Probing for new physics with the electron's Electric Dipole Moments: in atoms and molecules Selective account of the Past, Present, and Future

Kia Boon Ng, JILA & TRIUMF

06 March 2026

Lake Louise Winter Institute

Agenda

- Overview
  - Why electric dipole moments (EDMs)?
  - History of EDM measurements.

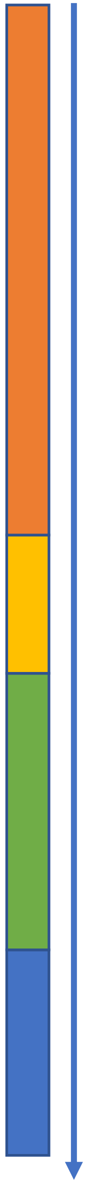
How eEDM experiments came to be

- Electron's EDM at JILA (HfF^+, ThF^+)
  - Current status (JILA Gen. II, Nov 2022).
  - JILA Gen. III (ongoing).

Diving into quantum-controlled molecules for fundamental physics

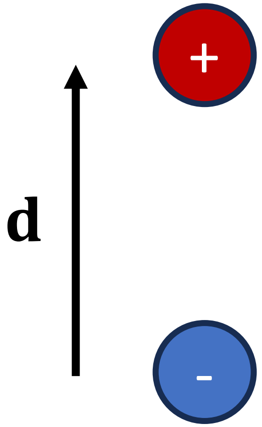
- What lies ahead
  - More advanced technology for eEDM.
  - Beyond eEDM.

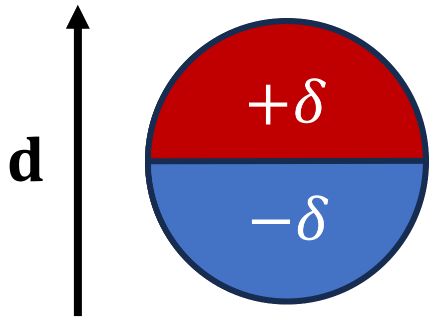
Broader landscape of quantum-controlled molecules for fundamental physics

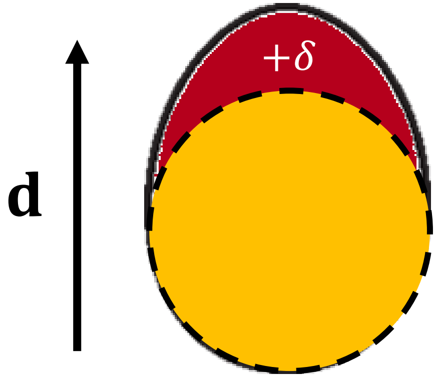


Time spent on each section

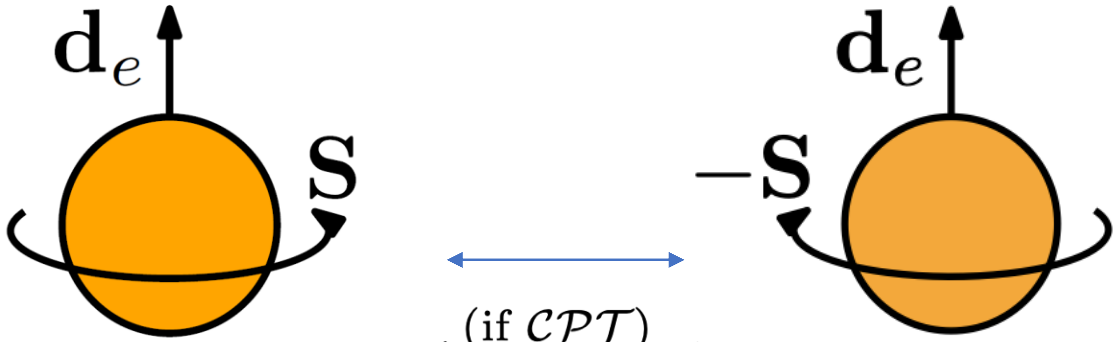
Electric Dipole Moments (EDMs)



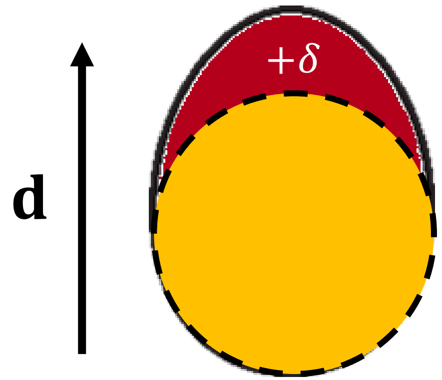




Electric Dipole Moments (EDMs)



$$\mathcal{T} \overset{(\text{if } \mathcal{CPT})}{\equiv} \mathcal{CP}$$



More accurately

For general appreciation of precision

"Egginess": semiclassical analogy of EDMs of elementary particles (and composite systems)

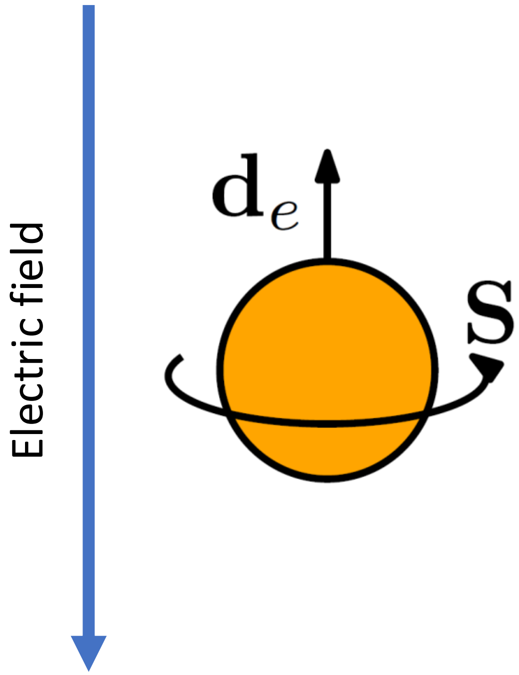
If EDM, then *CP* is not a good symmetry

Radical idea back in the days…

Backdrop (I)

- Pre 1950s
  - Fundamental symmetries, why not? → Elegance, power! / → Conservation laws, selection rules, etc.
- Purcell and Ramsey (1950)
  - Fundamental symmetries, why?
  - Elementary particles could have EDMs, why not?
- 1950s
  - Nature doesn't like fundamental symmetries?
    - Parity violation in weak interactions:
      e.g., Co-60 beta decay, π and μ decays, τ-θ puzzle.

Measuring EDM: spin precession experiment



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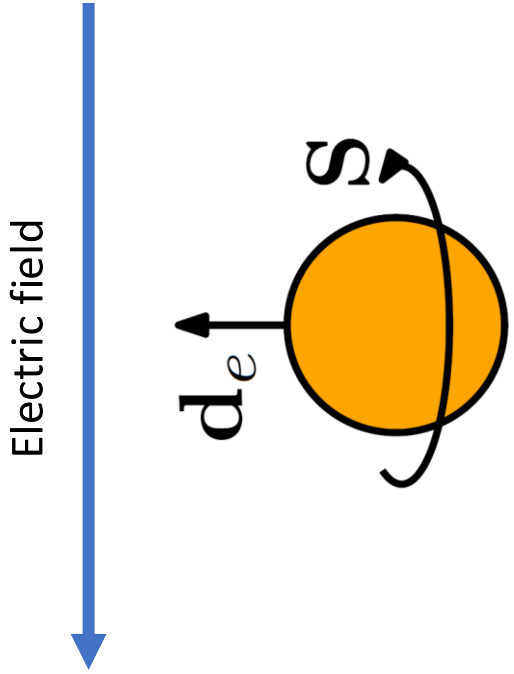
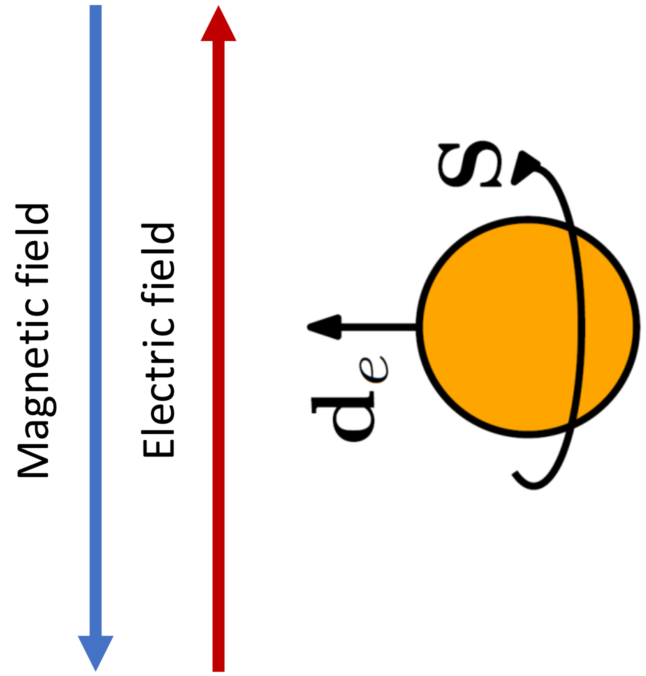


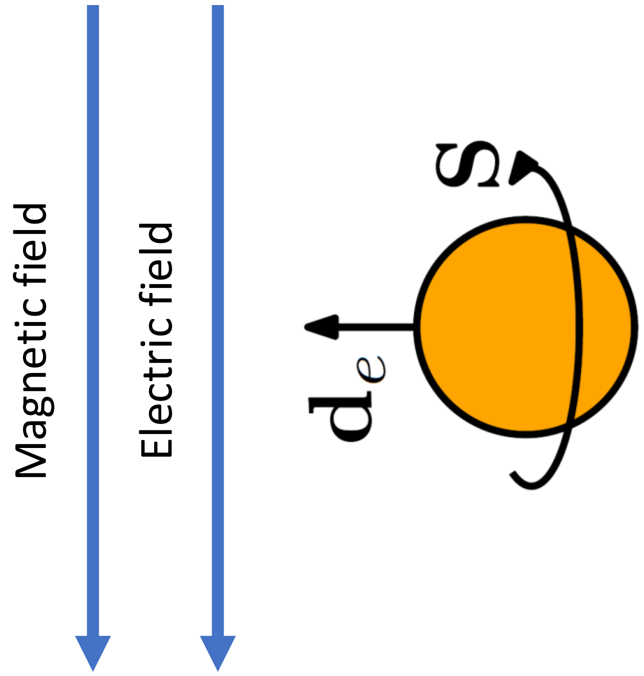
Precession rate proportional to gravity and how fast it spins on its axis.

Precession rate proportional to electric field strength and EDM.

Measuring EDM: spin precession experiment



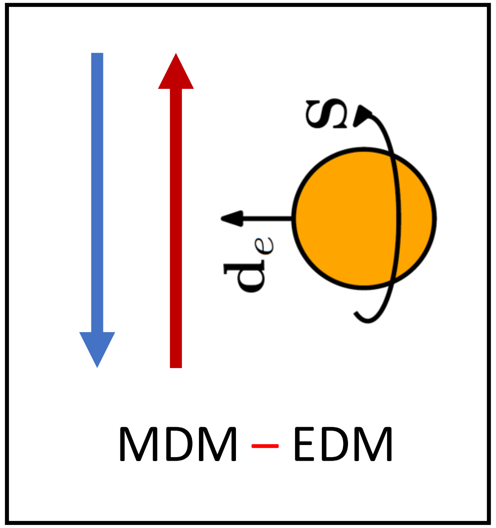
Precession = MDM – EDM

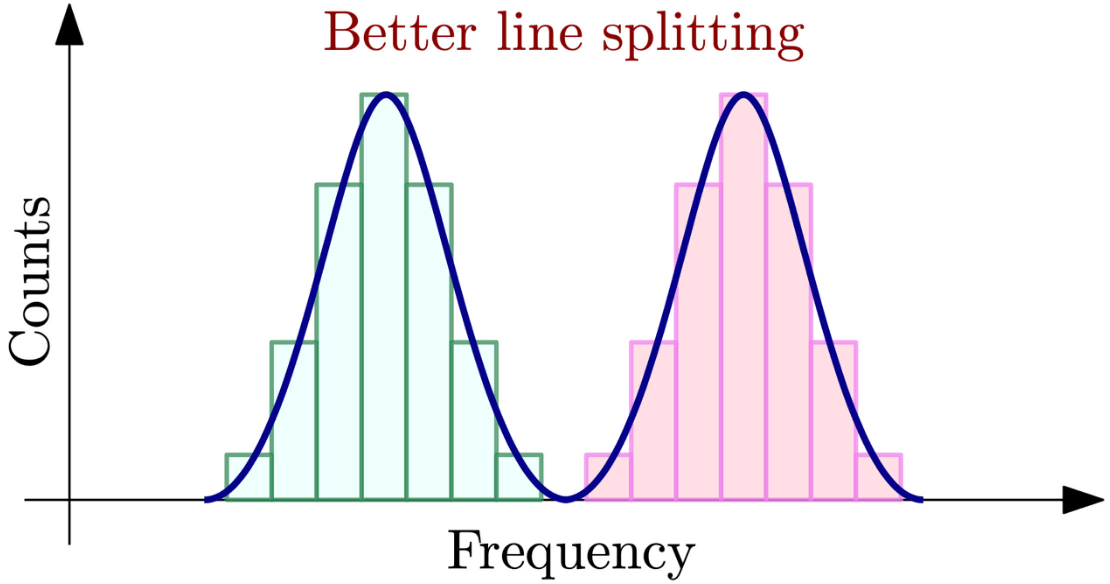


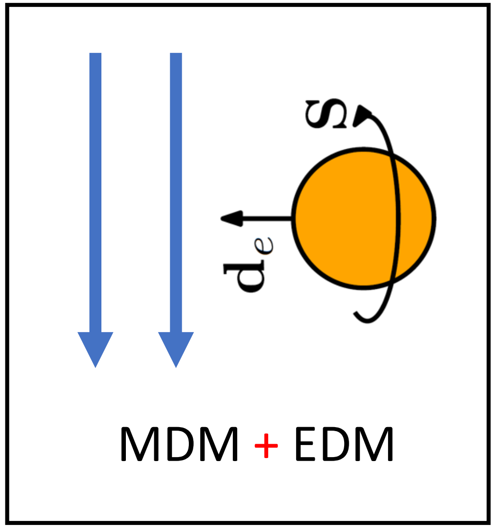
Precession = MDM + EDM

Measuring zero is hard, easier to measure difference between two non-zeroes.

Measuring EDM: spin precession experiment







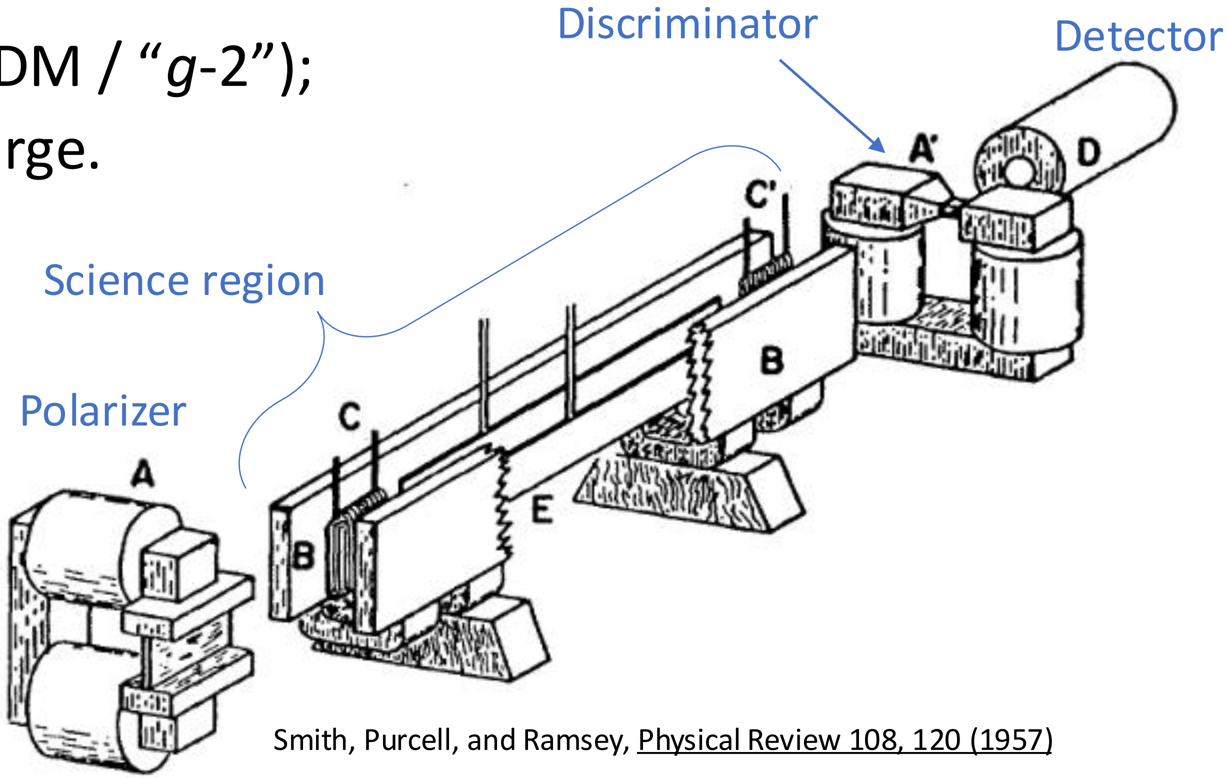
For precise measurement, we want:

- Large electric field ($\mathcal{E}_{\text{eff}}$)
- Long probing times (τ)
- Lots of counts ($\sqrt{N}$)

Backdrop (II)

- First EDM experiment (for neutrons)
  - ORNL + Harvard (1957);
  - Spin precession experiment (like MDM / “*g*-2”);
  - Neutrons, because zero electric charge.

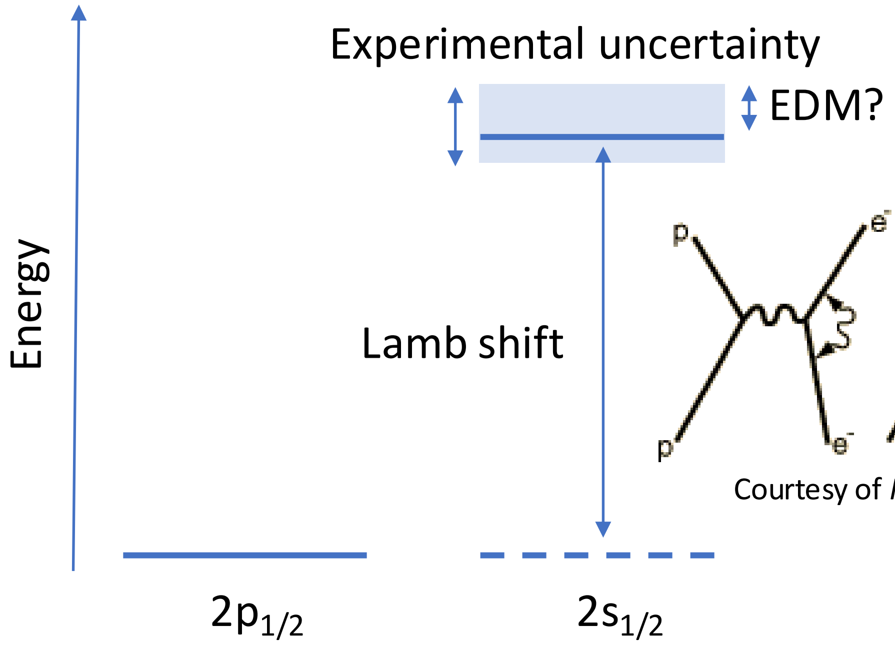
What about charged particles,
e.g., the electron?

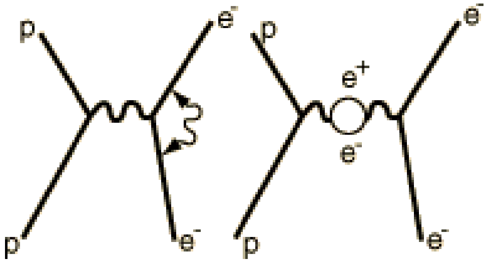


Smith, Purcell, and Ramsey, Physical Review 108, 120 (1957)

First attempts (~1960s)

- Forensics on available atomic data
  - Lamb shift.
  - Feinberg (1958) and Salpeter (1958).

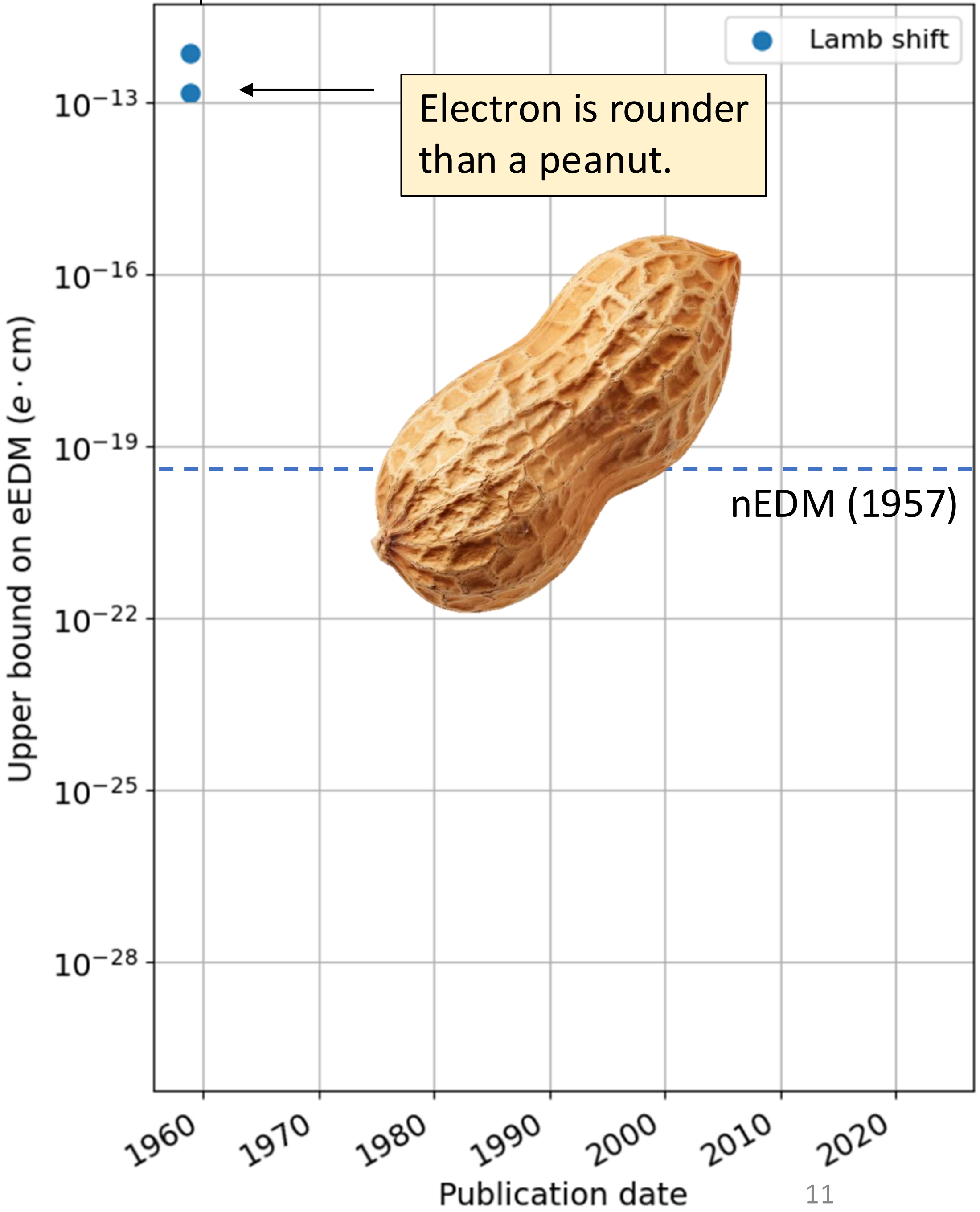




Courtesy of *HyperPhysics*

Schematic diagram of atomic levels in hydrogen

Adapted from Paul Hess's thesis



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  - Free electron spin precession.
  - Nelson et al. (1959).
  - Wilkinson and Crane (1963).

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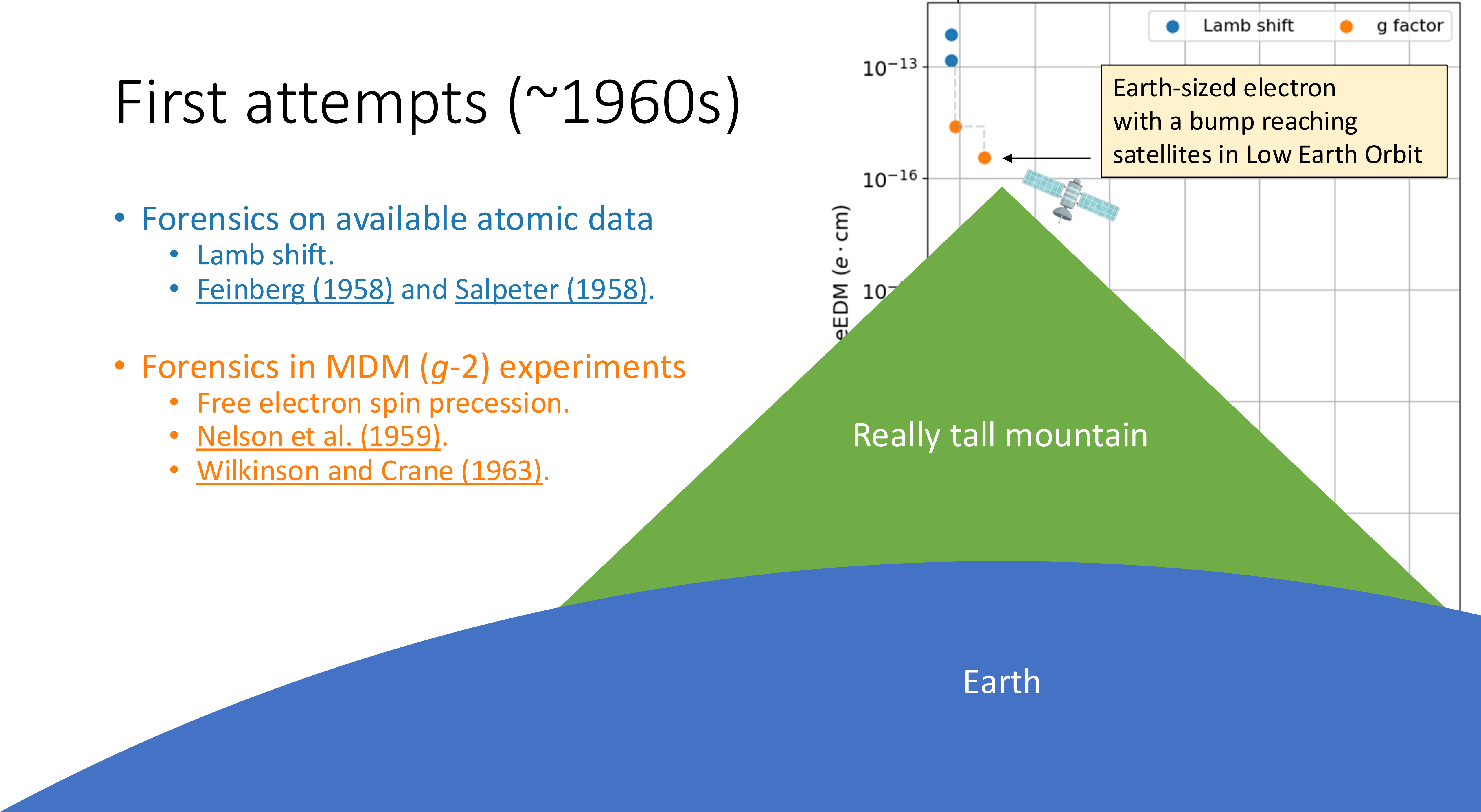

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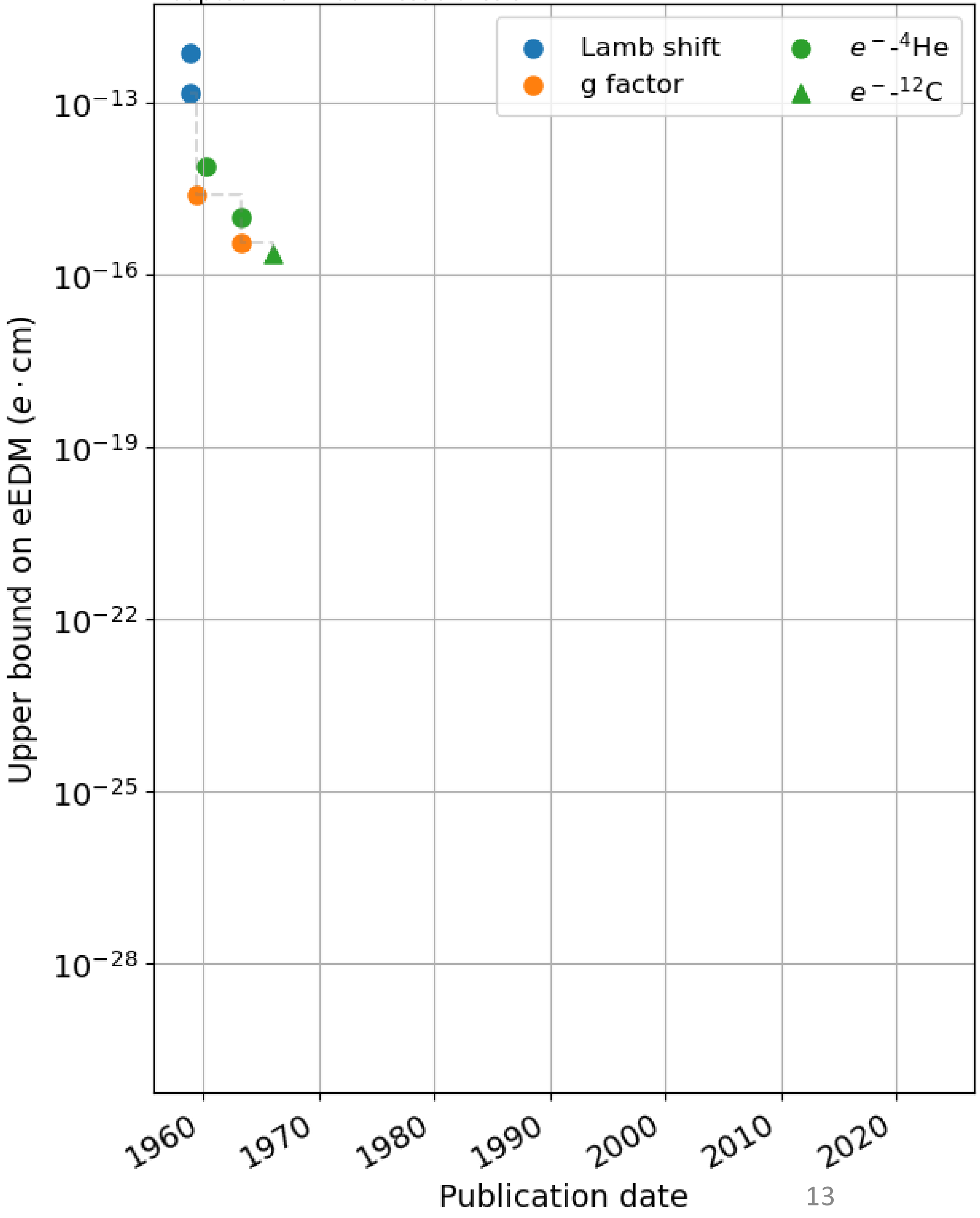
  - Free electron spin precession.
  - Nelson et al. (1959).
  - Wilkinson and Crane (1963).
- Scattering experiments
  - Form factors in differential cross sections.
  - Burleson and Kendall (1960).
  - Goldemberg and Torizuka (1963).
  - Rand (1965).

Adapted from Paul Hess's thesis



Concurrently (~1960s)

Can use same techniques as nEDM (and more)!

- More ideas to measure eEDM
  - Use atoms
    - E.g., Sandars (1965) and (1966)
    - Relativistic enhancement:
      eEDM can induce a much larger atomic EDM!
  - Use polar molecules
    - E.g., Sandars (1967)
    - Polar molecules can have even larger enhancements than atoms!
- Technology: lasers were just invented in early 1960s!

Table I. Upper limits on some electric-dipole moments.

Particle	Electric-dipole moment	Reference
Electron[a]	1.2×10^{-15} cm $\times e$	a
Muon[b]	2×10^{-16} cm $\times e$	b
Neutron[c]	2.4×10^{-20} cm $\times e$	c
Proton[d]	1.3×10^{-13} cm $\times e$	d

Sandars and Lipworth, Phys. Rev. Lett. **13**, 718 (1964)

About to trigger new era of EDM experiments!

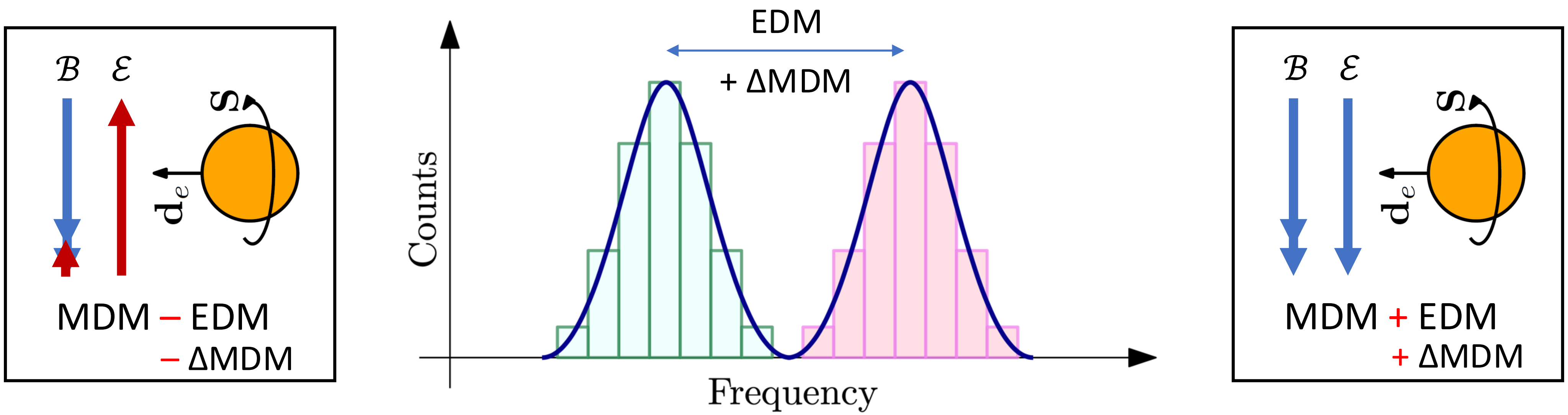
- On CP violation
  - Observed in the K^0 meson (1964).
  - Sakharov's conditions for baryogenesis (1967).
    (we need CP violation for dominance of matter over antimatter)

EDMs no longer just "*why not*", but are *strongly motivated*!

Era of the atoms

- Relativistic enhancements
  - Cesium: $d_{\text{Atom}}/d_e \sim 100$.
  - Thallium: $d_{\text{Atom}}/d_e \sim 600$.

- Systematic effects
  - E.g., spurious magnetic field effect that flips with E field ➔ mimics EDM.



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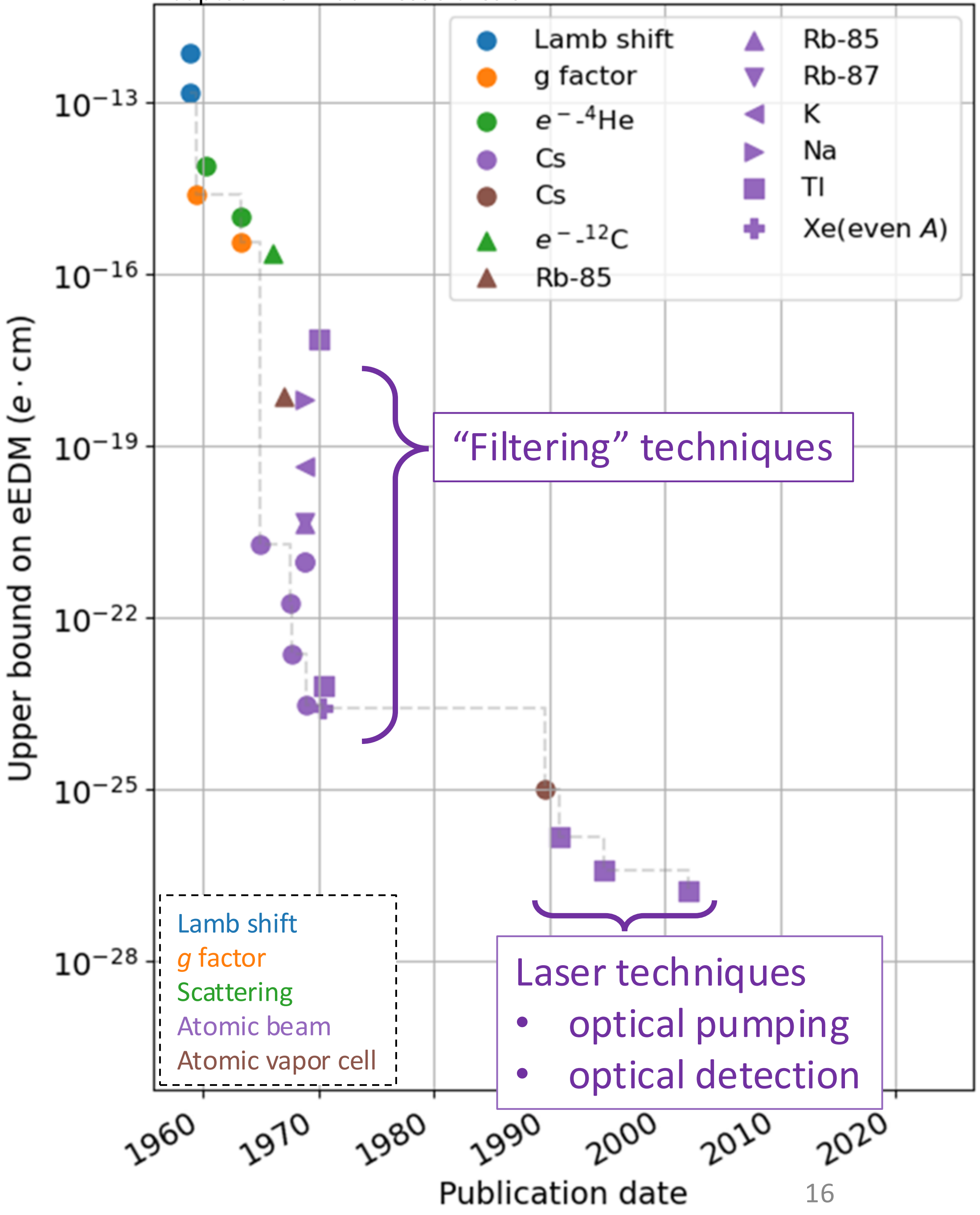
  - Thallium: $d_{\text{Atom}}/d_e \sim 600$.

- Systematic effects
  - E.g., spurious magnetic field effect that flips with E field ➔ mimics EDM.
  - Also bothering nEDM experiments.

- Each upgrade
  - Suppressing systematics
    - Counter-propagating atomic beams;
    - Sodium atoms for comagnetometry.
  - Increase probing time (τ).

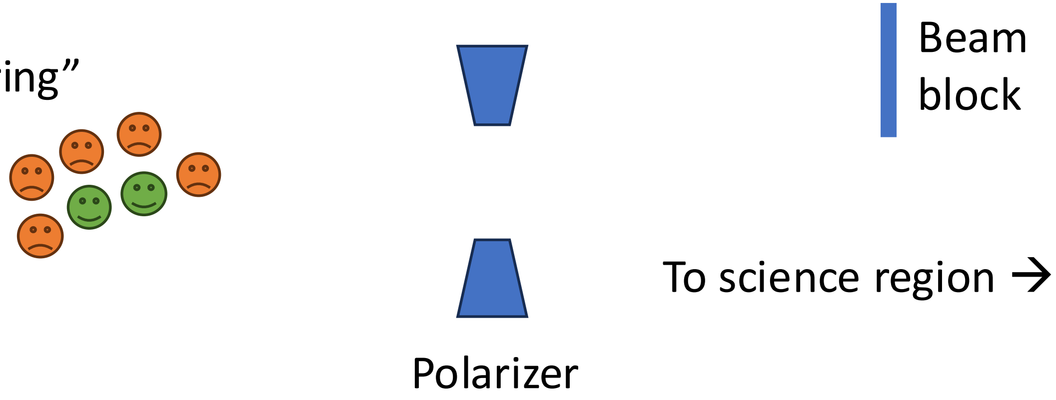
- Lasers greatly improve statistics ($\sqrt{N}$)!

Adapted from Paul Hess's thesis



Era of the atoms

"Filtering"

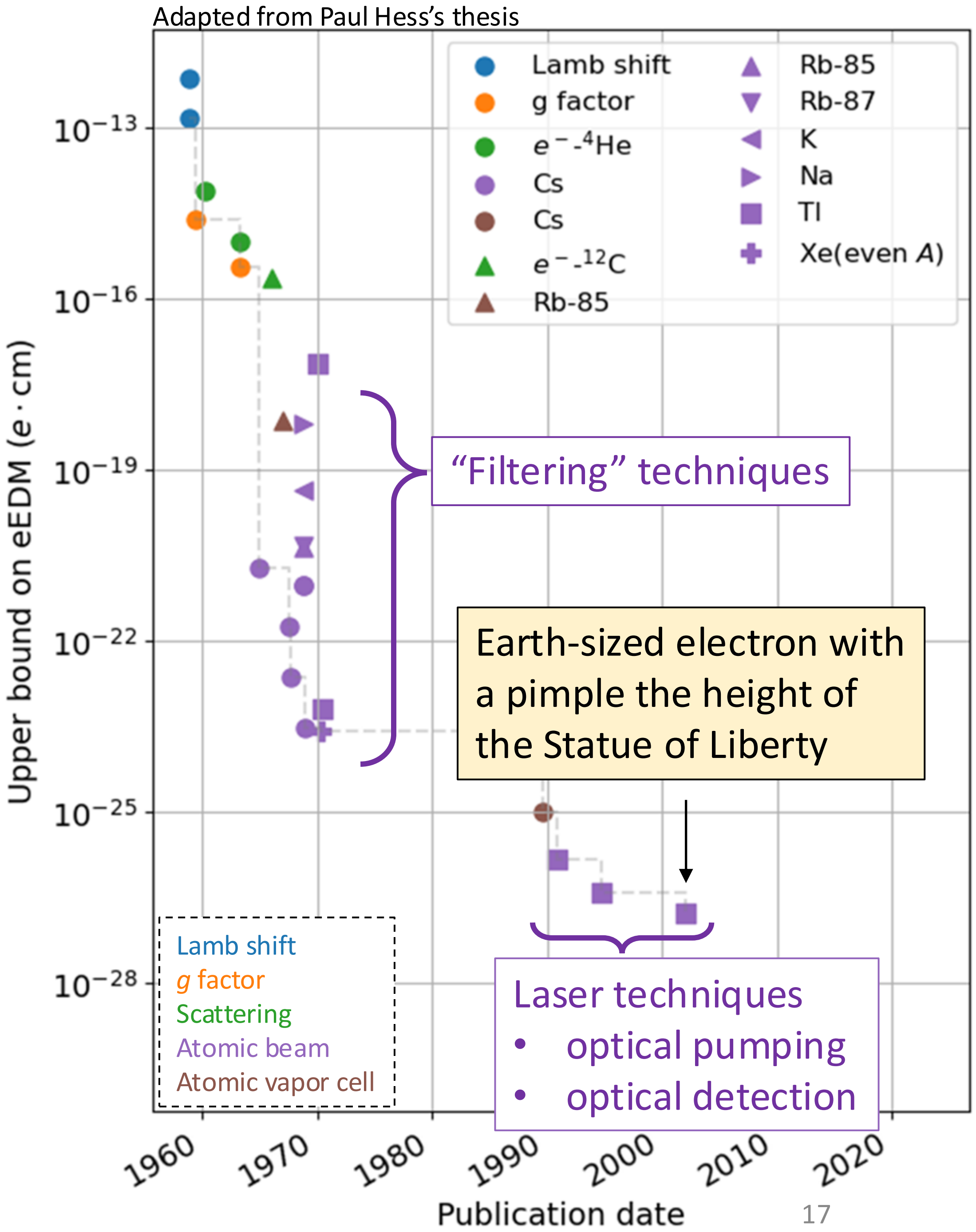


To science region →

Laser techniques

More atoms in EDM state!

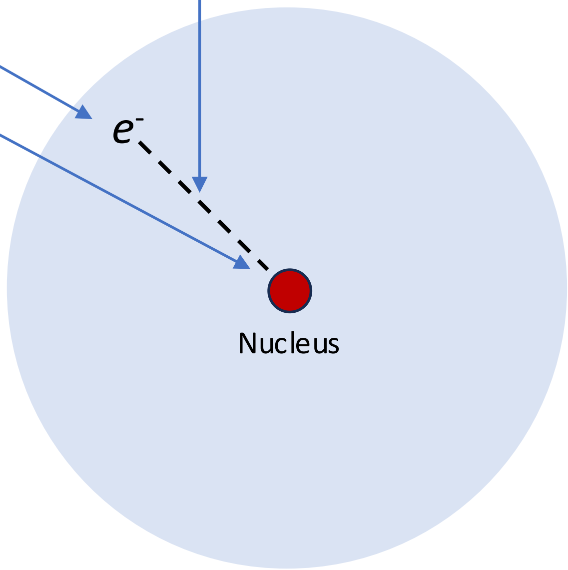
- Lasers greatly improve statistics ($\sqrt{N}$)!



Sidetrack from eEDM

Era of the atoms

- New understanding on multiple sources of atomic EDM (around 1980)
  - Electron's EDM;
  - Electron-nucleon interactions (semileptonic);
  - Nucleon EDM, CP-odd nuclear forces.



- Opens periodic table for more EDM searches.

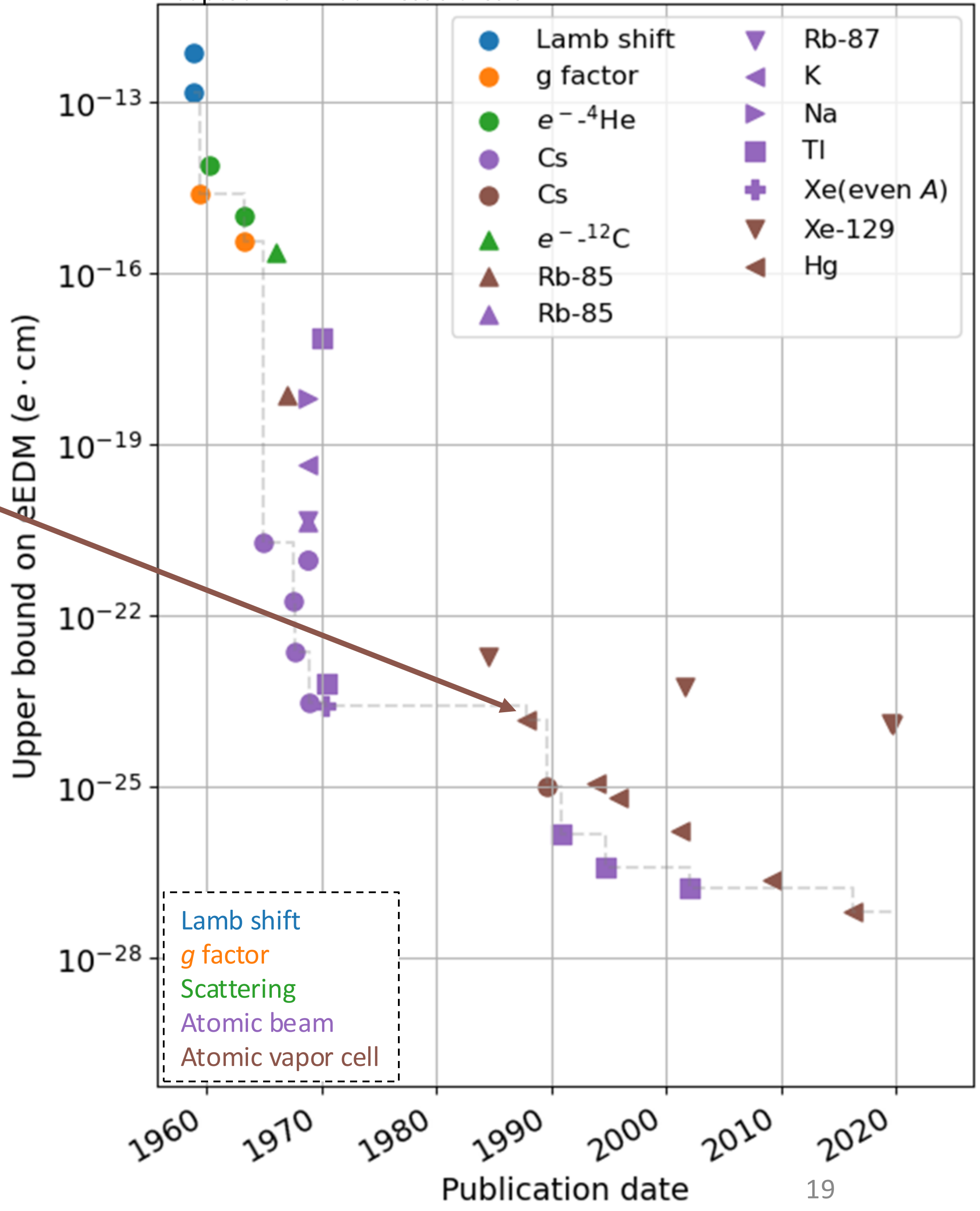
For today:
- Hg EDM
- TlF EDM

Sidetrack from eEDM

Era of the atoms

- Mercury in atomic vapor cell
  - Designed to probe nuclear EDM…
  - … managed to set record on eEDM anyway.
  - Optical pumping and detection.
  - Energy resolution @ $\delta E \sim 10\ \mathrm{pHz} \sim 10^{-26}\ \mathrm{eV}$ in 2016 result.
  - Blinded analysis (from 2009).
    - Suppress experimental bias e.g., when to stop data collection.

Adapted from Paul Hess's thesis

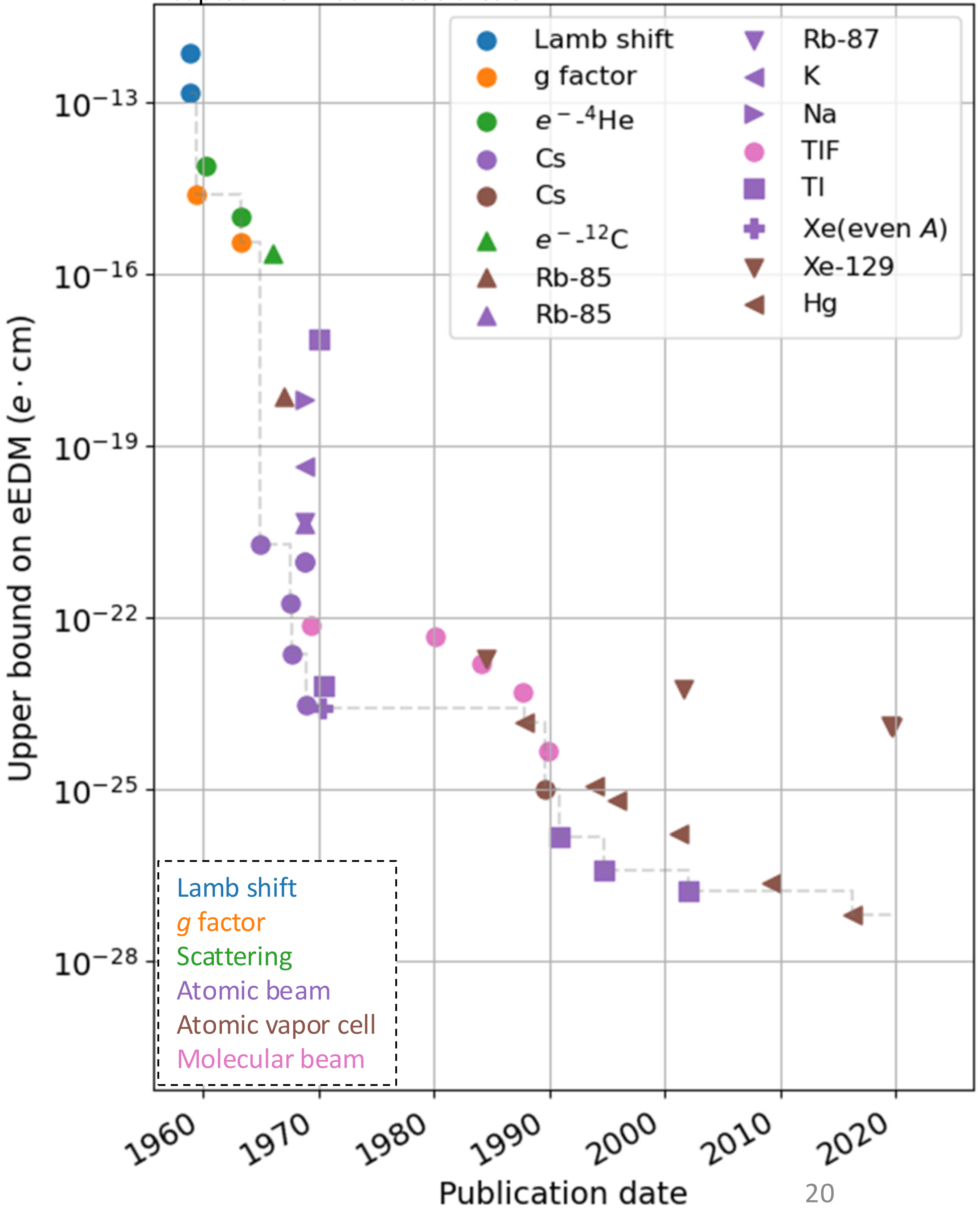


Sidetrack from eEDM

First attempts with polar molecules

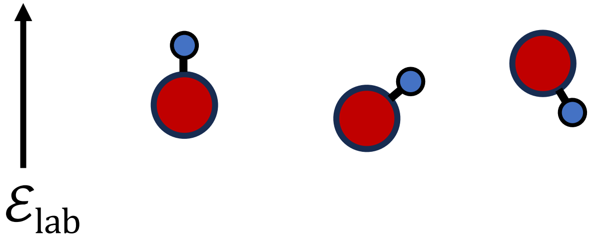
- Thallium monofluoride (^{205}TlF)
  - ^{205}Tl has 81 protons and 124 neutrons
    ➔ unpaired proton
    ➔ measure proton EDM!
  - eEDM comes in at higher order
    - d_p(TlF) ~ 260 d_e(TlF).

Adapted from Paul Hess's thesis



Era of the molecules

- Rich internal structures
  - Highly insensitive to magnetic fields
    ➔ Suppress magnetic-field systematics.
  - Comagnetometry with the same molecule
    ➔ Suppress magnetic-field systematics.
  - Easier to align molecules to lab frame
    ➔ Signal adds up instead of washing out.



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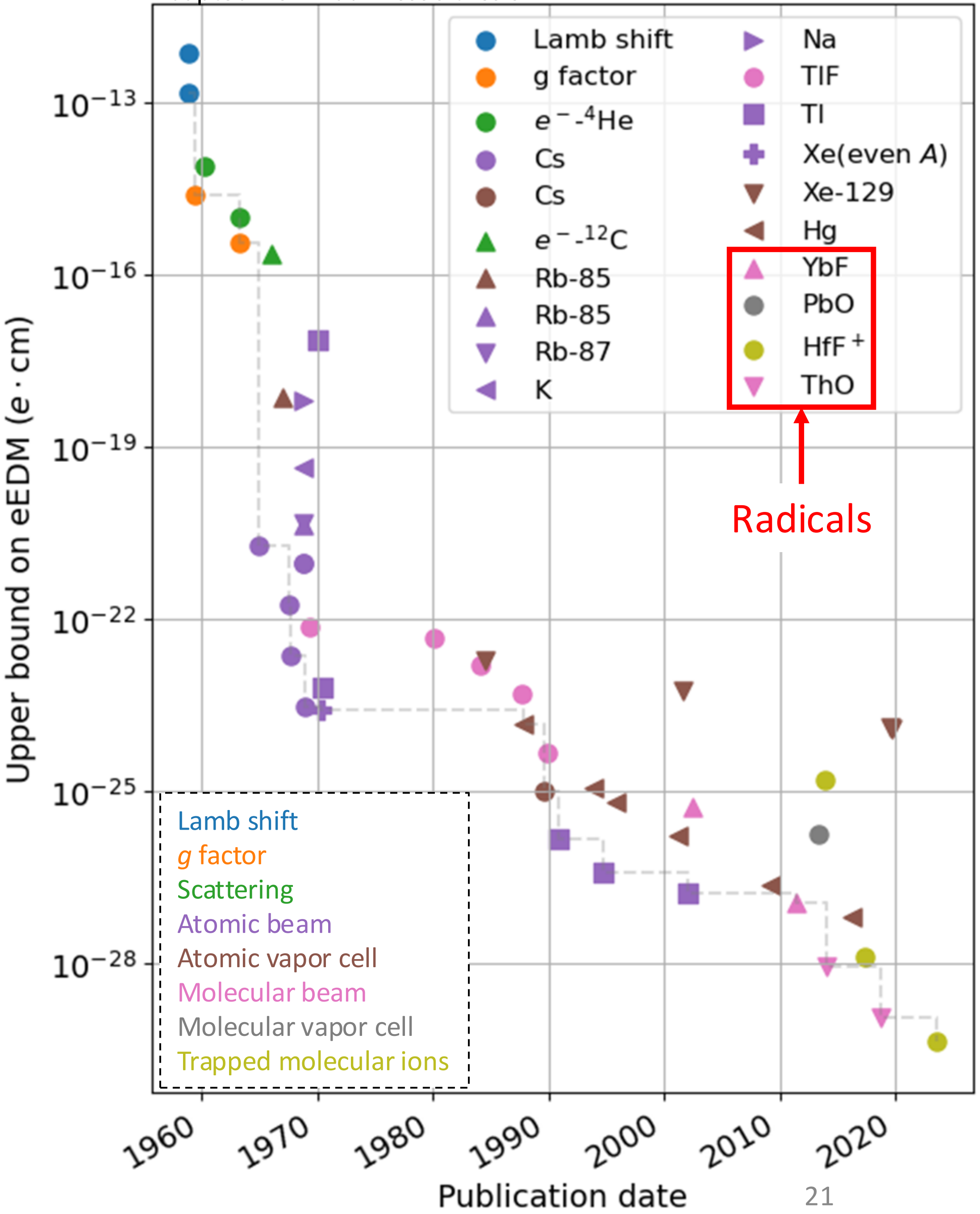
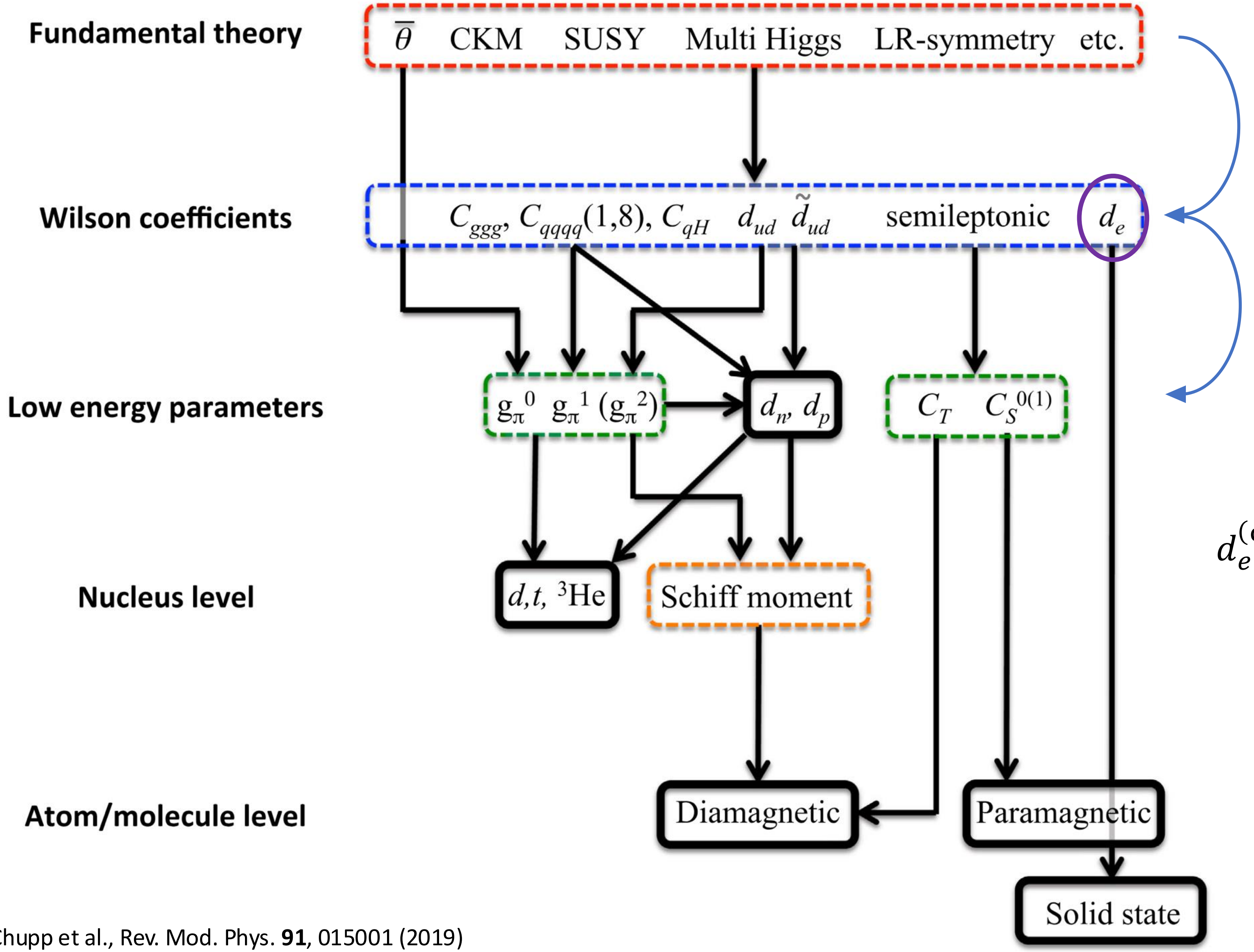




Integrating out degrees of freedom above relevant energy scales.

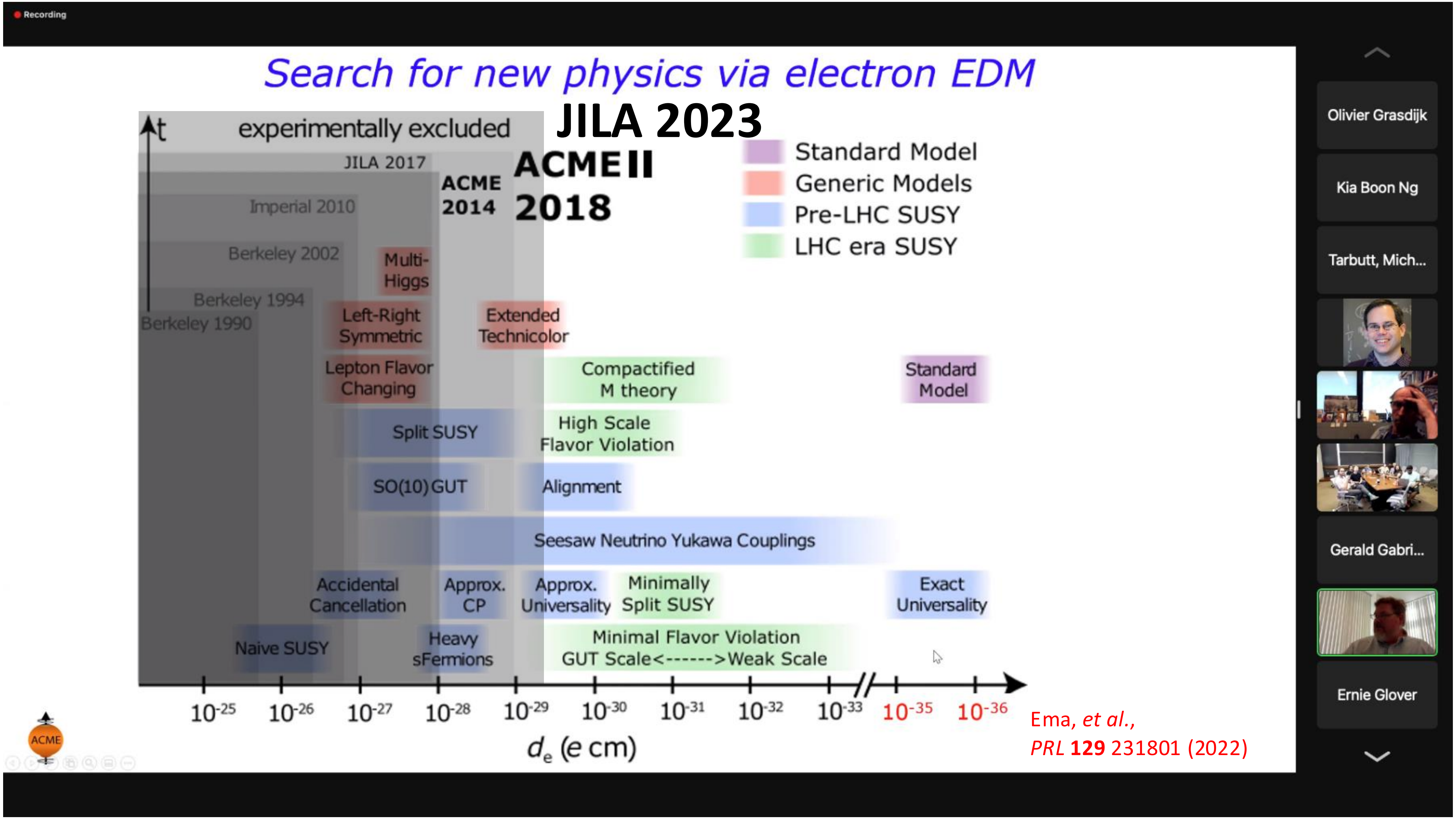
$$d_e^{(\text{one loop})} \sim \frac{e\hbar}{m_e c}\left(\frac{m_e}{M}\right)^2 g^2 \sin(\phi_{CP})$$

CP violating phase

Coupling strength

Mass of new particle

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

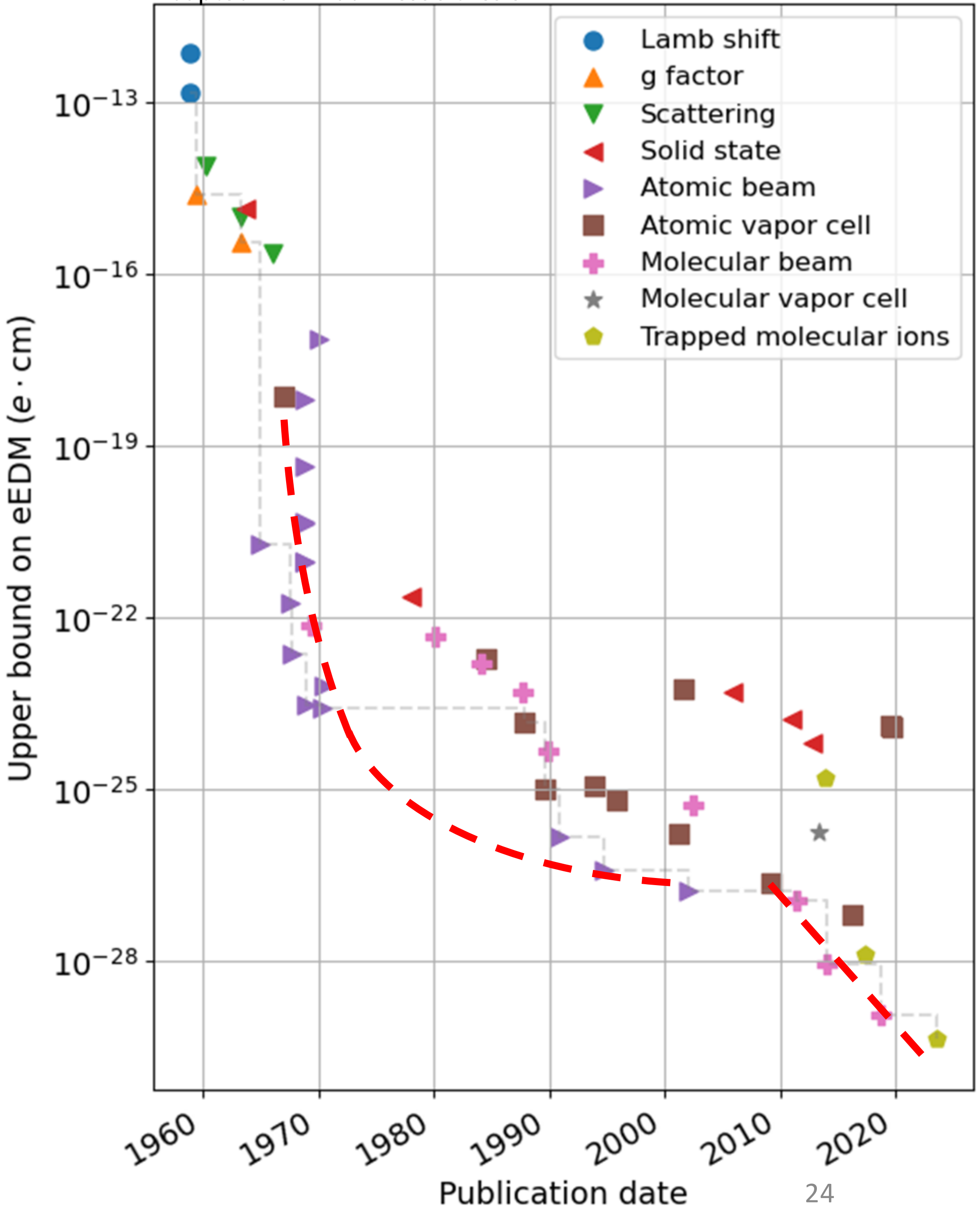


Courtesy of ACME, Sloan-Moore Annual eEDM Meeting (Sep 2022) start to the

What a great journey!

- eEDM through modern eyes
  - EDM violates CP
    ➔ good probe for CP-V new physics!
- Richness of atomic/molecular structure:
  - Relativistic effects boost sensitivity!
  - Powerful tool for quantum control and rejection of systematics!
  - Collaboration: theorists & experimentalists
    - Disentangle CP-V sources.
    - Identify better atoms/molecules and experimental techniques.

Adapted from Paul Hess's thesis



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  - Current status (JILA Gen. II, Nov 2022).
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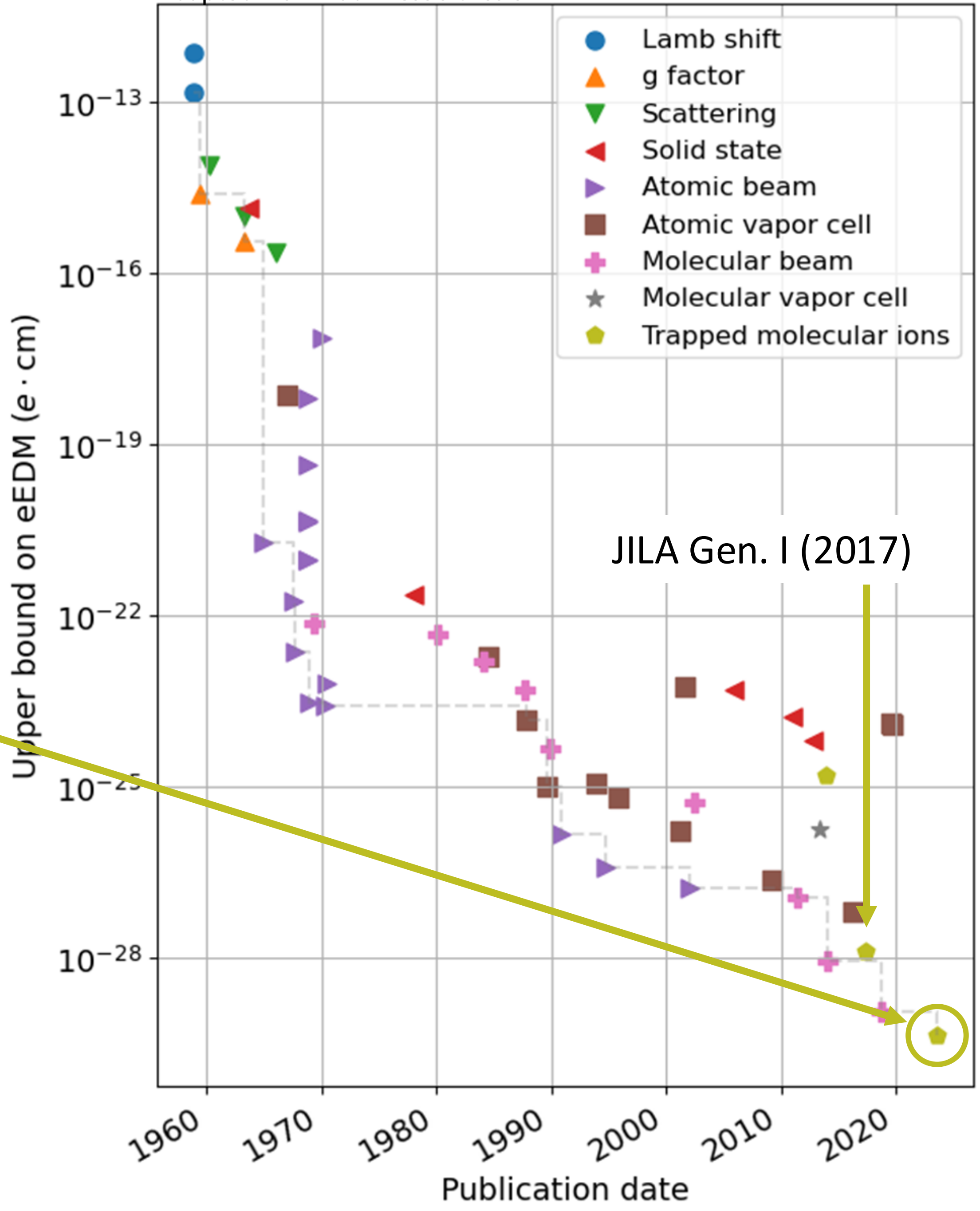
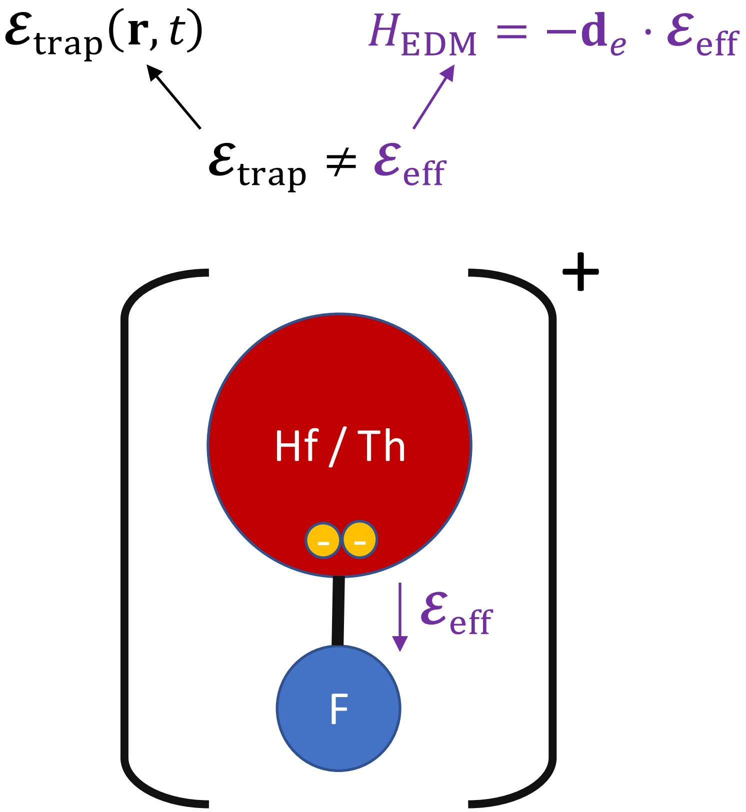
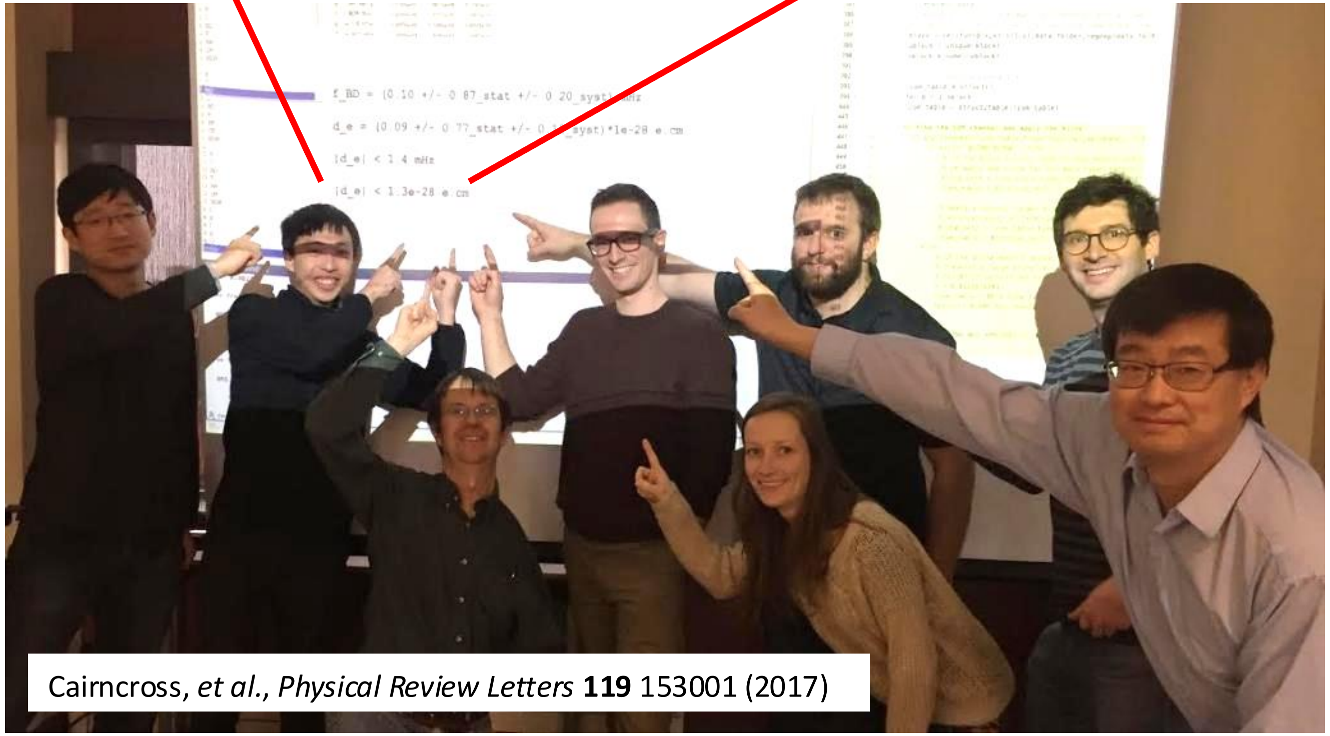


About HfF^+ (and ThF^+)

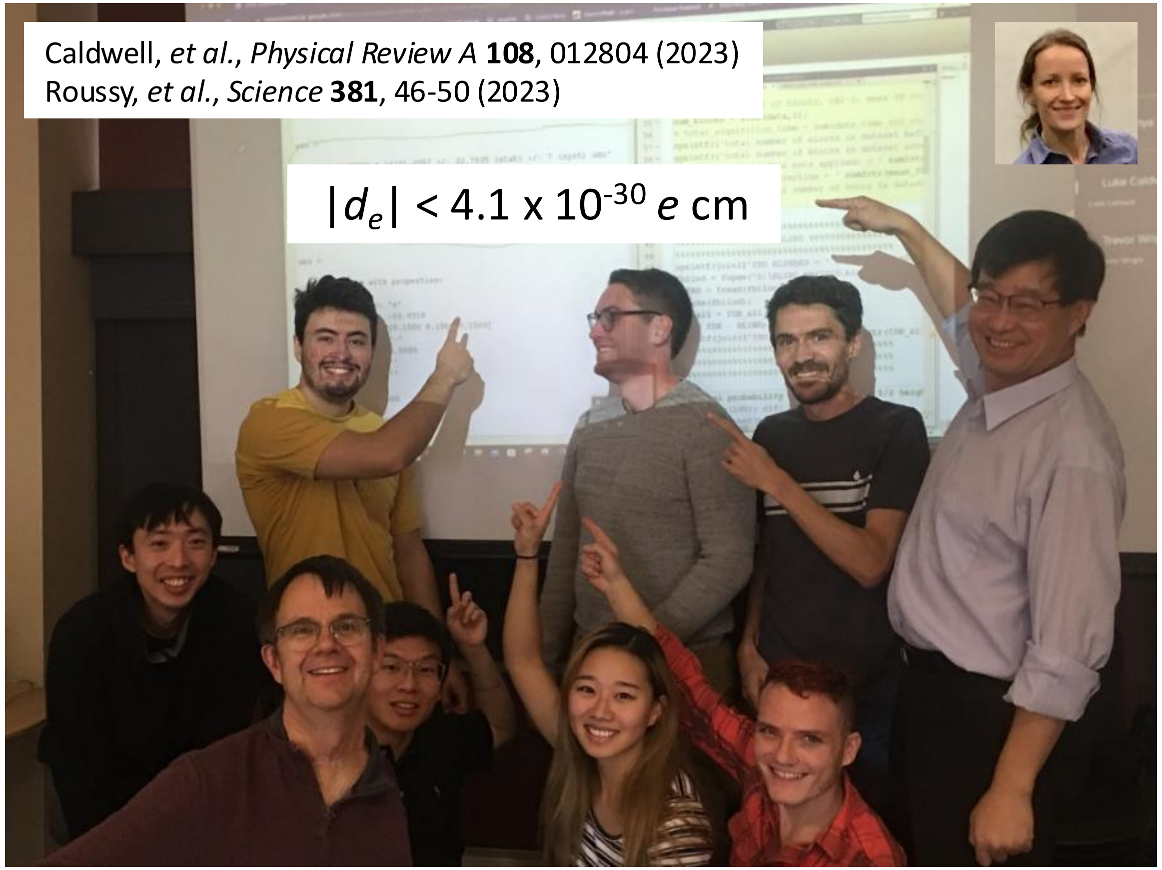
- Heavy atom for relativistic enhancement
  - Hf and Th.
- Electronegative atom for molecular enhancement
  - F.
- Ionize molecule
  - Electronic configuration for EDM measurement.
  - Confine molecular ion in ion trap.



JILA eEDM with HfF^+ (Gen. I 2017, Gen. II 2022)



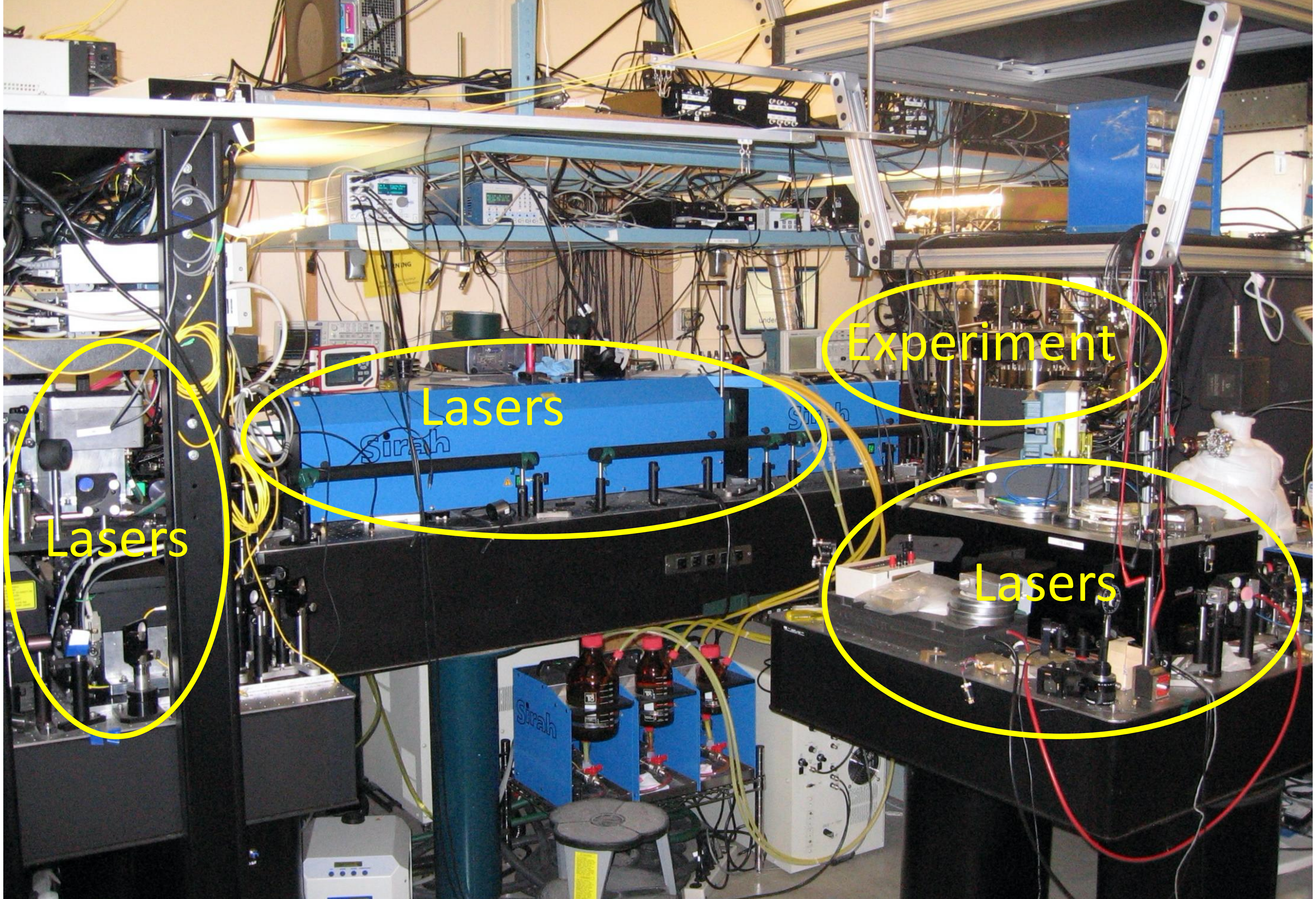
Cairncross, *et al.*, *Physical Review Letters* **119** 153001 (2017)



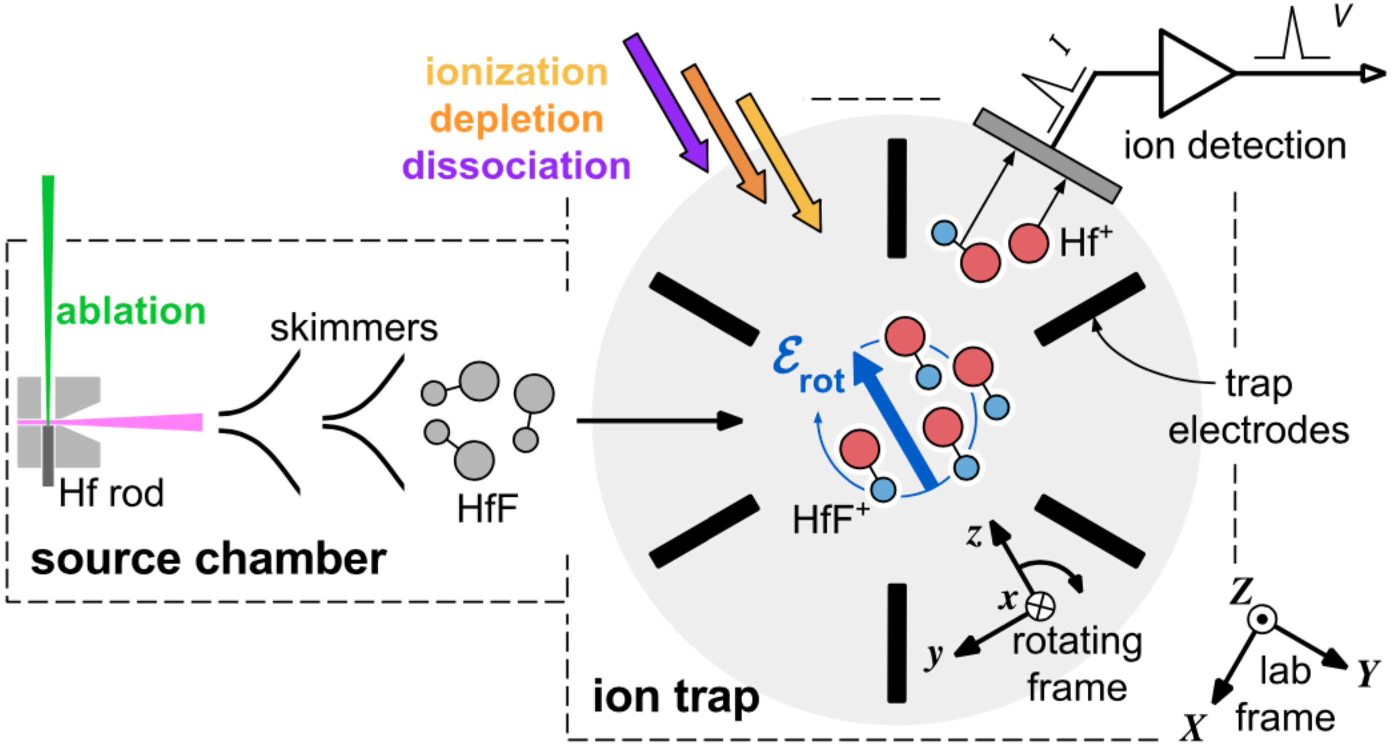
Caldwell, *et al.*, *Physical Review A* **108**, 012804 (2023)
Roussy, *et al.*, *Science* **381**, 46-50 (2023)

$|d_e| < 4.1 \times 10^{-30}\ e$ cm

Equivalent to measuring the Earth's roundness with a precision of the length of a school bus!


Experiment
Lasers
Lasers
Lasers

JILA eEDM



Cairncross, *et al.*, *Physical Review Letters* **119** 153001 (2017)

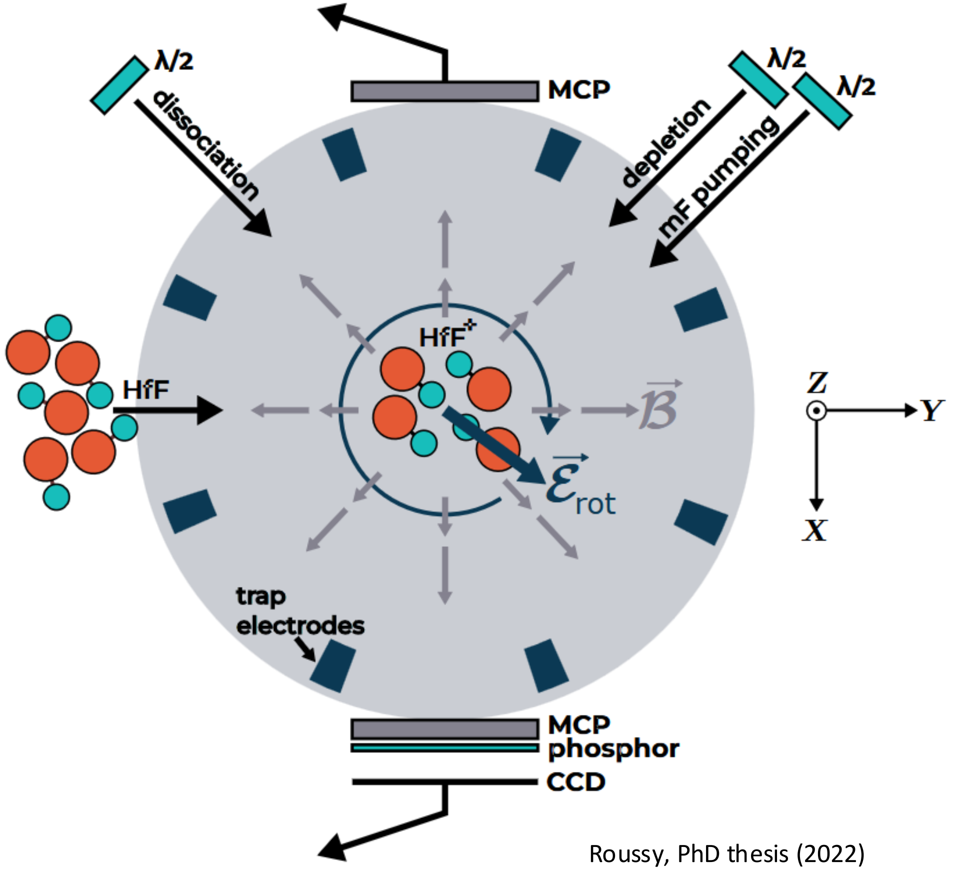
JILA I to JILA II:

- Ion trap upgrade.
- More efficient state preparation.
- Novel technique: two-state detection.

Two-state detection: the idea

Two-state detection: the reason

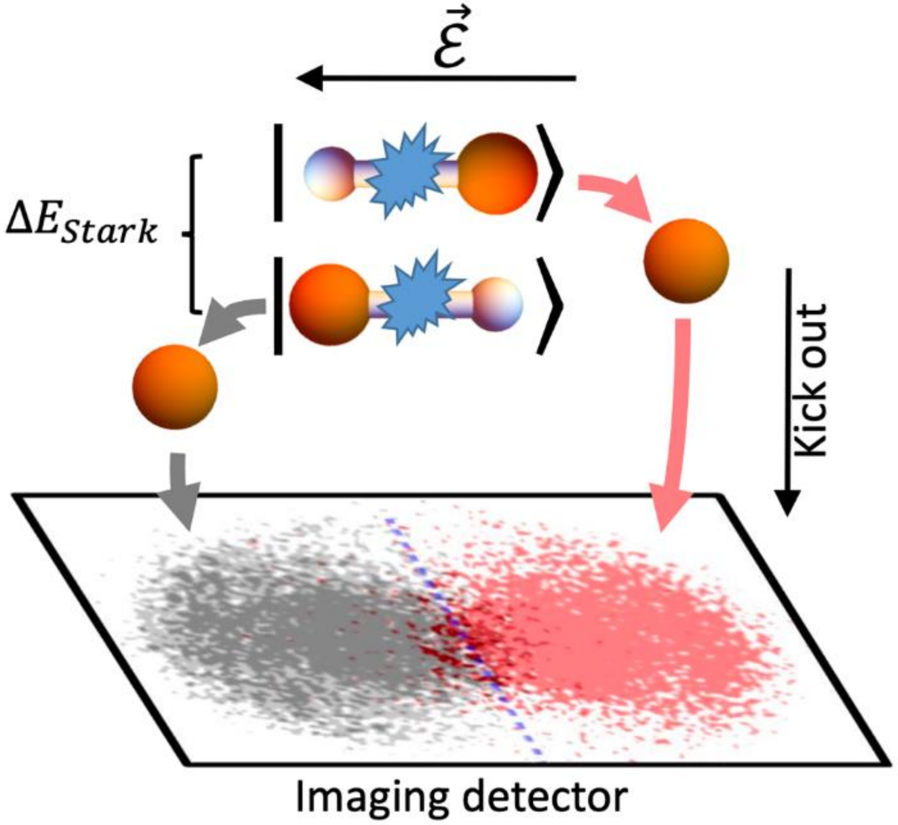
- Measuring a very small (EDM) frequency
  - Hard!
- Signal can be polluted by
  - Noise (statistics);
  - Unintended effects (systematics).
- Beneficial to do experiment in slightly different ways for checks
  - E.g., molecules oriented one way and another.



Roussy, PhD thesis (2022)

Two-state detection: the paper

- Dissociate molecules.
- Kick ions towards ion detector.
- Draw a calibrated line.
- Associate ions on one side with one state.
- Demonstrated with HfF^+ and ThF^+.



Zhou, *et al.*, *Physical Review Letter* **124** 053201 (2020)

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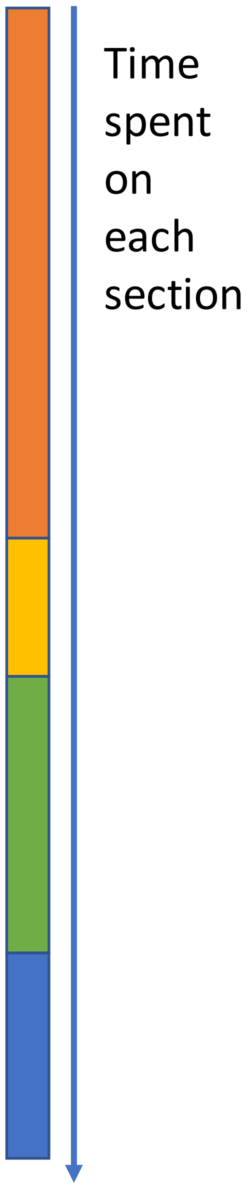
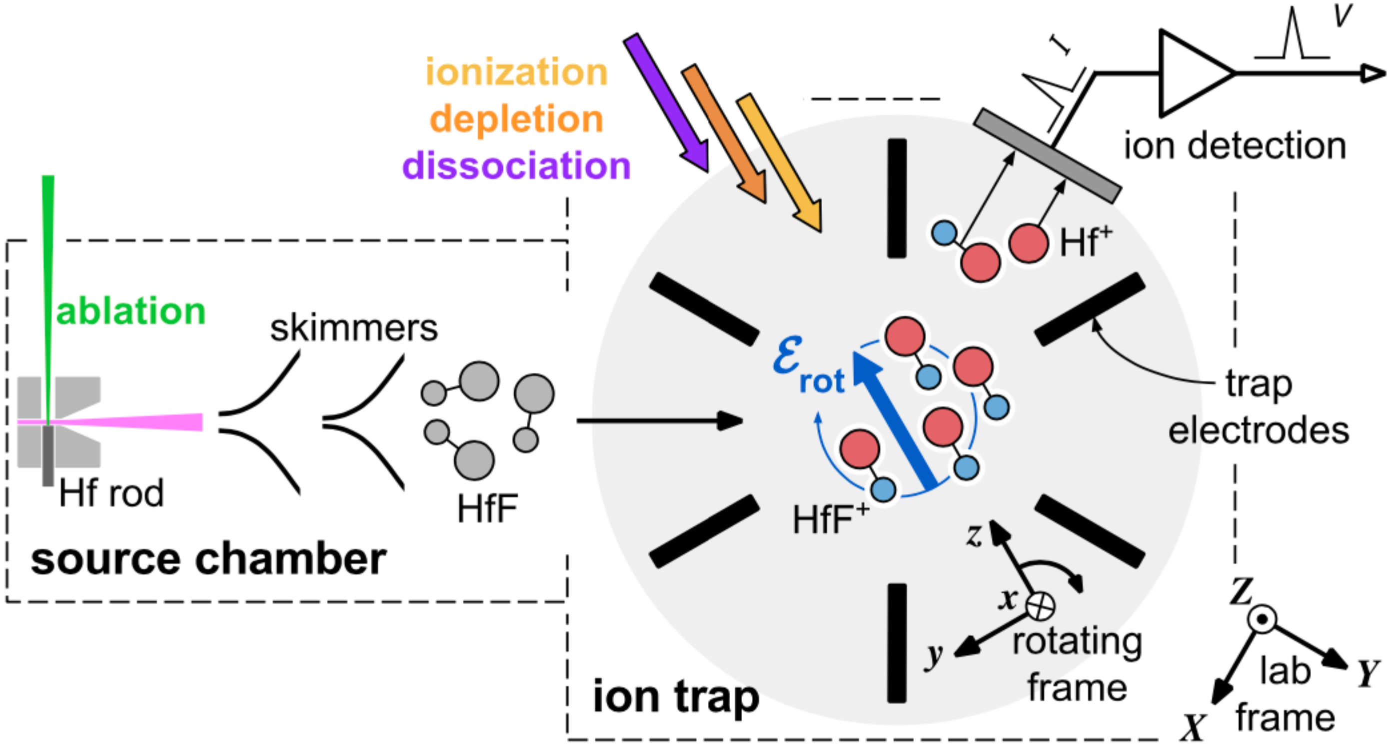
Broader landscape of quantum-controlled molecules for fundamental physics



JILA eEDM



Cairncross, *et al.*, *Physical Review Letters* **119** 153001 (2017)

JILA I to JILA II:
- Ion trap upgrade.
- More efficient state preparation.
- Novel technique: two-state detection.

JILA II to JILA III:
- Better molecule (ThF^+).
- Multiplexing.

Why ThF^+?

Spin precession rate proportional to electric field strength and eEDM.

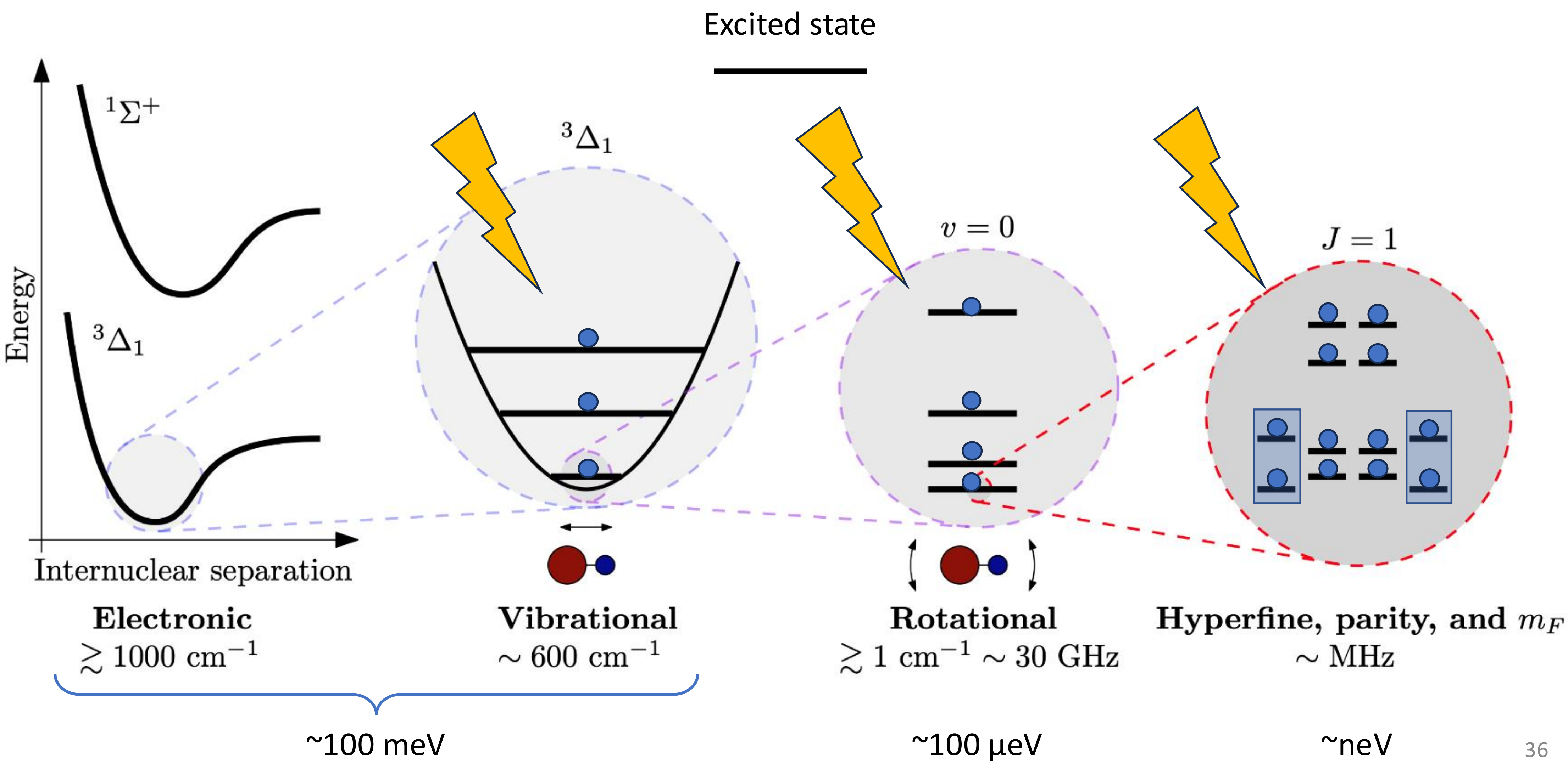
	ThF^+	HfF^+
Effective *E*-field	35.2 GV/cm	23.4 GV/cm
eEDM-sensitive state	(Ground state) 0 cm^{-1}	(Excited state) 977 cm^{-1}
Expected Coherence	~10 s	2 s

This restricts this

Higher sensitivity to new physics + More precise measurements = Better molecule!

Denis, et al., *New Journal of Physics* 17 (2015): 043005
Skripnikov, et al., *Physical Review A* 91 (2015): 042504
Petrov, et al., *Physical Review A* 76 (2007): 030501
Gresh et al., *Journal of Molecular Spectroscopy* 319, 1-9 (2016).
Cossel et al., *Chemical Physics Letters* 546, 1-11 (2012)

Molecules are complicated

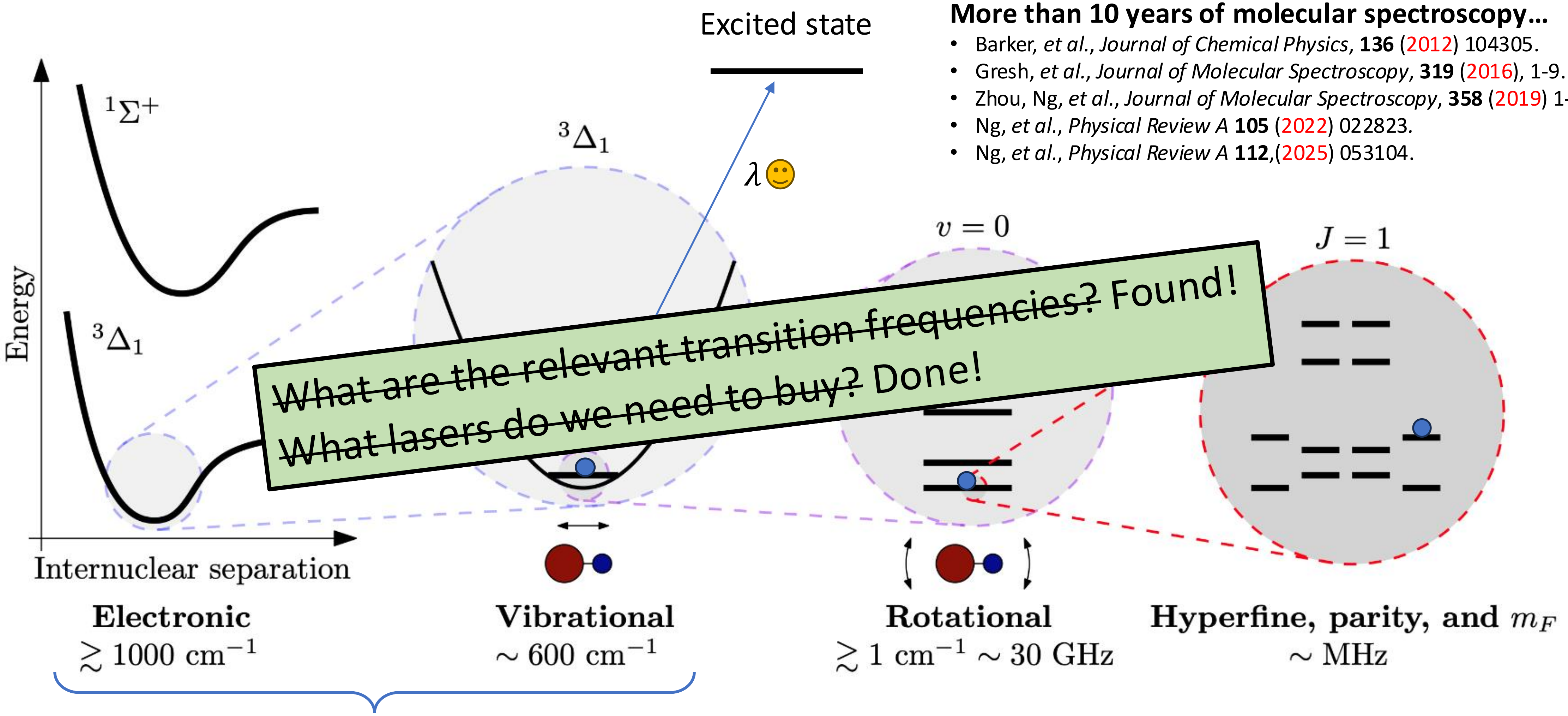


Molecules are complicated

Atomic transitions: NIST ✓
Nuclear energy levels: ENSDF ✓
Molecular levels: ???

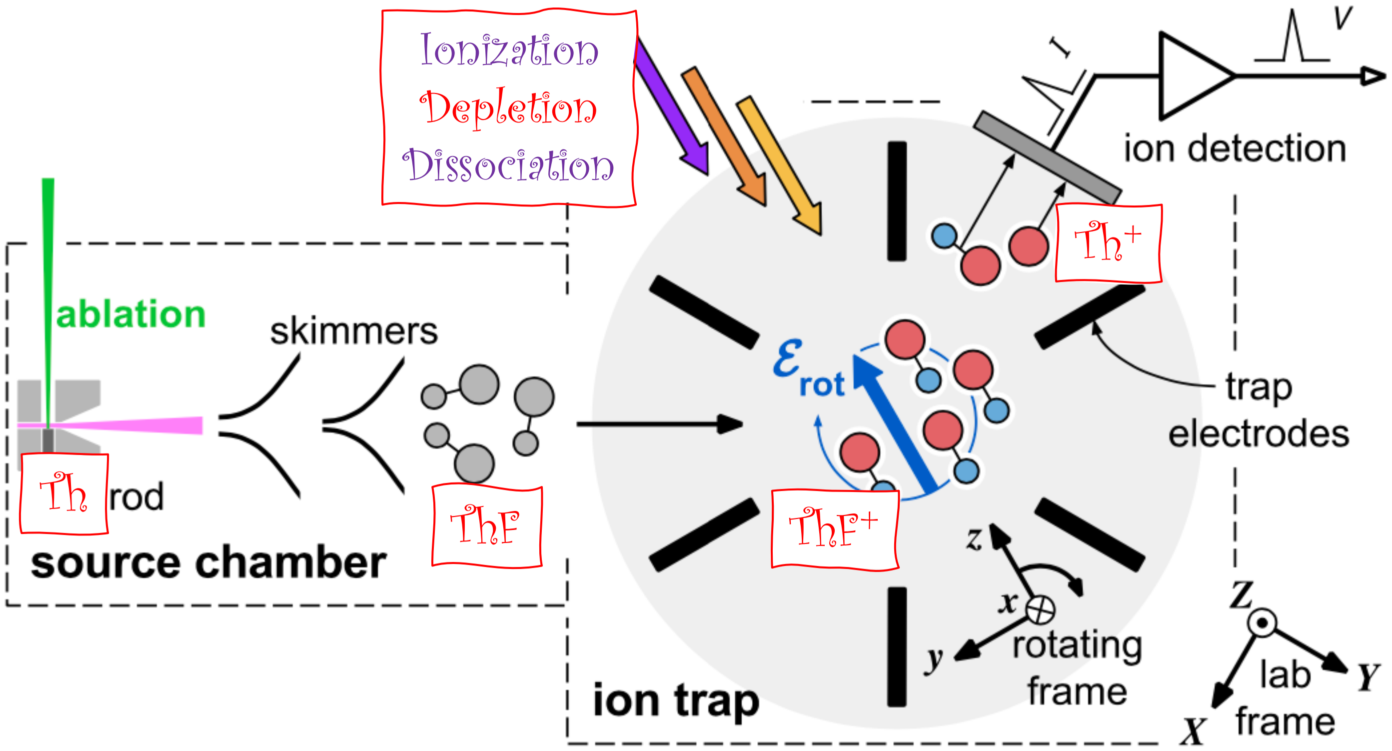
More than 10 years of molecular spectroscopy…

- Barker, *et al.*, *Journal of Chemical Physics*, **136** (2012) 104305.
- Gresh, *et al.*, *Journal of Molecular Spectroscopy*, **319** (2016), 1-9.
- Zhou, Ng, *et al.*, *Journal of Molecular Spectroscopy*, **358** (2019) 1-16.
- Ng, *et al.*, *Physical Review A* **105** (2022) 022823.
- Ng, *et al.*, *Physical Review A* **112**,(2025) 053104.



~~What are the relevant transition frequencies?~~ Found!
~~What lasers do we need to buy?~~ Done!

EDM experiment with ThF⁺



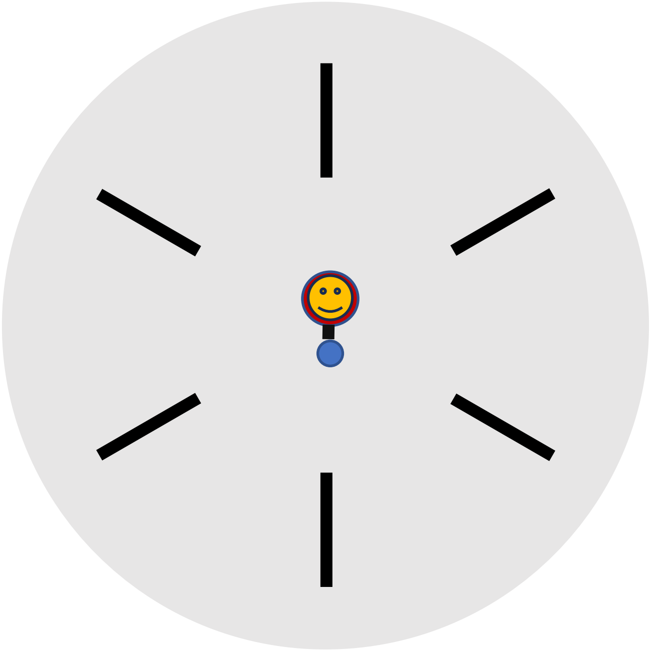
Cairncross, et al. *Physical review letters* 119.15 (2017): 153001.

EDM with ThF^+: inefficiency

State preparation
- ~ 100 ms

Spin precession
- ~ 10 s

State readout
- ~ 100 ms



EDM with ThF^+: multiplexing

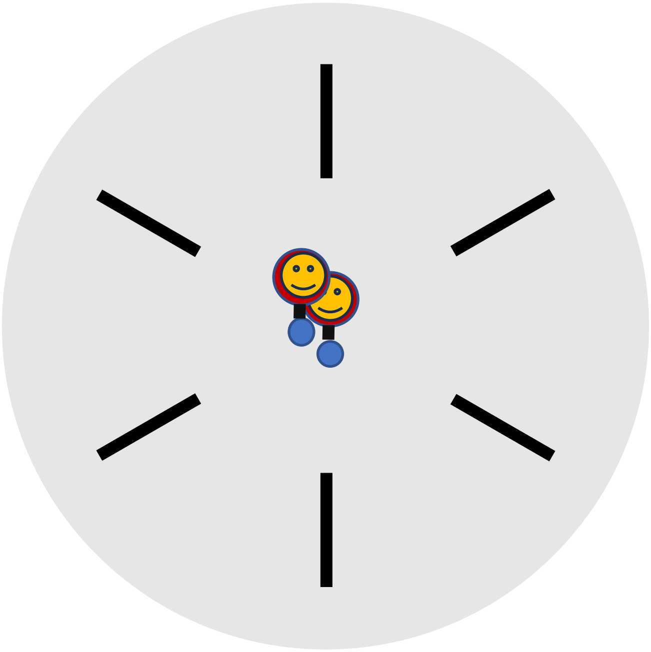
Greatly increasing repetition rate!

$f_{rep.} \sim \frac{1}{10\ \mathrm{s}}$ → $f_{rep.} \sim \frac{1}{100\ \mathrm{ms}}$

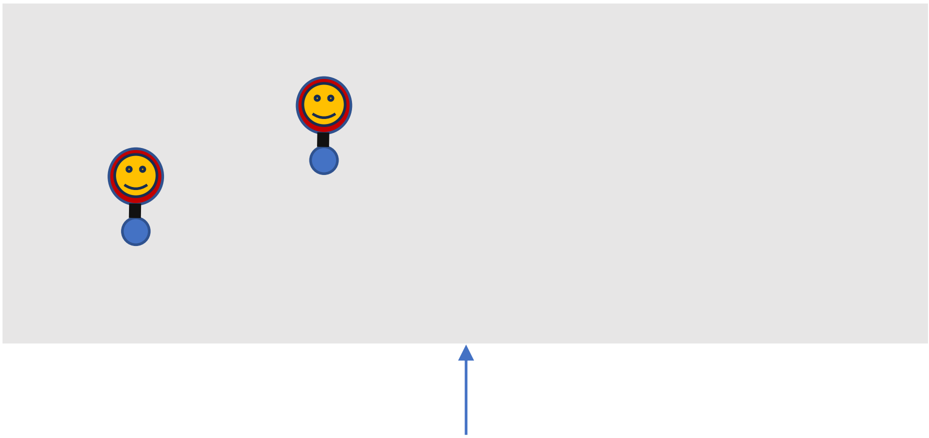
State preparation
- ~ 100 ms

Spin precession
- ~ 10 s

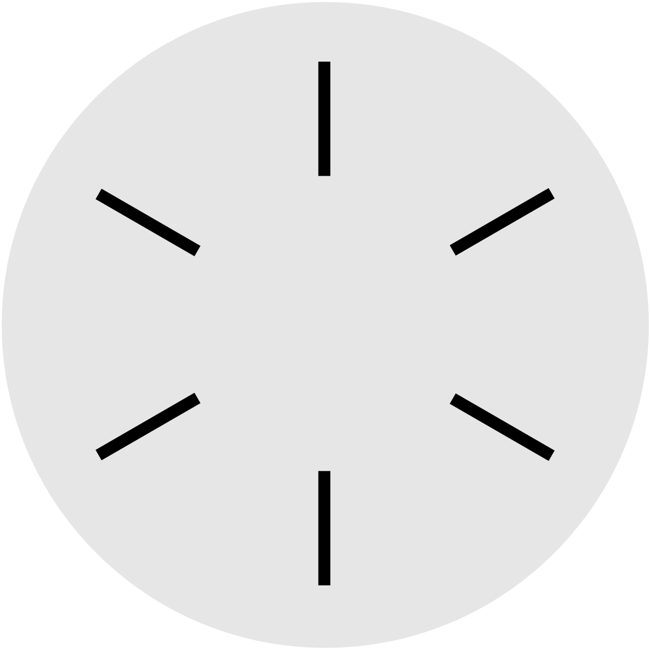
State readout
- ~ 100 ms

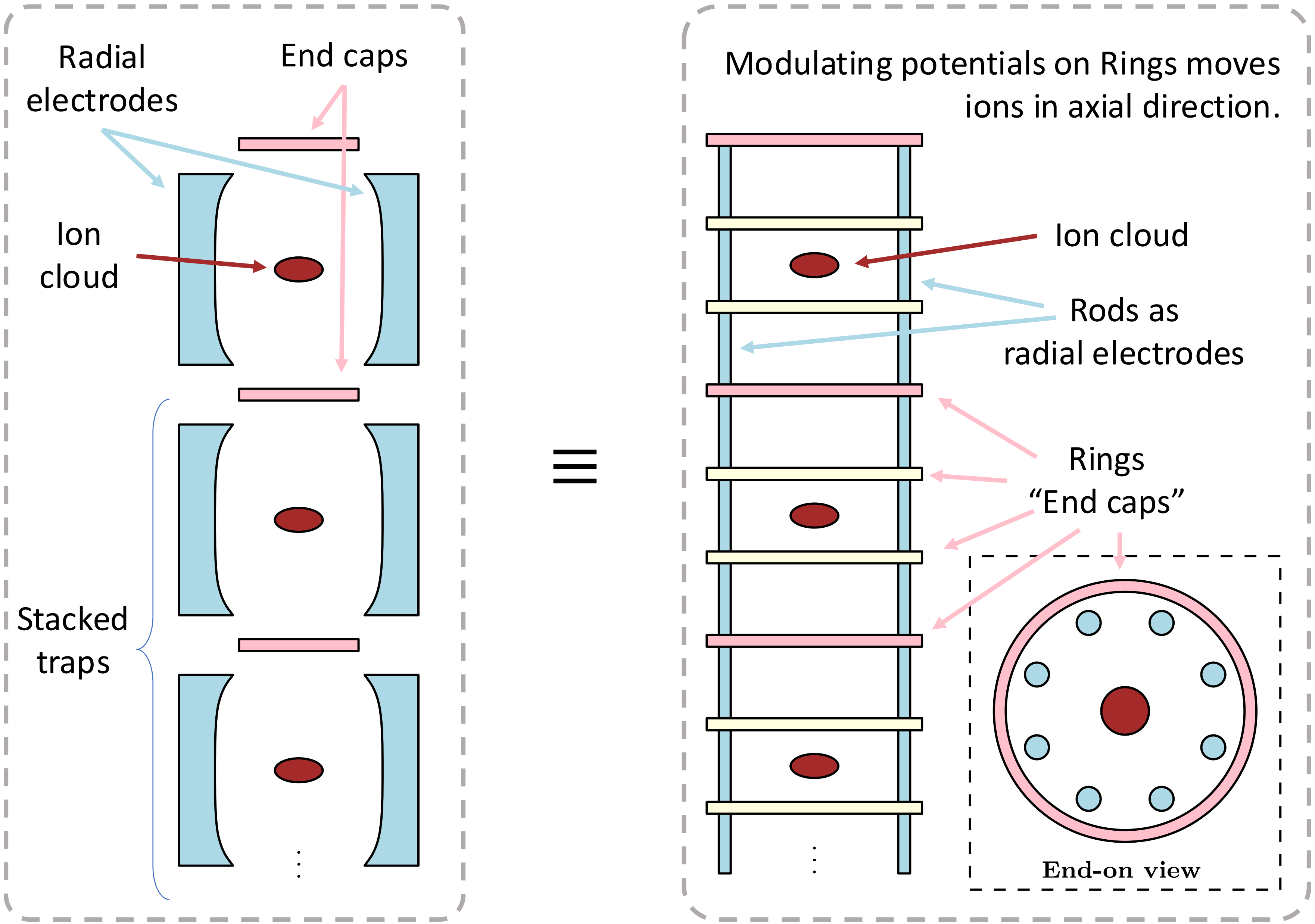


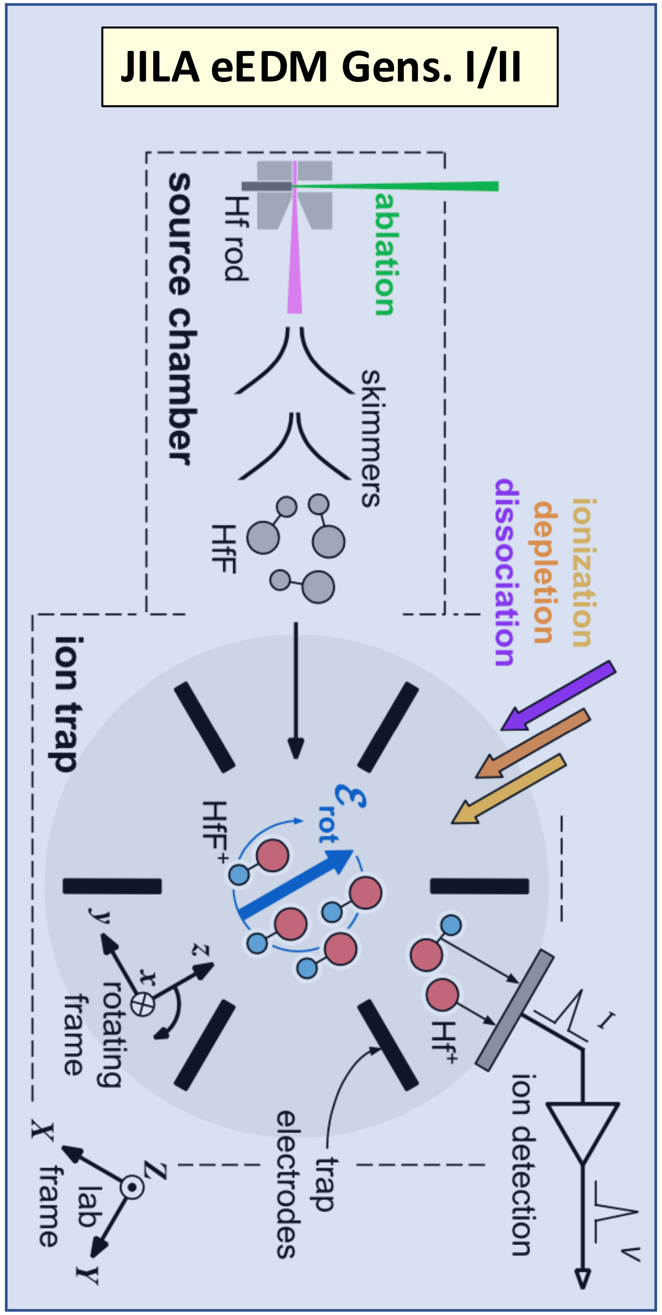
"Storage"



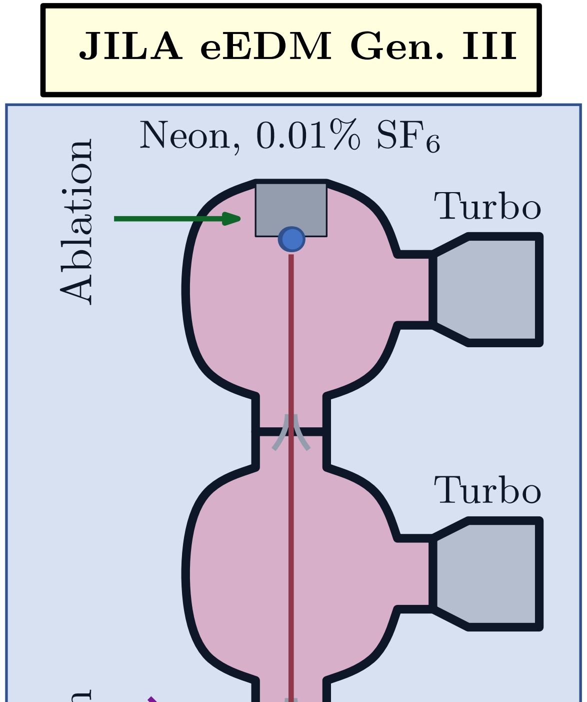
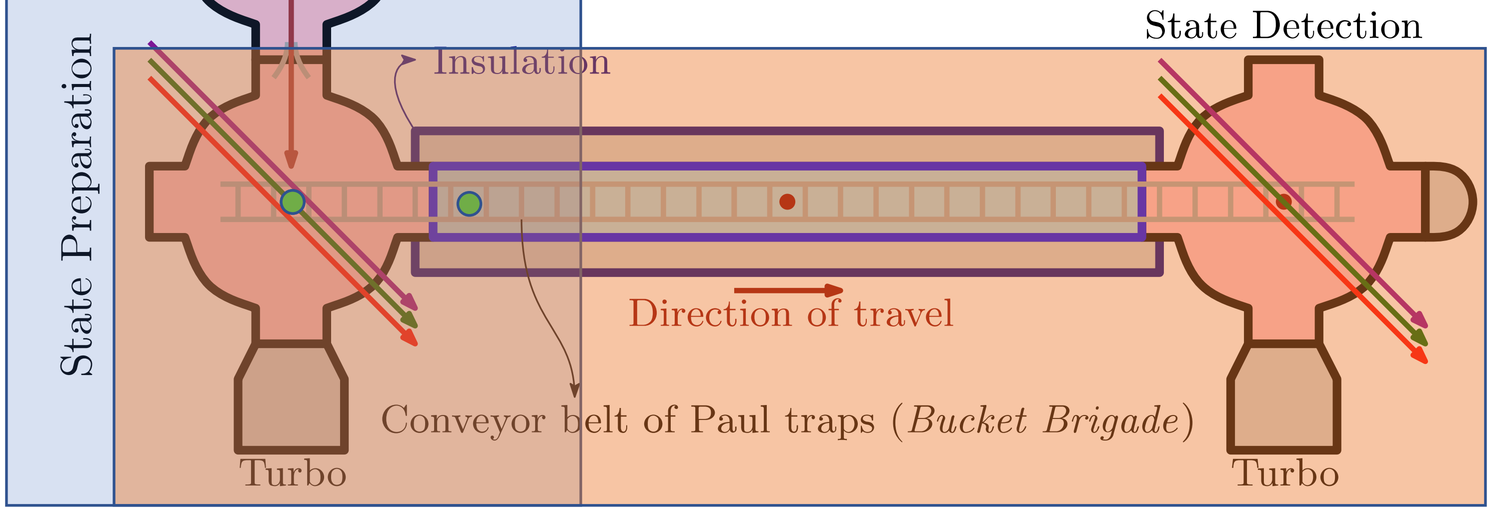
How can we make this?



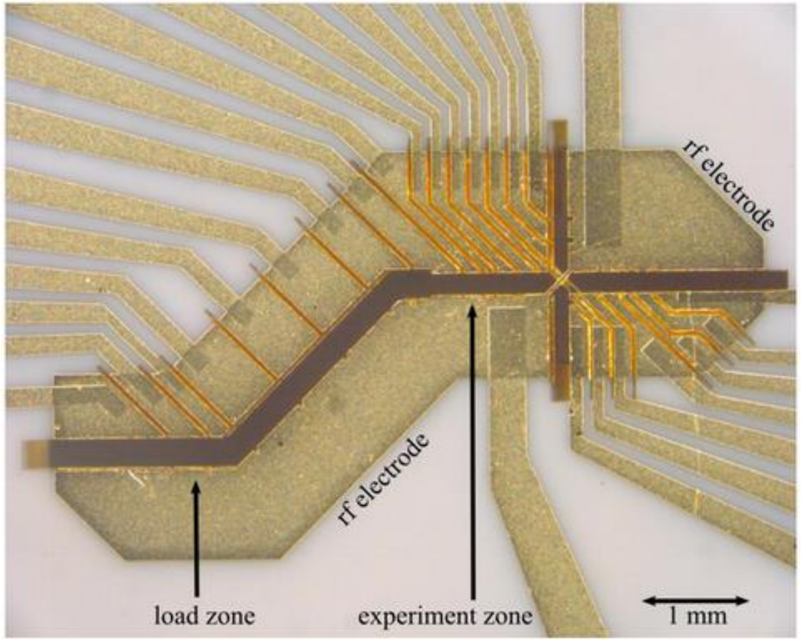

Radial electrodes
End caps
Ion cloud
Stacked traps
≡
Modulating potentials on Rings moves ions in axial direction.
Ion cloud
Rods as radial electrodes
Rings
“End caps”
End-on view



≡

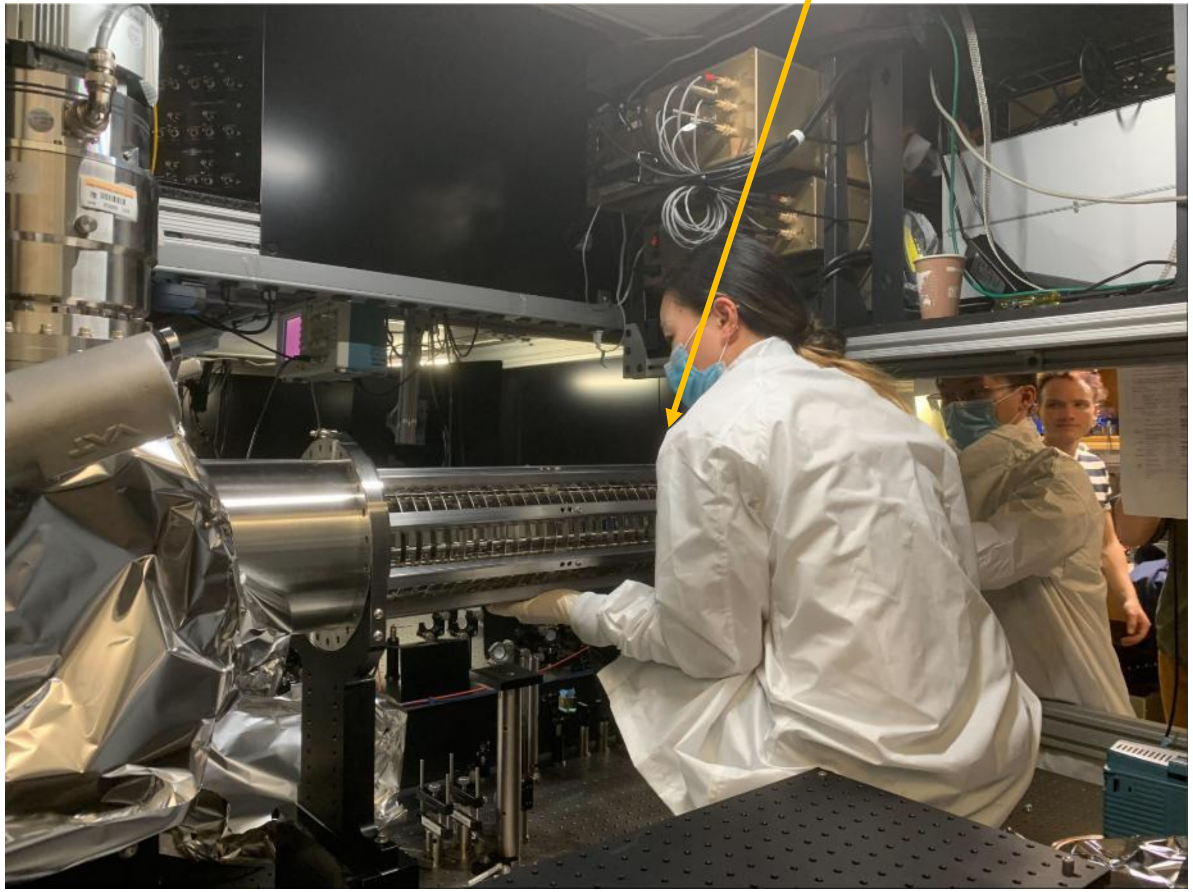



New in JILA eEDM Gen. III (*Bucket Brigade*)



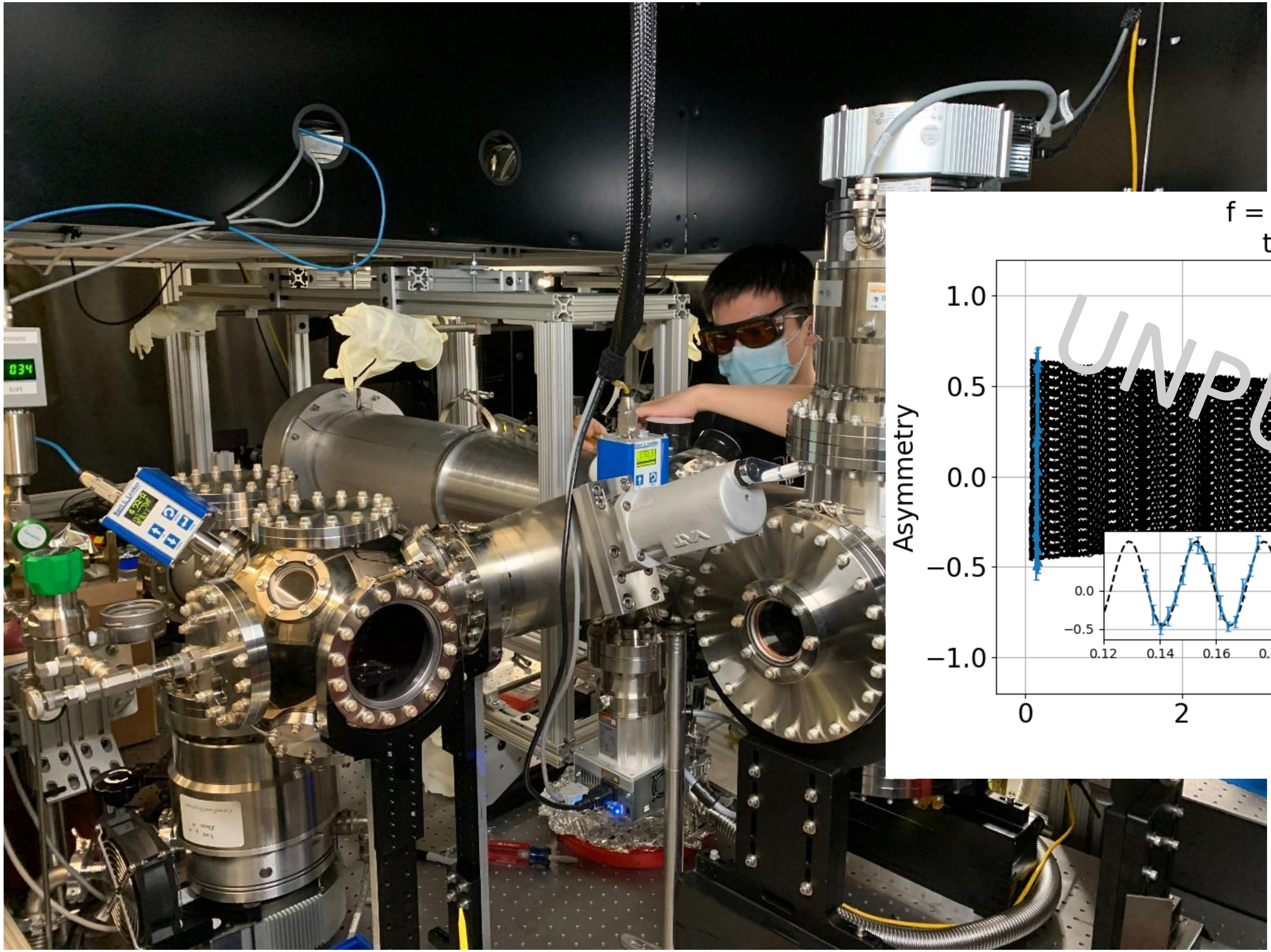
NIST, Ion Storage Group

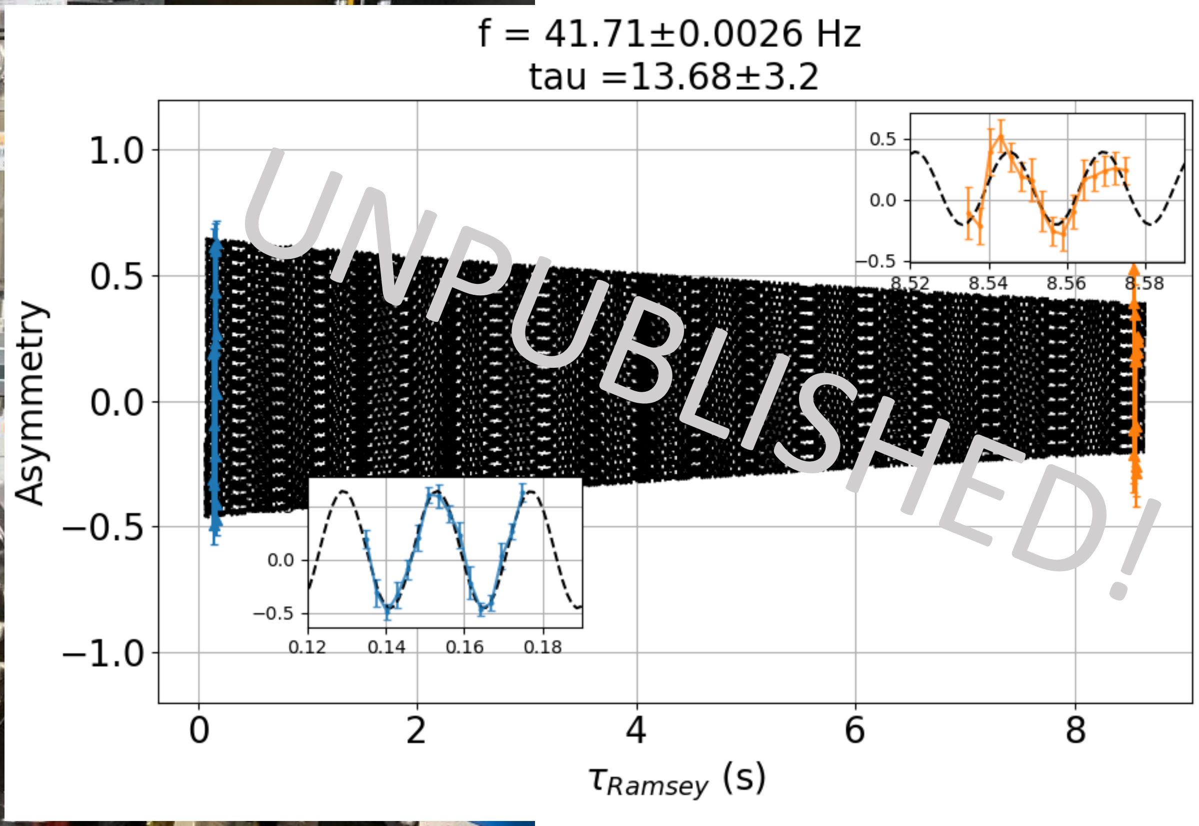
Electronics and structural design of the Baby Bucket



Loading the Baby Bucket into the vacuum chamber

Advisor included for scale





f = 41.71±0.0026 Hz
tau =13.68±3.2
Asymmetry
1.0
0.5
0.0
−0.5
−1.0
0
2
4
6
8
τ_{Ramsey} (s)
0.5
0.0
−0.5
8.52
8.54
8.56
8.58
0.0
−0.5
0.12
0.14
0.16
0.18
UNPUBLISHED!

Agenda

- Overview
  - Why electric dipole moments (EDMs)?
  - History of EDM measurements.

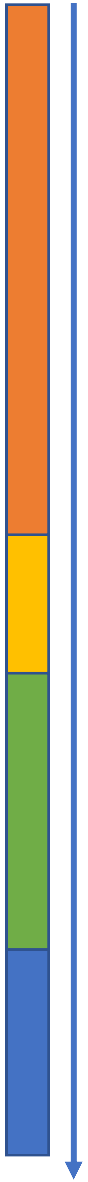
How eEDM experiments came to be

- Electron's EDM at JILA (HfF^+, ThF^+)
  - Current status (JILA Gen. II, Nov 2022).
  - JILA Gen. III (ongoing).

Diving into quantum-controlled molecules for fundamental physics

- What lies ahead
  - More advanced technology for eEDM.
  - Beyond eEDM.

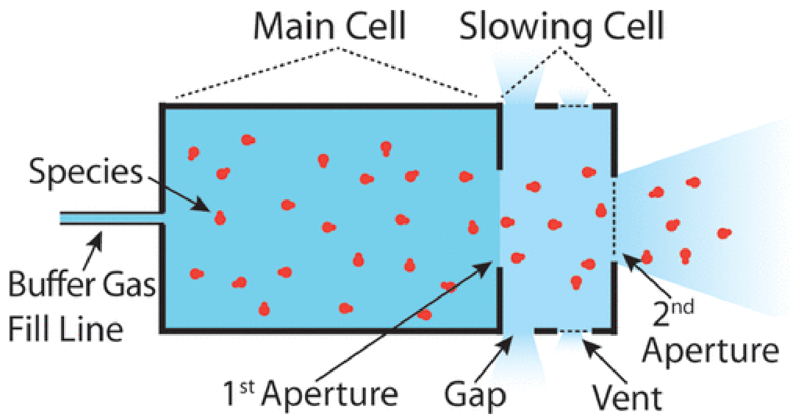
Broader landscape of quantum-controlled molecules for fundamental physics



Time spent on each section

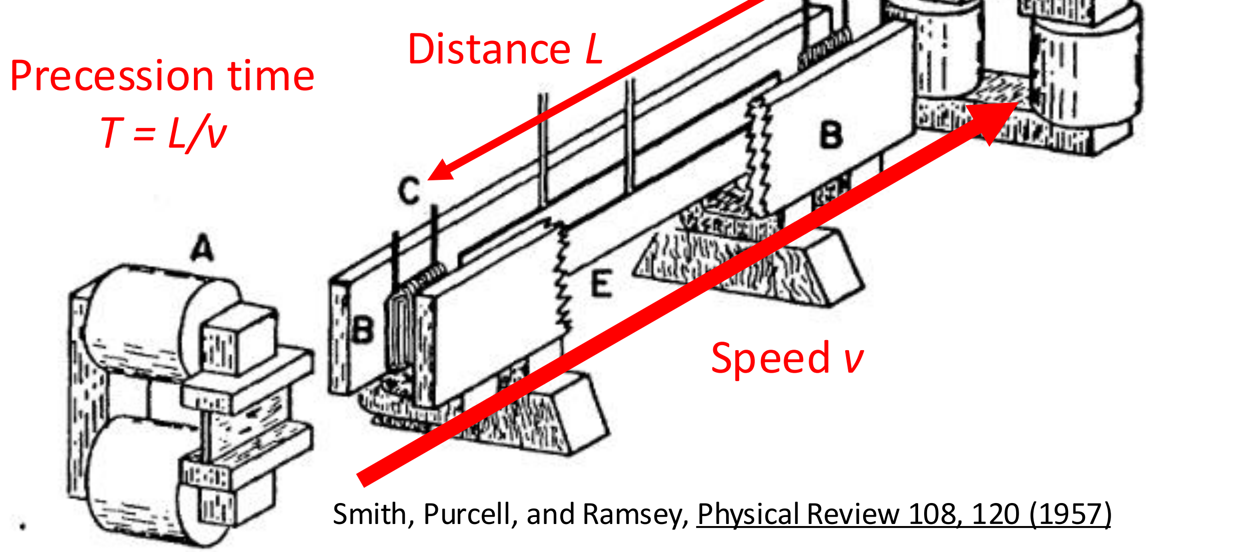
Common themes

- Increasing probing time
  - Reducing beam velocity
  - Trapping
- Increasing numbers
  - Refining chemical reaction
  - Focusing
  - Optical pumping
- Novel quantum manipulation
  - Polyatomic molecules



Hutzler et al., Chemical Reviews 112, 4803 (2012)

Precision of measurement
$\delta f \sim 1/T$



Smith, Purcell, and Ramsey, Physical Review 108, 120 (1957)

Common themes

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  - Trapping
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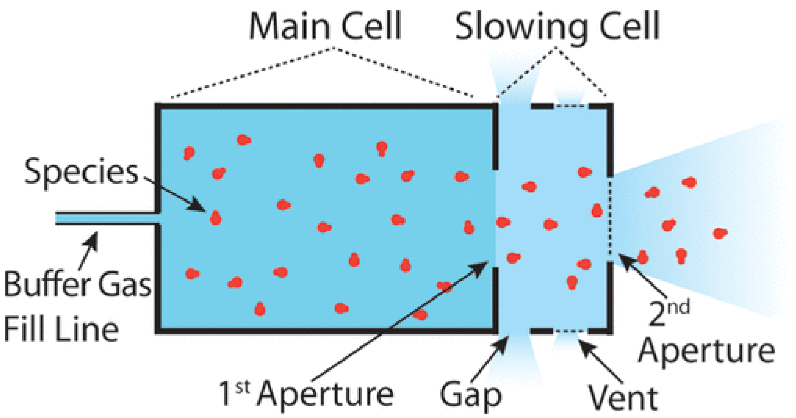

Hutzler et al., Chemical Reviews 112, 4803 (2012)

Precision of measurement
$\delta f \sim 1/T$

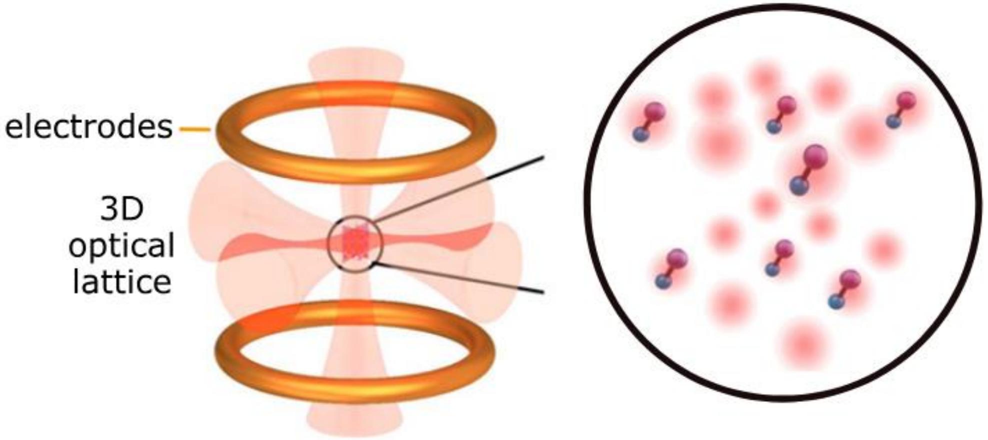

Figure 5. Illustration of a 3D optical lattice of YbF molecules for measuring the eEDM.

Fitch et al., Quantum Sci. Technol. **6** 014006 (2021)

Common themes

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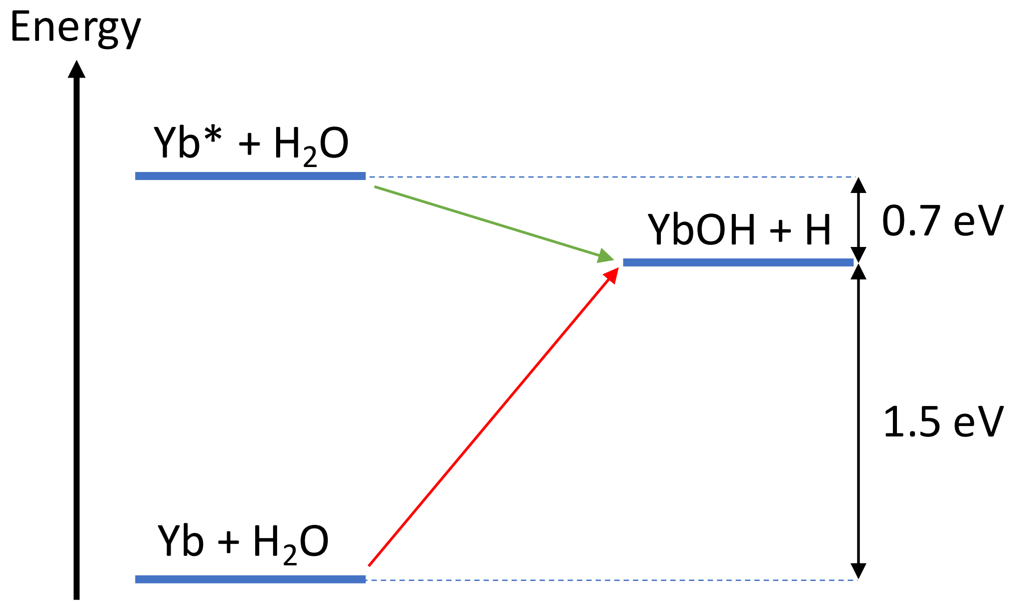
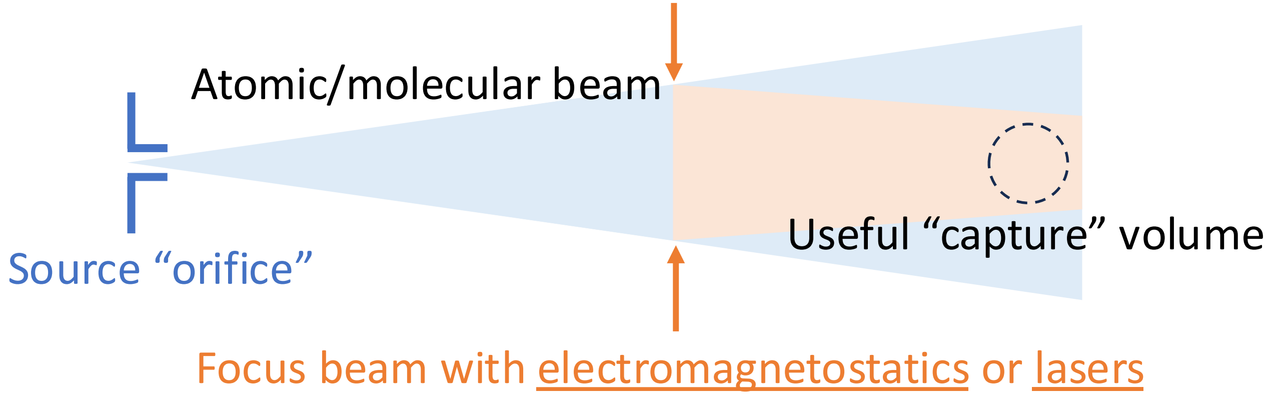


Adapted from Jadbabaie et al., New J. Phys. **22** 022002 (2020)



Common themes

- Increasing probing time
  - Reducing beam velocity
  - Trapping
- Increasing numbers
  - Refining chemical reaction
  - Focusing
  - Optical pumping
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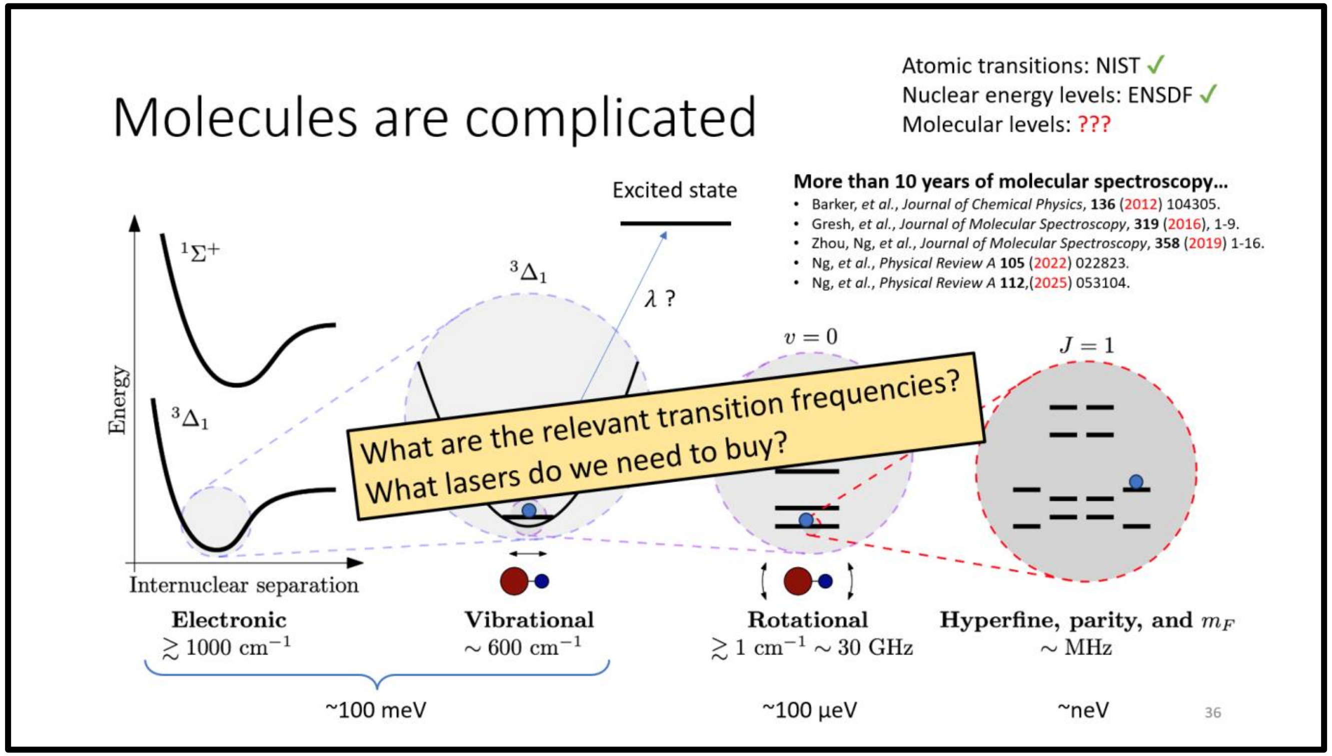
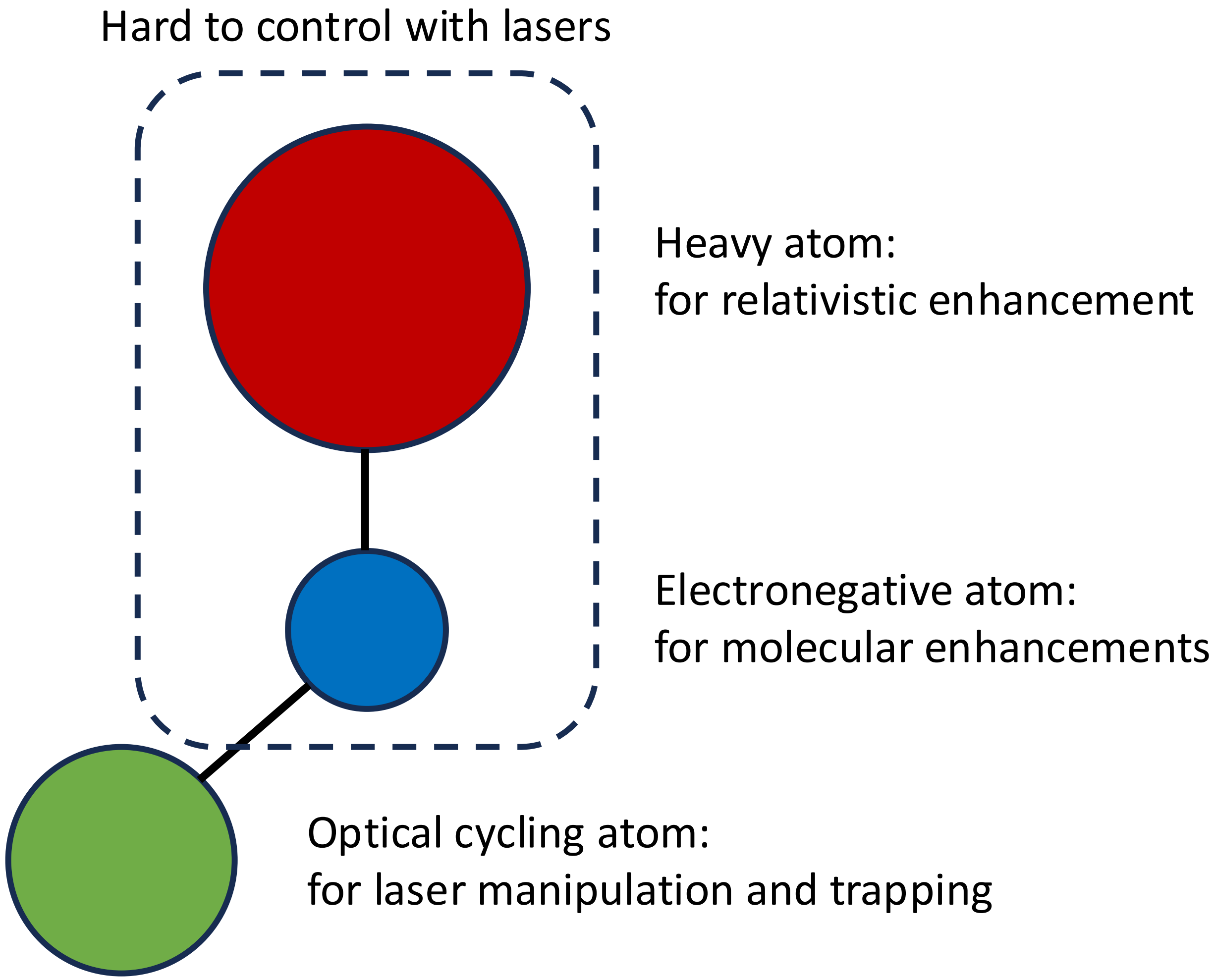
  - Polyatomic molecules



Common themes

- Increasing probing time
  - Reducing beam velocity
  - Trapping
- Increasing numbers
  - Refining chemical reaction
  - Focusing
  - Optical pumping
- Novel quantum manipulation
  - Polyatomic molecules

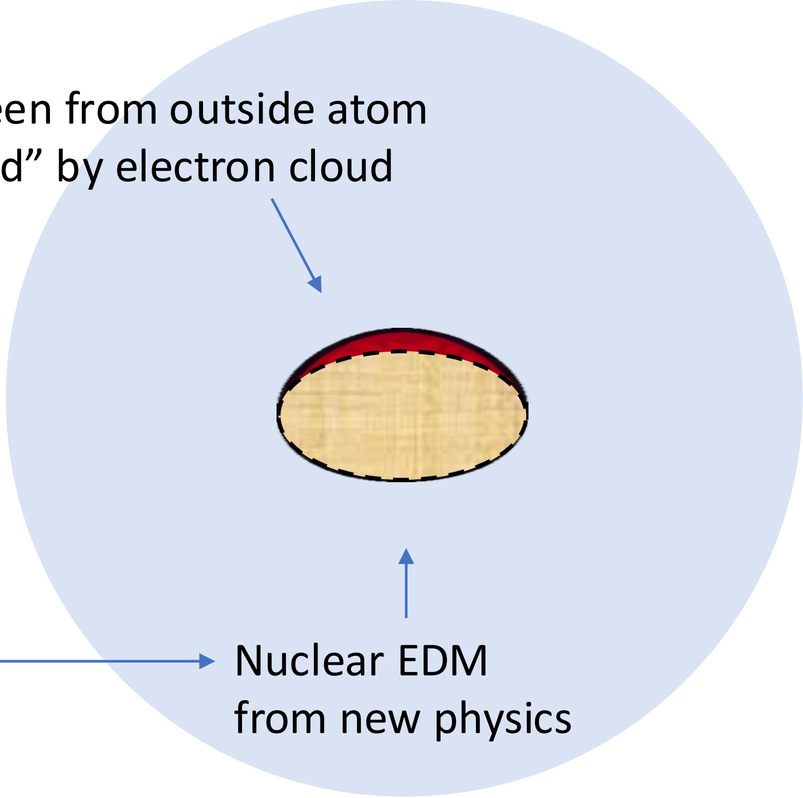
For example:



CP-V in atoms/molecules beyond eEDM

- Nuclear Schiff moment

Image seen from outside atom
“distorted” by electron cloud



Nuclear EDM
from new physics

Nucleon EDM + CP-violating nucleon-nucleon interactions

Image distorted by water droplets

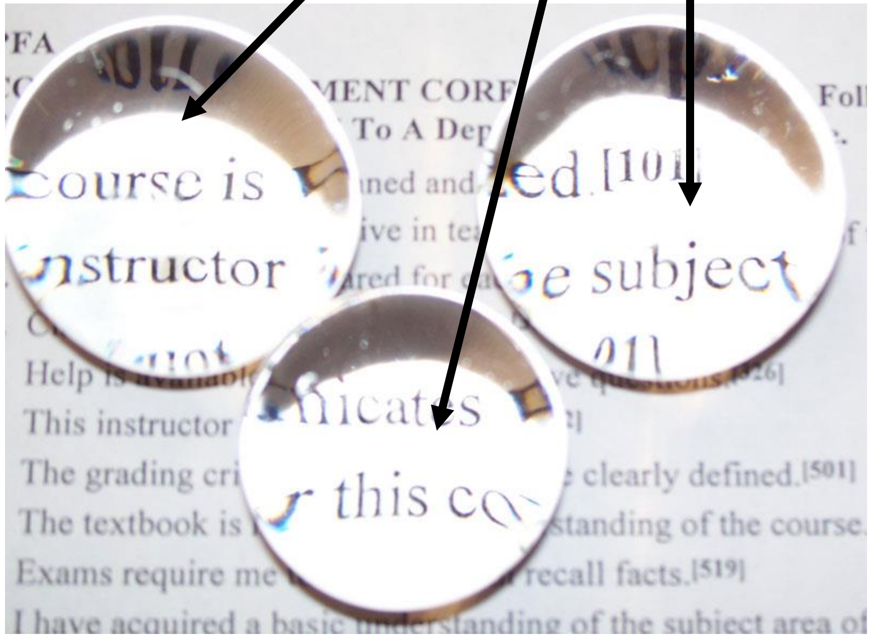


Image from the internet; no books were harmed in the process.

CP-V in atoms/molecules beyond eEDM

- Nuclear Schiff moment
  - Atoms
    - Hg, Xe, Ra
  - Molecules
    - TlF, RaX, FrAg, $^{227}ThF^{+}$, etc.

Happening at TRIUMF!

Nuclear enhancements from nuclear octupole deformation

Octupole-deformed nucleus:
usually short-lived ➔ accelerator facilities

Atomic/molecular techniques shared / co-developed between nuclear CP-V and eEDM experiments.

Conclusion

- Overview
  - Why electric dipole moments (EDMs)?
  - History of EDM measurements
- Electron's EDM at JILA (HfF^+, ThF^+)
  - Current status (JILA Gen. II, Nov 2022).
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- What lies ahead
  - More advanced technology for eEDM
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Adapted from Paul Hess's thesis