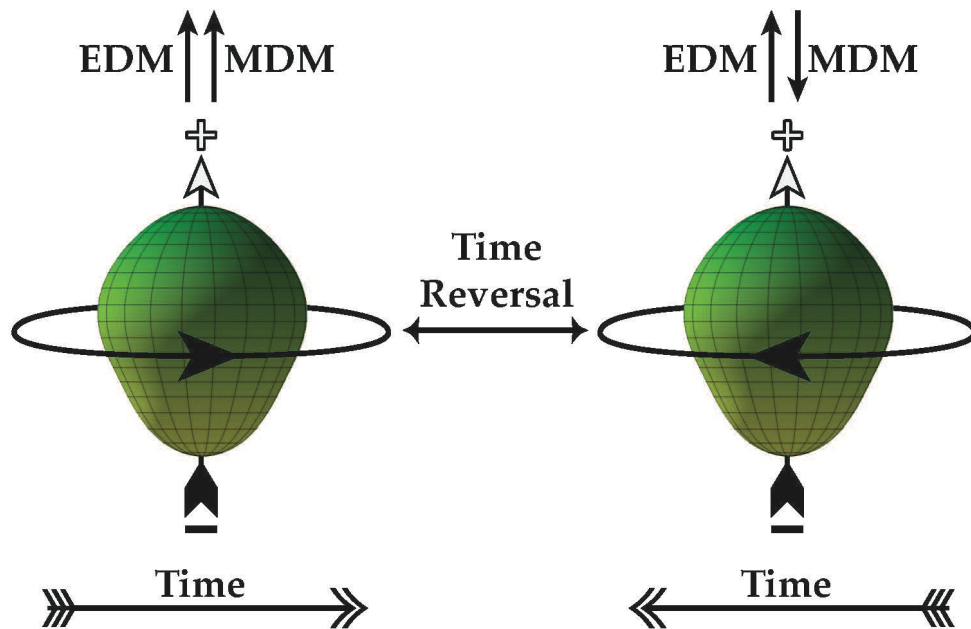


# Prospects for Determining If $^{229}\text{Pa}$ Is the Most Pear-Shaped Nucleus



Lise  
Meitner



Jaideep Taggart Singh (he/him/his)

Michigan State University / FRIB

CNRS-MSU IRL-NPA Mini-Symposium on  
Precision Measurements at Low Energies

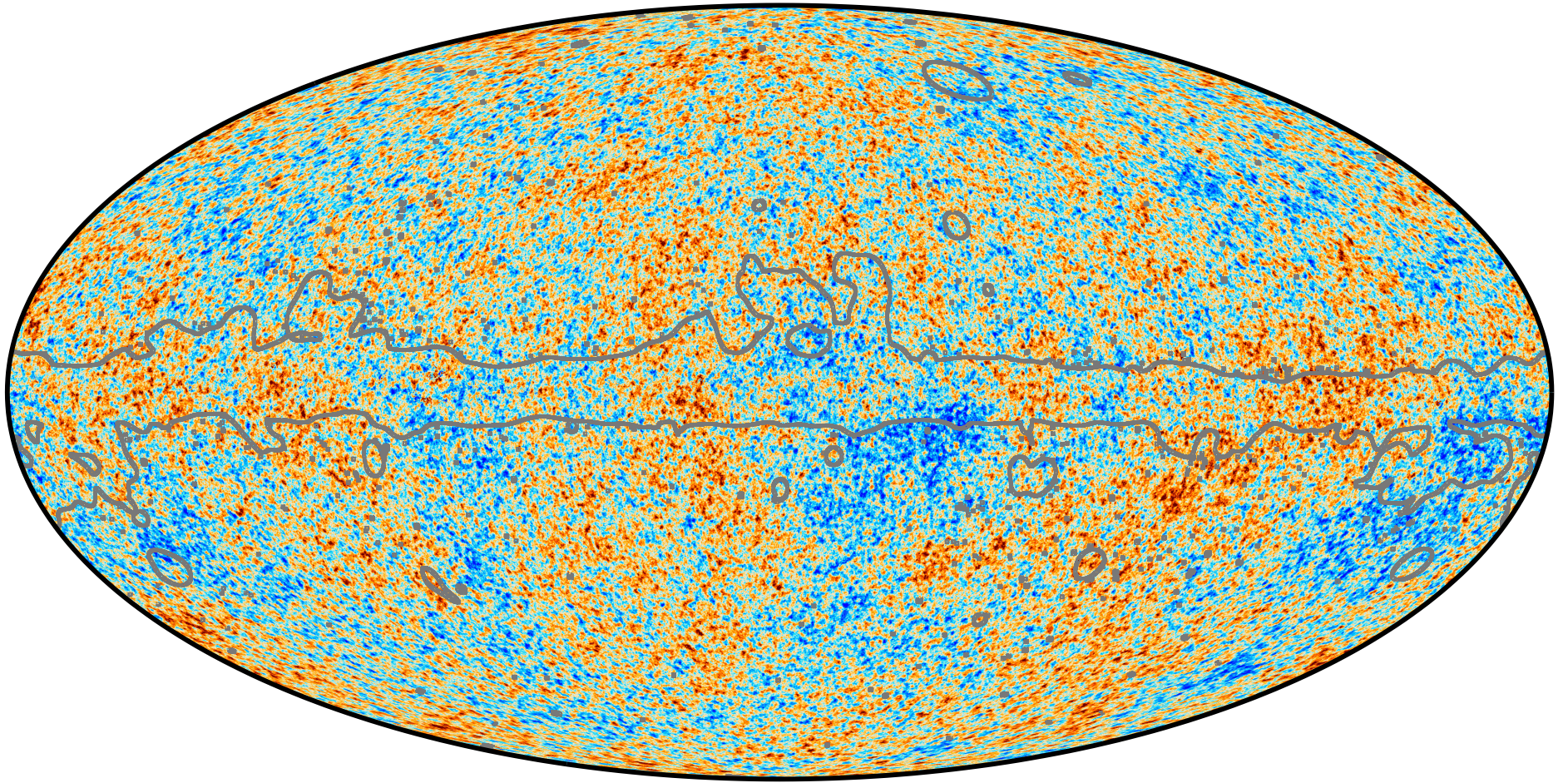
FRIB Lecture Hall 1200 – 13:30-14:10 March 15, 2025



SCAN ME



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



Planck 2018

<https://www.cosmos.esa.int/web/planck/picture-gallery>

-300



300  $\mu\text{K}$

~10 ppm  
fluctuations  
on top of 2.726 K



# Sakharov's Conditions: Need CP-Violation



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

A. D. Sakharov

Submitted 23 September 1966

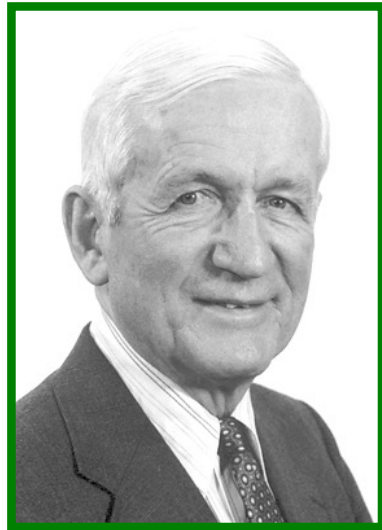
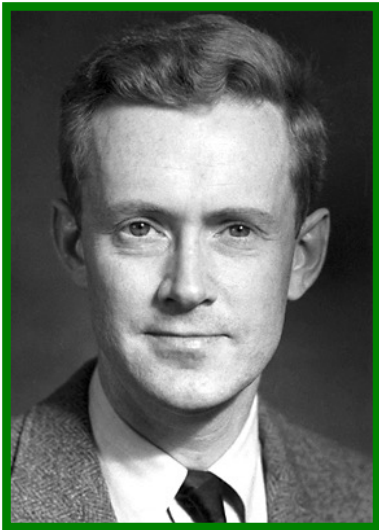
ZhETF Pis'ma 5, No. 1, 32-35, 1 January 1967

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.

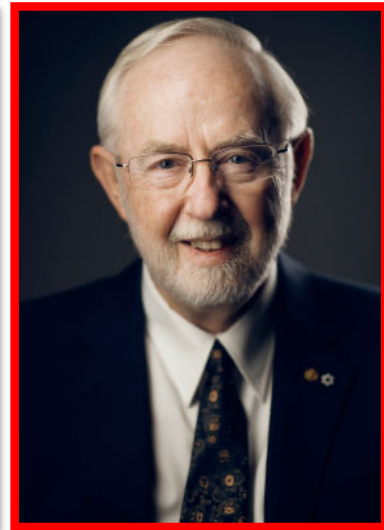
The Nobel Foundation

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

# Where do we look for more *CP*-violation?



The Nobel Foundation



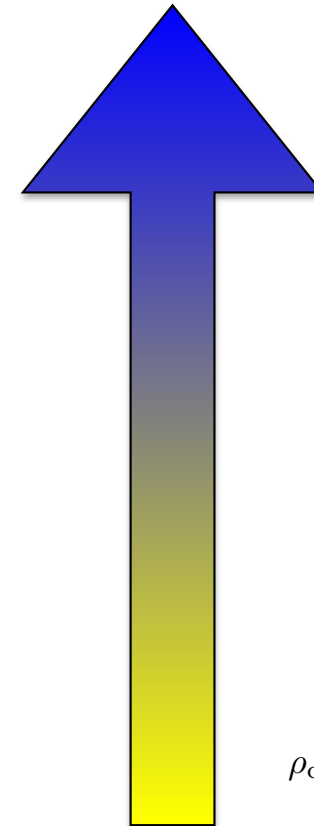
The Nobel Foundation

- Decays of *B*-mesons [Belle II] & Rare decays of b-hadrons [LHCb]
- Angular correlations in decay of positronium [MSU / Wittenberg] [See Oscar's talk](#)
- D-coefficient in beta-decay [MORA] [See talks by L. Lalanne & A. de Roubin](#)
- Nuclear magnetic quadrupole moments [Caltech, UNLV, ODU]
- Polarized neutron transmission through polarized nuclei [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

$$\vec{d} = \int \rho_{\text{charge}} \left( \vec{r} - \vec{R}_{\text{CM}} \right) d^3r = \langle \rho_{\text{charge}} \vec{r} \rangle - Q \vec{R}_{\text{CM}}$$



$\vec{d}$

$\rho_{\text{charge}}$  = charge distribution

$\vec{r}$  = position vector

$\vec{R}_{\text{CM}}$  = center of mass

$Q$  = net charge

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

# 2023 EDM Limits:

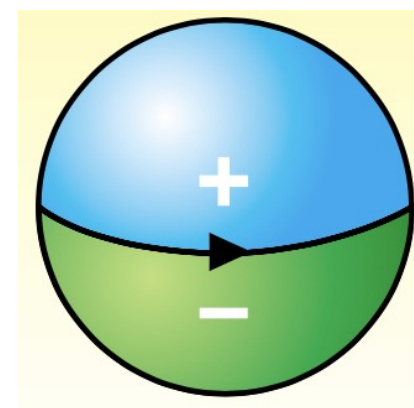
## “Free” of Standard Model (SM) “Backgrounds”

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-28 $e$ cm	SM estimate 1E-28 $e$ cm	Method (Location)
Neutron	220	$\sim 10^{-4}$	ultracold neutrons in a bottle (PSI)
“Electron”	0.11	$\sim 10^{-7}$	cold ThO beam (JHU / UC / Harvard / Northwestern)
	0.05		trapped HfF <sup>+</sup> (JILA / Boulder)
<sup>199</sup> Hg	0.074	$\sim 10^{-6}$	atoms in vapor cell (UW-Seattle)

Imagine a <sup>199</sup>Hg atom that is composed of two oppositely charged hemispherical shells each with charge magnitude  $e$ ...

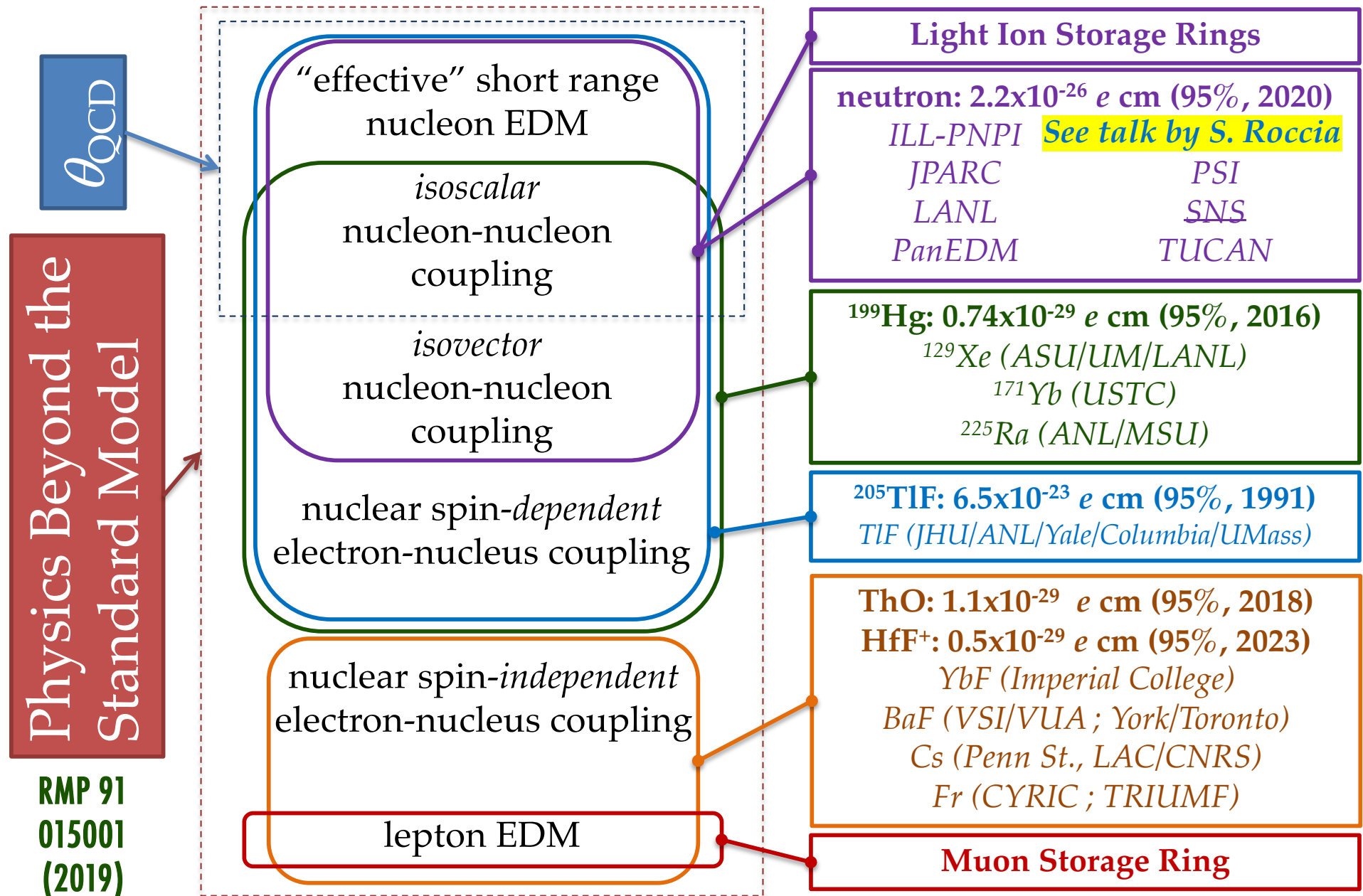
...if the <sup>199</sup>Hg atom was the size of the Earth, then the maximum thickness of these shells would be less than the diameter of a strand of human hair.



Physics Today, June 2003



# Different Sources of $\vec{T}$ in EDMs of Different Systems



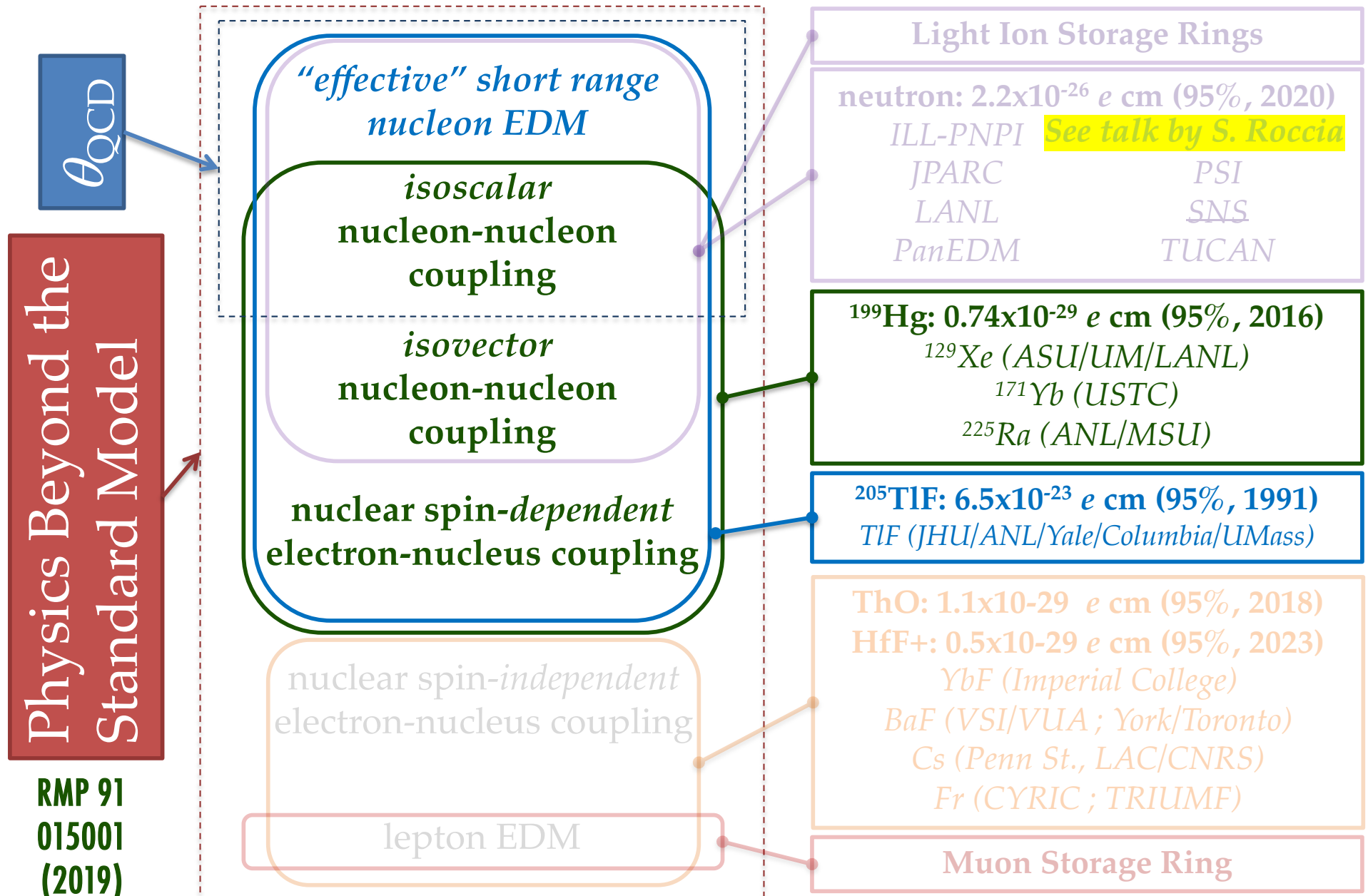
RMP 91  
015001  
(2019)

2025-03-15

PMLE Pa-229 (JTS)

7

# Different Sources of $\vec{T}$ in EDMs of Different Systems



Physics Beyond the  
Standard Model

RMP 91  
015001  
(2019)

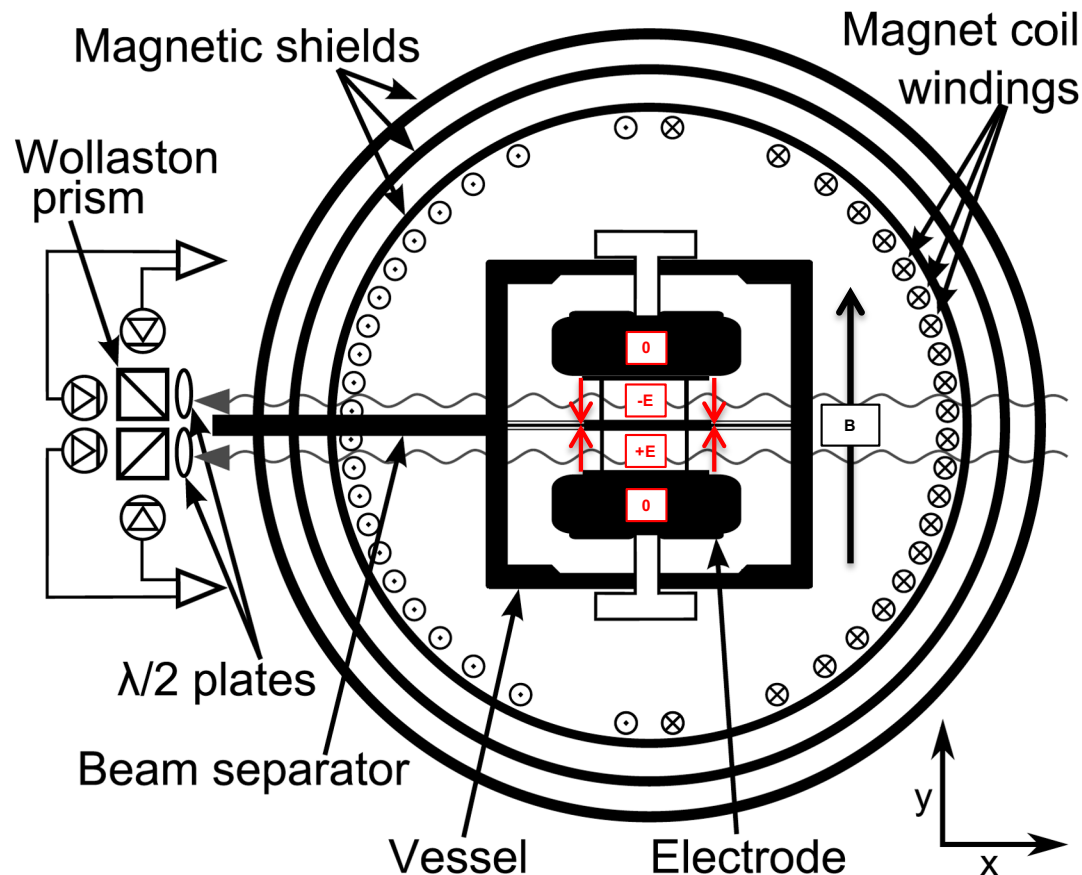
2025-03-15

PMLE Pa-229 (JTS)

8



# 2016: Atomic EDM of $^{199}\text{Hg}$ (Stable) In A Vapor Cell The Gold Standard For Over 40 Years!



- 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
- 40+ year old experiment!

Limiting systematic appears to be  $\sim 10$  nm scale motion of vapor cells when HV is switched in the presence of 2<sup>nd</sup> order  $B$ -field gradients.

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

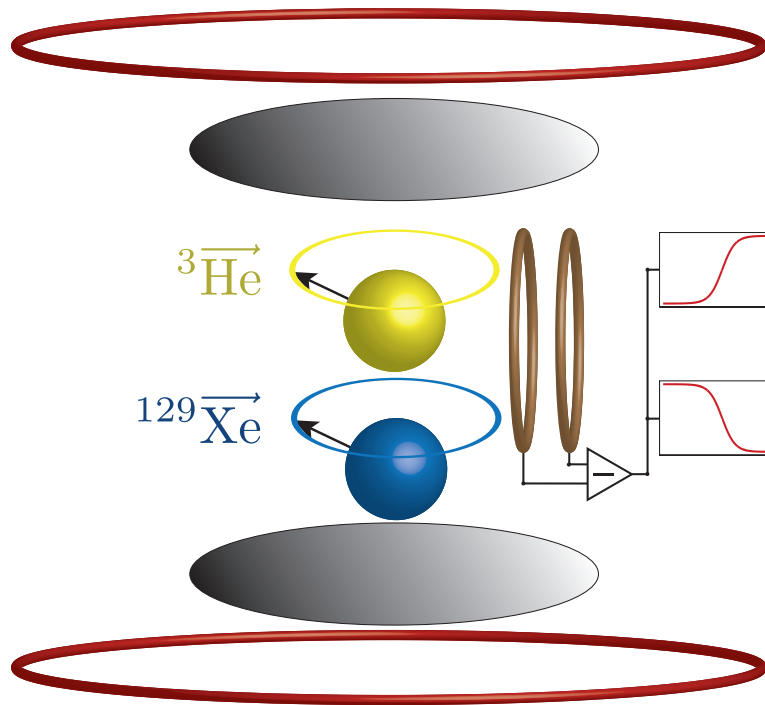
$$\Delta\nu \leq 0.1 \text{ nHz}$$

The best limit on atomic EDM:

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

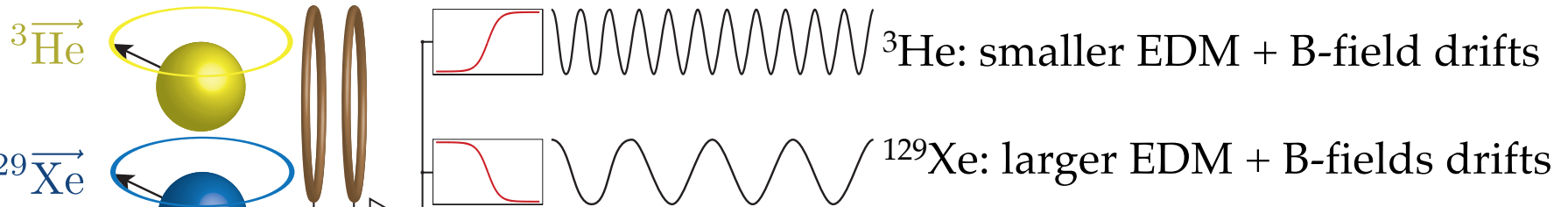
Graner et al., PRL 116:161601 (2016)

# 2019: Atomic EDM of $^{129}\text{Xe}$ (Stable) in Gas Cell Using SQUID Detection



## Polarized Noble Gases

- large magnetizations (30 pT) using SEOP
- polarized  $^3\text{He}$  for co-magnetometry
- very long spin precession times ( $10^4$  seconds)



## SQUID Detectors

- very sensitive detection (6 fT/ $\sqrt{\text{Hz}}$ )

## Magnetically Shielded Room (BMSR-2 & TUM)

- small ( $<1$  nT) and uniform ( $<10$  pT/cm) residual  $B$ -field
- high shielding factor ( $>10^8$ )

I. Altarev et al. J. Appl. Phys. 117, 233903 (2015)

I. Altarev et al. J. Appl. Phys. 117, 183903 (2015)



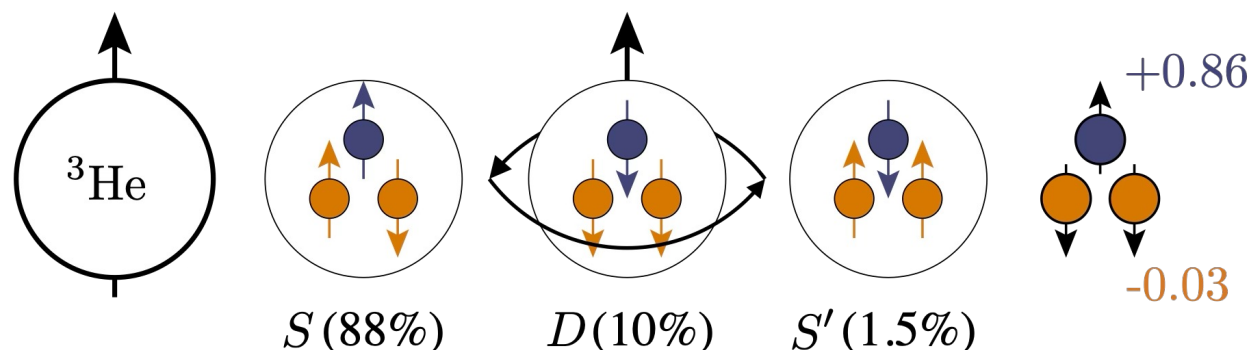
# “Hyperpolarized” Noble Gases for Nuclear Physics: The Charge Distribution of the Neutron



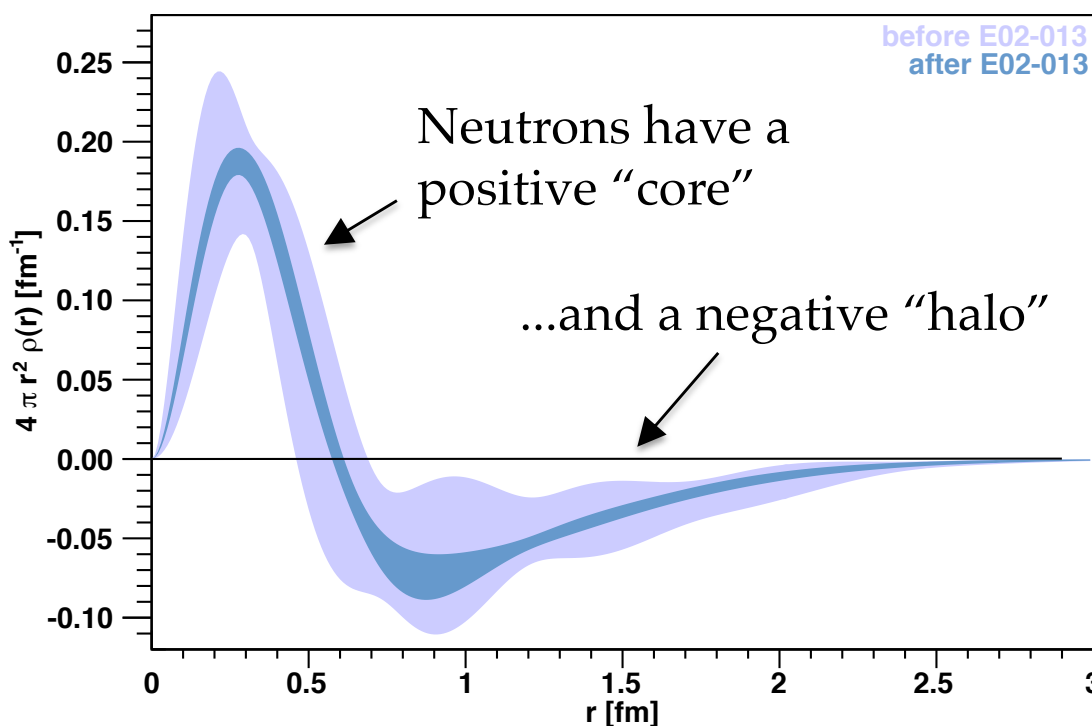
M.A. Bouchiat  
photo: Zolotrev

Density = 3 amg  
Volume = 30 cm<sup>3</sup>  
Polarization = 0.01%  
M.A. Bouchiat et al.  
Phys. Rev. Lett. 5, 373 (1960)

Density = 9 amg  
Volume = 400 cm<sup>3</sup>  
Polarization = 69%  
JTS et al.  
Phys. Rev. C 91, 055205 (2015)



Phys. Rev. C 29, 538 - 552 (1984) and Phys. Rev. C 42, 2310 - 2314 (1990)



Phys. Rev. Lett. 105, 262302 (2010)

# “Hyperpolarized” Noble Gases for Lung MRI



M.A. Bouchiat  
photo: Zolotrev

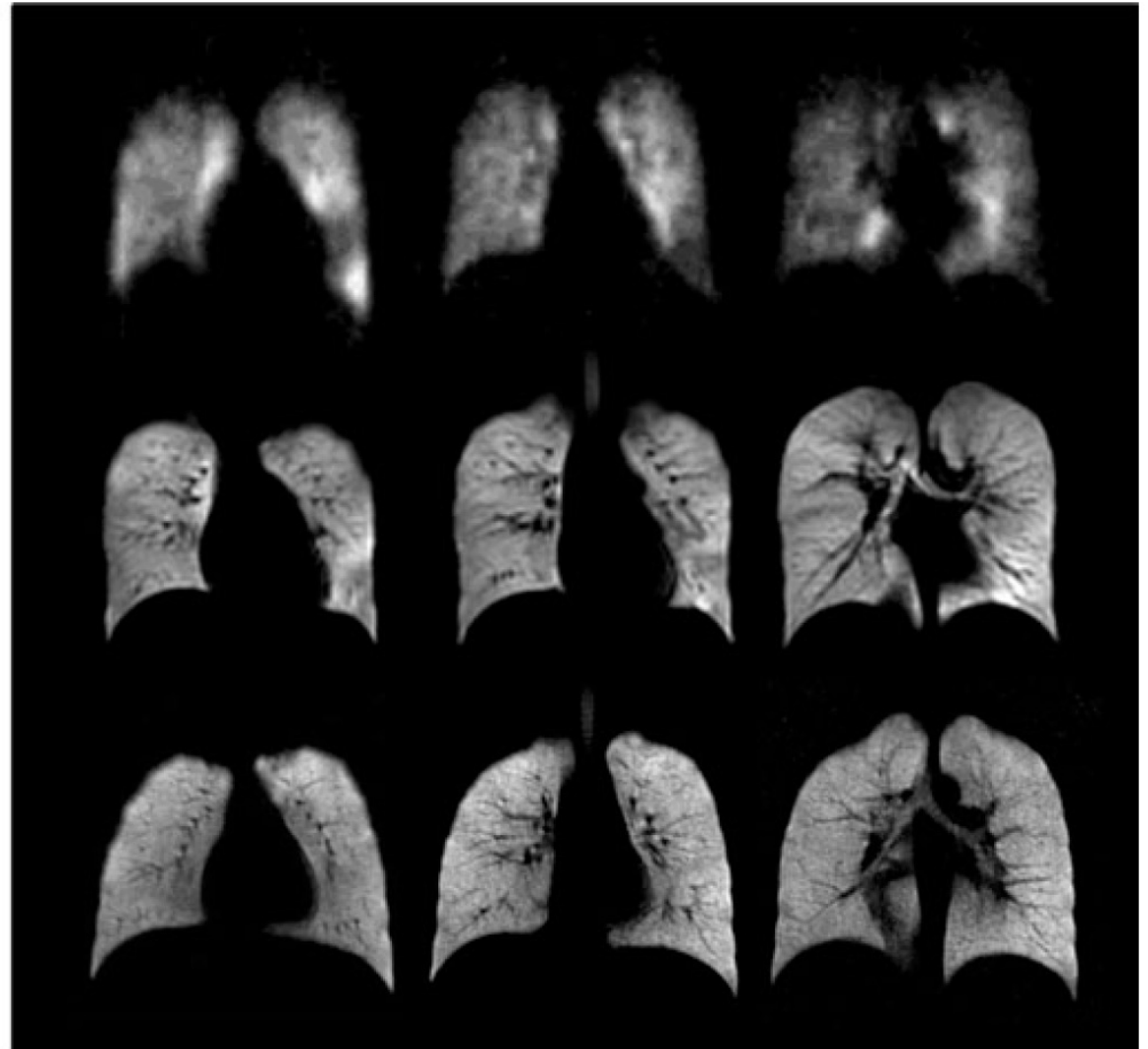
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M.A. Bouchiat et al.  
Phys. Rev. Lett. 5, 373 (1960)

Density = 9 amg  
Volume = 400 cm<sup>3</sup>  
Polarization = 69%  
JTS et al.  
Phys. Rev. C 91, 055205 (2015)

$^{129}\text{Xe}$   
(1996)

$^3\text{He}$   
(typical)

$^{129}\text{Xe}$   
(2009)



Mugler & Altes, JMRI 37 313 (2013)

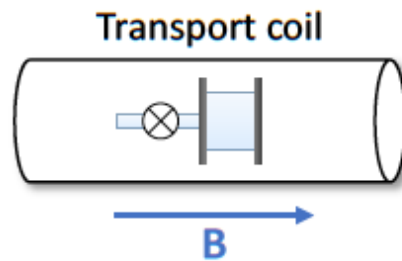
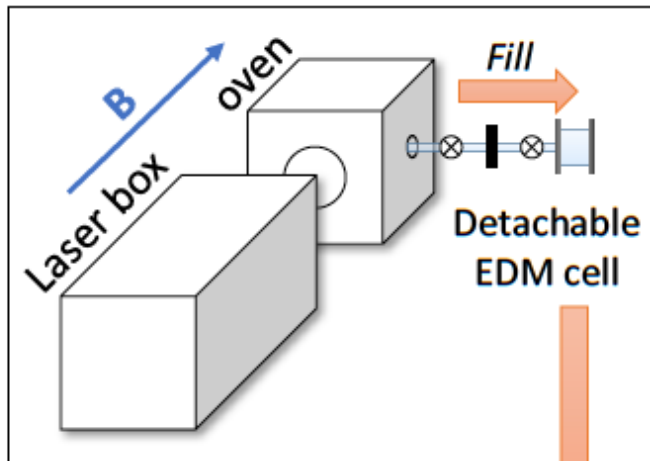


# Magnetically Shielded Room in Garching, Germany

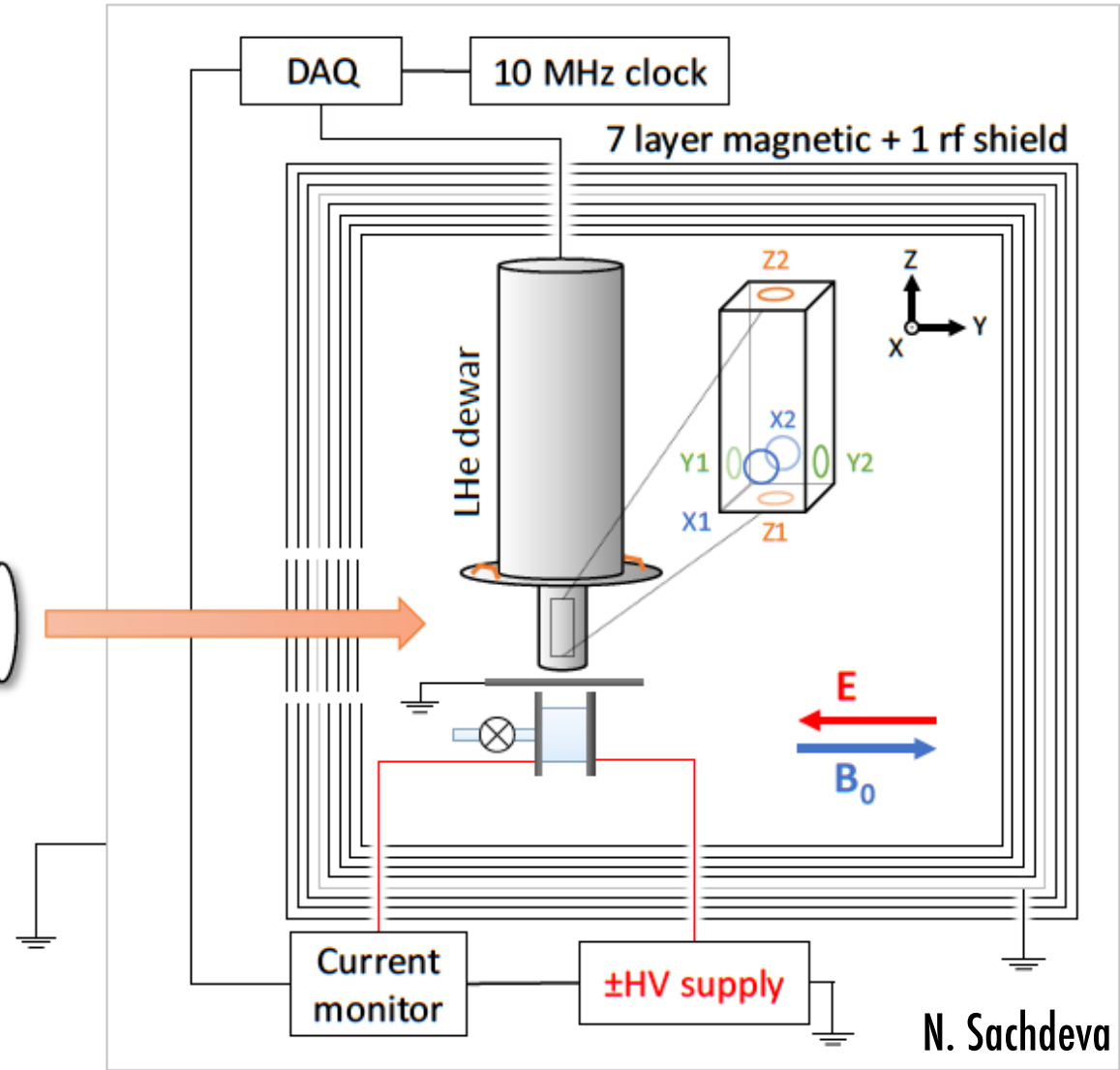


# 2019: The Atomic EDM of $^{129}\text{Xe}$ (Stable) in Berlin

*Polarizer room, ground floor*



*BMSR-2, first floor*



N. Sachdeva



# Main Systematic: Residual Longitudinal Polarization

PRL 123:143003 (2019)

PRA 100, 012502 (2019)

raw signal

integrated frequency sensitivity  
 $\sim 0.1 \mu\text{Hz}$  (30 days)

filtered signal

zoom in on filtered signal

$$\omega_{\text{co}} = \omega_{\text{Xe}} - \left[ \frac{\gamma_{\text{Xe}}}{\gamma_{\text{He}}} \right] \omega_{\text{He}}$$

“comagnetometer” signal

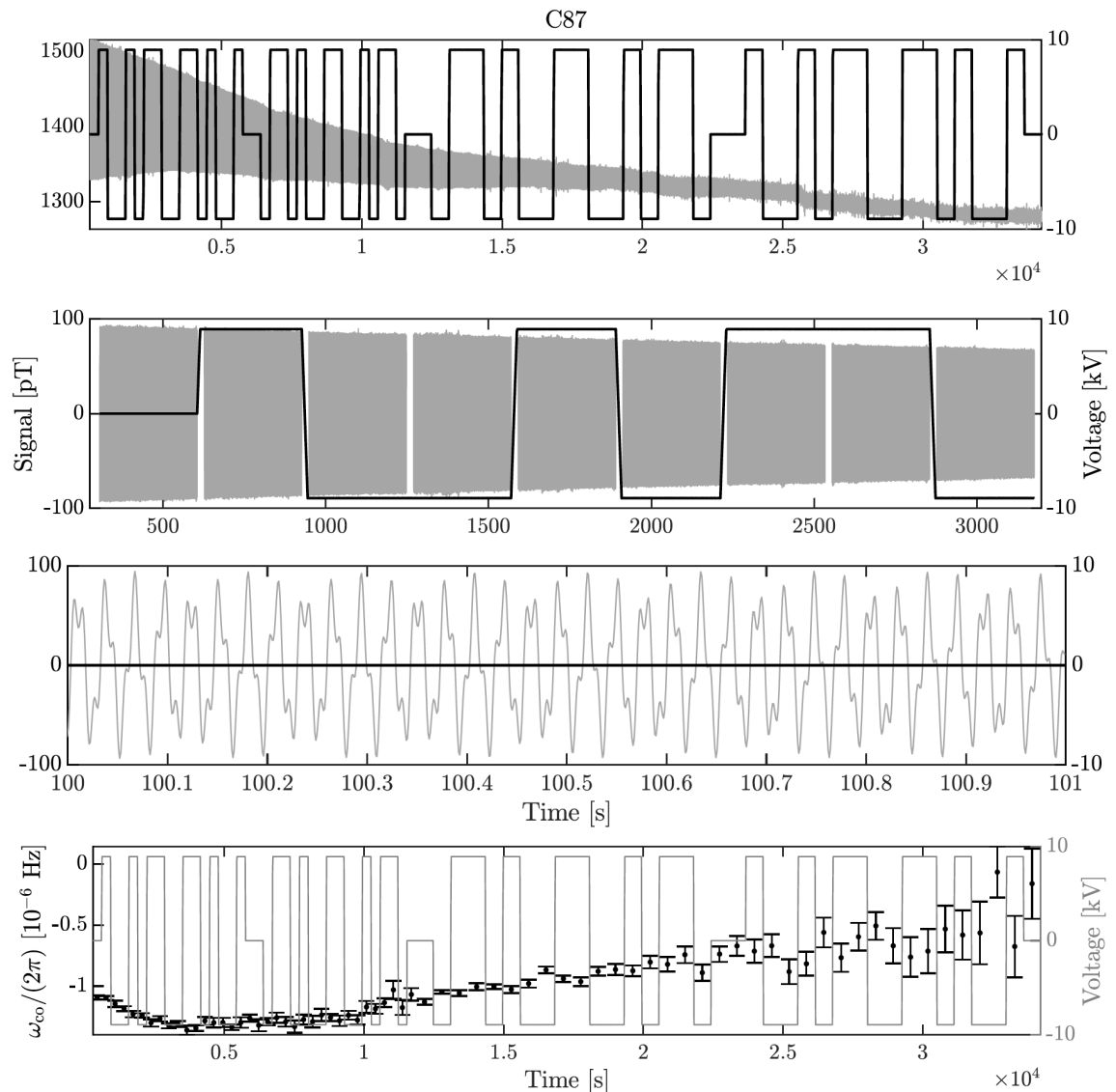
Next Generation  $^{129}\text{Xe}$  @ LANL:

W. Terrano (Arizona St.)

T. Chupp (Michigan)

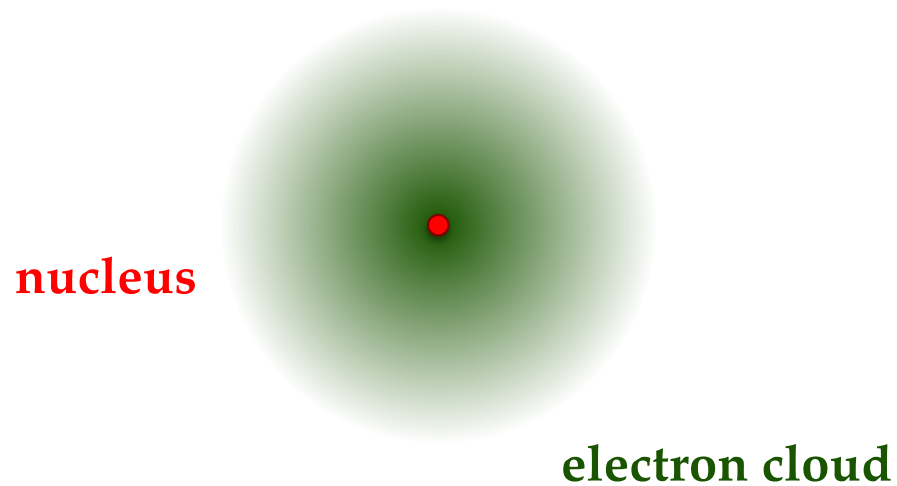
T. Ito (Los Alamos)

1000x improvement in statistical sensitivity is feasible using  
only demonstrated state of the art [Quantum Sci. Technol. 7 014001 (2022)]



# Diamagnetic Atoms: All Electrons Are Paired

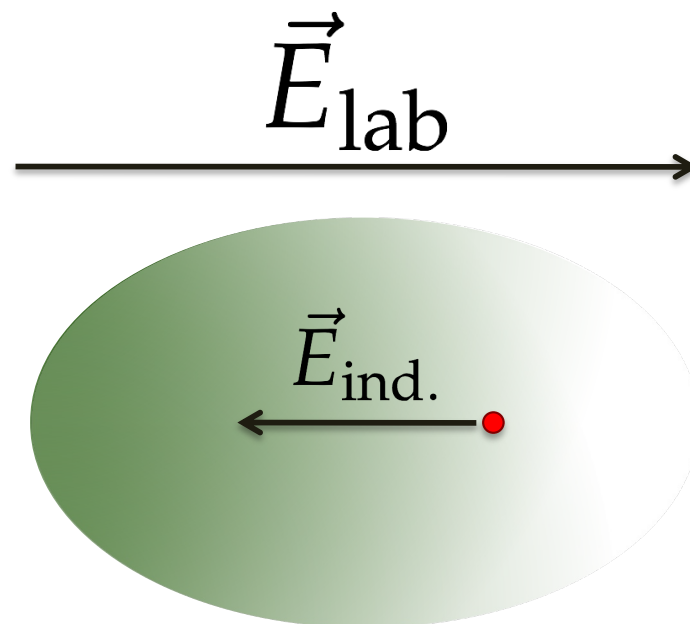
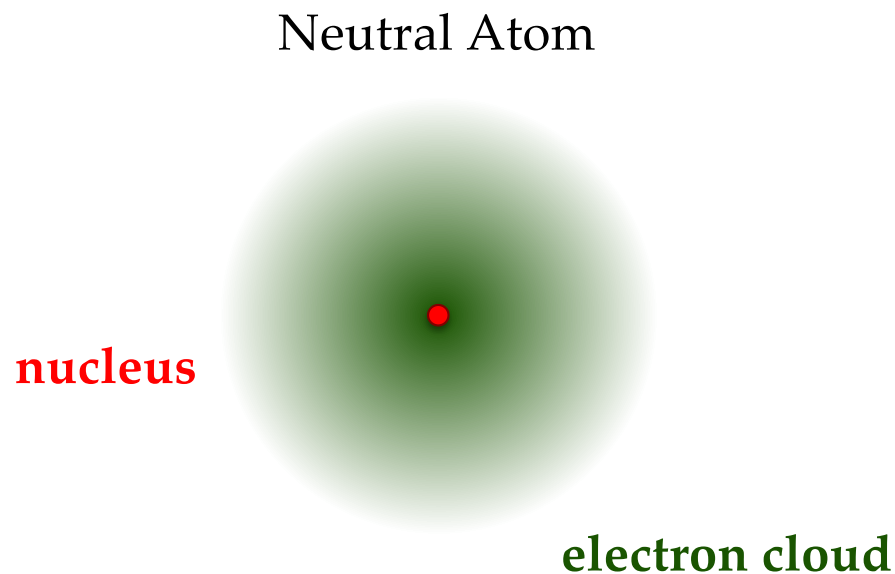
Neutral Atom



# Schiff Shielding in Diamagnetic Atoms

- Shielding in Diamagnetic Atoms

Schiff PR 132:2194 (1963)



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

# 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)

- $^{225}\text{Ra}$  vs  $^{199}\text{Hg}$  vs.  $^3\text{He}$ : 2.8 to 1 to  $10^{-5}$

JPB:AMOP 53:195004 (2020) & Phys. Rev. A 106, 022817 (2022)

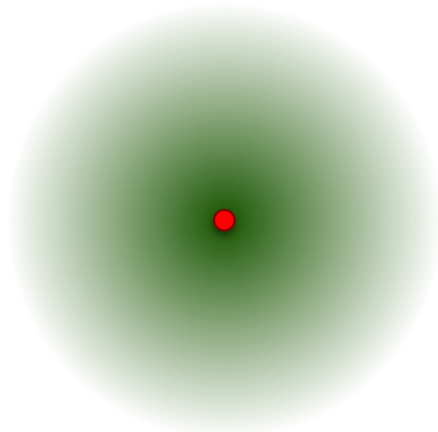
Madame  
Professor  
Marie-Anne  
Bouchiat



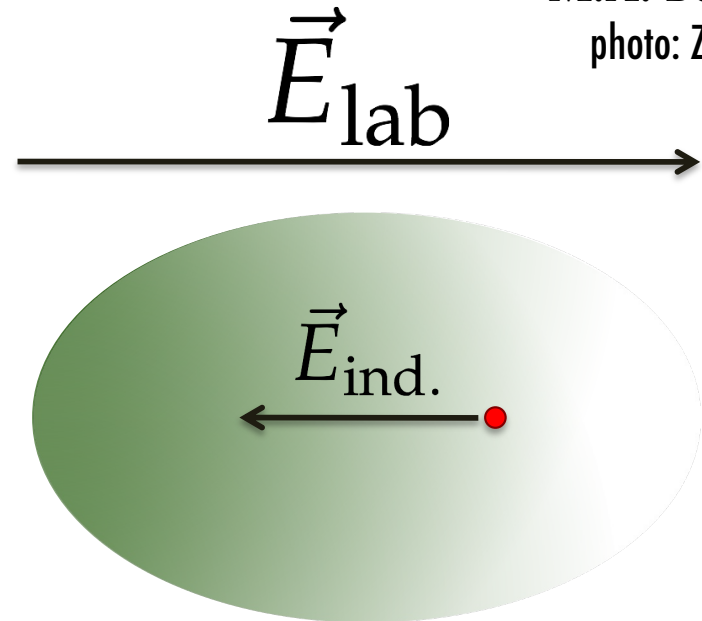
M.A. Bouchiat  
photo: Zolotrev

Neutral Atom

nucleus



electron cloud



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



# 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"**

Physics Letters 22:290 (1966) & Physics Letters 48B:111 (1974)

- $^{225}\text{Ra}$  vs  $^{199}\text{Hg}$  vs.  $^3\text{He}$ : 2.8 to 1 to  $10^{-5}$**

JPB:AMOP 53:195004 (2020) & Phys. Rev. A 106, 022817 (2022)

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

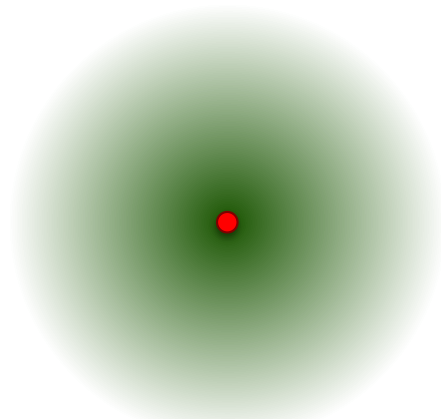
Schiff Moment

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

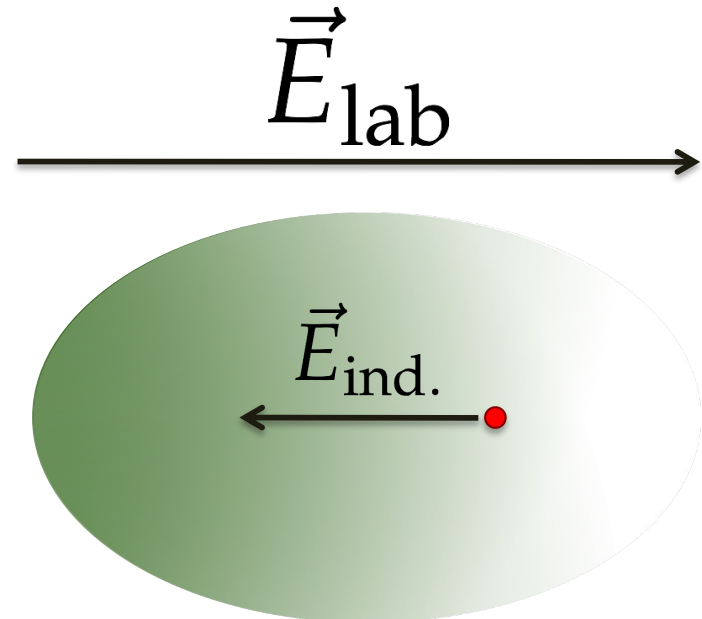
Zh. Eksp. Teor. Fiz. 87, 1521-1540 (1984)

Neutral Atom

nucleus



electron cloud



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

# $\mathbb{P}$ & $\mathbb{T}$ Physics: First Order Perturbation Theory

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

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

- The  $\mathbb{P}$  and  $\mathbb{T}$  physics that we seek (unknown & common to all isotopes)

# Isotopes With Nearly Degenerate Nuclear States

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

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

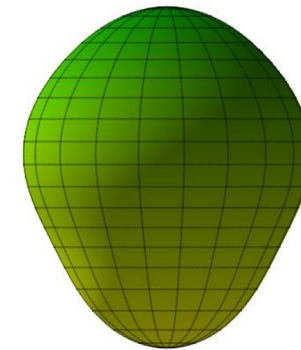
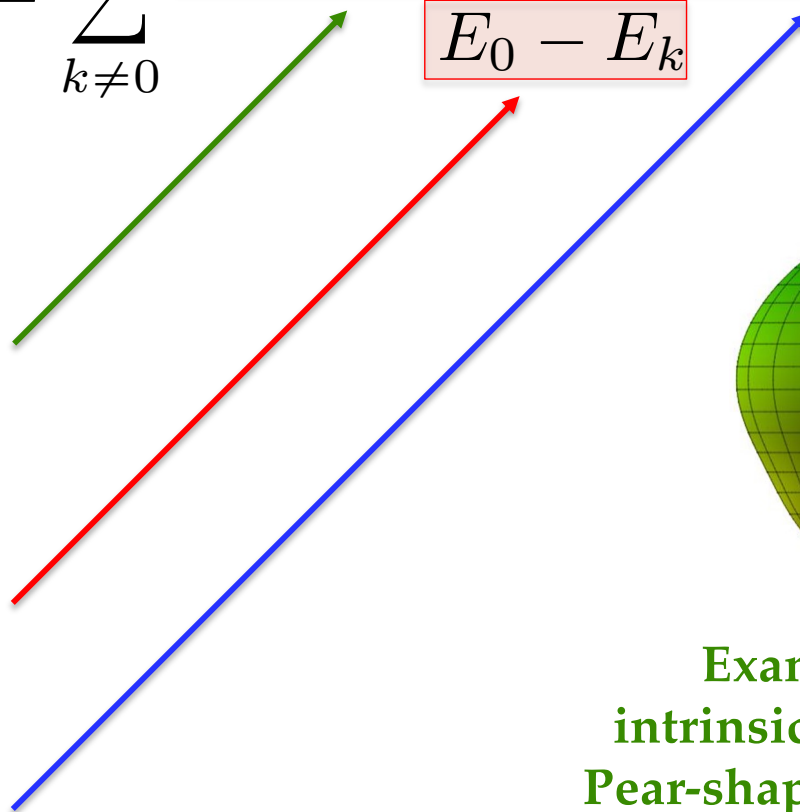
- Difference in lab-frame nuclear energy levels
- The  $\mathbb{P}$  and  $\mathbb{T}$  physics that we seek (unknown & common to all isotopes)

# 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”



# Pear-Shaped Nuclei = Nearly Degenerate Parity Doublets

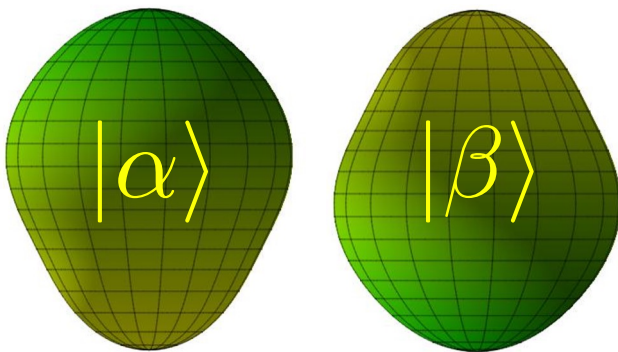
$$S_z = \frac{\langle er^2 z \rangle}{10} - \frac{\langle r^2 \rangle \langle ez \rangle}{6}$$

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

**Parity Doublet**

- **Nearly degenerate parity doublet**

Haxton & Henley PRL 51:1937 (1983)



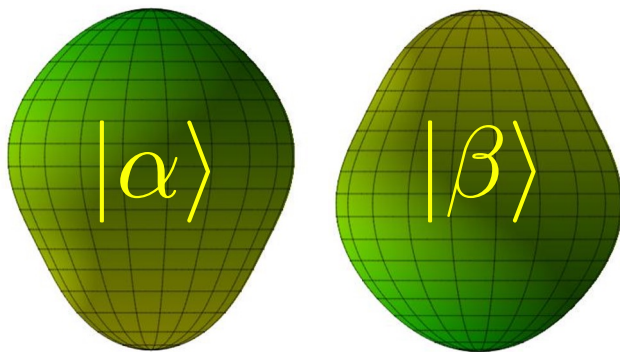
$$\begin{array}{l} \overline{\hspace{1cm}} \\ \uparrow \Delta E \\ \underline{\hspace{1cm}} \end{array} \quad \begin{array}{l} |\Psi_1\rangle = \frac{|\alpha\rangle \mp |\beta\rangle}{\sqrt{2}} \\ \\ |\Psi_0\rangle = \frac{|\alpha\rangle \pm |\beta\rangle}{\sqrt{2}} \end{array}$$

# Pear-Shaped Nuclei = Enhanced Intrinsic Schiff Moments

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

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

## Parity Doublet



- 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)

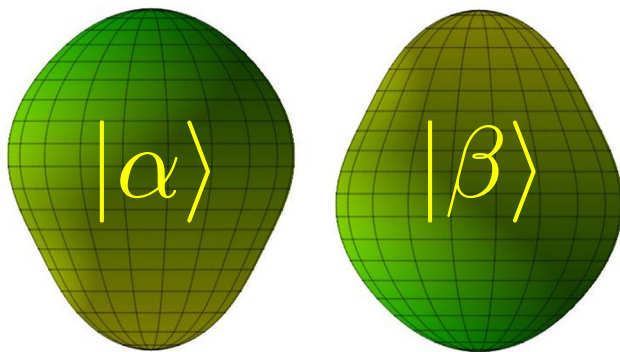
$$\begin{array}{l} \overline{\hspace{1cm}} \\ \uparrow \Delta E \\ \underline{\hspace{1cm}} \end{array} \quad \begin{array}{l} |\Psi_1\rangle = \frac{|\alpha\rangle \mp |\beta\rangle}{\sqrt{2}} \\ \\ |\Psi_0\rangle = \frac{|\alpha\rangle \pm |\beta\rangle}{\sqrt{2}} \end{array}$$

# Example: Enhanced Sensitivity in Radium-225

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

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

## Parity Doublet



- 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 ( $^{225}\text{Ra}$ ) / EDM ( $^{199}\text{Hg}$ )**

55 keV

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

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

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

$^{225}\text{Ra}$ : Dobaczewski & Engel PRL 94:232502 (2005)

$^{199}\text{Hg}$ : Ban et al. PRC 82:015501 (2010)

# Nuclear Structure Calculations Are Challenging!

type	$^{199}\text{Hg}$	$^{225}\text{Ra}$	ratio*2.8	references
SIII	0.005	7.0	4300	PRC 82 015501 (2010)
SkM*	-0.027	21.5	-2400	PRC 82 015501 (2010)
SLy4	-0.006	16.9	-8600	PRC 82 015501 (2010)
SkO'		6.0		
DE05	0.071			PRC 72 045503 (2005)
DS03	0.055			PAN 66 1940 (2003)
"Best"	+ / -(0.02)	6.0	+ / -(900)?	Prog. PNP 71 21 (2013)

- Isovector coupling is given by “chromo”-EDMs
- Nuclei are the most sensitive to this source of new physics
- Opportunity for  $^{225}\text{Ra}$  or other octupole deformed species



# Ongoing: The Atomic EDM of $^{225}\text{Ra}$ at Argonne

$|d(^{225}\text{Ra})| < 50 \times 10^{-23} \text{ e-cm (95\%)}$

PRL 114:233002 (2015)

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

equivalent to  $\sim 1000 \times \text{EDM}(^{199}\text{Hg})$

PRC 94:025501 (2016)

Upgrades underway to improve sensitivity by  $\times 1000$

Spectrochimica Acta Part B 172 105967 (2020)

$^{226}\text{Ra}$

nuclear spin = 0

$t_{1/2} = 1600 \text{ years}$

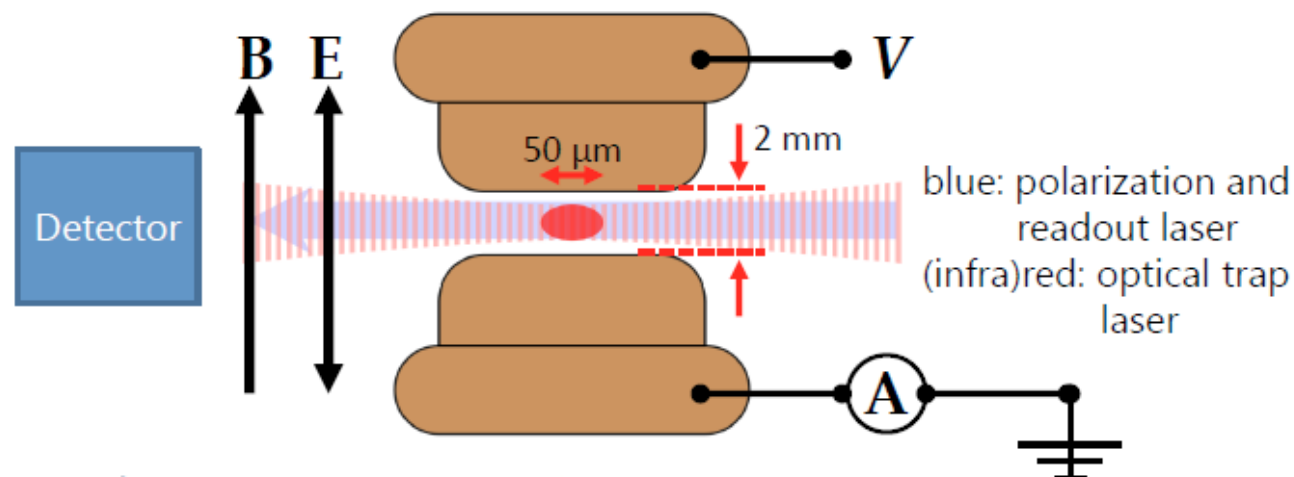
Low vapor pressure

$^{225}\text{Ra}$

Nuclear Spin =  $\frac{1}{2}$

$t_{1/2} = 15 \text{ days}$

Low vapor pressure



EDM search using atoms held in Optical Lattice

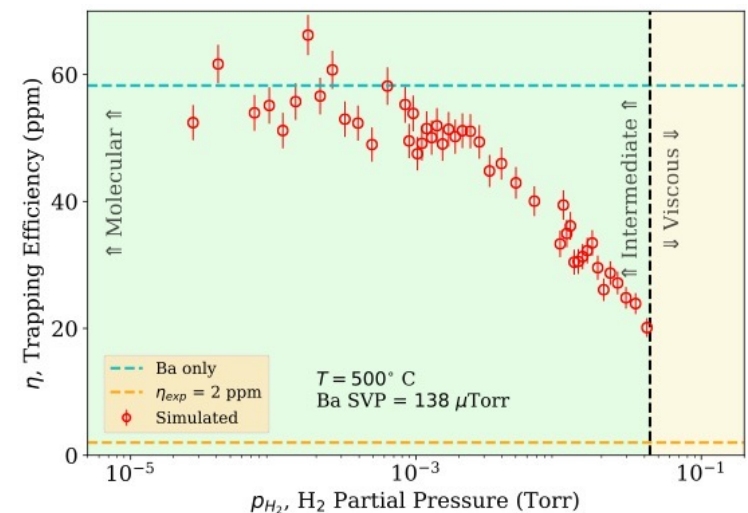
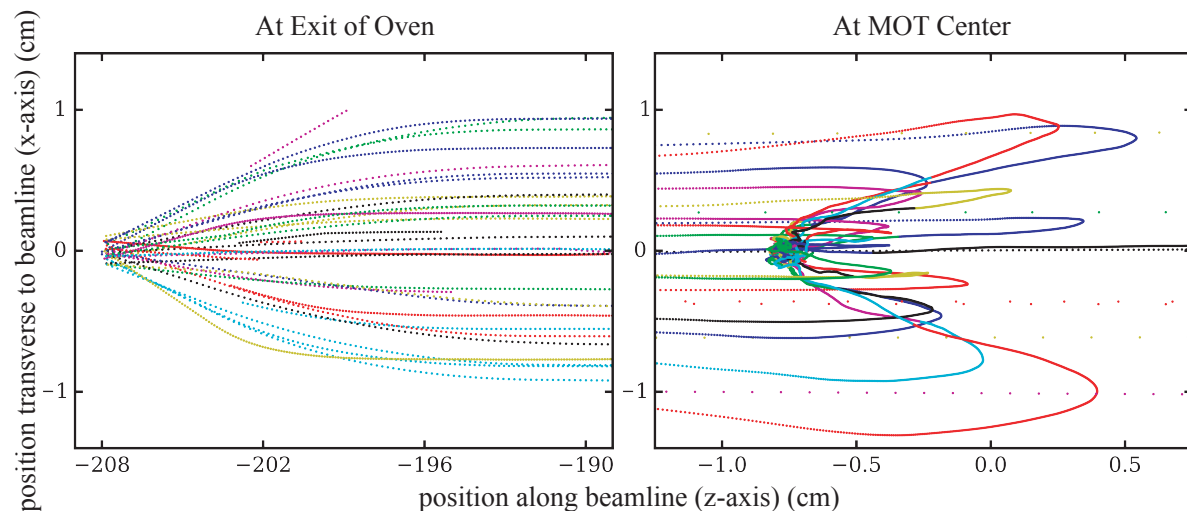
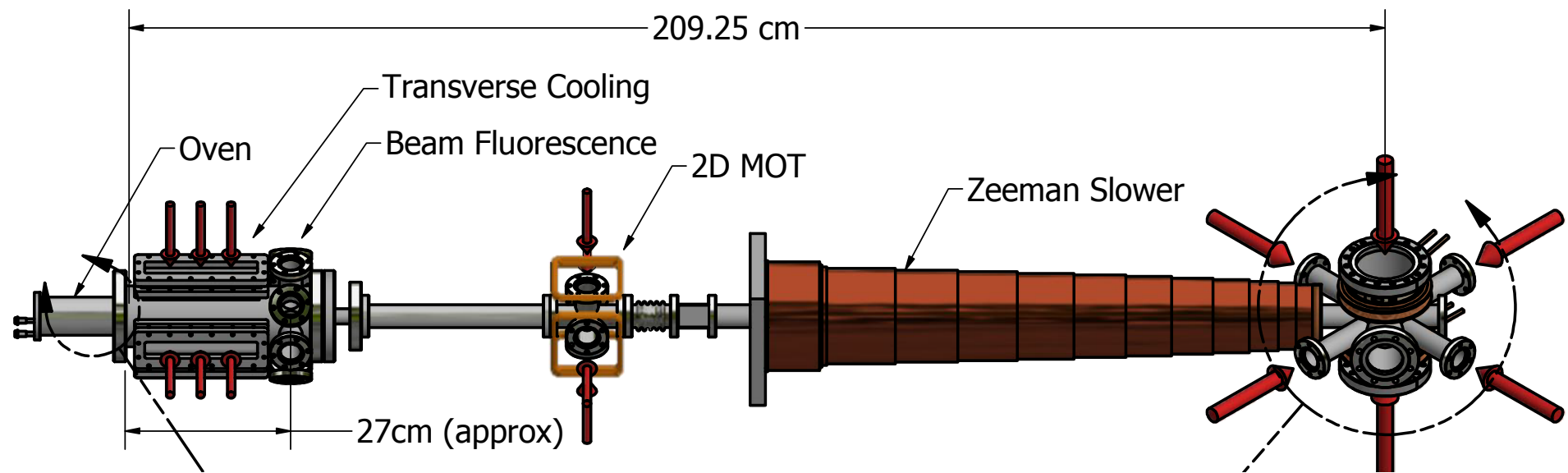
Romalis & Fortson PRA 59:4547 (1999)

Chin et al. PRA 63:033401 (2001)

Bishof et al. PRC 94:025501 (2016)

- Atoms concentrated in a very small region
- Long coherence time (100 s) PRL 129, 083001 (2022)
- negligible " $\mathbf{v} \times \mathbf{E}$ " systematics
- High electric field ( $> 300 \text{ kV/cm}$ ) in vacuum NIMA 1014 165738 (2021)
- Light-induced systematic effects can be controlled!

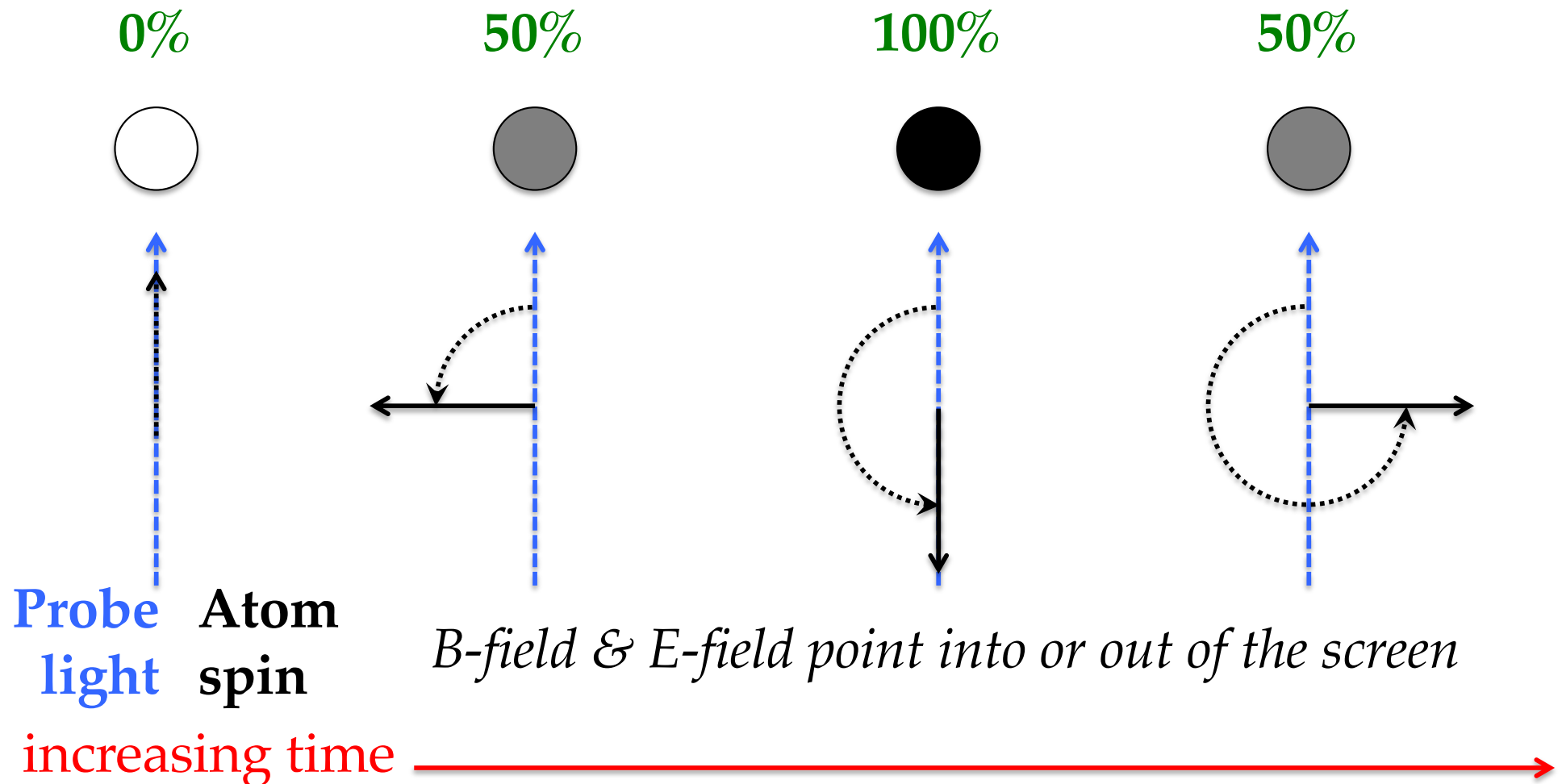
# Current Laser Trapping Efficiency: 2 ppm “Blue” Upgrade Being Implemented: >200 ppm



D. A. Potterveld, S. A. Fromm et al. (under review with PRA)

# The Absorption Probability Oscillates at the Spin Precession Frequency ( $\sim 20$ Hz)

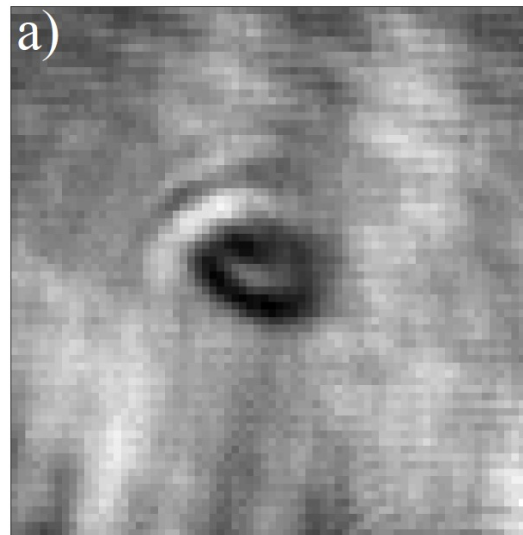
probability of absorbing probe light and creating a shadow:



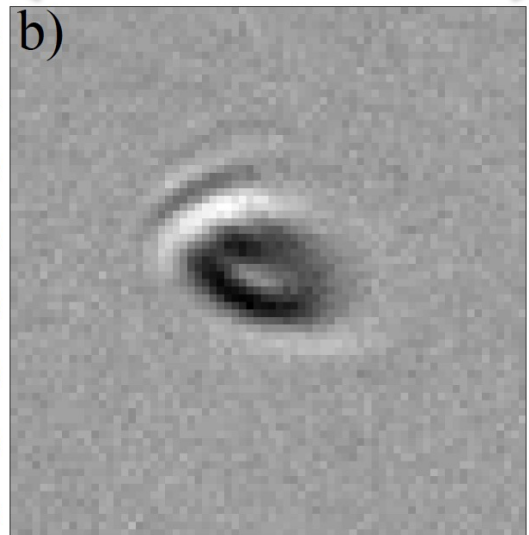


# Image Background & Distortion Corrections

average of 8  
raw images  
of  $^{226}\text{Ra}$

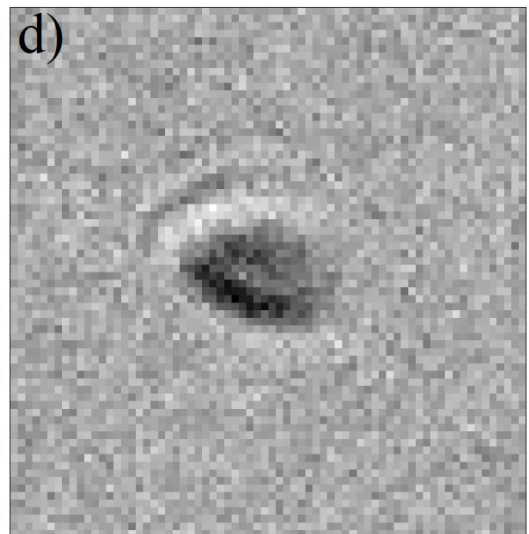
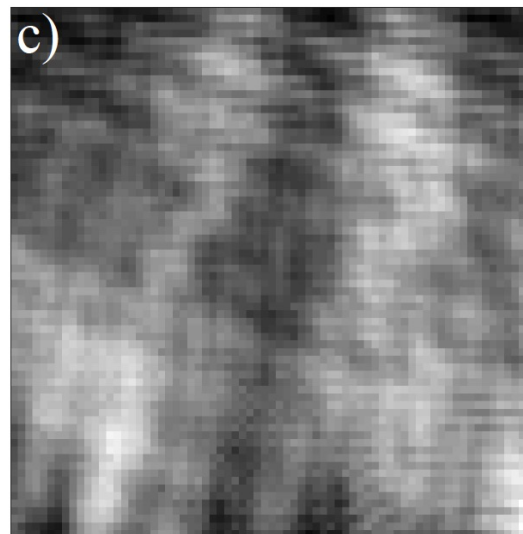


0.45 mm  
←→



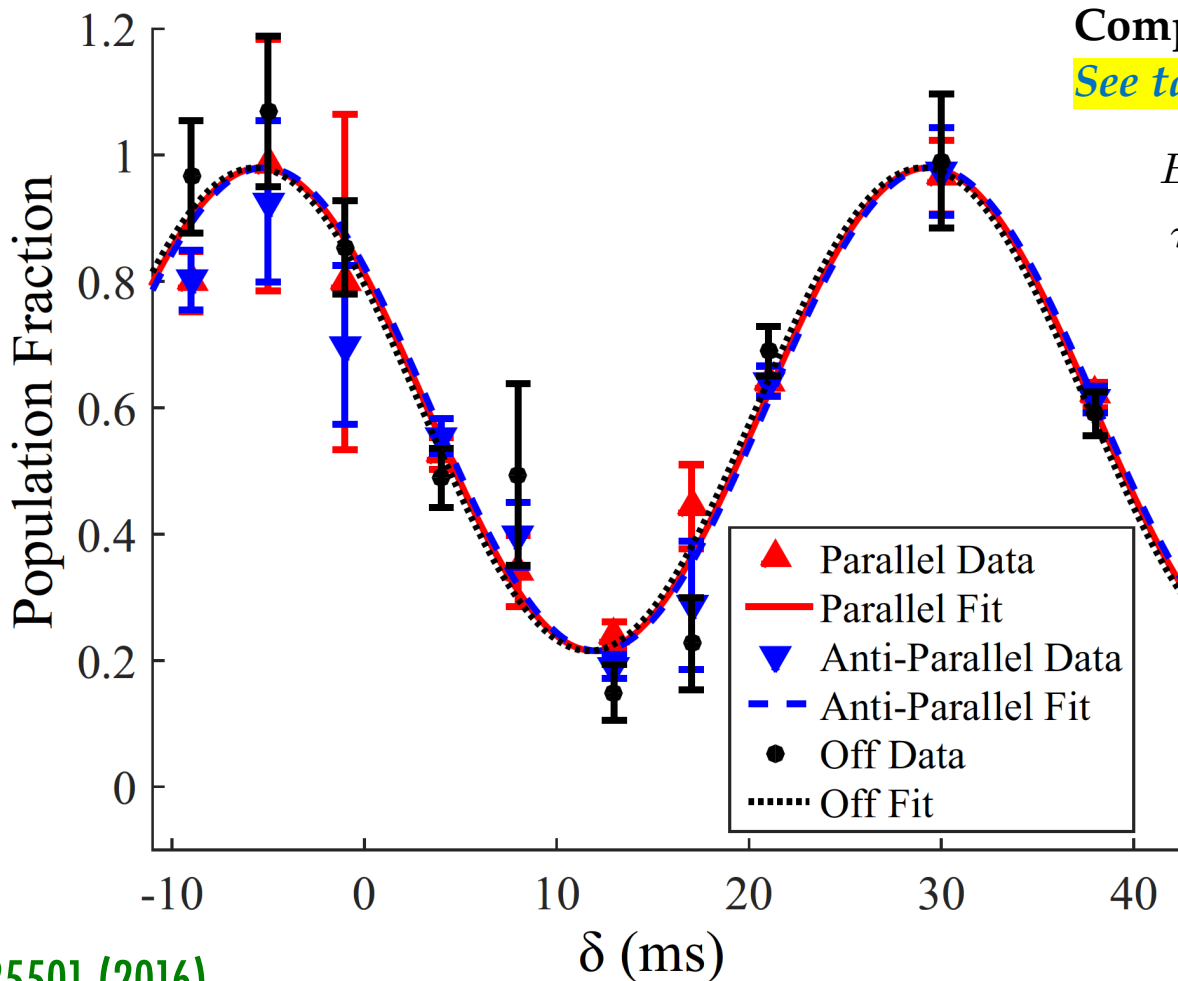
"clean"  
 $^{226}\text{Ra}$   
image  
 $10^4$  atoms

average of 63  
raw images  
of  $^{225}\text{Ra}$



"clean"  
 $^{225}\text{Ra}$   
image  
 $10^2$  atoms

# Reconstructed Spin Precession Curve After $\tau = 20$ s (Shadow Measurements Taken At Different Time Delays)



Completely Statistics Dominated  
*See talk by G. Arrowsmith-Kron*

$E = 67 \text{ kV/cm} \rightarrow 500 \text{ kV/cm}$

$\tau = 2 \text{ s} \rightarrow 20 \text{ s} \rightarrow 200 \text{ s}$

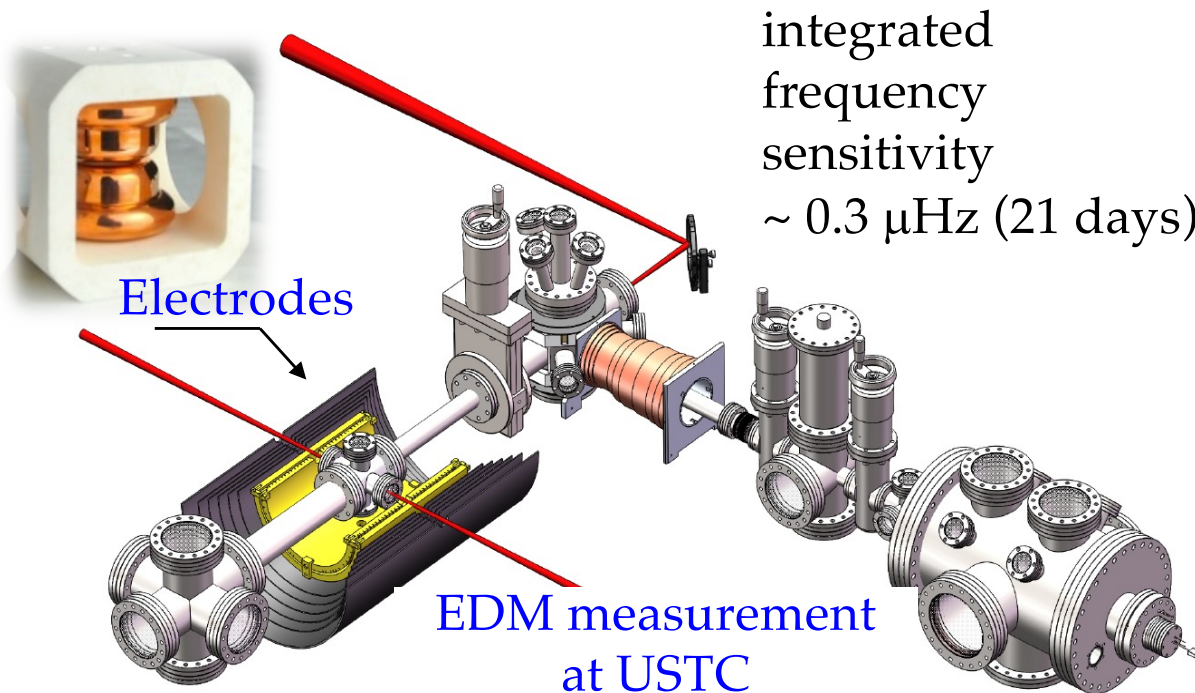
$$d = \frac{h(\Delta\phi)}{4E\tau}$$

integrated  
frequency  
sensitivity  
 $\sim 1 \text{ mHz (15 days)}$

PRC 94:025501 (2016)

$\Delta\phi$  = phase shift between red and blue curves

# 2022: Atomic EDM of $^{171}\text{Yb}$ (Stable) in a Laser Trap Using Laser Probing



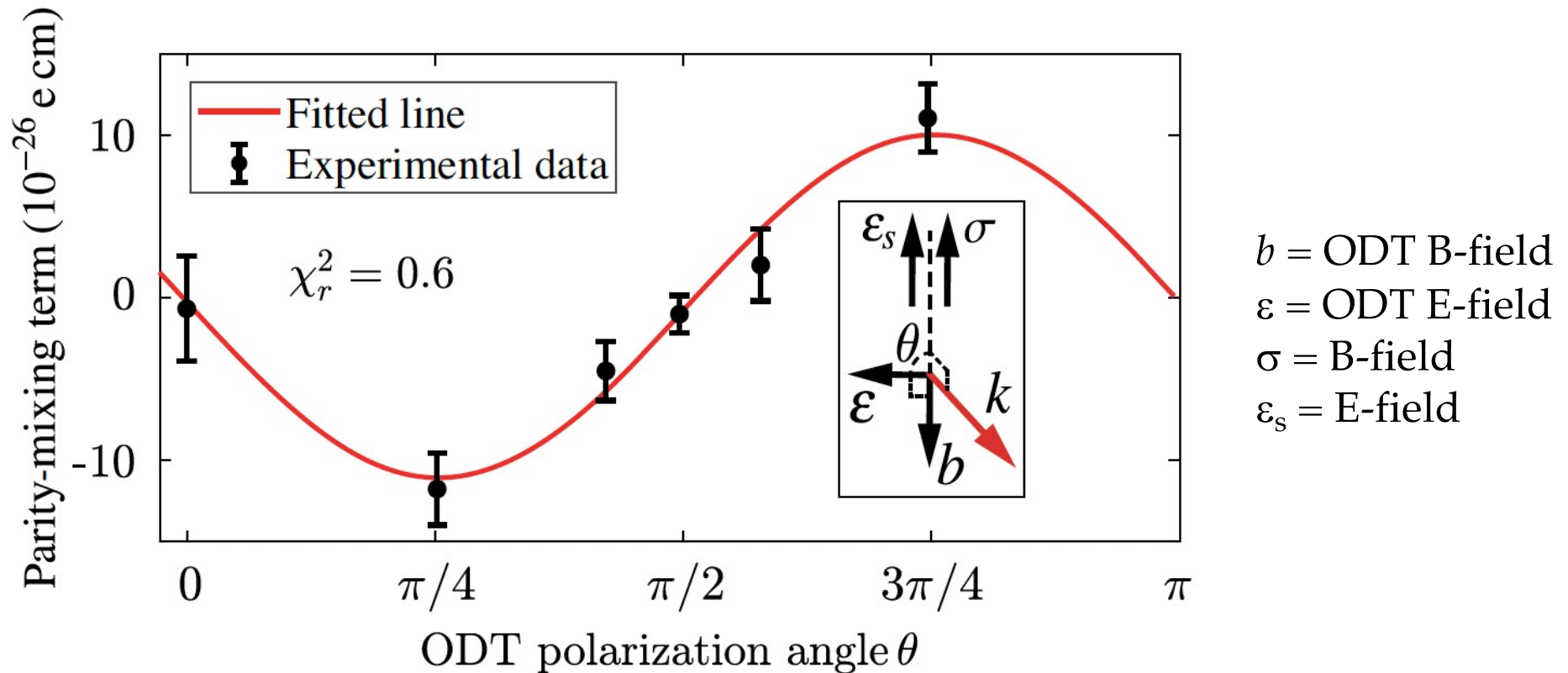
- Pathfinder experiment for  $^{225}\text{Ra}$  ( $10^5$  vs  $10^2$  atoms)
- Coherent spin precession time  $> 300$  s
- $\text{EDM}(^{171}\text{Yb}) < 1.5 \times 10^{-26} \text{ e-cm}$  (95% C.L.),  
equivalent to  $\sim 1000 \times \text{EDM}(^{199}\text{Hg})$

PRL 129, 083001 (2022)

slide from Z.-T. Lu

- Determined the magic ODT (optical dipole trap) wavelength  
PRA 102, 062805 (2020)
- Developed a quantum non-demolition (QND) method with a spin-detection efficiency of 50%  
Phys. Rev. App. 19, 054015 (2023)
- Observed the systematic due to parity mixing in ODT, and suppressed the effect by averaging measurements with ODTs in opposite directions
- Upgrades underway to improve sensitivity by  $\times 100$

# Measured Laser Trap Systematics for $^{171}\text{Yb}$ EDM is Good Enough for Next Generation $^{225}\text{Ra}$ EDM



$$\Delta\nu = \nu_1(\hat{b} \cdot \hat{\sigma})(\hat{\epsilon} \cdot \hat{\epsilon}_s) + \nu_2(\hat{b} \cdot \hat{\epsilon}_s)(\hat{\epsilon} \cdot \hat{\sigma})$$

Frequency shift scale factor of  $^{225}\text{Ra}$

[See talk by G. Arrowsmith-Kron](#)

PRL 129, 083001 (2022)

# Comparison of $^{129}\text{Xe}$ and $^{171}\text{Yb}$ Limits (Not Pear-Shaped)

$^{225}\text{Ra}$ : PRC 94:025501 (2016):  $< 1.4 \times 10^{-23} \text{ e cm (95\%)}$

(rare pear-shaped nuclei + laser trap experiment: 1 mHz)

$^{129}\text{Xe}$ : PRL 123:143003 (2019):  $< 1.4 \times 10^{-27} \text{ e cm (95\%)}$

(stable + gas cell experiment: 0.1  $\mu\text{Hz}$ )

$^{171}\text{Yb}$ : PRL 129:083001 (2022):  $< 1.5 \times 10^{-26} \text{ e cm (95\%)}$

(stable + laser trap experiment: 0.3  $\mu\text{Hz}$   
nearly identical to  $^{225}\text{Ra}$  experiment)

- 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.



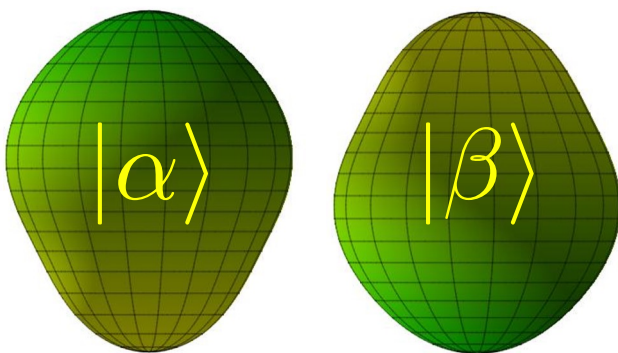
# Protactinium-229 \*May\* Be Unusually Sensitive!

Choose an isotope  
with large deformations

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

Unknown

Parity Doublet



$$\begin{aligned} |\Psi_1\rangle &= \frac{|\alpha\rangle \mp |\beta\rangle}{\sqrt{2}} \\ |\Psi_0\rangle &= \frac{|\alpha\rangle \pm |\beta\rangle}{\sqrt{2}} \end{aligned}$$

$\Delta E$

Pa-229: Haxton & Henley PRL 51:1937 (1983)

I. Ahmad et al Phys. Rev. C 92:024313 (2015)

Dobaczewski et al PRL 121, 232501 (2018)

Isotope	$\Delta E$ (keV)	$\tau_{1/2}$ (sec)	sensitivity
Hg-199	1800	stable	1
Rn-223	$\sim 10^2?$	$10^3$	$10^2$
Ra-225	55	$10^6$	$10^3$
<b>Pa-229</b>	<b>(0.06 +/- 0.05)?</b>	<b><math>10^5</math></b>	<b><math>10^6</math></b>

**FRIB will make lots of Pa-229!**

# General Indications of Octupole Deformations

1. Parity doubling (PD) is **“a necessary but not sufficient criterion for octupole deformation.” The size of the energy splitting “does not provide a useful measure of the amount of octupole deformation nor its stability...”**
2. Enhanced E3 matrix elements
3. Enhanced intrinsic (“body-frame” electric dipole moment)
4. “Decoupling factors” of the same magnitude and opposite sign for PD states
5. Alpha decay transitions to both parity bands of the daughter should be allowed with close to equal probability
6. Magnetic moment g-factors should be the same for both members of the PD

Leander & Sheline NPA 413(3):375-415 (1984)

# Indications of Octupole Deformation in $^{229}\text{Pa}$

1. Parity doubling (PD) is “a necessary but not sufficient criterion for octupole deformation.” The size of the energy splitting “does not provide a useful measure of the amount of octupole deformation nor its stability...

**Status: Looks good!**

PRC 92:024313 (2015)

2. Enhanced E3 matrix elements

**Status: Looks good! Some indirect measurements and many theory predictions.**

PRL 121, 232501 (2018) and PLB 96(1-2):7-10 (1980), PRC 37:2744-2778 (1988)

3. Enhanced intrinsic (“body-frame” electric dipole moment)

**Status: Looks good! Indirect evidence from measurements on Th-228 core**

Nature Physics 16:853-856 (2020)

4. “Decoupling factors” of the same magnitude and opposite sign for PD states

**Status: Looks good!**

NPA 576(2):267-307 (1994)

5. Alpha decay transitions to both parity bands of the daughter should be allowed with close to equal probability

**Status: Looks good!**

PRC 48(3):1003-1004 (1993)

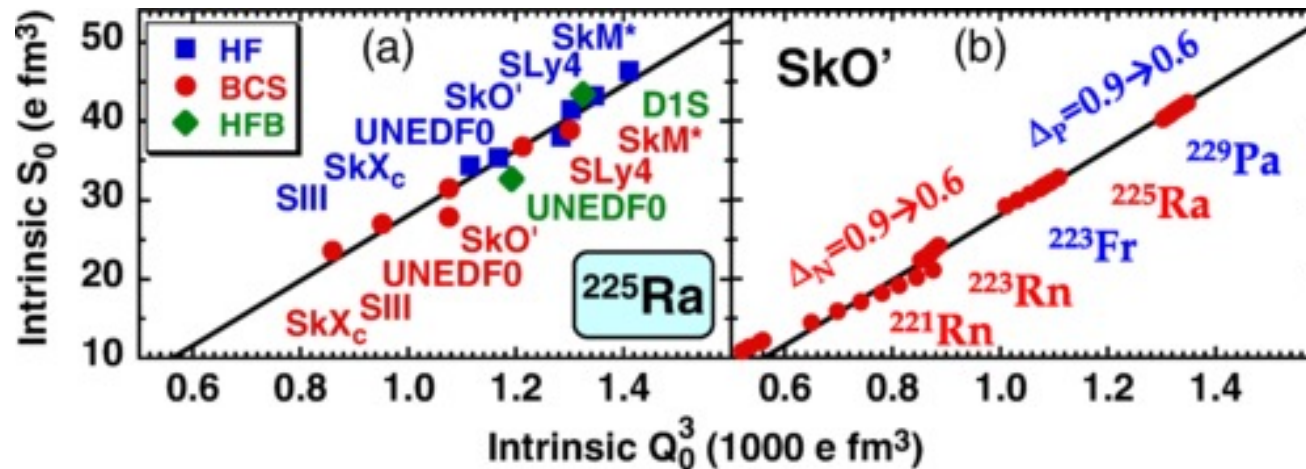
6. Magnetic moment g-factors should be the same for both members of the PD

**Status: No measurements yet**

Leander & Sheline NPA 413(3):375-415 (1984)

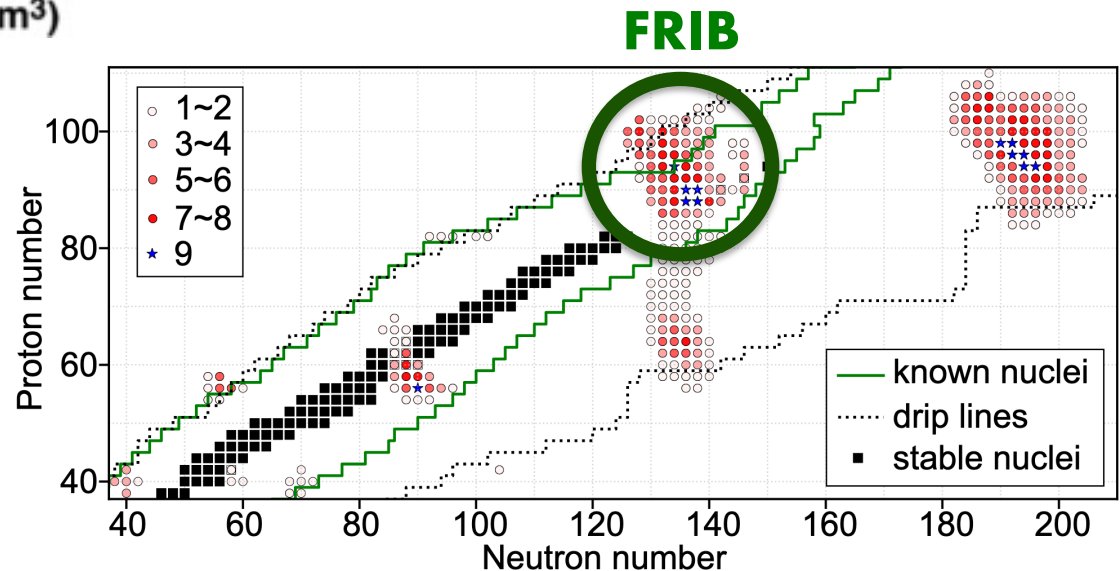
# Calibrating the Intrinsic Schiff Moment (Numerator)

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



PRL 121, 232501 (2018)  
Phys. Rev. C, 102:024311 (2020)

The Intrinsic Schiff Moment (numerator) varies by a factor of a few if not zero, but the Parity Doublet Splitting (denominator) could vary by orders of magnitude!



# Ground State Parity Doublet Energy Splitting (Denominator) The Muddled Picture for $^{229}\text{Pa}$ (1980-2015)

1980: Theoretical prediction: 100 eV

PLB 96(1-2):7-10

1982: Experimental claim from ANL: 220(50) eV

PRL 49:1758-61

1988: Theoretical calculation: 400 eV

PRC 37:2744-78



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PRC 44:R1728-31

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2015: Full reanalysis of 1982 ANL experiment + incorporation of data from German experiments: **Original claim of 220 eV conversion electrons is gone.**  
If there is a ground state parity doublet, then it must be 60(50) eV.  
PRC 92:024313 (2015)

# Ground State Parity Doublet Energy Splitting (Denominator) The Muddled Picture for $^{229}\text{Pa}$ (1980-2015)

## D. Splitting energy of the $5/2^\pm$ doublet

So far the  $5/2^-$  member of the parity doublet has not been identified. In this work we have established many excited states which would decay to the  $5/2^-$  level expected below 80 keV from the systematics. The fact that we have placed all observed transitions in the level scheme presented here leaves no  $\gamma$ -ray transition which could be attributed to the decay to the  $5/2^-$  level. From these observations we conclude that the  $5/2^-$  level is almost degenerate with some level below 80 keV, possibly the ground state. The  $5/2^\pm$  doublet in  $^{229}\text{Pa}$  was identified in Ref. [3] from two closed cycles of  $\gamma$  rays. One of the closed cycles,  $211.09 - (122.51 + 88.43)$ , cannot be used because the 211.06-keV  $\gamma$  ray is a doublet. The other closed cycle,  $241.84 - (122.52 + 119.26) = 60 \pm 50$  eV, still gives a positive number that makes  $5/2^-$  the  $^{229}\text{Pa}$  ground state. However, the large uncertainty in the energy difference makes the assignment of the  $5/2^-$  level uncertain.

“The state exists for sure!

I guarantee it!

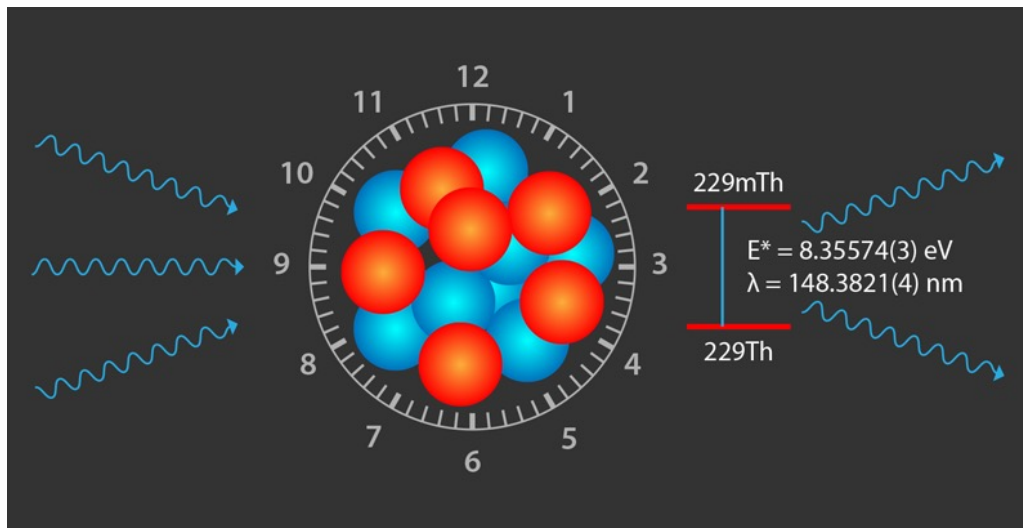
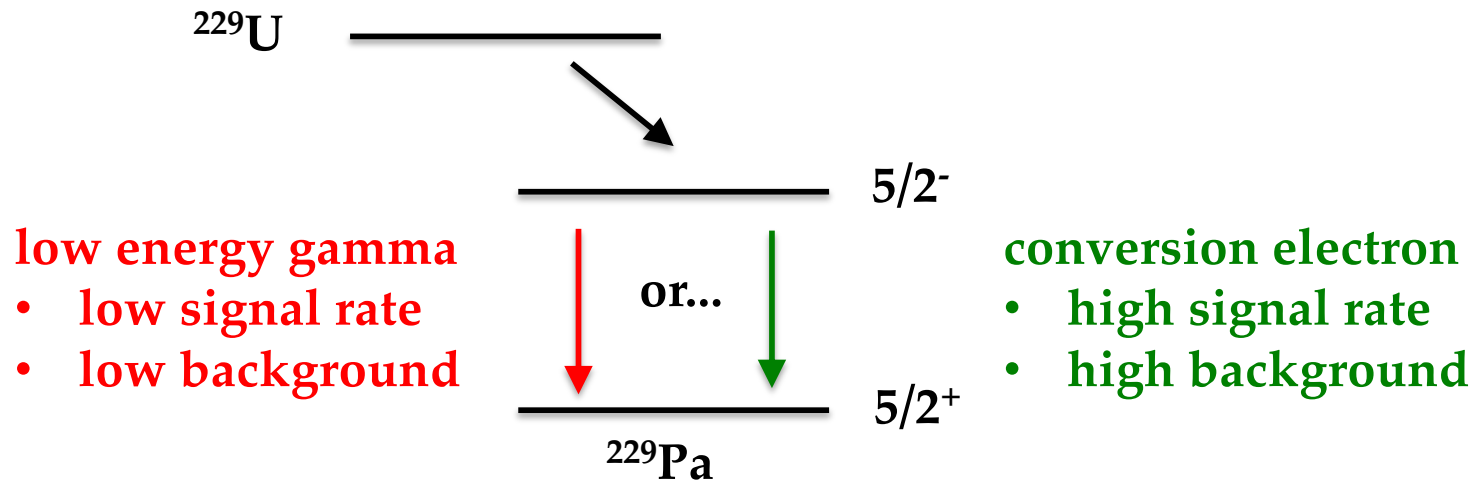
100%!

Just look for it now!”

Private Communication  
with IA (2014)

2015: Full reanalysis of 1982 ANL experiment + incorporation of data from German experiments: **Original claim of 220 eV conversion electrons is gone.**  
**If there is a ground state parity doublet, then it must be 60(50) eV.**  
PRC 92:024313 (2015)

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



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

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



# Low Energy Gamma Detectors (STJs) Now Exist!

We have used superconducting high-resolution radiation detectors to measure the energy level of metastable  $^{235m}\text{U}$  as  $76.737 \pm 0.018$  eV. The  $^{235m}\text{U}$  isomer is created from the  $\alpha$  decay of  $^{239}\text{Pu}$  and embedded directly into the detector. When the  $^{235m}\text{U}$  subsequently decays, the energy is fully contained within the detector and is

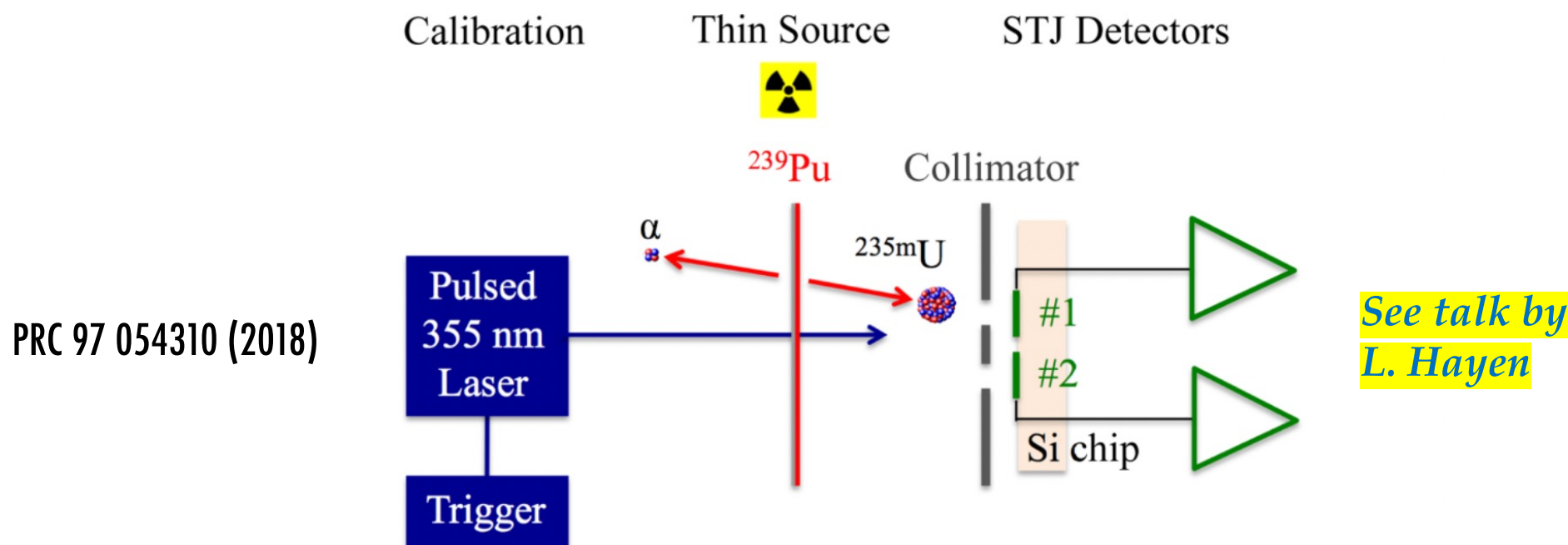
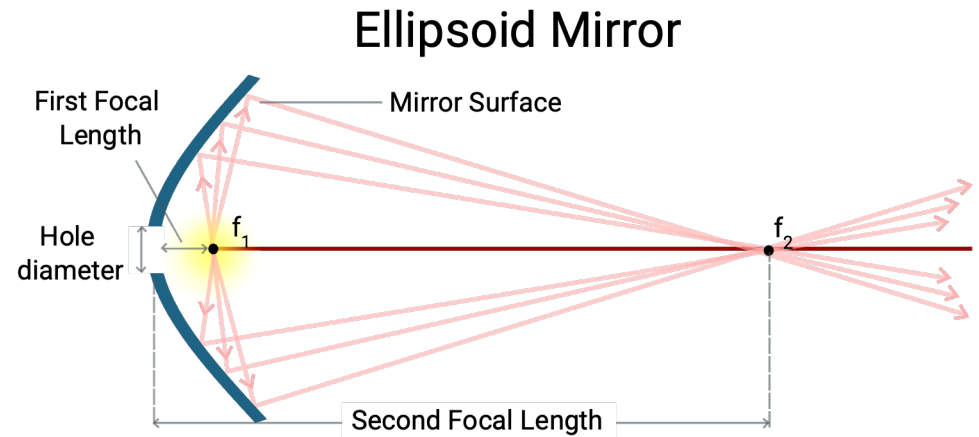
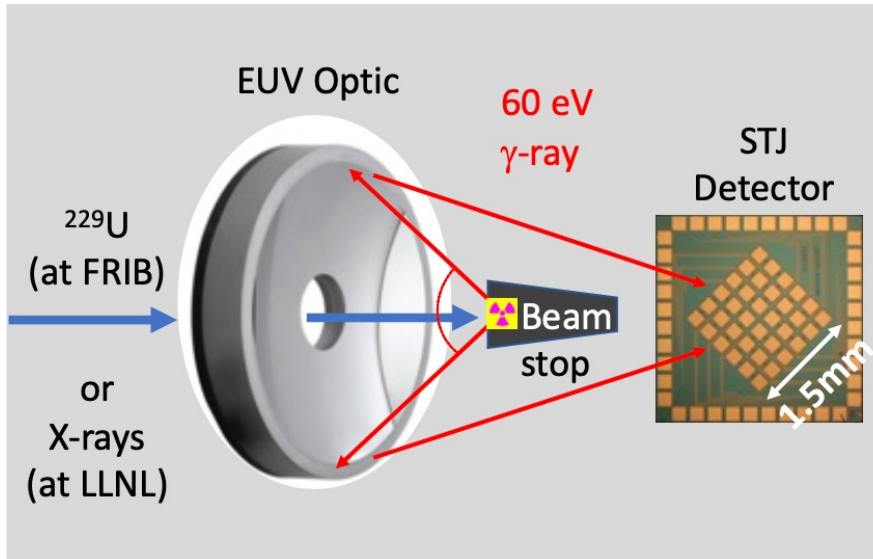
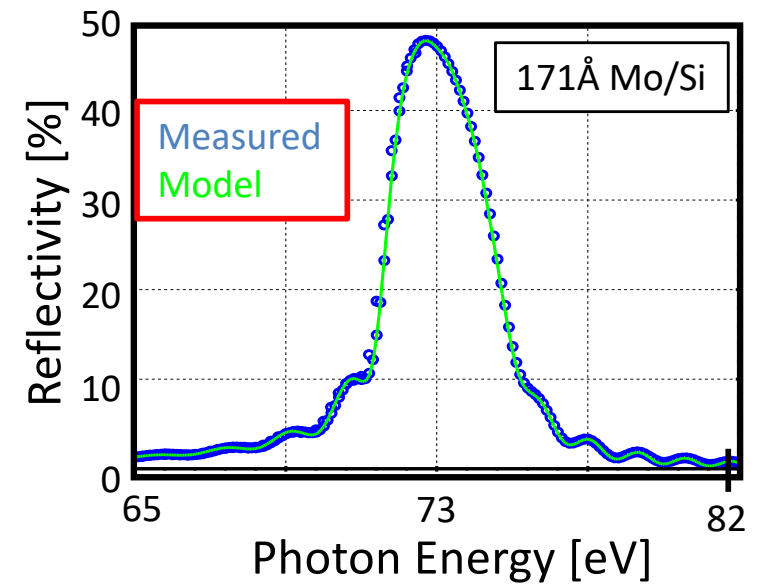
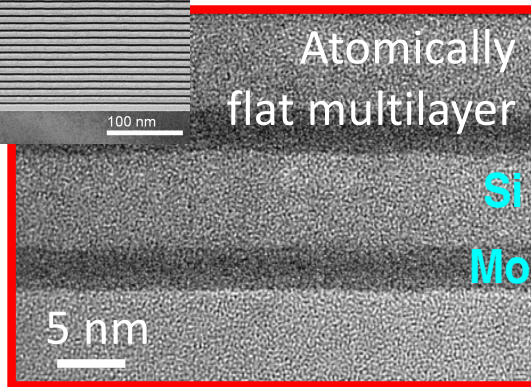
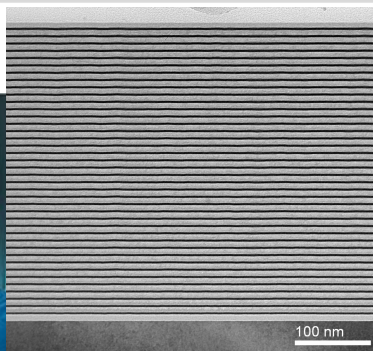
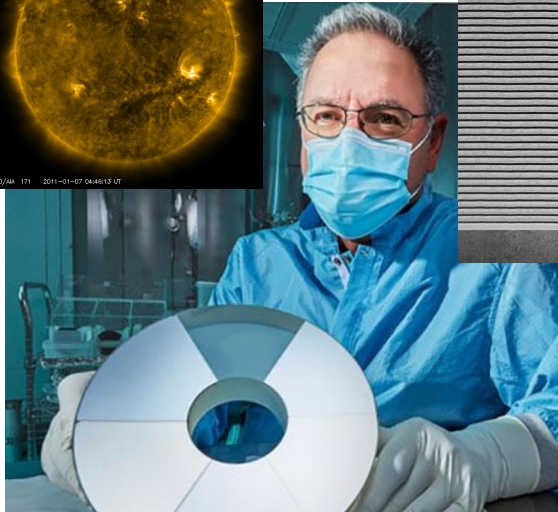
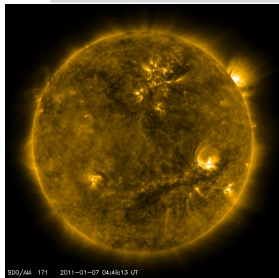


FIG. 1. Schematic of experimental setup:  $^{235m}\text{U}$  recoil ions produced by the decay of  $^{239}\text{Pu}$  are embedded in the STJ detectors, which measure their subsequent decay into the  $^{235}\text{U}$  ground state.

# Pending: Direct Gamma Detection (40-80 eV) w/ LLNL: Stephan Friedrich & Marie-Anne Descalle



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



# Pending: Direct Gamma Detection (40-80 eV) w/ LLNL: Stephan Friedrich & Marie-Anne Descalle

## Planned Work at LLNL:

1. Design EUV mirror
2. Fabricate mirror
3. Integrate mirror
4. Source: x-rays from Li K-shell electrons  
energy  $\sim 54$  eV
5. Source:  $^{239}\text{Pu}$  /  $^{235}\text{mU}$   
energy  $\sim 76$  eV
6. ship to FRIB

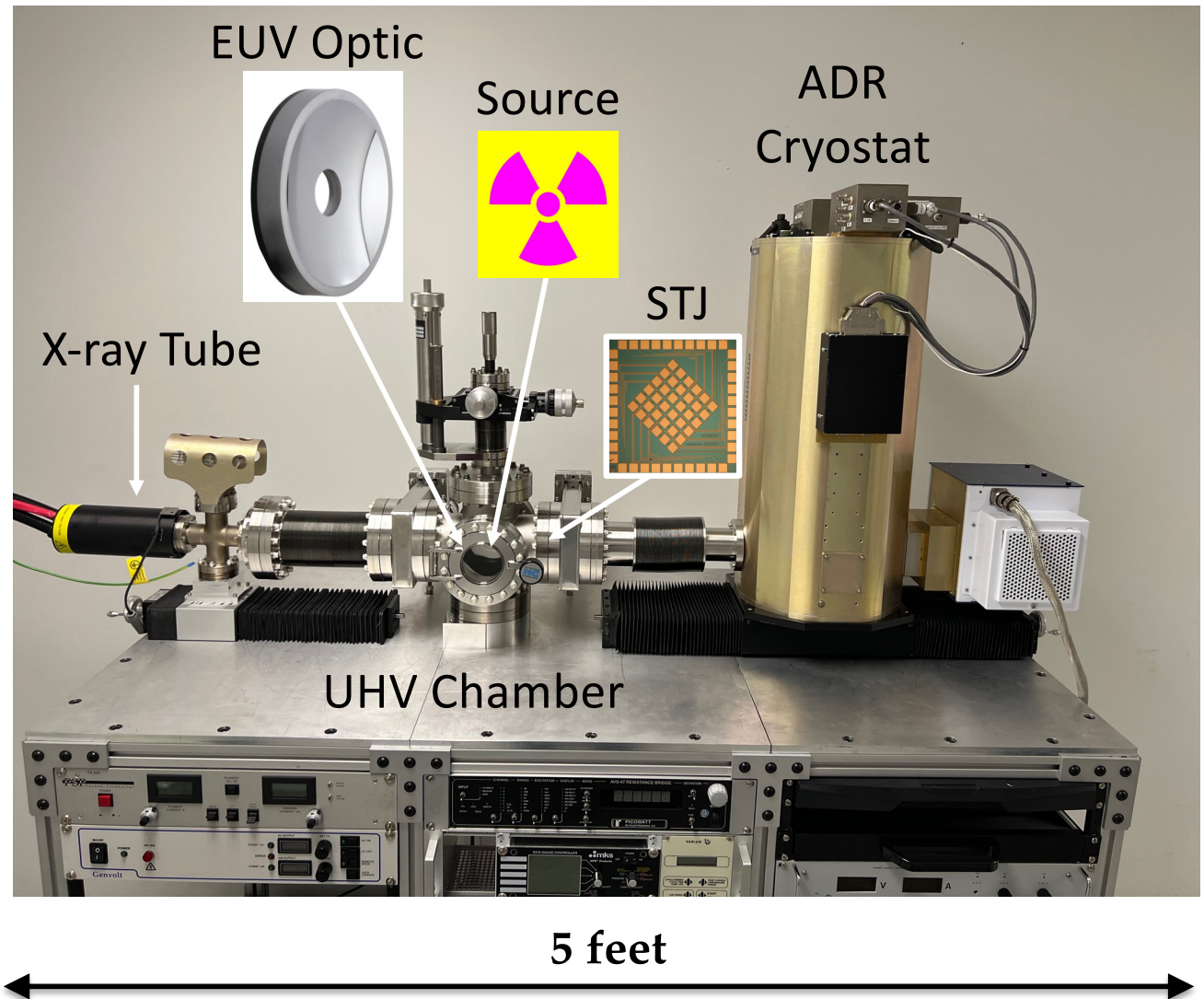
2023: LOI to FRIB PAC

2025-6: tests at LLNL

2023-7: GEANT simulations

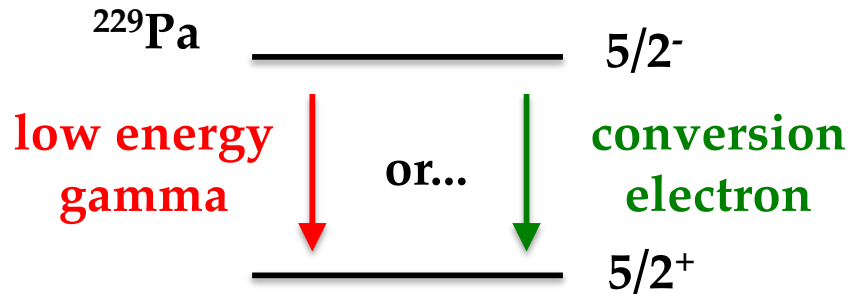
2027: submit PAC proposal

2027-8: run experiment





# The Internal Conversion Coefficient (ICC) is Very Large but Unknown

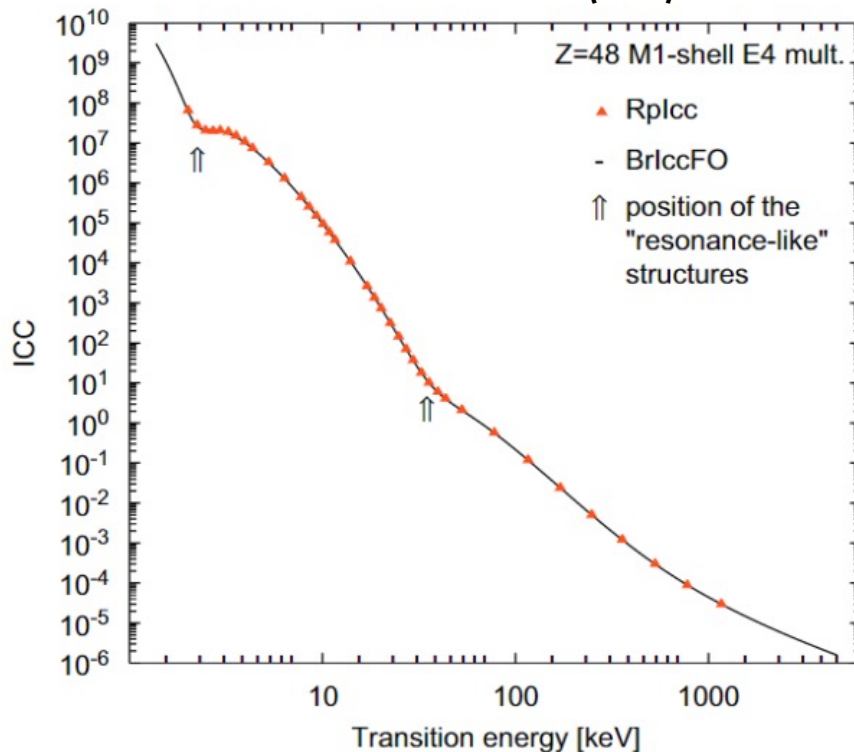


$$\frac{\text{electrons}}{\text{gammas}} \approx \alpha^4 Z^3 \left( \frac{L}{L+1} \right) \left( \frac{2m_e c^2}{\Delta E} \right)^{L+5/2} \approx 10^{18}$$

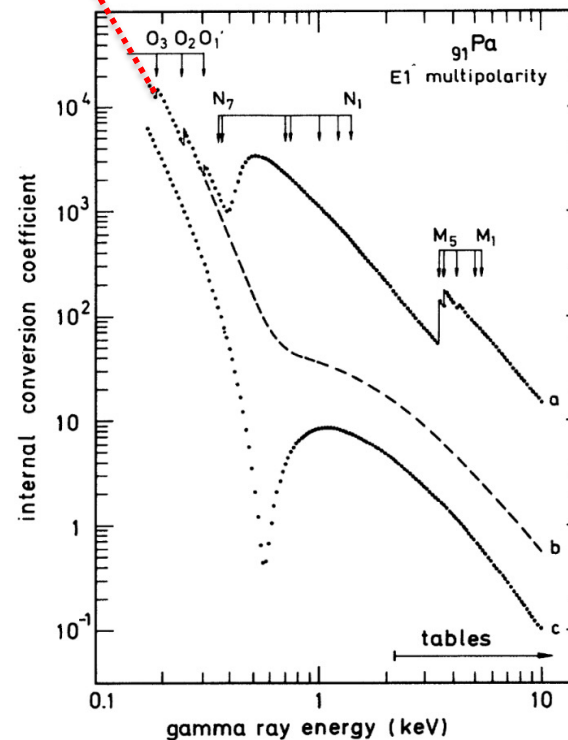
Introductory Nuclear Physics, 3<sup>rd</sup> (Krane)

10<sup>7</sup>?

BRICC: NIMA 589:202-229 (2008)



PRC 47(2):870-872 (1993)

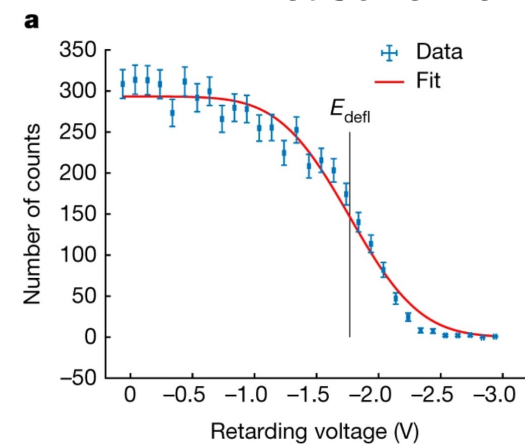
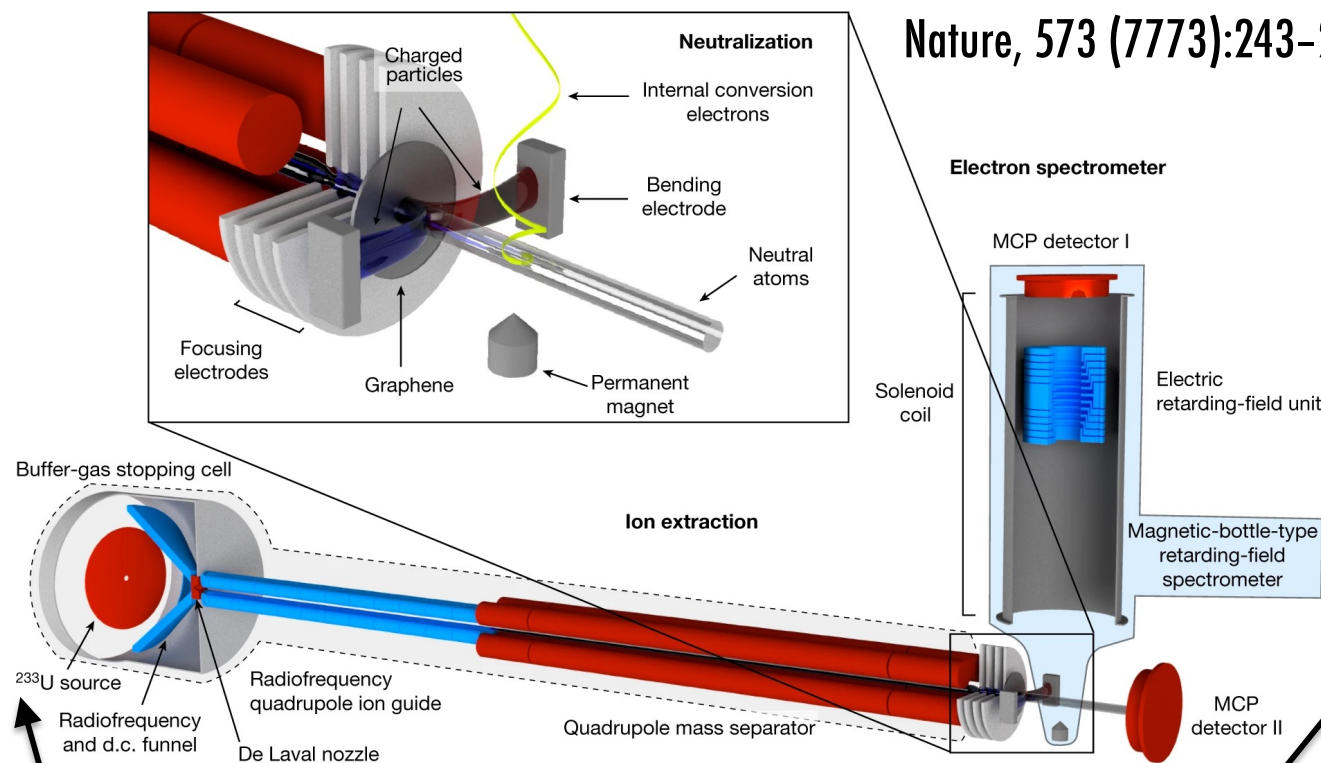


Planned experiments at LLNL using STJs with and without mirror using  $^{239}\text{Pu}/^{235\text{m}}\text{U}$  source will be used to experimentally measure the ICC for  $^{235\text{m}}\text{U}$  which is a proxy for  $^{229}\text{Pa}$ .

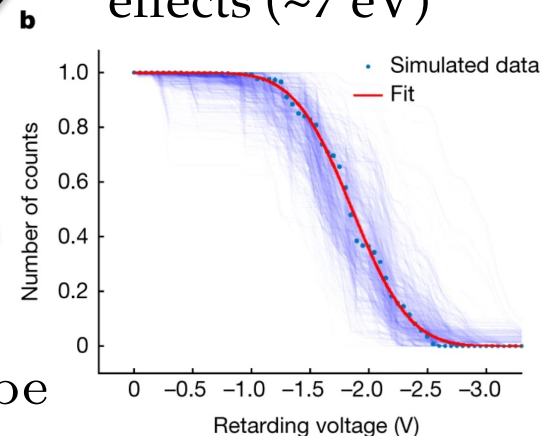
# Pending: The $^{229}\text{Th}$ Measurement Technique: Integrated Conversion Electron Spectroscopy

Nature, 573 (7773):243–246 (2019)

measurement



atomic binding  
effects ( $\sim 7$  eV)



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

We are just looking for nonzero counts and then the order of magnitude of  $\Delta E$  if a signal is observed. Unlike for  $^{229}\text{Th}$ , knowledge of atomic binding effects is not crucial.  
Main background: 8.36 eV from  $^{229}\text{Th}$ !

$^{229}\text{U}$  implanted from FRIB



# Embed and Probe $^{229}\text{Pa}$ Ions in Optical Crystals

- Large intrinsic sensitivity to BSM physics
  - **high  $Z$**  ( $^{199}\text{Hg}$ ,  $^{205}\text{Tl}$ ,  $^{225}\text{Ra}$ ,  $^{221,223}\text{Rn}$ ,  $^{229}\text{Pa}$ )
  - **octupole deformed nucleus** ( $^{225}\text{Ra}$ ,  $^{221,223}\text{Rn}$ ,  $^{229}\text{Pa}$ )
- Large  $E$ -field or  $B$ -field gradient to amplify observable
  - **local crystal fields (1-10 MV/cm) with large spin-orbit couplings (solids)**
- Repeat the measurement as many times as possible
  - ~~large number of nuclei (stable)~~
  - **long integration time (FRIB: steady supply for short  $\tau_{1/2}$ )**
  - **long trapping time: nuclei “stored” in the solid**
  - **long coherence time possible?**
- High efficiency extraction of experimental signal
  - **near unity capture and trapping efficiency in solid**
  - **optical detection via laser probing**
  - **optically-accessible nuclear spins?**
  - **inhomogenous broadening – address each nucleus individually?**

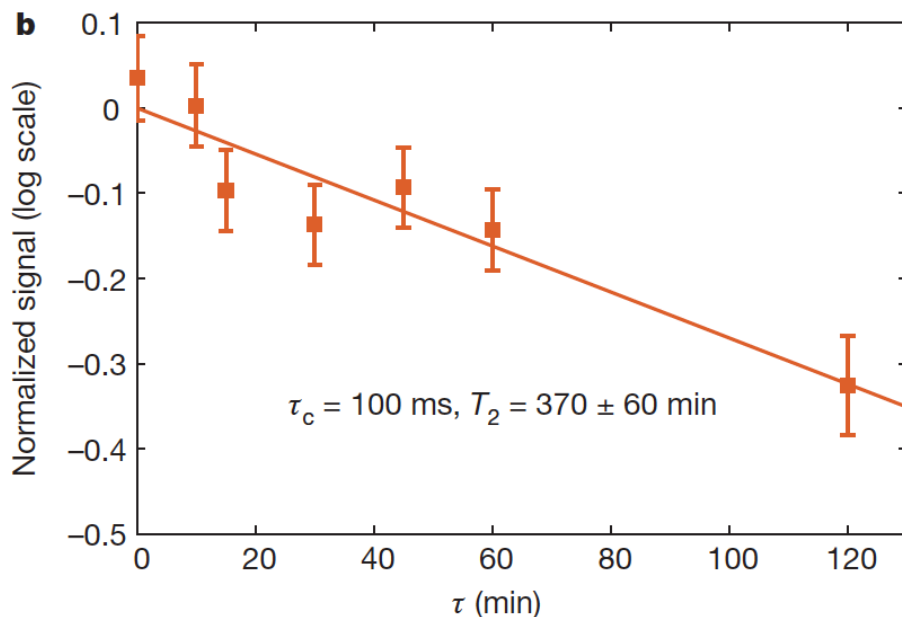
# Long Coherence Times of Lanthanide Ion Nuclei

doi:10.1038/nature14025

8 JANUARY 2015 | VOL 517 | NATURE | 177

## Optically addressable nuclear spins in a solid with a six-hour coherence time

Manjin Zhong<sup>1</sup>, Morgan P. Hedges<sup>1,2</sup>, Rose L. Ahlefeldt<sup>1,3</sup>, John G. Bartholomew<sup>1</sup>, Sarah E. Beavan<sup>1,4</sup>, Sven M. Wittig<sup>1,5</sup>, Jevon J. Longdell<sup>6</sup> & Matthew J. Sellars<sup>1</sup>



Under the right experimental conditions (magnetic field of 1.35 T and temperature of 2 K), using a specially designed pulse sequence (KDD<sub>x</sub>), **the  $T_2$  of  $^{151}\text{Eu}^{3+}$  ( $I=5/2$ ) embedded in  $\text{Y}_2\text{SiO}_5$  was measured to be over 6 hours.**

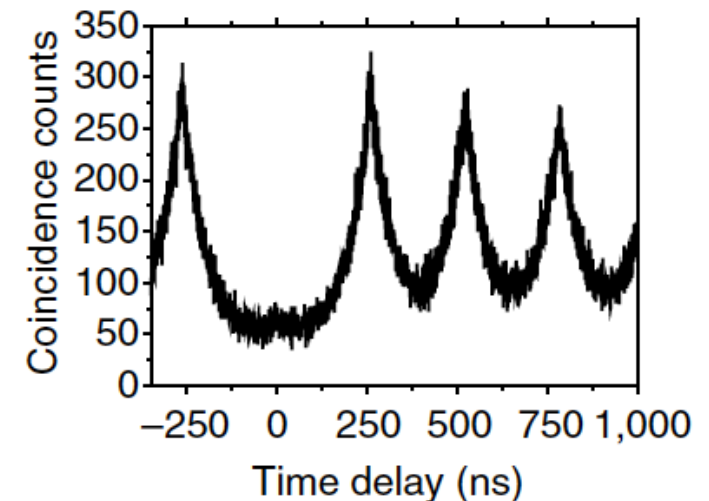
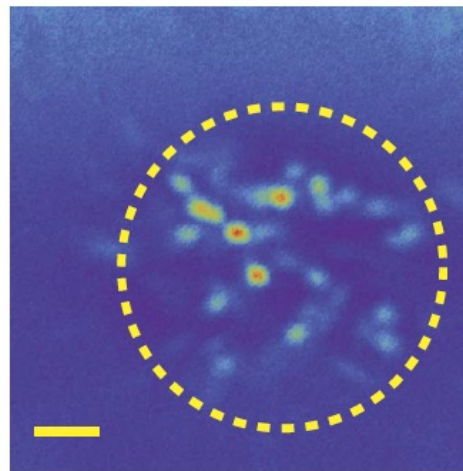
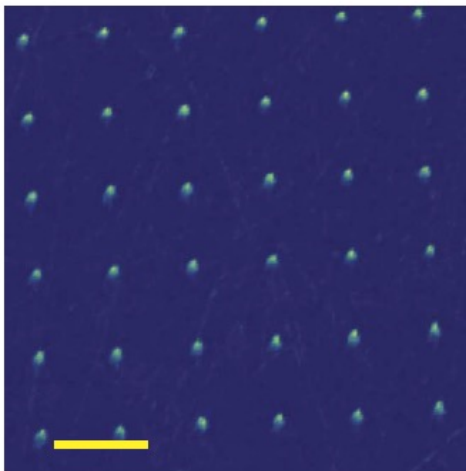
# Single Ion Implantation & Manipulation

Received 19 Jan 2014 | Accepted 15 Apr 2014 | Published 14 May 2014

DOI: 10.1038/ncomms4895

## Coherent properties of single rare-earth spin qubits

P. Siyushev<sup>1,\*</sup>, K. Xia<sup>1,\*</sup>, R. Reuter<sup>1</sup>, M. Jamali<sup>1</sup>, N. Zhao<sup>2</sup>, N. Yang<sup>3</sup>, C. Duan<sup>4</sup>, N. Kukharchyk<sup>5</sup>,  
A.D. Wieck<sup>5</sup>, R. Kolesov<sup>1</sup> & J. Wrachtrup<sup>1</sup>



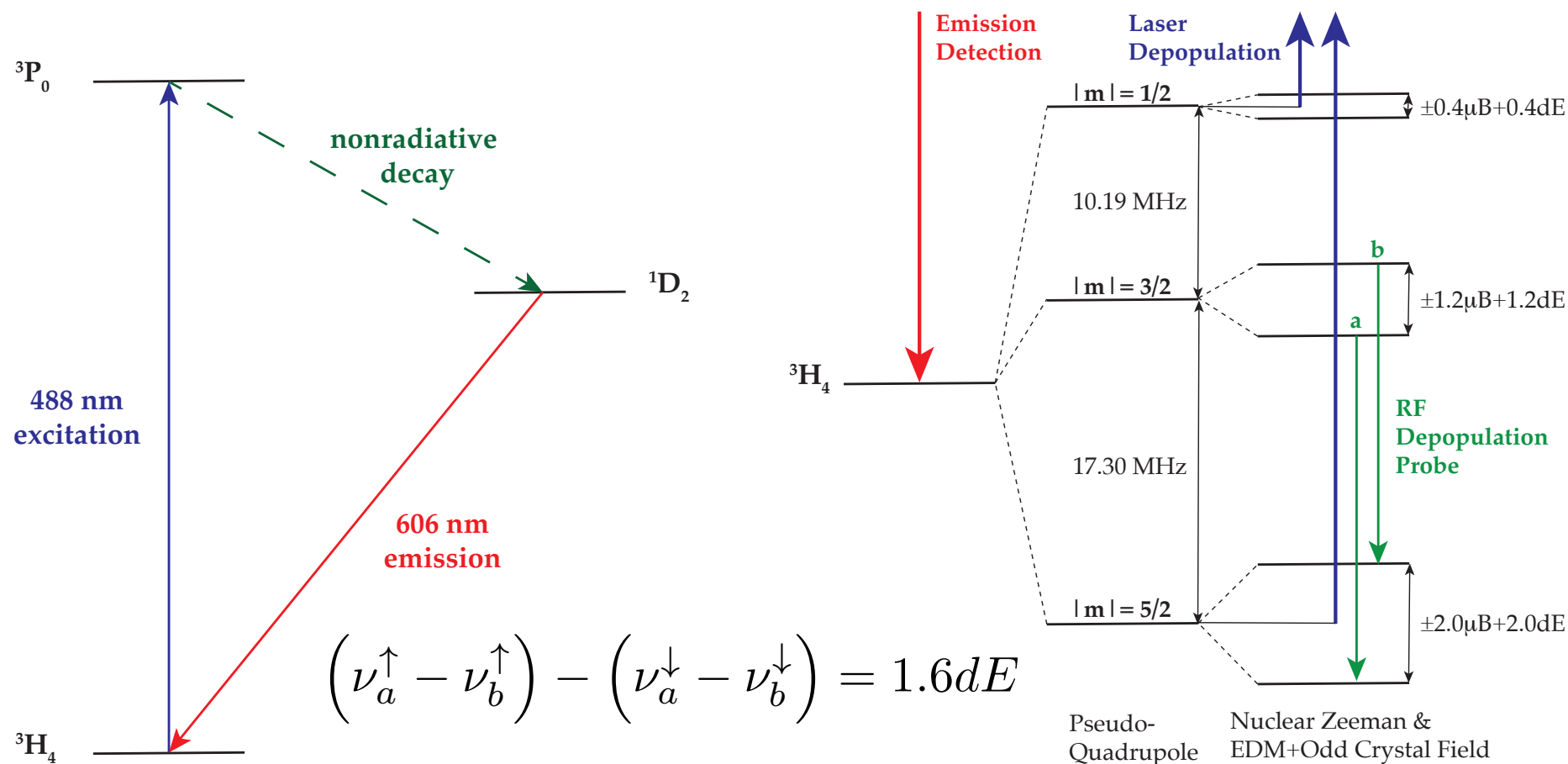
Left: well-controlled ion implantation of  $\text{Ce}^{3+}$  (yellow bar = 10 microns)

Middle: individual Ce site (yellow bar = 2 microns)

Right: antibunching in photon correlation data indicates single emitter

# Pending: Pathfinder $^{141}\text{Pr}^{3+}$ ( $I=5/2$ ) in YSO or Diamonds

## Jonas Becker (MSU-Physics) & Shannon Nicley (MSU-ECE)



$$\left(\nu_a^\uparrow - \nu_b^\uparrow\right) - \left(\nu_a^\downarrow - \nu_b^\downarrow\right) = 1.6dE$$

- Optically-detected shift in Larmor frequency with respect to two ensembles
- $^{229}\text{Pa}$  frequency sensitivity needed to match  $^{199}\text{Hg}$  New Physics sensitivity: 30 mHz per shot
- Integrated frequency sensitivity over 1.5 days ( $^{229}\text{Pa}$  half-life): 1 mHz

Hyp. Int. 240:29 (2019) & Phil. Trans. R. Soc. A.382: 20230169 (2024)

# 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

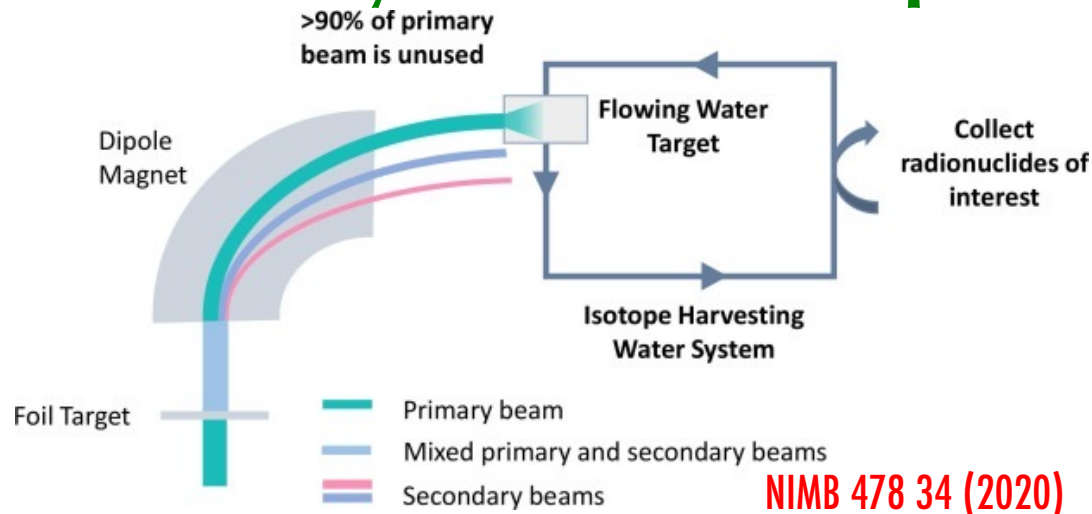
University of Michigan  
Ann Arbor, MI  
Very Good at American Football  
no FRIB



Google Maps & Wikipedia Commons



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



NIMB 478 34 (2020)



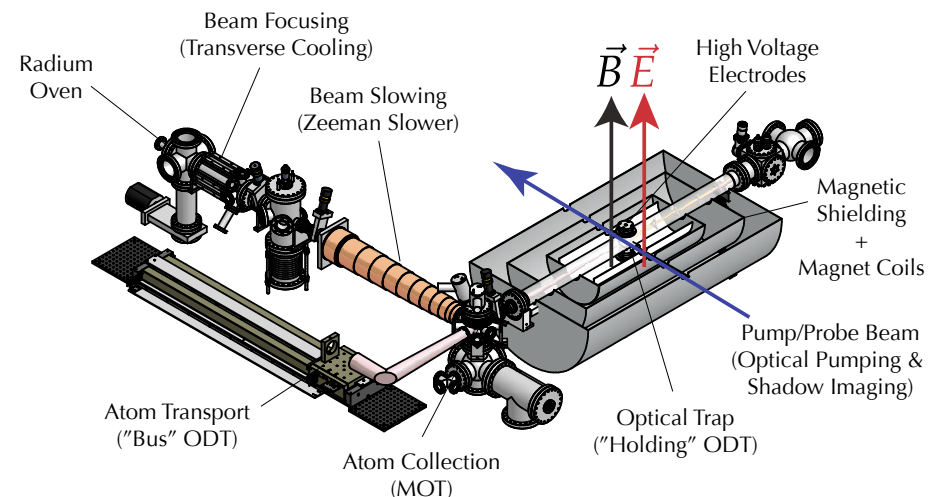
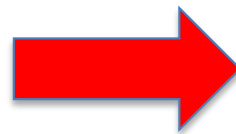
Adobe Stock

Isotope harvesting at FRIB: additional opportunities for scientific discovery [J. Phys. G: Nucl. Part. Phys. 46 100501 (2019)]

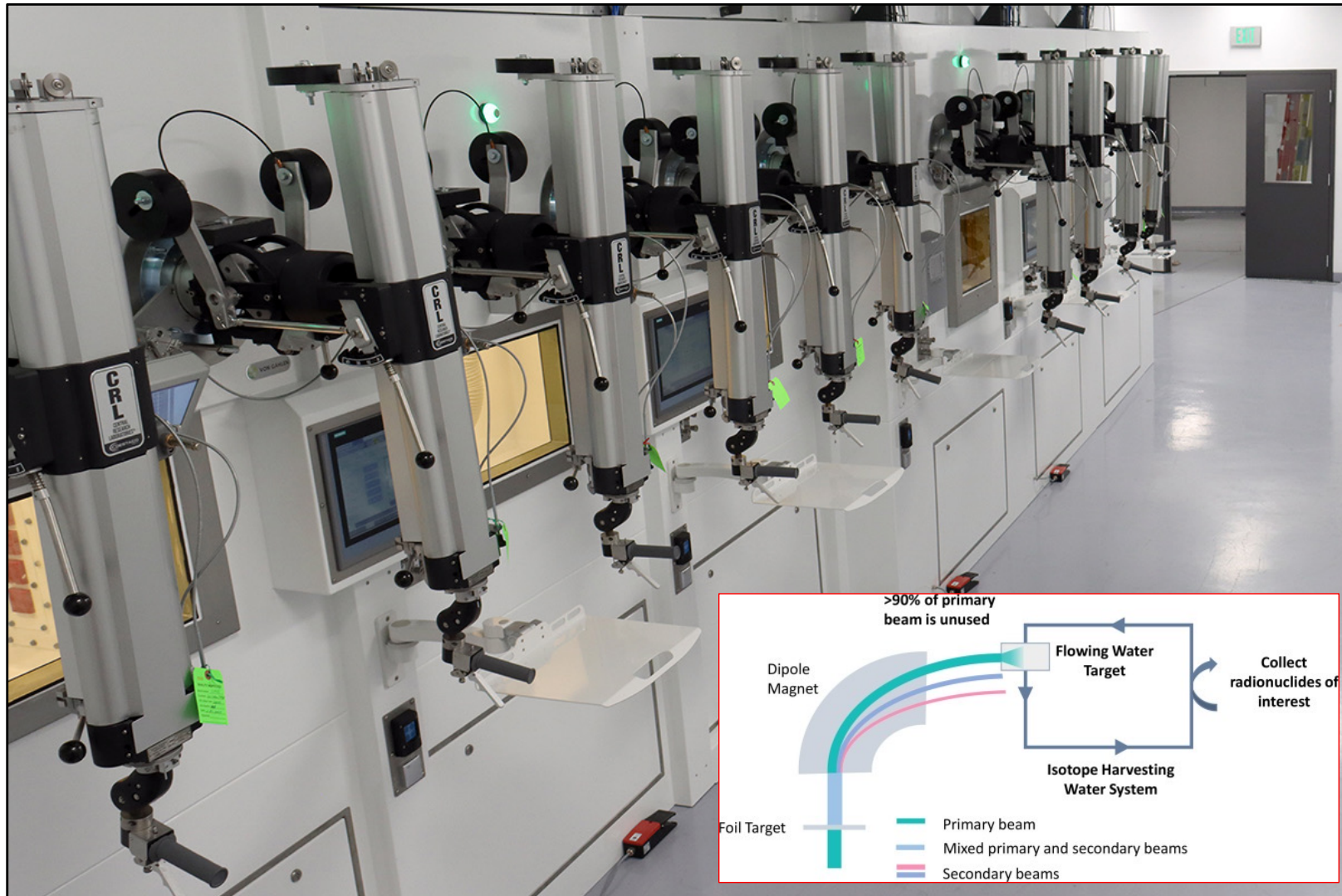


Adobe Stock

see talks by  
G. Arrowsmith-Kron  
A. Boyer



# Isotope Harvesting Vault Is Installed



30 feet

NIMB 478 34 (2020)

C. Vyas



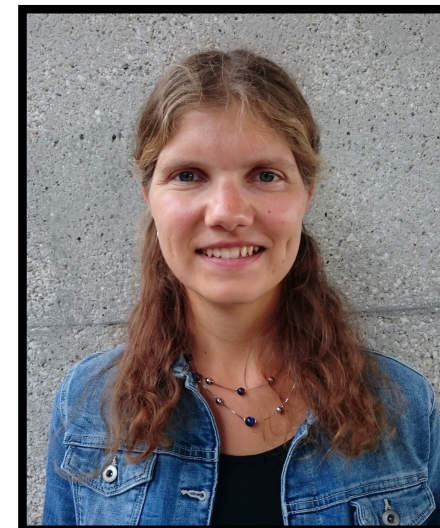
# The Radiochemistry Team at FRIB/MSU



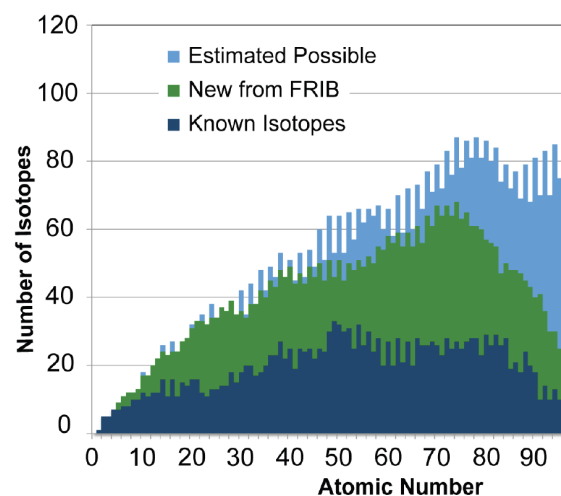
Prof. Greg Severin



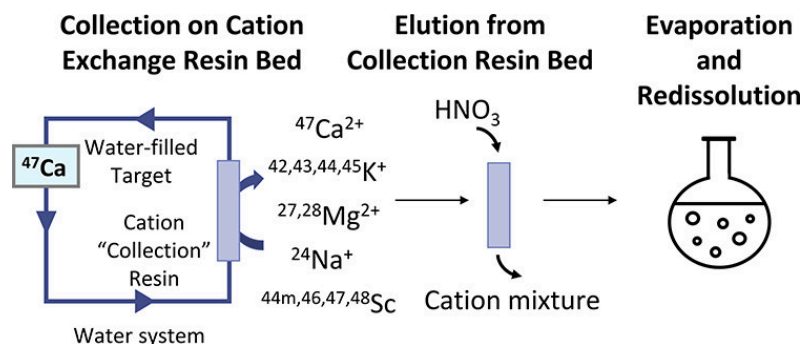
Prof. Alyssa Gaiser



Prof. Katharina Domnanich



Nature 486, 509–512 (2012)



Recovery of 92%  
to 99%  
of  $^{47}\text{Ca}$   
(surrogate for  
Radium)

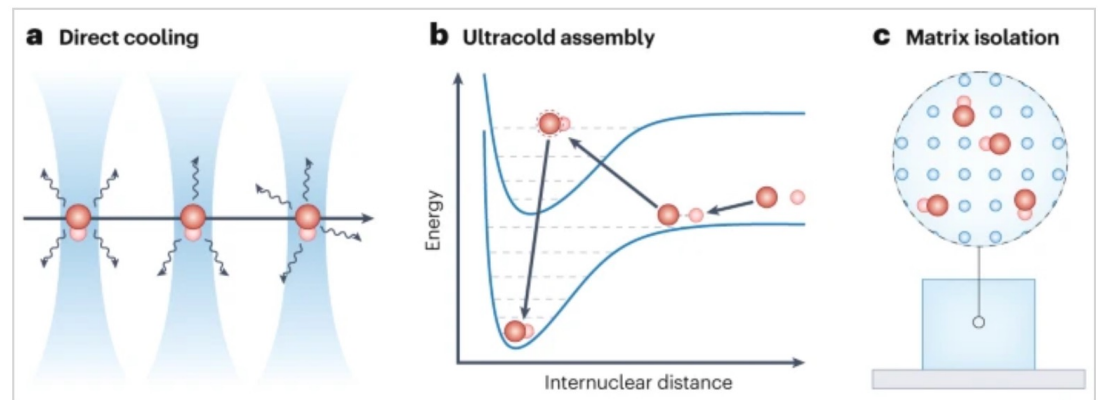
Abel et al., ACS Omega 5(43) 27864 (2020)

# Heavy (Rare) Pear-Shaped Nuclei Inside Molecules/Solids

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

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

**Opportunity:**  
Isotope harvesting @ FRIB:  
from “Beam to Beaker”  
( $^{225}\text{Ra}$ ,  $^{229}\text{Pa}$ , ...)



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:

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

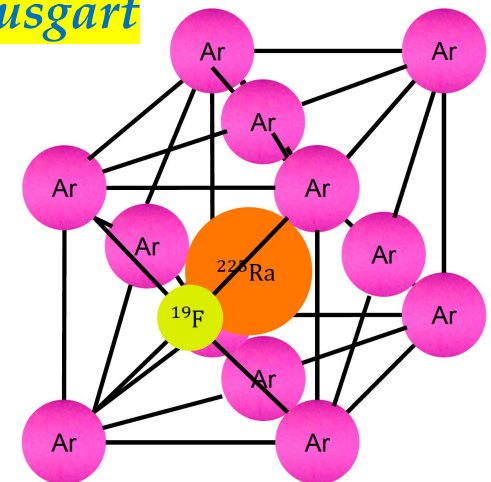
- Efficient trapping of a wide variety of species
- Very high number densities
- Stable and chemically inert confinement
- Transparent in the optical regime for optical probing
- Under certain conditions, polar molecules orient themselves along the crystal axes which allows for control of systematics: **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)**

see talks by

**K. Martirosova**

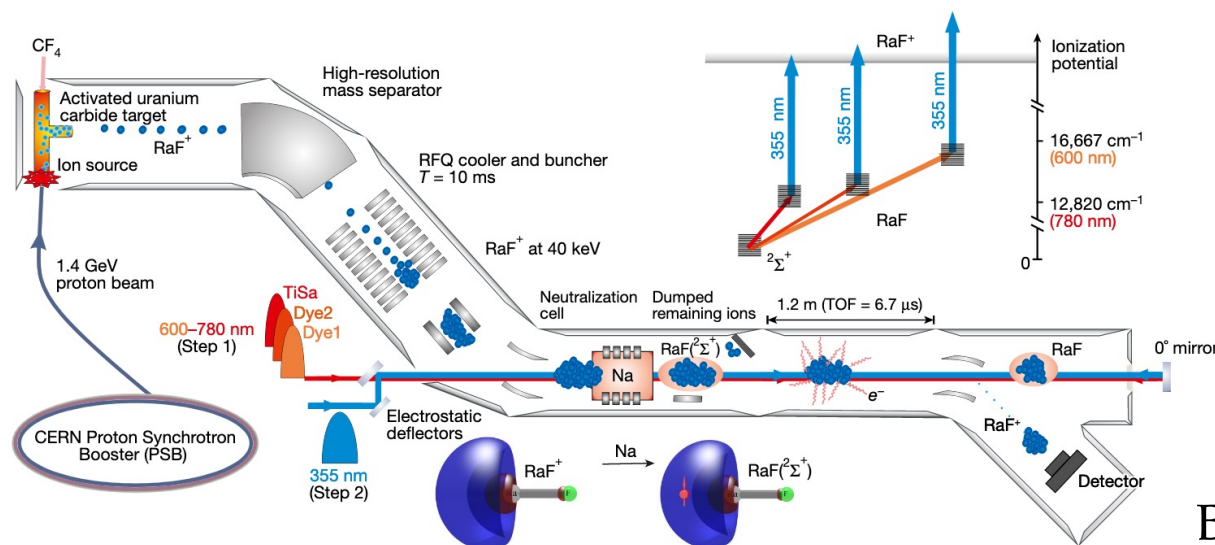
**A. Boyer**

**N. Nuscgart**



# Direct Laser Cooling of Neutral Molecules Into a Laser Trap

## QUESTLab (Xing Wu/FRIB) & RaX (MIT/Caltech/Harvard)



- Molecular spectroscopy of RaF is underway!
- RaF is the most laser coolable diatomic molecule!

**Nature 581:396 (2020)**

**PRL 127:033001 (2021)**

**R. Garcia Ruiz (MIT)**

### Benefits of Polyatomic Molecules

- Laser coolable & trappable
- Highly polarizable
- Comagnetometer states for control of systematics
- High CPV sensitivity
- Laser Cooling of  $\text{CaOCH}_3$

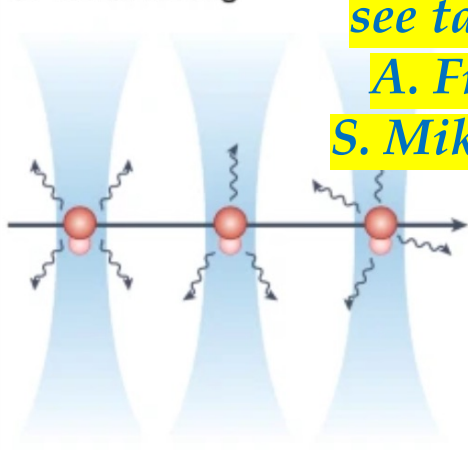
**PRL 119, 133002 (2017)**

**Quantum Science & Tech. 5, 044011 (2020)**

**Science 369, 1366–1369 (2020)**

**Nature Physics 20, p741–749 (2024)**

### a Direct cooling



*see talks by*

*A. Frenett*

*S. Miki-Silva*

**N. Hutzler (Caltech) & J. Doyle (Harvard)**



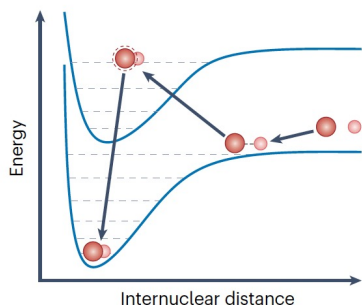
# Ultracold Assembly of Neutral Molecules Within A Laser Trap: $^{223}\text{FrAg}$ ( $\tau_{1/2} = 22$ minutes)

## Gen-I Estimate:

$\Rightarrow$  ~1000x projected improvement  
vs.  $^{199}\text{Hg}$  state of the art

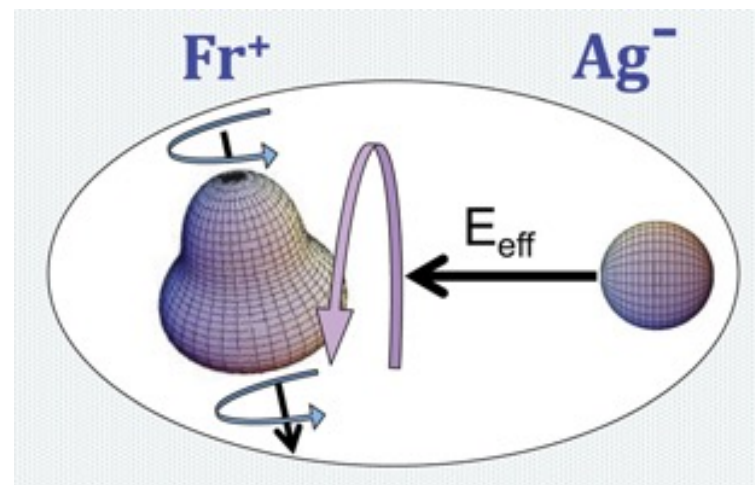
Needs major involvement of  
radiochemists,  
thermal ion beam source experts,  
radiological safety experts, ...  
to develop  $^{223}\text{Fr}^+$  ion source

**b** Ultracold assembly



Nature Physics 20, p741–749 (2024)

slide from D. DeMille  
(UChicago/Argonne)



All these parameters  
**ALREADY DEMONSTRATED**  
with stable bi-alkalis (!)

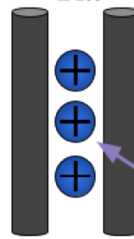
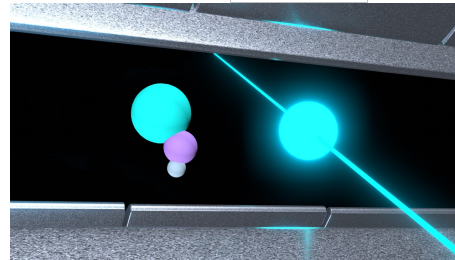
Theory calculations favorable:

New J. Phys. 23 113039 (2021)

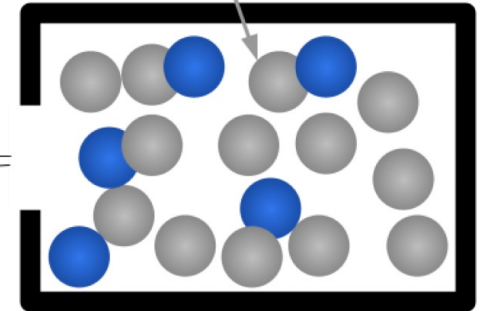
New J. Phys. 24 025005 (2022)

odd-proton nuclei like  $^{223}\text{Fr}$  probe  
largely orthogonal parameter  
space vs. odd-neutron species

# Quantum Logic Spectroscopy of Single Molecular Ions: $^{225}\text{RaOH}^+$ , $^{225}\text{RaSH}^+$ , & $^{225}\text{RaOCH}_3^+$ ( $\tau_{1/2} = 15$ days)



**solid thorium**



- Spectroscopy and atomic structure measurements of the logic ion  $\text{Ra}^+$   
PRL 122, 223001 (2019), PRA 100, 062512 (2019), PRA 100, 062504 (2019), PRA 102, 042822 (2020)  
PRA 105, 042801 (2022)

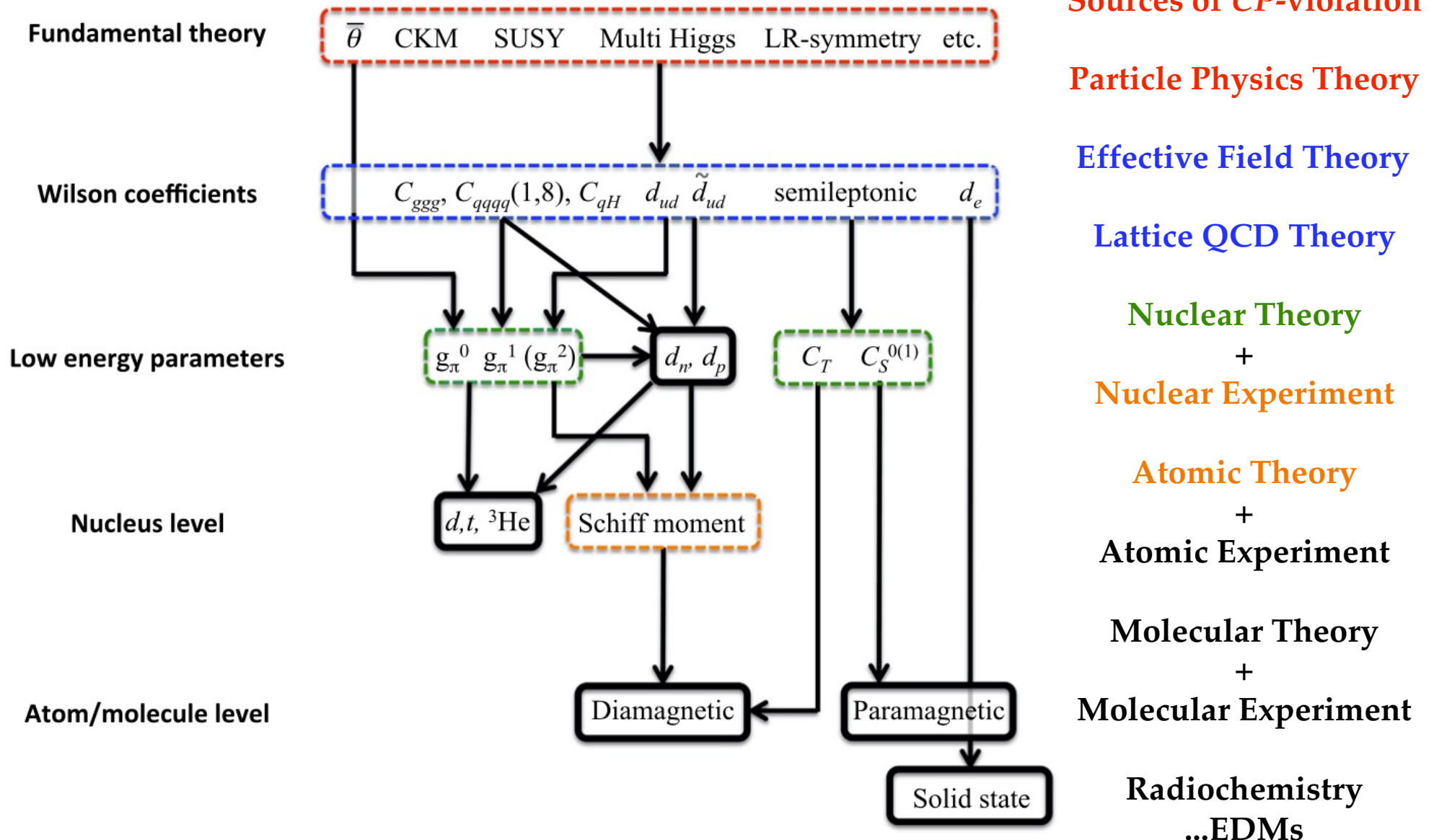
- Formation of relevant CPV-sensitive single molecular ions  
PRL 126, 023002 (2021)

- Identification of candidate molecular ions with pear-shaped nuclei with enhanced CPV sensitivity  
**PRL 126, 023003 (2021)** **slide from A. Jayich (UC Santa Barbara) DOE ECA 2021**

63

# Connecting New Physics to EDMs

T.E. Chupp, P. Fierlinger, M. Ramsey-Musolf, JTS, RMP 91:015001

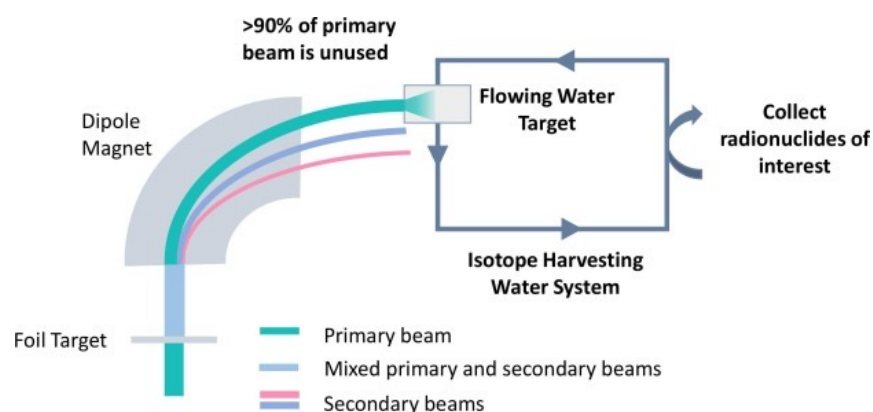


# The Nuclear Pear Factory: A Proposed Center

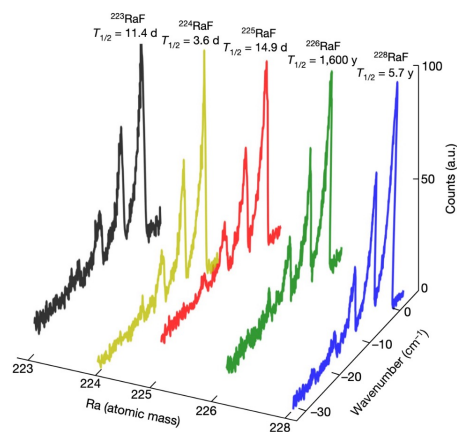


Nature 497:199 (2013)

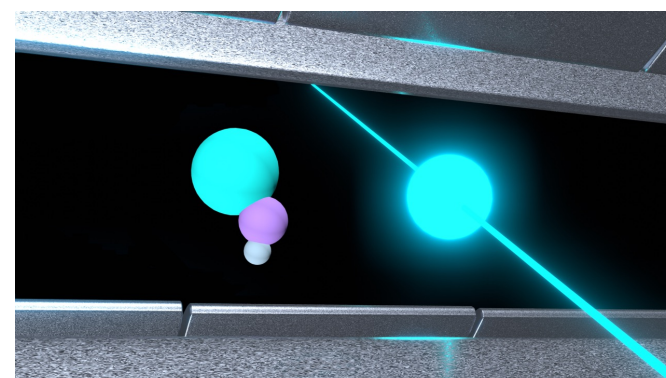
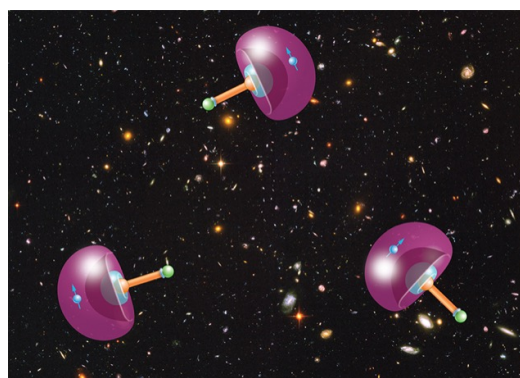
A joint Experiment/Theory & AMO/Nuclear/Radiochemistry effort to calibrate the new physics sensitivity of pear-shaped nuclei and to carry out the requisite precursory work leading to ultrasensitive EDM searches.



NIMB 478 34 (2020)



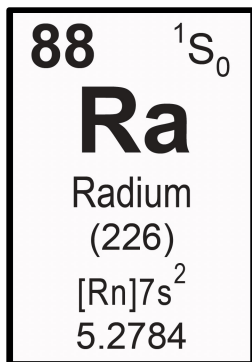
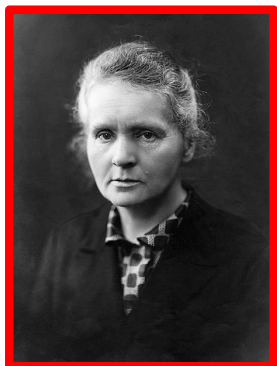
Nature 581:396 (2020)



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



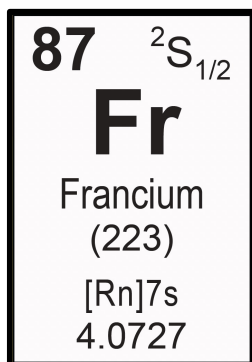
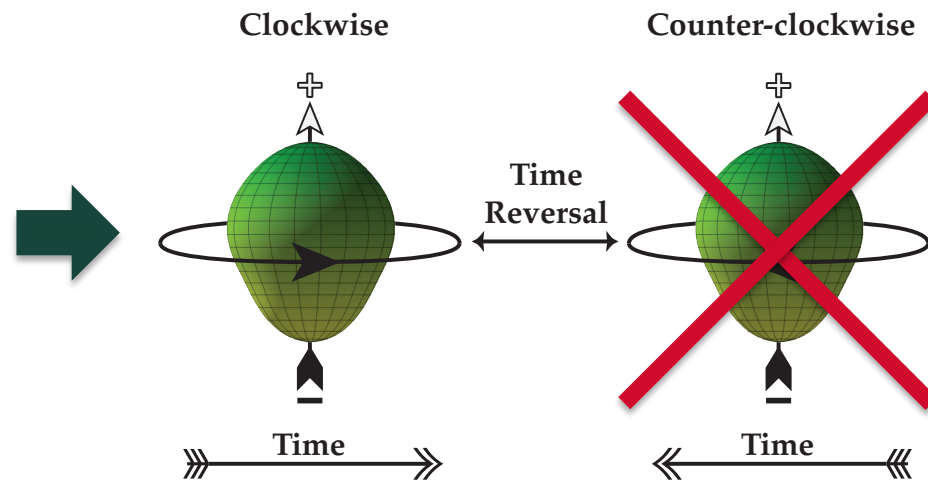
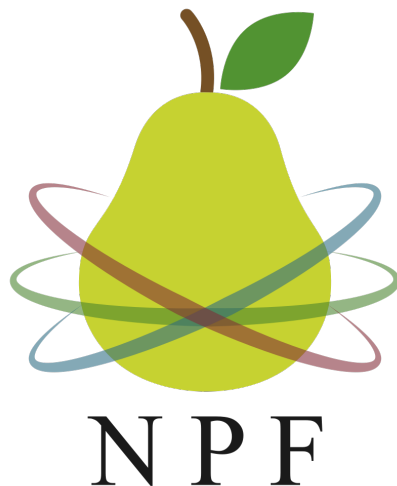
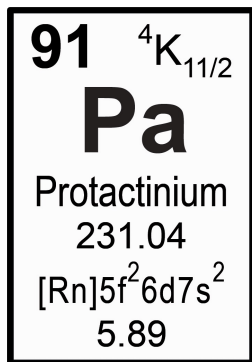
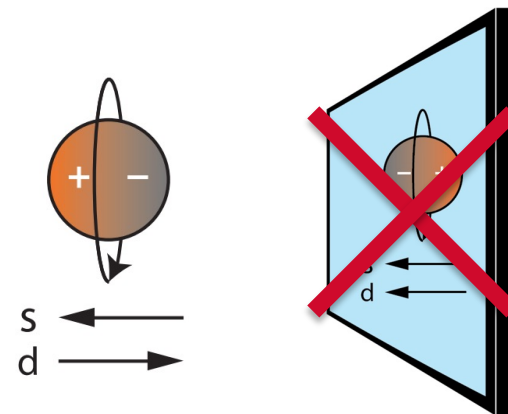
# We Are Following In The Footsteps Of Giants Towards A Transformational Discovery Within Our Student's Lifetime!



C.S. Wu



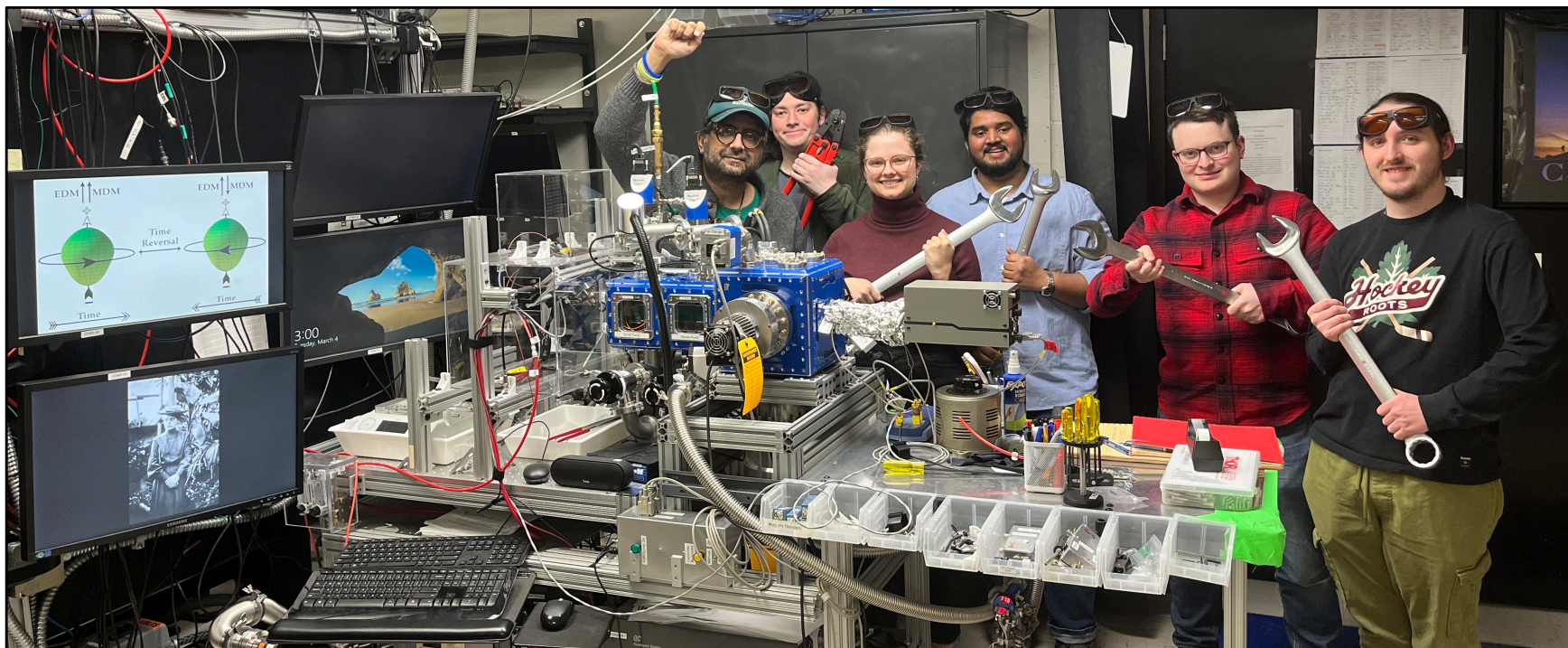
M.A. Bouchiat



**M. Curie (2/5), L. Meitner (0/49), & M. Perey (0/5)**

Wikipedia, NIST, AIP Emilio Segre Visual Archives, M. Zolotrev

# Thanks For Your Attention! More Questions?



U.S. DEPARTMENT  
*of* ENERGY

DE-SC0019015 (ECA-EDM3)

DE-SC0019455 (Ra EDM)

DE-NA0003996 (Pa-229/SAM)

DE-SC0025679 (Ra EDM+EDM3)



# 1654610  
(CAREER-SAM)  
# 2412951 (SAM)

GORDON AND BETTY  
**MOORE**  
FOUNDATION



E. A. Hessels  
@ York (Canada):  
GBMF8863  
G-2019-12503