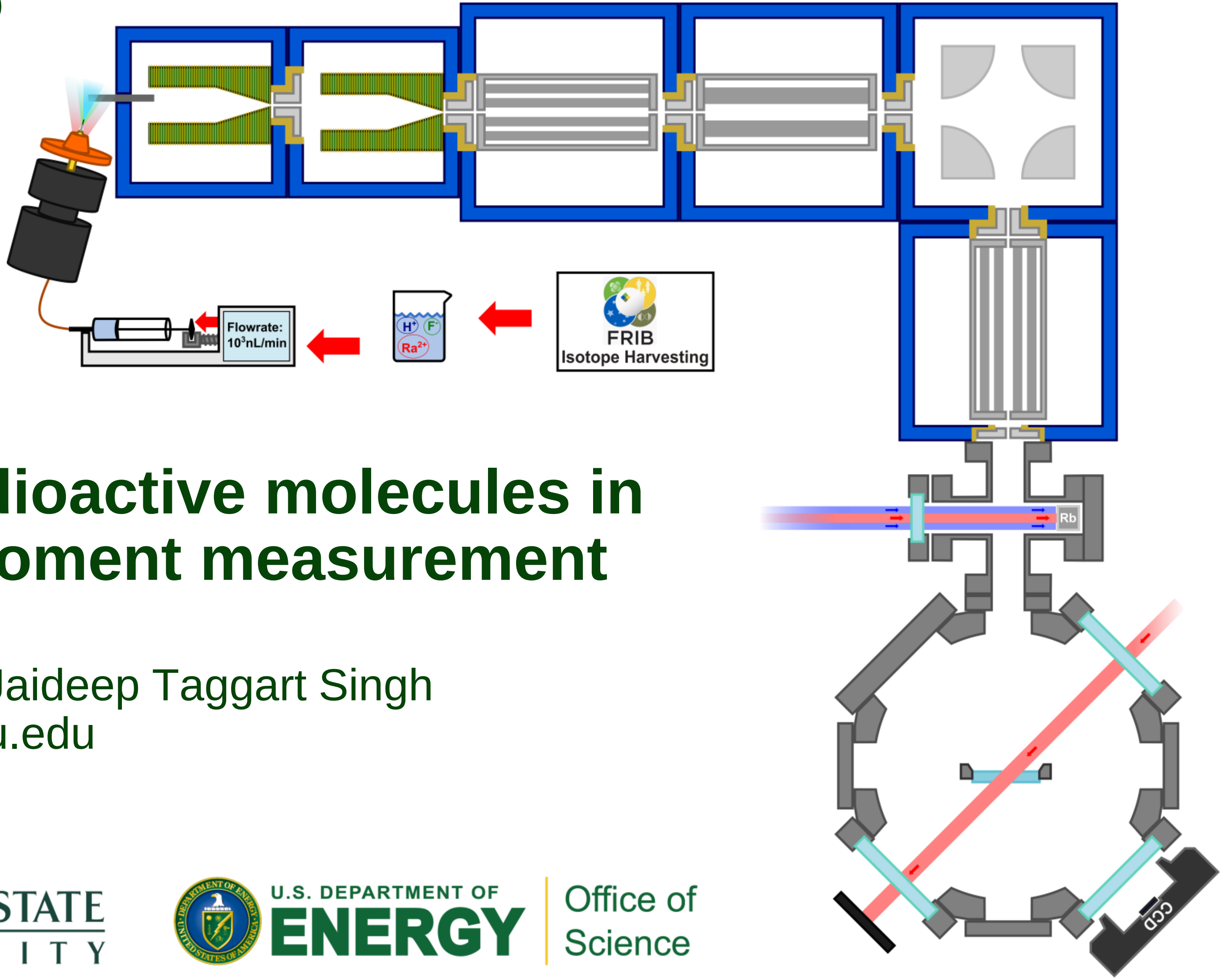
March 14, 14:40 – 15:20
Mini-symposium on Precision Measurements at Low Energies
Facility for Rare Isotope Beams, MSU, Lecture Hall 1200



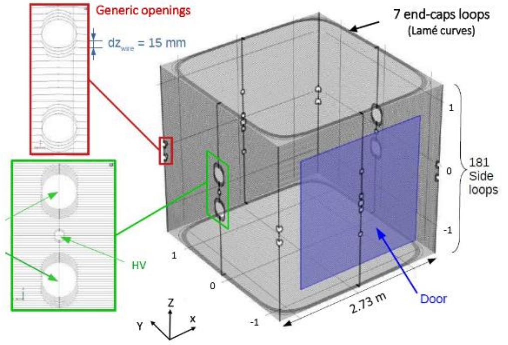
Progress towards forming radioactive molecules in solids for a nuclear Schiff moment measurement

Aiden Boyer*, Nicholas Nusgart, Jaideep Taggart Singh
*boyera@frib.msu.edu

This material is based upon work supported by the U.S. Department of Energy, Office of Science, Office of Nuclear Physics and used resources of the Facility for Rare Isotope Beams (FRIB) Operations, which is a DOE Office of Science User Facility under Award Number DE-SC0023633.
This work is supported by the U.S. Department of Energy, Office of Science, Office of High Energy Physics under Award Number DE-SC0022299
This work (EDM3) is supported by the U.S. Department of Energy, Office of Science, Office of Nuclear Physics under Award Number DE-SC0019015 and DE-SC0025679.

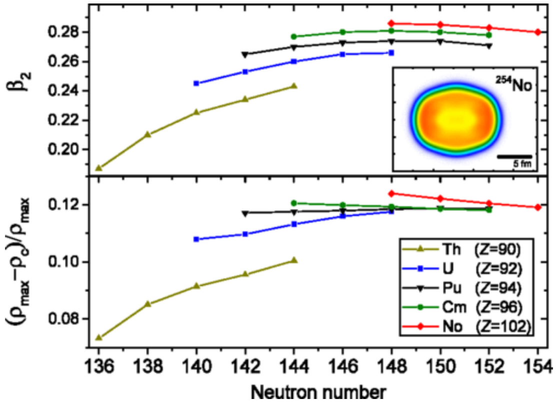
There Is a Lot of Exciting Work Ongoing in the Precision Frontier!

Neutron EDM: n2EDM @ PSI

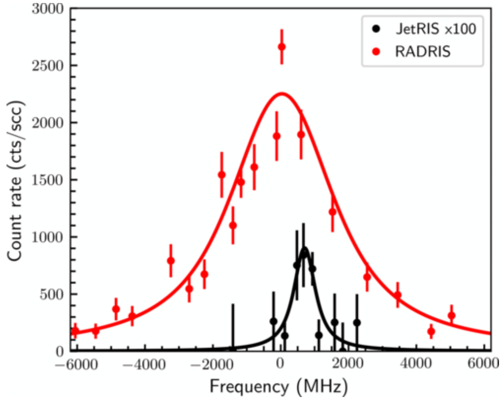


https://doi.org/10.48550/arXiv.2410.07914

Mass measurements and laser spectroscopy of superheavies

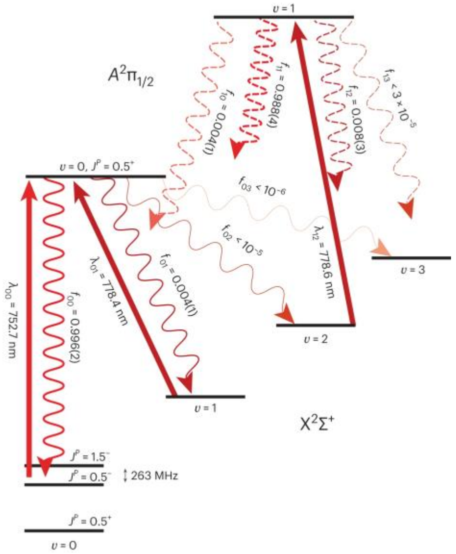


Raeder et al. PRL 120:232503 (2018)



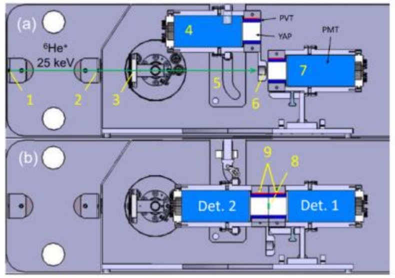
Lantis et al. PRR 6:023318 (2024)

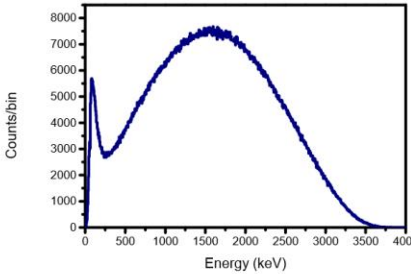
Spectroscopy of radium molecules (RaF)



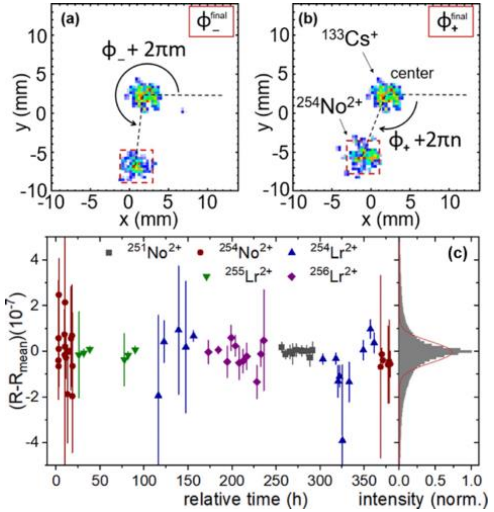
Udrescu et al. Nature Physics 20:202 (2024)

Tensor interaction search in nuclear beta decay of ^{6}He





Kanafani et al. EJP Web Conf. 282:01010 (2023)
Kanafani et al. PRC 106:045502 (2022)

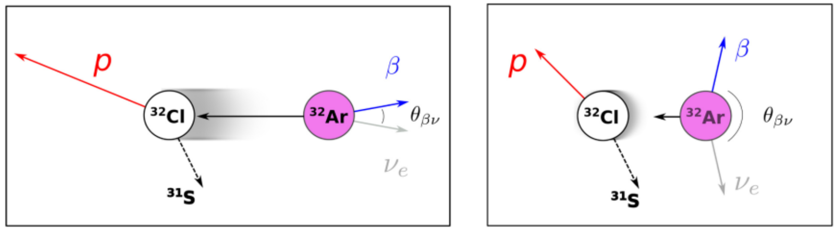


Kaleja et al. PRC 106:054325 (2022)

Decay recoil spectroscopy with superconducting quantum sensors

Bray et al. JLTP 218:74 (2025)
http://arxiv.org/abs/2411.08076

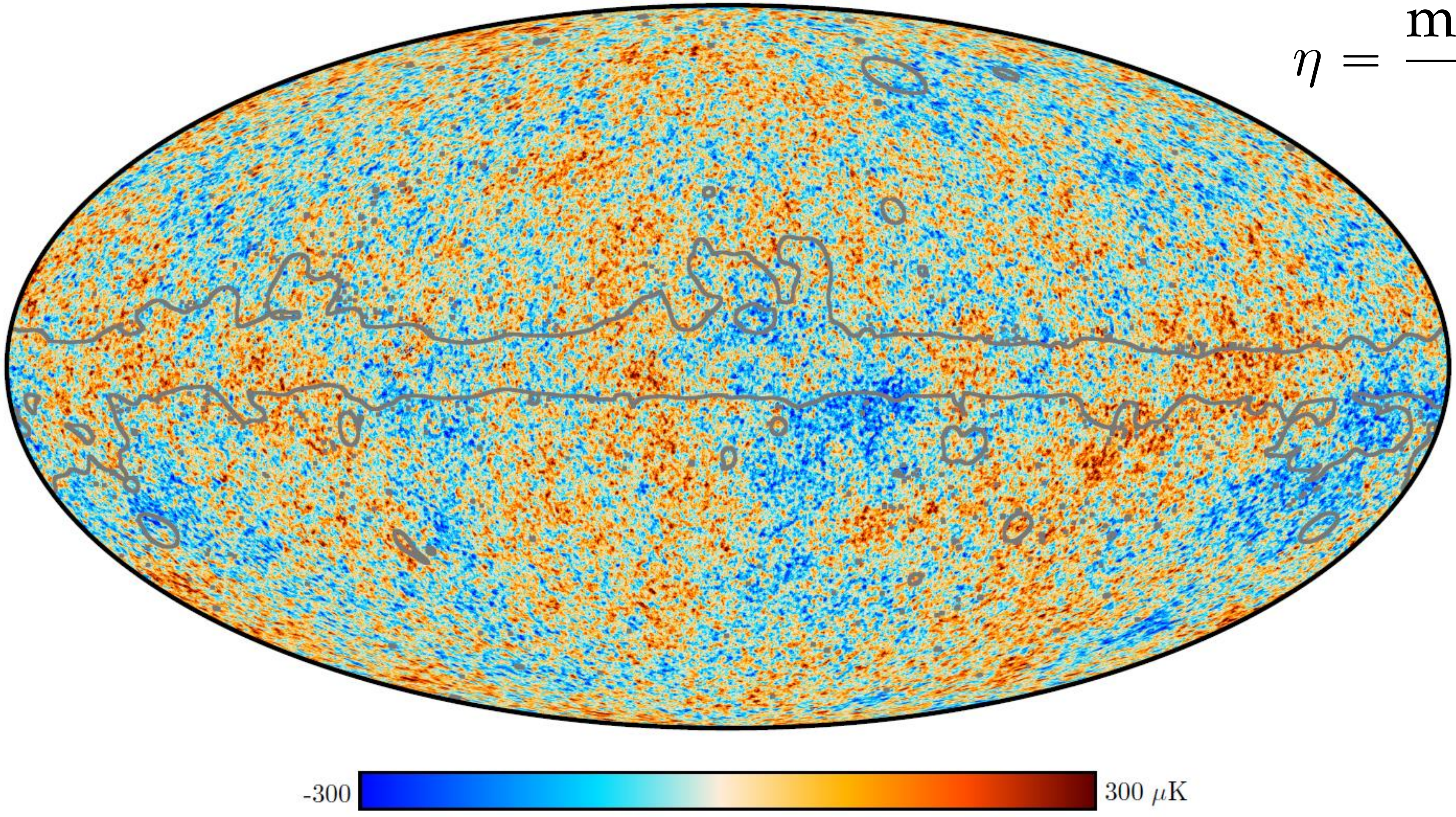
Scalar and tensor current searches with RIBs



(a) Vector interaction
(b) Scalar interaction

Atanasov et al. NIMA 1050:168159 (2023)

More Sources of CP Violation Needed To Explain Abundance Of Matter Over Antimatter In The Visible Universe



Planck (2018) https://www.cosmos.esa.int/web/planck/picture-gallery

$$\eta = \frac{\text{matter} - \text{antimatter}}{\text{relic photons}} \propto \sin(\delta)$$

$$\eta_{\text{exp}} \approx 10^{-9}$$

PDG2024

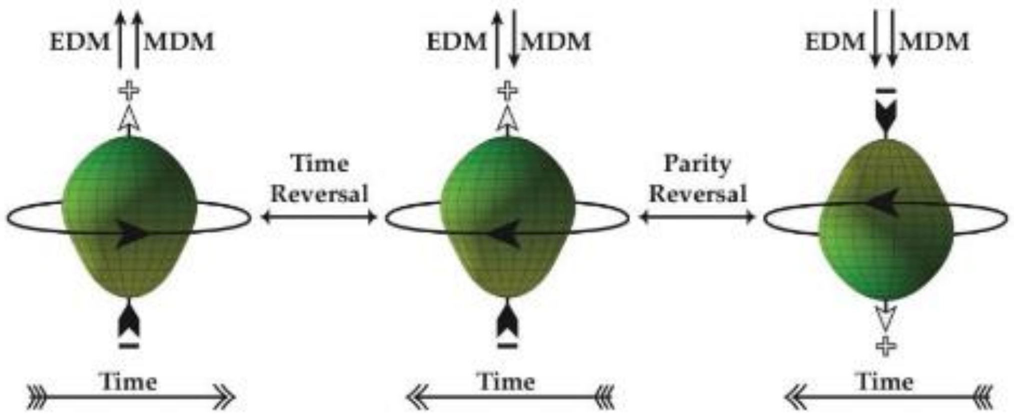
↕

$$\eta_{\text{CKM}} \approx 10^{-26}$$

Huet & Sather PRD 51:379 (1995)

Permanent Electric Dipole Moments Are A Signature Of T Violation

Quantity	P (Parity)	T (Time-reversal)
$\vec{J}$	Even (+)	Odd (-)
$\vec{B}$	Even (+)	Odd (-)
$\vec{E}$	Odd (-)	Even (+)
$\vec{J}\cdot\vec{B}$	Even (+)	Even (+)
$\vec{J}\cdot\vec{E}$	Odd (-)	Odd (-)



- EDMs measure a separation of charge

$$\vec{d} = \int \vec{r}\rho_Q d^3r = d\frac{\langle\vec{J}\rangle}{J}$$

$$\mathcal{H} = -\left(\vec{\mu}\cdot\vec{B} + \vec{d}\cdot\vec{E}\right) = -\frac{(\mu\vec{J}\cdot\vec{B} + d\vec{J}\cdot\vec{E})}{J}$$

J	Total angular momentum
B	Magnetic field
E	Electric field
d	Electric dipole moment
μ	Magnetic dipole moment
ρ_Q	Charge Distribution

CPT Theorem: T-Violation = CP-Violation

Experiments With Different Test Systems Are Sensitive to Different Sources of CP Violation

System	Best Limit (95%) 1E-28 *e* cm	SM estimate 1E-28 *e* cm Chupp et al. *RMP* 91:015001 (2019)	Method (Location)
Neutron	220	$\sim 10^{-4}$	ultracold neutrons in a bottle (PSI) Abel et al. *PRL* 124:081803 (2020)
“Electron”	0.11	$\sim 10^{-10}$	cold ThO beam (Chicago/Harvard/Northwestern) Andreev et al. *Nature* 562:7727 (2018)
	0.05		trapped HfF^+ (JILA/Boulder) Roussy et. al. *Science* 381:6653 (2023)
Hg-199	0.074	$\sim 10^{-6}$	atoms in vapor cell (UW-Seattle) Graner, et al. *PRL* 116:119901 (2017)

Current limits for permanent EDMs are free of SM backgrounds

Different Particles Are Sensitive To Different CP-Violating Parameters

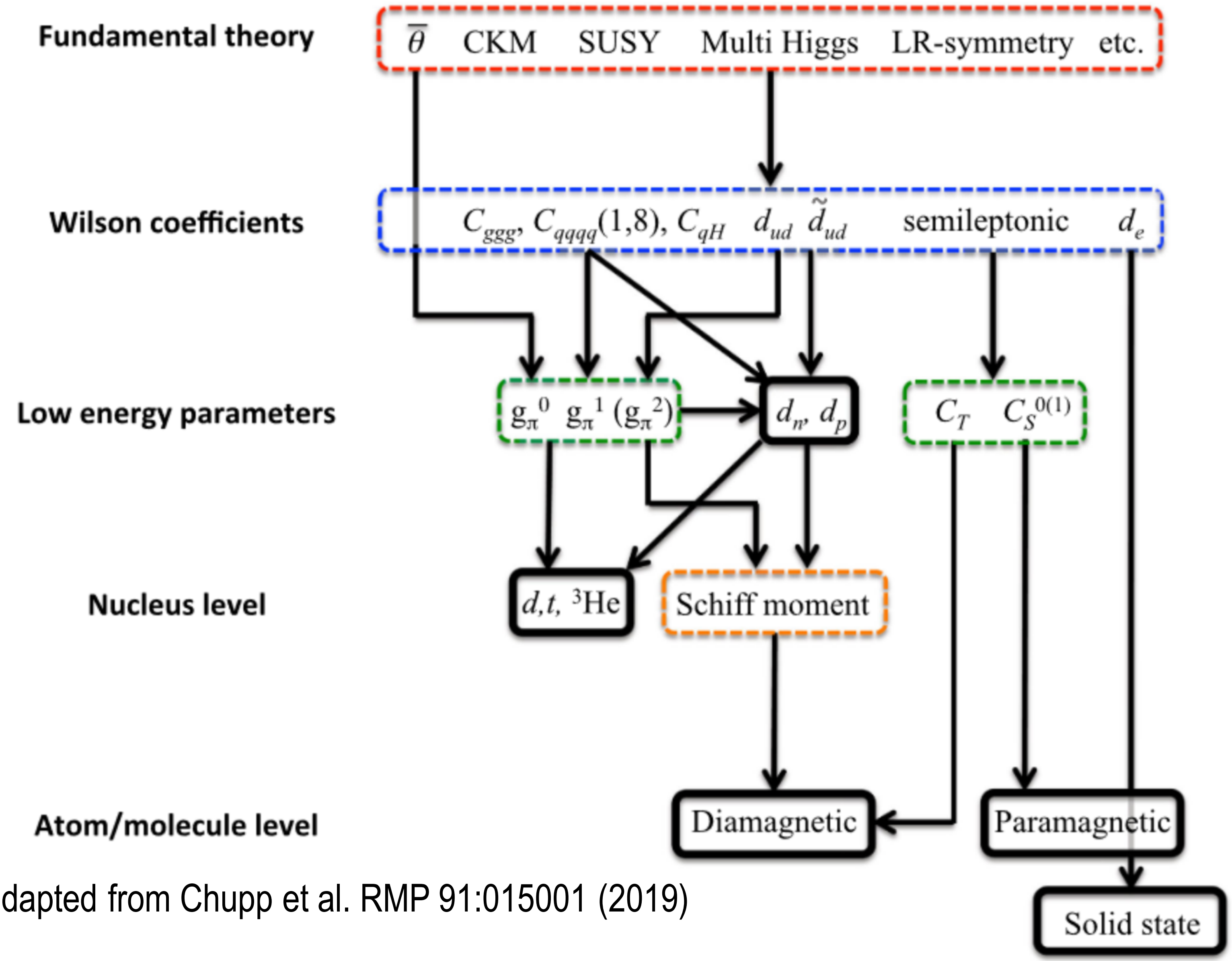


Figure adapted from Chupp et al. RMP 91:015001 (2019)

CP-violating
SM + BSM Lagrangian

Reduction to
effective-field-theory

Nuclear and atomic theory

Nuclear, atomic and
molecular experiments

Legend

Dominated by theory	🟥 🟦 🟩 🟧
Object of current or future experiments	▬

CP-Violating Observable in Diamagnetic Atoms: Nuclear Schiff Moment Example: Enhanced Sensitivity with Pear-Shaped Radium-225

$$S_z = \frac{\langle e r^2 z \rangle}{10} - \frac{\langle r^2 \rangle \langle e z \rangle}{6}$$

$$S \equiv \langle \Psi_0 | S_z | \Psi_0 \rangle = \sum_{k \neq 0} \frac{\langle \Psi_0 | S_z | \Psi_k \rangle \langle \Psi_k | V_{\cancel{PT}} | \Psi_0 \rangle}{E_0 - E_k} + \text{c.c.}$$

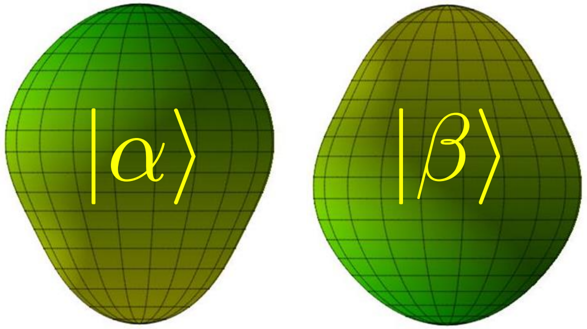
- **Nearly degenerate parity doublet**

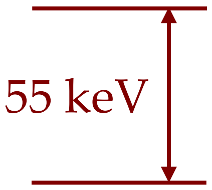
  Haxton & Henley PRL 51:1937 (1983)

- **Large intrinsic Schiff moment due to octupole deformation**

  Auerbach, Flambaum, & Spevak PRL 76:4316 (1996)

Ex: ^{225}Ra Parity Doublet





$$|\Psi_1\rangle = \frac{|\alpha\rangle - |\beta\rangle}{\sqrt{2}}$$

$$|\Psi_0\rangle = \frac{|\alpha\rangle + |\beta\rangle}{\sqrt{2}}$$

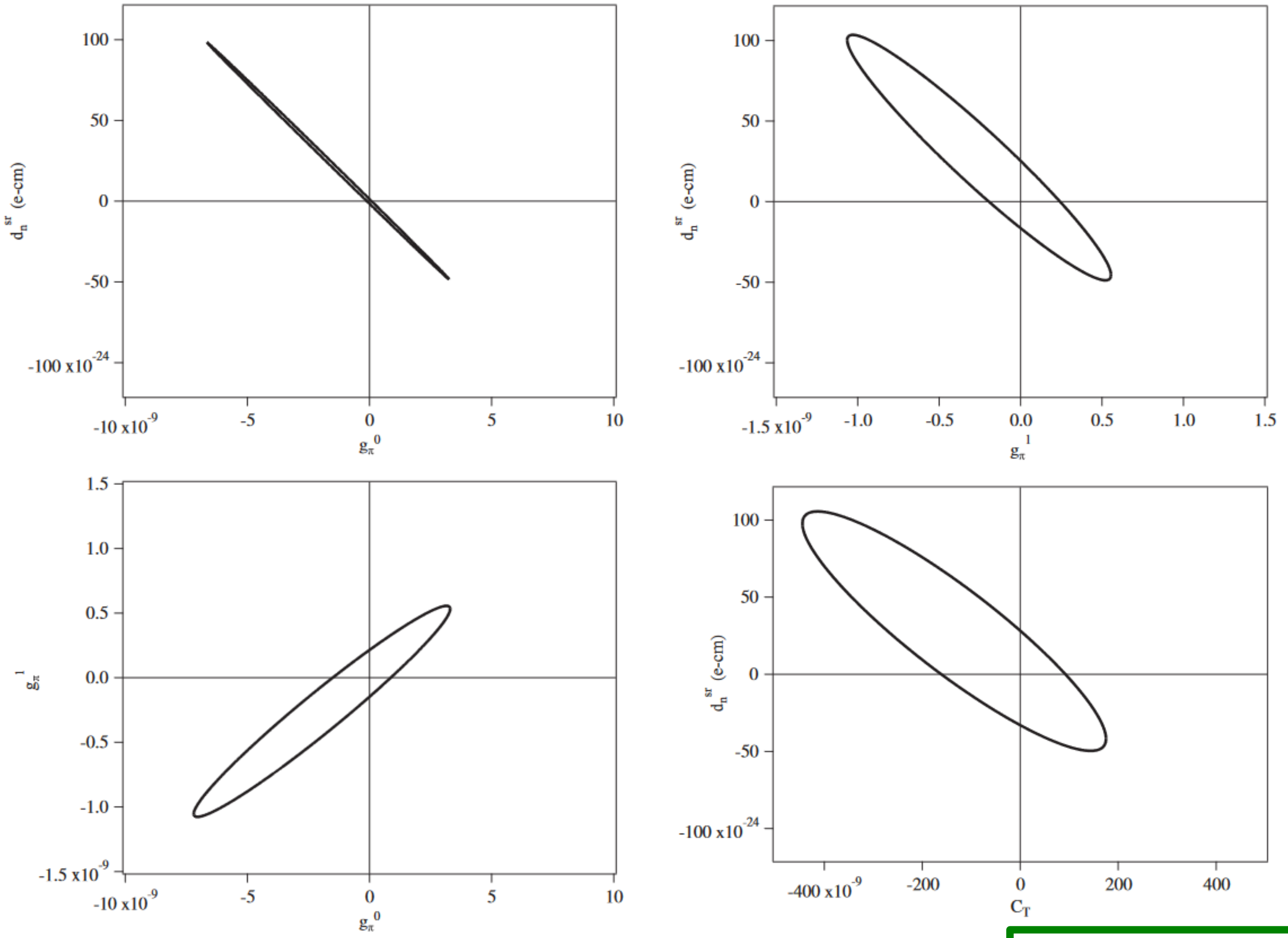
Total Enhancement Factor: EDM (^{225}Ra) / EDM (^{199}Hg)

Skyrme Model	Isoscalar	Isovector
SIII	300	4000
SkM*	300	2000
SLy4	700	9000

^{225}Ra: Dobaczewski & Engel PRL 94:232502 (2005)
^{199}Hg: Ban et al. PRC 82:015501 (2010)

Global Analysis Framework:
The Crucial Importance of Neutron EDM, CeNTREX, NSM, and Hg-199

Chupp et al. RMP 91:015001 (2019)



Four hadronic parameters:

$$d_n^{sr}\,,\ C_T\,,\ \bar{g}_\pi^{(0)}\,,\ \bar{g}_\pi^{(1)}\,,$$

Require at least four experiments to constrain:

1. **Neutron EDM**
2. **Atomic ^{199}Hg EDM**
3. **Pear-shaped NSM**
4. **^{205}Tl (CeNTREX)**

Stéphanie Roccia
Thursday March 13
11:00 – 11:40

Alex Frenett
Thursday March 13
14:40 – 15:20

Gordon Arrowsmith-Kron
Friday March 14
11:00 – 11:40

Aiden Boyer
Right Now!
14:40 – 15:20

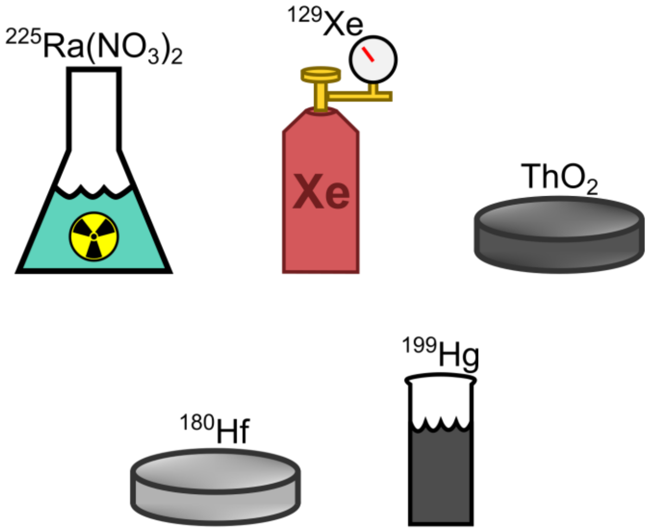
Sebastian Miki-Silva
Friday March 14
16:40 – 17:20

Nicholas Nusgart
Saturday March 16
09:40 – 10:20

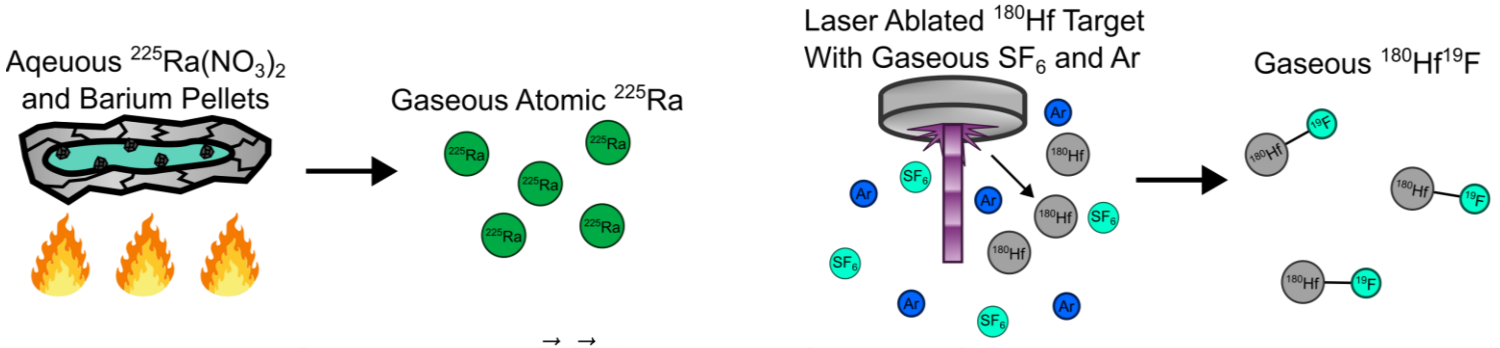
Jaideep Taggart Singh
Saturday March 16
14:00 – 14:40

The Generalized Recipe for an EDM Measurement

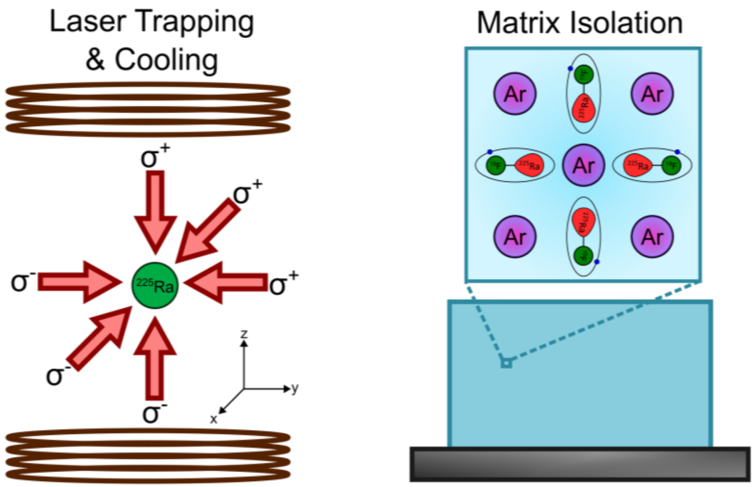
Step 1: Obtaining The Sample



Step 2: Sample Preparation

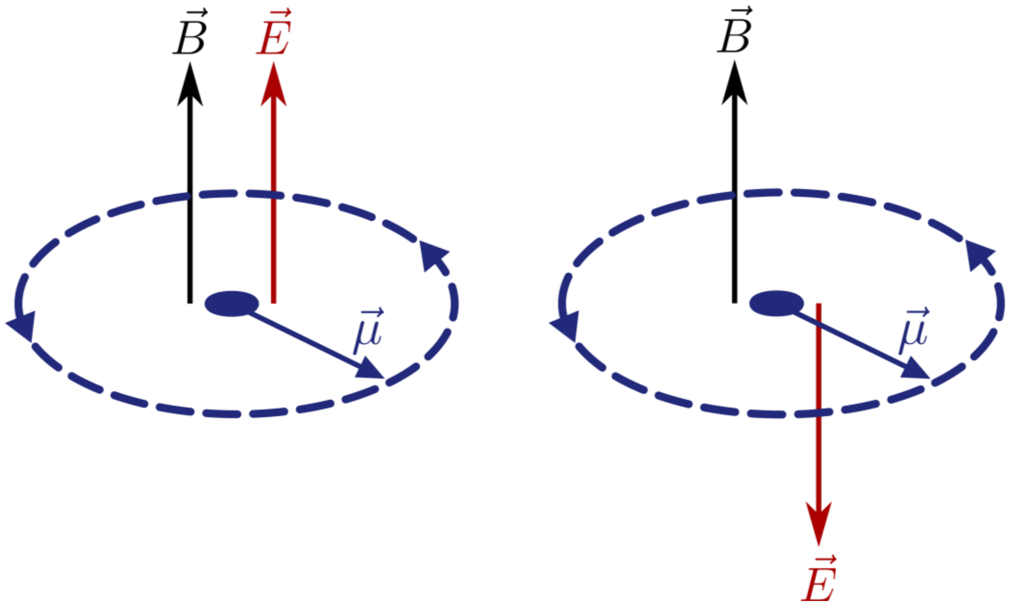


Step 3: Slowing And/Or Trapping

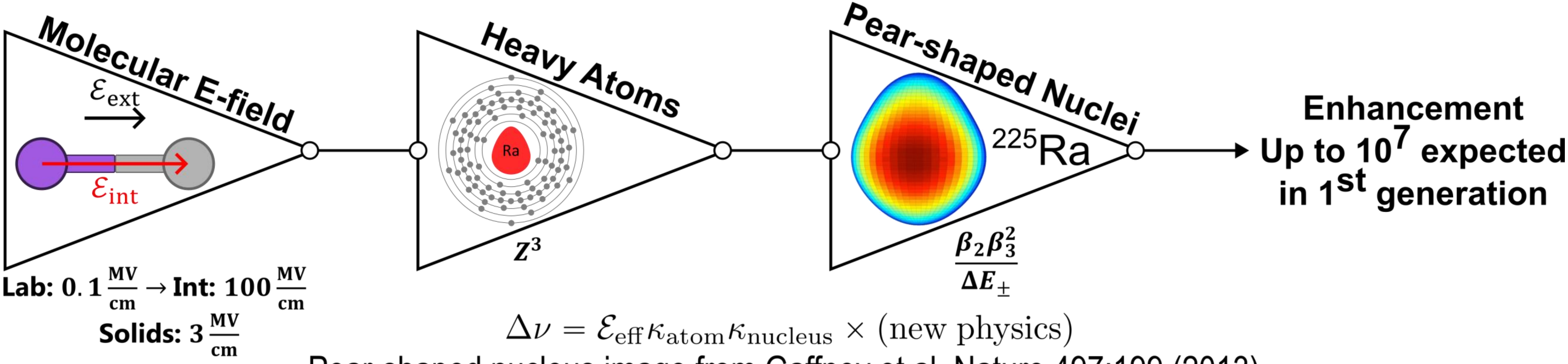


Step 4: Apply $\vec{B}$,$\vec{E}$ Fields and Observe Spin-Precession

$$h\nu_\uparrow = 2\left(\mu B_\uparrow + dE\right) \qquad h\nu_\downarrow = 2\left(\mu B_\downarrow - dE\right)$$



Borrow from Leptonic EDM Experiments: Combine Polar Molecules with Pear-shaped Nuclei



$$\Delta\nu = \mathcal{E}_{\text{eff}}\,\kappa_{\text{atom}}\,\kappa_{\text{nucleus}} \times (\text{new physics})$$

Pear-shaped nucleus image from Gaffney et al. Nature 497:199 (2013)

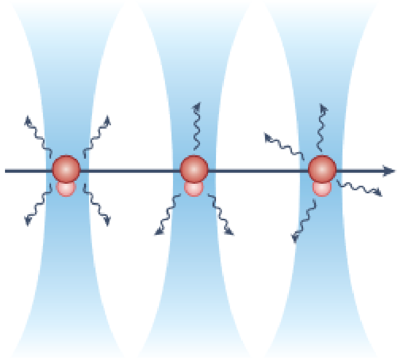
- Polar molecules have been demonstrated as an ultrasensitive tool for electron EDM searches
  - Easy to align molecule dipole moment with applied field
  - Molecule dependent co-magnetometry via energy splittings
  - Large internal fields produce larger splittings

- Polar molecules to be implemented in upcoming hadronic searches
  - CeNTREX: ^{205}TlF (Stable)
  - RaX: ^{225}RaF and ^{225}RaOH (Pear-shaped, not stable)
  - FrAg: ^{223}FrAg (Pear shaped, not stable)

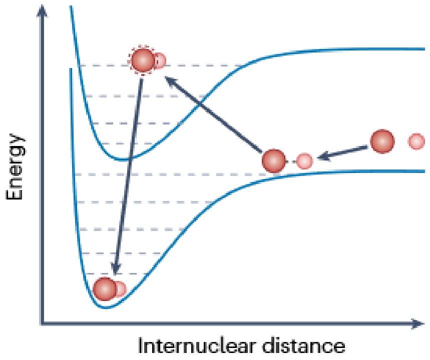
- Radioactive polar molecules challenging to use
  - Creation and handling of short-lived isotopes
  - How do we efficiently form molecules with these isotopes?

There Are Several Ways to Trap Radioactive Molecules for an EDM Measurement

Laser Cooling & Trapping



Ultracold Assembly



DeMille et al. Nature Physics 20:741 (2024)

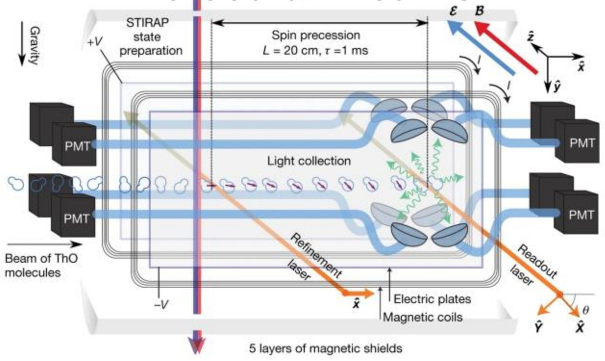
Opportunity

- Lots of progress with stable molecules, RaF favorable for laser cooling

Challenge

- Complex cooling schemes, Low efficiency, ~1%

Molecular Beams



Panda (Harvard PhD Thesis, 2018)

Opportunity

- Could be very efficient path to ultracold molecules

Challenge

- Limited to laser coolable atoms
- Species lifetime: current focus $^{223}\text{Fr}^{107}\text{Ag}$ (^{223}Fr $\tau_{1/2} = 22\ min$)

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- Lots of progress in making cold, intense beam of stable molecules

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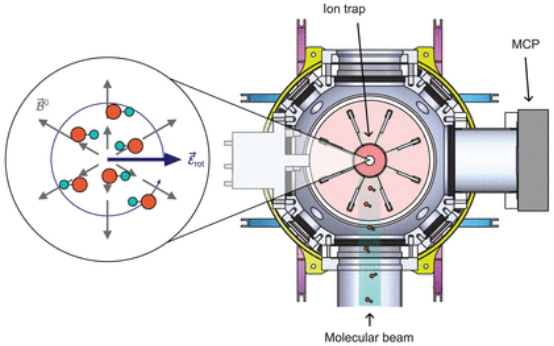
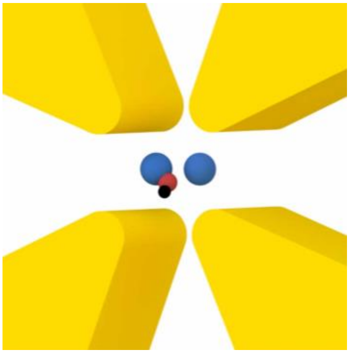
- Efficiency not characterized or optimized

Ion Trapping

Roussy et al. Science 381:46 (2023)



Arrowsmith-Kron et al. Rep. Prog. In Phys. 87:084301 (2024)



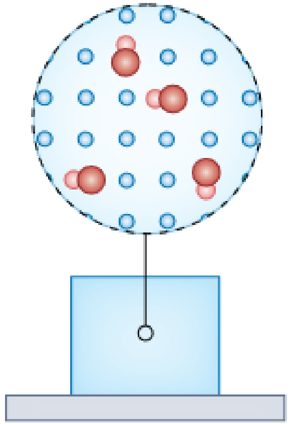
Opportunity

- Flexible for many species, long coherence times, amenable to small sample sizes

Challenge

- Limited trapping capacity due to Coulomb repulsion

Matrix Isolation



Opportunity

- Large number of molecules (10^{13}) can be trapped in a small volume (1mm^3)

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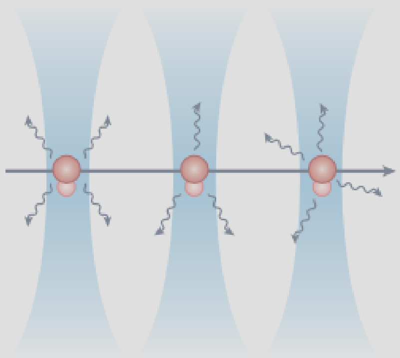

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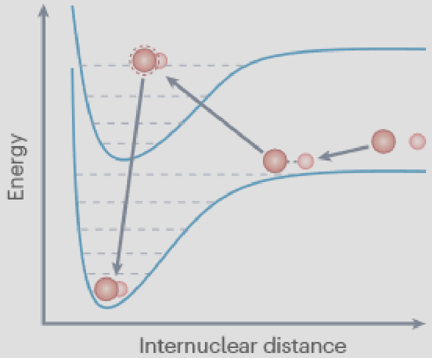

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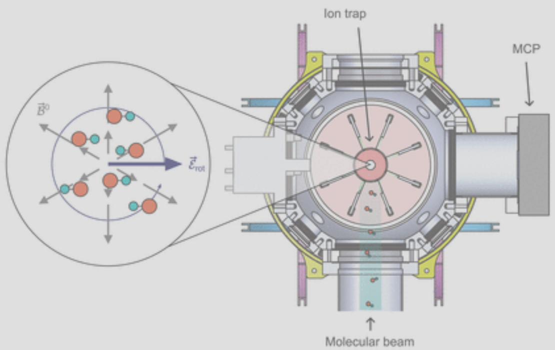

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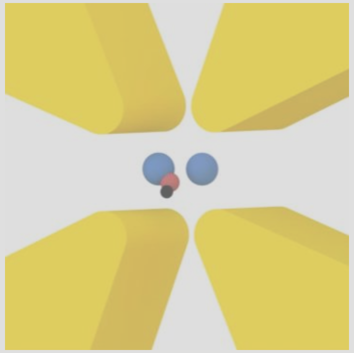

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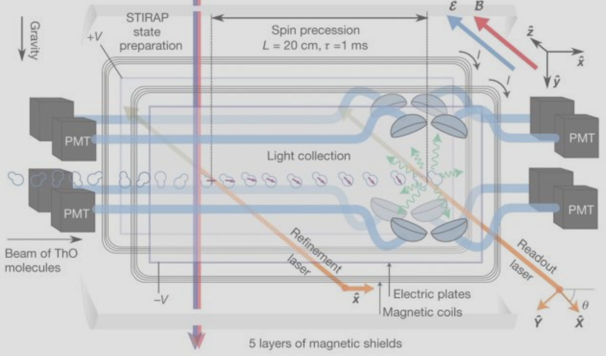

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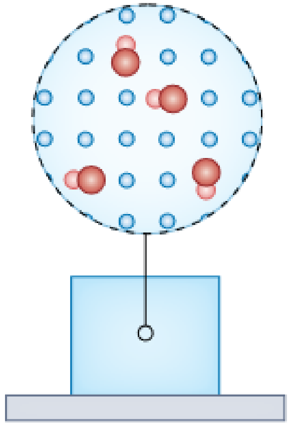

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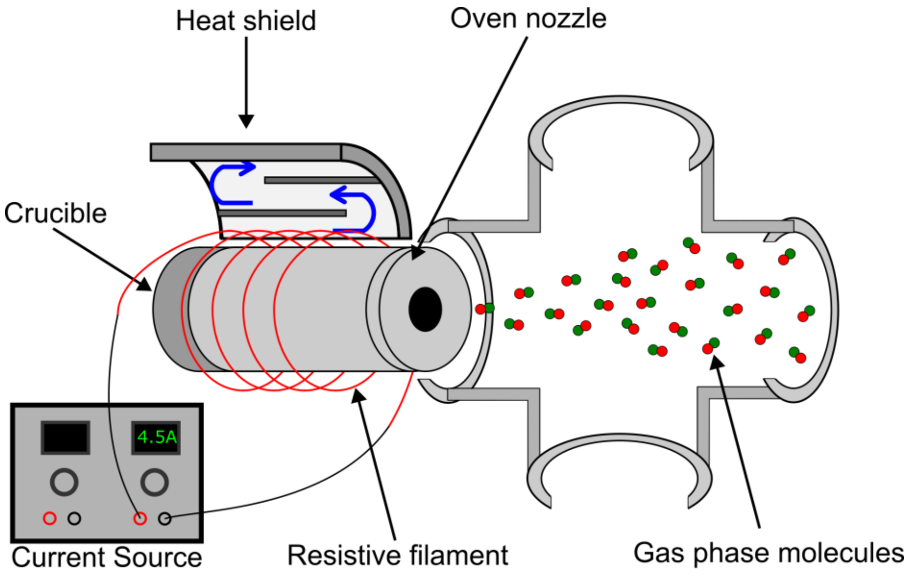
DeMille et al. Nature Physics 20:741 (2024)

Nicholas Nusgart
Saturday March 16
09:40 – 10:20

There Are Also Several Ways To Produce Molecules

Most Brute Force: High Temperature Oven



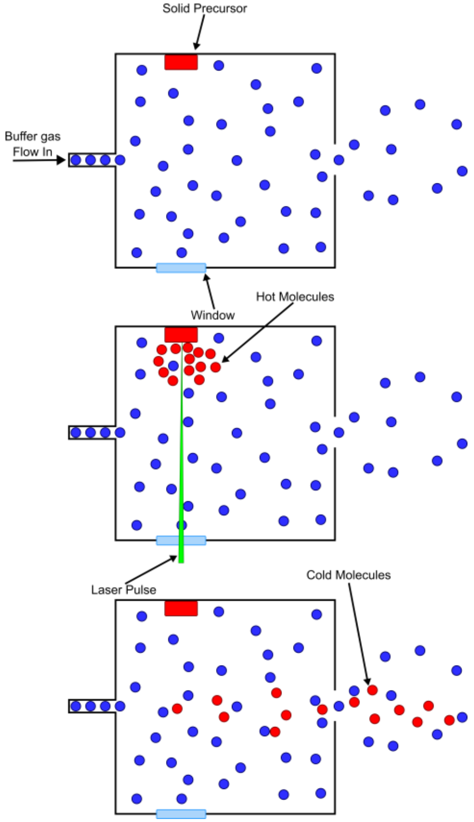
Opportunity

- Capable of creating neutral molecular beams from solid/liquid precursor

Challenge

- High temperatures required (~10^3 ºC) for radioactive molecules risks destroying them, efficiencies not well known

Less Brute Force: Laser Ablation + Cold Buffer Gas



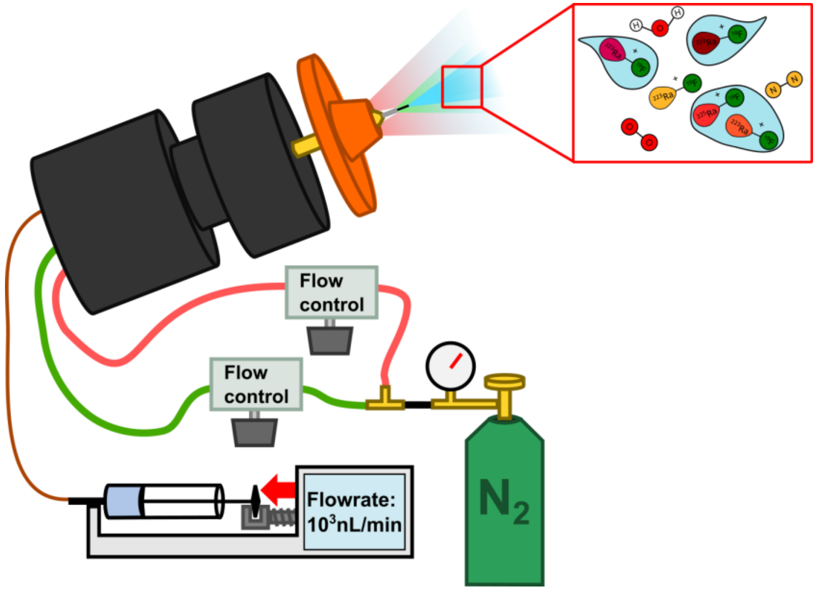
Opportunity

- Capable of creating slow, bright beams of neutral molecules

Challenge

- Macroscopic solid precursors required, efficiencies not characterized or optimized

Exotic And Weird: Electrospray Ionization



Opportunity

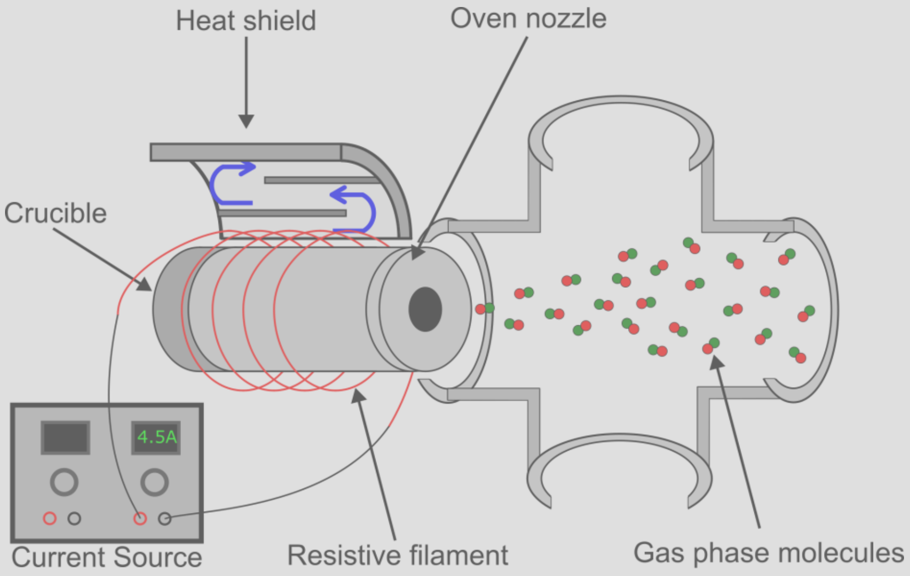
- Capable of creating molecular ions from liquid precursors with efficiencies up to 50%

Challenge

- Largely untested by fundamental symmetries community

There Are Also Several Ways To Produce Molecules

Most Brute Force: High Temperature Oven



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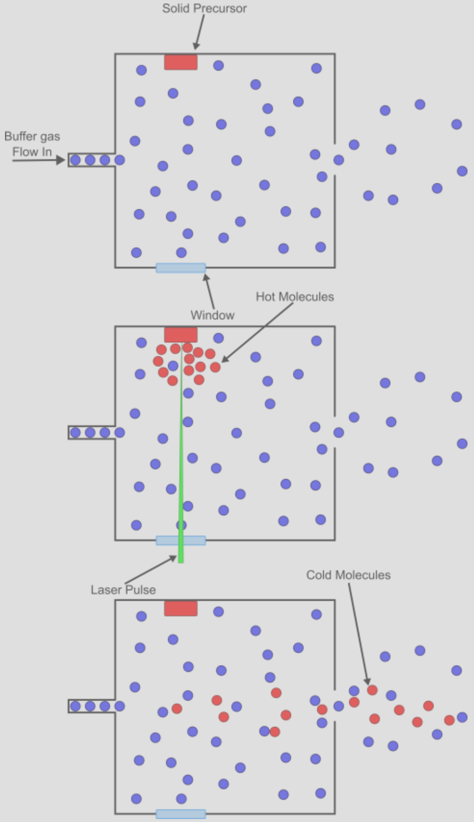

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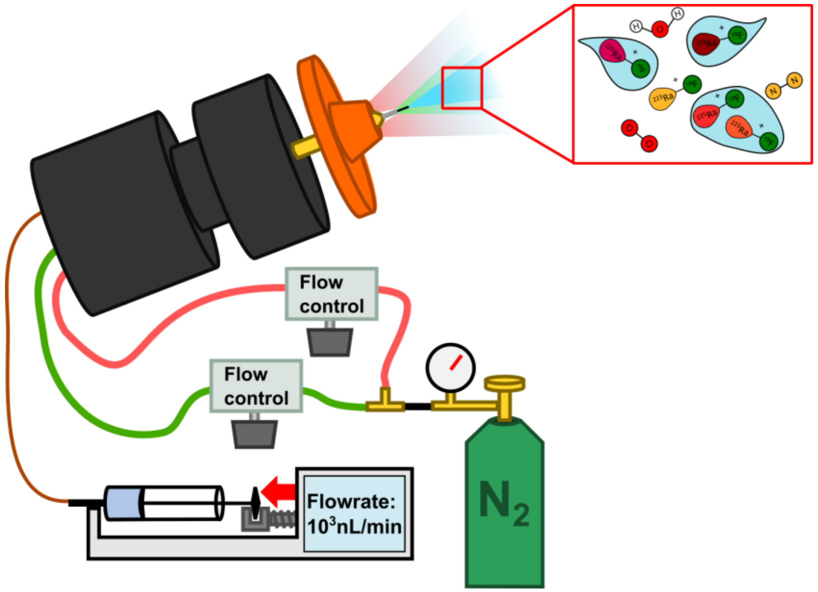

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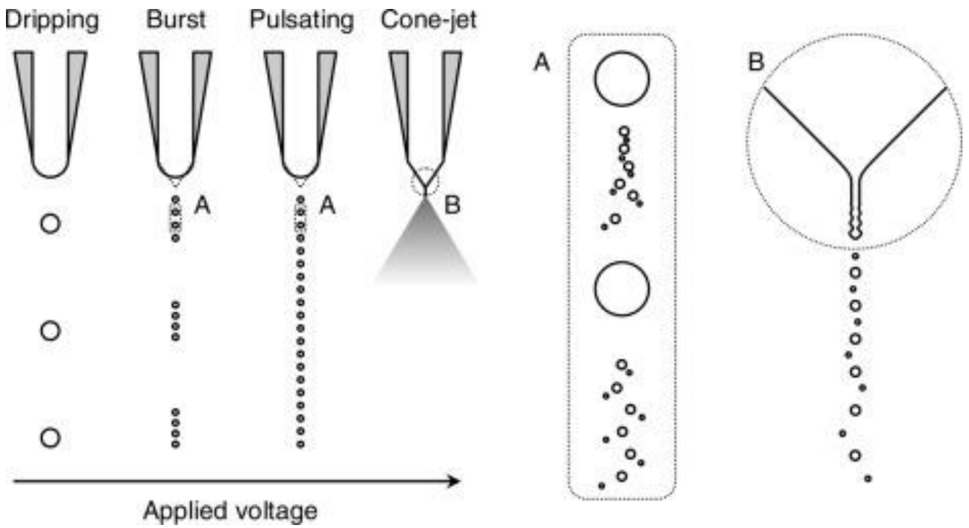
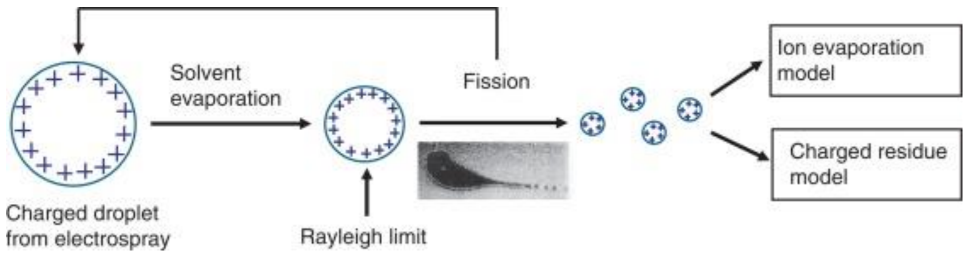
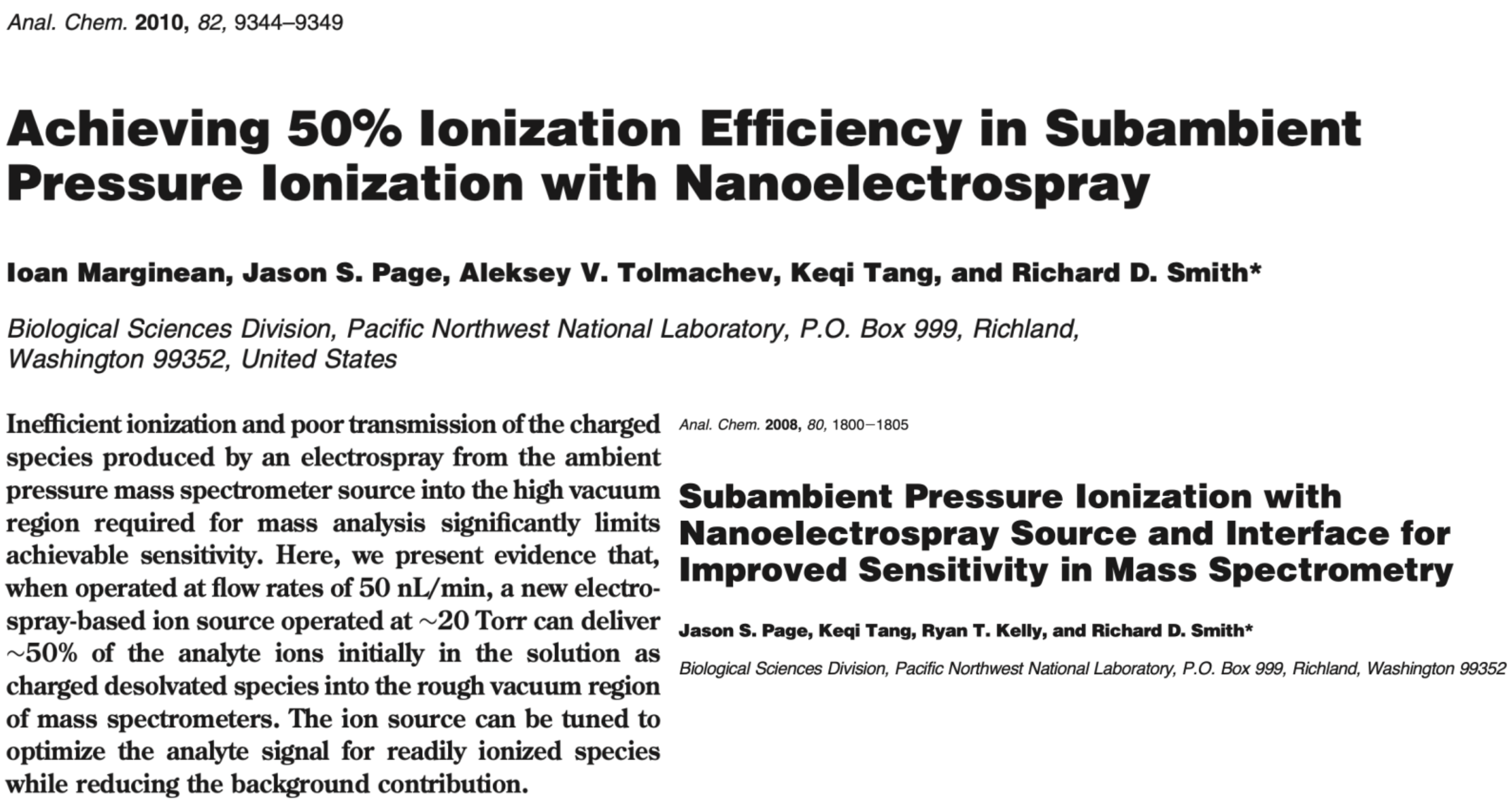
Electrospray Ionization:
It Could Be Very Efficient For Forming Radioactive Molecules



Encyclopedia of Spectroscopy and Spectrometry, Third Edition
https://doi.org/10.1016/B978-0-12-803224-4.00319-8





Anal. Chem. **2010,** *82,* 9344–9349

Achieving 50% Ionization Efficiency in Subambient Pressure Ionization with Nanoelectrospray

Ioan Marginean, Jason S. Page, Aleksey V. Tolmachev, Keqi Tang, and Richard D. Smith*

Biological Sciences Division, Pacific Northwest National Laboratory, P.O. Box 999, Richland, Washington 99352, United States

Inefficient ionization and poor transmission of the charged species produced by an electrospray from the ambient pressure mass spectrometer source into the high vacuum region required for mass analysis significantly limits achievable sensitivity. Here, we present evidence that, when operated at flow rates of 50 nL/min, a new electrospray-based ion source operated at ~20 Torr can deliver ~50% of the analyte ions initially in the solution as charged desolvated species into the rough vacuum region of mass spectrometers. The ion source can be tuned to optimize the analyte signal for readily ionized species while reducing the background contribution.

Anal. Chem. **2008,** *80,* 1800–1805

Subambient Pressure Ionization with Nanoelectrospray Source and Interface for Improved Sensitivity in Mass Spectrometry

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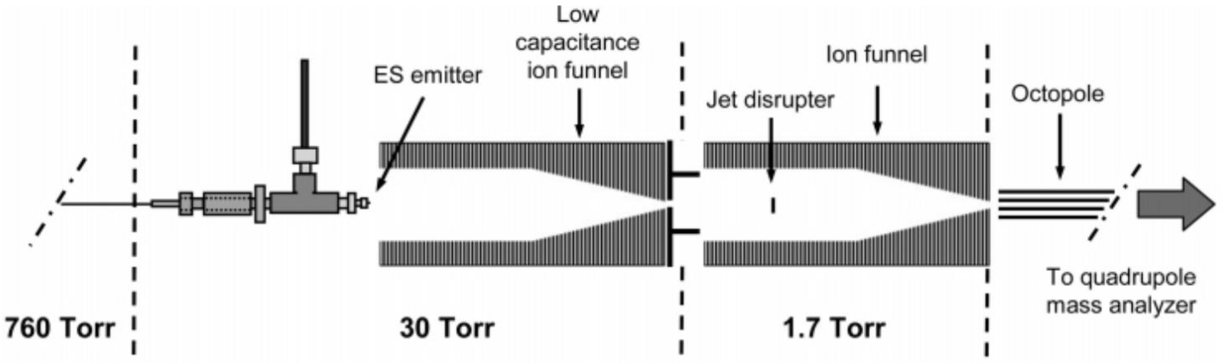
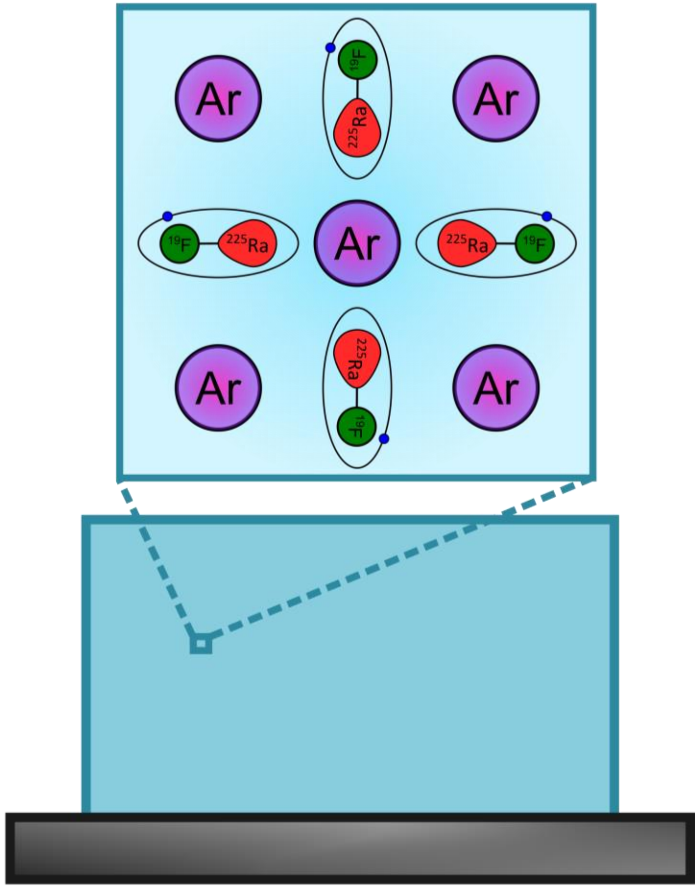


The FRIB-EDM3 Instrument:

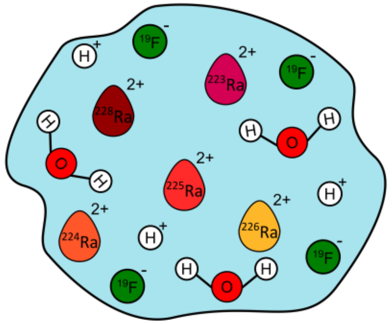
Form Radioactive Molecules (^{225}RaF, $\tau_{1/2}$ = 15 days) and Embed Them in Solids



The FRIB-EDM3 Instrument:
Radium Isotopes Will Be Harvested From FRIB Water Beam Dump

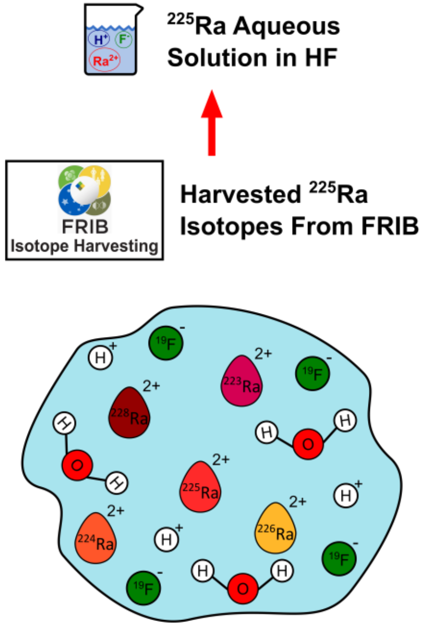
FRIB
Isotope Harvesting

Harvested ^{225}Ra Isotopes From FRIB



The FRIB-EDM3 Instrument:

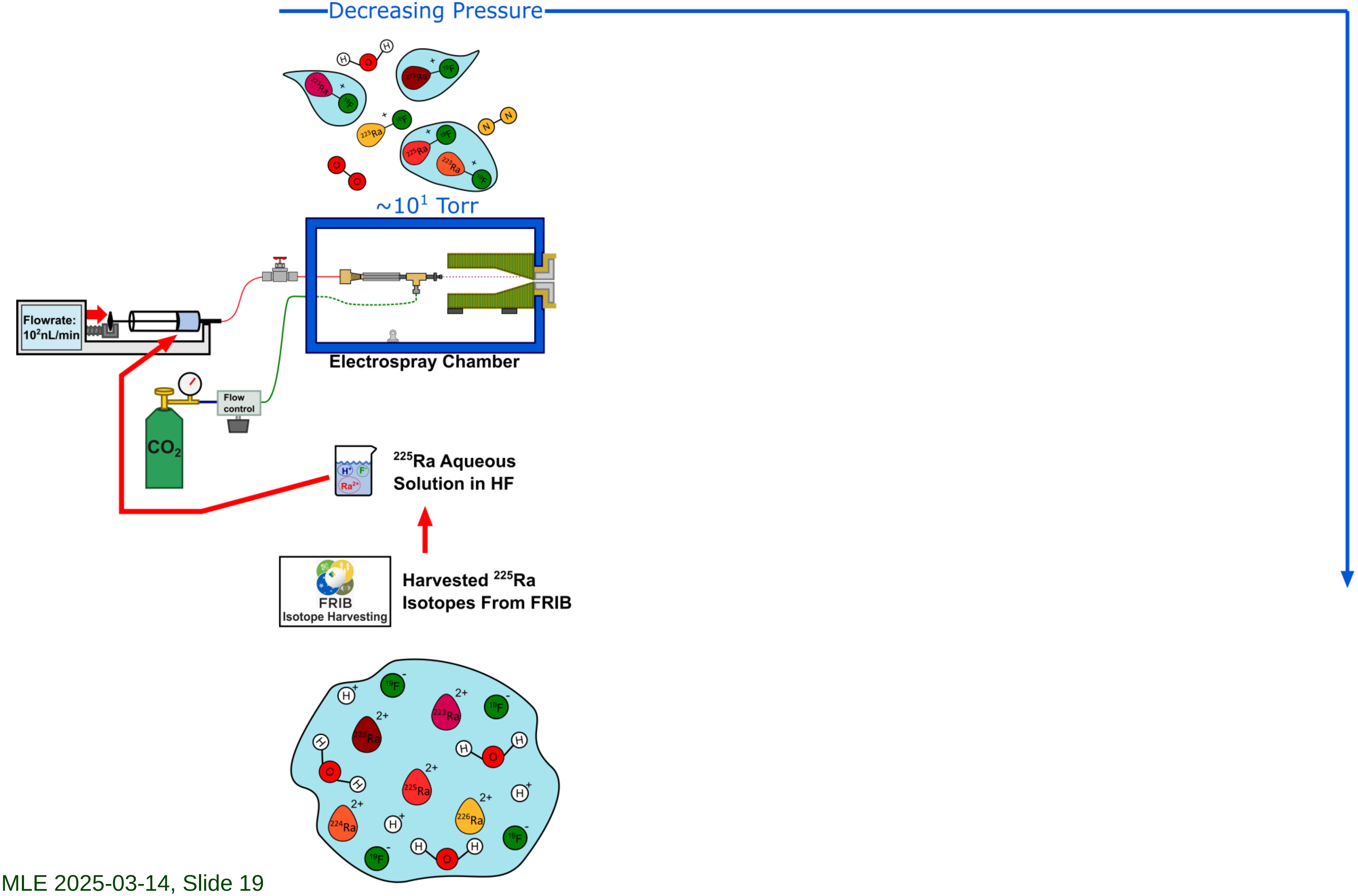
FRIB Isotope Harvesting Can Provide Us With Radium Solution



^{225}Ra Aqueous Solution in HF

Harvested ^{225}Ra Isotopes From FRIB

The FRIB-EDM3 Instrument:
Flow Solution Into An Electrospray Ionization Source To Make Molecular Ions



Decreasing Pressure

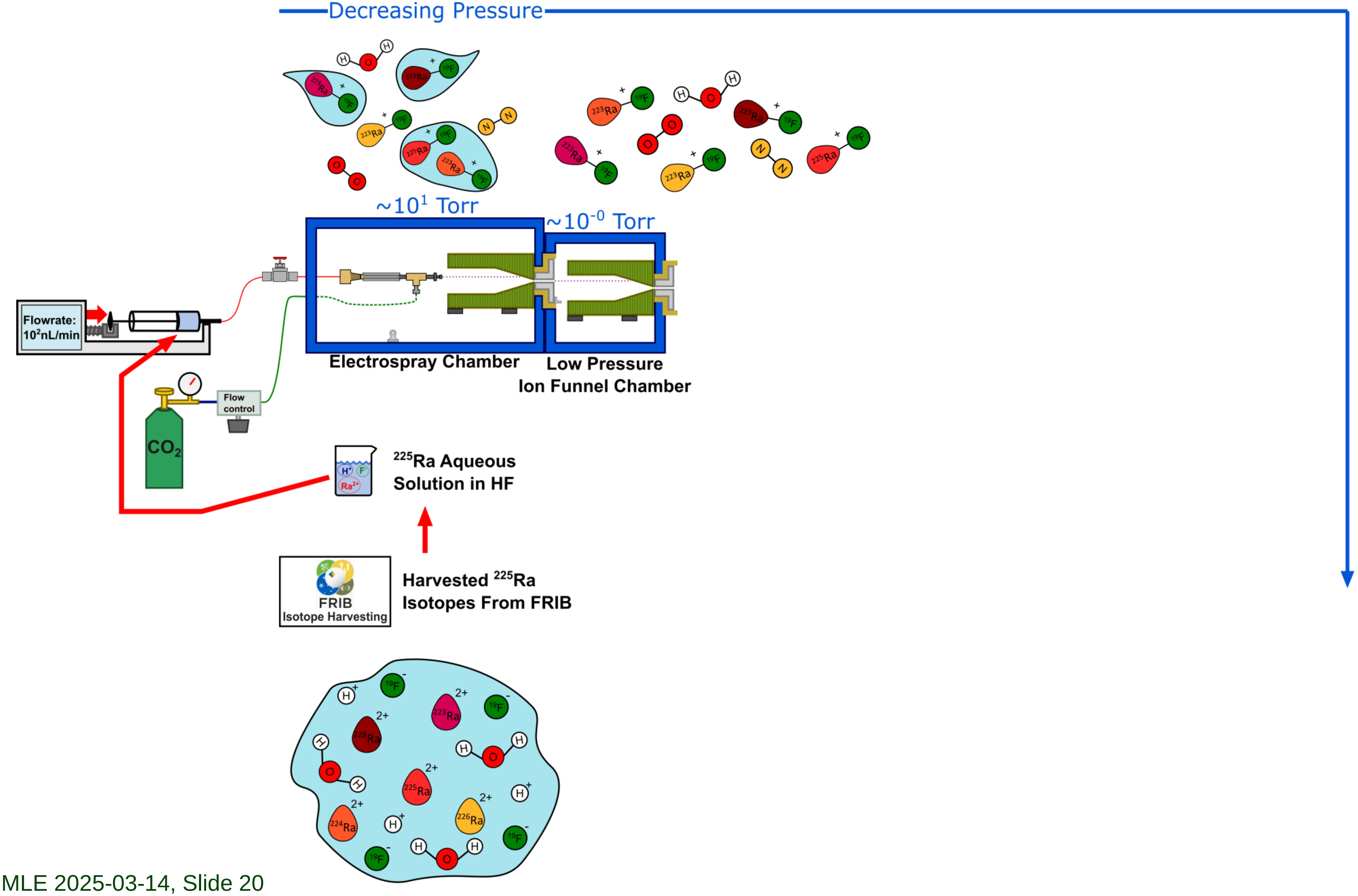
~10^1 Torr

Electrospray Chamber

^{225}Ra Aqueous Solution in HF

Harvested ^{225}Ra Isotopes From FRIB

The FRIB-EDM3 Instrument:
Collect Molecular Ions And Separate From Gas Load With Ion Funnels



Decreasing Pressure

~10^1 Torr

~10^{-0} Torr

Flowrate:
10^2nL/min

Flow control

CO_2

Electrospray Chamber

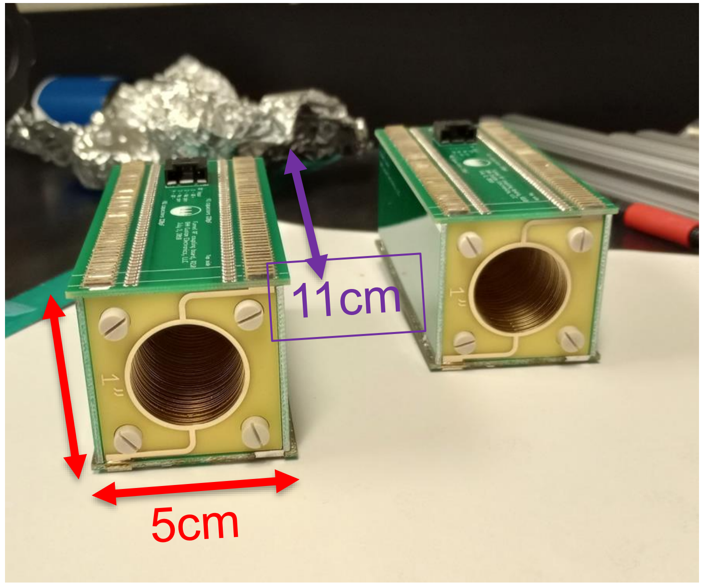
Low Pressure Ion Funnel Chamber

^{225}Ra Aqueous Solution in HF

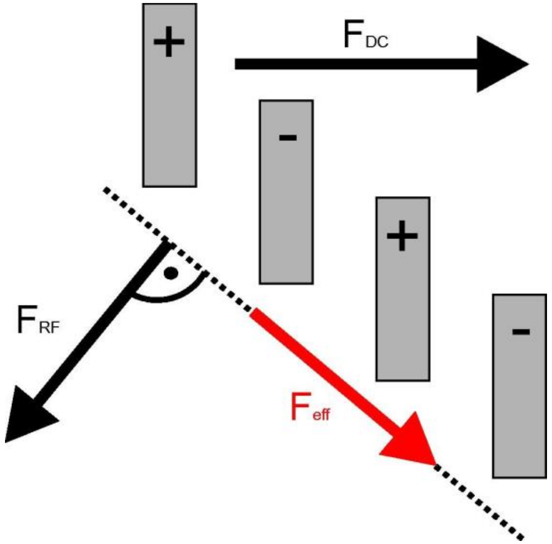
FRIB
Isotope Harvesting

Harvested ^{225}Ra Isotopes From FRIB

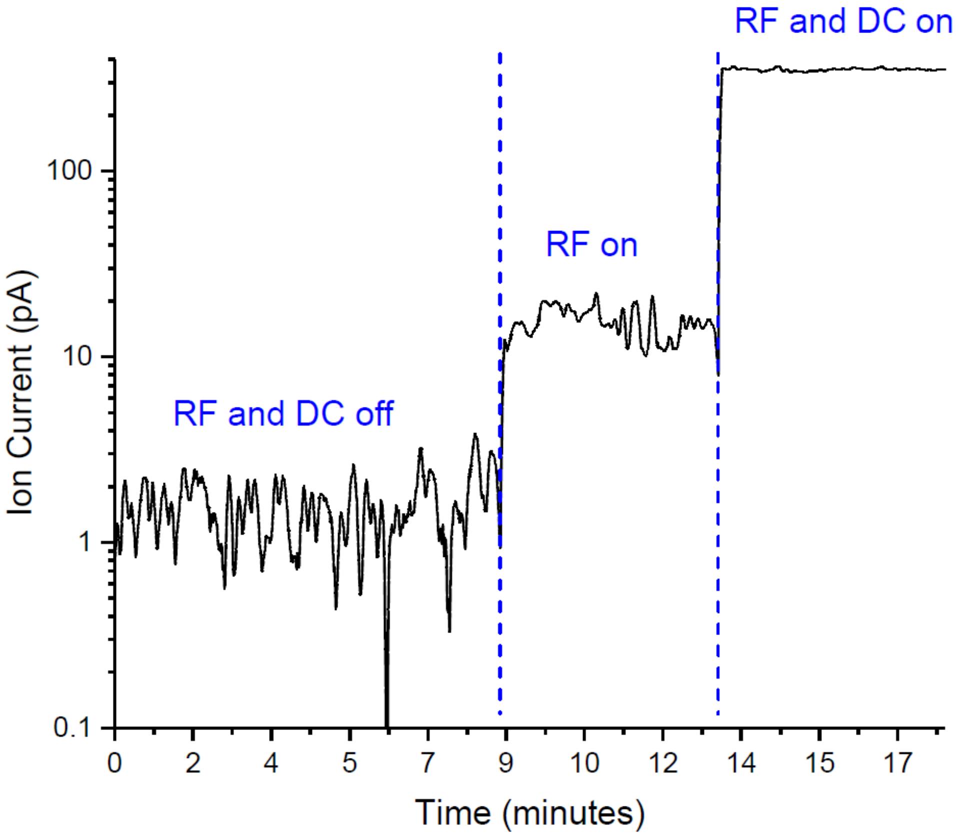
Testing With Isopropanol: Ion Funnels Work Well!

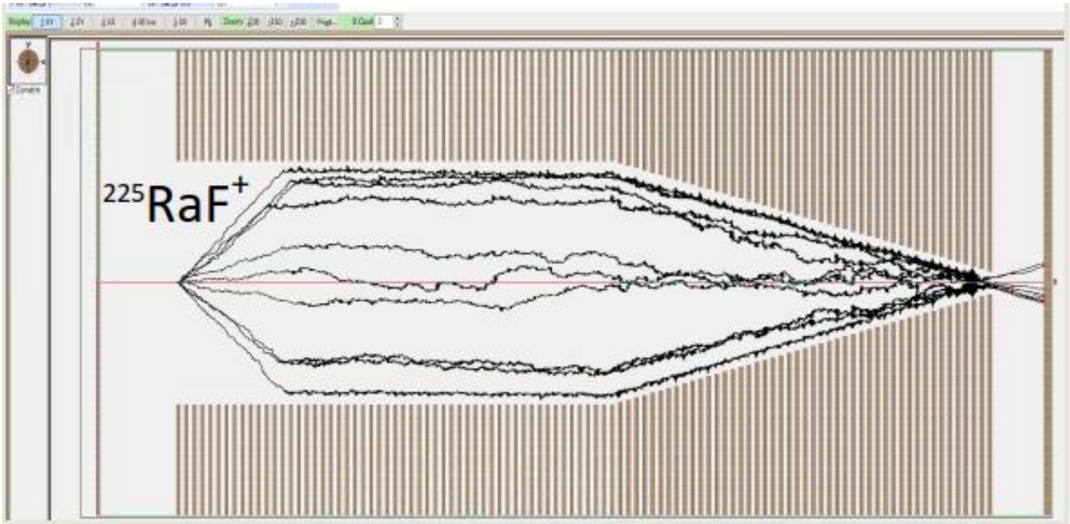


GAA Custom Electronics (Gordon Anderson)



Droese et al. NIMB 338:126 (2014)





Stable ion beams are possible:

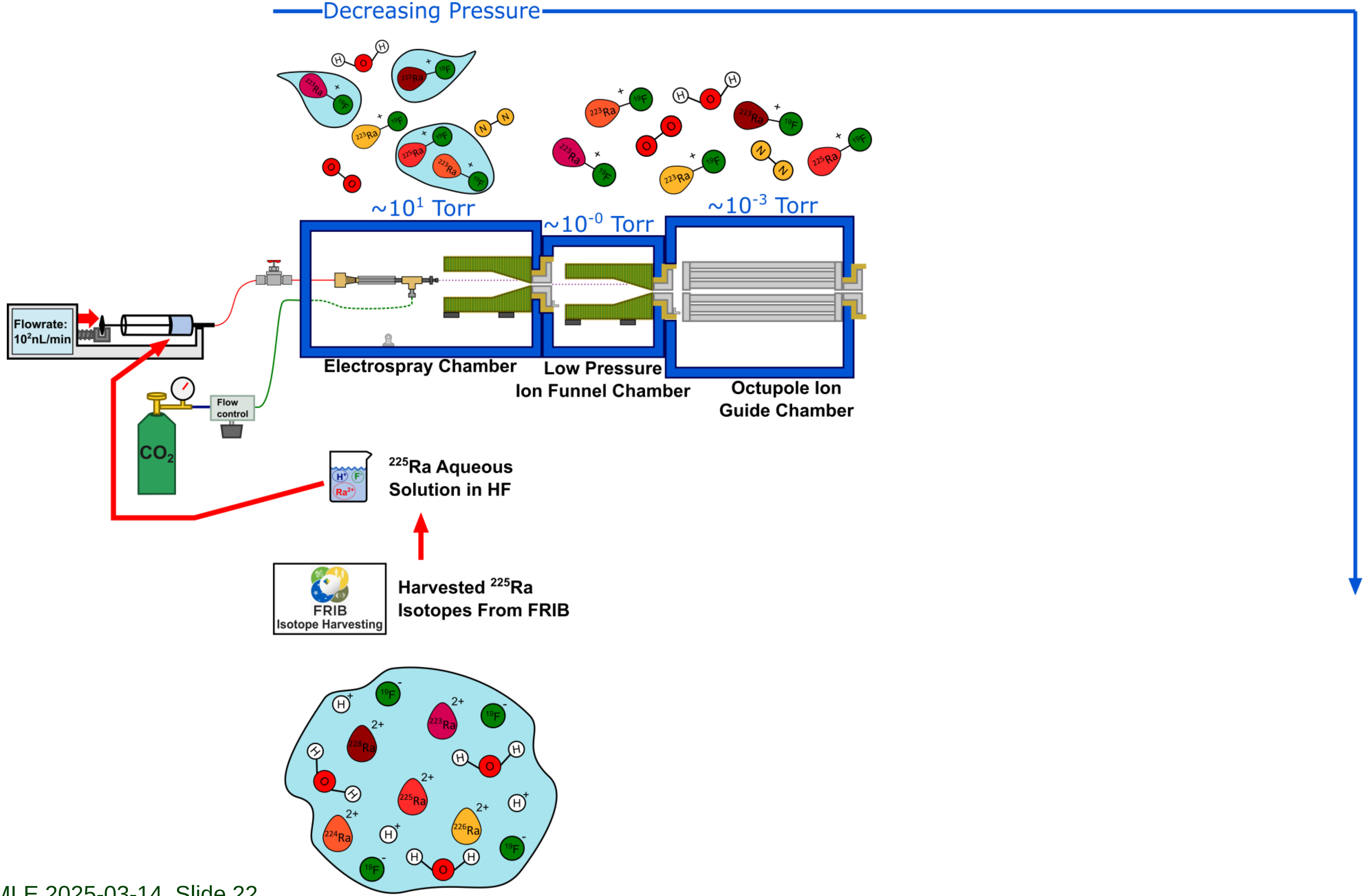
- Can be used during installation to test ion optics
- Optimizing applied bias

Challenges:

- Testing done with atmospheric electrospray
  - Low reproducibility
  - Lack of precise positioning and alignment

The FRIB-EDM3 Instrument:

Octupole Ion Guide Stage For Differential Pumping



Electrospray Chamber

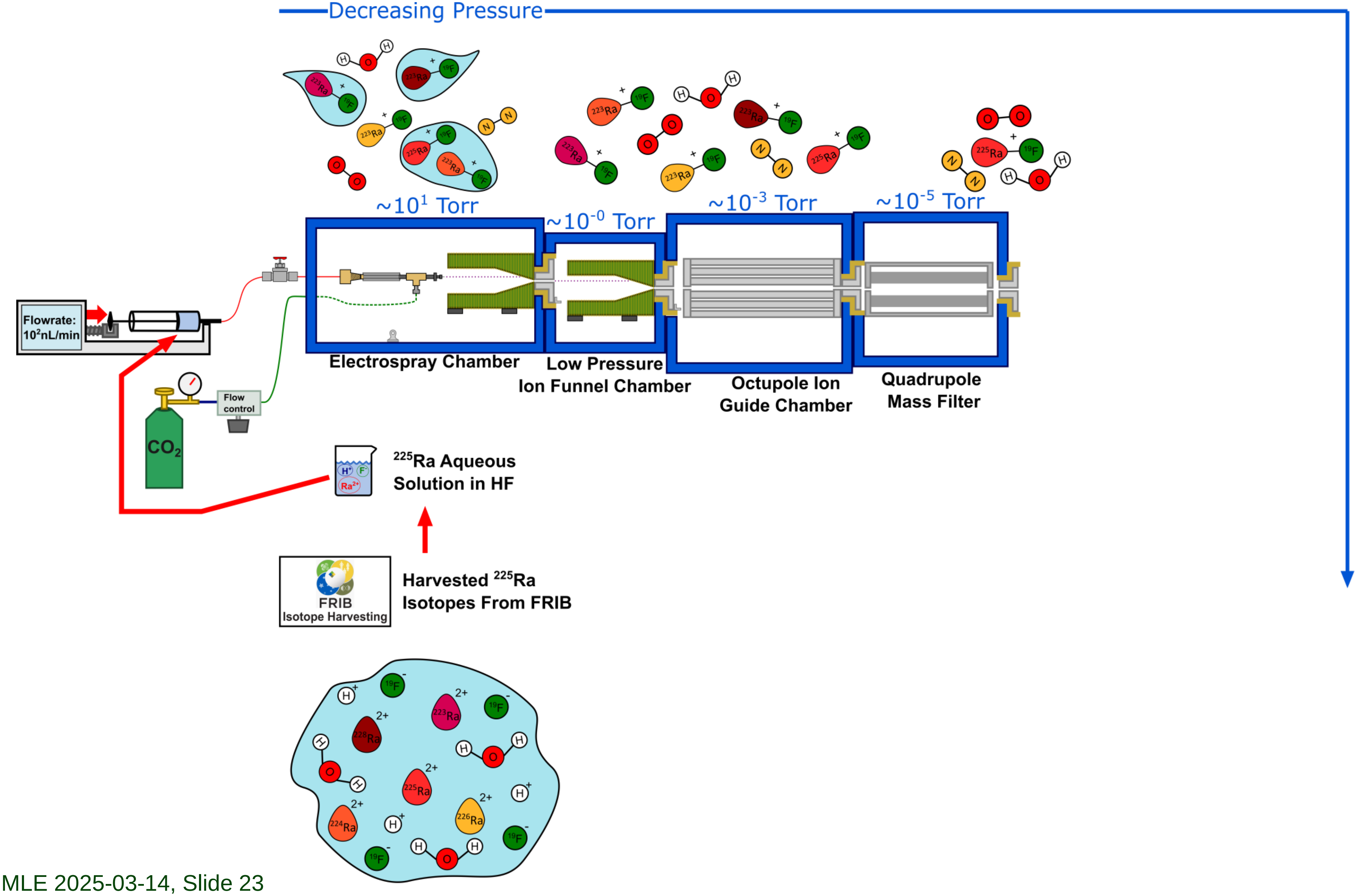
Low Pressure Ion Funnel Chamber

Octupole Ion Guide Chamber

^{225}Ra Aqueous Solution in HF

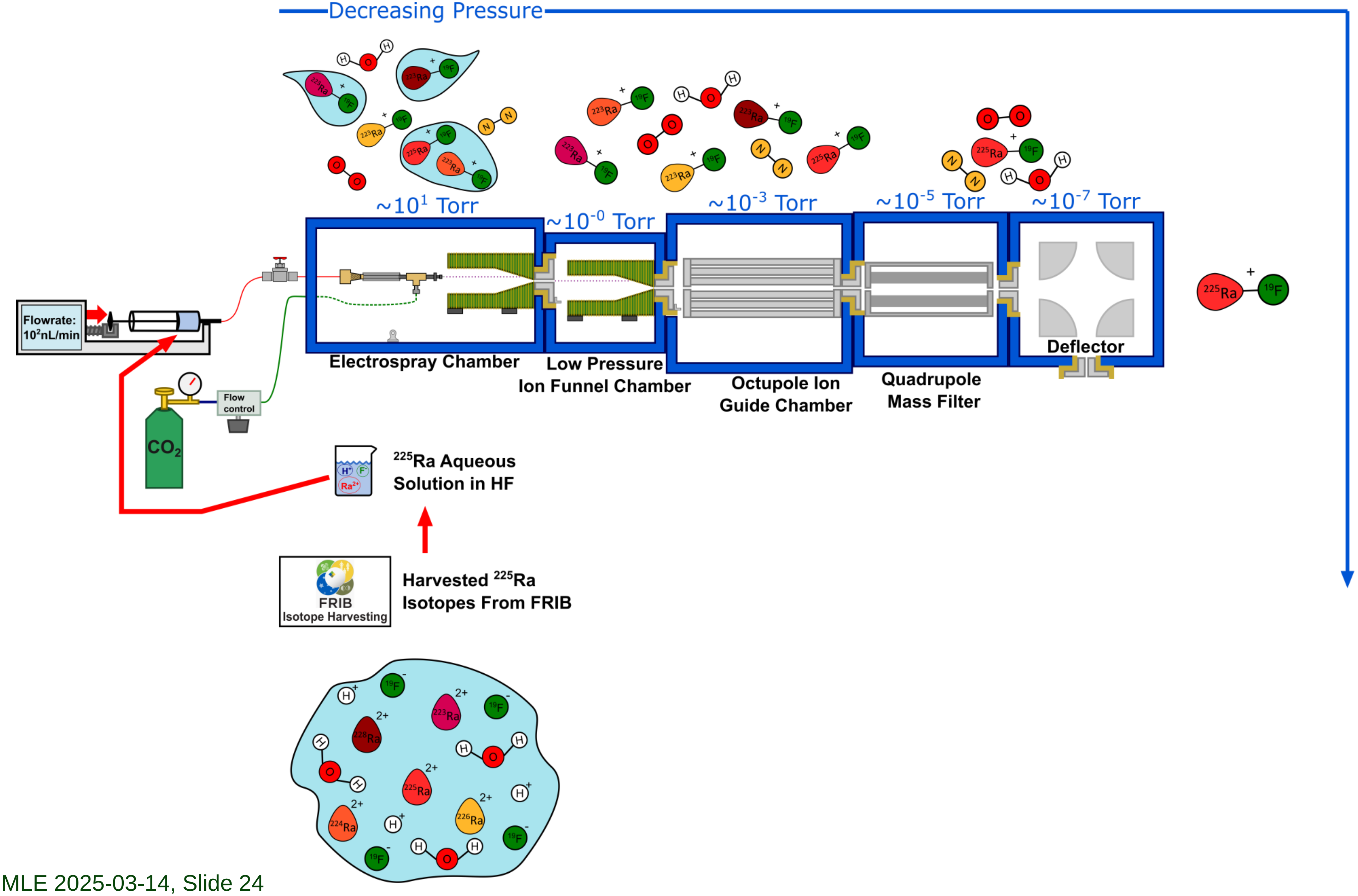
Harvested ^{225}Ra Isotopes From FRIB

The FRIB-EDM3 Instrument:
Quadrupole Mass Filter For Isotopic And Molecular Selection

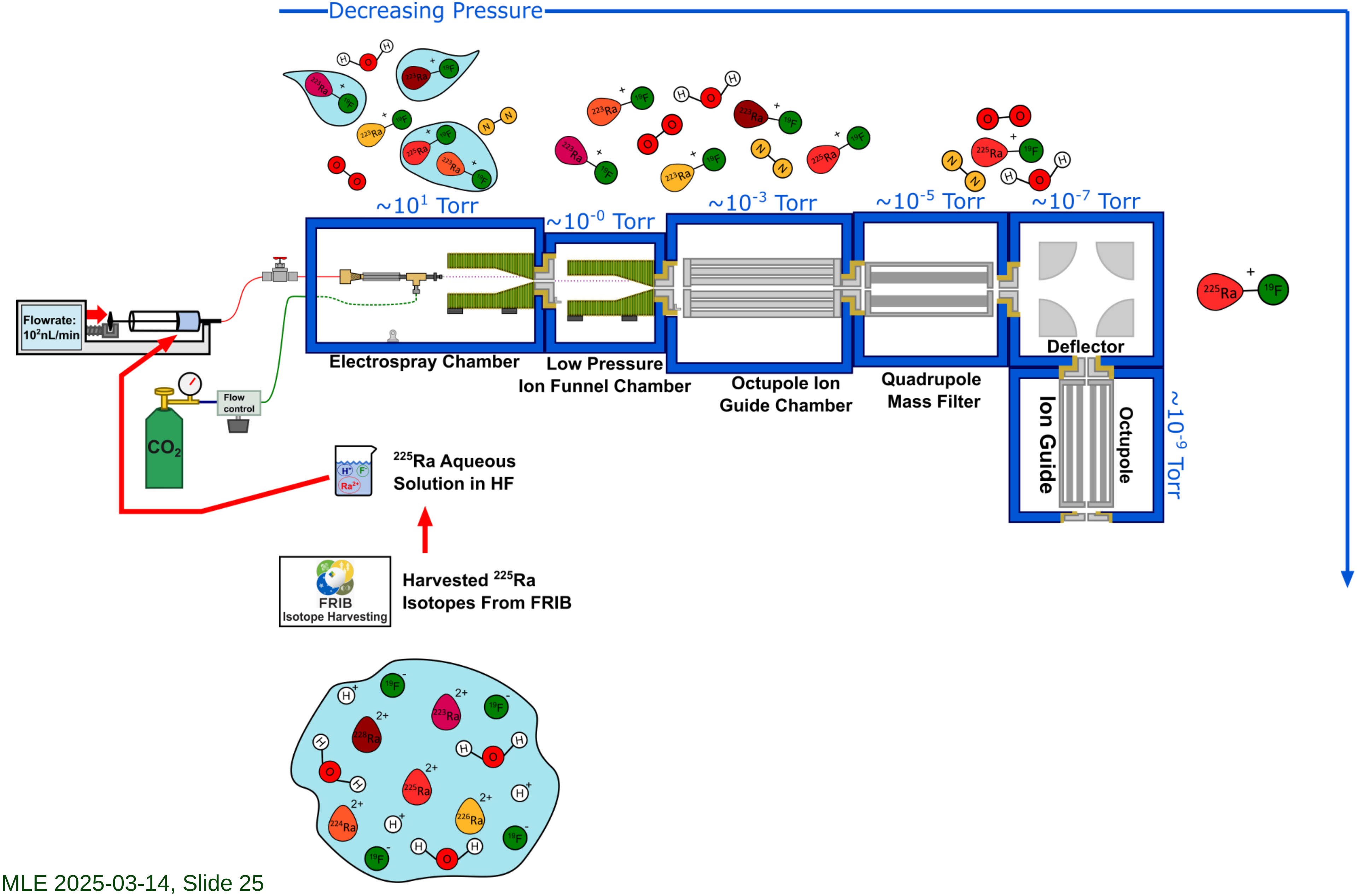


The FRIB-EDM3 Instrument:

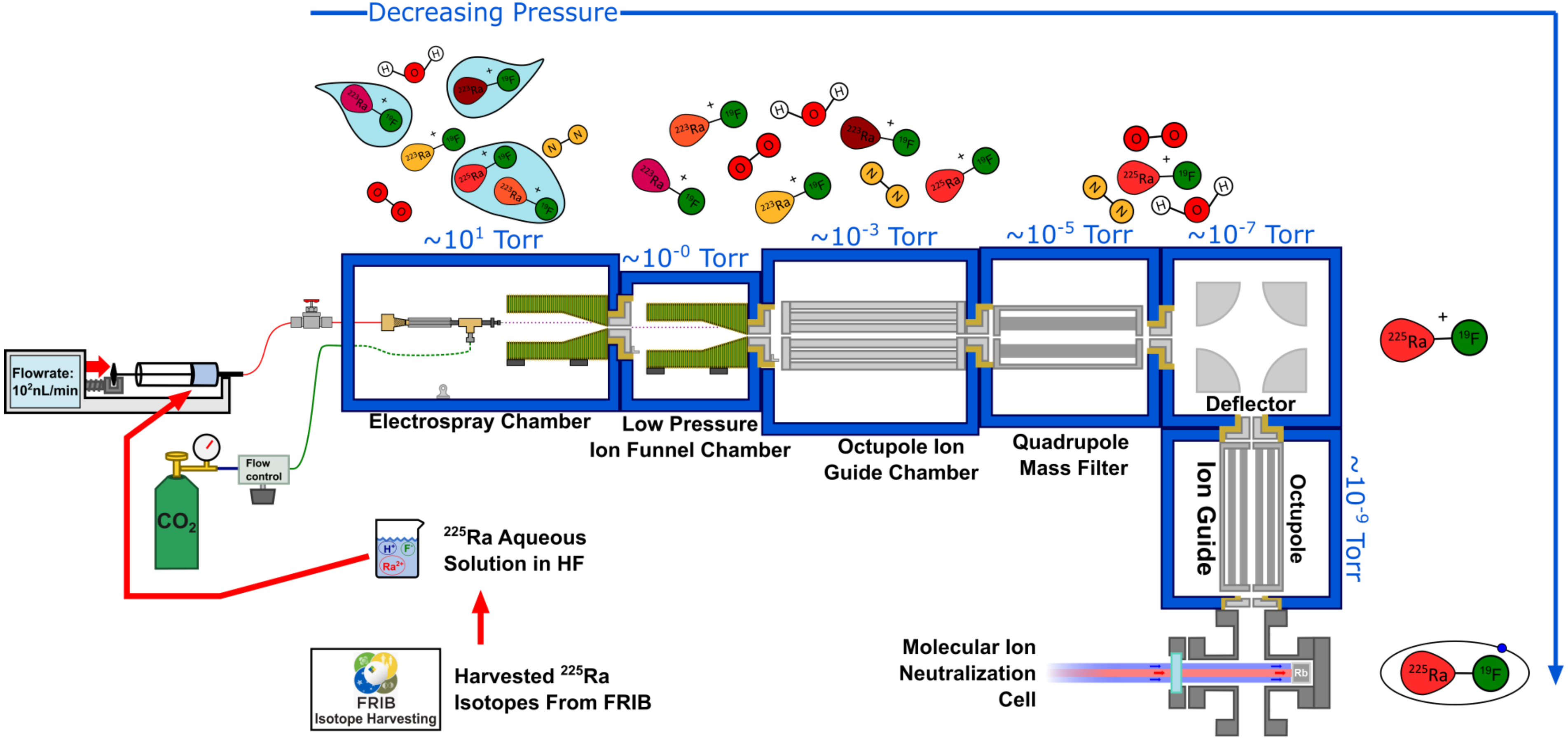
Electrostatic Deflector To Suppress Line Of Sight Neutrals

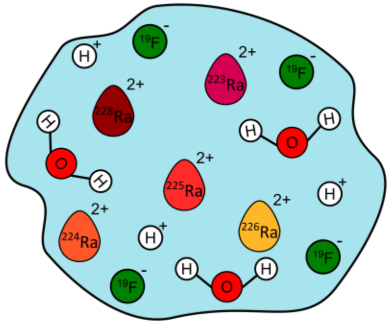


The FRIB-EDM3 Instrument:
Second Octupole Ion Guide Stage For More Differential Pumping



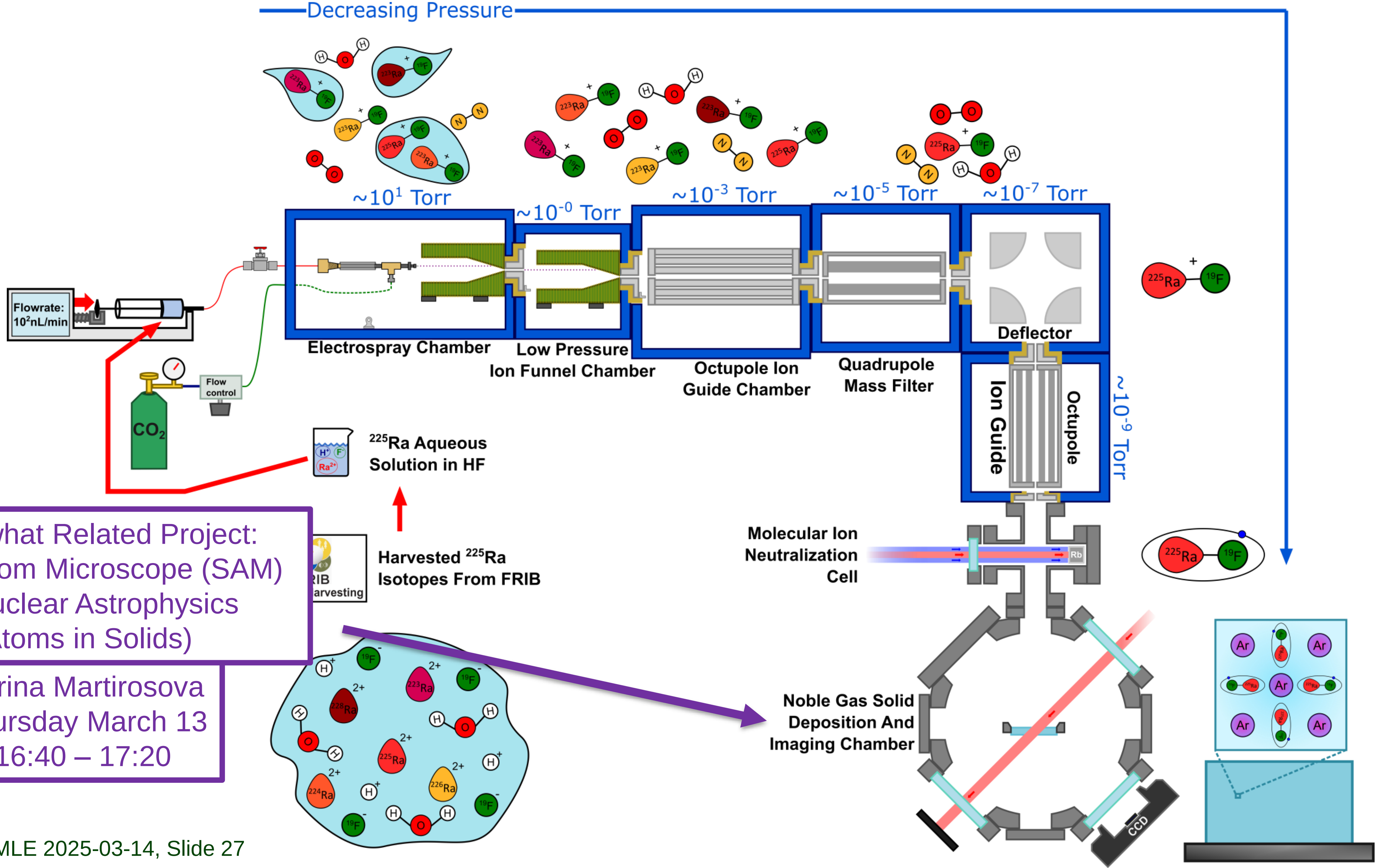
The FRIB-EDM3 Instrument:
Charge Exchange Cell For Neutralizing Molecular Ions





Alkali Rydberg atoms could be used for charge exchange to neutralize molecular ions (charge exchange cross sections scale like n^4)

The FRIB-EDM3 Instrument:
Co-deposit Neutral Molecules With Noble Gas Atoms Onto A Cryogenic Substrate



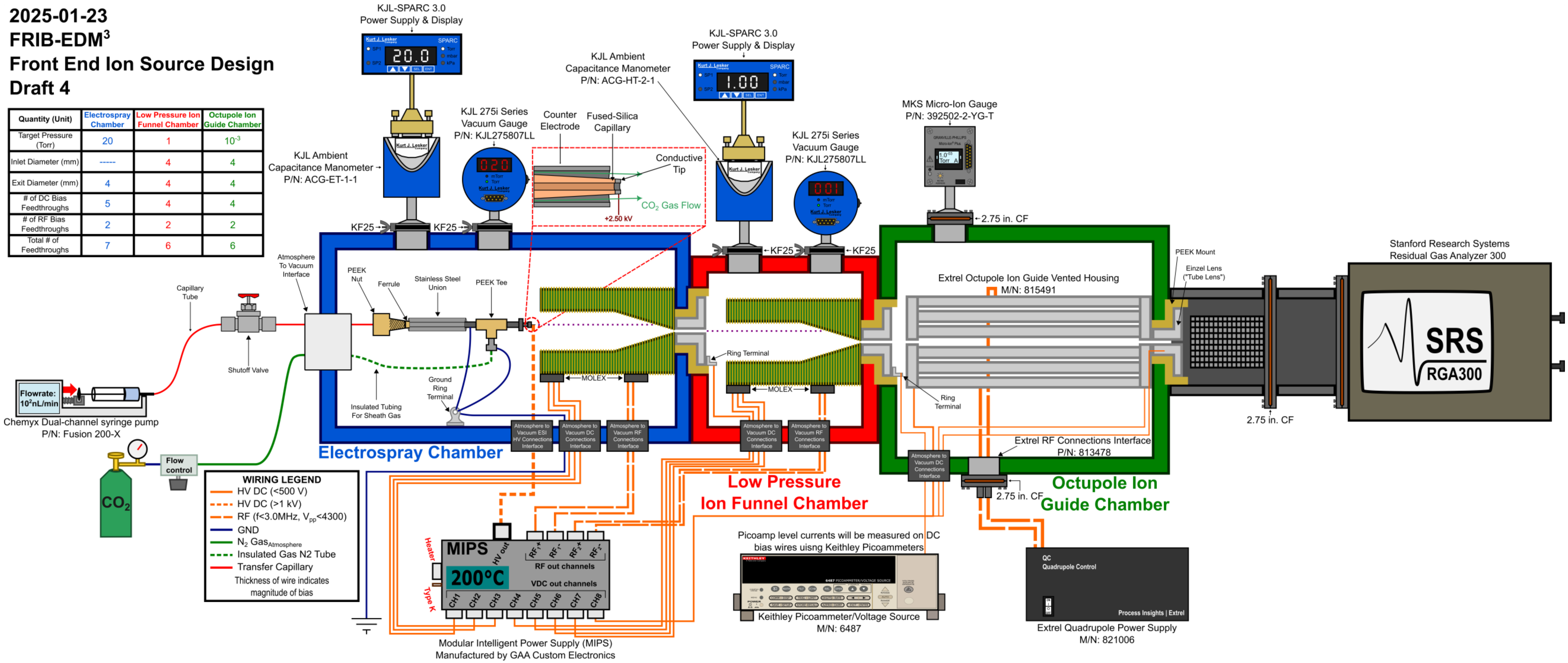
Somewhat Related Project:
Single Atom Microscope (SAM)
for Nuclear Astrophysics
(Atoms in Solids)

Karina Martirosova
Thursday March 13
16:40 – 17:20

Current Activities:
Complete Redesign of Front-End Interface

2025-01-23
FRIB-EDM^3
Front End Ion Source Design
Draft 4

Quantity (Unit)	Electrospray Chamber	Low Pressure Ion Funnel Chamber	Octupole Ion Guide Chamber
Target Pressure (Torr)	20	1	10^{-3}
Inlet Diameter (mm)	-----	4	4
Exit Diameter (mm)	4	4	4
# of DC Bias Feedthroughs	5	4	4
# of RF Bias Feedthroughs	2	2	2
Total # of Feedthroughs	7	6	6

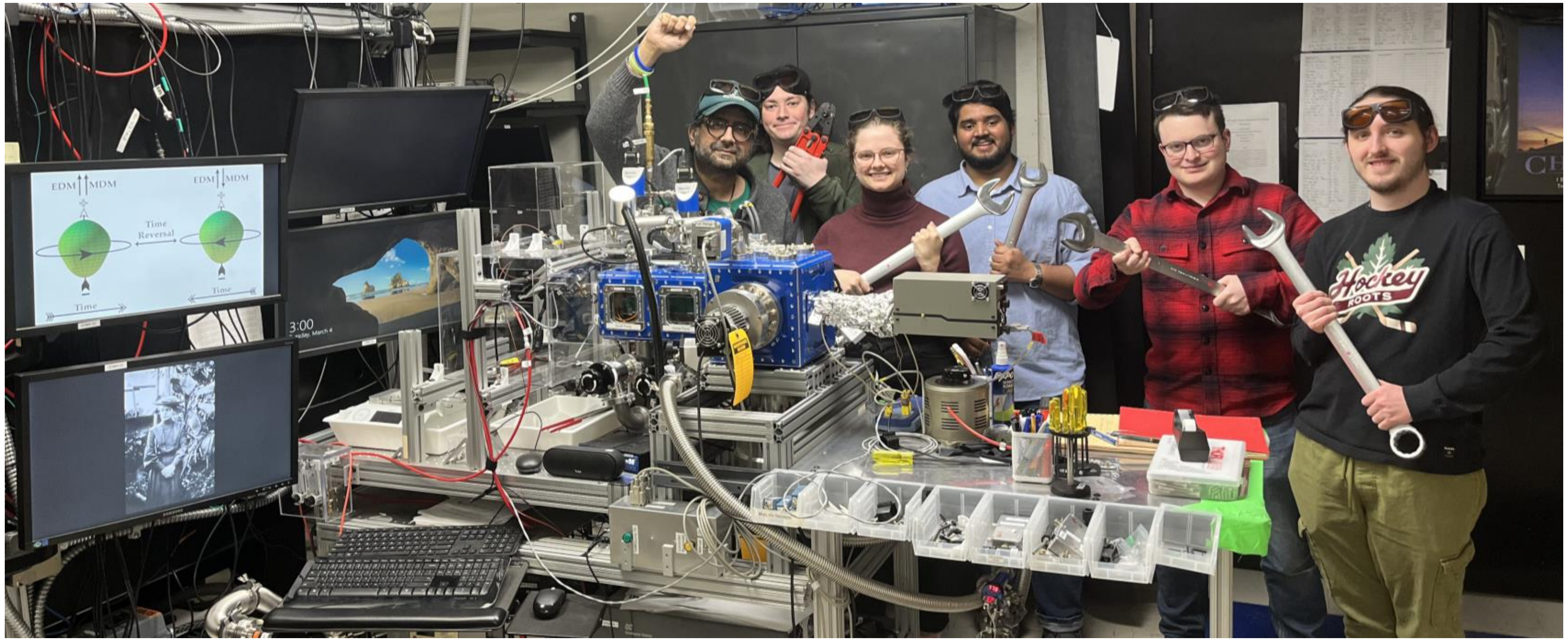


We believe the most efficient path forward is to take our electrospray efforts to a sub-ambient pressure at nanoliter per minute flow rates

Summary

- CP Violation could help account for discrepancy between the observed and predicted Baryon Asymmetry of the Universe (BAU)
- Non-zero EDMs and NSMs are a direct signature of T- and P-violation and thus also CP-violation
- Radioactive molecules could be a very useful tool for increasing statistics and offering additional degrees of freedom for control of systematics in Hadronic EDM/NSM searches
- Matrix isolation could be a powerful option for trapping many molecules in a small volume if we can exhibit enough control over effects in-medium
- Electrospray ionization could be an efficient path forward for producing radioactive molecules from small sample sizes

Thank You For Your Attention!



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Spinlab @ MSU:
PI: Jaideep Taggart Singh
Graduate Students: Erin White, Gordon Arrowsmith-Kron, Nicholas Nusgart, Aiden Boyer, Yousuf Alishan
Undergraduate Students: Skylar Milne, Lindsey Hickman, Rashawn Carter, Aesen Copeland, Myles Daugherty, Tanusree Makwana, Nick Koester, Graham Malone

U.S. DEPARTMENT OF ENERGY | Office of Science

DOE Early Career Award 2018
DE-SC0019015 (EDM3)
DE-SC0019455 (Ra EDM)
DE-SC0025679 (Ra EDM+EDM3)

BACKUP SLIDES

This material is based upon work supported by the U.S. Department of Energy, Office of Science, Office of Nuclear Physics and used resources of the Facility for Rare Isotope Beams (FRIB) Operations, which is a DOE Office of Science User Facility under Award Number DE-SC0023633.
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EDMs are screened in atomic systems resulting in nuclear Schiff moments

Neutral diamagnetic atoms: all electrons are paired

- Shielding in diamagnetic atoms
  - Schiff Phys. Rev. 132:2194 (1963)

Schiff Moment

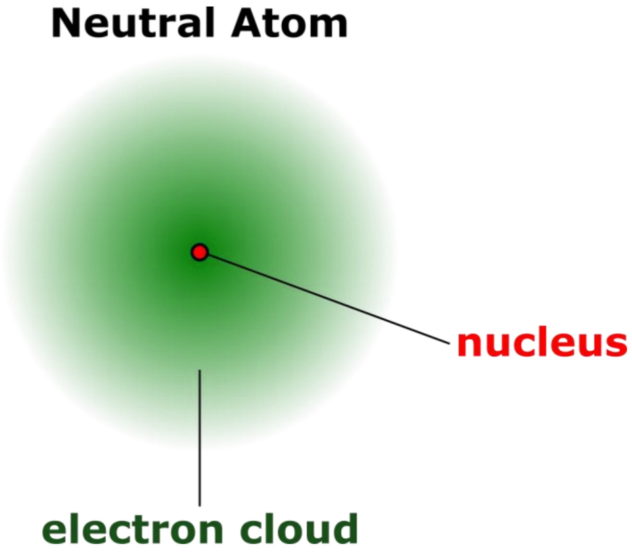
$$\vec{S} = \frac{\langle er^2\,\vec{r}\rangle}{10} - \frac{\langle r^2\rangle\langle e\vec{r}\rangle}{6}$$

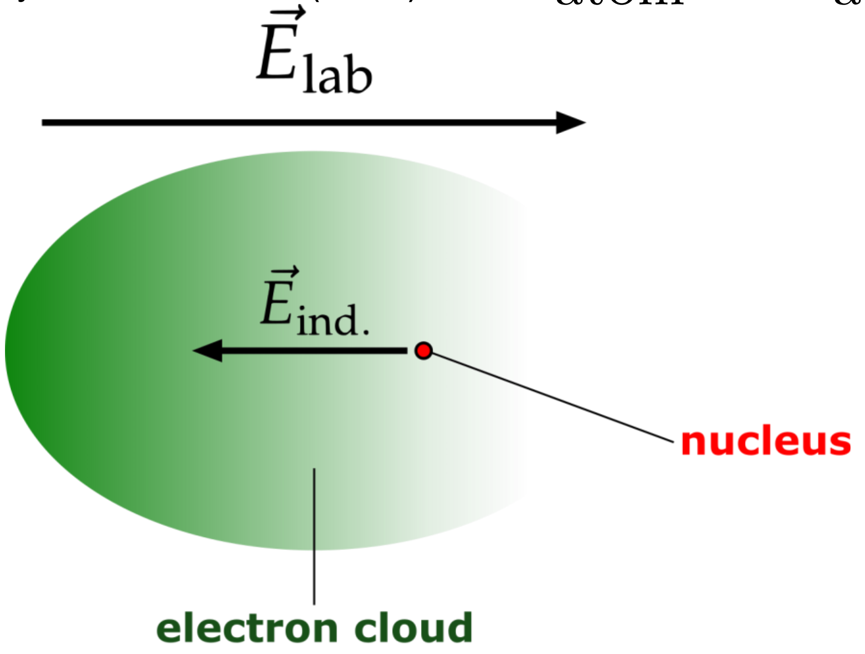
Sushkov et al. Zh. Eksp. Teor. Fiz 87:1521 (1984)

Relativistic atoms: Sandars-Bouchiat Z^3 scaling

- Shielding imperfect, leaving residual T-violating observable
- Atomic parity violating effects scale like Z^3
  - Sandars Phys. Lett. 14:194 (1965), Bouchiat and Bouchiat Phys. Lett. B 2:111 (1974)

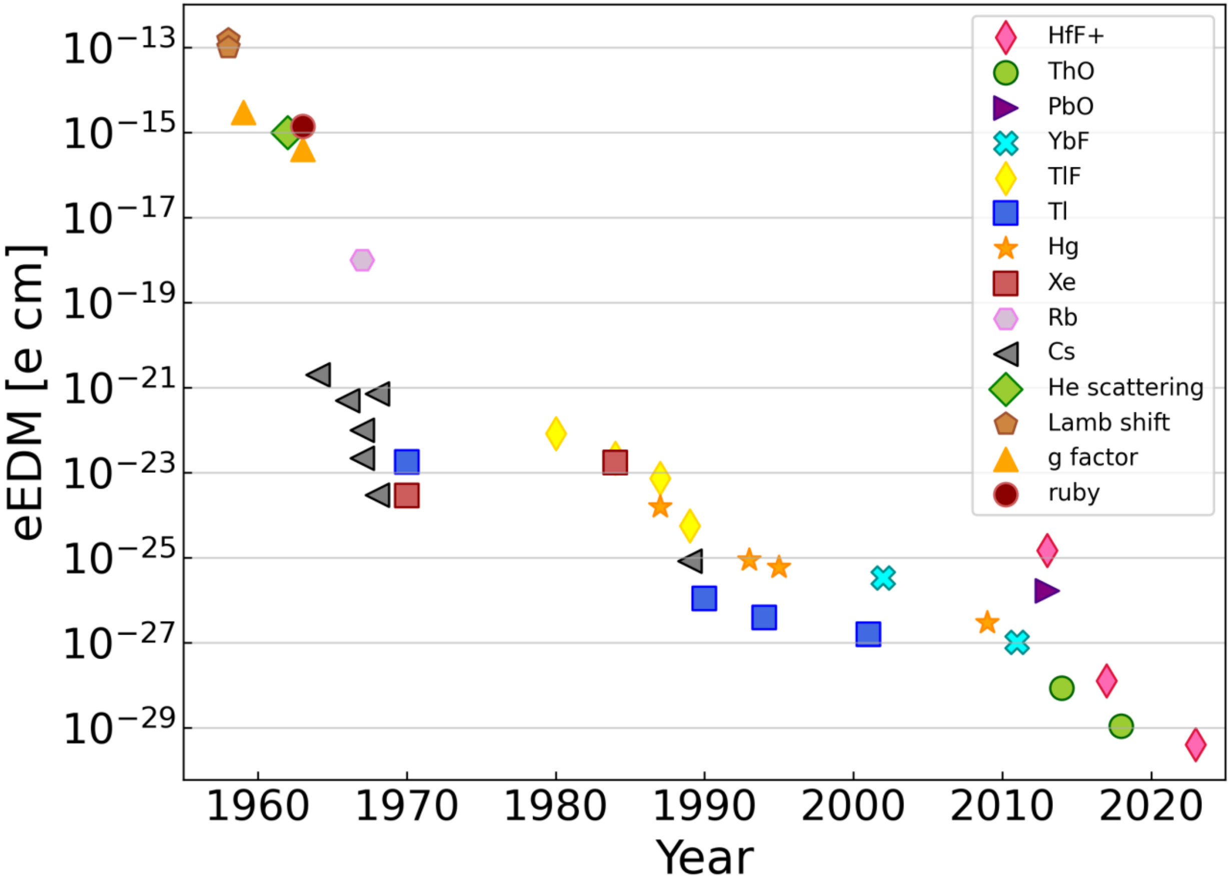
$$\vec{d}_{\text{atom}} = \kappa_{\text{atom}} Z^3 \vec{S}$$

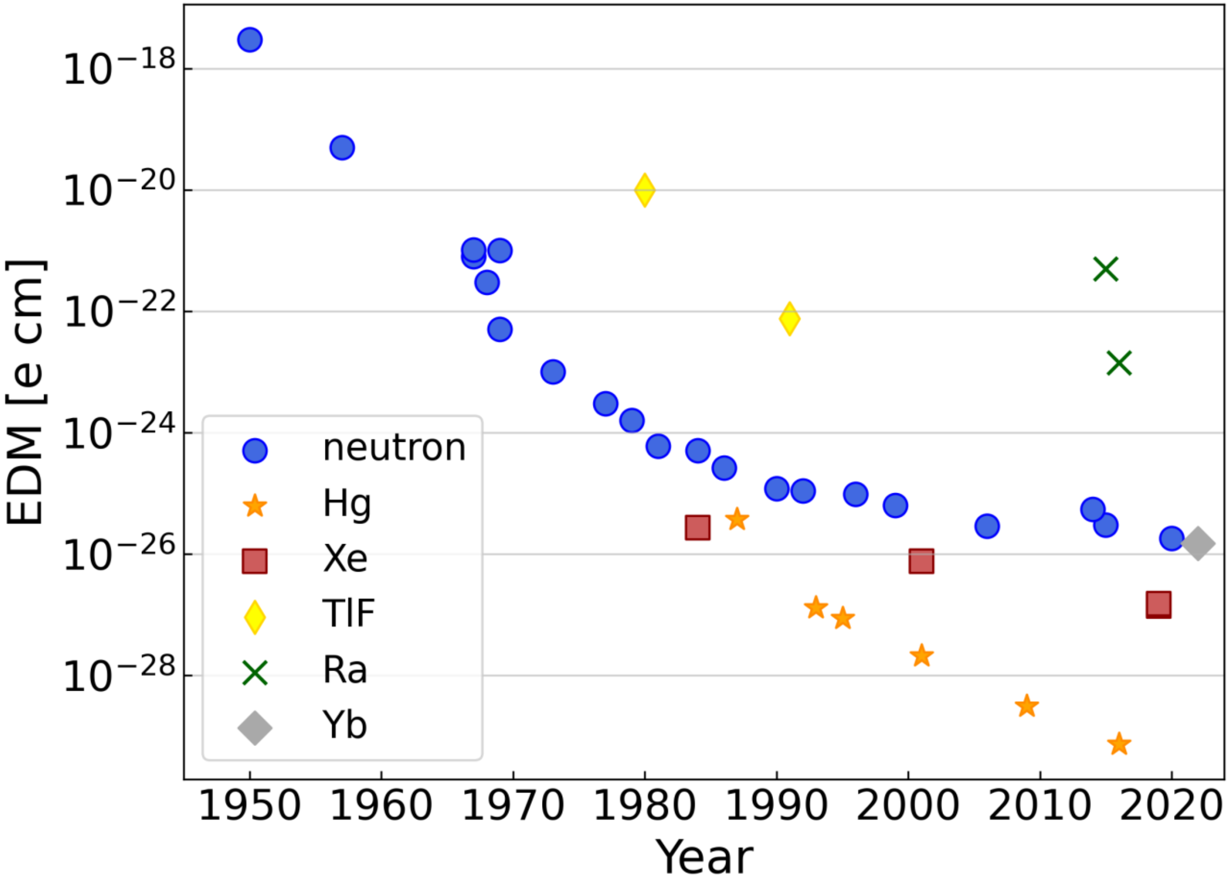




$$\vec{E}_{\text{ind.}} \approx -\vec{E}_{\text{lab}}$$

Pushing the Limits of Sensitivity with Atomic Systems: How Can We Improve?





A path towards more efficient production:
Sub-ambient Pressure Nanoelectrospray Ionization (SPIN)

- Atmospheric electrospray has two identified challenges:
  - Gas dynamics at the interface between atmosphere and the vacuum chamber induce heavy losses

Kebarle and Tang Analytical Chem 65:A972 (1993)

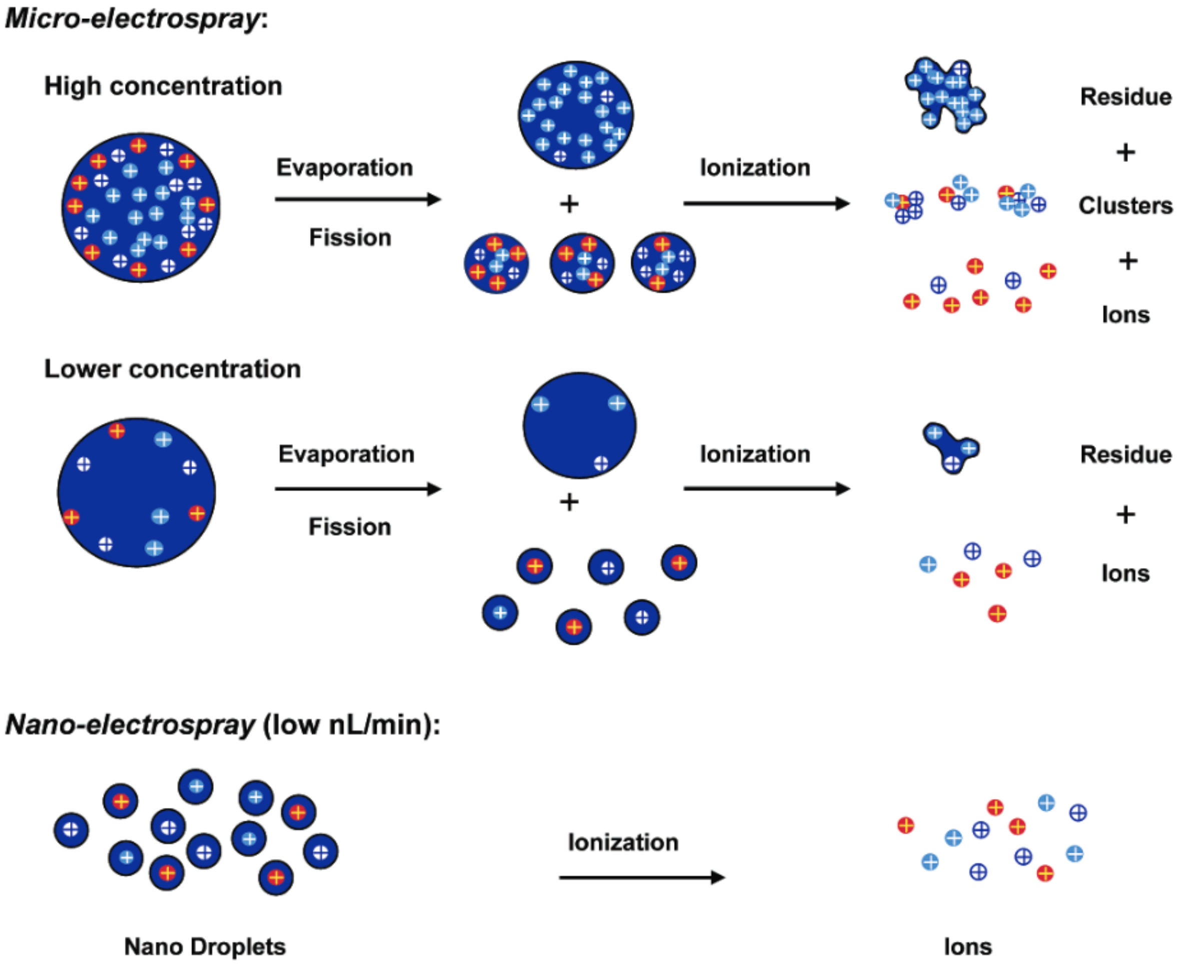
  - Liquid flow rates produce larger droplets, less likely to produce a gas-phase ion you are interested in

Smith et al. Acc. Of Chem. Research 37:269 (2004)

- Lower flow rates provide more efficient ionization
  - High ionization efficiency: droplet size is reduced and contain one analyte molecule on average

Gale and Smith Rapid Comm. In Mass. Spectrom. 7:1017 (1993)

Wilm and Mann Analytical Chem. 68:1 (1996)



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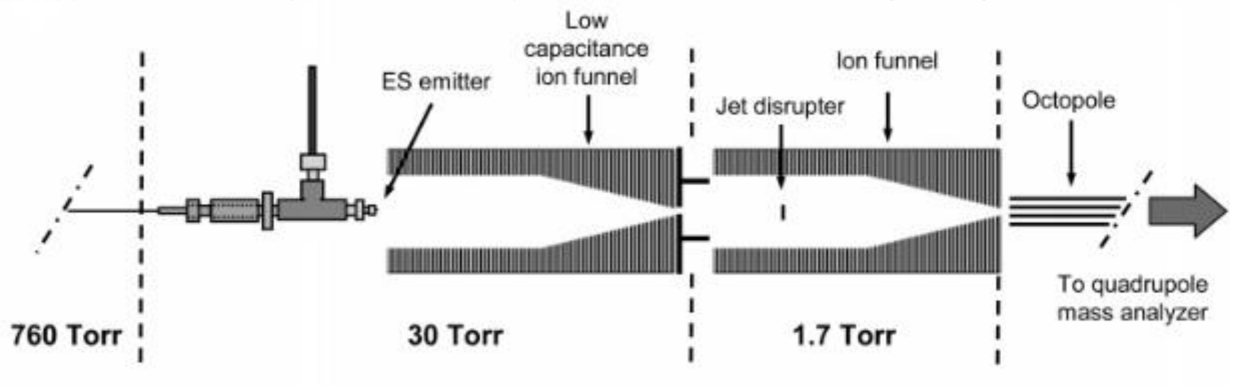
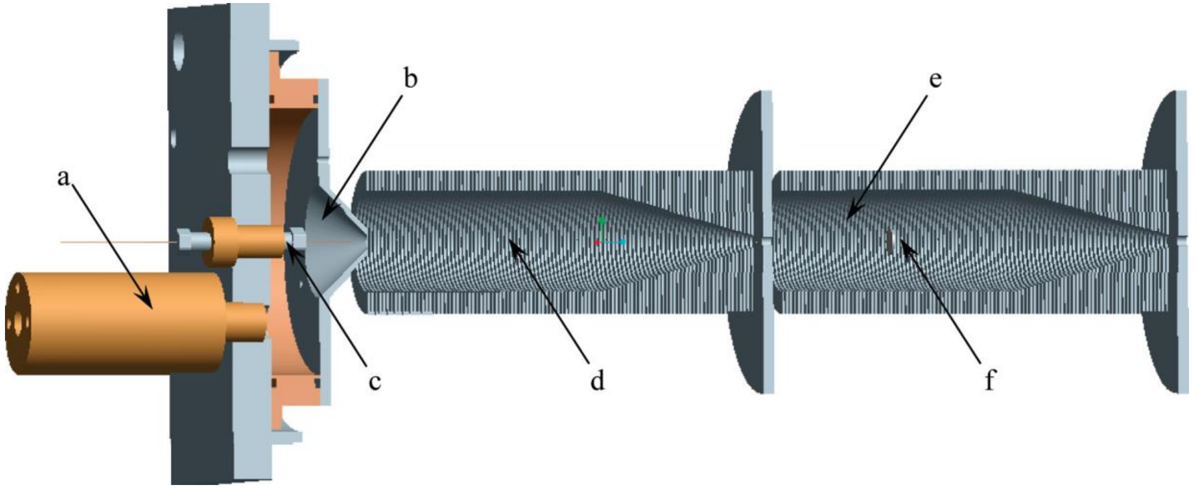
Wilm and Mann Analytical Chem. 68:1 (1996)

- Reduce pressure differential between electrospray and vacuum chamber
  - Allows placement of electrospray probe in vacuum chamber

Page et al. Analytical Chem 80:1800 (2008)





2nd generation SPIN source: 50% ionization efficiency

Marginean et al. Analytical Chem. 82:9344 (2010)

We Needed To Simulate Ion Funnel Performance For “Low” m/z

- Nick Nusgart has more details