Imaging individual Ba atoms in solid xenon for barium tagging in nEXO

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nEXO Collaboration

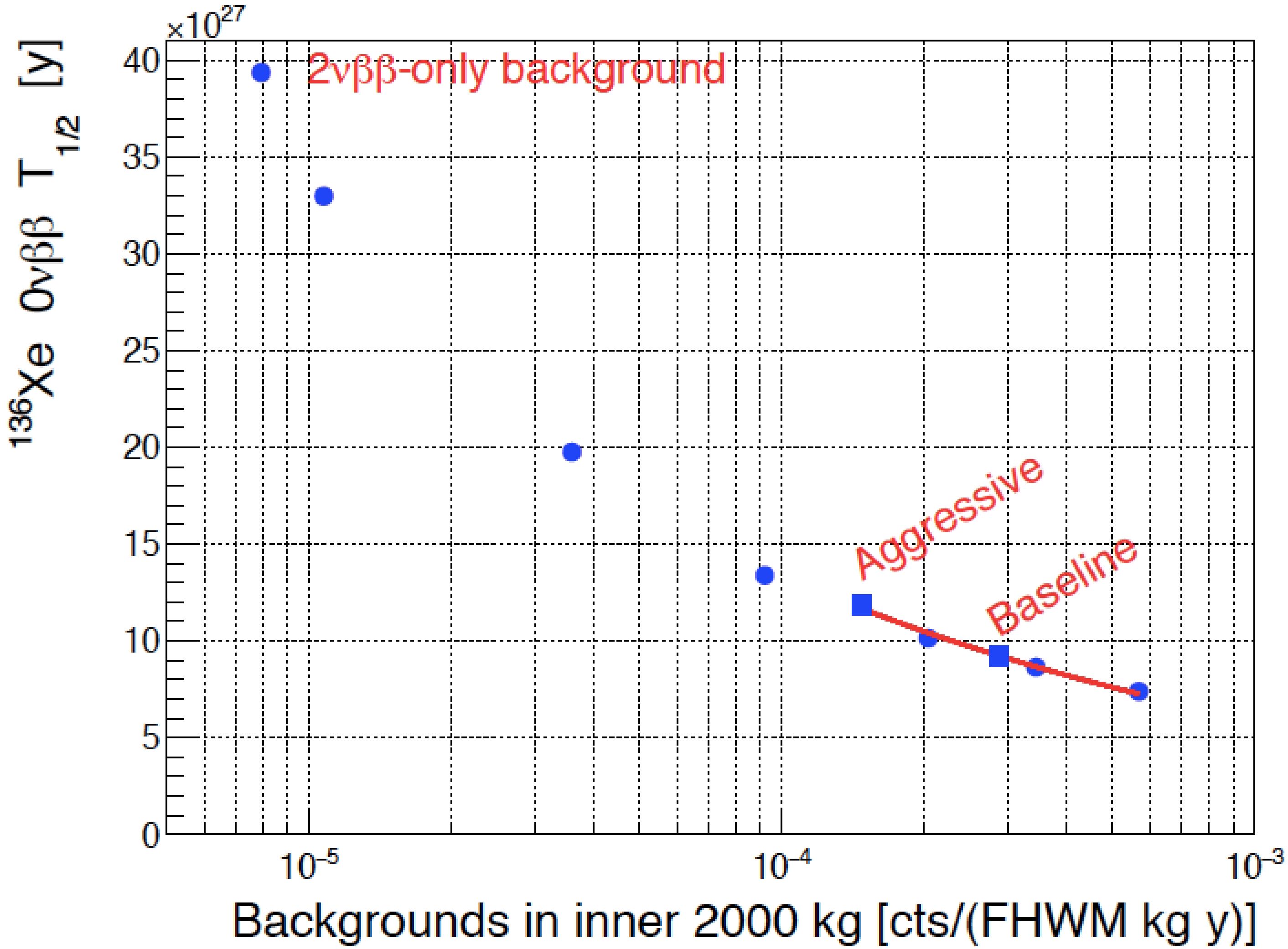
Extending the Sensitivity of Neutrinoless Double Beta Decay in the nEXO Detector

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

Current experiments detect the emitted electrons

- **Ba Tagging: also detect the daughter ^{136}Ba ion or atom located at the decay position.**
- **Potential to eliminate all but 2νββ backgrounds**

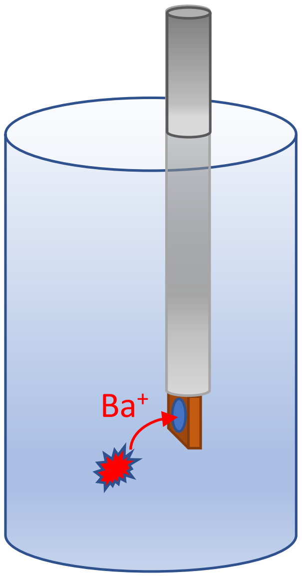
nEXO sensitivity vs. background

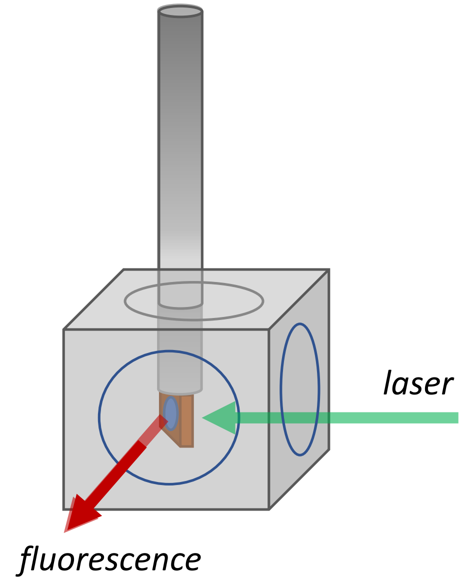


J. B. Albert et al., Phys. Rev. C 97, 065503 (2018)

Barium Tagging in Solid Xenon

- Locate the decay position with the TPC
- Insert a cryogenic probe and trap the Ba daughter in solid Xe
- Extract the probe and cool
- Tag the Ba daughter in the solid Xe via laser induced fluorescence





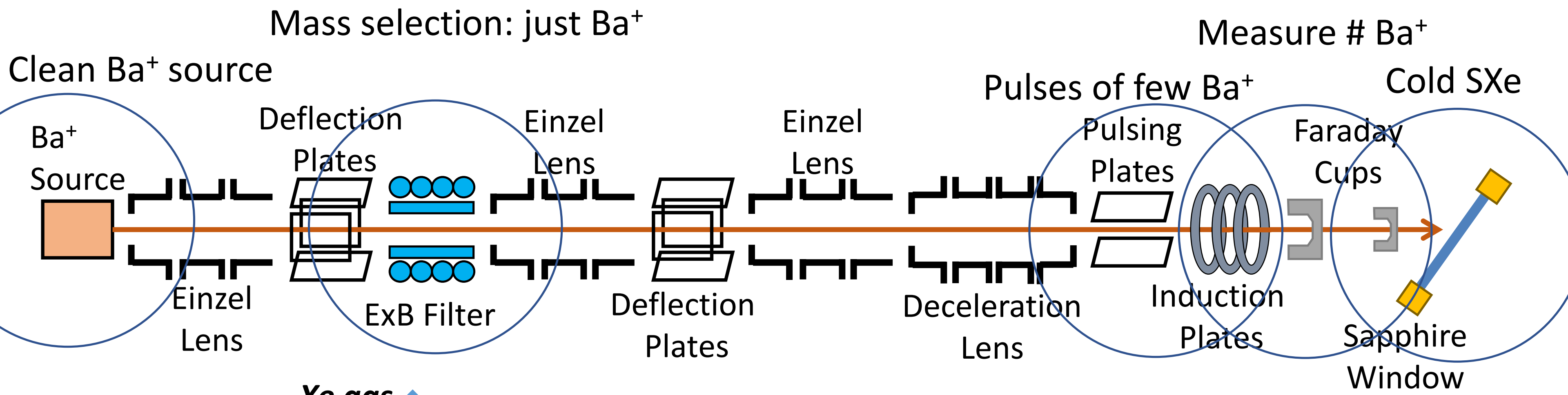
1 Ba ⟶ ββ decay

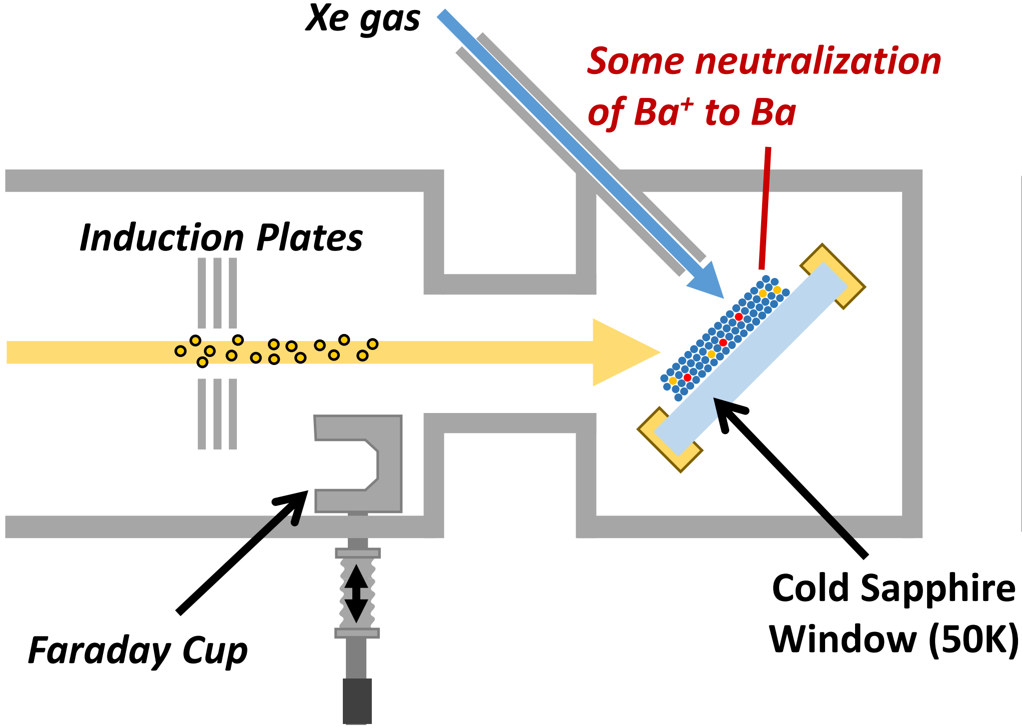
0 Ba ⟶ Not ββ decay

Probe in observation region - Use single Ba imaging technique we present in this work.

Requires counting of ***single*** Ba in solid Xe

Ba⁺ Ion Deposition System

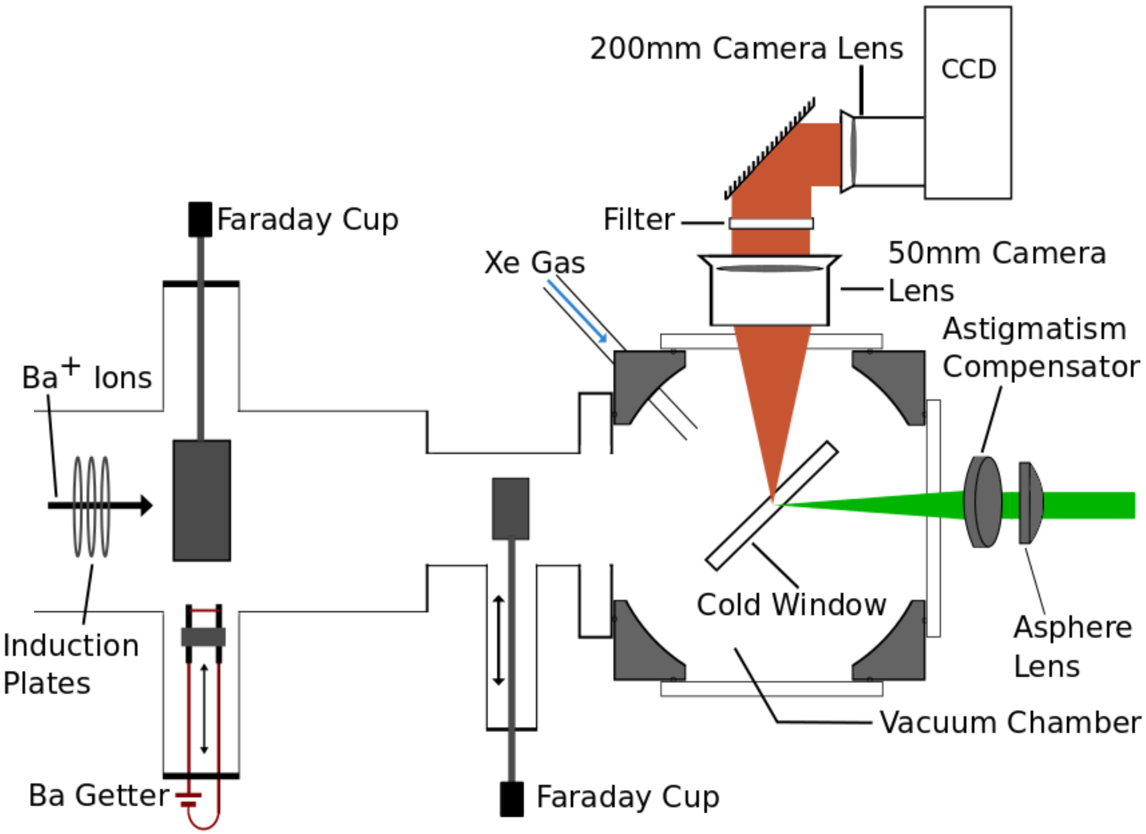


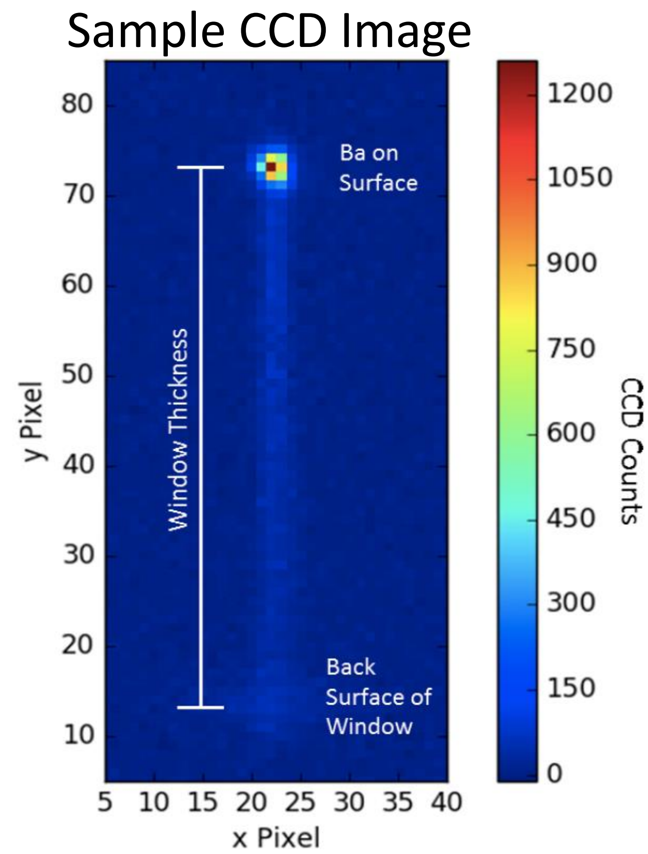


Deposition

1. Cool sapphire window to 50K
2. Begin Xe gas flow for ~ 6 s
3. Pulse Ba⁺ beam onto window
4. Stop Xe gas flow after ~ 6s
5. Cool window to 10K

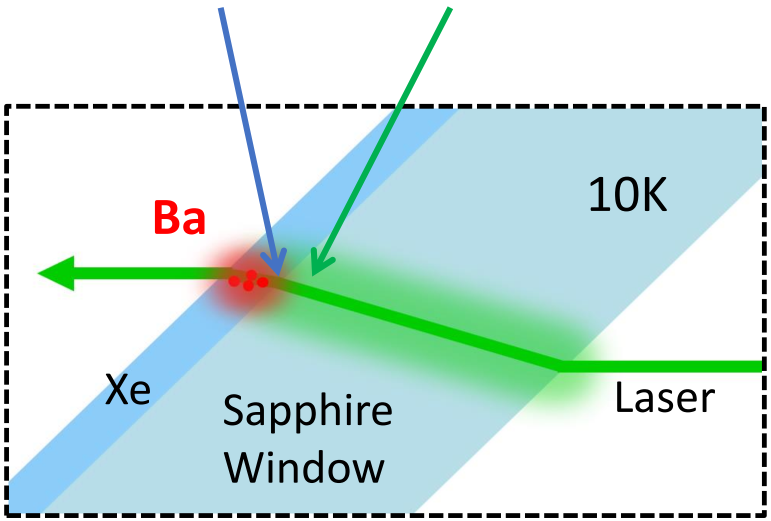
Observation of Ba in Solid Xe





Imaging and Background Reduction

Observed from above, there are two sources of background: surface, bulk

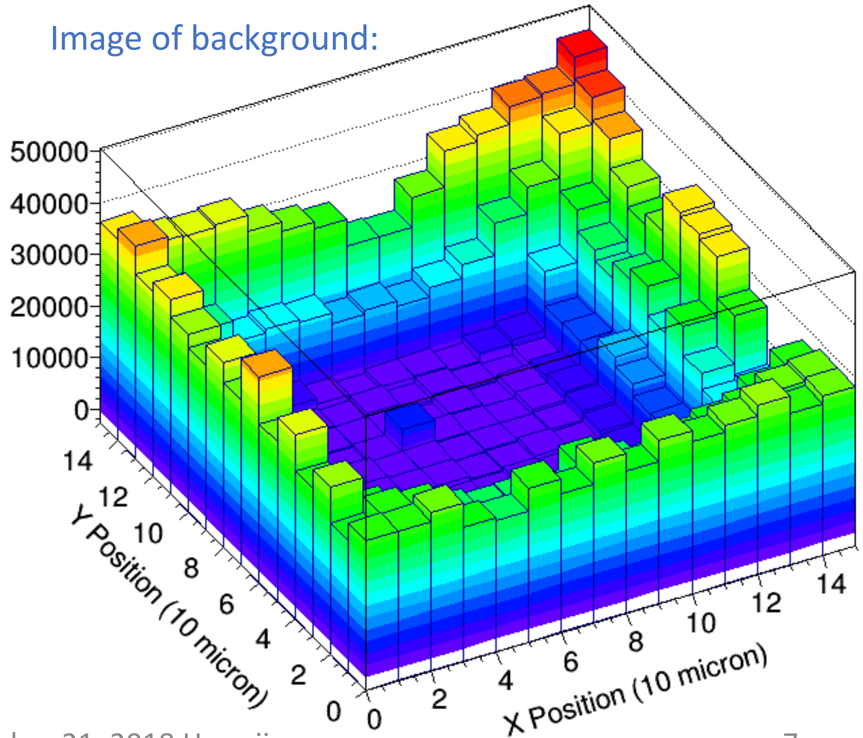


Cr^{3+} reduced with high purity windows

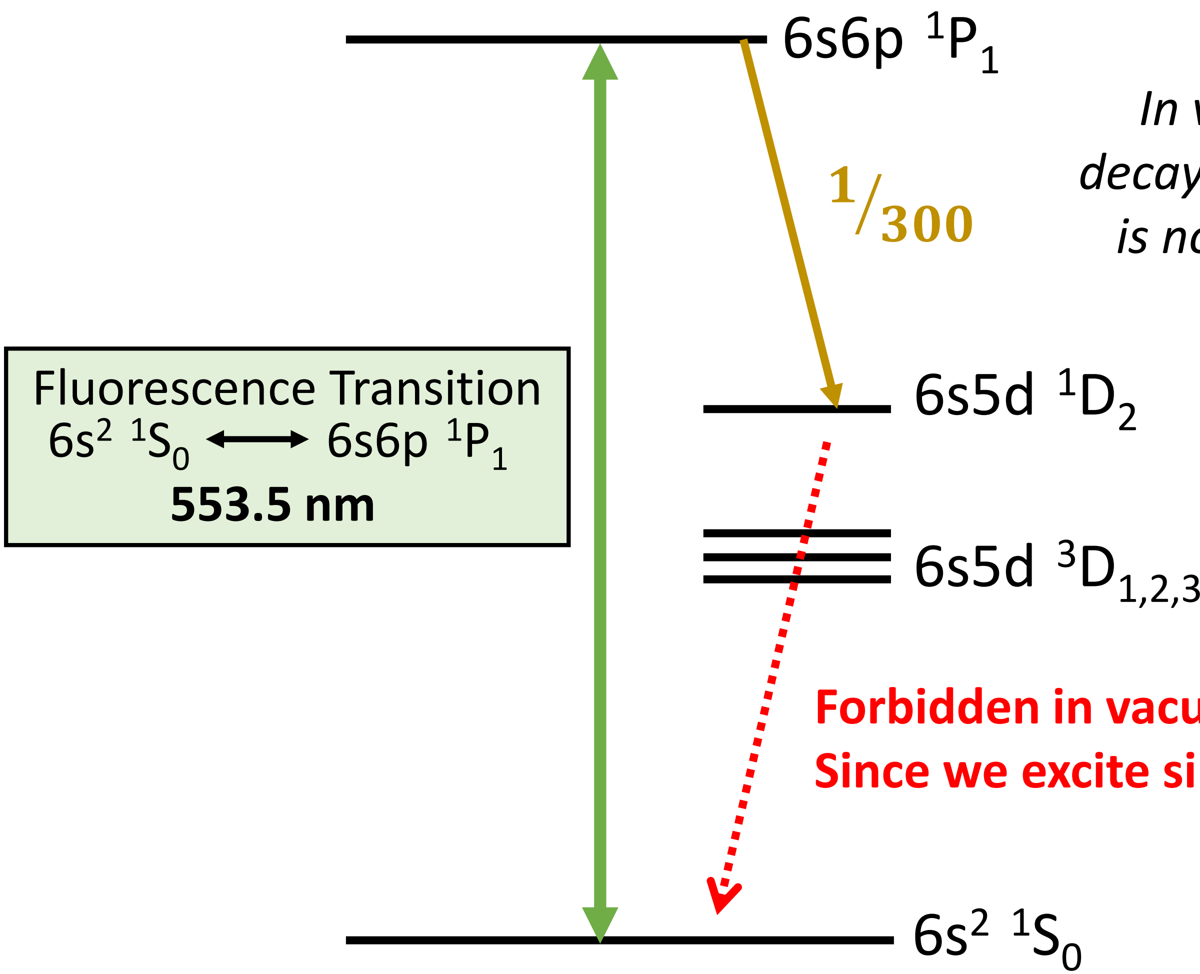
Surface background reduced via laser exposure

Raster 532 nm laser across window overnight at 100K → 30x background reduction in 90µm × 90µm area

Image of background:



Energy Levels of Ba in Vacuum



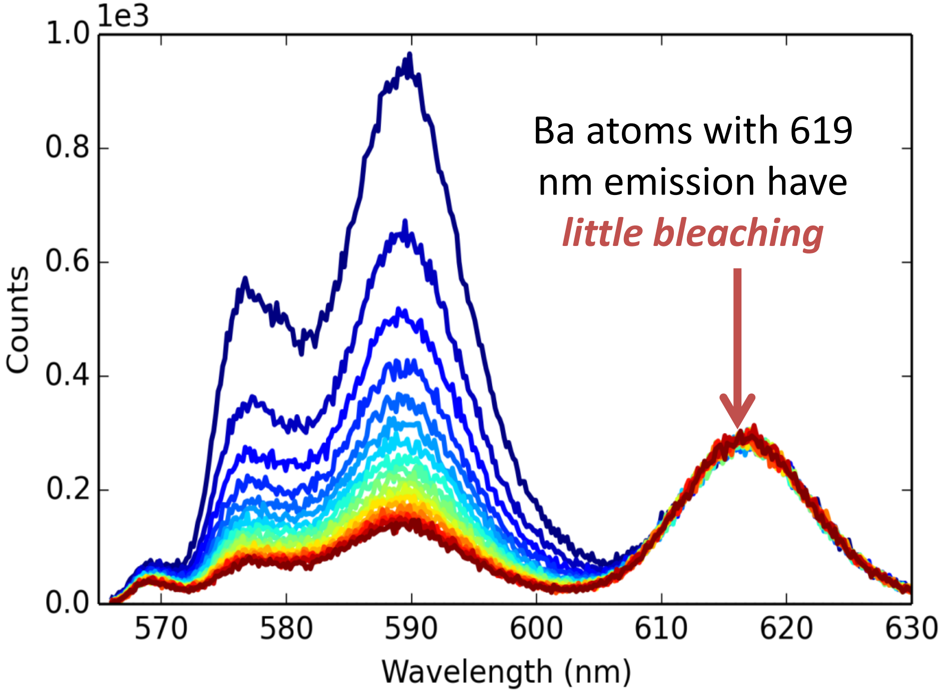
In vacuum, when the electron decays to the metastable D state, it is no longer excited by the laser.

Forbidden in vacuum but probably not in SXe
Since we excite single atoms up to 10^7 times.

Spectra of Ba in Solid Xe

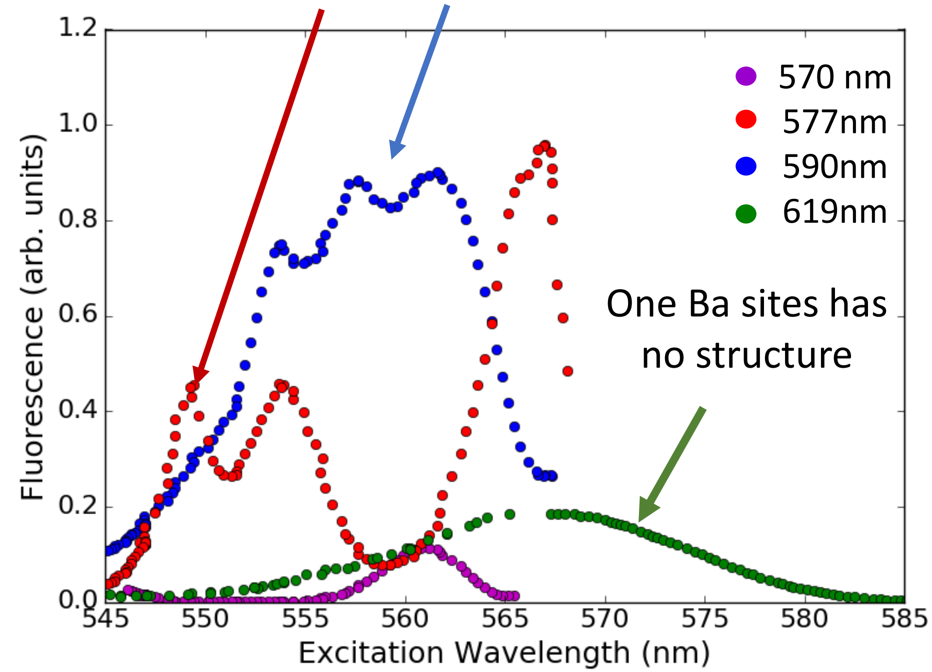
Ba Fluorescence Spectra

Three emission lines have
have more bleaching



Ba Excitation Spectra

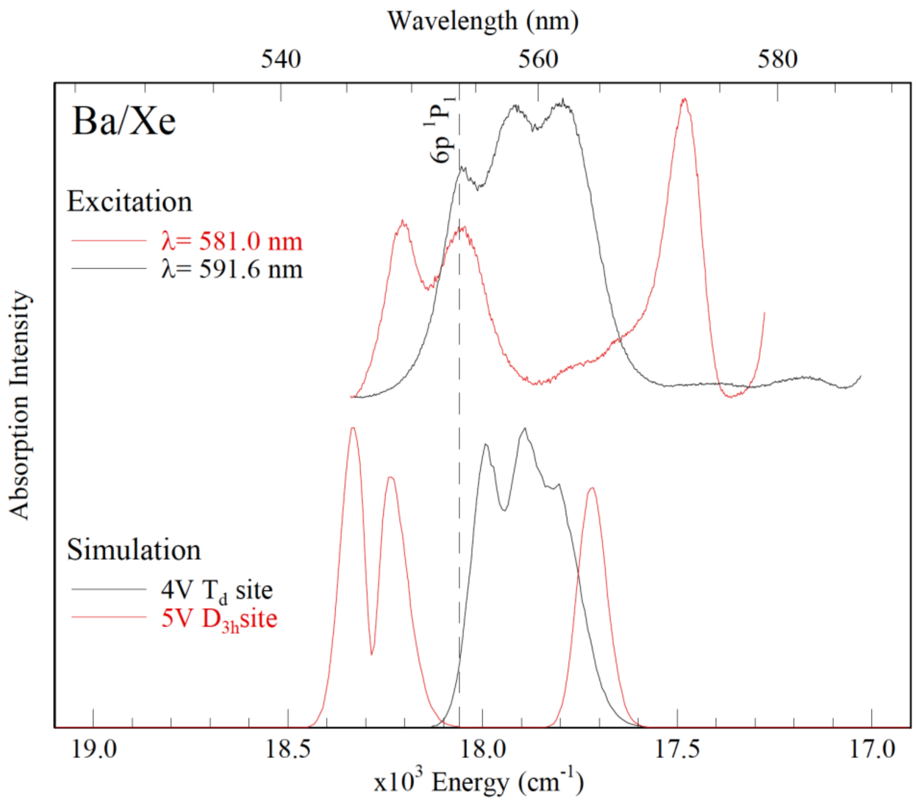
Two Ba sites have characteristic 3-peak excitation spectrum.



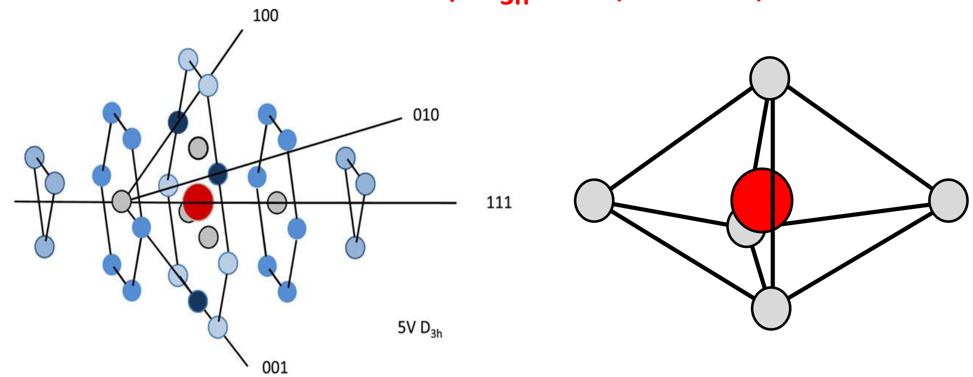
We have identified 4 distinct emission peaks, corresponding to 4 different matrix sites

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

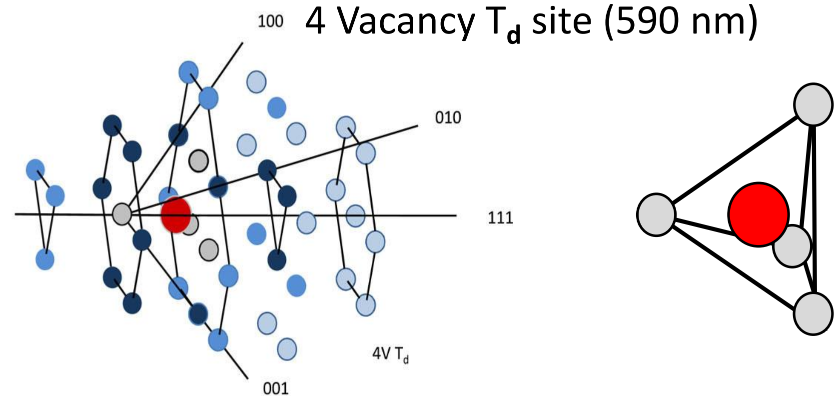
Identification of matrix sites of Ba in solid Xe for two peaks



5 Vacancy D_{3h} site (577 nm)

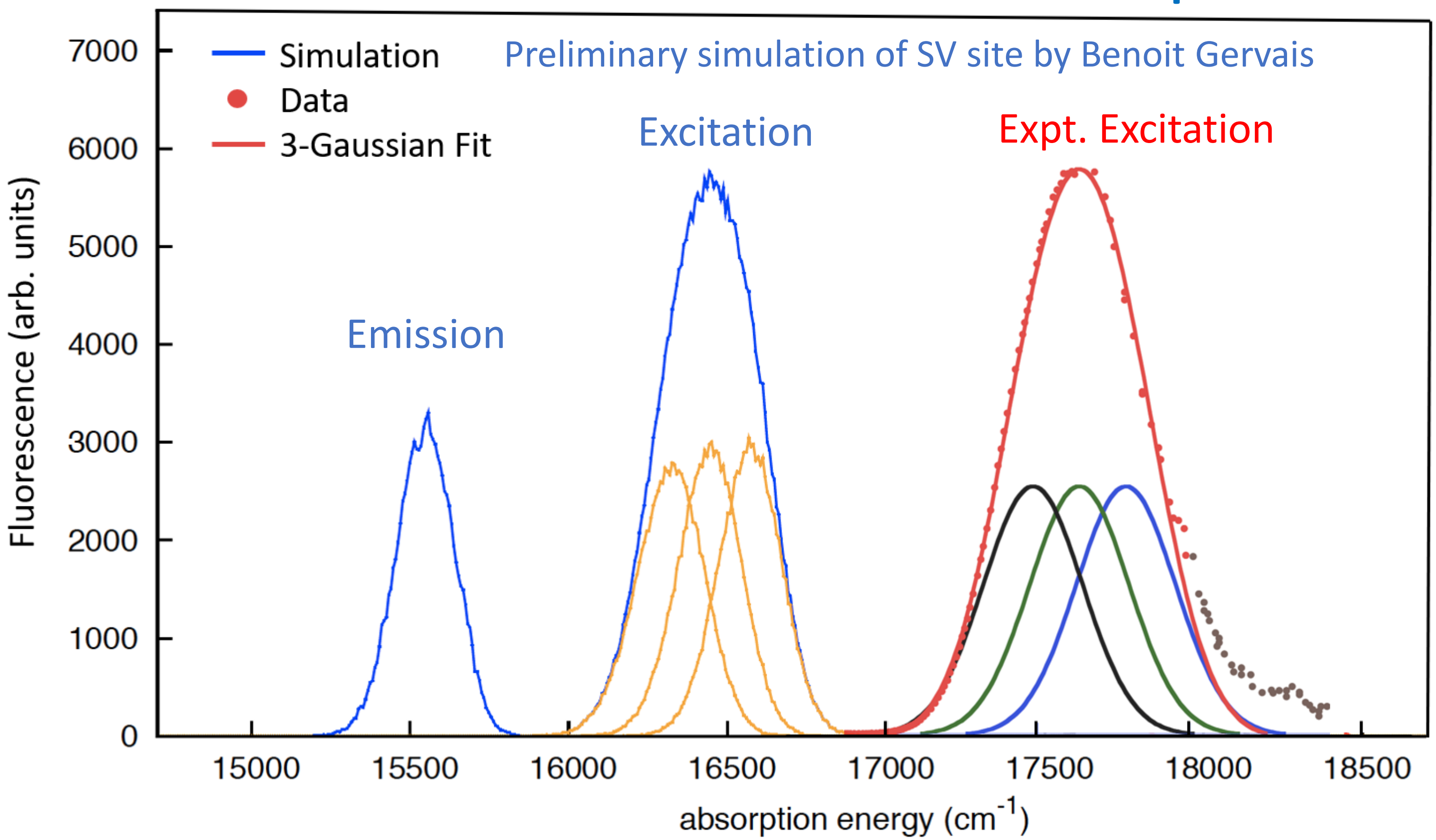


4 Vacancy T_d site (590 nm)



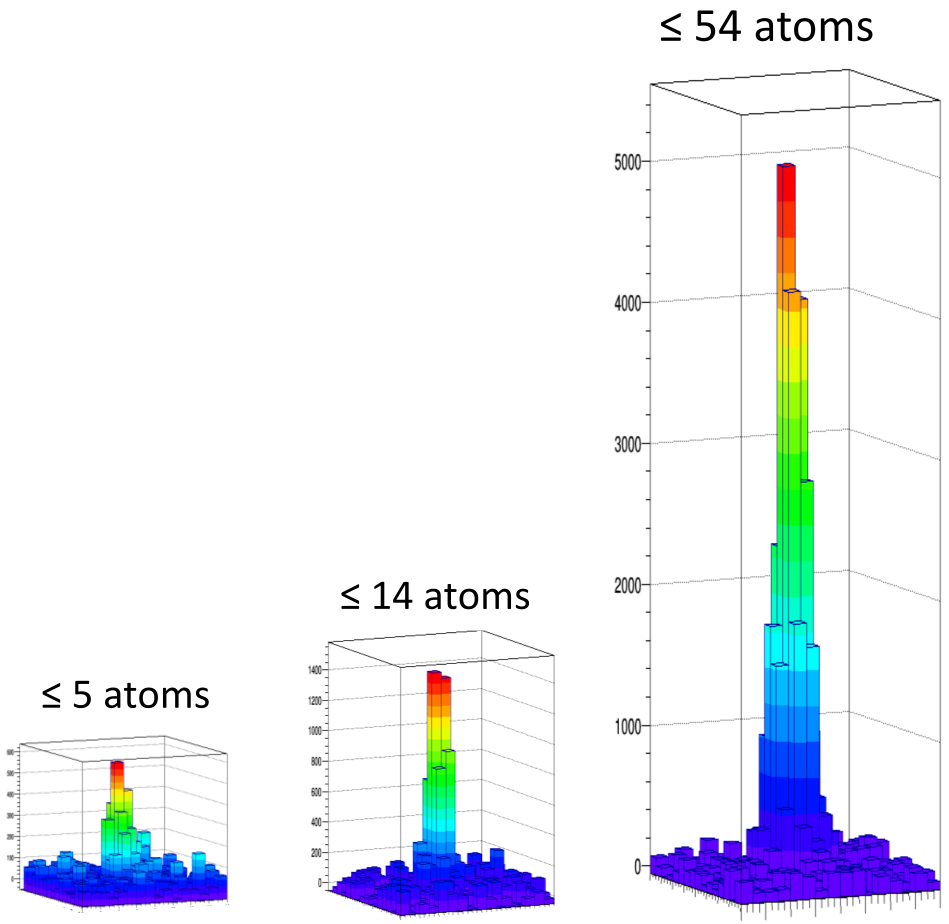
Davis, Gervais, and McCaffrey, *J. Chem. Phys.* **148**, 124308 (2018)

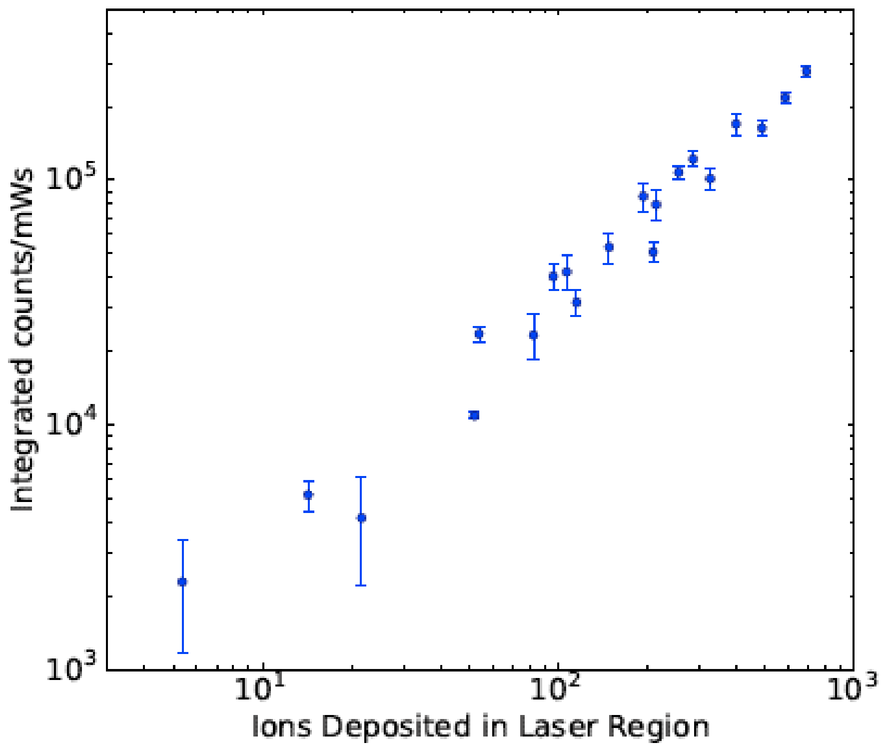
New: identification of 3rd matrix site of Ba in solid Xe: 619 nm peak



Single Vacancy (SV) site simulation shape qualitatively agrees with experiment
Cramped configuration is more sensitive to uncertainty in repulsive potential model

Fixed laser images of Ba in solid Xe

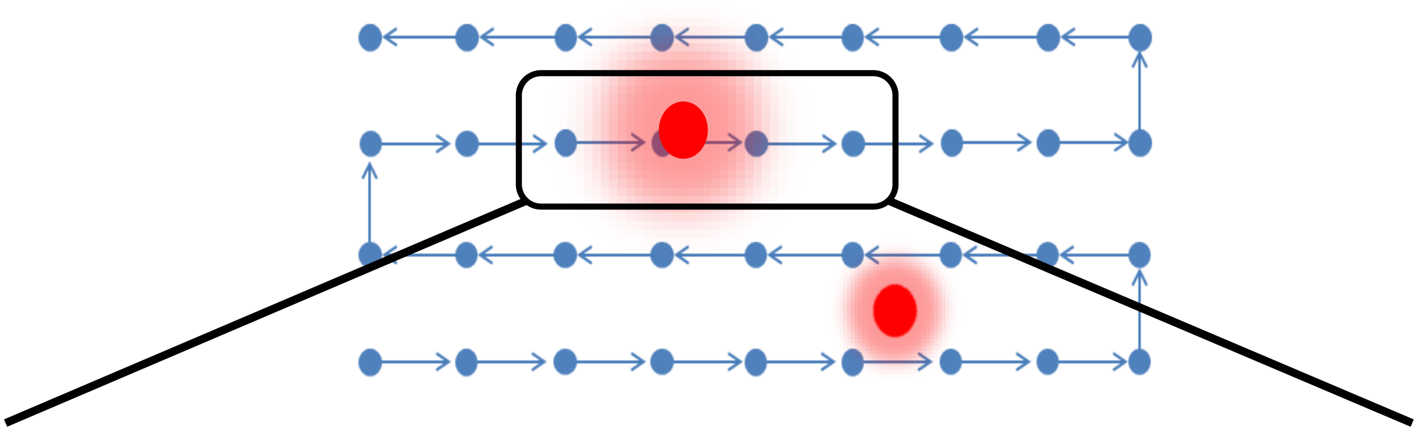


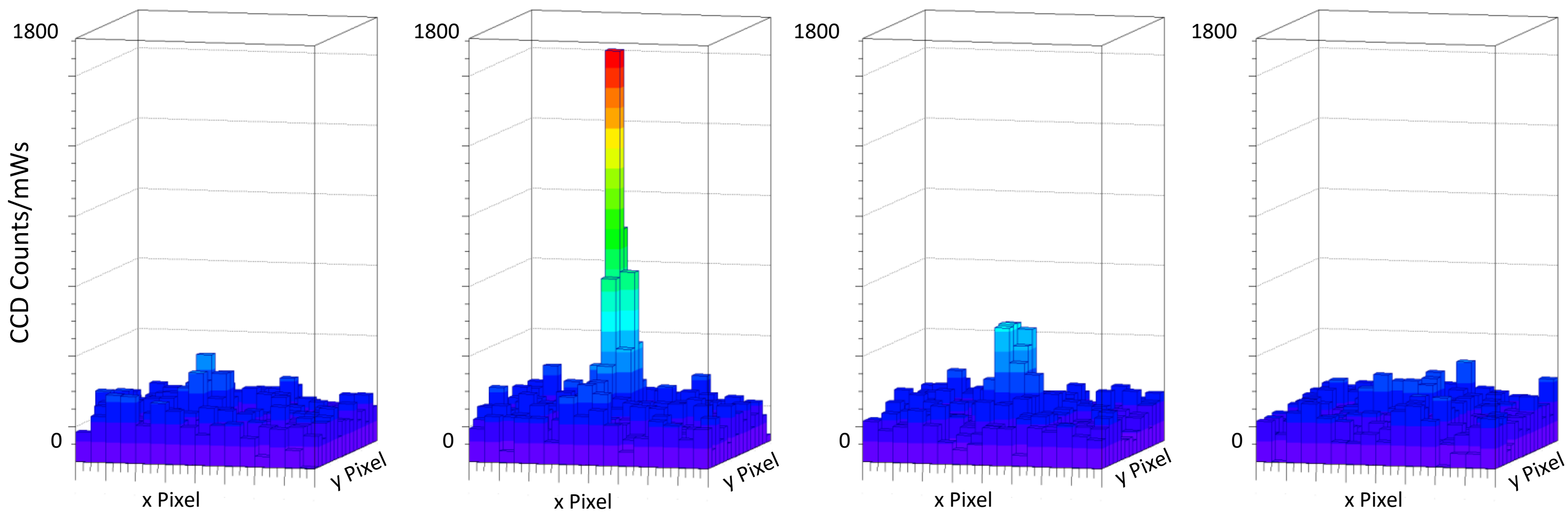


Fluorescence signal is linear with # of ions deposited: not Ba_n molecule!

Imaging single Ba atoms with laser scans

Each camera exposure is for a position in a grid:

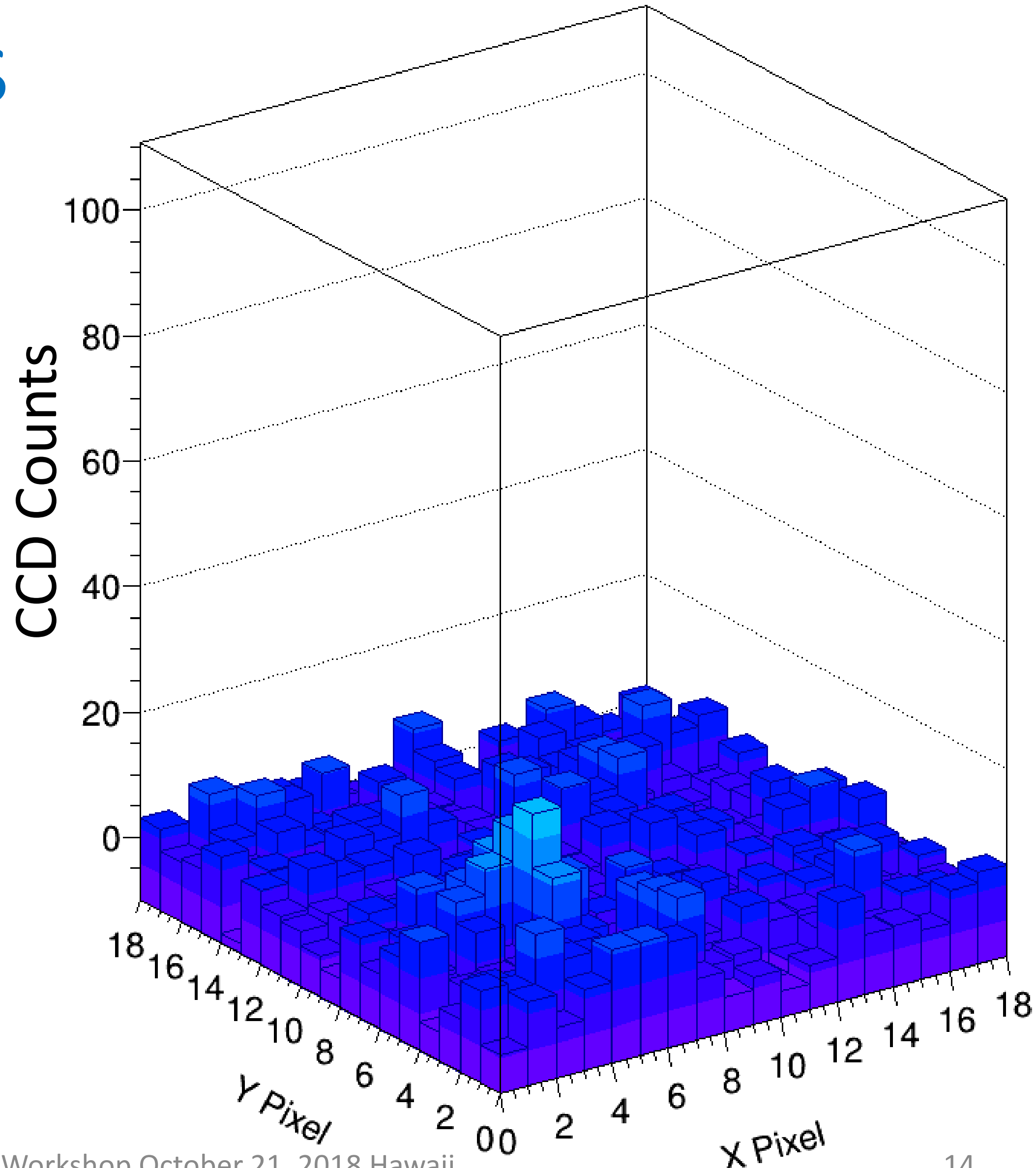




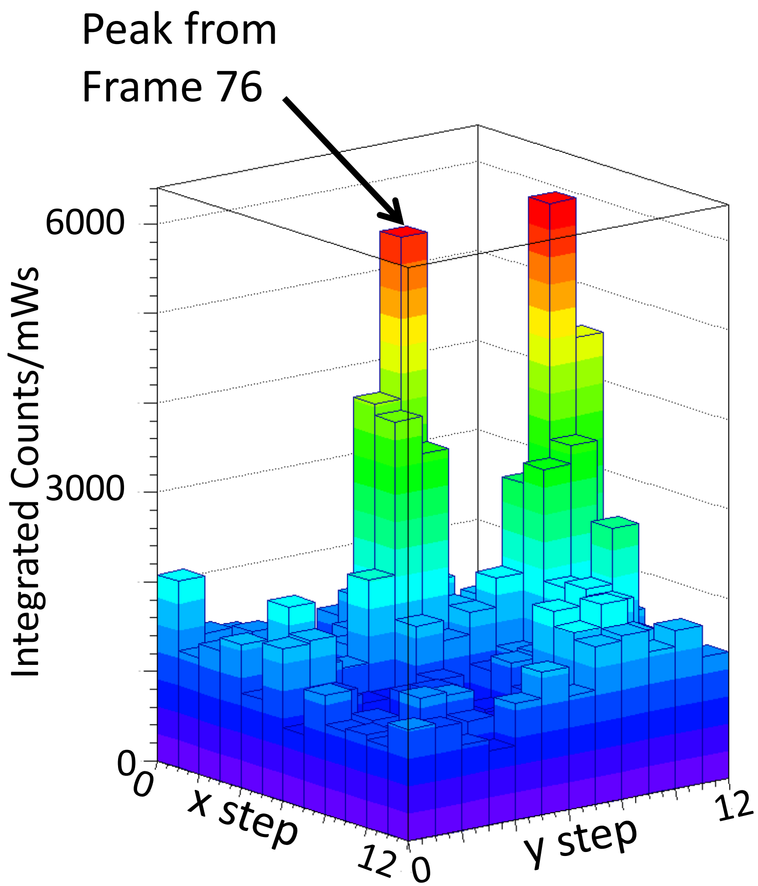
Scanning for Single Ba Atoms: raw CCD images

Scan Parameters

12 x 12 grid
x step: 4.0 µm
y step: 5.6 µm
Exposure: 3s/spot



Composite images of individual Ba atoms in solid Xe: counting Ba atoms



Making a Composite Image

Each frame is a CCD image of the laser at a grid spot

Between frames, the laser is moved to the next spot

Each frame is ntegrated around the laser region

Normalize by the laser exposure in mW*s

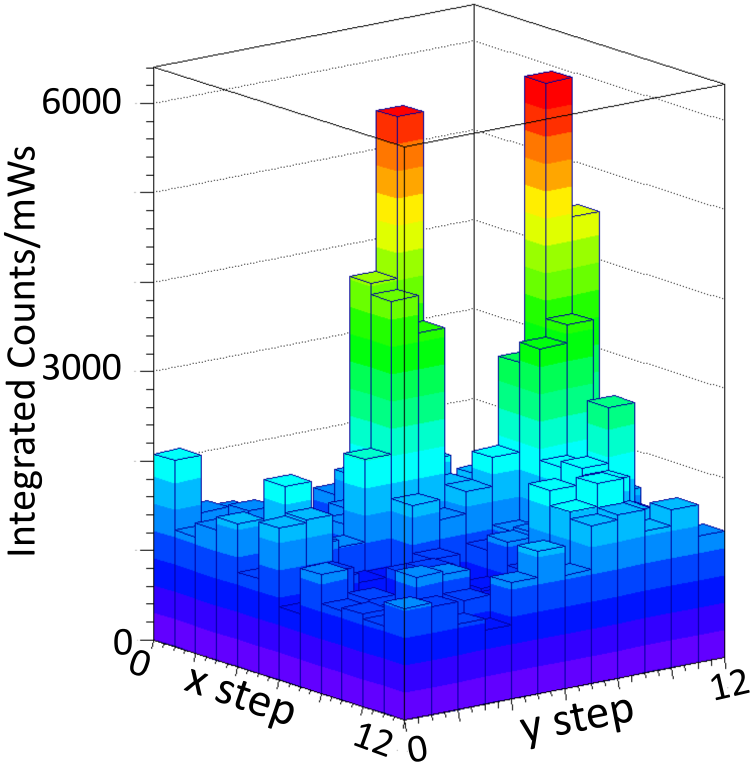
Signals plotted according to grid spot

Comment: count 2 detected Ba atoms in the scan area
48^{+5}_{-10} Ba^+ ions deposited in the scan area.

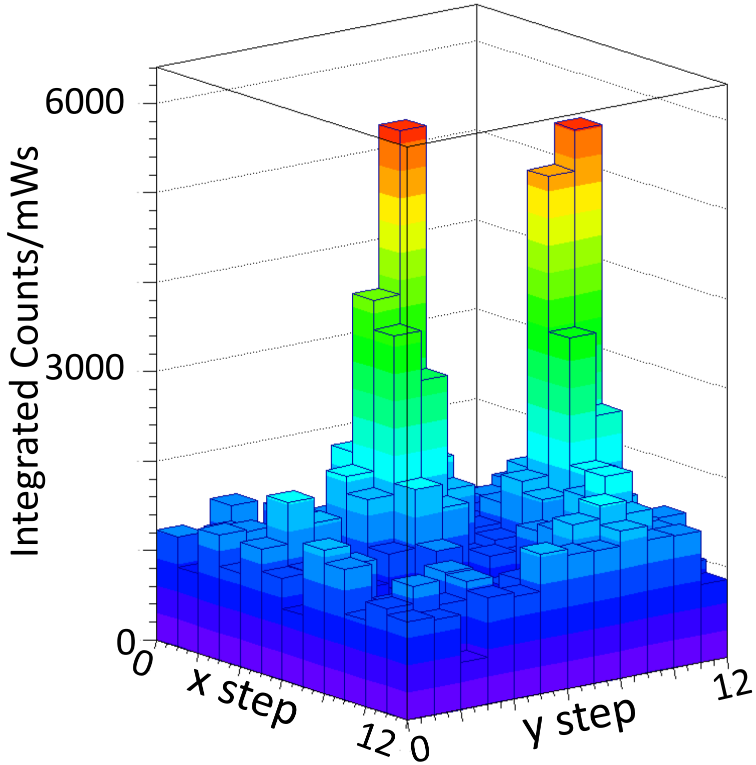
C. Chambers et al., arXiv 1806.10694, submitted to Nature

Composite images of individual Ba atoms in solid Xe: counting Ba atoms

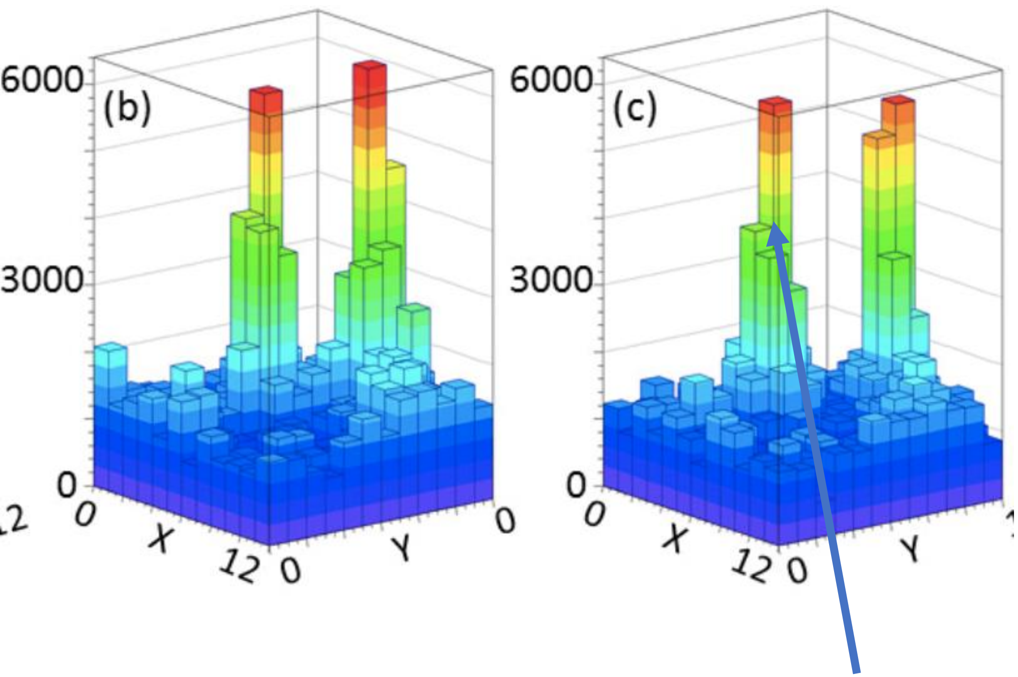
First Scan: 2 Ba atoms



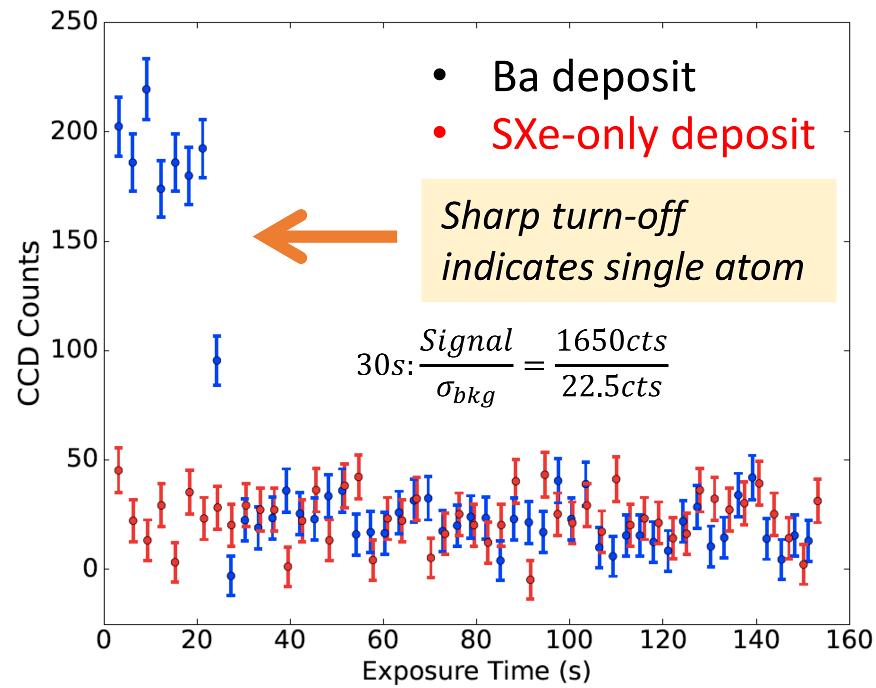
Repeat Scan: 2 Ba atoms still there



Sitting on one Ba atom



Counts in laser beam region per CCD frame



Then laser moved to near the left peak.

3s images are taken for 150 s.

Atoms emit for ~25s more, then turn off: *(>300 s for other atoms)*

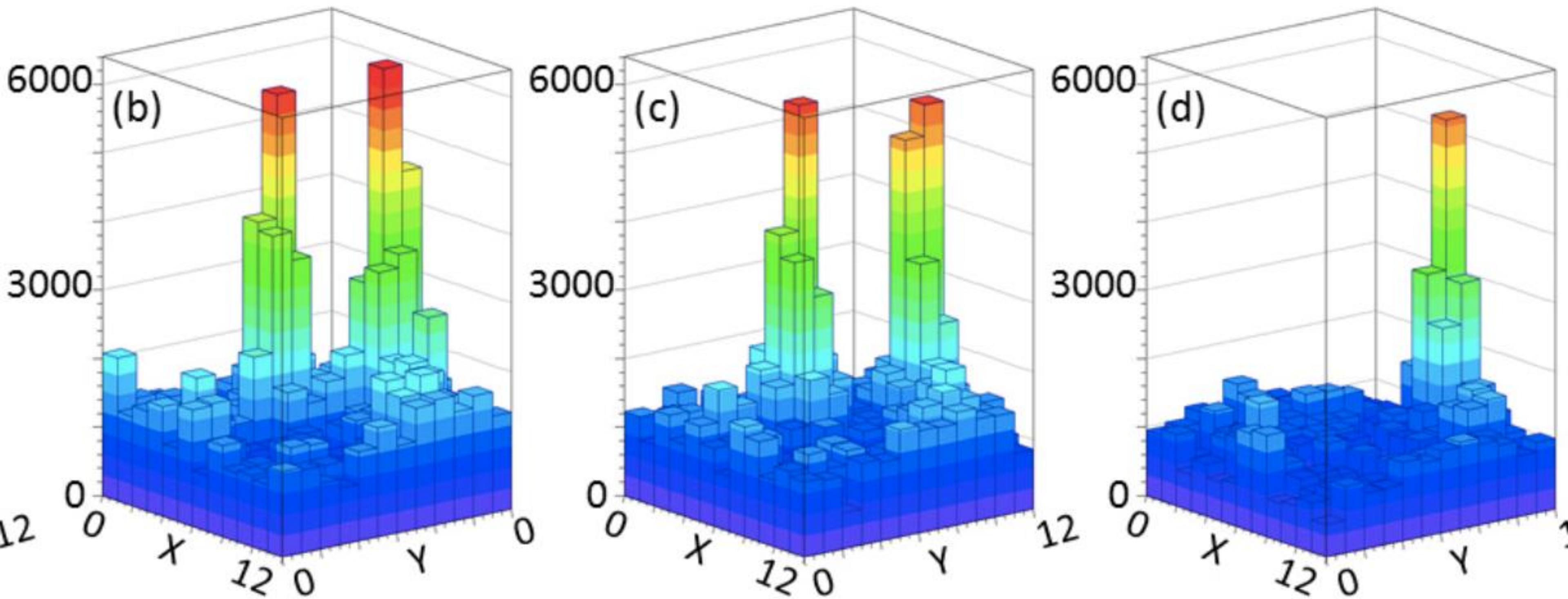
Lots of photons emitted by one Ba atom! (700,000 - >10^7)

Repeat scan after sitting on one Ba atom

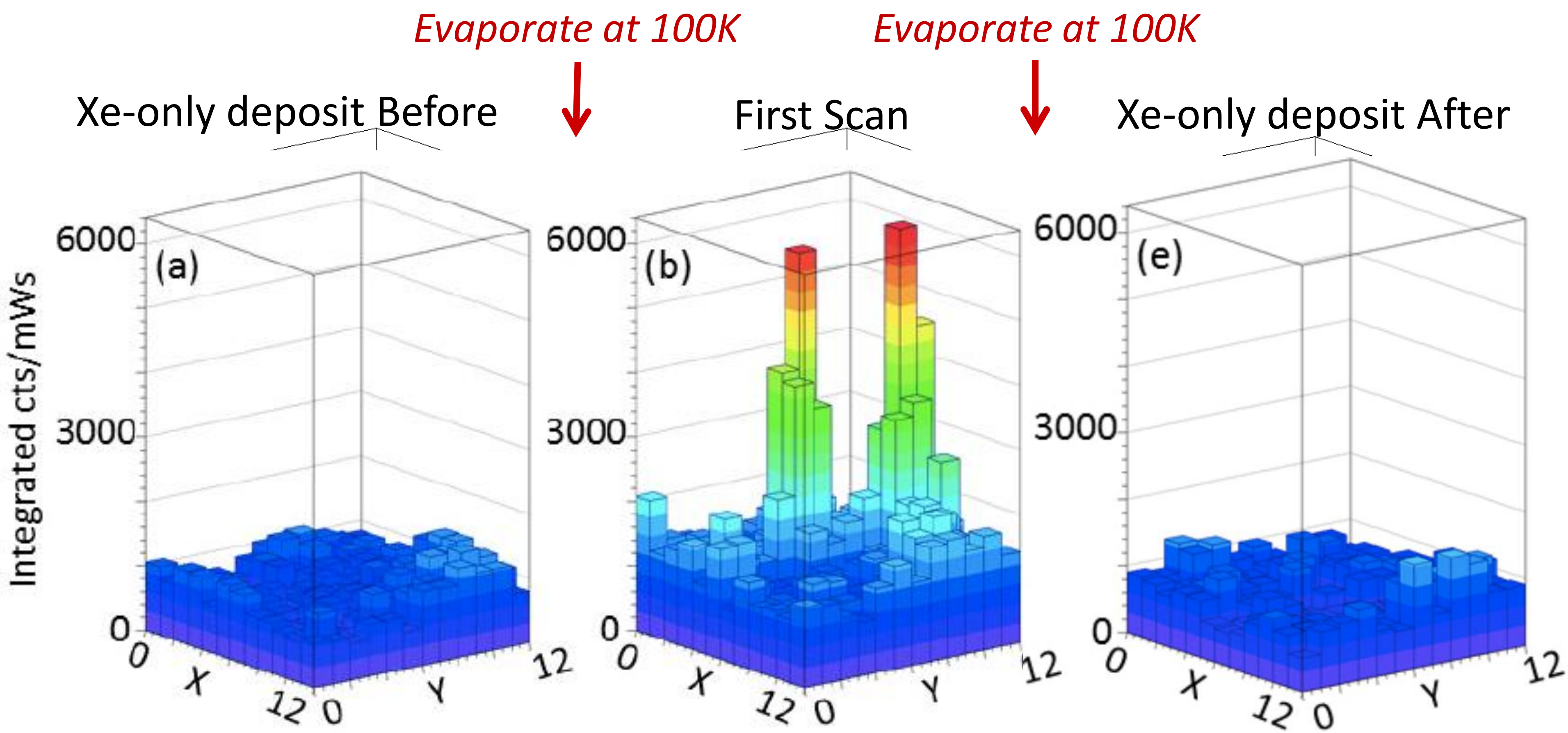
First Scan: 2 Ba

Repeat Scan: 2 Ba

Repeat Scan: after sitting near peak: one Ba is gone



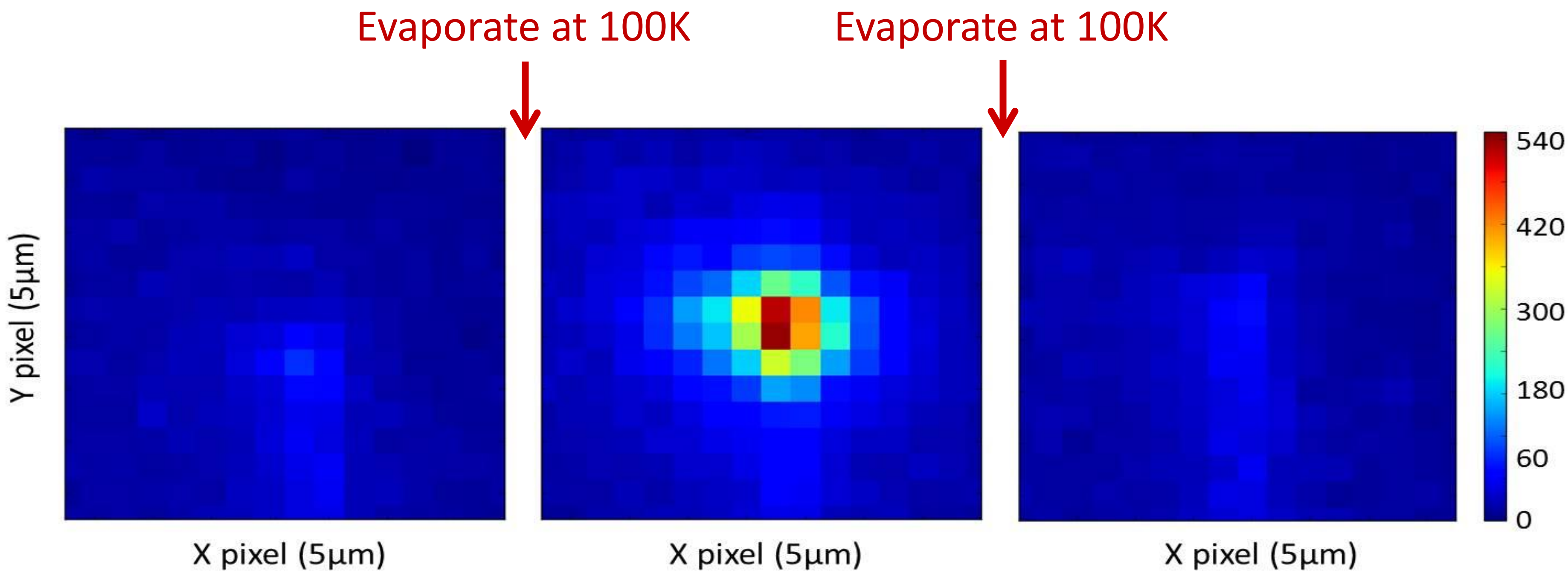
Comparing scans of deposits with and without Ba pulses



No Ba left behind after evaporation!

C. Chambers et al., arXiv 1806.10694, submitted to Nature

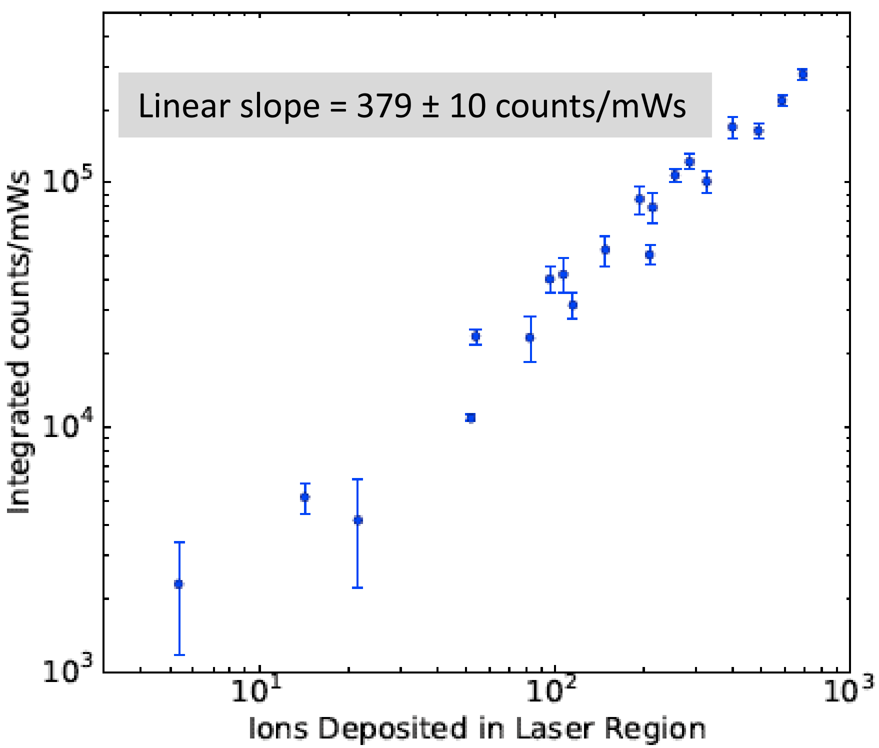
Erasing a large Ba deposit



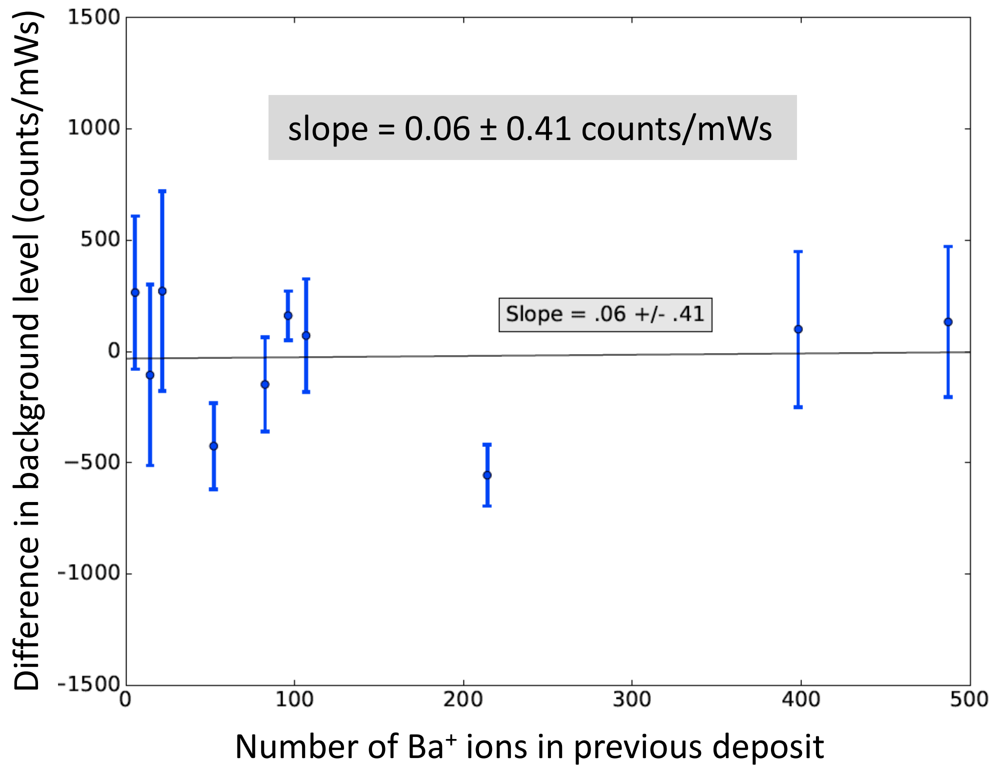
Even after a large deposit (7000 ions) all detectable Ba atoms are removed
Thus no "history effect" interfering with subsequent deposits

Quantifying erasure of Ba deposits

Ba counts/mWs



After-Before SXe only counts/mWs

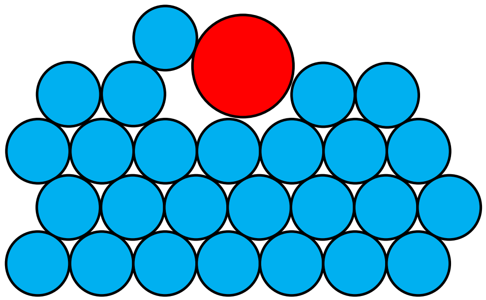


Limit of <0.16% Ba signal after evaporation
Thus no "history effect" interfering with subsequent deposits

C. Chambers et al., arXiv 1806.10694, submitted to Nature

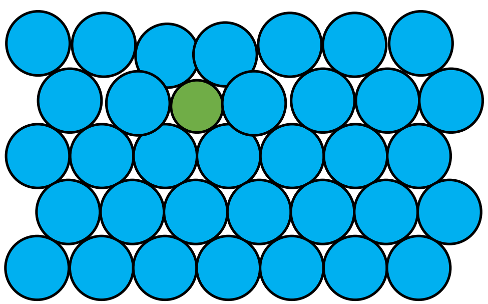
Comment on formation of matrix sites of Ba in Solid Xe

Incident Ba Atom
$r_{Ba\text{-}Xe} = 5.5$ Å



Xe
$r_{Xe\text{-}Xe} = 4.4$ Å

Incident Ba^+ Ion
$r_{Ba^+\text{-}Xe} = 3.6$ Å



Ba atoms are too large to fit in a single vacancy (SV) site, preferring the 4- and 5-vacancy sites.

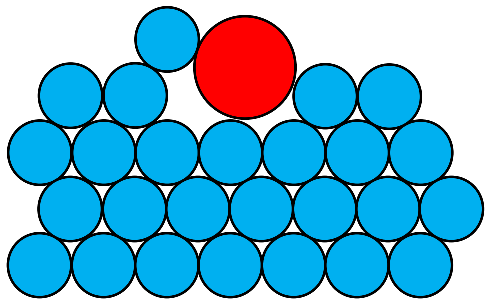
Ba implanted as an ion has a much tighter bond to Xe, thus preferring the SV site

This has already been demonstrated experimentally for Na^+ ions in SAr: D. C. Silverman and M. E. Fajardo, J. Chem. Phys. 106, 8964 (1997).

Comment on formation of matrix sites of Ba in Solid Xe

Incident Ba Atom

$r_{Ba\text{-}Xe}$ = 5.5 Å



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Xe

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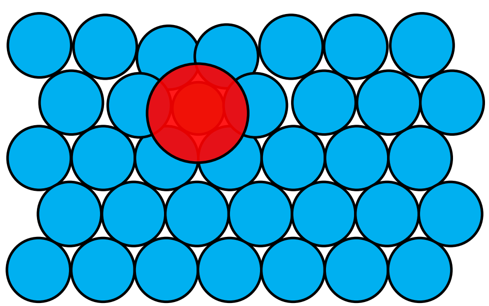
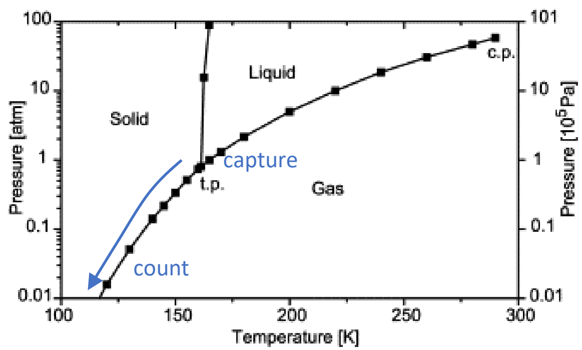
Incident Ba^+ Ion

$r_{Ba^+\text{-}Xe}$ = 3.6 Å

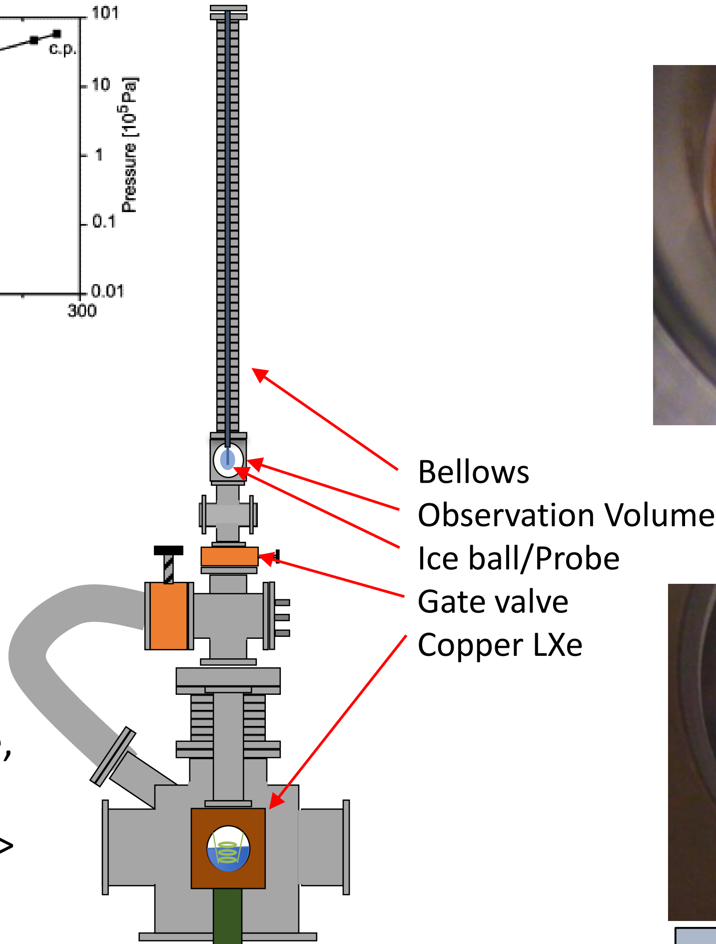


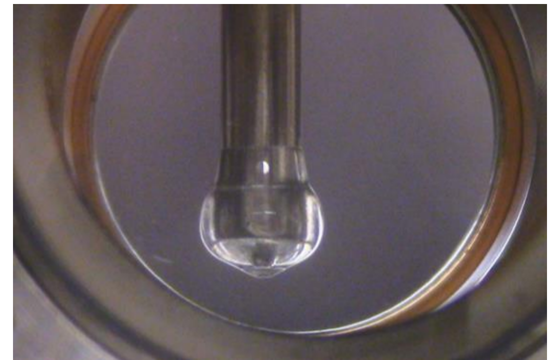
Ba^+ then neutralizes later to Ba, but is trapped in the cramped SV site by the Xe matrix.

Apparatus to capture Ba in SXe on a cryoprobe



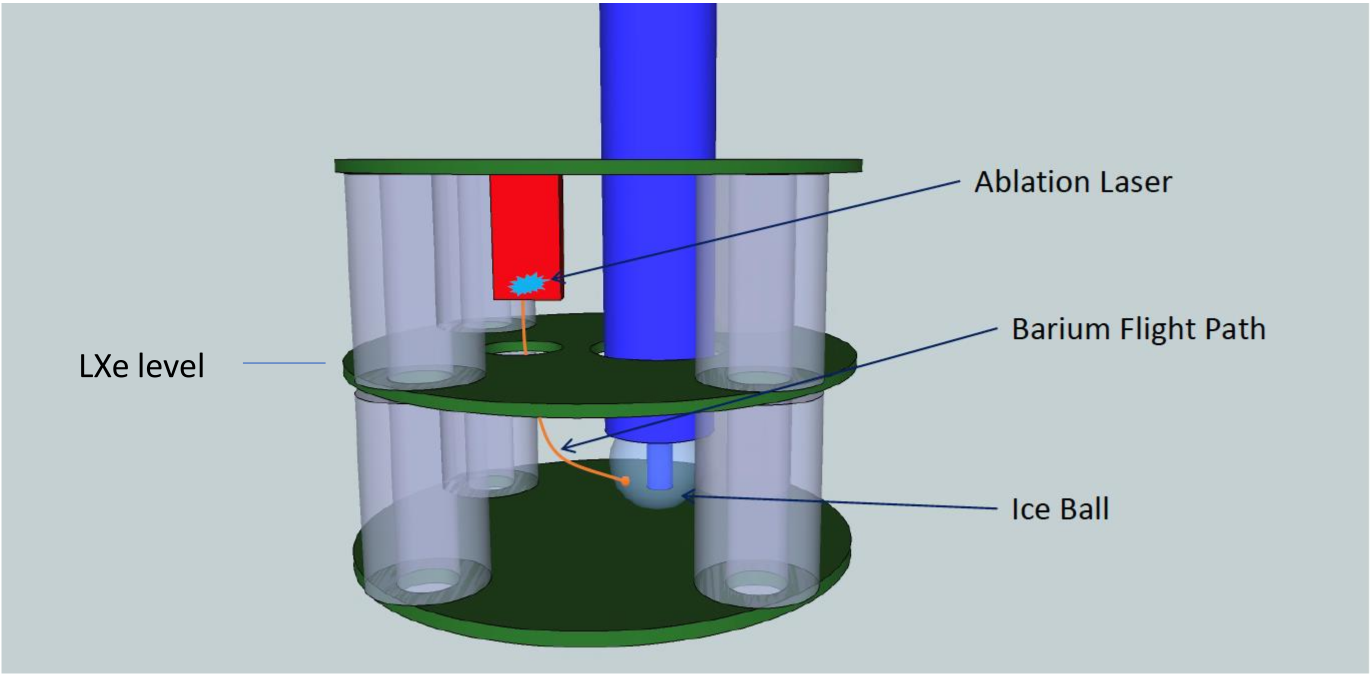
- Capture Ba in SXe at >161K.
- Raise probe to observation region.
- After close gate valve, reduce probe T and reduce Xe gas pressure, following gas-solid vapor pressure curve -> 10K.





Successful freezing from LXe and extraction to observation volume

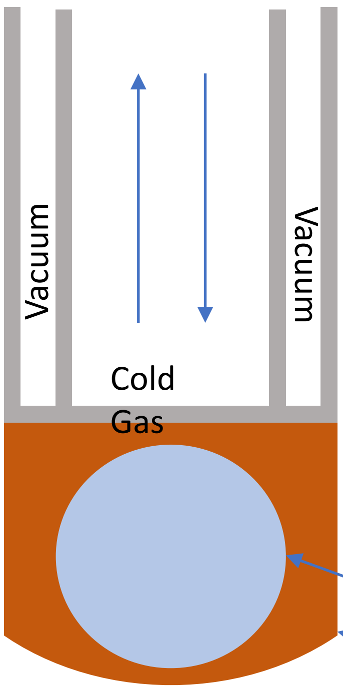
Working on implantation of Ba^+ ions into SXe on cryoprobe

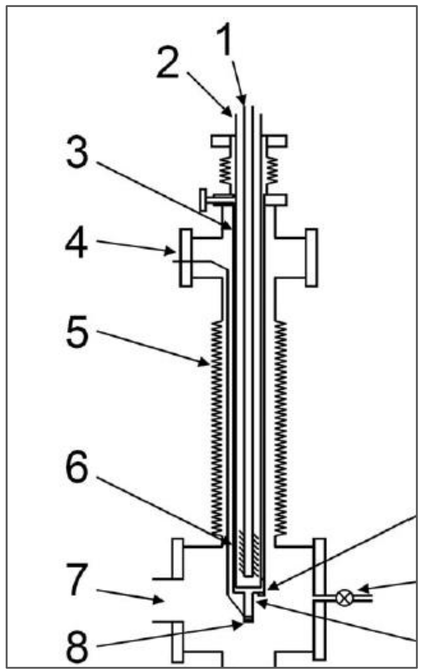


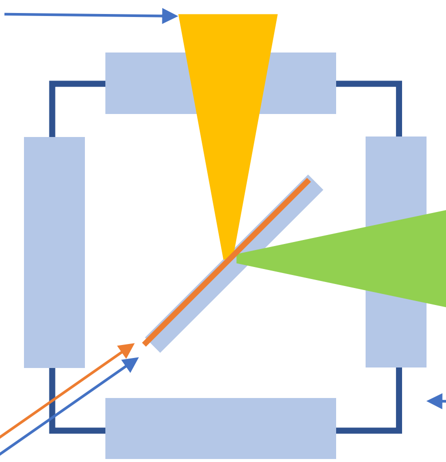
- Ba^+ ions created by laser ablation in Xe gas
- E-field to drift ions into LXe and to cryoprobe.
- Then spectroscopy of Ba in SXe in observation chamber

A better cryoprobe for single Ba imaging

Mount sapphire window on end of cryoprobe







- Probe will dip into LXe and extract barium in SXe to upper observation cube
- *Similar optics of ion beam setup*

We are working to adapt Peter Fierlinger's helium cooled probe that included capacitive measurement of SXe thickness.

P. Fierlinger et al., Rev. Sci. Instrum. 79, 045101 (2008).

Summary
First imaging of single atoms in solid rare gas, a major step for Ba tagging in nEXO

Key features:

- Single Ba atoms can be counted with $S/\sigma \approx 70$.
- *Ba deposit is "erased" by evaporating and re-freezing a new solid Xe coating.*
- *No sensitivity to any stray Ba atoms on window surface.*
- *Ba^{+} ions preferentially go into single vacancy sites – if they neutralize after they form in SV site, they will be in the site for which we have demonstrated single Ba imaging.*
  (This might occur for capture from LXe!)

nEXO Collaboration

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IME Beijing, People's Republic of China
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Indiana University
Laurentian University, Canada
Lawrence Livermore National Laboratory
University of Massachusetts
McGill University, Canada
University of North Carolina, Wilmington
Oak Ridge National Laboratory
Pacific Northwest National Laboratory
Rensselaer Polytechnic Institute
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