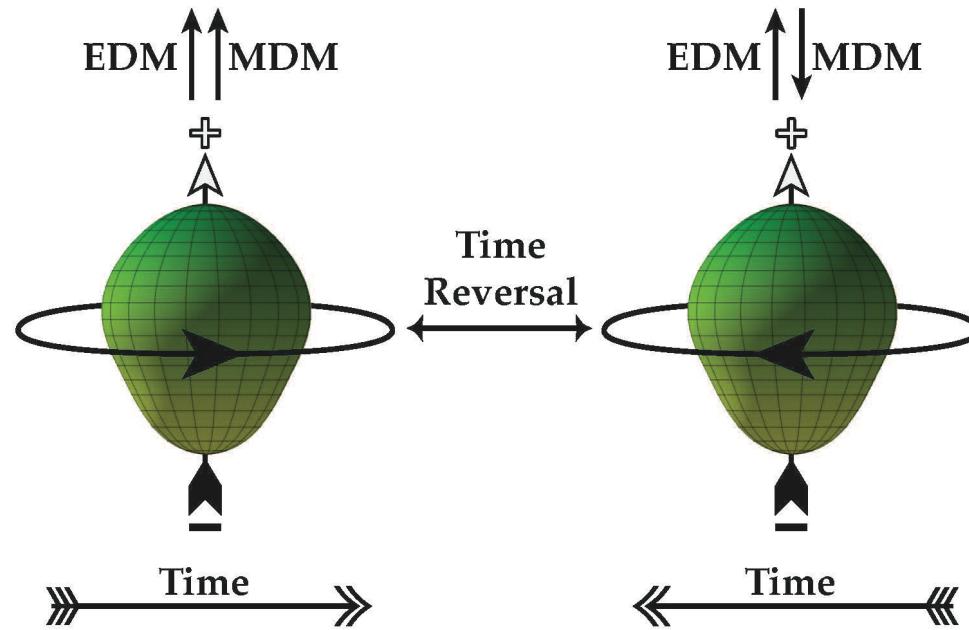


Prospects for Determining If ^{229}Pa Is the Most Pear-Shaped Nucleus



Lise
Meitner

Jaideep Taggart Singh (he/him/his)

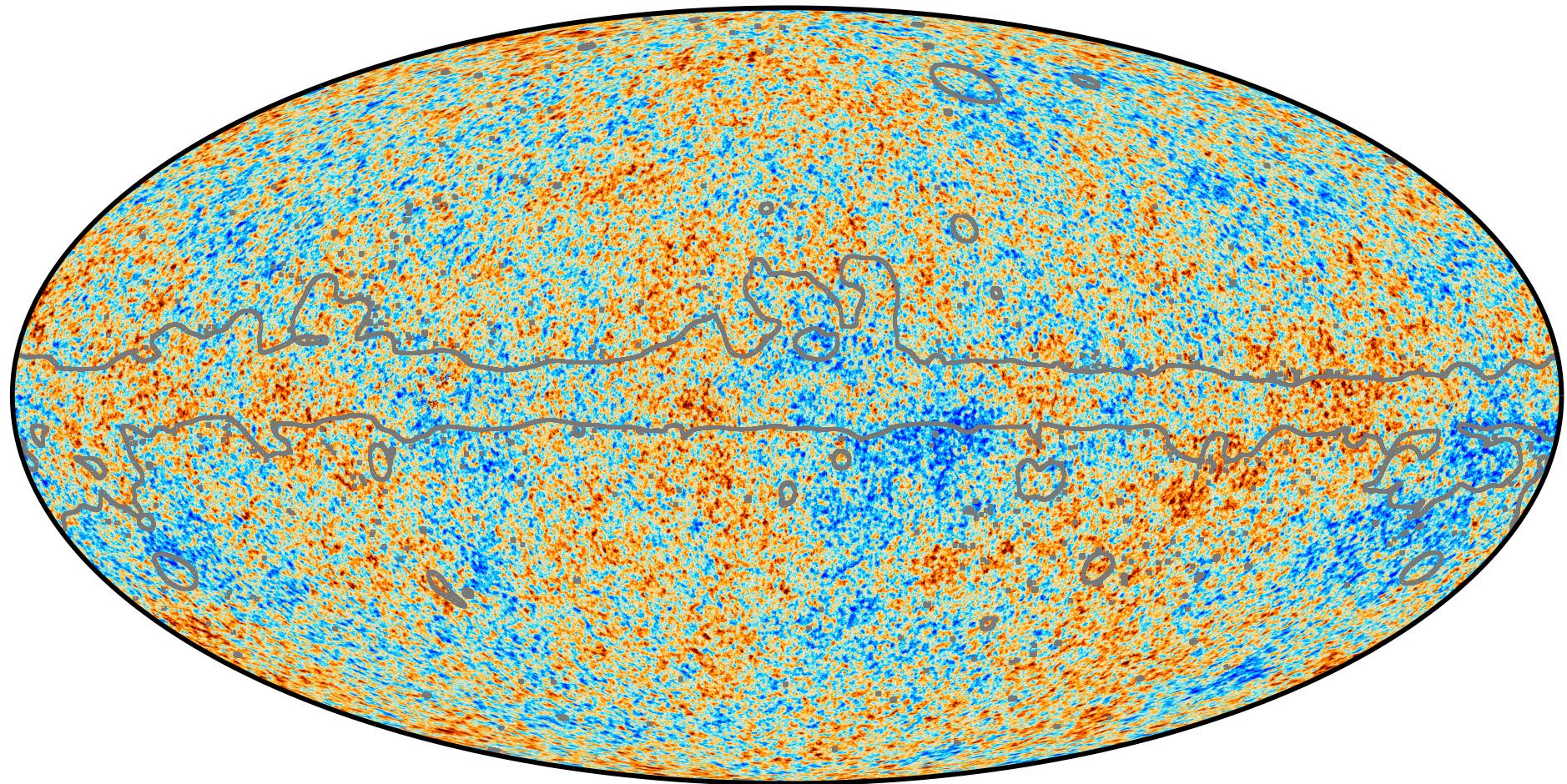
Michigan State University / FRIB

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

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



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



Planck 2018

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

-300  300 μK

~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

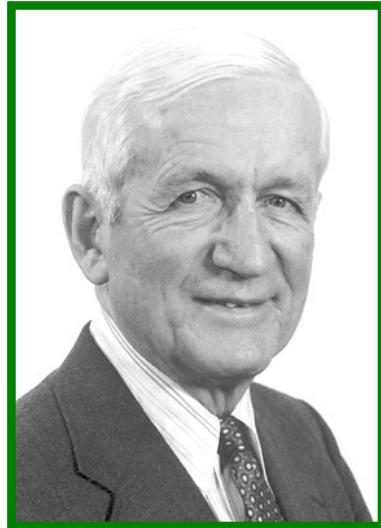
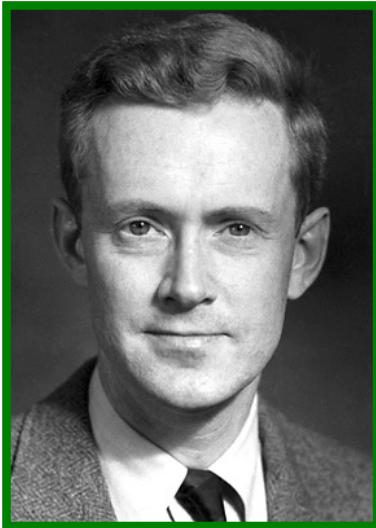
ZhETF Pis'ma 5, No. 1, 32-35, 1 January 1967

The theory of the expanding Universe, which presupposes a superdense initial state of matter, apparently excludes the possibility of macroscopic separation of matter from antimatter; 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

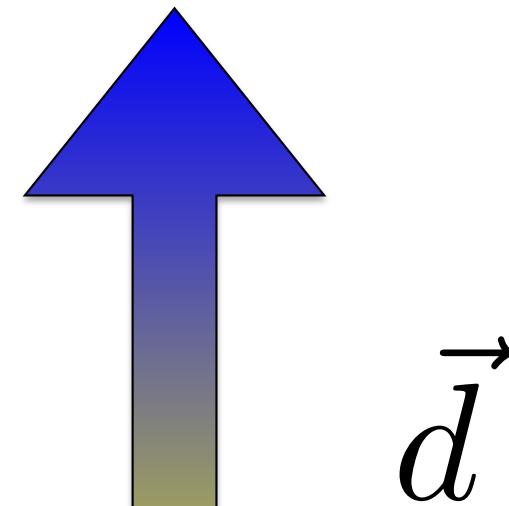


The Nobel Foundation

- 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 + $0\nu2\beta$]
- *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

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



ρ_{charge} = charge distribution

\vec{r} = position vector

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

Q = net charge

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

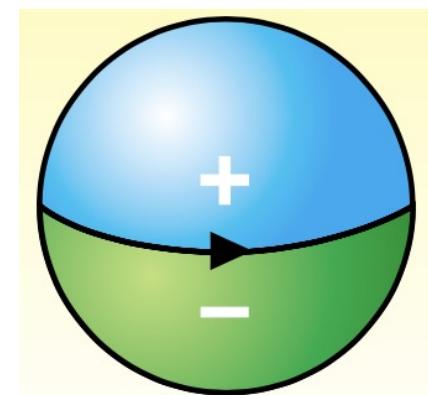
2023 EDM Limits: “Free” of Standard Model (SM) “Backgrounds”

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 ⁺ (JILA/Boulder)
¹⁹⁹ Hg	0.074	$\sim 10^{-6}$	atoms in vapor cell (UW-Seattle)

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

...if the ¹⁹⁹Hg 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

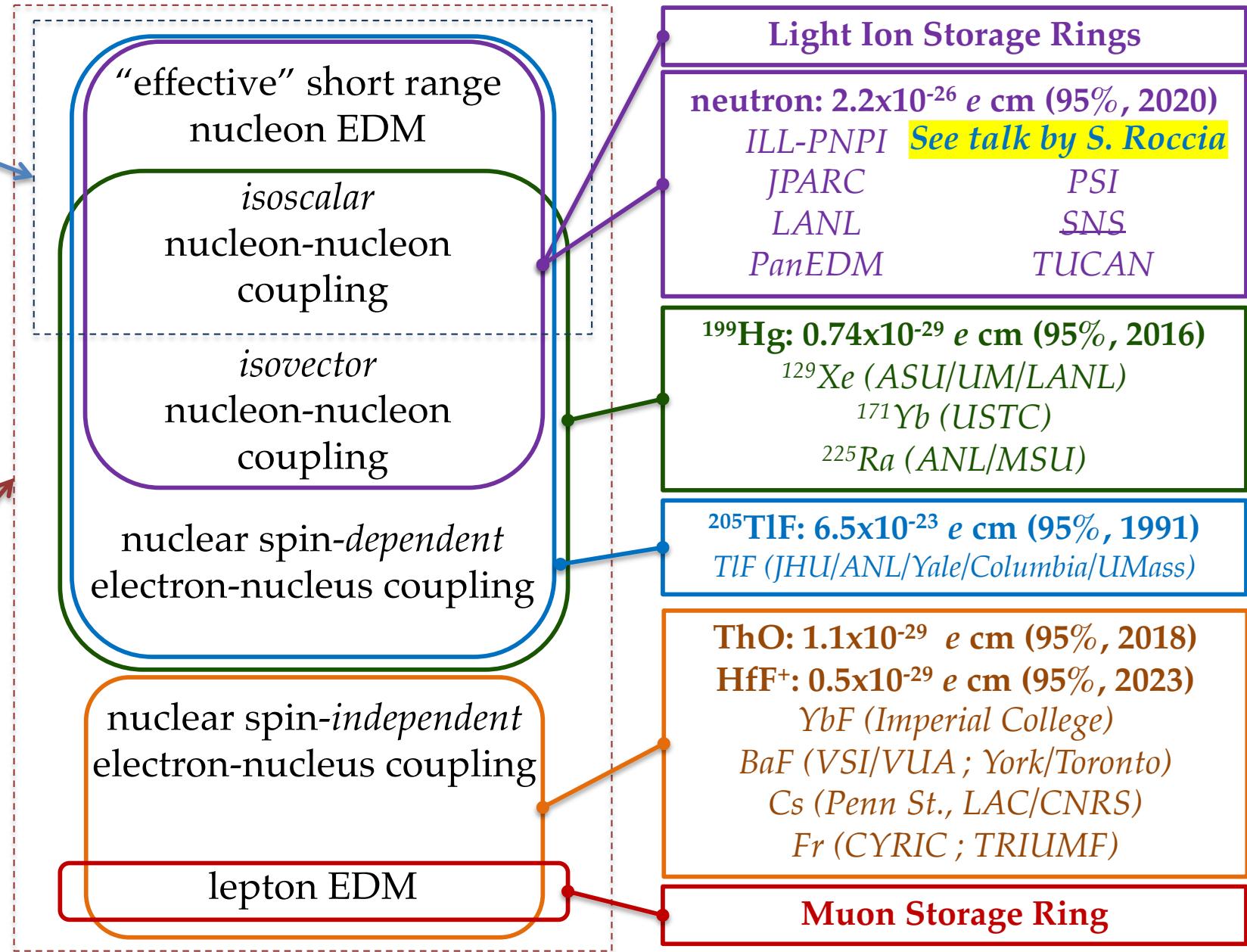
Different Sources of \mathcal{T} in EDMs of Different Systems

Physics Beyond the Standard Model

RMP 91
015001
(2019)

2025-03-15

θ_{QCD}



Different Sources of \mathcal{T} in EDMs of Different Systems

Physics Beyond the Standard Model

RMP 91
015001
(2019)

2025-03-15

θ_{QCD}

"effective" short range nucleon EDM

isoscalar nucleon-nucleon coupling

isovector nucleon-nucleon coupling

nuclear spin-dependent electron-nucleus coupling

nuclear spin-independent electron-nucleus coupling

lepton EDM

Light Ion Storage Rings

neutron: $2.2 \times 10^{-26} e \text{ cm}$ (95%, 2020)

ILL-PNPI **See talk by S. Roccia**

JPARC

LANL

PanEDM

PSI

SNS

TUCAN

^{199}Hg : $0.74 \times 10^{-29} e \text{ cm}$ (95%, 2016)

^{129}Xe (ASU/UM/LANL)

^{171}Yb (USTC)

^{225}Ra (ANL/MSU)

^{205}TlF : $6.5 \times 10^{-23} e \text{ cm}$ (95%, 1991)

TlF (JHU/ANL/Yale/Columbia/UMass)

ThO : $1.1 \times 10^{-29} e \text{ cm}$ (95%, 2018)

HfF^+ : $0.5 \times 10^{-29} e \text{ cm}$ (95%, 2023)

YbF (Imperial College)

BaF (VSI/VUA ; York/Toronto)

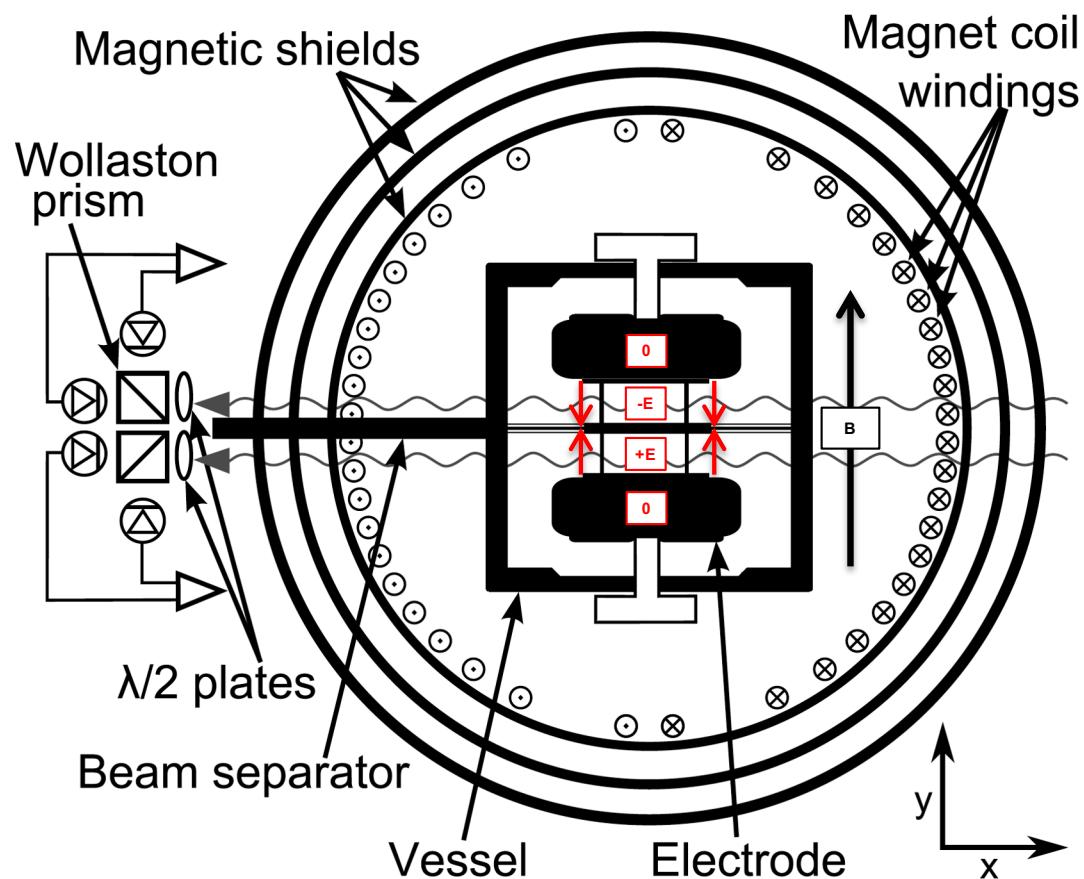
Cs (Penn St., LAC/CNRS)

Fr (CYRIC ; TRIUMF)

Muon Storage Ring

2016: Atomic EDM of ^{199}Hg (Stable) In A Vapor Cell

The Gold Standard For Over 40 Years!



- diamagnetic, $^1\text{S}_0$ 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 ~ 10 nm scale motion of vapor cells when HV is switched in the presence of 2nd order B -field gradients.

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

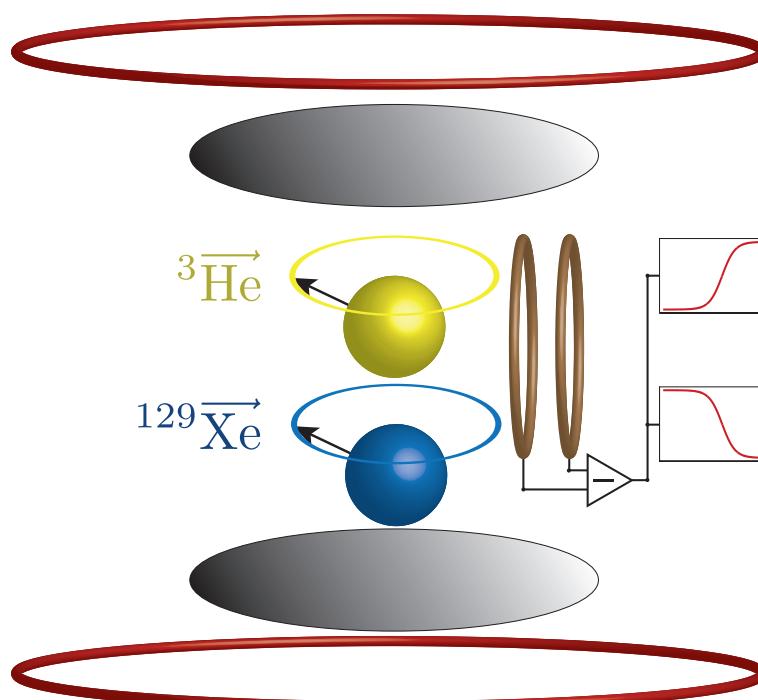
$$\Delta\nu \leq 0.1 \text{ nHz}$$

The best limit on atomic EDM:

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

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

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



Polarized Noble Gases

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

^3He : smaller EDM + B-field drifts

^{129}Xe : larger EDM + B-fields drifts

SQUID Detectors

- very sensitive detection (6 fT / root-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)

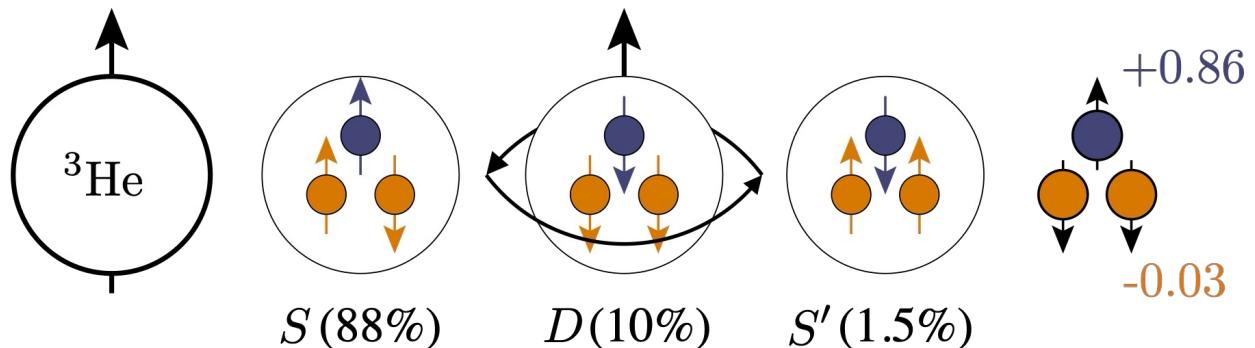
“Hyperpolarized” Noble Gases for Nuclear Physics: The Charge Distribution of the Neutron



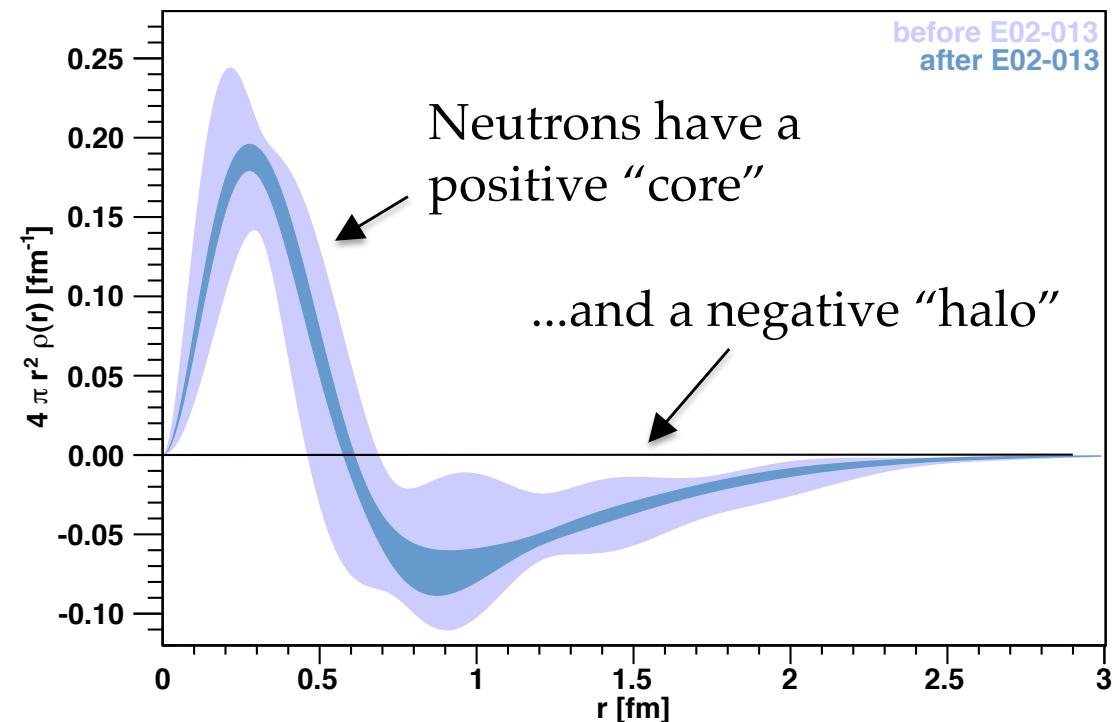
M.A. Bouchiat
photo: Zolotrev

Density = 3 amg
Volume = 30 cm³
Polarization = 0.01%
M.A. Bouchiat et al.
Phys. Rev. Lett. 5, 373 (1960)

Density = 9 amg
Volume = 400 cm³
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

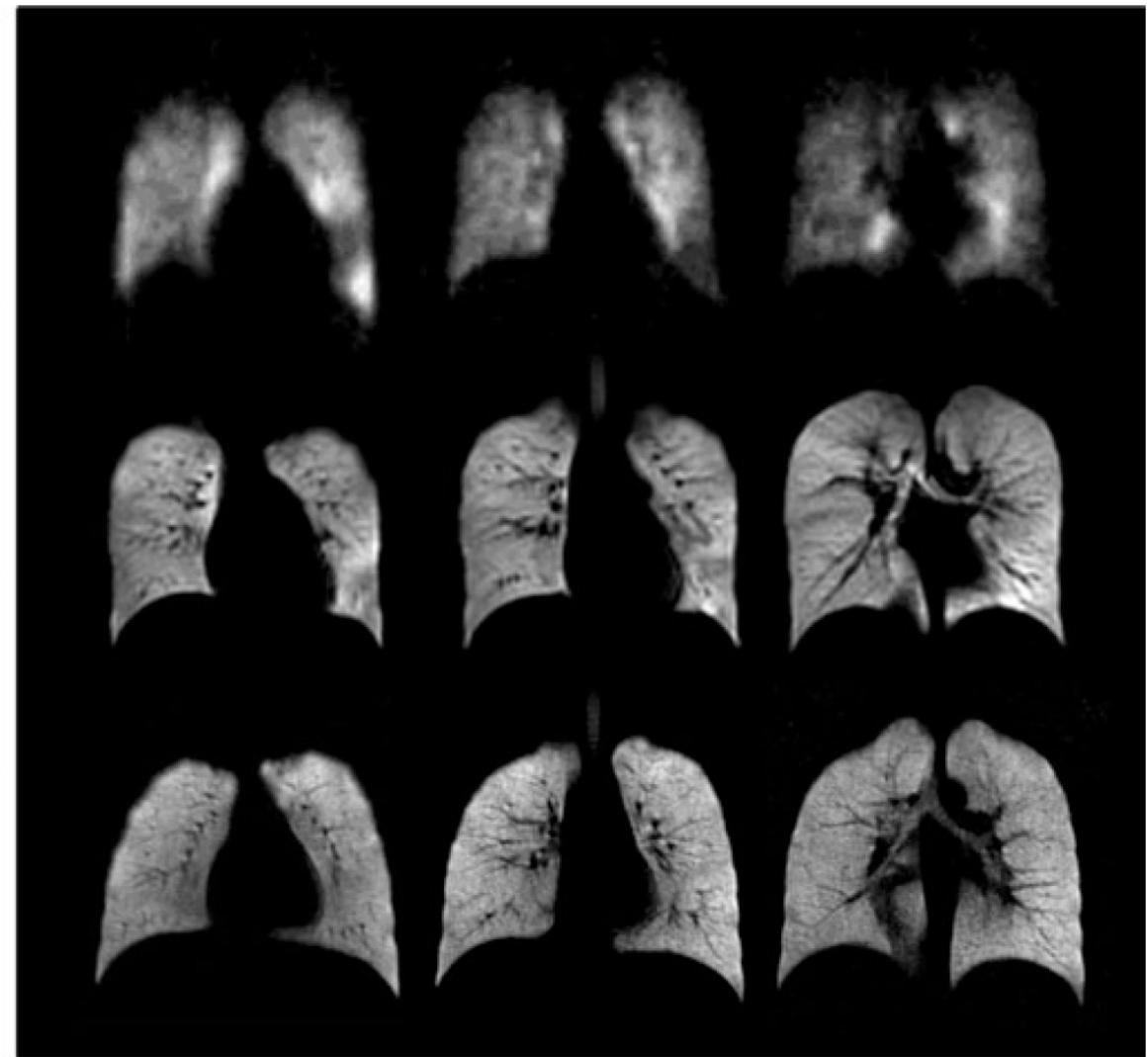


M.A. Bouchiat
photo: Zolotrev

Density = 3 amg
Volume = 30 cm³
Polarization = 0.01%
M.A. Bouchiat et al.
Phys. Rev. Lett. 5, 373 (1960)

Density = 9 amg
Volume = 400 cm³
Polarization = 69%
JTS et al.
Phys. Rev. C 91, 055205 (2015)

¹²⁹Xe
³He
¹²⁹Xe
(typical) (1996)
(2009)



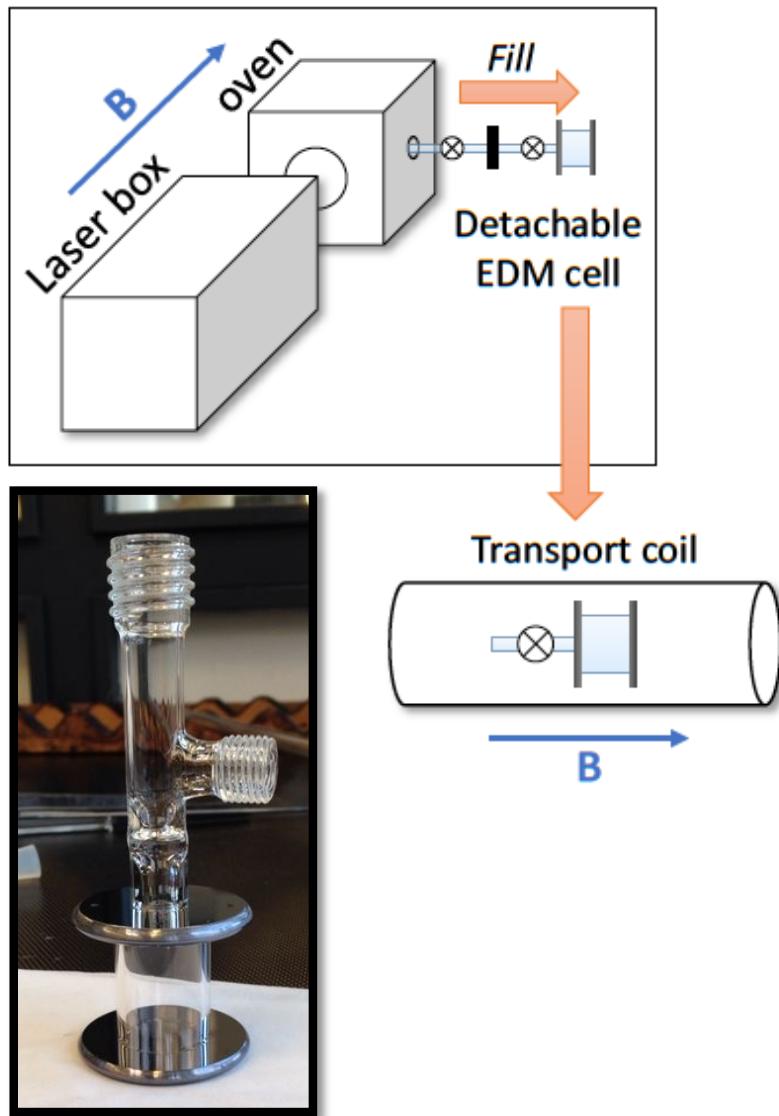
Mugler & Altes, *JMR* 37 313 (2013)

Magnetically Shielded Room in Garching, Germany

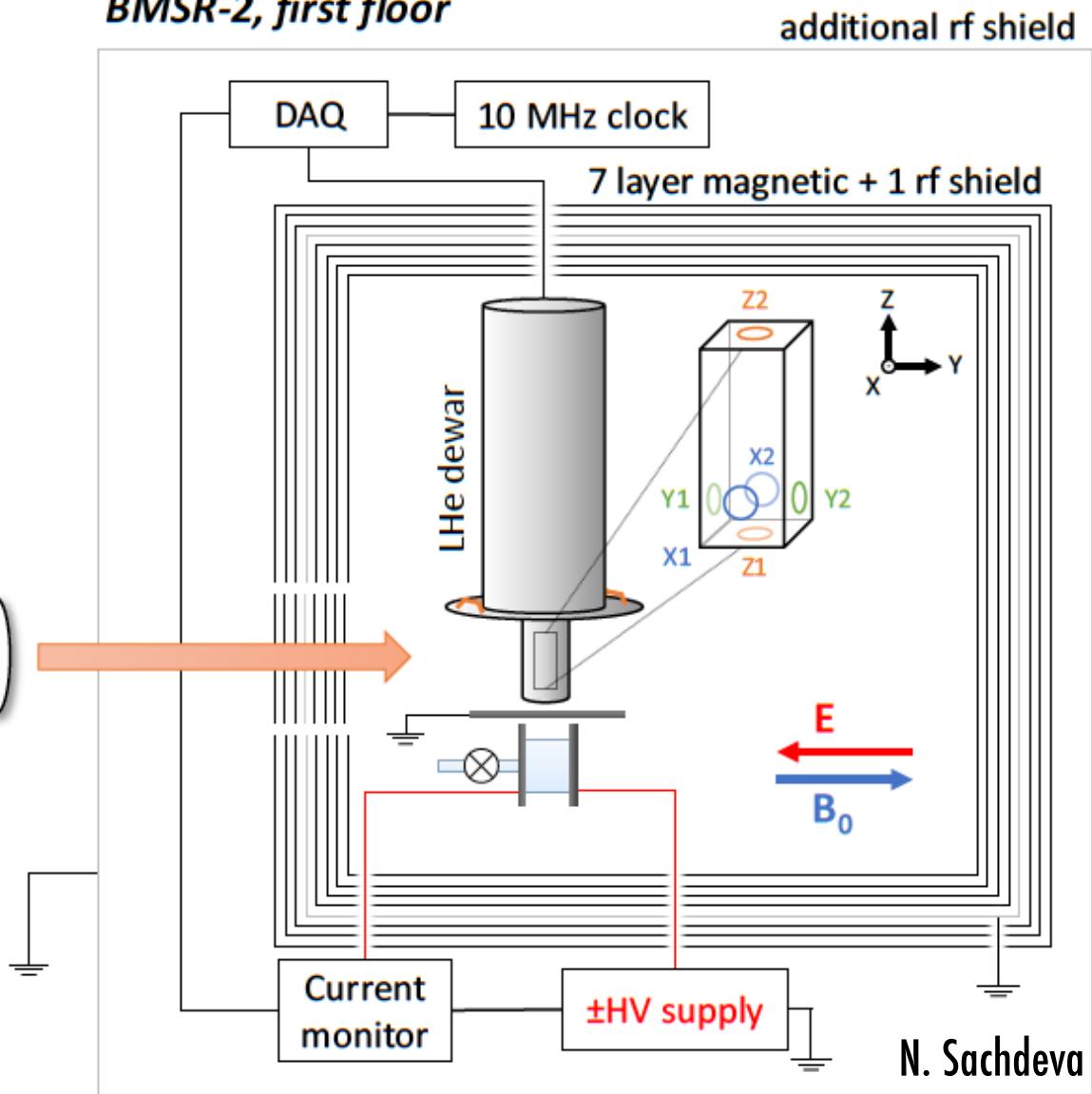


2019: The Atomic EDM of ^{129}Xe (Stable) in Berlin

Polarizer room, ground floor



BMSR-2, first floor



Main Systematic: Residual Longitudinal Polarization

PRL 123:143003 (2019)
PRA 100, 012502 (2019)

raw signal

integrated frequency sensitivity
~ 0.1 μ Hz (30 days)

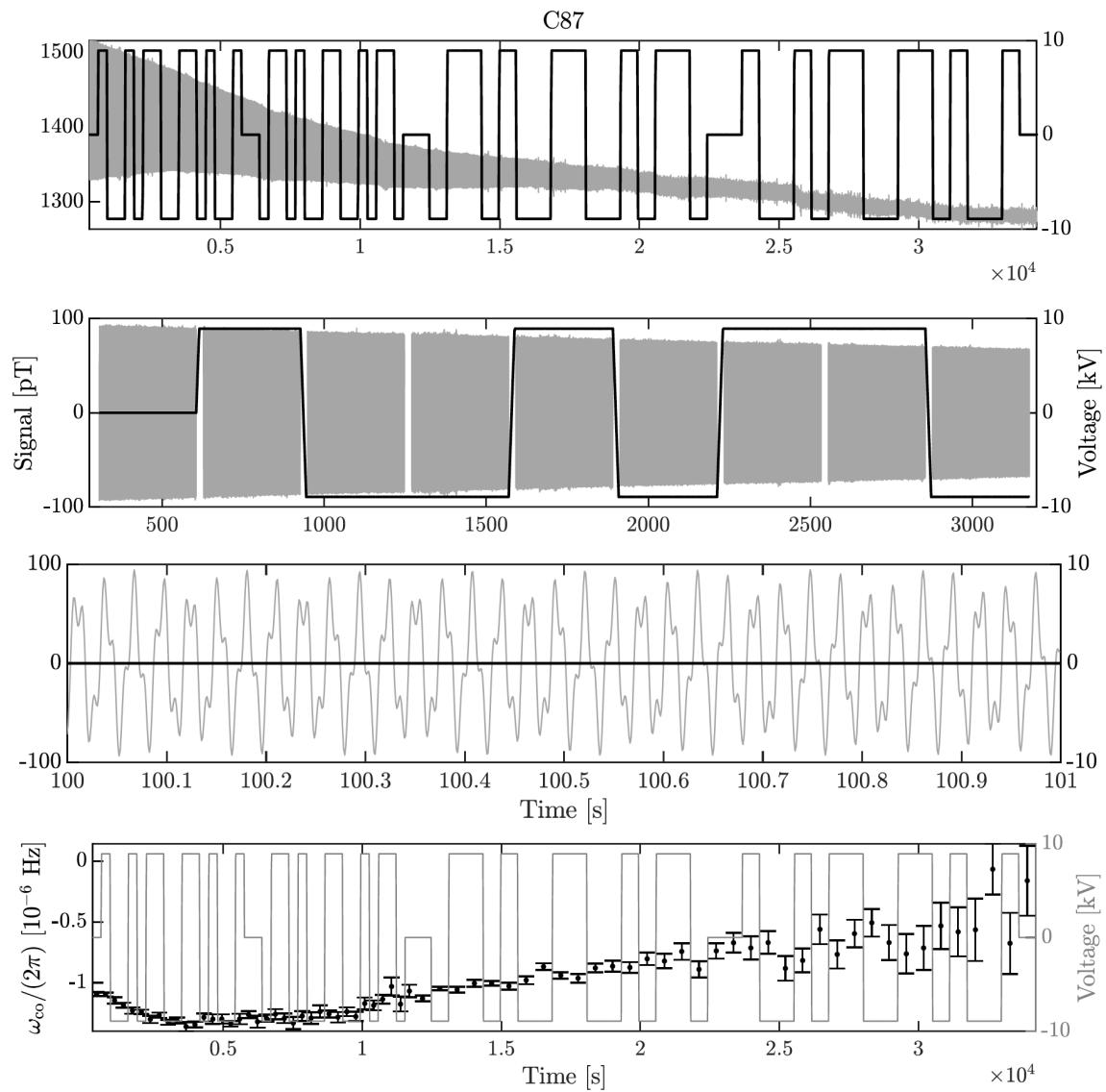
filtered signal

zoom in on filtered signal

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

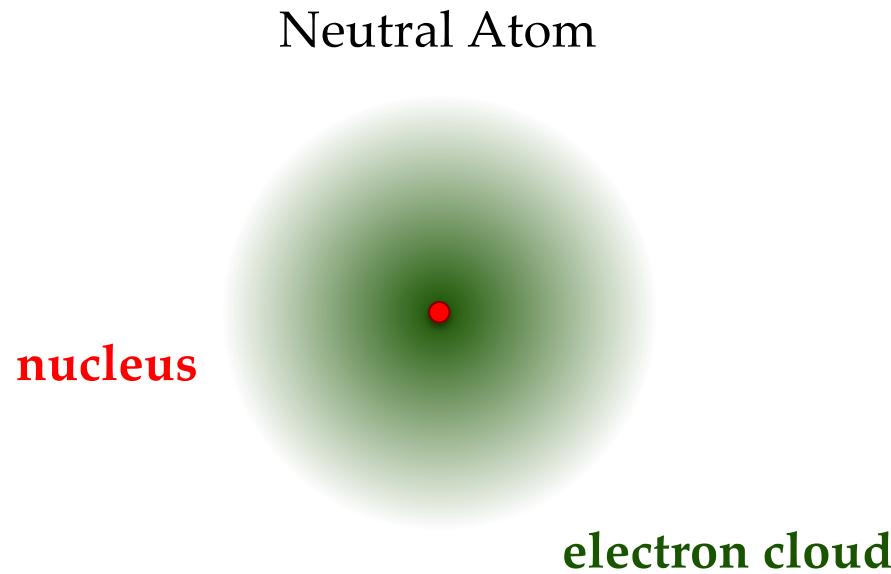
“comagnetometer” signal

Next Generation ^{129}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)]

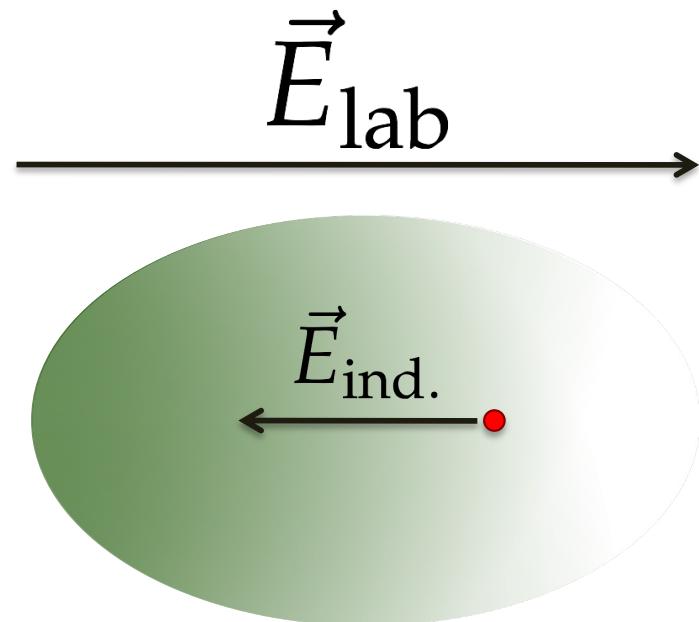
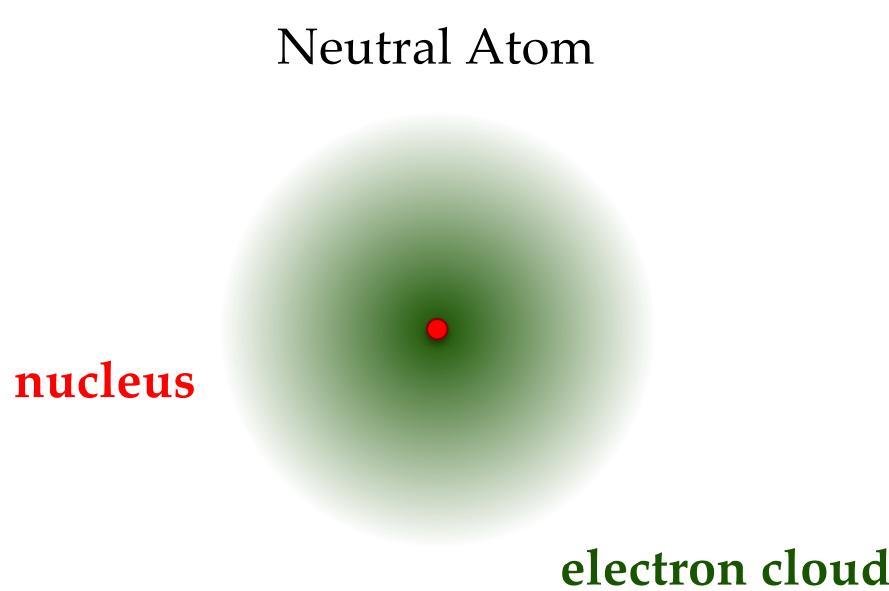
Diamagnetic Atoms: All Electrons Are Paired



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

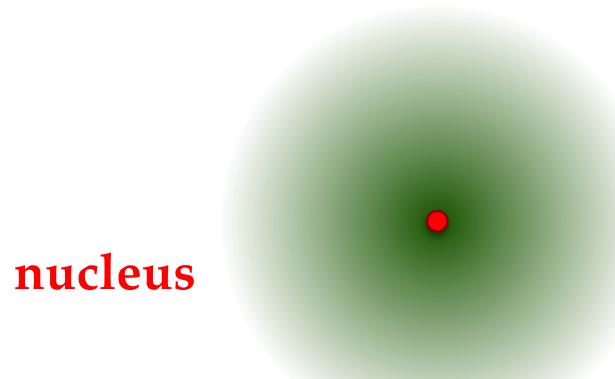
- **Shielding in Diamagnetic Atoms**
Schiff PR 132:2194 (1963)
- **Relativistic atoms: The Sandars-Bouchiat Z^3 "Law"**
Physics Letters 22:290 (1966) & Physics Letters 48B:111 (1974)
- **^{225}Ra vs ^{199}Hg vs. ^3He : 2.8 to 1 to 10^{-5}**
JPB:AMOP 53:195004 (2020) & Phys. Rev. A 106, 022817 (2022)

Madame
Professor
Marie-Anne
Bouchiat

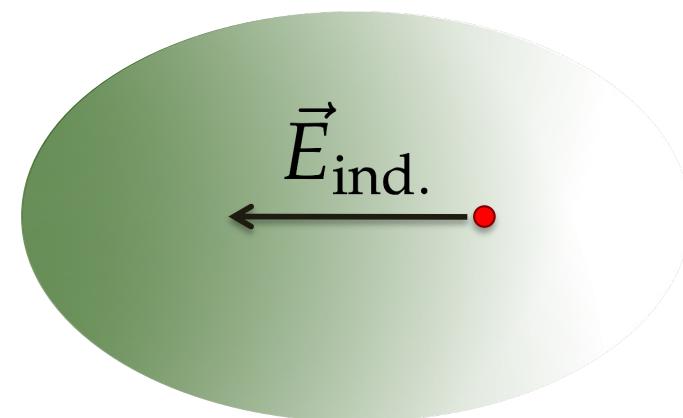


M.A. Bouchiat
photo: Zolotrev

Neutral Atom



electron cloud



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

Residual ρ & τ Observable: Nuclear Schiff Moment

- Shielding in Diamagnetic Atoms

Schiff PR 132:2194 (1963)

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

Schiff Moment

- Relativistic atoms: The Sandars-Bouchiat Z^3 "Law"

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

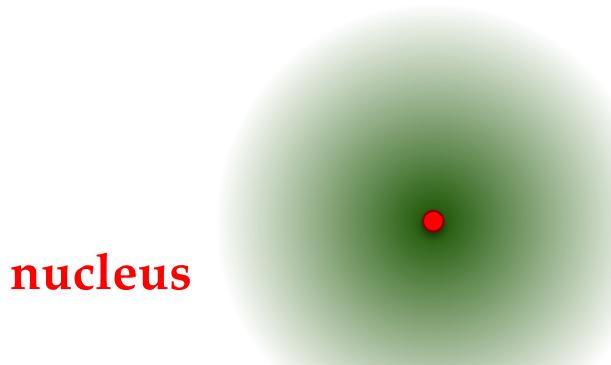
$$\vec{S} = \frac{\langle e \vec{r}^2 \vec{r} \rangle}{10} - \frac{\langle \vec{r}^2 \rangle \langle e \vec{r} \rangle}{6}$$

- ^{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)

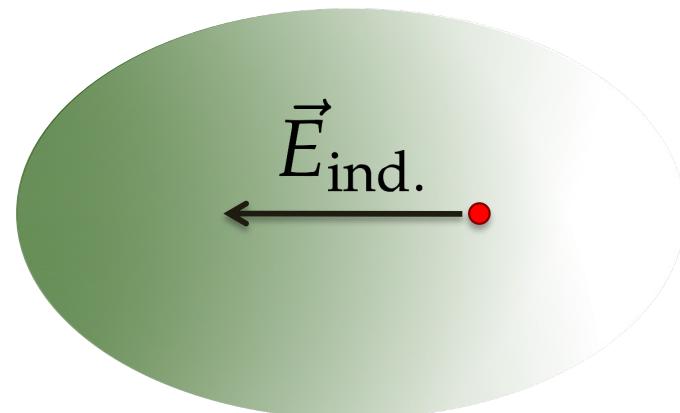
Zh. Eksp. Teor. Fiz. 87, 1521-1540 (1984)

Neutral Atom



electron cloud

$$\vec{E}_{\text{lab}}$$



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

P & T Physics: First Order Perturbation Theory

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

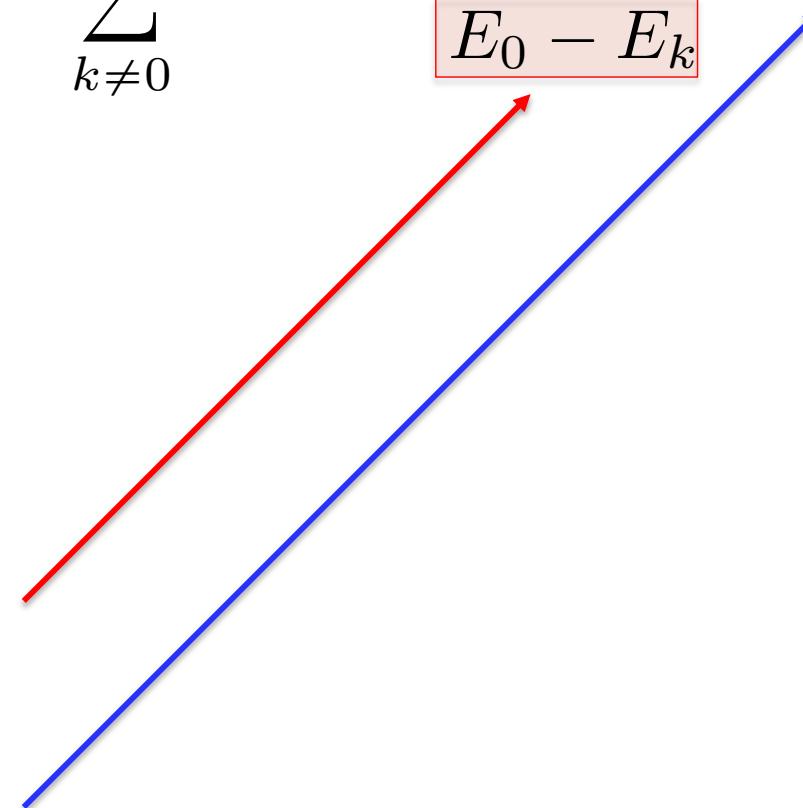
$$S \equiv \langle \Psi_0 | S_z | \Psi_0 \rangle = \sum_{k \neq 0} \frac{\langle \Psi_0 | S_z | \Psi_k \rangle \langle \Psi_k | V_{\text{PT}} | \Psi_0 \rangle}{E_0 - E_k} + \text{c.c.}$$

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

Isotopes With Nearly Degenerate Nuclear States

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

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



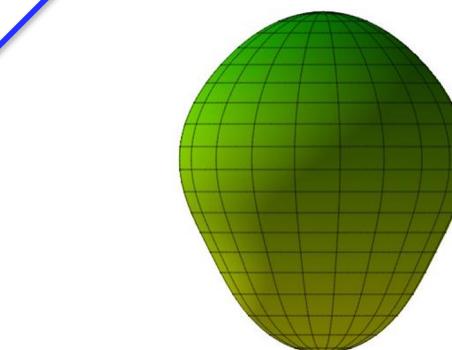
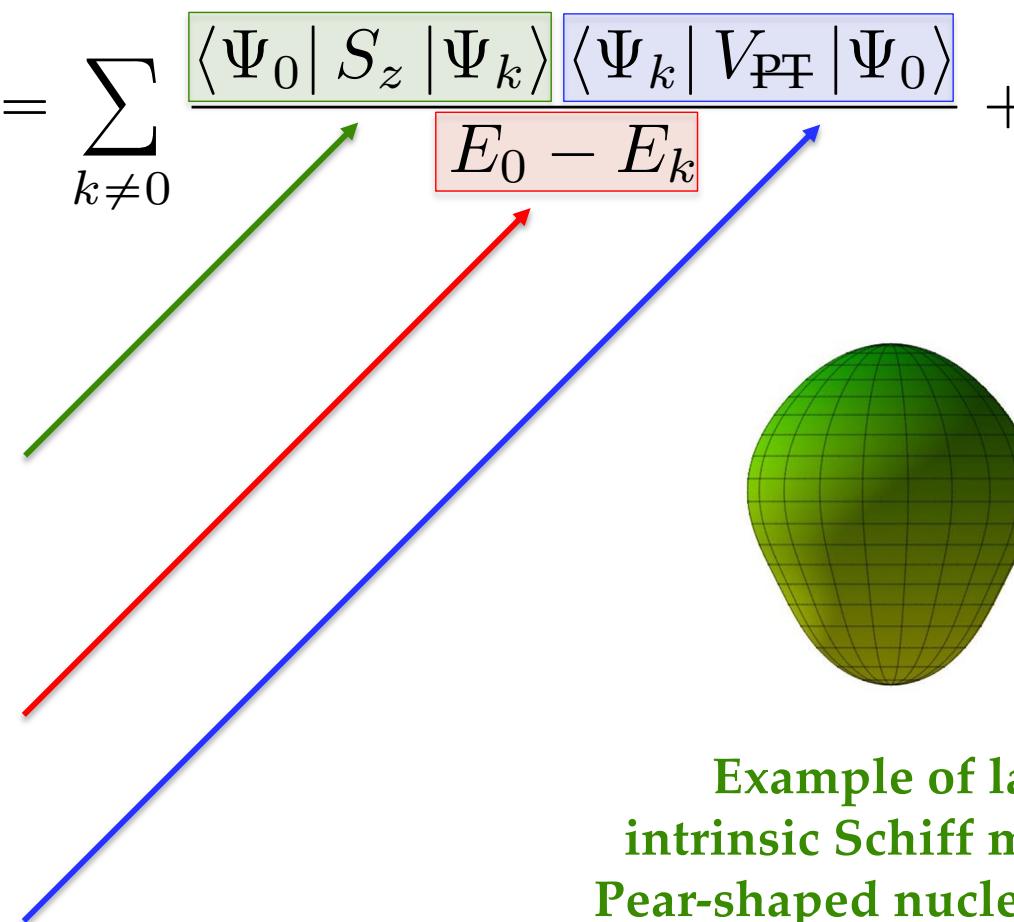
- Difference in lab-frame nuclear energy levels
- The P and T physics that we seek (unknown & common to all isotopes)

Nuclear Schiff Moment in the Lab Frame

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

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

- Body-frame Schiff moment – large when there are intrinsic nuclear deformations
- Difference in lab-frame nuclear energy levels
- The P and 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

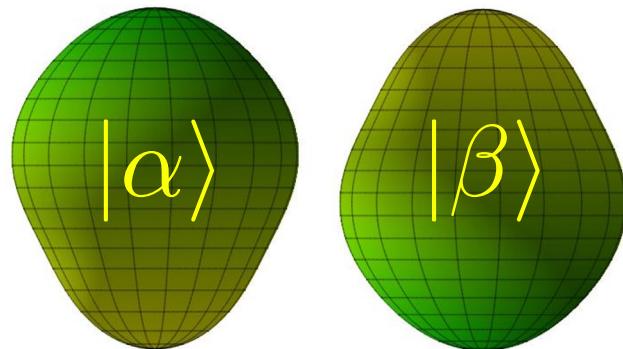
$$S_z = \frac{\langle er^2 z \rangle}{10} - \frac{\langle r^2 \rangle \langle ez \rangle}{6}$$

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

Parity Doublet

- Nearly degenerate parity doublet

Haxton & Henley PRL 51:1937 (1983)



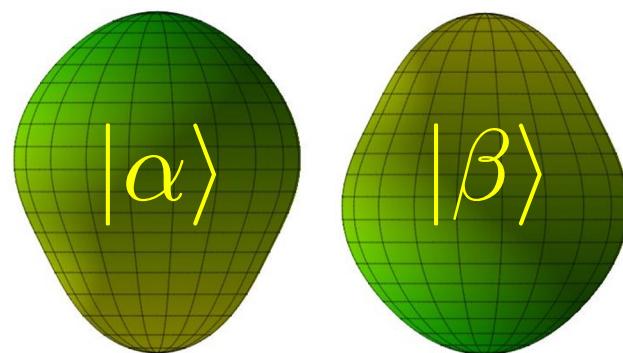
$$\Delta E$$
$$| \Psi_1 \rangle = \frac{|\alpha\rangle \mp |\beta\rangle}{\sqrt{2}}$$
$$| \Psi_0 \rangle = \frac{|\alpha\rangle \pm |\beta\rangle}{\sqrt{2}}$$

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)

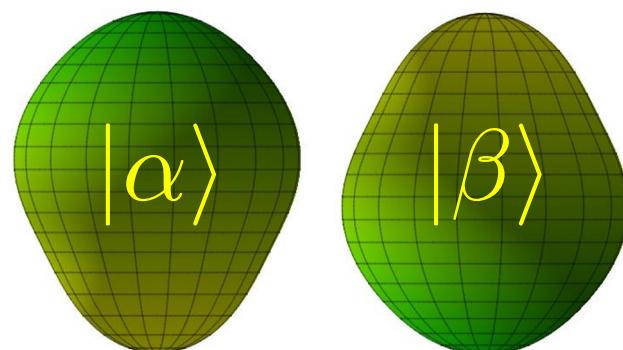
$$\Delta E$$
$$| \Psi_1 \rangle = \frac{|\alpha\rangle \mp |\beta\rangle}{\sqrt{2}}$$
$$| \Psi_0 \rangle = \frac{|\alpha\rangle \pm |\beta\rangle}{\sqrt{2}}$$

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



55 keV

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

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

- **Nearly degenerate parity doublet**
Haxton & Henley PRL 51:1937 (1983)
- **Large intrinsic Schiff moment due to octupole deformation**
Auerbach, Flambaum, & Spevak PRL 76:4316 (1996)

Total Enhancement Factor: EDM (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)

Nuclear Structure Calculations Are Challenging!

type	^{199}Hg	^{225}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}Ra or other octupole deformed species

Ongoing: The Atomic EDM of ^{225}Ra at Argonne

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

PRL 114:233002 (2015)

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

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

PRC 94:025501 (2016)

Upgrades underway to improve sensitivity by x1000

Spectrochimica Acta Part B 172 105967 (2020)

^{226}Ra

nuclear spin = 0

$t_{1/2} = 1600$ years

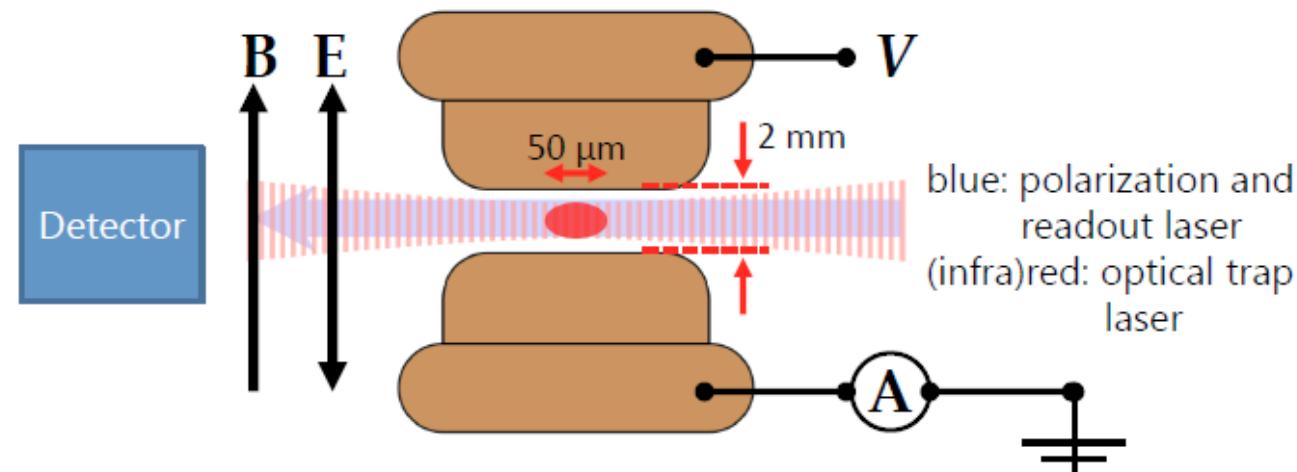
Low vapor pressure

^{225}Ra

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

$t_{1/2} = 15$ days

Low vapor pressure



EDM search using atoms held in Optical Lattice

Romalis & Fortson PRA 59:4547 (1999)

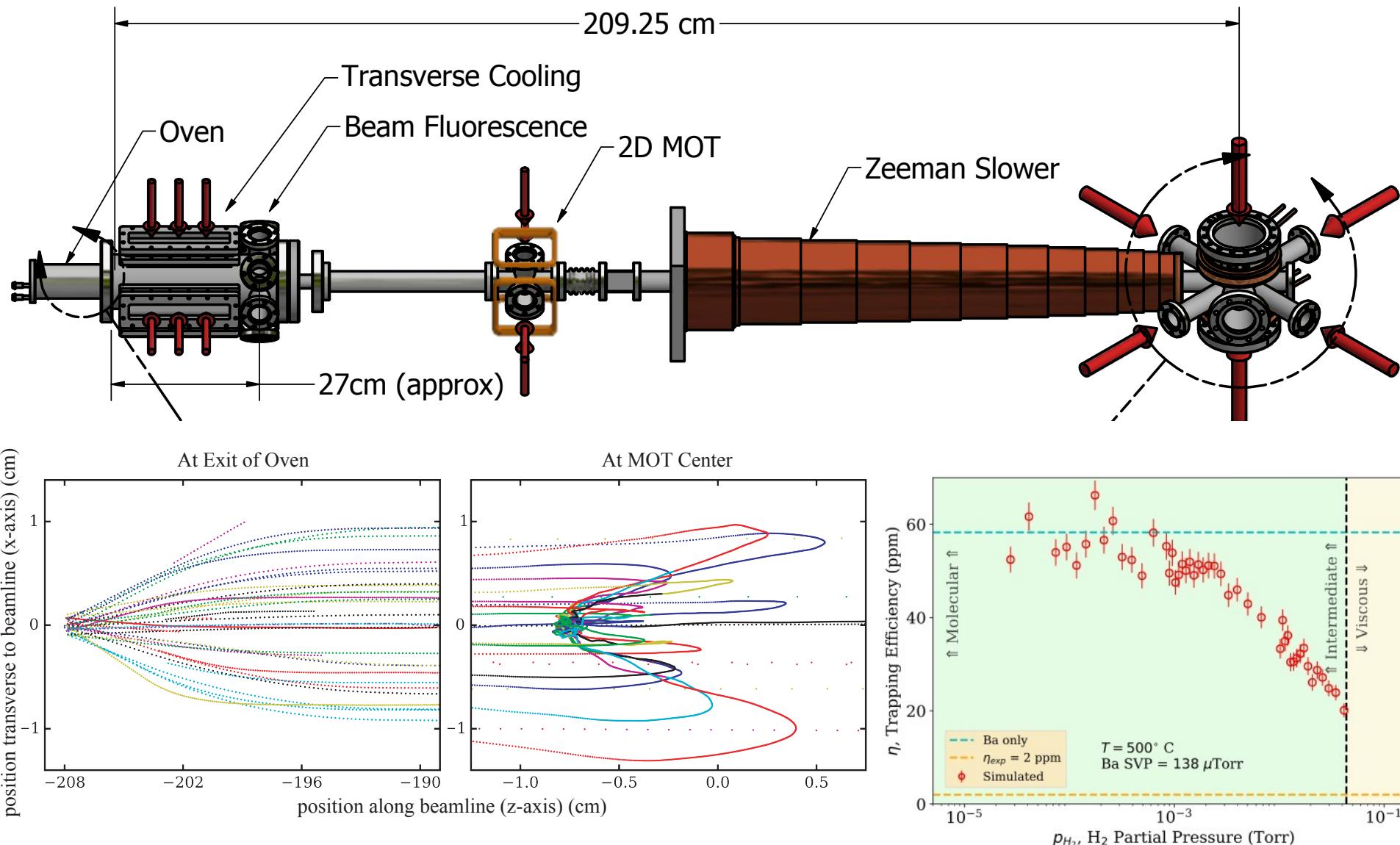
Chin et al. PRA 63:033401 (2001)

Bishof et al. PRC 94:025501 (2016)

- Atoms concentrated in a very small region
- Long coherence time (100 s) PRL 129, 083001 (2022)
- negligible “ $v \times E$ ” 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 (~20 Hz)

probability of absorbing probe light and creating a shadow:

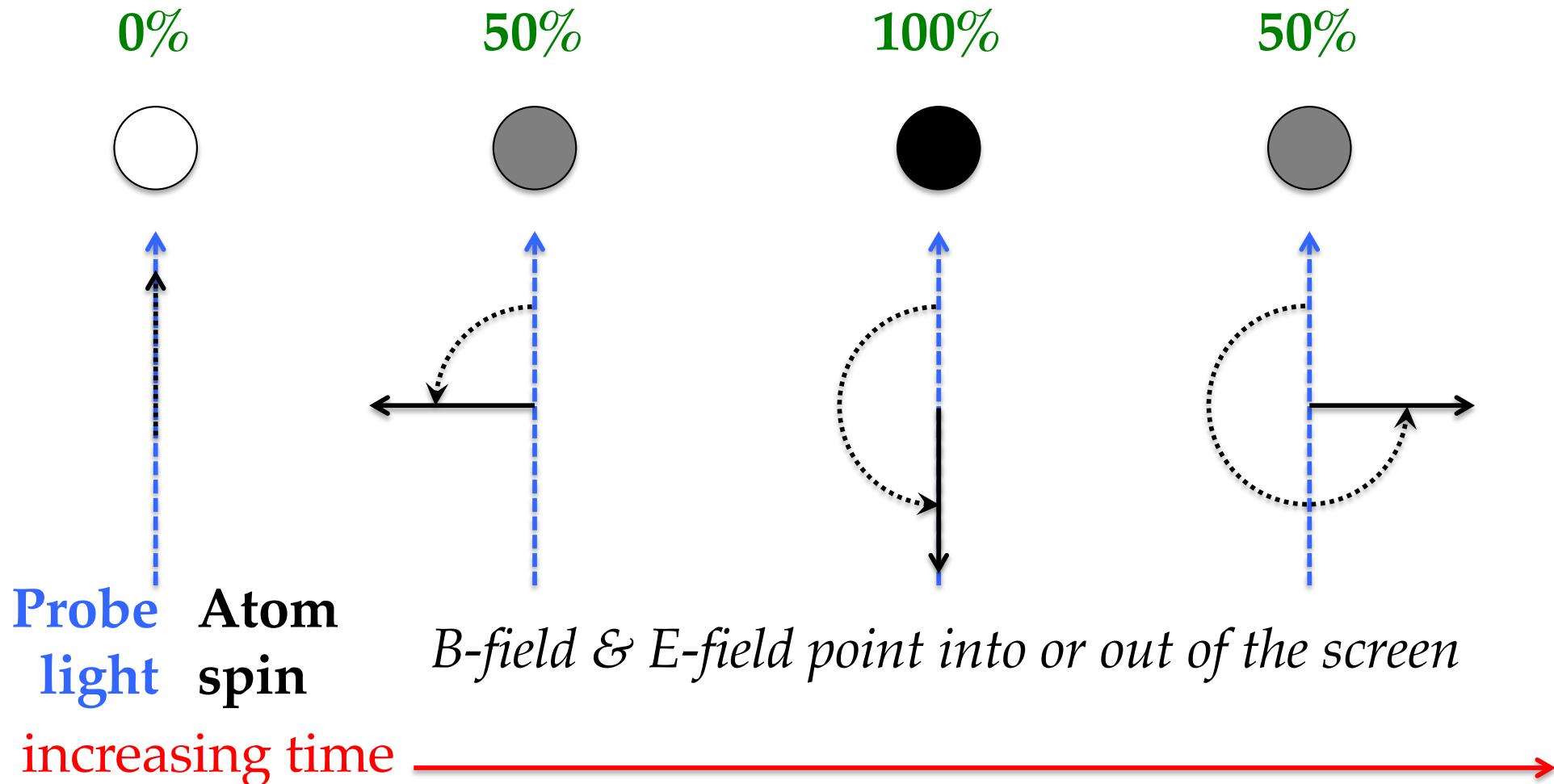
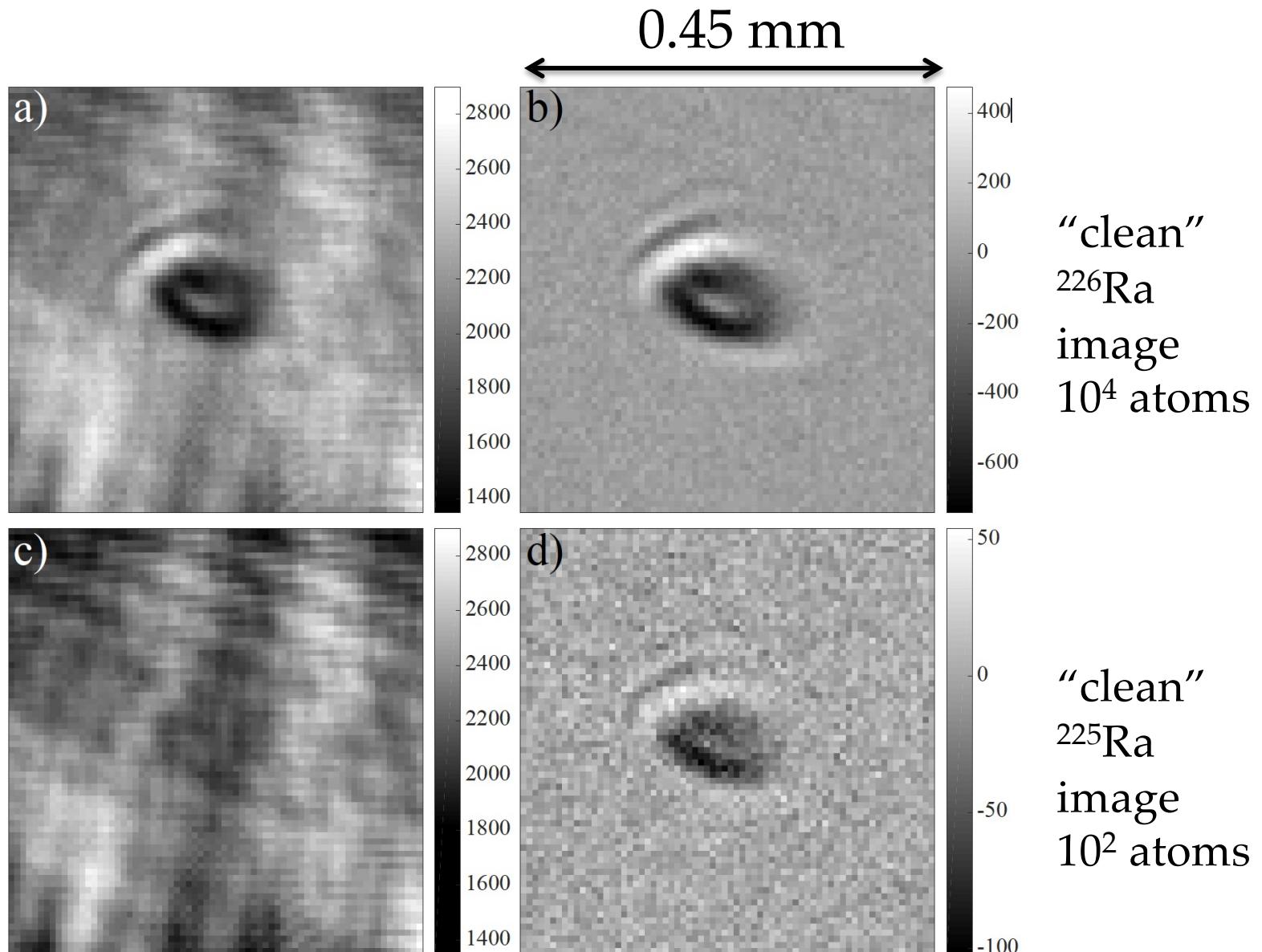
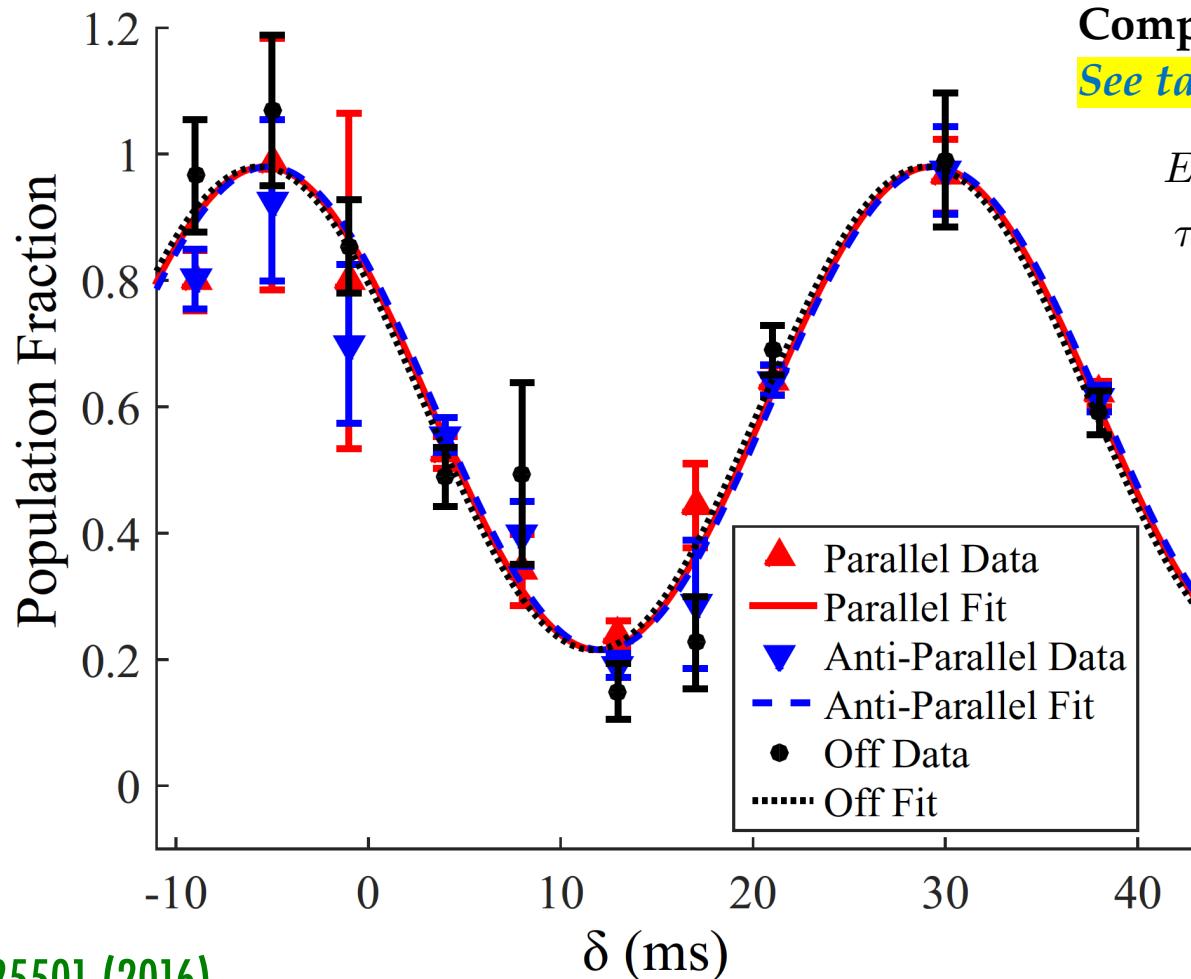


Image Background & Distortion Corrections

average of 8
raw images
of ^{226}Ra



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



Completely Statistics Dominated
See talk by G. Arrowsmith-Kron

$E = 67 \text{ kV/cm} \rightarrow 500 \text{ kV/cm}$
 $\tau = 2 \text{ s} \rightarrow 20 \text{ s} \rightarrow 200 \text{ s}$

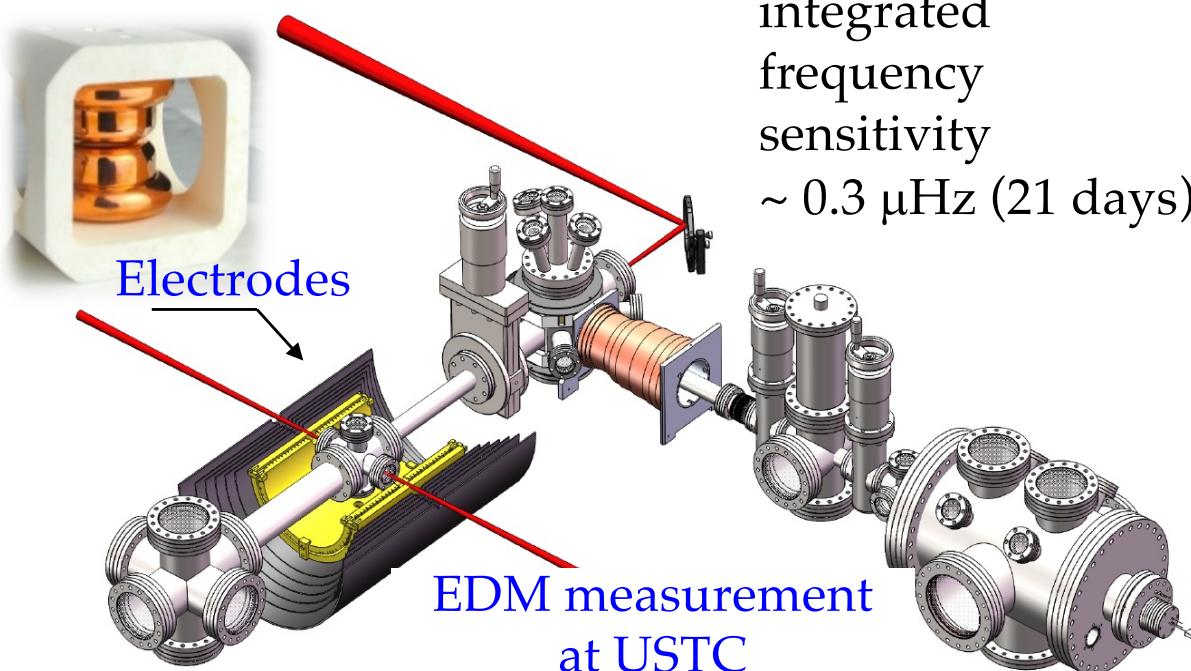
$$d = \frac{h(\Delta\phi)}{4E\tau}$$

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}Yb (Stable) in a Laser Trap Using Laser Probing



integrated frequency sensitivity
 $\sim 0.3 \mu\text{Hz}$ (21 days)

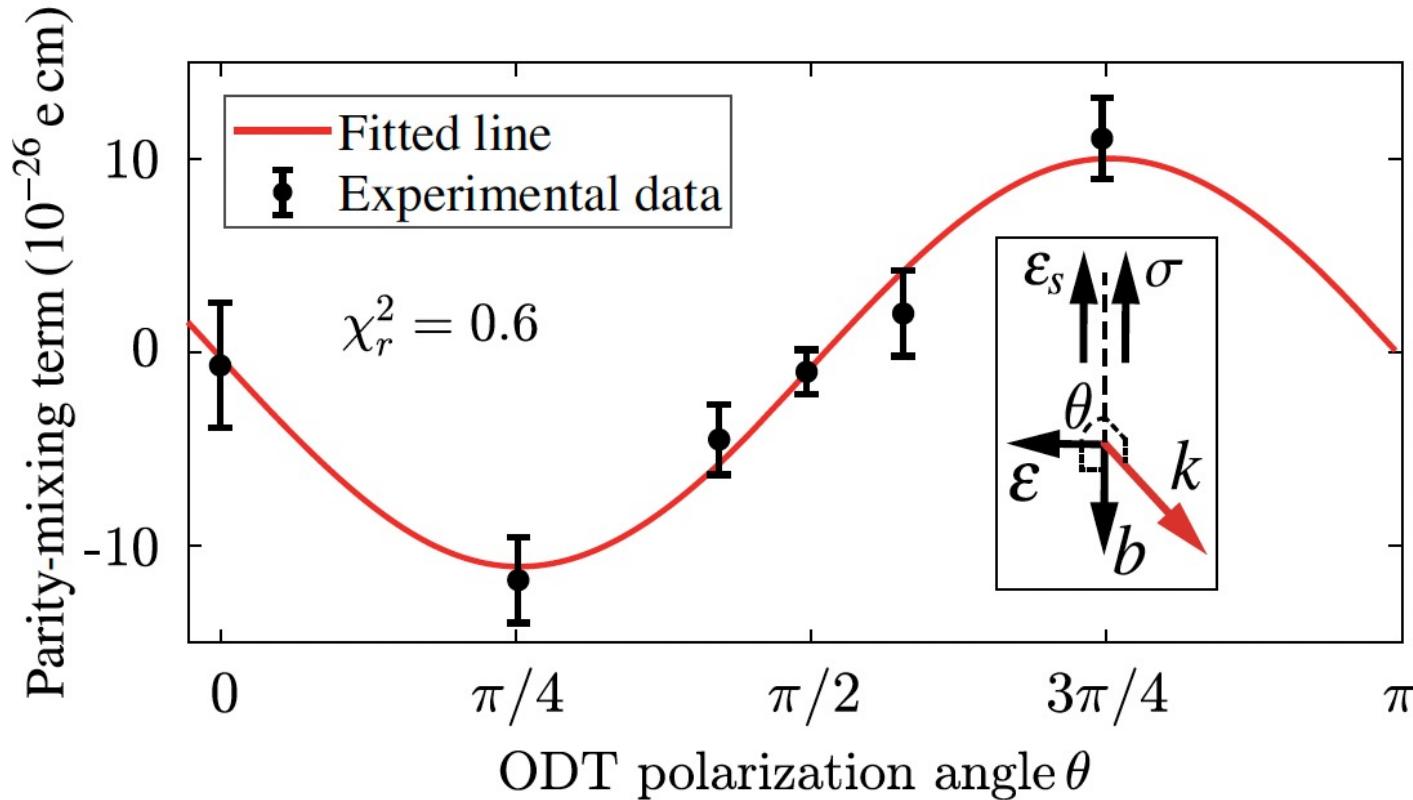
- Pathfinder experiment for ^{225}Ra (10^5 vs 10^2 atoms)
- Coherent spin precession time > 300 s
- **EDM(^{171}Yb) $< 1.5 \times 10^{-26} e\text{-cm}$ (95% C.L.), equivalent to $\sim 1000 \times$ EDM(^{199}Hg)**

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}Yb EDM is Good Enough for Next Generation ^{225}Ra EDM



b = ODT B-field
 ε = ODT E-field
 σ = B-field
 ε_s = E-field

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

Frequency shift scale factor of ^{225}Ra
See talk by G. Arrowsmith-Kron

PRL 129, 083001 (2022)

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

^{225}Ra : PRC 94:025501 (2016): $< 1.4 \times 10^{-23} e \text{ cm}$ (95%)
(rare pear-shaped nuclei + laser trap experiment: 1 mHz)

^{129}Xe : PRL 123:143003 (2019): $< 1.4 \times 10^{-27} e \text{ cm}$ (95%)
(stable + gas cell experiment: 0.1 μHz)

^{171}Yb : PRL 129:083001 (2022): $< 1.5 \times 10^{-26} e \text{ cm}$ (95%)
(stable + laser trap experiment: 0.3 μHz
nearly identical to ^{225}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.

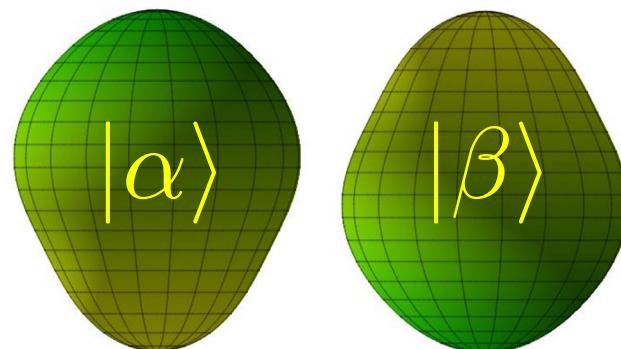
Protactinium-229 *May* Be Unusually Sensitive!

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

Choose an isotope with large deformations

Unknown

Parity Doublet



$$\Delta E$$

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

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

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	ΔE (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
Pa-229	(0.06 +/- 0.05)?	10^5	10^6

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}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)

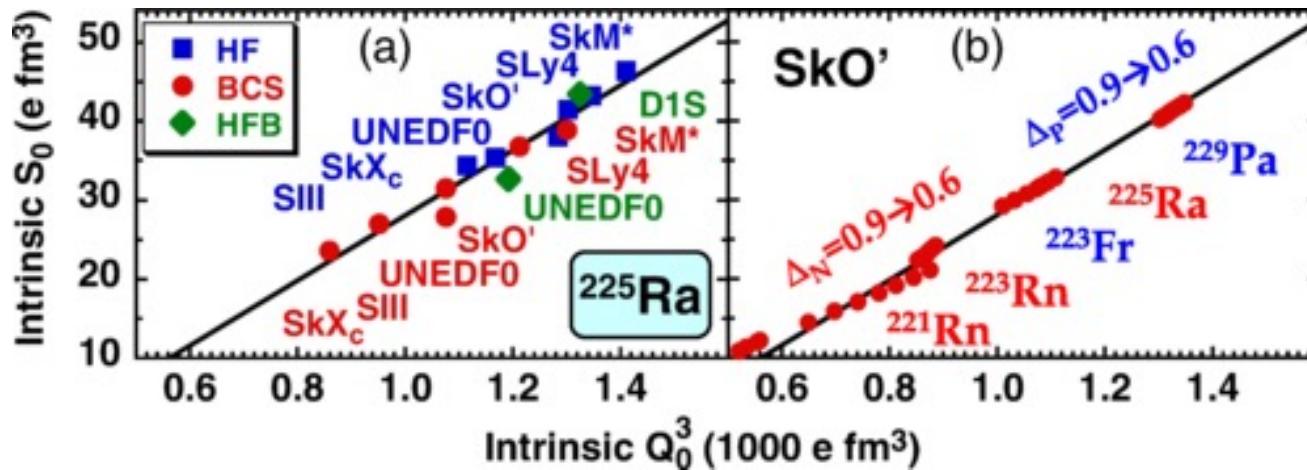
6. Magnetic moment g-factors should be the same for both members of the PD

Status: No measurements yet

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

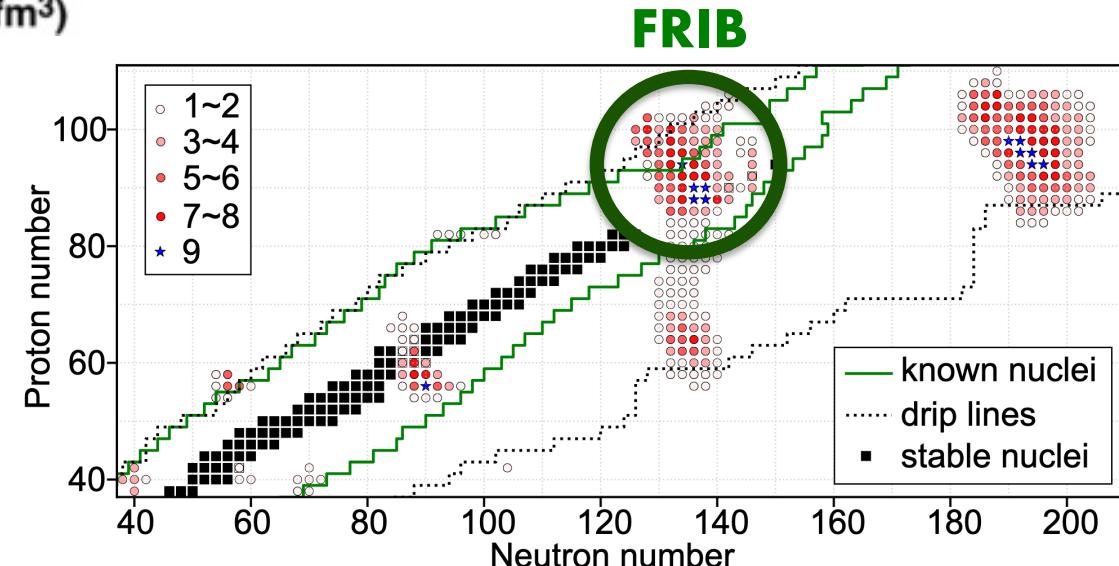
Calibrating the Intrinsic Schiff Moment (Numerator)

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



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}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

Ground State Parity Doublet Energy Splitting (Denominator) The Muddled Picture for ^{229}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

1991: No ground state parity doublet found in German experiment!

PRC 44:R1728-31

1993: No evidence for octupole deformation from German experiment!

PPNP 28:429-34

Ground State Parity Doublet Energy Splitting (Denominator) The Muddled Picture for ^{229}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

1991: No ground state parity doublet found in German experiment!

PRC 44:R1728-31

1993: No evidence for octupole deformation from German experiment!

PPNP 28:429-34

1994: Weak octupole collectivity now claimed by German experiments!

NPA 576:267-307

Ground State Parity Doublet Energy Splitting (Denominator) The Muddled Picture for ^{229}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

1991: No ground state parity doublet found in German experiment!

PRC 44:R1728-31

1993: No evidence for octupole deformation from German experiment!

PPNP 28:429-34

1994: Weak octupole collectivity now claimed by German experiments!

NPA 576:267-307

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}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 γ -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}Pa was identified in Ref. [3] from two closed cycles of γ rays. One of the closed cycles, $211.09 - (122.51 + 88.43)$, cannot be used because the 211.06-keV γ 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}Pa ground state. However, the large uncertainty in the energy difference makes the assignment of the $5/2^-$ level uncertain.

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)

“The state exists for sure!

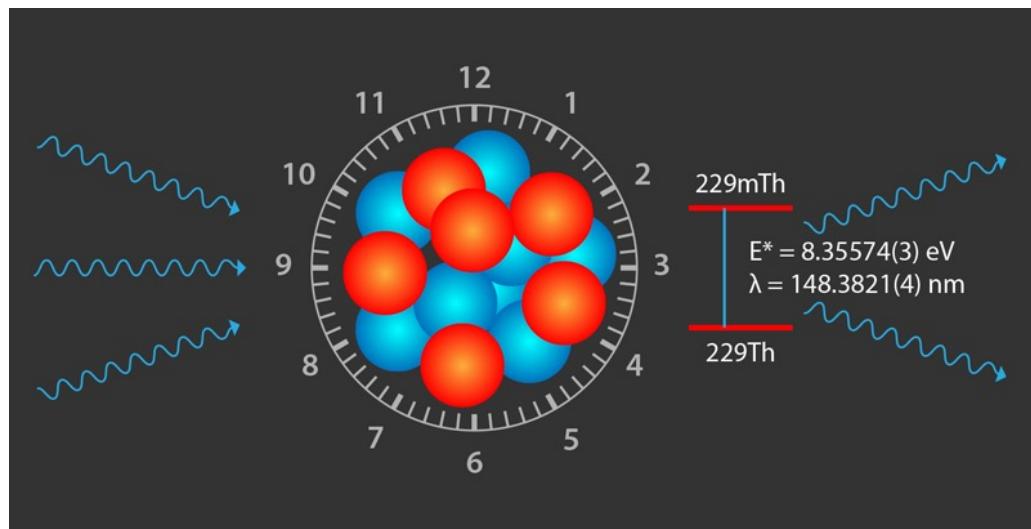
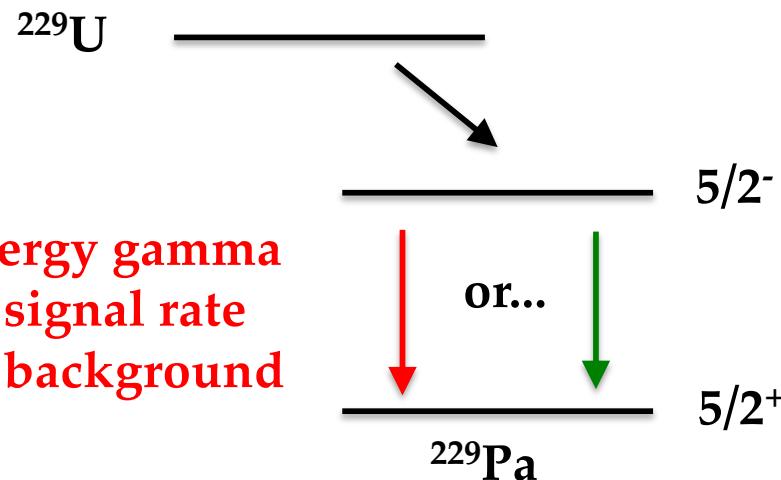
I guarantee it!

100%!

Just look for it now!”

Private Communication
with IA (2014)

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



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

<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}U as 76.737 ± 0.018 eV. The ^{235m}U isomer is created from the α decay of ^{239}Pu and embedded directly into the detector. When the ^{235m}U subsequently decays, the energy is fully contained within the detector and is

PRC 97 054310 (2018)

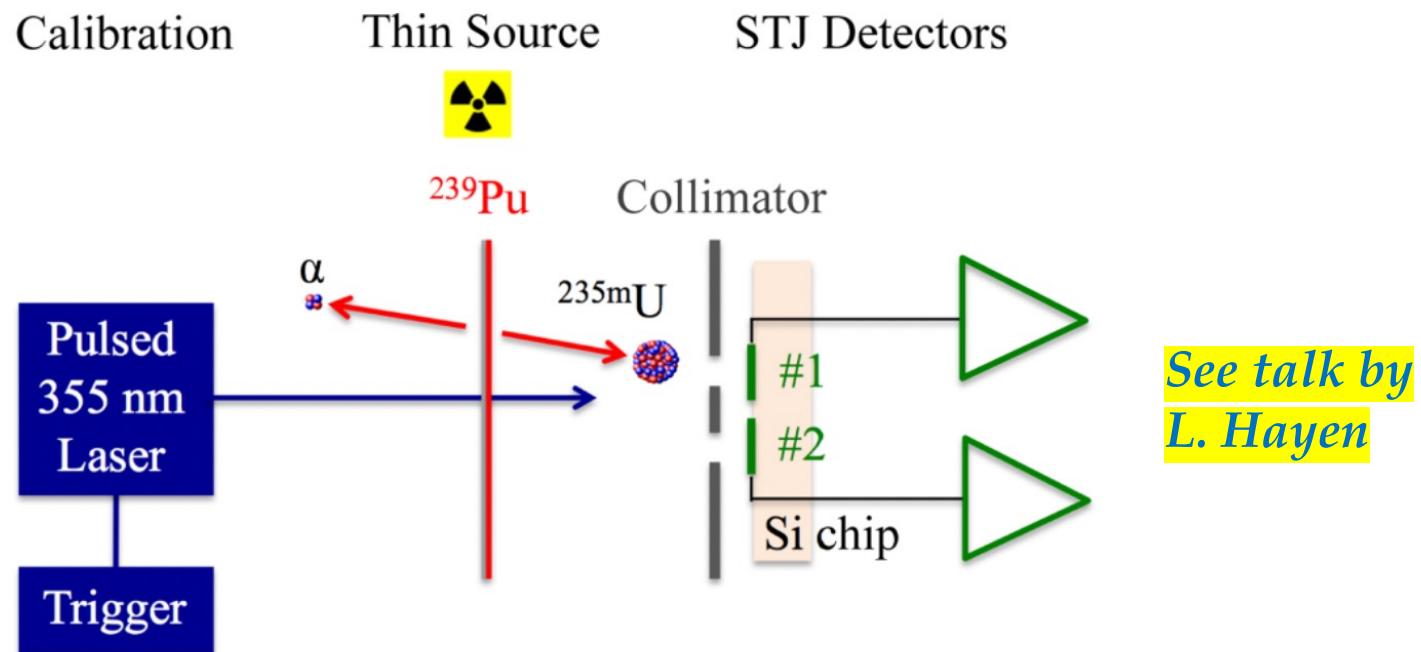
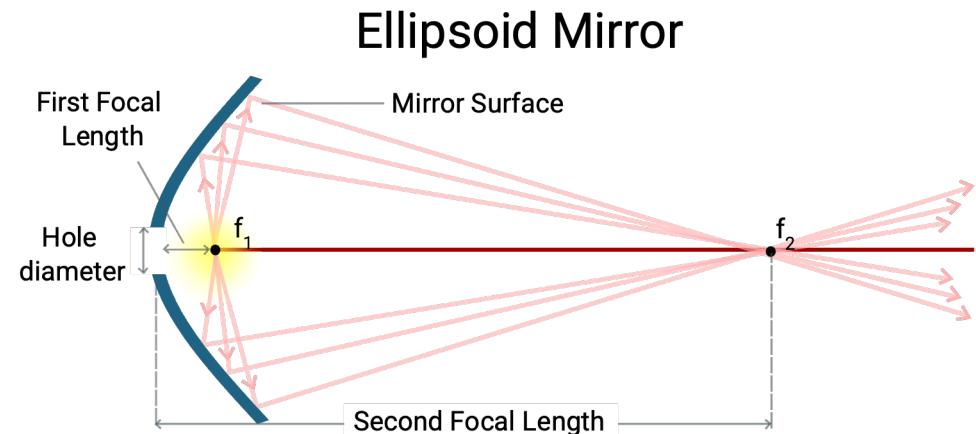
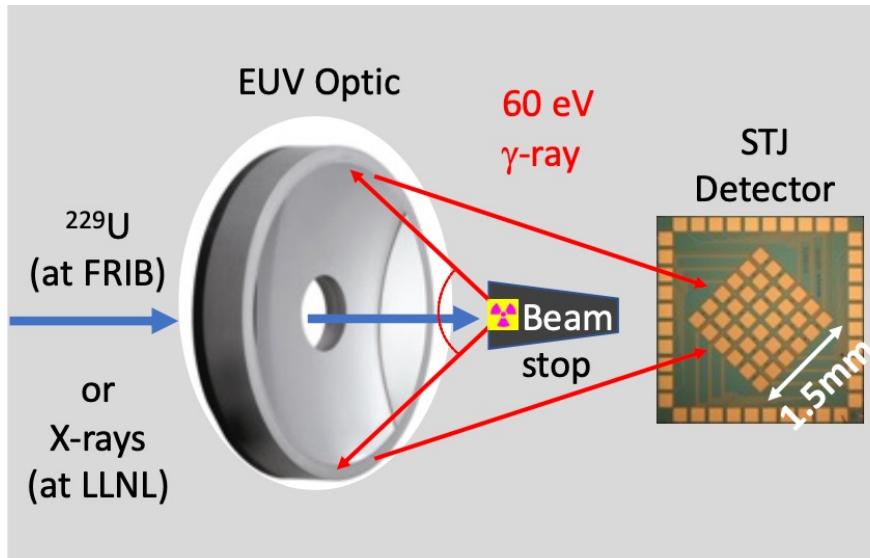
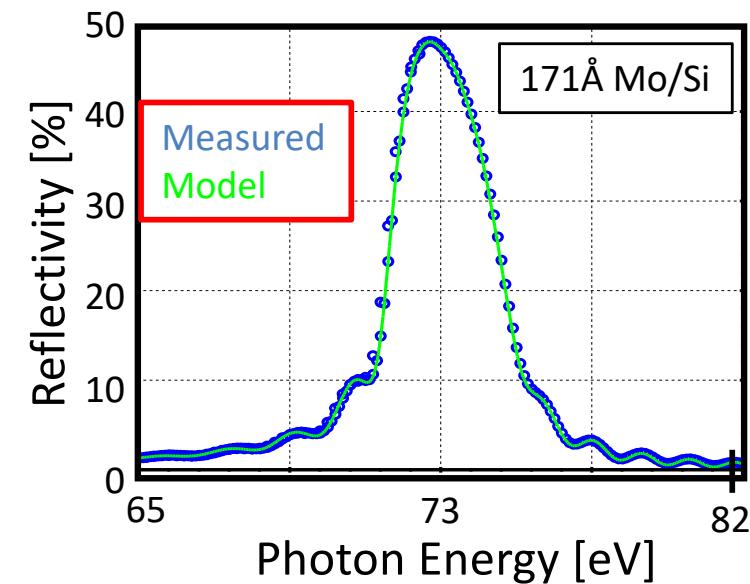
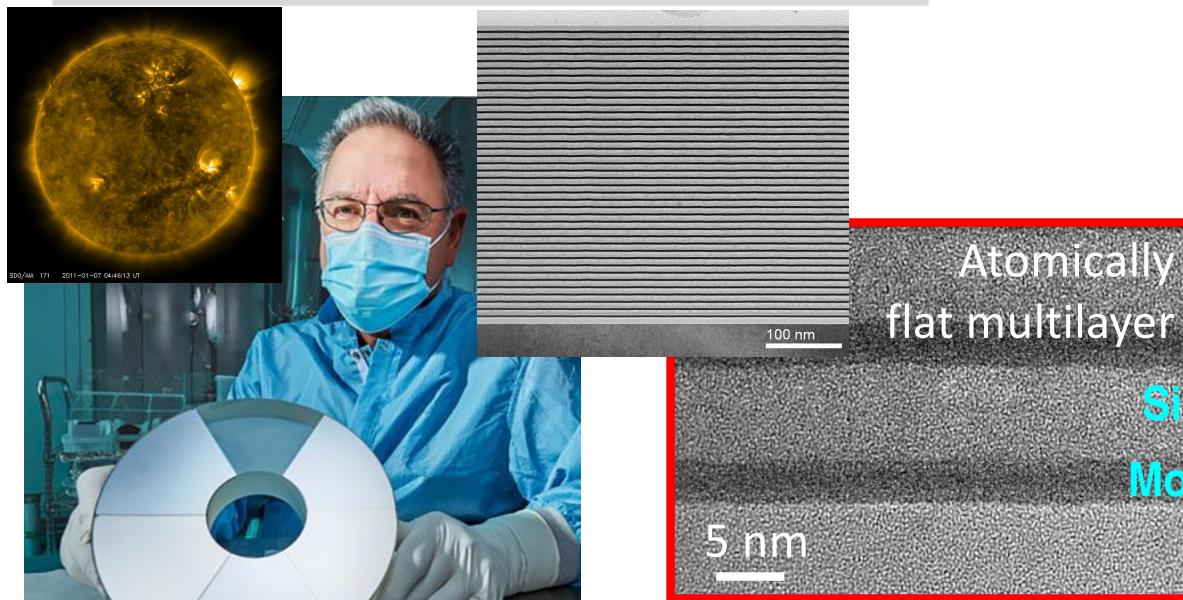


FIG. 1. Schematic of experimental setup: ^{235m}U recoil ions produced by the decay of ^{239}Pu are embedded in the STJ detectors, which measure their subsequent decay into the ^{235}U ground state.

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 ~ 54 eV
5. Source: $^{239}\text{Pu} / ^{235}\text{mU}$
energy ~ 76 eV
6. ship to FRIB

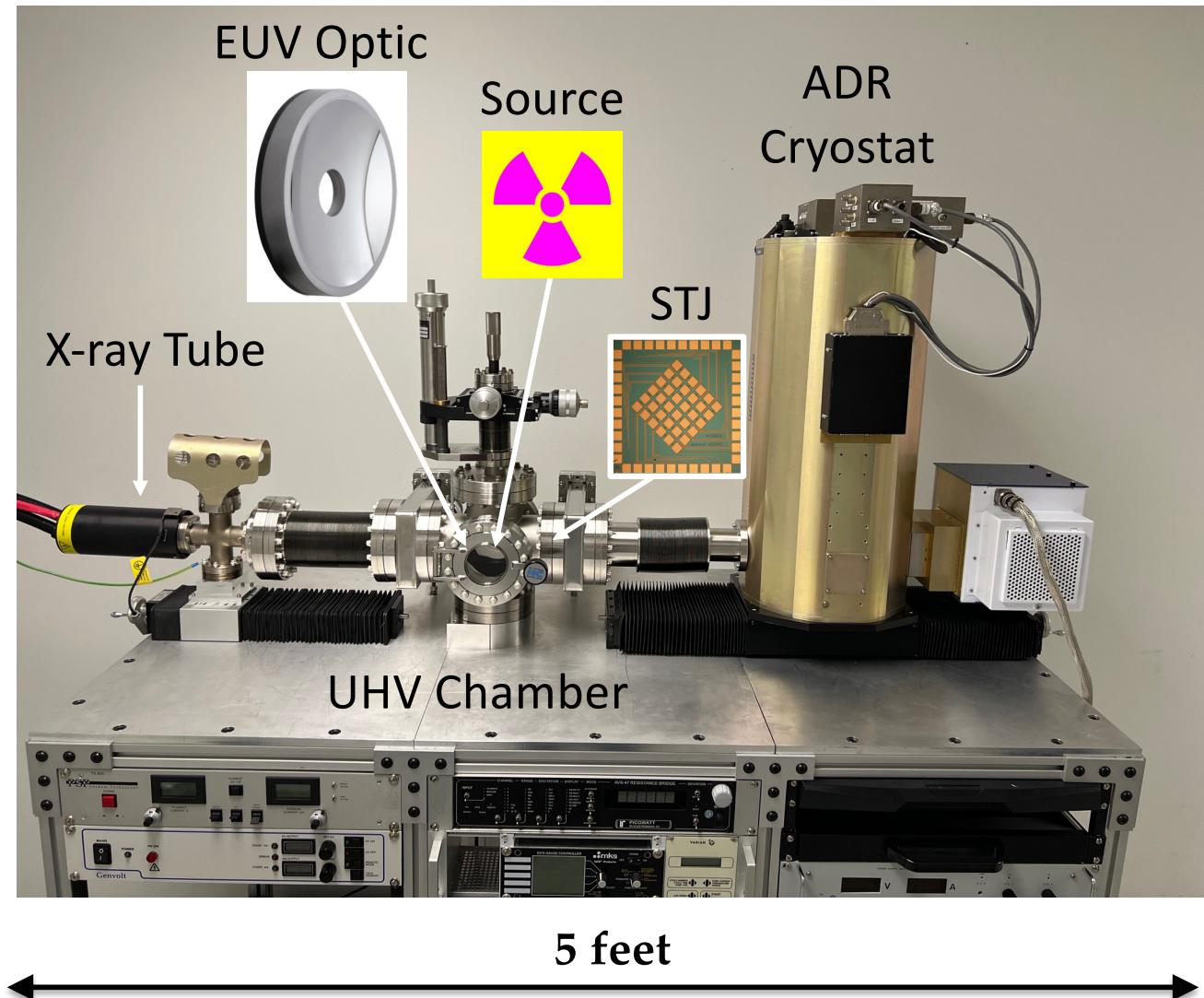
2023: LOI to FRIB PAC

2025-6: tests at LLNL

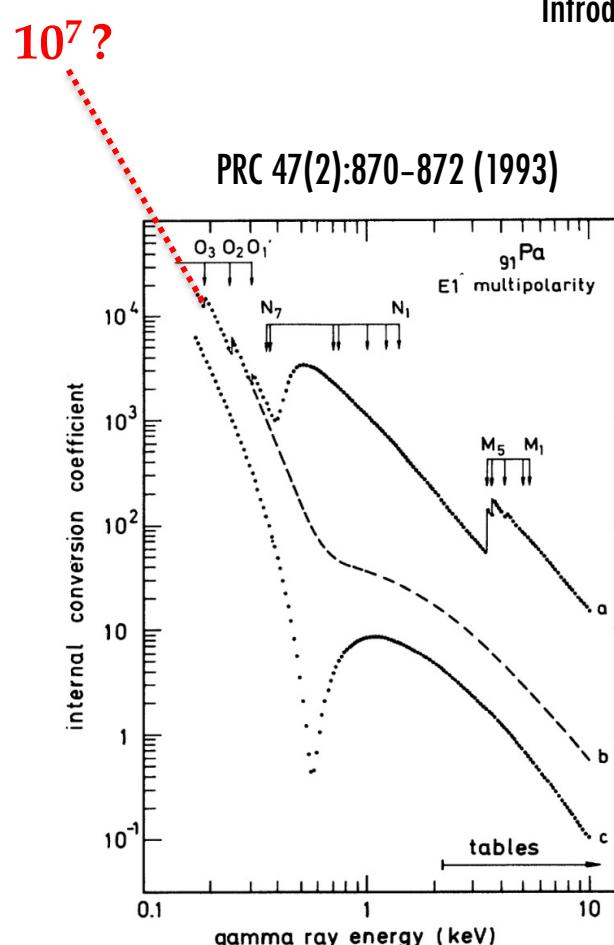
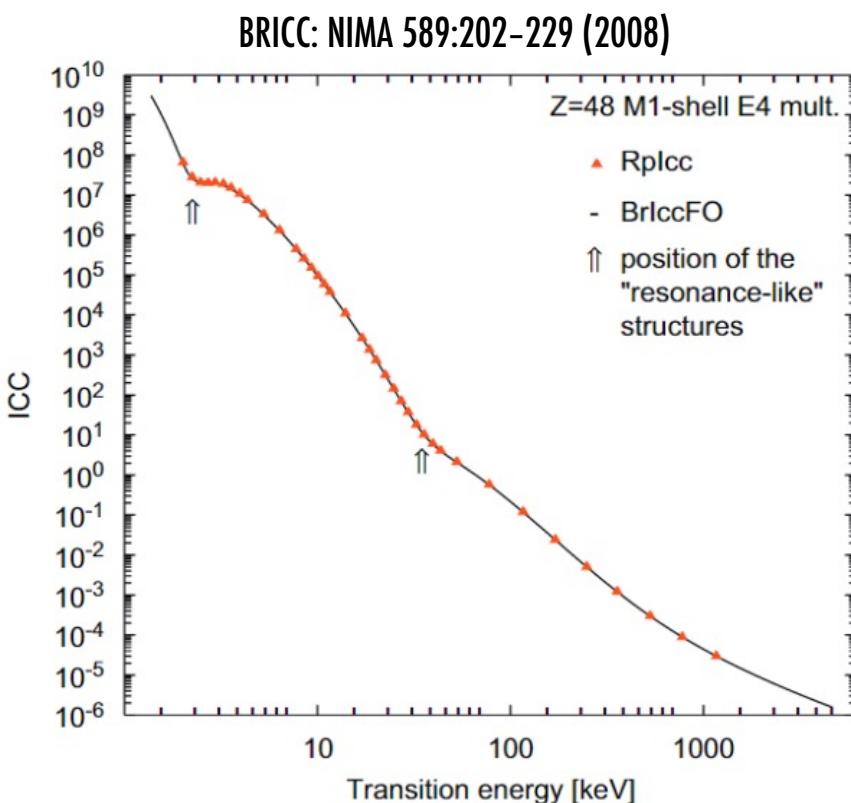
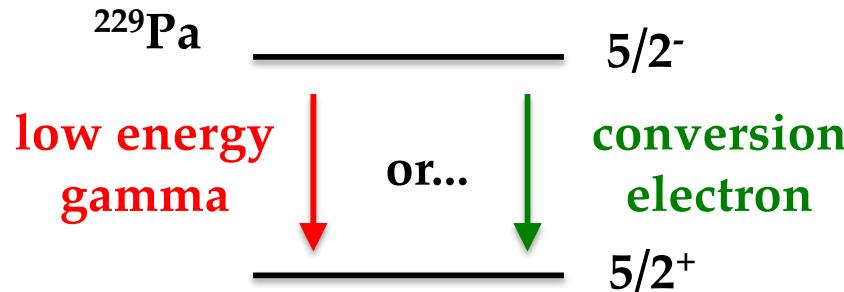
2023-7: GEANT simulations

2027: submit PAC proposal

2027-8: run experiment



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

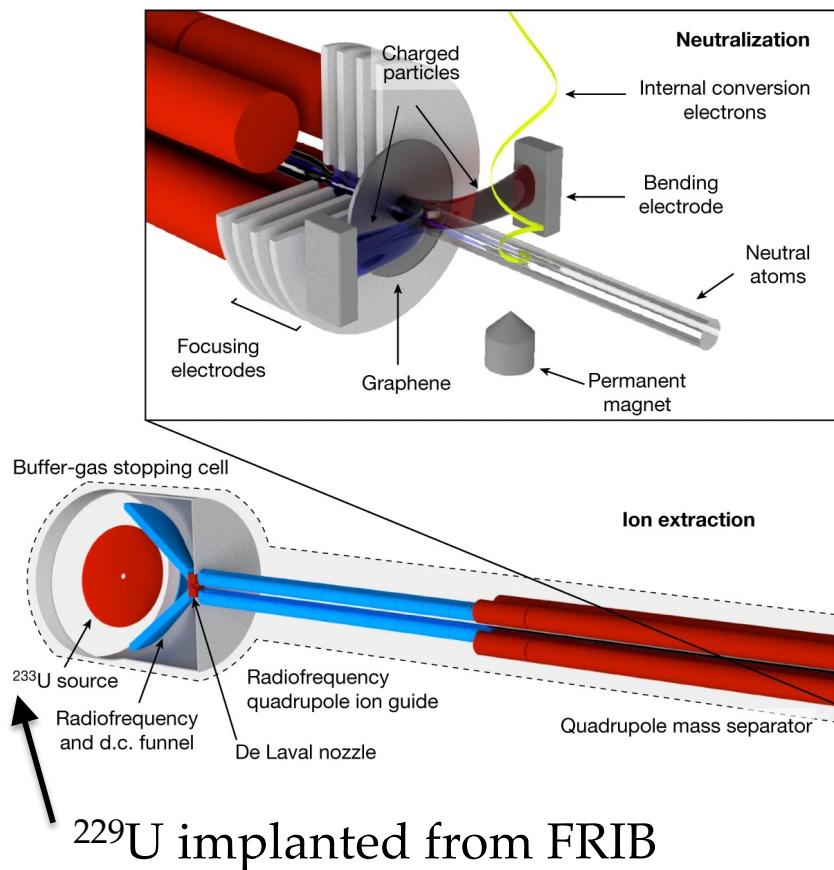


$$\frac{\text{electrons}}{\text{gammas}} \approx \alpha^4 Z^3 \left(\frac{L}{L+1} \right) \left(\frac{2m_e c^2}{\Delta E} \right)^{L+5/2} \approx 10^{18}$$

Introductory Nuclear Physics, 3rd (Krane)

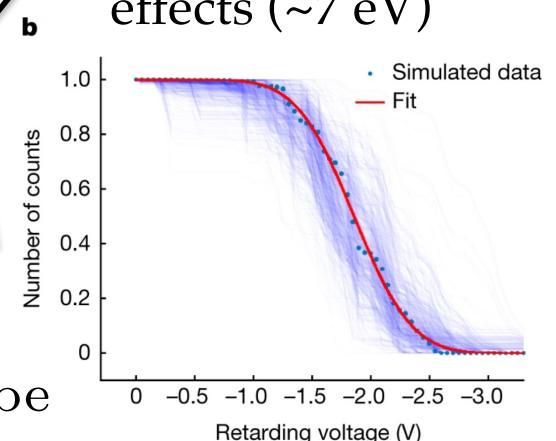
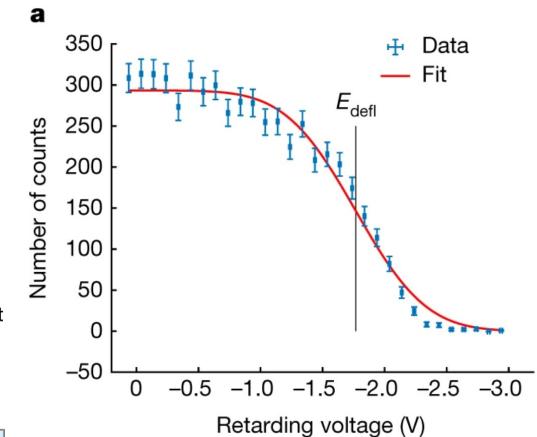
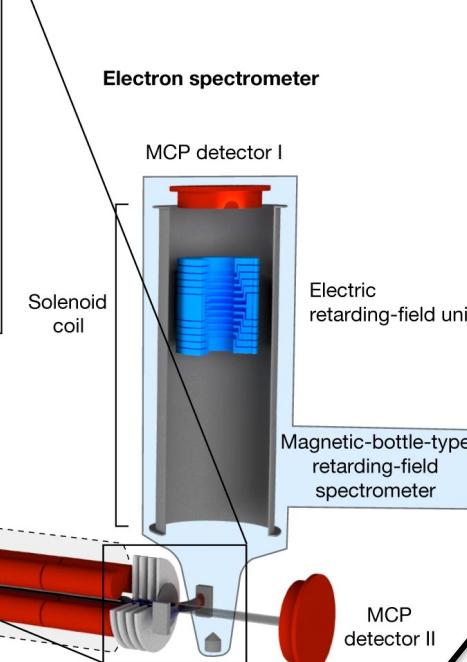
Planned experiments at LLNL using STJs with and without mirror using $^{239}\text{Pu}/^{235m}\text{U}$ source will be used to experimentally measure the ICC for ^{235m}U which is a proxy for ^{229}Pa .

Pending: The ^{229}Th Measurement Technique: Integrated Conversion Electron Spectroscopy



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

measurement



atomic binding effects (~ 7 eV)

We are just looking for nonzero counts and then the order of magnitude of ΔE if a signal is observed. Unlike for ^{229}Th , knowledge of atomic binding effects is not crucial.

Main background: 8.36 eV from ^{229}Th !

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

Embed and Probe ^{229}Pa Ions in Optical Crystals

- Large intrinsic sensitivity to BSM physics
 - **high Z** (^{199}Hg , ^{205}Tl , ^{225}Ra , $^{221,223}\text{Rn}$, ^{229}Pa)
 - **octupole deformed nucleus** (^{225}Ra , $^{221,223}\text{Rn}$, ^{229}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?**

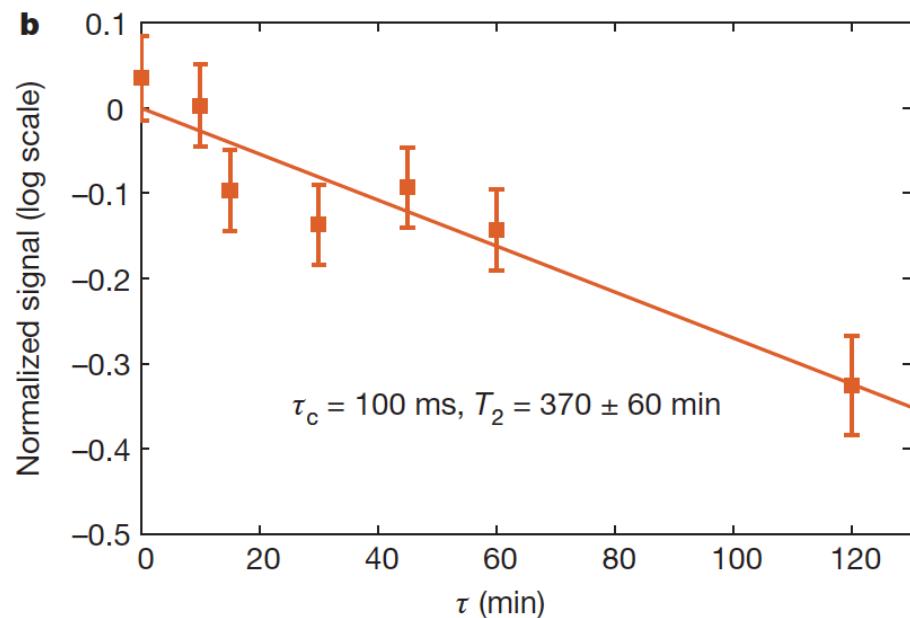
Long Coherence Times of Lanthanide Ion Nuclei

≡ 8 JANUARY 2015 | VOL 517 | NATURE | 177

doi:10.1038/nature14025

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

Manjin Zhong¹, Morgan P. Hedges^{1,2}, Rose L. Ahlefeldt^{1,3}, John G. Bartholomew¹, Sarah E. Beavan^{1,4}, Sven M. Wittig^{1,5}, Jevon J. Longdell⁶ & Matthew J. Sellars¹



Under the right experimental conditions (magnetic field of 1.35 T and temperature of 2 K), using a specially designed pulse sequence (KDD_x), the T₂ of ¹⁵¹Eu³⁺ (I=5/2) embedded in Y₂SiO₅ was measured to be over 6 hours.

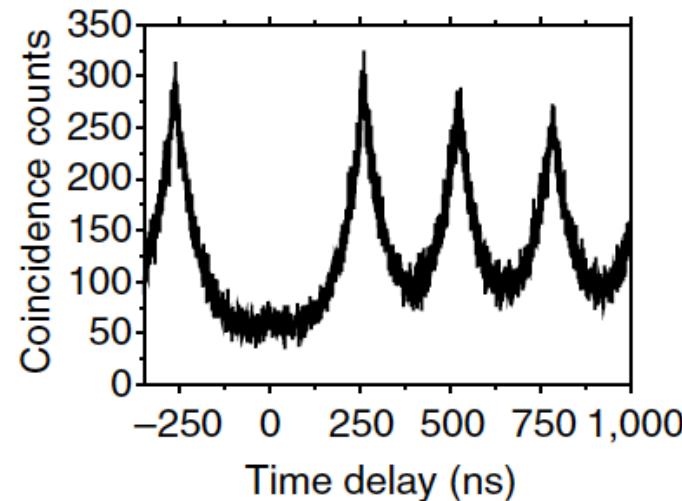
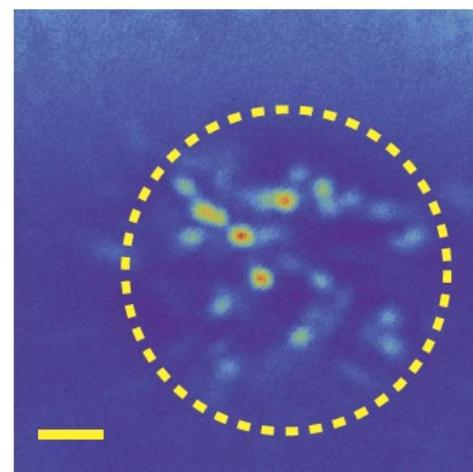
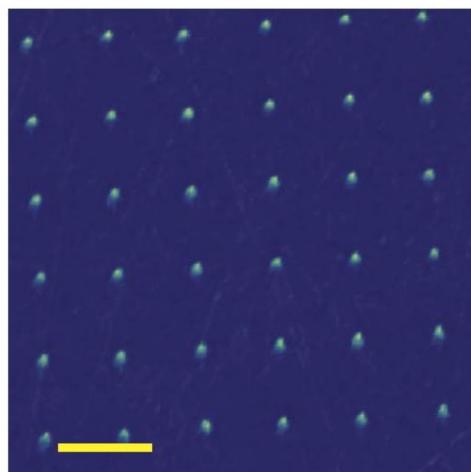
Single Ion Implantation & Manipulation

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

DOI: [10.1038/ncomms4895](https://doi.org/10.1038/ncomms4895)

Coherent properties of single rare-earth spin qubits

P. Siyushev^{1,*}, K. Xia^{1,*}, R. Reuter¹, M. Jamali¹, N. Zhao², N. Yang³, C. Duan⁴, N. Kukharchyk⁵, A.D. Wieck⁵, R. Kolesov¹ & J. Wrachtrup¹



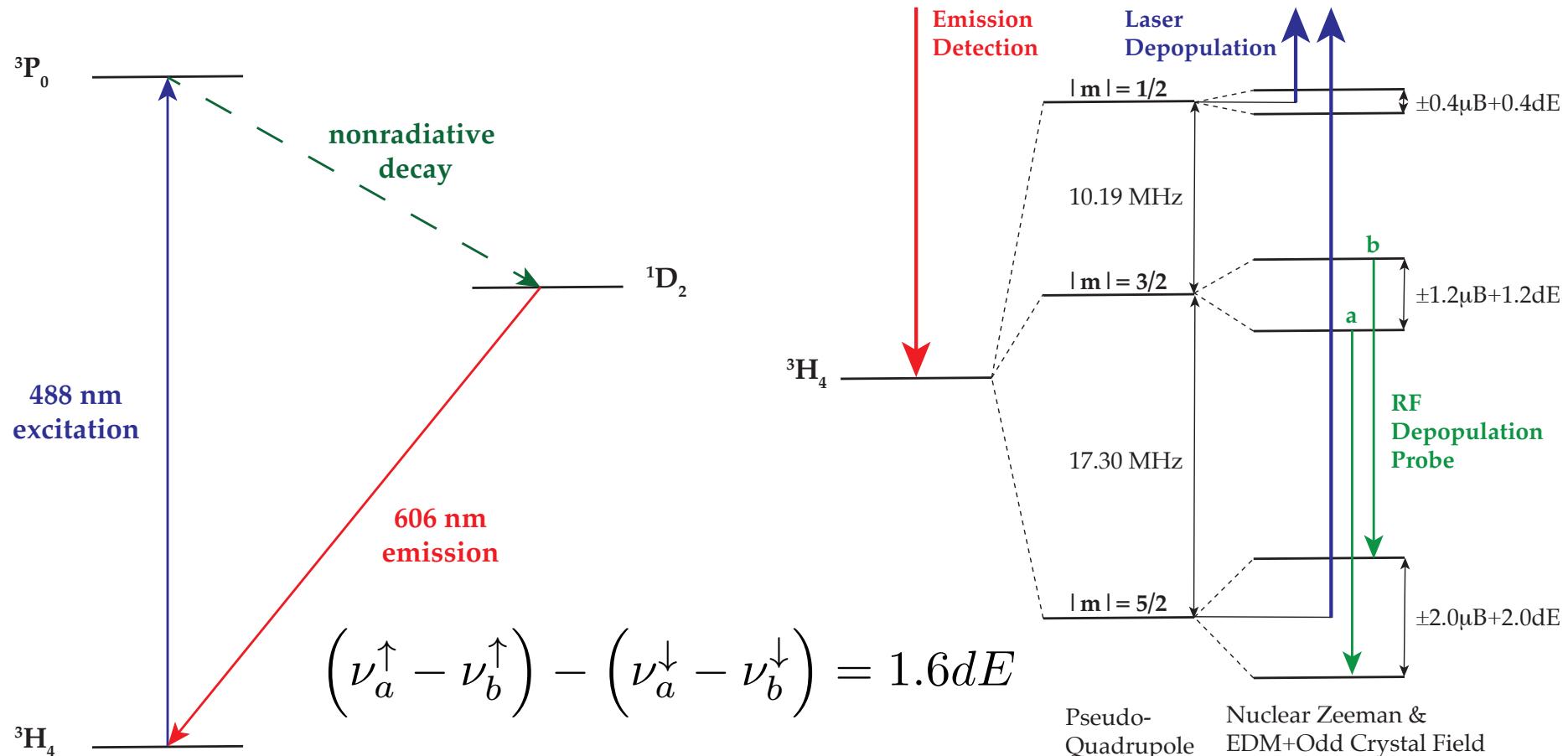
Left: well-controlled ion implantation of Ce³⁺ (yellow bar = 10 microns)

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)



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

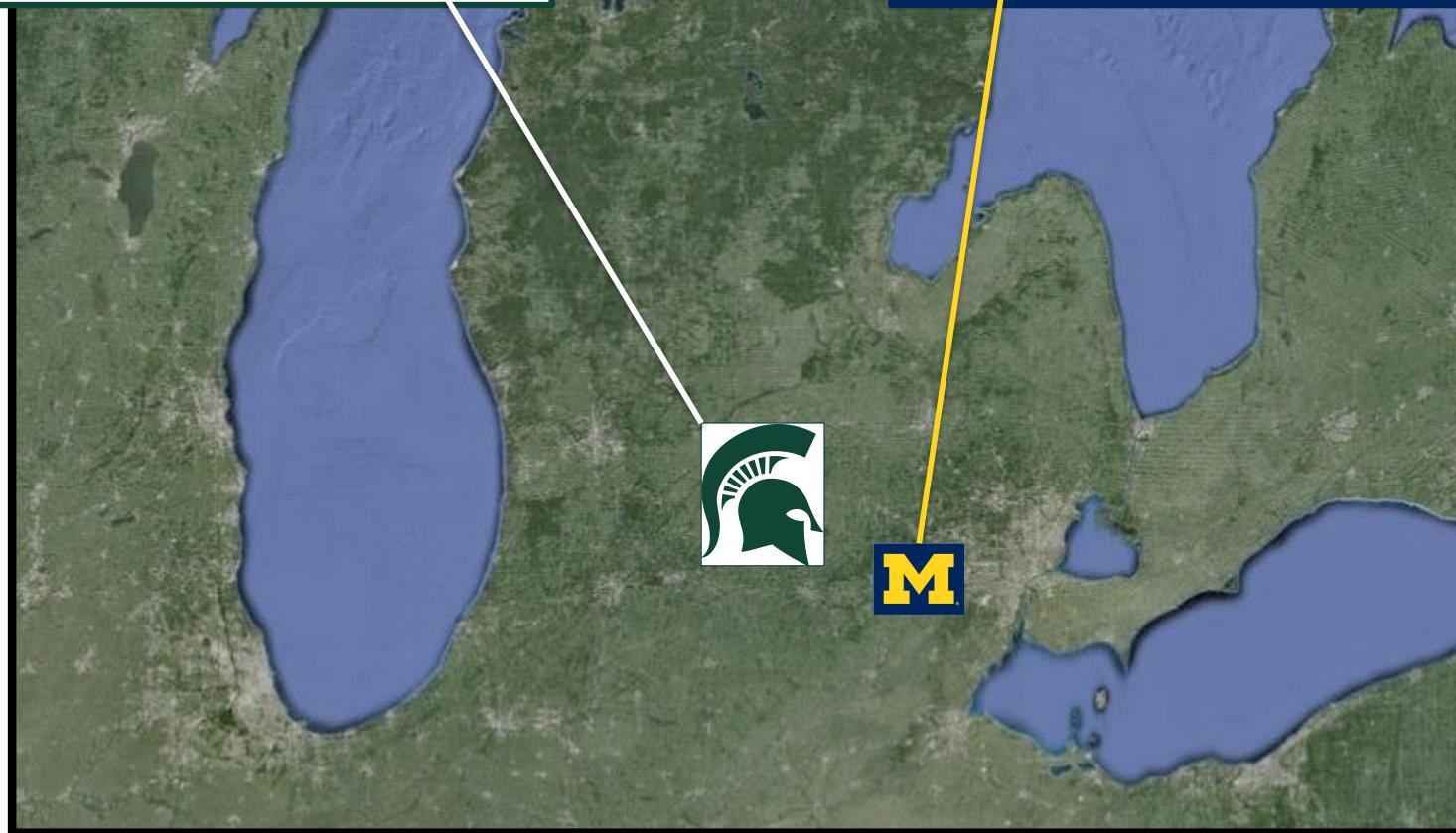


Google Maps & Wikipedia Commons

Facility for Rare Isotope Beams @ MSU

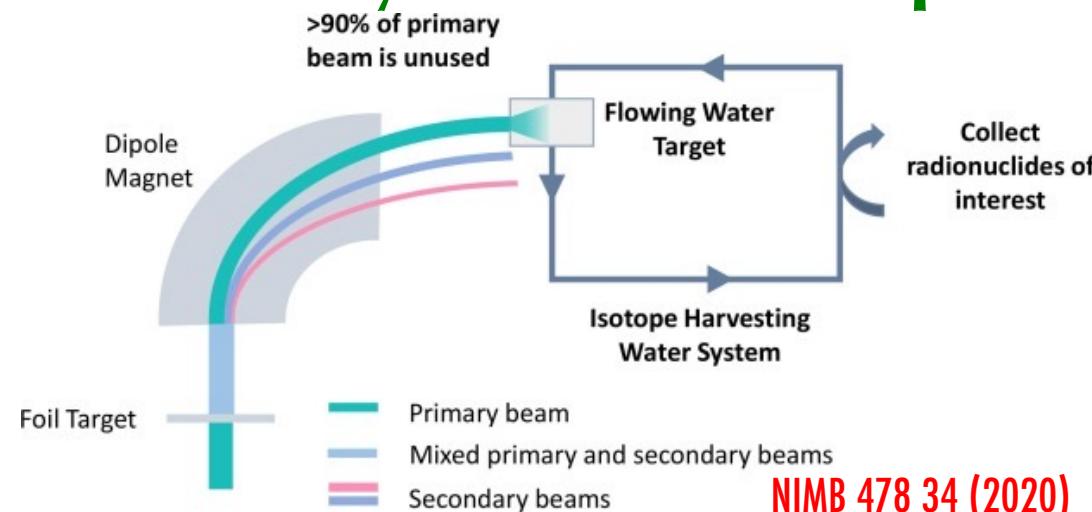
Michigan State University
East Lansing, MI
Very Bad at American Football
Home of FRIB

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

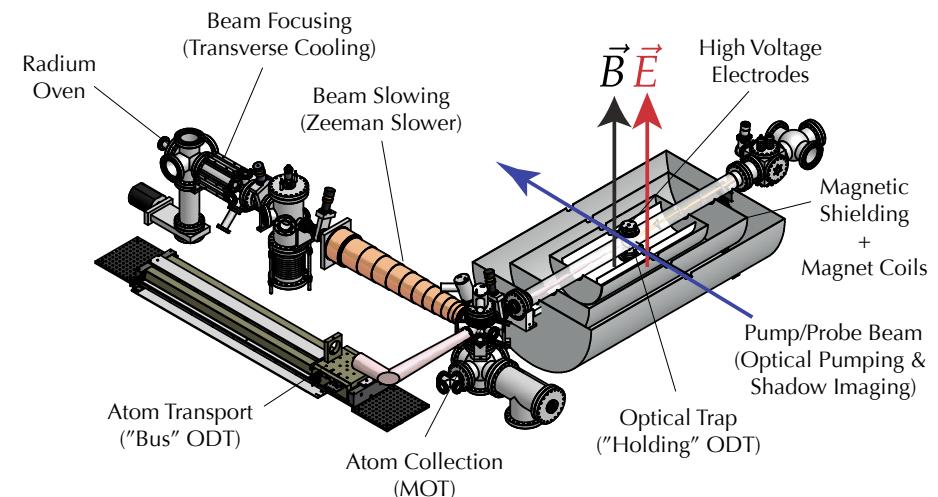
Isotope harvesting at FRIB: additional opportunities for scientific discovery [J. Phys. G: Nucl. Part. Phys. 46 100501 (2019)]



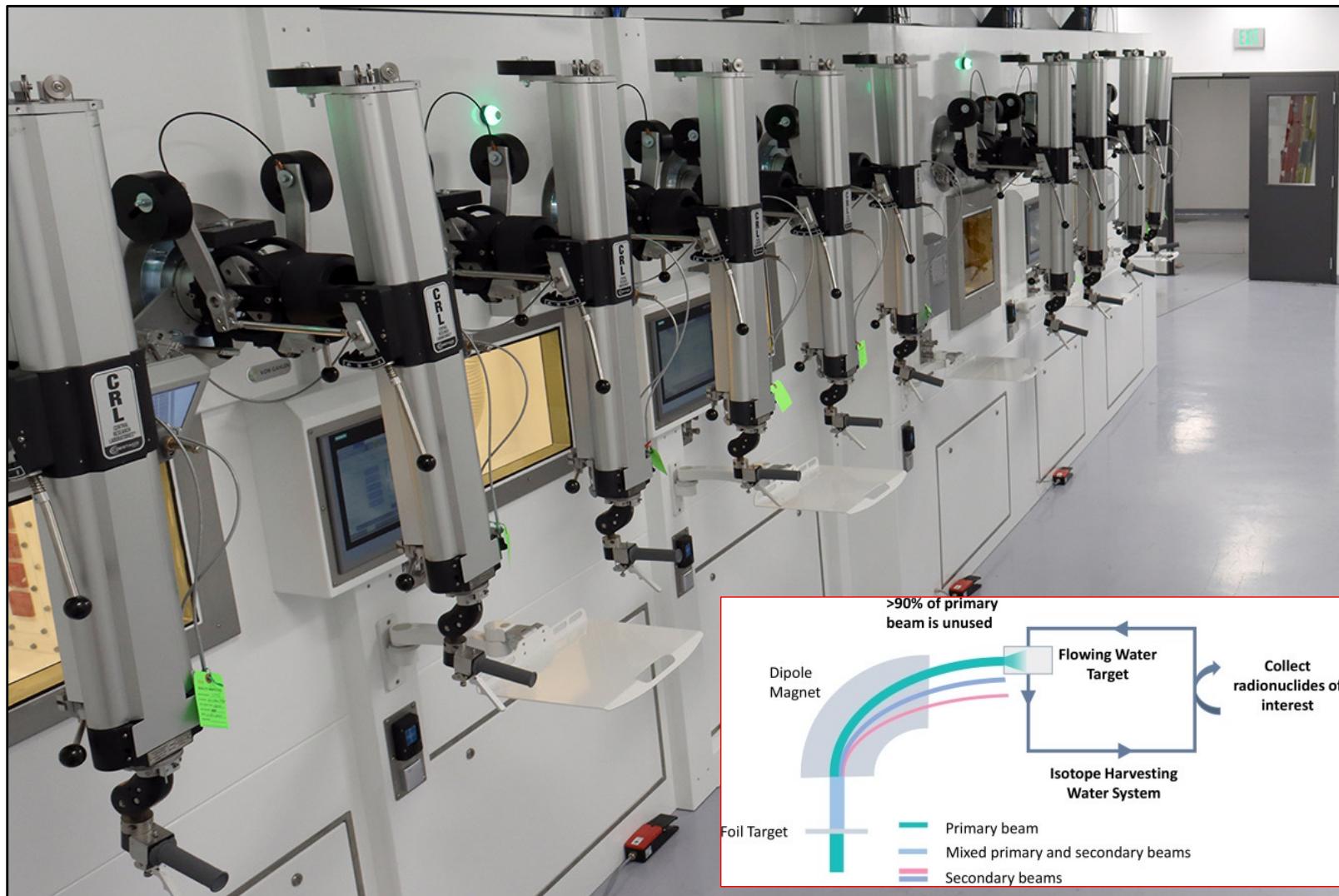
see talks by
G. Arrowsmith-Kron
A. Boyer



Adobe Stock



Isotope Harvesting Vault Is Installed



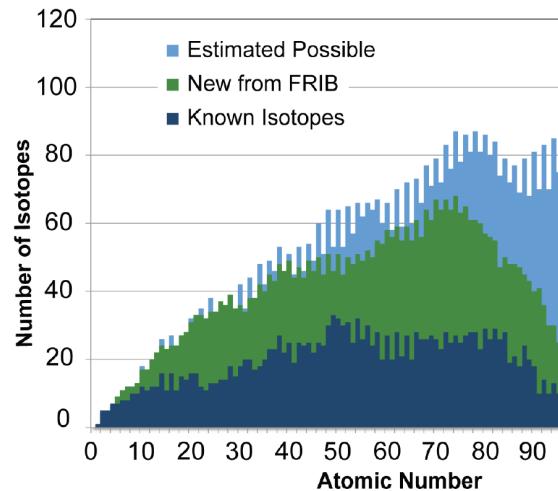
C. Vyas

NIMB 478 34 (2020)

The Radiochemistry Team at FRIB/MSU



Prof. Greg Severin

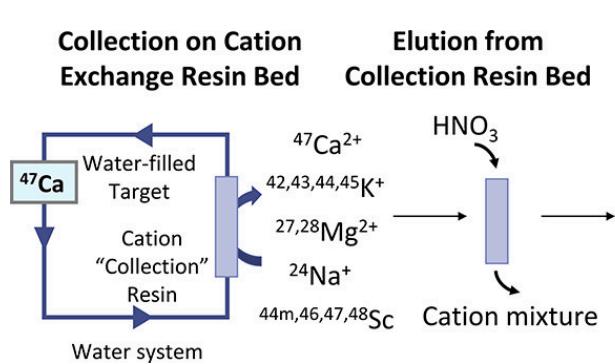


Nature 486, 509–512 (2012)

2025-03-15



Prof. Alyssa Gaiser



Prof. Katharina Domnanich

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

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

PMLE Pa-229 (JTS)

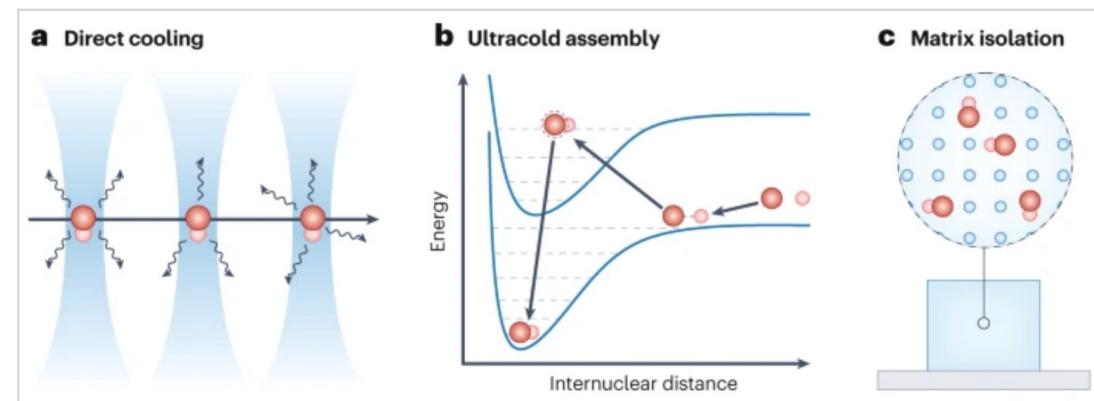
58

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

Enhancements: nuclear Schiff moment enhancement of x1000 (^{225}Ra) to maybe(!?!) x1000000 (^{229}Pa)
and $\sim 100 \text{ MV/cm}$ effective internal E -field (lab $< 1 \text{ MV/cm}$)
[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.

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



Nature Physics 20, p741-749 (2024)

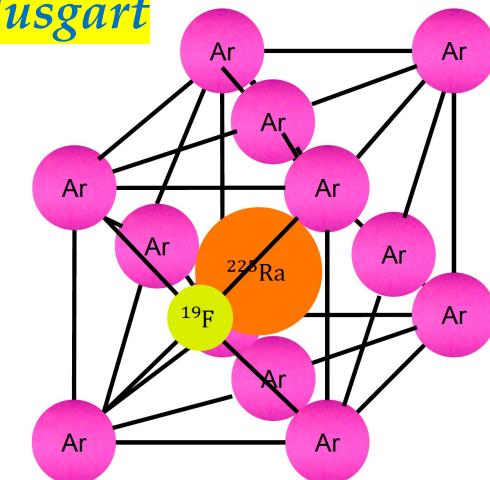
Challenges:

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

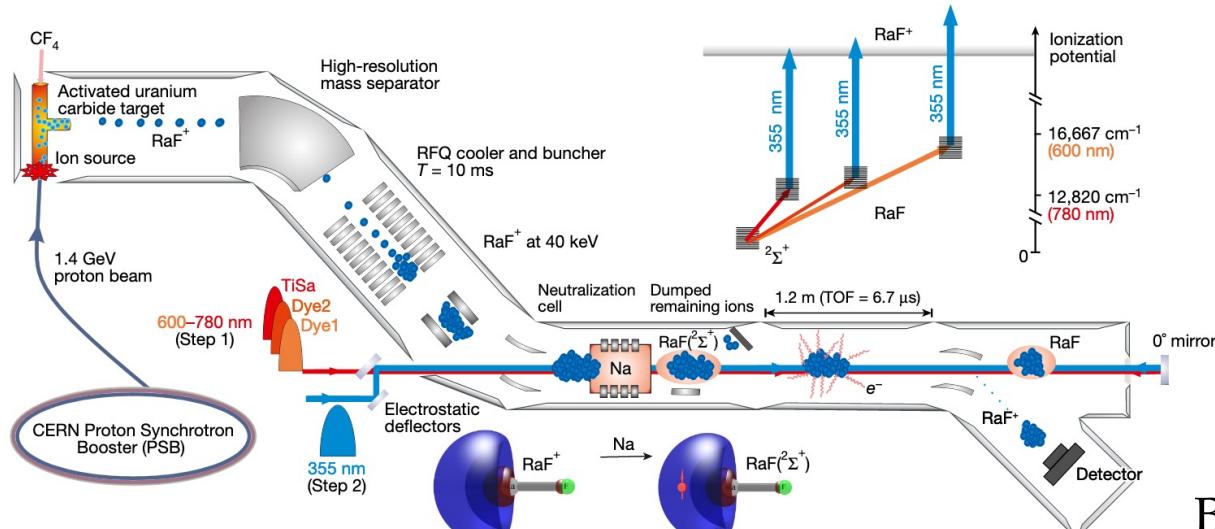
- Efficient trapping of a wide variety of species
- Very high number densities
- Stable and chemically inert confinement
- Transparent in the optical regime for optical probing
- Under certain conditions, polar molecules orient themselves along the crystal axes which allows for control of systematics: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

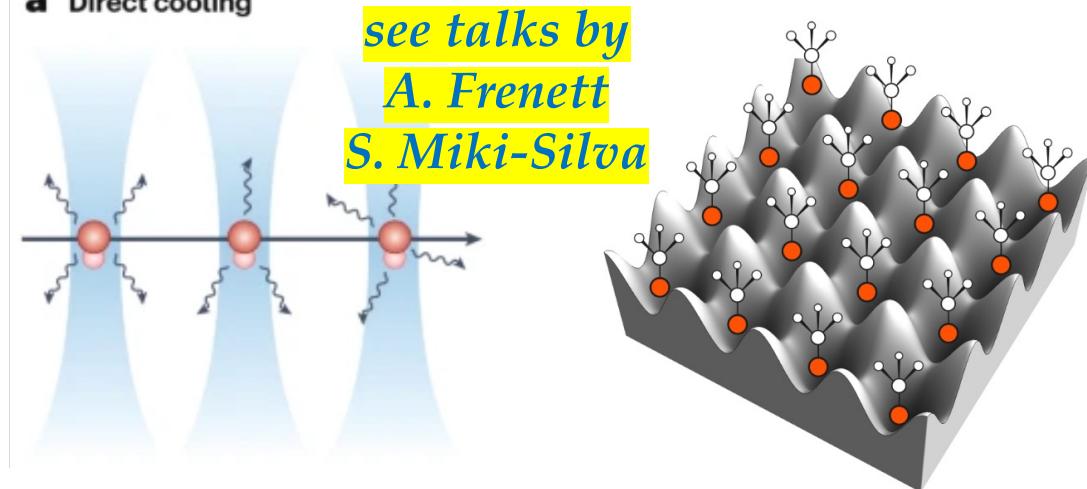


Direct Laser Cooling of Neutral Molecules Into a Laser Trap

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



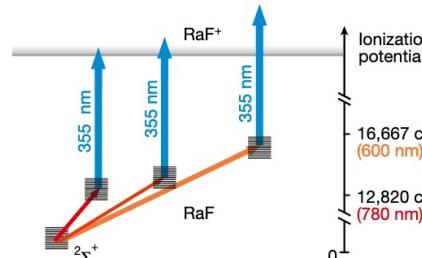
a Direct cooling



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

2025-03-15

PMLE Pa-229 (JTS)



- 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 CaOCH₃

PRL 119, 133002 (2017)

Quantum Science & Tech. 5, 044011 (2020)

Science 369, 1366–1369 (2020)

Nature Physics 20, p741–749 (2024)

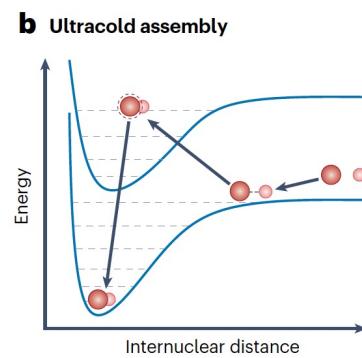
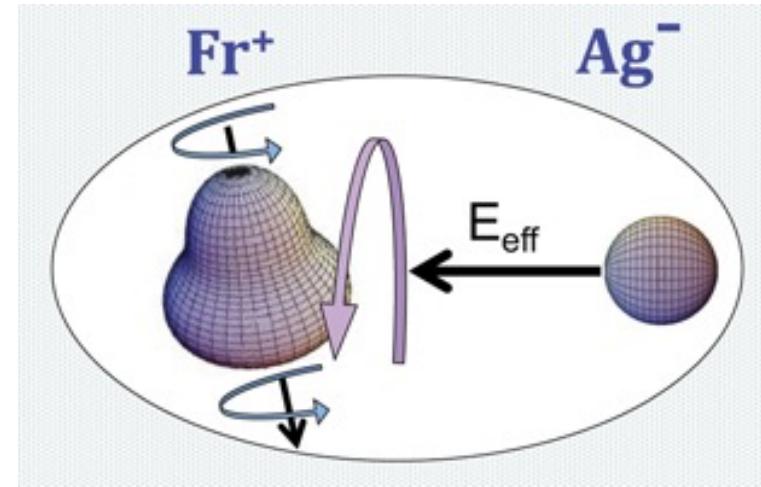
61

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

Gen-I Estimate:

⇒ ~1000x projected improvement
vs. ^{199}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



slide from D. DeMille
(UChicago/Argonne)

Nature Physics 20, p741–749 (2024)

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

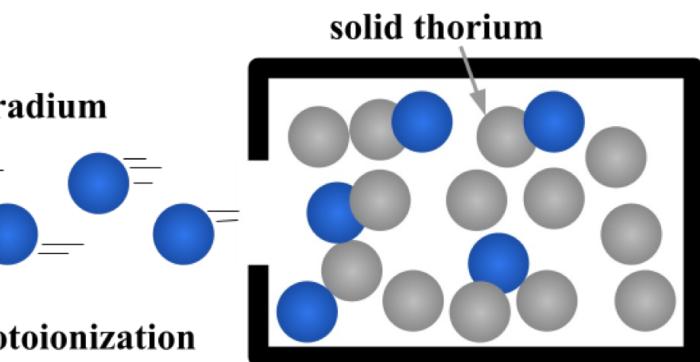
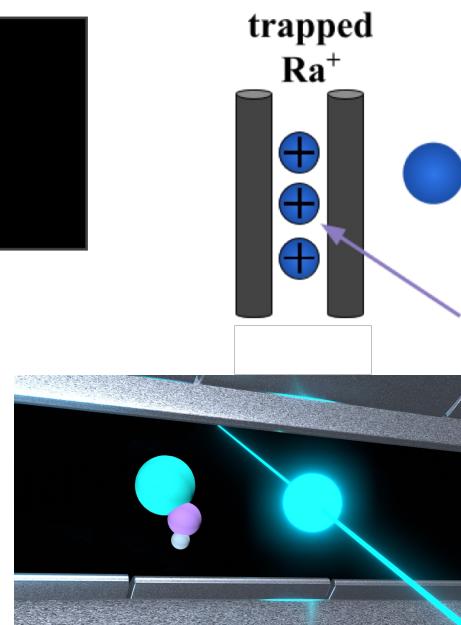
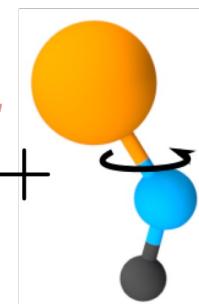
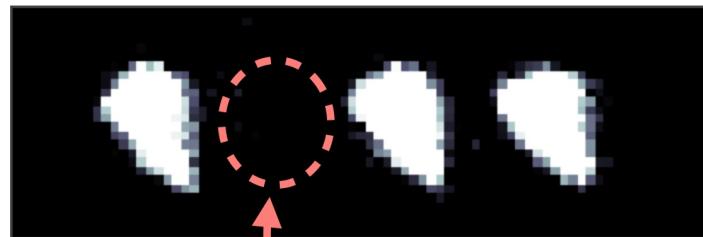
Theory calculations favorable:

New J. Phys. 23 113039 (2021)

New J. Phys. 24 025005 (2022)

odd-proton nuclei like ^{223}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)



Santra et al., PRA 90, 040501(R) (2014)

- Spectroscopy and atomic structure measurements of the logic ion 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

Connecting New Physics to EDMs

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

Fundamental theory

$\bar{\theta}$ CKM SUSY Multi Higgs LR-symmetry etc.

Wilson coefficients

$C_{ggg}, C_{qqqq}(1,8), C_{qH}, d_{ud}, \tilde{d}_{ud}$ semileptonic d_e

Low energy parameters

$g_\pi^0, g_\pi^1 (g_\pi^2)$ d_n, d_p

Nucleus level

$d, t, {}^3\text{He}$ Schiff moment

Atom/molecule level

Diamagnetic Paramagnetic

Solid state

Sources of CP -violation

Particle Physics Theory

Effective Field Theory

Lattice QCD Theory

Nuclear Theory

+

Nuclear Experiment

Atomic Theory

+

Atomic Experiment

Molecular Theory

+

Molecular Experiment

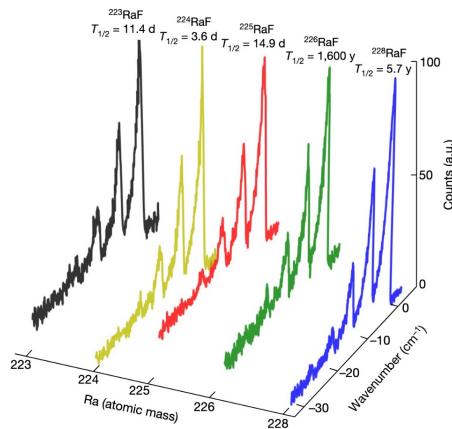
Radiochemistry

...EDMs

The Nuclear Pear Factory: A Proposed Center

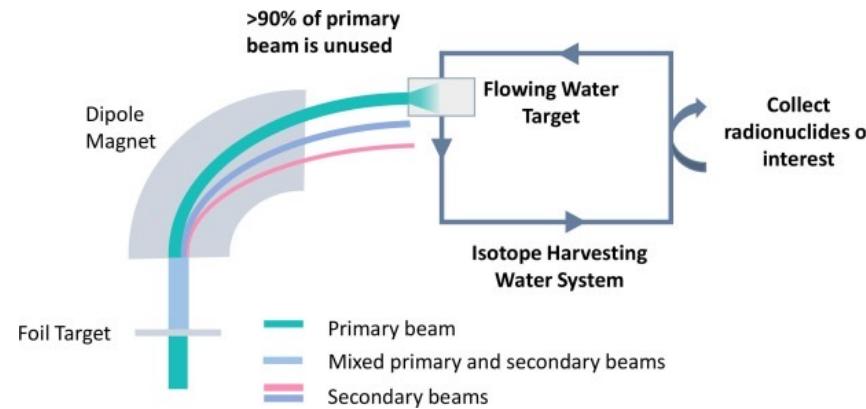


Nature 497:199 (2013)

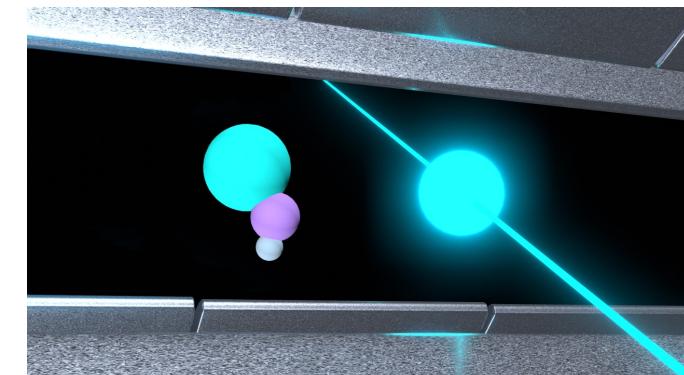
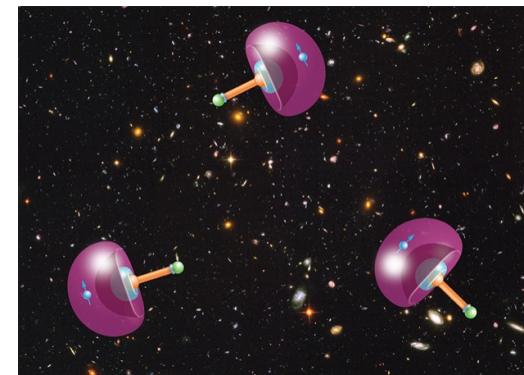


Nature 581:396 (2020)

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.

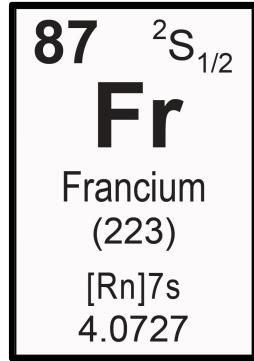
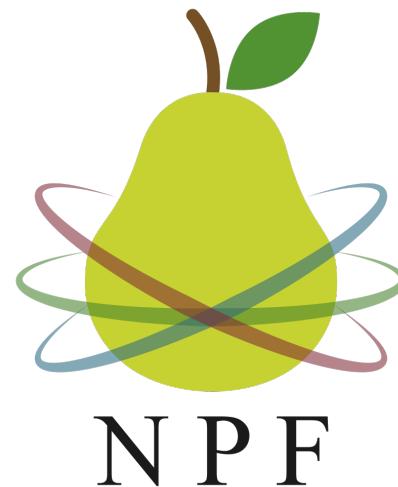
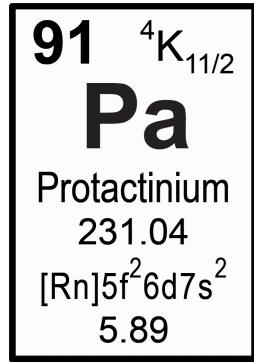
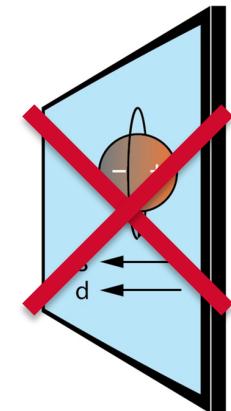
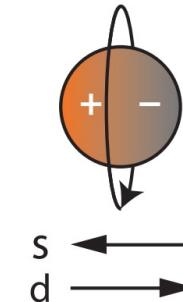
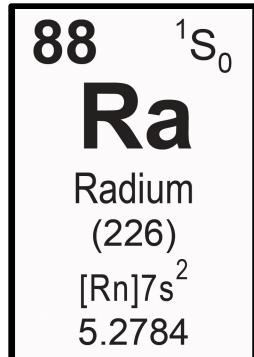


NIMB 478 34 (2020)

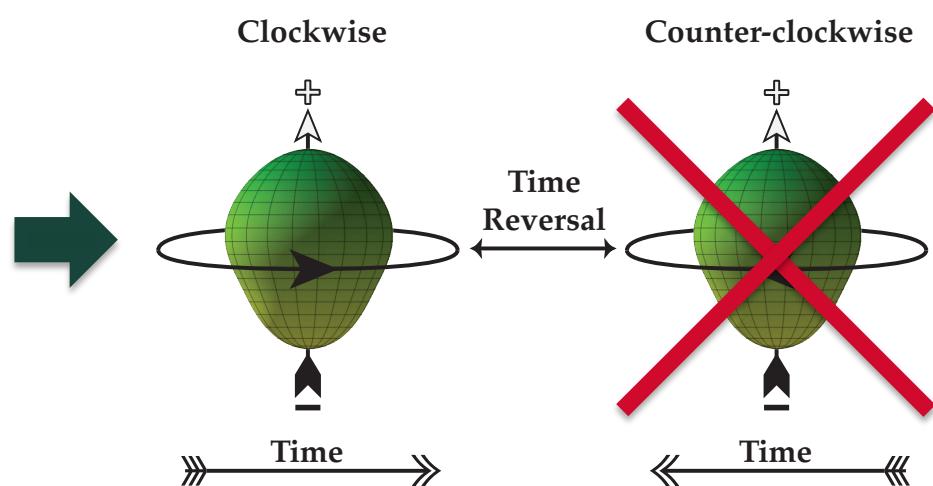


<https://physics.aps.org/articles/v14/103> & A.M. Jayich

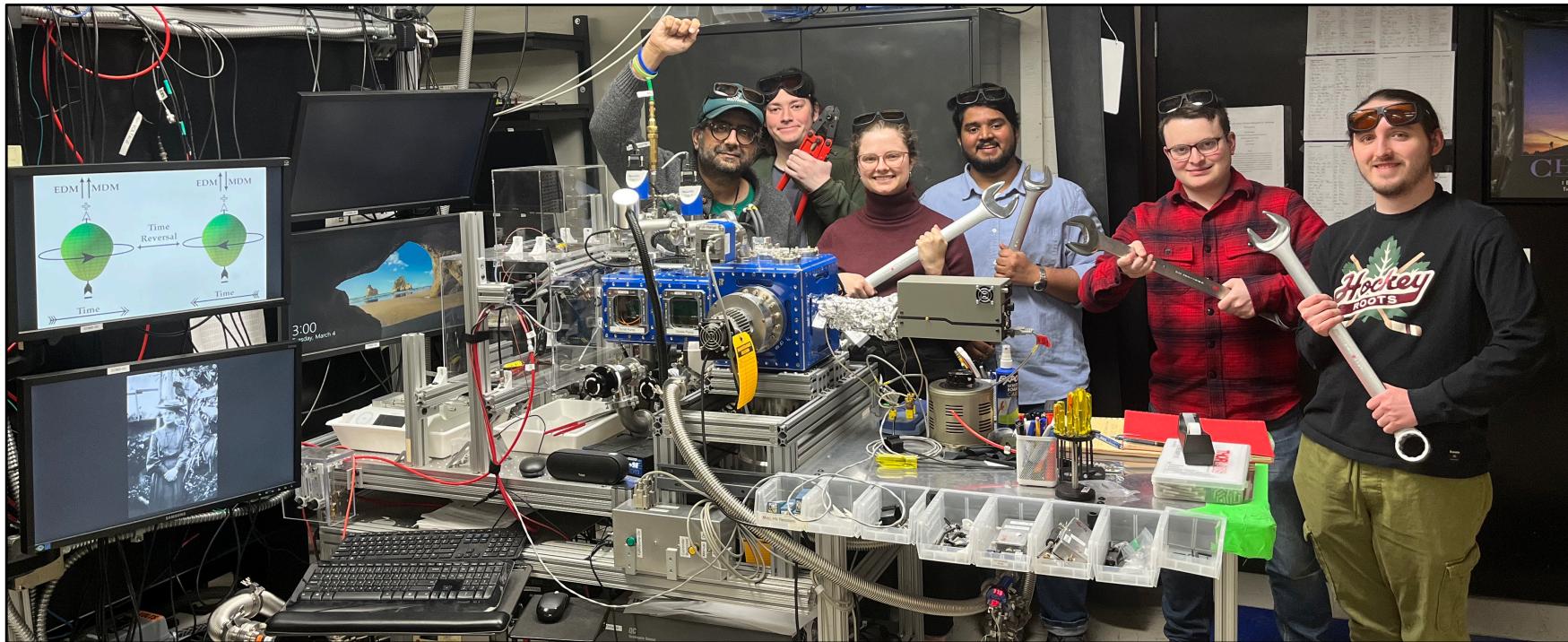
We Are Following In The Footsteps Of Giants Towards A Transformational Discovery Within Our Student's Lifetime!



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. Hessel
@ York (Canada):
GBMF8863
G-2019-12503