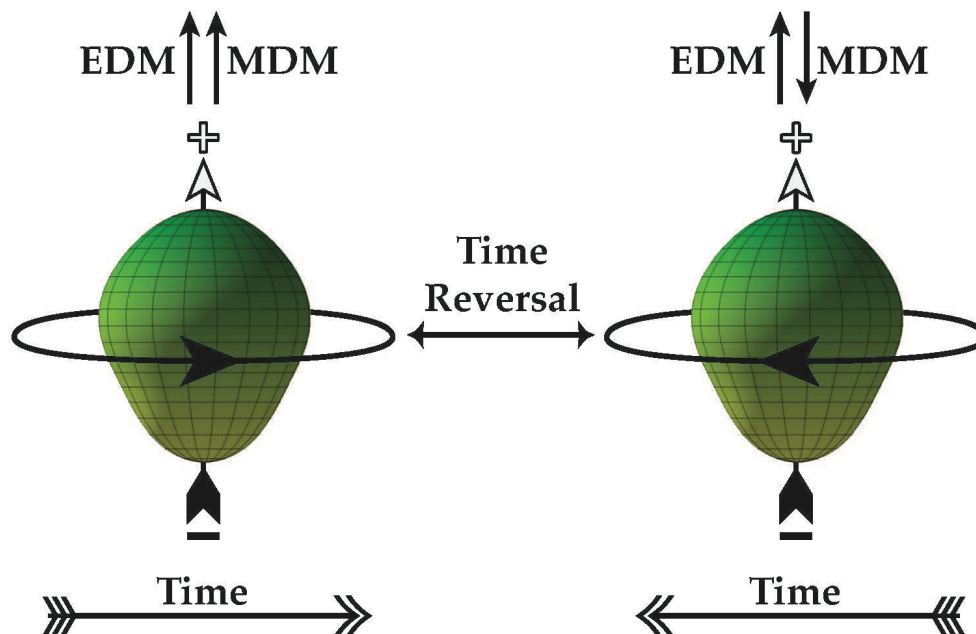


The Nuclear Pear Factory: Searching for Time-Reversal Violation Using Pear-Shaped Nuclei in the FRIB Era



Jaideep Taggart Singh

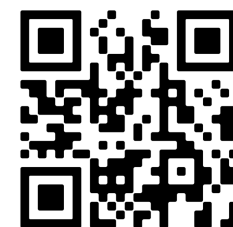
Michigan State University / FRIB

DNP T1-6: 0900-0930, June 30, 2023

2023 CAP Congress, Fredericton, NB, Canada

University of New Brunswick, UNB Kinesiology (Rm. 201)

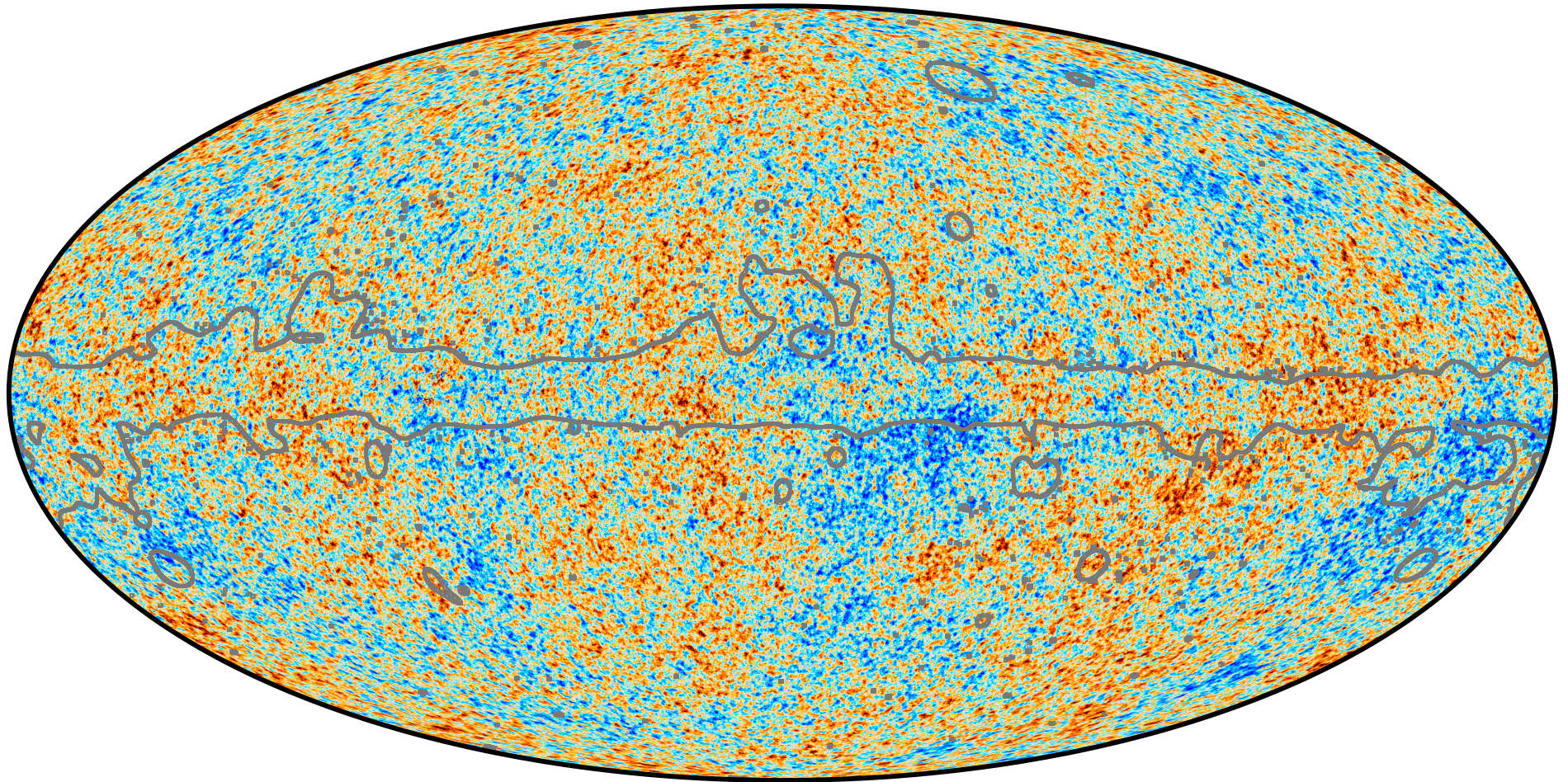
Marie-Anne
Bouchiat



SCAN ME



Cosmic Microwave Background Anisotropy: More matter than antimatter in the visible universe



Planck 2018

-300

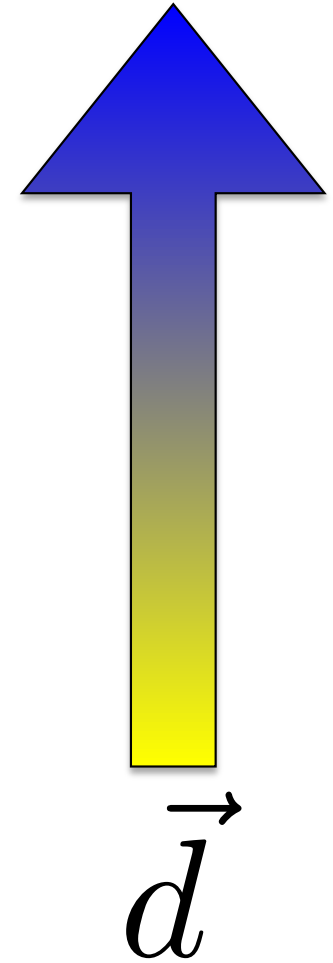


300 μK

~ 100 ppm
fluctuations

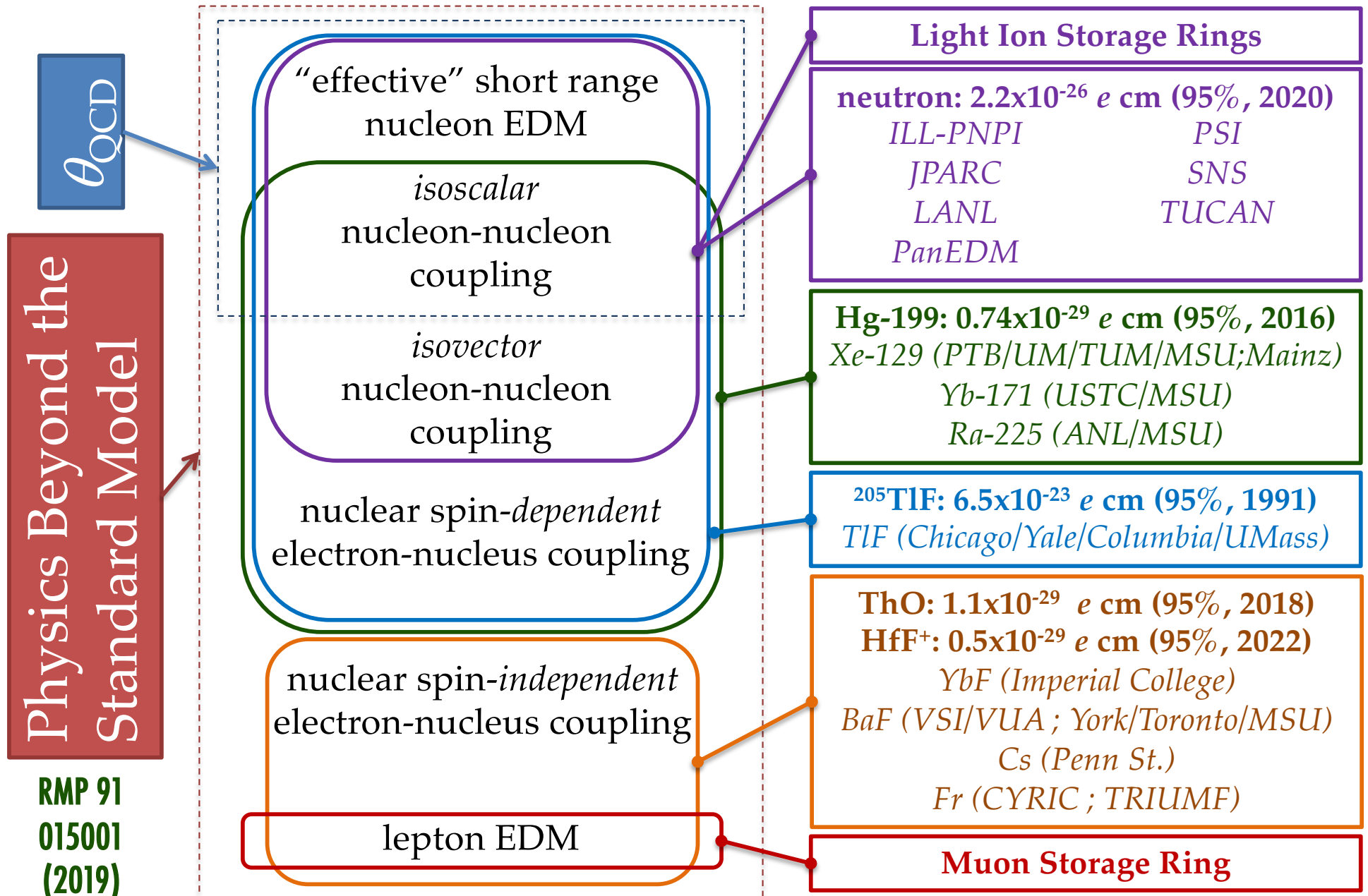
<https://www.cosmos.esa.int/web/planck/picture-gallery>

EDM: Measures the Separation of Charges



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

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

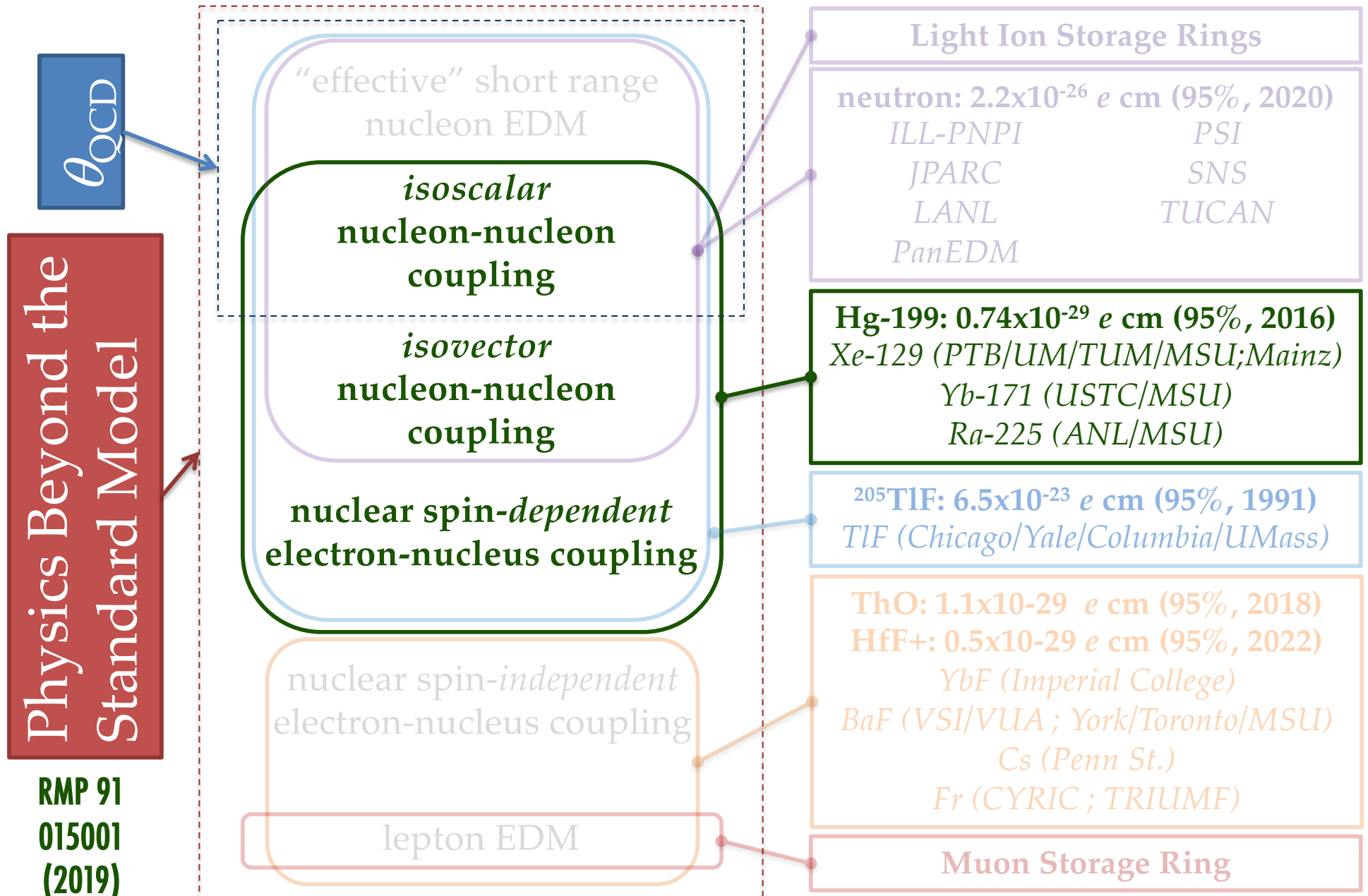


RMP 91
015001
(2019)

2023-06-30

2023 CAP Congress, Fredericton, NB

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



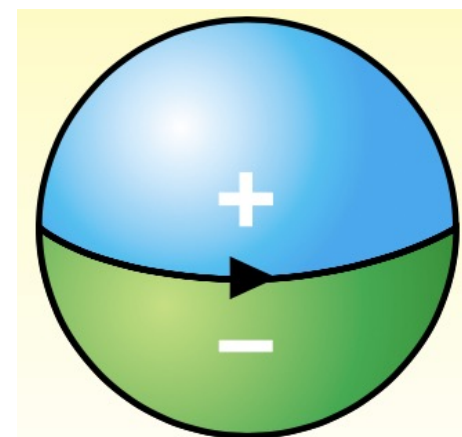
2023 EDM Limits: Free of SM “Backgrounds”

Chupp, Fierlinger, Ramsey-Musolf, JTS RMP 91:015001 (2019) & Nature 562:355 (2018) & PRL 124:081803 (2020) & arxiv:2212.11841

System	Best Limit (95%) 1E-28 <i>e</i> cm	SM estimate 1E-28 <i>e</i> cm	Method (Location)
Neutron	220	$\sim 10^{-4}$	ultracold neutrons in a bottle (PSI)
“Electron”	0.11 0.05	$\sim 10^{-10}$	cold ThO beam (Chicago / Harvard / Northwestern) trapped HfF ⁺ (JILA / Boulder)
Hg-199	0.074	$\sim 10^{-6}$	atoms in vapor cell (UW-Seattle)

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

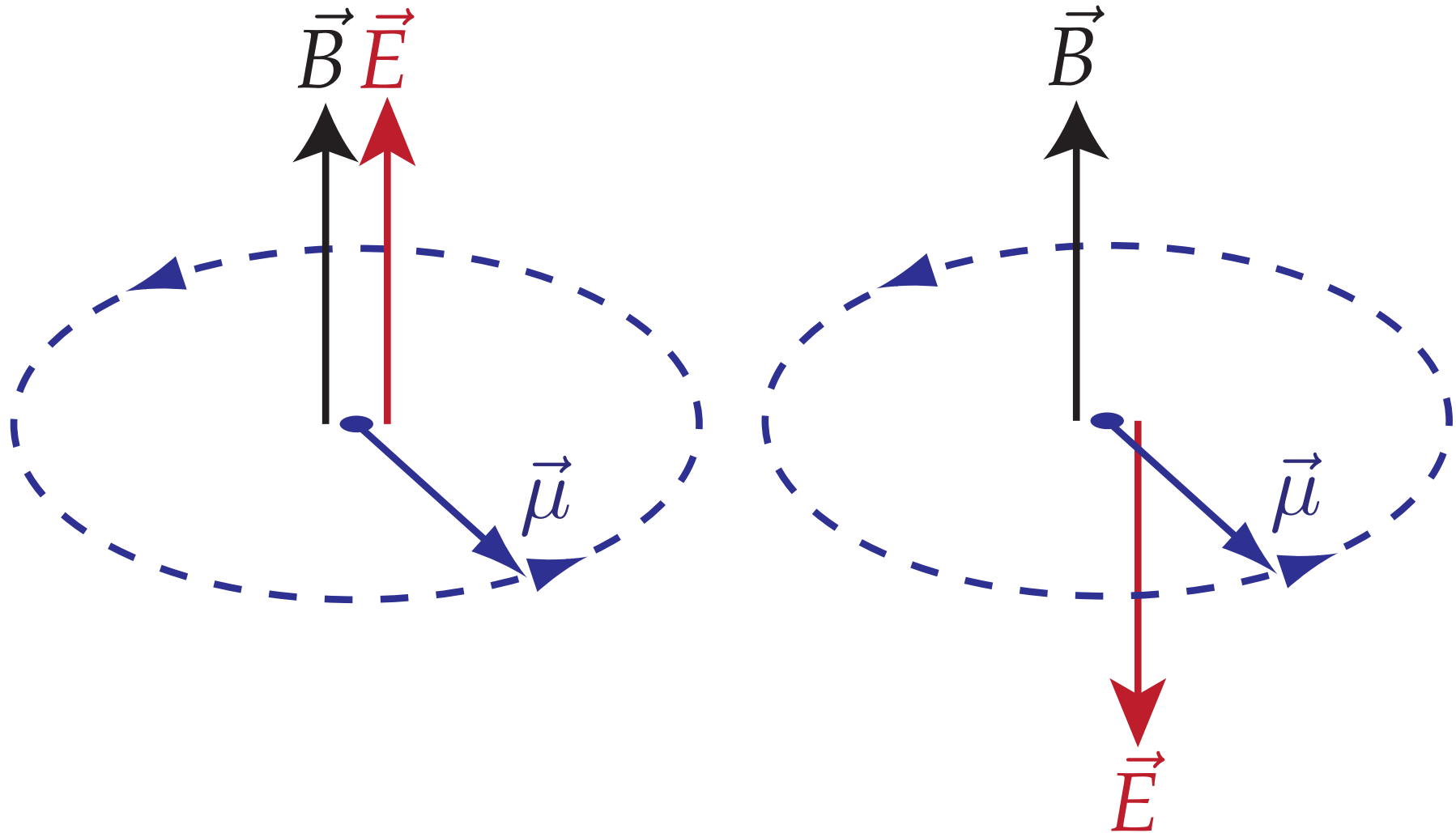
If the Hg-199 atom was the size of the Earth, then the maximum thickness of these shells would be less than the diameter of a strand of human hair.



Physics Today, June 2003

Always Measure Frequency

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



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

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

Statistical Sensitivity

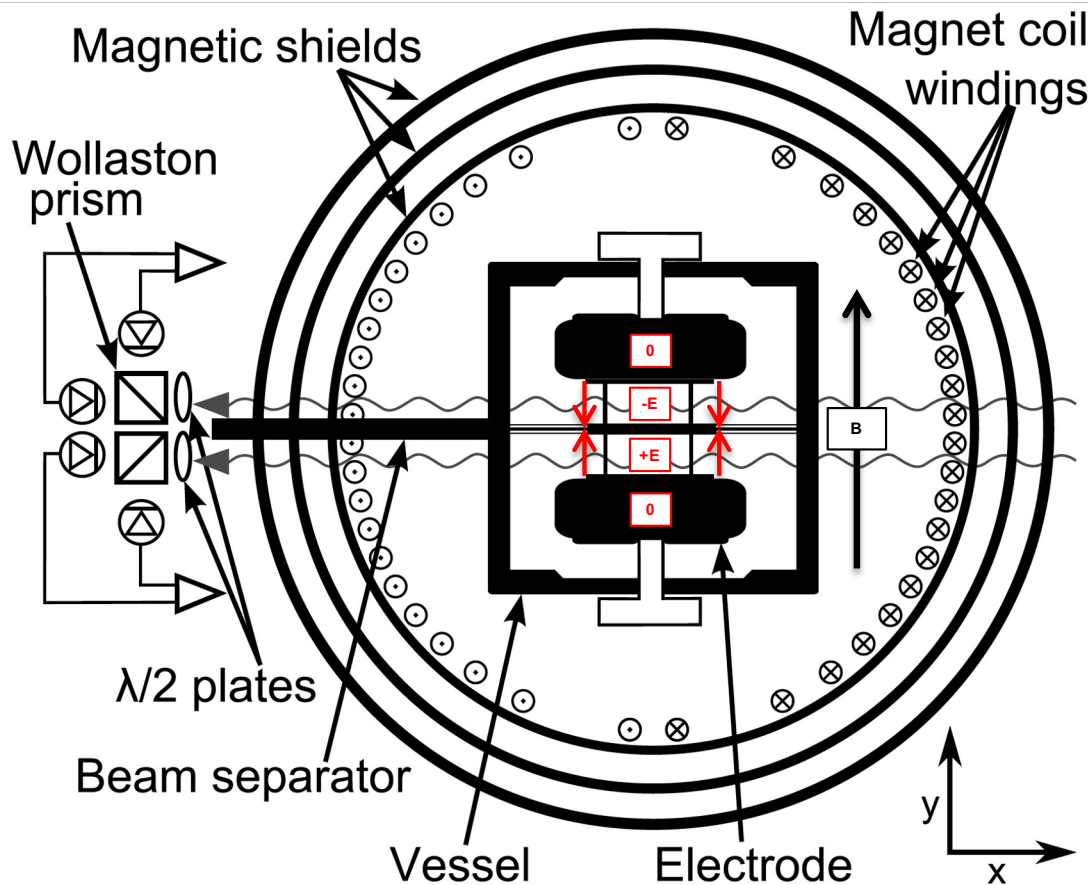
$$\Delta\nu = \nu_{\uparrow} - \nu_{\downarrow} = \frac{4dE}{h} \quad \sigma_{\nu} = \frac{\Gamma_{\text{linewidth}}}{\text{SNR}}$$

Quantum Projection Noise:

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

Electric field number of detected particles integration time interrogation time

The Gold Standard: Hg-199 EDM Search



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

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

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

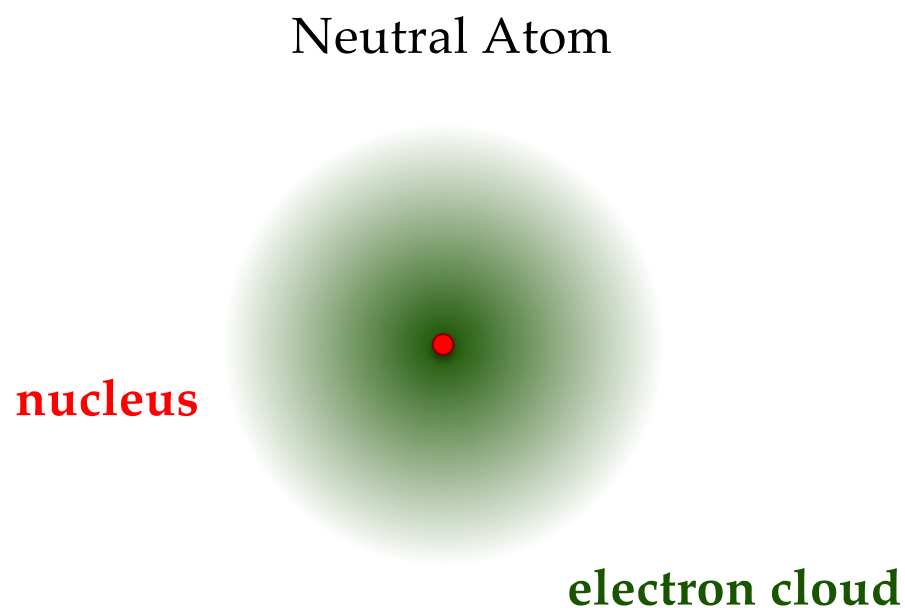
$$\Delta\nu \leq 0.1 \text{ nHz}$$

The best limit on atomic EDM:

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

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

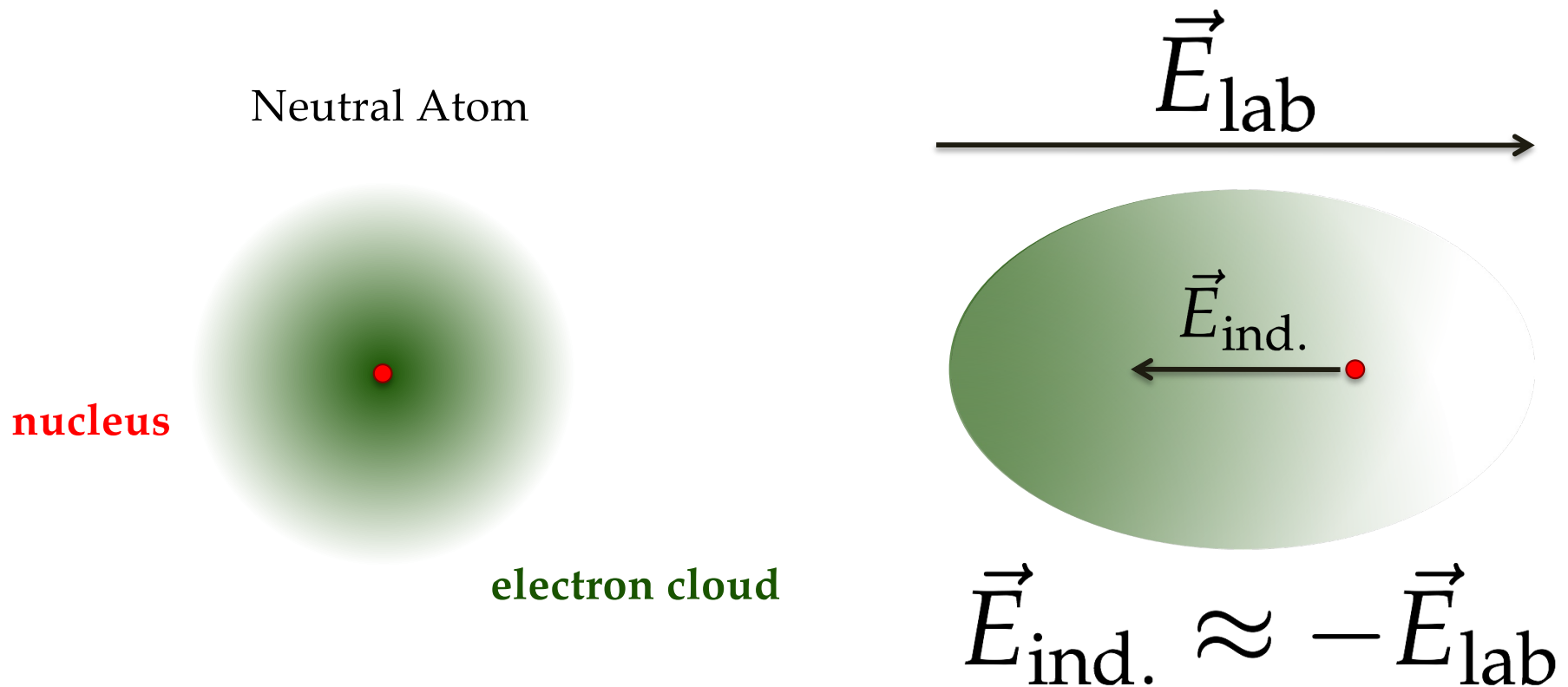
Diamagnetic Atoms: All electrons are paired.



Schiff Shielding in Diamagnetic Atoms

- Shielding in Diamagnetic Atoms

Schiff PR 132:2194 (1963)



Shielding Imperfect with Relativistic Atoms & Finite Nuclei

- Shielding in Diamagnetic Atoms

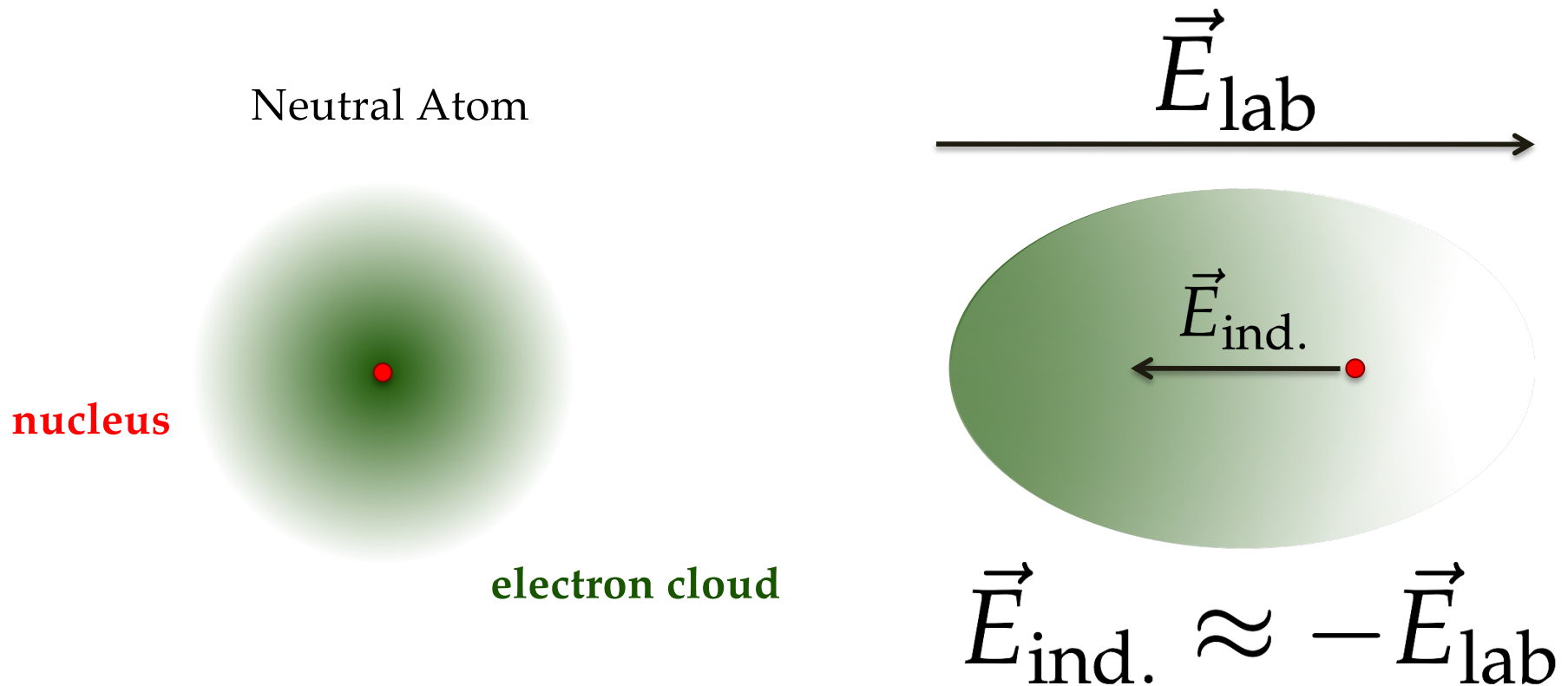
Schiff PR 132:2194 (1963)

- Relativistic atomic structure ($^{225}\text{Ra}/^{199}\text{Hg} \sim 3$)

PRA 66:012111 (2002) & PRL 120:203001 (2018) & PRA 92:022502 (2015)

Schiff Moment

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

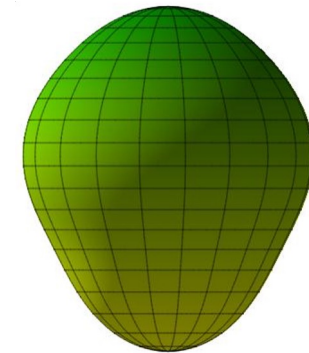


Nuclear Schiff Moment in the Lab Frame

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

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

1. Body-frame nuclear deformation with large intrinsic Schiff moment
2. Difference in lab-frame nuclear energy levels
3. The CP-violating physics that we seek to measure (unknown)



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

Enhanced Nuclear Moments with Parity Doublets

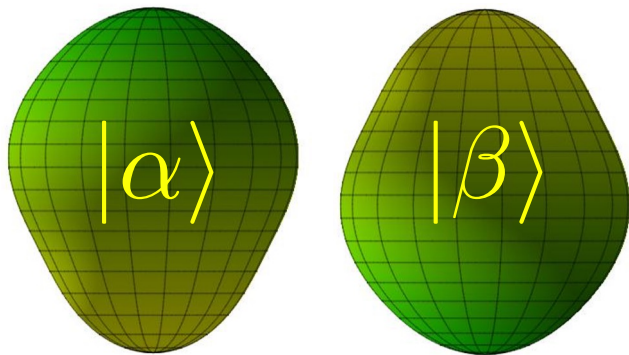
$$S_z = \frac{\langle er^2 z \rangle}{10} - \frac{\langle r^2 \rangle \langle ez \rangle}{6}$$

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

Parity Doublet

- Nearly degenerate parity doublet

Haxton & Henley PRL 51:1937 (1983)



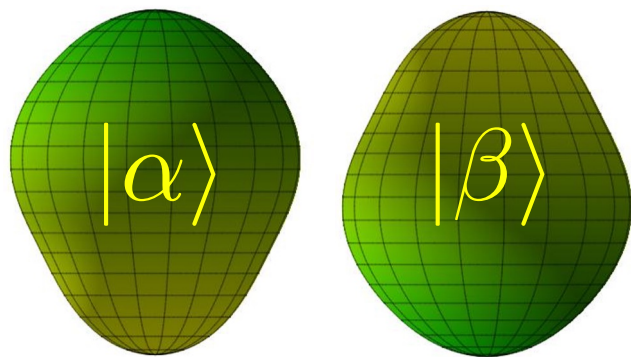
$$\begin{array}{l} \text{---} \\ \uparrow \Delta E \\ \text{---} \end{array} \quad \begin{array}{l} |\Psi_1\rangle = \frac{|\alpha\rangle - |\beta\rangle}{\sqrt{2}} \\ \\ |\Psi_0\rangle = \frac{|\alpha\rangle + |\beta\rangle}{\sqrt{2}} \end{array}$$

Enhanced Schiff Moments in Deformed Nuclei

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

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

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)

$$\begin{array}{l} \text{---} \\ \uparrow \Delta E \\ \text{---} \end{array} \quad |\Psi_1\rangle = \frac{|\alpha\rangle - |\beta\rangle}{\sqrt{2}}$$

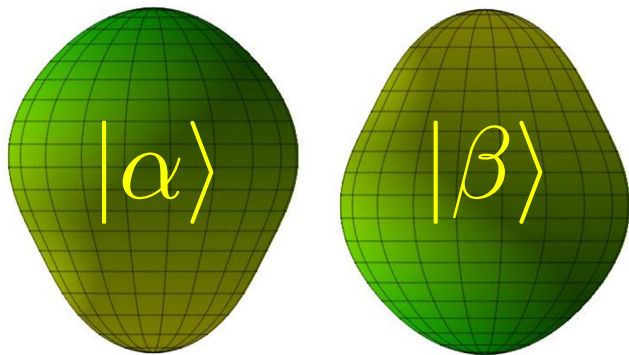
$$\text{---} \quad |\Psi_0\rangle = \frac{|\alpha\rangle + |\beta\rangle}{\sqrt{2}}$$

Enhanced Sensitivity in Radium-223/Radium-225

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

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

Parity Doublet



55 keV

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

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

- Nearly degenerate parity doublet

Haxton & Henley PRL 51:1937 (1983)

- Large intrinsic Schiff moment due to octupole deformation

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

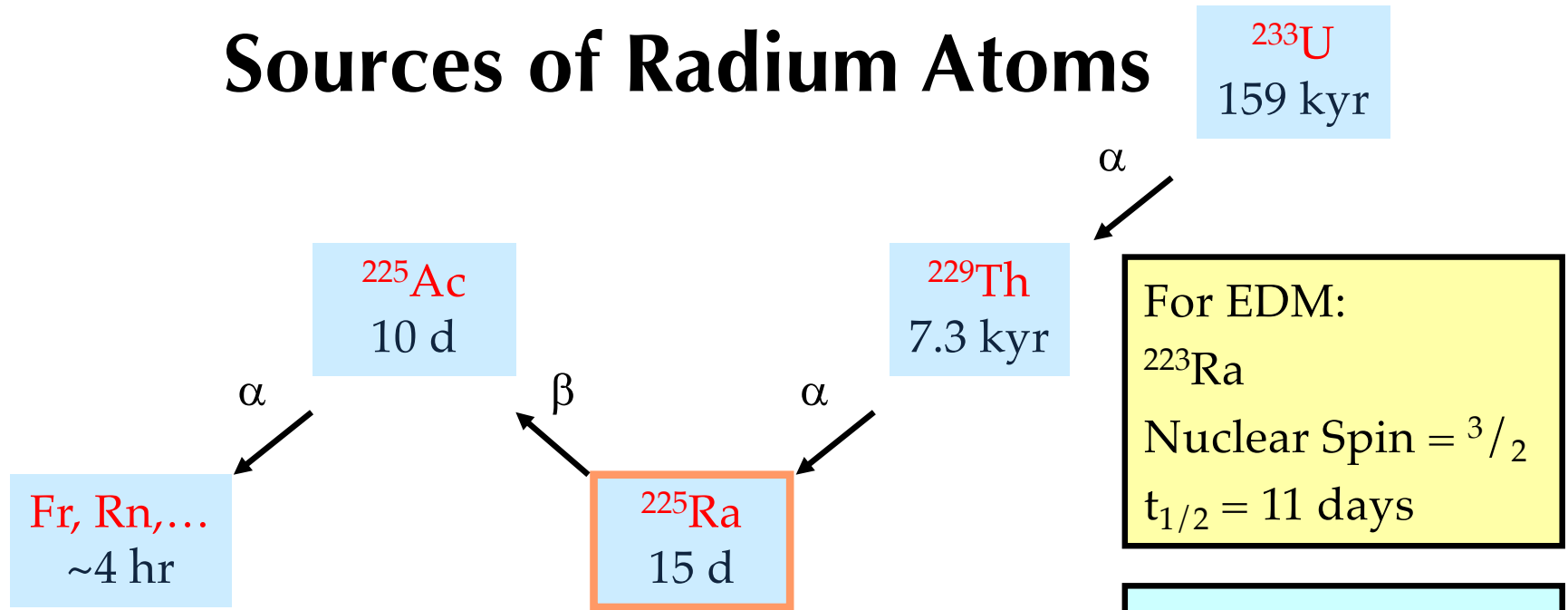
Total Enhancement Factor: EDM (²²⁵Ra) / EDM (¹⁹⁹Hg)

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)

Sources of Radium Atoms



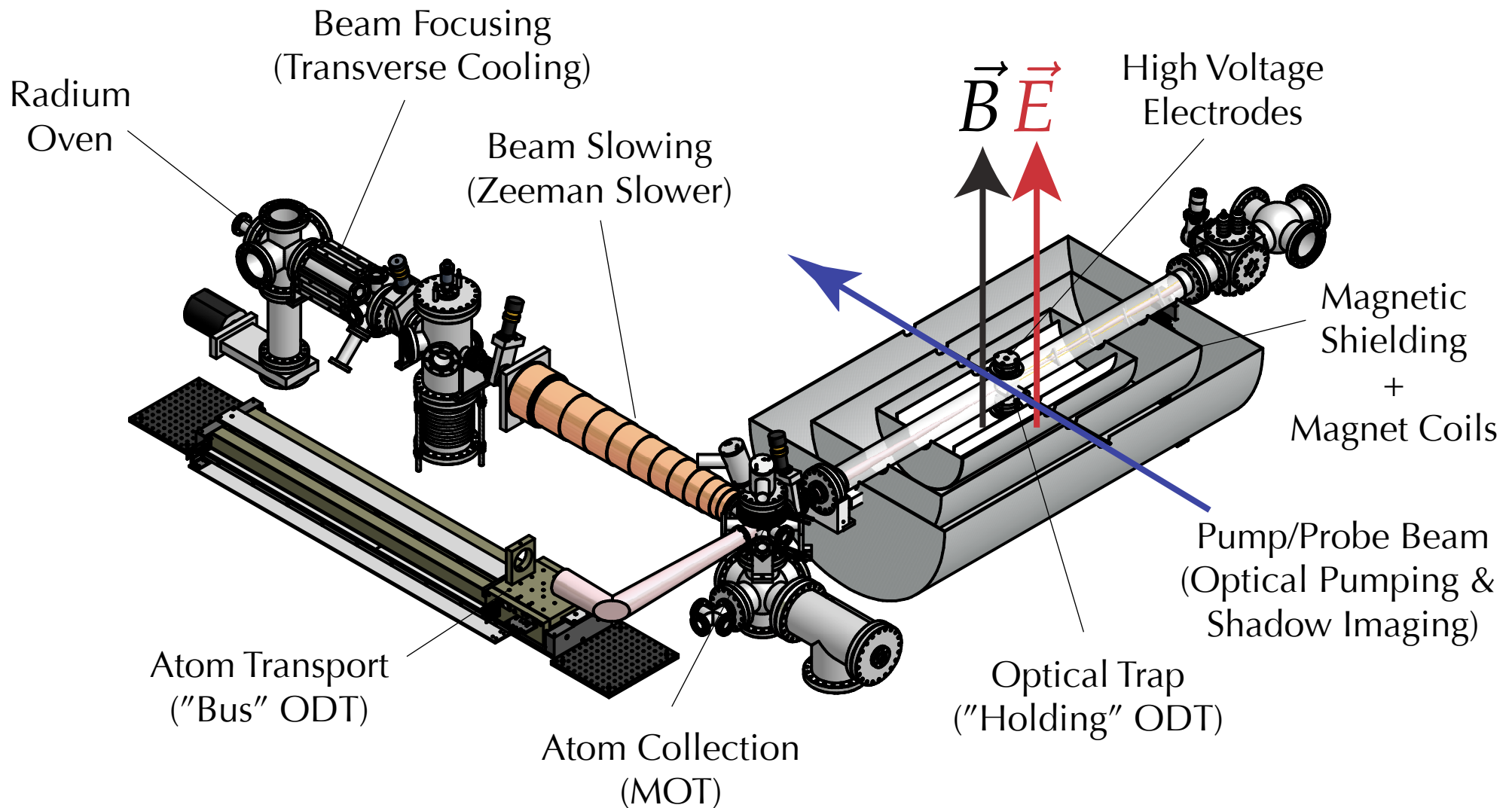
For EDM:
 ^{223}Ra
 Nuclear Spin = $3/2$
 $t_{1/2} = 11$ days

For EDM:
 ^{225}Ra
 Nuclear Spin = $1/2$
 $t_{1/2} = 15$ days

For Testing:
 ^{226}Ra
 Nuclear Spin = 0
 $t_{1/2} = 1600$ yrs

- 2 mCi (50 ng) ^{225}Ra sources from:
 National Isotope Development Center (Oak Ridge, TN)
- Test source: 1 μCi (1 mg) ^{226}Ra
- Integrated Atomic Beam Flux $\sim 10^8/\text{s}$
- **^{225}Ra is reserved for cancer research (targeted alpha therapy using ^{225}Ac), but we have a plan to use a ^{229}Th source.**
Eventually, the Facility for Rare Isotope Beams will produce ^{225}Ra , ^{229}Pa , etc.

The Laser Trap Ra EDM Experiment @ Argonne



Ra EDM: Completely Statistics Limited

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

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

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

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

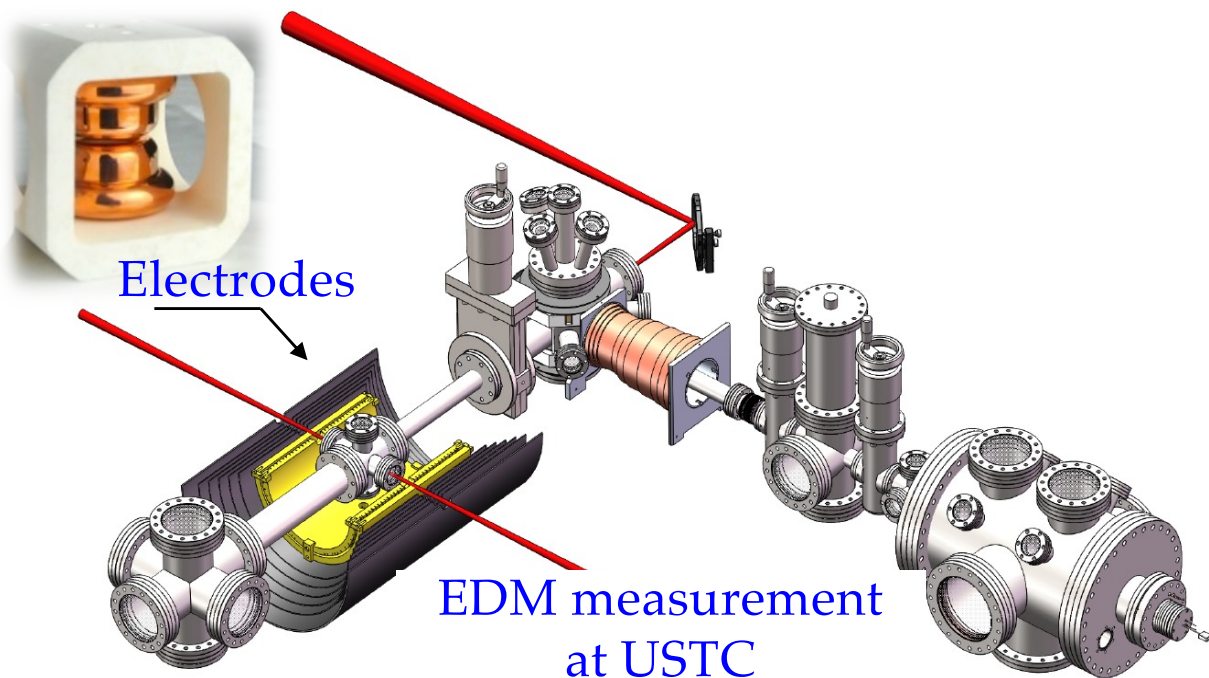
More efficient detection of atoms: optical cycling

More efficient laser cooling and trapping: 1 ppm to 100 ppm

Higher electric field: 70 kV / cm to >200 kV / cm

Goal is $<10^{-26} e \text{ cm}$ over 4 years and then $10^{-28} e \text{ cm}$ long term

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



- Pathfinder experiment for $^{223,225}\text{Ra}$
- Coherent spin precession time > 300 s
- **EDM(^{171}Yb) $< 1.5 \times 10^{-26}$ e-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 $\times 100$

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

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

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

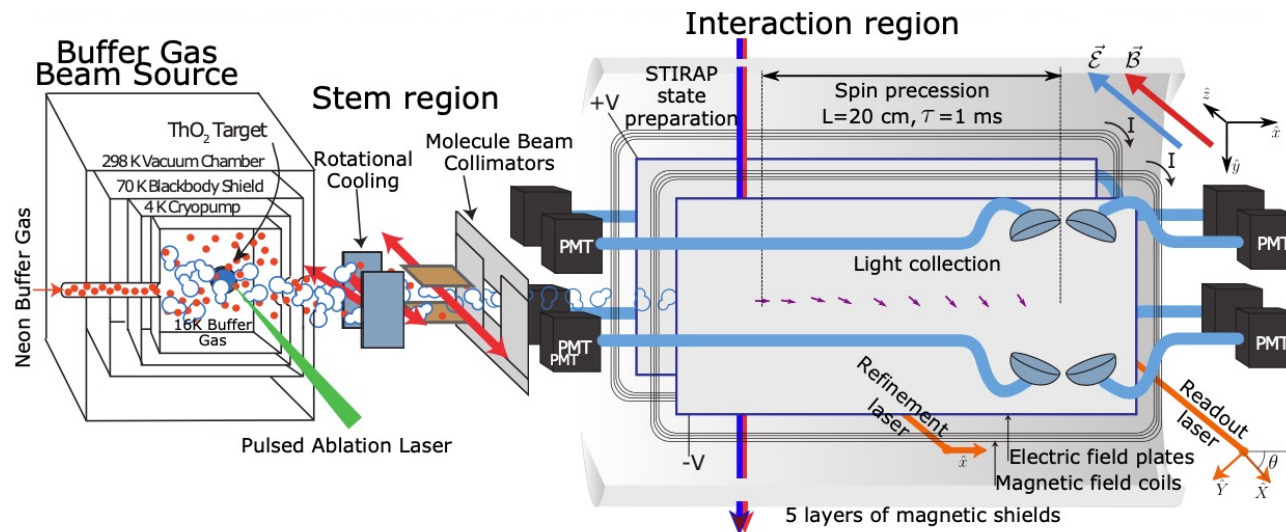
Yb-171: PRL 129:083001 (2022): $< 1.5 \times 10^{-26} e \text{ cm}$ (95%)
(laser trap experiment, very similar to Ra experiment)

- 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.

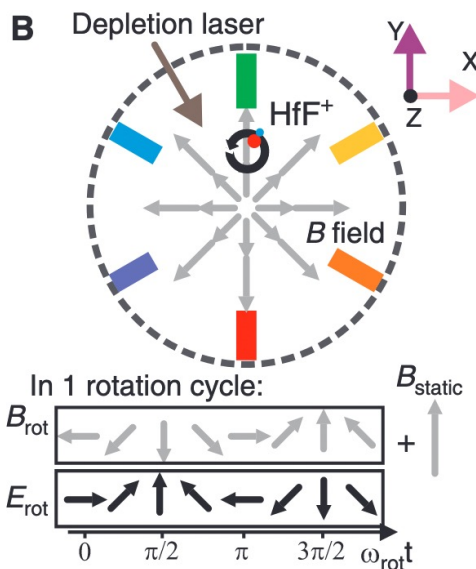
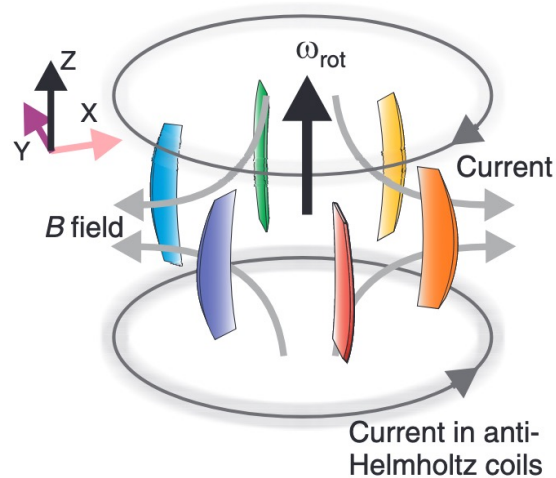
Molecular Electron EDM Experiments: Large Internal E-field and Control of Systematics

ACME – ThO*
Neutral Beam
(Chicago/
Harvard/
Northwestern)

C. Panda (Harvard 2018)
Nature 562 355 (2018)



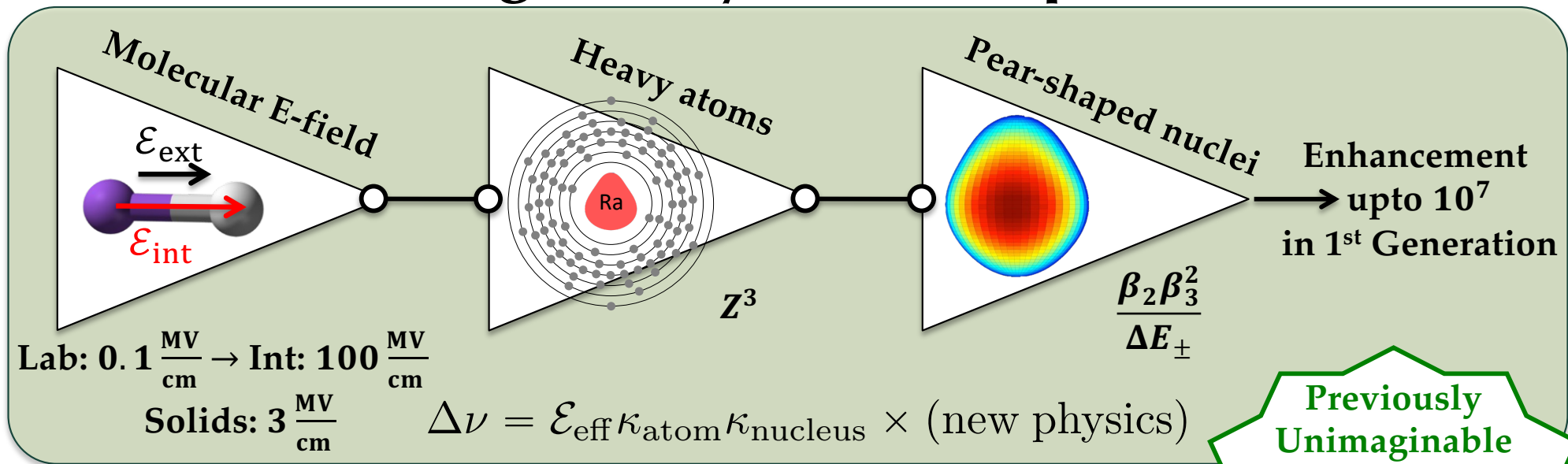
A $\vec{E}_{rot} = E_{rot} (\cos(\omega_{rot}t) \hat{x} + \sin(\omega_{rot}t) \hat{y})$



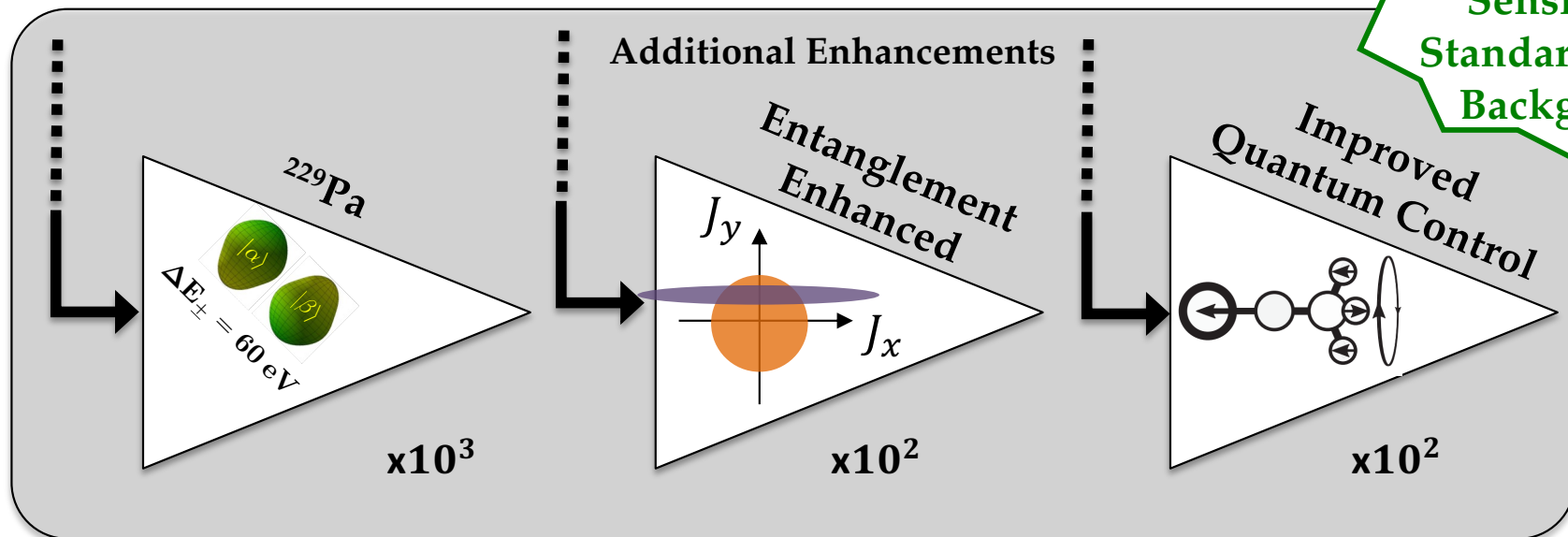
HfF⁺ / ThF⁺
Ion Trap
(JILA)

Science 342 (6163) 1220 (2013)
PRL 119 153001 (2017)
arxiv:2212.11841 (2022)

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



Previously Unimaginable Sensitivity: Standard Model Background

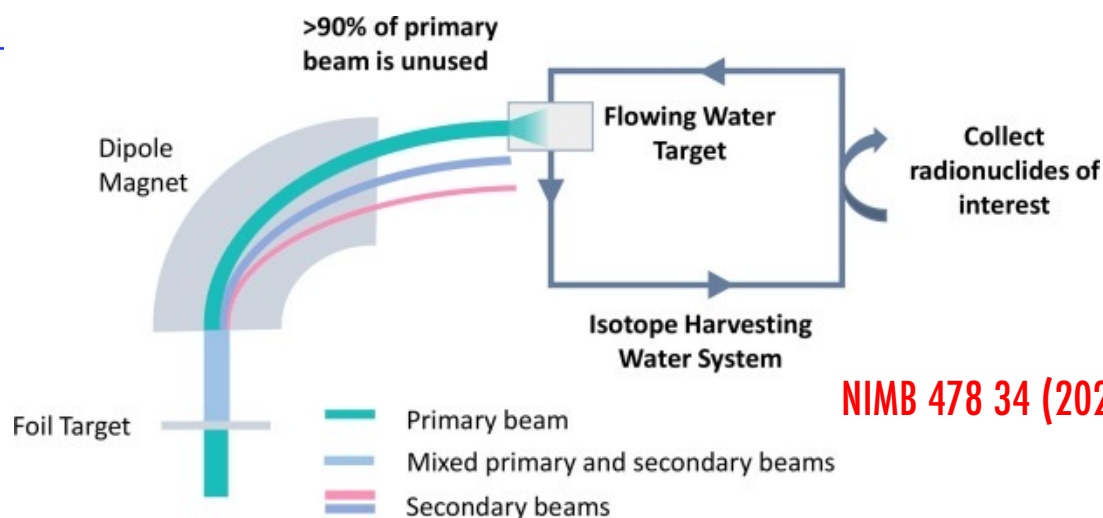


Opportunity for Nuclear Schiff Moments: Short-Lived Pear-Shaped Nuclei Inside Molecules

Enhancements: nuclear Schiff moment enhancement of $\times 1000$ (^{225}Ra)
to maybe(!?!) $\times 1000000$ (^{229}Pa)
and ~ 100 MV/cm effective internal E-field (lab < 1 MV/cm)

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 , ...)



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?

Facility for Rare Isotope Beams @ MSU



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

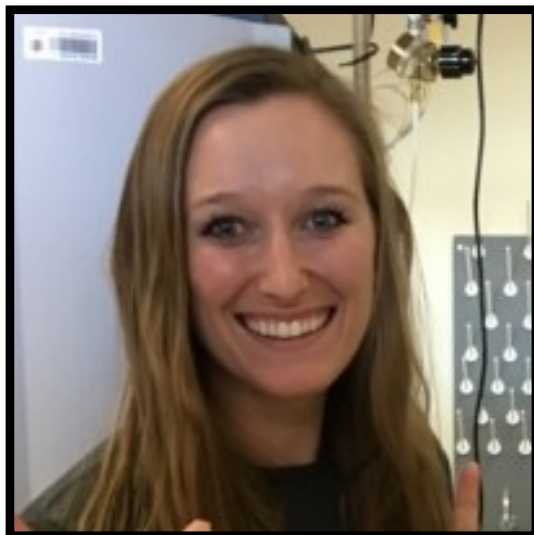
University of Michigan
Ann Arbor, MI
Good at American Football
no FRIB

Google Maps & Wikipedia Commons

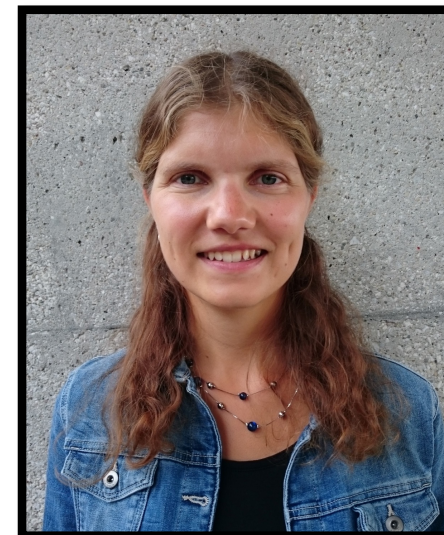
“Isotope Harvesting” at The Facility for Rare Isotope Beams (MSU/East Lansing)



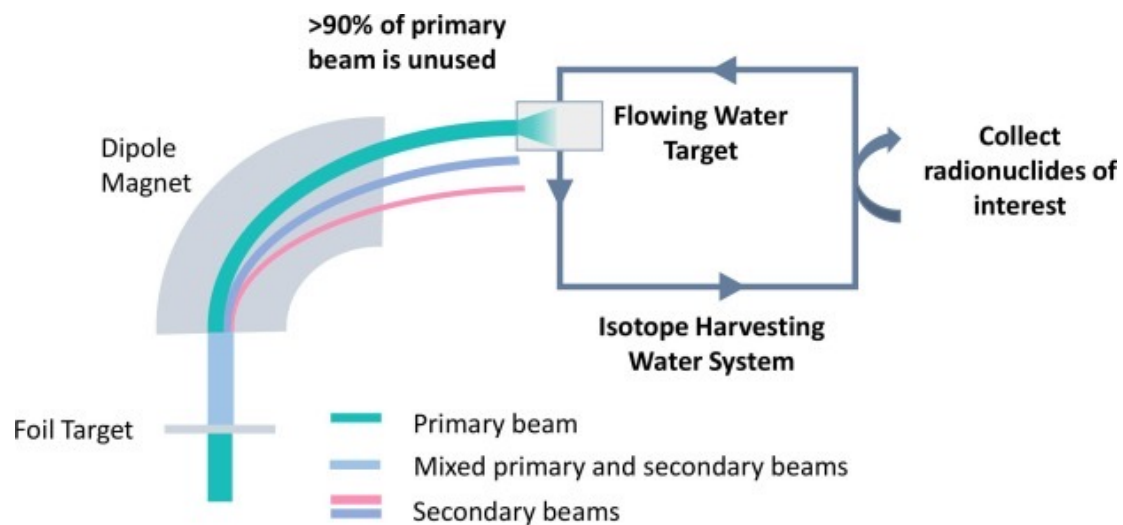
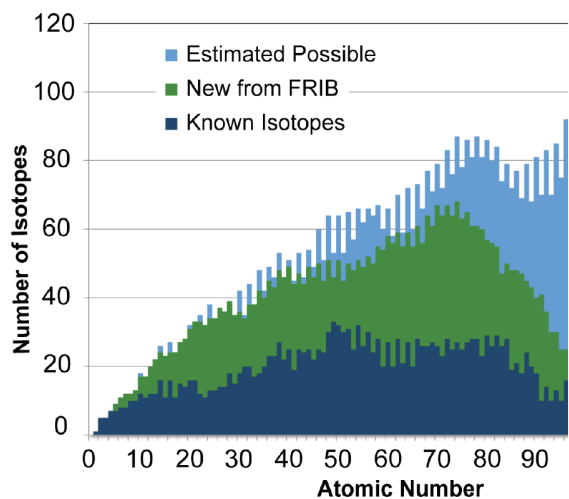
Prof. Greg Severin



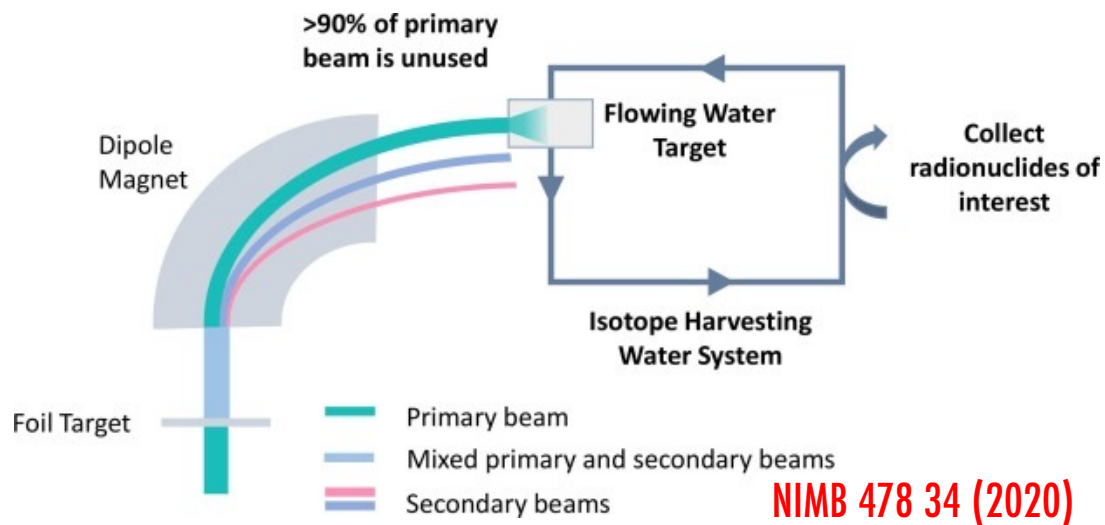
Prof. Alyssa Gaiser



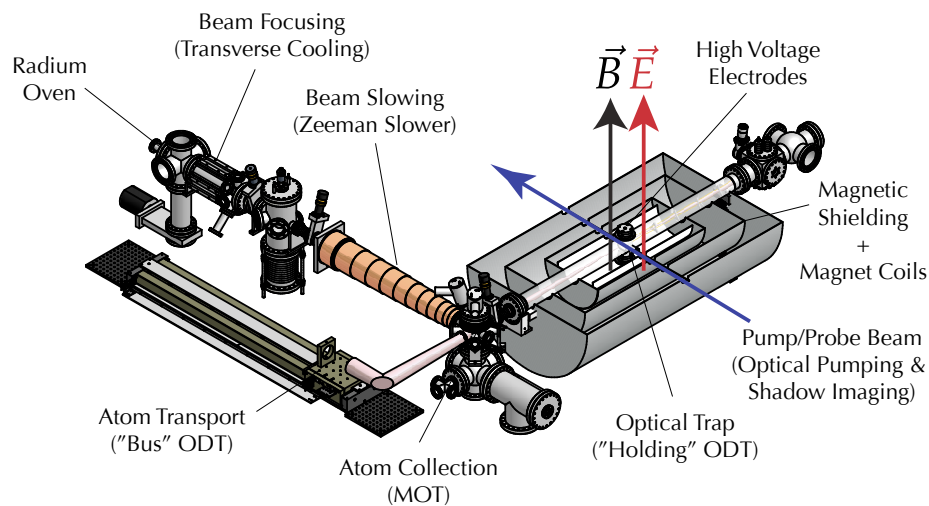
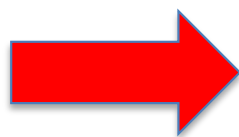
Prof. Katharina Domnanich



\$upport Needed For “Beaker to Experiment”

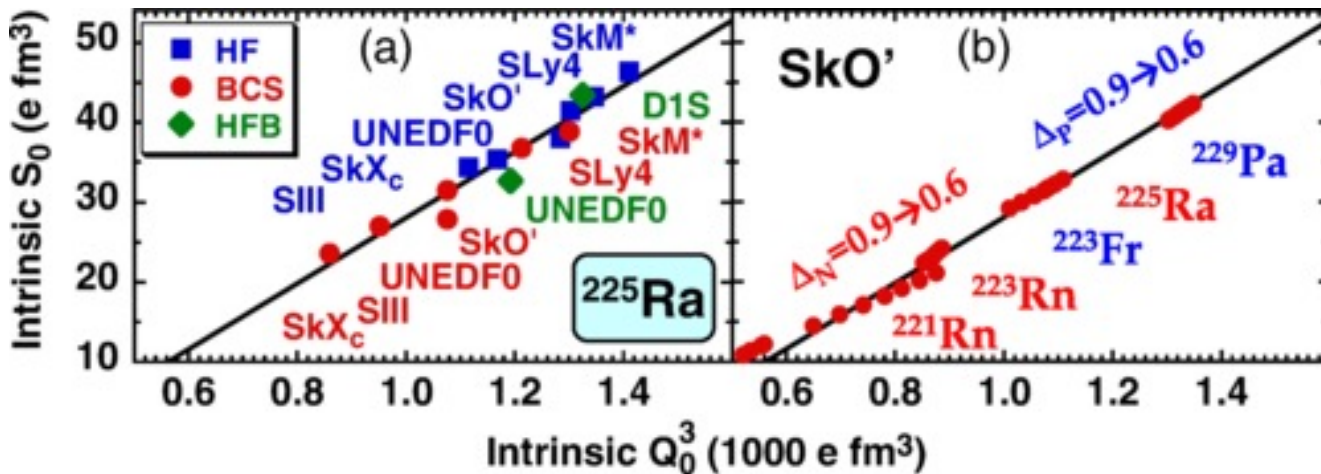


- FRIB Operations is supported by DOE-NP
- Isotope Harvesting @ FRIB is supported by DOE-Isotopes



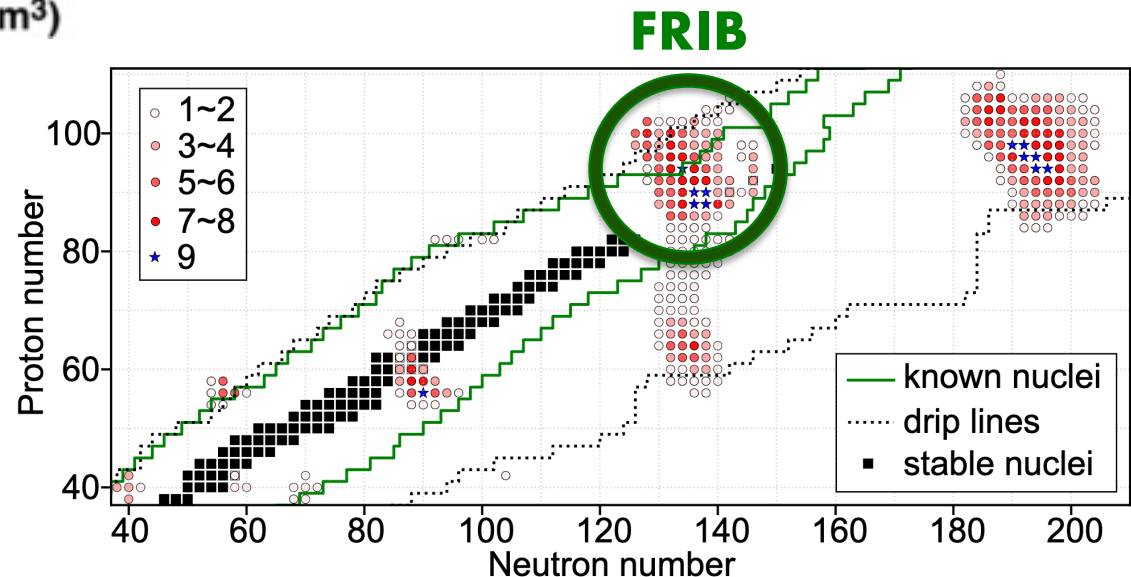
Calibrating the Intrinsic Schiff Moment

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



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

Nuclear structure measurements combined with nuclear theory can calibrate the new physics sensitivity of “enhancer” isotopes with uncertainty quantification!



Protactinium-229 (^{229}Pa) *may* be unusually sensitive!

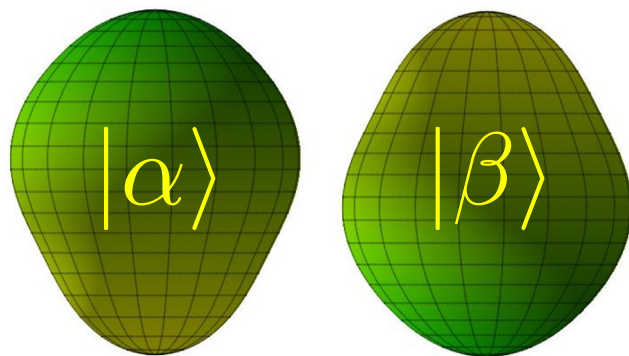
$$S_z = \frac{\langle er^2z \rangle}{10} - \frac{\langle r^2 \rangle \langle ez \rangle}{6}$$

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



$$\begin{aligned} \Delta E & \begin{array}{l} \uparrow \\ \downarrow \end{array} \\ |\Psi_1\rangle &= \frac{|\alpha\rangle - |\beta\rangle}{\sqrt{2}} \\ |\Psi_0\rangle &= \frac{|\alpha\rangle + |\beta\rangle}{\sqrt{2}} \end{aligned}$$

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

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

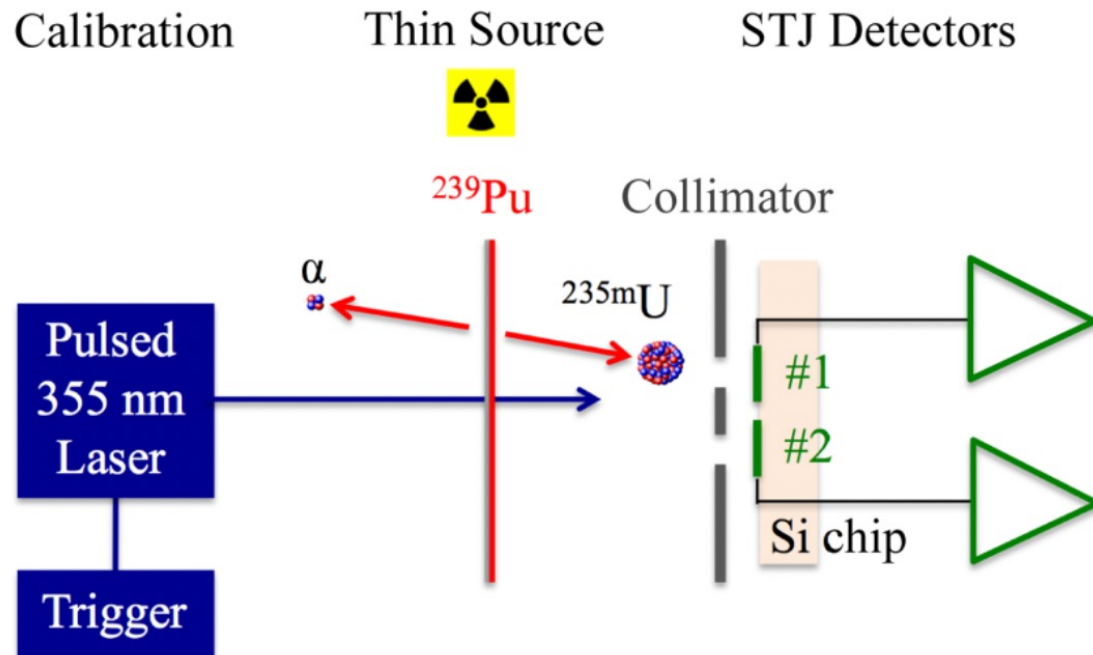
Dobaczewski et al PRL 121, 232501 (2018)

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!

Planned Pa-229 Nuclear Spectroscopy @ FRIB!

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



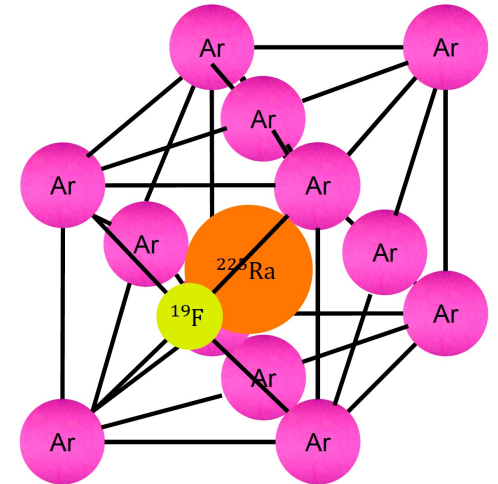
PRC 97 054310 (2018)

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.

Pear-Shaped Nuclei Implanted In Cryogenic Solids:

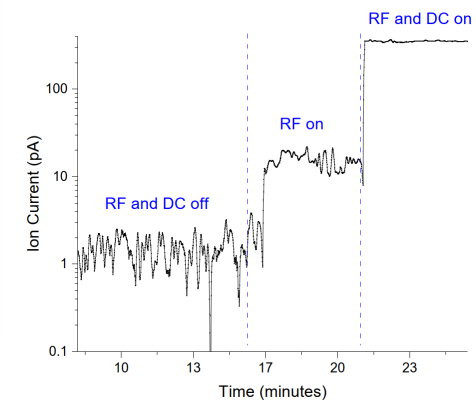
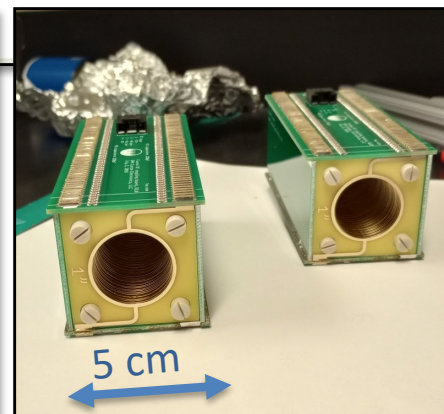
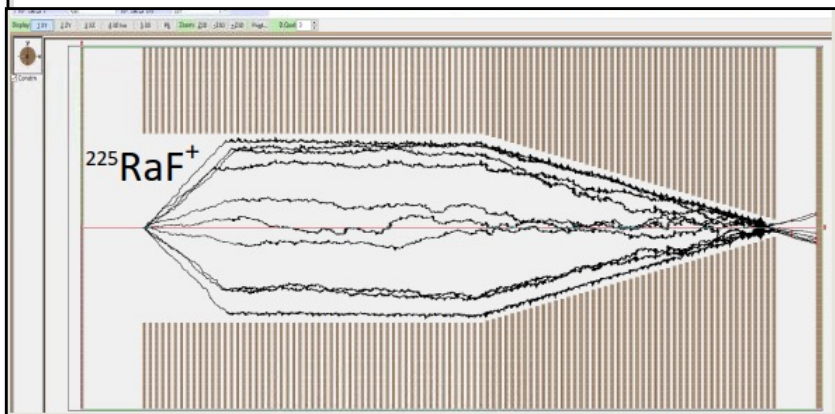
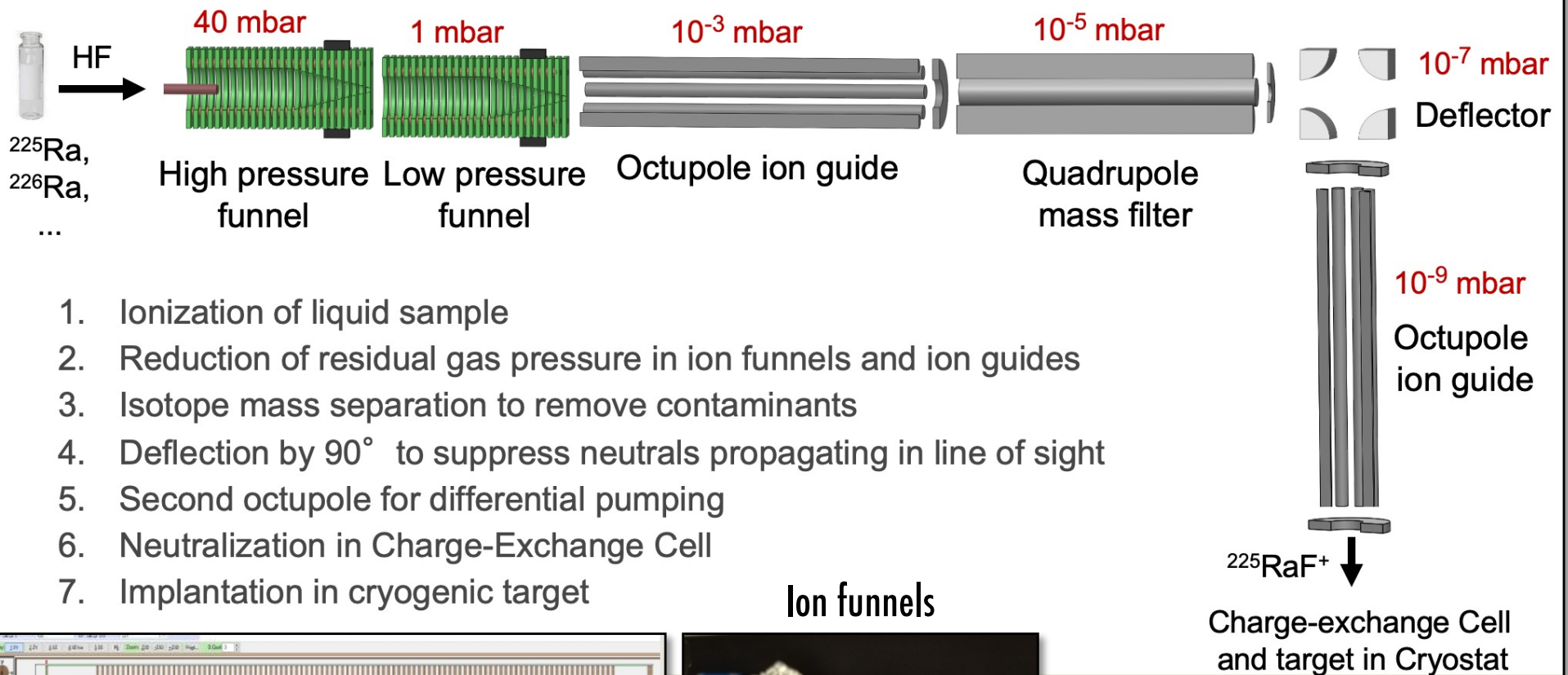
^{225}RaF ($t_{1/2} = 15$ days) & ^{229}Pa ($t_{1/2} = 1.5$ days)

- **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 allowing for optically-addressable nuclear spins [Hyp. Int. 240:29 \(2019\)](#), [arXiv:2305.05781 \(2023\)](#), [arXiv:2304.10331 \(2023\)](#)
- Implanted ions can sit at two distinct sites with opposite pointing internal E-fields which allows for control of systematics [PR 131 1912 \(1963\)](#)
- **Efforts are underway to form & implant molecules & ions into solids**



JTS DOE ECA 2018

RaF & RaOH in Noble Gas Solids (MSU/York/Toronto)



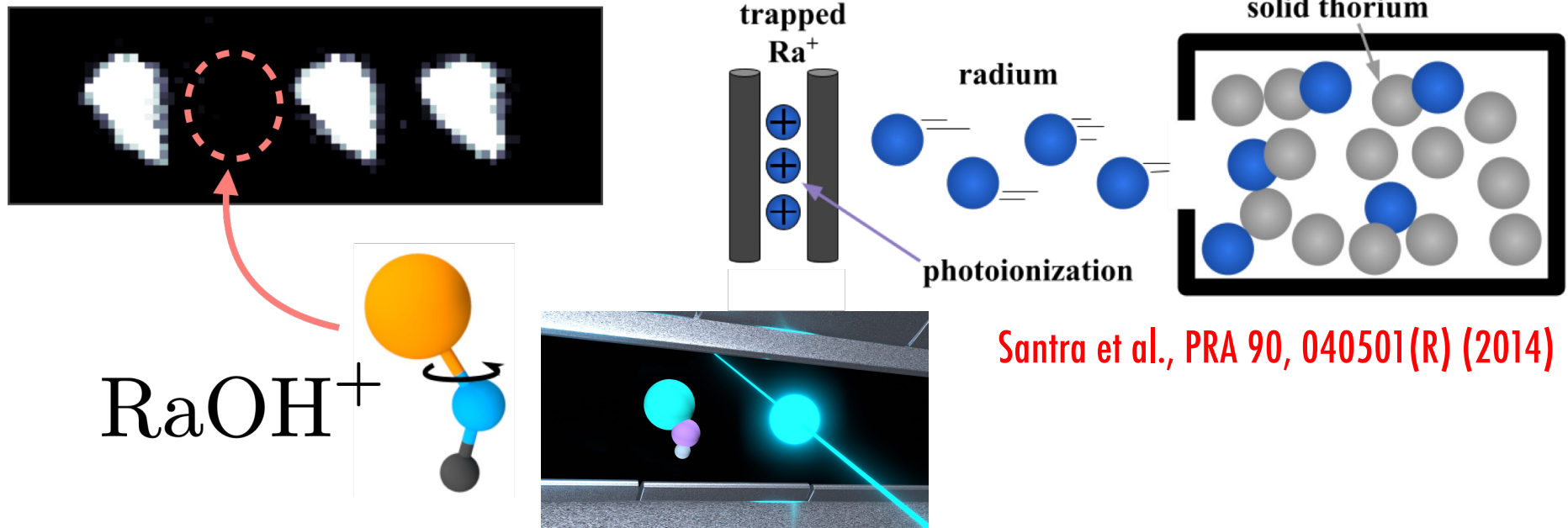
N. Nusgart, ICAP 2022

2023-06-30

GAA Custom Electronics

2023 CAP Congress, Fredericton, NB

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

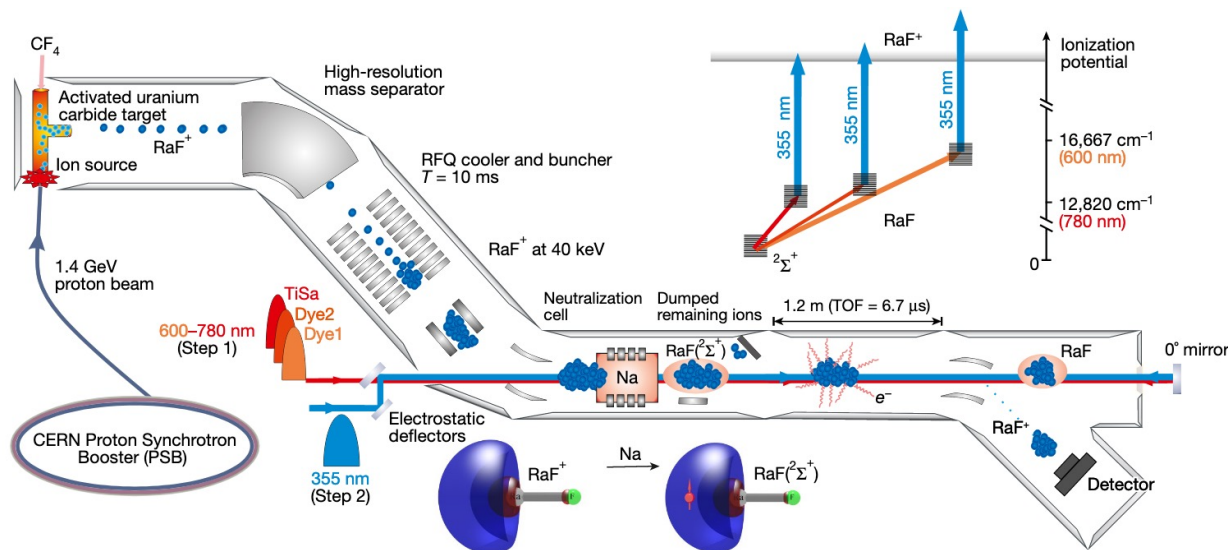


Santra et al., PRA 90, 040501(R) (2014)

- 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

Direct Laser Cooling of Neutral Molecules Into a Laser Trap: ^{225}RaF & $^{225}\text{RaOH}$ ($t_{1/2} = 15$ days)



- Molecular spectroscopy of RaF is underway!
- Laser cooling of RaF appears feasible and scheme is under development

Nature 581:396 (2020)

PRL 127:033001 (2021)

slide from R. Garcia Ruiz

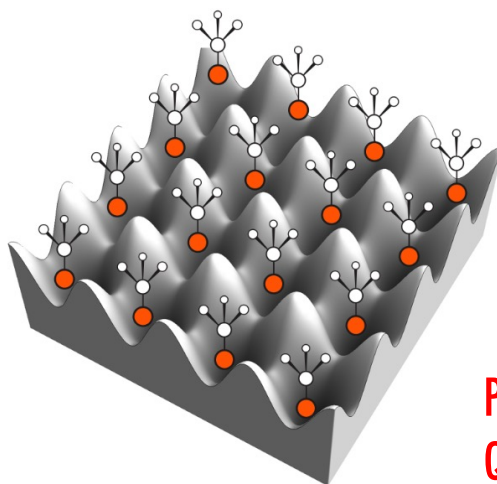
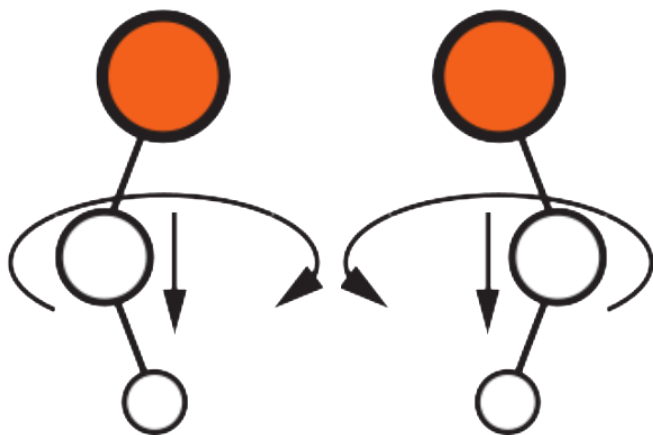
Benefits of Polyatomic Molecules

- Laser coolable & trappable
- Highly polarizable
- Comagnetometer states for control of systematics
- High CPV sensitivity

PRL 119, 133002 (2017)

Quantum Science & Tech. 5, 044011 (2020)

slide from N. Hutzler



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

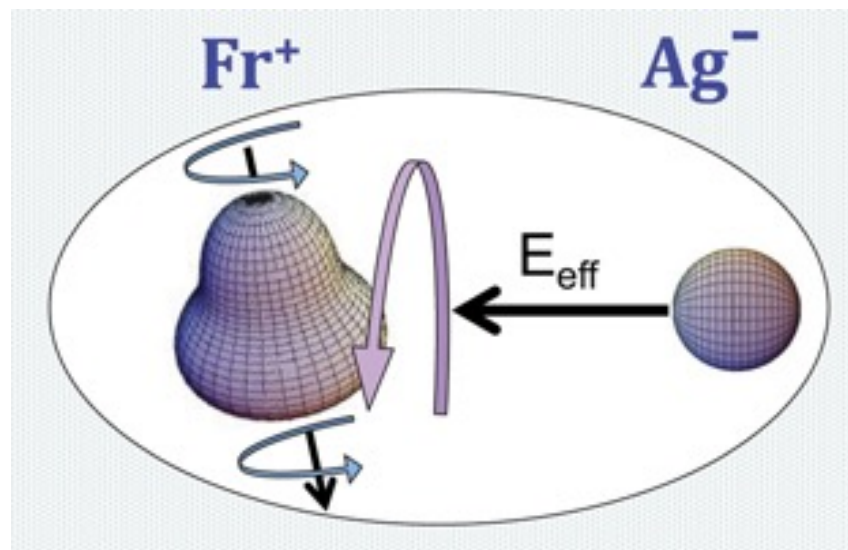
- Included in Gen-I Estimate:

- 300x NSM enhancement
- near-ideal molecular structure
- $t_{\text{coh}} \sim 10$ S [Cornish, Zwierlein, etc.]
- ~100% detection efficiency
- $n = 10^4$ molecules

⇒ ~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



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

Theory calculations favorable:

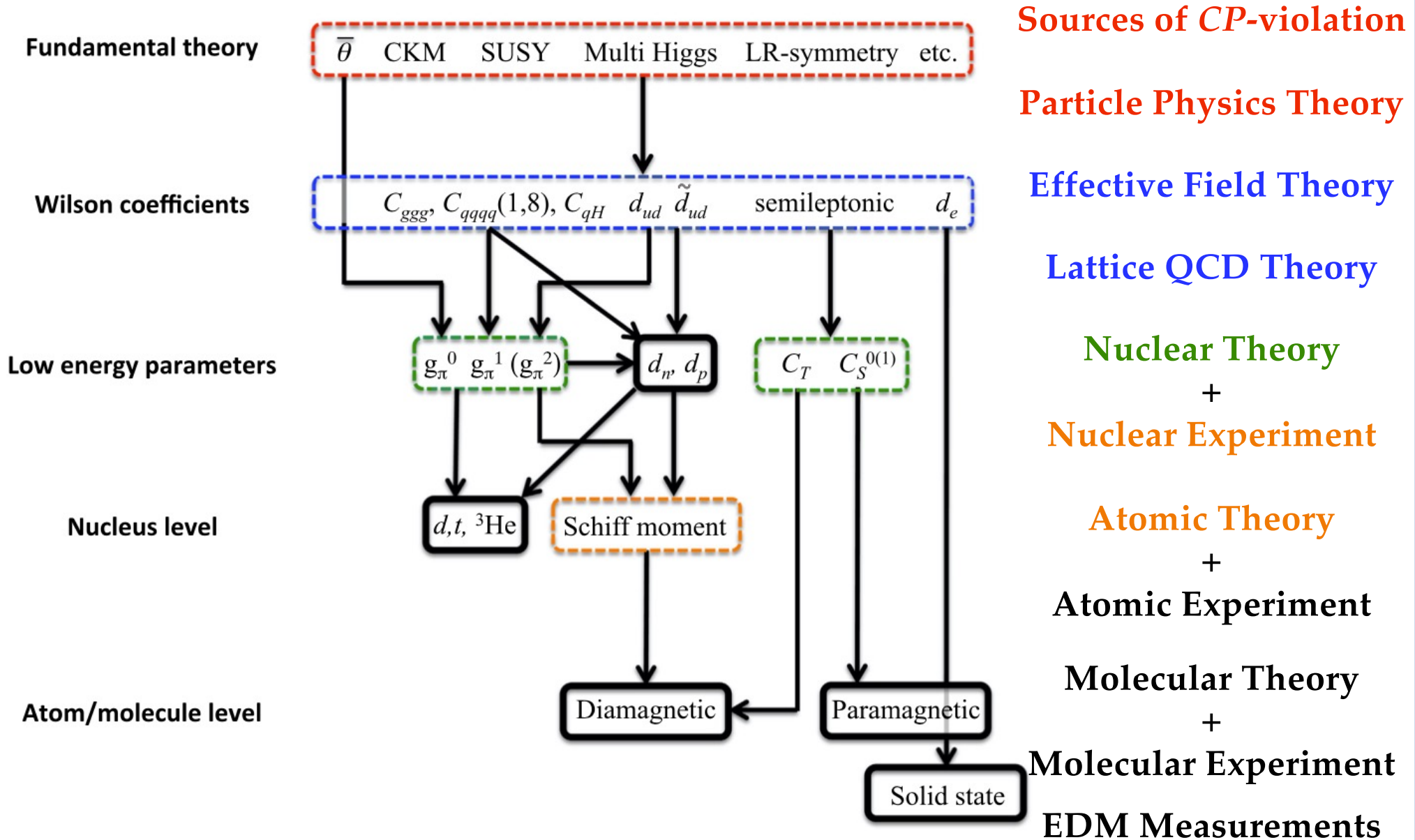
New J. Phys. 23 113039 (2021)

New J. Phys. 24 025005 (2022)

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

Connecting New Physics to EDMs

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

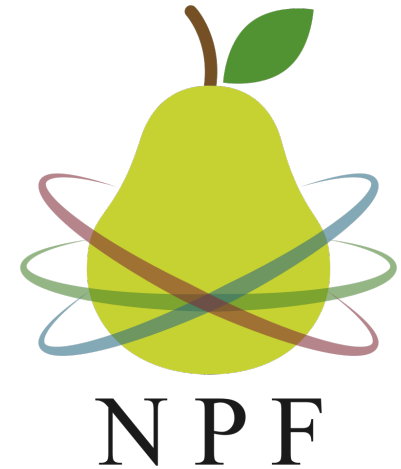
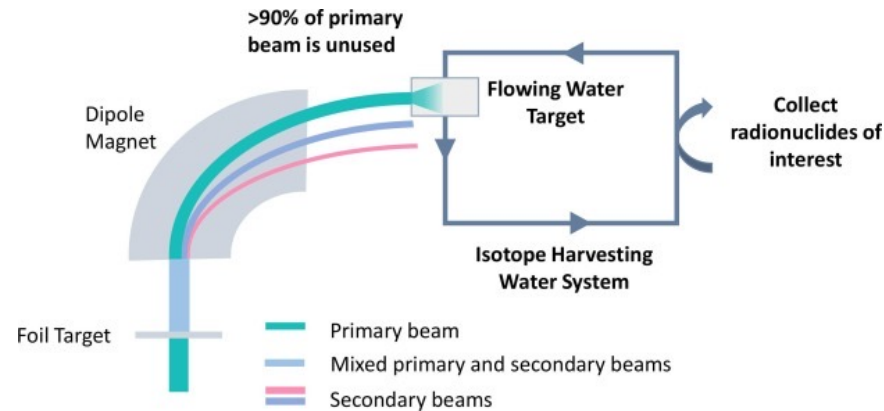


The Nuclear Pear Factory: A Proposed Center

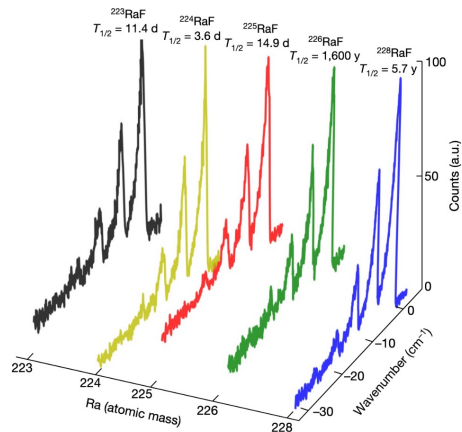


Nature 497:199 (2013)

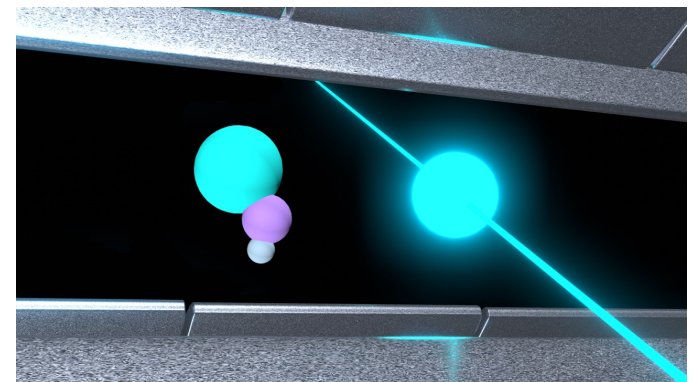
A joint Experiment/Theory & AMO/Nuclear effort to calibrate the new physics sensitivity of pear-shaped nuclei and to carry out the requisite precursory work leading to ultrasensitive EDM searches.



NIMB 478 34 (2020)



Nature 581:396 (2020)



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

Thanks For Your Attention!

1. Detecting a non-zero EDM would be an unambiguous signature of physics Beyond the Standard Model of Particle Physics.
2. Pear-shaped nuclei such as Radium-225 and Protactinium-229 have significantly enhanced sensitivity to *CP*-violation originating within the nuclear medium.
3. **Short-lived radioactive molecules potentially have $\times 10^5$ to $\times 10^{10}$ more new physics sensitivity than Hg-199 in the hadronic sector on a per atom basis.**
4. **Isotope harvesting and radiochemistry at FRIB enables access to these enhancer isotopes in practical quantities for ultrasensitive EDM searches.**
5. **We propose a center, The Nuclear Pear Factory, to realize the unprecedented discovery potential made possible by short-lived radioactive molecules.**

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