

Precision Measurements with Radioactive Molecules

Nick Hutzler Caltech



Outline

- Brief review: atoms and molecules for CPV searches
- Advantages of quantum-controlled molecules
- Advantages of radioactive molecules
- Experimental developments
- Outlook and future directions



Atoms and molecules for CPV searches

Low Energy Observables

- The baryon asymmetry suggests new CP-violating physics
- Leads to CPV electromagnetic moments of regular matter
 - Electric dipole moment (EDM)
 - Nuclear Schiff moment (NSM)
 - Magnetic quadrupole moment (MQM)
- Enables sensitive probes of new physics with tabletop scales



The basic idea

- Look for CPV spin precession in the internal fields in atoms/molecules
- CPV shifts generally ~(spin)·(internal E field)
 - We just say "EDM measurements" even though some are not
- Measure spin precession between time-reversed states
 - Coherent measurement



CP-Violating Observables



Electron electric dipole moment (EDM)



Nuclear magnetic quadrupole moment (MQM)

Sensitive to nuclear CPV

Enhanced in heavy species



Nuclear Schiff moment (NSM)

Sensitive to nuclear CPV

Enhanced in heavy species

Enhanced in quadrupoledeformed nuclei Enhanced in octupoledeformed nuclei

Sensitive to electroncoupled CPV Enhanced in heavy species



Sensitivity

- Goal: measure some CPV electromagnetic moment in the electrons/nuclei
 - eEDM, NSM, or MQM
 - Ultimately arises from some CPV parameter $C = d_e$, d_n , $g_{\pi NN}^{(1)}$, ...
- Observable: energy shifts in an atom or molecules, ΔE
- Related by three factors: $\Delta E = C \times Q_{AM} \times Q_{nuc} \times Q_{pol} = C \times Q$
 - Q_{AM} = sensitivity of atom/molecule to the effect
 - How much does the energy shift given the presence of this electromagnetic moment?
 - "Internal fields"
 - Q_{nuc} = sensitivity of the nucleus to the effect
 - How large of a CPV moment is induced in the nucleus given *C*?
 - $Q_{pol} = polarization factor$
 - What fraction of the "internal fields" in the atom/molecule can be aligned in the lab?
 - $Q = Q_{AM} \times Q_{nuc} \times Q_{pol}$ = overall sensitivity of species to CPV
 - Punchline: Q is maximized for radioactive molecules
- See Snowmass EDM whitepaper 2203.08103 for even more details



Sensitivity

• Observable is phase ϕ after free precession for time τ

$$\phi = \tau \Delta E / \hbar = C Q \tau / \hbar$$

Repeated measurements:



Uncertainty in CPV parameter

Maximizing Q_{AM}

- Atoms/molecules have extreme internal electromagnetic environments
- Directly enhances electron EDM
 - Maximum lab field ~100 kV/cm
 - Q_{AM} given by effective field \mathcal{E}_{eff}
- Directly enhances NSM, MQM
 - More complicated than simple "internal field" picture... more later
 - Due to strong electron/nucleus interactions
- $Q_{AM} \sim Z^{2-3}$ generally
 - For species with similar electronic structure, e.g. periodic table columns



K. Gaul and R. Berger, PRA 101, 012508 (2020)

Maximizing Q_{pol} : Atoms vs. molecules

- Need to align internal fields in the lab frame
 - Otherwise, zero average direction → zero average energy shift
 - True for all static CPV effects
- Atoms
 - $\Delta \sim 100$ THz (electronic)
 - Q_{pol} ~ 10⁻³ @ 100 kV/cm
- Molécules
 - - Sometimes even smaller, more on that later
 - $Q_{pol} \gtrsim \mathcal{O}(1) @ 10 \text{ kV/cm}$
- "Molecules are 1000x more sensitive than atoms"
 - $Q_{AM} \sim \text{same}, Q_{pol} \text{ much larger}$



Molecular eEDM Searches



YbF, Imperial

- Spin precession in pulsed supersonic beam
- First to beat atomic TI limits
- |d_e| < 1.1 × 10⁻²⁷ e cm (2011)



HfF⁺, JILA/Boulder

- Spin precession in ion trap
- |d_e| < 1.3 × 10⁻²⁸ e cm (2017)
- $|d_{e}| < 4.1 \times 10^{-30} \text{ e cm} (2023)$



ACME, ThO, Harvard/Chicago/Northwestern

- Spin precession in cryogenic beam
- |d_e| < 8.7 × 10⁻²⁹ e cm (2014)
- |d_e| < 1.1 × 10⁻²⁹ e cm (2018)
 - Electron CPV only
 - 100x in 10 years
 - Each is being upgraded
 - More are under way
 - Methods are being extended to nuclear CPV

Future Improvements 10⁻²³ 0.002 0.05 10^{-24} CsĻ_{ŢI} One-Loop New Physics Reach [TeV] e 10⁻²⁵ 0.5 0.02 Electron EDM Limit [e cm] 10⁻²⁶ Reach Т YbF 10⁻²⁷ Experimental Limits 0.2 5 Two-Loop New Physics ThO 10⁻²⁸ ThO 10⁻²⁹ 50 2 10⁻³⁰ Improvements to existing experiments \rightarrow rovements Next-generation trapping \rightarrow 10⁻³¹ 500 Deformed Nuclei \rightarrow 20 Advanced quantum control→ 10⁻³² 10⁻³³ 5000 200 10⁻³⁴ 1985 1990 1995 2000 2005 2010 2015 2020 2025 2030 2035 2040 Year

Similar improvements in hadronic CPV are also anticipated

From 2022 Snowmass EDM whitepaper, arXiv:2203:08103 – Already out of date

Nuclear Schiff Moments

- Non-overlap of charge, spin in nucleus creates Nuclear Schiff Moment (NSM)
 - Electric field $\vec{E}_{NSM} \propto \vec{I}$ confined within the nucleus
 - Mixes core-penetrating, opposite parity atomic orbitals
- Need atomic *s,p* orbitals to penetrate nuclear core
- Need $\vec{I} > 0$ to align \vec{E}_{NSM}



V. V. Flambaum and J. S. M. Ginges, PRA 65, 032113 (2002)

Octupole Deformations

- NSMs can be enhanced in octupole-deformed (β₃) nuclei
 - Increases *Q_{nuc}* by **100-1,000**
- Combines with molecular Q_{pol} enhancements $\rightarrow \sim 10^6$ sensitivity gain vs. atoms with spherical nuclei
- The ability to trap and probe one molecule at a time would reach the frontiers of hadronic CPV
 - Atomic experiments are very advanced, continue to advance, are hard to beat
- ²²⁹Pa might have a further Q_{nuc} enhancement of ~ 10³



L. P. Gaffney et al., Nature 497, 199 (2013)

		Q _{nuc}	Q_{AM}	$Q_{nuc} \times Q_{AM}$	
88	²²⁵ Ra	-1	-8.25	8	
80	¹⁹⁹ Hg	0.005	-2.50	-0.013	
Ζ	Atom	$[e\mathrm{fm}^3\bar{\theta}]$	$10^{-17}S[e{\rm fm}^3]$	$10^{-17}\overline{ heta}$	
		\boldsymbol{S}	$d_A[e]$	cm]	

Why Octupole?

- In absence of symmetry violation, nucleus should be in superposition → octupole shape averages away
- PV, TV nuclear forces can mix these states
- Large charge, mass mismatch from asymmetric shape
- Deformed nuclei often have low-lying opposite parity states, ≤ 100 keV
 - Makes theory much more tractable → only few relevant states



N. Auerbach, V. V. Flambaum, & V. Spevak, PRL 76, 4316 (1996) J. Dobaczewski, J. Engel, M. Kortelainen & P. Becker, PRL 121, 232501 (2018), J. Engel et al., PRC 35, 1973R (1987)

Radioactive Nuclei

- Heavy, spinful, octupoledeformed nuclei are radioactive
- Nuclei must be in a molecule amenable to quantum control
 - Synthesis, spectroscopy, measurement protocol development, ...
- All of these pieces are coming together



lsotope	Half-life	
²²³ Fr	22 min	
²²⁵ Ra	15 d	
²²³ Ra	11 d	
²²⁷ Ac	22 yr	
²²⁹ Th	7,900 yr	
²²⁹ Pa	2 d	

Combining Enhancements



Radioactive molecules offer a unique opportunity to probe far beyond the current frontiers of fundamental symmetries 10⁶ molecules 100 s coherence time Heavy, deformed nucleus — Quantum control Robust error rejection



>PeV-scale CP-violating physics @ 1 loop >100 TeV-scale CP-violating physics @ 2 loops Both leptonic and hadronic sectors Extreme precision, $\theta_{QCD} < 10^{-14}$ Near Standard Model limit for NSMs

> Future orders-of-magnitude improvements from quantumenhanced metrology, highly exotic nuclei, ...

How can we implement advanced quantum control in CPV-sensitive molecules?



Advanced Quantum Control

Photon cycling in molecules

- The ability to scatter many photons is useful for quantum control in gas phase molecules
 - Efficient state control through optical pumping
 - Efficient detection
 - Optical forces/ cooling/trapping
 - Critical ingredient in ultracoherent atomic experiments (clocks)
- Decay to other states stops the cycling process
 - Vibration is limiting factor
 - Rotation, hyperfine → "solved"



Electronic structure for optical cycling

- Generally works for single, metal-centered s orbital
 - Alkaline-earth (s²)
 - Single bond to halogen (F)
- Orbital hybridization pushes electron away from bond
 - Decouples electronic structure from molecular excitations
- CaF, SrF, BaF, YbF, RaF,...
- Generally holds for pseudohalogens
 F → OH, CCH, OCH₃, ...
- This structure also gives good CPV sensitivity
 - Core-penetrating electrons



Parity Doublets

- Challenge: CPV experiments also rely on high polarizability
 - CPV sensitivity ∝ polarization
- Current best experiments utilize "parity doublets"
 - Each level in rotational ladder is split into a doublet
- Large polarization, small fields
 - Typically $\mathcal{O}(1)$ in ≤ 100 V/cm
 - Significantly easier engineering
 - Many systematics are ∝ E field
- Suppresses E,B systematics
 - States with oppose EDM, magnetic energy shifts
 - "Internal co-magnetometers"
 - Critical for leading experiments
- Laser coolable ²Σ diatomics don't have these



Polyatomic Molecules

- One solution: polyatomics generically have parity doublets
 - Arise from symmetry-lowering mechanical motions
- Use ligand as a resource to engineer molecule
 - Maintains laser cooling (usually)
 - Gives parity doublets
 - Tune electromagnetic sensitivity
- Solves two important limitations with laser-coolable diatomics
 - No doublets \rightarrow hard to polarize
 - Unpaired electron \rightarrow magnetic



I. Kozyryev and NRH, PRL 119, 133002 (2017)

¹⁷³YbOH NMQM @ Caltech

- MQM search in ¹⁷³YbOH
- ¹⁷³Yb has large MQM sensitivity
- Cryogenic buffer gas beam
- Laser cooling in future generations
- Parity doubling due to vibrational bending
 - Polarize in ~100 V/cm
 - Internal co-magnetometers
- All-optical spin precession and readout







Spin Precession

- Observed electron spin precession in bending mode
- Use two-photon transition to prepare, read superposition
- Can engineer fieldinsensitive clock transitions to suppress decoherence
 - Generally useful for a wide range of species
 - Helps address problems from large nuclear hyperfine structure (relevant!)



Y. Takahashi, C. Zhang, A. Jadbabaie, NRH, PRL 131, 183003 (2023)



EDM Pathfinder

- Pathfinder experiment using trapped, ultracold CaOH @ Harvard with John M. Doyle
 - Trapped at tens of μK
 - EDM sensitivity is small
 - Science state X(010) structure is identical to other [AE]-OH
- Demonstrated PolyEDM
 - Full EDM protocol
 - Directly applicable to similar species (Sr, Ba, Yb, Ra)OH
 - PolyEDM science species: SrOH





Loïc Anderegg Harvard University



Engineering Coherence

- Able to tune-out magnetic sensitivity
- Tune magnetic projection via E field





L. Anderegg, N. B. Vilas, C. Hallas, P. Robichaud, A. Jadbabaie, J. M. Doyle, and NRH, Science 382, 665 (2023)

Engineering Coherence





L. Anderegg, N. B. Vilas, C. Hallas, P. Robichaud, A. Jadbabaie, J. M. Doyle, and NRH, Science 382, 665 (2023)



Radioactive Molecules

Can we find a species which combines everything I have discussed?

- Heavy, octupole nucleus \rightarrow NSM enhancements
- Laser coolable \rightarrow Advanced quantum control
- Polyatomic structure \rightarrow Systematic error control

Where to start?



Octupole nucleus

Radium

This contributed to motivation for groundbreaking RaF work



R. F. Garcia Ruiz *et al.*, Nature 581, 396 (2020) T. A. Isaev, S. Hoekstra, and R. Berger, PRA 82, 052521 (2010)

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Neutral Radium Polyatomics

- RaOH is the simplest molecule combining everything I have discussed
 - Laser cooling
 - Molecular enhancement
 - Octupole deformed nucleus
 - High polarizability
 - Tunable magnetic moments
 - Advanced quantum control
 - ...
 - Demonstrated protocols
- Predicted to be very laser-coolable
 - Periodic trends help the ability to cycle photons in this case
- More complex ligands in future
- Could probe boundaries of leptonic and hadronic CPV with only a few trapped molecules
- Need to produce and cool to ~few K to apply the methods I discussed



Number of Lasers

C. Zhang, NRH, L. Cheng, J. Chem. Theory Comput. 19, 4136 (2023)

See also: T. A. Isaev, A. V. Zaitsevskii, E. Eliav, J. Phys. B. 50, 225101 (2017) I. Kozyryev and NRH, Phys. Rev. Lett. 119, 133002 (2017)

RaOH Spectroscopy

- Recently started studying ²²⁶RaOH at Caltech
- Tabletop apparatus
- Cooled to 4 K
- Main challenge: quantity(Obviously)
- Once the quantity problem is addressed, we can use the full quantum control toolbox



Molecular production

- MOH are free radicals
 - M=Ca, Sr, Ba, Yb, Ra
- Create via cryogenic buffer gas cooling
- Vaporize solid targets with pulsed laser
 - Some combination of M, O, H
 - We have tried many, many things
- Cool to 4 K with cryogenic helium
 - Translationally and internally
- Optically drive chemical reactions to create desired species
 - Excited M can overcome reaction barriers
 - Increase yield ~10x
 - Has been demonstrated in all of these different species
- Can make cold, slow, intense beams with an aperture in the gas cell



A. Jadbabaie, N. Pilgram, J. Kłos, S. Kotochigova, NRH, New J. Phys. 22, 022002 (2020) NRH, H. Lu, and J. M. Doyle, Chem Rev 112, 4803 (2012) 35

Target Production

- Currently fabricating ablation targets in-house
- 10 µCi ²²⁶RaCl solution from Eckert & Ziegler
 - 370 kBq, 10 μg, ~3 x 10¹⁶ atoms
- Pipette ("dropcast") solution onto substrate to évaporate
- Surround with $(something)_{x}(OH)_{y}$
 - We tried several things
- This is an area of major potential improvement
 - More later

RaCl₂ surrounded by reaction precursors

Isotope Lab @ Caltech



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Synthesis and Detection

- Vaporize Ra and Al(OH)₃ with pulsed YAG
- $Ra^{+}H_2O \rightarrow RaOH+H$
 - Excite Ra to metastable state
 - ${}^{1}S_{0} {}^{3}P_{1}$, 714 nm
- Cool RaOH with 4 K He
 - Molecules are cold, stopped
 - Same starting point as other molecular precision measurement, laser cooling experiments
 - Engineering challenges remain
- Detect RaOH through resonant fluorescence spectroscopy
- Also produced and detected RaOD with deuterated targets





Species selectivity

- Ablation creates many species uncontrollably
 - How can we distinguish RaOH vs. ???
- Optically-driven M*+H₂O chemistry is *resonant* with metal M
 - M-containing molecules will change
 - Others will not
- Can even use to disentangle isotopologues since 4 K Doppler < Isotope shifts
- First demonstrated with YbOH spectroscopy
 - Very congested since multiple isotopes and isotope ~ hyperfine ~ rotation



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Robust spectroscopy with dense and contaminated spectra



N. H. Pilgram, A. Jadbabaie, Y. Zeng, NRH, T. C. Steimle. J. Chem Phys. 154, 244309 (2021)

Search Approach

- Small quantity and theoretical uncertainties are challenges
 - 10 μ Ci \rightarrow few thousand shots
 - Theory uncertainty \rightarrow ~10 THz
 - Linewidth ~100 MHz
 - (10 THz)/(100 MHz) ~ 10⁵
- Solution: phased search
 - ~40 GHz \rightarrow ~3 GHz \rightarrow ~100 MHz
 - This is common, but adapting to our system took work
 - (Typical ns pulsed lasers present challenges)

Partially depleted Ba target



Cold RaOH Spectroscopy - Preliminary





Improvements

- Previously: resolution limited by inconsistent production
 - Large shot-to-shot fluctuations
 - → too many averages needed
- Improvements in last few months
 - Mostly with BaOH, handled in equivalent forms and amounts, as a proxy
 - Improved atomic production via dropcasting tweaks
 - Atomic normalization via strong ${}^{1}S_{0} {}^{1}P_{1}$ line (483 nm in Ra, 553 nm in Ba)
 - Optimized multi-pulse ablation
 - Aggressive background/noise reduction
 - Better amplifiers/digitizer
 - Lock-in detection
 - Helium cryoplasma mitigation
- Now: consistent single-shot BaOH fluorescence with signal/noise > 100
 - With old methods, never saw BaOH on a single shot
- Next radium run starting on Thursday!



RaOH 10 μCi Last year SNR ~ few Inconsistent





Going Forward

- Improved ablation target production
 - Time consuming (hand pipetting)
 - Drop-on-demand?
 - Electrochemistry?
 - Something else?
 - Too expensive, pre-made RaCl \$\$\$
 - Buy from DOE
 - Use other forms?
- Other ligands
 - Straightforward to change ligand
 - "We do this all the time"
 - Optimizing production still takes work
 - Can vary solid precursor or flow reactive gas
- Other species entirely?
 - Nuclear structure, nuclear astrophysics, exotic chemistry, ... ?
 - In general, if we can get ≥ 10 ug of material we can probably make molecules out of it
 - Smaller quantities in the future as we push on the optimization
- Combine with radioactive beam methods
- Shorter lived isotopes
- This method would benefit greatly from increased interactions with the community!

Calcium-containing molecules which have been buffer gas cooled



D. Mitra et al., J. Phys. Chem. Lett. 13, 7029 (2022), G. Lao et al., J. Phys. Chem. Lett. 13, 11029 (2022), G.-Z. Zhu et al., Nat. Chem. 14, 995 (2022)

Many other approaches

- See 2302.02165
- Other neutral molecules
 RaF, AcX, ThX, FrX, PaX, ...
- Photoassociated neutrals
 - FrAg, RaAg, ...
- Molecular ions
 - RaX⁺, ThF⁺, AcOH⁺, PaF₃⁺, ...
- Cryogenic matrices
 - RaX, Pa, ...
- Atoms
 - Ra, Rn, ...
- We need experiments in multiple systems
 - There is no best approach







Solids



EDM, NSM, MQM – Complementarity

Wilson coefficient	Operator (dimension)	Number	$-ImC_{eq}(v/\Lambda^{-})$ $-6 \times 10^{-9} -4 -2 0 2 4 6$
δ_e	Electron EDM (6)	1	-4×10^{-6}
Im $C^{(1,3)}_{\ell equ}$, Im $C_{\ell eqd}$	Semi-leptonic (6)	3	1.0 -
$ar{ heta}$	Theta term (4)	1	0.5 -
δ_q	Quark EDM (6)	2	
$\tilde{\delta}_q$	Quark chromo EDM (6)	2	
$C_{\tilde{G}}$	Three-gluon (6)	1	-0.5 -
$\operatorname{Im} C_{auad}^{(1,8)}$	Four-quark (6)	2	
$\operatorname{Im} C_{\varphi ud}$	Induced four-quark (6)	1	-1.0 — ThO
Total		13	-1.5×10^{-27} -5×10^{-8} 0 5
	Wilson coefficient δ_e $\operatorname{Im} C_{\ell equ}^{(1,3)}$, $\operatorname{Im} C_{\ell eqd}$ $\bar{\theta}$ δ_q δ_q δ_q $\tilde{\delta}_q$ $C_{\tilde{G}}$ $\operatorname{Im} C_{quqd}^{(1,8)}$ $\operatorname{Im} C_{\varphi ud}$ Total	Wilson coefficientOperator (dimension) δ_e Electron EDM (6) $\operatorname{Im} C_{\ell equ}^{(1,3)}, \operatorname{Im} C_{\ell eqd}$ Semi-leptonic (6) $\bar{\theta}$ Theta term (4) δ_q Quark EDM (6) δ_q Quark chromo EDM (6) $\tilde{\delta}_q$ Four-quark (6) $\operatorname{Im} C_{quqd}^{(1,8)}$ Four-quark (6) $\operatorname{Im} C_{\varphi ud}$ Induced four-quark (6)TotalTotal	Wilson coefficientOperator (dimension)Number δ_e Electron EDM (6)1 $\operatorname{Im} C_{\ell equ}^{(1,3)}, \operatorname{Im} C_{\ell eqd}$ Semi-leptonic (6)3 $\bar{\theta}$ Theta term (4)1 δ_q Quark EDM (6)2 δ_q Quark chromo EDM (6)2 $\tilde{\delta}_q$ Three-gluon (6)1 $\operatorname{Im} C_{quqd}^{(1,8)}$ Four-quark (6)2 $\operatorname{Im} C_{\varphi ud}$ Induced four-quark (6)1TotalTotal13

J. Engel, M. J. Ramsey-Musolf, and U. van Kolck, Prog. Part. Nucl. Phys. 71, 21 (2013) Snowmass ED

Snowmass EDM white paper, arXiv:2203.08103

- Complex CPV parameter space <u>requires</u> multiple systems to obtain physically meaningful values
 - Atom/molecule CPV measures a sum of sources
 - Need >1 eEDM and NSM and MQM, but also nucleon EDM
 - Dependencies are generally complementary

An exciting time – everything is coming together



Radioactive beam spectroscopy



Radiochemistry, isotope access

Trapping/control

-









Rapid broadband spectroscopy

Advanced quantum control

Sensitivity Outlook Pathy hadron

Pathway to seeing hadronic CKM physics



* One-loop mass reach through quark chromo-EDMs. Different species have different mass reach for different hadronic CPV sources. 47



Thanks for your attention!

Please get in touch, or learn more at:





PolyEDM Collaboration <u>www.polyedm.com</u>



SLAM! Community <u>www.slamcommunity.com</u>



Hutzler Lab, Summer 2023 Standing: Daniel, Phelan, Yi, Nick, Chandler Sitting: Ashay, Arian, Madison, Taz, Adele Hovering: Tim, Chi, Yuiki





Caltech DeLogi Science and Technology Grant

Collaborators

PolyEDM

John M. Doyle (Harvard) Tim Steimle (ASU/Caltech) Amar Vutha (Toronto)

Molecular Theory

Anastasia Borschevsky (Groningen) Bill Goddard (Caltech)

SLAM!

Lan Cheng (JHU) Alyssa Gaiser (MSU) Ronald Garcia Ruiz (MIT) Andrew Jayich (UCSB) Jaideep Singh (MSU/FRIB)





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Backup Slides

RaX vs. Hg

- Nuclei have different sensitivity to underlying sources of new physics
 - Makes comparing "how much more sensitive" difficult
- One metric: what is ΔE for NSM induced by CKM?
 - Disclosure: the theory uncertainties on these is large/unknown
 - Disclosure: this involved combining many different units and conventions
- Hg
 - CKM predicts NSM-induced atomic EDM |d^S_{Hg}| = 4.2 × 10⁻³⁵ e cm
 Experiment uses 10 kV/cm → | ΔE/h | ≈ 1 × 10⁻¹⁶ Hz
- RaX (X=F, OH, OH, ... all fairly similar)
 - CKM predicts $|d_{Ra}^{S}| = 7.4 \times 10^{-32} e cm$
 - Atomic sensitivity $|K_a^S| = 8.8 \times 10^{-17} \text{ cm}/(fm)^3$
 - CKM predicts $|S_{Ra}| = 8.3 \times 10^{-16} e (fm)^3$
 - RaX sensitivity $\Delta E/S_{Ra} \approx 2.5 \times 10^4 \ e/4\pi\epsilon_0 a_0^4$
 - Energy shift $|\Delta E/h| \approx 1 \times 10^{-9} Hz$
- $|\Delta E_{RaX,CKM}/\Delta E_{H.g,CKM}| \approx 10^7$

See V. V. Flambaum, Phys. Rev. C 99, 035501 (2019), N. Yamanaka and E. Hiyama, J. High Energy Phys. 2016, 67 (2016), and references contained therein