Progress in tagging of ^{136}Ba daughter from ^{136}Xe $0\nu\beta\beta$ decay

Bill Fairbank

Contributions from the NEXT and nEXO Collaborations

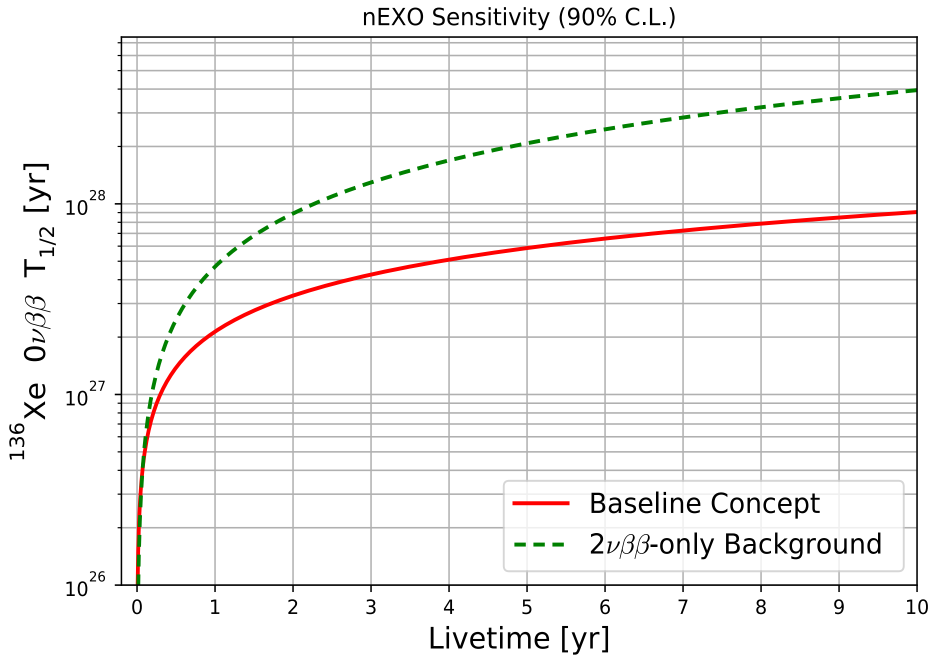
Tagging the Ba daughter of Neutrinoless Double Beta Decay could increase the ultimate sensitivity of nEXO and NEXT

$$^{136}Xe \rightarrow {}^{136}Ba^{++} + 2e^{-}$$

"Tagging" Ba daughter has potential to eliminate all but 2νββ backgrounds

M. Moe, Phys. Rev. C 44, R931 (1991)

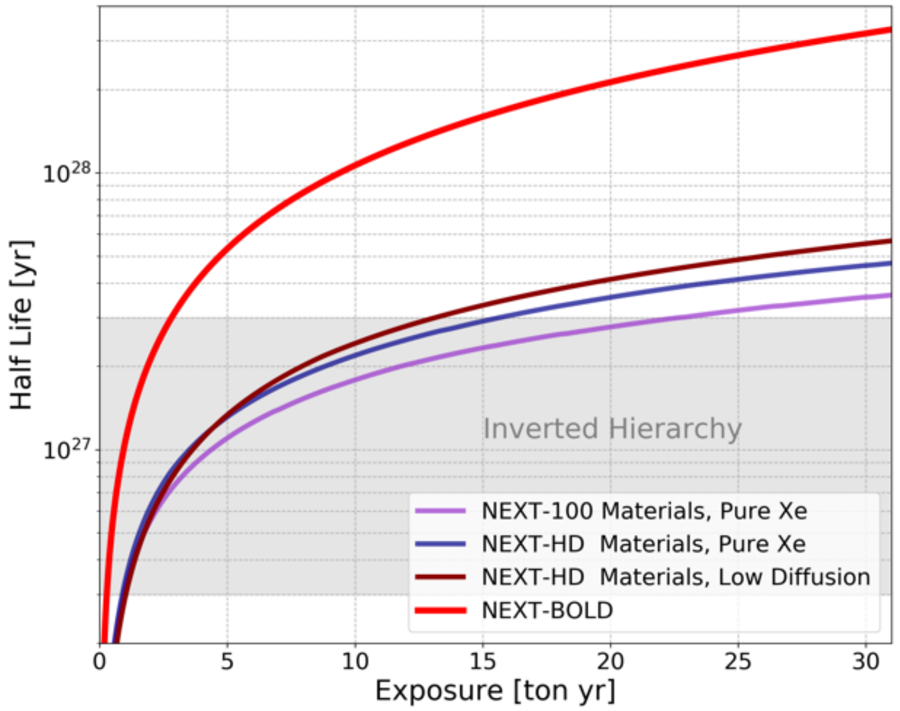
In nEXO, eliminating other backgrounds could give up to 4x higher sensitivity



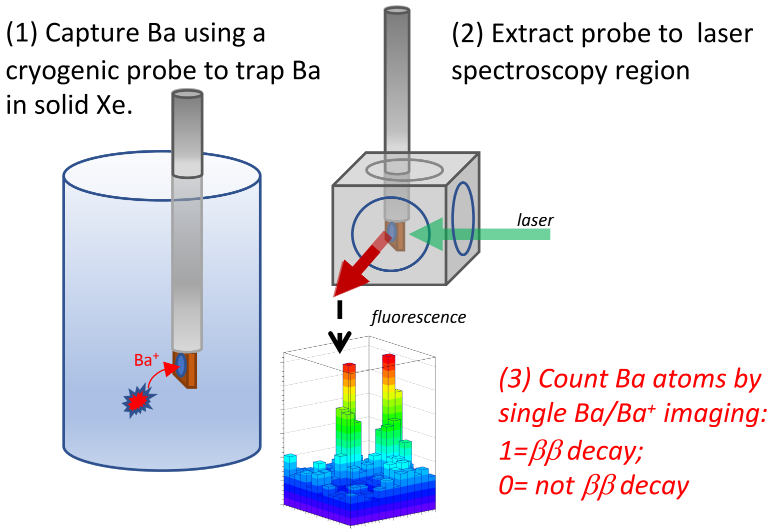
nEXO Collaboration, *Phys. Rev. C* **97**, 065503 (2018)

Ba tagging is a possible upgrade after 5 yrs of nEXO operation

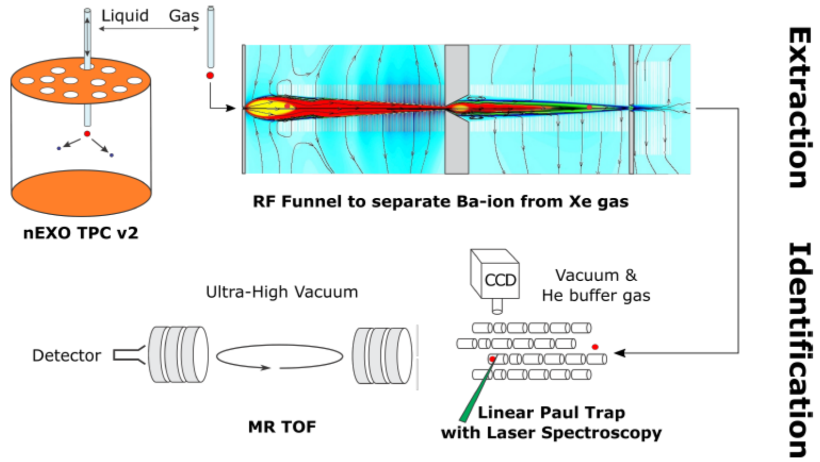
In NEXT, higher efficiency with Ba tagging and eliminating other backgrounds could provide up to a factor of 6 higher sensitivity



nEXO Barium tagging in SXe



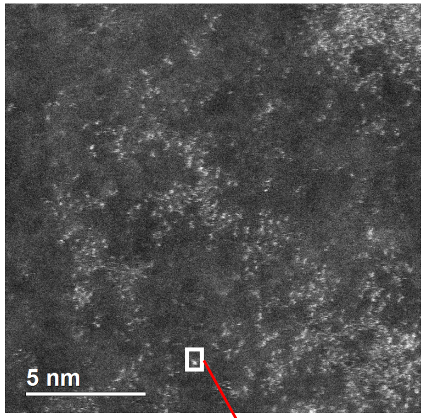
nEXO Barium tagging using capillary, funnel, trap, TOF



- Extract $Ba^{+(+)}$ from liquid Xe TPC into a Xe gas environment
- Extract $Ba^{+(+)}$ with a Xe gas jet into a low pressure chamber
- After nozzle, pump Xe gas away and guide $Ba^{+(+)}$ to identification

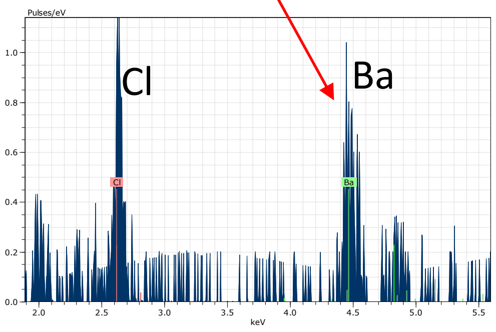
nEXO Barium tagging using single Ba imaging with electrons and x-rays

ADF Image

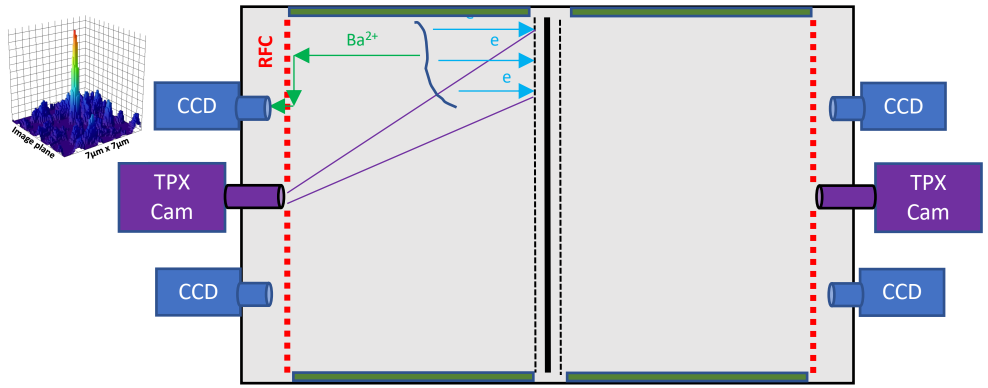


Single "molecules" of $BaCl_2$

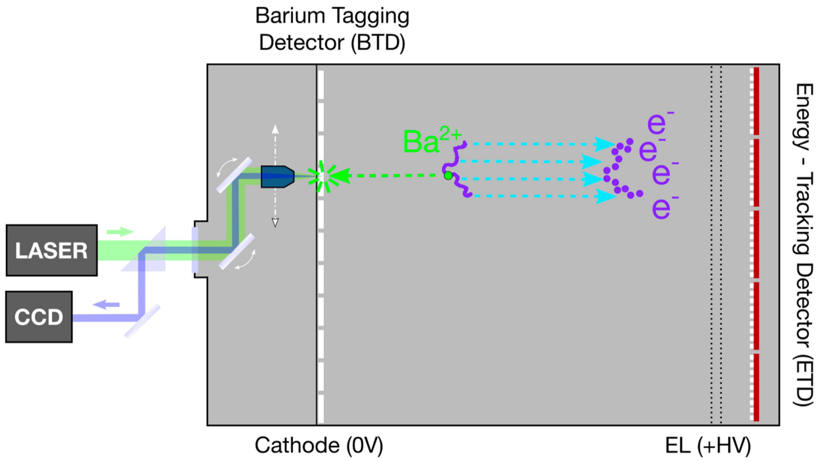
EDX Spectra of isolated molecule of $BaCl_2$



NEXT "CRAB" concept with RF carpet concentrators and camera-based topology measurement



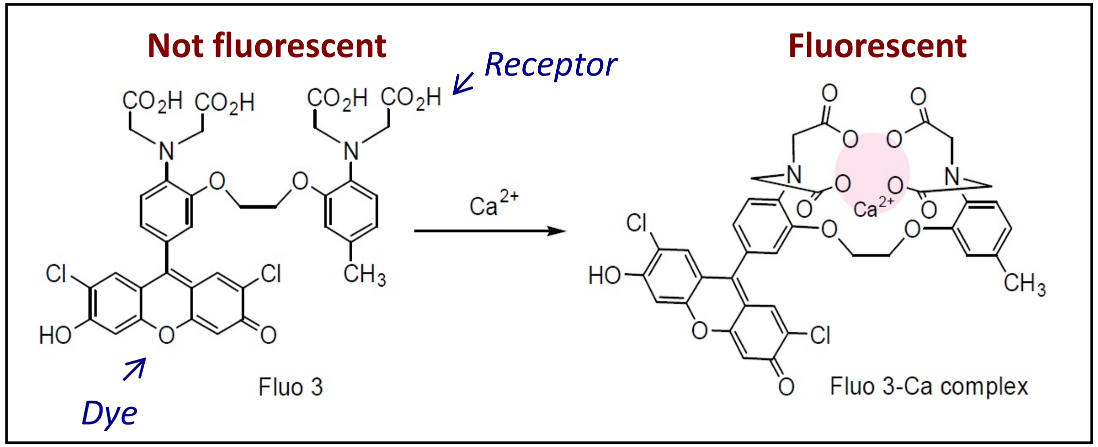
NEXT "Barium Tagging Detector" concept with fully active cathode and SiPM—based tracking + energy plane

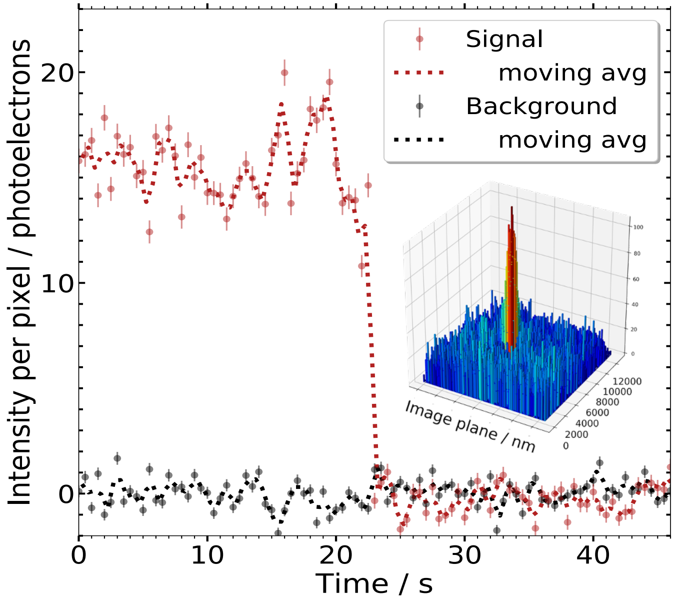


Barium Tagging Progress in NEXT

Barium Tagging in Xenon Gas with New Single-Molecule Fluorescence Imaging Techniques

- In xenon gas, barium daughter ions are expected to become ~100% doubly- charged – but there are no low-lying fluorescent transitions.
- Chelation of Ba^{++} by fluorescent chemosensors generates transitions in the visible spectrum under near-UV or two-photon IR excitation.
- New, highly selective chemistry has been realized for Ba^{++} ions.
- Single-step photo-bleaching demonstrates single-molecule sensitivity, with ±2 nm rms super-resolution — well beyond the diffraction limit.





Single-step photo-bleaching in 7 bars of argon gas.

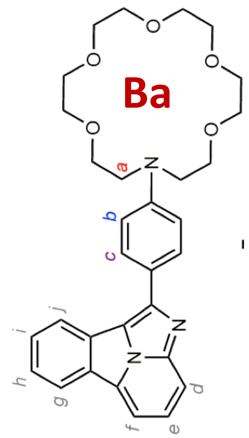
J.Phys.Conf.Ser. 650 (2015) 1, 012002
JINST 11 (2016) 12, P12011
Phys.Rev.Lett. 120 (2018) 13, 132504

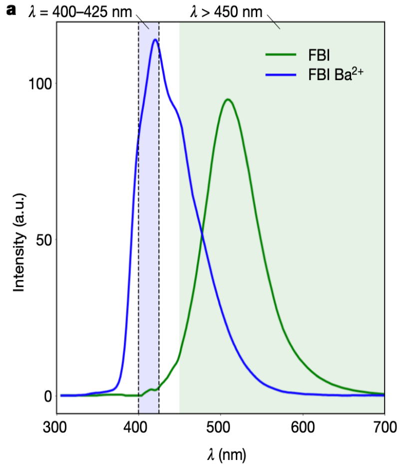
New Chemistry

- Conventional ion chemosensors are not suitable for imaging in dry environments such as xenon gas
- NEXT has developed new, dry-phase imaging of barium ions using crown-ether derivatives linked to optimized fluorophores.
- Crown-ether receptor can be tuned to bind efficiently and selectively to barium cations.
- Thus far, two types have been developed within this class of crown-ether chemosensors: **on-off**, and **bi-color.**
- Computational chemistry is predictive for molecular fluorescence and binding.

Nature Sci Rep 9, 15097 (2019)
Nature 583, 48–54 (2020)
arXiv: 2006.09494 (submitted to JACS)

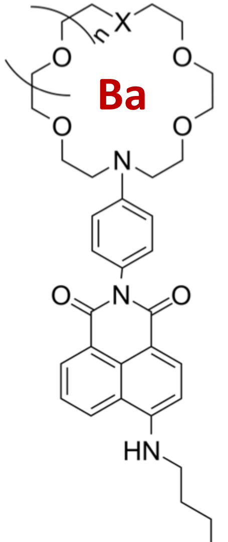
In-vacuo capture from $Ba(ClO_4)_2$ with bi-color response



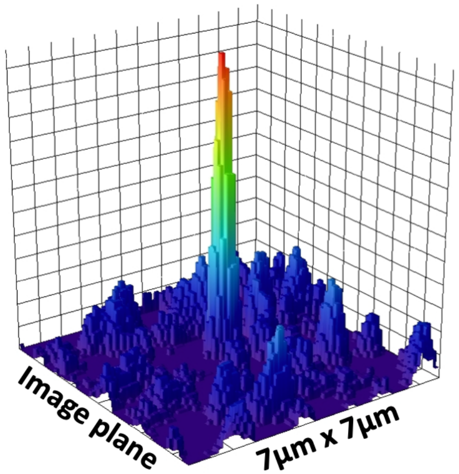


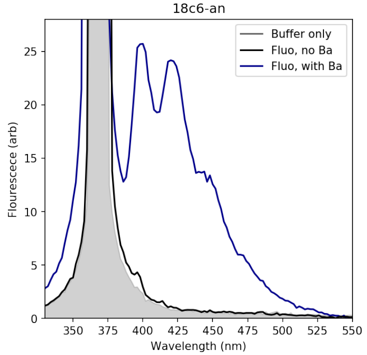
Fluorescent Bi-color chemosensor switches from green to blue upon chelation; filter removes green light background allowing clean separation

Dry single Ba^{++} ion detection with on-off fluorescence



1A-1E





First evidence for $Ba(ClO_4)_2$ reaction in dry medium using the bi-color chemosensor

Nature 583, 48–54 (2020)

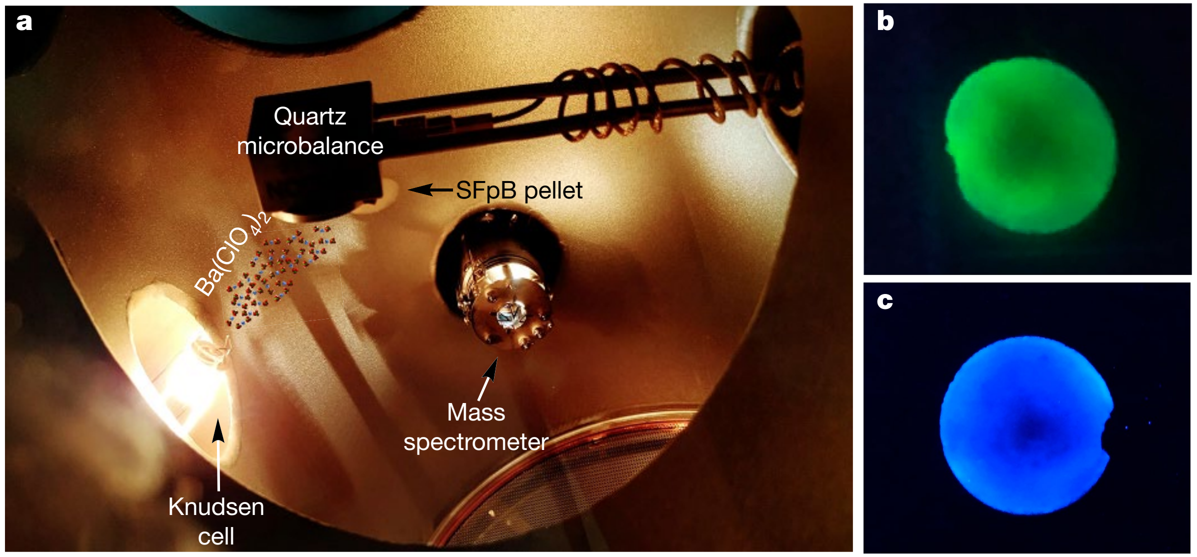
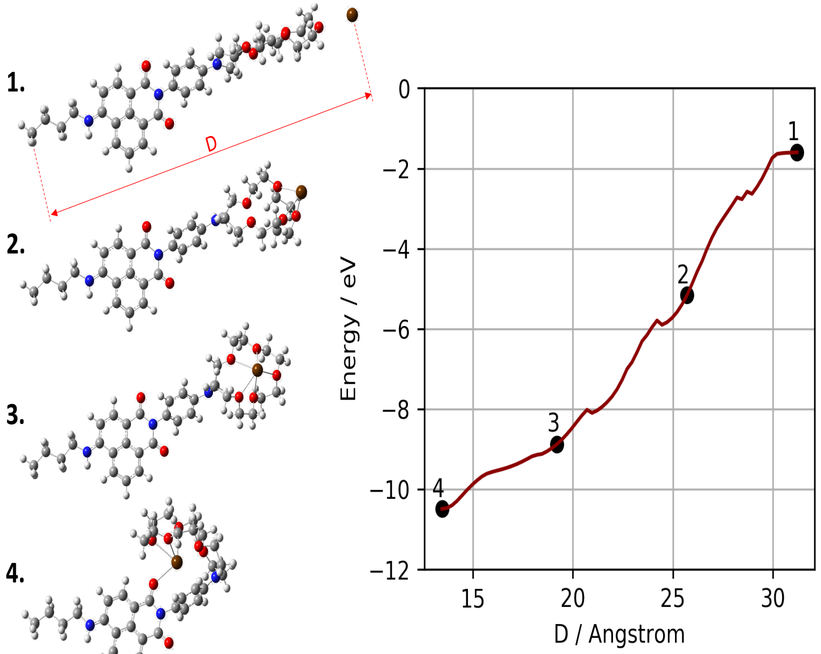


Fig. 3 | Sublimation of $Ba(ClO_4)_2$ on the FBI. a, Experimental setup. Photograph of the interior of the UHV chamber used for sublimation. The positions of the pellet, evaporator, quartz microbalance and mass spectrometer are indicated. **b**, **c**, Photographs of the pellet before (**b**) and after (**c**) the sublimation. In both cases, the excitation light is 365 nm. We note the characteristic green colour of unchelated FBI before the sublimation and the blue shift after the sublimation, which shows a large density of chelated molecules.

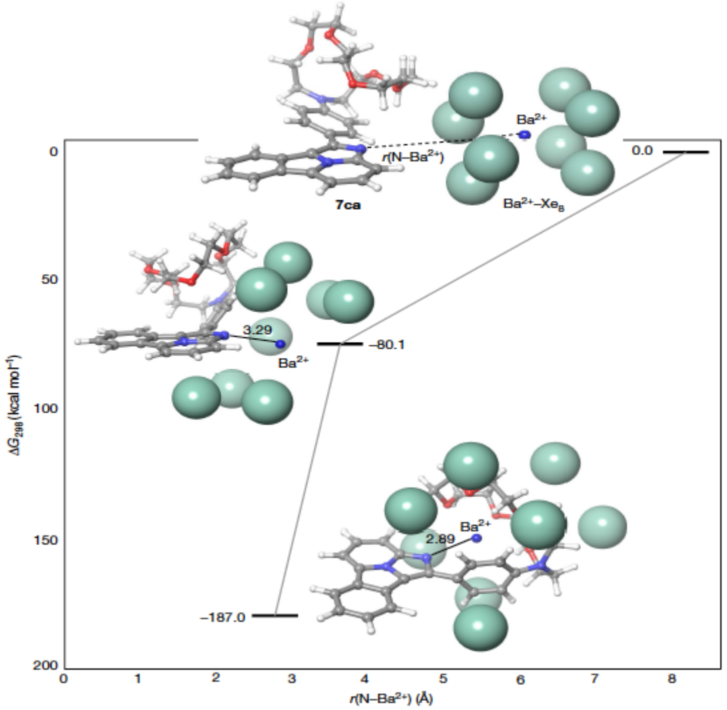
This result indicates likely chelation with Ba^{++} in dry xenon gas

Computations indicate that Ba^{++} is expected to chelate efficiently in high-pressure xenon gas



As Ba^{++} moves close to crown-ether, it becomes very strongly bound

arXiv: 2006.09494 (submitted to JACS)



Presence of xenon atoms in solvation shell around Ba^{++} does not inhibit chelation

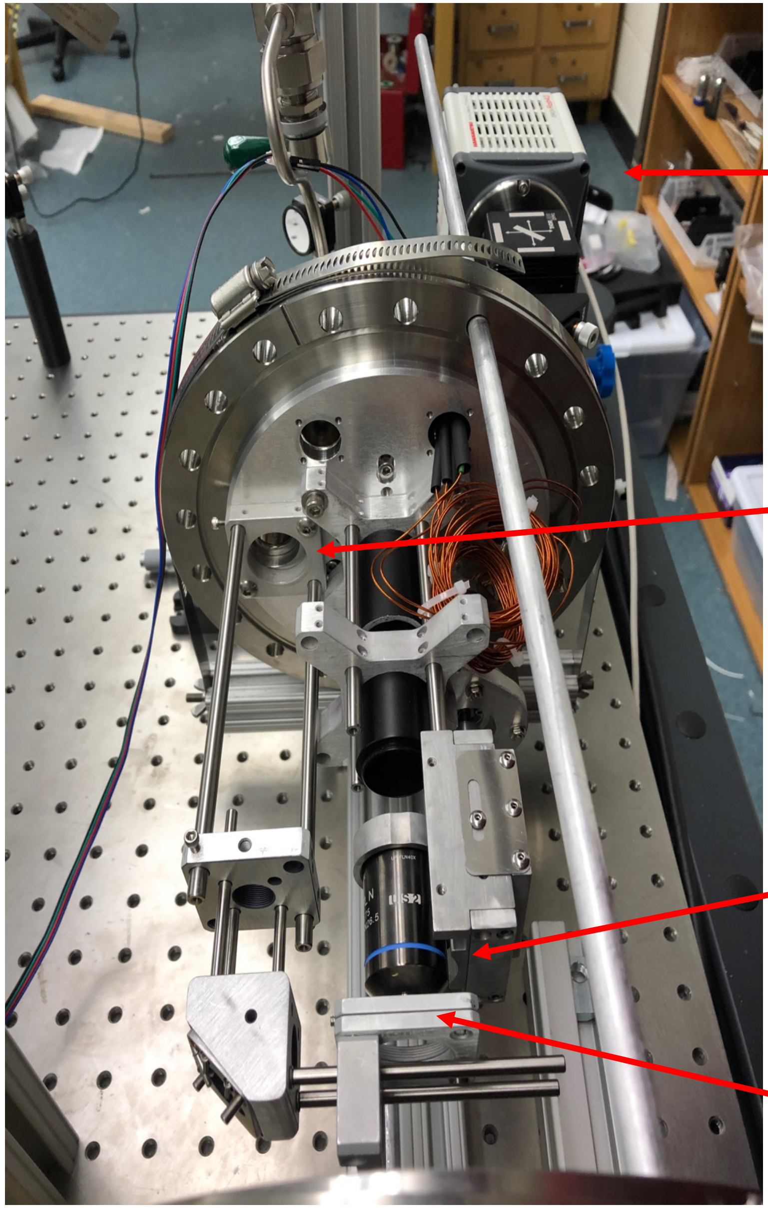
Nature 583, 48–54 (2020)

High-Pressure Microscope Demonstration

- TIRF and epi-fluorescent microscopy have both been recently demonstrated as single-ion capable modalities within high-pressure gaseous xenon (HPGXe) systems.
- Images of single-molecule fluorescence show no dependence on pressure from 1 to 10 bars of xenon or argon gas
- Sample exploits chelated Ba^{++} immobilized within PVA matrix

DPF2019 proc, arXiv: 1909.04677
Publication in Preparation

Single ion imaging demonstrated across 1mm^2 in HPGXe



EMCCD Camera

Laser beam enters here

Objective

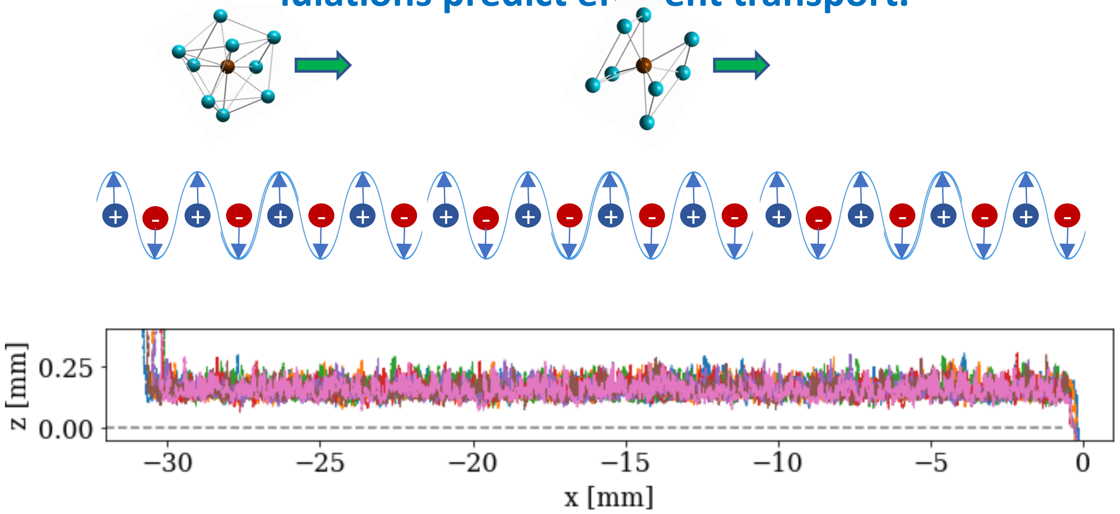
Sample with Ba^{++}

Ion Transport & Concentration: RF Carpets

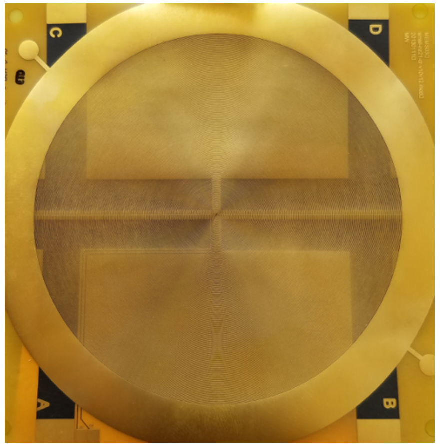
- One concept for ion collection from a large cathode plane is through concentration with RF carpets at 10 bar onto ~1mm^2 sensor plane with a monolayer of chemosensors.
- Simulations and HV tests suggest that efficient ion transport to sensor plane is achievable at 10 bar xenon, even with solvation shell.
- Program of R&D at the CARIBU facility will test high pressure RF carpet, scheduled for 2020 (COVID permitting).

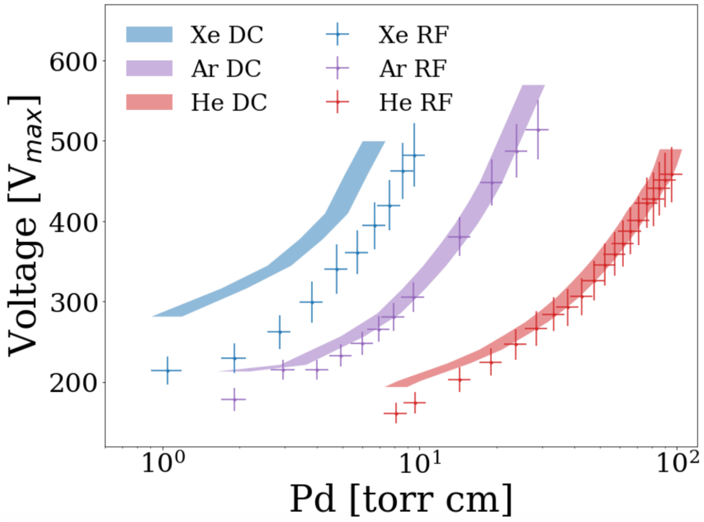
JINST 15 (2020) 04, P04022
Phys.Rev.A 97 (2018) 6, 062509

DFT calculations predict ion clustering and RF carpet ulations predict eff[illegible]ient transport.



RF HV strength of Xe is sufficient for RF transport at 10bar.





"NEXT" Steps

- Ion beams using $^{222}Ra^{2+}$ from thorium decay are under development for single-ion tests.
  (RITA program, Spain)

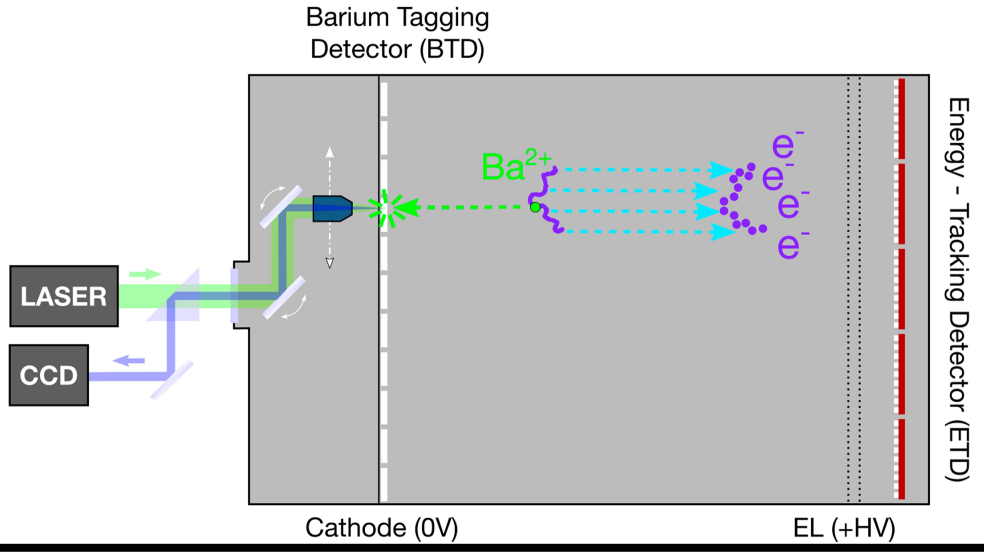
- Beam tests planned at ANL CARIBU will use $^{144}Ba^{2+}$ mass-selected from ^{252}Cf fission.
  (GodXilla program, USA)

- The ultimate test-beam is ββ2ν !

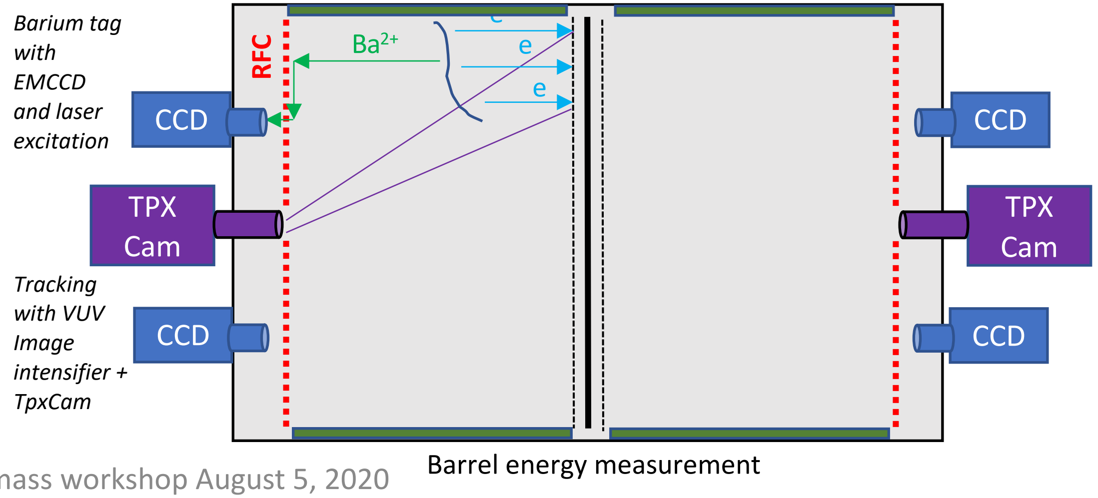
- Demonstrator phases at 10kg-scale are being planned for ~2024-2025.

- Multiple full system concepts under exploration, to be guided by ongoing R&D.

"Barium Tagging Detector" concept with fully active cathode and SiPM—based tracking + energy plane

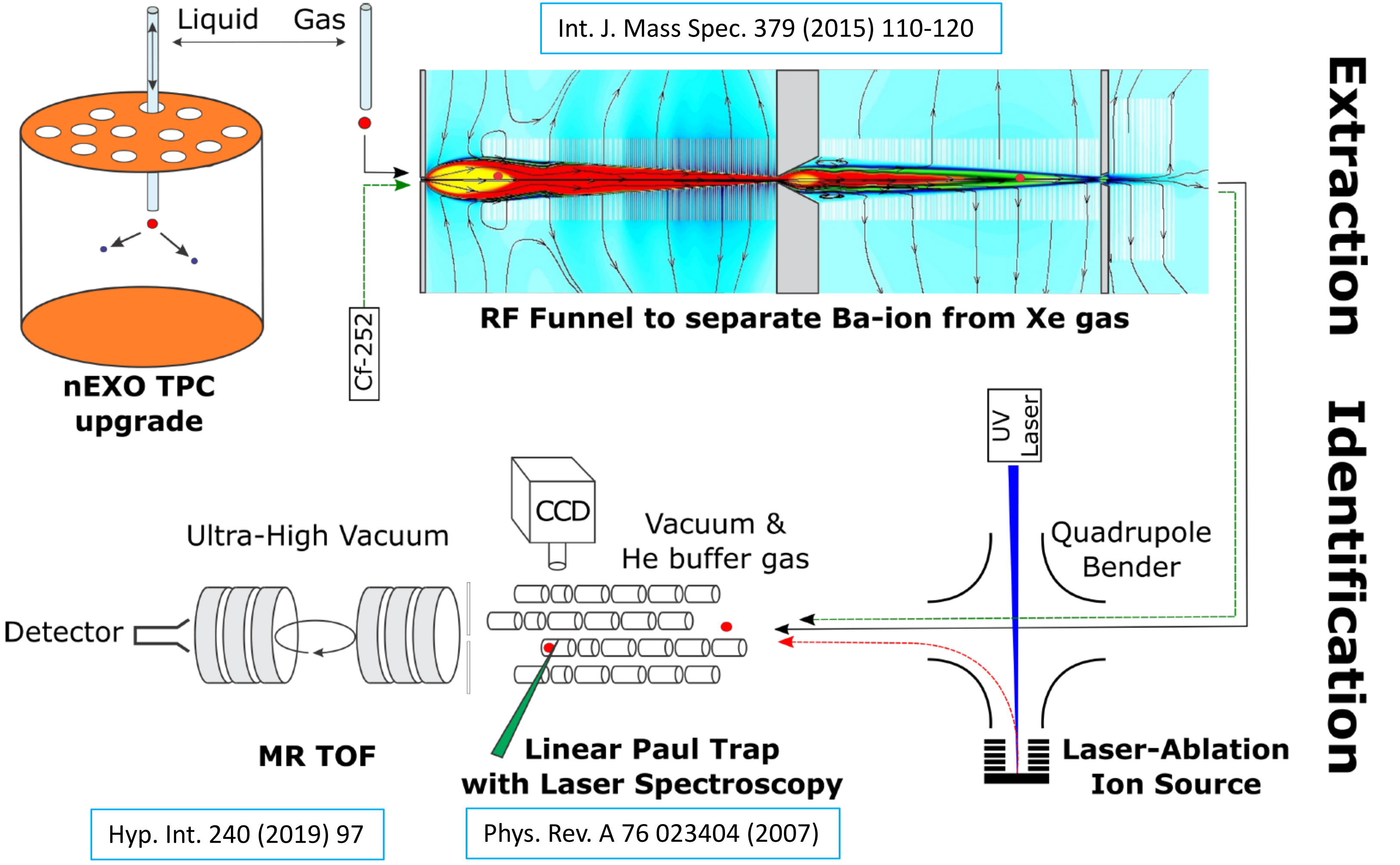


"CRAB" concept with RF carpet concentrators and camera-based topology measurement



Barium Tagging Progress in nEXO

Ba-ion tagging – the Canadian approach



- Extract $Ba^{+(+)}$ from liquid Xe TPC into a Xe gas environment
- Extract $Ba^{+(+)}$ with a Xe gas jet into a low pressure chamber
- After nozzle, pump Xe gas away and guide $Ba^{+(+)}$ to identification

Trans-Canadian Effort

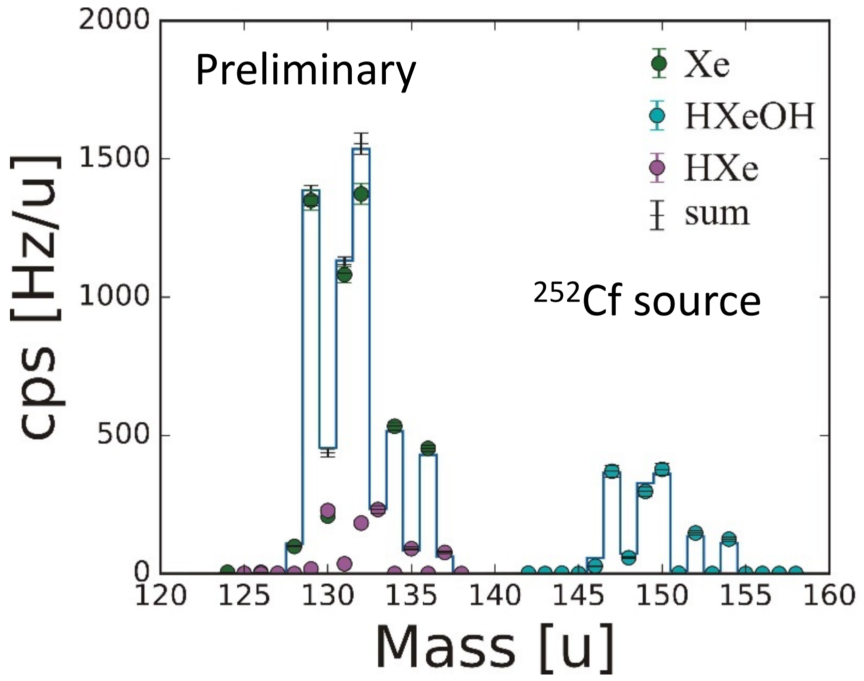
Carleton University:
- Laser system for ion identification (ID)
- Displacement device for ion extraction from LXe to GXe

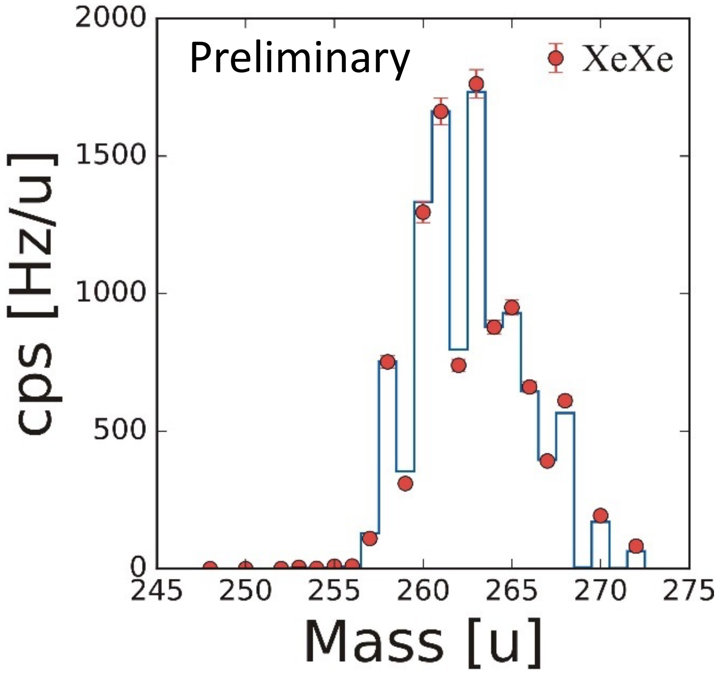
TRIUMF:
- Linear Paul Trap (LPT) for ion ID and bunching
- Online-extraction studies with radioactive ions

McGill University:
- Radio-Frequency Funnel for ion extraction in vacuum
- Multi-reflection time-of-flight mass spectrometer (MR TOF) for m/q ID

Ion extraction from xenon gas with RF funnel

Mass spectra of ions extracted from 2.1 bar Xe using Cf-252 source and single RF funnel

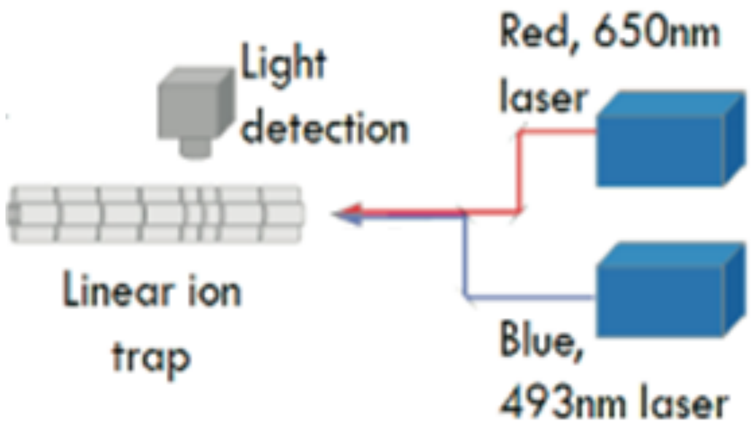




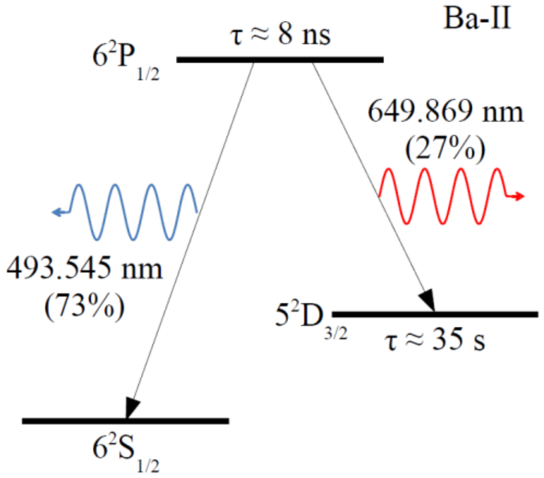
- Ions extracted up to 10 bar!
- Gd-148 and Cf-252 ion sources used
- Ions extracted from Ar, Kr, and Xe

- Ba-ions not identified
- Fission products not identified
- Ion extraction efficiency unknown!

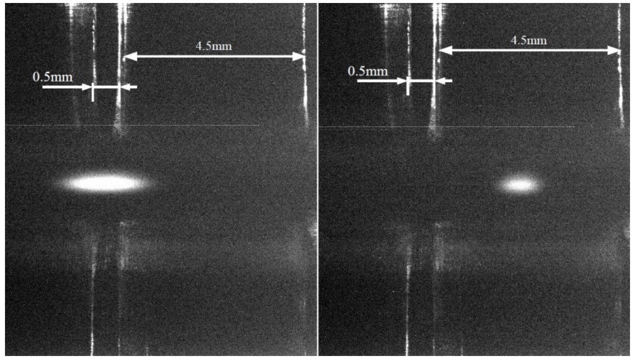
Single Ba ion detection & identification



Using a relatively simple and well understood fluorescing system

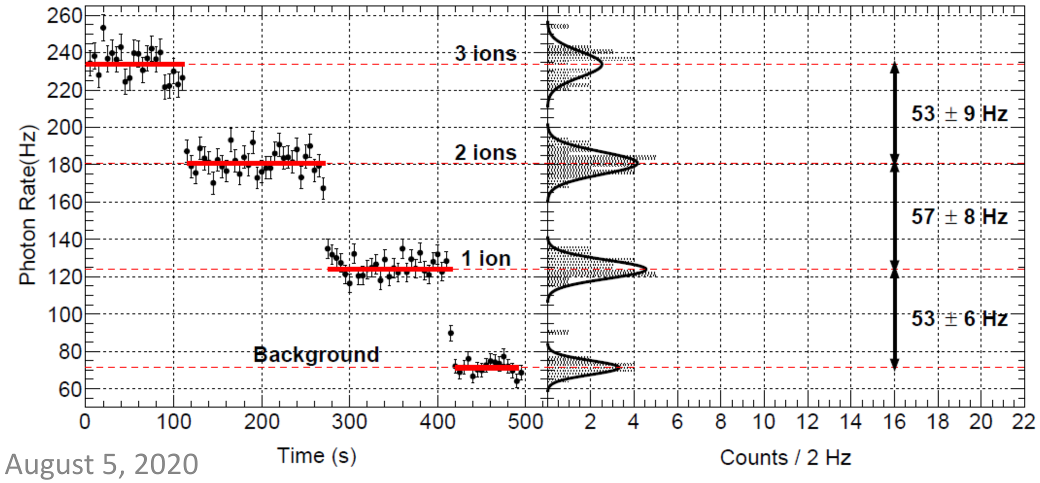


Demonstrated ion cloud imaging and accurate position control



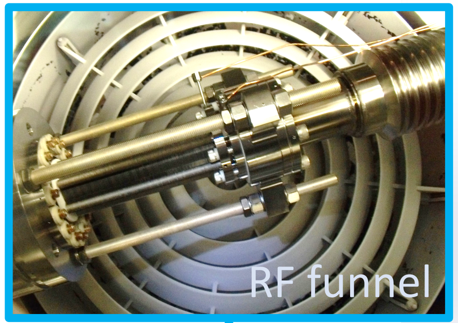
Demonstrated by M. Green et al., Phys. Rev. A **76** 023404 (2007)

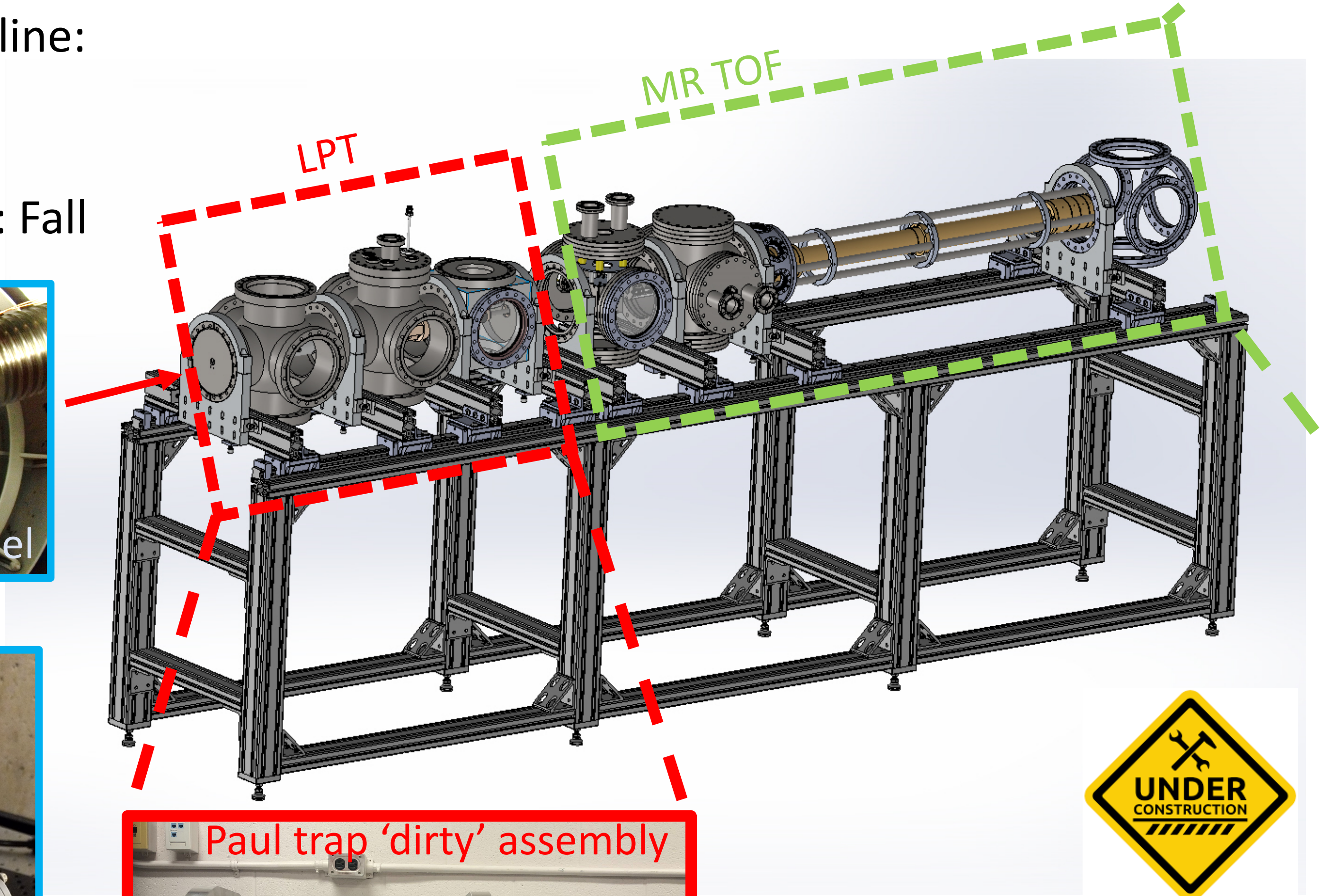
Demonstrated single ion sensitivity using intermodulation technique (background control)

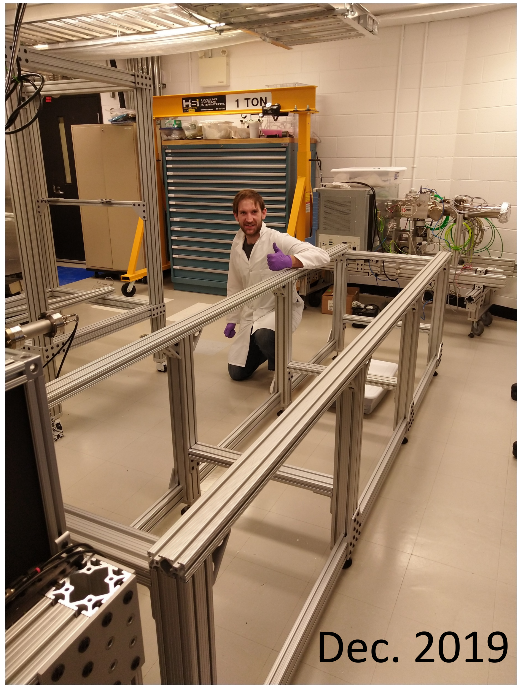


Ba-tagging in Canada

- Part assembly resumed early July!
- Commissioning Timeline:
  - MR TOF: Aug.
  - Paul trap: Sept.
  - LTP and MR TOF: Fall

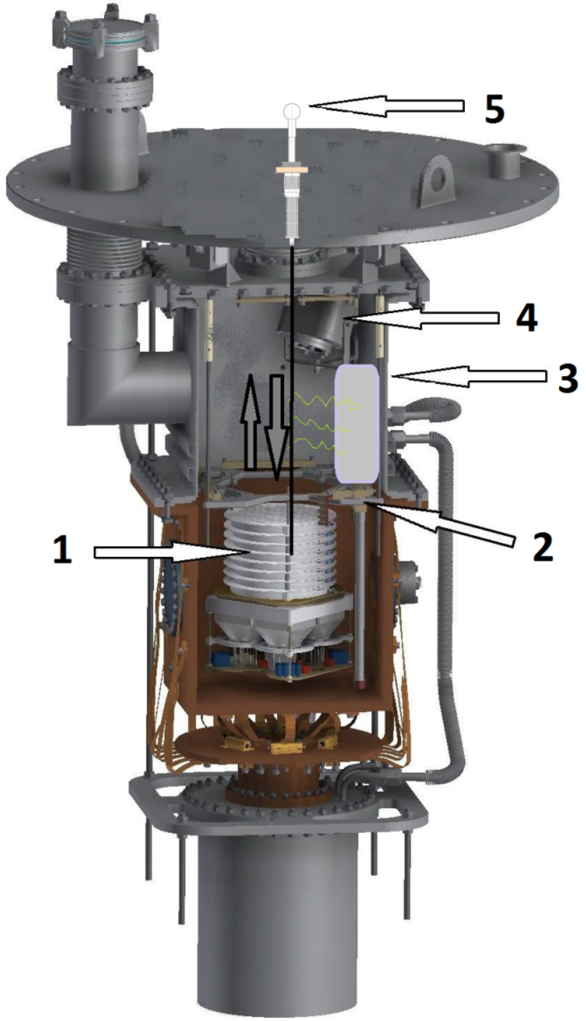






- Ion extraction from GXe with upgraded RF funnel & Cf-252 anticipated 2021
- Ion extraction from LXe at TRIUMF anticipated 2023

Tests of ion extraction from LXe



- **Cryogenic test setup**
  - 1) Test device (electric drift field plus scintillation light detector, no tracking)
  - 2) Slow control instrumentation (LXe level control, heaters)
  - 3) Scintillation light detector in gaseous Xe (mainly for alpha spectrometry)
  - 4) Cryogenic camera (for control of mechanical motion)
  - 5) Mechanical manipulator and feedthrough
- (Radioactive) ion injection done through the gas purification/recirculation system (not shown in the drawing beside)
- **Two strategies are considered**
  - **Mechanical extraction using a dry probe carrying HV which dips in and out of LXe**
    - Easy to implement for Po-218 ions using Rn-222 injection
    - Alpha decay of Po-218 ions leads to Xe gas ionization and then the production of a large amount of scintillation light
    - Good energy resolution and particle identification required to be able to reject backgrounds (electron recoils and alpha decays from U/Th chains)
  - **Mechanical manipulator only drives a capillary tube made of stainless steel to allow for the ion extraction from LXe (along with large amounts of Xe) into a low pressure chamber for further ion manipulation and analysis (not shown here)**

Status and Plan

- Ion extraction from Xe gas up to 10 bar demonstrated
- Single Ba^{+}-ion identification demonstrated with excellent signal to noise
- Ion identification under development for sensitive mass and fluorescence spectroscopy of extracted ions
- R&D required to develop extraction of ion from LXe to GXe
- Developing plan to study ion-extraction using radioactive ions at accelerator facility

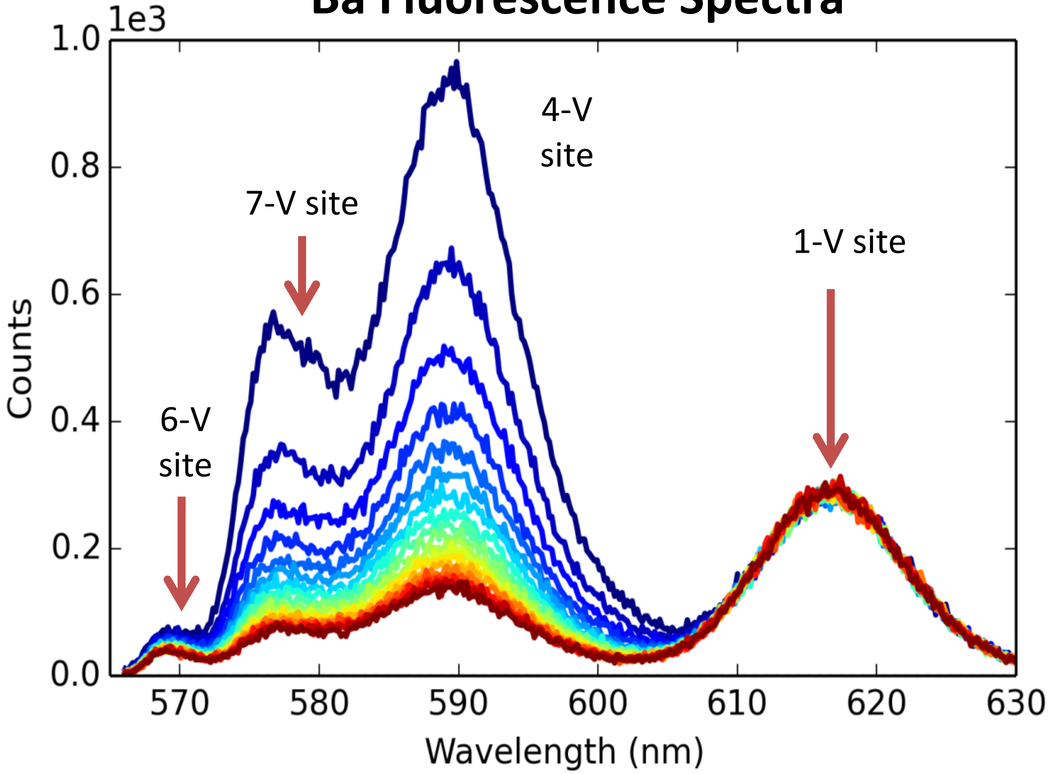
Ba tagging in SXe: Ba exists in SXe in 4 matrix sites with unique spectra

because Ba is bigger than Xe

Each Ba site has a different fluorescence and excitation spectrum.

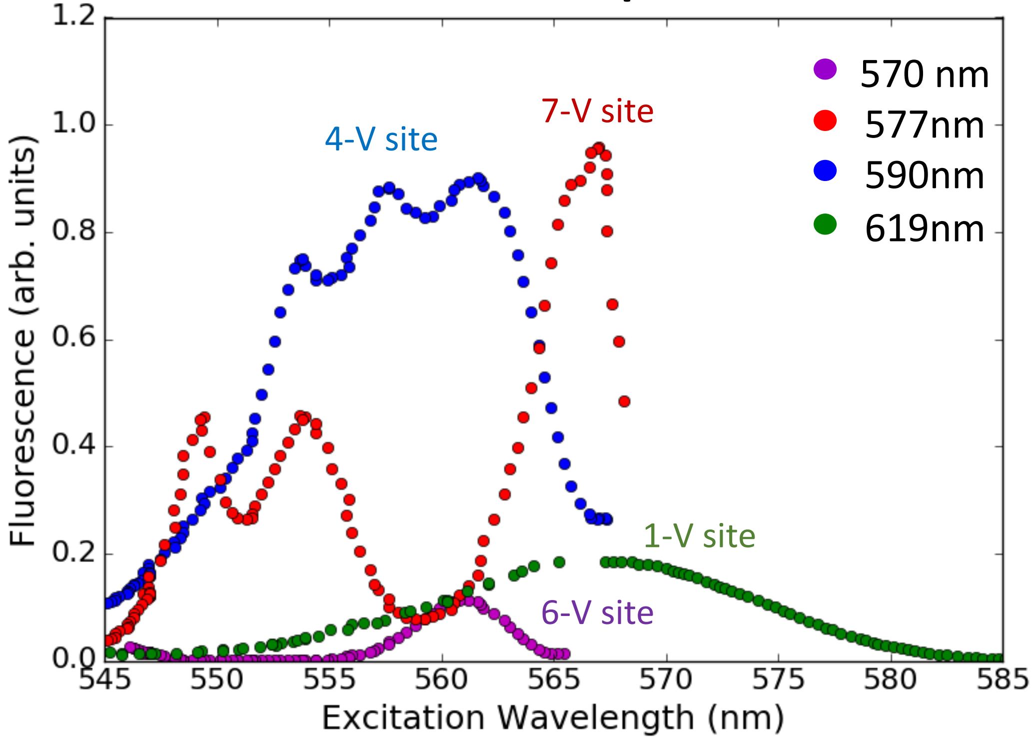
Site identification determined by theoretical calculations (Kleshchina et al, J. Chem. Phys. 151,121104 (2019))

Ba Fluorescence Spectra



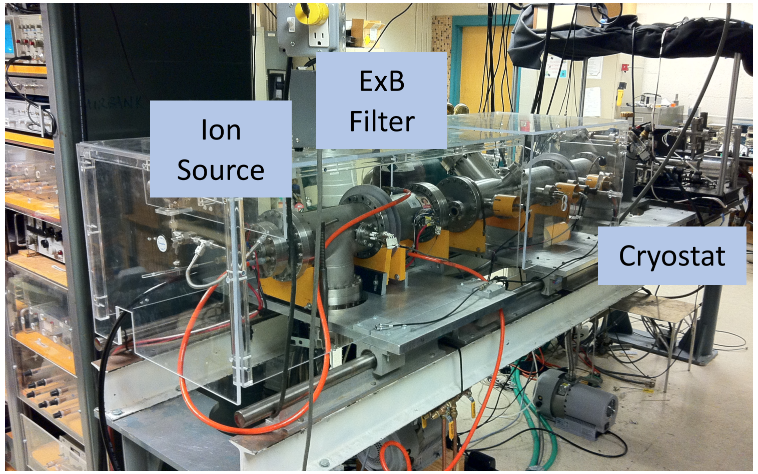
Every 10th spectrum shown – 3 peaks exhibit bleaching

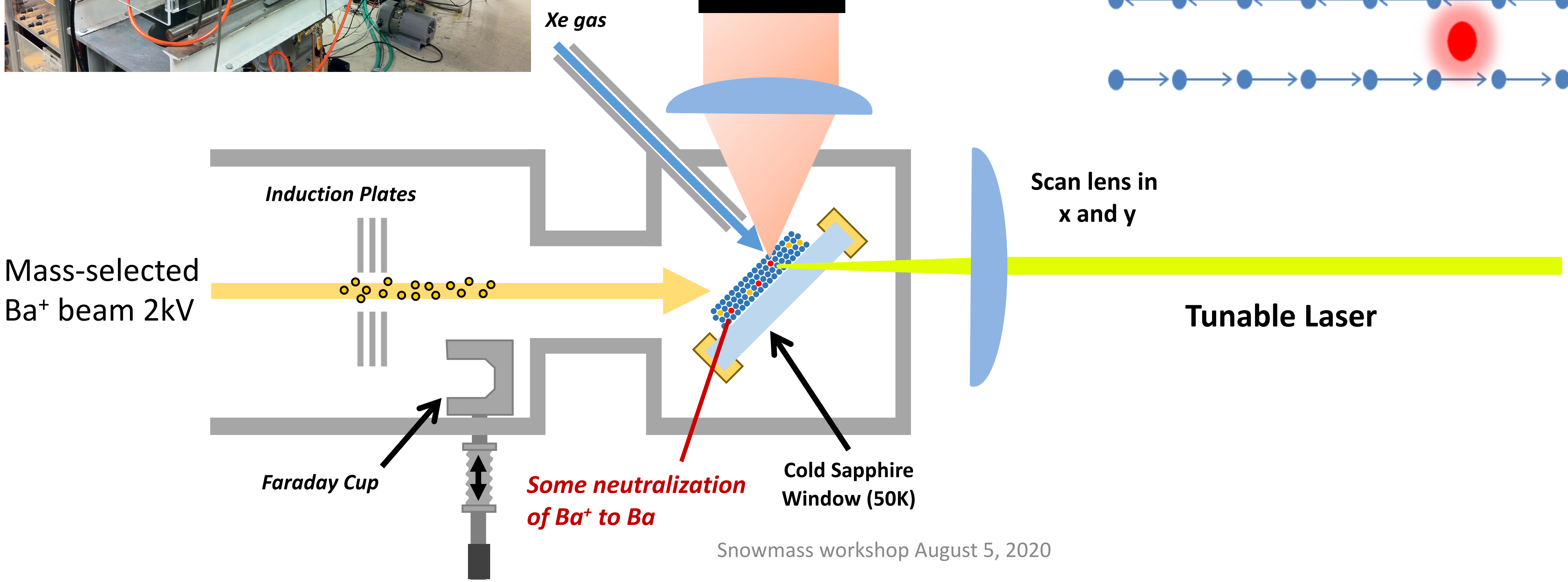
Ba Excitation Spectra



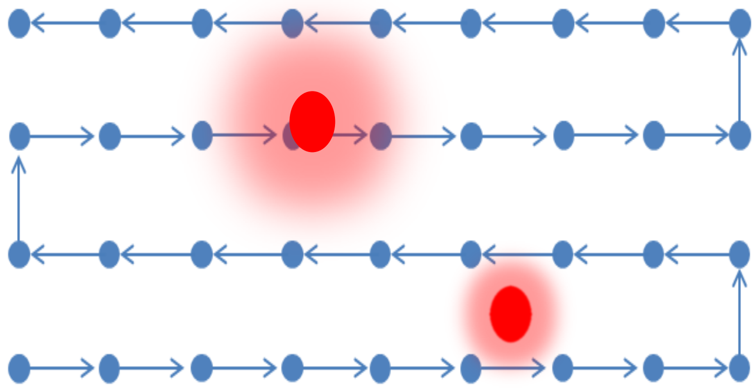
B. Mong et. al, *Phys. Rev. A* **91**, 022505 (2015)

Imaging of single Ba/Ba$^+$: Deposit pulses of few Ba$^+$ ions in SXe





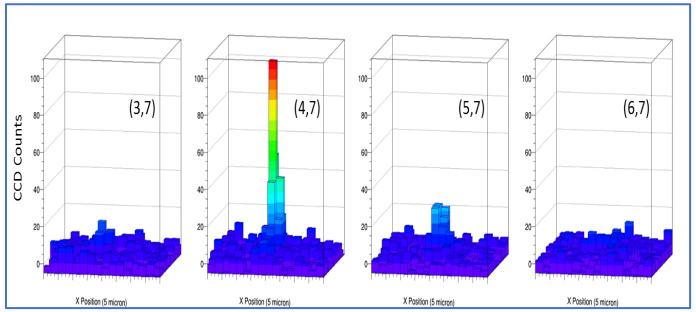
Scan laser across deposit



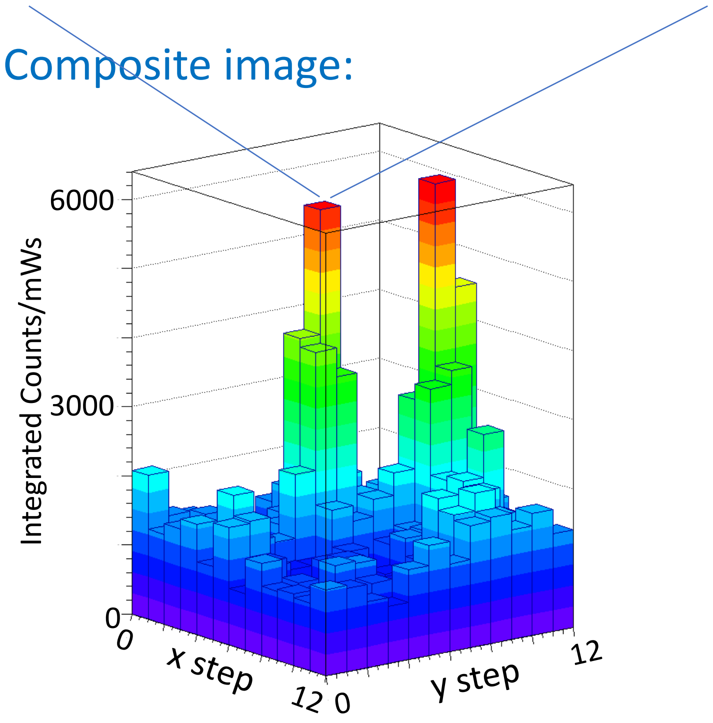
Scanning and counting single Ba atoms in SXe:

in 1-V site

raw CCD images:

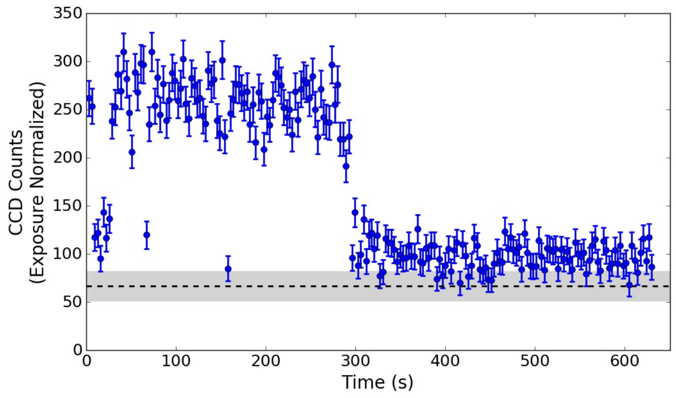


Composite image:

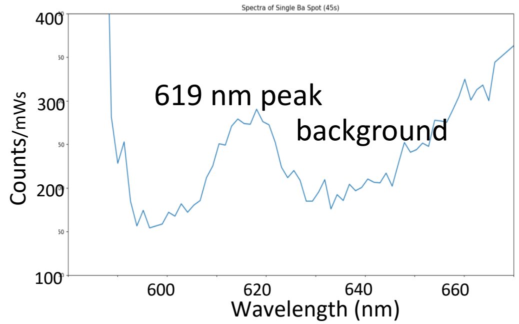


Nature **569,** 203 (2019)

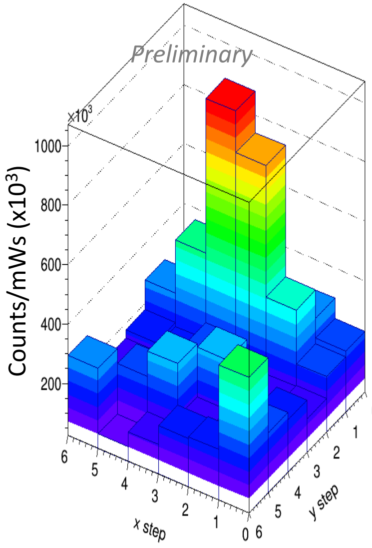
Looking at one Ba atom: sudden turn off, blinking, spectrum



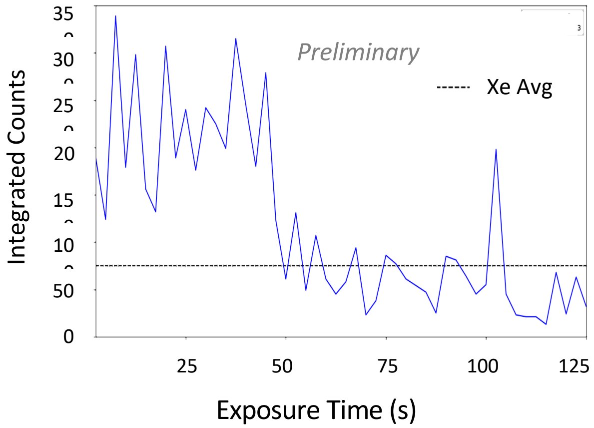
Spectrum of a single Ba atom



in 7-V site



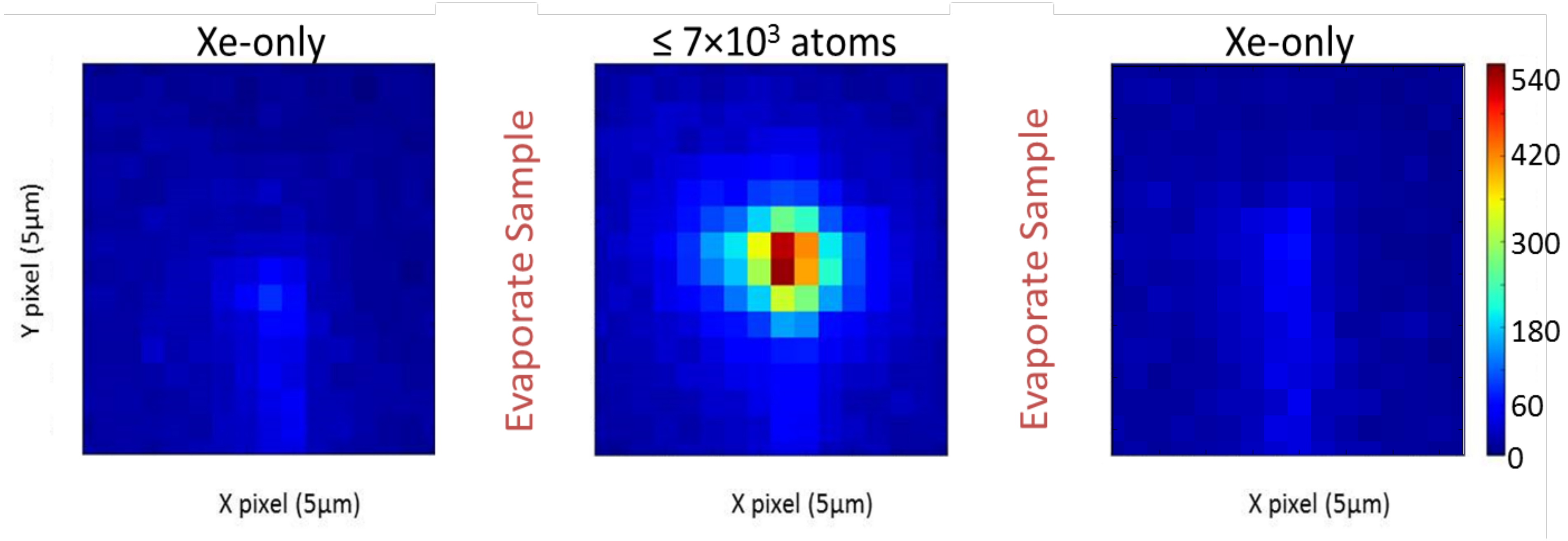
Sit on a peak: see characteristic turnoff of a single atom



We are working on imaging of Single Ba atoms in the other two sites.

An important feature of Ba tagging in SXe:

Even in a large Ba deposit of ~7000 Ba^+ ions, evaporation "erases" the Ba signal (619 nm 1-V site):



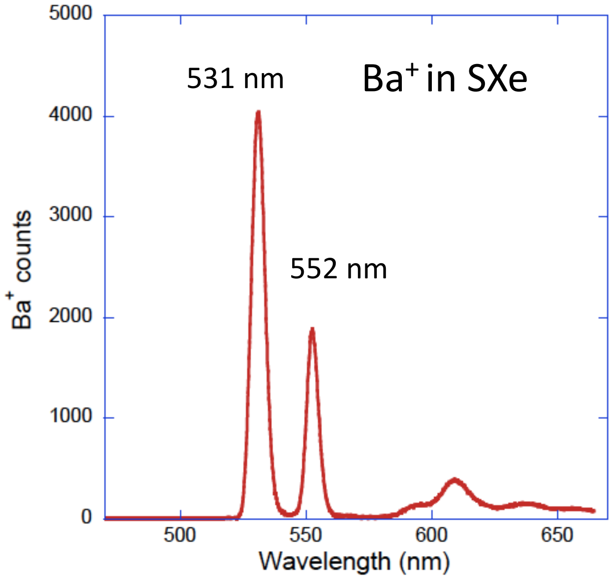
AND: If there are other Ba atoms on the window, we don't see them.

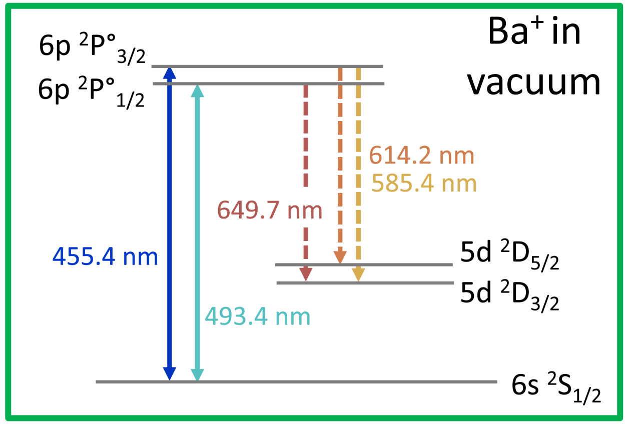
As far as we have observed, this is true in other Ba sites, as well as for Ba^+.

Imaging of small numbers of Ba^+ ions in SXe with spectrometer

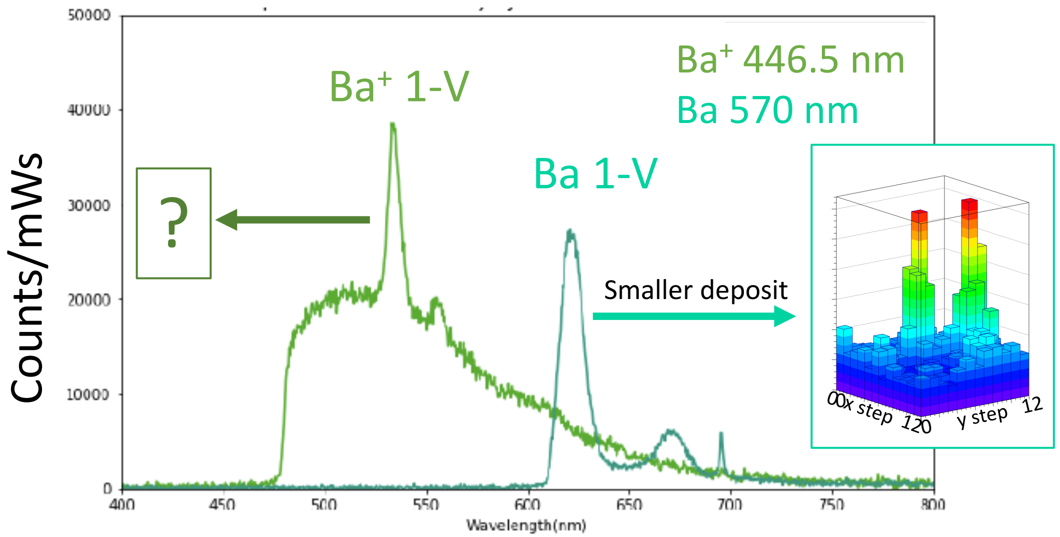
Ba^+ is smaller than Xe: ***Prefers 1-V site only***

Ba^+ in SXe spectrum interpretation





- Signal from 1-V Ba^+ on same deposit is similar size to 1-V Ba signal
- There is more background from surface fluorescence in the green that competes with Ba^+ peaks

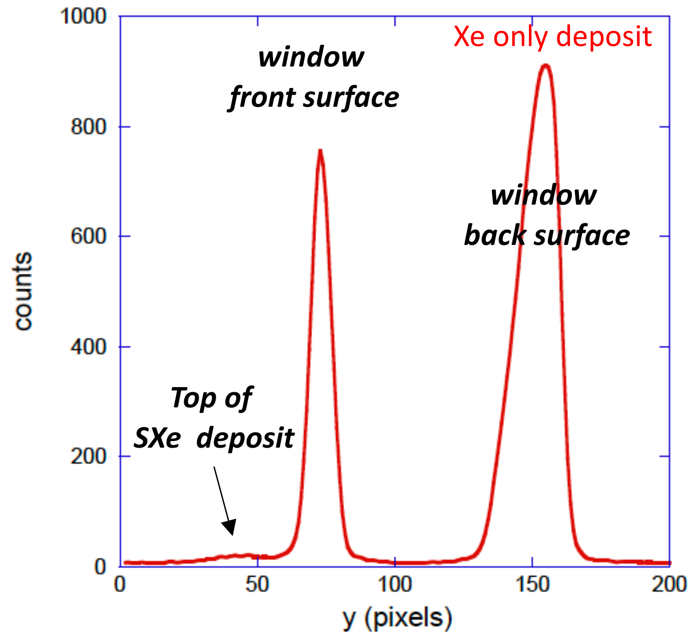


Note:

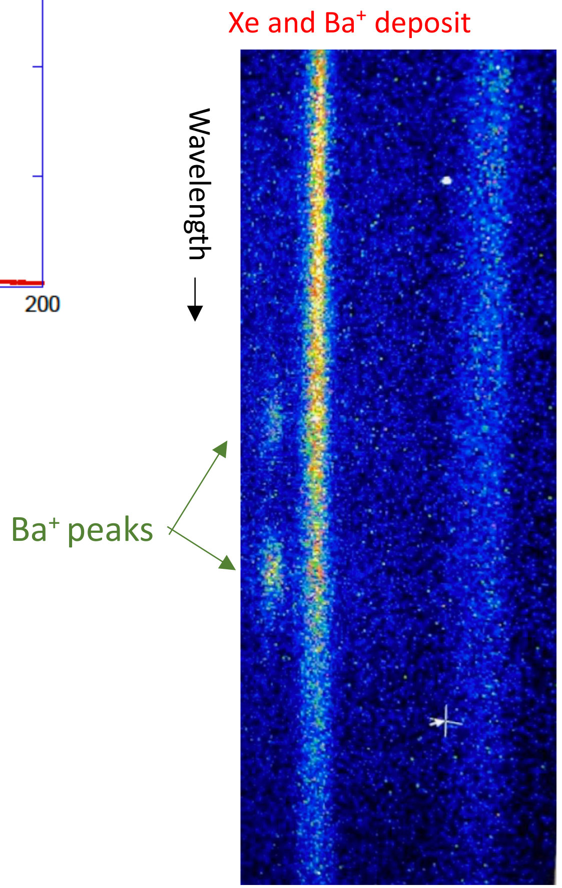
- Background spectrum similar to weak OH emission with UV excitation
- *May be H_2O accumulating on cold window or SXe surface???*

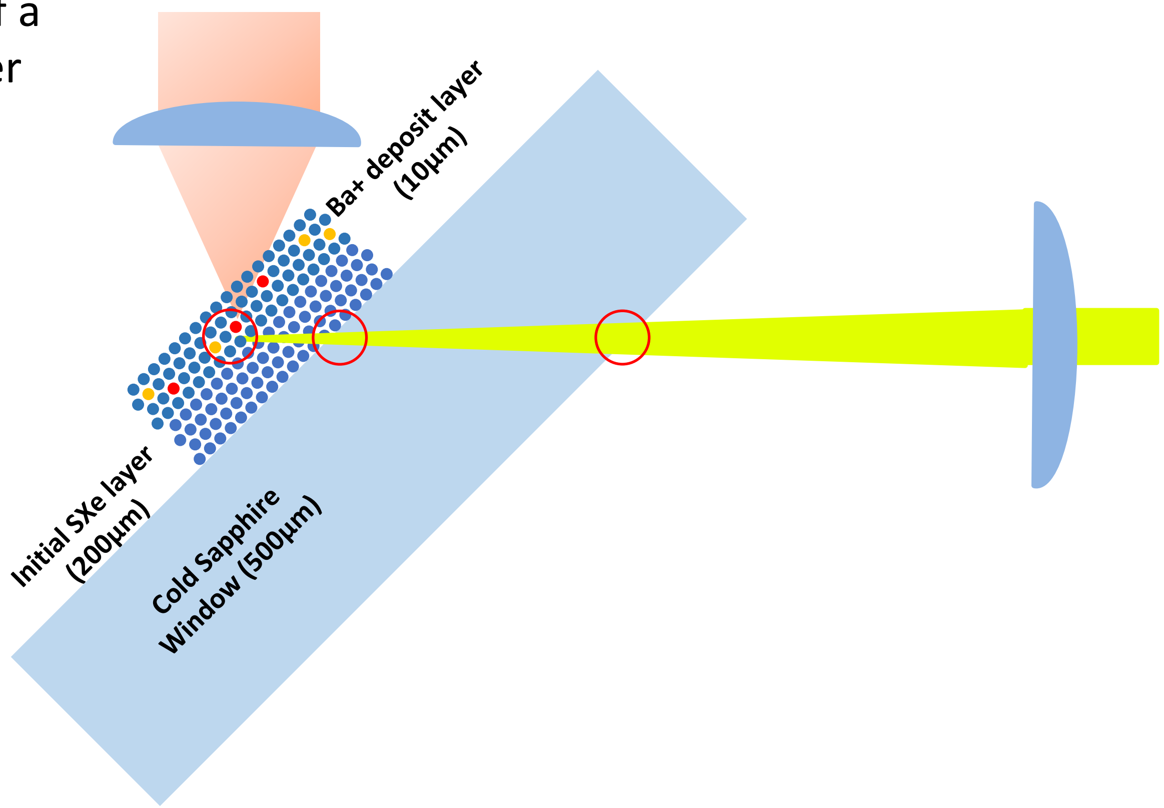
Progress in reducing background for imaging of single Ba^+ ions in SXe

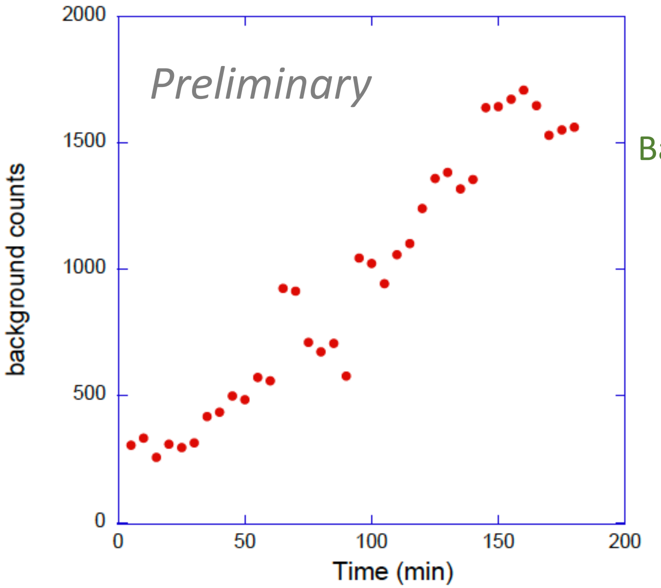
Transverse profile of spectrum frame



(1) Background at surface of a thick SXe layer is much lower than at window surface.





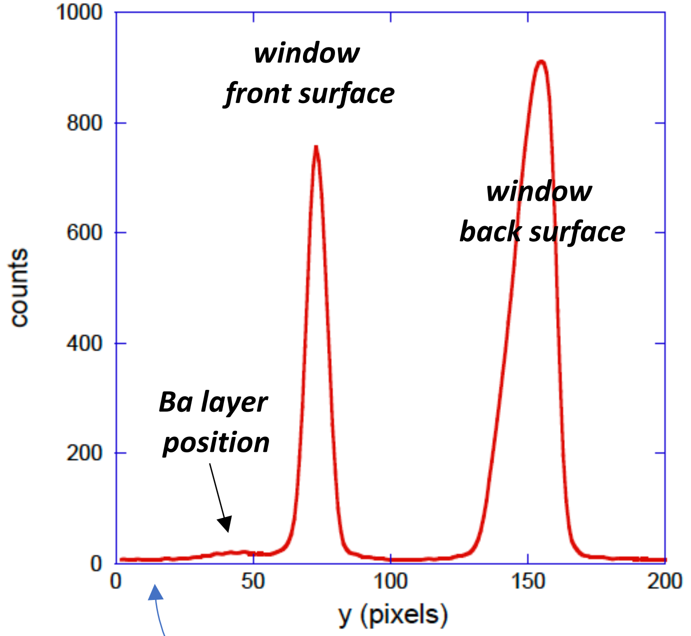


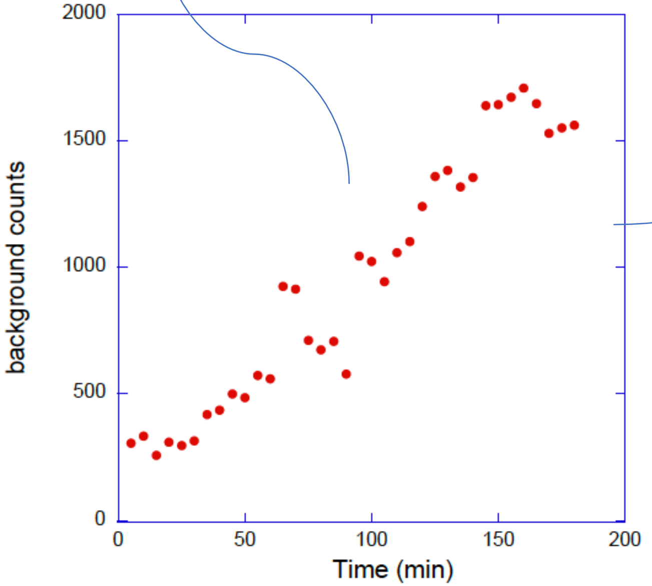
(2) Background at SXe surface increases linearly with time.

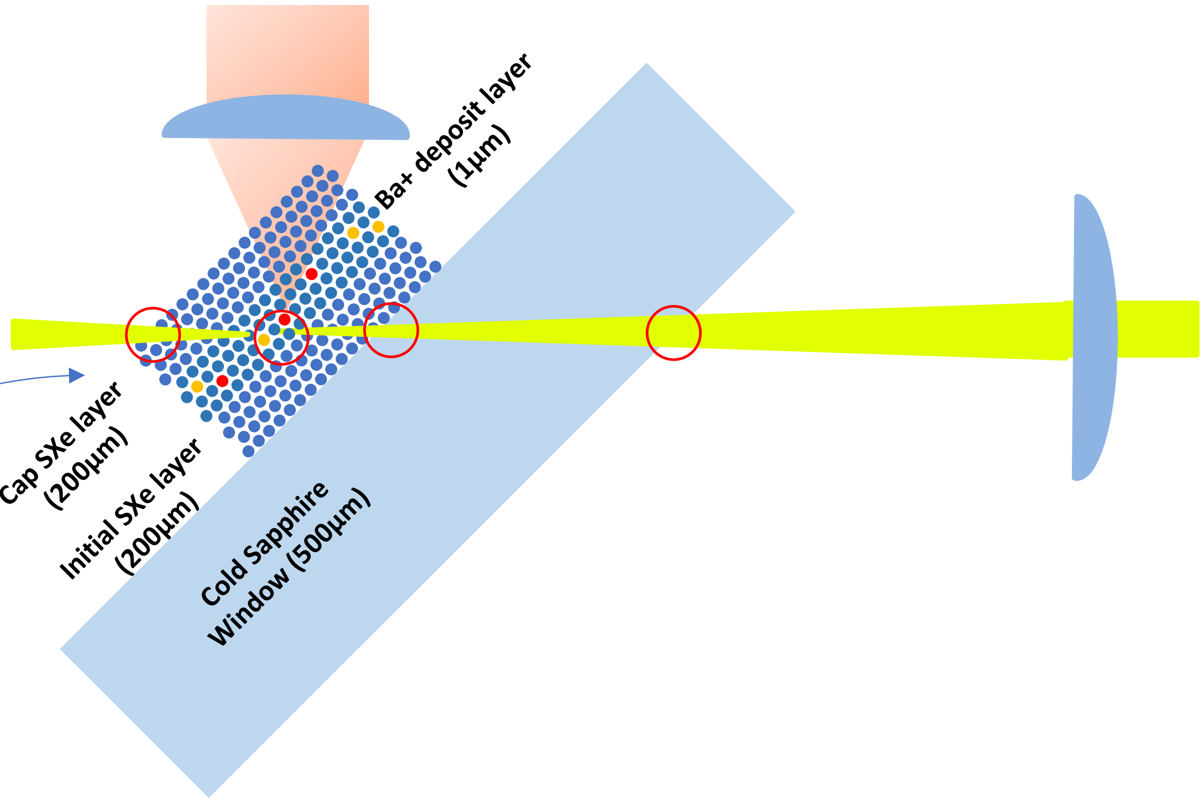
(3) Background seems to be due to residual gas H_2O depositing on SXe surface. *(would be none in nEXO)*

Reduce background by adding a SXe cap layer

Transverse profile of spectrum frame



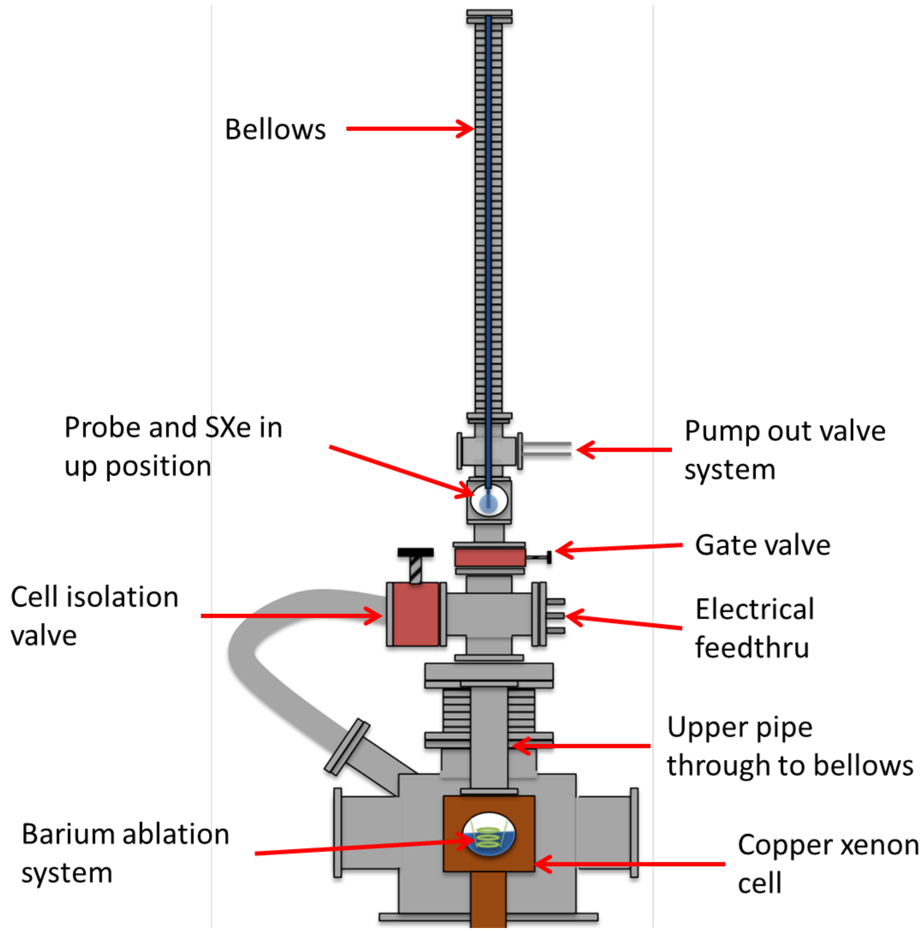




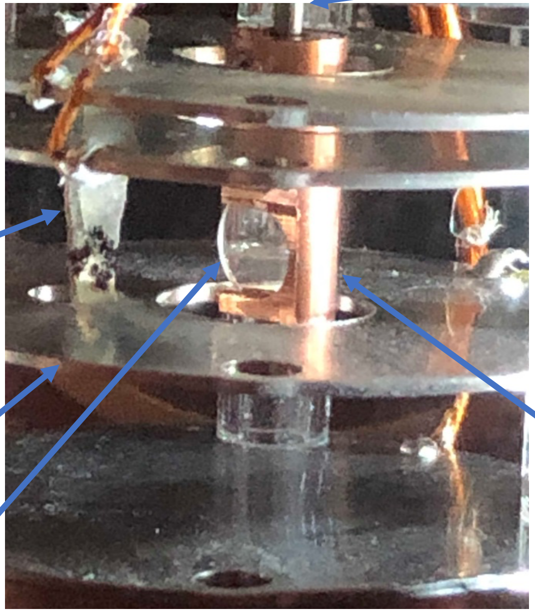
(4) With SXe cap layer the background will grow in region displaced from Ba^{+} deposit

Experiments on Ba/Ba$^+$ extraction on a cryogenic probe

Cryogenic probe and extraction system



Initial tests to see laser-induced fluorescence of barium atoms in SXe frozen from GXe



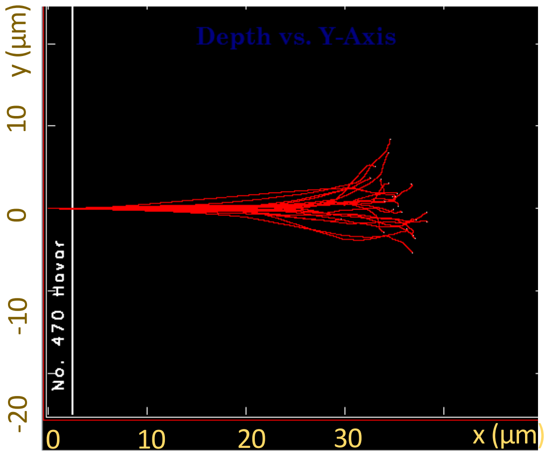
- Ions created using a pulsed Nd:YAG laser drawn to window during freezing of SXe from GXe.
- Probe raised to up position for laser spectroscopy

- *First spectroscopy has been done on Ba atoms in 1-V state at ~155 K on a sample of laser ablated Ba$^+$ ions in SXe collected in GXe and raised to extracted position.*
- *Now installing a probe that can be cooled with LN2 or LHe.*

Plans for a definitive online test of Barium tagging method

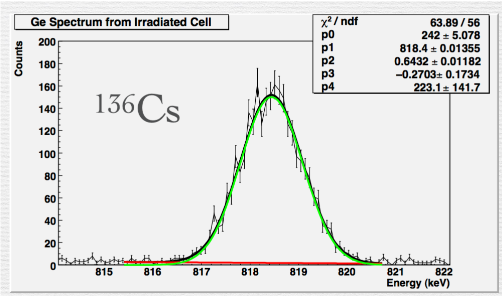
We plan to use radioactive ^{A}Cs or ^{A}La implanted or created in a LXe cell to demonstrate the locating, retrieval, and detection of the single beta decay daughter ^{A}Ba.

- ANL: Radioactive isotope are produced through ^{252}Cf fission. After stopping by the Gas catch, the radioactive ions are selected and accelerated in the CARIBU facility.
  - Accelerated beams: 2 MeV/nucleon, charge state around 10^{+} to 20^{+}, ~ 250 MeV total energy for Cs beams (^{139}Cs, ^{140}Cs…)
  - *Early experiments at ANL used low energy beams to study Ba desorption from SXe or metal surfaces.*
- TUNL or UK: Proton irradiation of natural Xe produces ^{136}Cs via $^{136}Xe(p,n)$, which beta-decays to ^{136}Ba. Production in GXe or LXe possible.
  - *Pilot demonstration at TUNL saw 30 nCi produced in 10 min on 1 atm ^{nat}Xe gas cell using 60 nA 7 MeV proton beam.*
- TRIUMF: Radioactive isotopes produced by 480 MeV p on UC. Fission and spallation products separated by ISOL.
  - Accelerated beams of higher mass Cs and lower mass La isotopes available in charge state 13^{+}-15^{+} to get ~250 MeV total energy.



250 MeV Cs beam goes through 2.5 μm Havar and will stop in 33 μm SXe

819 keV gamma from decay scheme observed at TUNL



Initial test in near term: simple 1D extraction and γ detection of Cs/La produced directly in liquid.

Conclusions

- Lots of progress on Ba tagging
- Some of the basic principles of several promising techniques have been demonstrated experimentally and by simulations
- Cautiously optimistic about future