



Second workshop on Neutrinoless double beta decay search in Xe
next generation experiment

Barium Tagging

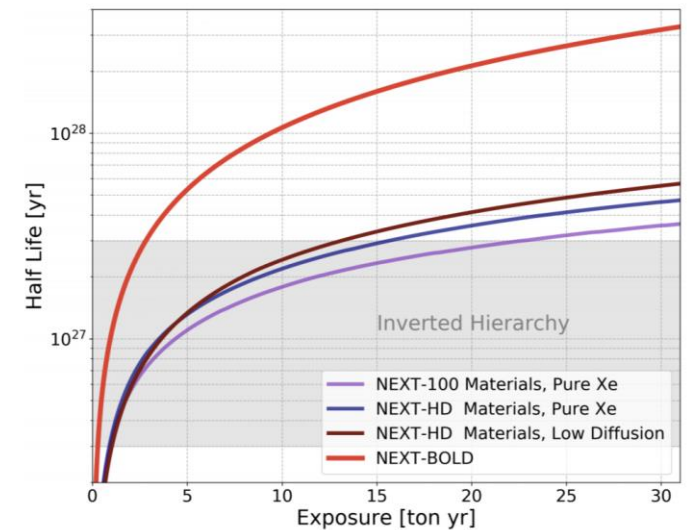
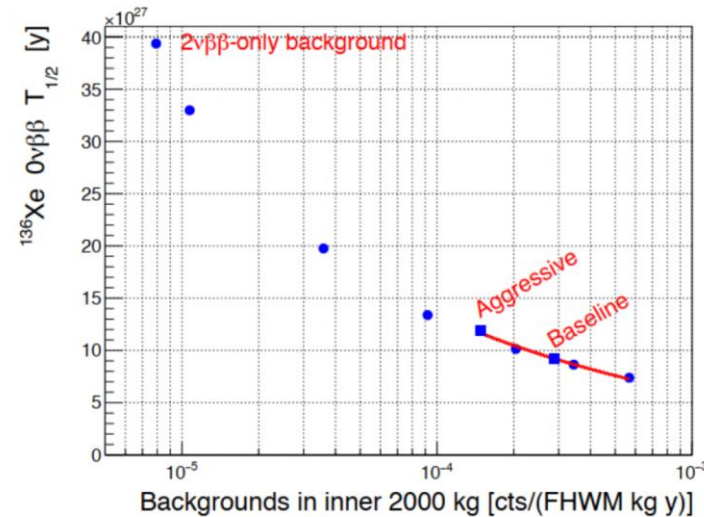
Enakshi Dey

University of Manchester & University of Texas at Arlington

Barium Tagging



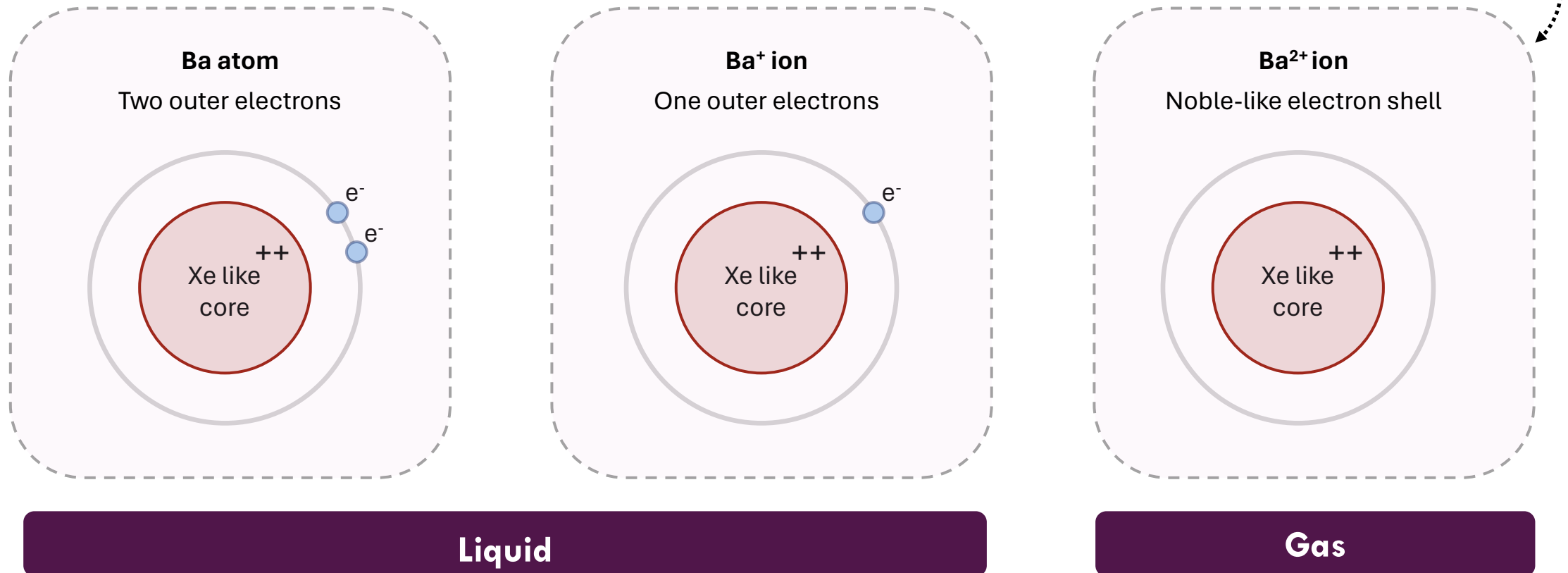
- Ba ion is produced in a true $\beta\beta$ decay event, not in other natural radioactive decays
- Detecting Ba ion in coincidence with the two electrons energy would result in a background-free experiment



Is it possible to detect one Ba ion or atom in a ton of xenon?

Understanding Ba charge state in Xenon

- Inside the detector Ba is born in a high charge state as emerging beta electrons disrupt the atom
- It captures electrons from xenon to reach the Ba^{2+} state
- In **gas** electron-ion recombination to Ba^+/Ba is suppressed, so it essentially stops at Ba^{2+}



Moe's Original Proposal

New approach to the detection of neutrinoless double-beta decay

M. K. Moe

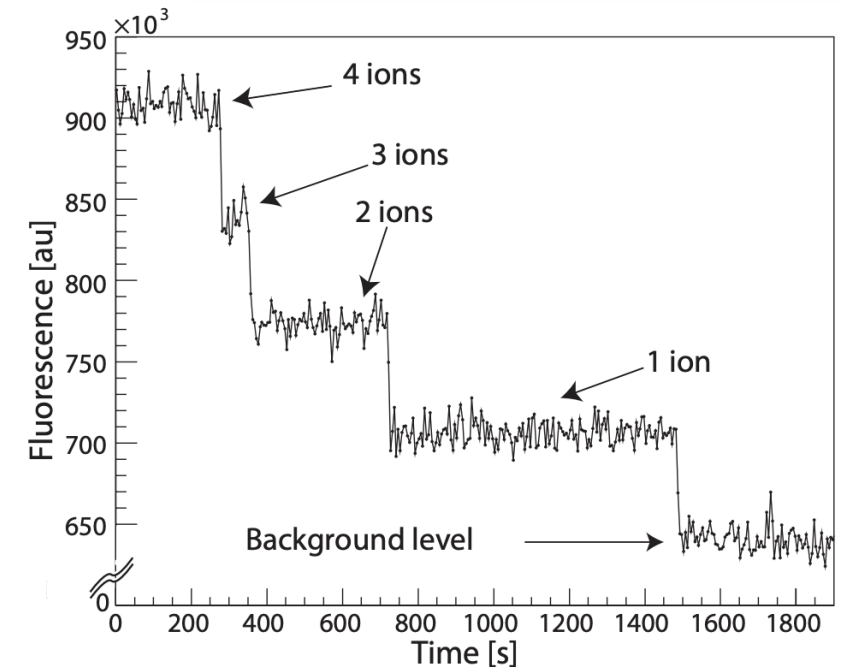
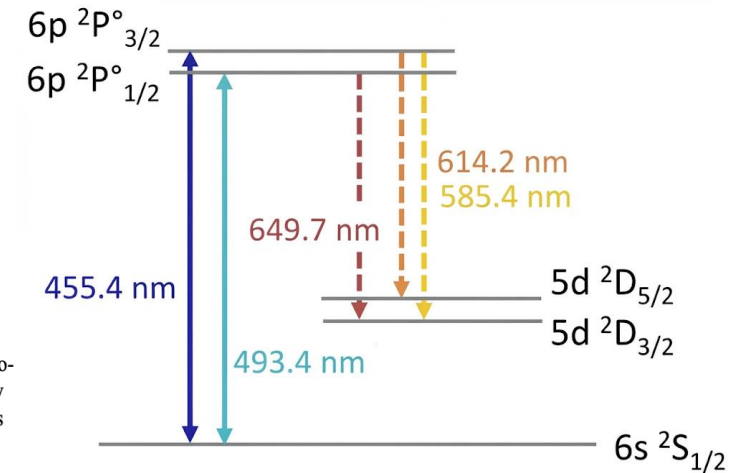
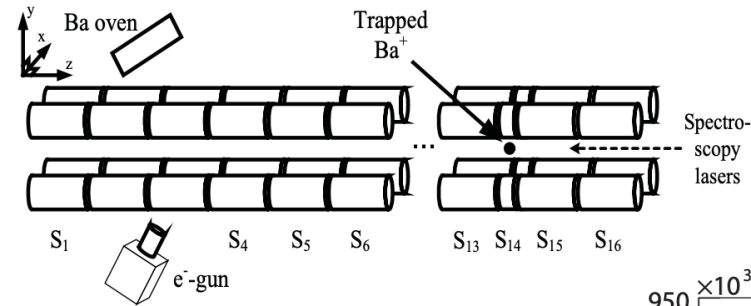
Department of Physics, University of California, Irvine, California 92717

(Received 2 April 1991)

A distinctive feature of the double-beta-decay signature that has been neglected in direct counting experiments is the appearance of the daughter atom. The newly created nucleus, usually being stable, is not easily detected. The atomic physics of the daughter, however, may be considerably more accommodating, especially in the case of ionized ^{136}Ba arising from the double-beta decay of ^{136}Xe . The barium ion isolated in the xenon matrix may be detectable by its laser fluorescence. Coincident detection of the ion and the beta particles could well render background nonexistent.

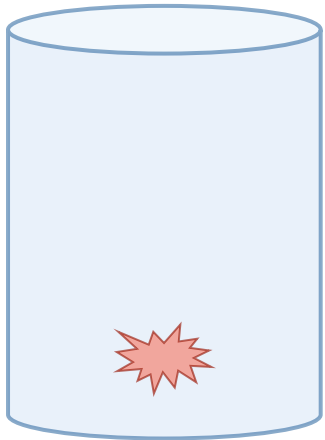
- First proposed by Moe in 1991 the possibility of detecting ^{136}Ba in a xenon detector using laser spectroscopy
- Since then identification of Ba has been demonstrated in **Ba atom, Ba^+ , and Ba^{2+}**

Moe's idea was demonstrated by EXO group in 2007 by trapping 1,2,3,4 ions in a linear paul trap and detecting them by laser fluorescence



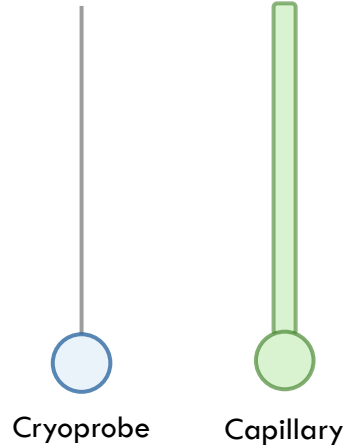
nima.2007.05.123
B. Flatt et al.

Barium Tagging in Liquid Xenon



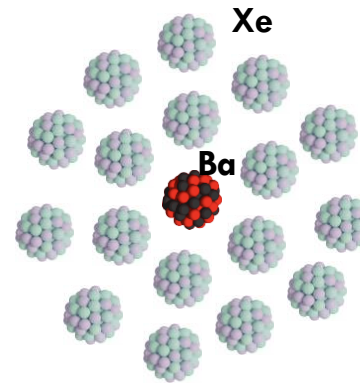
Localize

Find the decay site



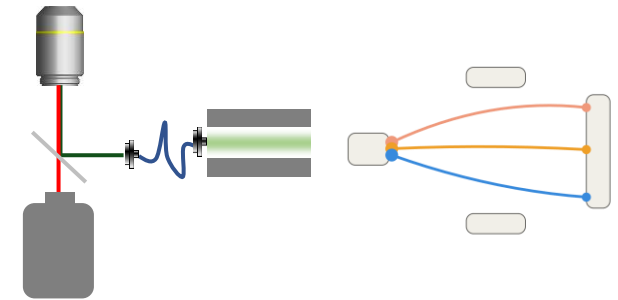
Extract

Two parallel routes



Separate

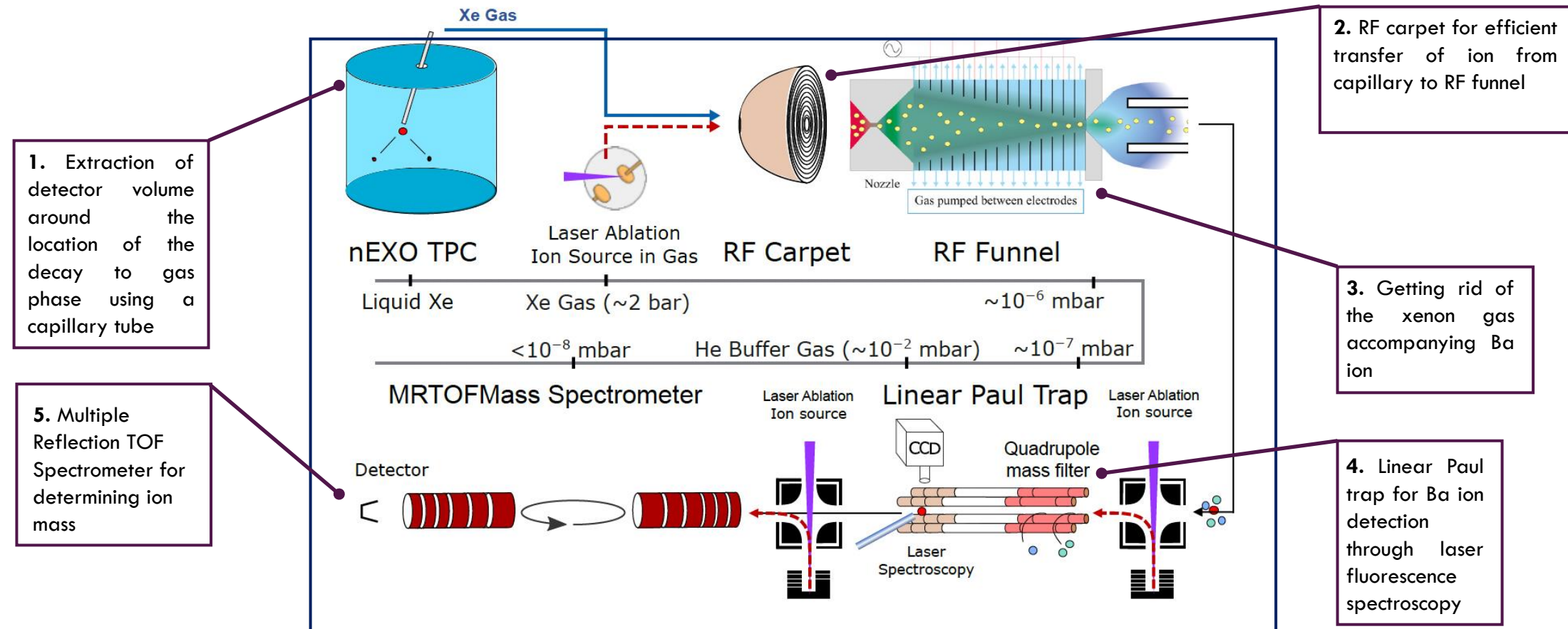
Remove the background
Xe



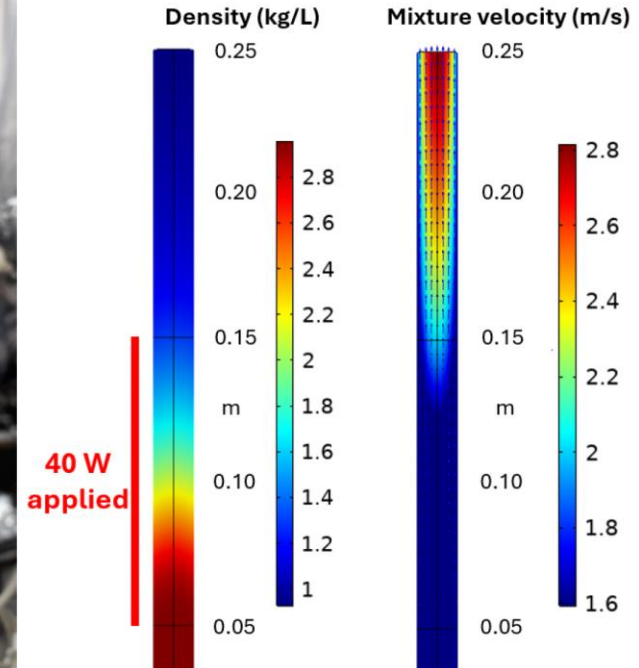
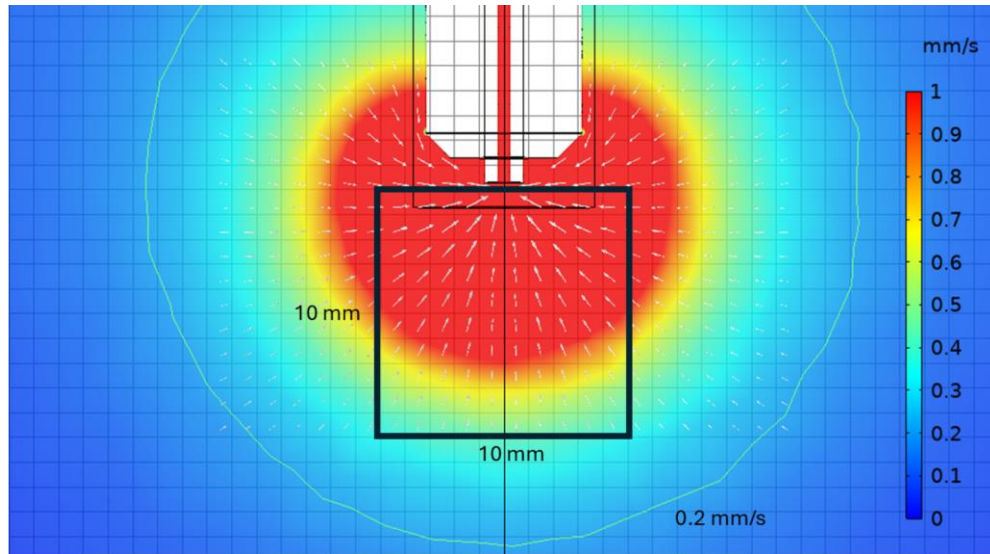
Identify

Confirm it's Ba

Extracting Single Ba Ion from Liquid Xenon Using Capillary



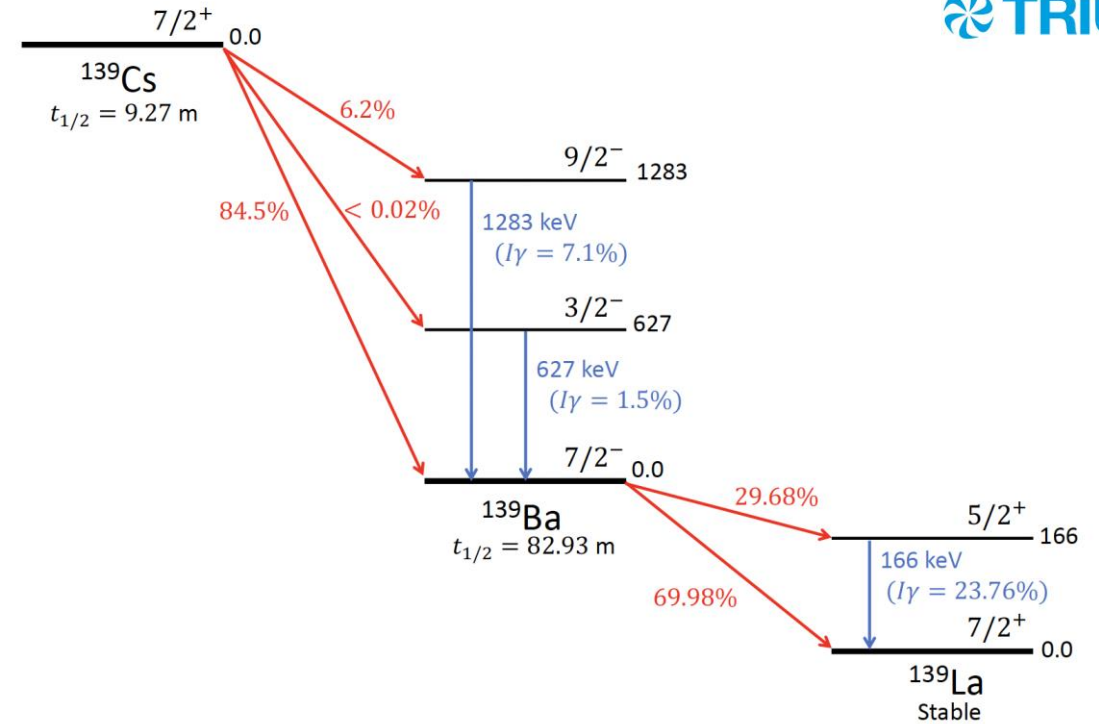
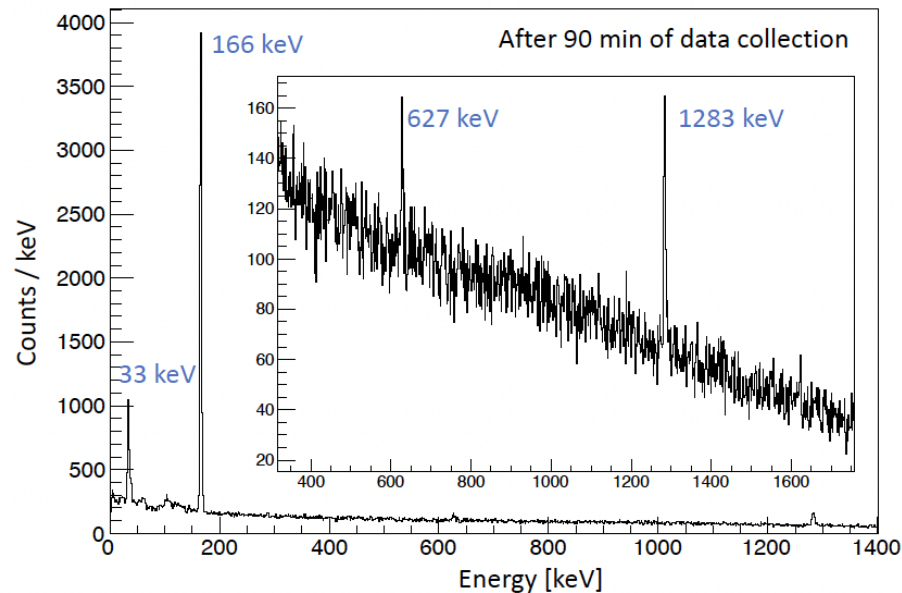
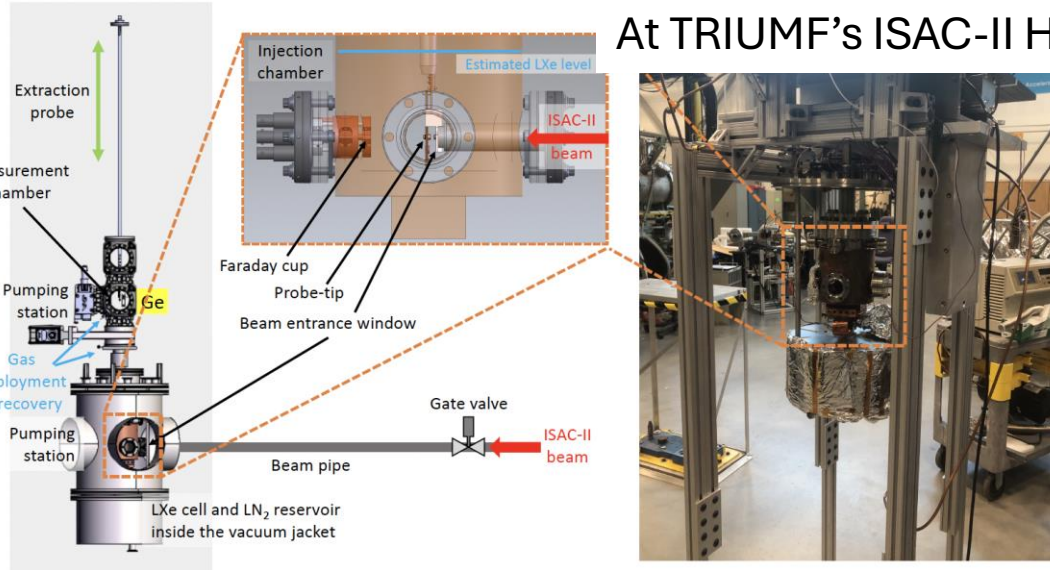
Ion Capture and Transport



- The capillary tip pulls in any ion that comes near it — its "capture zone"
- The drift field distorts near its metal surface, pushing ions to the wall, so it's switched off while collecting
- Xenon boils to gas mid capillary — flow stays laminar, so the ion isn't lost to the wall

Ion Source for Testing Ba Ion Extraction

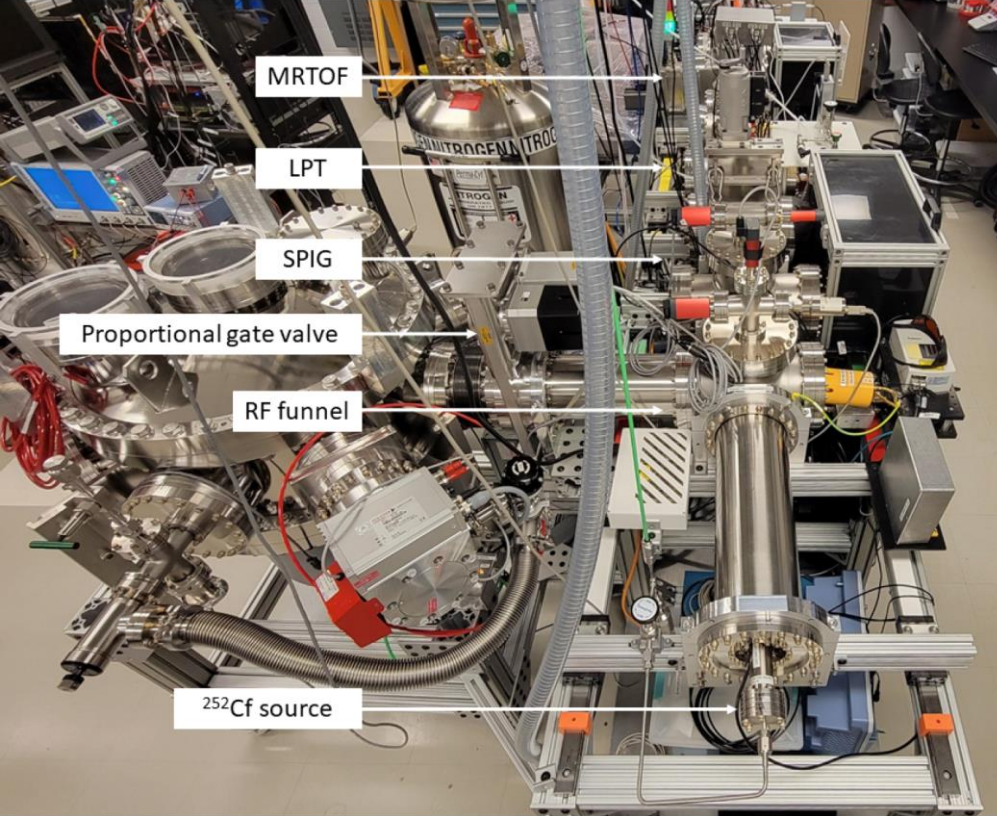
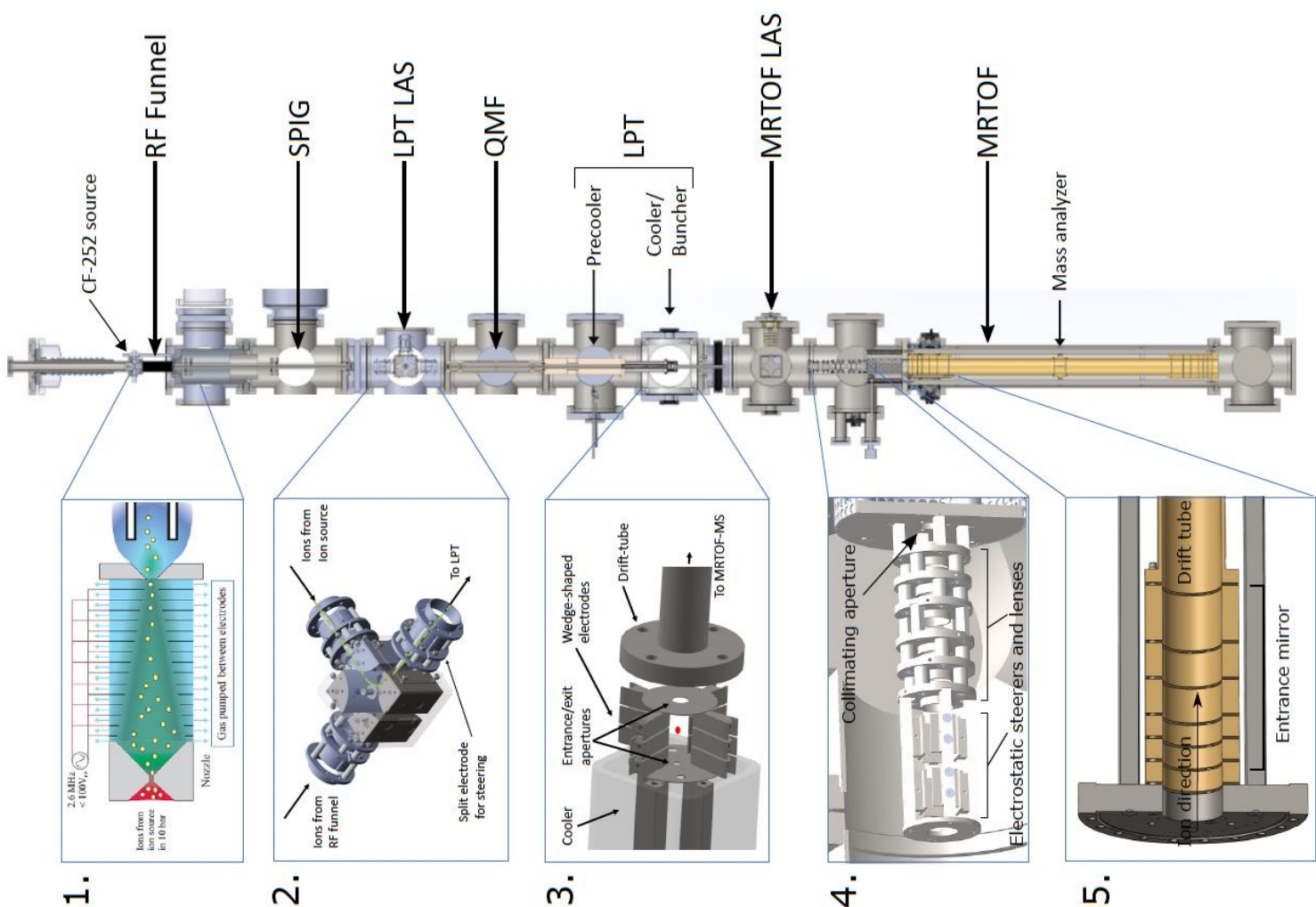
At TRIUMF's ISAC-II Hall



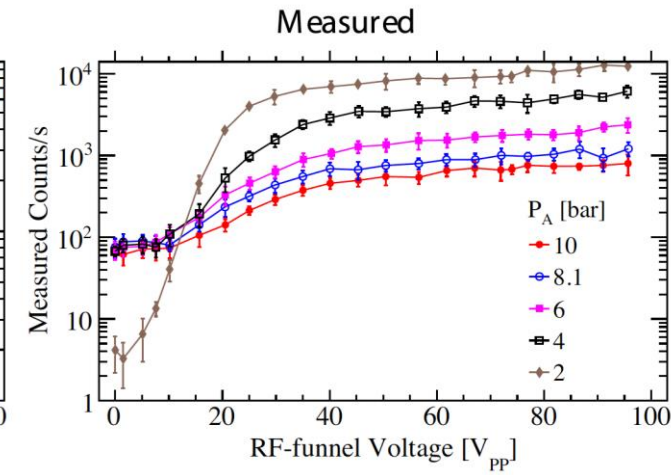
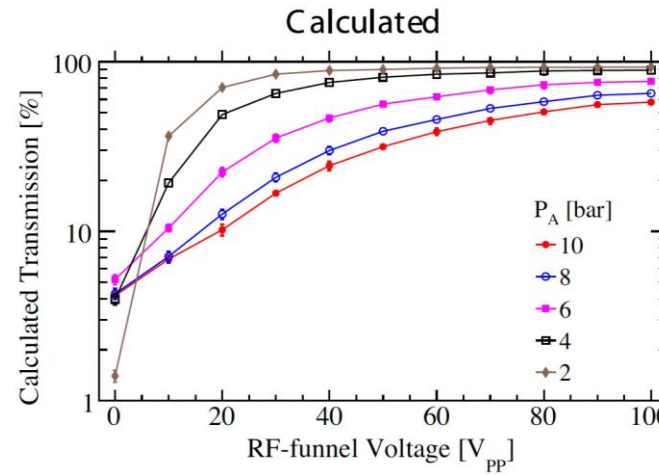
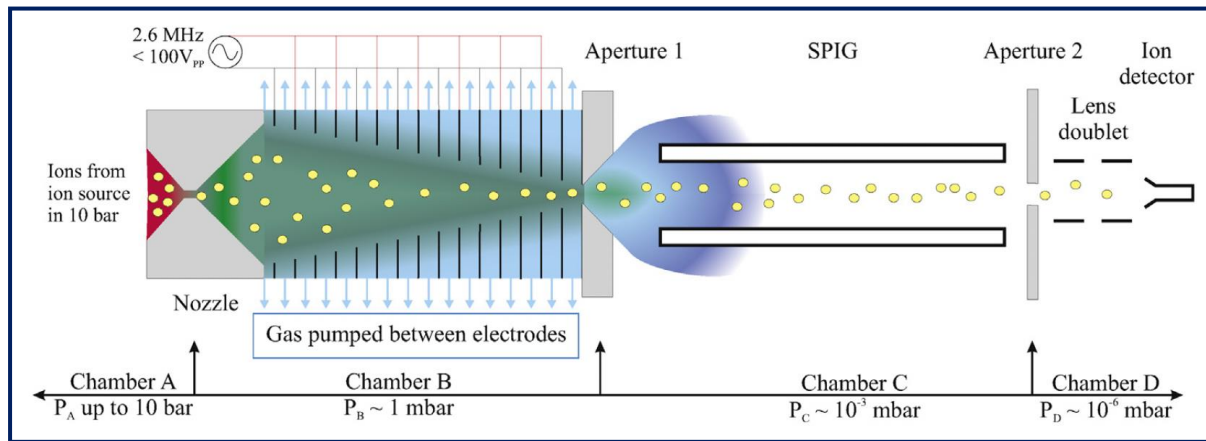
- Need a controlled, known ion source to validate extraction
- ^{139}Cs beam stopped in LXe, daughter ^{139}Ba collected electrostatically, identified via HPGe γ -spectroscopy (166 keV line)

G C Ball et al 2016 Phys. Scr. **91** 093002

Ion Transport and Identification Steps



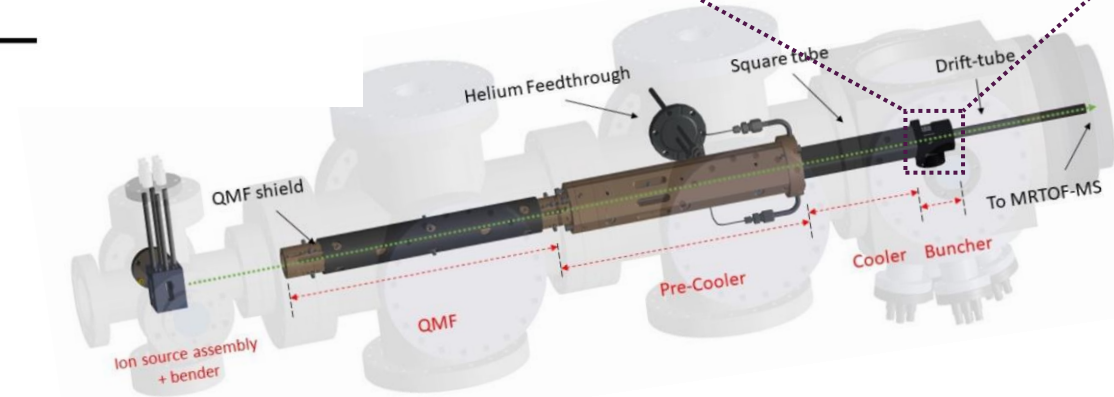
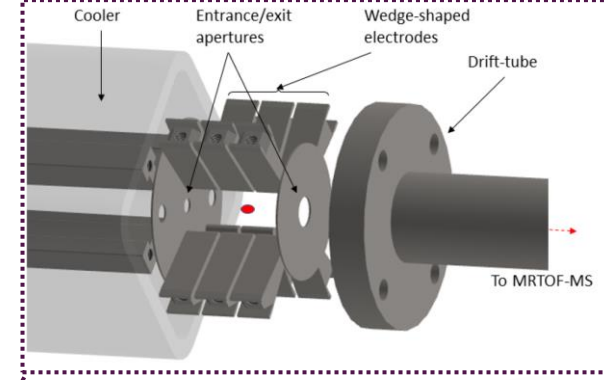
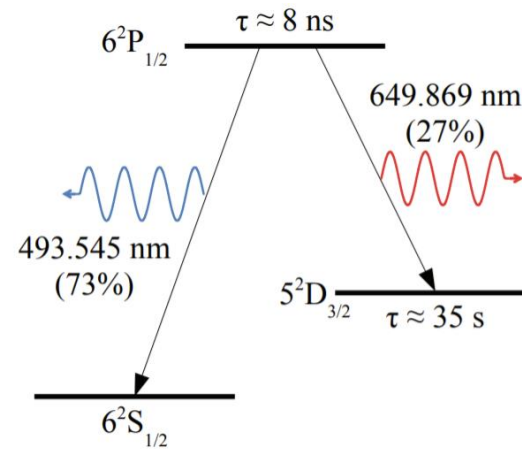
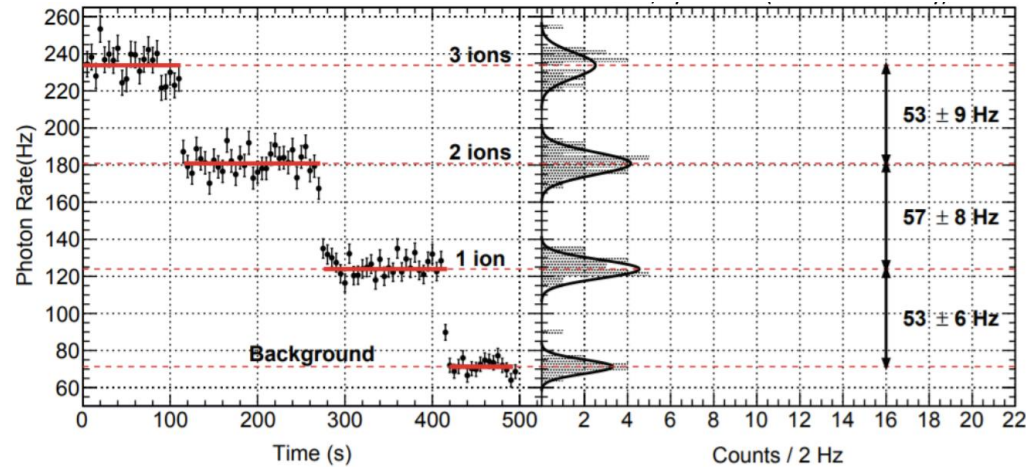
Separating Ba⁺ from Xe Gas Using RF Funnel



- Downstream instruments need vacuum, but the ion arrives at 10 bar normal pressure-reduction methods lose most of the ions
- RF funnel lets gas escape radially through gaps between electrodes, dropping pressure from 10 bar to 1 mbar
- Ion stays confined to the axis by the RF field and can't escape sideways with the gas, gas flow alone carries the ion forward
- Ion extraction up to 10 bar

International Journal of Mass Spectrometry 379 (2015) 110–120
T. Brunner et al.

Trapping and Identifying Ba Ion : LPT

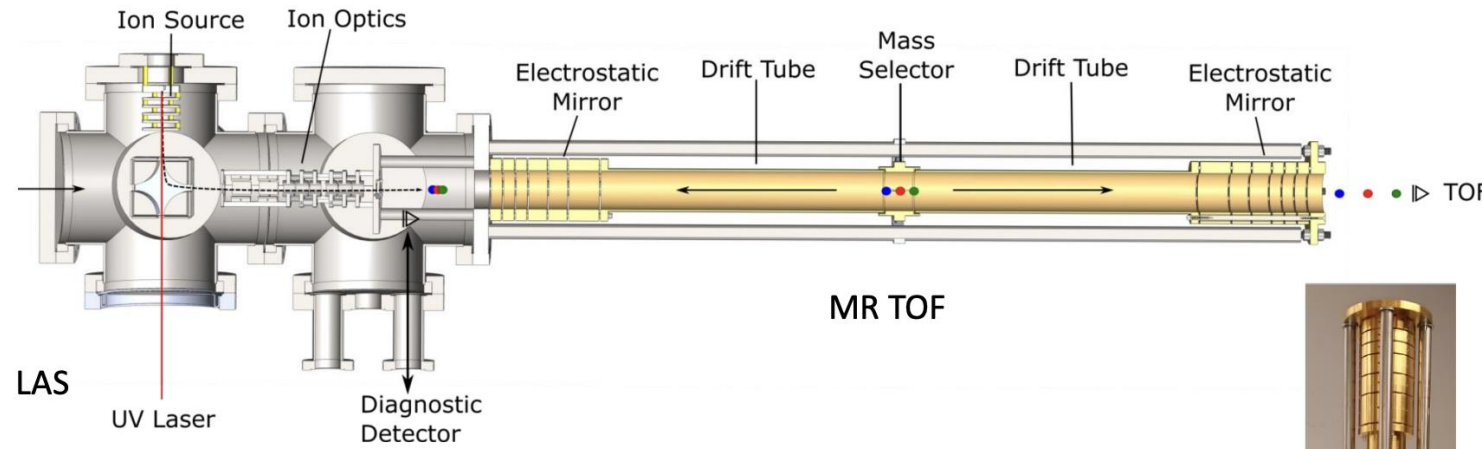
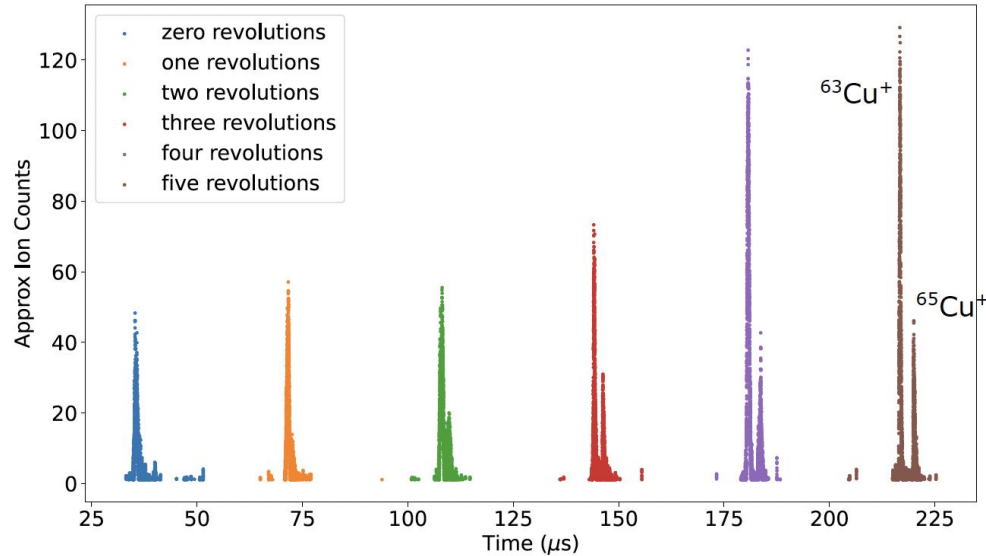


Single Ba ion detection has been demonstrated at Carleton University

- After the RF funnel, the ion arrives too energetic to trap, it first needs a Quadrupole Mass Filter (QMF) to reject unwanted masses, then two cooling stages to bring it down to sub-eV energy using helium buffer gas
- Cooled ions are collisionally trapped in a buncher (Linear paul trap) confining ions radially with RF and axially with a DC potential well
- Trapped Ba⁺ ion is identified by laser induced fluorescence

Lan, Y. A Linear Paul Trap for Barium Tagging of Neutrinoless Double Beta Decay in nEXO. Ph.D. Thesis,

Confirming Ba^+ Mass : MRTOF



Is it really $^{136}\text{Ba}^+$, or is $^{136}\text{Xe}^+$ riding along?

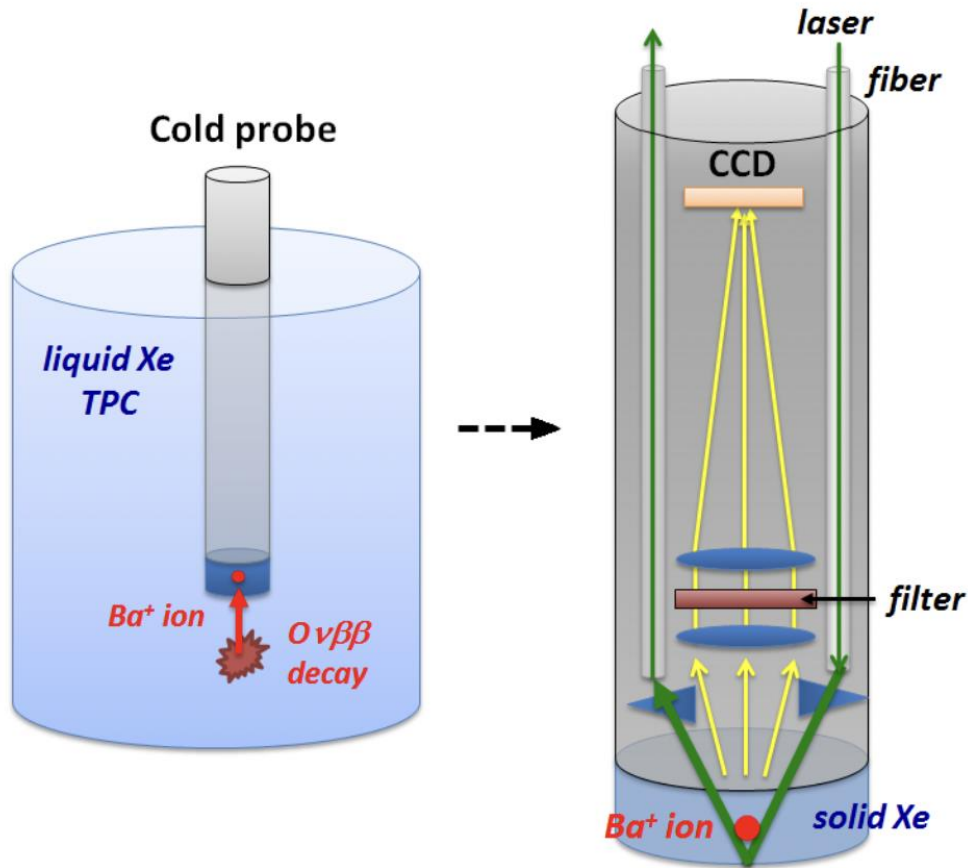
- Ions with different mass to charge can be resolved
- MRTOF - bouncing the ion between two mirrors - hundreds of reflections in ~ 1 m
- Simulated with real LPT ions: resolving power $> 100,000$ — enough to separate Ba from Xe
- Tested with copper ions : 400 reflections $\rightarrow ^{63}\text{Cu}^+ / ^{65}\text{Cu}^+$ cleanly separated, resolving power = 20,000

Fully assembled
MRTOF



Murray, K. Design and Commissioning of a Multi-Reflection Time-of-Flight Mass-Spectrometer for Barium Tagging with nEXO. Ph.D. Thesis

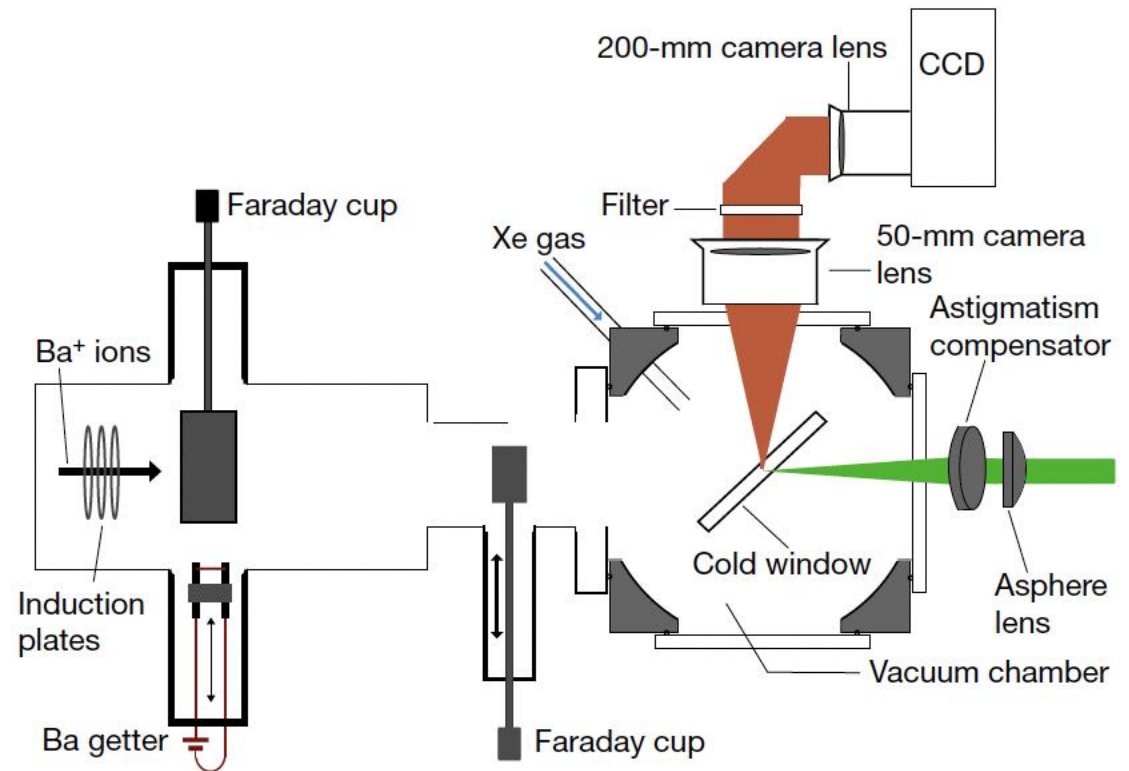
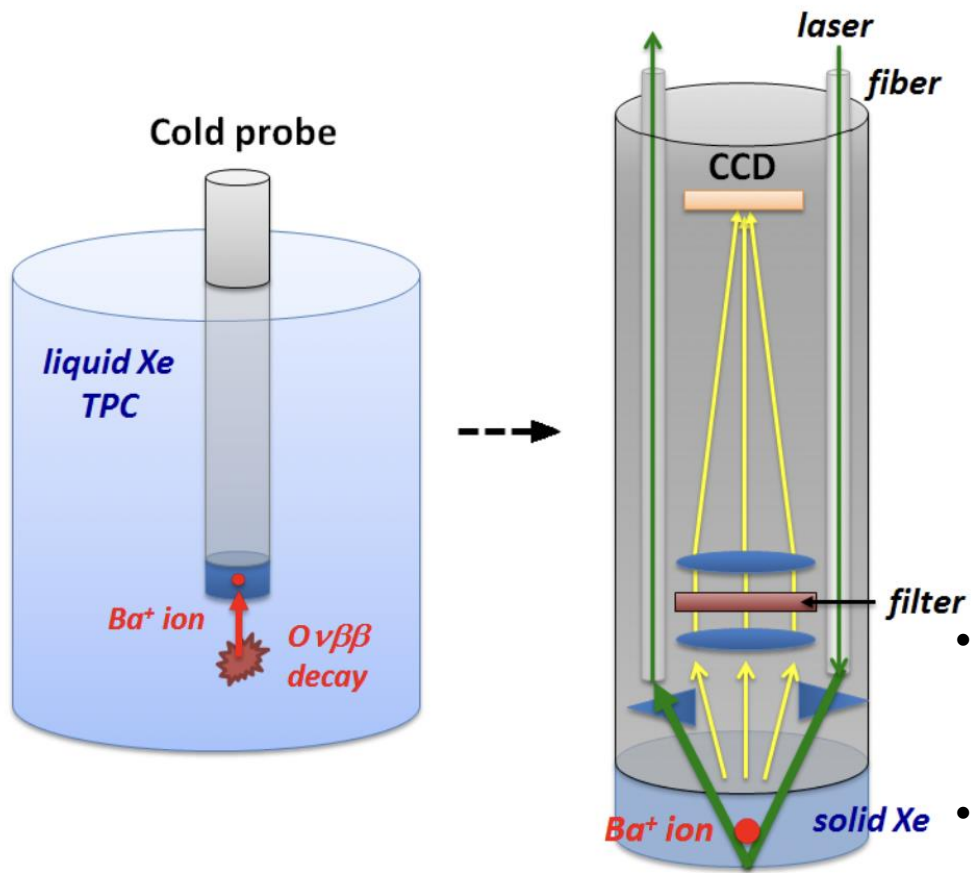
Ba Tagging in Solid Xenon



Concept :

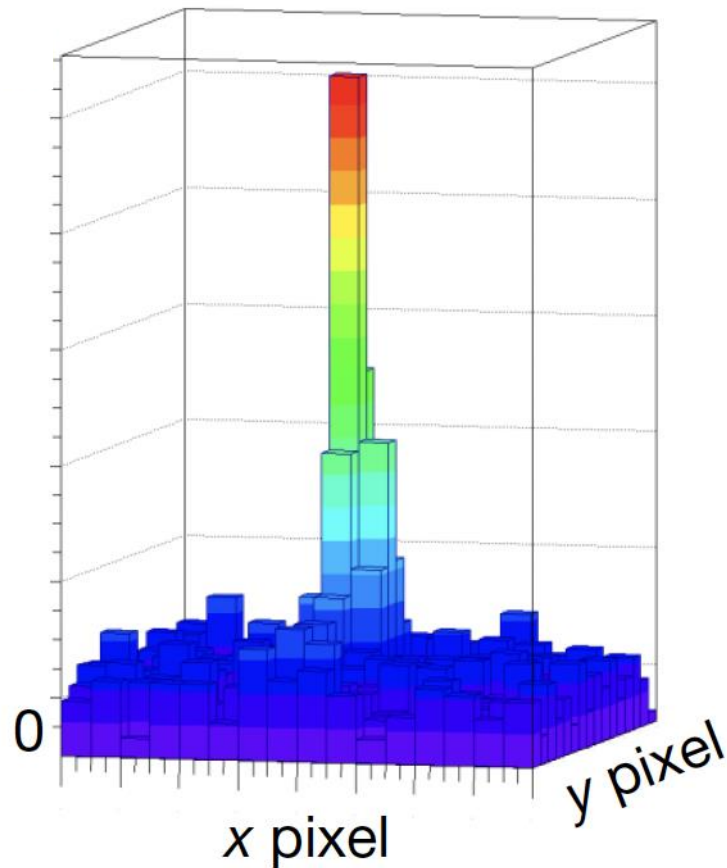
- Cold probe inserted into LXe TPC at candidate $0\nu\beta\beta$ decay site, freezes thin SXe layer, capturing Ba^+/Ba in the ice
- Trapped atom excited via fiber-coupled laser; fluorescence collected and imaged on CCD

Ba Tagging in Solid Xenon



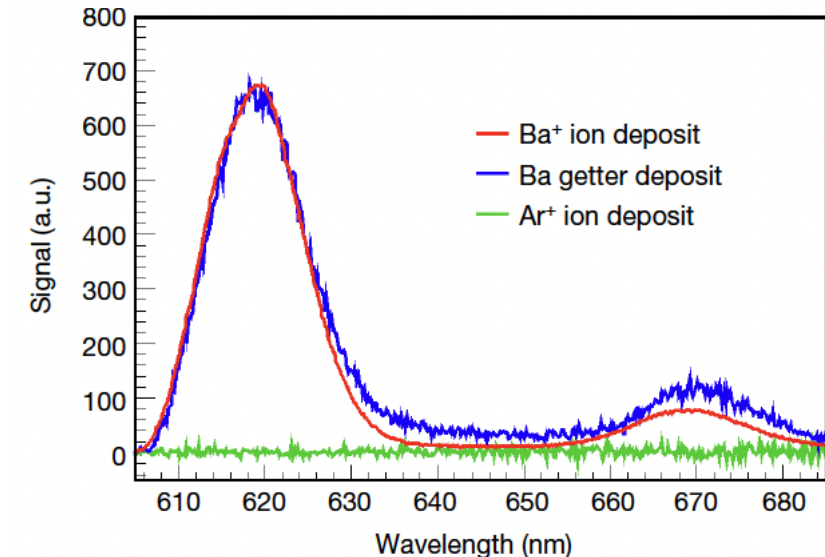
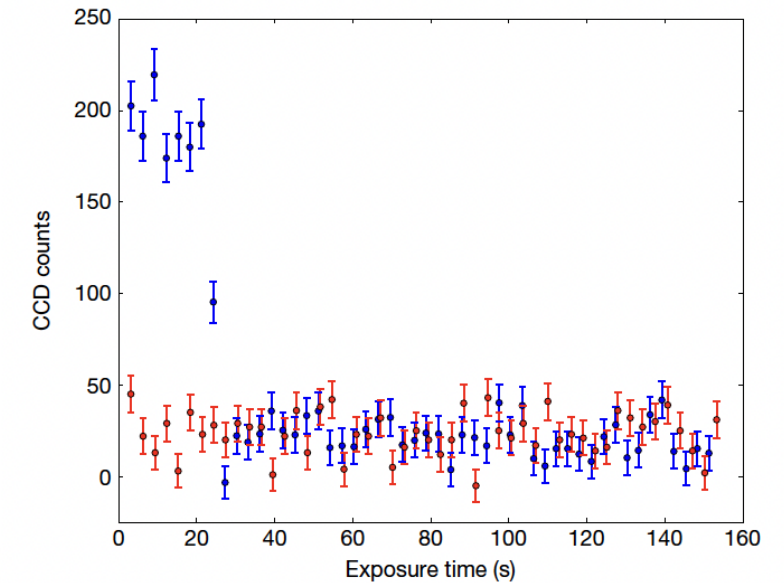
- Two Ba sources — thermal getter and pulsed Ba⁺ ion beam — deposit onto cold sapphire window
- Faraday cups + induction plates calibrate ion number and beam alignment
- Laser excites the deposit; fluorescence is filtered and imaged onto a EMCCD camera

Single Barium Atom/Ion Detection in Solid Xenon Matrix



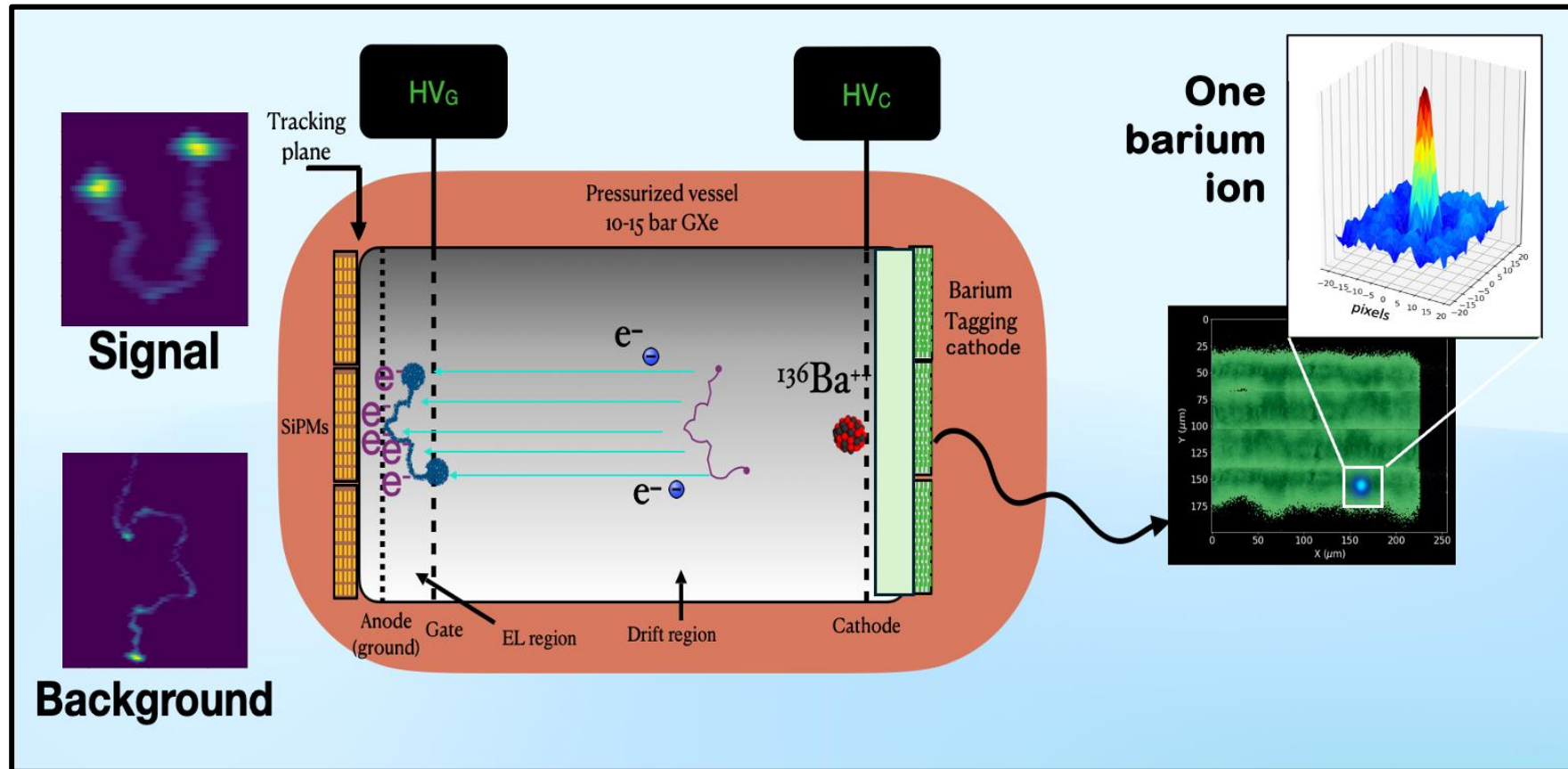
Single Ba atom peak

- Signal (blue) drops sharply to background (red) after ~ 30 s
- The sharp cutoff is the single-atom signature
- Absence of signal with an Ar^+ ion beam rules out ion-damage color centers



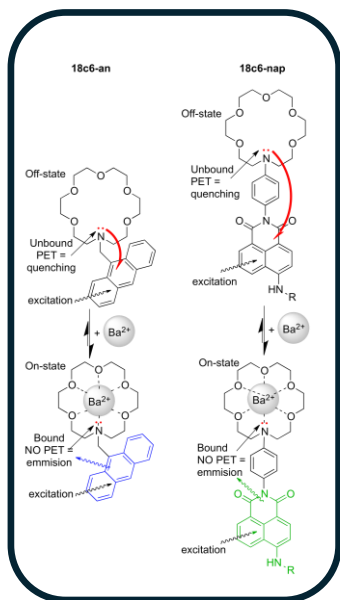
Nature volume 569, pages203–207 (2019)

Detecting Ba^{2+} Ion in Xenon Gas

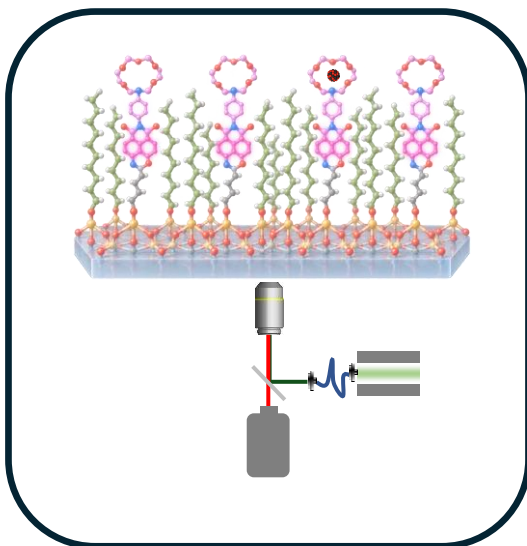


- Ba^{2+} doesn't have any outer shell electrons
- It's not easy to do atomic fluorescence

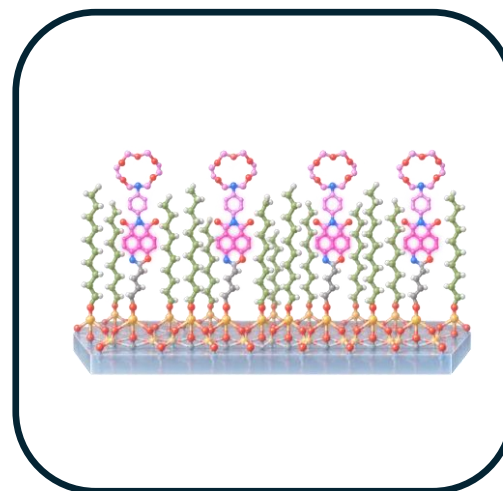
Steps to Detect Ba^{2+} Ion



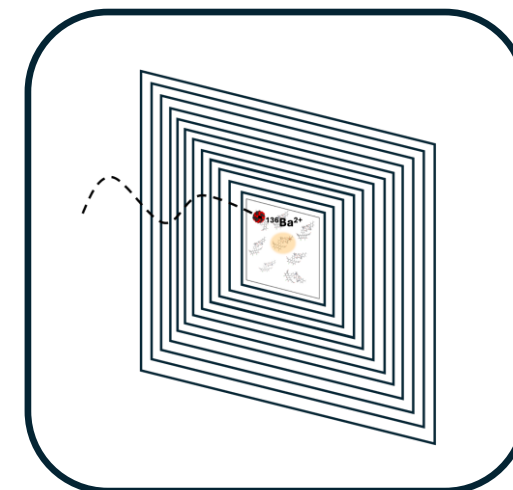
1. Designing and synthesizing the desired molecule



2. Unambiguous optical detection of the molecule attached to the barium ion



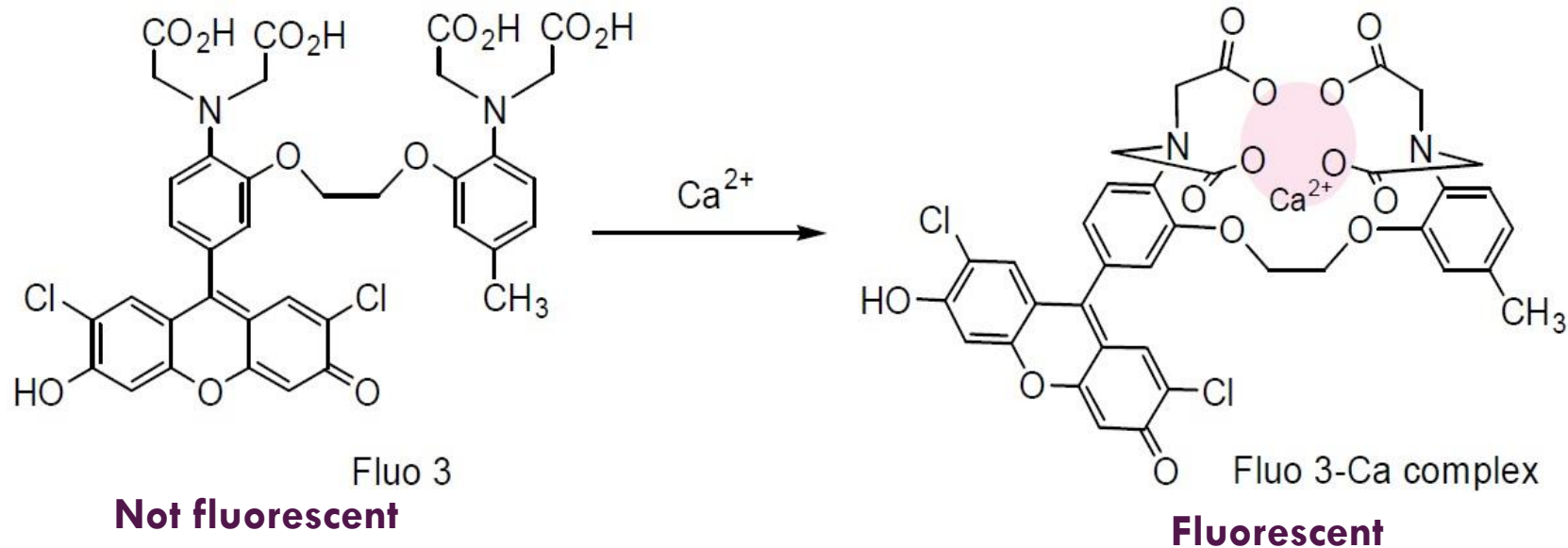
3. Growing a Self assembled monolayer



4. Transporting the ion to the sensor

Single Molecule Fluorescence Imaging

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



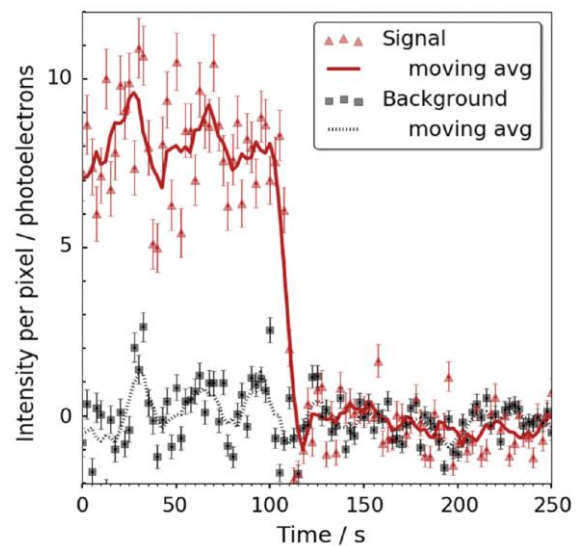
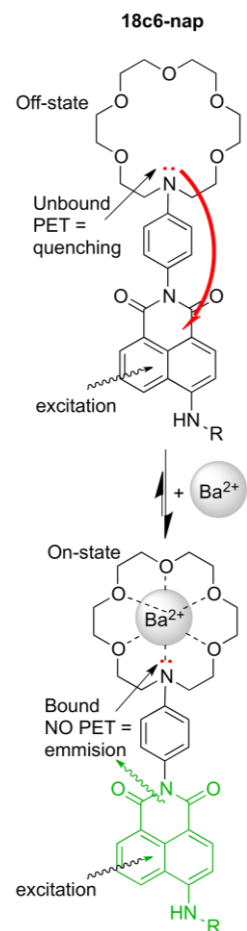
Since calcium and barium belong to the same chemical family, dyes designed to detect calcium often respond to barium as well

But fluorescein dye doesn't shine without solvent, deprotonation of the carboxylic acids to accept the ion depends on it

*Credit : David Nygren
J.Phys.Conf.Ser. 650 (2015) no.1, 012002*

Exploring Molecules

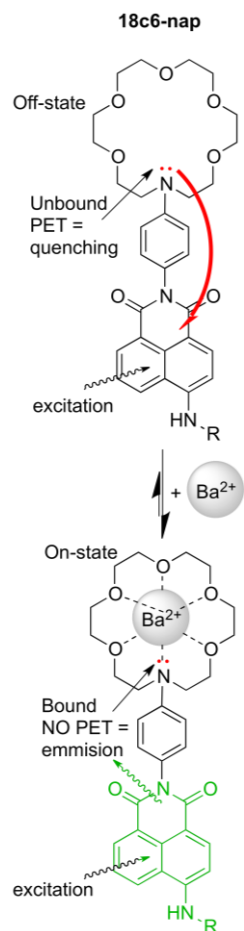
Turn on
Fluorescence
Indicator



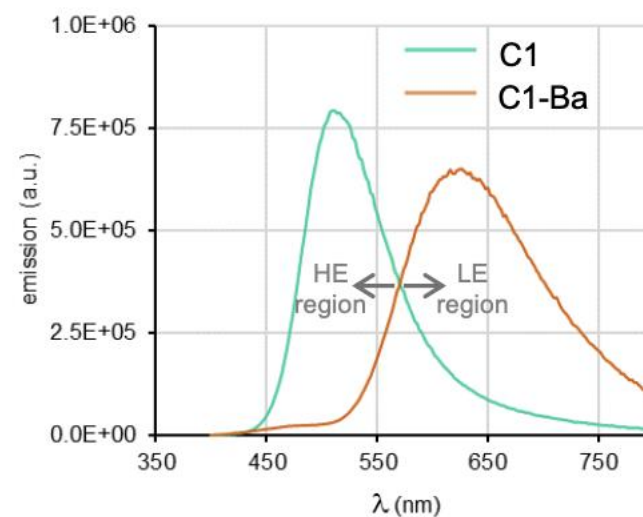
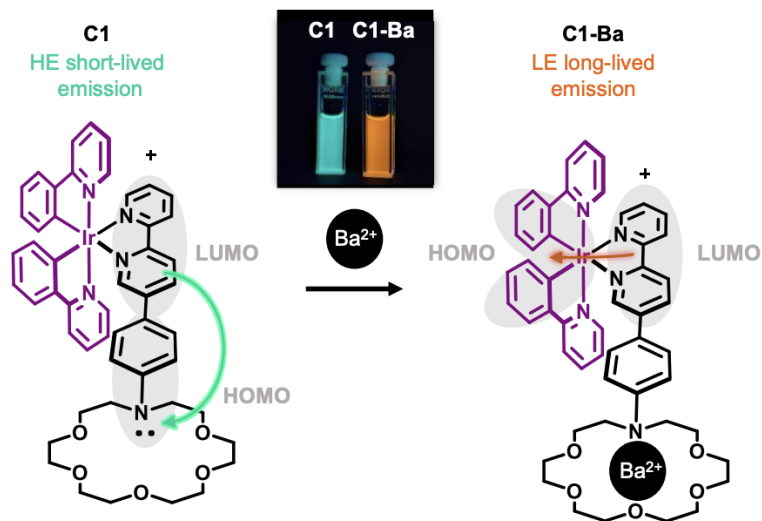
ACS Sensors 6, 1, 192-20 (2021)

Exploring Molecules

Turn on
Fluorescence
Indicator



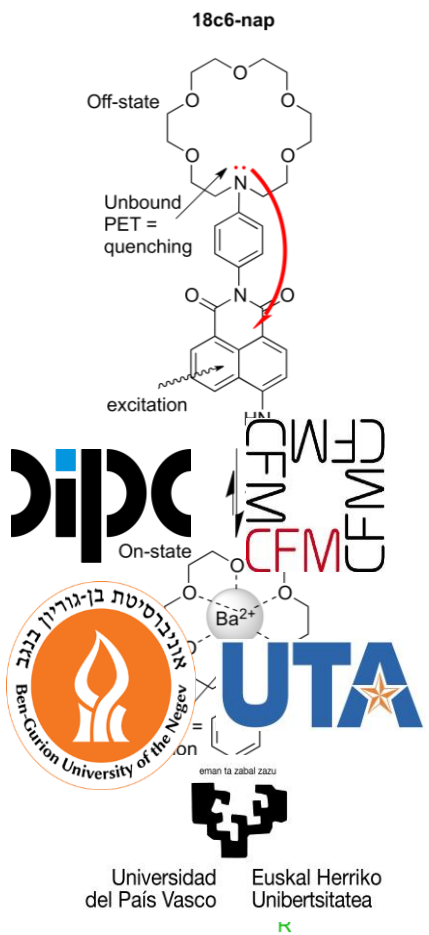
Time Resolved
Fluorescence Bi-
color Indicator



chemrxiv-2024-t81cl

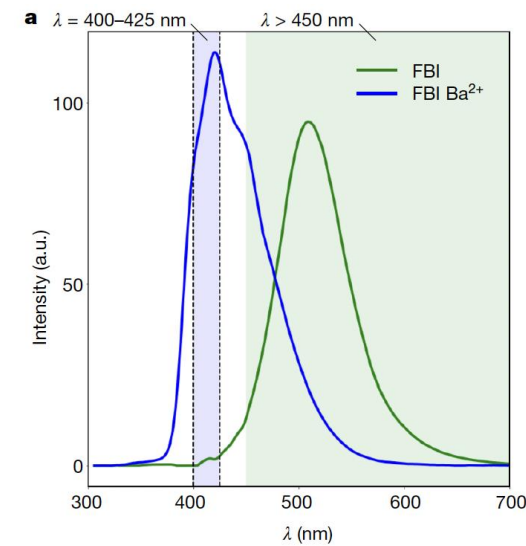
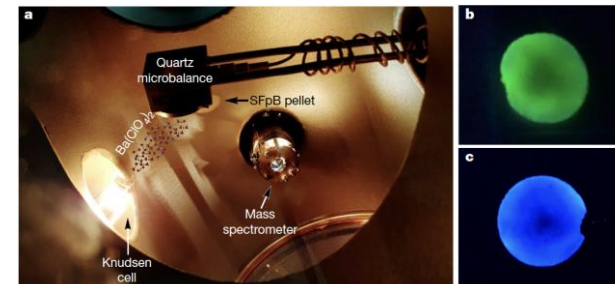
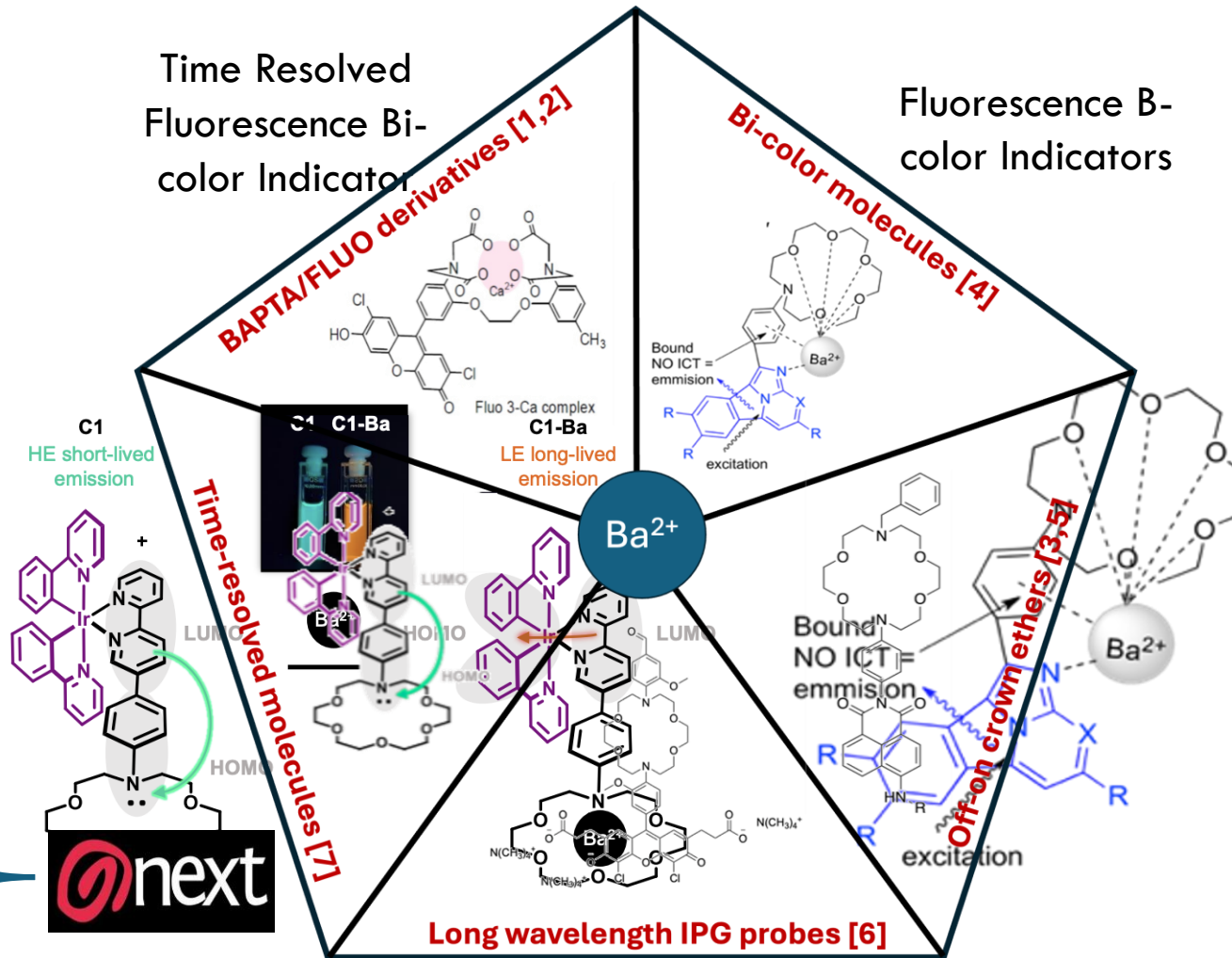
Exploring Molecules

Turn on
Fluorescence
Indicator



Time Resolved Fluorescence Bi- color Indicator

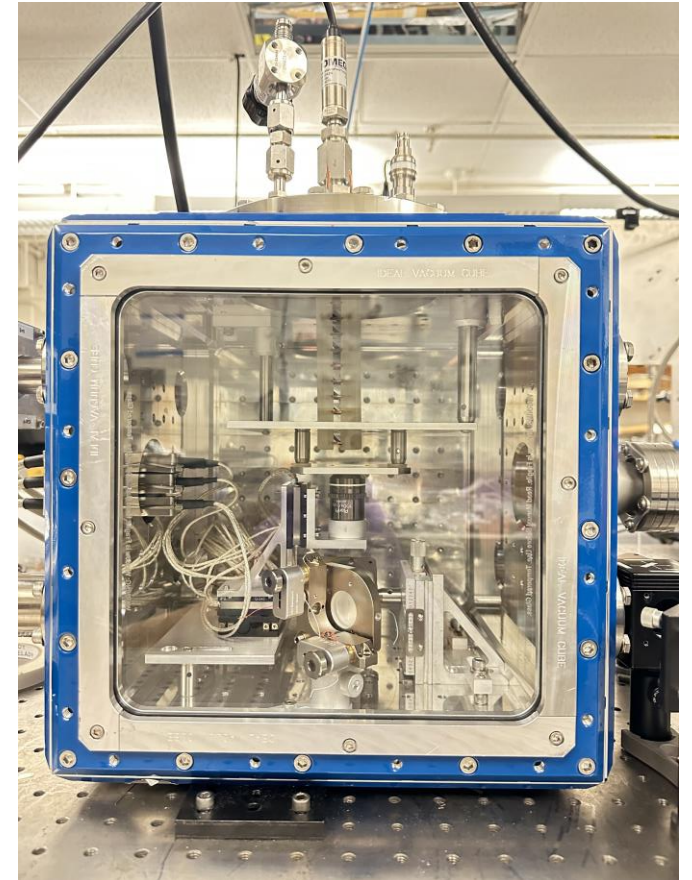
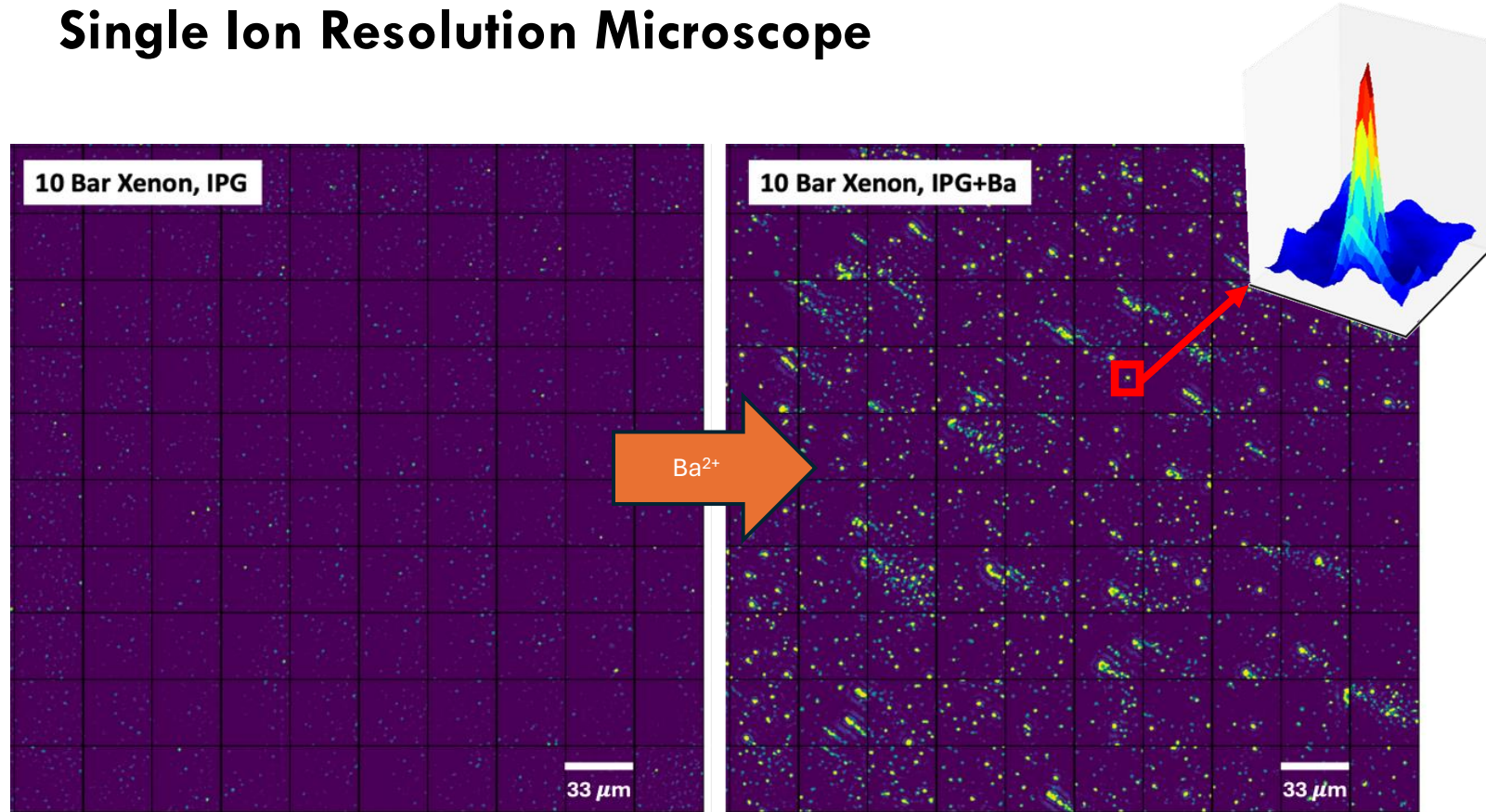
Fluorescence B-color Indicators



Nature 583, 48 (2020)

- [1]. JINST 11 P12011 (2016)
[2]. Phys. Rev. Lett. 120, 132504 (2018)
[3]. Sci Rep 9: 15097 (2019)
[4]. Nature 583, 48 (2020)
[5]. ACS Sensors 6, 1, 192-20 (2021)
[6]. 10.26434/chemrxiv-2023-wxpbh (2023)
[7]. 10.26434/chemrxiv-2024-t81cl (2024)

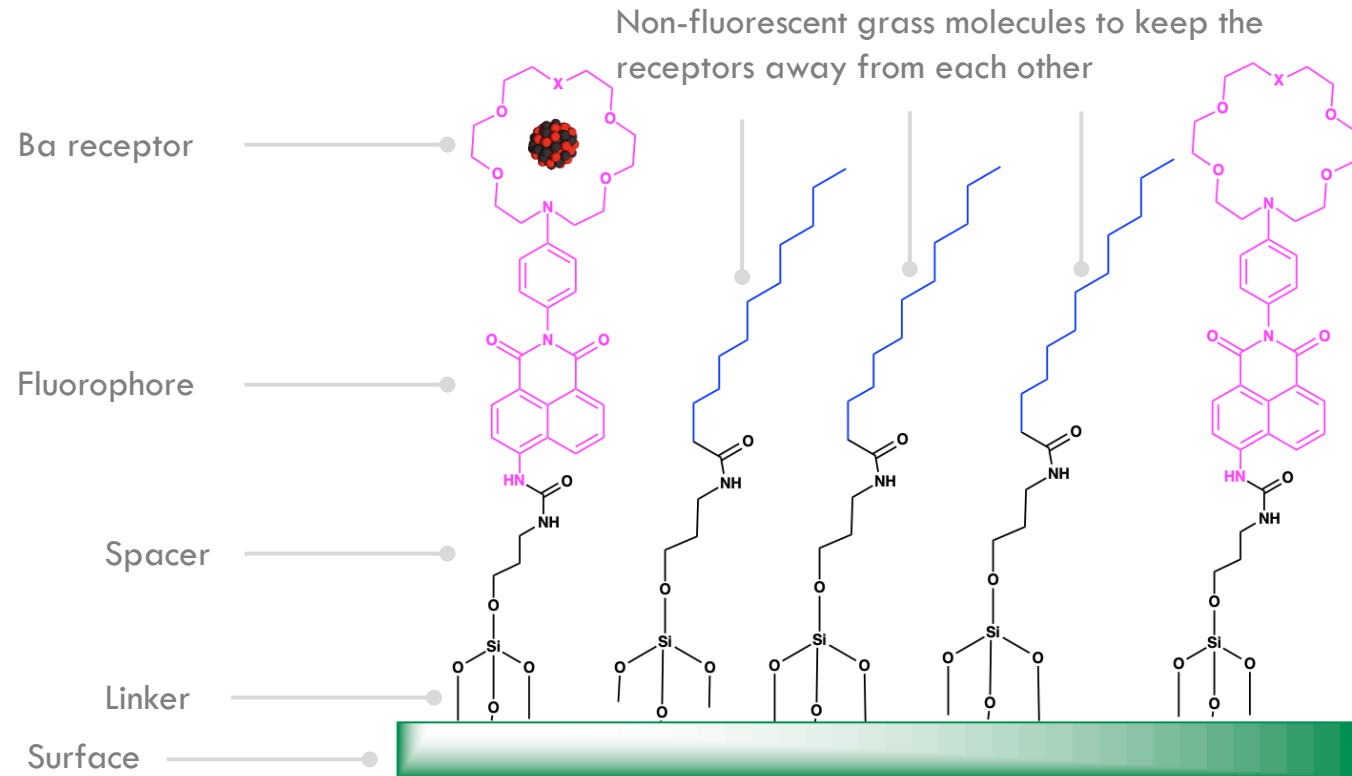
Single Ion Resolution Microscope



A novel fluorescence microscope was developed with wide-field scanning and operational in high-pressure (10 bar) xenon gas.

Nature Communications, 14(10595)

Surface Functionalization

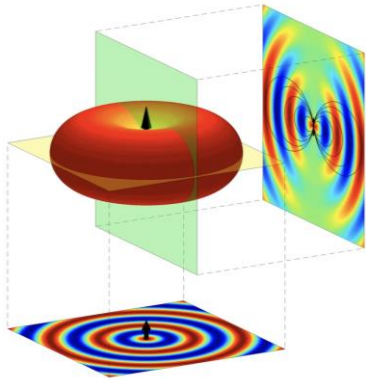


We ideally want:

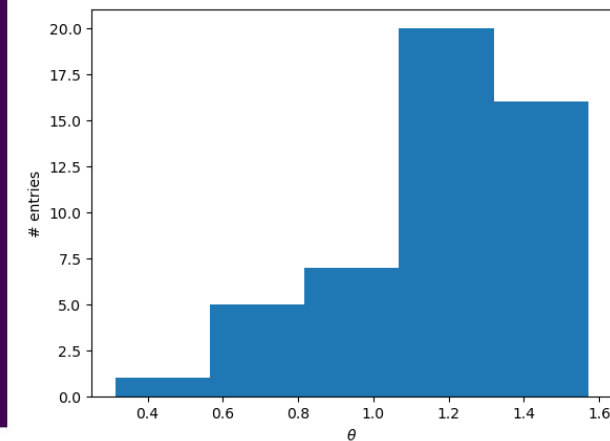
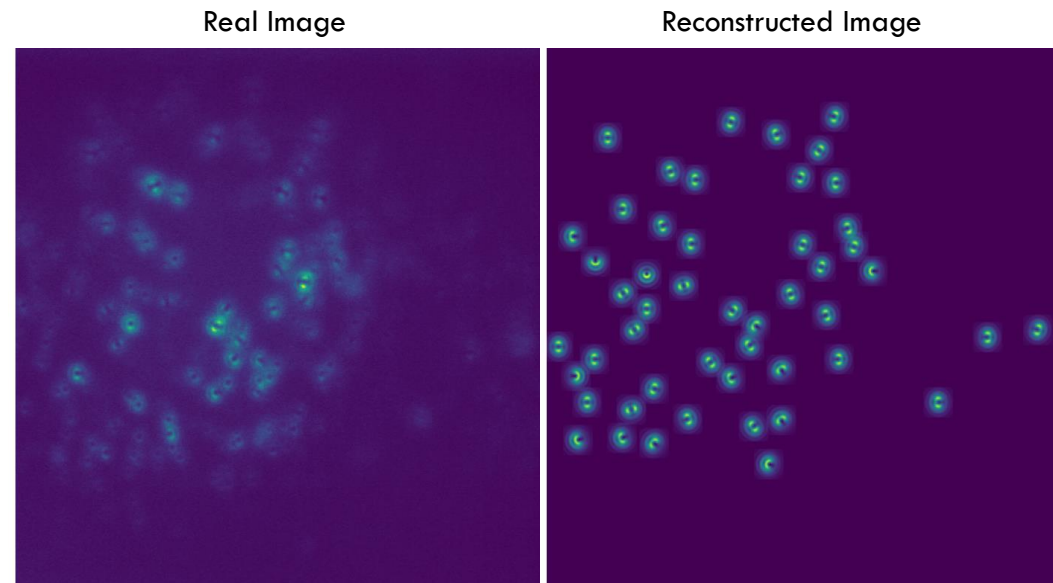
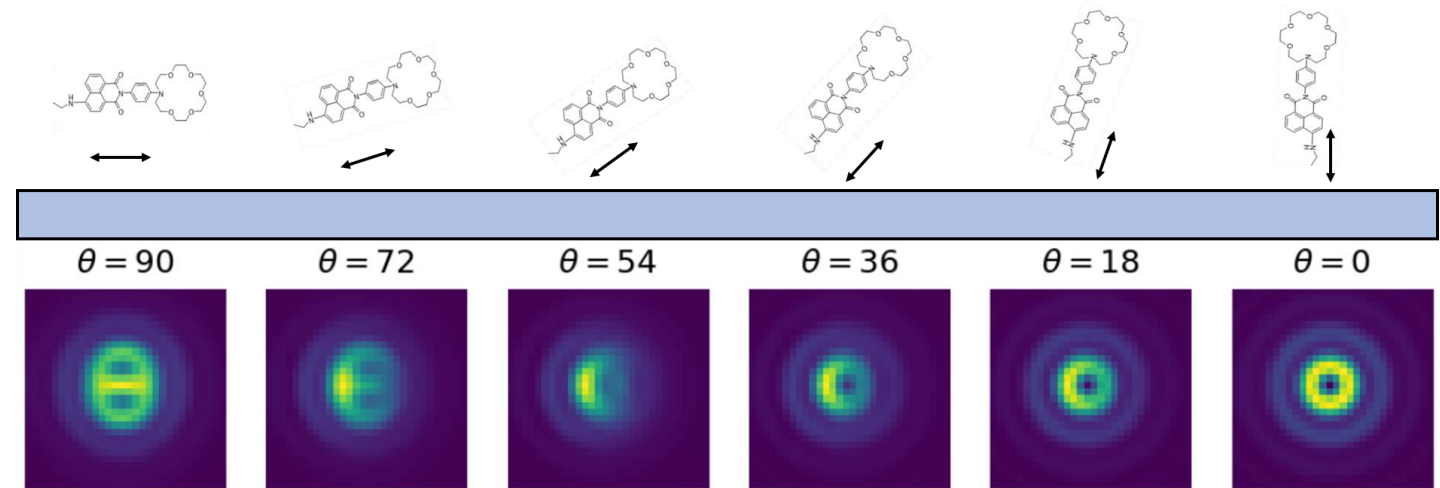
- Receptors close together, but not too close
- Receptors away from the surface for Ba to bind
- High efficiency, low background

Can be achieved only by growing Self Assembled Monolayer (SAM)

Orientation of Molecule on Surface

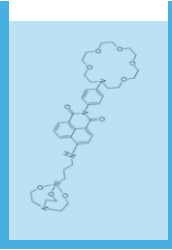


- Fluorescent molecules emit light in dipole radiation pattern.
- In-focus molecules appear as simple dots in the image.
- Defocused (~ 500 nm) images show unique ring patterns that encode the 3D orientation of each molecule's dipole.

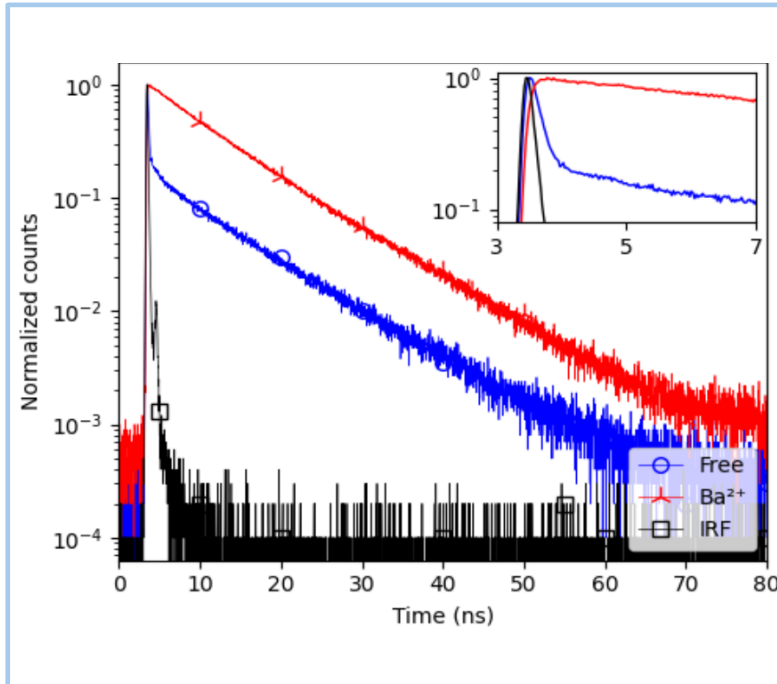


J. Opt. Soc. Am. A 43, 50-58 (2026)

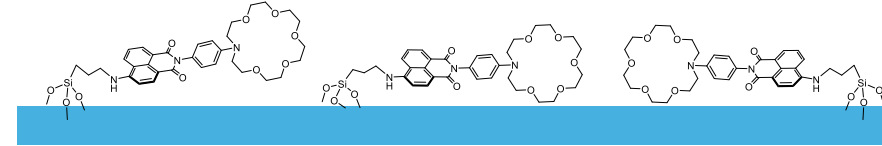
Photobleaching Time Study of Functionalized Molecule



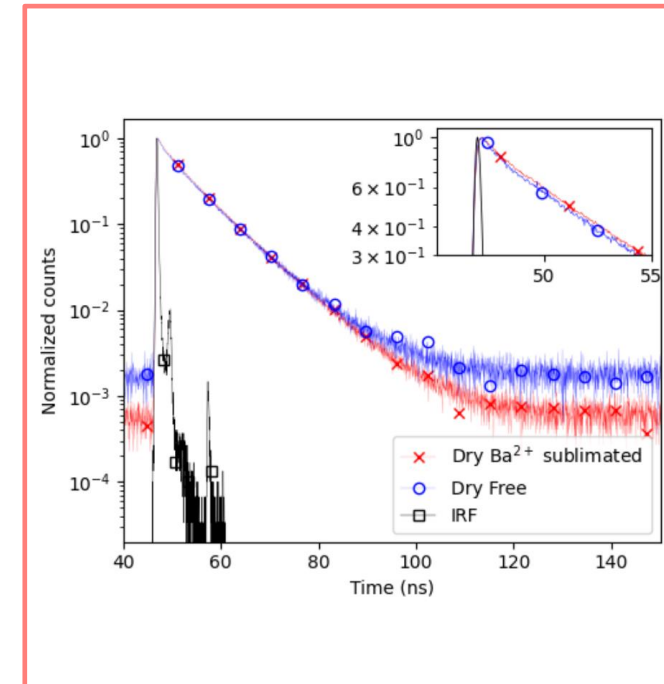
PET ACTIVE



In solution



PET BLOCKED

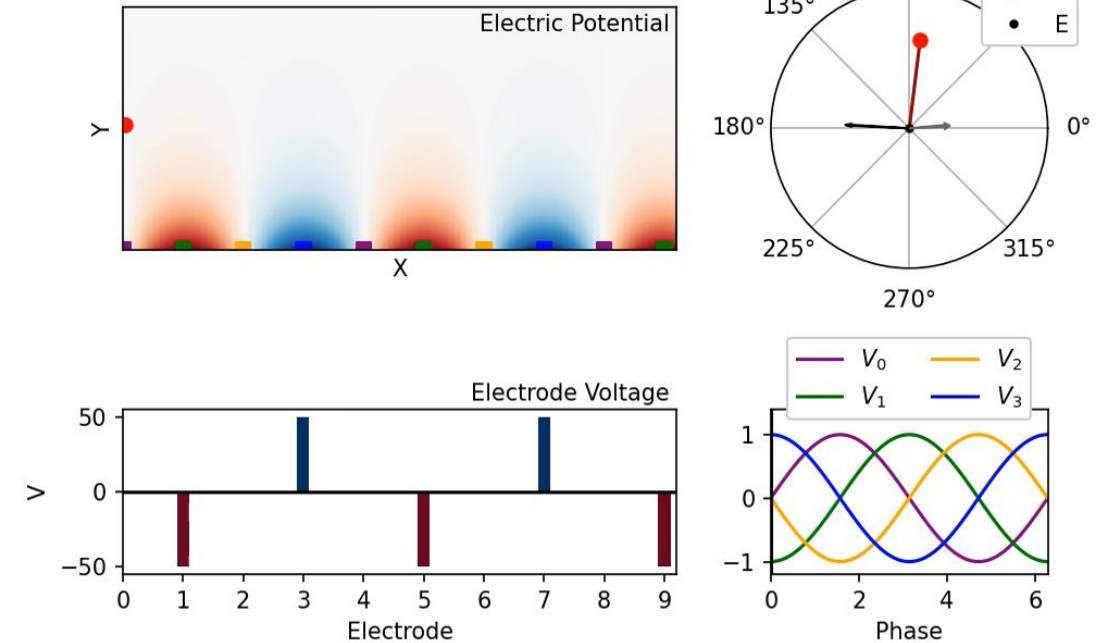
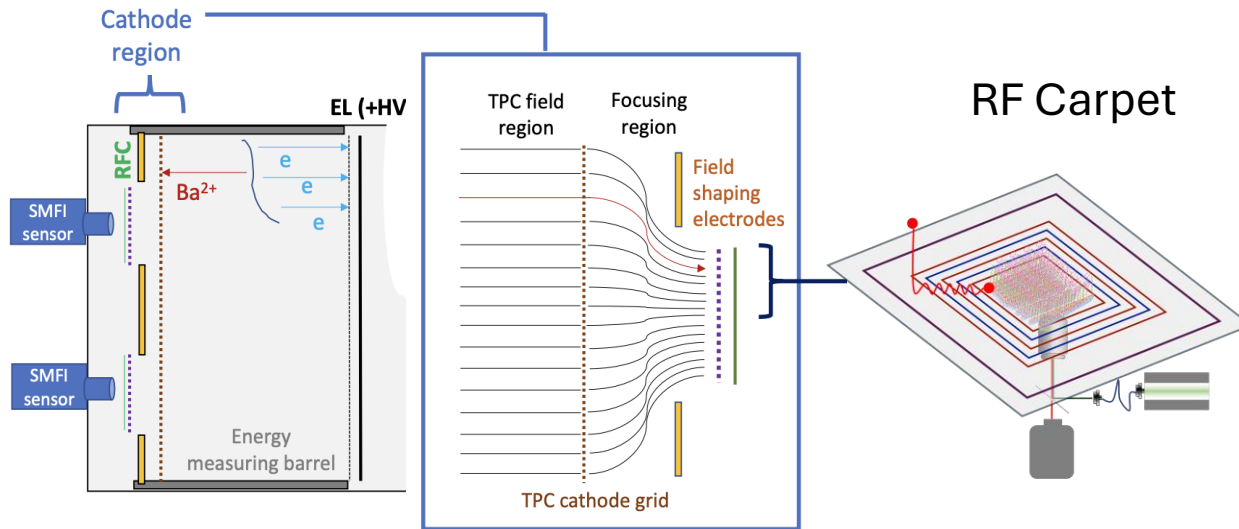


Dry, on surface (air)

Surface immobilization of functionalized molecule almost completely suppresses PET

*Plot credit Mikel Elorza
Paper under collaboration review*

Ion Transport to Sensor



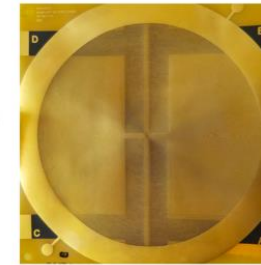
- Array of RF several carpets, each concentrating ions to one single molecule fluorescent imaging ion sensor

- N-phased RF voltages create a traveling wave pattern
- Time-averaged force from electrodes keeps ions levitated above the surface

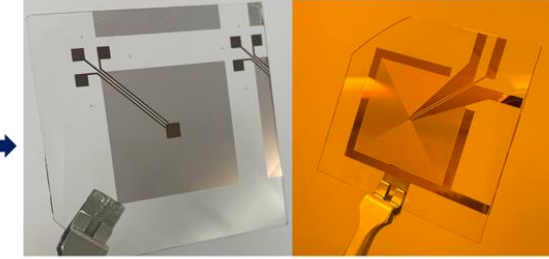
nima.2022.167000
Jones et al

Radiofrequency Carpet for Ba Ion Transport

ν BIT

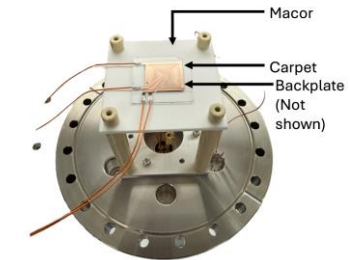
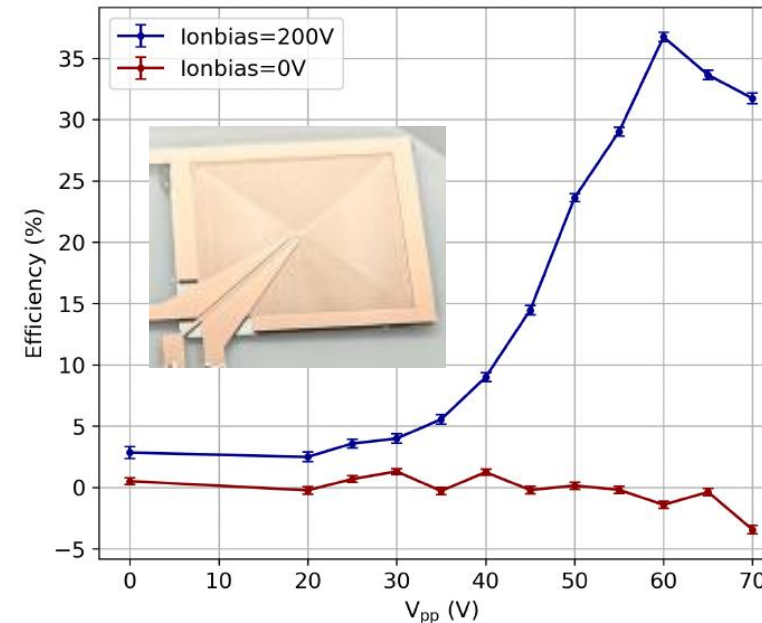


Current gen:
Pitch=160um
PCB on polyimide

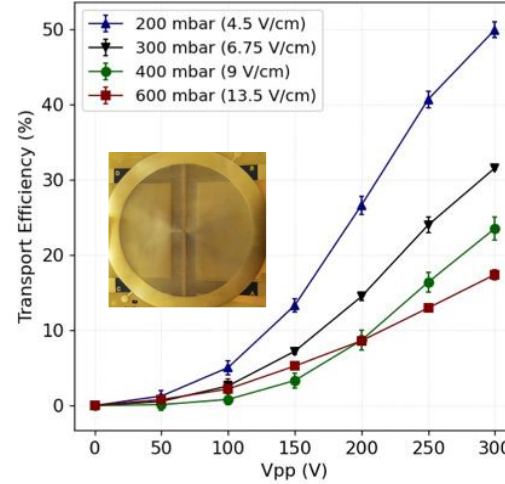
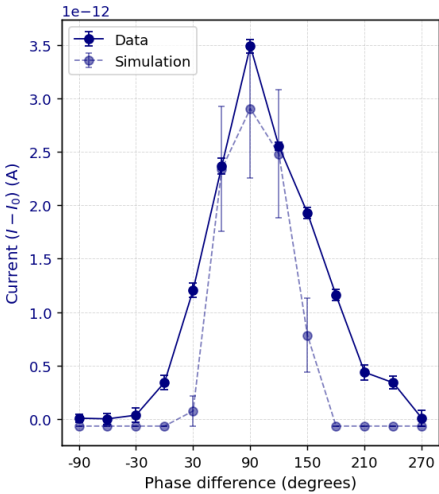


Next gen:
Pitch=50um
Cr+Cu microfab on Fused Silica
(1-5 bar)

- To go higher in pressure you need lower pitch RF carpet

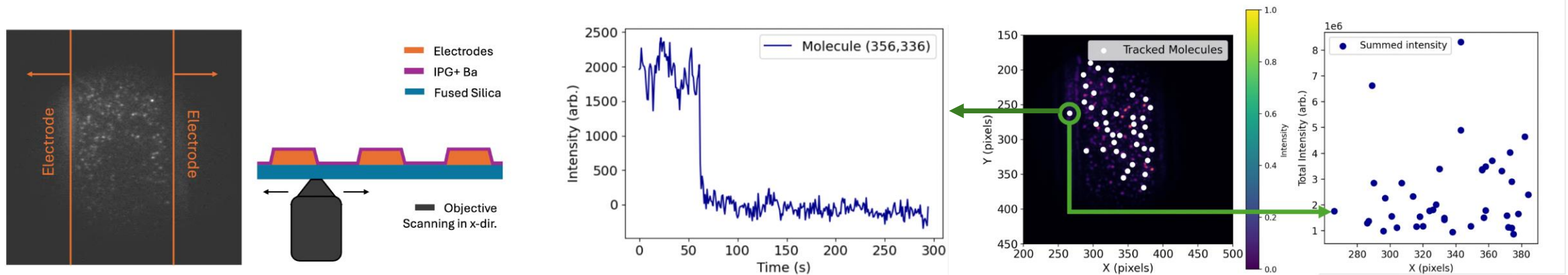


- Preliminary result ν BIT working at 500 mbar
- It works in 1 bar pressure



- Cs⁺ used as mass-proxy for Ba²⁺
- 4-phase mode confirms transport is driven by the traveling wave: signal peaks at 90°, vanishes at 0°/180°

SMFI on vBIT



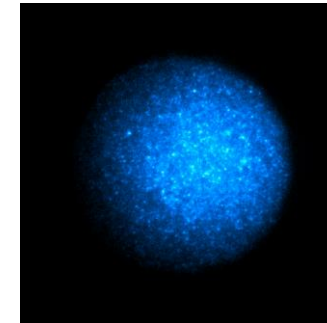
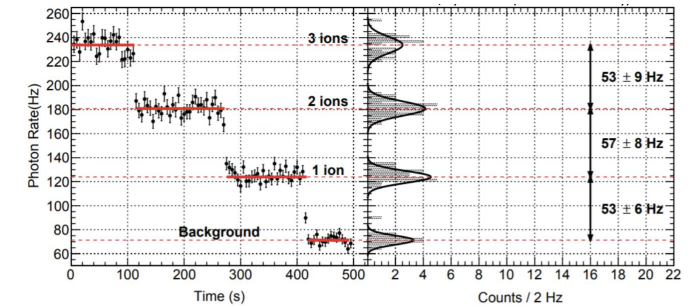
- vBIT: a photonic chip-based sensor combining optical microscopy with microfabricated RF carpets, for full-detector coverage.
- SMFI has been demonstrated directly on vBIT.

Summary

Barium tagging, if realized, can dramatically increase the sensitivity of a $0\nu\beta\beta$ search.

Barium tagging has been explored in liquid, solid, and gas xenon.

- **Liquid Xe:** single Ba^+ ion trapped and identified by laser fluorescence.
- **Solid Xe:** single Ba atom/ion detected via cold-probe capture and CCD imaging.
- **Gas Xe:** single Ba^{2+} ion imaged using fluorescent sensor molecules.



Each approach has some milestones achieved and some still open — continuous work is ongoing in all three sectors to demonstrate the full end-to-end technology.

Thank You