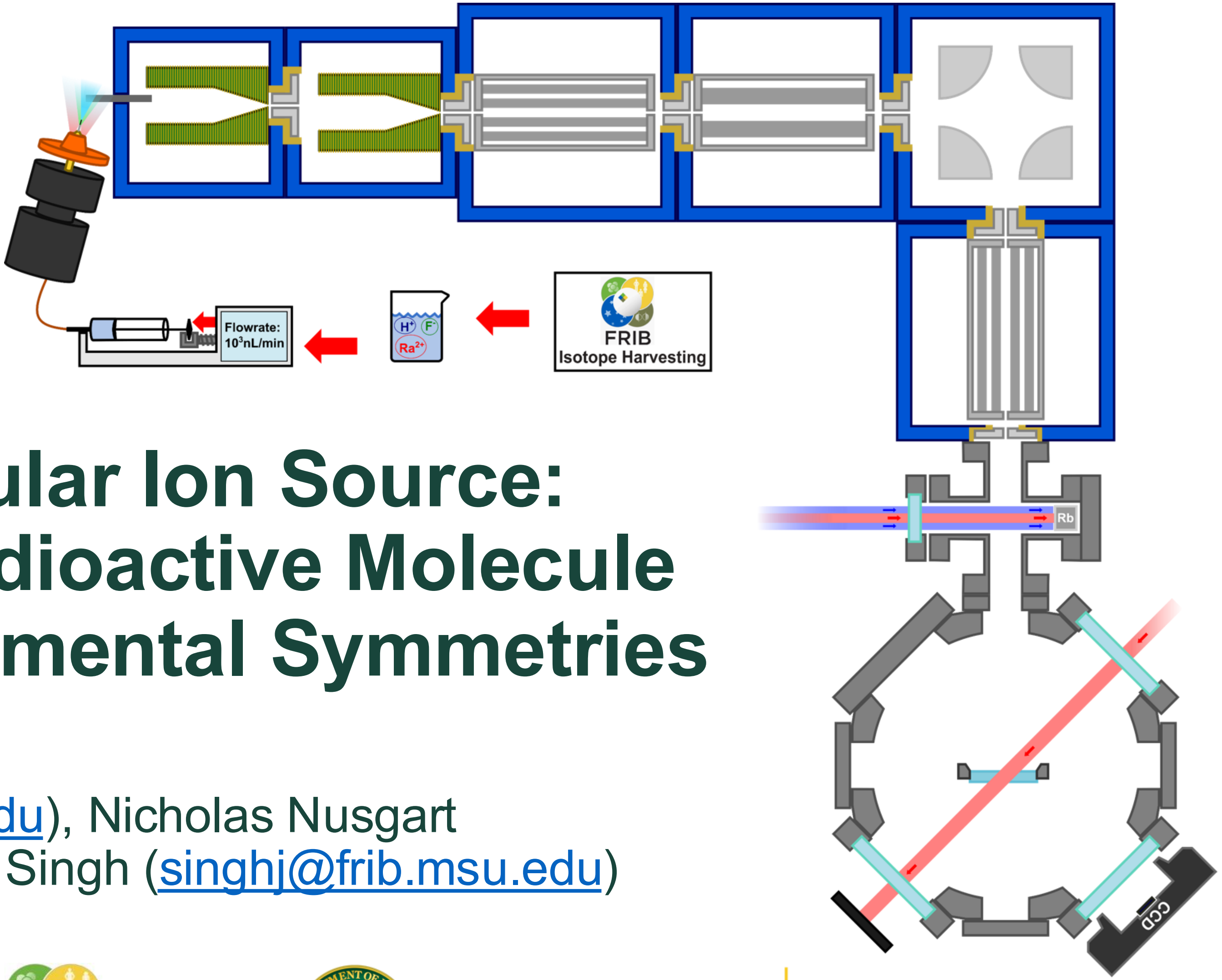
FRIB-EDM3 Talk 2/2

June 10, 20:00 – 20:20 (18+2)

15th Conference on the Intersections of Particle and Nuclear Physics

Memorial Union, Pan Hellenic Room, University of Wisconsin-Madison



The FRIB-EDM3 Molecular Ion Source: Designing An Efficient Radioactive Molecule Source For Tests Of Fundamental Symmetries

Aiden Boyer (boyera@frib.msu.edu), Nicholas Nusgart (Nusgart@frib.msu.edu), Jaideep Taggart Singh (singhj@frib.msu.edu)

MICHIGAN STATE UNIVERSITY

FRIB

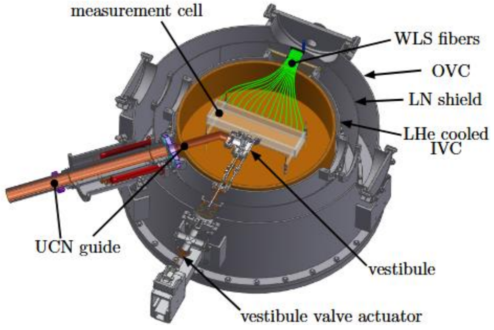
U.S. DEPARTMENT of ENERGY | Office of Science

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's A Lot Of Exciting Topics Being Discussed At CIPANP! Precision Measurements, Fundamental Symmetries, BSM

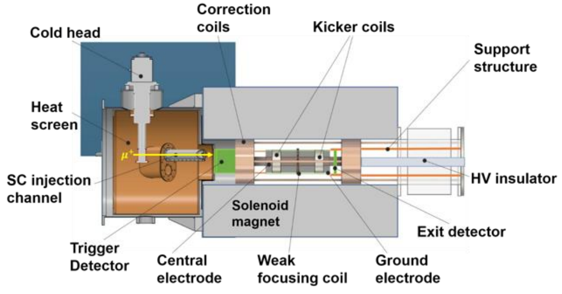
Fundamental Symmetries: Permanent Electric Dipole Moments

Skyler Degenkolb, Wednesday June 11, 08:15



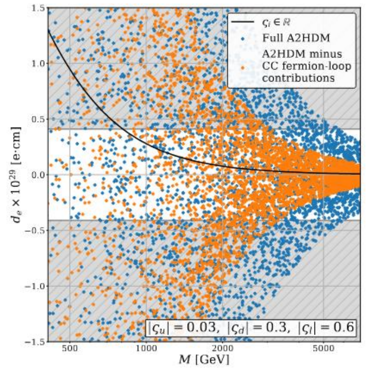
SNS nEDM → nEDMSF

Ciancolo et al. https://arxiv.org/abs/2411.03337v2 (2025)



Muon EDM – Phase I

Hu et al. http://arxiv.org/abs/2502.11186 (2025)



Model-independent eEDM contributions

Dávila et al. http://arxiv.org/abs/2504.16700 (2025)

Decay Recoil Spectroscopy With Superconducting Quantum Sensors

Kyle Leach, Tuesday June 10, 11:00

THE BeEST SALER

Bray et al. JLTP 218:74 (2025)

http://arxiv.org/abs/2411.08076

Cosmic Microwave Background: Upcoming And Ongoing Measurements

Maximilliano Silva-Feaver, Wednesday June 11, 10:45

SIMONS OBSERVATORY CMB-S4 Next Generation CMB Experiment

Logos: Simons Observatory, CMB-S4

Weak Sector Symmetry Violations: Electron Scattering, Polarized Neutrons

NOPTREX Neutron Optical Parity and Time-Reversal EXperiment

MOLLER

Logos: NOPTREX, MOLLER

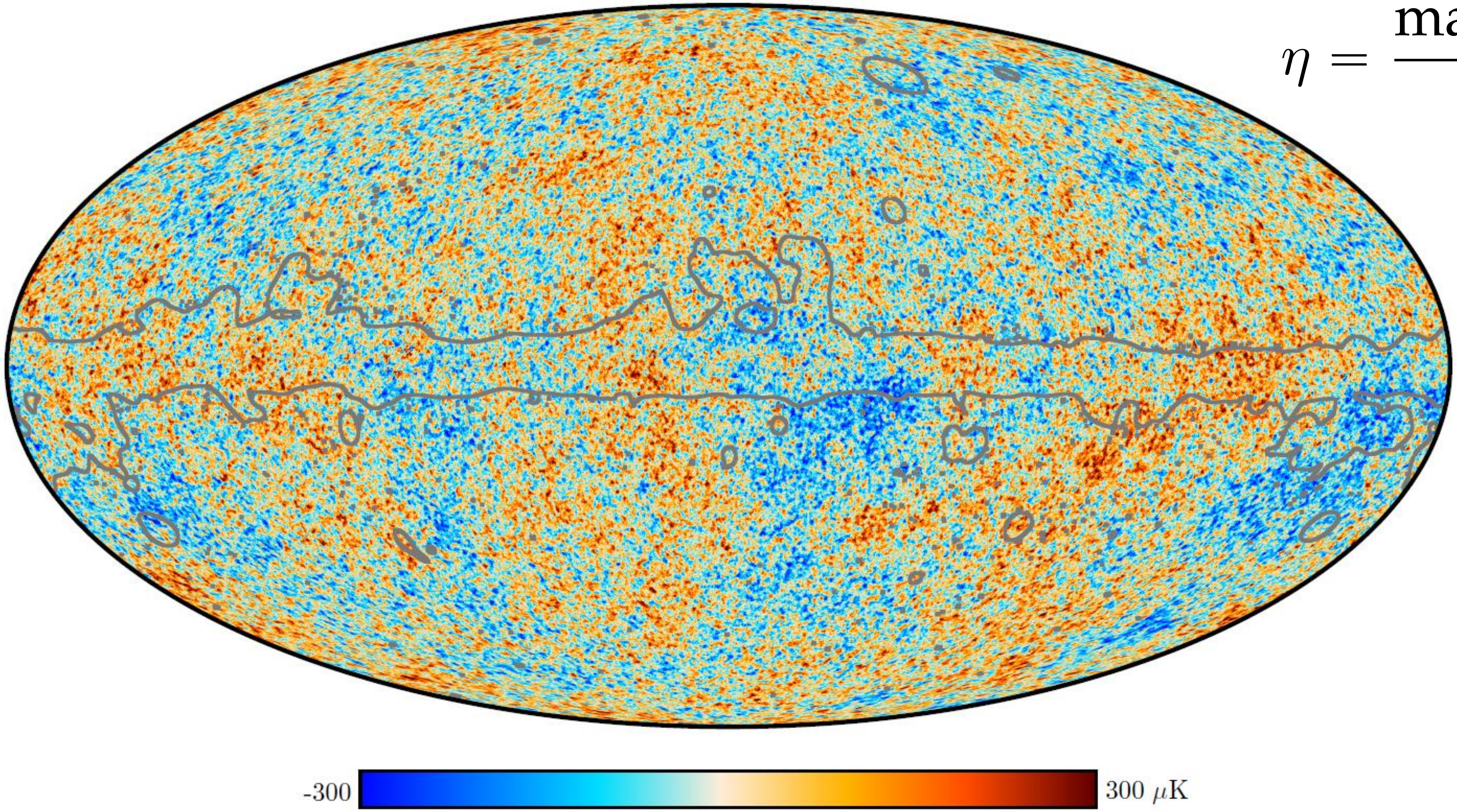
Beyond Standard Model Searches @ LHC

Shivani Lomte, Thursday June 12, 10:10

CMS ATLAS EXPERIMENT

Logos: CMS, ATLAS

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

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

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

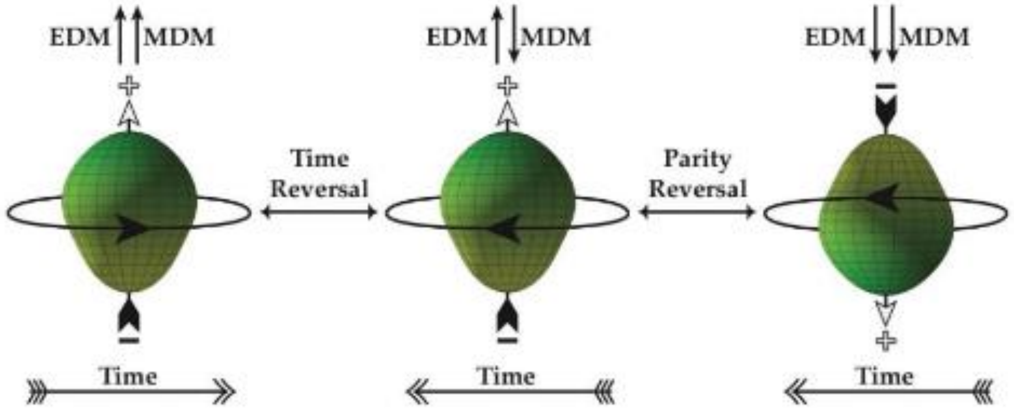
PDG2024

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

Huet & Sather PRD 51:379 (1995)

Permanent Electric Dipole Moments: 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

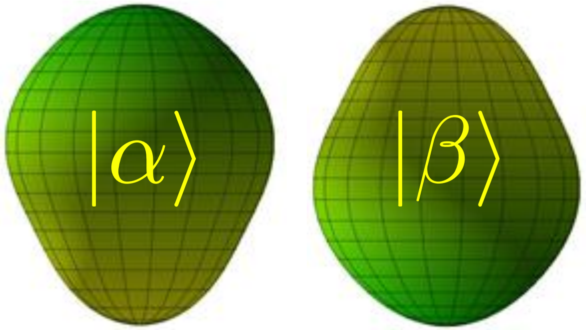
CP-Violating Observable In Diamagnetic Atoms: Nuclear Schiff Moment With Pear-Shaped Radium-225

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

$$S \equiv \langle \Psi_0 | S_z | \Psi_0 \rangle = \sum_{k \neq 0} \frac{\langle \Psi_0 | S_z | \Psi_k \rangle \langle \Psi_k | V_{\not{P}\not{T}} | \Psi_0 \rangle}{E_0 - E_k} + \text{c.c.}$$

Western Pear In A Chair (2016) by Audry Handler
Chazen Museum of Art

Ex: ^{225}Ra Parity Doublet



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

55 keV

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

- **Nearly degenerate parity doublet**

  Haxton & Henley PRL 51:1937 (1983)

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

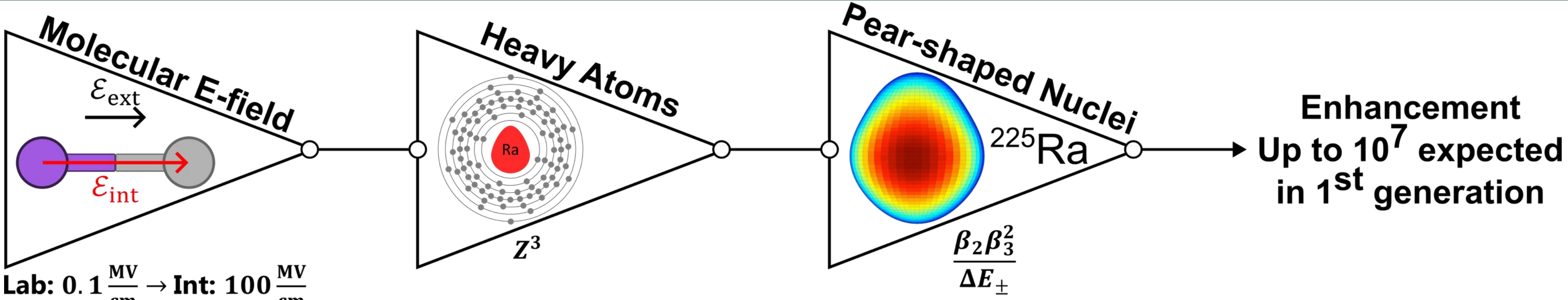
  Auerbach, Flambaum, & Spevak PRL 76:4316 (1996)

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)

Taking Inspiration From Leptonic EDM Searches: Combine Polar Molecules With Pear-Shaped Nuclei



Lab: $0.1\,\frac{\text{MV}}{\text{cm}} \rightarrow$ **Int:** $100\,\frac{\text{MV}}{\text{cm}}$

Solids: $3\,\frac{\text{MV}}{\text{cm}}$

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

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

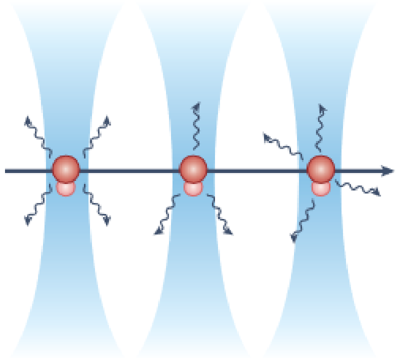
- Polar molecules have been demonstrated as an ultrasensitive tool for electron EDM searches
  - Easy to align molecule dipole moment with applied field
  - Molecule dependent co-magnetometry via energy splittings
  - Large internal fields produce larger splittings

- 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 Slow And Trap Radioactive Molecules

Laser Cooling & Trapping



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

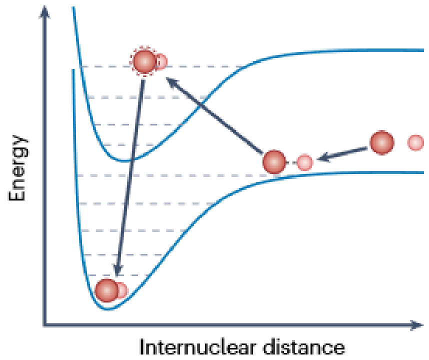
Opportunity

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

Challenge

- Complex cooling schemes, Low efficiency, ~1%

Ultracold Assembly



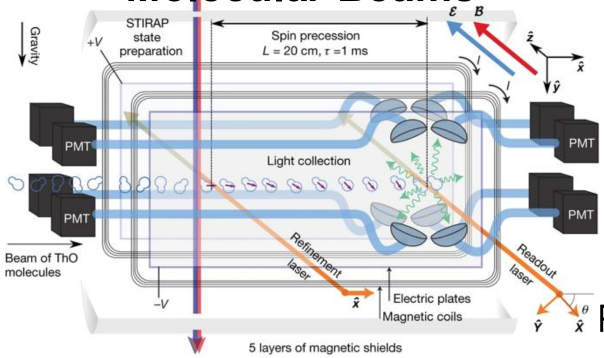
Opportunity

- Could be very efficient path to ultracold molecules

Challenge

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

Molecular Beams



Panda (Harvard PhD Thesis, 2018)

Opportunity

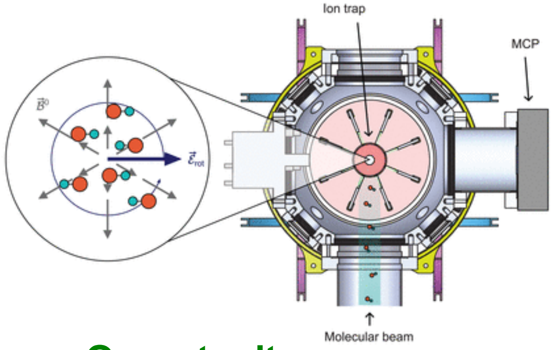
- Lots of progress in making cold, intense beam of stable molecules

Challenge

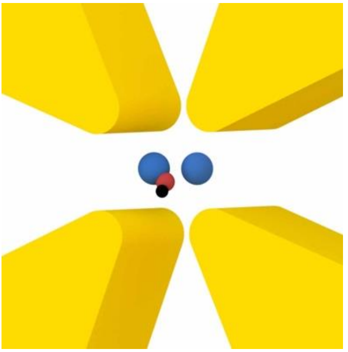
- Efficiency not characterized or optimized

Ion Trapping

Roussy et al. Science 381:46 (2023)



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



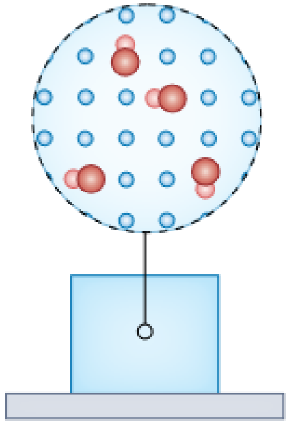
Opportunity

- 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 ($1mm^3$)

Challenge

- Inhomogeneities, broad linewidths obscure sensitivity

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

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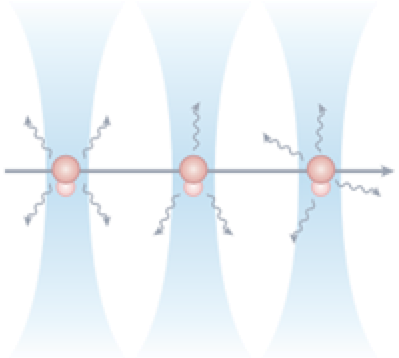

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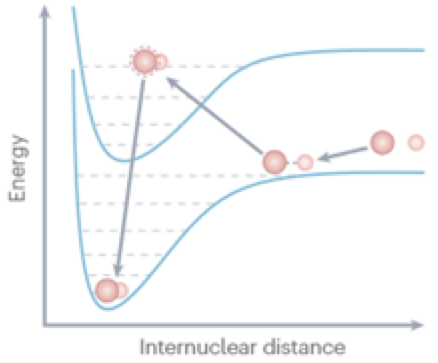

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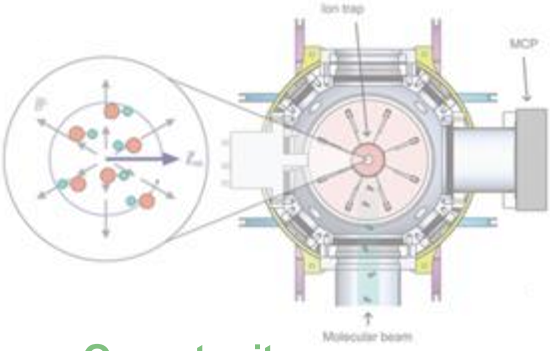

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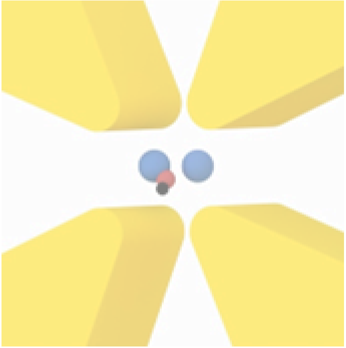

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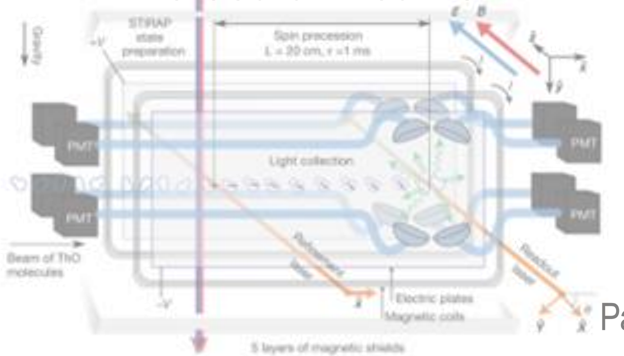

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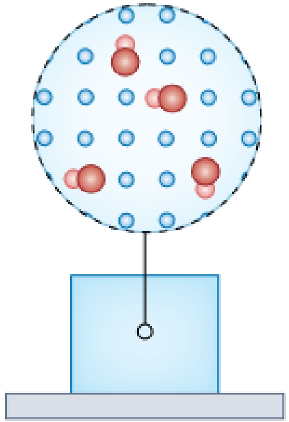
Matrix Isolation

Nicholas Nusgart
Tuesday June 10
19:40 – 20:00



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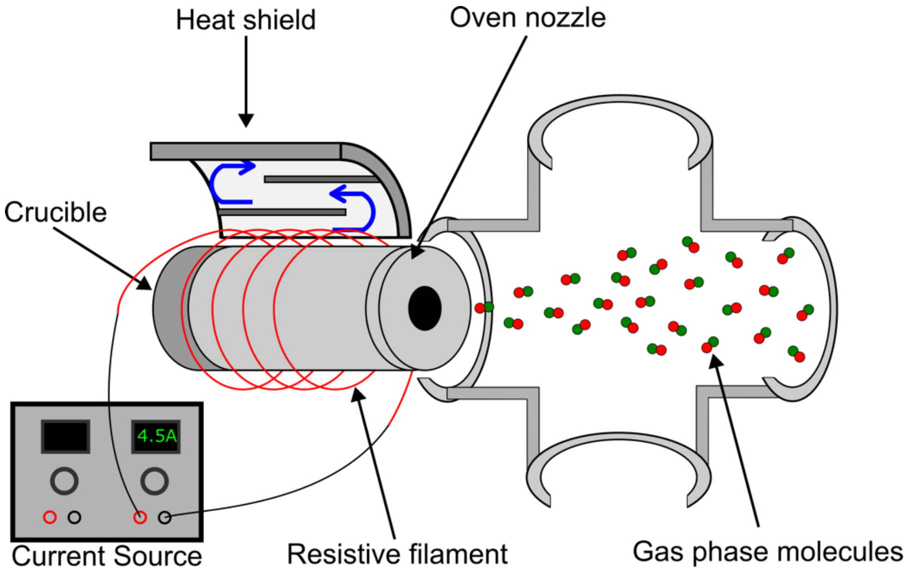
There Are Also Several Ways To Produce Molecules

Most Brute Force: High Temperature Oven



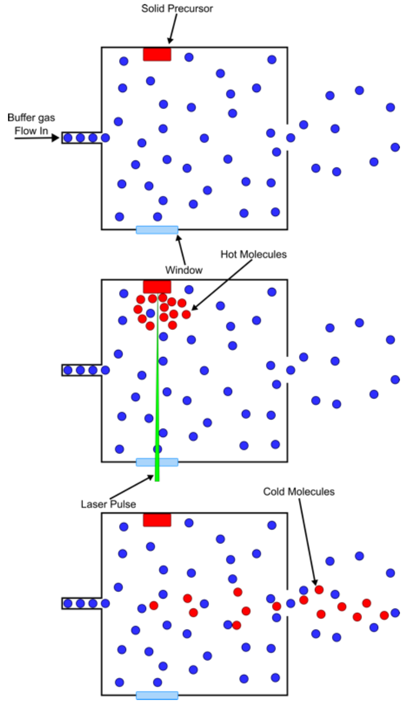
Opportunity

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

Challenge

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

Less Brute Force: Laser Ablation + Cold Buffer Gas



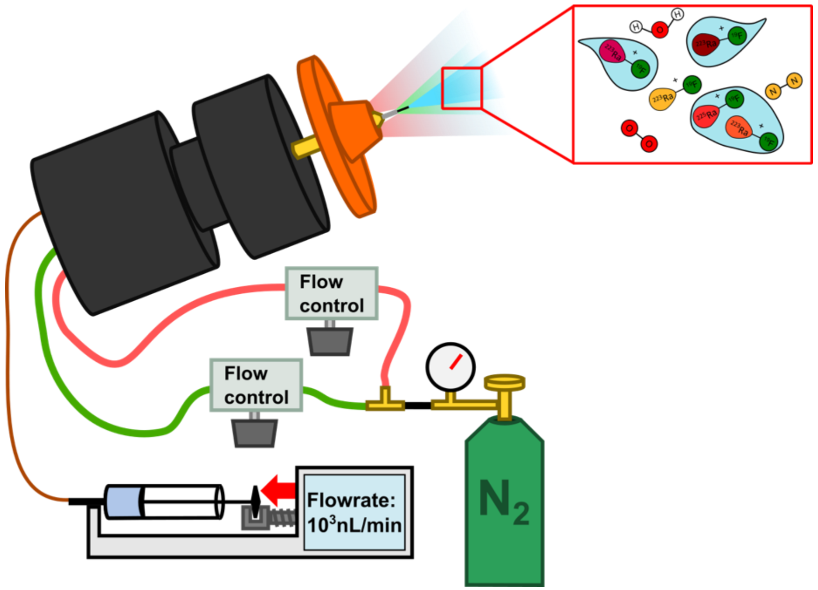
Opportunity

- 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

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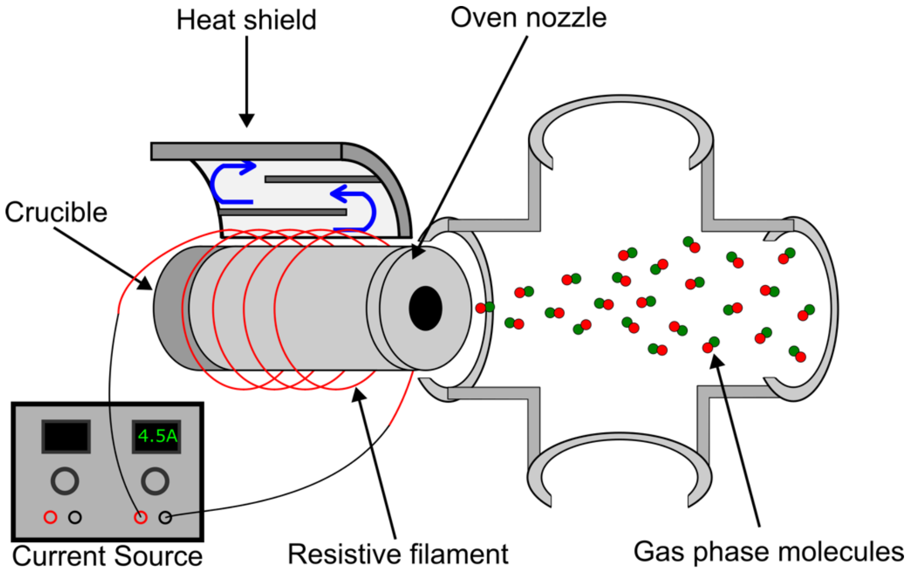

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(Gas phase neutral atoms)
Gordon Arrowsmith-Kron
Tuesday June 10
19:20 – 19:40

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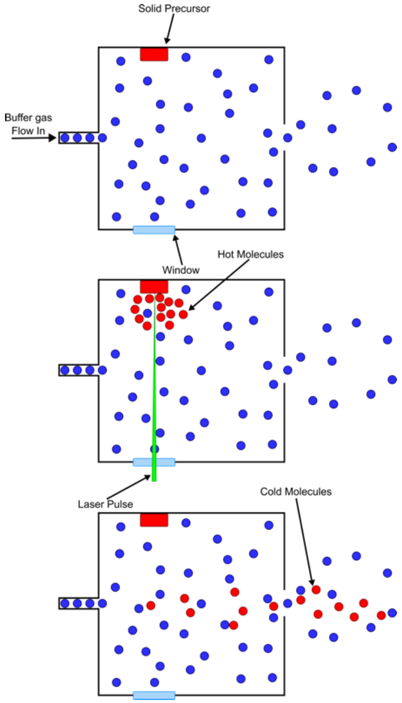

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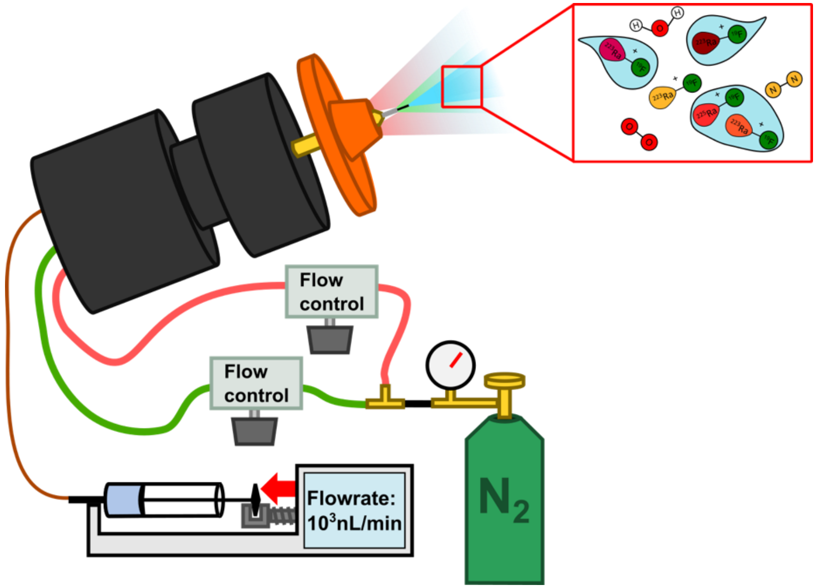

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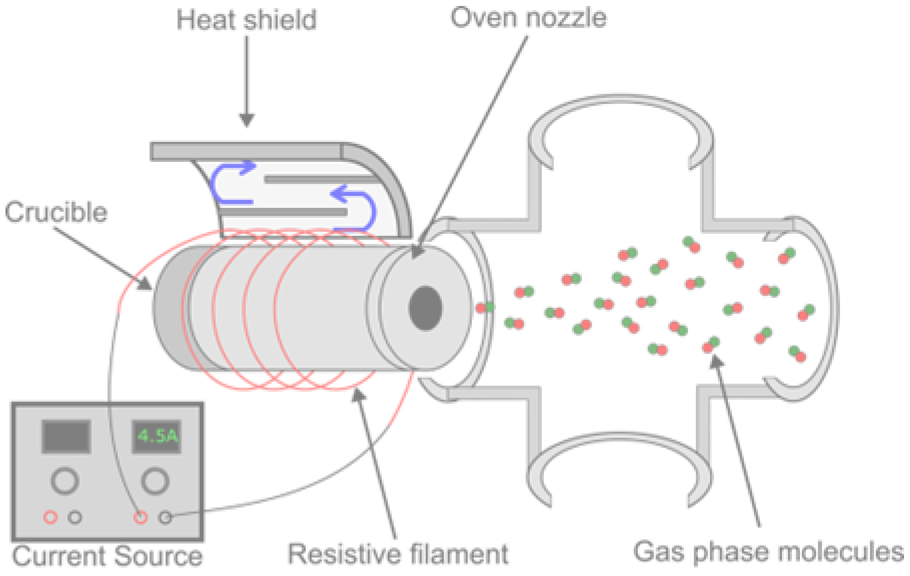

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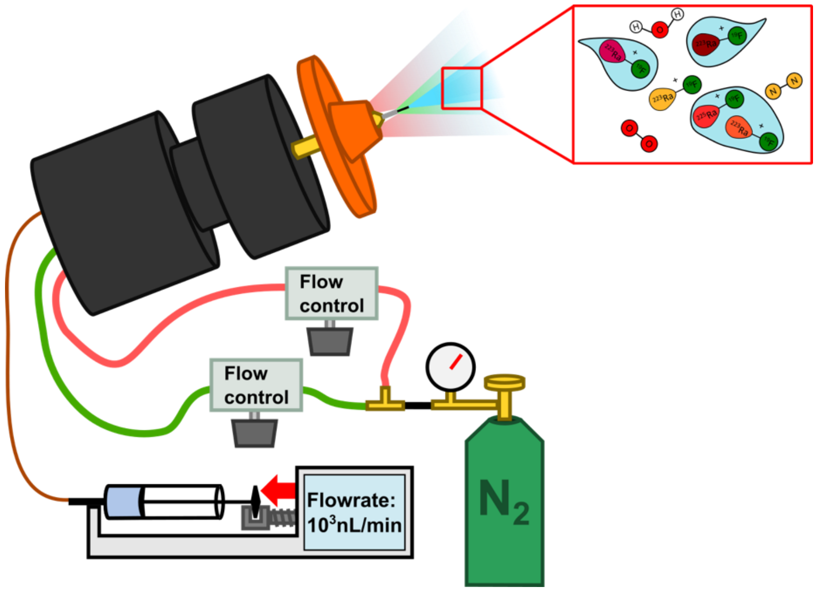
Exotic And Weird: Electrospray Ionization



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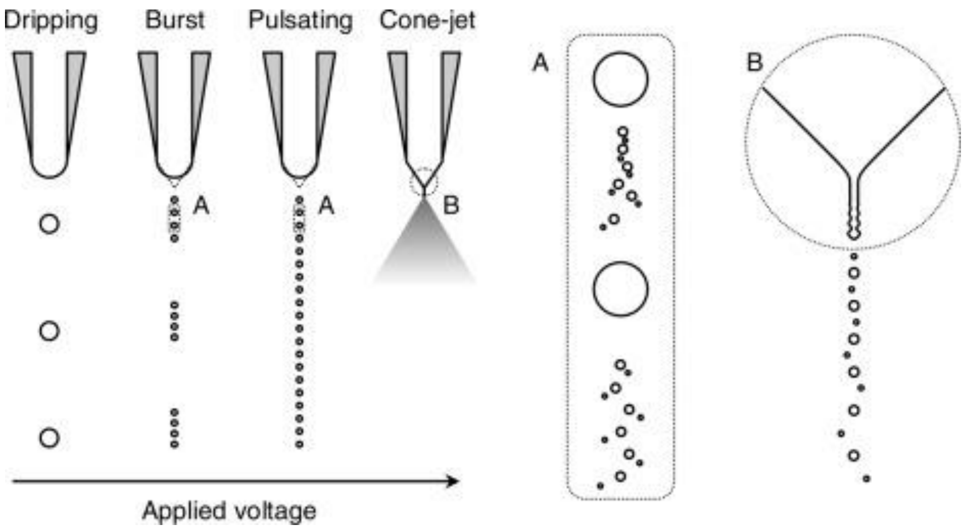
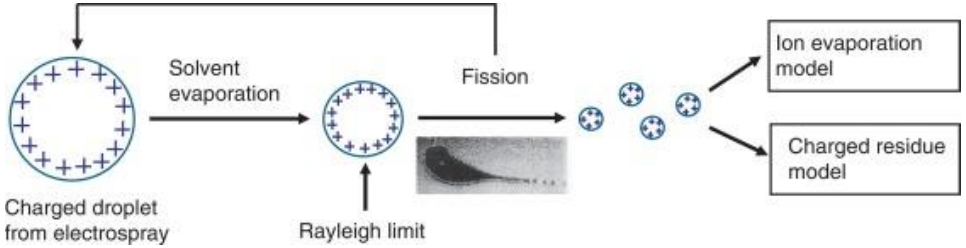
Electrospray Ionization: It Could Be Very Efficient For Forming Radioactive Molecules



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

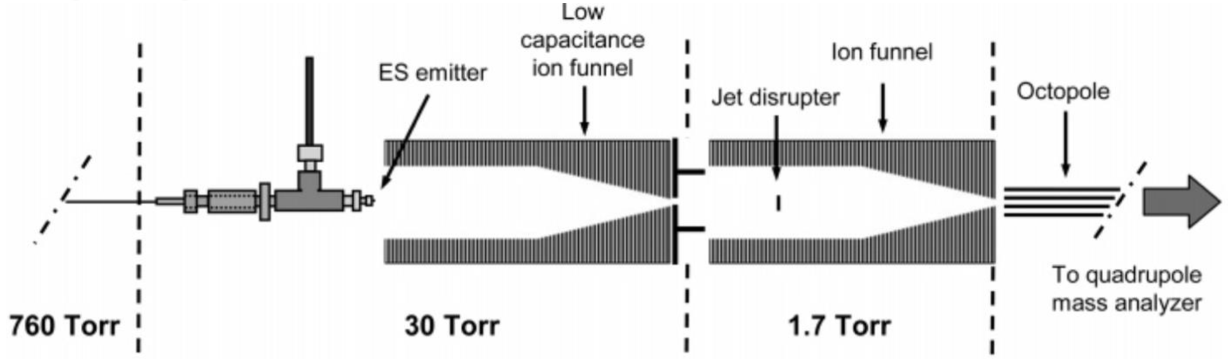
Inefficient ionization and poor transmission of the charged species produced by an electrospray from the ambient pressure mass spectrometer source into the high vacuum region required for mass analysis significantly limits achievable sensitivity. Here, we present evidence that, when operated at flow 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

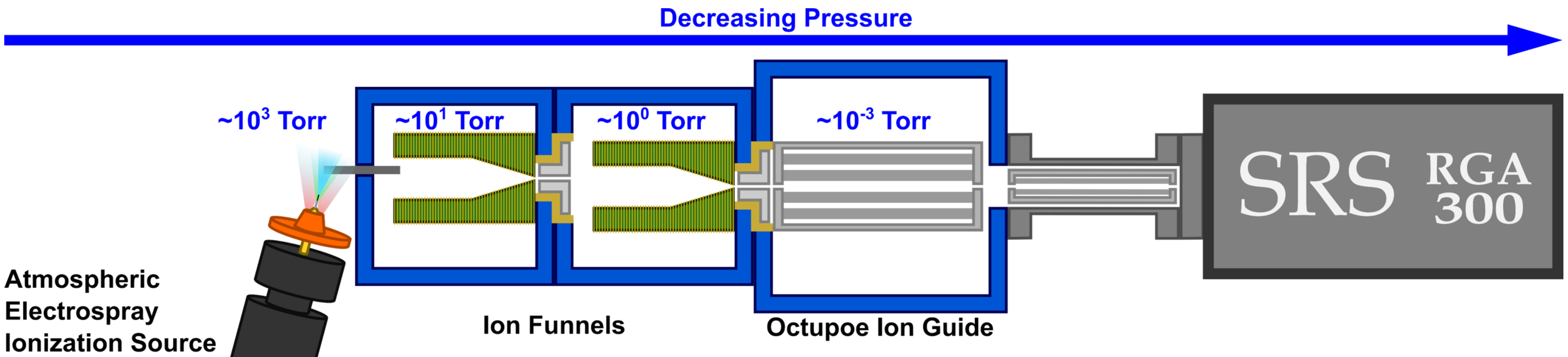
Subambient Pressure Ionization with Nanoelectrospray Source and Interface for Improved Sensitivity in Mass Spectrometry

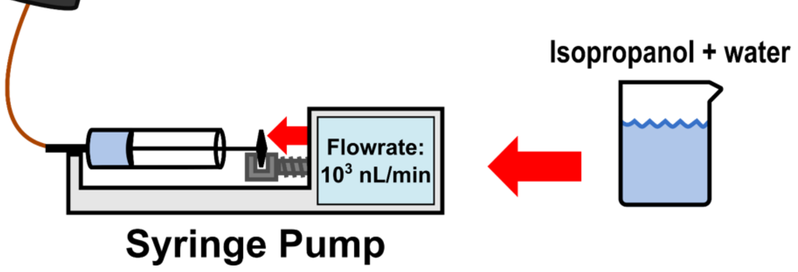
Jason S. Page, Keqi Tang, Ryan T. Kelly, and Richard D. Smith*

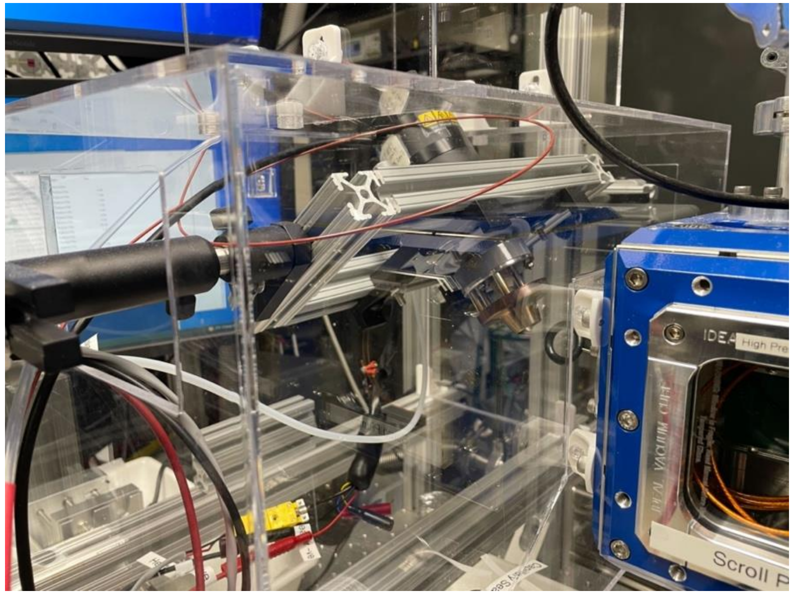
Biological Sciences Division, Pacific Northwest National Laboratory, P.O. Box 999, Richland, Washington 99352



We Have Prior Experience With Electrospray



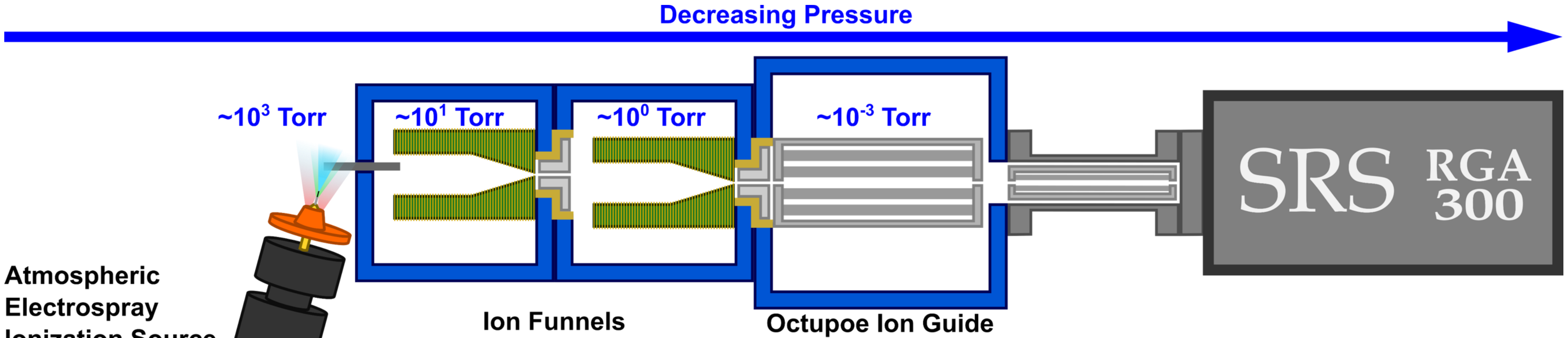


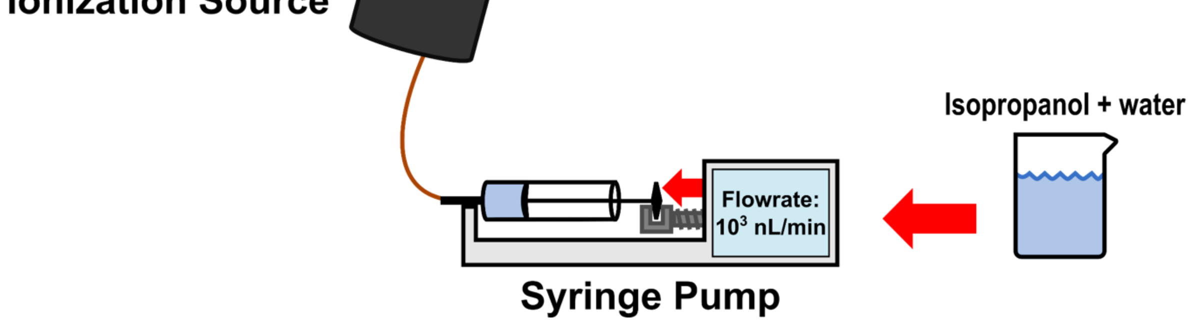


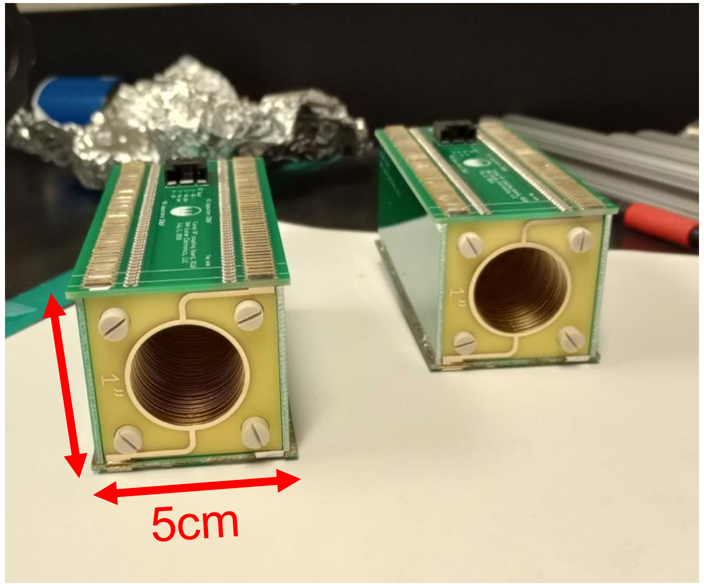
Electrospray Ionization Source:

- DC High Voltage (3-5 kV)
- Collinear gas flow
- Difficult to precisely position and align
- Subject to atmospheric conditions of room

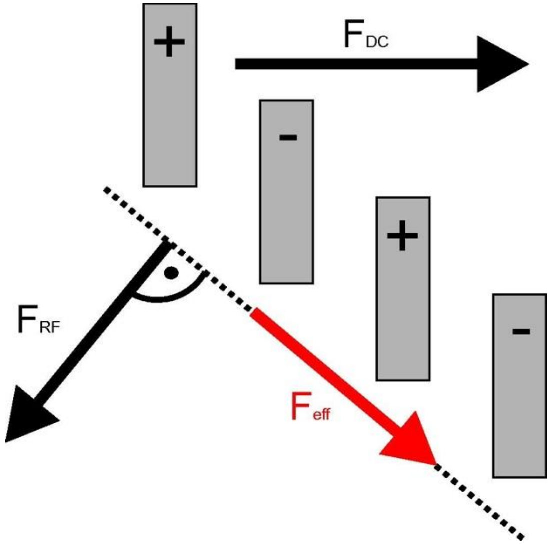
We Have Prior Experience With Electrospray







GAA Custom Electronics (Gordon Anderson)

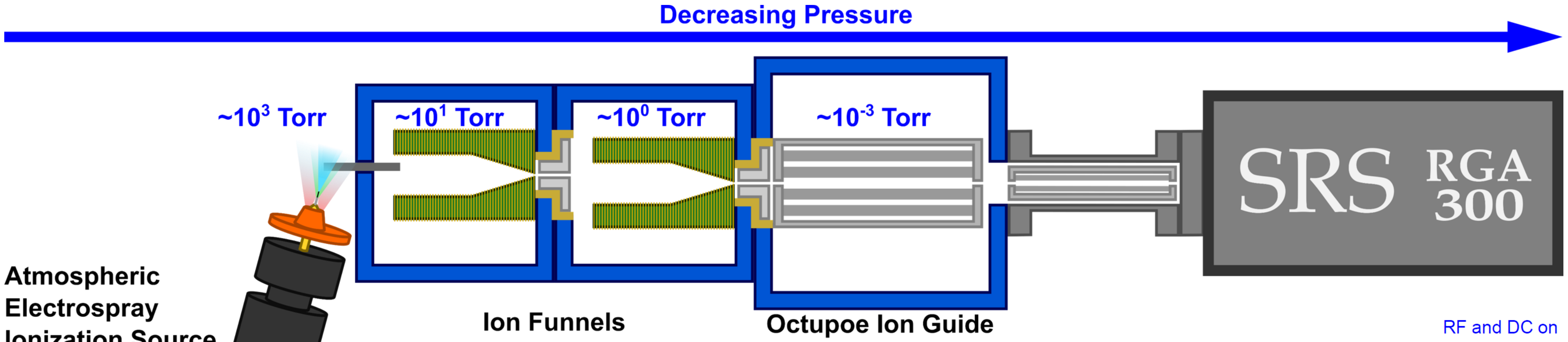


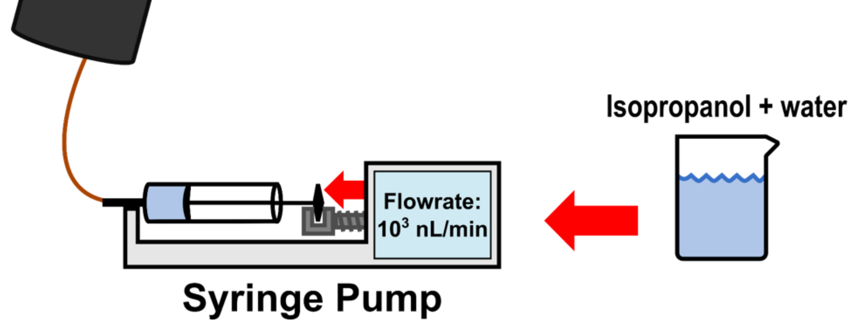
Droese et al. NIMB 338:126 (2014)

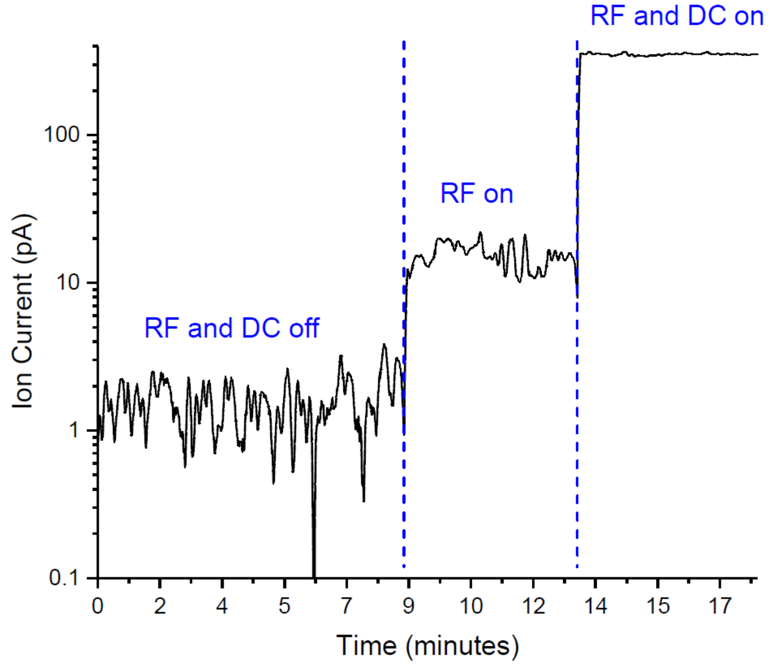
Electrodynamic Ion Funnels:

- DC Gradient (~ 130 V)
- Alternating-phase RF (250-500 V, 2-3 MHz) V_{pp}
- Commercial product complete with electronics to operate

We Have Prior Experience With Electrospray







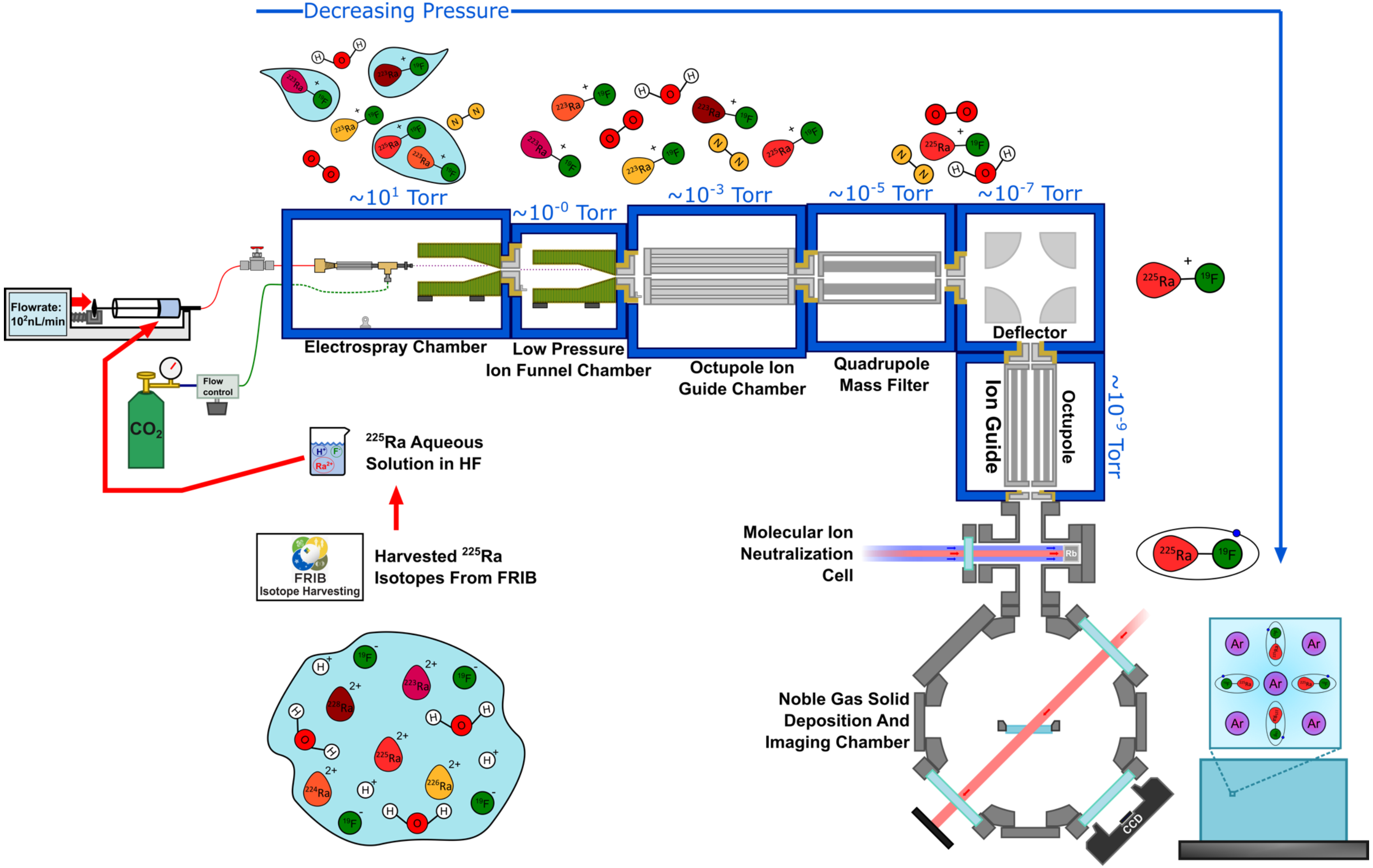
Stable ion beams are possible:

- Magnitude and stability affected by:
    - Applied DC/RF
    - Ambient conditions around electrospray
    - Solution flow rate into electrospray source
    - Source positioning and alignment

Challenges:

- Low reproducibility of test conditions
- Lack of control over positioning and alignment

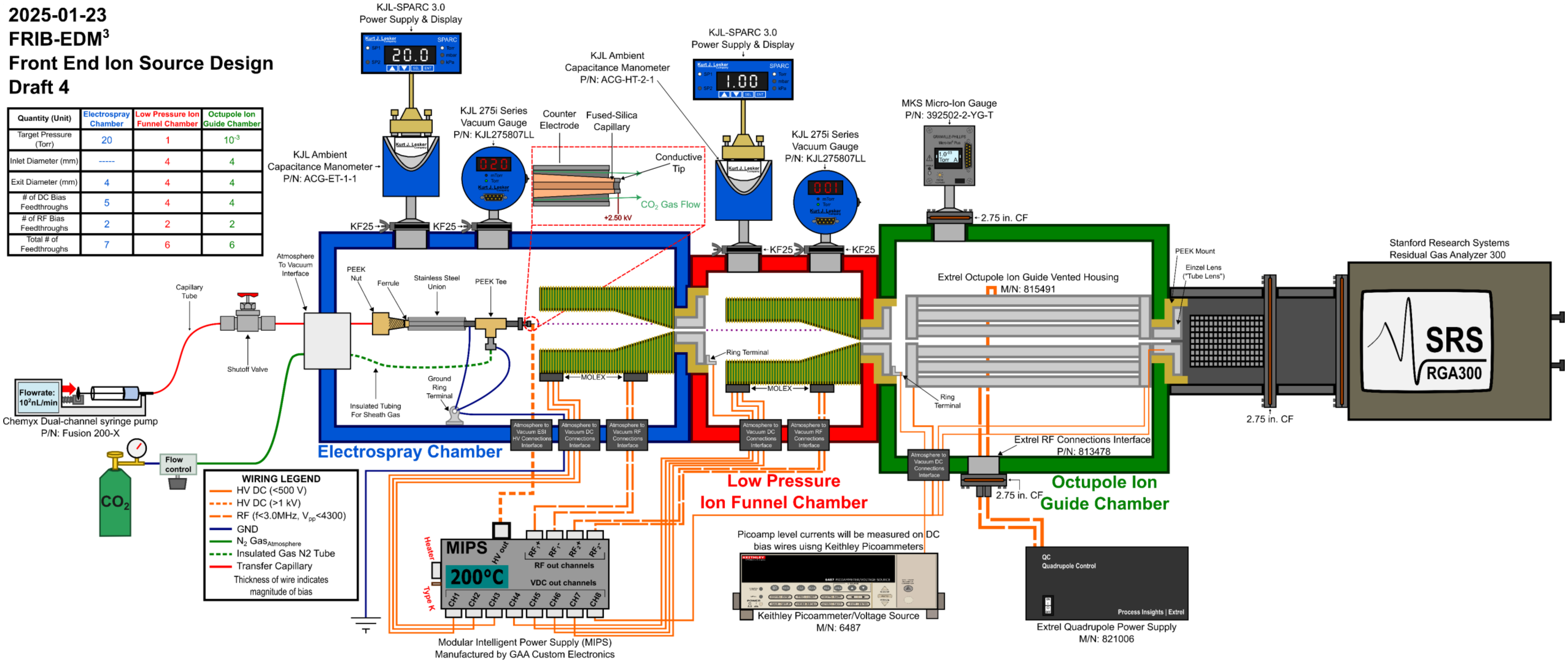
The FRIB-EDM3 Instrument Plan: Co-deposit Neutral Molecules With Noble Gas Atoms Onto A Cryogenic Substrate



Current Activities: Complete Redesign of Front-End Interface

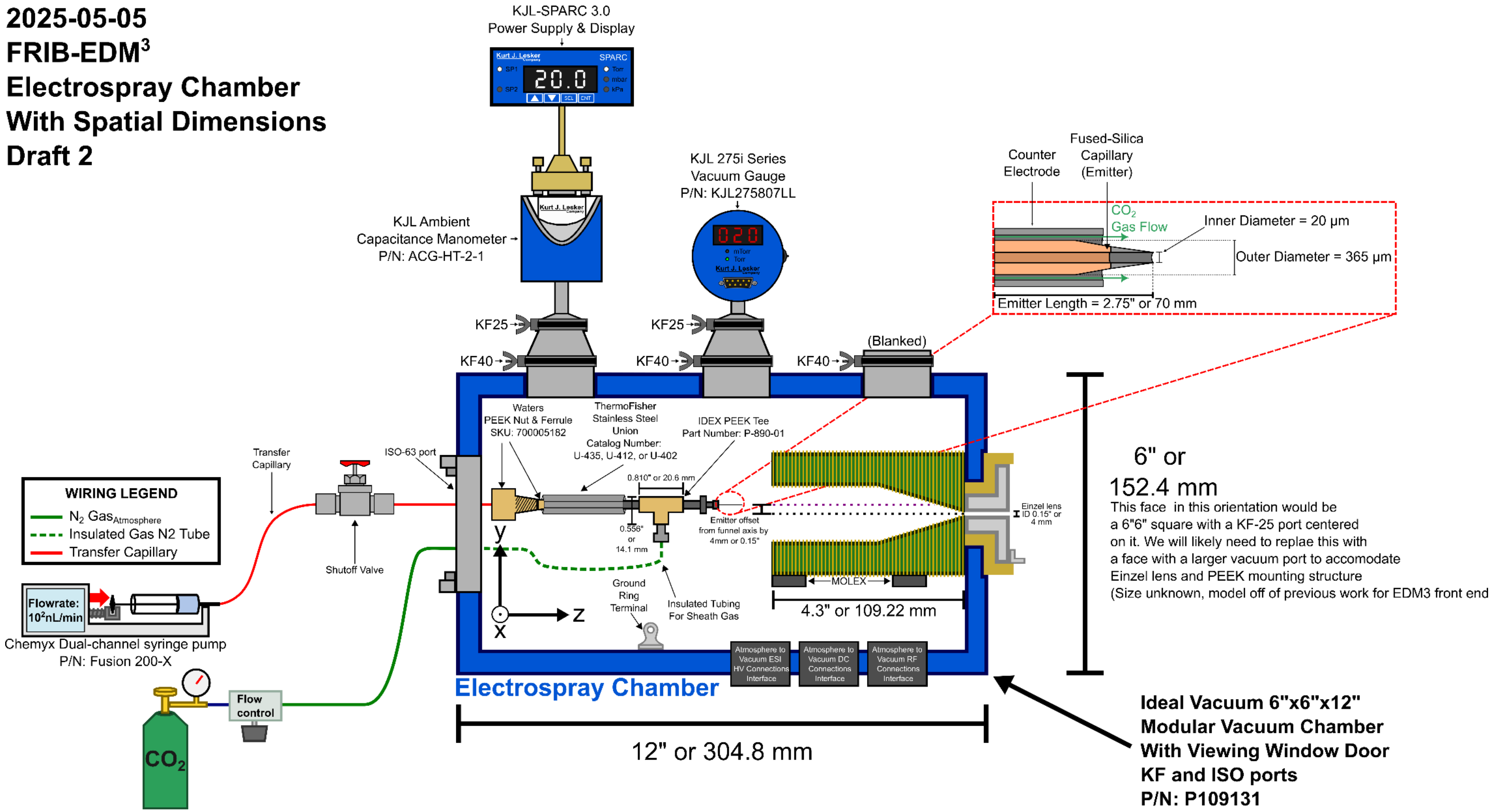
2025-01-23
FRIB-EDM³
Front End Ion Source Design
Draft 4

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

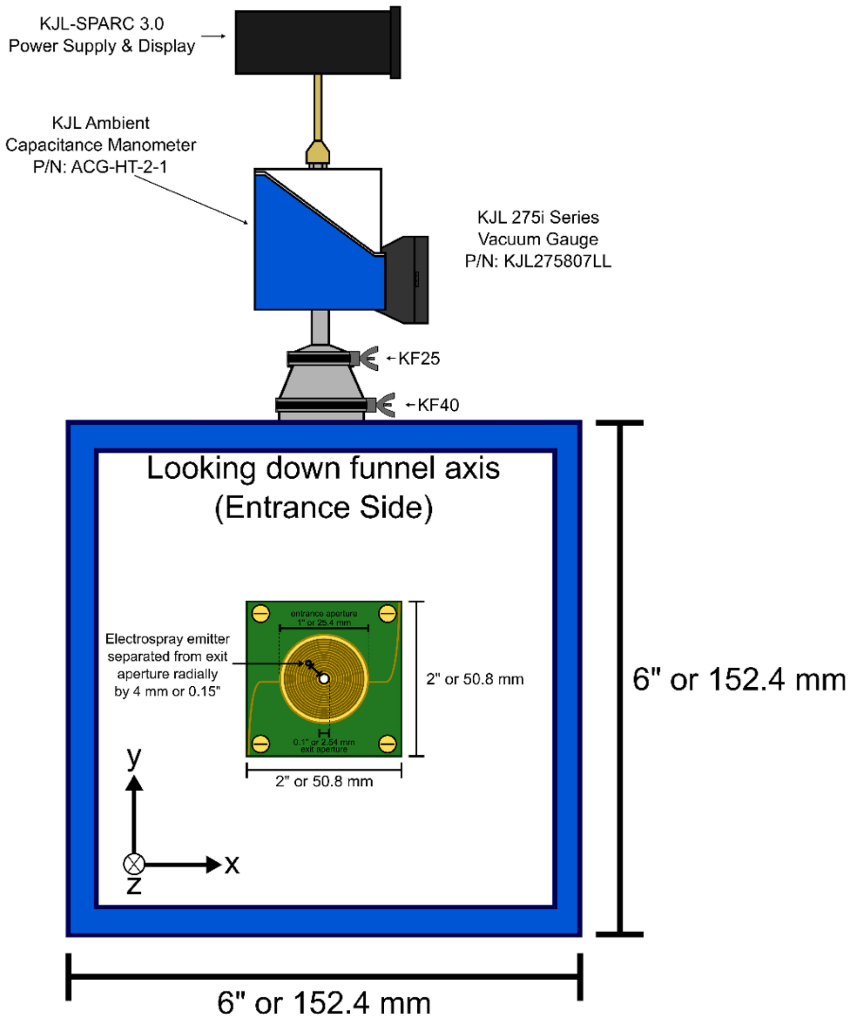


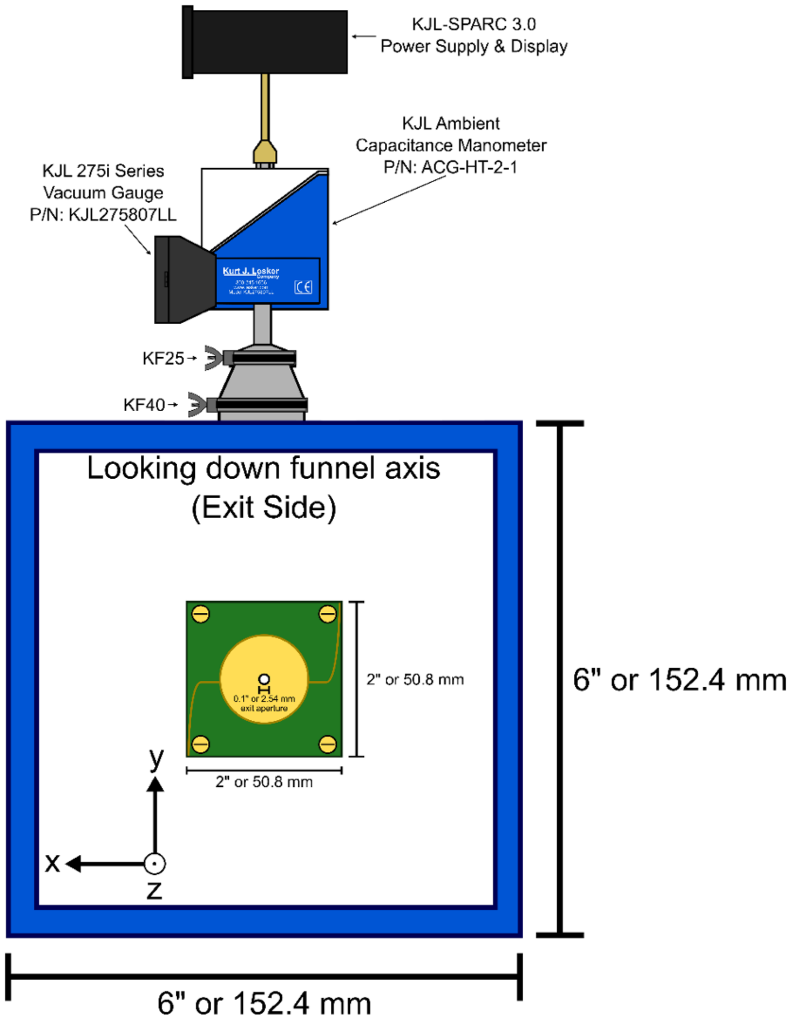
Current Activities:
More Granular Drawings For Our Mechanical Design Team

2025-05-05
FRIB-EDM[3]
Electrospray Chamber
With Spatial Dimensions
Draft 2



Current Activities: More Granular Drawings For Our Mechanical Design Team

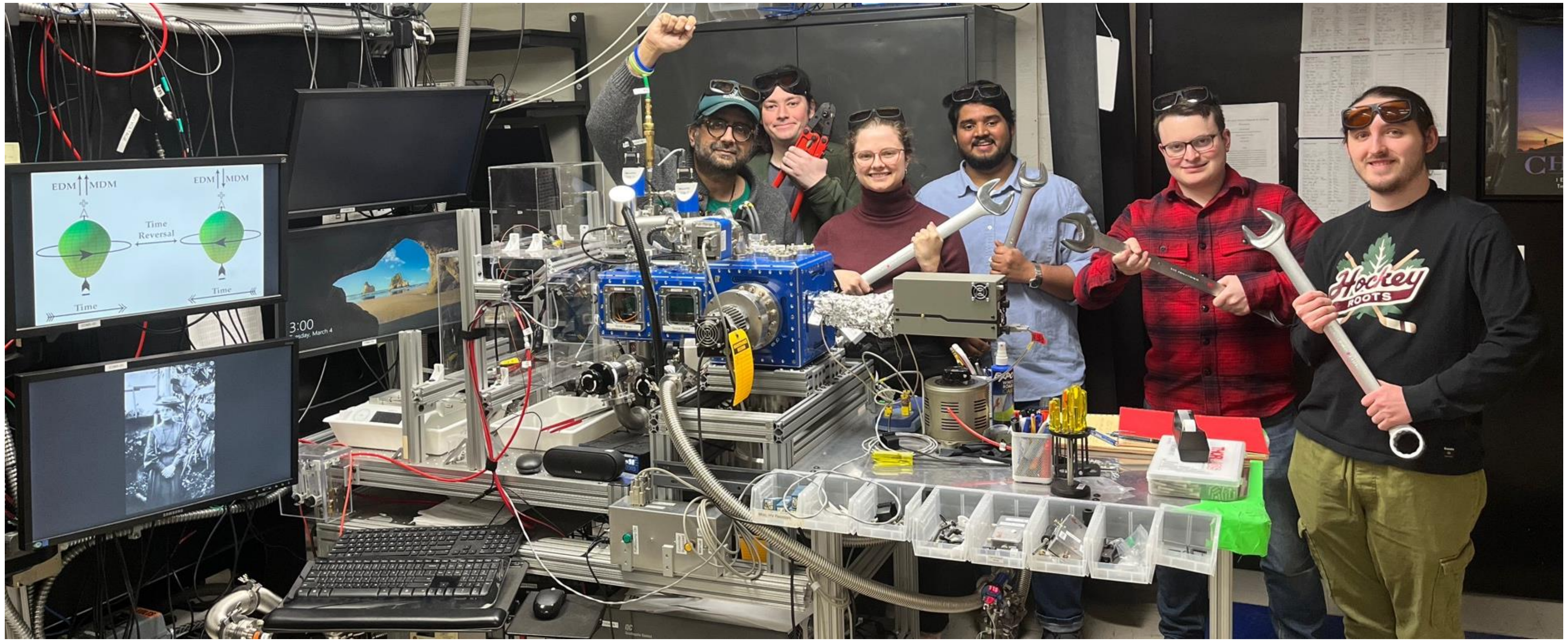




Summary

- 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
- We believe we can improve upon on test bench electrospray by utilizing lower precursor flow rates and taking the electrospray pressure to rough vacuum

Thank You For Your Attention!



Special thanks to:
Jochen Ballof, Sebastian Miki-Silva, Oscar Naviliat-Cuncic, Tom-Erik Haugen, Mia Au, Peyton Lalain, Sebastian Rothe, Ben Arend, Peader Richards, Brandon Ewert, Patrick Glennon, Eric Hessels, Greg Koyanagi, David Leimbach, Ryan Ringle, Stefan Schwarz, Amar Vutha

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

What Is The Expected Magnitude Of The Schiff Moment For Pear-shaped Systems Predicted By Current Calculations? (1)

The Schiff Moment is calculated in two ways depending on whether you ask an atomic or nuclear theorist. In general, it depends on the size scale of the **~~P~~**, **~~T~~** violating physics.

For Atomic theorists, calculating a nuclear Schiff moment involves calculating $A_{nuclear}$:

$$d_{atom} = A_{electron} d_e + A_{nucleus} S + A_{NSI} C_{NS\,I} + A_{NSD} C_{NSD}$$

For paramagnetic atoms with nuclear spin = 0

$$d_{atom} = \underbrace{A_{electron}}_{\textbf{LARGE}} d_e + \underbrace{A_{nucleus}}_{\sim\textbf{0}} S + \underbrace{A_{NSI}}_{\textbf{LARGE}} C_{NS\,I} + \underbrace{A_{NSD}}_{\sim\textbf{0}} C_{NSD}$$

For diamagnetic atoms with nuclear spin ≠ 0

$$d_{atom} = \underbrace{A_{electron}}_{\textbf{small}} d_e + \underbrace{A_{nucleus}}_{\textbf{LARGE}} S + \underbrace{A_{NSI}}_{\textbf{small}} C_{NS\,I} + \underbrace{A_{NSD}}_{\textbf{LARGE}} C_{NSD}$$

What Is The Expected Magnitude Of The Schiff Moment For Pear-shaped Systems Predicted By Current Calculations? (2)

The Schiff Moment is calculated in two ways depending on whether you ask an atomic or nuclear theorist. In general, it depends on the size scale of the ~~P~~, ~~T~~ violating physics.

For Nuclear theorists, calculating a nuclear Schiff moment involves calculating all A_{things}:

$$S = A_{SRN}\tilde{d}_n^{SR} + A_{SRP}\tilde{d}_p^{SR} + A_{isoscalar}g_0 + A_{isovector}g_1 + A_{isotensor}g_2$$

What Aqueous Precursor Will You Use?

Initially, we though we'd use HF, but HF is a nasty chemical to work with. Using it in our electrospray was the first thing we thought would introduce fluorine to form molecular bonds with radium. But… it would be corrosive AND radioactive and very hazardous to work with

Alyssa Gaiser (Radiochemist @ FRIB) told us about how you can add fluorine to ammounium (or ammonium derivatives) readily to f-shell elements and how she expects that you'd be able to do so for alkaline earth elements too:

"With the small quantities I believe you anticipate working with, precipitation will not be an issue, unless for some reason you have another alkali or alkaline earth carrier salt present in precipitatable quantities (i.e. mgs of Ba salt), you should experience little to no issue."
Alyssa Gaiser, personal communication, March 11, 2025

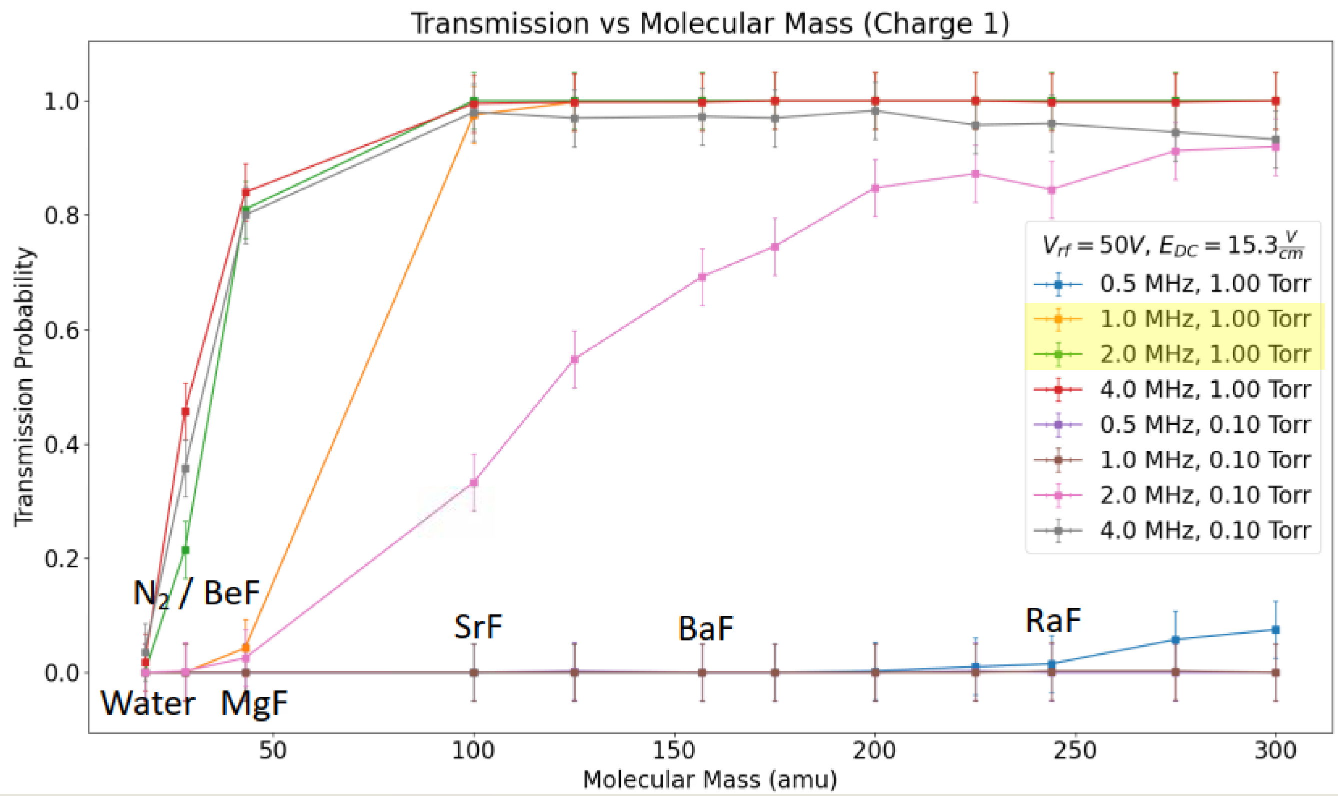
Fluorine + Ammonium for f-shell: Russo and Haendler Journal of Inorganic and Nuclear Chemistry 36:763 (1974)
Link: https://www.sciencedirect.com/science/article/pii/0022190274808080

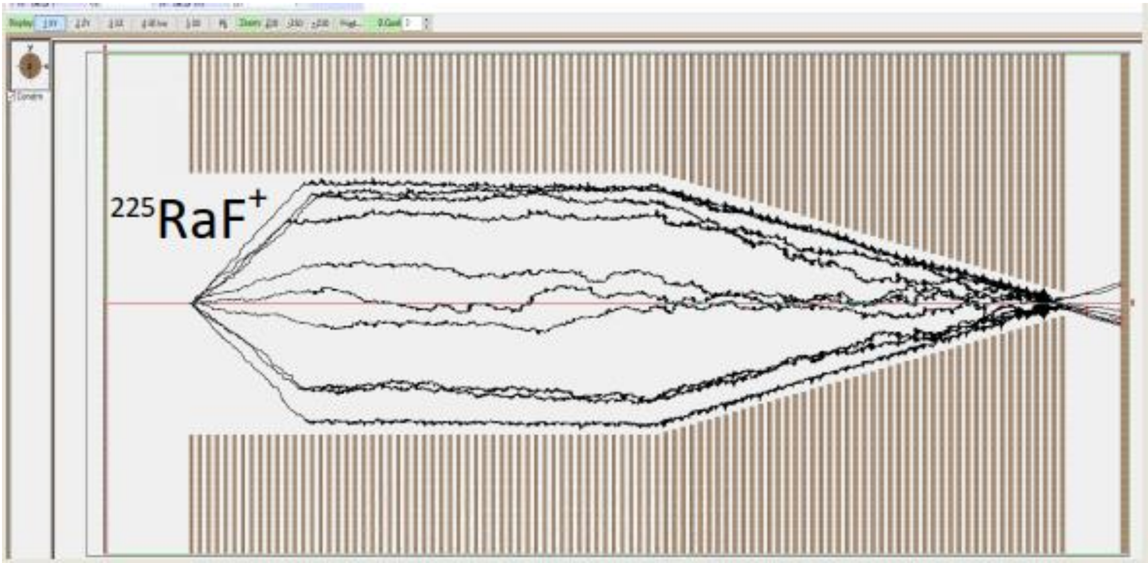
Precipitation of RaF_2 from $Ra(NO_3)_2$ in NH_4F: Butkalyuk et al. Radiochemistry 63:21 (2021)
Link: https://link.springer.com/article/10.1134/S1066362221010045

Simulating Ion Funnel Performance For "Low" m/z

Electrospray Ionization is typically used on molecular ions with masses ~ 10^3 Daltons (Da)
Nick Nusgart (nusgart@frib.msu.edu) performed the simulations and has more details

$$1\,Da = 1\,m_u = \frac{1}{12} m(^{12}C)$$





Offset Between The Funnels And Electrospray?

We intentionally offset funnels from each other, and the first funnel from the electrospray to suppress the line-of-sight gas load from the electrospray source