# PROGRESS ON BARIUM TAGGING FOR NEXT

**Ben Jones**

University of Texas at Arlington

UNIVERSITY OF TEXAS ARLINGTON

# NEXT

## USA

University of Texas Arlington

Argonne National Laboratory

Harvard (VE RI TAS)

Fermilab

ATM

Iowa State University

Pacific Northwest National Laboratory

*co-spokespersons:*

***David Nygren***

***JJ Gomez Cadenas***

## Spain

DIPC

IFIC Institut de Física Corpuscular

Universitat de Girona

USC Universidade de Santiago de Compostela

Universidad de Zaragoza

Universitat Politècnica de València

## Portugal, Israel, Russia, Columbia

U C

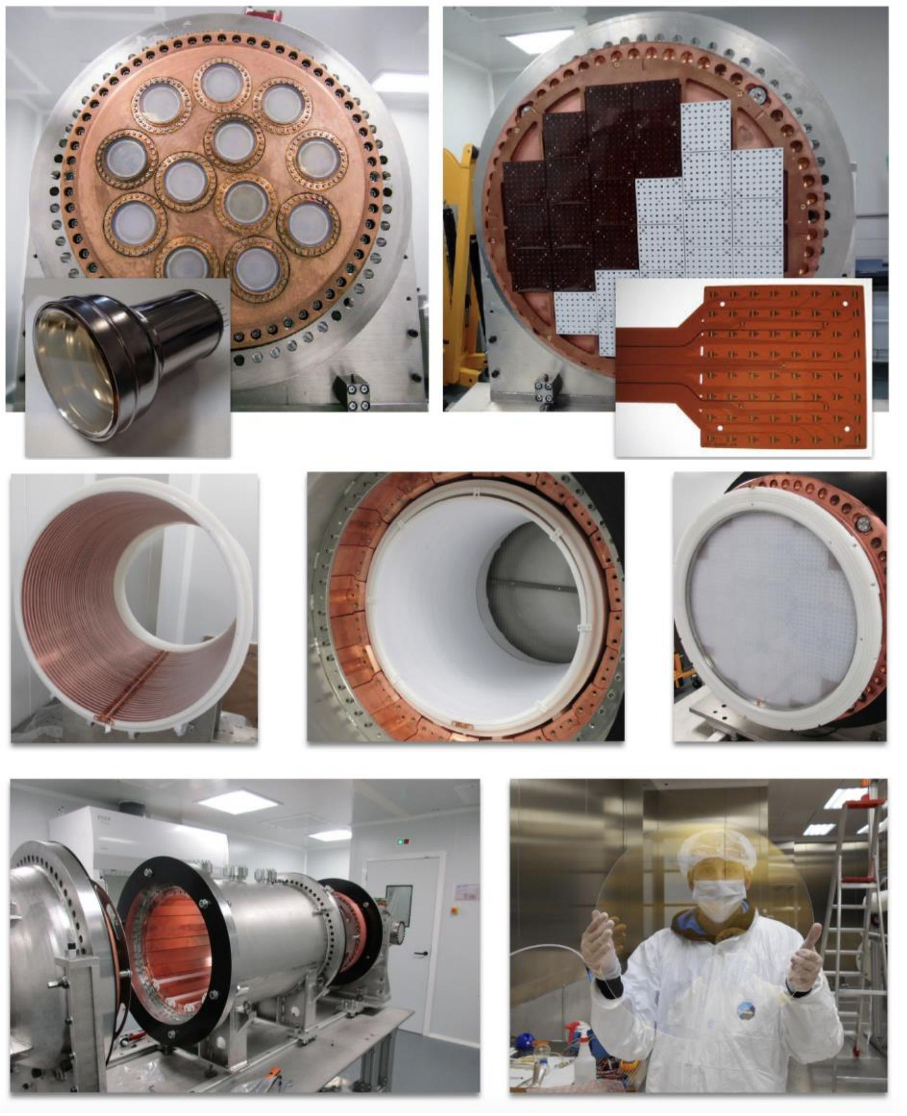
JINR Dubna

universidade de aveiro

Ben-Gurion University of the Negev

UAN Universidad Antonio Nariño

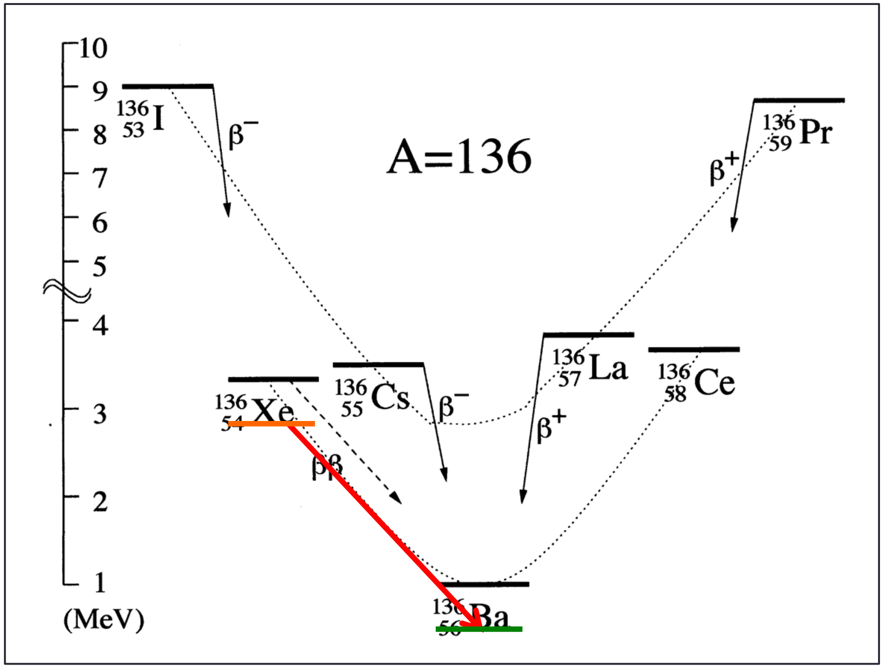
## *Lior Arazi already talked about NEXT and HPGXe –*
## *so I will skip introductions and get right to the point…*



# Barium Tagging

- Barium ion is only produced in a true ββ decay, not in any other radioactive event.
- Identification of Ba ion plus ~1% FWHM energy measurement would give a background-free experiment.
- ***Various experimental techniques exist to image single atoms or ions.***
- ***Can any of them be applied to efficiently detect an individual barium ion or atom in a ton of material?***

$^{136}Xe \rightarrow {}^{136}Ba + e + e$

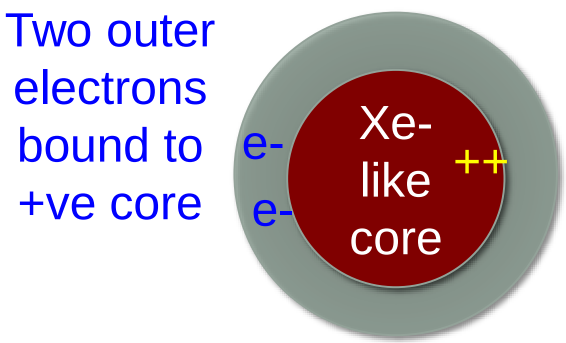



**Barium tagging for 0nubb has been actively explored in liquid and gaseous xenon for >15 years, with the holy grail is a scalable, efficient, single ion sensitive technology.**

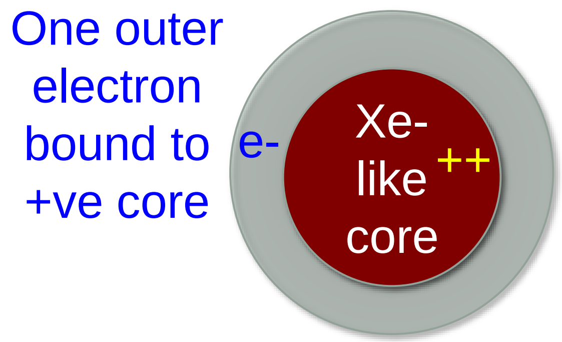
# Barium Atoms and Ions

- Barium is born in a high charge state as emerging beta electrons disrupt the atom
- Quickly captures electrons from xenon to reach the $Ba^{++}$ state
- In gas, it ~stops there. In liquid, further recombination happens.

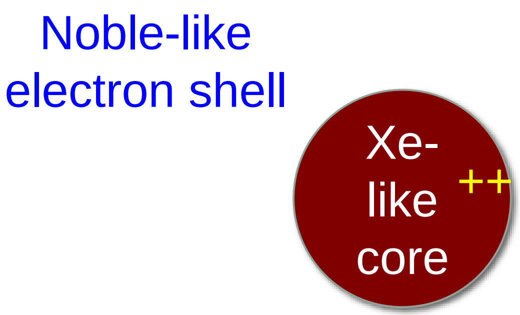
Structure of the **Ba** atom




Structure of the **Ba+** ion



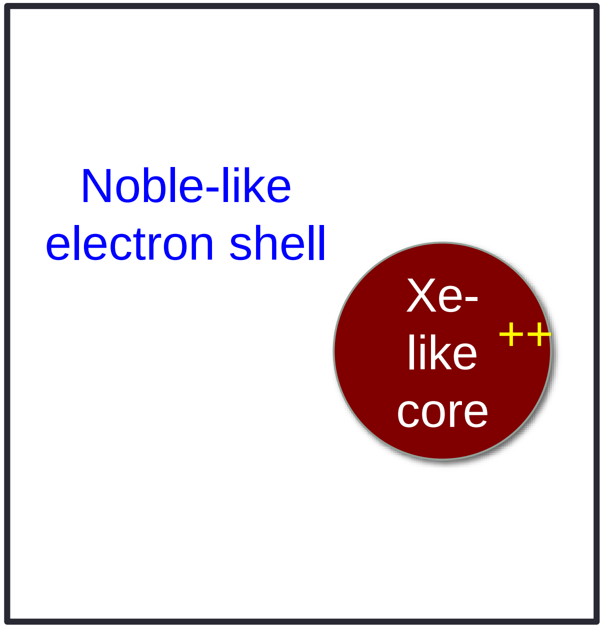

Structure of the **Ba$^{++}$** ion




**Liquid –**
Some distribution

**Gas –**
Mostly this

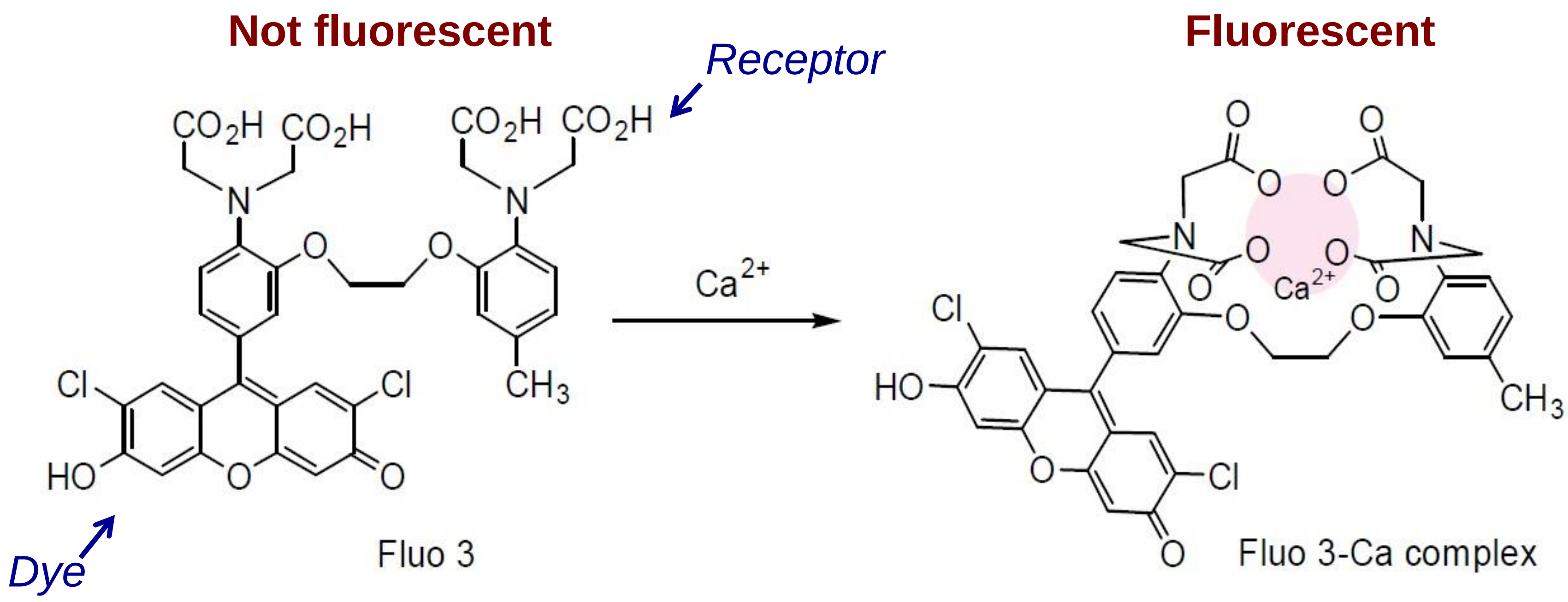
# Q: How do you make $Ba^{++}$ shine?



***Concept to adapt SMFI for Ba tagging:***
***D.R. Nygren,* J.Phys.Conf.Ser. 650 (2015) no.1, 012002**

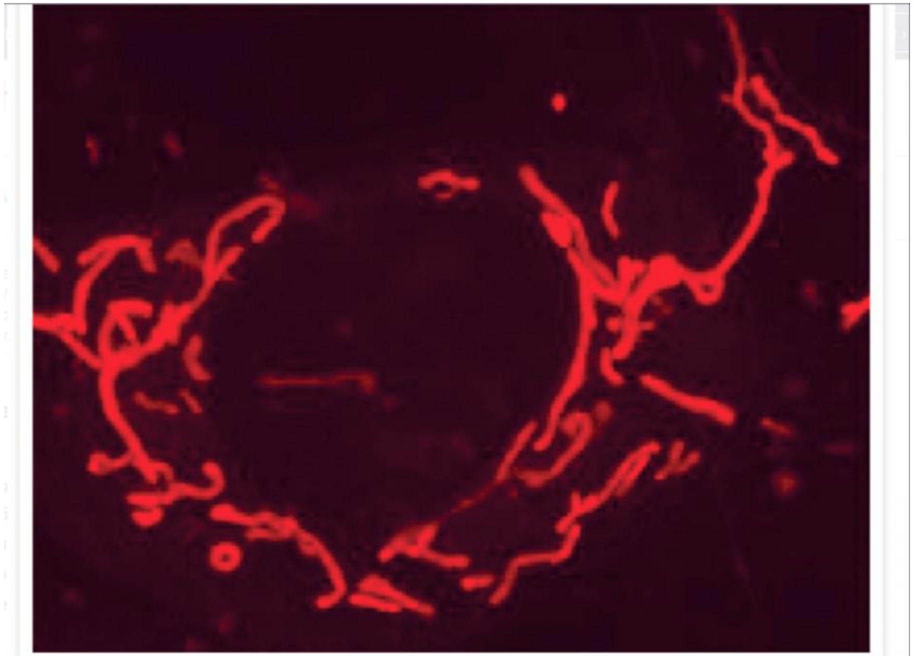
# SMFI:

- A non-fluorescent molecule becomes fluorescent (or vice versa) upon chelation with an incident ion.




*Calcium and barium are congeners – many dyes developed for calcium are also expected to respond to barium*

## SMFI is a technique from biochemistry with demonstrated single-ion resolution.



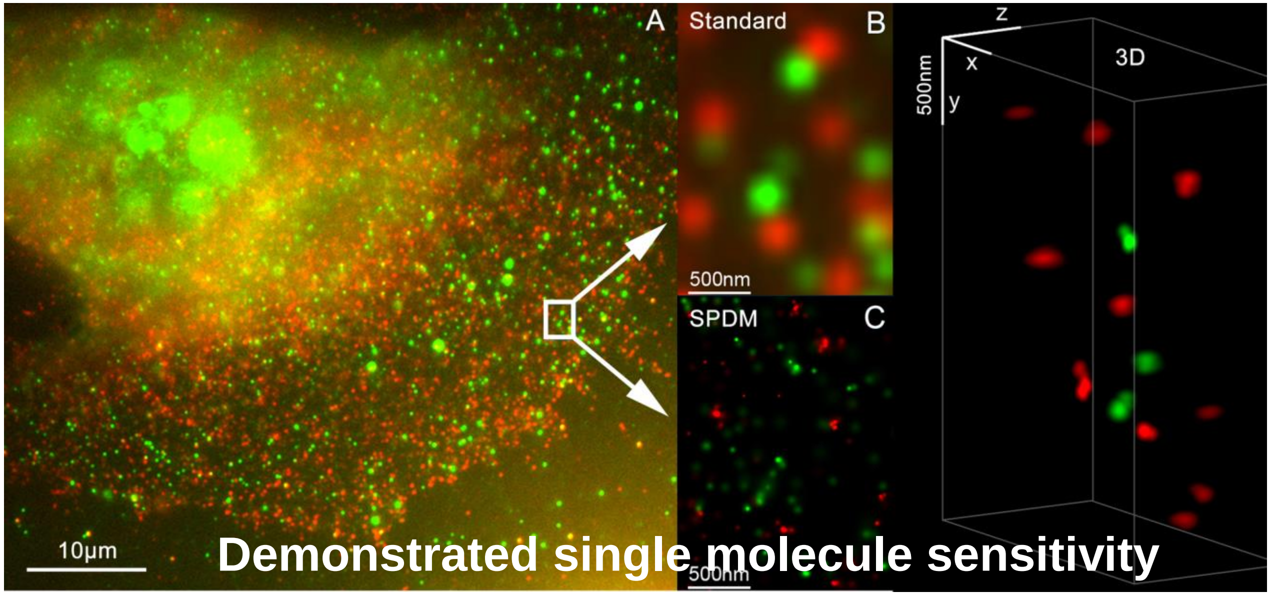
← Rhod-2 sensing $Ca^{++}$ production in rat astrocyte cells

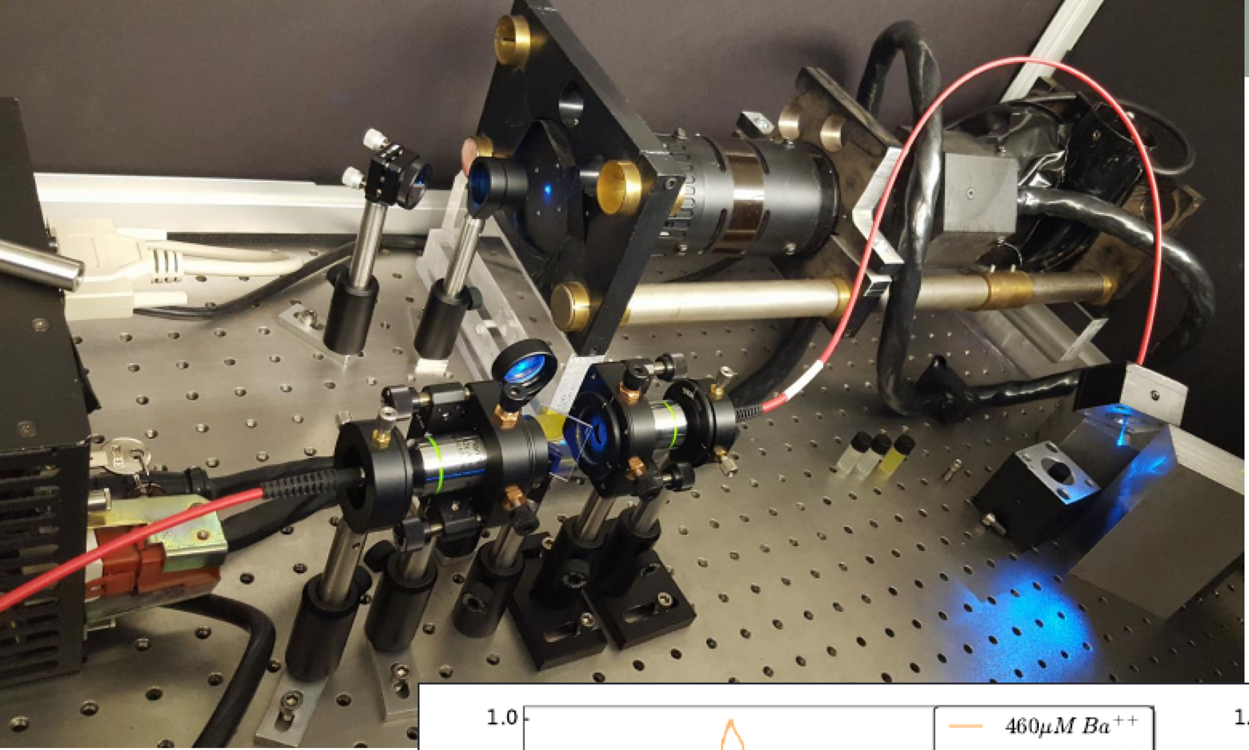
**J Microsc. 2011 Apr;242(1):46-54**

**J Cell Biol 145, 795 (1999).**

Single molecule tracking using SMFI is the basis of super-resolution microscopy→

*These methods won the Nobel Prize in chemistry in 2014.*

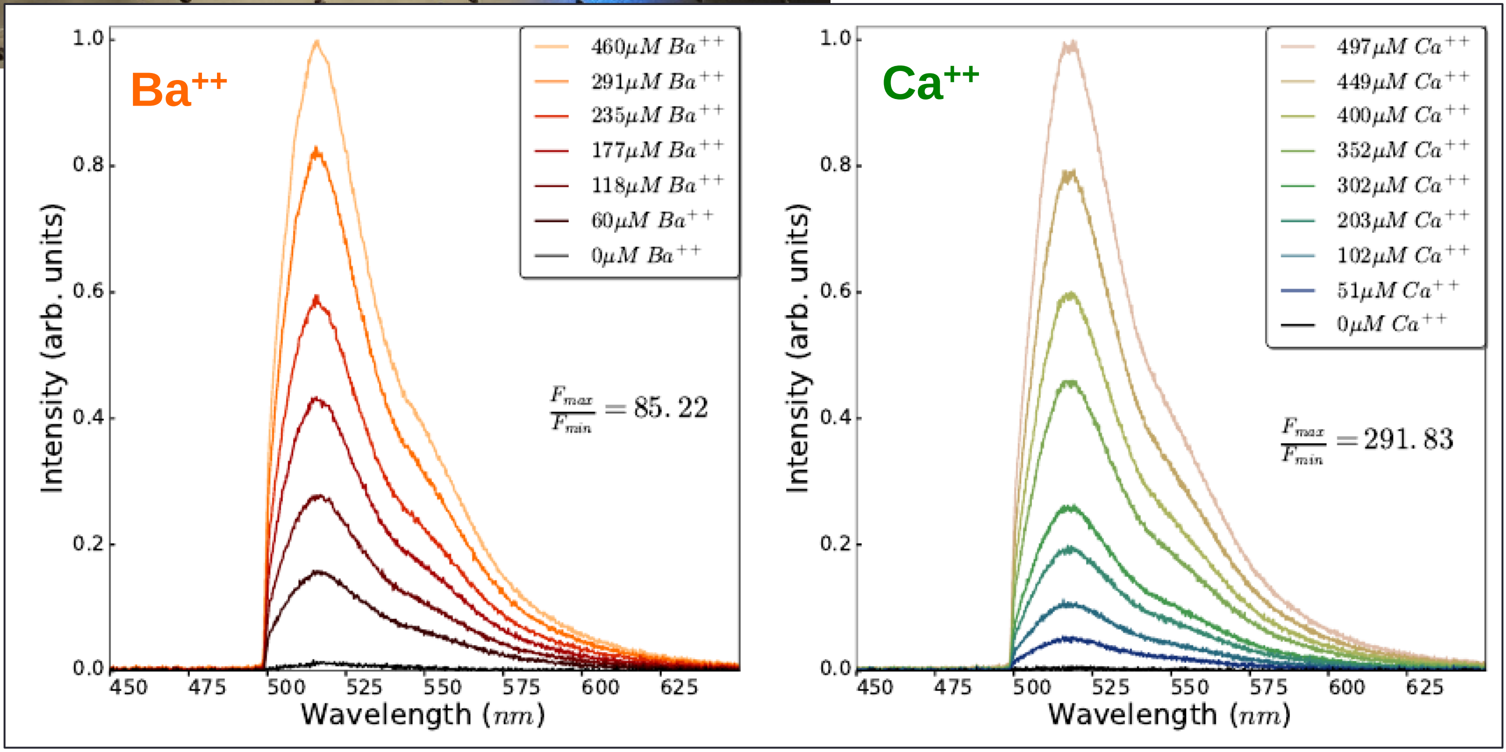


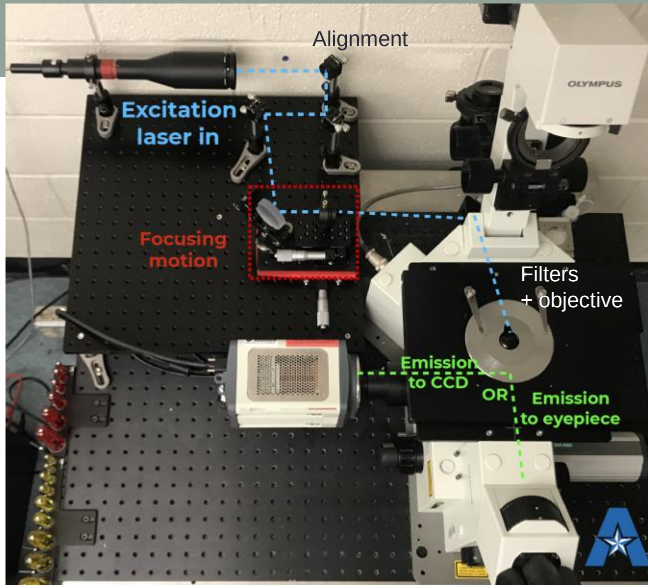


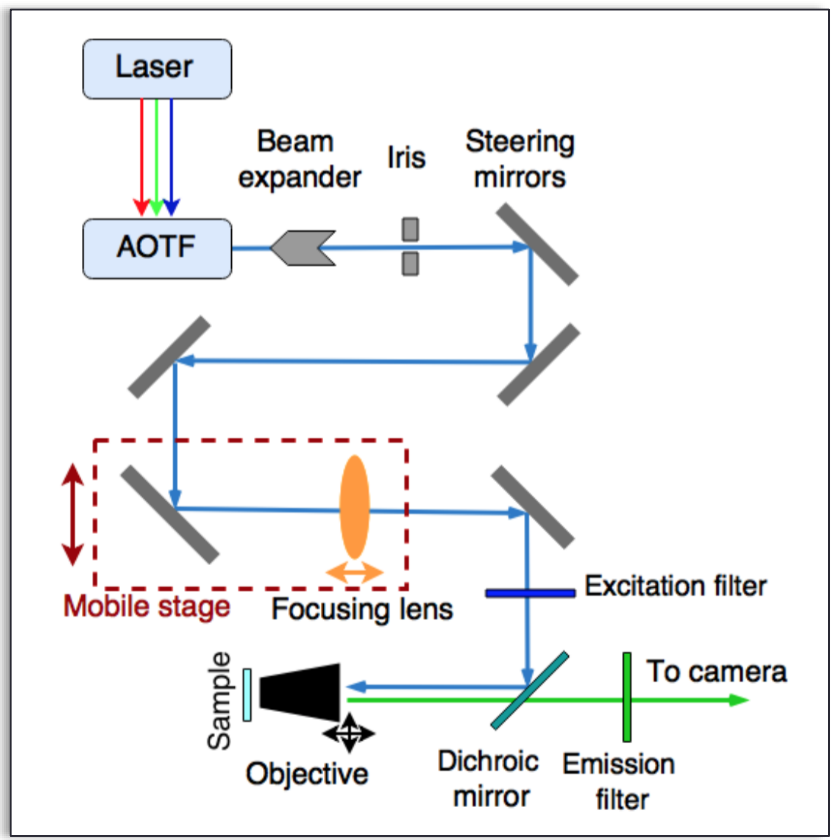
← First dabbling - developed a bespoke fluorescence sensor to study barium production at the end of a fiber.

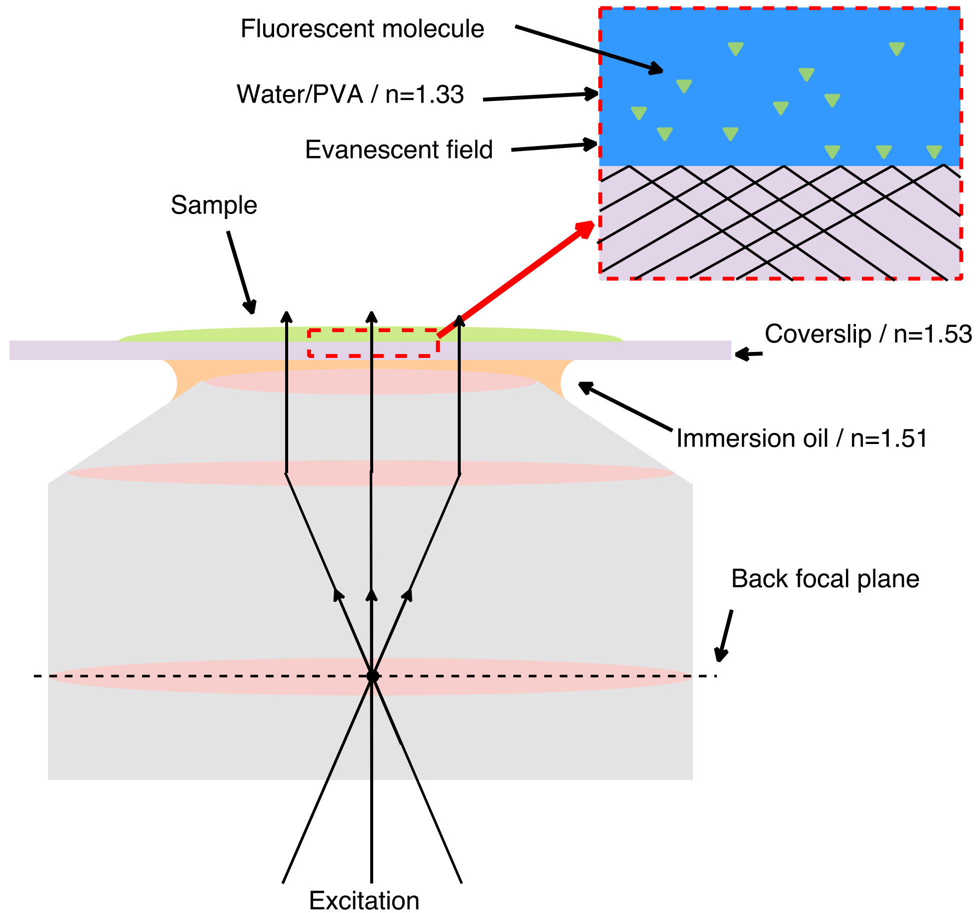
***Single molecule fluorescence imaging as a technique for barium tagging in neutrinoless double beta decay***
***Jones, McDonald, Nygren, JINST (2016) 11 P12011***

**We find strong fluorescence from Fluo3 and Fluo4 under chelation with $Ba^{++}$ ions →**



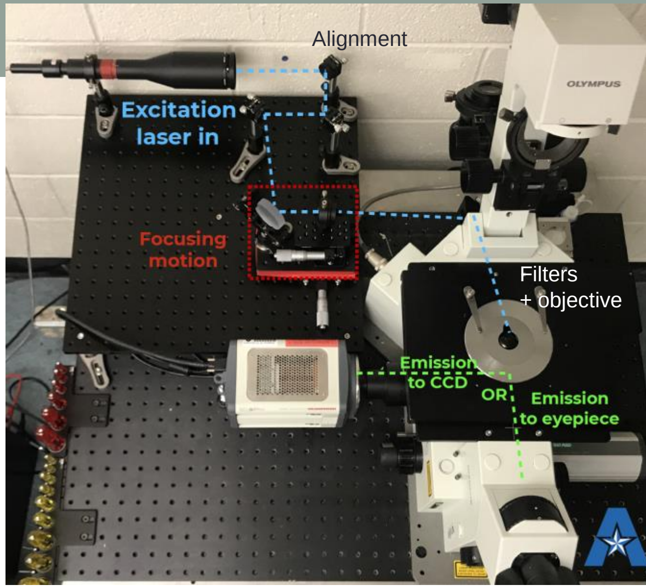


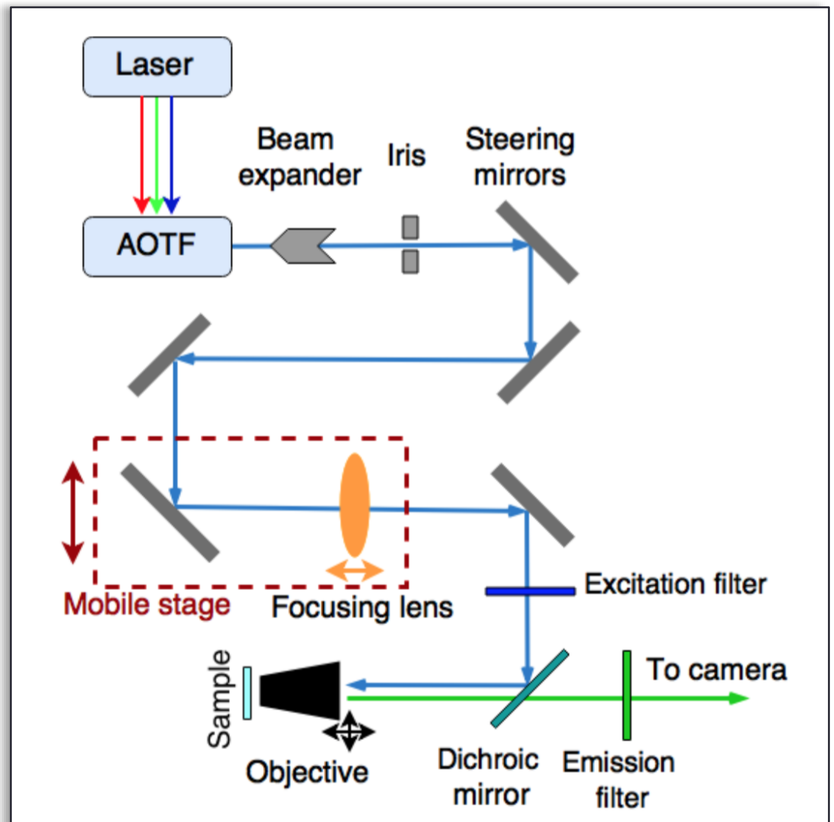



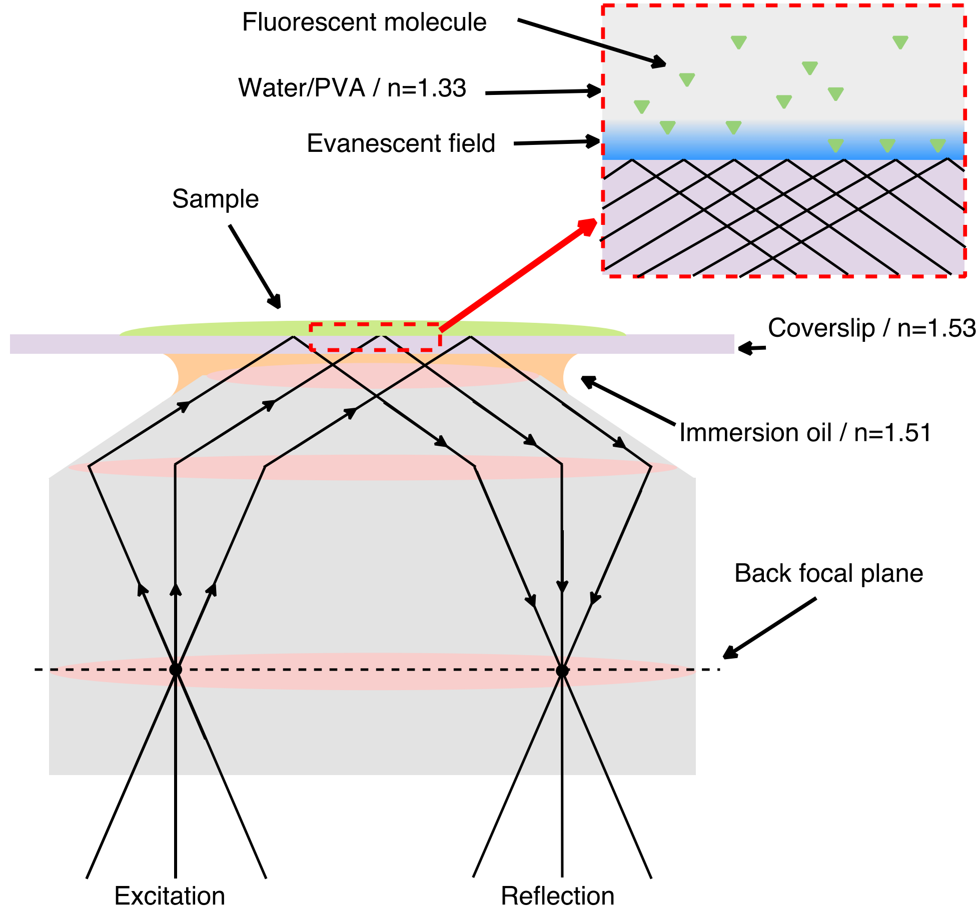




# TIRF

Total Internal Reflection Fluorescence microscopy



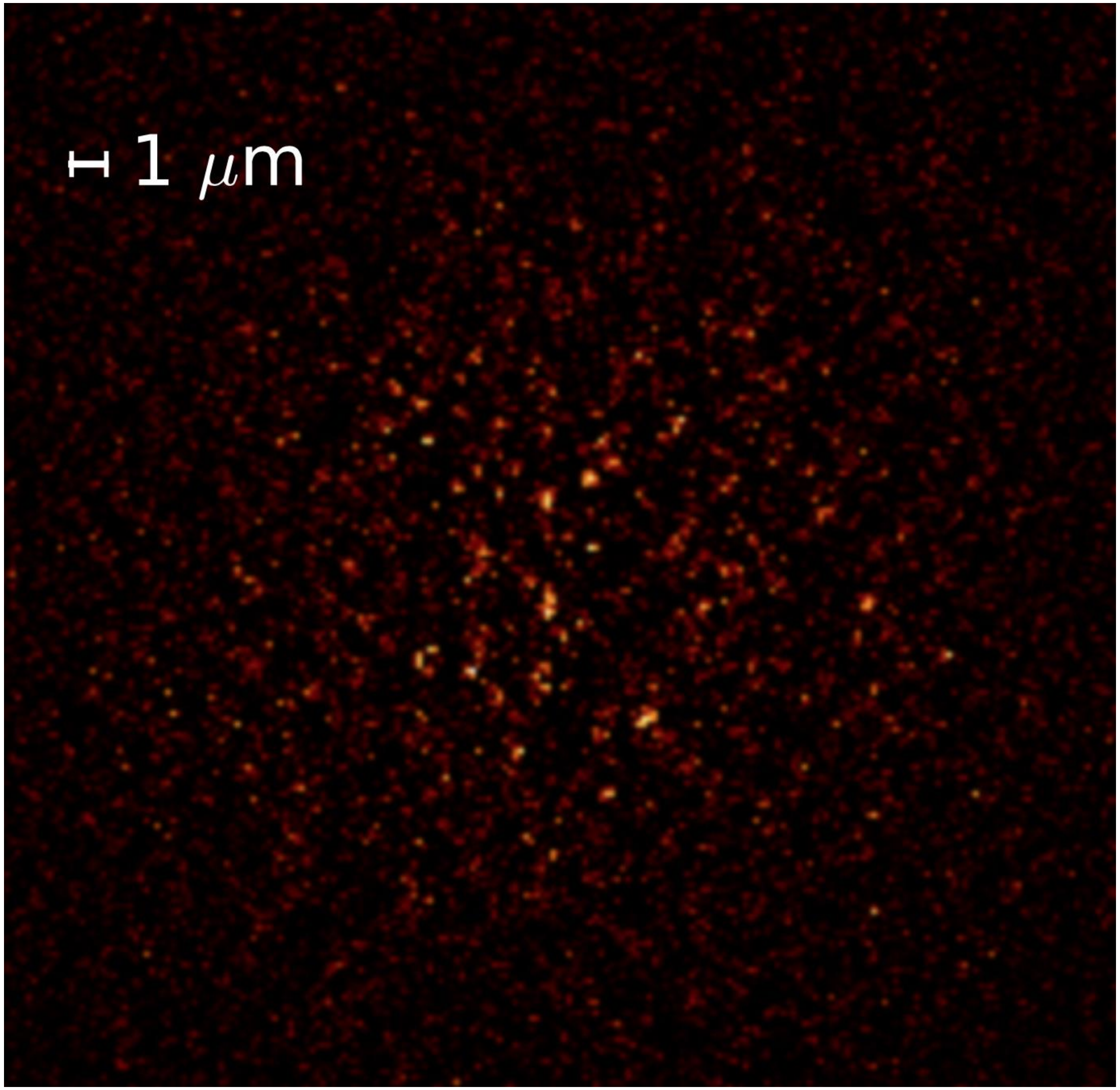




# TIRF

Total Internal Reflection
Fluorescence microscopy

# Single $Ba^{++}$ TIRF images

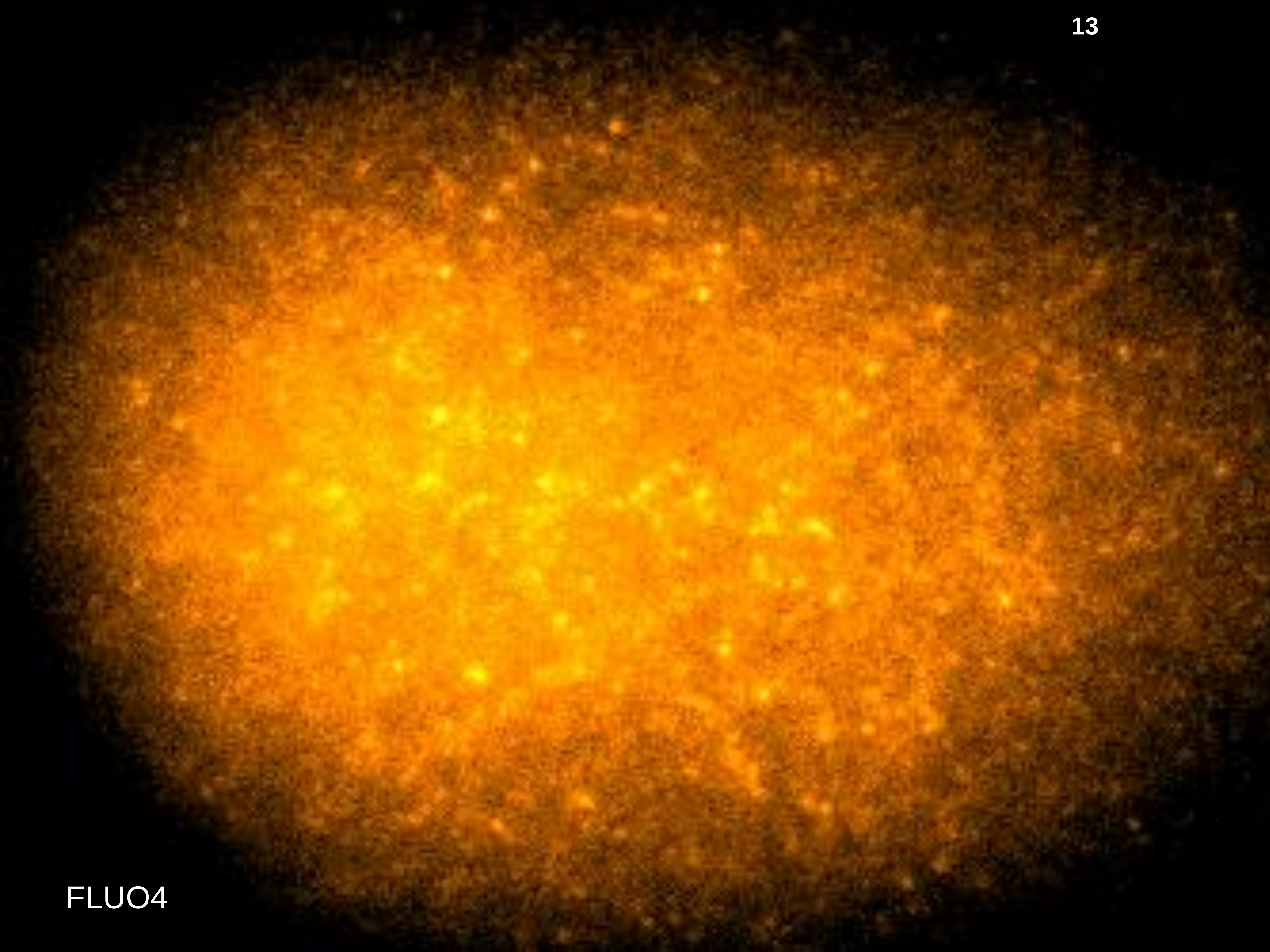


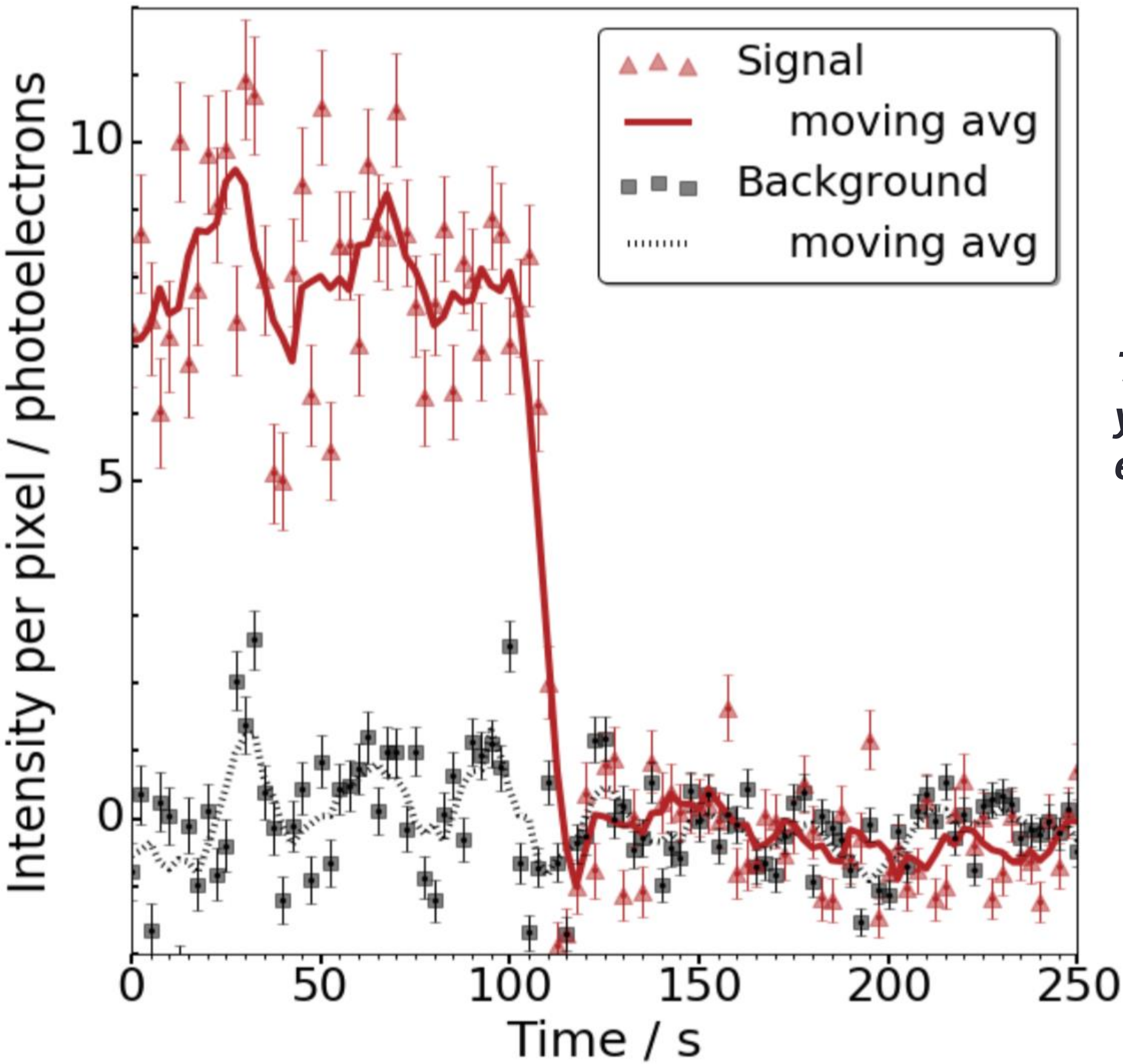

← **This image shows a weak solution of barium perchlorate salt on our sensor.**

Each spot is a **single barium ion.**

Brighter spots are near the TIRF surface, dimmer ones are deeper in the sample.

In a xenon detector, dye deposited as a monolayer and only brightest spots at constant depth expected.

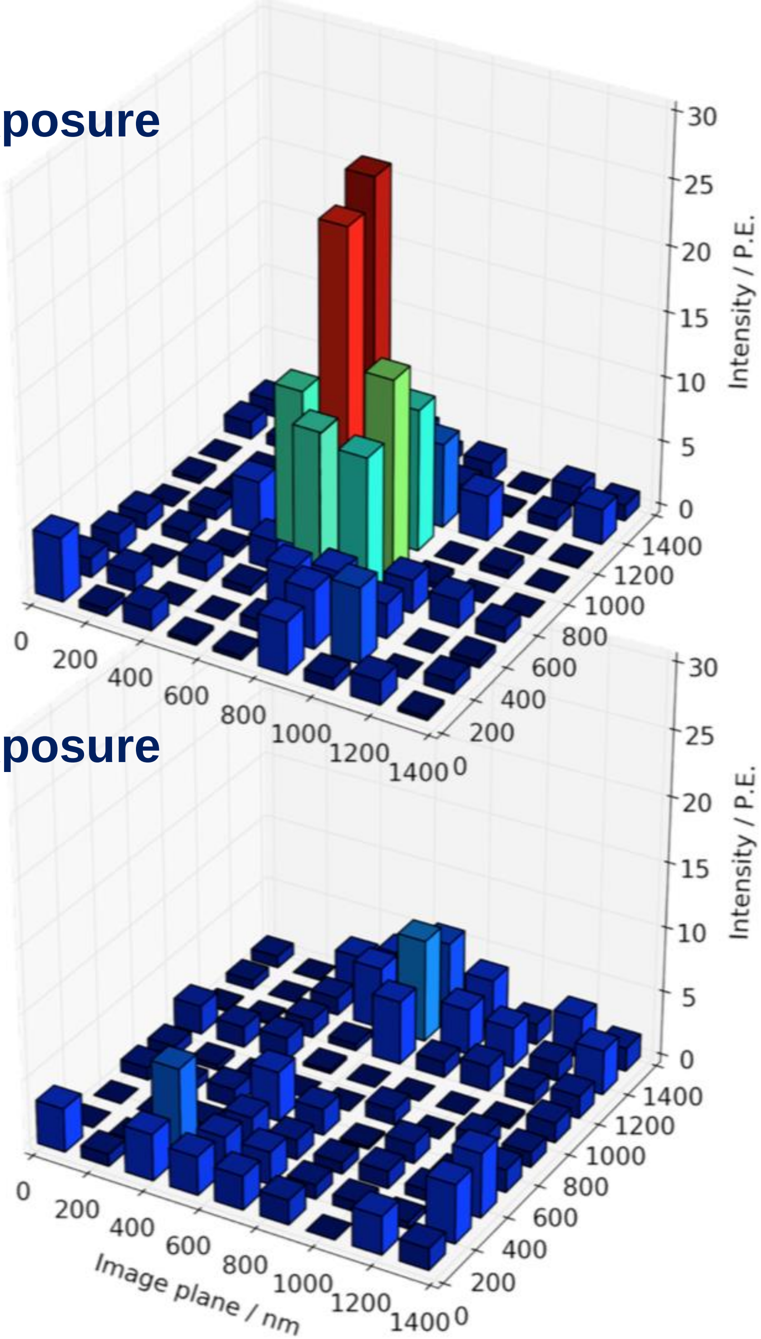

FLUO4




***This "step" is how you know it is exactly one ion.***

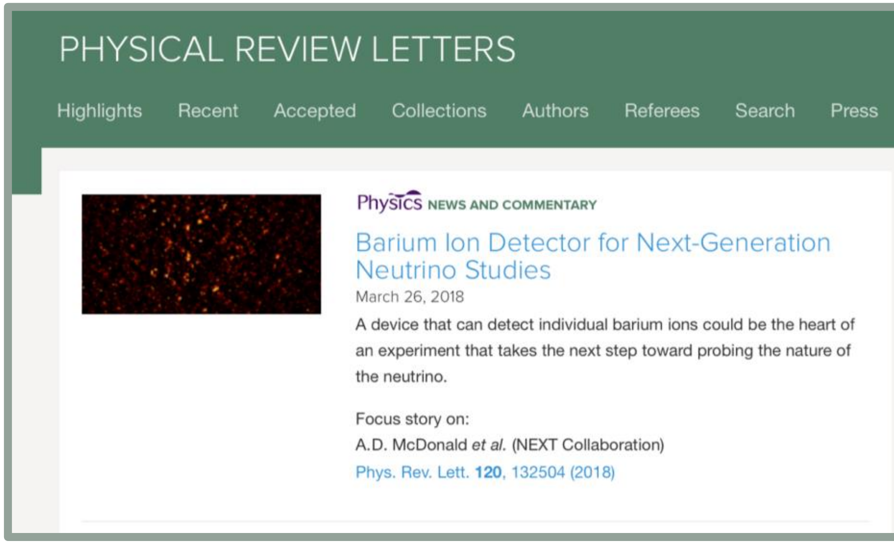
**0.5s exposure before:**

**0.5s exposure after:**




***Single ions barium resolved with 2nm super-resolution and 12.9 sigma stat. significance.***

***Phys.Rev.Lett. 120 (2018) no.13, 132504***


PHYSICAL REVIEW LETTERS

Highlights Recent Accepted Collections Authors Referees Search Press

Physics NEWS AND COMMENTARY

Barium Ion Detector for Next-Generation Neutrino Studies

March 26, 2018

A device that can detect individual barium ions could be the heart of an experiment that takes the next step toward probing the nature of the neutrino.

Focus story on:

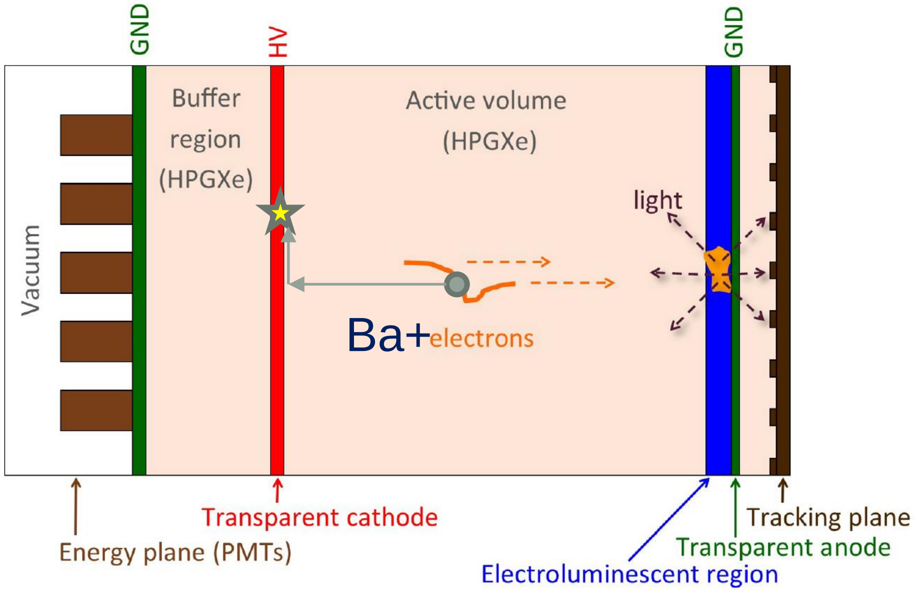
A.D. McDonald *et al.* (NEXT Collaboration)

Phys. Rev. Lett. **120**, 132504 (2018)

**First demonstration of single Ba++ ion resolution.**

# Next steps: Making it work in gas

- 1. Barium ion test beam
- 2. Barium drift characterization
- 3. Dry phase microscopy
- 4. Ion concentration to sensors
- 5. Dry SMFI molecule design
- 6. Combine into a working sensor for NEXT prototype




PROUDLY SUPPORTED BY:
U.S. DEPARTMENT OF ENERGY

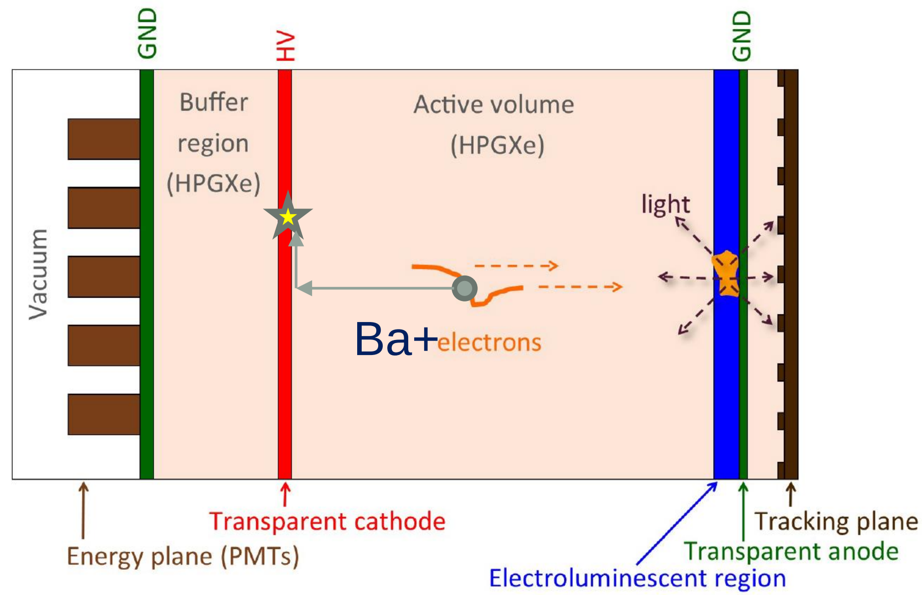
# Next steps: Making it work in gas

- 1. Barium ion test beam
- 2. Barium drift … cterization
- 3. D… … ase microscopy
- 4. Ion concentration to sensors

***Focus on new stuff here:***

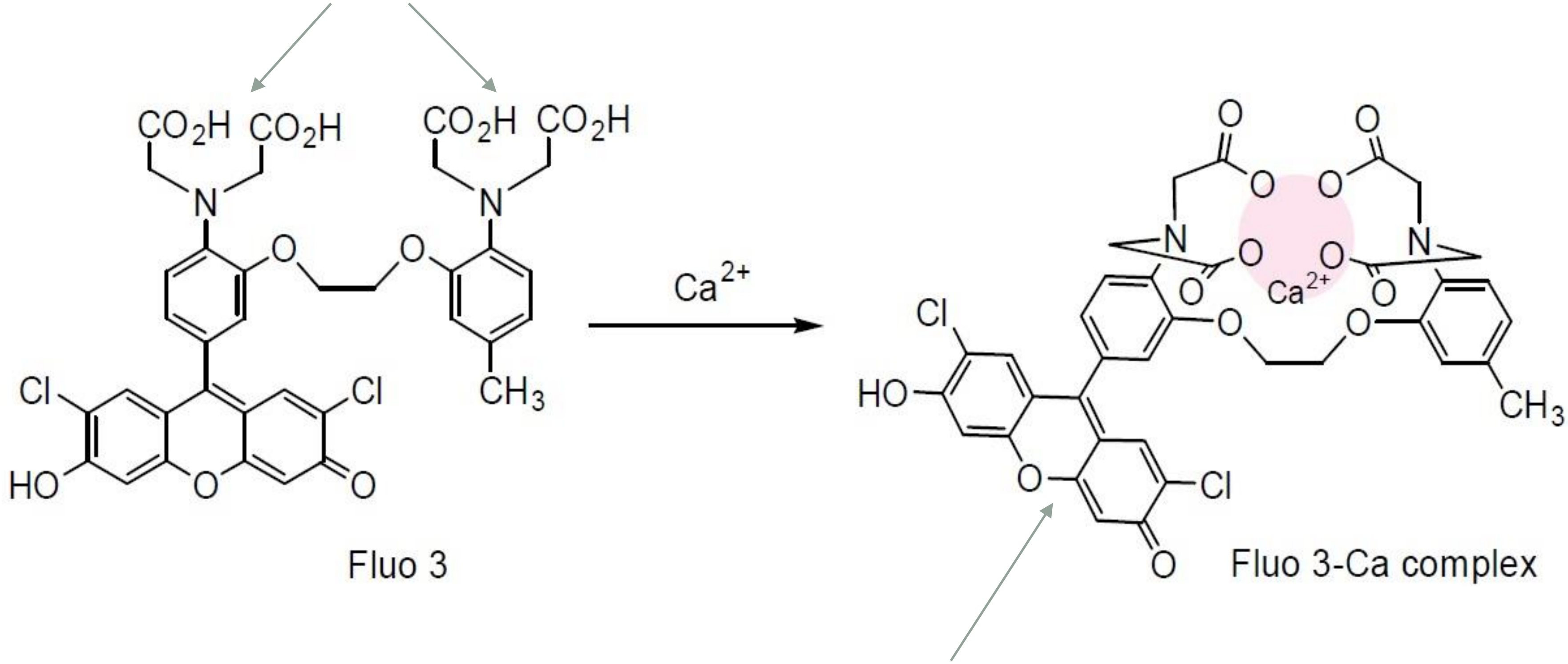
- 5. Dry SMFI molecule design
- 6. Combine into a working sensor for NEXT prototype

**No time to tell you about it all!**

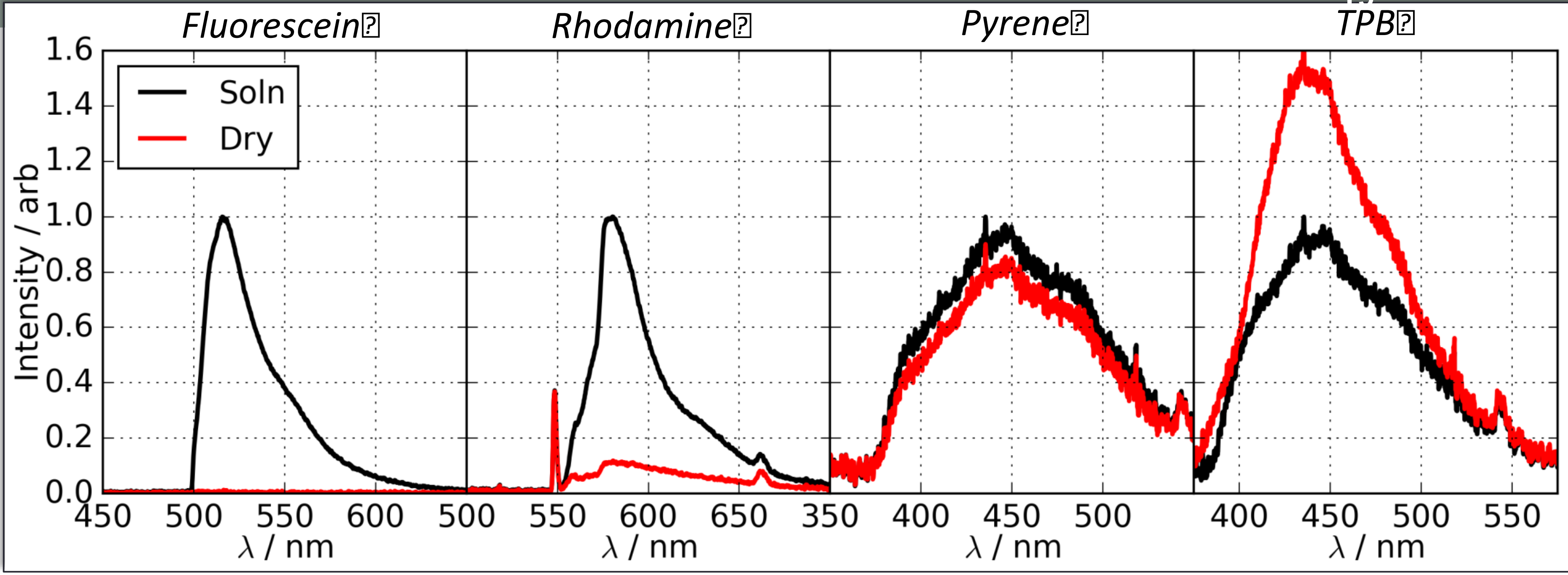


# Inadequacies of the FLUO family

Deprotonation of carboxylic acids is required to accept the ion – we observe the characteristic pH dependence of this in solution

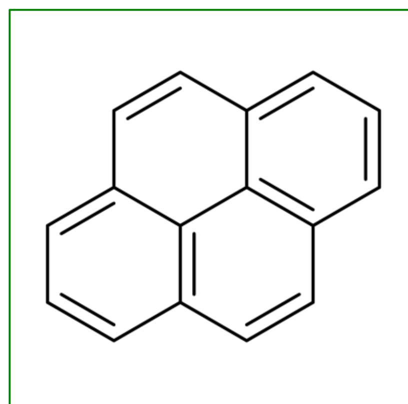


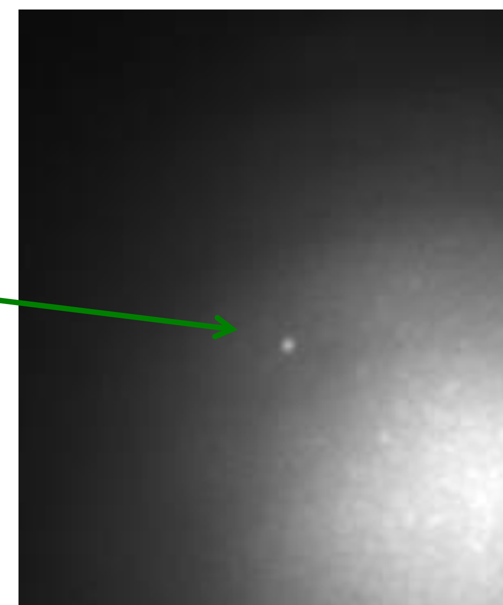

Fluorescein does not shine dry




Fluo-3 may not be ideal for HPGXe, since fluorescein dye is not bright in dry phase.

But pyrene works. We can resolve single molecules of it too.

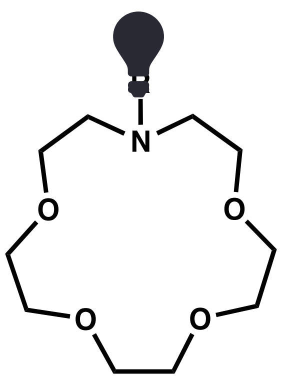




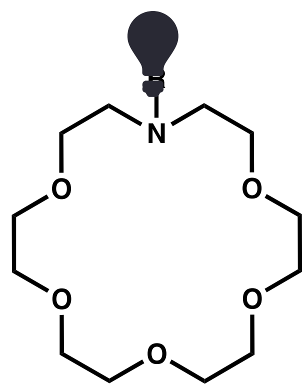
FOSS LAB
@ UTA

# Molecule Development

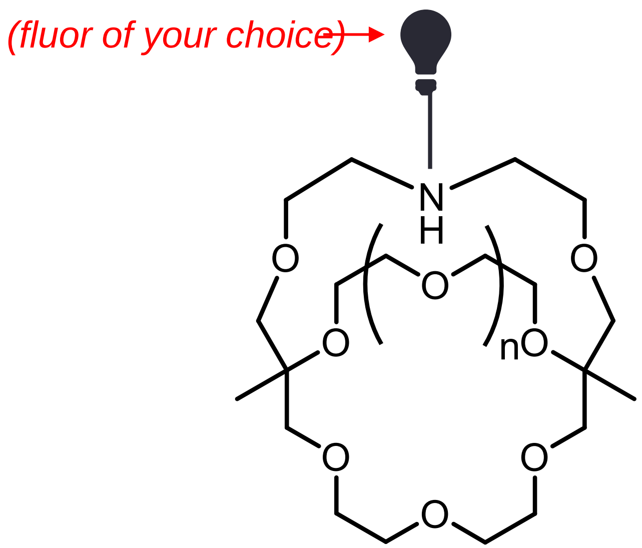
- As a first step we have targeted a systematic exploration of azo-crown and azo-cryptand derivatives.
- Showing you the first results from these studies today.
- Pyrene has been our first choice fluor. Due to the design of the synthesis, it is “plug and play” to install new ones.



**15-crown-5**



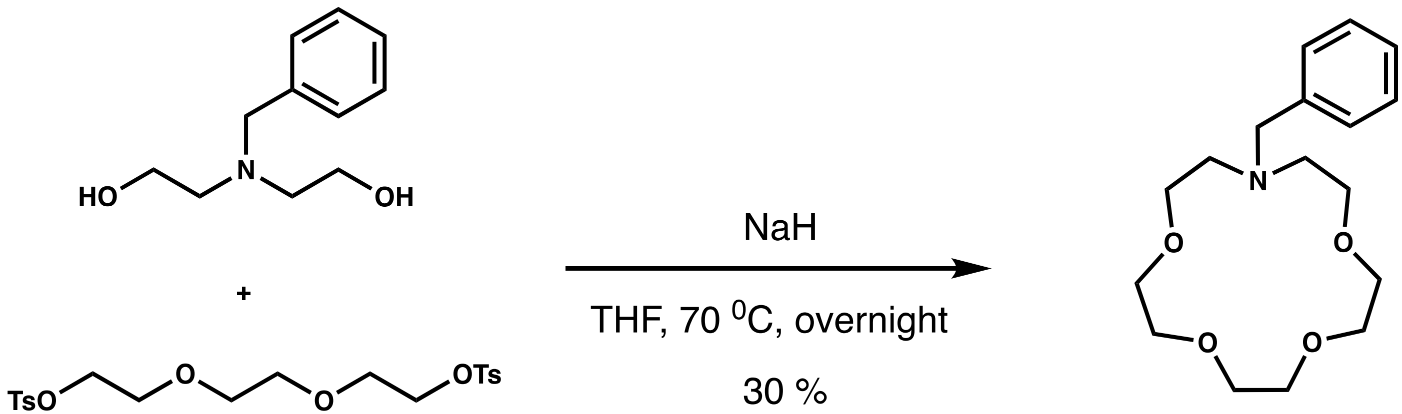
**18-crown-6**

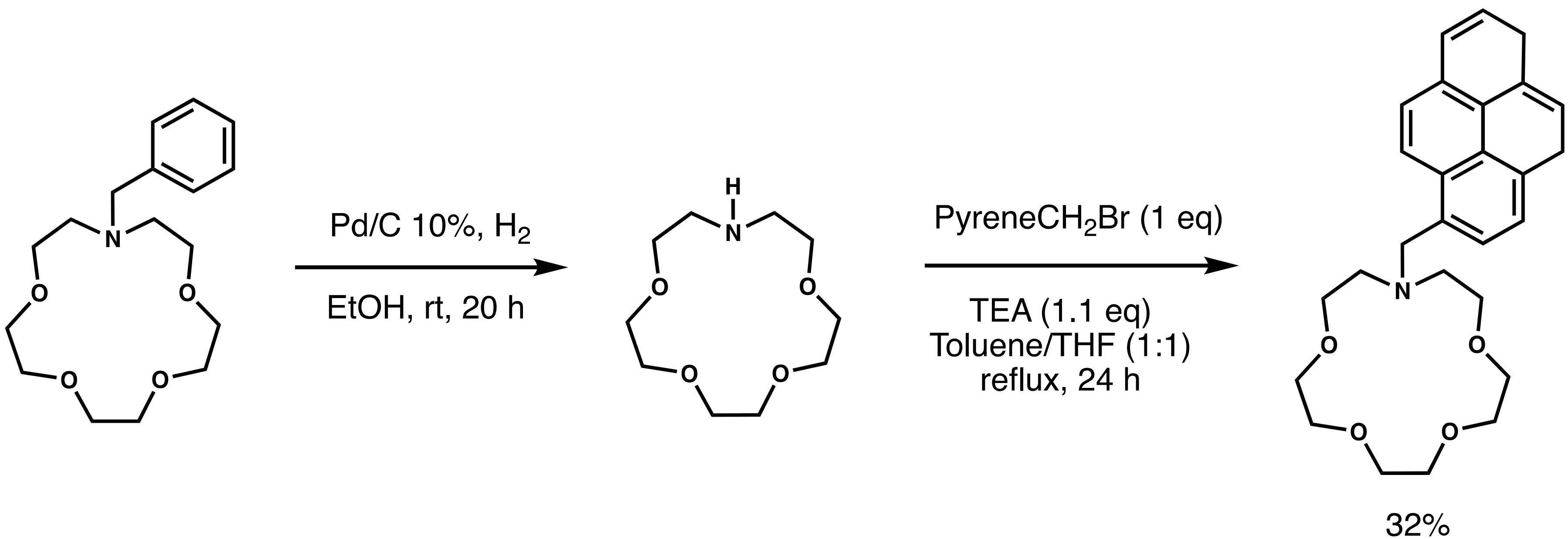



**MAC-NH (Monoaza Cryptand)**

## Example of the synthesis for 15c5:

FOSS LAB
@ UTA




***Then into the column to clean it up…***

# Installing Alternative Fluorophores

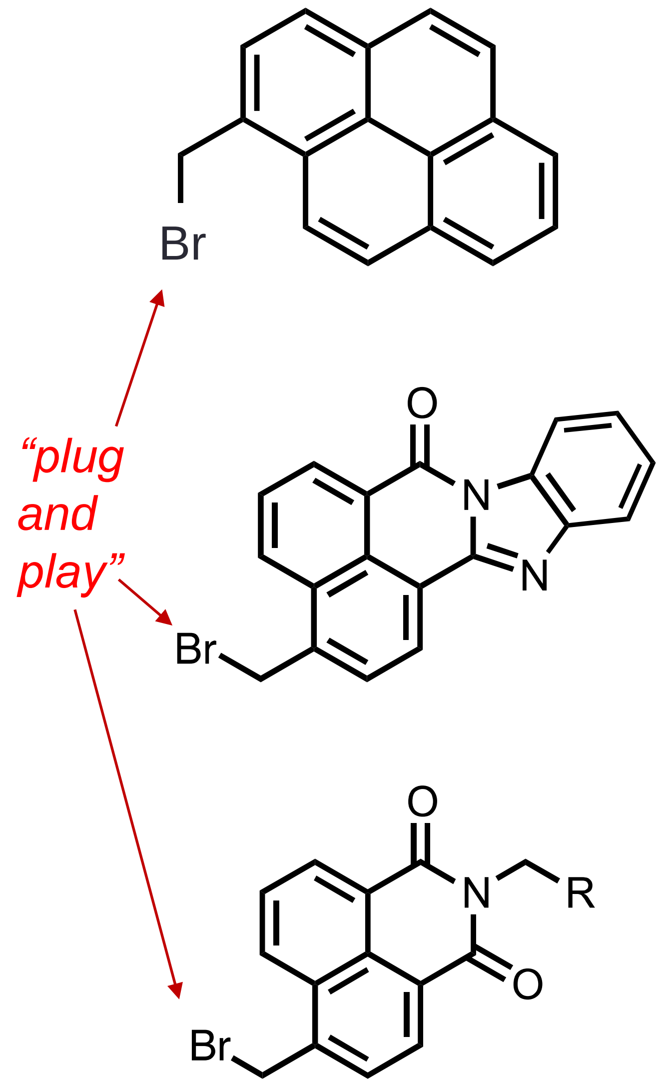
FOSS LAB @ UTA

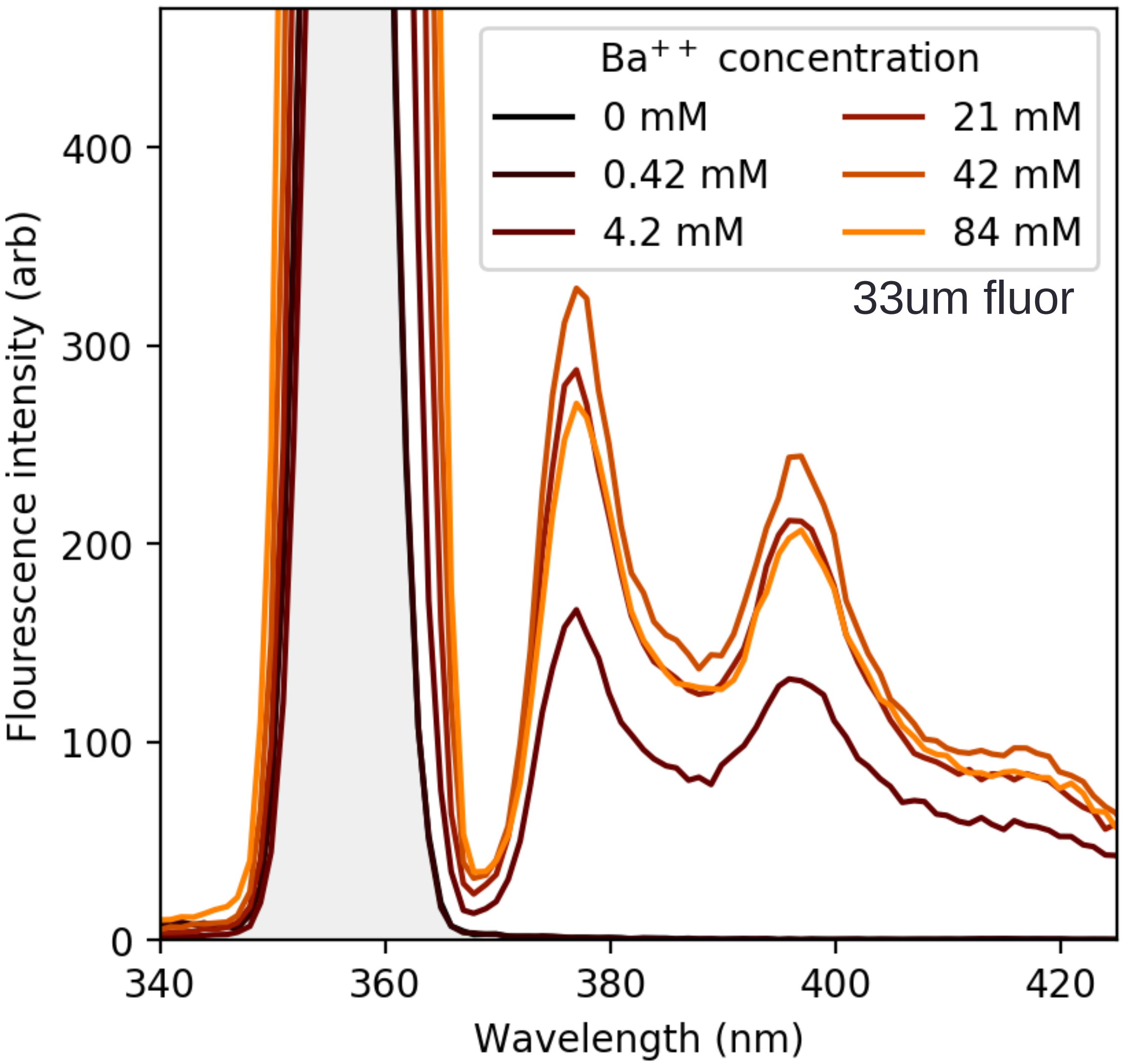
168 SINGLE MOLECULE FLUORESCENCE SPECTROSCOPY

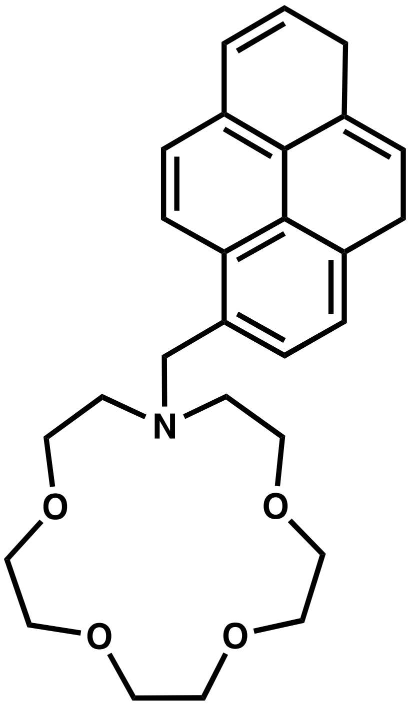
**Table 4.1** Photophysical properties of some common dyes with potential for single molecule fluorescence studies

| Fluorophore | $\lambda^{1p}_{ex}$ (nm) | $\lambda^{1p}_{em}$ (nm) | QY | $\varepsilon$ ($cm^{-1}M^{-1}$) | SS (nm) | $\tau_f$ (ns) | $\lambda^{2p}_{ex}$ (nm) | $\lambda^{2p}_{em}$ (nm) | Reference |
|---|---|---|---|---|---|---|---|---|---|
| FITC | 495 | 520 | 0.7 | 73,000 | 25 | — | 947 | 530 | [87–89] |
| FAM | 495 | 520 | 0.7 | 83,000 | 25 | — | — | — | [88] |
| TMR | 554 | 585 | 0.2–0.5 | 95,000 | 31 | 2.1 | 849 | 570 | [5,90,91] |
| R6G | 530 | 556 | — | 105,000 | 26 | — | — | — | [92] |
| Cy2 | 489 | 506 | — | — | 17 | — | 905 | 520 | [87,93] |
| Cy3 | 550 | 570 | 0.14 | 150,000 | 20 | ~1 | 1032 | 578 | [3,87,90] |
| Cy5 | 650 | 670 | 0.15 | 250,000 | 20 | ~1 | — | — | [3,5,90] |
| Cy5.5 | 675 | 694 | — | 250,000 | 19 | — | — | — | [3] |
| Cy7 | 743 | 767 | 0.02 | 250,000 | 24 | ~0.8 | — | — | [3,90,94] |
| ECFP | 458 | 472 | 0.4 | 26,000 | 14 | — | — | — | [94] |
| EGFP | 395,470 | 509 | 0.8 | 30,000 | 39 | 3.2 | — | — | [5,90,94] |
| EYFP | 514 | 527 | 0.6 | 84,000 | 13 | 3.7 | — | — | [90,94] |
| DsRed | 532 | 582 | 0.29 | 22,500 | 50 | 2.8 | — | — | [90] |
| Bodipy Fl | 504 | 510 | — | 70,000 | 6 | — | 920 | 526 | [87,88] |
| Bodipy R6G | 528 | 547 | — | 70,000 | 19 | — | — | — | [88] |
| AF488 | 495 | 520 | 0.5–0.9 | 80,000 | 25 | — | 985 | 530 | [5,87] |
| AF546 | 554 | 570 | — | 112,000 | 16 | — | 1028 | 582 | [87,95] |
| AF555 | 555 | 565 | — | 150,000 | 10 | — | — | — | [3] |
| AF594 | 590 | 617 | — | 92,000 | 27 | — | 1074 | 619 | [87,95] |
| AF633 | 632 | 647 | — | 100,000 | 15 | 3.2 | — | — | [3,90] |

*Handbook of Single Molecule Fluorescence*, Gell, C. Brockwell, D. Smith, A. OUP Oxford, 2006, New York p. 168-9.



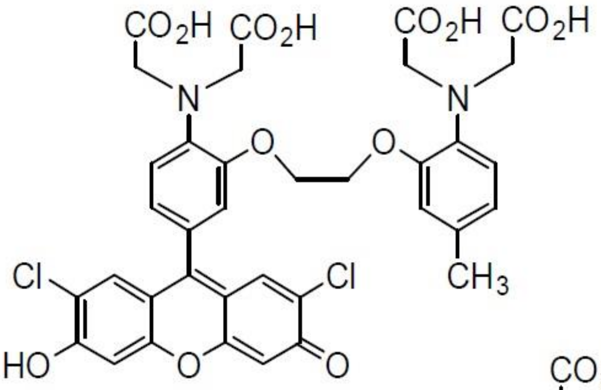


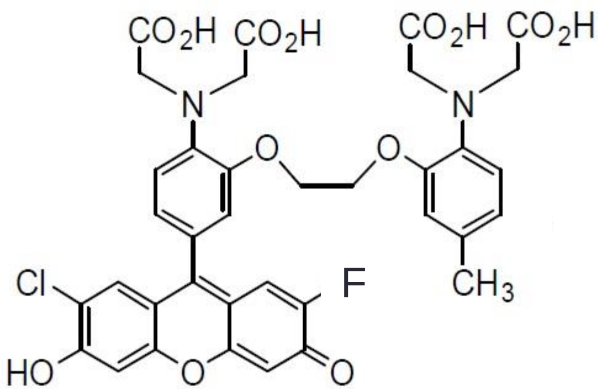



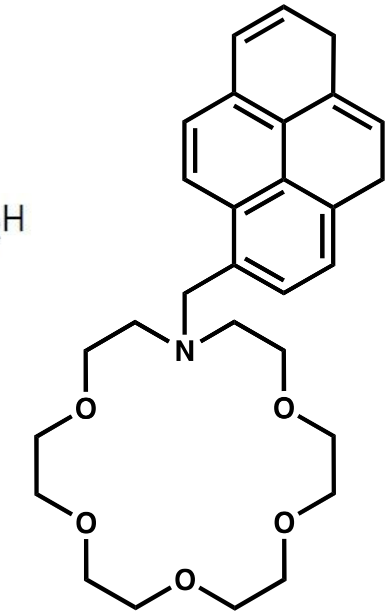

"NEXT-2"

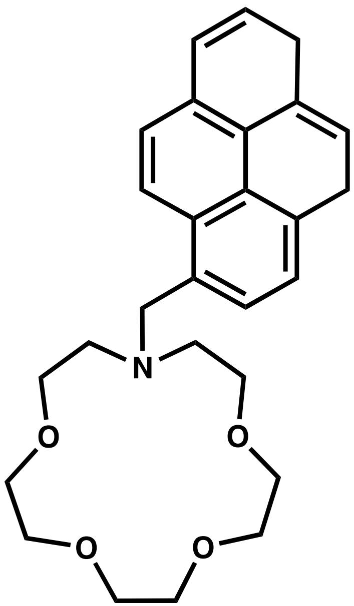
**Home made fluorophore with extremely strong off/on response!**

*(preliminary numbers – still optimizing…)*



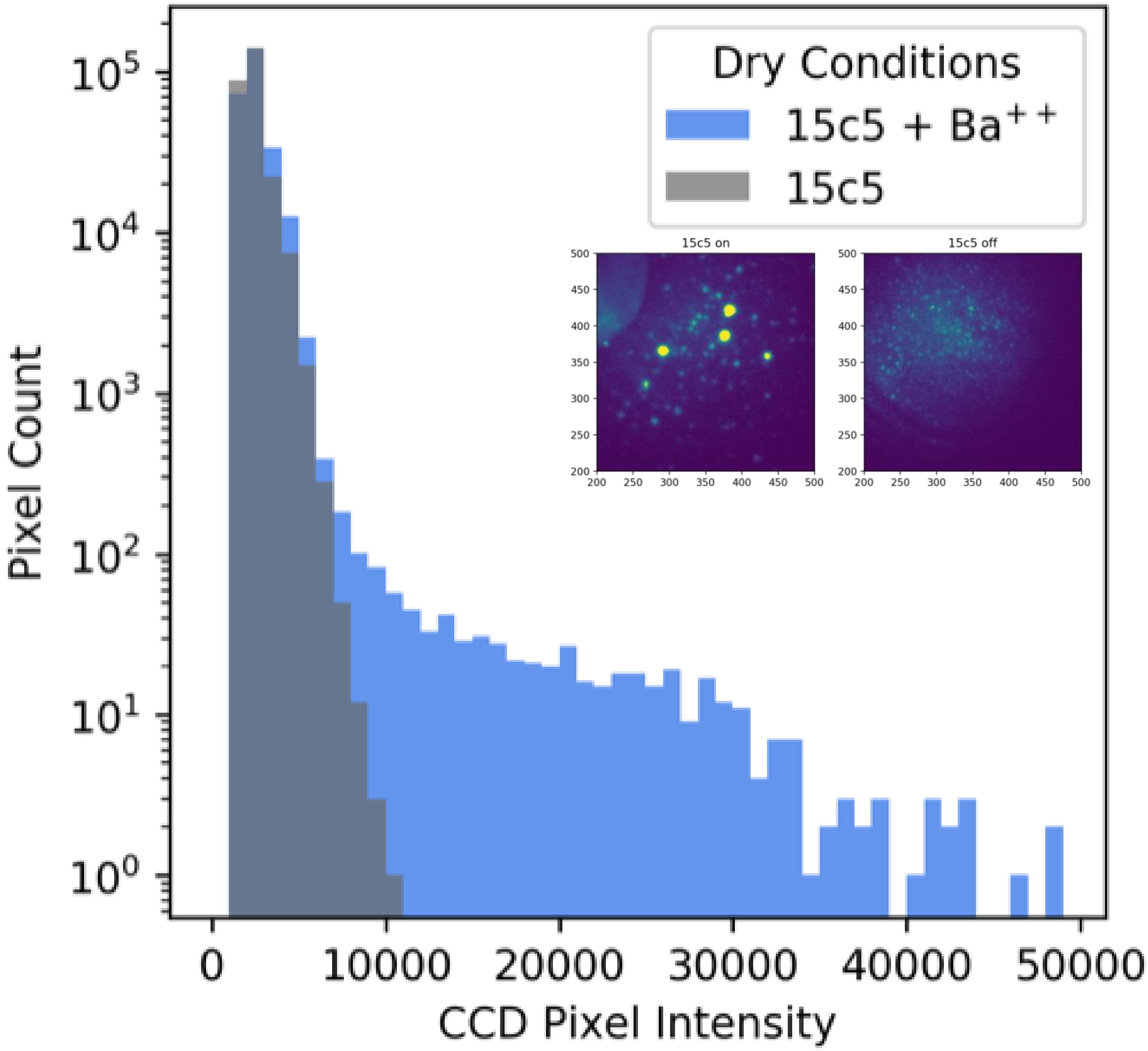

| | BAPTA, fluorescein-Cl | BAPTA, fluorescein-F | 18c6, pyrene: | 15c5, pyrene: |
|---|---|---|---|---|
| | **FLUO-3** | **FLUO-4** | **NEXT-1** | **NEXT-2** |
| **Max. Frac. $Ba^{++}$ Response:** | **17 x** | **85 x** | **6 x** | **205 x** |

# “Dry” SMFI response with NEXT-2:

Dry microscopy on chelated samples shows strong on/off behavior.

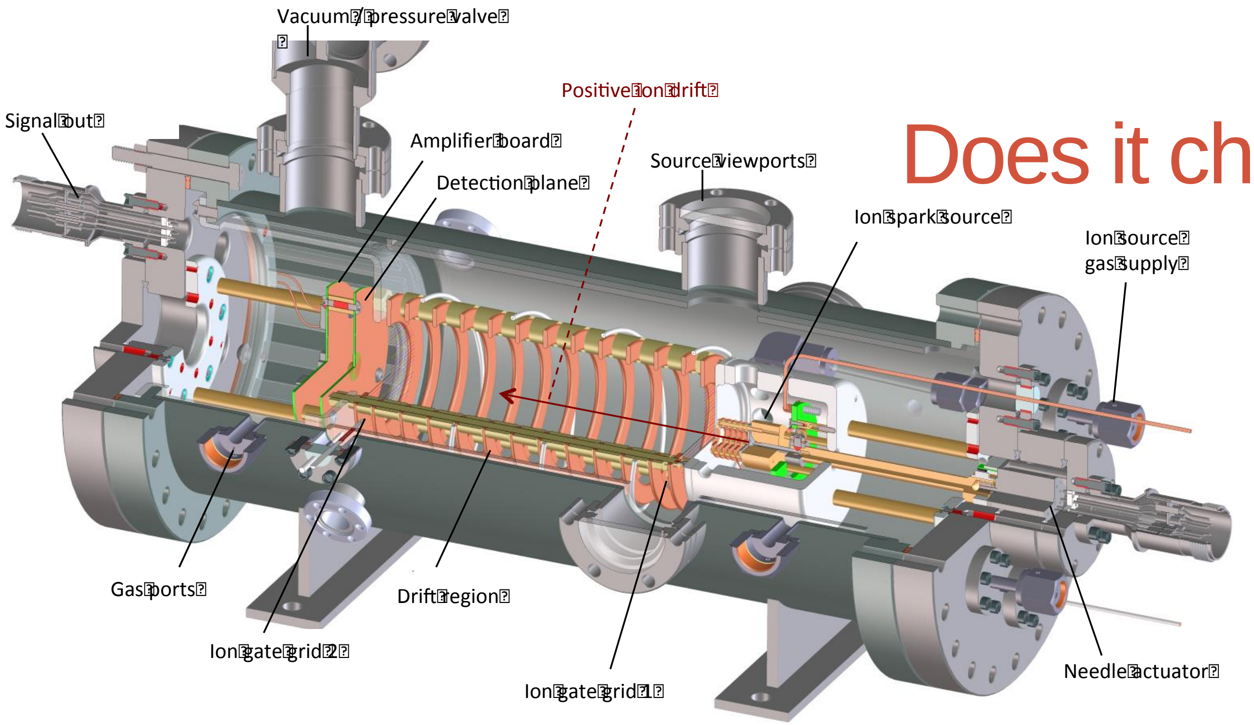
This has not been achievable with commercial fluorophores.

**First demonstration of dry Ba++ fluorescent chemo-sensor.**

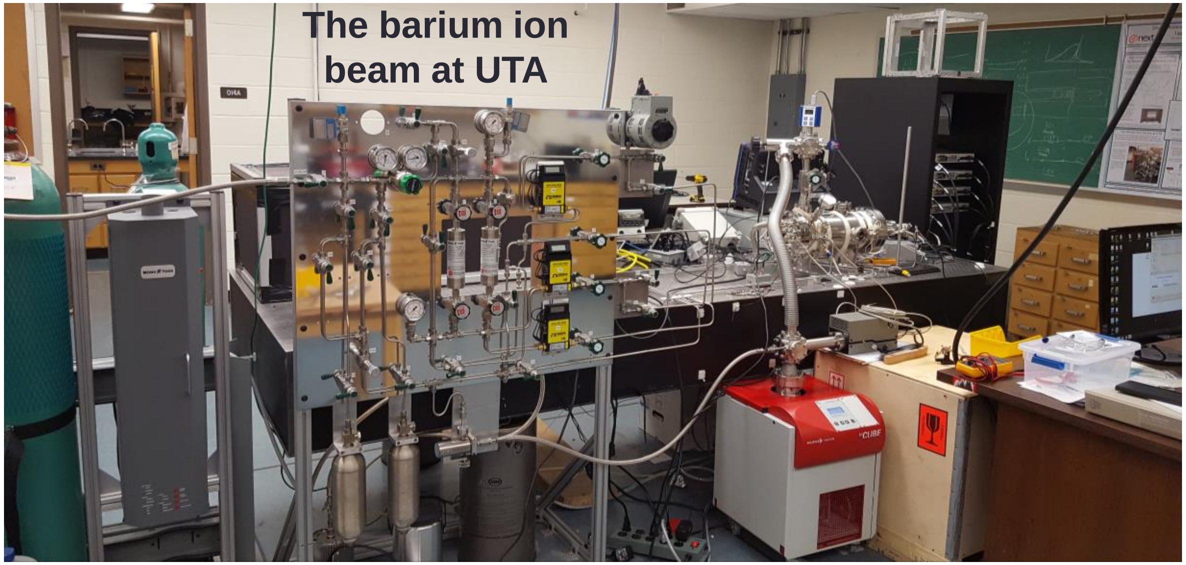


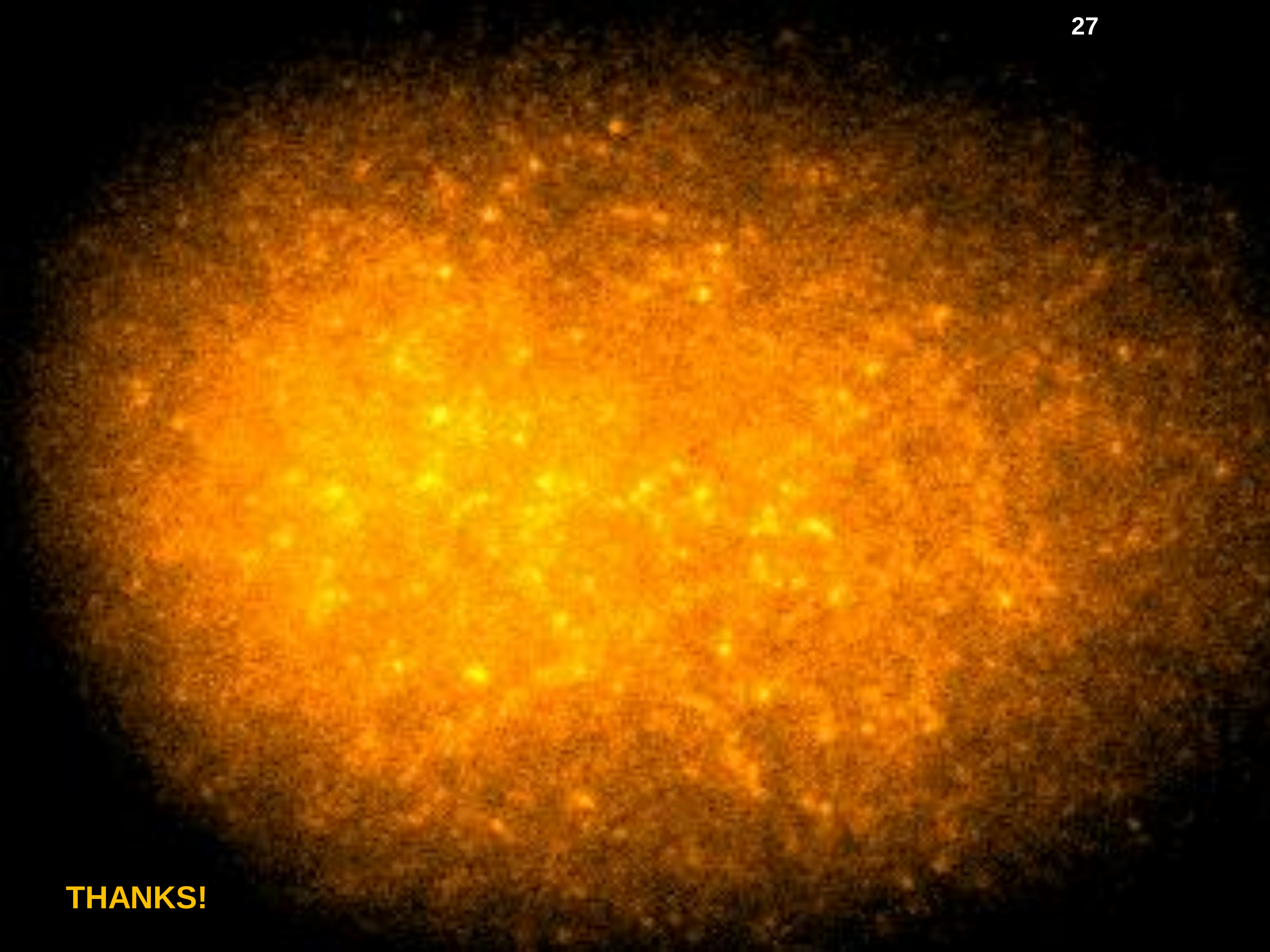
# It shines dry.

# Does it chelate $Ba^{++}$ dry?




***Implement a sensor based on NEXT-2 molecule in barium ion beam to find out!***

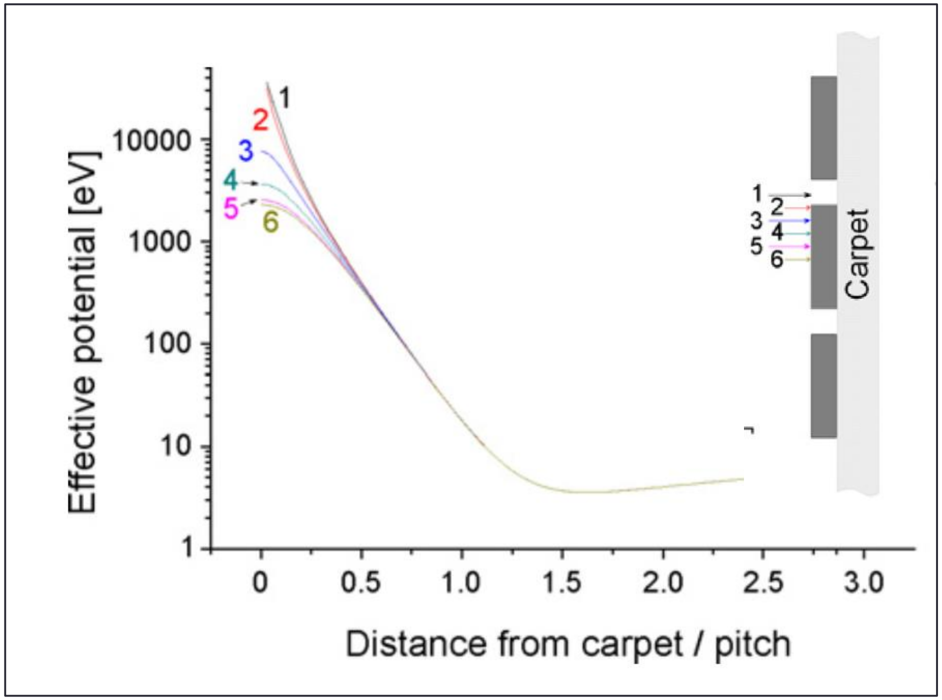


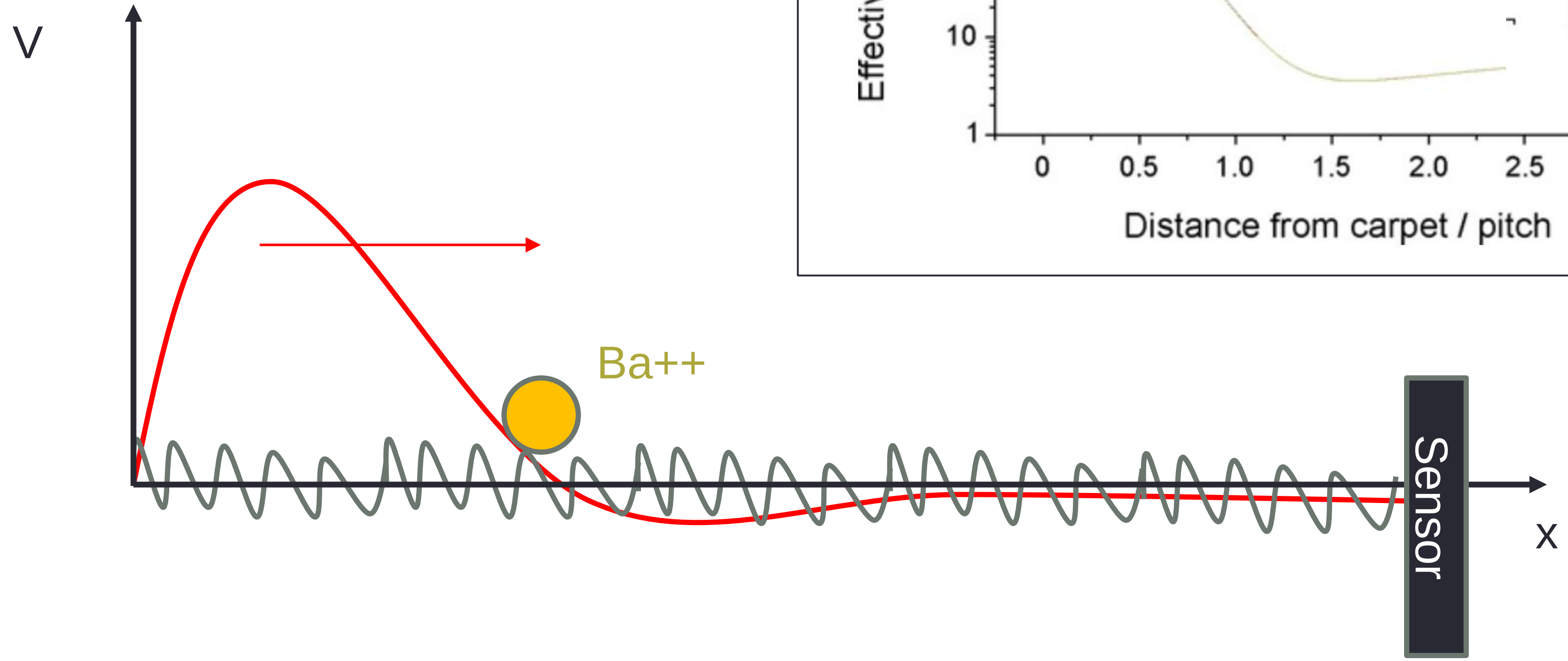


**THANKS!**

# RF Carpet fields

***International Journal of Mass Spectrometry 299 (2011) 71–77***



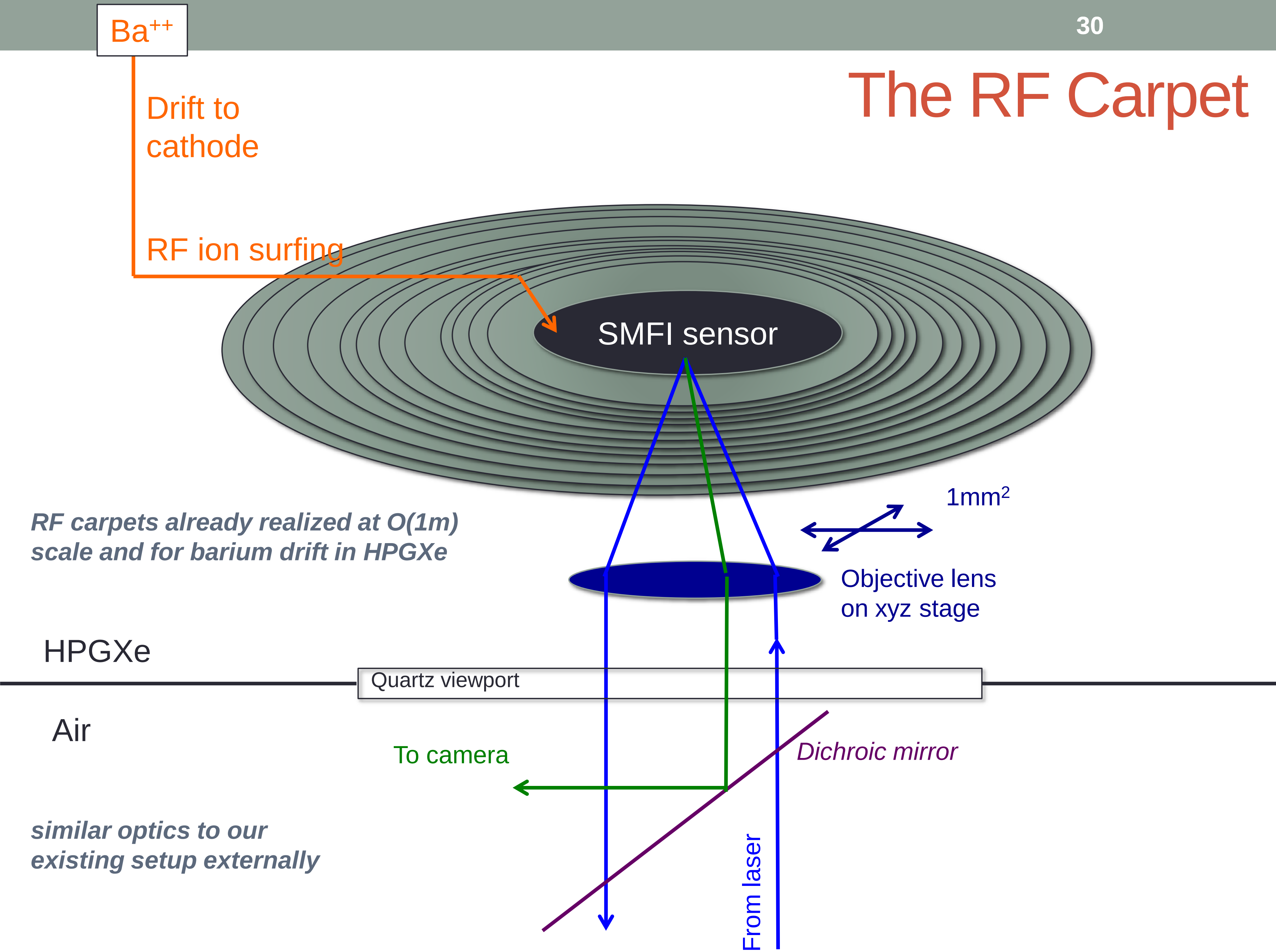




Superposed fields:

RF creates effective potential that levitates ions without neutralization

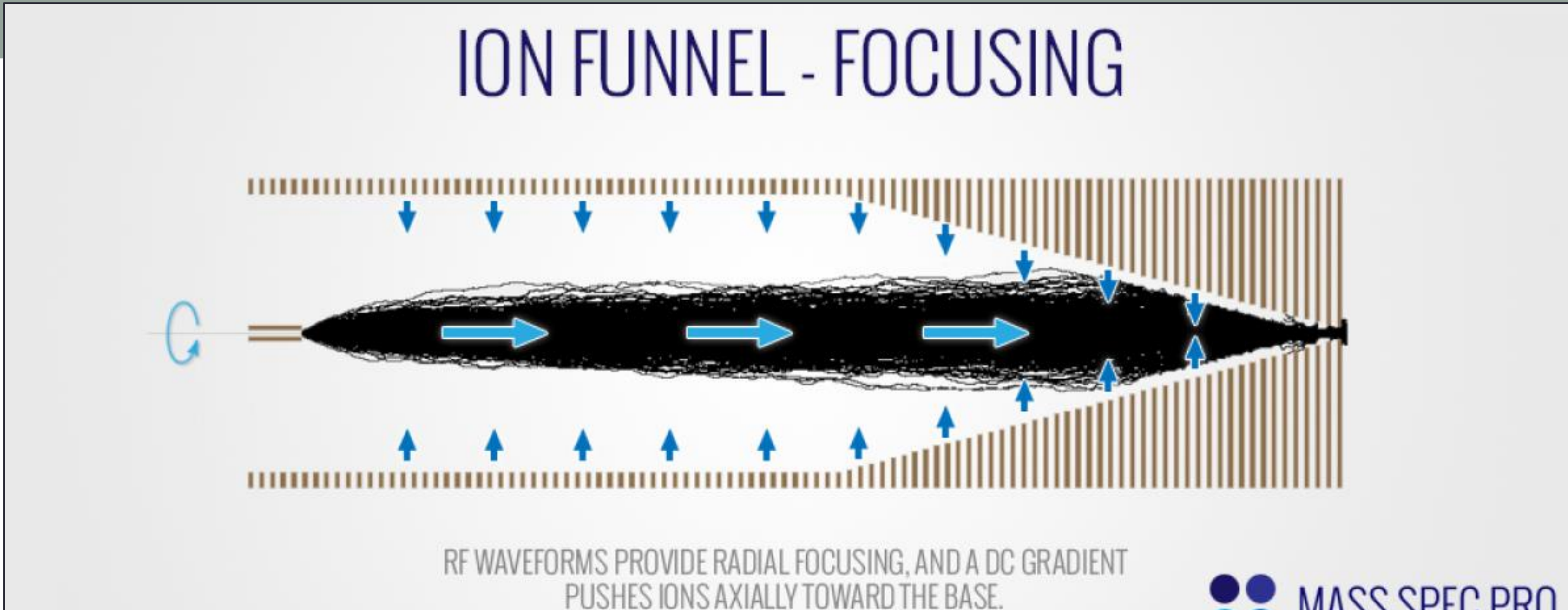
Surfing field sweeps ions to center,

# The RF Carpet

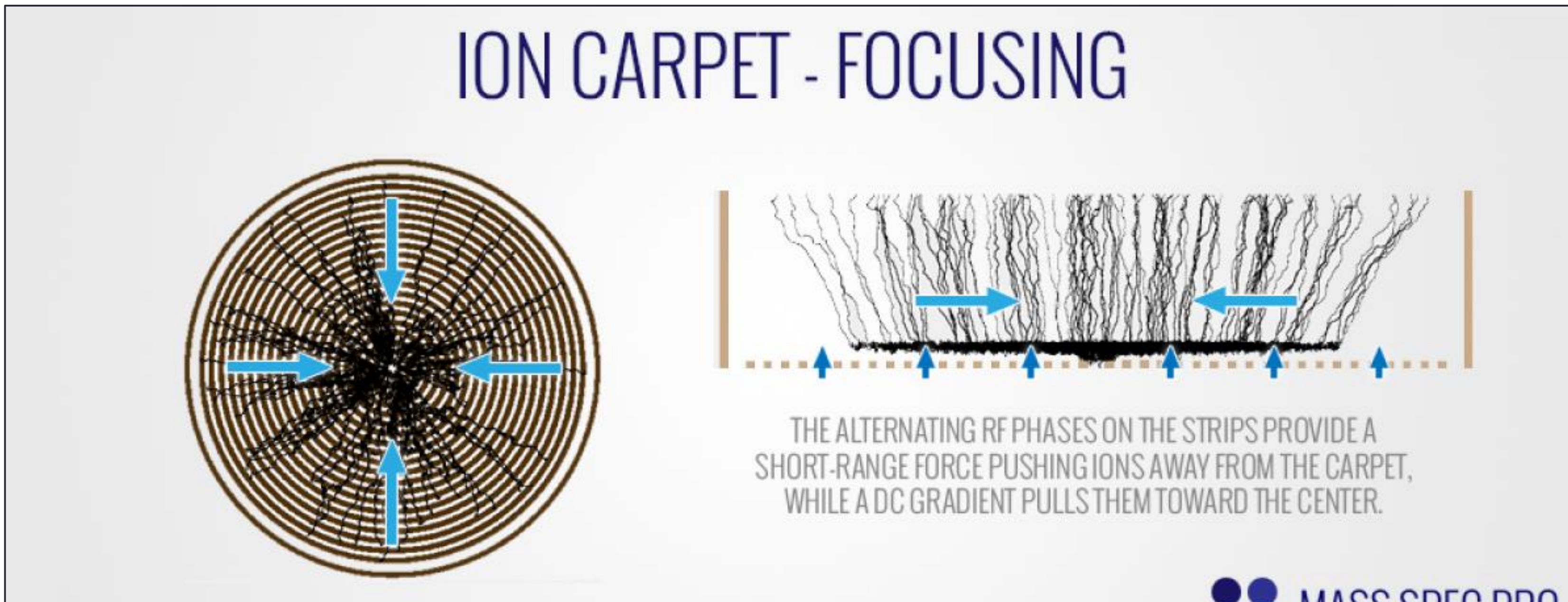



***RF carpets already realized at O(1m) scale and for barium drift in HPGXe***

***similar optics to our existing setup externally***



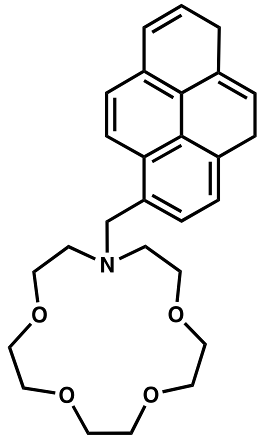

**As explored by Brunner / nEXO for extraction to trap**




**We plan to prototype and demonstrate a small (5cm) RF carpet at UTA**

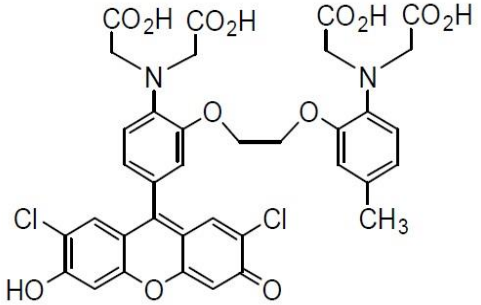
# Names:

- This molecule is called:
  - 13-pyrenylmethyl-1,4,7,10-tetraoxa-13-aza-cyclopentadecane



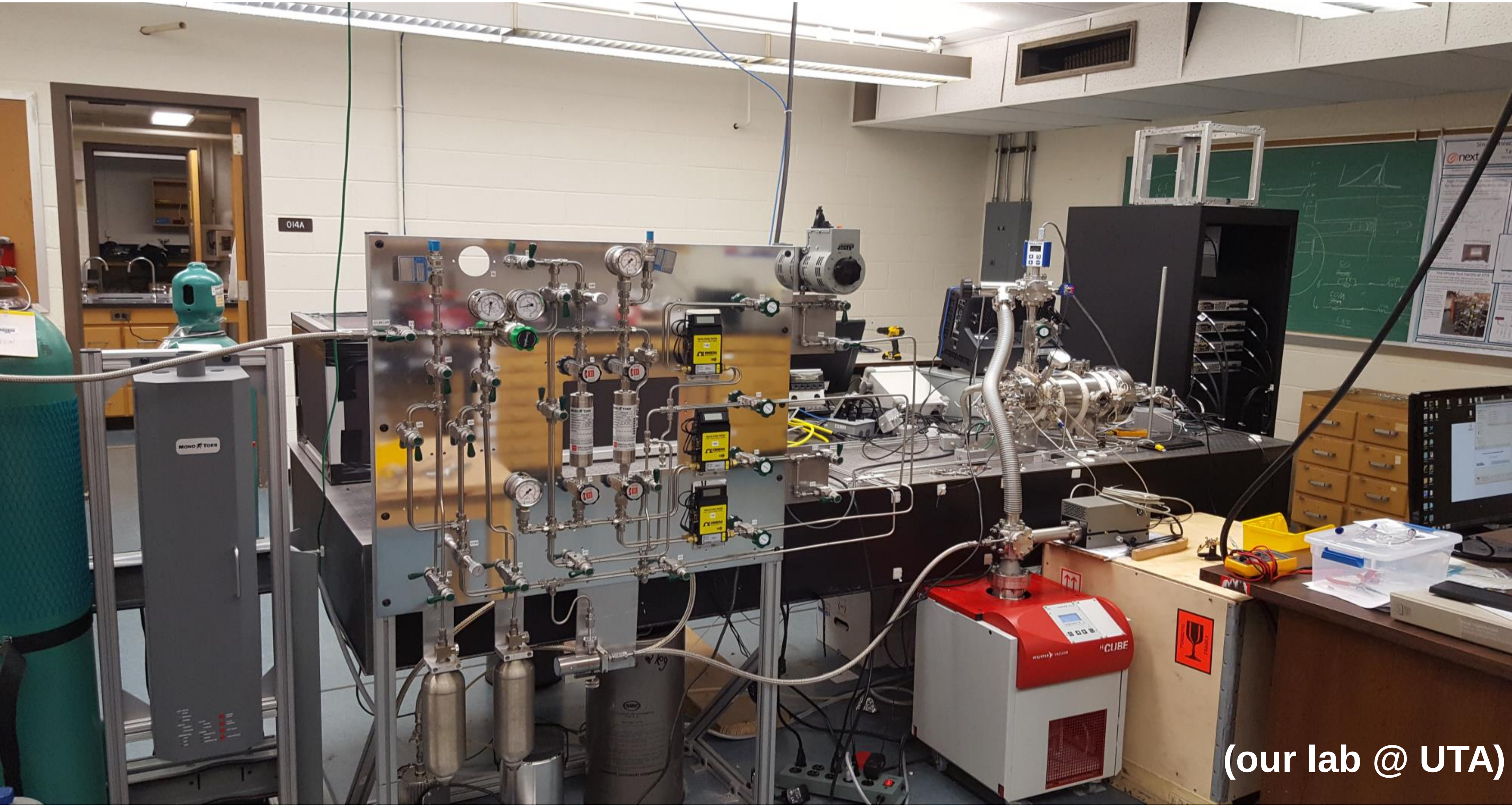

- Under IUPAC rules we may also call it:
  - N-prenylmethyl-azo-15-crown-5
- "Standard" in SMFI is 4-letter word and number
  - {[2-(2-{2-[Bis(carboxymethyl)amino]-5-(2,7-dichloro-6-hydroxy-3-oxo-3*H*-xanthen-9-yl)phenoxy}ethoxy)-4-methylphenyl](carboxymethyl)amino}acetic acid

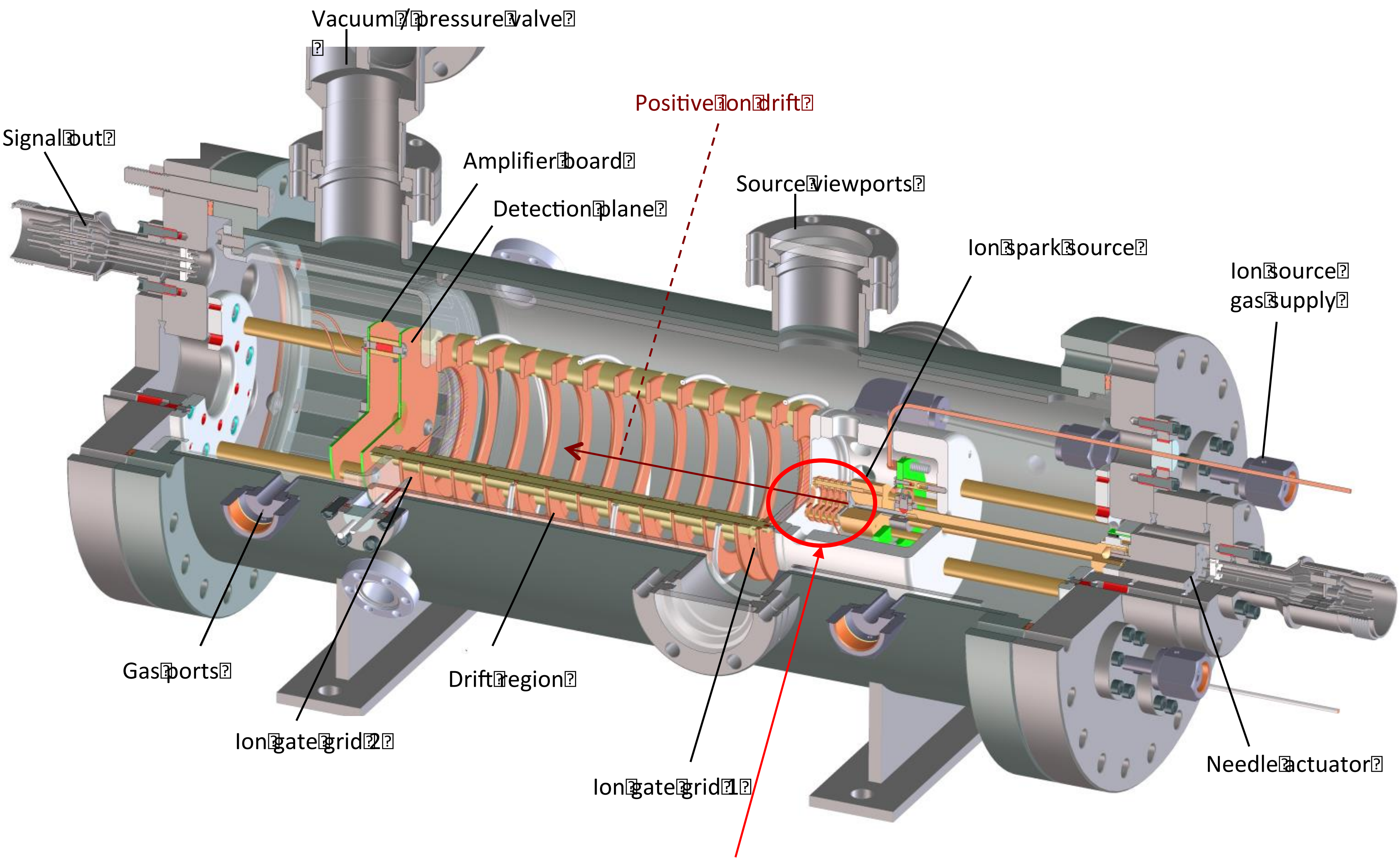
→ **FLUO-3**



# The barium beam

- The next major step is to test barium sensing dyes in HPGXe environment
- Expect better performance than in solution from both energetics and reactivity considerations

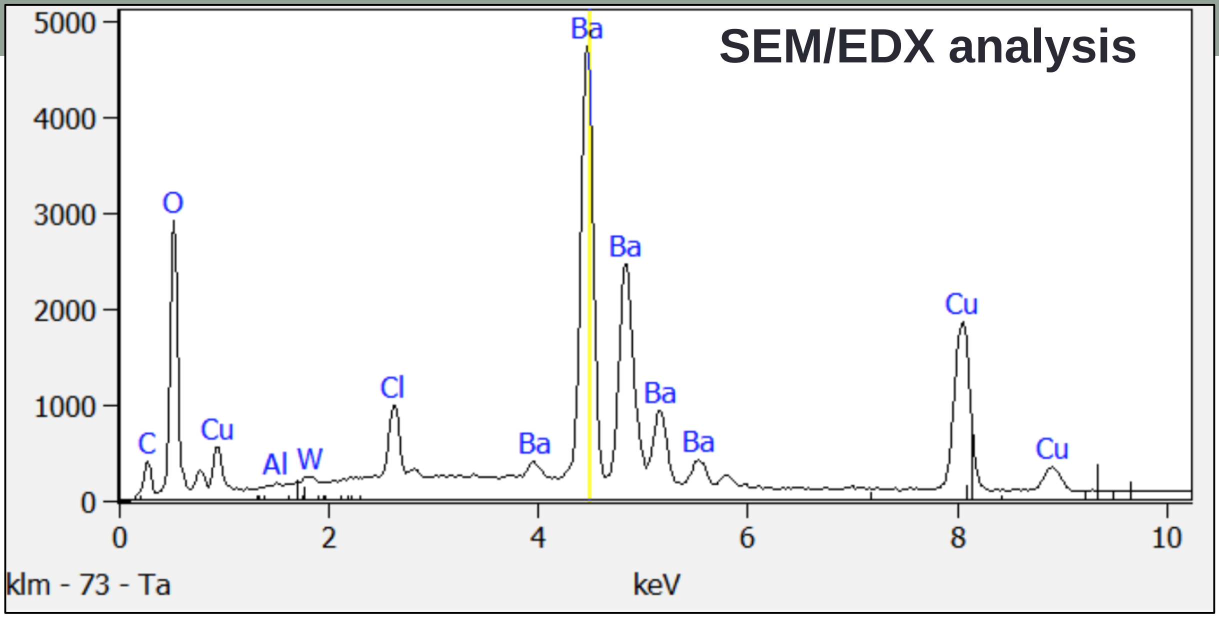




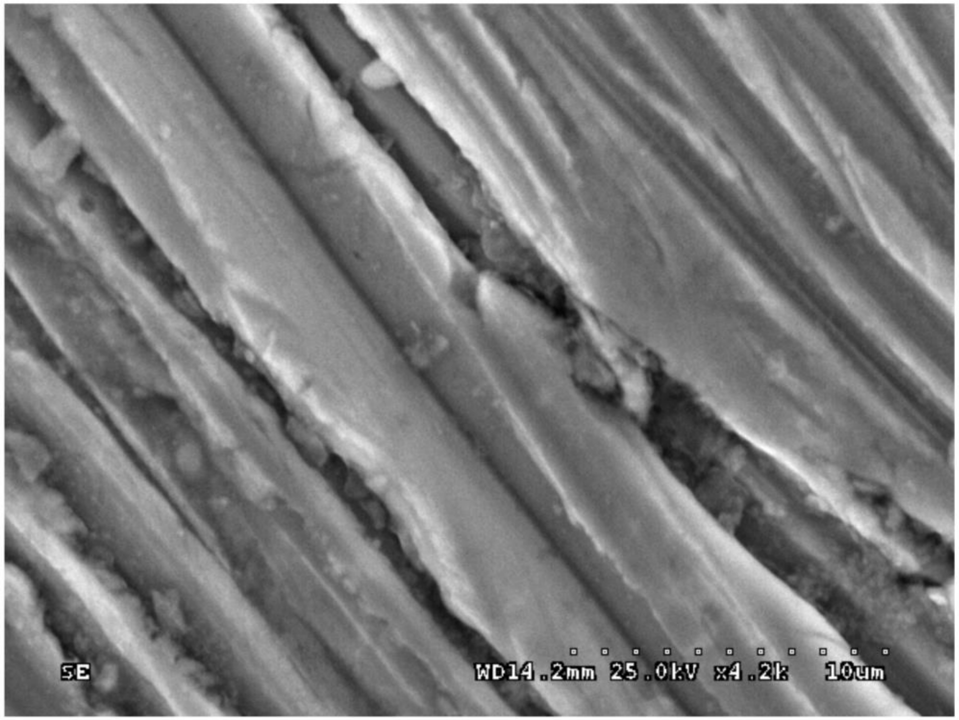

Barium or barium coated needle goes here

Custom electroplating method deposits a stable barium-rich coating onto copper for spark source from methanol.

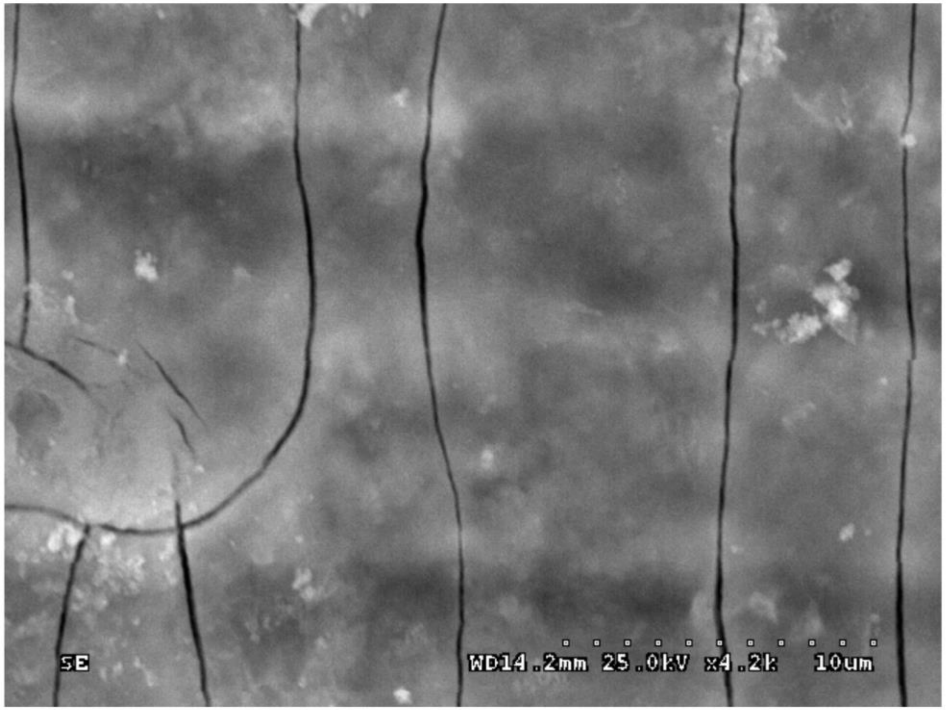
Removes difficulty of barium metal handling.




## Bare copper



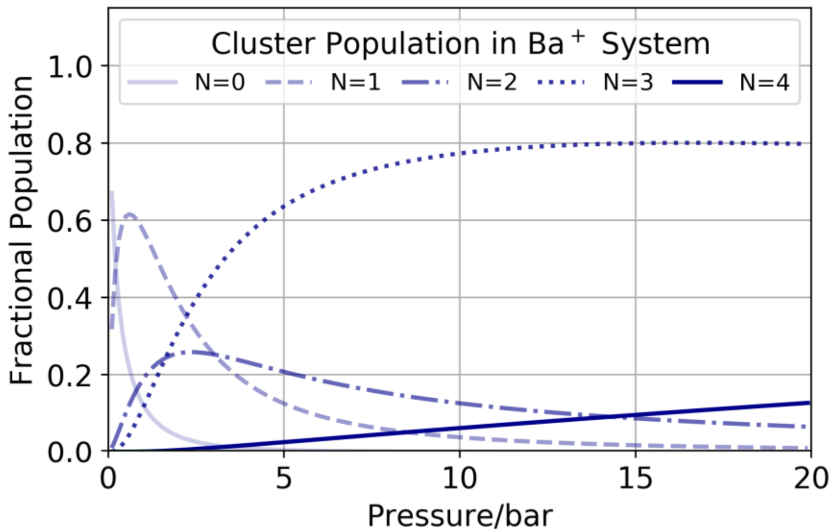

## Barium electroplated copper



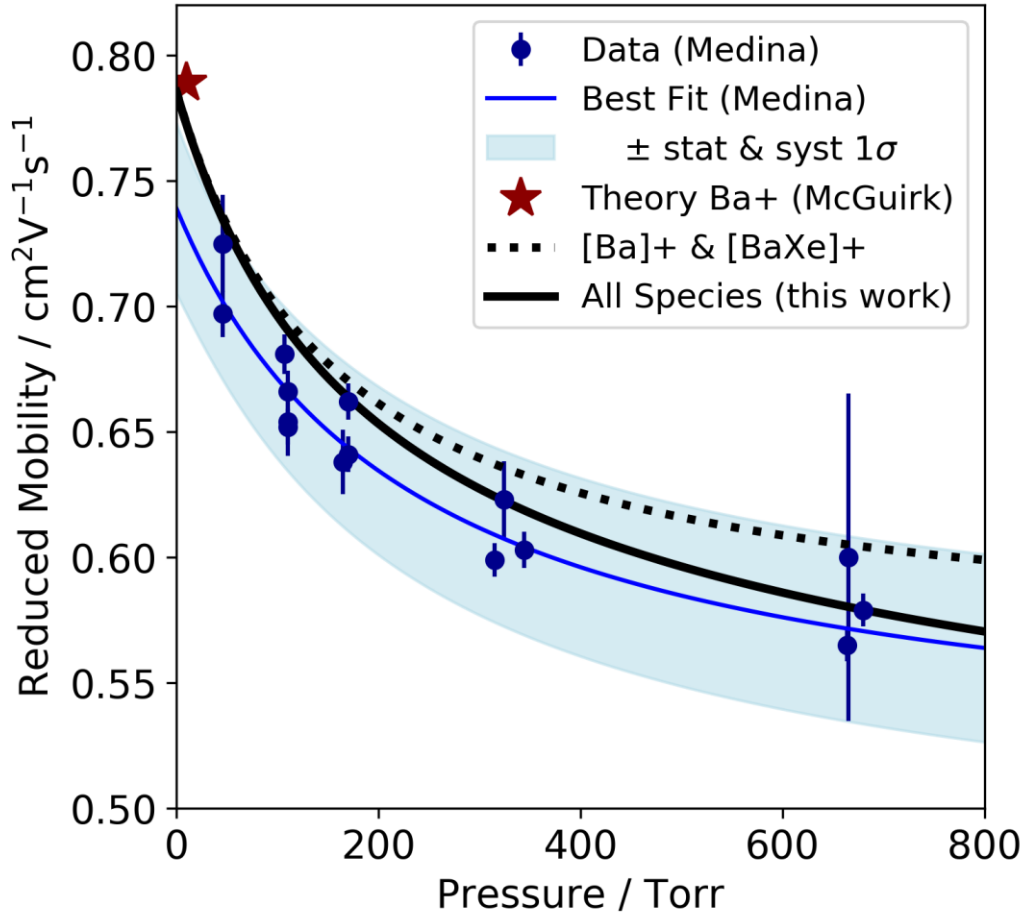
# Barium mobility in gas – in theory:

Not a single ion drifting!

$$Ba^+ + Xe + Xe \longleftrightarrow BaXe^+ + Xe$$




**Excellent agreement with data for Ba+**

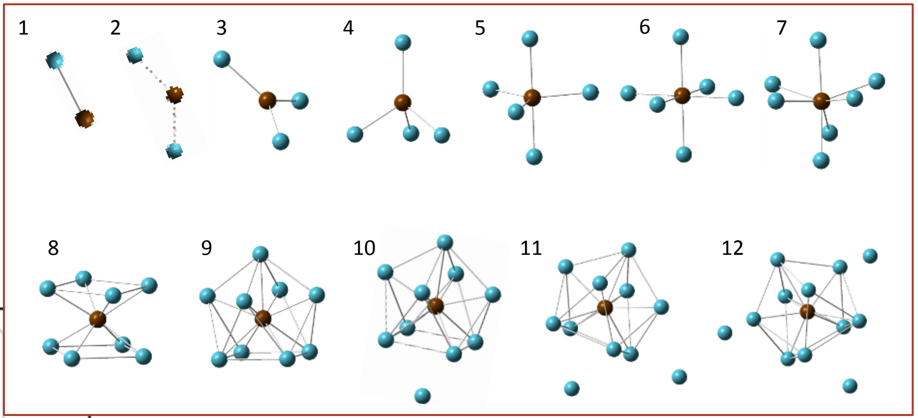


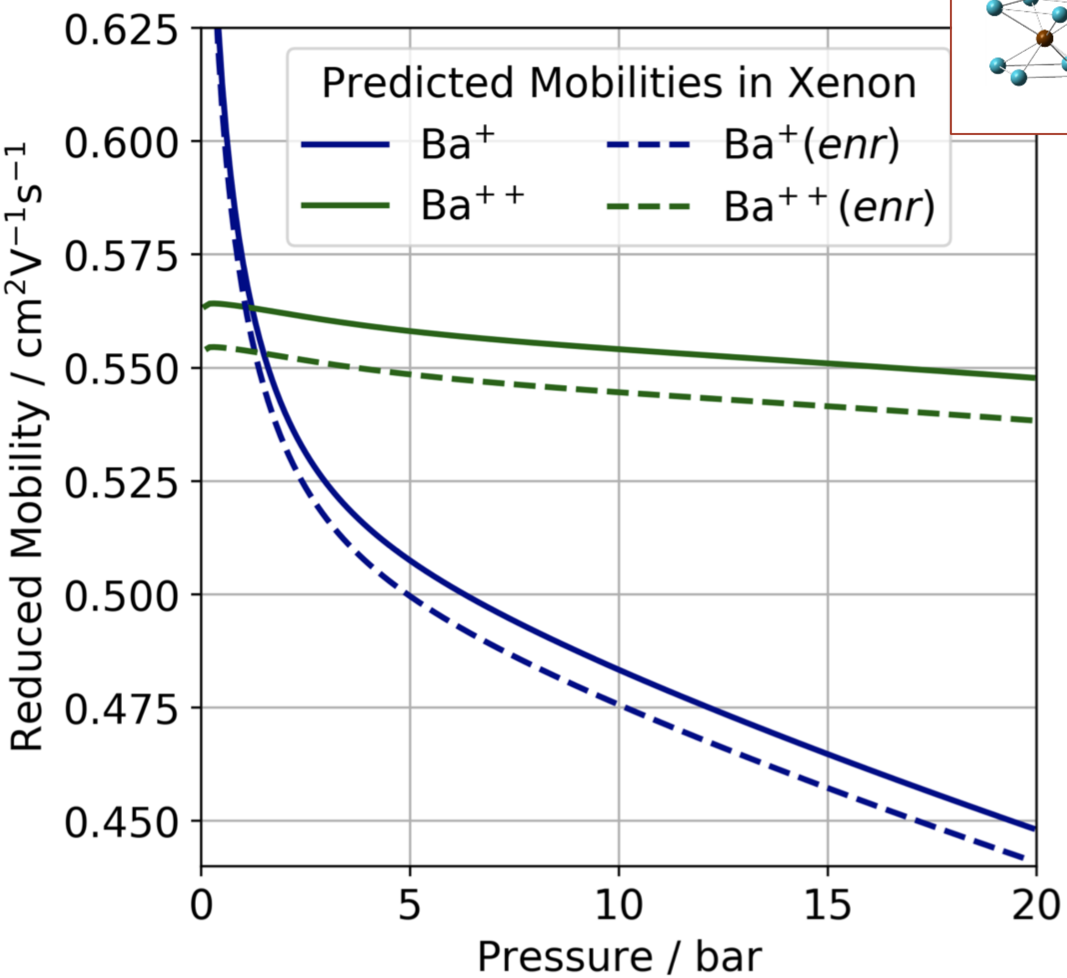


**Mobility and Clustering of Barium Ions and Dications in High Pressure Xenon Gas**
E. Bainglass, B.J. P. Jones, et. al. Phys.Rev. A97 (2018) no.6, 062509

# For Ba++ things get more complicated.

***Calculated Ba++ clusters:***



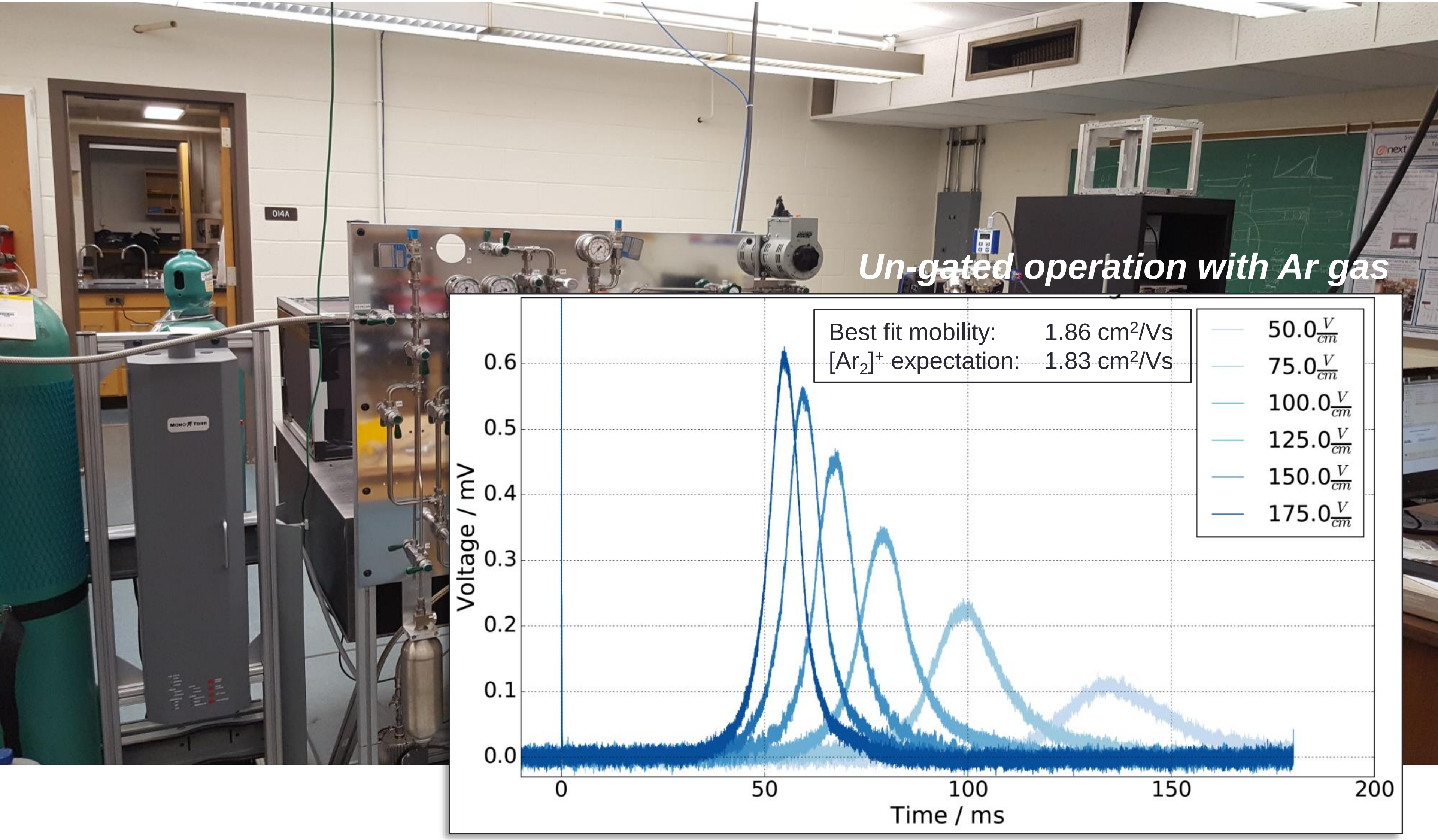



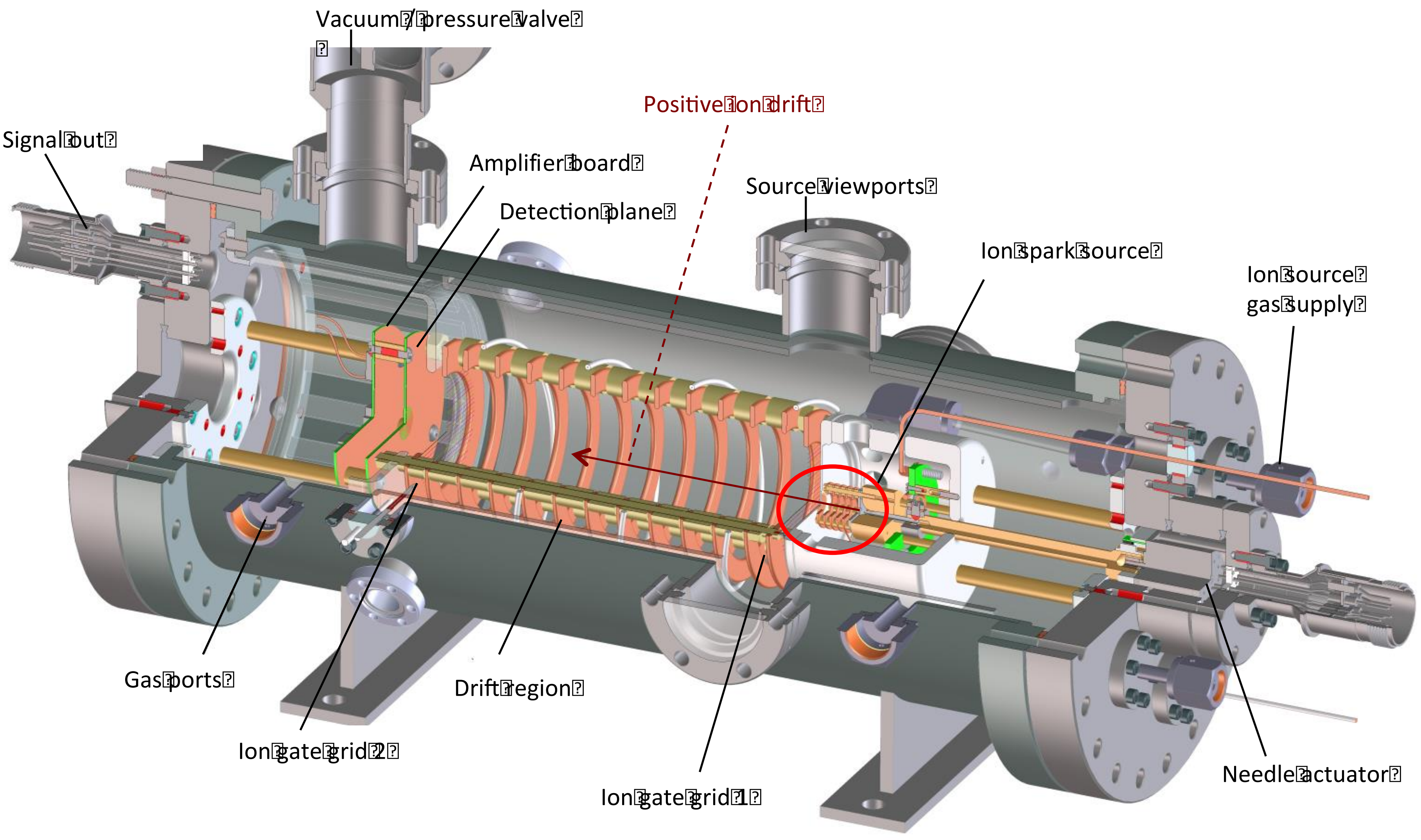

Bigger clusters more similar to each other, so less pressure dependence in Ba++ than Ba+

Isotopic composition changes scattering kinematics, so %-level differences with enriched xenon

**We will test this with experimental data soon!**

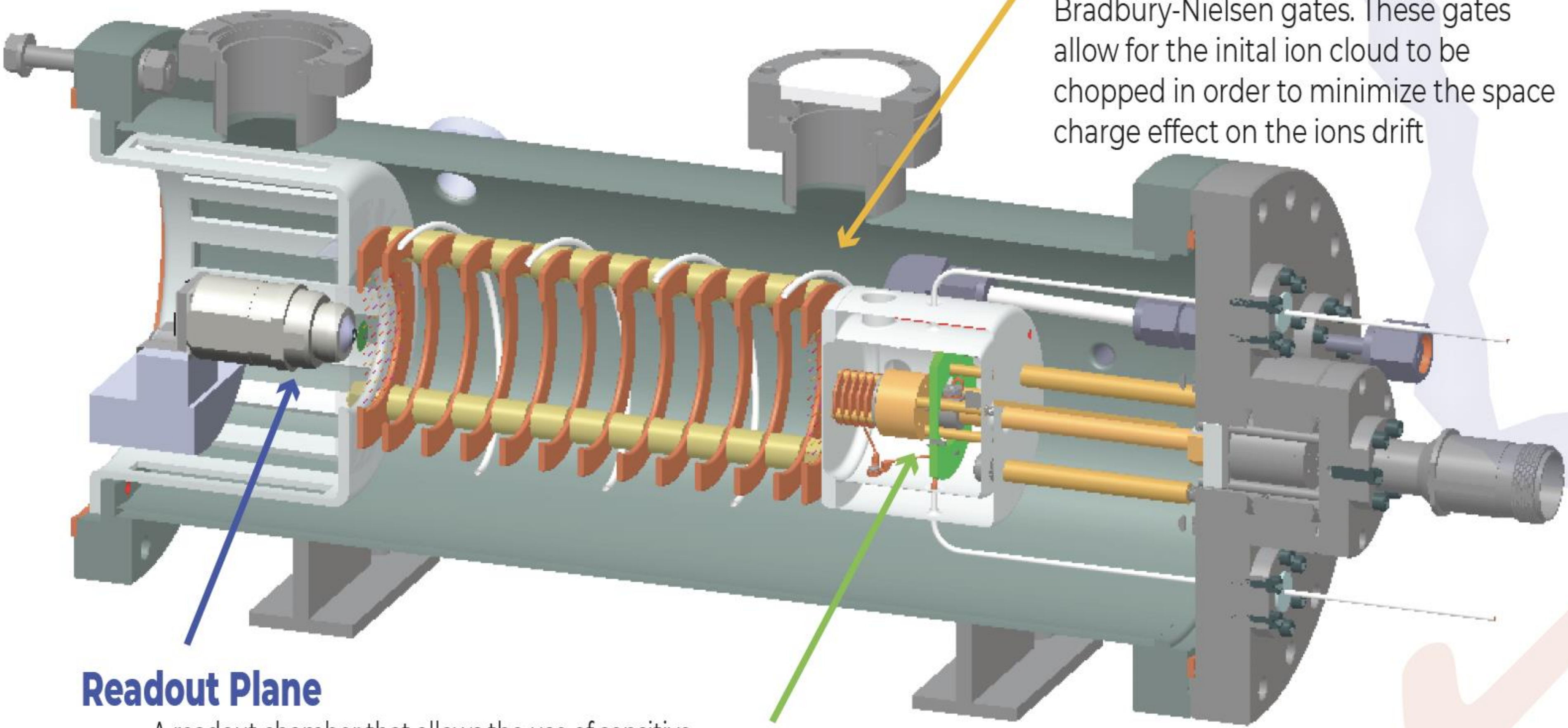
# The ion beam is coming…




Vacuum / pressure valve
Positive ion drift
Signal out
Amplifier board
Source viewports
Detection plane
Ion spark source
Ion source gas supply
Gas ports
Drift region
Ion gate grid 2
Ion gate grid 1
Needle actuator

## Gated Drift Region

20cm uniform drift region that utilized Bradbury-Nielsen gates. These gates allow for the inital ion cloud to be chopped in order to minimize the space charge effect on the ions drift



## Readout Plane

A readout chamber that allows the use of sensitive eletrical amplifier or optical readout. The ions are guided here via an eletric field.

## Ion Source

A needle and ring electrode that is adjustable allowing for an adjustable spark  gap to acomadate Various pulse and pressure conditions.

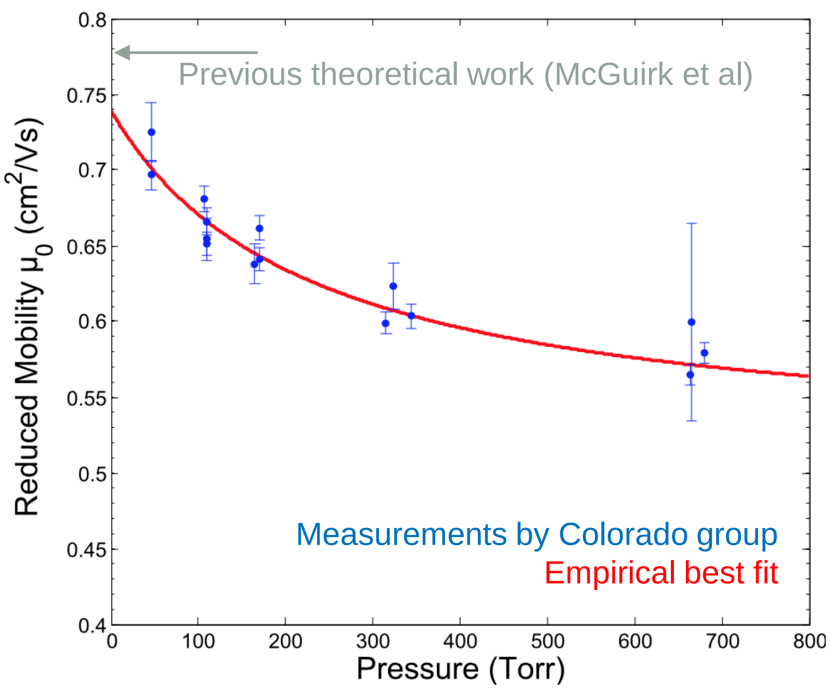
# Barium ion mobility in gas

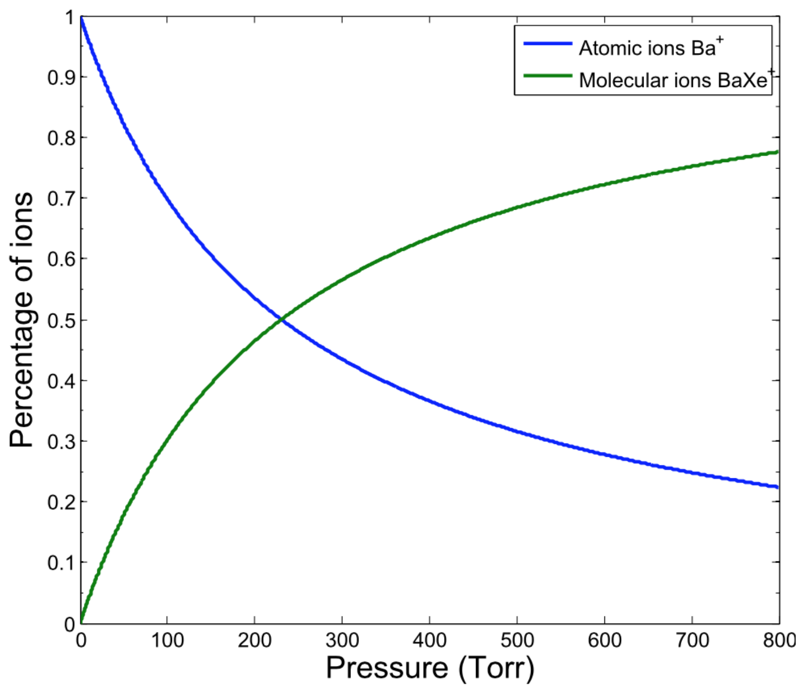
- Measurements from Medina's PhD thesis illustrate the importance of molecular ion formation.

$$\mathrm{Ba}^{+} + \mathrm{Xe} + \mathrm{Xe} \longleftrightarrow \mathrm{BaXe}^{+} + \mathrm{Xe}$$

- Never measured in $Ba^{++}$ to our knowledge
- And not measured above 1 bar to our knowledge either

From **"Mobility and fluorescence of barium ions in xenon gas for the EXO experiment" – Julio Cesar Benitez Madina, PhD thesis, Colorado State University**

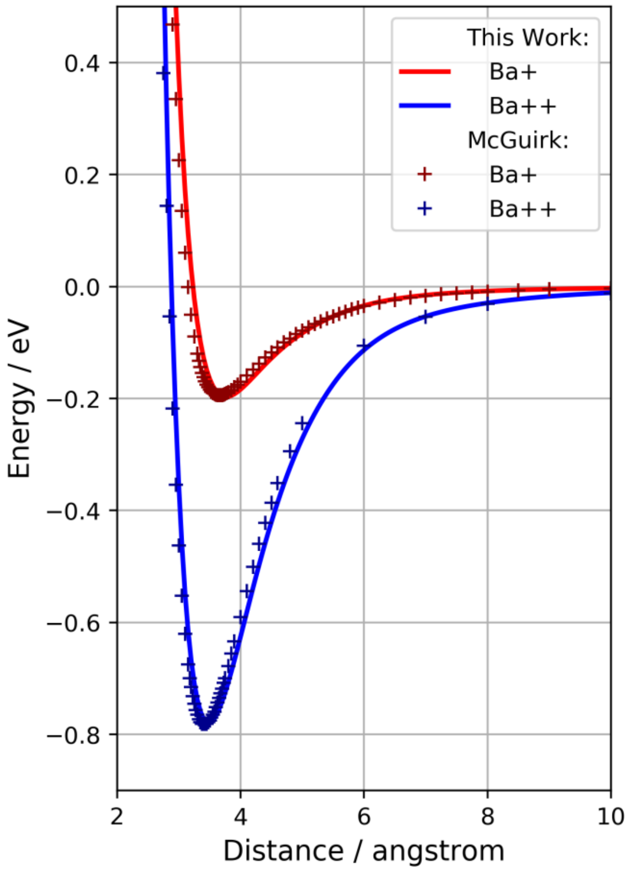


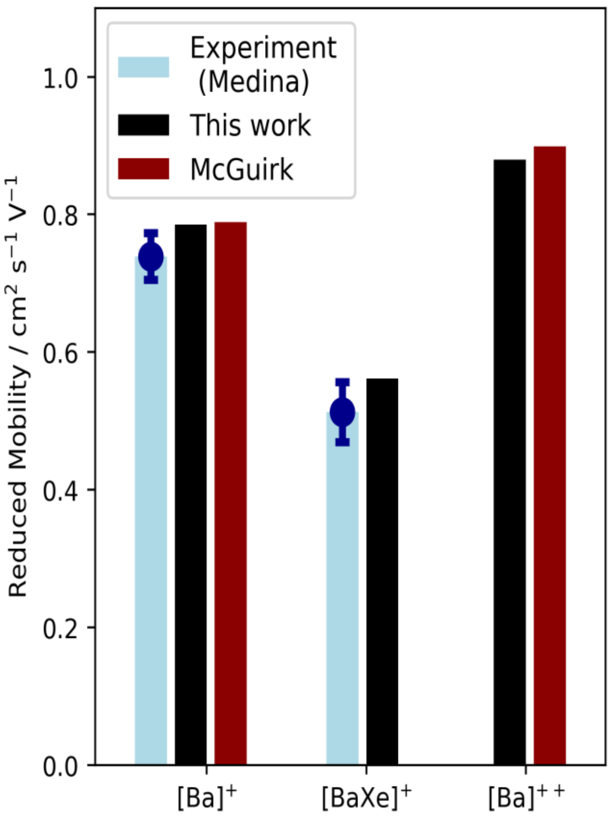
# DFT vs coupled clusters

- Past published calcs used coupled cluster theory – good for bare ion ONLY.
- We calculate cluster distributions and cross sections using density functional theory then evaluate mobility.
- DFT is not expected to be as accurate. But too many atoms for coupled cluster method.
- We benchmark our DFT + ion mobility calculations for bare ions against McGuirk, and experimentally extracted data for BaXe system.

**%-level agreement**

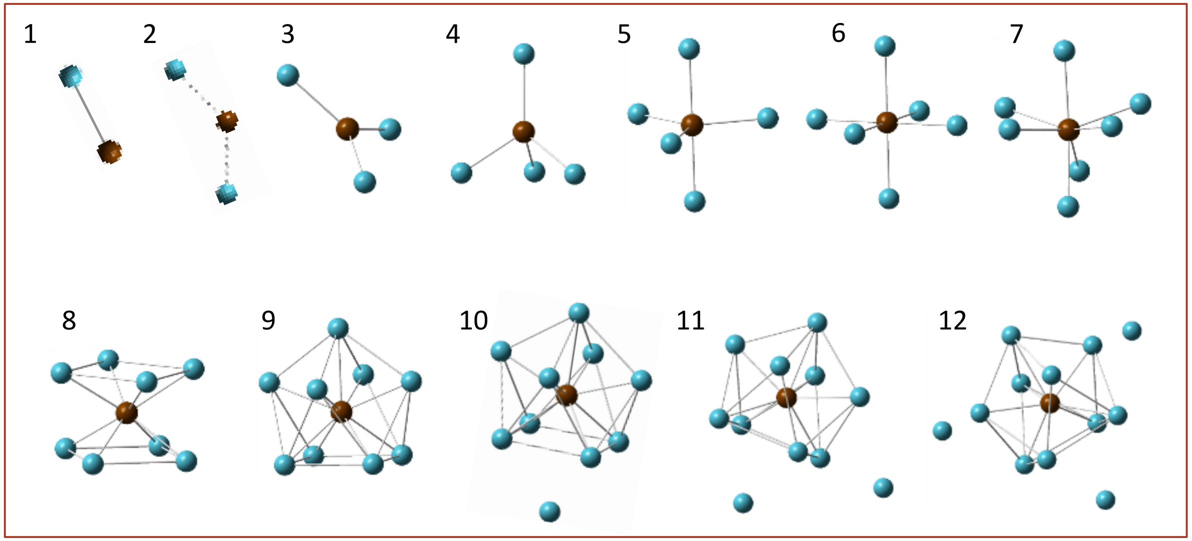



***Comparison of potentials***



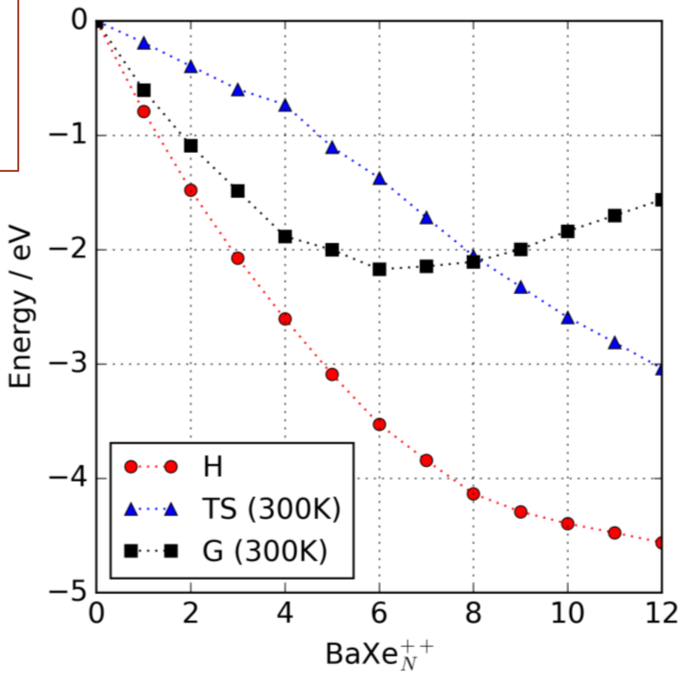

***Comparison of mobilities***

# Ba++ clusters much more




← *Our calculated DFT clusters*

*Our calculated thermo quantities* ↓

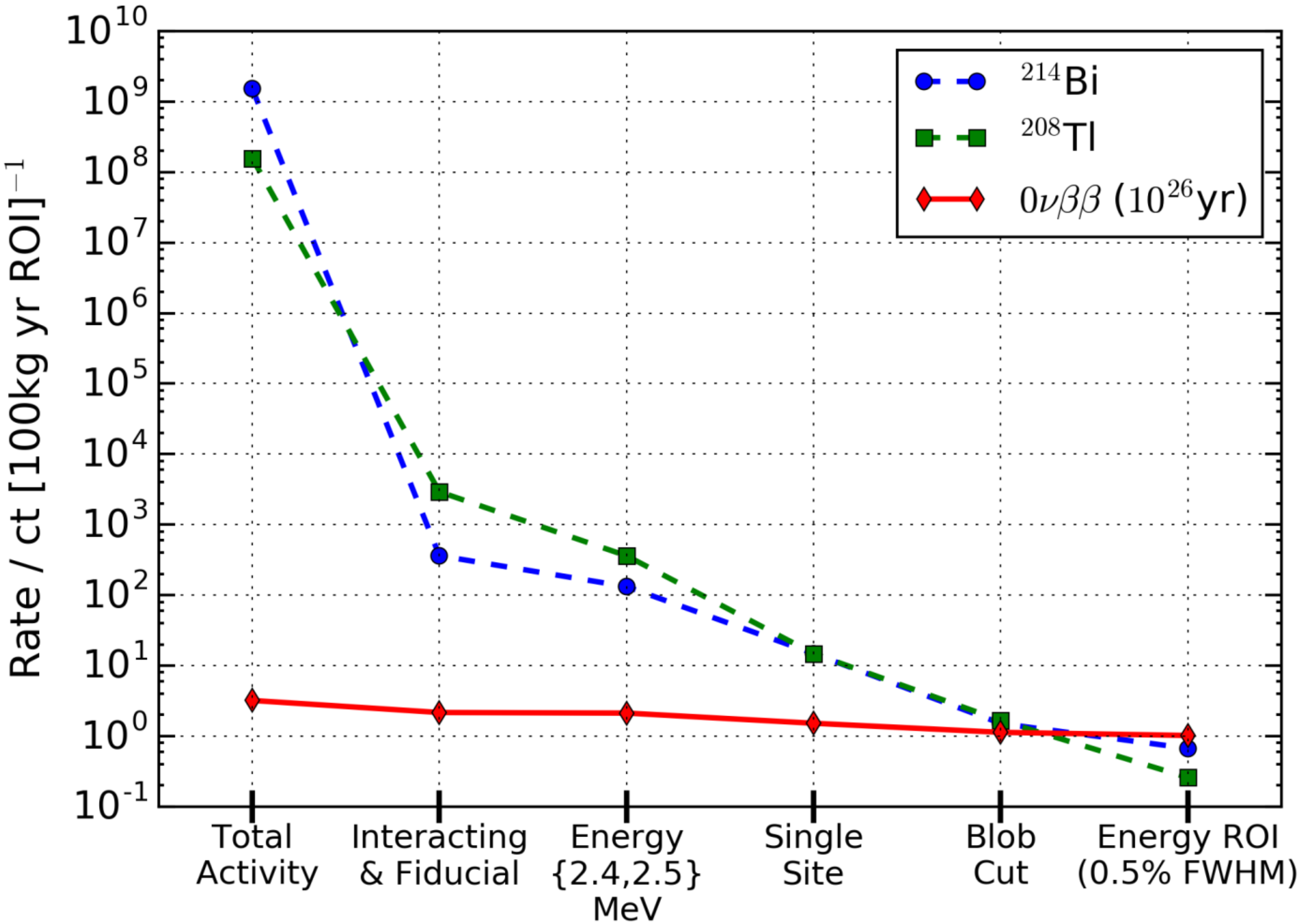



Enthalpy pushes this way →

$$[\mathrm{BaXe}_{N-1}]^{++} + \mathrm{Xe} \leftrightarrow [\mathrm{BaXe}_N]^{++}$$

← Entropy pushes this way

# NEXT-100 projected background index performance




← Approximately Background free @ 100kg-scale

*World-leading in xenon by an order of magnitude, but still ~5-7 cts/yr at ton scale*