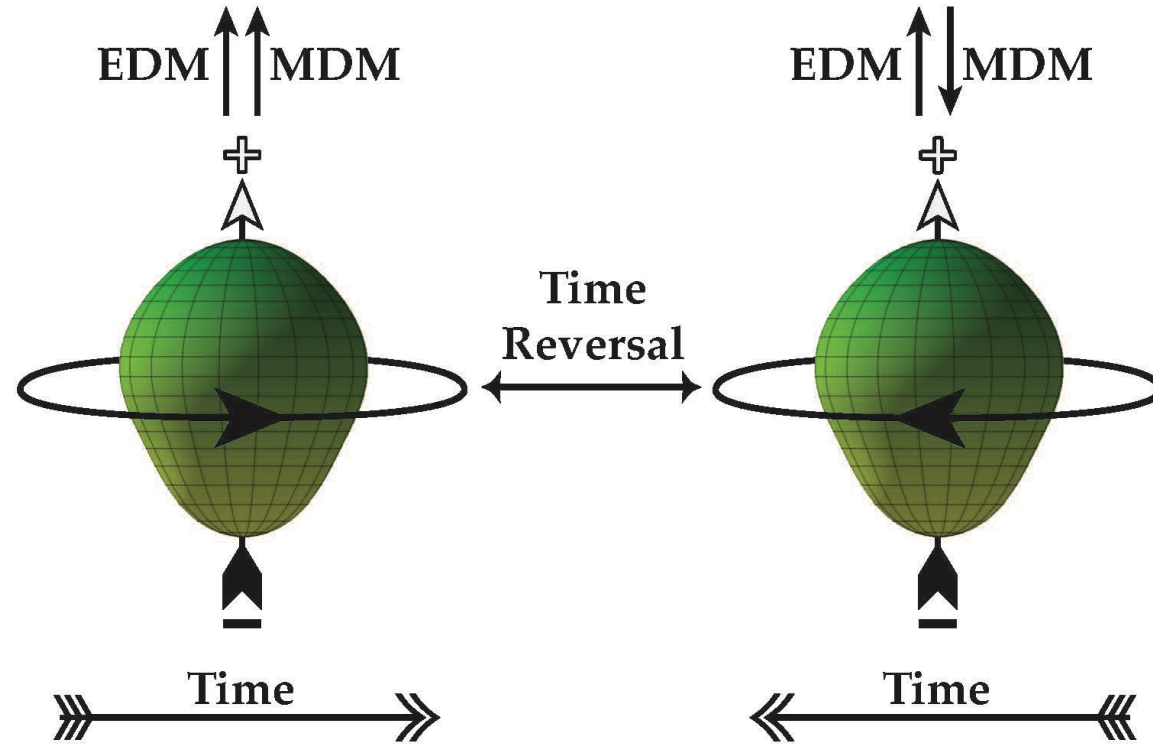


Towards an Ultrasensitive Search for Time-Reversal Violation Using Heavy Rare Pear-Shaped Nuclei



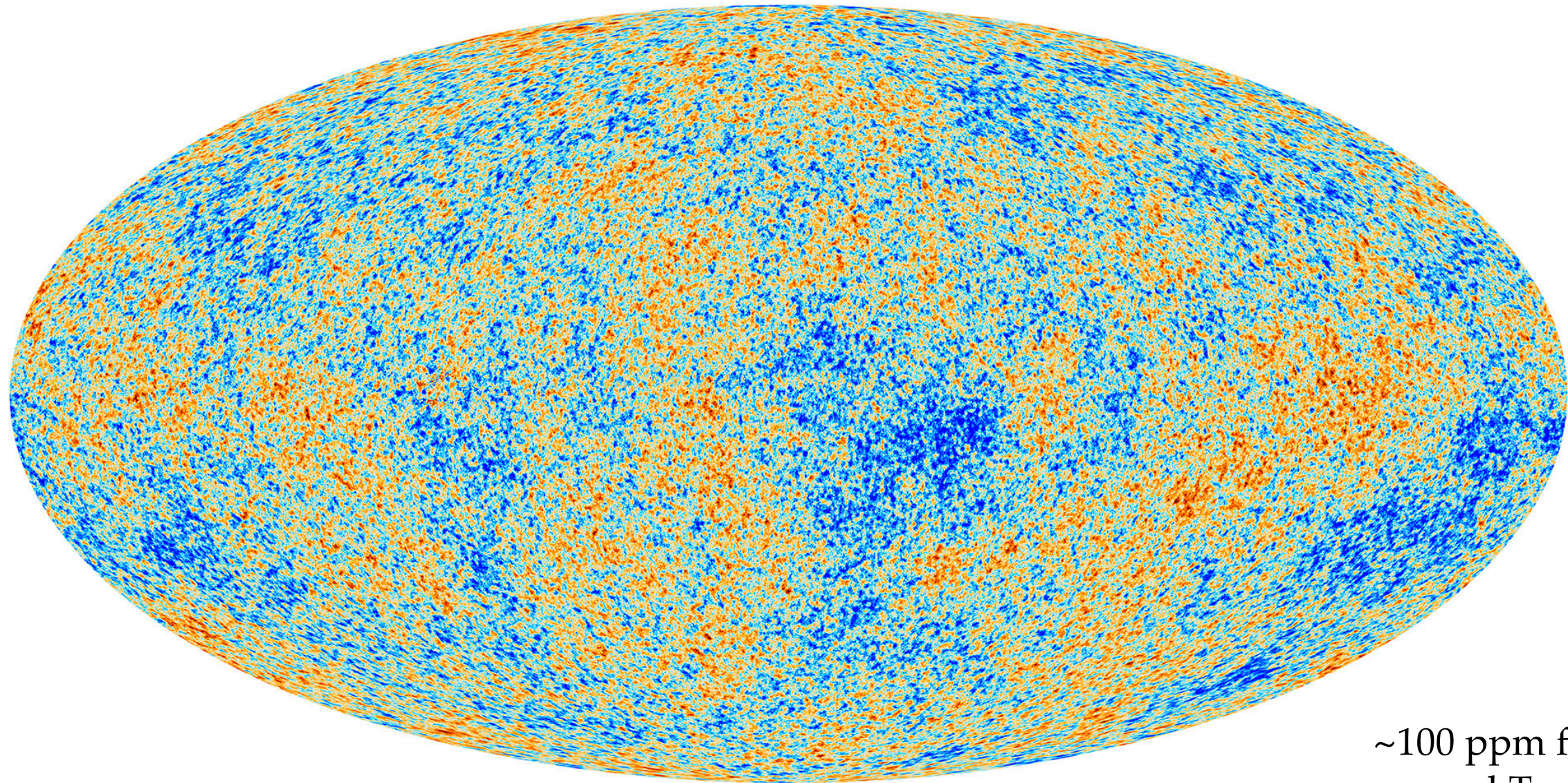
Lise
Meitner



Jaideep Taggart Singh (he/him/his)
Facility for Rare Isotope Beams/Michigan State University
PSI Center for Neutron and Muon Sciences
Laboratory for Particle Physics - LTP / PSI Thursday Colloquium
March 12, 2026, 16:00, WBGB / 019



Cosmic Microwave Background Anisotropy: “Baby Picture” of the Universe



~100 ppm fluctuations
around $T = 2.7255 \text{ K}$

Planck 2018

https://www.esa.int/Science_Exploration/Space_Science/Planck/Planck_and_the_cosmic_microwave_background

There is no visible antimatter in the Universe.

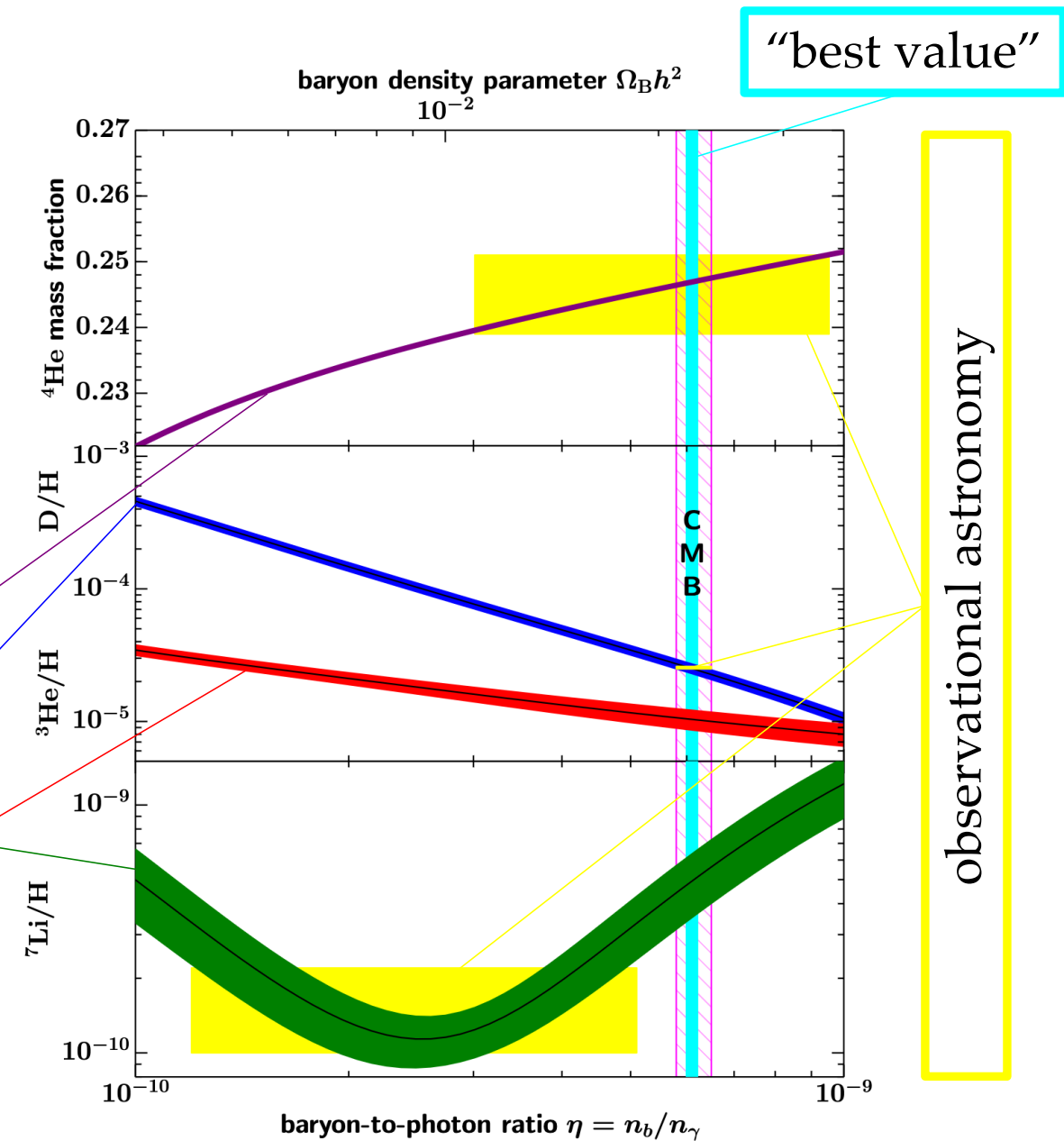
$$\eta \text{ " = " } \frac{(\text{matter}) - (\text{antimatter})}{\text{relic photons}}$$

$$\eta = 0.0000000006115 \text{ (0.6\%)}$$

$$\approx 10^{-9}$$

nuclear physics

PDG2025



CMBR

observational astronomy

Sakharov's Conditions: Need CP -Violation

The Nobel Foundation



VIOLATION OF CP INVARIANCE, C ASYMMETRY, AND BARYON ASYMMETRY OF THE UNIVERSE

A. D. Sakharov

Submitted 23 September 1966

ZhETF Pis'ma 5, No. 1, 32-35, 1 January 1967

The theory of the expanding Universe, which presupposes a superdense initial state of matter, apparently excludes the possibility of macroscopic separation of matter from anti-matter; it must therefore be assumed that there are no antimatter bodies in nature, i.e., the Universe is asymmetrical with respect to the number of particles and antiparticles (C asymmetry). In particular, the absence of antibaryons and the proposed absence of baryonic neutrinos implies a non-zero baryon charge (baryonic asymmetry). We wish to point out a possible explanation of C asymmetry in the hot model of the expanding Universe (see [1]) by making use of effects of CP invariance violation (see [2]). To explain baryon asymmetry, we propose in addition an approximate character for the baryon conservation law.

1. A baryon number violating interaction exists.
2. Departure from thermal equilibrium.
3. *Both C - & CP -symmetry must be violated.*

Standard Model CP -Violation: Not Enough

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

$$\eta_{\text{exp}} \approx 10^{-9} \quad \text{PDG2025}$$

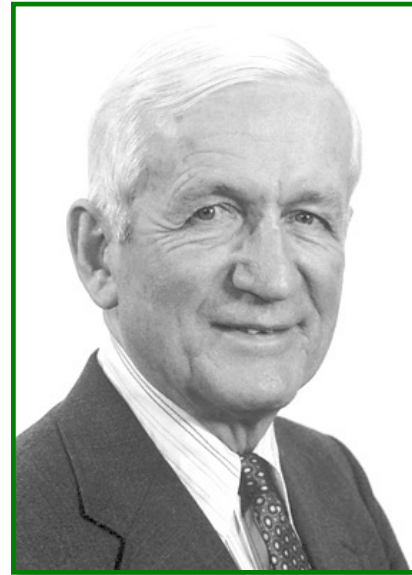
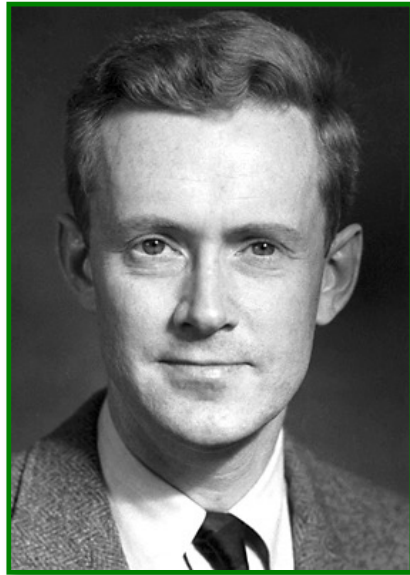
$$\eta_{\text{CKM}} \approx 10^{-26} \quad \text{Huet \& Sather PRD 51:379 (1995)}$$

$$V_{\text{CKM}} = \begin{bmatrix} c_{12}c_{13} & s_{12}c_{13} & s_{13} \exp(-i\delta) \\ -s_{12}c_{23} - c_{12}s_{23}s_{13} \exp(+i\delta) & +c_{12}c_{23} - s_{12}s_{23}s_{13} \exp(+i\delta) & s_{23}c_{13} \\ +s_{12}s_{23} - c_{12}c_{23}s_{13} \exp(+i\delta) & -c_{12}s_{23} - s_{12}c_{23}s_{13} \exp(+i\delta) & c_{23}c_{13} \end{bmatrix}$$

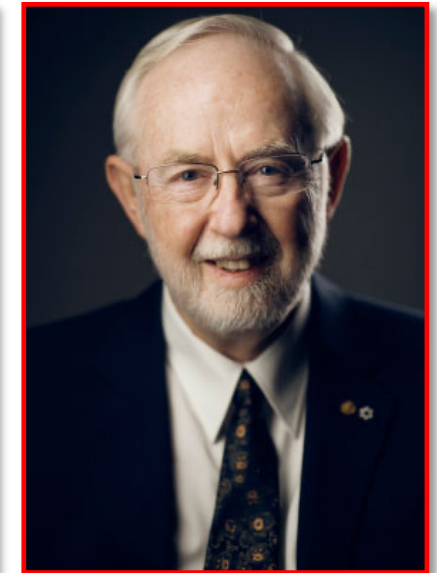
$$c_{12} = \cos(\theta_{12})$$

$\delta = CP$ -violating “phase”

Where do we look for more CP -violation?



The Nobel Foundation



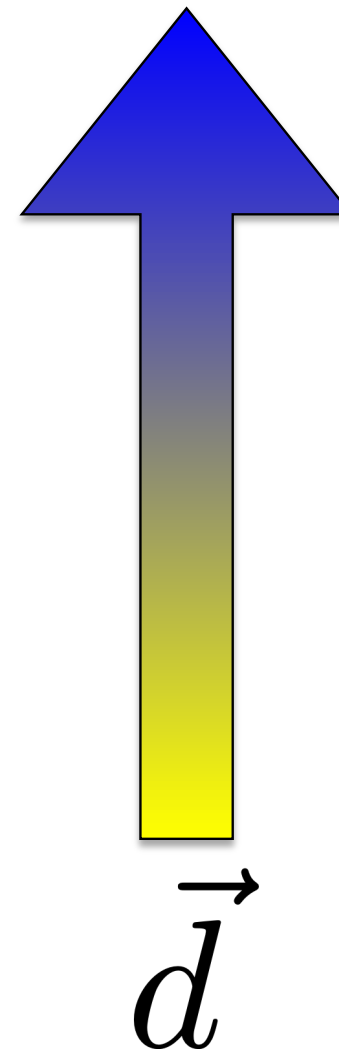
The Nobel Foundation

- Decays of B -mesons [Belle II]
- precision Higgs-ology [ATLAS/CMS]
- Rare decays of b -hadrons [LHCb]
- Angular decay correlations of positronium [Witt./MSU]
- D -coefficient in nuclear beta-decay [The MORA Project]
- Nuclear magnetic quadrupole moments [Caltech, UNLV, ODU]
- Double polarized neutron transmission [NOPTREX]
- **Neutrinos have mass! (PMNS matrix) [neutrino oscillations + $0\nu 2\beta$]**
- ***electric dipole moments: If CPT is good, then T -violation can be used to search for new sources of CP -violation!***

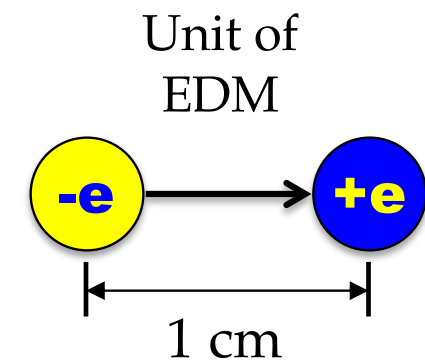
Electric Dipole Moment (EDM): Measures the Separation of Charges



"Thunder Cloud as Generator #2" (1971) by Paterson Ewen [Art Gallery of Ontario]



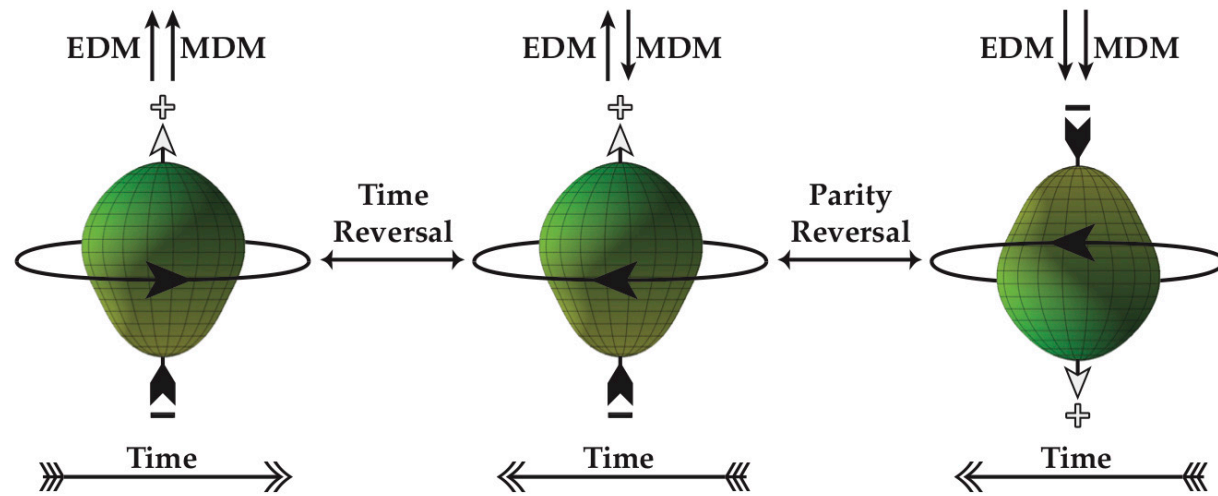
The existence of
"permanent" EDMs
violate
Time-reversal,
Parity, and
Charge-Parity
symmetries:
~~T, P, CP~~



EDMs to E-fields as MDMs to B-fields

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

adapted from B. Filippone:

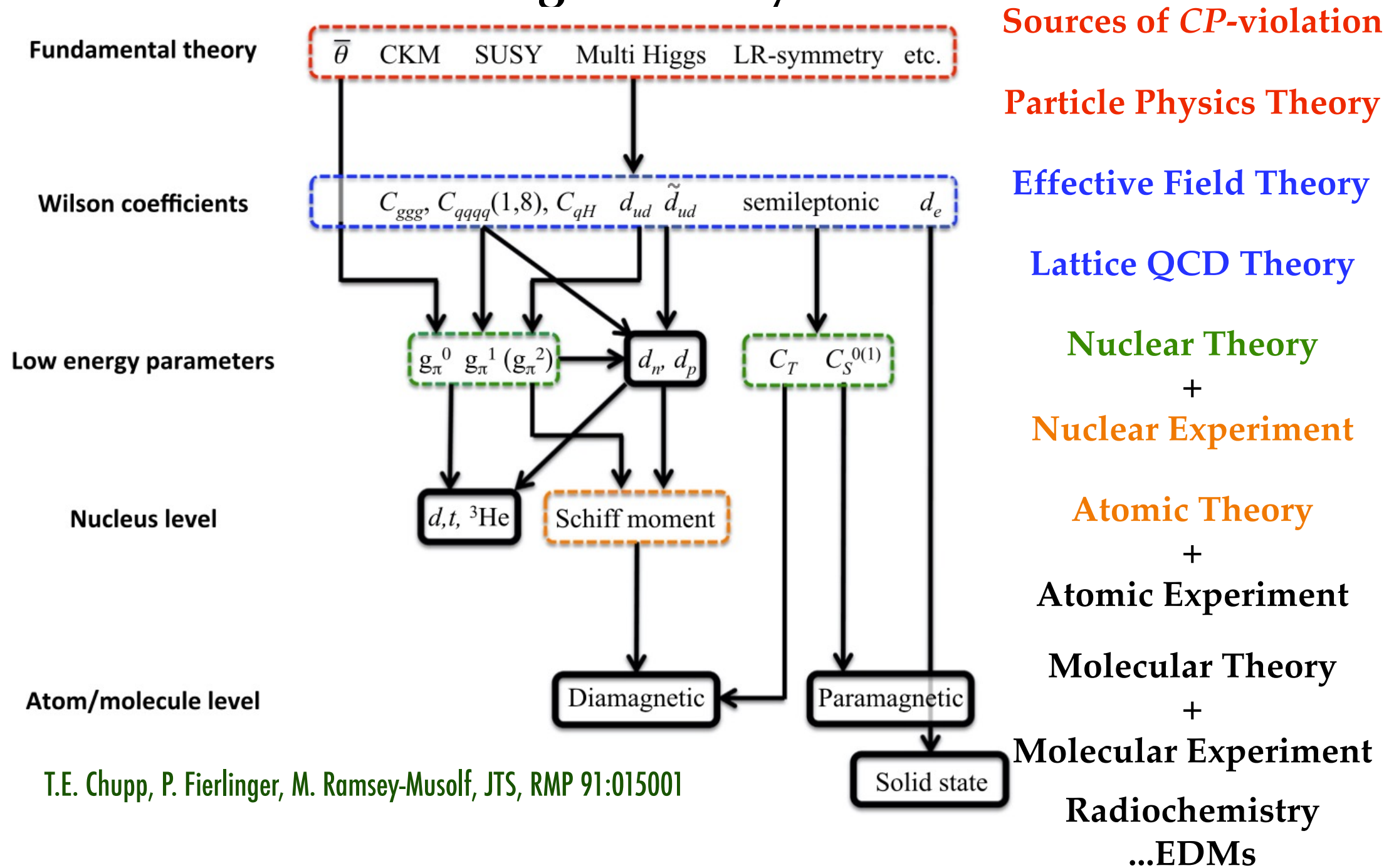


	<i>P</i> -parity	<i>T</i> -time reversal
\vec{S}	+	-
\vec{B}	+	-
\vec{E}	-	+
$\vec{S} \cdot \vec{B}$	+	+
$\vec{S} \cdot \vec{E}$	-	-

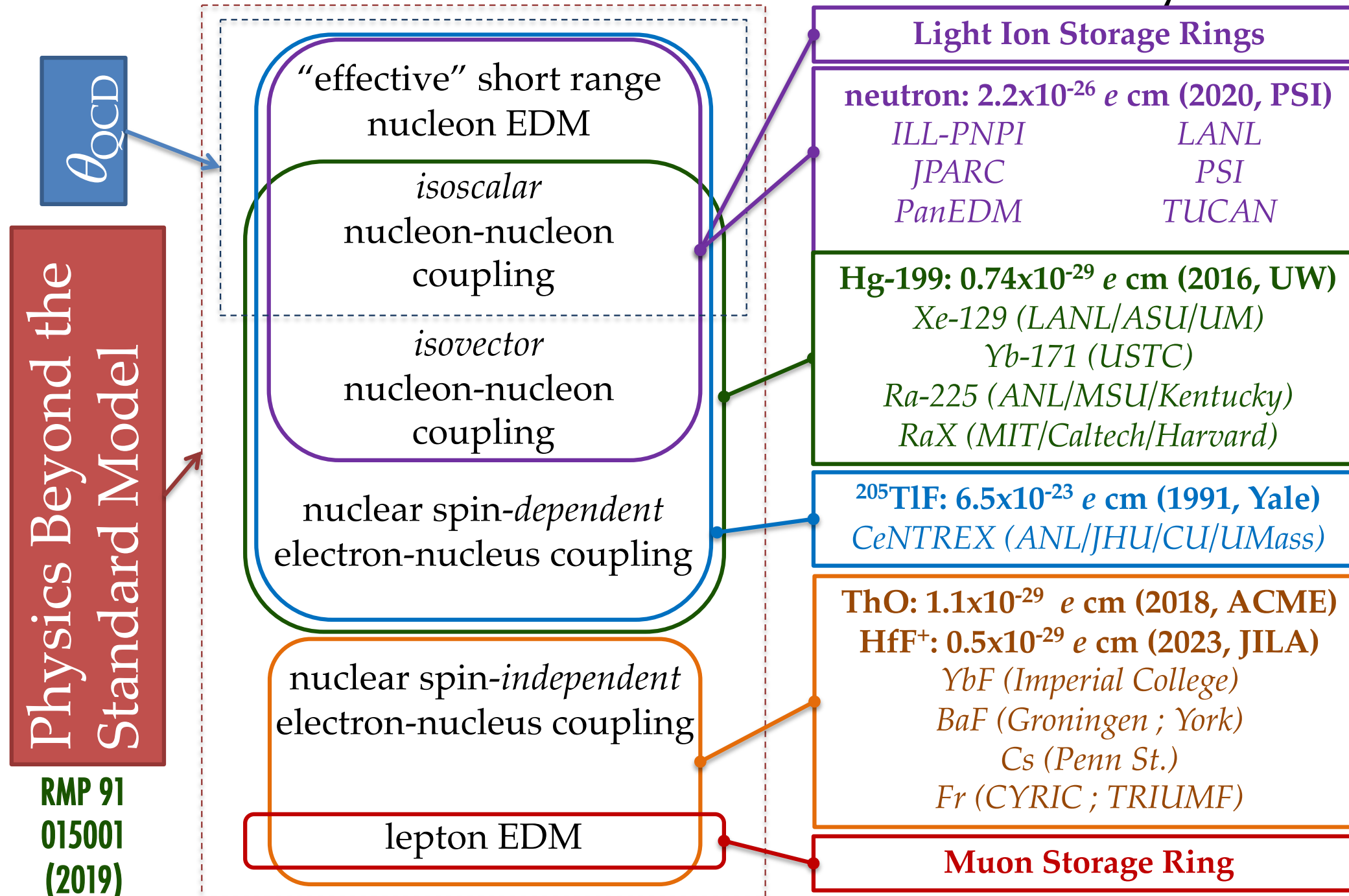
Theorist: ...trivial application of the Wigner-Eckart Theorem...

Experimentalist: ...blah blah blah Wigner-someone something...

Connecting New Physics to EDMs



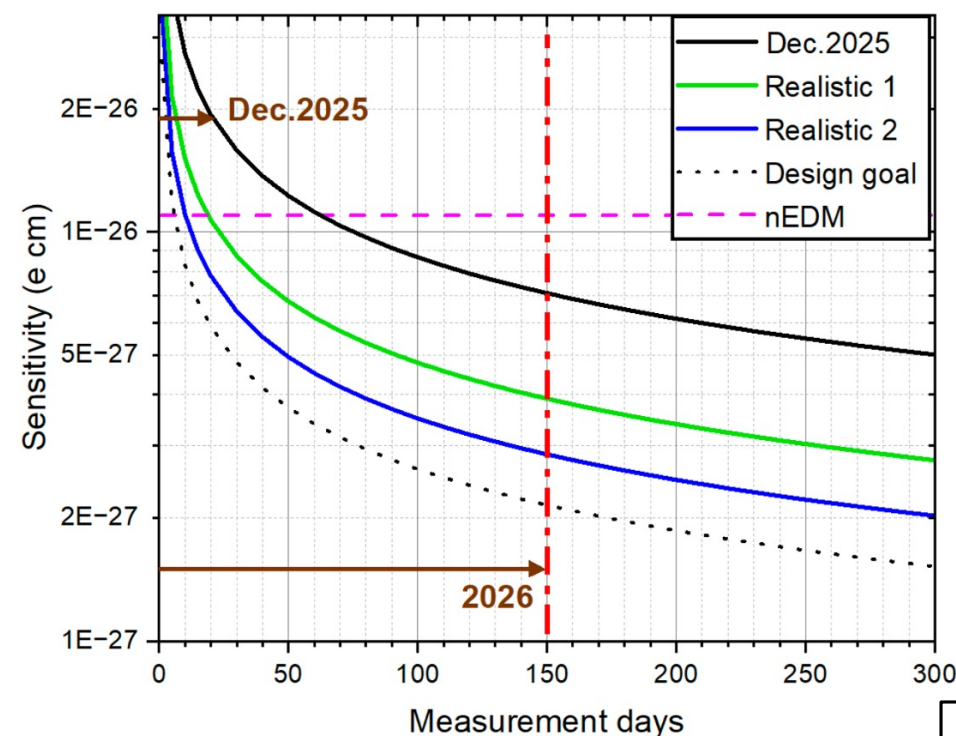
Different Sources of $\mathcal{T} \leftrightarrow$ EDM of Different Systems



Neutron EDM is Crucial - Congratulations!

Outlook for 2026

- Address HV performance
- Finish commissioning of external (Cs) magnetometers for **systematic control**
- Start blinded data-taking in June



n2EDM slide from E. Segarra (PSI)
Les Houches EDM 2026

Neutron EDM:

- is the most interpretable hadronic signal of CPV
- is the most sensitive observable to underlying quark EDMs

At least two corroborating experiments are required!

Exciting to see the progress in the Muon EDM “storage ring” approach

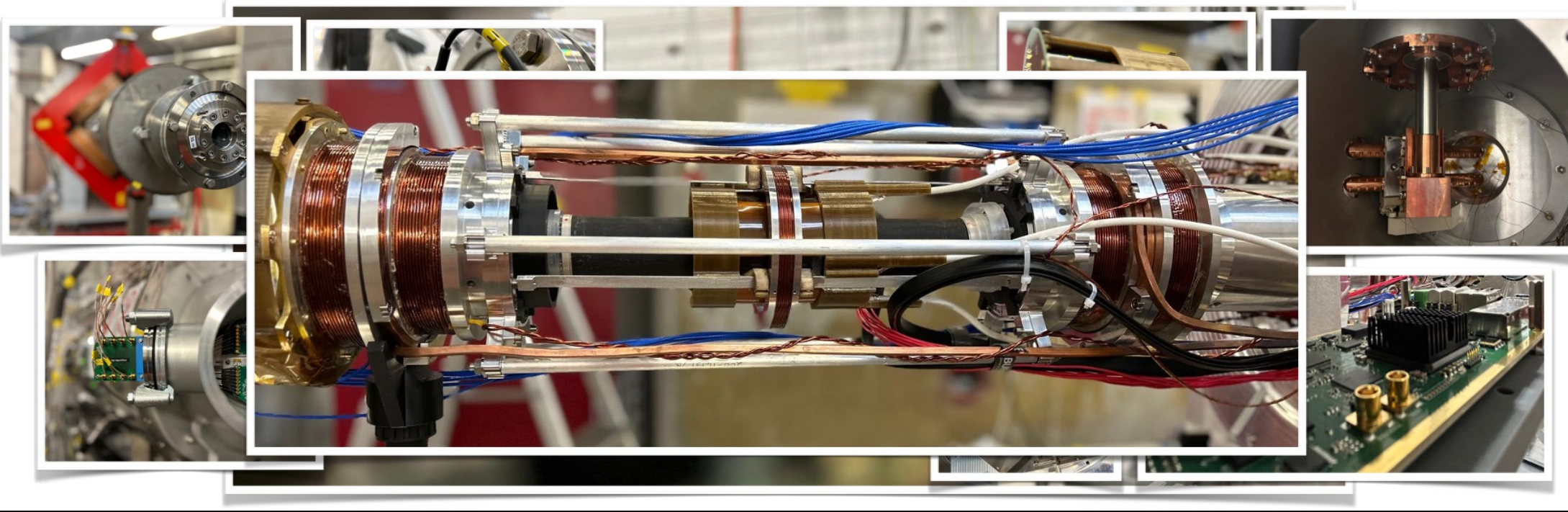


Current status

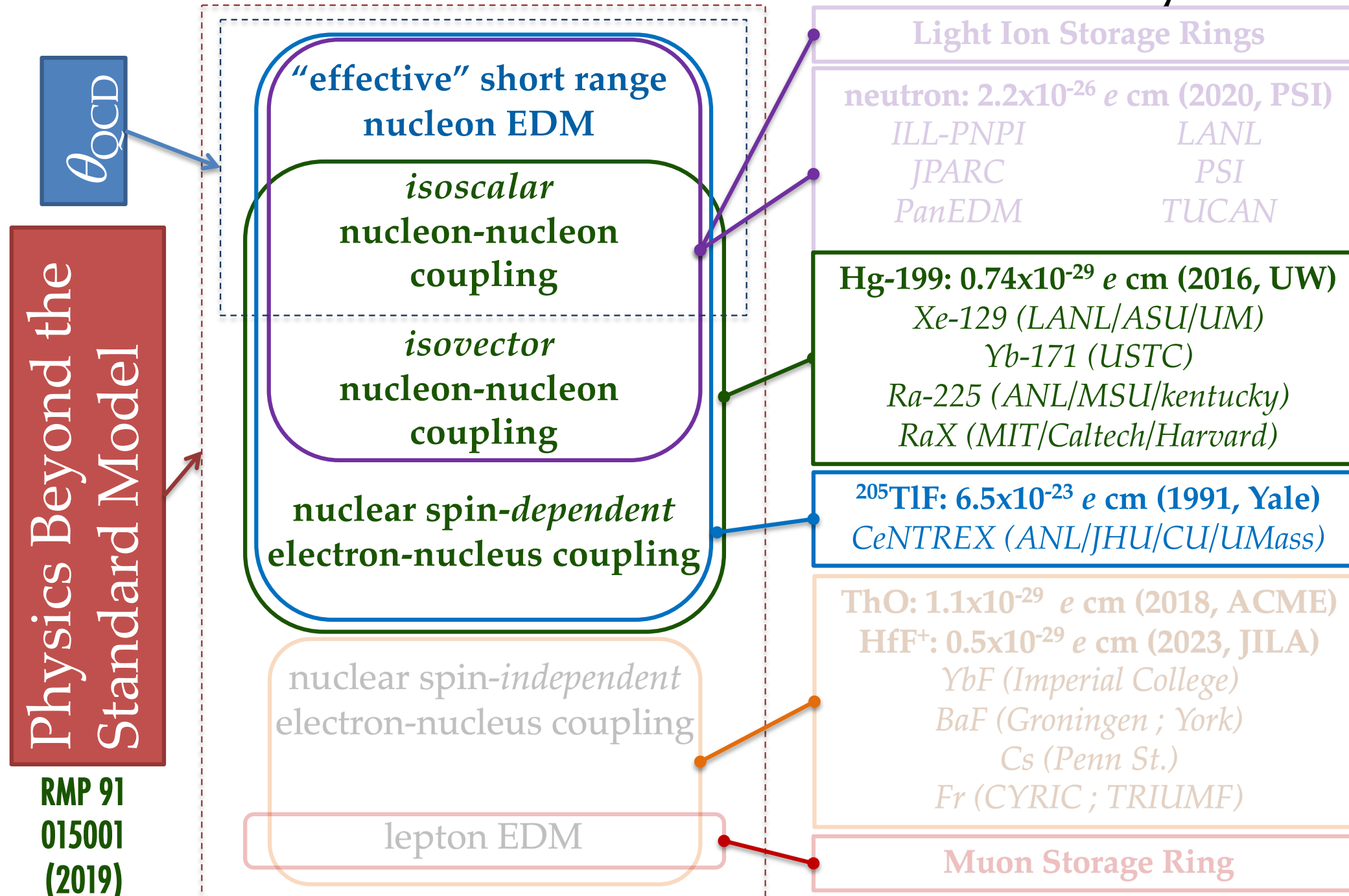


- The 2025 in a nutshell: Construction, assembly and integration of ALL items (whole/partially, prototype/final version)
 - A major milestone towards
 - the final assembly of the experimental setup, the proof of the frozen spin technique and base sensitivity (2026)
muEDM data taking (2027)

muEDM slide from
P. Schmidt-Wellenburg (PSI)
Les Houches EDM 2026



Different Sources of $\mathcal{T} \Leftrightarrow$ EDM of Different Systems



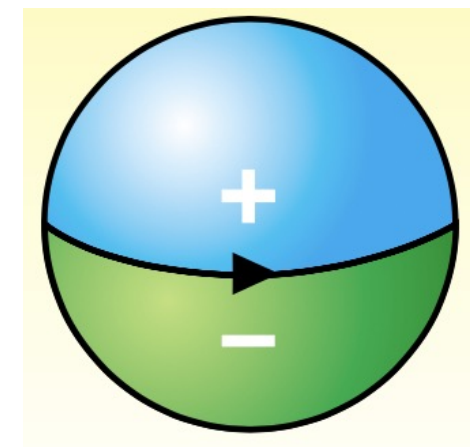
2023 EDM Limits: Free of Standard Model (SM) “Backgrounds”

Chupp, Fierlinger, Ramsey-Musolf, JTS RMP 91:015001 (2019) & Nature 562:355 (2018)
& PRL 124:081803 (2020) & PRL 129:231801 (2022) & Science 381:46 (2023)

System	Best Limit (95%) 1E-32 <i>e</i> cm	SM estimate 1E-32 <i>e</i> cm	Method (Location)
Neutron	2200000	~1	ultracold neutrons in a bottle (PSI)
“Electron”	1100	~0.00001	cold ThO beam (ACME-III 30x soon) (Chicago / Harvard / Northwestern)
	500		trapped HfF ⁺ (JILA / Boulder)
Hg-199	740	~0.0001	atoms in vapor cell (UW-Seattle)

Imagine a Hg-199 atom that is composed of two oppositely charged hemispherical shells each with charge magnitude *e*...

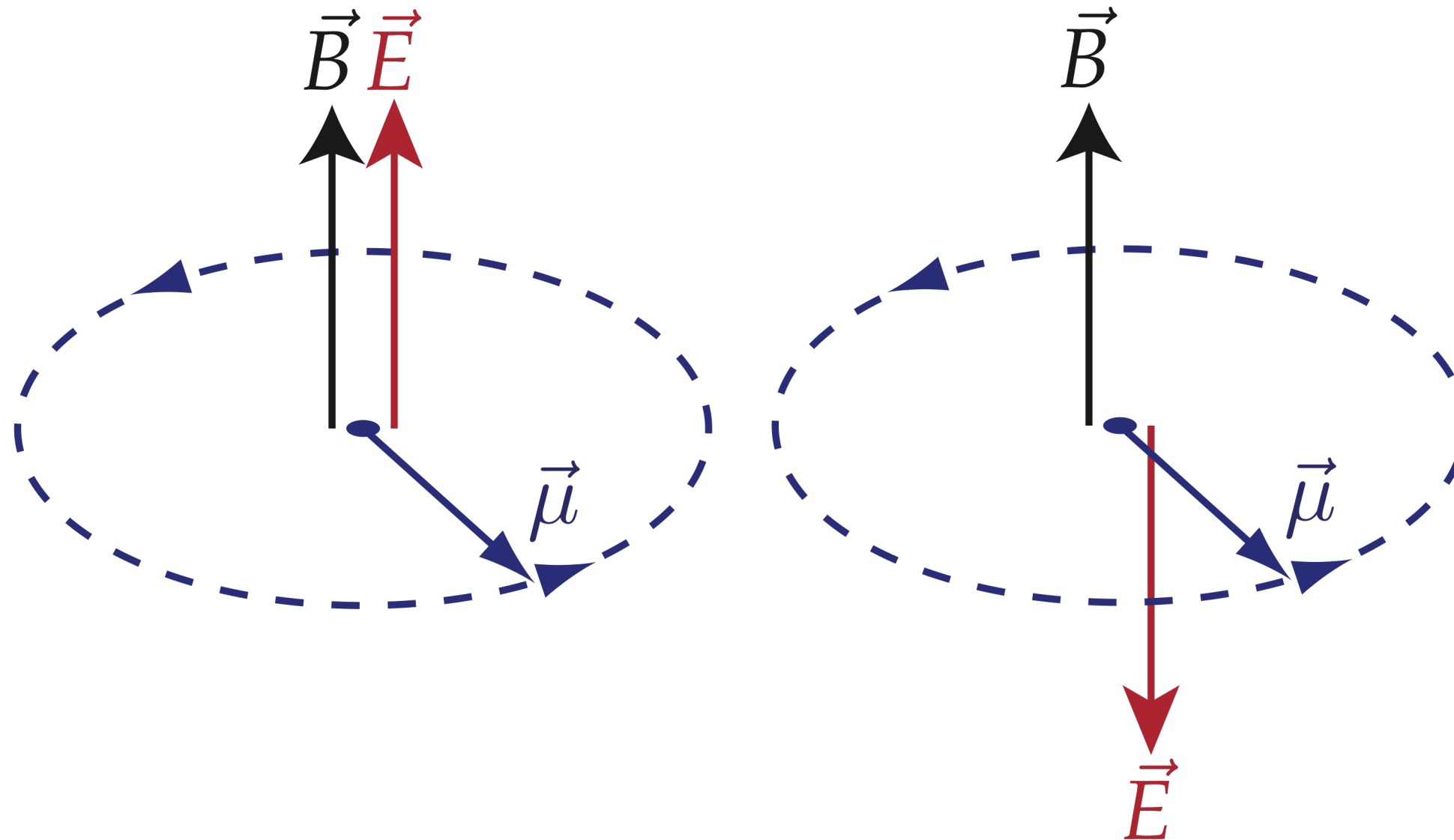
...if the Hg-199 atom was the size of the Earth, then the maximum thickness of these shells would be less than the diameter of a strand of human hair.



Physics Today, June 2003

Always Measure Frequency

Example: Spin Precession of a Spin-1/2 Particle



$$h\nu_{\uparrow} = 2(\mu B_{\uparrow} + dE)$$

$$h\nu_{\downarrow} = 2(\mu B_{\downarrow} - dE)$$

Statistics & Systematics

$$\sigma_\nu = \frac{\Gamma_{\text{linewidth}}}{\text{SNR}}$$

$$\Delta\nu = \nu_\uparrow - \nu_\downarrow = \frac{4dE}{h} + \frac{2\mu(B_\uparrow - B_\downarrow)}{h}$$

Quantum Projection Noise:

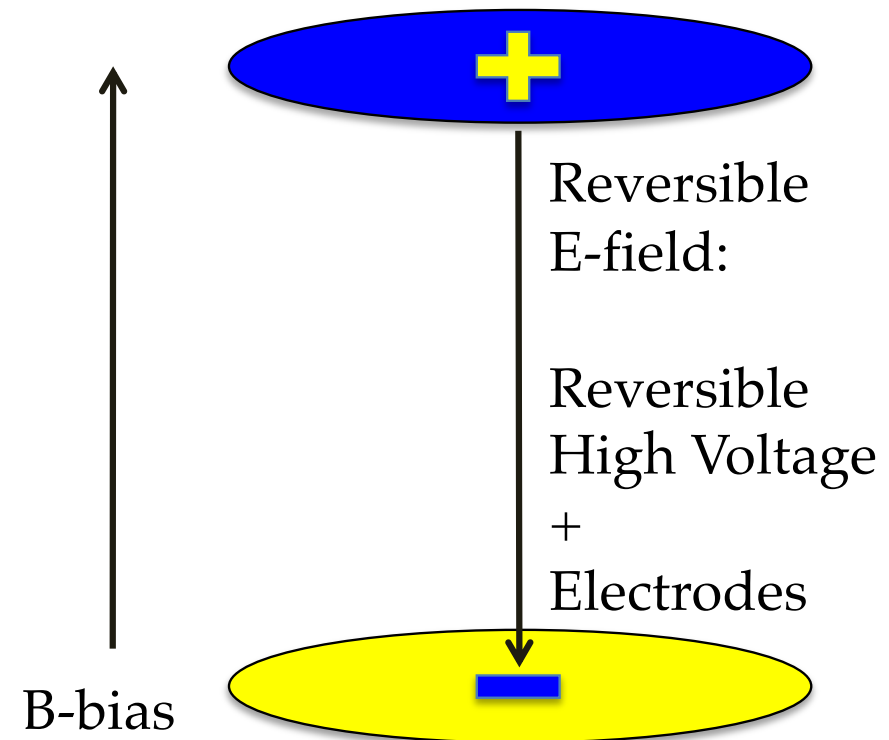
$$\frac{\sigma_d}{\sqrt{N_m}} = \frac{\hbar}{4E\sqrt{N_d T \tau}}$$

Electric field number of detected particles integration time interrogation time

Magnetic Field Instabilities: Annoying

$$\Delta\nu = \nu_{\uparrow} - \nu_{\downarrow} = \frac{4dE}{h} + \frac{2\mu(B_{\uparrow} - B_{\downarrow})}{h}$$

Instabilities adds noise & limits the statistical precision.



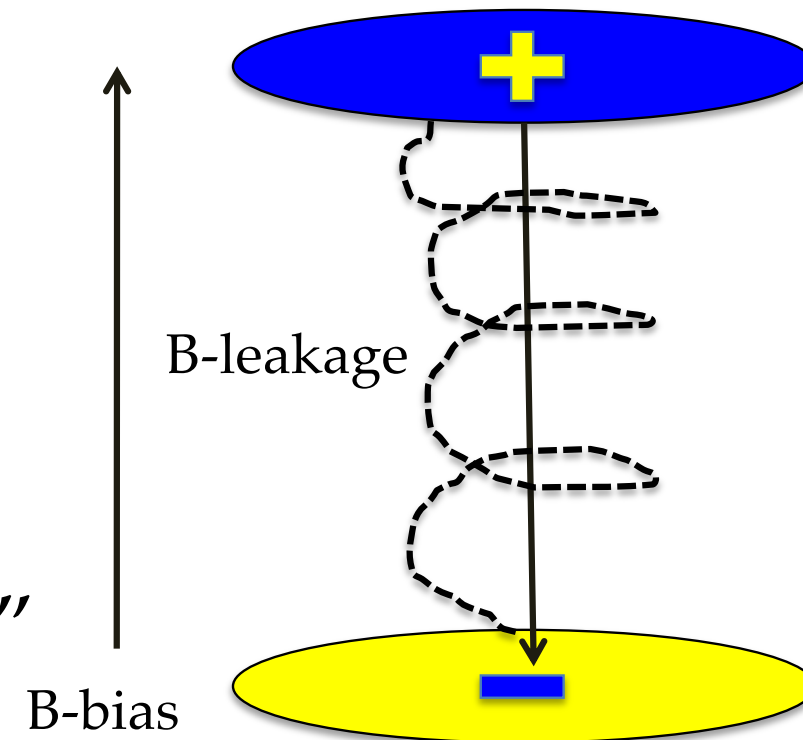
Electric Field-Correlated Systematic: Killer

$$\Delta\nu = \nu_{\uparrow} - \nu_{\downarrow} = \frac{4dE}{h} + \frac{2\mu(B_{\uparrow} - B_{\downarrow})}{h}$$

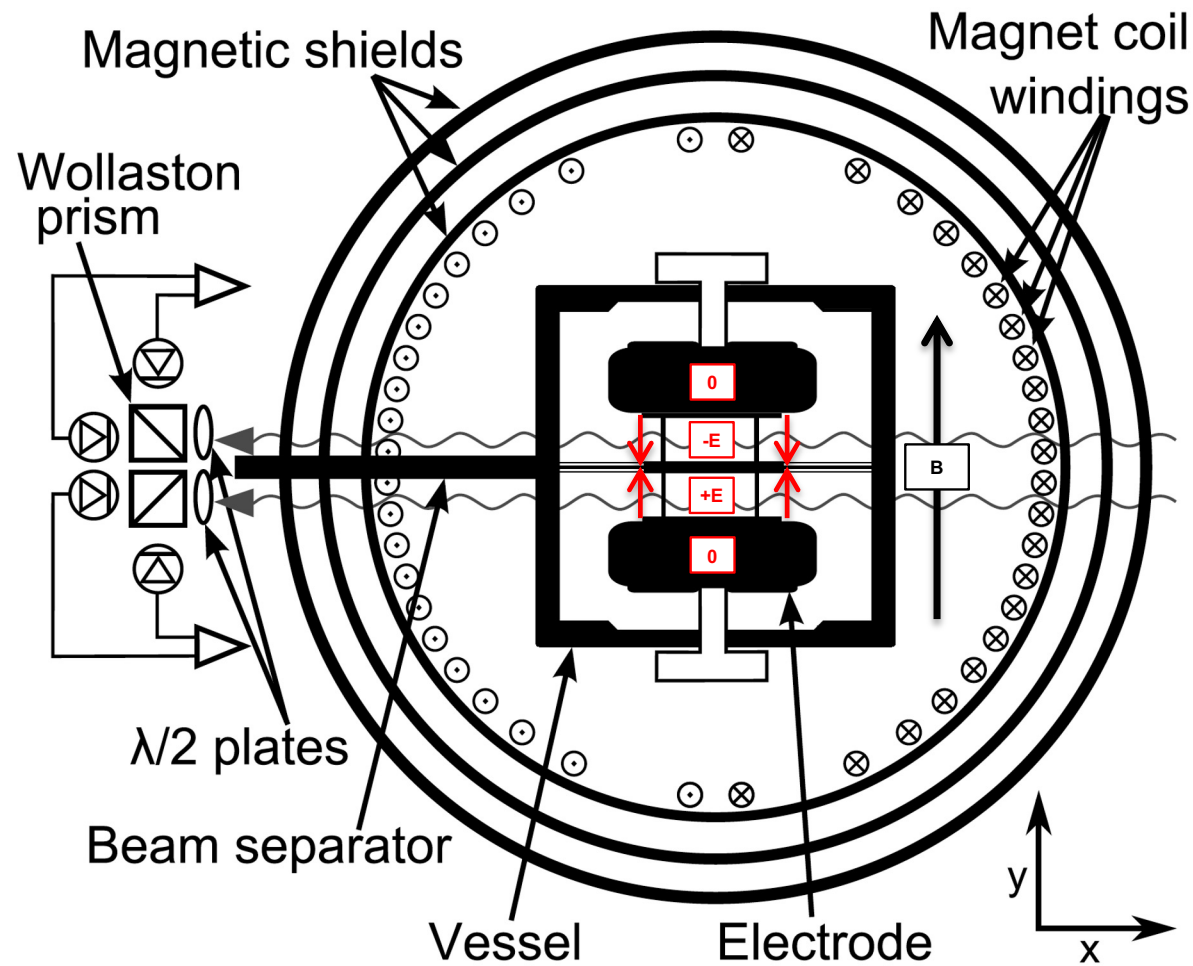
challenge!

Instabilities adds noise & limits the statistical precision.

“False” effects, things which change sign with the electric field, are nasty: “leakage current”



The Gold Standard: Hg-199 EDM Search



- diamagnetic, 1S_0 ground state
- $I = 1/2$, no elect. quad. moment
- high Z , (80) rel. atomic struct.
- stable, (17% n.a.) 92% enriched
- high vapor pressure, ($10^{13} / \text{cm}^3$)
- modest electric field, 10 kV / cm
- 30+ year old experiment!

Limiting systematic appears to be ~ 10 nm scale motion of vapor cells when HV is switched in the presence of 2nd order B -field gradients.

$$\nu = 8.3 \text{ Hz}$$

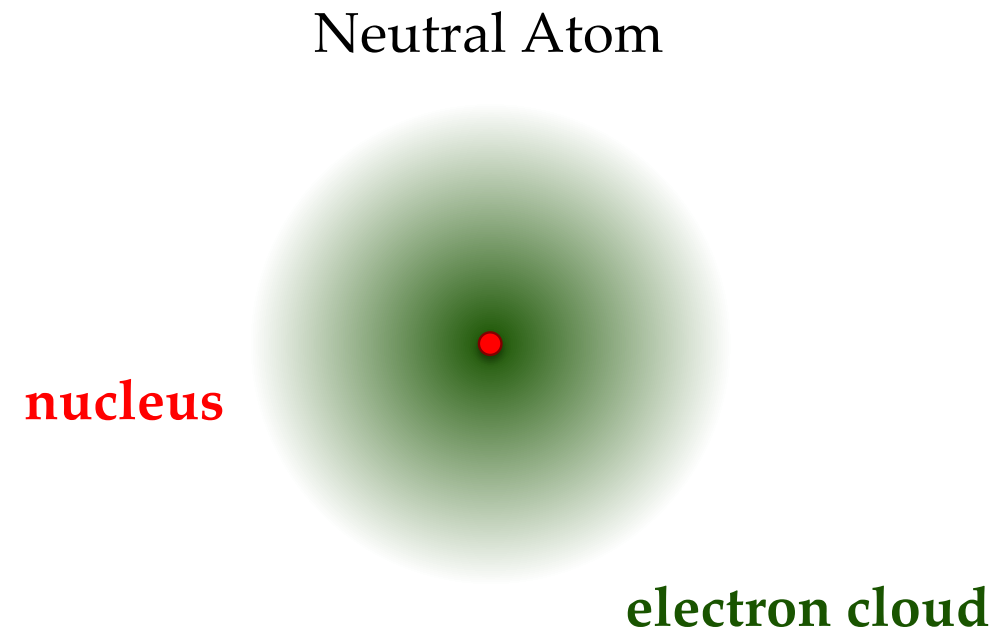
$$\Delta\nu \leq 0.1 \text{ nHz}$$

The best limit on atomic EDM:

$$\text{EDM}(^{199}\text{Hg}) < 0.74 \times 10^{-29} \text{ e-cm (95\% C.L.)}$$

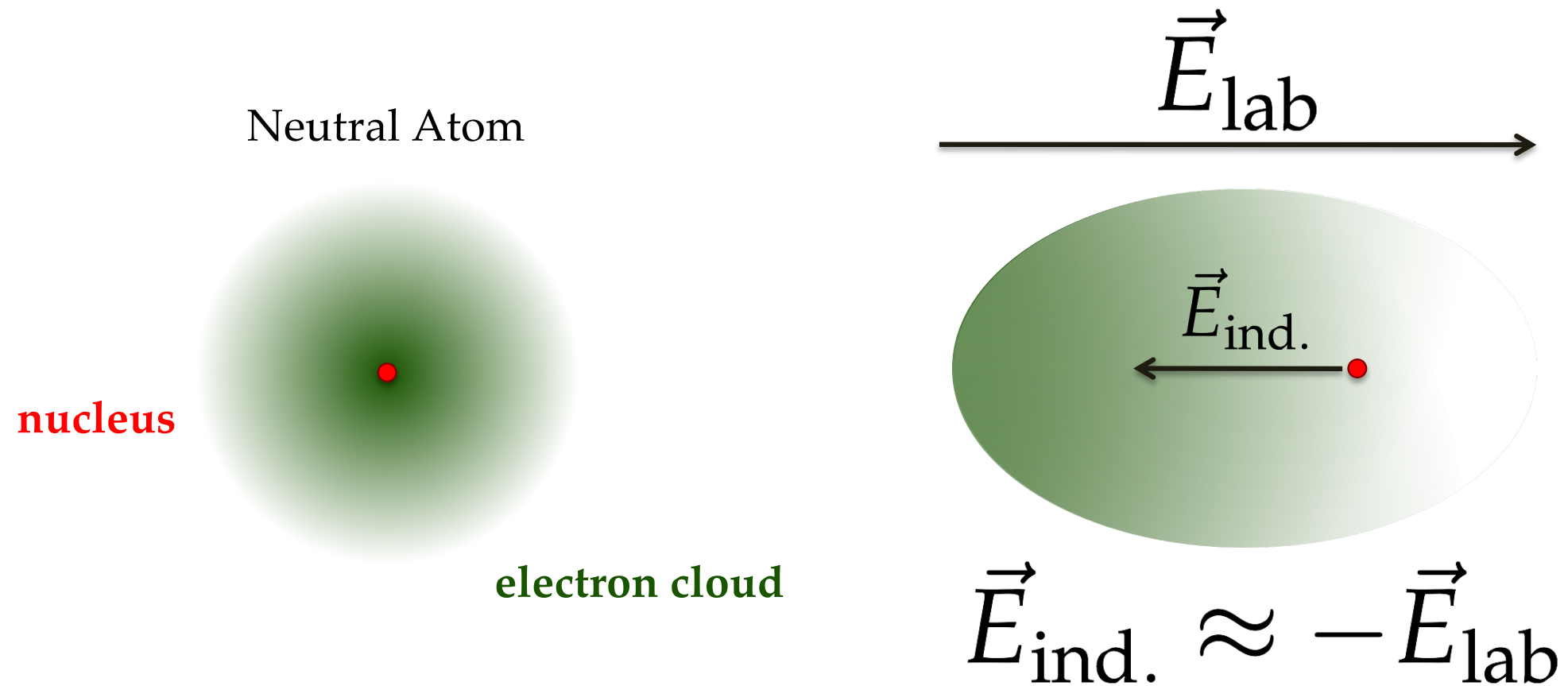
Graner et al., PRL 116:161601 (2016)

Diamagnetic Atoms: All electrons are paired.



Schiff Shielding in Diamagnetic Atoms

- **Shielding in Diamagnetic Atoms**
Schiff PR 132:2194 (1963)



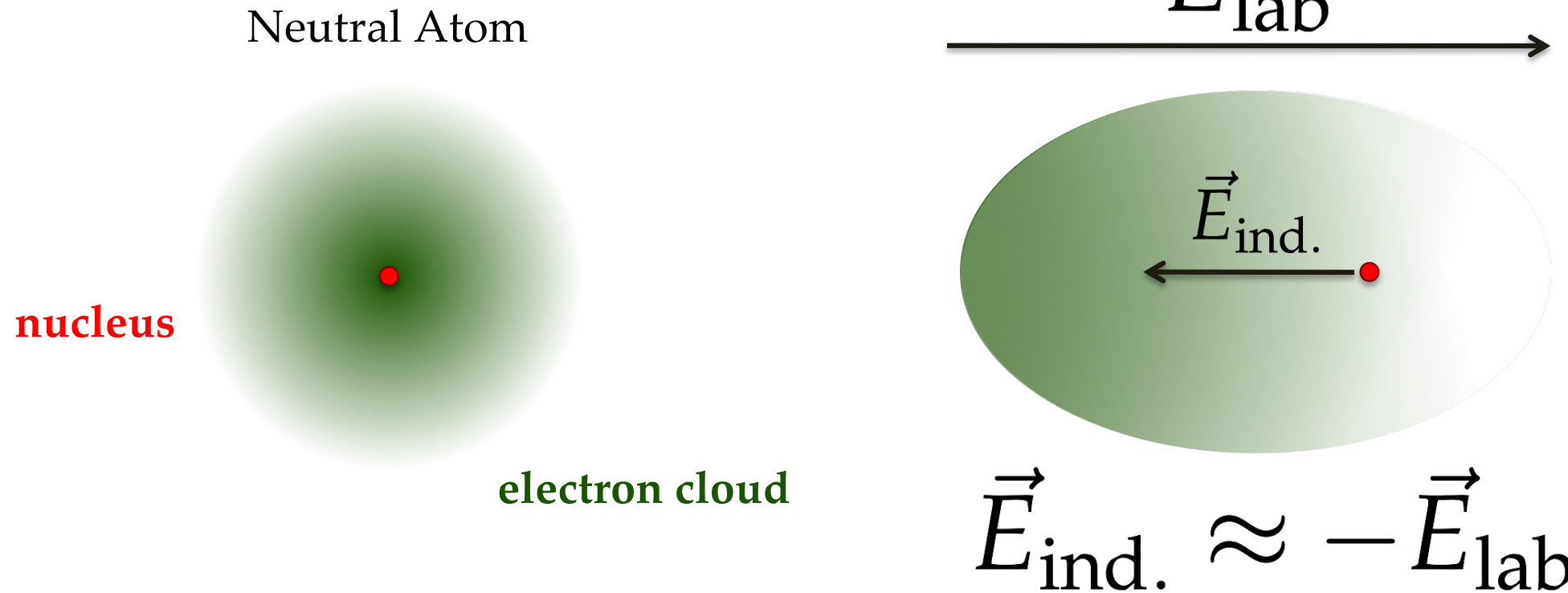
Shielding Imperfect in Relativistic Atoms with Nonzero Nuclear Size

- **Shielding in Diamagnetic Atoms**
Schiff PR 132:2194 (1963)
- **Relativistic atoms: The Sandars-Bouchiat Z^3 "Law"**
Physics Letters 22:290 (1966) & Physics Letters 48B:111 (1974)
- **^{225}Ra vs ^{199}Hg vs. ^3He : 2.8 to 1 to 10^{-5}**
JPB:AMOP 53:195004 (2020) & Phys. Rev. A 106, 022817 (2022)

Madame
Professor
Marie-Anne
Bouchiat



M.A. Bouchiat
photo: Zolotrev



Residual \mathbb{P} & \mathbb{T} Observable: Nuclear Schiff Moment

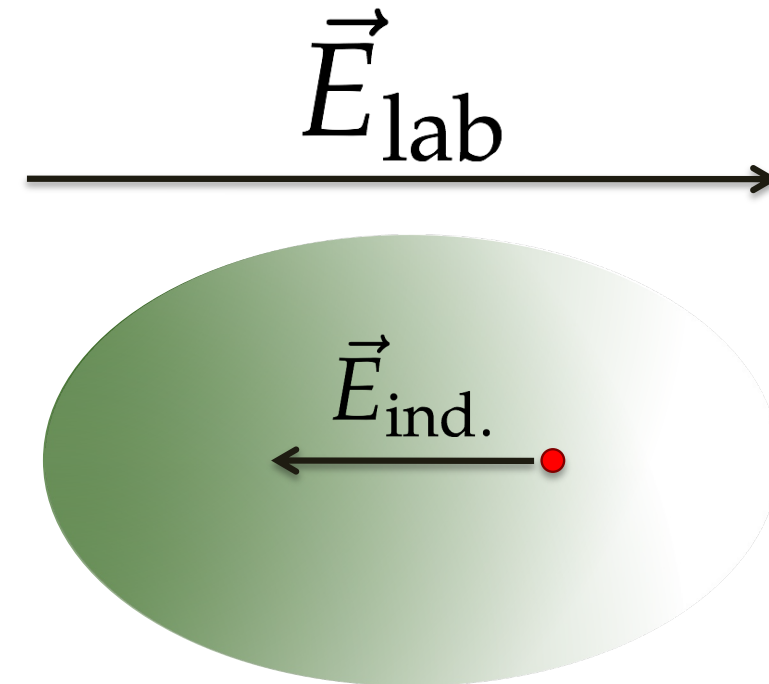
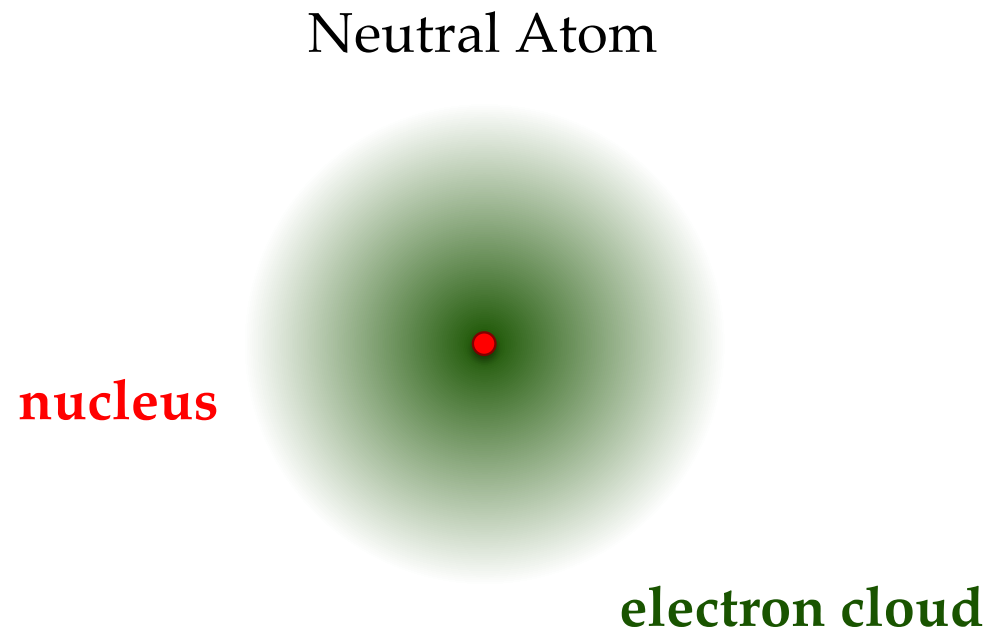
- **Shielding in Diamagnetic Atoms**
Schiff PR 132:2194 (1963)

- **Relativistic atoms: The Sandars-Bouchiat Z^3 "Law"**
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- **^{225}Ra vs ^{199}Hg vs. ^3He : 2.8 to 1 to 10^{-5}**
JPB:AMOP 53:195004 (2020) & Phys. Rev. A 106, 022817 (2022)

Zh. Eksp. Teor. Fiz. 87, 1521-1540 (1984)

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



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

P & T Physics: First Order Perturbation Theory

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

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

- The P and T physics that we seek (unknown & common to all isotopes)

Isotopes With Nearly Degenerate Nuclear States

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

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

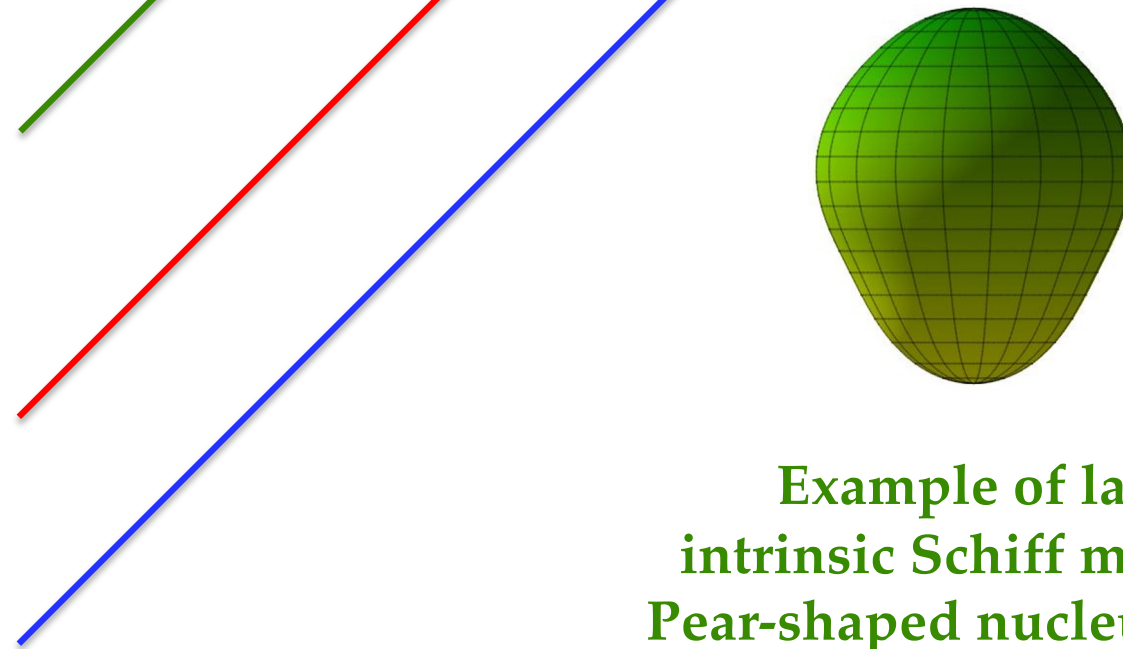
- Difference in lab-frame nuclear energy levels
- The \mathbb{P} and \mathbb{T} physics that we seek (unknown & common to all isotopes)

Nuclear Schiff Moment in the Lab Frame

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

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

- Body-frame Schiff moment – large when there are intrinsic nuclear deformations
- Difference in lab-frame nuclear energy levels
- The \mathbb{P} and \mathbb{T} physics that we seek (unknown & common to all isotopes)



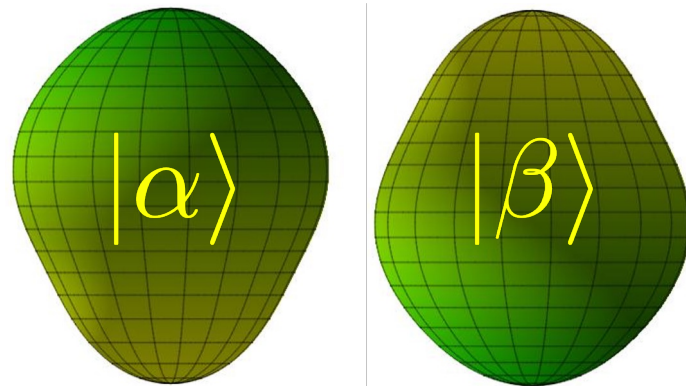
Example of large intrinsic Schiff moment: Pear-shaped nucleus in the “body-frame”

Example: Enhanced Sensitivity in Radium-225

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

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

Parity Doublet



$$\begin{array}{l}
 \text{---} \\
 \uparrow 55 \text{ keV} \\
 |\Psi_1\rangle = \frac{|\alpha\rangle - |\beta\rangle}{\sqrt{2}} \\
 \downarrow \\
 \text{---} \\
 |\Psi_0\rangle = \frac{|\alpha\rangle + |\beta\rangle}{\sqrt{2}}
 \end{array}$$

- 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)

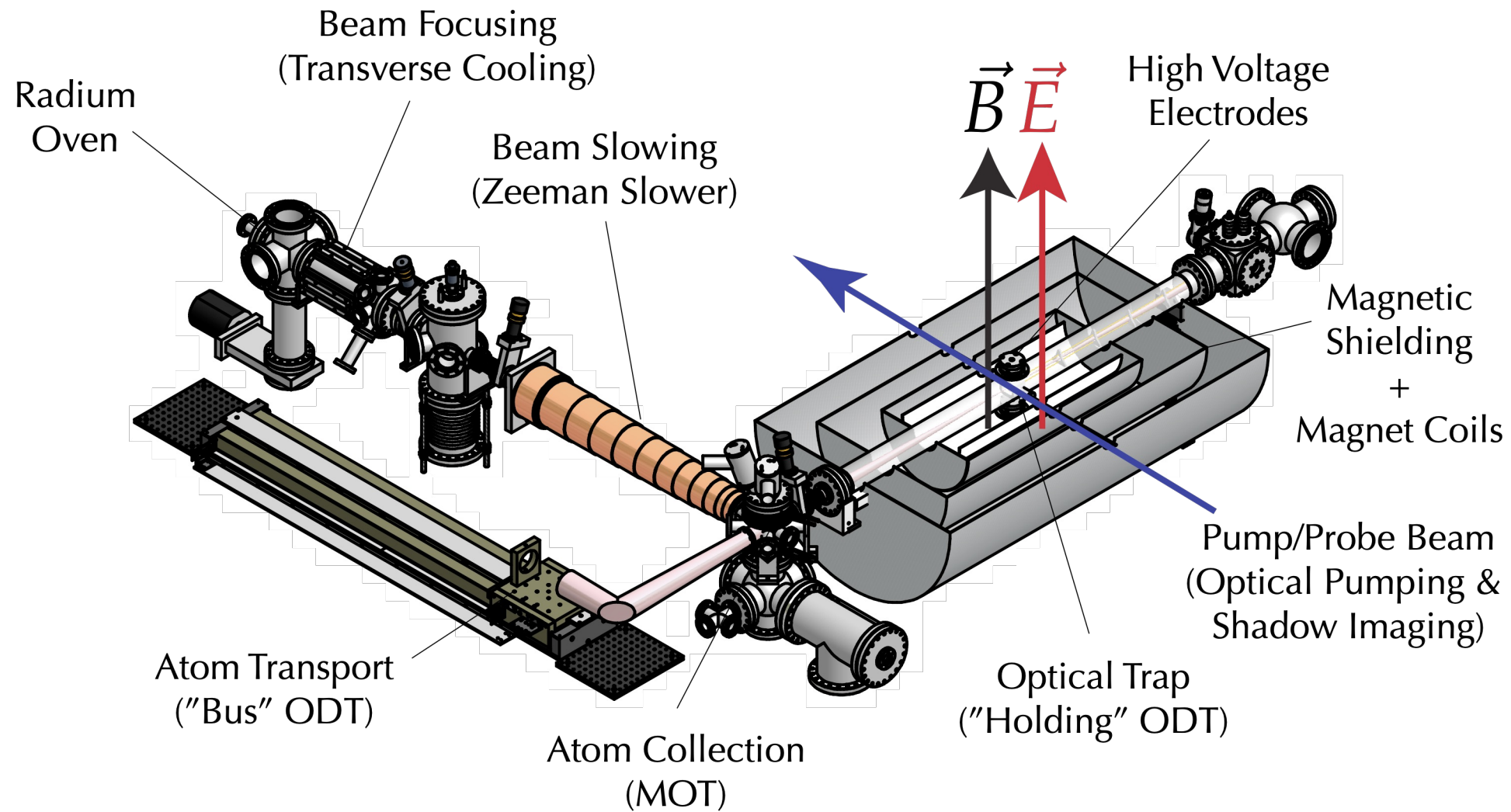
Total Enhancement Factor: EDM (²²⁵Ra) / EDM (¹⁹⁹Hg)

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

²²⁵Ra: Dobaczewski & Engel PRL 94:232502 (2005)

¹⁹⁹Hg: Ban et al. PRC 82:015501 (2010)

Laser Trap Atomic Ra EDM @ ANL w/ MSU+Kentucky



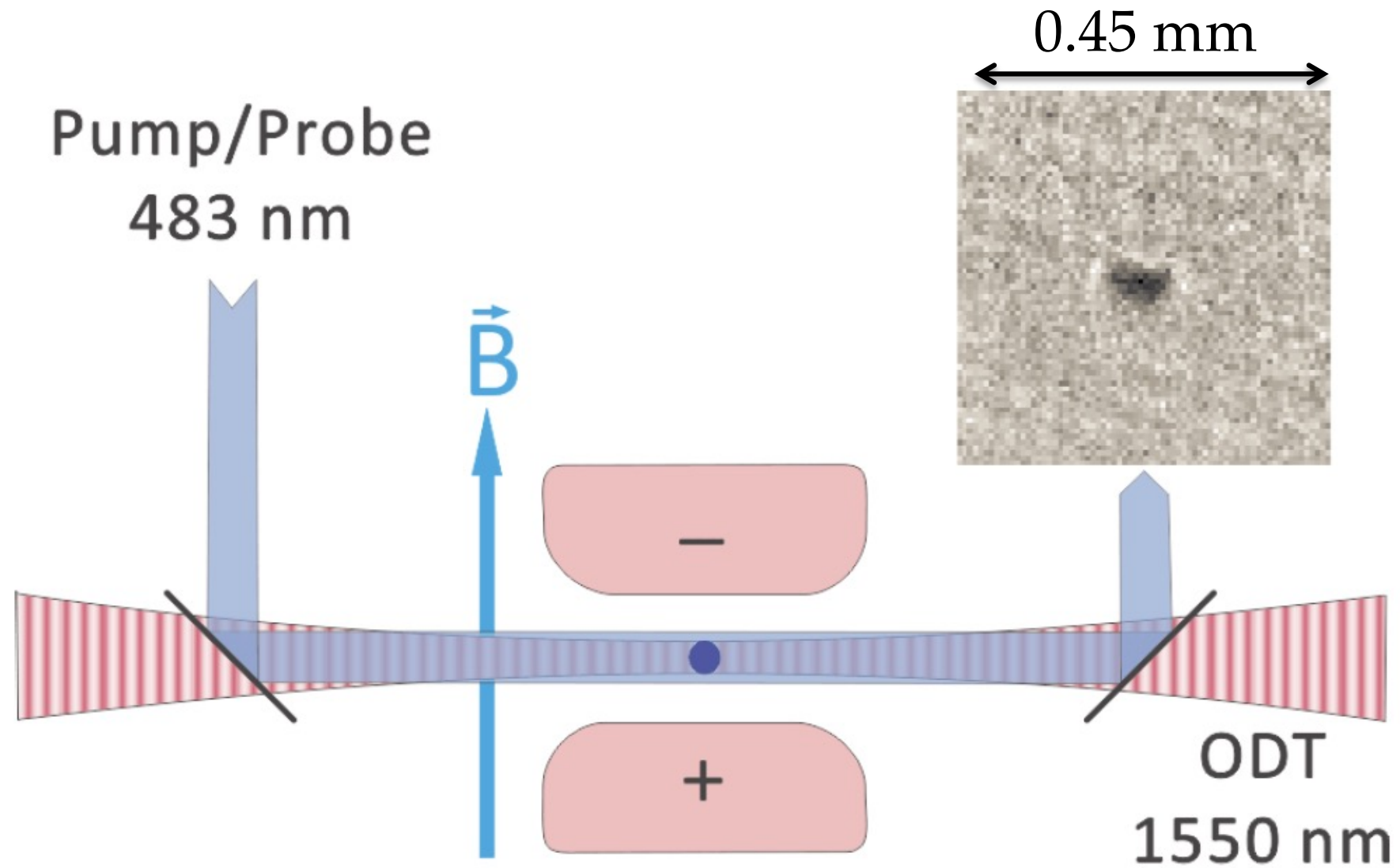
PRC 94:025501

$|d(\text{Ra-225})| < 1.4 \times 10^{-23} e \text{ cm} (95\%)$

completely statistics limited

several upgrades underway

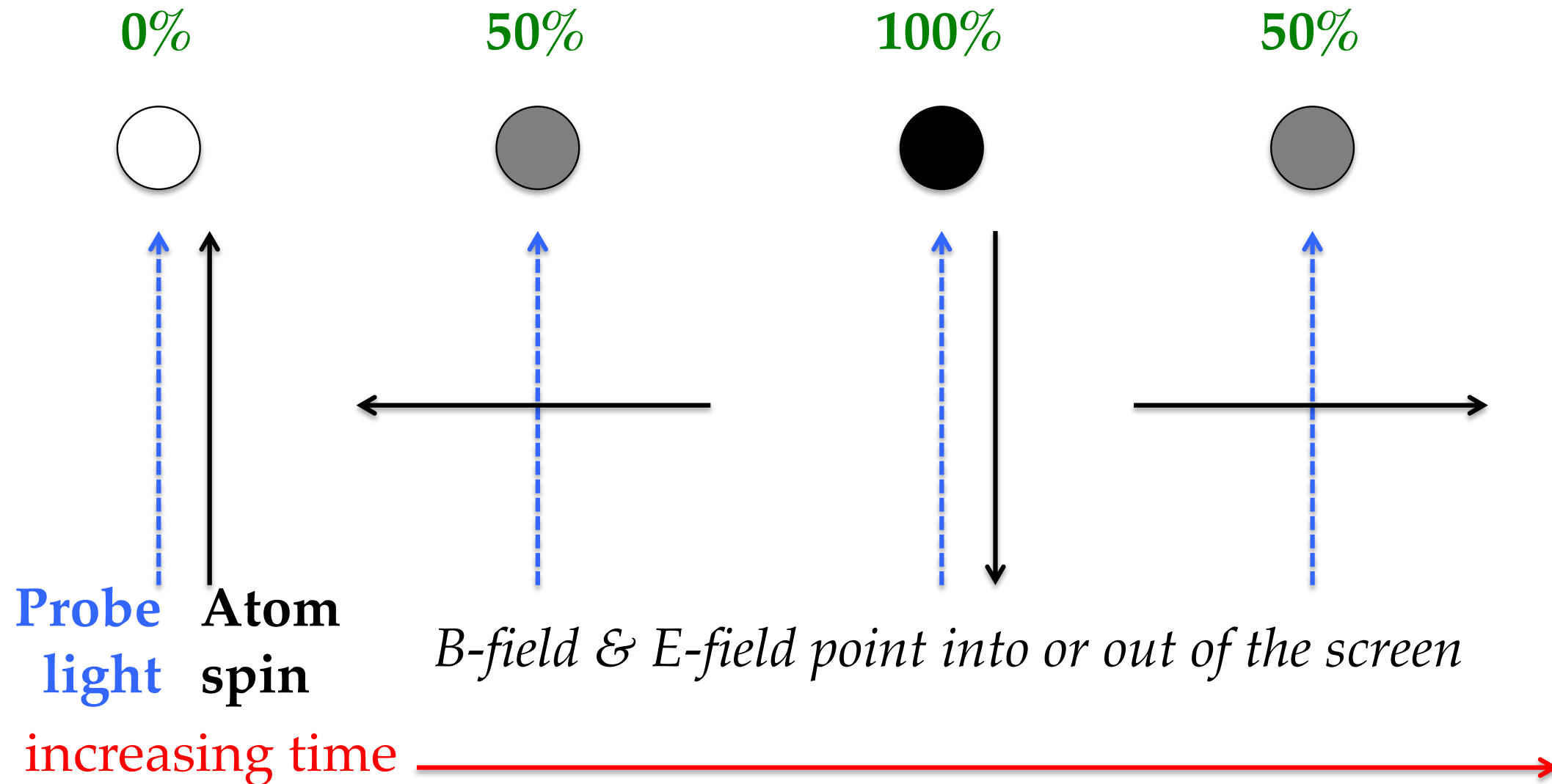
Atoms Create a Shadow by Absorption



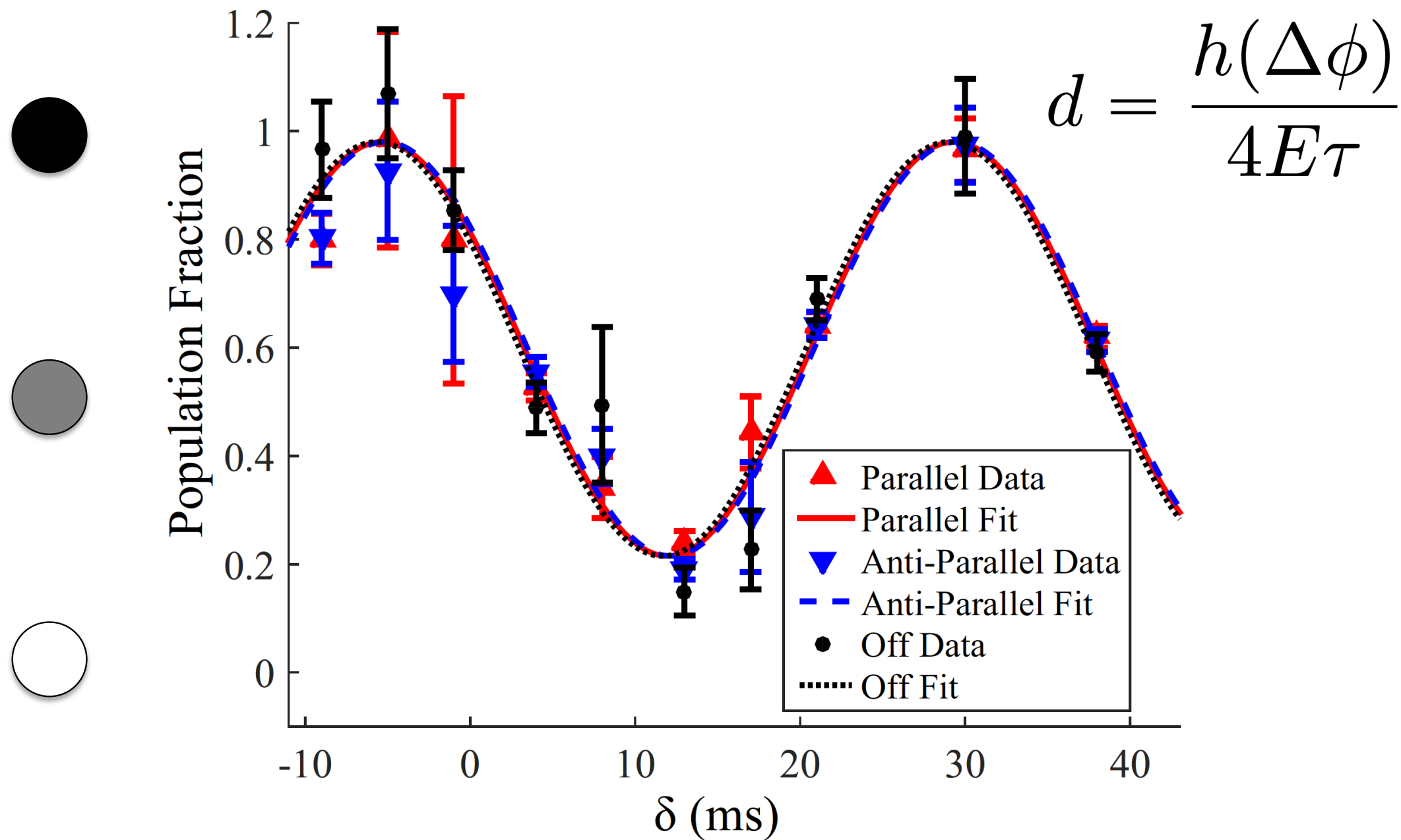
Parker et al. Phys. Rev. Lett. 114, 233002 (2015)

Absorption probability oscillates at ~20 Hz

probability of absorbing probe light and creating a shadow:



Reconstructed Spin Precession Curve After 20s



Towards More Statistics

Dec 2014: PRL 114:233002: $|d(\text{Ra-225})| < 50 \times 10^{-23} e \text{ cm}$ (95%)

June 2015: PRC 94:025501: $|d(\text{Ra-225})| < 1.4 \times 10^{-23} e \text{ cm}$ (95%)

Effect	Current uncertainty	α scenario uncertainty	β scenario uncertainty
E-squared effects	1×10^{-25}	7×10^{-29}	7×10^{-31a}
B-field correlations	1×10^{-25}	5×10^{-27}	3×10^{-29a}
Holding ODT power correlations	6×10^{-26}	9×10^{-30}	9×10^{-32a}
Stark interference	6×10^{-26}	2×10^{-27}	3×10^{-29a}
E-field ramping	9×10^{-28}	2×10^{-29}	N/A
Blue laser power correlations	7×10^{-28}	1×10^{-31}	1×10^{-31}
Blue laser frequency correlations	4×10^{-28}	8×10^{-30}	8×10^{-30}
$\mathbf{E} \times \mathbf{v}$ effects	4×10^{-28}	7×10^{-30}	N/A
Leakage current	3×10^{-28}	9×10^{-29}	N/A
Geometric phase	3×10^{-31}	7×10^{-30}	5×10^{-33}
Total	2×10^{-25}	5×10^{-27}	4×10^{-29a}

^aThis uncertainty will improve with the statistical sensitivity of the experiment.

1. More efficient detection of atoms: **Tenzin Rabga (Ph.D. 2020)**
2. Higher electric field: **Roy Ready (Ph.D. 2021)**
3. More efficient laser cooling & trapping: **GAK**
4. More atoms: (Isotope Harvesting at FRIB)



Dr. Gordon Arrowsmith-Kron
MSU Physics & Astronomy
Department:
Best Thesis Prize, 2025

Recent Results in Xe-129 and Yb-171 (Not Pear-Shaped)

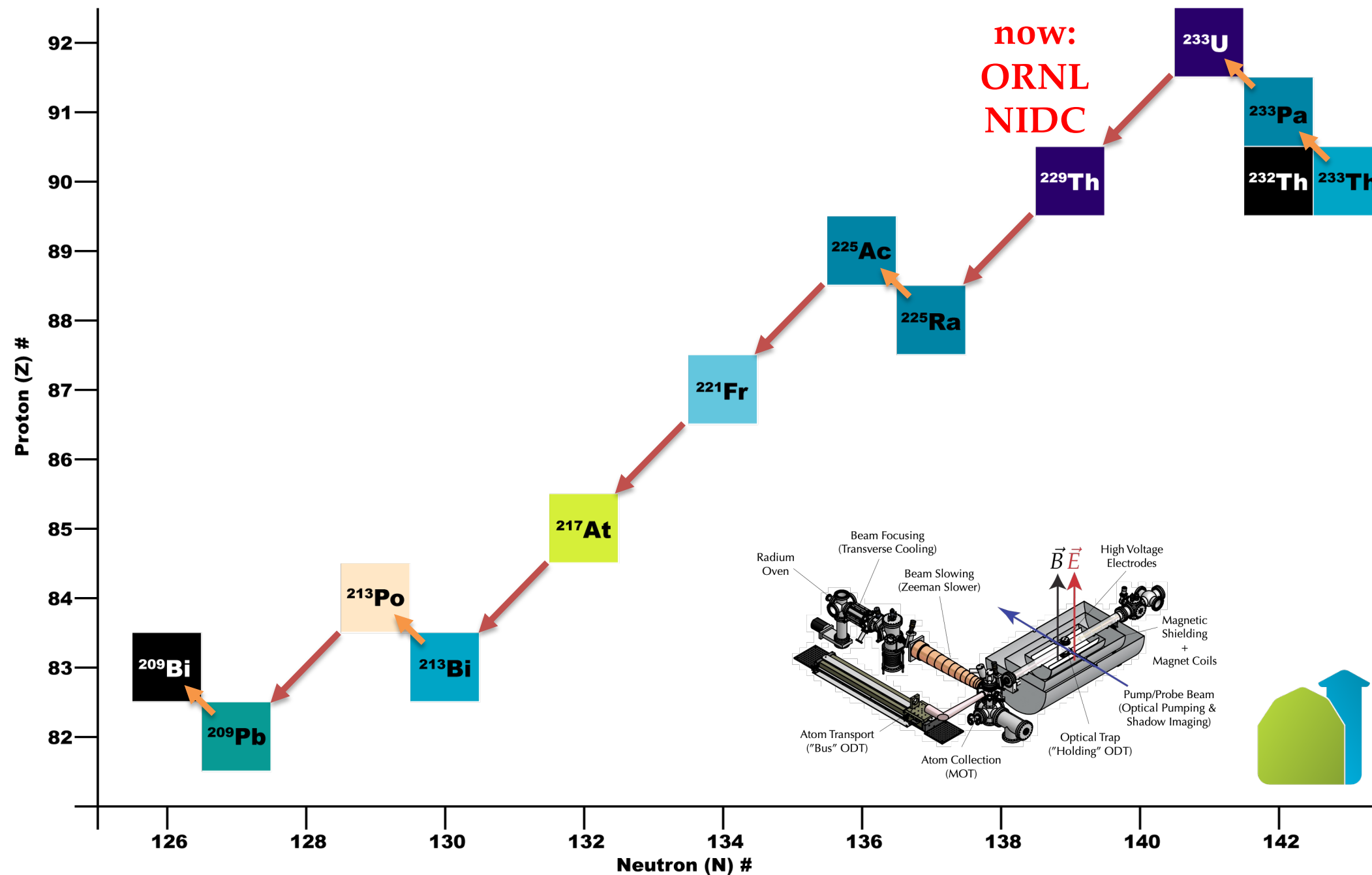
Ra-225: PRC 94:025501 (2016): $< 14000 \times 10^{-27} e \text{ cm}$ (95%)
(laser trap experiment – 1000x in a “few” years)

Xe-129: PRL 123:143003 (2019): $< 1.4 \times 10^{-27} e \text{ cm}$ (95%)
(gas cell experiment – new effort @ LANL)

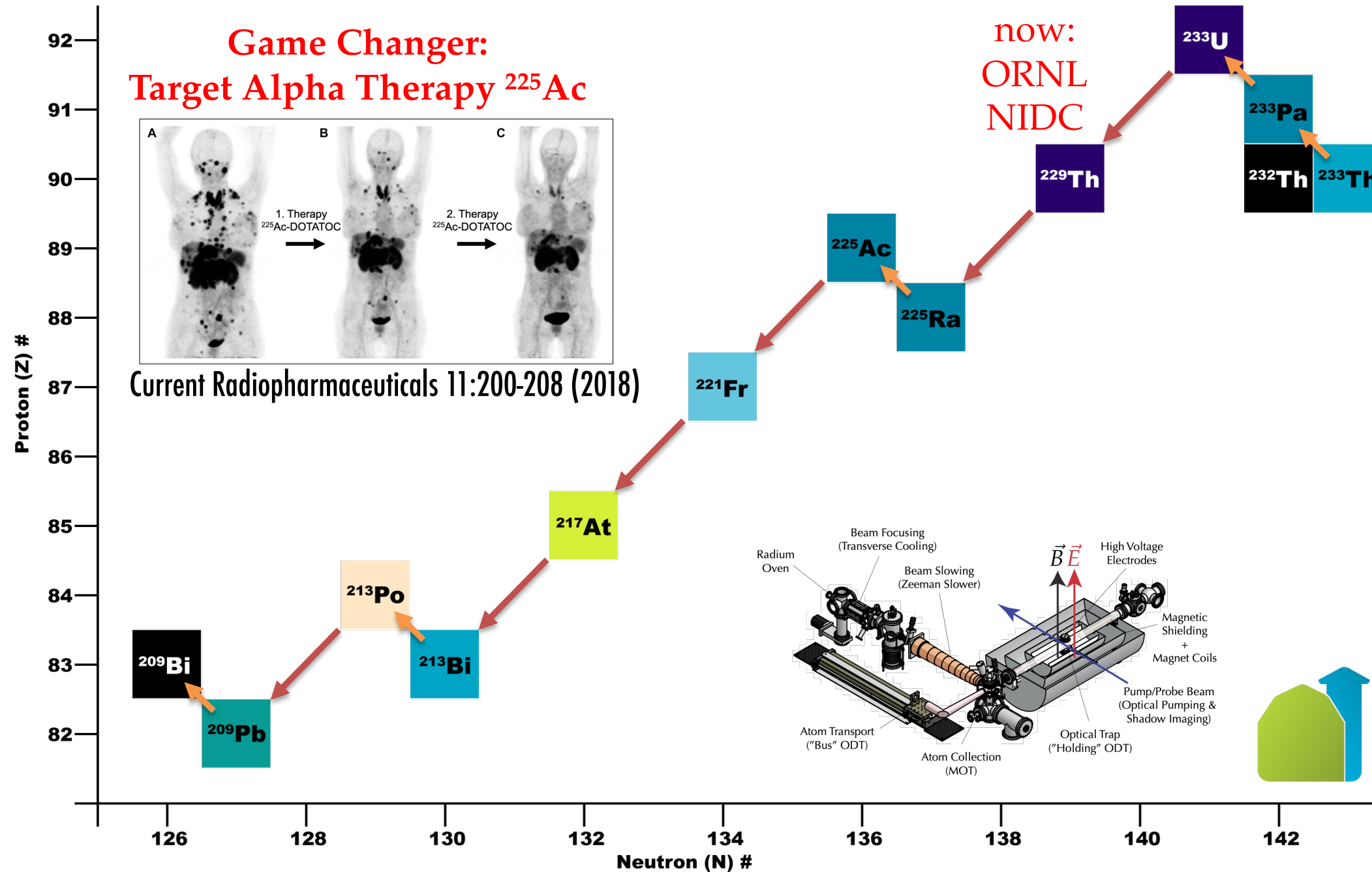
Yb-171: PRL 129:083001 (2022): $< 15 \times 10^{-27} e \text{ cm}$ (95%)
(laser trap experiment – 30x data taking complete)

- At the moment, the new physics constraints within the hadronic sector for all three of these experiments are roughly equal.
- The Yb experiment validates the laser trap approach for Ra for at least another three orders of magnitude.

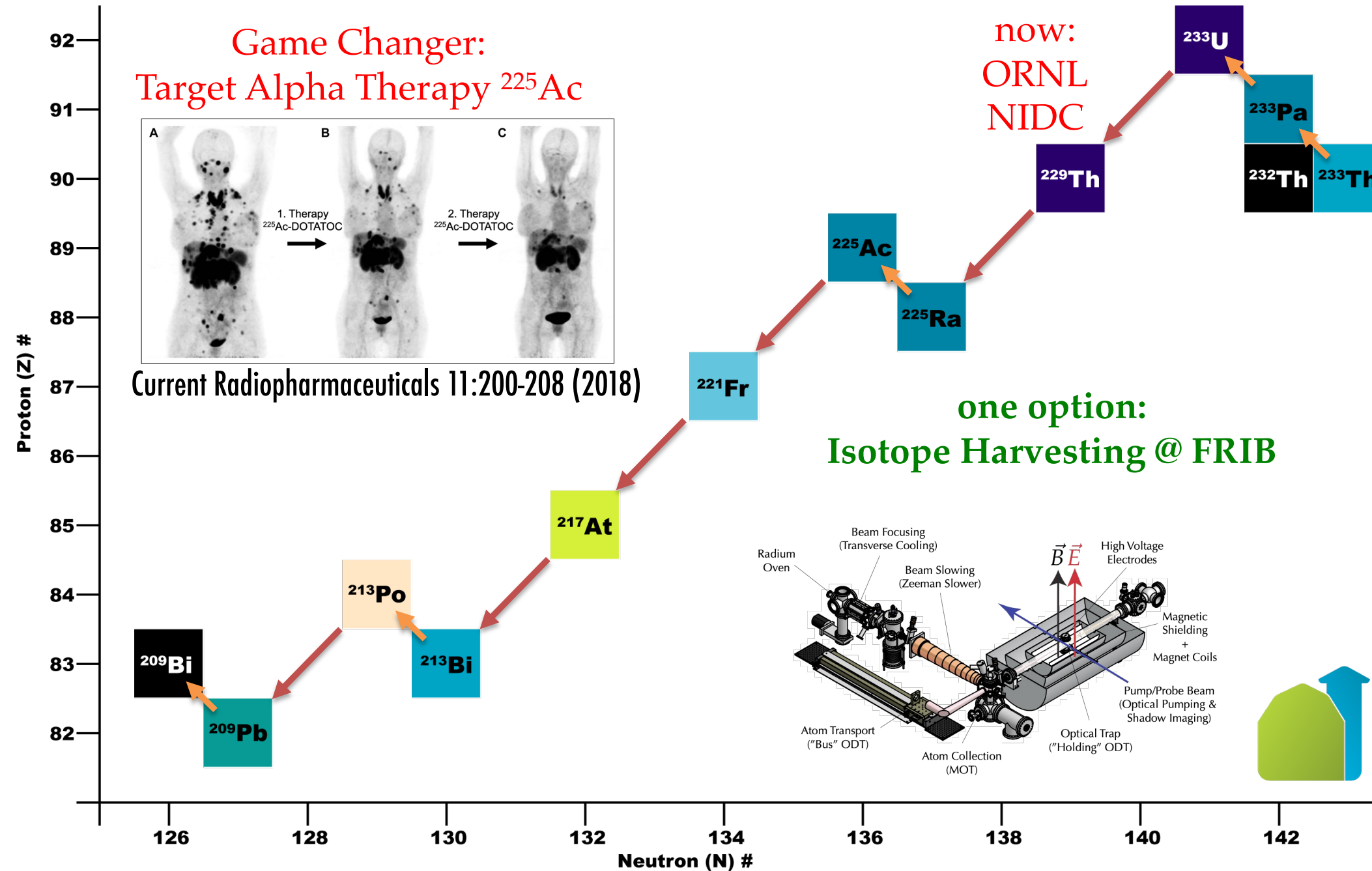
Source of Radium-225 Atoms ($\tau_{1/2} = 15$ days)



Source of Radium-225 Atoms ($\tau_{1/2} = 15$ days)



Source of Radium-225 Atoms ($\tau_{1/2} = 15$ days)



Facility for Rare Isotope Beams @ MSU

Michigan State University
East Lansing, MI
Very Bad at American Football
Home of FRIB



Google Maps & Wikipedia Commons

Facility for Rare Isotope Beams @ MSU

Michigan State University
East Lansing, MI
Very Bad at American Football
Home of FRIB

University of Michigan
Ann Arbor, MI
Very Good at American Football
no FRIB



Google Maps & Wikipedia Commons

“Isotope Harvesting” at The Facility for Rare Isotope Beams (MSU/East Lansing)



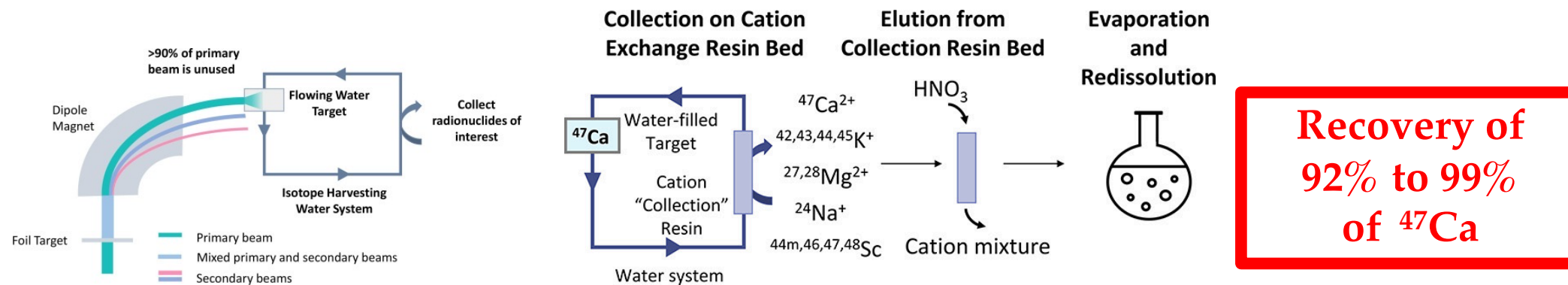
Prof. Greg Severin



Prof. Alyssa Gaiser



Prof. Katharina Domnanich



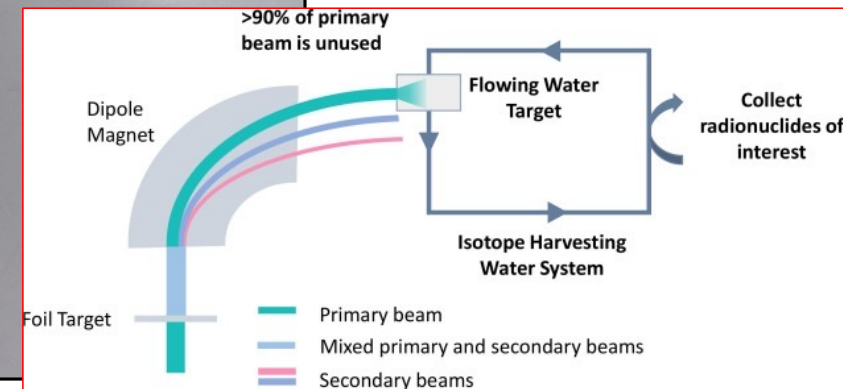
NIMB 478 34 (2020)

Abel et al., ACS Omega 5(43) 27864 (2020)

Isotope Harvesting Vault has been installed!



C. Vyas



30 feet

NIMB 478 34 (2020)

^{225}Ra and ^{229}Pa Are High Sensitivity EDM Candidates that Are Also Favorable Cases for FRIB Production and Radiochemistry

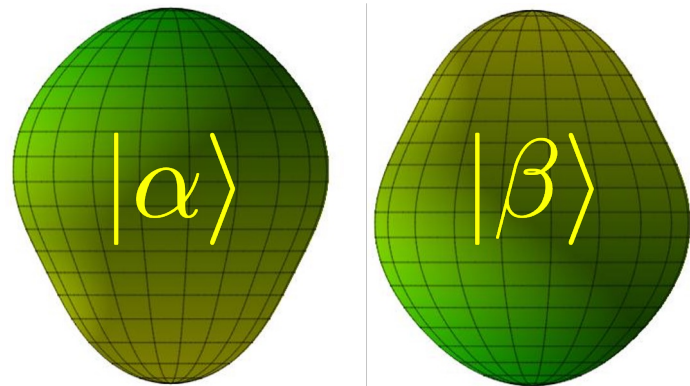
Nuclear Schiff Moment (NSM) = $S_z = \frac{\langle er^2 z \rangle}{10} - \frac{\langle r^2 \rangle \langle ez \rangle}{6}$

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

Choose isotope to maximize prefactor

ARNPS 75 129 (2025)

Parity Doublet



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

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

ΔE

- 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)

EDM (^{225}Ra) / EDM (^{199}Hg)

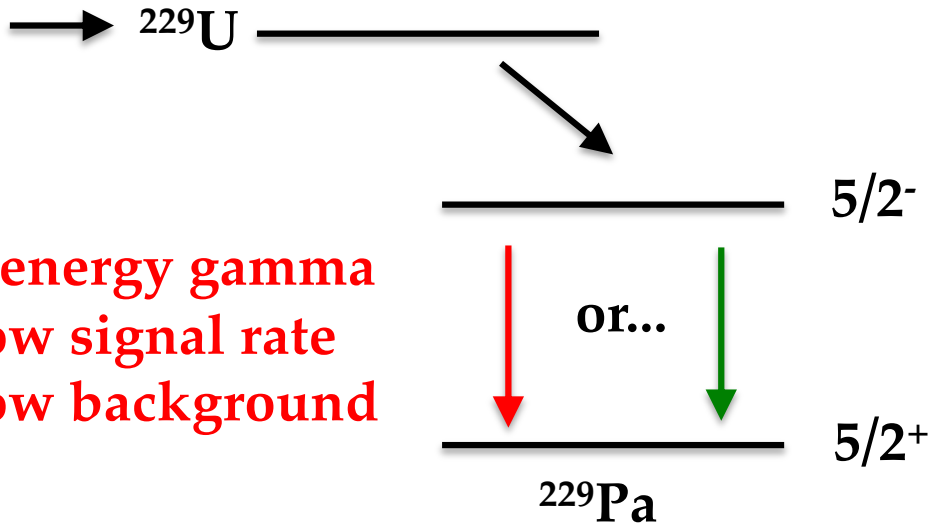
Skyrme Model	Isoscalar	Isovector
SIII	300	4000
SkM*	300	2000
SLy4	700	9000



^{225}Ra : PRL 94:232502 (2005)
 ^{199}Hg : PRC 82:015501 (2010)
 ^{229}Pa : PRC 92:024313 (2015)
 atoms: JPG 46 100501 (2019)

	^{225}Ra	^{229}Pa
pear?	best case known	probably...???
ΔE (keV)	(55.16)	(0.06 +/- 0.05)?
sens. rel. to ^{199}Hg	10^3	$10^6?$
$t_{1/2}$ (d)	14.8	1.55
atoms (20 kW)	$>10^{12}$	$>10^{13}$
atoms (400 kW)	$>10^{13}$	$>10^{14}$

Two Complementary Paths To Search For a Low-Lying Nuclear State in ^{229}Pa

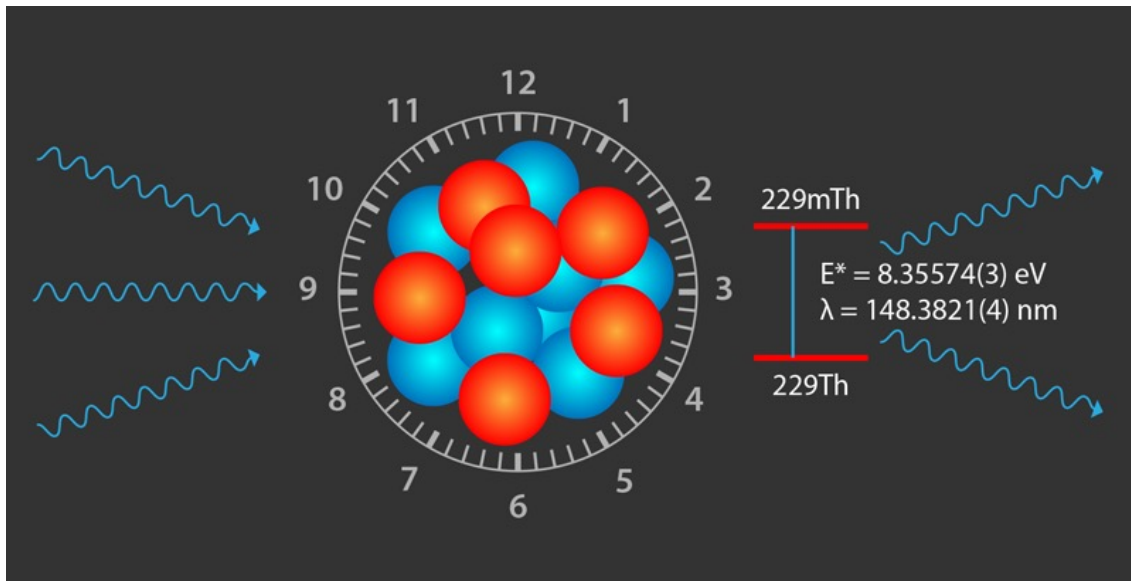


- low energy gamma**
- low signal rate
 - low background

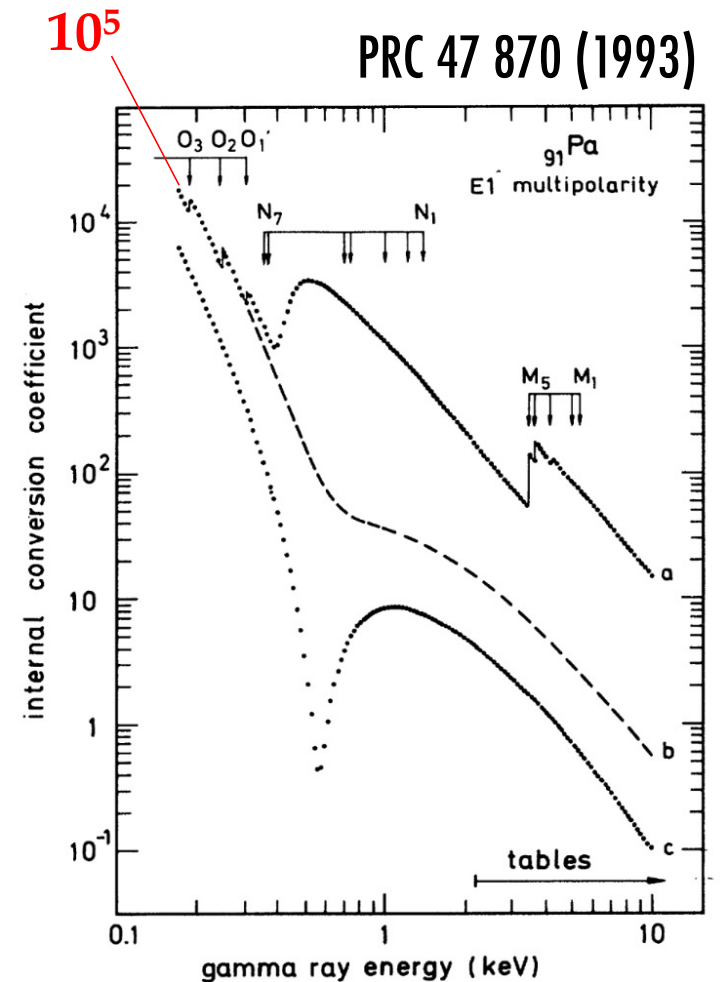
$$\frac{\text{electrons}}{\text{gammas}} \approx \kappa_{\text{Schiff}} \alpha^4 Z^3 \left(\frac{L}{L+1} \right) \left(\frac{2m_e c^2}{\Delta E} \right)^{L+5/2} \approx 10^6 \text{ to } 10^{18}$$

- conversion electron**
- high signal rate
 - high background

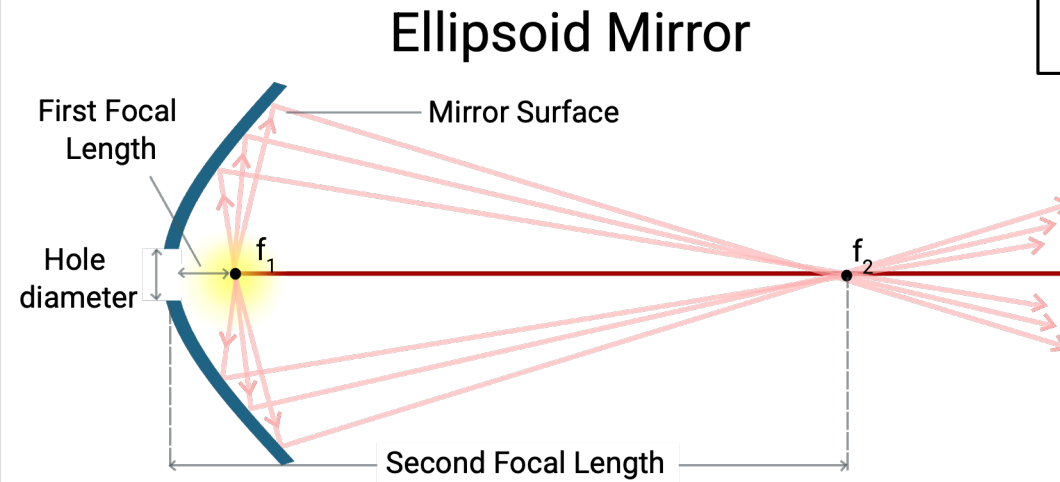
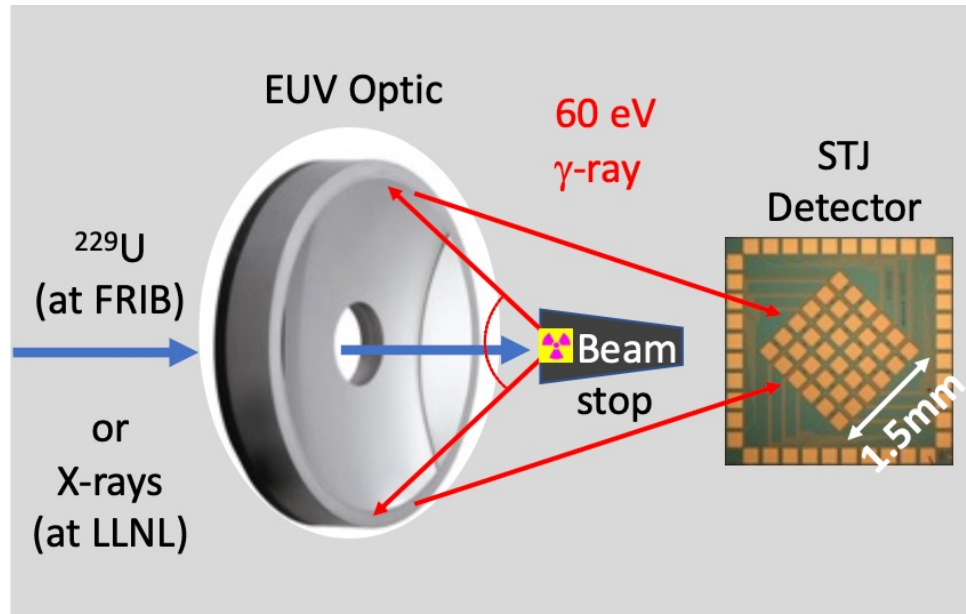
<https://physics.aps.org/articles/v17/71>



	^{229}Th	^{229}Pa
A	229	229
Z	90	91
N	139	138
ΔE	8.36 eV	(60 +/- 50) eV?
PD?	$5/2^+$ $3/2^+$	$5/2^+$ $5/2^-$



Direct Low Energy Gamma Detection (40-80 eV) w/ LLNL



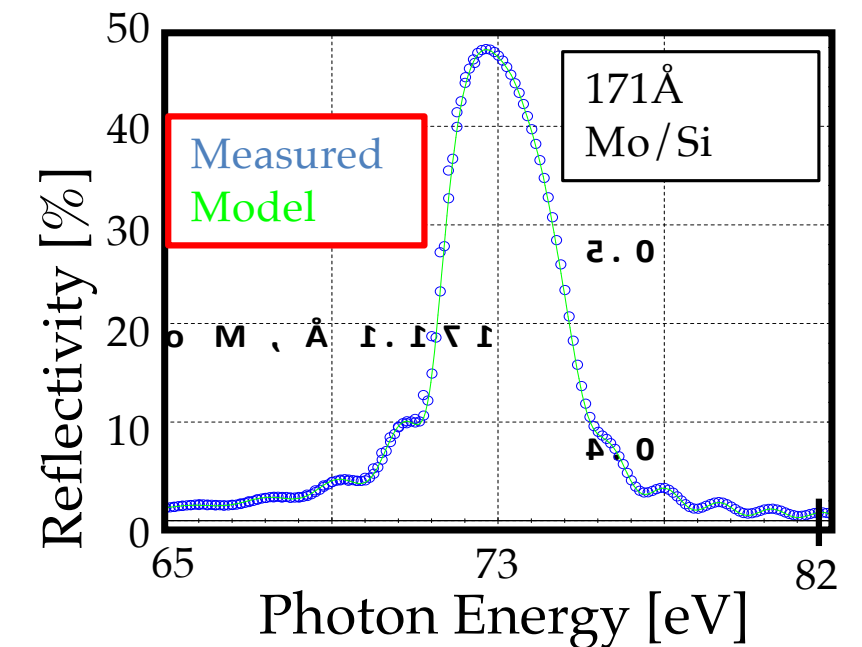
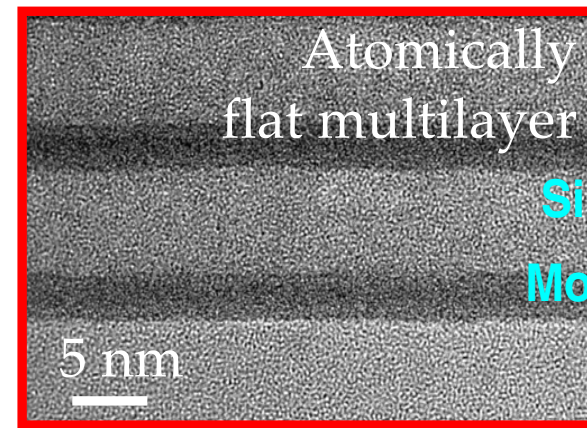
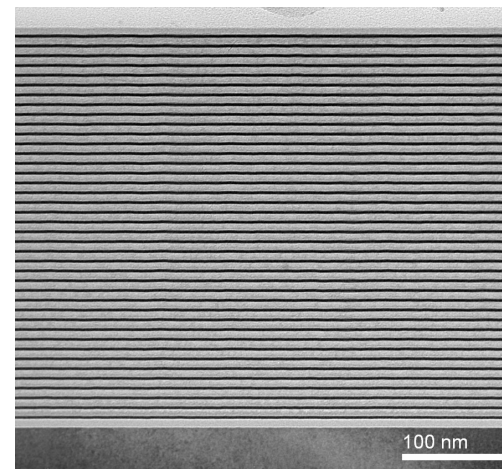
<https://www.meetoptics.com/academy/ellipsoidal-mirrors#what-are-ellipsoidal-mirrors>

STJ: Stephan Friedrich

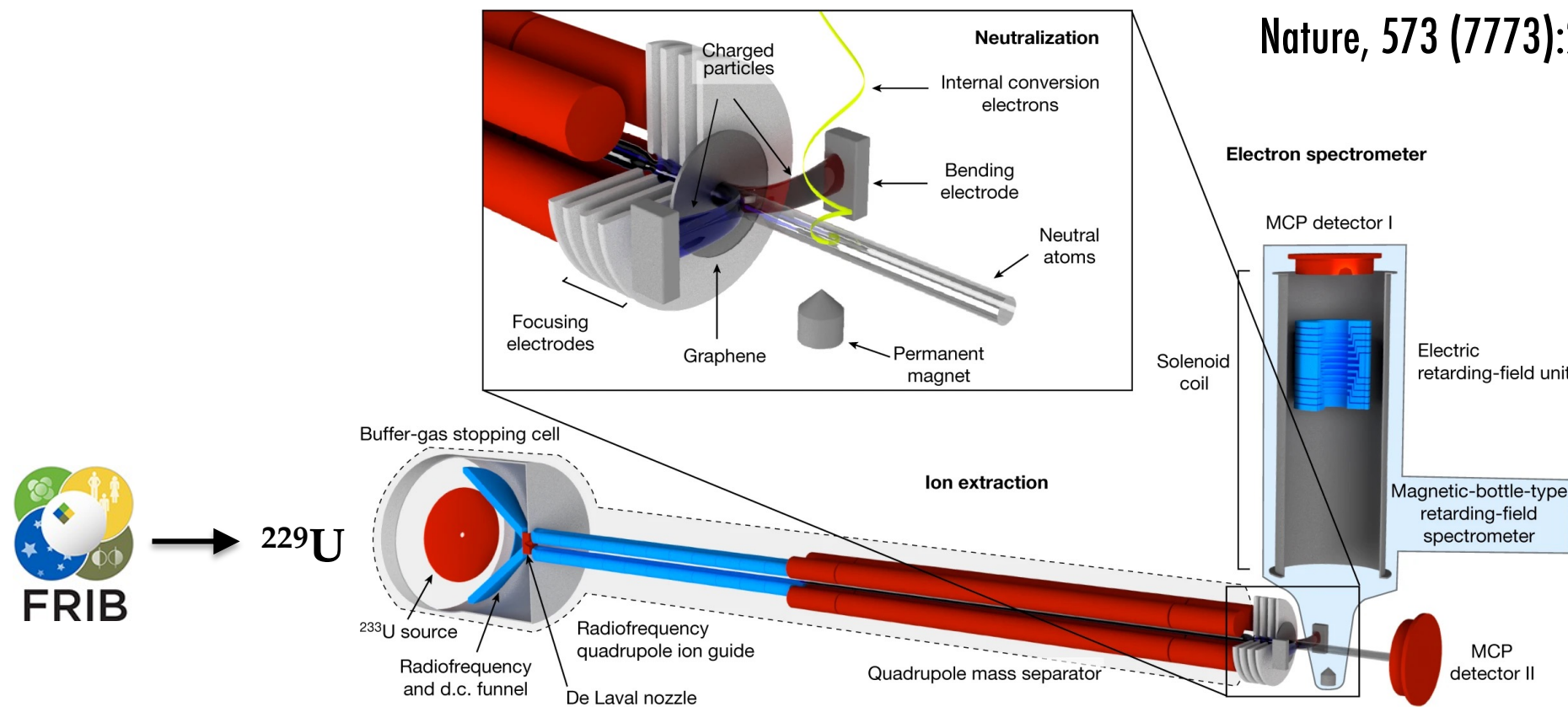
The EUV optic filters the signal gammas from gammas outside the transmission window as well as alphas and betas and focuses them into the STJ.
GEANT simulations underway as well as a proposal paper.



EUUV: Marie-Anne Descalle



^{229}Th Method: Integrated Conversion Electron Spectroscopy

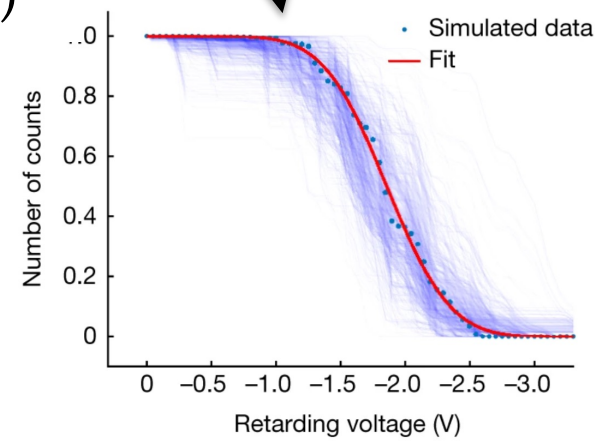
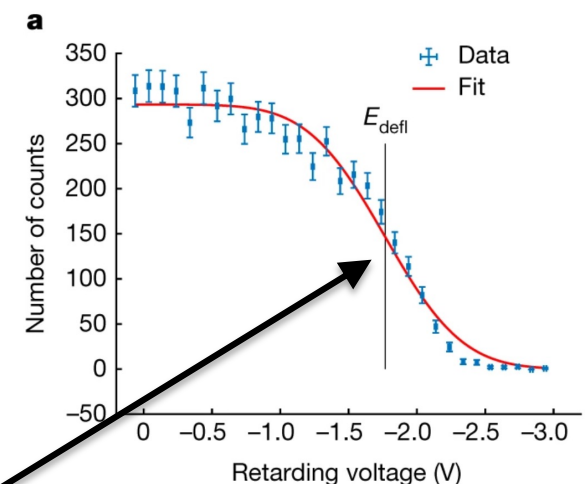


Nature, 573 (7773):243-246 (2019)

^{229}Th measurement

$$\Delta E = E_{\text{defl}} + E_{\text{abe}}$$

atomic binding effects (~7 eV)



This exact instrument is available for use from NIST. We are studying the design carefully to determine if this existing instrument is suitable for ^{229}Pa .

The atomic binding effects had to be calculated accurately to infer the energy for ^{229}Th . In our case, we mostly want to confirm or deny the existence of the state and a 10 eV uncertainty in the energy if observed is more than tolerable.

Molecular Electron EDM Experiments: Large Internal E-field and Control of Systematics

ACME

Neutral ThO* Beam

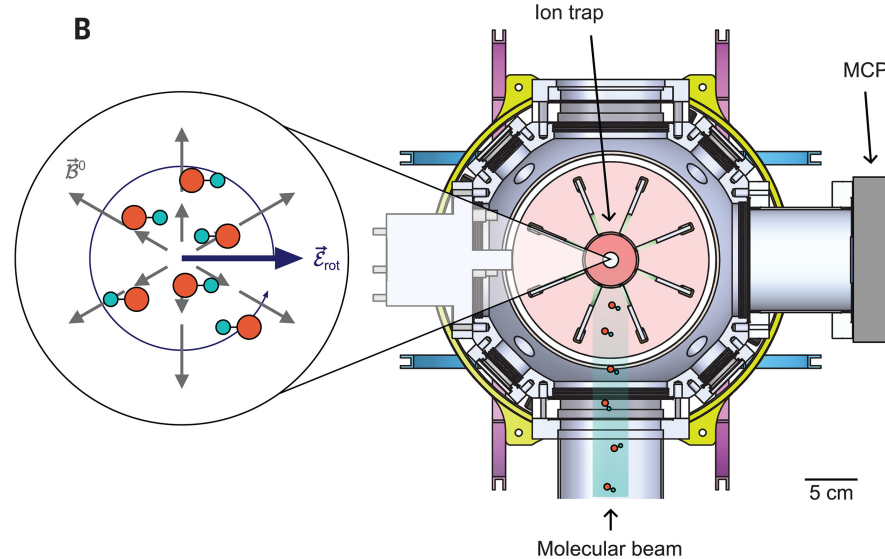
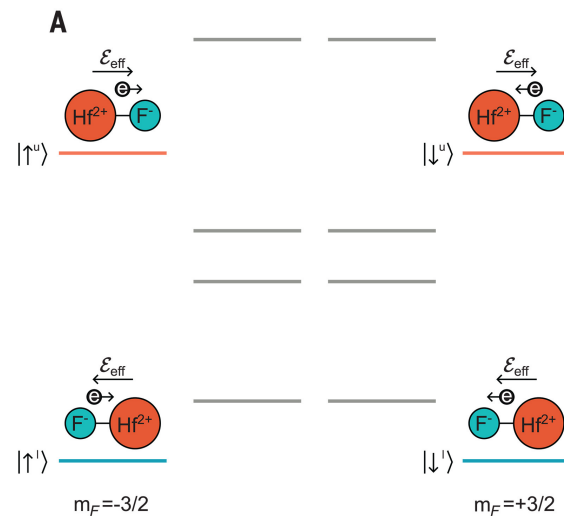
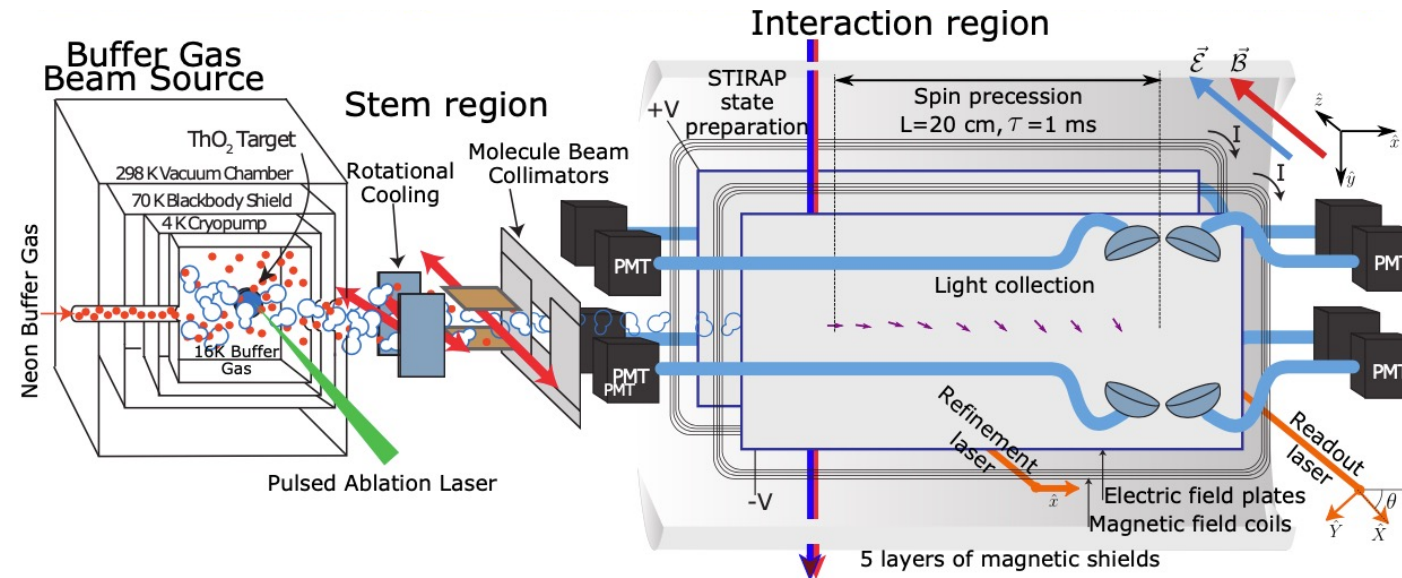
(Chicago /

Harvard /

Northwestern)

C. Panda (Harvard 2018)

Nature 562 355 (2018)

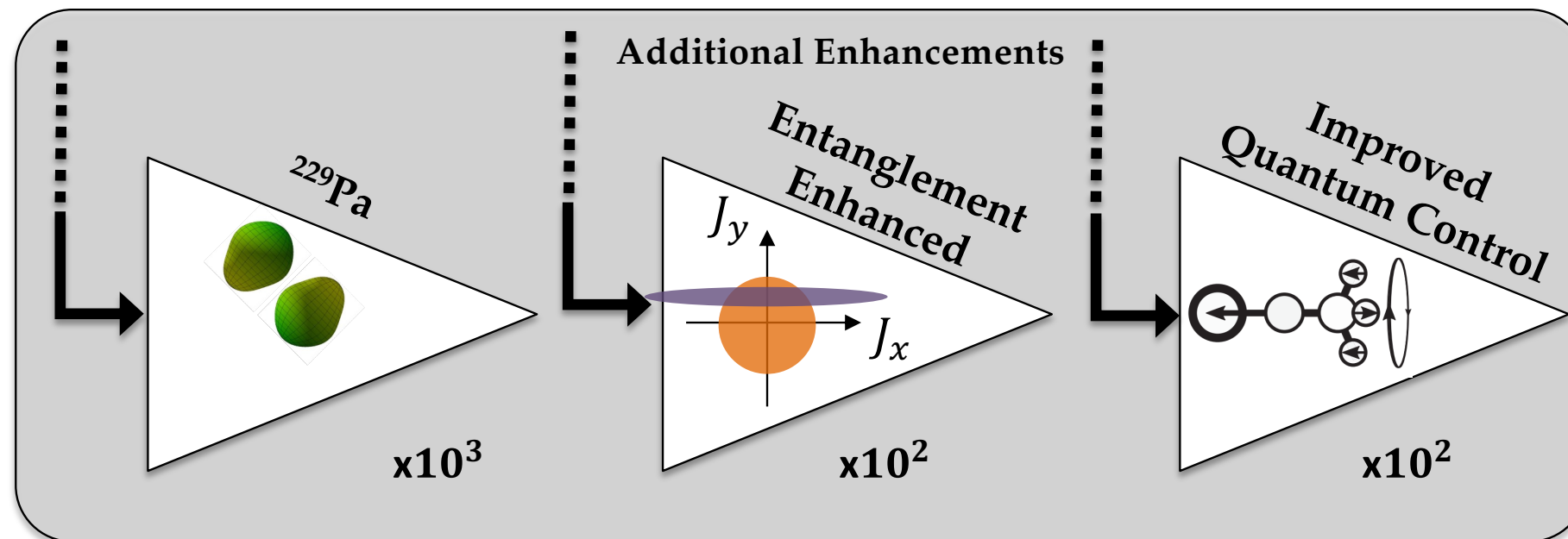
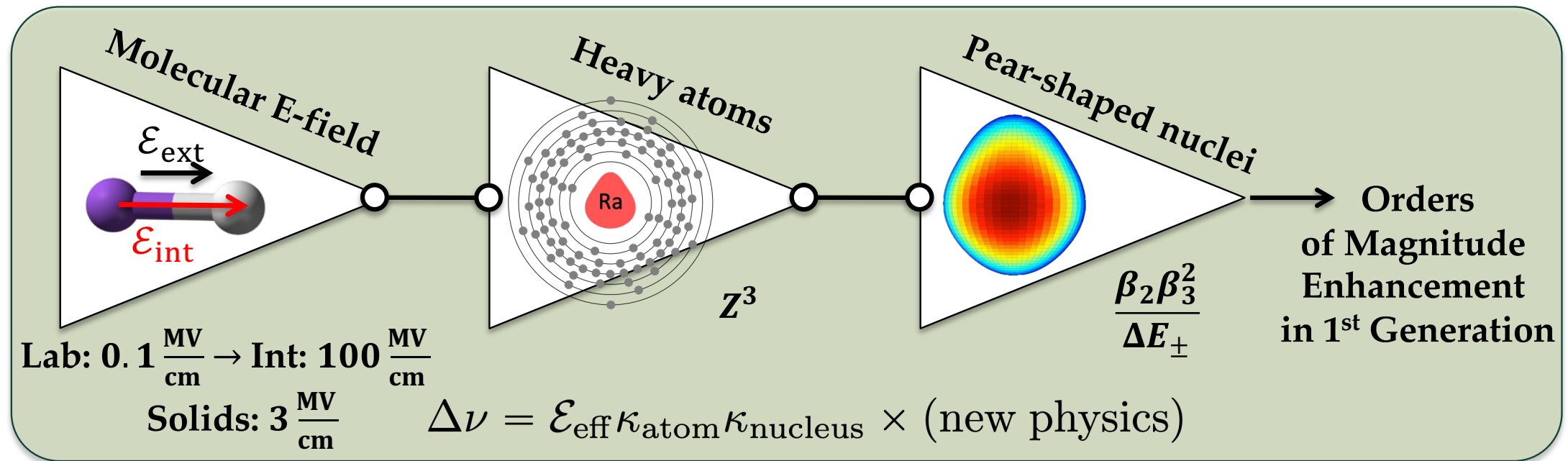


HfF⁺ / ThF⁺
Ion Trap
(JILA)

PRL 119 153001 (2017)

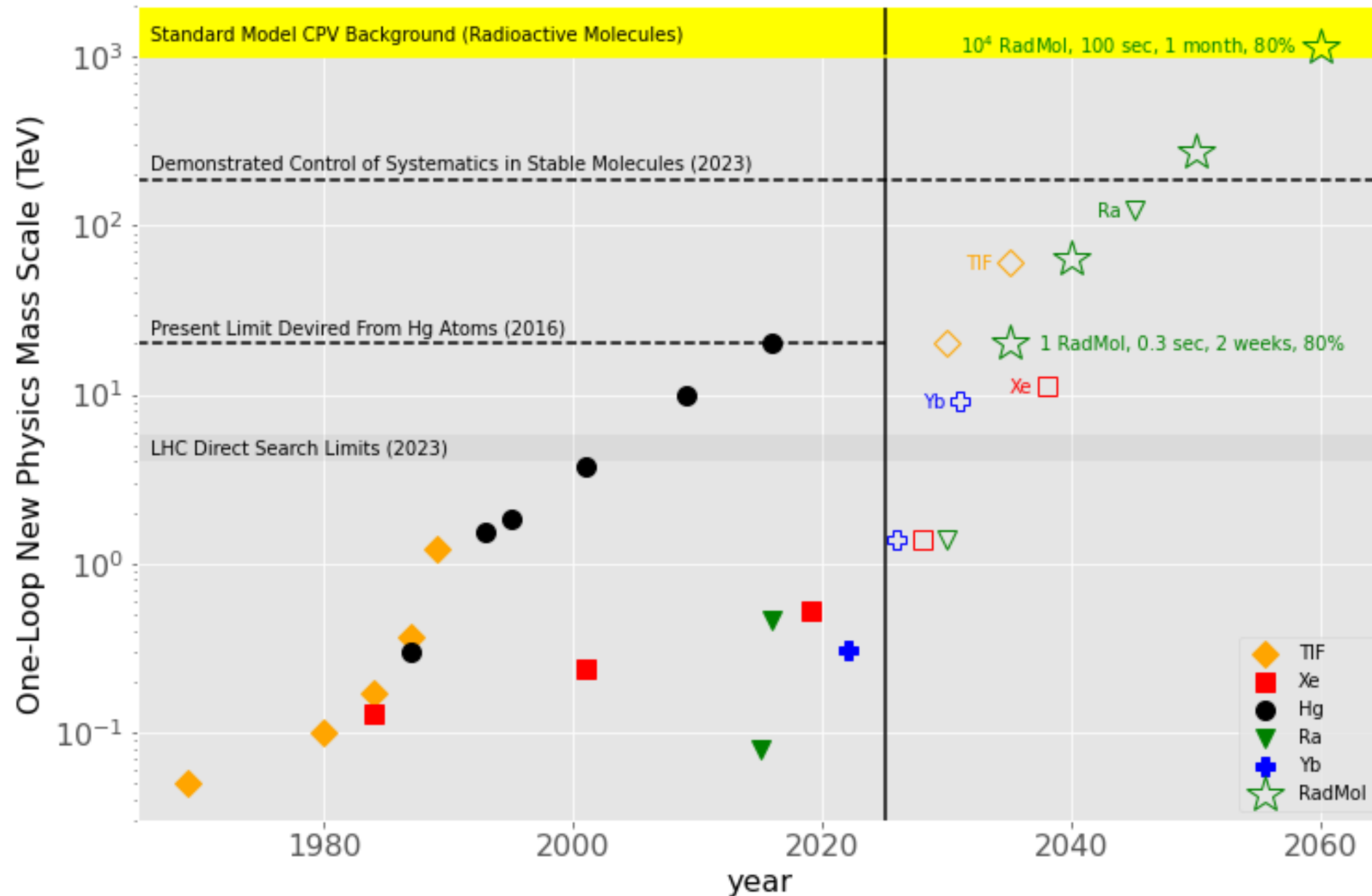
Science 381:46 (2023)

New Laboratory: Trapped Radioactive Molecules Containing Heavy Pear-Shaped Nuclei



Xing Wu
MSU 2023

Radioactive Molecule “EDM” Experiments Have the Potential to Span the Entire New Physics Discovery Window for Hadronic CP -Violation

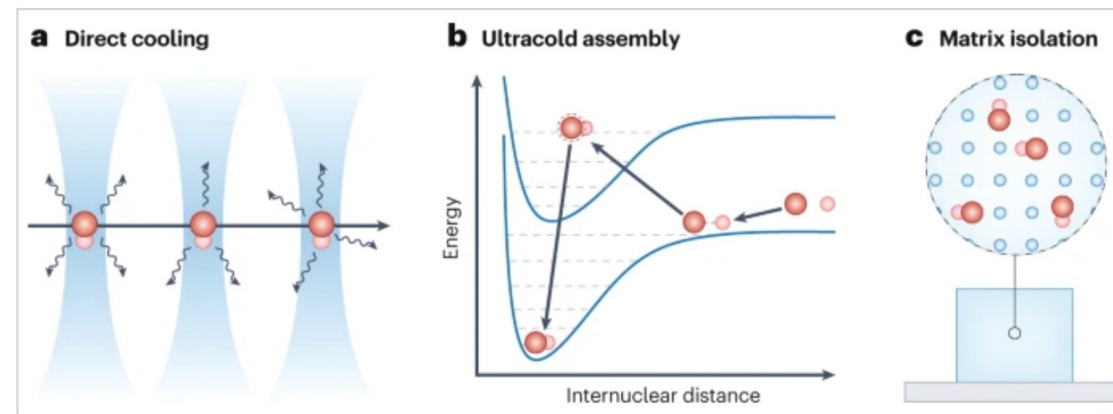


Many Complementary Paths: Different Isotopes + Different Molecules In Laser Traps, Ion Traps, & Solids

Enhancements: nuclear Schiff moment enhancement of $\times 1000$ (^{225}Ra)
to maybe(!?!) $\times 1000000$ (^{229}Pa)
and ~ 100 MV / cm effective internal E -field (lab < 1 MV / cm)
[N.B. the nucleus feels a different E_{eff} than the electrons!]

Potential: $\times 10^5$ to $\times 10^{10}$ more new physics sensitivity than the ^{199}Hg experiment on a per atom basis.

Opportunities for
FRIB, TRIUMF, ISOLDE, ...
[2024 Rep. Prog. Phys. 87 084301](#)



Nature Physics 20, p741–749 (2024)

Challenges:

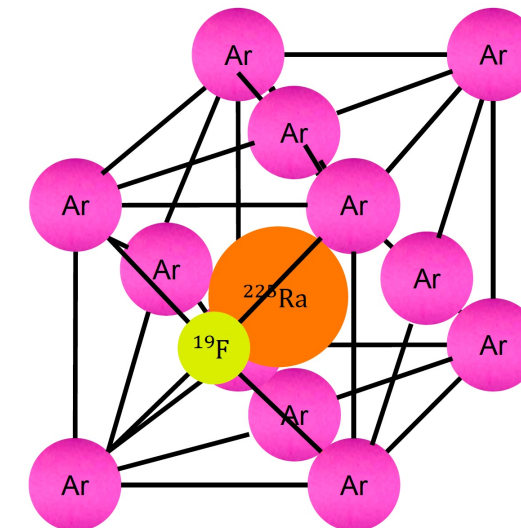
- How do we get the harvested isotopes from “Beaker” into an experiment?
- How do we calibrate the new physics sensitivity of these “enhancer isotopes” inside of molecules?
- How do we efficiently form & probe short-lived radioactive molecules?

Pear-Shaped Nuclei Implanted In Cryogenic Solids:

^{225}RaF ($\tau_{1/2} = 15$ days) & ^{229}Pa ($\tau_{1/2} = 1.5$ days)

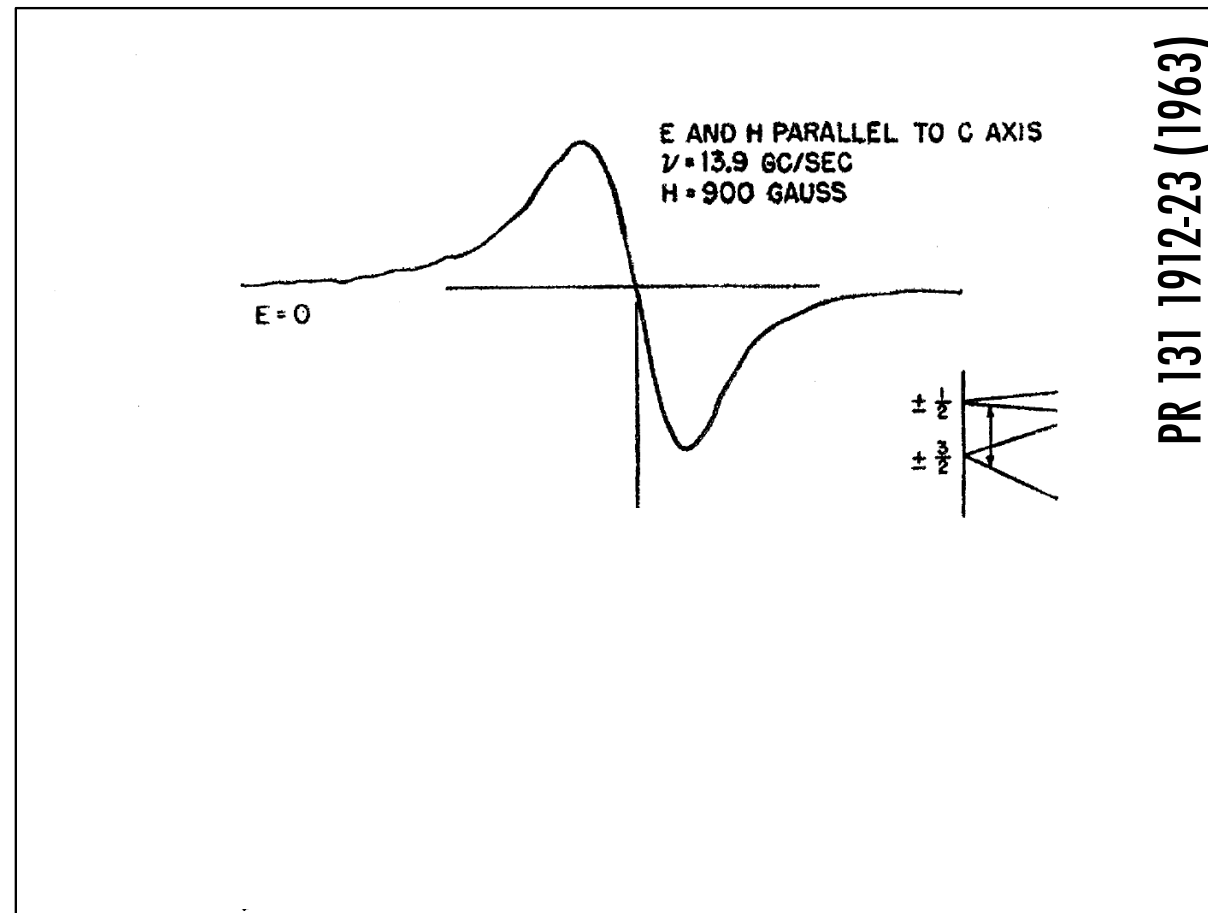
Inspired by EDM³ **Eric Hessels (York)** and **Amar Vutha (Toronto)**

- **Efficient trapping of a wide variety of species**
- **Very high number densities**
- Stable and chemically inert confinement
- Transparent in the optical regime for optical probing
- Under certain conditions, polar molecules orient themselves along the crystal axes which allows for control of systematics: **PRA 98:032513 (2018)**
- **Challenge: quantum control in rare gas solids**
- Ions implanted in optical crystals or diamonds allowing for optically-addressable nuclear spins
Hyp. Int. 240:29 (2019), Phil. Trans. R. Soc. A.382: 20230169 (2024), PRA 108, 012819 (2023)
- Implanted ions can sit at two distinct sites with opposite pointing internal E-fields which allows for control of systematics **PR 131 1912 (1963)**



Controlling Systematics in Solids: The RB-Experiment

Cr^{3+} in Al_2O_3 : Royce & Bloembergen Phys. Rev. 131 1912 (1963)

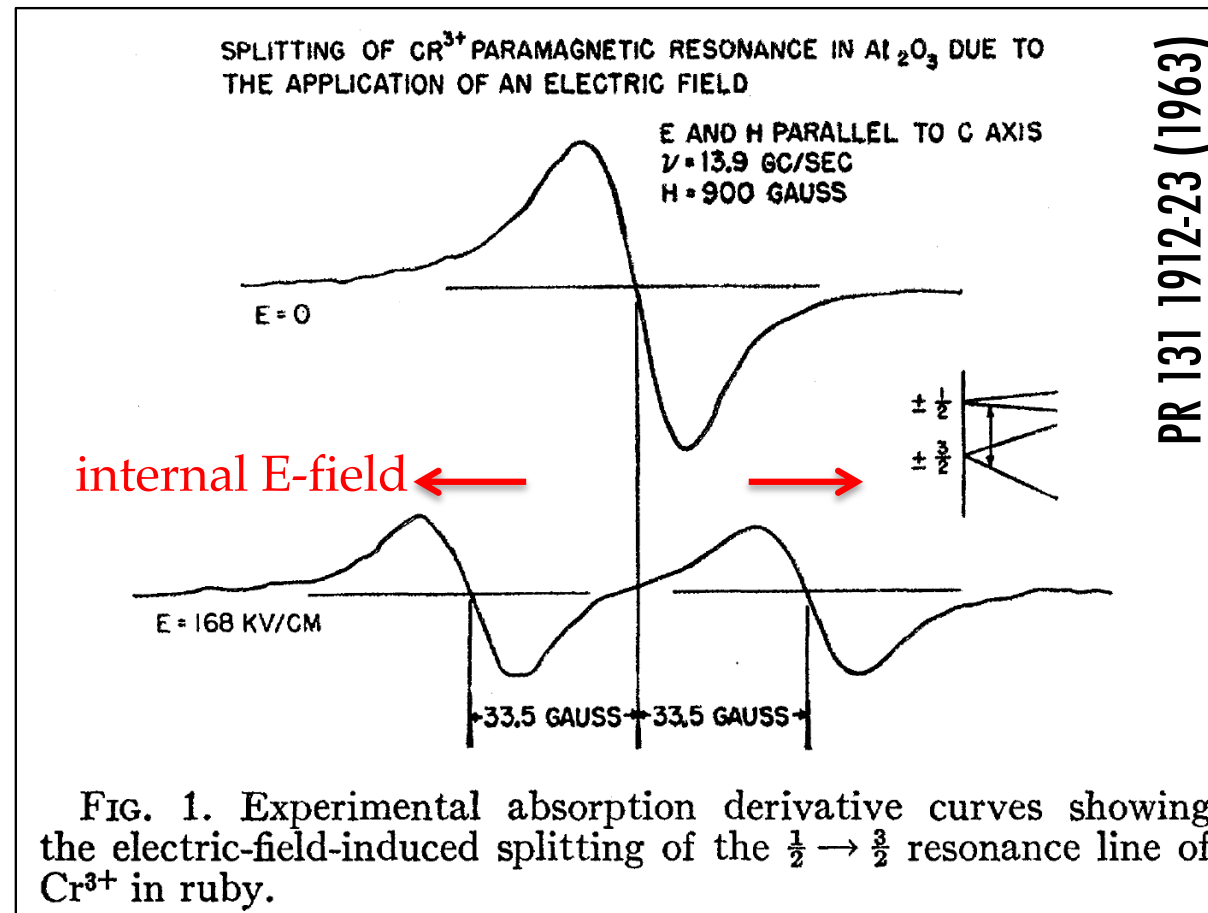


→ external B-field

Key Concept: In the absence of external fields, the non-degeneracy of a Kramers Doublet is an indication of time-reversal symmetry violation.

“Stark Splitting:” There Are Two Ensembles of Ions!

Cr³⁺ in Al₂O₃: Royce & Bloembergen Phys. Rev. 131 1912 (1963)

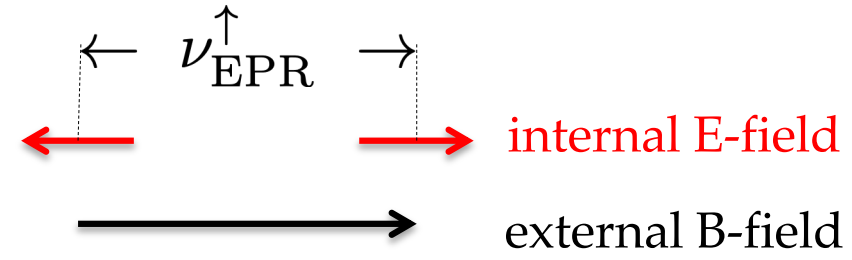
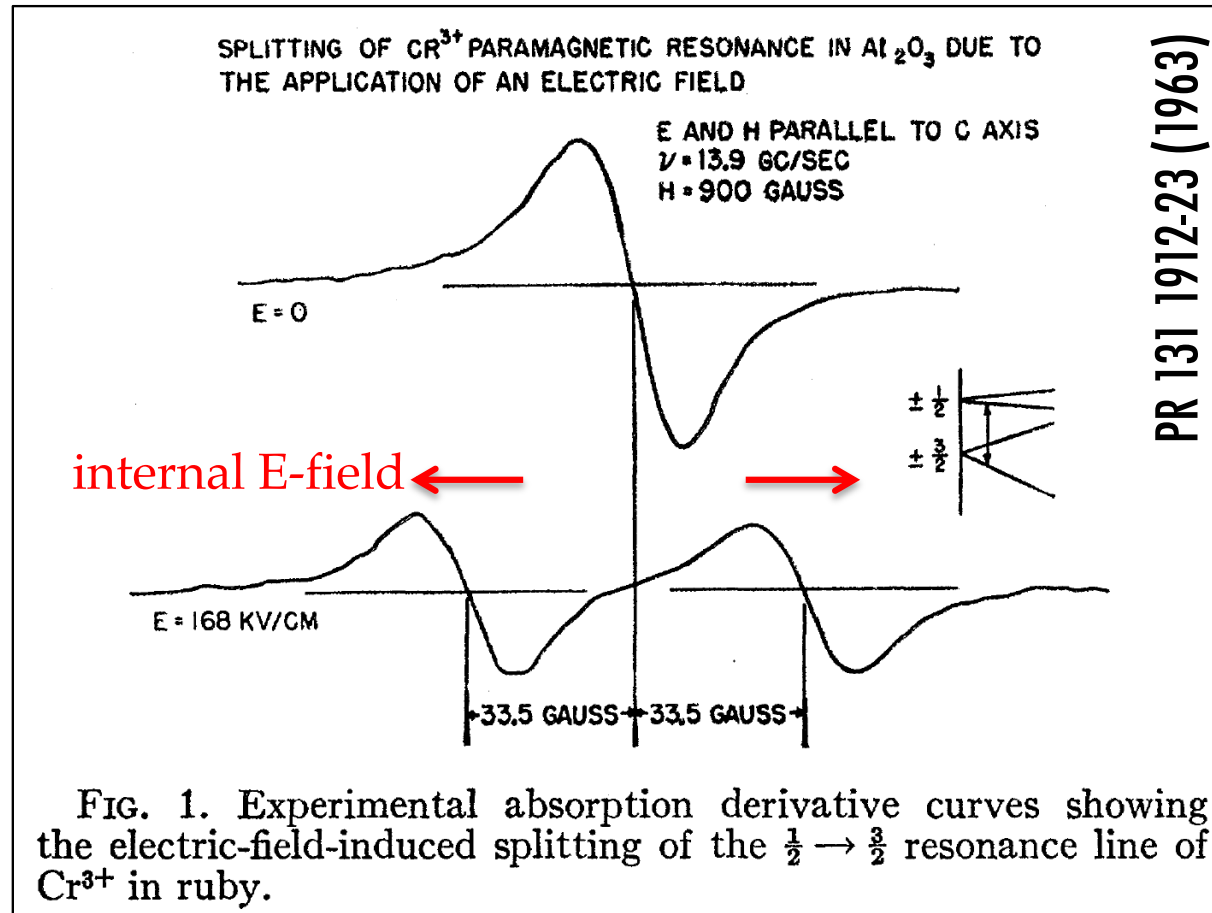


→ external B-field

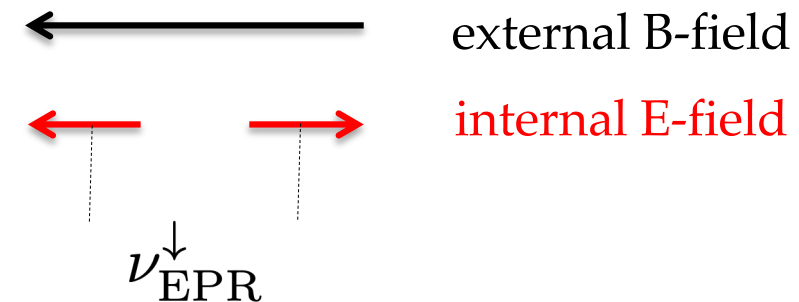
Key Concept: Under the right conditions, ions (& molecules...) in solids are oriented along the crystal axes.

Apply E-Field Only During State Detection!

Cr³⁺ in Al₂O₃: Royce & Bloembergen Phys. Rev. 131 1912 (1963)

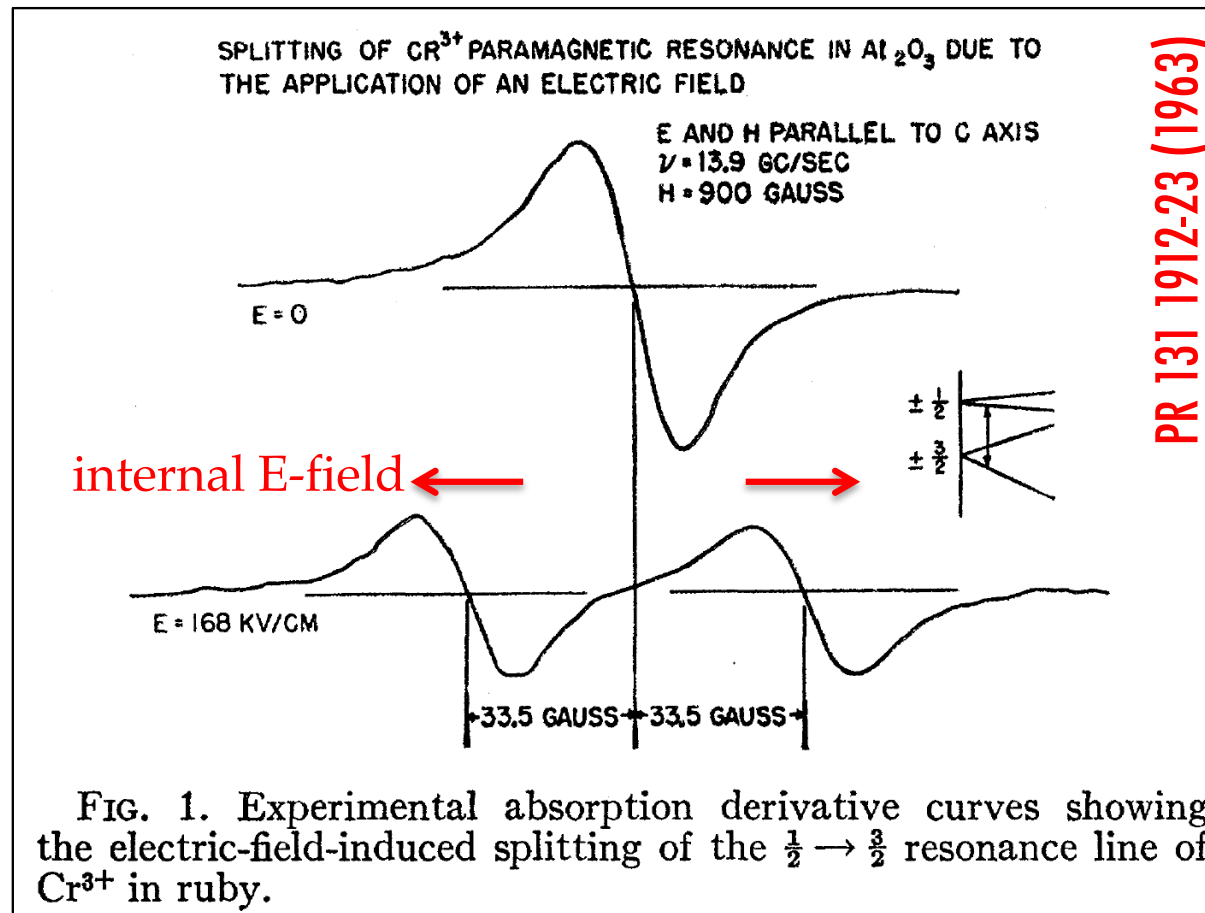


$$\Delta\nu_{\text{EPR}} = \frac{4\Delta mdE}{J}$$

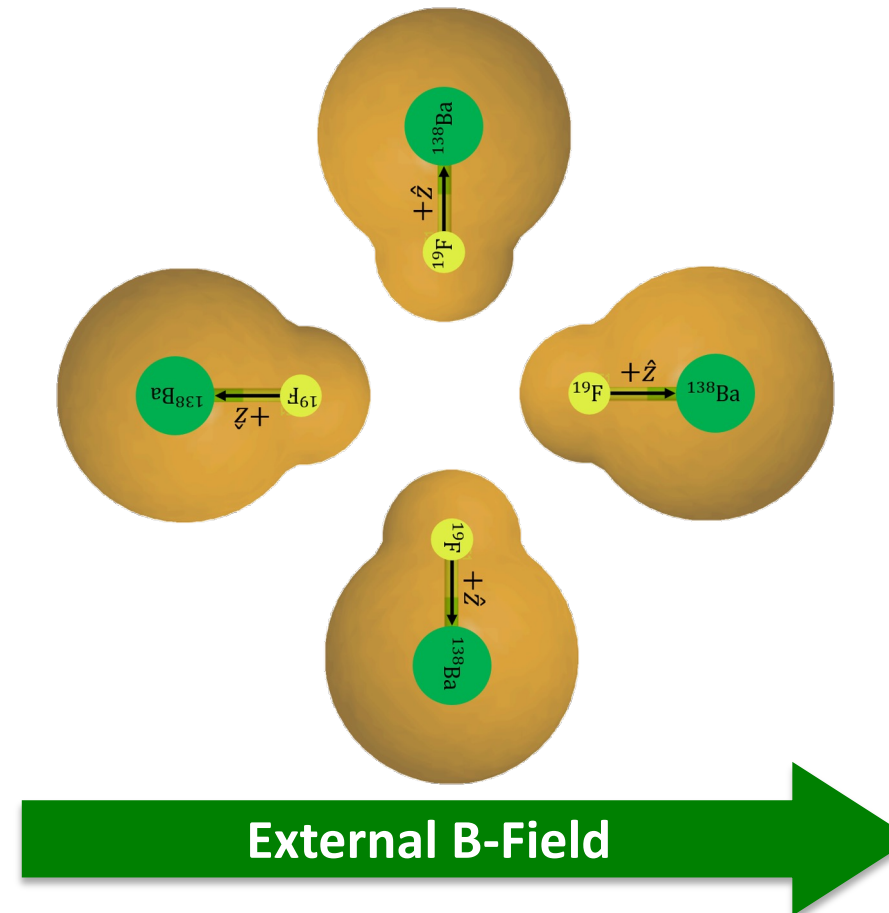


Key Concept: Apply a lab E-field only during the detection/readout step just to spectroscopically distinguish the two ensembles!

Key Idea to Control Systematics in Solids: Two Nearby Sites Where Effective E-fields Are Opposed



PR 131 1912-23 (1963)



PRA 98:032513 (2018)

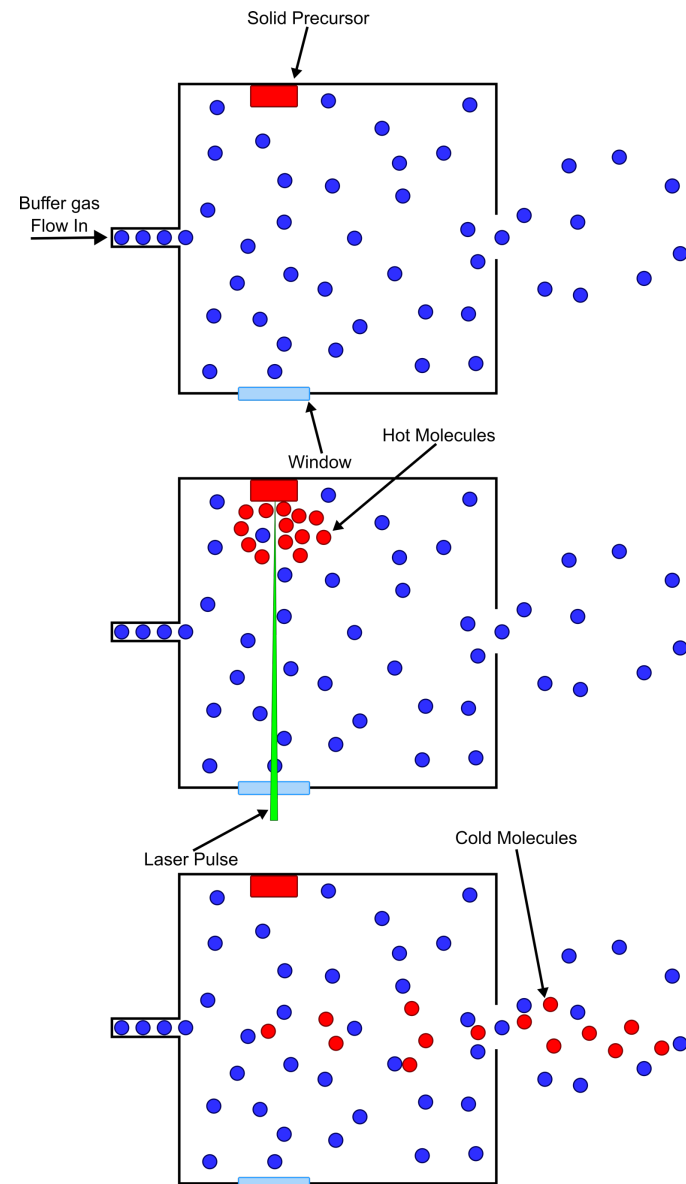
BaF/RaF orientations
(in/out not depicted here)

external B-field

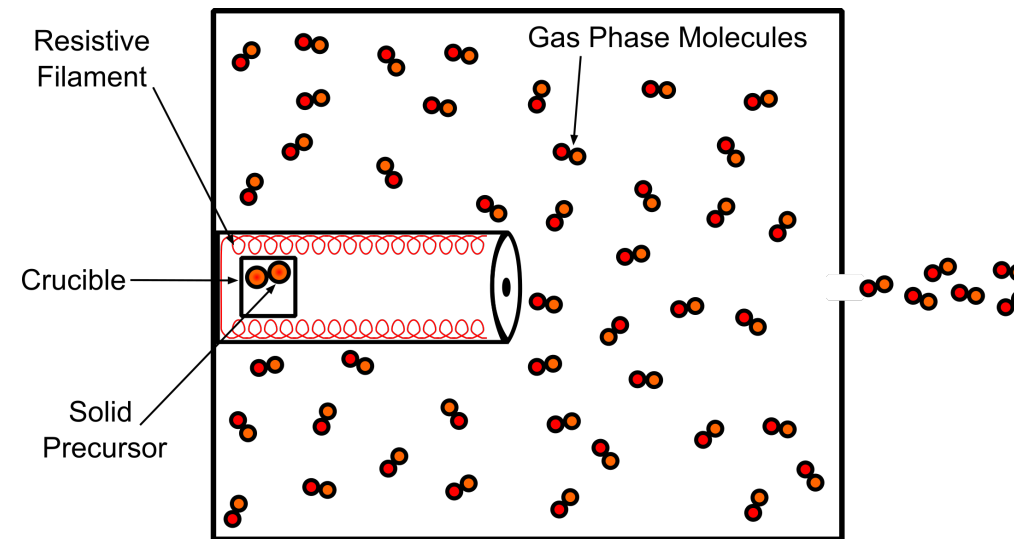
Key Concept: No lab E -field during the spin precession step minimizes the sensitivity of the frequency measurement to external E -fields.

Challenge: How Can We *Efficiently* Make The Molecules of Interest?

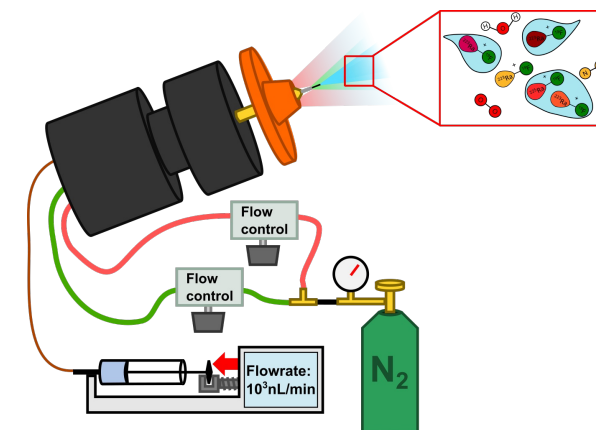
Less Brute Force:
Laser Ablation + Cold Buffer Gas



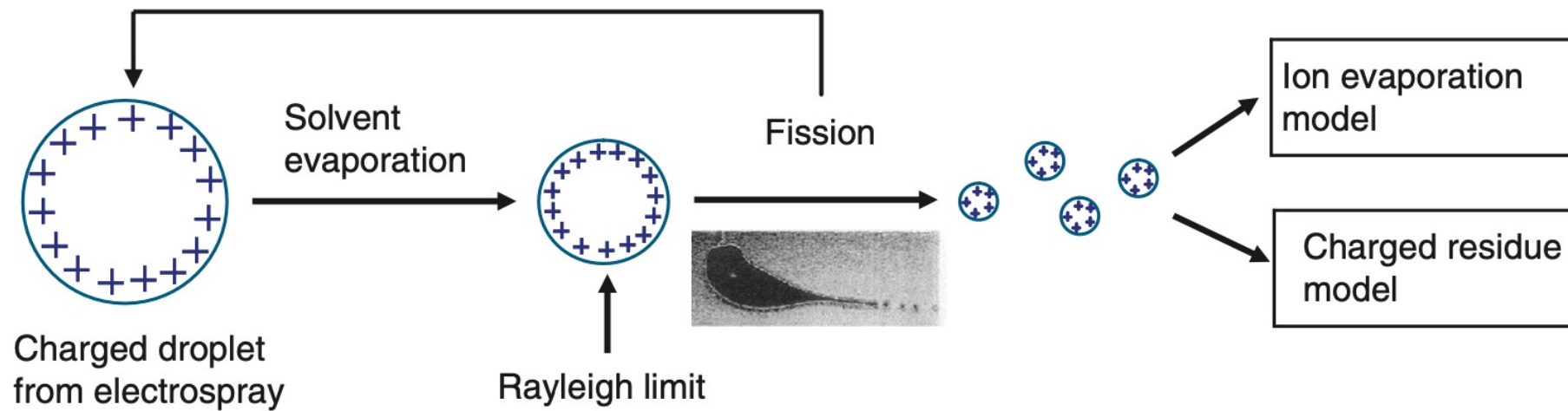
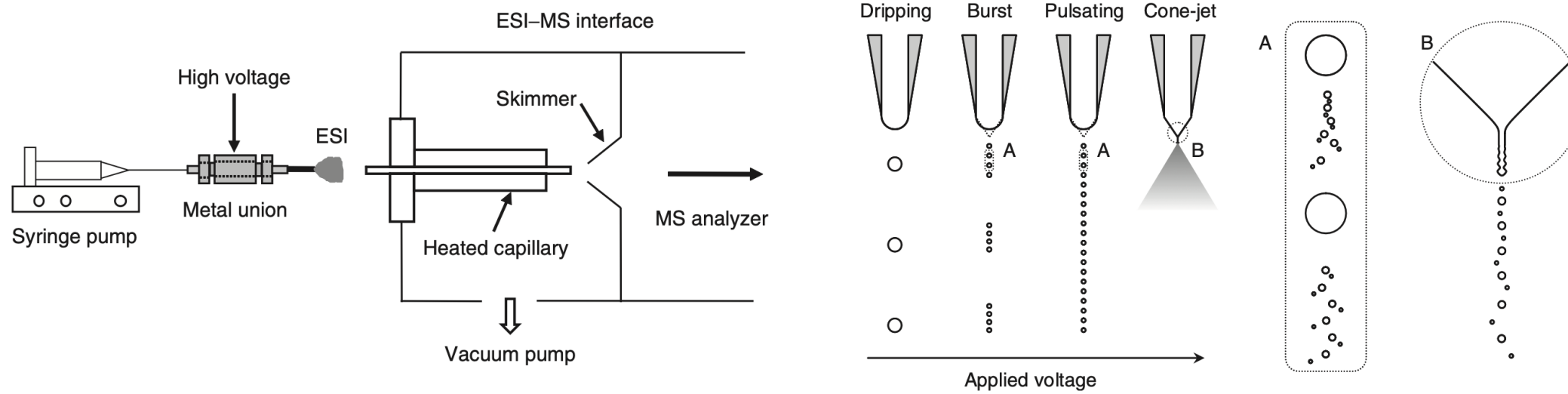
Most Brute Force:
High Temperature Oven



Crazy Enough to Work:
Electrospray Ionization



Electrospray Ionization Basics



Encyclopedia of Spectroscopy and Spectrometry, Third Edition

<http://dx.doi.org/10.1016/B978-0-12-803224-4.00319-8>

Nobel Prize Chemistry 2002: John B. Fenn

Electrospray Ionization Could Be Very Efficient!

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

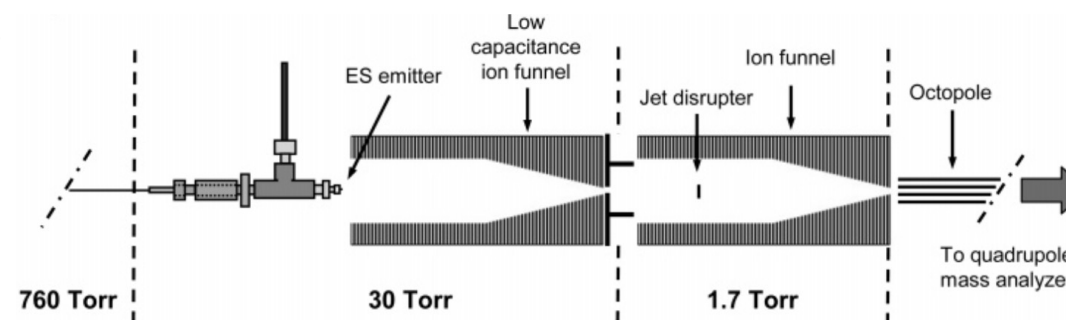
Inefficient ionization and poor transmission of the charged species produced by an electrospray from the ambient pressure mass spectrometer source into the high vacuum region required for mass analysis significantly limits achievable sensitivity. Here, we present evidence that, when operated at flow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

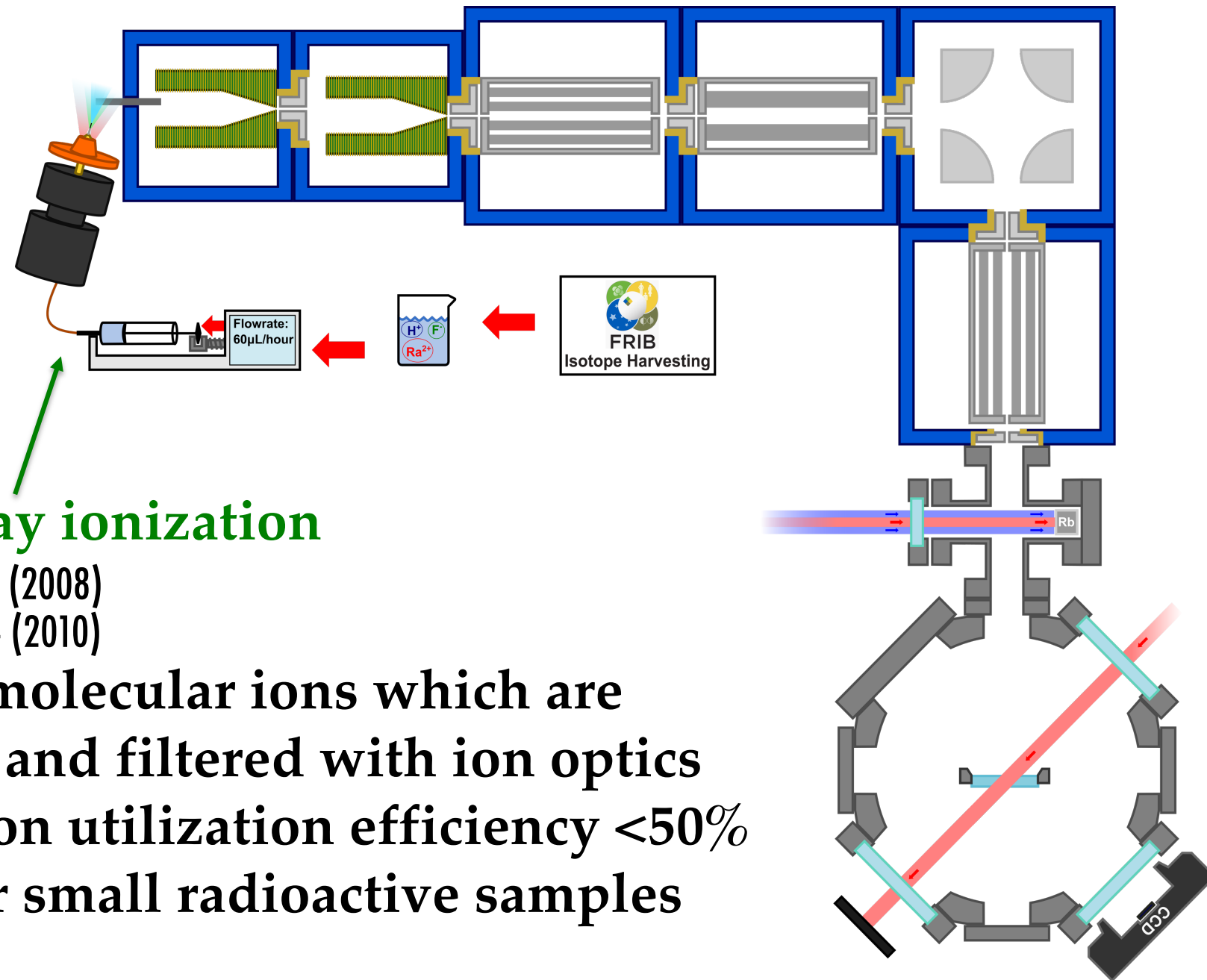
Subambient Pressure Ionization with Nanoelectrospray Source and Interface for Improved Sensitivity in Mass Spectrometry

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



FRIB-EDM3: ^{225}RaF in Noble Gas Solids



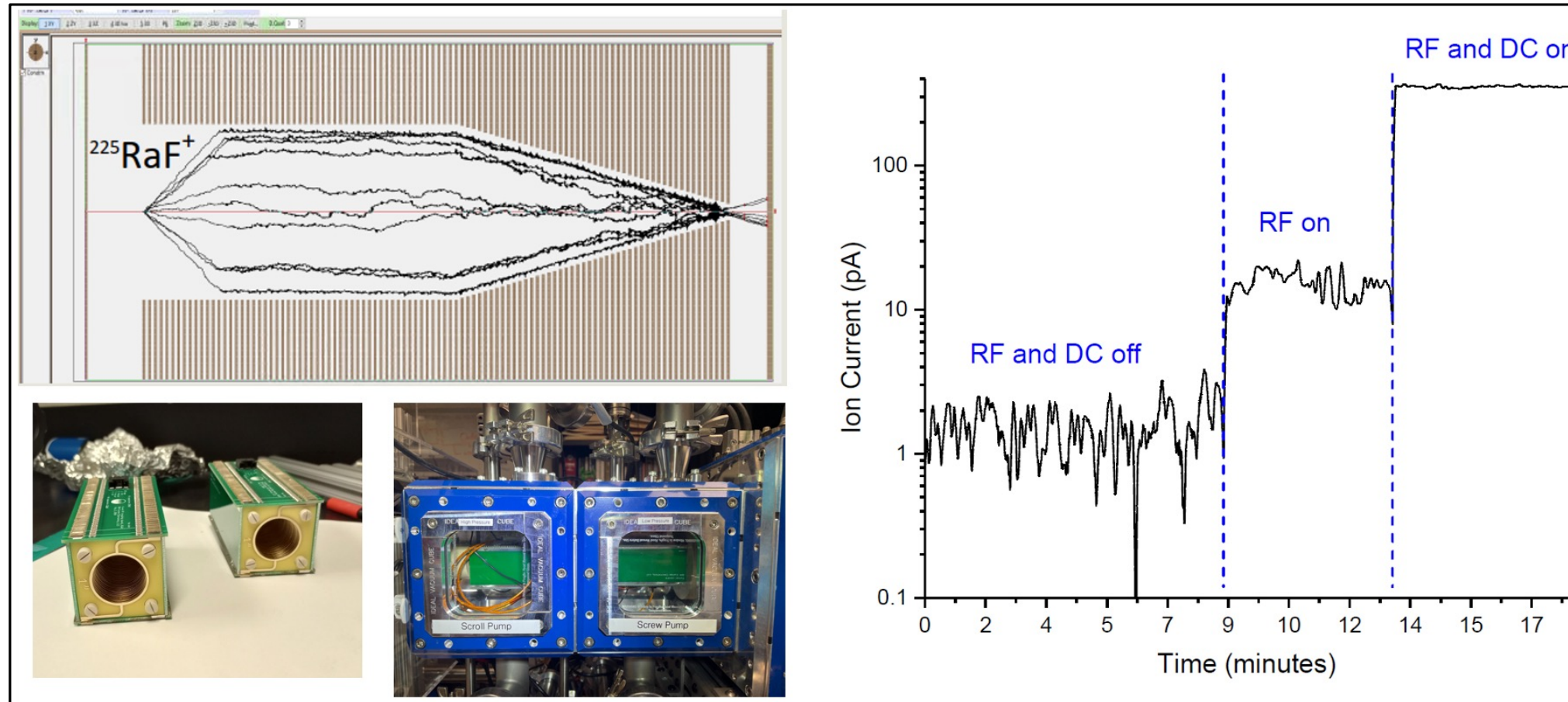
Electrospray ionization

Anal. Chem.80:1800 (2008)

Anal. Chem.82:9344 (2010)

- creates molecular ions which are focused and filtered with ion optics
- ionization utilization efficiency <50%
- ideal for small radioactive samples

Prototype: Testing With Isopropanol: Ion Funnel Work Well! (~\$5k off the shelf)



Stable ion beams are possible:

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

Challenges:

- Low reproducibility
- Lack of control over positioning and probe alignment

FRIB-EDM3 v2.0: Design and Procurement Complete Assembly Underway Now, Testing This Summer!

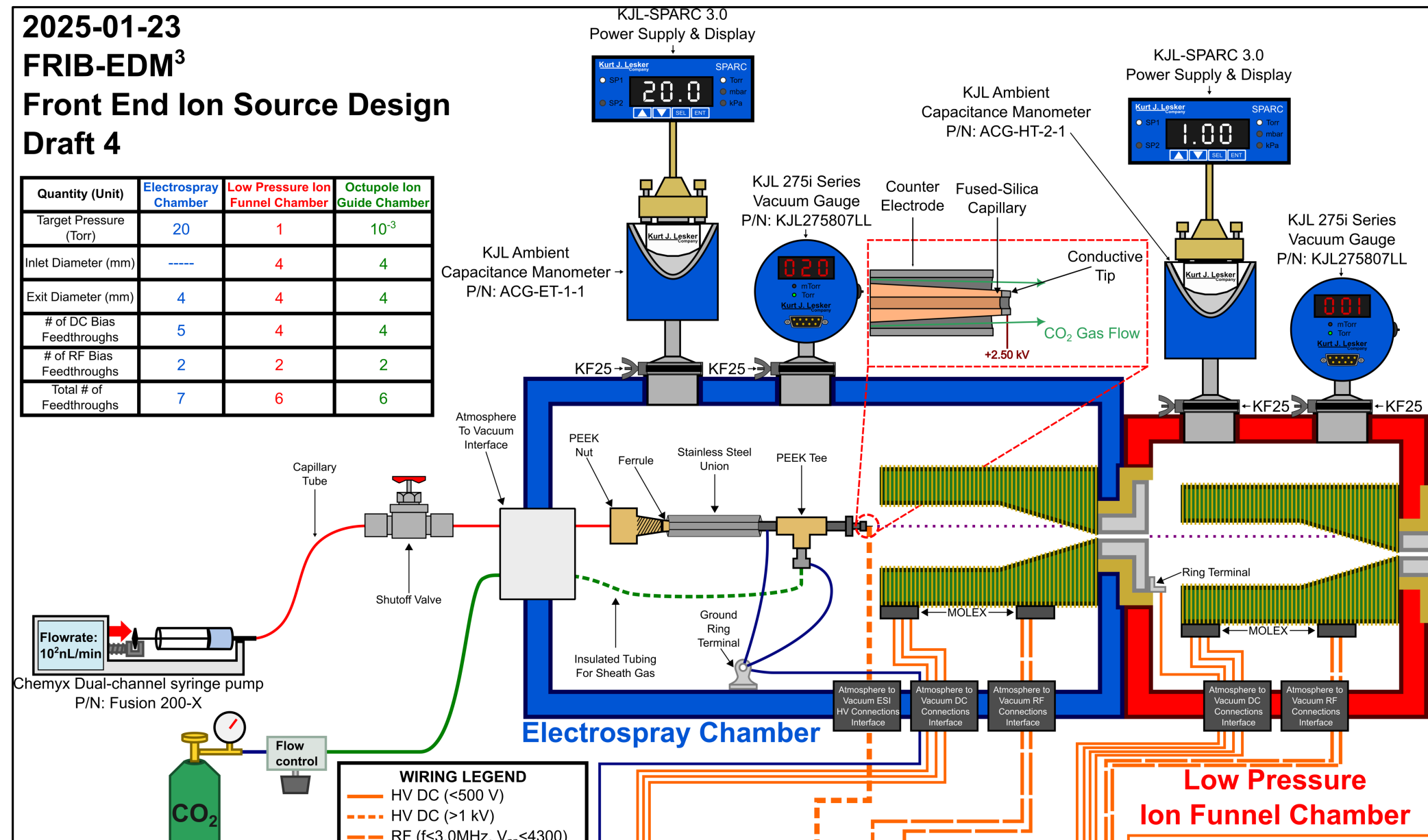
2025-01-23

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



Alternative Approach: Atomic Ions In Crystals

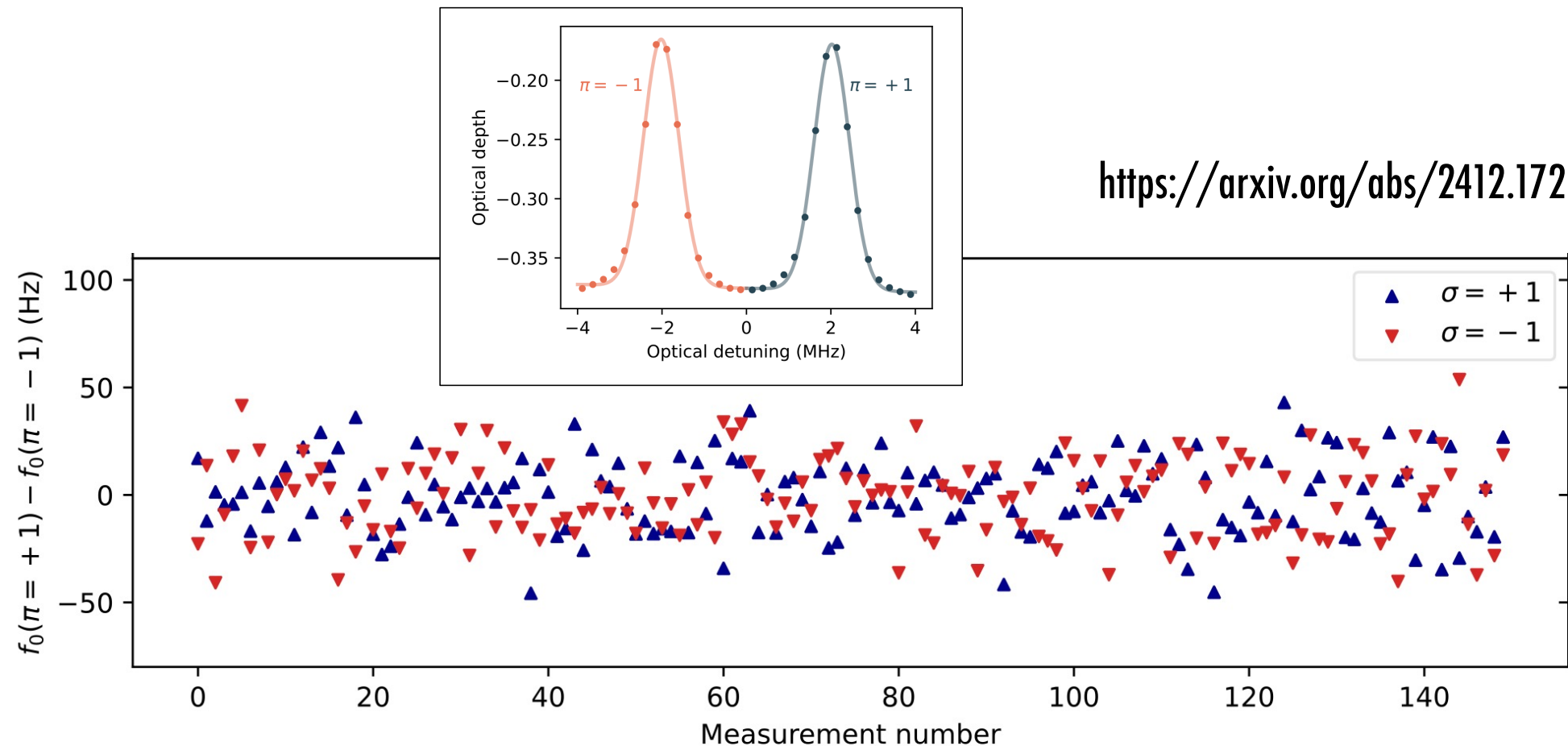
Another Example of a Modern “RB-Experiment”

Comagnetometry using mirror-symmetric ions in a crystal

Bassam Nima,¹ Mingyu Fan,¹ Aleksandar Radak,¹ Andrew Jayich,² and Amar Vutha¹

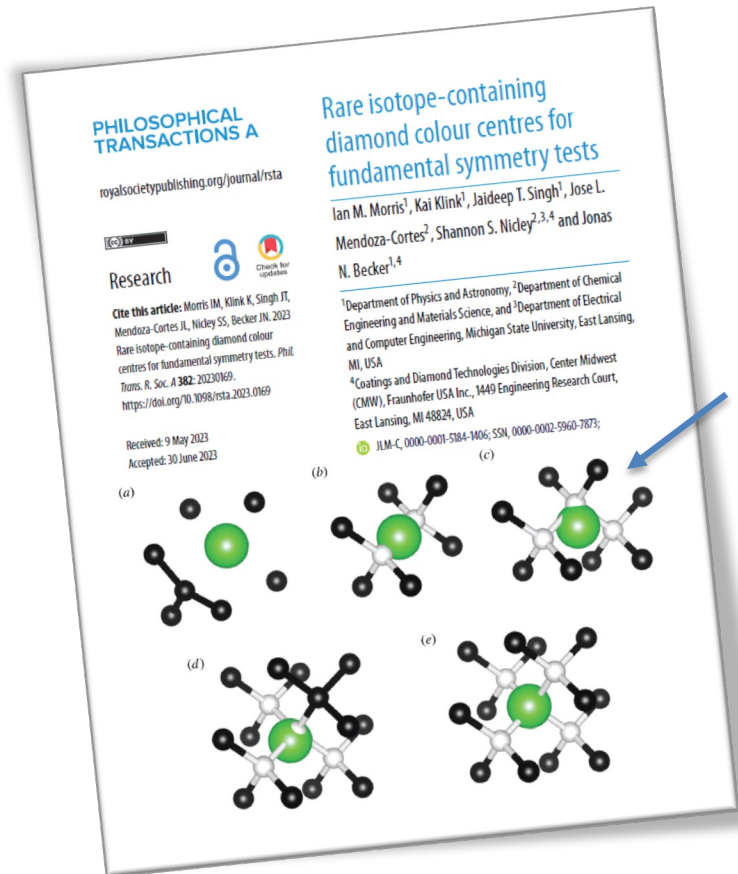
¹*Department of Physics, University of Toronto, Toronto ON M5S 1A7, Canada*

²*Department of Physics, University of California - Santa Barbara, Santa Barbara CA 93106, USA*



<https://arxiv.org/abs/2412.17276>

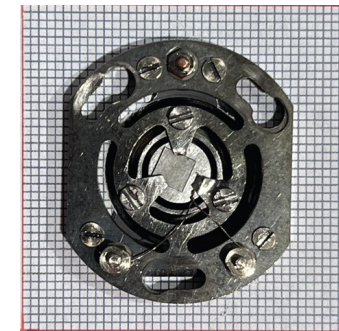
Prepare & Study Rare Isotopes in Diamonds



Pa-231 Doped
Diamond

The defect we
are interested in

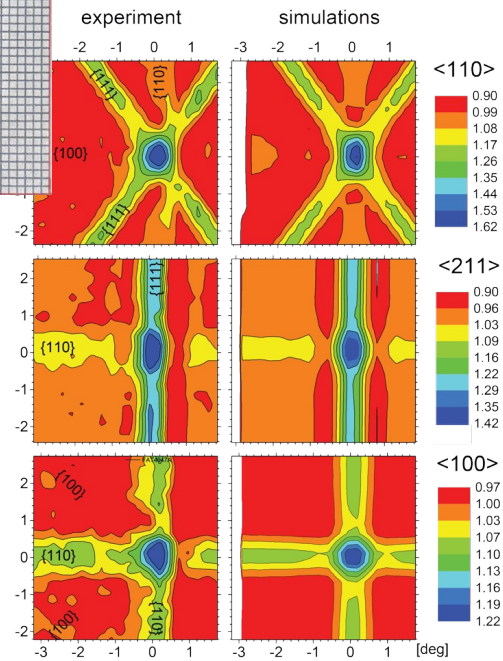
One
Inch



Does it exist?

NOW: First ion implantations and emission channeling experiments at CERN /Switzerland **motivated by our paper** (collaborator: Lino da Costa Pereira, KU Leuven Belgium and CERN)

Yes!



2023: Team theoretically predicted existence of several rare isotope defects in diamond, **unfunded student class project**

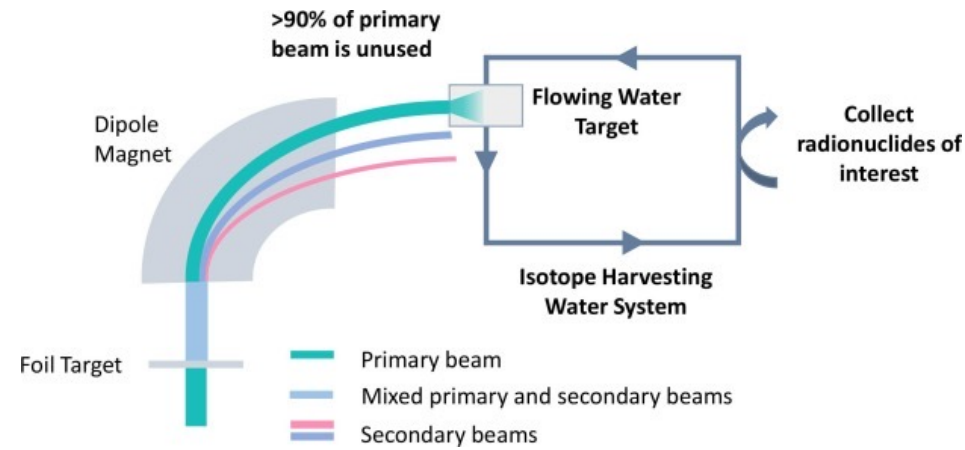
MSU Team:
 Defects: Jonas Becker
 Diamonds: Shannon Nicley
 Implantation: Paul Gueye
 Radiochemistry: Alyssa Gaiser
 DFT: Jose Mendosa
 PER: Daryl McPadden

The Nuclear Pear Factory: A Proposed Center

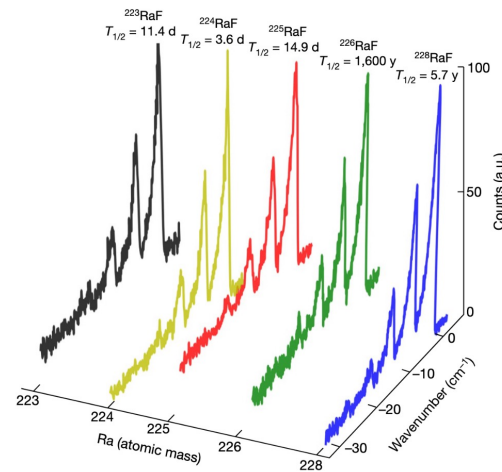
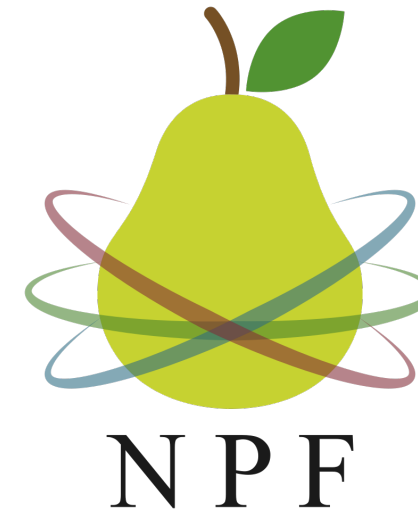


Nature 497:199 (2013)

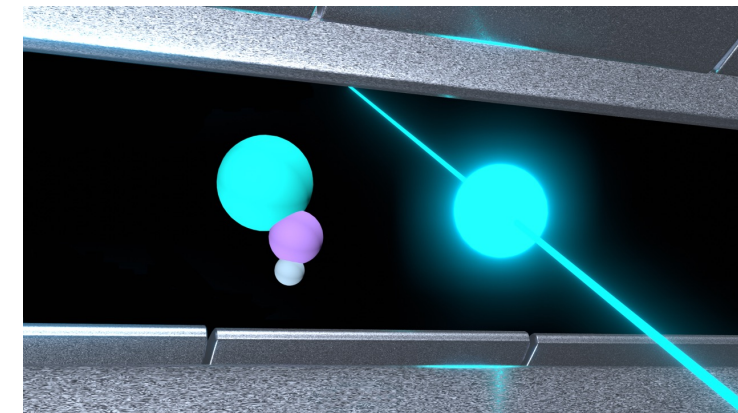
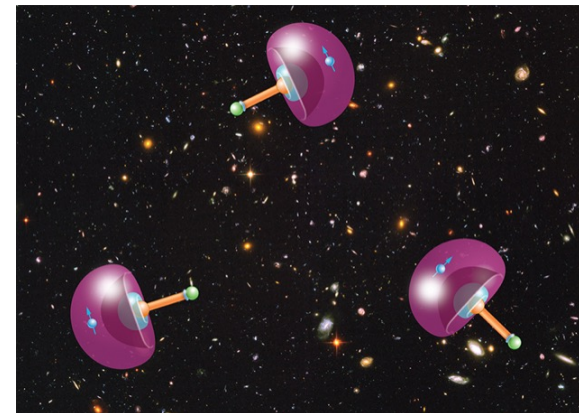
A joint Experiment/Theory & AMO/Nuclear/Radiochemistry effort to calibrate the new physics sensitivity of pear-shaped nuclei and to carry out the requisite precursory work leading to ultrasensitive EDM searches.



NIMB 478 34 (2020)



Nature 581:396 (2020)



<https://physics.aps.org/articles/v14/103> & A.M. Jayich

Complementarity: EDMs are the Elephant Butt of New Physics Searches.

LHC Direct Searches

$$\sigma_{\text{LHC}} \propto |C_{ij}|^2$$

Precision Muon Measurements

Neutrino Oscillations

modified from
Stéphanie Rocca's
original concept



Particle Astrophysics

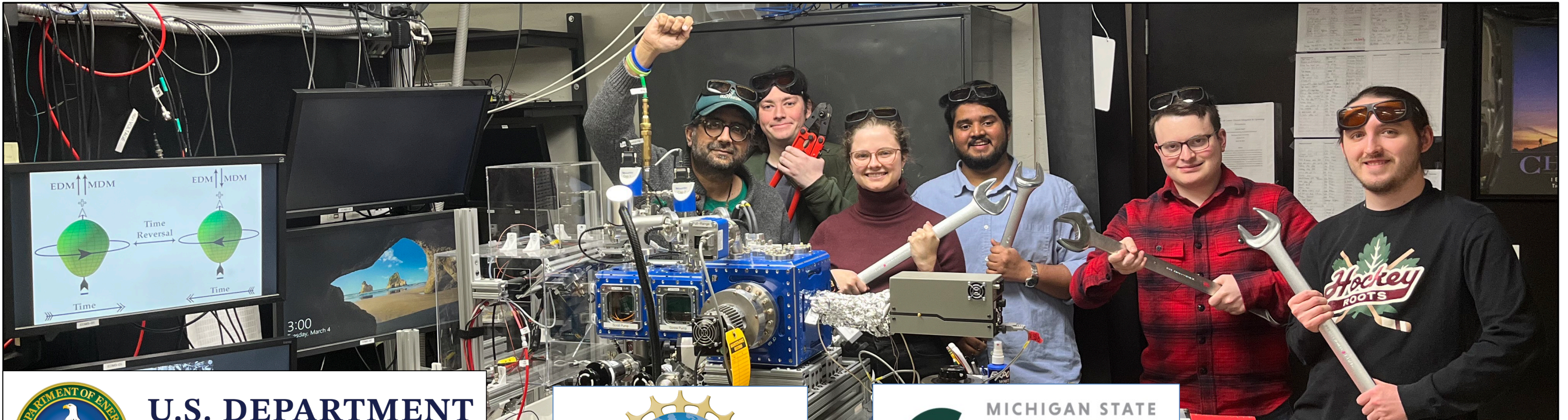
EDMs

$$d_{\text{edm}} \propto \text{Im } C_{ij}$$

Neutrinoless Double
Beta Decay

Thanks For Your Attention!

photo: S. Grant



U.S. DEPARTMENT
of ENERGY

DE-SC0019015 (ECA-EDM3)
DE-SC0019455 (Ra EDM)
DE-NA0003996 (Pa-229)
DE-SC0025679 (Ra EDM+EDM3)



1654610
(CAREER-SAM)
2412951 (SAM)



MICHIGAN STATE
UNIVERSITY
RESEARCH
FOUNDATION

SPG: Center
Development (NPF)

PCLB (Pa-229)

GORDON AND BETTY
MOORE
FOUNDATION

2025 Experimental
Physics Investigator
GBMF13799 (Pa-229)