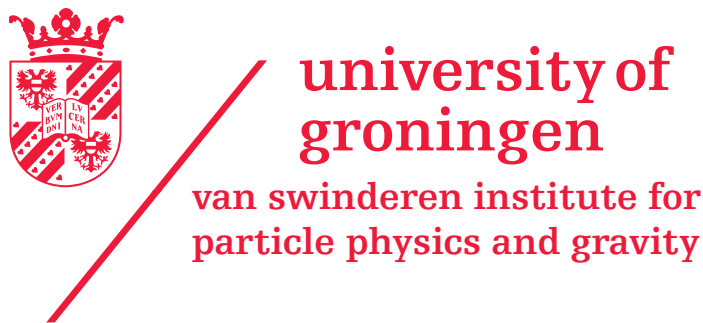
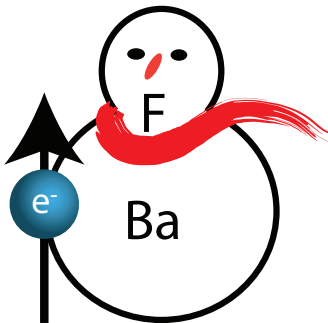
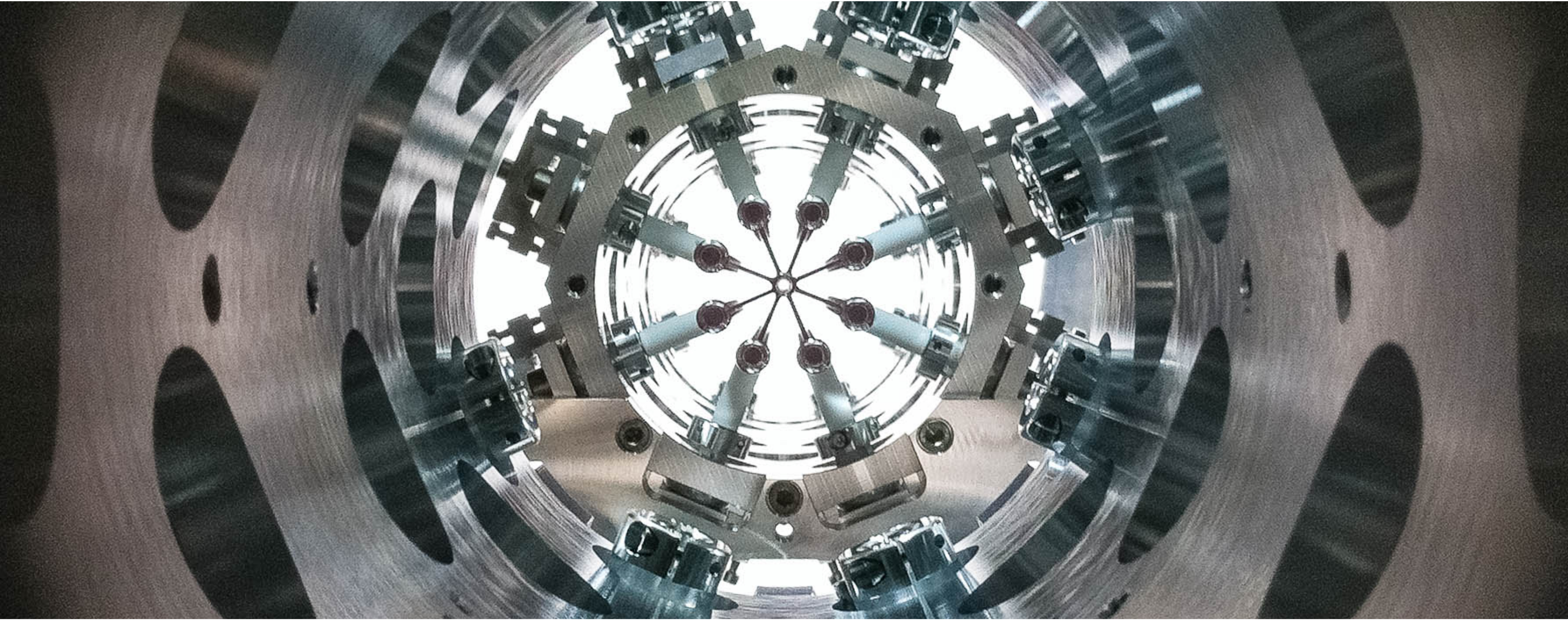


Molecular systems for tests of fundamental physics

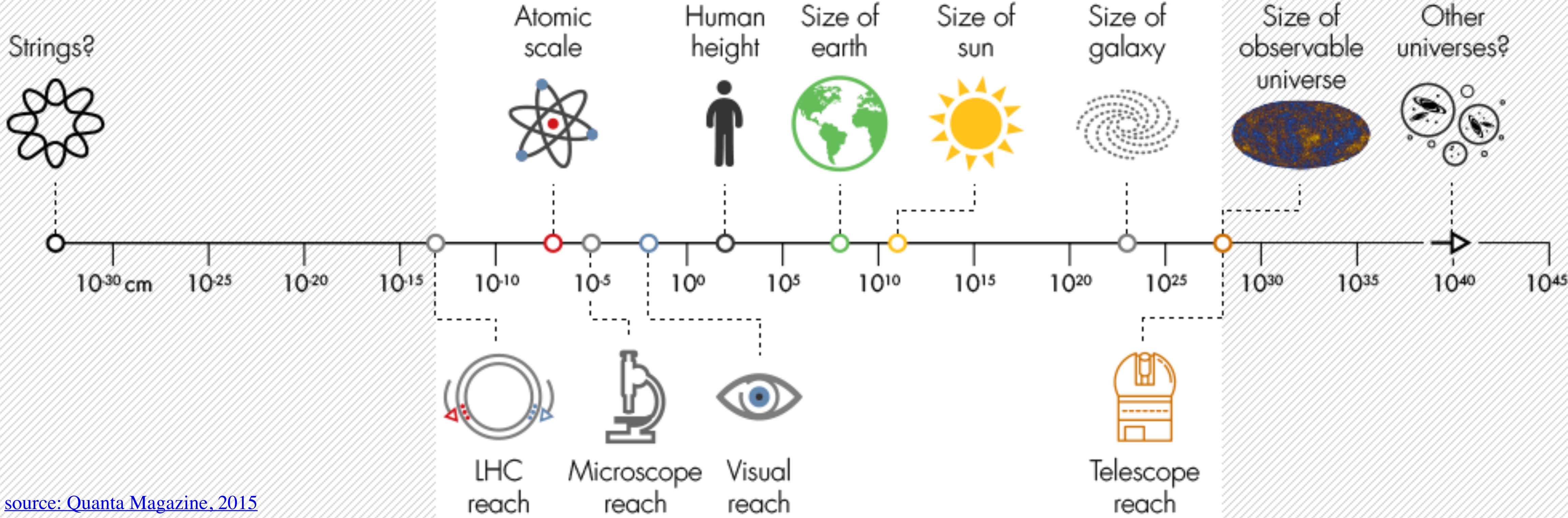
Steven Hoekstra, RUG & Nikhef



Motivation: reach beyond the current limits of observation

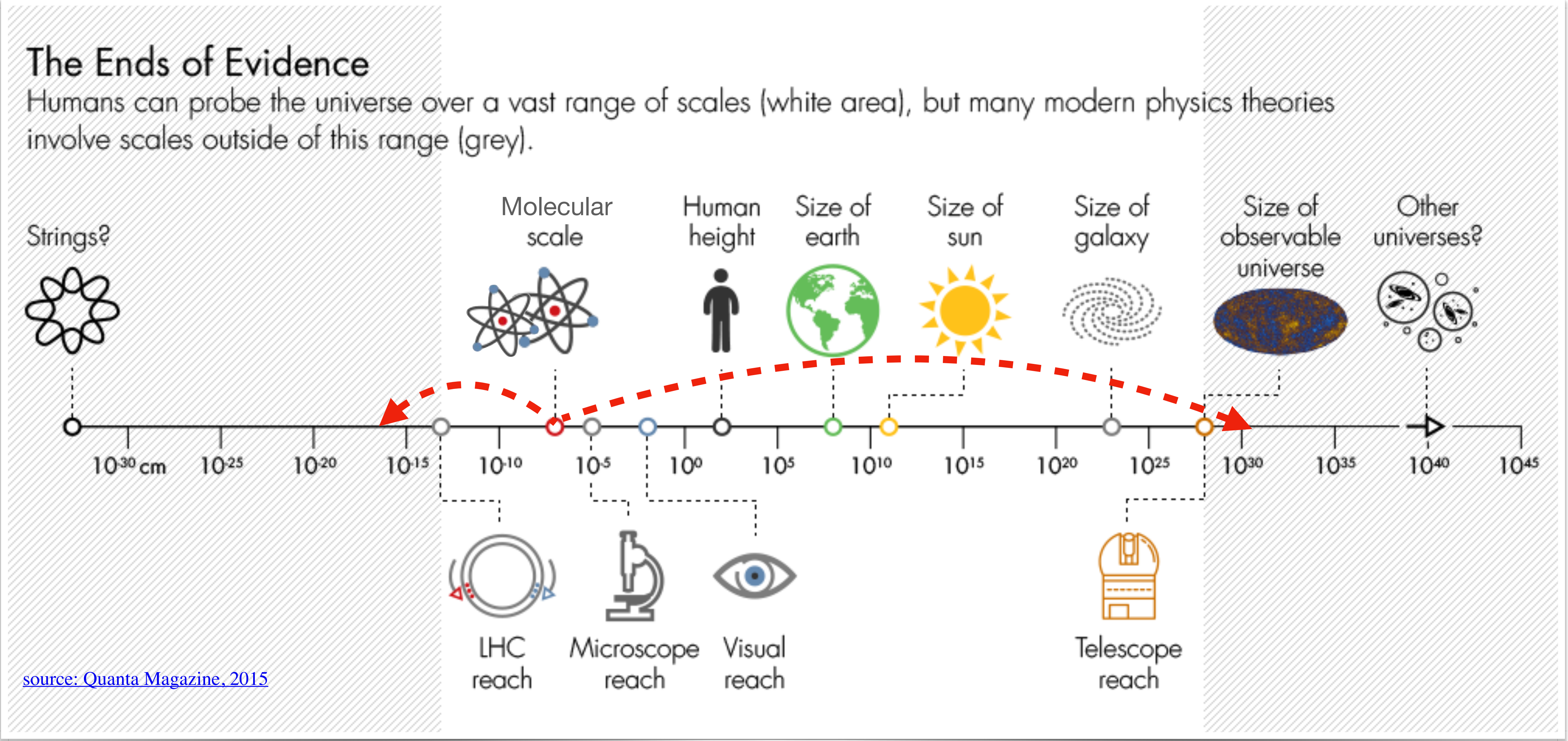
The Ends of Evidence

Humans can probe the universe over a vast range of scales (white area), but many modern physics theories involve scales outside of this range (grey).



source: [Quanta Magazine, 2015](#)

Motivation: reach beyond the current limits of observation



Molecules can be extremely sensitive quantum sensors for fundamental physics!

Molecules vs atoms

Extra complexity brings experimental challenges and new possibilities

Close-lying opposite parity levels: study parity violation
(also chirality)

Heavy polar molecules: hugely enhanced electron-EDM sensitivity

Tunneling in molecular motion: extremely sensitive to value of constants

Molecules vs atoms

Extra complexity brings experimental challenges and new possibilities

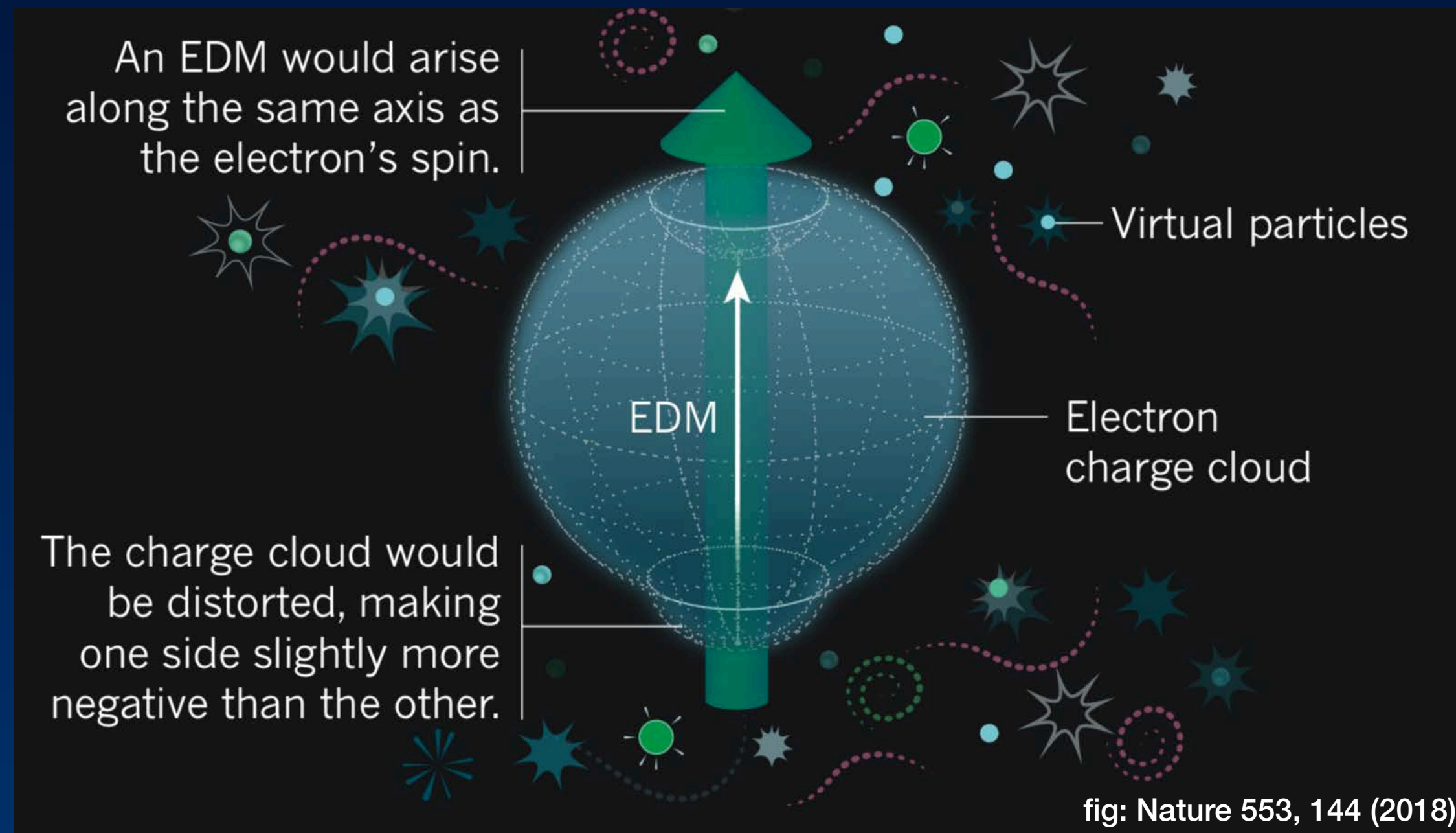
Close-lying opposite parity levels: study parity violation
(also chirality)

Heavy polar molecules: hugely enhanced electron-EDM sensitivity

Tunneling in molecular motion: extremely sensitive to value of constants

The electron's Electric Dipole Moment (eEDM)

probing CP violation beyond the standard model



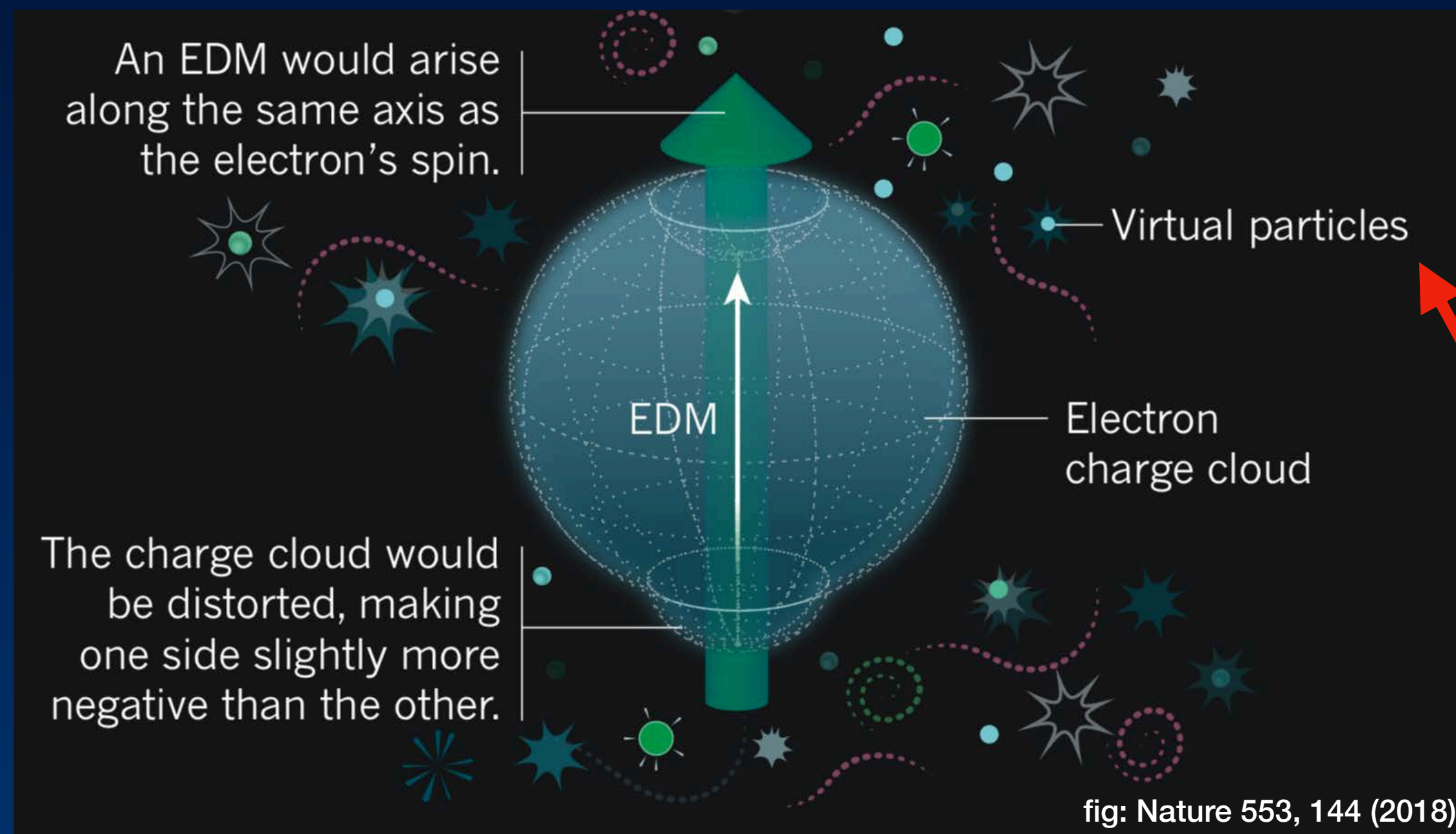
eEDM violates P, T and CP symmetry (provided CPT holds)

eEDM magnitude
(e cm)



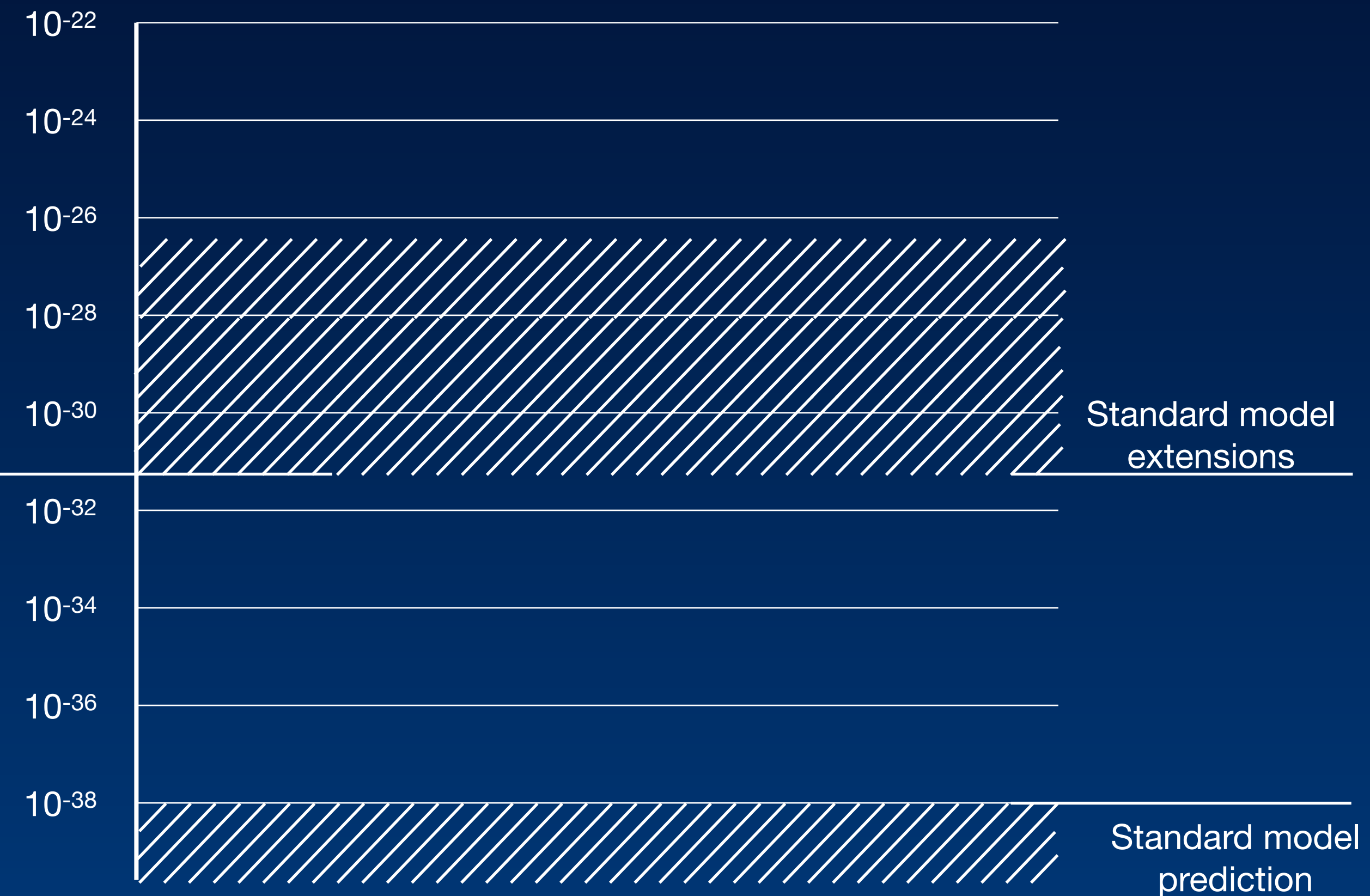
The electron's Electric Dipole Moment (eEDM)

probing CP violation beyond the standard model



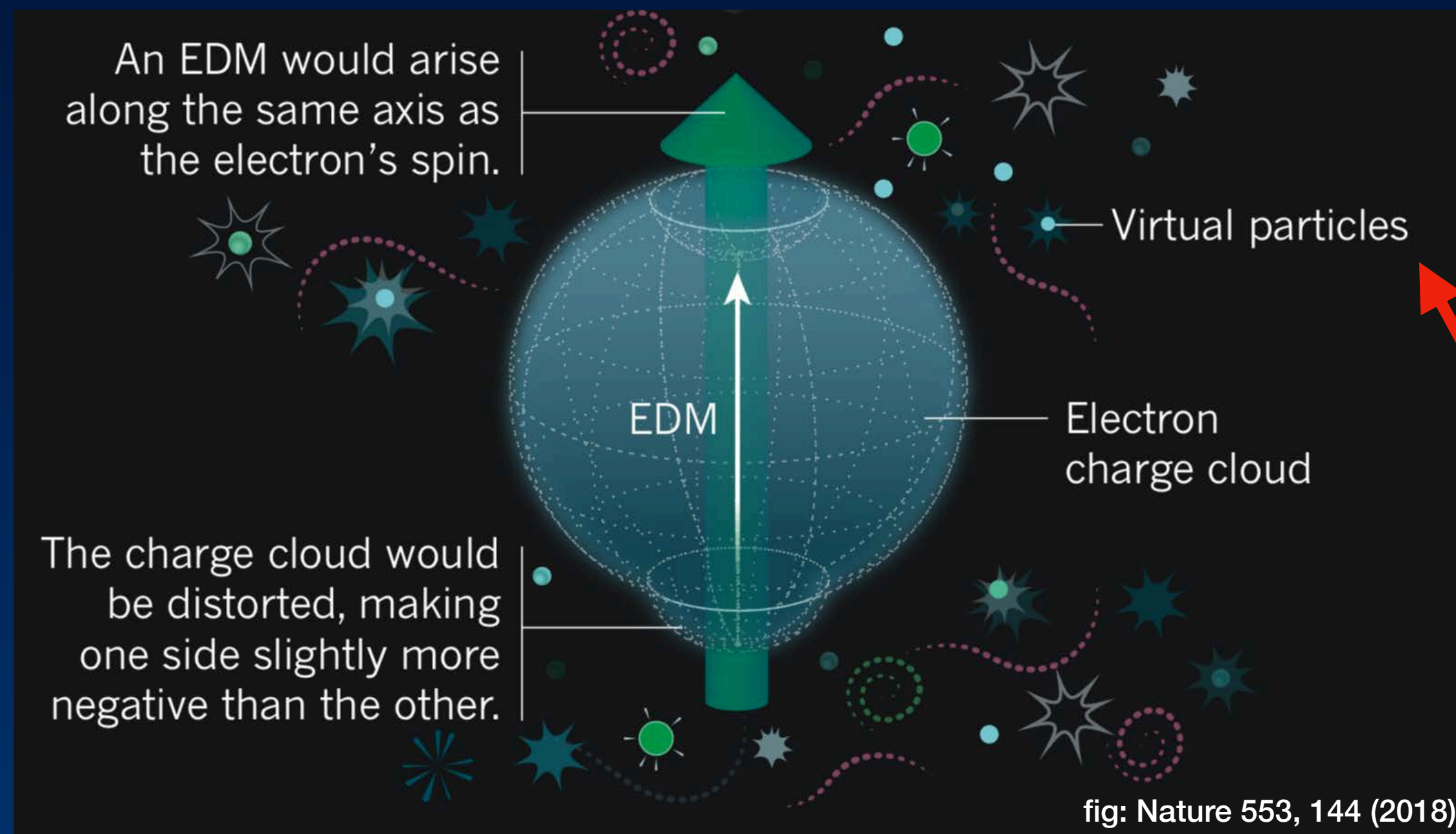
eEDM violates P, T and CP symmetry (provided CPT holds)

eEDM magnitude
(e cm)



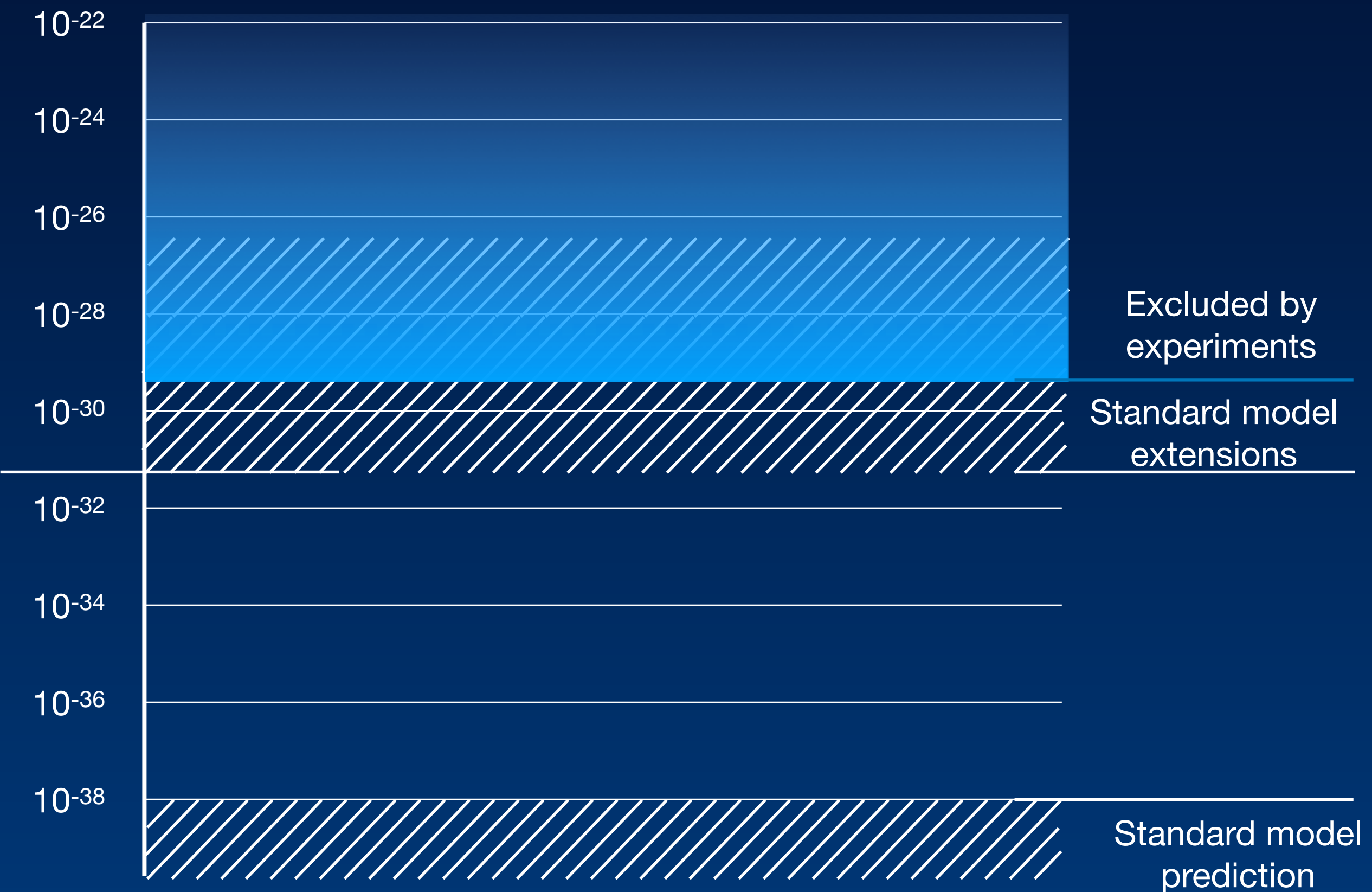
The electron's Electric Dipole Moment (eEDM)

probing CP violation beyond the standard model

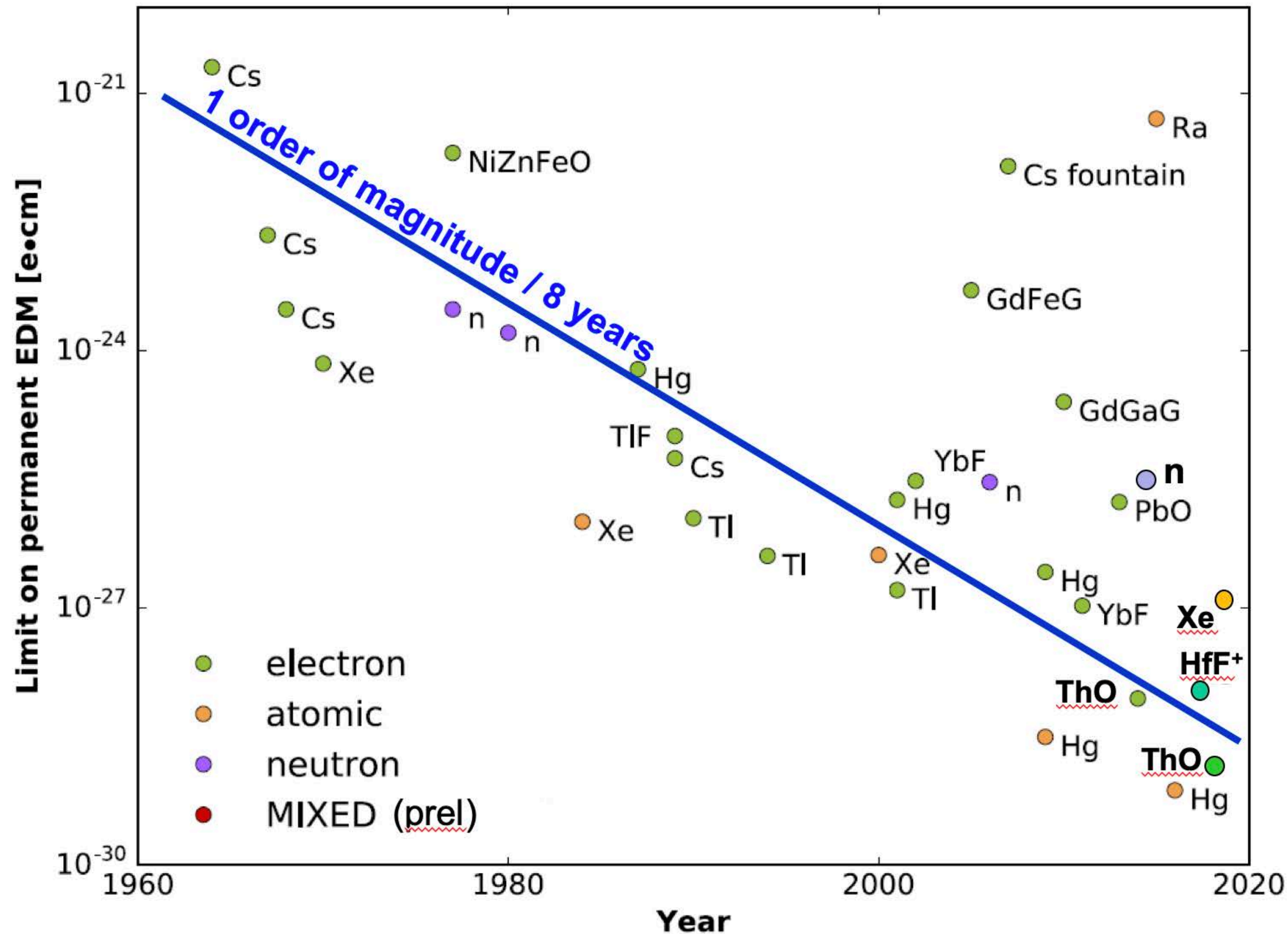


eEDM violates P, T and CP symmetry (provided CPT holds)

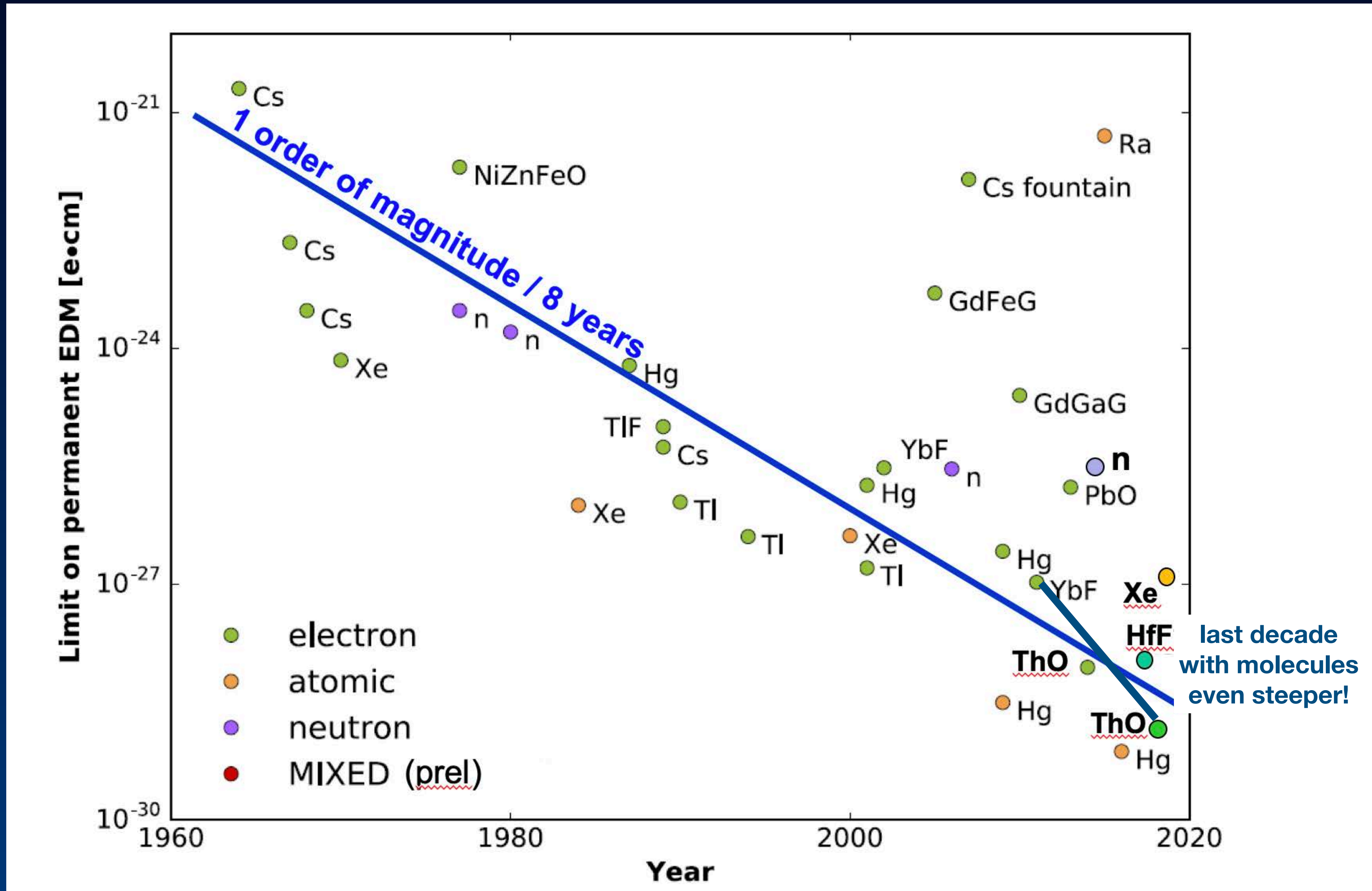
eEDM magnitude
(e cm)



A history of measurements with steady progress



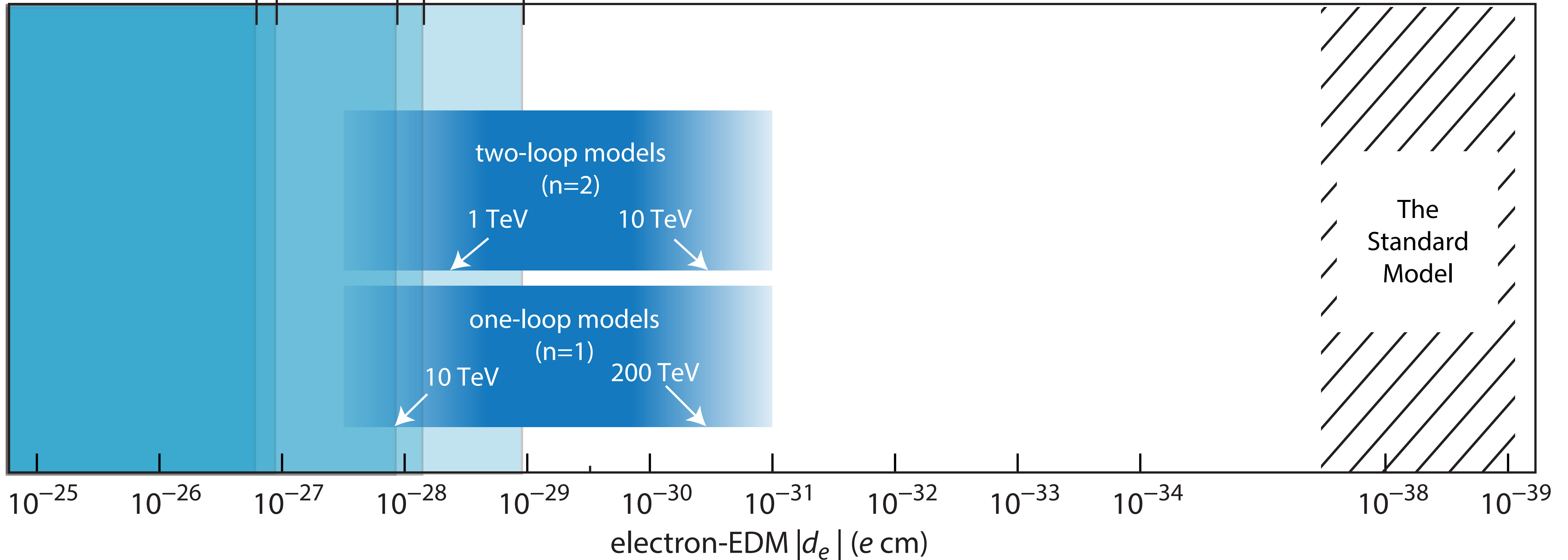
A history of measurements with steady progress



The electron's electric dipole moment (eEDM)

effectively a background-free method to probe new physics

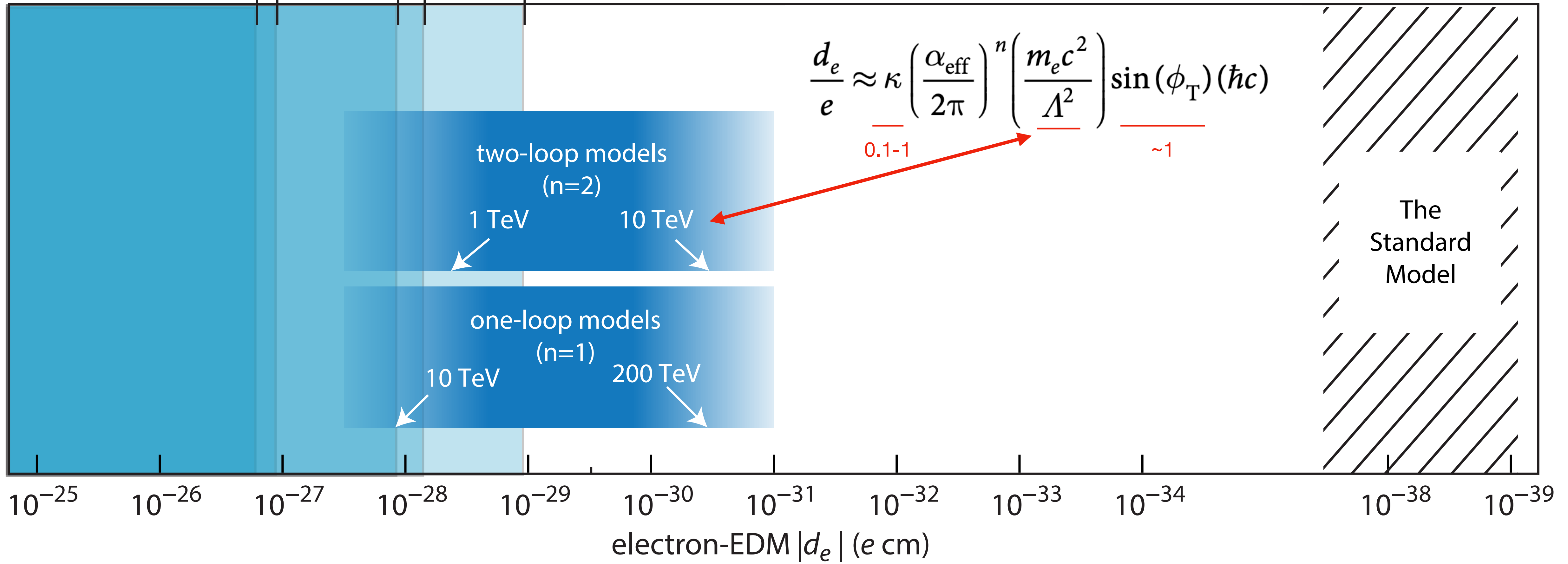
Tl YbF (2002) (2011) HfF⁺ ThO (2017) (2014) ThO (2018)



The electron's electric dipole moment (eEDM)

effectively a background-free method to probe new physics

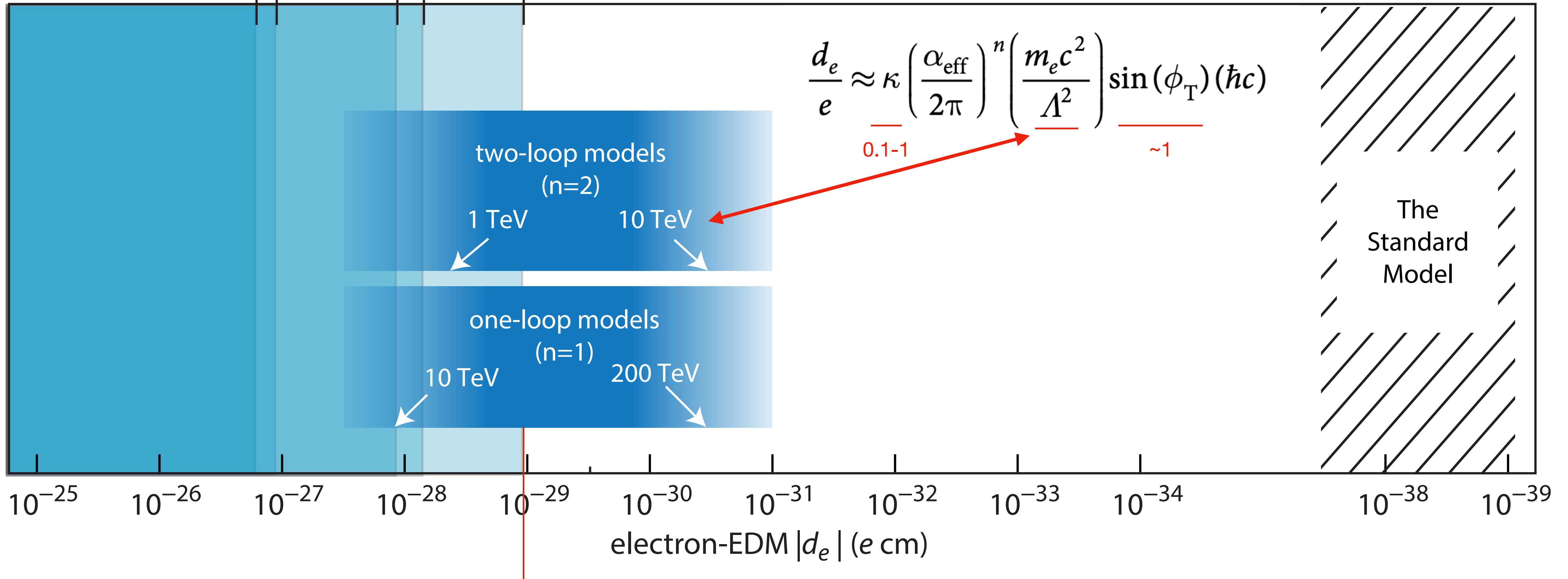
TI YbF (2002) HfF⁺ ThO (2017) ThO (2014) ThO (2018)



The electron's electric dipole moment (eEDM)

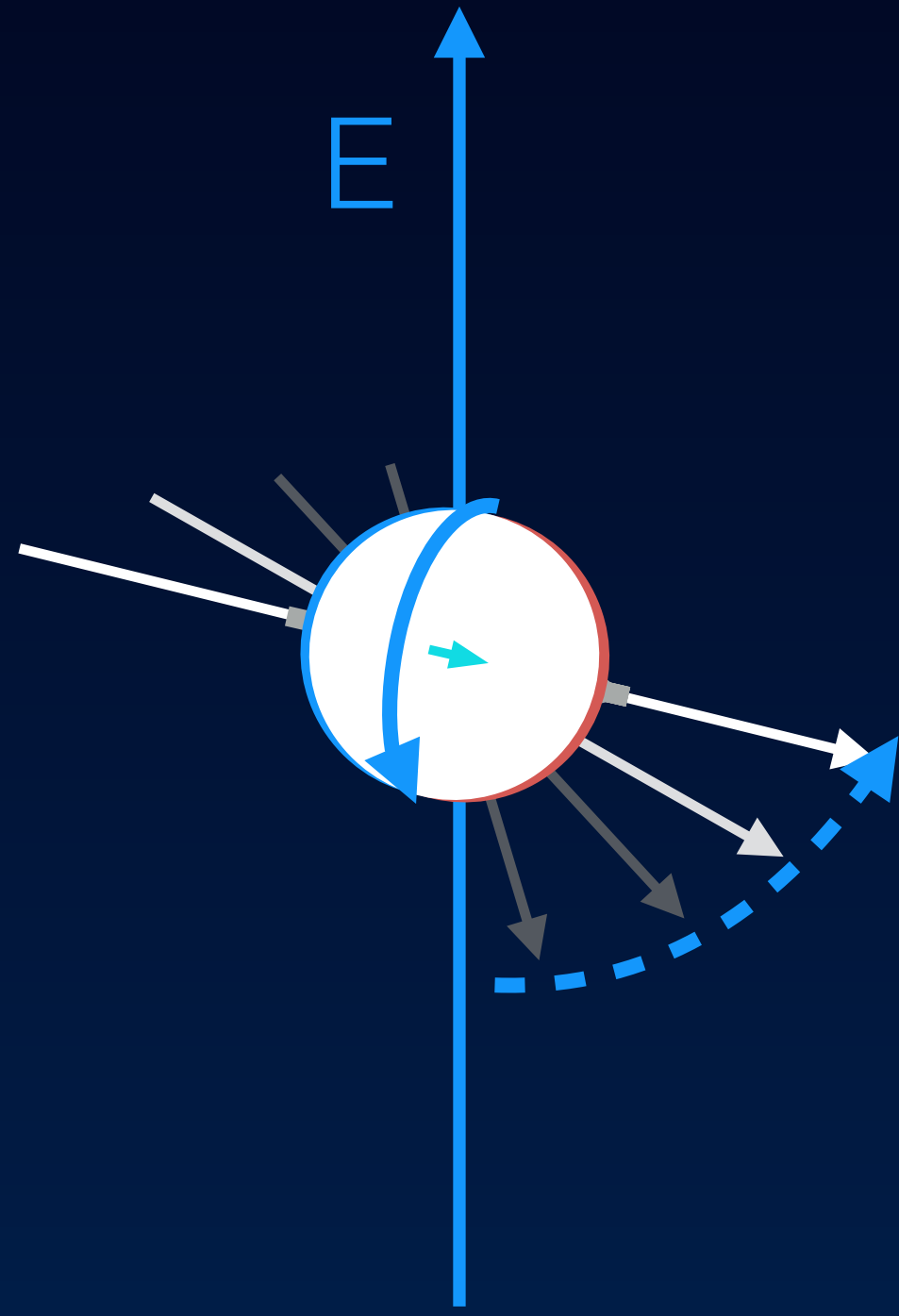
effectively a background-free method to probe new physics

TI YbF (2002) HfF⁺ ThO (2017) ThO (2014) ThO (2018)

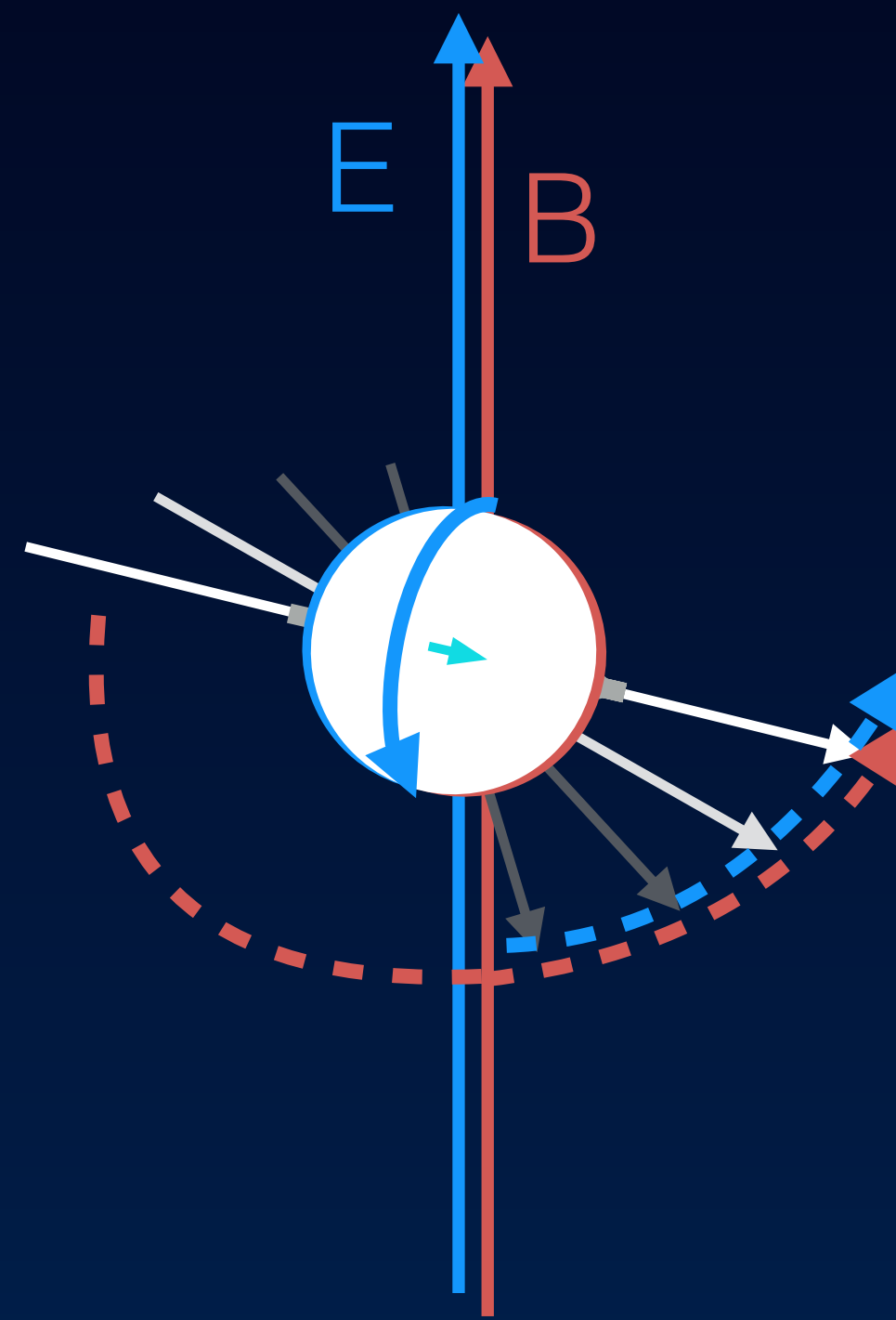


The ThO result limits time-reversal-symmetry-violating new physics to energy scales above $\Lambda \approx 30 \text{ TeV}$ or $\Lambda \approx 3 \text{ TeV}$, for $n=2$ or 1 , respectively

How to measure a dipole moment?

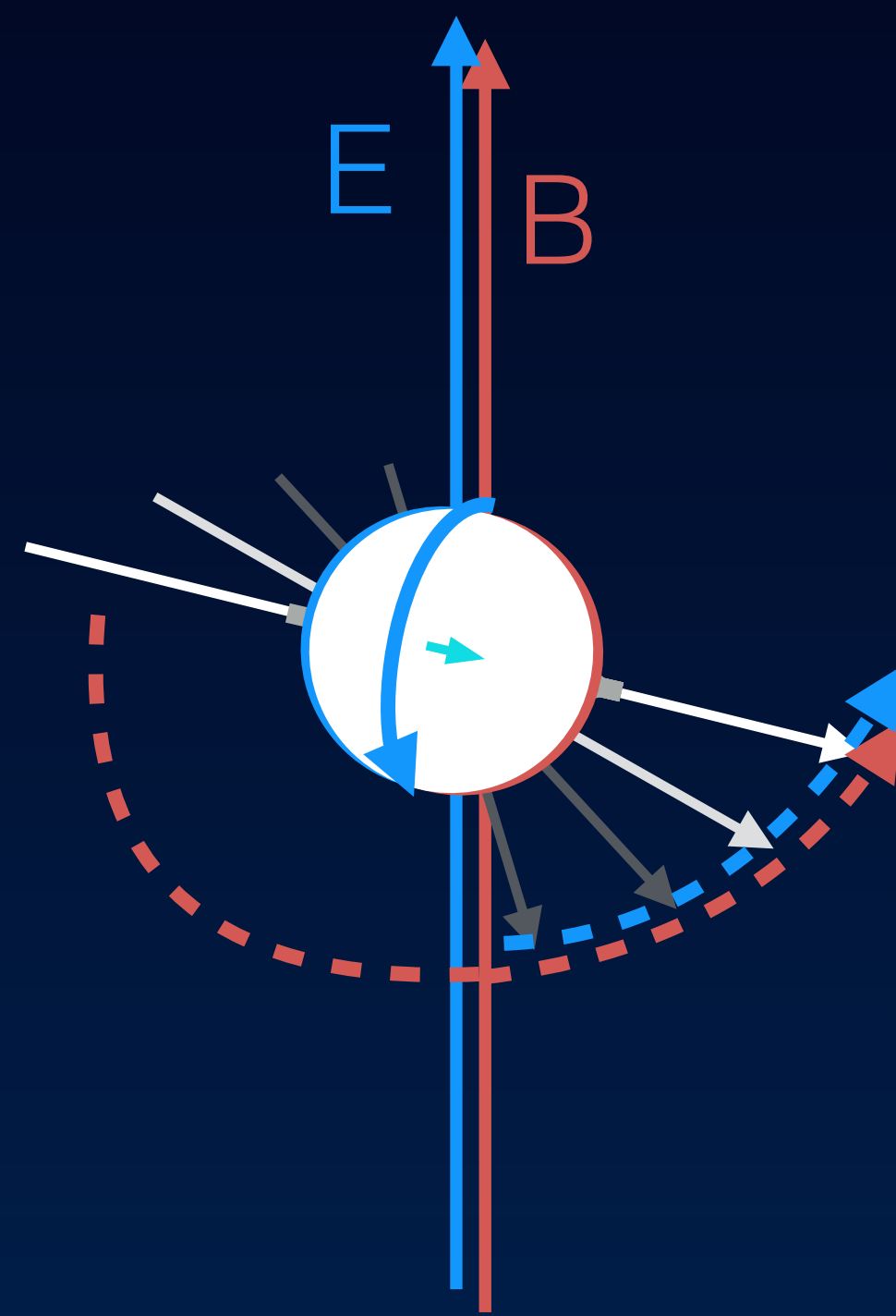
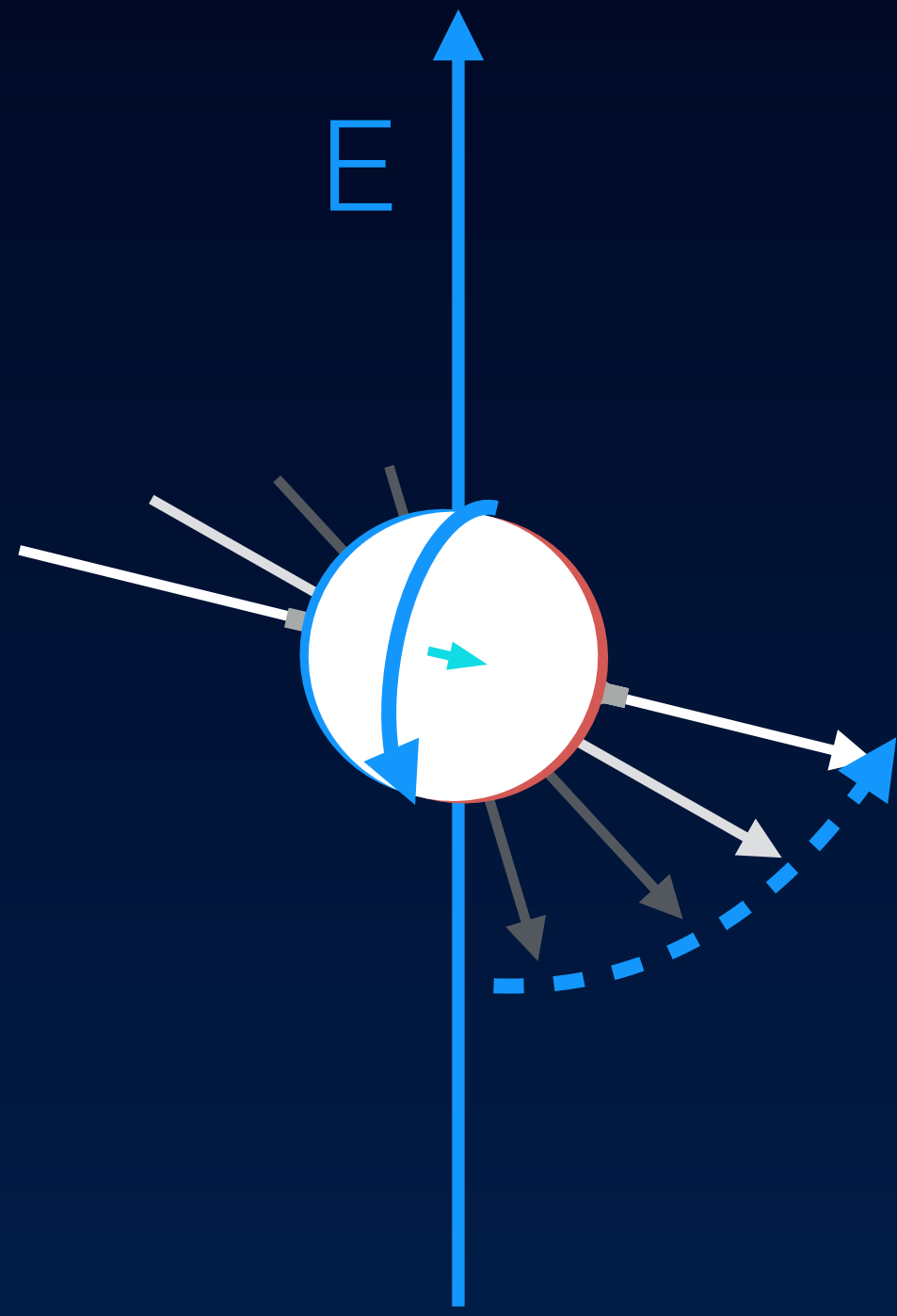


precession!

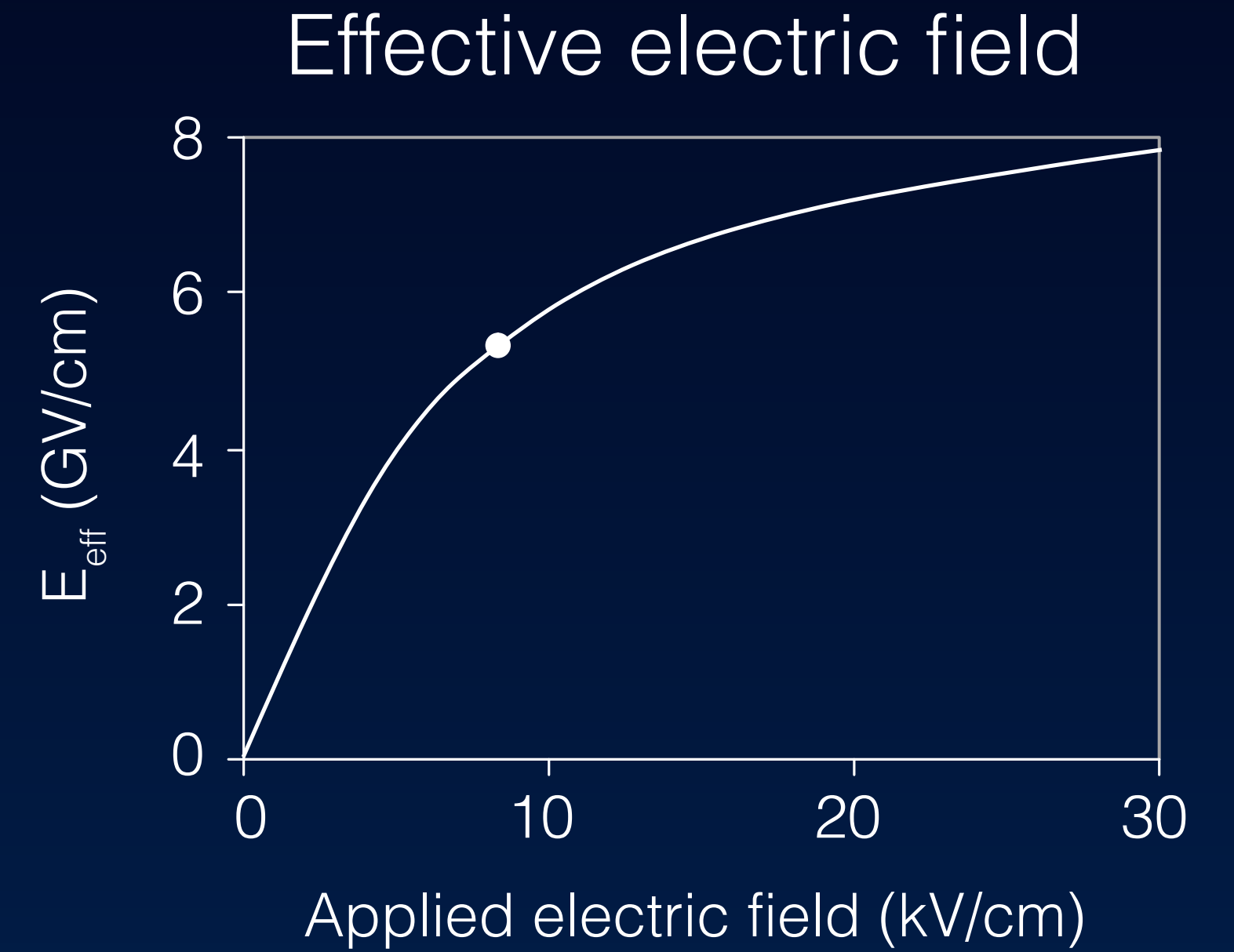


However, electron also has
magnetic dipole moment
(and charge!)

How to measure a dipole moment?



polar molecule



precession!

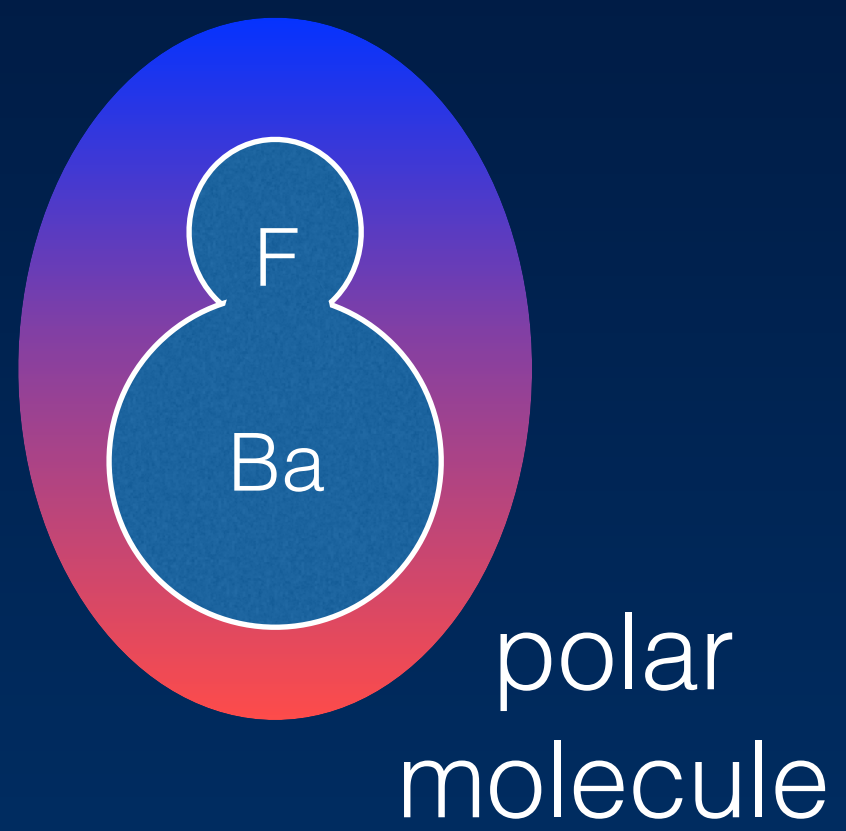
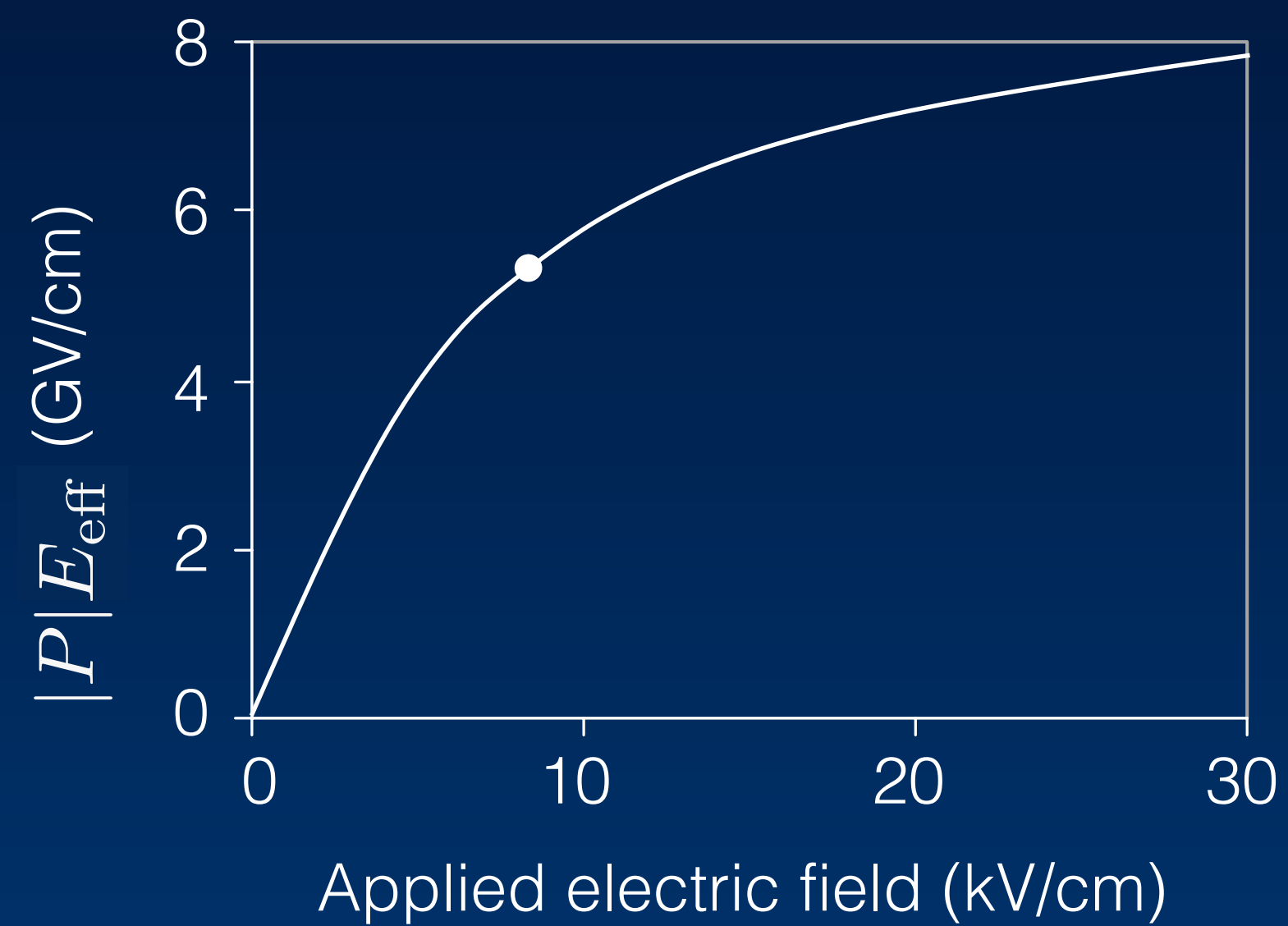
However, electron also has magnetic dipole moment (and charge!)

Solution:
use electron embedded in a polar molecule!

Enhances E
Shields B

Increasing the eEDM sensitivity

Effective electric field

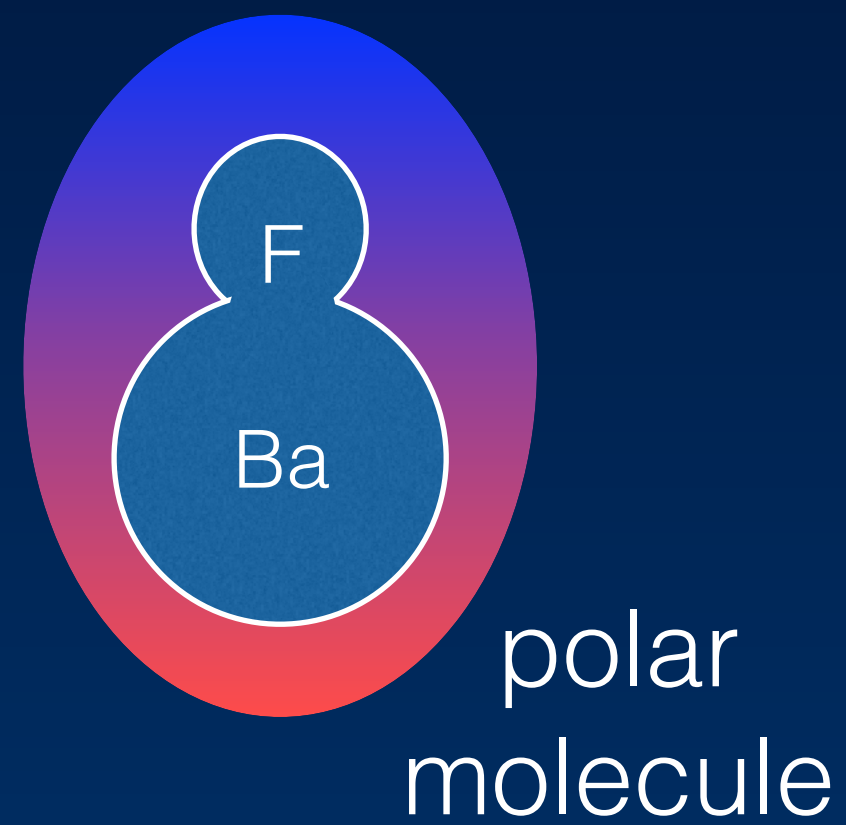
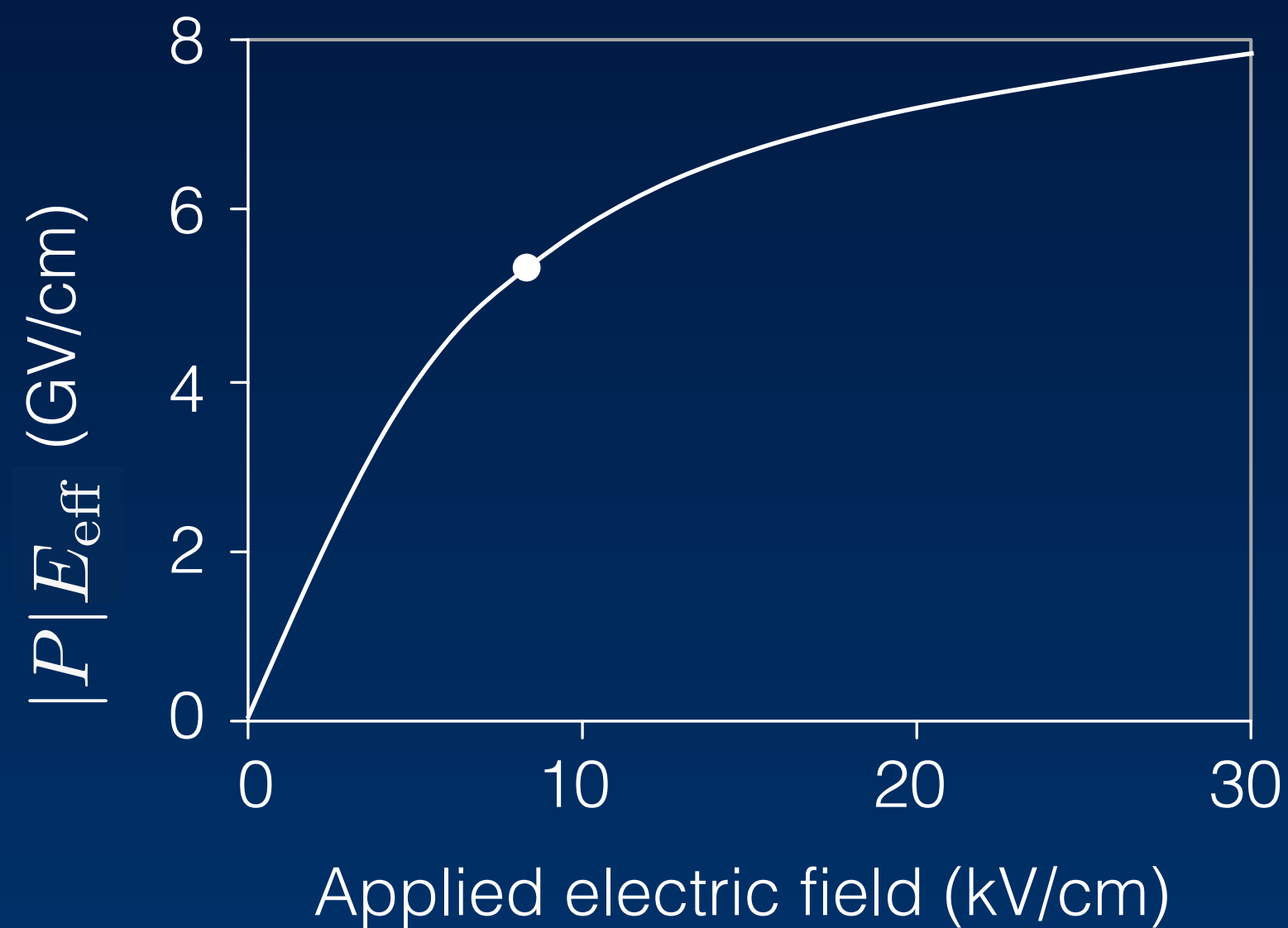


Increasing the eEDM sensitivity

Measure shift of molecular energy level
that correlates with electric field direction reversal

statistical error:
$$\sigma_d = \frac{\hbar}{e} \frac{1}{2|P|E_{\text{eff}}\tau\sqrt{\dot{N}T}}$$

Effective electric field



Increasing the eEDM sensitivity

Measure shift of molecular energy level
that correlates with electric field direction reversal

statistical error:
$$\sigma_d = \frac{\hbar}{e} \frac{1}{2|P|E_{\text{eff}}\tau\sqrt{\dot{N}T}}$$

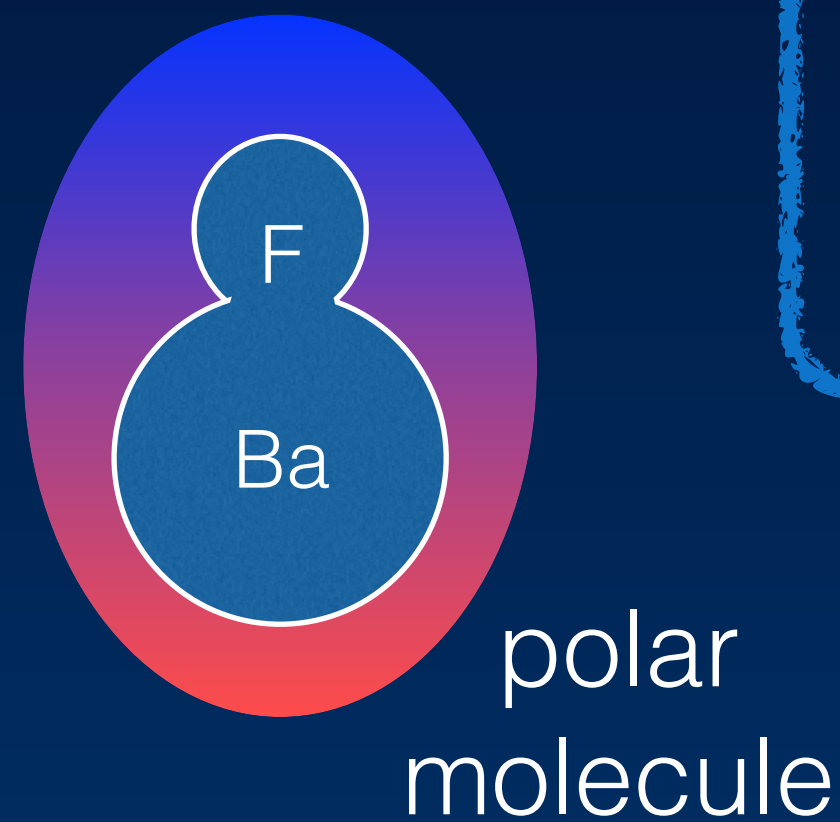
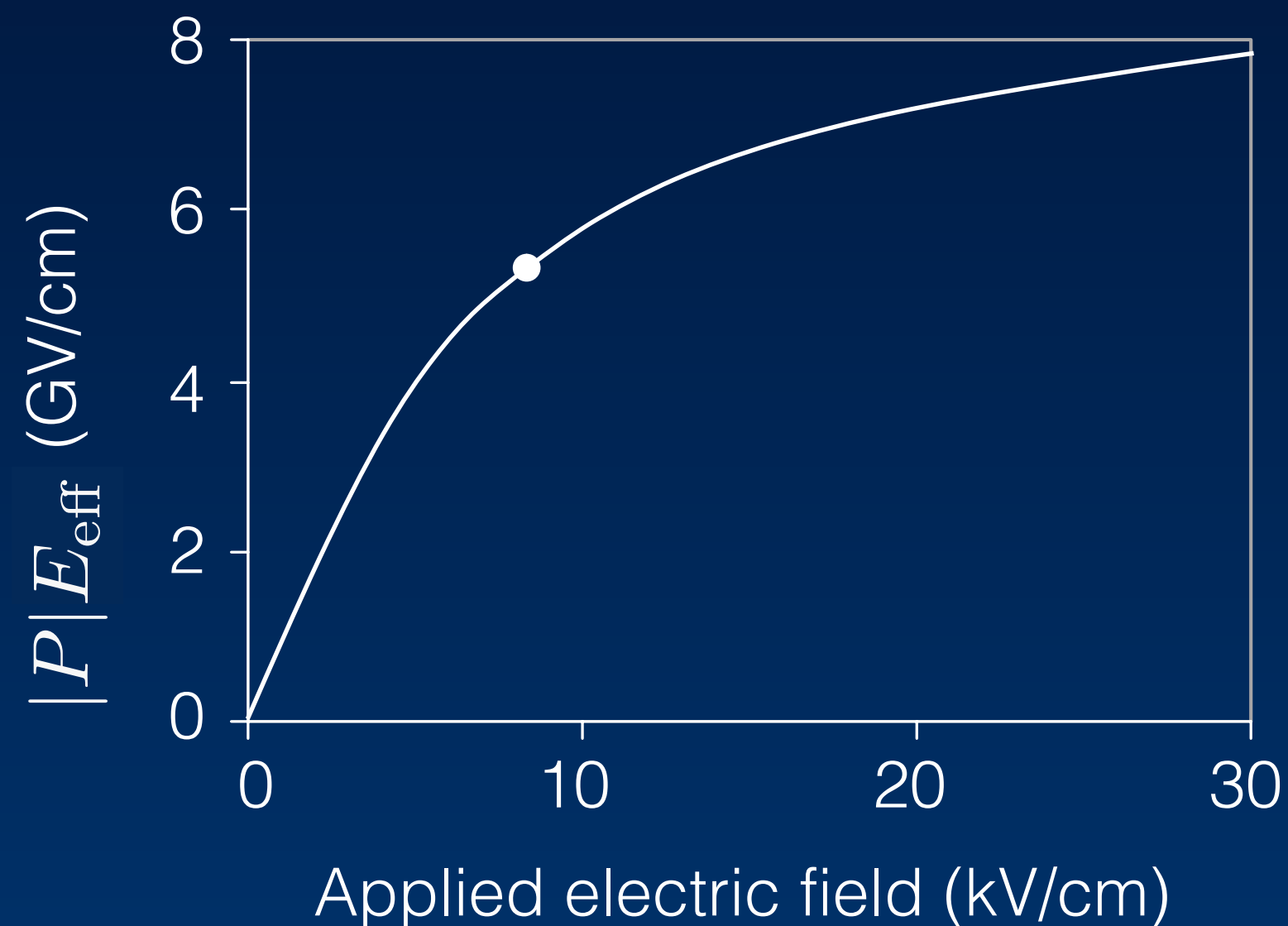
Cold Molecules

Quantum tricks

Number of detected molecules

Coherent interaction time

Effective electric field



Coherent interaction time

Key technique: Ramsey spin interferometer

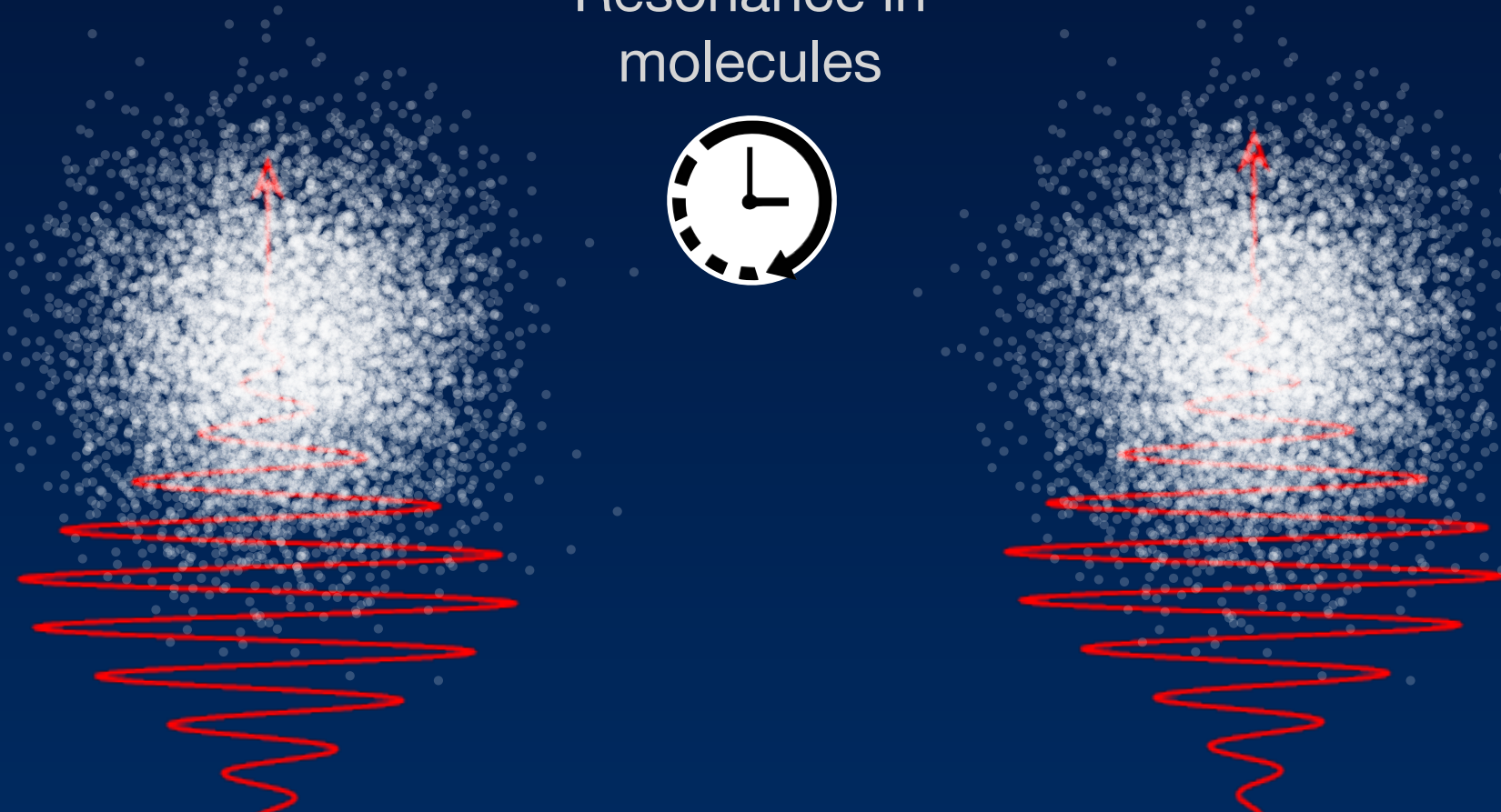
laser pulse 1:

Creates a quantum superposition, creating coherent excitation of all molecules

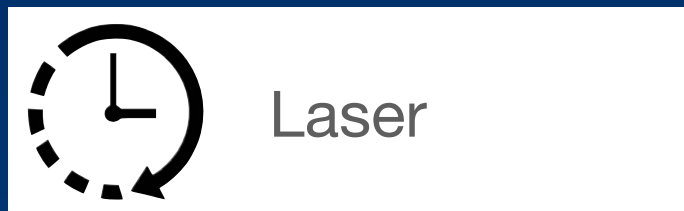
laser pulse 2:

Measures state of the molecules through interference

Resonance in molecules



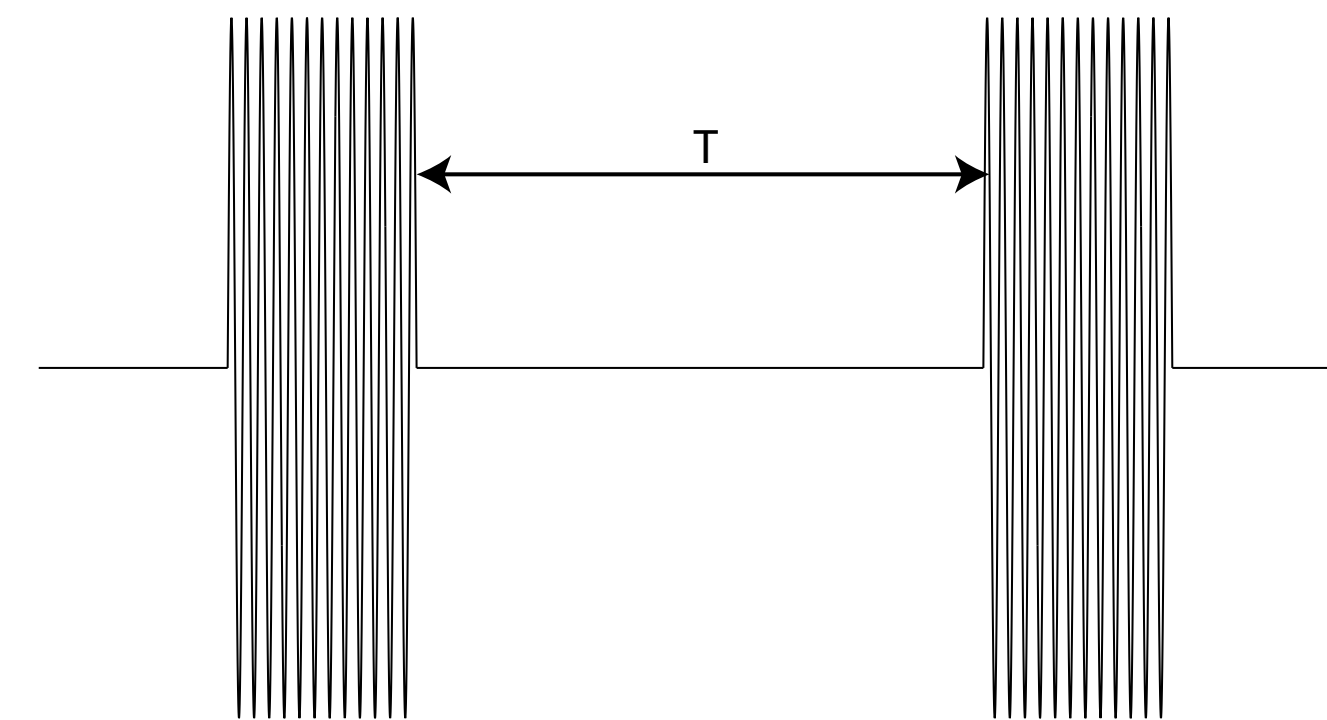
Time T



Laser

Frequency set by external reference, tuned to molecular resonance

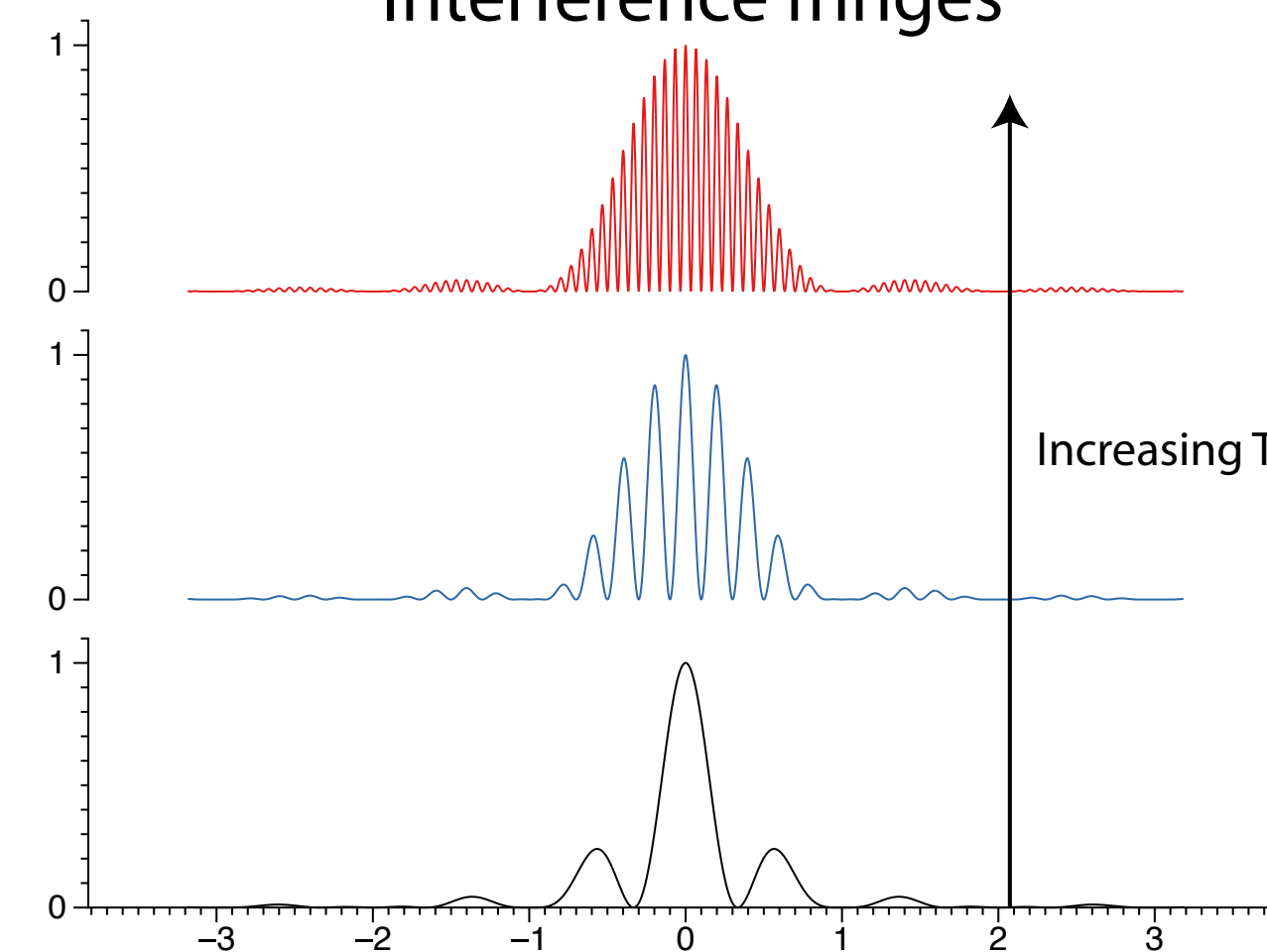
Ramsey $\pi/2$ pulses



$\pi/2$ pulse

$\pi/2$ pulse

Interference fringes



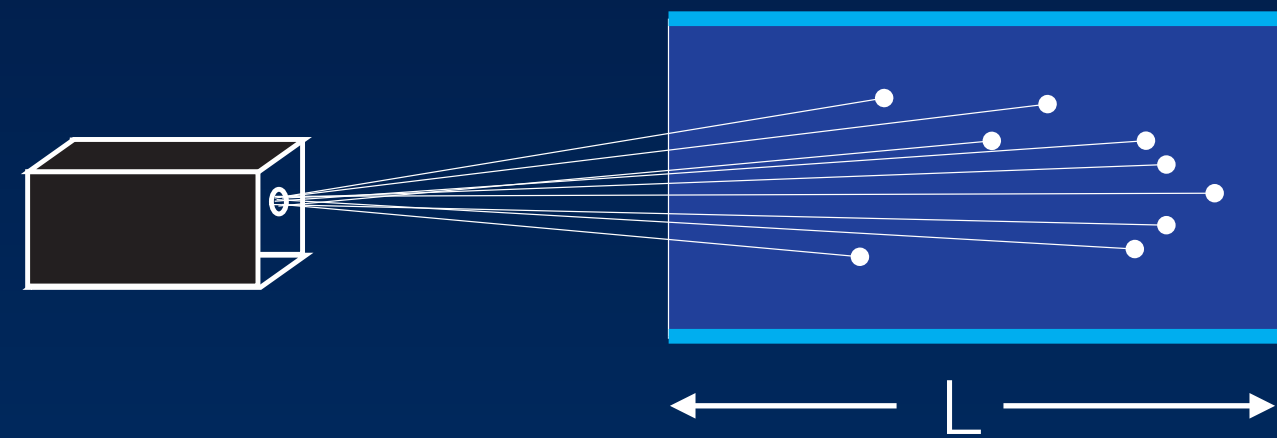
Cold molecules offer longer coherent interaction times

fast beam

$$\tau \sim 1-2 \text{ ms}$$

$$L \sim 0.5 \text{ m}$$

$$v \sim 250-500 \text{ m/s}$$



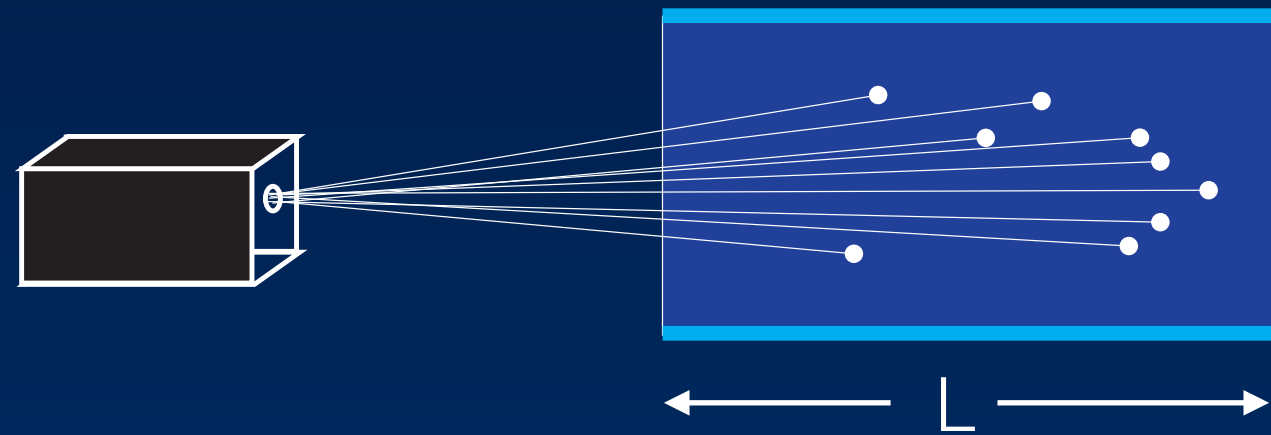
Cold molecules offer longer coherent interaction times

fast beam

$$\tau \sim 1-2 \text{ ms}$$

$$L \sim 0.5 \text{ m}$$

$$v \sim 250-500 \text{ m/s}$$

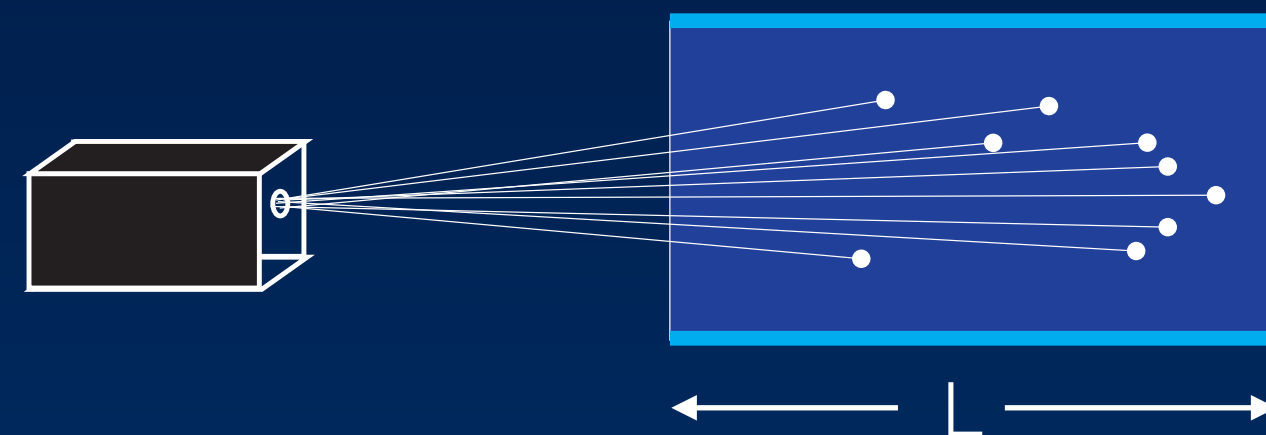


slow beam

$$\tau \sim 15 \text{ ms}$$

$$L \sim 0.5 \text{ m}$$

$$v \sim 30 \text{ m/s}$$



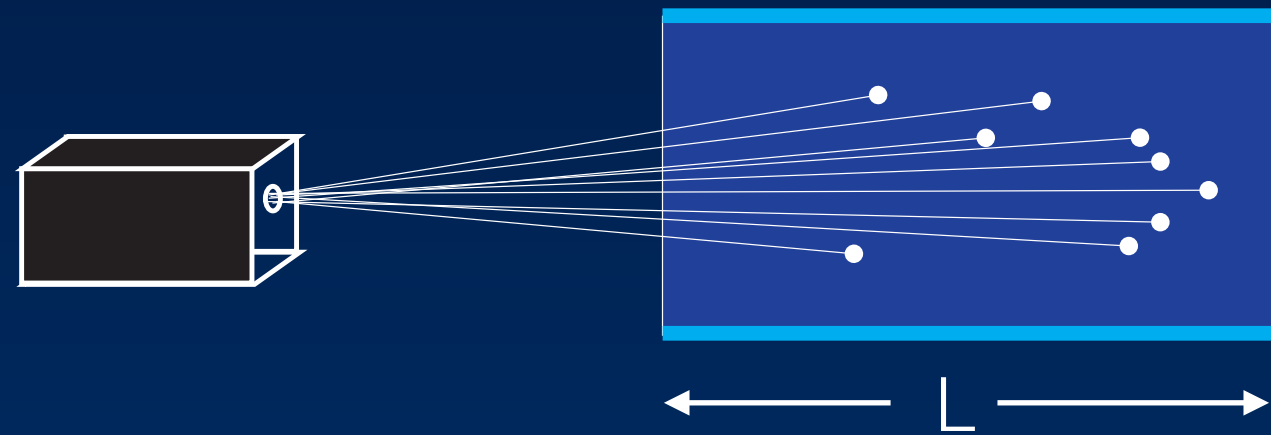
Cold molecules offer longer coherent interaction times

fast beam

$$\tau \sim 1-2 \text{ ms}$$

$$L \sim 0.5 \text{ m}$$

$$v \sim 250-500 \text{ m/s}$$

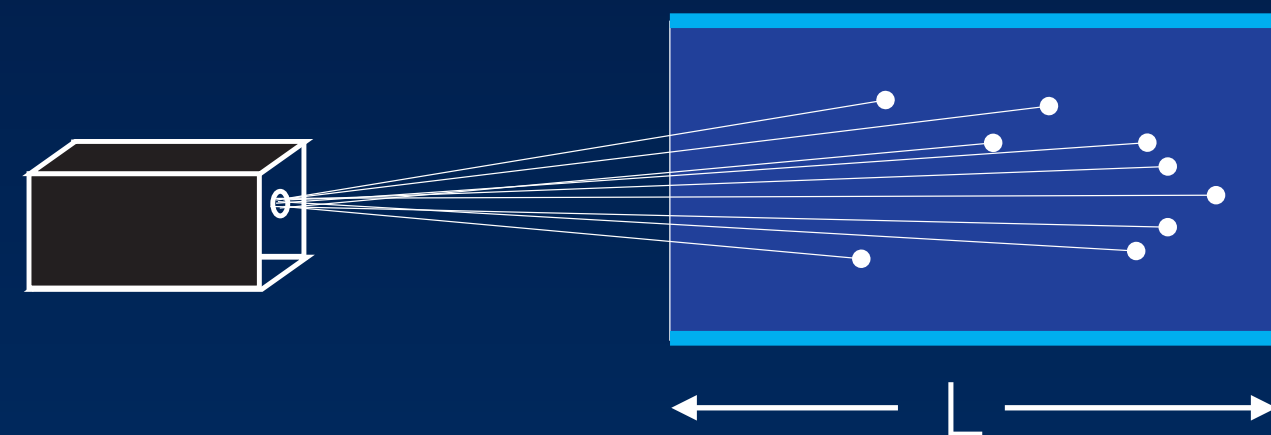


slow beam

$$\tau \sim 15 \text{ ms}$$

$$L \sim 0.5 \text{ m}$$

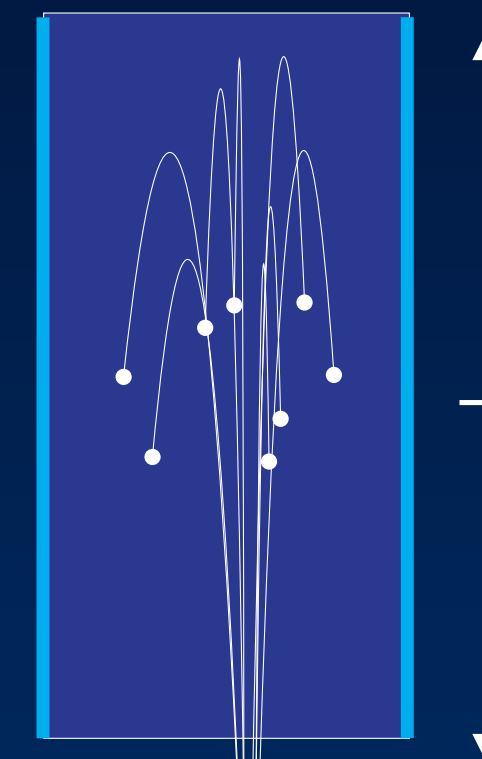
$$v \sim 30 \text{ m/s}$$



fountain

$$\tau \sim 100 \text{ ms}$$

$$L \sim 0.5 \text{ m}$$

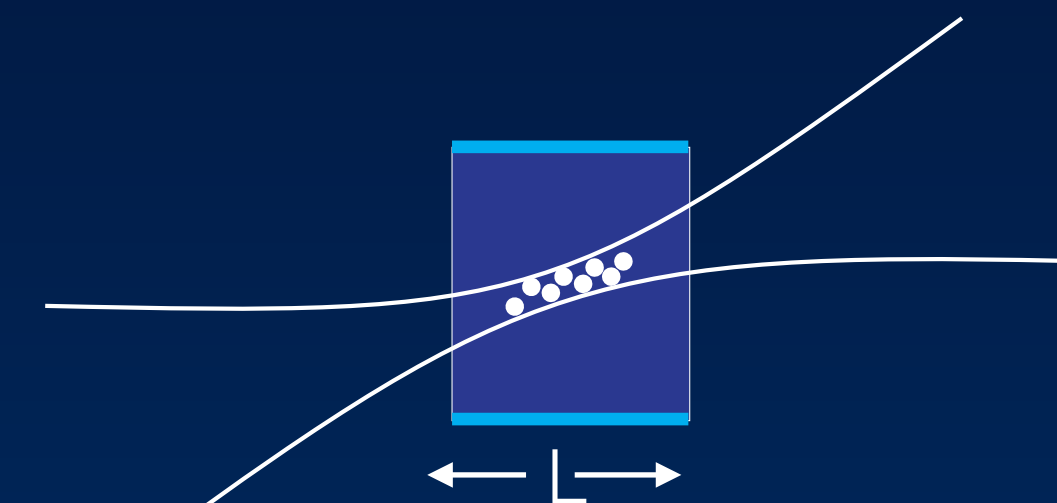


slow vertical beam

trap

$$\tau \sim 1-10 \text{ s}$$

$$L \sim 0.5 \text{ mm}$$

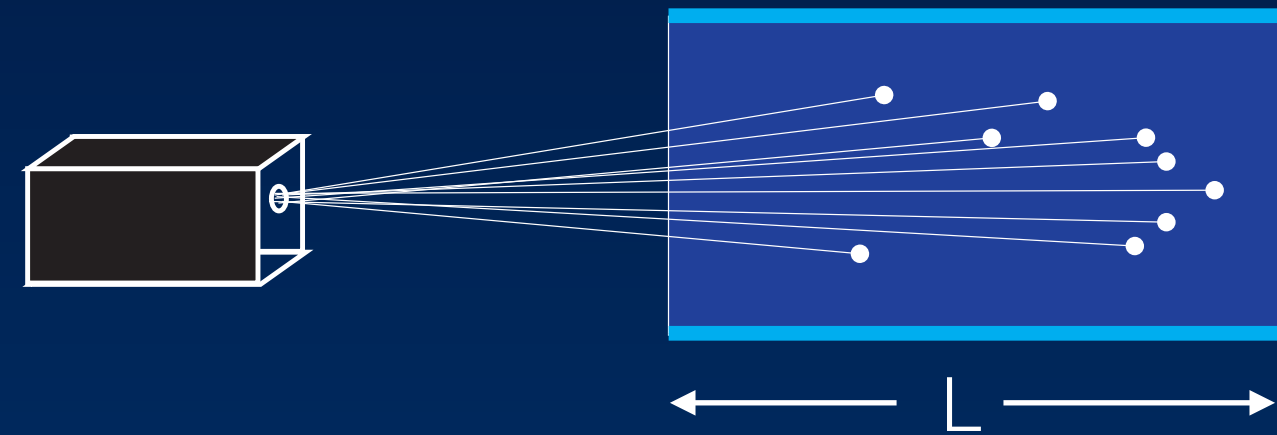


molecules trapped in
laser focus

Cold molecules offer longer coherent interaction times

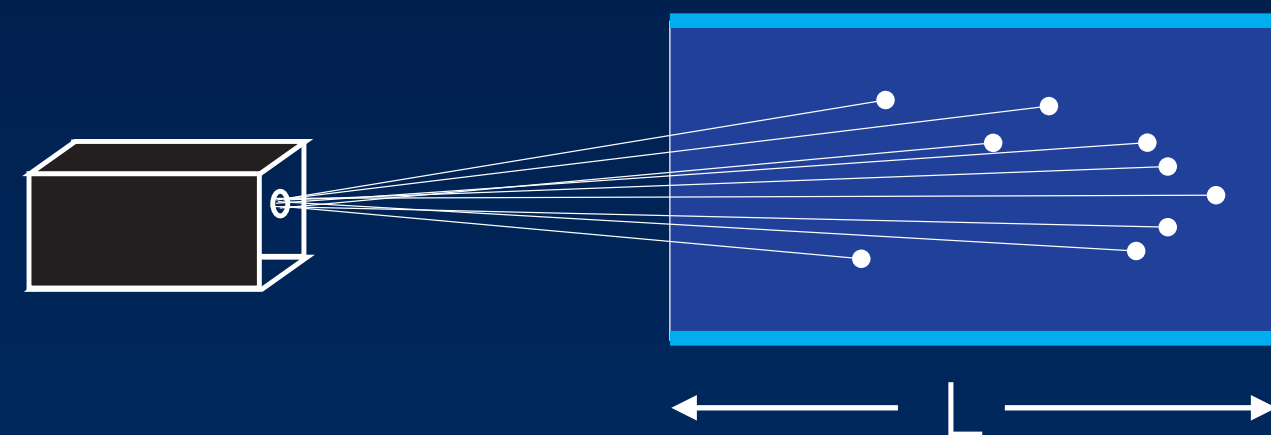
fast beam

$\tau \sim 1-2$ ms
 $L \sim 0.5$ m
 $v \sim 250-500$ m/s



slow beam

$\tau \sim 15$ ms
 $L \sim 0.5$ m
 $v \sim 30$ m/s



fountain

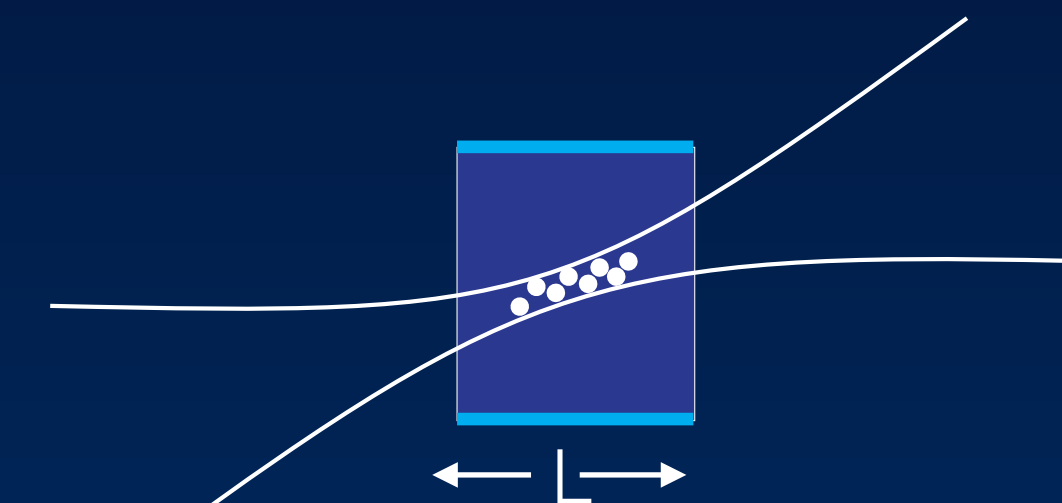
$\tau \sim 100$ ms
 $L \sim 0.5$ m



slow vertical beam

trap

$\tau \sim 1-10$ s
 $L \sim 0.5$ mm



molecules trapped in
laser focus

Main challenge:

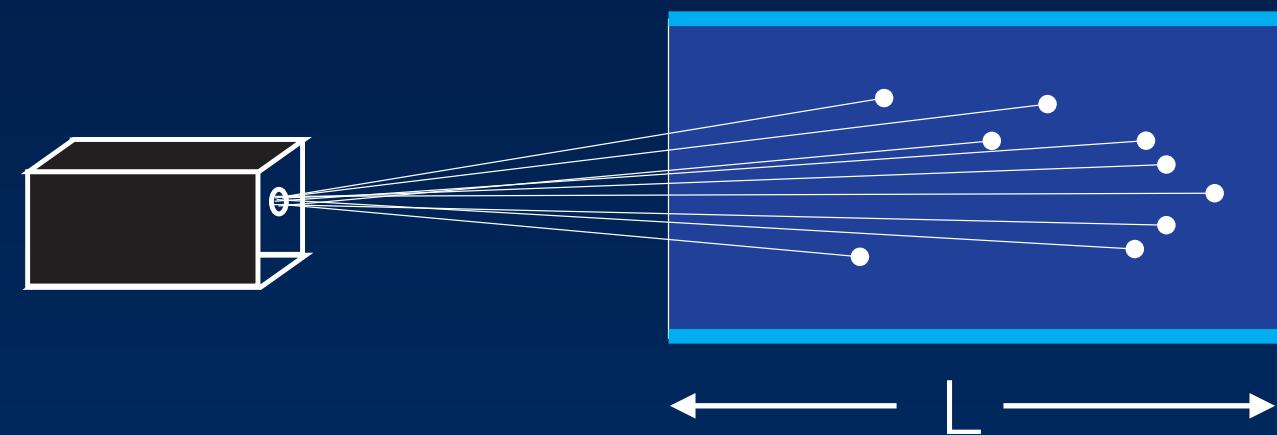
how to maintain N while increasing t

Strongly connected to choice of molecule!

Cold molecules offer longer coherent interaction times

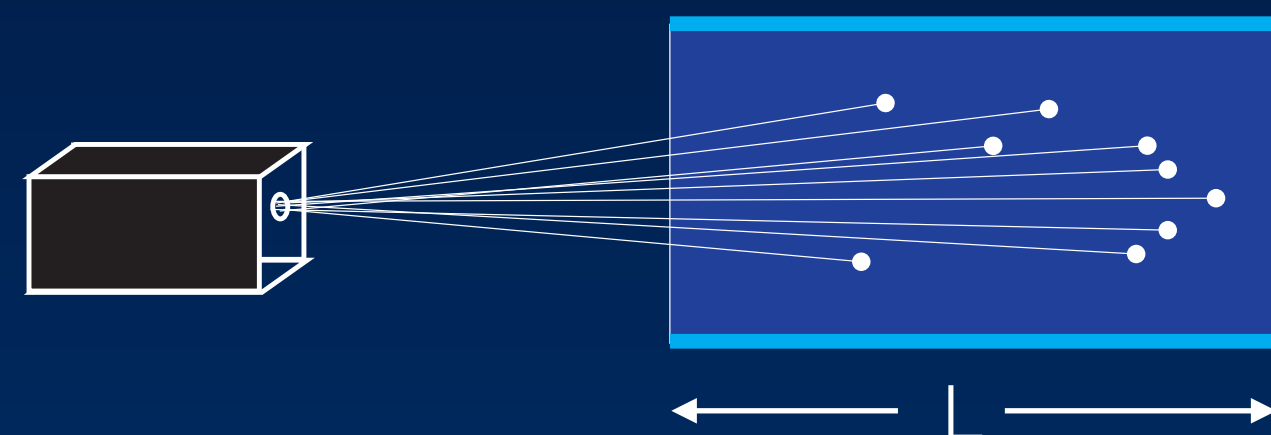
fast beam

$\tau \sim 1-2$ ms
 $L \sim 0.5$ m
 $v \sim 250-500$ m/s



slow beam

$\tau \sim 15$ ms
 $L \sim 0.5$ m
 $v \sim 30$ m/s



fountain

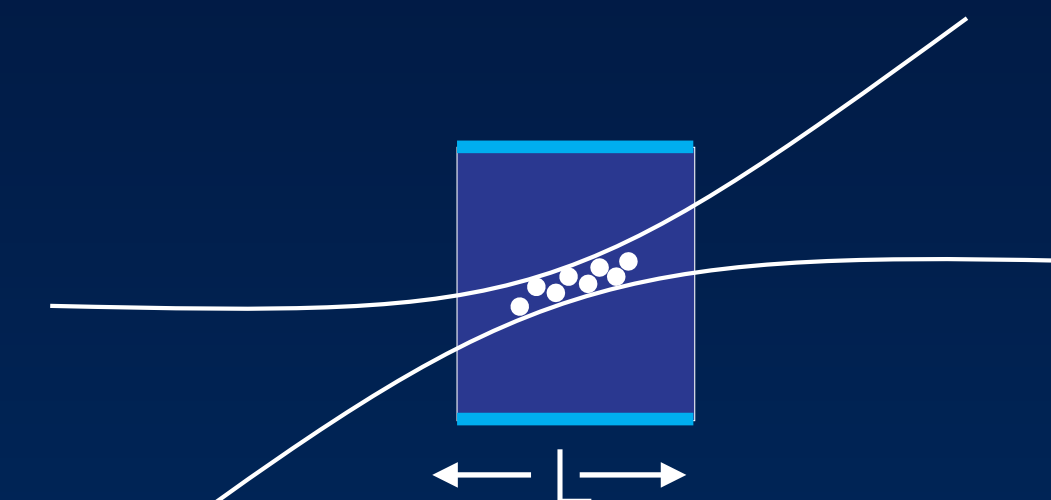
$\tau \sim 100$ ms
 $L \sim 0.5$ m



slow vertical beam

trap

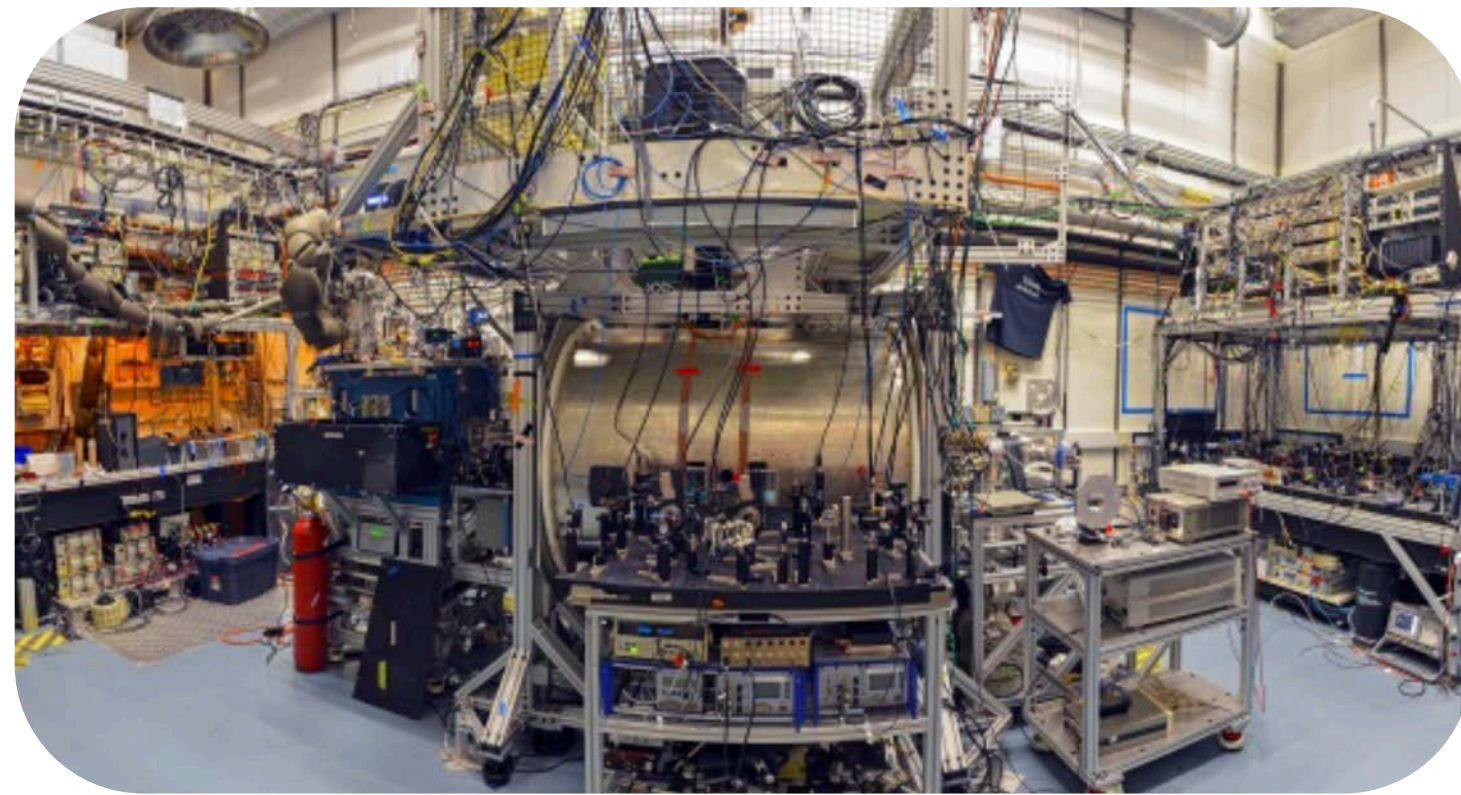
$\tau \sim 1-10$ s
 $L \sim 0.5$ mm



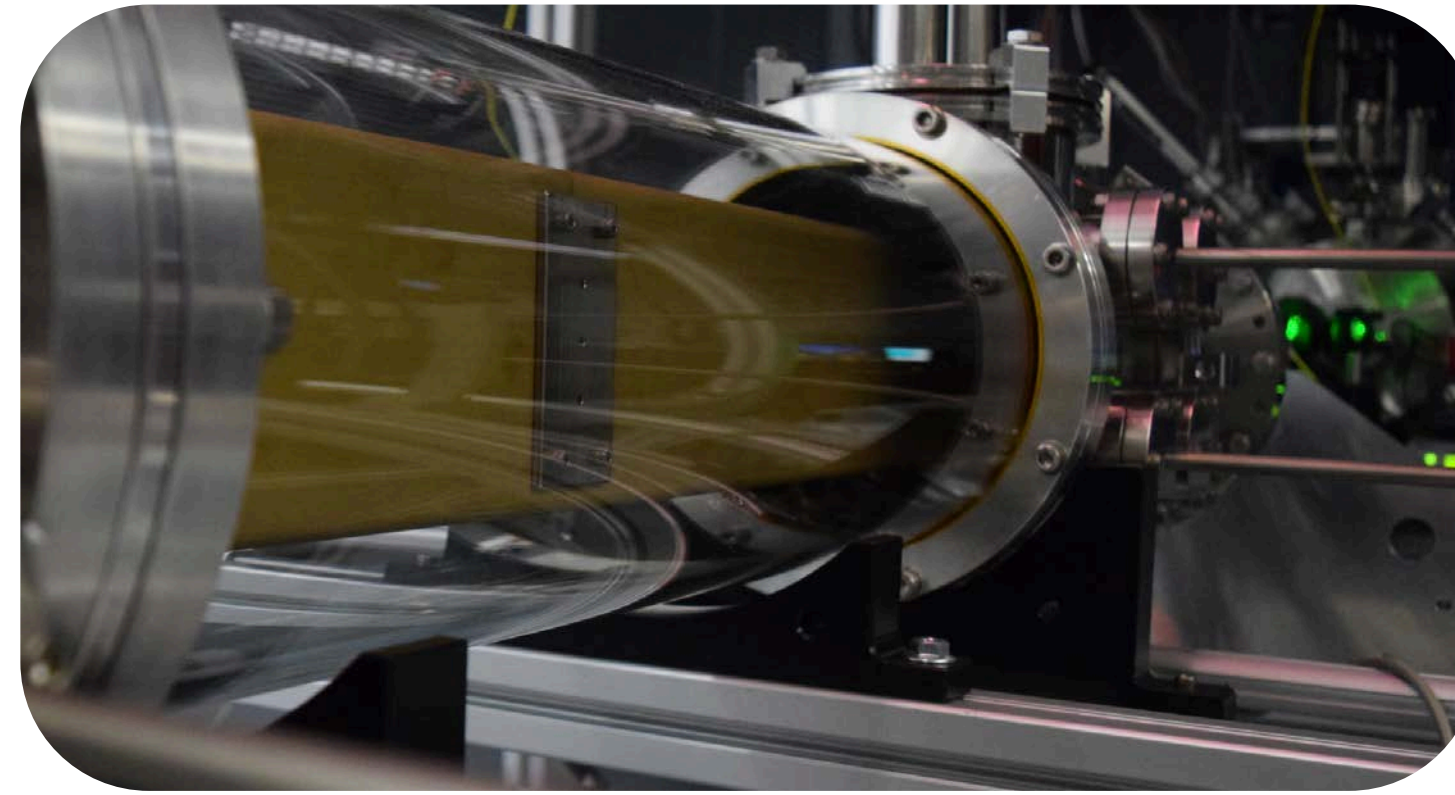
molecules trapped in
laser focus

Main challenge:
how to maintain N while increasing t
Strongly connected to choice of molecule!

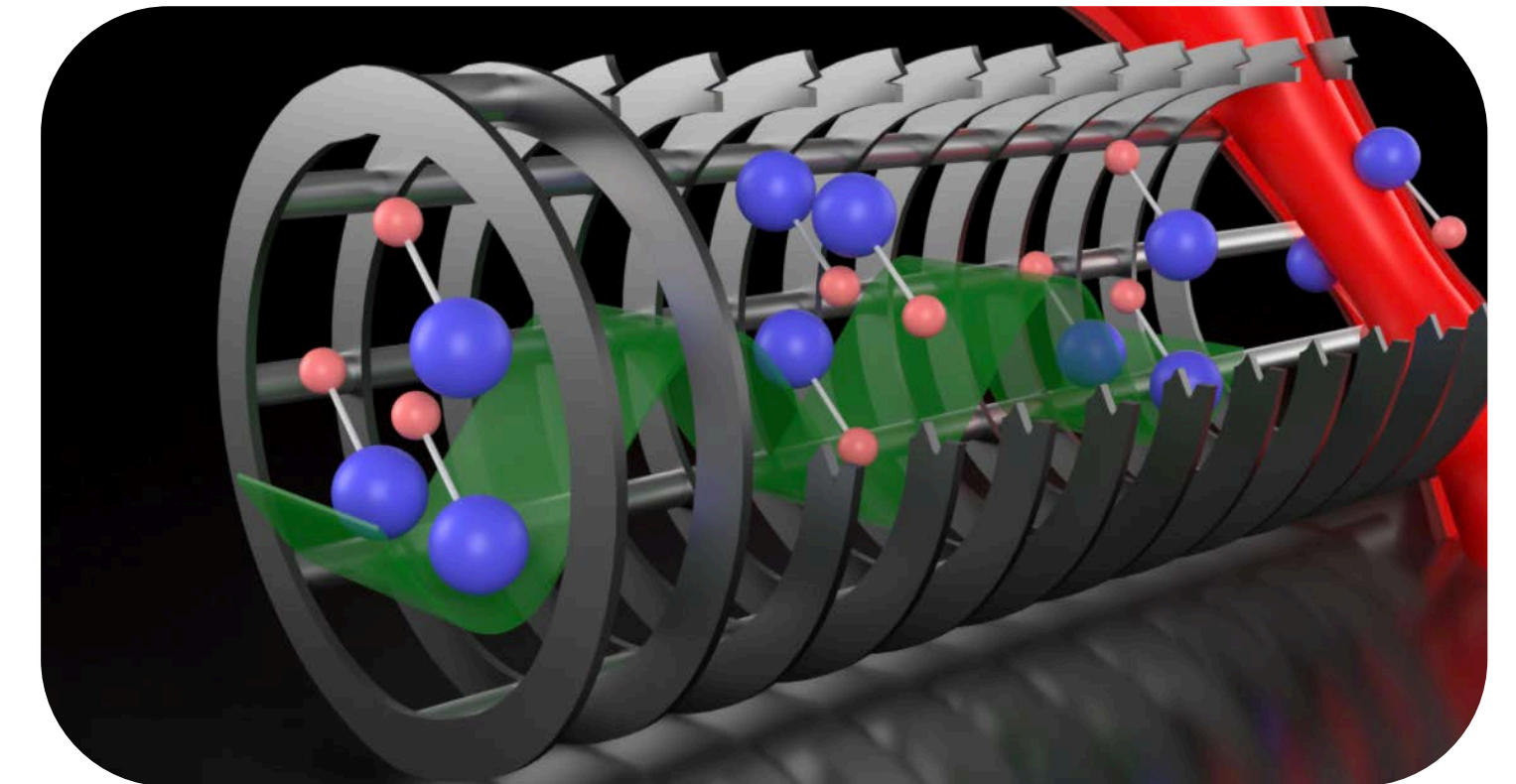
eEDM experiments using molecules



ACME - beam of ThO molecules
John Doyle, David DeMille,
Gerald Gabrielse



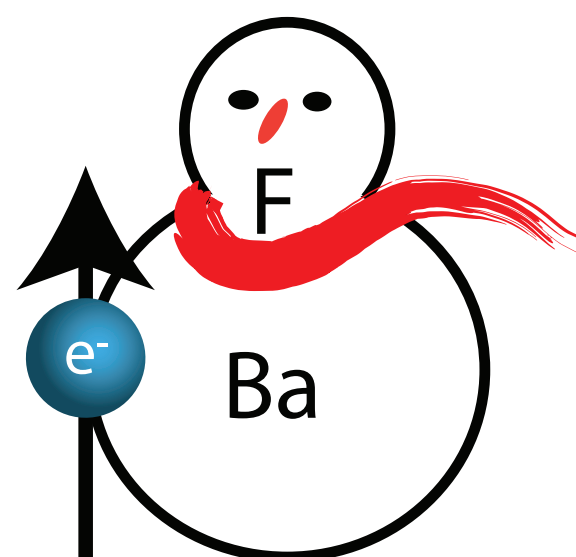
Imperial College London - beam of
YbF molecules
Mike Tarbutt, Ben Sauer, Ed Hinds



JILA - trapped HfF⁺ ions
Eric Cornell, Jun Ye

Others are being set up:

Electric Dipole Measurements using Molecules within a Matrix



Decelerated BaF beam
experiment in Groningen,
The Netherlands
(NL-eEDM)

Search for eEDM in cryogenic crystals

PHYDES:
Para-Hydrogen and Diatomic for eEDM Study

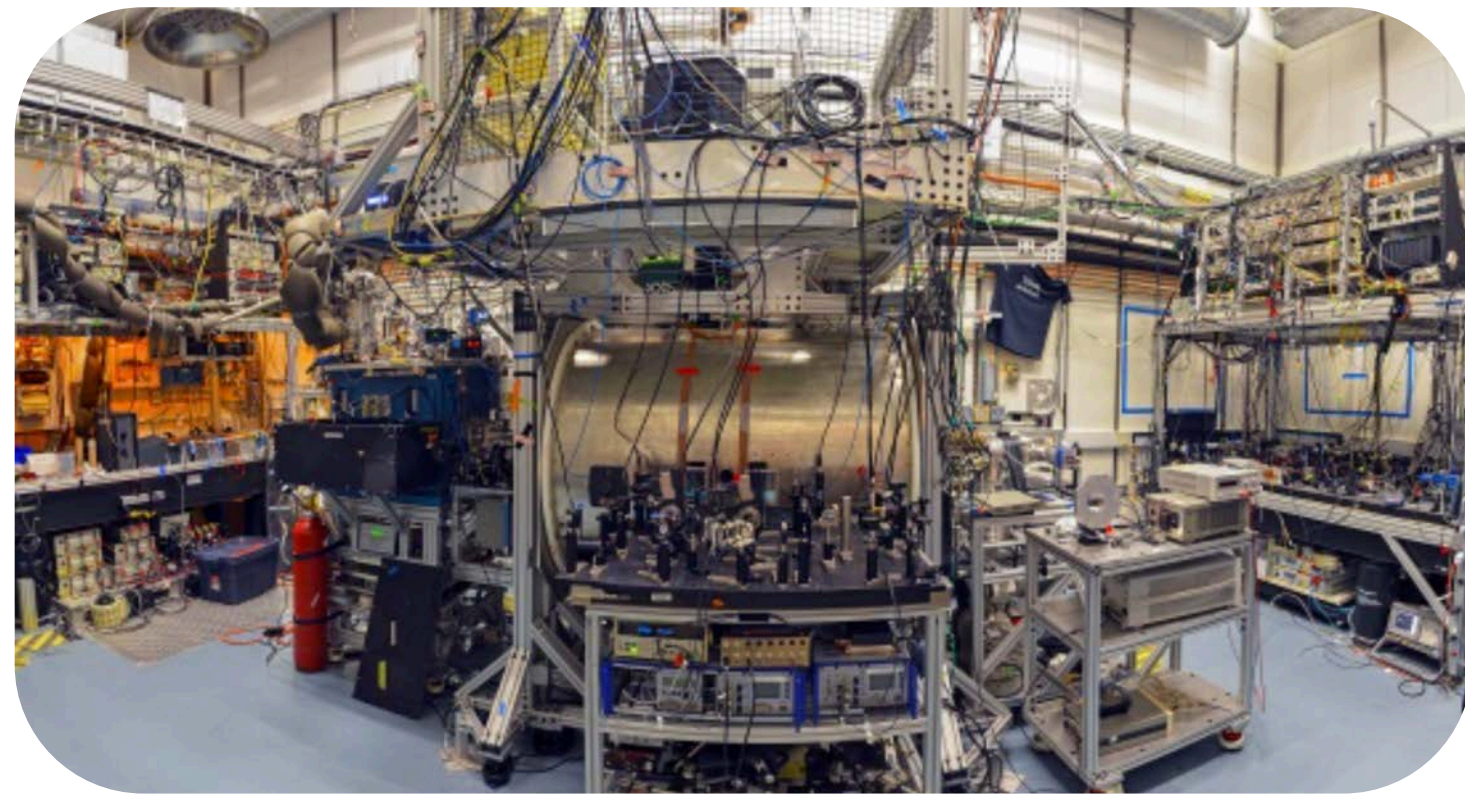


- York University
- Michigan State University
- University of Toronto

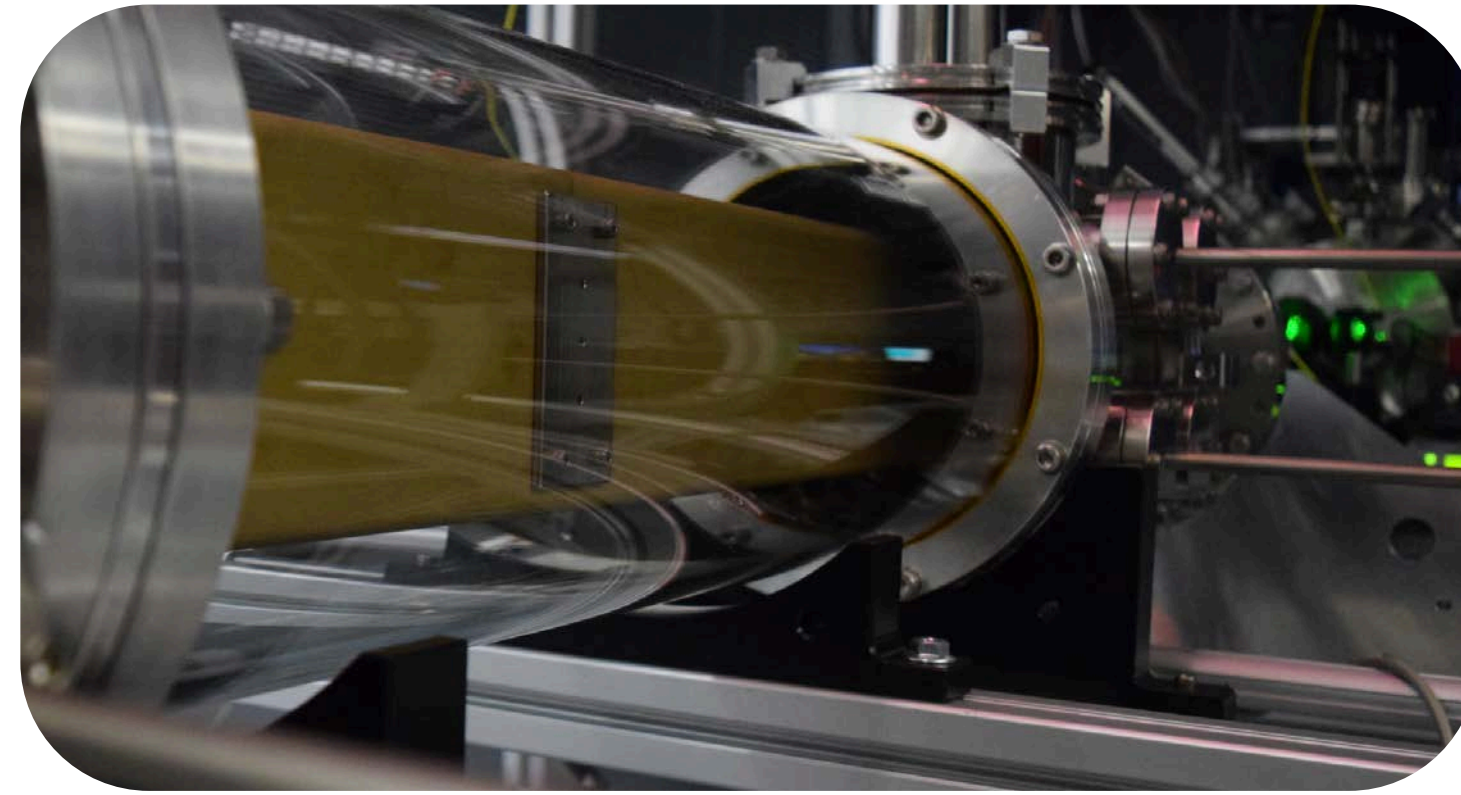


EDM³ Collaboration

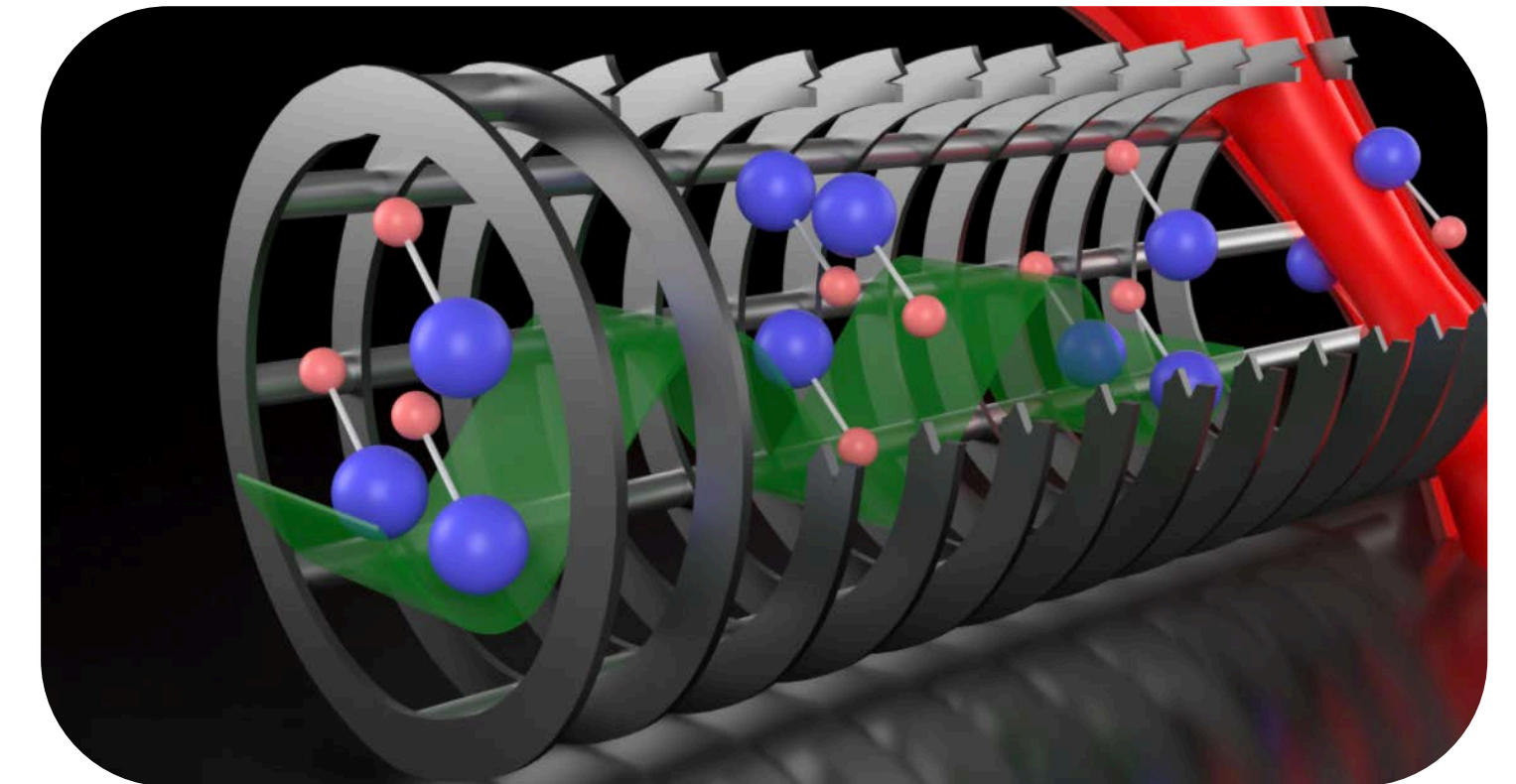
eEDM experiments using molecules



ACME - beam of ThO molecules
John Doyle, David DeMille,
Gerald Gabrielse



Imperial College London - beam of
YbF molecules
Mike Tarbutt, Ben Sauer, Ed Hinds



JILA - trapped HfF⁺ ions
Eric Cornell, Jun Ye

Others are being set up:

Electric Dipole Measurements using Molecules within a Matrix

Decelerated BaF beam
experiment in Groningen,
The Netherlands
(NL-eEDM)

Search for eEDM in cryogenic crystals

PHYDES:
Para-Hydrogen and Diatomic for eEDM Study

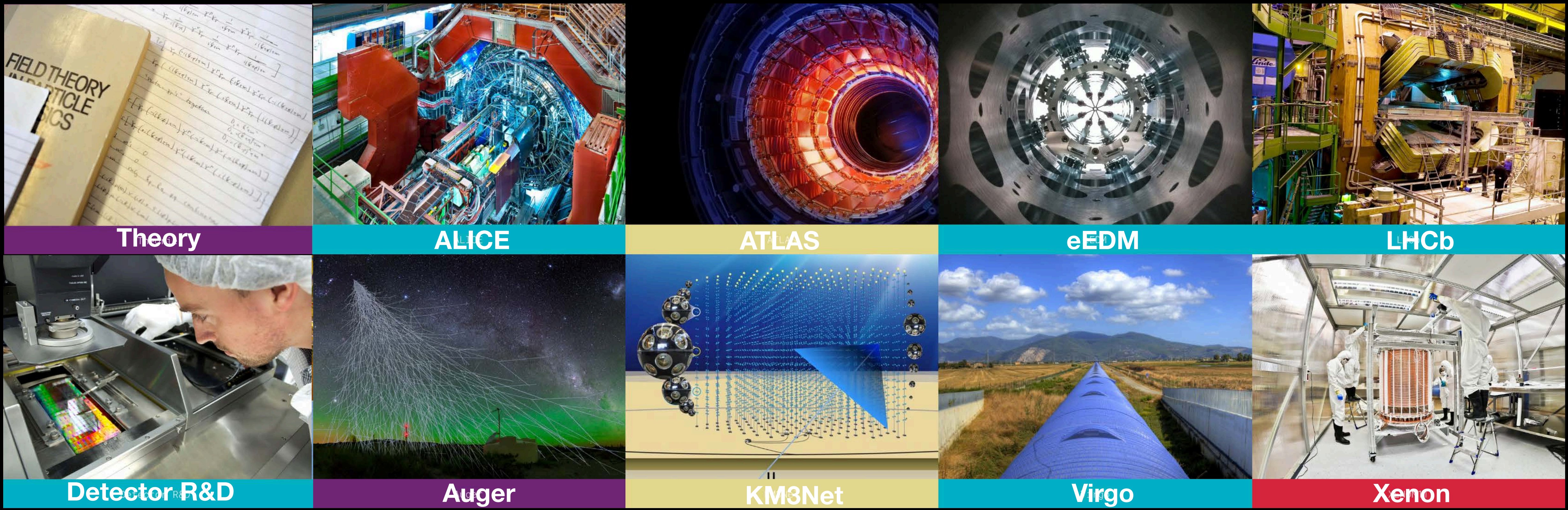


- York University
- Michigan State University
- University of Toronto



EDM³ Collaboration

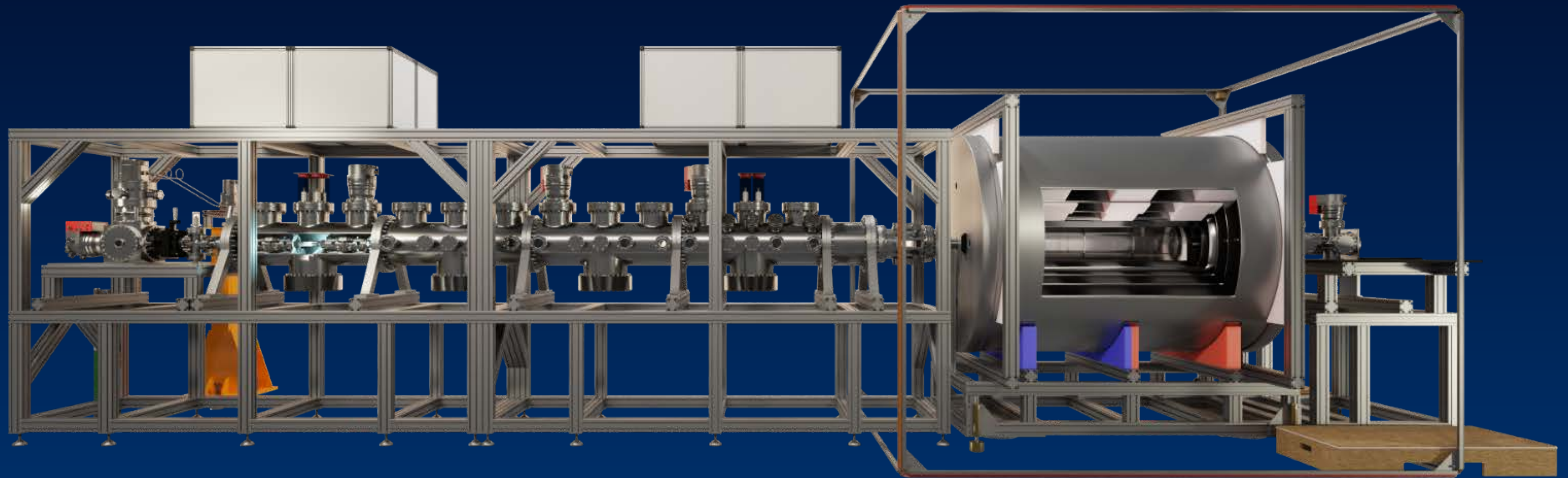
NL-eEDM: A Nikhef research programme started in 2017...



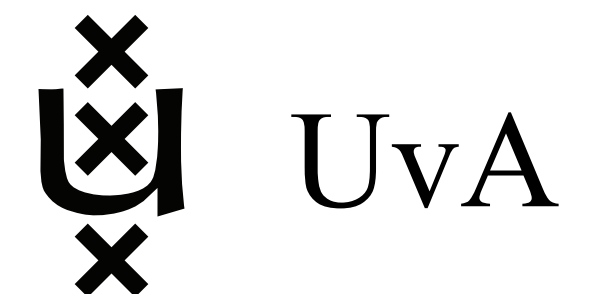
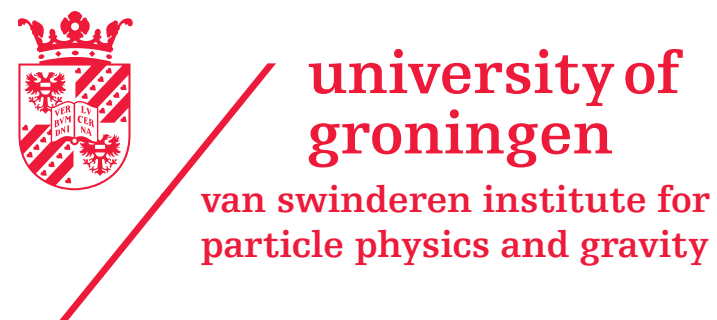
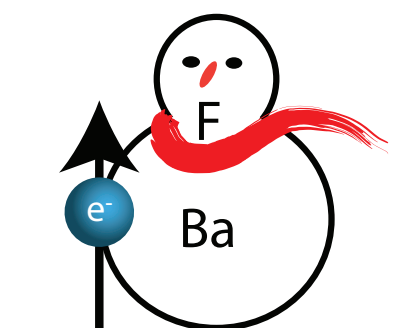
Dutch National Institute for (astro)Particle Physics

...using slow BaF molecules and lasers!

Cold molecules offer longer coherent interaction times



*The NL-eEDM collaboration, Measuring the electric dipole moment of the electron in BaF, *European Phys J D* 72, 197 (2018).*

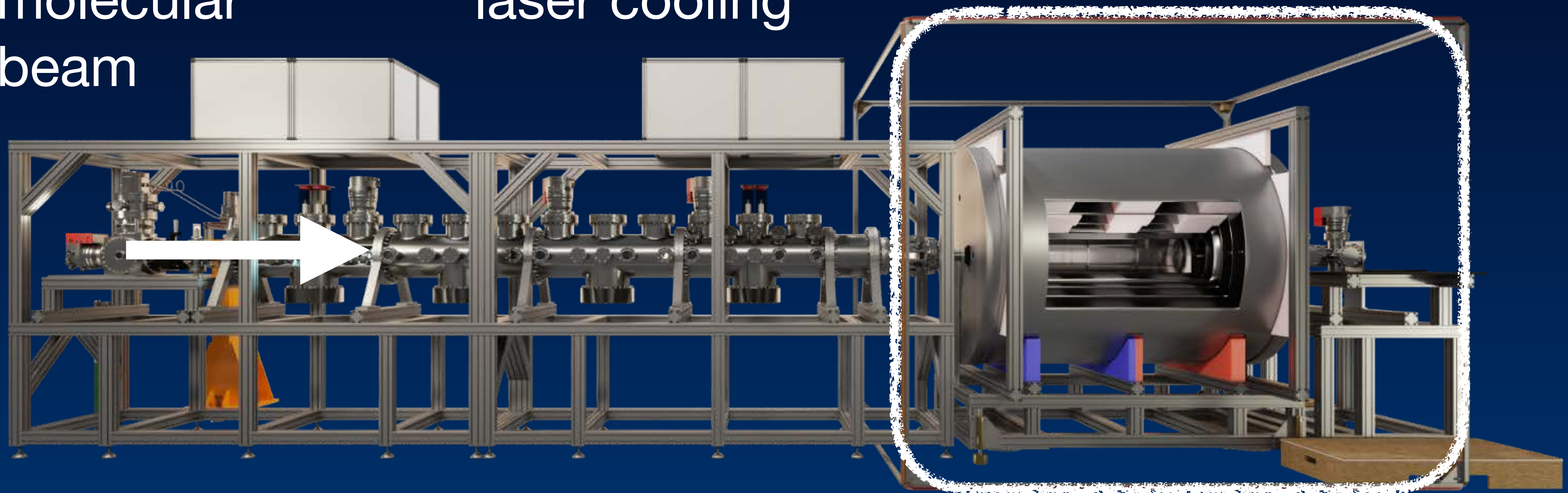


Cold molecules offer longer coherent interaction times

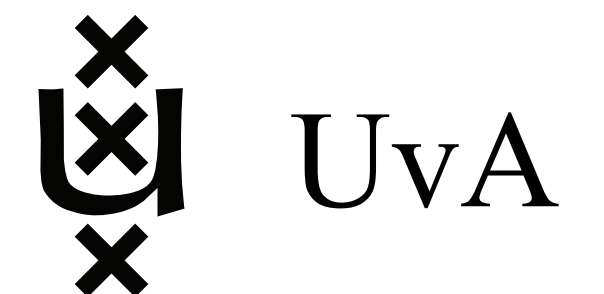
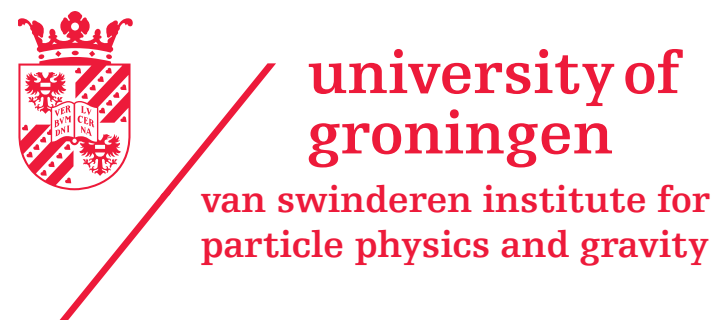
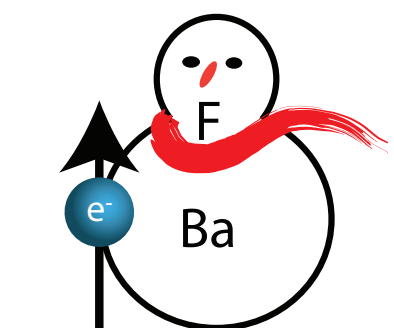
Pulsed
molecular
beam

deceleration &
laser cooling

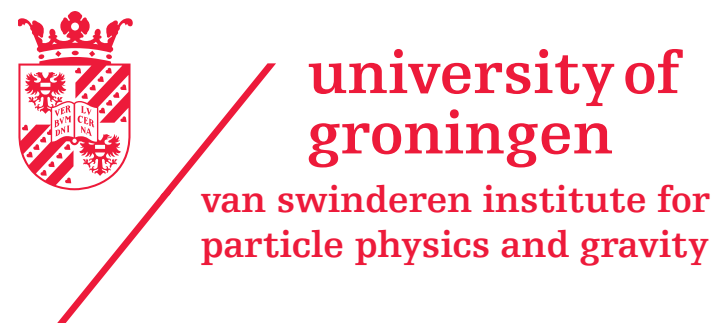
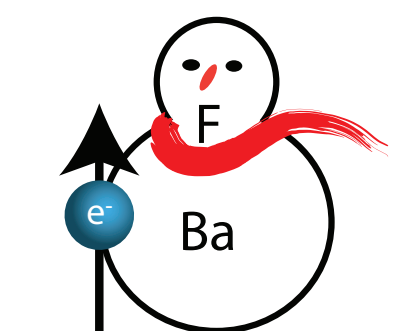
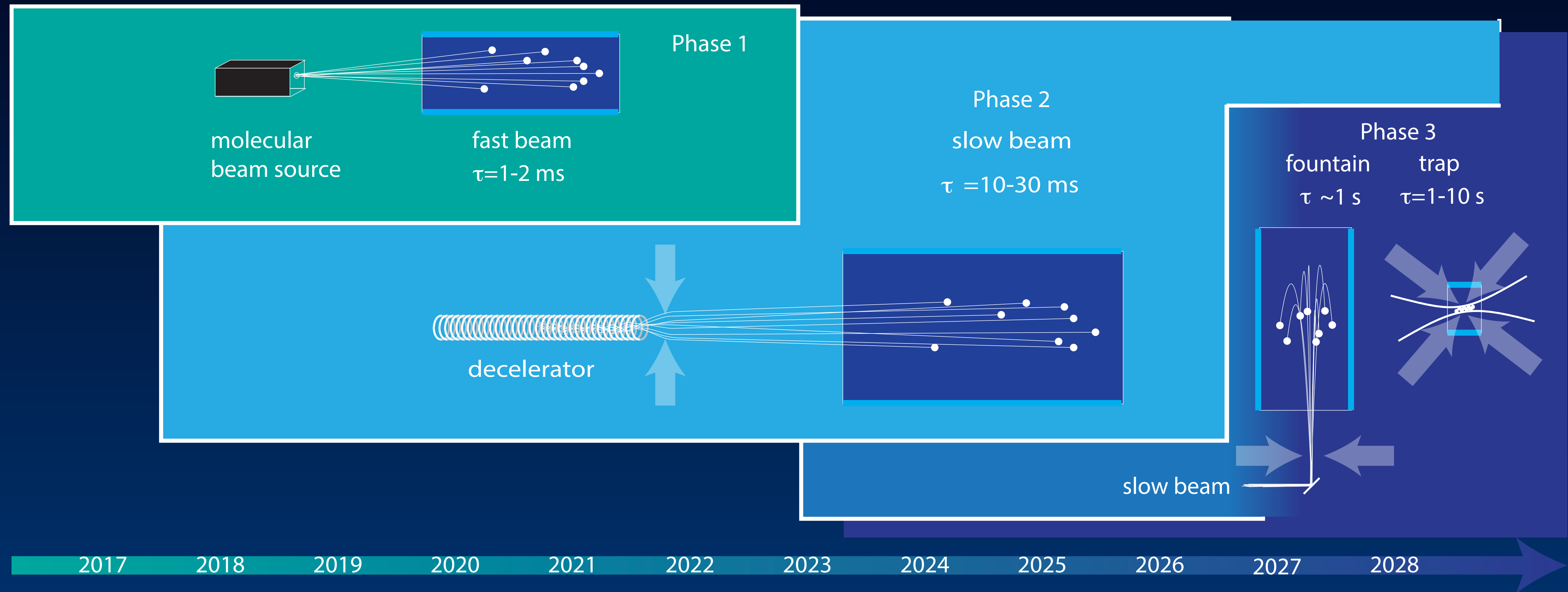
spin interferometer
to probe eEDM



The NL-eEDM collaboration, Measuring the electric dipole moment of the electron in BaF, European Phys J D 72, 197 (2018).



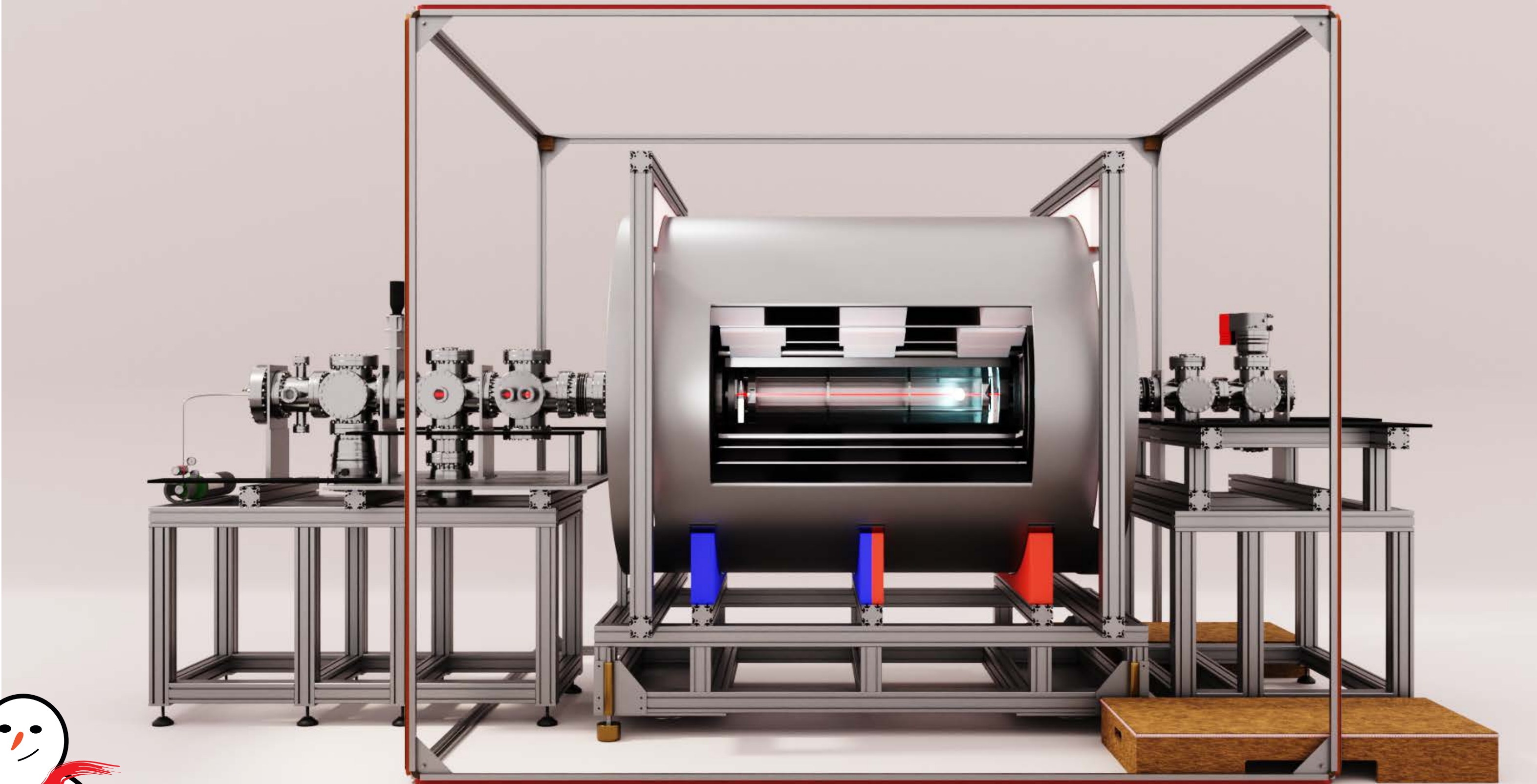
Cold molecules offer longer coherent interaction times



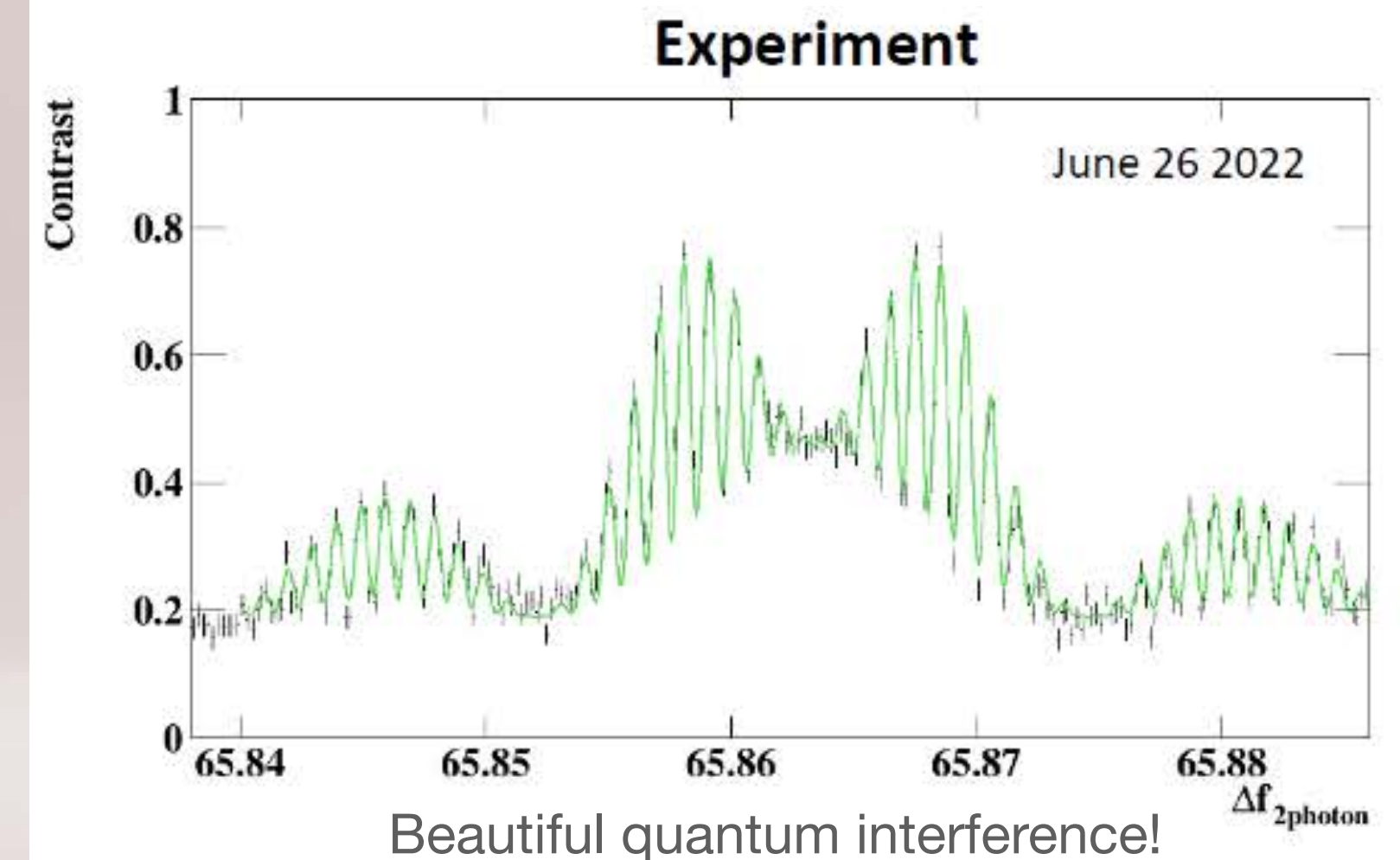
Interference data using fast molecular beam

to demonstrate control over systematic effects

Create molecular beam → Quantum interference → Readout by fluorescence



Compare to theory that includes the full interaction of the molecule with light, electric and magnetic fields (optical Bloch equations)

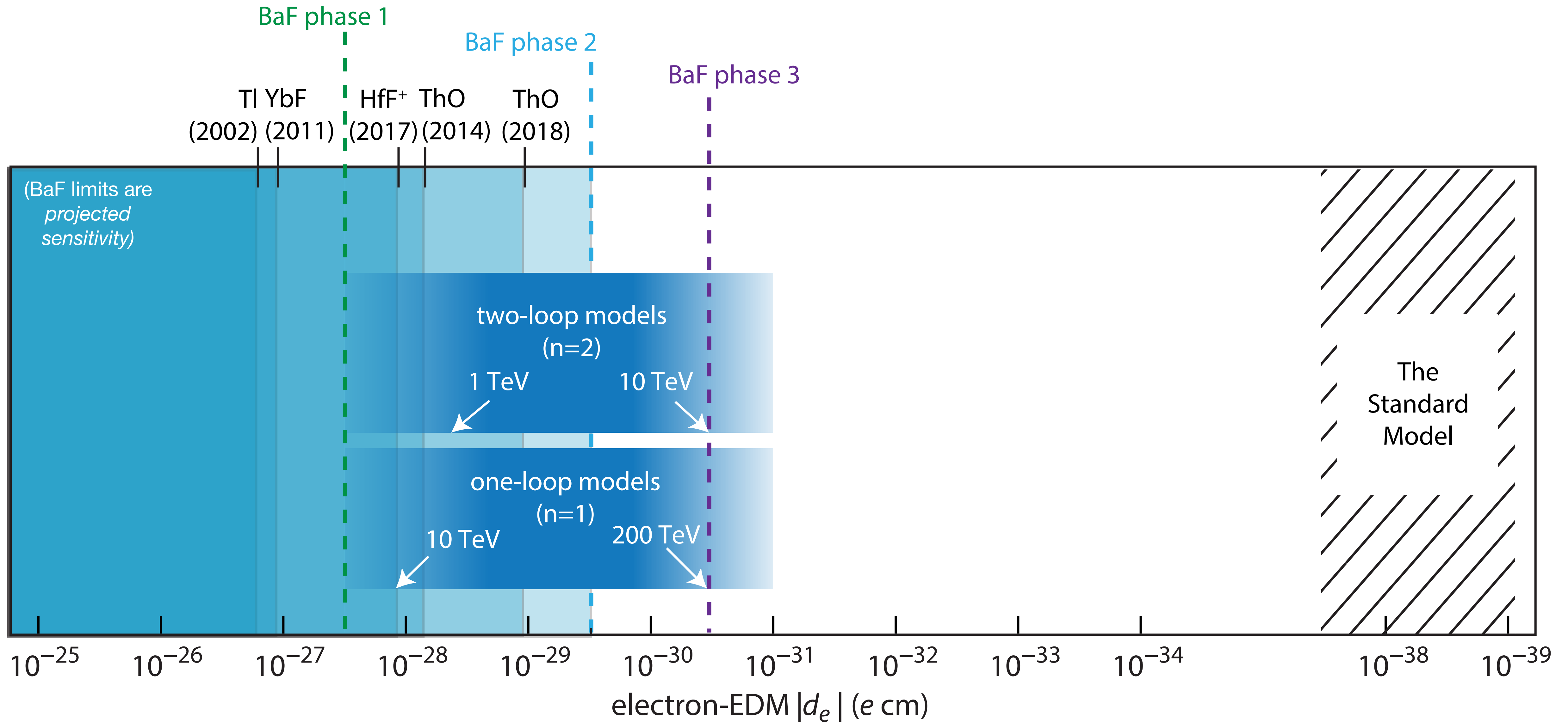


Contains all relevant experimental parameters
Crucial for reduction of systematic effects
(A.Boeschoten et al, NL-eEDM collaboration, in prep.)



NL-eEDM

... and a corresponding increase in eEDM sensitivity!



The choice of molecule

Exciting developments - some examples

Molecules containing radioactive elements

Proposal:

Isaev, T. A., Hoekstra, S. & Berger, R. Laser-cooled RaF as a promising candidate to measure molecular parity violation, *Phys Rev A* 82, 052521 (2010).





IN THE NEWS

[Physicists measure a short-lived radioactive molecule for first time](#)

Molecules containing heavy and deformed radioactive nuclei may help scientists to measure symmetry-violating phenomena and identify signs of dark matter.

Article | [Open Access](#) | [Published: 27 May 2020](#)

Spectroscopy of short-lived radioactive molecules

[R. F. Garcia Ruiz](#) , [R. Berger](#) , [J. Billowes](#), [C. L. Binnersley](#), [M. L. Bissell](#), [A. A. Breier](#), [A. J. Brinson](#), [K. Chrysalidis](#), [T. E. Cocolios](#), [B. S. Cooper](#), [K. T. Flanagan](#), [T. F. Giesen](#), [R. P. de Groote](#), [S. Franchoo](#), [F. P. Gustafsson](#), [T. A. Isaev](#), [Á. Koszorús](#), [G. Neyens](#), [H. A. Perrett](#), [C. M. Ricketts](#), [S. Rothe](#), [L. Schweikhard](#), [A. R. Vernon](#), [K. D. A. Wendt](#), [F. Wienholtz](#), [S. G. Wilkins](#) & [X. F. Yang](#) — [Show fewer authors](#)

[Nature](#) **581**, 396–400 (2020) | [Cite this article](#)



Polyatomic molecules

Proposal:

Kozyryev, I. & Hutzler, N. R. Precision Measurement of Time-Reversal Symmetry Violation with Laser-Cooled Polyatomic Molecules, *Physical Review Letters* 119, 133002 (2017).



[John M. Doyle](#)

Harvard University



[Nick Hutzler](#)

Caltech



[Tim Steimle](#)

Arizona State
University



[Amar Vutha](#)

University of Toronto

Advanced cooling techniques

Direct laser cooling - now extending to heavier and more complex species

Anderegg *et al.*, *Science* **365**, 1156–1158 (2019)

COLD MOLECULES

An optical tweezer array of ultracold molecules

Loïc Anderegg^{1,2*}, Lawrence W. Cheuk^{1,2}, Yicheng Bao^{1,2}, Sean Burchesky^{1,2}, Wolfgang Ketterle^{2,3}, Kang-Kuen Ni^{1,2,4}, John M. Doyle^{1,2}

Trapping of C_2^- in a digital ion trap

Alexander Hinterberger, Sebastian Gerber , Emanuel Oswald, Christan Zimmer, Julian Fesel and Michael Doser

CERN, European Laboratory for Particle Physics, 1211 Geneva, Switzerland

Journal of Physics B: Atomic, Molecular and Optical Physics, Volume 52, Number 22

Towards laser cooling of molecular anions - for antimatter physics

nature
physics

LETTERS

PUBLISHED ONLINE: 28 AUGUST 2017 | DOI: 10.1038/NPHYS4241

Molecules cooled below the Doppler limit

S. Truppe, H. J. Williams, M. Hambach, L. Caldwell, N. J. Fitch, E. A. Hinds, B. E. Sauer and M. R. Tarbutt*

Advanced cooling techniques

Exciting developments - some examples

A molecular fountain

PRL 117, 253201 (2016)

PHYSICAL REVIEW LETTERS

week ending
16 DECEMBER 2016



Molecular Fountain

Cunfeng Cheng, Aernout P. P. van der Poel, Paul Jansen,^{*} Marina Quintero-Pérez,[†] Thomas E. Wall,[‡]

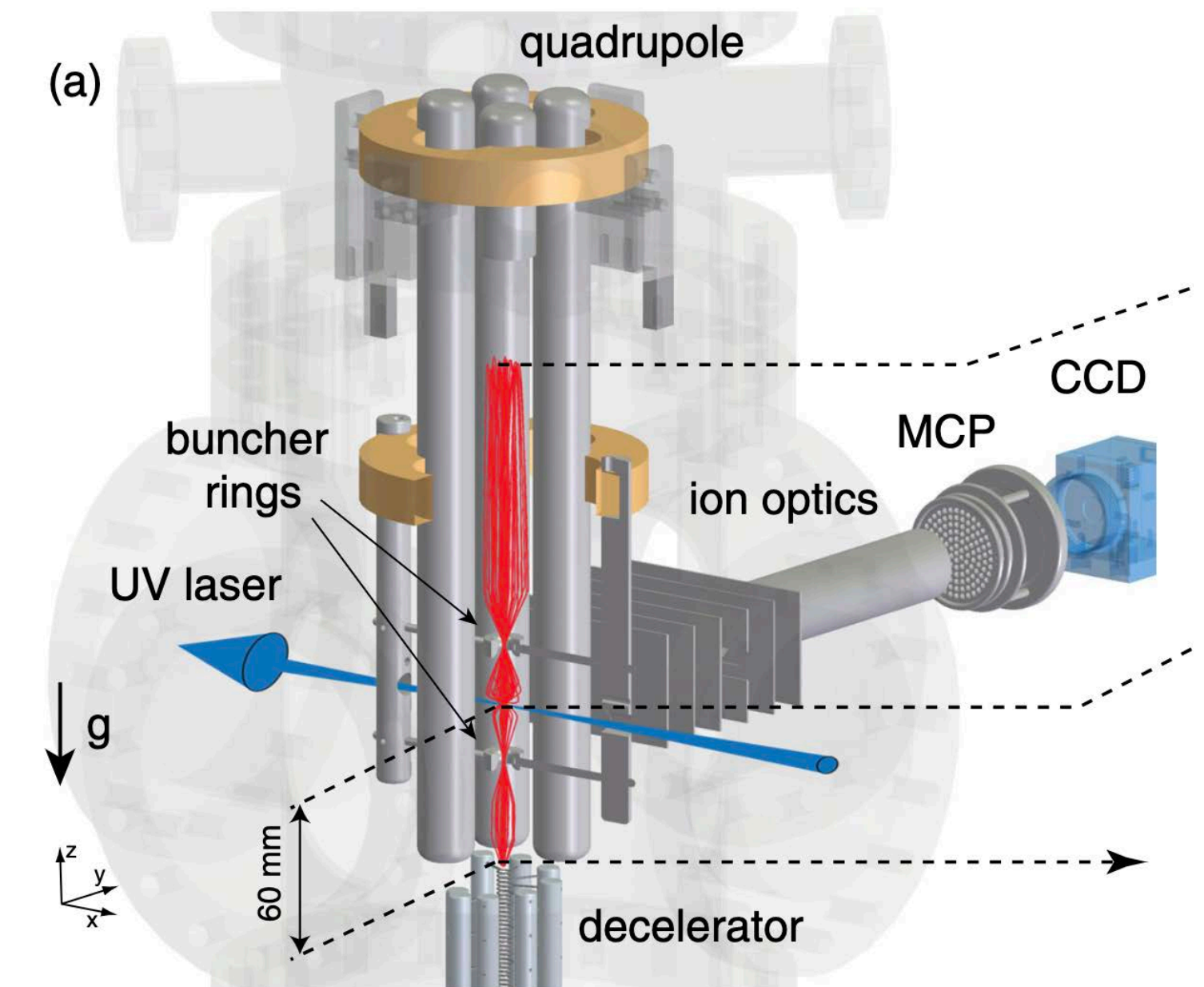
Wim Ubachs, and Hendrick L. Bethlem

LaserLaB, Department of Physics and Astronomy, Vrije Universiteit, De Boelelaan 1081,
1081 HV Amsterdam, The Netherlands

(Received 17 October 2016; published 13 December 2016)

PRL 117, 253201 (2016)

PHYSICAL REVIEW



Deceleration and trapping of heavy diatomic molecules

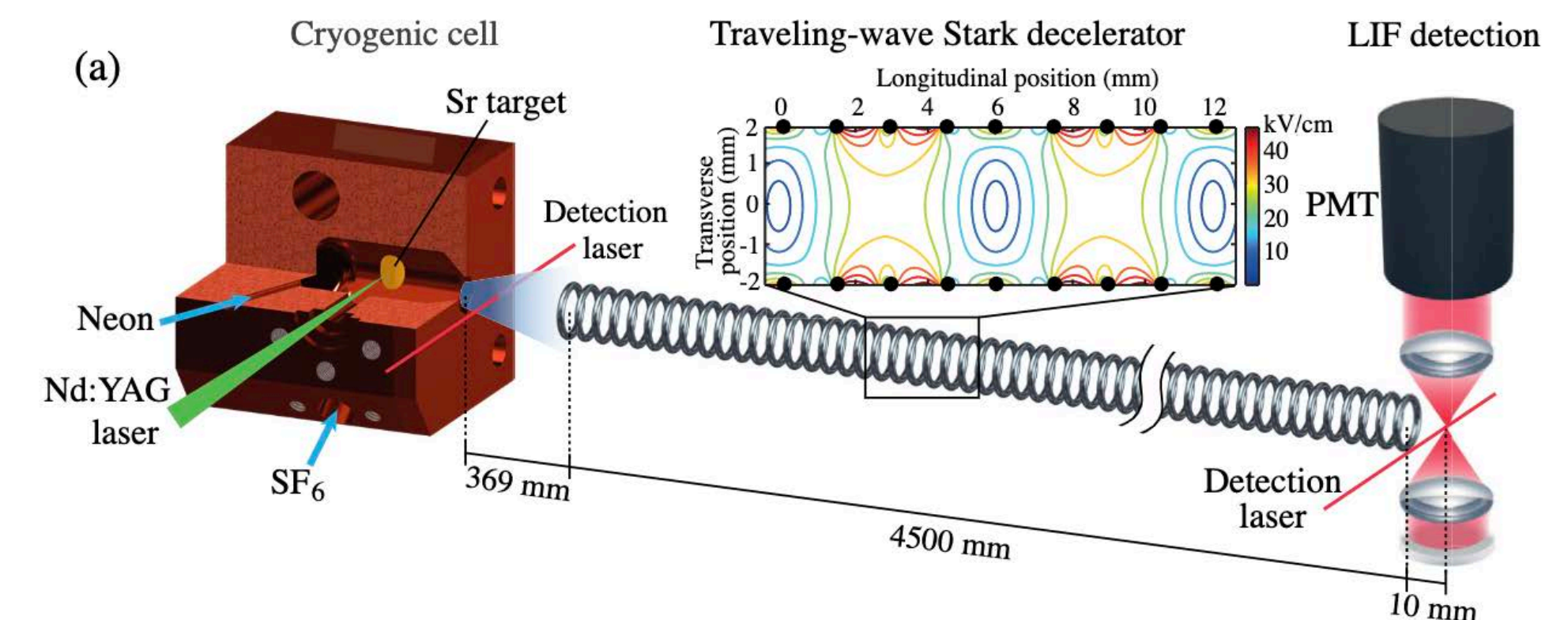
PHYSICAL REVIEW LETTERS 127, 173201 (2021)

Featured in Physics

Deceleration and Trapping of SrF Molecules

P. Aggarwal^{1,2,*} Y. Yin^{1,2,*} K. Esajas^{1,2} H. L. Bethlem^{1,3} A. Boeschoten^{1,2} A. Borschevsky^{1,2} S. Hoekstra^{1,2,†}
K. Jungmann^{1,2} V. R. Marshall^{1,2} T. B. Meijknecht^{1,2} M. C. Mooij^{2,3} R. G. E. Timmermans^{1,2} A. Touwen^{1,2}
W. Ubachs³ and L. Willmann^{1,2}

(NL-eEDM Collaboration)



Outlook: beyond Ramsey interferometry

Make use of entanglement and squeezing techniques

Science 352, 1552–1555 (2016)

REPORTS

QUANTUM PHYSICS

Quantum phase magnification

O. Hosten, R. Krishnakumar, N. J. Engelsen, M. A. Kasevich*

Quantum metrology exploits entangled states of particles to improve sensing precision beyond the limit achievable with uncorrelated particles. All previous methods required detection noise levels below this standard quantum limit to realize the benefits of the intrinsic sensitivity provided by these states. We experimentally demonstrate a widely applicable method for entanglement-enhanced measurements without low-noise detection. The method involves an intermediate quantum phase magnification step that eases implementation complexity. We used it to perform squeezed-state metrology 8 decibels below the standard quantum limit with a detection system that has a noise floor 10 decibels above the standard quantum limit.

NL-eEDM: Teamwork!



Scientific staff:
Anastasia Borschevsky
Rick Bethlem
Steven Hoekstra
Klaus Jungmann
Rob Timmermans
Wim Ubachs
Jordy de Vries
Lorenz Willmann

