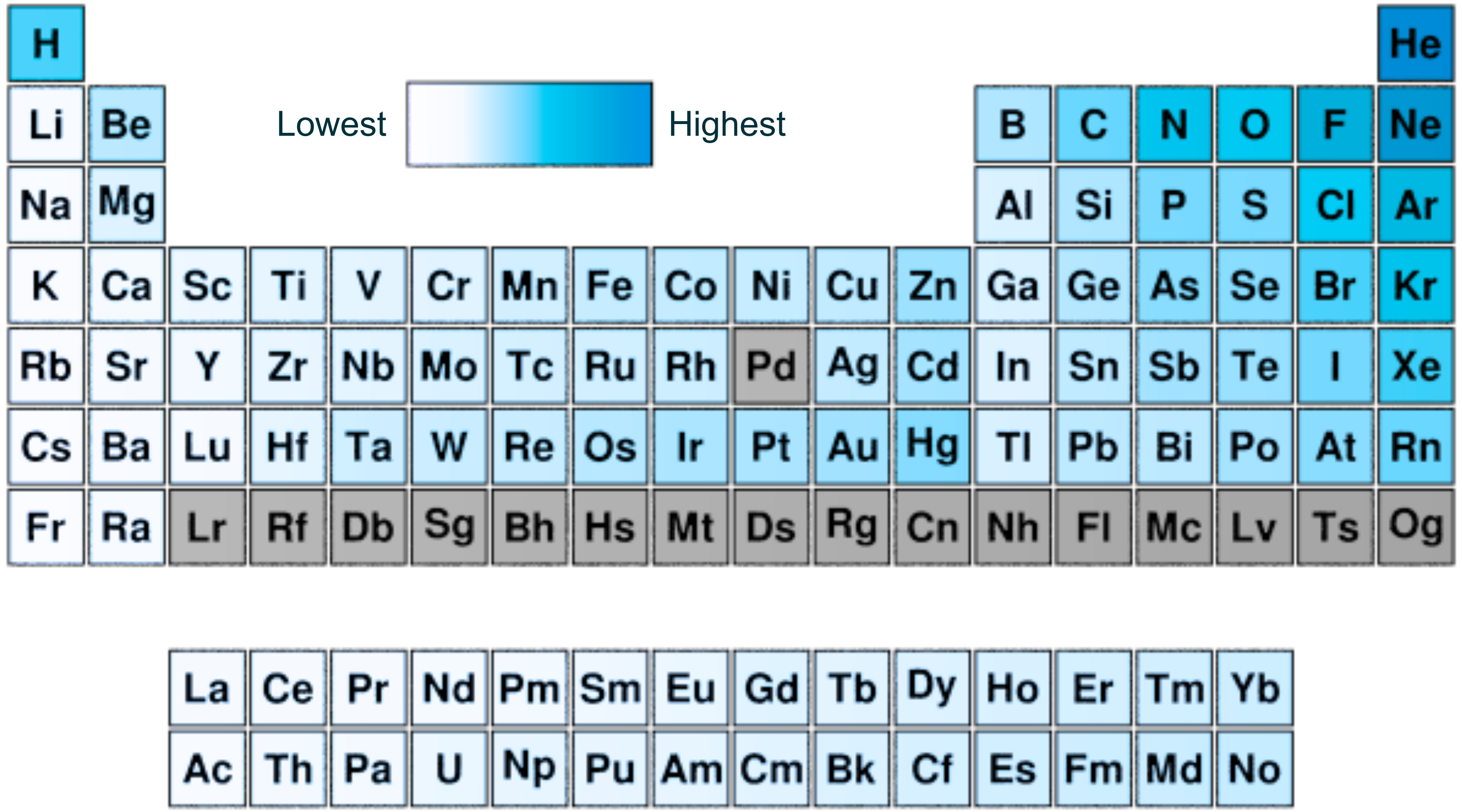
June 2024

GAME TIME!

Only Rule: Do not google it!

Quiz #1

What is the quantity represented in the color code of this Periodic Table?



Modified from www.webelements.com

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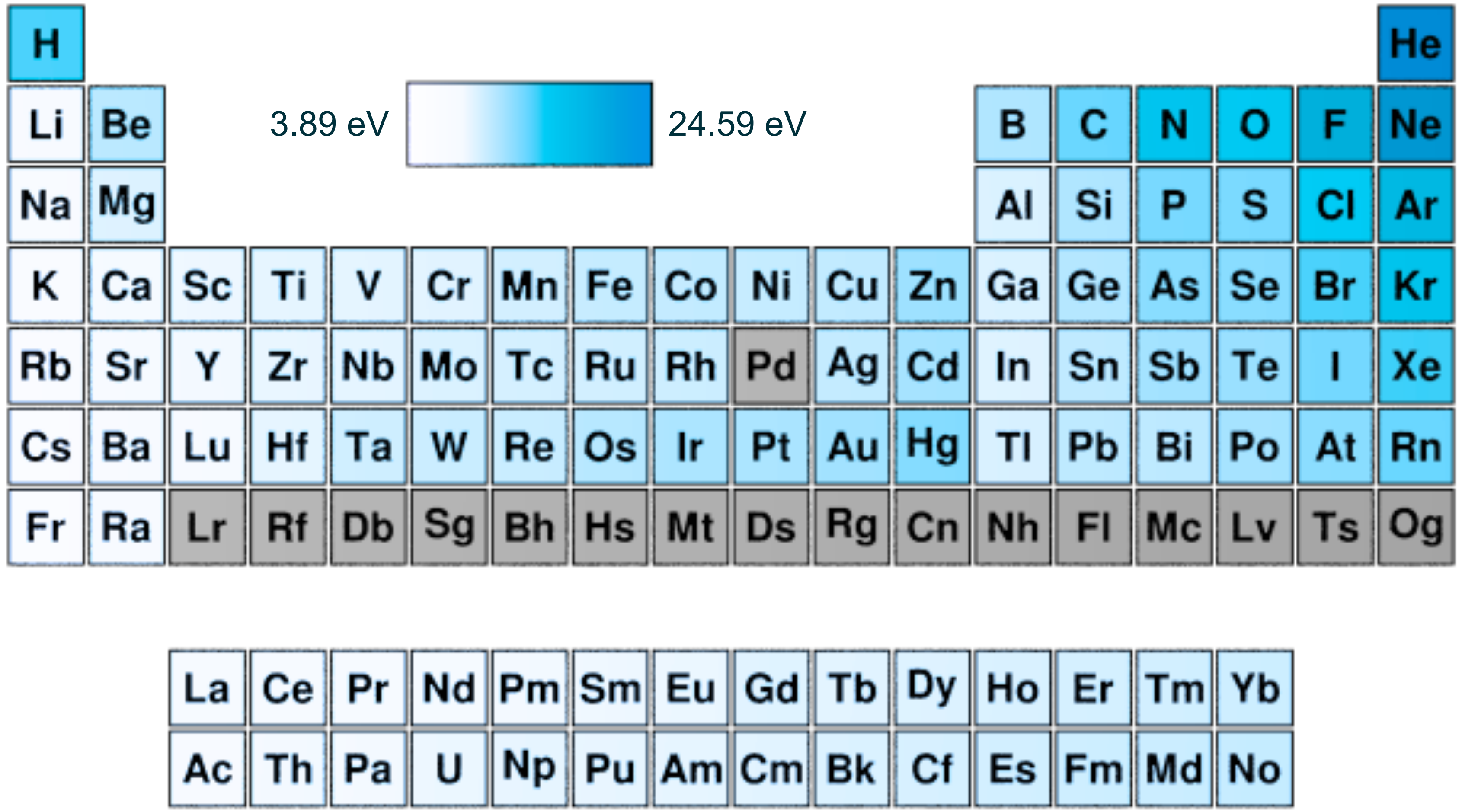

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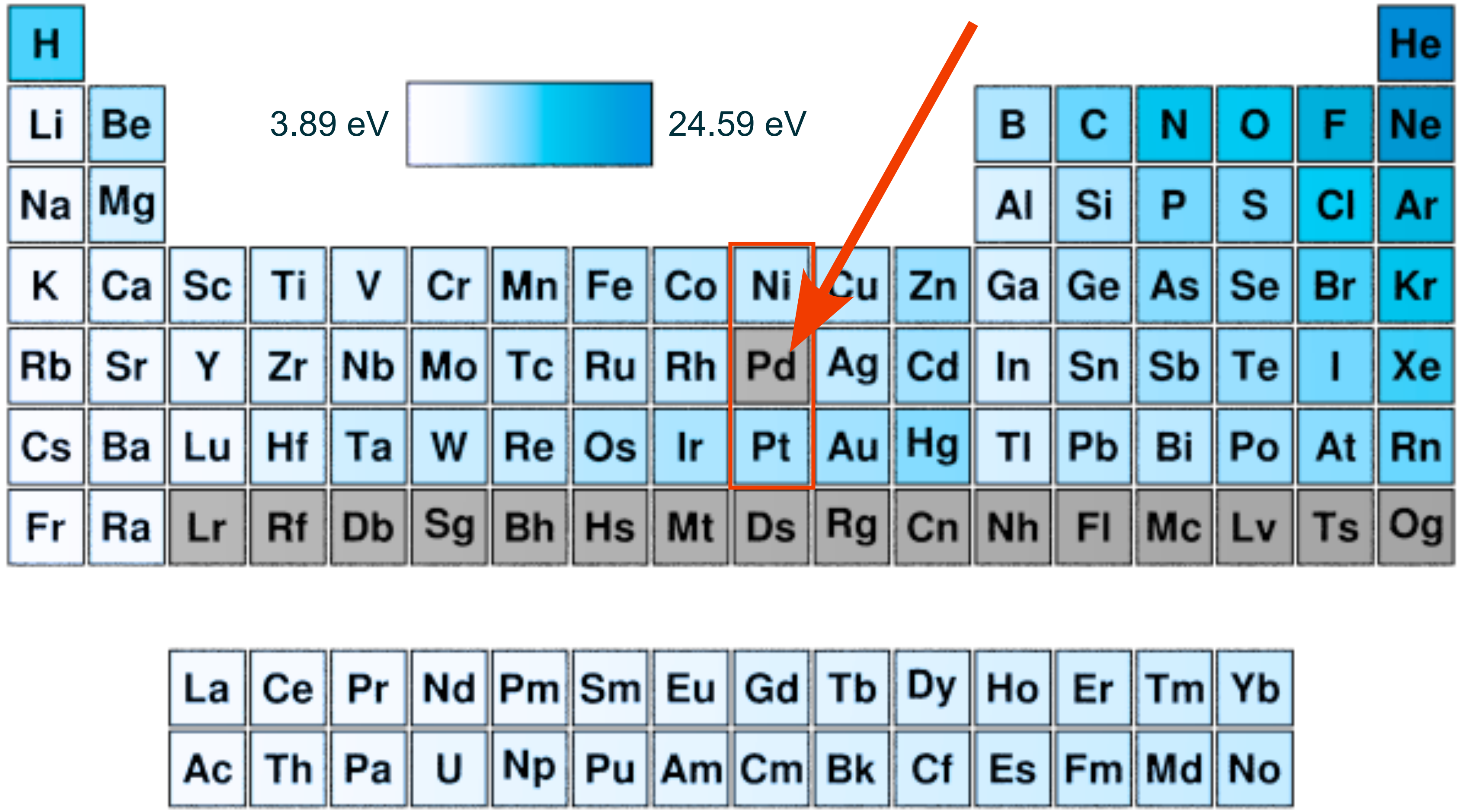
1st Ionization Potential

Least amount of energy needed to remove an electron from a neutral atom

Quiz #2

What is the IP of Paladium?



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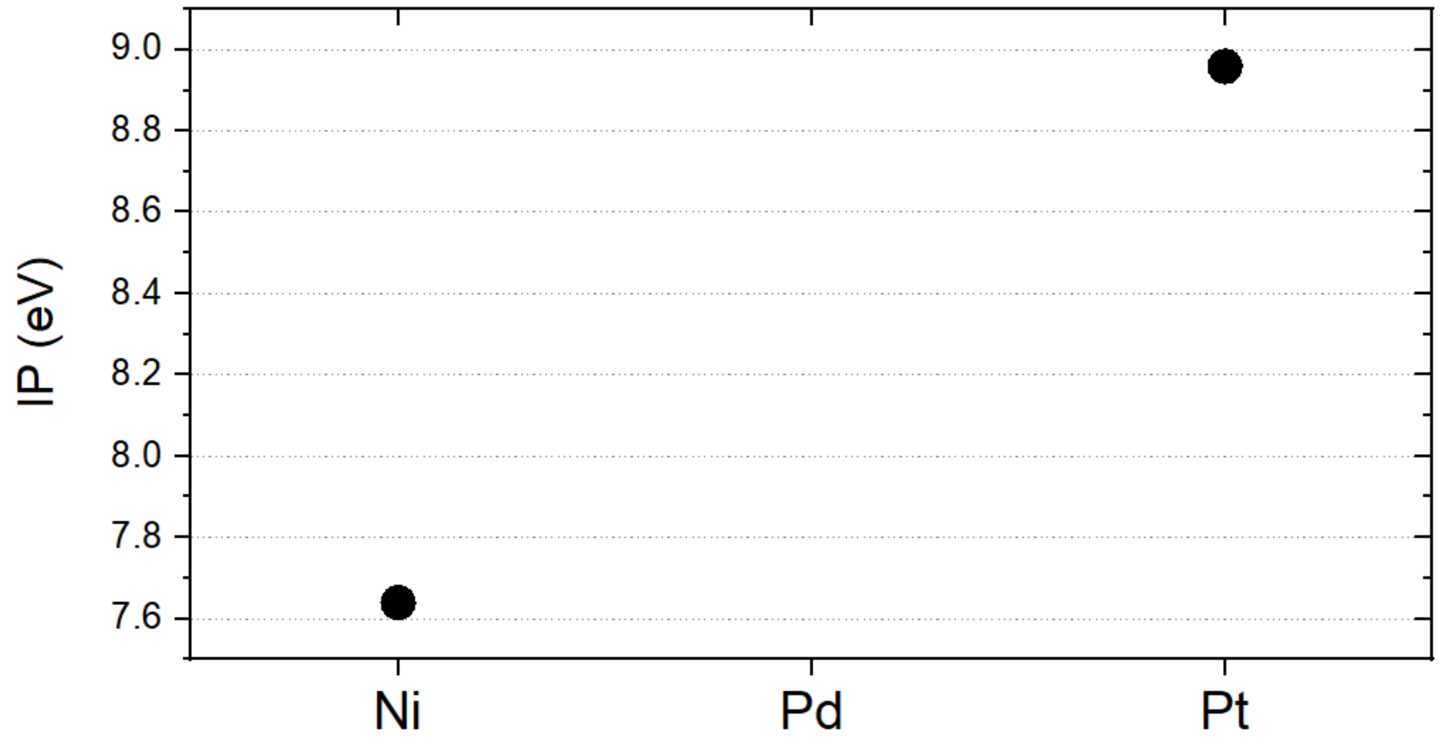

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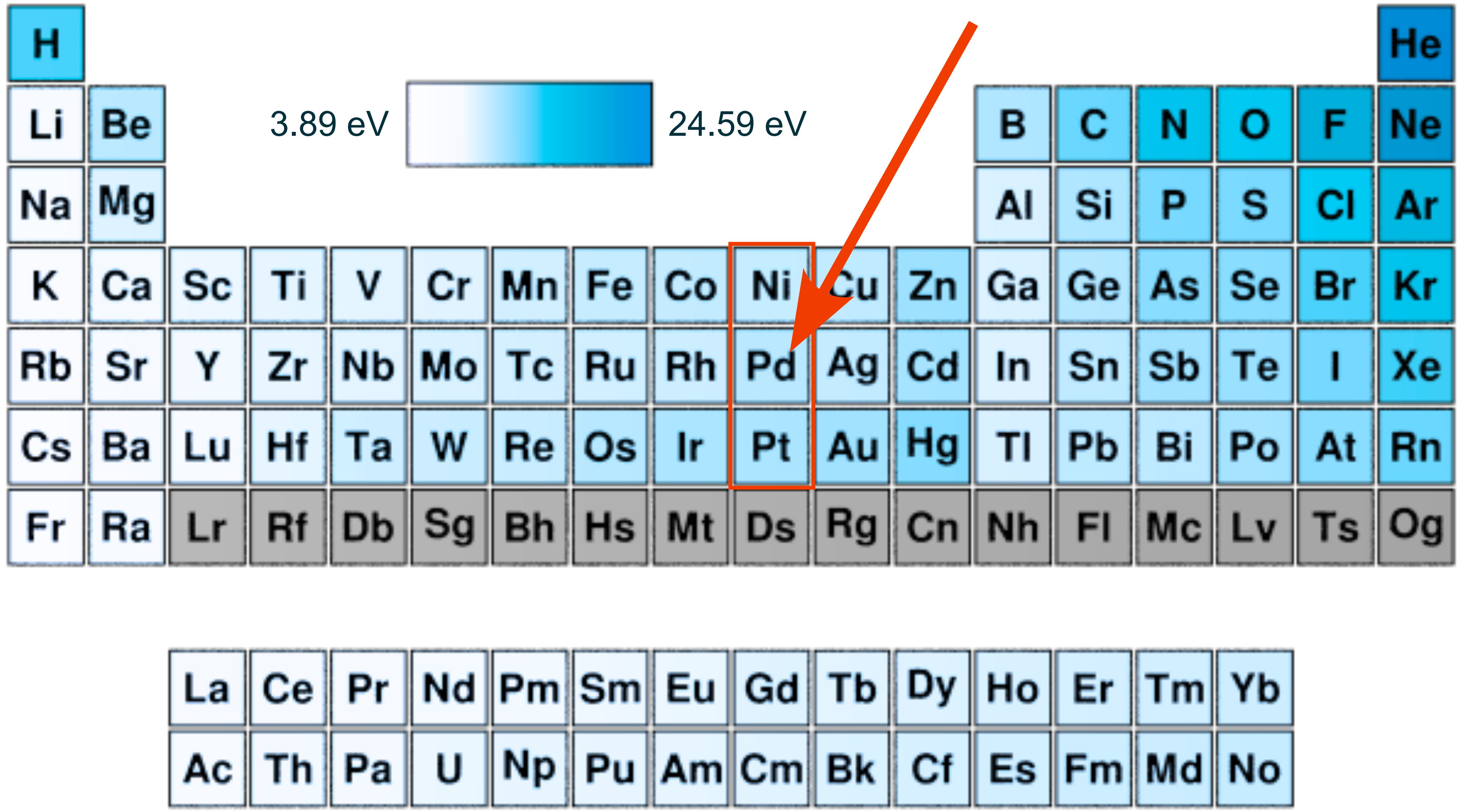

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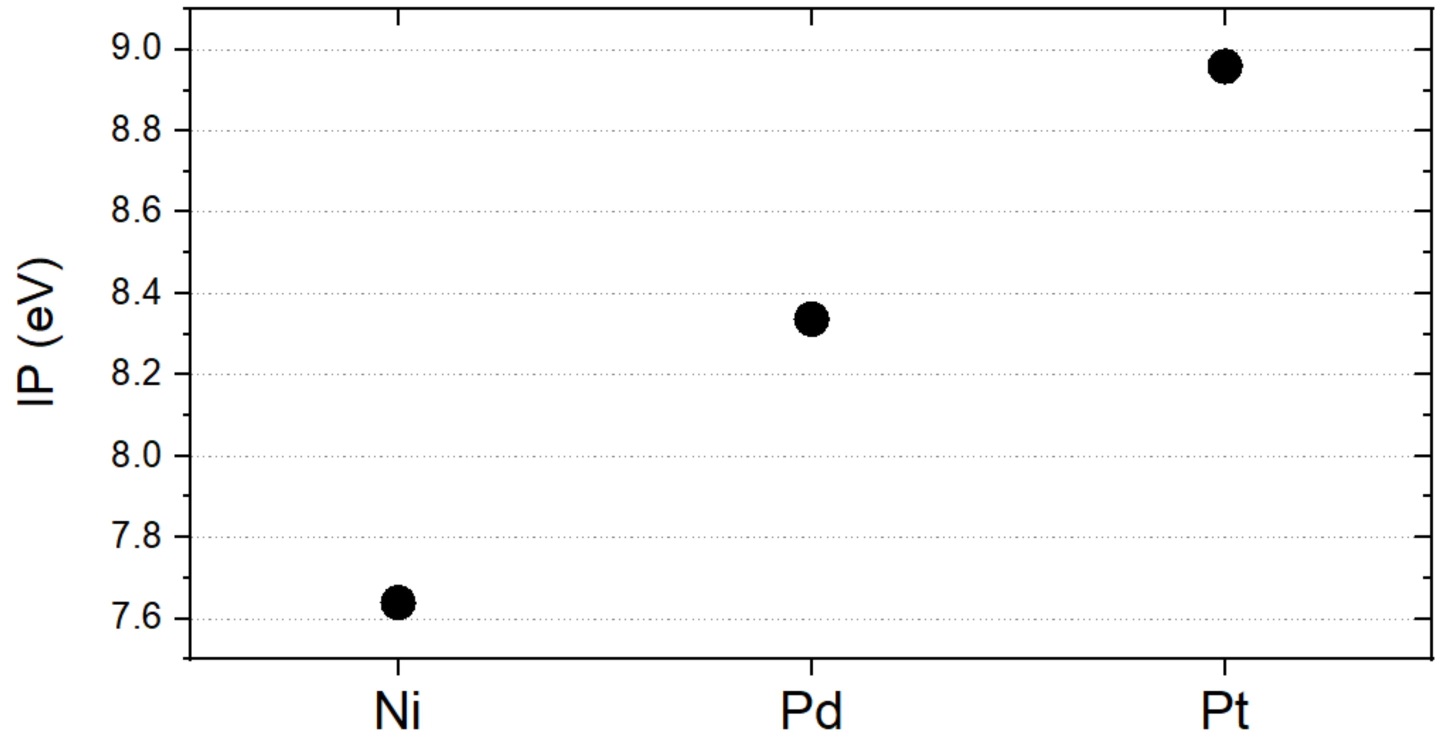
8.3369(1) eV



Quiz #3

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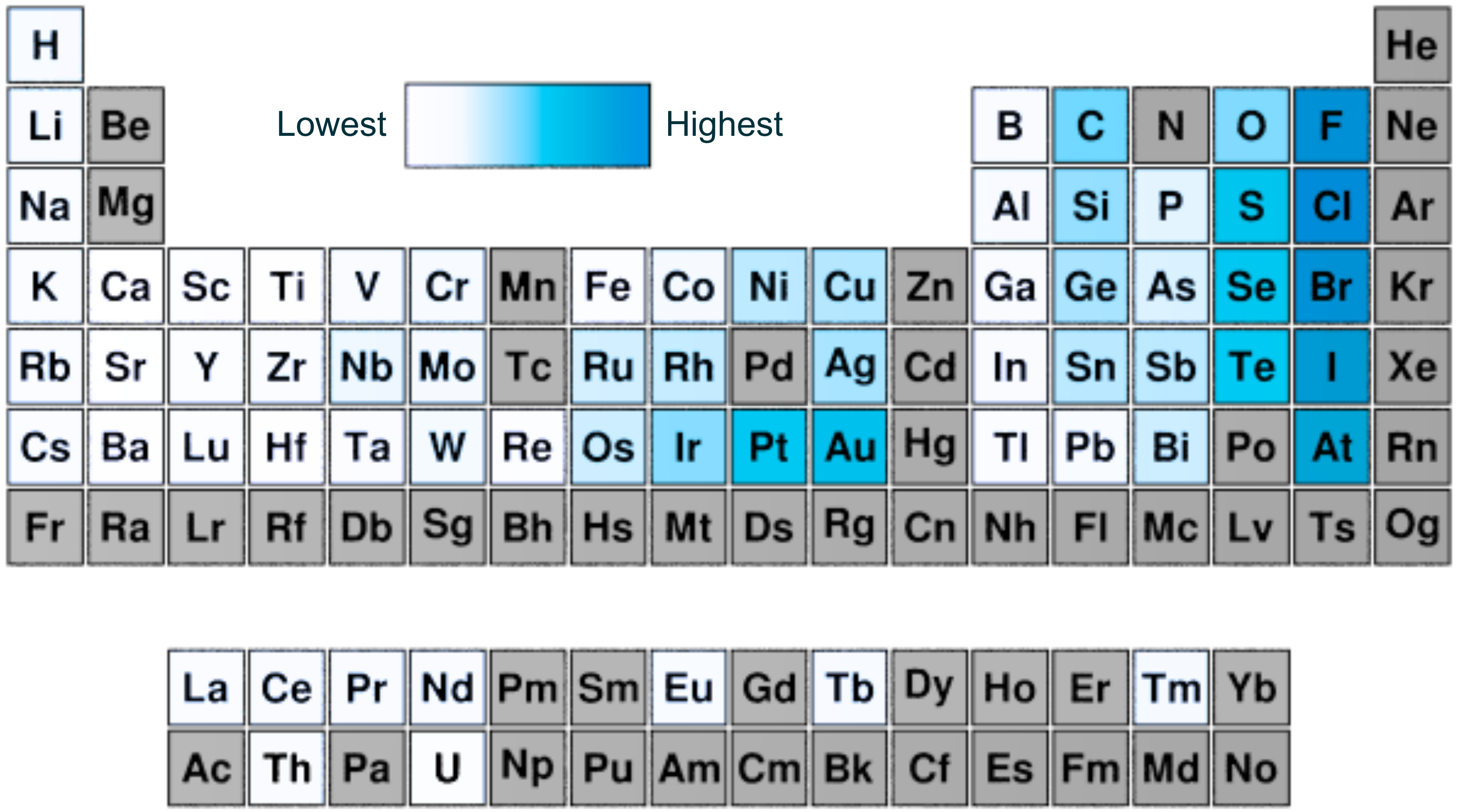

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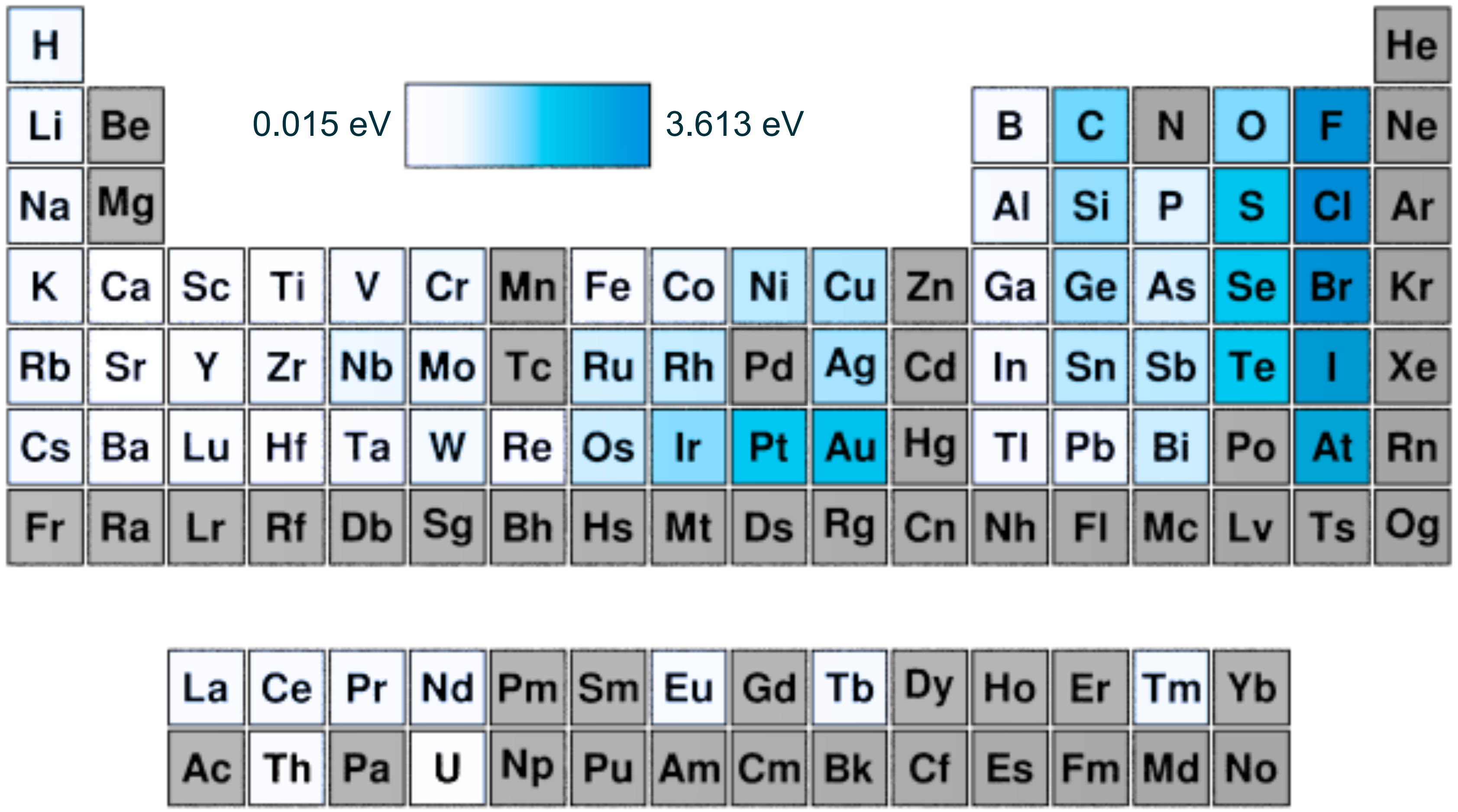


Modified from www.webelements.com

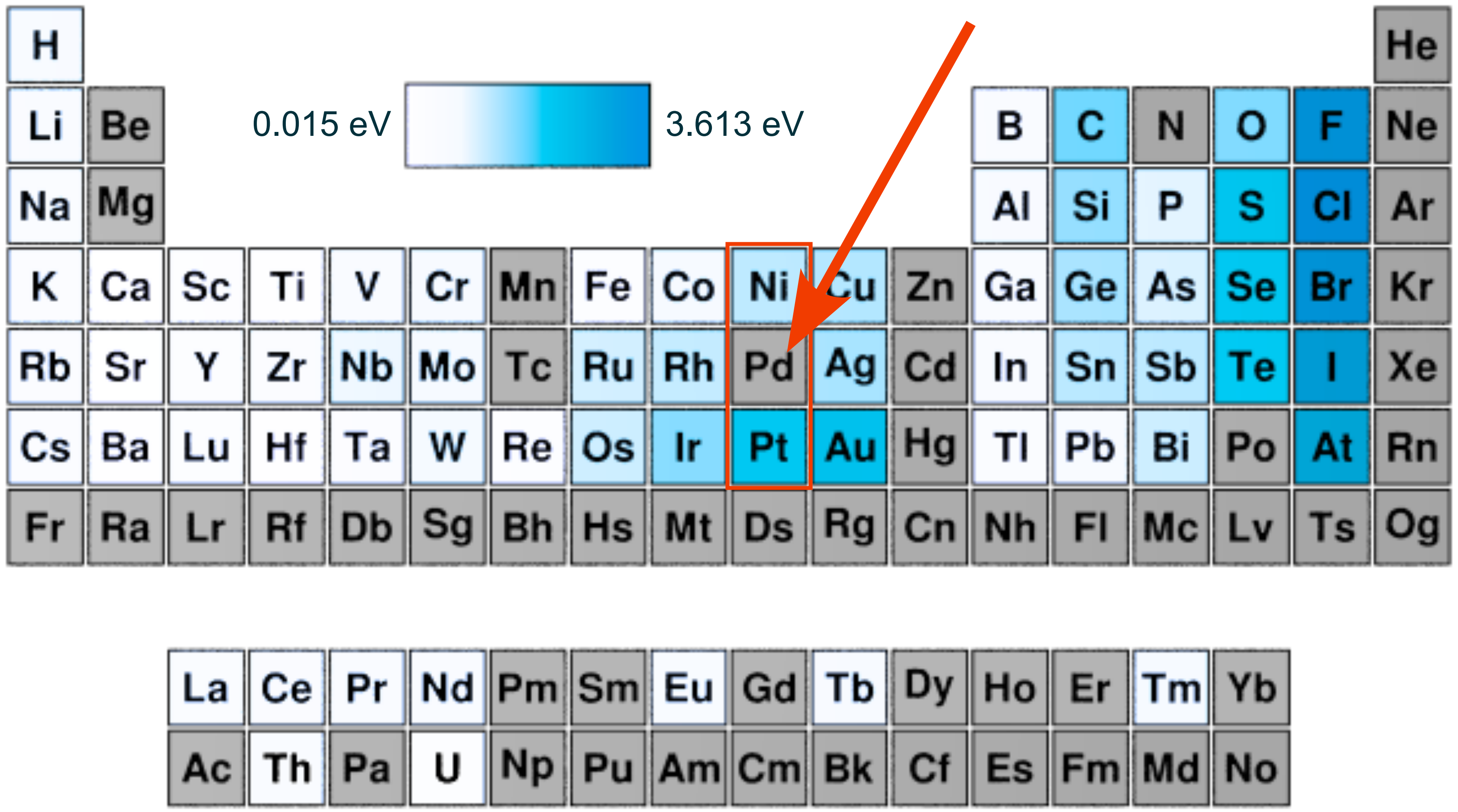
Electron Affinity

Energy needed to remove an electron from a **negative ion**

Of fundamental importance for **chemistry**: Strongly related to how much an element is prone to form chemical bonds by sharing electrons

Quiz #4

What is the EA of Paladium?



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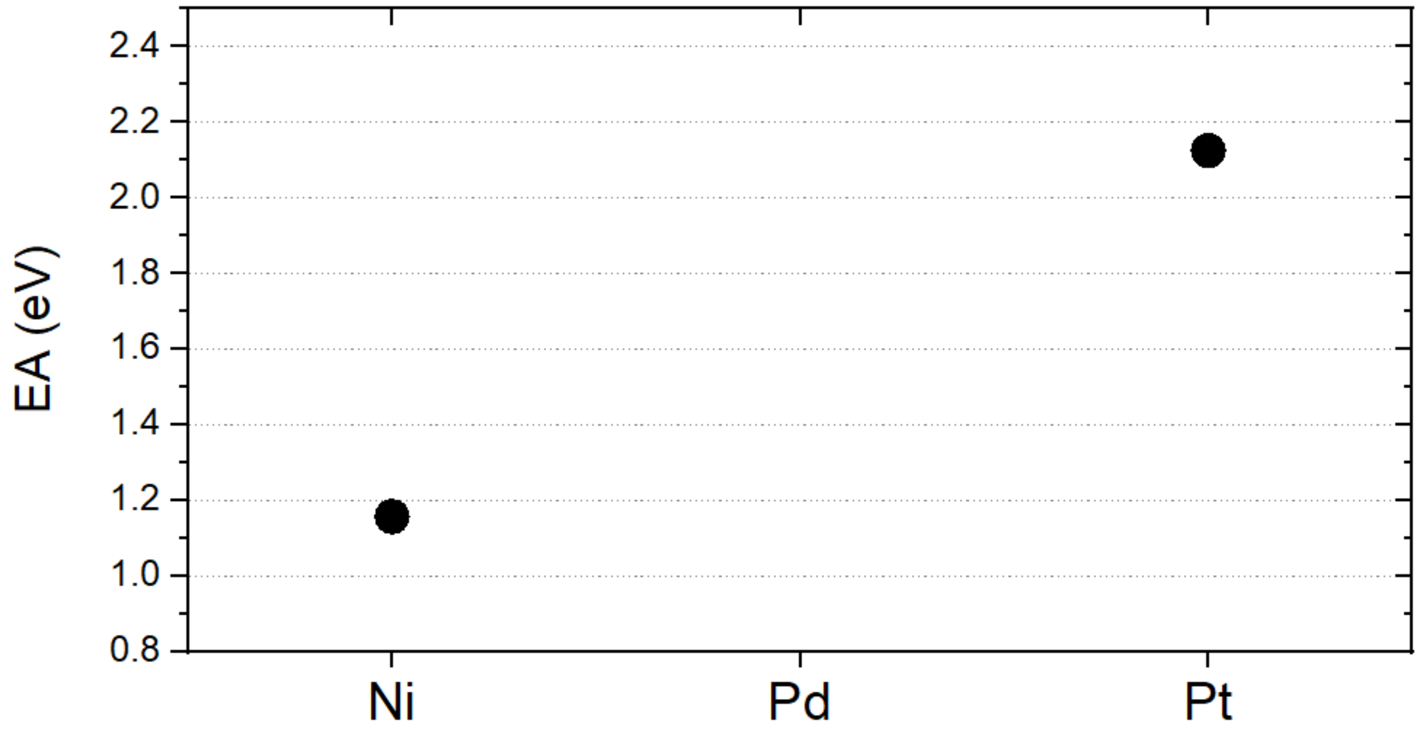
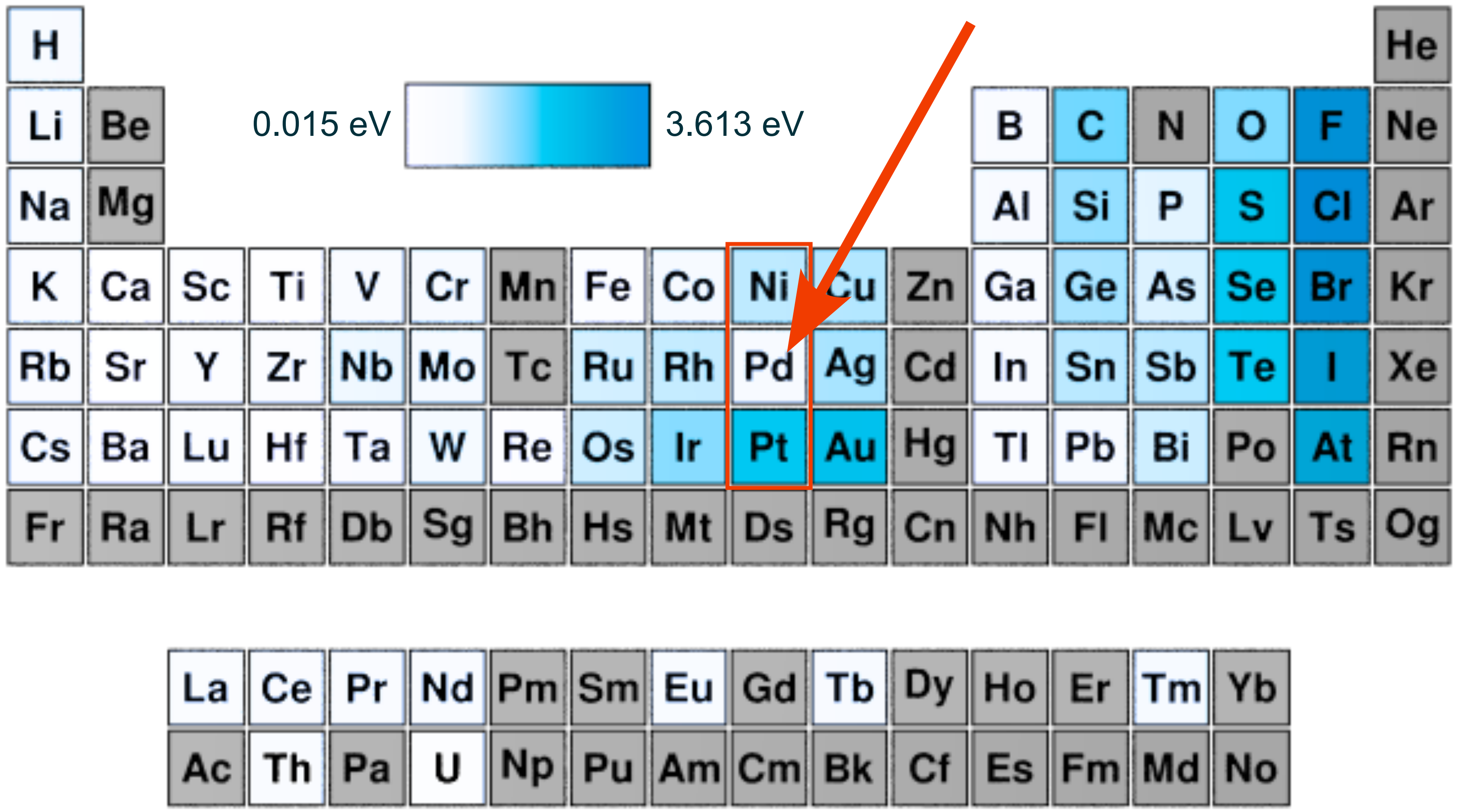


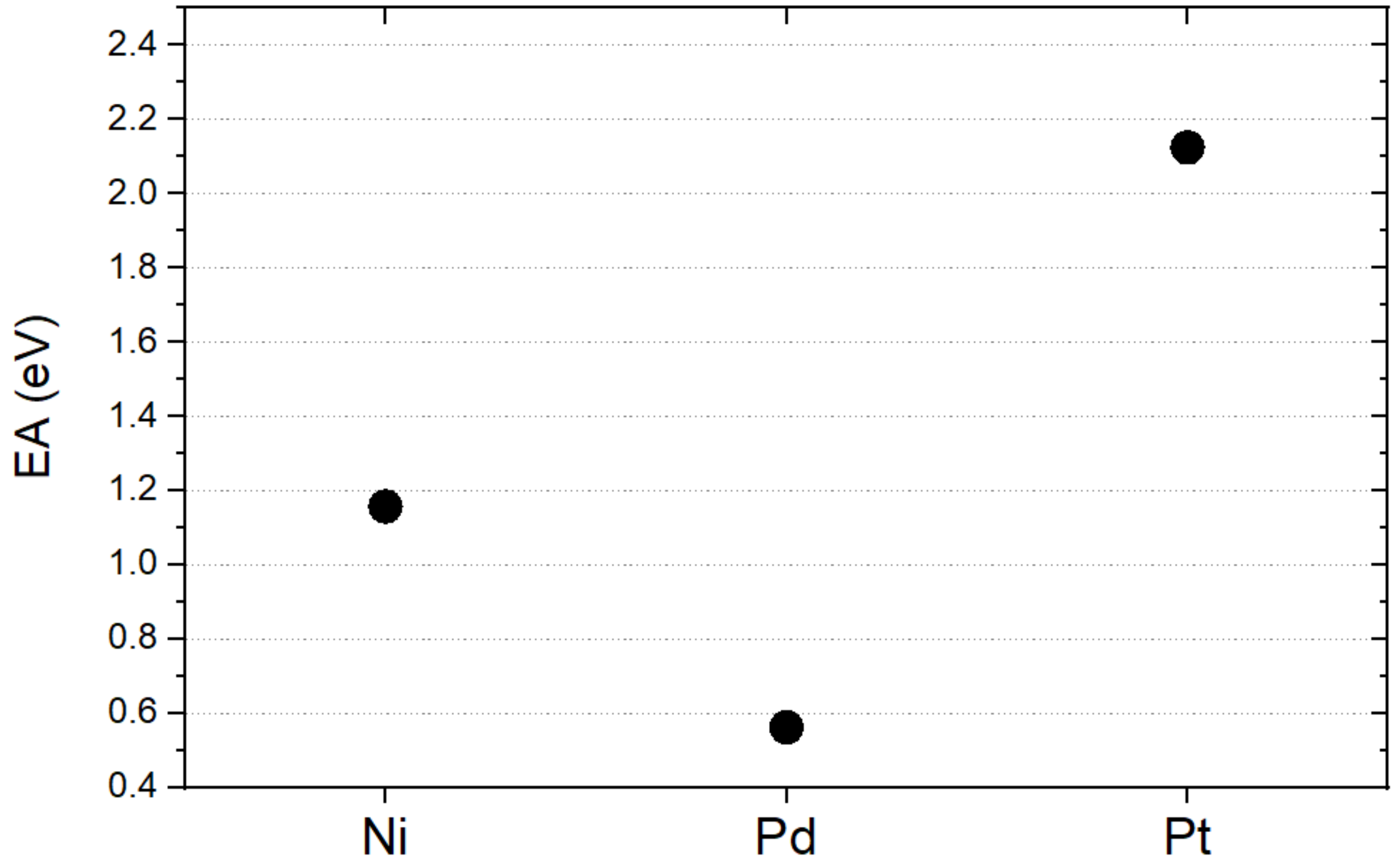
Quiz #4

What is the EA of Paladium?



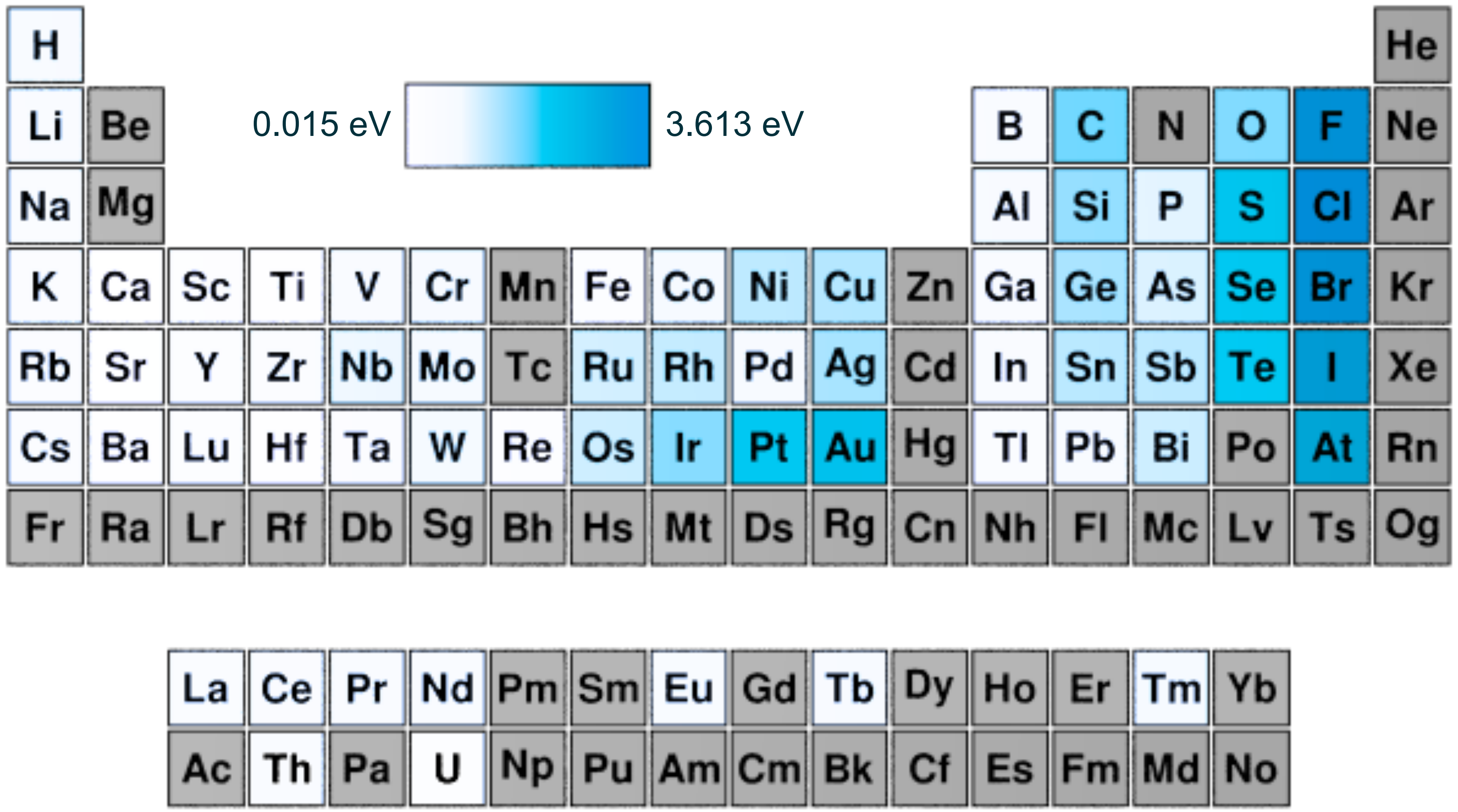
Modified from www.webelements.com

0.5621(1) eV



The Electron Affinity Landscape

Negative Ions are challenging…



Modified from www.webelements.com

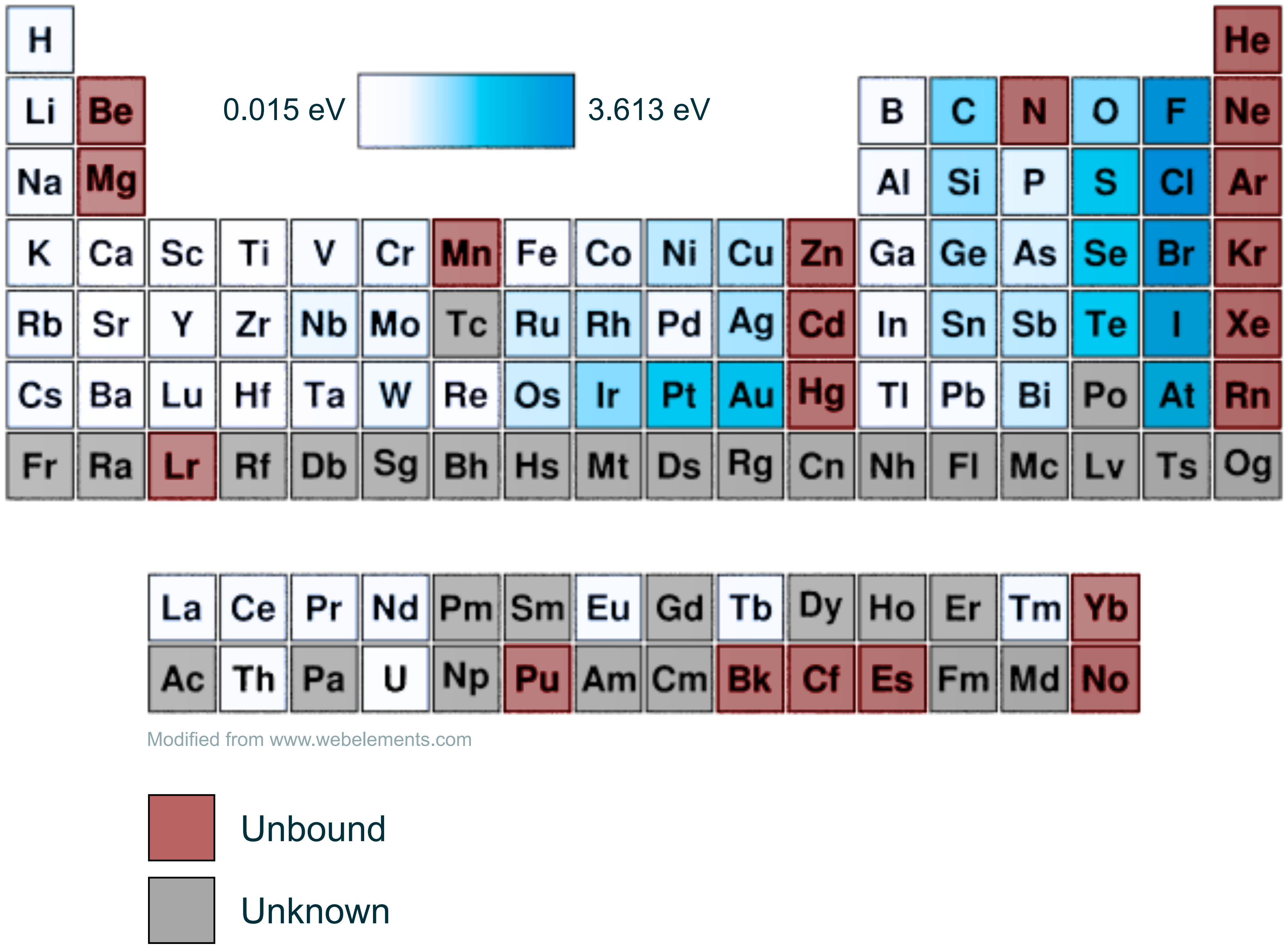
… theoretically …

Mostly bound by **electron-electron correlations**
Therefore they cannot be described through mean-field approaches.

Of fundamental importance for **atomic physics:**
Ideal systems to benchmark atomic theories by their ability to describe many-body dynamics.

The Electron Affinity Landscape

Negative Ions are challenging…



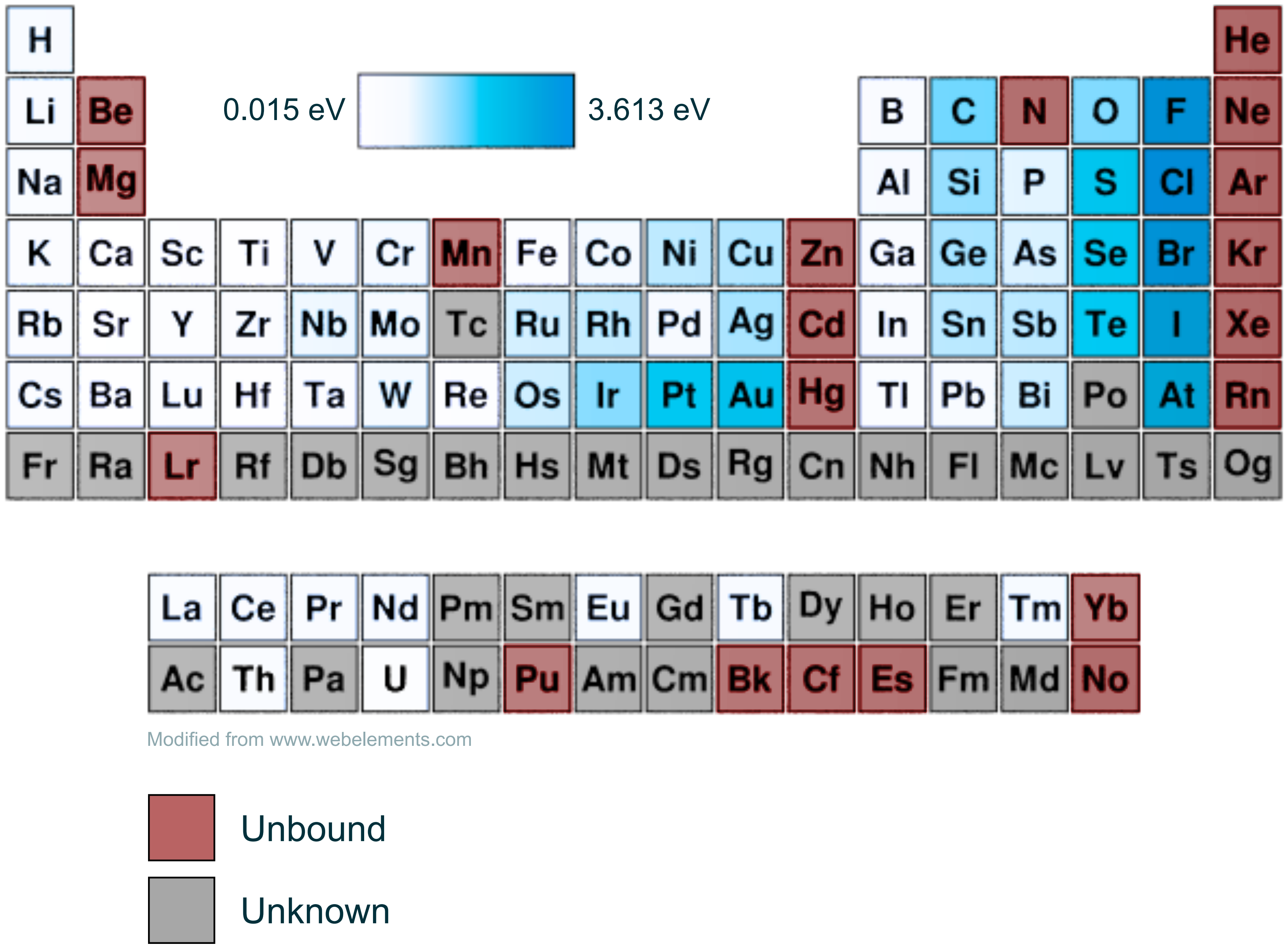
Modified from www.webelements.com

… and experimentally!

Techniques often require macroscopic quantities
Not sensitive enough to handle rare elements, which mostly affects actinides/superheavies.

The Electron Affinity Landscape

~1/3 of EAs in the Periodic Table are unknown



Missing opportunities on:

Fundamental & applied chemistry:
Radioprotection
Nuclear medicine
F-block chemistry

Fundamental atomic physics:
SHE and Actinides: highly-correlated systems, relativistic effects very pronounced

The AETHER Project at LBNL

Advanced Electrostatic Trap for Heavy Element Research

1. **The sensitivity gap:** how do we plan to solve the problem with Negative Ion Spectroscopy?

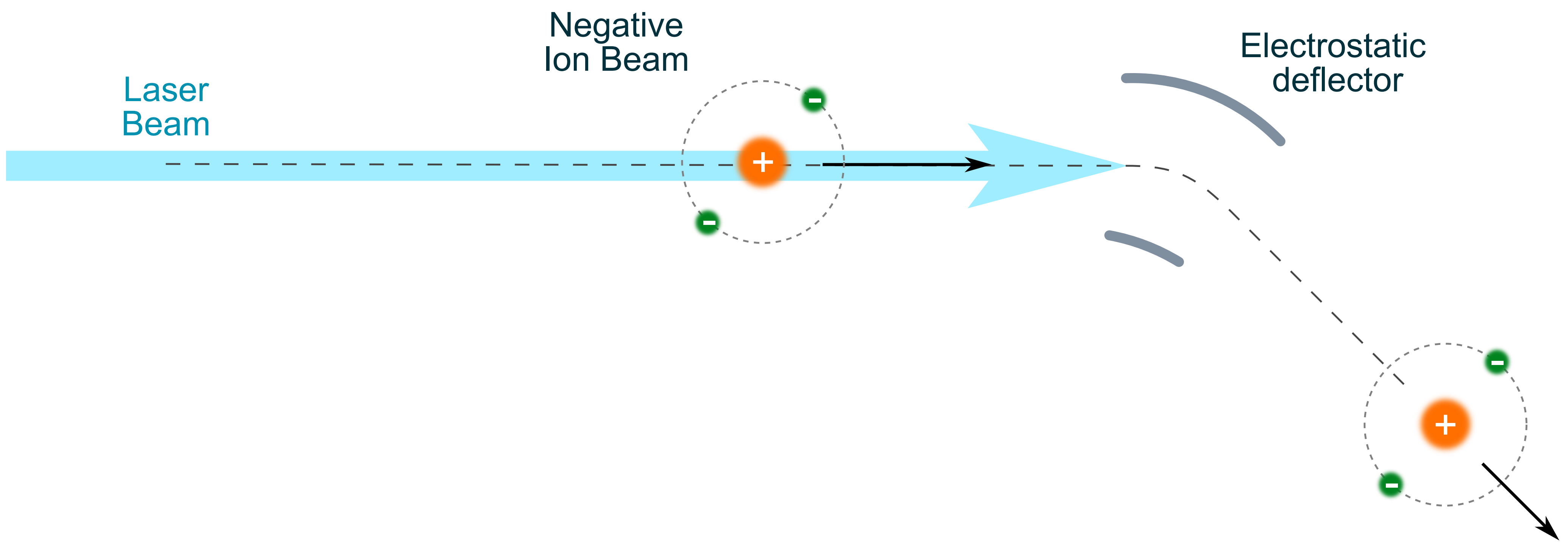
2. **AETHER's Concept:** our plans for the future infrastructure at LBNL

3. **Its status:** how far is our progress on assembly?

4. **Its "side quest":** high precision mass spectrometry for nuclear structure

The Sensitivity Gap

Laser Photodetachment Threshold (LPT) probes the energy to dismantle the negative ion:



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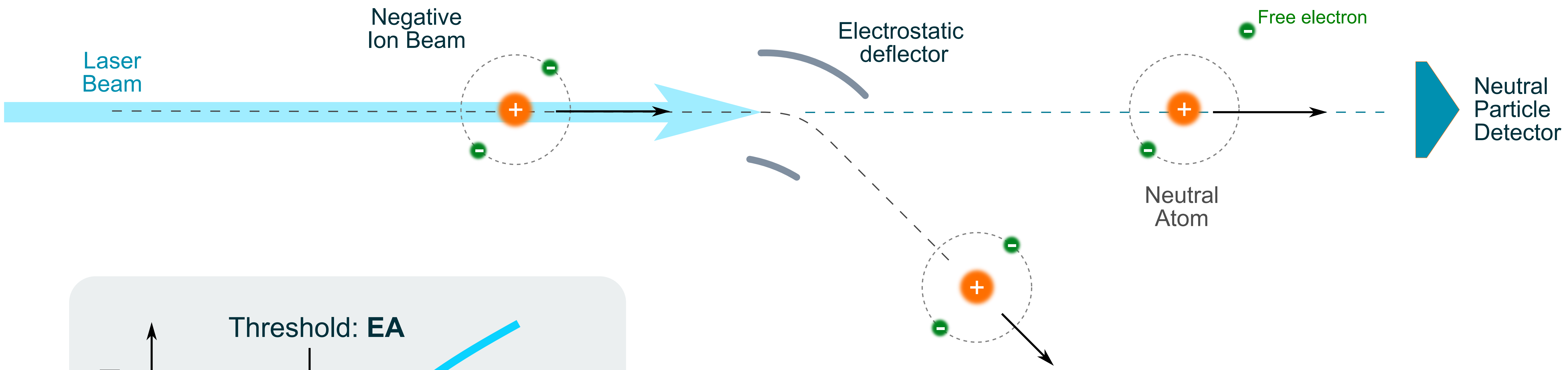

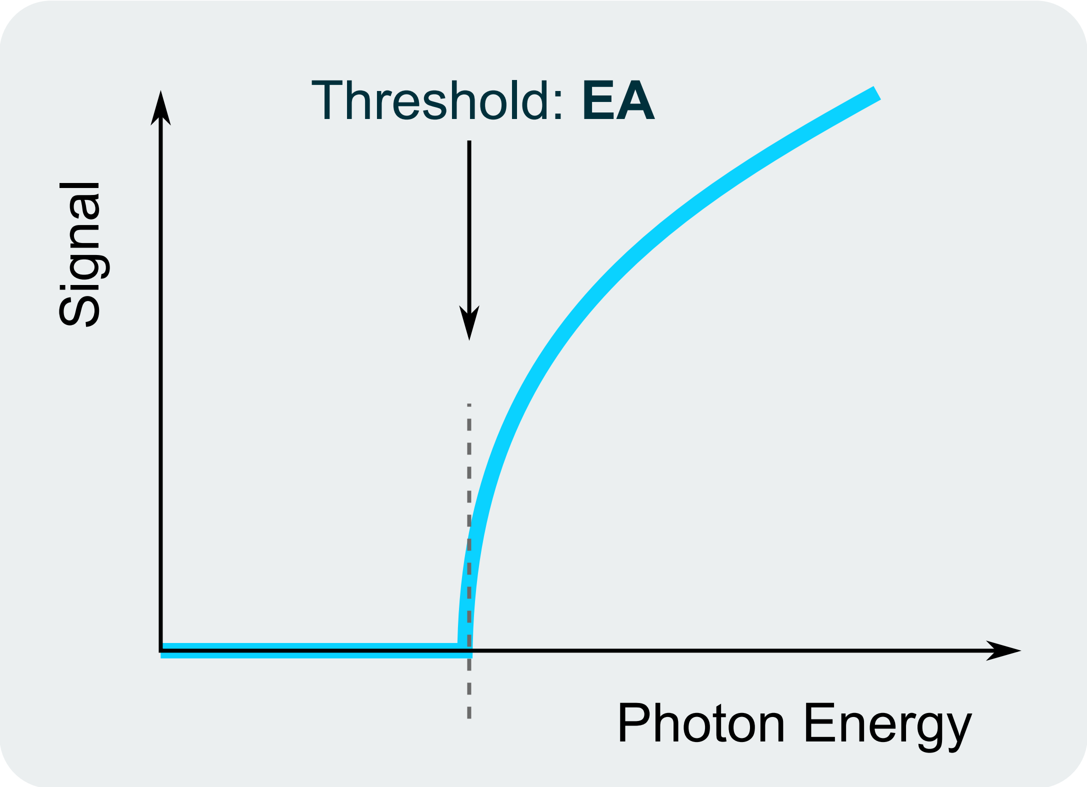

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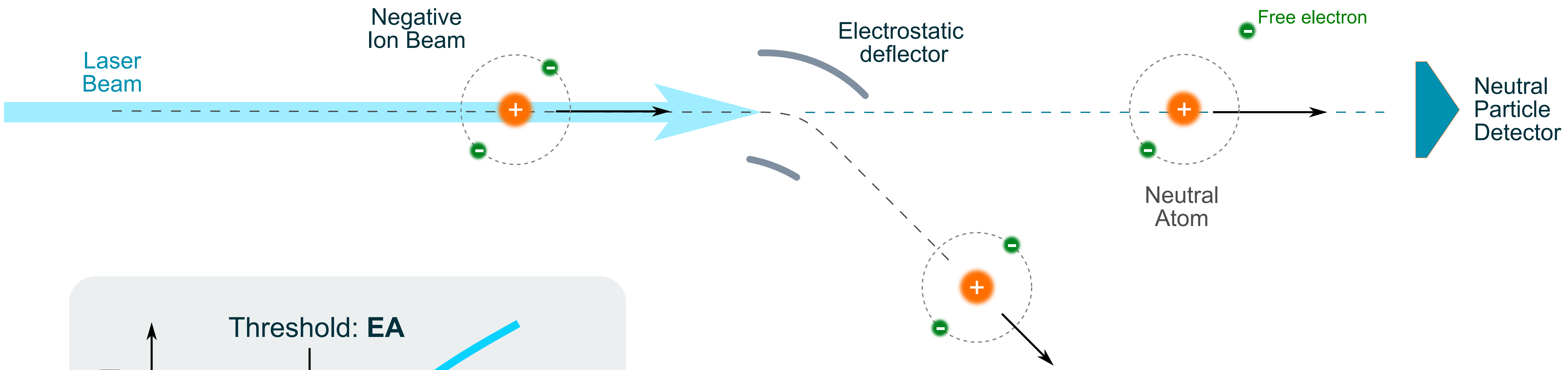
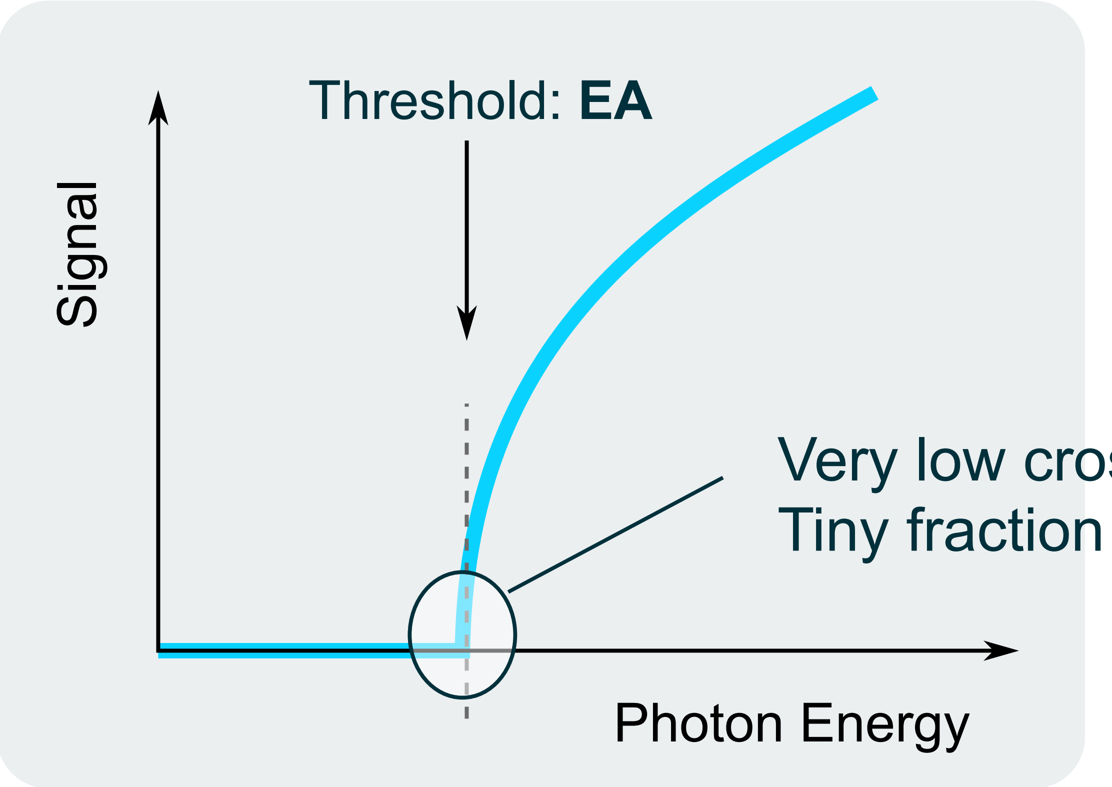
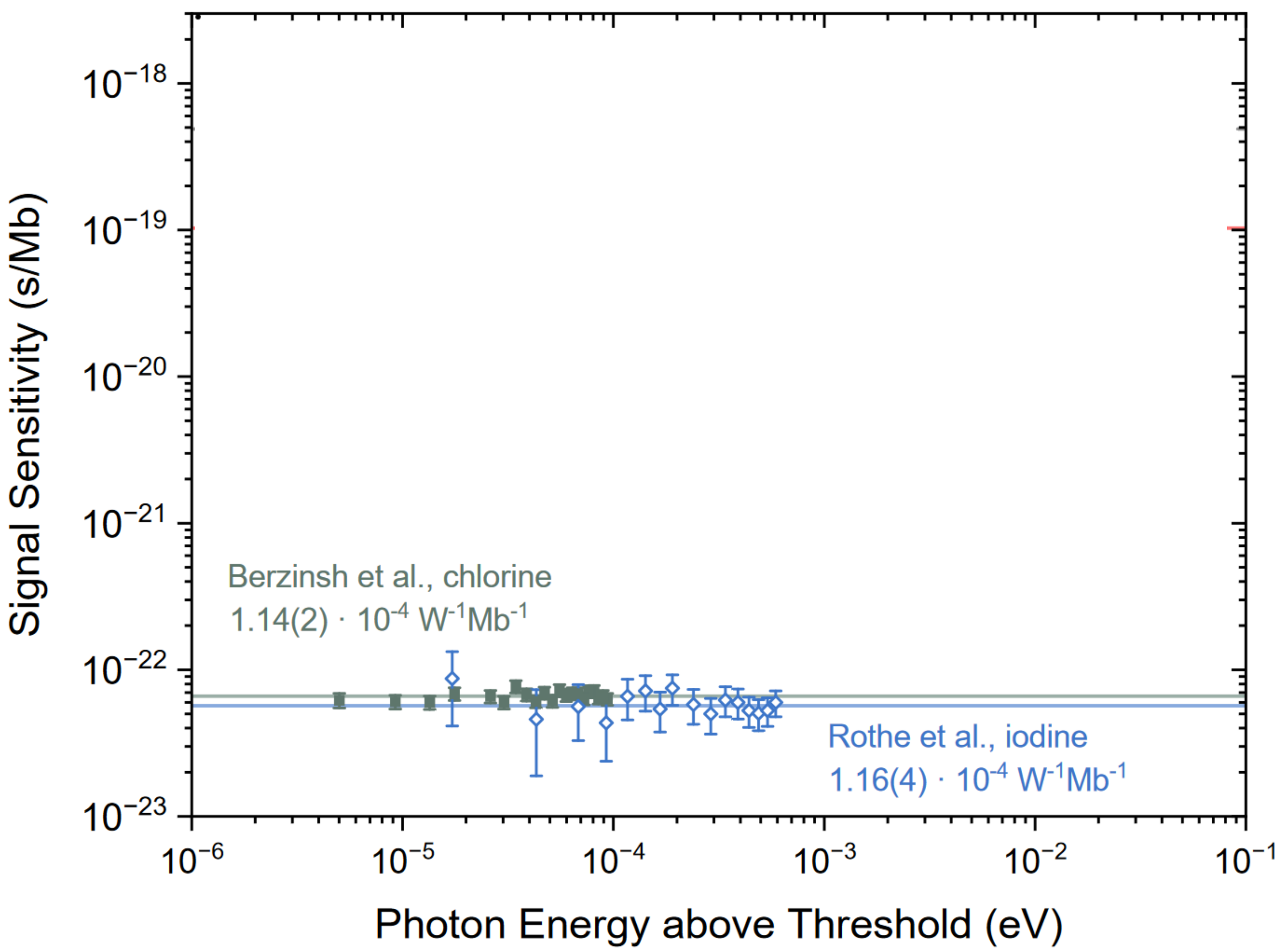




Very low cross section around threshold
Tiny fraction of ions are photodetached, large quantities needed = **low sensitivity!**

The Sensitivity Gap

What is the magnitude of the problem?



Sensitivity

Detected photodetachment event rate

$$\Gamma = \frac{N_{ev}}{N_{ion} \cdot \Phi \cdot \sigma}$$

Ion rate

Photon flux

Photodetachment Cross section

The Sensitivity Gap

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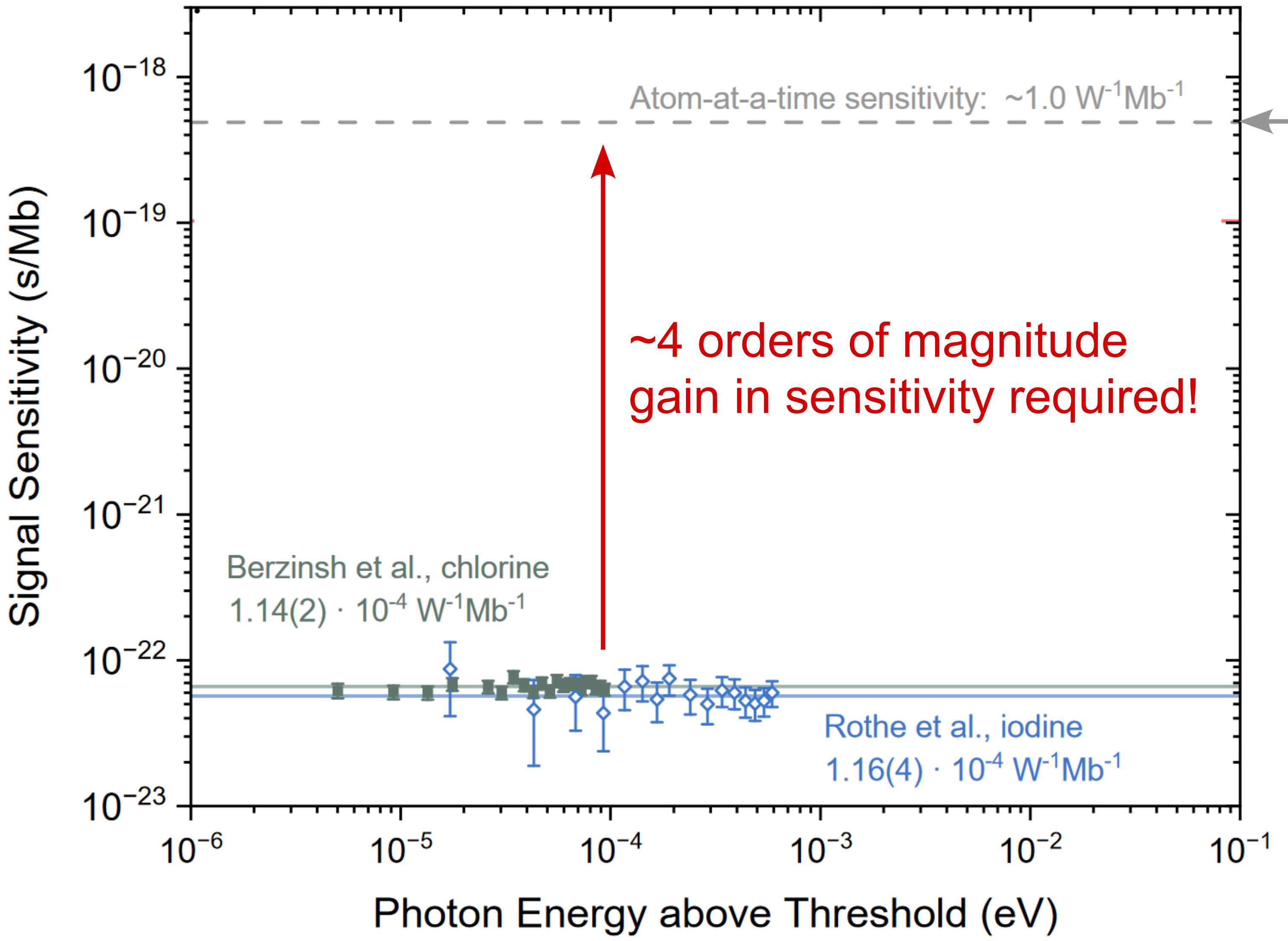


Extremely rare species require atom-at-a-time sensitivity!

Therefore....

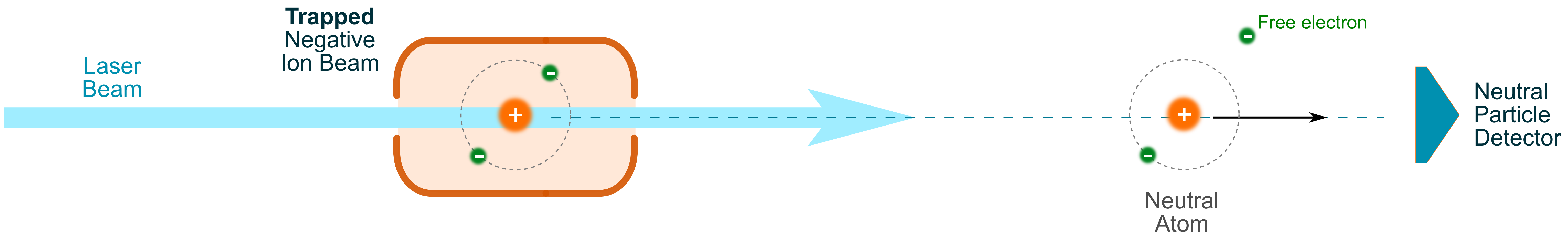
$$N_{ev}/N_{ion} \approx 1 \qquad \Gamma = \frac{N_{ev}}{N_{ion} \cdot \Phi \cdot \sigma}$$

at a reasonable target precision above threshold
(here we take 0.001 eV, or $\sigma \approx 1 \text{ Mb}$)

at a technologically attainable laser power
(here we take 1 W)

The Sensitivity Gap - Our Solution

Increase ion exposure to lasers by confinement



Ions are confined in an ion trap

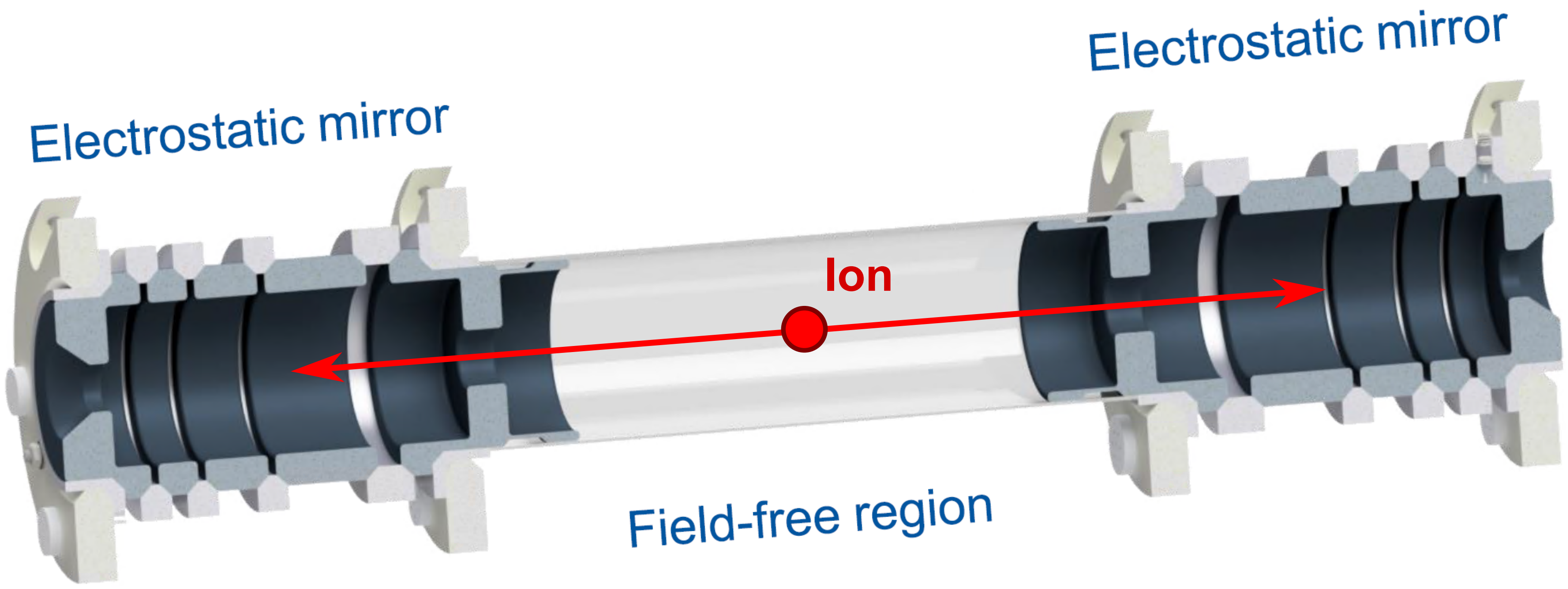
Particles stay exposed to laser for much longer time - requires less particles for same signal

Expected gain of orders of magnitude in sensitivity

... but storage medium must direct neutralized atoms towards detector

The Sensitivity Gap - Our Solution

Our trap of choice: MR-TOFs



Multiple-Reflection Time-of-Flight devices

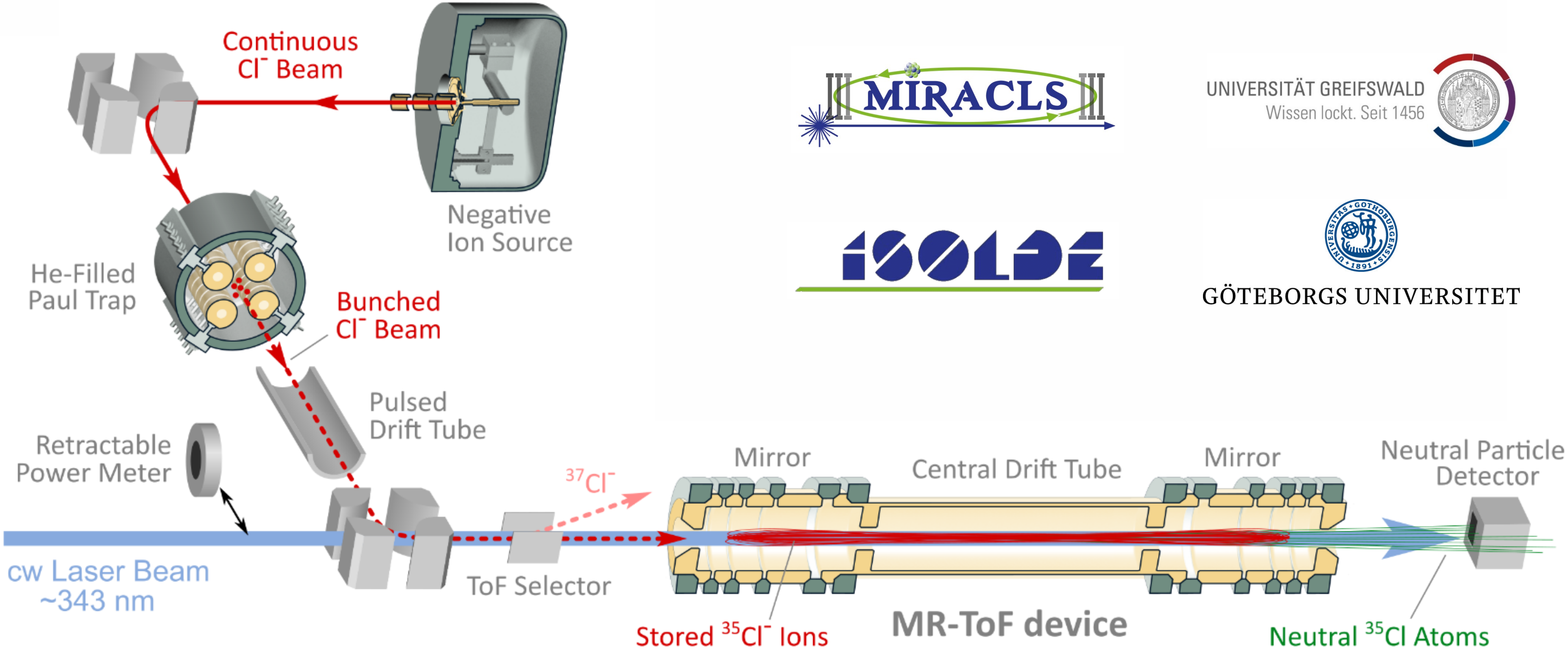
Widely used in rare isotope sciences as high precision **mass spectrometers** for over a decade

Expertise on their use to increase sensitivity of several laser spectroscopy techniques with

MIRACLS

The Sensitivity Gap - Our Solution

Proof-of-Concept at CERN-ISOLDE with MIRACLS



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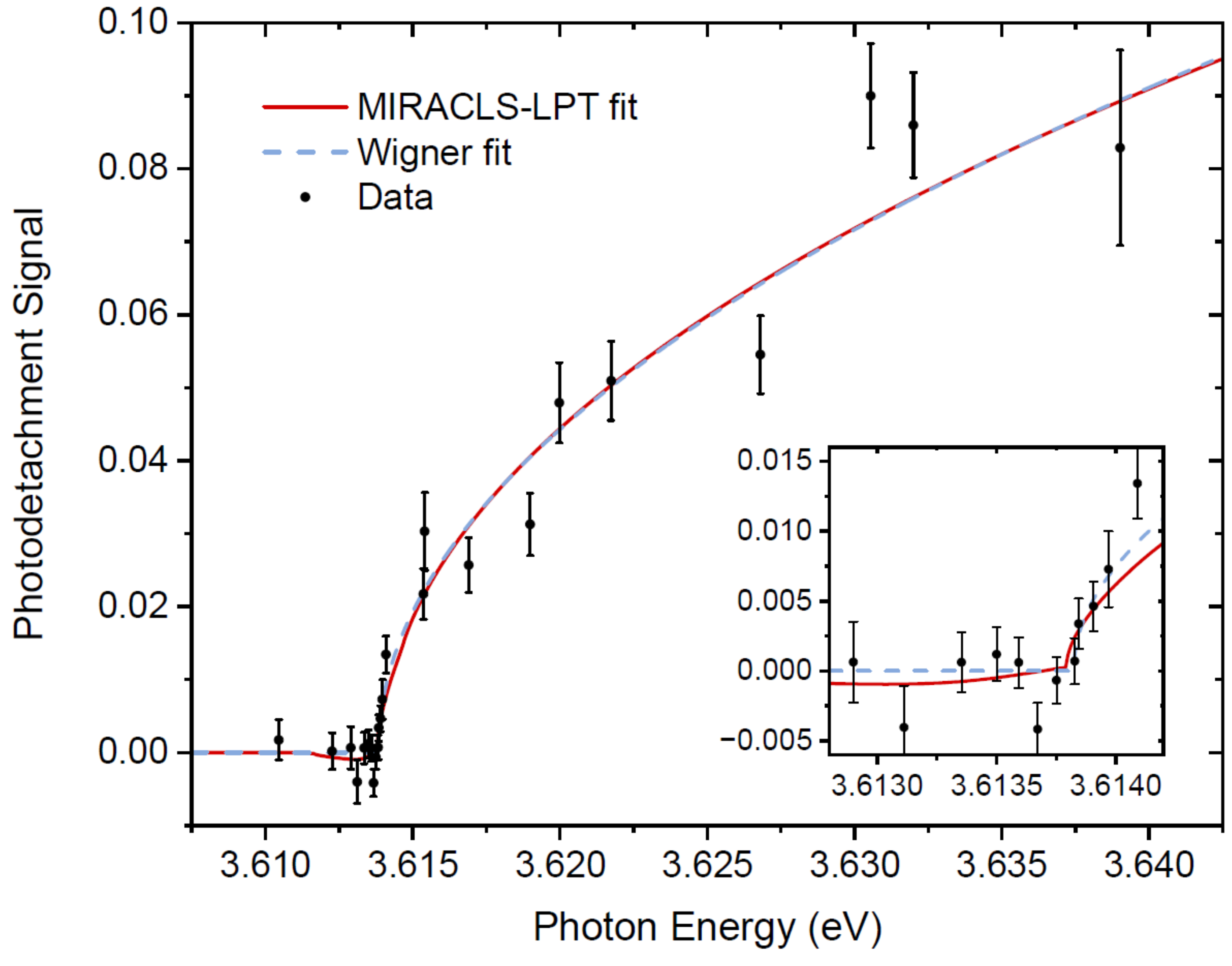

(non-Doppler corrected)

Re-evaluated EA of ^{35}Cl:

	MIRACLS	Berzinsh et al.*
EA (eV)	3.612720(44)	3.612726(27)
Ion rate (pps)	~3000	~10^9
Avg. laser power (mW)	~2.0	~13.0
Ion-laser exposure	520 ms 60k "passes"	few μs single pass

* U. Berzinsh, et al. Phys. Rev. A 51, 231 (1995)

The Sensitivity Gap - Our Solution

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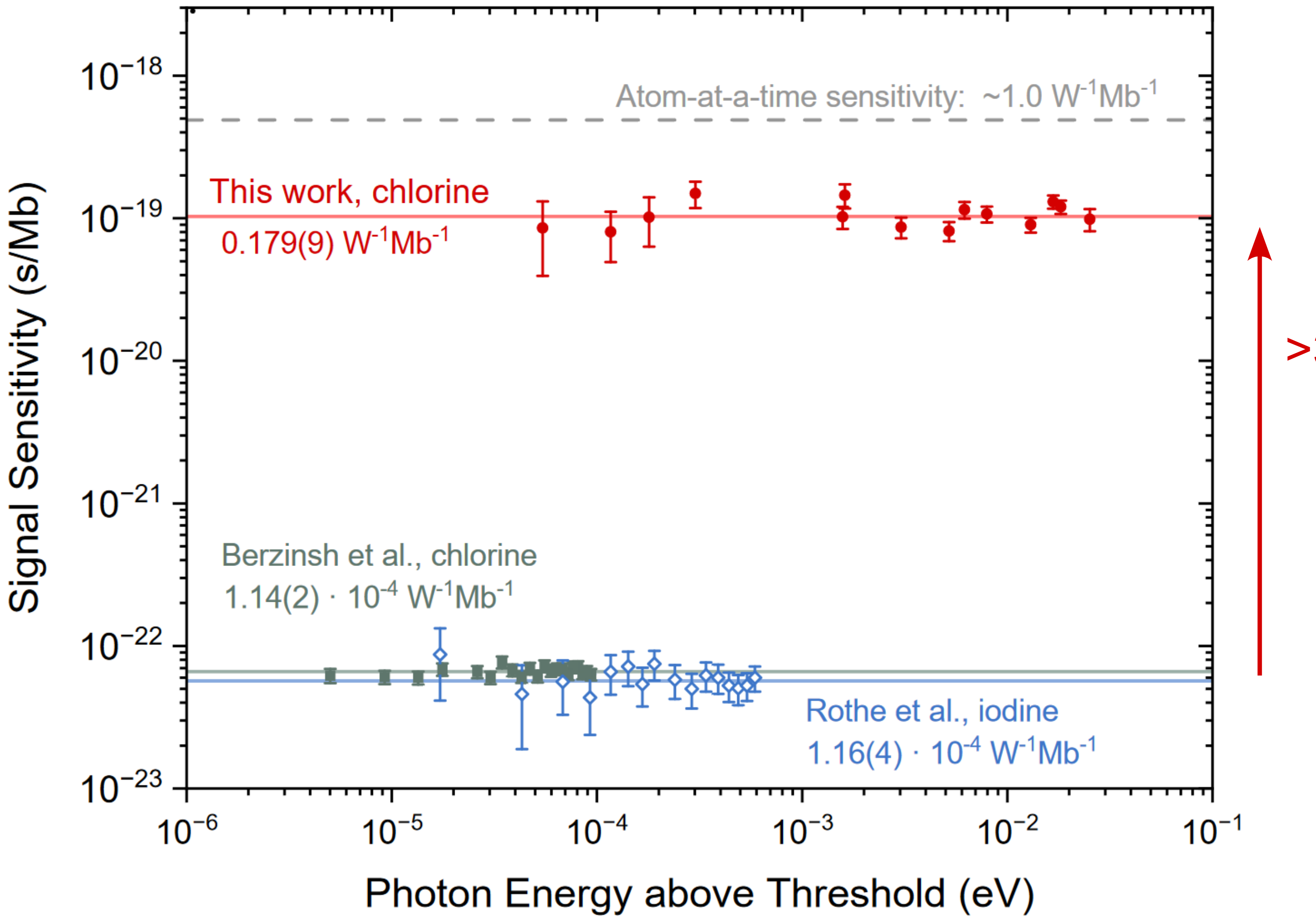

>3 orders of magnitude gain in sensitivity!

The Sensitivity Gap - Our Solution

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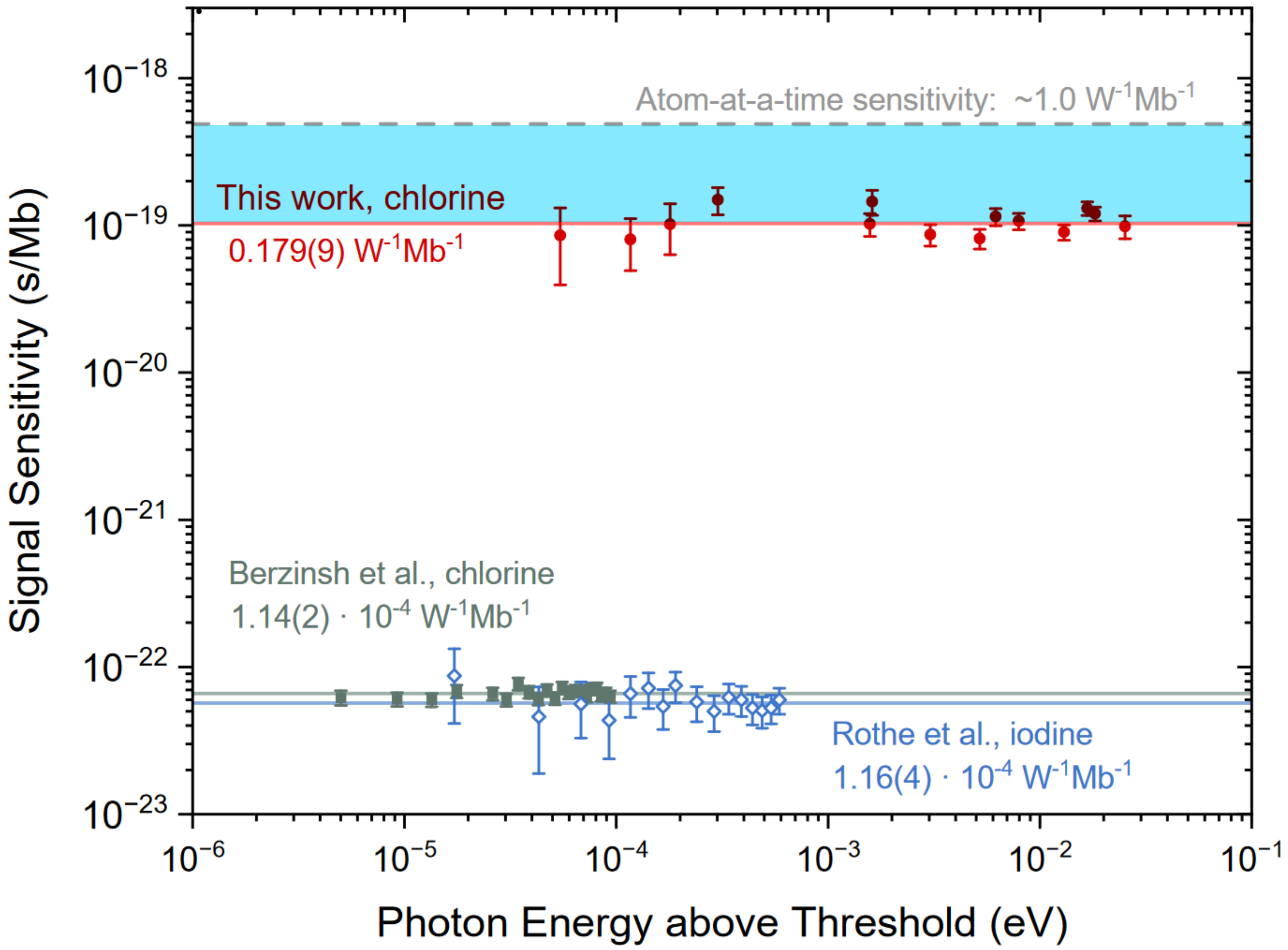


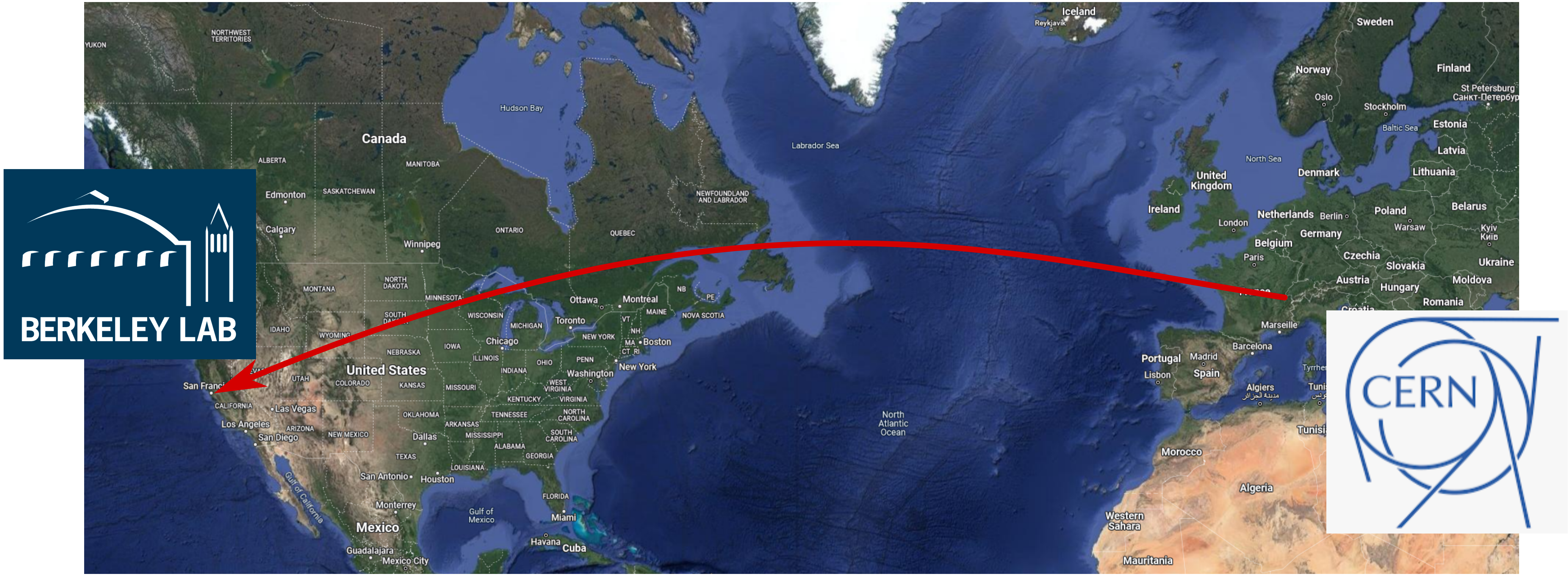
Detector employed detected about **1.5%** of all photodetachment events.

Mostly due to geometry...
- Detector displaced to avoid direct laser incidence
- Detected atoms only at one side of the MR-TOF

... and quantum efficiency:
- Detector not suitable to detect low-energy atoms

Towards AETHER...

MIRACLS Proof-of-Principle MR-TOF was sent to LBNL to seed the new infrastructure



AETHER's Concept

Create sample of element → Prepare sample for spectroscopy → Create negative Ion → Probe Sample → Detect Signal

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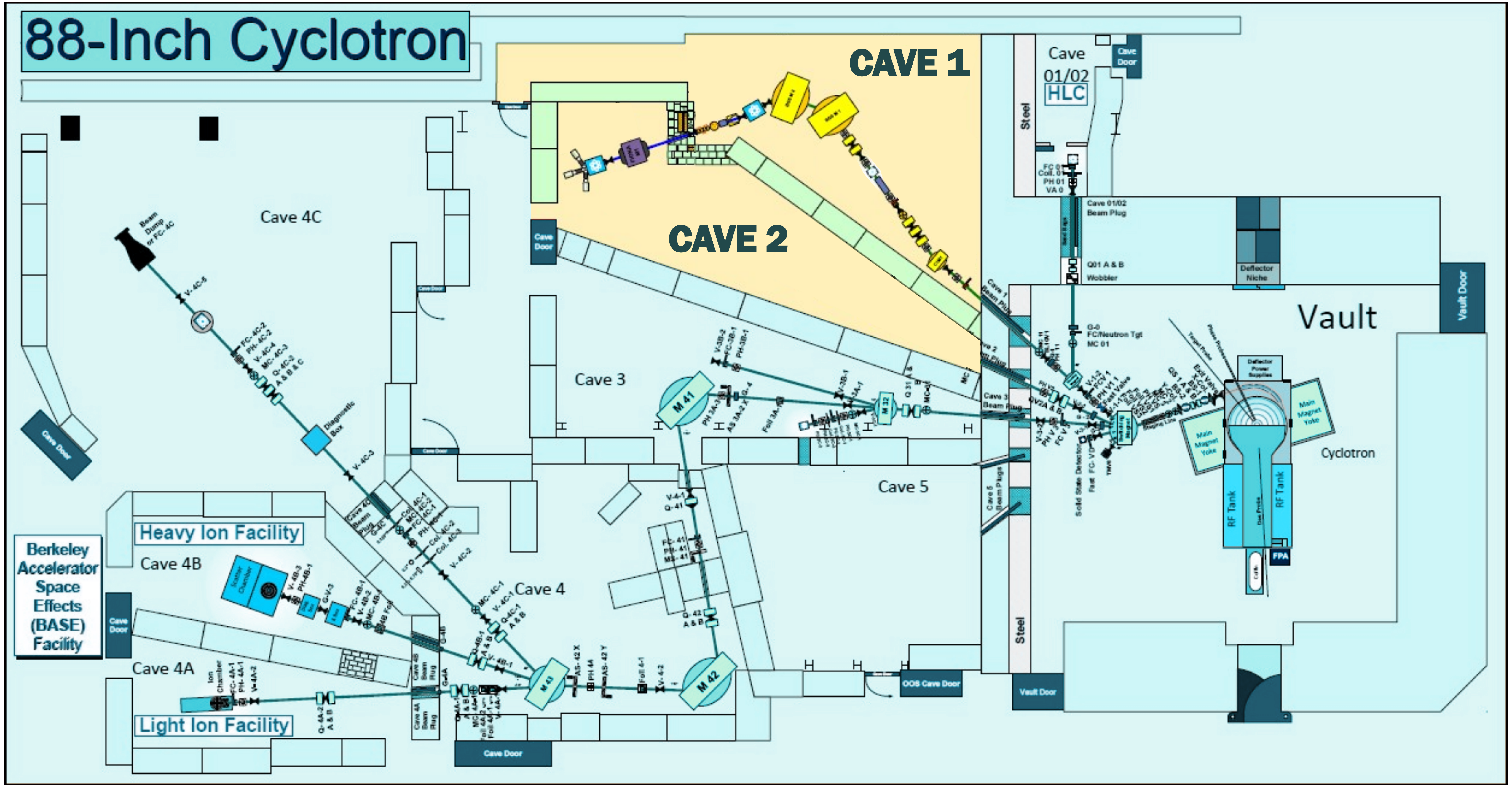

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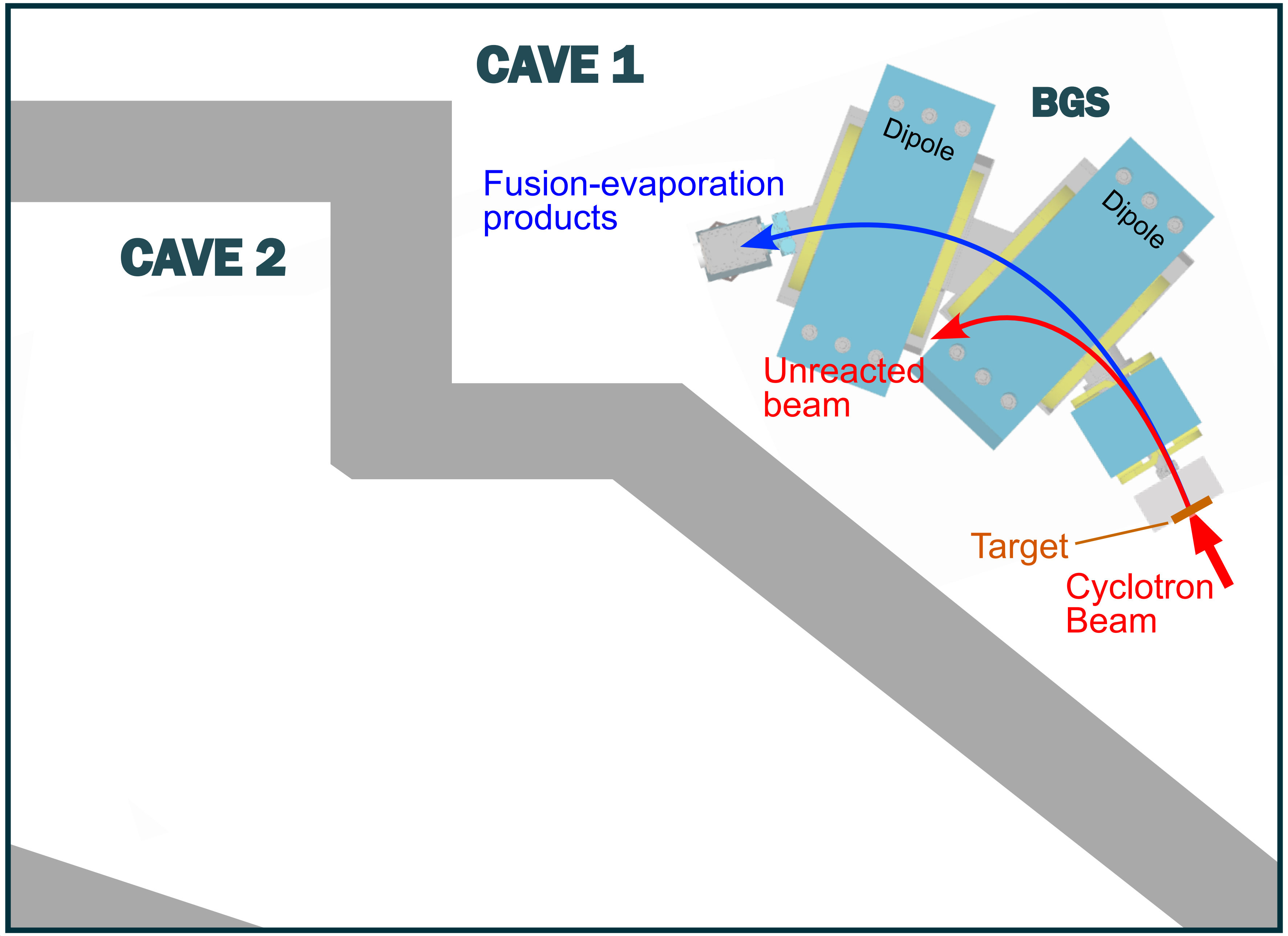

Artificial production of nuclides by fusion-evaporation nuclear reactions

Efficient separation at **Berkeley Gas-filled Separator**

Low production: few samples per second at best

One of very few systems in the world capable of producing ALL of the elements whose EA is unknown

AETHER's Concept

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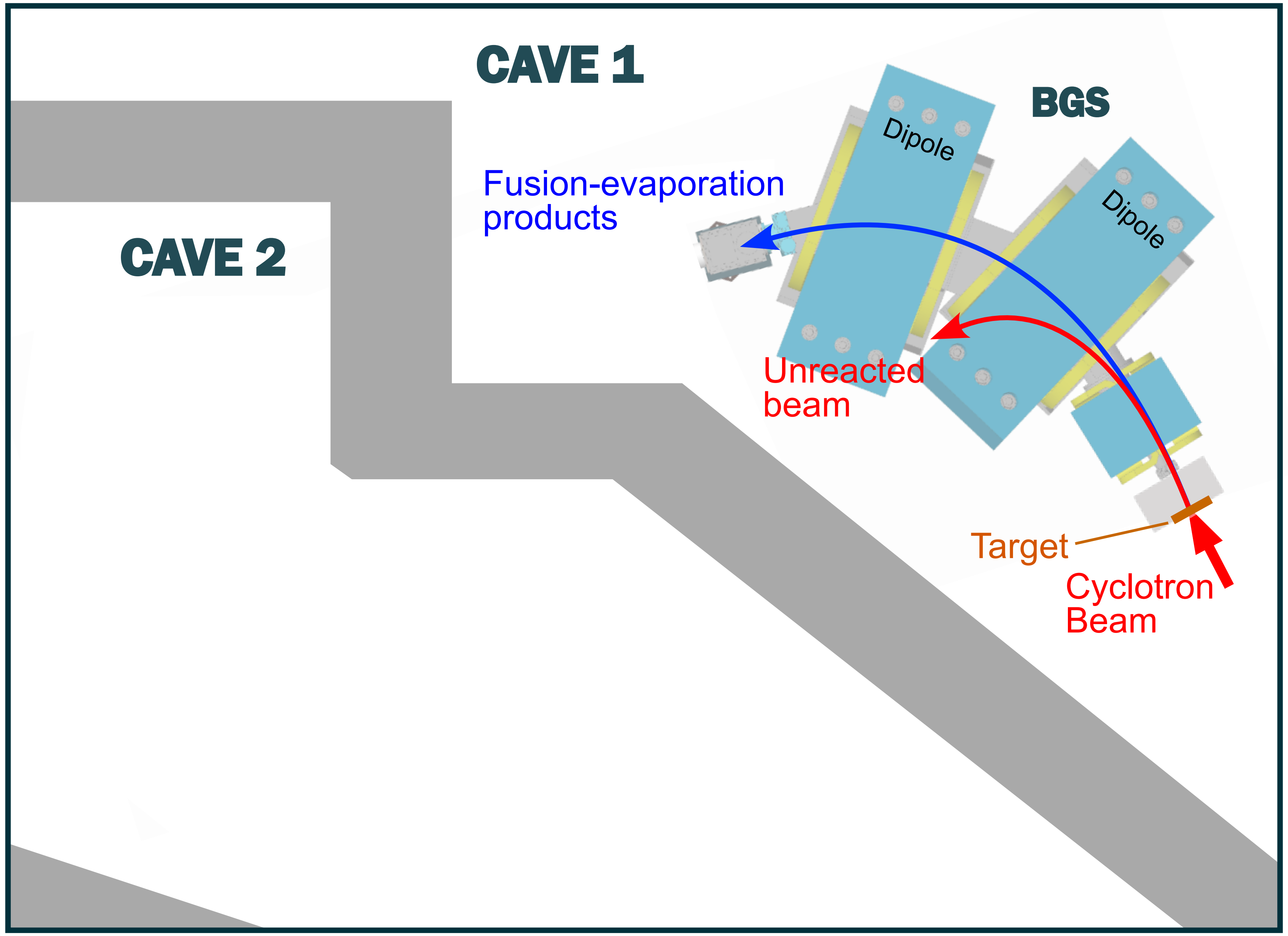

Ions out of BGS are highly energetic and disperse

Spectroscopy experiments require cold (low emitance), low energy ion bunches

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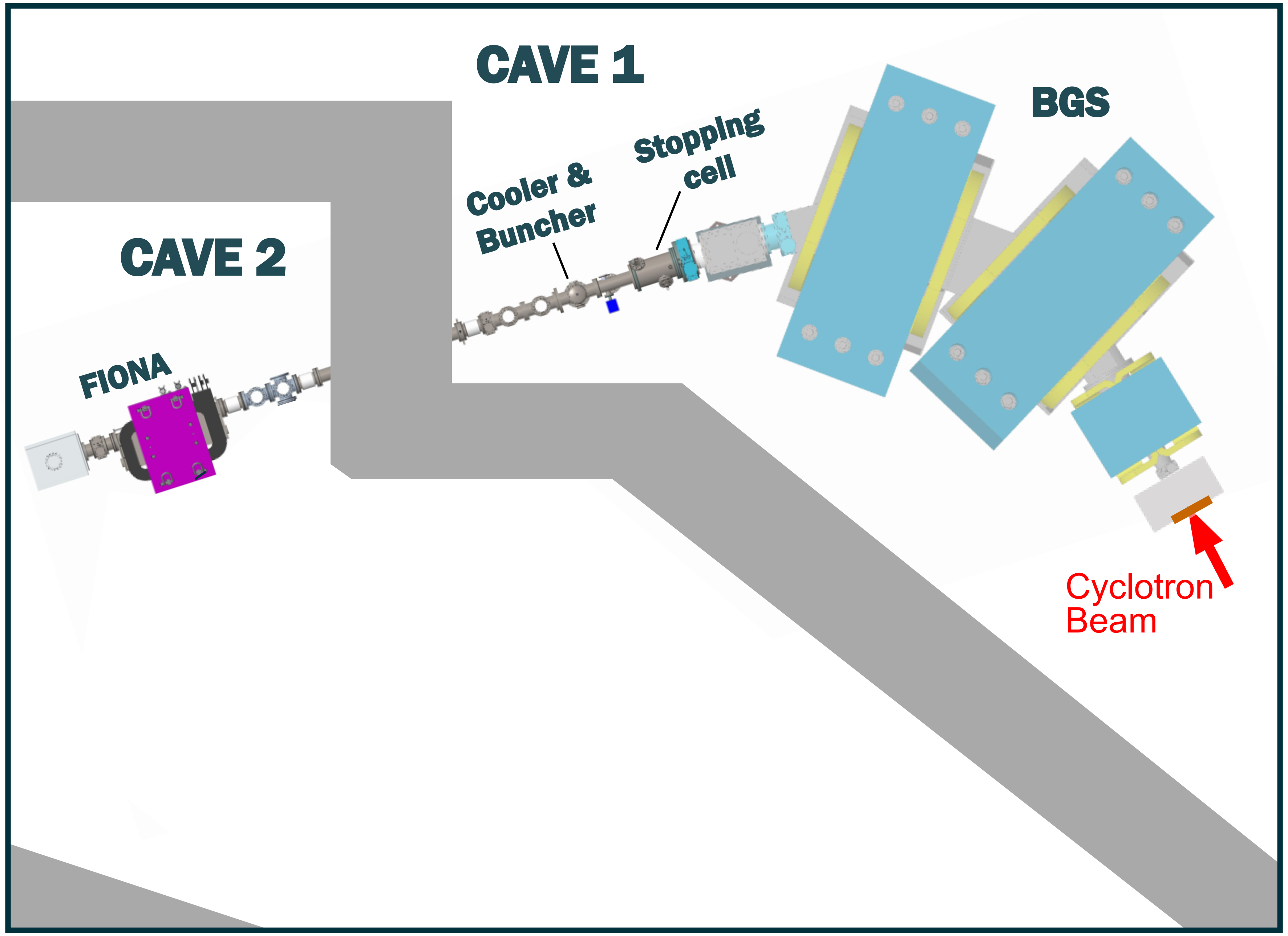

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Spectroscopy experiments require cold (low emitance), low energy ion bunches

FIONA mass spectrometer also require such properties!

A stopping cell and a cooler & buncher are already in place

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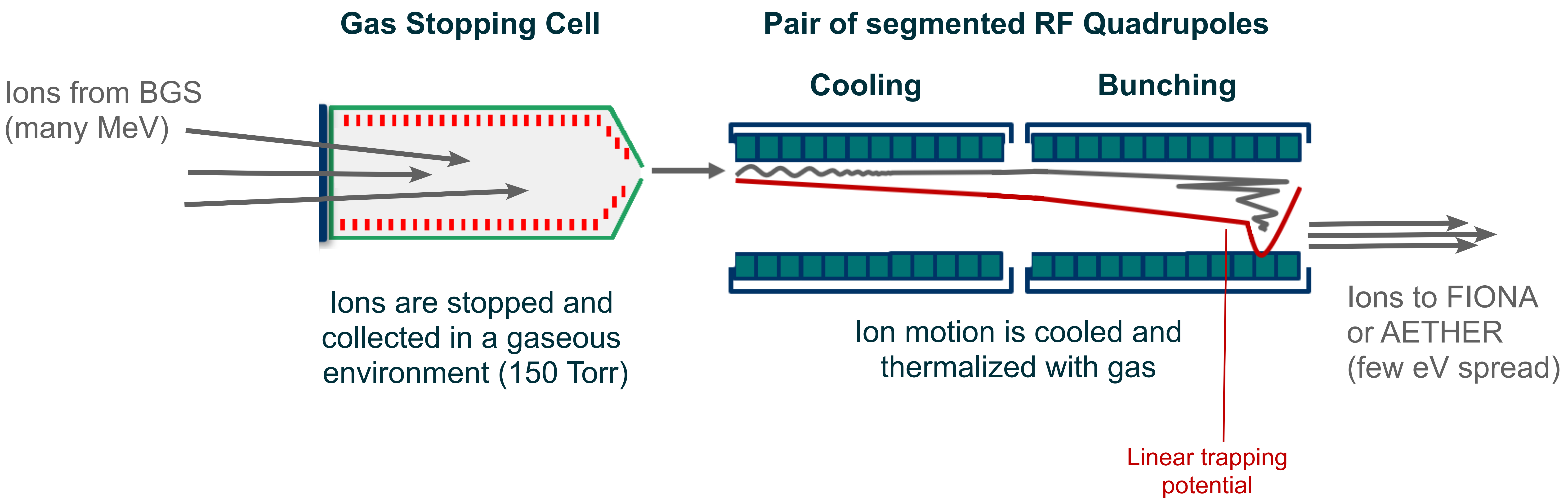

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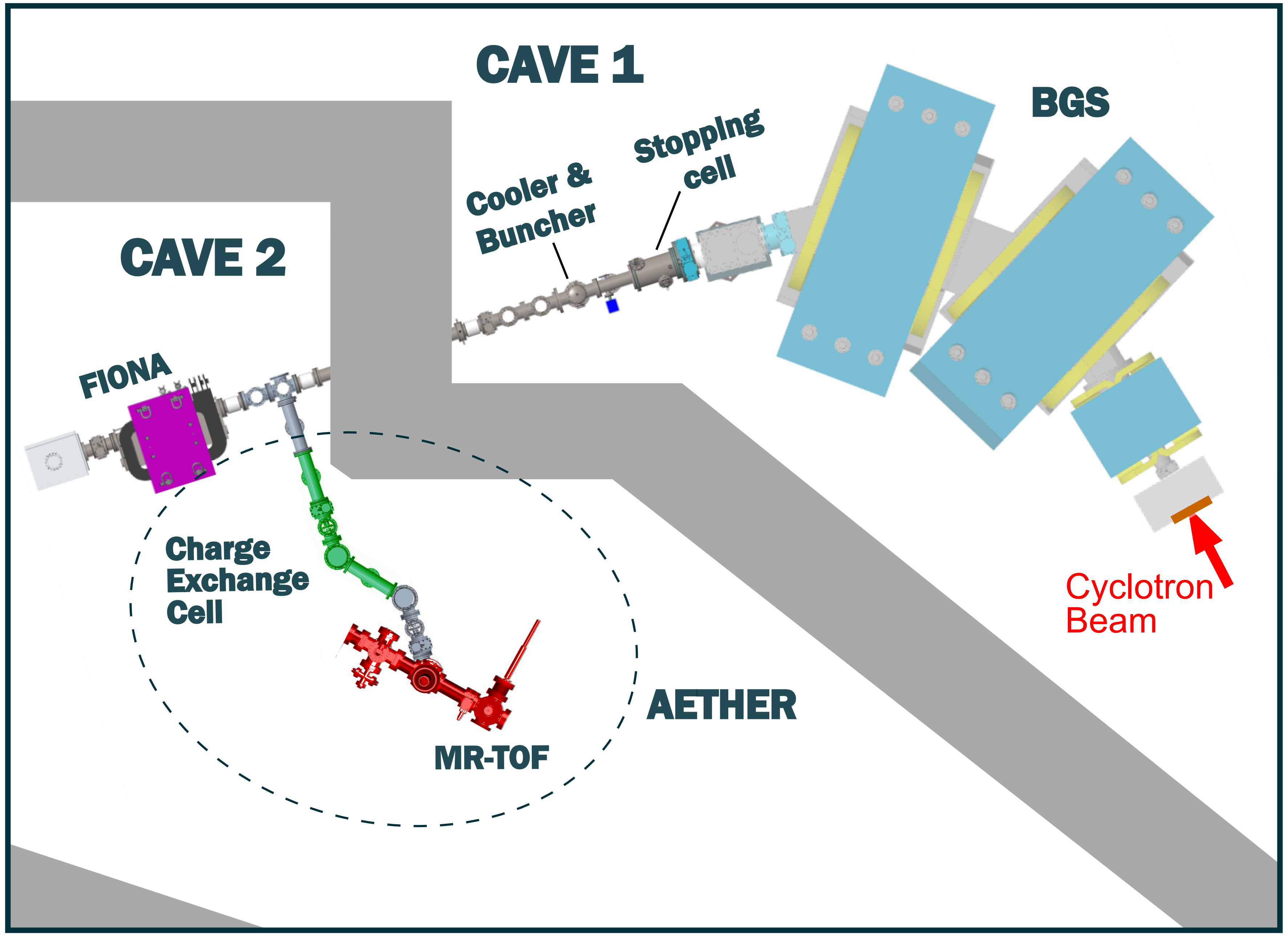
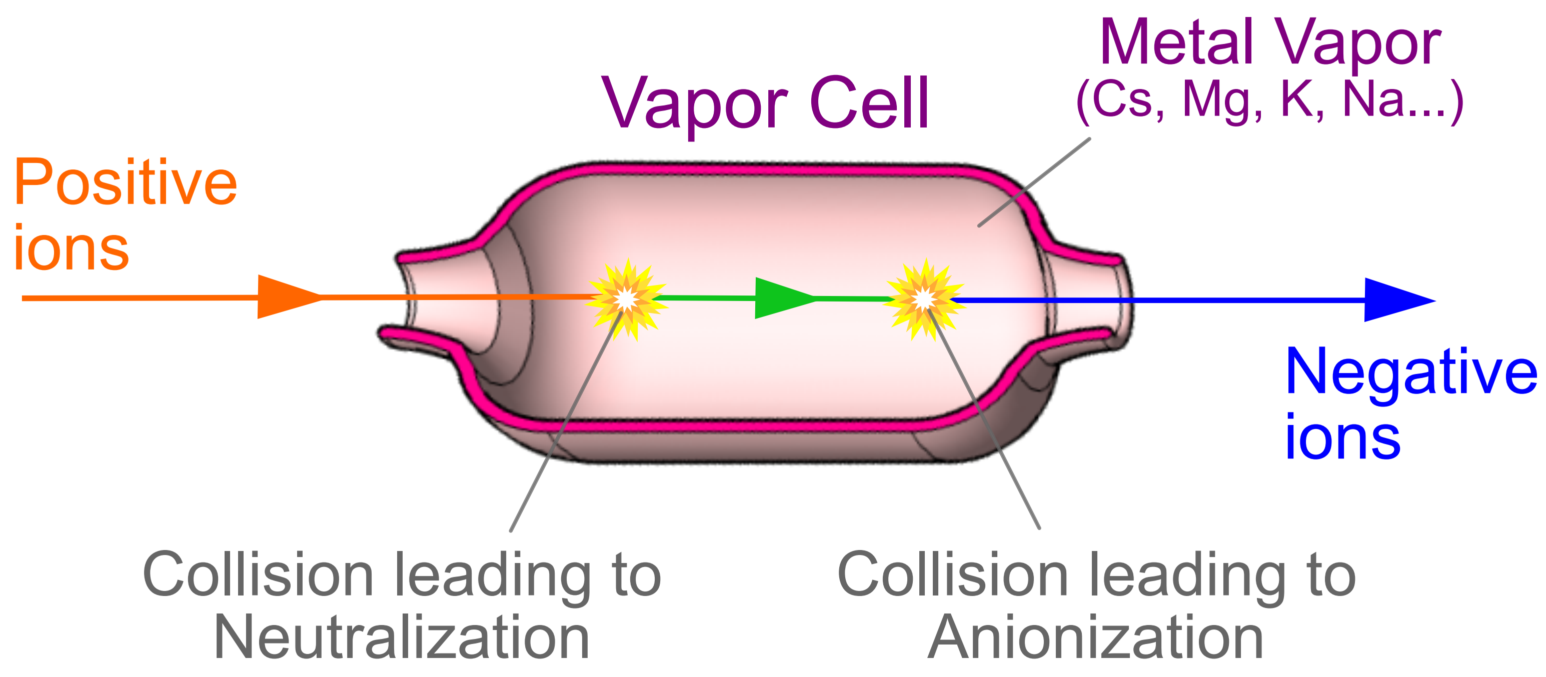


AETHER's Concept

Create sample of element → Prepare sample for spectroscopy → **Create negative Ion** → Probe Sample → Detect Signal

Double charge-exchange with metal vapors:



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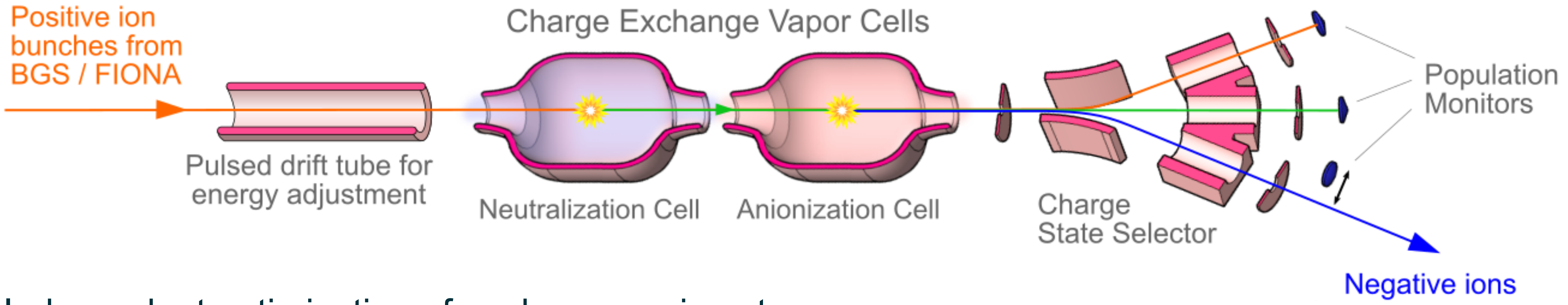
Create sample of element → **Prepare sample for spectroscopy** → **Create negative Ion** → **Probe Sample** → **Detect Signal**

Double-cell approach:



- Independent optimization of each conversion step
- Full parameter-space flexibility

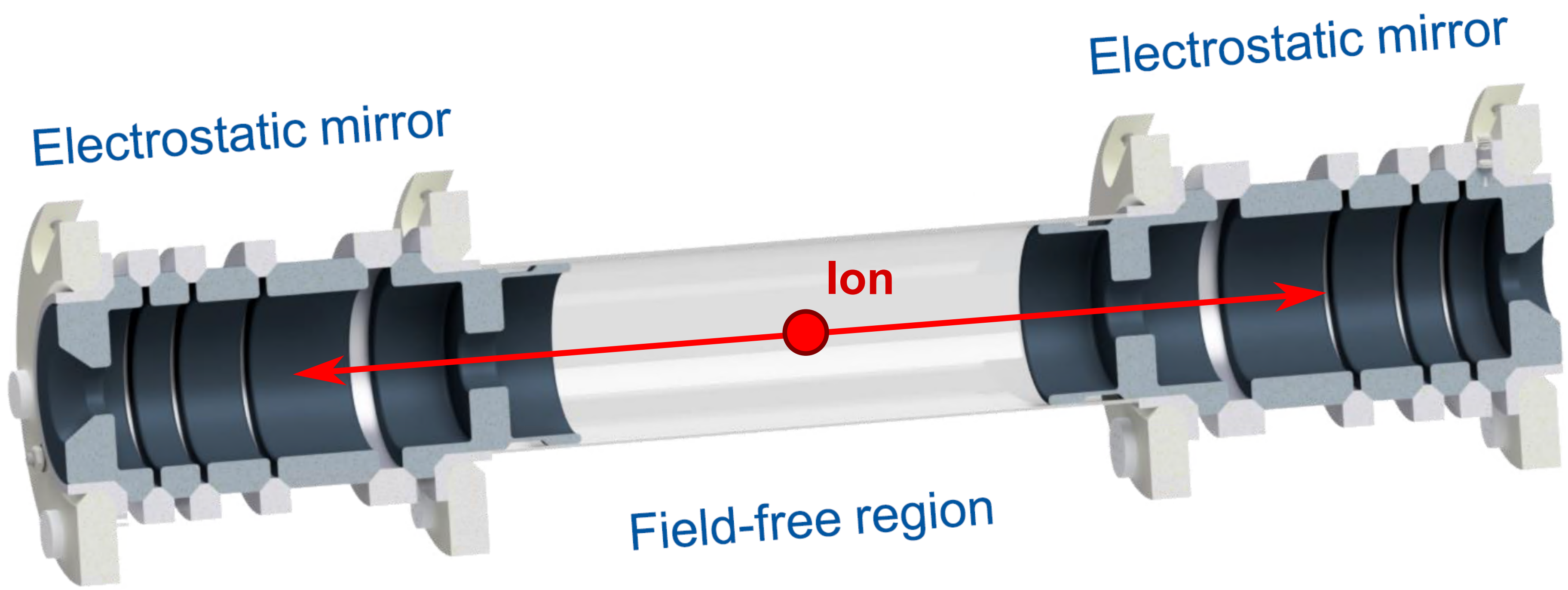
- Theory support from Dr. Remi Cabrera Trujillo (UNAM) for <u>charge exchange cross sections</u>

Bonus: indirect determination of EA and atomic radii!

AETHER's Concept

Create sample of element → Prepare sample for spectroscopy → Create negative Ion → Probe Sample → Detect Signal

First cases: MIRACLS PoP spectrometer



CERN

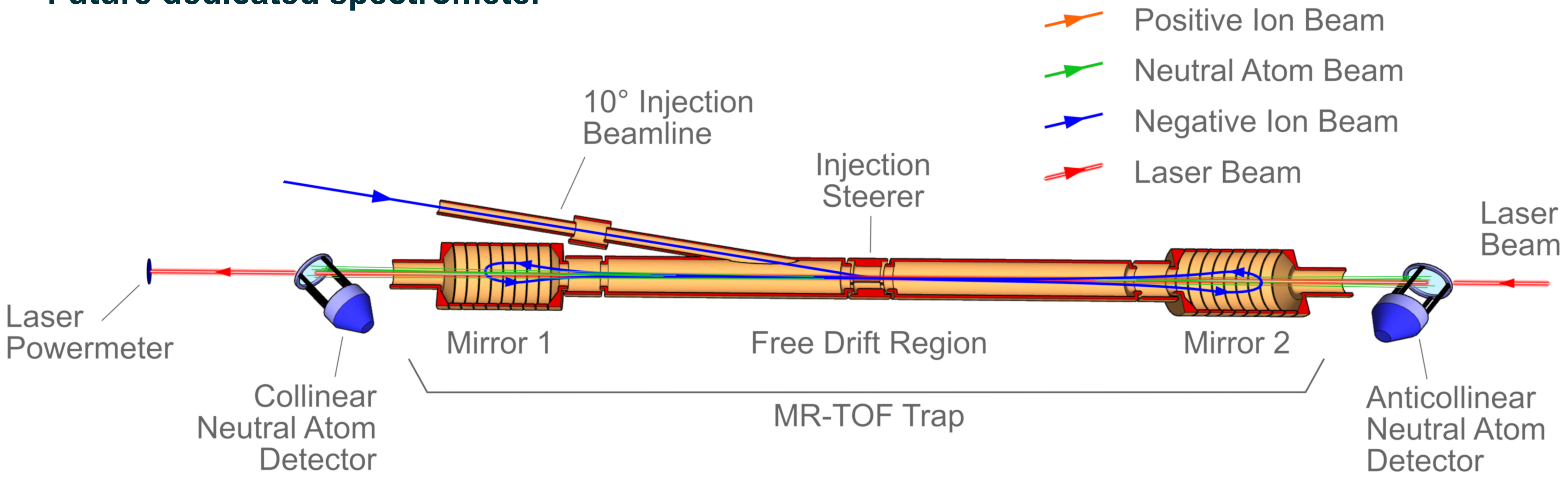
MIRACLS

UNIVERSITÄT GREIFSWALD
Wissen lockt. Seit 1456

AETHER's Concept

Create sample of element → Prepare sample for spectroscopy → Create negative Ion → **Probe Sample** → Detect Signal

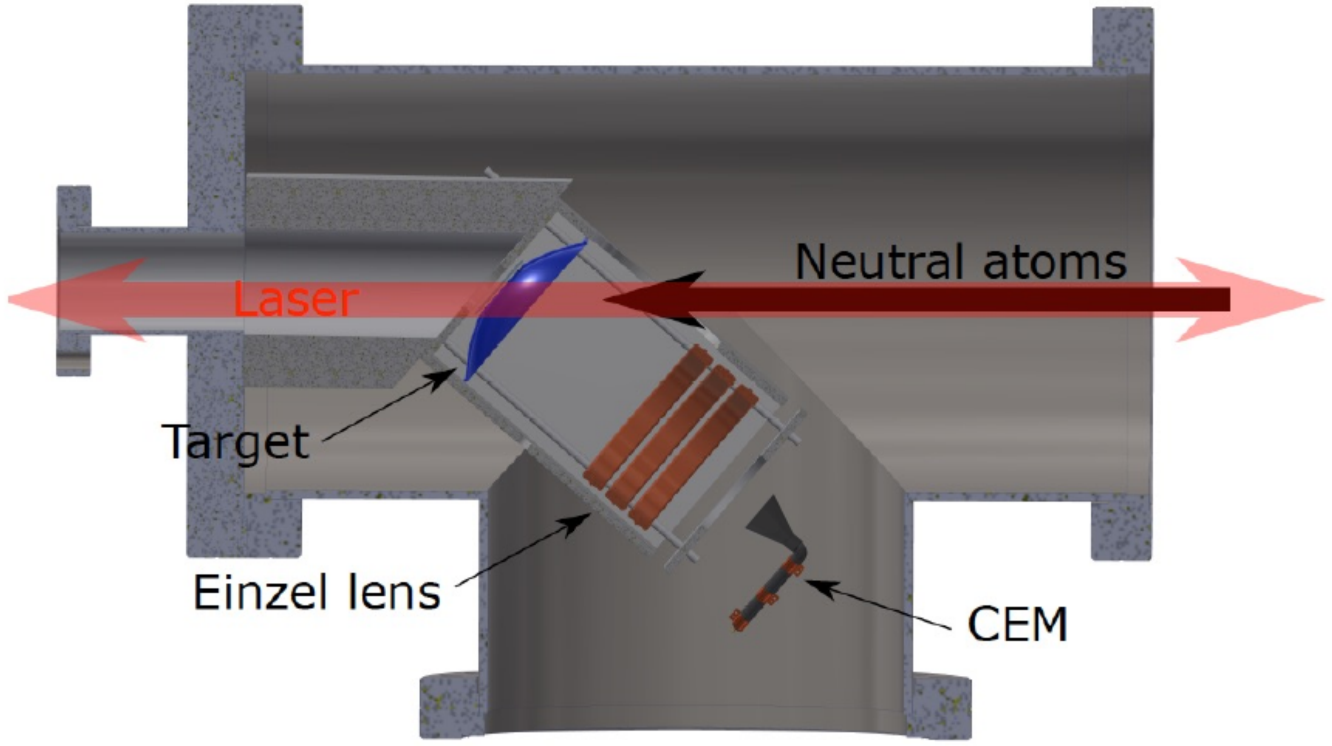
Future dedicated spectrometer



AETHER's Concept

Create sample of element → Prepare sample for spectroscopy → Create negative Ion → Probe Sample → Detect Signal

Transparent Neutral Particle Detector



J. Warbinek et al. Appl. Phys. Lett. 114, 061902 (2019)

GANDALPH

GÖTEBORGS UNIVERSITET

State-of-the-art detectors are crucial for required sensitivity!

- Large area
- Transparent to laser (graphene impact plate)
- CW-laser friendly - low photoelectron emission
- Built at University of Gothemburg

To be developed:

- Alpha-tagging capabilities
  (to be built from the work of E.M. Lykiardopoulou)

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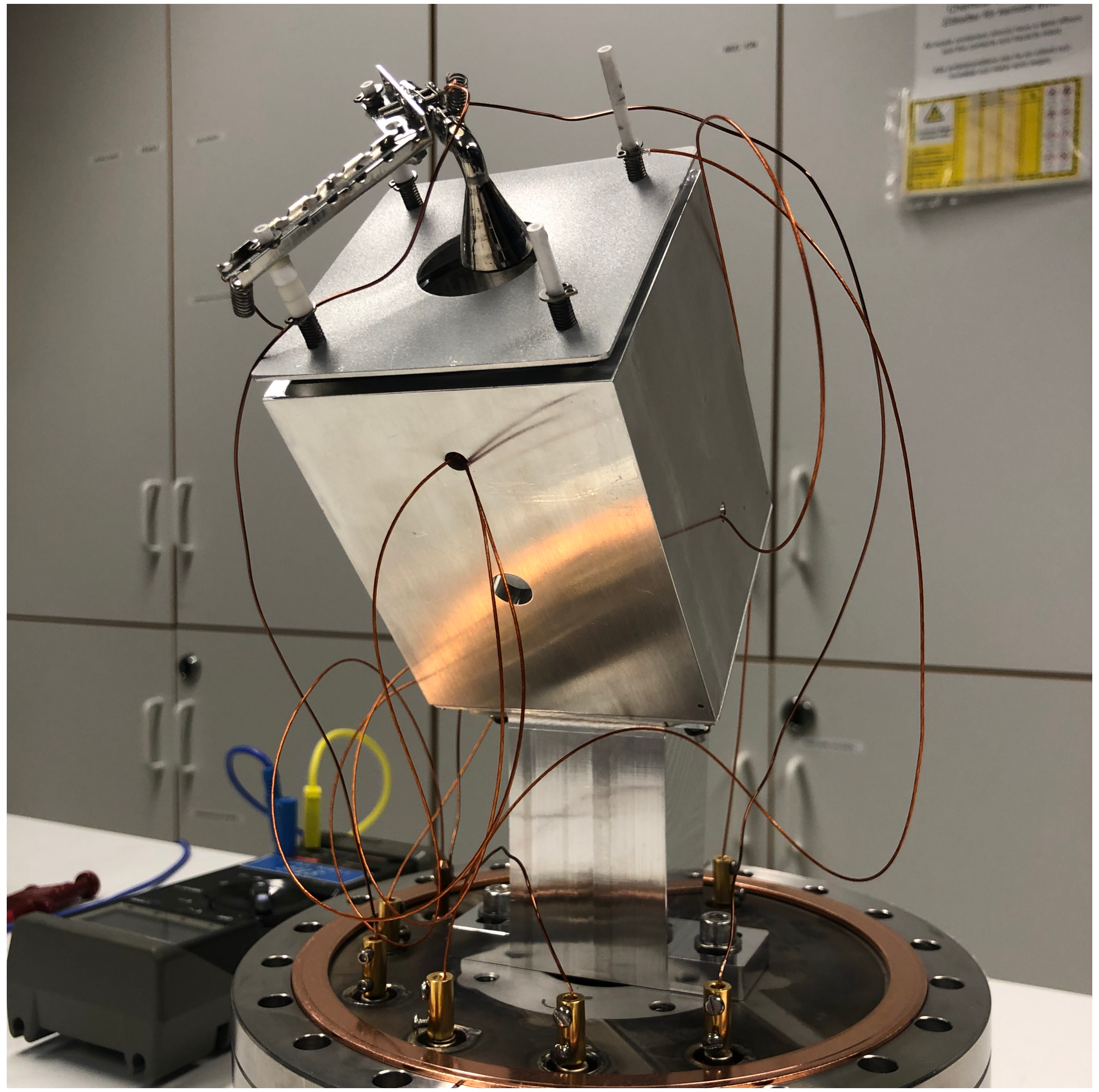
Transparent Neutral Particle Detector



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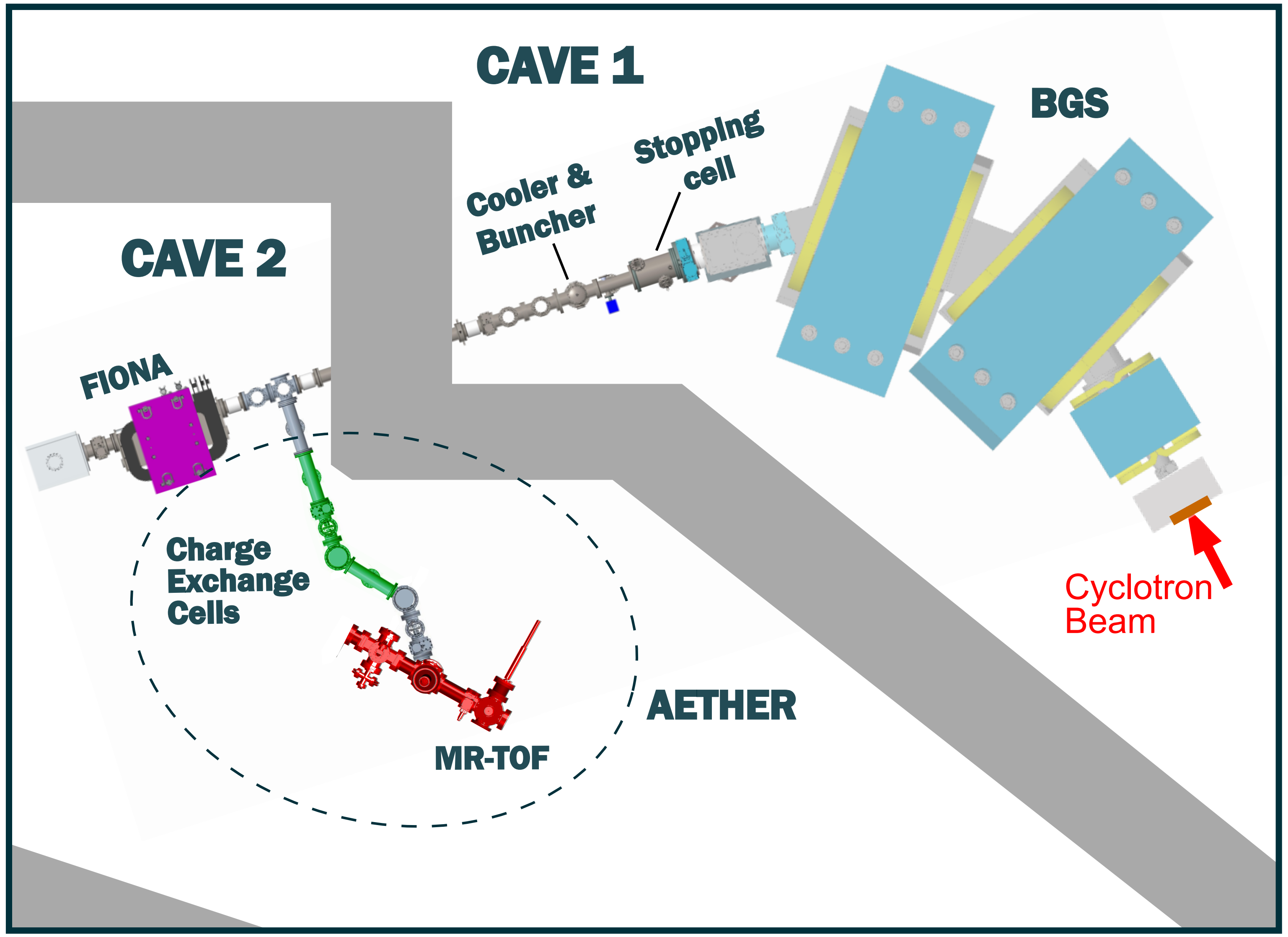
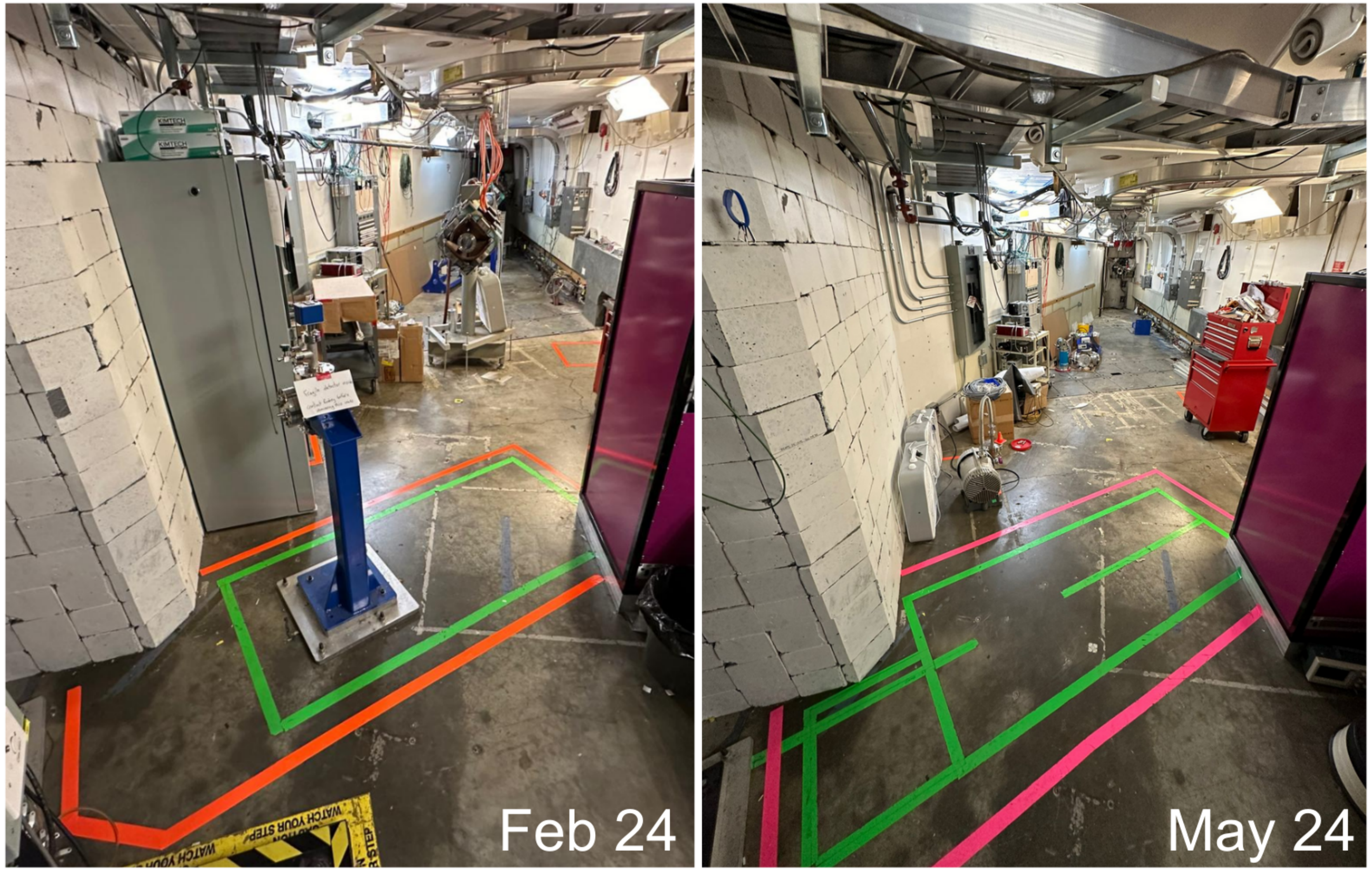
AETHER's Status

Create sample of element	Prepare sample for spectroscopy	Create negative Ion	Probe Sample	Detect Signal
Ready!	Ready!	Securing funding	Spectrometer at LBL!	1 Detector ready
			Installation ongoing	Upgrades under dev.
			Laser system to be designed	1 copy to be built



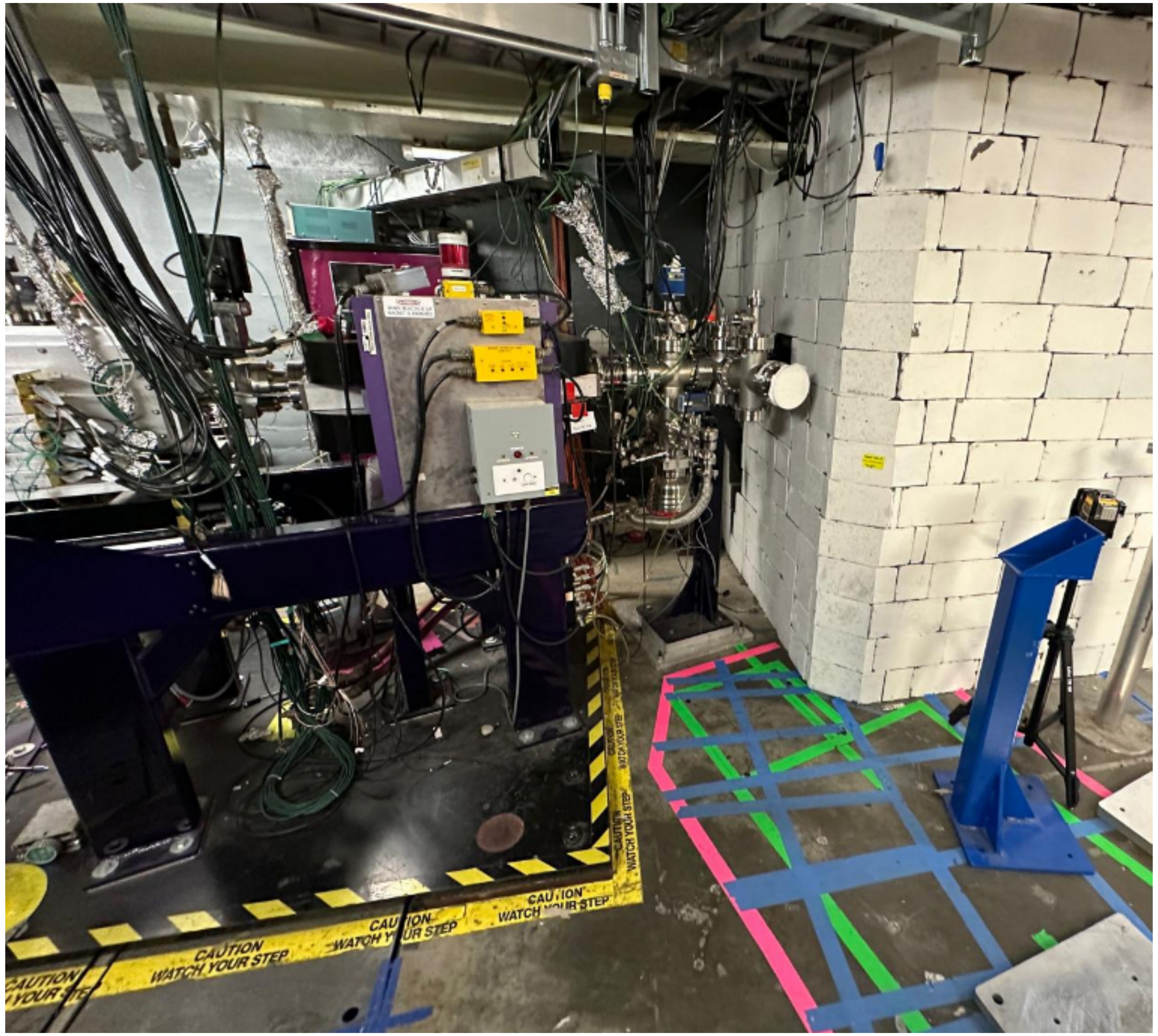
AETHER's Status

October 2023
MR-TOF arrives from CERN

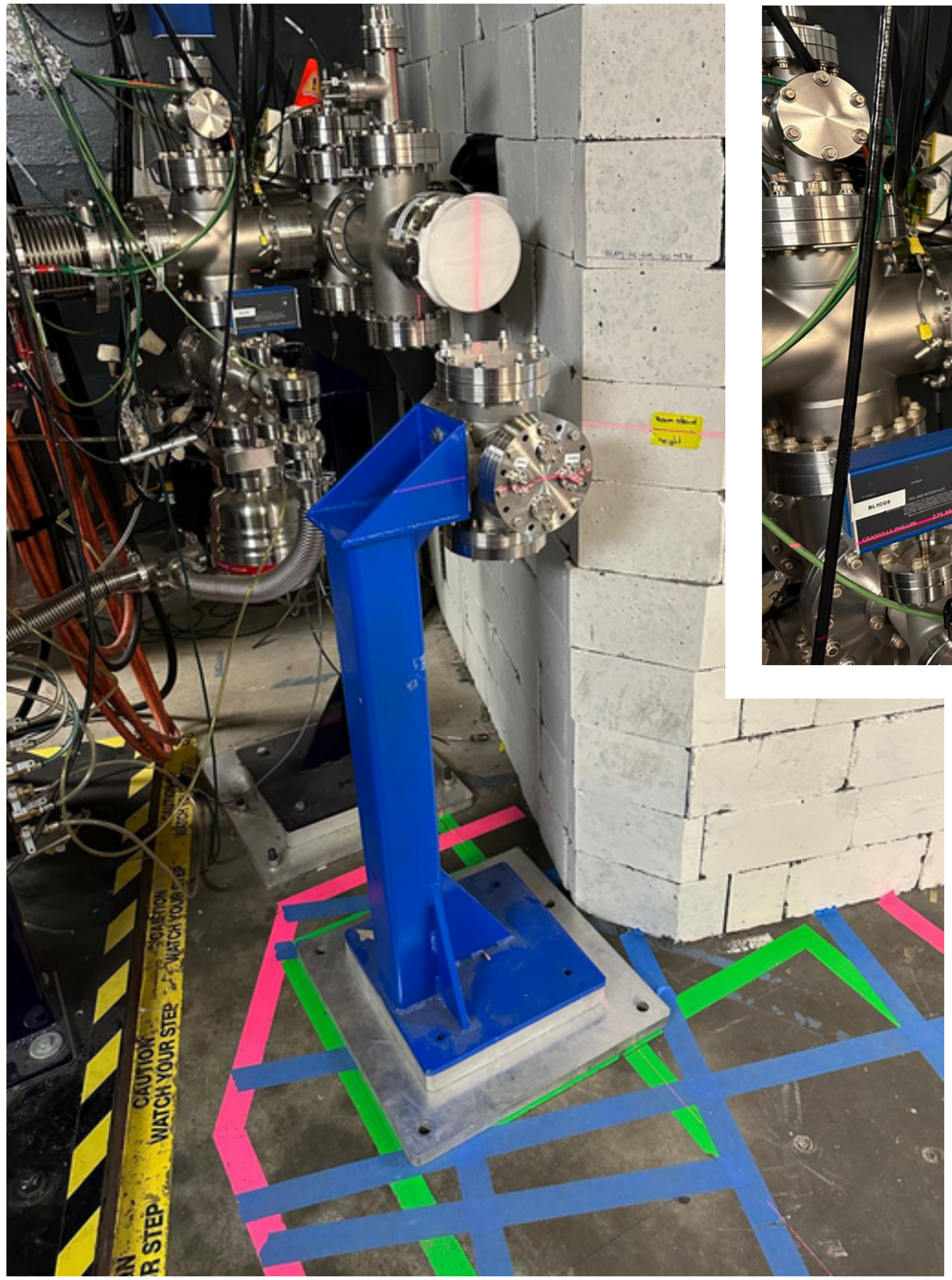


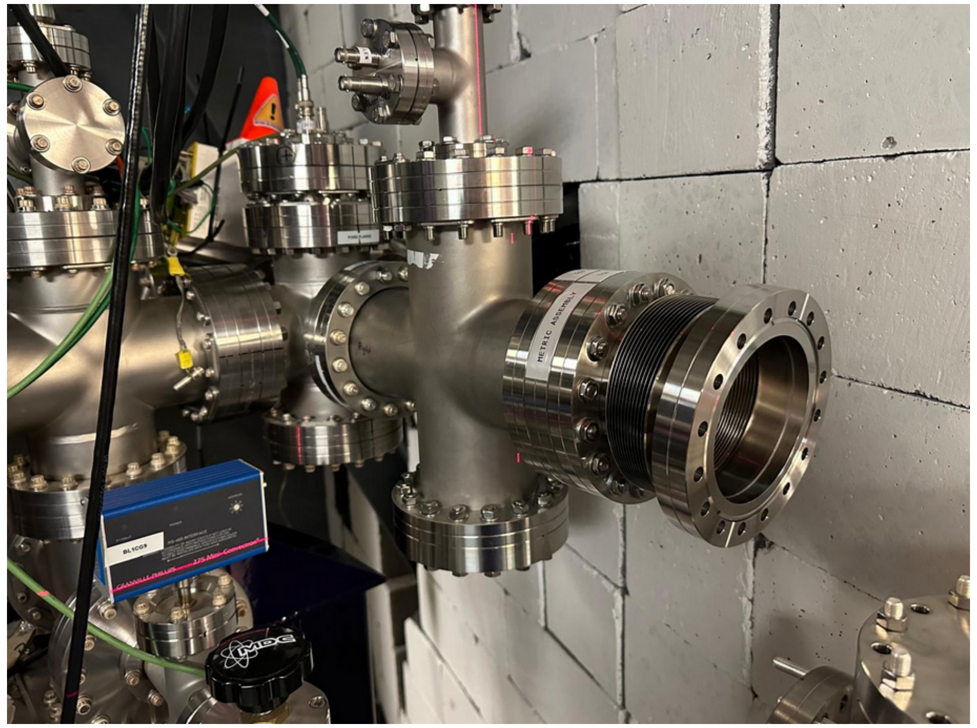
Ongoing in 2024
Cave 2 cleared from older beamline
Seismic provisions ready
FIONA equipment to be rearranged

AETHER's Status



Ongoing in 2024
Coupling beam line under development





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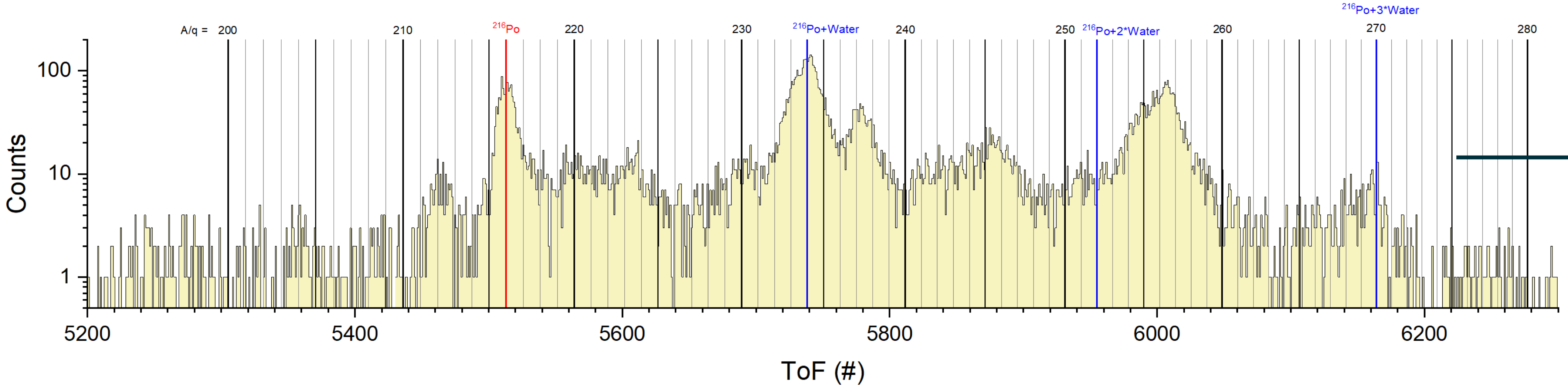
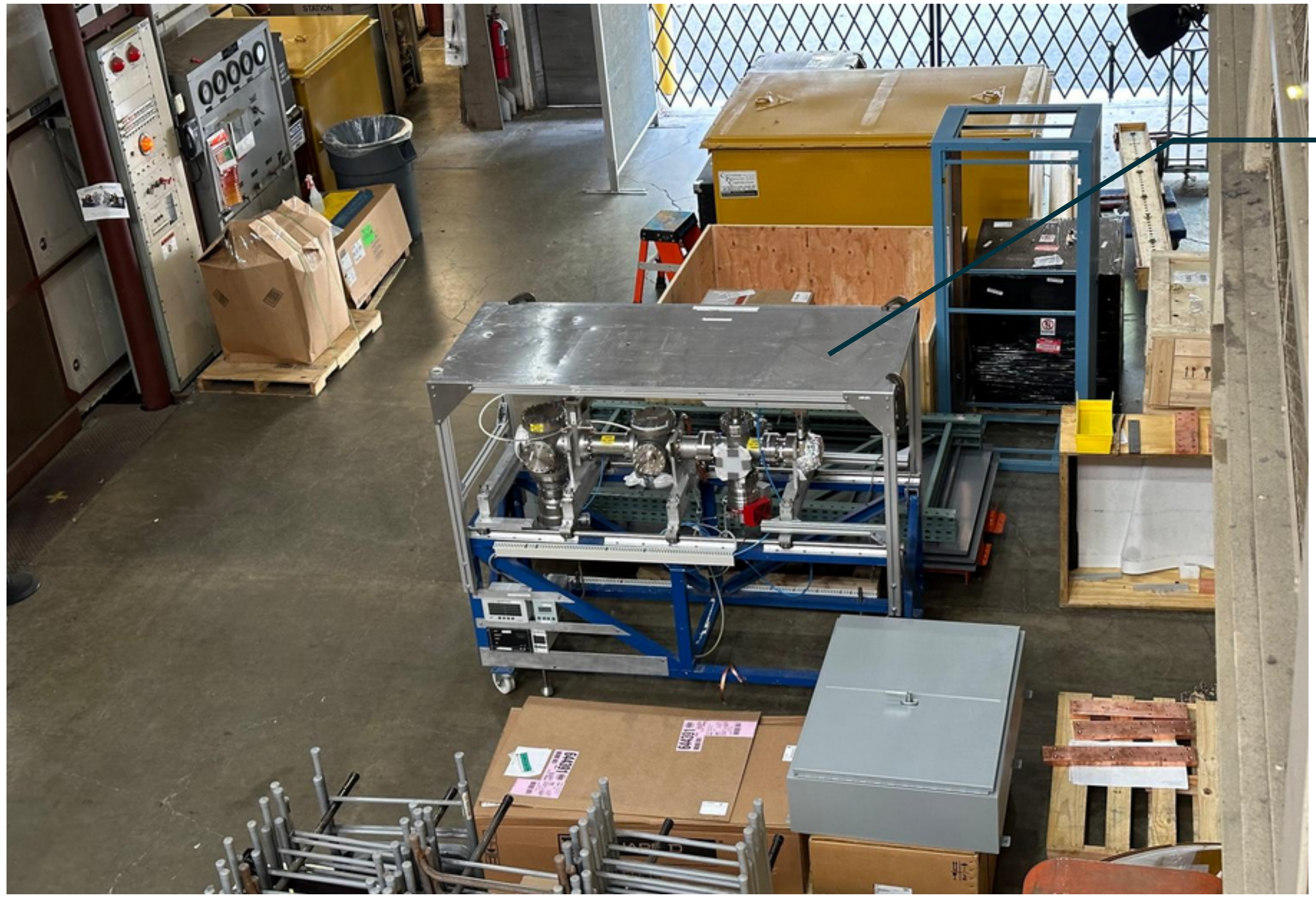
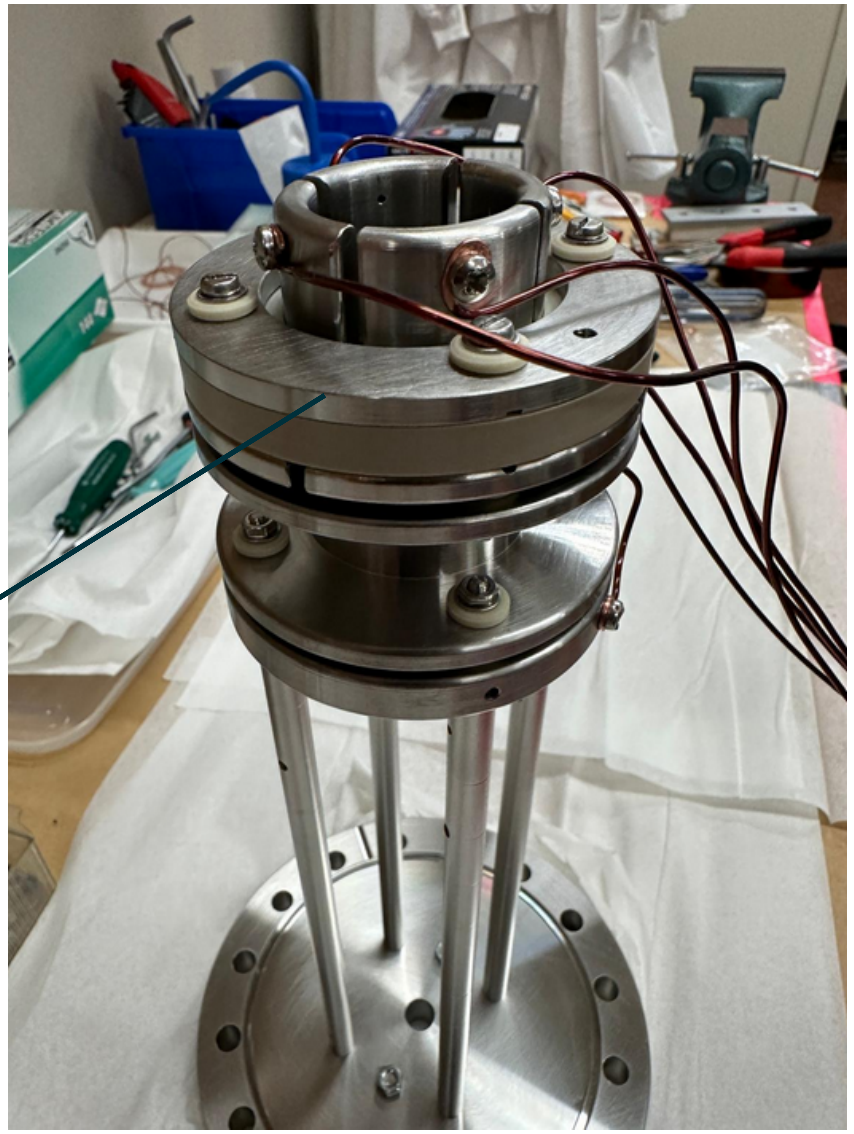


First ions in the new beam line segment
New DAQ comissioned



MR-TOF ready to be craned into Cave 2

Other modifications ongoing



AETHER's Status

Secured space for a laser lab right above Cave 2

Legacy laser system to serve as a starting point

AETHER's "Side Quest"

MR-TOFs are also superb TOF-MS mass spectrometers

Highly sensitive (up to ~1 count/day)
Precise/accurate to measure **nuclear binding energy** (~10s keV/c^2)
Fast (~50 ms measurement cycle)
Highly tolerant to sample contamination

Ideal for nuclear structure investigations of rare,
short-lived species!

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Precise/accurate to measure **nuclear binding energy** (~10s keV/c^2)
Fast (~50 ms measurement cycle)
Highly tolerant to sample contamination

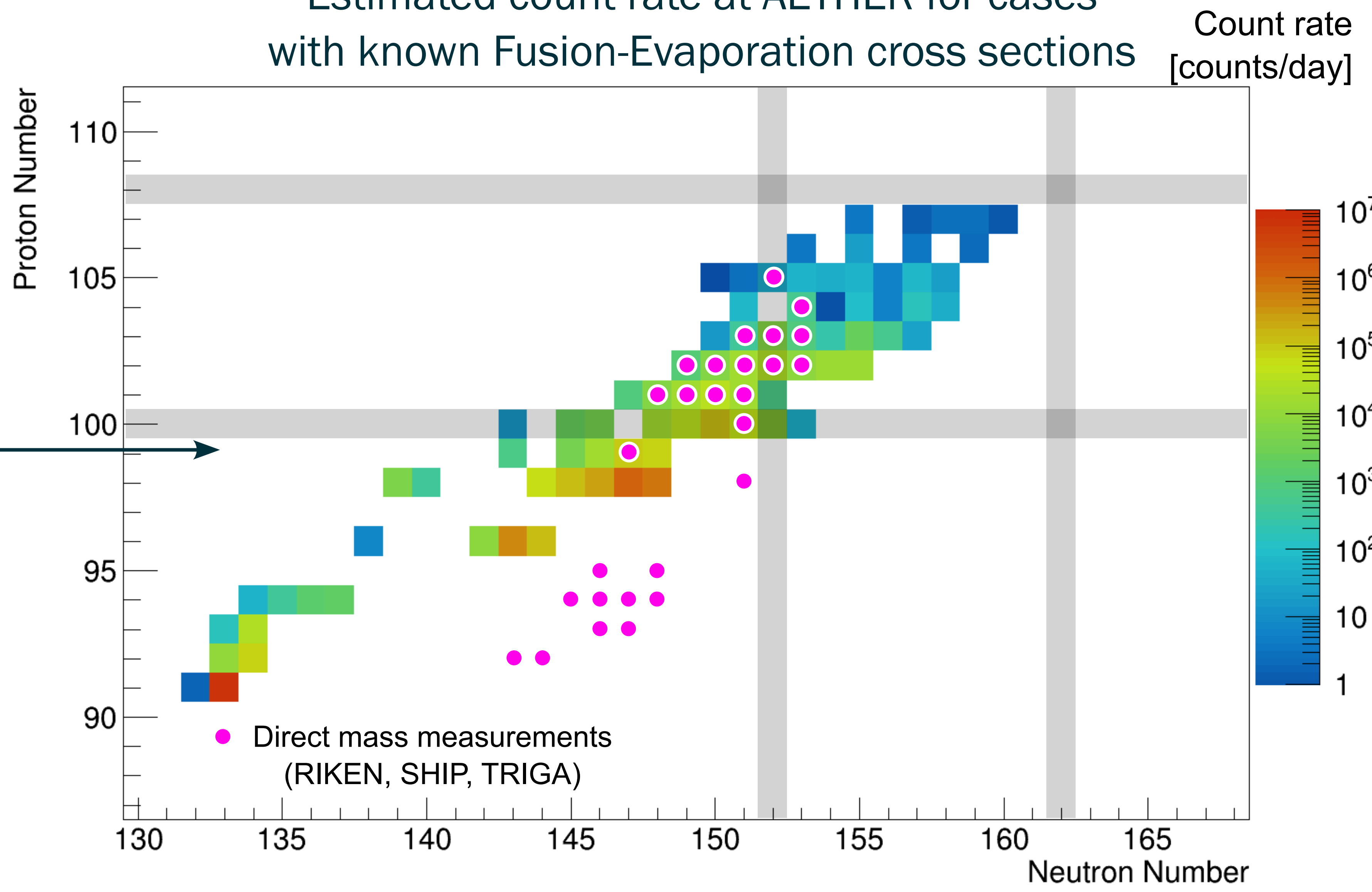
Ideal for nuclear structure investigations of rare, short-lived species!

Several unique opportunities with nuclides produced by the BGS

… specially with late actinides and SHE (e.g. deformed shell closures).

First online beam time mid/late 2025 (?)

Estimated count-rate at AETHER for cases with known Fusion-Evaporation cross sections



Outlook

1. Electron Affinities are fundamental observables to chemistry and atomic physics.

Provide insights into an element's reactivity and electron-electron correlations.

Experimental techniques lack sensitivity to measure EAs of rare elements.

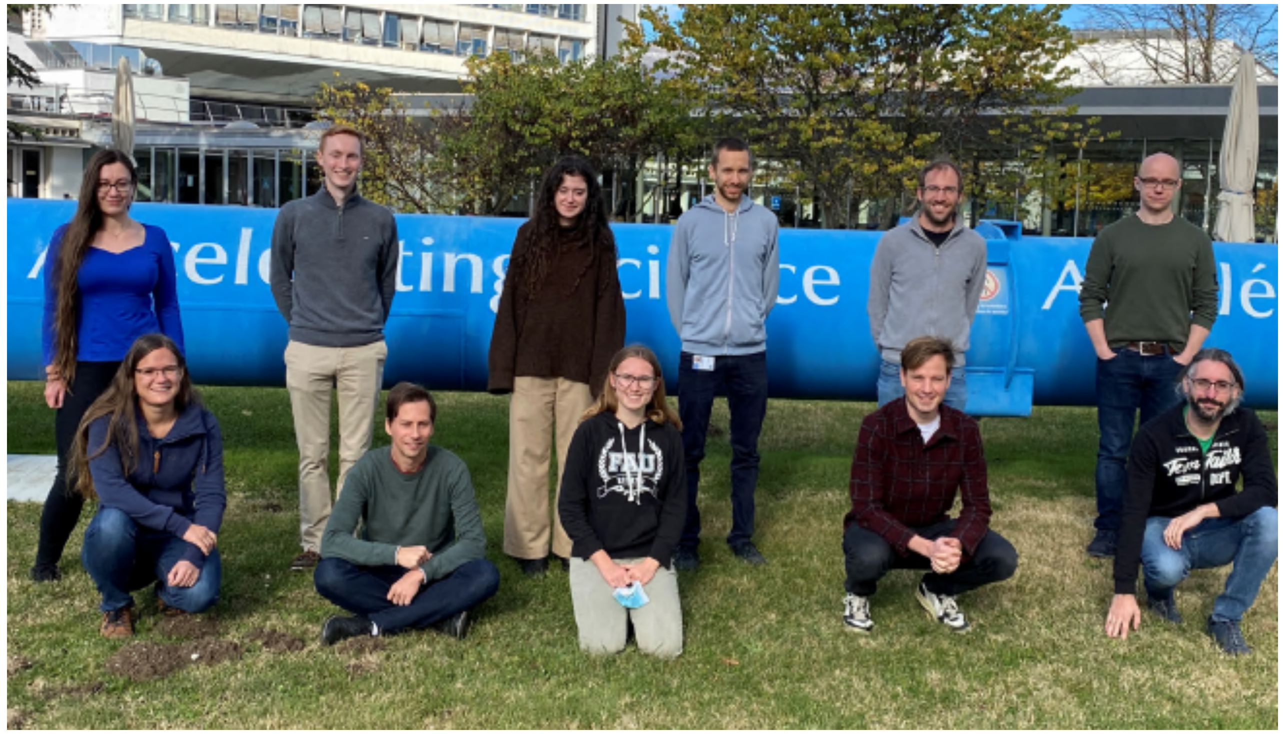
2. AETHER will be a dedicated setup at LBNL to measure the rarest EAs in the Periodic Table.

Atom-at-a-time sensitivity by increased observation time using MIRACLS technique.

Benefit from samples uniquely produced by the BGS.

3. AETHER will also perform high precision mass spectrometry for nuclear structure investigations.

Thank you!



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BERKELEY LAB
Lawrence Berkeley National Laboratory

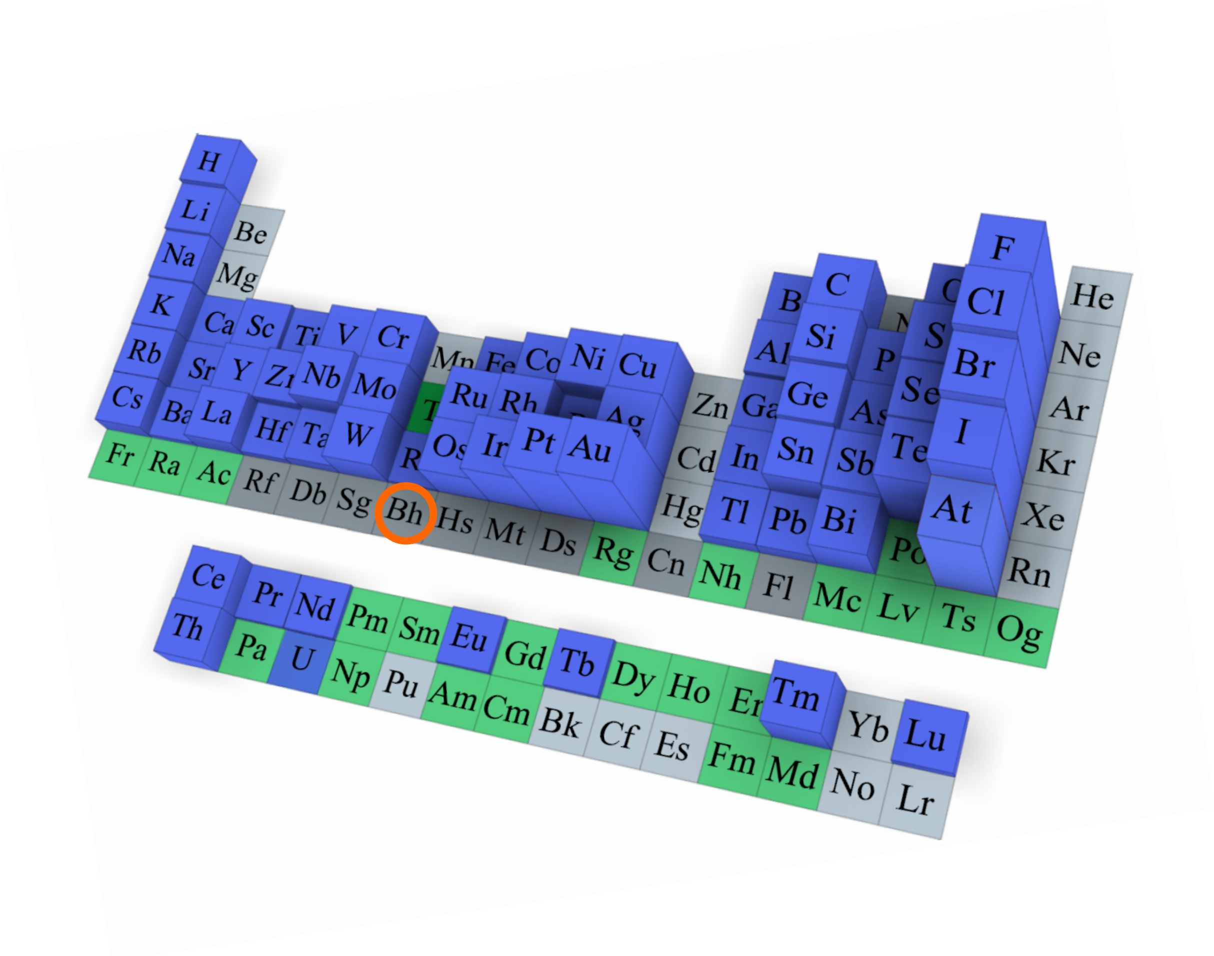
Heavy Elements
Lawrence Berkeley National Laboratory

MIRACLS

Reach with BGS

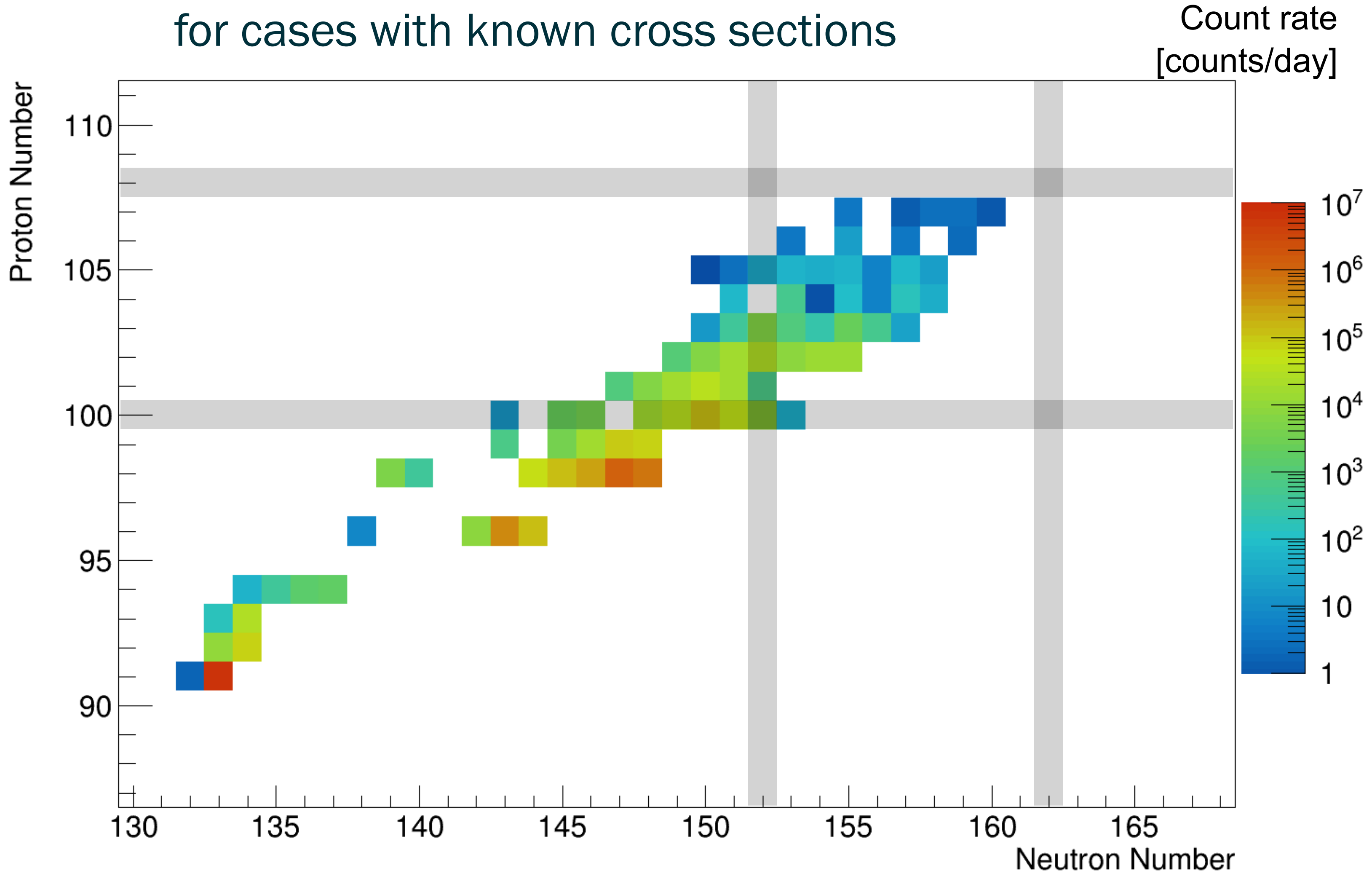
Mostly limited by efficiency through gas cell and beam conversion efficiency

Toughest scenario would require about 10 counts
Or ~1/day for a roughly 1 week beamtime



Adapted from D. Leimbach et al, Nat. Comm. (2020)

Estimated count-rate at the BGS MR-TOF for cases with known cross sections



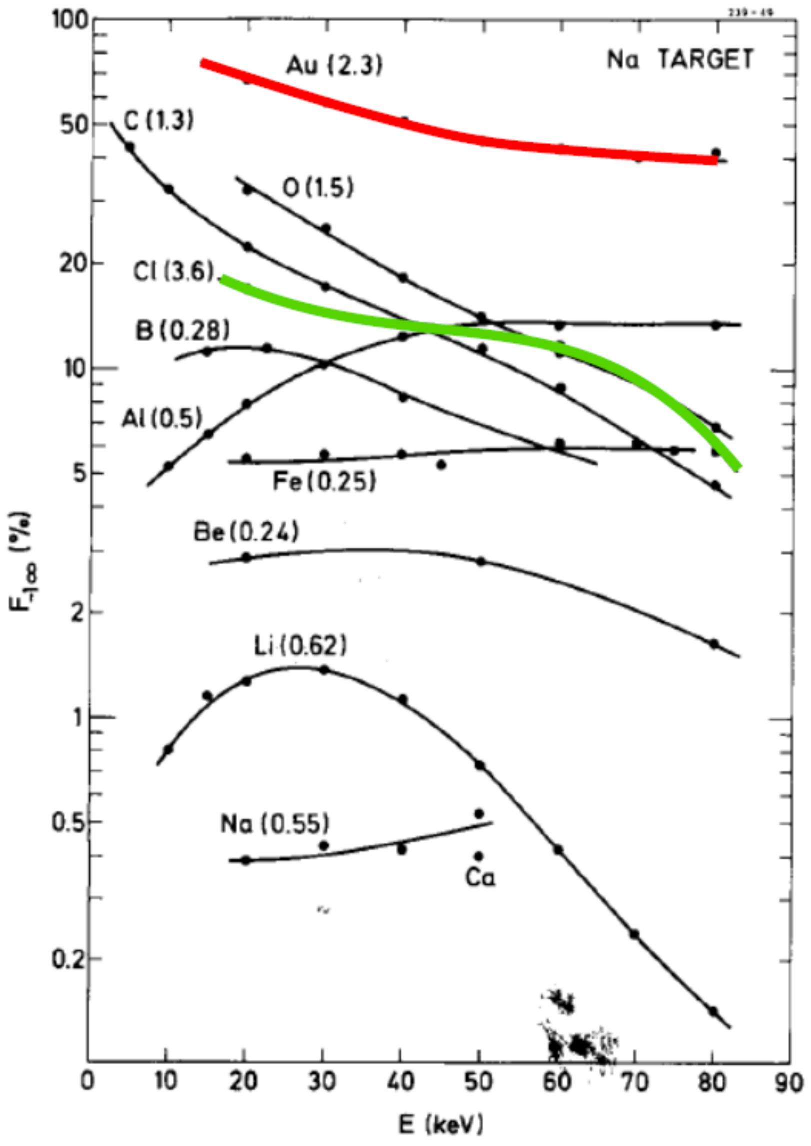
Charge exchange in metal vapors

Cross sections highly depend on EA, atomic radii and other properties of both target and projectile.

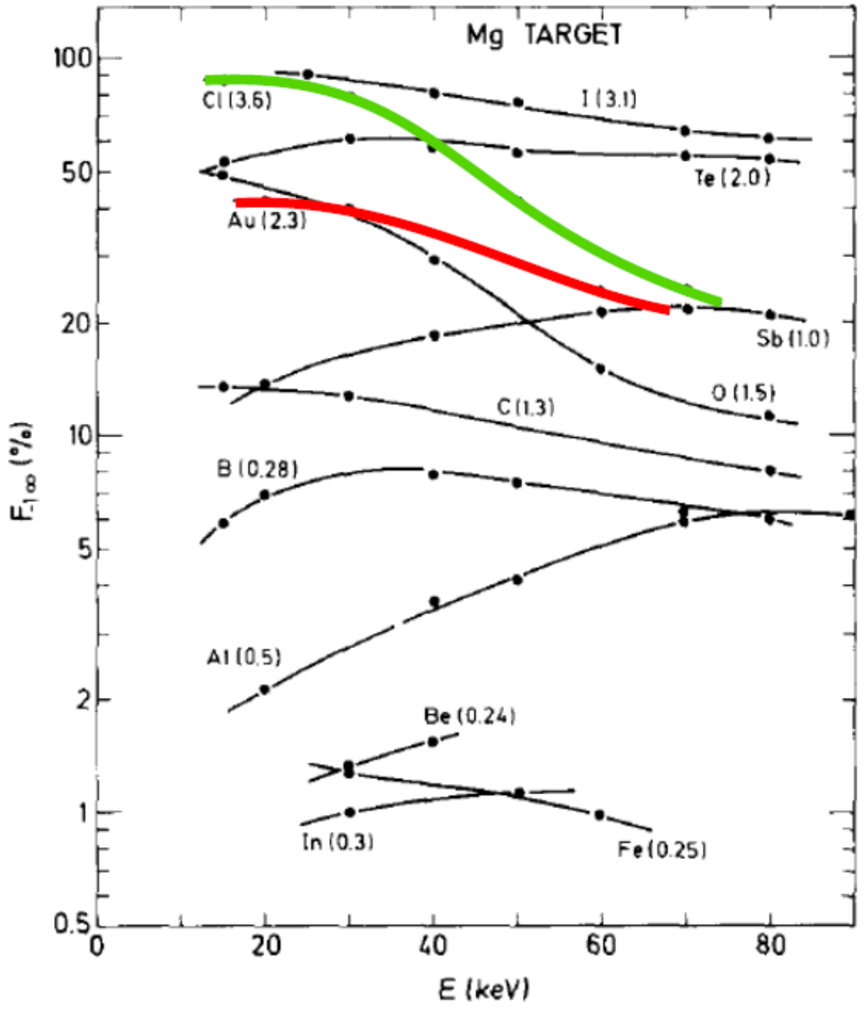
Studying the collisional process that leads to the creation of negative ions already provides a wealth of information.

$$B^+ + T^0 \rightarrow B^0 + T^+ + \Delta E\,(IP_B - IP_T)$$
$$B^0 + T^0 \rightarrow B^- + T^+ + \Delta E\,(EA_B - IP_T)$$

Na vapor



Mg vapor



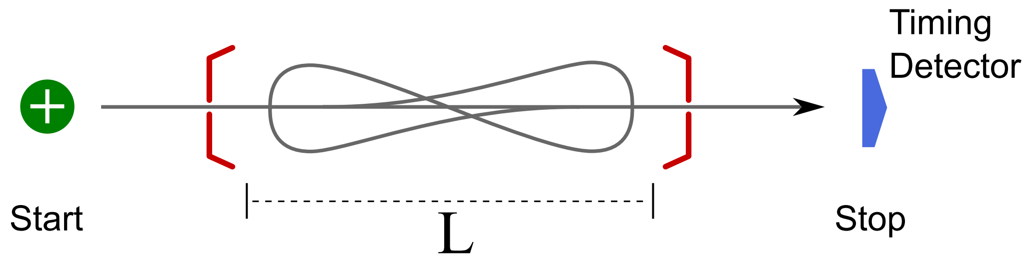
Heinemeier & Hvelplund, NIM 148 (1978)

MR-TOF 101

Multiple Reflection Time-of-Flight Mass Spectrometry

Drift through a field-free region

Time of flight: $$tof = L\sqrt{\frac{m}{2\,E_{kin}}}$$



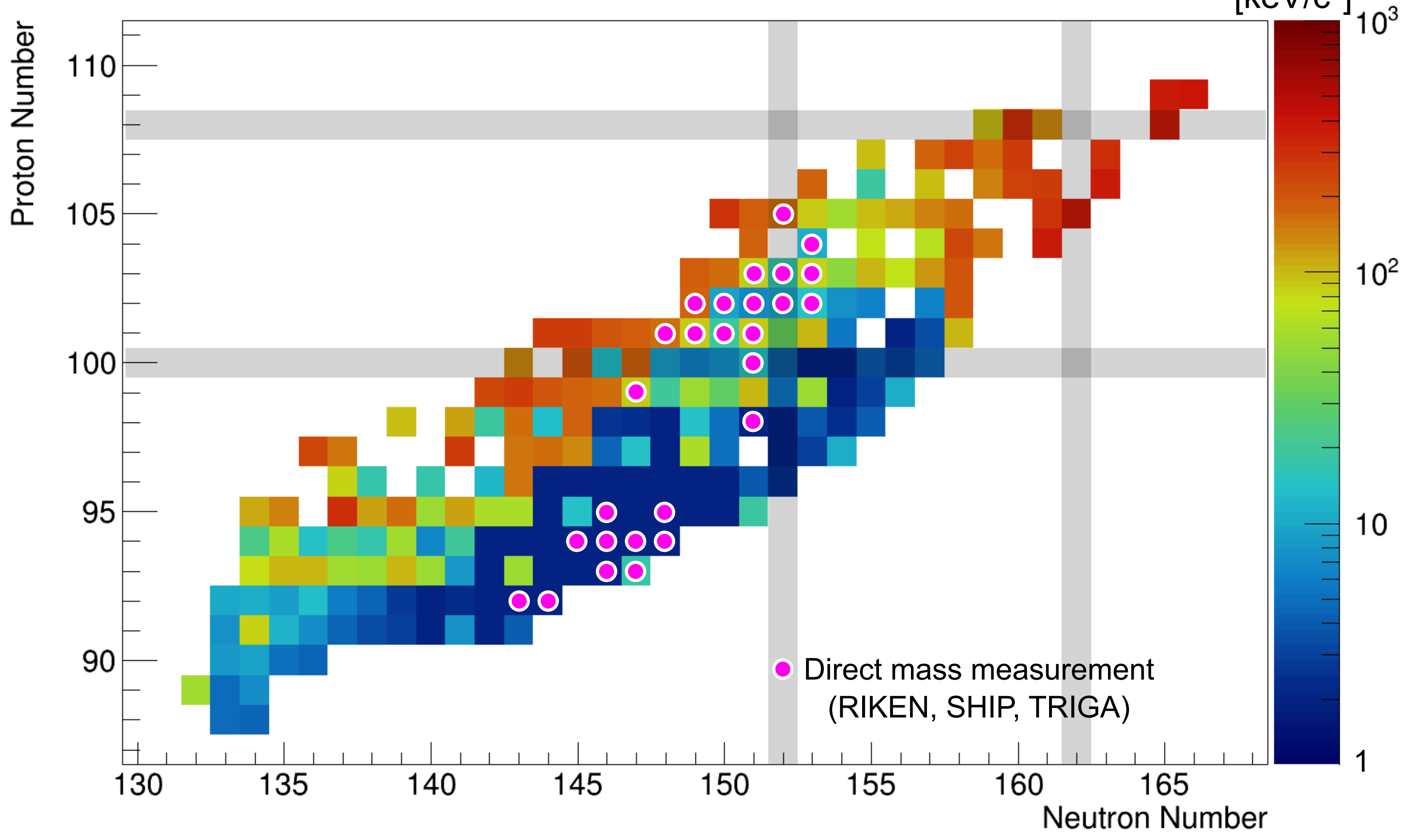
Resolution scales with ***L*** - recycle ***L*** many times

In MR-TOF-MS, flight path length can reach several km in a compact device.
Resolution is close to what PTMS offers.

Mass Spectrometry for Nuclear Structure

Opportunities at the BGS: (3) Transuraniums & Superheavy Elements

Attainable by fusion-evaporation,
with known half-lives >50 ms



Estimated count-rate at the BGS MR-TOF
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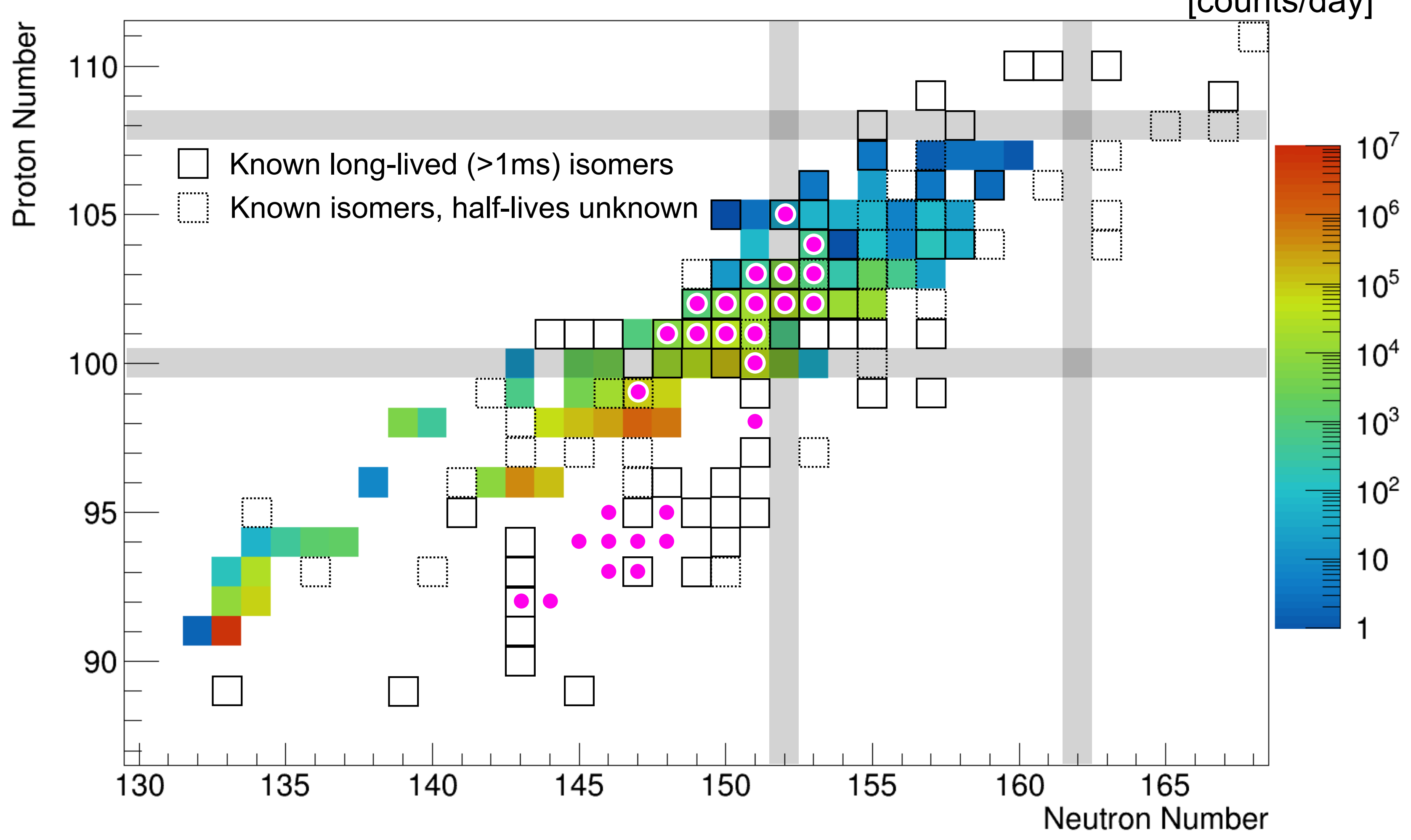
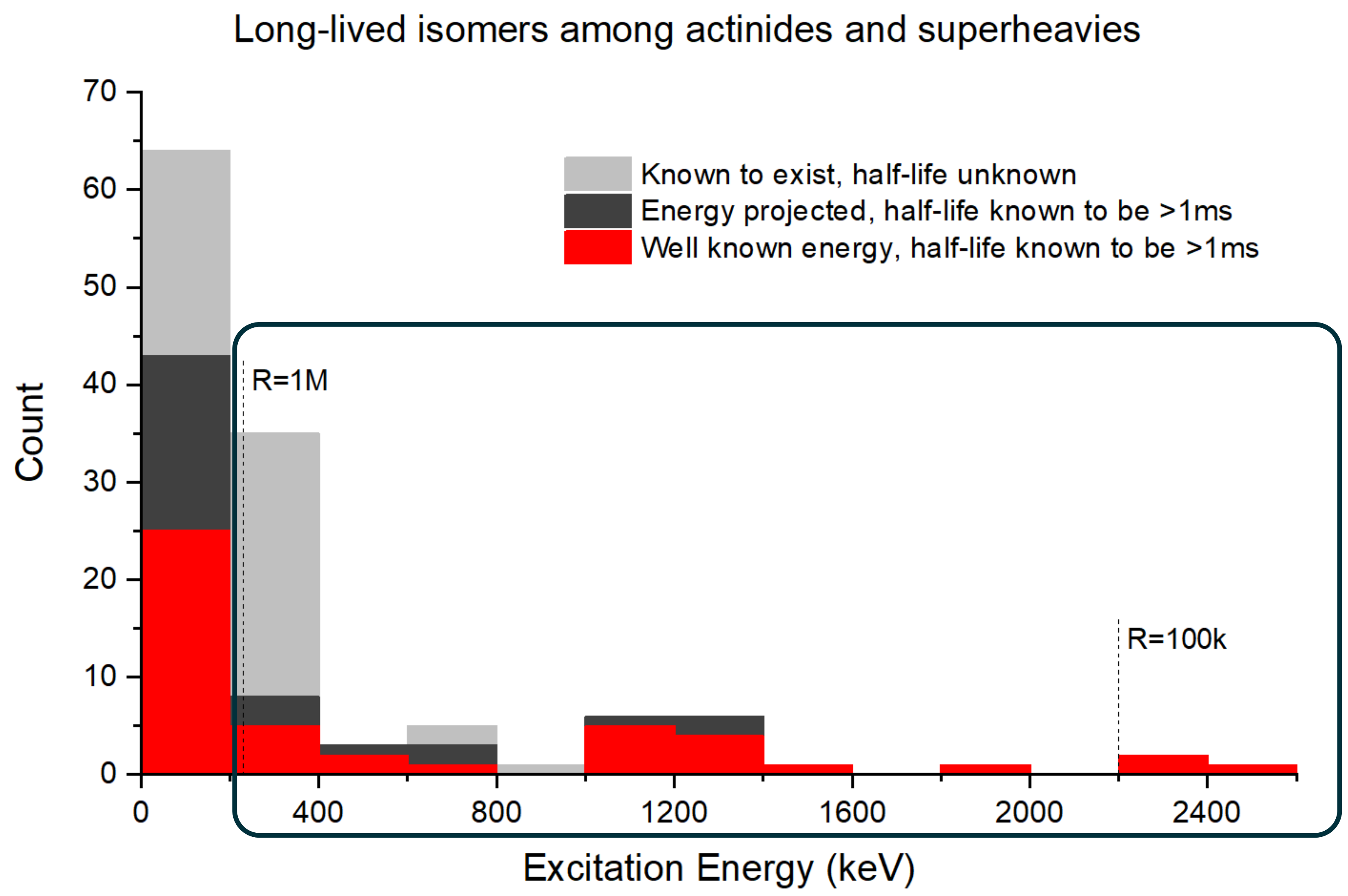


What could be done with a High-Res MR-TOF?

State-of-the-art: R = 1 M



40% of cases fully resolved!

Electron Affinities of SHE

Why do they matter?

- (One of the) ultimate tests of atomic theory

  Very pronounced relativistic effects

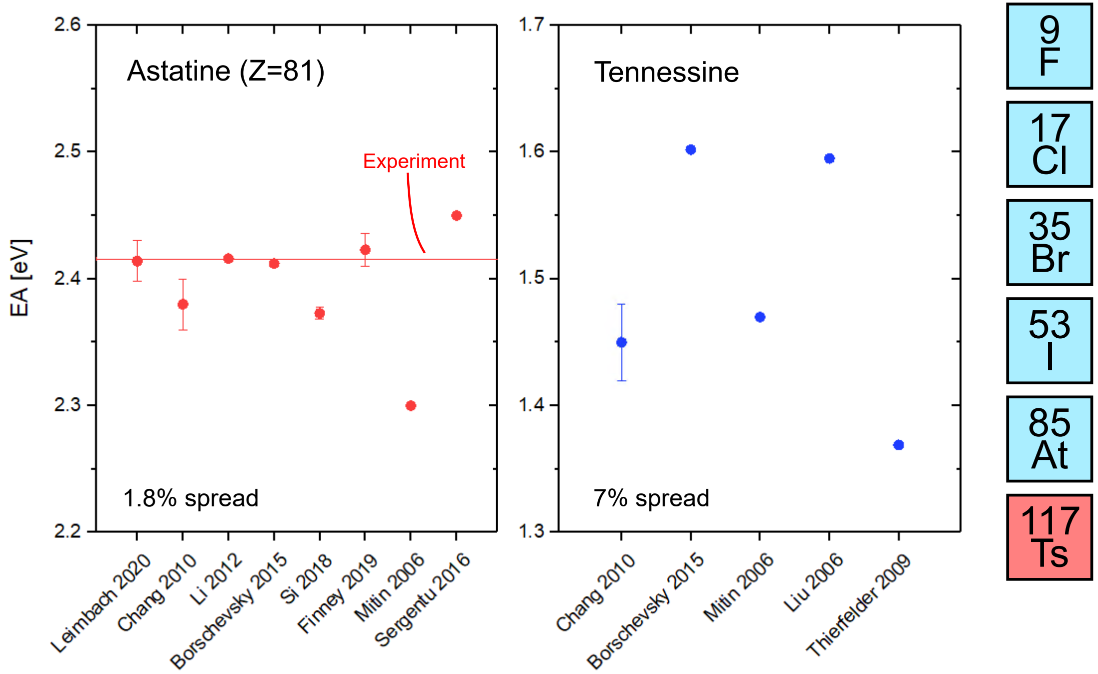
  Highly correlated electron systems

  Higher order QED effects sizeable

- Support SHE chemistry experiments

- Inspect validity of periods in the Periodic Table of Elements

Tennessine (Z=117)



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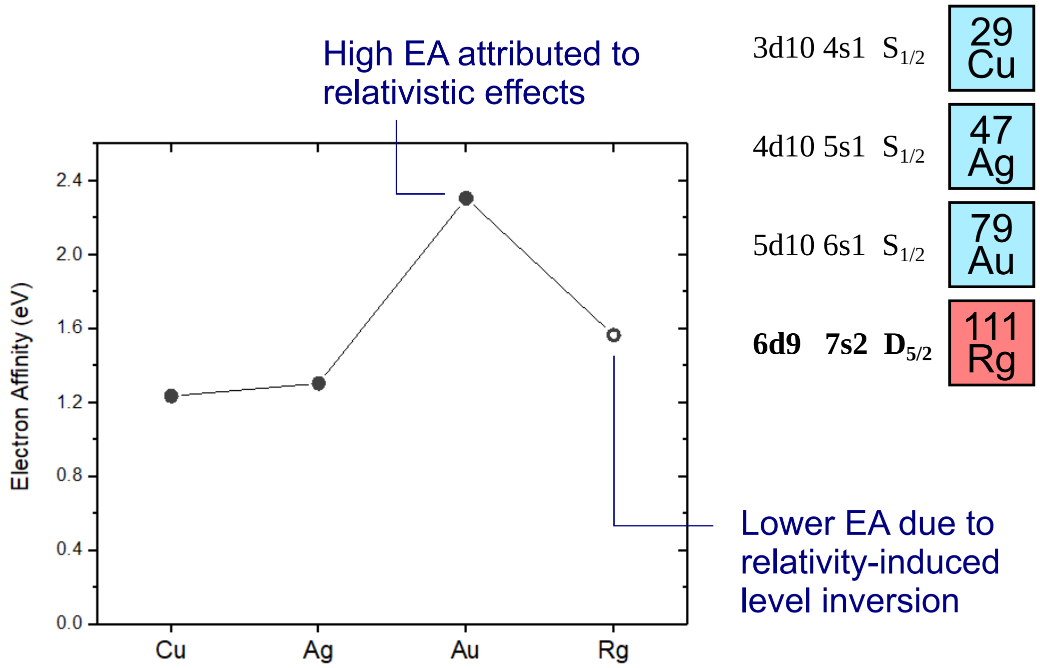
Roentgenium (Z=111)

Extreme relativistic effects



E. Eliav, et al. Phys. Rev. Lett. 73, 3203 (1994)

Electron Affinities of SHE

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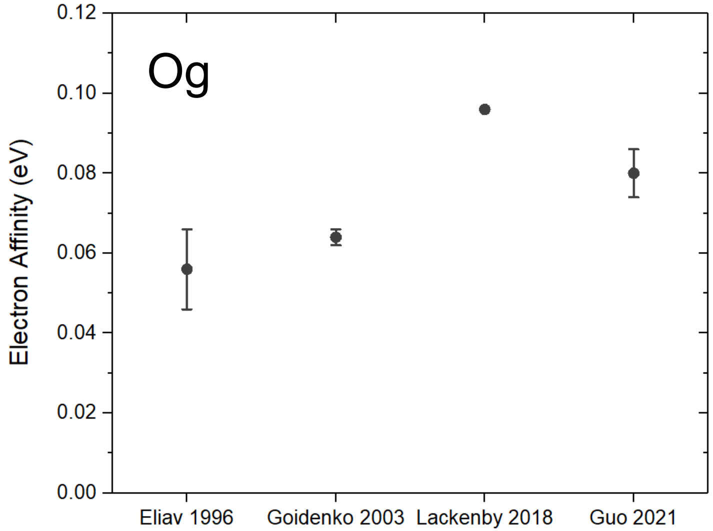
Oganesson (Z=118)

Is it a Noble Gas?

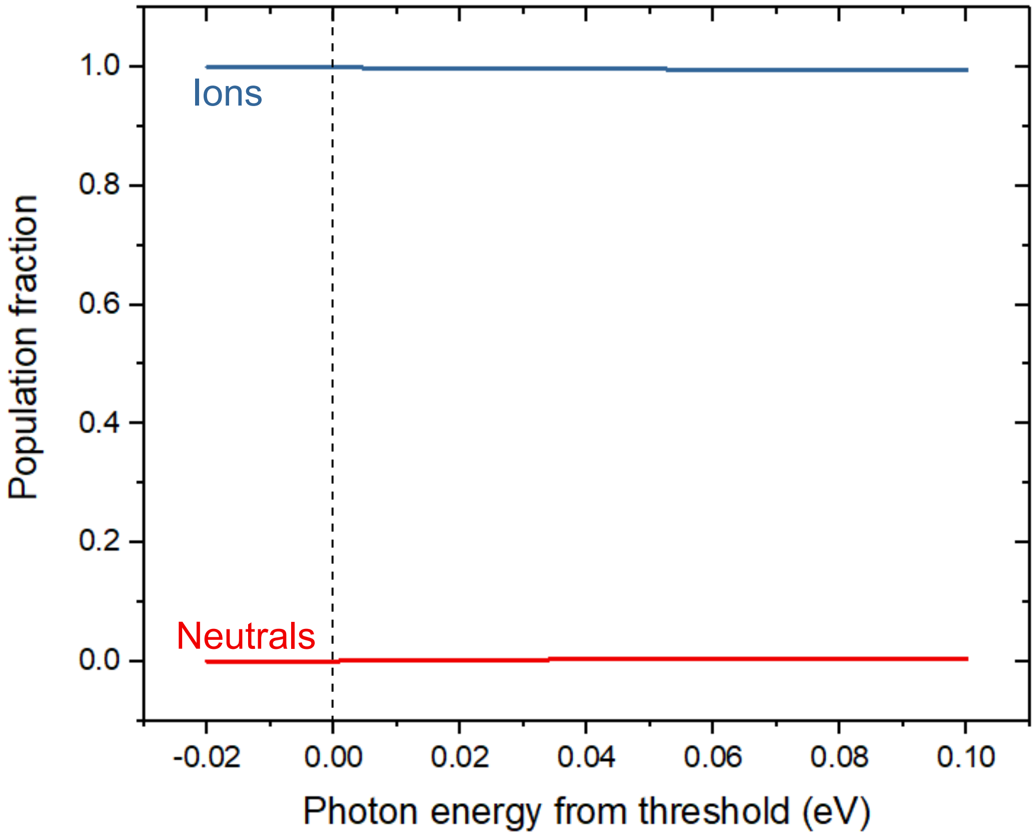


Unbound	2 He
Unbound	10 Ne
Unbound	18 Ar
Unbound	36 Kr
Unbound	54 Xe
Unbound	86 Rn
Predicted Bound!!!	118 Og

Just the observation of Og^- would demonstrate a remarkable deviation from PTE patterns!

Electron Affinities of SHE

LPT, one atom at a time



Goal: 100% photodetachment probability
with less than 0.1 eV from threshold

Halogen-like (Tennessine)

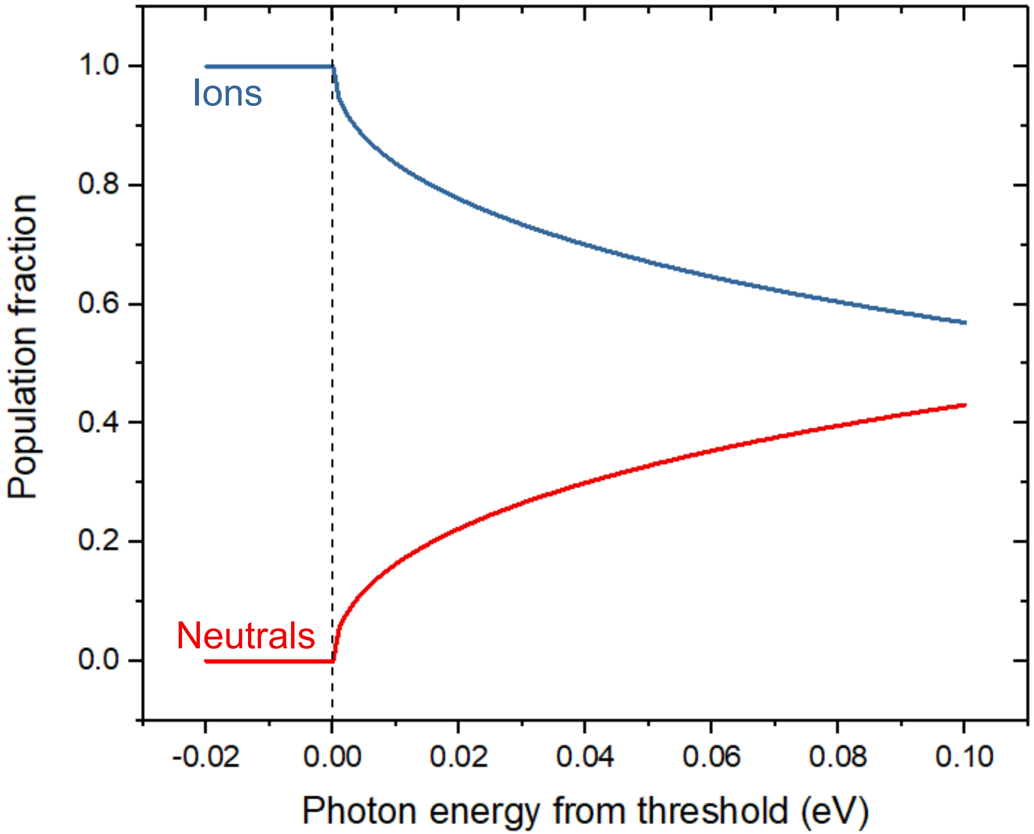
1.0 W continuous laser power

Single pass, 50 μs of laser exposure

1.5 mm laser radius, 70% laser-ion overlap, 60% of trapping time inside drift tube

Electron Affinities of SHE

LPT, one atom at a time



Goal: 100% photodetachment probability
with less than 0.1 eV from threshold

Halogen-like (Tennessine)

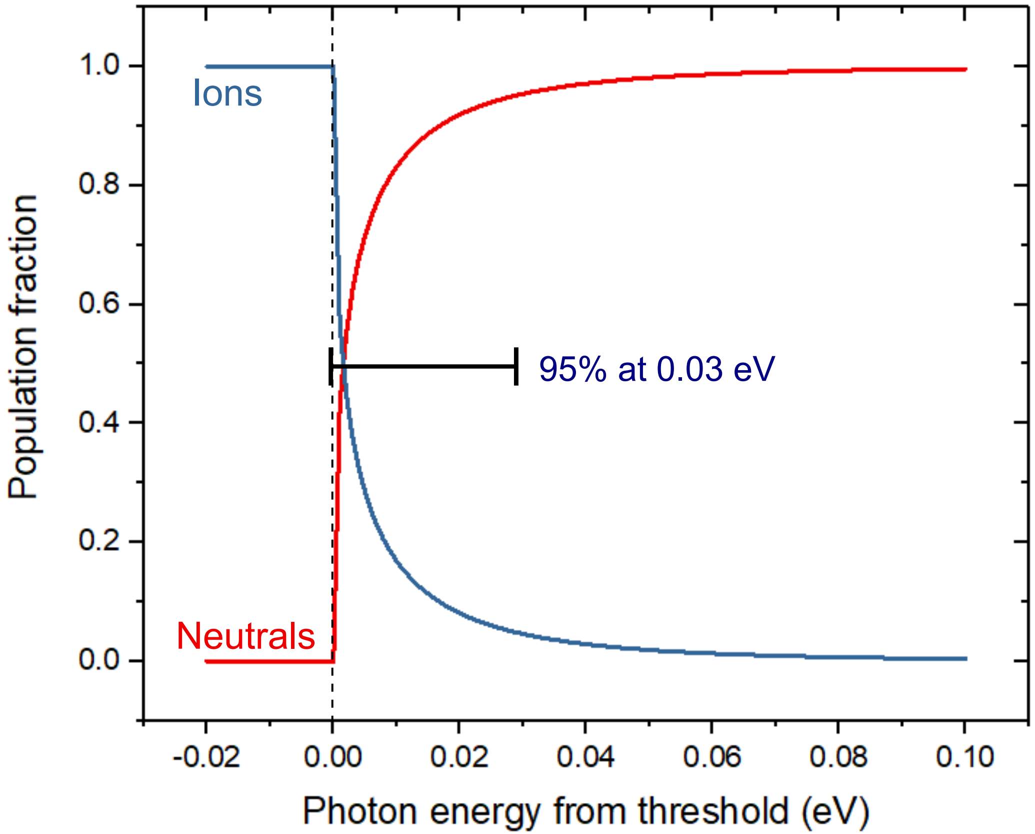
1.0 W continuous laser power

10 ms trapping

1.5 mm laser radius, 70% laser-ion overlap, 60% of trapping time inside drift tube

Electron Affinities of SHE

LPT, one atom at a time



Goal: 100% photodetachment probability
with less than 0.1 eV from threshold

Halogen-like (Tennessine)

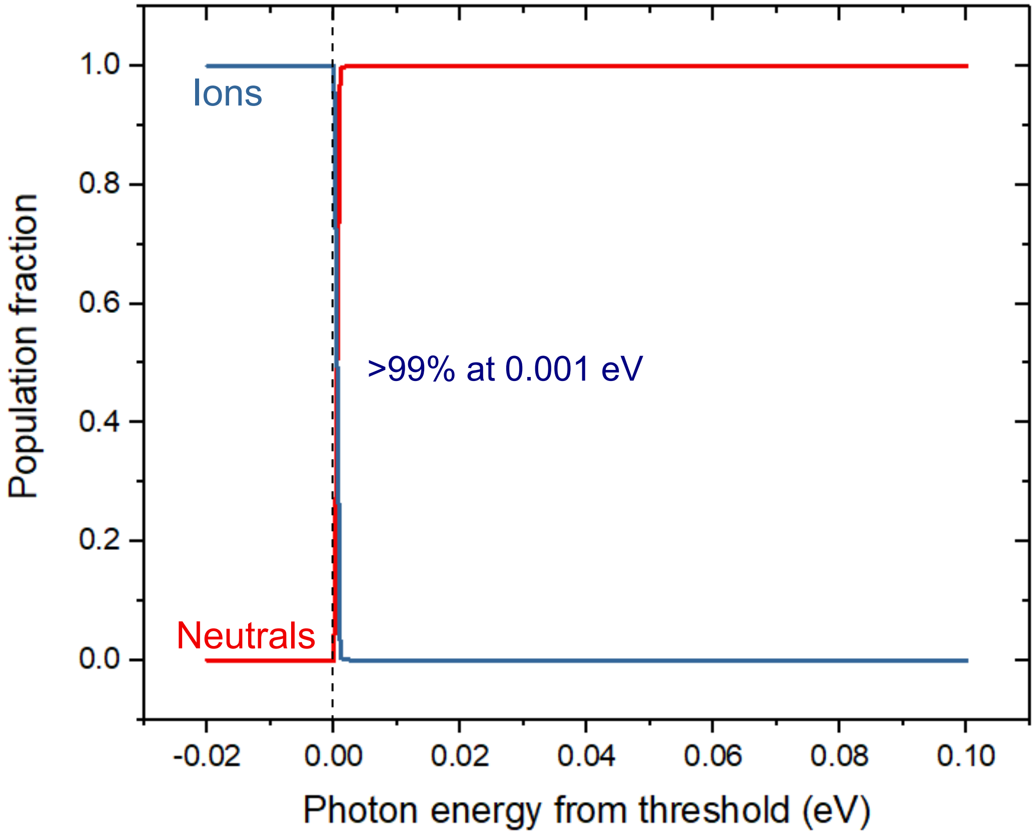
1.0 W continuous laser power

100 ms trapping

1.5 mm laser radius, 70% laser-ion overlap, 60% of trapping time inside drift tube

Electron Affinities of SHE

LPT, one atom at a time



Goal: 100% photodetachment probability with less than 0.1 eV from threshold

Halogen-like (Tennessine)

1.0 W continuous laser power

1000 ms trapping

Measurement strategy:
- successive approximations, approach threshold as statistics permit
- no need to search for a narrow resonance!
- not much reliant on reference transitions or accurate calculations

1.5 mm laser radius, 70% laser-ion overlap, 60% of trapping time inside drift tube