BETTER 0NUBB THROUGH BIOCHEMISTRY

SEARCHING FOR MAJORANA NEUTRINOS IN XENON GAS WITH SINGLE MOLECULE FLUORESCENCE IMAGING

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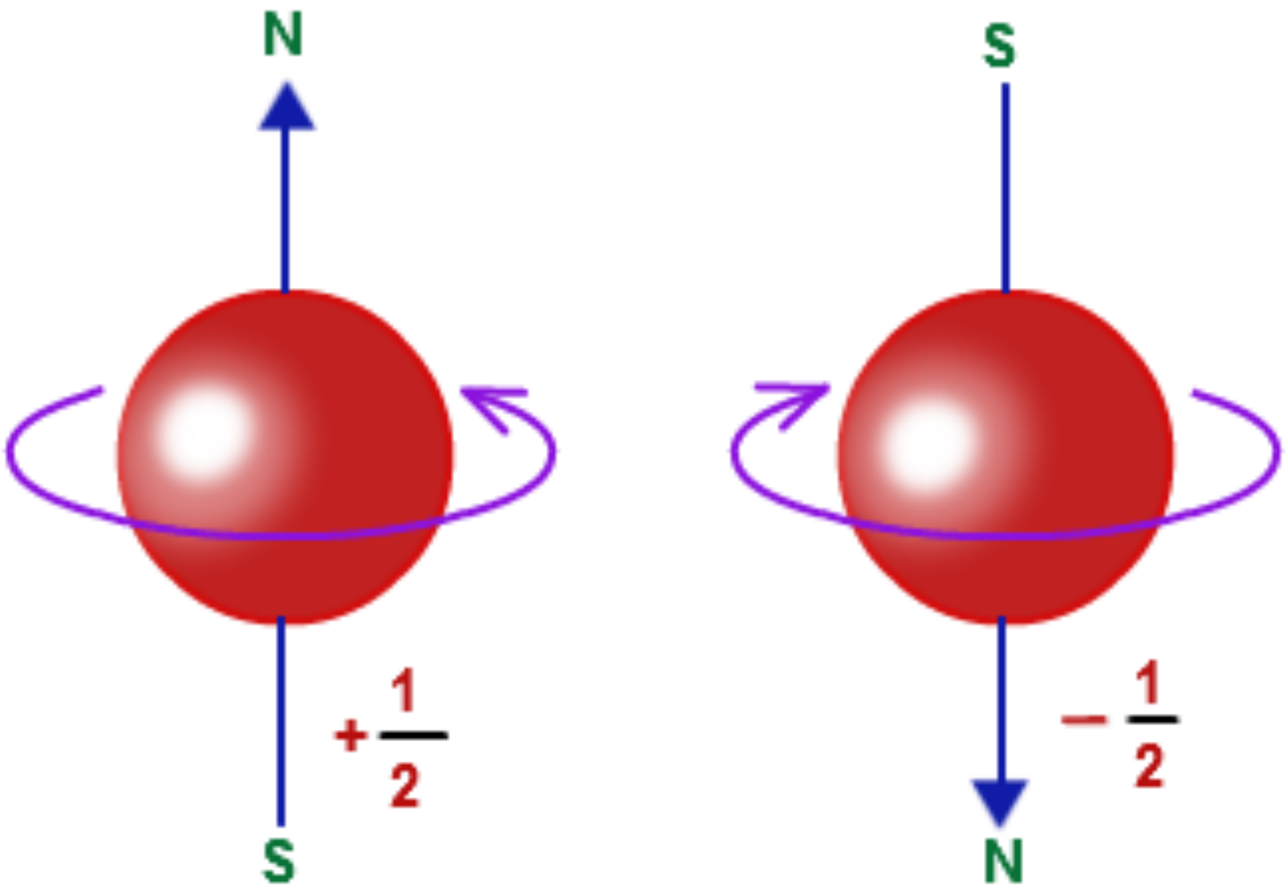
LBNL Physics Colloquium

Majorana Neutrinos in a Nutshell



Behaves like matter **Behaves like antimatter**

Robust observation of Majorana neutrinos would tell us 6 things about nature before breakfast:

1) Lepton number conservation is violated.

2) Massive fermions exist that are neither matter or antimatter but something else (Majorana fermions)

3) The SM with the Majorana term is non-renomalizible → SM is definitely a low energy effective theory.

4) There are other mass generating mechanisms in nature beyond the Higgs mechanism (though the Higgs may be involved, nonetheless).

5) Like G_F tells us the weak scale and ultimately leads to W and Z mass predictions, m_ν tells us the next scale.

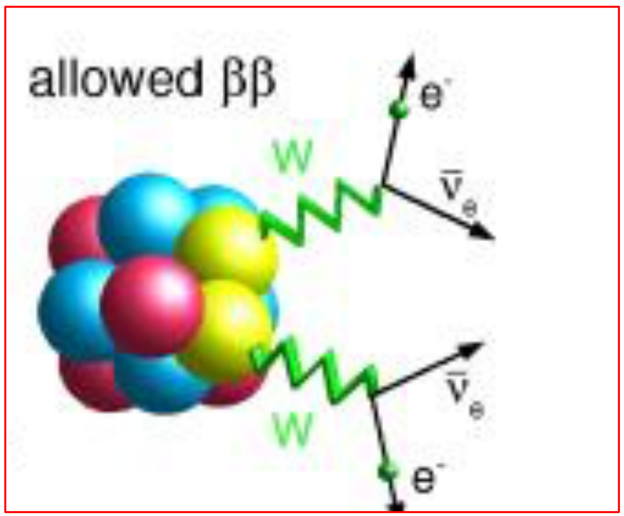
6) Majorana neutrinos are a prediction of the theory of Leptogenesis that may generate observed matter/anti-matter asymmetry of the Universe (given enough CPV – see also: DUNE)

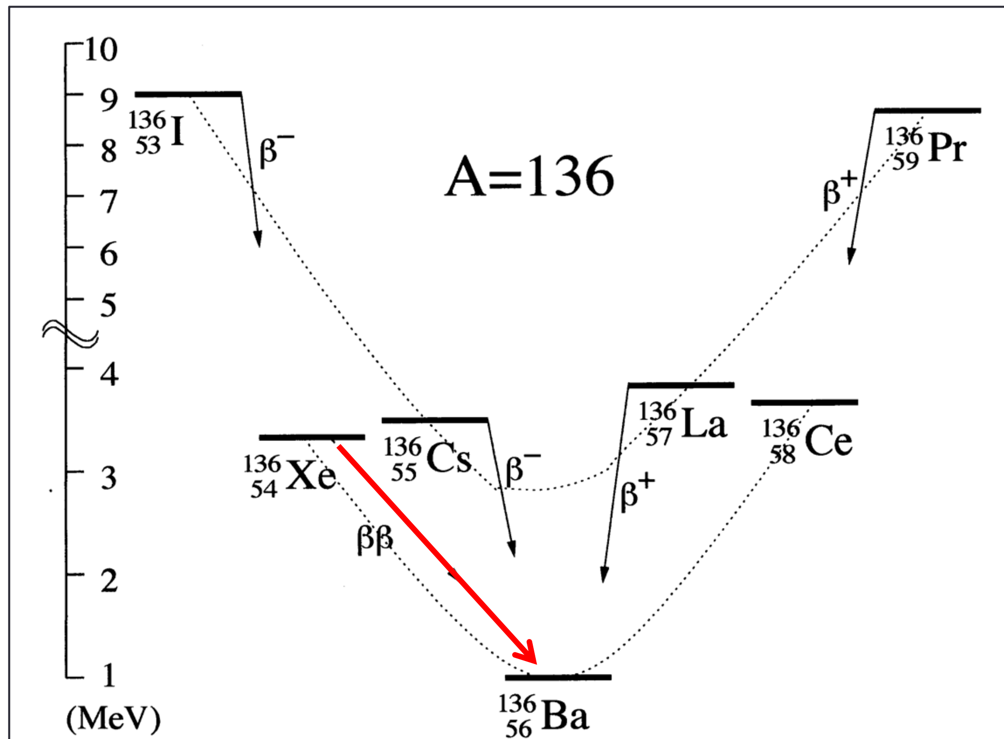
Double-Beta Decay

A rare radioactive process, energetically allowed for some even-even nuclei where $m_{Z+1} > m_Z > m_{Z+2}$

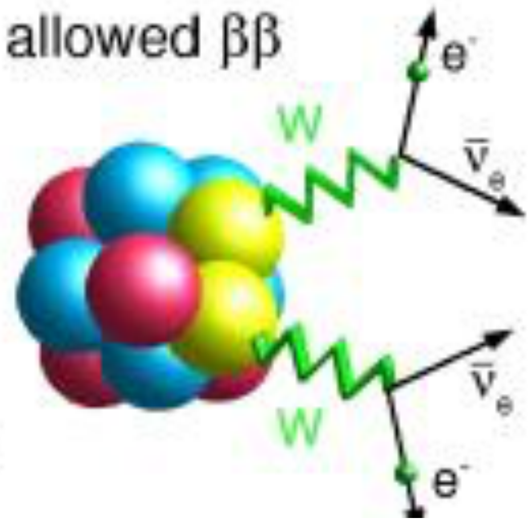
- $(Z,A) \rightarrow (Z+2,A) + e^-_1 + \underline{\nu}_1 + e^-_2 + \underline{\nu}_2$

Discovered in 1987 in ^{82}Se and now seen in multiple isotopes





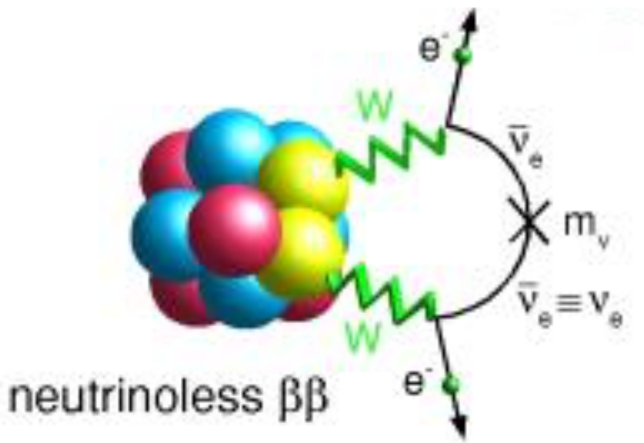
Types of double beta decay



A known standard model process and an important calibration tool

$$T_{\frac{1}{2}} \approx 10^{19-21}\, yrs.$$

Final state: $e^- e^- \nu_e \nu_e$

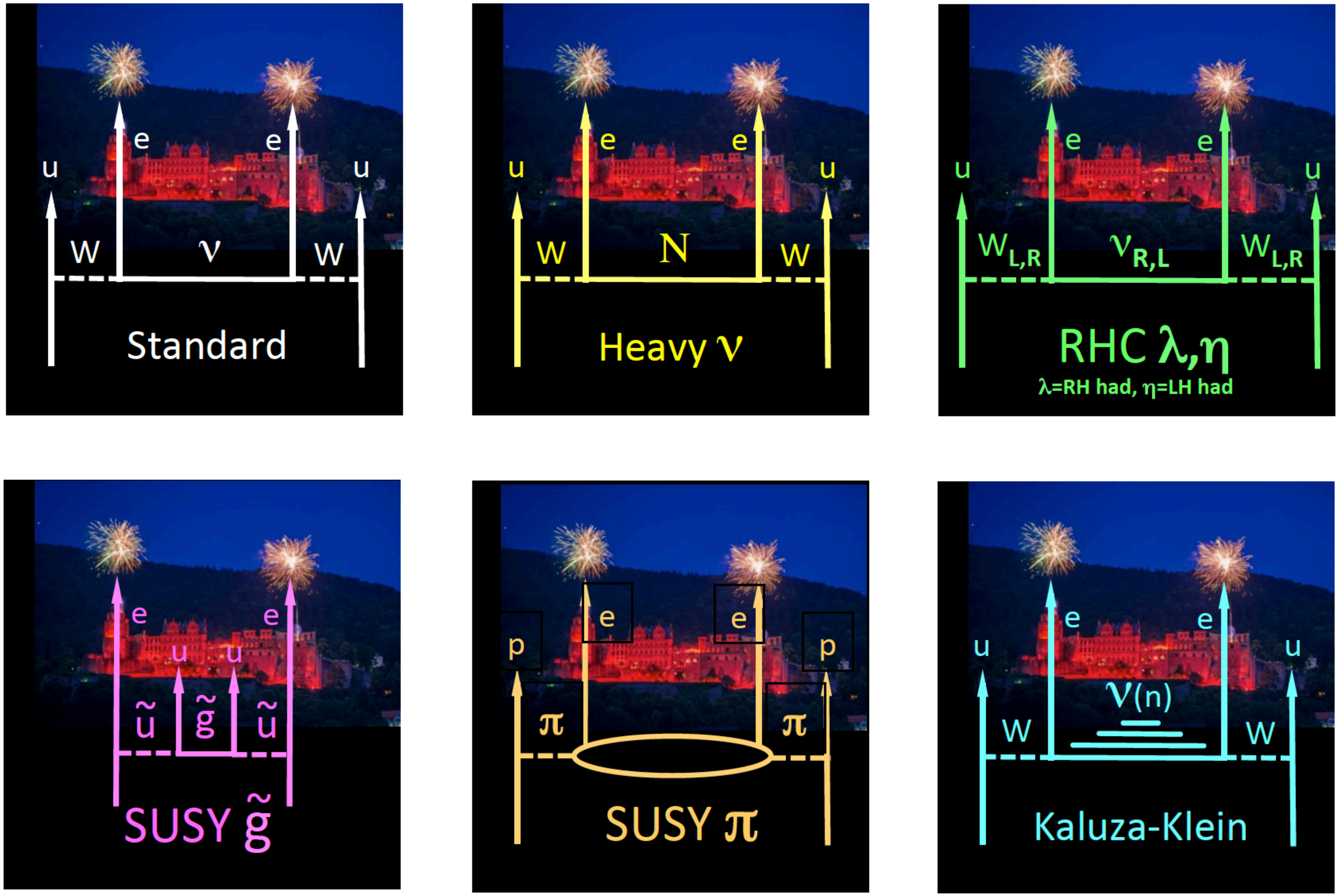


Observation would prove that the neutrino is a Majorana fermion

$$\frac{1}{T_{\frac{1}{2}}} = G \times \|\mathbf{M}\|^2 \times m_{\bar{\nu}}{}^2$$

Final state: $e^- e^-$

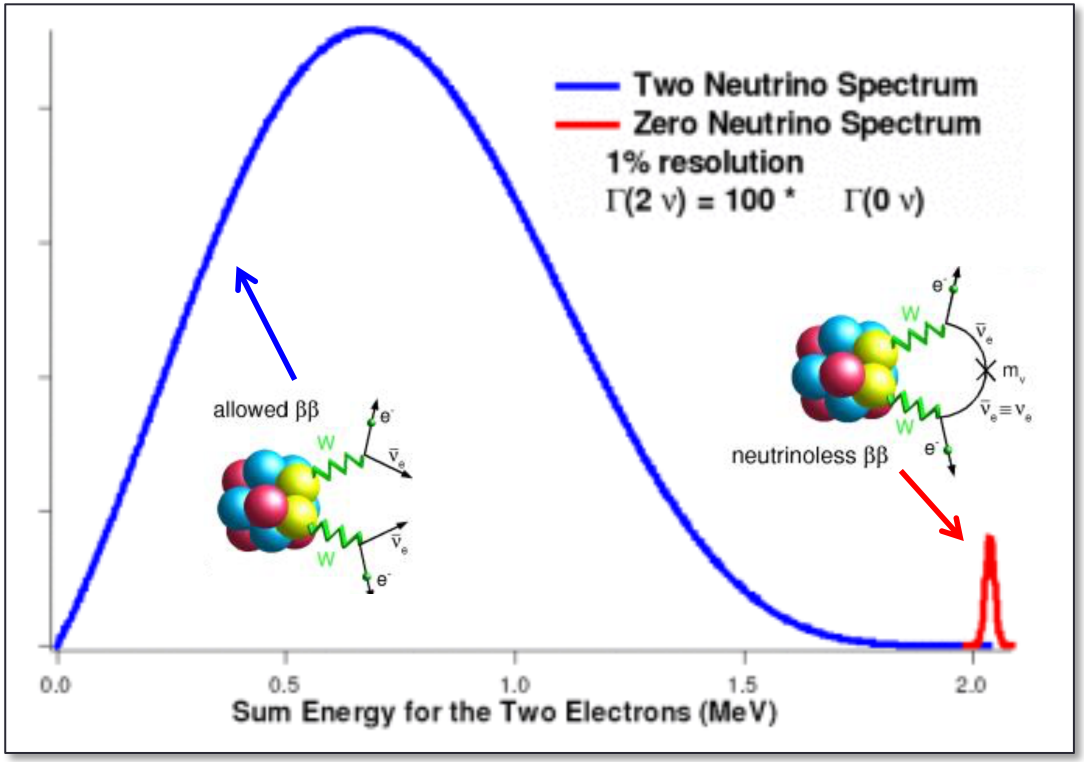
Warning: **don't stick to $m_{\beta\beta}$ metric, just go on with $T_{1/2}$!** Variety of $0\nu\beta\beta$ mechanisms:



$0\nu\beta\beta$ from any mechanism → **Majorana nature of ν would be established anyway**

Slide from The Mid and Long Term Future of Neutrinoless Double Beta Decay, Andrea Giuliani, Neutrinno2018, https://doi.org/10.5281/zenodo.1286915

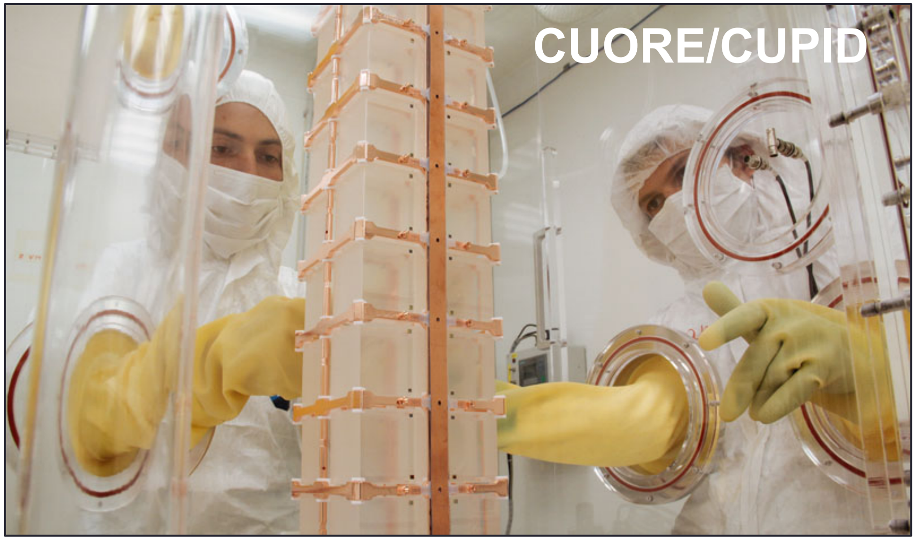
The ideal experiment:



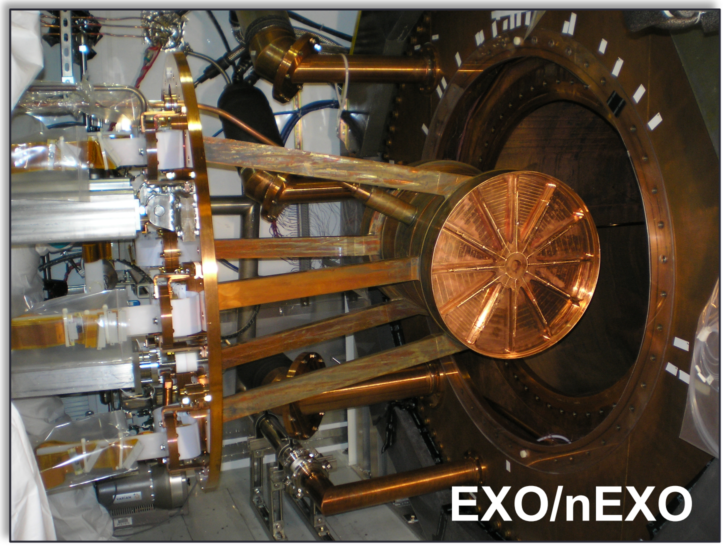
- ***MANDATORY:***
  - Resolution better than ~2% FWHM to fully reject two-neutrino mode

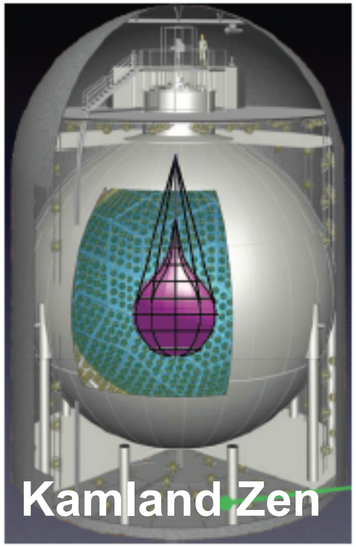
- ***Then just watch and wait…***

Neutrinoless Double Beta Decay Searches



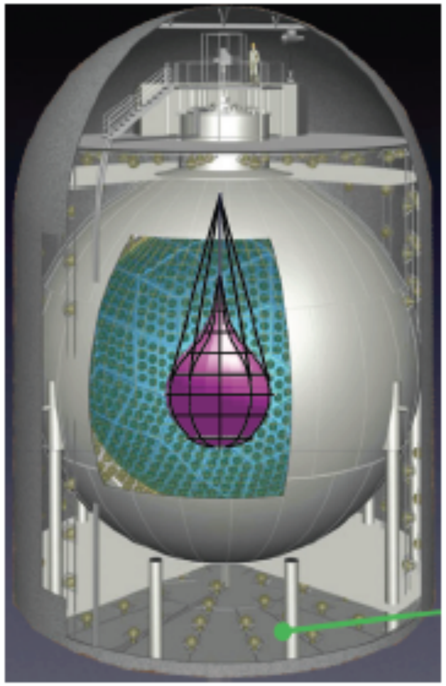
Many completely different technologies…

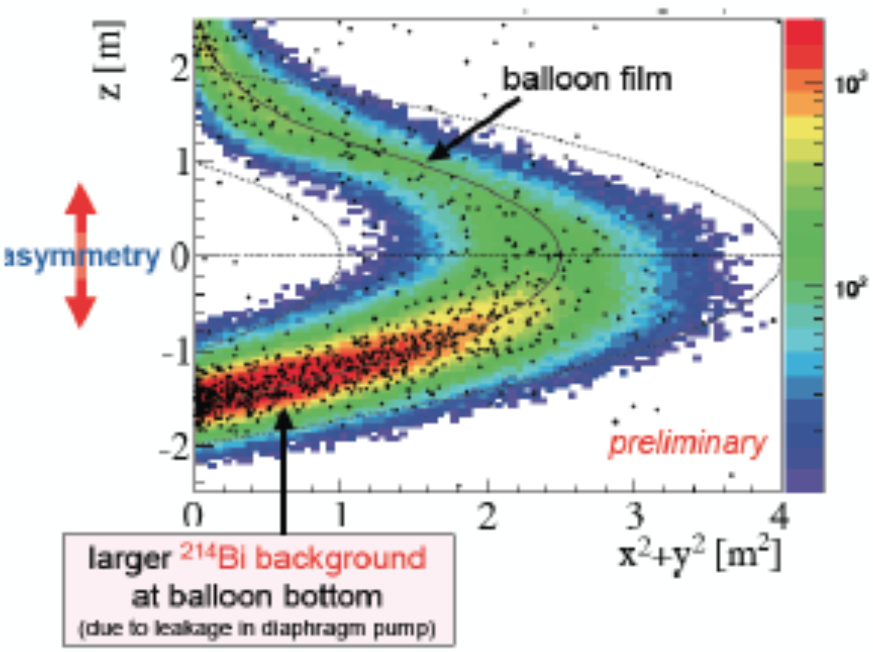




KAMLAND-ZEN

- **Present World Leader.**
- Liquid scintillator with dissolved ^{136}Xe
  - Kamland-Zen Phase1 : 179 kg ^{136}Xe
  - Kamland-Zen Phase2 : 383 kg ^{136}Xe
  - Kamland-Zen2 : 800 kg ^{136}Xe





VITAL STATISTICS	*NSAC goals*
Energy resolution: 17% FWHM	*<1%*
Background index: *Initial : 210* *Reduced by ~2 after filtration*	*<1*
Present 0νββ Limit: 1.07×10^{26} yrs	*>n*10²⁷*

KAM

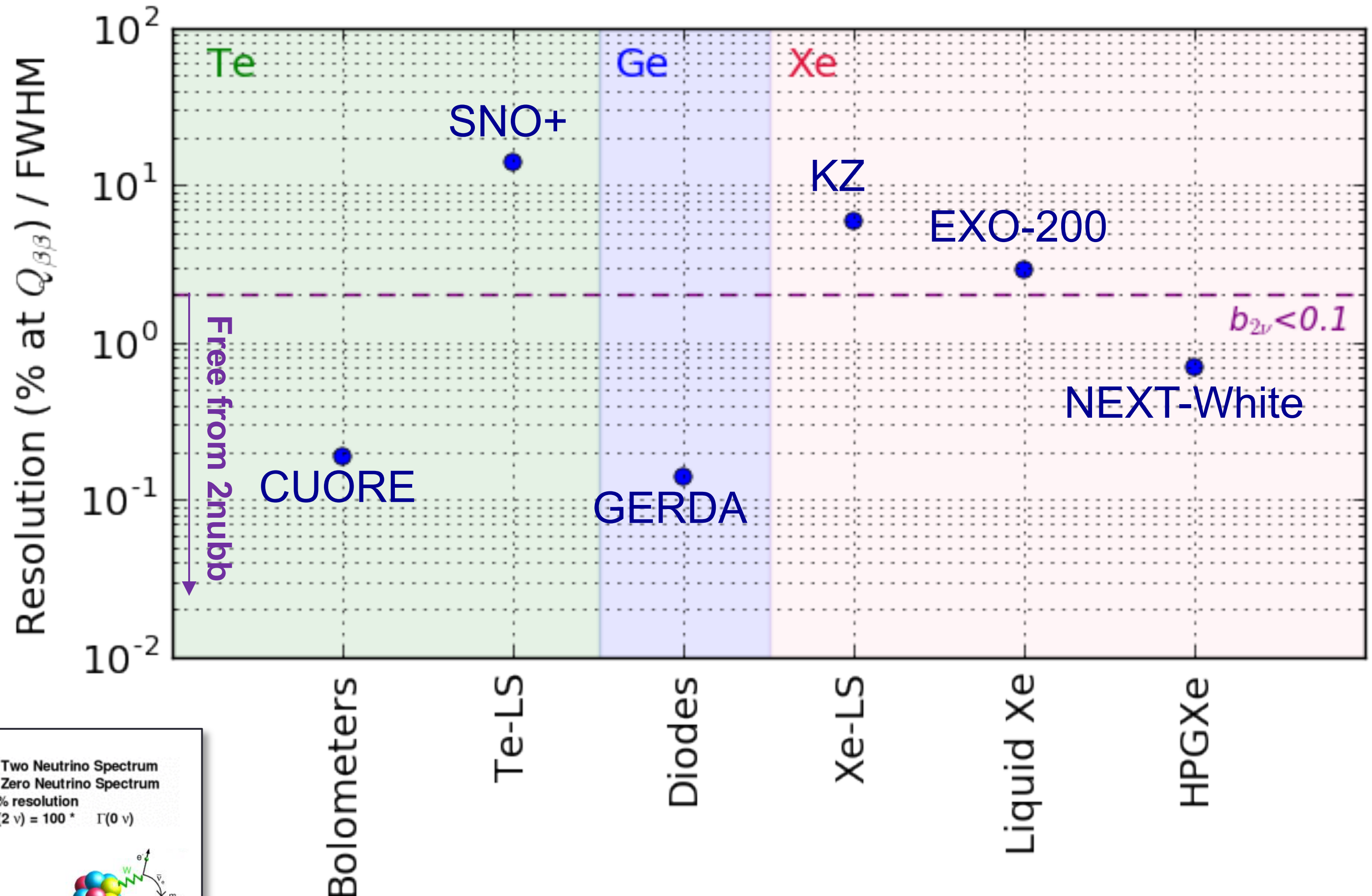
- **Preser**
- Liquid
  - Kar
  - Kar
  - Kar

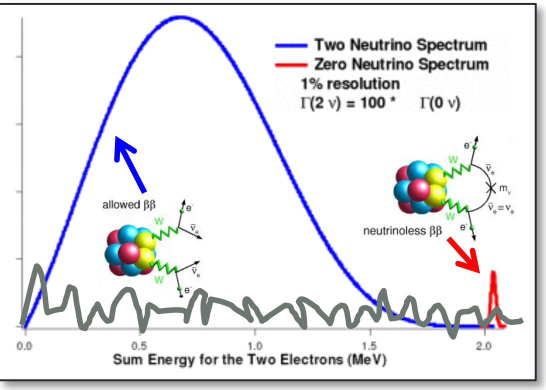
	Period-1 (270.7 days)		Period-2 (263.8 days)	
Observed events	22		11	
Background	Estimated	Best-fit	Estimated	Best-fit
^{136}Xe $2\nu\beta\beta$	-	5.48	-	5.29
Residual radioactivity in Xe-LS				
^{214}Bi (^{238}U series)	0.23 ± 0.04	0.25	0.028 ± 0.005	0.03
^{208}Tl (^{232}Th series)	-	0.001	-	0.001
^{110m}Ag	-	8.5	-	0.0
External (Radioactivity in IB)				
^{214}Bi (^{238}U series)	-	2.56	-	2.45
^{208}Tl (^{232}Th series)	-	0.02	-	0.03
^{110m}Ag	-	0.003	-	0.002
Spallation products				
^{10}C	2.7 ± 0.7	3.3	2.6 ± 0.7	2.8
^{6}He	0.07 ± 0.18	0.08	0.07 ± 0.18	0.08
^{12}B	0.15 ± 0.04	0.16	0.14 ± 0.04	0.15
^{137}Xe	0.5 ± 0.2	0.5	0.5 ± 0.2	0.4

Note: Significant 2nubb background due to limited energy resolution

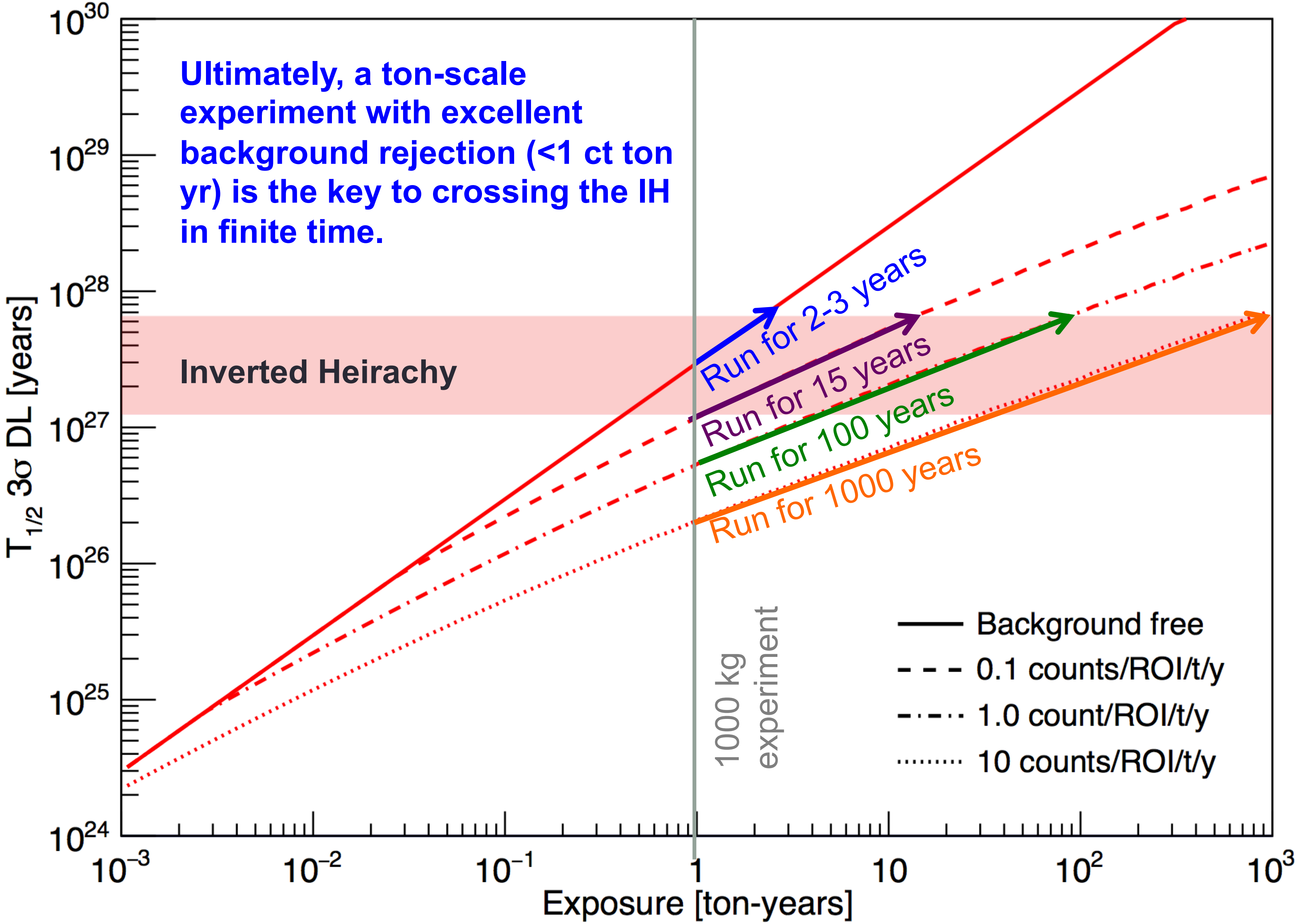
NSAC goals
<1%
<1
$>n*10^{27}$

Measuring Energy



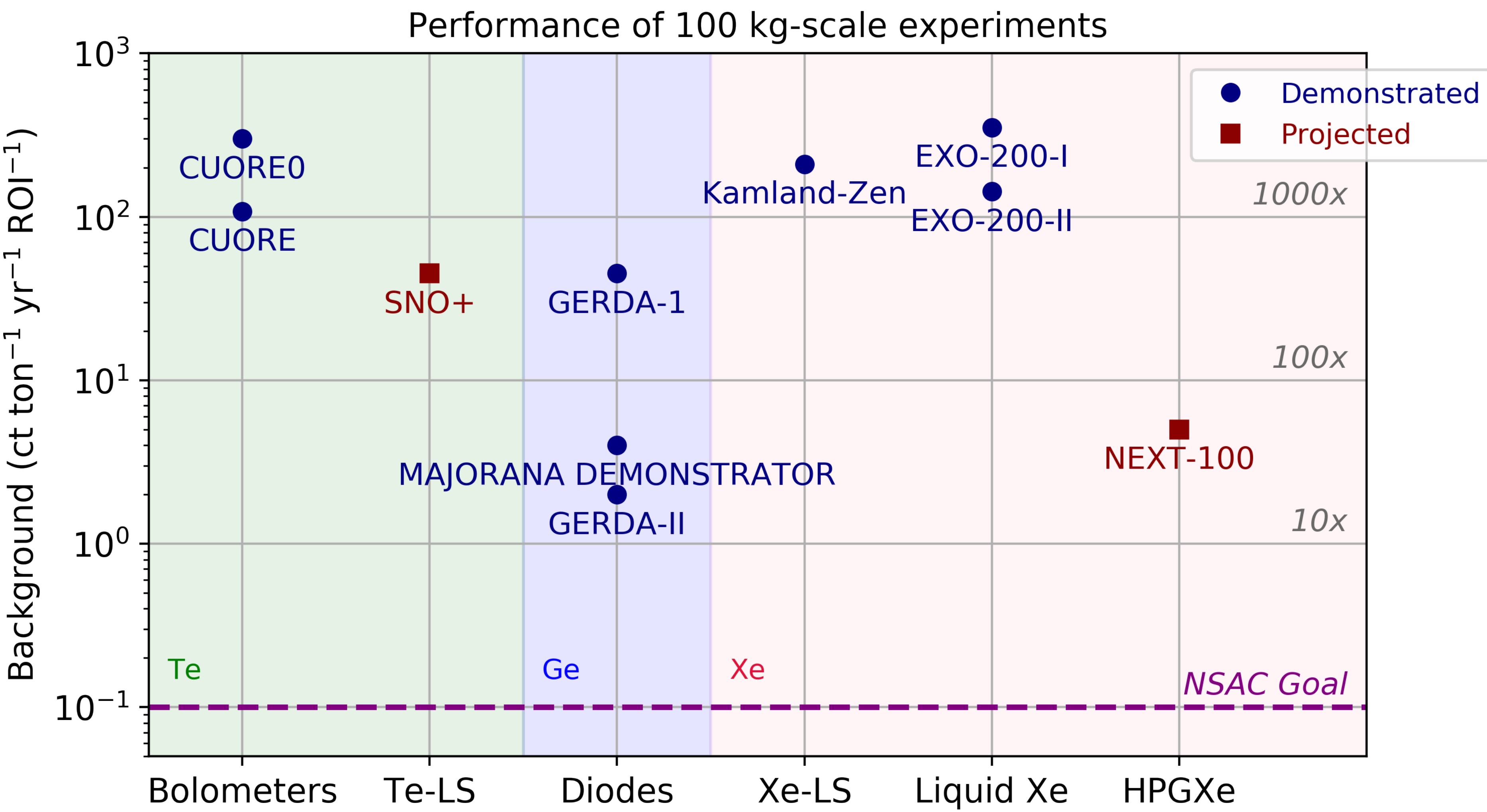


Once free from 2nubb, radioactive and cosmogenic backgrounds become dominant

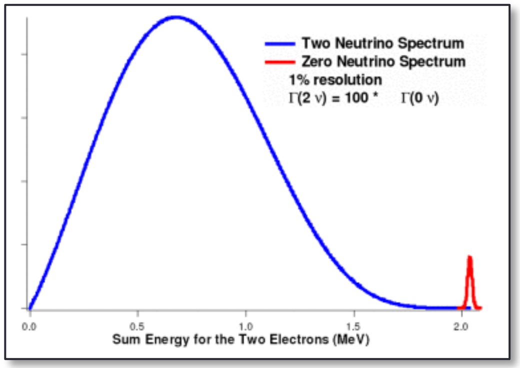


Ultimately, a ton-scale experiment with excellent background rejection (<1 ct ton yr) is the key to crossing the IH in finite time.
Inverted Heirachy
Run for 2-3 years
Run for 15 years
Run for 100 years
Run for 1000 years
1000 kg experiment
Background free
0.1 counts/ROI/t/y
1.0 count/ROI/t/y
10 counts/ROI/t/y
$T_{1/2}$ 3σ DL [years]
Exposure [ton-years]
10^{30}
10^{29}
10^{28}
10^{27}
10^{26}
10^{25}
10^{24}
10^{-3}
10^{-2}
10^{-1}
1
10
10^{2}
10^{3}

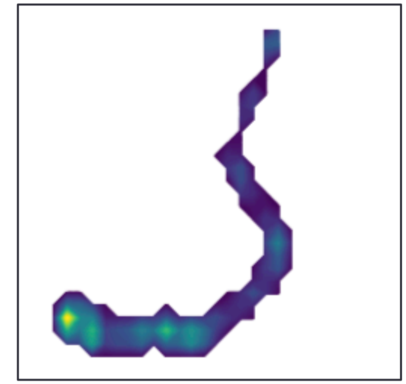
100kg-class experiments:

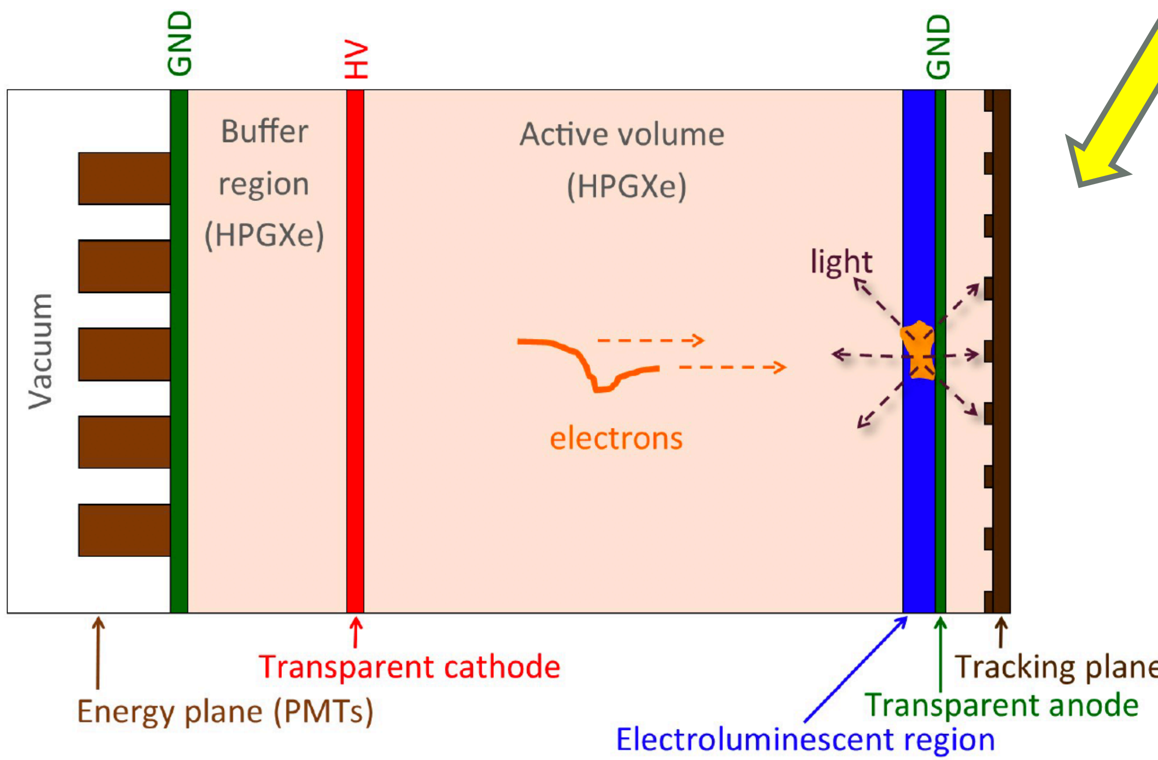


Measure energy this end



Measure topology this end





The NEXT Program

- **Sequence of HPGXe TPCs, focused on achieving big, very low background xenon 0νββ detector**

→ NEXT-DBDM

(Berkeley, US)

→ NEXT-DEMO

(Valencia, Spain)

→ **NEXT-White**

(Canfranc, Spain)

→ NEXT-100

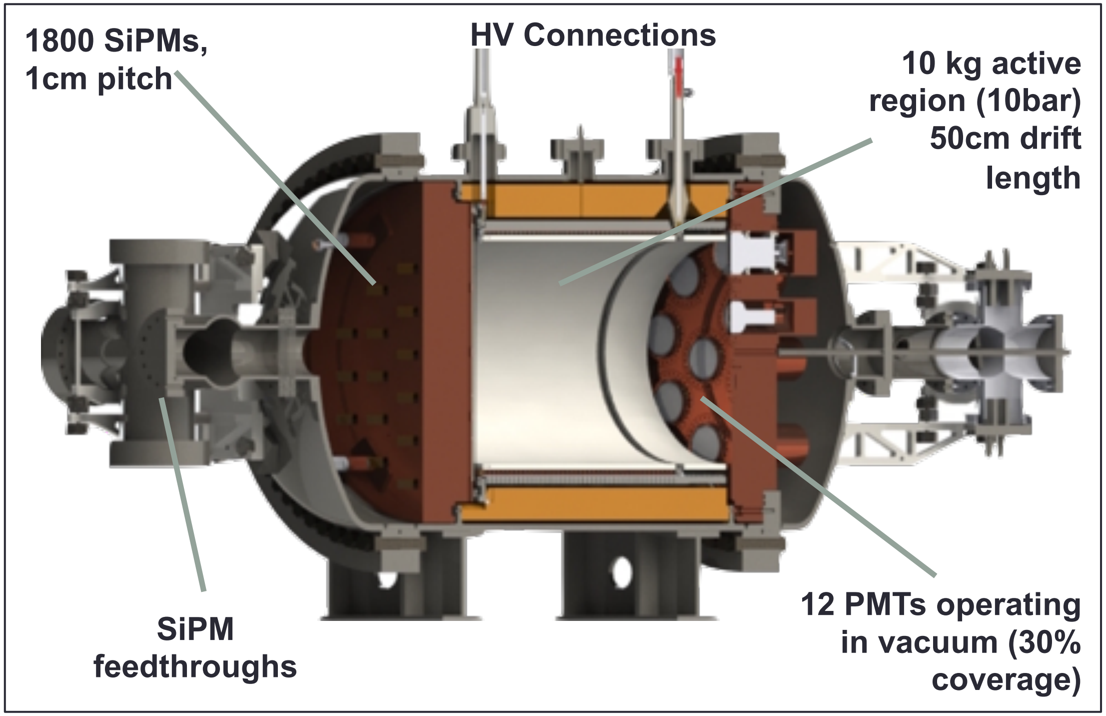
(Canfranc, Spain)

→ NEXT-HD

(~2023)

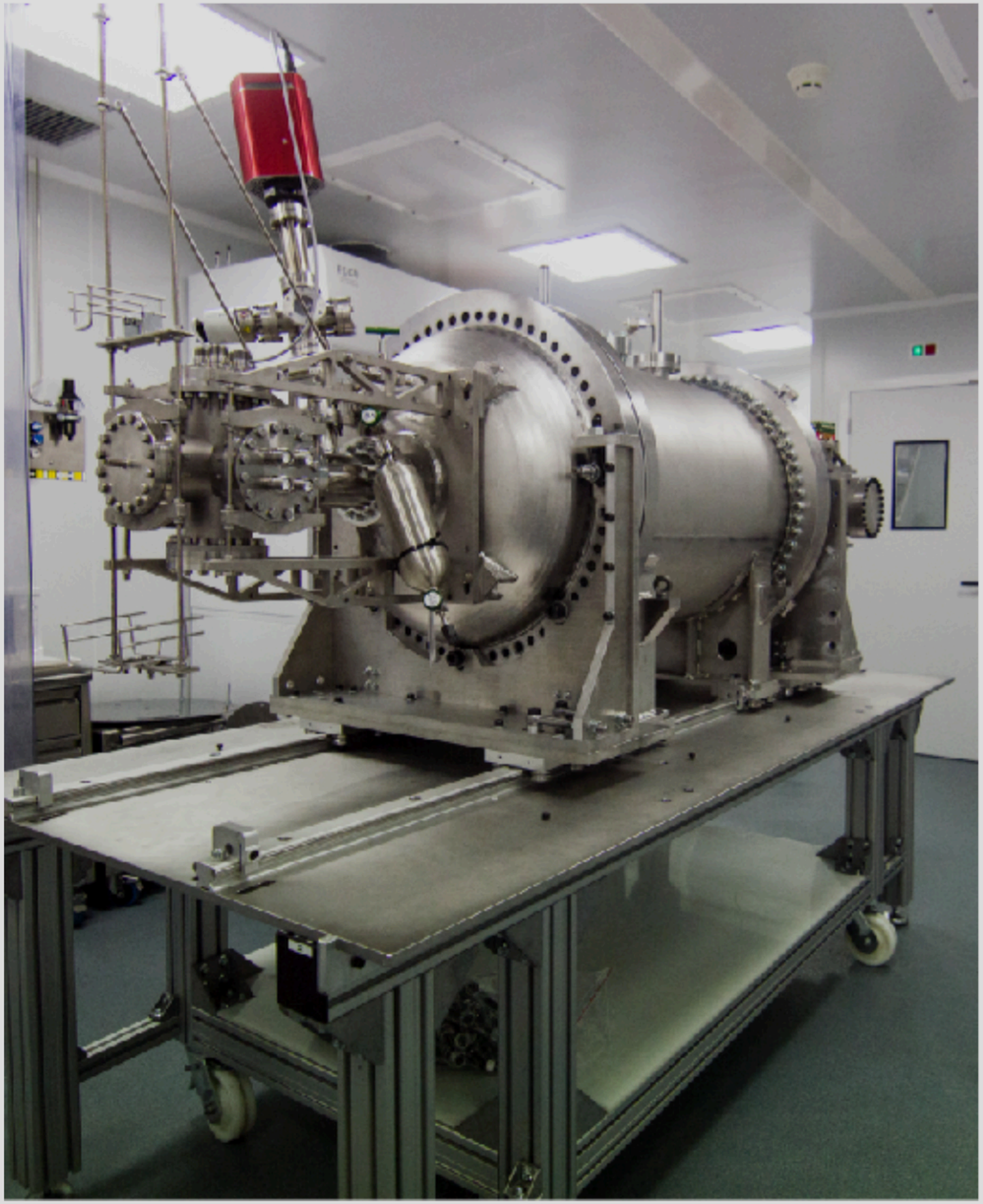
→ *NEXT-BOLD*

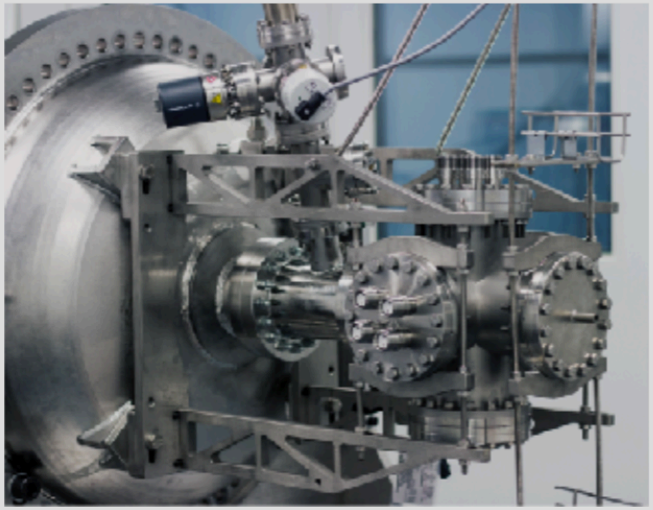
(~2026)



NEXT-White operating now

Full underground technology demonstrator @10kg scale

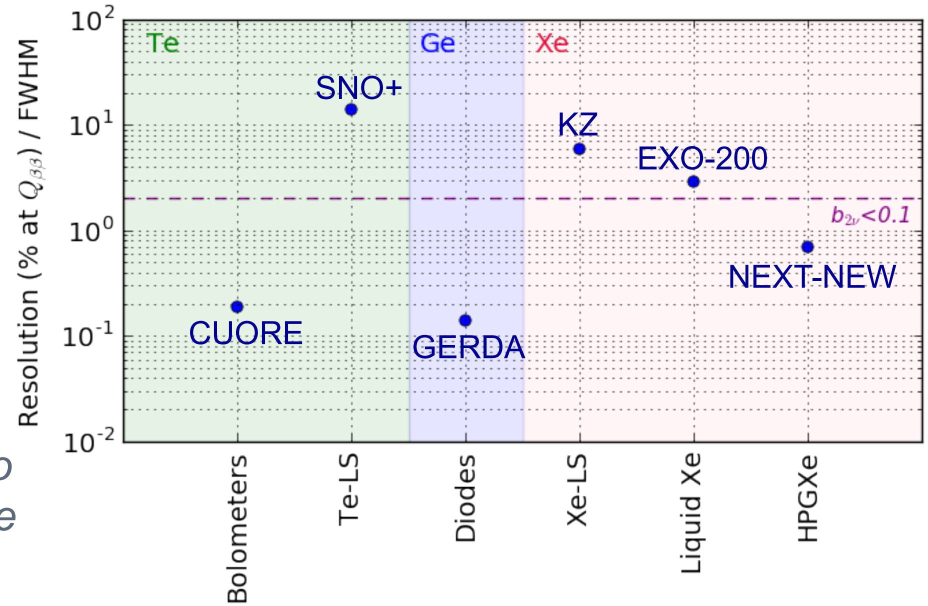




Why Xenon Gas?

- 1) Energy resolution
- 2) Topology
- 3) Practicality at large scale

Why Xenon Gas?

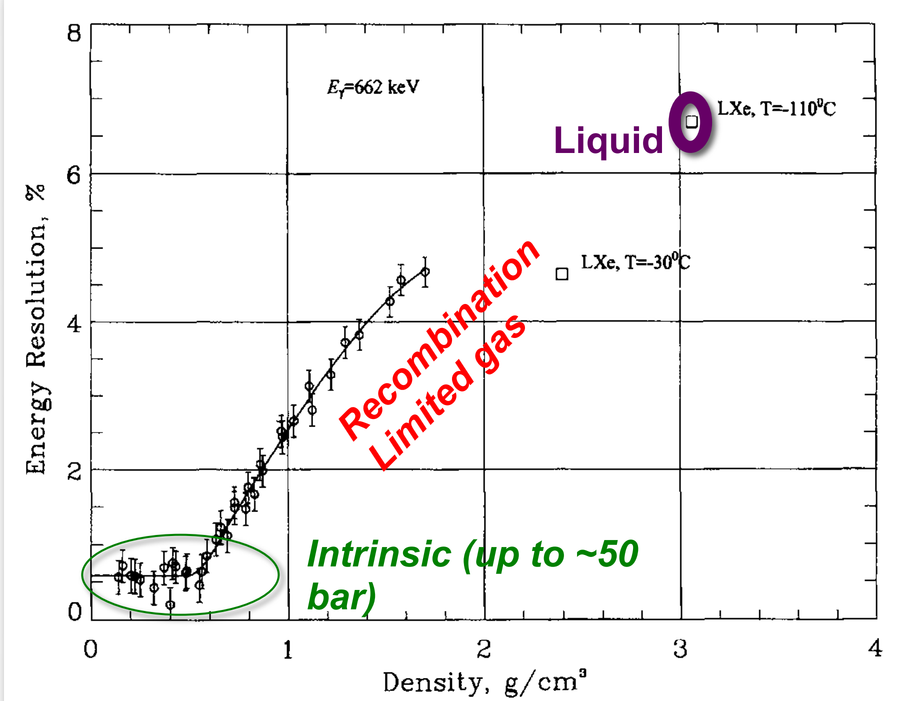


- **1) Energy resolution**

  Fluctuation-less EL gain and low Fano factor produces resolution comparable with solid-state technologies in a monolithic TPC experiment

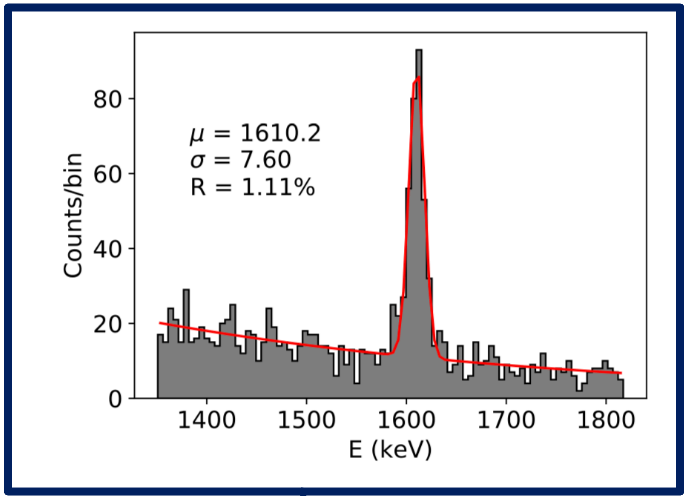
- 2) Topology

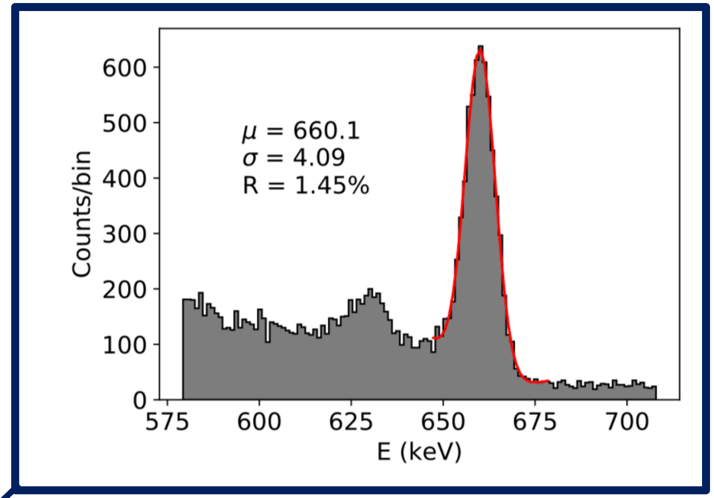
- 3) Practicality at large scale



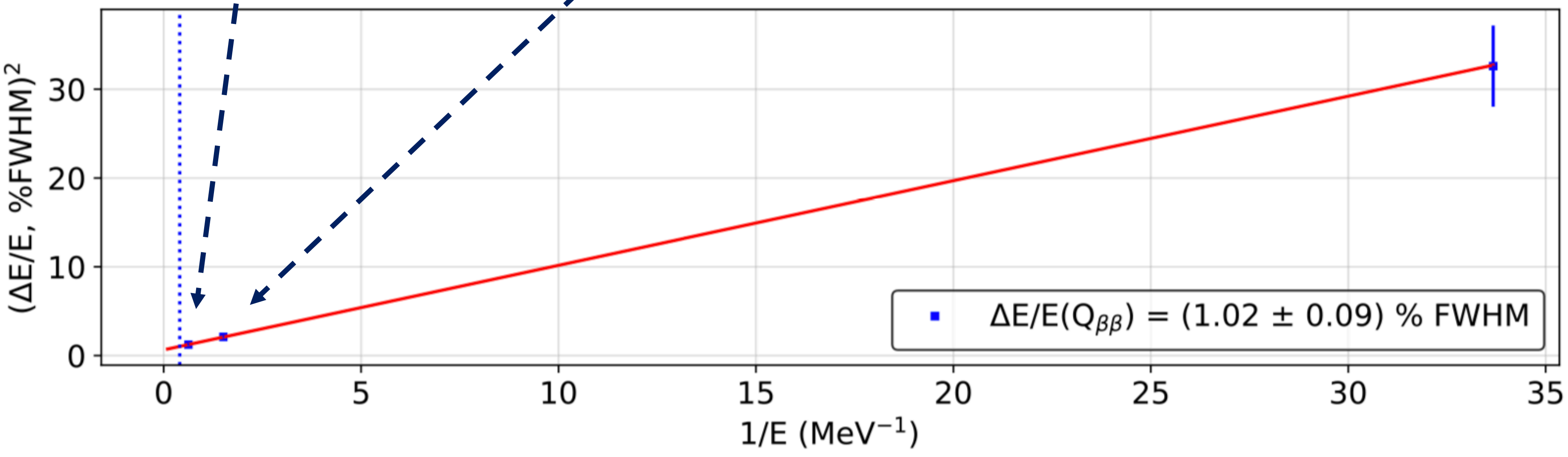
Bolotnikov and Ramsey. "The spectroscopic properties of high-pressure xenon."NIM *A* 396.3 (1997): 360-370

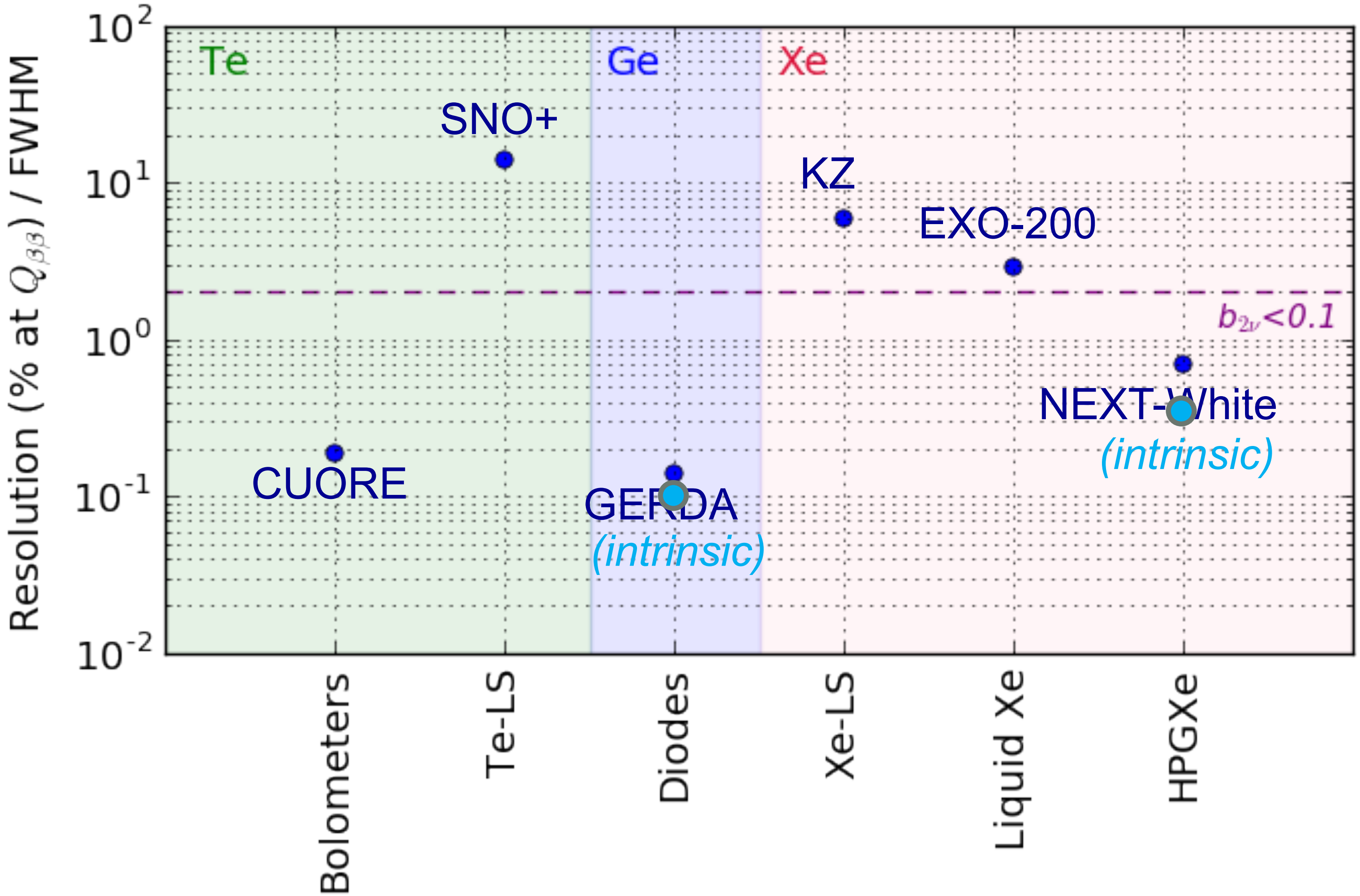
Initial results on energy resolution of the NEXT-White detector
J. Renner et al, arXiv 1808:01804
Energy calibration of the NEXT-White detector with 1% FWHM resolution near Qββ of 136Xe
J Renner et al, arXiv:1905.13110





Energy calibrations and stability still improving: presently sit at ~1% at Q_{bb}





- Still some room for improvement in energy resolution – factor ~2-3 from intrinsic
- Continuing improvements in analysis and understanding detector effects reap continuing rewards.

Why Xenon Gas?

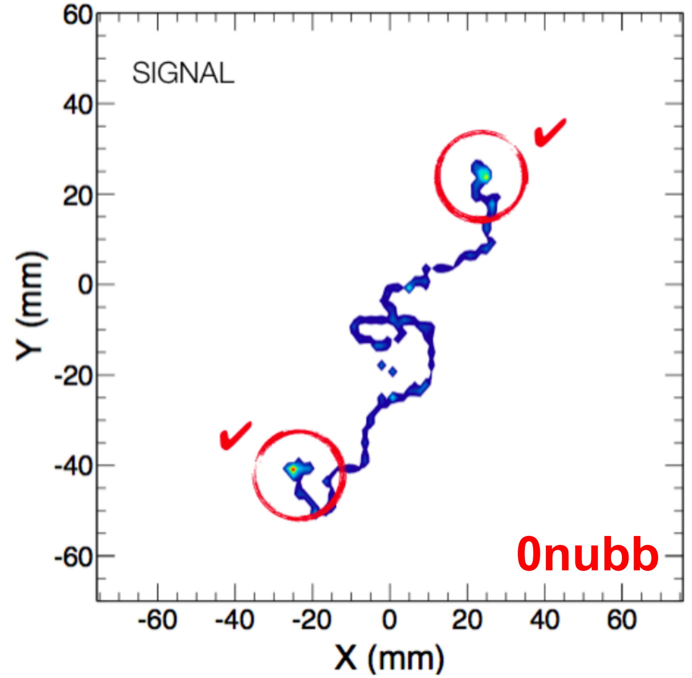
- 1) Energy resolution

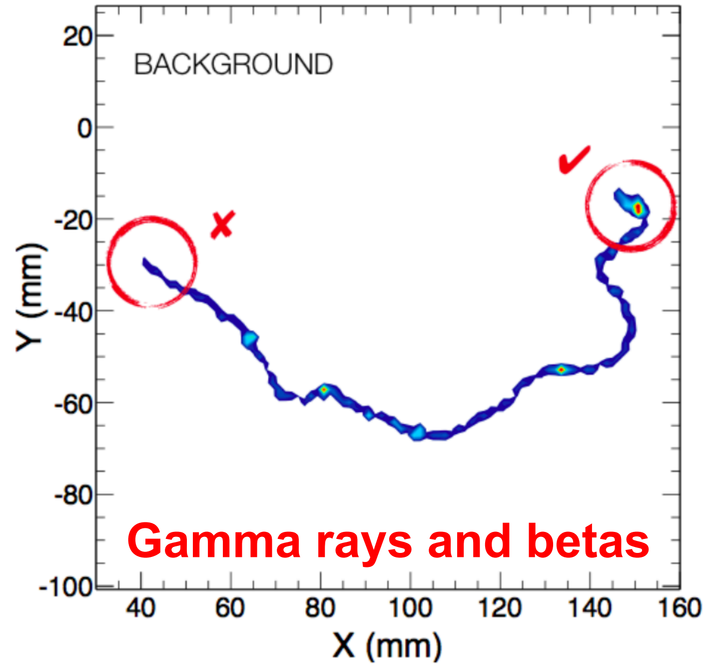
  Fluctuation-less EL gain produces resolution comparable with solid-state technologies in a monolithic TPC experiment

- **2) Topology**

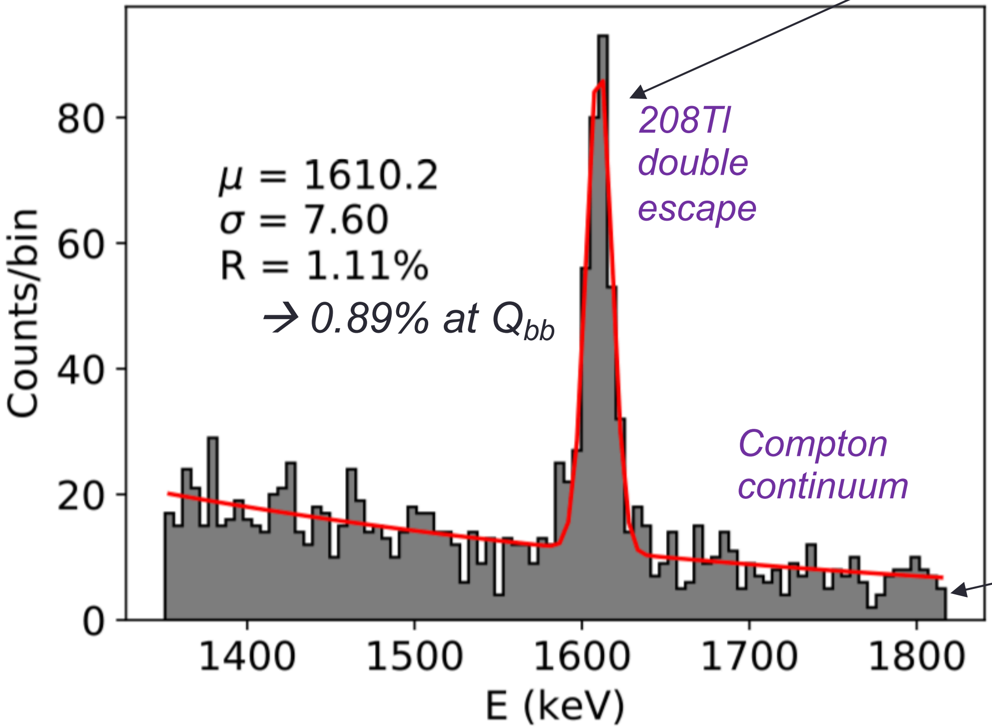
  Lower density allows powerful single-vs-multi electron and single-vs-multi-site topological background rejection

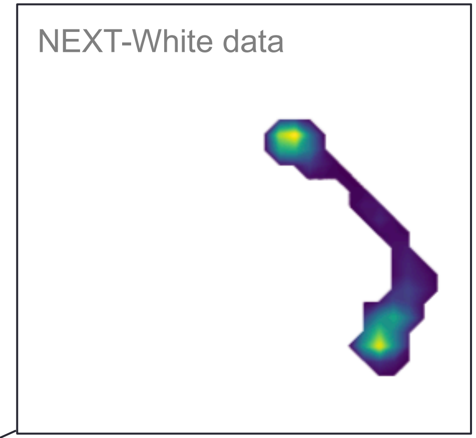
- 3) Practicality at large scale



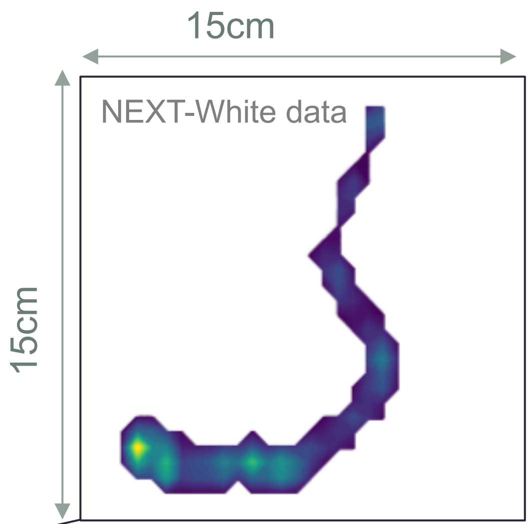


Topological Reco with Double Escape Peaks



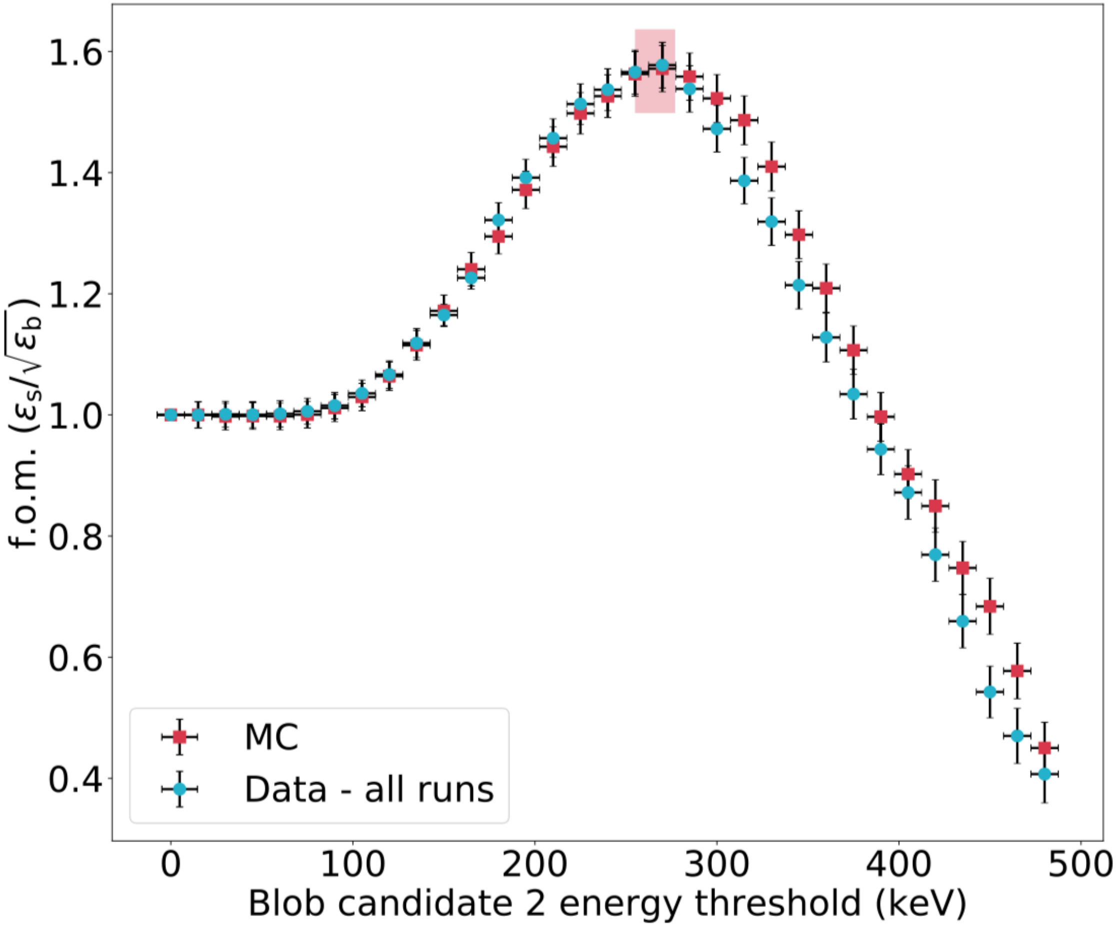


On double escape peak



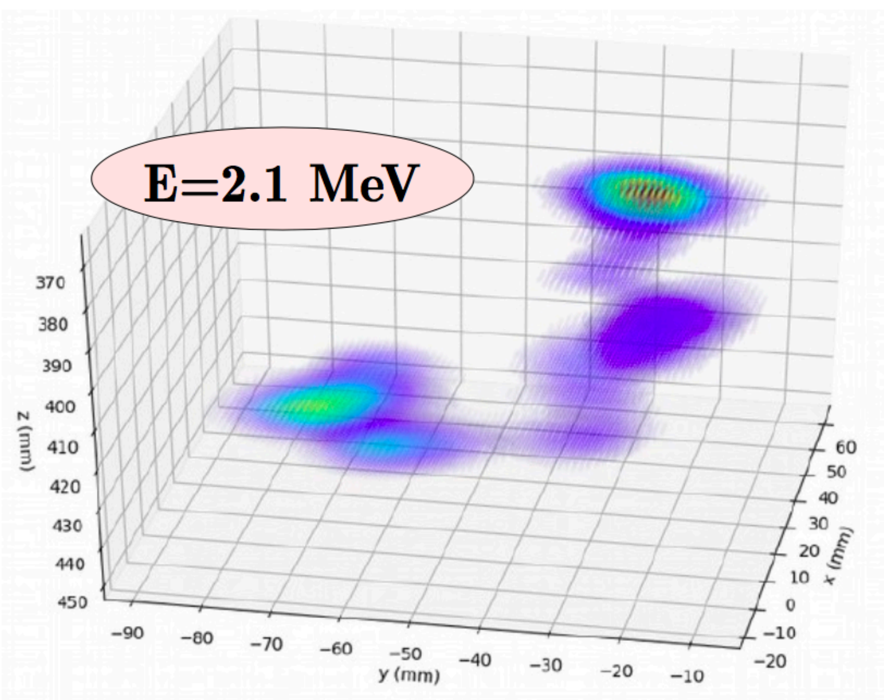
Off double escape peak

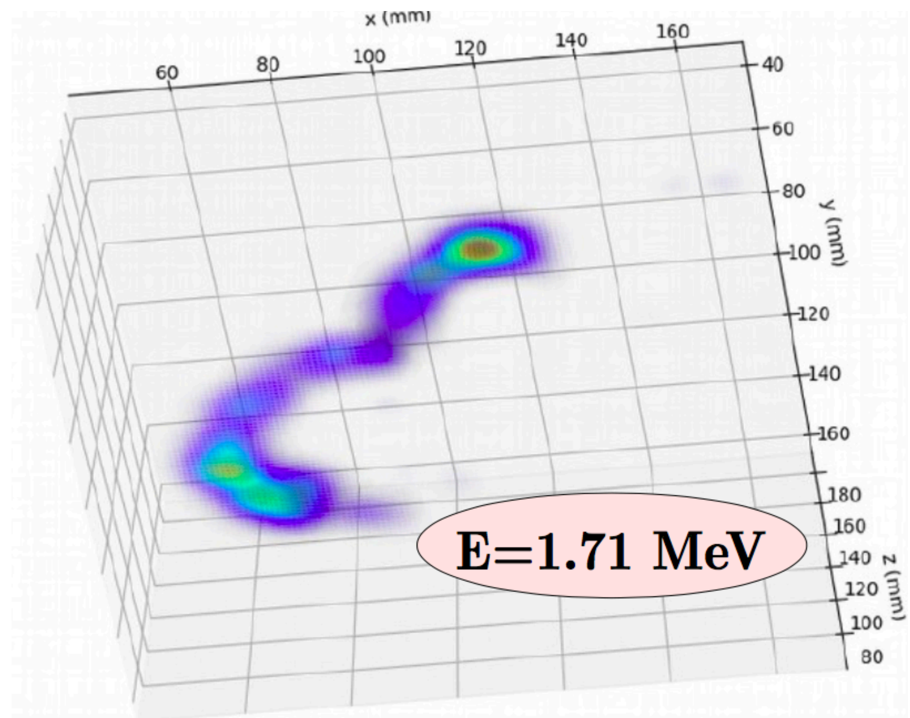
Data / MC agreement on topological signature



[arXiv:1905.13141](https://arxiv.org/abs/1905.13141)
Efficiency of the 2-electron topological signature in the NEXT-White detector

Two-neutrino double beta decay candidates





NEXT-White data

Topologically identified and away from double escape peaks

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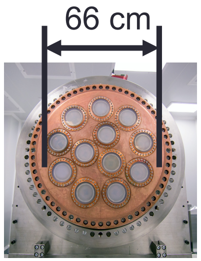
- 2) Topology

  Lower density allows powerful single-vs-multi electron and single-vs-multi-site topological background rejection

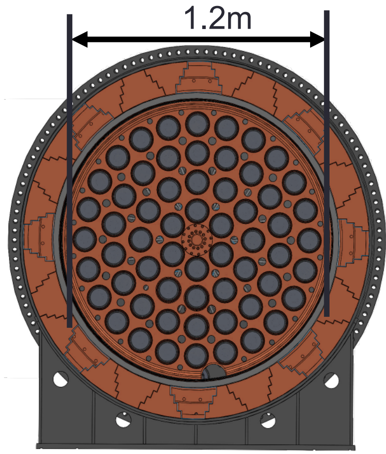
- **3) Practicality at large scale**

  Reliance on active background rejection rather than self shielding means program uses isotope efficiently and can be phased

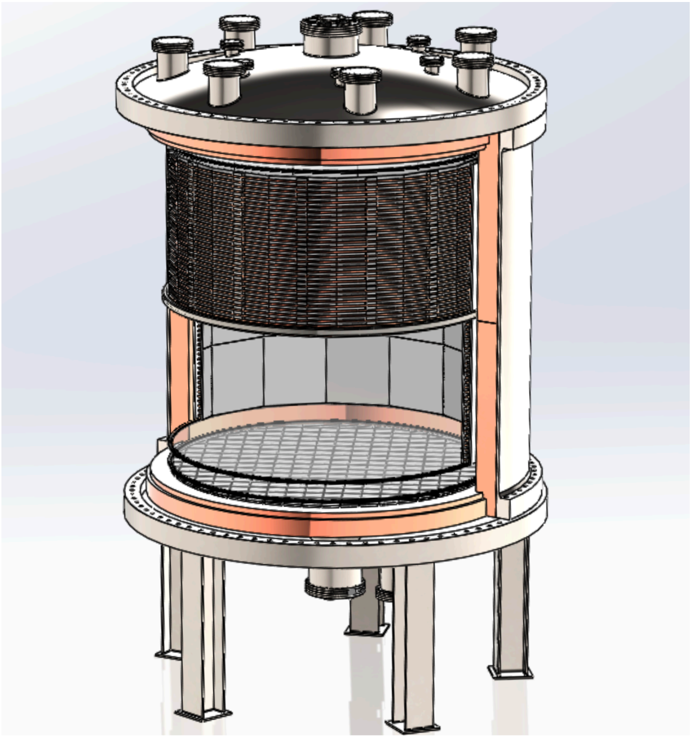
NEXT-NEW
Running



NEXT-100
2019



NEXT-HD: *2023*



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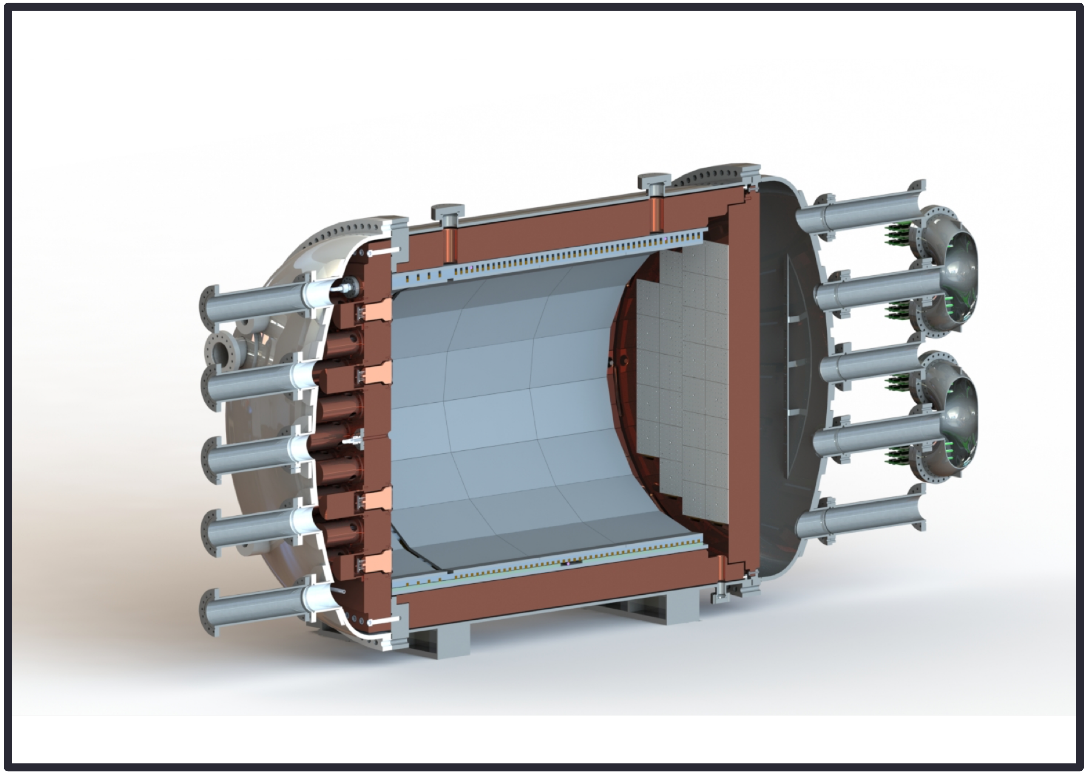
(~2023)

→ *NEXT-BOLD*

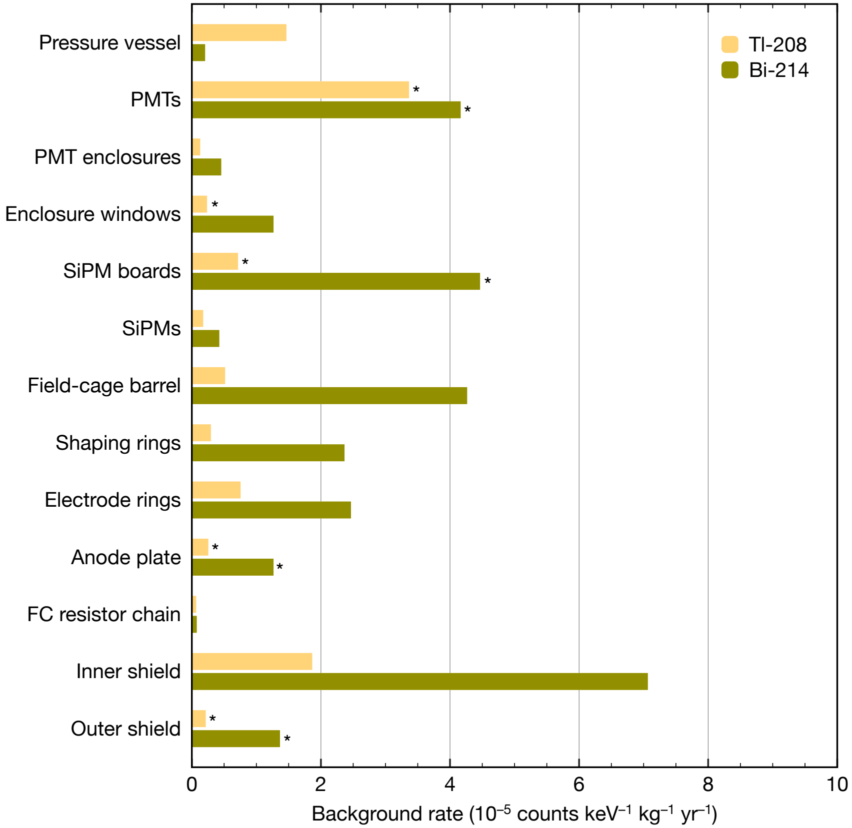
(~2026?)

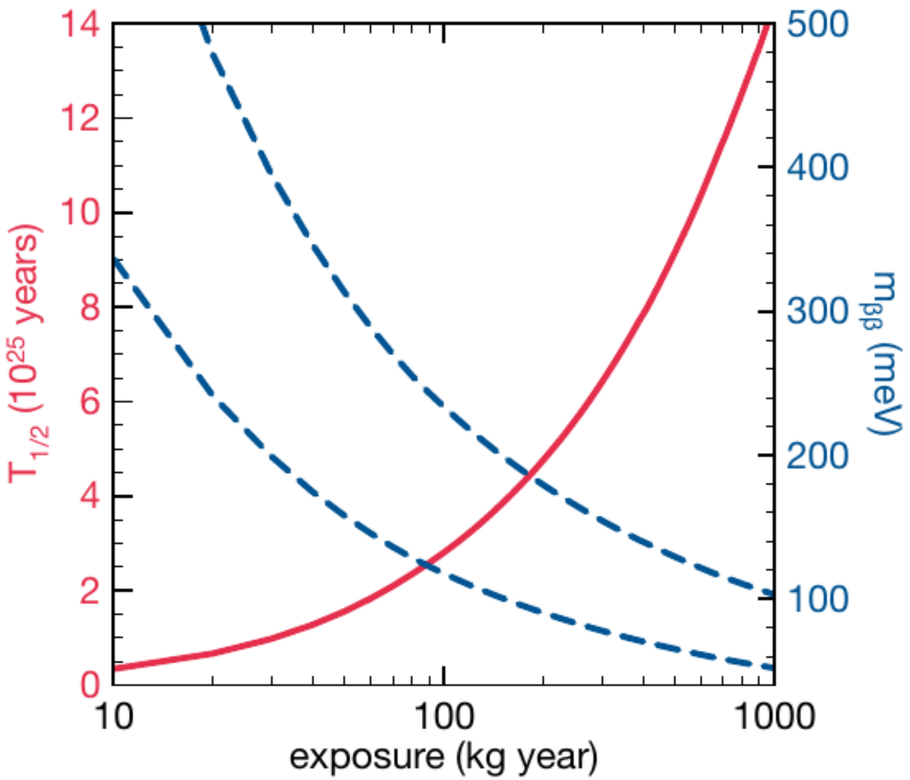


100 kg scale neutrinoless double beta decay search and background-study for ton-scale

NEXT-100 Sensitivity

- Projected near-background-free performance at 100kg scale - Total BG: 5x10-4 c/keV/kg/y, validated with NEXT-White.
- Presently under construction for operation in 2020.





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(~2023)

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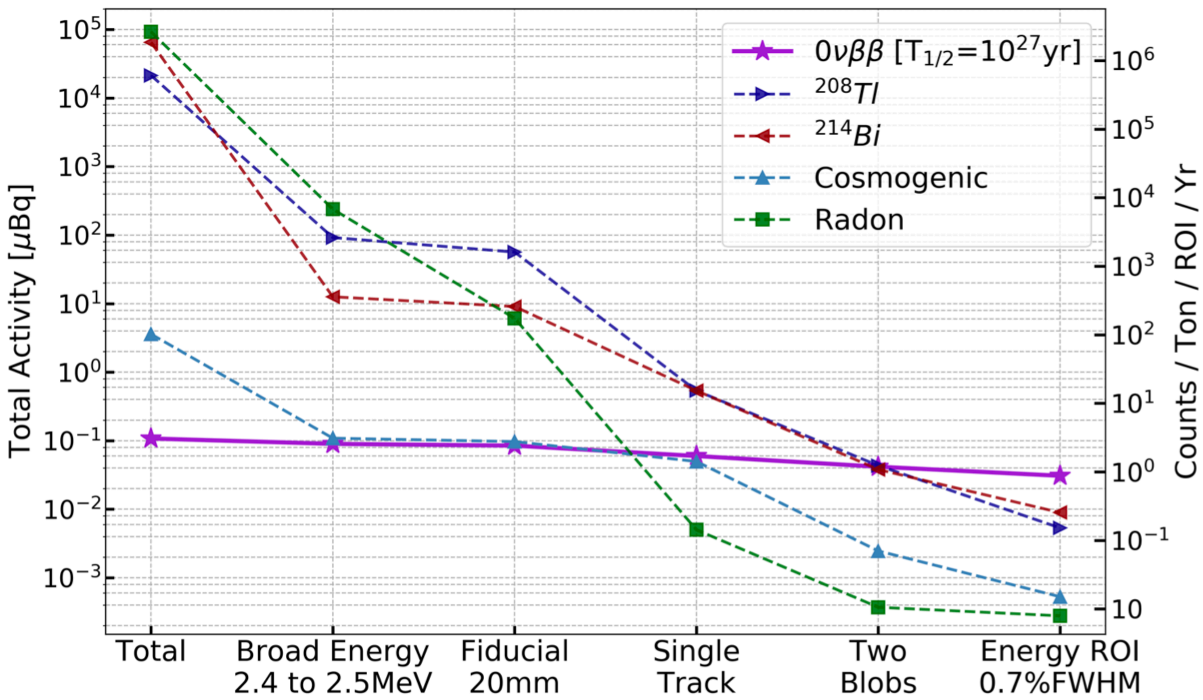
(~2026?)

Ton-scale experiment in conceptual design stage
I present projections and selected ongoing R&D



← NEXT-HD event selection assuming 0.7% energy resolution and demonstrated topological cut performance

NEXT-100 Background Model:

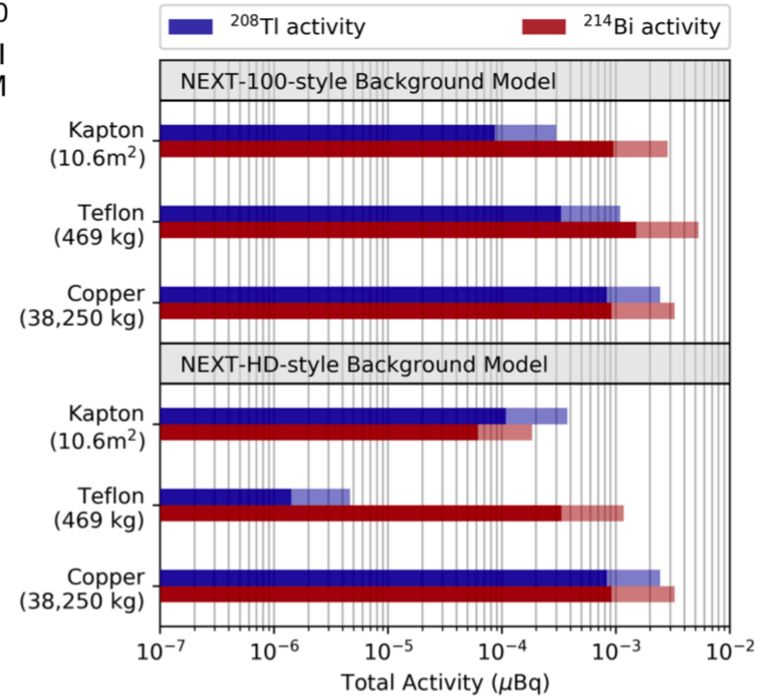
Only assayed materials for NEXT-100.

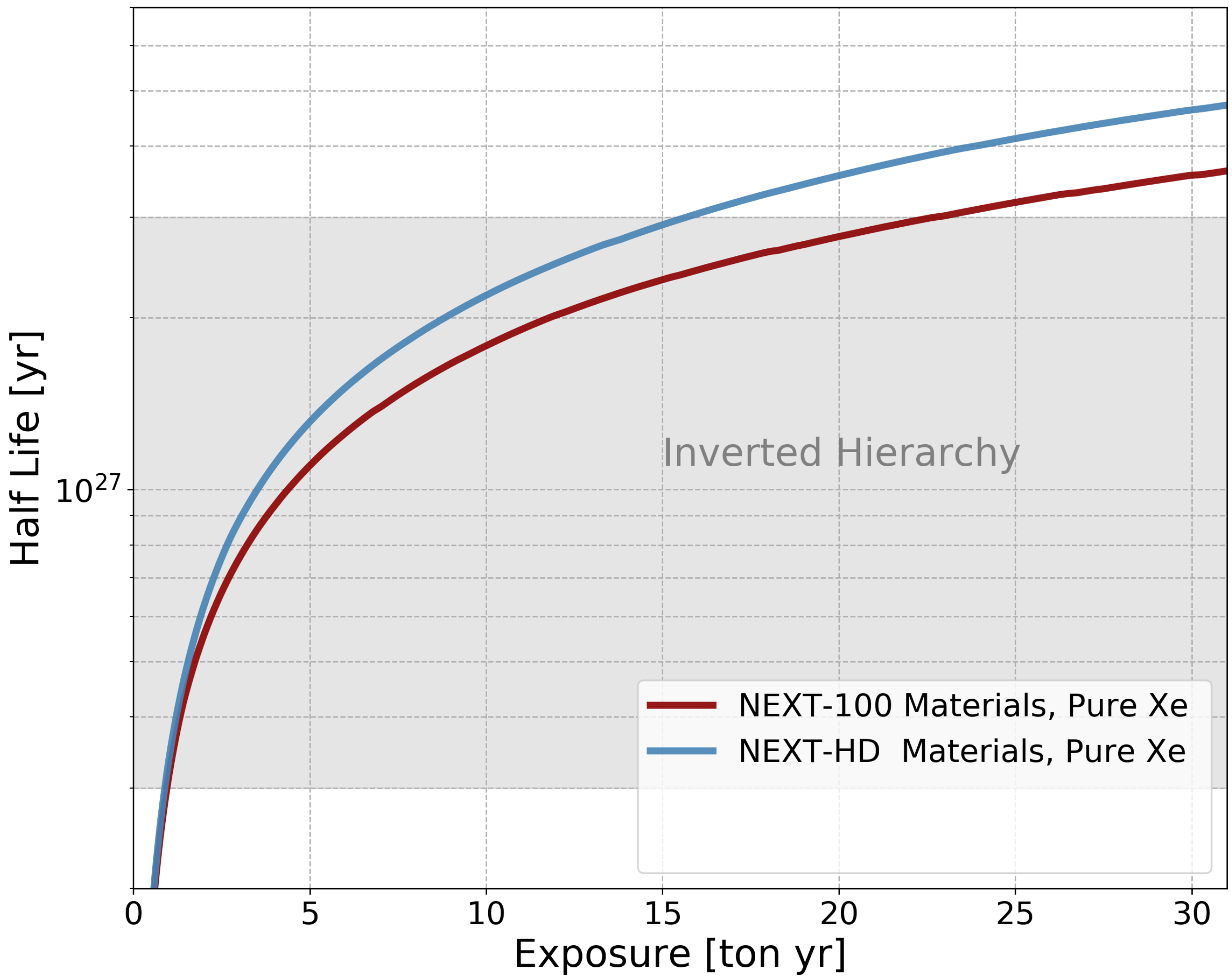
- Pure Xe: 0.5 / [ct ton yr ROI];
- Low Diffusion: 0.15 / [ct ton yr ROI]

NEXT-HD Background Model:

Cleaner Teflon and Kapton located by other collaborations:

- Pure Xe: 0.25 / [ct ton yr ROI];
- Low Diffusion: 0.08 / [ct ton yr ROI]





- **NEXT-100 background model** includes all assayed NEXT-100 materials.
- **NEXT-HD background model** takes advantage of cleaner materials (Teflon and Kapton) already identified by other collaborations.

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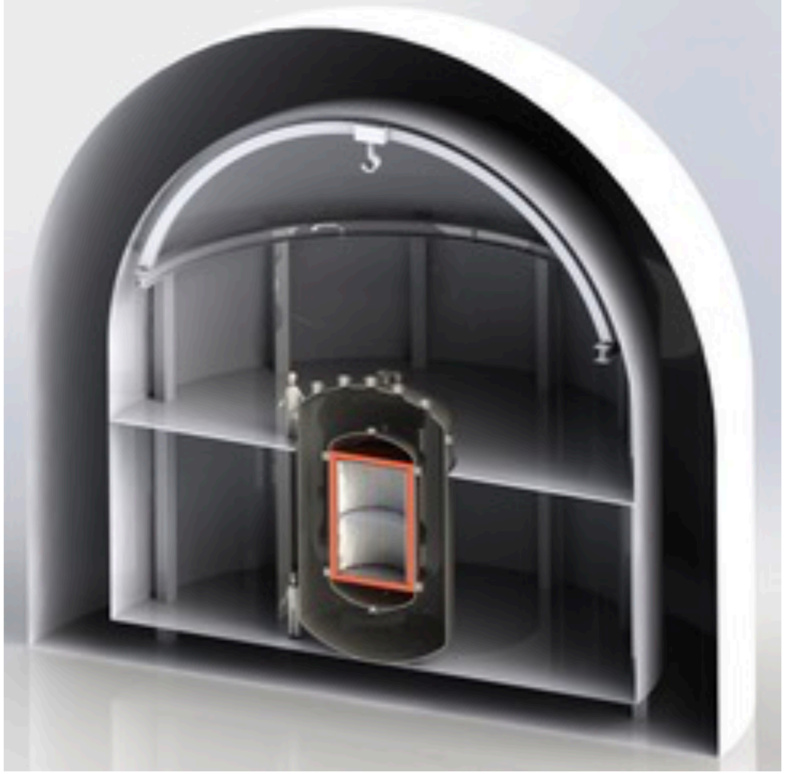
(~2023)

→ ***NEXT-BOLD***

(~2026?)



Ton-scale experiment in conceptual design stage
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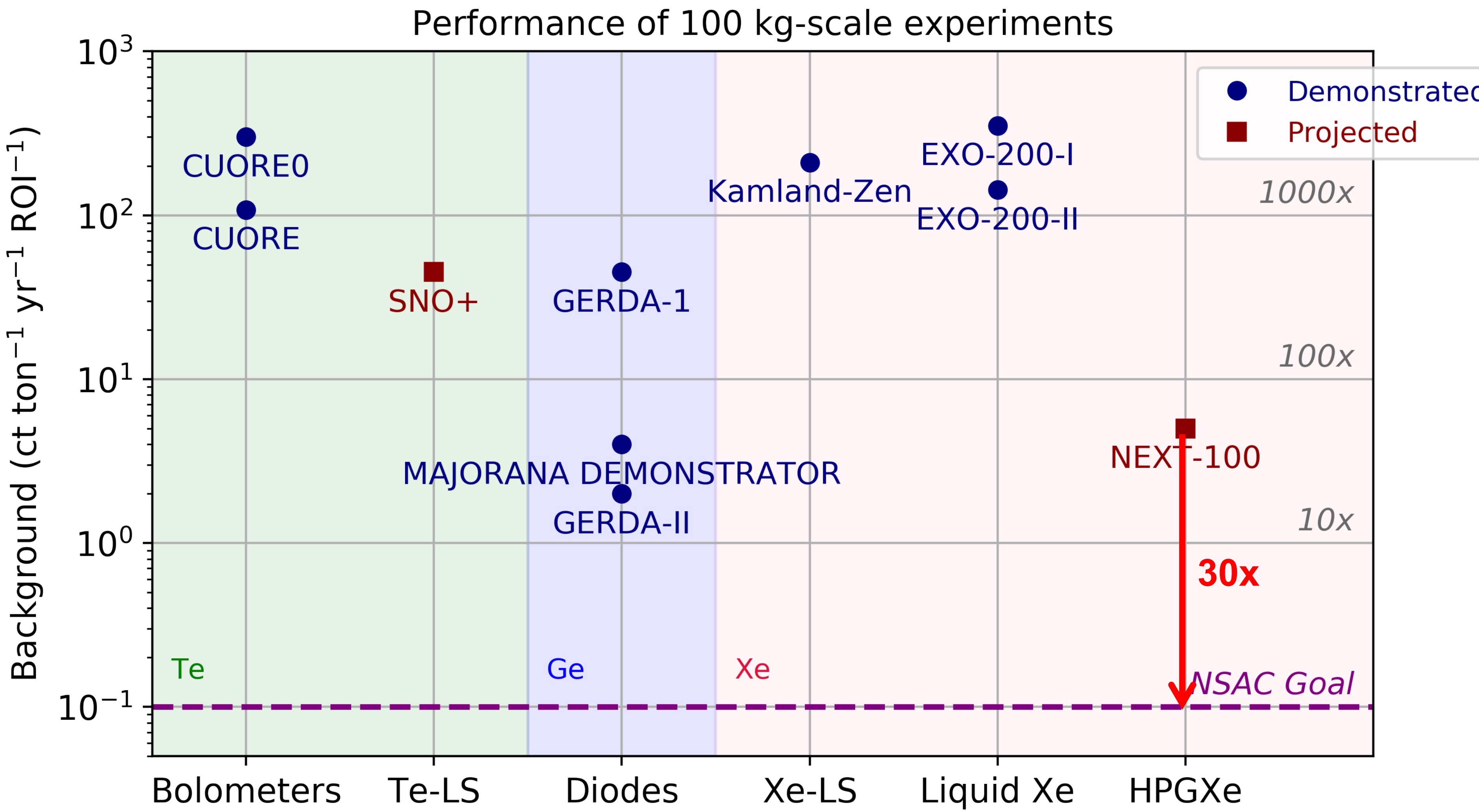
NEXT-100 will be comfortably world leading in xenon, and competitive with world leaders in other isotopes.

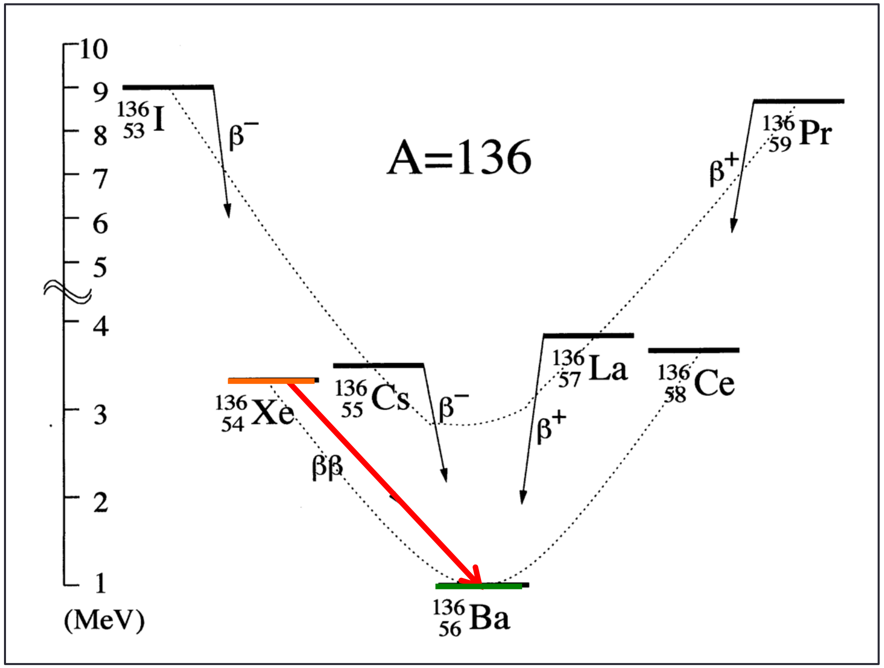


But, world-leading is not enough.

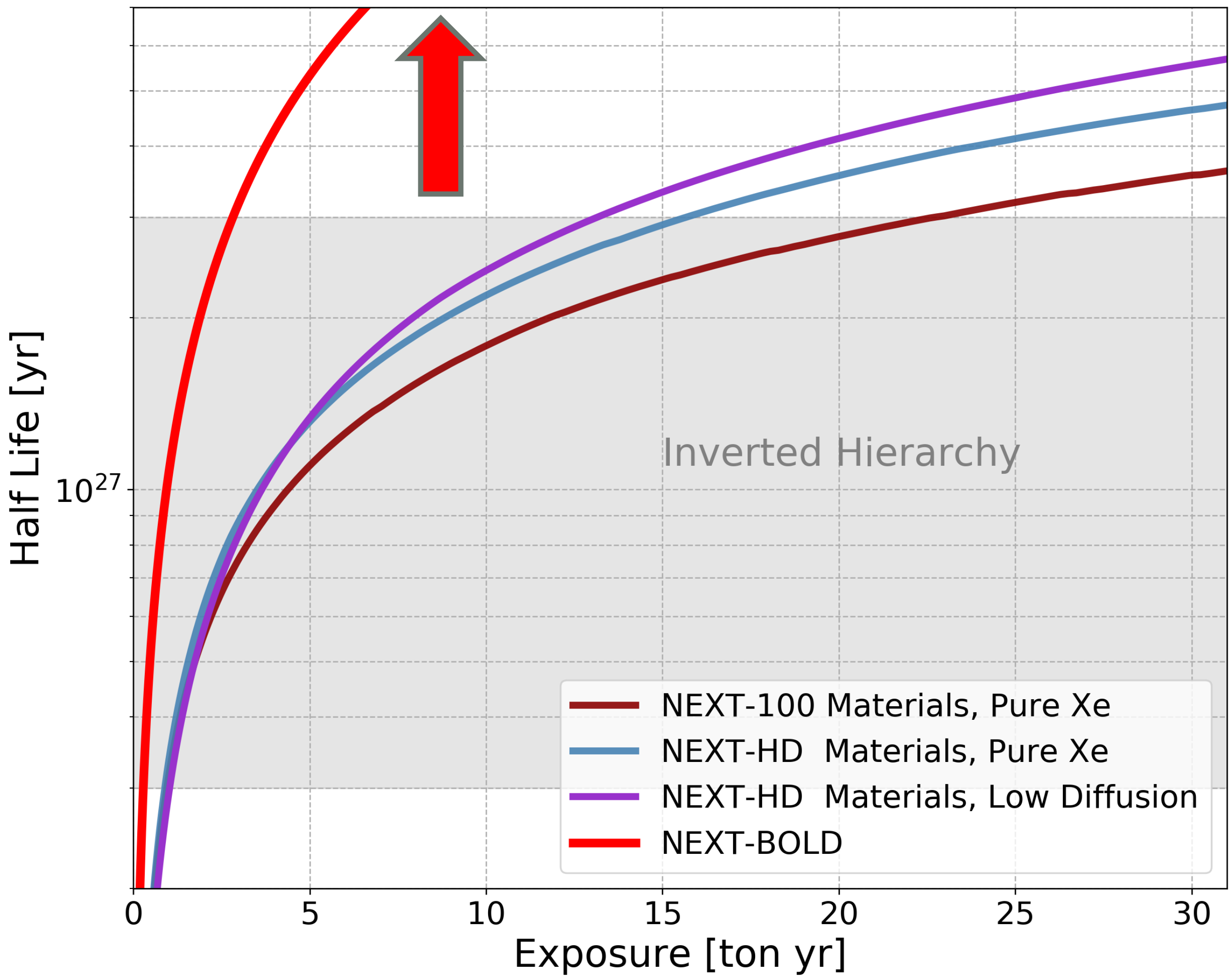
Barium Tagging

- Barium ion is only produced in a true ββ decay, not in any other radioactive event.
- Identification of Ba ion plus ~1% FWHM energy measurement would give a background-free experiment.
- ***Is it plausible to detect an individual barium ion or atom in a ton of material?***

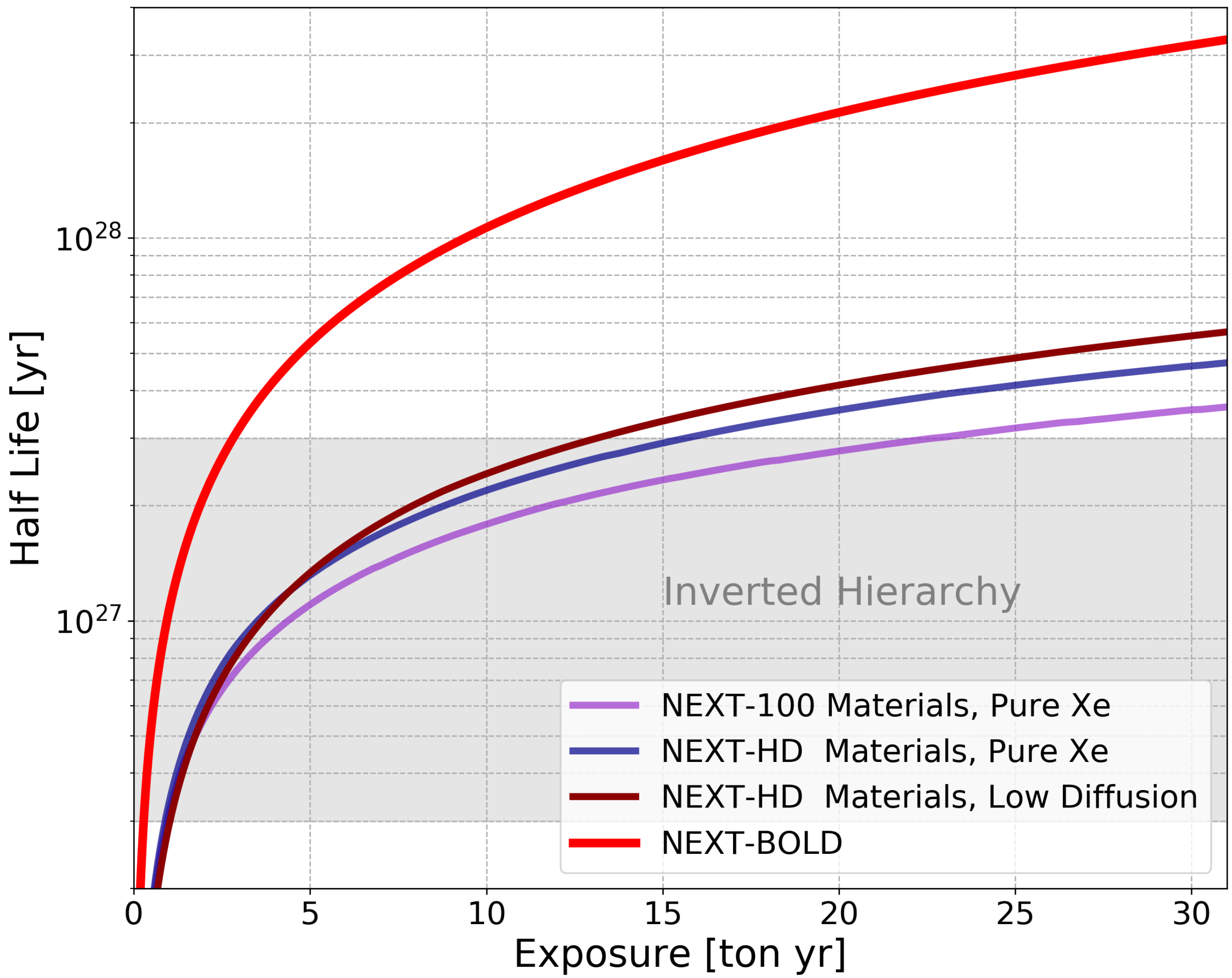
$^{136}Xe \rightarrow {}^{136}Ba + e + e$



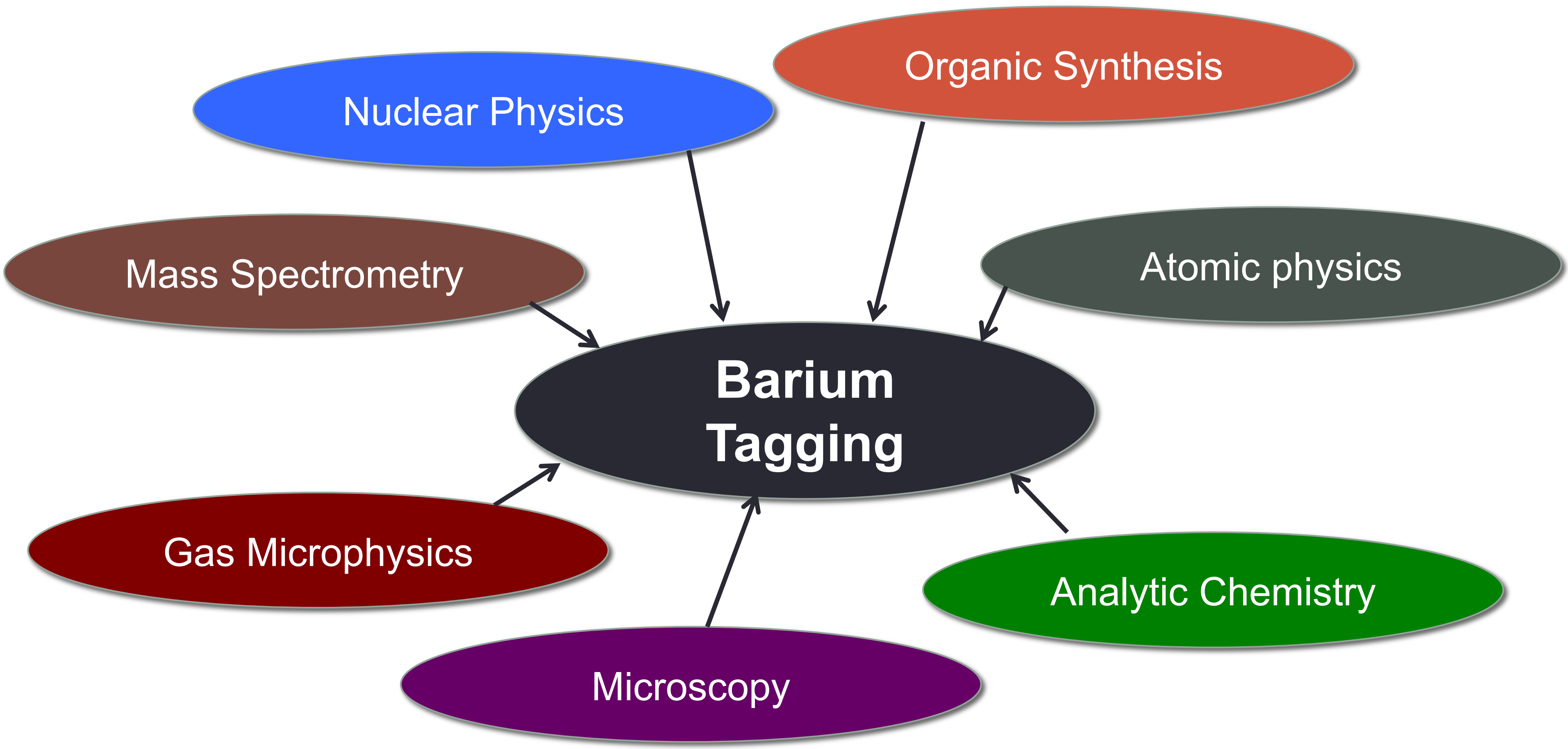
Barium tagging for 0nubb has been actively explored in liquid and gas xenon for ~30 years, with the holy grail is a scalable single ion sensitive technology.



- **NEXT-BOLD would represent a dramatic sensitivity improvement through combination of signal efficiency increase and background reductions**



- **NEXT-BOLD would represent a dramatic sensitivity improvement through combination of signal efficiency increase and background reductions**

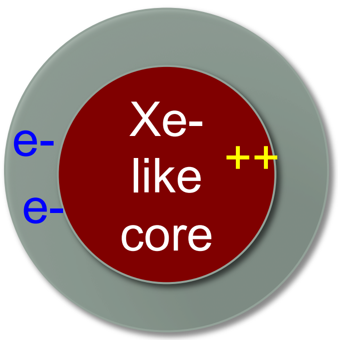

Organic Synthesis
Nuclear Physics
Atomic physics
Mass Spectrometry
Barium Tagging
Gas Microphysics
Analytic Chemistry
Microscopy

Basics - Barium Atoms and Ions

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

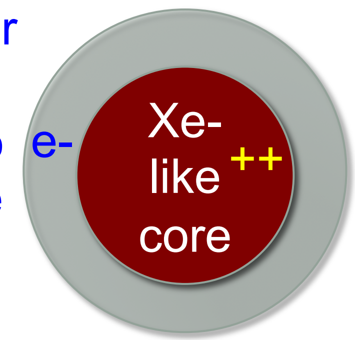
Structure of the **Ba** atom

Two outer electrons bound to +ve core



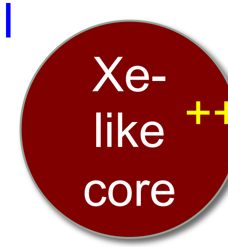
Structure of the **Ba+** ion

One outer electron bound to +ve core



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

Noble-like electron shell

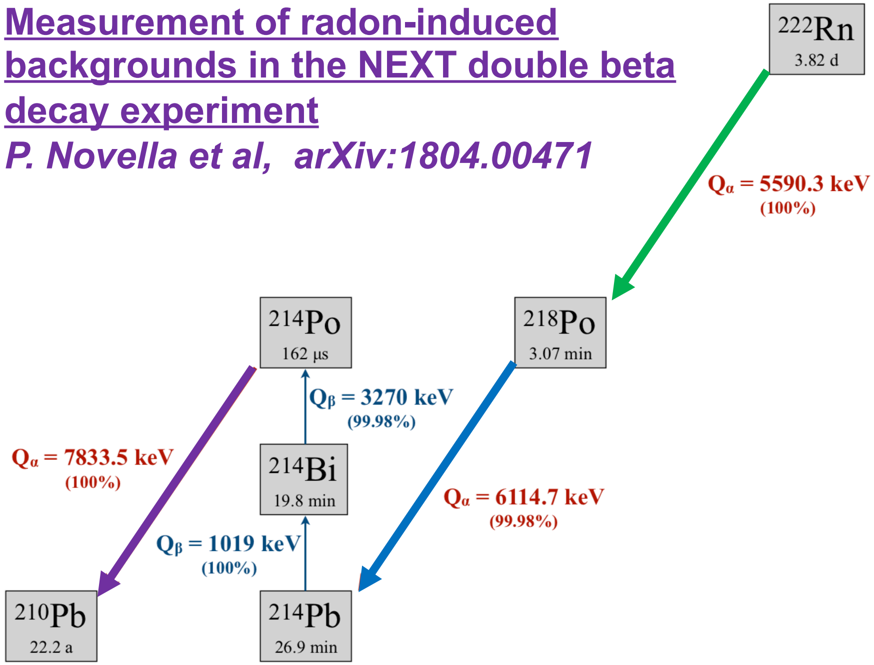


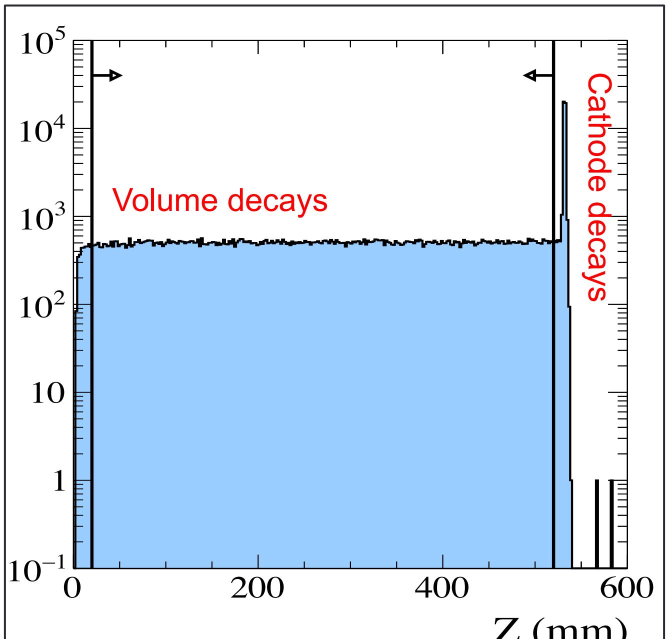
Liquid –
Some distribution

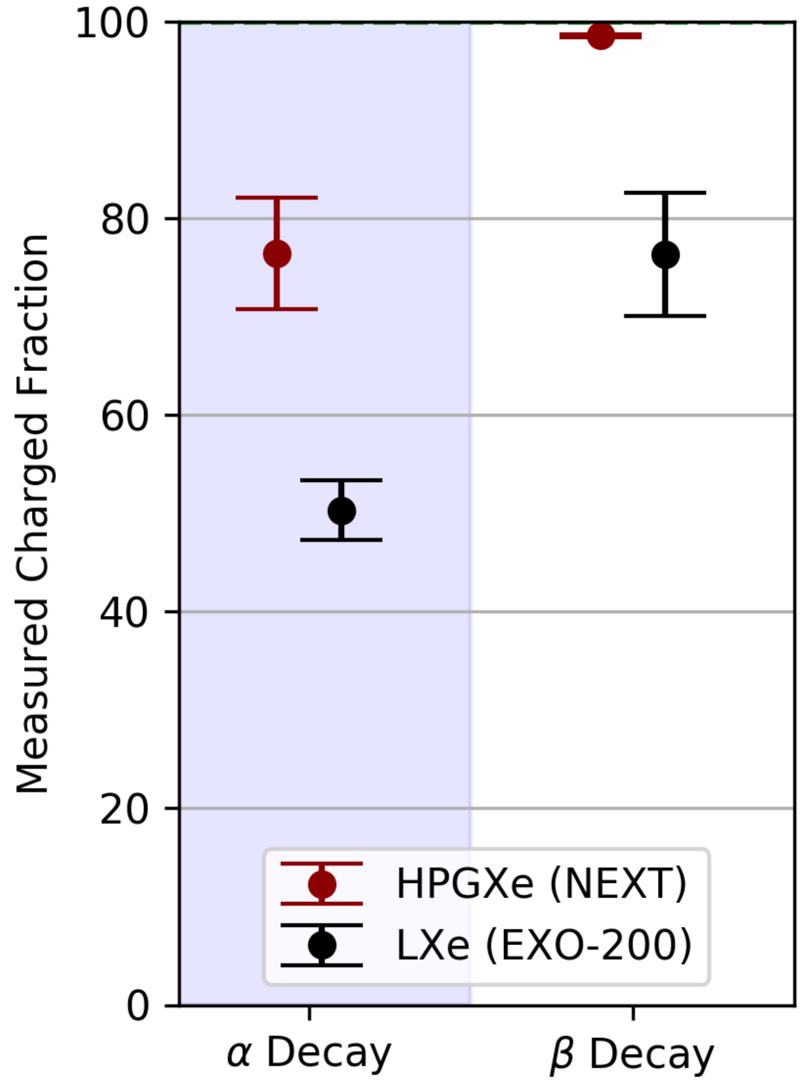
Gas –
Mostly this

Ionic Charge State in Liquid and Gas

Measurement of radon-induced backgrounds in the NEXT double beta decay experiment
P. Novella et al, arXiv:1804.00471



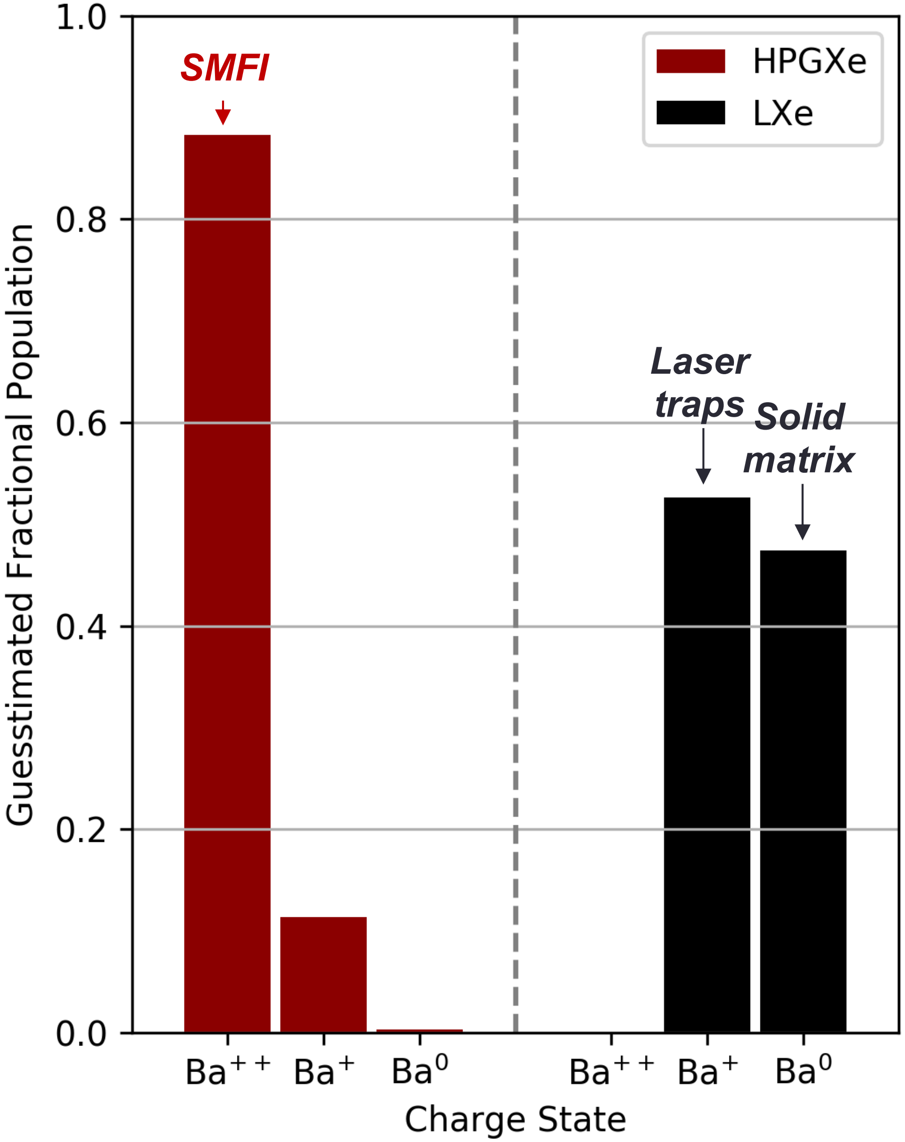




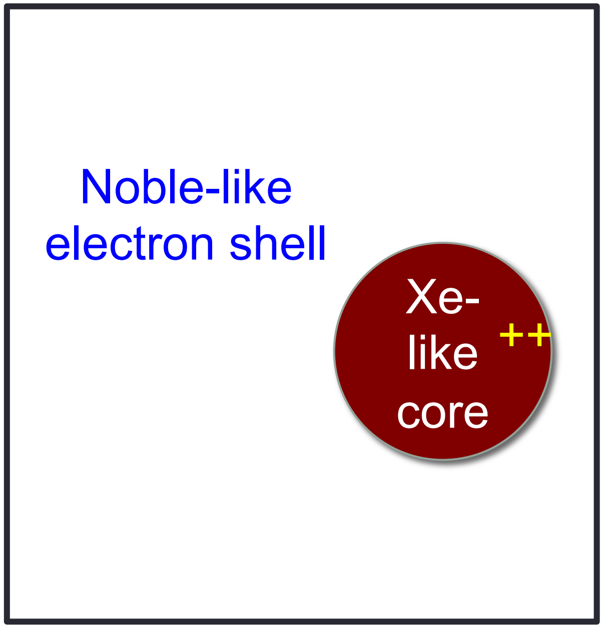
Extrapolating…

- Using "best guess" assumptions:
  - Ba++ has 2x larger Onsager radius than Ba+, so expect 4x larger capture cross section for recombination.
  - In double beta decay, two emerging tracks at nucleus, so 2x recombination of single decay.
- Guesstimate Ba charged fractions in double beta decay →

My back-of-envelope guesstimates:
(nb: reasonable people disagree)



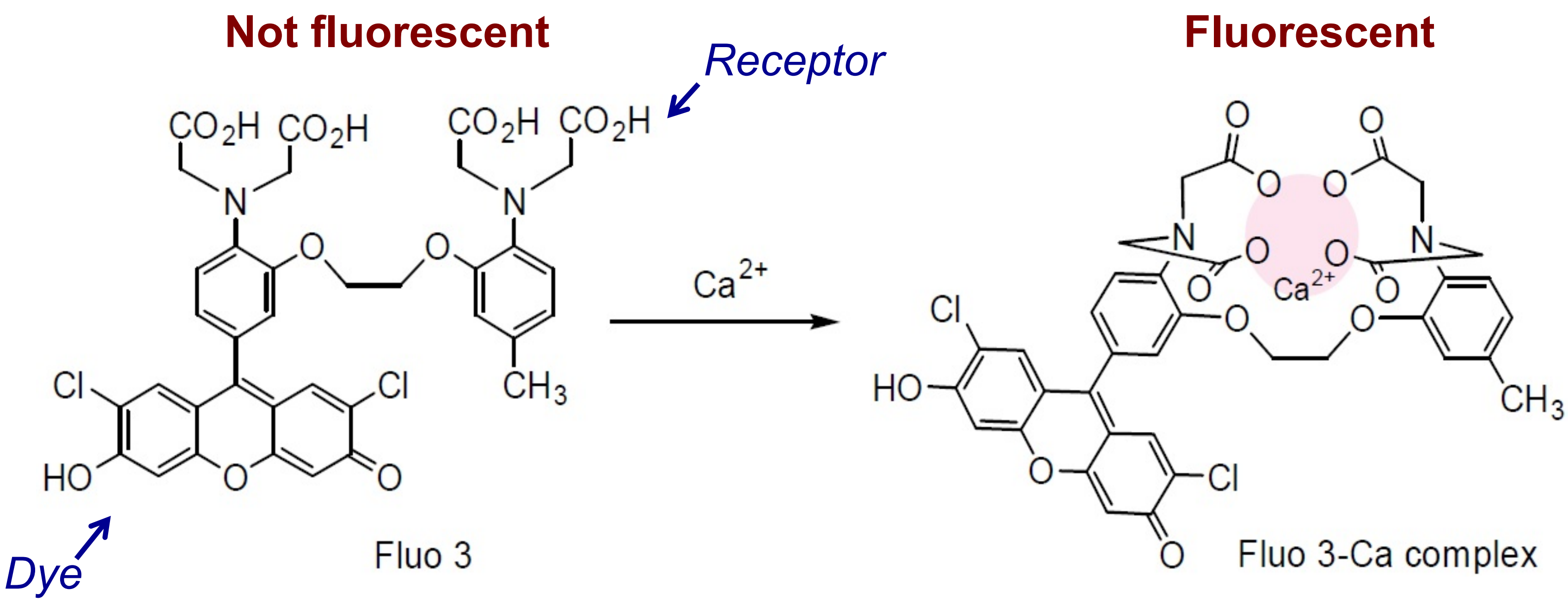
Q: How do you make Ba^{++} shine?



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

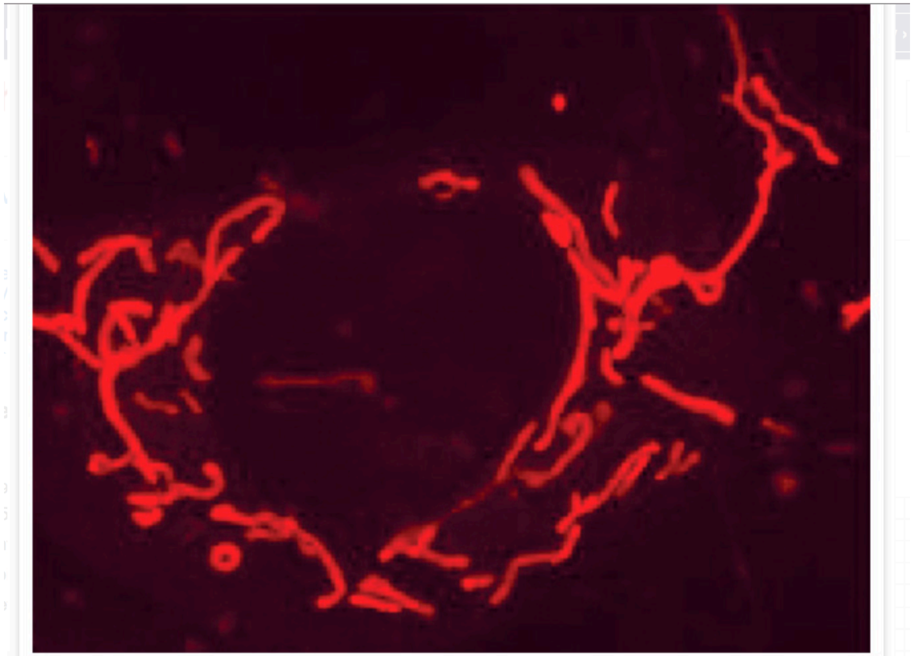
SMFI:

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



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

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



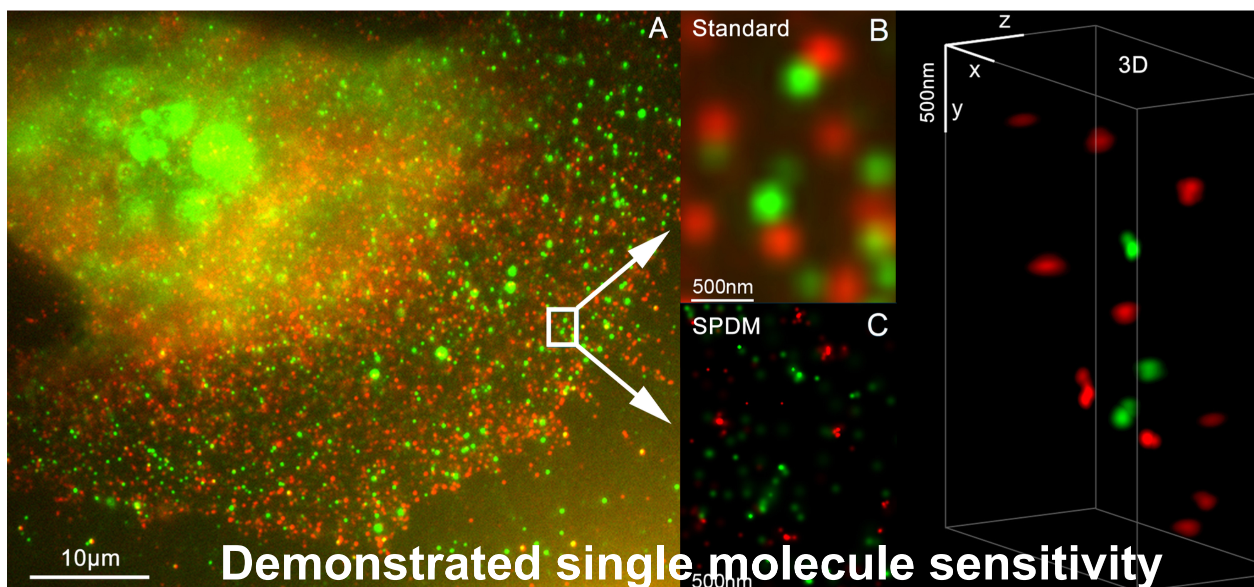
← Rhod-2 sensing Ca^{++} production in rat astrocyte cells

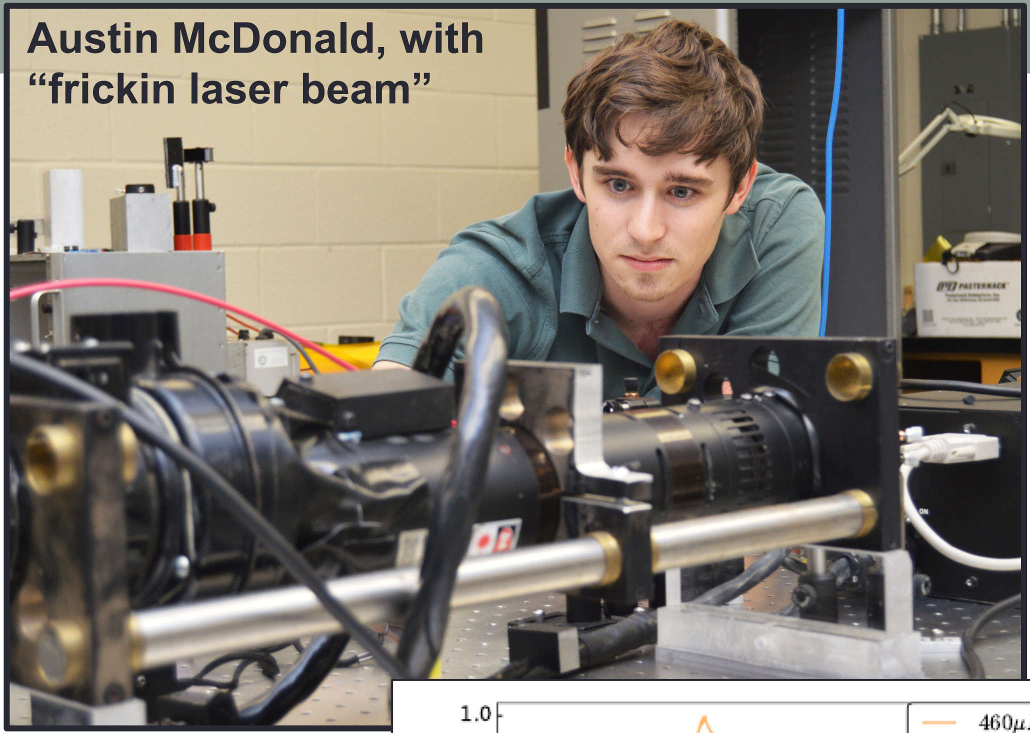
J Microsc. 2011 Apr;242(1):46-54

J Cell Biol 145, 795 (1999).

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

These methods won the Nobel Prize in chemistry in 2014.

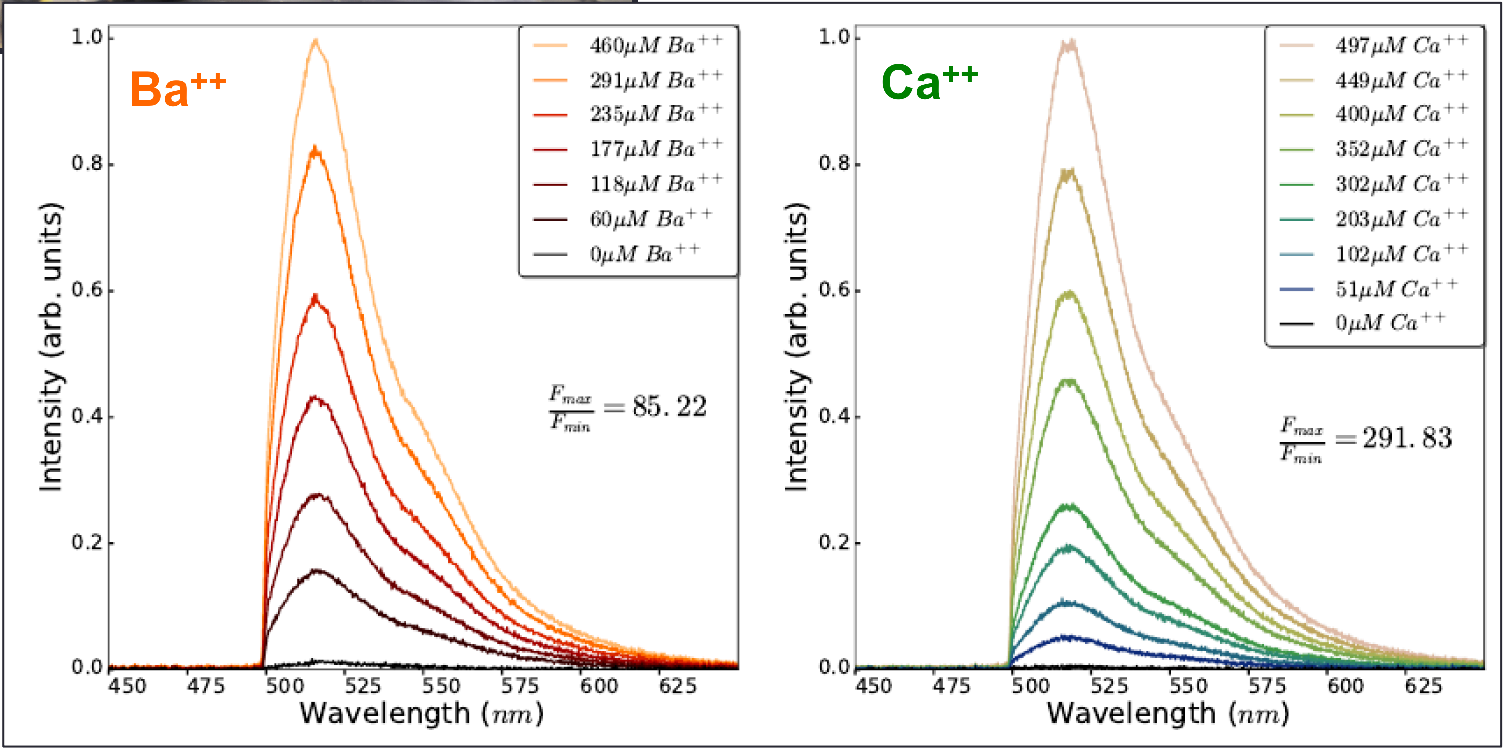


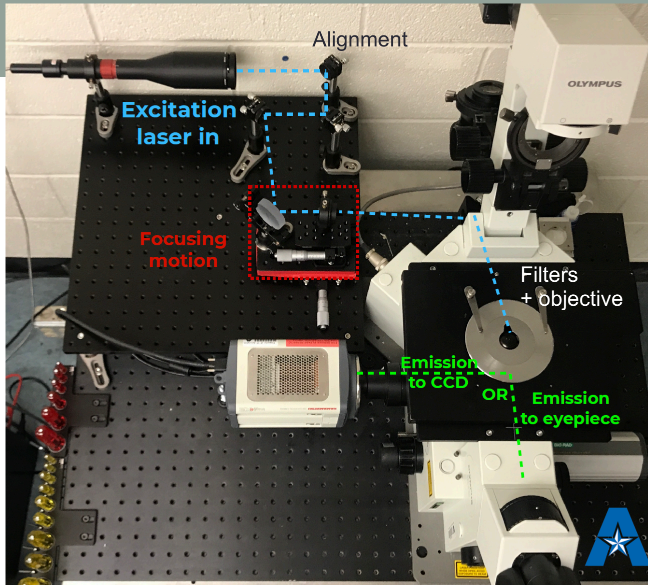


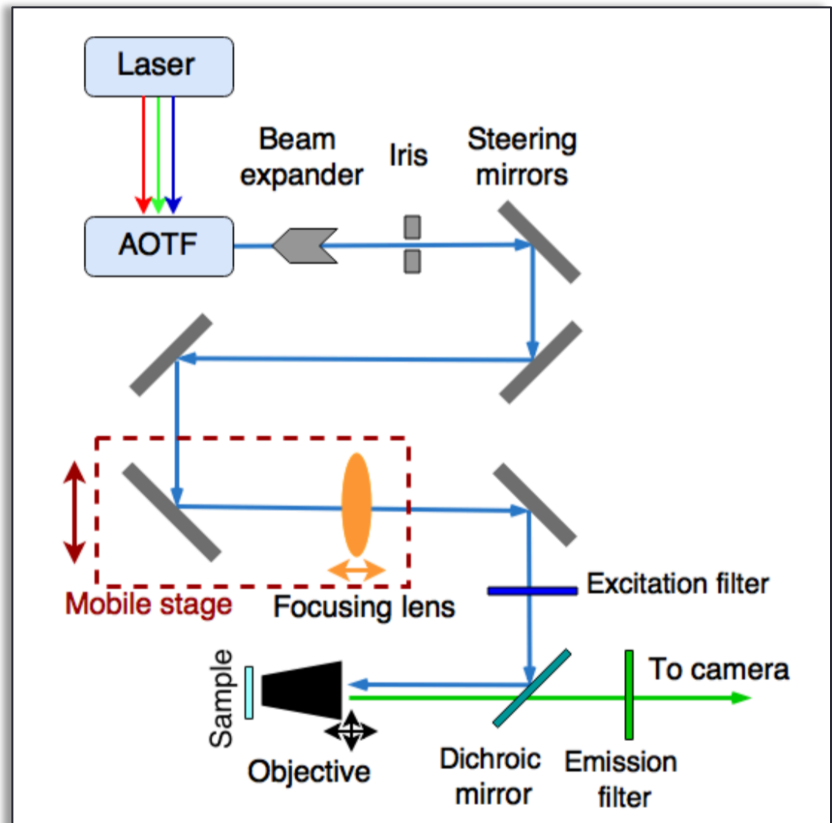
← First dabbling - developed a bespoke fluorescence sensor to study barium production at the end of a fiber.

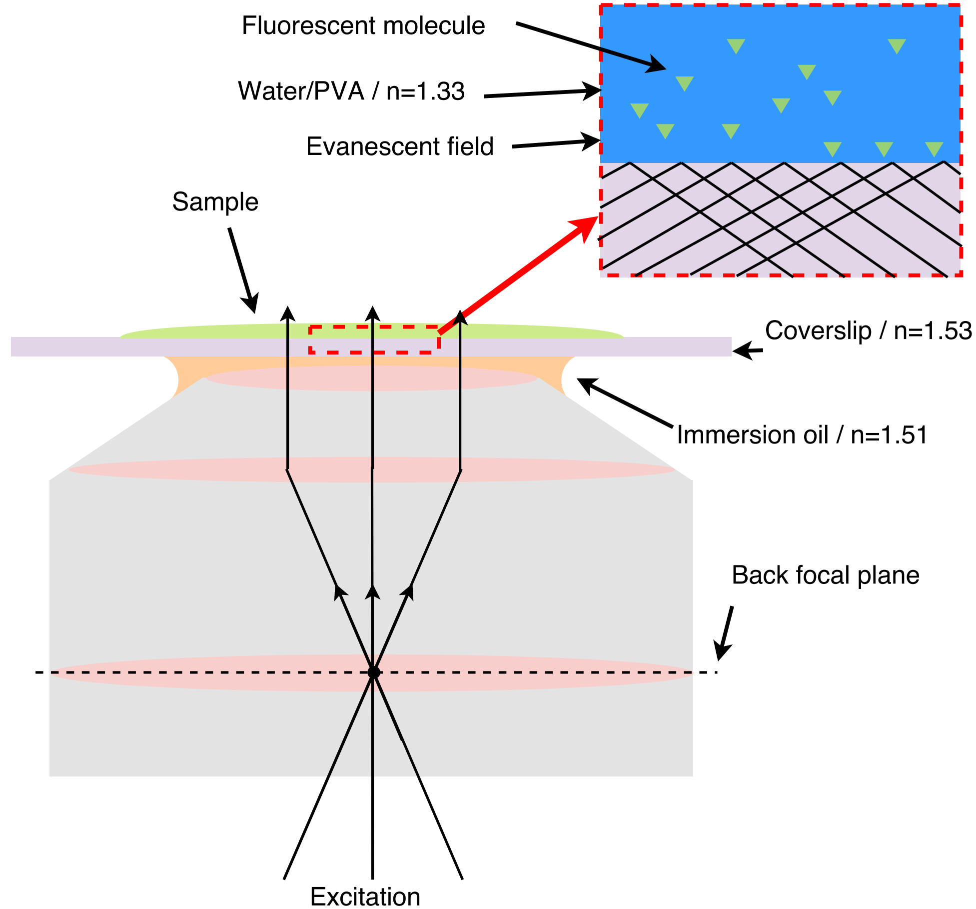
Single molecule fluorescence imaging as a technique for barium tagging in neutrinoless double beta decay
Jones, McDonald, Nygren, JINST (2016) 11 P12011

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



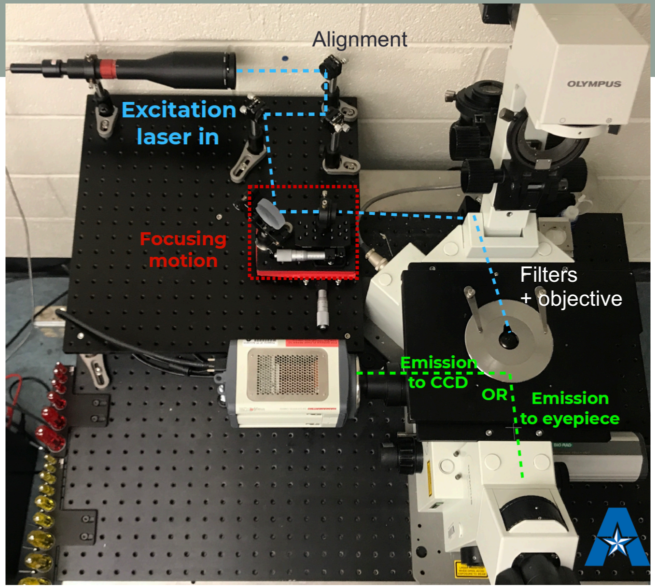


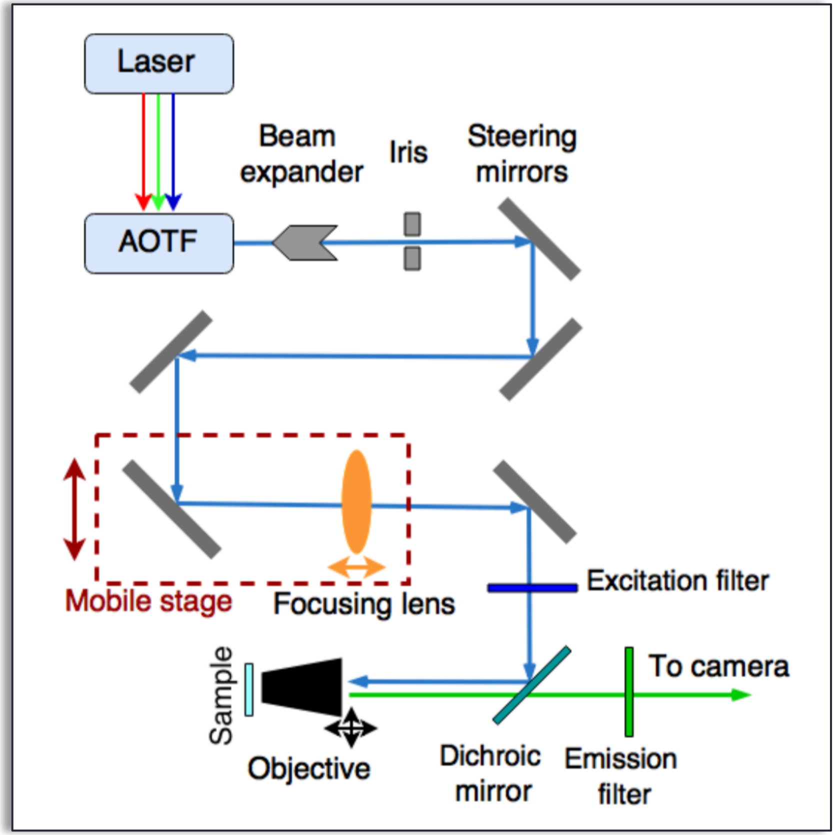


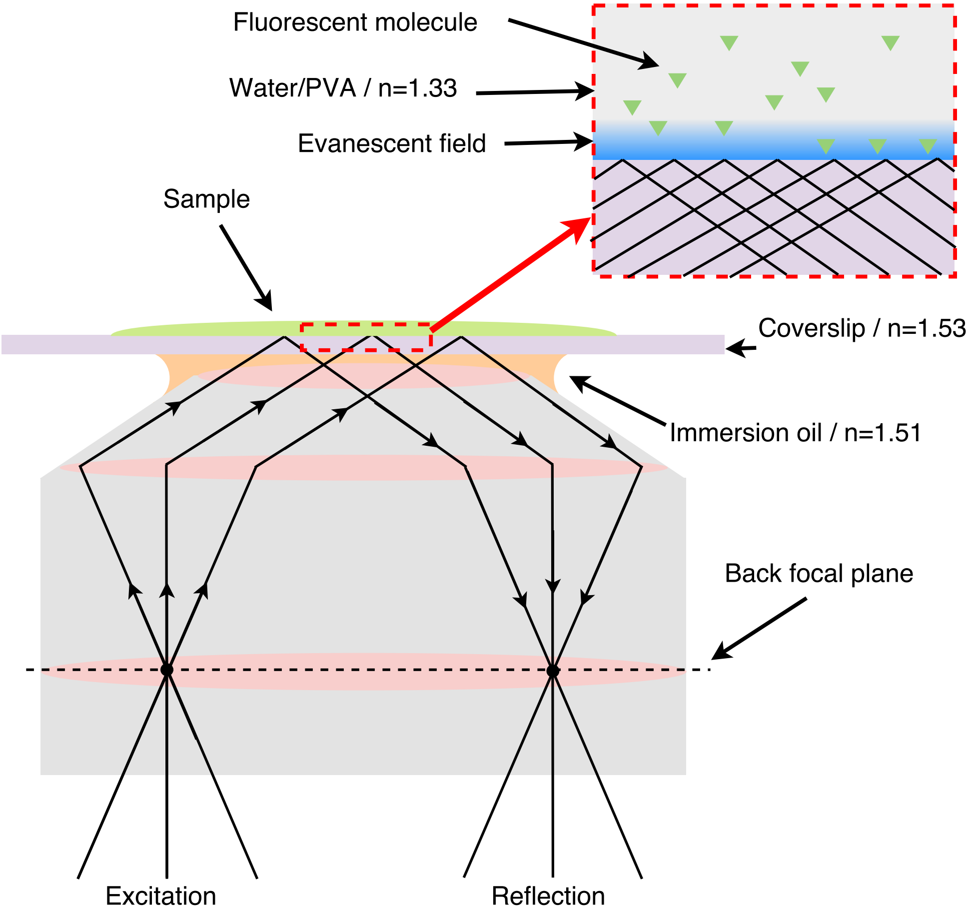


TIRF

Total Internal Reflection Fluorescence microscopy

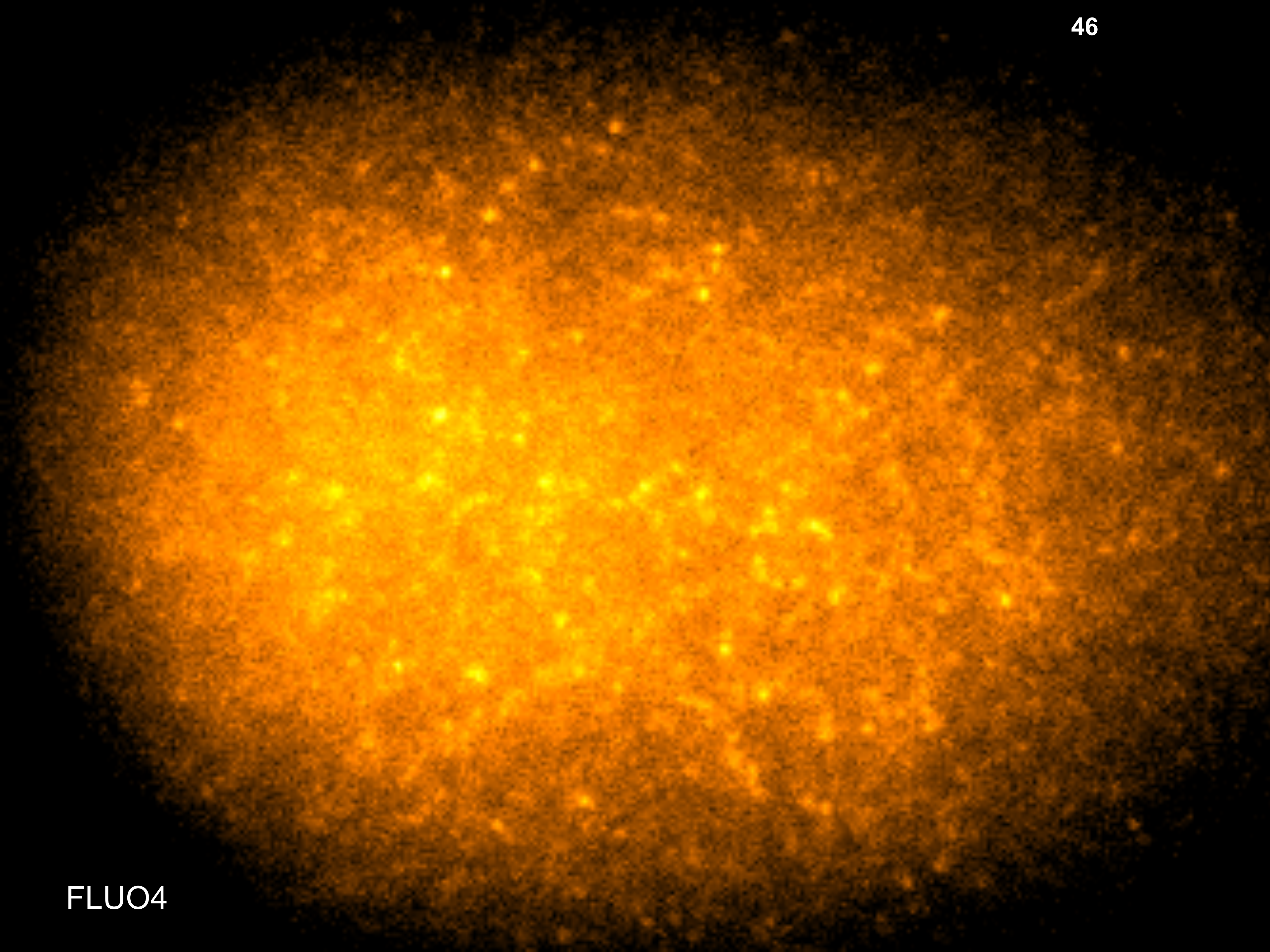


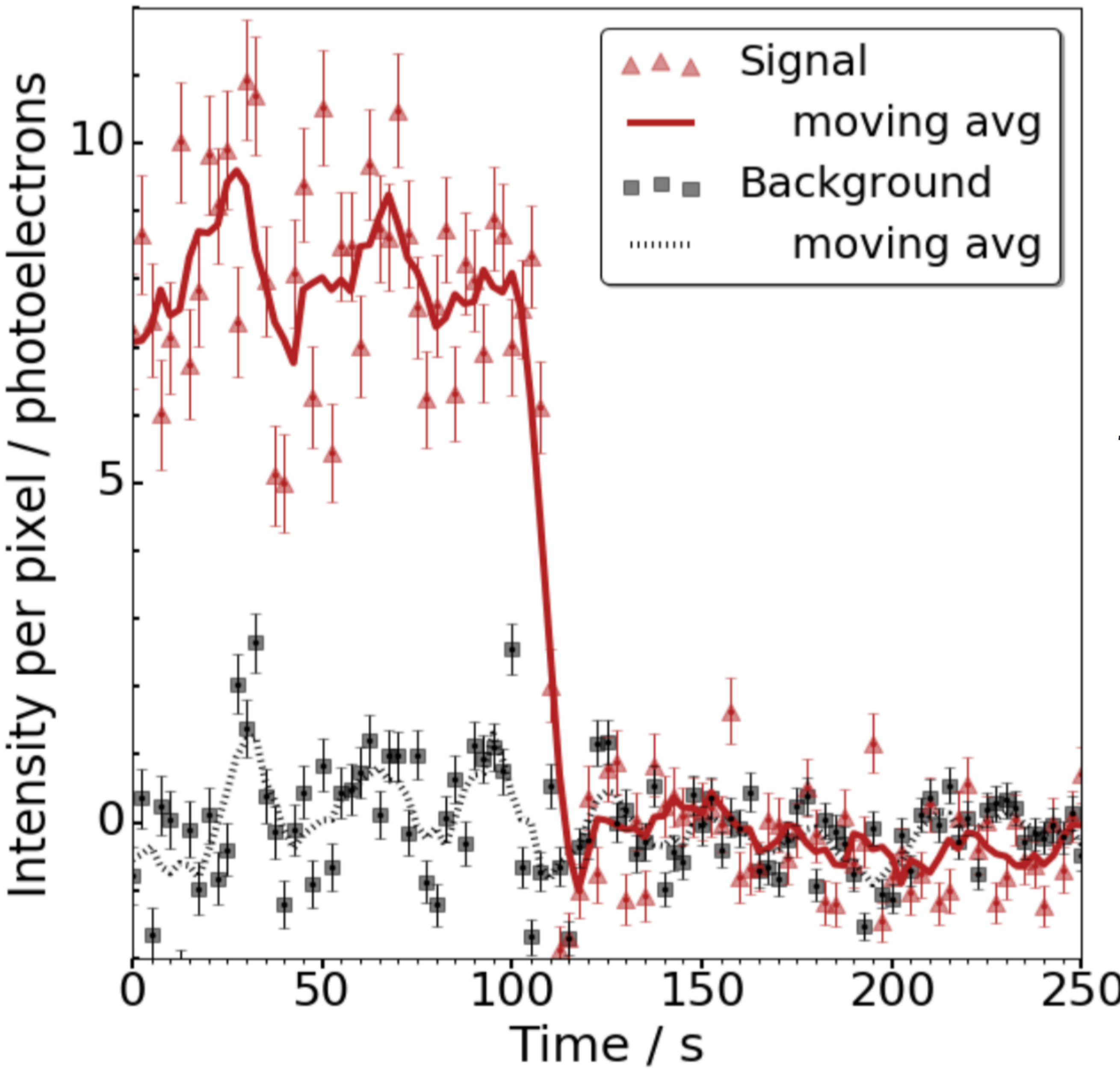




TIRF

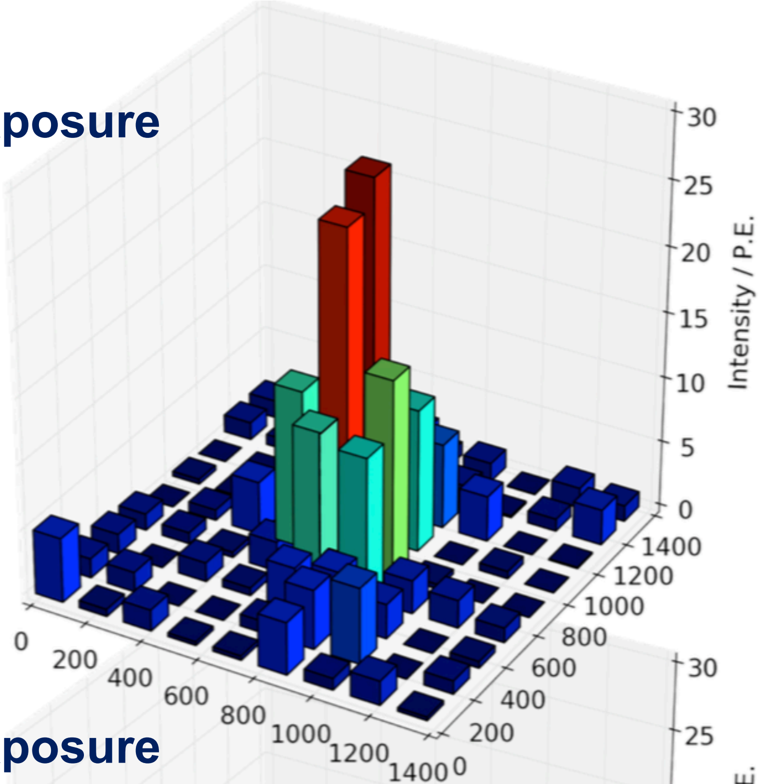
Total Internal Reflection
Fluorescence microscopy


FLUO4

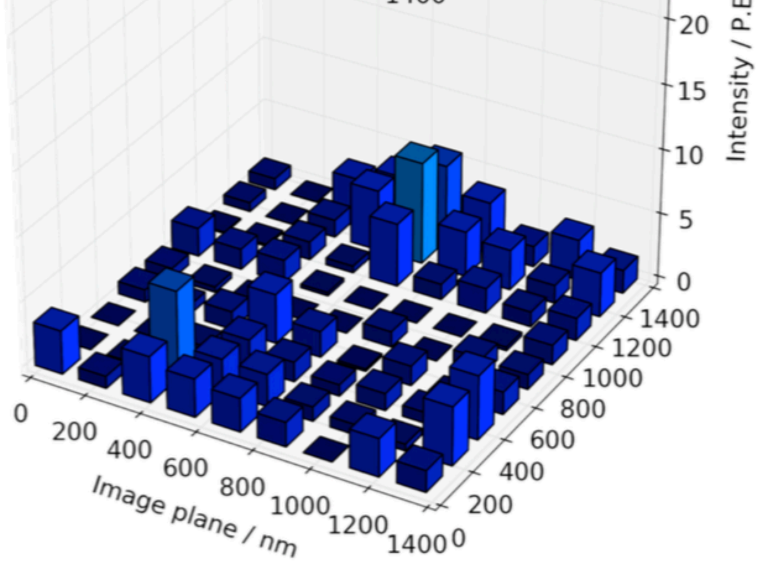


This "step" is how you know it is exactly one ion.

0.5s exposure before:

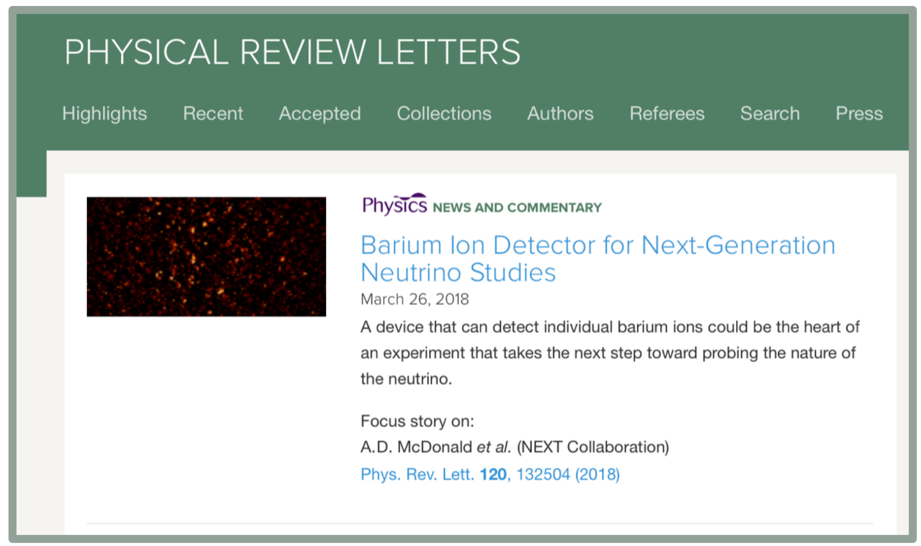


0.5s exposure after:



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

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



First demonstration of single Ba++ ion resolution.

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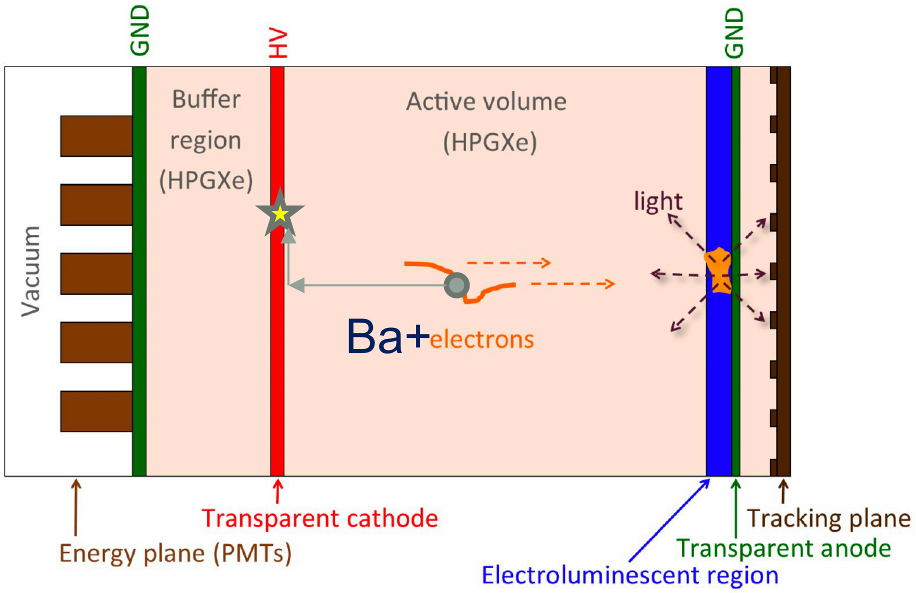
- 5. Combine into a working sensor for NEXT prototype

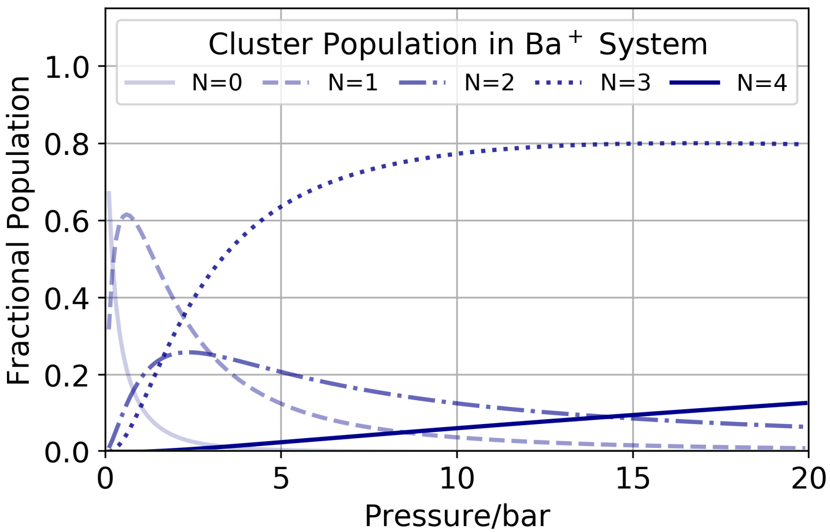


SUPPORTED BY:
U.S. DEPARTMENT OF ENERGY

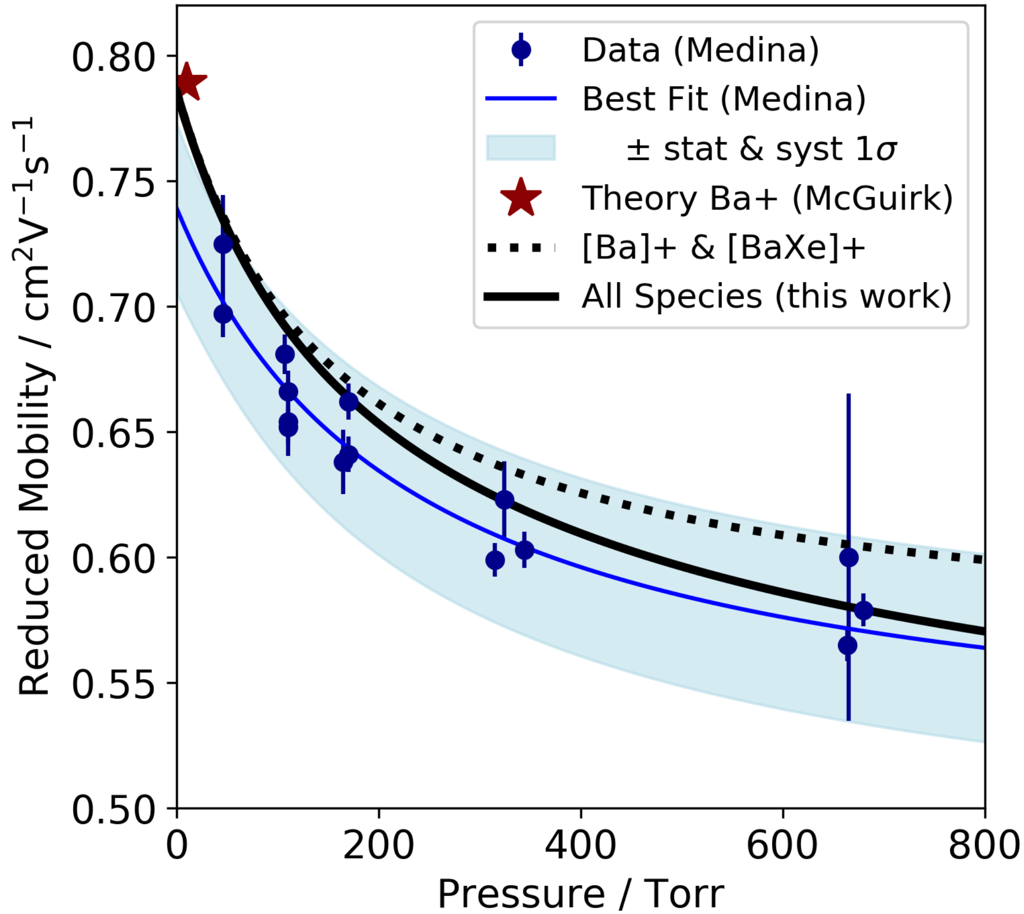
Barium mobility in gas – in theory:

Not a single ion drifting!

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



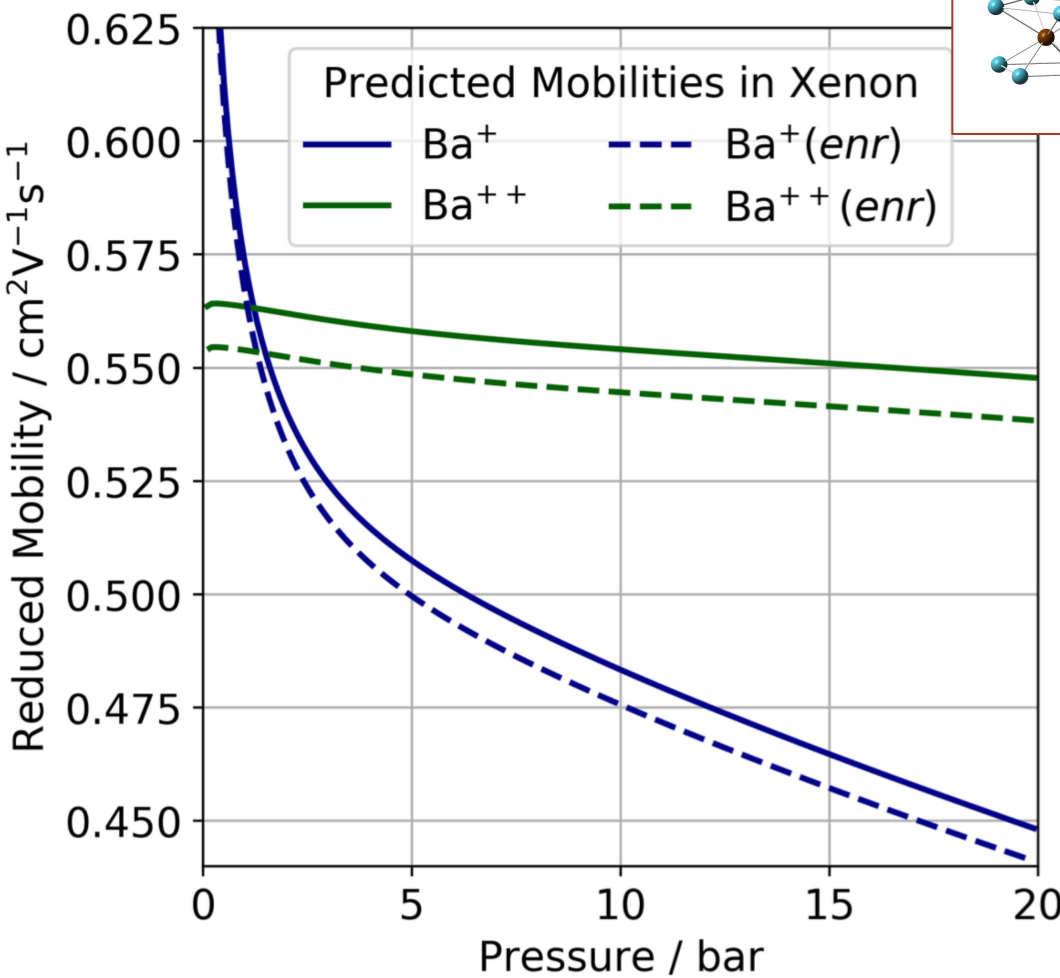
Excellent agreement with data for Ba+



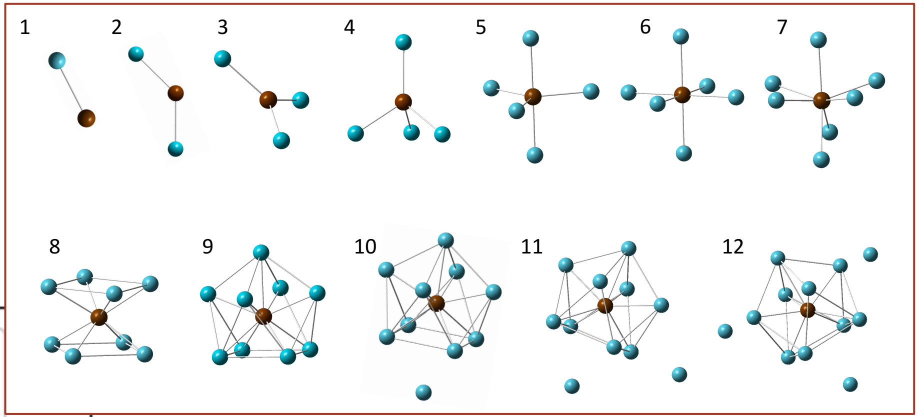
Mobility and Clustering of Barium Ions and Dications in High Pressure Xenon Gas

E. Bainglass, B.J. P. Jones, et. al. Phys.Rev. A97 (2018) no.6, 062509

For Ba++ things get more complicated.



Calculated Ba++ clusters:



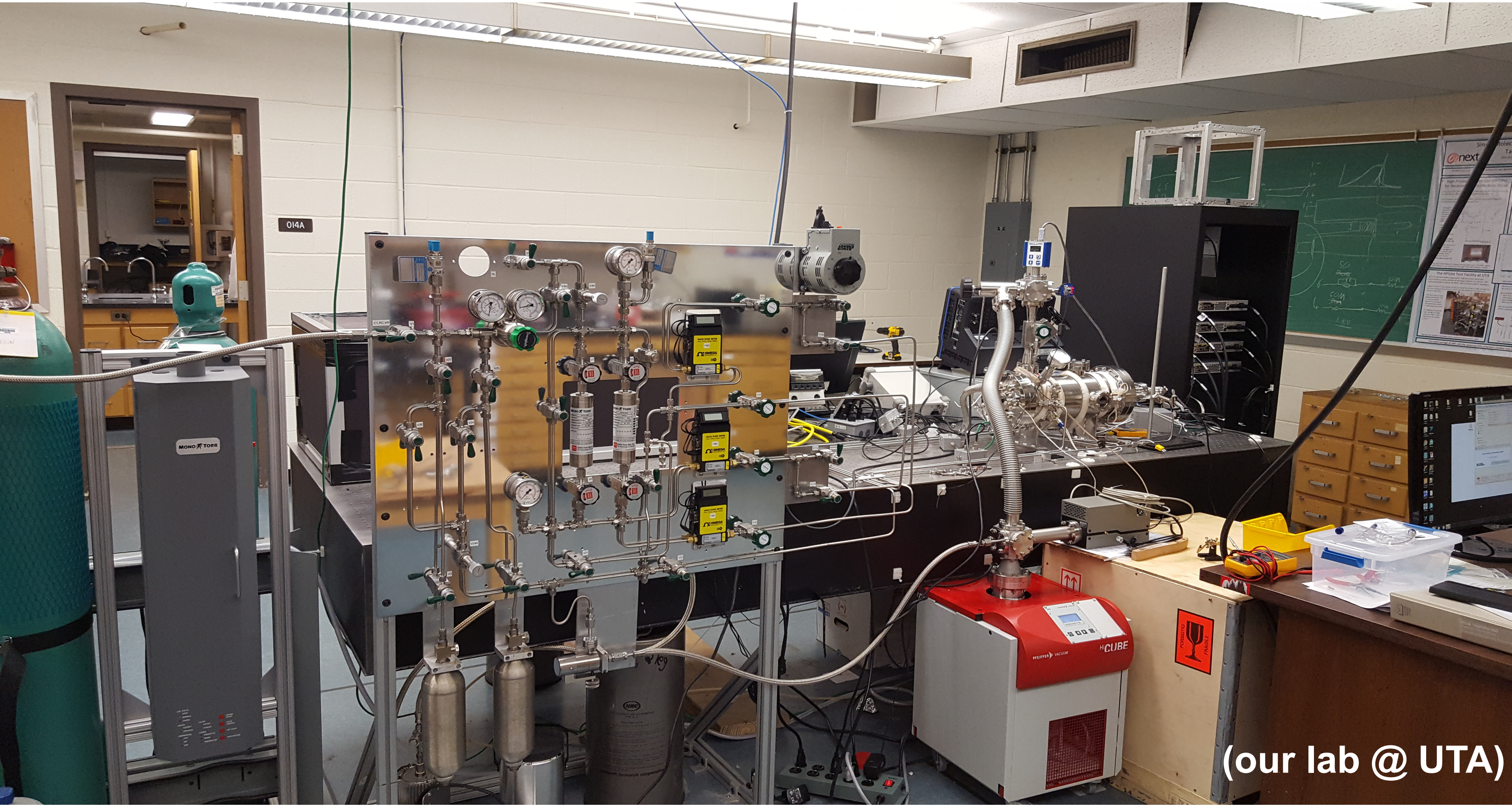
Bigger clusters more similar to each other, so less pressure dependence in Ba++ than Ba+

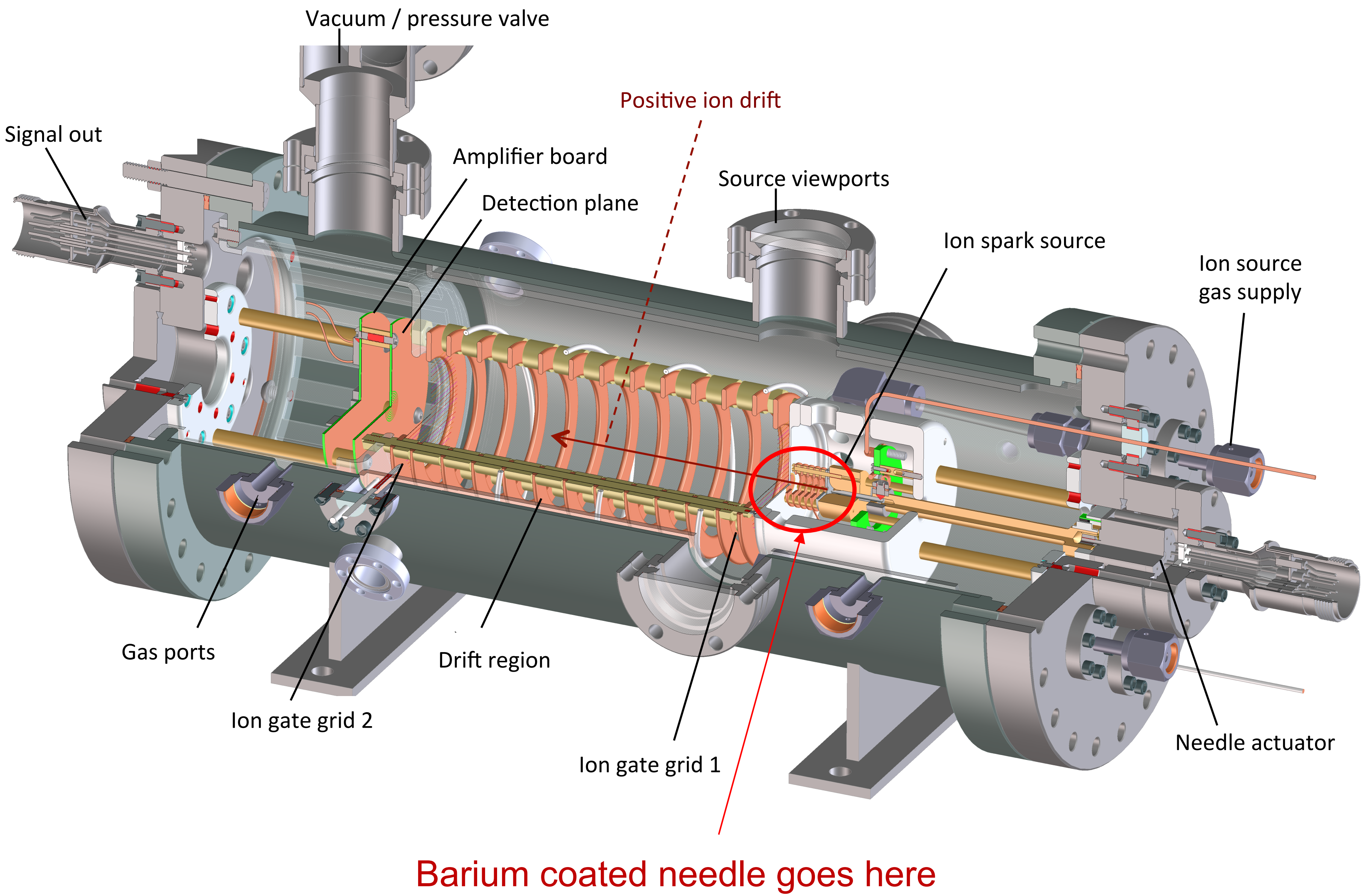
Isotopic composition changes scattering kinematics, so %-level differences with enriched xenon

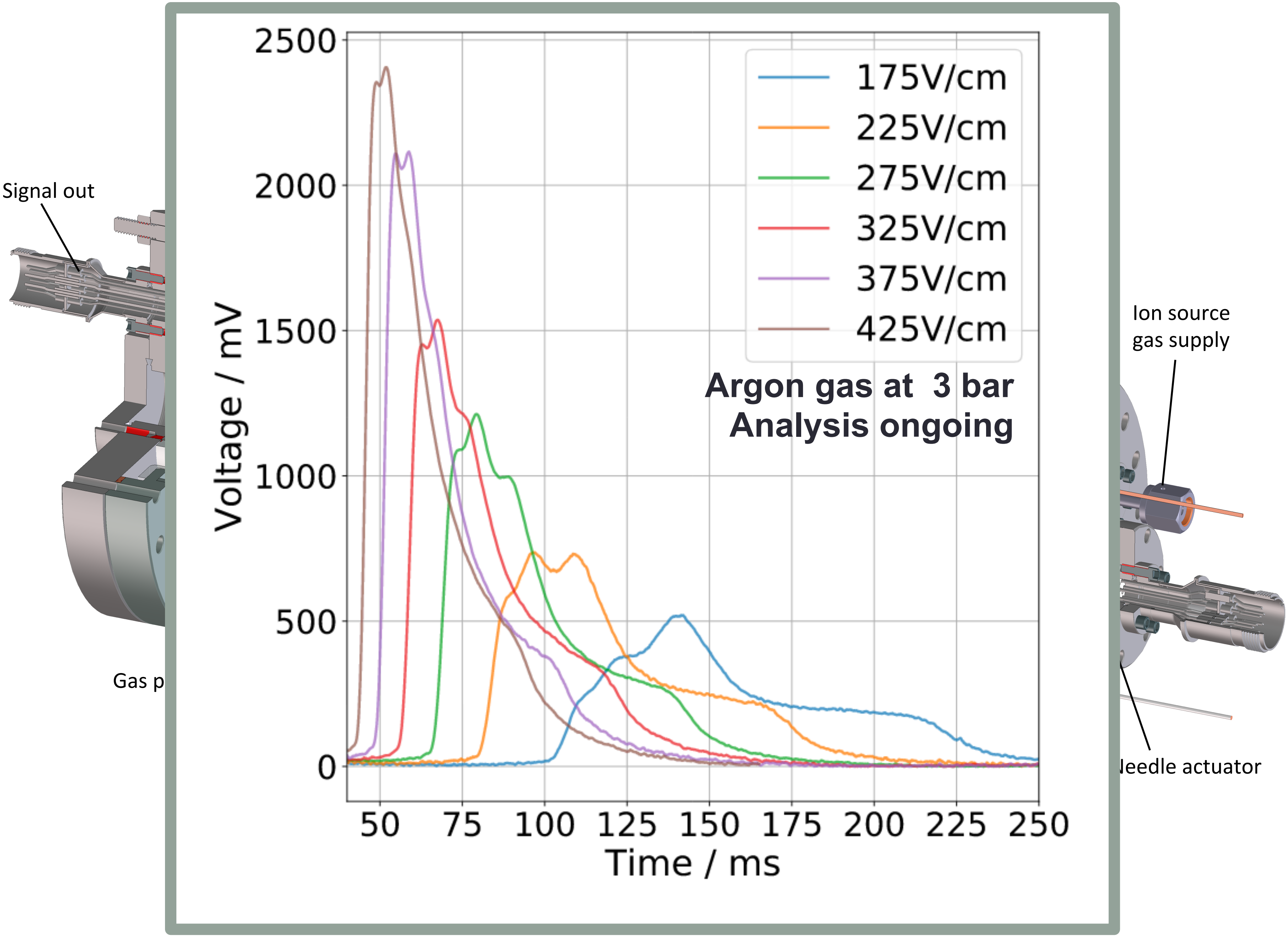
We will test this with experimental data soon!

The barium beam

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




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


Signal out
Gas p
Ion source
gas supply
Jeedle actuator
175V/cm
225V/cm
275V/cm
325V/cm
375V/cm
425V/cm
Argon gas at 3 bar
Analysis ongoing
Voltage / mV
0
500
1000
1500
2000
2500
50
75
100
125
150
175
200
225
250
Time / ms

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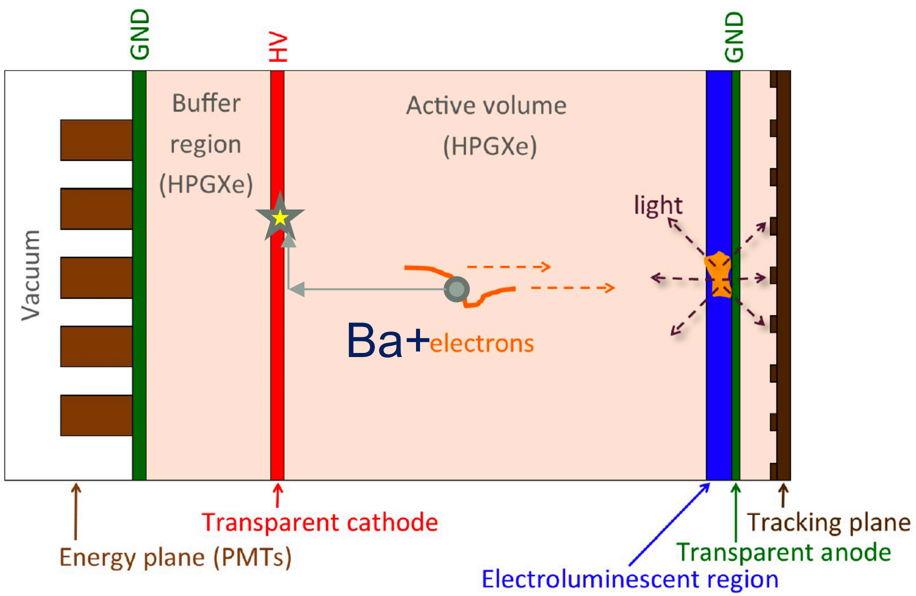
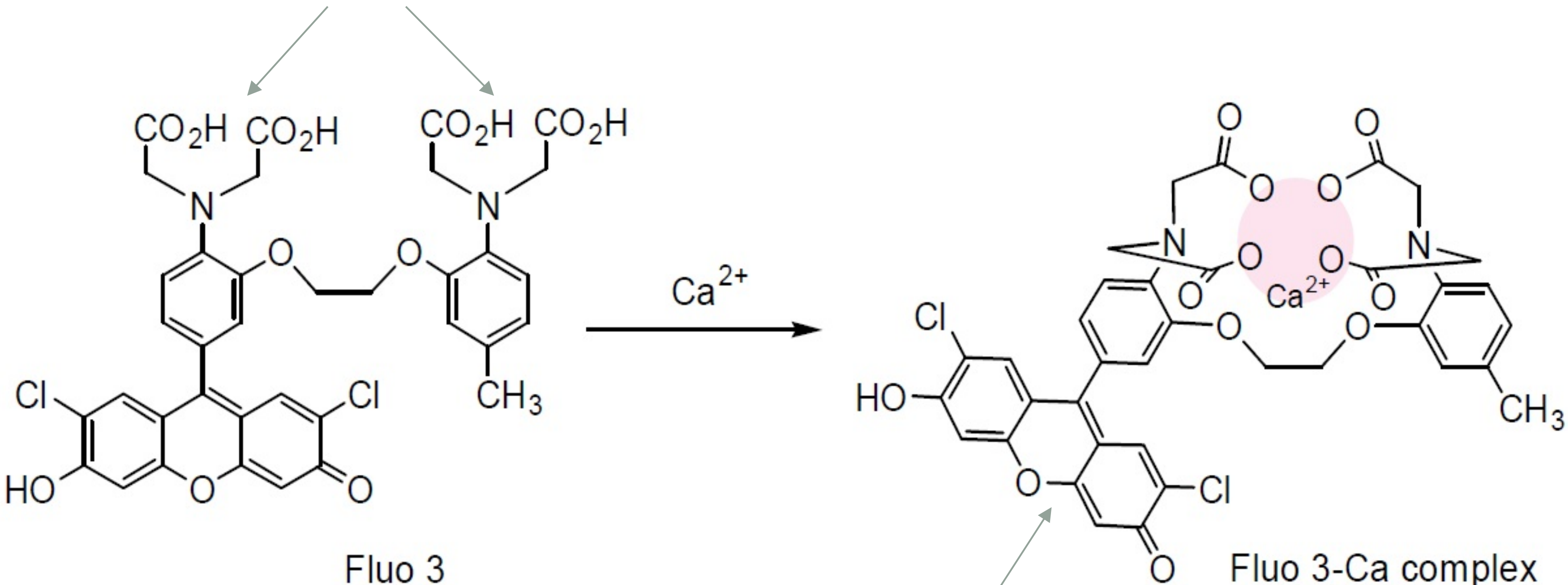


SUPPORTED BY:
U.S. DEPARTMENT OF ENERGY

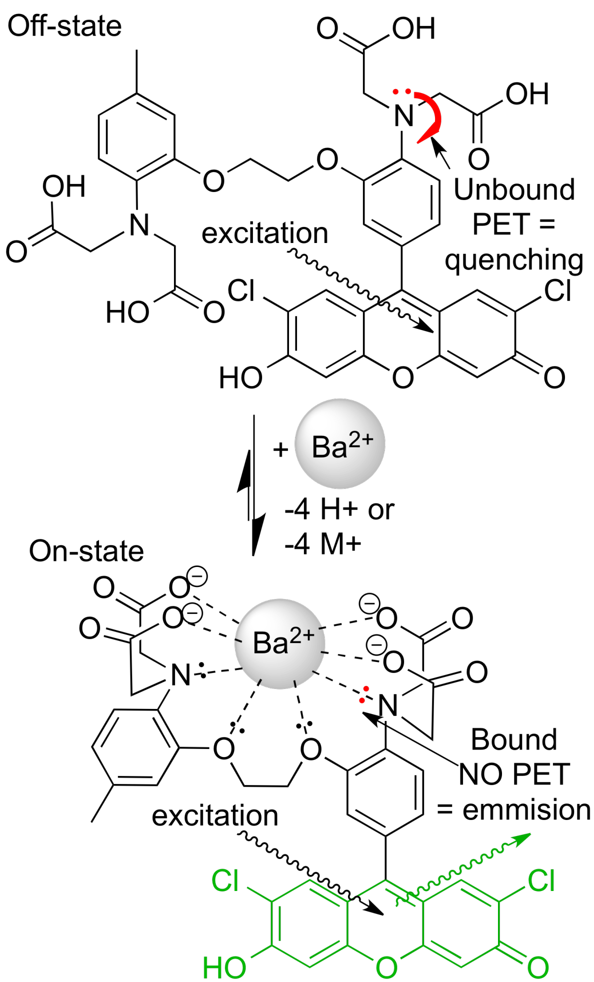
Issues with the FLUO family for dry sensing

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

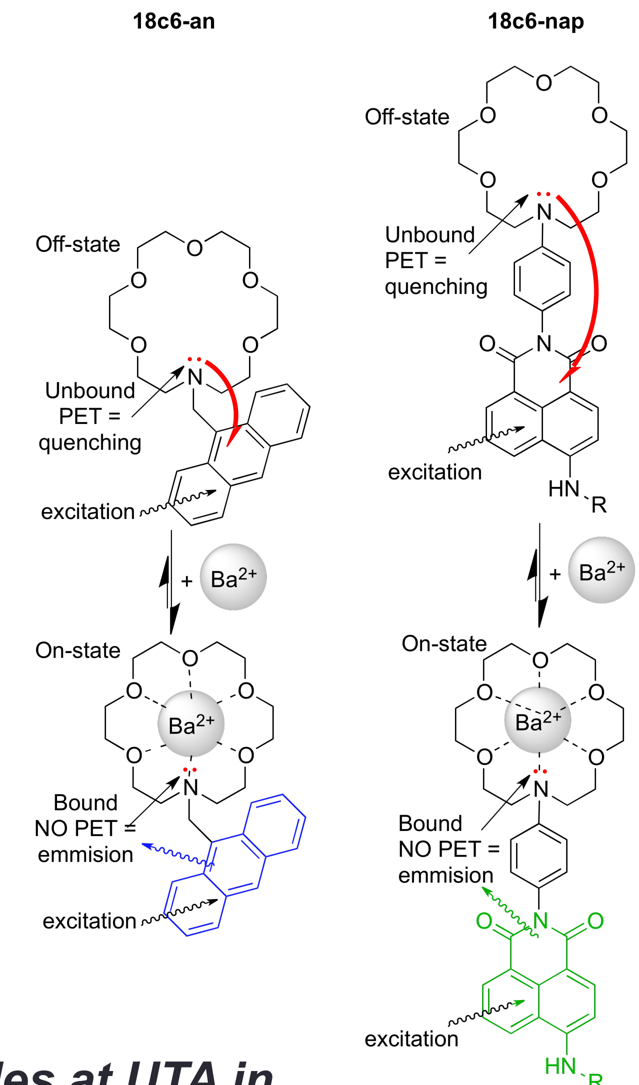


Fluorescein does not shine dry

Out with the old…

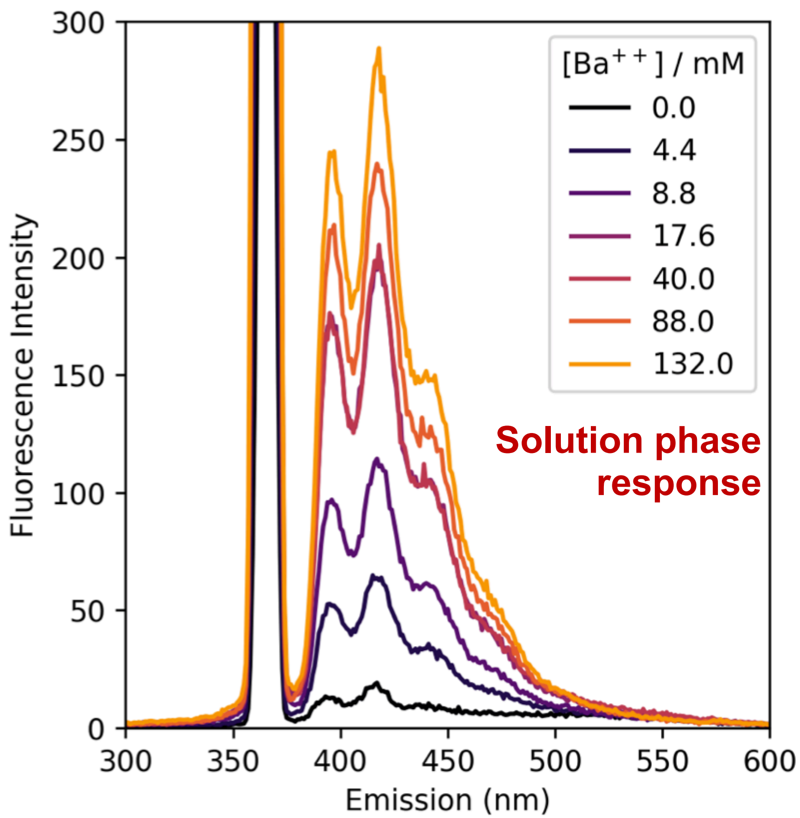


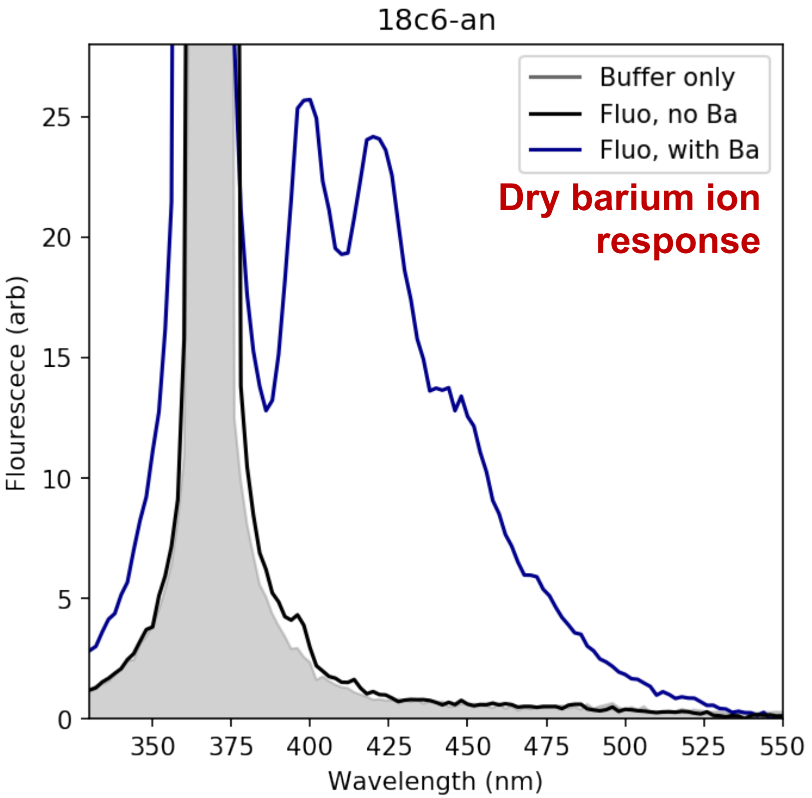
And in with the new!



Designing and making our own SMFI molecules at UTA in collaboration with Foss organic chemistry lab

Solving the dry ion sensing problem

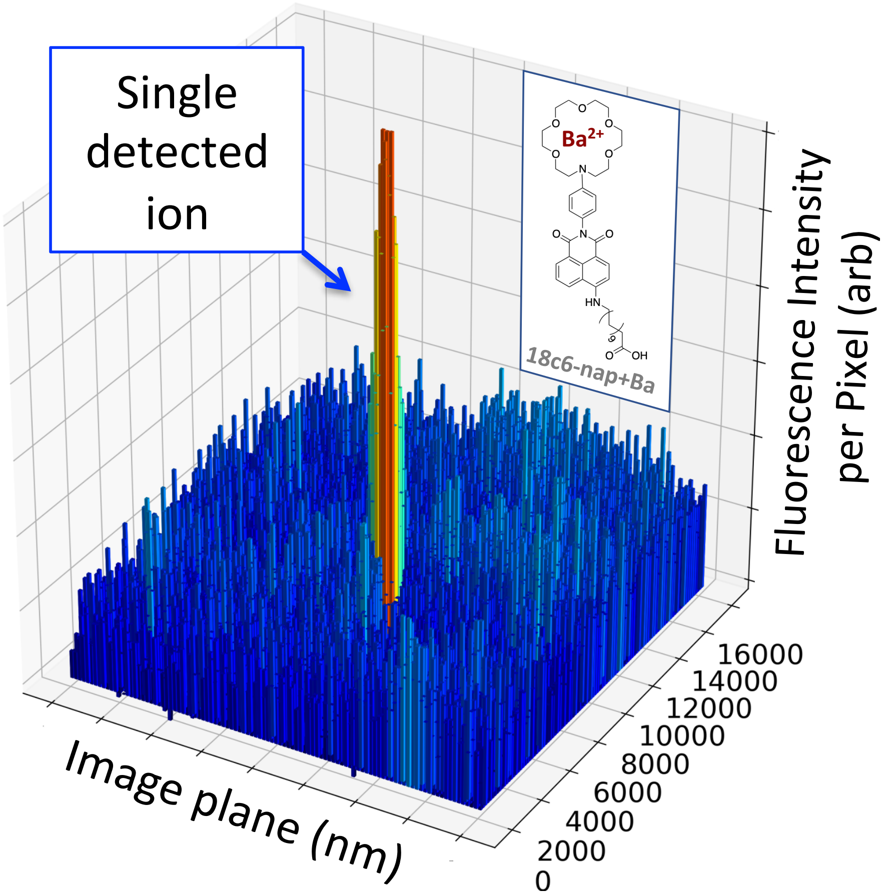




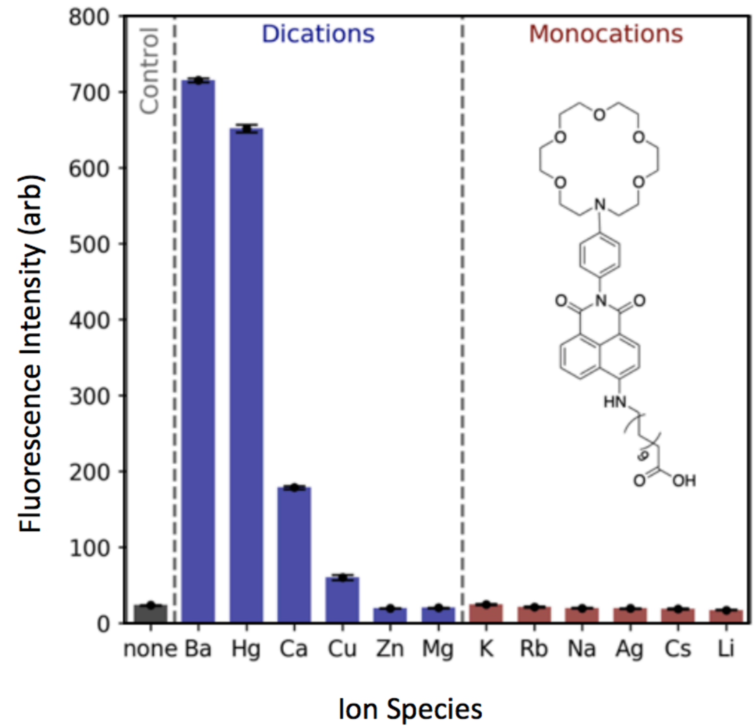
Nature Sci. Rep. 9, 15097 (2019)

Barium Chemosensors with Dry-Phase Fluorescence for Neutrinoless Double Beta Decay

P. Thapa[1,*], N. Byrnes[1], F. W. Foss Jr.[1], B. J. P. Jones[1], A. A. Denisenko[1], D. R. Nygren[1], A. D. McDonald[1], and K. Woodruff[1]



High selectivity to barium
Rejection of mono-cations mitigates possible Xe+ background



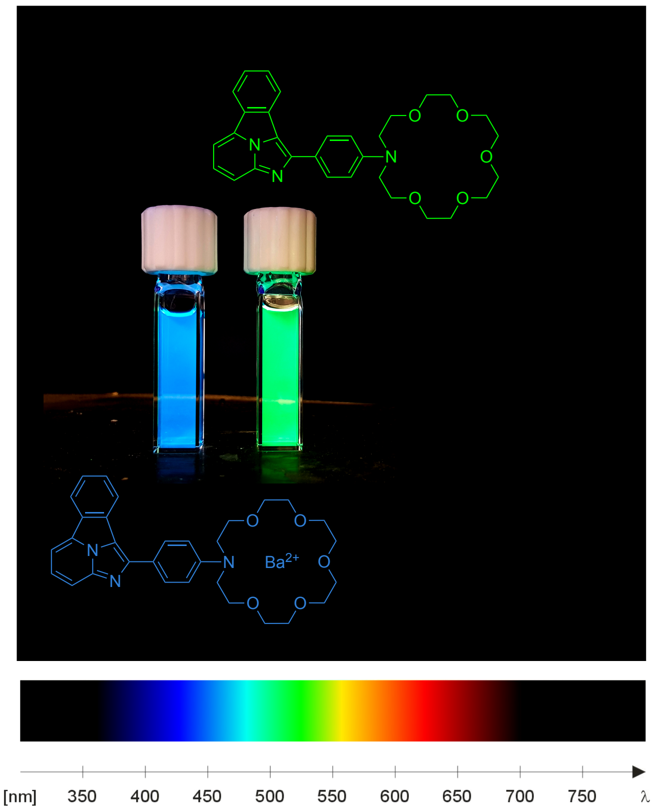
arXiv:1904.05901
APS DPF proceeding

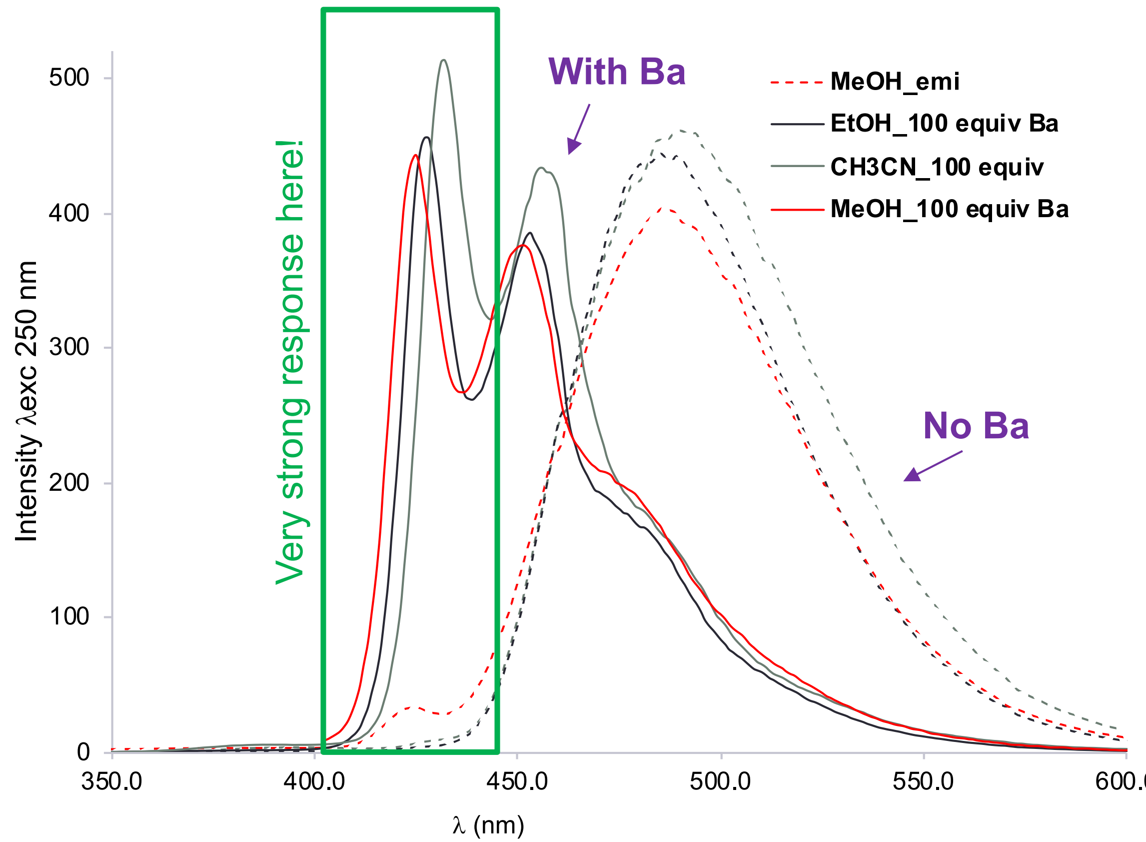
Barium Tagging with Selective, Dry-Functional, Single Molecule Sensitive On-Off Fluorophores for the NEXT Experiment

N.K. Byrnes[1], A. A. Denisenko, F.W. Foss Jr., B.J.P. Jones, A.D. McDonald, D.R. Nygren, P. Thapa, K. Woodruff for the NEXT Collaboration

Department of Physics, University of Texas at Arlington

An alternative path: color-changing fluorophores



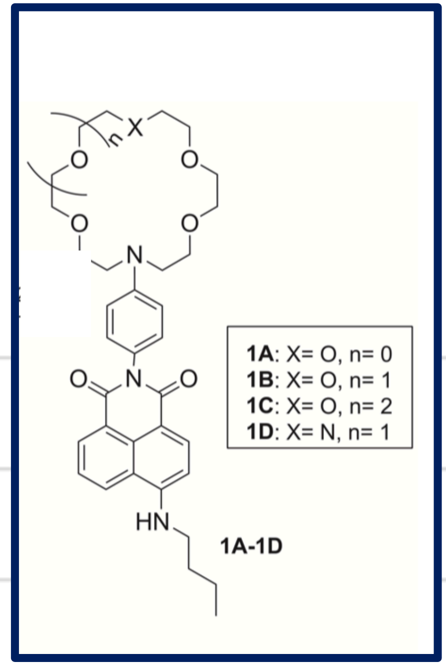


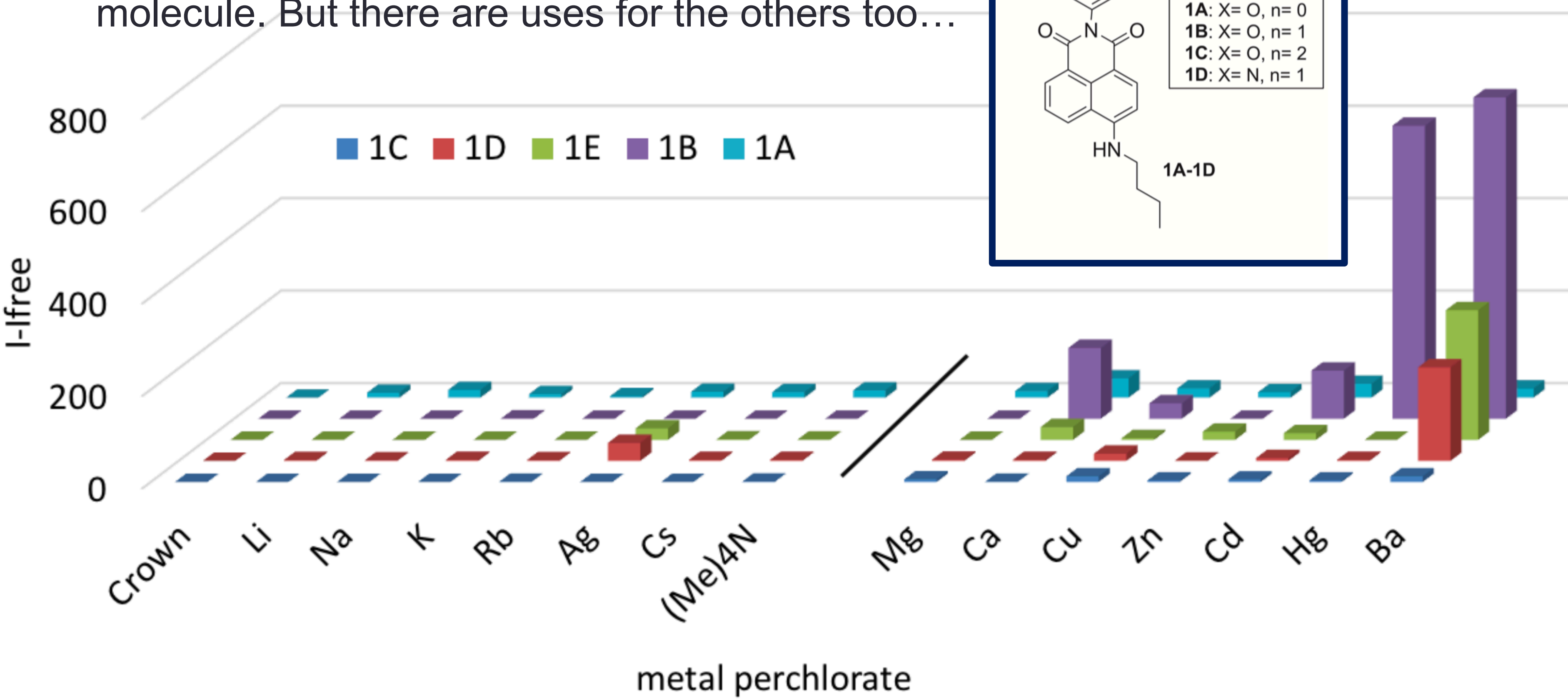
New ideas and new molecules from our collaborators at DIPC may open new and promising avenues !

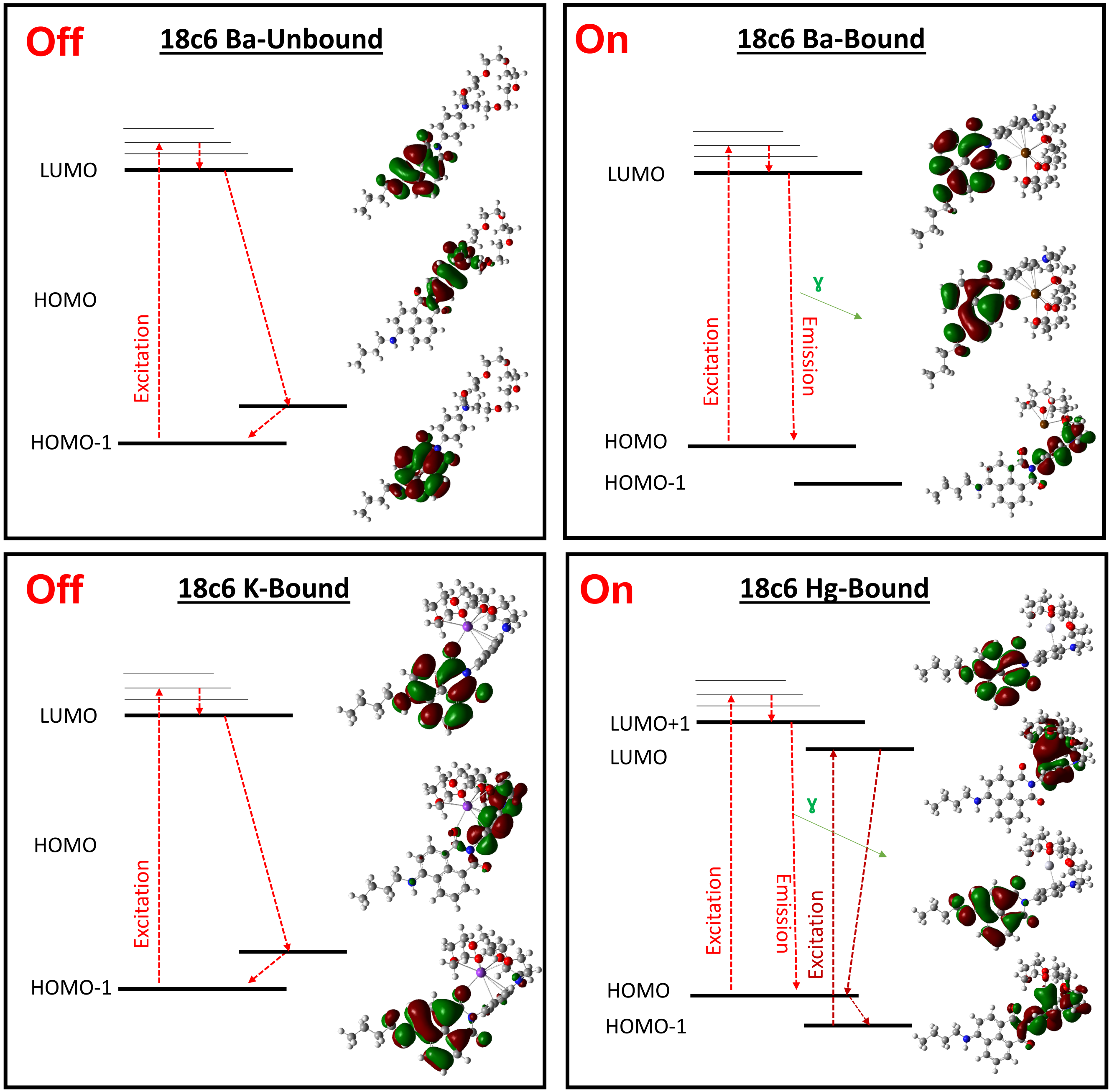
Tuning receptor selectivity

By changing the receptor we can adapt for selectivity to different ionic species.

Targeting an ultra-Barium sensitive, selective molecule. But there are uses for the others too…







From guesswork to precision molecular design

Computations with DFT and TDDFT now allow us to be predictive of which receptor / fluor combinations respond to which ions and why.

We are just starting to explore a wide space with this new tool.

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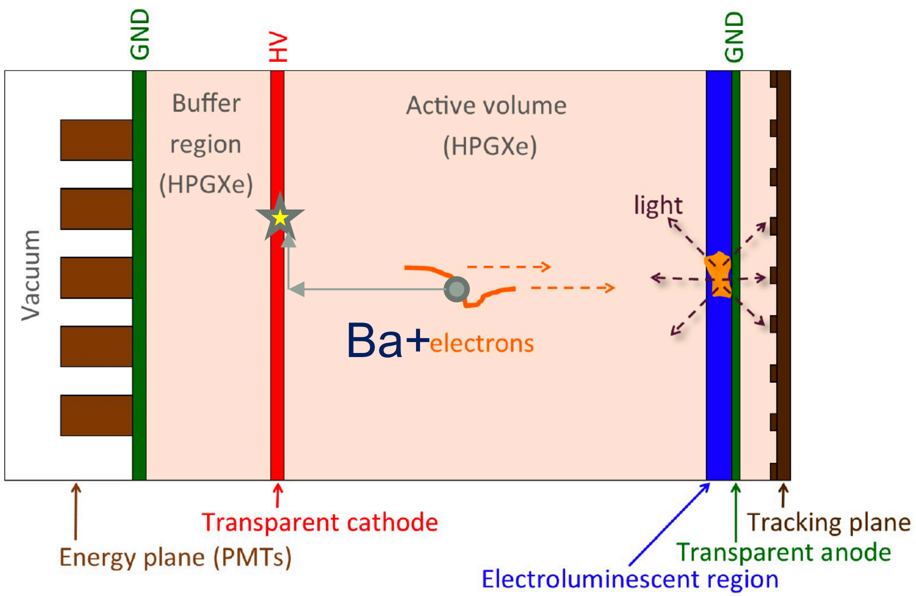
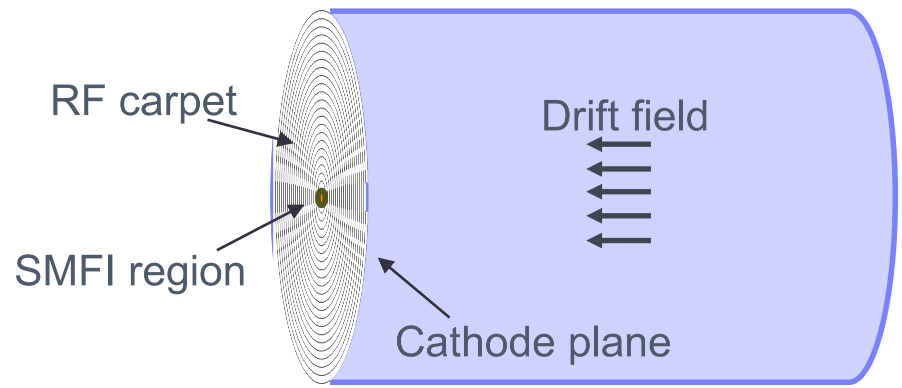
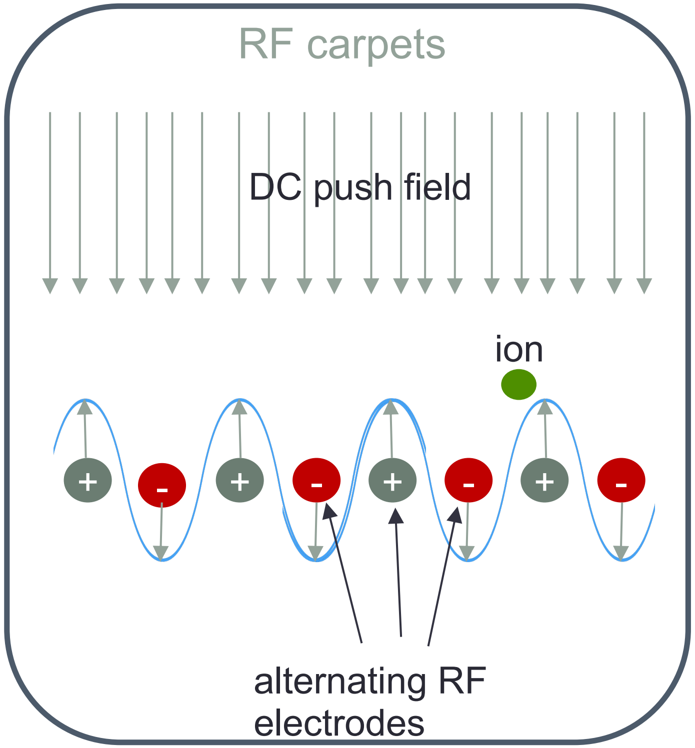


SUPPORTED BY:
U.S. DEPARTMENT OF ENERGY

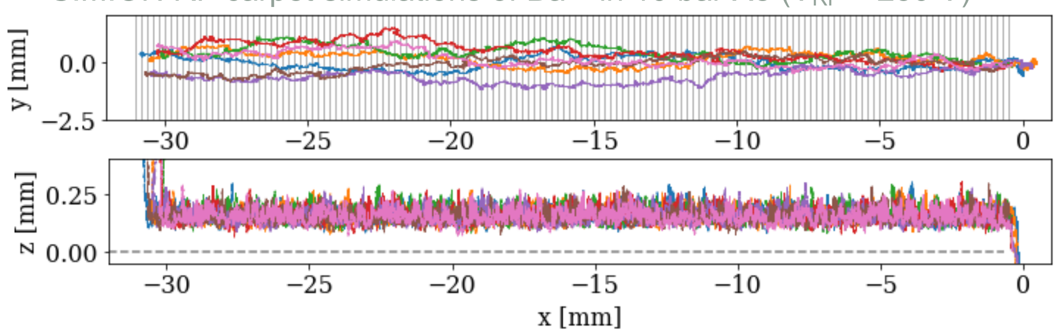
Ion transport using RF carpets



- Need to transport Ba^{++} ion to small (~cm^2) scan region
- RF carpets are being investigated for high pressure transport across the cathode
  - Commonly used for ion transport in low pressure helium
  - Have been shown to operate up to 300 mbar - NIM B: 376:221–224 (2016)
- Simulations suggest strong transmission in xenon gas can be achieved.



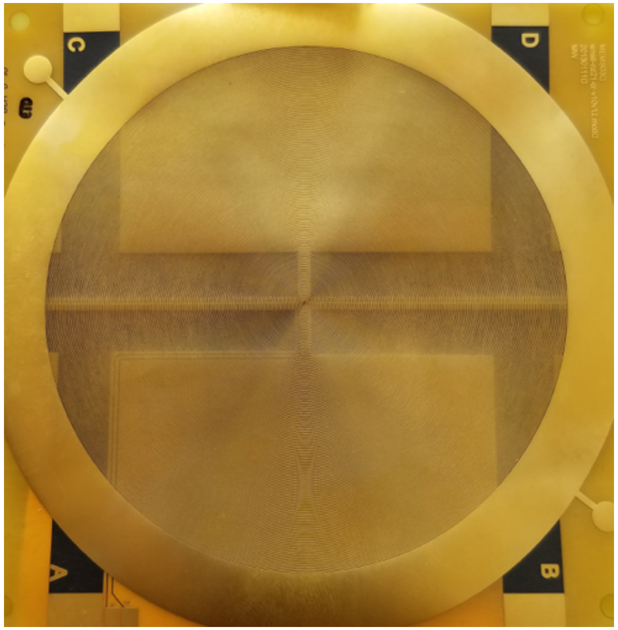
SIMION RF carpet simulations of Ba^{++} in 10 bar Xe (V_{RF} = 250 V)



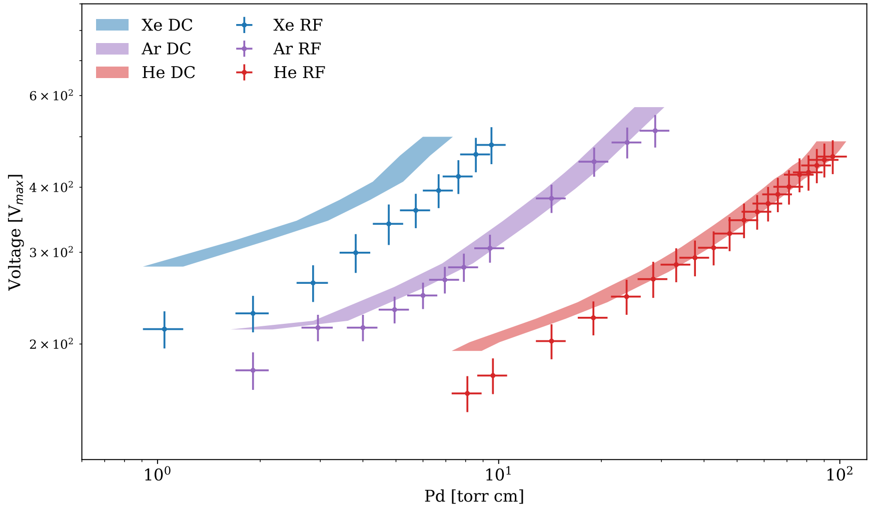
Ion transport using RF carpets

- Preliminary simulations and calculations show RF carpet transport is possible at 10 bar
  - Need high voltage and small inter-electrode distances
- Tested high voltage RF breakdown across electrodes in high pressure noble gases
- Presently setting up a test at ANL's CARIBU accelerator to demonstrate performance in xenon.

RF carpet for CARIBU tests



RF voltage breakdown in noble gases



Submitted to JINST

Radio Frequency and DC High Voltage Breakdown of High Pressure Helium, Argon, and Xenon

---

The NEXT Collaboration

K. Woodruff,[1,a] J. Baeza-Rubio,[1] D. Huerta,[1] B.J.P. Jones,[1] A.D. McDonald,[1] L. Norman,[1] D.R. Nygren,[1] C. Adams,[10] V. Álvarez,[17] L. Arazi,[6] I.J. Arnquist,[18] C.D.R Azevedo,[4]

Next steps: Making it work in gas

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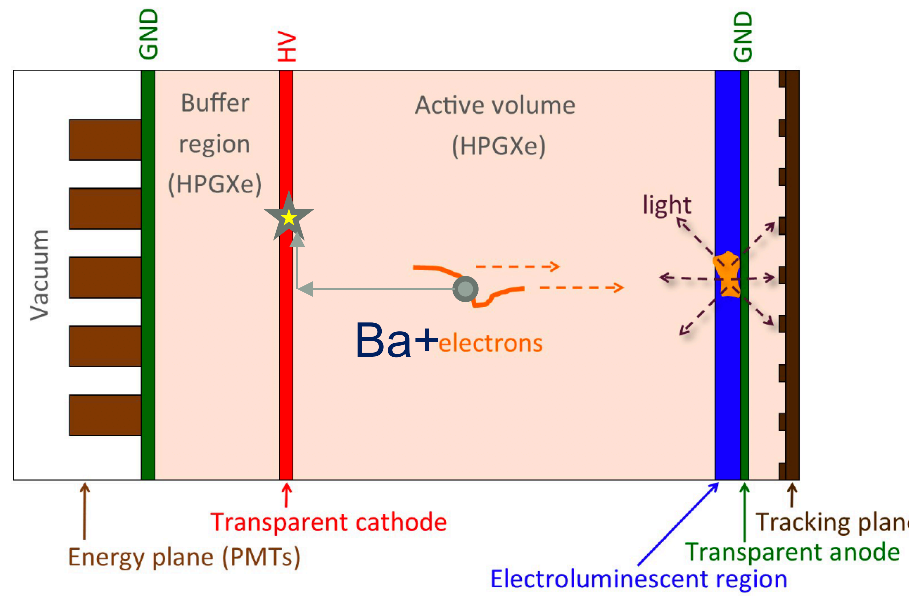
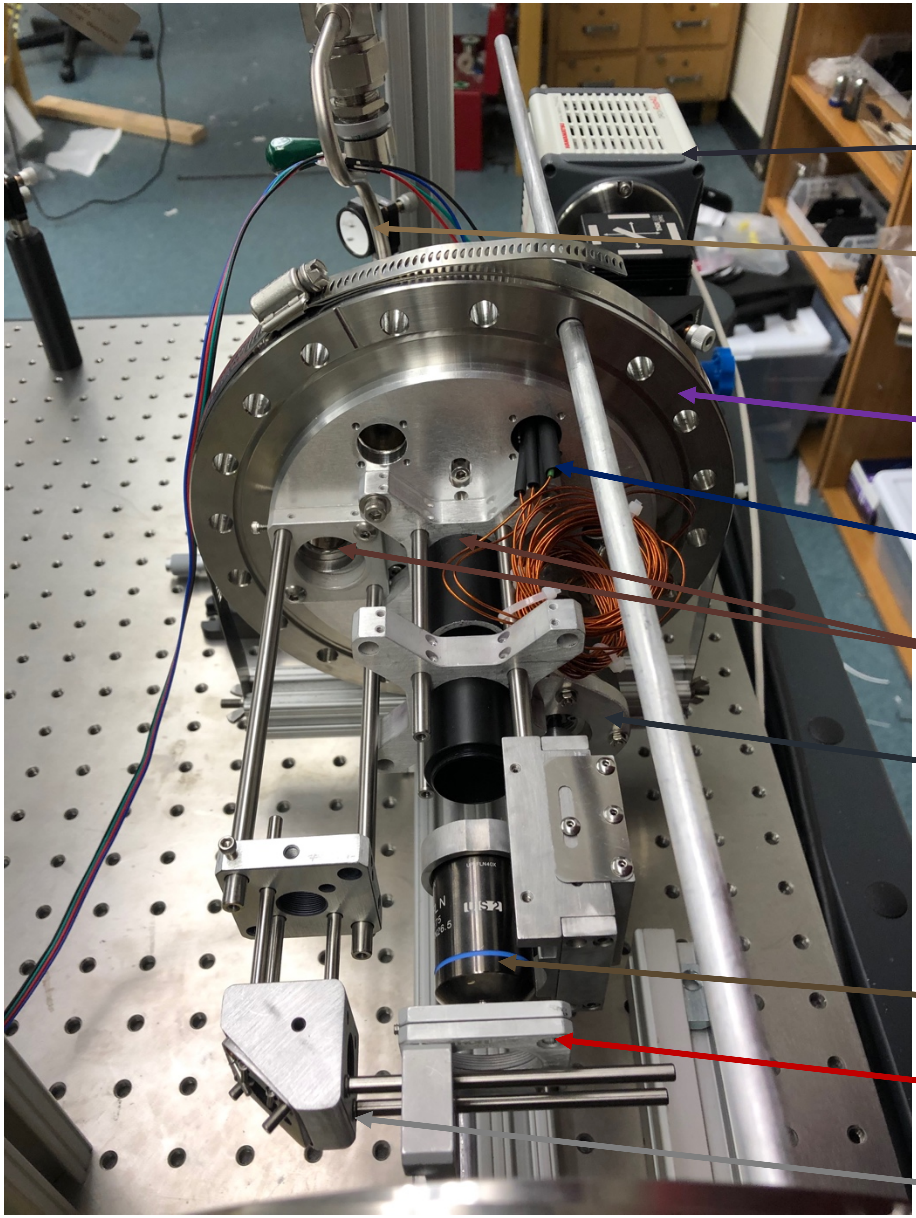
- **4. High Pressure Microscopy**
- 5. Combine into a working sensor for NEXT prototype



SUPPORTED BY:
U.S. DEPARTMENT OF ENERGY

The vacuum/pressure microscope



External imaging optics

Gas line on ¼” pipe feedthrough

8” CF flange

Pressure-rated 10pin feedthrough

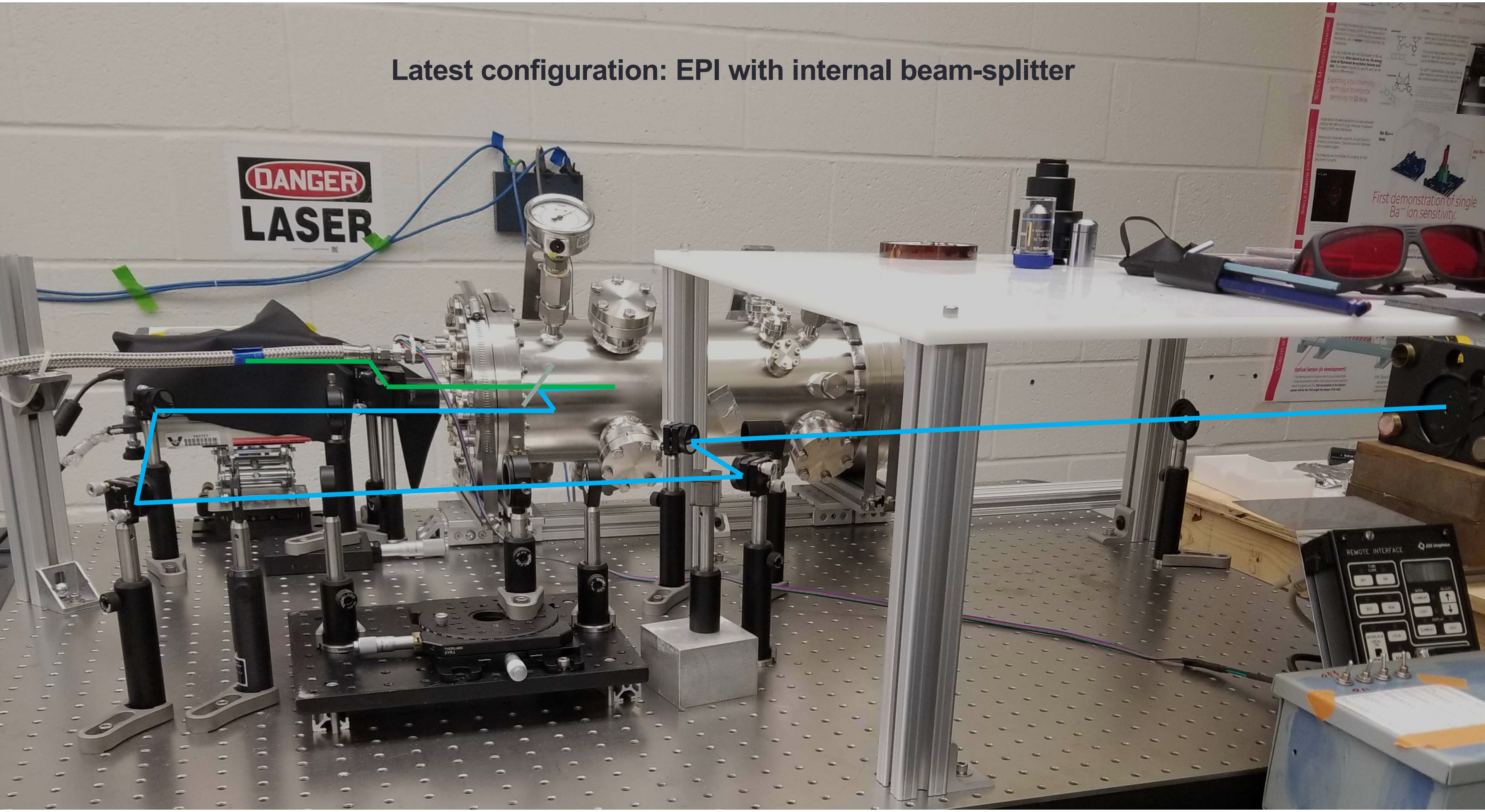
Sapphire optical windows (EM & EX)

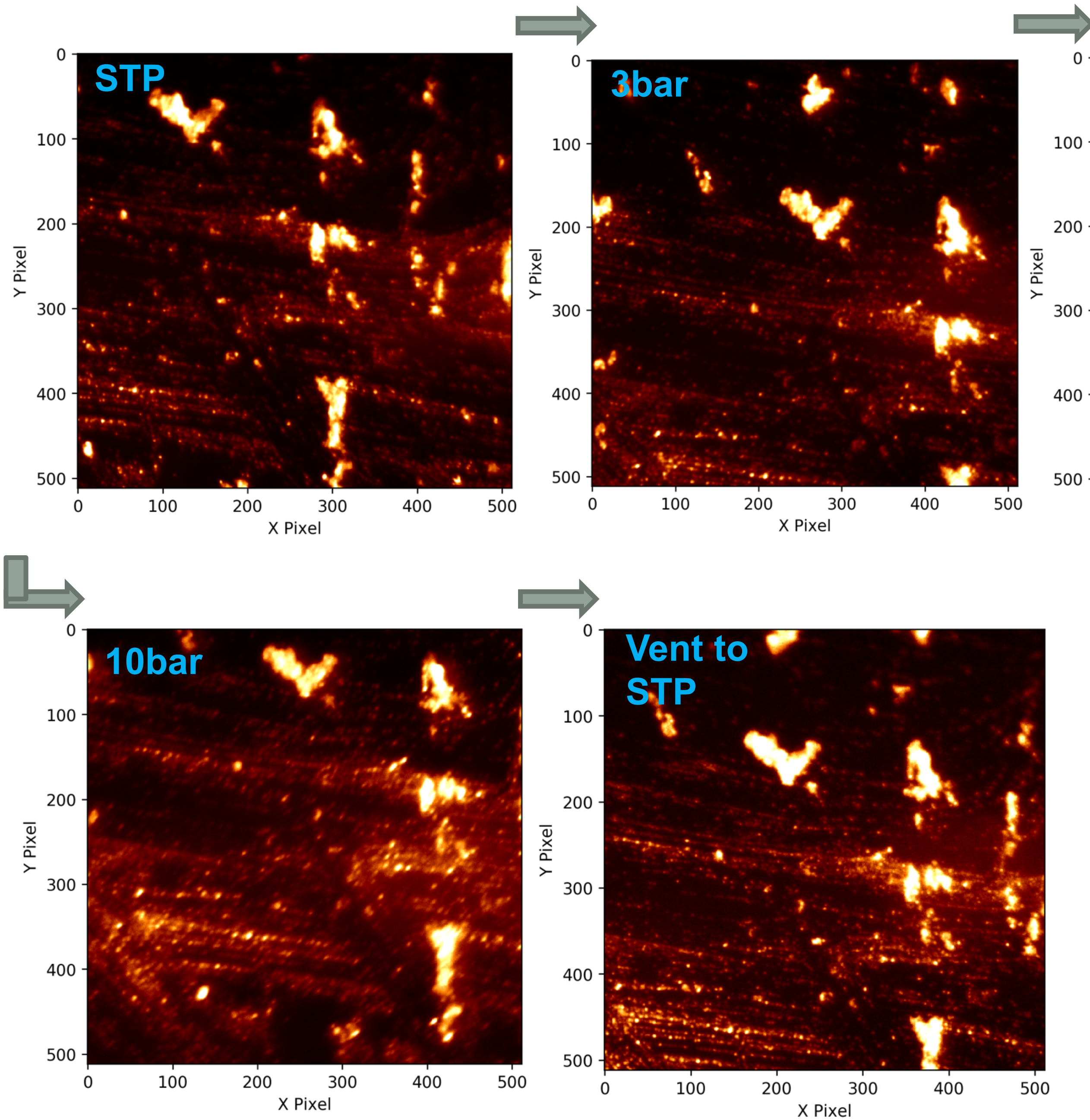
Vacuum stepper motor +
micrometer screw stage

Customized microscope objective

Slide holder

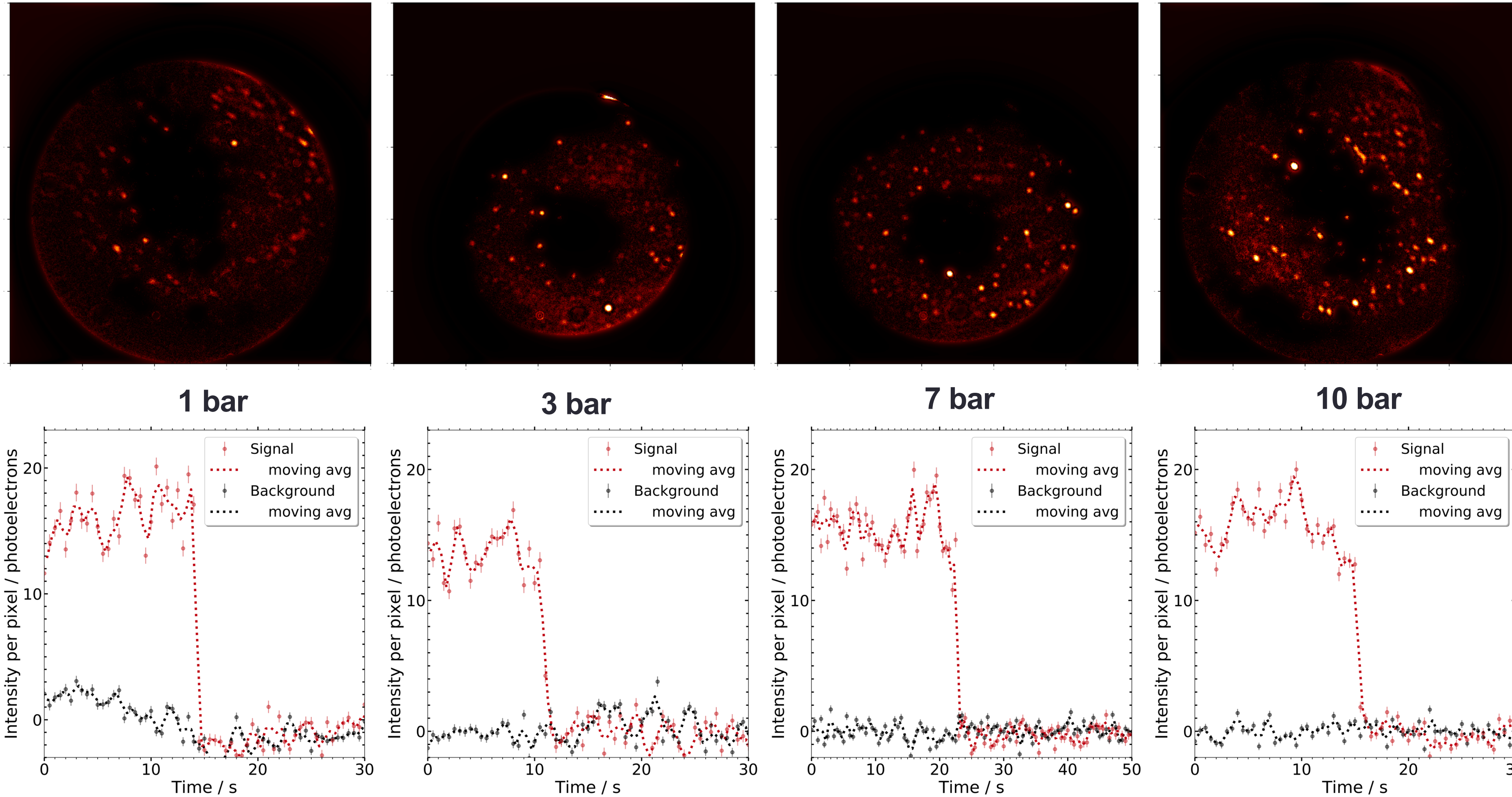
Internal 90-degree mirror


Latest configuration: EPI with internal beam-splitter
DANGER
LASER
First demonstration of single Ba⁺⁺ ion sensitivity
REMOTE INTERFACE



High pressure calibration images of fluorescent clusters – low mag test (20x)

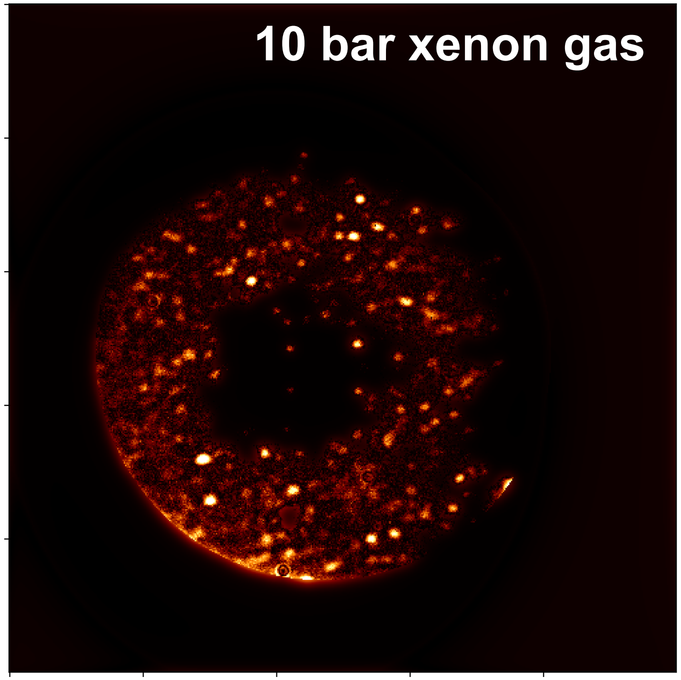
New Data from High Pressure, Single Ion Microscopy

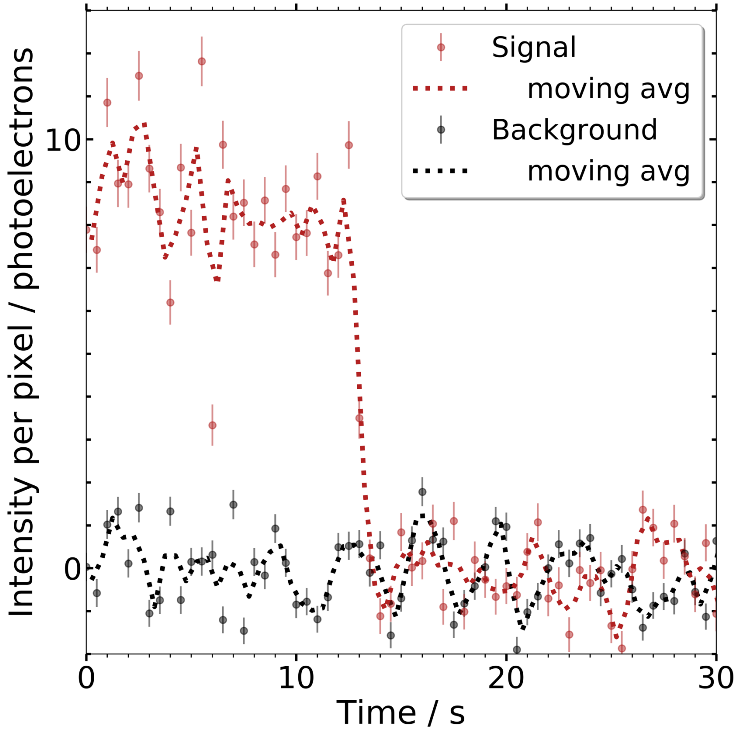


FIRST HIGH PRESSURE SINGLE MOLECULE FLOURESCENT IMAGES (ARGON GAS).

Single Ion Images in HPGXe

- **The first single ion image taken within the working medium of a neutrinoless double beta decay experiment.**

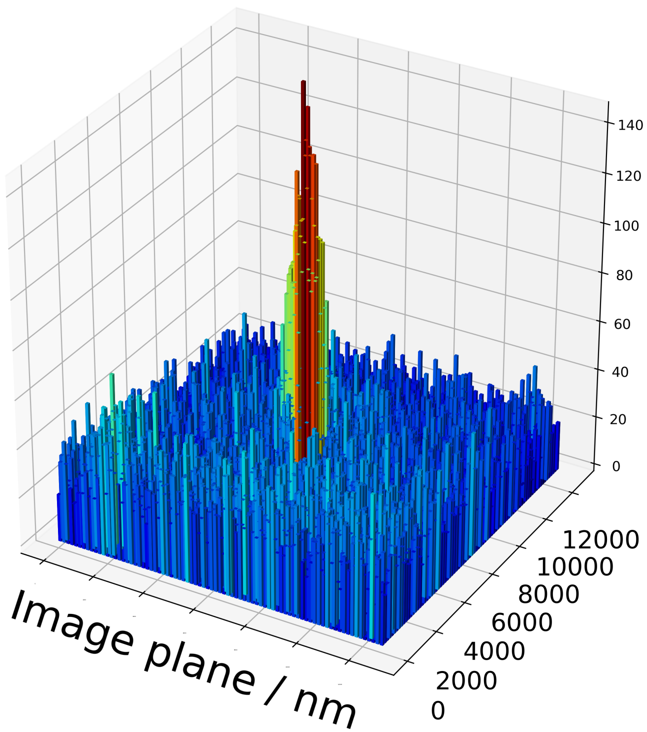




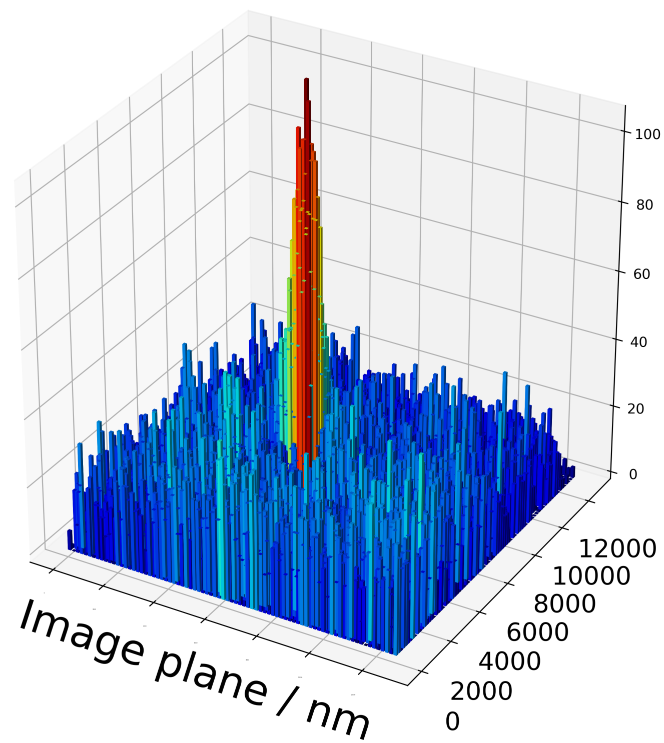
- **A critical advance for barium tagging, and *(we claim)* for neutrinoless double beta decay!**

First ever single molecule images in a double beta medium:

single barium ions in 10 bar argon and xenon



Ba^{++} ion / 10 bar xenon



Ba^{++} ion / 10 bar argon

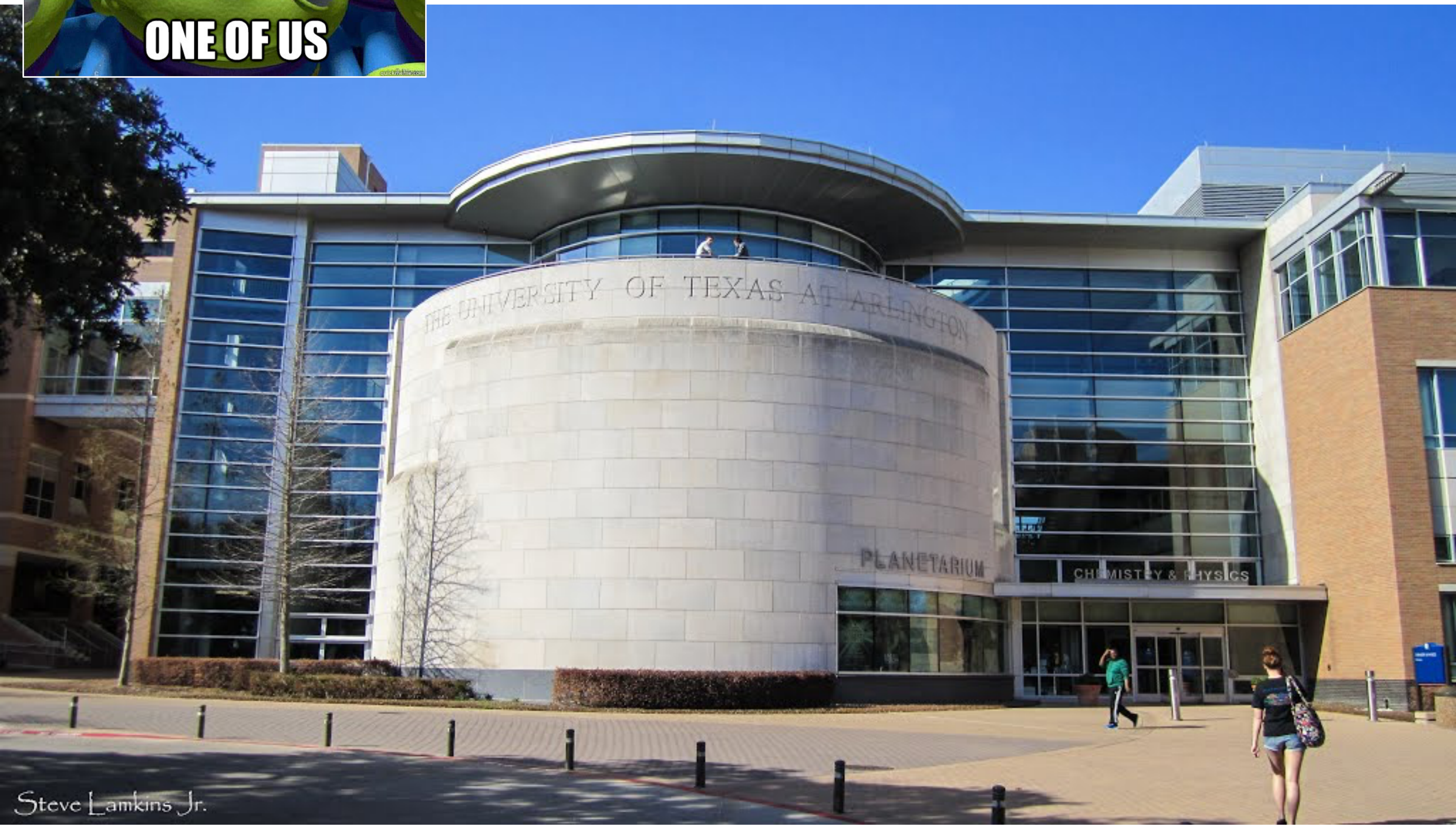
Publication in preparation

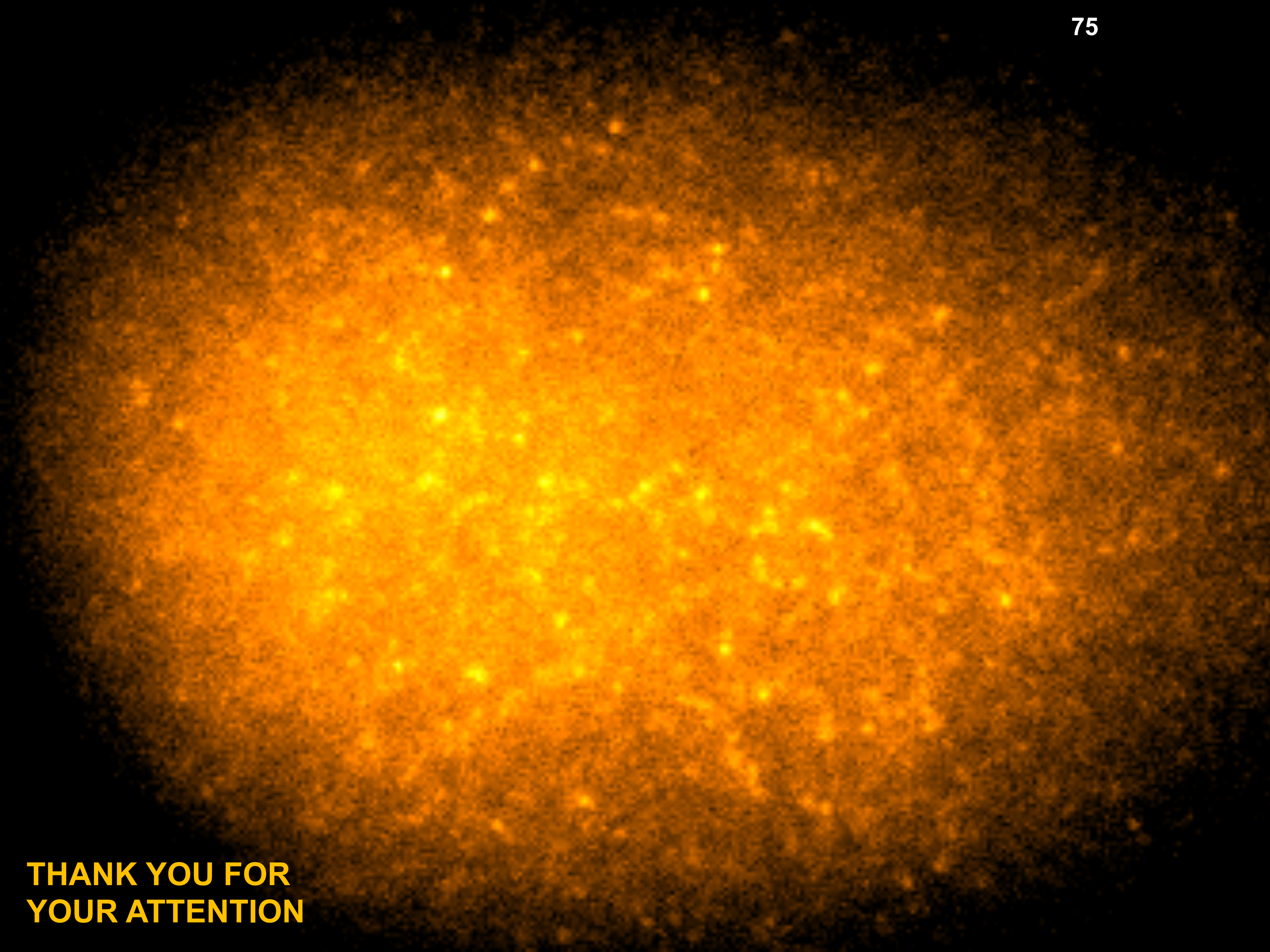
Summary

- High pressure xenon gas is an especially compelling technique for neutrinoless double beta decay, due to energy resolution, topology, scalability.
- Barium tagging may offer a major increase in sensitivity if it can be realized.
- Single ion imaging with SMFI shows great promise.
- Barium dications have been individually imaged in high pressure xenon gas environment
- Ion concentration onto sensors is under continuing development.
- Progress continues rapidly!

We are looking 1-2 a new graduate students to work with us on barium tagging – if you are one or know one, reach out!

UNIVERSITY OF TEXAS ARLINGTON




THANK YOU FOR
YOUR ATTENTION