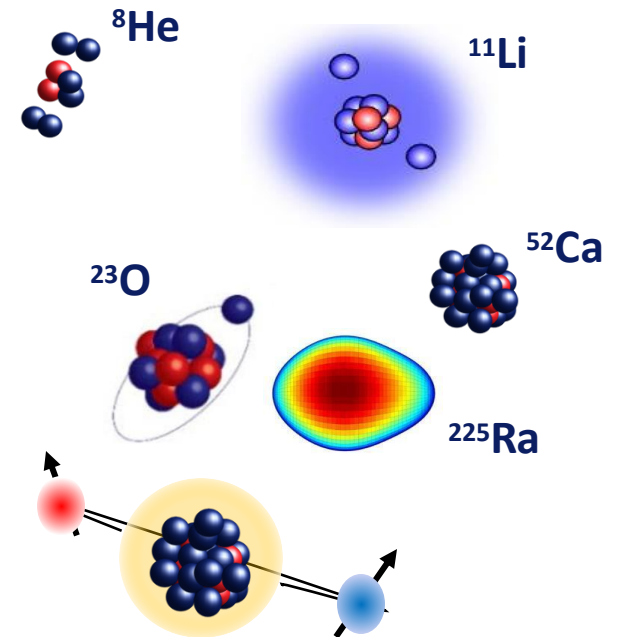


Radioactive Atoms and Molecules as Quantum Sensors for Nuclear Physics



Figure modified from <https://sphereofinfluence360.com/>



Ronald F. Garcia Ruiz, MIT

Quantum Information Science on the Intersections of Nuclear and AMO Physics

UMass Boston, January, 2025



Graduate Students



S. Udrescu
(-> U. Chicago)



A. Brinson
NSF Fellowship



H. Kakiota
Ezoe Memorial
Fellowship



S. Moroch
Hertz Foundation
Fellowship



F. Pastrana
Santo Domingo
Fellowship



D. Gonzales
(Harvard)



J. Munoz
MIT Fellowship



M. Fulghieri



F. Shungo

Postdoctoral Researchers



S. Wilkins



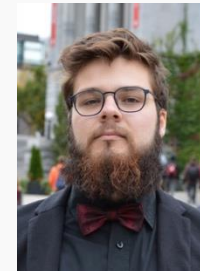
J. Karthein
Now Faculty at Texas A&M



S. Ebadi
Pappalardo Fellowship



A. Jadbabaie
NSF MPS Ascend Fellow



A. Belly
NSERC Fellow

PI



R.F. Garcia Ruiz

Undergraduate Students



K. Khusainova



S. Munoz



A. Fernandez



R. Hernandez



S. Becerra



**Bates
Lab**

Thanks to...



Nuclear & Atomic & Molecular

Theory

Experiment

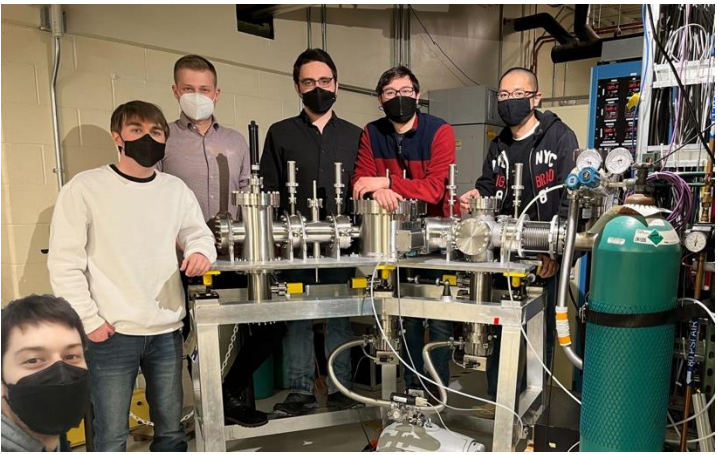


J. Dobaczewski (York)
J. Holt (TRIUMF)
R. Stroberg (U ND)
W. Nazarewicz (FRIB/MSU)

R. Berger (Malbroune)
A. Borschevsky (Groningen)
L. Skripnikov (Petersburg)



RISE @ FRIB



RaX Collaboration



NEPTUNE Project

Caltech

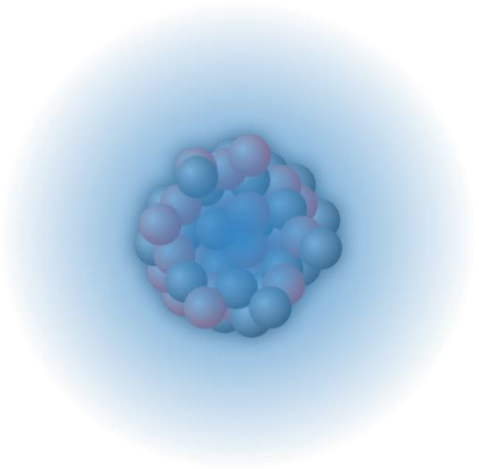


Caltech



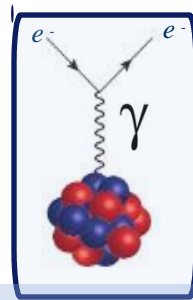
Why Atoms & Molecules?

Ra⁺



Atom ($S_{1/2}$)

Long range (> 1 fm)

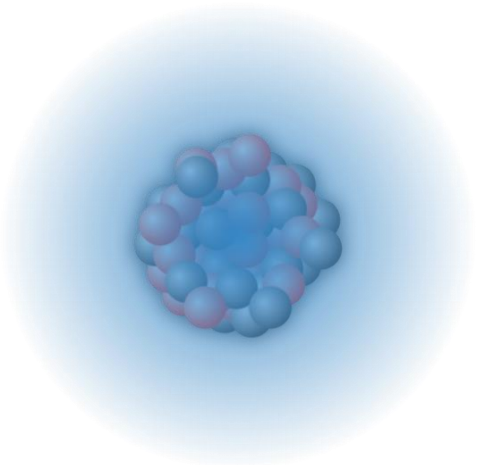


**Electromagnetic nuclear
properties $\langle r^2 \rangle, I, \mu, Q, \dots$**

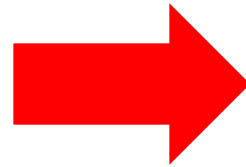


Why Atoms & Molecules?

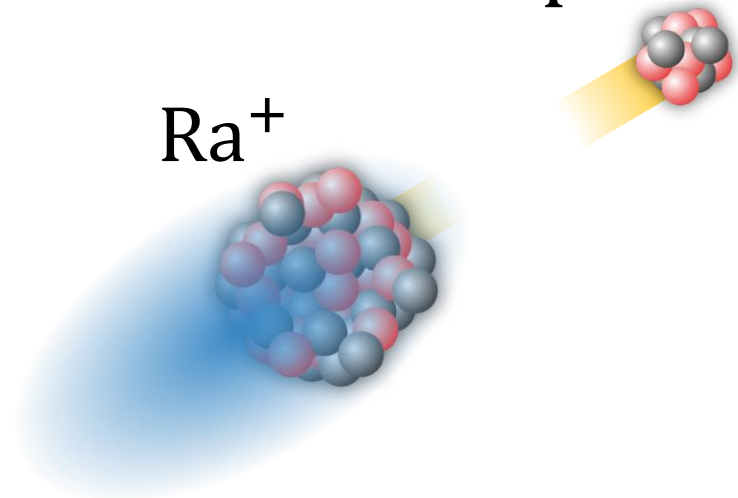
Ra⁺



Atom ($S_{1/2}$)



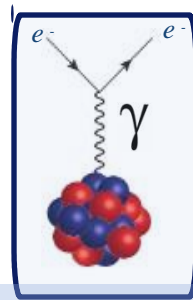
F⁻



Ra⁺

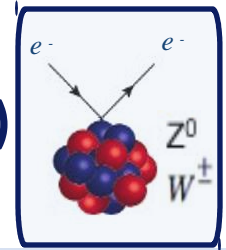
Molecule ($\Sigma_{1/2}$)

Long range (> 1 fm)



Electromagnetic nuclear properties $\langle r^2 \rangle, I, \mu, Q, \dots$

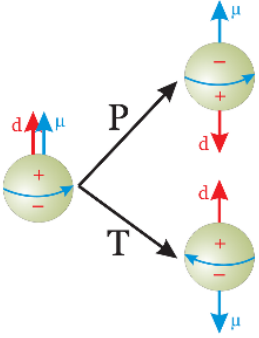
Short range (< 1 fm)



Electroweak nuclear properties (P,T - violation⁵)

Why Atoms & Molecules?

Eric Cornell's Talk
Tuesday at 10 am



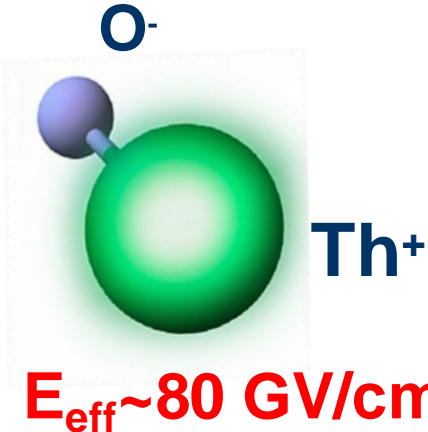
[Sandars Phys. Rev. Lett. 18, 1396 (1967)]
[Hudson et al. Nature 473 493 (2011)]

Nuclear spin = 0

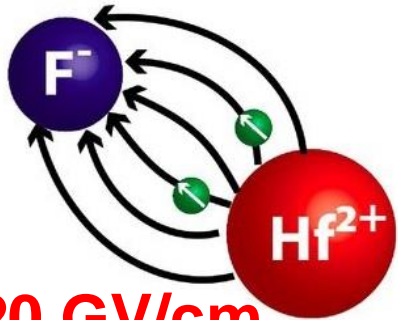
[1 eV=241.8 THz]

$$H_{mol} = H_e + H_{vib} + H_{rot} + \dots + H_{hfs} + H_{PV} + H_{PTV} + d_e E_{eff}$$

eV ~ 2 10^{-2} 10^{-5} $< 10^{-20}$



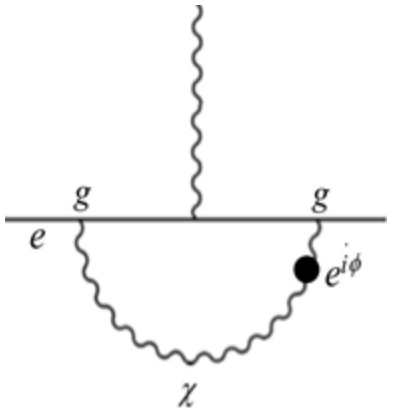
[ACME, Nature 562, 355 (2018)]



$E_{eff} \sim 20 \text{ GV/cm}$

$|d_e| \leq 4.1 \times 10^{-30} \text{ e} \cdot \text{cm}$

[Roussy et al. Science 381, 46 (2023)]



$$d_e \sim \mu_B \left(\frac{g^2}{2\pi} \right)^N \left(\frac{m_e}{m_\chi} \right)^2 \sin \phi$$

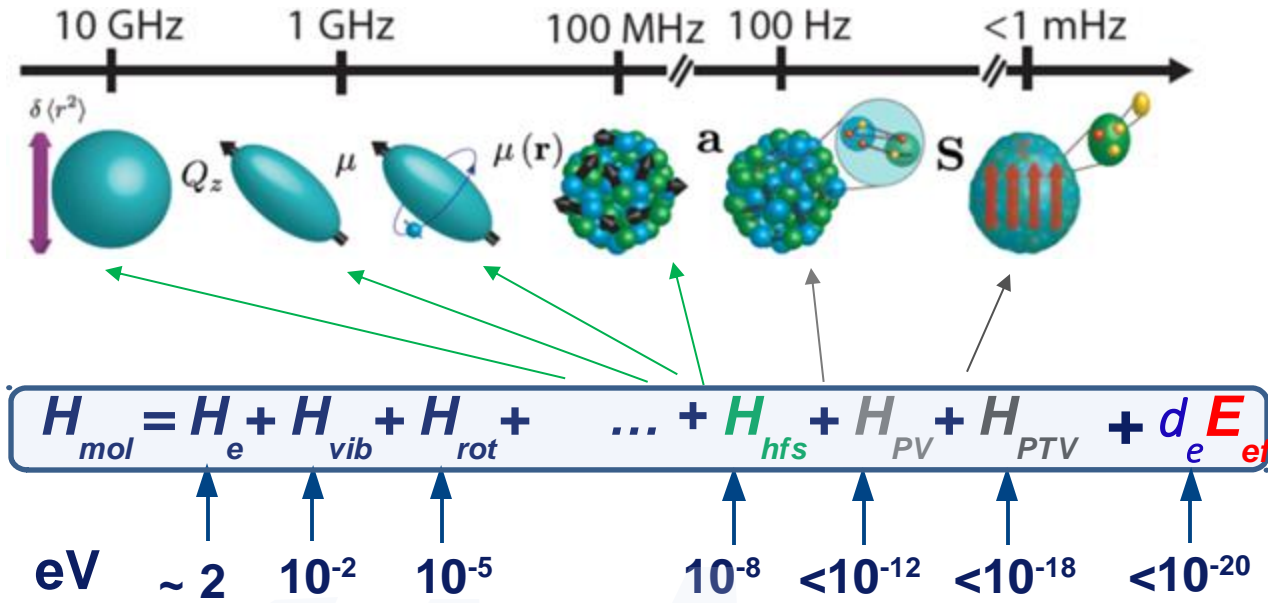
Probing physics @ TeV scale!

Why Atoms & Molecules?

Molecules with heavy, exotic nuclei: Ra(Z=88)

Nuclear spin > 0

[1 eV=241.8 THz]



$$H_{mol} = H_e + H_{vib} + H_{rot} + \dots + H_{hfs} + H_{PV} + H_{PTV} + d E_{eff}$$



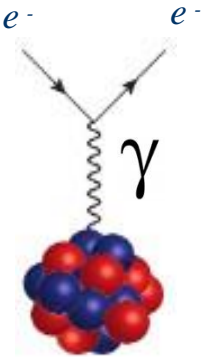
[Sandars Phys. Rev. Lett. 18, 1396 (1967)]
 [Hudson et al. Nature 473 493 (2011)]

Why atoms & molecules?

$$H_{mol} = H_e + H_{vib} + H_{rot} + \dots + H_{hfs} + H_{PV} + H_{PTV}$$

Dean Lee's Talk
Monday at 11:30 am

H_{hfs}



P,T-even

- Nuclear structure
- Nuclear matter

$\langle r^2 \rangle, I, \mu, Q, \dots$

How do nuclear phenomena emerge from QCD?

What are the properties of nuclear matter (e.g. neutron stars)?



Nuclei



Neutron star

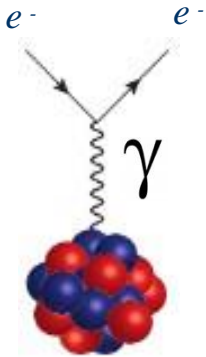
Recent highlights:

- Nature Physics 20,1719 (2024)
- Phys. Rev. Lett. 133, 033003 (2024)
- Nature Physics 20, 202 (2024)
- Physical Review Letters 132, 162502 (2024)
- Rep. Prog. Phys. 87 084301 (2024)
- Physical Review Letters 131, 222502 (2023)
- Nature 607, 260 (2022)
- Physical Review Letters 128, 022502 (2022)

Why atoms & molecules?

$$H_{mol} = H_e + H_{vib} + H_{rot} + \dots + H_{hfs} + H_{PV} + H_{PTV}$$

H_{hfs}

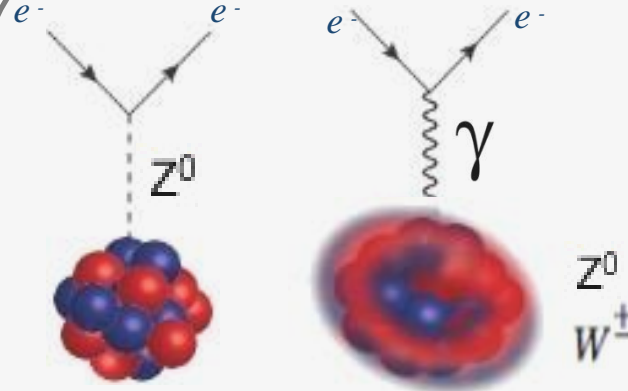


P,T-even

- Nuclear structure
- Nuclear matter

$\langle r^2 \rangle, I, \mu, Q, \dots$

H_{PV}

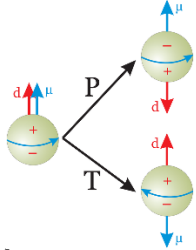


P-violation

- Electro weak structure
- Precision Standard Model tests
- Dark Mater properties?
- New forces?

Why atoms & molecules?

$$H_{mol} = H_e + H_{vib} + H_{rot} + \dots + H_{hfs} + H_{PV} + H_{PTV}$$



H_{hfs}

Antimatter

Matter

- Nuclear structure
- Nuclear matter

$\langle r^2 \rangle, I, \mu, Q, \dots$

H_{PV}

Strong CP problem

$$\mathcal{L} = \theta \frac{1}{16\pi^2} F_{\mu\nu}^a \tilde{F}^{\mu\nu a}$$

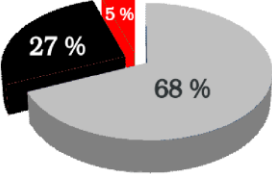
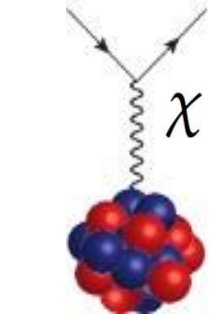
- Electro weak structure
- Precision Standard Model tests
- Dark Mater properties?
- New forces?

H_{PTV}

EDM MDM ^{223}Ra

T-violation

- Matter-antimatter asymmetry
- New particles?

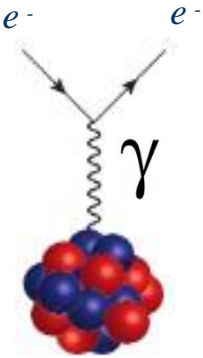


■ Ordinary Matter
■ Dark Matter
■ Dark Energy

Why atoms & molecules?

$$H_{mol} = H_e + H_{vib} + H_{rot} + \dots + H_{hfs} + H_{PV} + H_{PTV}$$

H_{hfs}

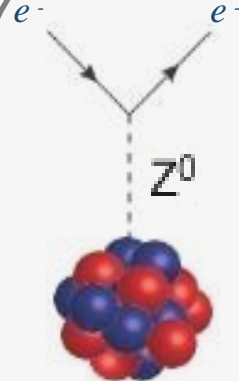


P,T-even

- Nuclear structure
- Nuclear matter

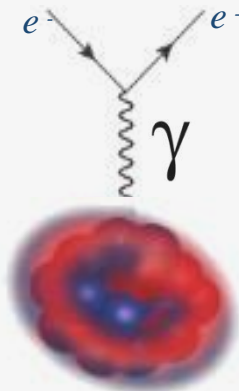
$\langle r^2 \rangle, I, \mu, Q, \dots$

H_{PV}



P-violation

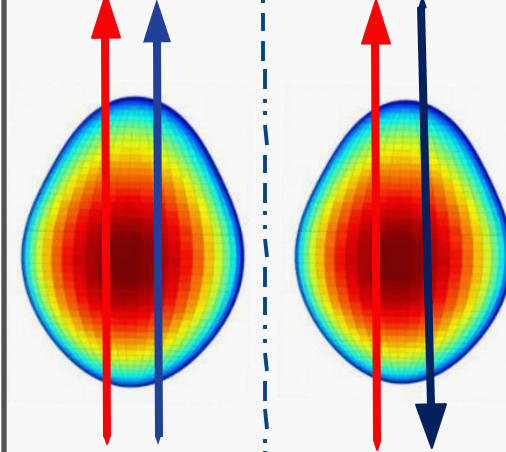
- Electro weak structure
- Precision Standard Model tests
- Dark Mater properties?
- New forces?



Z^0
 W^\pm

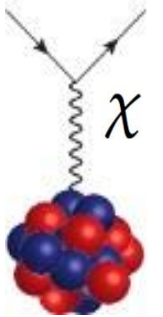
H_{PTV}

EDM MDM ^{223}Ra

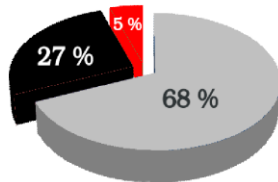


T-violation

- Matter-antimatter asymmetry
- New particles?



χ



■ Ordinary Matter
■ Dark Matter
■ Dark Energy

Why atoms & molecules?

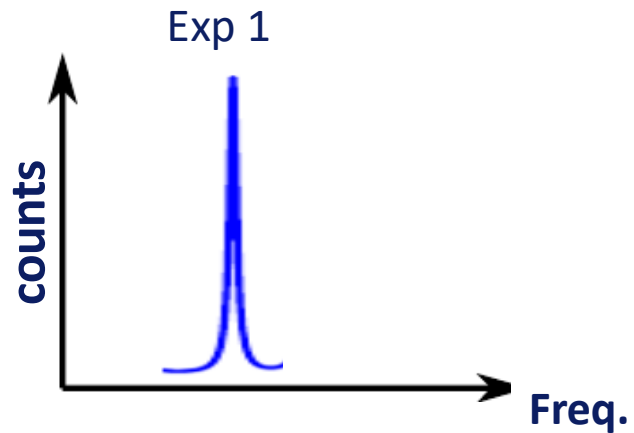
$$H_{mol} = H_e + H_{vib} + H_{rot} + \dots + H_{hfs} + H_{PV} + H_{PTV}$$

$\sim O_{Nucl} F_{mol}$

↑ ↑ ↑ ↑ ↑ ↑

eV ~ 2 10⁻² 10⁻⁵ 10⁻⁸ <10⁻¹² <10⁻¹⁸

$$H_{mol} - H'_{mol} \sim O_{Nucl} F_{mol} \quad \rightarrow \text{Direct measurements!}$$



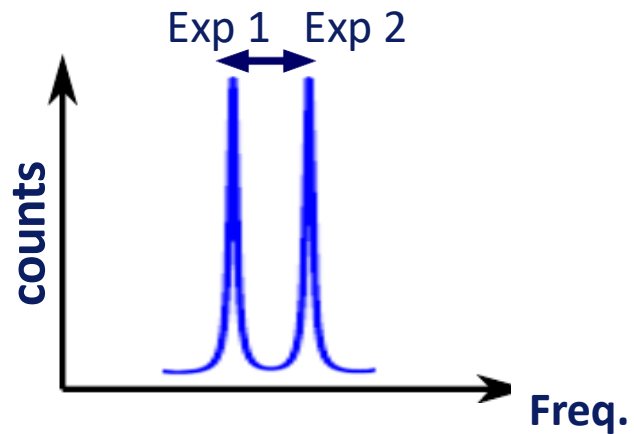
Why atoms & molecules?

$$H_{mol} = H_e + H_{vib} + H_{rot} + \dots + H_{hfs} + H_{PV} + H_{PTV}$$

$\sim O_{Nucl} F_{mol}$

eV ~ 2 10^{-2} 10^{-5} 10^{-8} $<10^{-12}$ $<10^{-18}$

$$H_{mol} - H'_{mol} \sim O_{Nucl} F_{mol} \quad \rightarrow \text{Direct measurements!}$$

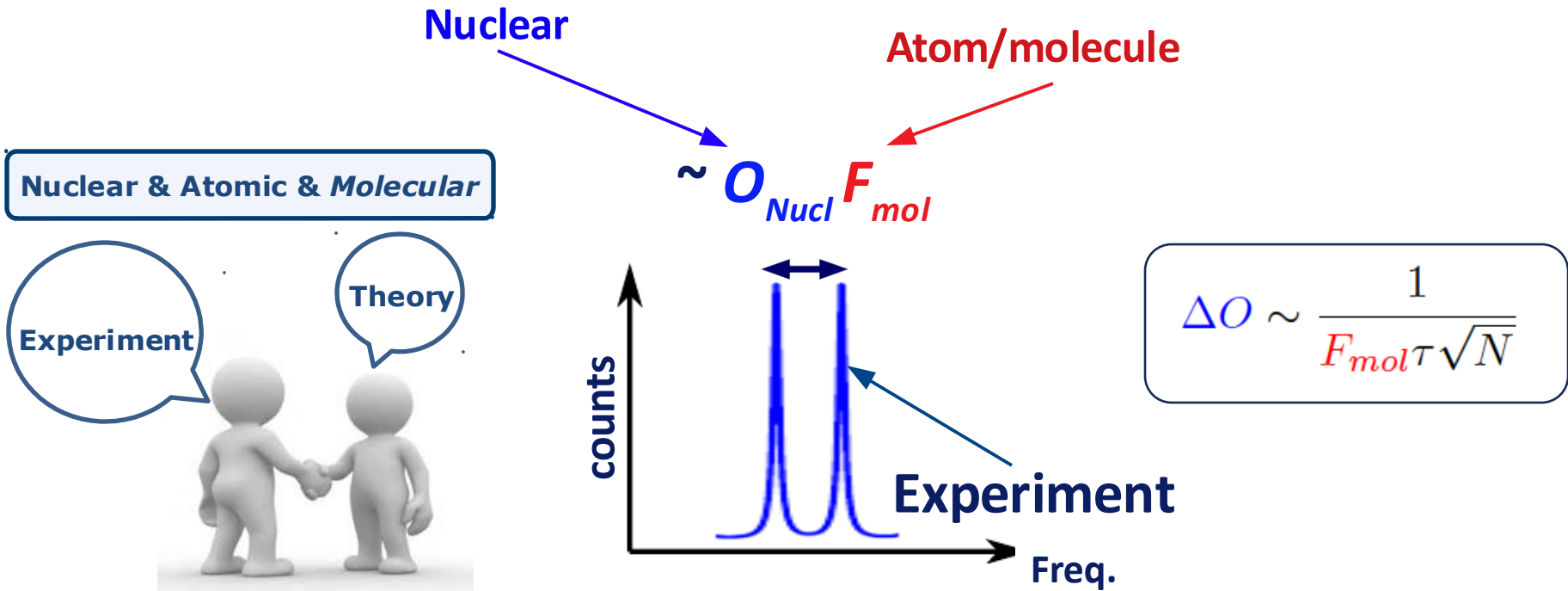


Why atoms & molecules?

$$H_{mol} = H_e + H_{vib} + H_{rot} + \dots + H_{hfs} + H_{PV} + H_{PTV} \quad \sim O_{Nucl} F_{mol}$$

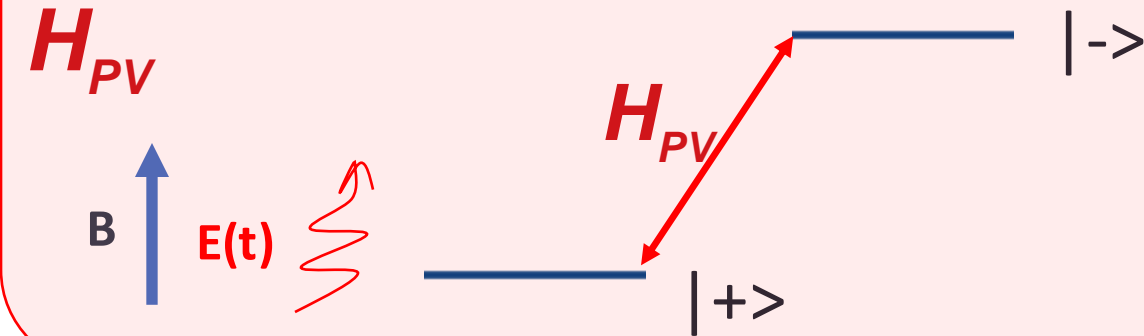
$\uparrow \quad \uparrow \quad \uparrow \quad \uparrow \quad \uparrow \quad \uparrow$
 eV ~ 2 10^{-2} 10^{-5} 10^{-8} $<10^{-12}$ $<10^{-15}$

$$H_{mol} - H'_{mol} \sim O_{Nucl} F_{mol} \quad \rightarrow \text{Direct measurements!}$$

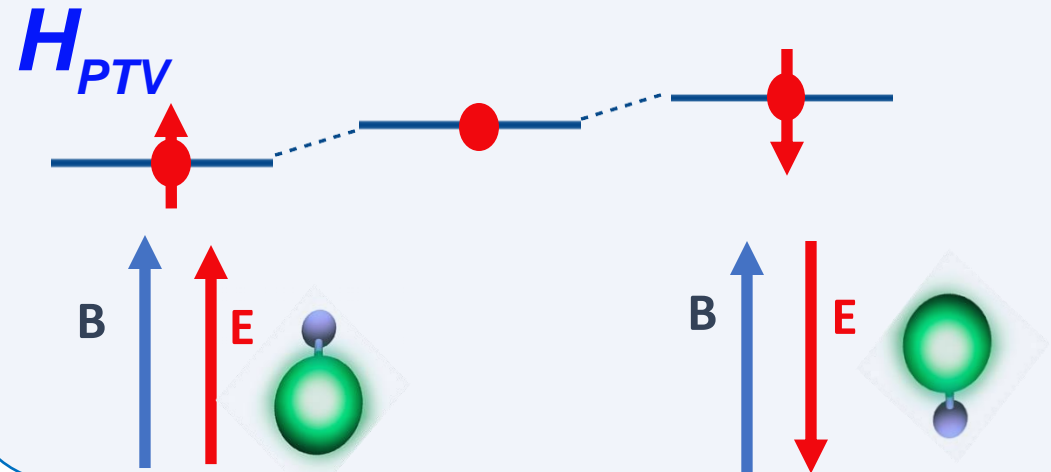


Molecules as “quantum sensors”

[Phys Rev Lett 120, 142501 (2018)]



[Nature 562, 355 (2018)] [Roussy et al. Science 381, 46 (2023)]



Eric Cornell's Talk
Tuesday at 10 am

Recent reviews:

- “Searches for new sources of CP violation using molecules as quantum sensors”. Hutzler et al. arXiv:2010.08709 (2020)
- “Opportunities for fundamental physics research with radioactive molecules”. Arrowsmith-Kron et al. Rep. Prog. Phys. 87 084301 (2024)

Radioactive molecules as sensitive probes of P,T-violating nuclear properties

$$\begin{array}{ccccccc}
 & & & & & \underbrace{\hspace{10em}} & \sim O_{Nucl} F_{mol} \\
 & & & & & \boxed{H_{mol} = H_e + H_{vib} + H_{rot} + \dots + H_{hfs} + H_{PV} +} & \\
 H_{PTV} & \uparrow & \uparrow & \uparrow & & \uparrow & \uparrow & \uparrow \\
 \text{eV} & \sim 2 & 10^{-2} & 10^{-5} & & 10^{-8} & < 10^{-12} & < 10^{-18}
 \end{array}$$

Radioactive Molecules for studies of P, T violation

^{225}Ra

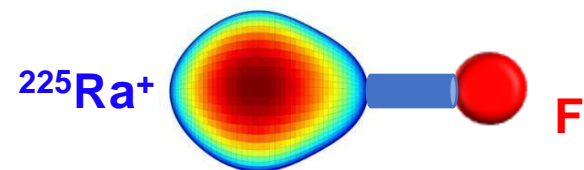
- ✓ Large Z, A
- ✓ Nuclear spin I > 0
- ✓ $\beta_2 \beta_3 > 0$

$$\begin{aligned}
 & \text{Nuclear} > 10^3 & \text{Atom/molecule} > 10^3 \\
 & \sim Z^a A^b \beta_2 \beta_3^2 / (E_+^N - E_-^N) & \sim Z^e / (E_+^{e-} - E_-^{e-}) \\
 & \searrow & \swarrow \\
 & \sim O_{\text{Nucl}} & F_{\text{mol}} \\
 & & E_+^N - E_-^N \sim 10^{-5} \text{ eV}
 \end{aligned}$$



[Gaffney et al. Nature 497, 199 (2013)]

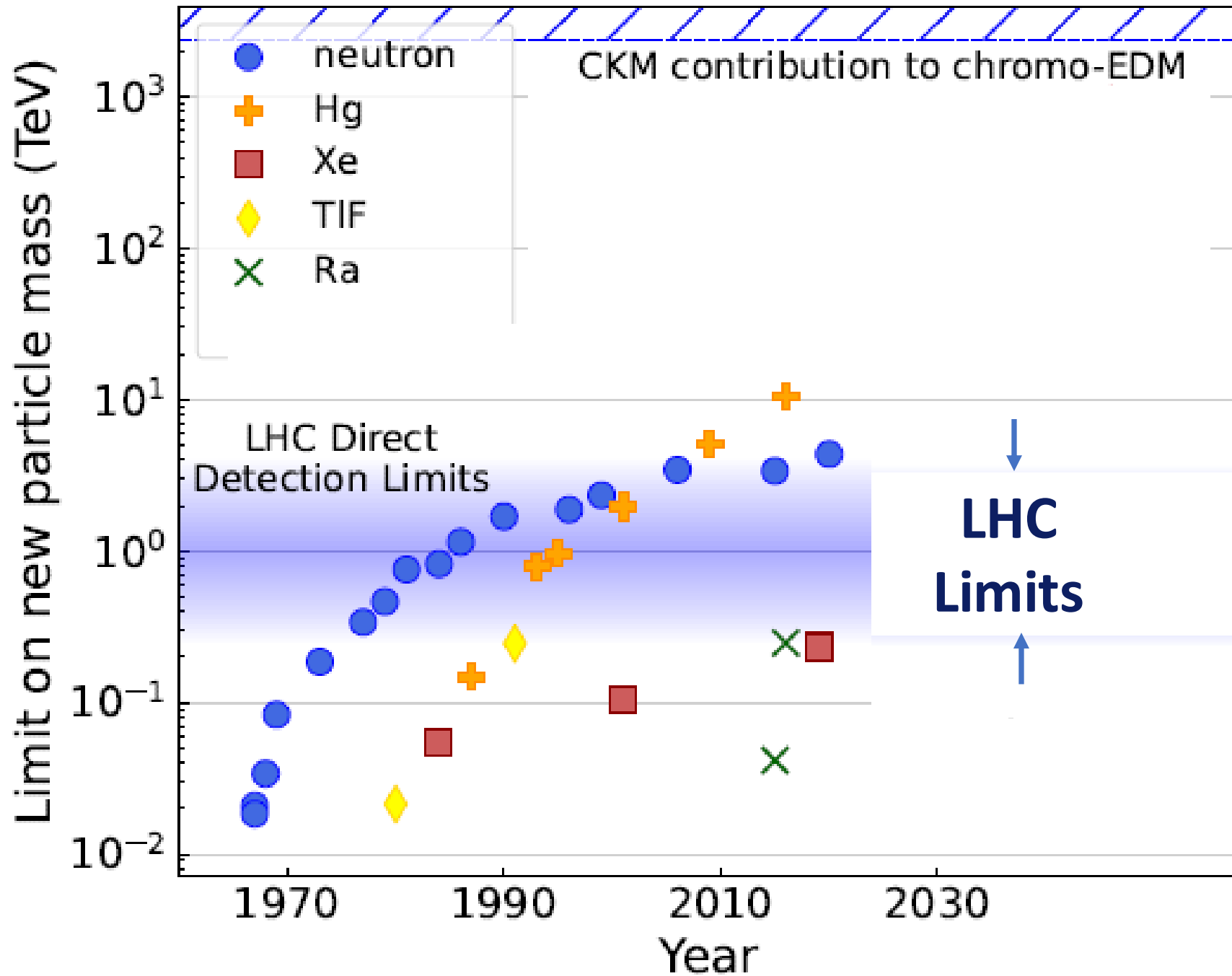
[Ra@ ANL: Parker et al. PRL 114, 233002 (2015)]



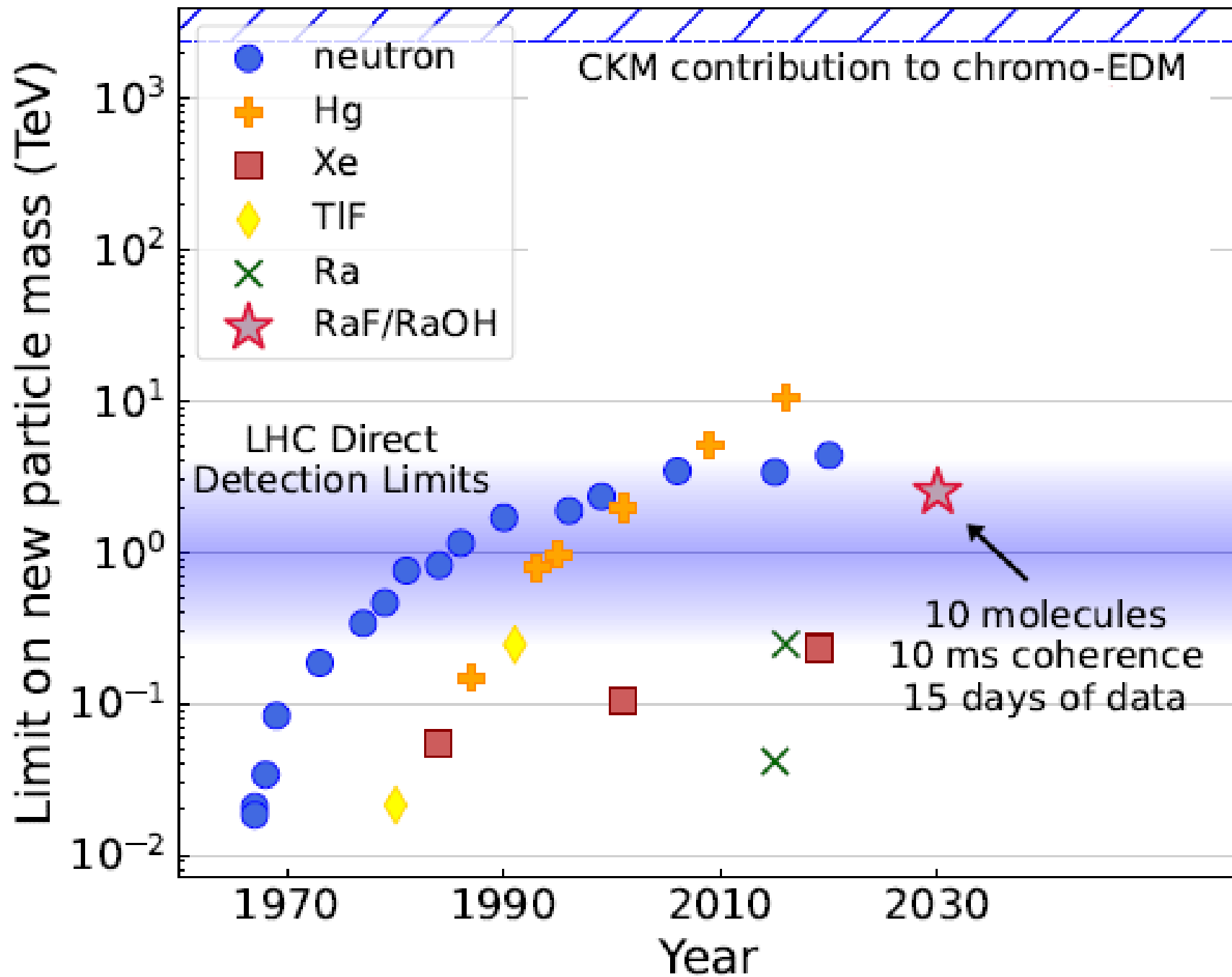
Radioactive molecules => **Best of all worlds!**

=> $> 10^6$

Hadronic EDMs



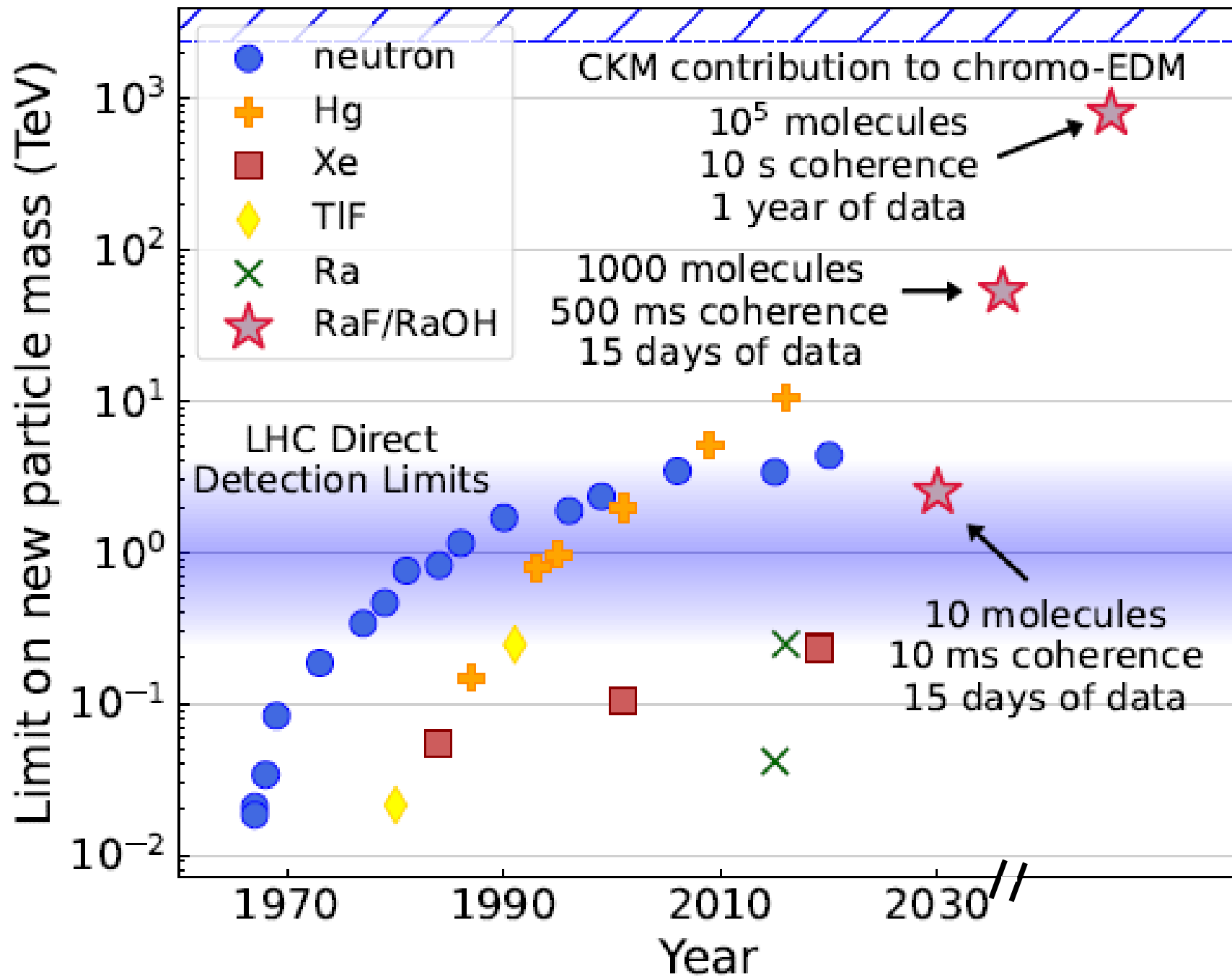
Hadronic EDMs



Window of Discovery



Hadronic EDMs

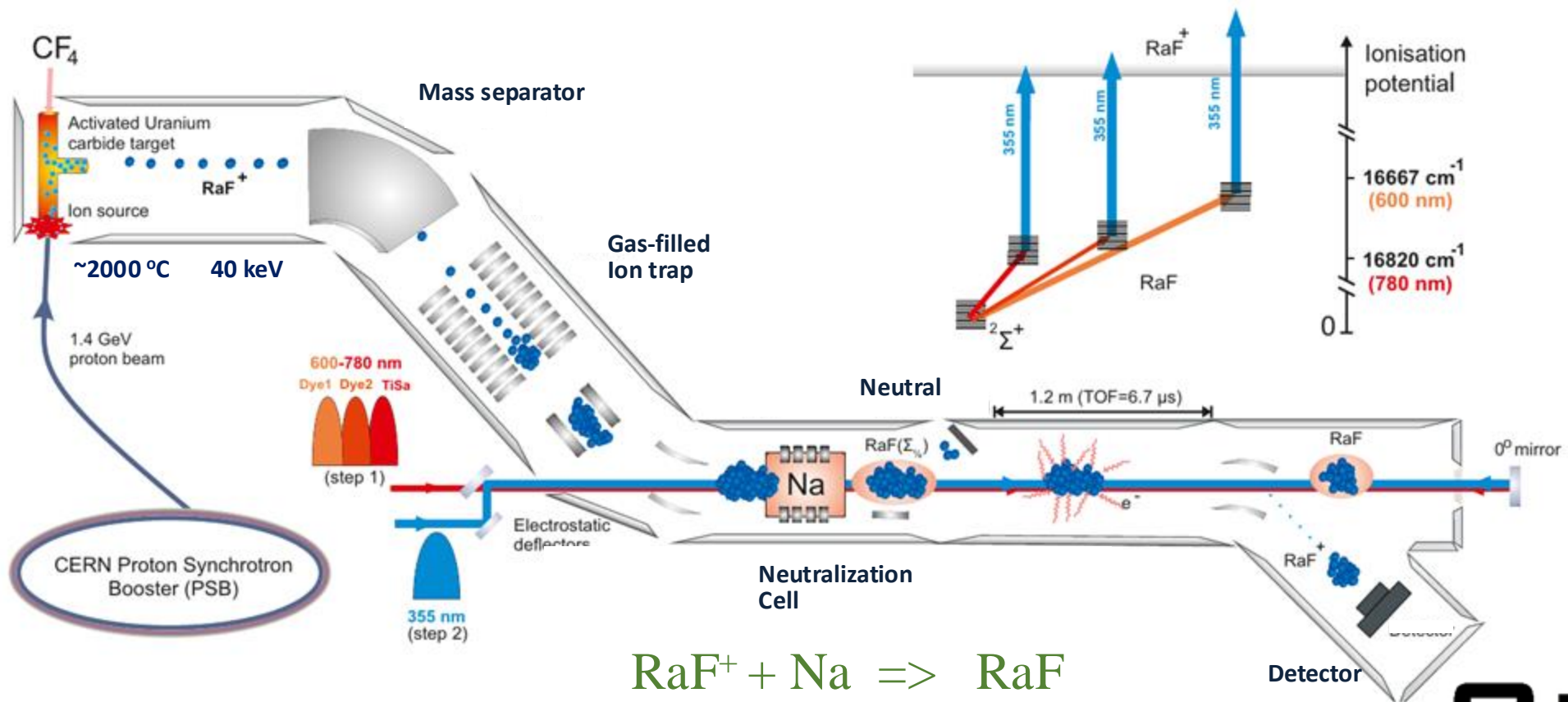


Window of Discovery



Recent Results (RaF)

ISOLDE



CRIS

Recent Results (RaF)



nature

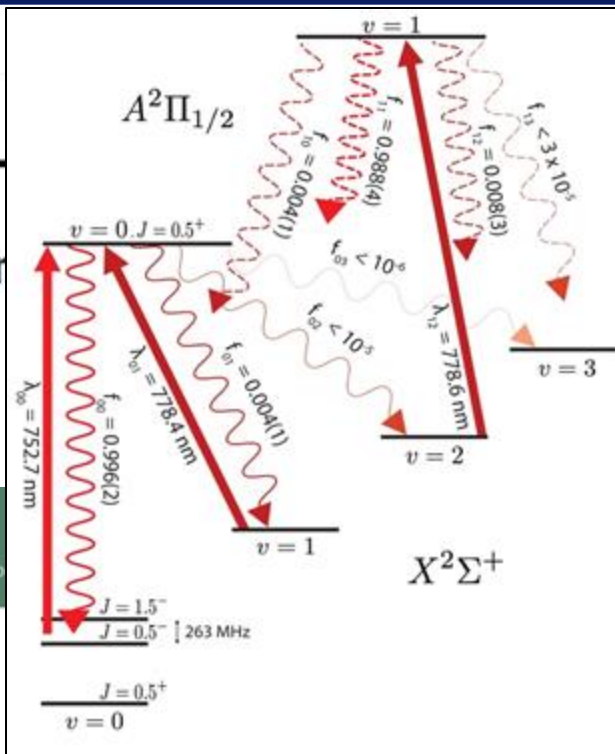
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Article | Open Access | Published: 27 May 2020

Spectroscopy of short-lived radioactive

R. F. Garcia Ruiz ✉, R. Berger ✉, [...]

Nature 581, 396–400 (2020) | Cite this article



Nature 581, 396 (2020)

PRL 127, 033001 (2021)

Nature Phys 20, 202 (2024)

(2024)
under review



PHYSICAL REVIEW LETTERS

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Featured in Physics Editors' Suggestion Open Access

Isotope Shifts of Radium Monofluoride Molecules

S. M. Udrescu et al.
Phys. Rev. Lett. 127, 033001 – Published 14 July 2021

PhysICS See Viewpoint: Sizing up Exotic Nuclei with Radioactive Molecules



$$H_{mol} = H_e + H_{vib} + H_{rot} + \dots + hfs + H_{PV} + H_{PTV}$$

eV ~ 2 10⁻² 10⁻⁵ 10⁻⁸ <10⁻¹² <10⁻¹⁵

naturephysics

nature > naturephysics > articles > article

Article | Published: 09 January 2024

Precision spectroscopy and laser-cooling scheme of a radium-containing molecule

S. M. Udrescu ✉, S. G. Wilkins ✉, A. A. Breier, M. Athanasakis-Kaklamanakis, R. F. Garcia Ruiz ✉, M. Au, I.

+ Show authors

Nature Physics 20, 202–207 (2024) | Cite this article

Recent studies of
RaF + Na => RaF⁻



S. Udrescu



S. Wilkins

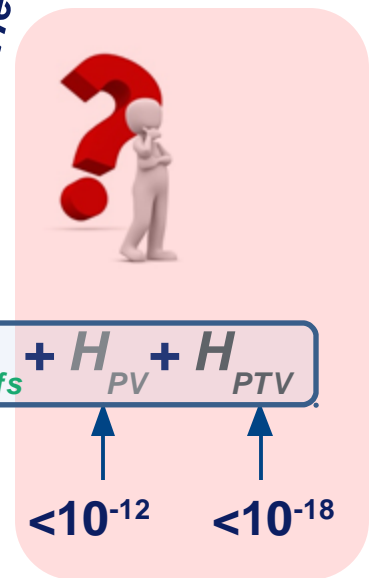


M. Athanasakis

Recent Results (RaF)



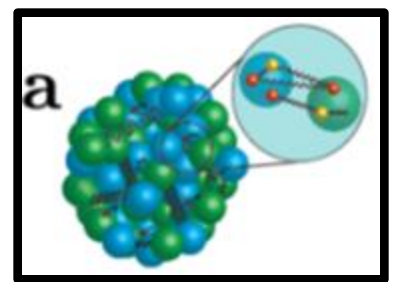
Nature 581, 396 (2020) ✓
PRL 127, 033001 (2021) ✓
Nature Phys 20, 202 (2024) ✓
 (2024) Under review ✓



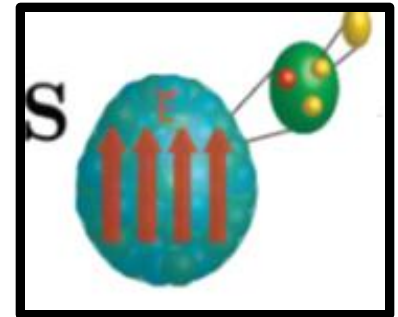
$$\Delta O \sim \frac{1}{F_{mol} \tau \sqrt{N}}$$

$$H_{mol} = H_e + H_{vib} + H_{rot} + \dots + H_{hfs} + H_{PV} + H_{PTV}$$

eV ~ 2 10⁻² 10⁻⁵ 10⁻⁸ <10⁻¹² <10⁻¹⁸

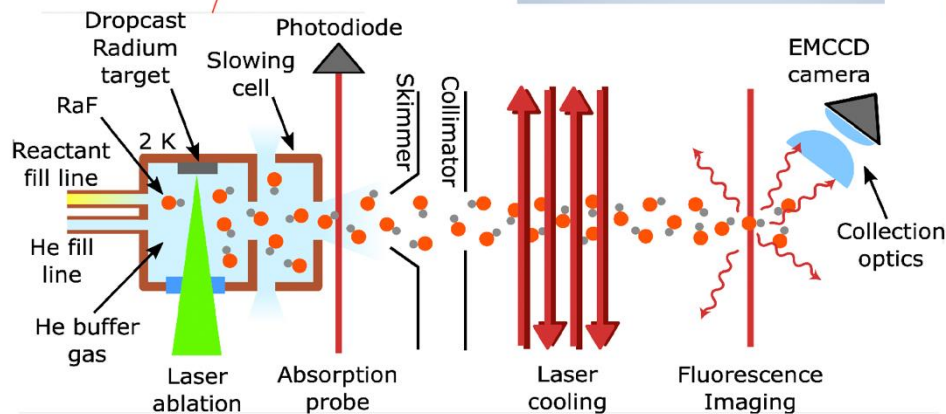
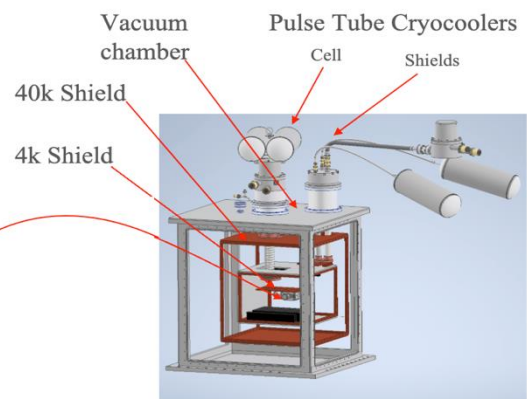
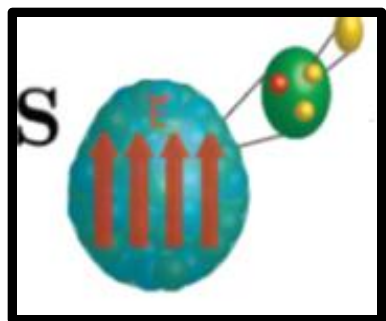
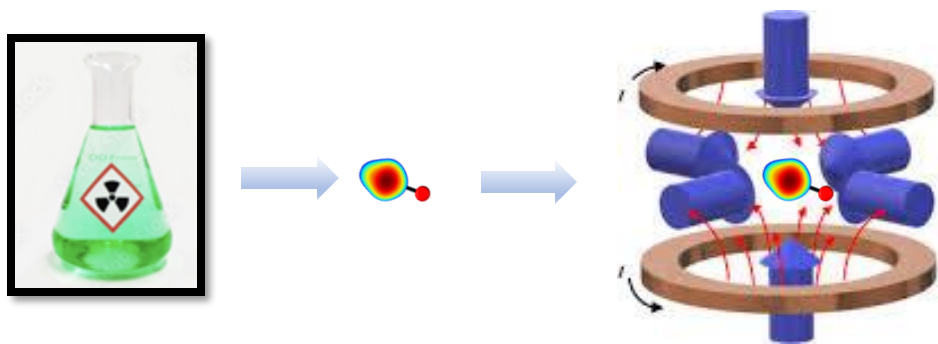


NEPTUNE Project



RaX Collaboration

P,T-violation measurements with RaX Molecules

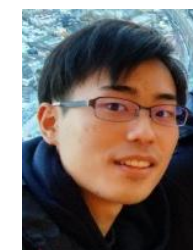


RaX Collaboration

*J. Doyle (Harvard), N. Hutzler (Caltech),
R.F. Garcia Ruiz (MIT) & FRIB*



Caltech



S. Ebadi

A. Jadbabaie

J. Munoz

M. Fulghieri

F. Shungo

K. Khusainova

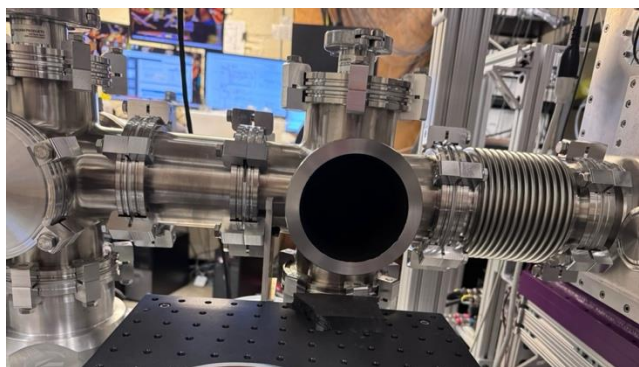
S. Munoz

RaX Collaboration

- ✓ Beam box fabrication
- ✓ Cryocooler testing
- ✓ Laser Installation
- ✓ Beamline construction



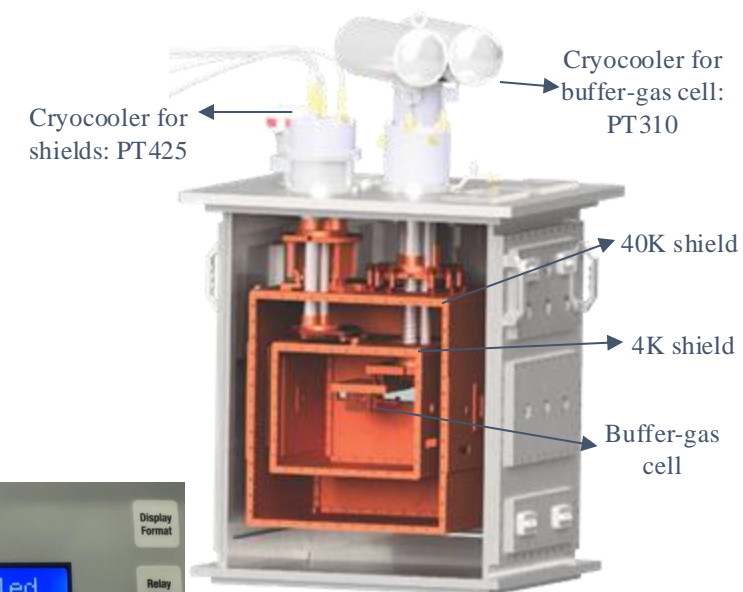
Lab @ Harvard



Blackened beamline for fluorescence collection



Ti:Sa laser installed



PT310 cryocooler:
2.2K base temp,
1W@3K, 51W@34K

Radioactive Molecules for Fundamental-Symmetry Tests

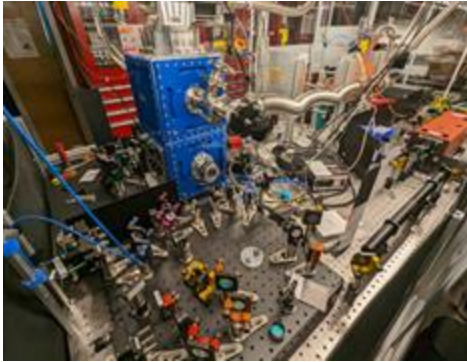
→ RaOH, RaOH⁺ and RaOCH₃⁺

[Hutzler, Jayich, ...]

[Fan et al. Phys. Rev. Lett. 126, 023002 (2021)]

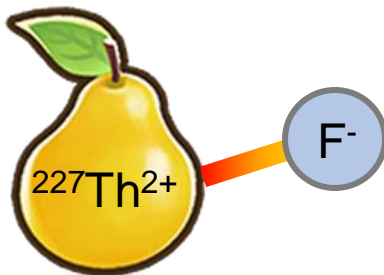
[Yu & Hutzler Phys. Rev. Lett. 126, 023003 (2021)]

Hutzler's group: RaOH @ Caltech



→ ²²⁷ThF⁺

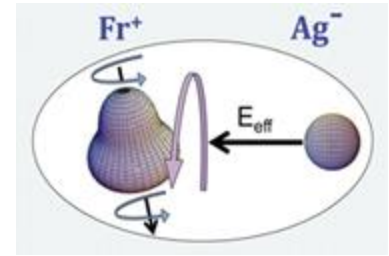
[Boon Ng, Cornell, Fang, Malbrunot ...]



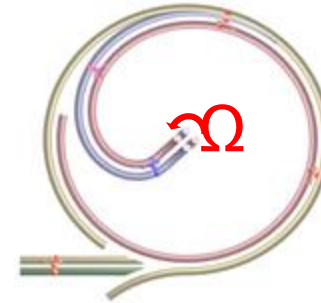
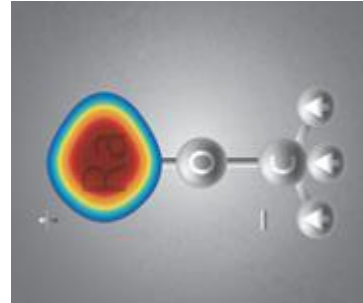
→ Assembling molecules from cold atoms [Demille]

- RaAg
- FrAg

[New J. Phys. 23, 113039 (2021)]



→ Centrifuge Deceleration



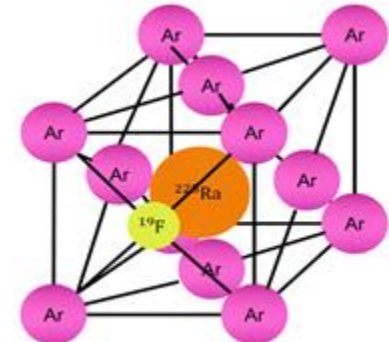
Xing Wu

RaOCH₃ @ FRIB/MSU

→ Molecules in Noble Gas Solids (York/Toronto/MSU)

[Singh, Vutha,...]

[Vutha et al. PRA 98:032513 (2018)]

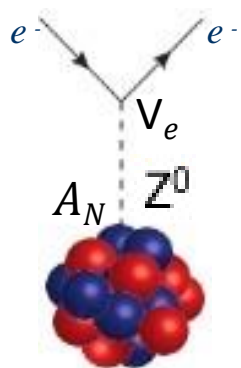
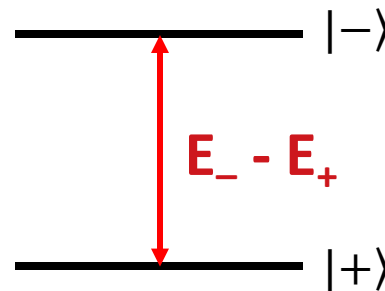


Molecules as sensitive probes of the nuclear
electroweak (EW) structure

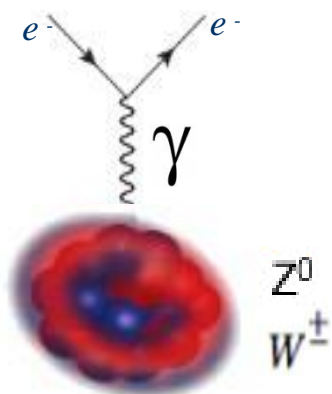
Molecules as sensitive probes of the nuclear EW structure

$$H_{PV} \sim Z^c / (E_+ - E_-)$$

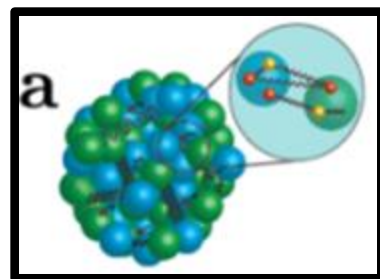
$$|+\rangle' = |+\rangle + \frac{\langle + | V_{PV} | - \rangle}{E_+ - E_-} |-\rangle$$



C_{2u}, C_{2d}



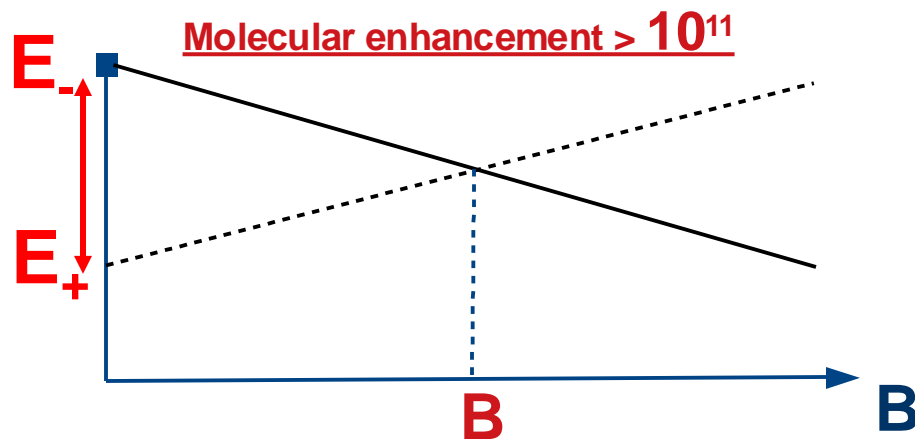
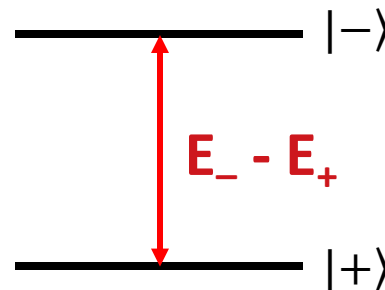
Anapole



Molecules as sensitive probes of the nuclear EW structure

$$H_{PV} \sim F(Z^c) / (E_+ - E_-)$$

$$|+\rangle' = |+\rangle + \frac{\langle + | V_{PV} | - \rangle}{E_+ - E_-} |-\rangle$$

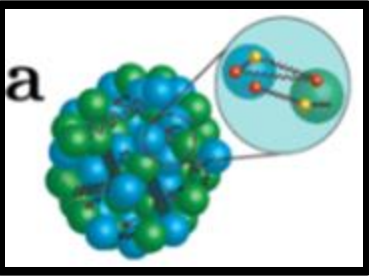


$> 10^{11}$ Enhancement sensitivity to electroweak nuclear properties!

Demille's Group [Altunas et al. Phys. Rev. Lett. 120, 142501 (2018)]

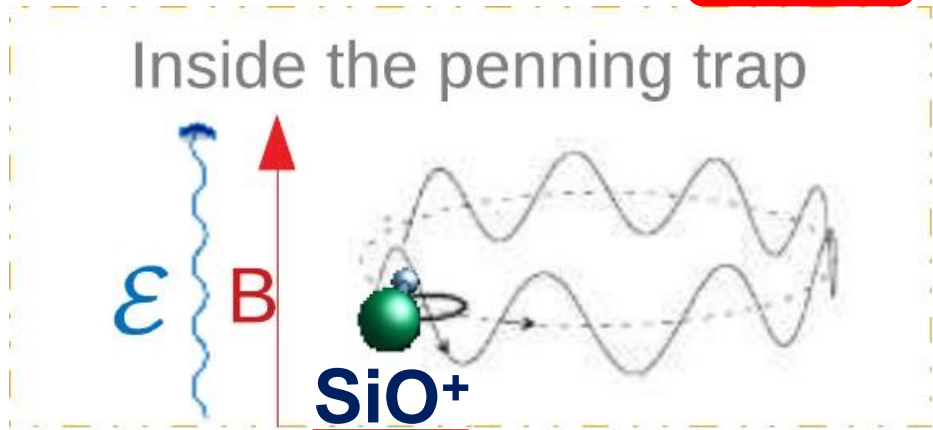
NEPTUNE – Project

Nuclear Electroweak Measurements in a Penning Trap Using Near-degenerate Energy States of Molecules



$$H_{PV} \sim F(Z^c) / (E_+ - E_-)$$

$$|+\rangle' = |+\rangle + \frac{\langle + | V_{PV} | - \rangle}{E_+ - E_-} |-\rangle$$



J. Karthein



S. Udrescu



S. Moroch



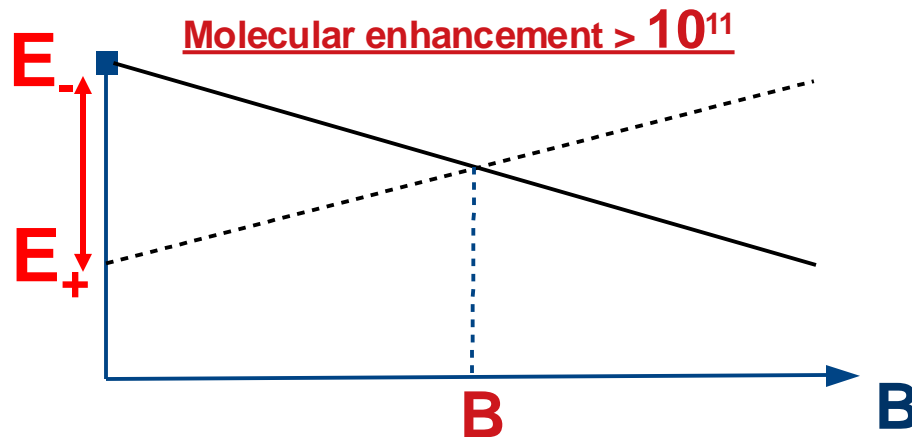
H. Kakiota

PHYSICAL REVIEW LETTERS

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Electroweak Nuclear Properties from Single Molecular Ions in a Penning Trap

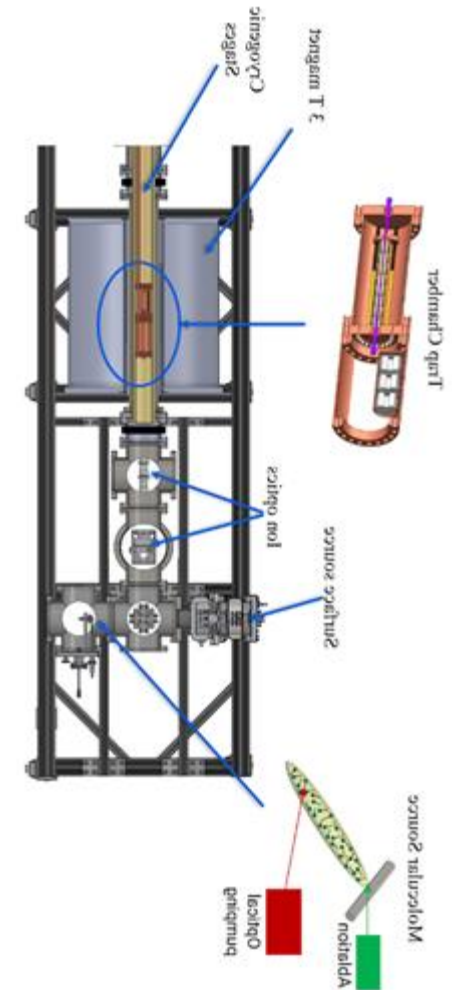
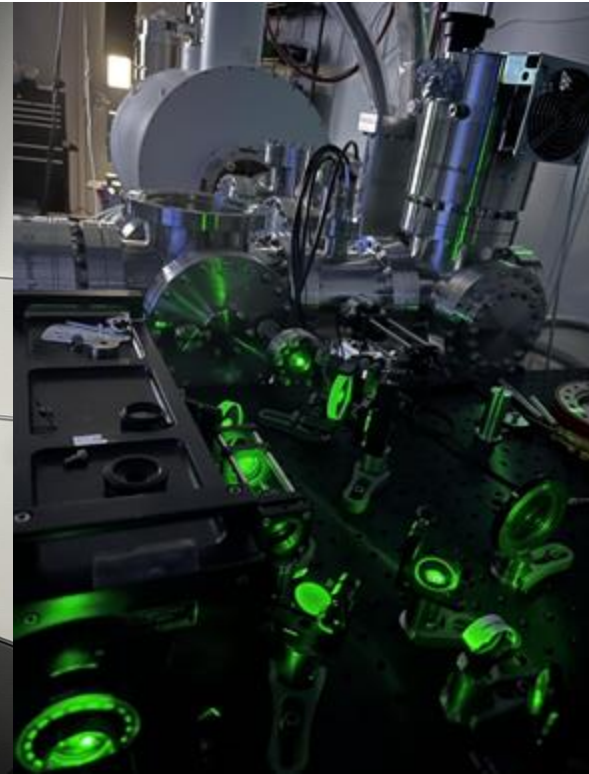
J. Karthein, S. M. Udrescu, S. B. Moroch, I. Belosevic, K. Blaum, A. Borschevsky, Y. Chamorro, D. DeMille, J. Dilling, R. F. Garcia Ruiz, N. R. Hutzler, L. F. Pašteka, and R. Ringle
 Phys. Rev. Lett. **133**, 033003 – Published 19 July 2024



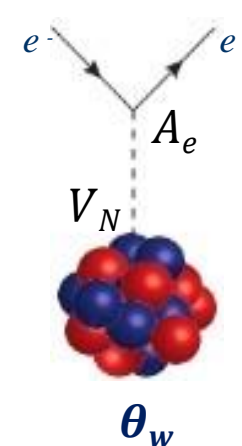
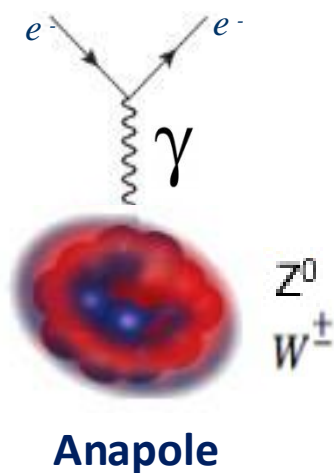
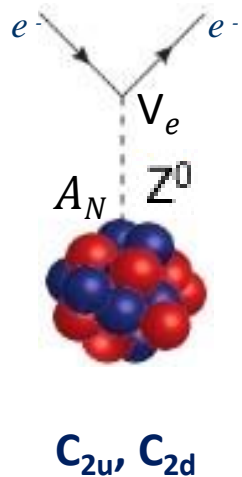
NEPTUNE – Project

Nuclear Electroweak Measurements in a Penning Trap Using Near-degenerate Energy States of Molecules

Lab
@MIT



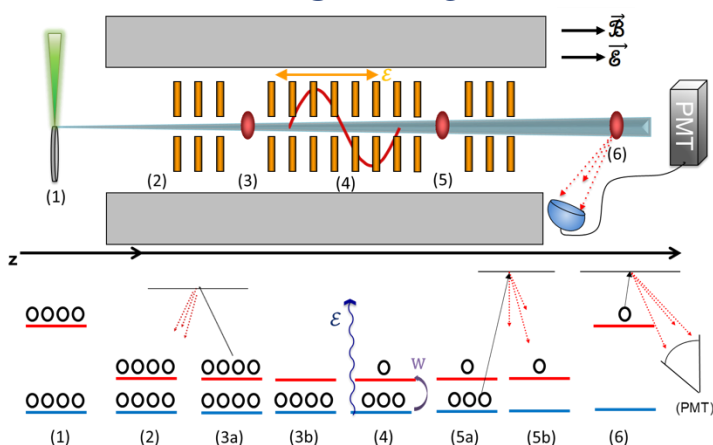
Molecules as sensitive probes of the nuclear EW structure



Fr @ TRIUMF

[Zang et al. Phys. Rev. Lett. 115, 042501 (2015)]

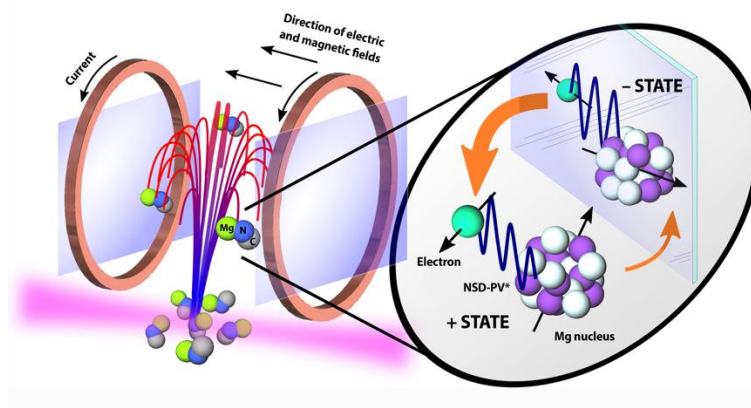
ZOMBIES



U Chicago, ANL

[Altunas et al. PRL 120, 142501 (2018)]

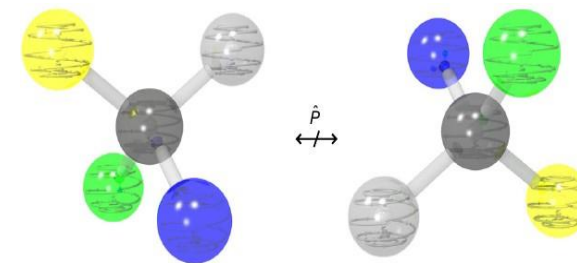
Molecular fountain



NIST

[Norrsgard et al. Comm. Phys. 2, 77 (2019)]

$$PV \sim Z^5$$



[Erez et al. arXiv:2206.03699 (2022)]

[Gaul et al. Phys. Rev. Lett. 125, 123004 (2021)]

Summary and Outlook

$$H_{mol} = H_e + H_{vib} + H_{rot} + \dots + H_{hfs} + H_{PV} + H_{PTV}$$

H_{hfs}

P,T-even

- Nuclear structure
- Nuclear matter

H_{PV}

P-violation

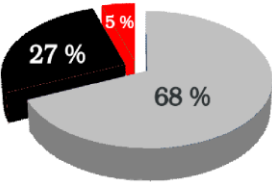
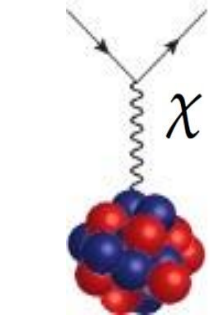
- Electro weak structure
- Precision Standard Model tests
- Dark Mater properties?
- New forces?

H_{PTV}

EDM MDM ^{223}Ra

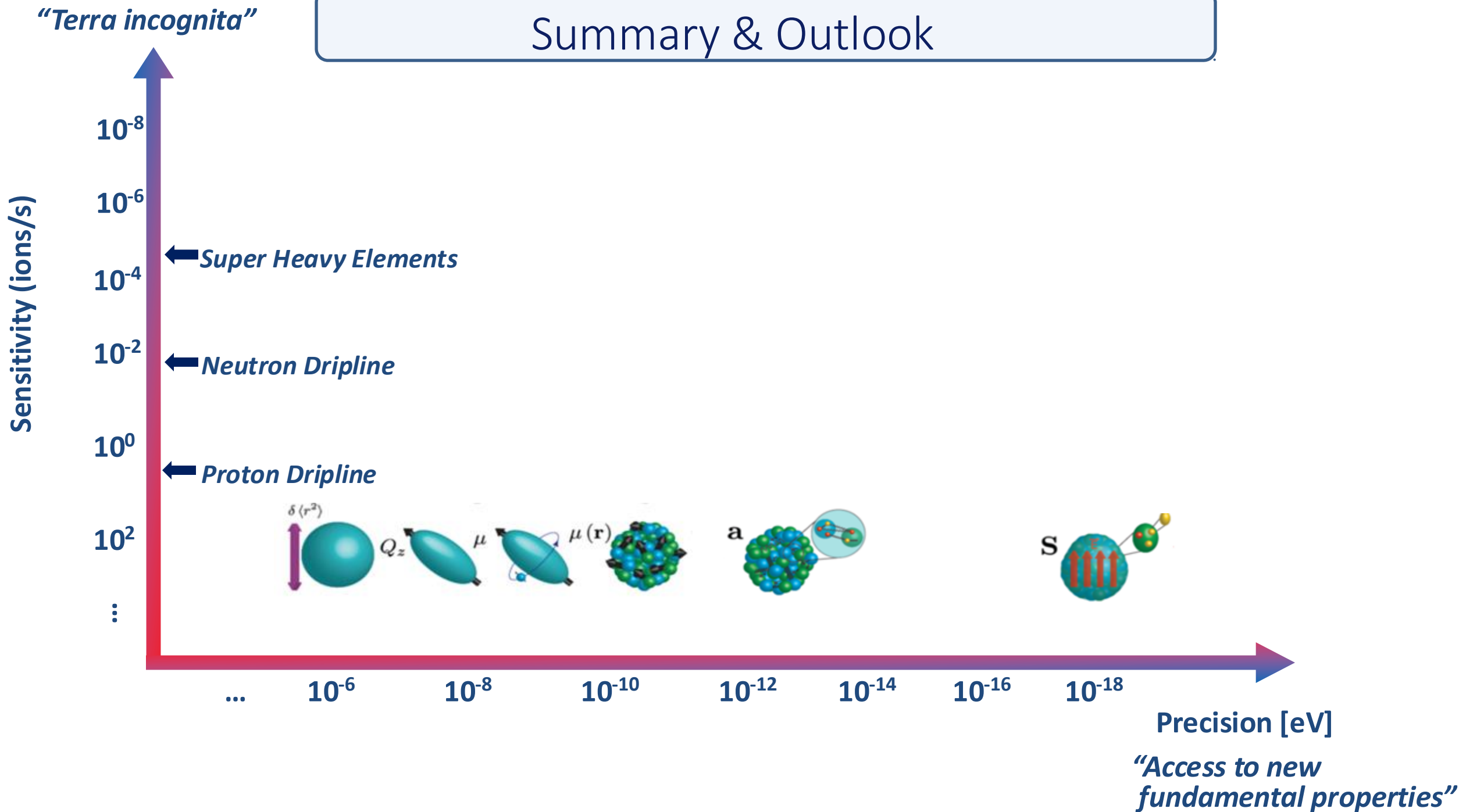
T-violation

- Matter-antimatter asymmetry
- New particles?



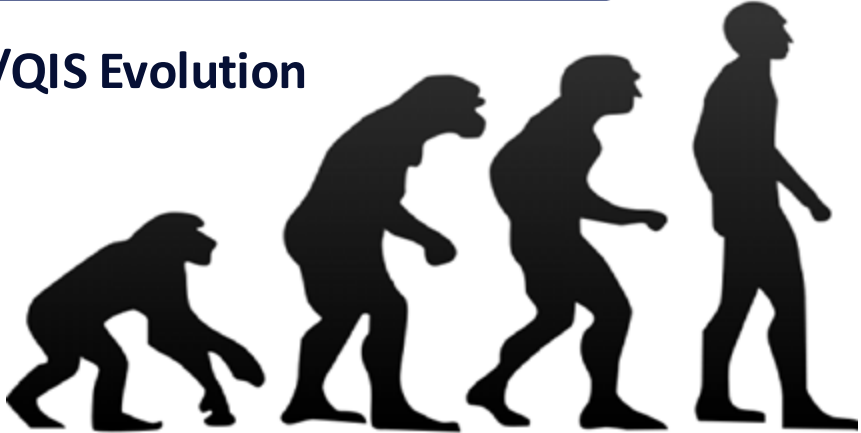
■ Ordinary Matter
■ Dark Matter
■ Dark Energy

Summary & Outlook

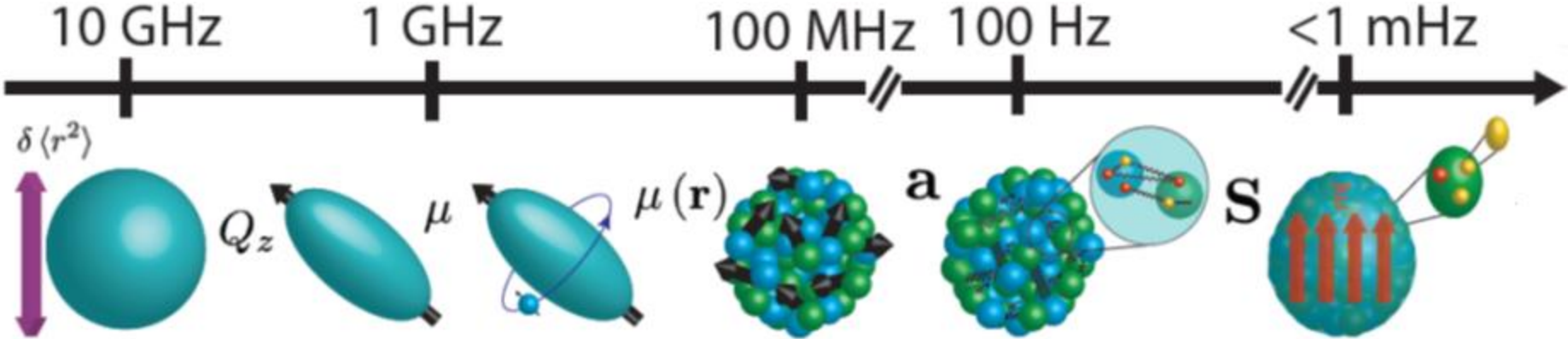


Summary and Outlook

AMO/QIS Evolution



We are here!



$$H_{mol} = H_e + H_{vib} + H_{rot} + H_{sr} + \dots + H_{hfs} + H_{PV} + H_{PTV}$$

eV ~ 2 10^{-2} 10^{-5} 10^{-6} 10^{-8} $<10^{-12}$ $<10^{-15}$

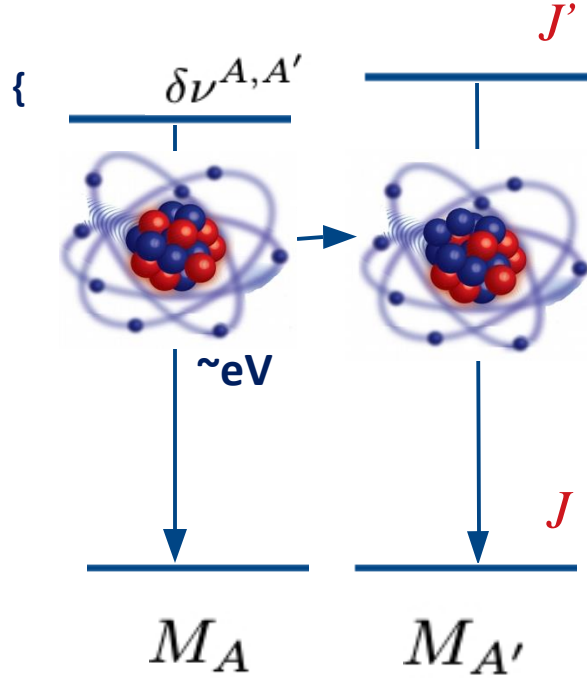
New opportunities!

- **New Facilities (FRIB, TRIUMF, ISOLDE, FAIR,....)**
- **New Era of Precision (Atomic, Molecular, Nuclear) Physics**

Isotope Shifts for Nuclear Structure and BSM Physics

Isotope Shifts for Nuclear Structure and BSM Physics

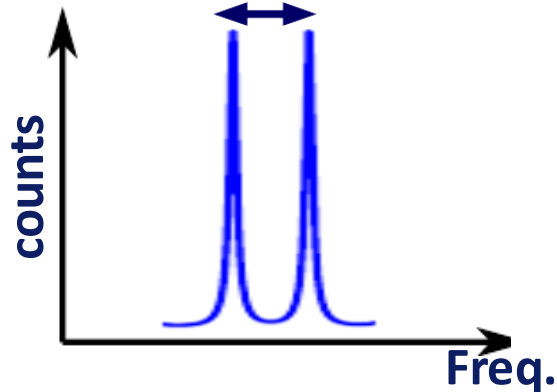
Isotope shift
 MHz $< 10^{-6}$ eV



$I = 0$

$$\sim F\delta\langle r^2 \rangle^{A,A'}$$

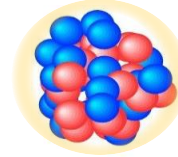
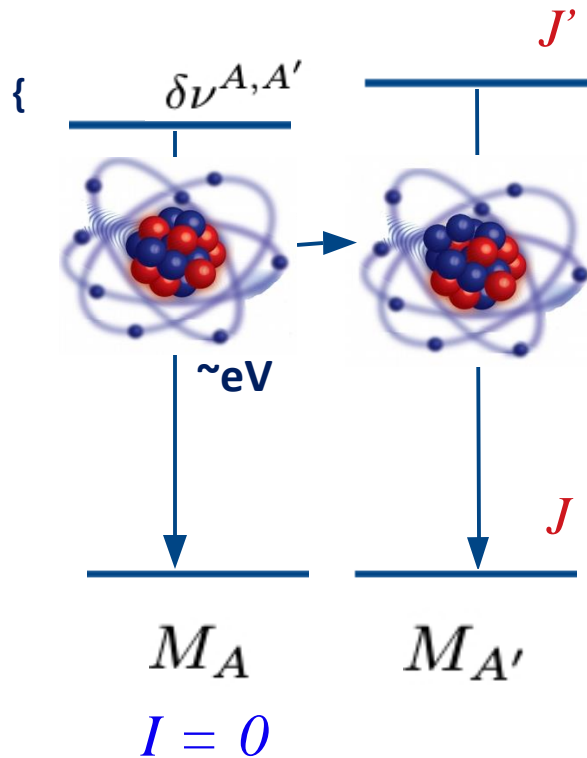
Atom/molecule
 Nuclear



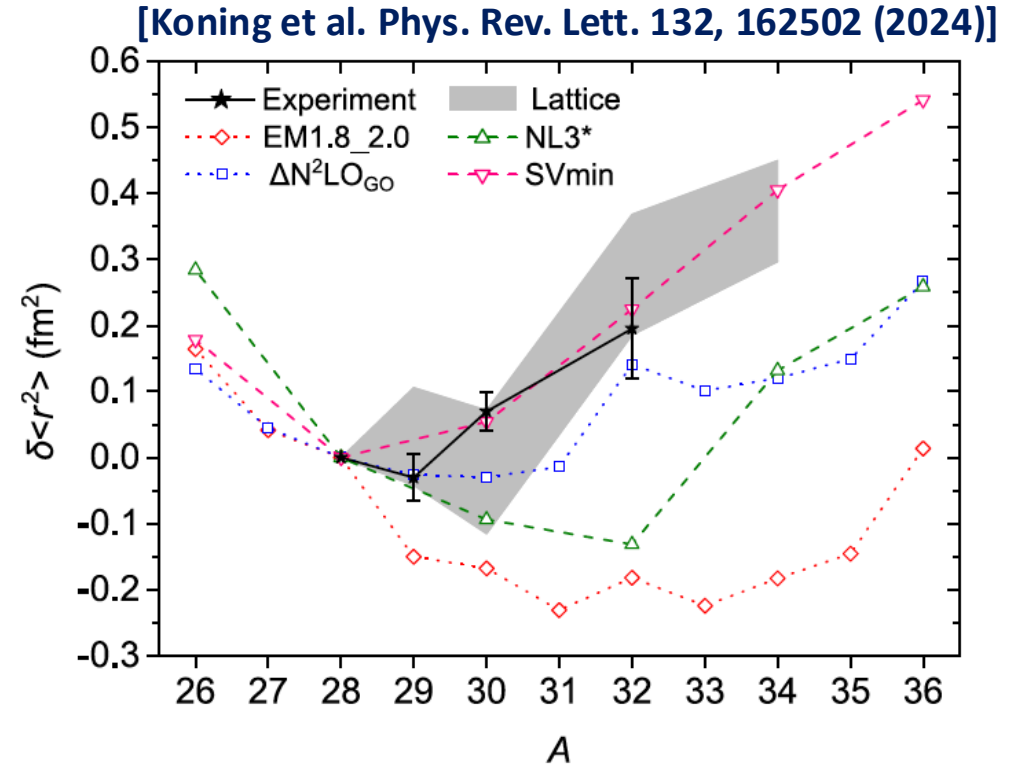
$$\delta\nu^{A,A'} = K_{MS} \frac{M_{A'} - M_A}{M_{A'} M_A} + F\delta\langle r^2 \rangle^{A,A'}$$

Isotope Shifts for Nuclear Structure and BSM Physics

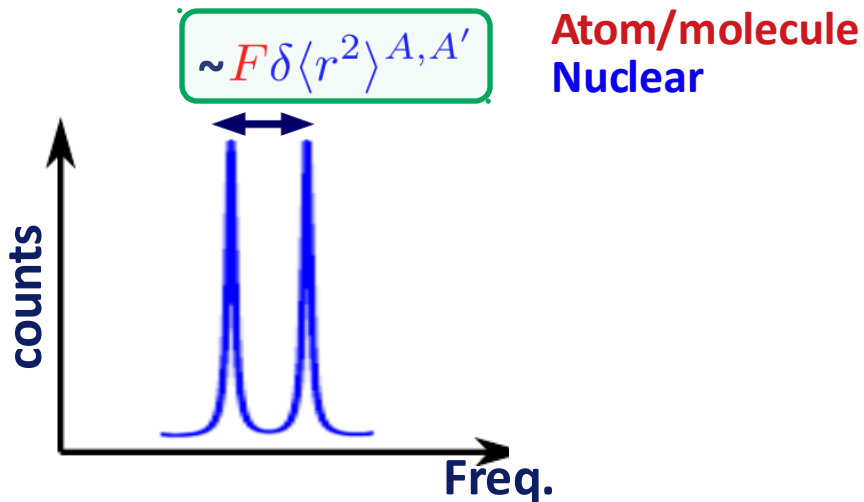
Isotope shift
MHz $< 10^{-6}$ eV



Si(Z=14)



Uncertainty ~ 1 MHz



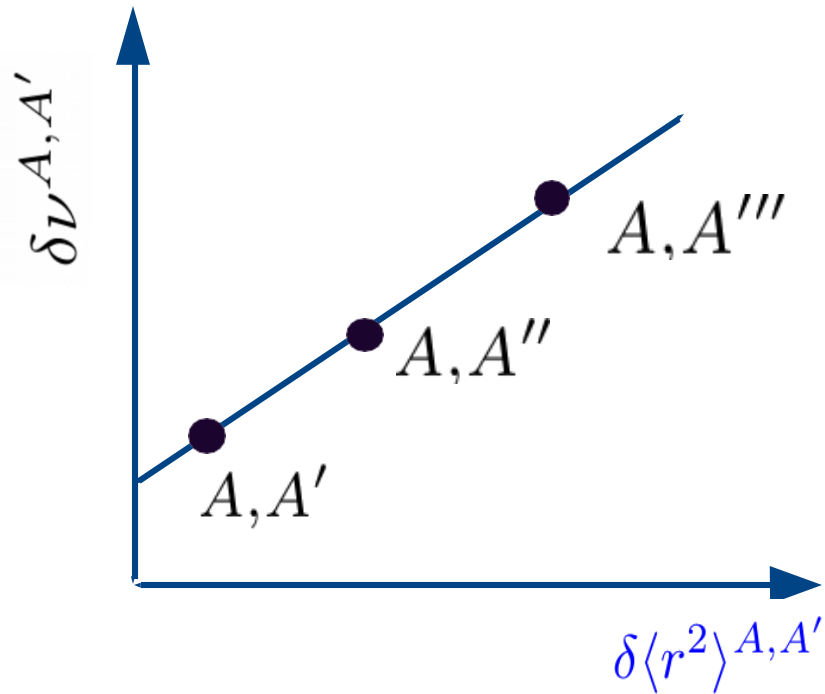
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Dean Lee's Talk
Monday at 11:30 am

Isotope Shifts for Nuclear Structure and BSM Physics

Atom/molecule
Nuclear

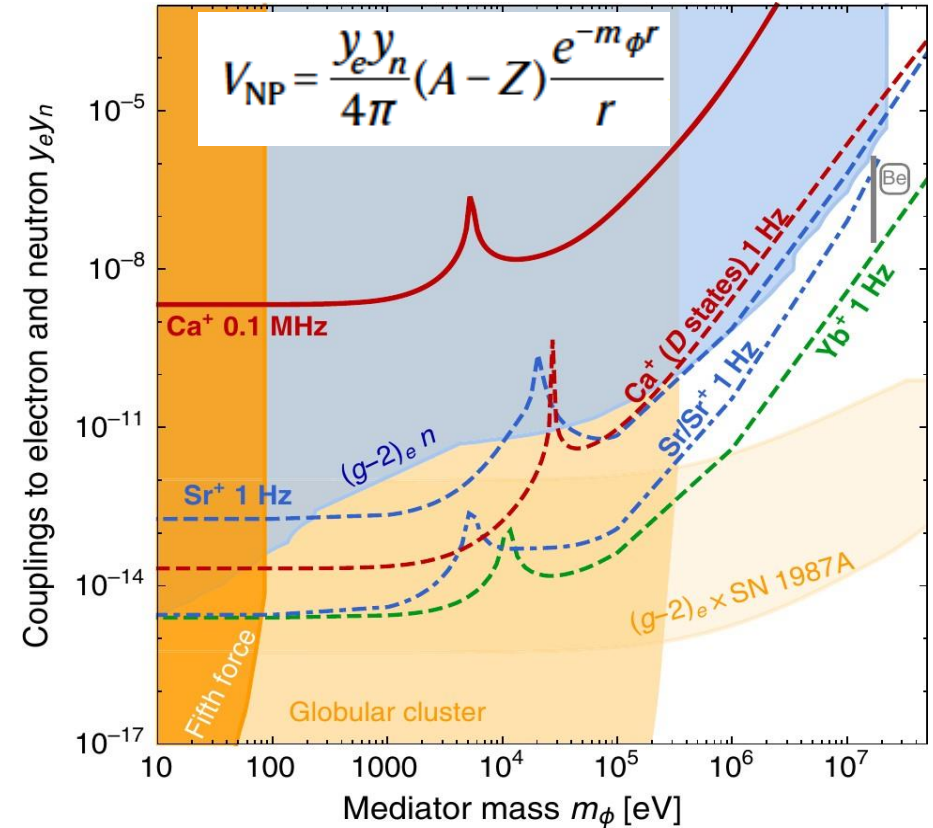
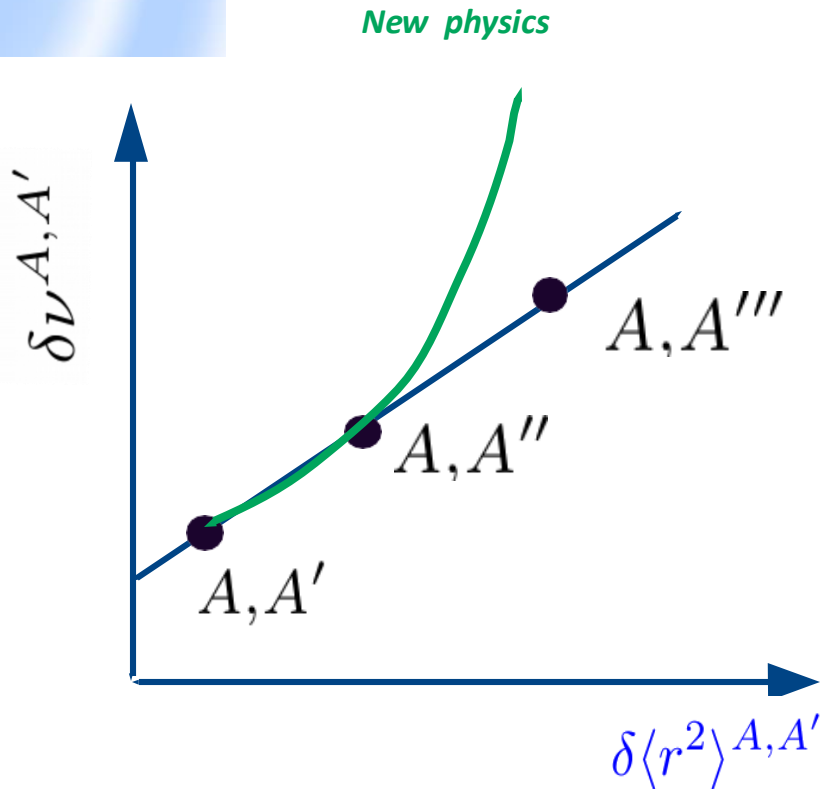
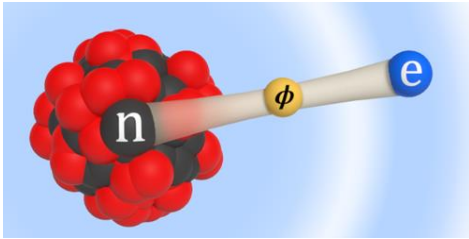
$$\delta\nu^{A,A'} = K_{MS} \frac{M_{A'} - M_A}{M_{A'} M_A} + F \delta\langle r^2 \rangle^{A,A'}$$



Isotope Shifts for Nuclear Structure and BSM Physics

Atom/molecule
Nuclear

$$\delta\nu^{A,A'} = K_{MS} \frac{M_{A'} - M_A}{M_{A'} M_A} + F \delta\langle r^2 \rangle^{A,A'} + \alpha_{NP} X_i \gamma_{AA'}$$



[Berenguet et al. Phys. Rev. Lett 120, 091801 (2018)]

Isotope Shifts for Nuclear Structure and BSM Physics

Atom/molecule
Nuclear

$$\delta\nu^{A,A'} = K_{MS} \frac{M_{A'} - M_A}{M_{A'} M_A} + F \delta\langle r^2 \rangle^{A,A'} + \alpha_{NP} X_i \gamma_{AA'} + G \delta\langle r^4 \rangle^{A,A'}$$

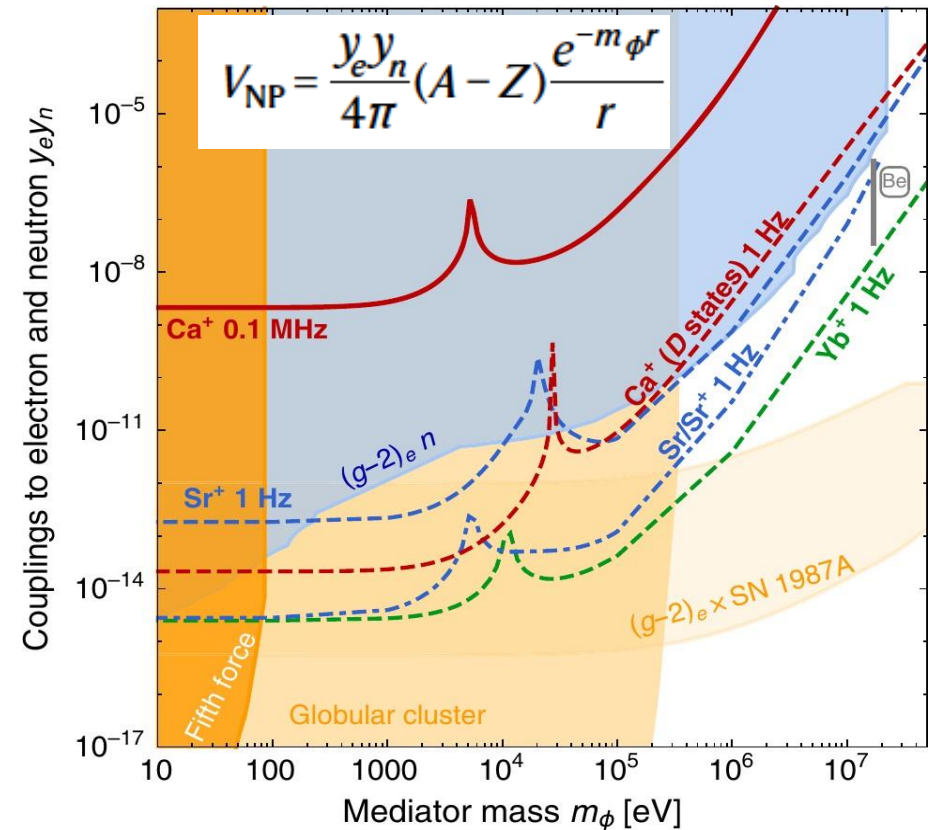
Yb⁺ → Vuletic's group at MIT

Uncertainty ~ 10 Hz (stable)

[Counts et al. Phys. Rev. Lett. 125, 123002 (2020)]
 [J. Hur et al. Phys. Rev. Lett. 128, 163201 (2022)]
 [Door et al. arXiv:2403.07792 (2024)]

$\delta\langle r^4 \rangle^{A,A'}$ → Related to the surface thickness of the nuclear density

[Reinhard et al. Phys. Rev. C 101, 021301(R) (2020)]

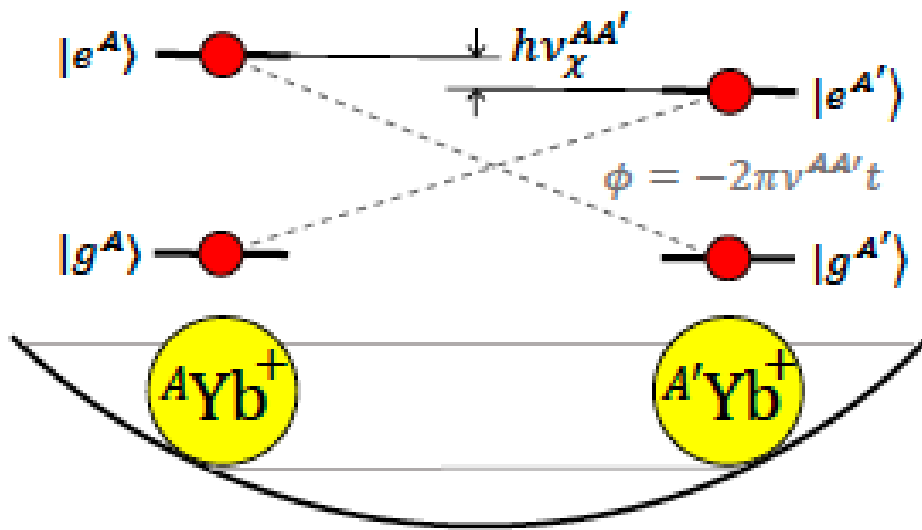


[Berengut et al. Phys. Rev. Lett 120, 091801 (2018)]

Isotope Shifts for Nuclear Structure and BSM Physics

Atom/molecule
Nuclear

$$\delta\nu^{A,A'} = K_{MS} \frac{M_{A'} - M_A}{M_{A'} M_A} + F \delta\langle r^2 \rangle^{A,A'} + \alpha_{NP} X_i \gamma_{AA'} + G \delta\langle r^4 \rangle^{A,A'}$$



Entanglement

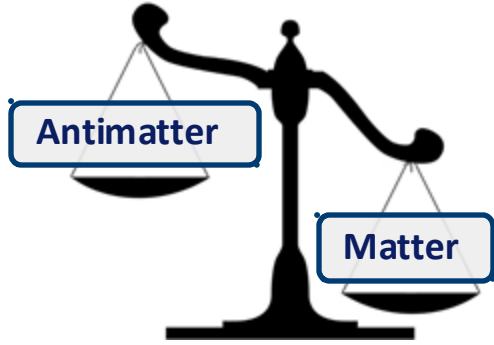
$$|\psi\rangle = \frac{1}{\sqrt{2}} (|g^A\rangle|g^{A'}\rangle + e^{-2\pi\nu^{AA'}t} |e^A\rangle|e^{A'}\rangle)$$

[T. Manovitz et al., PRL 123, 203001 (2019)]

Sr⁺: Uncertainty ~ 10 mHz (stable)

Major Open Questions in Physics and Cosmology

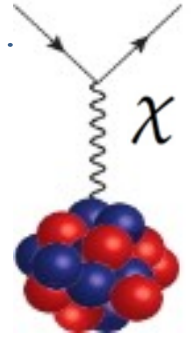
Why is there more matter?



Strong CP problem

$$\mathcal{L} = \theta \frac{1}{16\pi^2} F_{\mu\nu}^a \tilde{F}^{\mu\nu a}$$

Are there new particles?



New sources of Time-reversal (CP) violation
in nuclei can provide answers to these problems

Fundamental Symmetries

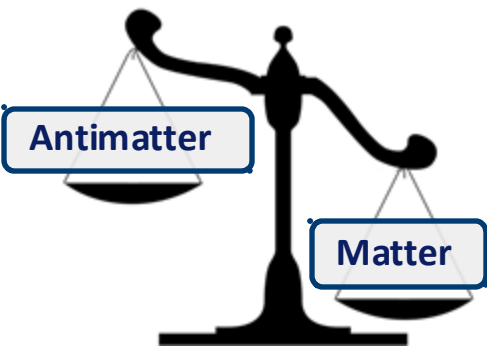
Parity (P): $r \rightarrow -r$

Time (T): $t \rightarrow -t$

Charge (C): $c \rightarrow \bar{c}$

Major Open Questions in Physics and Cosmology

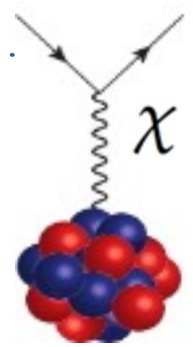
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Strong CP problem

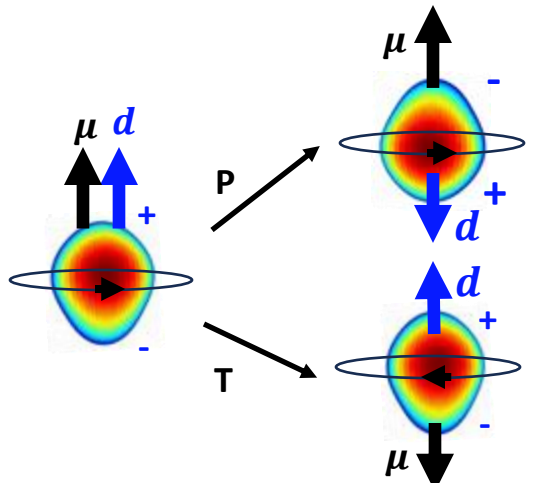
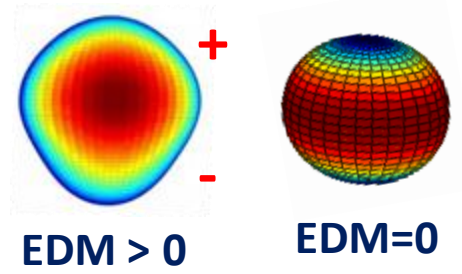
$$\mathcal{L} = \theta \frac{1}{16\pi^2} F_{\mu\nu}^a \tilde{F}^{\mu\nu a}$$

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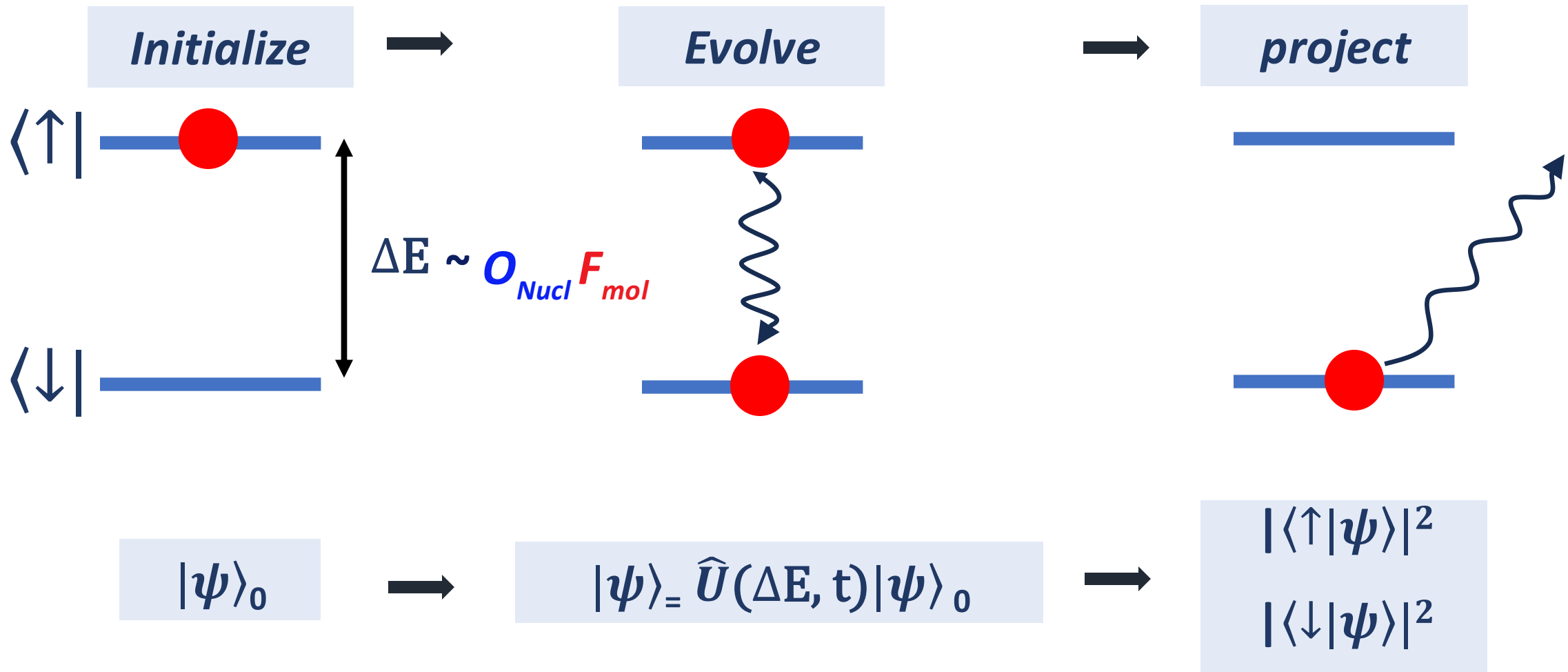
New sources of Time-reversal (CP) violation in nuclei can provide answers to these problems

Electric Dipole Moment (EDM) \rightleftarrows Time-reversal violation



Fundamental Symmetries
 Parity (P): $r \rightarrow -r$
 Time (T): $t \rightarrow -t$
 Charge (C): $c \rightarrow \bar{c}$

Molecules as “quantum sensors”

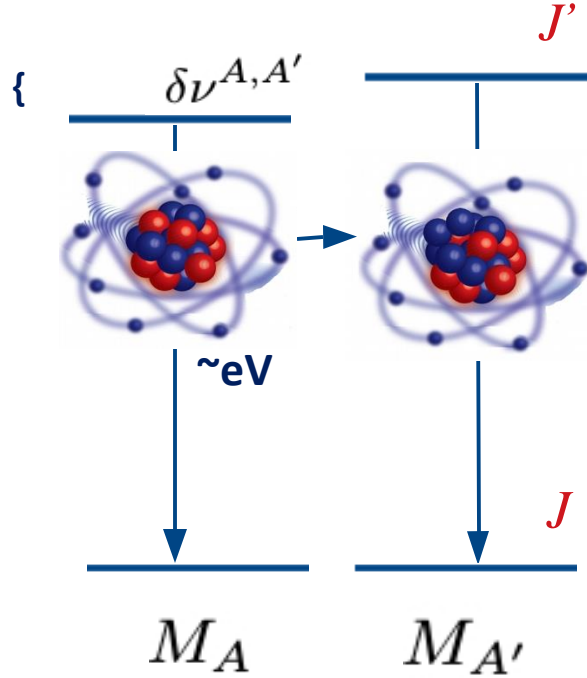


Recent reviews:

- “Searches for new sources of CP violation using molecules as quantum sensors”. Hutzler et al. arXiv:2010.08709 (2020)
- “Opportunities for fundamental physics research with radioactive molecules”. Arrowsmith-Kron et al. Rep. Prog. Phys. 87 084301 (2024)

Isotope Shifts for Nuclear Structure and BSM Physics

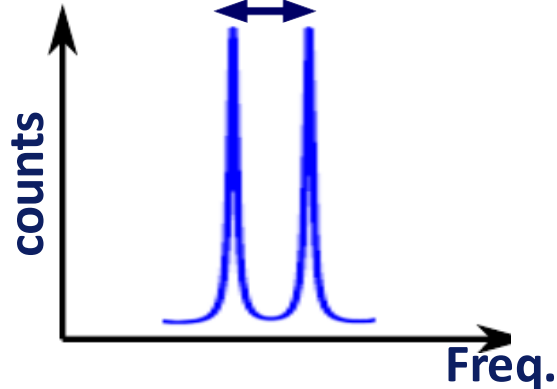
Isotope shift
MHz $< 10^{-6}$ eV



$I = 0$

$$\sim F\delta\langle r^2 \rangle^{A,A'}$$

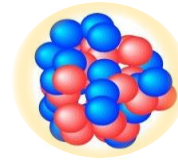
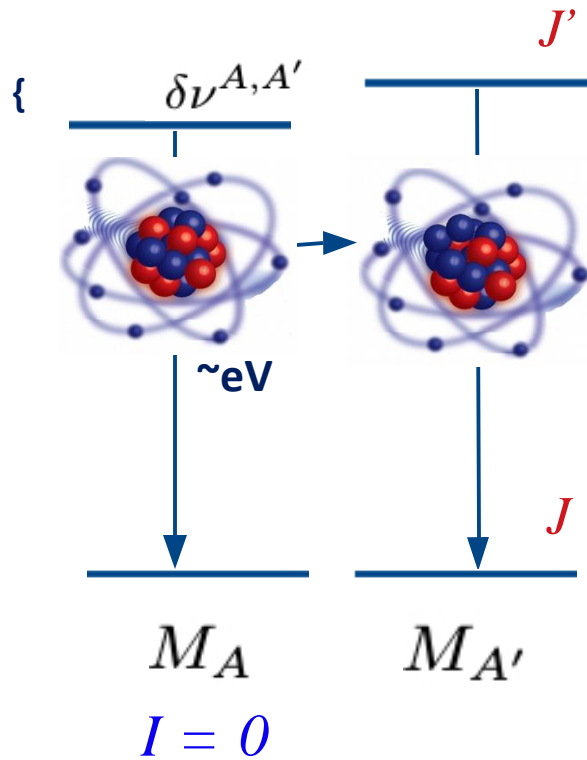
Atom/molecule
Nuclear



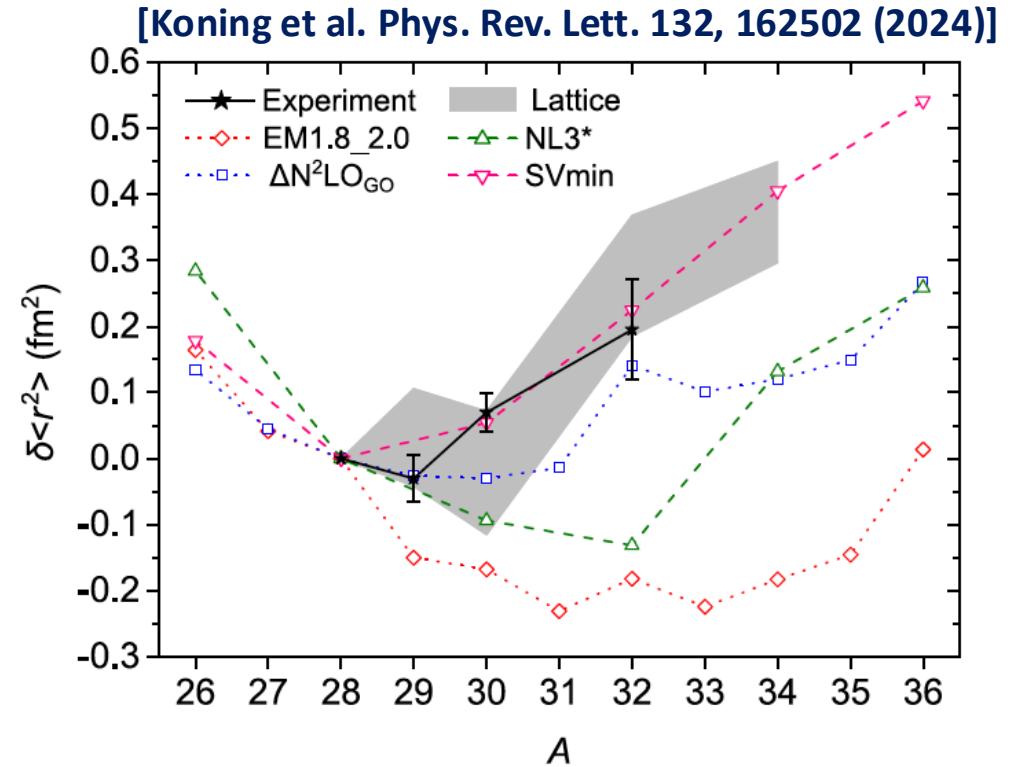
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Isotope Shifts for Nuclear Structure and BSM Physics

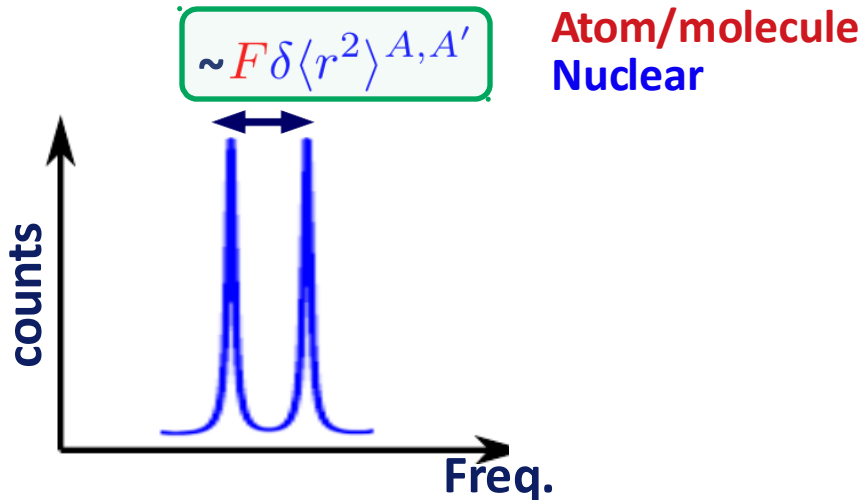
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Uncertainty ~ 1 MHz



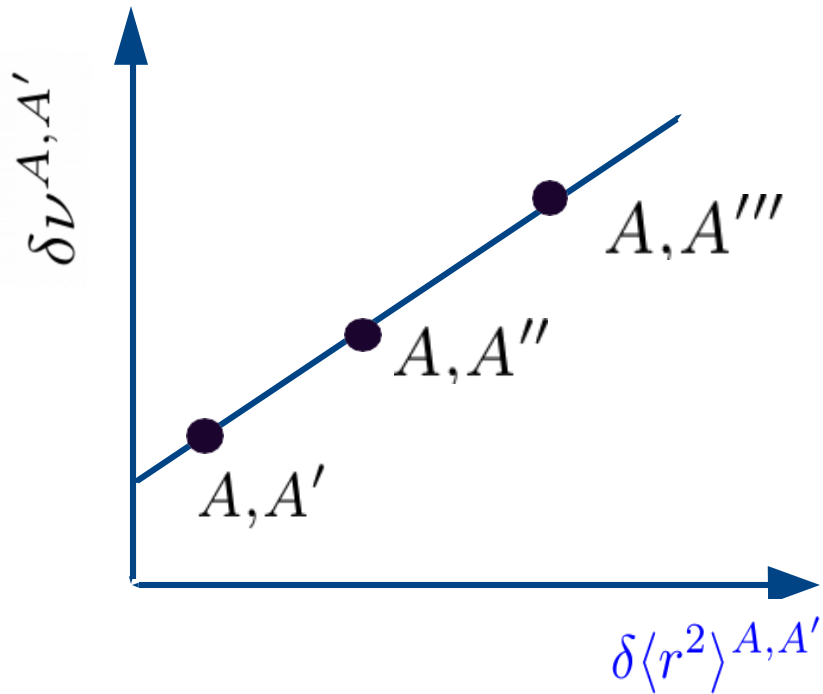
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Dean Lee's Talk
Monday at 11:30 am

Isotope Shifts for Nuclear Structure and BSM Physics

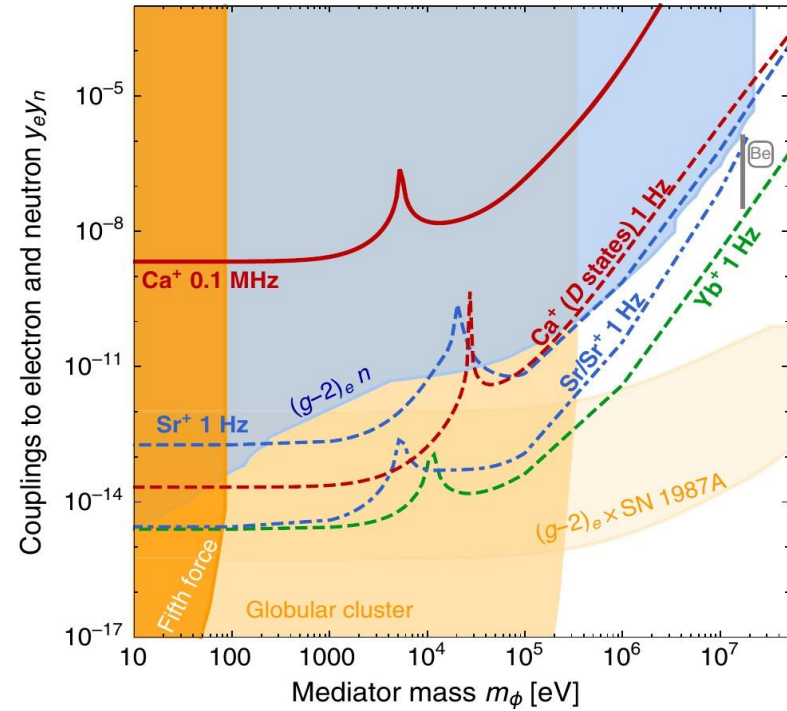
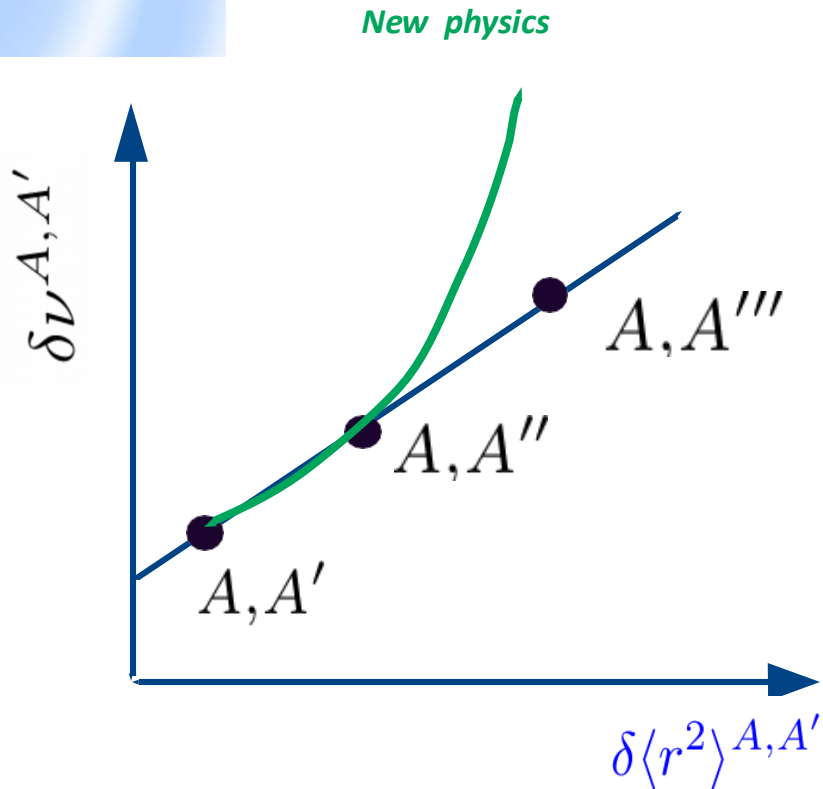
Atom/molecule
Nuclear

$$\delta\nu^{A,A'} = K_{MS} \frac{M_{A'} - M_A}{M_{A'} M_A} + F \delta\langle r^2 \rangle^{A,A'}$$



Atom/molecule
Nuclear

$$\delta V^{A,A'} = K_{MS} \frac{M_{A'} - M_A}{M_{A'} M_A} + F \delta \langle r^2 \rangle^{A,A'} + \alpha_{NP} X_i \gamma_{AA'}$$



[Berenguet et al. Phys. Rev. Lett 120, 091801 (2018)]

Isotope Shifts for Nuclear Structure and BSM Physics

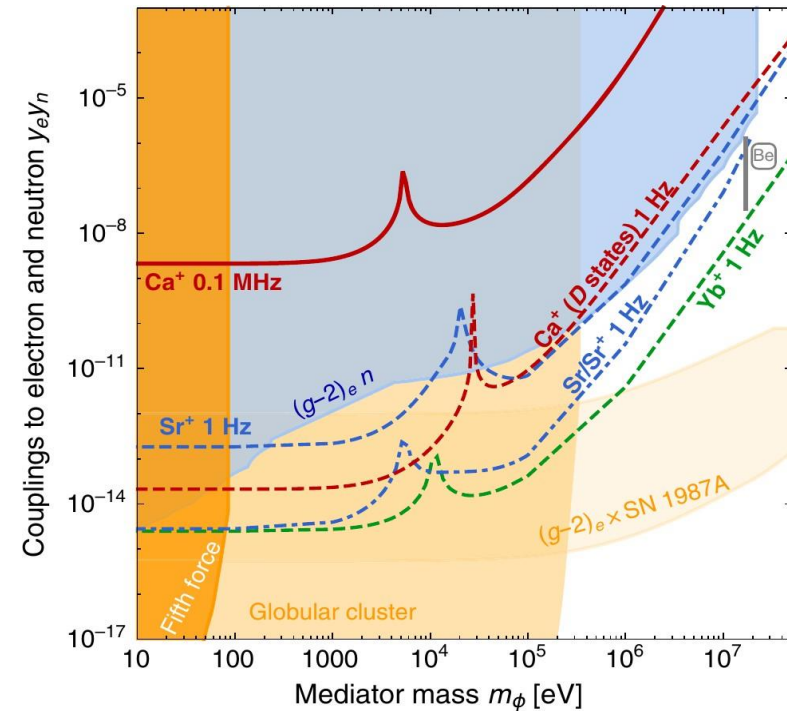
Atom/molecule
Nuclear

$$\delta\nu^{A,A'} = K_{MS} \frac{M_{A'} - M_A}{M_{A'} M_A} + F \delta\langle r^2 \rangle^{A,A'} + \alpha_{NP} X_i \gamma_{AA'} + G \delta\langle r^4 \rangle^{A,A'}$$

Yb^+ → Vuletic's group at MIT

Uncertainty ~ 10 Hz (stable)

[Counts et al. Phys. Rev. Lett. 125, 123002 (2020)]
[J. Hur et al. Phys. Rev. Lett. 128, 163201 (2022)]

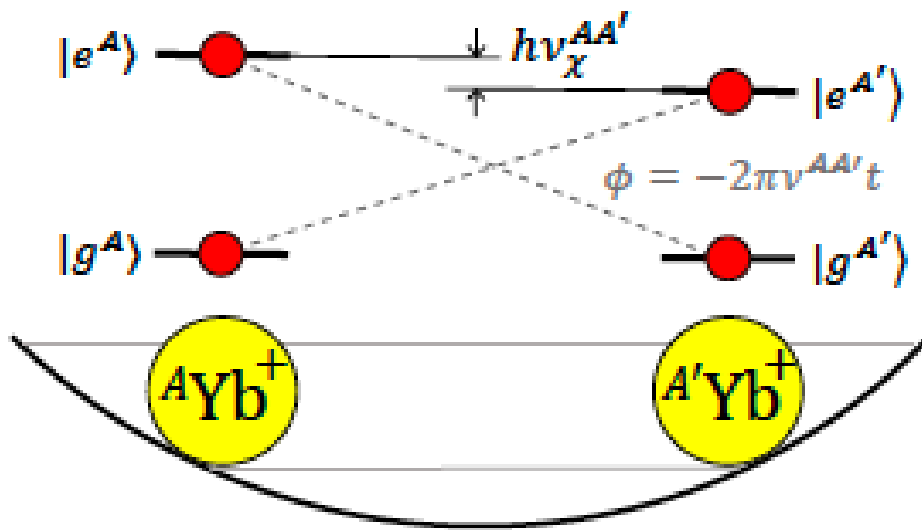


[Berengut et al. Phys. Rev. Lett 120, 091801 (2018)]

Isotope Shifts for Nuclear Structure and BSM Physics

Atom/molecule
Nuclear

$$\delta\nu^{A,A'} = K_{MS} \frac{M_{A'} - M_A}{M_{A'} M_A} + F \delta\langle r^2 \rangle^{A,A'} + \alpha_{NP} X_i \gamma_{AA'} + G \delta\langle r^4 \rangle^{A,A'}$$



Entanglement

$$|\psi\rangle = \frac{1}{\sqrt{2}} (|g^A\rangle|g^{A'}\rangle + e^{-2\pi\nu^{AA'}t} |e^A\rangle|e^{A'}\rangle)$$

[T. Manovitz et al., PRL 123, 203001 (2019)]

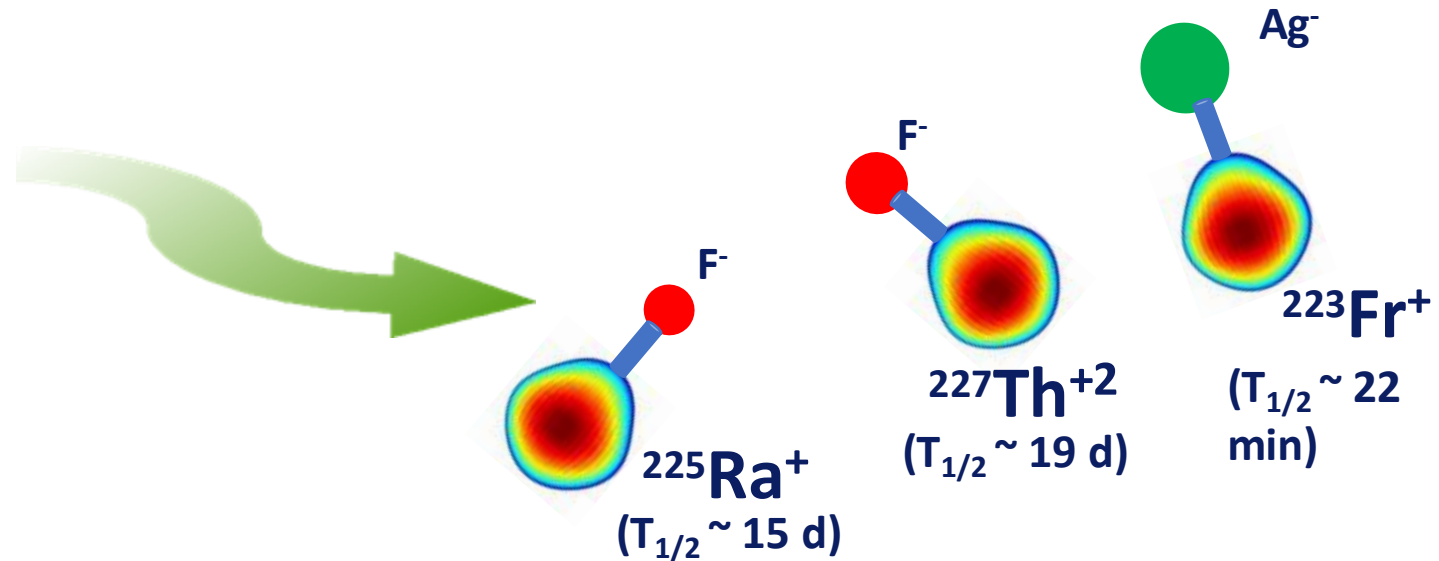
Sr⁺: Uncertainty ~ 10 mHz (stable)

Buckup Slides

Probing Symmetry-Violating Nuclear Properties with Molecules

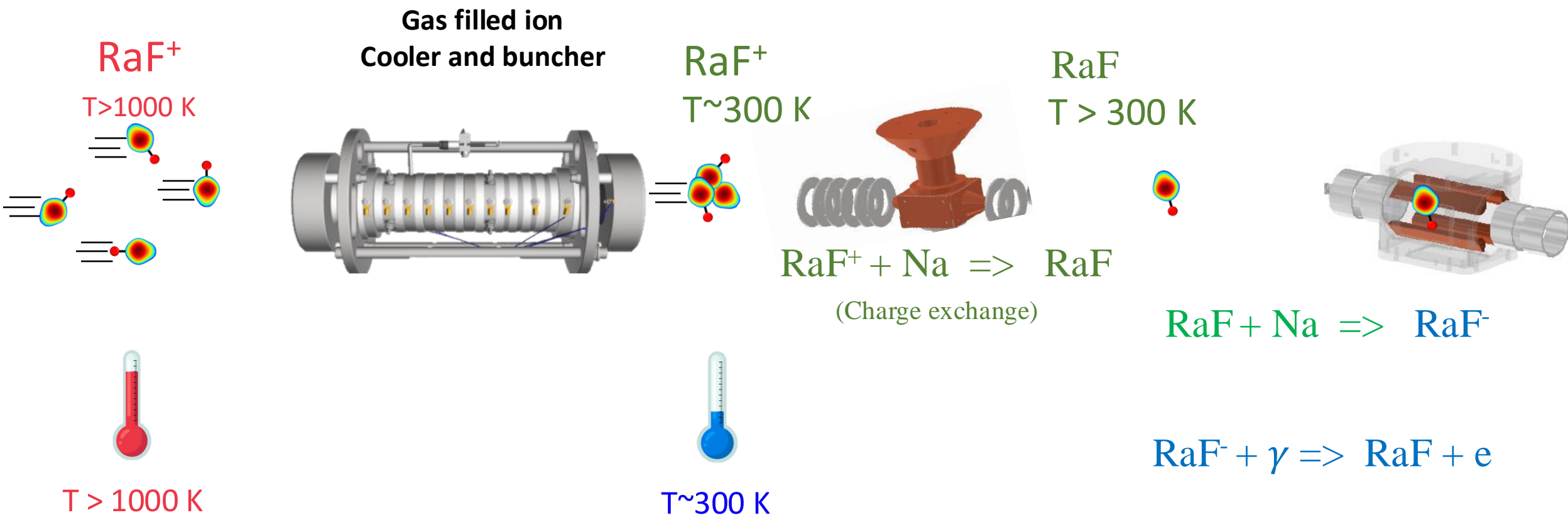


Figure modified from <https://sphereofinfluence360.com/>



Radioactive molecule production

$$\delta H_{PTV} \propto \frac{1}{\tau \sqrt{\dot{N} T}}$$



RaF⁻ Experiment @ CRIS-CERN

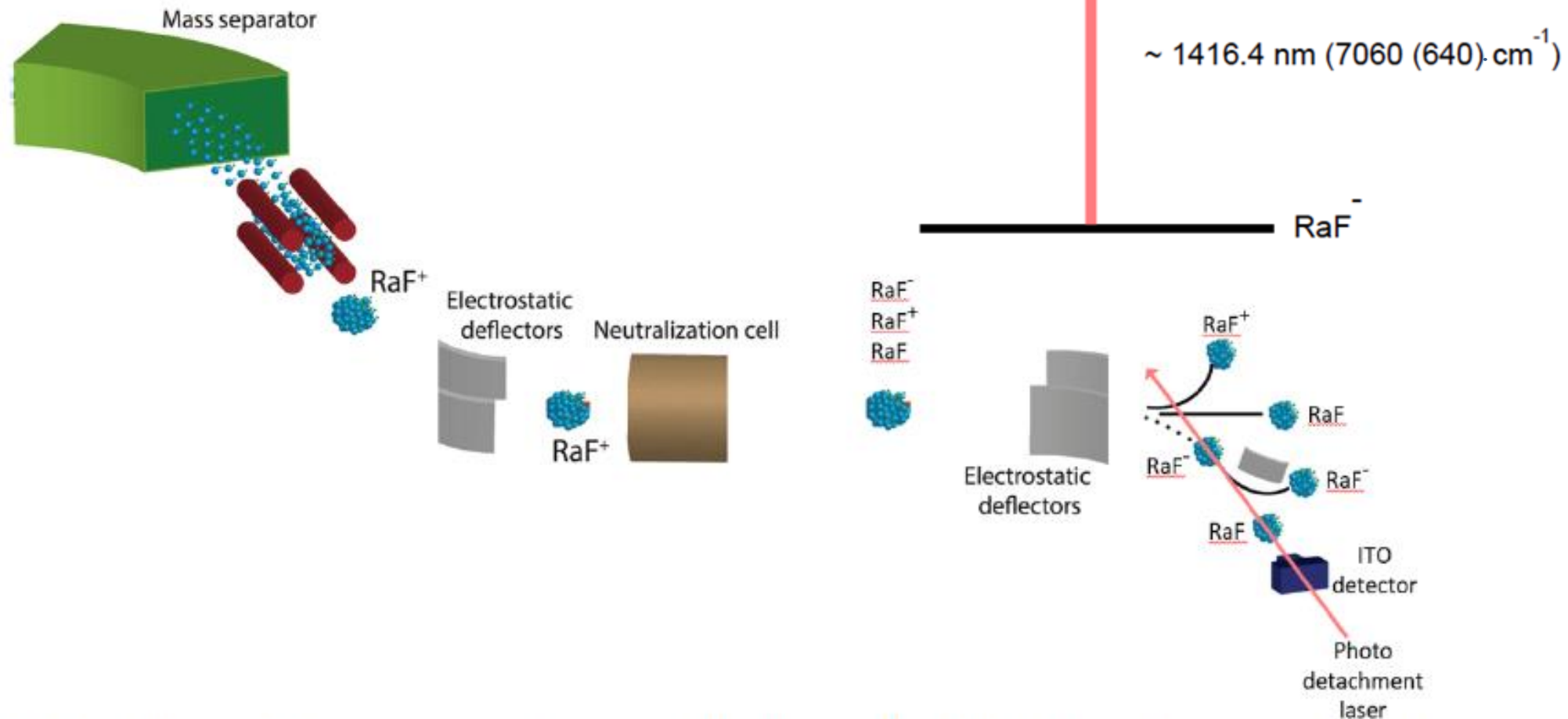


Fig 2. Schematic for anion production and colinear photodetachment measurements.

RaF⁻ Experiment @ CRIS-CERN

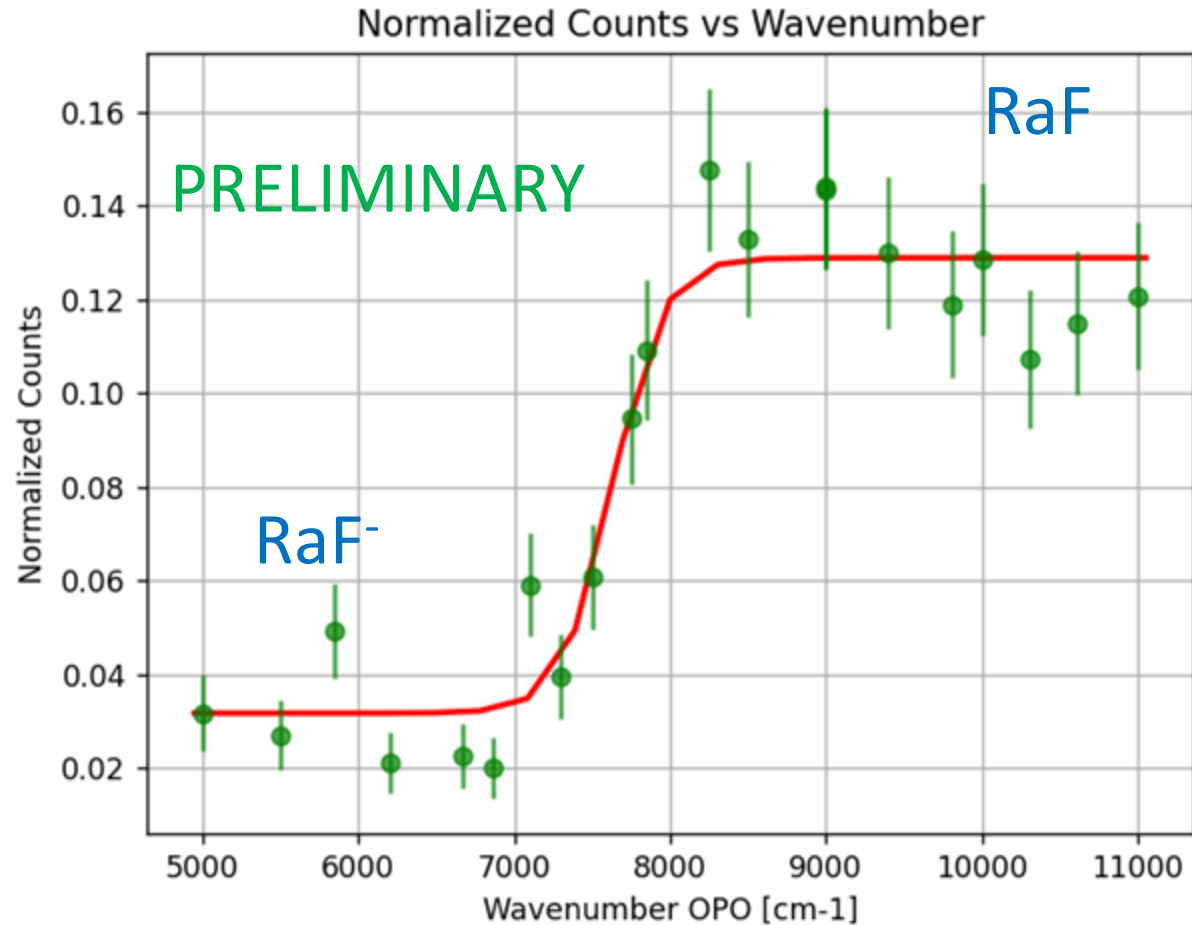
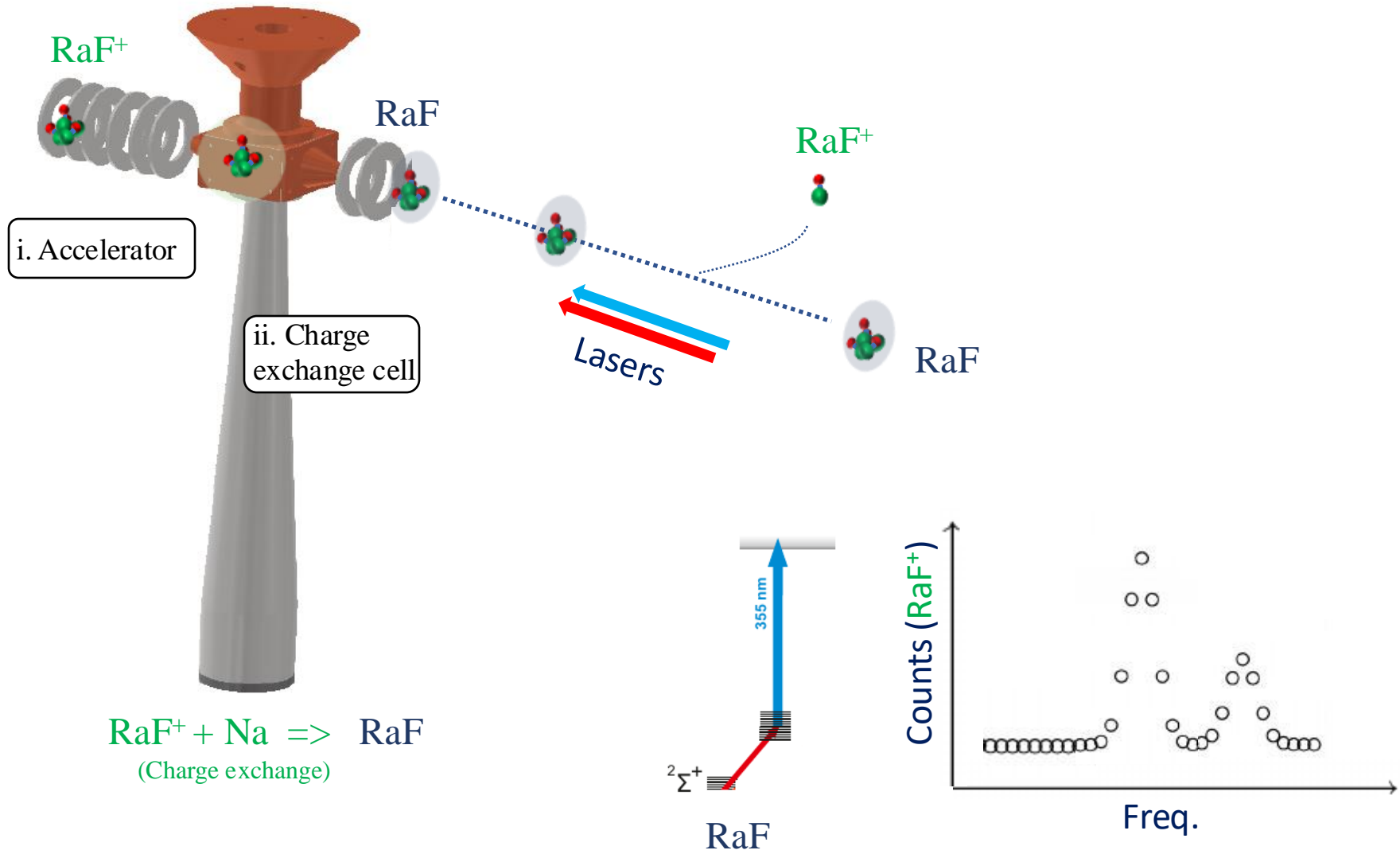


Table I. Dissociation wavenumbers \tilde{D}_e , adiabatic photoelectron detachment wavenumbers (ADW), equilibrium bond length r_e and harmonic vibrational wavenumber $\tilde{\omega}_e$ of MF⁻ computed at the level of RECP-CCSD(T)

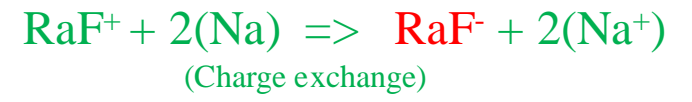
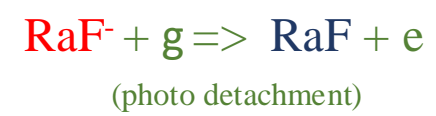
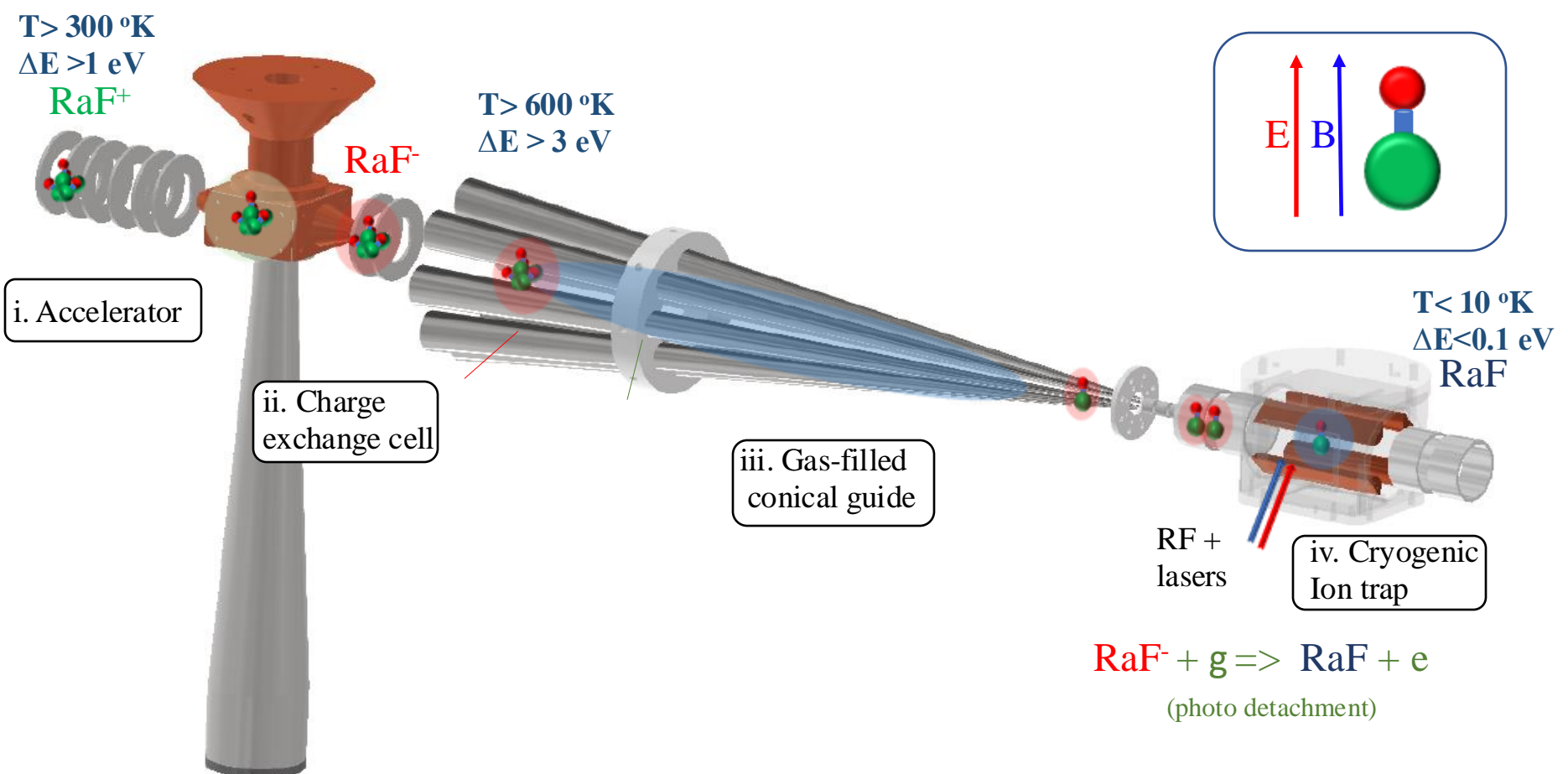
Molecule	$\tilde{D}_e/\text{cm}^{-1}$	ADW/cm ⁻¹	$r_e/\text{\AA}$	$\tilde{\omega}_e/\text{cm}^{-1}$
BeF ⁻	28 700	8590	1.42	1060
MgF ⁻	21 600	10 800	1.82	577
CaF ⁻	26 200	8300	2.01	516
SrF ⁻	25 000	7950	2.15	434
BaF ⁻	26 800	6770	2.27	439
RaF ⁻	23 600	7210	2.36	401

Gaul, Garcia Ruiz, Berger. arXiv:2403.09320 (2024)

Symmetry-violating measurements with RaF

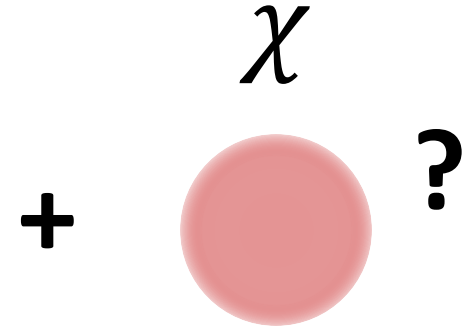


Symmetry-violating measurements with RaF



What are the fundamental particles and forces of nature?

three generations of matter (fermions)			interactions / force carriers (bosons)		
	I	II	III		
mass	$\approx 2.16 \text{ MeV}/c^2$	$\approx 1.273 \text{ GeV}/c^2$	$\approx 172.57 \text{ GeV}/c^2$	0	$\approx 125.2 \text{ GeV}/c^2$
charge	$\frac{2}{3}$	$\frac{2}{3}$	$\frac{2}{3}$	0	0
spin	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	1	0
QUARKS	u up	c charm	t top	g gluon	H higgs
	d down	s strange	b bottom	γ photon	
LEPTONS	e electron	μ muon	τ tau	Z Z boson	SCALAR BOSONS
	ν_e electron neutrino	ν_μ muon neutrino	ν_τ tau neutrino	W W boson	
	$\approx 0.511 \text{ MeV}/c^2$	$\approx 105.66 \text{ MeV}/c^2$	$\approx 1.77693 \text{ GeV}/c^2$	$\approx 91.188 \text{ GeV}/c^2$	
	-1	-1	-1	0	
	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	1	
	$< 0.8 \text{ eV}/c^2$	$< 0.17 \text{ MeV}/c^2$	$< 18.2 \text{ MeV}/c^2$	$\approx 80.3692 \text{ GeV}/c^2$	
	0	0	0	± 1	
	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	1	



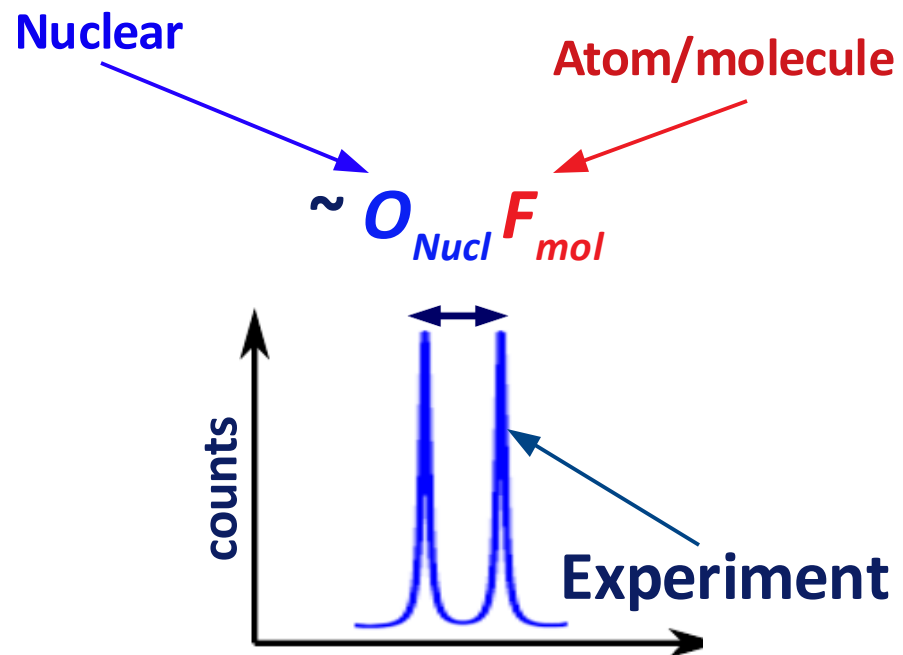
Why radioactive molecules?

$$H_{mol} = H_e + H_{vib} + H_{rot} + \dots + H_{hfs} + H_{PV} + H_{PTV}$$

$\sim O_{Nucl} F_{mol}$

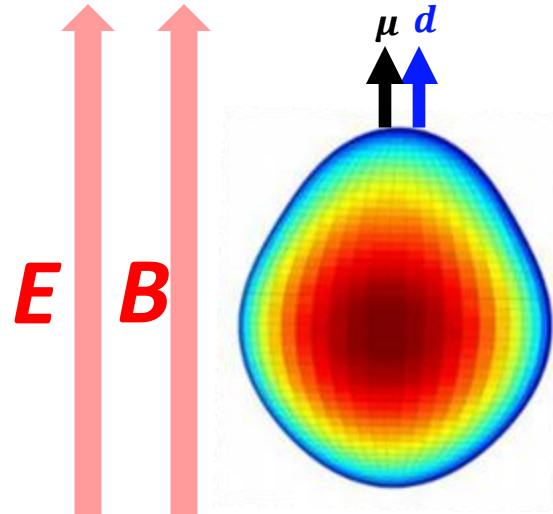
eV ~ 2 10^{-2} 10^{-5} 10^{-8} $<10^{-12}$ $<10^{-18}$

$$H_{mol} - H'_{mol} \sim O_{Nucl} F_{mol} \quad \rightarrow \text{Direct measurements!}$$



Electromagnetic properties of a charge distribution

(Produced by
the electrons)



Transient fields in reaction
 $B \sim 10^3$ Tesla
 $\nabla E \sim 10^{21}$ V/cm²

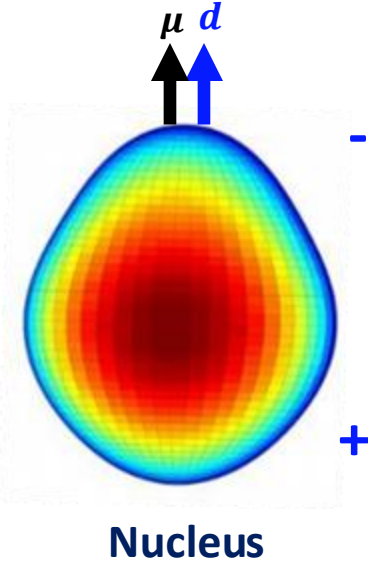
Atom/molecule
Nuclear

Atom
 $B \sim 3$ Tesla
 $\nabla E \sim 10^{18}$ V/cm²

Violate Parity and
Time reversal

$$H_{sym} = \dots + c^1_{(I,J)} \mu \cdot B + c^{1'}_{(I,J)} d \cdot E_{eff} + c^2_{(I,J)} Q \cdot \nabla E + c^{2'}_{(I,J)} Q_m \cdot \nabla B + \dots$$

Nuclei as a Rich Source of CP Violation



$$=$$

 Neutron EDM + Proton EDM + Quark EDM + Nucleon-nucleon interactions + ...

The diagram shows the decomposition of the nuclear EDM into four components:

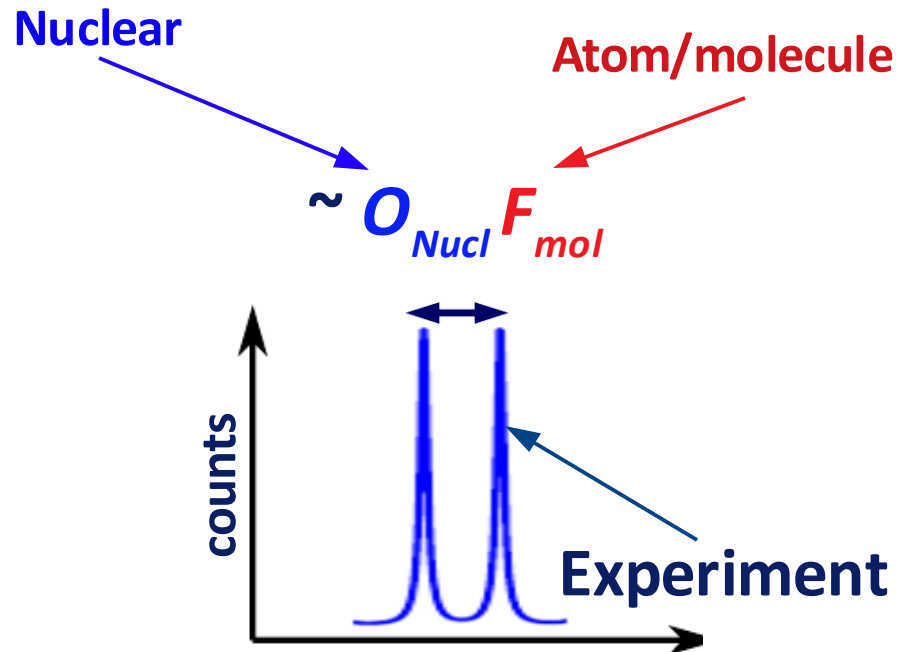
- Neutron EDM:** Three red circles labeled 'n' with upward-pointing arrows.
- Proton EDM:** Three blue circles labeled 'p' with upward-pointing arrows.
- Quark EDM:** Two stacked circles, the top one labeled 'u d' and the bottom one labeled 'u d'.
- Nucleon-nucleon interactions:** A red circle labeled 'n', a blue circle labeled 'p', and a blue circle labeled 'p'.

Why radioactive atoms & molecules?

$$H_{mol} = H_e + H_{vib} + H_{rot} + \dots + H_{hfs} + H_{PV} + H_{PTV} \quad \sim O_{Nucl} F_{mol}$$

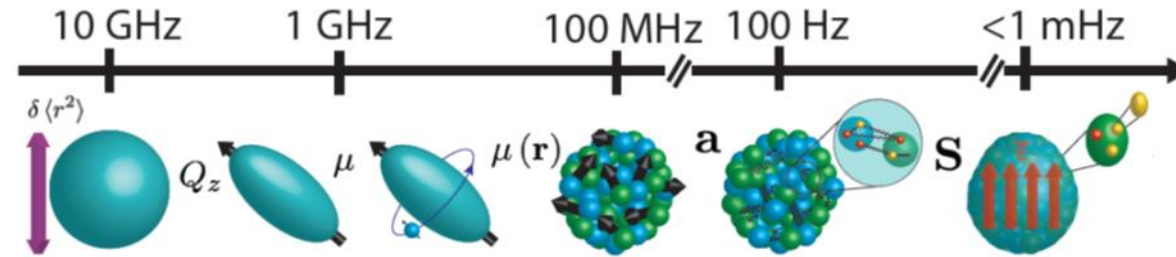
$\uparrow \quad \uparrow \quad \uparrow \quad \uparrow \quad \uparrow \quad \uparrow$
 eV ~ 2 10^{-2} 10^{-5} 10^{-8} $<10^{-12}$ $<10^{-18}$

$$H_{mol} - H'_{mol} \sim O_{Nucl} F_{mol} \quad \rightarrow \text{Direct measurements!}$$



Why (Radioactive) Molecules?

Heavy exotic
Nuclei Ra(Z=88)



$$H_{mol} = H_e + H_{vib} + H_{rot} + \dots + H_{hfs} + H_{PV} + H_{PTV}$$

[1 eV=241.8 THz]



$$\sim O_{Nucl} E_{mol}$$

Nuclear

$$\sim Z^3 A^{1/3} \beta_2 \beta_3^2 / (E_{+}^{N_+} - E_{-}^{N_-})$$

Molecule

$$\sim Z^3 / (E_{+}^{e_+} - E_{-}^{e_-})$$

Electric field ~ 100 GV/cm

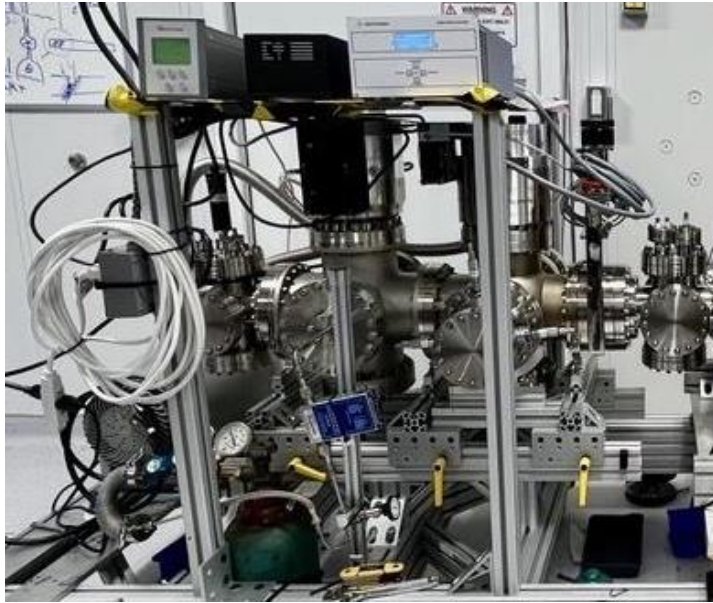
Radioactive molecules => **Best of all worlds!**

[Sandars Phys. Rev. Lett. 18, 1396 (1967)]
 [ACME, Nature 562, 355 (2018)]
 [Roussy et al. Science 381, 46 (2023)]

Molecules as sensitive probes of electroweak nuclear properties

Setup @ MIT

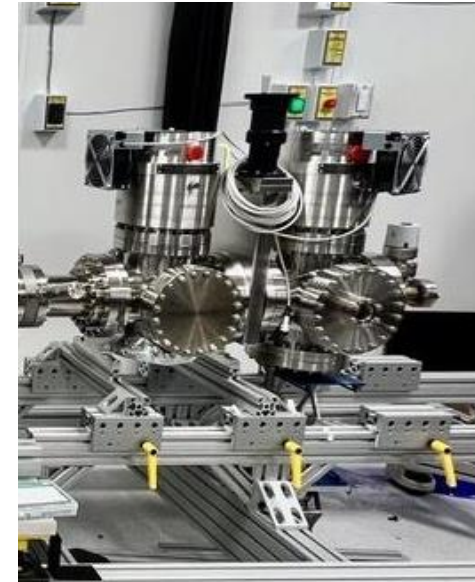
~3 m



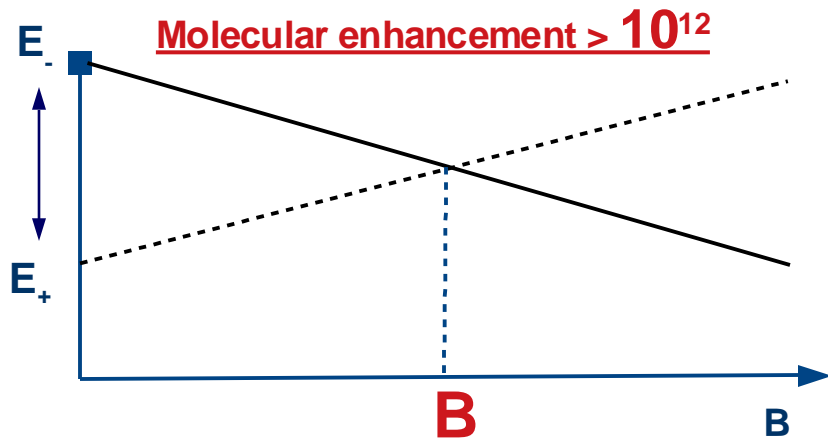
Paul trap



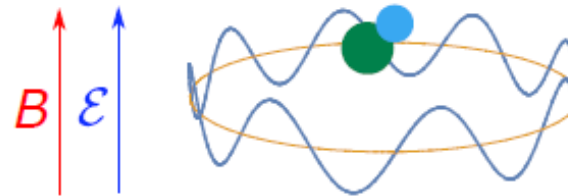
Penning trap



Ion source

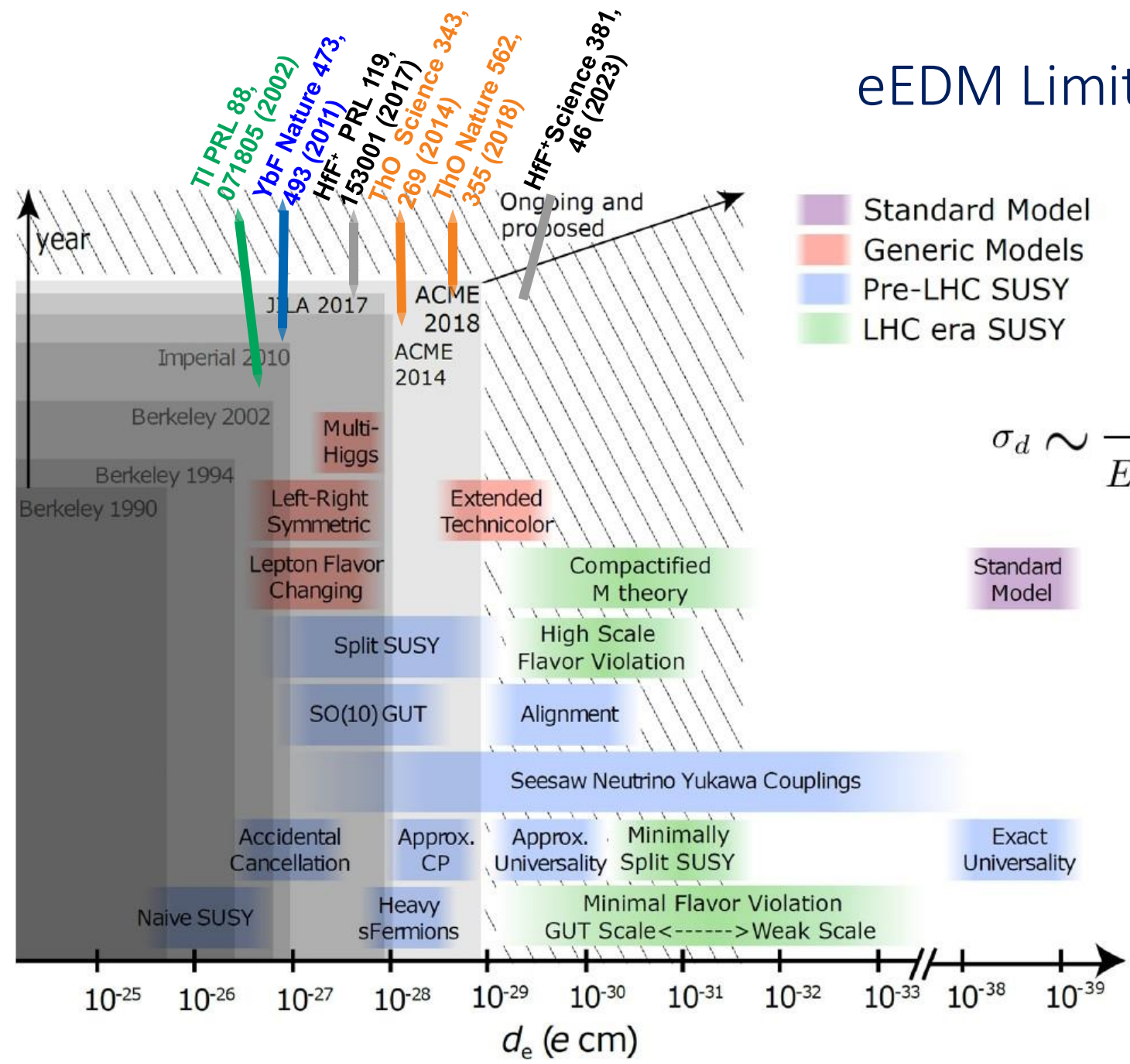


Inside Penning trap

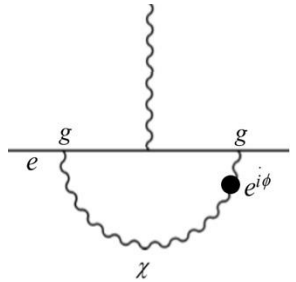


[Kartehin, Udrescu, Moroch et al. Under review in *Physical Review Letters* (arXiv:2310.11192) (2023)]

eEDM Limits



$$\sigma_d \sim \frac{1}{E_{\text{eff}} \tau \sqrt{N T}}$$



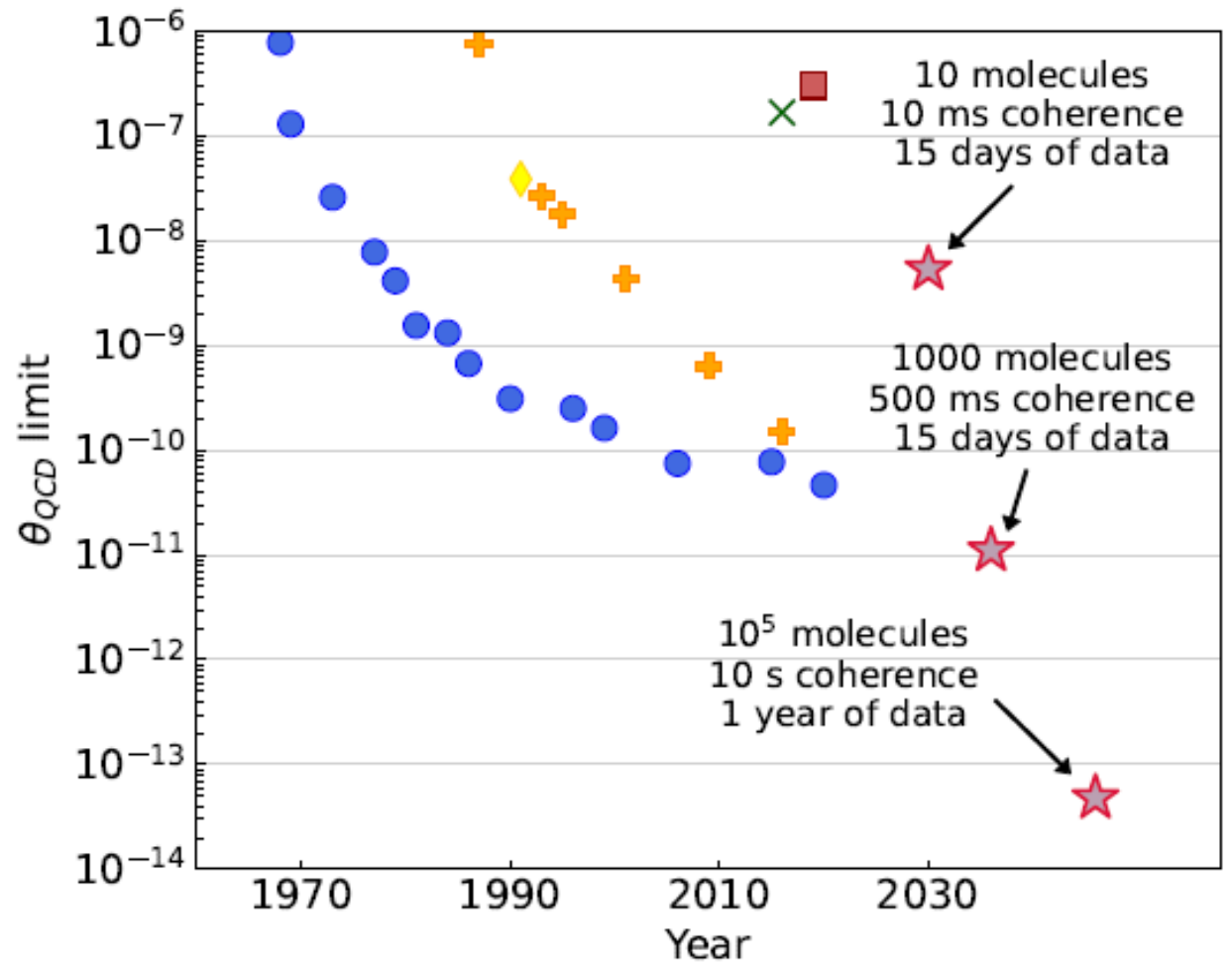
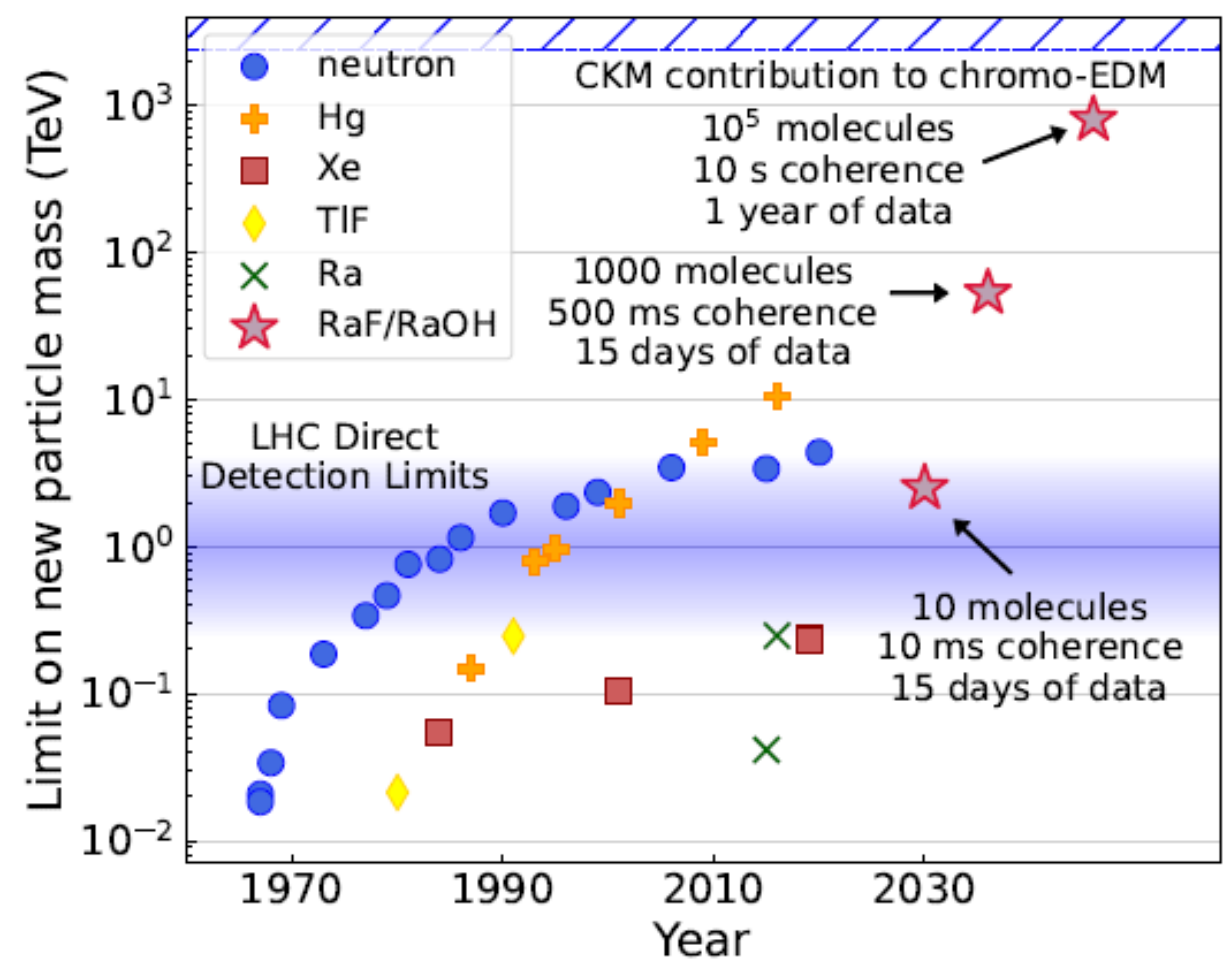
$$d_e \sim \mu_B \left(\frac{g^2}{2\pi} \right)^N \left(\frac{m_e}{m_\chi} \right)^2 \sin \phi$$

Probing physics @ TeV scale!

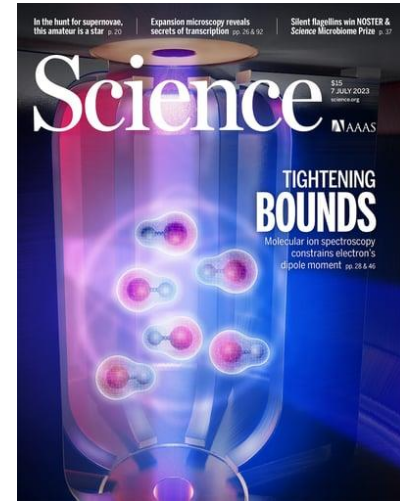
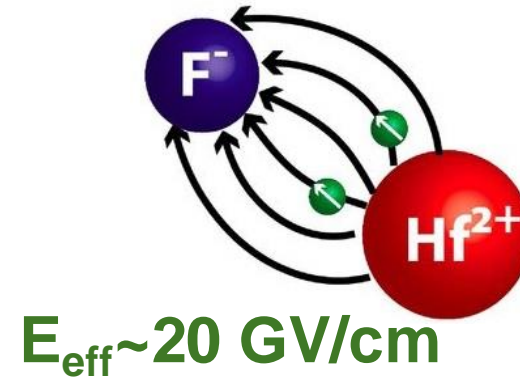
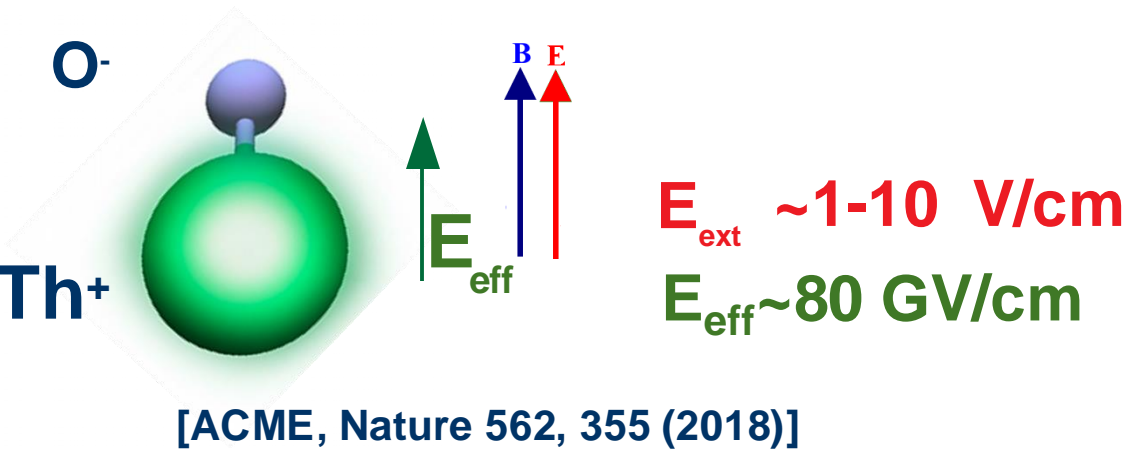
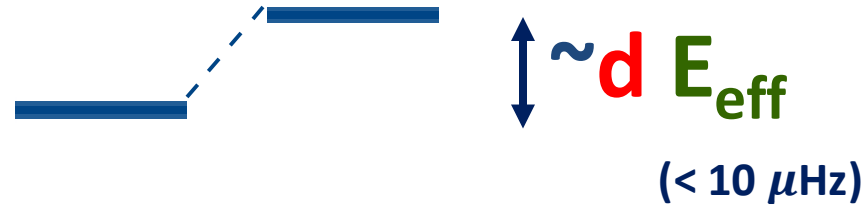
[Source: D. DeMille. Manipulating Quantum Systems: An Assessment of Atomic, Molecular, and Optical Physics in the United States (2019)]



Hadronic EDM Limits



Molecules for studies of P, T violation

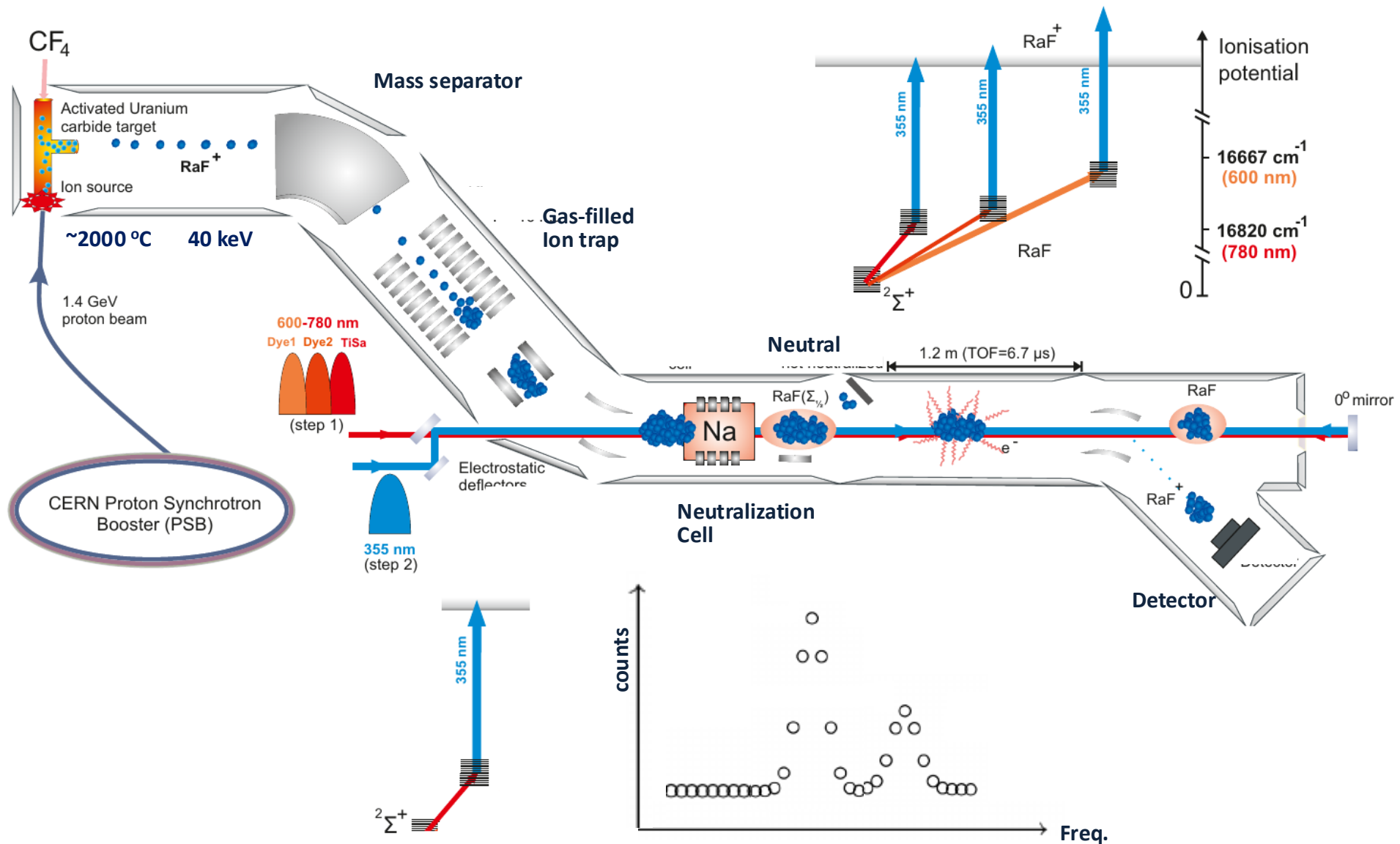


$$|d_e| \leq 4.1 \times 10^{-30} \text{ e} \cdot \text{cm}$$

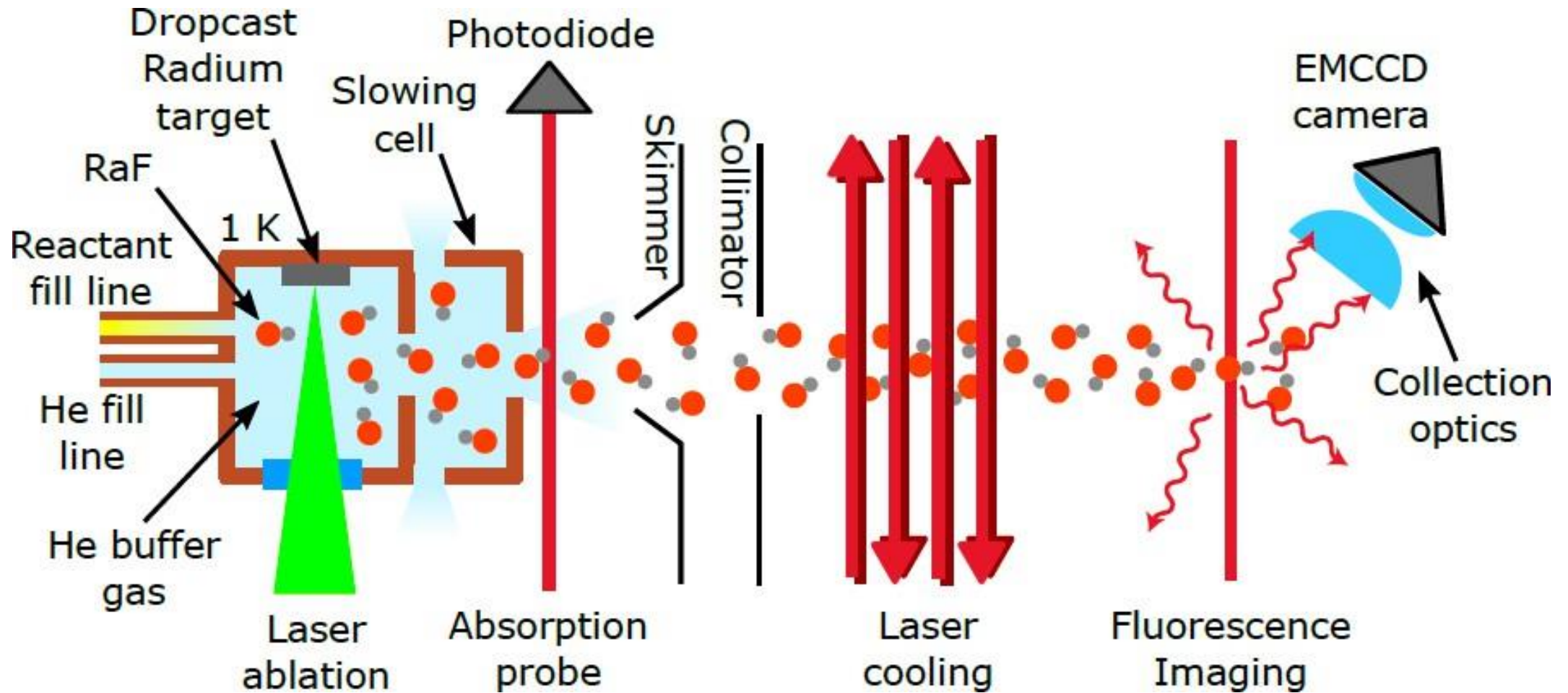
[Roussy et al. Science 381, 46 (2023)]

[Baron et al. Science 343, 269 (2014)]
 [Sandars Phys. Rev. Lett. 18, 1396 (1967)]

Recent Results (RaF)



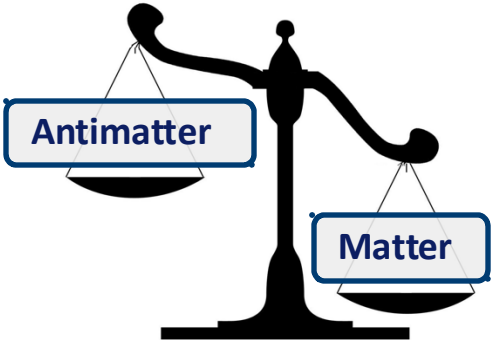
Laser Cooling of RaX Molecules



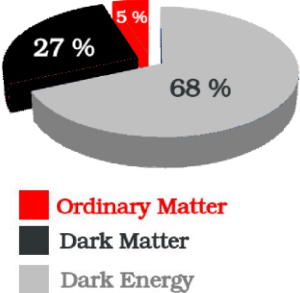
In Collaboration with J. Doyle (Harvard) & N. Hutzler (Caltech)

Major Open Questions in Physics and Cosmology

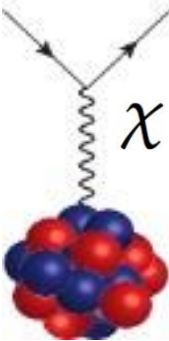
Why is there more matter?



What is Dark Matter?



Are there new particles?

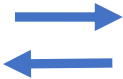


Strong CP problem

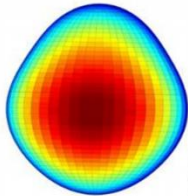
$$\mathcal{L} = \theta \frac{1}{16\pi^2} F_{\mu\nu}^a \tilde{F}^{\mu\nu a}$$

New sources of Time-reversal (CP) violation in nuclei can solve all of these problems

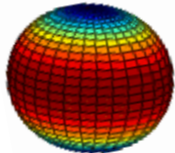
Time-reversal violation



Electric Dipole Moment (EDM) Of fundamental particles



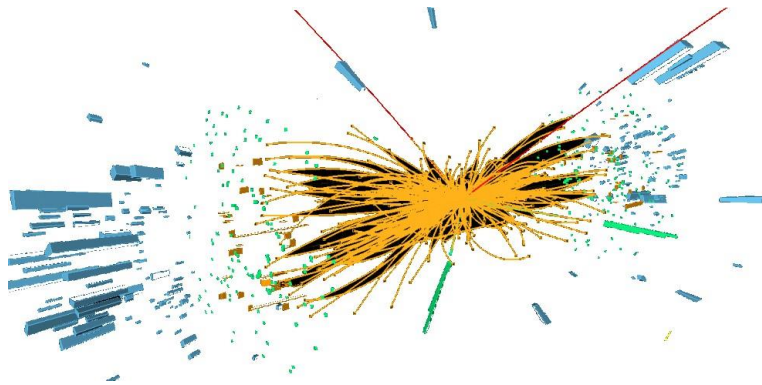
EDM > 0



EDM=0

Major Open Questions in Physics and Cosmology

Energy frontier



TeV

$<10^{-14}$

GeV

10^{-9}

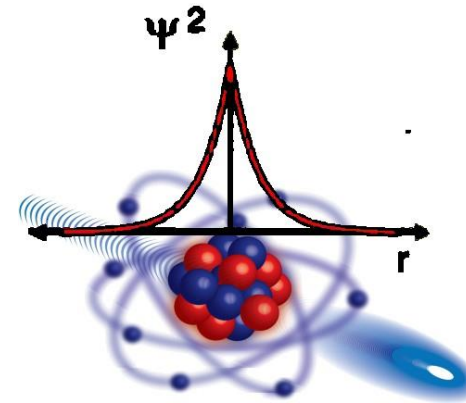
MeV

10^{-6}

eV

10^0

Precision frontier



Recent Results (RaF)



nature

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Article | Open Access | Published: 27 May 2020

Spectroscopy of short-lived radioactive molecules

R. F. Garcia Ruiz ✉, R. Berger ✉, [...]

Nature 581, 396–400 (2020) | Cite this article

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DOI:10.1063/PT.6.1.20200419a

11 Jun 2020 in Research & Technology

Spectroscopy of molecules with unstable nuclei

Pinning down the energy transitions of radium monofluoride, and eventually other short-lived molecules, could reveal the ways they are influenced by the properties of heavy radioactive nuclei.

Andrew Grant

physicsworld

ATOMIC AND MOLECULAR | RESEARCH UPDATE

Exotic radioactive molecules could reveal physics beyond the Standard Model

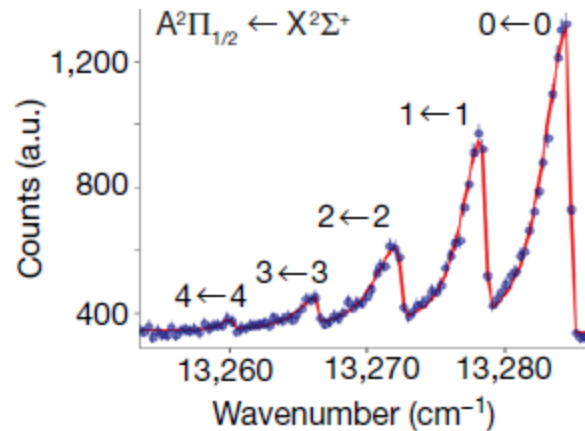
05 Jun 2020

CHEMISTRY WORLD

Molecular experiments hope to reveal new physics

BY ANDY EXTANCE | 11 JUN 2020

Detecting extremely short-lived radium fluoride can explore standard model's limits



Nature 581, 396 (2020)



$$H_{mol} = H_e + H_{vib} + H_{rot} + \dots + H_{hfs} + H_{PV} + H_{PTV}$$

eV ~ 2 10⁻² 10⁻⁵ 10⁻⁸ <10⁻¹² <10⁻¹⁸

Recent Results (RaF)



PHYSICAL REVIEW LETTERS

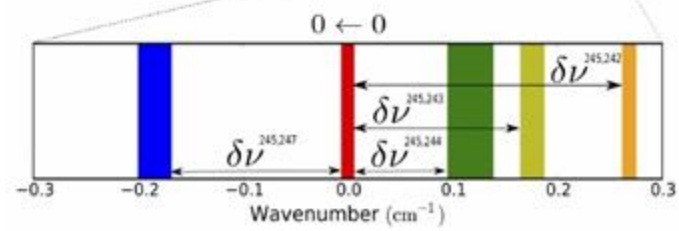
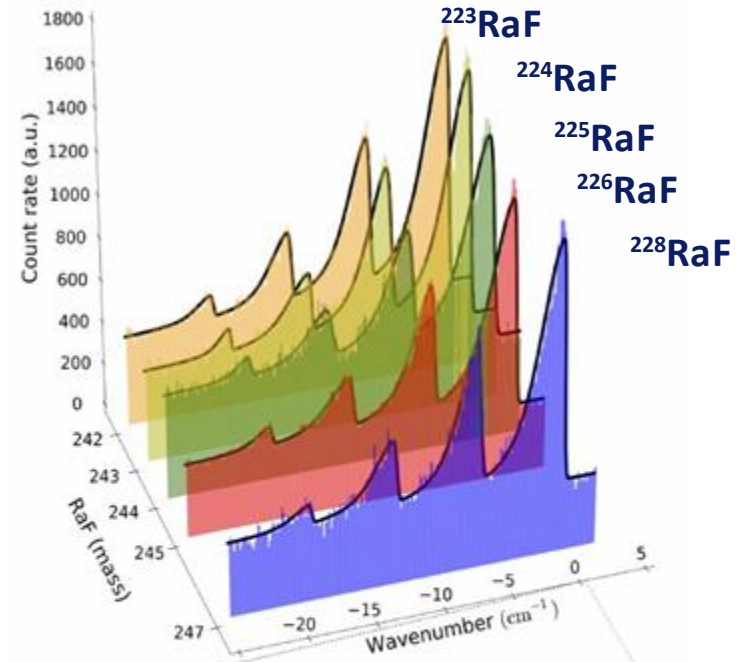
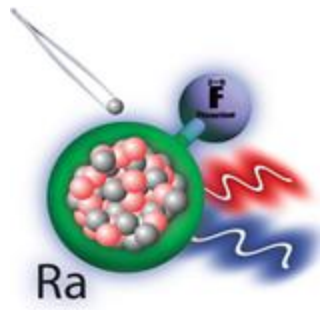
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Isotope Shifts of Radium Monofluoride Molecules

S. M. Udrescu *et al.*
Phys. Rev. Lett. **127**, 033001 – Published 14 July 2021

PhysiCS See Viewpoint: Sizing up Exotic Nuclei with Radioactive Molecules



Nature 581, 396 (2020) ✓
PRL 127, 033001 (2021) ✓

$$H_{mol} = H_e + H_{vib} + H_{rot} + \dots + H_{hfs} + H_{PV} + H_{PTV}$$

eV ~ 2 10⁻² 10⁻⁵ 10⁻⁸ <10⁻¹² <10⁻¹⁸

Recent Results (RaF)



nature > nature.physics > articles > article

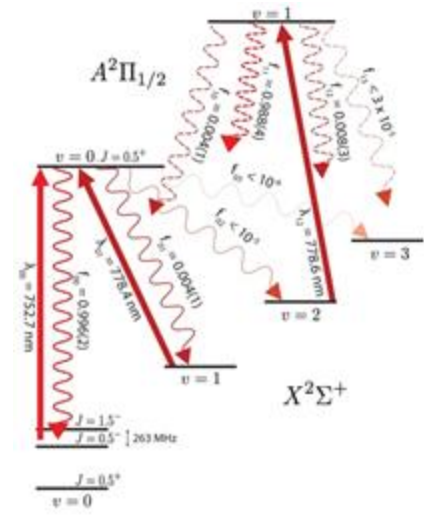
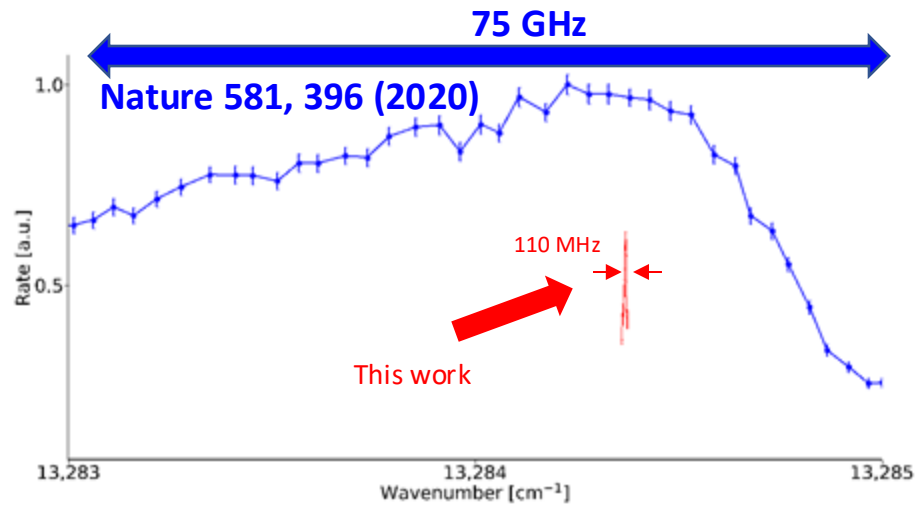
Article | Published: 09 January 2024

Precision spectroscopy and laser-cooling scheme of a radium-containing molecule

S. M. Udrescu, S. G. Wilkins, A. A. Breier, M. Athanasakis-Kaklamanakis, R. F. Garcia Ruiz, M. Au, I.

+ Show authors

Nature Physics 20, 202–207 (2024) | Cite this article

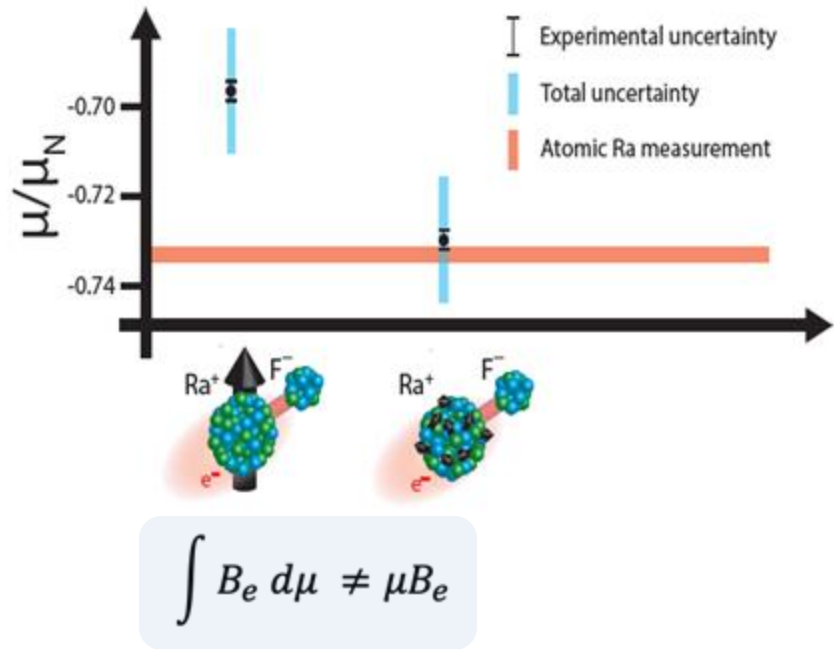
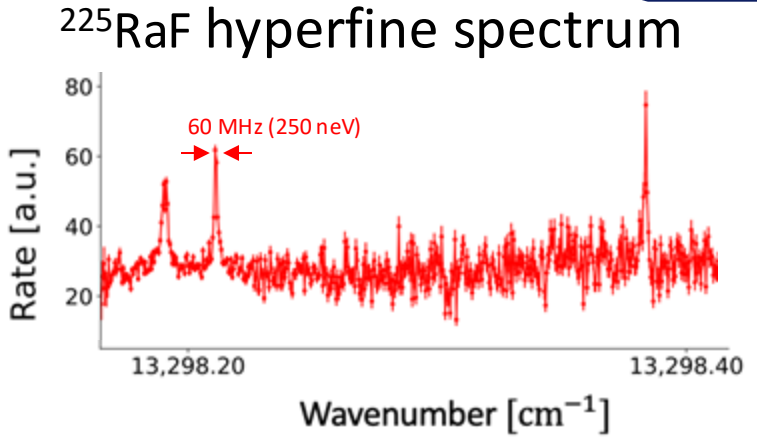


Nature 581, 396 (2020) ✓
PRL 127, 033001 (2021) ✓
Nature Phys 20, 202 (2024) ✓

$$H_{mol} = H_e + H_{vib} + H_{rot} + \dots + H_{hfs} + H_{PV} + H_{PTV}$$

eV ~ 2 10^{-2} 10^{-5} 10^{-8} $<10^{-12}$ $<10^{-18}$

Recent Results (RaF)



Nature 581, 396 (2020)

PRL 127, 033001 (2021)

Nature Phys 20, 202 (2024)

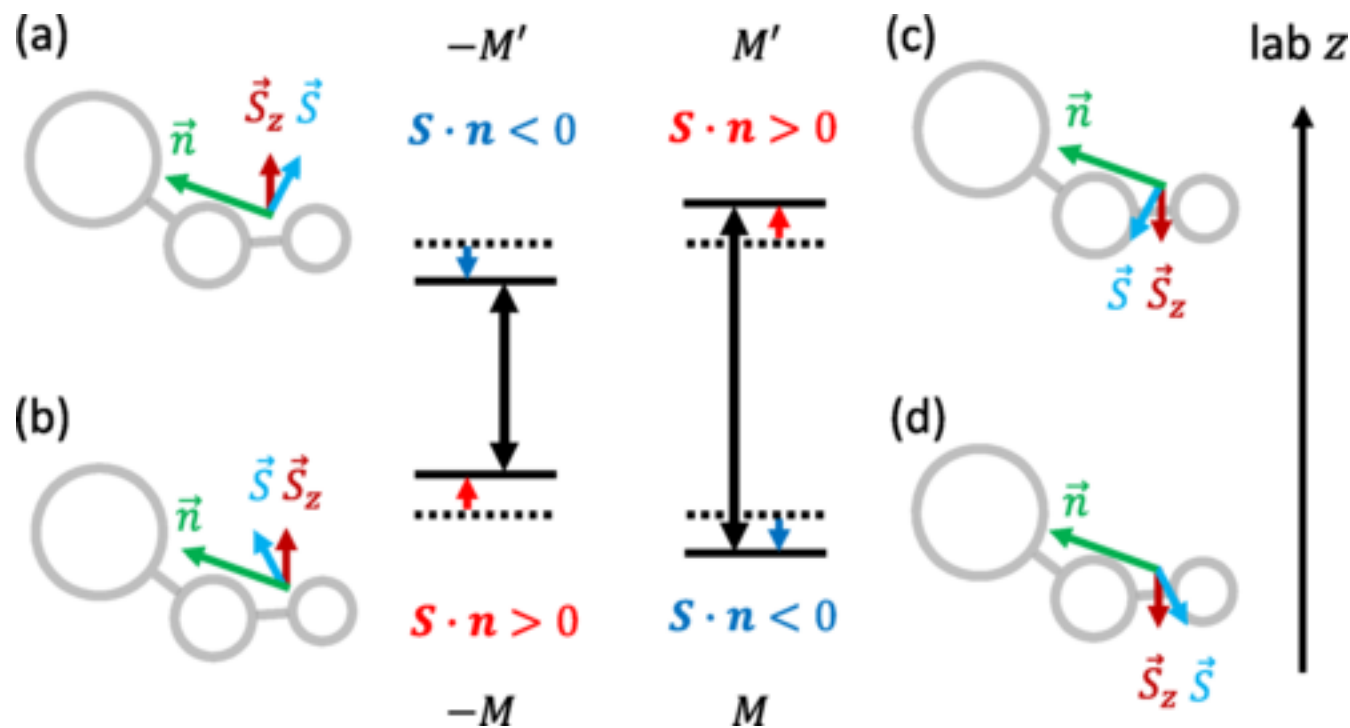
$$H_{mol} = H_e + H_{vib} + H_{rot} + \dots + H_{hfs} + H_{PV} + H_{PTV}$$

eV ~ 2 10⁻² 10⁻⁵ 10⁻⁸ <10⁻¹² <10⁻¹⁸



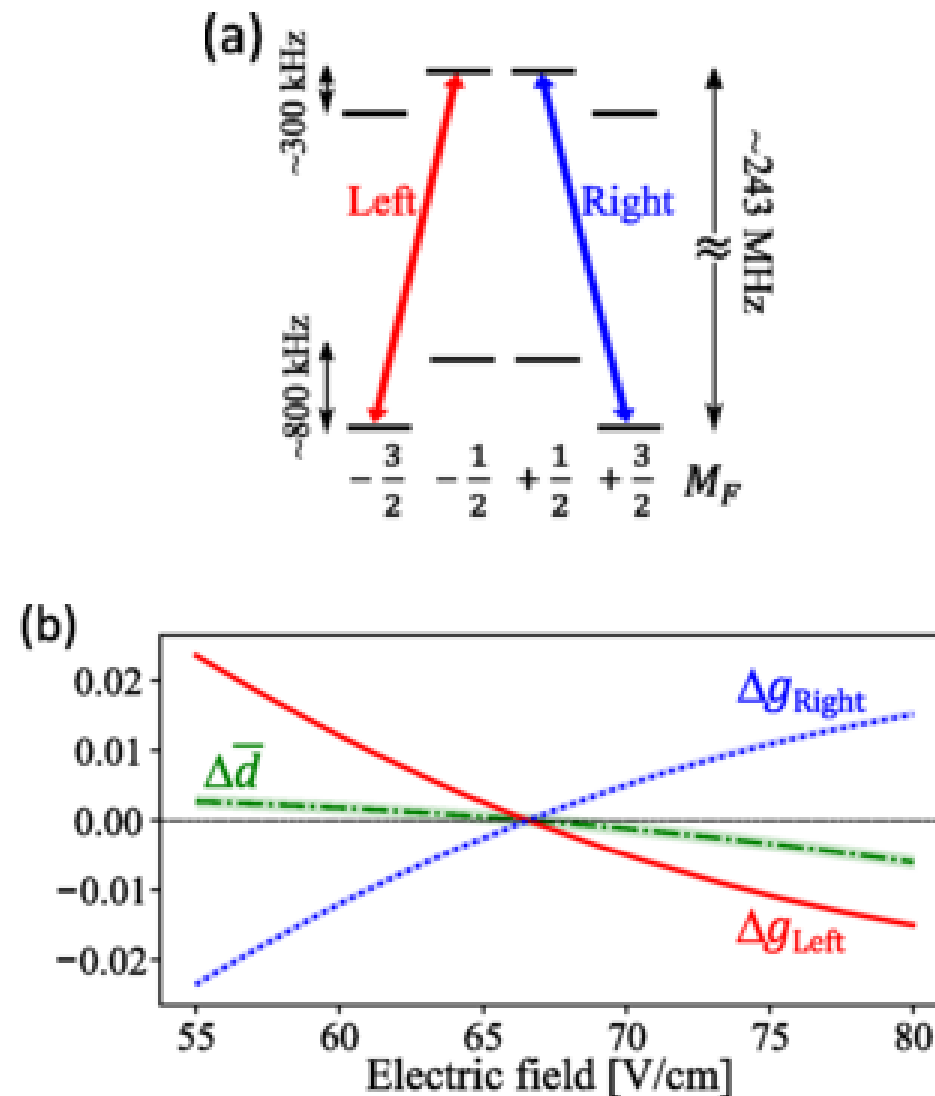
Engineering field-insensitive molecular clock transitions for symmetry violation searches

Takahashi, Zhang, Jadbabaie, Hutzler [Phys. Rev. Lett. 131, 183003 (2023)]

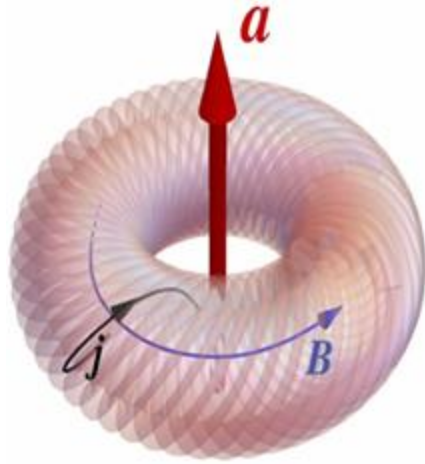


$$H = -D \vec{n} \cdot \vec{E} - g\mu_B \vec{S} \cdot \vec{B}$$

$$+ W_d d_e \vec{S} \cdot \vec{n} + W_Q \frac{Q}{I} \vec{l} \cdot \vec{n} - W_M \frac{M}{2I(2I-1)} \vec{S} \hat{T} \vec{n}.$$



Nuclear Anapole Moment

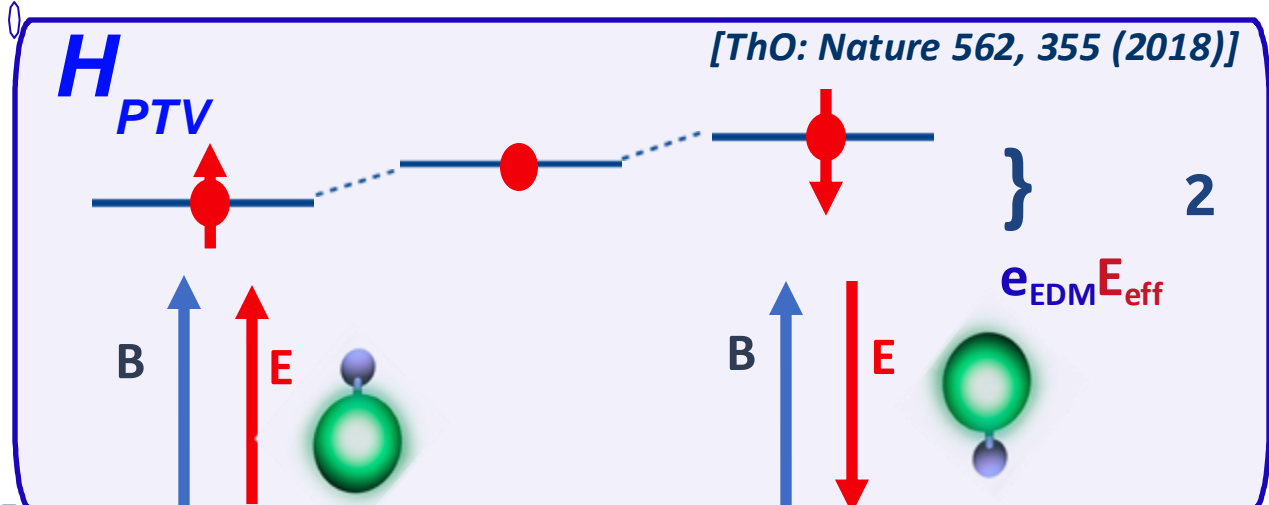
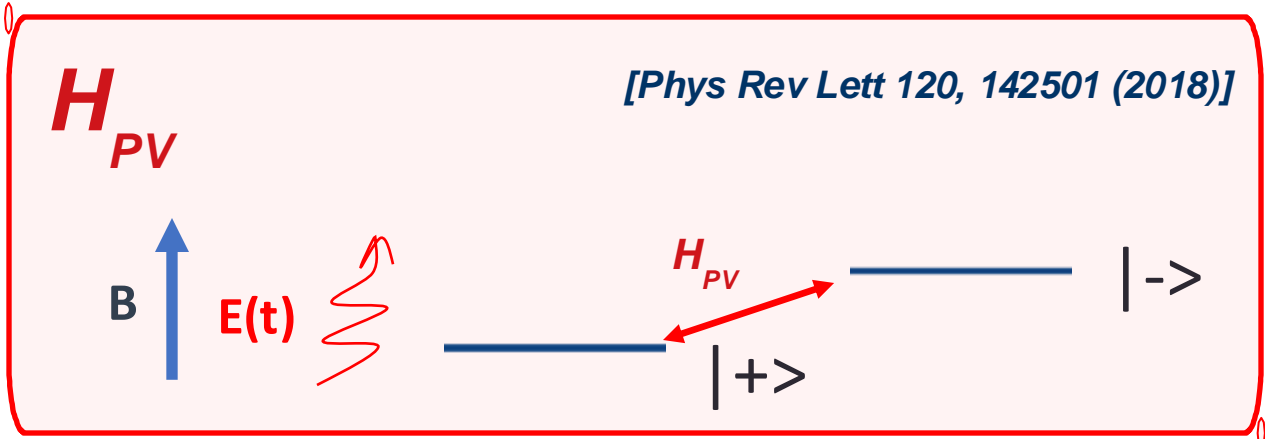


$$\sigma_d \sim \frac{1}{E_{\text{eff}} \tau \sqrt{\dot{N} T}}$$

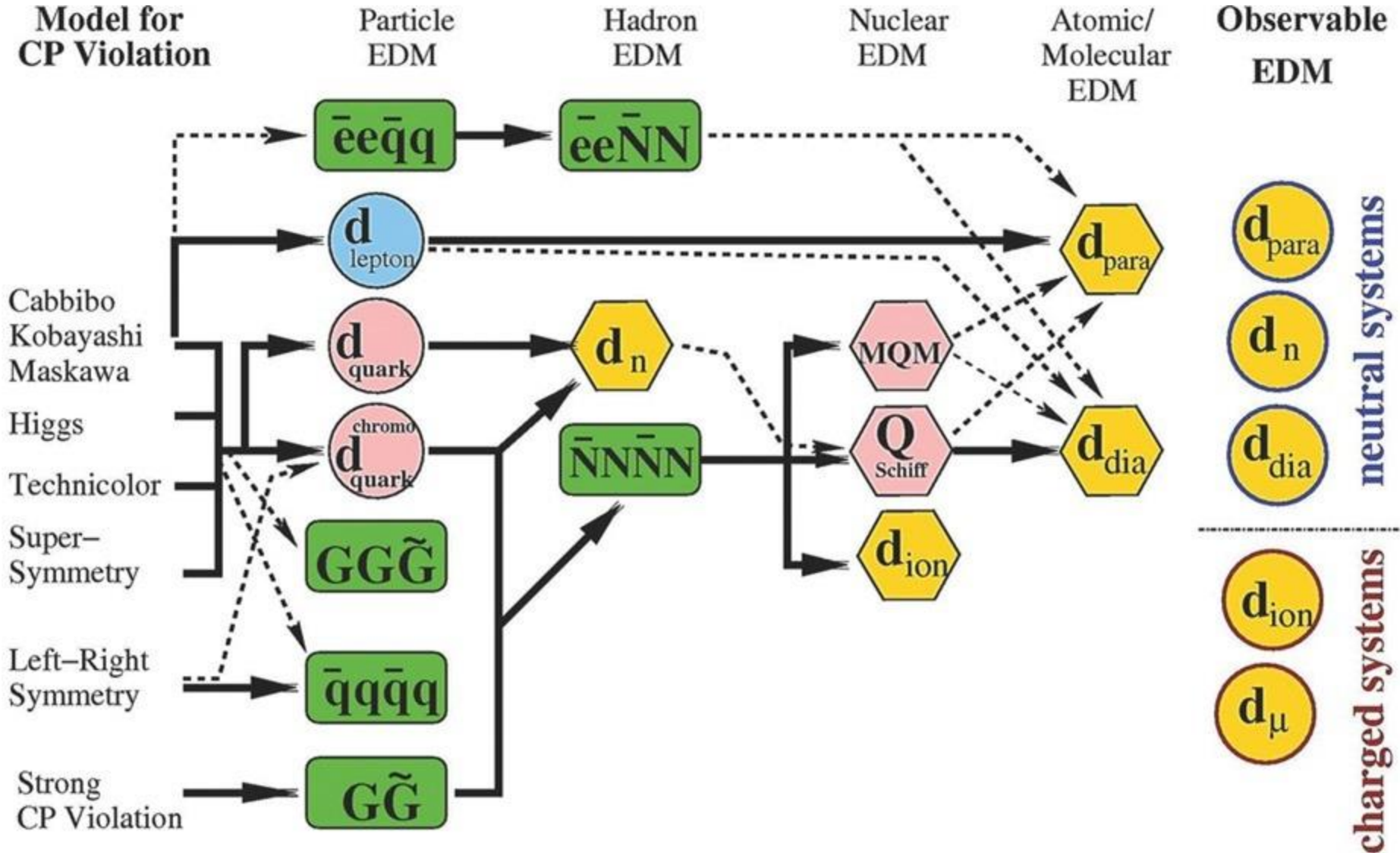
[Desplanques, Donoghue, Holstein et al. Ann. Phys. 124, 449-495 (1980)]

Can we achieve efficient cooling & trapping?

$$H_{mol} = H_e + H_{vib} + H_{rot} + \dots + H_{hfs} + \underbrace{H_{PV} + H_{PTV}}_{?}$$



[HfF⁺: PRL 119, 153001 (2017)]



Why radioactive nuclei?

Schiff

Moments

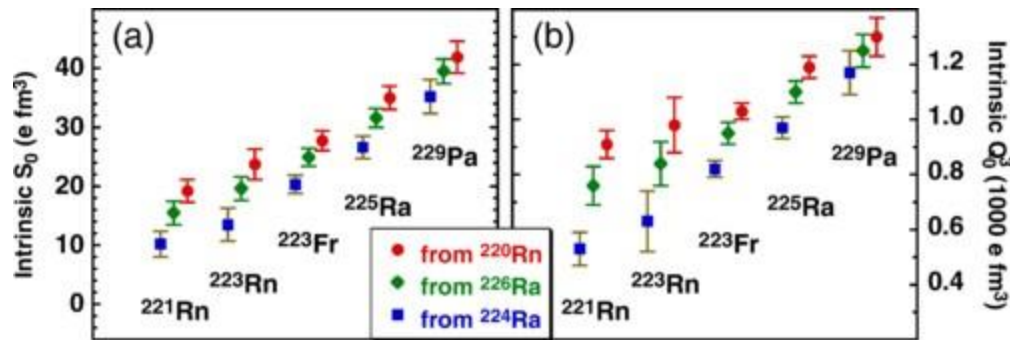
$$S = \langle \Psi_0 | S_z | \Psi_0 \rangle = \frac{\langle \Psi_+ | S_z | \Psi_- \rangle \langle \Psi_+ | V_{PT} | \Psi_- \rangle}{E_+ - E_-}$$

P,T-odd nucleon-nucleon interaction

Nuclear structure

Theory + Experiment

Experiment

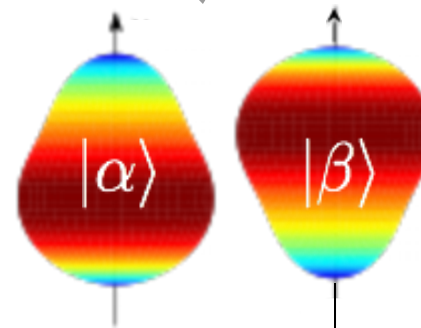


Dobaczewski et al. Phys. Rev. Lett. **121**, 232501 (2018)

Chupp et al. Rev. Mod. Phys. **91**, 015001 (2019)

$$\hat{S}_0 = \frac{e}{10} \sqrt{\frac{4\pi}{3}} \sum_i \left(r_i^3 - \frac{5}{3} r_{ch}^2 r_i \right) Y_0^1(\Omega_i) + \dots$$

DE: Energy splitting of opposite parity states



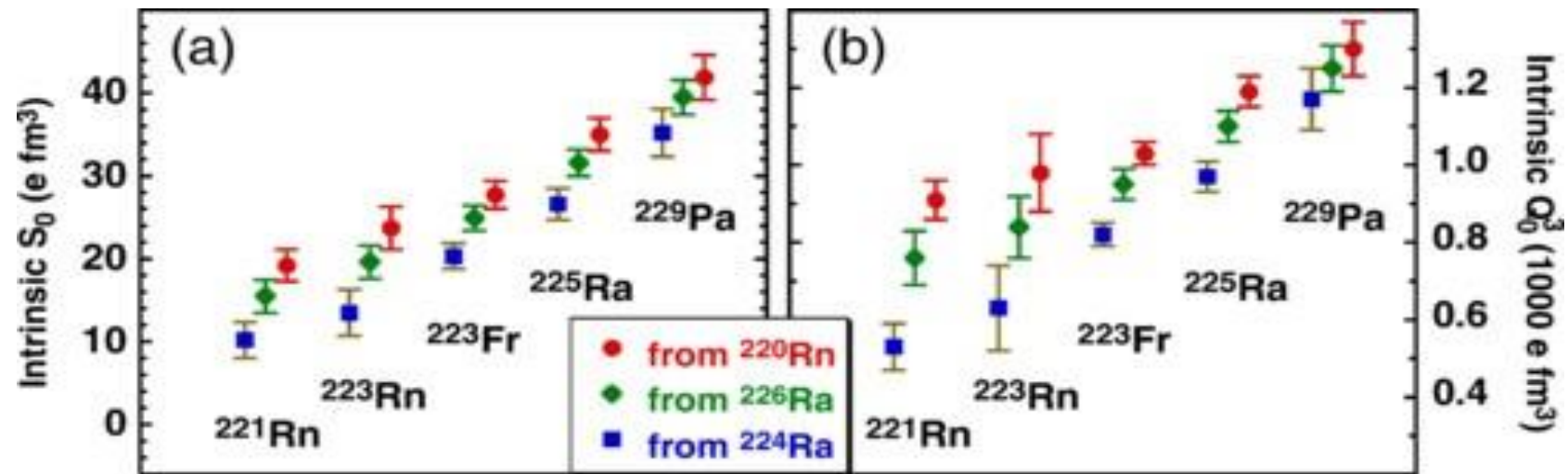
²²⁵Ra

DE = 55 keV



Slide from L. Gaffney

[Gaffney et al. Nature 497, 199 (2013)]



Dobaczewski et al. Phys. Rev. Lett. **121**, 232501 (2018)