The FRIB-EDM^3 Instrument: A Tool For Creating Molecules Using Octupole Deformed Nuclei For Tests Of Fundamental Symmetries

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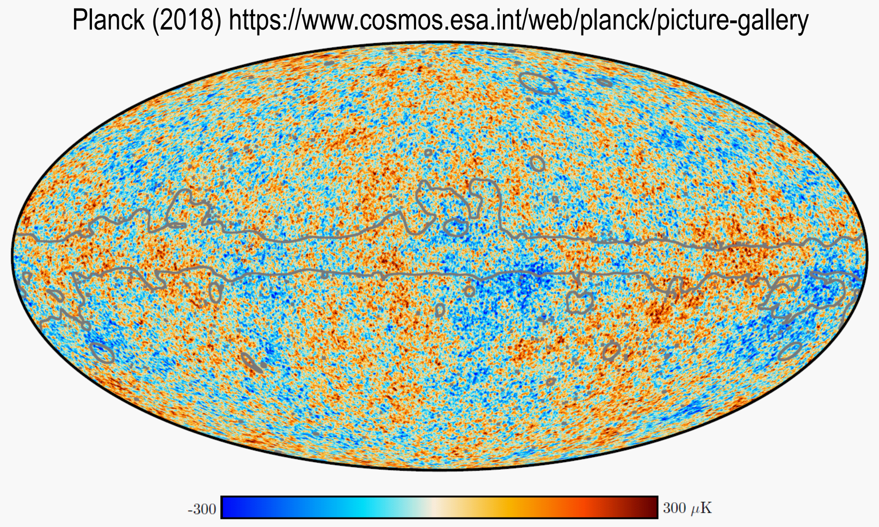
FACILITY FOR RARE ISOTOPE BEAMS

Introduction

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



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

$\eta_{exp} \approx 10^{-9}$ PDG2024 $\eta_{CKM} \approx 10^{-26}$ Huet & Sather PRD 51:379 (1995)

Sakharov Conditions
1. A baryon number violating interaction exists
2. Must be a departure from thermal equilibrium
3. Both C- & CP-symmetry violation required

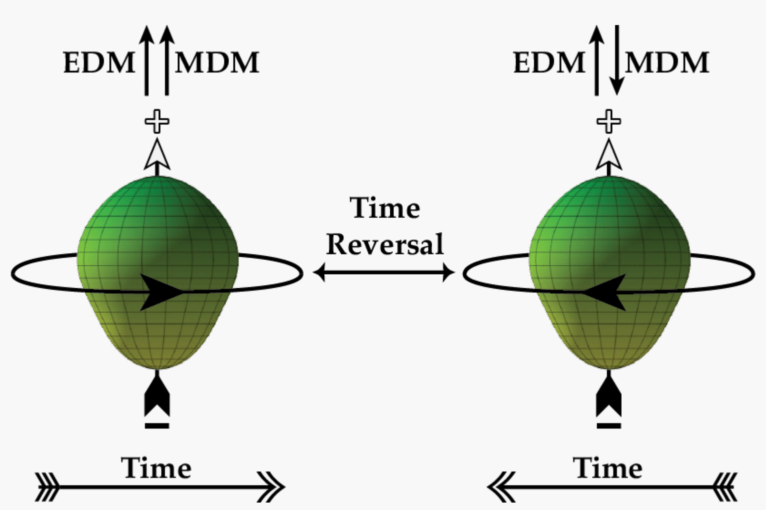
A. D. Sakharov JETP Letters, 5:24 (1967)

Electric Dipole Moments (EDMs) measure a separation of charge:

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

Dipole moments can couple to electromagnetic fields:

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



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 (-)

CPT Theorem:
T-Violation = CP-Violation

The CP-violating observable in nuclear systems that corresponds to an EDM is called a **nuclear Schiff moment:**

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

Nuclear Schiff moments in the lab frame:

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

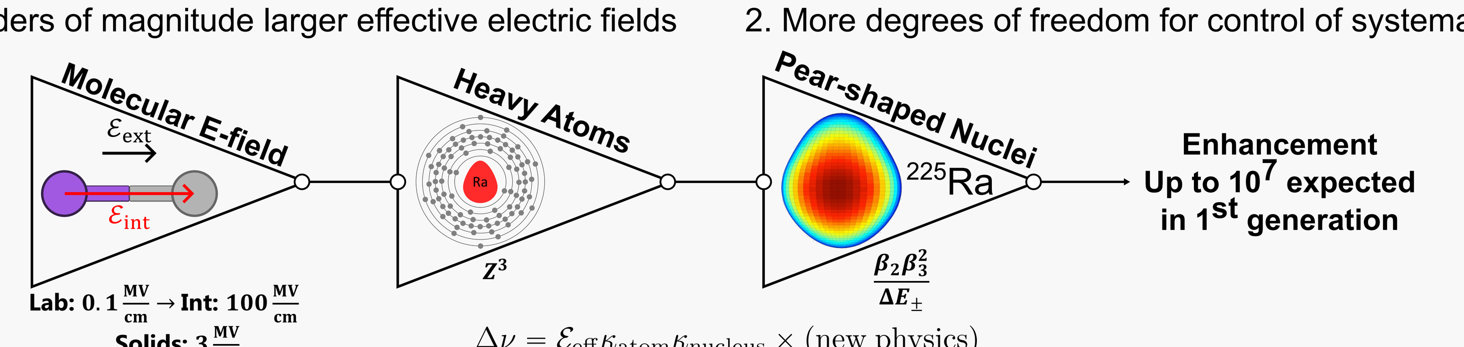
In the lab frame, octupole deformed nuclei amplify our ability to see **symmetry violating physics** because of their:

1. Nearly degenerate parity doublets Haxton & Henley PRL 51:1937 (1983)

2. Large intrinsic Schiff moment Auerbach, Flambaum, & Spevak PRL 76:4316 (1996)

Molecules offer two classes of enhancement:

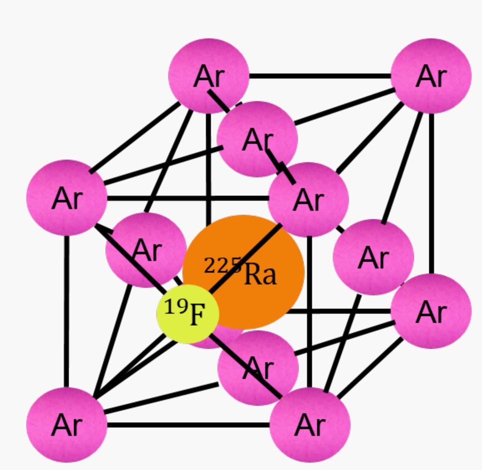
1. Orders of magnitude larger effective electric fields
2. More degrees of freedom for control of systematics



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

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

A polar molecule with a heavily deformed nucleus like ^{225}Ra allows us to take advantage of these enhancements. To perform an NSM search, we need to trap our molecules. We are interested in embedding them in noble gas solids to trap them because:
- Large densities of guest molecules ($\leq 10^{13}$/mm^3) can be hosted without them interfering with each other
- Noble gas solids lock the orientation of certain guest molecules Ex: BaF in neon: Li et al. NJP 25:082001 (2023)
- Stable, chemically inert environment



Acknowledgements

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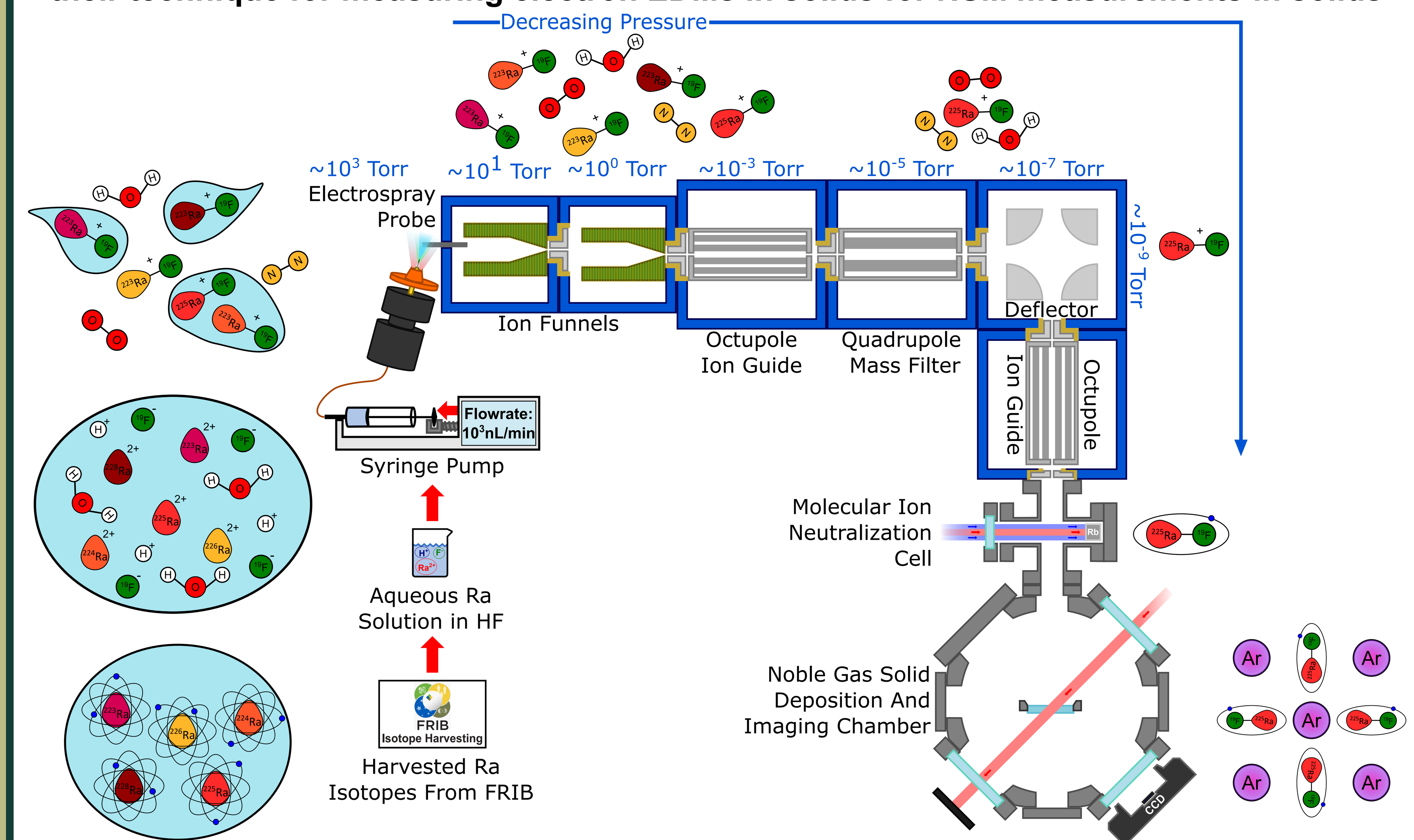
FRIB-EDM^3

We are collaborating with York University (Canada) and the University of Toronto to adapt their technique for measuring electron EDMs in solids for NSM measurements in solids



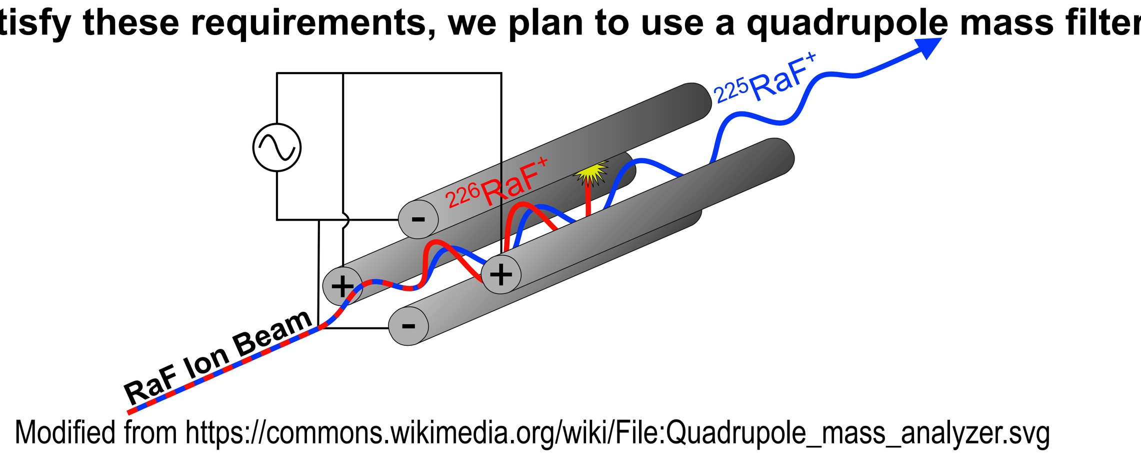
We want a technique that:
- Can form an isotopically pure beam of neutral molecules
- Is UHV compatible
- Has a high formation efficiency

See also: Vutha et al. PRA 98:032513 (2018), J. T. Singh Hyp. Int. 240:29 (2019), Ramachandran & Vutha, PRA 108 :012819 (2023), and Li et. al NJP 25:082001 (2023)

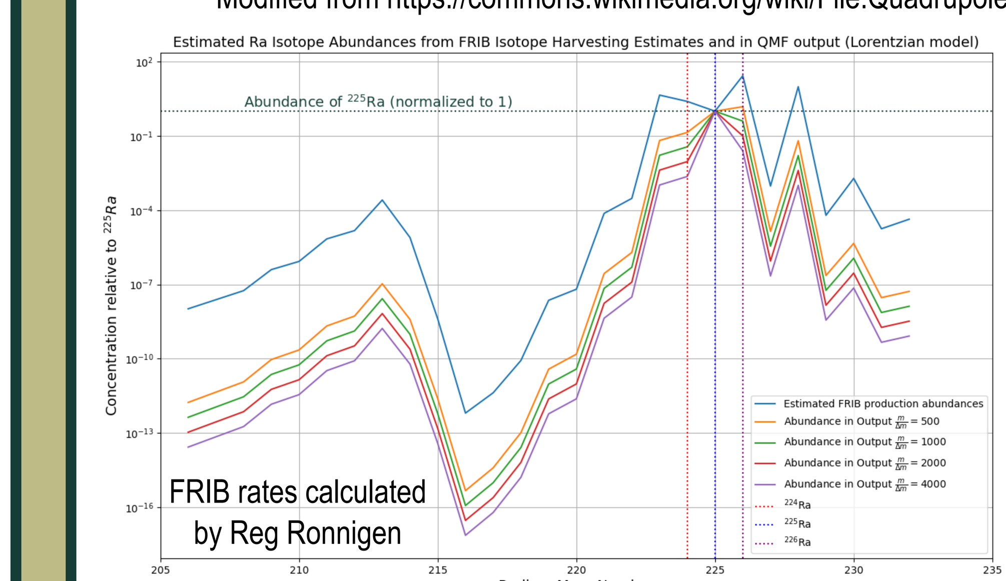
Isotopic Filtering with a Quadrupole Mass Filter

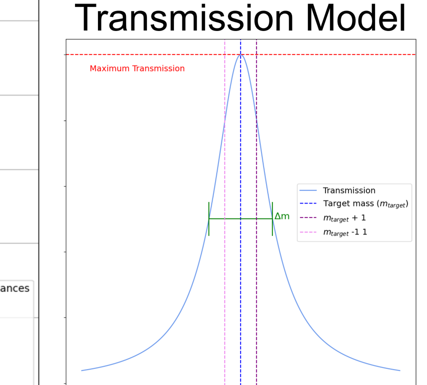
We plan to source Ra from FRIB's Isotope Harvesting in the form of an aqueous solution of Ra in HF. To ensure isotopic purity, while preserving our statistics, we need to be able to:
- Separate RaF molecules containing ^{225}Ra from those containing other Ra isotopes
- Transmit our desired molecules with high efficiency

To satisfy these requirements, we plan to use a quadrupole mass filter.



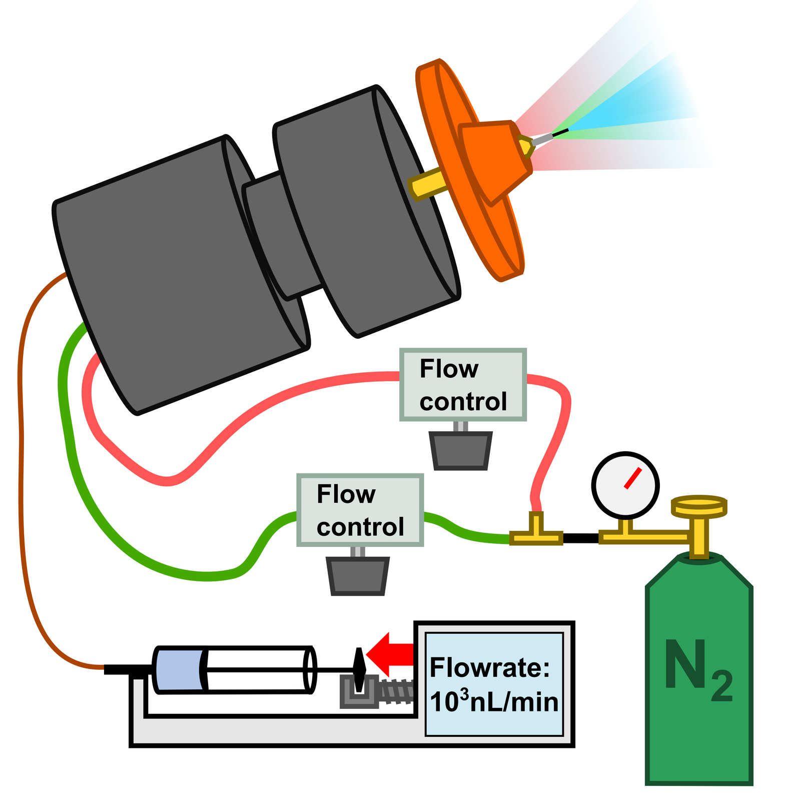
Modified from https://commons.wikimedia.org/wiki/File:Quadrupole_mass_analyzer.svg





Abundance in QMF output = (Input Abundance)(QMF Transmission)

Molecule Formation with Electrospray Ionization

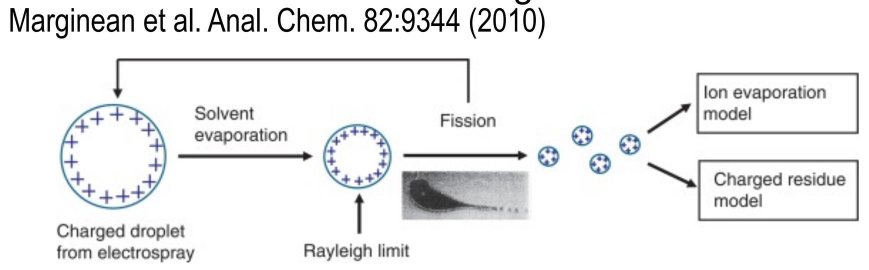


Common Approach: Cryogenic Buffer Gas Beam
- Creates molecular ions from solid precursor
- Molecule extraction efficiencies vary (20% for ThO)
- Estimated final yield of desired molecules ~1% (3% for ThO, 0.03% for SrF)

Hutzler et al. PCCP 42:18976 (2011) Barry et al PCCP 13:18936 (2011)

Our Approach: Electrospray Ionization
- Creates molecular ions from aqueous precursor
- Ideal for small radioactive samples (~10^5 nL)
- Ion utilization efficiencies as high as 50%

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

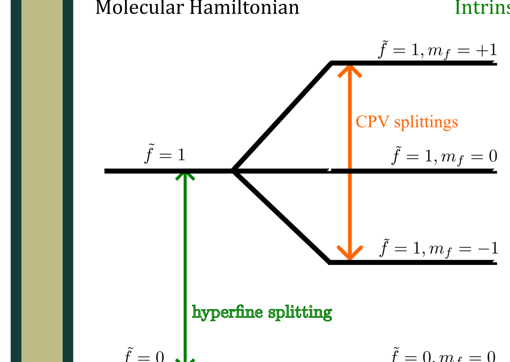


Tang et al. Encyclopedia of Spectroscopy and Spectrometry 476 (2017)

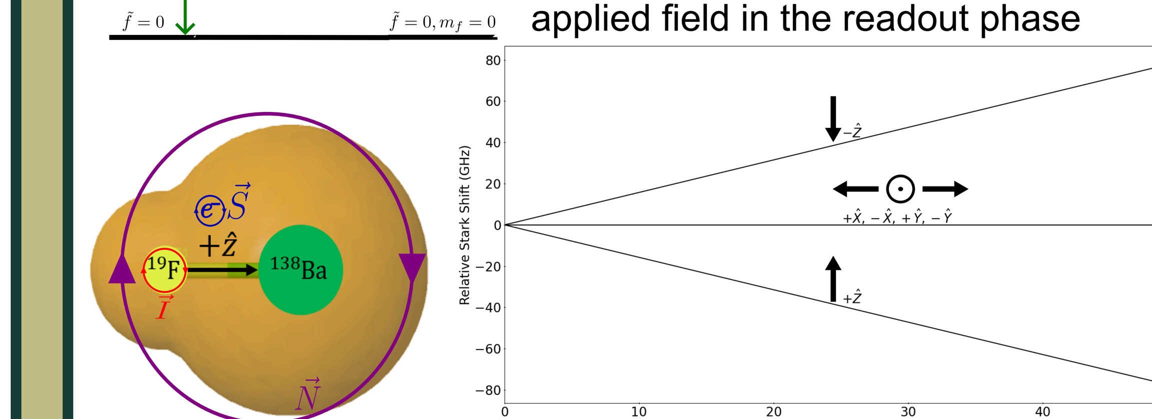
Rotational Hyperfine Structure Calculations

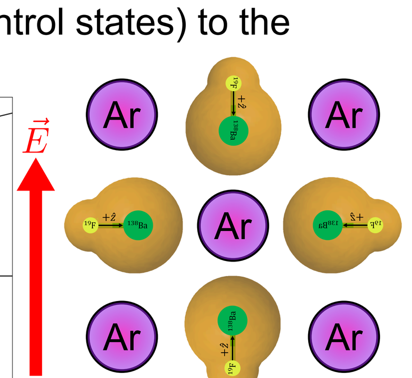
$$\text{H}(N,J,F,M_F) = \text{H}_{rot}(N,J) + \text{H}_{hfs}(\vec{I_F},\vec{S}) + \text{H}_{stk}(N,J,F,M_F,\vec{E_{lab}}) + \text{H}_{Dev}(N,J,F,M_F) + \text{H}_{CPV}$$

Molecular Hamiltonian; Intrinsic to Molecule; External Field; Model of matrix effects (Devonshire potential); CP-violating physics



Proposing a measurement scheme requires knowledge of the rohyperfine structure in-medium for our molecule:
- Started with a similar and simpler molecule, ^{138}Ba^{19}F
- Can use an electric field to spectroscopically distinguish states aligned with or against (potential science states), and perpendicular (potential systematic control states) to the applied field in the readout phase

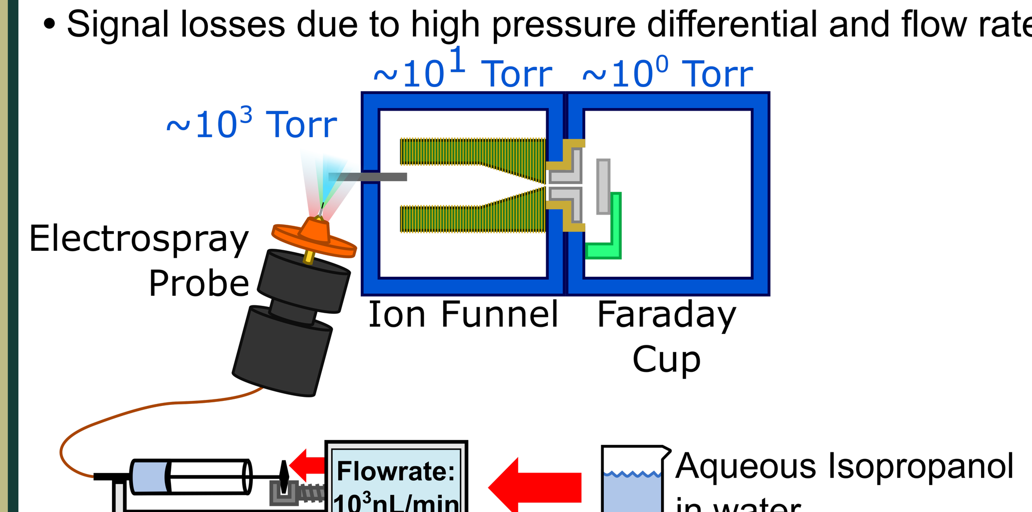


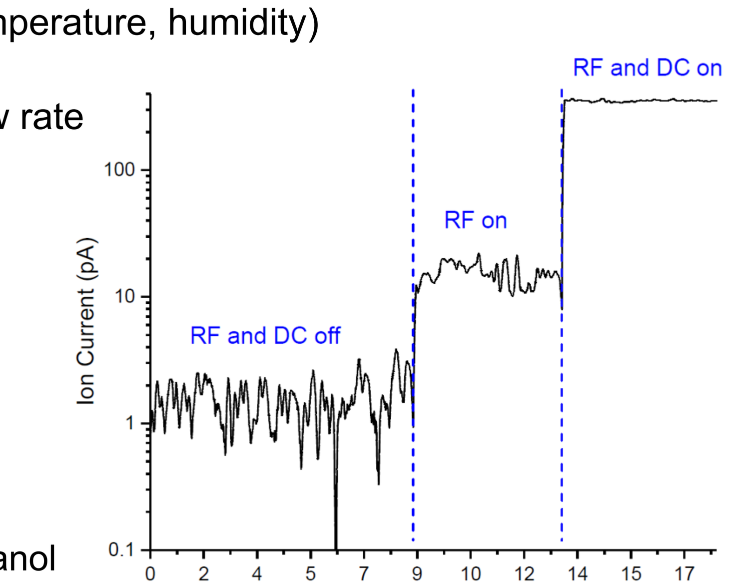


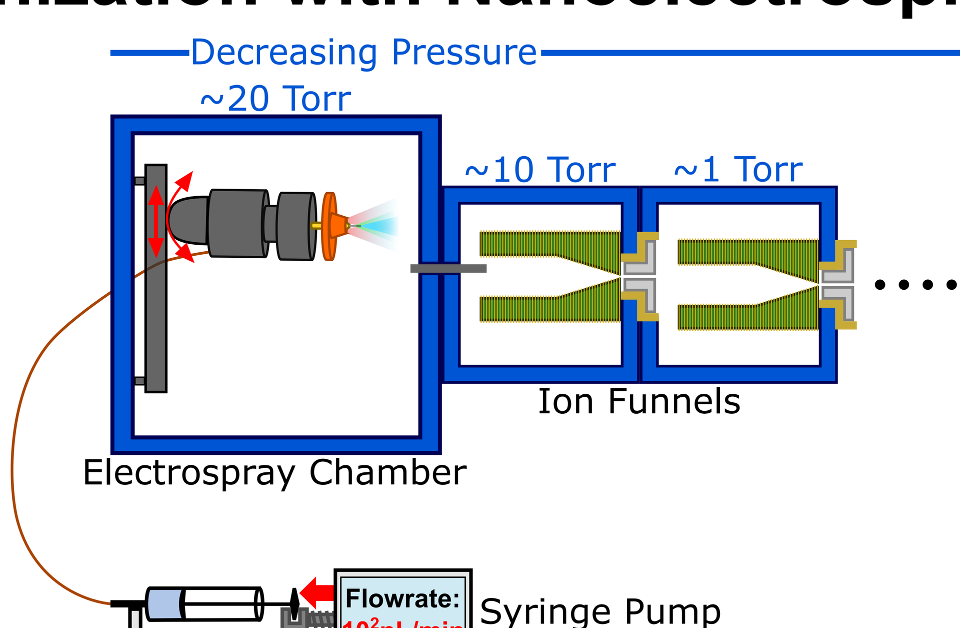
Orientation Locking in ^{138}Ba^{19}F:Ar

Upgrading to Subambient Pressure Ionization with Nanoelectrospray

Our atmospheric electrospray ion source allows us to form molecular ions, however:
- Stability sensitive to ambient conditions (pressure, temperature, humidity)
- Stability sensitive to probe positioning and alignment
- Signal losses due to high pressure differential and flow rate







We are designing new electrospray ion source that will allow for:
- Improved ion transmission via low pressure differential
- Increased spray stability with lower flow rates
- Precise positioning and alignment of spray probe