ECFA Detector R&D Roadmap Symposium of Task Force 5
Quantum and Emerging Technologies

Novel ionic, atomic and molecular systems

thorium nuclear clock

https://thoriumclock.eu/

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Q-SEnSE
An NSF Quantum Leap Challenge Institute

https://www.colorado.edu/research/qsense/

NSF

NIST
National Institute of Standards and Technology
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Office of Naval Research
ONR
Science & Technology

erc
European Research Council

Many thanks to Klaus Blaum, José R. Crespo López-Urrutia, David Hanneke, Nick Hutzler, Andrew Jayich, Jeff Martoff, Piet Schmidt, and Ronald Fernando Garcia Ruiz for providing information on the present status of the experiments, perspectives on the future technological advances and sensitivities, and especially timeline information!

ECFA Detector R&D Roadmap Symposium of Task Force 5
Quantum and Emerging Technologies

Which quantum technologies are likely to lead to disruptive discoveries in fundamental physics in the next 10-20 years?

How do we define “quantum technology” and “quantum sensor”?

A technology or device that is naturally described by quantum mechanics is considered ``quantum''.

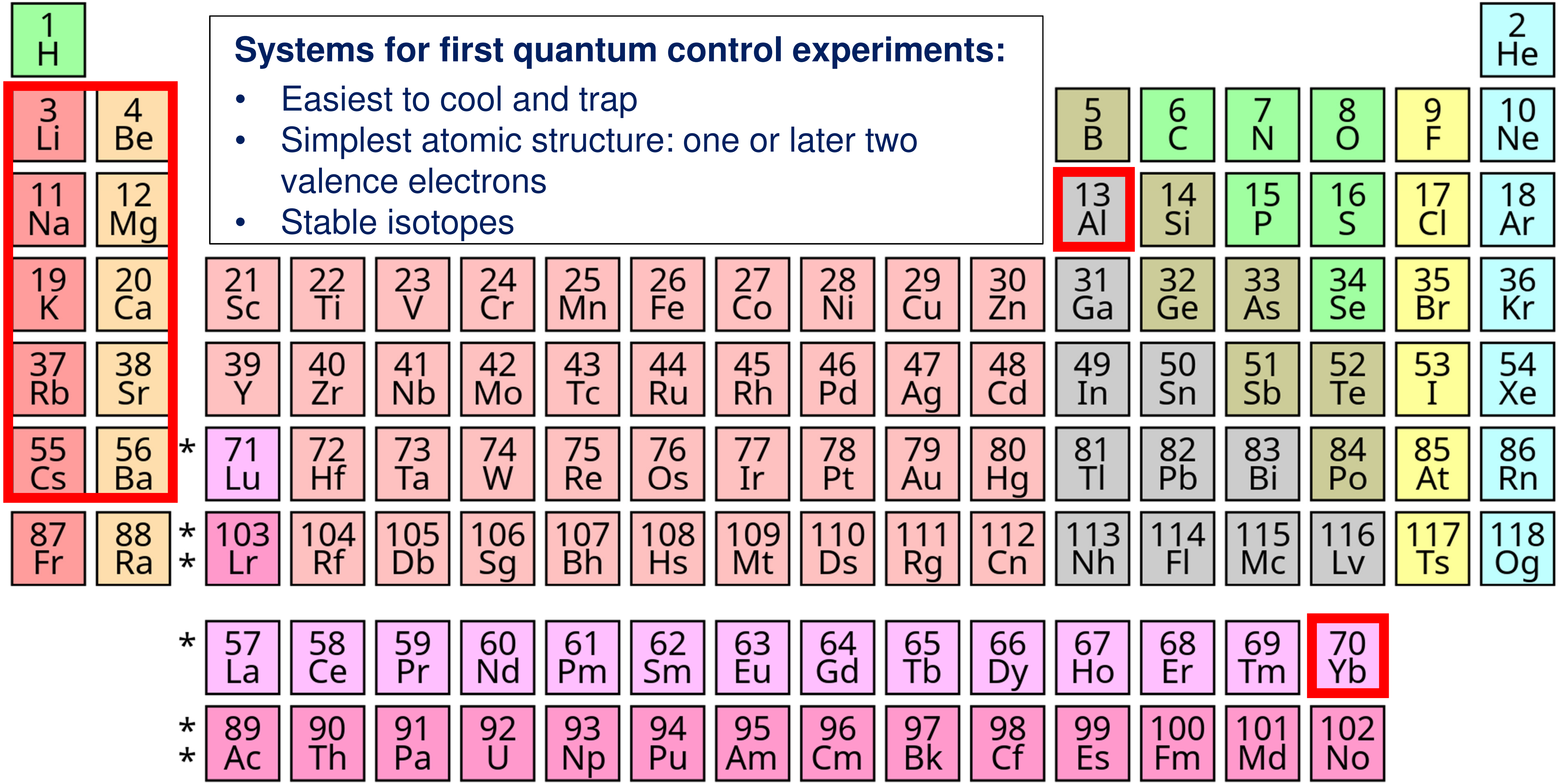
Then, ***a "quantum sensor" is a device, the measurement (sensing) capabilities of which are enabled by our ability to manipulate and read out its quantum states***.

Quantum Technology and the Elephants
Quantum Science and Technology Editorial
Marianna Safronova & Dmitry Budker

Why use novel systems?

Systems for first quantum control experiments:

- Easiest to cool and trap
- Simplest atomic structure: one or later two valence electrons
- Stable isotopes



Wikimedia Commons

Why use novel systems?

- **Much higher sensitivity for new physics or sensitivity to different new physics**

  Enhancements in heavy atoms, ions, and molecules with heavy atoms
  - Relativistic effects
  - Heavy nuclei (Z^3 or similar scaling)
  - Octupole deformed nuclei
  - Larger effective electric field (molecules for eEDM)

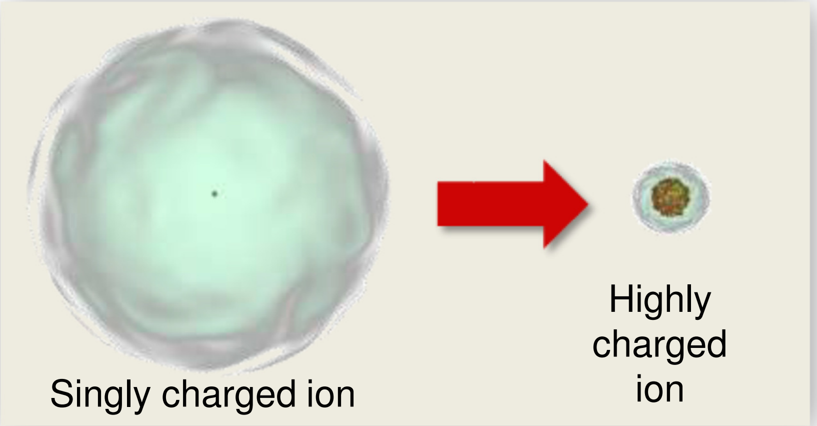
  Different types of transitions are available – sensitivity to different fundamental constants (molecules and molecular ions, highly-charged ions, nuclear clock)

  Need more isotopes or need a radioactive isotope

- **New systems have properties not available in currently used systems allowing for reduced systematics or better statistics**

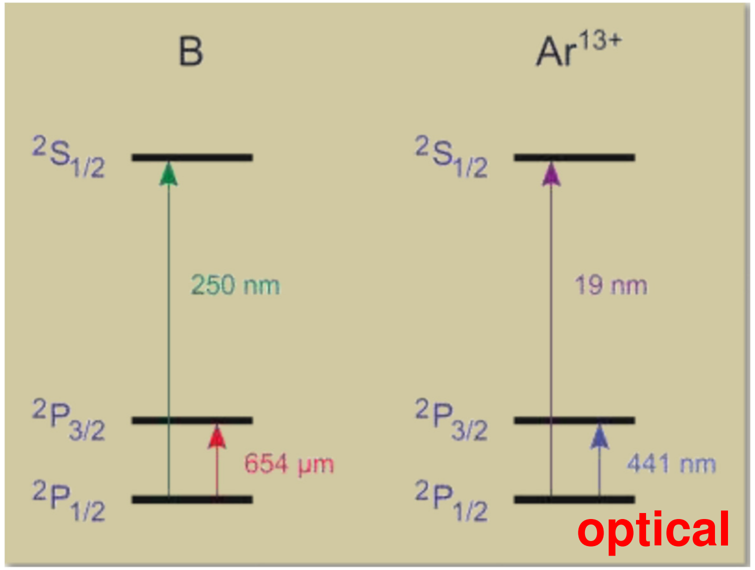
From building quantum sensors to dedicated new physics experiments

Novel systems: highly charged ions (HCIs)



Scaling with a nuclear charge Z

Binding energy	$\sim Z^2$
Hyperfine splitting	$\sim Z^3$
QED effects	$\sim Z^4$
Stark shifts	$\sim Z^{-6}$

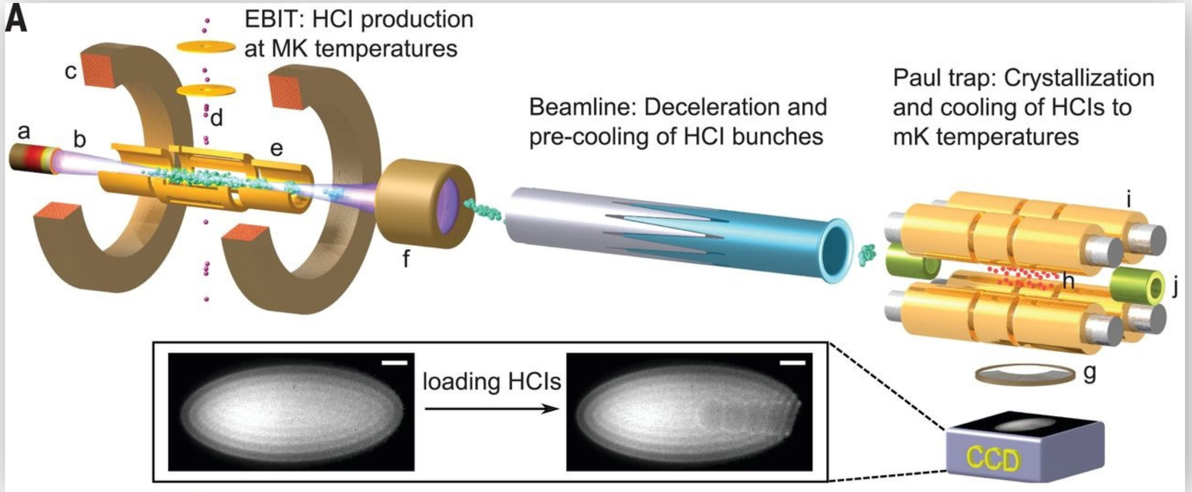


- Fine-structure, hyperfine-structure, and level-crossing transitions in range of table-top lasers
- Much higher sensitivity to new physics due to relativistic effects
- Rich variety of level structure not available in other systems
- Reduced systematics due to suppressed Stark shifts

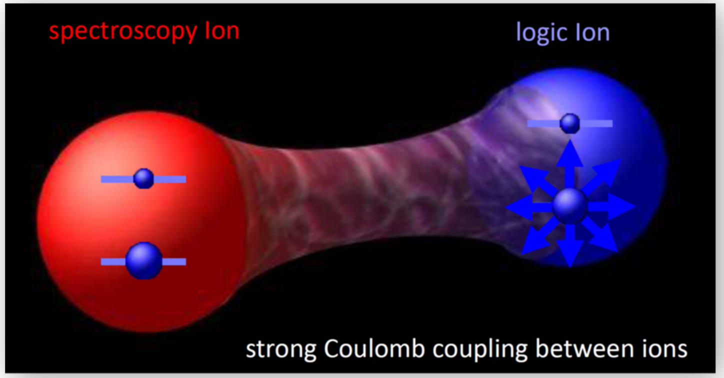
Review on HCIs for optical clocks: Kozlov *et al.*, Rev. Mod. Phy. **90**, 045005 (2018)

Picture credit: Piet Schmidt

HCIs for ultra-precise clocks (Paul traps): present status



No direct laser-cooling transitions:
use sympathetic cooling with Be^+



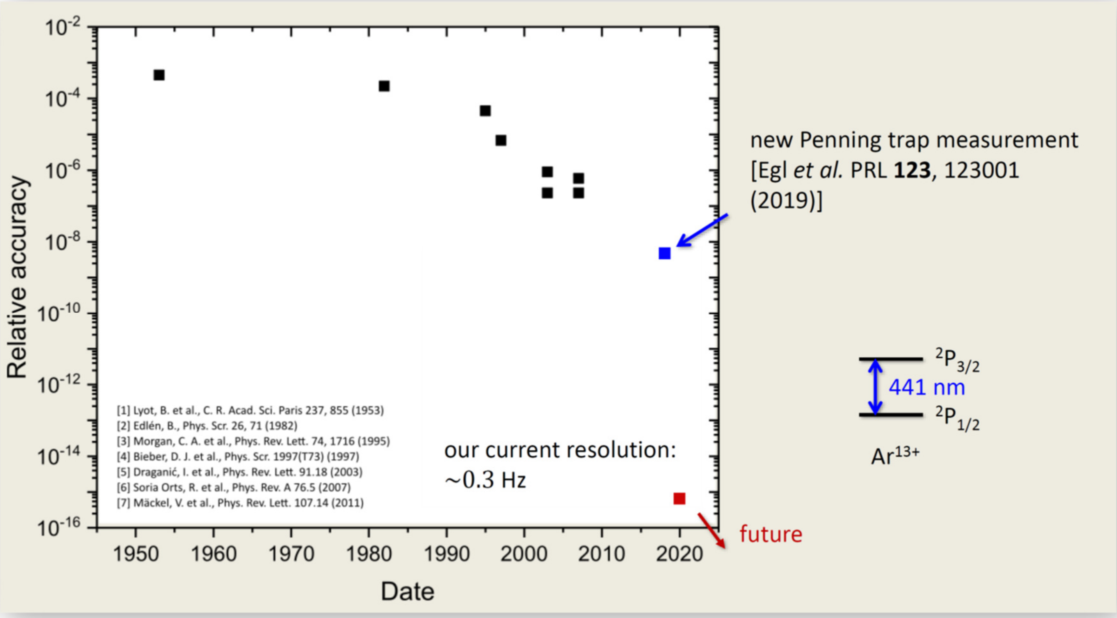
See Dave Hume's talk for more on quantum logic spectroscopy

2015: First sympathetic cooling of HCIs:
L. Schmöger et al., Science 347, 1233 (2015),
Heidelberg

2020: Coherent laser spectroscopy of highly charged ions using quantum logic, P. Micke et al., Nature 578, 60 (2020)

7 orders of magnitude improvement !!!

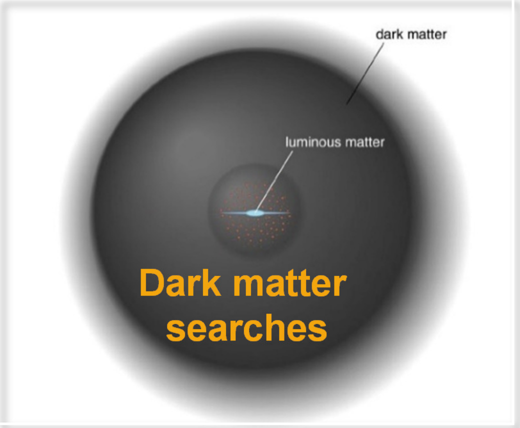
First prototype optical clock, PTB, Germany

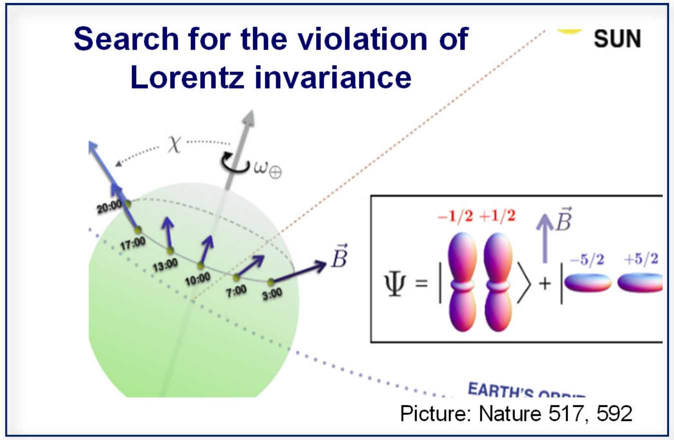


HCIs for ultra-precise clocks : applications & future

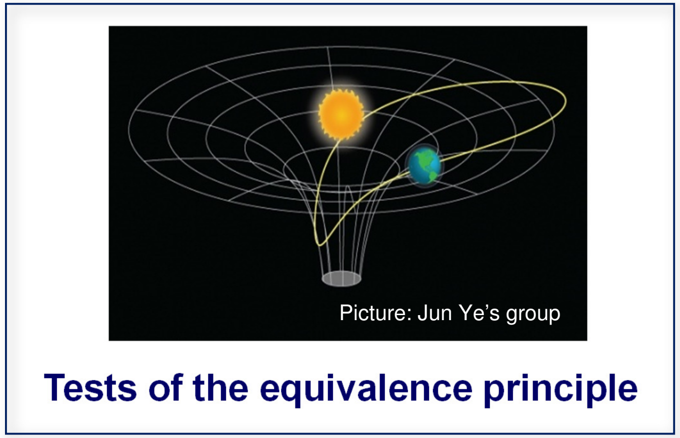
HCIs: **much larger** sensitivity to variation of α and dark matter searches then current clocks

- Enhancement factor K>100, most of present clocks K<1, Yb^+ E3 K=6
- Hyperfine HCI clocks sensitive to m_e/m_p ratio and m_q/Λ_{QCD} ratio variation
- Additional enhancement to Lorentz violation searches



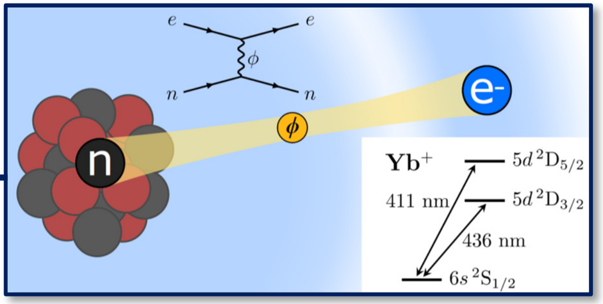


Picture: Nature 517, 592



Tests of the equivalence principle

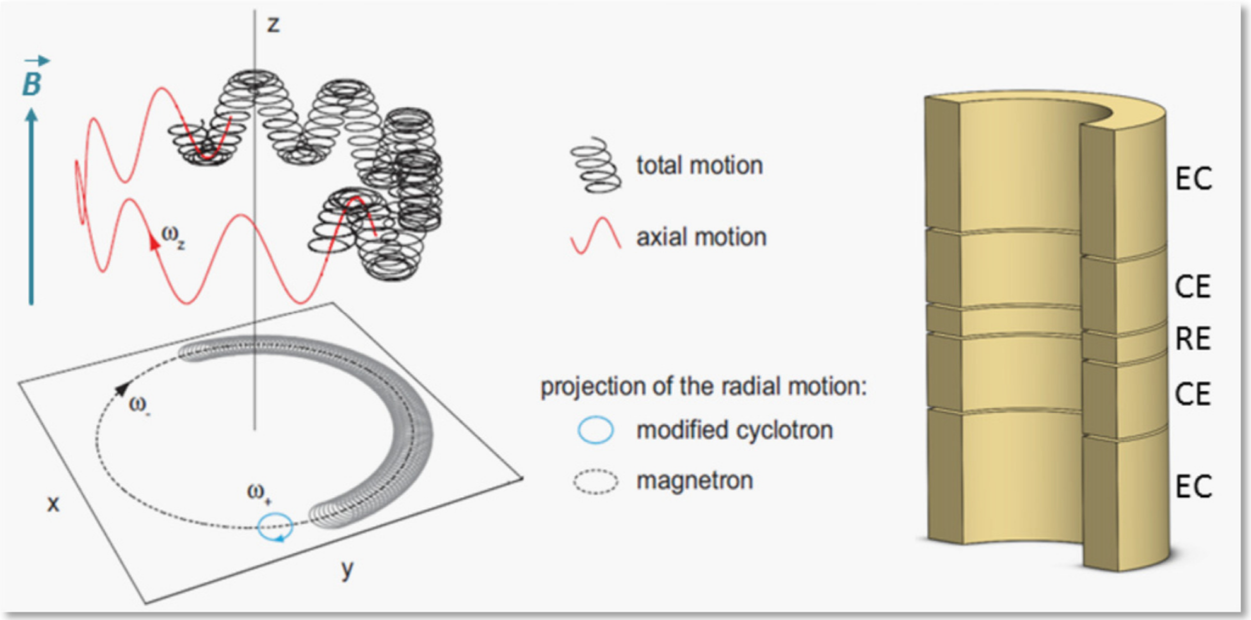
- Searches for the variation of fundamental constants
- Tests of QED: precision spectroscopy
- Fifth force searches: precision measurements of isotope shifts with HCIs to study non-linearity of the King plot



5 years: Optical clocks with selected HCIs will reach 10^{-18} accuracy

10 years: Strongly α-sensitive transitions in HCIs will reach of 10^{-18} uncertainty, multi-ion HCI clocks

HCIs in Penning traps for fundamental physics



Review: K. Blaum *et al.*, *Quantum Sci. Technol.* **6** 014002 (2021)

Advantage of highly charged ions:
the observable, the cyclotron frequency, scales with charge q

Precision magnetic moment measurements of bound electrons

- Stringent test of bound-state QED
- Measurement of fundamental constants

Present status: g-factor differences for different isotopes at the 10^{-13} level

Precision Penning-trap mass spectrometry

- Nuclear masses for test of special relativity
- Nuclear masses for fifth force searches (need actual isotope masses)
- Nuclear masses for neutrino physics studies
- Determination of electron binding energies and test of QED

Present status: $5{\times}10^{-12}$ relative mass precision

R. X. Schüssler et al., Nature 581, 42 (2020)

HCI in Penning traps for fundamental physics: future

Klaus Blaum, MPIK, Heidelberg

Main limitations:

(1) the magnetic field strengths and homogeneities (the cyclotron frequency scales with B)
(2) the stability of the power supplies providing the trapping voltages
(3) the temperature and thus the oscillation amplitude of the single stored ion.

5 years

Limitation (2) can be overcome by at least a factor of 10 by using Josephson junctions for a voltage reference with outstanding stability.
Limitation (3) can be addressed by doing sympathetic laser cooling of the stored particle.

10 years

All exotic species can be sympathetically laser cooled - a real breakthrough in precision Penning trap experiments. The limits for mass and g-factor precision can be improved a factor of 10 – 100.

20 years

Development of stronger magnets with better stability to overcome limitation (1)
Present: 7T magnetic field strengths provided by a NbTi superconducting magnet with relative field stabilities of 1×10^{-10} per hour
Need: magnets up to 20T with temporal and spatial stabilities similar or better to those of NbTi.

Thorium nuclear clocks for fundamental tests of physics

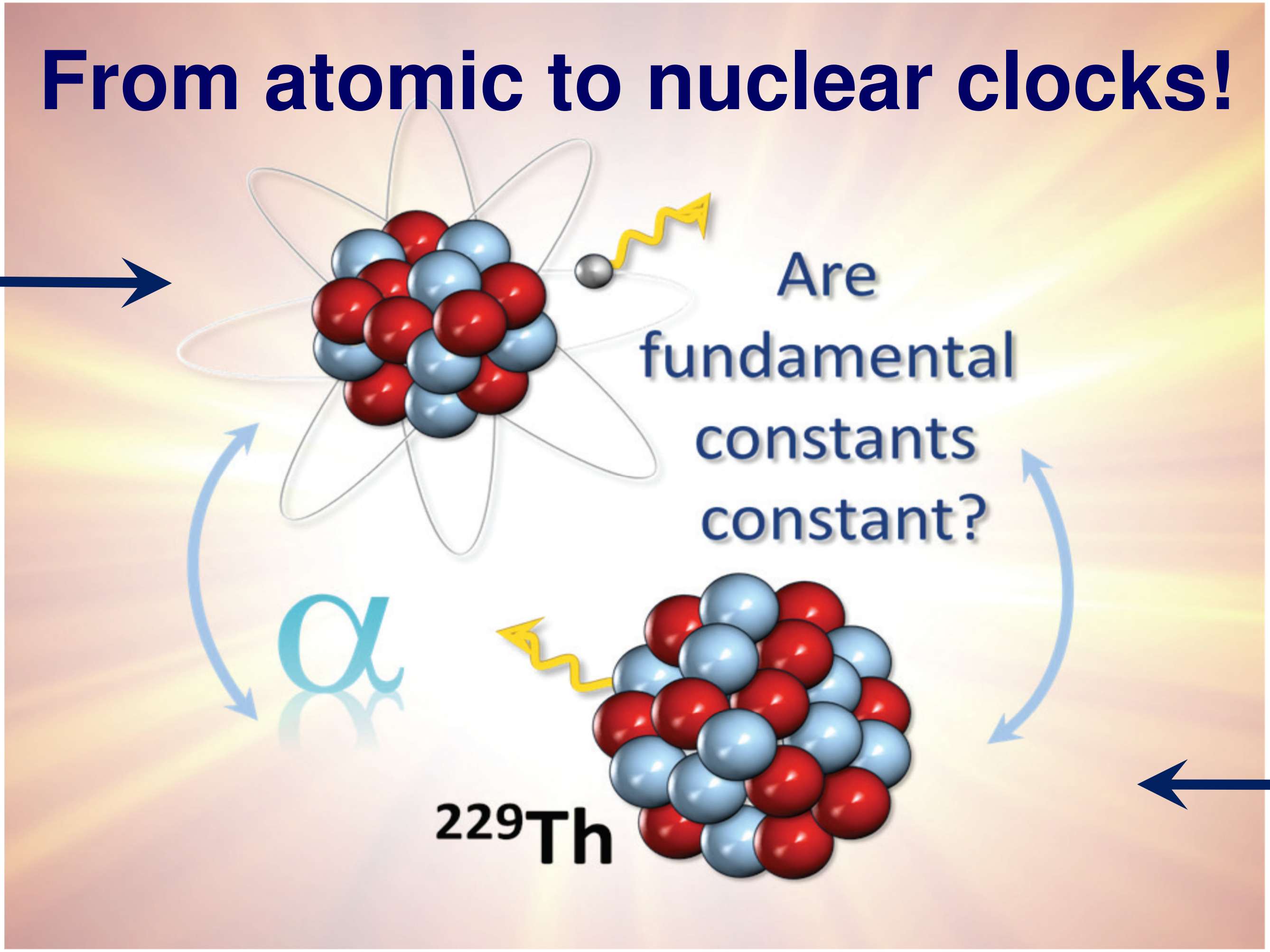
thorium
nuclear clock

https://thoriumclock.eu/

Thorsten Schumm, TU Wein
Ekkehard Peik, PTB
Peter Thirolf, LMU
Marianna Safronova, UDel

erc

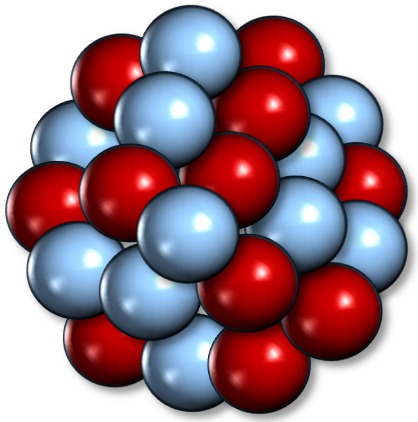
Clock based on transitions in atoms

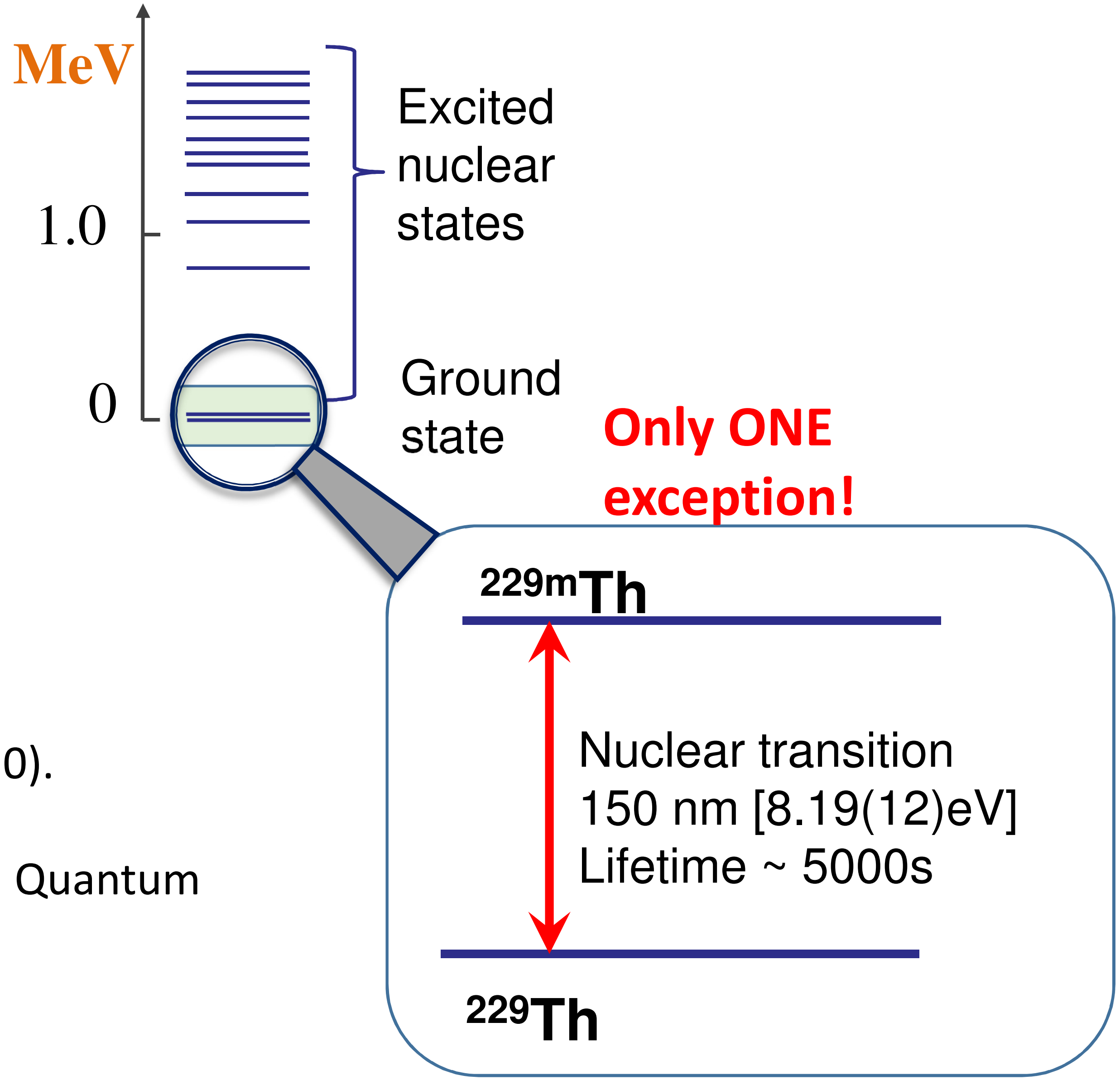


Clock based on transitions in nuclei

Obvious problem: typical nuclear energy levels are in MeV
Six orders of magnitude from ~few eV we can access by lasers!

Atomic
Nucleus

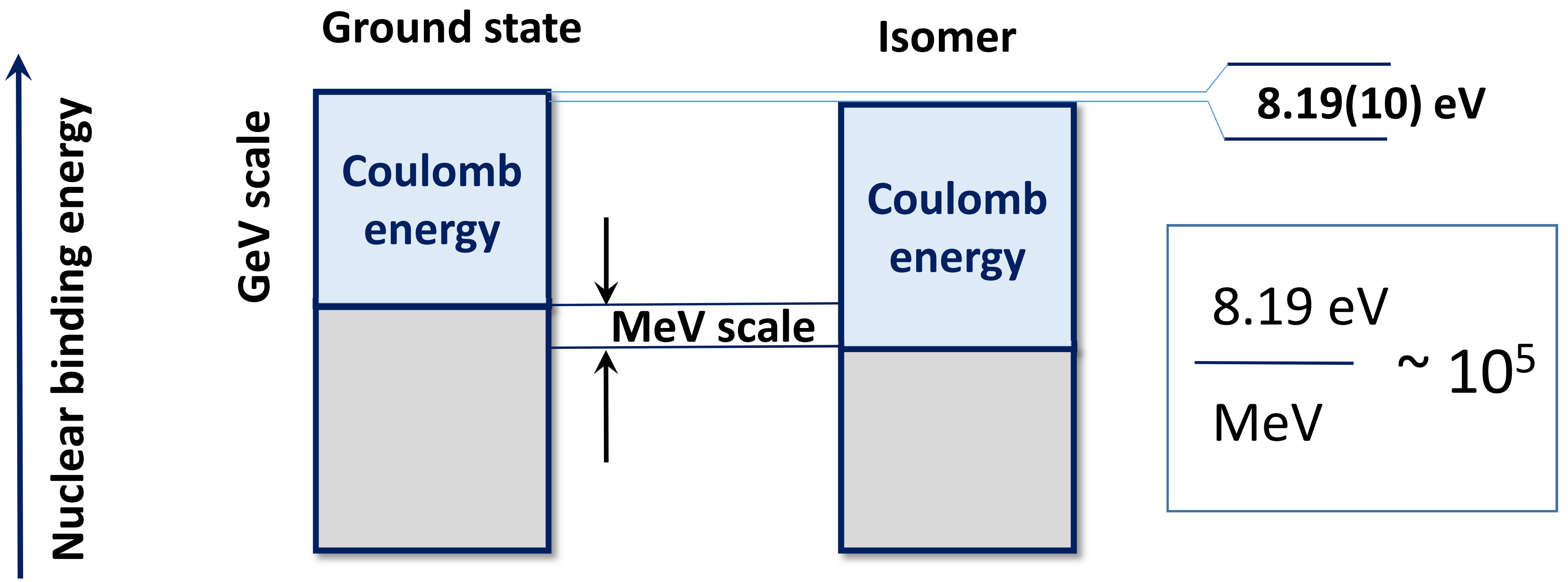




Energy of the ^{229}Th nuclear clock transition:
Seiferle *et al*., Nature 573, 243 (2019)
T. Sikorsky et al., Phys. Rev. Lett. 125, 142503 (2020).

Review: E. Peik, et al., arXiv:2012.09304, in press, Quantum Science and Technology (2021).

Th nuclear clock: Exceptional sensitivity to new physics



Much higher predicted sensitivity (K = 10000-100000) to the variation of α and $\frac{m_q}{\Lambda_{QCD}}$.

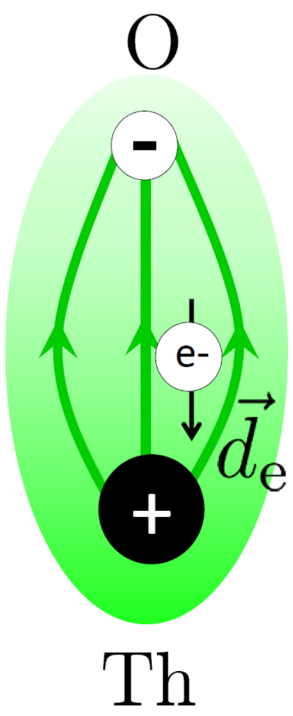
Nuclear clock is sensitive to coupling of dark matter to the nuclear sector of the standard model.

5 years: prototype nuclear clocks, based on both solid state and trapped ion technologies
Measure isomer properties to establish of sensitivity to new physics
Variation of fundamental constant and dark matter searches competitive with present clock

10 years: 10^{-18} – 10^{-19} nuclear clock, 5 - 6 orders improvement in current clock dark matter limits

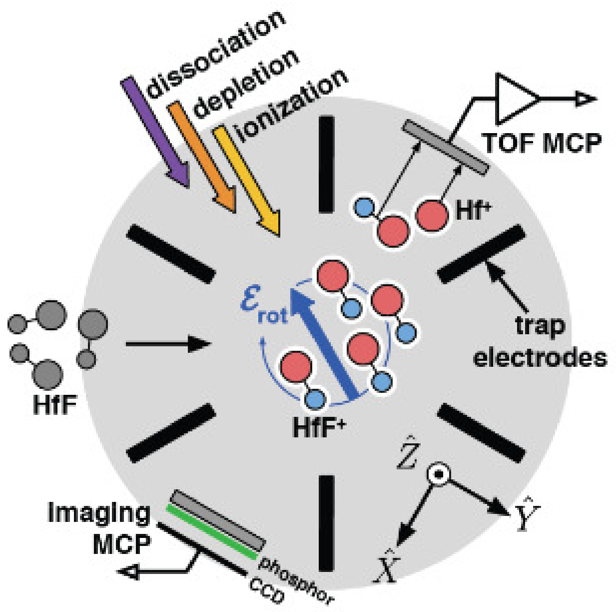
Searches for electron EDM with molecules
Present status: experiments with reported results

Advanced ACME



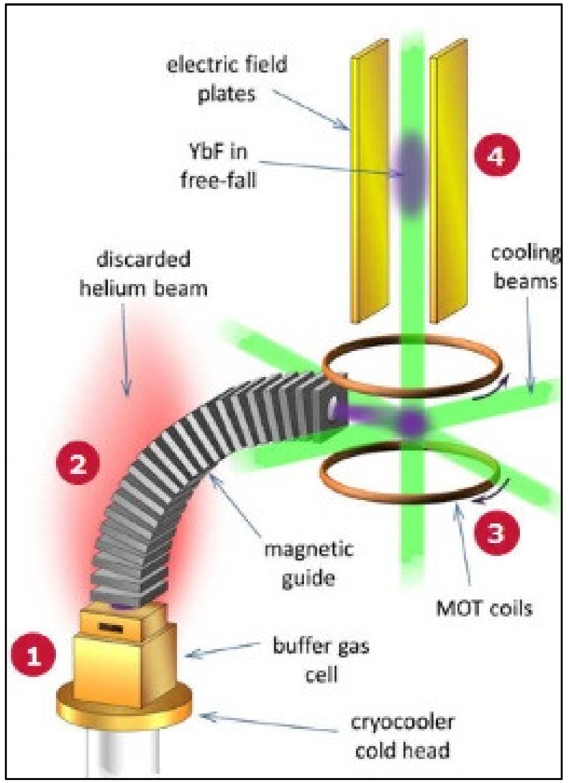
ThO

JILA eEDM



HfF^+, (now also ThF^+)

Imperial College



YbF

Expected an order or magnitude improvement in ~5 years

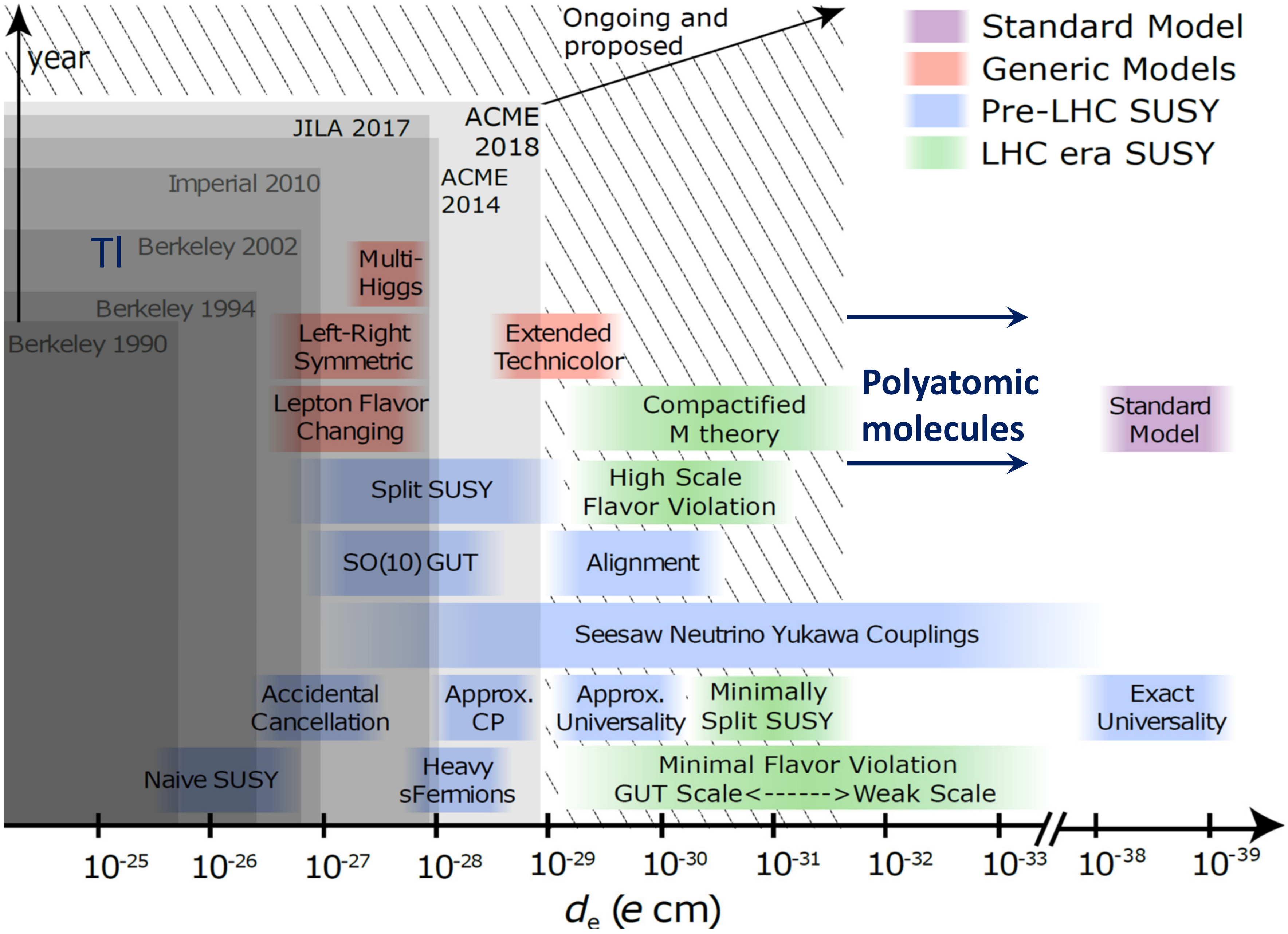
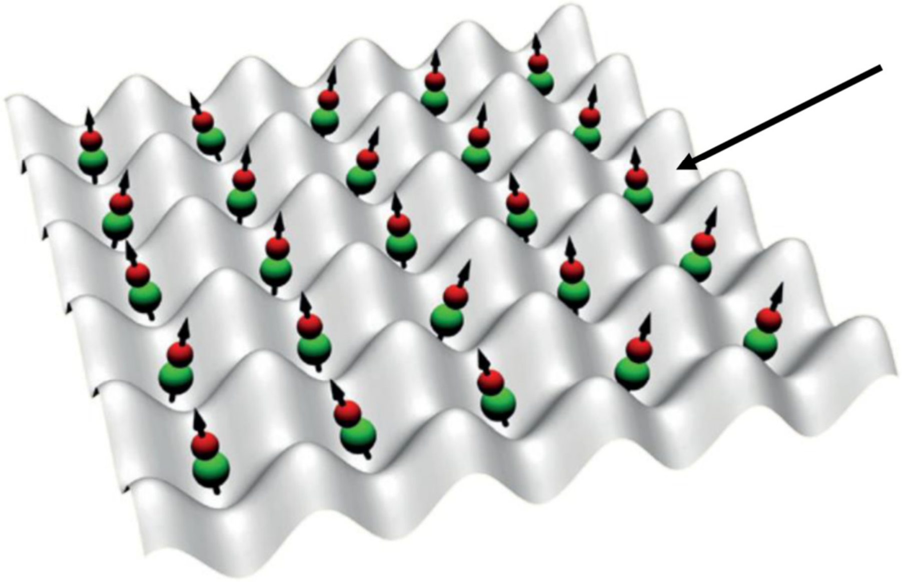


Figure is from 2020 USA AMO Decadal survey (Credit: Dave DeMille)

https://www.nationalacademies.org/amo

Electron EDM experiments: (1) laser-cooled molecules



Heavy, polar molecule sensitive to new physics

Need to trap at ultracold temperatures

Laser slowed, cooled, and trapped in 3D: SrF, CaF, and YO
Laser-cooled, but not yet trapped: YbF, BaH, SrOH, CaOH, YbOH, and $CaOCH_3$

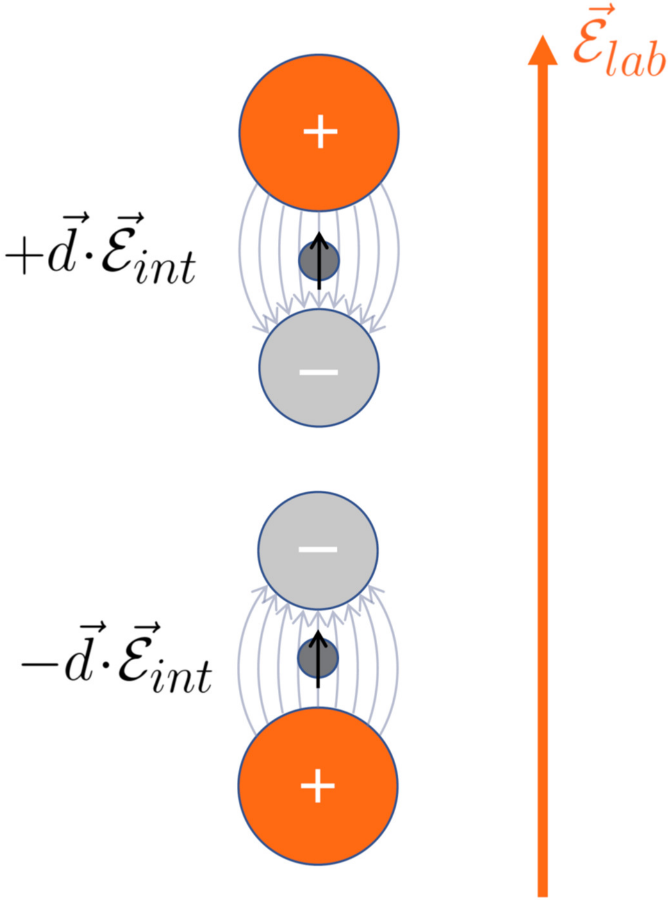
- 10^6 molecules
- 10 s coherence
- Large enhancement(s)
- Robust error rejection
- 1 week averaging

$M_{new\ phys}$ ~ 1,000 TeV

Even before implementing advanced quantum control, such as entanglement-based squeezing

Slide from: Nick Hutzler

Electron EDM experiments: (2) internal co-magnetometer



Need "internal co-magnetometer" states

No need to reverse electric field

ACME and JILA eEDM
ThO HfF$^+$

You can not laser cool any diatomic molecule with co-magnetometer states!

Numerous internal states give rise to many leakage channels out of a cycling transition.

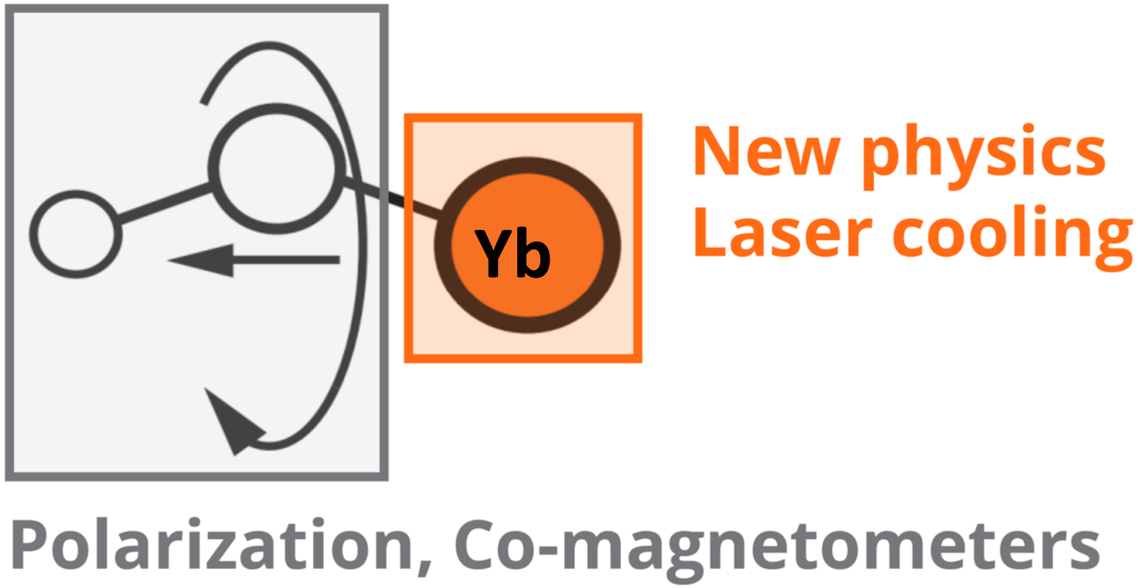
Note: there are other cooling methods besides laser cooling (sympathetic, evaporative, or optoelectrical) and trapped molecular ions enable very sensitive measurements without the need for laser cooling.

Picture from: Nick Hutzler

eEDM experiments with polyatomic laser-cooled

Caltech
Harvard

YbOH



Proposal: Ivan Kozyryev and N. R. Hutzler, Phys. Rev. Lett. **119**, 133002 (2017)
Review: N. R. Hutzler, *Quantum Sci. Technol.* **5** 044011 (2020)

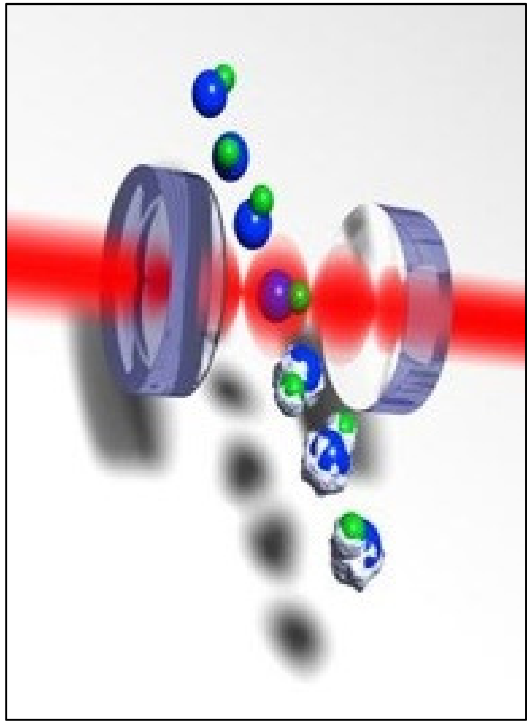
5 years: An electron EDM result with trapped ultracold YbOH, initial goal 10^{-31} e cm
8 years: Improvements in coherence time and number trapped molecules: 10^{-32} e cm
12 years: Very large numbers of trapped molecules or many operating in parallel, 10^{-33} e cm
Further improvement with squeezing?

Picture & timeline from: Nick Hutzler

Hadronic T-violation searches with molecules

CP-violation in the nucleus: manifest as a nuclear Schiff moment (NSM) or nuclear magnetic quadrupole moment (MQM). Arises from nucleon EDMs, new CP-violating nuclear forces, strong force CP-violation (θ).

CeNTREX: see arXiv:2010.01451



TlF (proton EDM)

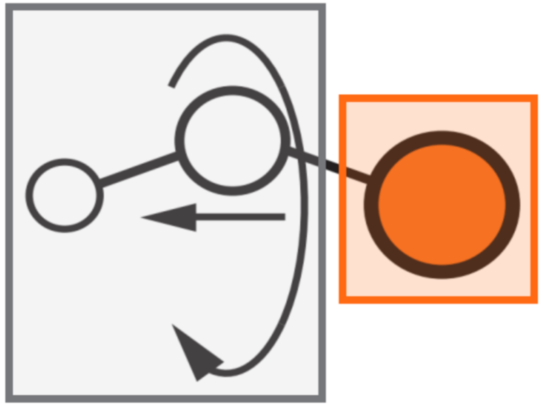
The observable signature of a Schiff moment will be a shift in the NMR frequency of ^{205}Tl nuclei when the molecules are polarized by a strong electric field.

First generation: a cryogenic molecular beam of TlF

Second generation: laser cool and trap the TlF molecules for increased sensitivity.

YbOH nuclear MQM

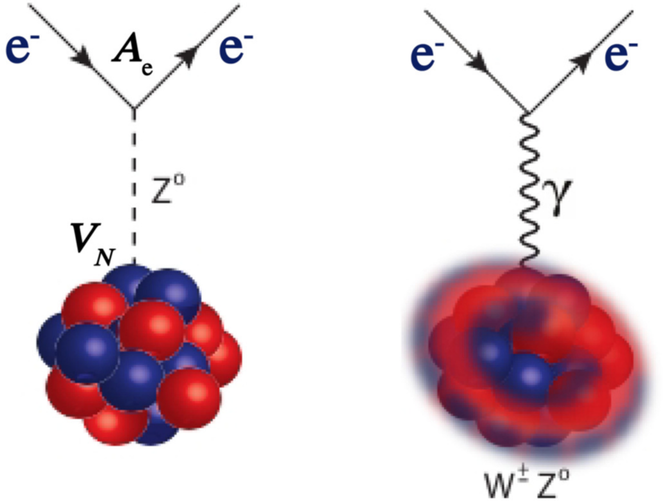
Theory: J. Chem. Phys. 152, 084303 (2020)



3 years:
beam-based measurements

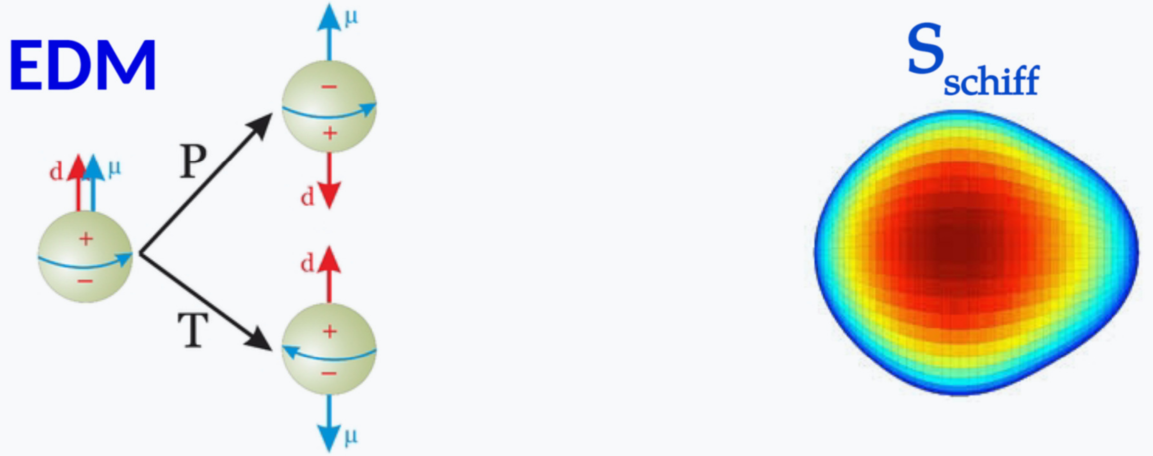
Fundamental symmetries: radioactive atoms and molecules

Parity violation



$\sim Z^3$ $\sim Z^2 A^{2/3} R(Z)$

T-violation



$\sim Z^2 R(Z)$ RaF

$S \sim Q_2 Q_3 Z A^{2/3} / (E_+ - E_-)$

ZOMBIES (Yale, BaF)
Yb (Mainz)

Fr (TRIUMF, Tokyo)
Ra^+ (UCSB)

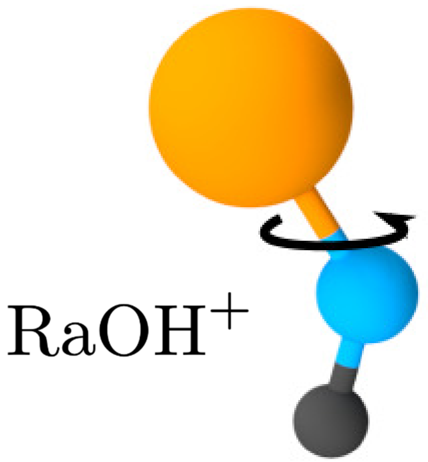
Ra and Ra-based molecules have a further enhancement due to an octupole deformation of the ^{225}Ra nucleus: an intrinsic Schiff moment 1000 times larger than in spherical nuclei such as Hg.

Collinear resonance ionization spectroscopy of RaF molecules
[Garcia Ruiz, Berger et al. CERN-INTC-2018-017 (2018)]

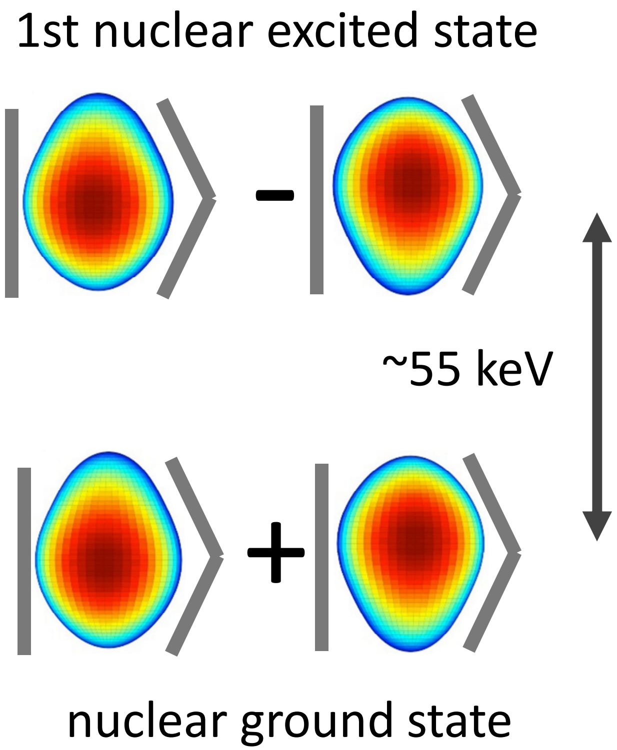
Picture credits: Ronald Fernando Garcia Ruiz

T-violation with radioactive molecular ions

Theory: nuclear Shiff moments sensitivity investigated for RaOH, RaOH+, $ThOH^+$, and $RaOCH_3^+$



$RaOH^+$ and $RaOCH_3^+$ having been recently created and cooled in an ion trap [UCSB, Fan et al., PRL 126, 023002 (2021)].



Octupole deformation ^{225}Ra

T- violating forces mix the states

Enhancement: ~1000x

from Andrew Jayich

Other candidate: ^{229}Pa, the splitting is 50(60) eV - we don't know if the state exists.

^{229}Pa may be 100,000 times more sensitive than ^{199}Hg.

Currently no significant source of ^{229}Pa (1.5 day half-life). Plans to harvest at the Facility for Rare Isotope Beams at Michigan State University.

J. T. Singh, Hyperfine Int. 240, 29 (2019)

T-violation with radioactive atoms and molecules: timeline

From Andrew Jayich and Ronald Fernando Garcia Ruiz

1-2 years : Laser cooling of $^{225}Ra^+$, measurements of Ra+ properties, Ac^{2+} with radium quantum logic
Investigate RaOH, RaOH+, other polyatomic molecules containing Ra, ^{229}ThO, $^{229}ThF^+$ at ISOLDE
RaF: Precision experiments: rotational and hyperfine structure

3 years: First quantum logic spectroscopy (QLS) of radioactive molecules using Ra+ as the logic qubit
QLS of radium-based triatomic molecules: RaOH+, RaSH+, RaCN+
RaF: Deacceleration and trapping

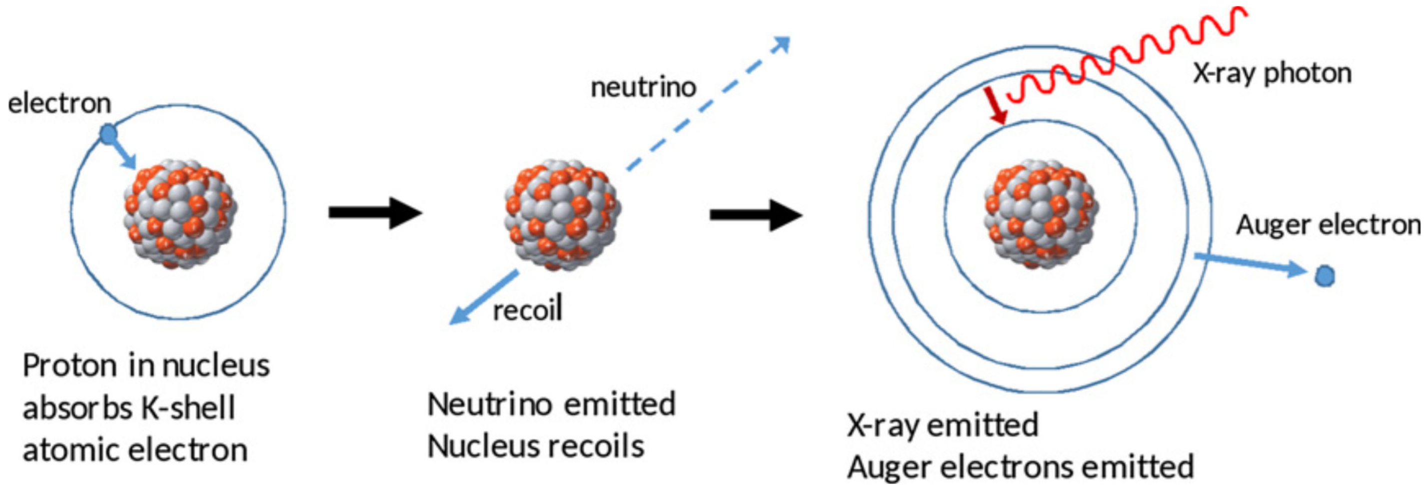
5 years: First QLS-based single molecular ion EDM measurement with Ra
Measure the nuclear energy level structure of ^{229}Pa
RaF: first laser cooling tests and systematic studies, symmetry-violating measurements

8 years: New θ_{QCD} bounds

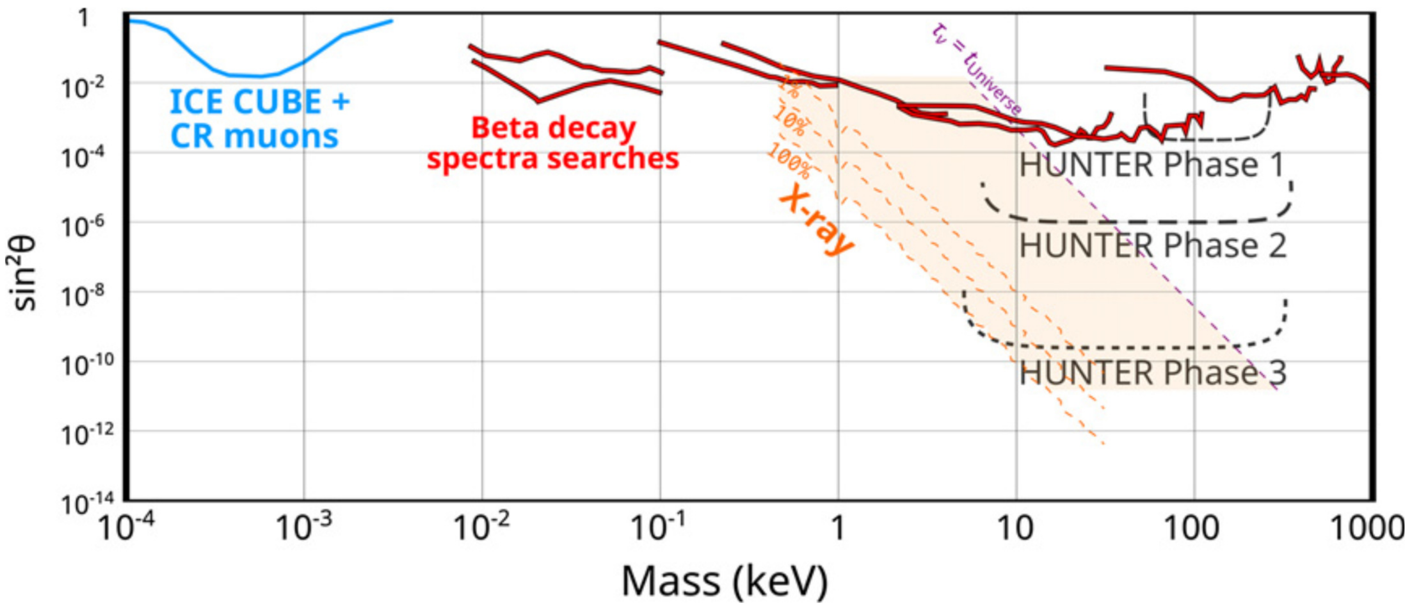
10-15 years: If ^{229}Pa enhancement as predicted new θ_{QCD} bounds pushing the low few 10^{-13} range

HUNTER: precision massive-neutrino search based on a laser cooled atomic source

$$^{131}\mathrm{Cs} \rightarrow {}^{131}\mathrm{Xe}^{*} + \nu_e$$



Cs atoms are trapped in a MOT. Complete kinematical reconstruction is possible, allowing the neutrino mass to be determined event-by-event.



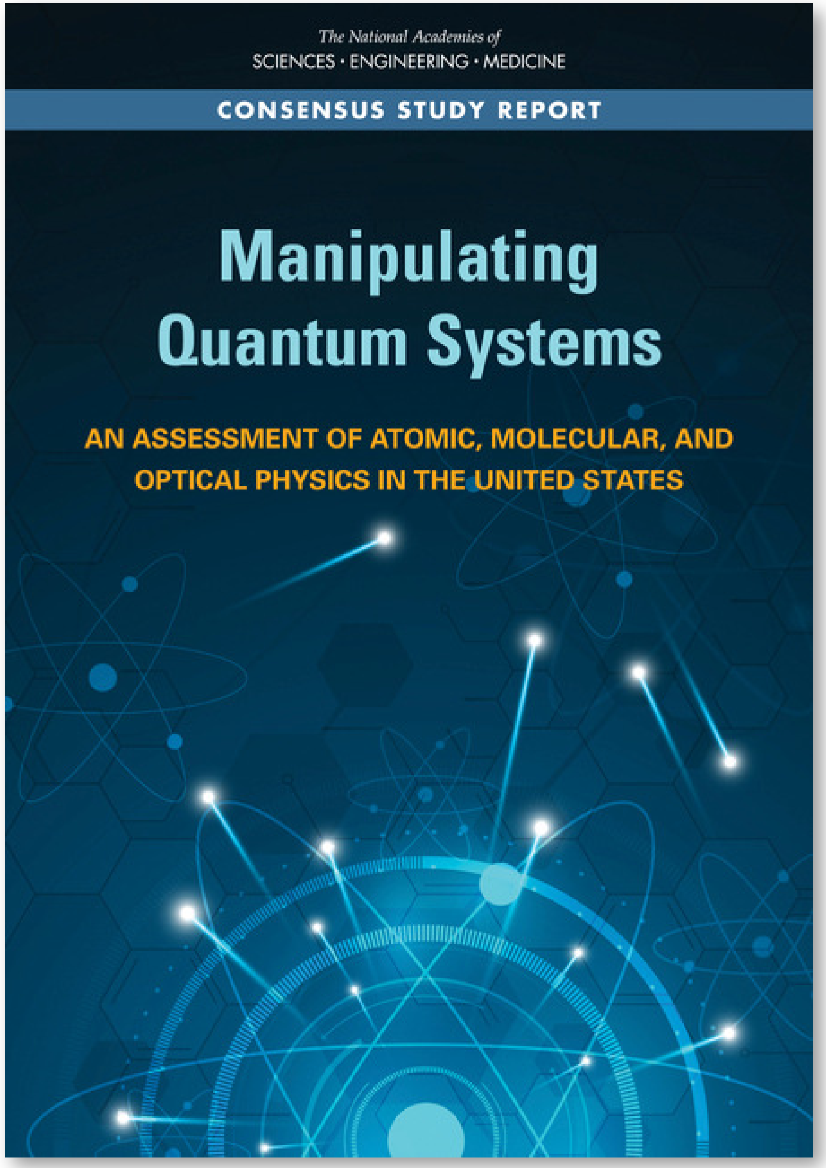
Limits on sterile neutrino coupling strength vs mass. Dashed lines (orange) show astrophysical limits permitting sterile neutrinos to be the galactic dark matter

From: C. J. Martoff *et al., Quantum Sci. Technol.* **6** 024008 (2021)

Novel ionic, atomic and molecular systems: other topics

- Need extensive atomic, molecular and nuclear theory development – many properties are unknown. Theory will aid to select best candidates, develop laser-cooling schemes, reduce systematics, explain anomalies. Need large-scale high-performance computing, new ides with AI.
- Need to set up theory cyberinfrastructure: data and code portals (atomic portal version 1 to be released at UD).
- Need much stronger ties of AMO and particle physics communities – new ideas for uses of QT for fundamental physics.
- Development of VUV, XUV, and soft X- ray frequency combs and laser to drive them for HCI spectroscopy. Included in the timeline.
- Molecular and molecular ions clocks – searches for the variation of the m_e/m_p ratio and corresponding dark matter searches. KRb, Sr_2, N_2^+, O_2^+, H_2^+, HD^+, TeH^+, SrOH, and others, including those proposed for T-violation studies. See David Hanneke et al., Quantum Sci. Technol. 6, 014005 (2021). Included in the timeline.
- Lorentz violation tests and proposals with trapped ions [Nature 567, 204 (2019), Phys. Rev. Lett. 120, 103202 (2018).
- Precision spectroscopy experiments for dark matter searches (King plot nonlinearities – need radioactive isotopes and theory for standard model signals). Phys. Rev. Lett. 125, 123002 (2020) and others [D. Antypas et al., Quantum Sci. Technol. 6 034001 (2021)].
- Muonic hydrogen, proton radius puzzle and related hydrogen spectroscopy experiments.

2020 USA Decadal Assessment and Outlook Report on AMO Science and other recourses



PDF and html versions are available (free) online:
https://www.nationalacademies.org/amo

Chapter 6
PRECISION FRONTIER AND FUNDAMENTAL NATURE OF THE UNIVERSE

Recent review:
Search for new physics with atoms and molecules, M. S. Safronova, D. Budker, D. DeMille, Derek F. Jackson-Kimball, A. Derevianko, and Charles W. Clark, Rev. Mod. Phys. 90, 025008 (2018). **106 pages, over 1100 references**

Forthcoming Focus Issue in Quantum Science and Technology
Quantum Sensors for New-Physics Discoveries
Editors: Marianna Safronova and Dmitry Budker

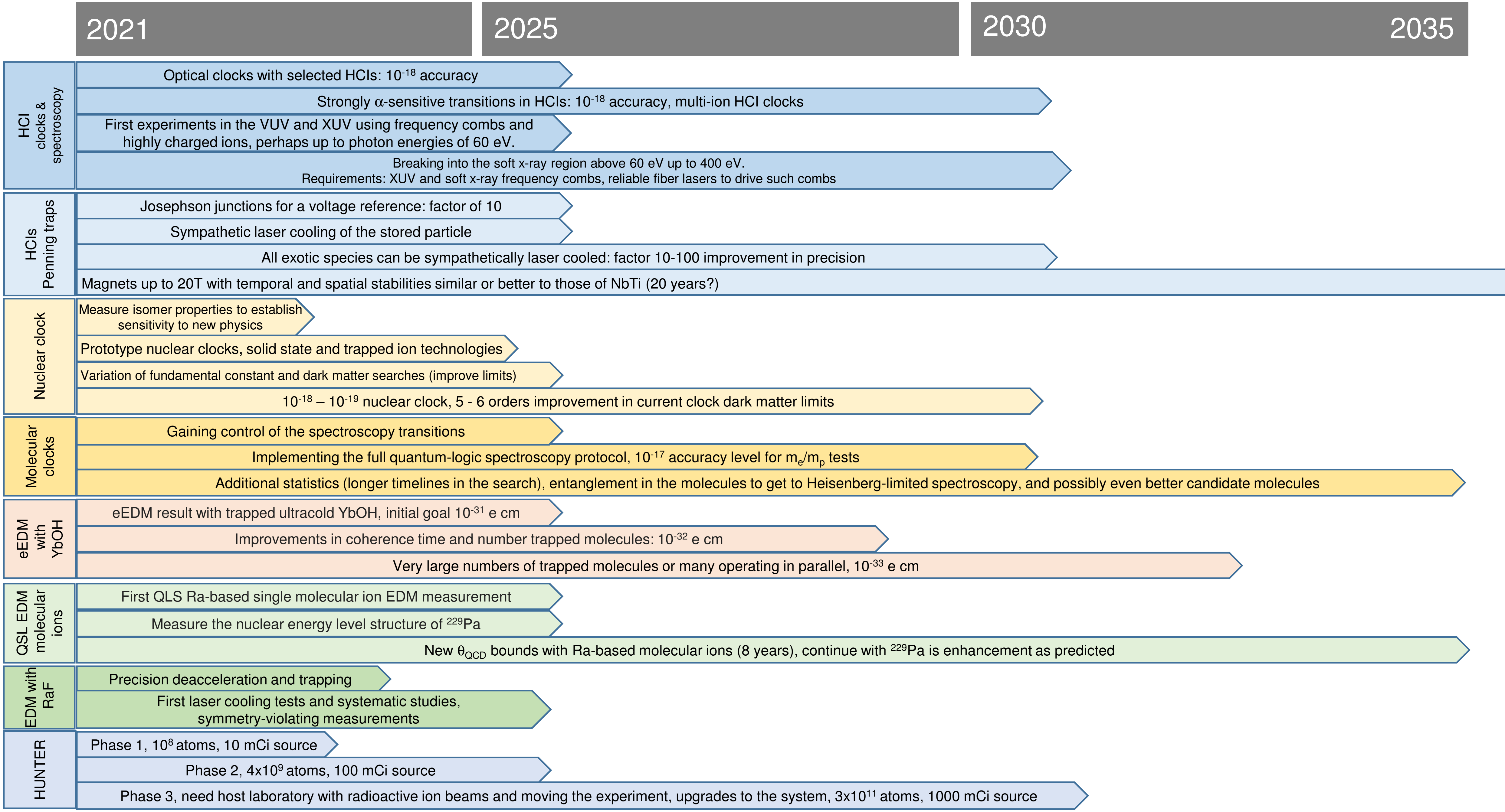
+18 articles will appear focusing on the future decade
https://iopscience.iop.org/journal/2058-9565/page/Focus-issues

Difficulty of preparing 20+ timeline: these technologies did not exist 20 years ago!

AMO experiments are funded in a 3-year (rarely 5-year) cycle

Very rapid development

- 2014 - 2015: First proposals to search for dark matter with clocks
- 2015 First ultracold HCIs
- 2017: Proposal to use polyatomic molecules for EDMs, laser cooling already demonstrated
- 2020: First quantum logic spectroscopy with HCIs
- 2021: $RaOH^+$ and $RaOCH_3^+$ created and cooled in an ion trap


2021
2025
2030
2035
HCI clocks & spectroscopy
Optical clocks with selected HCIs: 10^{-18} accuracy
Strongly α-sensitive transitions in HCIs: 10^{-18} accuracy, multi-ion HCI clocks
First experiments in the VUV and XUV using frequency combs and highly charged ions, perhaps up to photon energies of 60 eV.
Breaking into the soft x-ray region above 60 eV up to 400 eV. Requirements: XUV and soft x-ray frequency combs, reliable fiber lasers to drive such combs
HCIs Penning traps
Josephson junctions for a voltage reference: factor of 10
Sympathetic laser cooling of the stored particle
All exotic species can be sympathetically laser cooled: factor 10-100 improvement in precision
Magnets up to 20T with temporal and spatial stabilities similar or better to those of NbTi (20 years?)
Nuclear clock
Measure isomer properties to establish sensitivity to new physics
Prototype nuclear clocks, solid state and trapped ion technologies
Variation of fundamental constant and dark matter searches (improve limits)
10^{-18} – 10^{-19} nuclear clock, 5 - 6 orders improvement in current clock dark matter limits
Molecular clocks
Gaining control of the spectroscopy transitions
Implementing the full quantum-logic spectroscopy protocol, 10^{-17} accuracy level for m_e/m_p tests
Additional statistics (longer timelines in the search), entanglement in the molecules to get to Heisenberg-limited spectroscopy, and possibly even better candidate molecules
eEDM with YbOH
eEDM result with trapped ultracold YbOH, initial goal 10^{-31} e cm
Improvements in coherence time and number trapped molecules: 10^{-32} e cm
Very large numbers of trapped molecules or many operating in parallel, 10^{-33} e cm
QSL EDM molecular ions
First QLS Ra-based single molecular ion EDM measurement
Measure the nuclear energy level structure of ^{229}Pa
New θ_{QCD} bounds with Ra-based molecular ions (8 years), continue with ^{229}Pa is enhancement as predicted
EDM with RaF
Precision deacceleration and trapping
First laser cooling tests and systematic studies, symmetry-violating measurements
HUNTER
Phase 1, 10^8 atoms, 10 mCi source
Phase 2, $4x10^9$ atoms, 100 mCi source
Phase 3, need host laboratory with radioactive ion beams and moving the experiment, upgrades to the system, $3x10^{11}$ atoms, 1000 mCi source