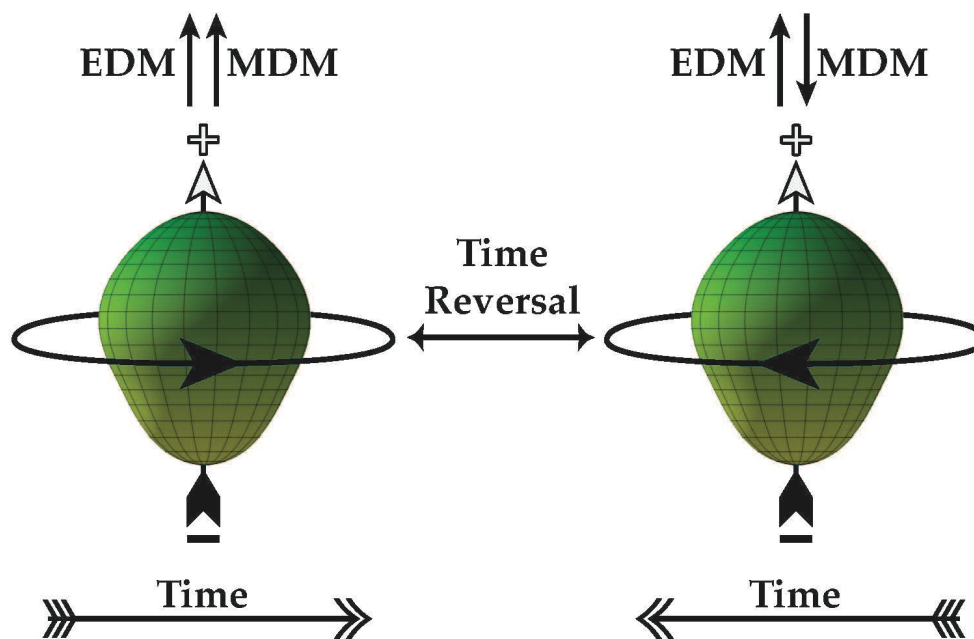


Prospects of Using Pear-Shaped Nuclei in Cryogenic Solids for Tests of Time-Reversal Symmetry



Jaideep Taggart Singh (he/him/his)

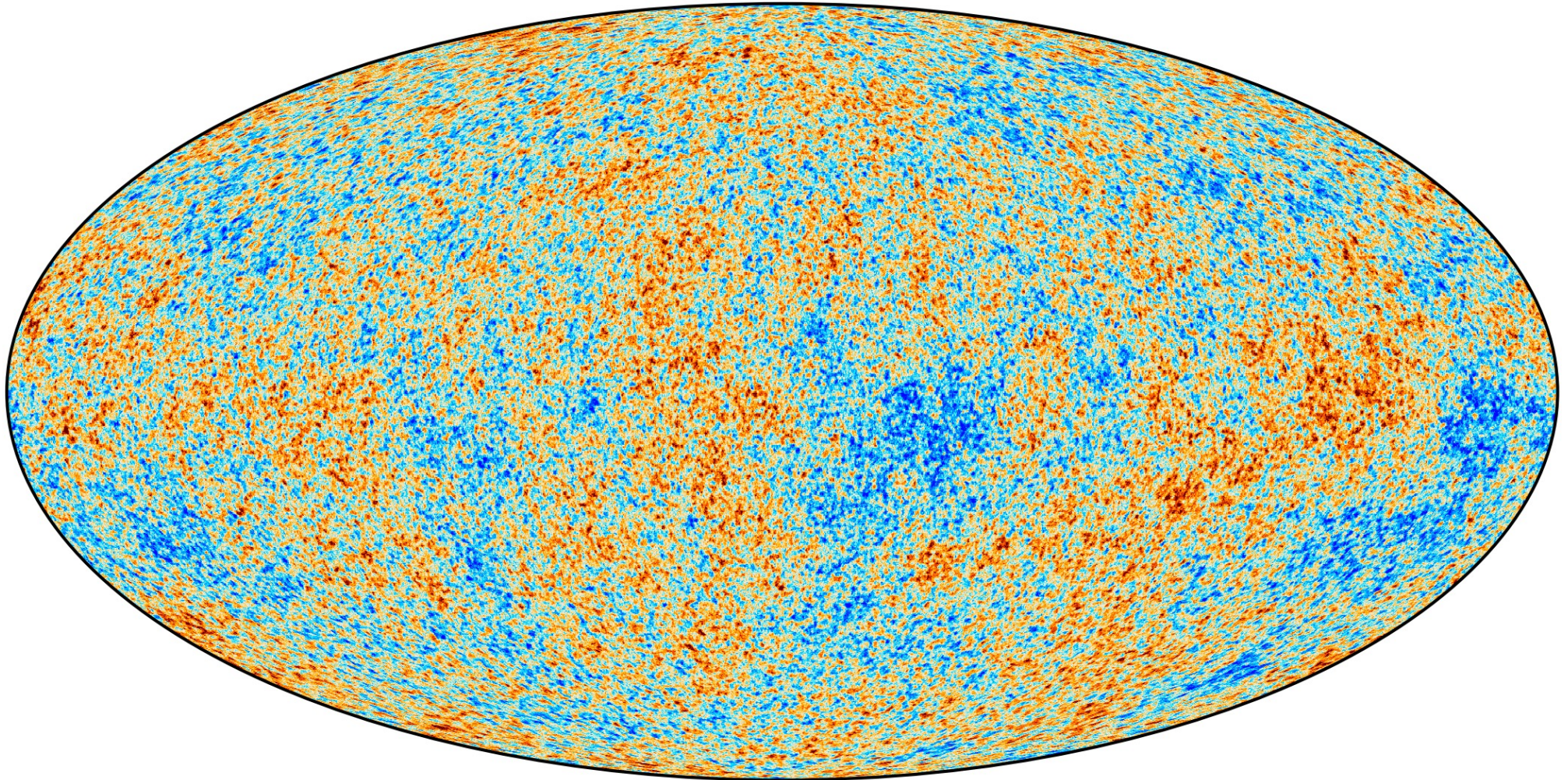
Michigan State U./FRIB/NSCL

12:10-12:45 June 28, 2021 (Virtual Meeting - MIT)

**New Opportunities for Fundamental Physics
Research with Radioactive Molecules**



Cosmic Microwave Background Anisotropy



Planck 2015

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

~10 ppm
fluctuations

There is no visible antimatter in the Universe.

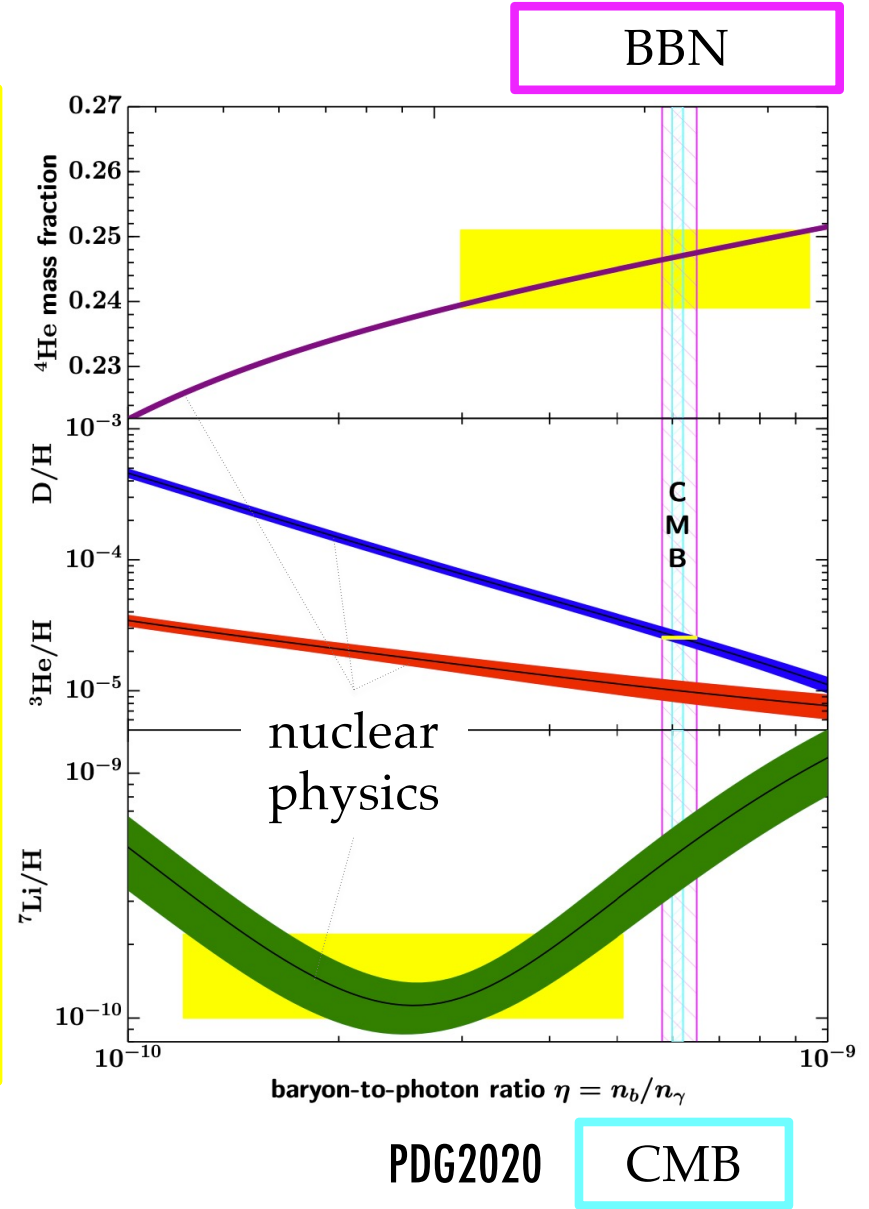
$$\frac{(\text{matter}) - (\text{antimatter})}{\text{relic photons}}$$

$$\eta = \frac{n_B - n_{\bar{B}}}{n_\gamma}$$

$$= 0.000000000061 \text{ (1\%)}$$

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

observational astronomy



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.*

Standard Model CP -Violation: Not Enough

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

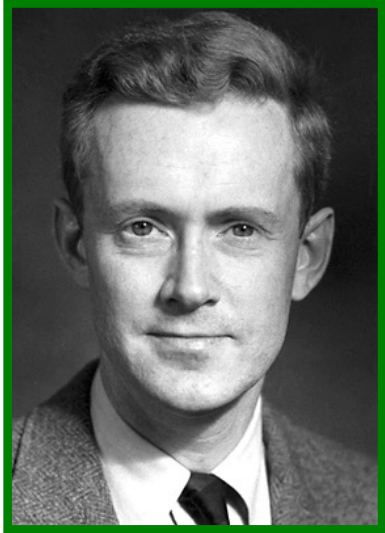
$$\eta_{\text{exp}} \approx 10^{-9} \quad \text{PDG2020}$$

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

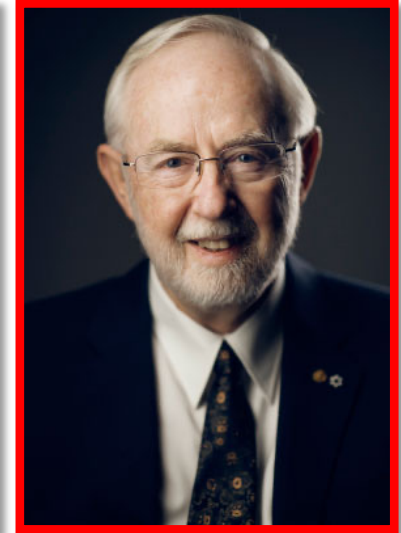
$$V = \begin{pmatrix} c_{12}c_{13} & s_{12}c_{13} & s_{13}e^{-i\delta} \\ -s_{12}c_{23} - c_{12}s_{23}s_{13}e^{i\delta} & c_{12}c_{23} - s_{12}s_{23}s_{13}e^{i\delta} & s_{23}c_{13} \\ s_{12}s_{23} - c_{12}c_{23}s_{13}e^{i\delta} & -c_{12}s_{23} - s_{12}c_{23}s_{13}e^{i\delta} & c_{23}c_{13} \end{pmatrix}$$

$\delta = CP$ -violating “phase”

Where do we look for more CP -violation?



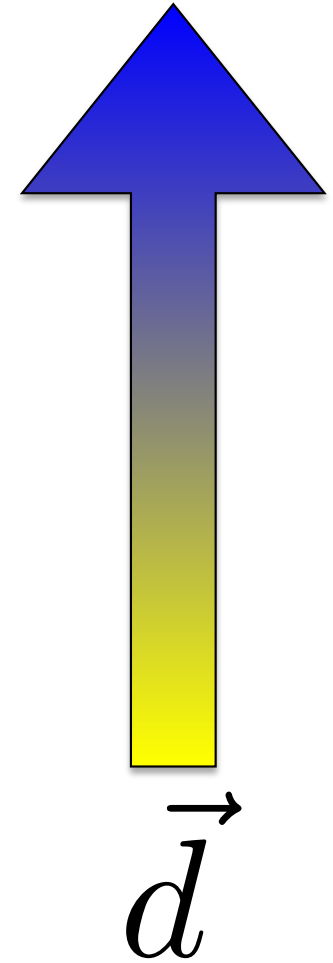
The Nobel Foundation



The Nobel Foundation

- Decays of B-mesons (like Kaons) [BABAR ; KEK]
- Rare decays of baryons [LHCb]
- Angular correlations in the 3γ -decay of ortho-positronium [MSU ; MIT ; TUNL ; Krakow]
- D-coefficient in nuclear beta-decay [The MORA Project]
- **Neutrinos have mass! (PMNS matrix) [T2K! + $0\nu 2\beta$]**
- ***electric dipole moments: If CPT is good, then T-violation can be used to search for new sources of CP-violation!***

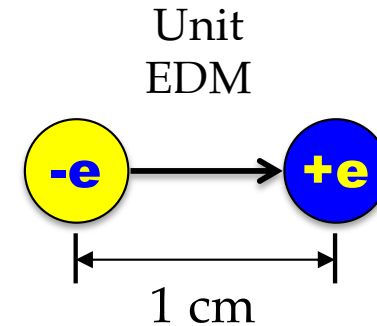
EDM: Measures the Separation of Charges



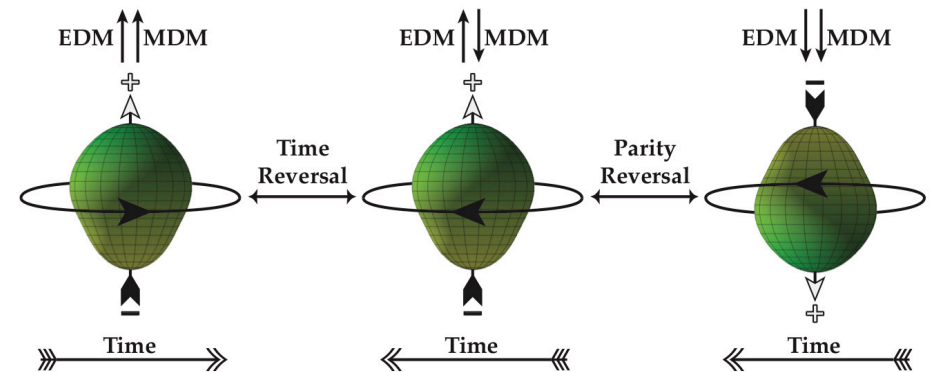
"Thunder Cloud as Generator #2" (1971) by Paterson Ewen [Art Gallery of Ontario]

EDMs to E-fields as MDMs to B-fields

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



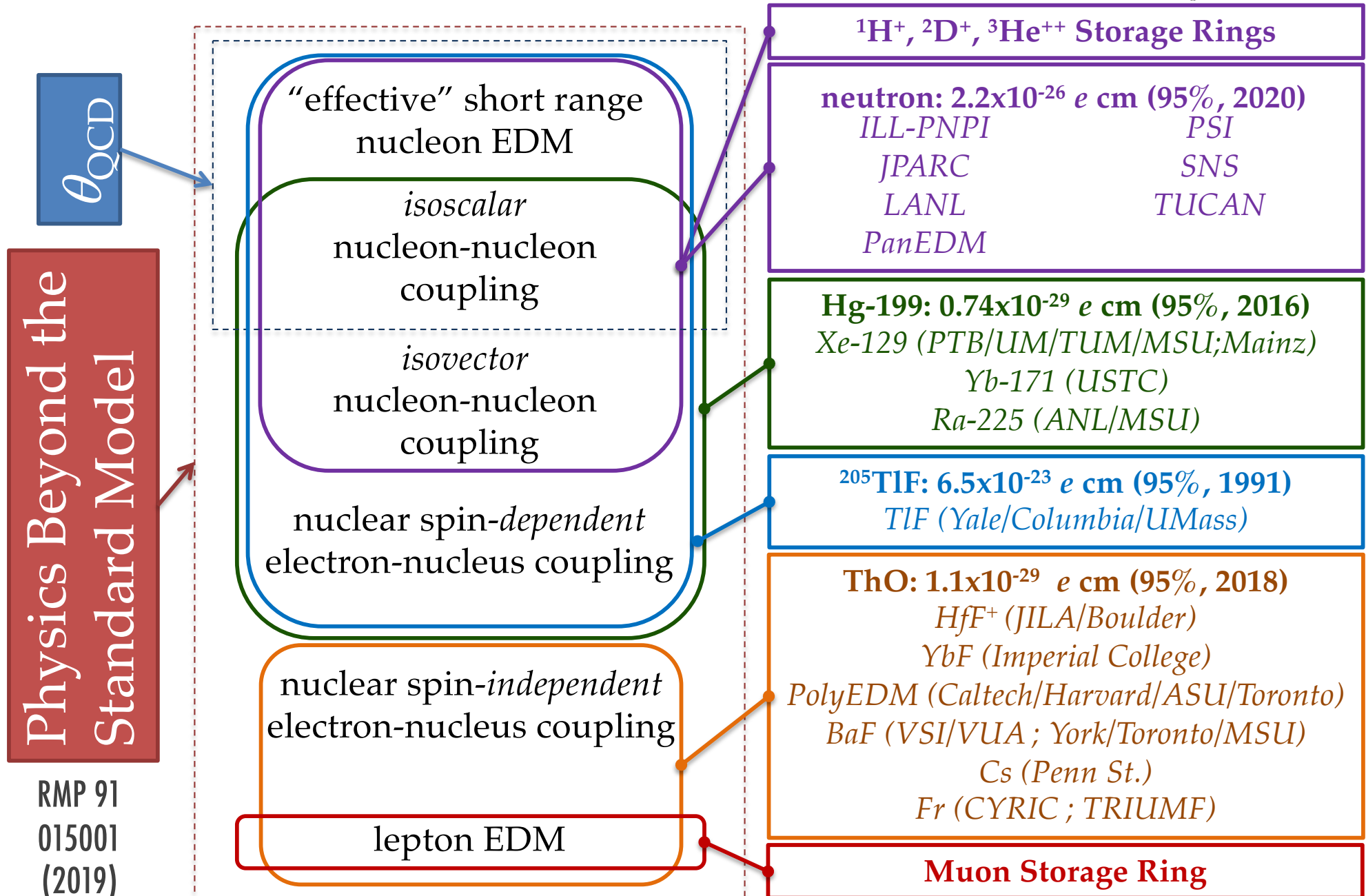
	<i>P</i> -parity	<i>T</i> -time reversal
\vec{S}	+	-
\vec{B}	+	-
\vec{E}	-	+
$\vec{S} \cdot \vec{B}$	+	+
$\vec{S} \cdot \vec{E}$	-	-



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

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

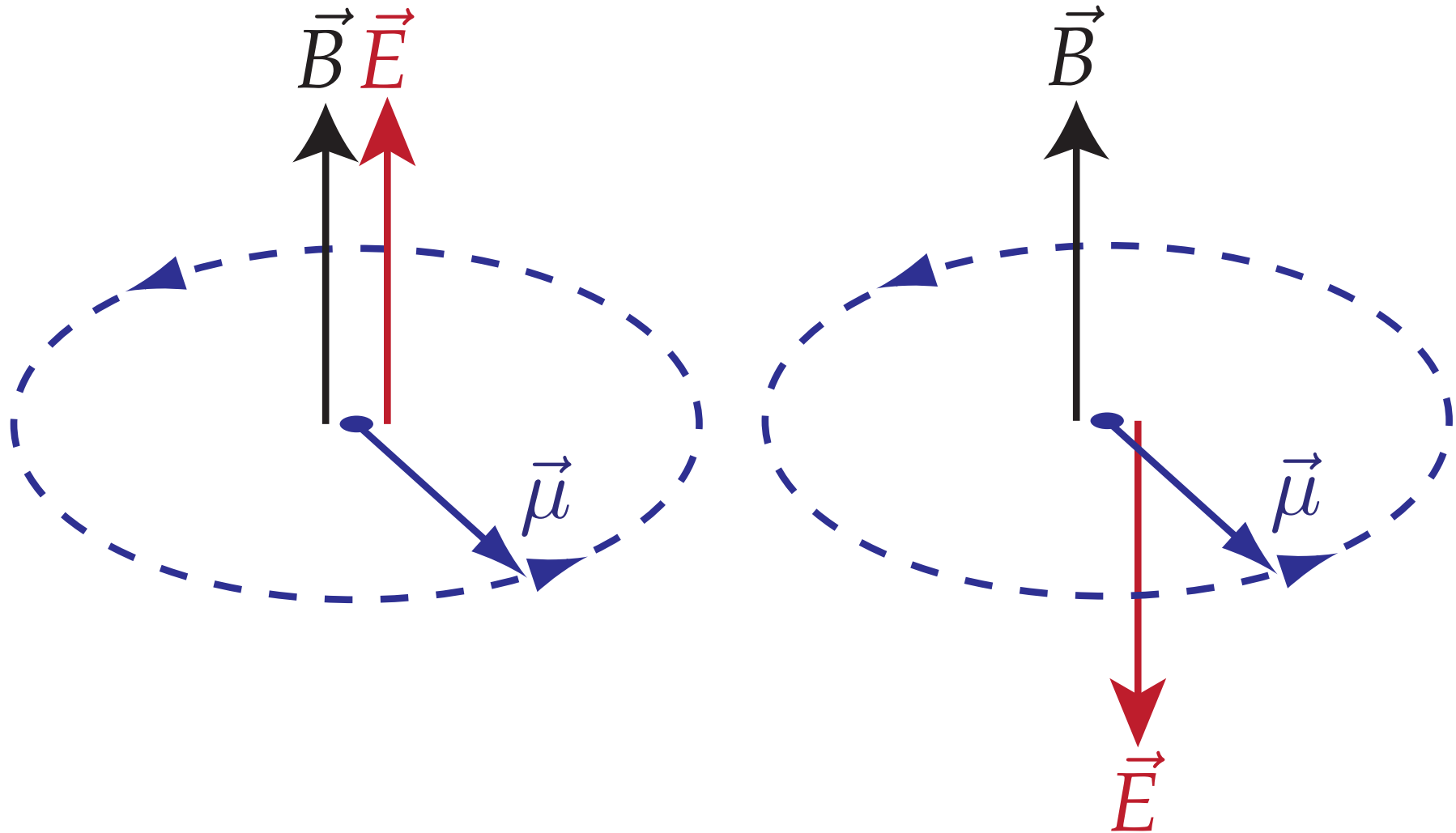
Different Sources of $\mathcal{CP} \Leftrightarrow$ “EDMs” of Different Systems



RMP 91
015001
(2019)

2021-06-28

Always Measure Frequency: Spin Precession



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

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

Ultimate Statistical Sensitivity

$$\Delta\nu = \nu_{\uparrow} - \nu_{\downarrow} = \frac{4dE}{h}$$

statistical sensitivity:

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

Electric
field

number of
particles

integration
time

interrogation
time

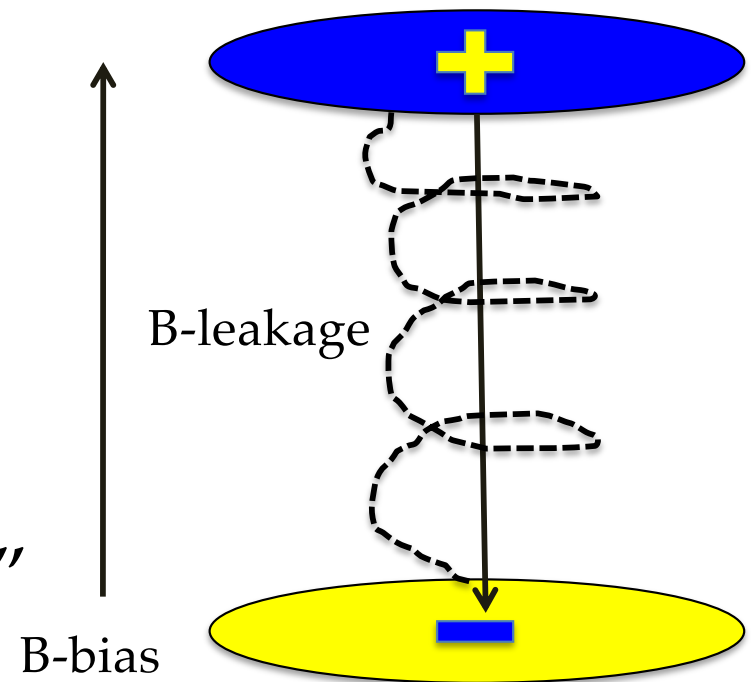
Electric Field-Correlated Systematic: Killer

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

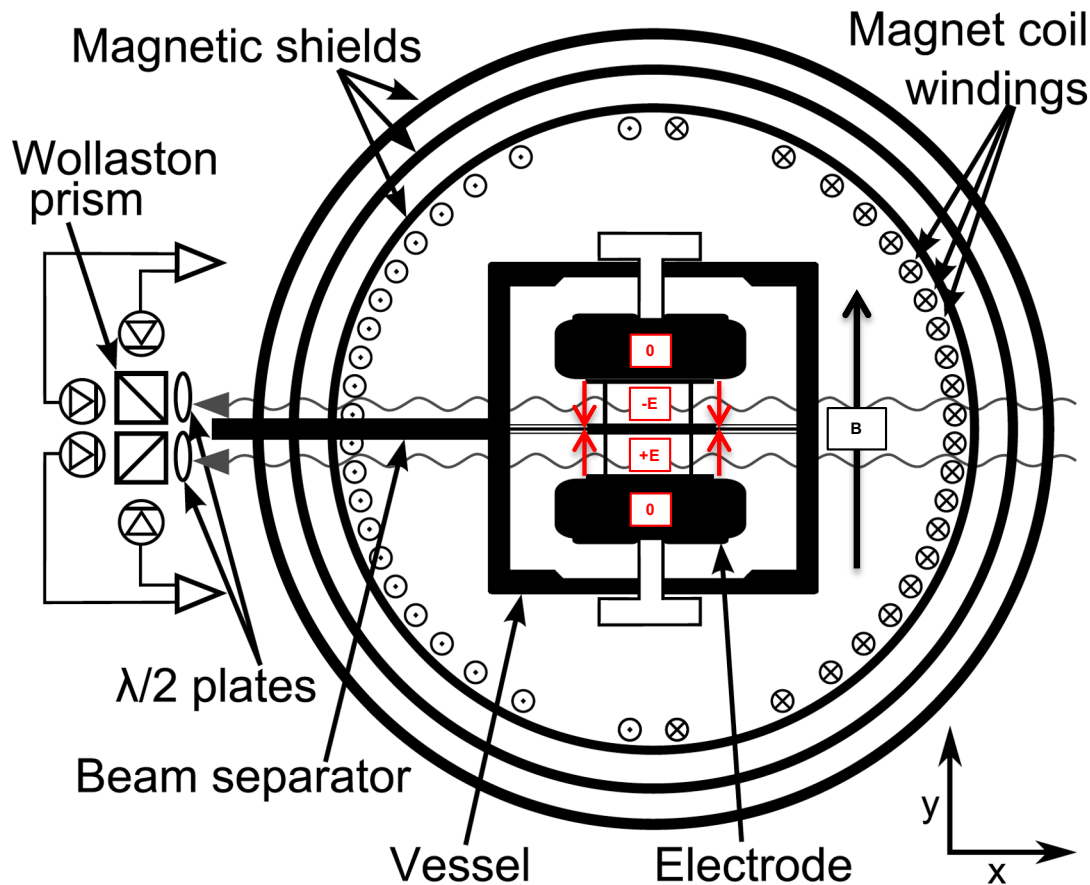
challenge!

Instabilities adds noise & limits the statistical precision.

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



The Seattle 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, ($10^{13} / \text{cm}^3$)
- 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)

Shielding Imperfect with Relativistic Atoms & Finite Nuclei

- Shielding in Diamagnetic Atoms

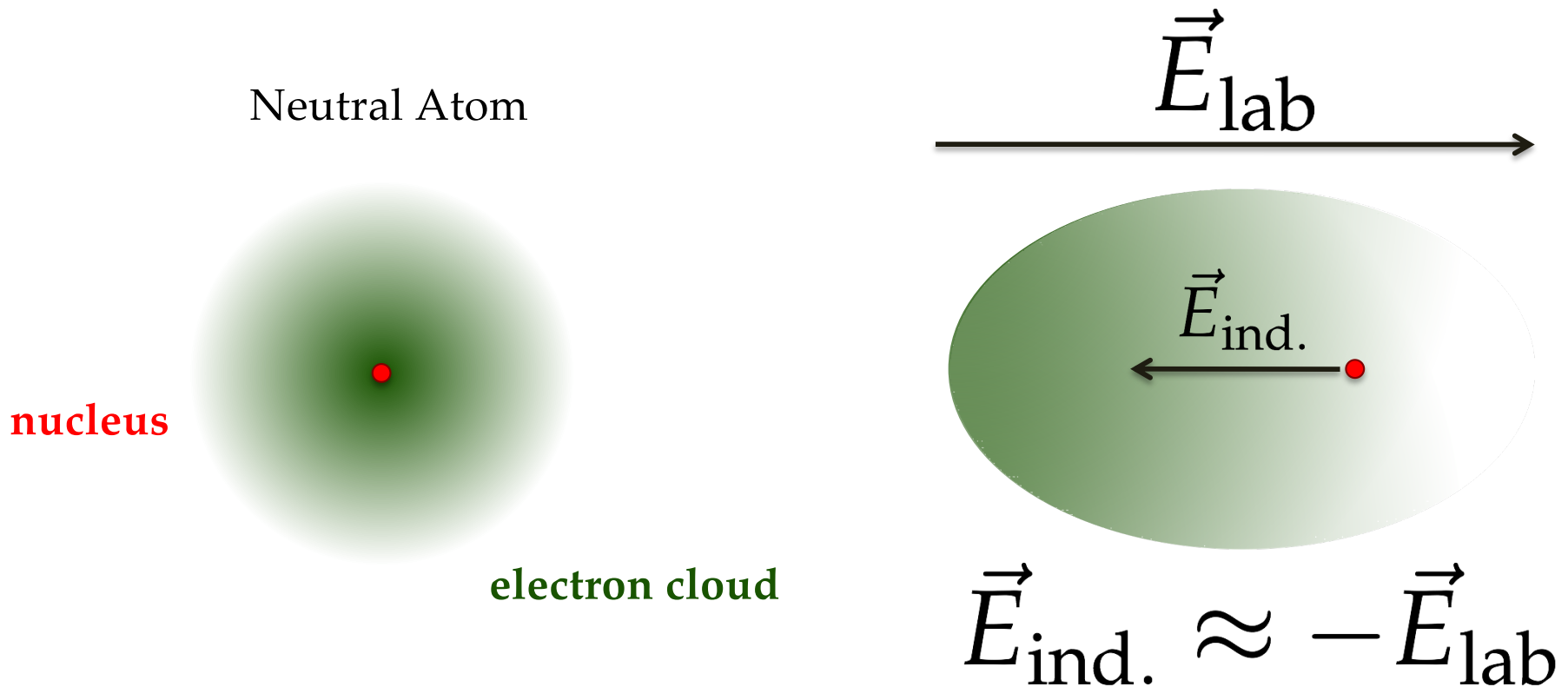
Schiff PR 132:2194 (1963)

- 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

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



Enhanced Sensitivity in “Pear”-Shaped Nuclei Like Radium-225

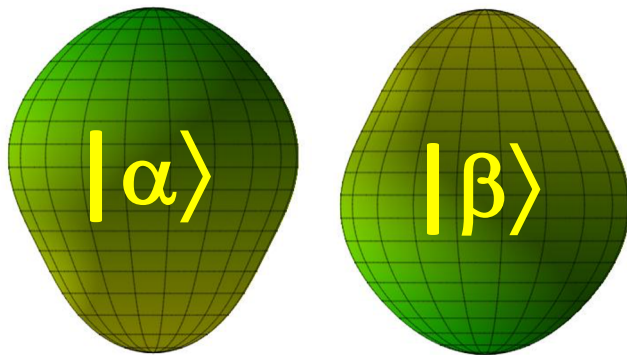
$$S_z = \frac{\langle er^2z \rangle}{10} - \frac{\langle r^2 \rangle \langle ez \rangle}{6}$$

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



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

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

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

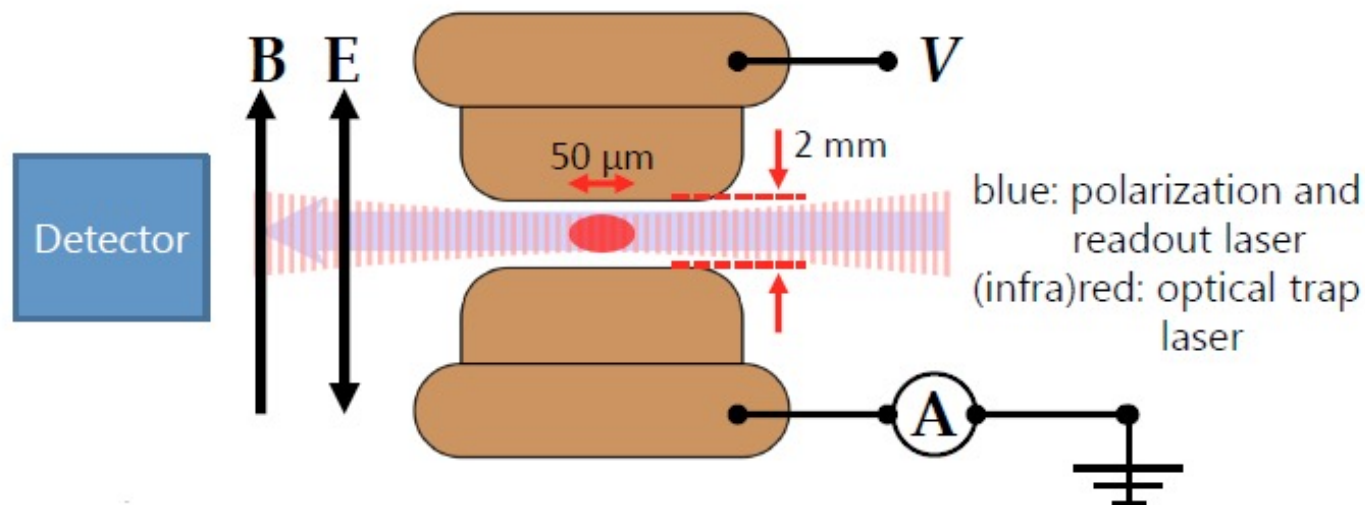
^{199}Hg : Ban et al. PRC 82:015501 (2010)

55 keV

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

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

The ANL Laser Trap Atomic Ra EDM Search



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)

^{225}Ra

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

Electronic Spin = 0

$t_{1/2}$ = 15 days

Low vapor pressure

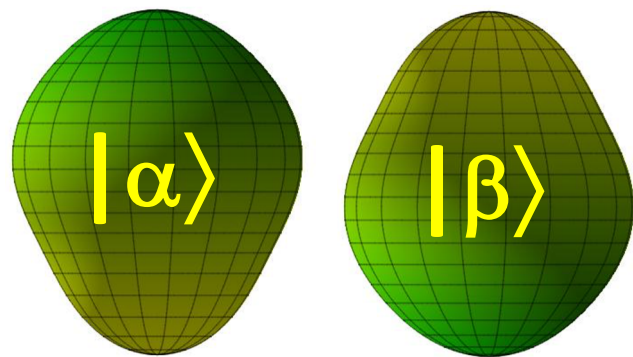
- Long coherence time (100 s)
- negligible “ $v \times E$ ” systematics
- High electric field (0.1-0.5 MV/cm) in vacuum
- Light-induced systematic effects can be controlled!
- Statistics-dominated “proof-of-principle” experiments

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

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

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

Parity Doublet



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

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

ΔE

Pa-229: 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 produce $>10^9$ /sec of both
Ra-225 & Pa-229!**



Towards Precision Pa-229 Nuclear Spectroscopy

PHYSICAL REVIEW C **97**, 054310 (2018)

Accurate measurement of the first excited nuclear state in ^{235}U

F. Ponce, E. Swanberg, J. Burke, R. Henderson, and S. Friedrich

Lawrence Livermore National Laboratory, 7000 East Avenue, Livermore, California 94550, USA



(Received 13 July 2017; revised manuscript received 27 February 2018; published 10 May 2018)

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 independent of the decay mode or the chemical state of the uranium. The detector is calibrated using an energy comb from a pulsed UV laser. A comparable measurement of the metastable ^{229m}Th nucleus would enable a laser search for the exact transition energy in $^{229}\text{Th} - ^{229m}\text{Th}$ as a step towards developing the first ever nuclear (baryonic) clock.

DOI: [10.1103/PhysRevC.97.054310](https://doi.org/10.1103/PhysRevC.97.054310)

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

- Large intrinsic sensitivity to BSM physics JTS Hyp. Int. 240:29 (2019)
 - **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 (MQM) to amplify observable
 - **local crystal fields (1-10 MV/cm) (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?**
- **Control of systematics – co-magnetometer ($^{141}\text{Pr}^{+3}$, $I=5/2$, stable)?**

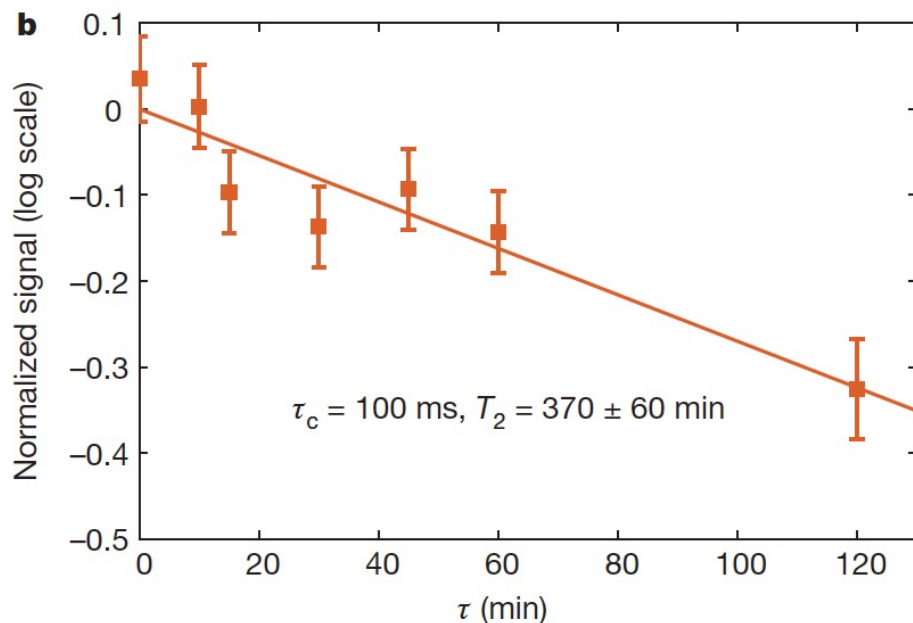
Long Coherence Times of Lanthanide Ion Nuclei

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 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 T_2 of $^{151}\text{Eu}^{3+}$ ($I=5/2$) embedded in Y_2SiO_5 was measured to be over 6 hours.**

Stable Molecule Experiments

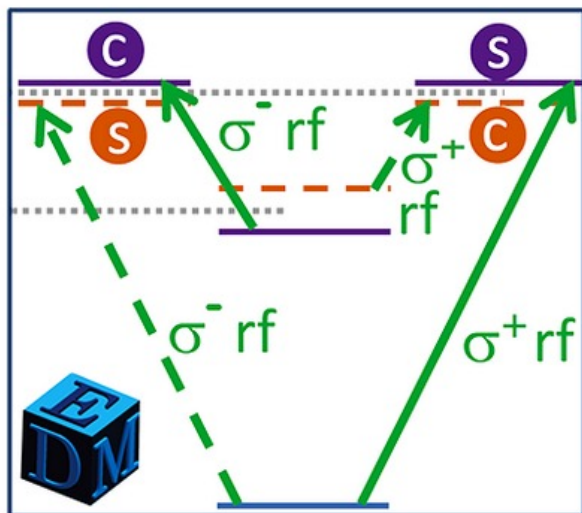
Polar molecules have been very successful for electron EDM searches! [Nature 562:355 \(2018\)](#) & [PRL 119:153001 \(2017\)](#)

- improved the electron EDM limit by 2 orders of magnitude over the last 10 years
- large “internal” electric fields (>10 GV/cm)
- rich internal molecular structure allows for exquisite control of systematics

Can the same be done in the hadronic sector?

- CeNTREX (Yale/UMass/Columbia) [arXiv:2010.01451](#)
 - nuclear Schiff moment of Thallium-205 via TlF
- Polyatomic systems (Caltech) [QST 5 044011](#)
 - magnetic quadrupole moment of Yb-173 via YbOH
- Trapped Molecular Ions (UNLV) [J. Mol. Spec. 358, \(2019\) 1-16](#)
 - magnetic quadrupole moment of Ta-181 via TaO⁺

Oriented Molecules in Noble Gas Solids



EDM³

York – Hessels
Toronto - Vutha
MSU – JTS
with generous
support from
Moore and Sloan
Foundations!

EDITORS' SUGGESTION

Orientation-dependent hyperfine structure of polar molecules in a rare-gas matrix: A scheme for measuring the electron electric dipole moment

The Stark shift of the hyperfine states of polar molecules embedded in a solid rare-gas matrix is found to depend on the molecular orientation. This finding may significantly improve the measurements of the electron electric dipole moment by using large ensembles of polar molecules trapped in rare-gas matrices with orientation-dependent detections.

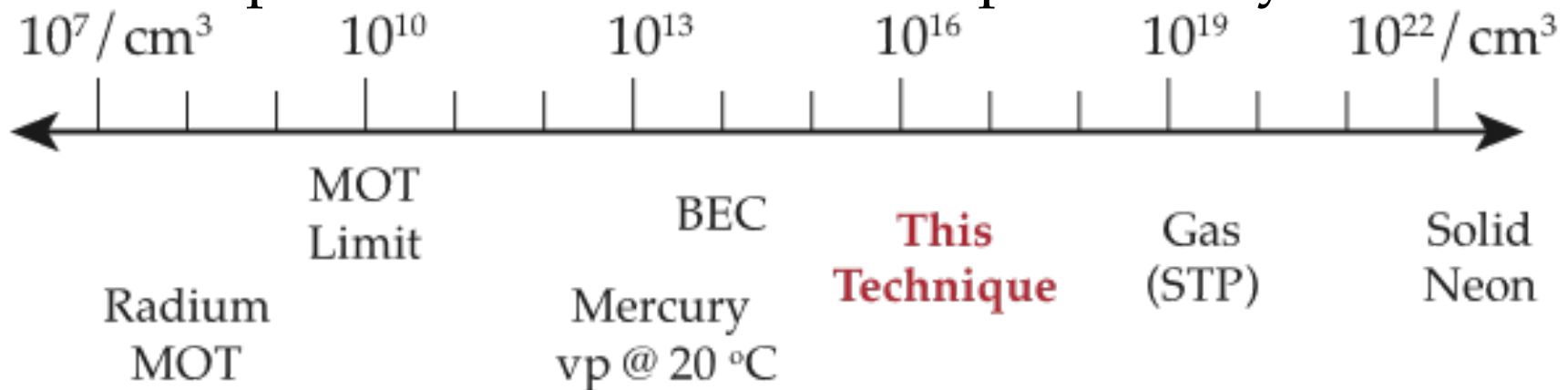
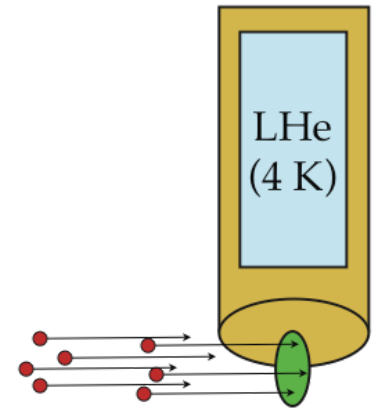
A. C. Vutha, M. Horbatsch, and E. A. Hessels

[Phys. Rev. A 98, 032513 \(2018\)](#)

co-magnetometry: nearby pairs of anti-aligned molecules

Advantages of Noble Gas Solids

1. Trapping of a wide variety of species
2. Stable confinement
3. Chemically inert (electronic ground state 1S_0)
4. Transparent in the optical regime
5. long T_1 : solid Xe-129 ($I=1/2$) 10^2 s @ 10 G & 77 K
 Gatzke, Cates, et al. PRL 70 693 (1993) 10^6 s @ 10^3 G & 4.2 K
6. long T_2 : 10^3 s for 1 ppm diamagnetic (μ_N) spin impurities
 Van Vleck PR 74 1168 (1948)
7. tunable particle number / nuclear spin density:



Oriented Radioactive Molecules In Noble Gas Solids

Opportunity: nuclear Schiff enhancement $\sim 10^{3+}$
and number densities $> 10^{13} / \text{cm}^3$
and $\sim 10^2$ MV / cm effective field

PRA 87:020102 (2013)

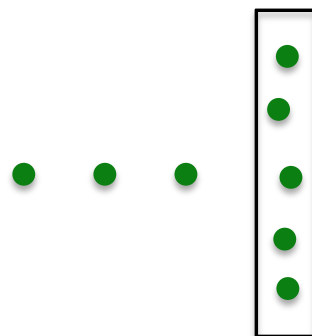
PRA 90:052513 (2014)

- $^{225}\text{Ra}^{19}\text{F}$ in NGS – nuclear Schiff moment (NSM)
 - stable surrogates: $^{138}\text{Ba}^{19}\text{F}$, $^{171}\text{Yb}^{19}\text{F}$
 - coherence times < 1 s (looks like an electron)
- $^{225}\text{Ra}^{16}\text{O}$ in NGS – nuclear Schiff moment
 - stable surrogates: $^{138}\text{Ba}^{16}\text{O}$, $^{171}\text{Yb}^{16}\text{O}$
 - coherence times $\sim 10^3$ s (looks like a nucleus)
- some ^{229}Pa containing molecule: NSM or MQM
- **One Challenge: how do we efficiently produce radioactive molecules?**

Form, Filter, Neutralize, and Co-Deposit Molecules

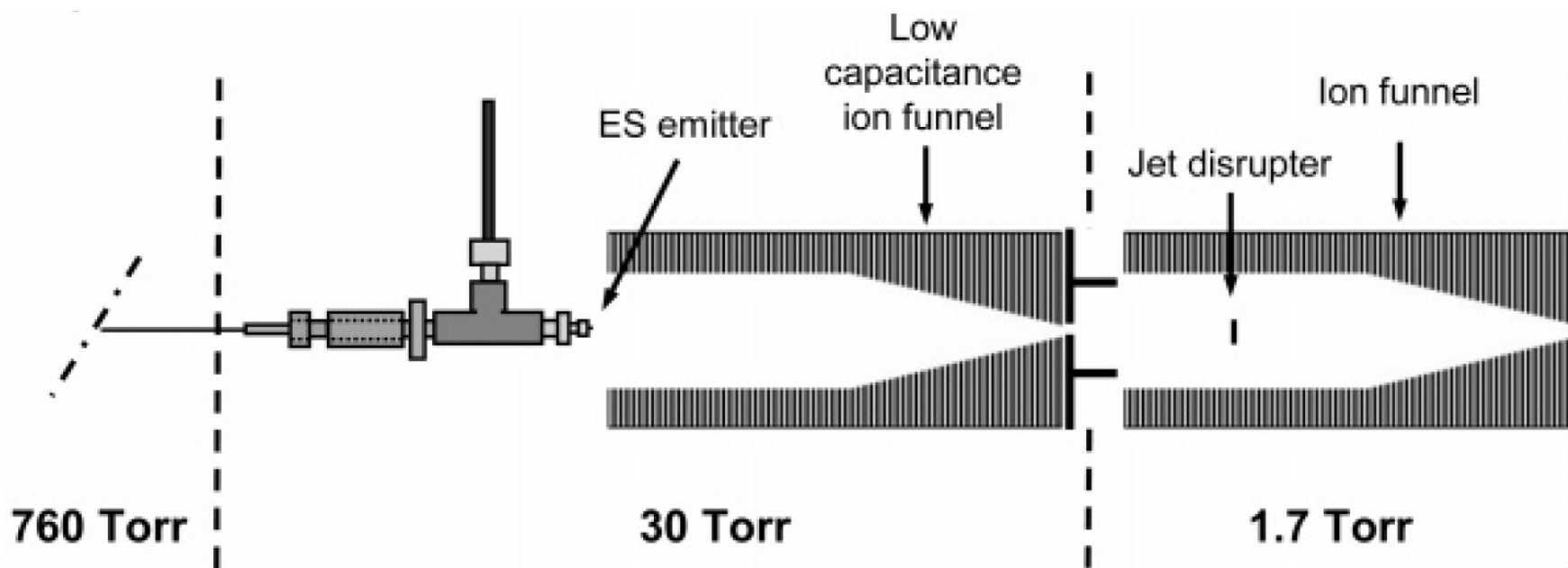


Nanoelectrospray
Ionization Source



Efficiencies of 10-50%
have been reported in
these ion sources.

J. Am. Soc. Mass Spectrom. (2015) 26:55-62



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Amitchell125 at English Wikipedia [CC BY-SA 3.0 (<https://creativecommons.org/licenses/by-sa/3.0>)], via Wikimedia Commons

Big Open Questions

- How efficiently can we make molecules using a nano-electrospray ionization source?
- What are the optical pumping & cycling properties of molecules in medium?
- To what degree are molecules oriented in medium?
- To what degree can the quantity of spin impurities be minimized in medium?
- What is a viable measurement scheme for nuclear Schiff moments of oriented molecules in medium?

These are our current research goals!

Thank For Your Attention!

1. Detecting a non-zero EDM would be an unambiguous signature of Physics beyond the Standard Model.
2. Pear-shaped nuclei such as Radium-225 and Protactinium-229 have significantly enhanced sensitivity to CP-violation originating within the nuclear medium.
3. Maximizing the discovery potential of rare isotopes requires an efficient technique for trapping and probing the nuclei.
4. Implantation into solids allows for high number densities, may allow for high efficiency optical probing, provides large internal electric fields for oriented molecules or ions in optical crystals, and allows for co-magnetometry.

**My group is looking for a Postdoc for this project! Come find me or:
singhj@frib.msu.edu web: spinlab.me twitter: [@spinlabmsu](https://twitter.com/spinlabmsu)**



**US Department of Energy, Office of Science,
Office of Nuclear Physics: DE-SC0019015 (ECA)**