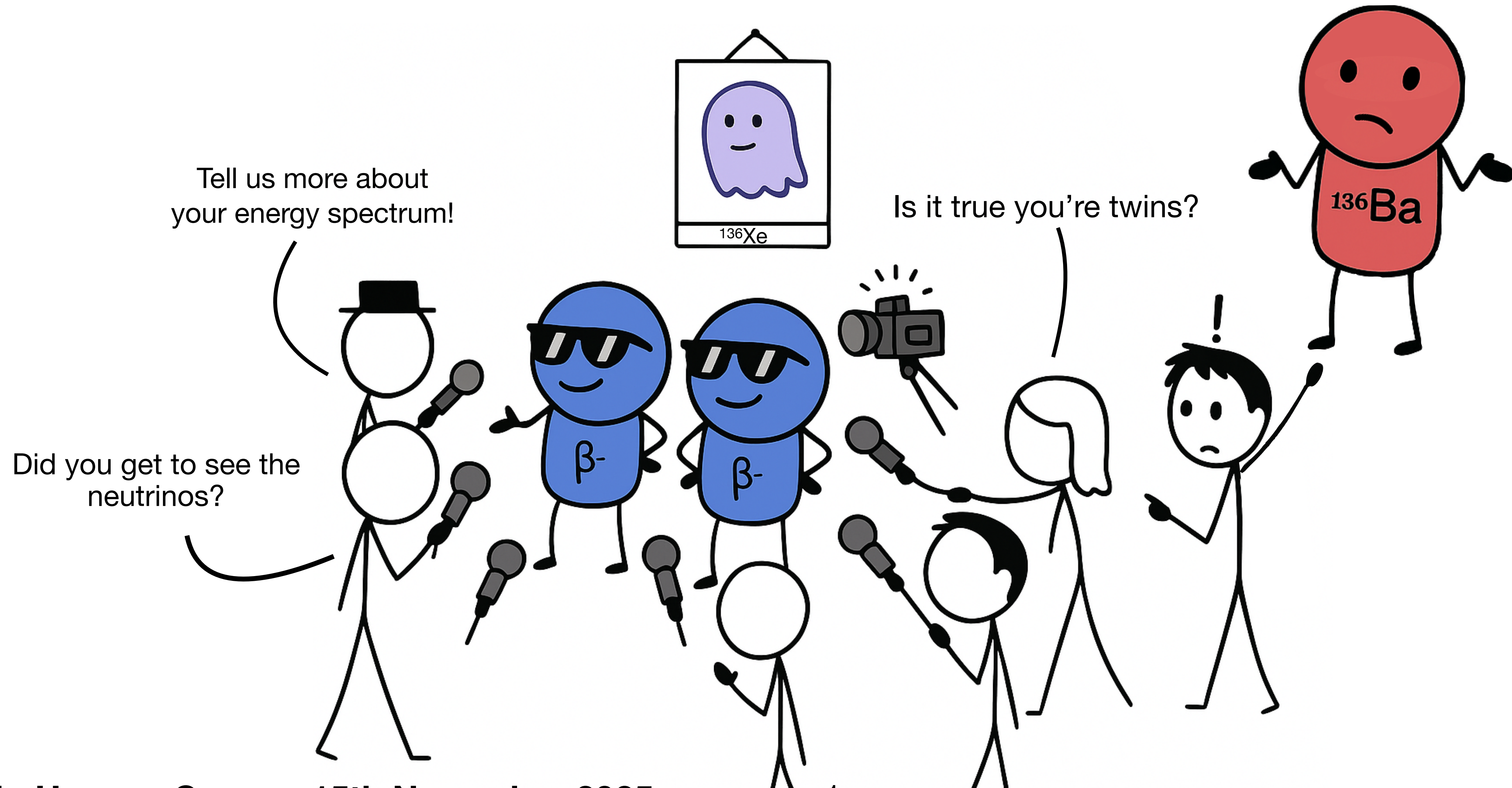


Barium Tagging for $0\nu\beta\beta$ searches

Overview and current efforts in NEXT and nEXO



Why do we care about Barium?

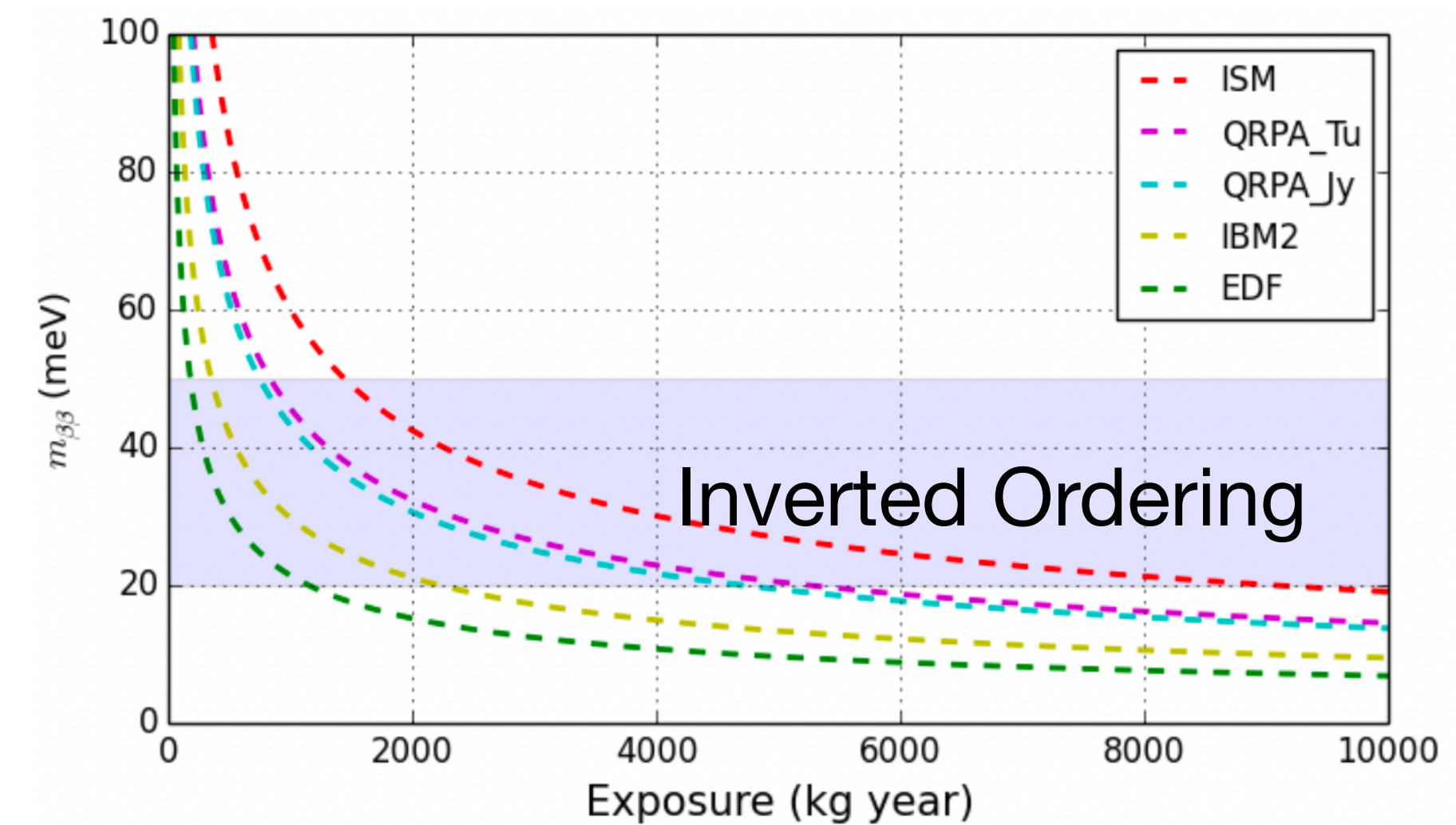
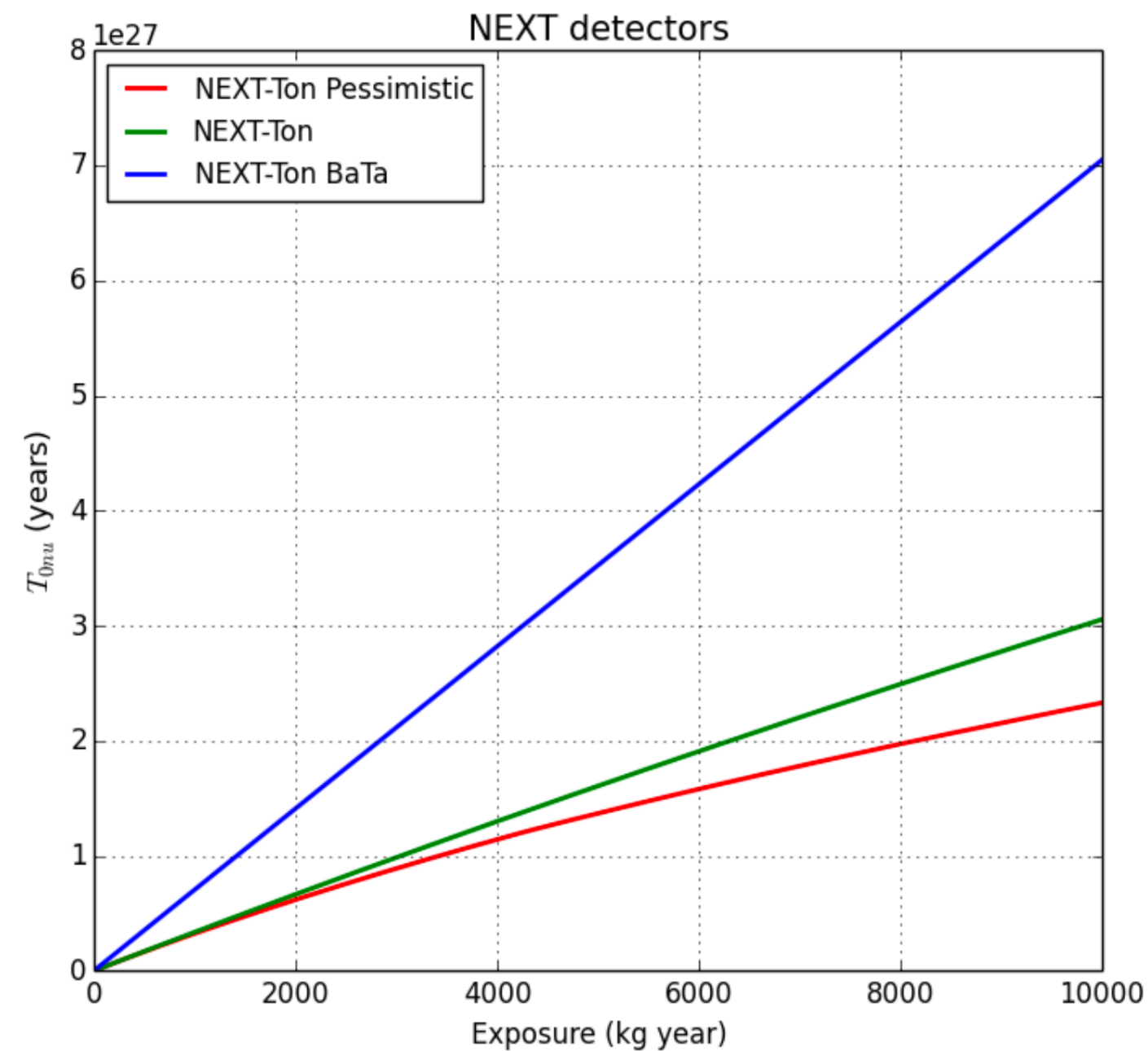
- Next generation Xenon TPCs need to reach lifetimes $> 10^{27}$ years with one or several tons of Xe.
- At the same time, background needs to be mitigated down to the level of 10^{-3} counts/kg/year.
- The presence of Barium is an indisputable evidence of the $\beta\beta$ reaction occurrence:



- Different approaches for GXe (Ba^{2+}) and LXe (Ba^+).

How far could we go?

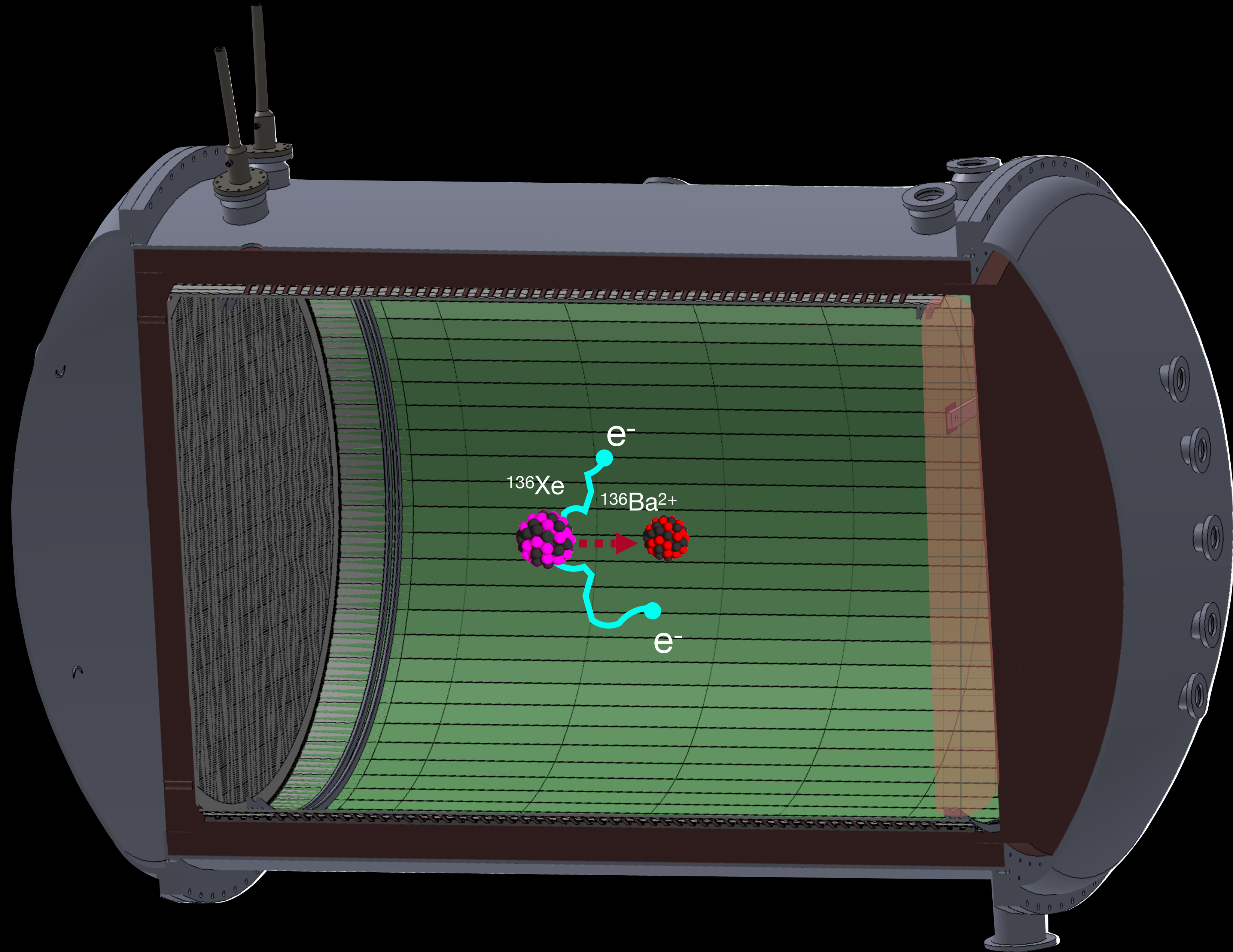
- Linear scale of sensitivity to $\tau_{0\nu\beta\beta}$ would with exposure if zero background.
- The full IO (blue shaded area) of $m_{\beta\beta}$ could be probed with 1 ton \times 10 years exposure.

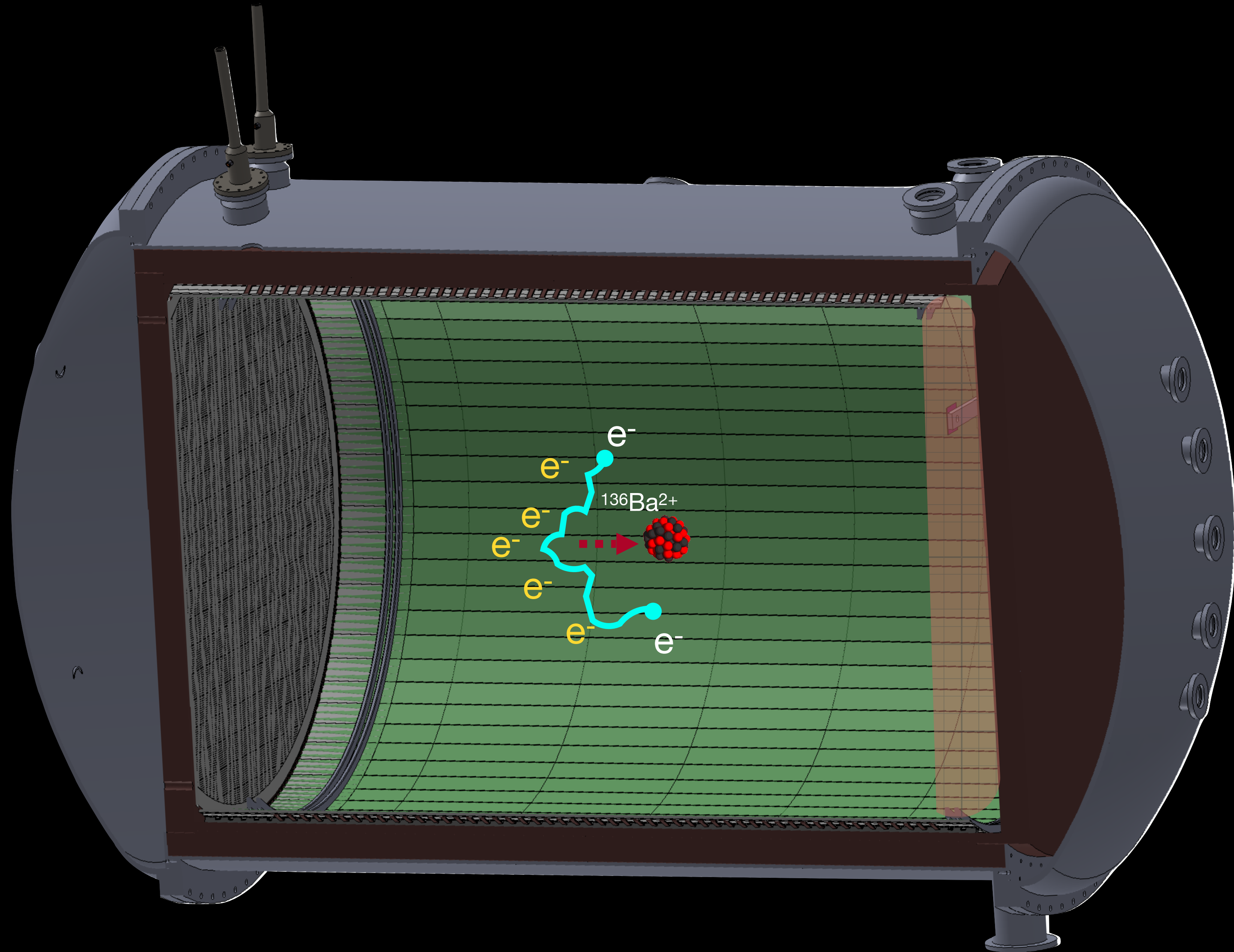


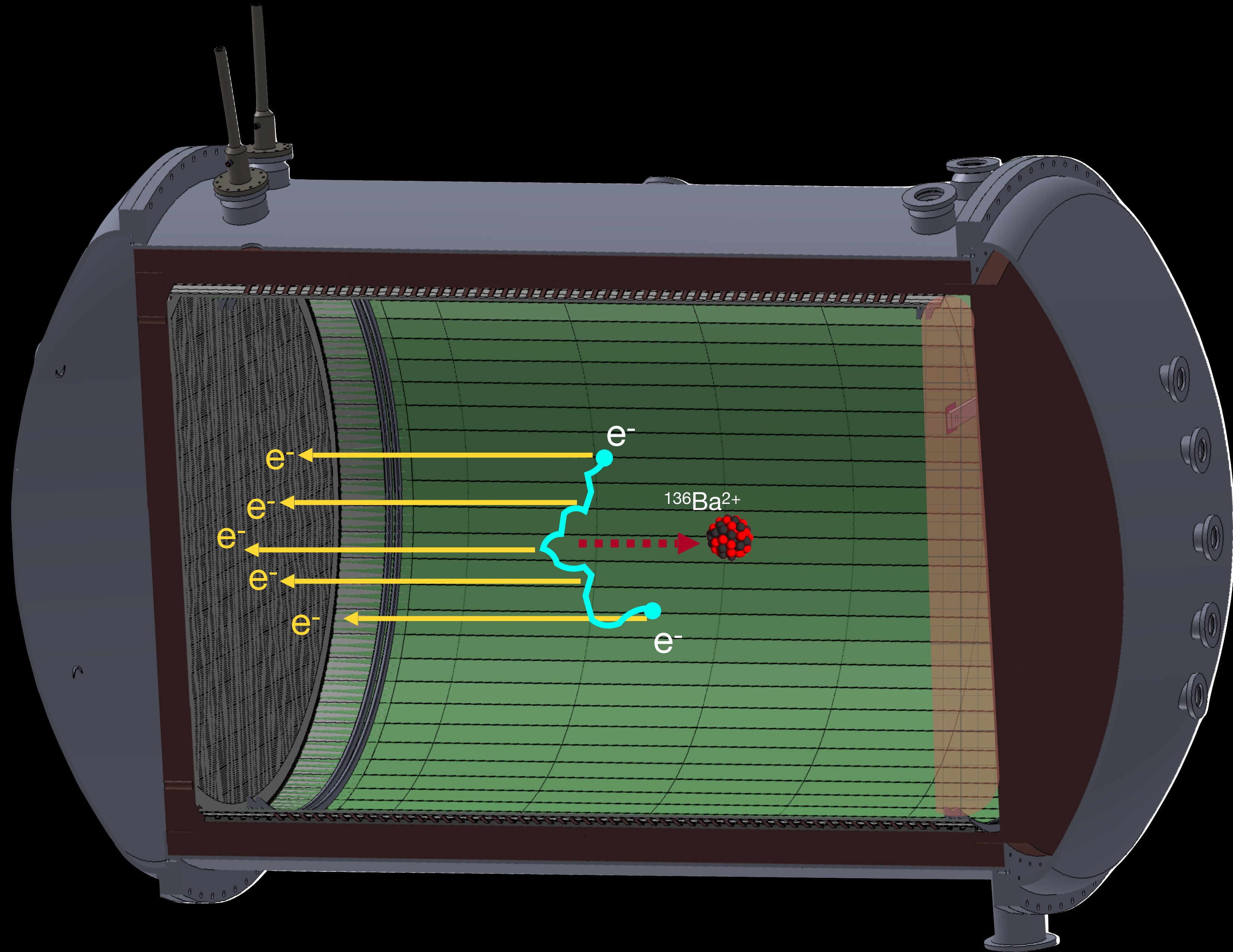
From J. Muñoz (2018) PhD Thesis

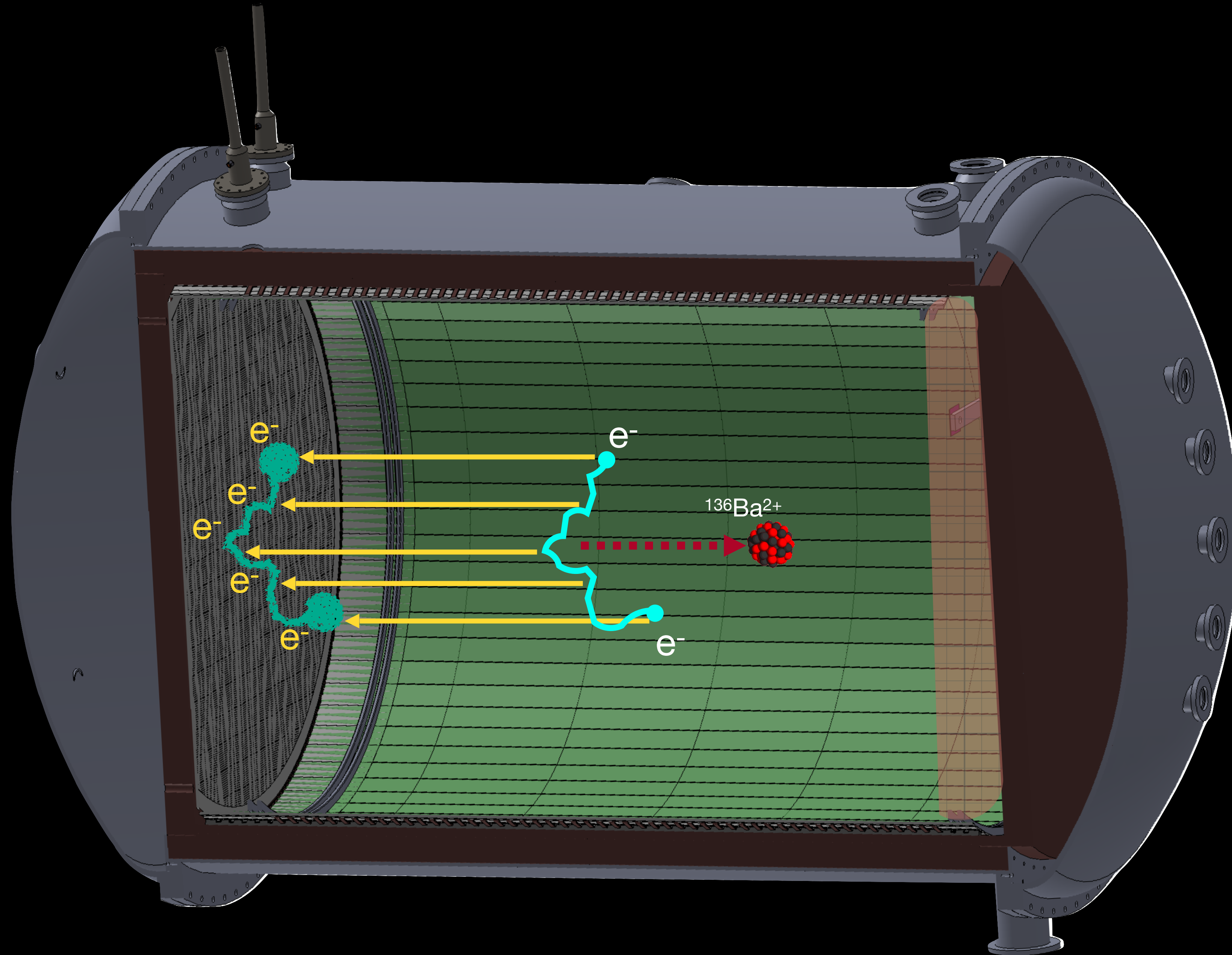
Overview

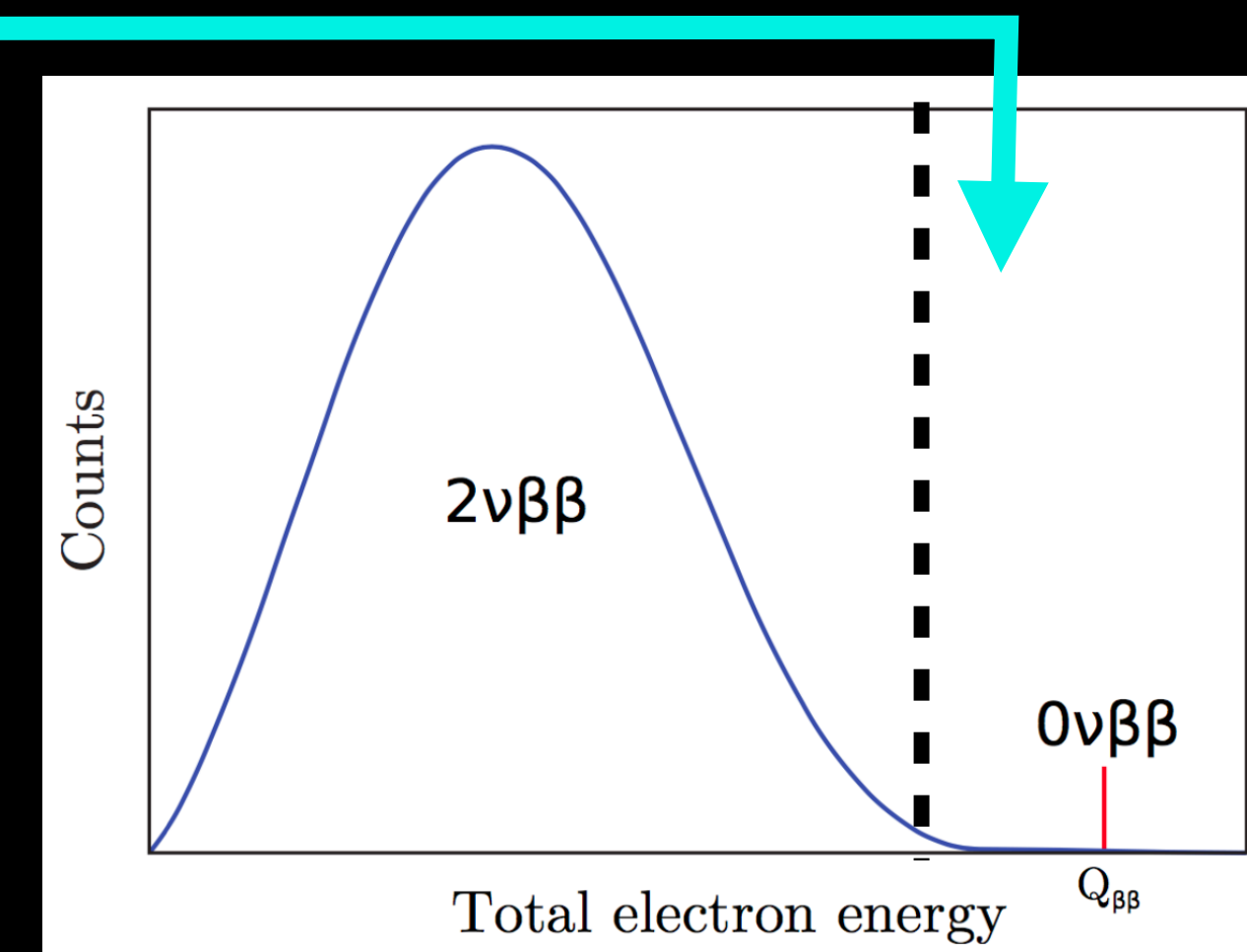
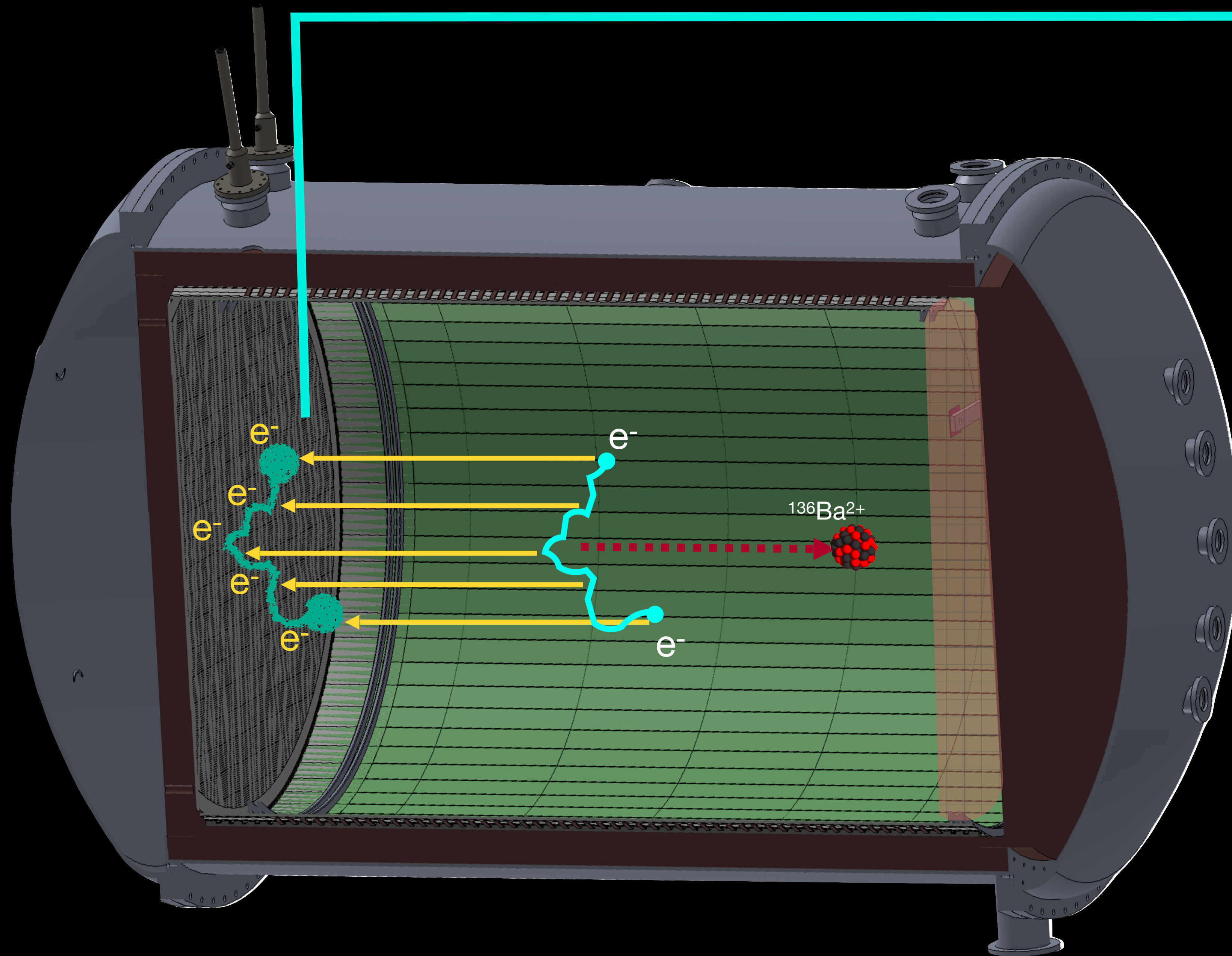
- ☒ Introduction to Ba-tagging
- ☐ **Ba-Tagging in NEXT**
 - ☐ **System design**
 - ☐ Chemo-sensors characterization
 - ☐ Optical detection
 - ☐ Ion sources and guiding systems
- ☐ Ba-Tagging in nEXO
- ☐ The road forward

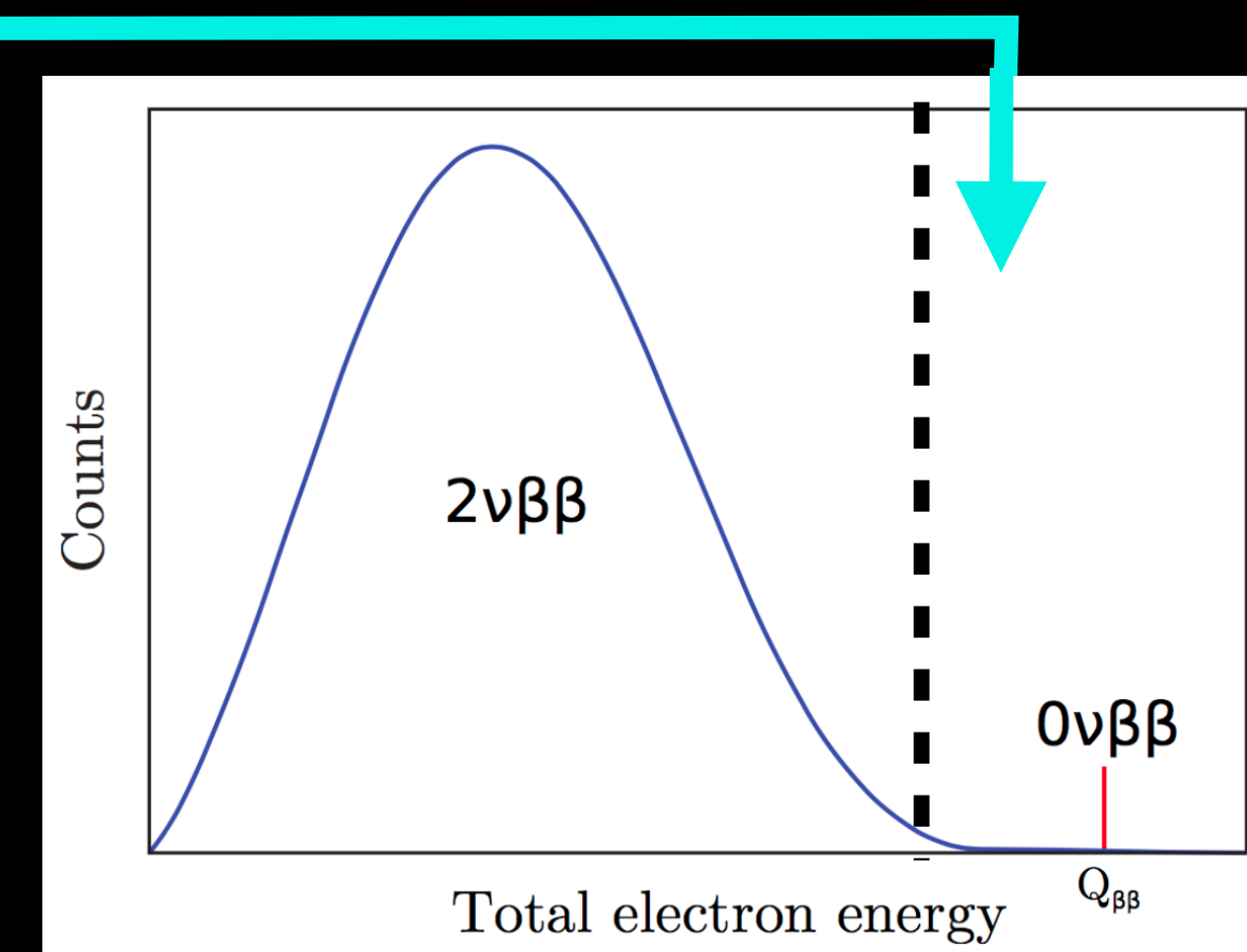
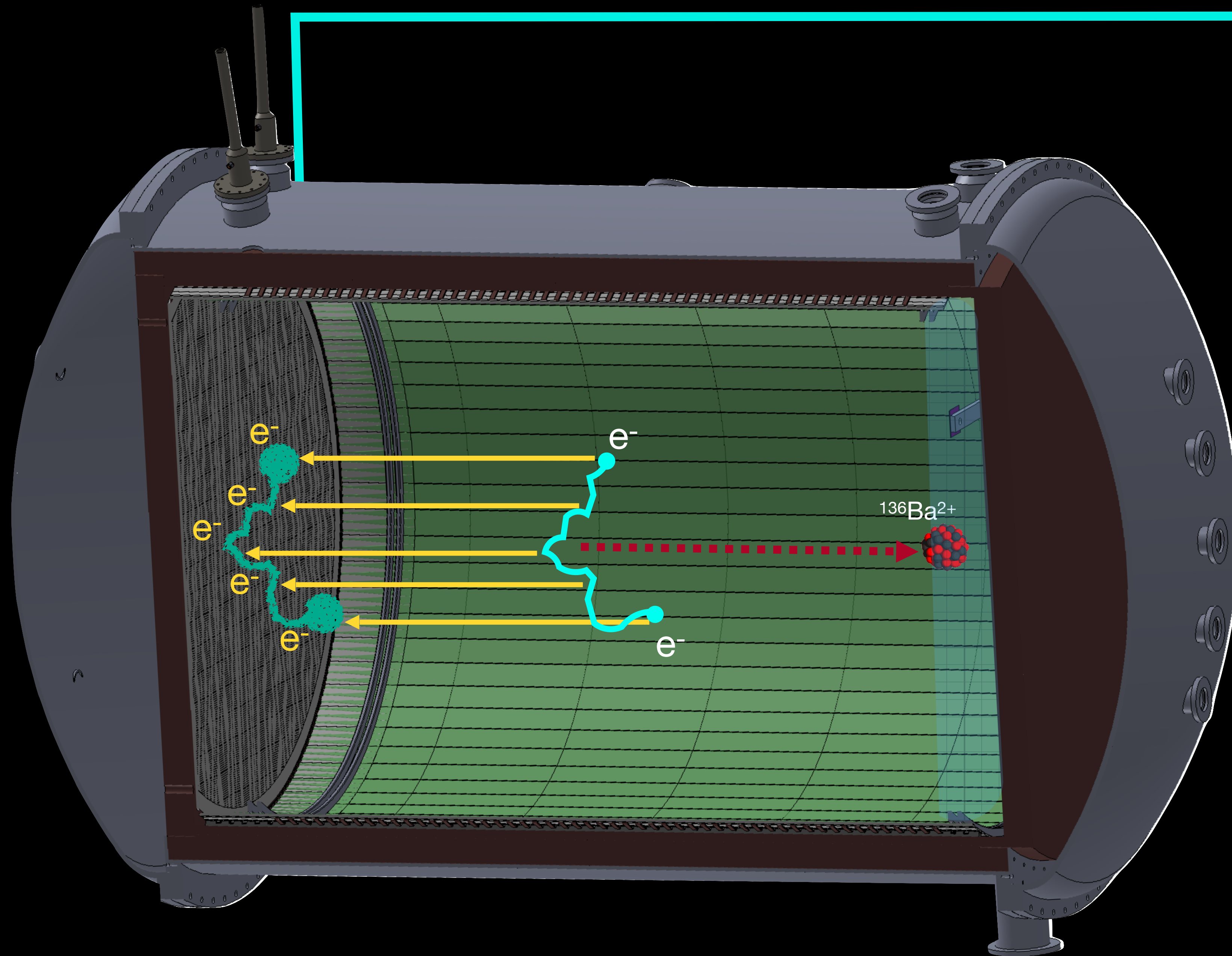


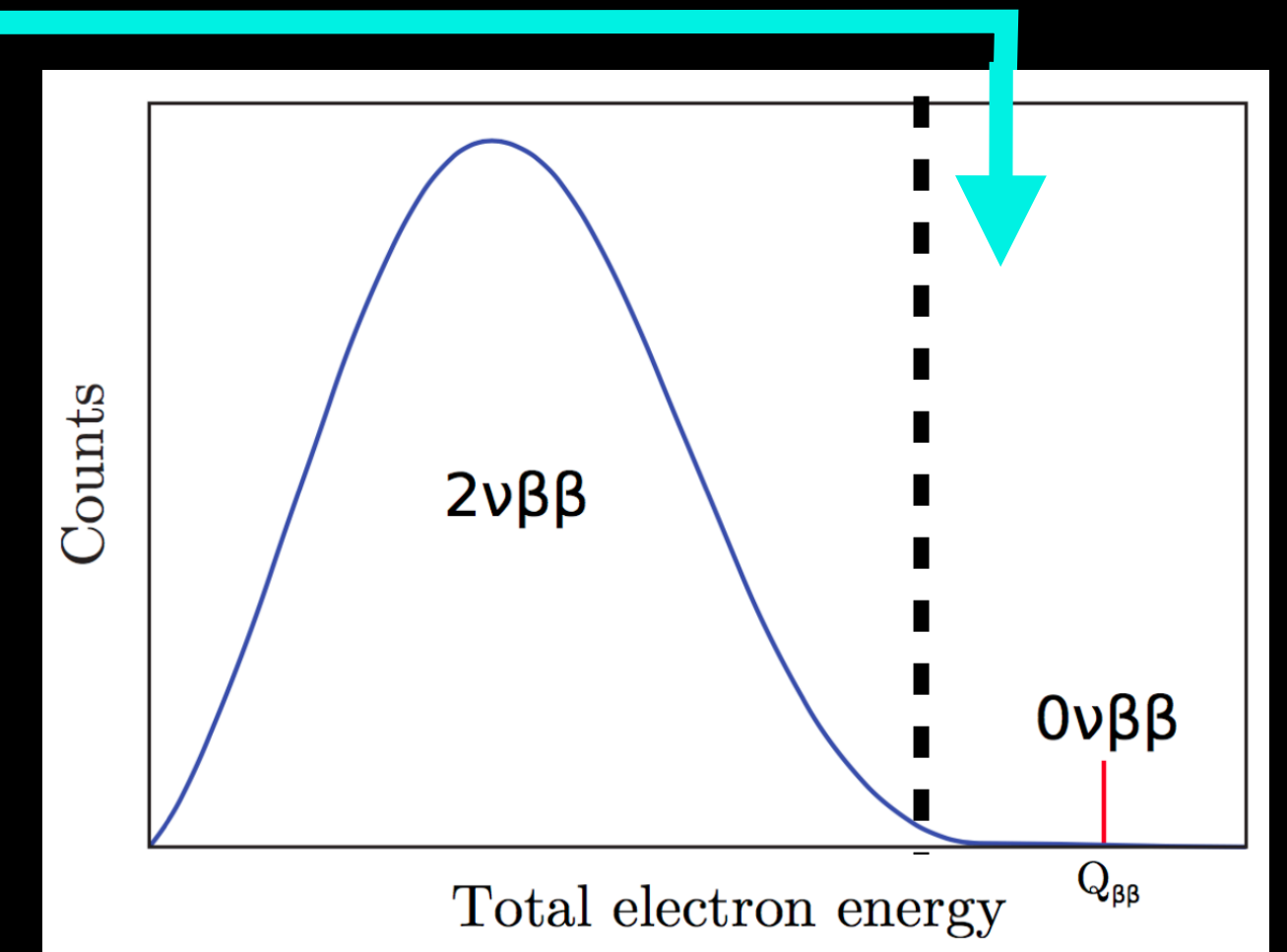
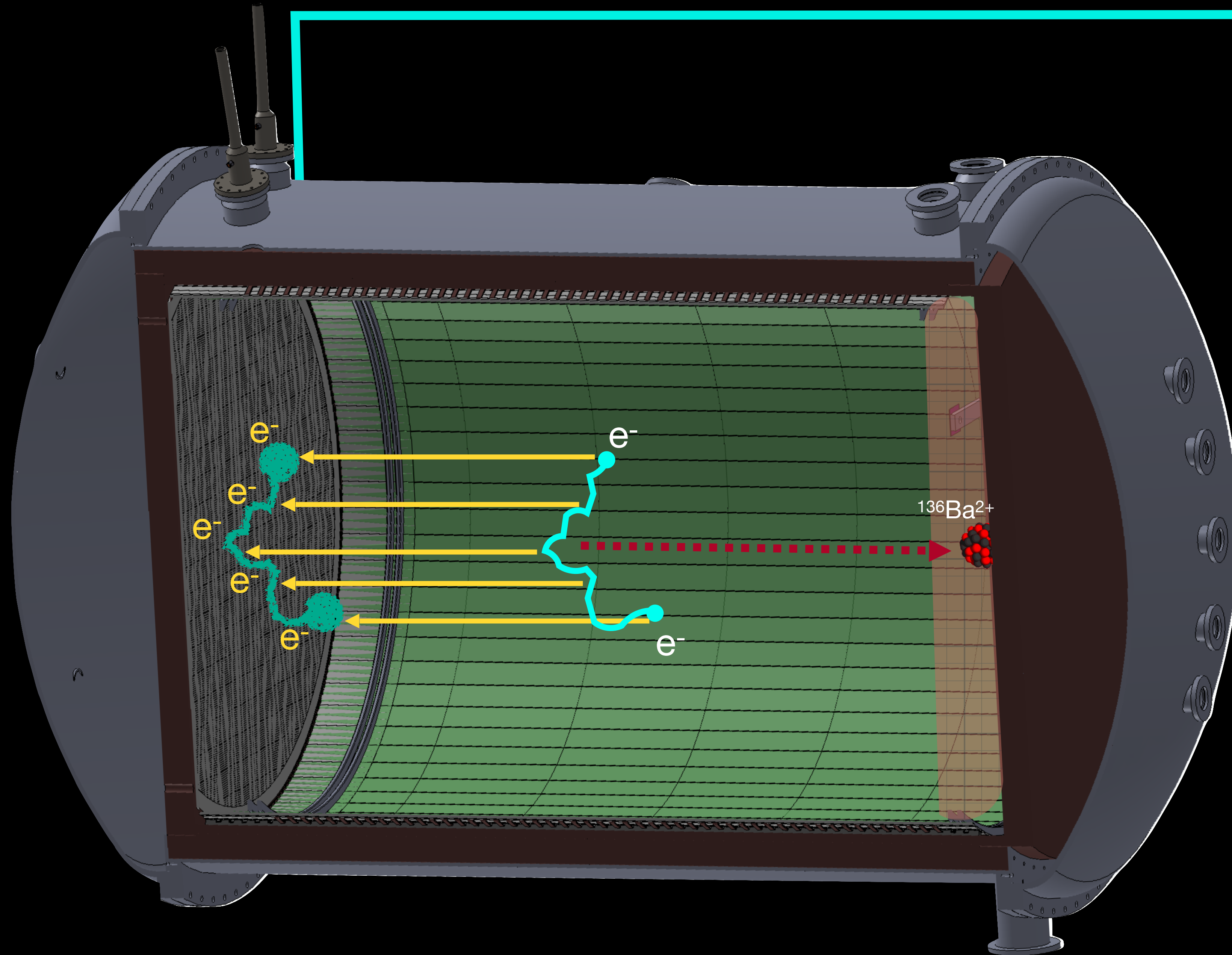


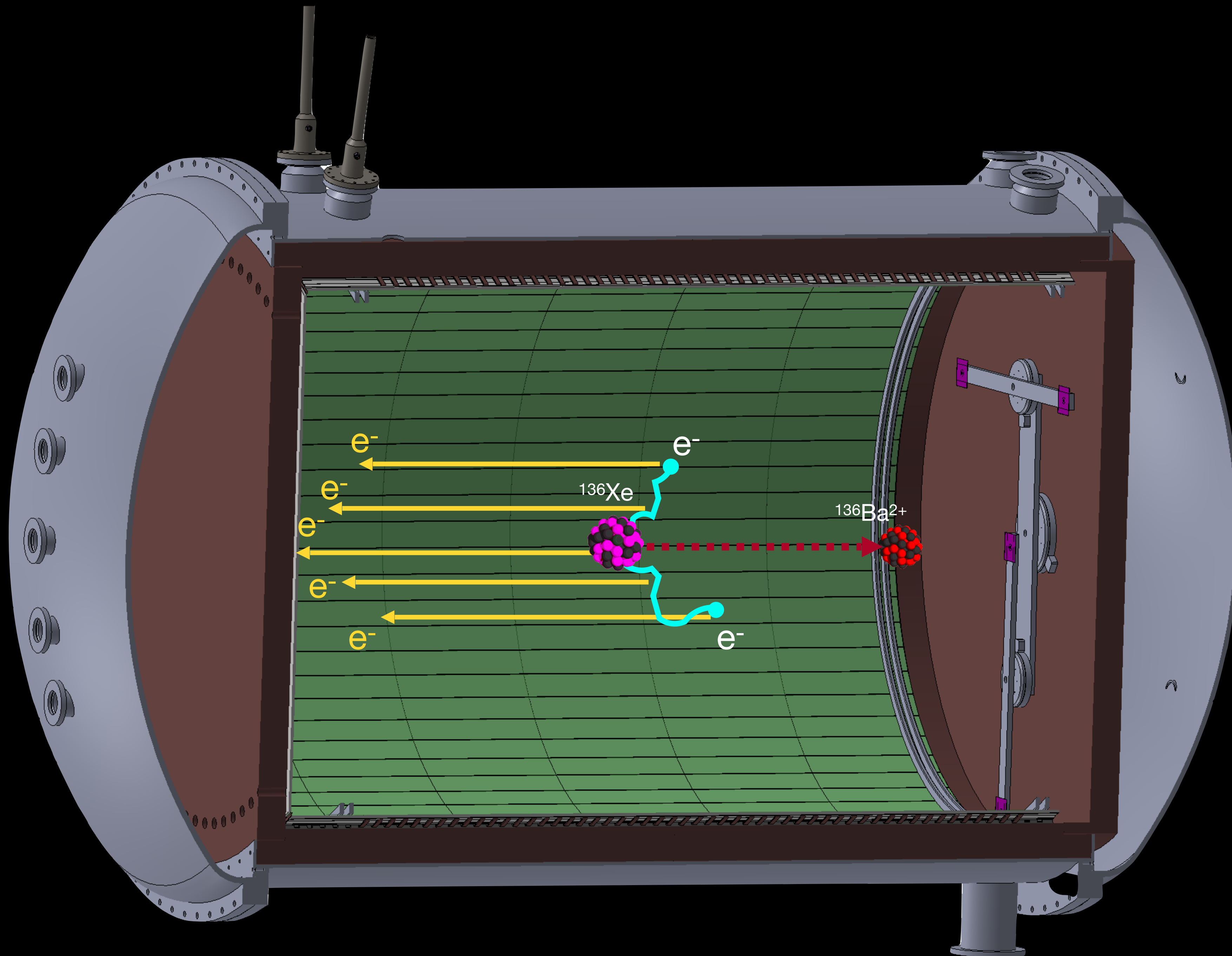


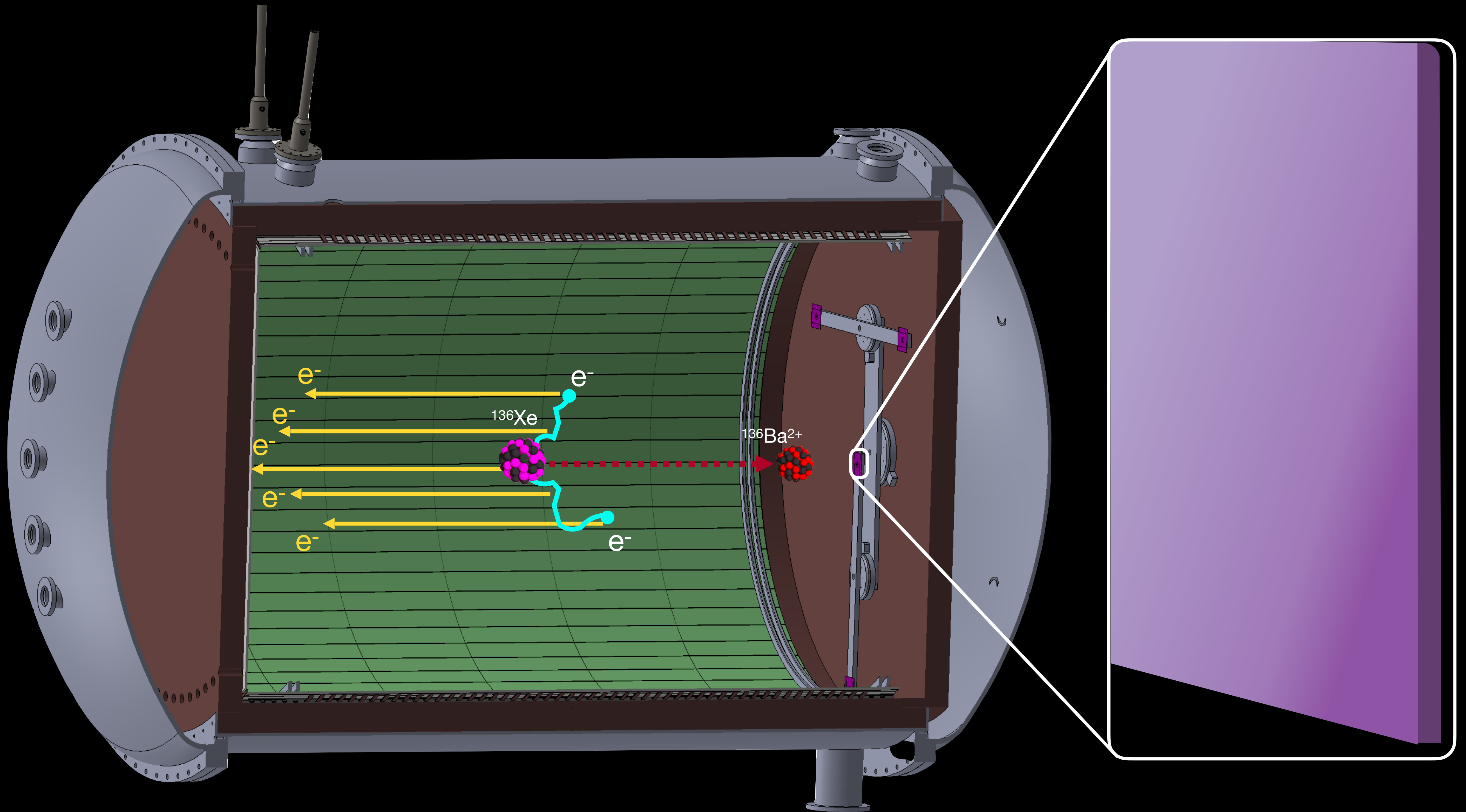


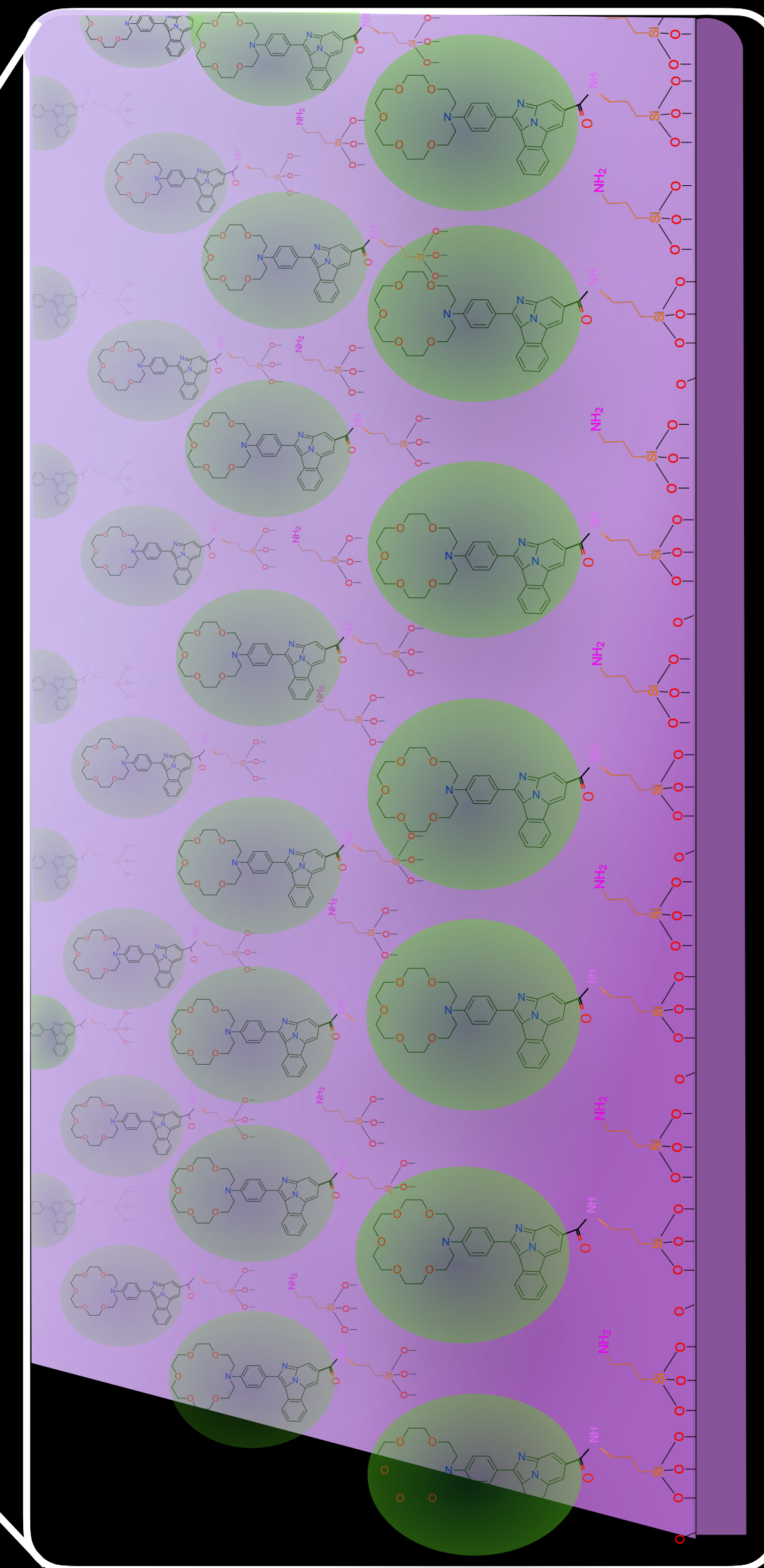
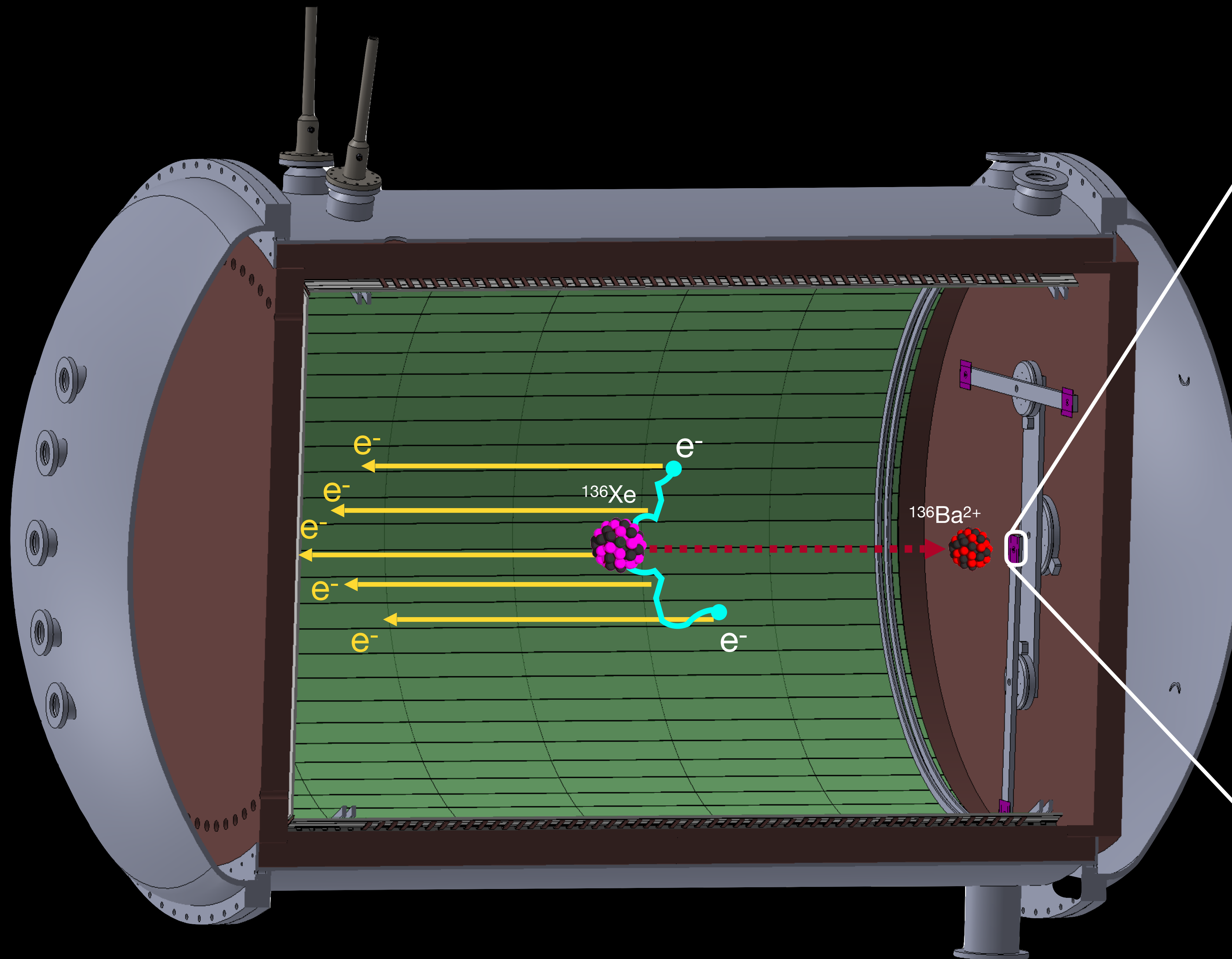


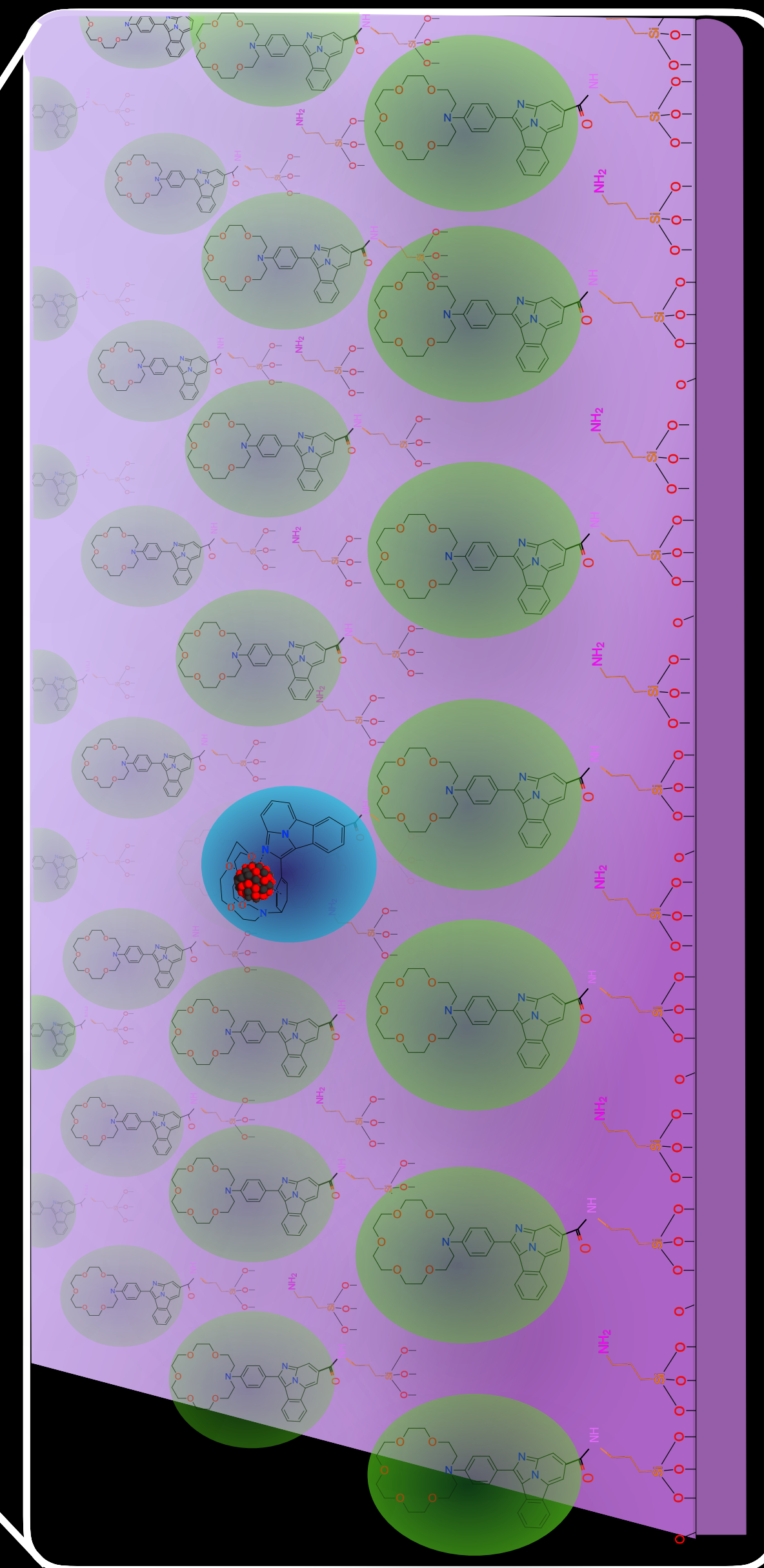
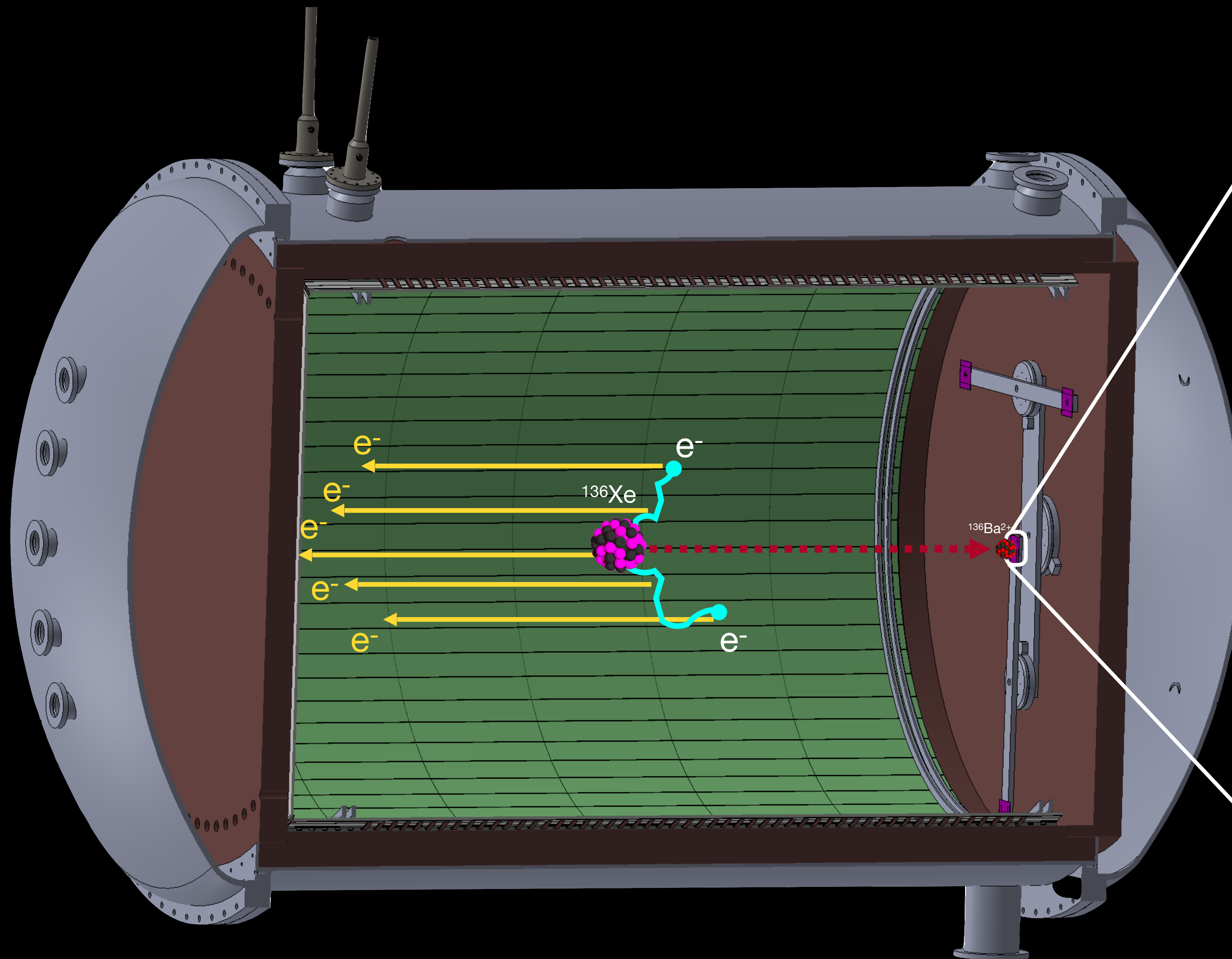










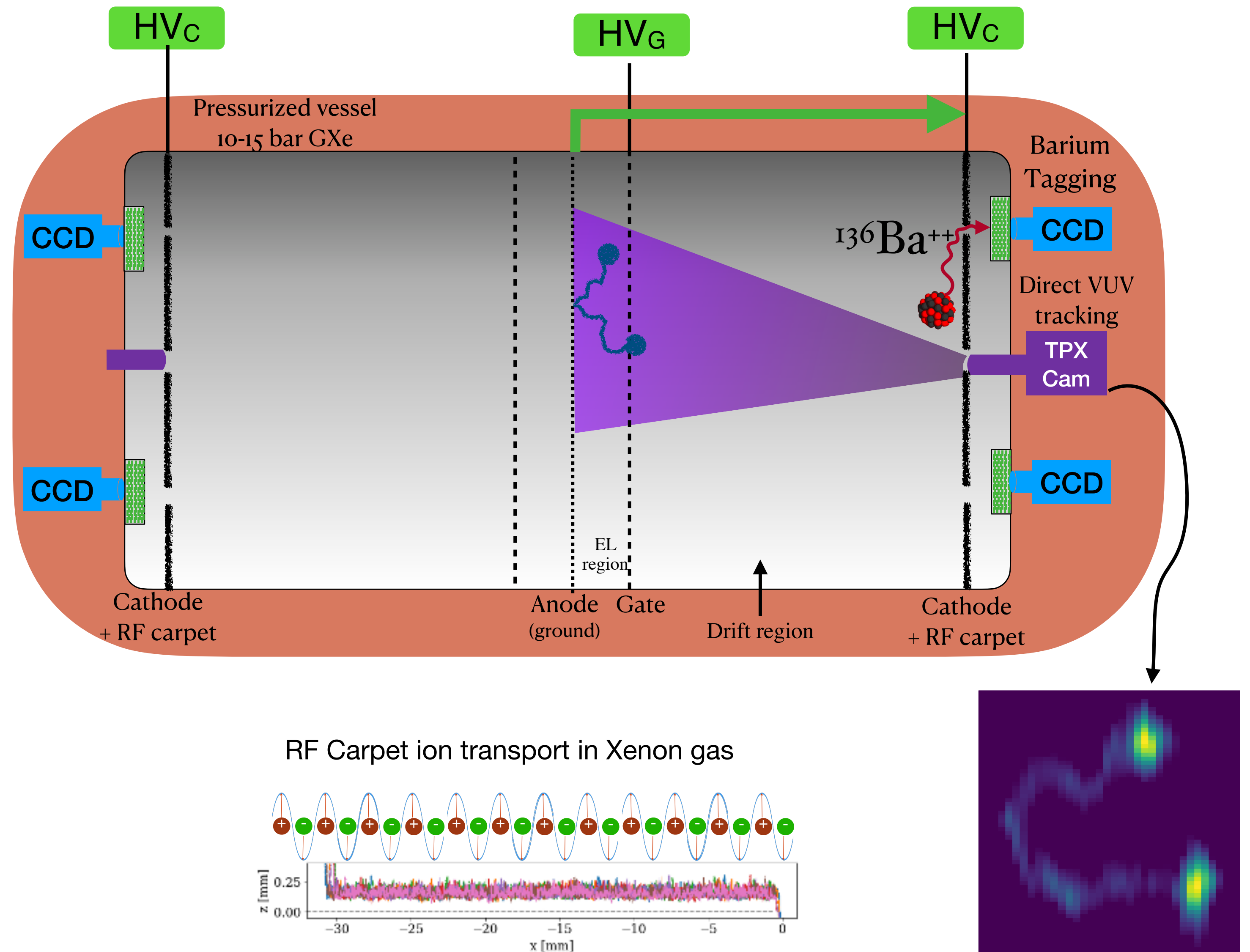


Sensor design lines in GXe

[a] JINST 18 P08006 (2023)

The CRAB concept^[a]

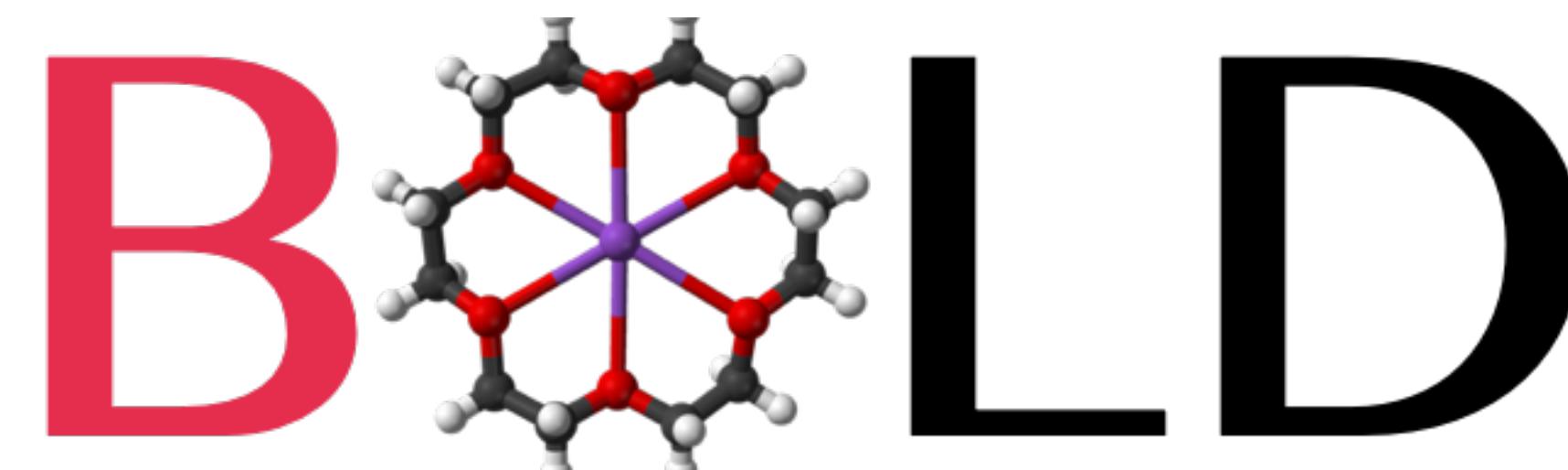
- TimePix camera to detect the e- track is detected
- Ba^{2+} ions concentrated by RF carpets into small sensor areas
- In-situ detection by laser microscopy for delayed coincidence signal.



R&D into Chemosensors

The building blocks of the BaTag system

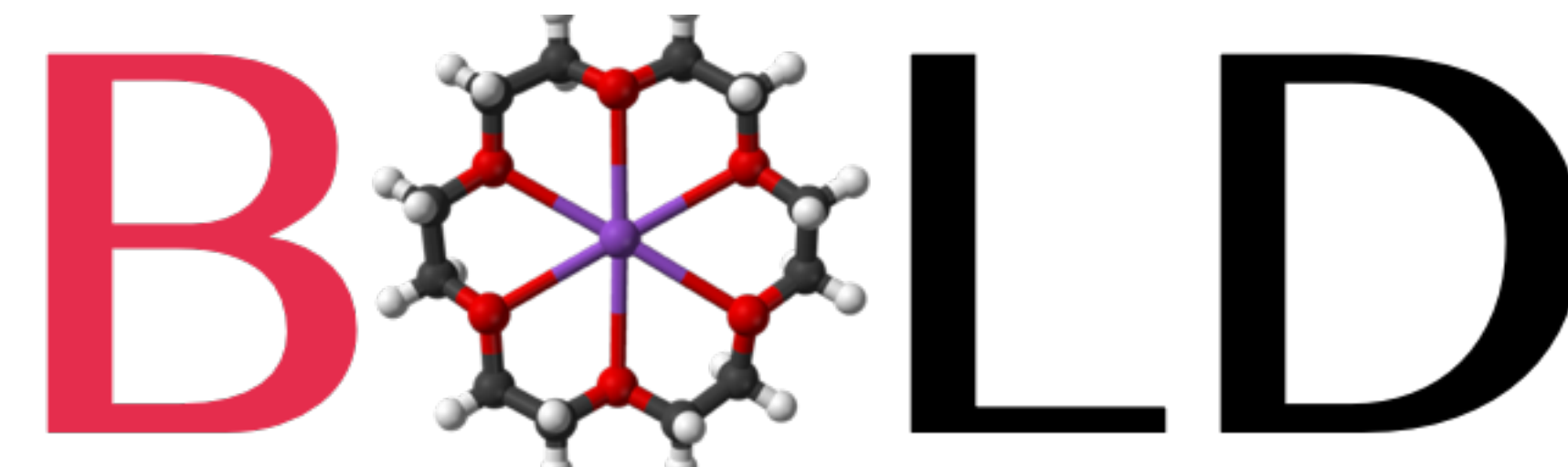
- Exploit three properties of light emission:



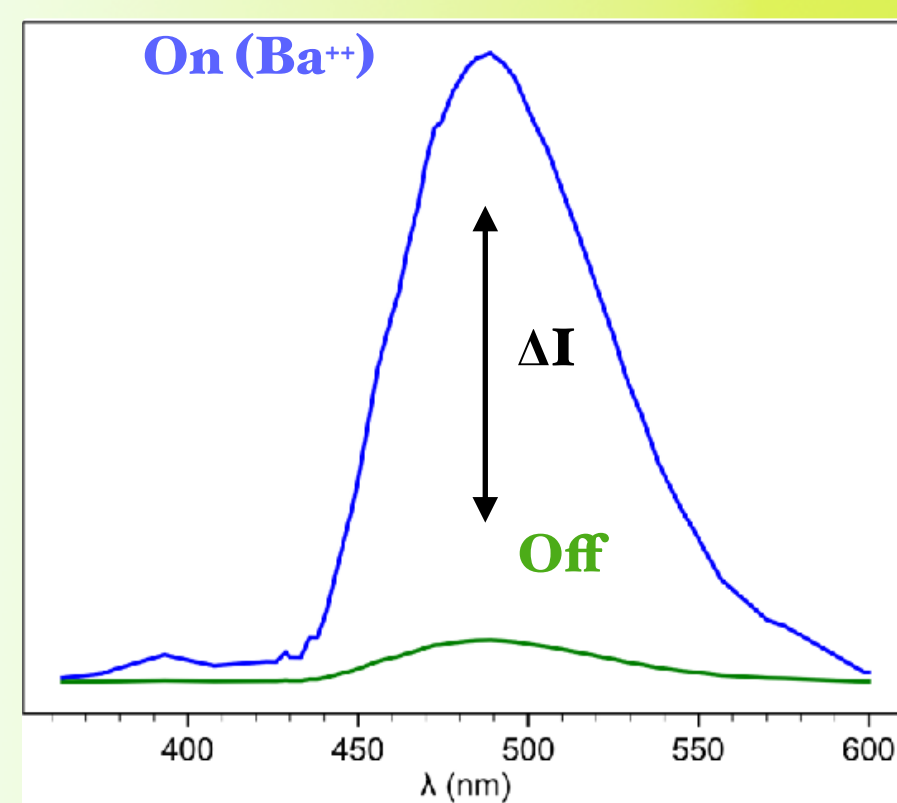
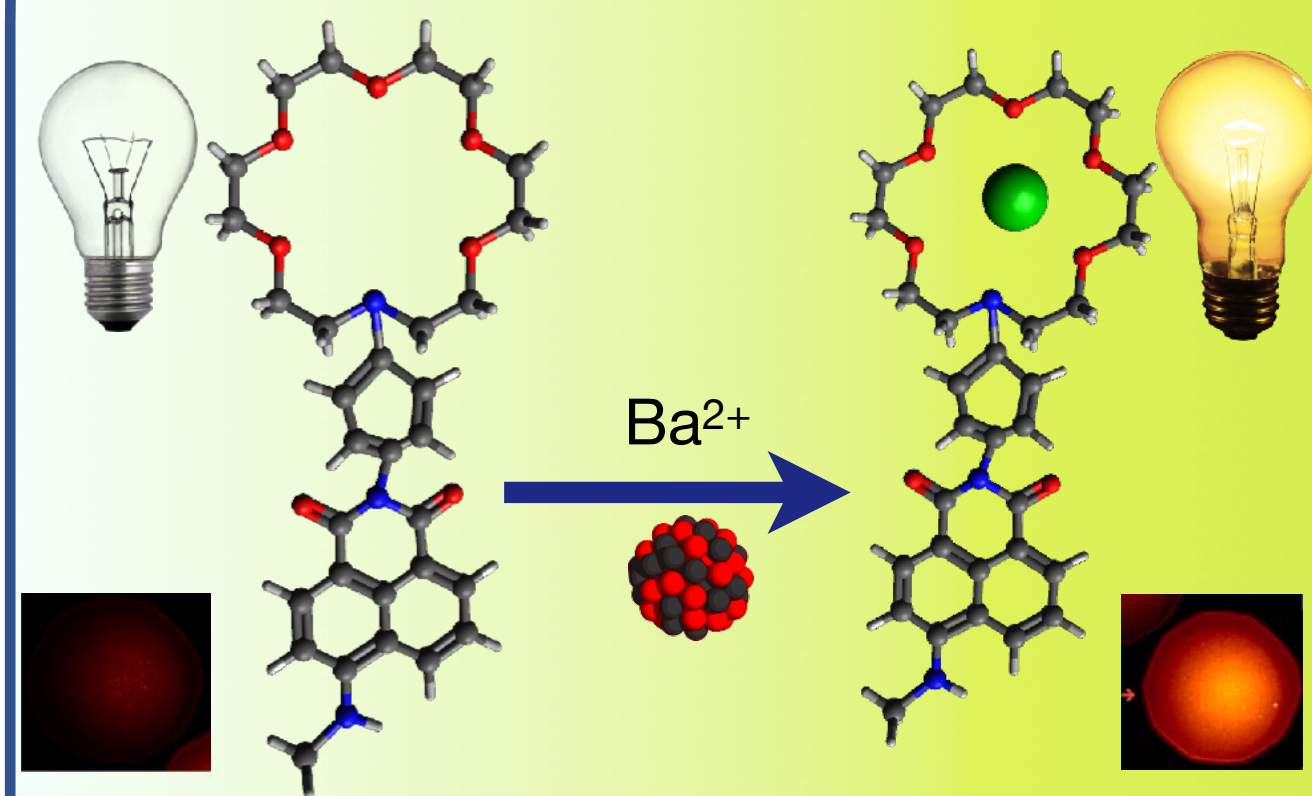
R&D into Chemosensors

The building blocks of the BaTag system

- Exploit three properties of light emission:



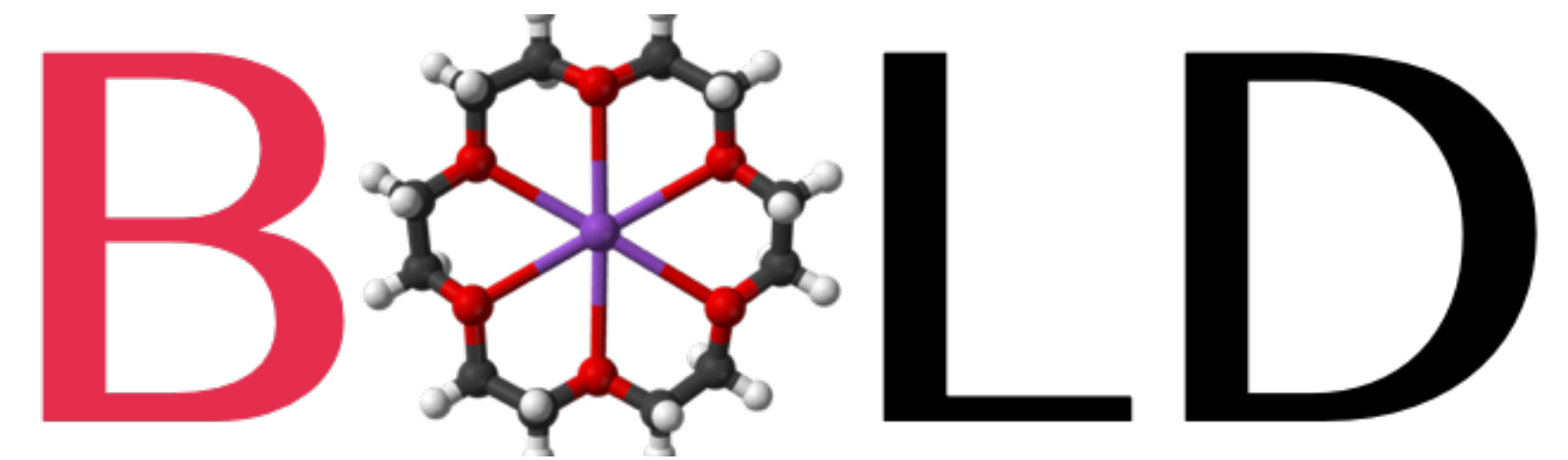
a) Intensity: Off-On Indicators



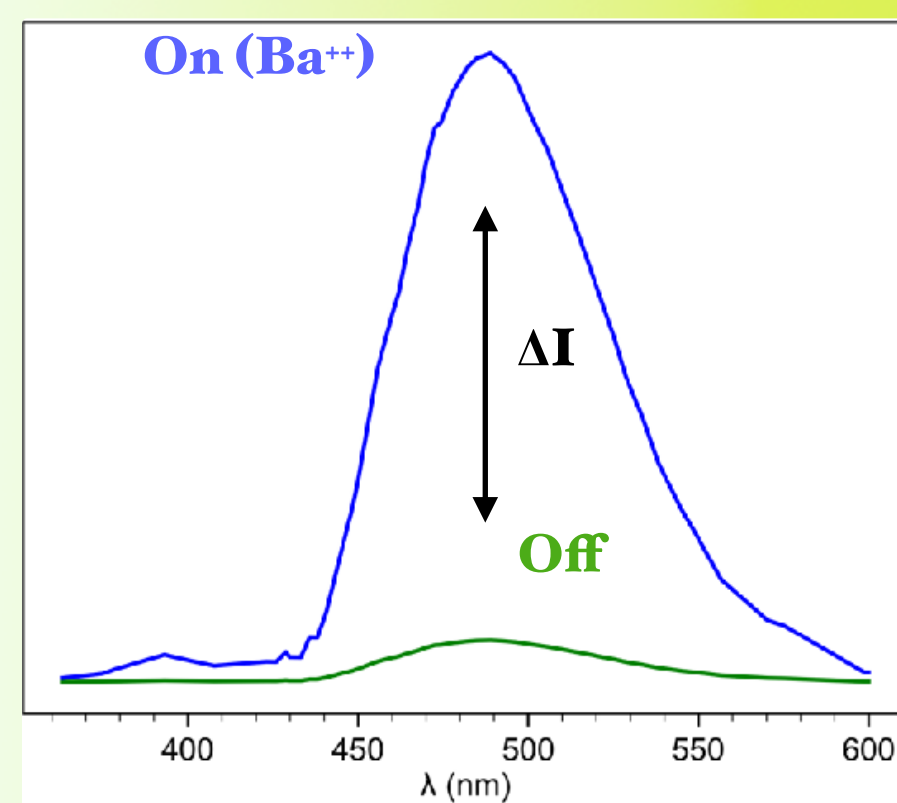
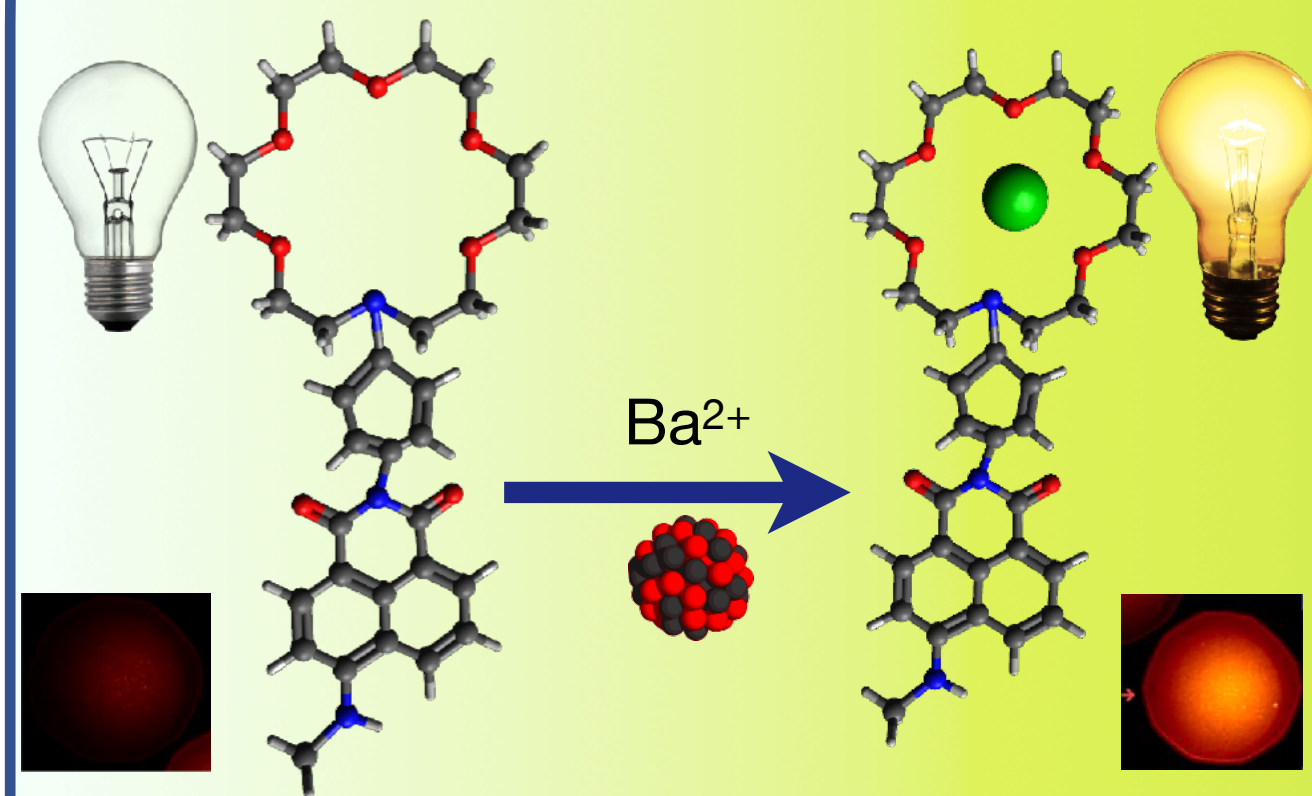
R&D into Chemosensors

The building blocks of the BaTag system

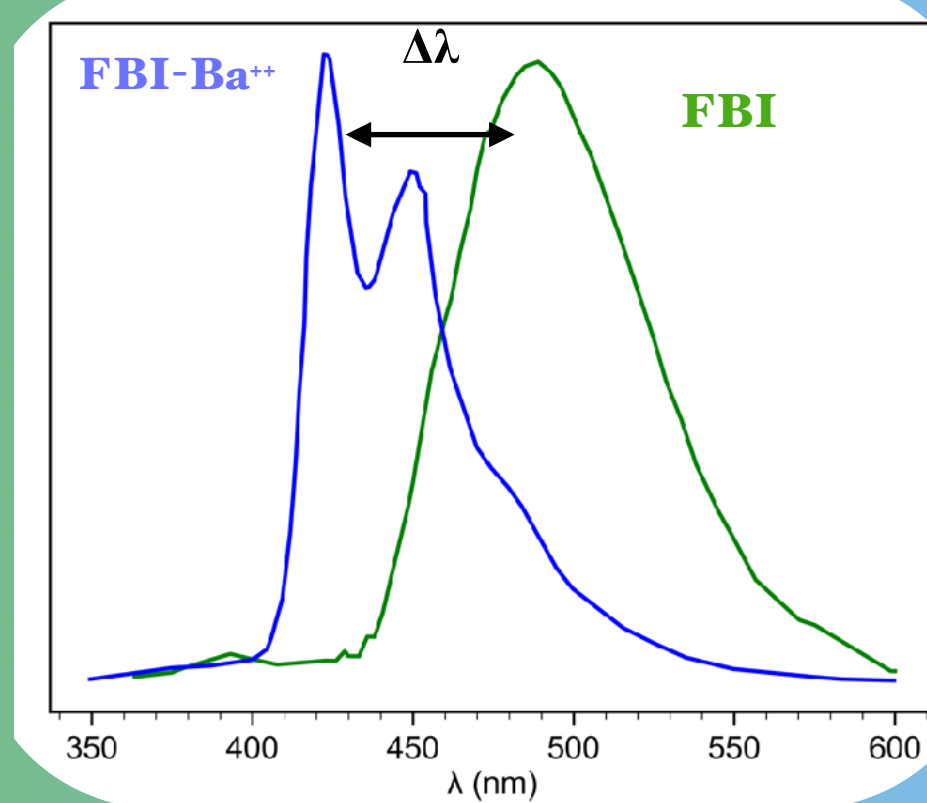
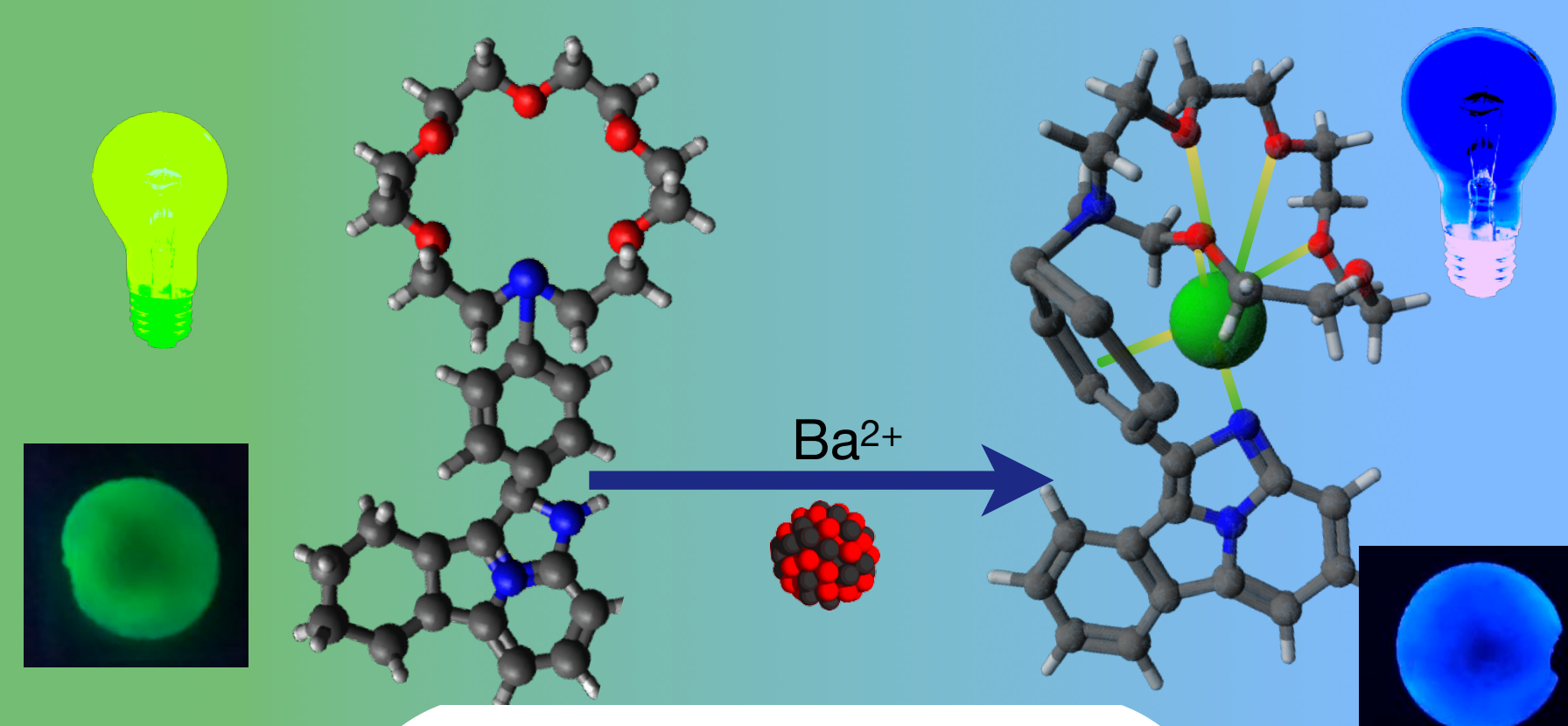
- Exploit three properties of light emission:



a) Intensity: Off-On Indicators



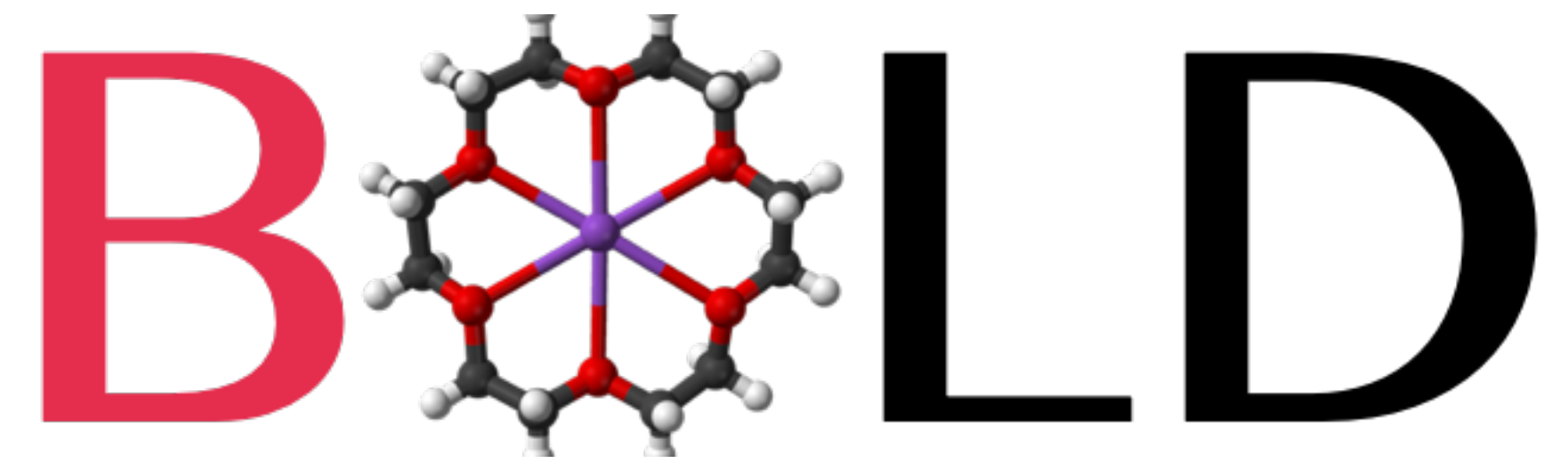
b) Wavelength: Bicolor Indicators (FBI)



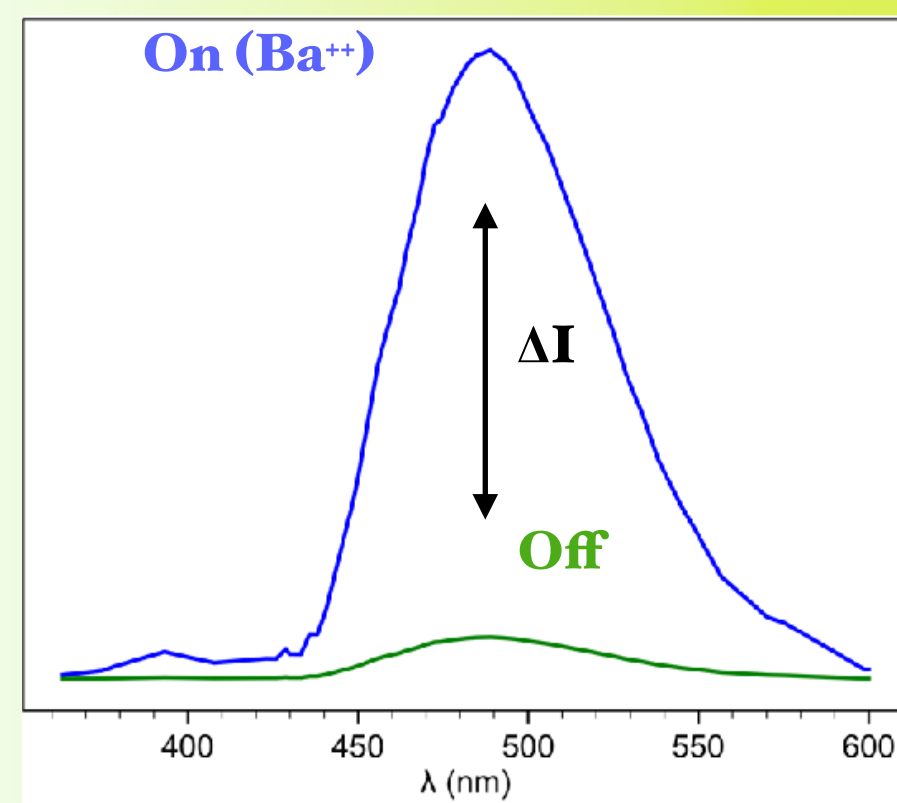
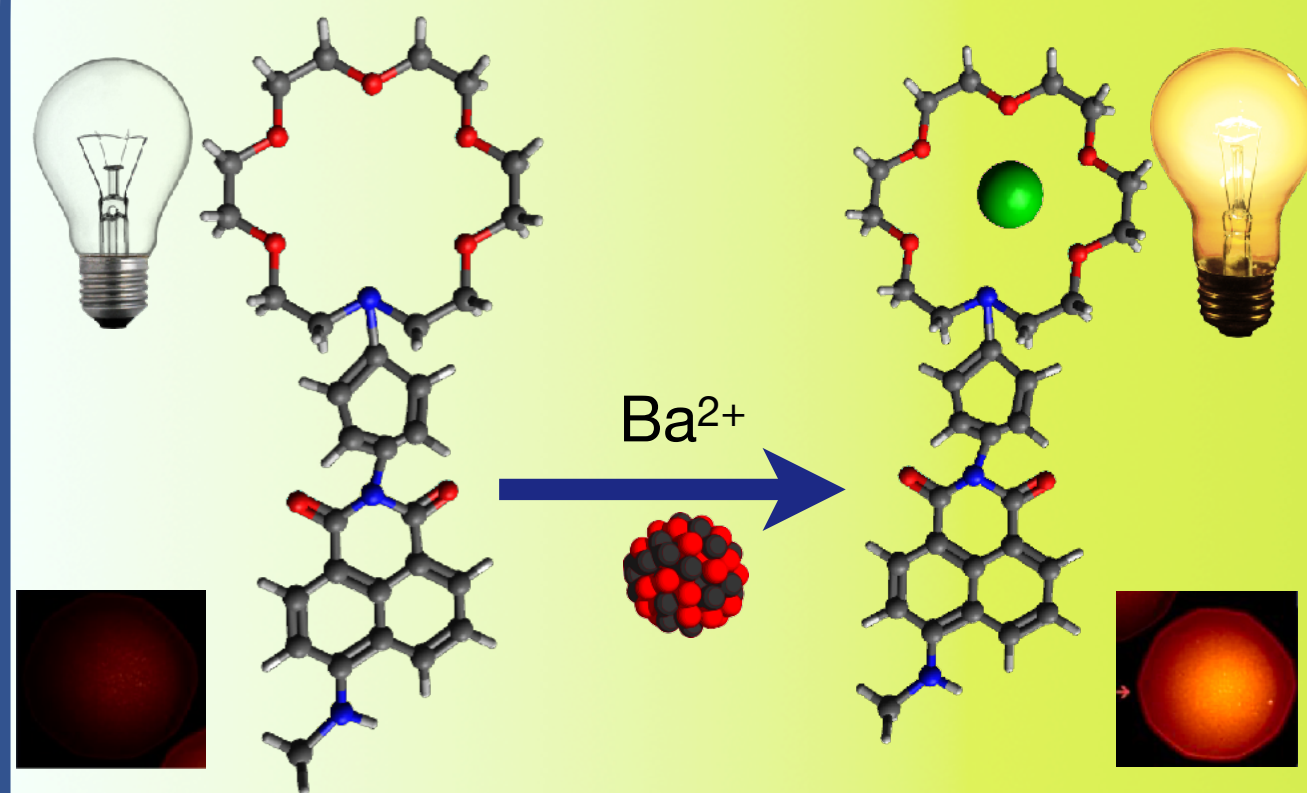
R&D into Chemosensors

The building blocks of the BaTag system

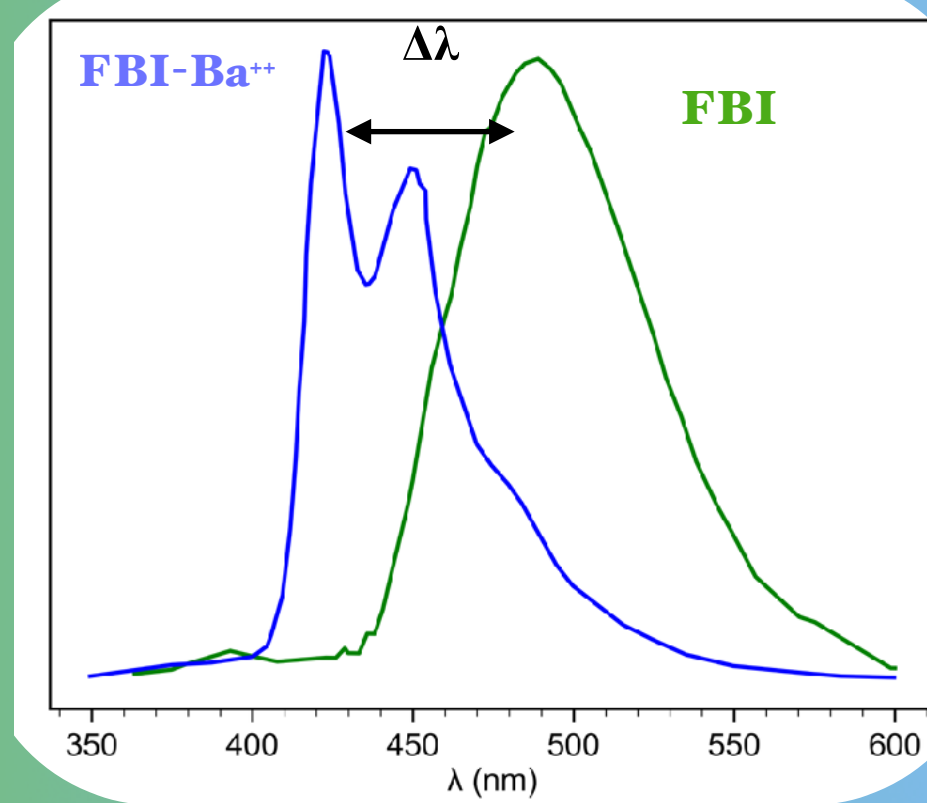
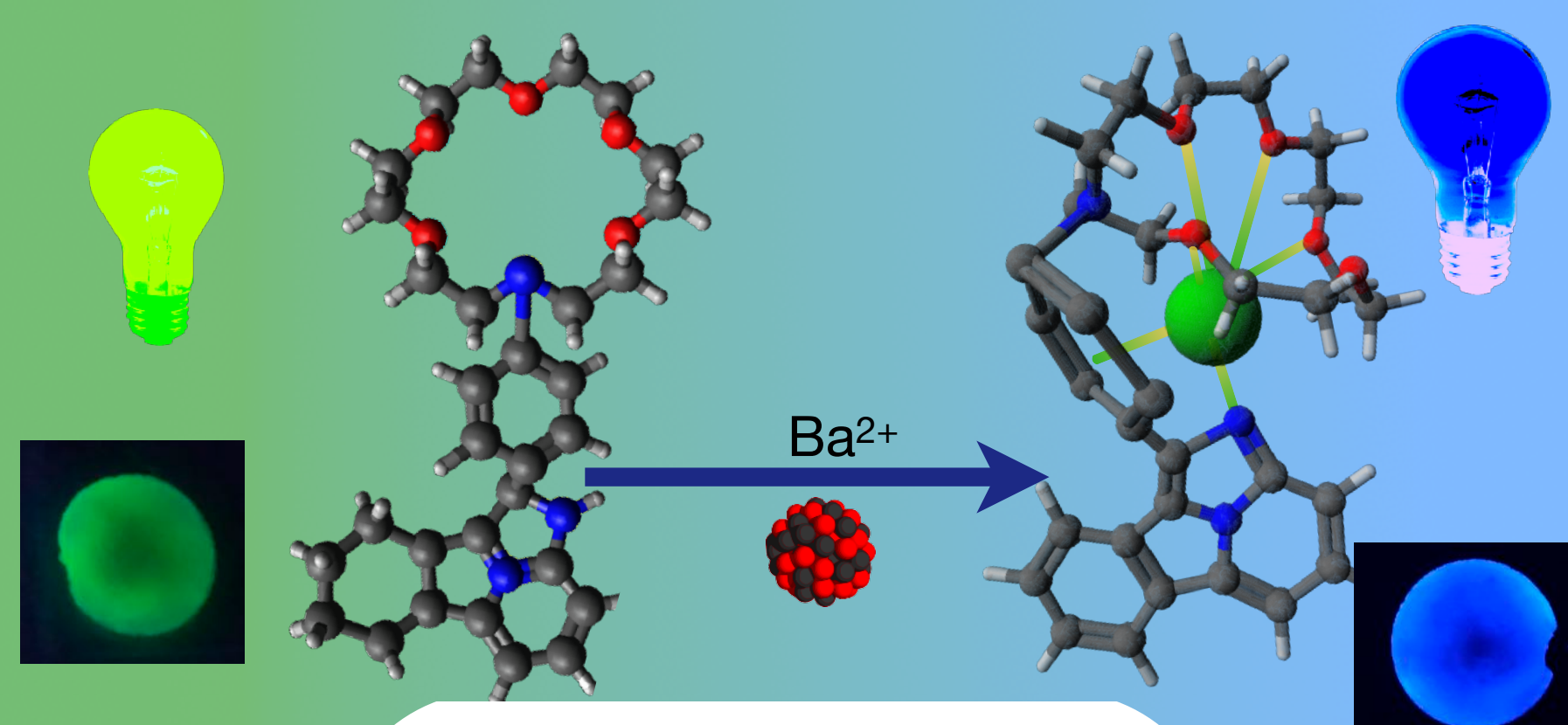
- Exploit three properties of light emission:



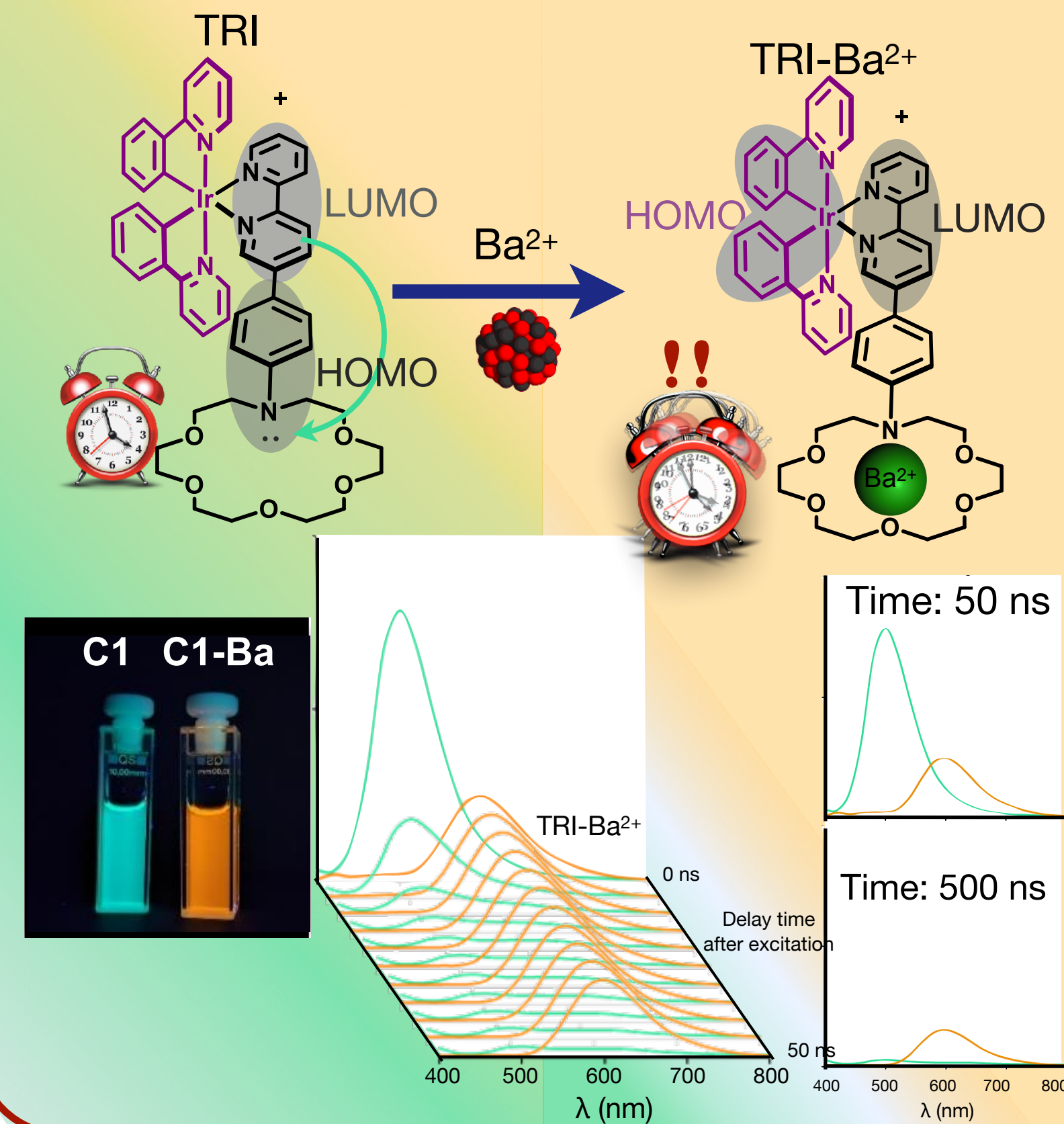
a) Intensity: Off-On Indicators



b) Wavelength: Bicolor Indicators (FBI)

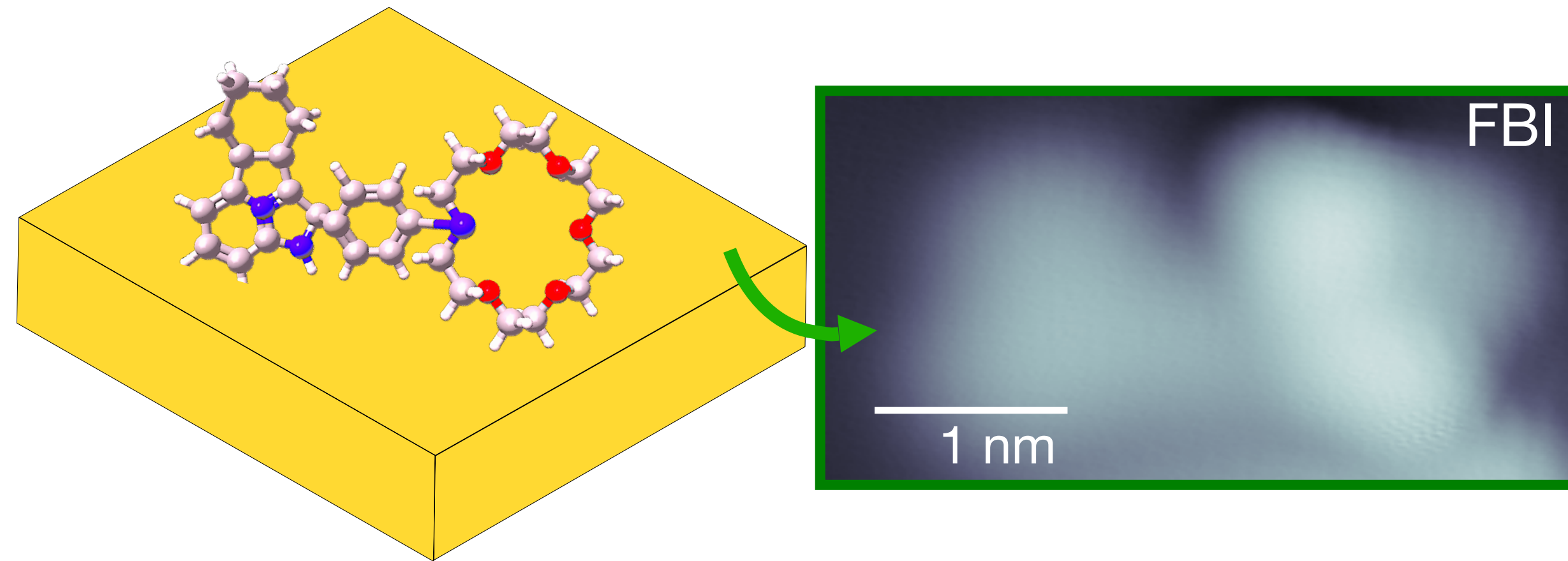


c) Relaxation time: Time-resolved Indicators (TRI)



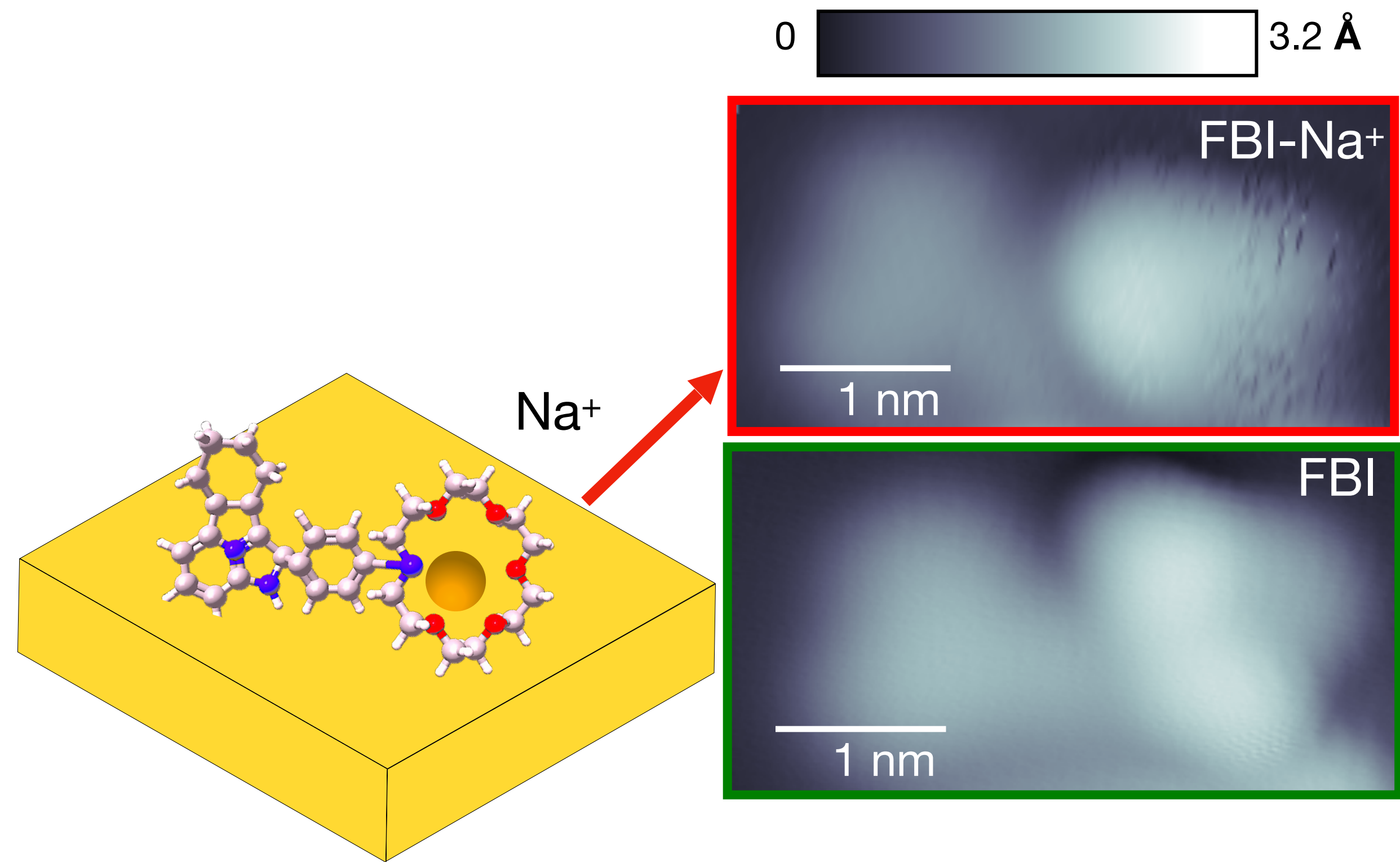
Molecule characterization

Capture demonstrated in UHV



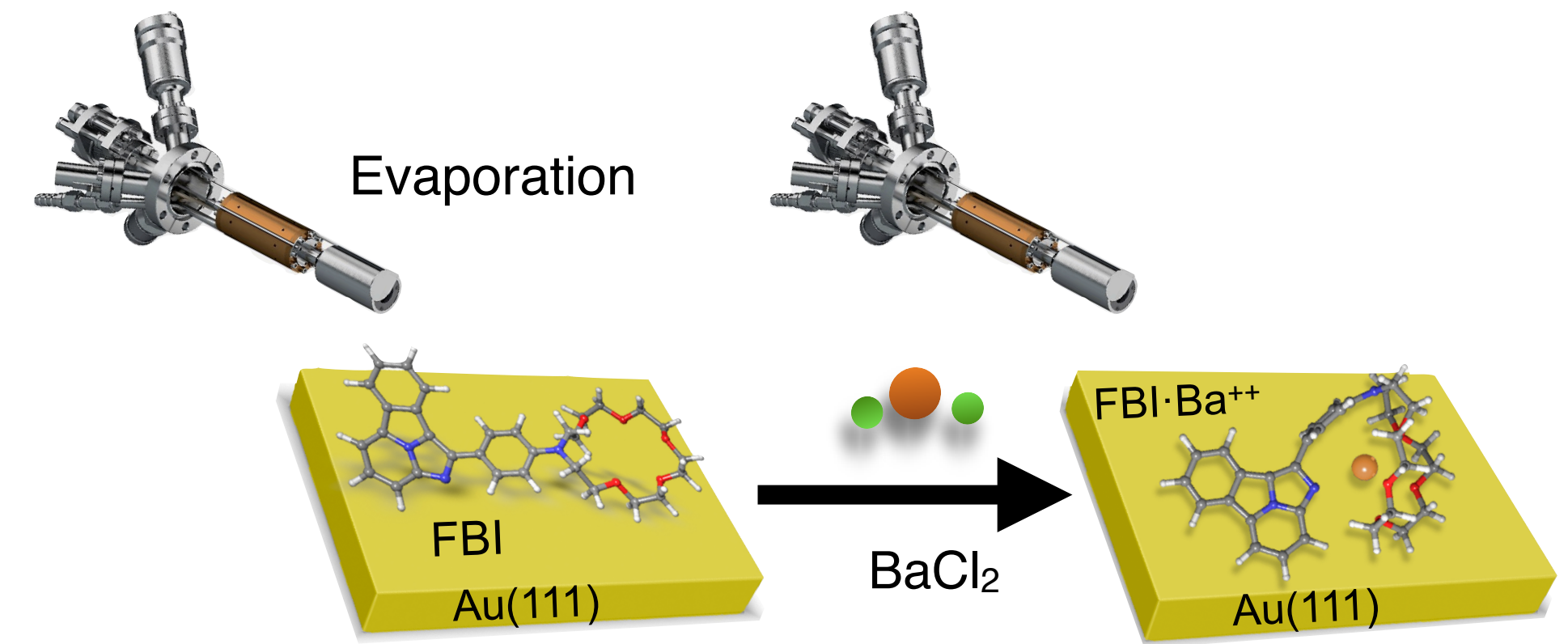
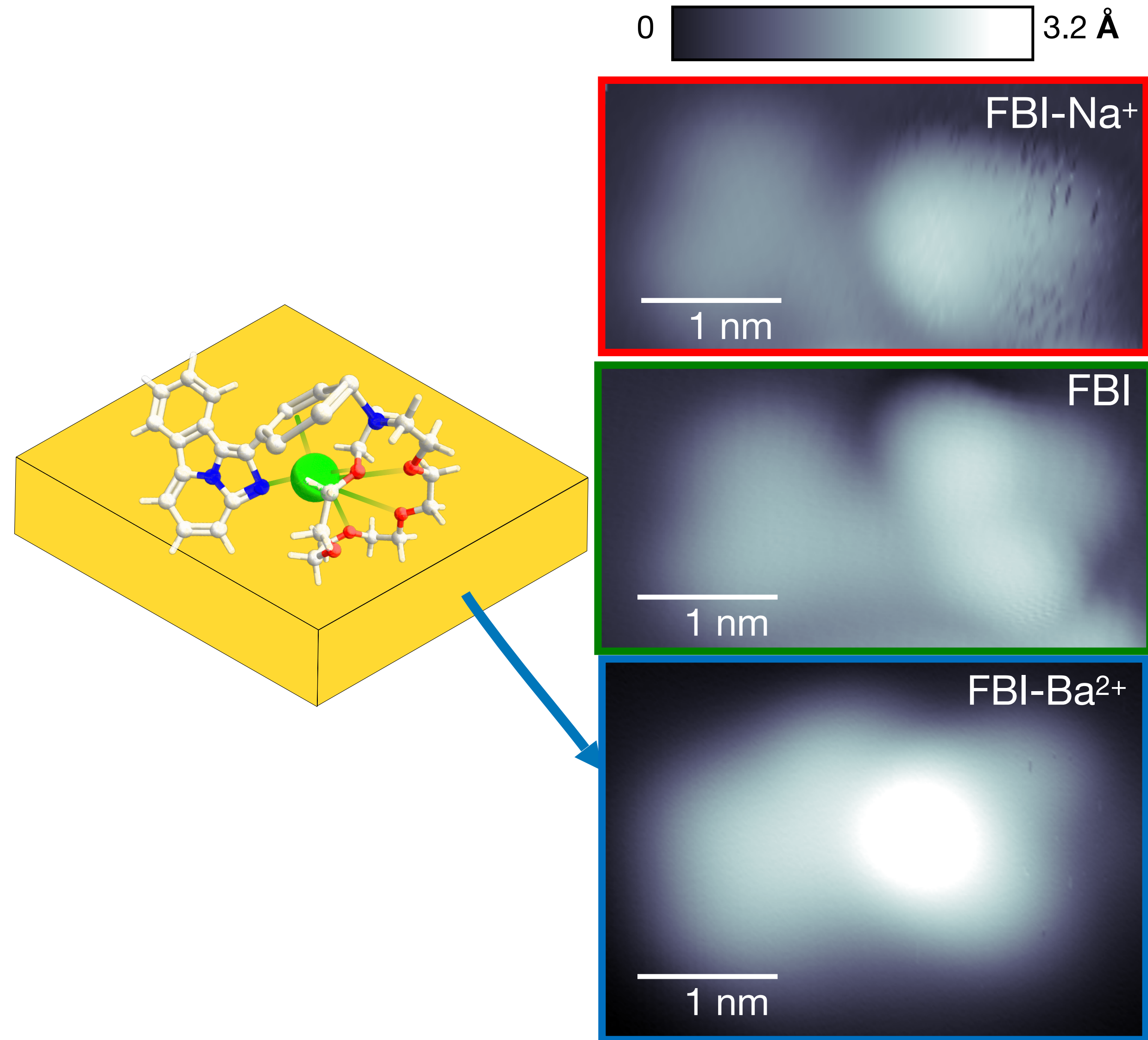
Molecule characterization

Capture demonstrated in UHV



Molecule characterization

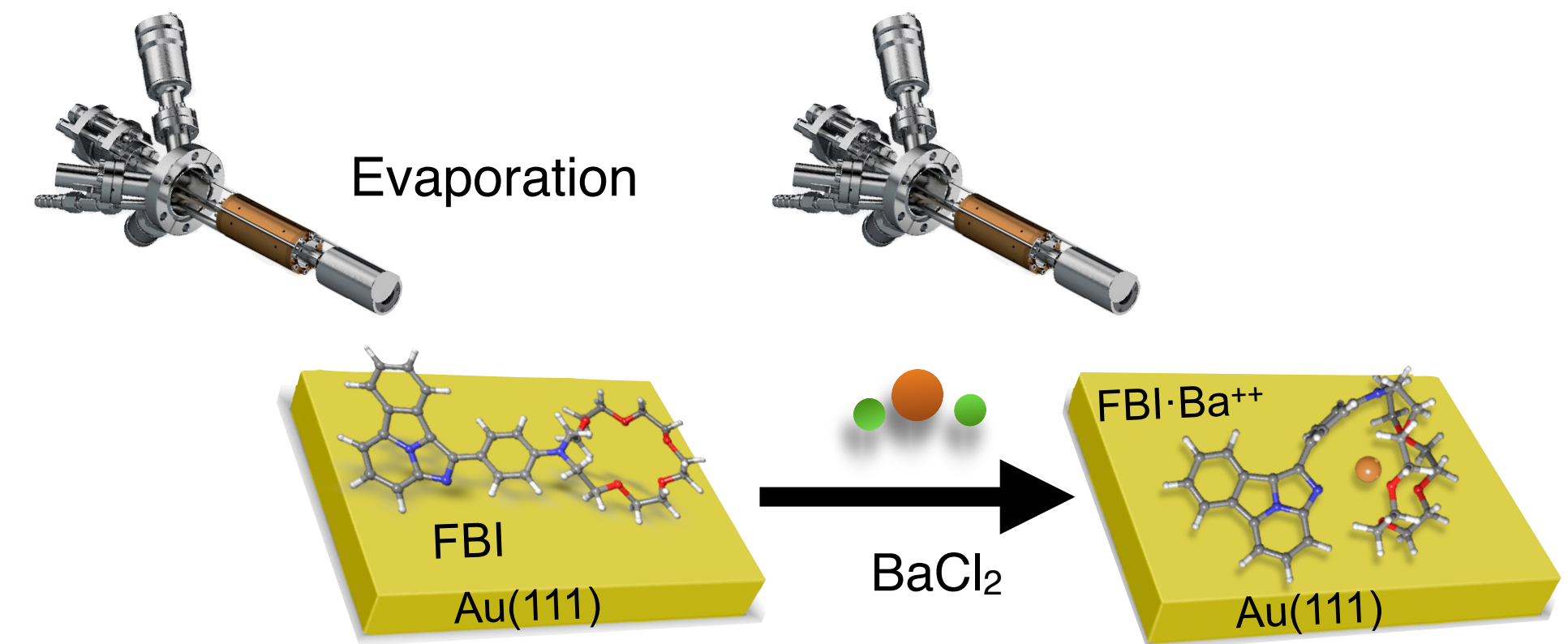
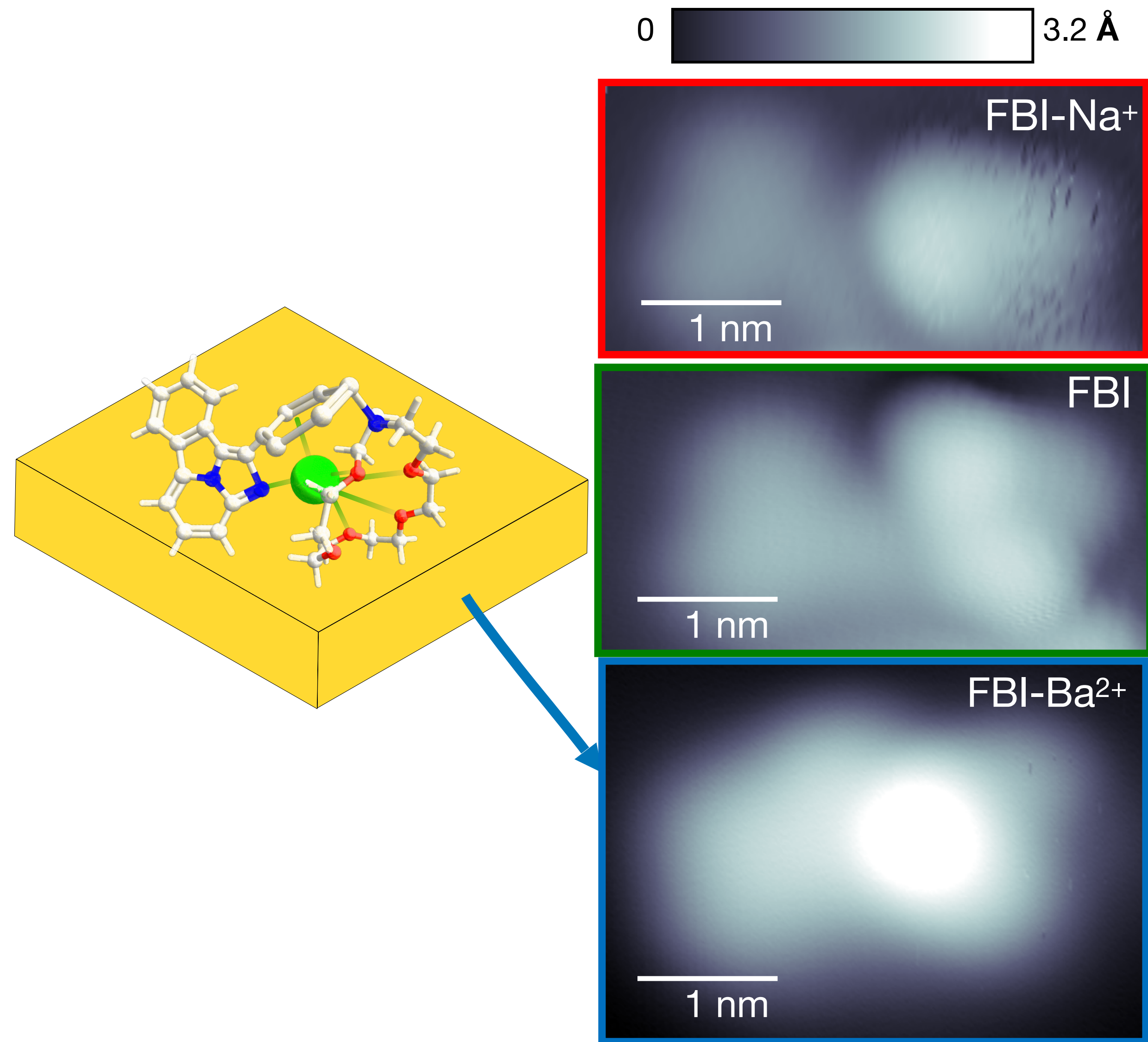
Capture demonstrated in UHV



- ✓ Structural changes (STM)
- ✓ Gap widening compatible with shift in absorption (STS)

Molecule characterization

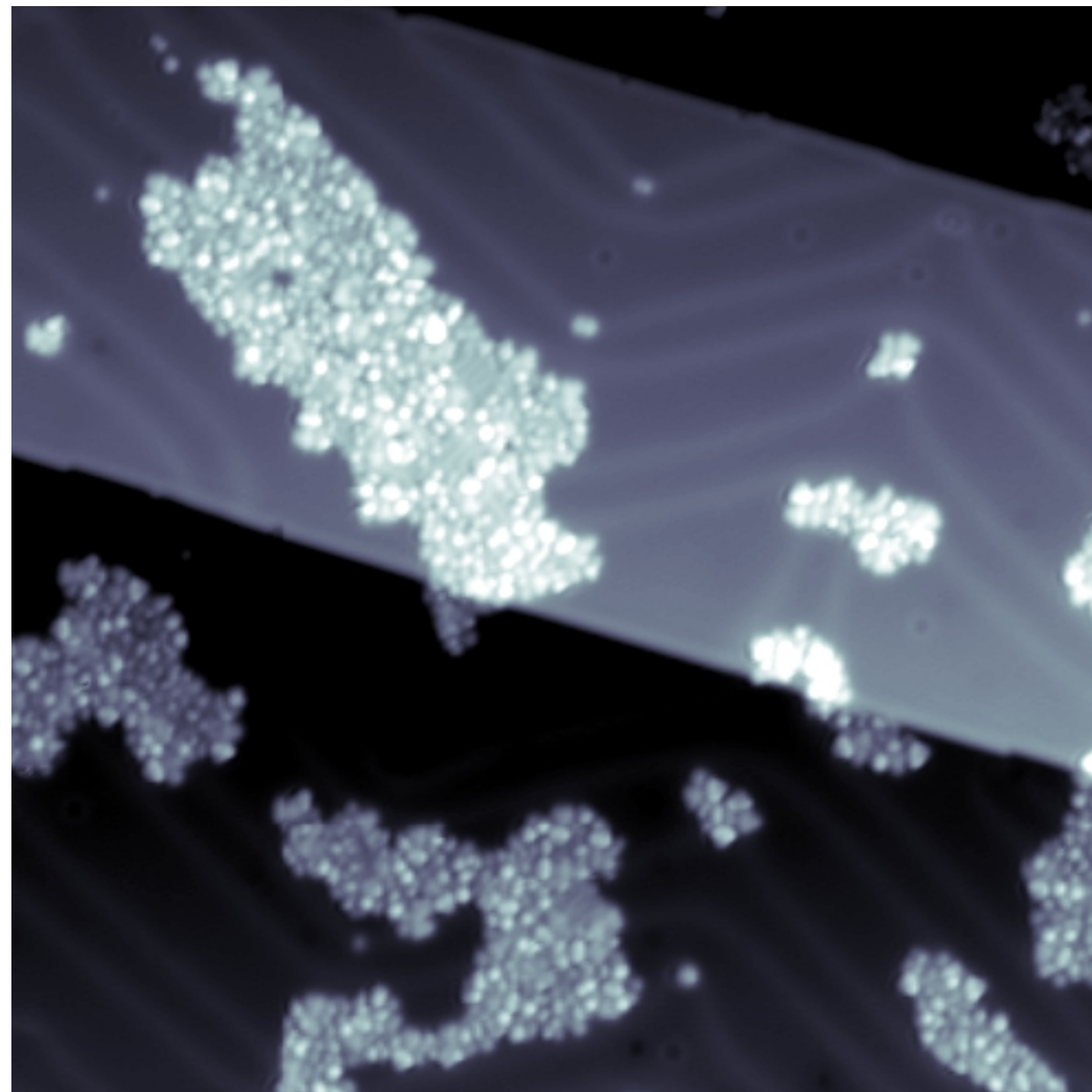
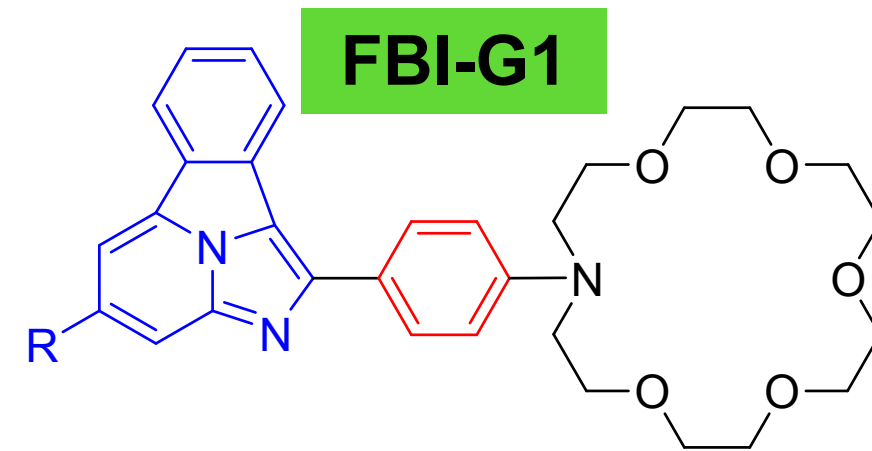
Capture demonstrated in UHV



- ✓ Structural changes (STM)
- ✓ Gap widening compatible with shift in absorption (STS)
- ✓ Different effect for Na⁺ (selectivity)
- ✓ Chemical interaction (XPS)
- ✓ Independent of substrate (Au, Cu, ITO, silica)

Arrangement of molecules on Au

Towards assembling the sensor

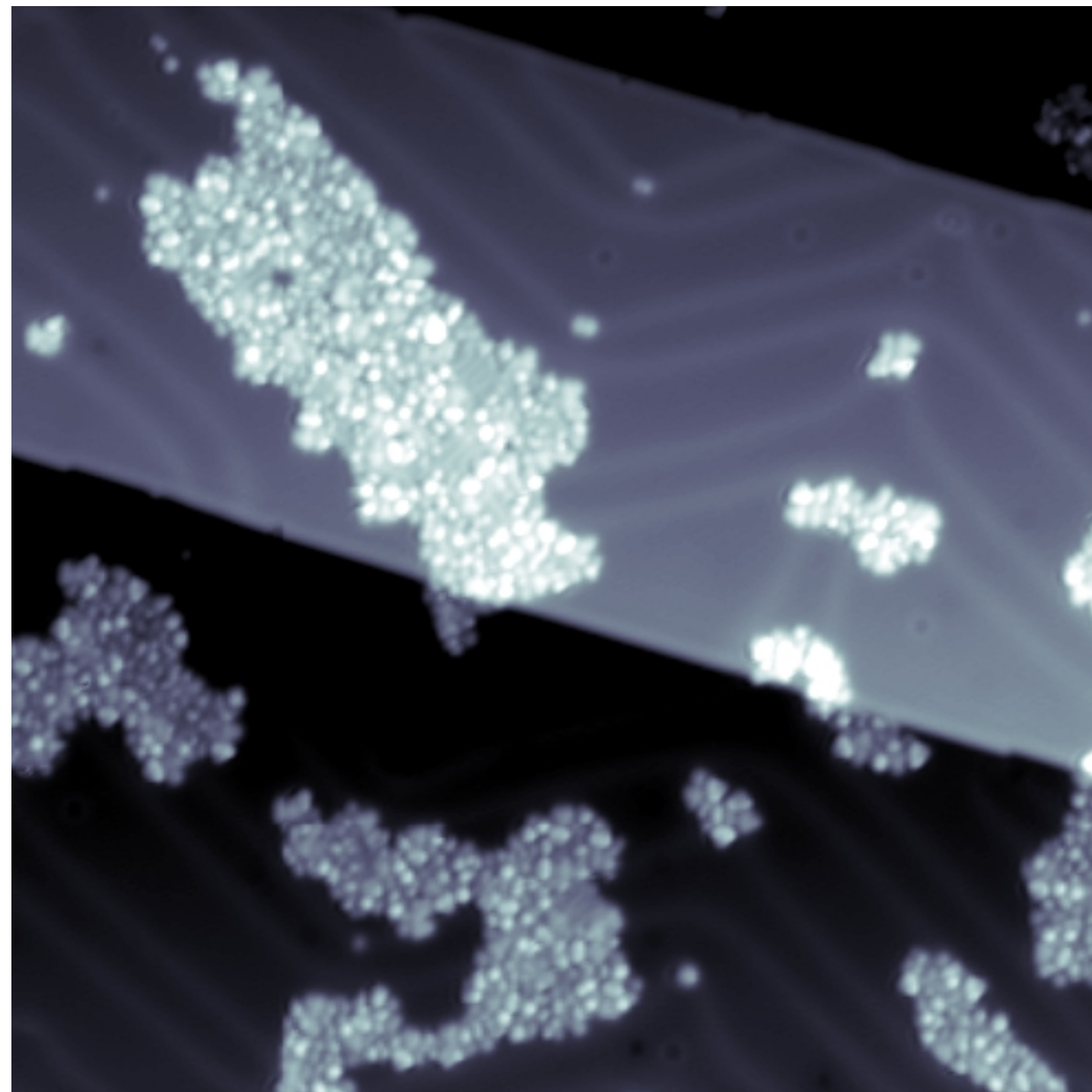
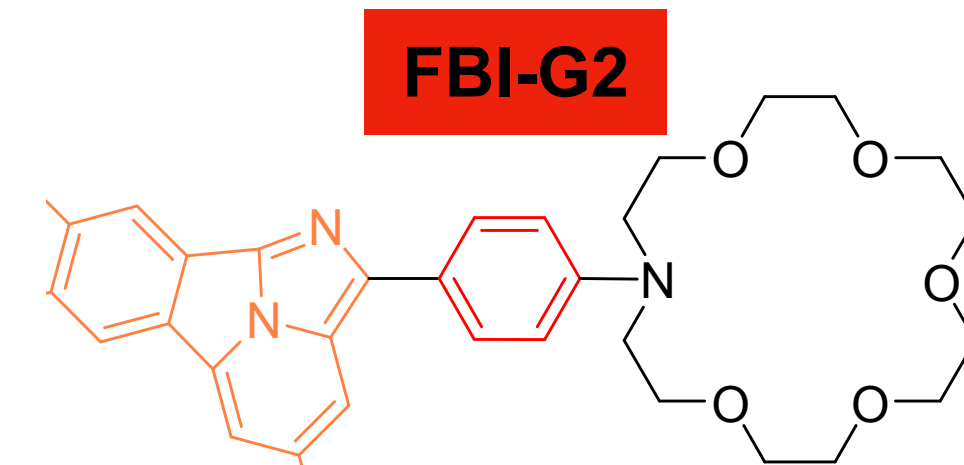
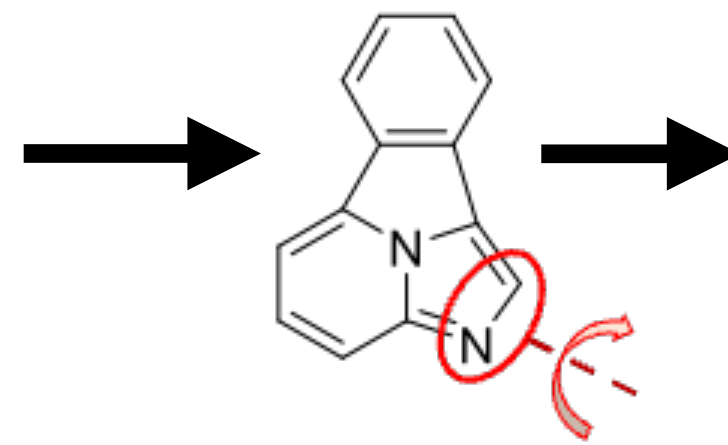
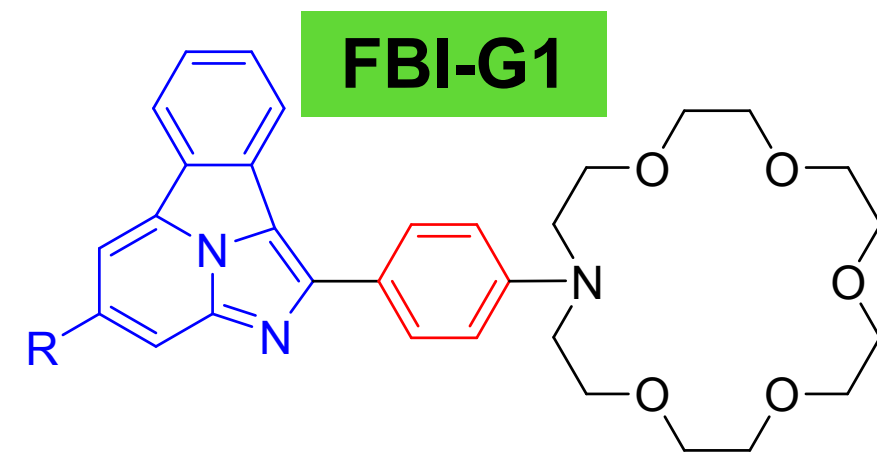


- Mainly islands

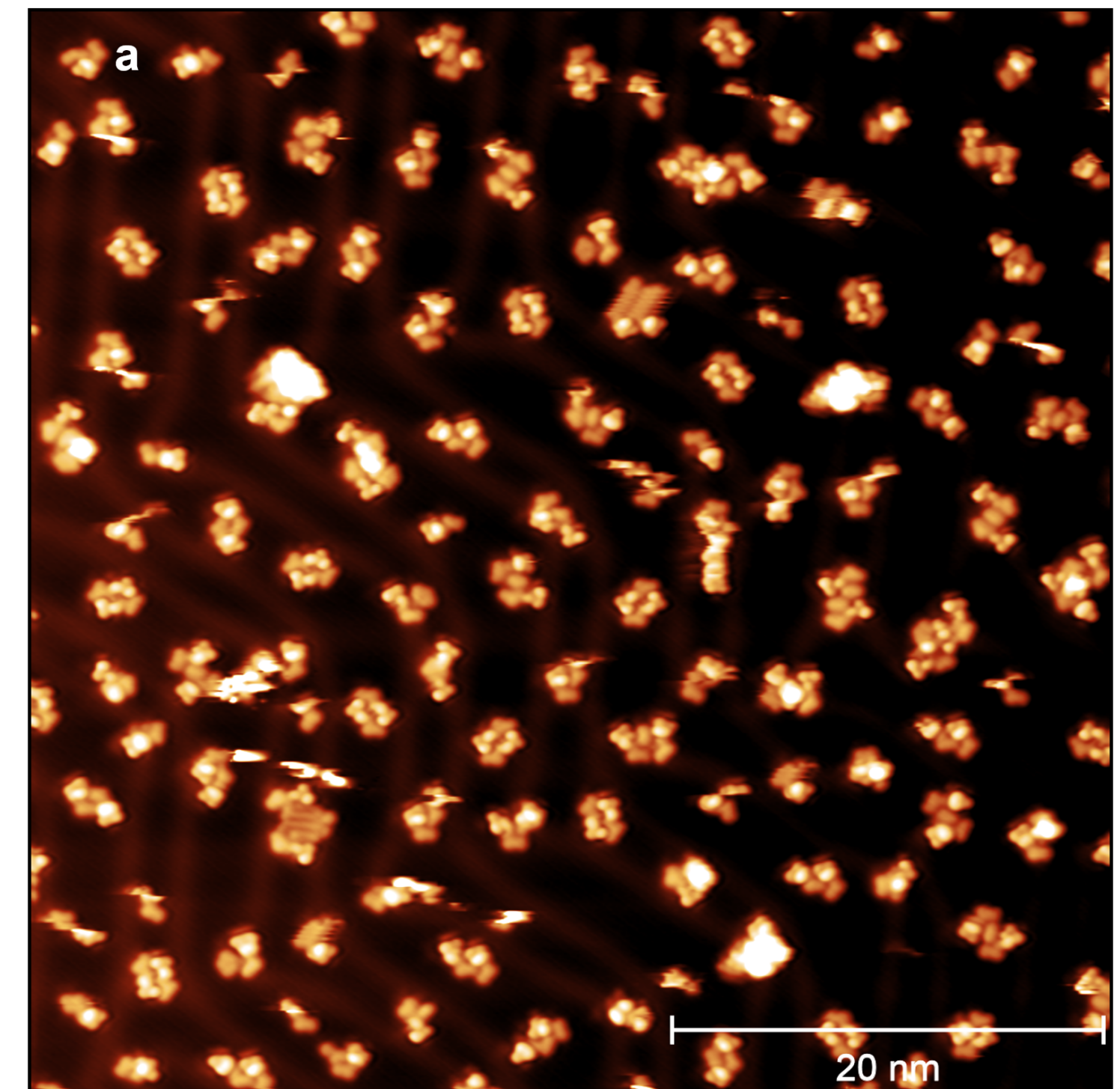
Arrangement of molecules on Au

Towards assembling the sensor

- A single C-N switch changes the arrangement completely



- Mainly islands

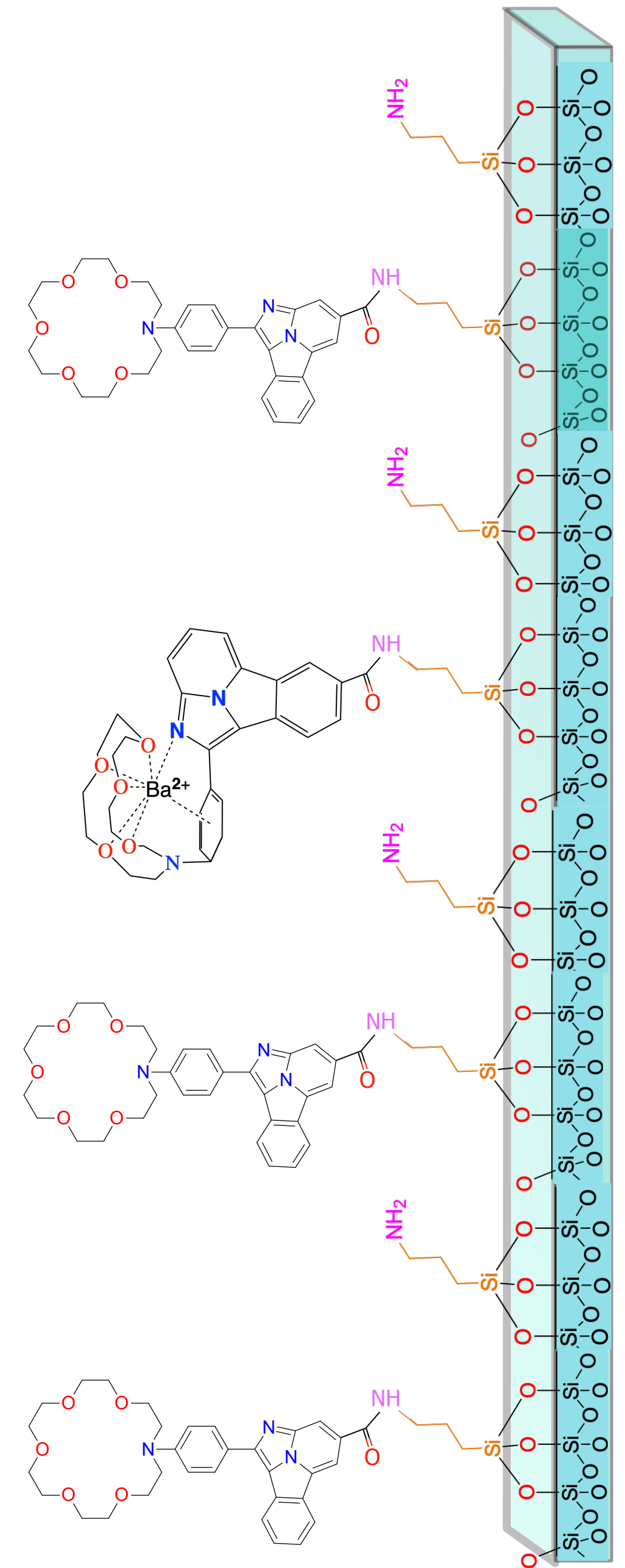


- Mainly dimers

Monolayer formation

Sensor structure in the nano-microscale

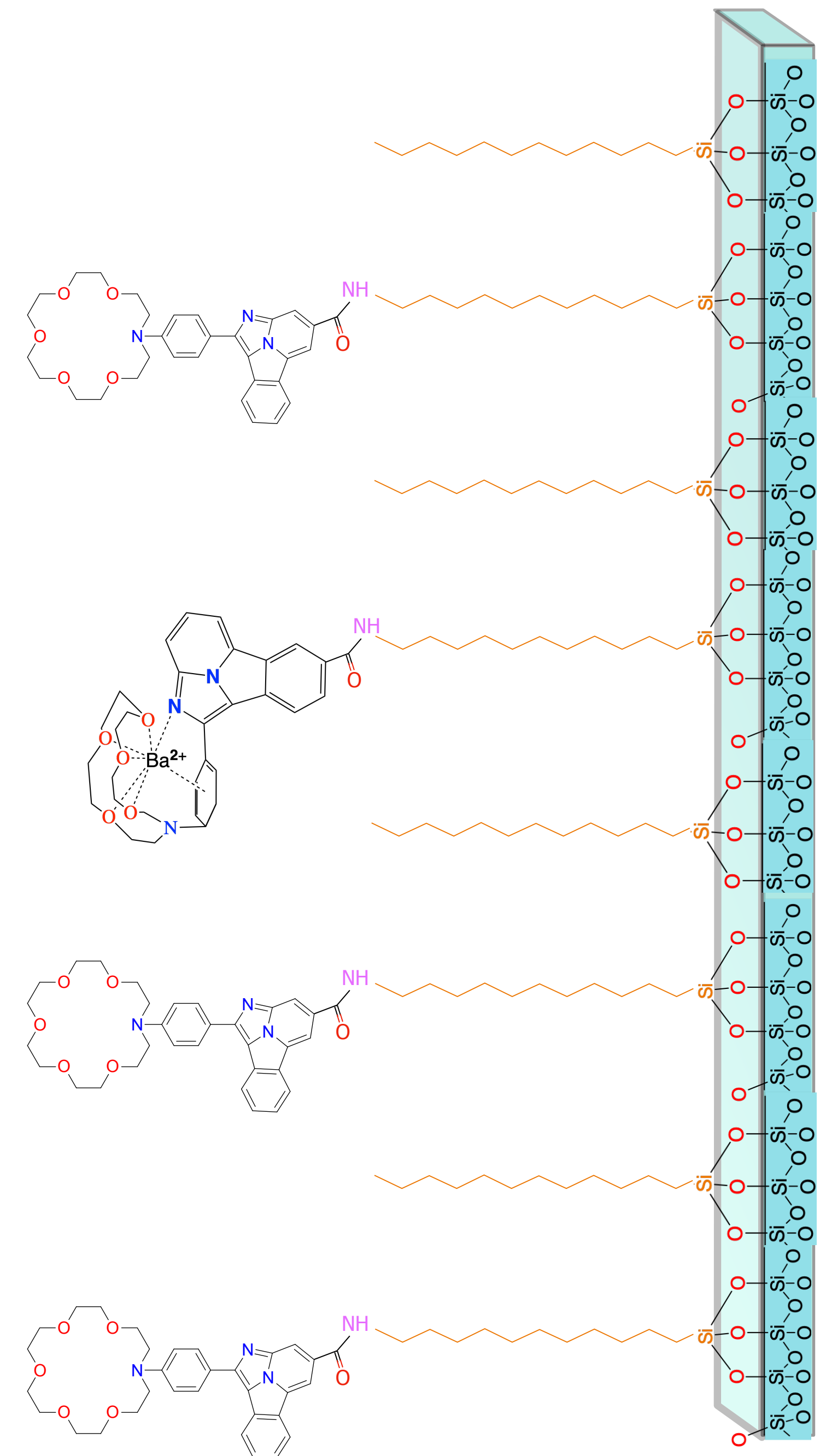
- Chemical bond between molecule and sensor
- Stable location of molecule improves single-molecule detection



Monolayer formation

Sensor structure in the nano-microscale

- Chemical bond between molecule and sensor
- Stable location of molecule improves single-molecule detection
- Chain length affects interaction with substrate and sensor efficiency



Overview

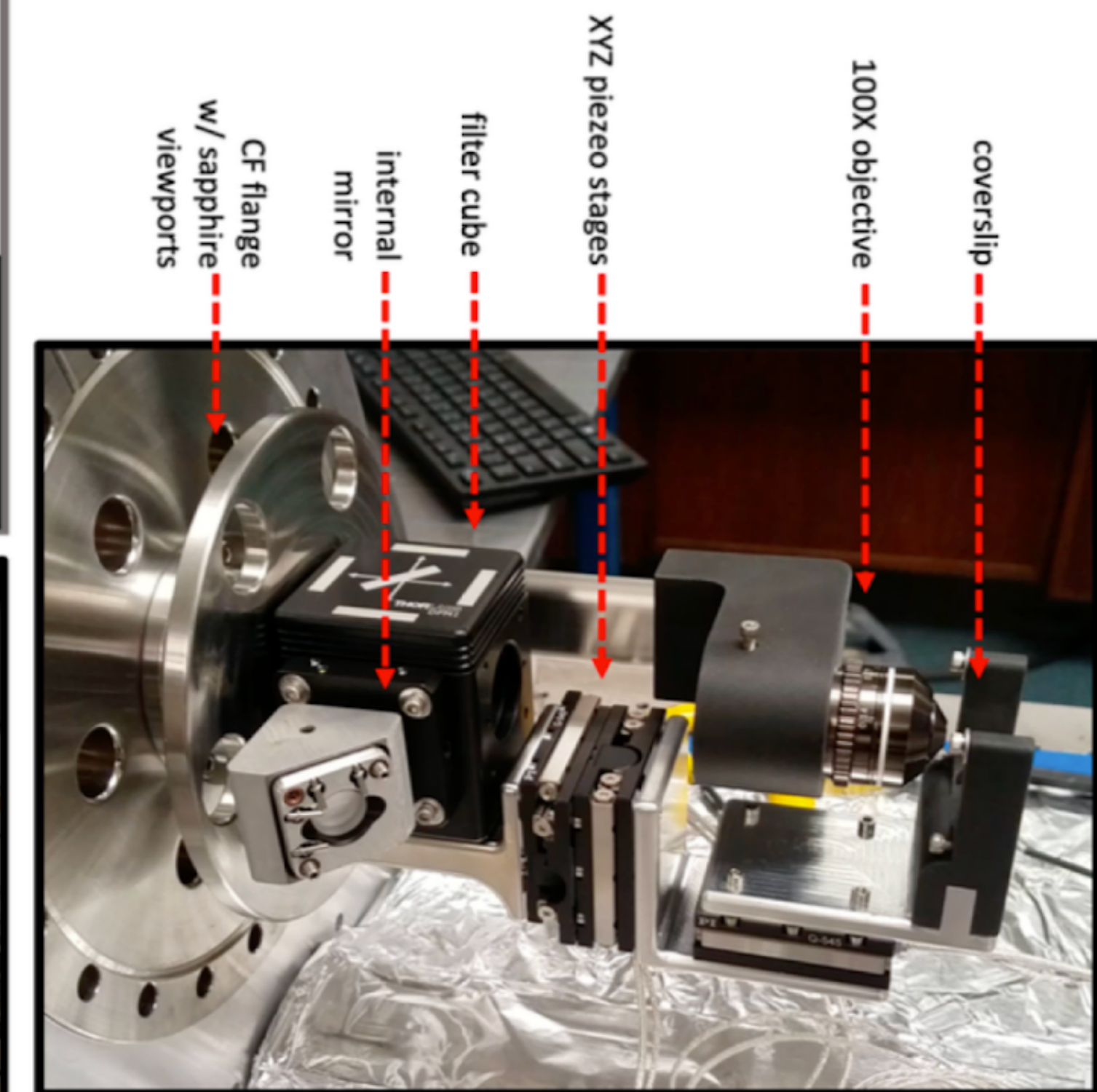
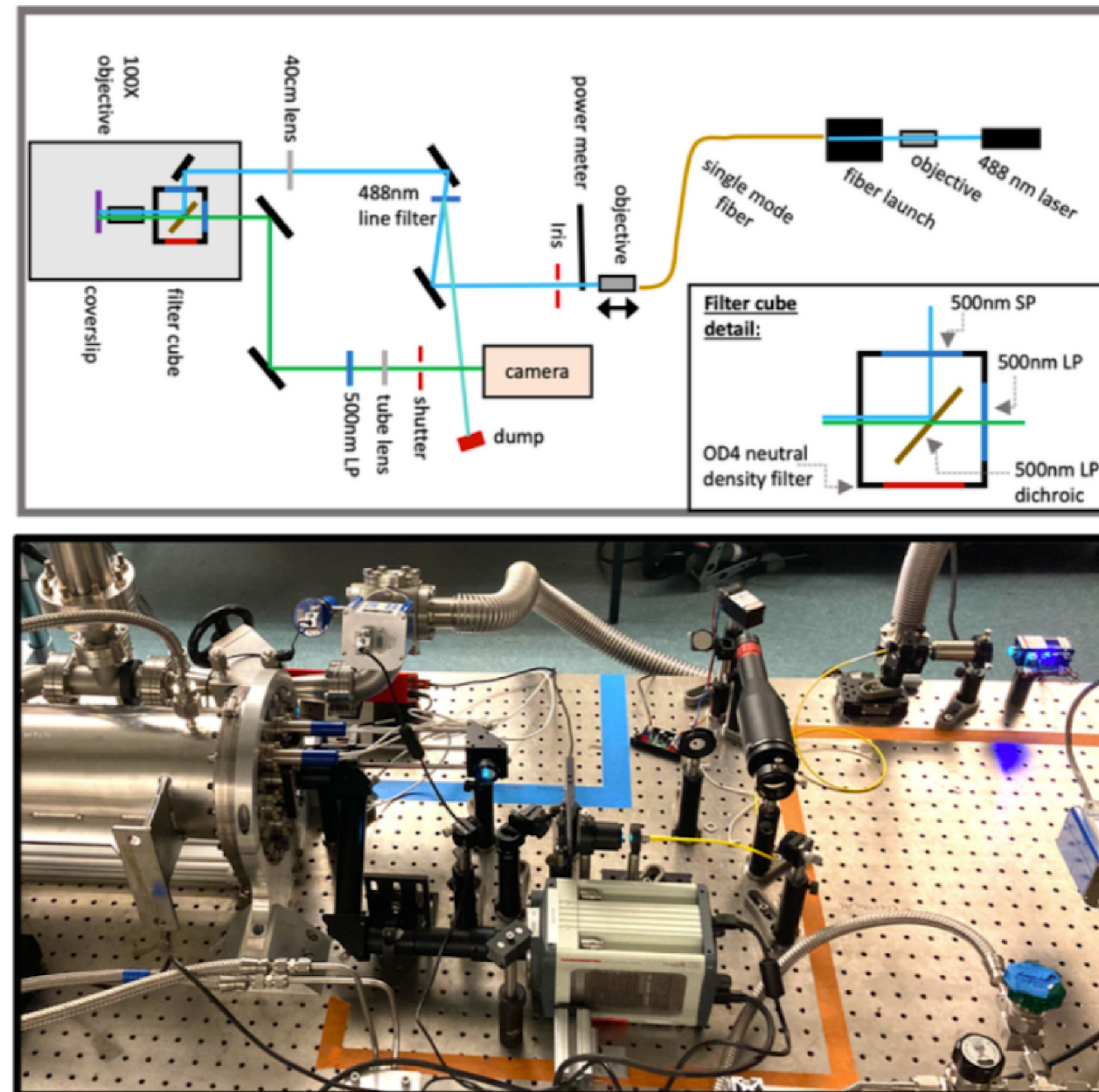
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- ☐ Ba-Tagging in nEXO
- ☐ The road forward

Microscopy efforts

[a] Nat. Commun. 15, 10595 (2024).

SMFI in High-pressure Xe

- Observation of single-molecule events in HPGXe
- Microscope Objective (MO) inside pressure chamber

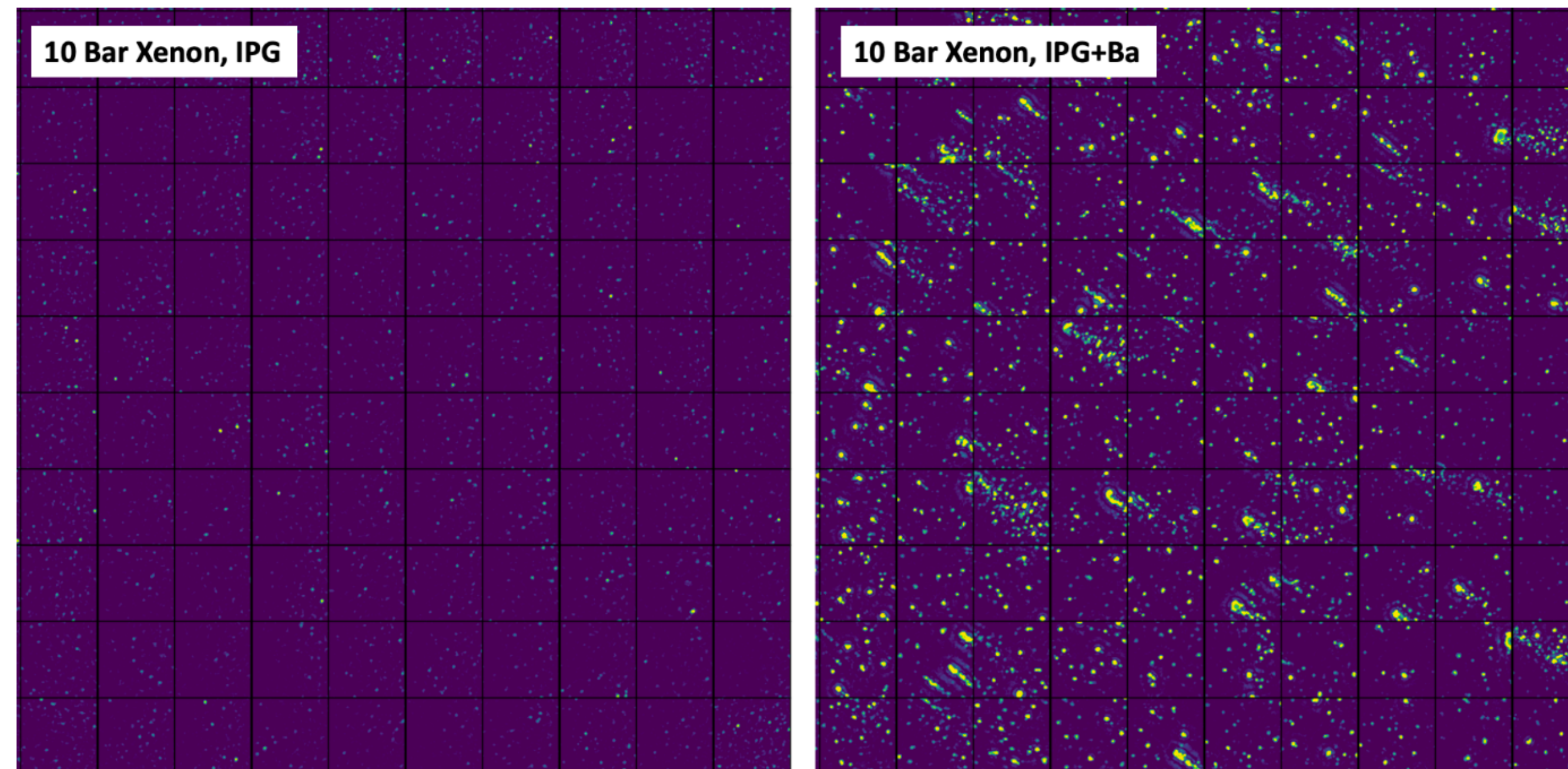


Microscopy efforts

[a] Nat. Commun. 15, 10595 (2024).

SMFI in High-pressure Xe

- Raster scans for large-scale characterization of sample fluorescence

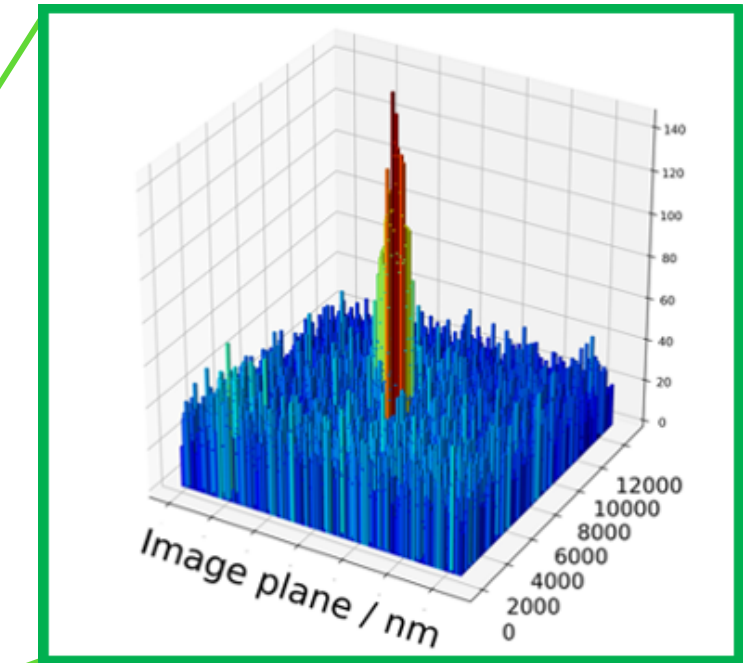
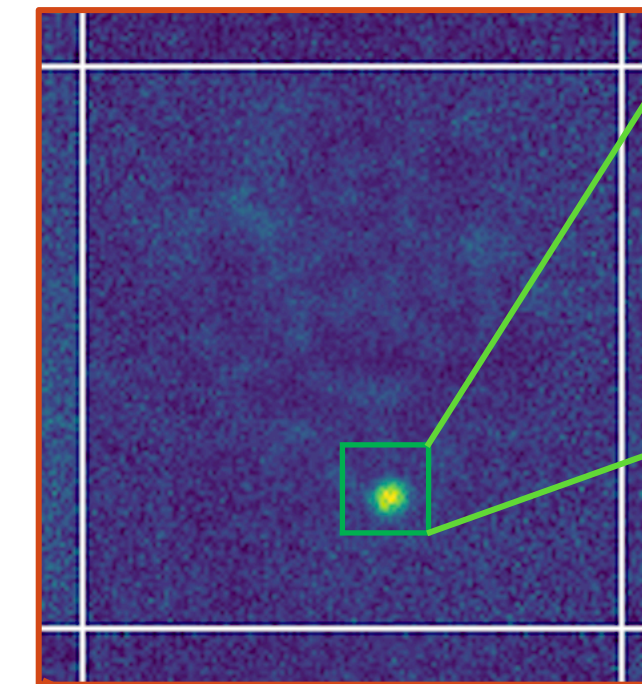
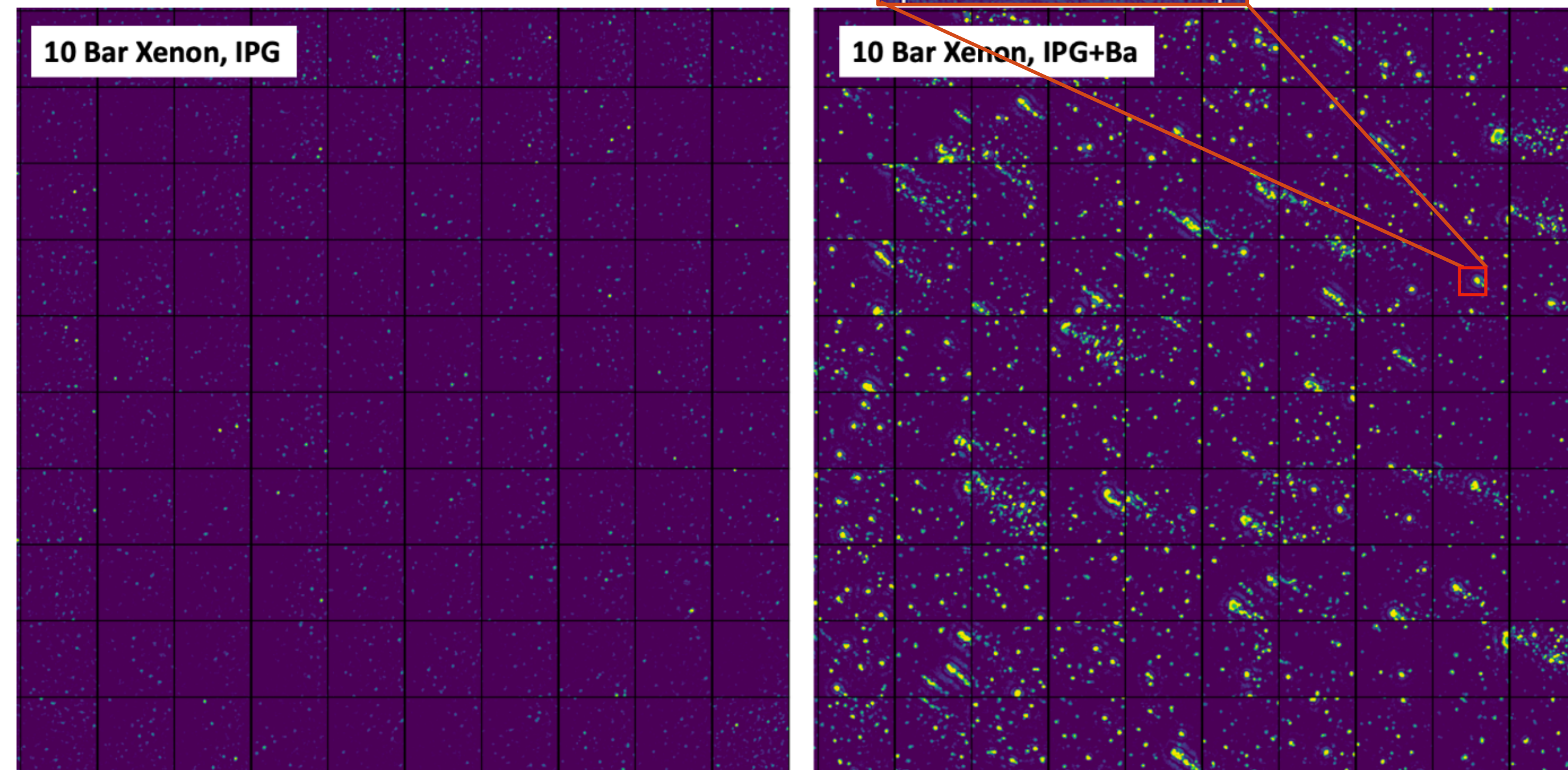


Microscopy efforts

SMFI in High-pressure Xe

- Raster scans for large-scale characterization of sample fluorescence

[a] Nat. Commun. 15, 10595 (2024).

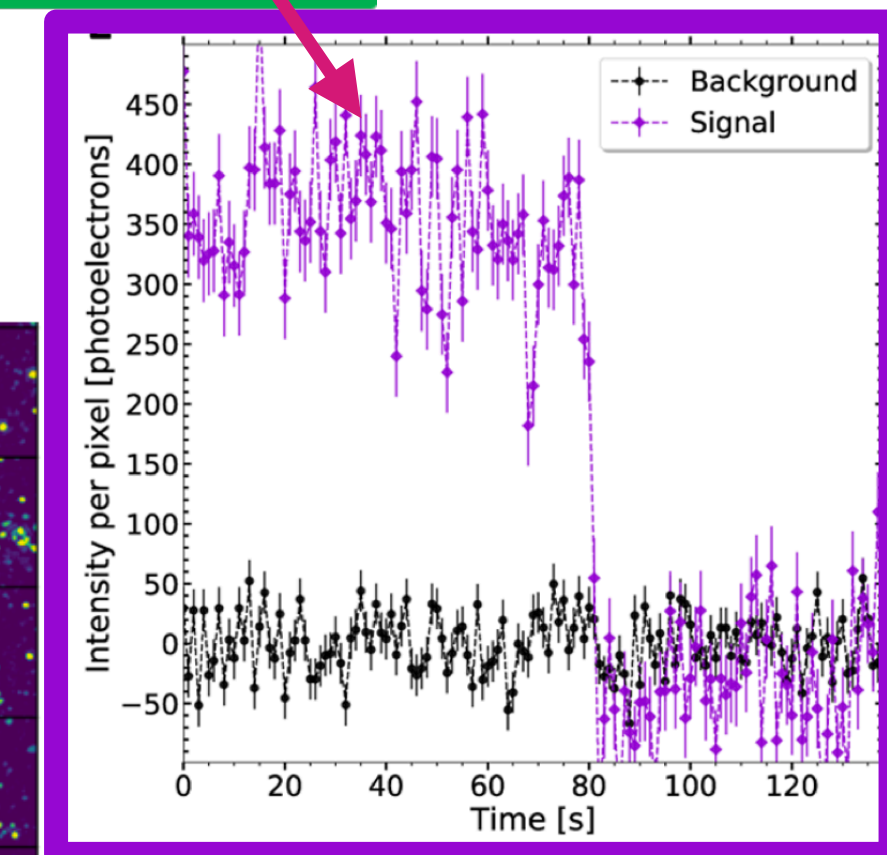
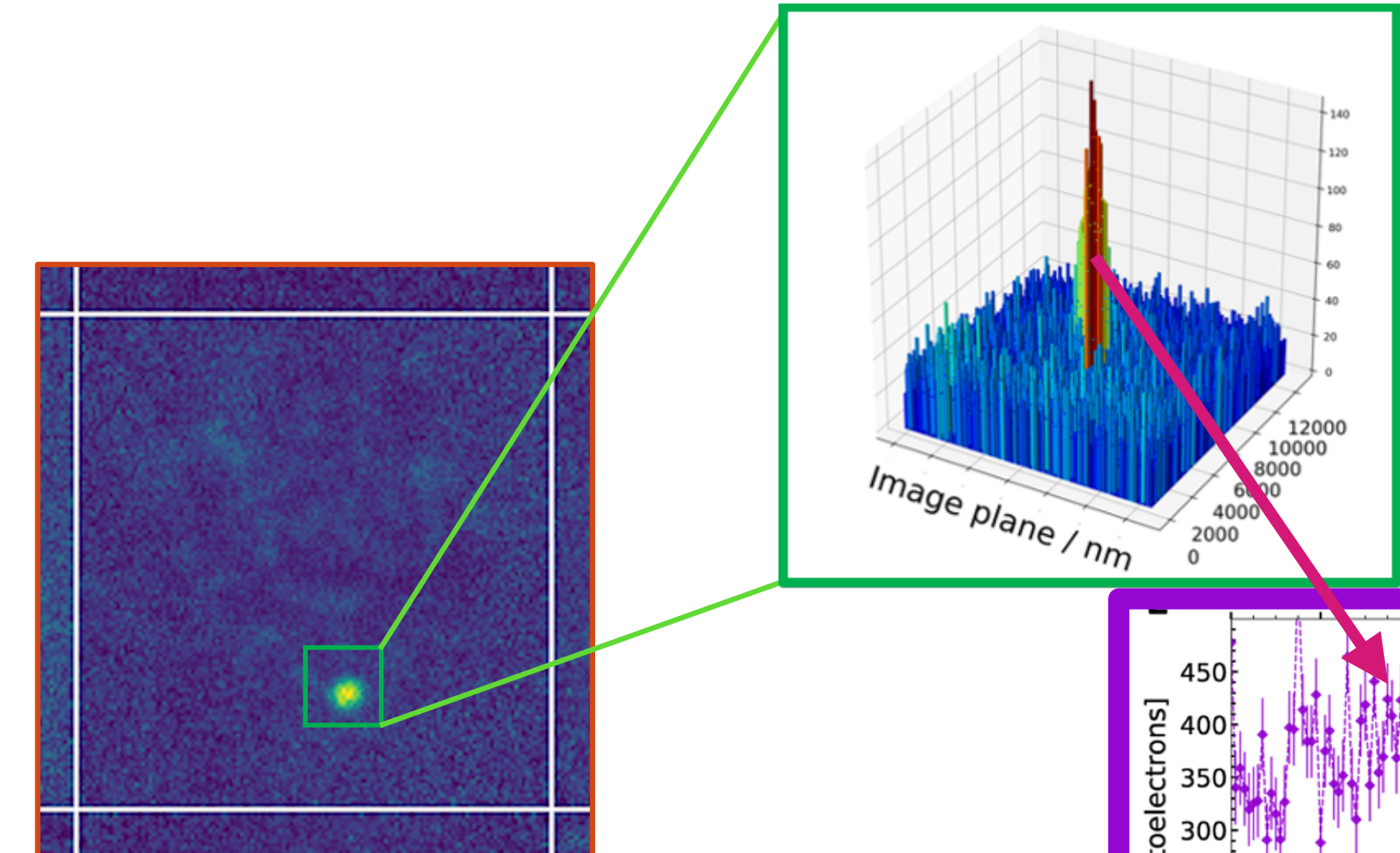
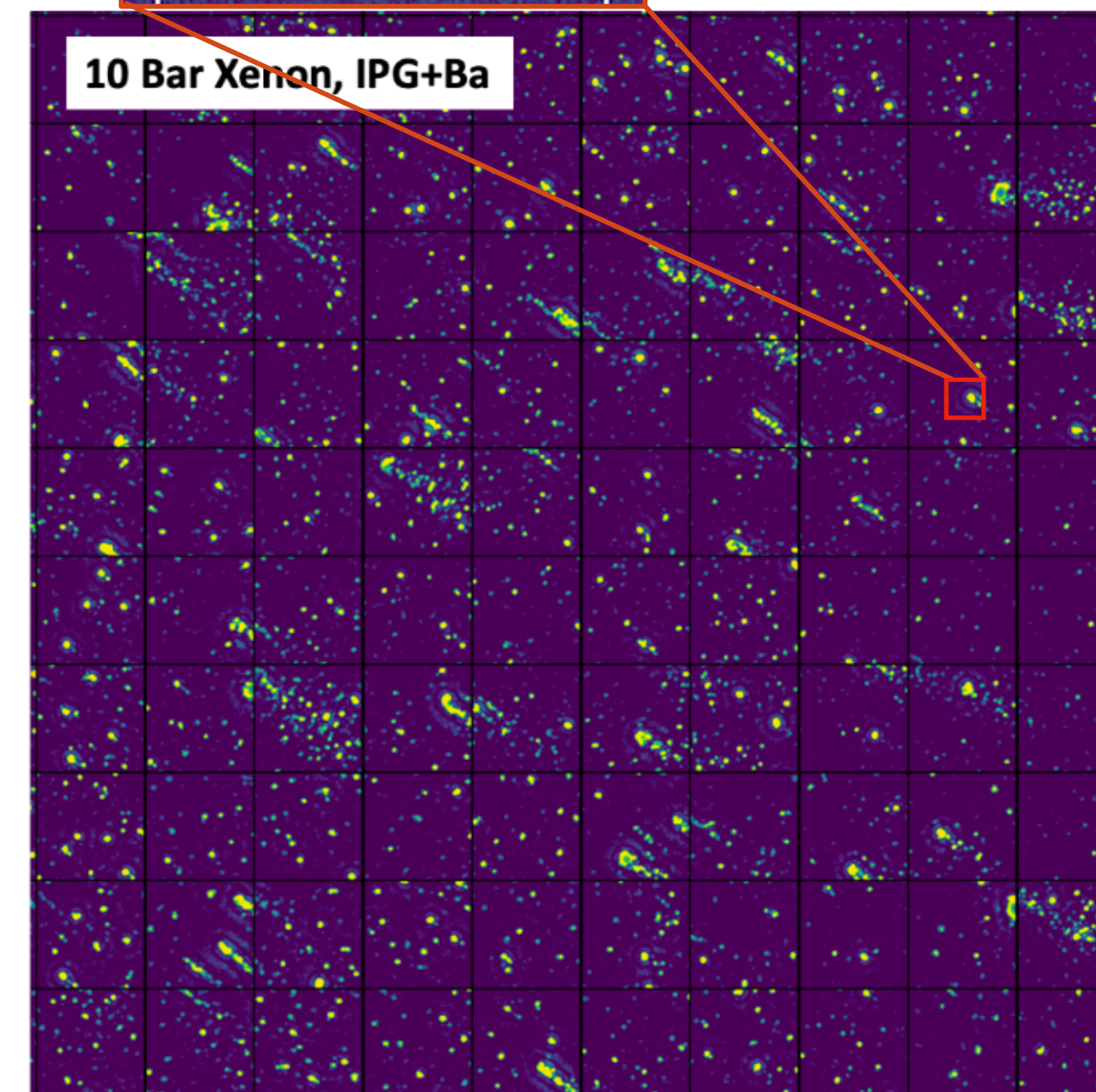
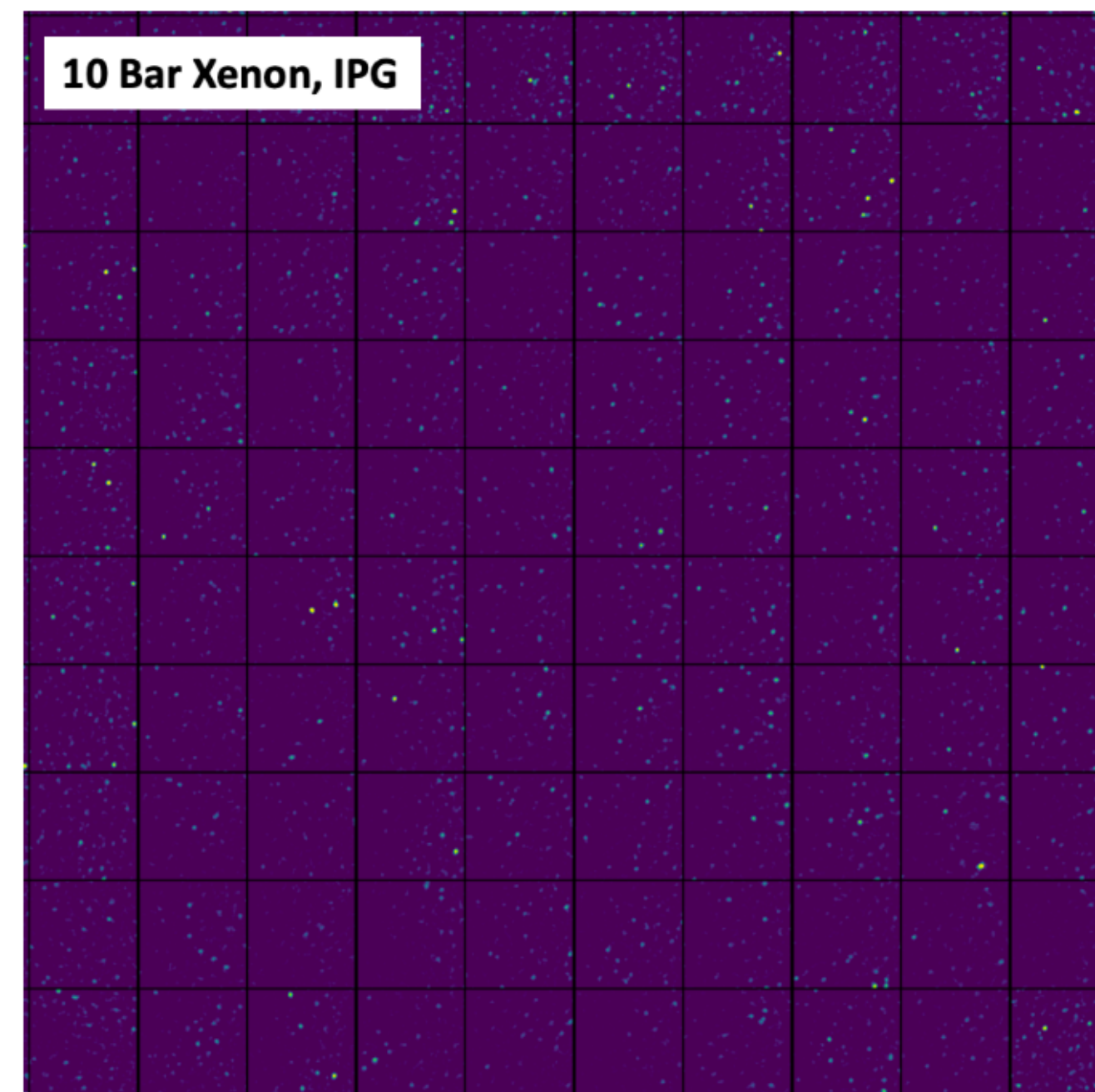


Microscopy efforts

SMFI in High-pressure Xe

- Raster scans for large-scale characterization of sample fluorescence
- Single-step detection of IPG (+Ba²⁺) molecules
- Autofocus algorithm with piezo stage

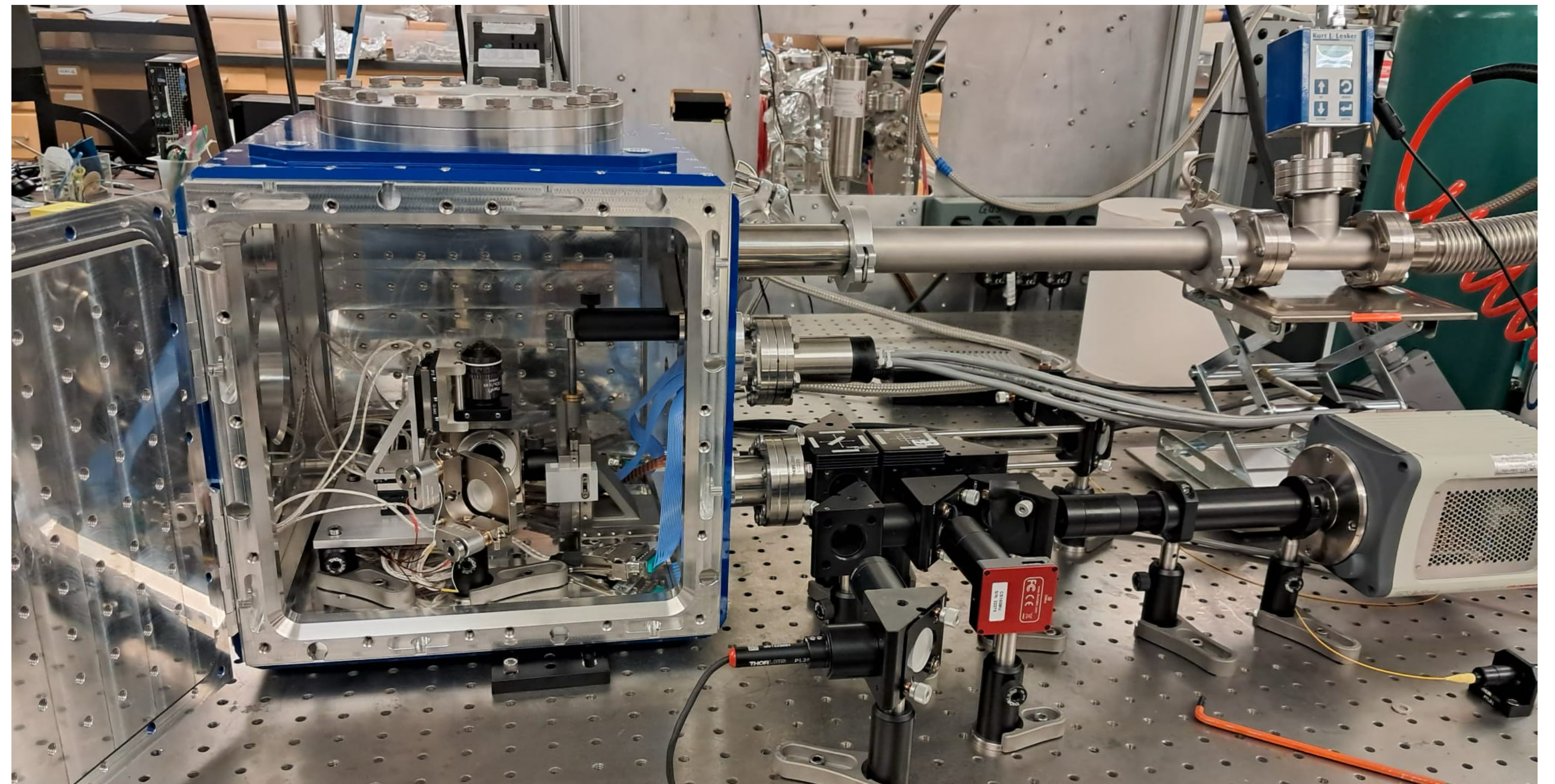
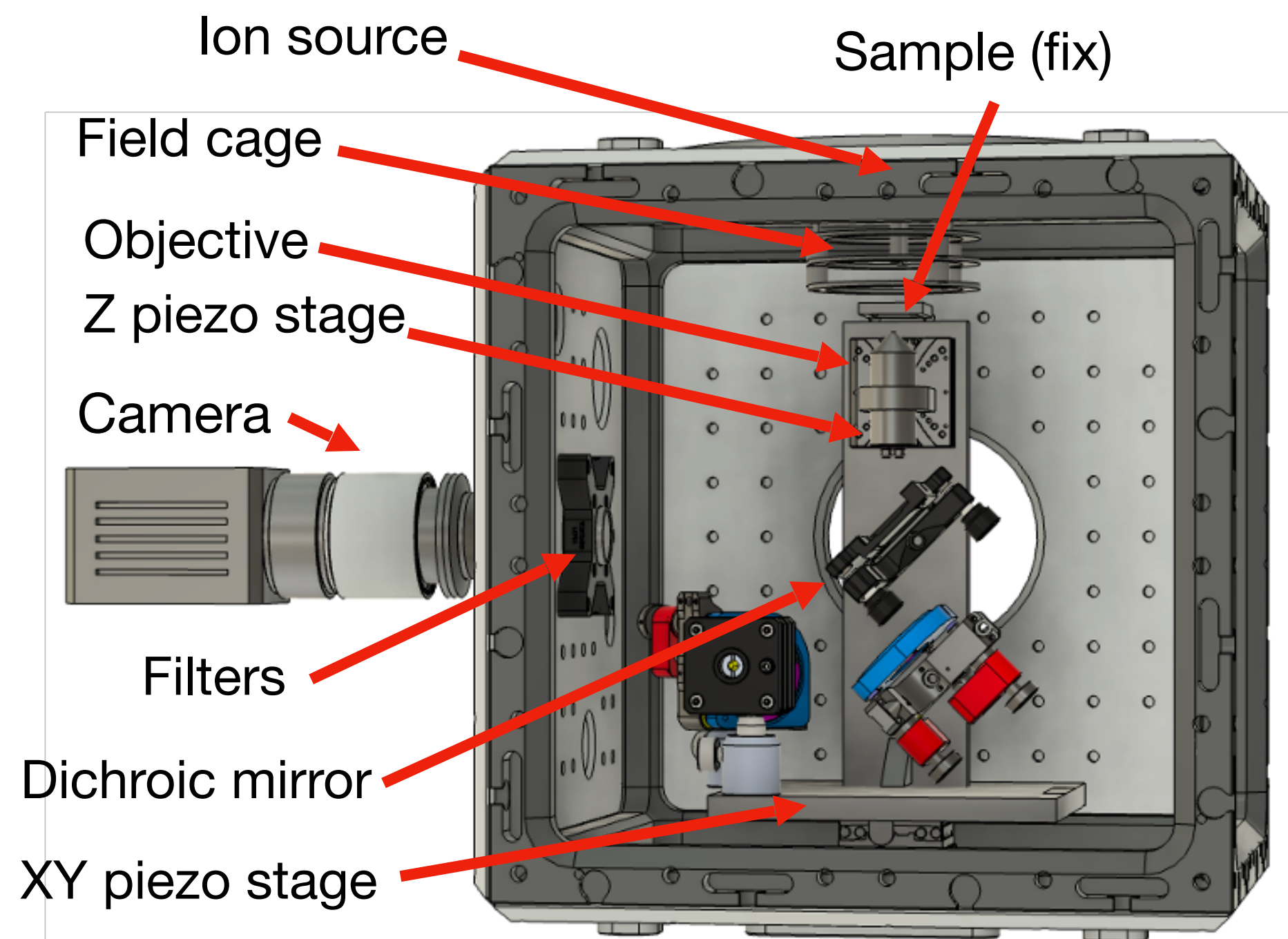
[a] Nat. Commun. 15, 10595 (2024).



Microscopy efforts

HP Microscope upgrade

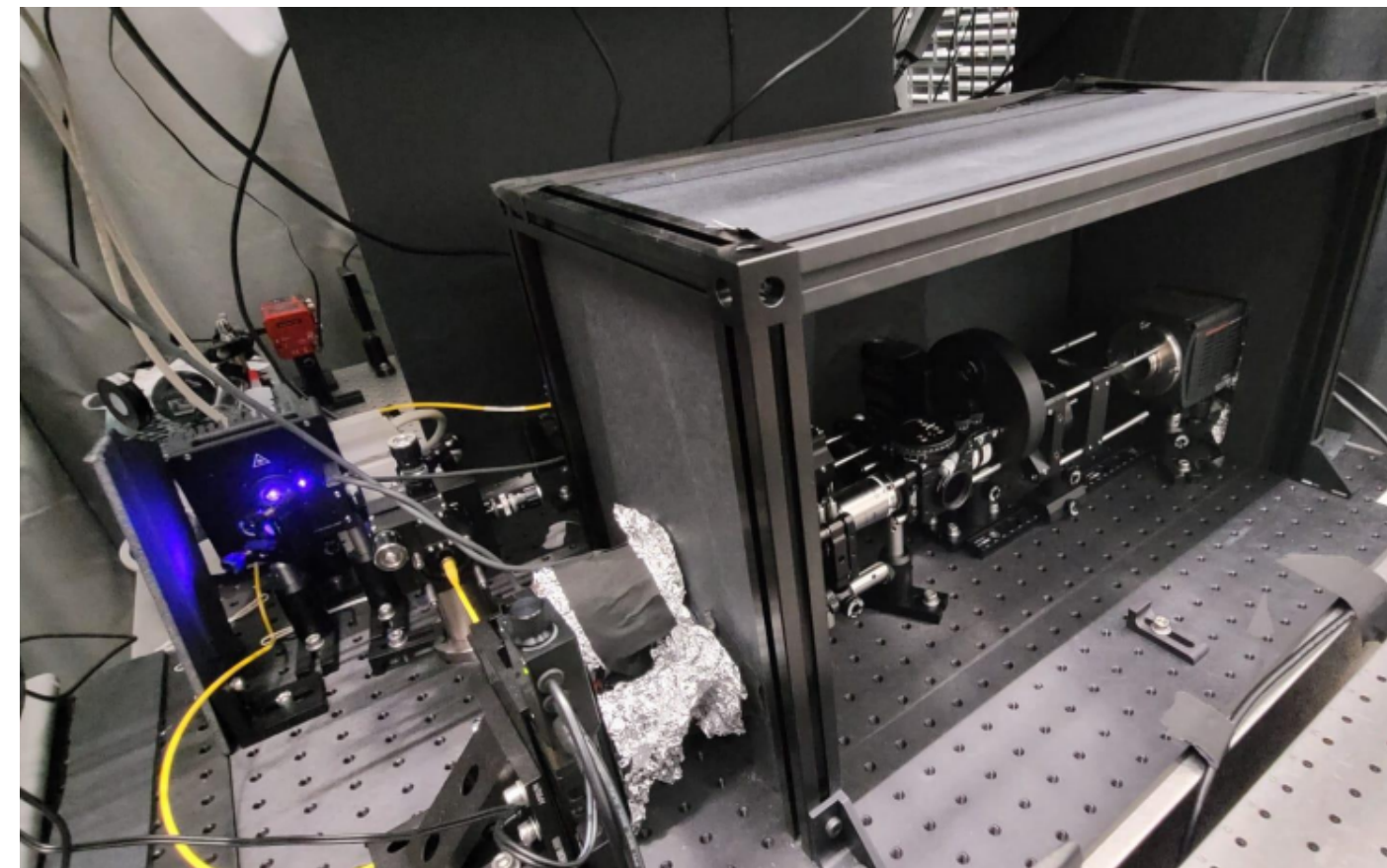
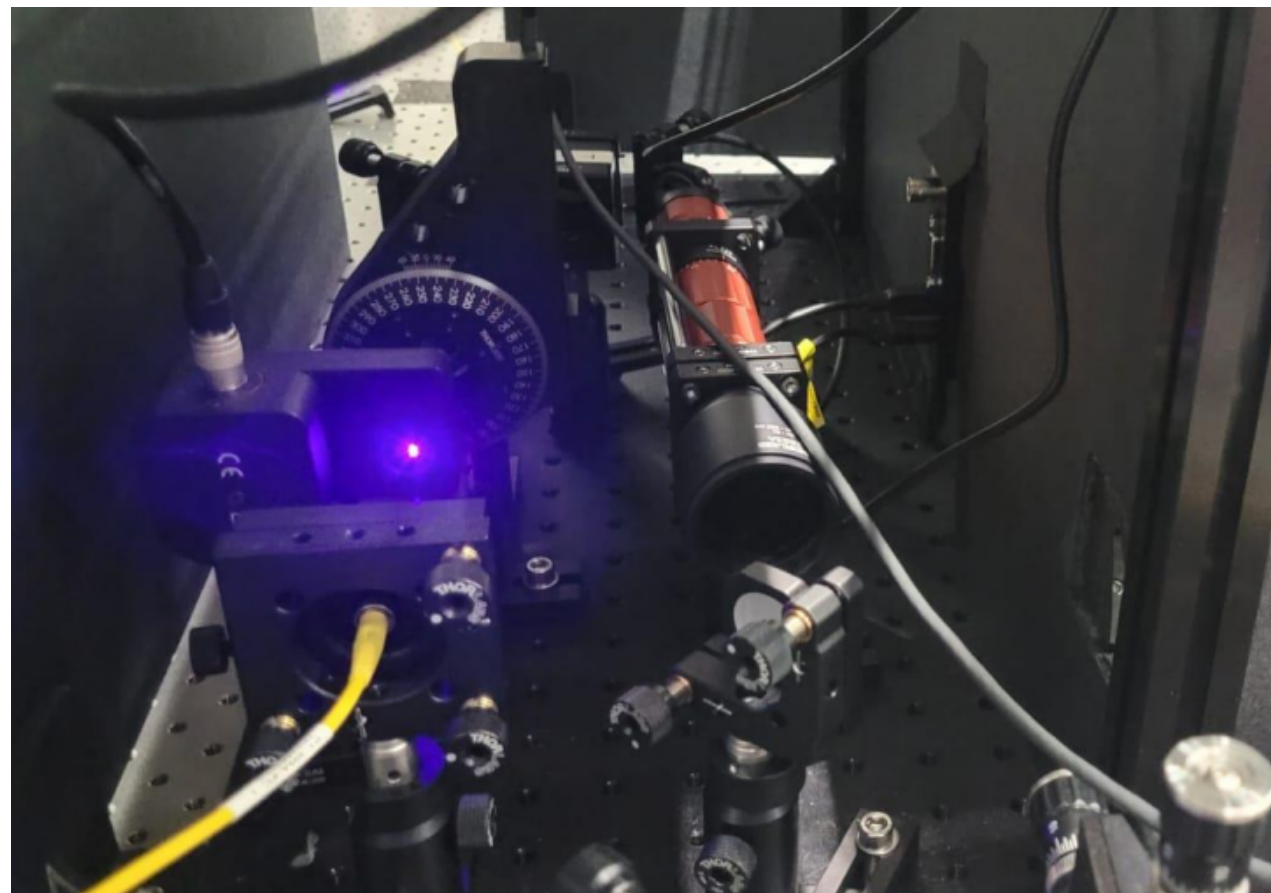
- Operating in HPGXe, coupling to Ba^{2+} source
- MO and optics move in scan, not sample



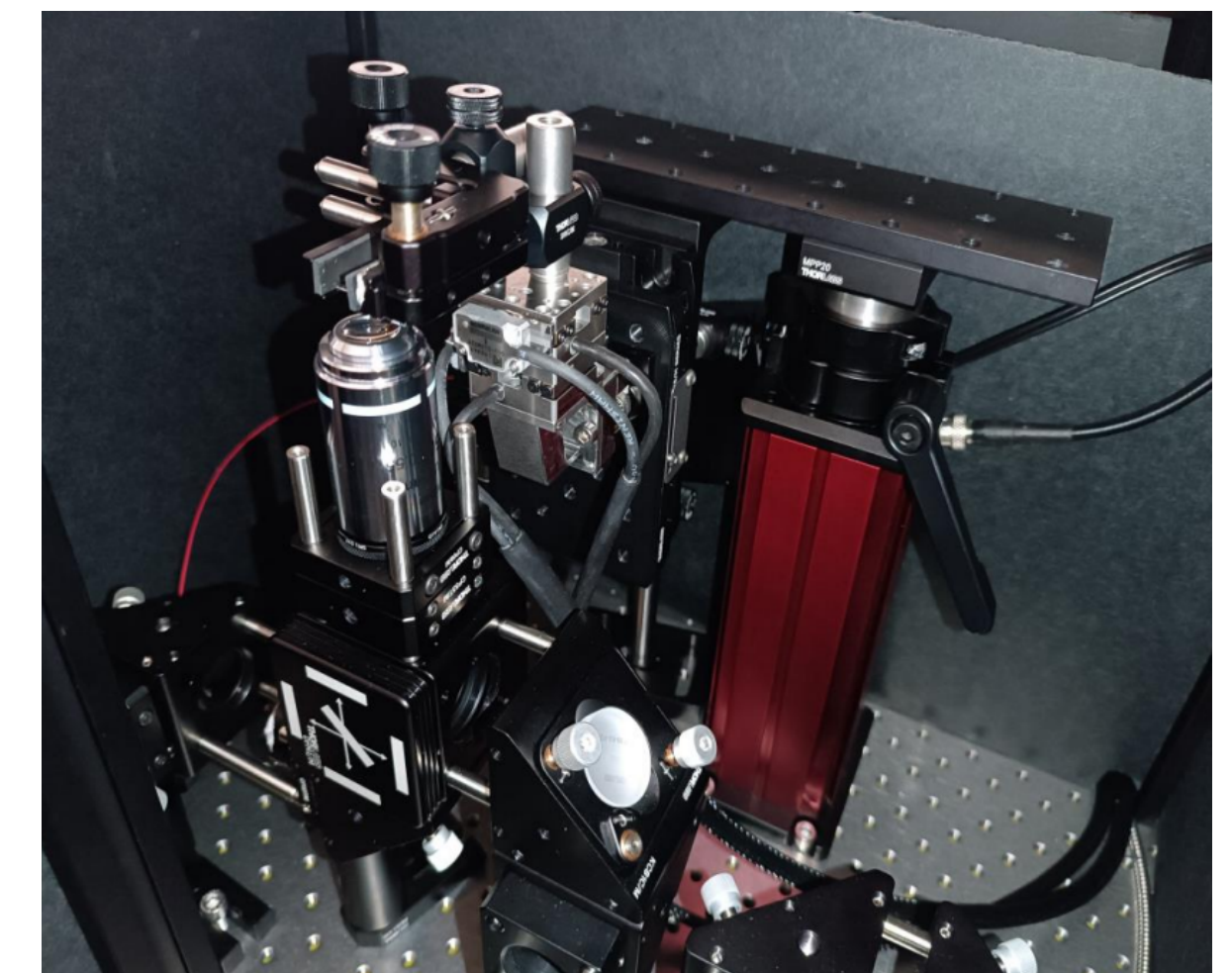
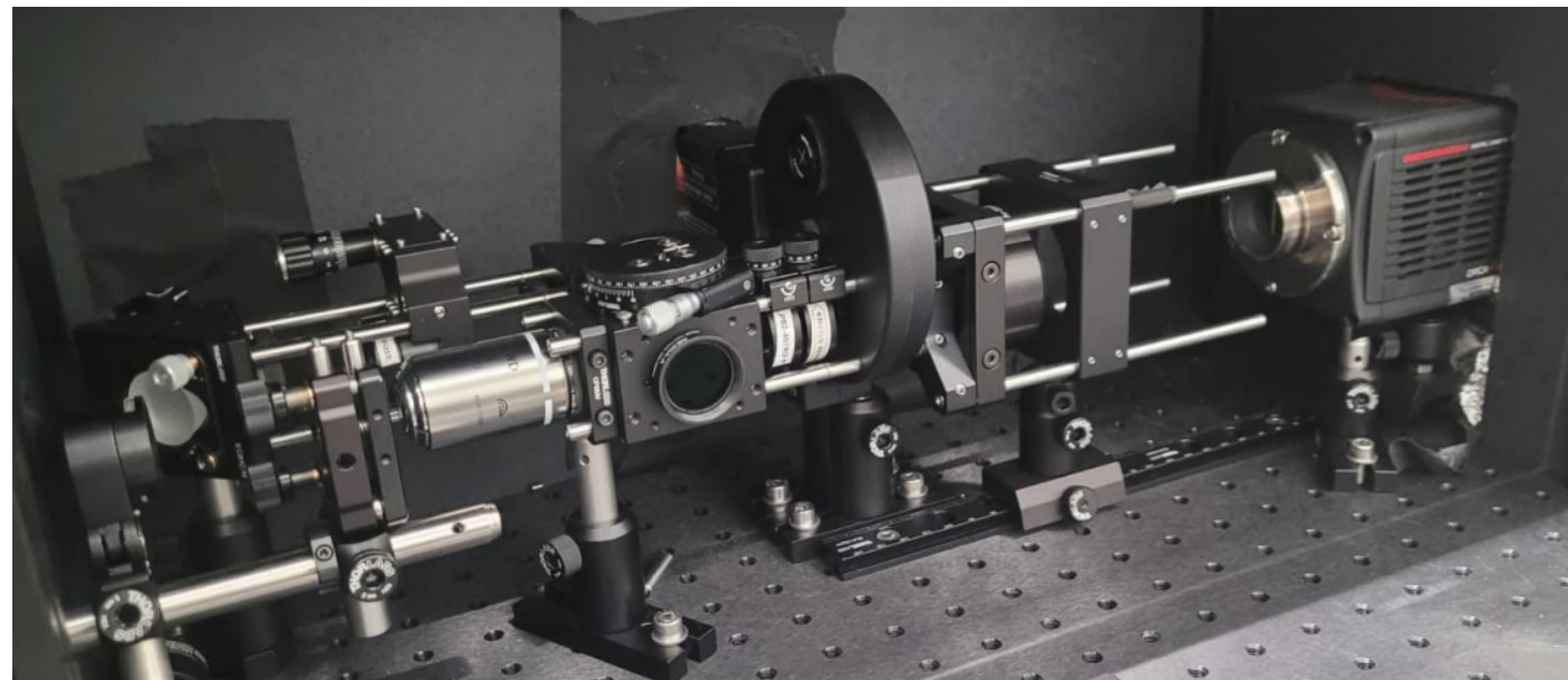
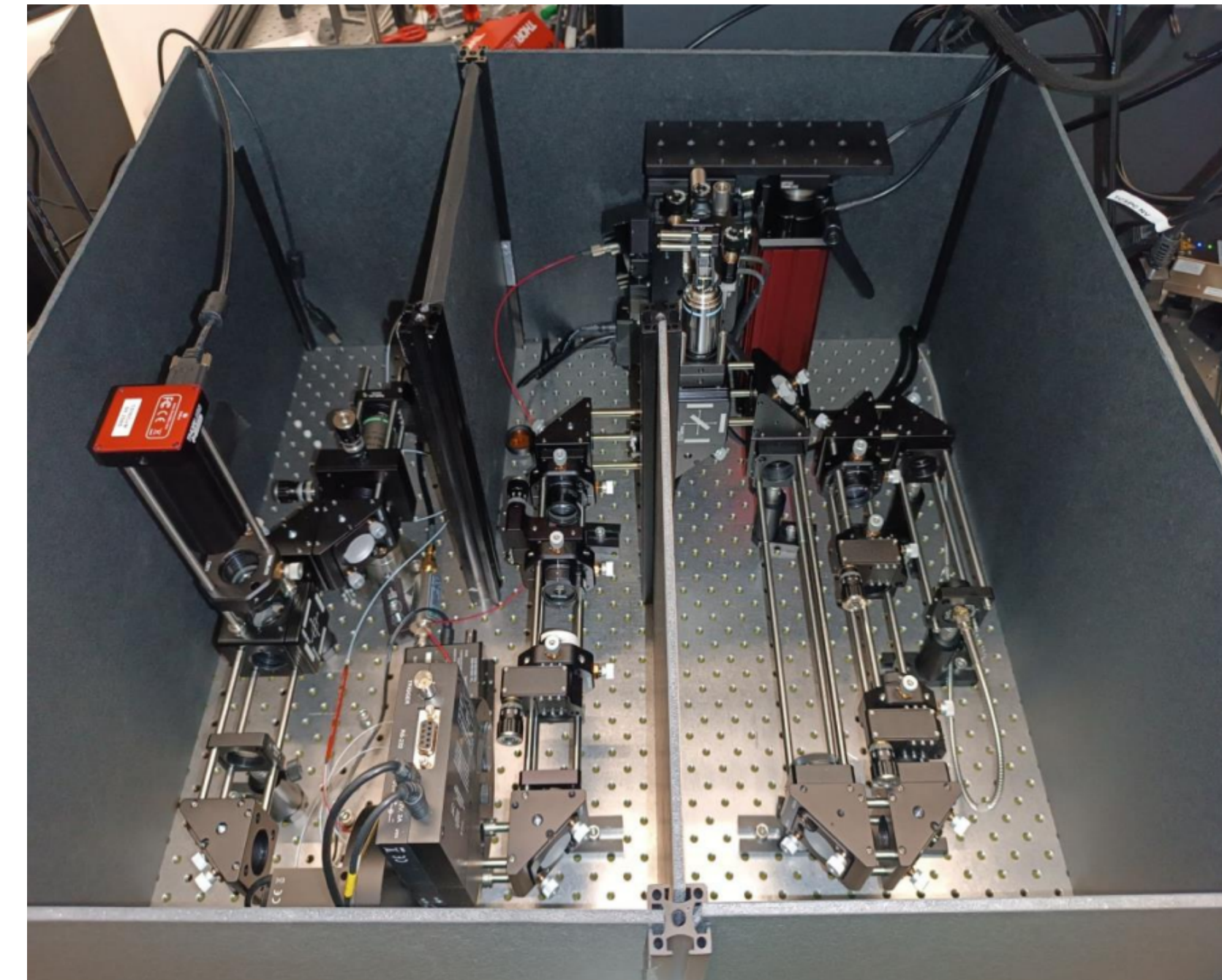
Microscopy efforts

Flexible setup configurations

Wide-field microscopy



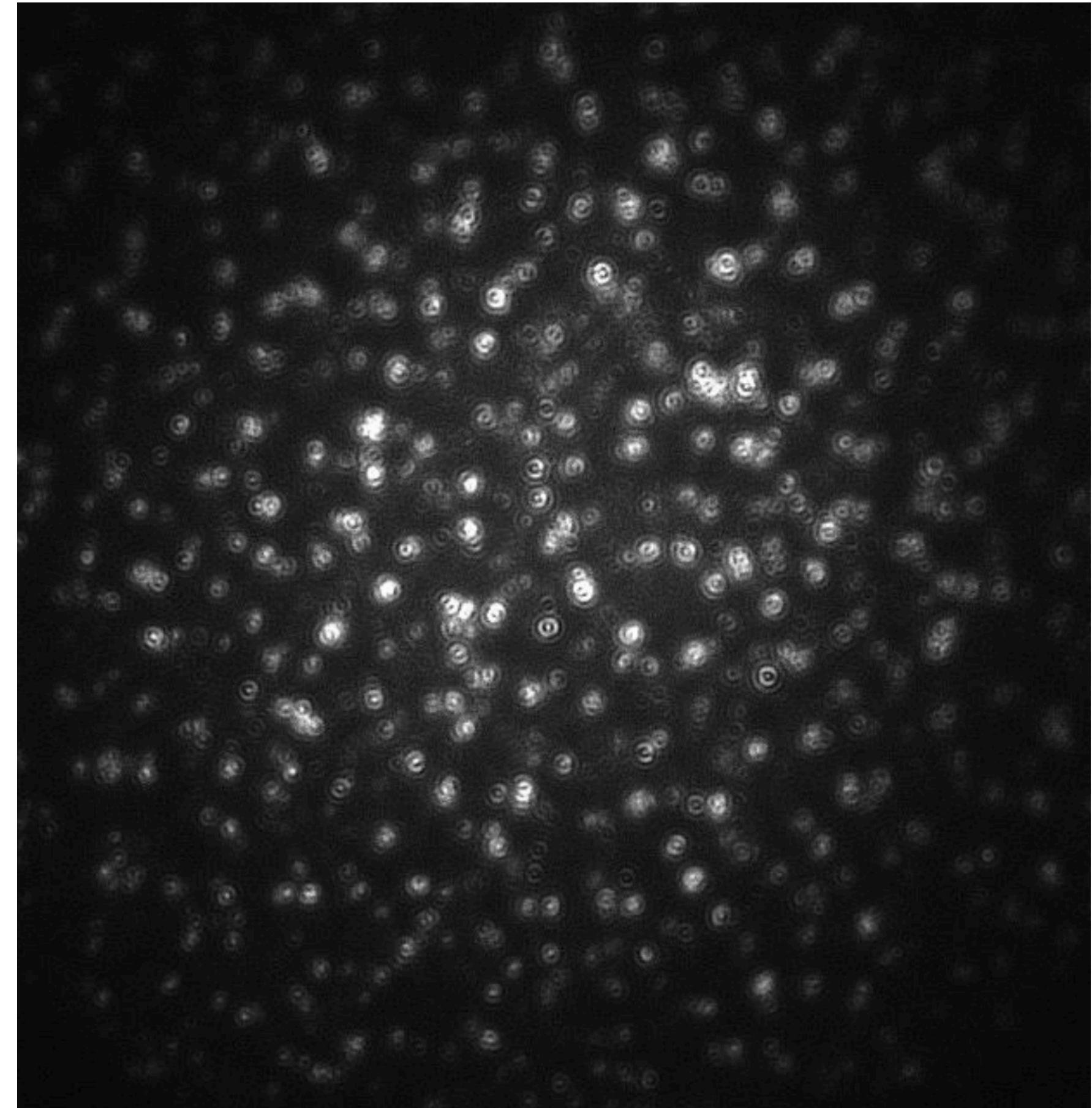
Prototyping/testing new techniques



Microscopy efforts

Photochemistry properties

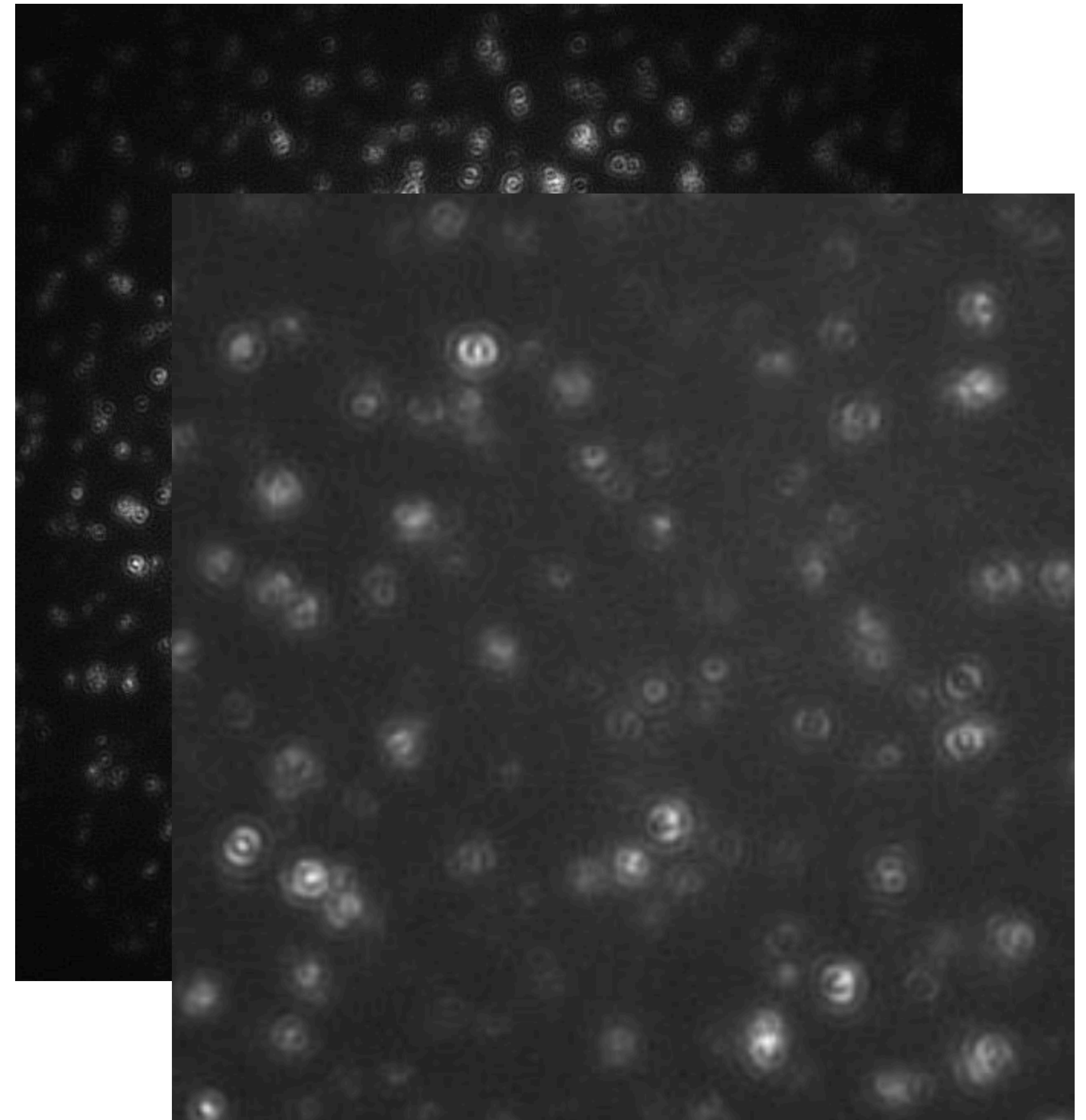
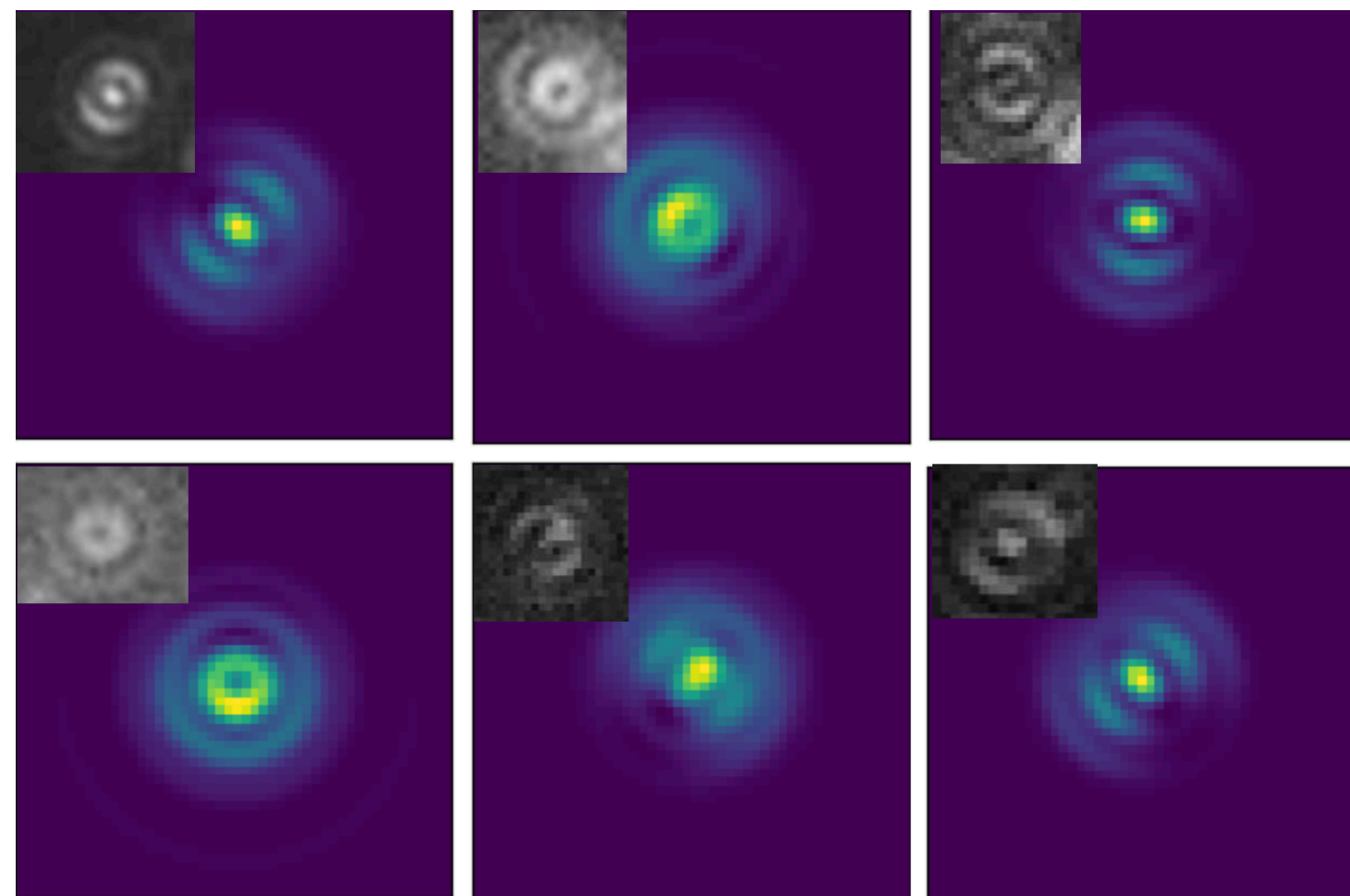
- SMFI and molecular orientation
 - Modeled as dipoles in electric field
 - Match pattern with simulation



Microscopy efforts

Photochemistry properties

- SMFI and molecular orientation
 - Modeled as dipoles in electric field
 - Match pattern with simulation



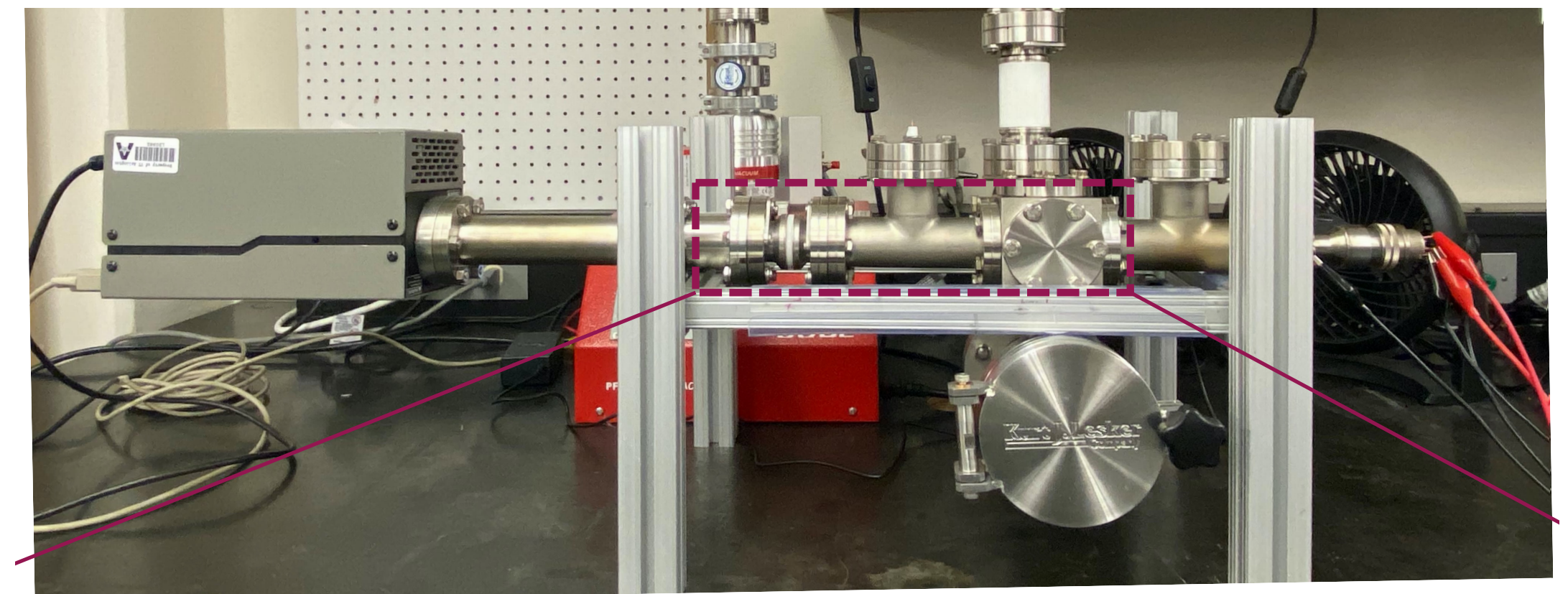
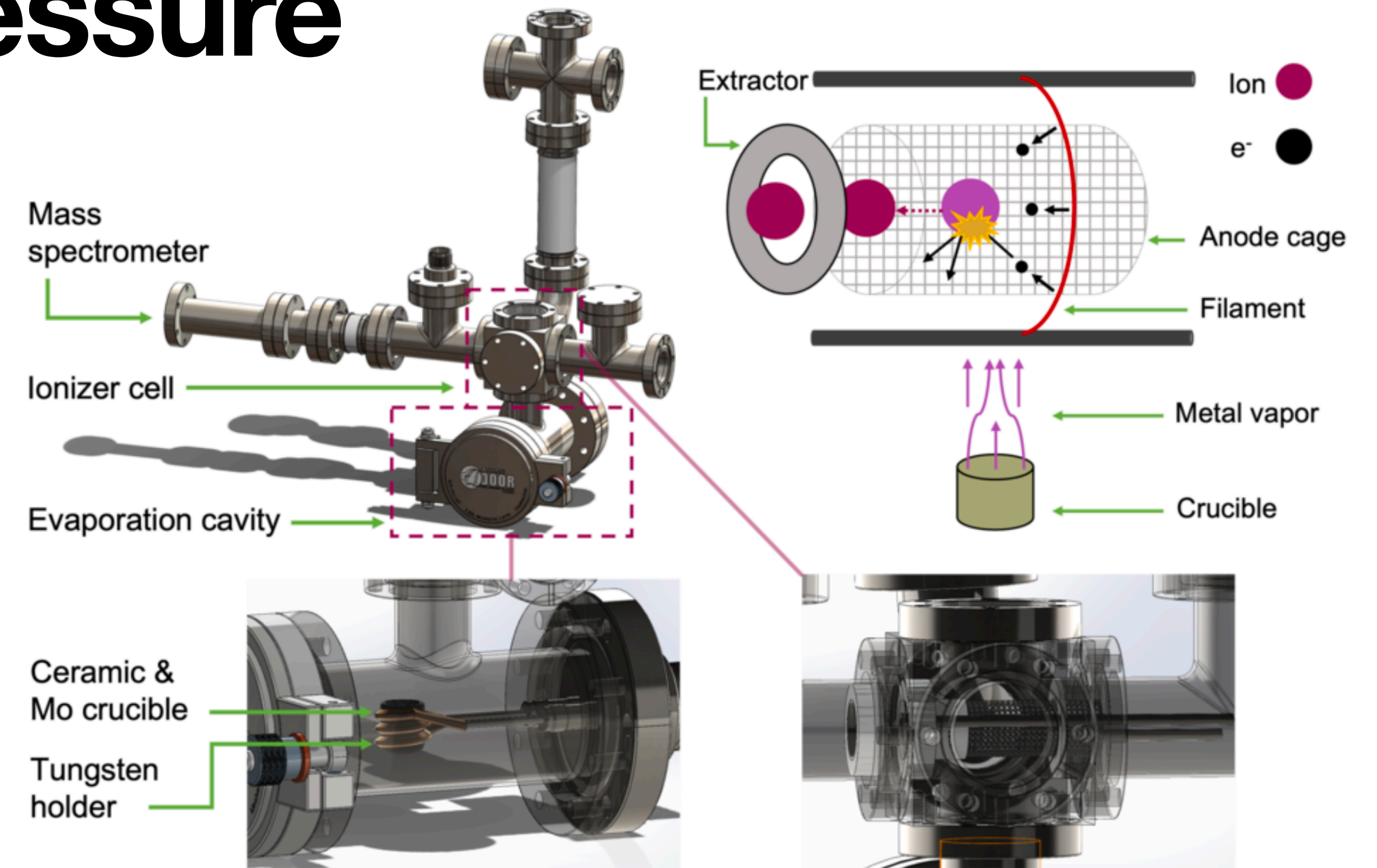
Overview

- ☒ Introduction to Ba-tagging
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Ba²⁺ beam in high pressure

Controlled energy ions

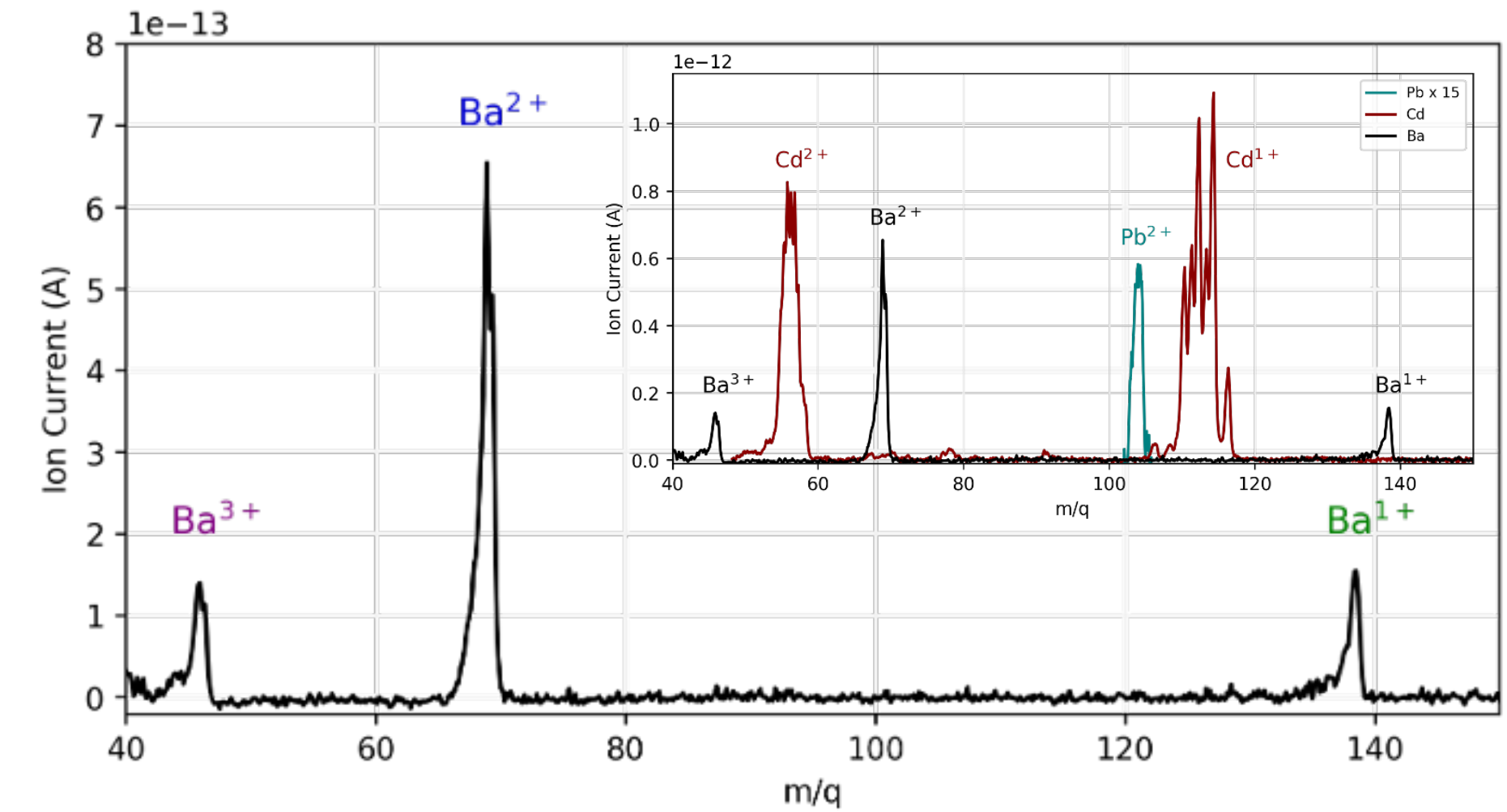
- Compact, solvent-free Ba²⁺ ion source
- Combines metal vaporization + electron-impact ionization with tunable voltages.



Ba²⁺ beam in high pressure

Controlled energy ions

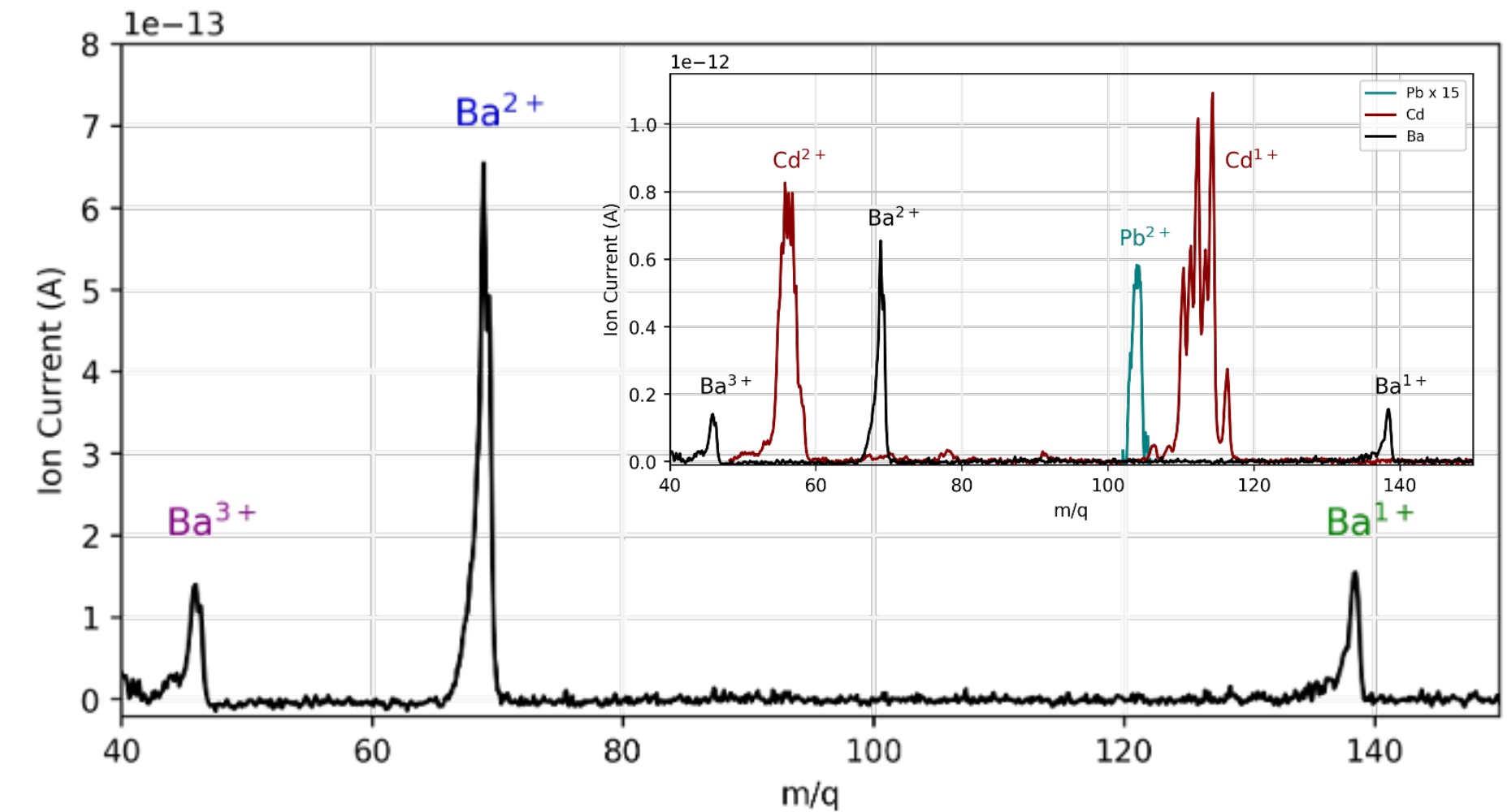
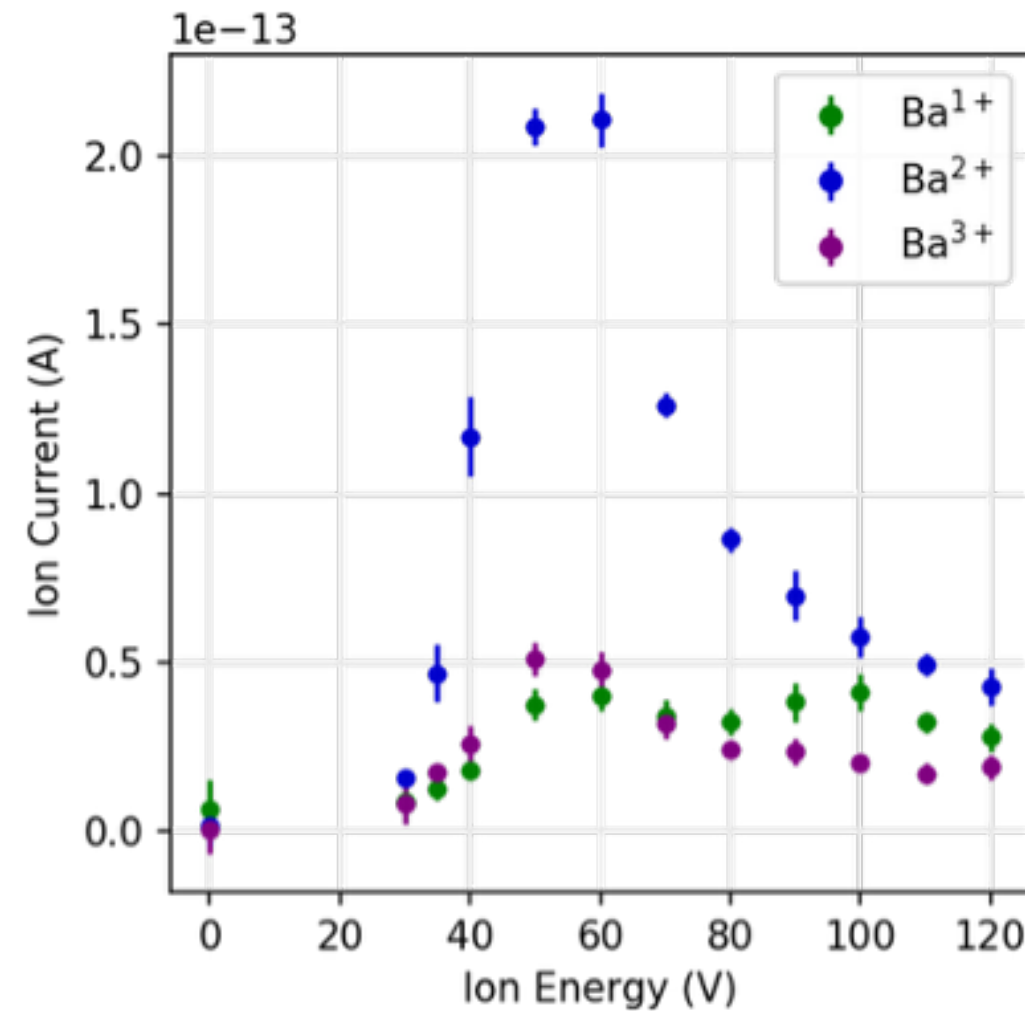
- Compact, solvent-free Ba²⁺ ion source
- Combines metal vaporization + electron-impact ionization with tunable voltages.



Ba²⁺ beam in high pressure

Controlled energy ions

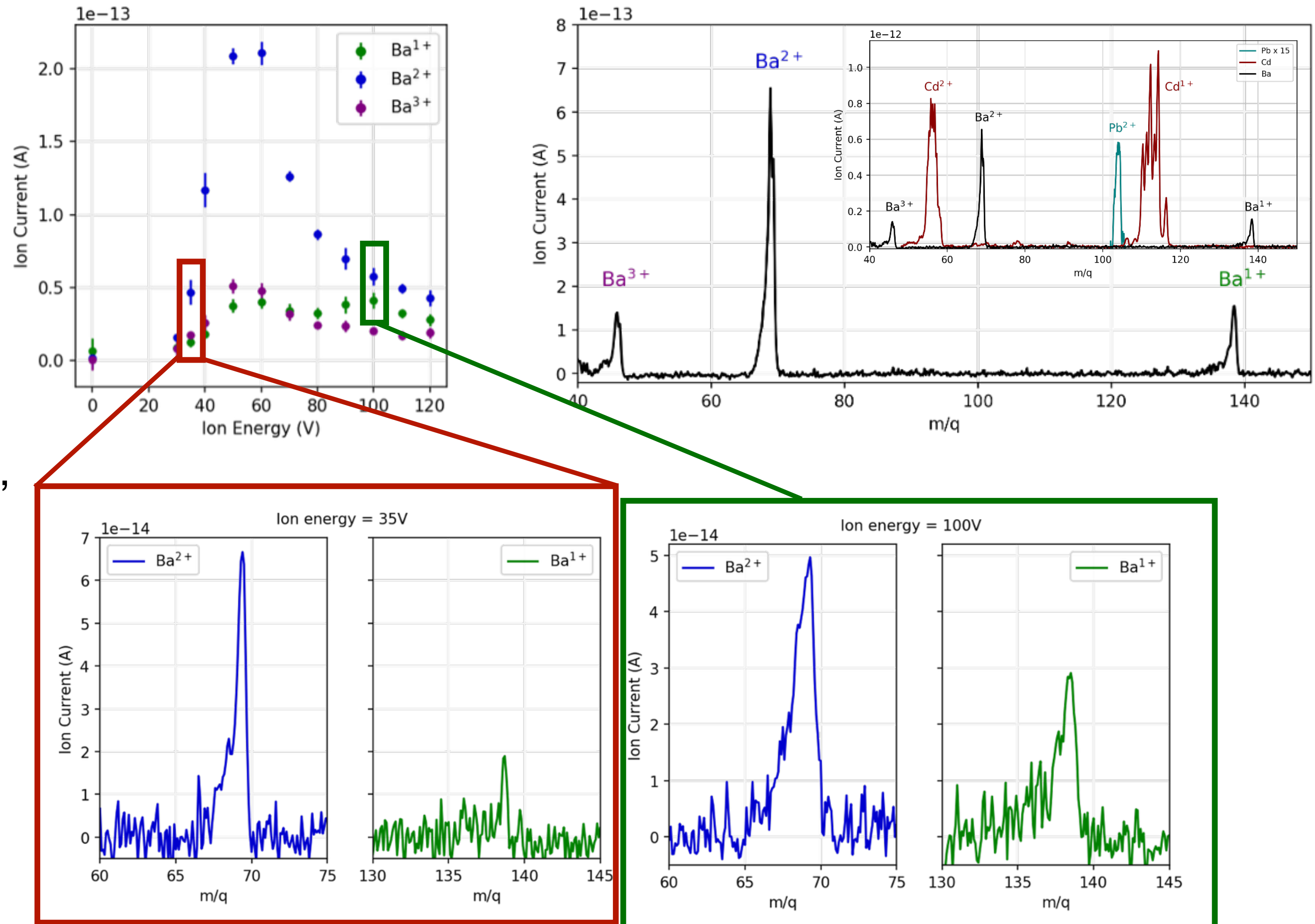
- Compact, solvent-free Ba²⁺ ion source
- Combines metal vaporization + electron-impact ionization with tunable voltages.
- Achieved stable, controllable Ba²⁺, Pb²⁺, Cd²⁺ beams confirmed via mass spectrometry.
- Optimized parameters (I_e , V_e , V_i) → majority Ba²⁺ flux.



Ba²⁺ beam in high pressure

Controlled energy ions

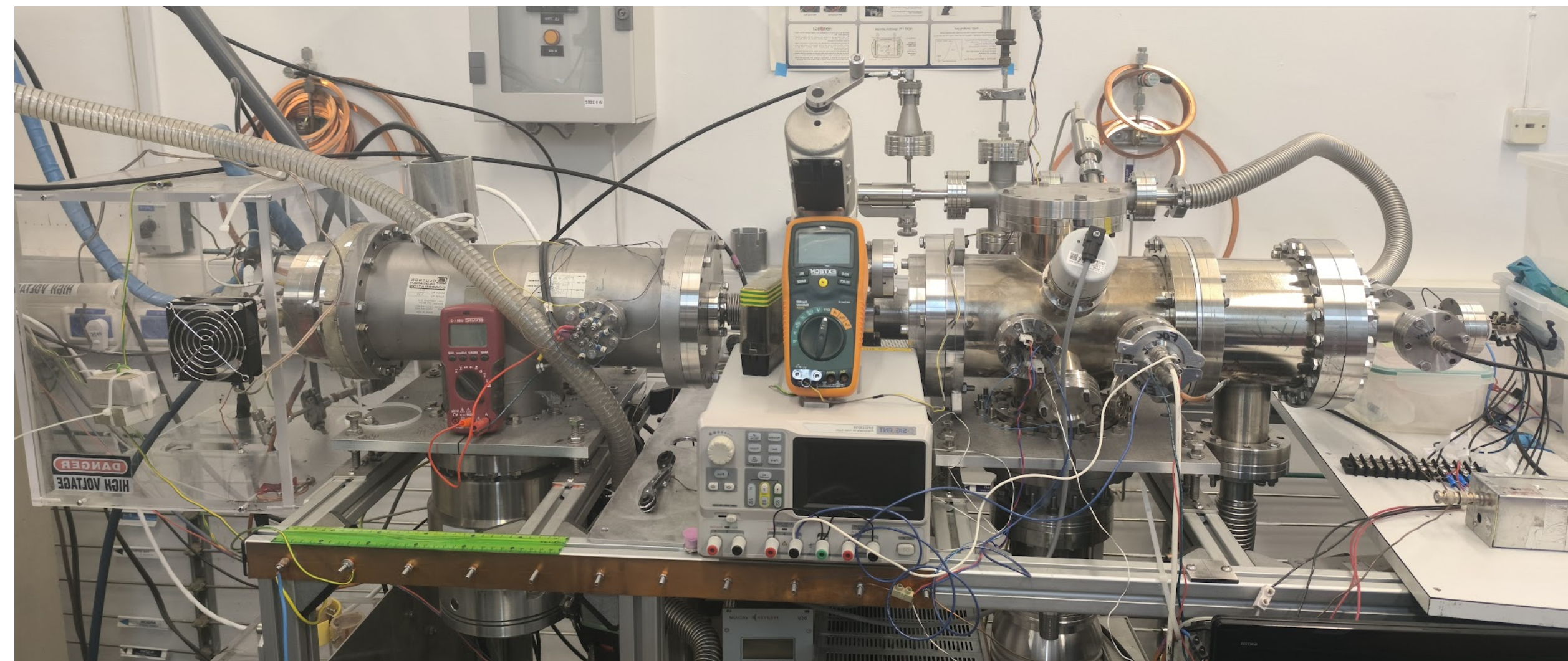
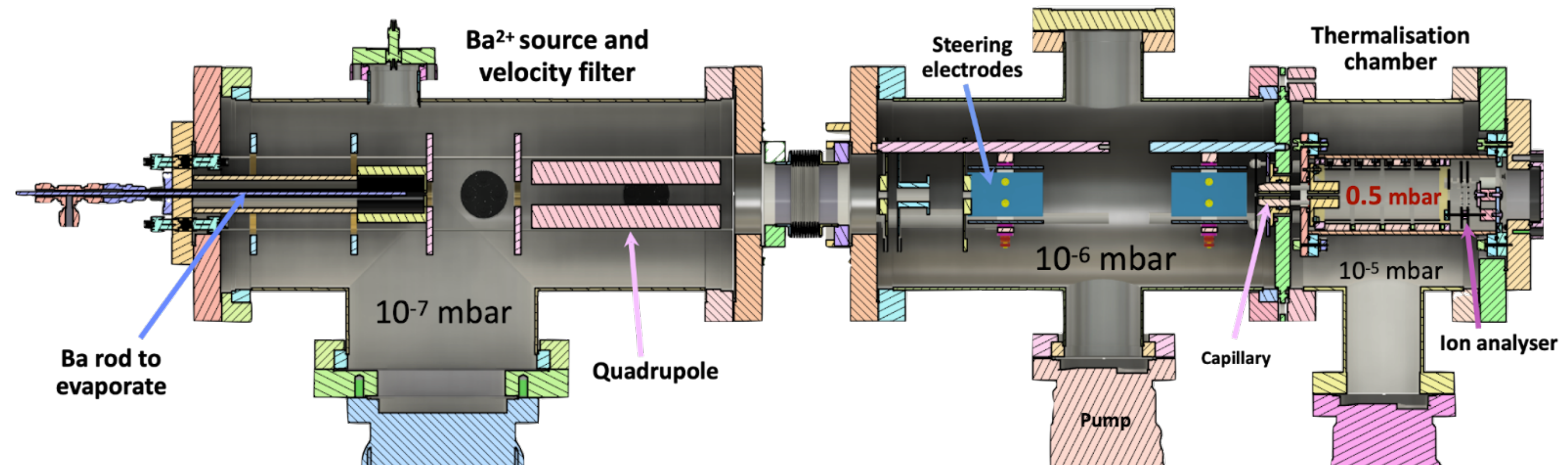
- Compact, solvent-free Ba²⁺ ion source
- Combines metal vaporization + electron-impact ionization with tunable voltages.
- Achieved stable, controllable Ba²⁺, Pb²⁺, Cd²⁺ beams confirmed via mass spectrometry.
- Optimized parameters (I_e , V_e , V_i) → majority Ba²⁺ flux.
- Need thermalization of ions for testing with molecules



Ba²⁺ beam in low pressure

Thermal ions

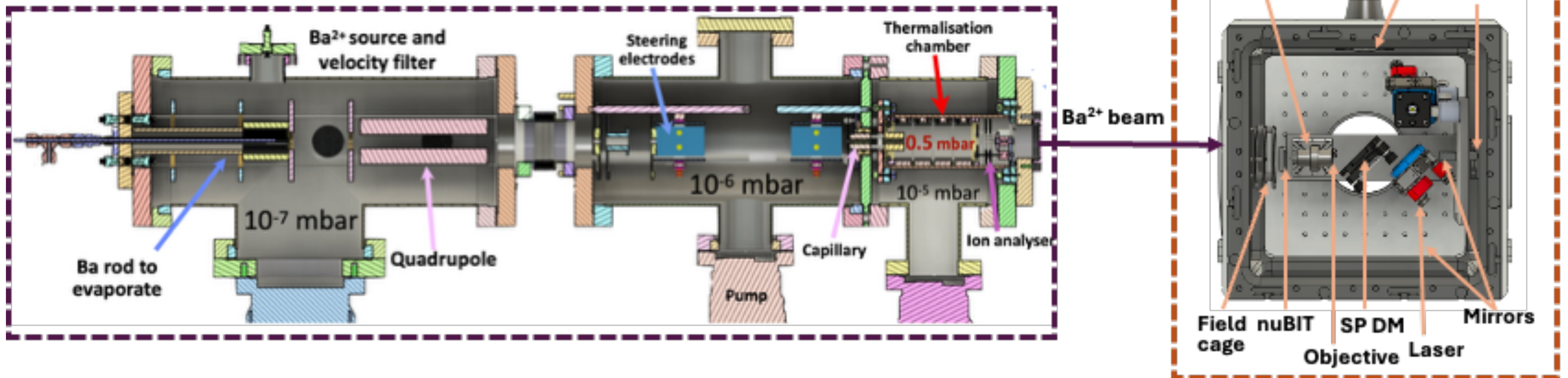
- Sublimation of Ba → filtering from other ions
- Steering into thermalization chamber though differential pumping
- Collision with GHe at ~0.5 mbar



Ba²⁺ beam in low pressure

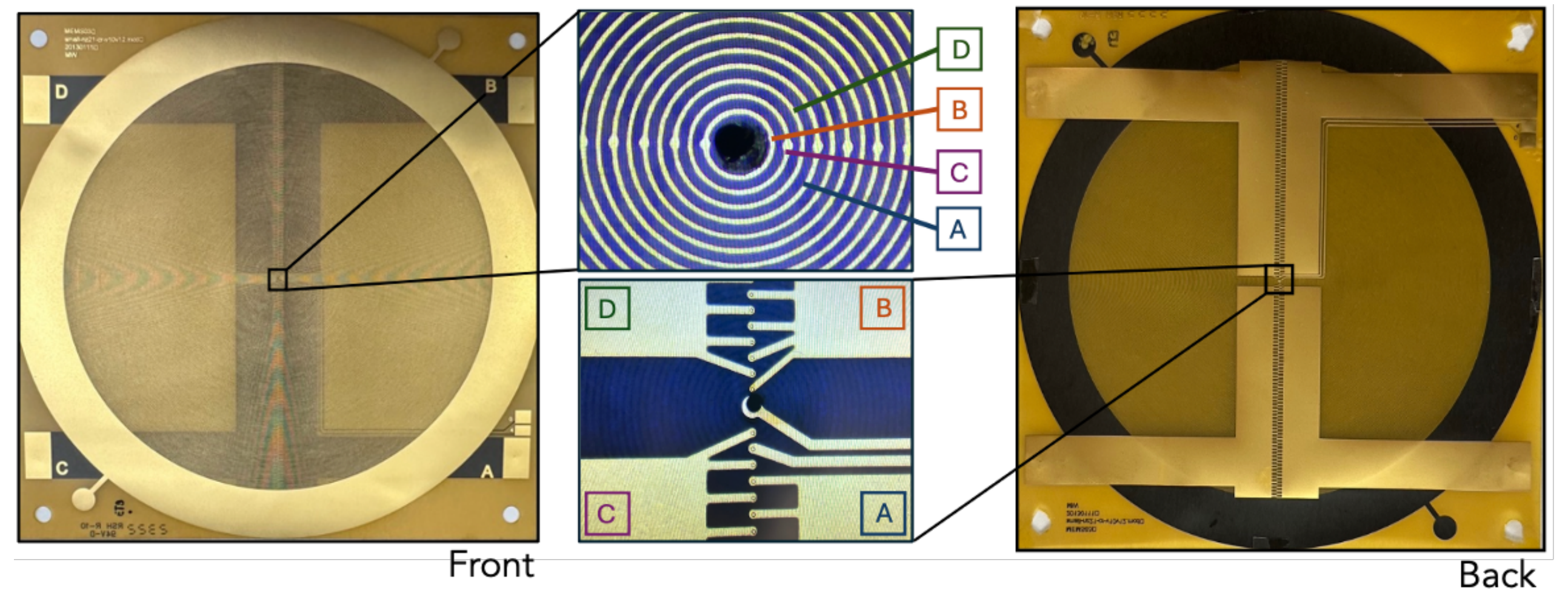
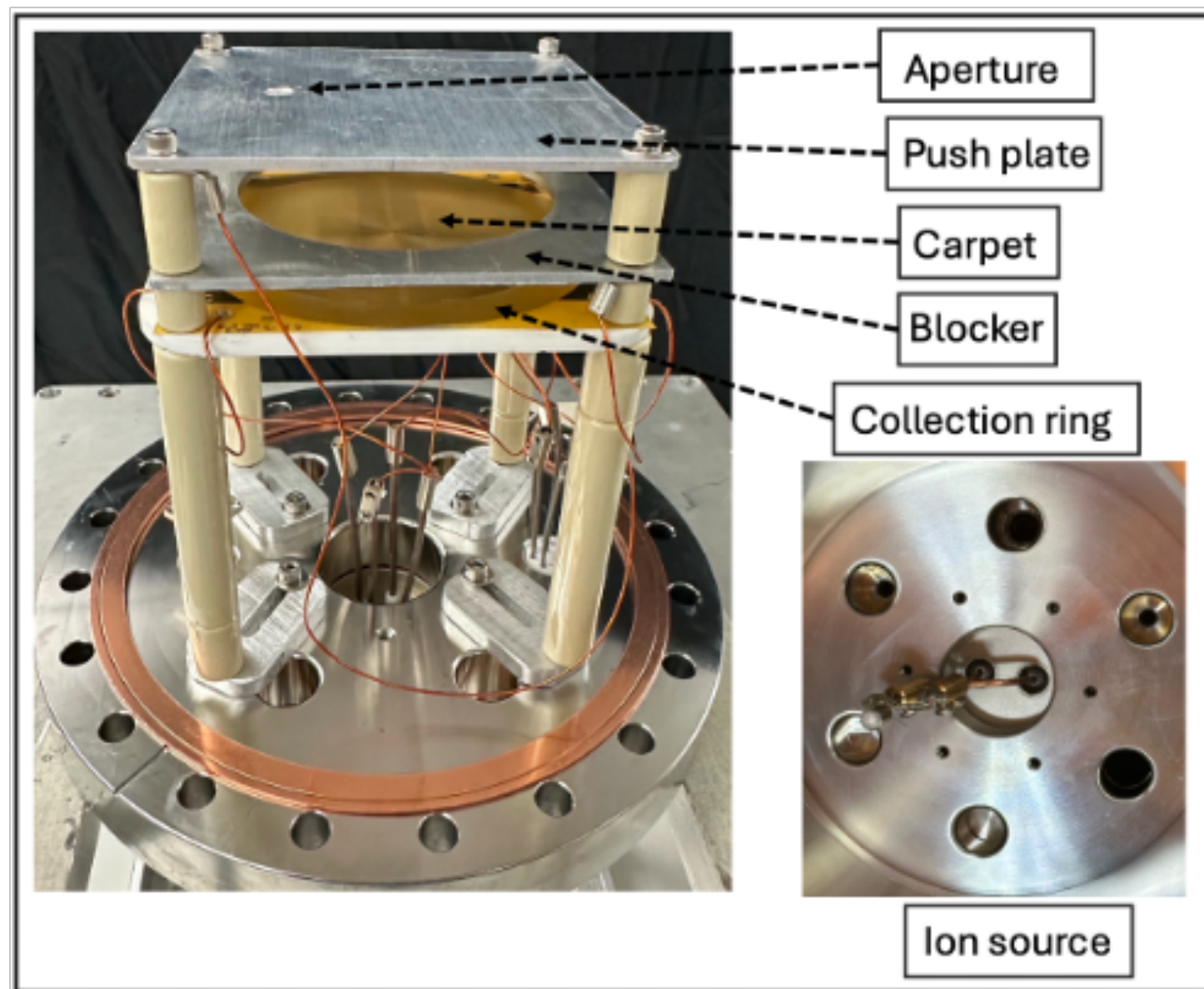
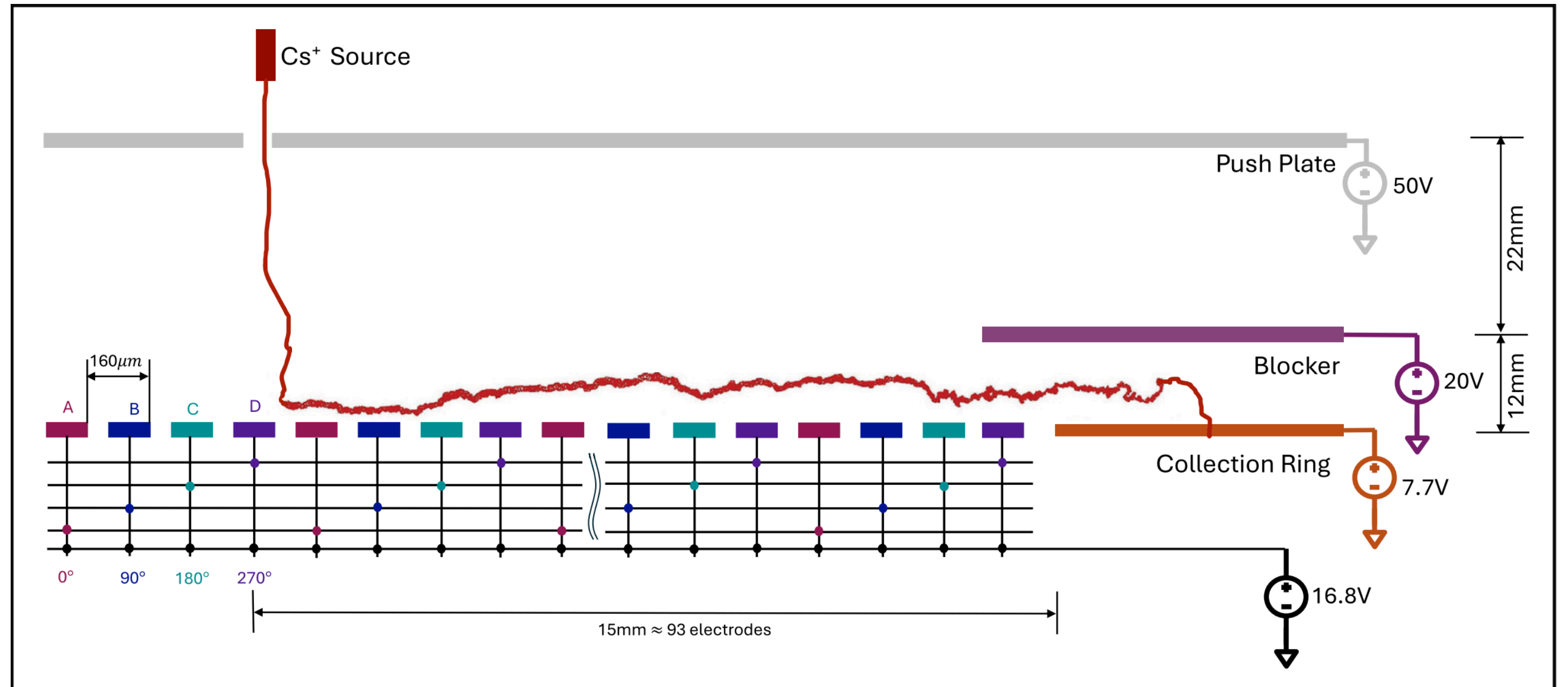
Coupling to microscope

- Aim: Testing detection of single Ba-chelated molecules *in-situ* at vacuum with thermal ions.
- Optimized HP microscope already available, coupling to low-energy beam planned.



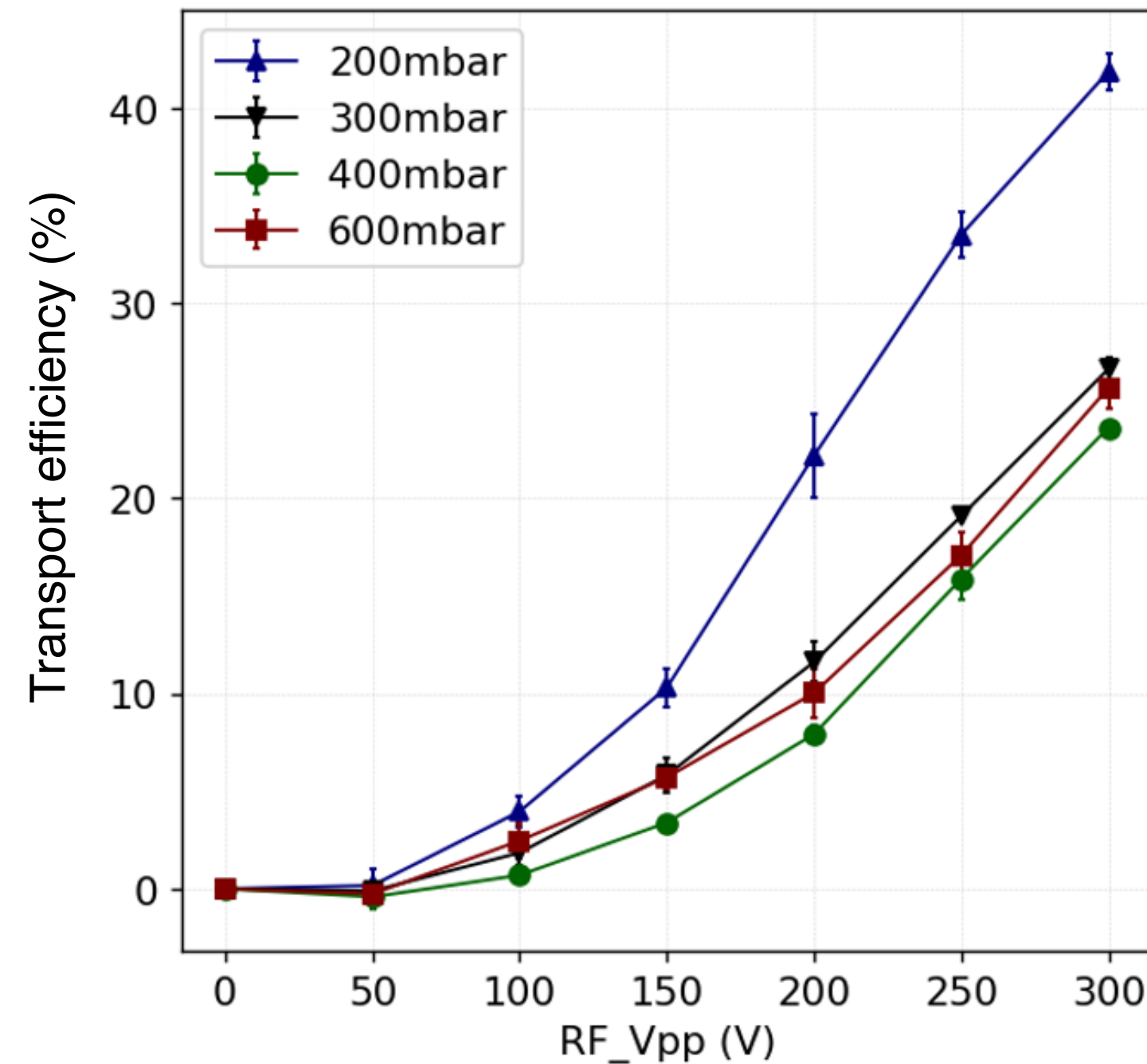
Ba²⁺ guide in GXe with RF carpet

- Using Cs⁺ as Ba²⁺ conservative stand-in
- 160 μm electrode pitch, at 2 MHz, up to 300 V
- Maximum efficiency for four-phase mode, as predicted from simulation



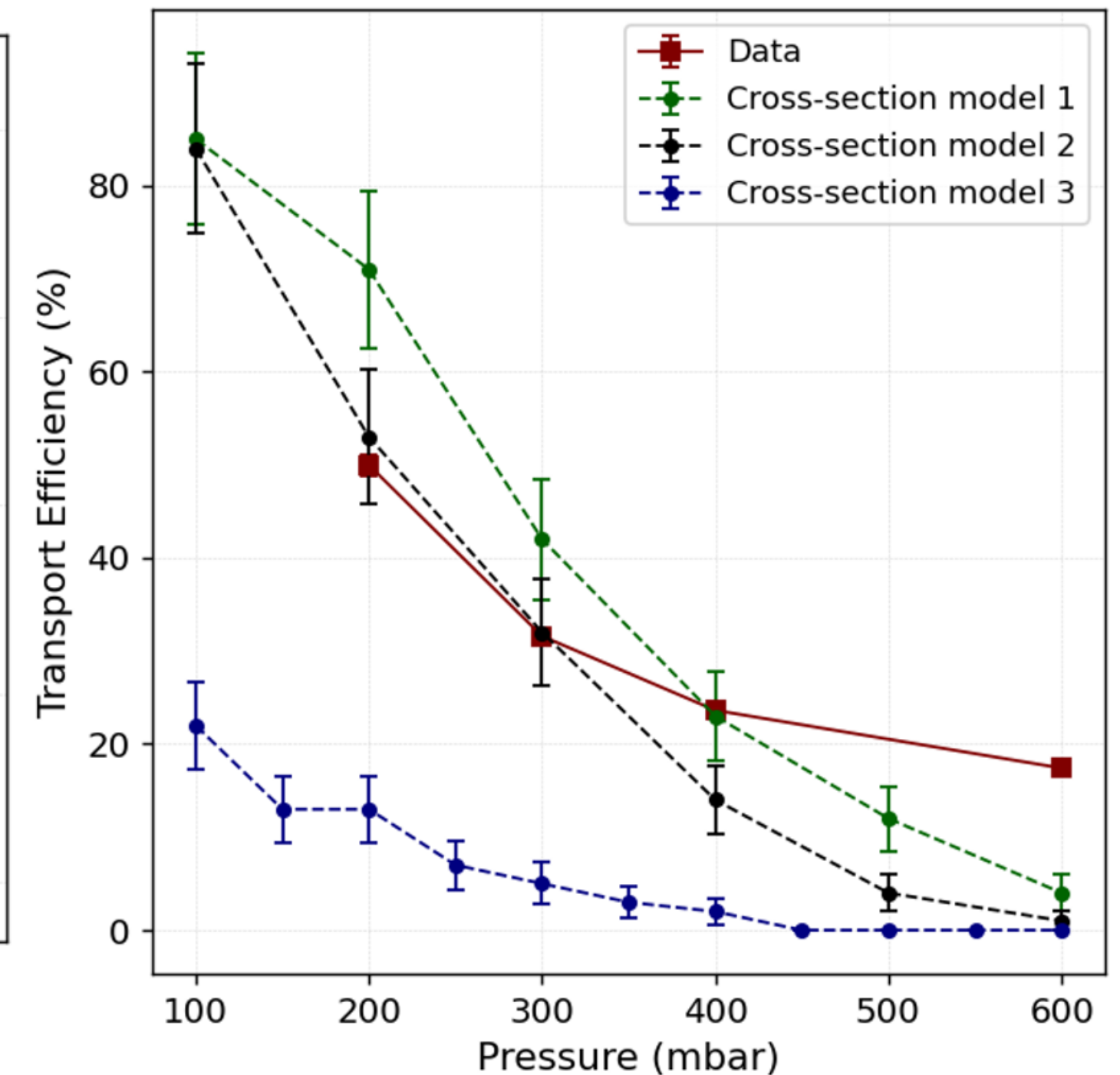
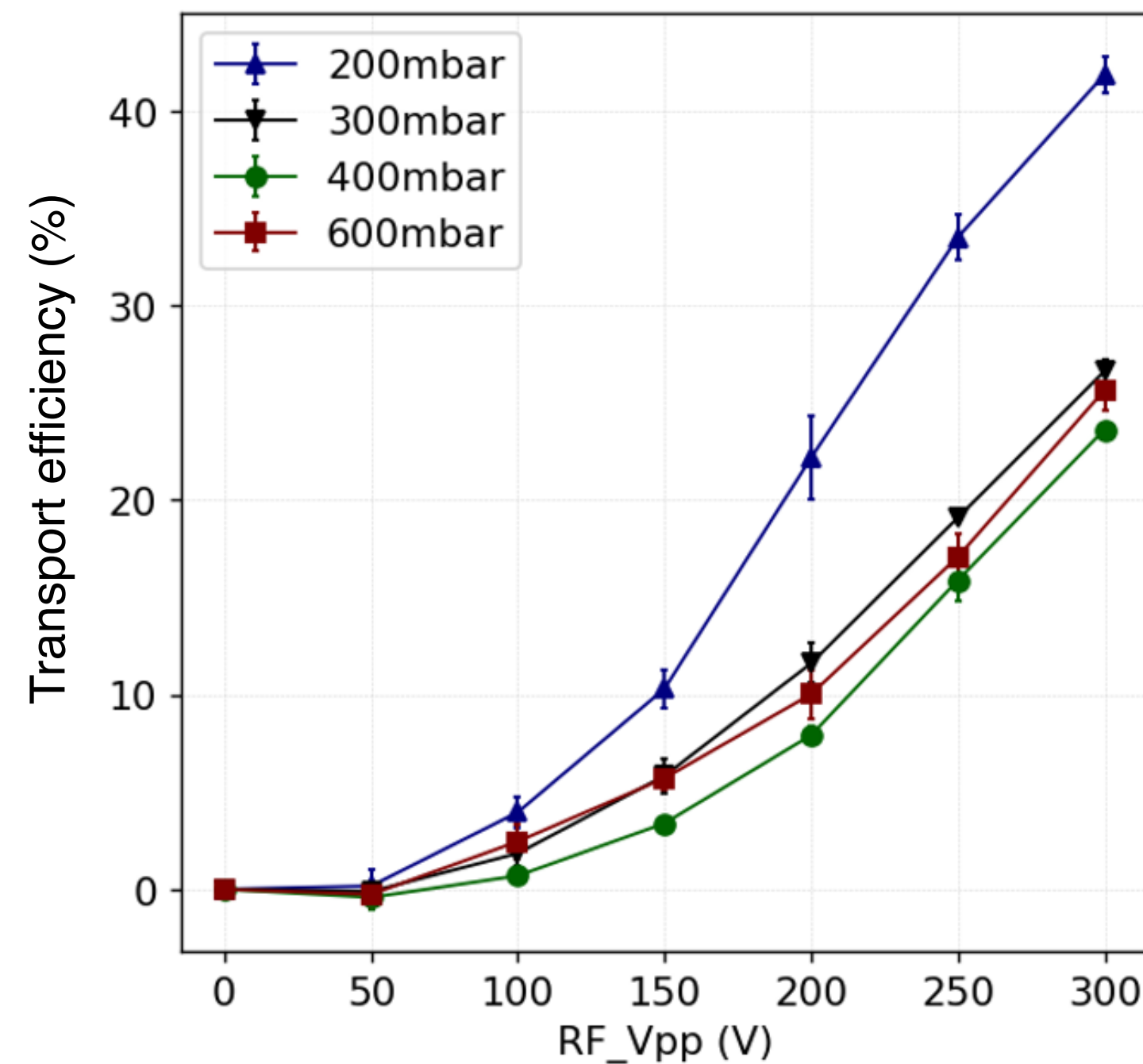
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- Transport efficiency depends on with RF voltage and phase difference



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- Successful ion transport at up to 600 mbar
- Transport efficiency depends on with RF voltage and phase difference
- 3 cross-section models in qualitative agreement; clustering model (3) too restrictive



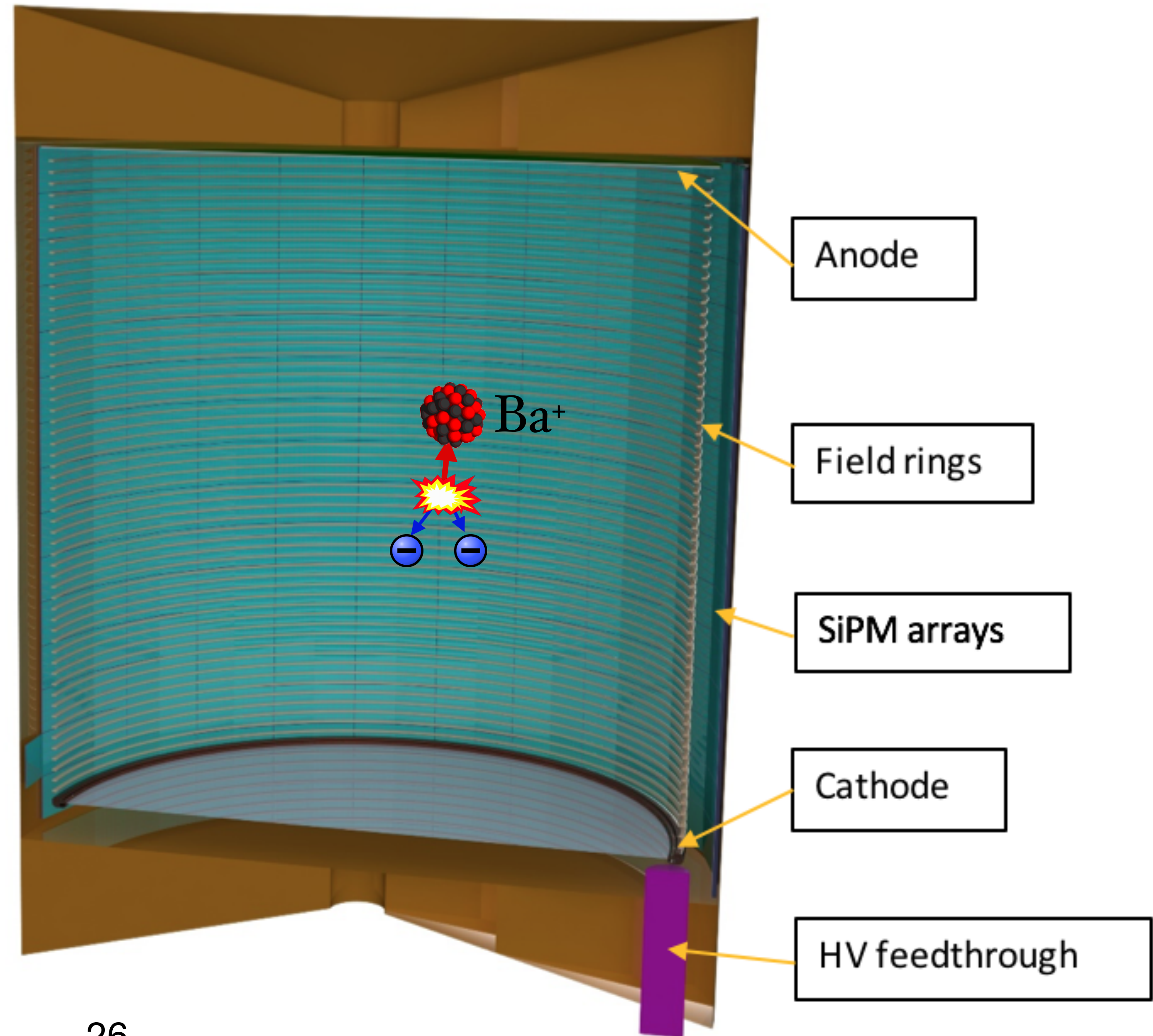
More on clustering: Phys. Rev. A. 97, 062509 (2018)

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- ☐ The road forward

Ba-Tagging in LXe

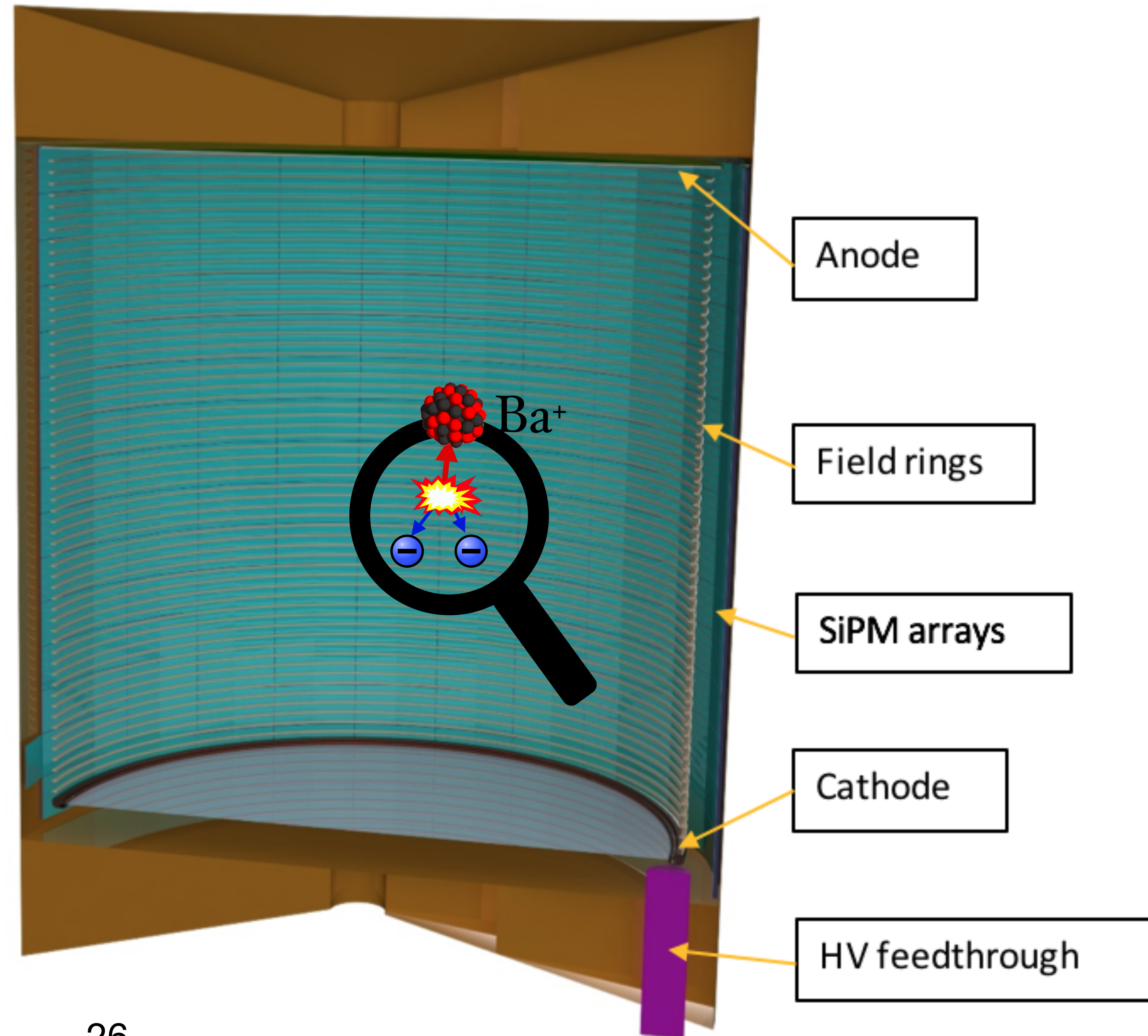
Concept



Ba-Tagging in LXe

Concept

1. Localization^[a]

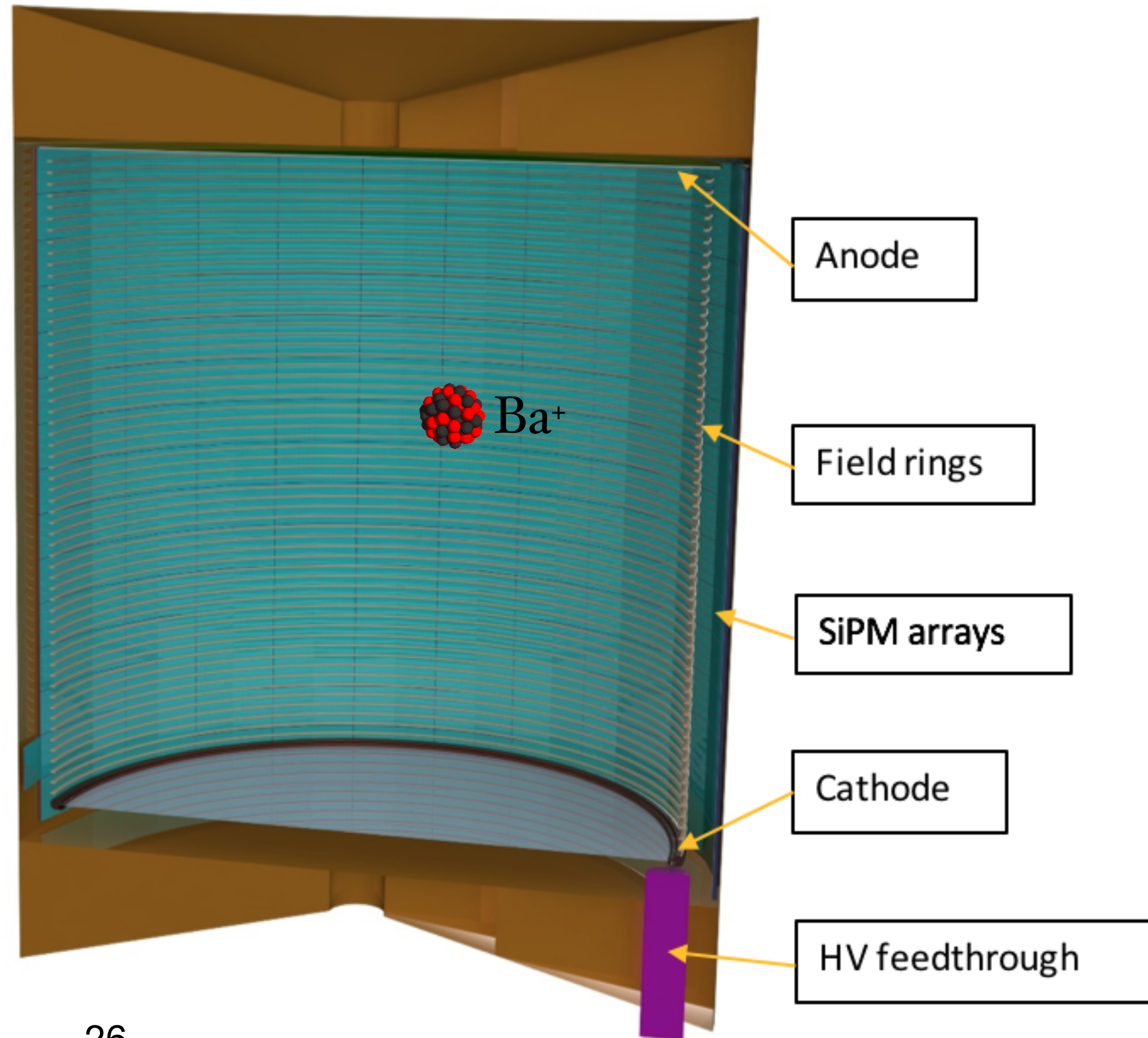


^[a] Phys. Rev. C 89, 015502 (2014) (EXO-200 $T_{2\nu\beta\beta}$ meas.)

Ba-Tagging in LXe

Concept

1. Localization^[a]
2. Extraction



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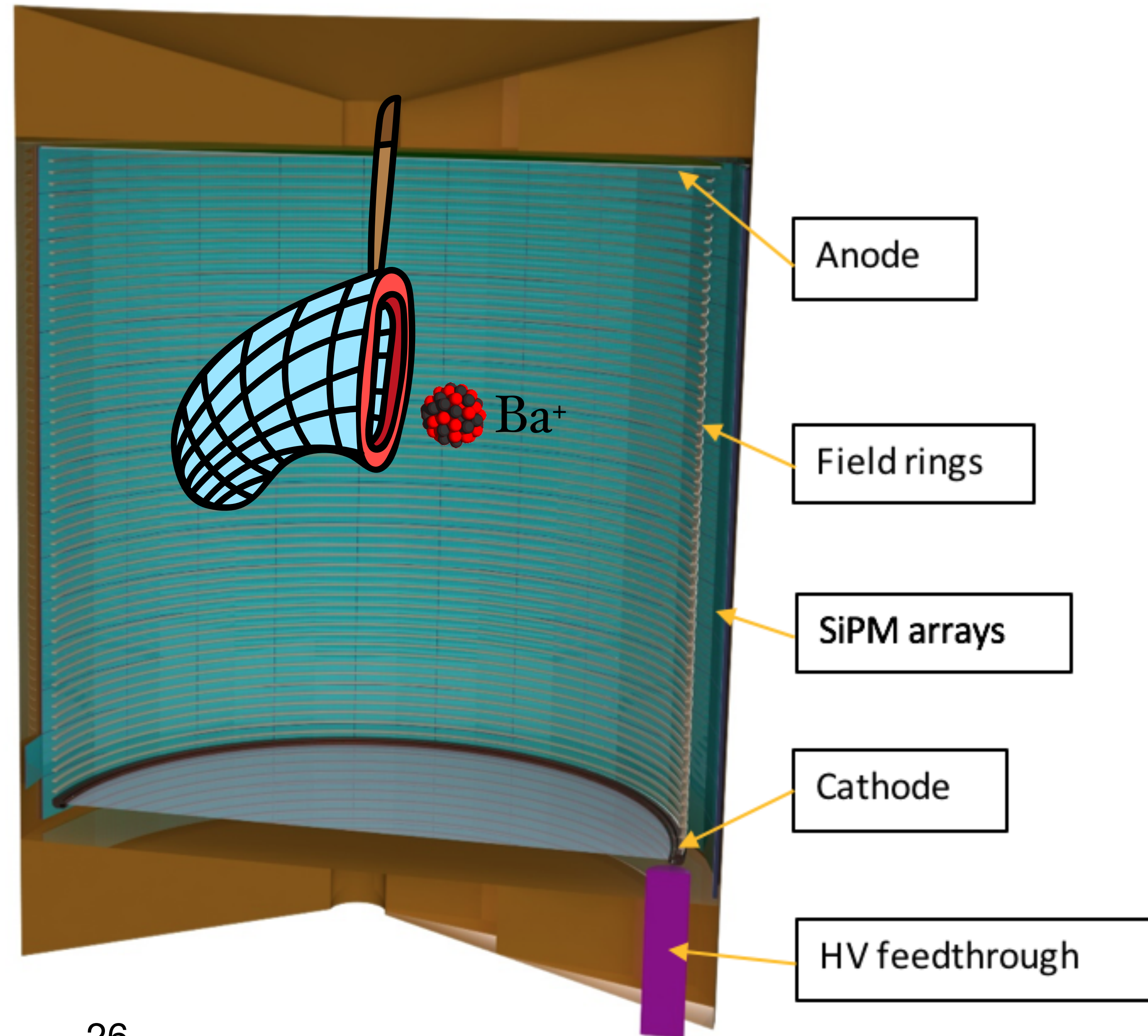
Ba-Tagging in LXe

Concept

1. Localization^[a]

2. Extraction

A. Cryoprobe

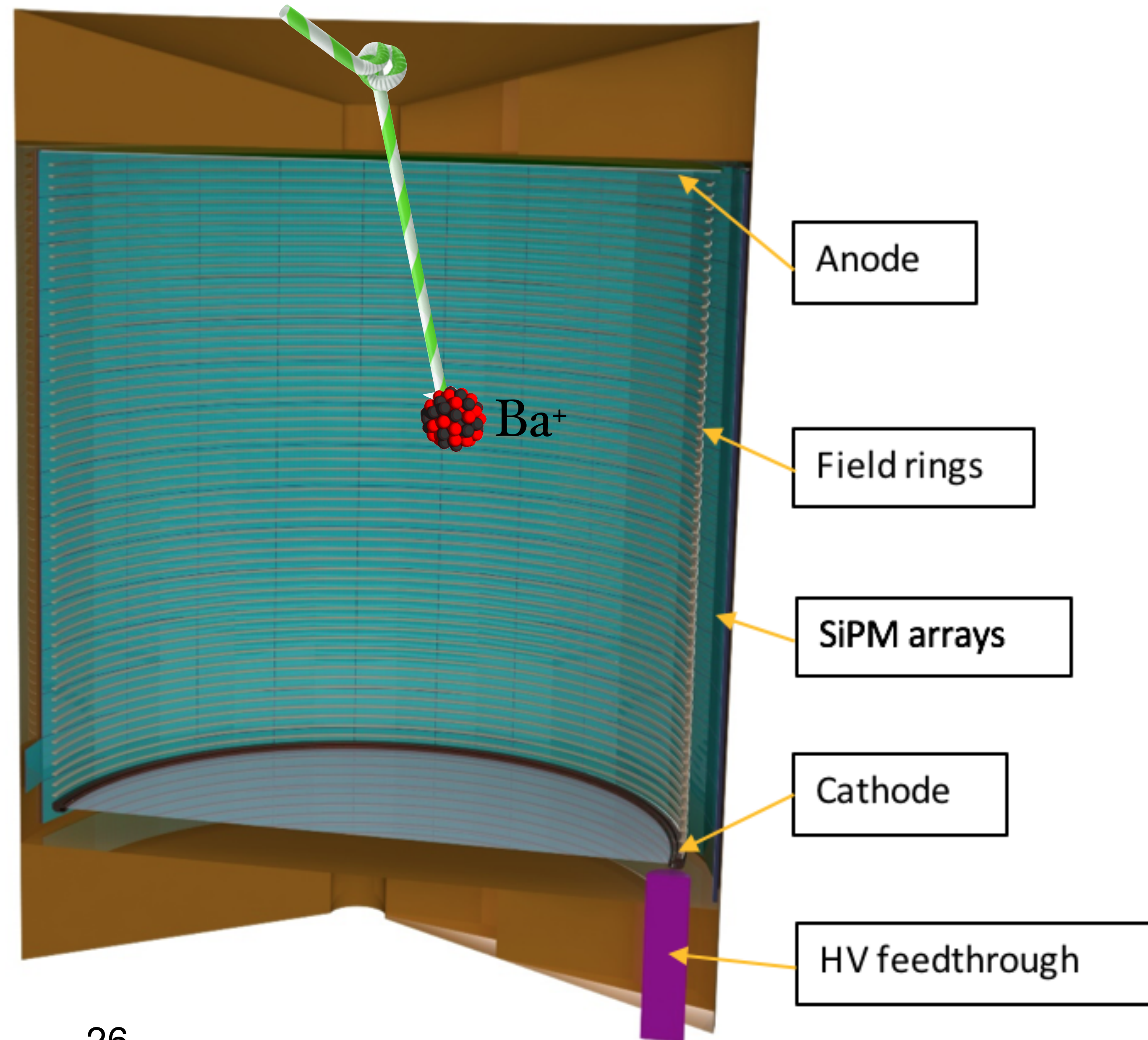


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Ba-Tagging in LXe

Concept

1. Localization^[a]
2. Extraction
 - A. Cryoprobe
 - B. Capillary tube

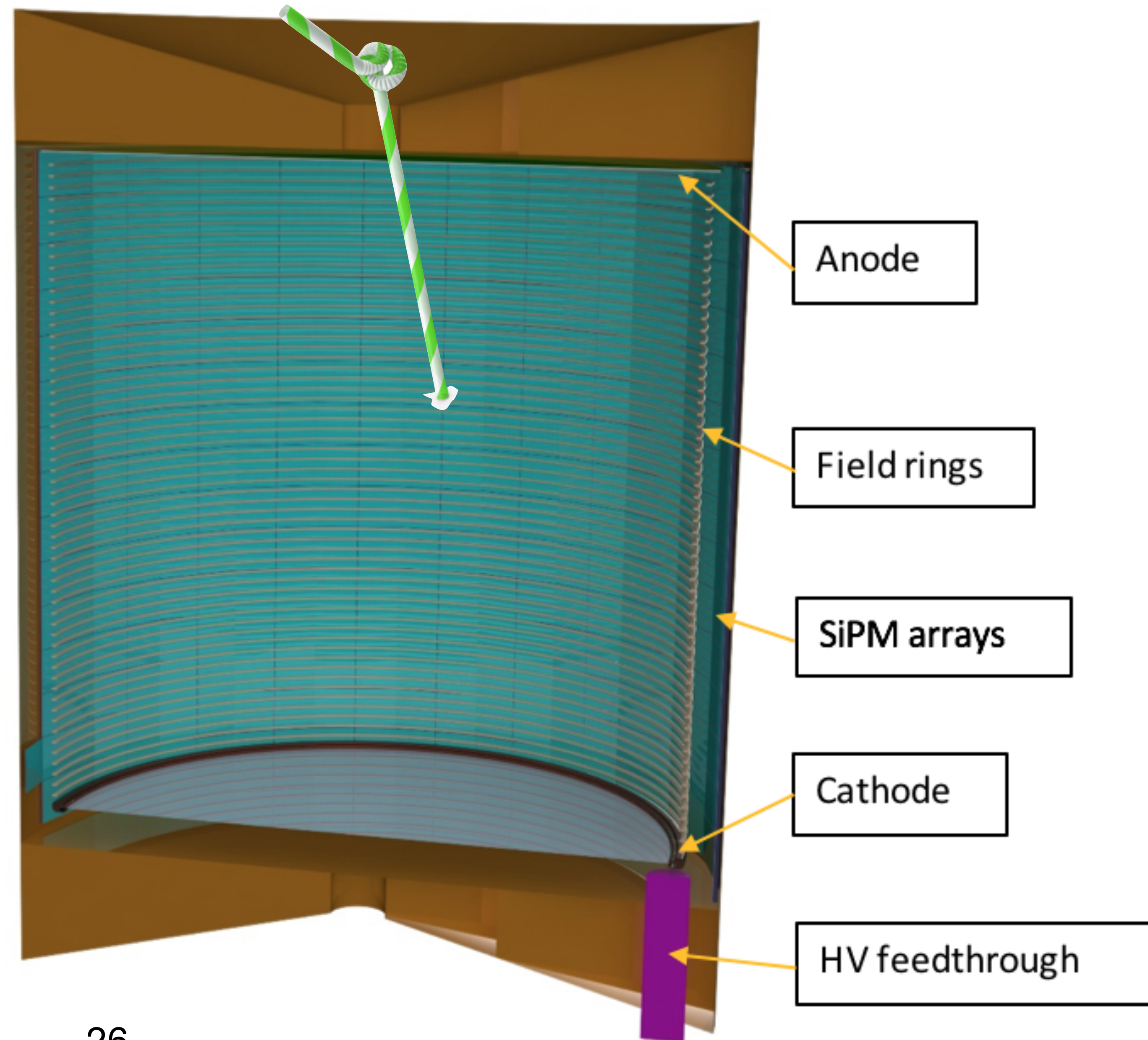


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Ba-Tagging in LXe

Concept

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3. (Separation)

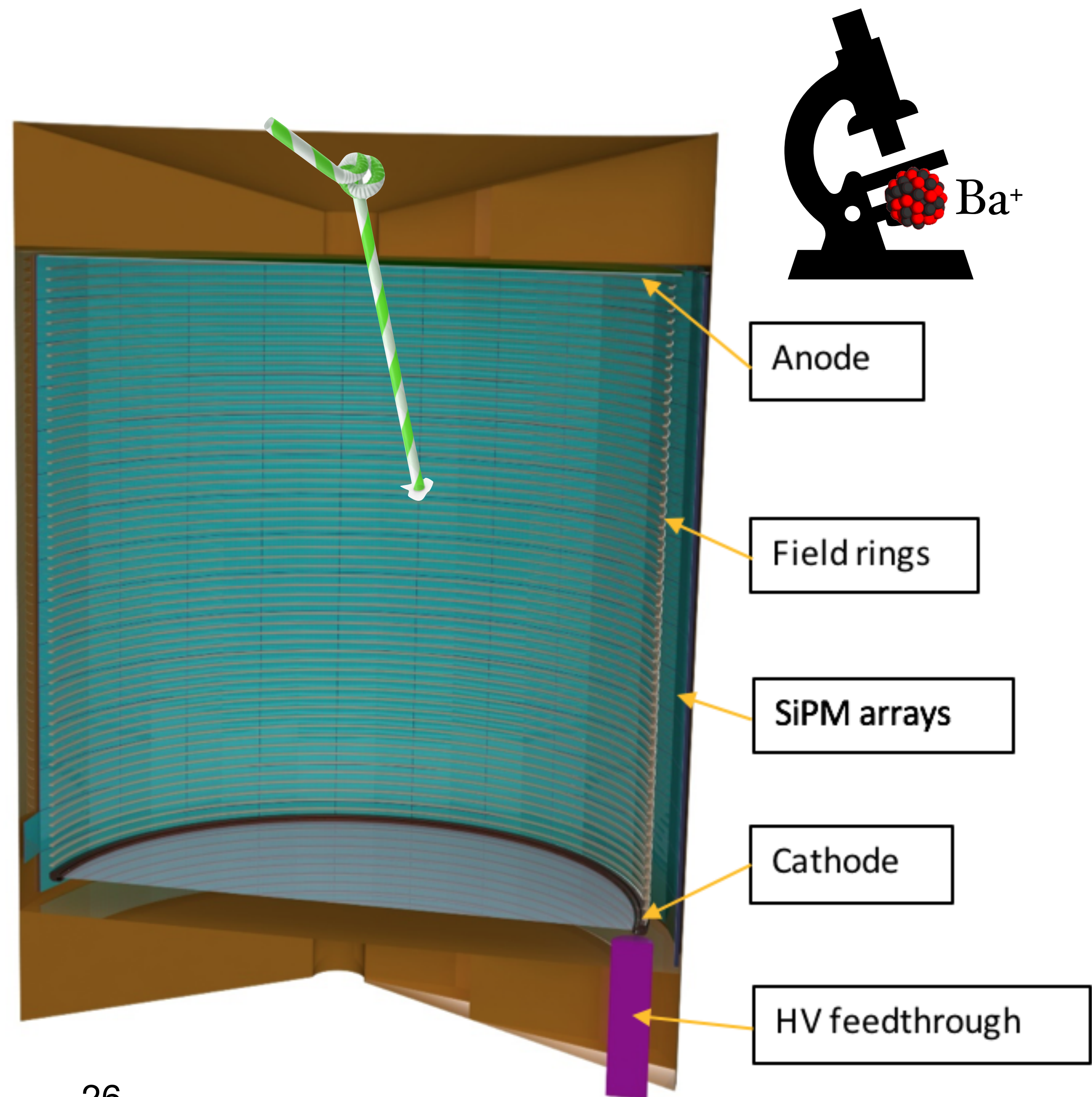


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Ba-Tagging in LXe

Concept

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4. Identification

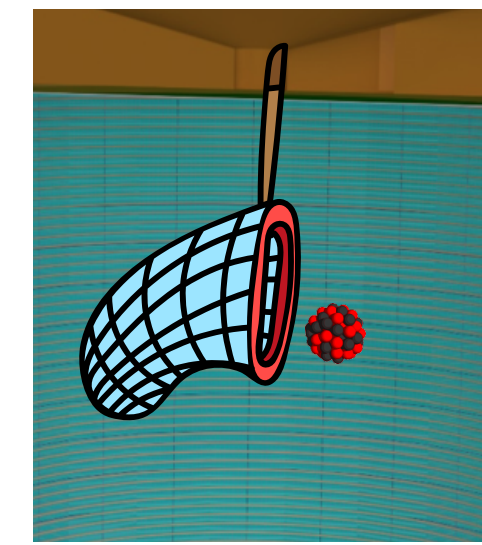


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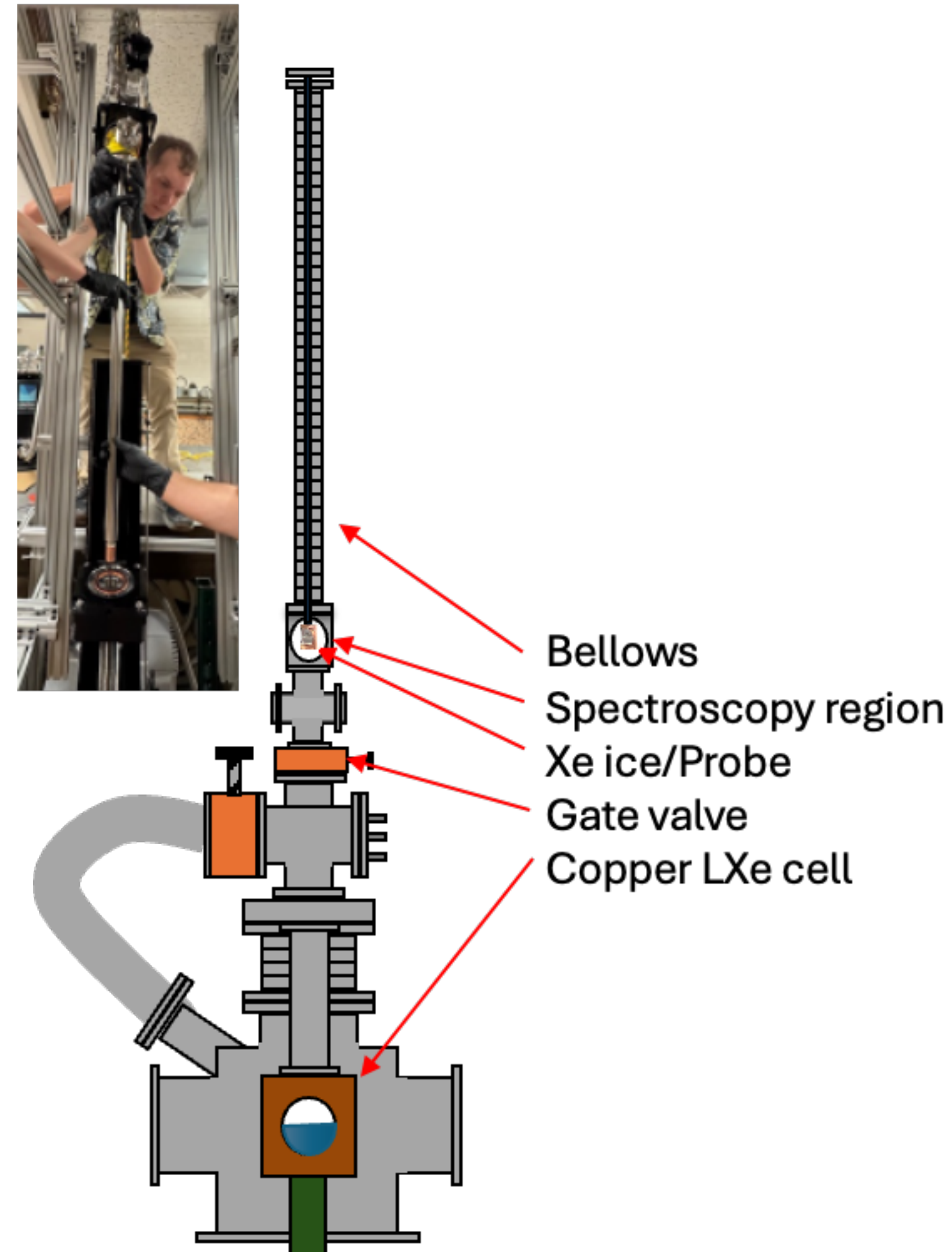
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Extraction with cryoprobe (cont.)

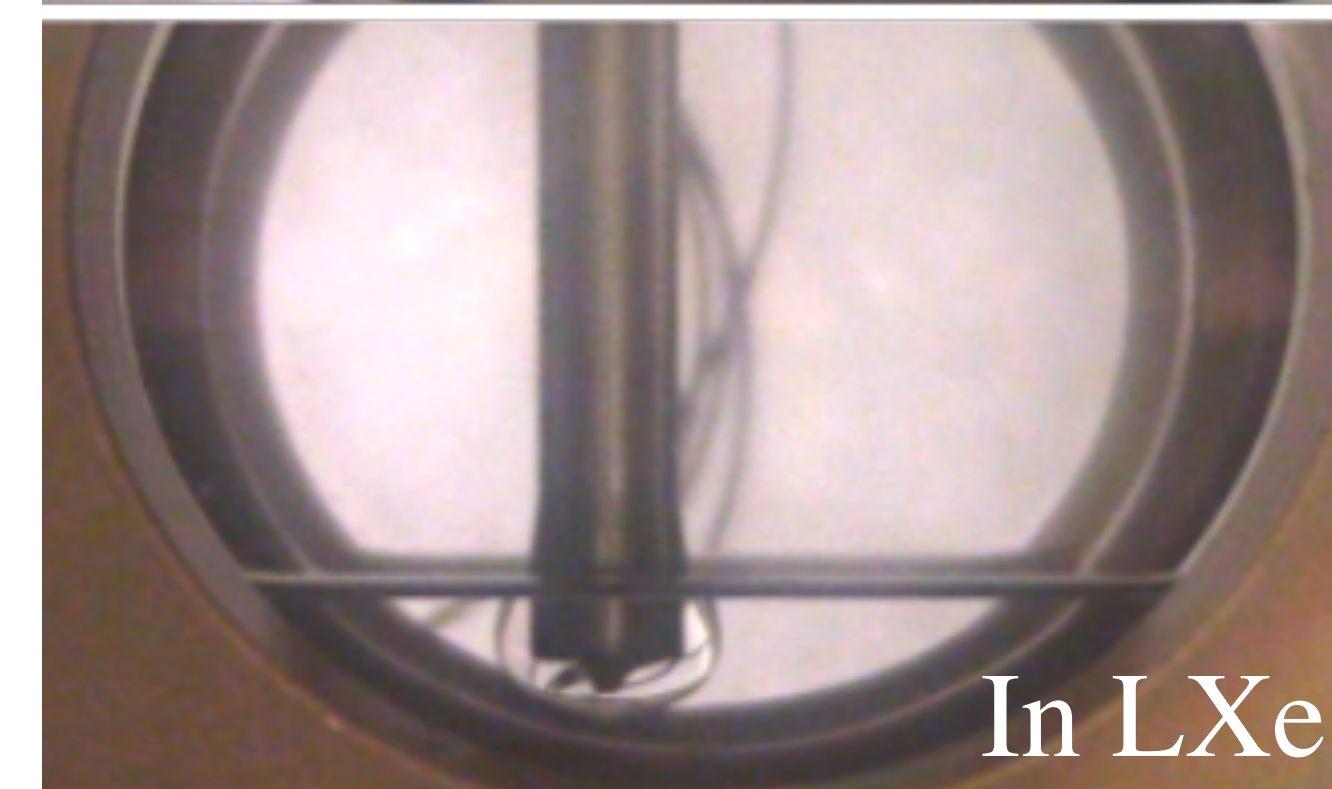
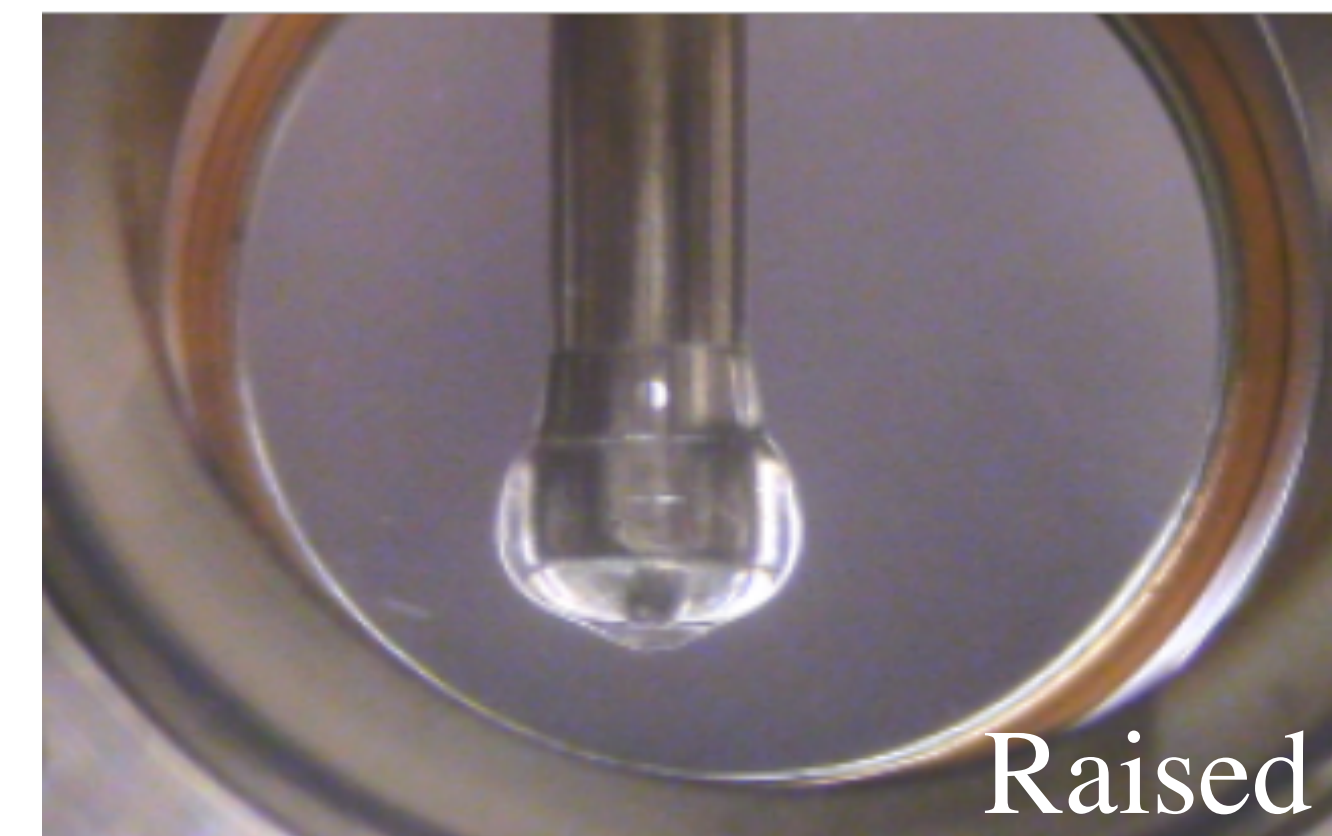
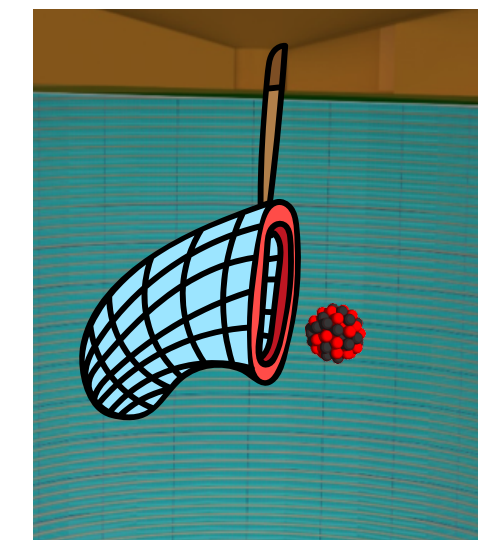
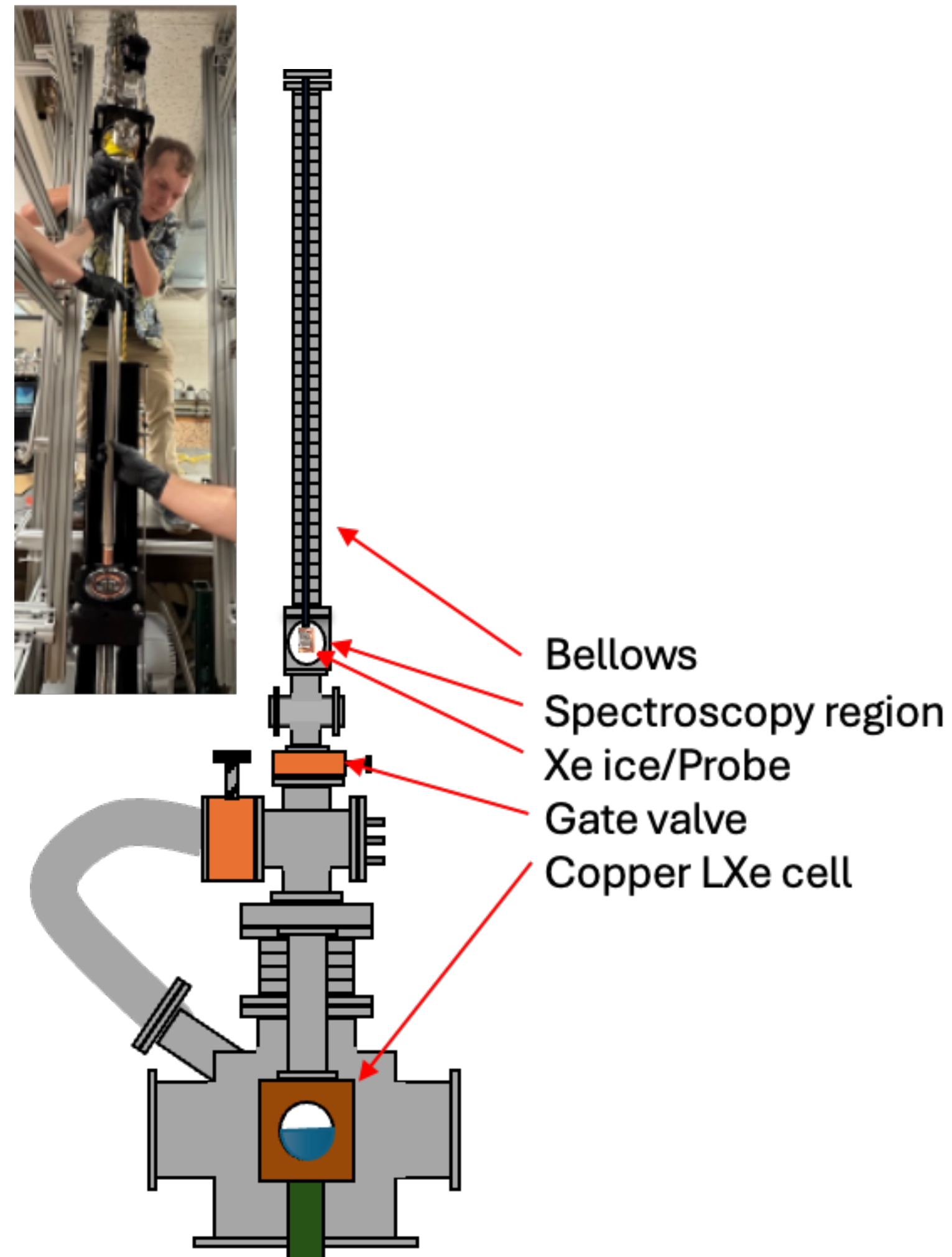


- Capture Ba in SXe from LXe at $>161\text{K}$.
- Proper SXe ice is transparent, not frosty or cracked
- After gate valve, reduce probe T and $p \rightarrow 10\text{-}40\text{ K}$ for single Ba/Ba⁺ imaging.

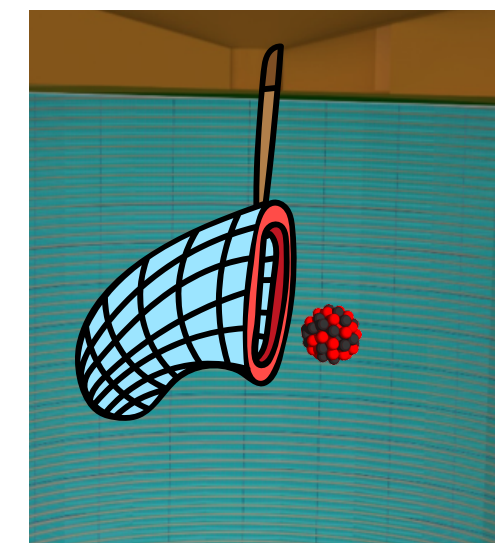


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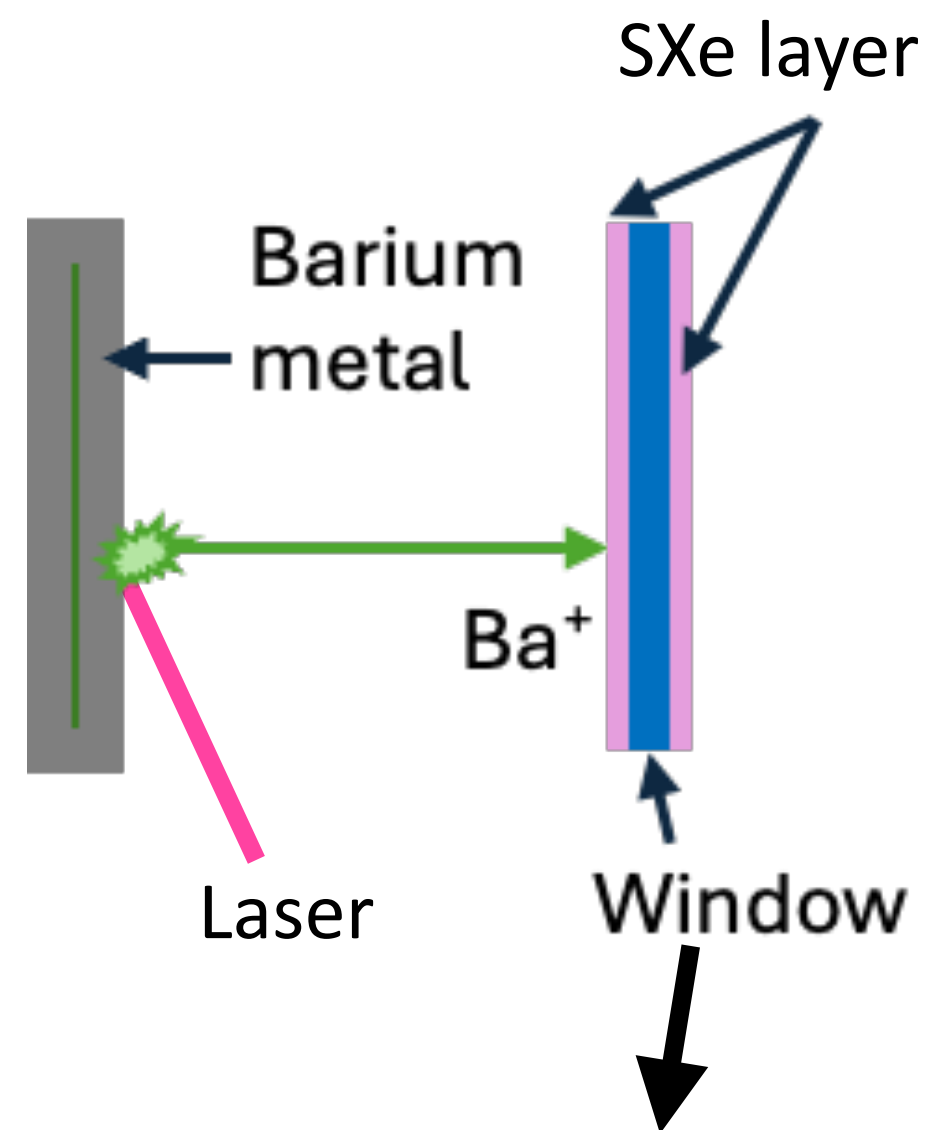


Extraction with cryoprobe (cont.)



Previous work:

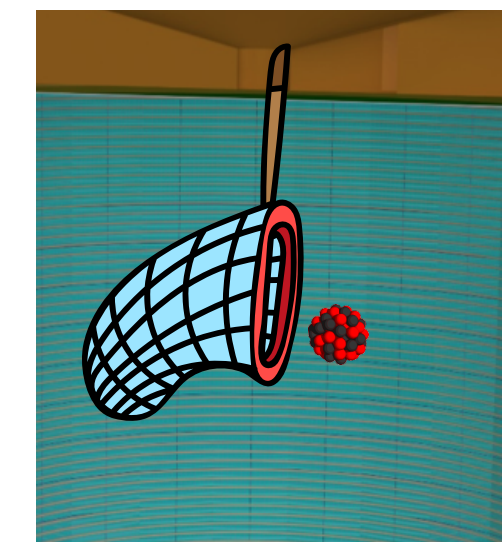
- Laser desorption in **GXe**
- Ba fluorescence at 160 K
- Expanding LN₂/Ar for cryoprobe



Barium metal →

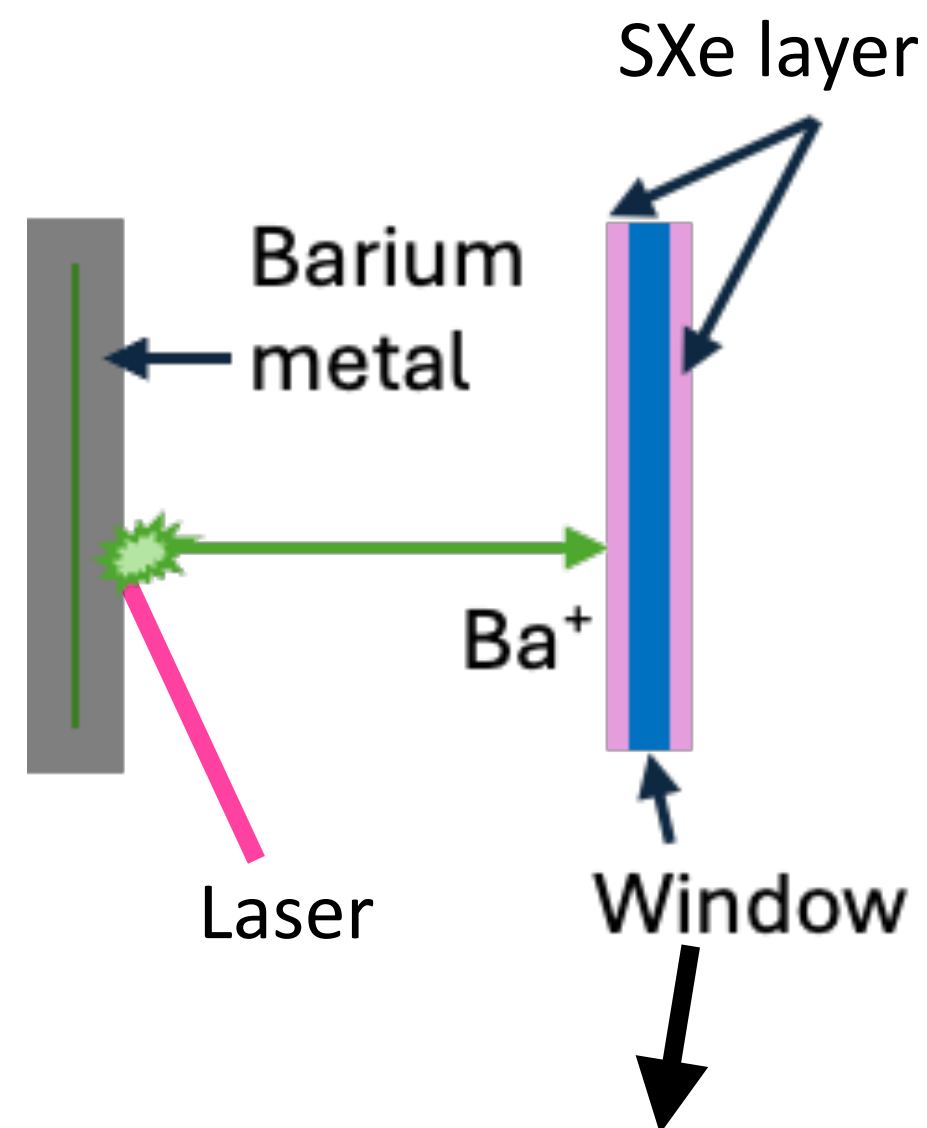


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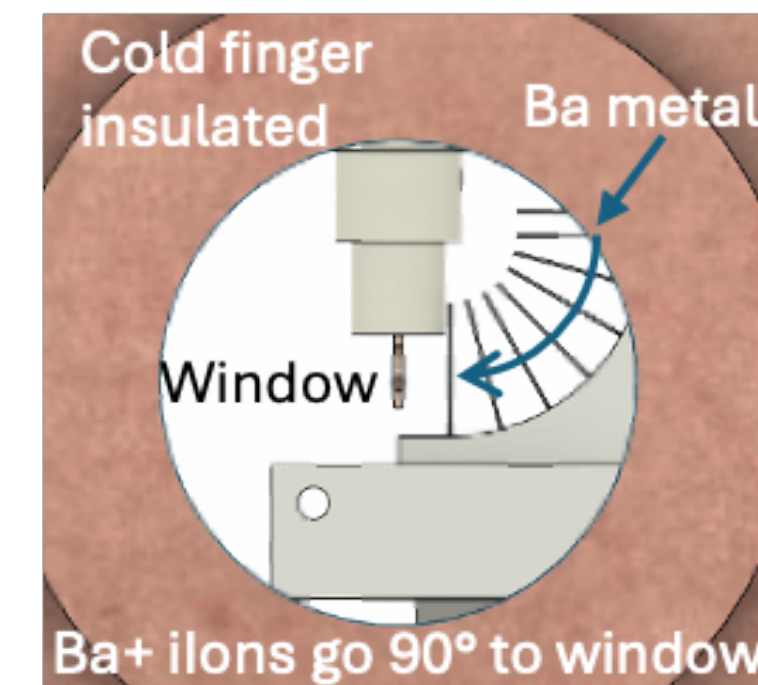


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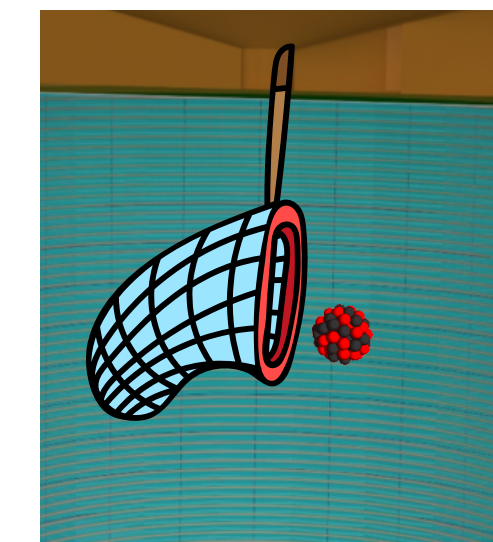


Latest upgrades:

- Electrodes to guide Ba⁺ in **LXe** to SXe window
- Commercial He cryoprobe → 10-40 K

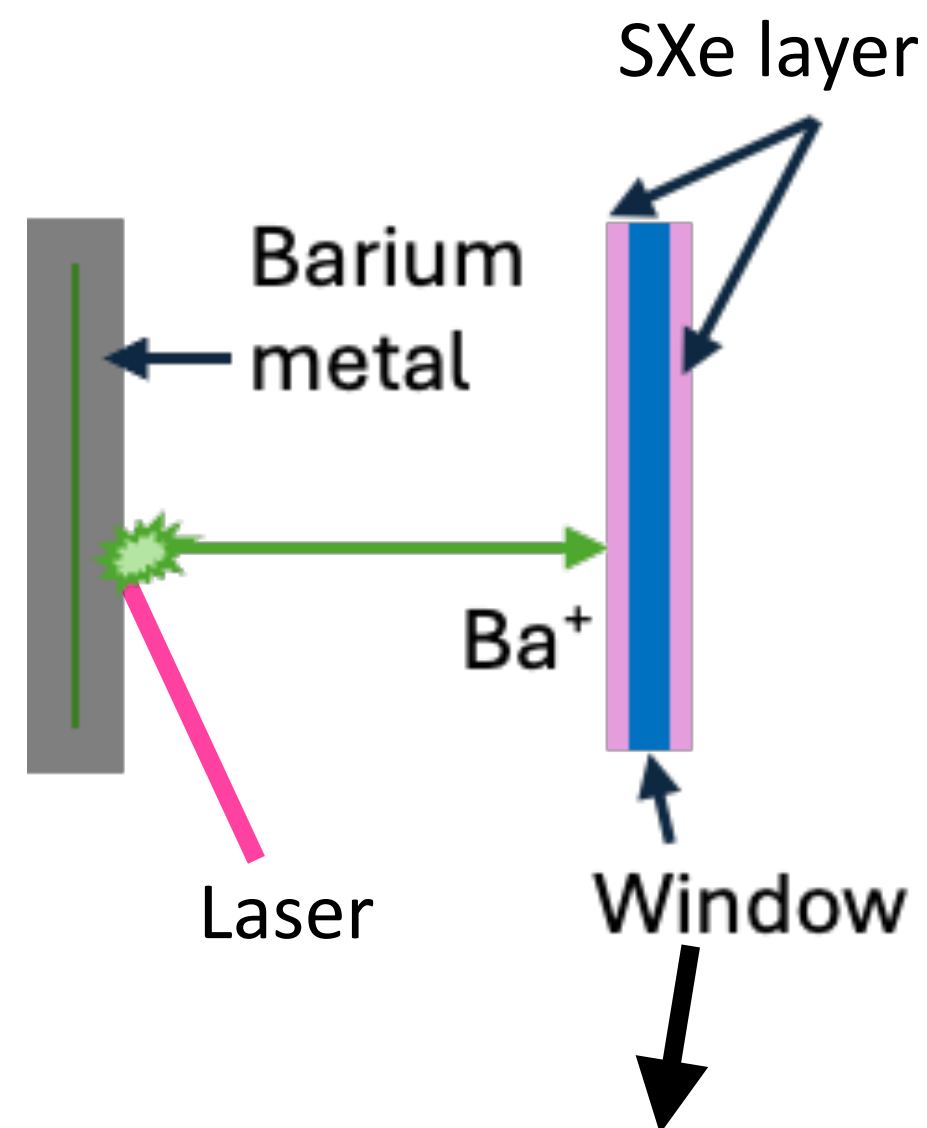


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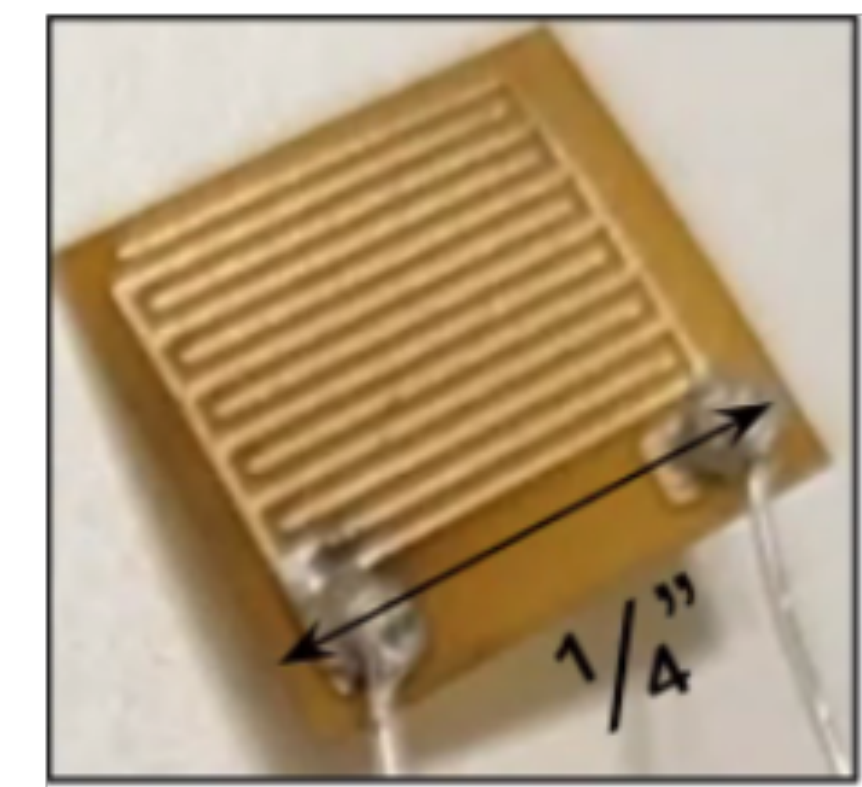
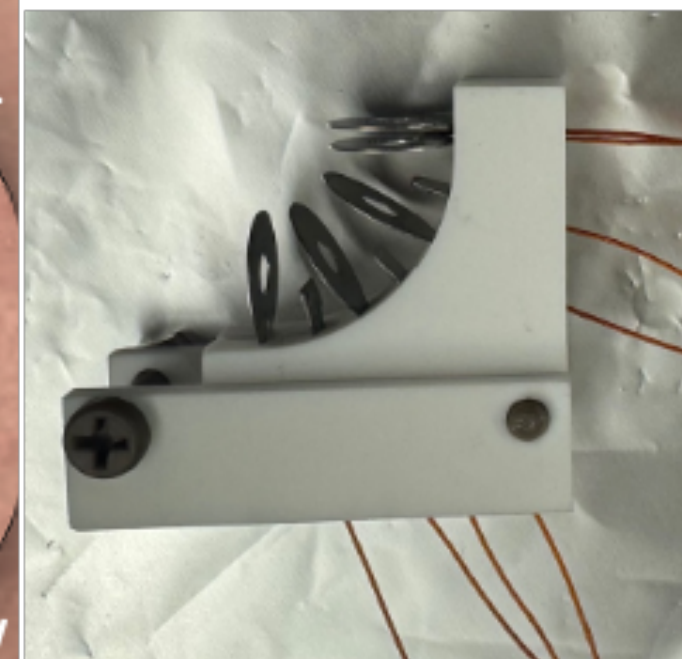
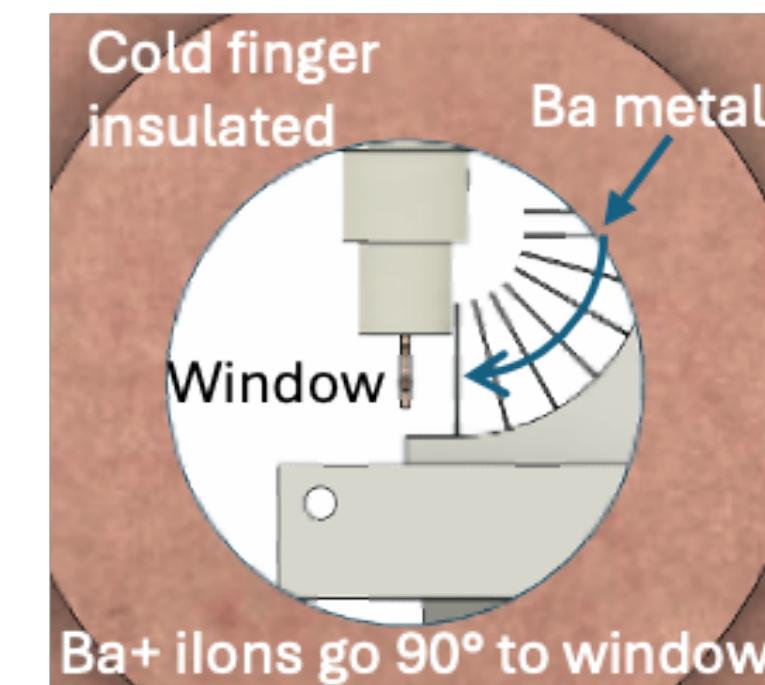


Barium metal →



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- Commercial He cryoprobe → 10-40 K
- Thickness control of SXe: ~10's μm

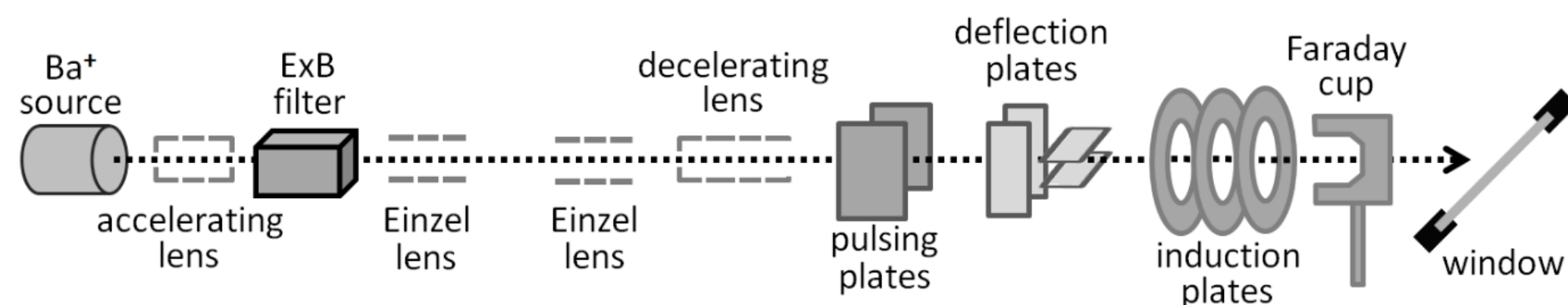
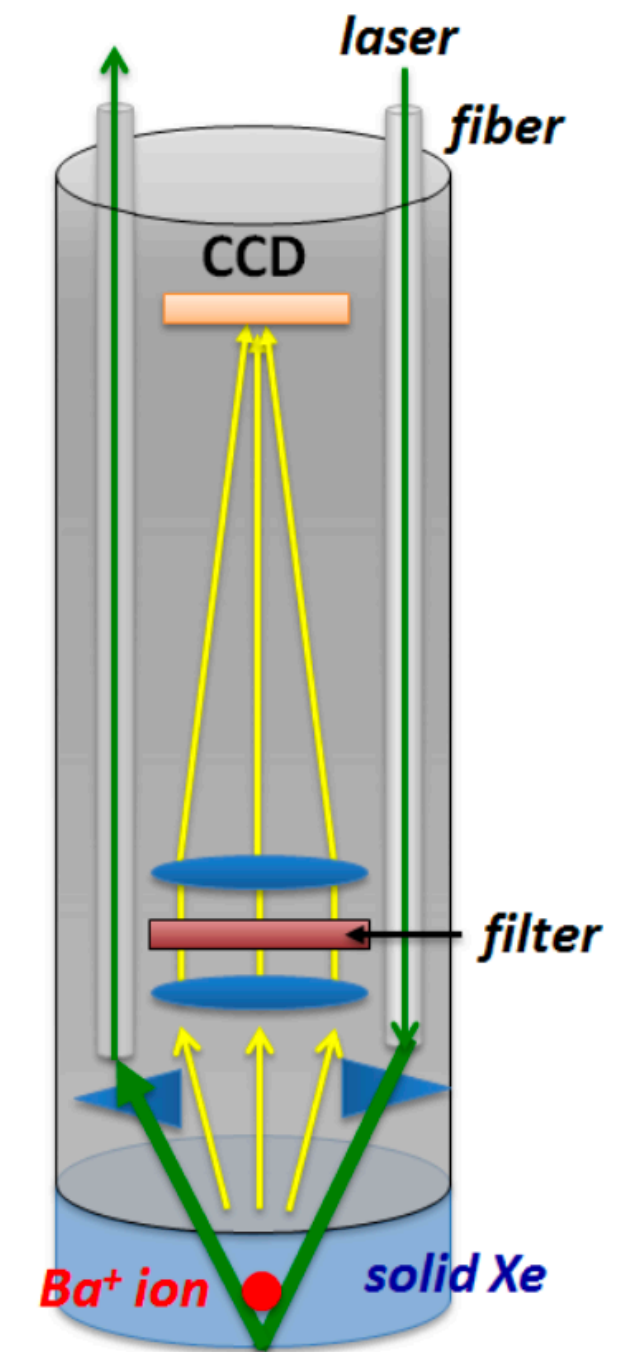
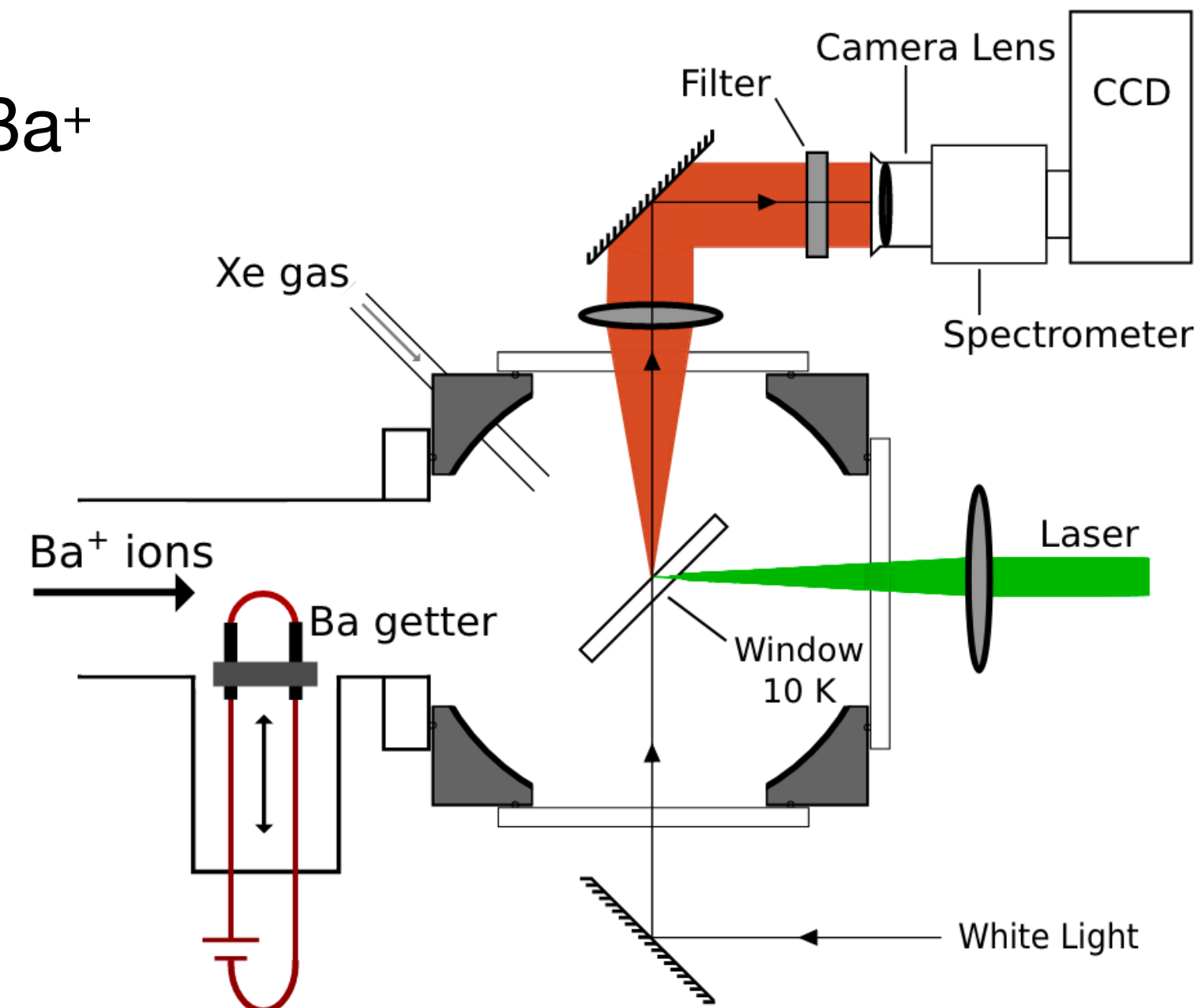
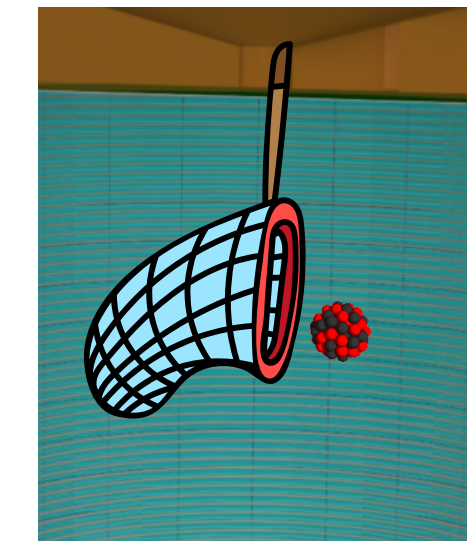


Flexible Capacitance sensor
measures SXe thickness UCSD

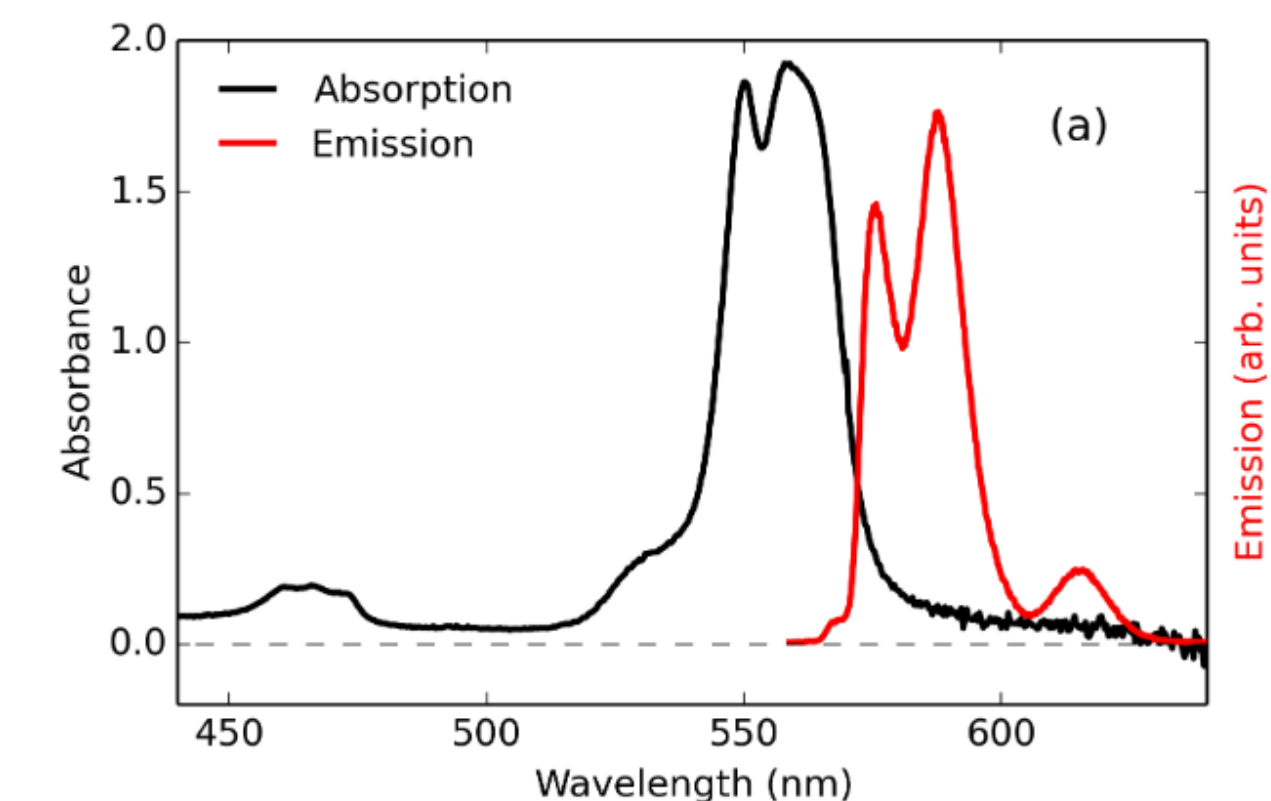
Implantation of Ba⁺ on SXe

First laser spectroscopy results

- Absorption and fluorescence spectra of Ba/Ba⁺ in SXe and SAr.
- Ba⁺ ion source and neutral Ba from getter.
- Multiple emission bands corresponding to different matrix sites and partial Ba⁺ neutralization
- Fluorescence images of $\approx 10^4$ Ba atoms



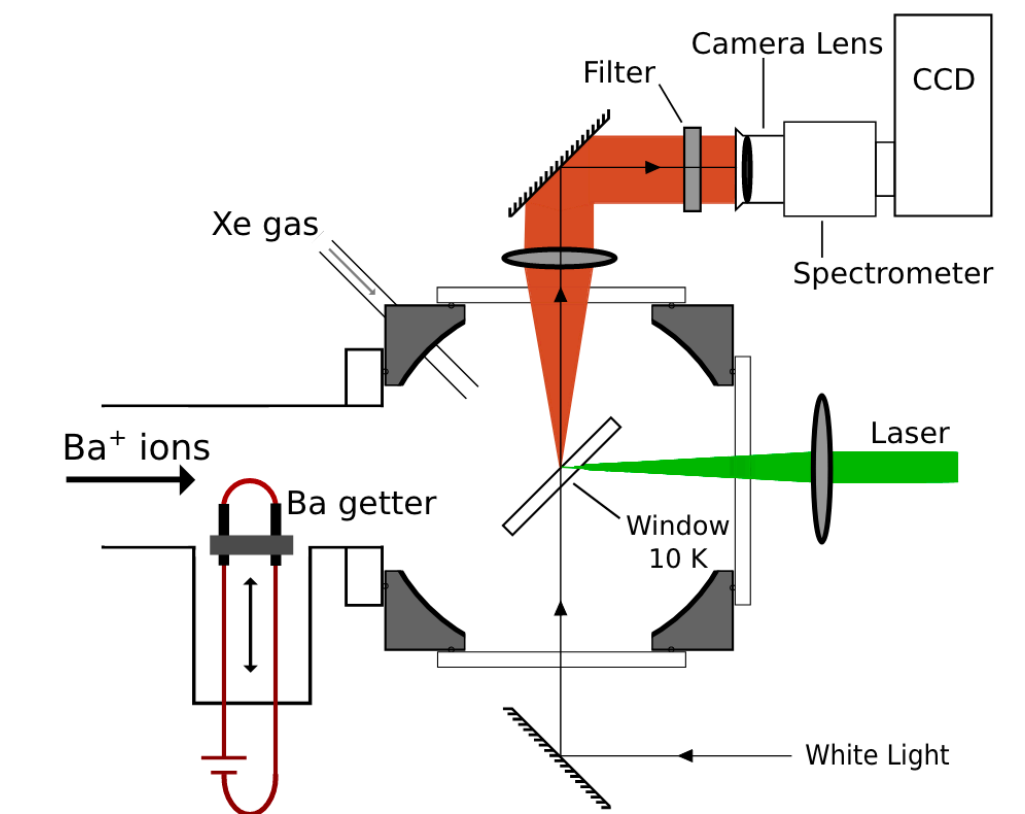
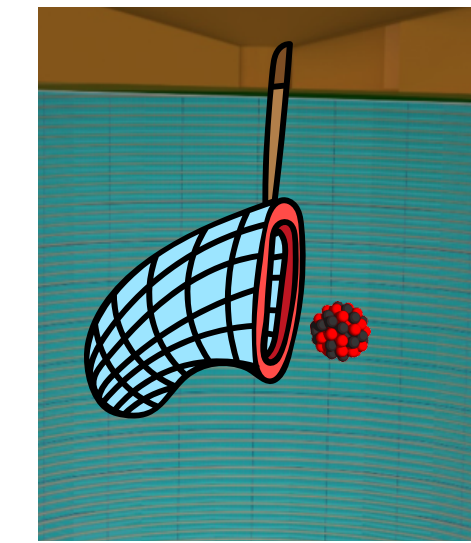
Absorption/emission spectra of Ba in SXe



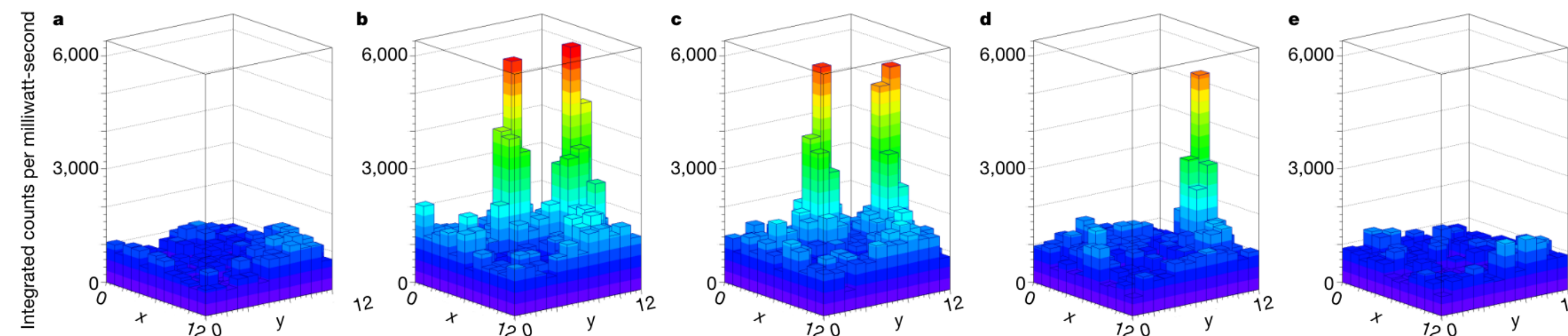
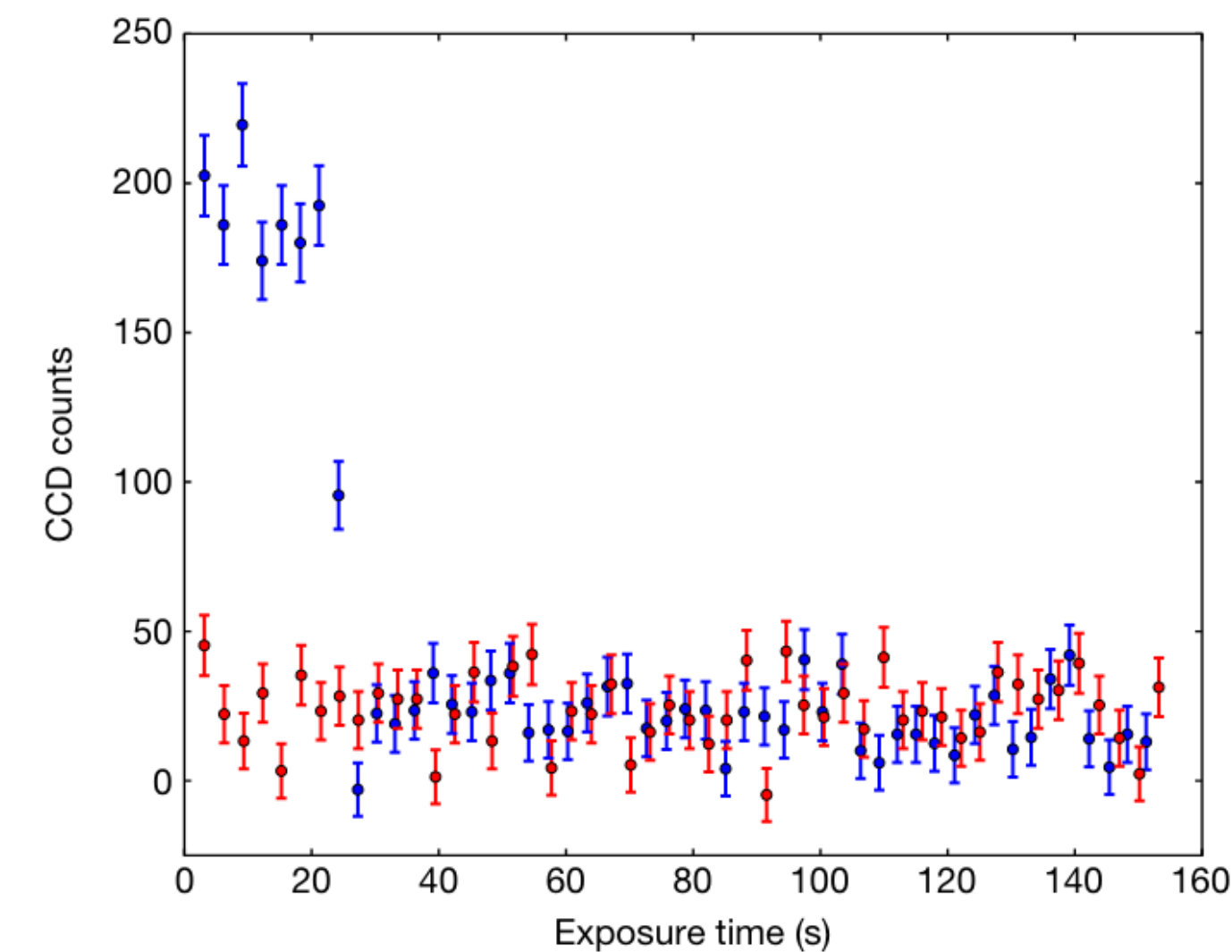
Single-ion detection in SXe

Matrix Vacancy study

- Extended result of Mong et al. at the single Ba level (observed single photobleaching step)
- Corresponding to single-vacancy Ba in SXe matrix.
- Different behavior of the Tetra- and Hexa-Vacancy.
- Annealing causes fluorescence recovery (anti-bleaching) at the HV site (577 nm)



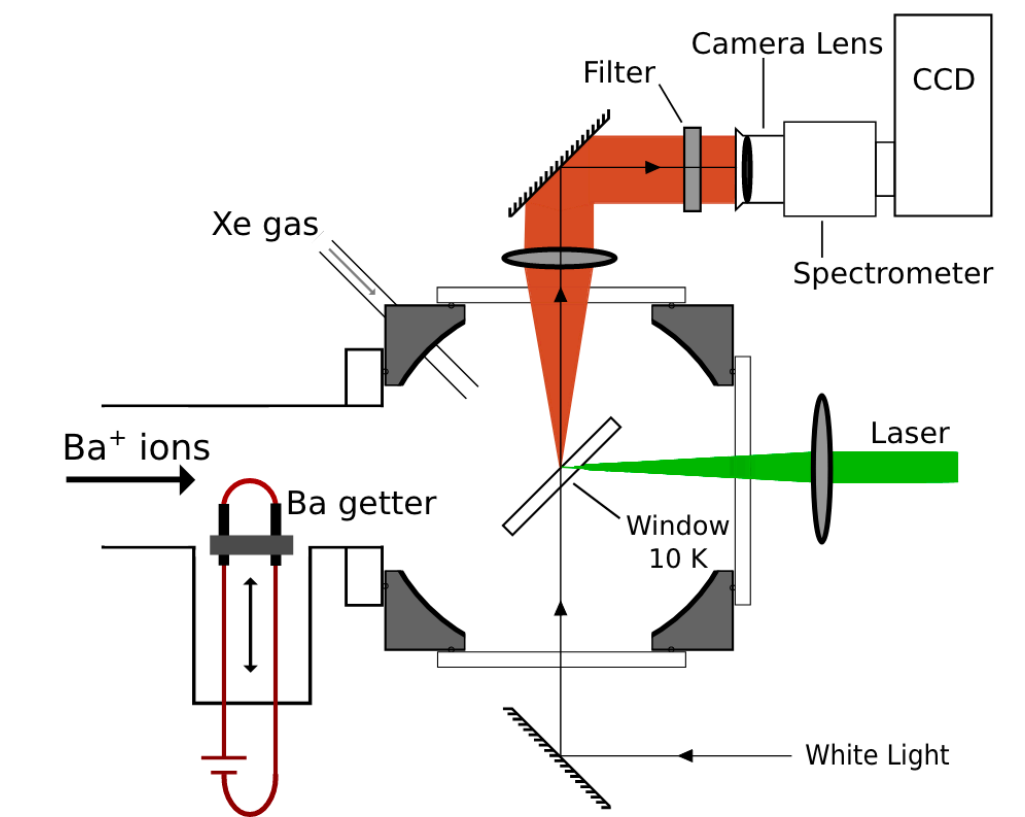
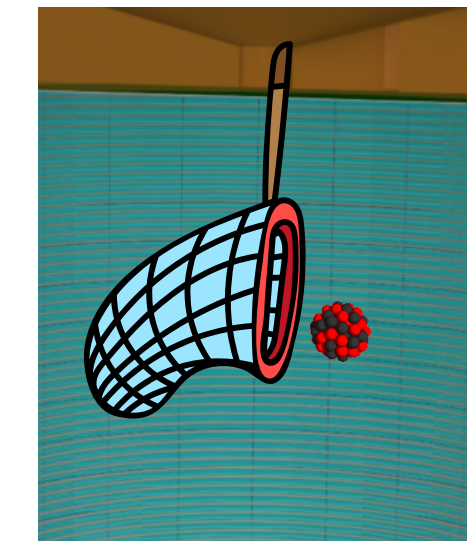
Single-steps of Ba in SXe



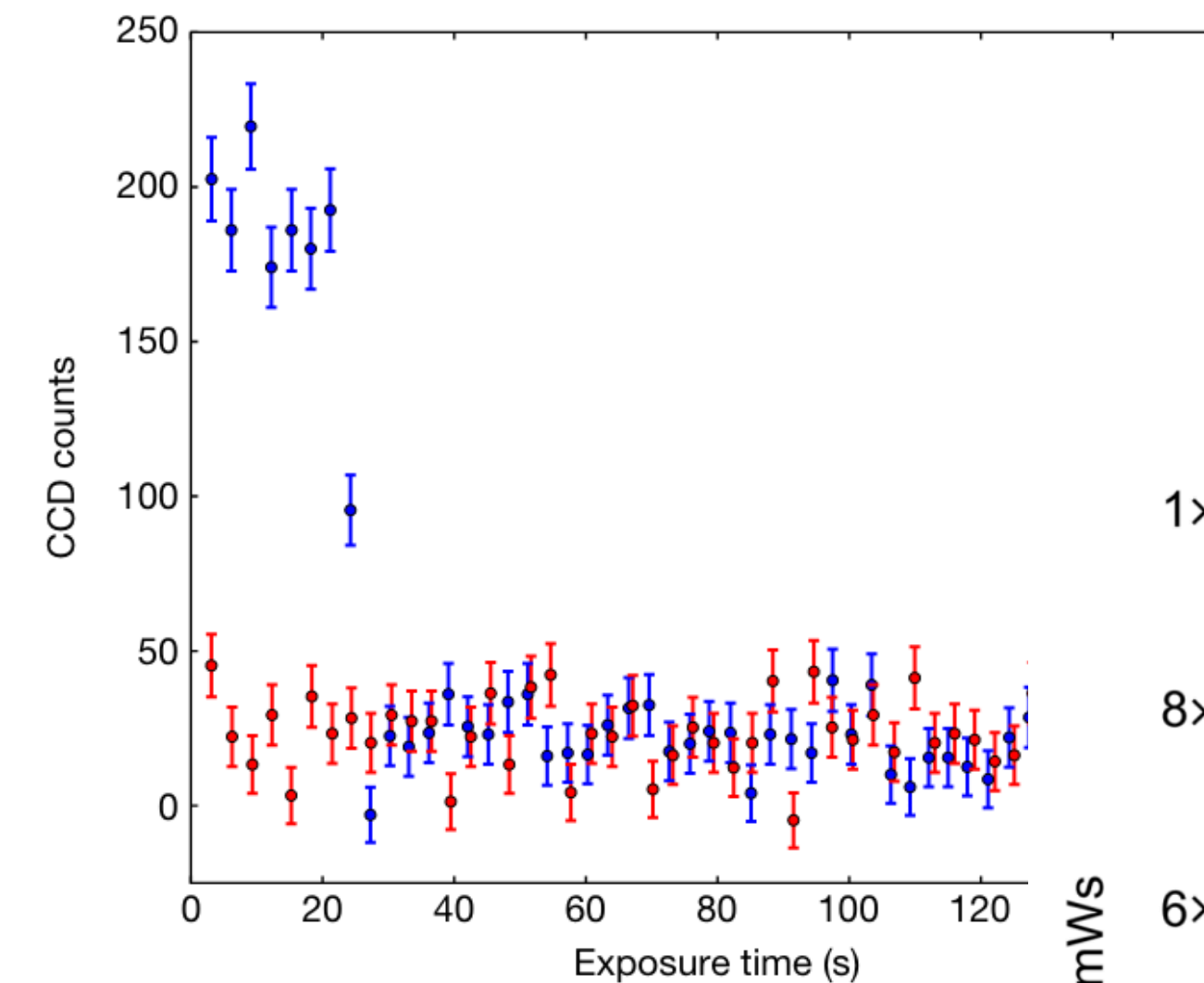
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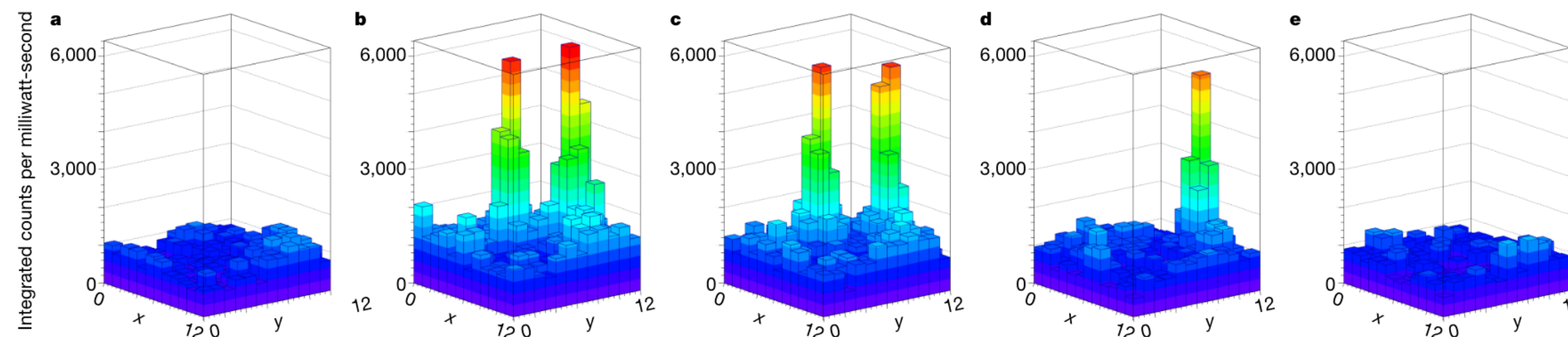
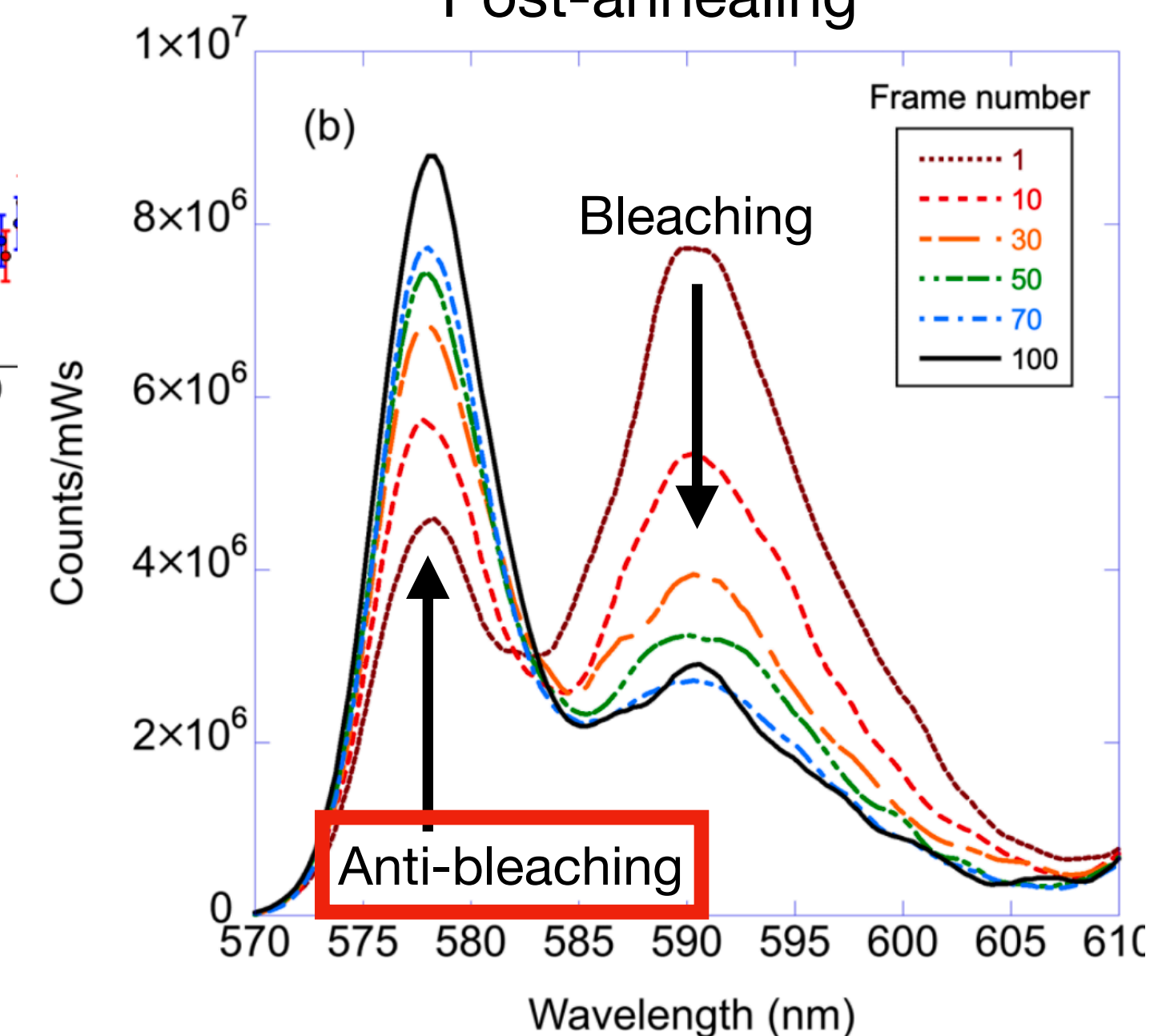
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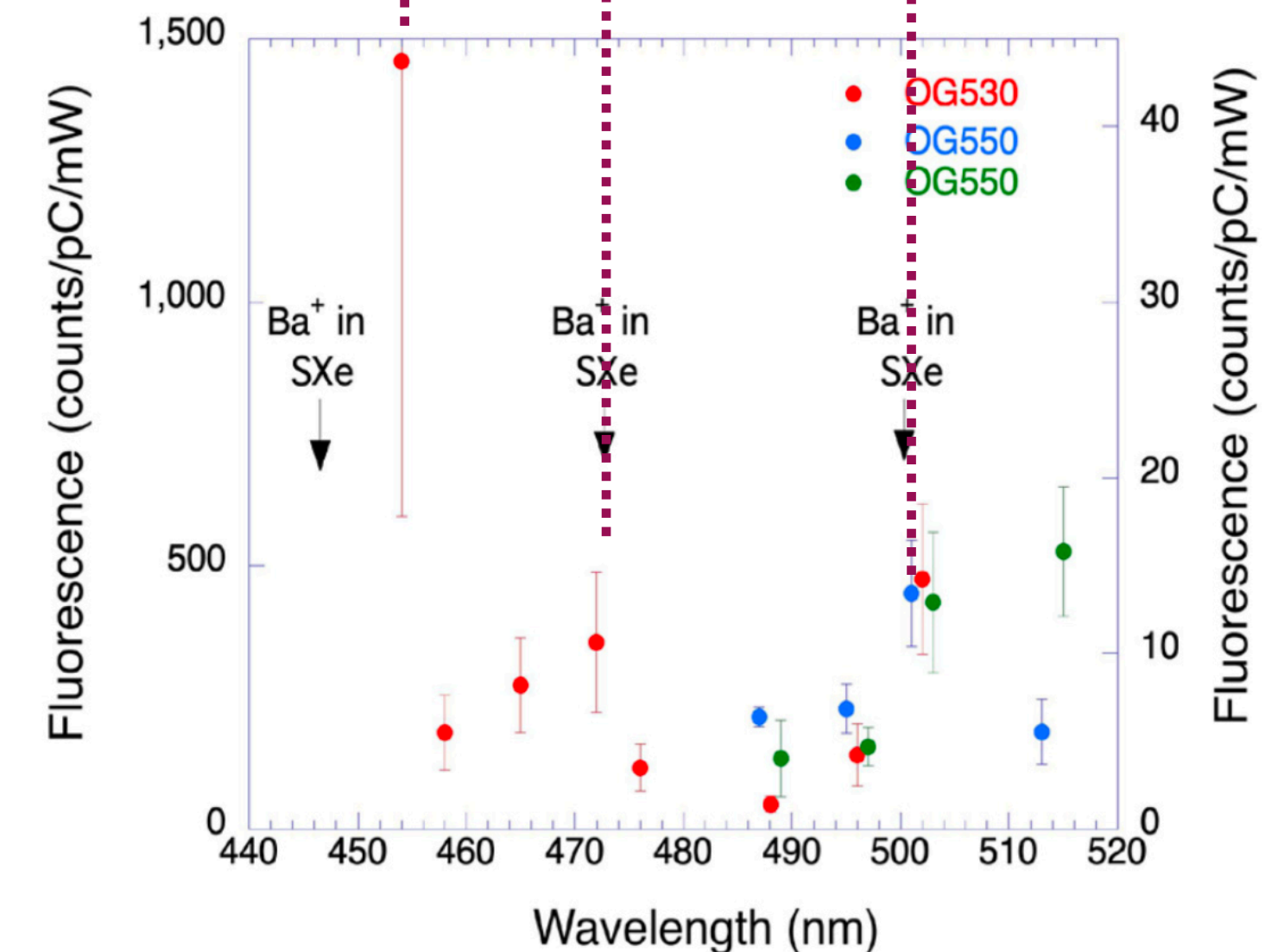
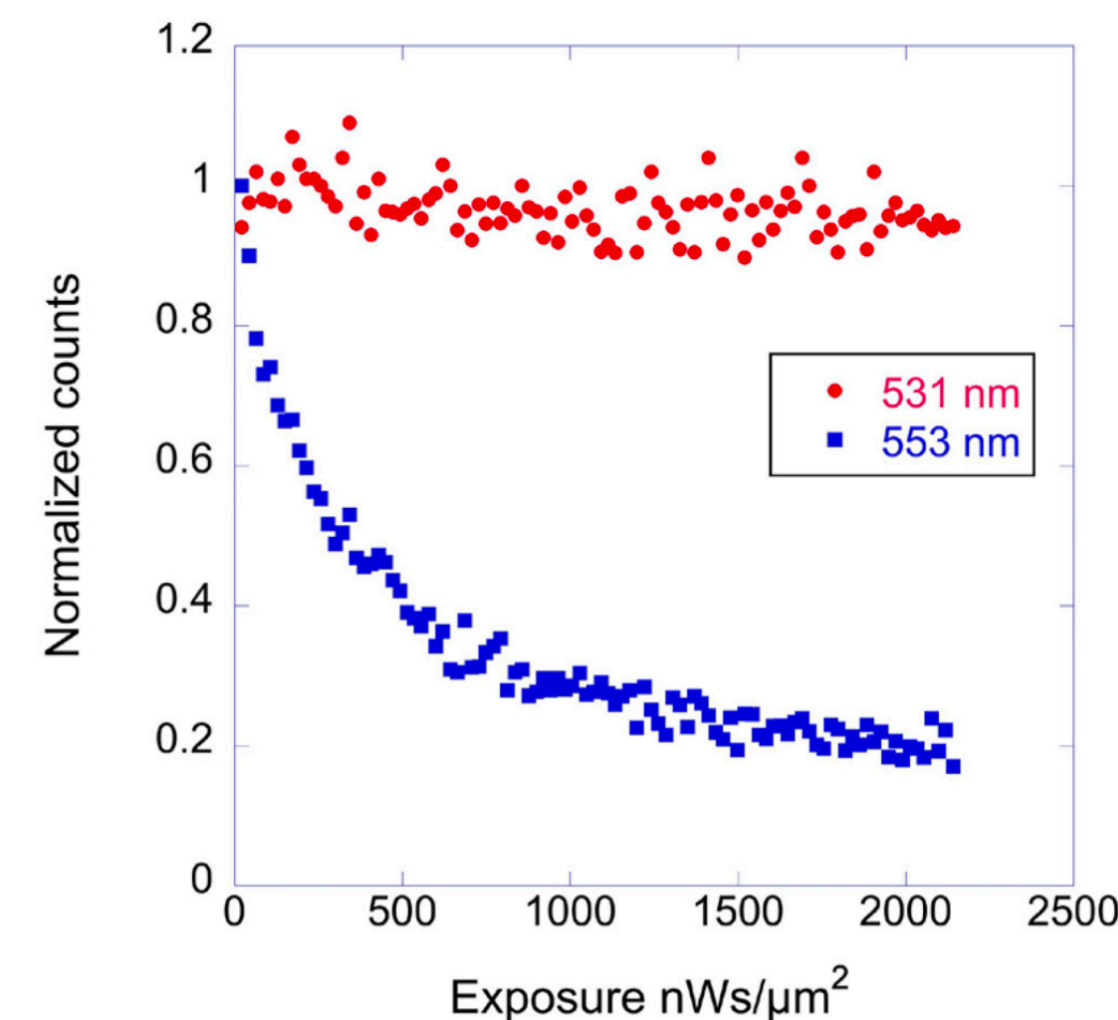
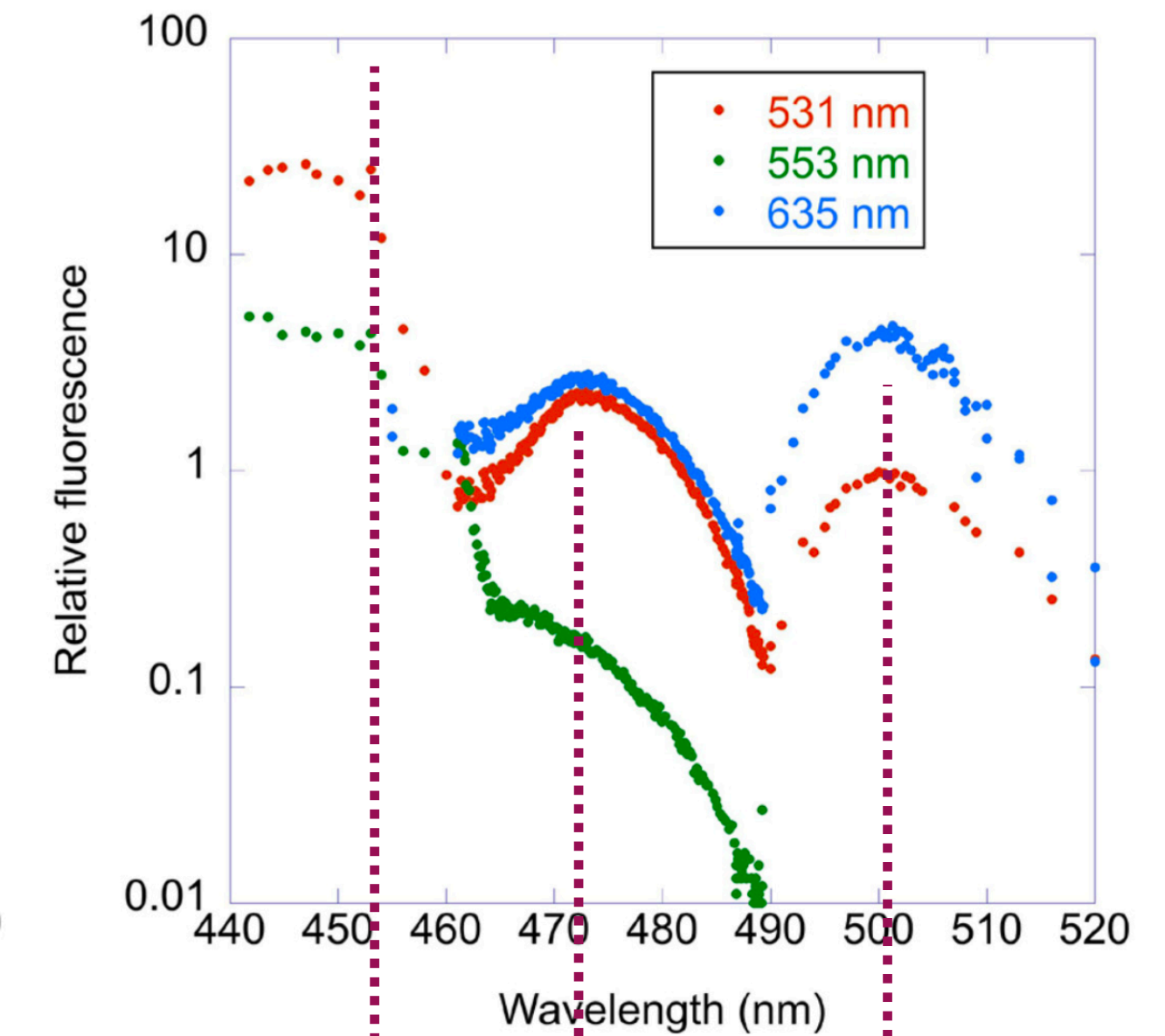
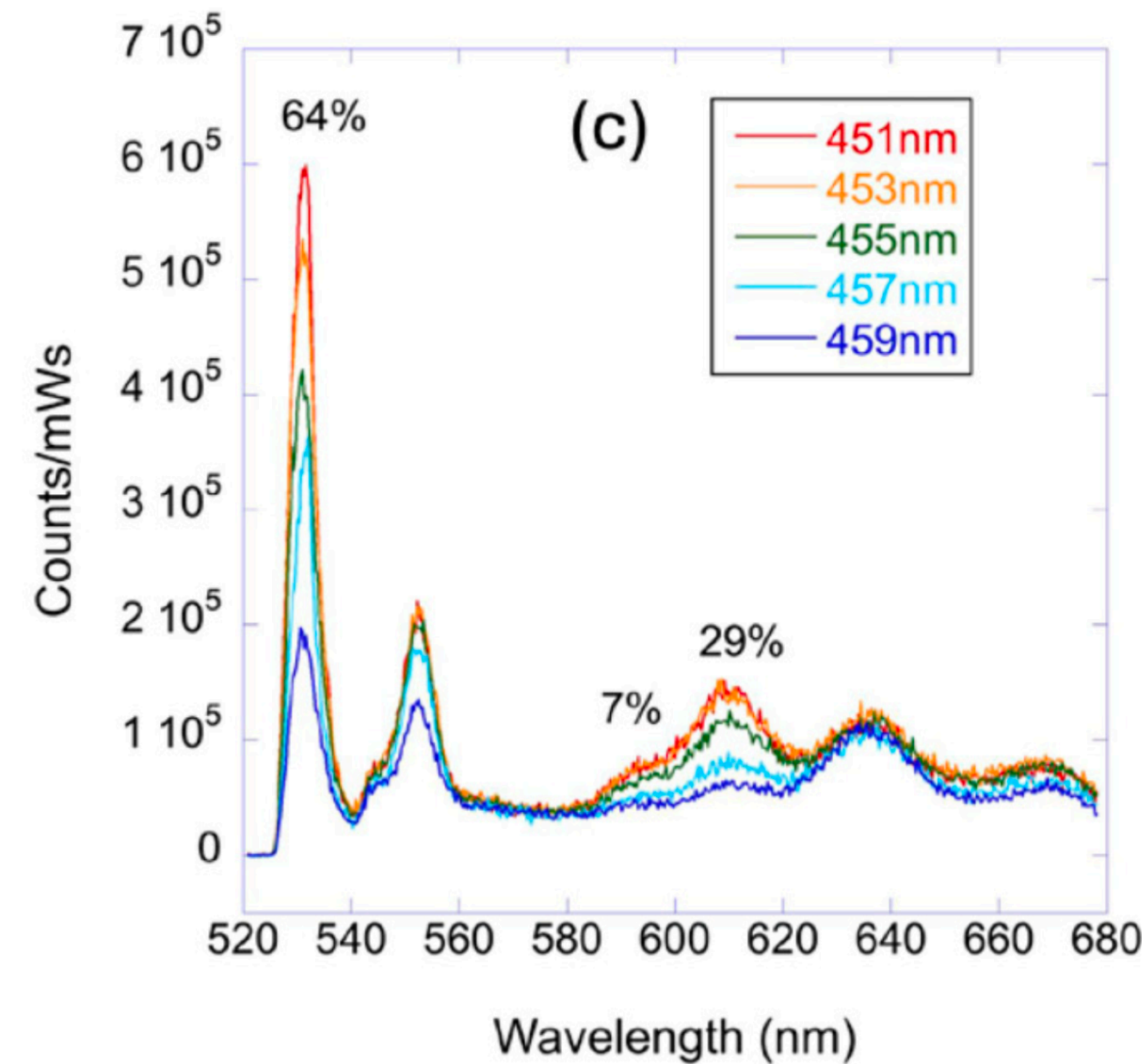


Post-annealing



Complete spectroscopy of Ba⁺ in SXe

- 3 excitation bands (446, 473, 500 nm) and 2 emission groups tied to different SXe sites.
- Moderate bleaching, high fluorescence → feasible for single-ion imaging
- Thick SXe layers cut surface background by ×10
- Spectra match Ba⁺ in liquid Xe, confirming Ba⁺ tagging viability for nEXO

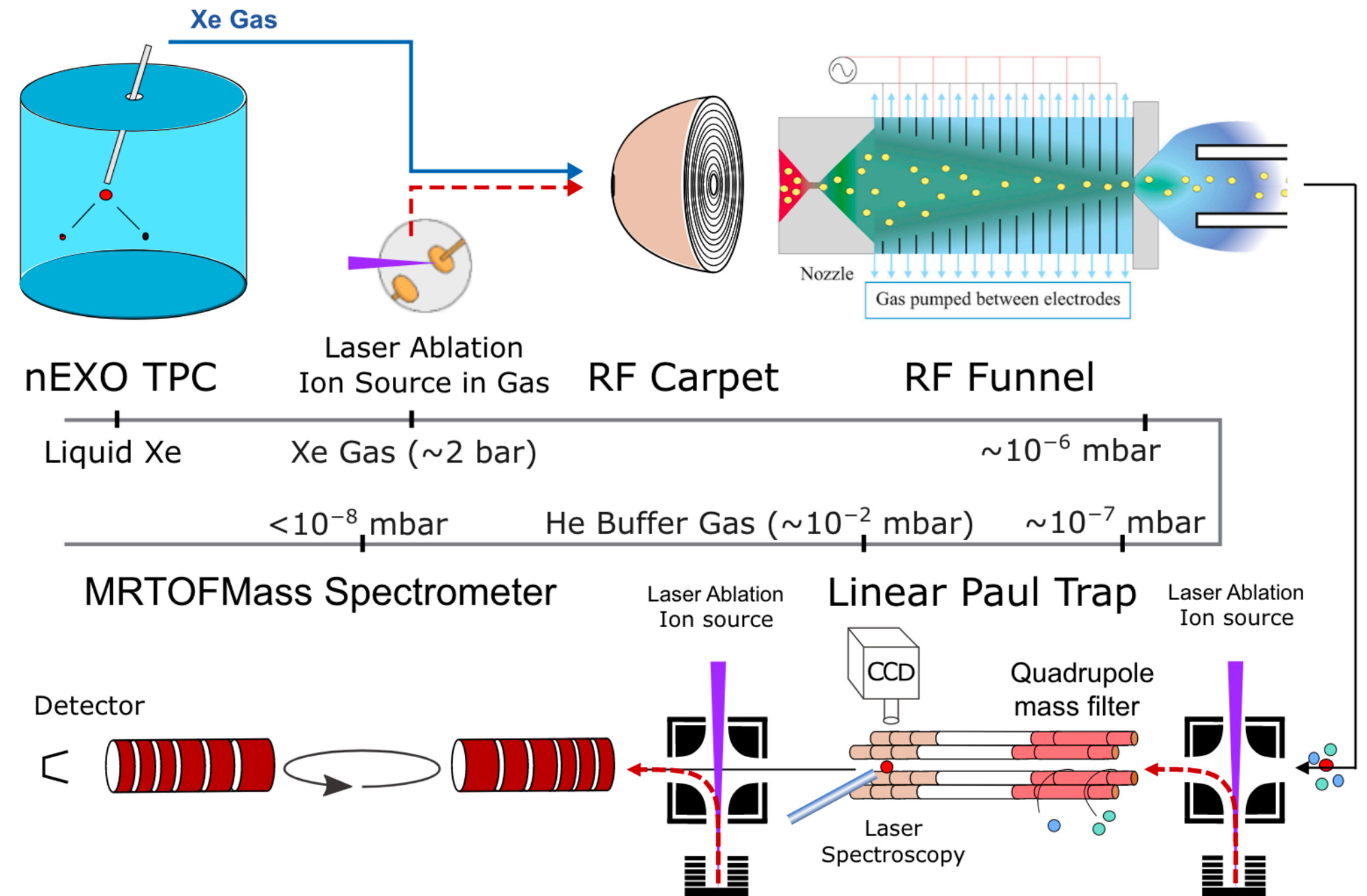


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Extraction with capillary

The Canadian approach

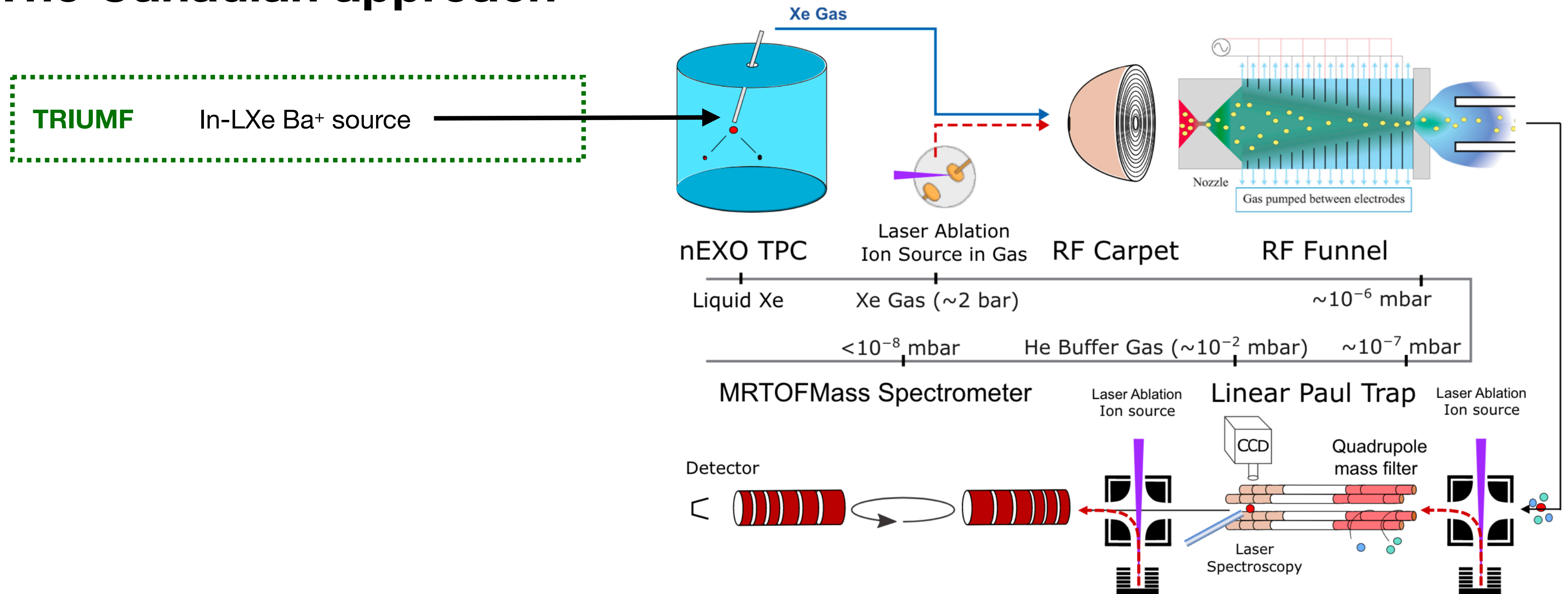


[a] Atoms 2024, 12, 71 (2024) (Ba⁺ extraction from LXe to vacuum)

[b] Hyperfine Interact 240: 97 (2019) (MRTOF design)

Extraction with capillary

The Canadian approach

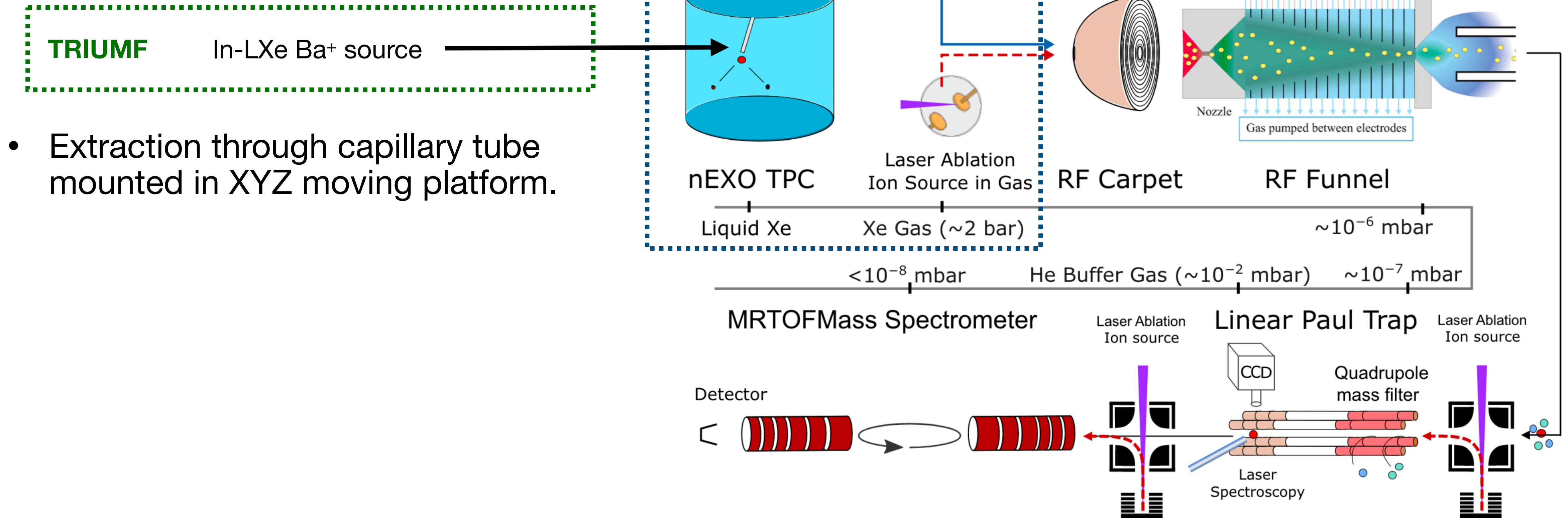


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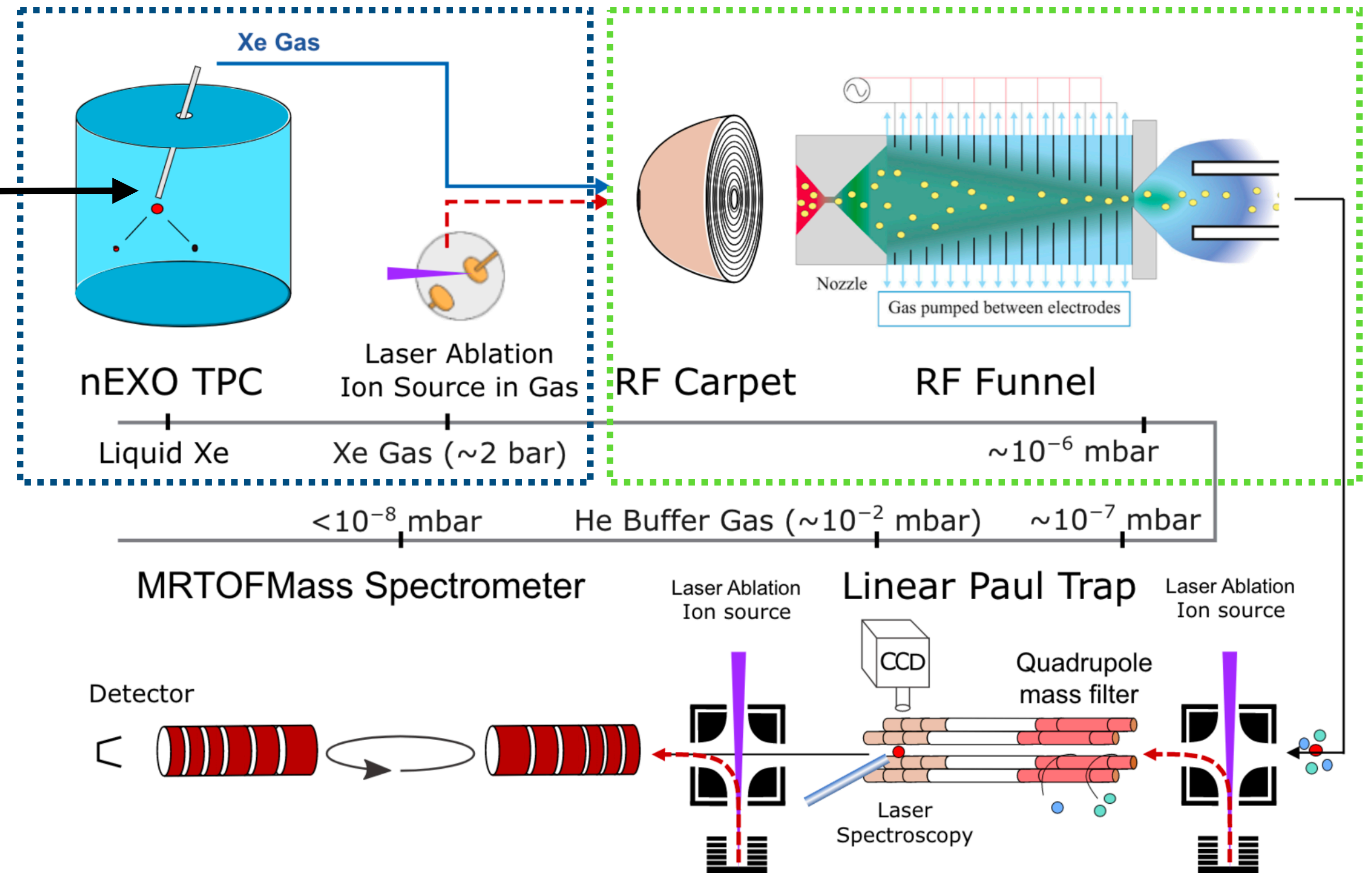
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Extraction with capillary

The Canadian approach

- **TRIUMF** In-LXe Ba⁺ source
- Extraction through capillary tube mounted in XYZ moving platform.
- Differential pumping in RF Funnel to separate Ba⁺ from GXe.



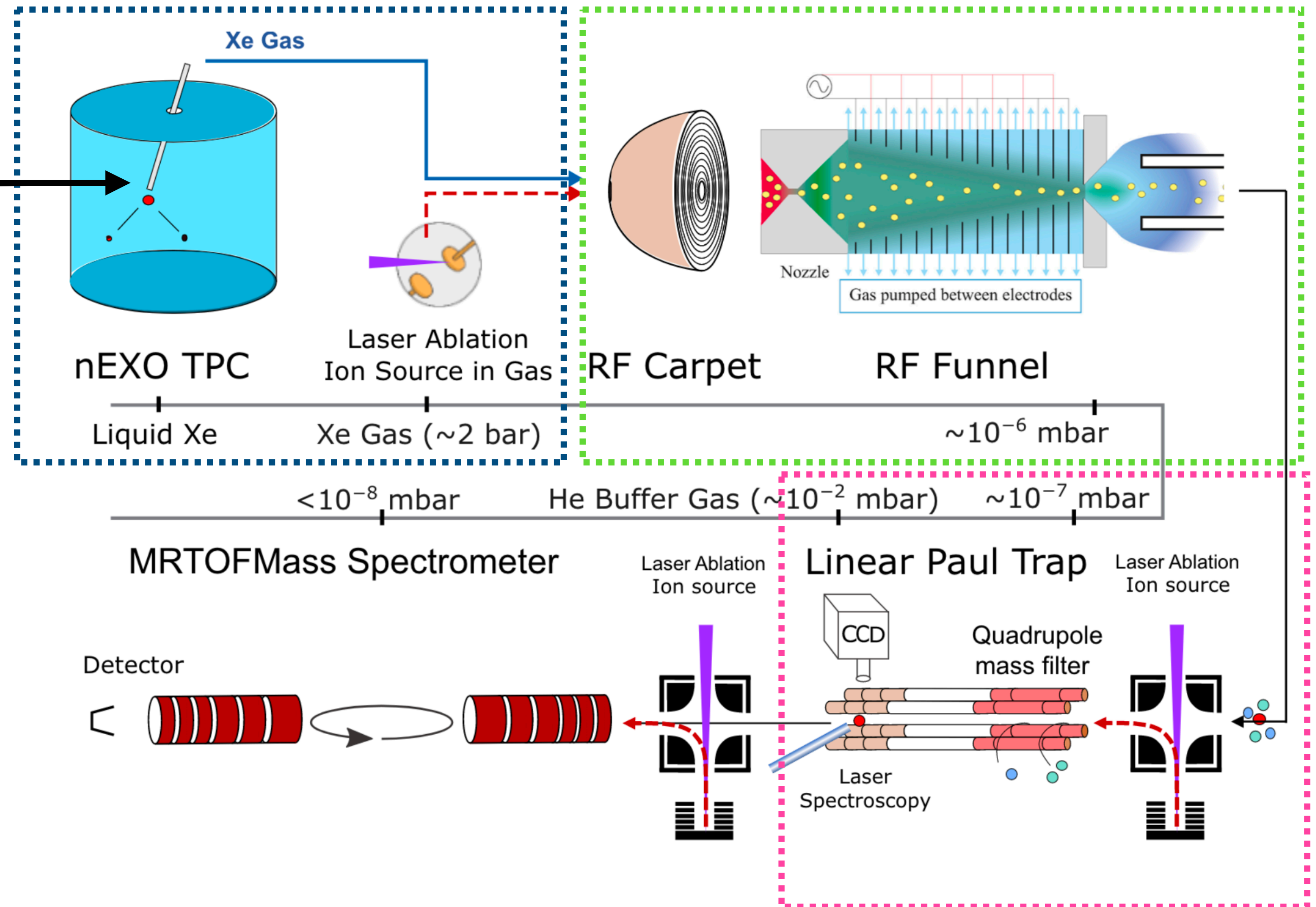
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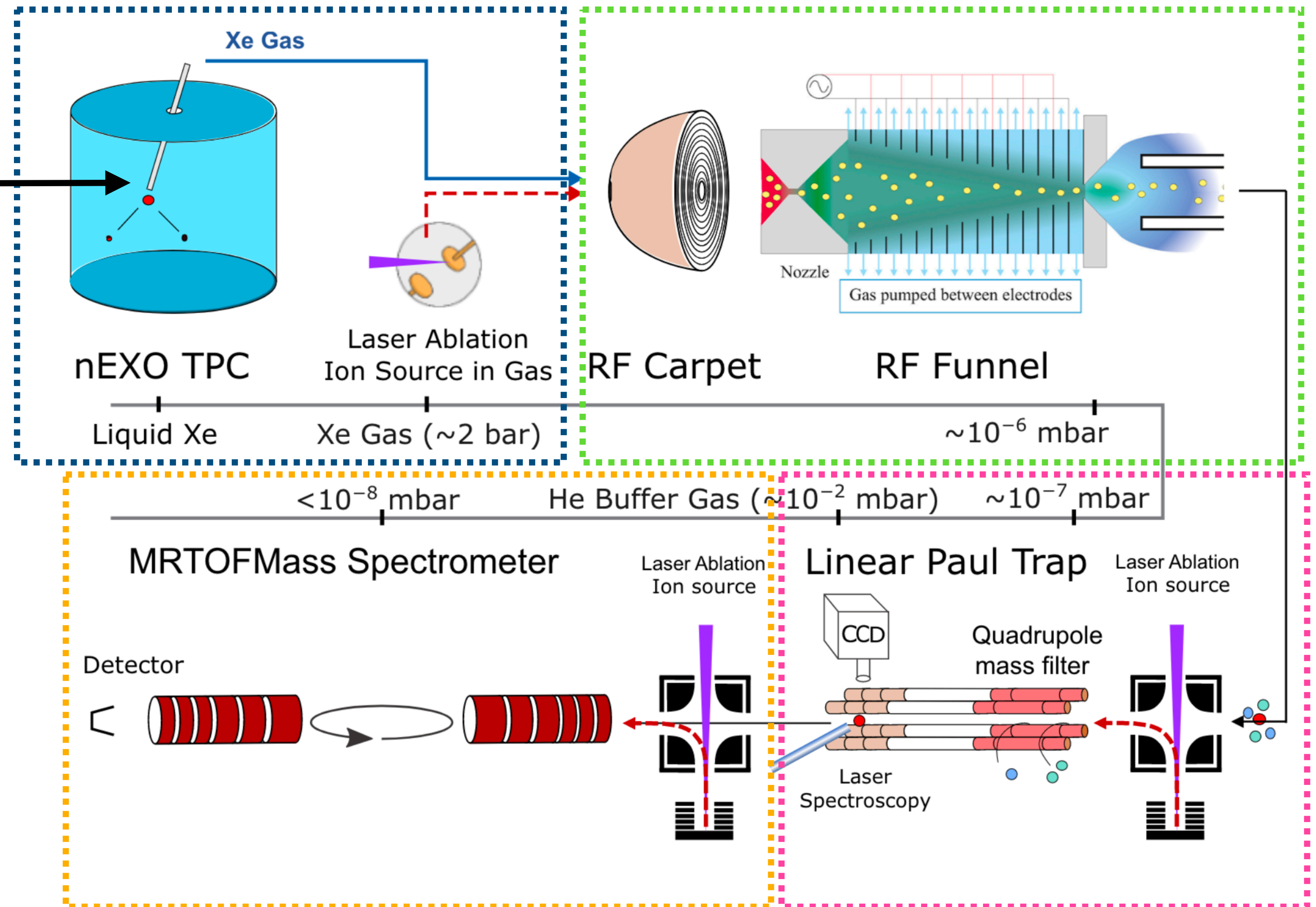
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- Identification of ¹³⁶Ba isotope through MRTOF-MS



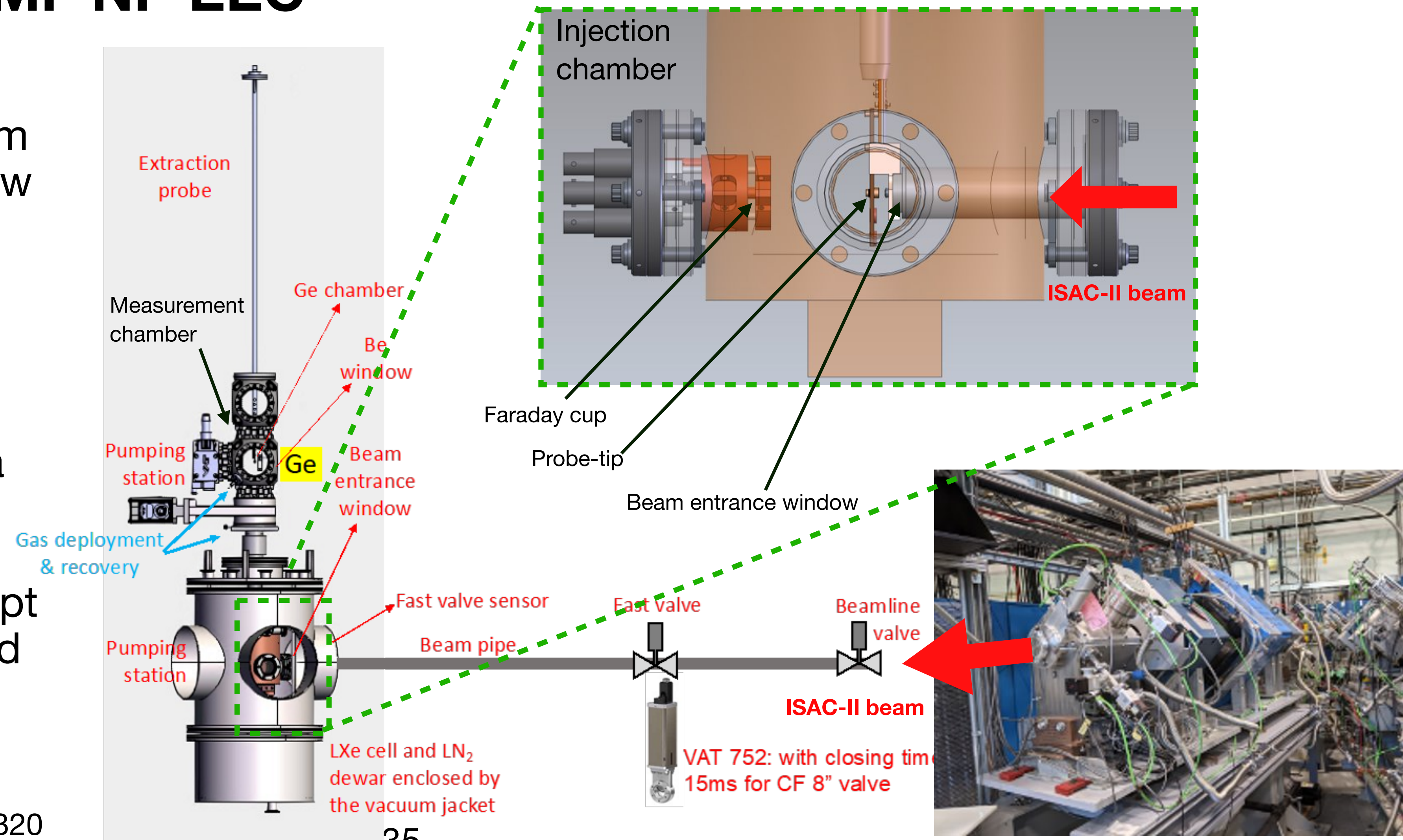
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In-LXe ion source

Proposal for TRIUMF NP EEC

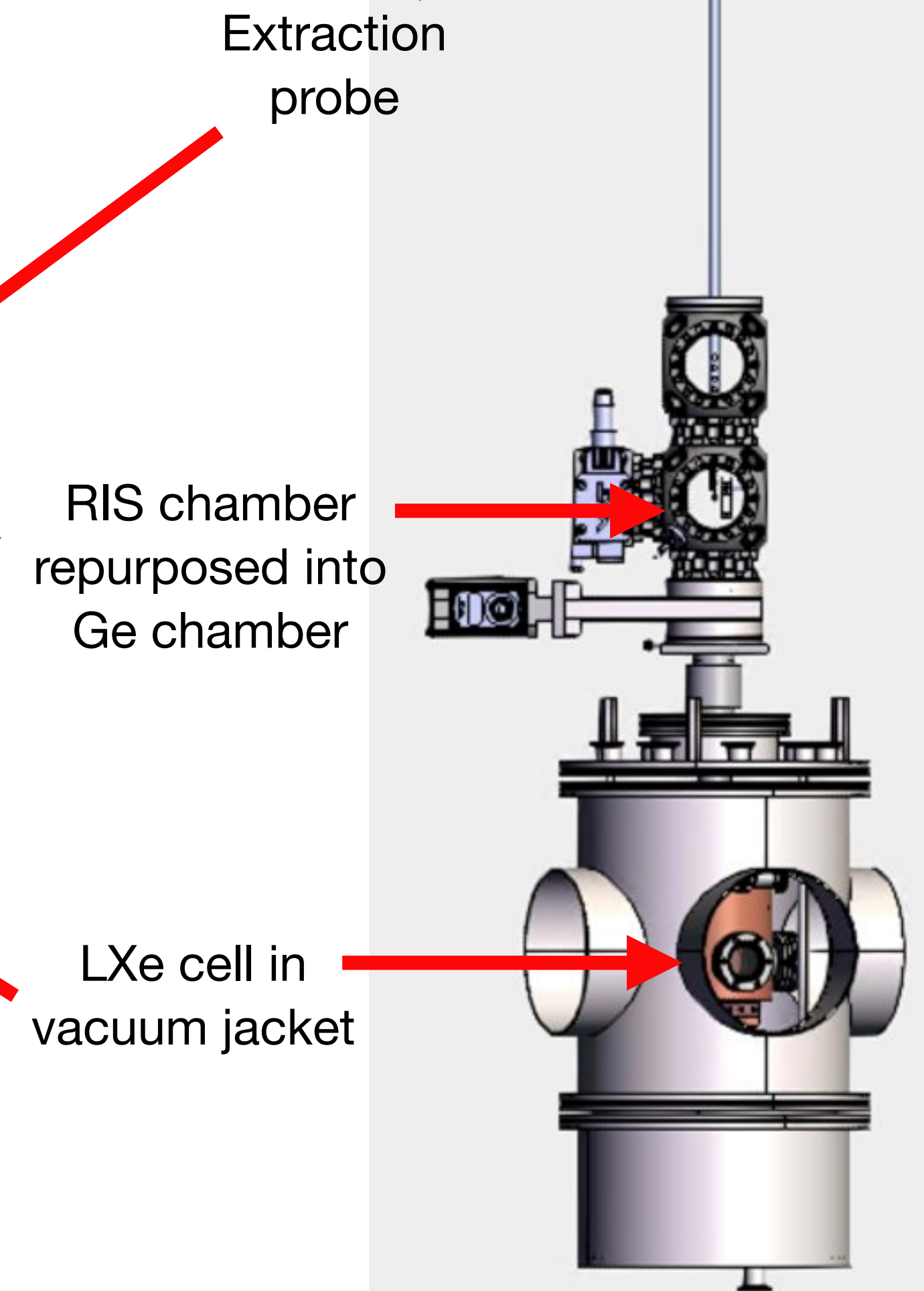
- Inject ^{139}Cs or ^{136}Cs from ISAC through Be window
- Stopped ions in LXe
- Cs decays into Ba
- Collect the daughter Ba with electroprobe
- Future upgrades to adapt source to cryoprobe and capillary



In-LXe ion source

Proposal for TRIUMF NP EEC

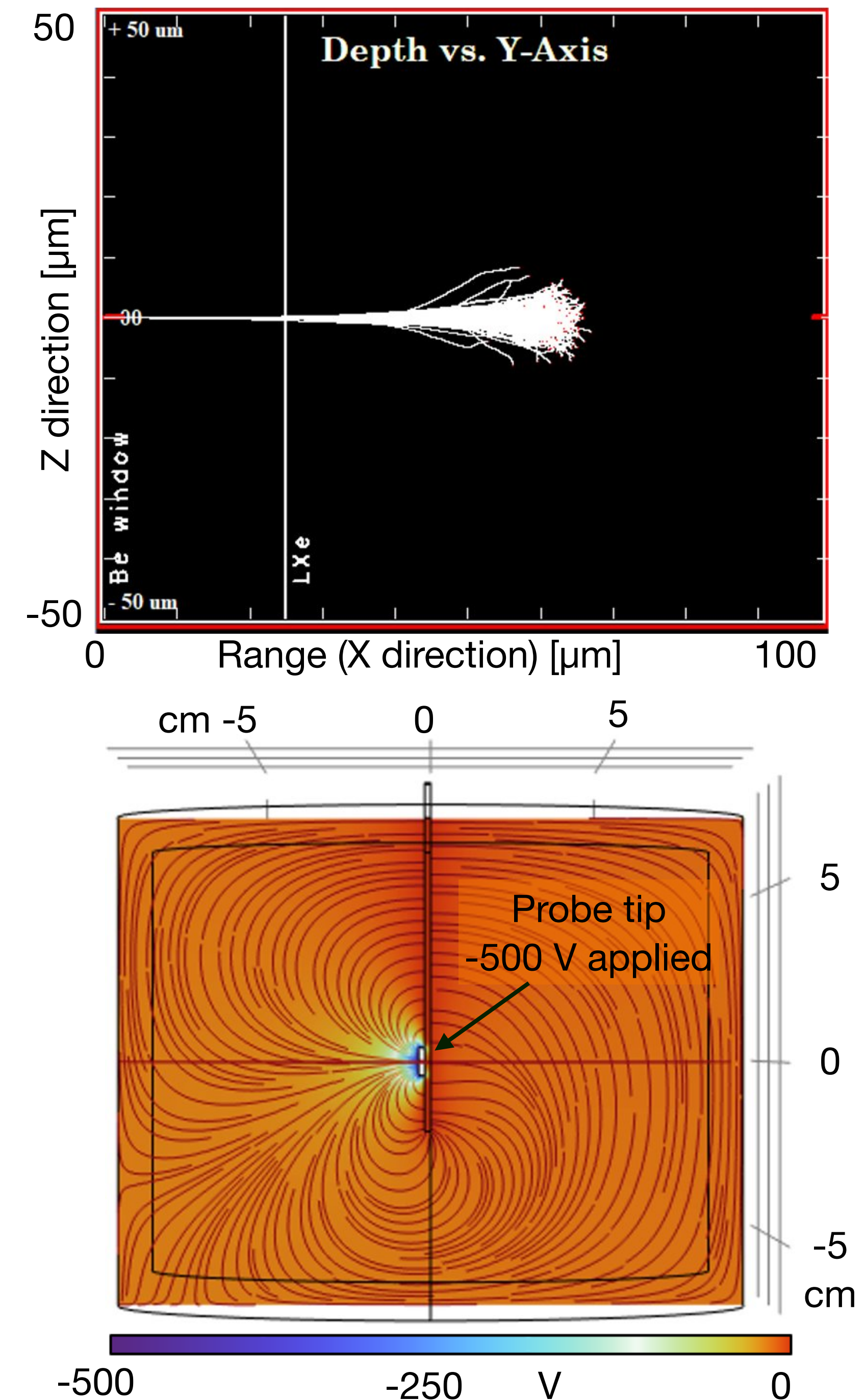
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In-LXe ion source

Proposal for TRIUMF NP EEC

- Stopping 4 MeV/u ^{139}Cs after 25 μm Be window and 35 μm in LXe
- Solved electrostatic and fluid dynamics with COMSOL
- 100% collection efficiency at -500 V after ~ 100 s

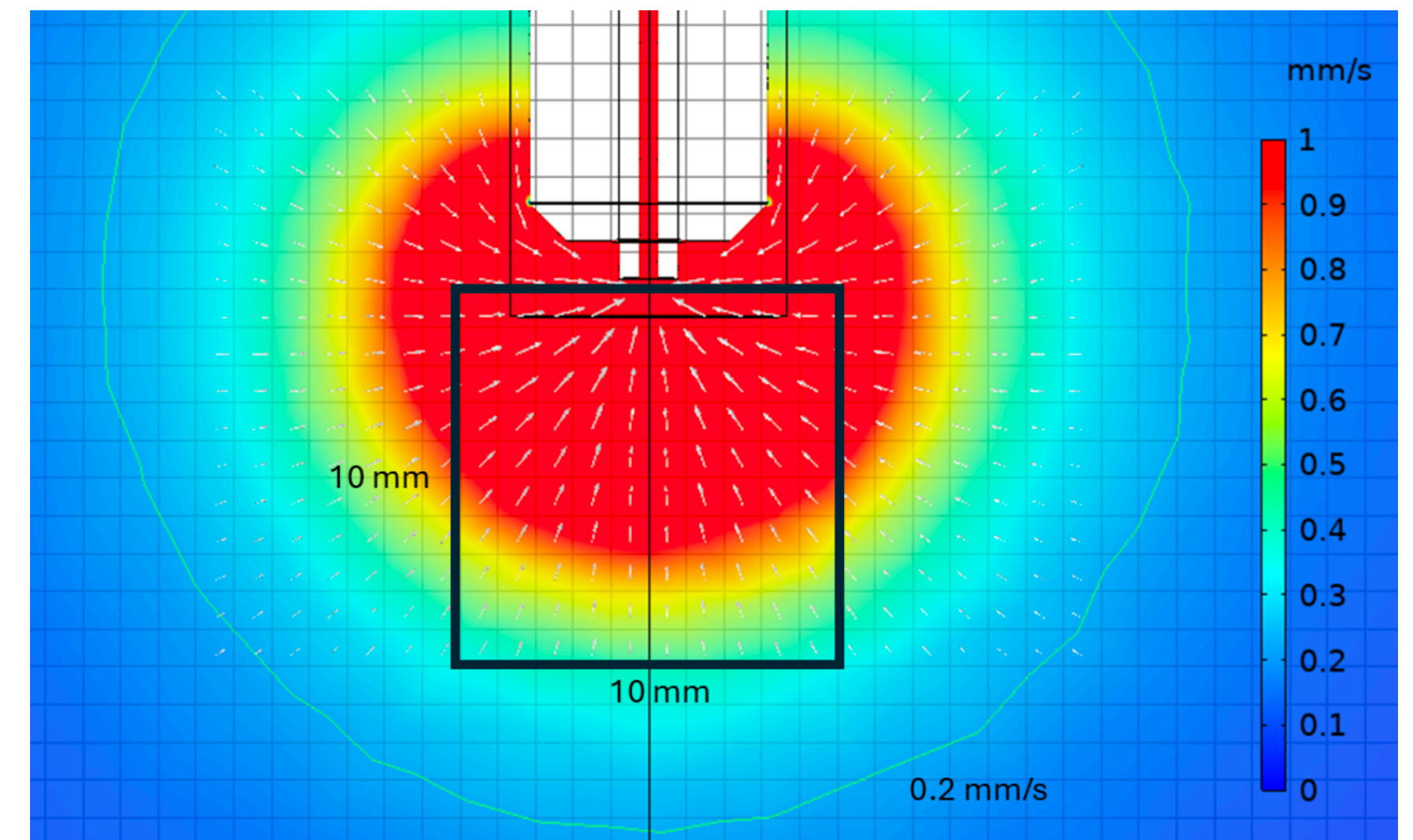
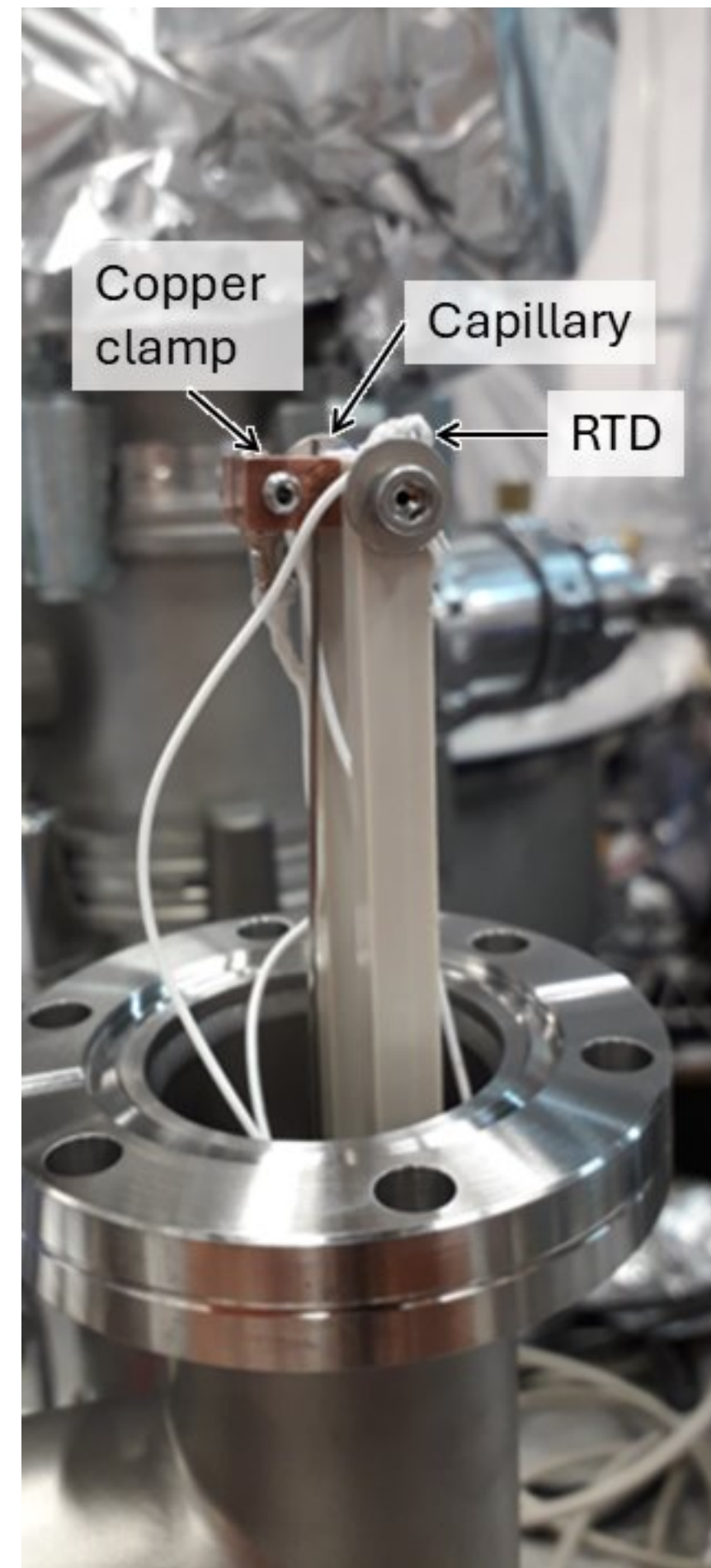


The capillary

Ba⁺ extraction from LXe



- Target volume: 1 cm³. Precision of 0.11 mm
- Once inside, the tube is heated to transport Ba in LXe-GXe mixture
- Successful proof-of-principle untargeted ion transfer using ²²²Rn

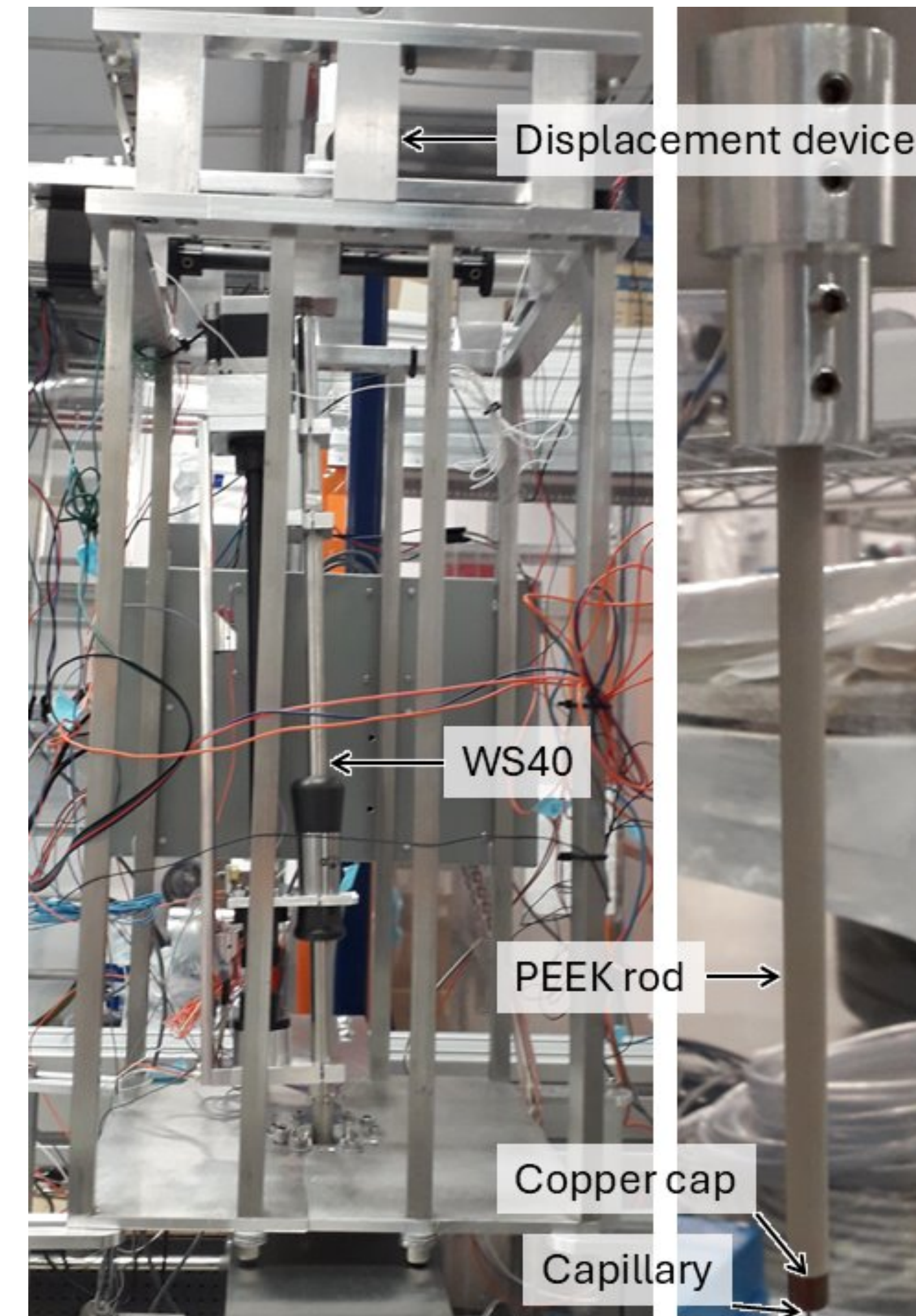
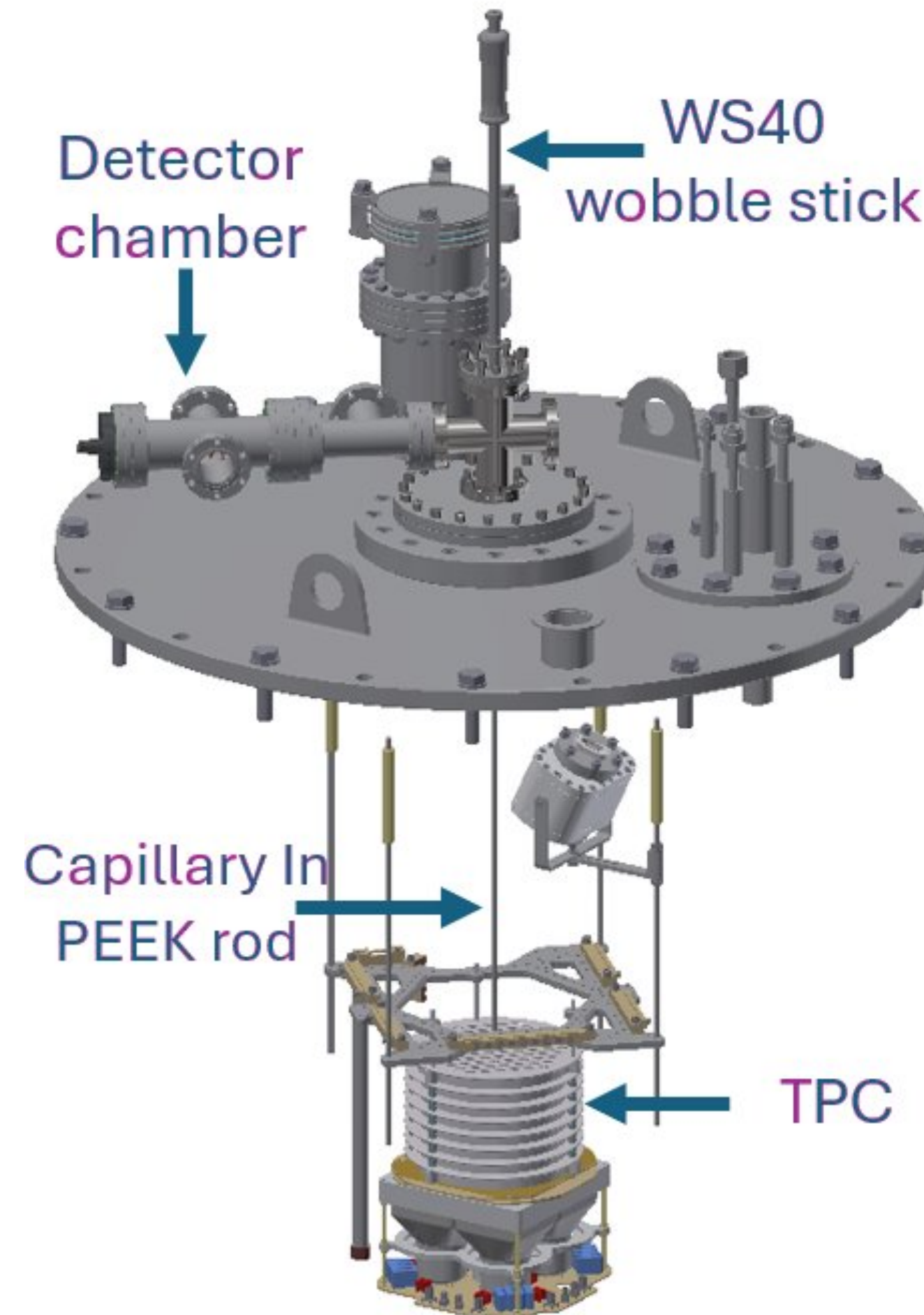


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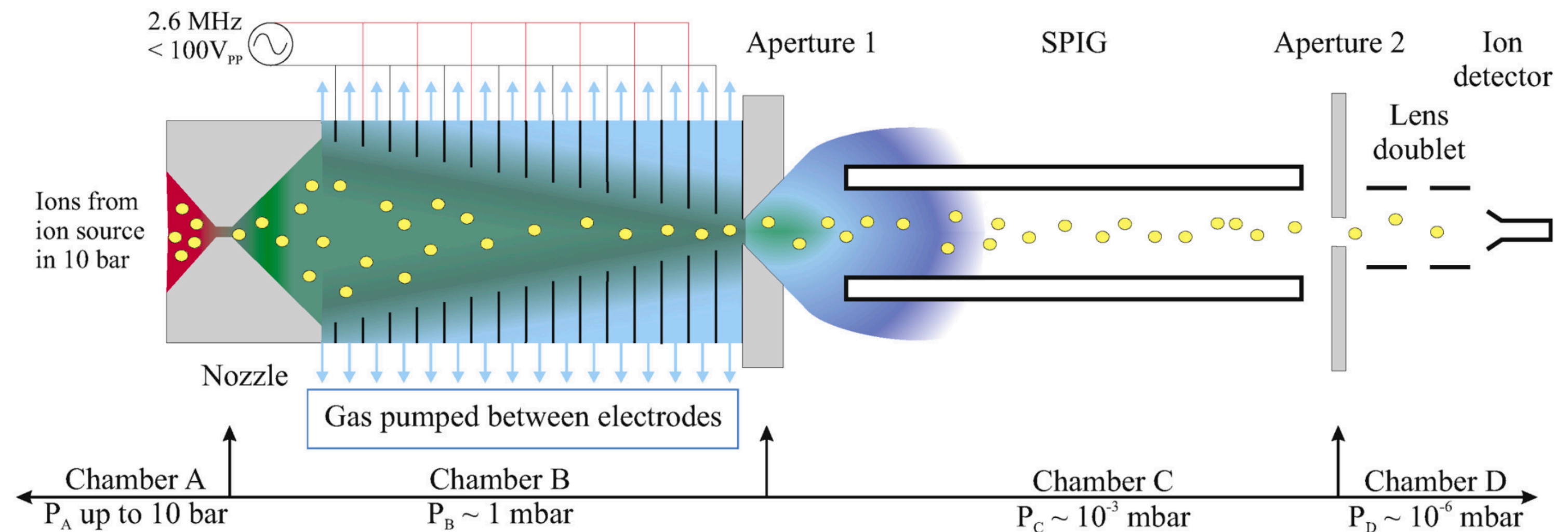
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Separation of Ba from GXe

RF funnel + diff. pumping from 10 bar to 10^{-6} mbar

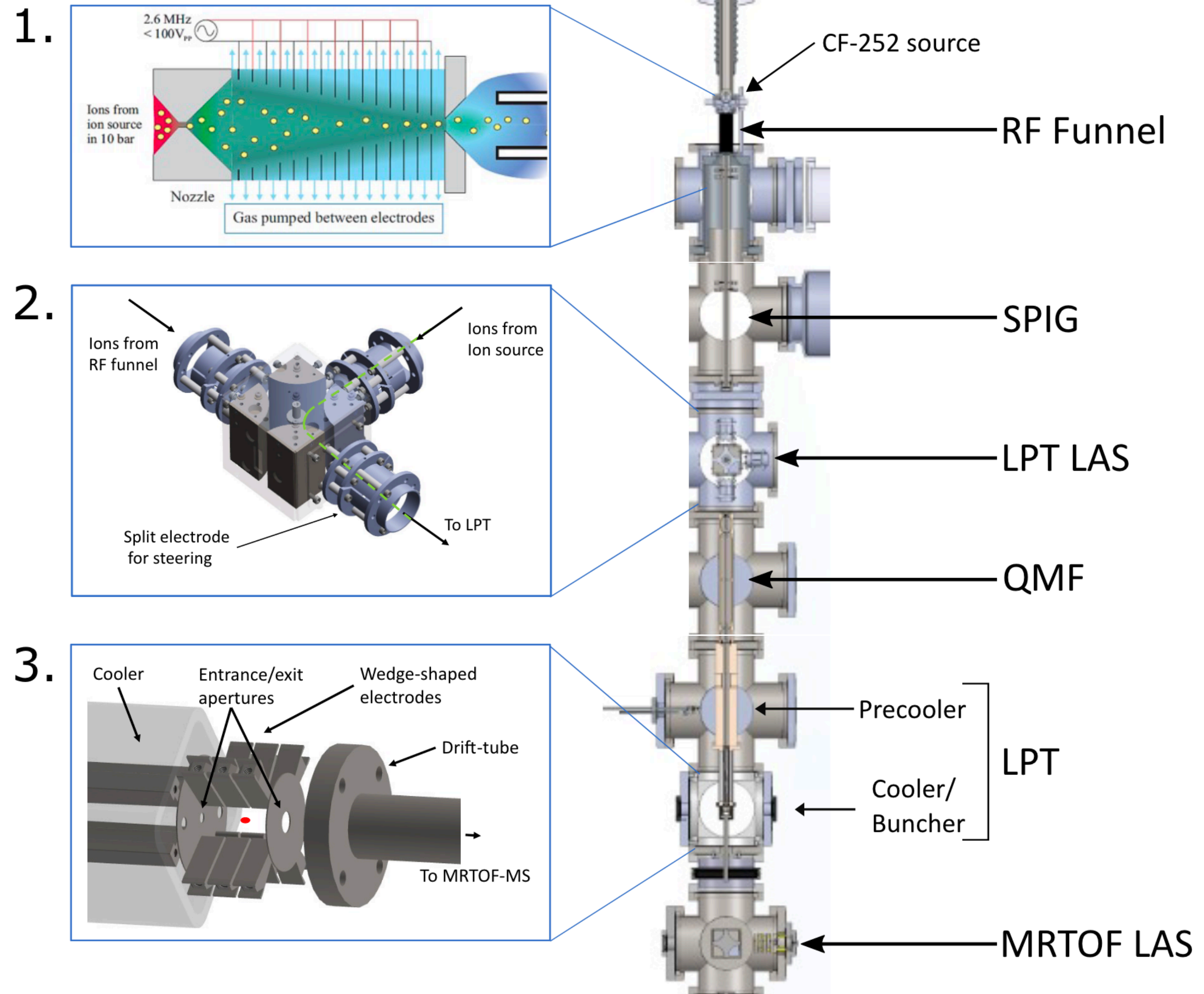
- Extracted GXe up to 10 bar
- RF-only \rightarrow no axial drift field \rightarrow less contaminants
- Mass spectrum with Sixtupole Ion Guide (SPIG)
- Faraday/CEM detector operating at 10^{-6} mbar



Ba element identification

Linear Paul Trap (LPT) + Laser spectroscopy

- Can introduce different ions by LAS + bender for calibration
- A Quadrupole Mass Filter removes out remaining BaH, BaOH
- Cooling with 0.1 mbar He to $E < 1$ eV in pre-cooler
- Identification of bunches through laser-induced fluorescence + CCD



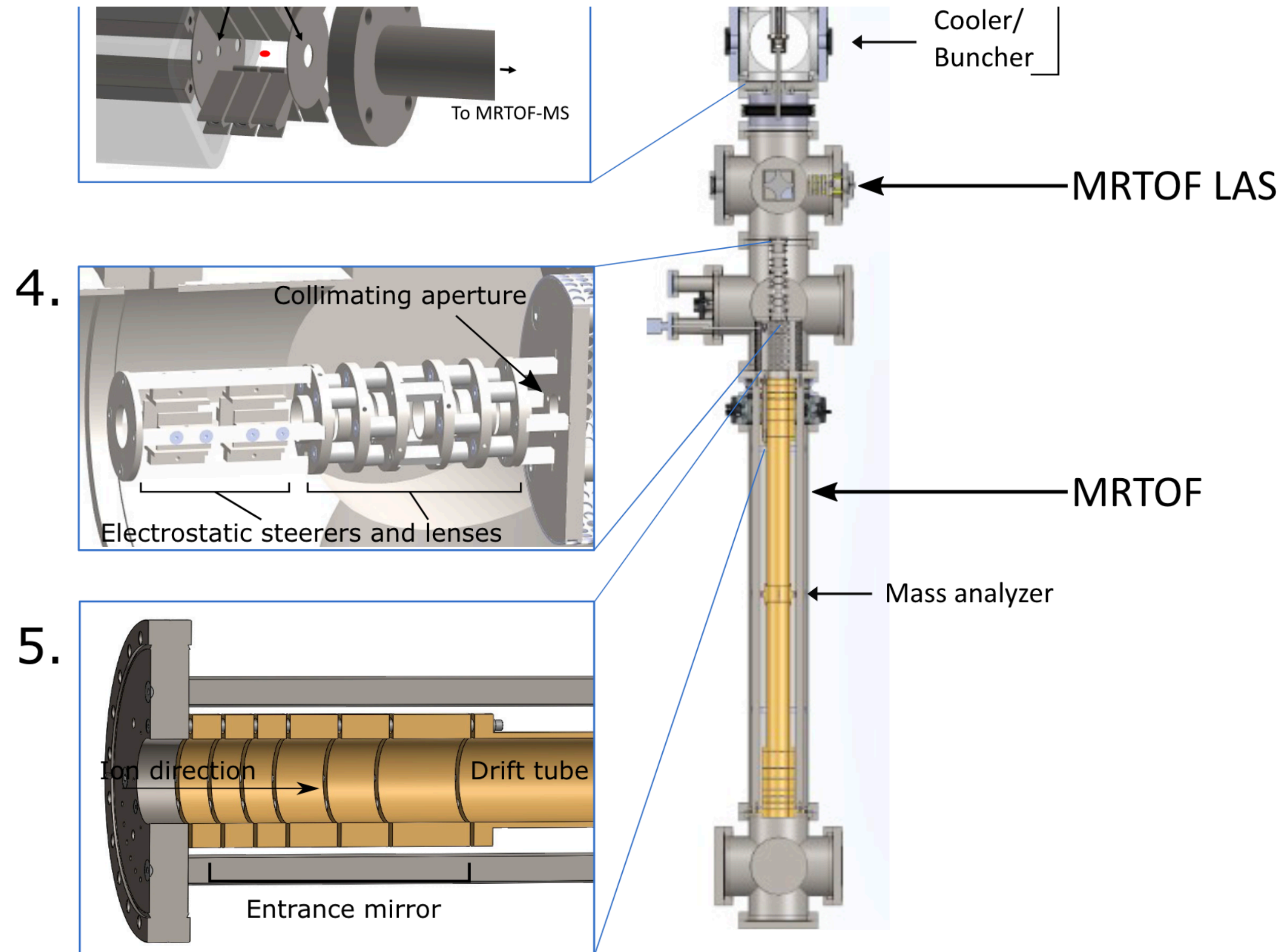
[a] Atoms 2024, 12, 71 (2024)

[b] Nucl. Instrum. Methods Phys. Res., B 541 (2023) 298–300

Isotopic identification of ^{136}Ba

Multiple Reflection Time-of-Flight Mass Spectrometer (MRTOF-MS)

- Reflections increase difference in their TOF and thus m/q .
- 2 electrostatic mirrors, each of 6 electrodes
- Steerers and focusing (einzeln) lenses for injection
- Achieved $m/\Delta m \sim 20,000$ with unbunched ions from LAS



Overview

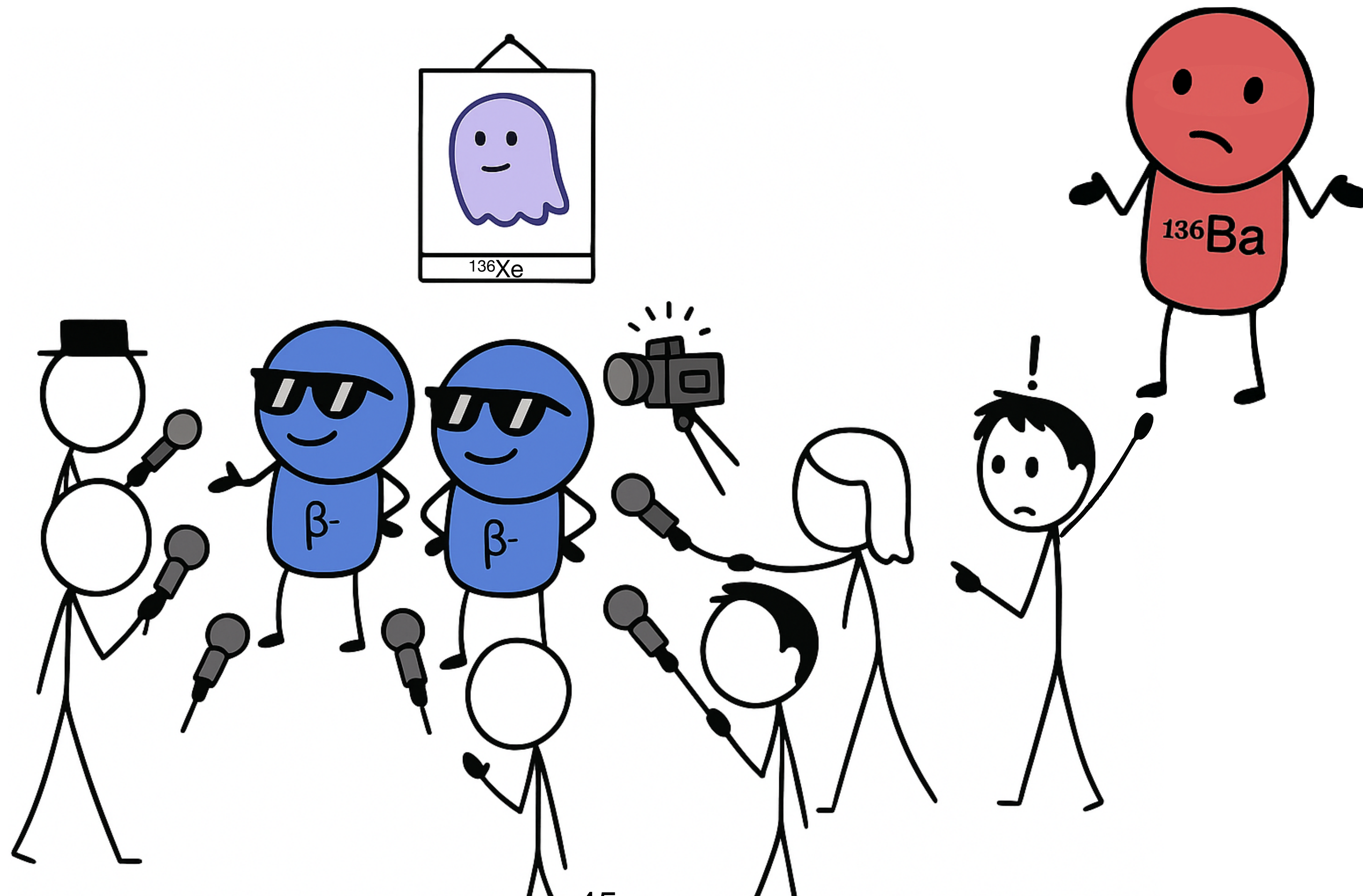
- ☒ Introduction to Ba-tagging
- ☒ Ba-Tagging in NEXT
- ☒ Ba-Tagging in nEXO
- ☐ **The road forward**

Synergies in Ba-Tagging

Regardless of LXe/GXe

- Can we neutralize Ba^{2+} in GXe to Ba^+ and use laser spectroscopy?
- Can we use chemosensors with neutral Ba or mono cation Ba^+ ?
- ...

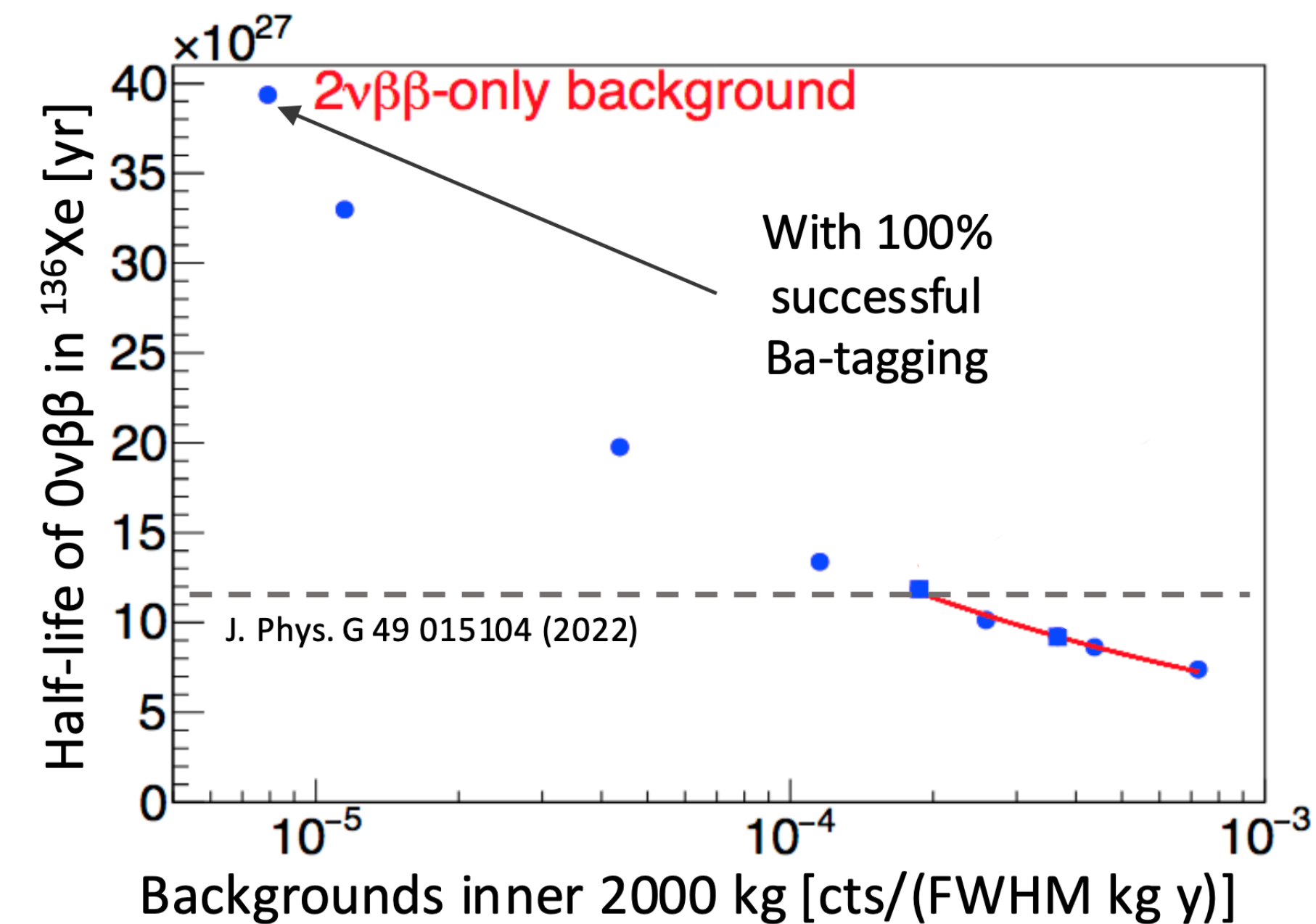
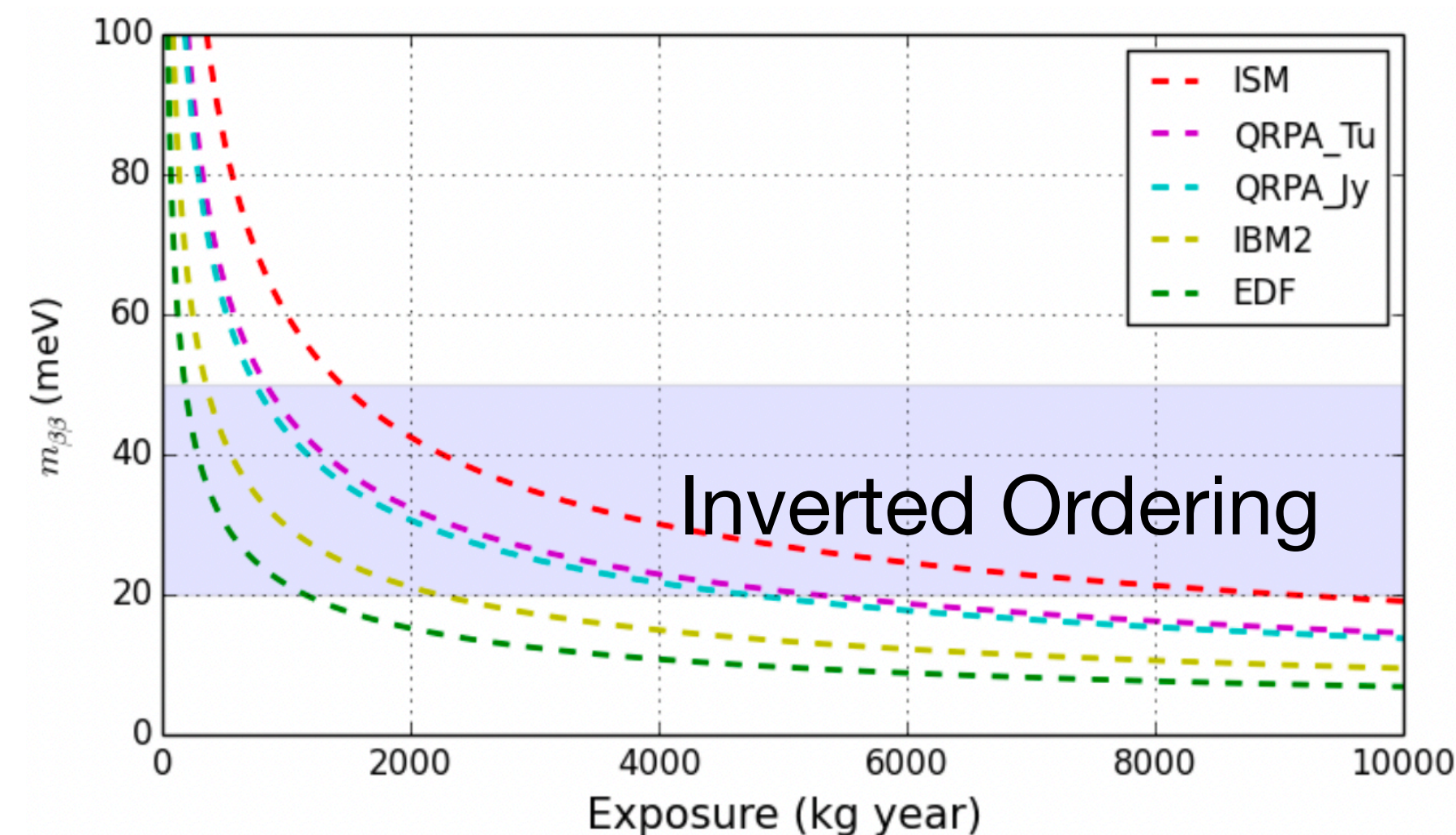
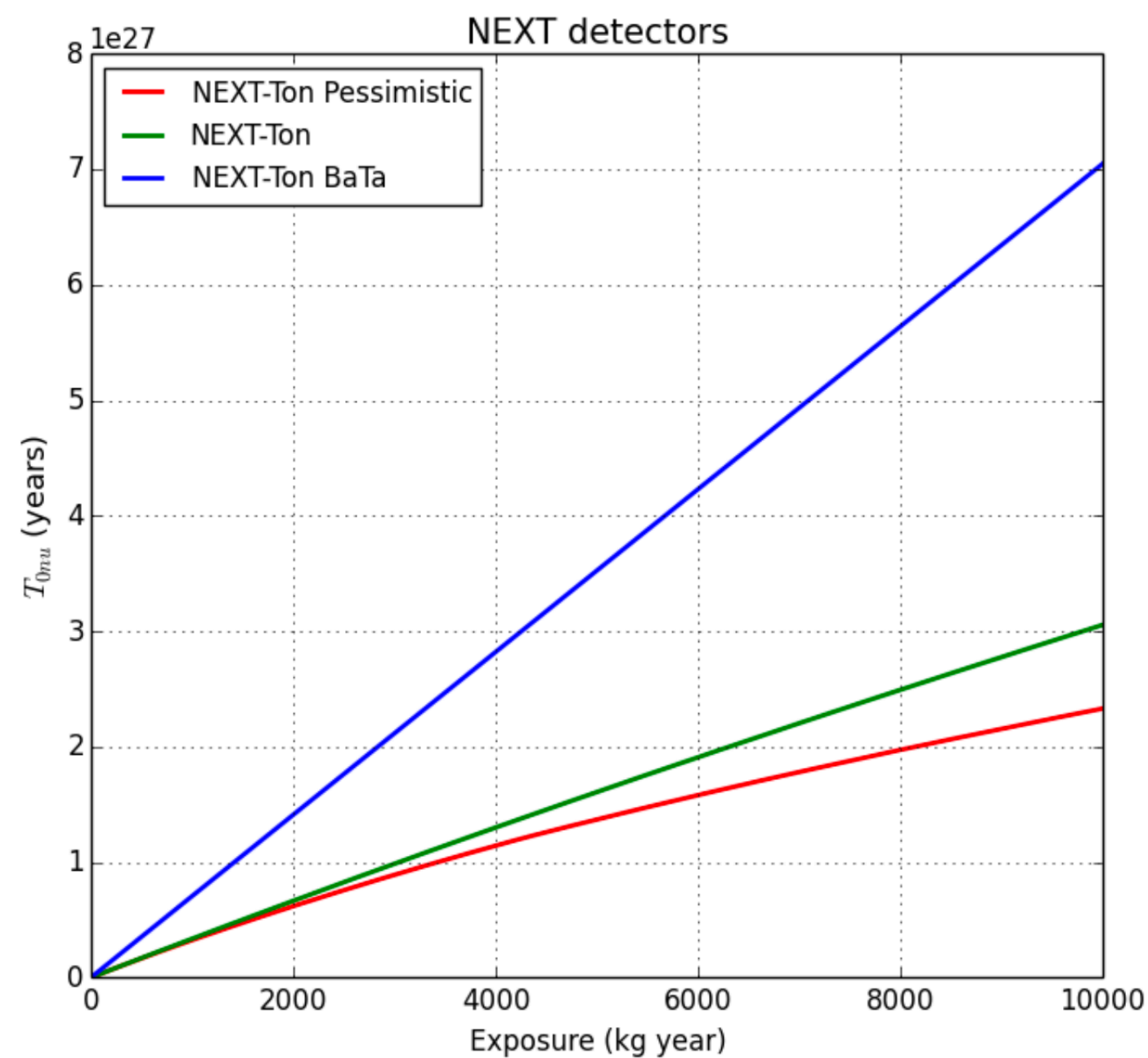
Thank you for your attention!



Backup slides (NEXT)

How far could we go?

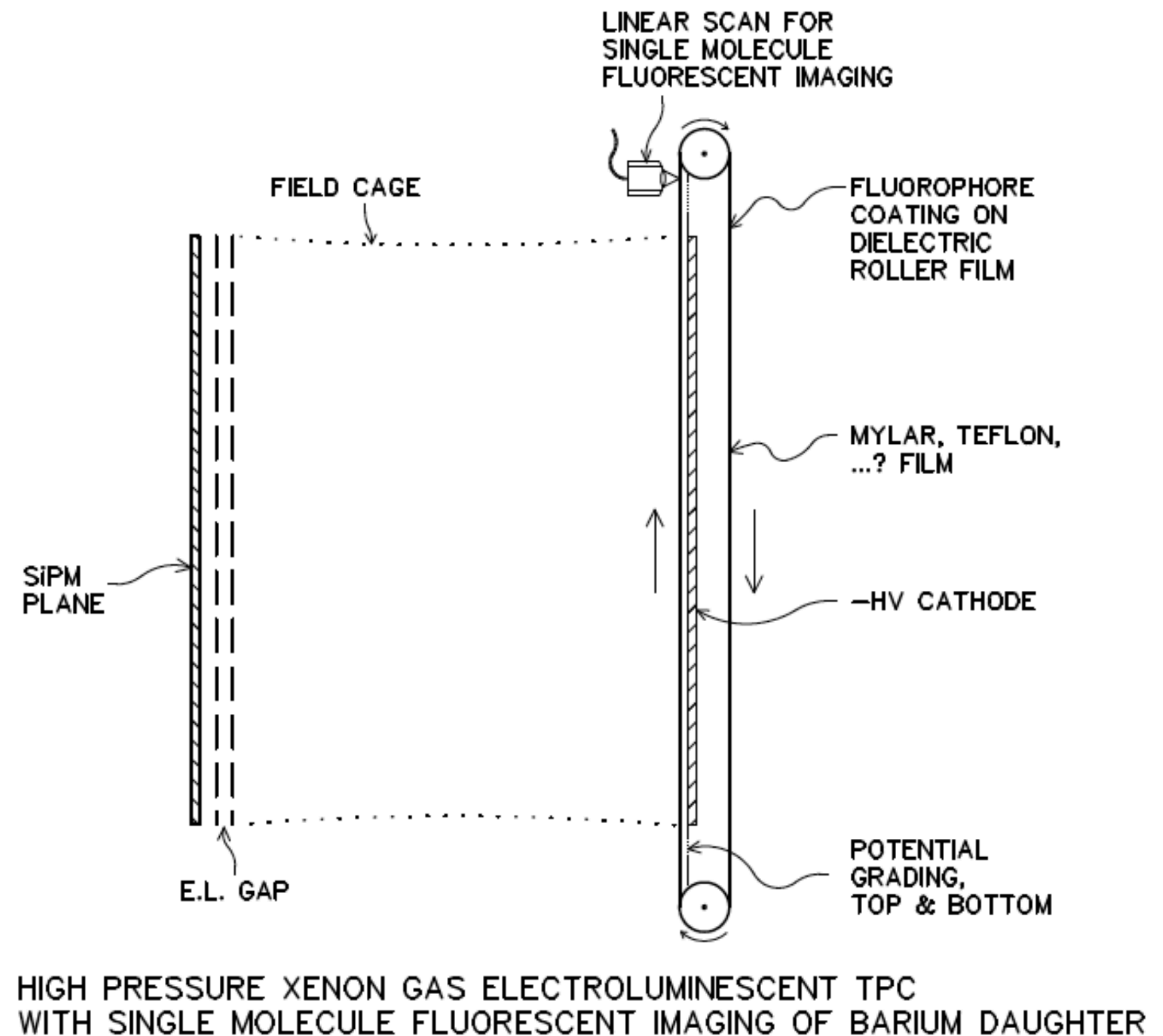
- Linear scale of sensitivity to $\tau_{0\nu\beta\beta}$ would with exposure if zero background.
- The full IO (blue shaded area) of $m_{\beta\beta}$ could be probed with 1 ton \times 10 years exposure.



From J. Muñoz (2018) PhD Thesis

Historic precedents

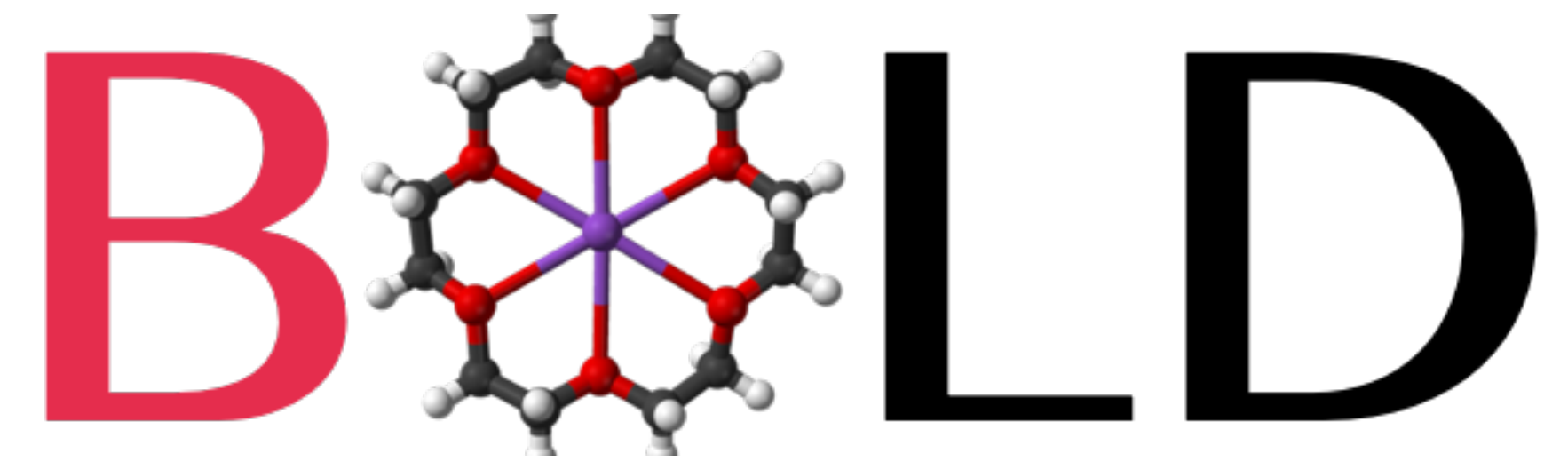
- M. K. Moe (1991) first proposed using **Ba⁺** as a handle for detection of $0\nu\beta\beta$ in coincidence with the 2 electrons.
- D. Nygren (2015) suggests using **Ba²⁺** detection via Microscopy Imaging of chemosensors in GXe.



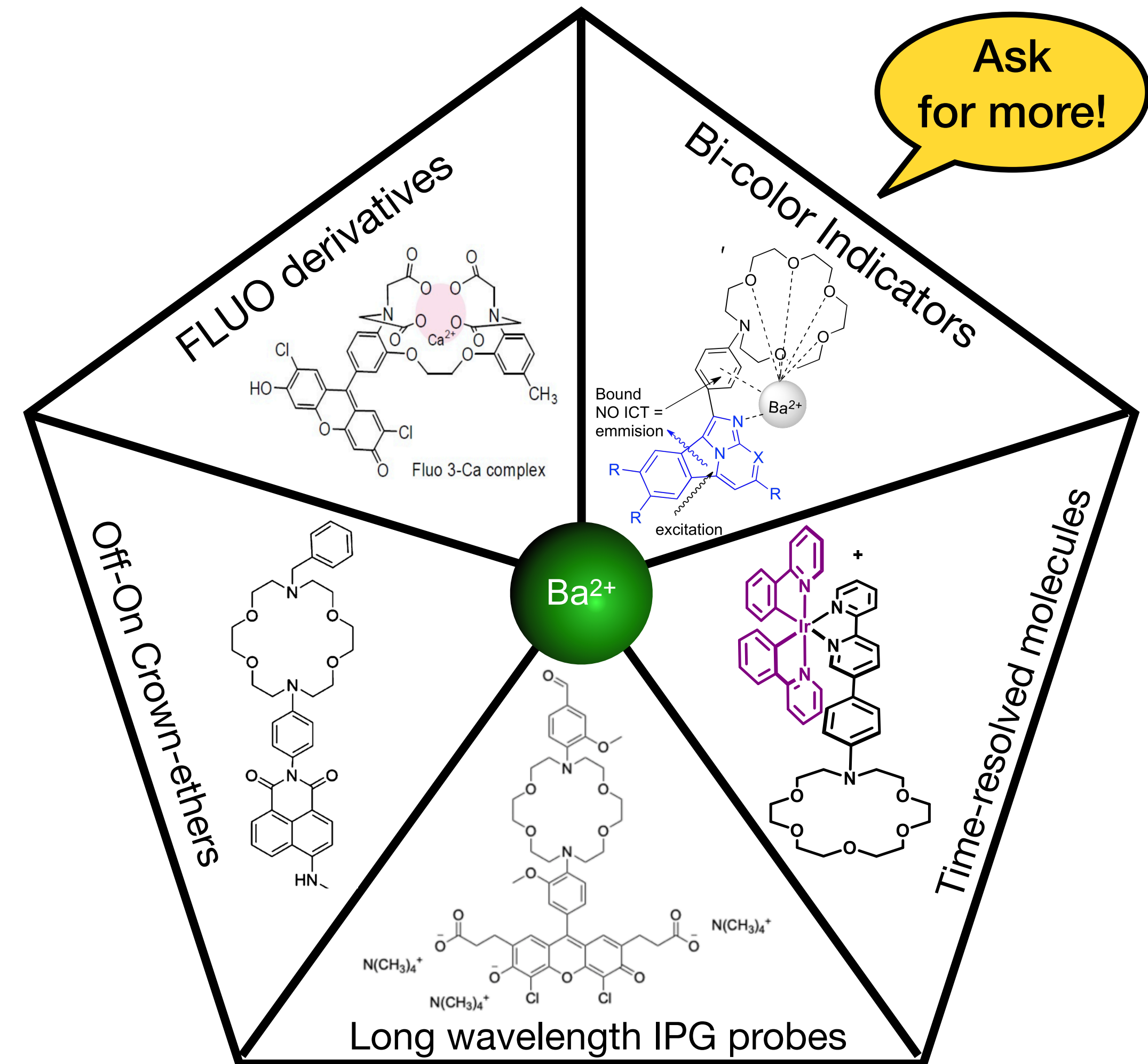
From D. Nygren J. Phys.: Conf. Ser. 650 012002 (2015)

R&D into Chemosensors

The building blocks of the BaTag system



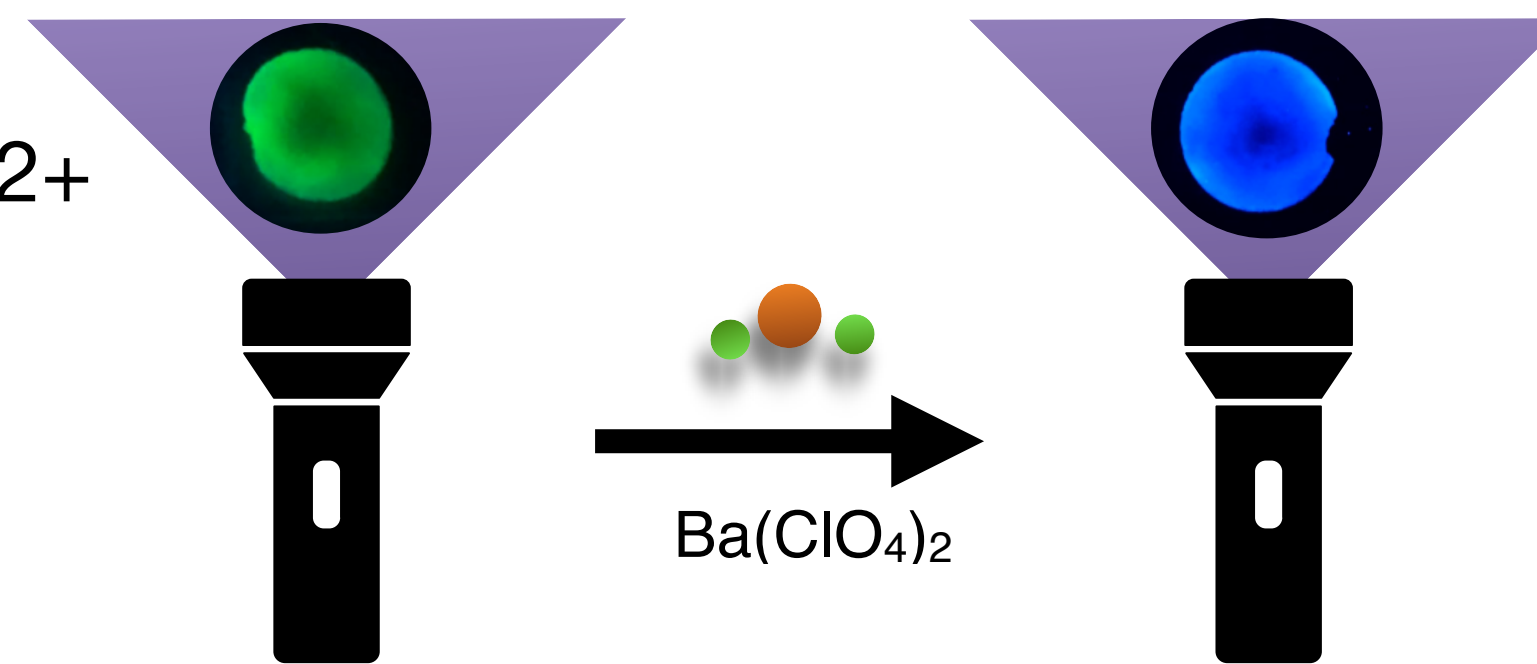
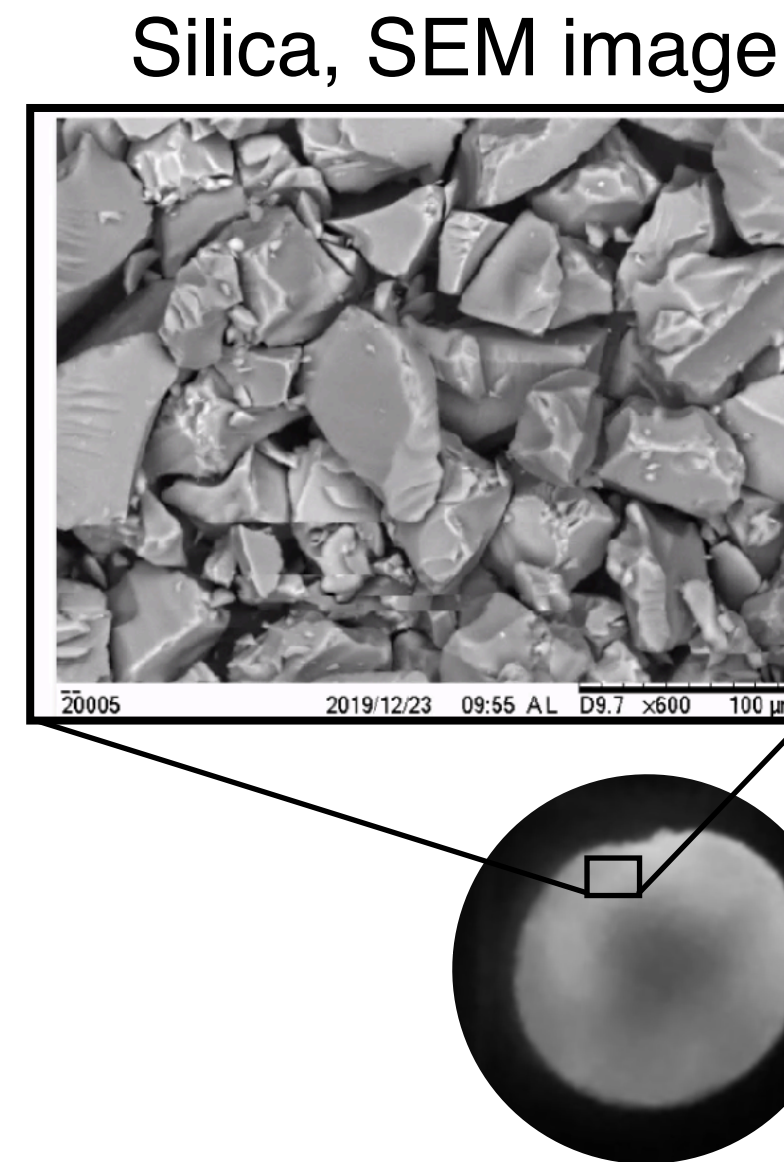
- Most based on crown-ether
- Highly selective to Ba^{2+} ions



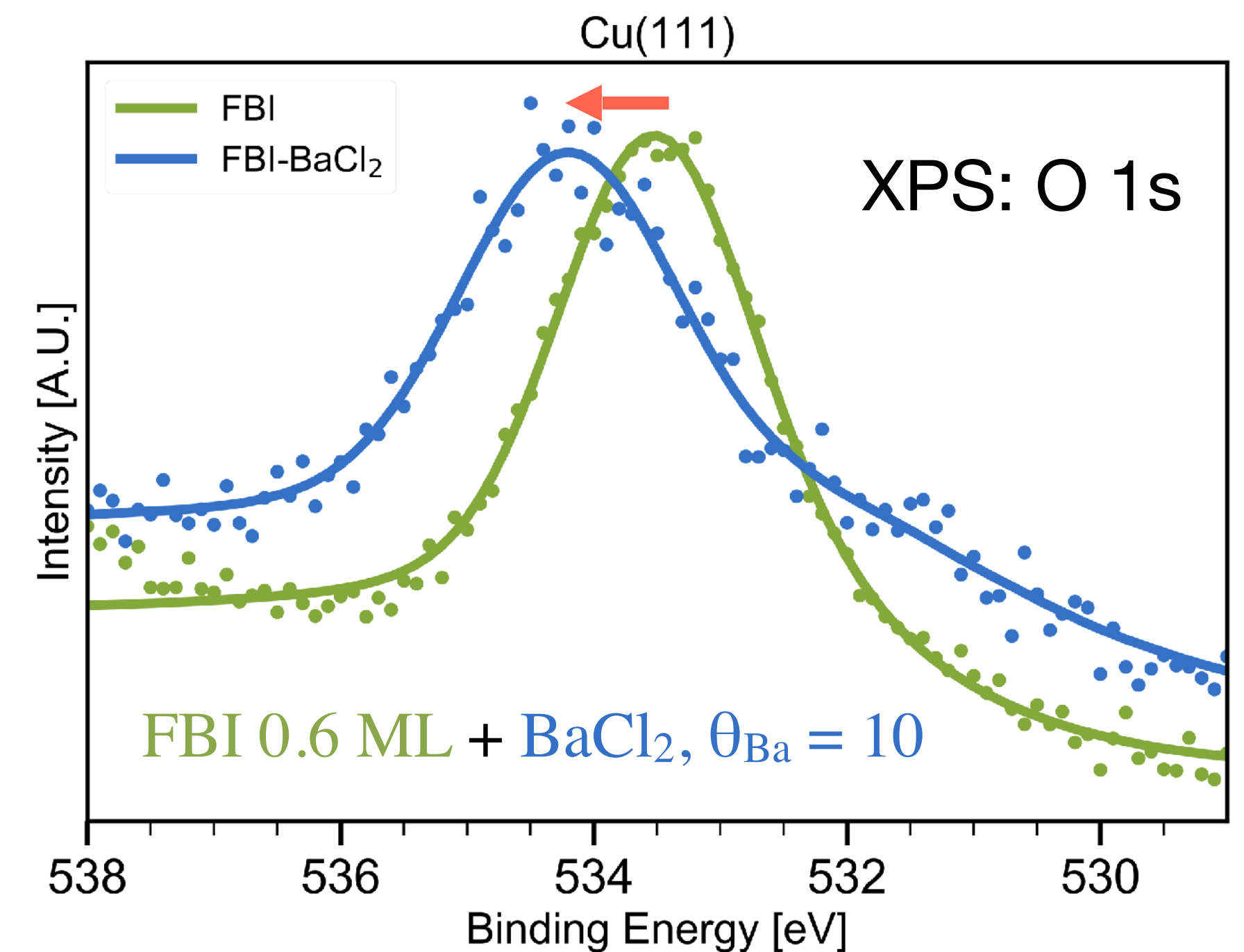
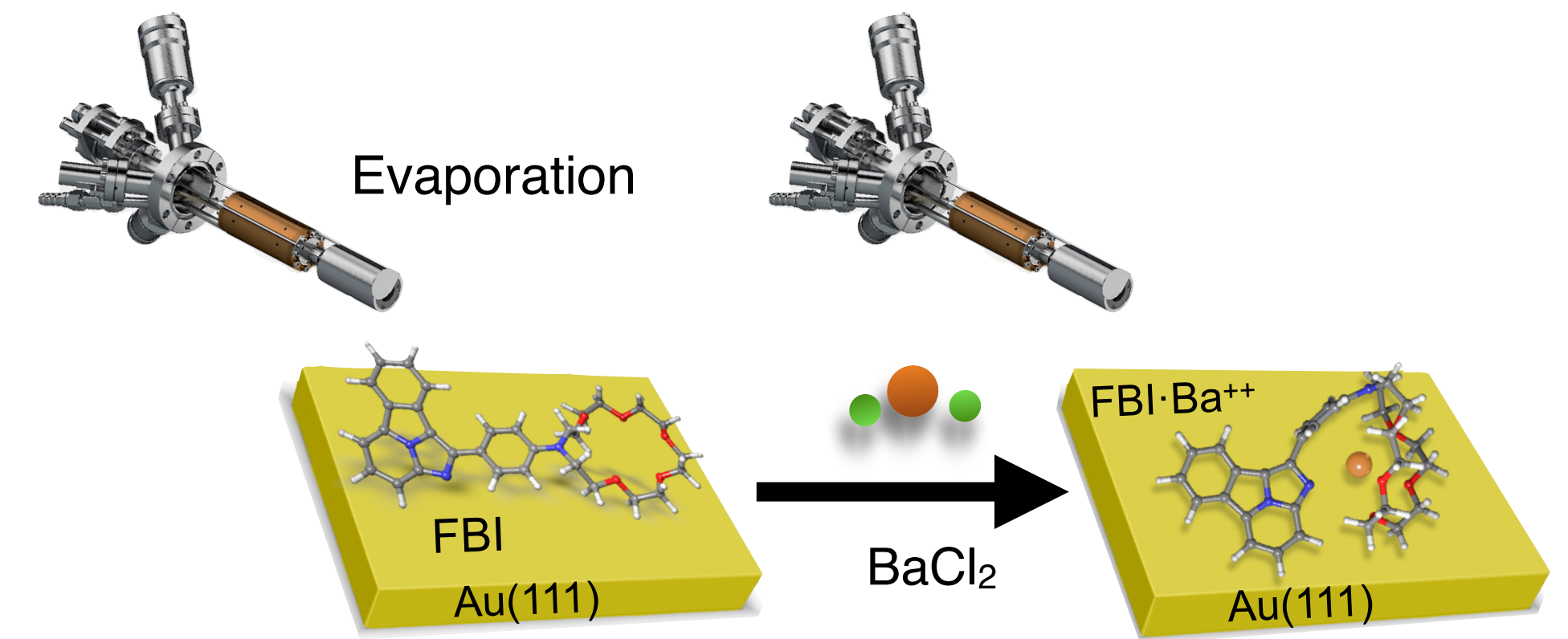
Molecule characterization

Chelation demonstrated in UHV

- 4 different substrates:
➡ Au, Cu, ITO^[b], silica^[a]
- Fluorescence color change preserved in silica pellets^[a]
- Evidence of chemical interaction^[b] of the crown-ether with Ba²⁺
- BaCl₂ added in UHV conditions



[a] *Nature* 583, 48–54 (2020)

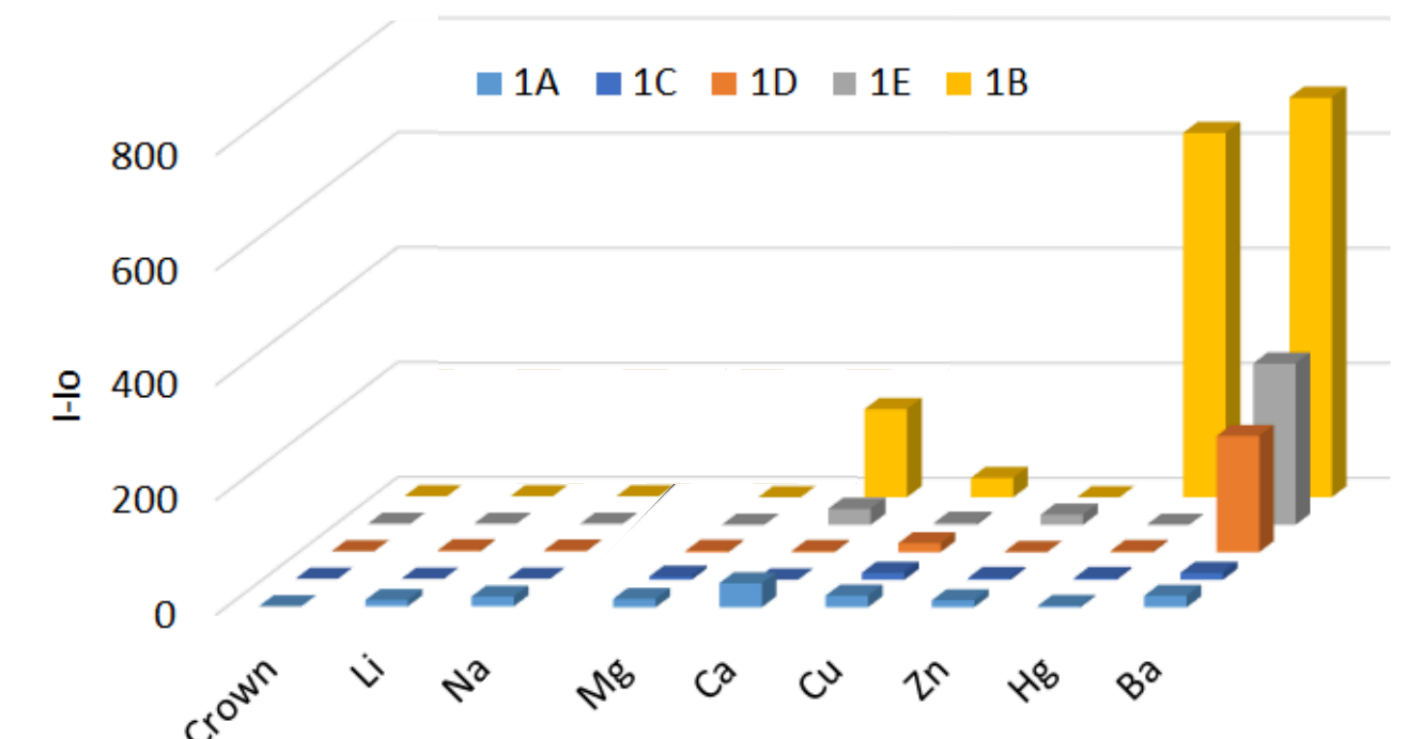
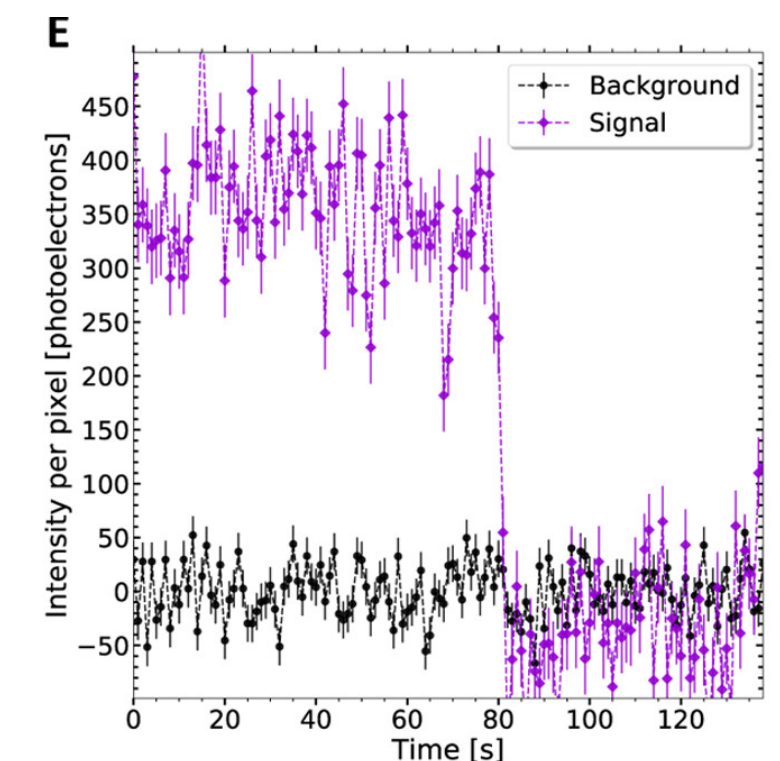
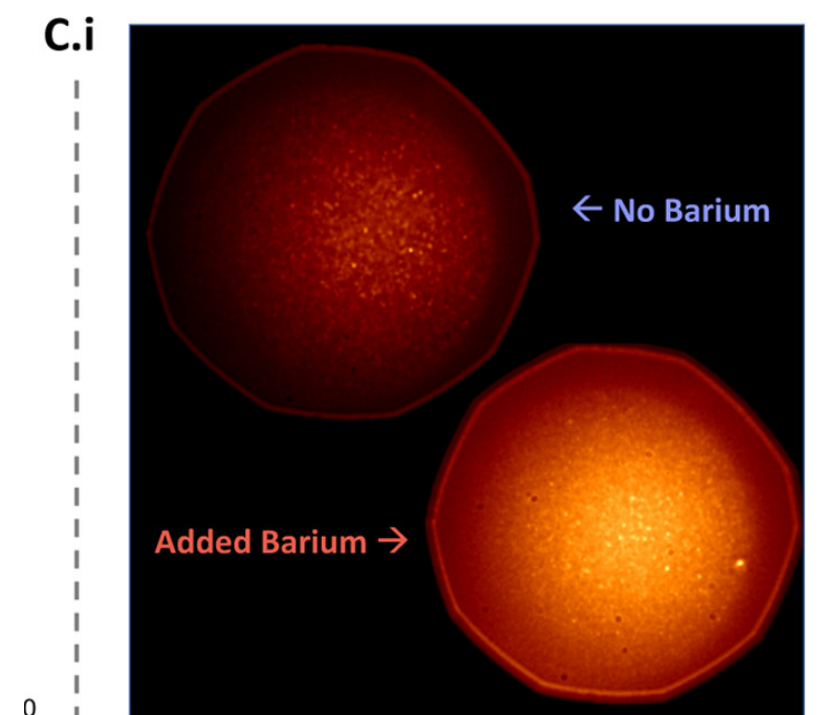
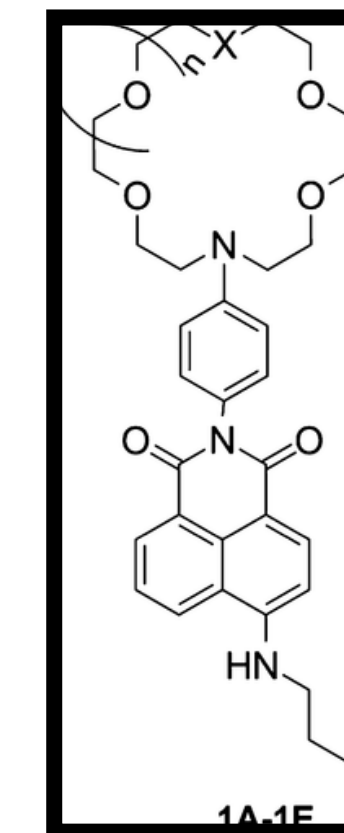
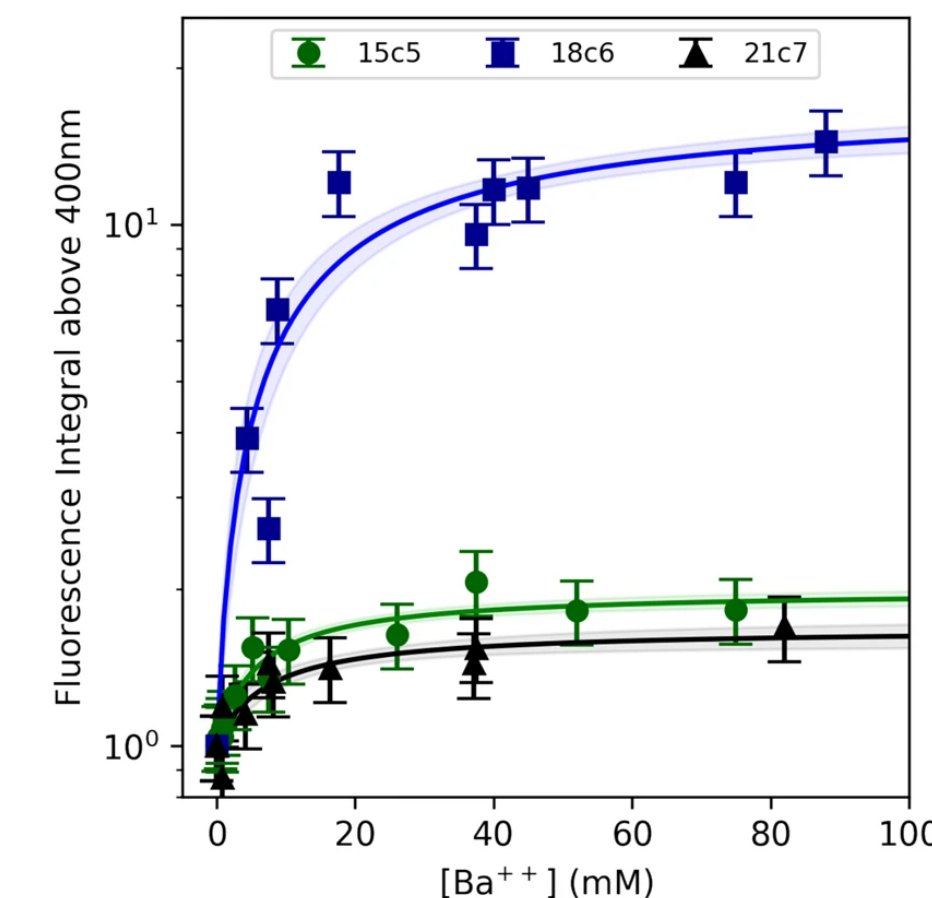
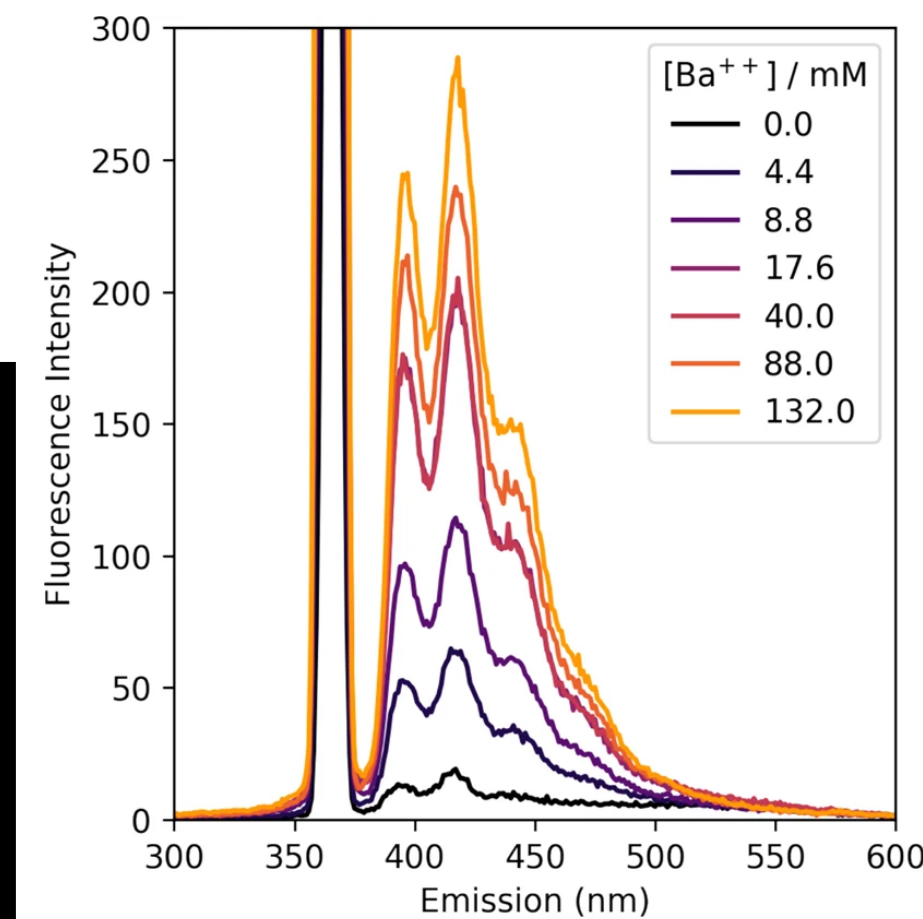
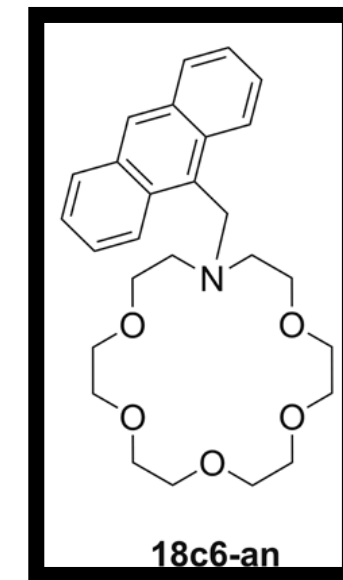
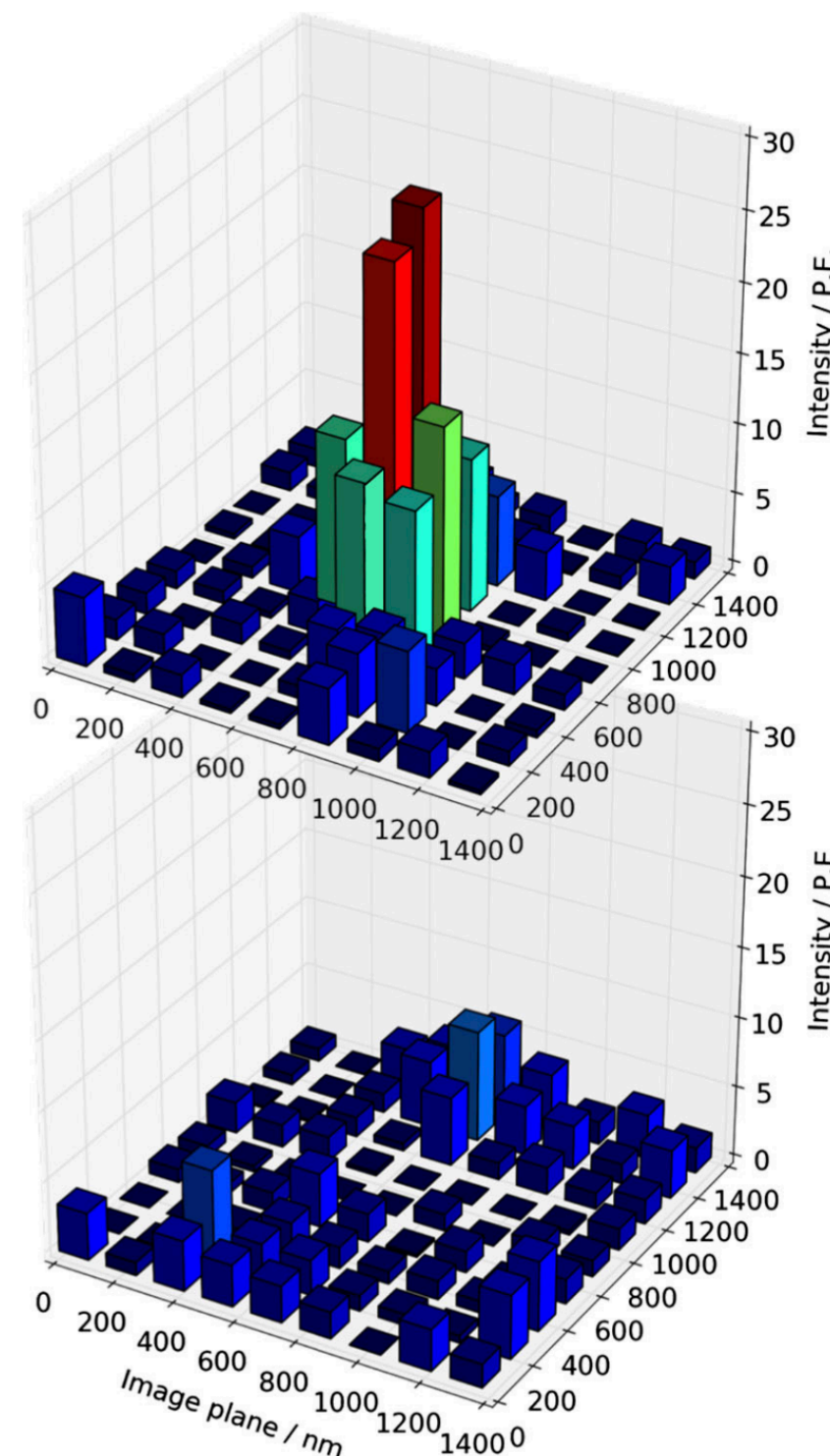
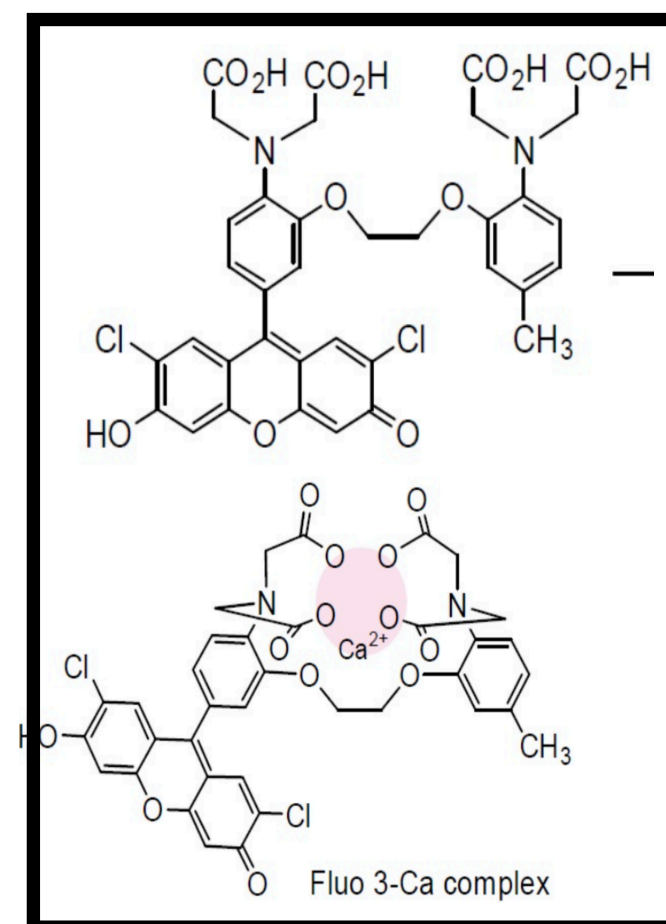


[b] *Nat. Commun.* 13, 7741 (2022)

SMFI with on-off molecules

Extensive research with several chemosensor families

- Studies in solution and dry surfaces



[a] JINST 11 P12011 (2016)

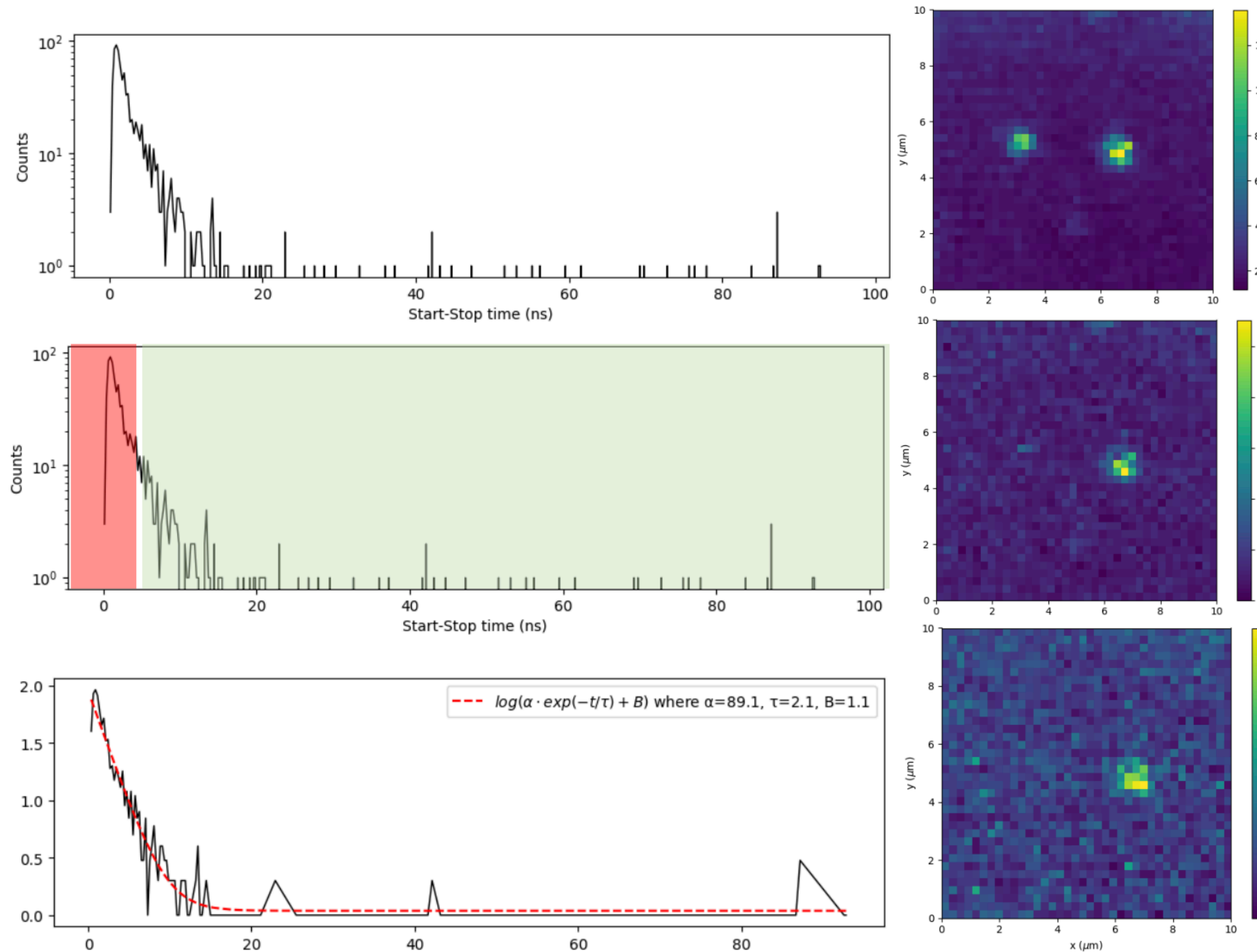
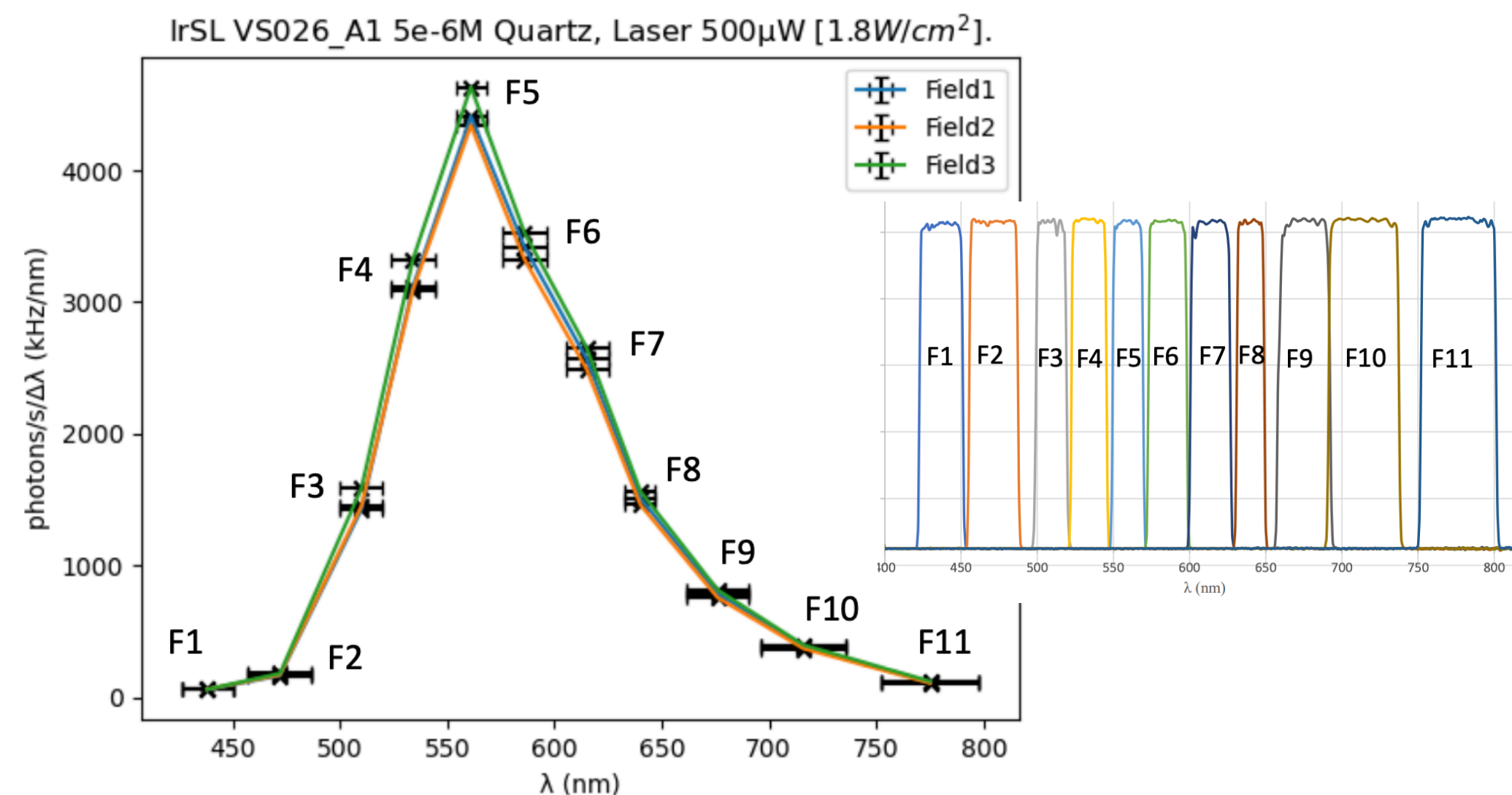
[b] Phys. Rev. Lett. 120, 132504 (2018)

[c] Sci. Rep. 9, 1, 15097 (2019)

[d] ACS Sens. 6, 1, 192–202 (2021)

Other approaches:

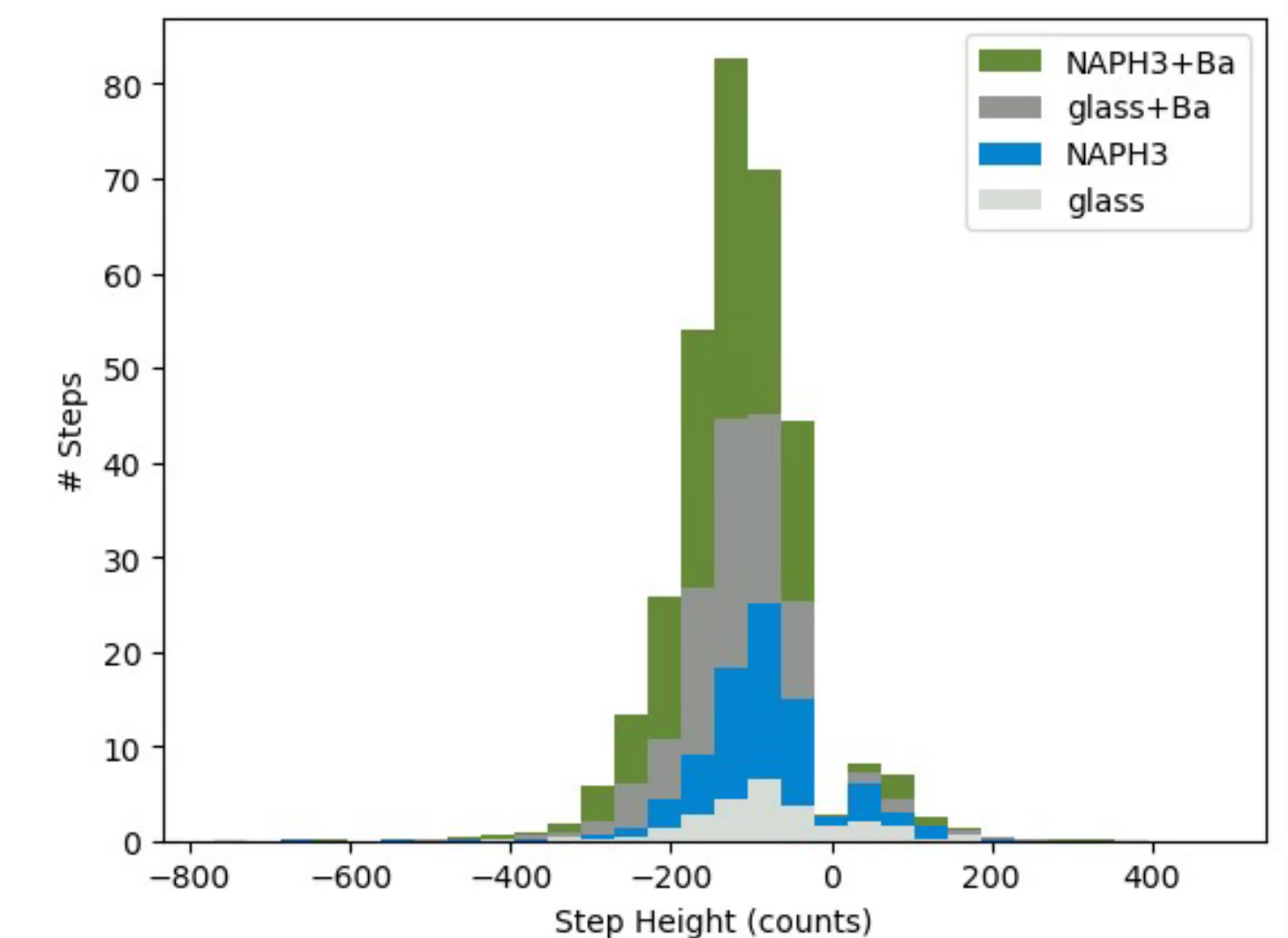
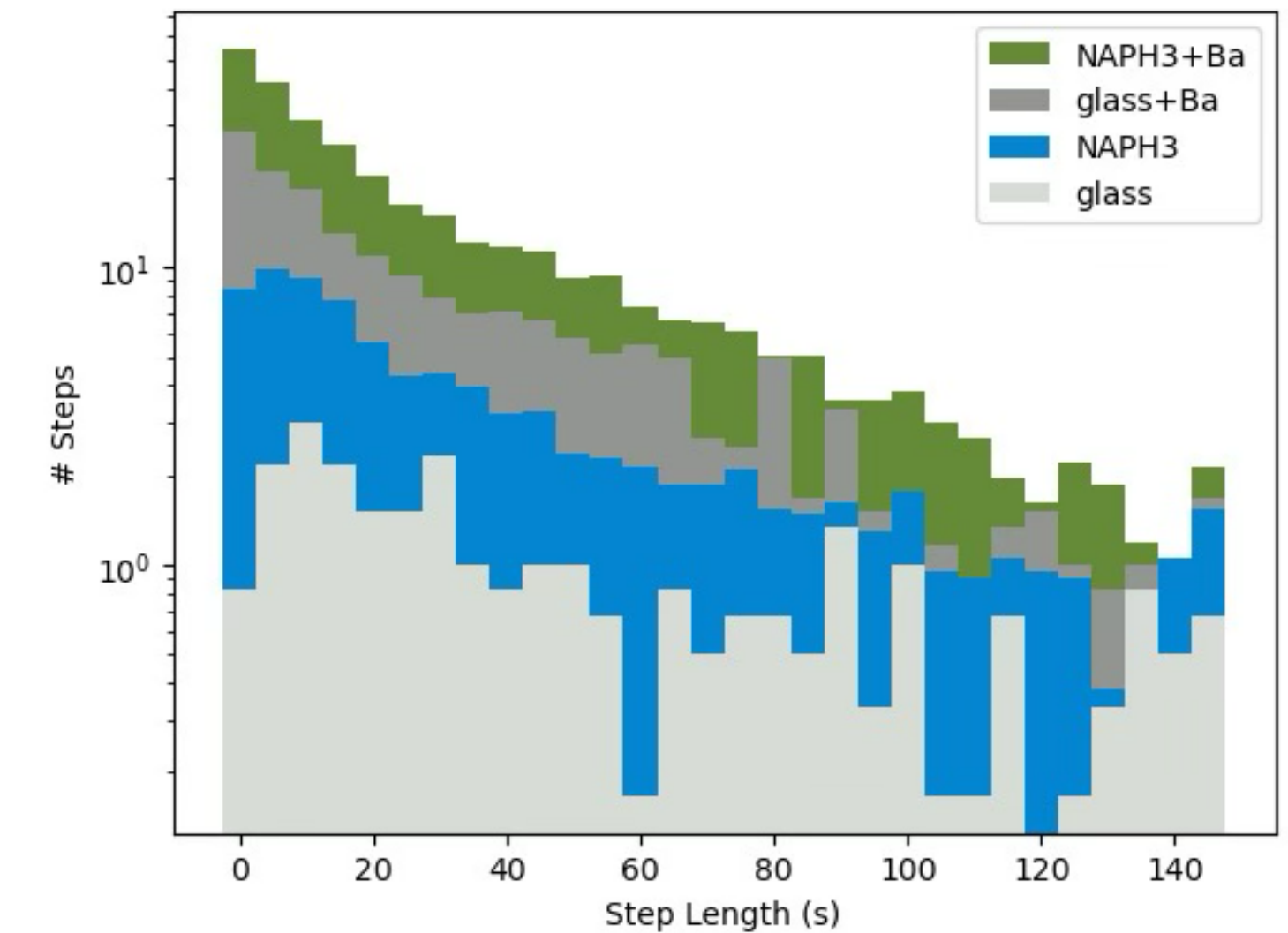
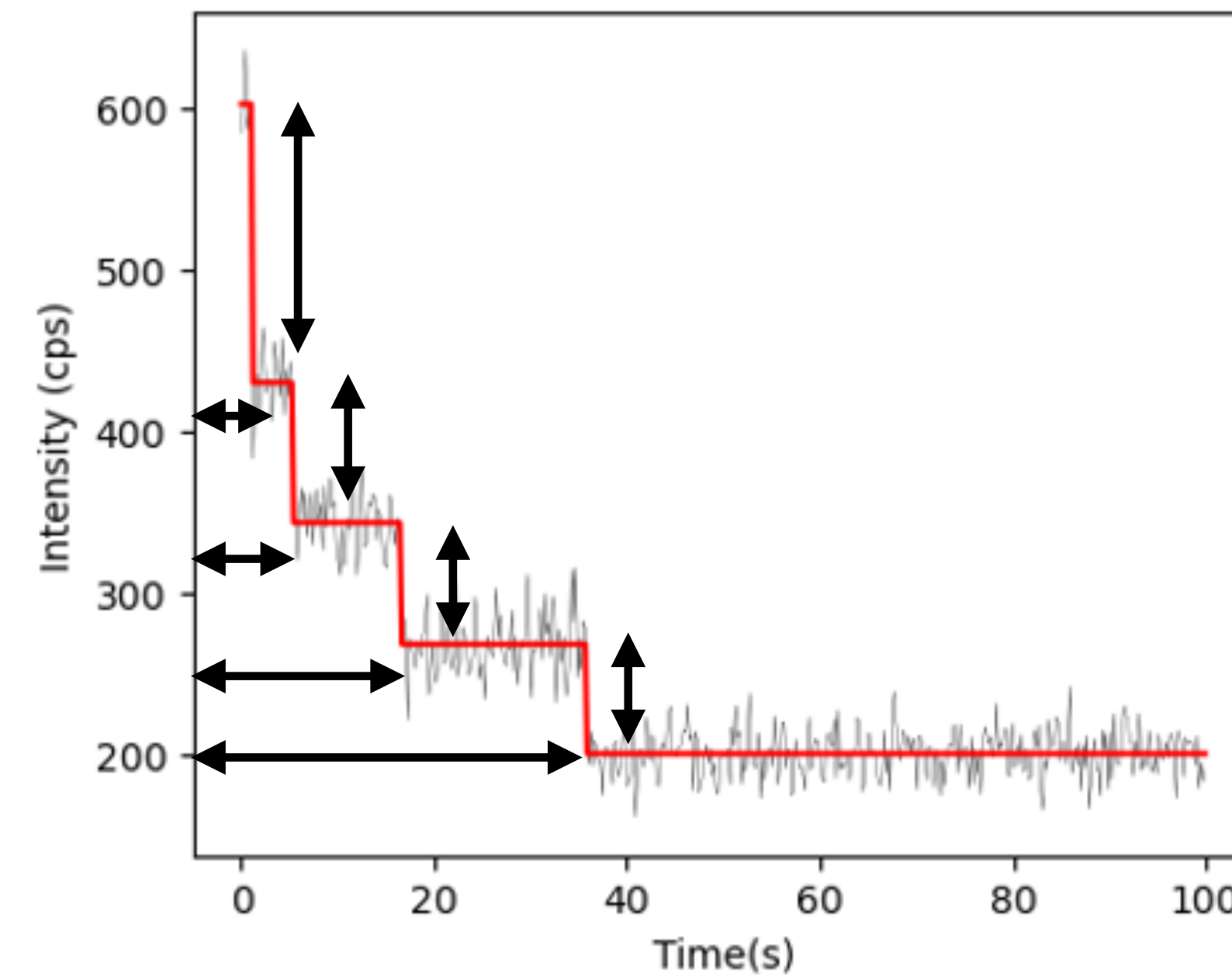
- TIRF vs epi-illumination, wide-field, confocal, 2PA.
- Emission spectrum reconstruction (bicolor molecules)
- Fluorescence-lifetime imaging microscopy (FLIM) for temporal molecules



Microscopy efforts

Statistics on step properties

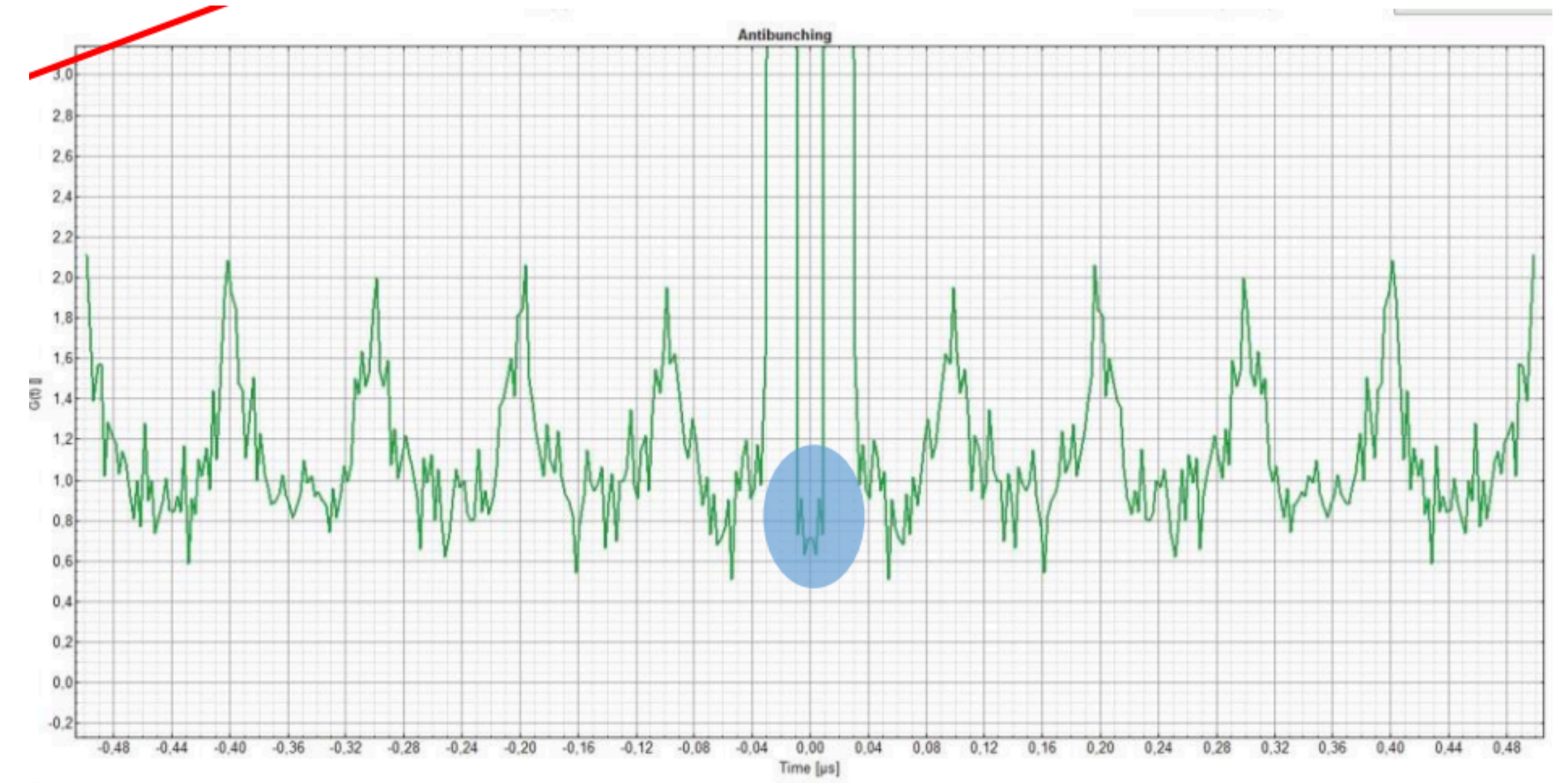
- SMFI and molecular orientation
- Photochemical studies from step distributions
- Development of efficient step finding algorithms



Ask
for more!

Other approaches:

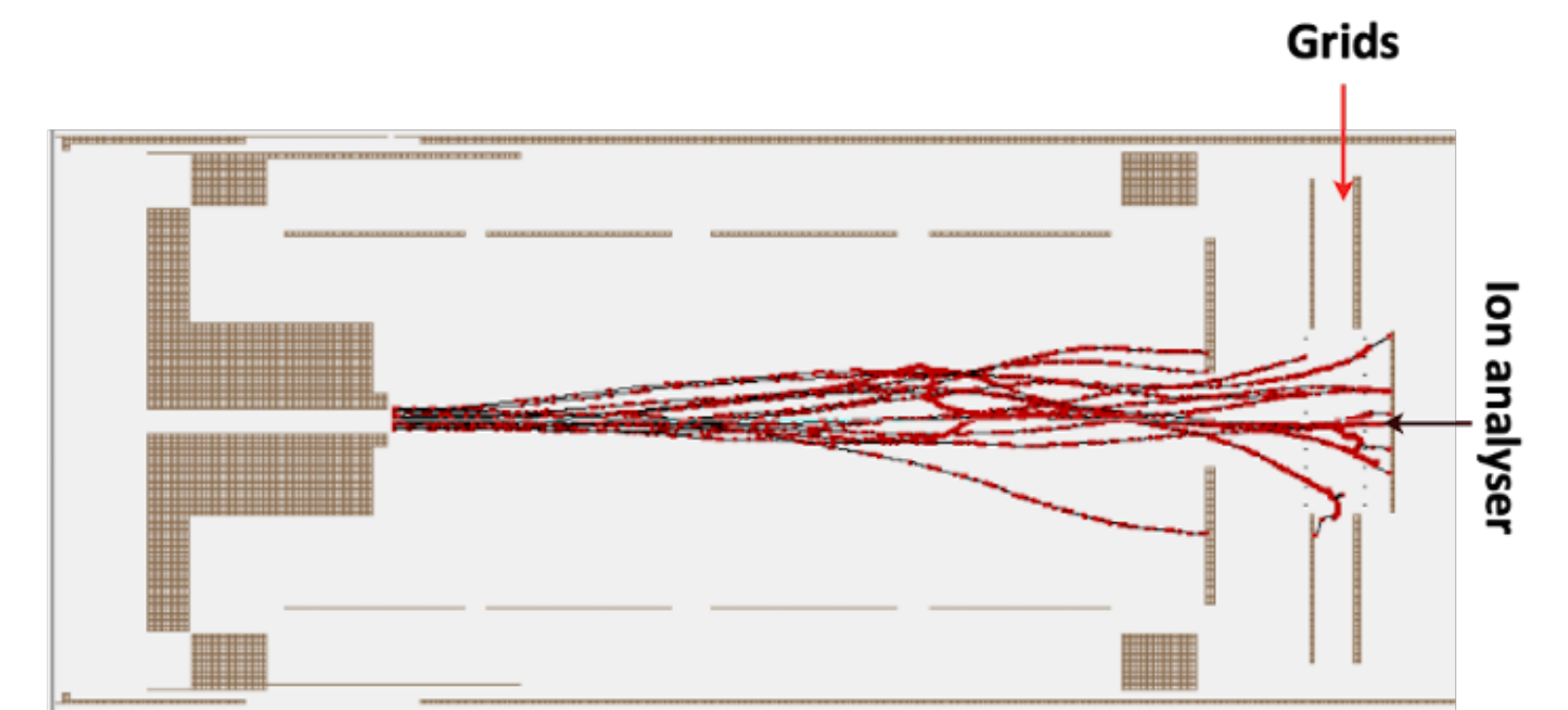
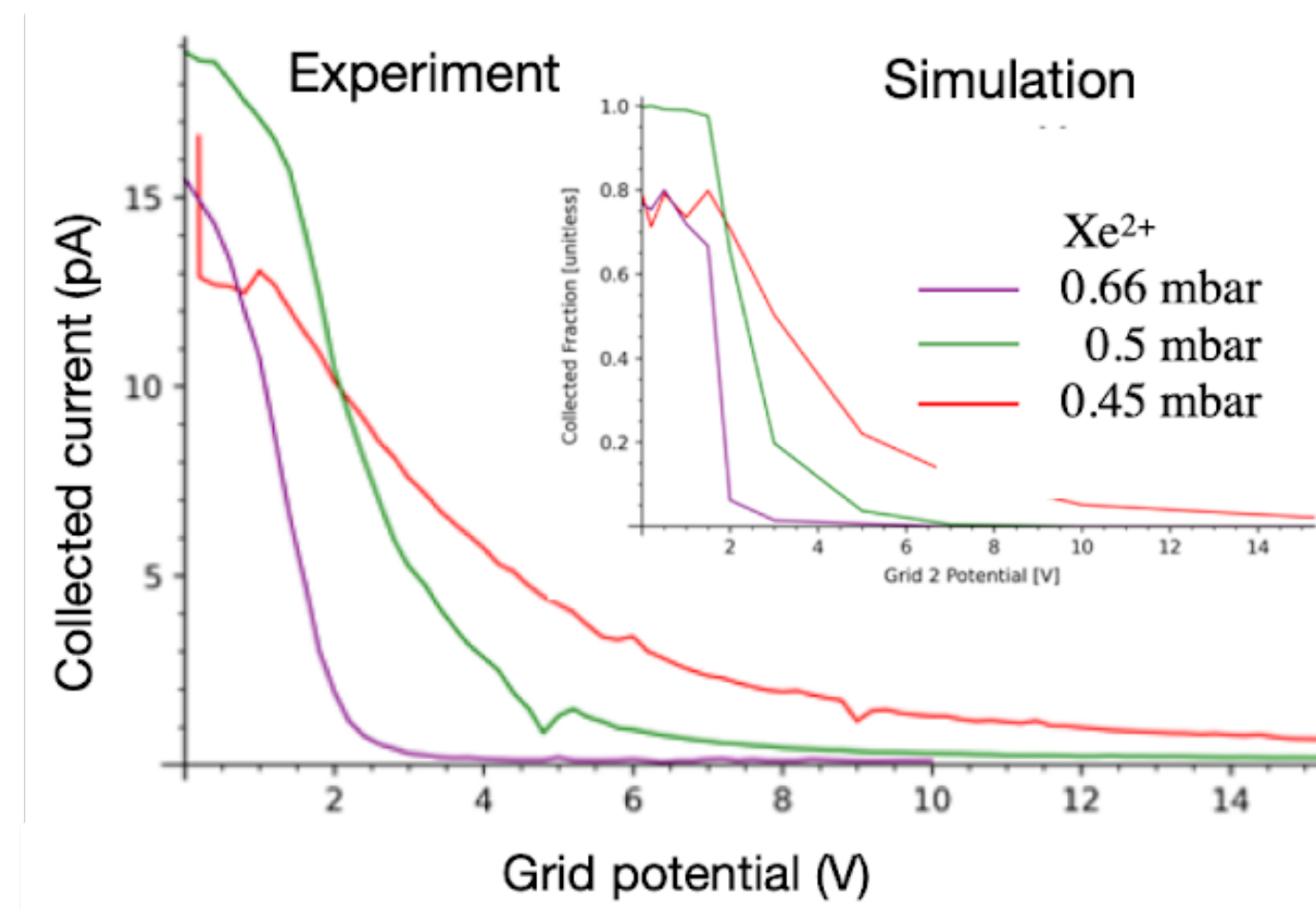
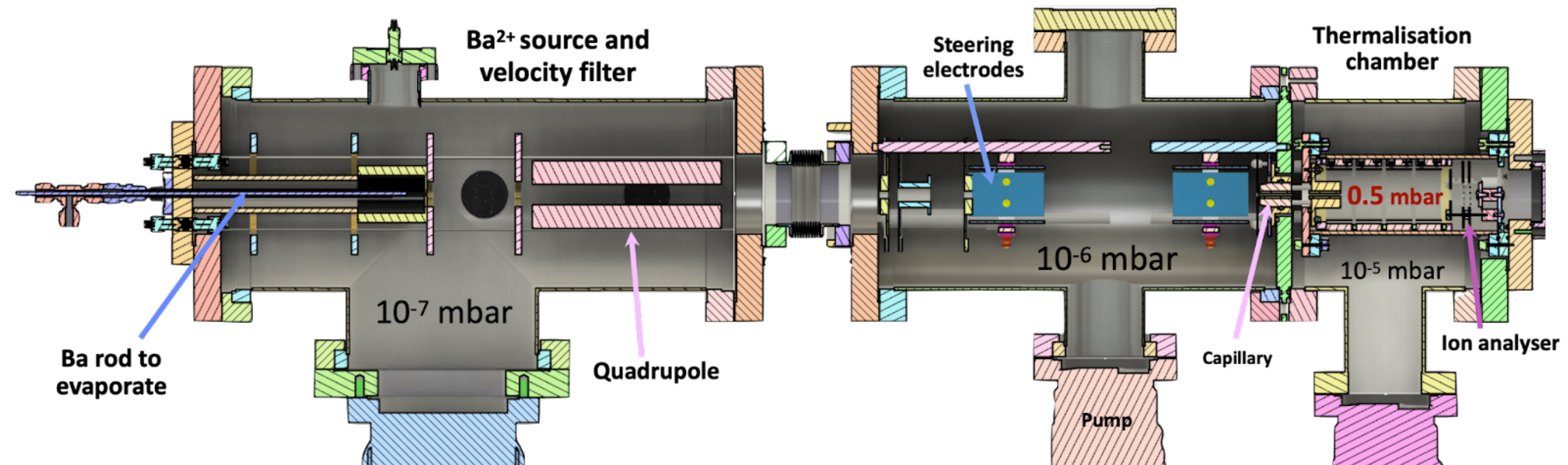
- Illumination through TIRF vs epi-illumination (wide-field, confocal), 2PA.
- Emission spectrum reconstruction (bicolor molecules)
- Fluorescence-lifetime imaging microscopy (FLIM) for temporal molecules
- Non-destructive SMFI: Antibunching



Ba²⁺ beam in low pressure

Thermal ions

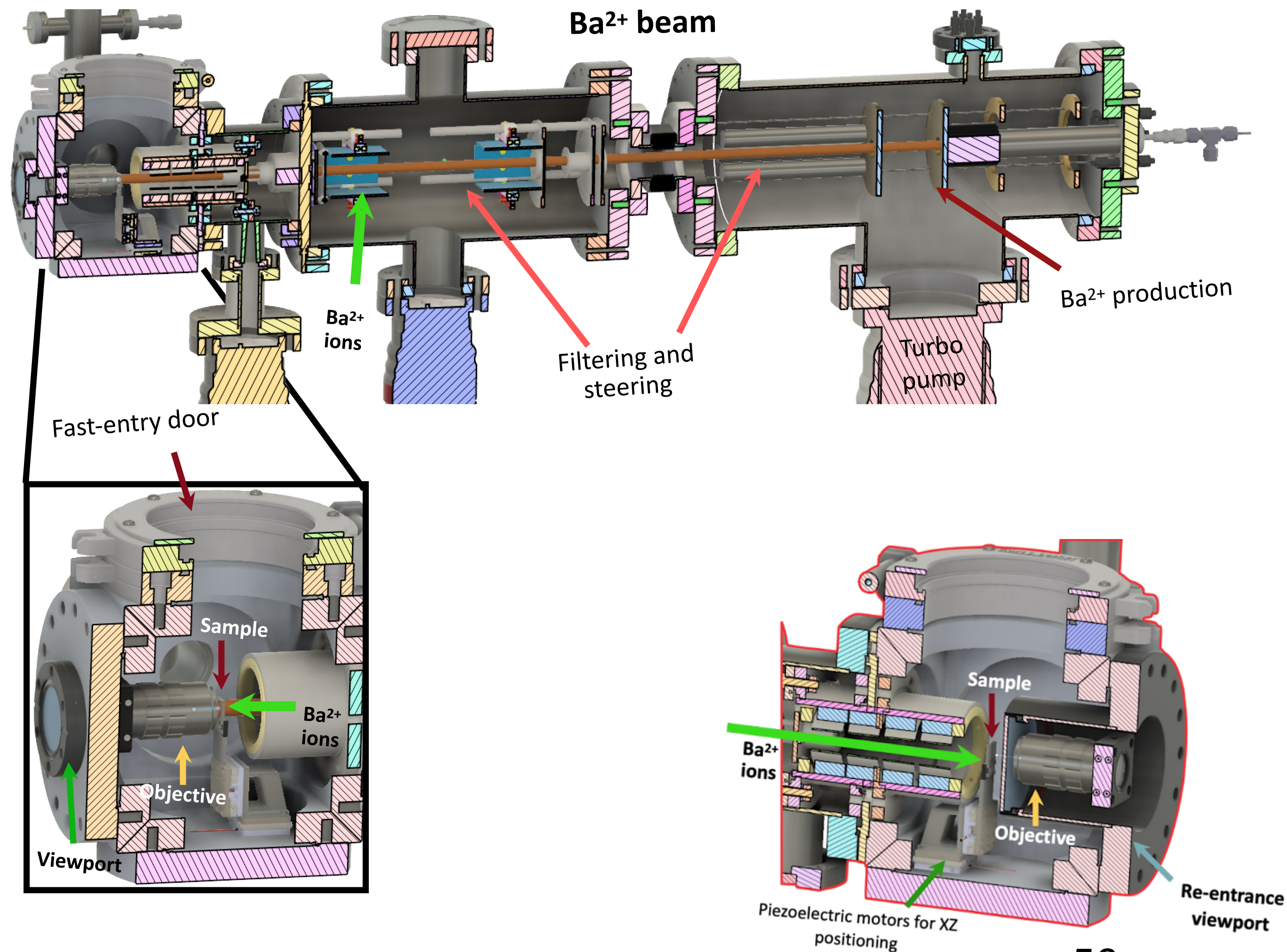
- Sublimation of Ba → filtering from other ions
- Steering into thermalization chamber though differential pumping
- Collision with GHe at ~0.5 mbar
- Beam characterization with Xe²⁺
- Optimization with Ba²⁺ undergoing



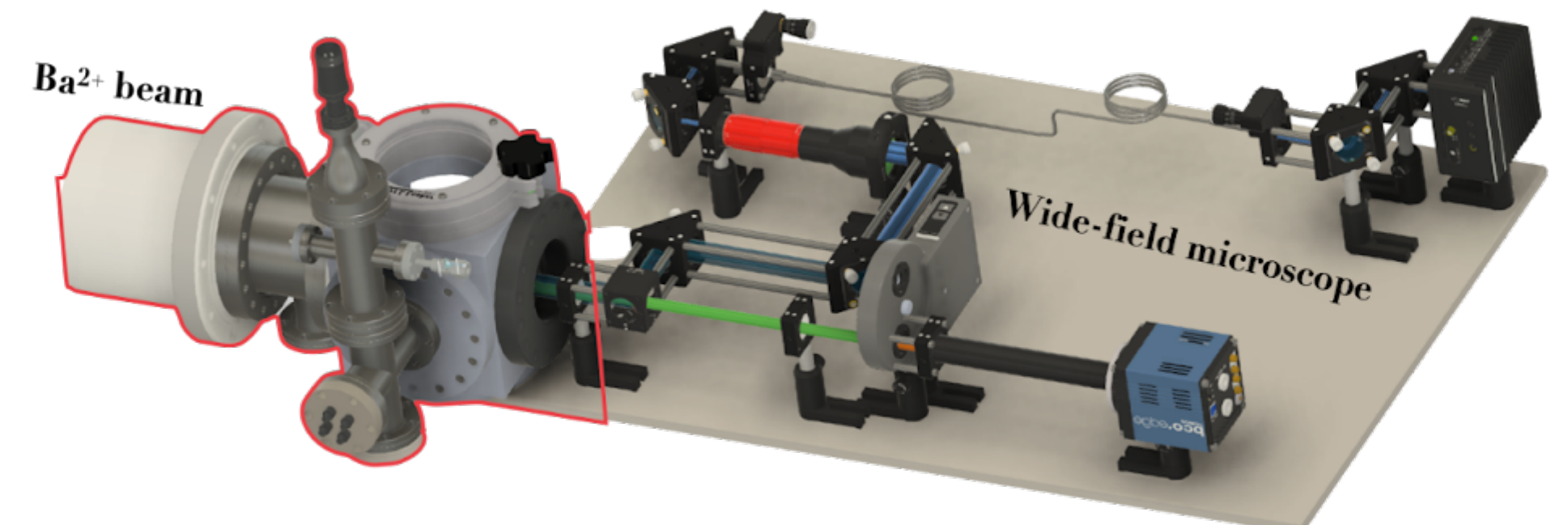
Thermalization chamber simulation, ions passing through the capillary can reach the analyser

Ba²⁺ beam in low pressure

Coupling to microscope



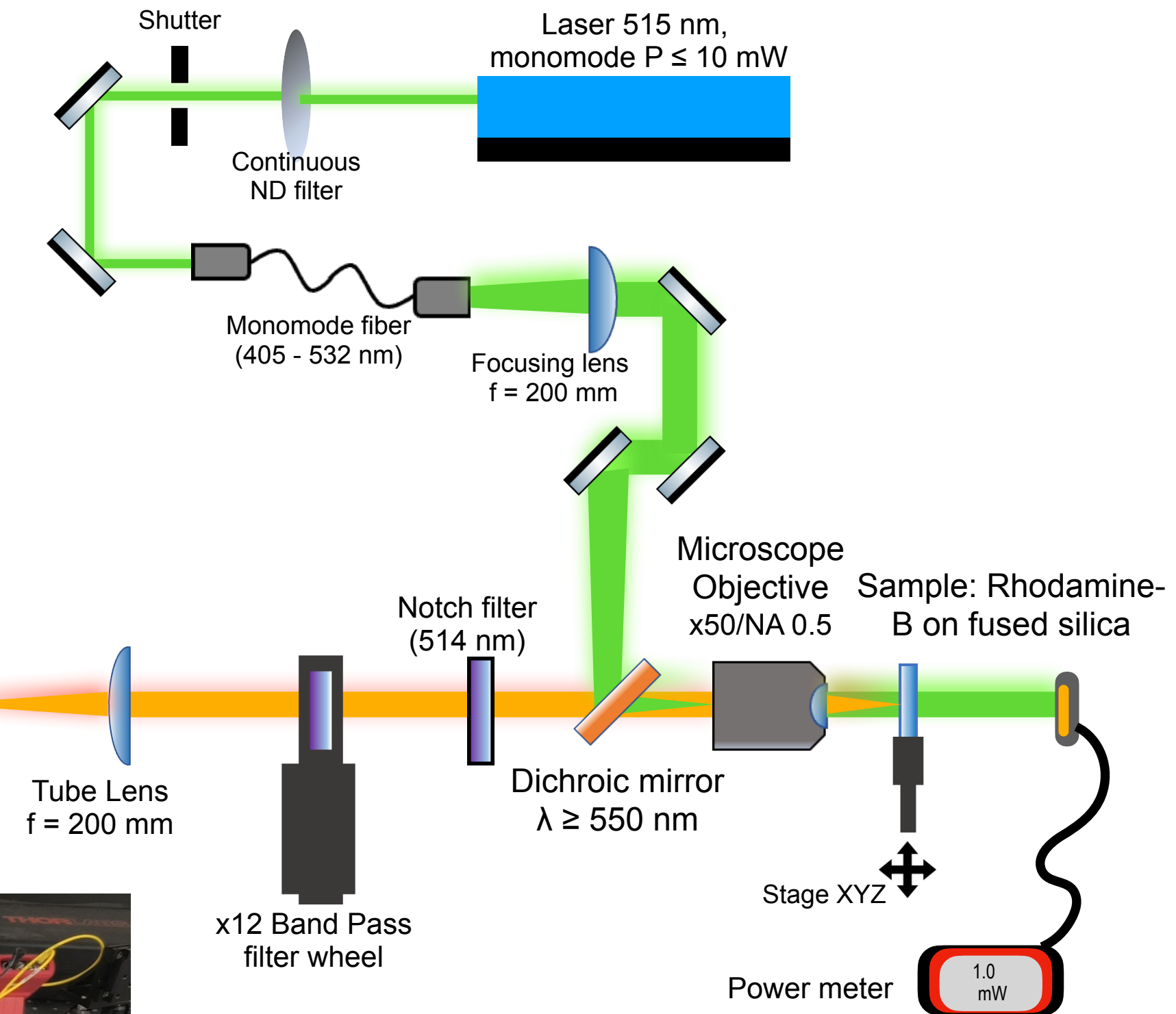
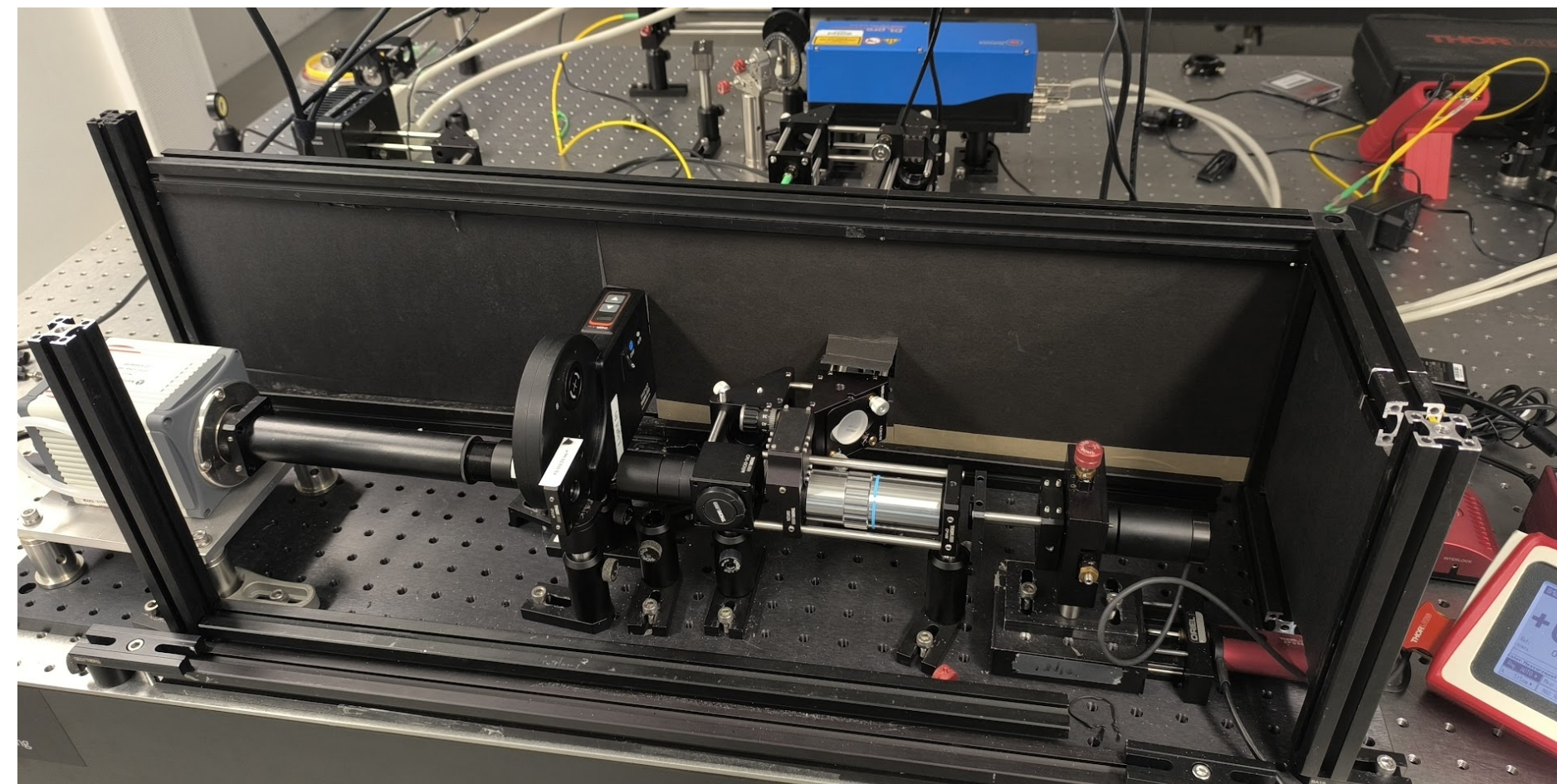
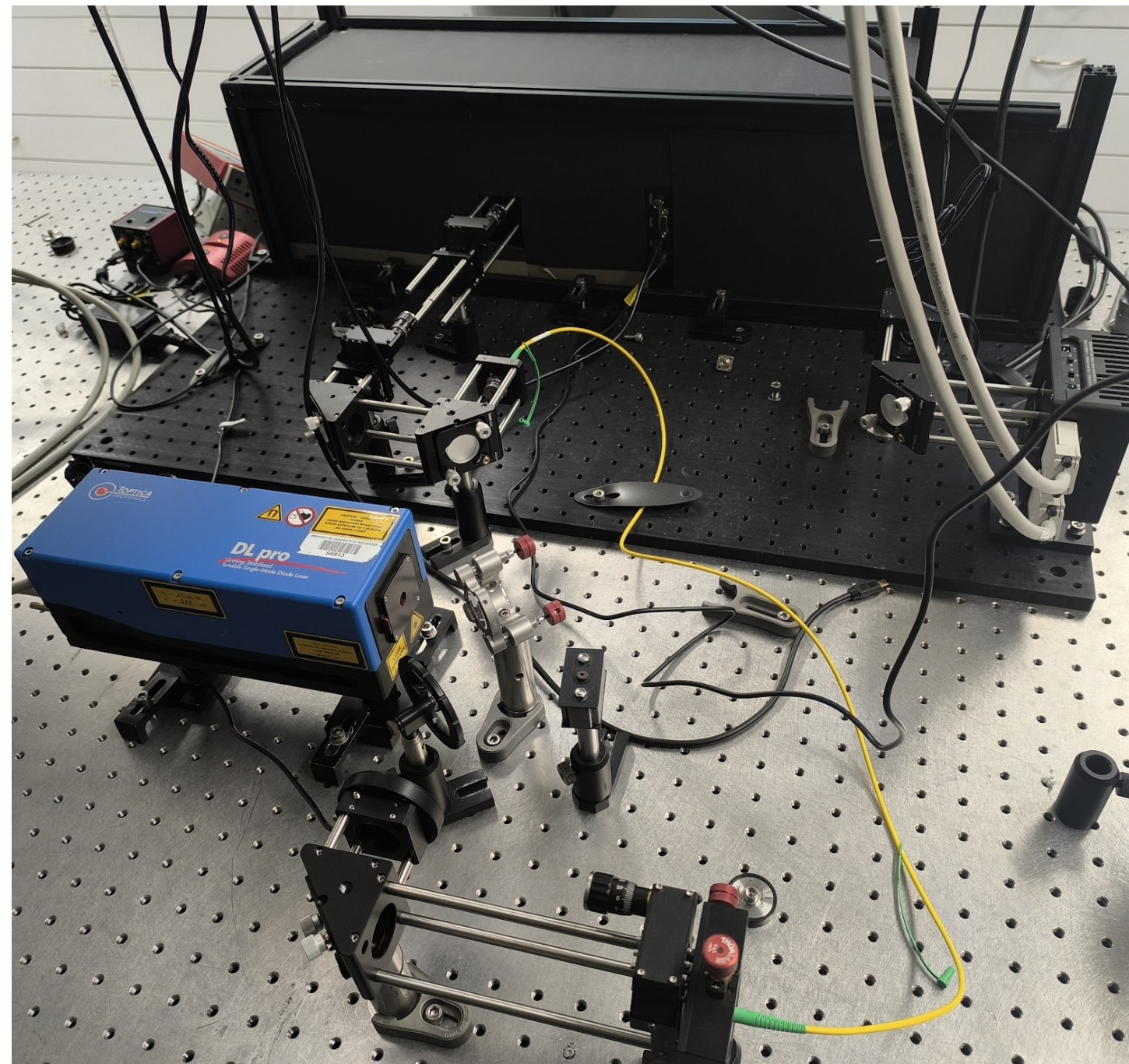
- Beam coupled to chamber with MO, working at low pressure ~1mbar
- Alternative: long WD MO outside the chamber
- Reconciling with high NA limitation is challenging



Ba²⁺ beam in low pressure

Coupling to microscope

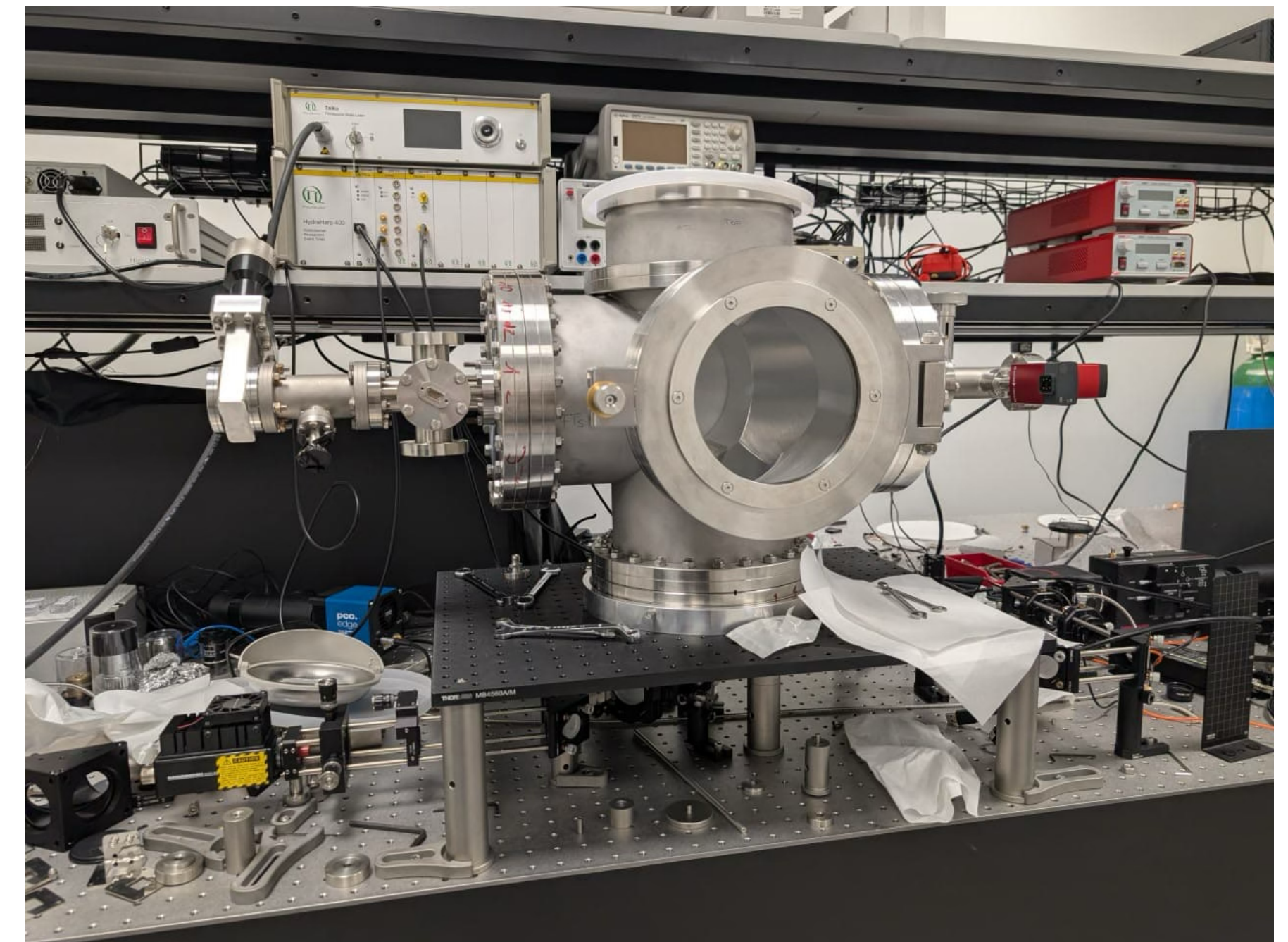
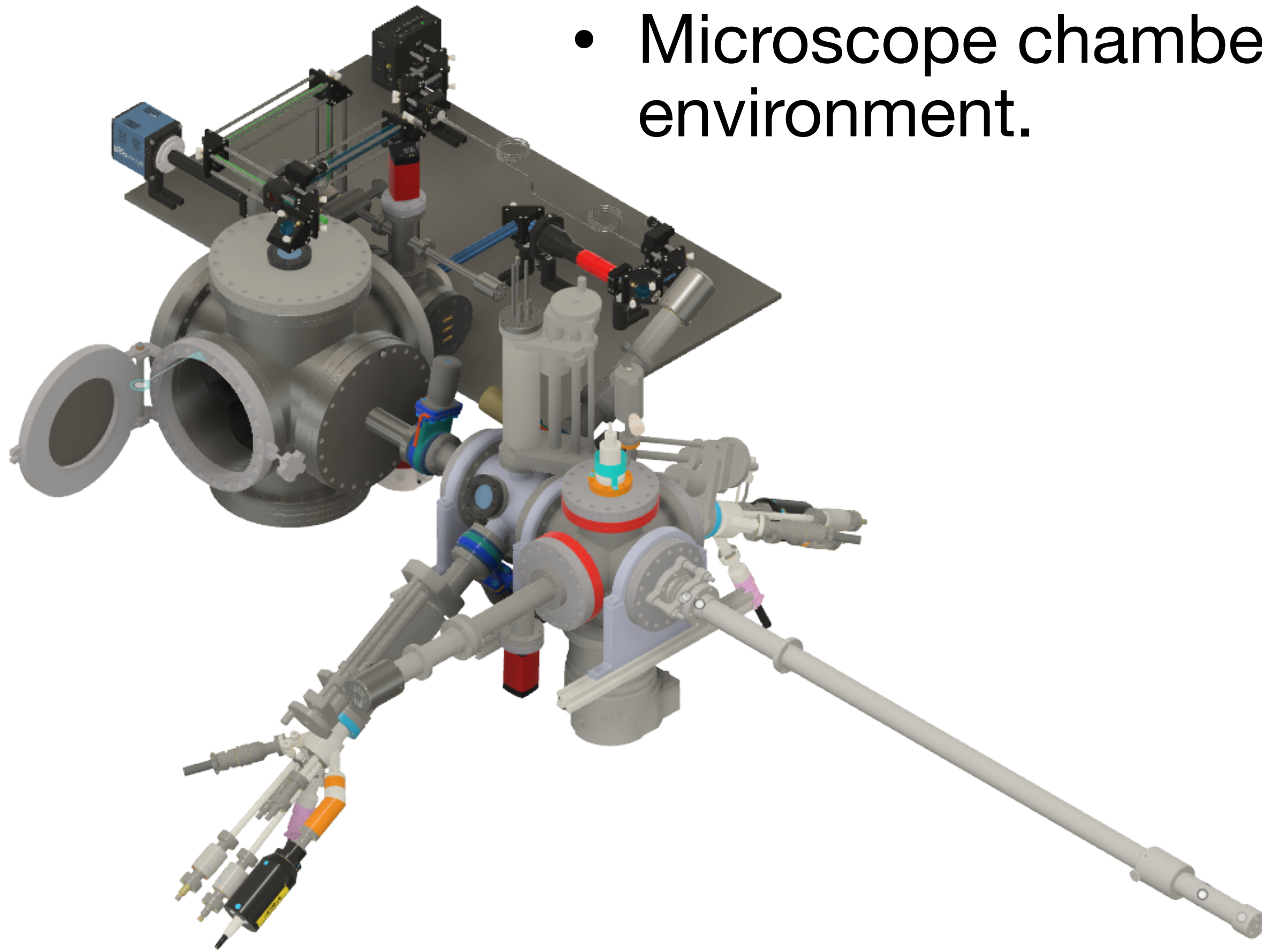
- Wide-field microscope for coupling to beam



Microscopy efforts

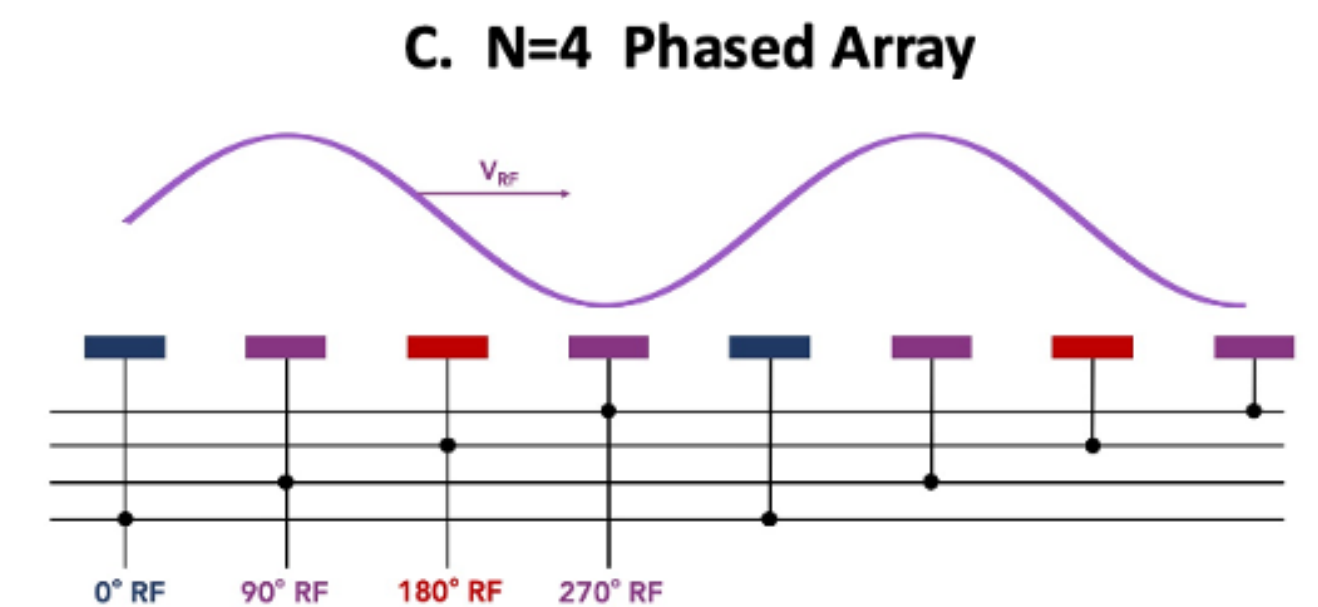
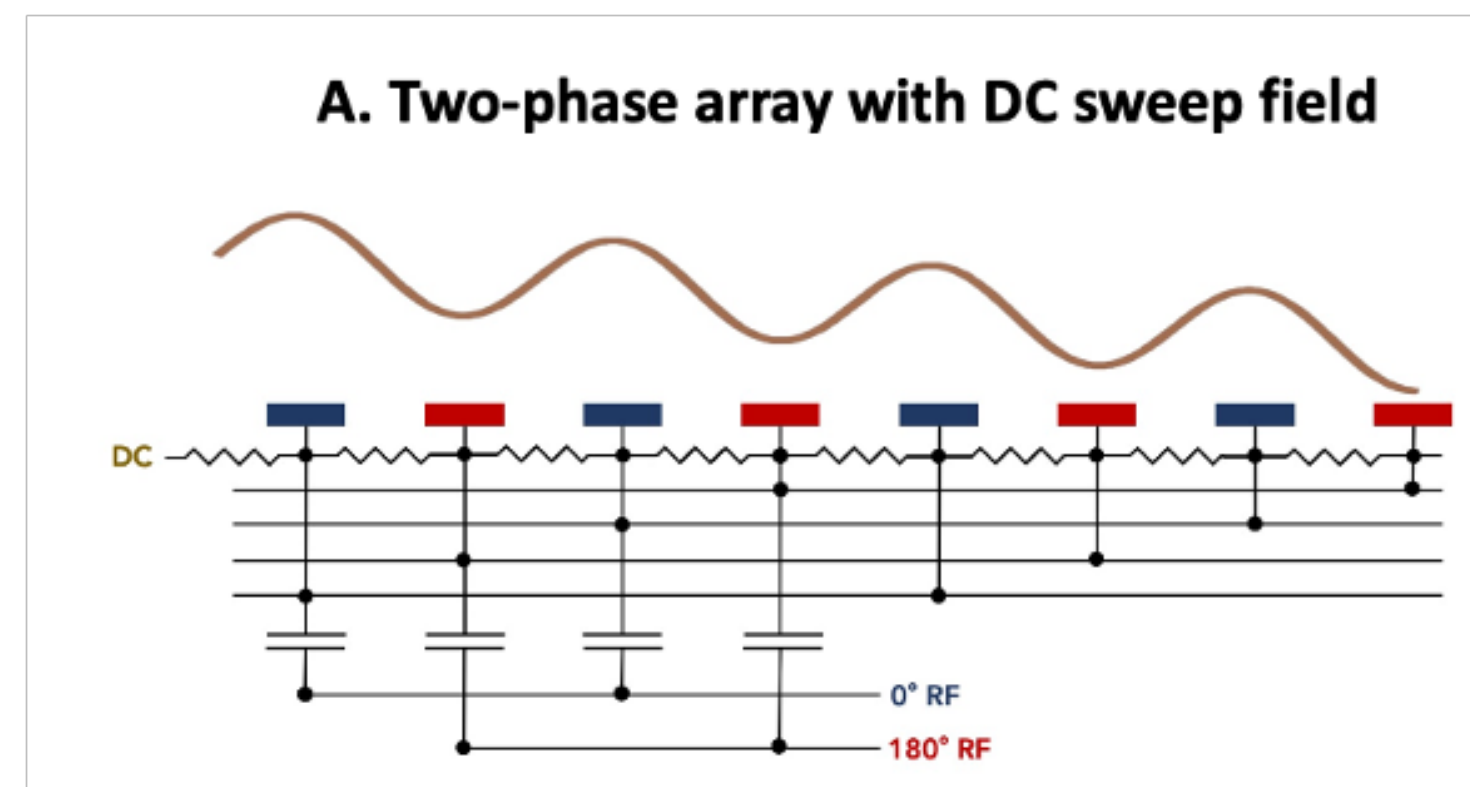
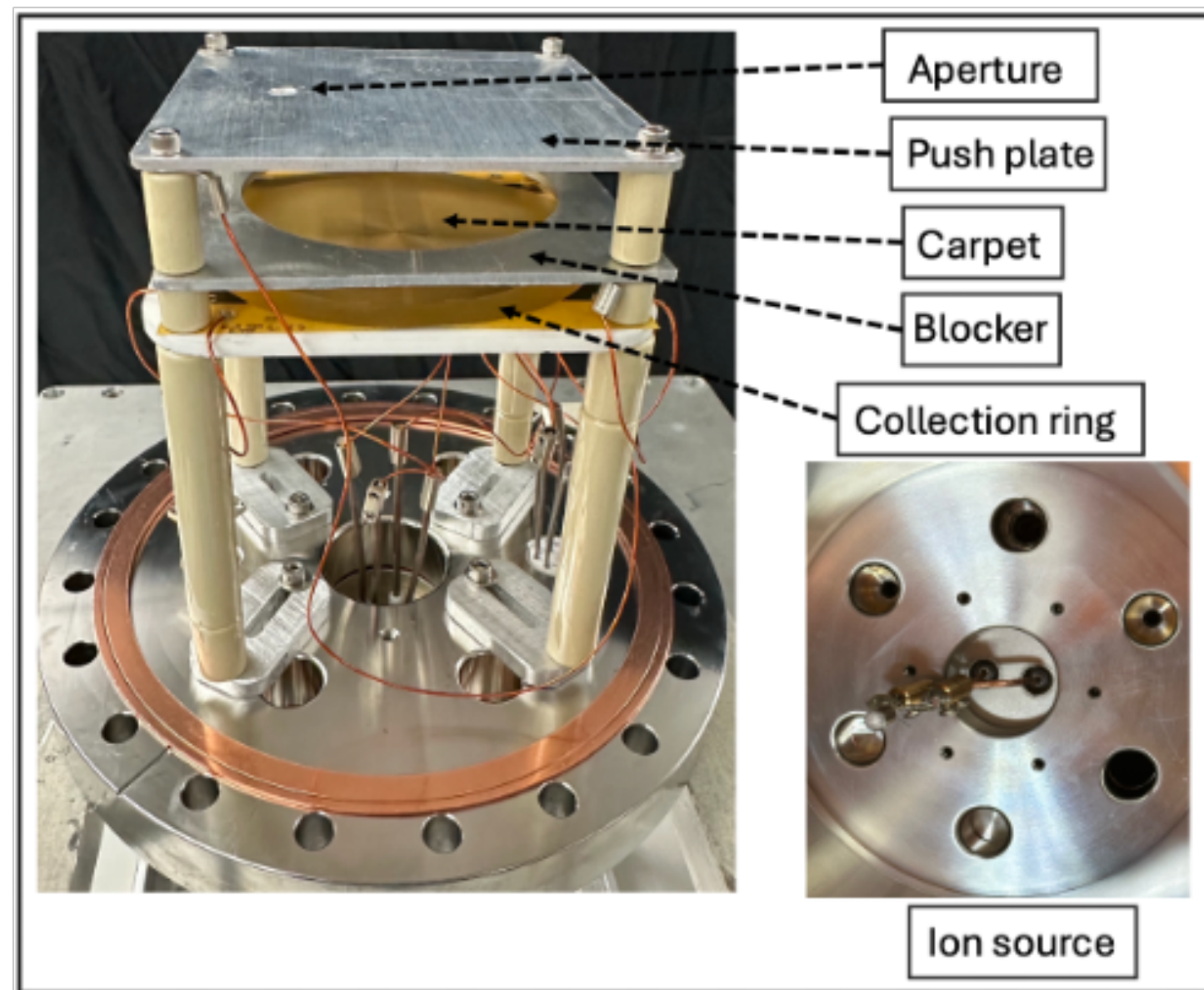
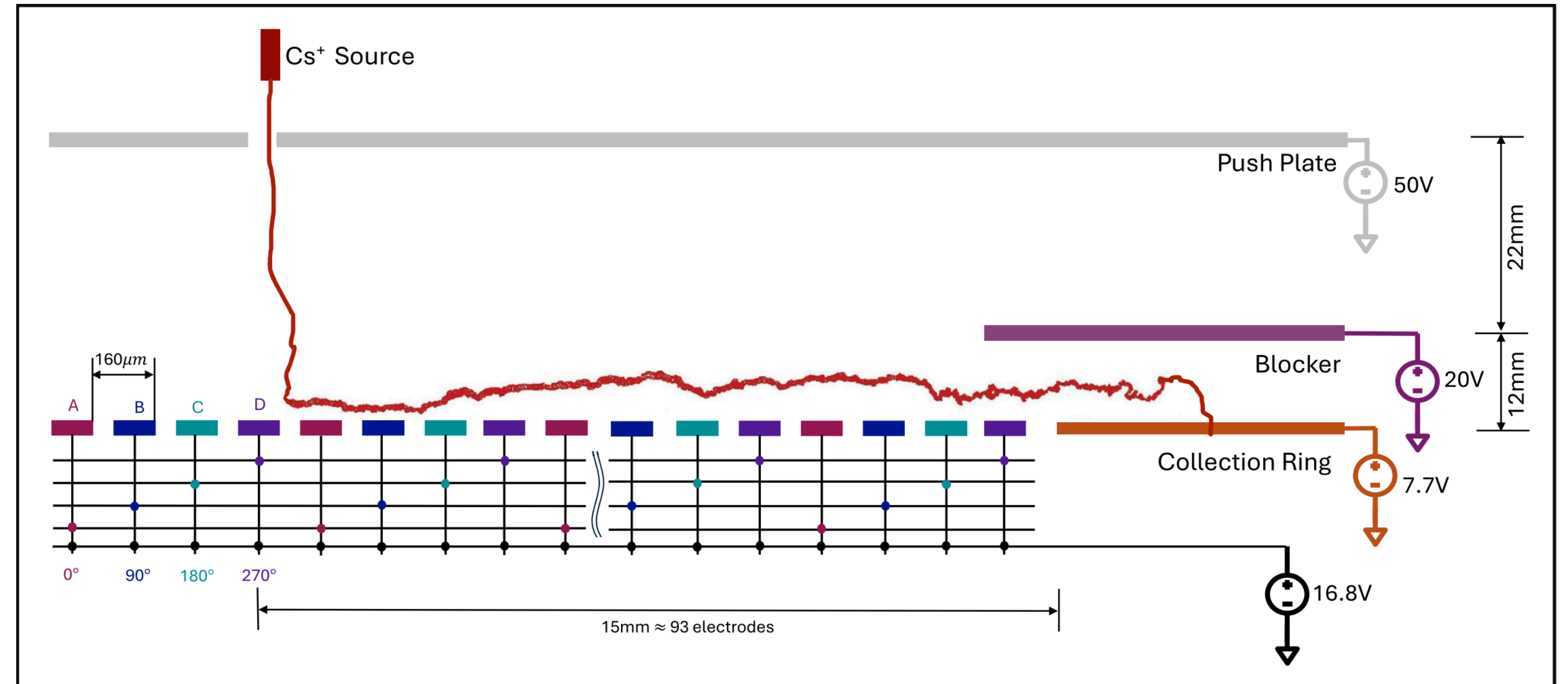
Coupling to Ba/molecular evaporators

- Next step: couple to evaporation chamber (in vacuum)
- Grow Monolayer and deposit Ba^{2+} in UHV
- Microscope chamber can operate in controlled 1 atm Ar environment.



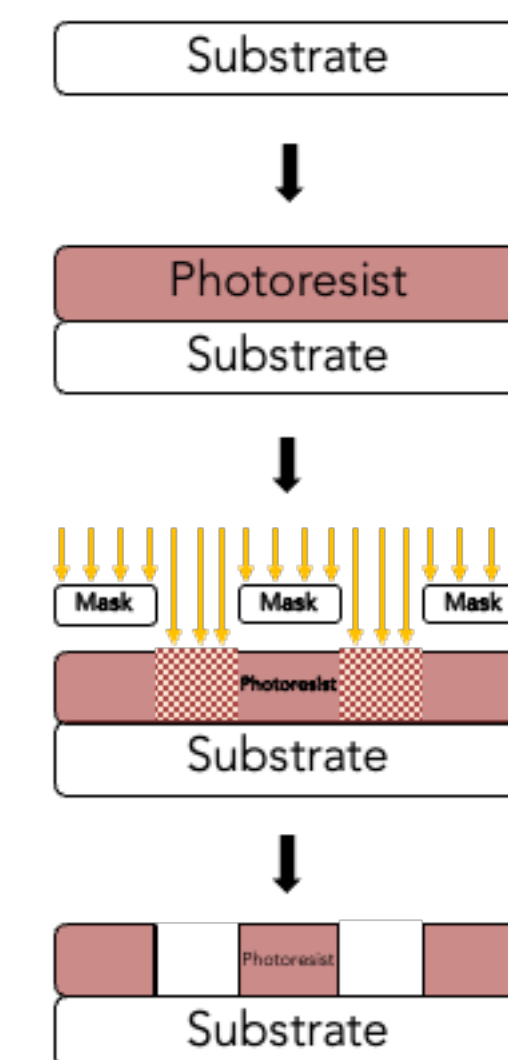
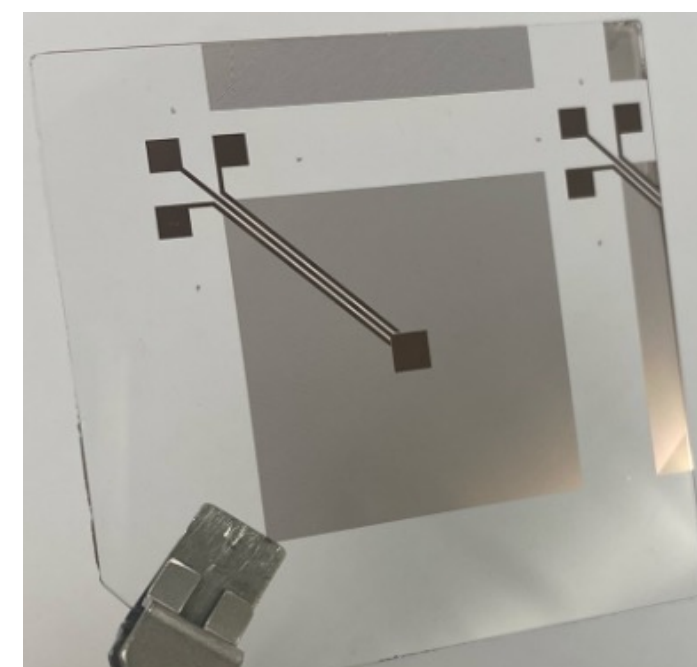
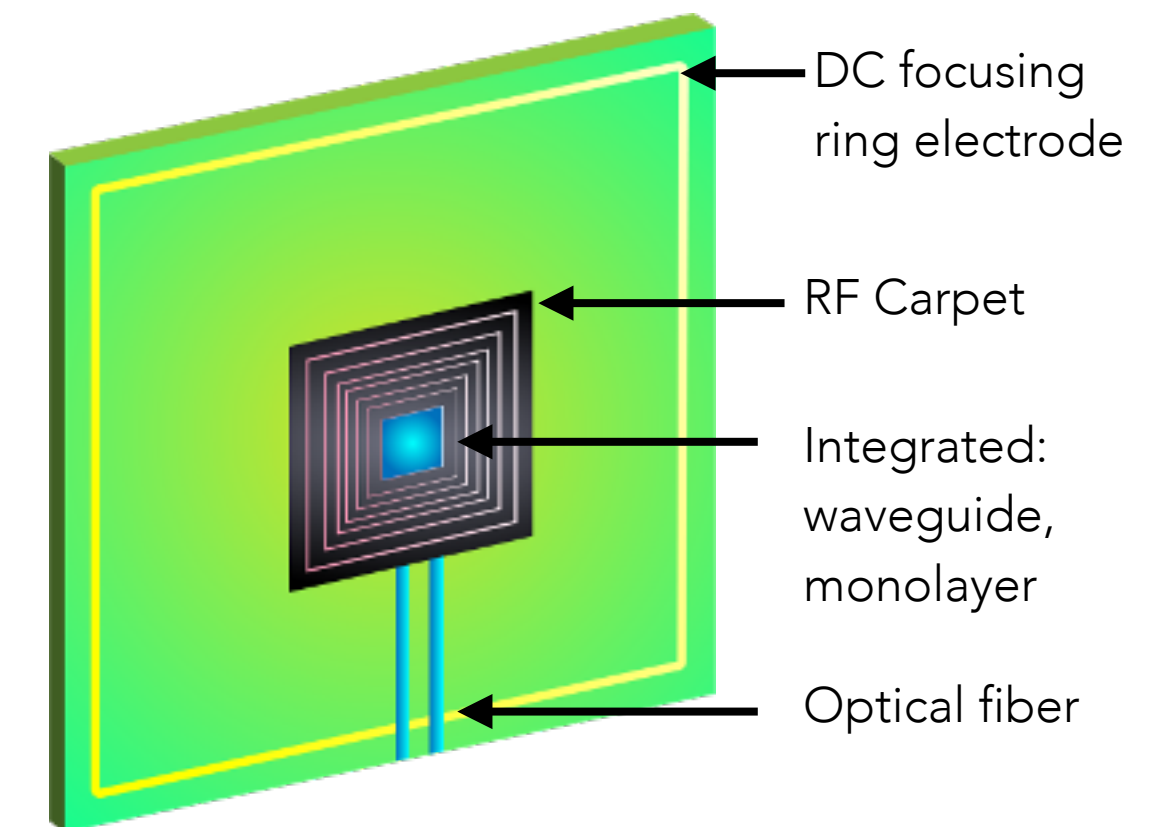
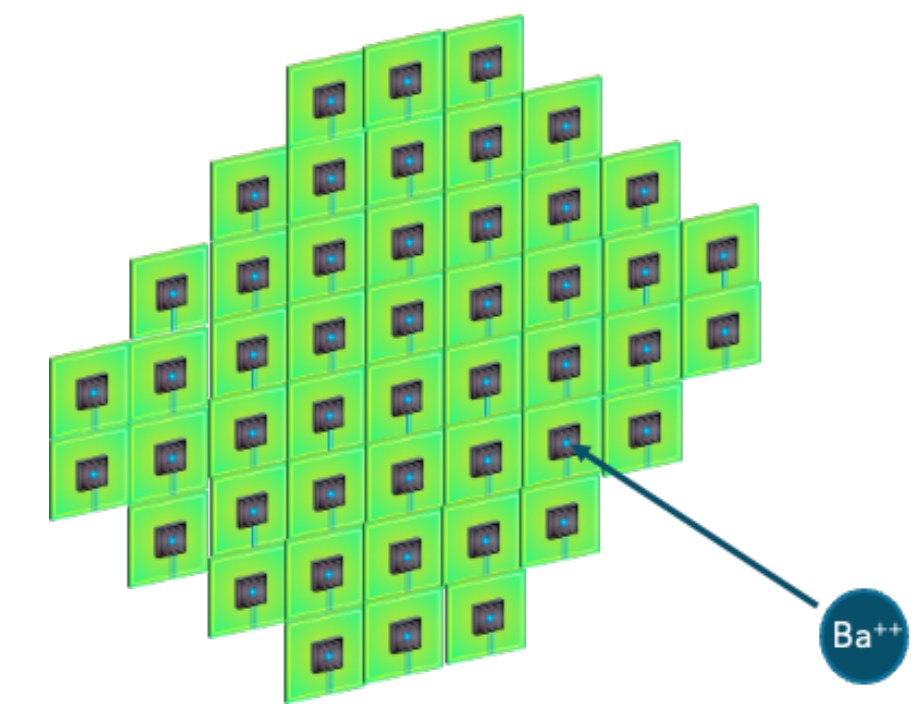
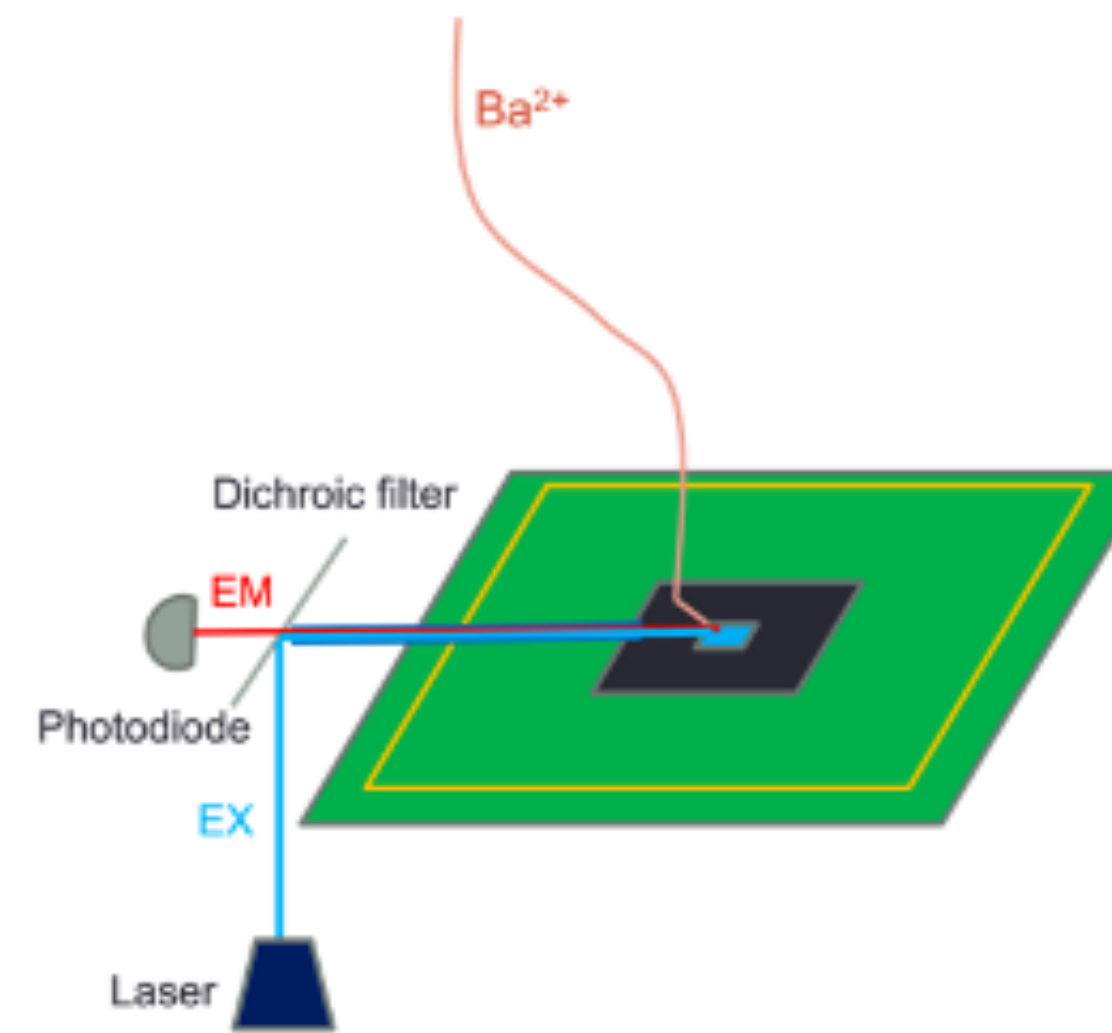
Ba²⁺ guide in GXe with RF carpet

- Using Cs⁺ as Ba⁺ stand-ins
- 160 μm electrode pitch, at 2 MHz, up to 300 V
- Maximum efficiency for four-phase mode, as predicted from simulation



Ba²⁺ collection and sensing: vBit

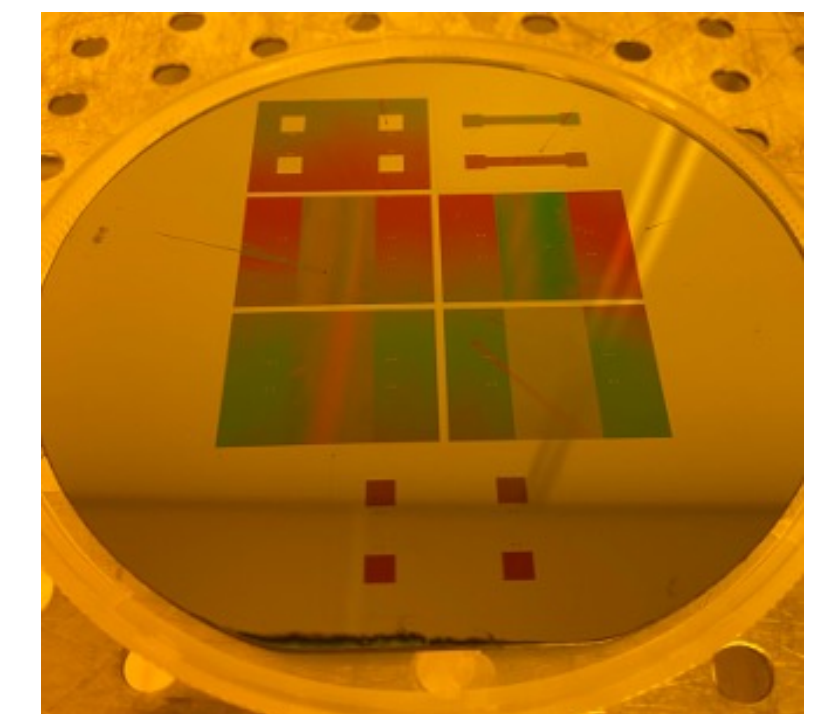
- Array of small-pitch RF carpets with molecular sensing region: vBIT
- Optimization of production pipeline is being finished
- Working on integrating with Fused silica and spin-coated molecules



Apply photoresist

Expose to UV light

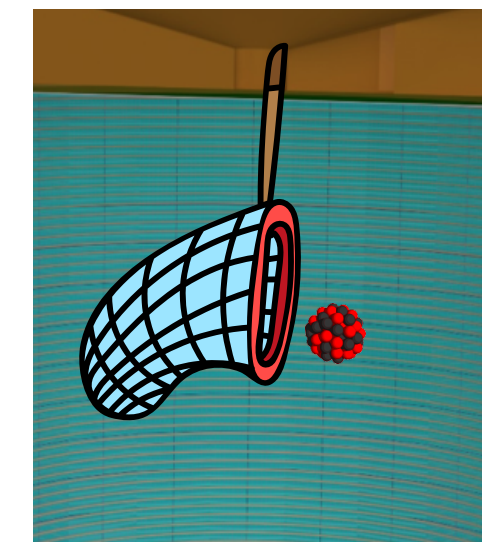
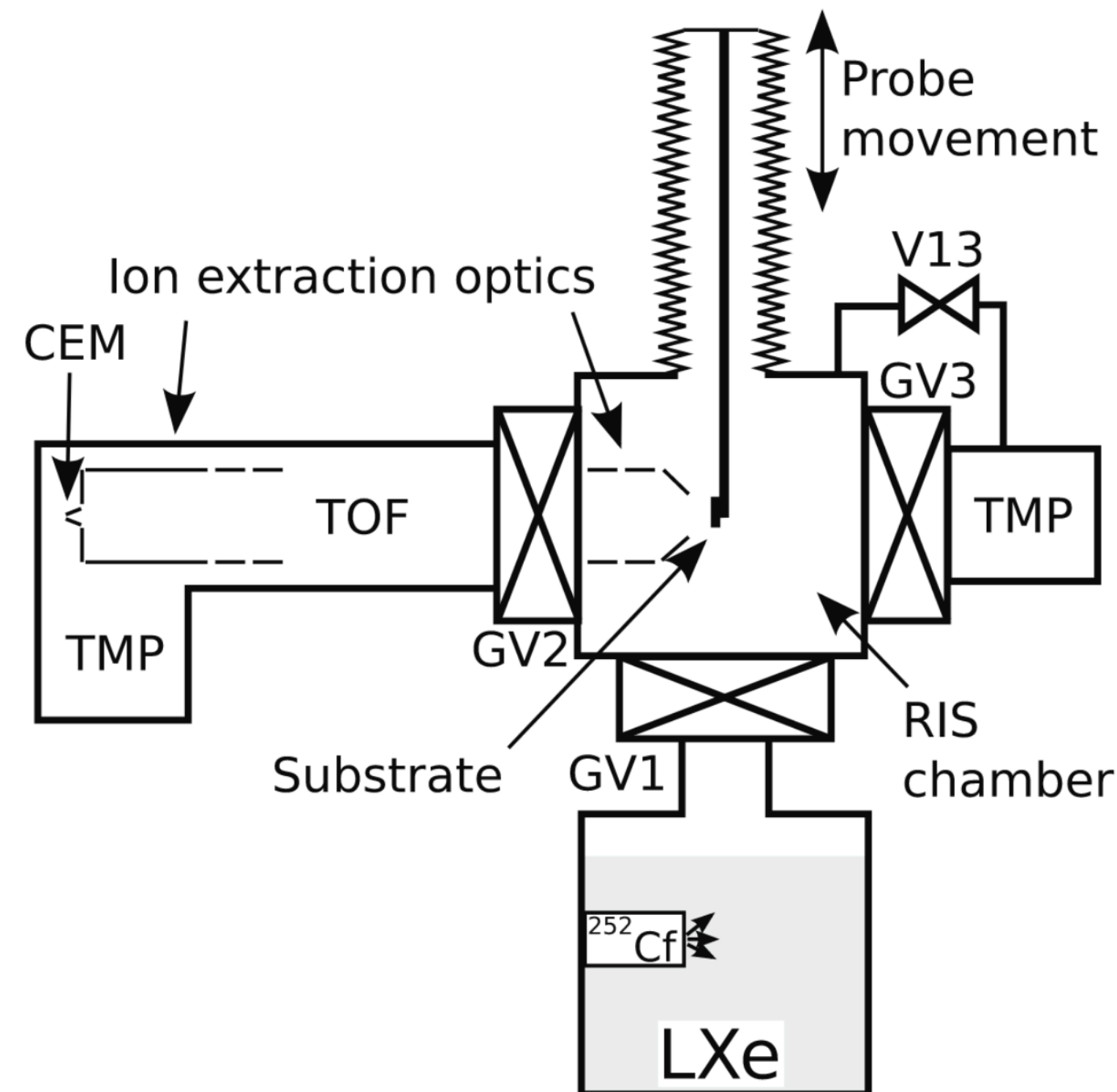
Apply developer



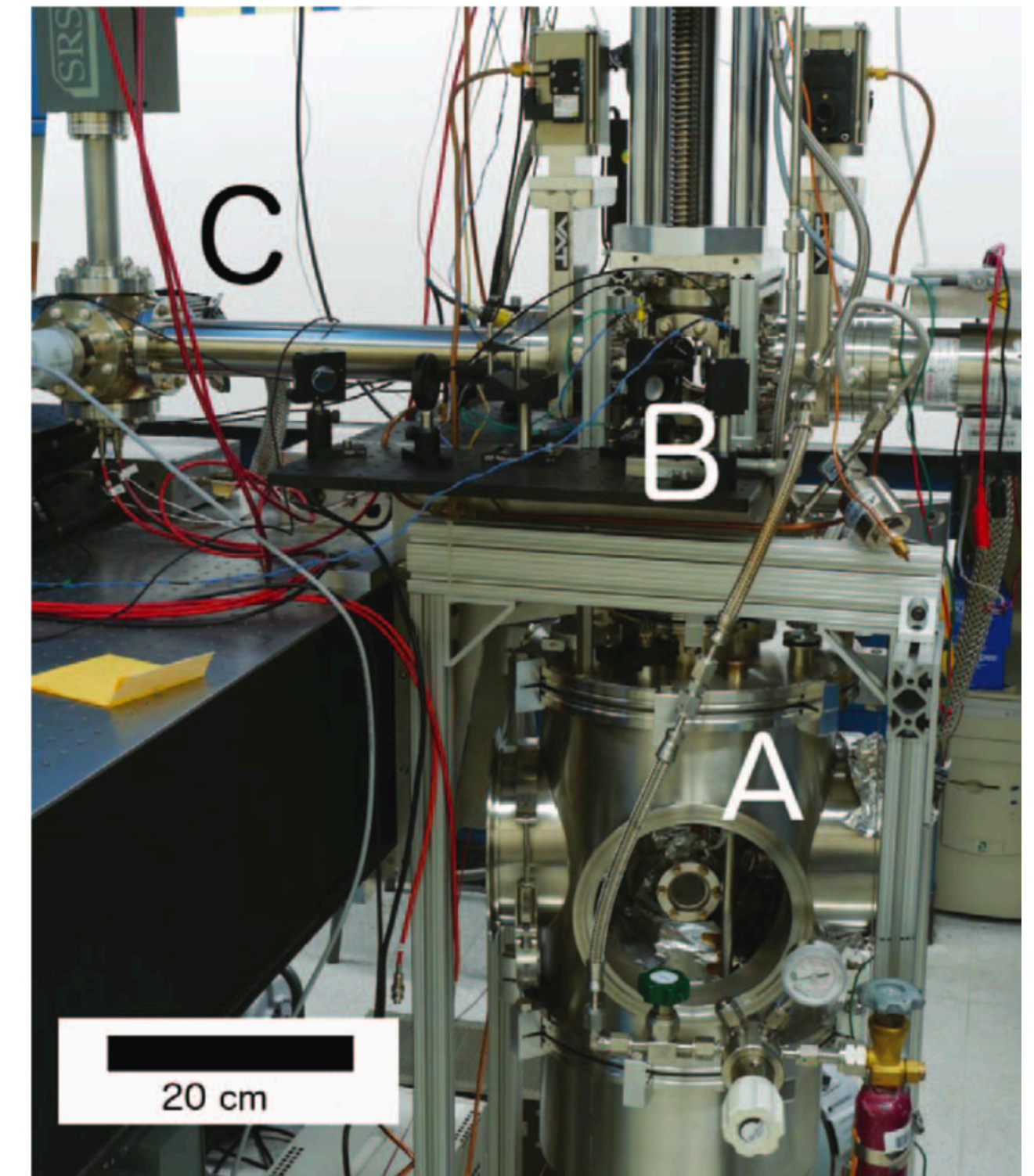
Backup slides (nEXO)

Extraction with cryoprobe

- ^{252}Cf source in LXe TPC
- Laser Induced Thermal Desorption (LITD) of Ba from probe
- Detection through Resonance Ionization Spectroscopy (RIS) and Time of Flight (TOF) Spectrometry



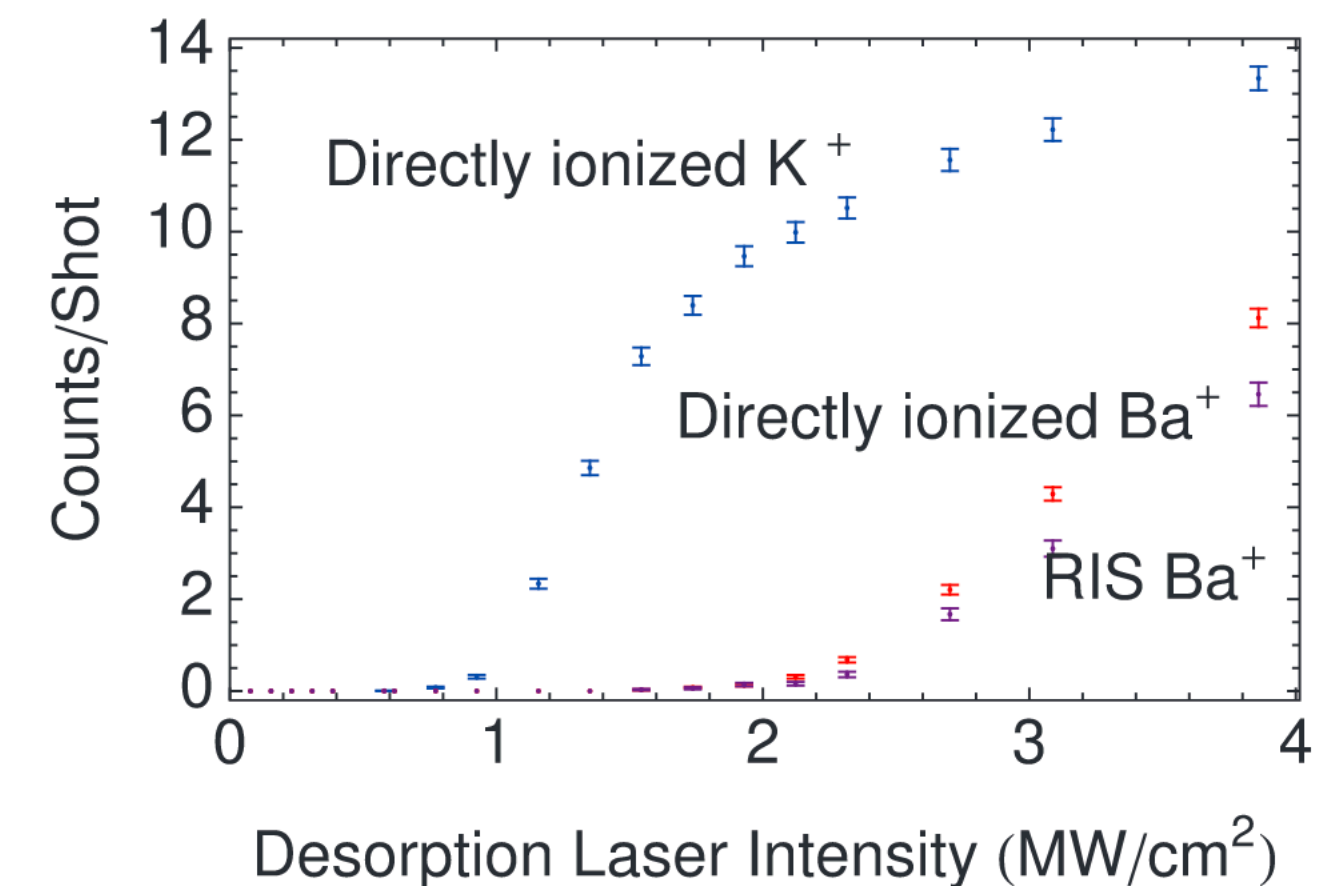
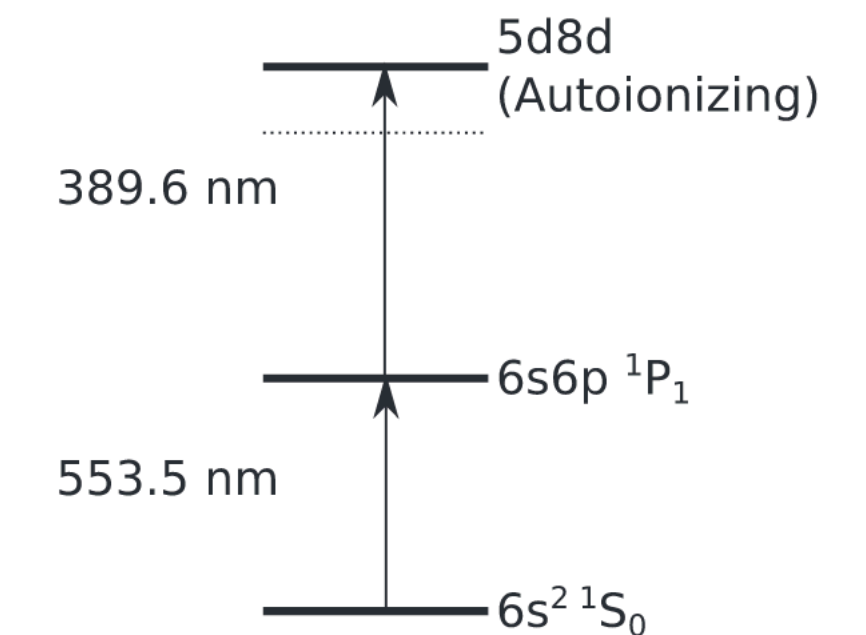
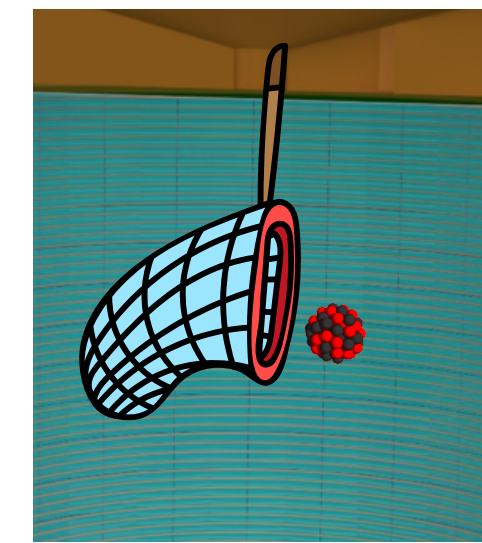
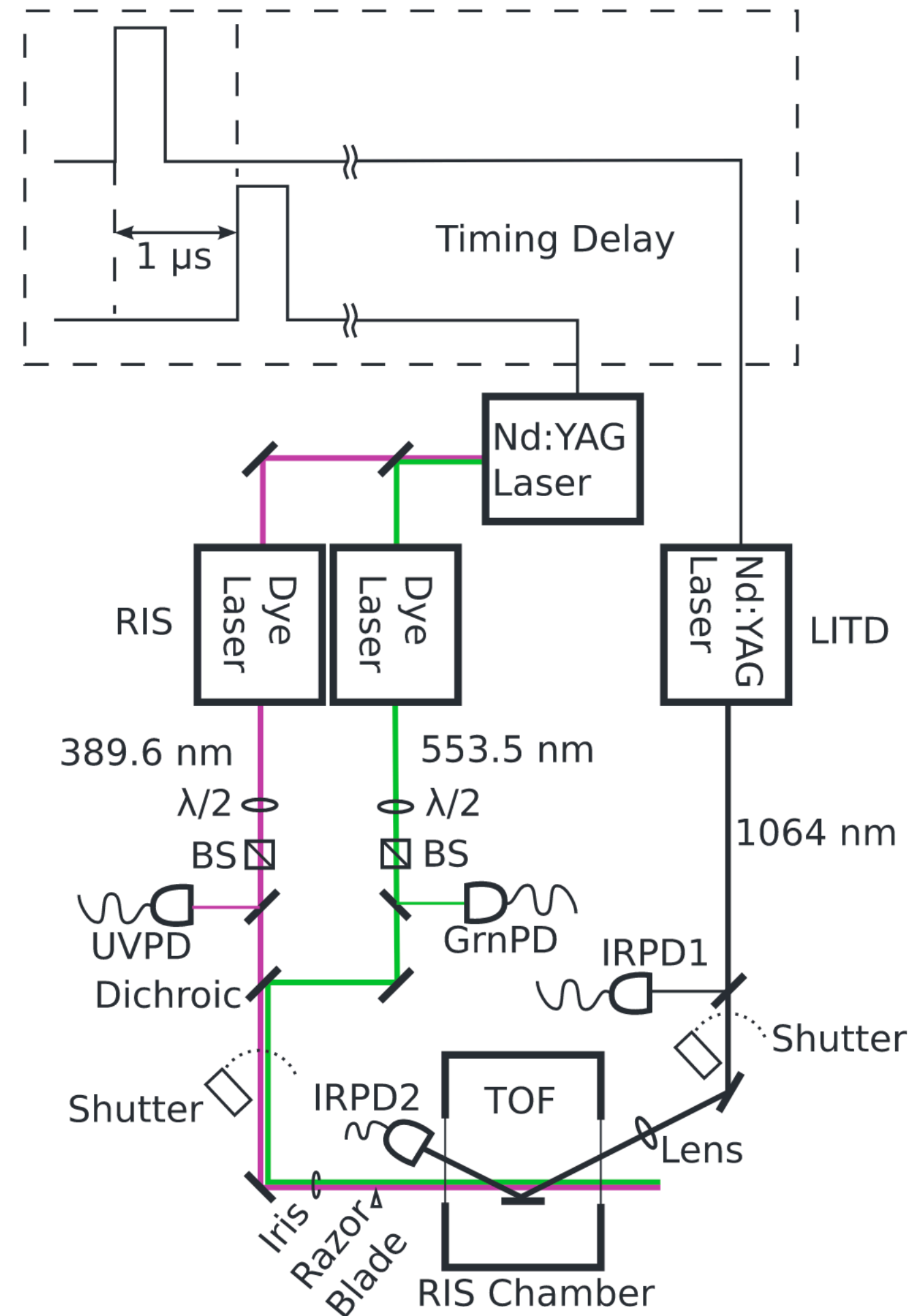
Ask
for more!



Extraction with cryoprobe

The RIS approach

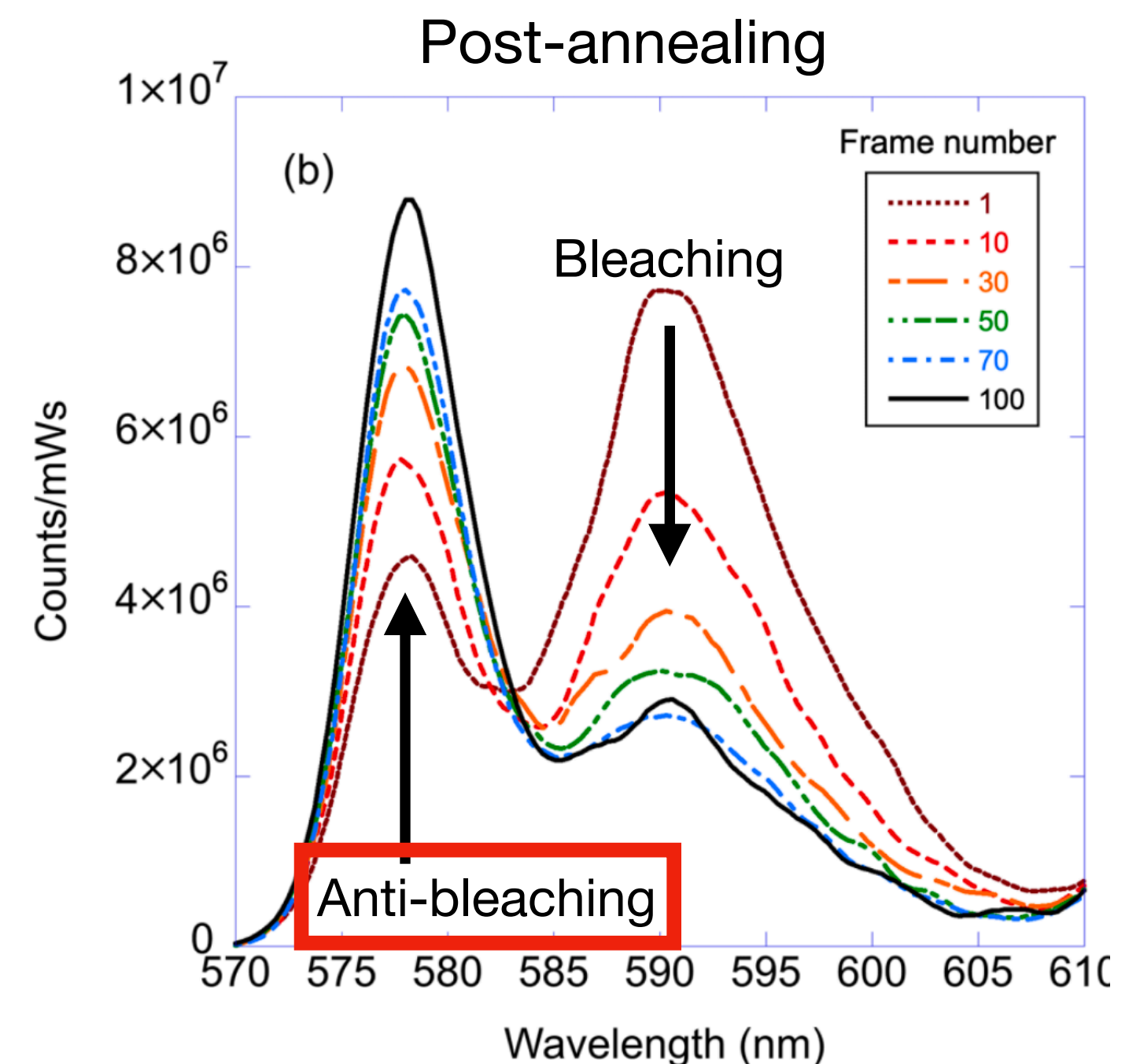
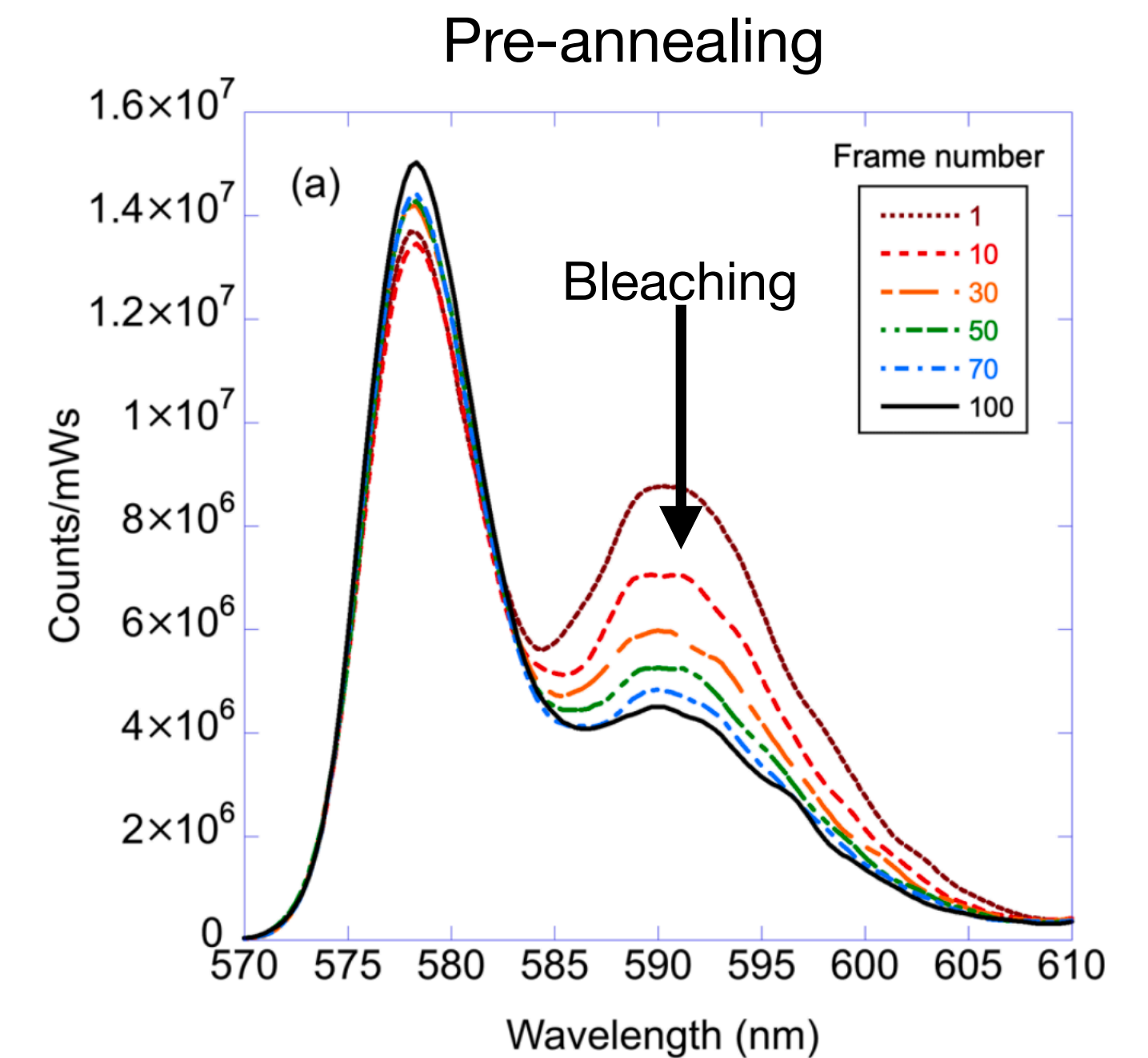
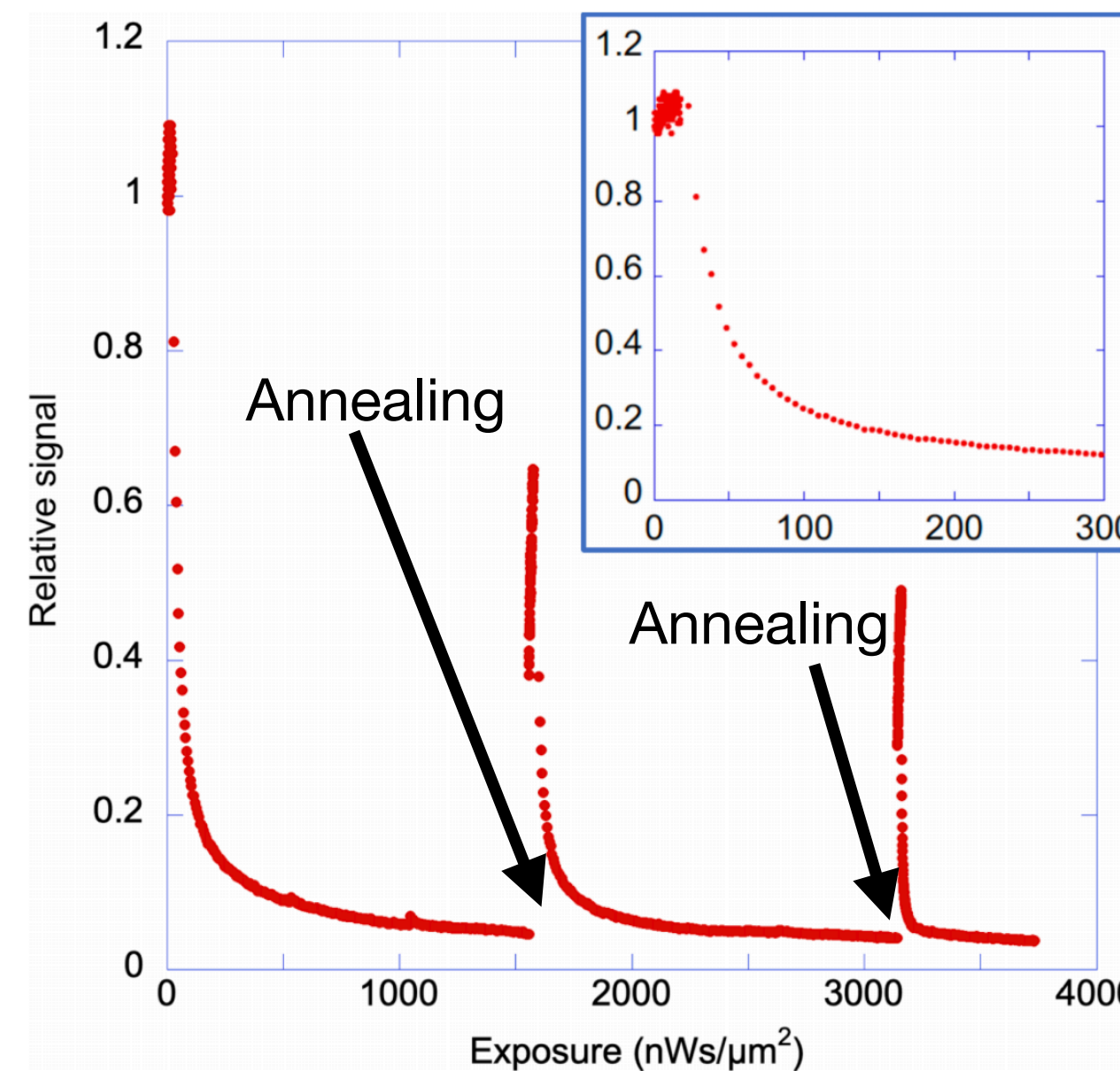
- Demonstration of LITD+RIS tech (Selective ionization of Ba^+).
- Discrimination of direct desorption and non-resonant ionization Ba^+ from background.
- Measure different desorption thresholds from Si for K^+ and Ba^+ .



Single-ion detection in SXe

Overcoming photobleaching

- Extended result of SV Ba in SXe matrix for Tetra- (TV) and Hexa-Vacancy (HV) sites.
- Annealing causes fluorescence recovery (anti-bleaching) at the HV site (577 nm)
- Bleaching is irreversible in the TV site (590 nm).



System calibration

Laser Ablation Source (LAS) + Quadrupole bender

- Custom ablation target:
Ti, Mb, Ni, Ta, W, CuSn, SS...
- Used to calibrate MRTOF and
LPT with other elements

