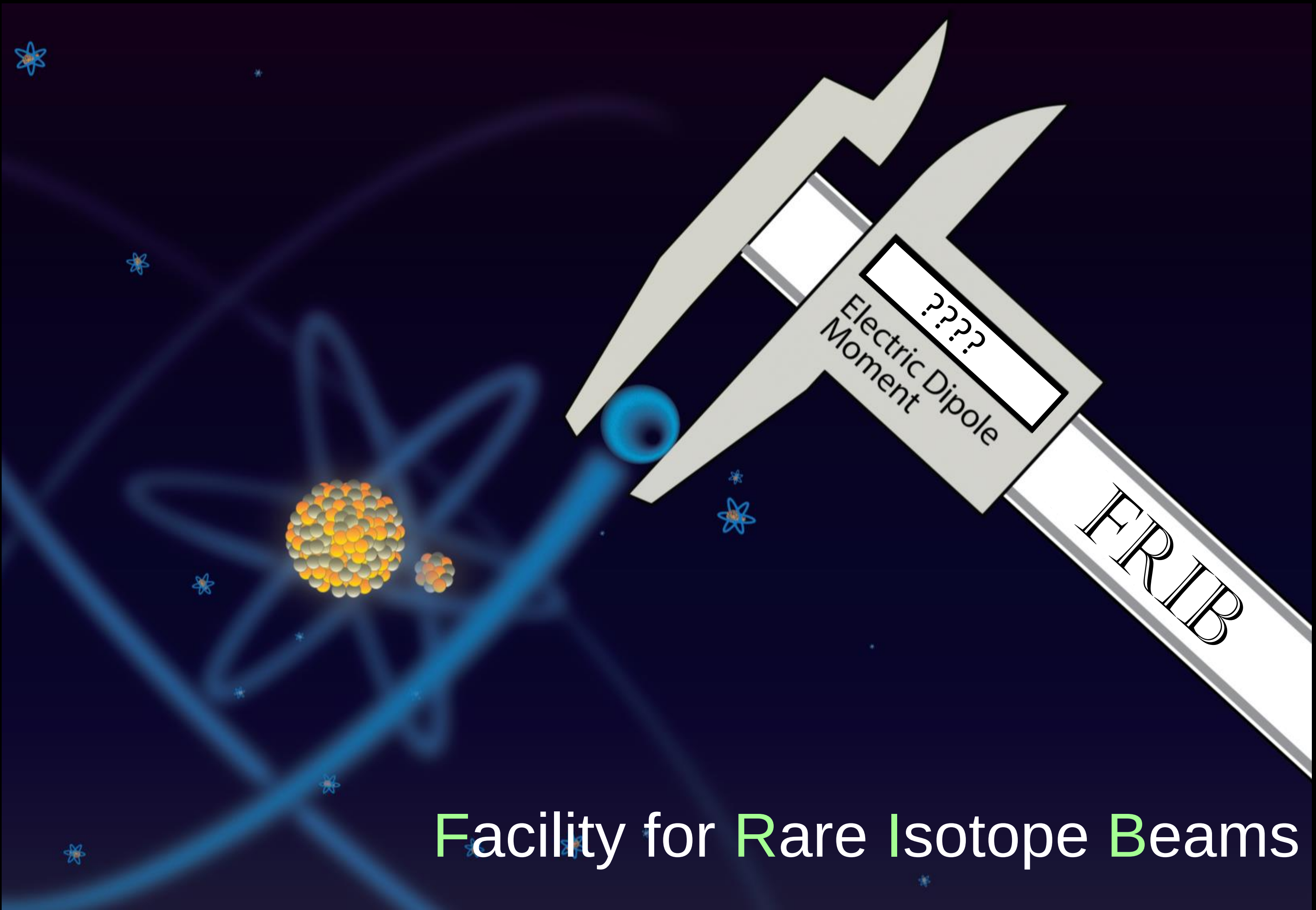
Advancing electric dipole moment (EDM) searches with ultracold radioactive molecules at FRIB



Xing Wu

FRIB &
Michigan State U

$$$$ Sponsored by DOE $$$$

U.S. DEPARTMENT OF ENERGY | Office of Science

ACME collaboration

University of Chicago
David DeMille (PI)
Zhen Han (graduate student)
Peiran Hu (graduate student)

Northwestern University
Gerald Gabrielse (PI)
Xing Fan (Res. Asst. Professor)
Daniel Ang (*Harvard grad student*)
Cole Meisenhelder (*Harvard grad student*)
Siyuan Liu (graduate student)
Maya Watts (graduate student)
Collin Diver (graduate student)

Harvard University
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Zack Lasner (postdoc)

Okayama University
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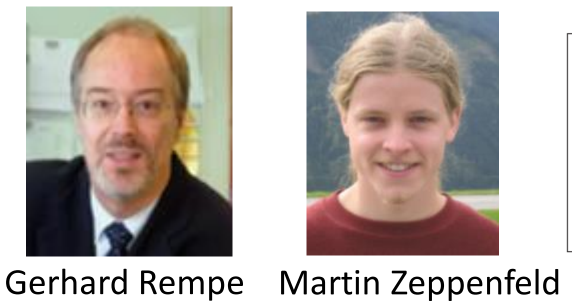
Noboru Sasao
Satoshi Uetake
Takahiko Masuda
Ayami Hiramoto

Other collaborators
Cris Panda (Berkeley)
Nick Hutzler (Caltech)
Xing Wu (FRIB/MSU)

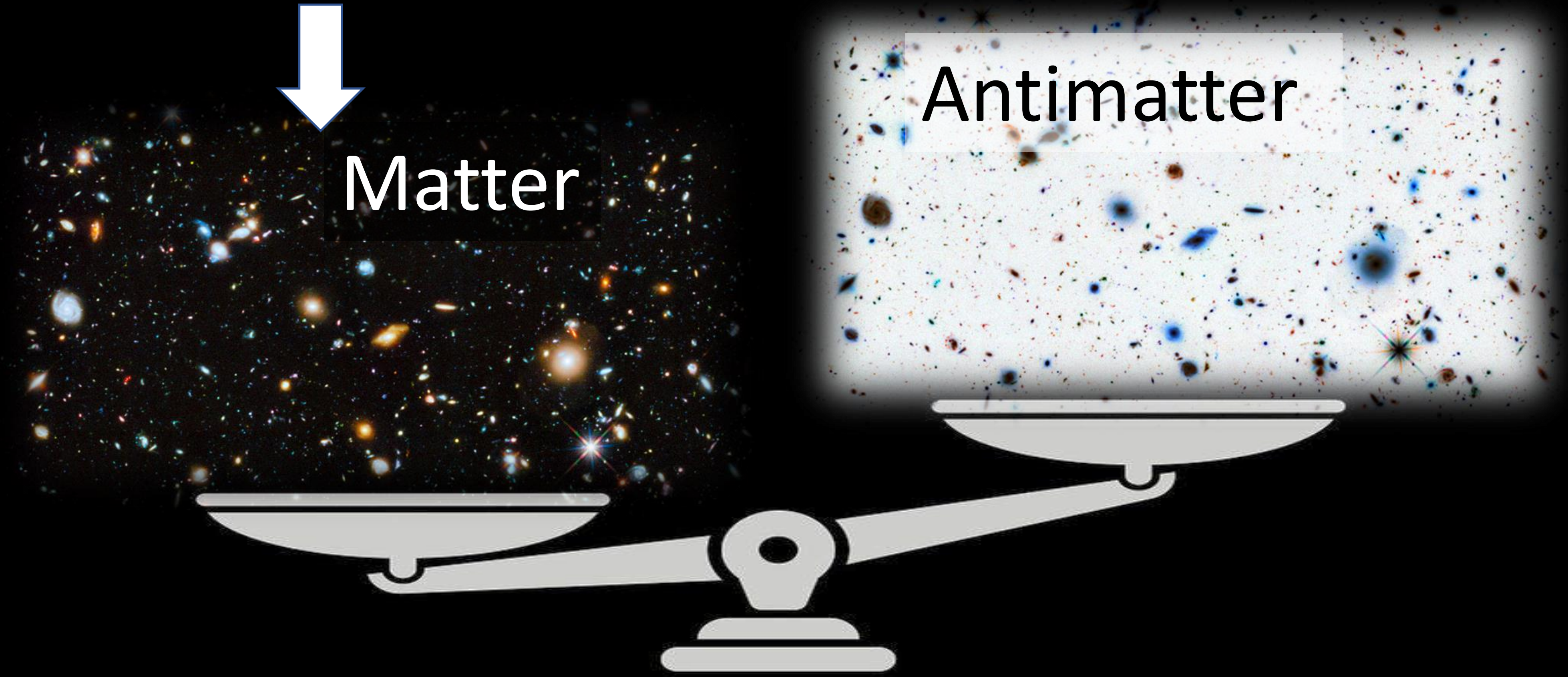
FRIB, MSU

MPI of Quantum Optics
Garching, DE:



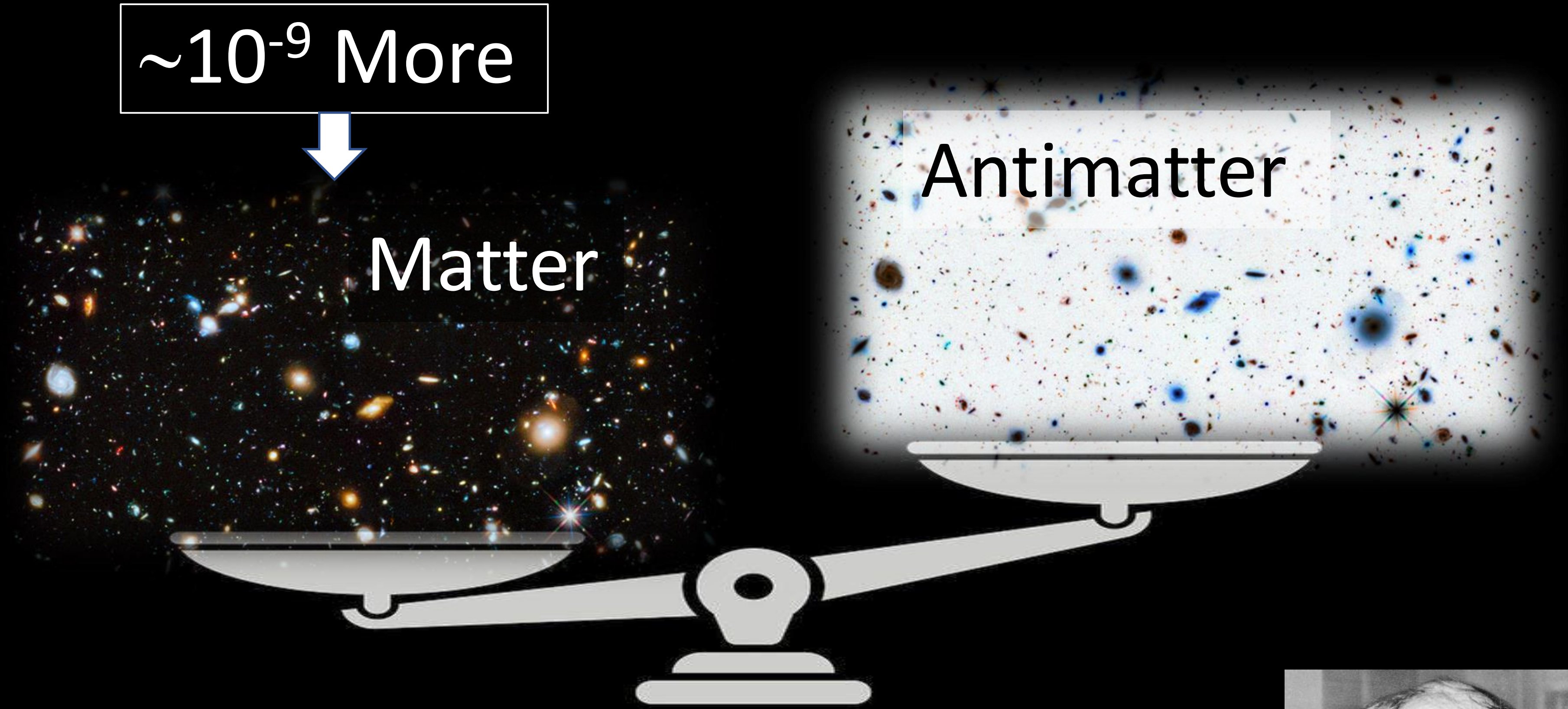
After the Big Bang:

~1 part per billion more matter than antimatter



The remaining ~10^{-9} after Annihilation makes up the entire observable Universe!

Supported by n_B/n_γ from Cosmology Observation



- Matter-favoring mechanism must violate *CP*-symmetry *JETP Lett. 5, 27 (1967)*
- *CP:* Charge & Parity reversal

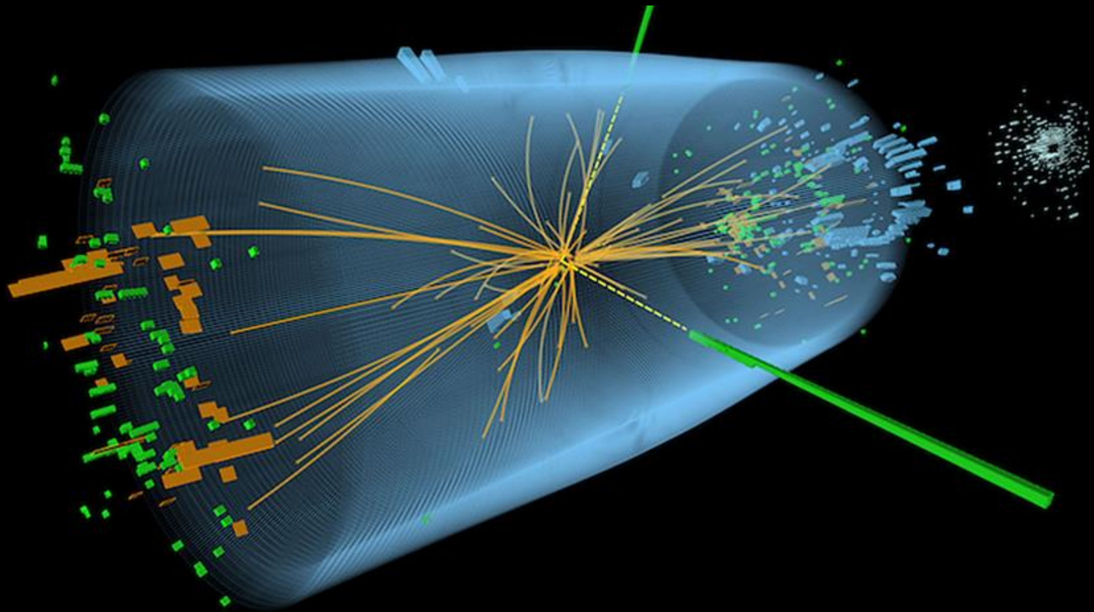
CP ↔ *T:* Time reversal
Assuming *CPT* invariance

A.D. Sakharov

Peace Prize 1975

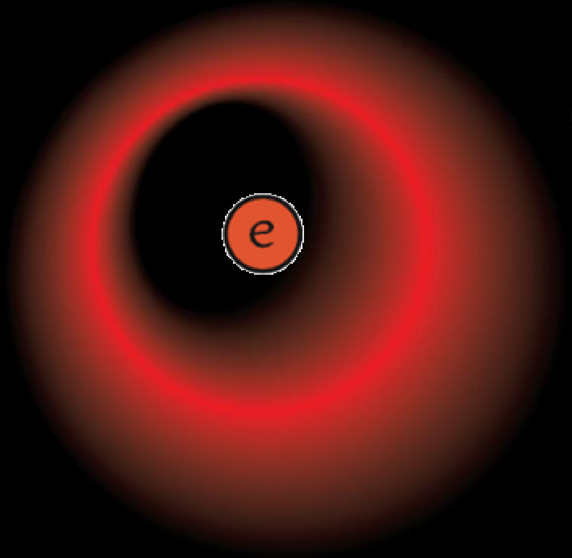
Two Ways to Hunt for the Exotic Particles

‘Conventional’ Approach: Particle Colliders



Higgs Boson @ LHC, Cern

Search for: Electric Dipole Moment (EDM)



Asymmetry in charge distribution

Norman Ramsey
Proposed In 1950

Physics 1989

Two Ways to Hunt for the Exotic Particles

≲10 TeV
(Direct Search)

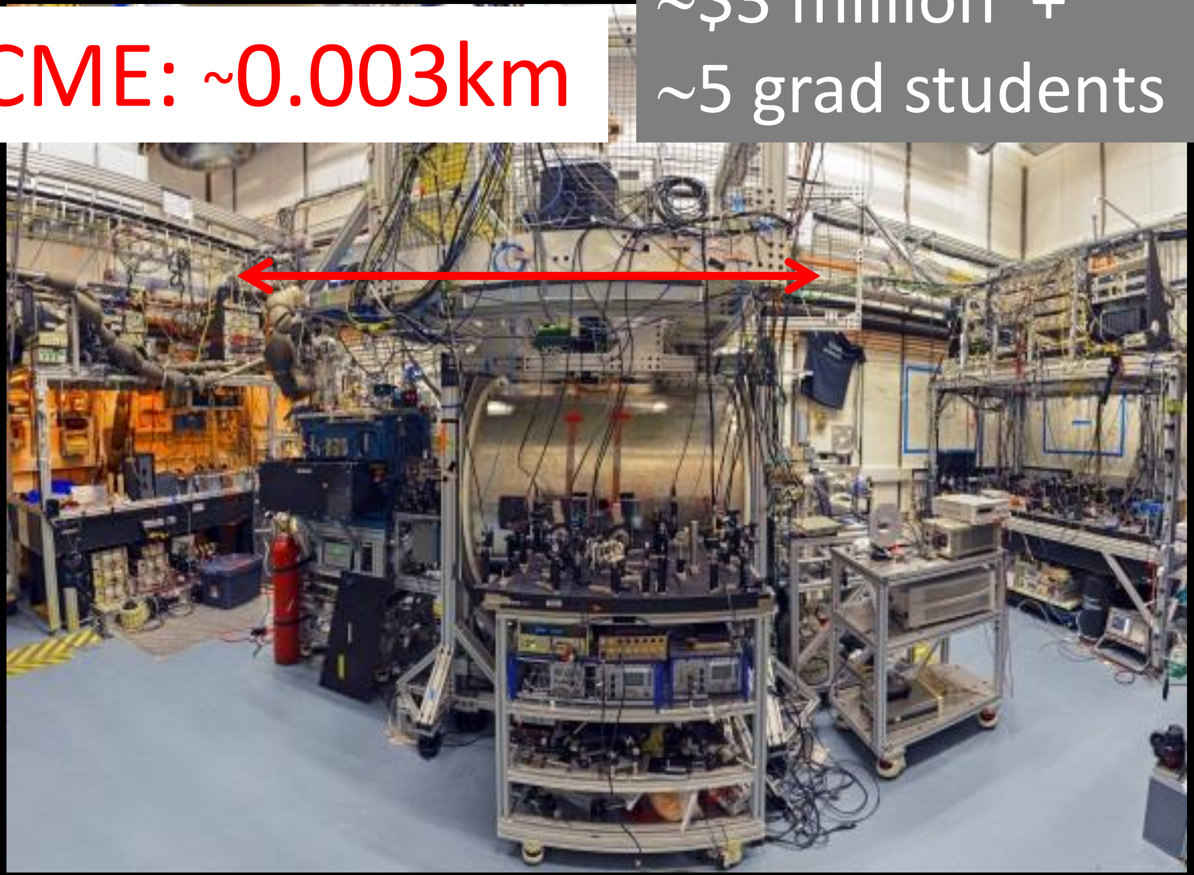
LHC: ~30km

~$5 billion +
~$1 billion/yr

~30 TeV & beyond
(Model Testing)

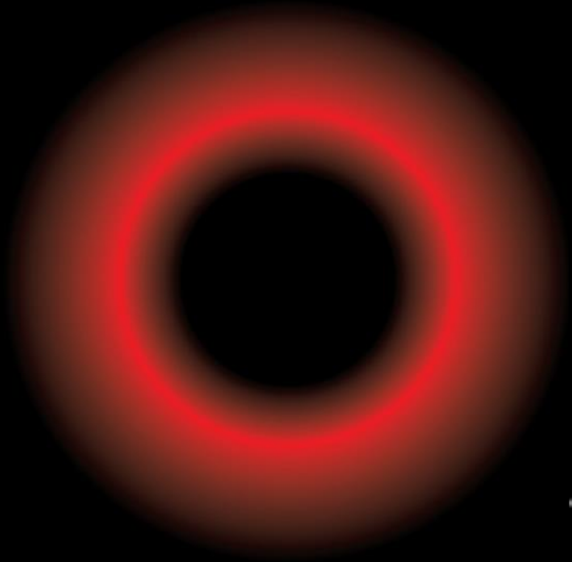
ACME: ~0.003km

~$3 million +
~5 grad students

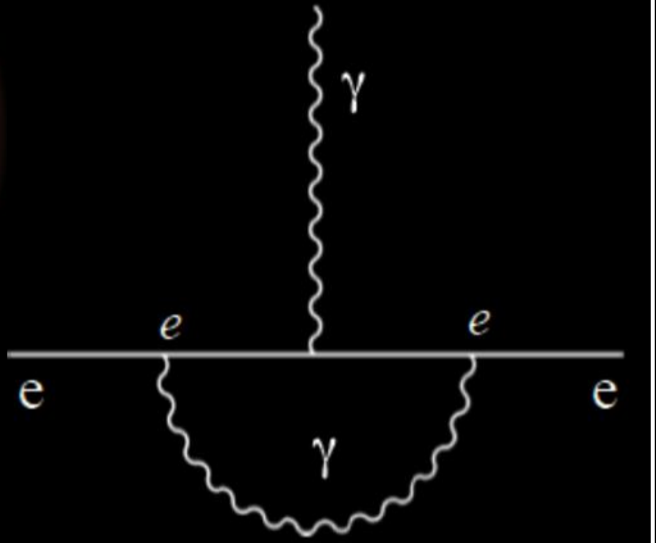


Electron EDM Violates *CP*

Standard Model



Magnetic Moment anomaly

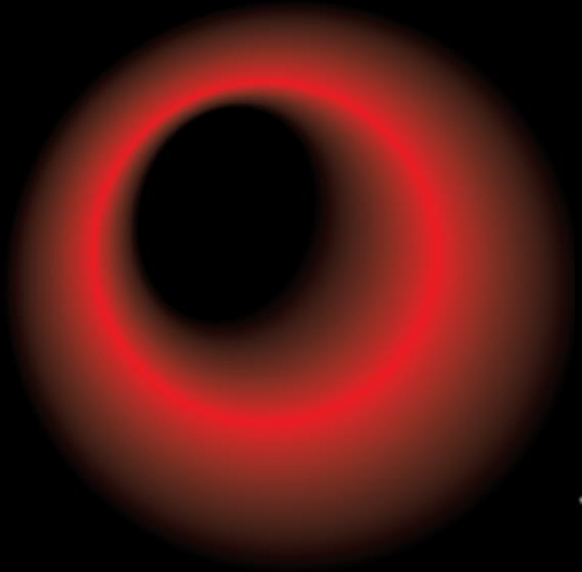


Electron is addressed by Virtual Particle – g-2

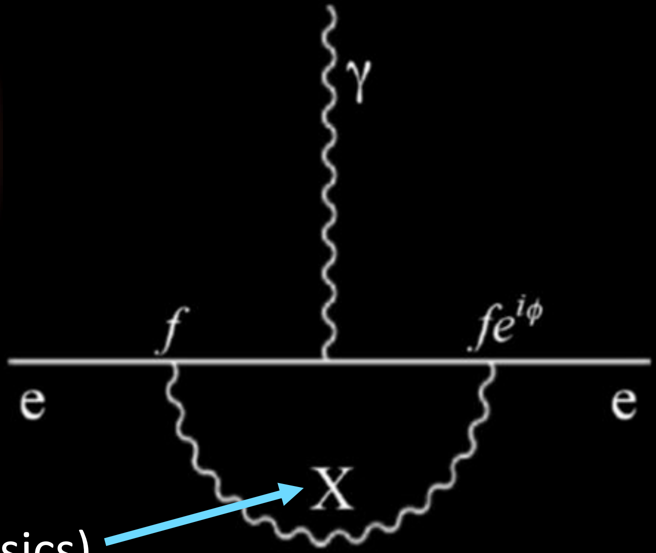
Well-known in QED to 0.13 parts per trillion

g/2=1.001 159 652 180 59 (13)
X. Fan et al. *PRL. 130, 071801 (2023)*

Beyond SM



Electric Dipole Moment ?

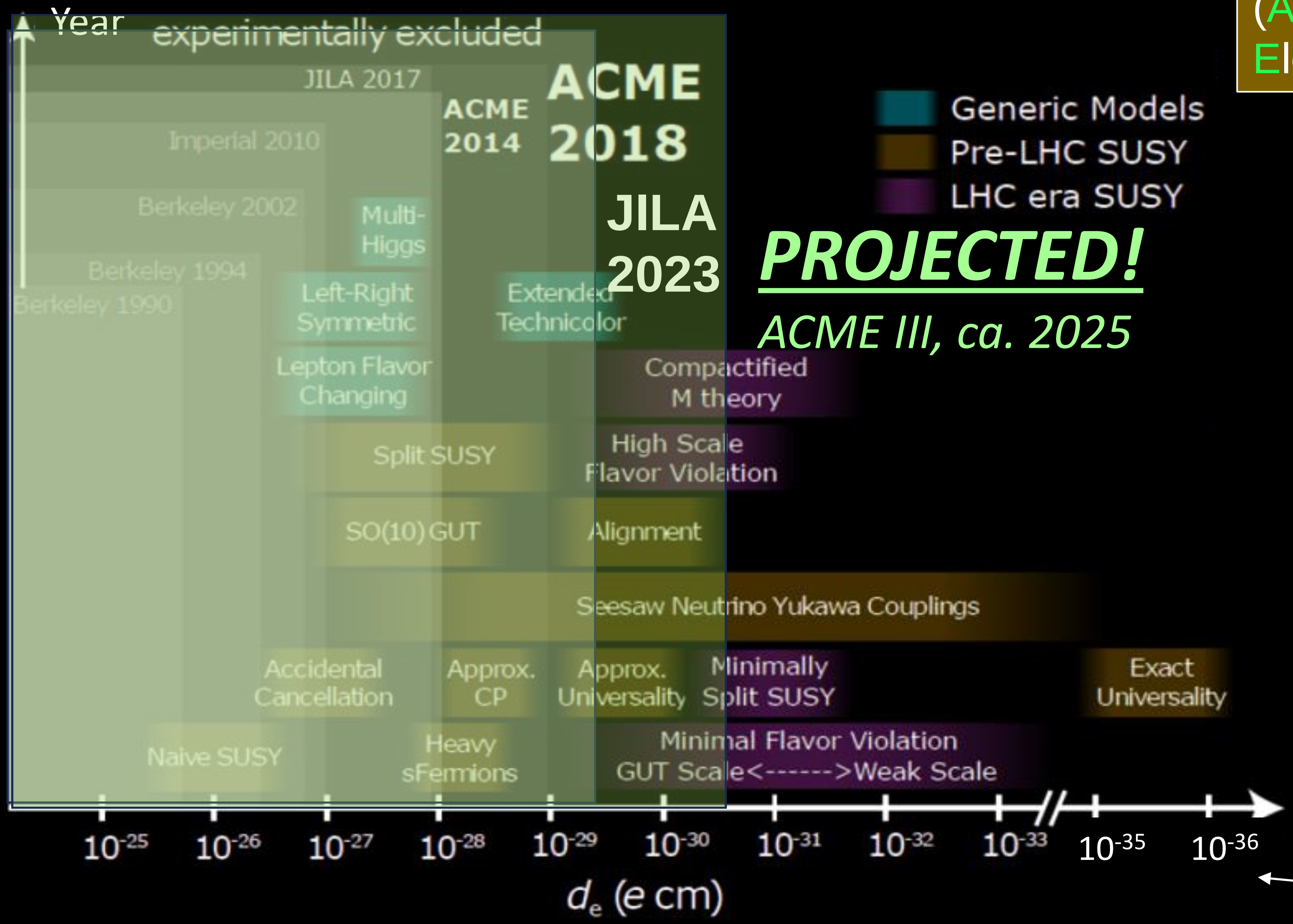


Electron is addressed by Virtual Particle, **X** (new physics)

1st order perturbation, intrinsic *CP*-violating phase

EDM Narrows the Search for Theory Beyond SM

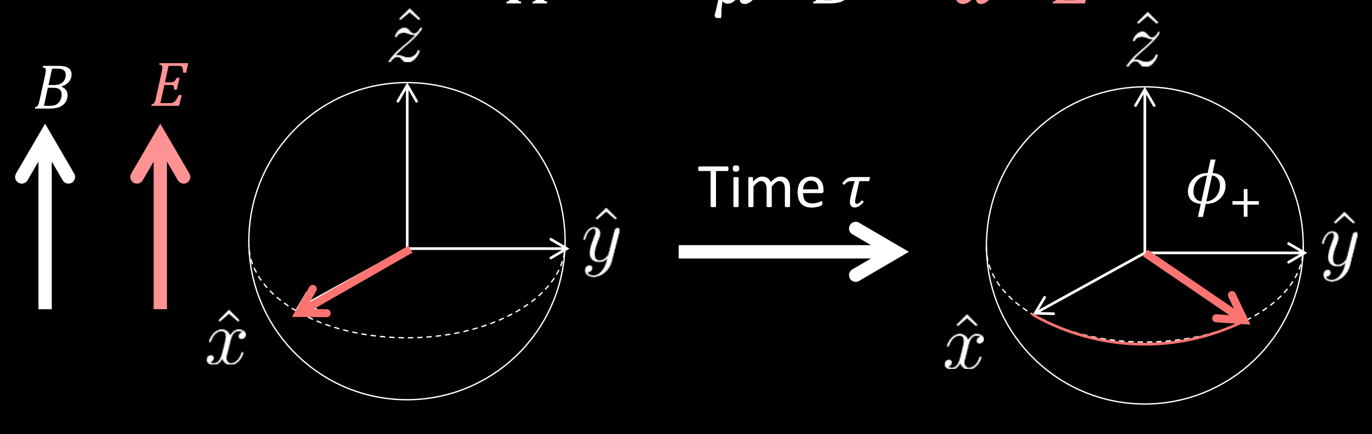
(ACME: Advanced Cold Molecule Electron EDM search)



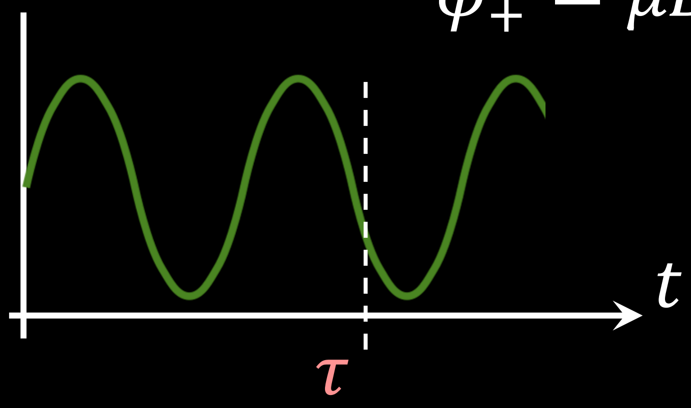
Y. Ema, T. Gao, M. Pospelov
***PRL** 129, 231801 (2022)*

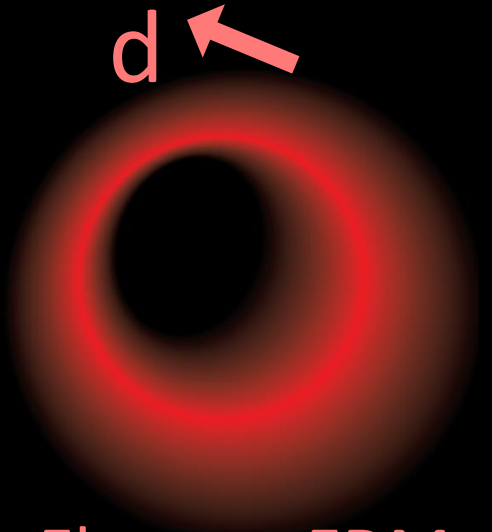
Search EDMs by Ramsey Interference Measurement

$$H = -\mu \cdot B - d \cdot E$$



$$\phi_+ = \mu B\tau + dE\tau$$

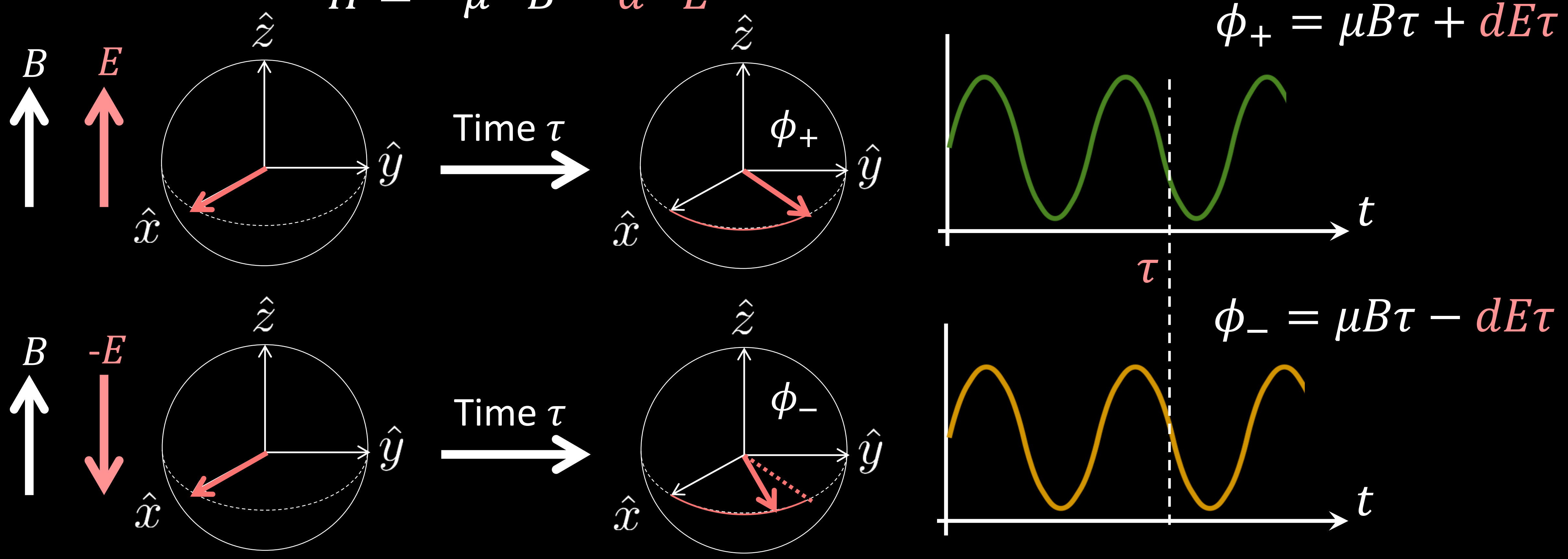




Electron EDM

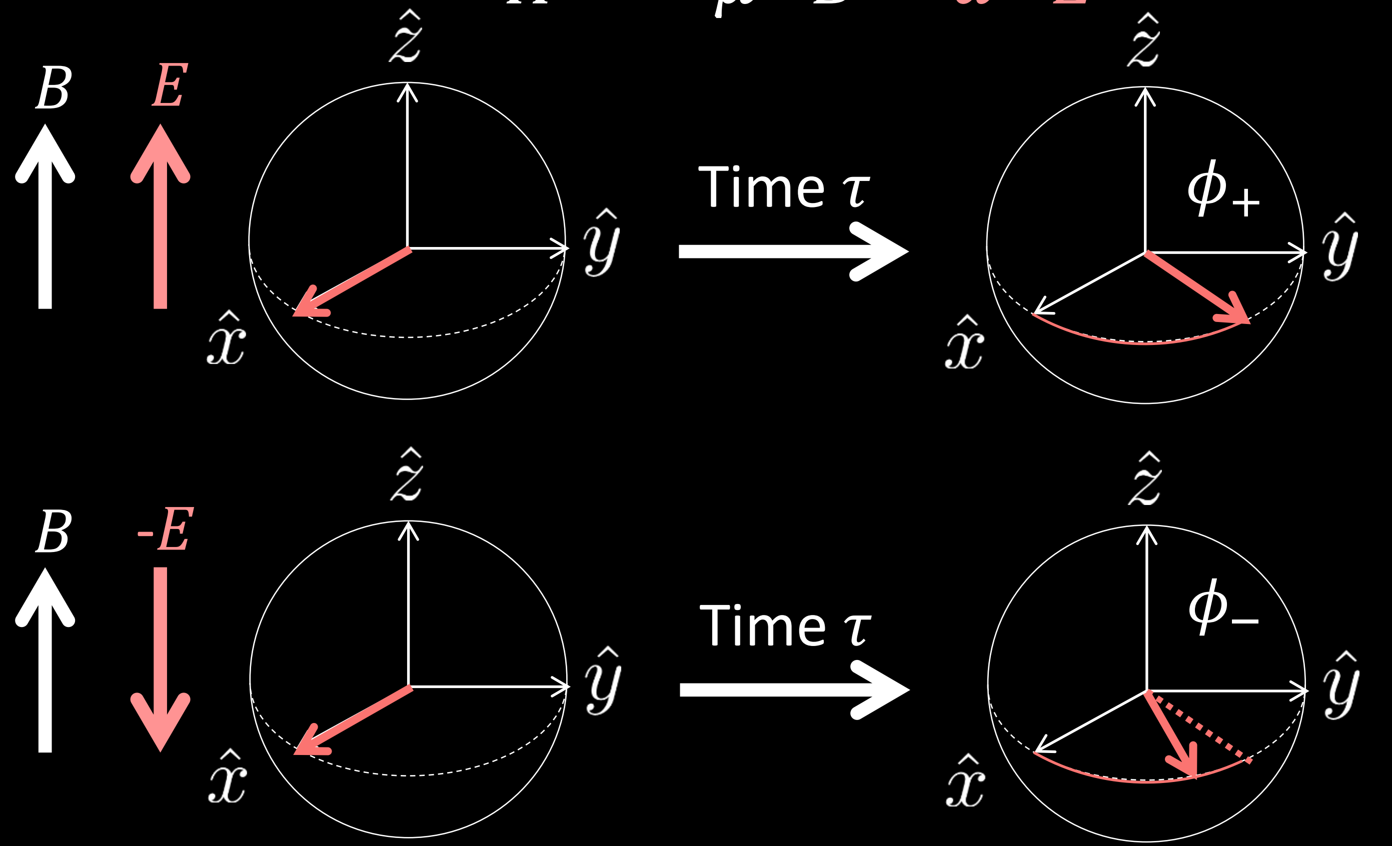
Extract EDM by Reversing the E-field

$$H = -\mu \cdot B - d \cdot E$$



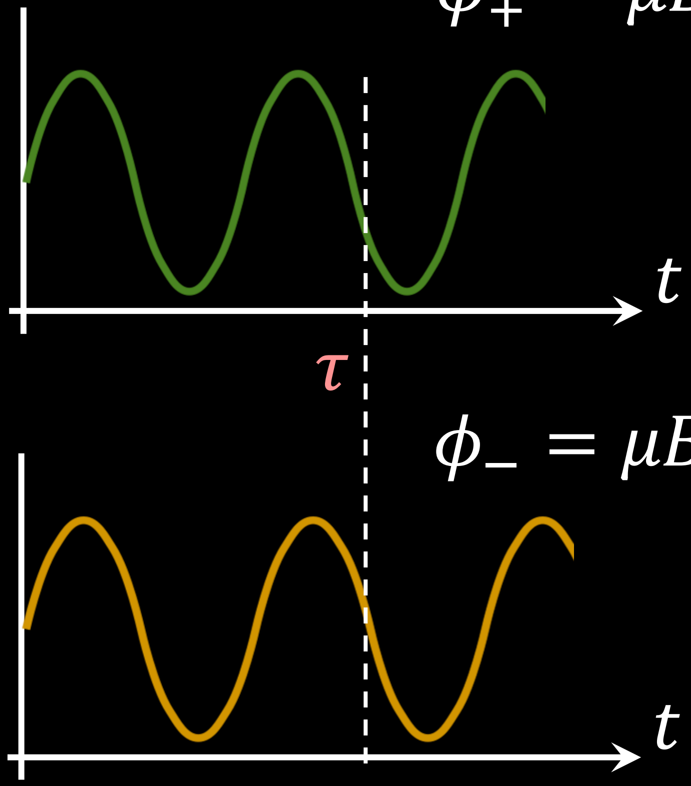
Extract EDM by Reversing the E-field

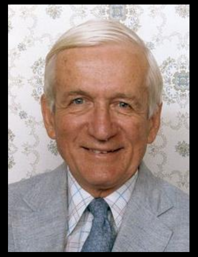
$$H = -\mu \cdot B - d \cdot E$$



$$\phi_+ = \mu B\tau + dE\tau$$

$$\phi_- = \mu B\tau - dE\tau$$





Norman Ramsey

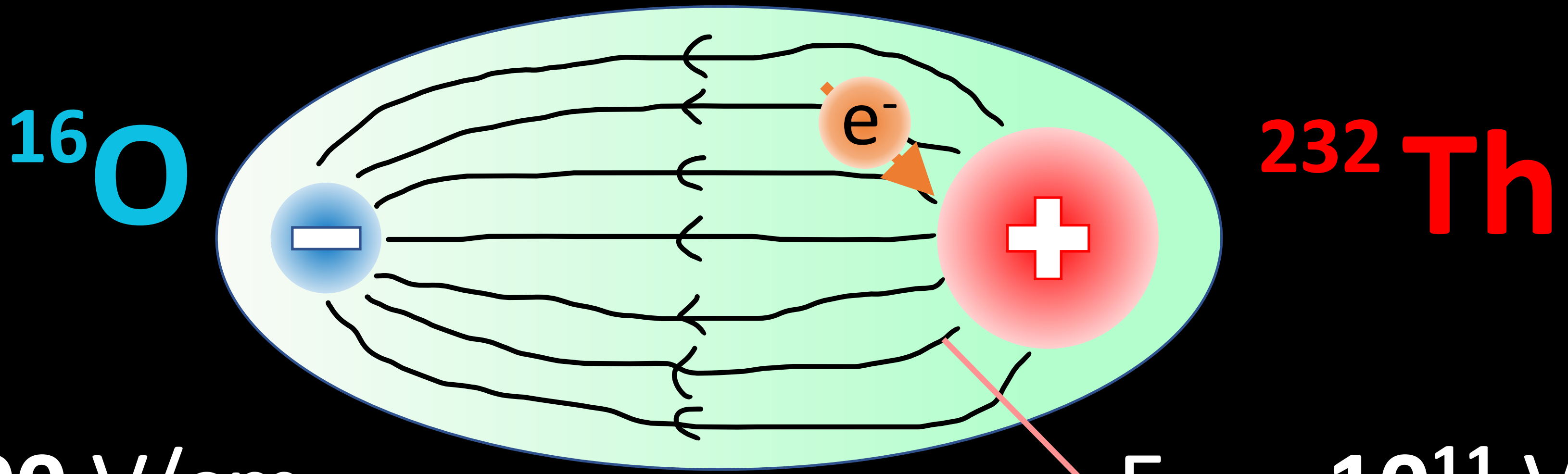
Physics 1989

Electron EDM can be extracted: $d \propto \phi_+ - \phi_-$

Molecule Amplifies EDM Interaction!

ACME uses ThO molecules:

$E_{eff} \propto Z^3$



E_{lab} ~ **100** V/cm:

Applied E-field to polarize molecules

E_{eff} ~ **10^{11}** V/cm:

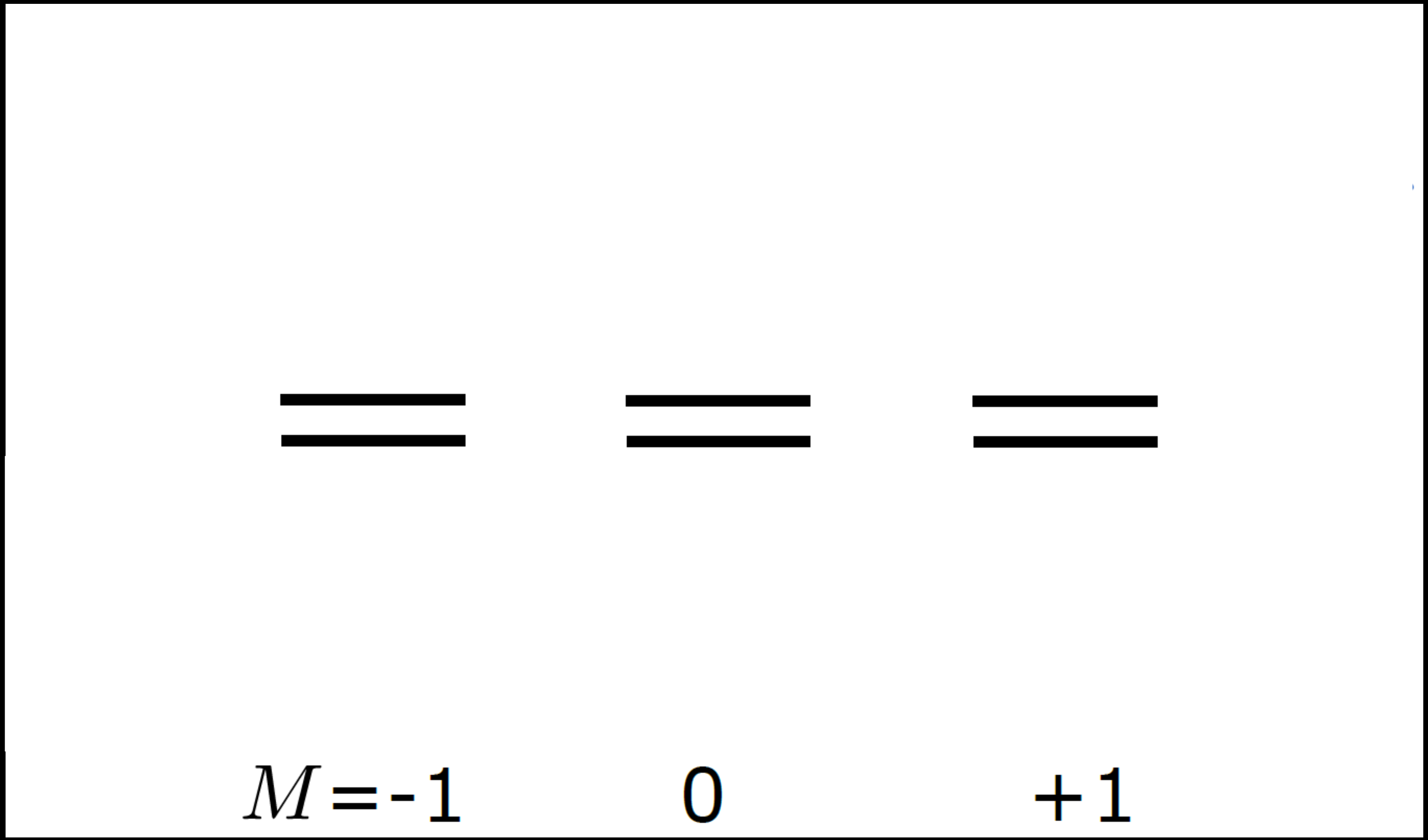
Effective E-field around heavy nucleus

$H\ ^3\Delta_1$ state of ThO: Excellent for EDM Search

- Insensitive to magnetic field noise
  - Σ=1, Λ=2, but they are anti-parallel, so the magnetic moments nearly cancel out
  - Measured to be μ_H=0.00440(5) μ_B
- Ω= Λ + Σ = 2 + (-1) = 1, giving parity doublet

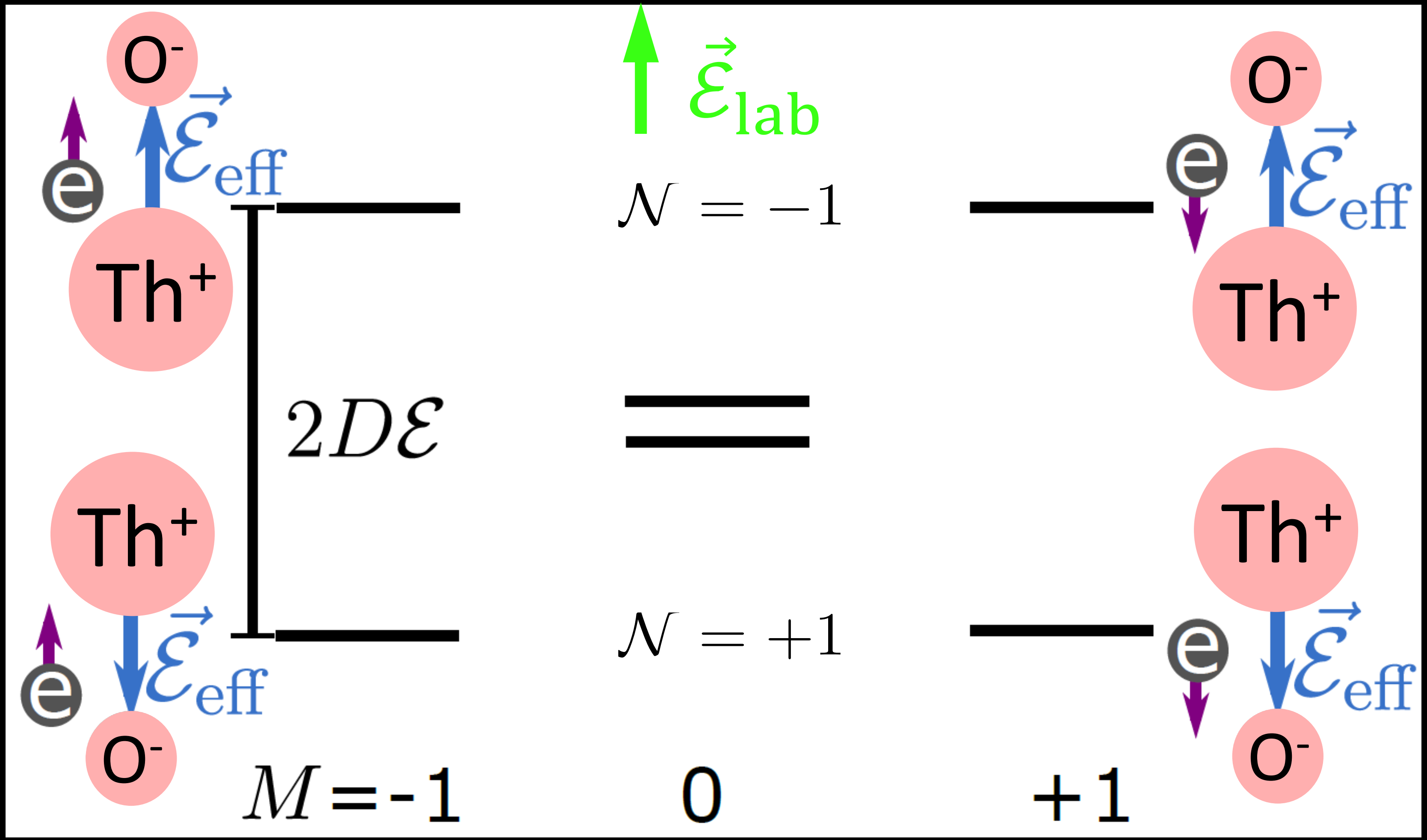
$H\,^3\Delta_1$ state of ThO: Excellent for EDM Search

- Easily polarizable



$H\,^3\Delta_1$ state of ThO: Excellent for EDM Search

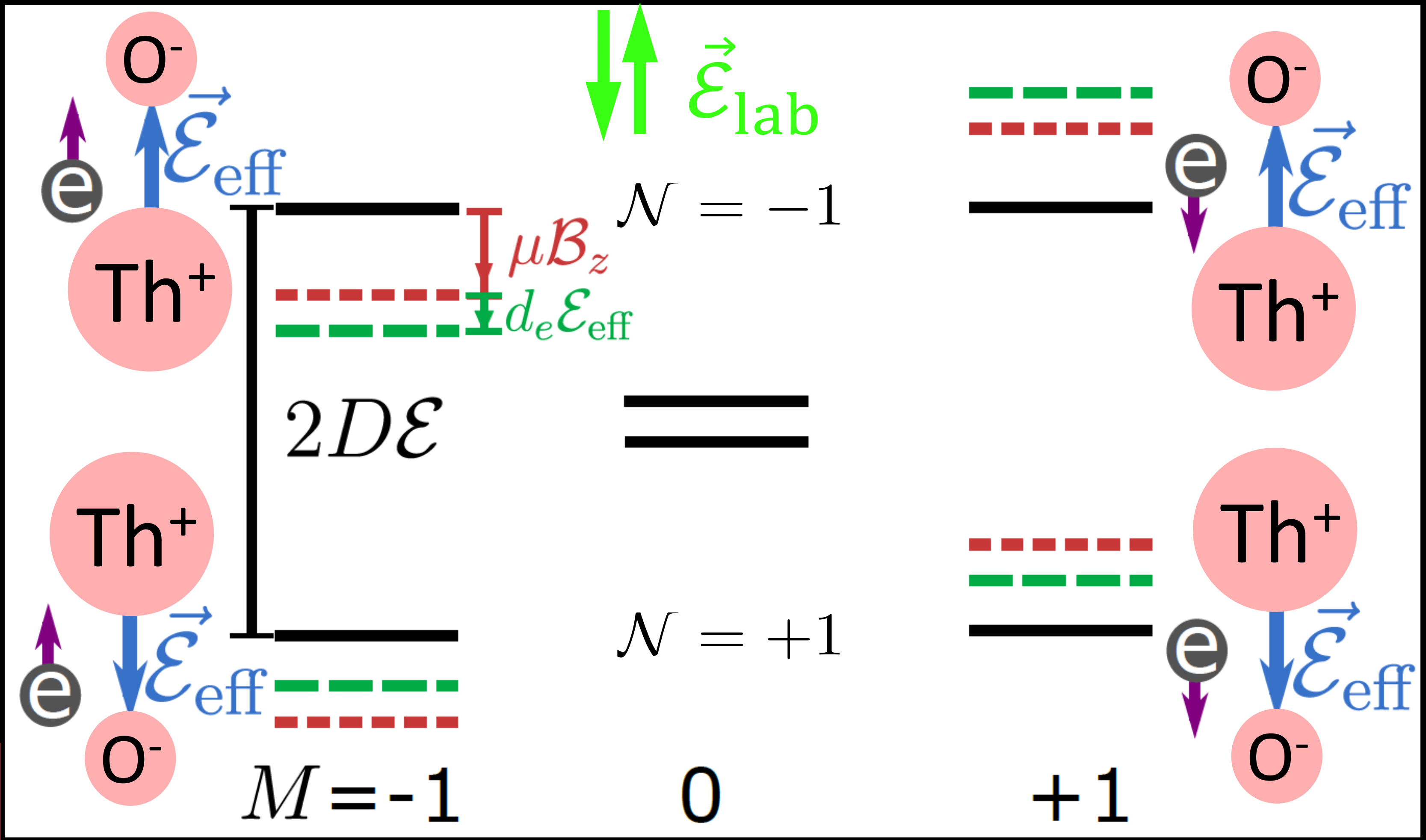
- Easily polarizable
- Can reverse $\vec{\mathcal{E}}_{\text{eff}}$ by either reversing:
  - Molecule-orientation, $\mathcal{N}$;
  - Lab electric field, $\vec{\mathcal{E}}_{\text{lab}}$



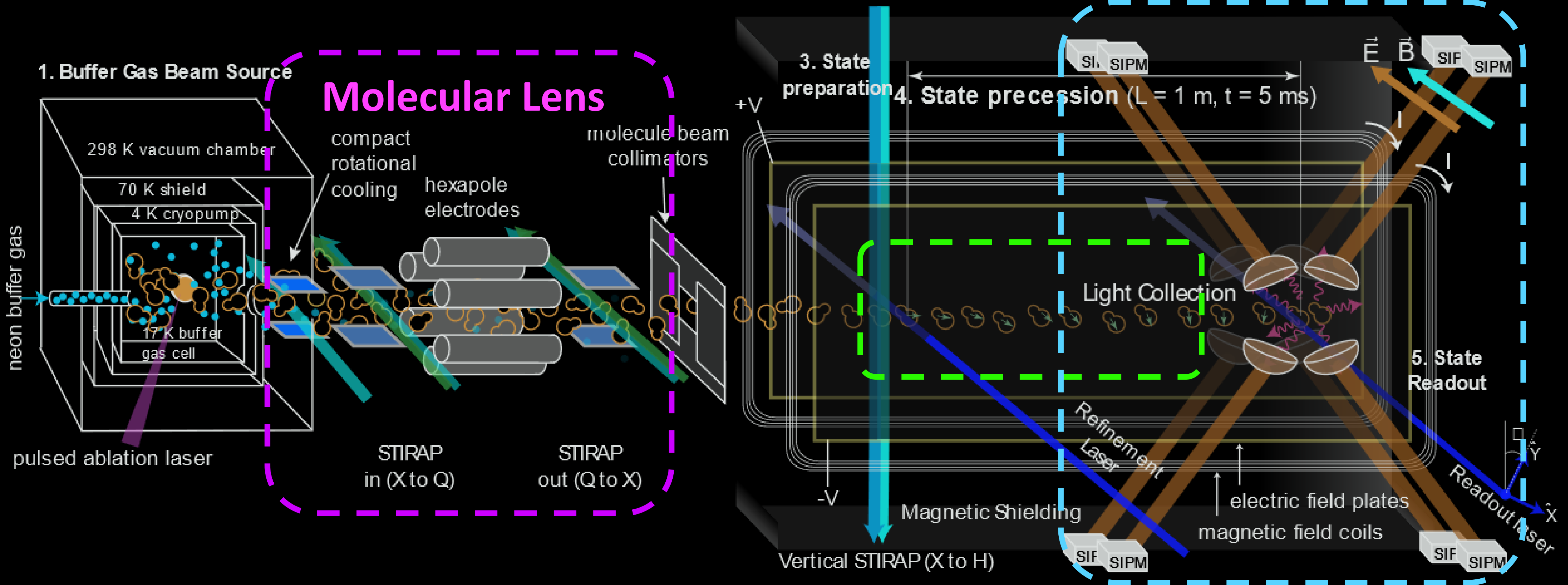
$H\ ^3\Delta_1$ state of ThO: Excellent for EDM Search

- Easily polarizable
- Can reverse $\vec{\mathcal{E}}_{\text{eff}}$ by either reversing:
  - Molecule-orientation, $\mathcal{N}$;
  - Lab electric field, $\vec{\mathcal{E}}_{\text{lab}}$

$$d_e \mathcal{E}_{\text{eff}} = -\hbar\omega^{\mathcal{N}\mathcal{E}}$$



ACME III will improve EDM sensitivity by ~30 times



- Molecular Lens increases molecular flux by x19 times
- x5 times longer spin precession time
- Better detection efficiency w/ SiPM and improved collection optics
- Additional technical improvements to control systematics and noises

Tested upgrades for ACME III

Upgrades	Signal Gain	EDM Sensitivity Gain
Electrostatic Lens	19	4.3
Increased Precession Time	.2	2.1
SiPM Detector & Photon Collection Upgrade	4	2
Timing Jitter Noise Reduction	1	1.7
Total	12	30

Figure of merit:

$$\frac{1}{\Delta d} \propto E\tau\sqrt{\dot{N}T}$$

X19 larger $\dot{N}$ ✓ XW, et al, *NJP* 24 073043 (2022) XW, et al, *NJP* 22 023013 (2020)

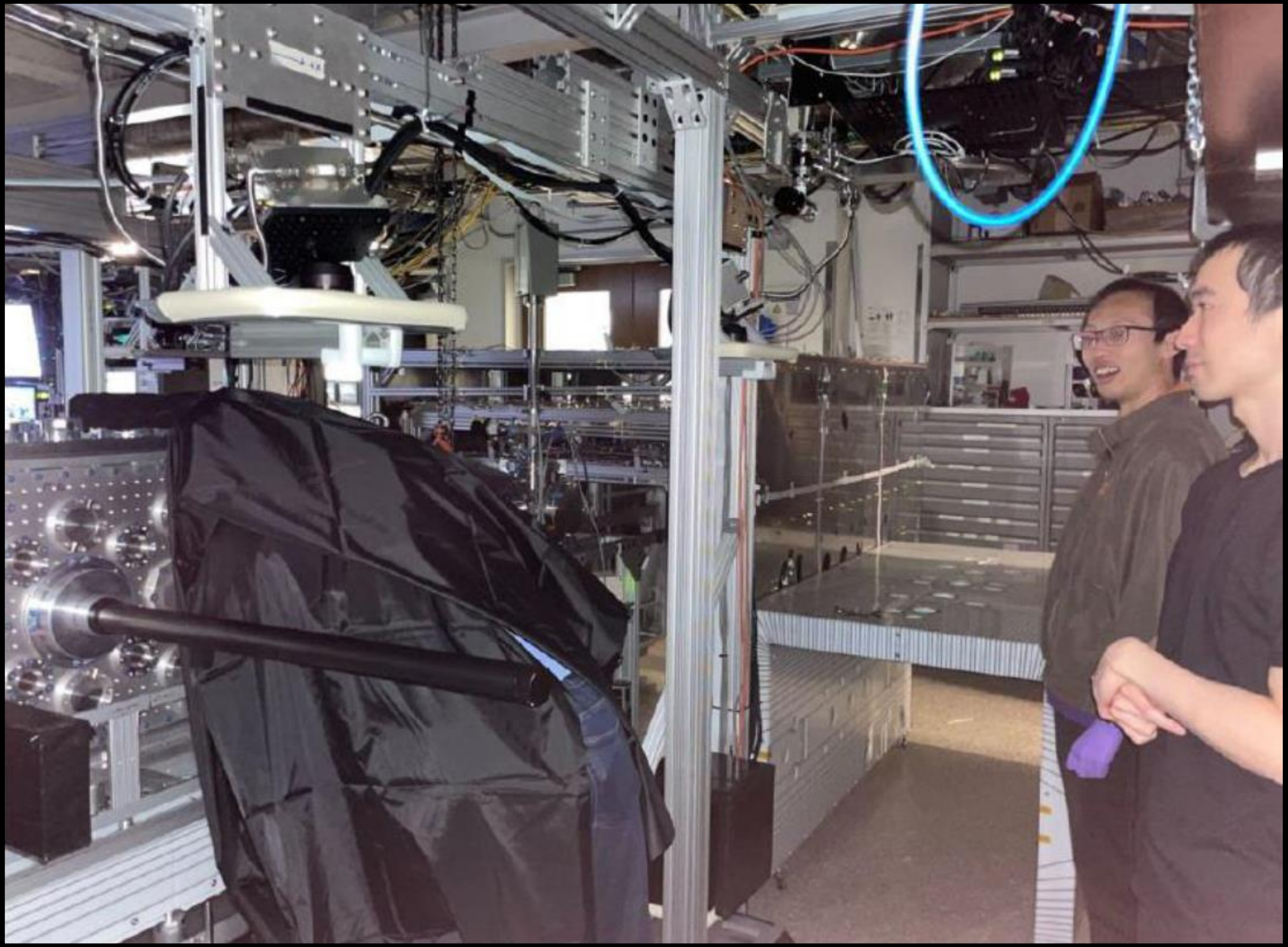
x 5 longer τ ✓ D.Ang, XW, et al, *PRA* 106 (2), 022808 (2022)

x4 larger $\dot{N}$ ✓ T Masuda, XW, et al, *Optics Express* 29, 11, 16914 (2021)

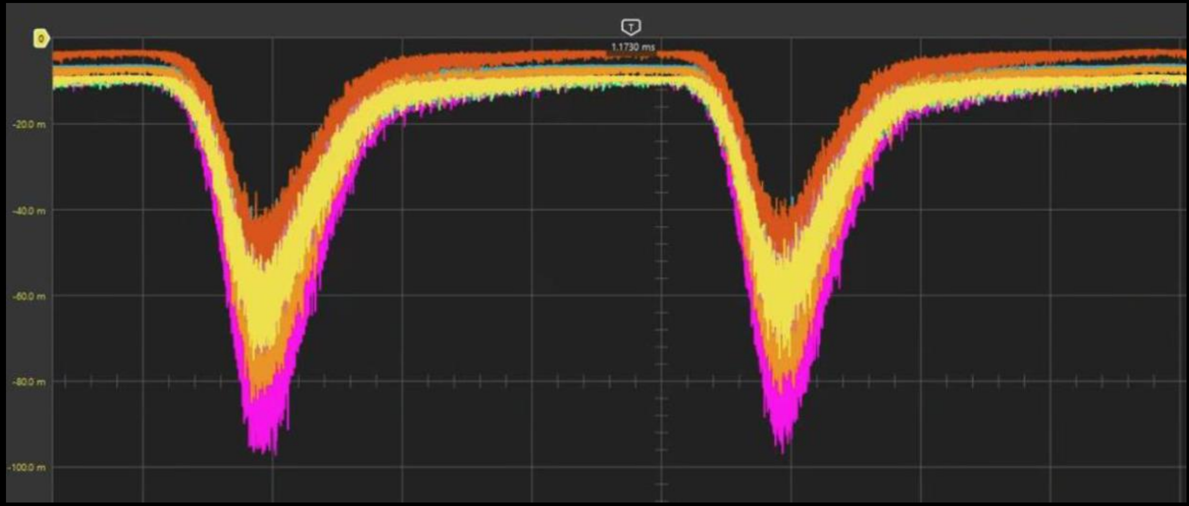
x3 reduction in noise ✓ C.Panda, XW, et al, *J. Phys. B* 52, 235003 (2019)

Projected EDM sensitivity gain over ACME II

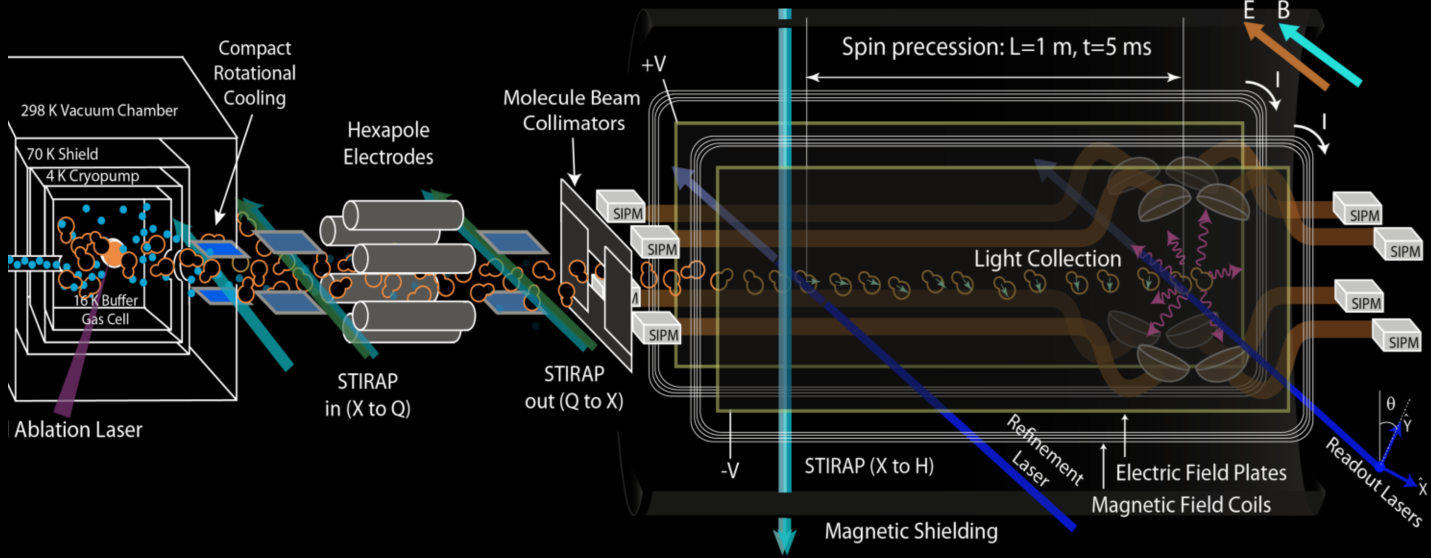
First molecular signal from ACME III beamline



- Signal rate is approximately what we have expected from all the upgrades
- Excellent prospects to probe eEDM at $<3\times10^{-31}$ *e·cm* level



State of the Art



ACME collaboration (Harvard, U Chicago, Northwestern)

Neutral ThO molecular beam:

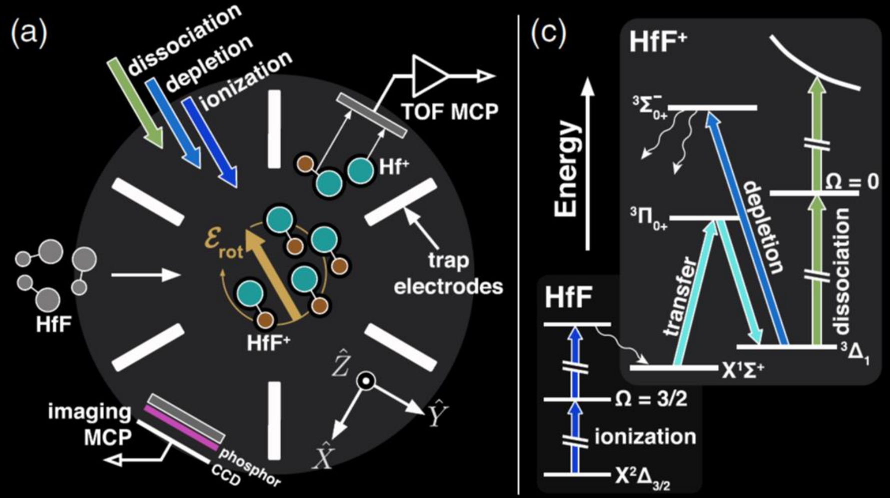
$|d_e|<1.1 \times 10^{-29}$ e cm

Nature 562, 355 (2018)

☺ Project to reach $<3 \times 10^{-31}$ e cm in the coming generation

☹ interrogation time is limited by EDM state lifetime (~ 4ms)

E. Cornell



JILA eEDM (Eric Cornell, Jun Ye)

Trapped HfF^+ molecular ions:

$|d_e|< 5 \times 10^{-30}$ e cm

Science **381**, 46 (2023)

☺ ~1s spin precession time

☹ Severely limited by N, due to Coulomb repulsion

One Strategy: Neutral Molecules in a Trap!

neutral YbF

Mike Tarbutt
@ Imperial College London

neutral BaF

Steven Hoekstra
@ NL-eEDM collaboration

neutral RaF

Ronald Garcia-Ruiz,
John Doyle, Nick Hutzler

☺ YbF, BaF, and RaF can be nominally laser cooled!

(Alkaline-earth fluoride molecules are analogous to the Alkali atoms)

☺ Perspective to load into Magneto-Optical Trap, optical dipole trap, and optical lattice

☹ They do not possess the powerful $^3\Delta_1$ co-magnetometer structure (which both ThO and HfF^+ have)

Polyatomic Molecules

- Take a molecule that is like Alkaline-earth fluoride, but with –F atom replaced by a ligand group that produces co-magnetometer structure (e.g. –OH, -OCH_3)

✓ Alkaline-earth element stays as photon-cycling center
✓ Ligand group provides the parity doublet

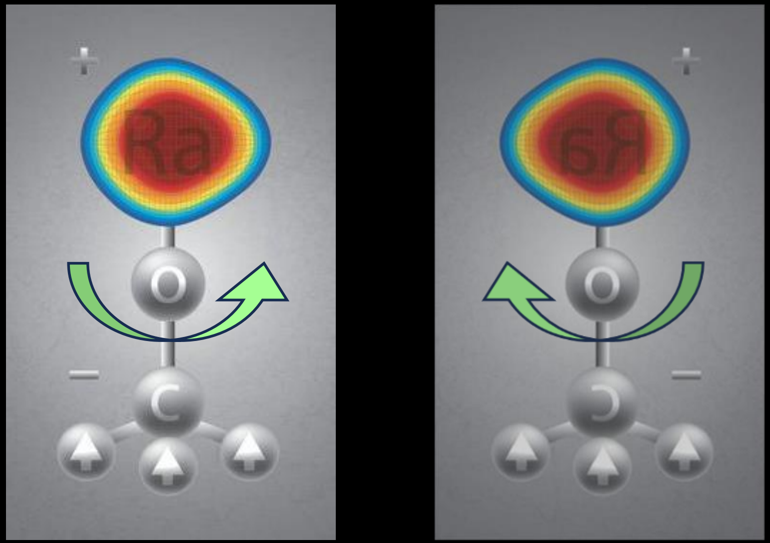


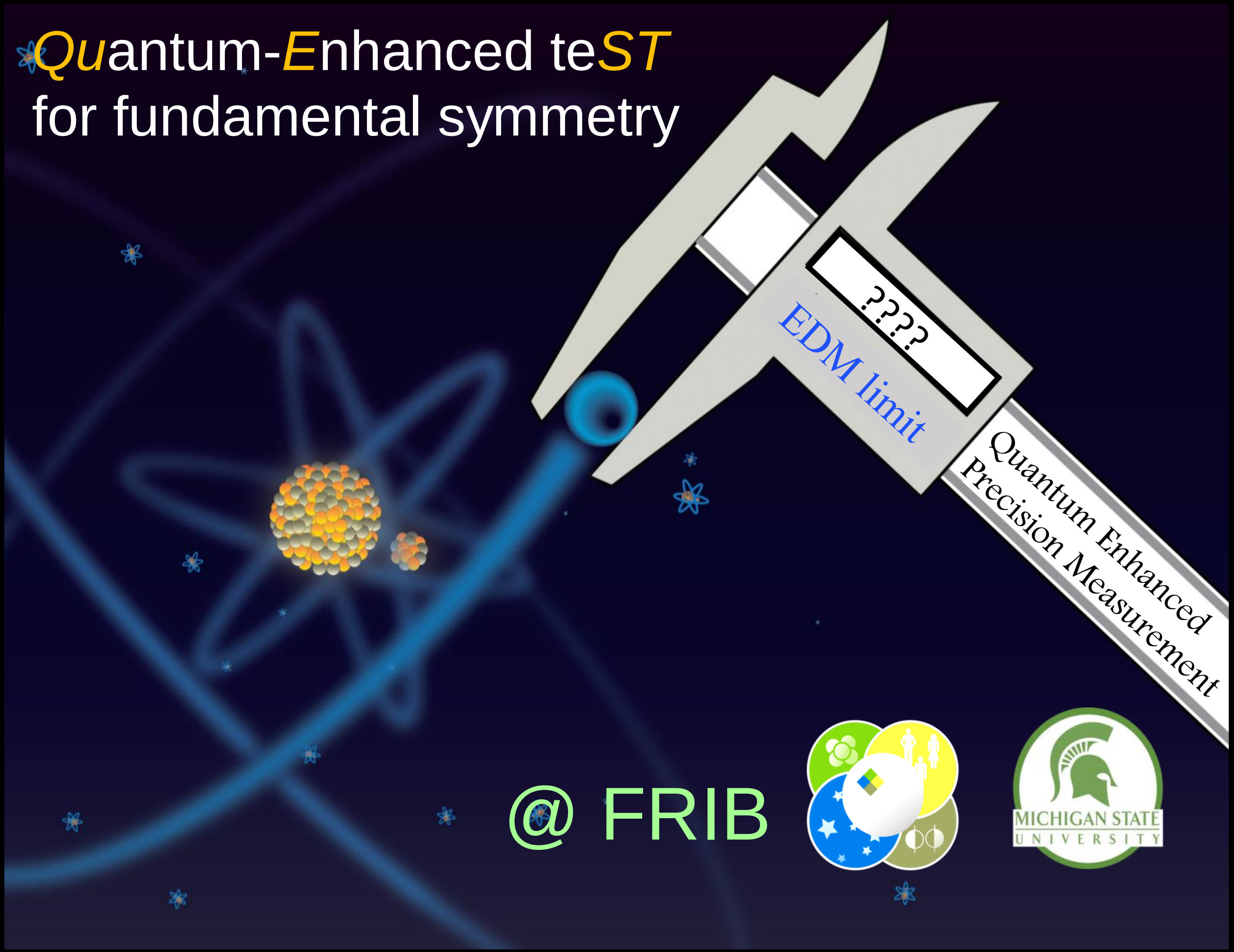
Image modified from Hutzler Group @ *CalTech*

$RaOCH_3$

N. Hutzler @ *CalTech*

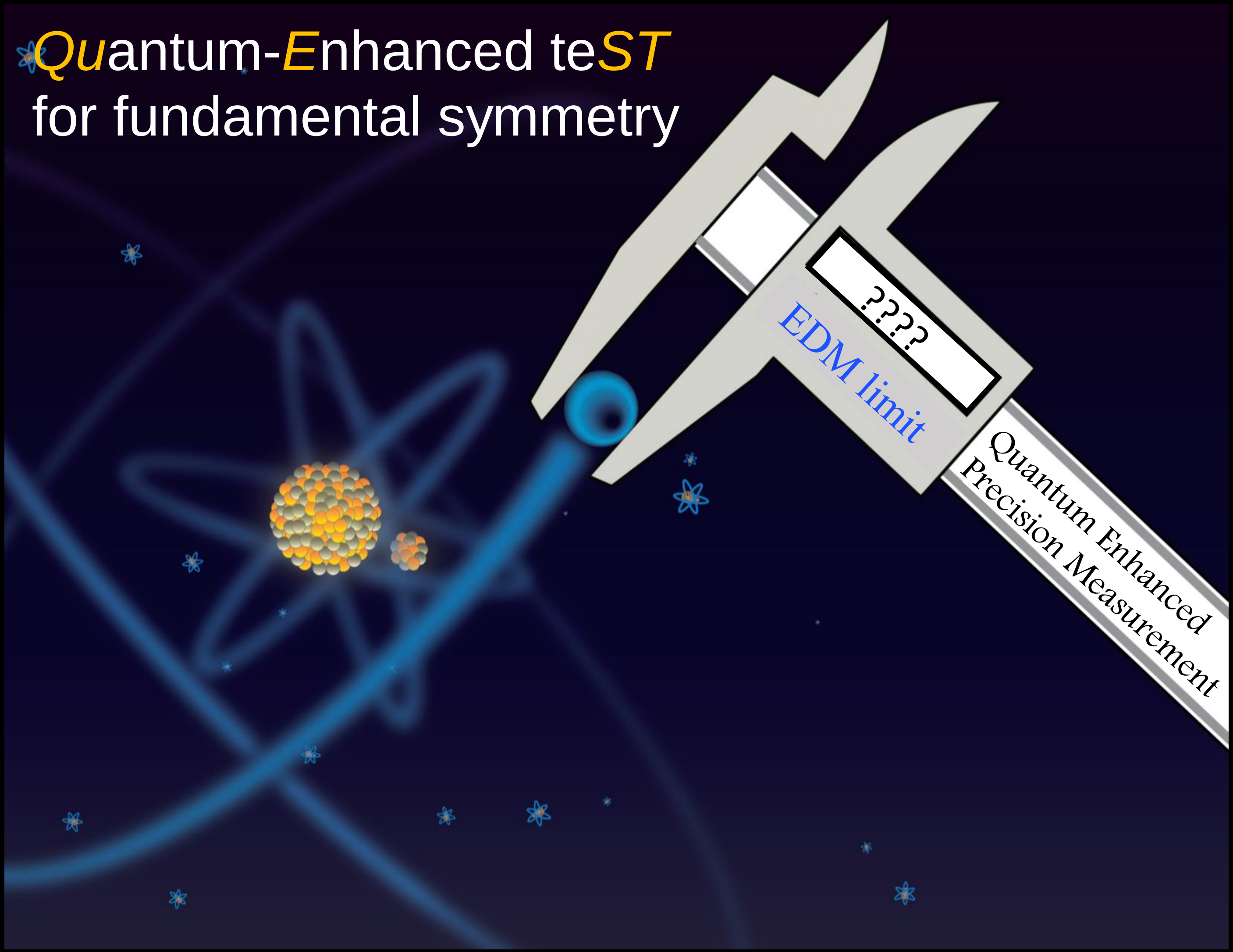
I. Kozyryev and N. Hutzler, PRL 119, 133002 (2017)

Fall 2023: *QuEST* Project Starts at FRIB/MSU



To establish a new generation EDM measurement that outperforms the current best limits by several orders of magnitude

Fall 2023: *QuEST* Project Starts at FRIB/MSU

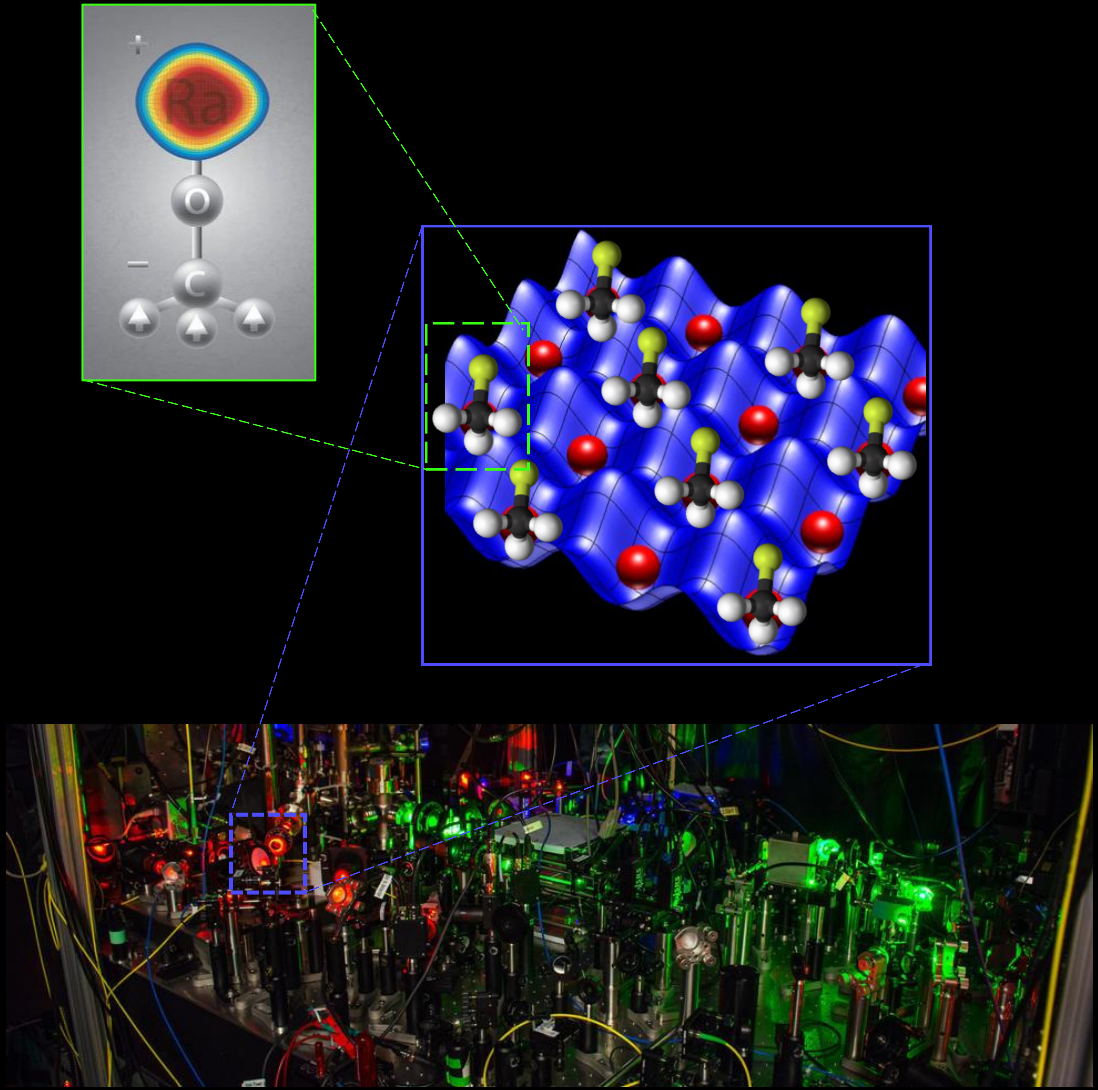


Rare Isotope Access at FRIB:

Octupole deformation from pear-shaped nuclei (^{225}Ra, ^{223}Fr)→ nuclear enhancement to *T*-violating new physics

Facility for Rare Isotope Beams (FRIB) Michigan State University

Fall 2023: *QuEST* Project Starts at FRIB/MSU



‘Conventional’ Laser Cooling of Molecules

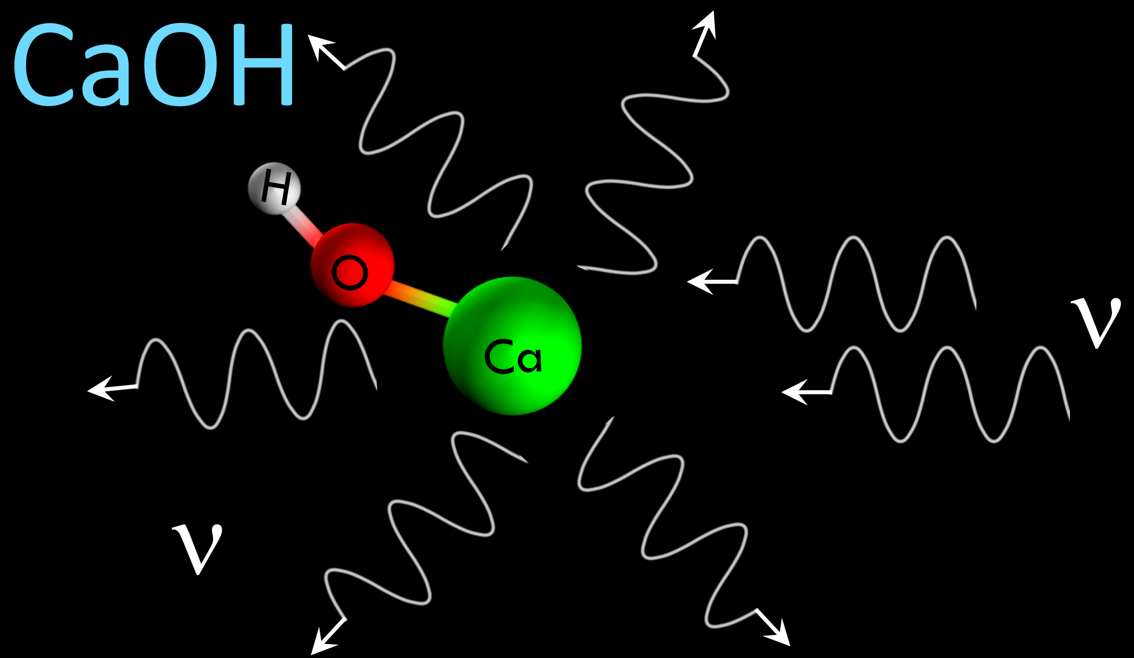
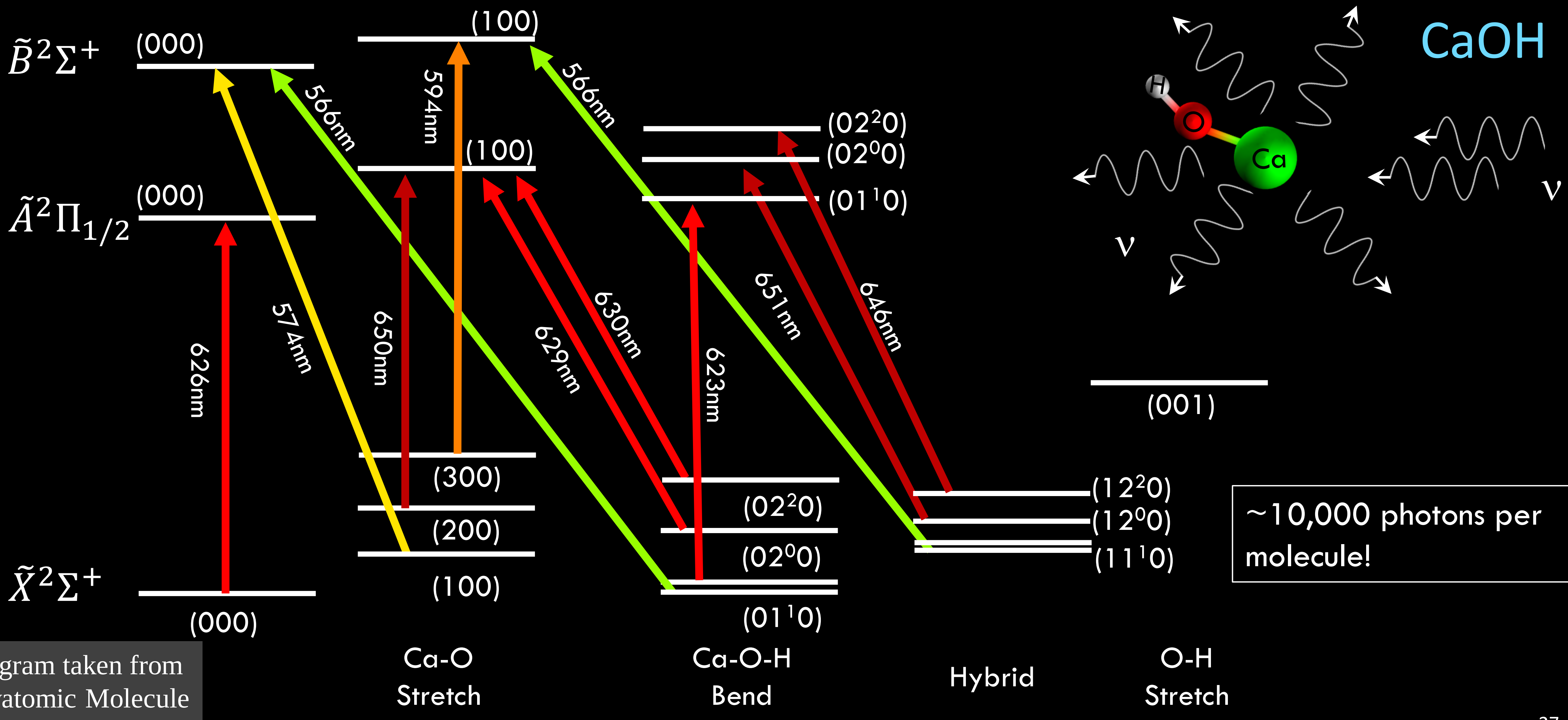


Image taken from Polyatomic Molecule Project @ *Harvard*

John Doyle @ Harvard
David DeMille @ U Chicago
Jun Ye @ JILA/Boulder
……

- Photon-cycling on Optical-Cycling-Center (e.g. Ca, Sr, Yb, etc)
- Many Atom-Cooling techniques are ready to apply
- Reach ~5 micro-Kelvin (for CaF).
- But, it requires ~12 Lasers or more for polyatomic molecules…. ☹

Probably Need More than 12 Lasers to Re-pump!



~10,000 photons per molecule!

Diagram taken from Polyatomic Molecule Project @ *Harvard*

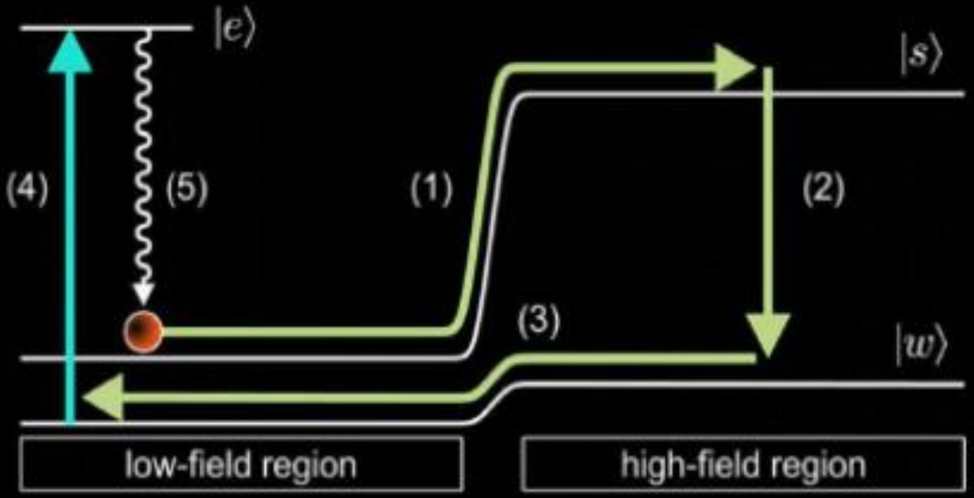
Opto-electric Sisyphus Cooling

- Only need about 1 laser!! ☺
- Only require to scatter ~100 photons
- Rely on E-field to remove energy of molecules
- Optical transition pumps molecules from weaker Stark-states (|w>) to stronger Stark-states (|s>).
- Demonstrated for CH_2O, CH_3F, reaching ~300 micro-Kelvin!

But, cooling rate is very slow, ~10 Hz, vs. ~10 MHz for conventional laser cooling ☹

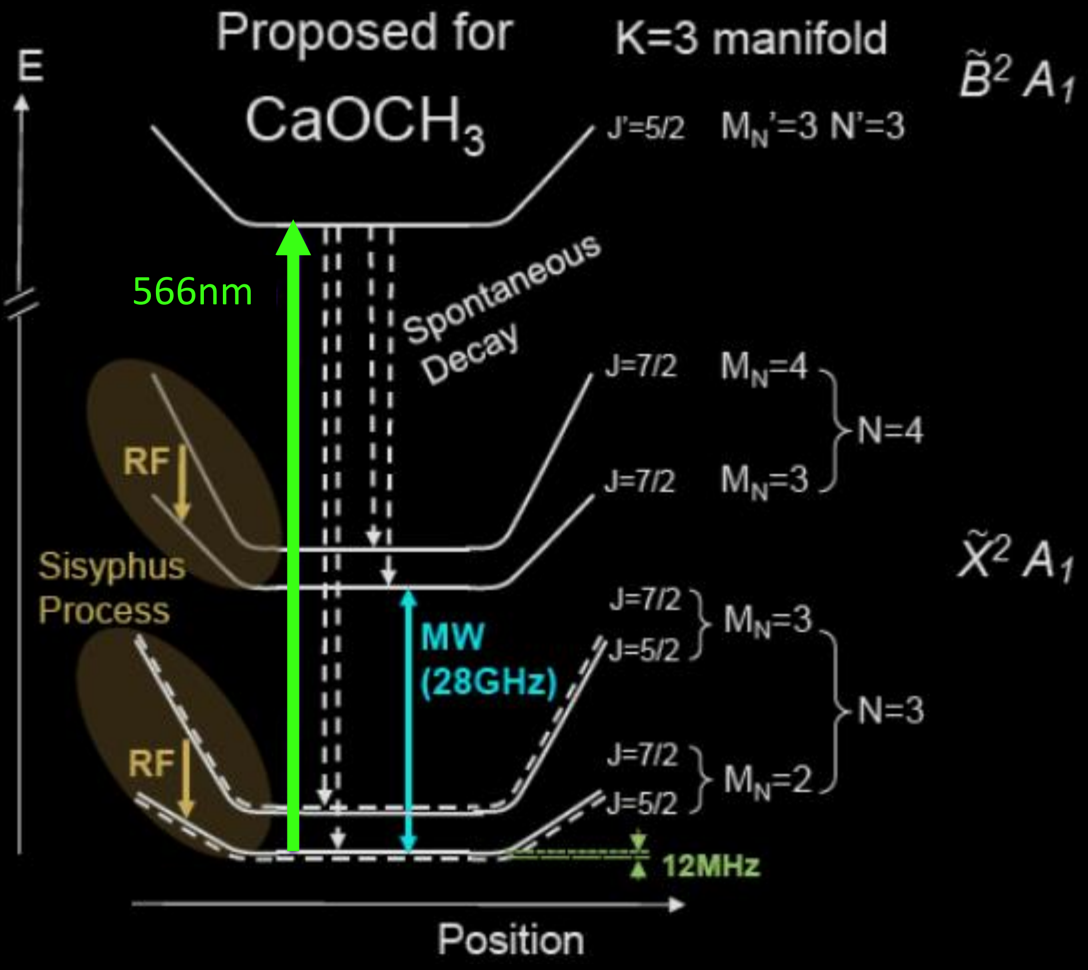
M. Zeppenfeld & G. Rempe @ MPQ

Nature 491, 570–573 (2012),
Phys. Rev. Lett. 116, 063005 (2016)



Would it be Feasible to Marry the Two Most Successful Methods for Direct Cooling?

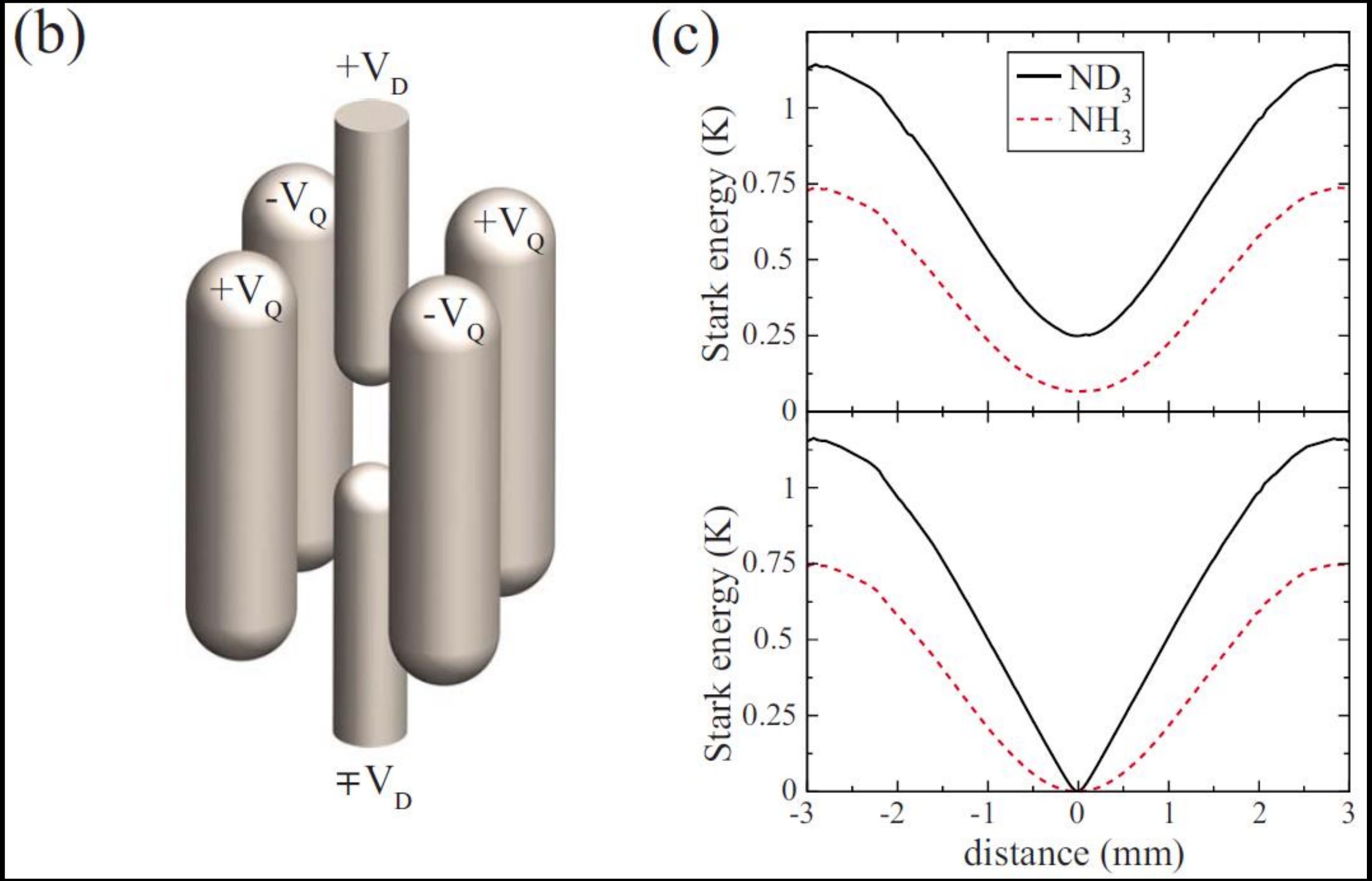
YES!



XW, manuscript in prep. (2024)

- ✓ x10^6 faster cooling rate!!
- ✓ Only need ~1 re-pumping lasers, instead of 15 or more!!!
- ✓ Fundamental cooling limit is the photon recoil at ~ 400nK

Electric Ioffe-Pritchard trap for polar molecules

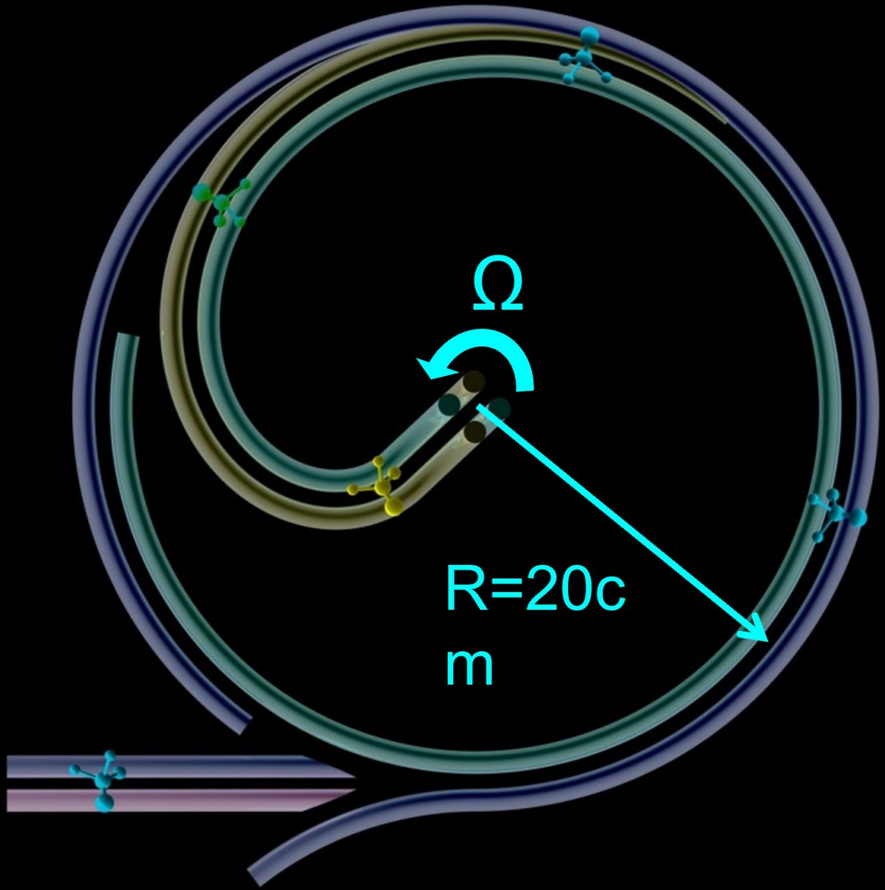


PRA 79, 051401 (R) 2009

Schnell & Meijer @ *FHI*, Berlin

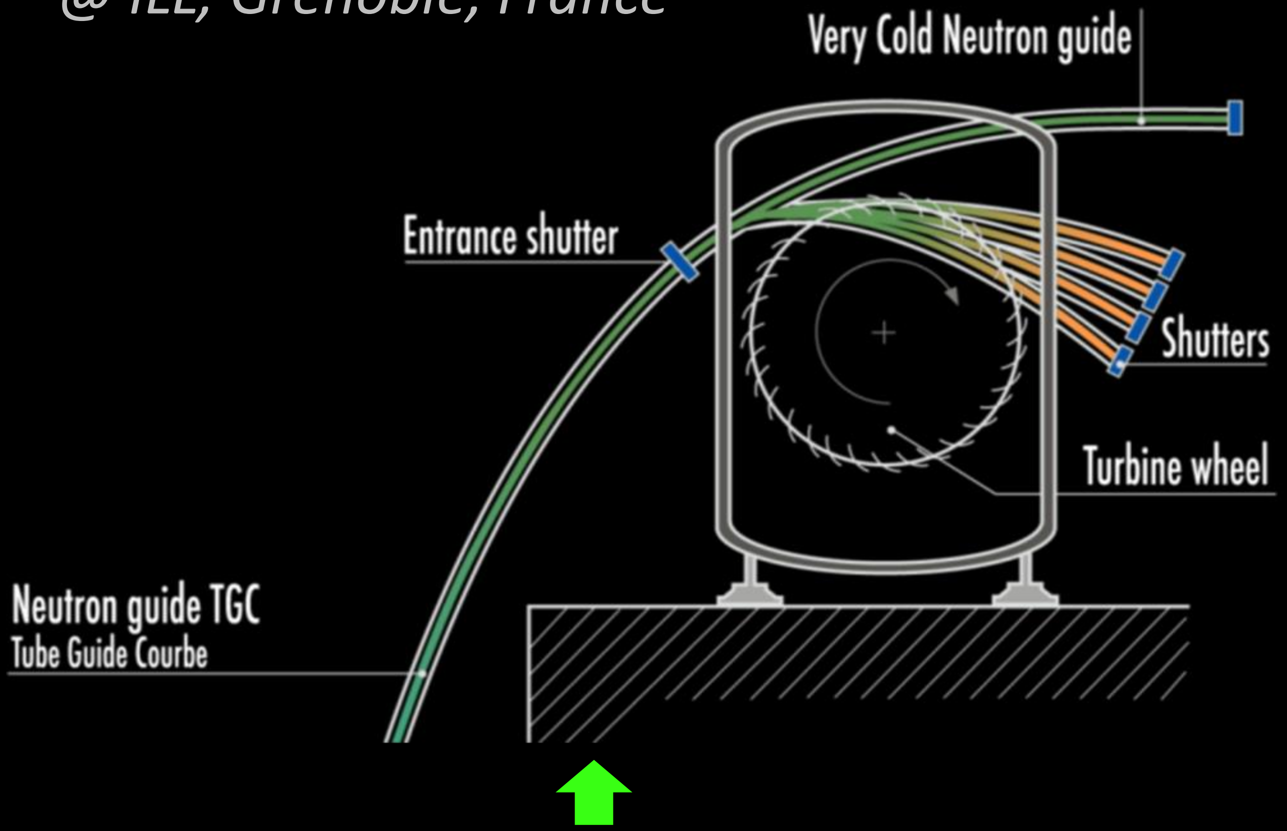
Decelerate Molecular Beams by Centrifugal Force!

Demonstrated in my doctoral thesis



Same principle was used for producing ultracold neutron beams

@ ILL, Grenoble, France

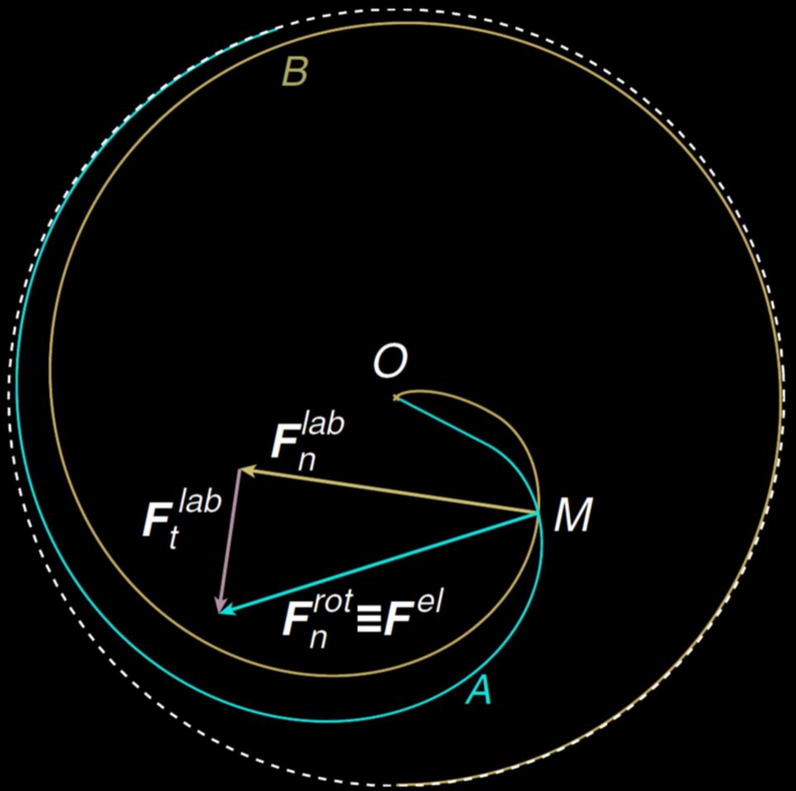


XW, PhD Thesis, TU München (2017)
XW et al., Science 358, 645 (2017)
S Chervenkov, XW et al., PRL, 112, 013001 (2014)

For searching Neutron EDM!

Physics in the Lab Frame

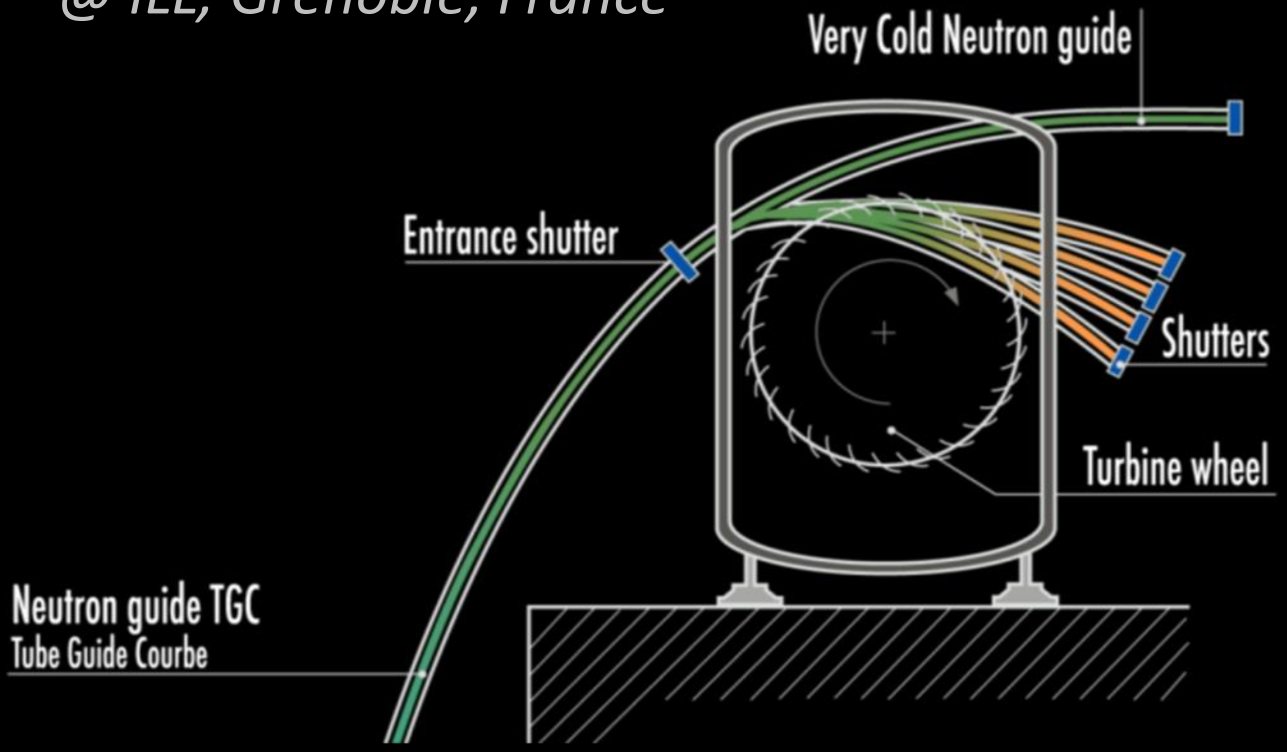
A: trajectory in the rotating frame
B: trajectory in the lab frame



XW, PhD Thesis, TU München (2017)
S Chervenkov, XW et al., PRL, 112, 013001 (2014)

Same principle was used for producing ultracold neutron beams since 1990s

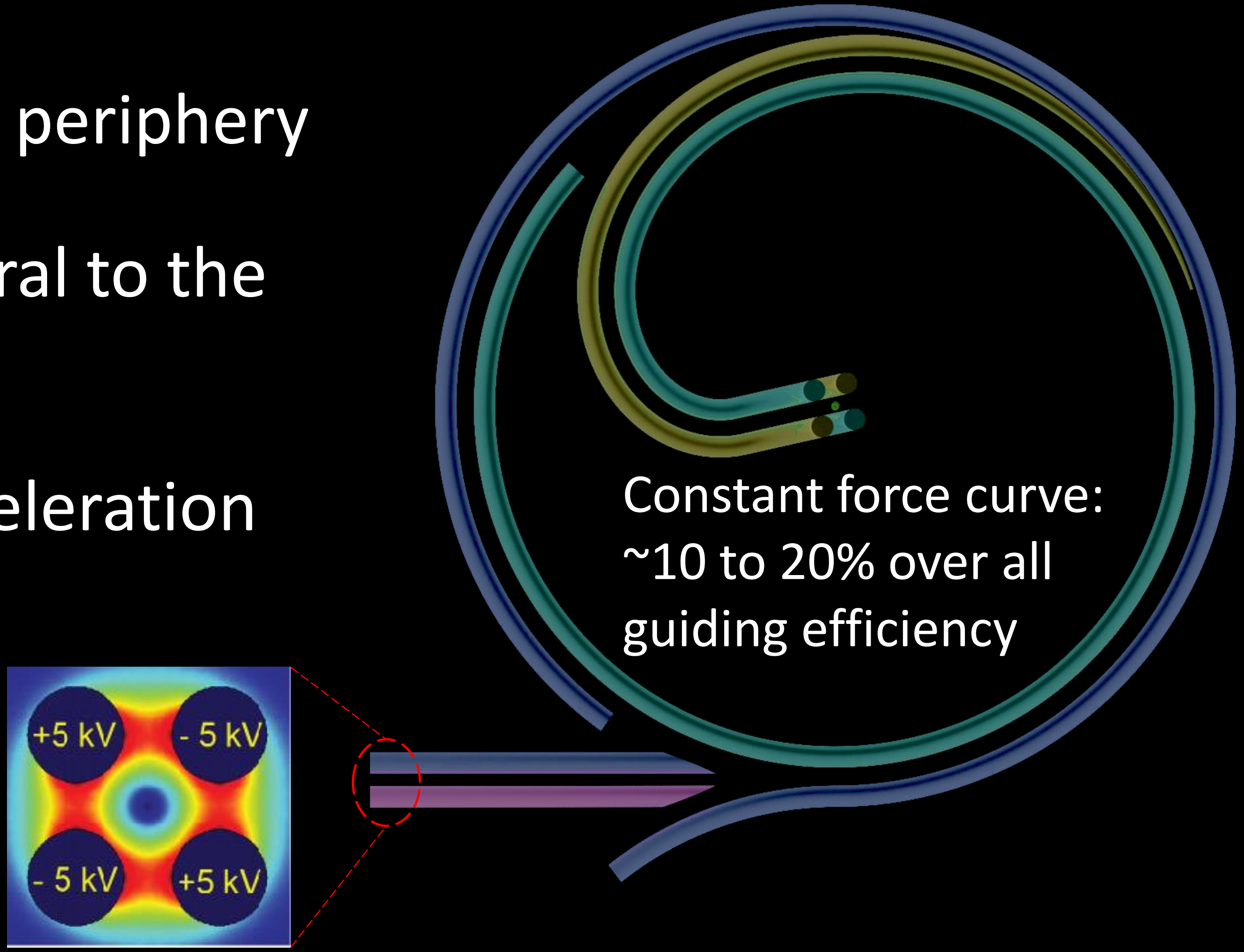
@ ILL, Grenoble, France



For searching Neutron EDM!

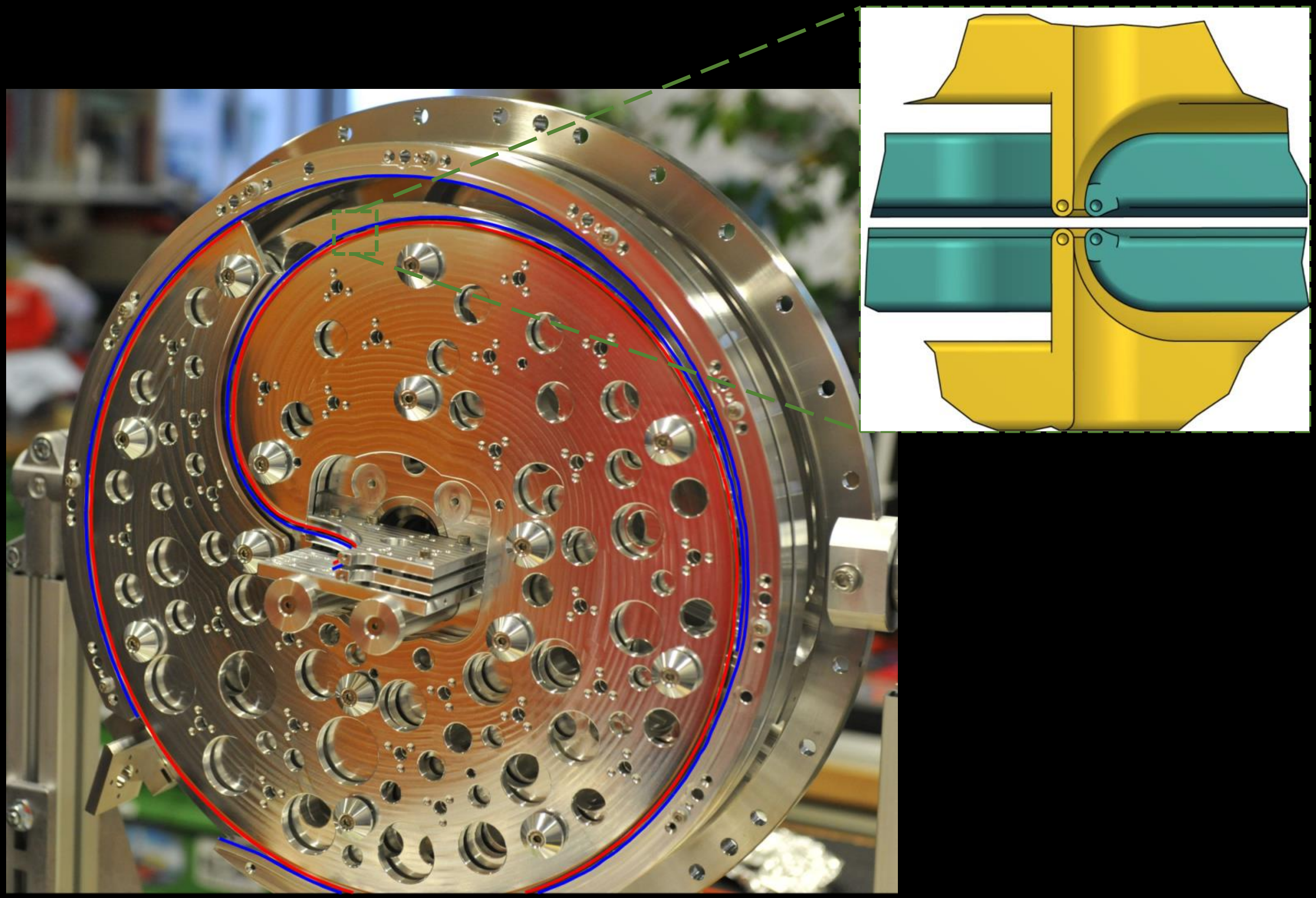
High efficiency

- Static electrodes at the periphery
- Rotating electrodes spiral to the centre
- Enable continuous deceleration



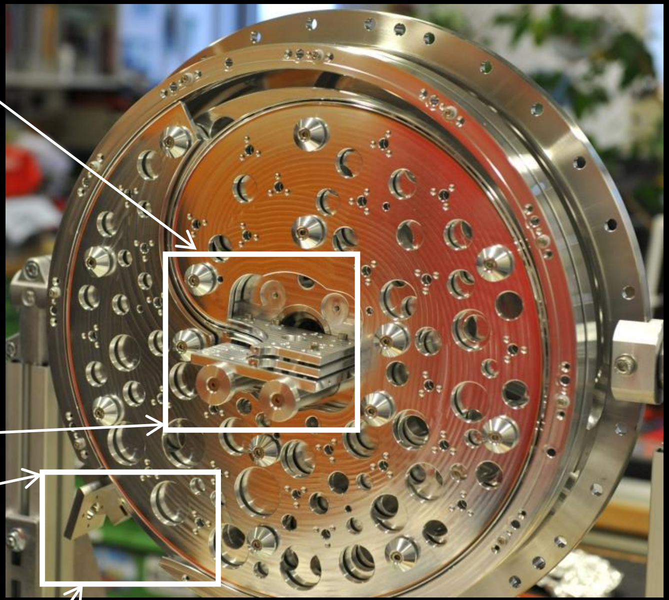
Constant force curve:
~10 to 20% over all
guiding efficiency

How does a Real Centrifuge Look Like?



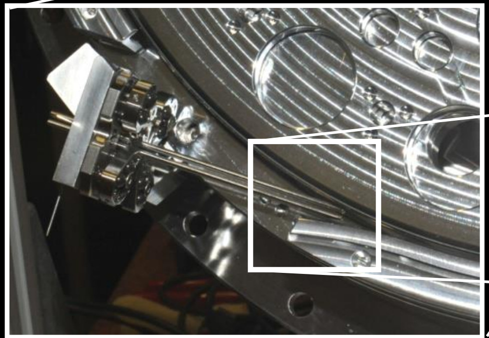
How does a Real Centrifuge Look Like?

Bend-up

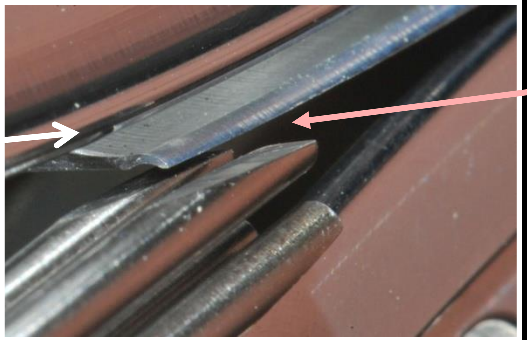


V_p= 174 mph
(*or* 280 km/h)

Injector



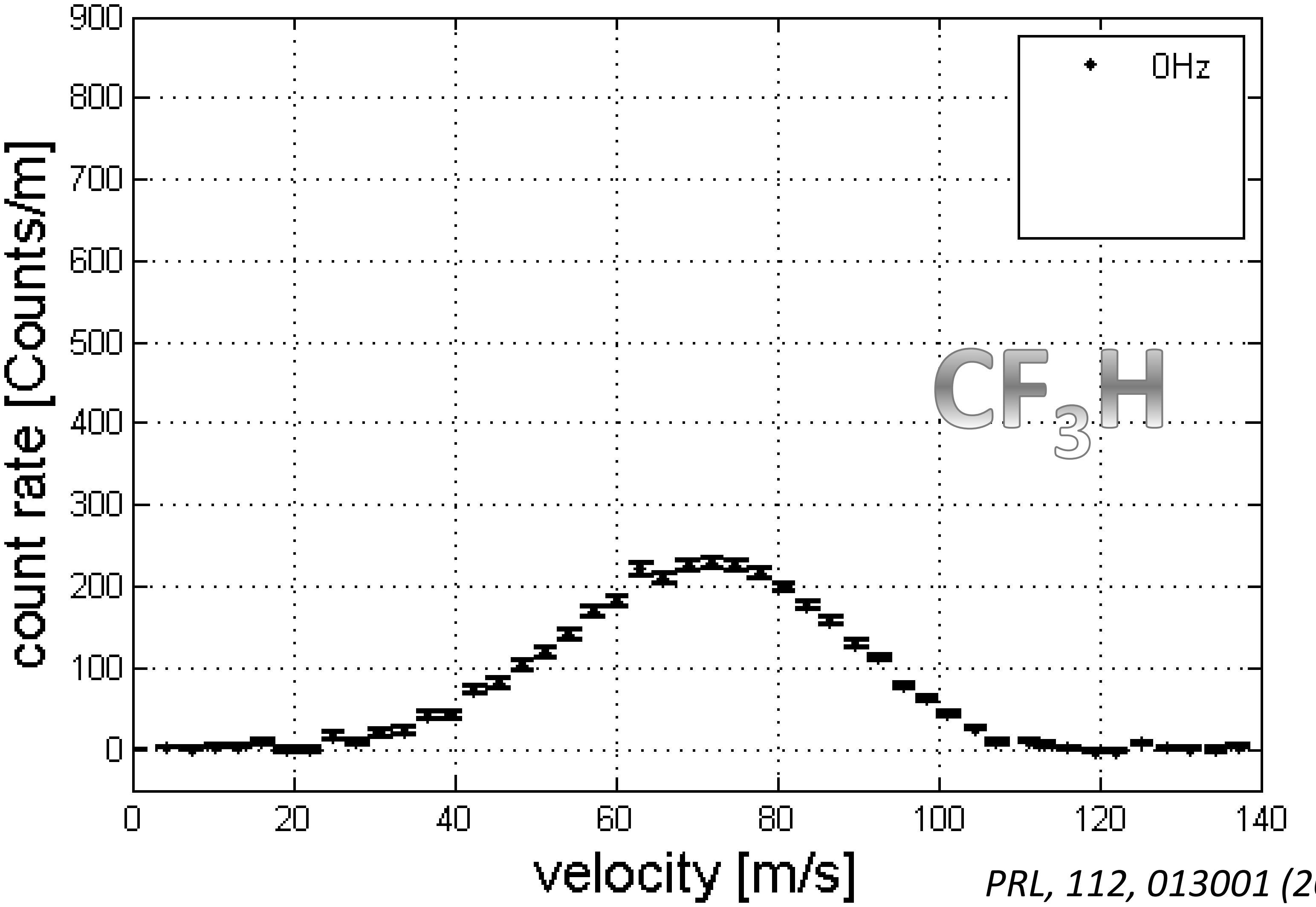
Injector (close-up)



Rotating electrode

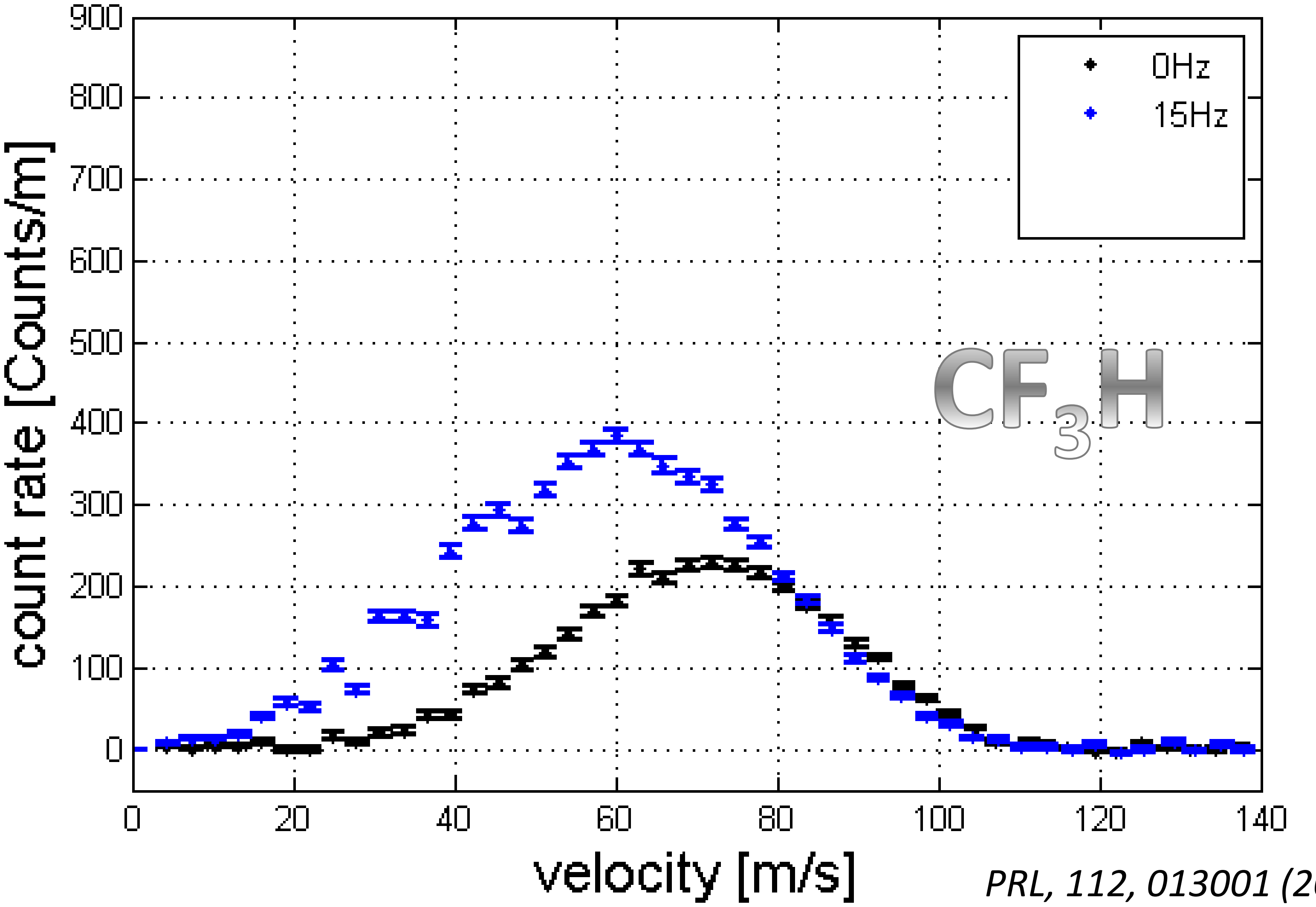
200μm gap

Deceleration



PRL, 112, 013001 (2014)

Deceleration



PRL, 112, 013001 (2014)

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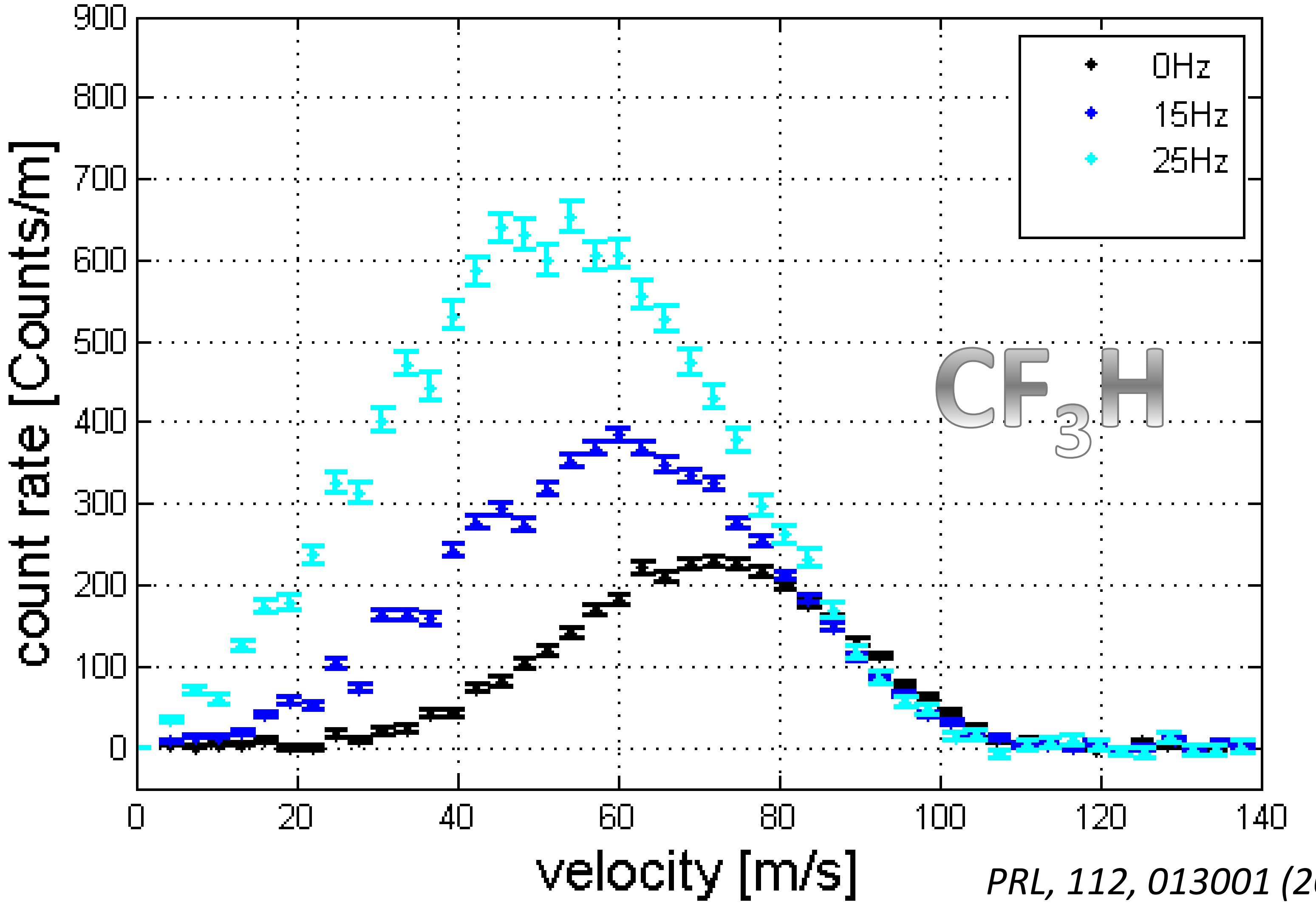
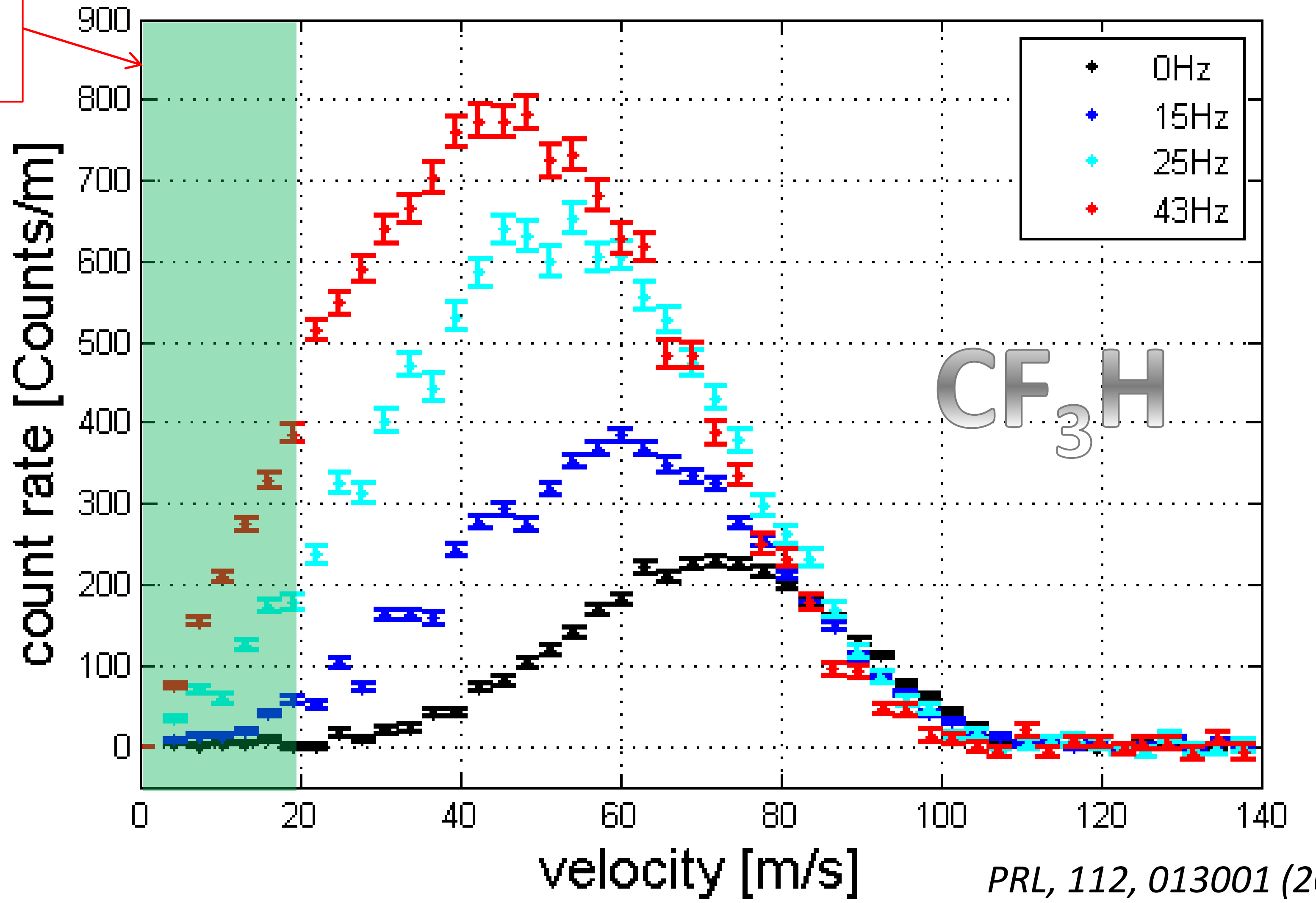


PRL, 112, 013001 (2014)

Deceleration

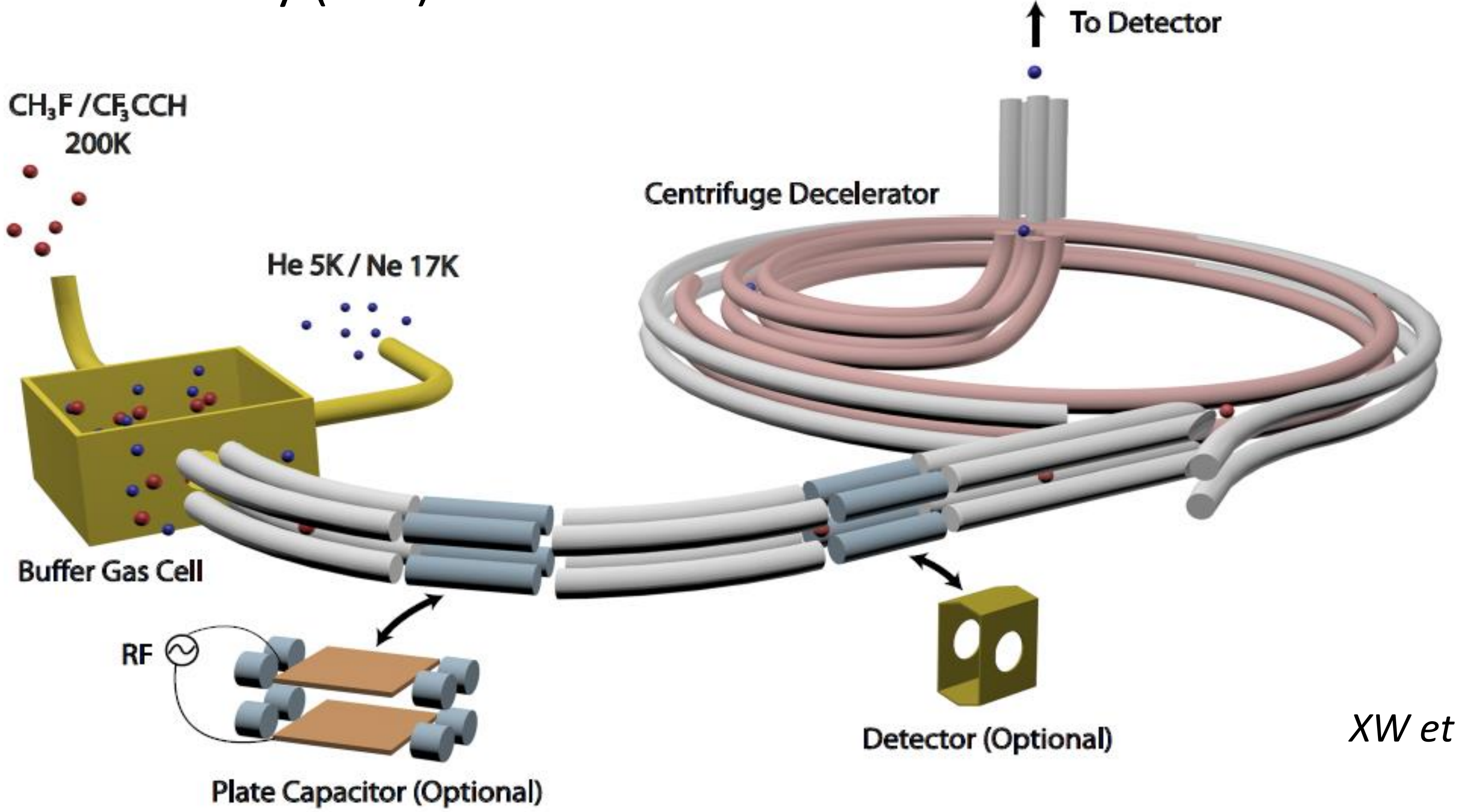
~ 1K (*or* 86μeV), trappable!



PRL, 112, 013001 (2014)

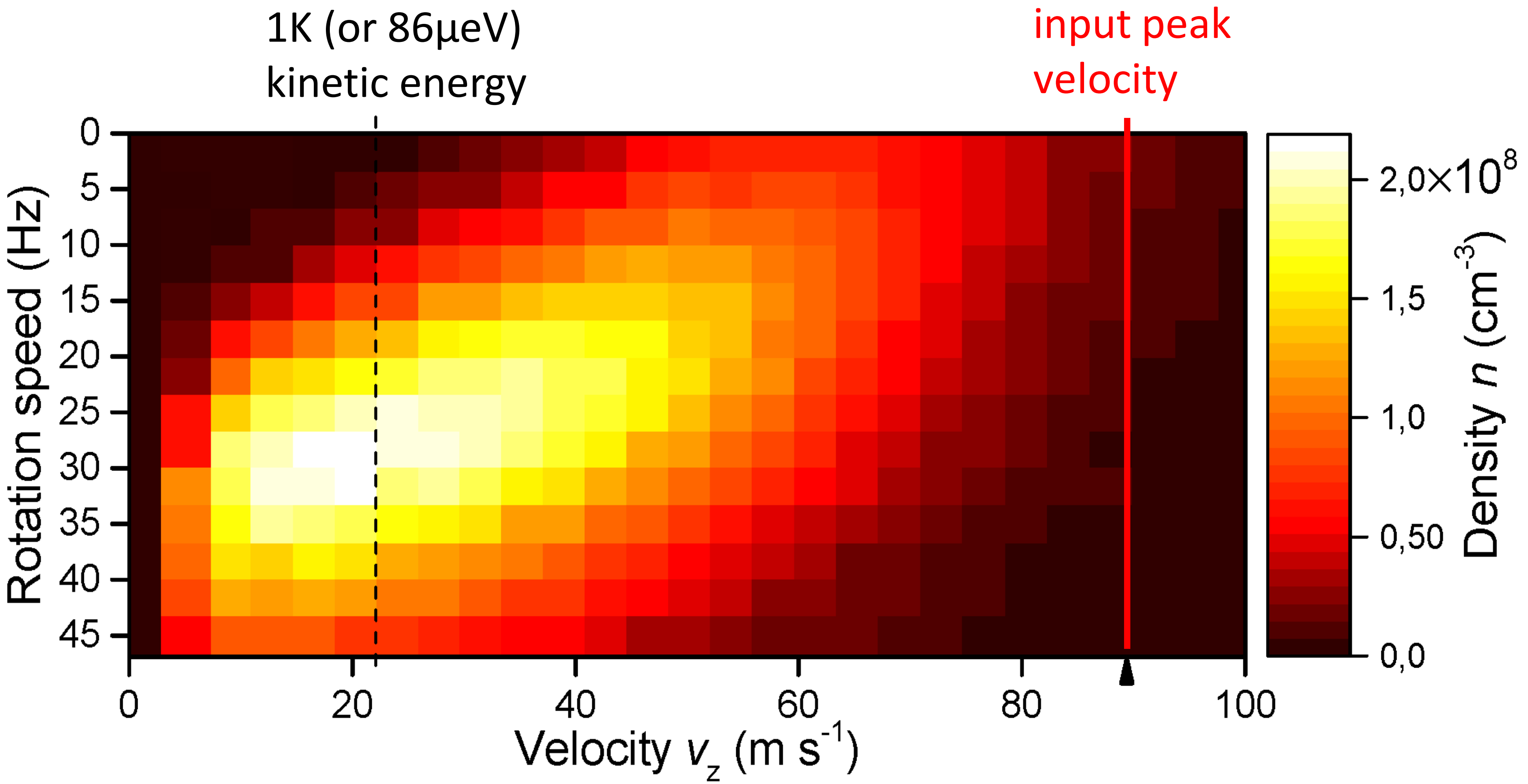
Cryofuge

- Combine a cryogenic beam source with the centrifuge
  - flux (<1K) ≈ $1.2 \times 10^{10} s^{-1}$
  - density (<1K) ≈ $1.0 \times 10^{9} cm^{-3}$



XW et al., Science 358, 645 (2017)

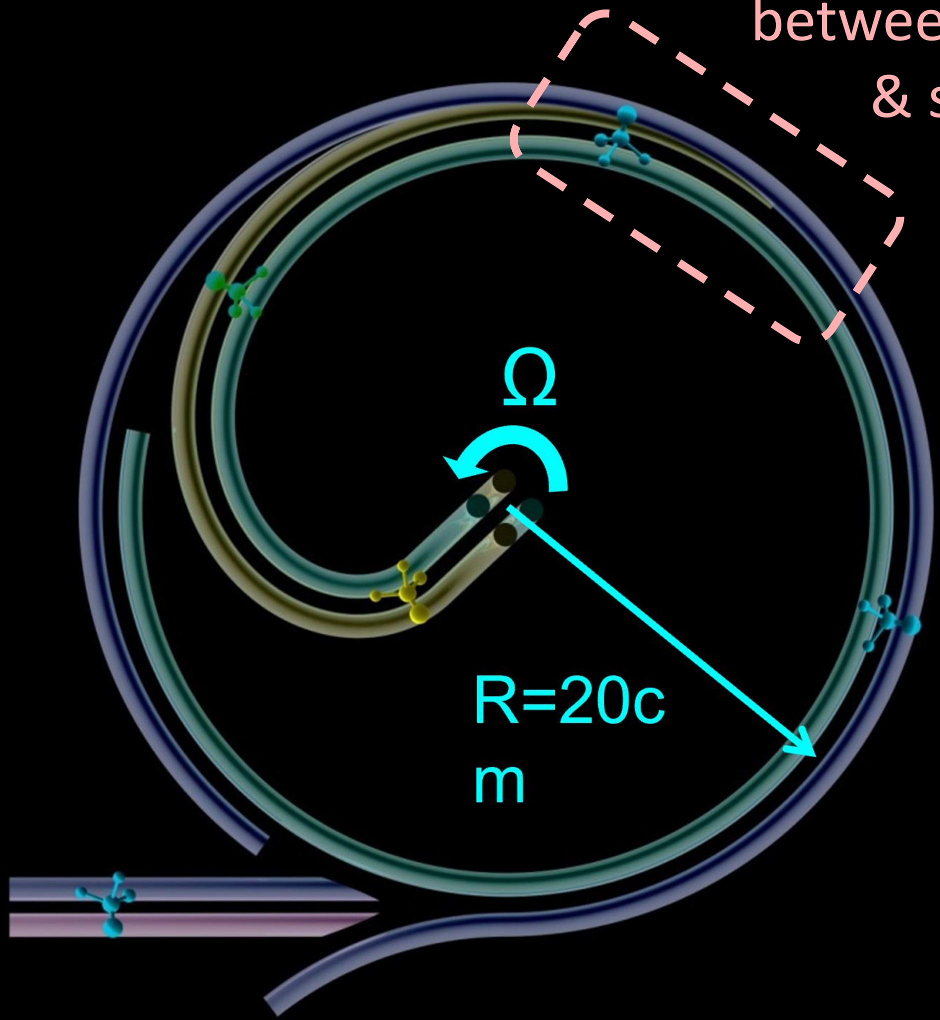
Deceleration results



XW et al., Science 358, 645 (2017)

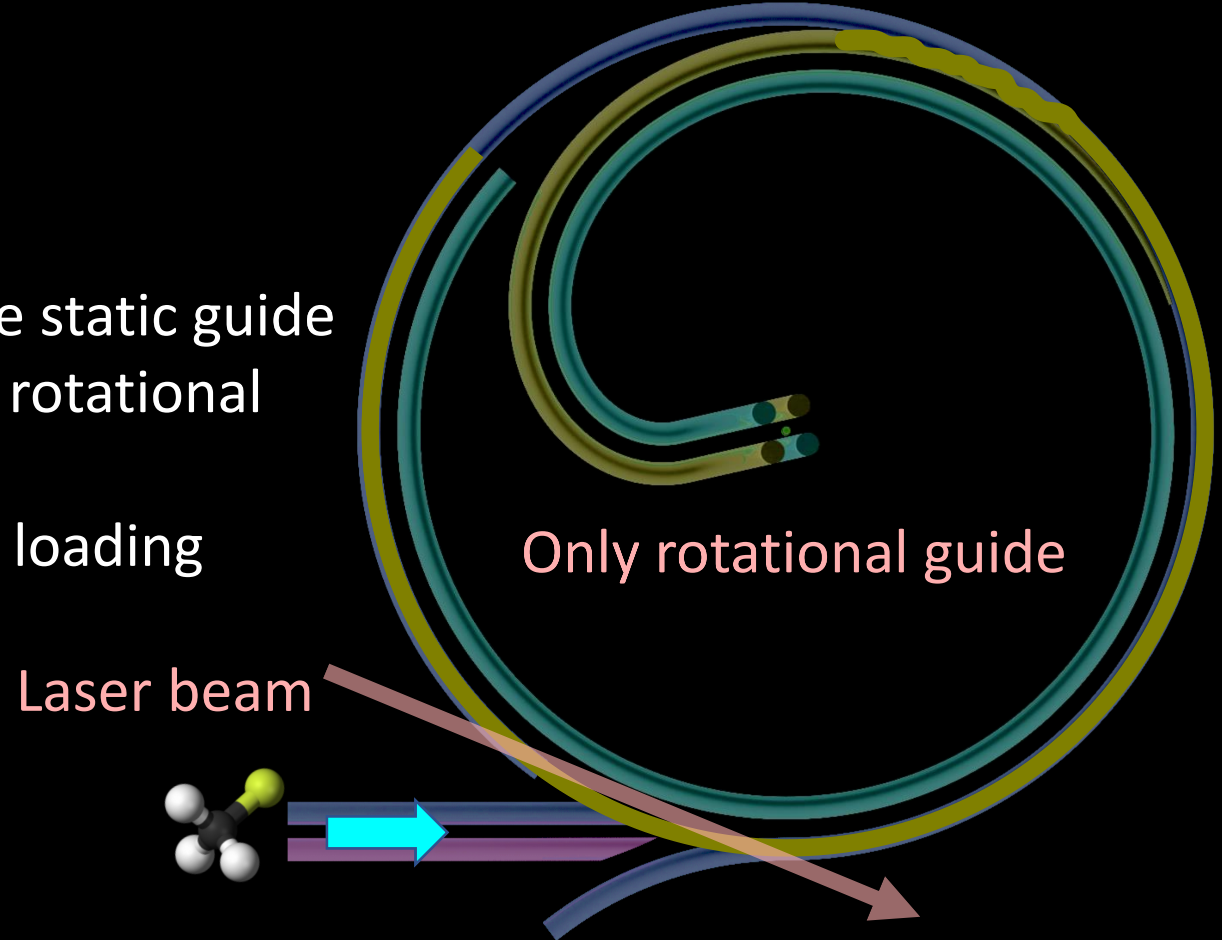
Plan for Simplifying the Mechanical Structure

Key Challenge: Interface between rotational & static guides



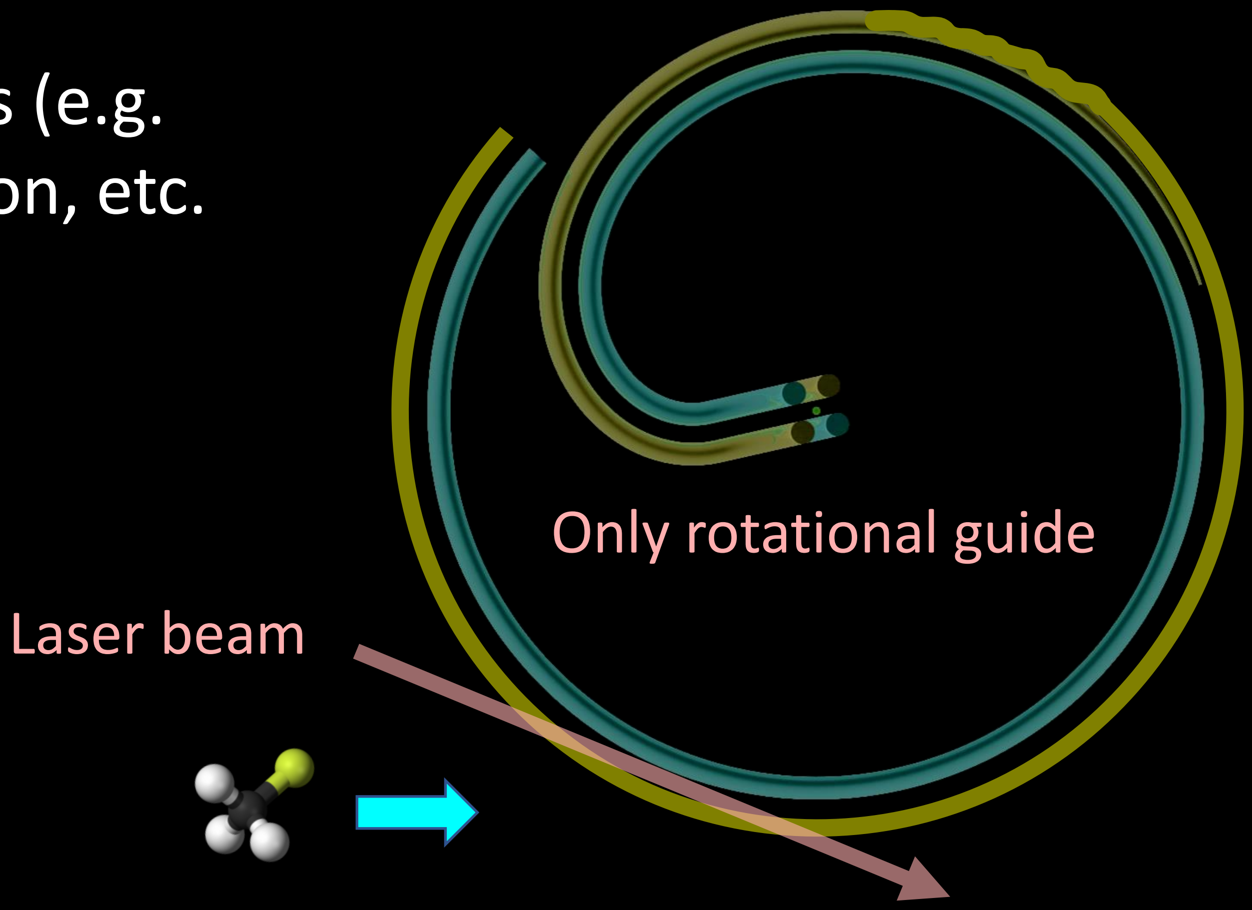
- Remove static guide
- Extend rotational guide
- Optical loading

Solution: completely avoid the static guide



Truly Generic Approach to Decelerate Particles

- The guide can be either electric, or magnetic, or surface scattering
- Applicable: polar molecules, atoms (e.g. Hydrogen and Tritium), cold neutron, etc.
- Optical loading can be replaced by microwave/RF pumping



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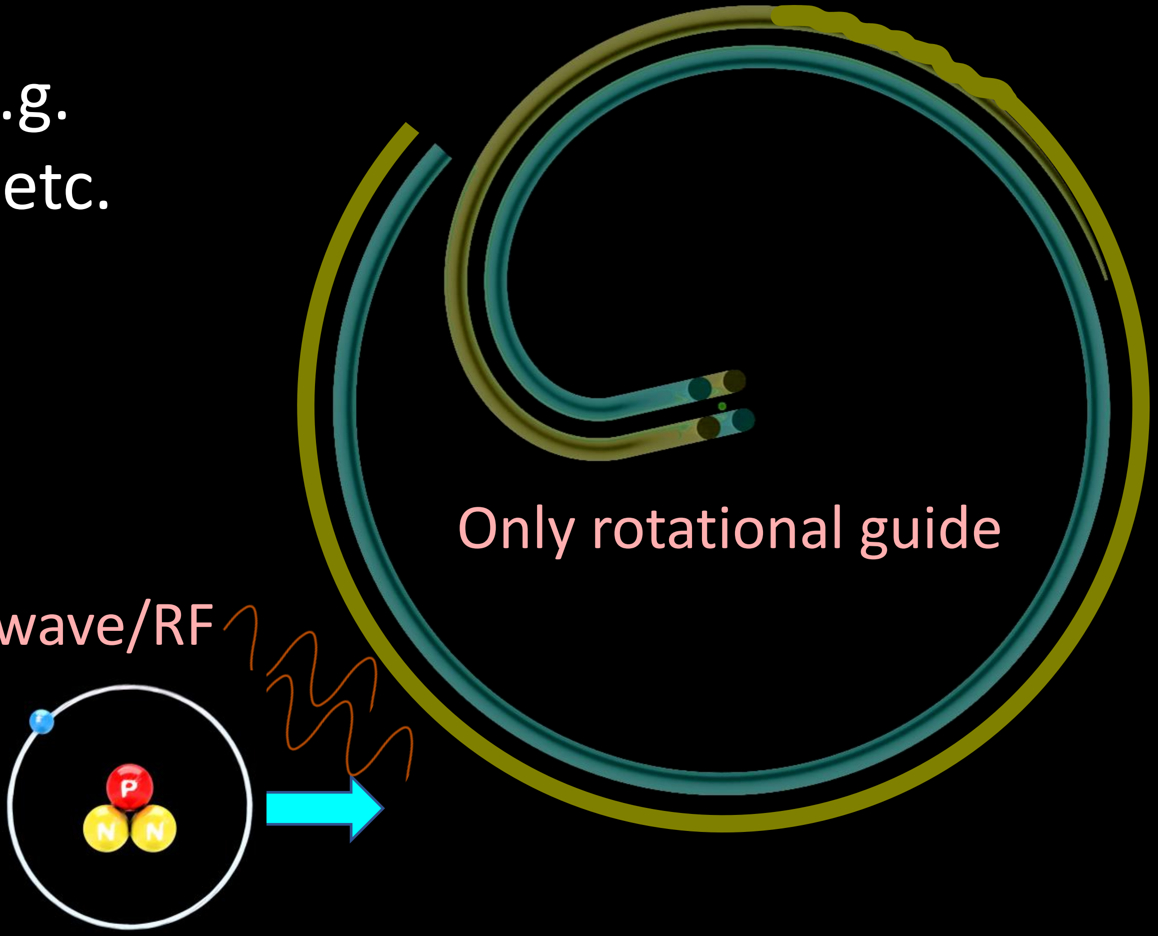
- Optical loading can be replaced by microwave/RF pumping

Hamish Robertson & Elise Novitski
@ UW Seattle

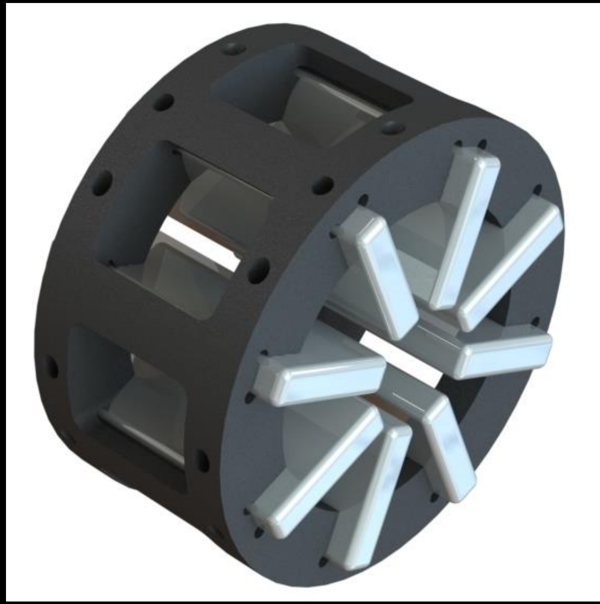
“Project 8”: probe neutrino mass from Tritium β-decay

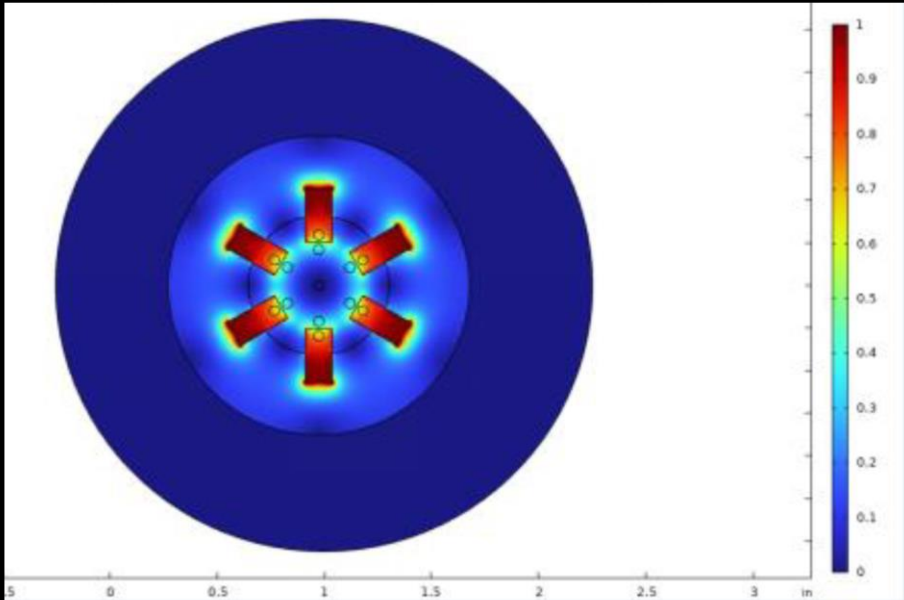


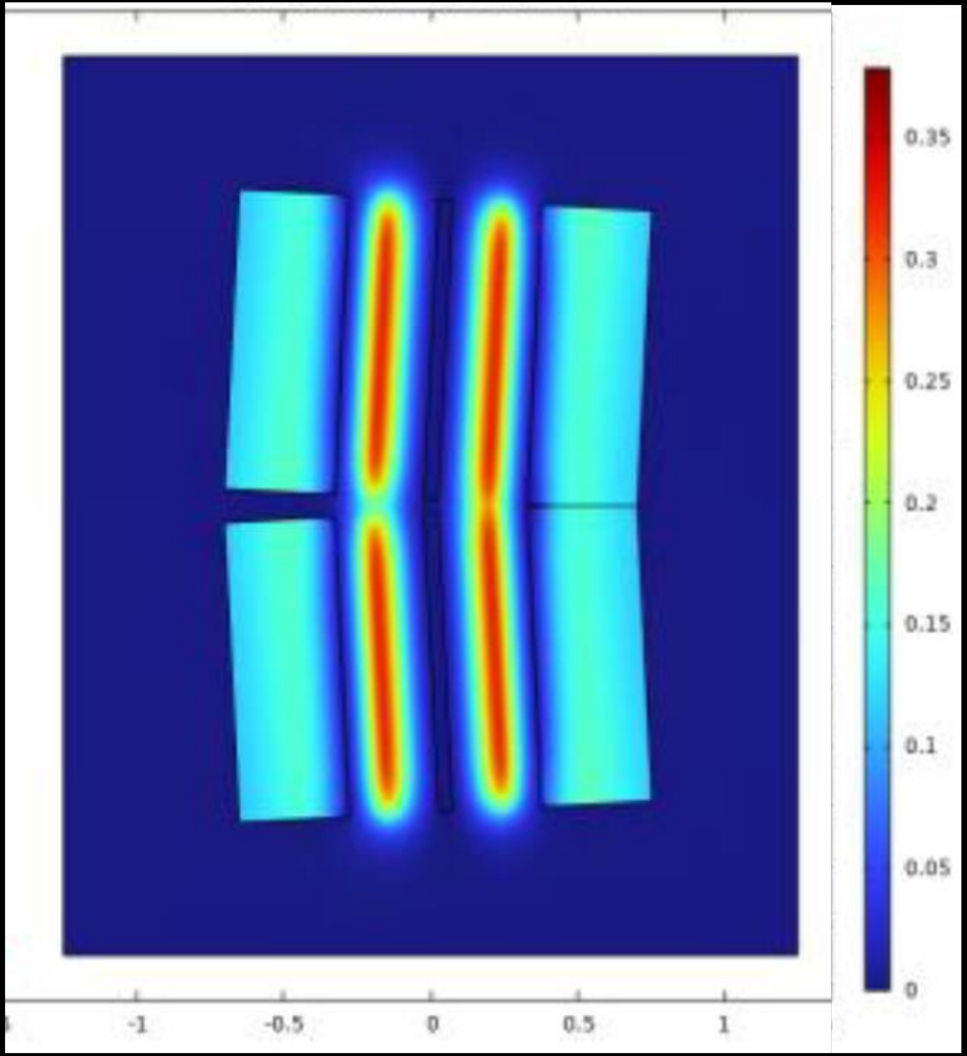
Start with Hexapole Permanent Magnetic Guide

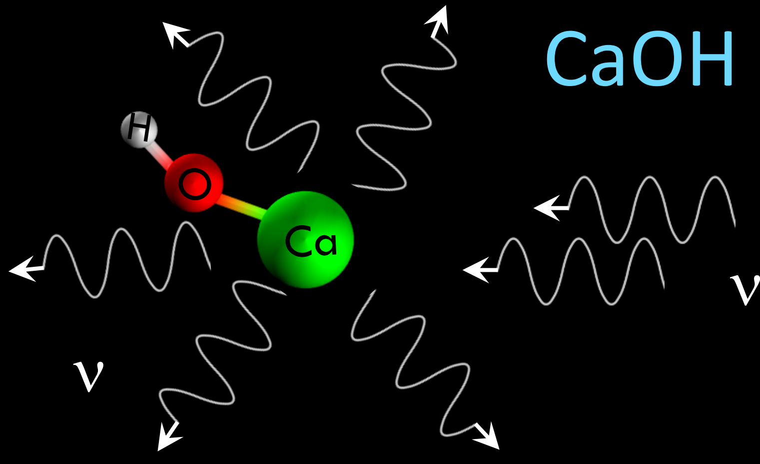
Sebastian Miki-Silva
@ FRIB/MSU

- 1st Grad student started design & simulation work.
- Plan to benchmark with Rb/Li atomic beams before $CaOCH_3$ (and eventually $RaOCH_3$)







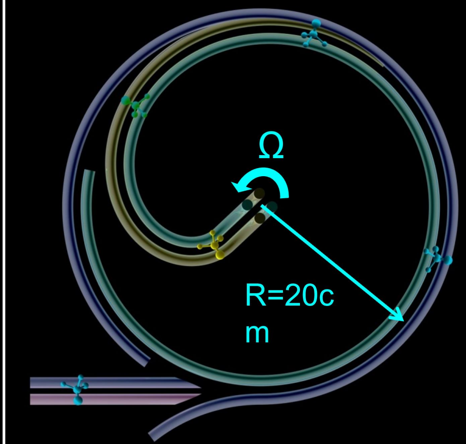


CaOH

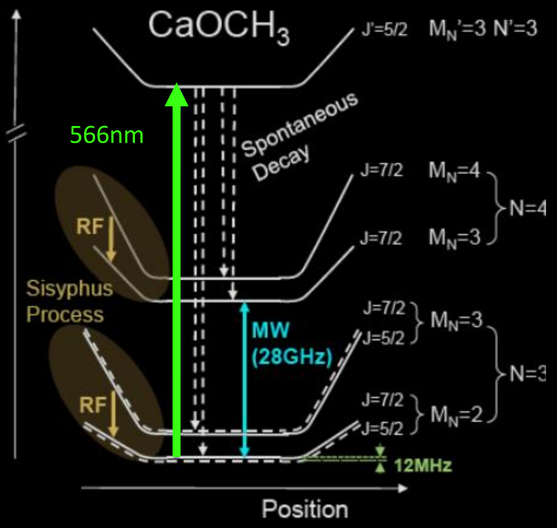
+ ~15 lasers

@ Harvard

Only 1~2 lasers



+



@ FRIB/MSU

An Ultracold-Molecule Experiment by the People, for the People!!

Long-Range Interaction

Quantum-Controlled Reaction

Chiral Molecules

Chemistry

Time Variation of Fundamental Constants

Quantum Simulation

Dipolar Quantum Physics

Biology

Quantum Memory

Astrophysics

EDM Search

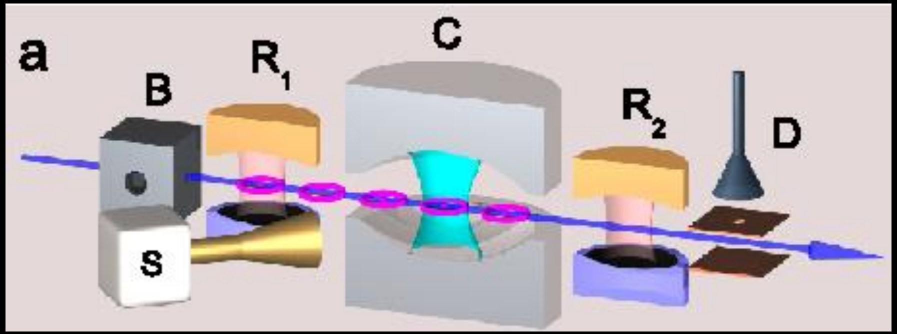
Nuclear Physics

Quantum Blackbody Thermometry

Quantum Sensing

Quantum Measurement with Molecules Coupled in Microwave Resonator

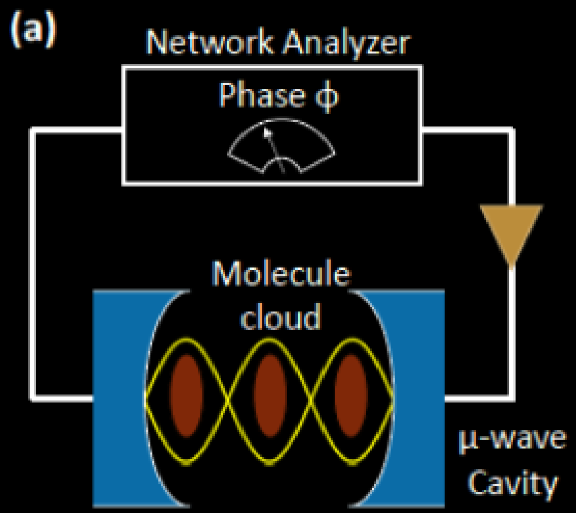
Quantum Non-Demolition (QND) Measurement with Rydberg atoms in µWave-Resonator



Serge Haroche

Physics 2012

Molecules in a µWave-Resonator



QND Measurement-based Spin Squeezing

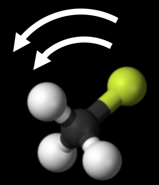
Demonstrated with atoms in optical resonators:

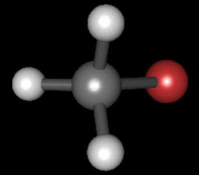
V. Vuletić @ *MIT*
J. Thompson @ *JILA, Boulder*
Schleier-Smith, Kasevich @ *Stanford*

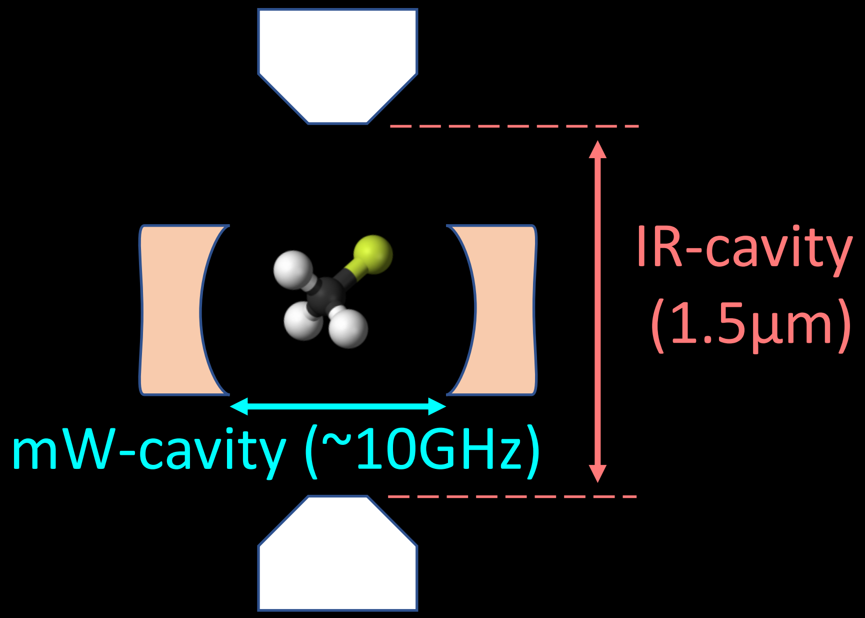
Cavity QED with Molecules

- In addition to metrology …:
  - Molecules in a microwave-IR cross cavity

Molecule has
- rotational transitions (6~60GHz)
- vibrational transition (in IR range)







Quantum transducer:
to convert qubits between mW
and telecom wavelength

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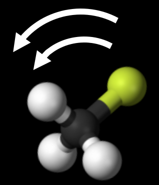

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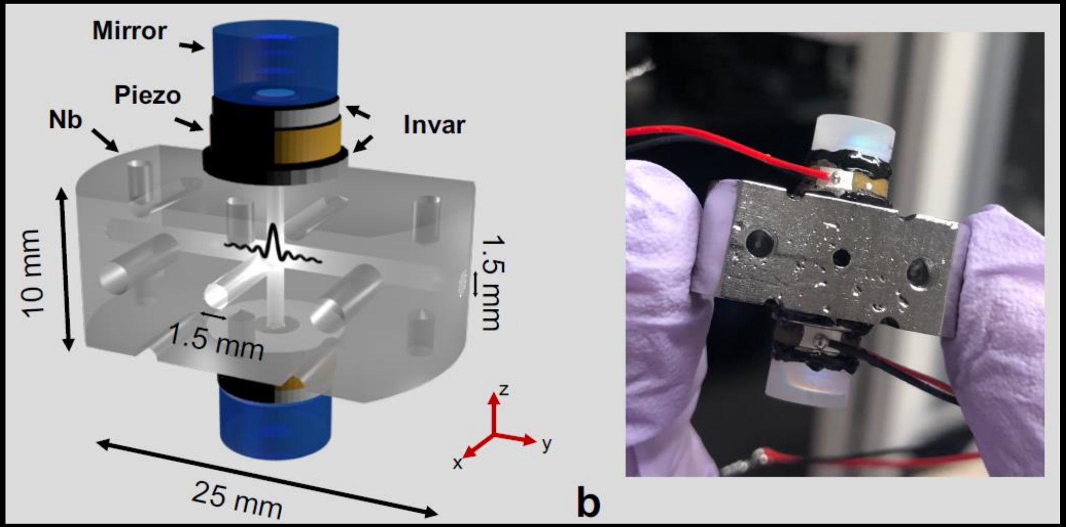
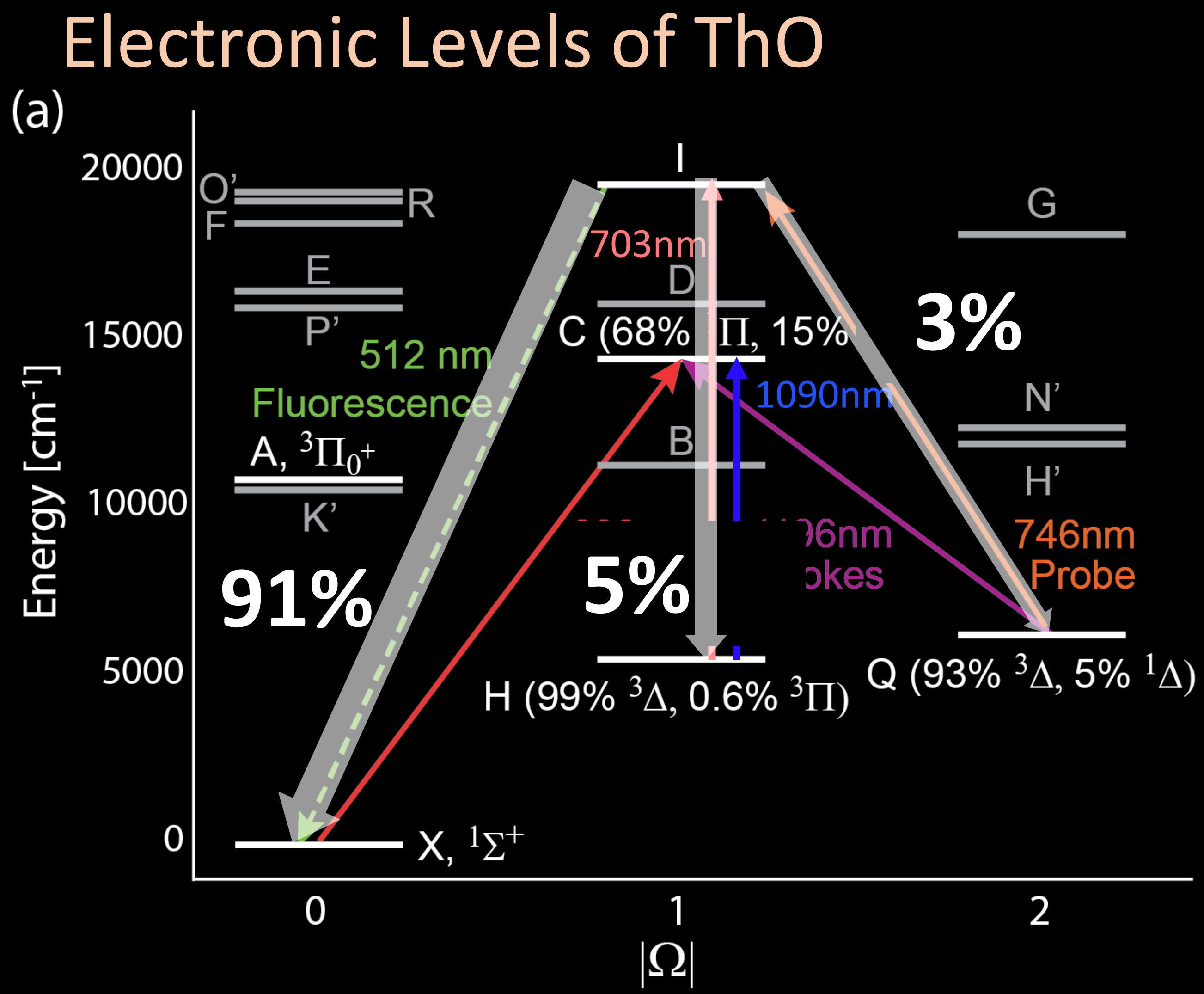


D. Schuster @ *U Chicago*

ACME II Setup Moved from Harvard to East Lansing this Year

Great perspective for cycling >10 photons on ThO X→I transition



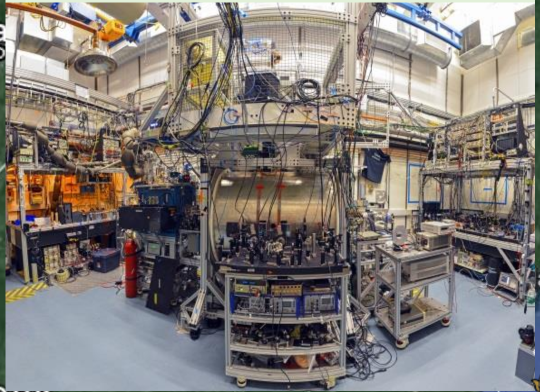
- X→I transition (512nm) has 91% branching ratio back to X, v=0 state.
- Cycling >10 photons + other improvement can significantly enhance eEDM sensitivity
- Potentially cycle ~100 photons by closing Q→I and H→I transitions:
  - ➢ Maybe more than just photon-cycling???

ACME II Setup Moved from Harvard to East Lansing this Year

Alex Frenett from PolyEDM project @ Harvard will JOIN FRIB as postdoc fellow this September

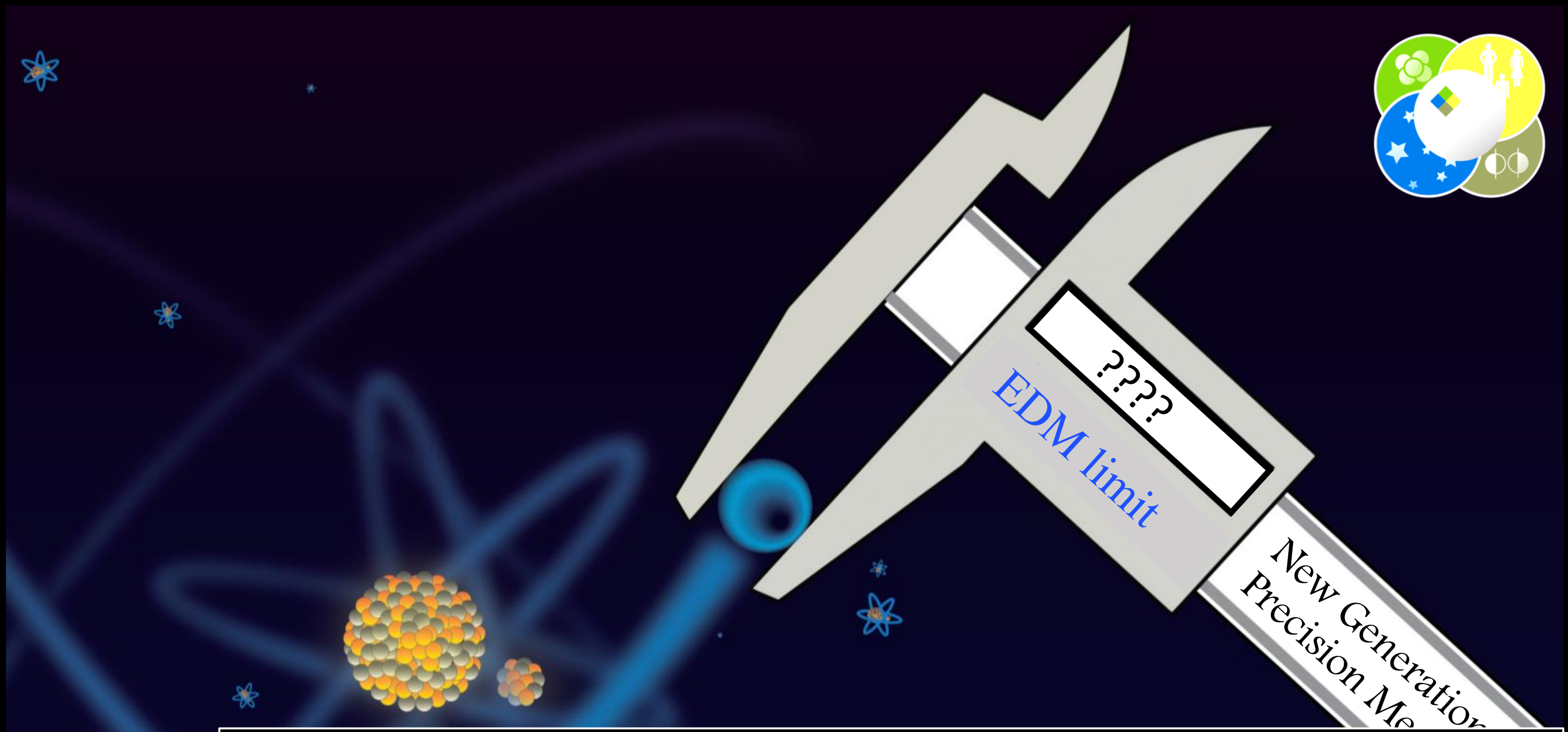
FRIB

Historic ACME II setup



Test New Upgrades for … post-ACME-III

Rare Isotopes Gives Further Enhancement



Eventually cover the entire parameter space, up to the Standard Model prediction!

Quantum Control of Molecules

Enhancement: 2 orders of magnitude

Quantum-Enhanced Sensing:

Enhancement: 1~2 orders of magnitude

Nuclear Enhancement to Hadronic Sector of *T*-Violation

Octupole Deformation in e.g. ^{225}Ra, ^{223}Fr

Enhancement: up to 4 Orders of Magnitude

Test Fundamental Symmetries using Radioactive Molecules @ FRIB

Contact: wux@frib.msu.edu

Xing Wu

An excellent team needs excellent Students & Postdocs

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