KU LEUVEN

Fundamental Physics studies at ISOLDE-CERN and PSI

Gerda Neyens

On behalf of the KU Leuven scientists active at ISOLDE and PSI

KU LEUVEN fwo EOS THE EXCELLENCE OF SCIENCE

ESPP European Strategy for Particle Physics meeting,
February 5, 2025 Antwerp, Belgium

Precision measurements to test the Standard Model

now the most promising route towards discovering New Physics

Precision spectroscopy of heavy (thus radioactive) atoms and molecules can provide stringent constraints on CP violation via searches for permanent electric dipole moments and other CP-odd properties in leptons, hadrons, and nuclei. Flambaum and Dzuba, PRA101, 042504 (2020)

Ongoing projects since decades:

- Searches for a neutron Electric Dipole Moment (nEDM)
- Searches for an electron EDM (eEDM)
- Searches for a nuclear Shiff Moment (nuclear EDM)

New breakthroughs and opportunities:

- x1000 enhanced sensitivity to eEDM and Shiff moment using **radioactive molecules** !
- The **^{229m}Th nuclear clock** to test fundamental constants (are they constant?)
- Search for a **5th force** using isotope shift measurements on **radioactive atoms and ions**

nEDM and n2EDM collaborations at PSI

(KU Leuven member since 2007)

Second generation neutron EDM experiment with experience of previously successful measurement

PSI: $d_n < 1.8 \times 10^{-26} e\ cm\ (90\%\ C.L.)$

Standard Model expectation: $10^{-32} e\ cm$

KU Leuven PhD thesis Elise Wursten, 2021

Ultra cold neutron source and n2EDM

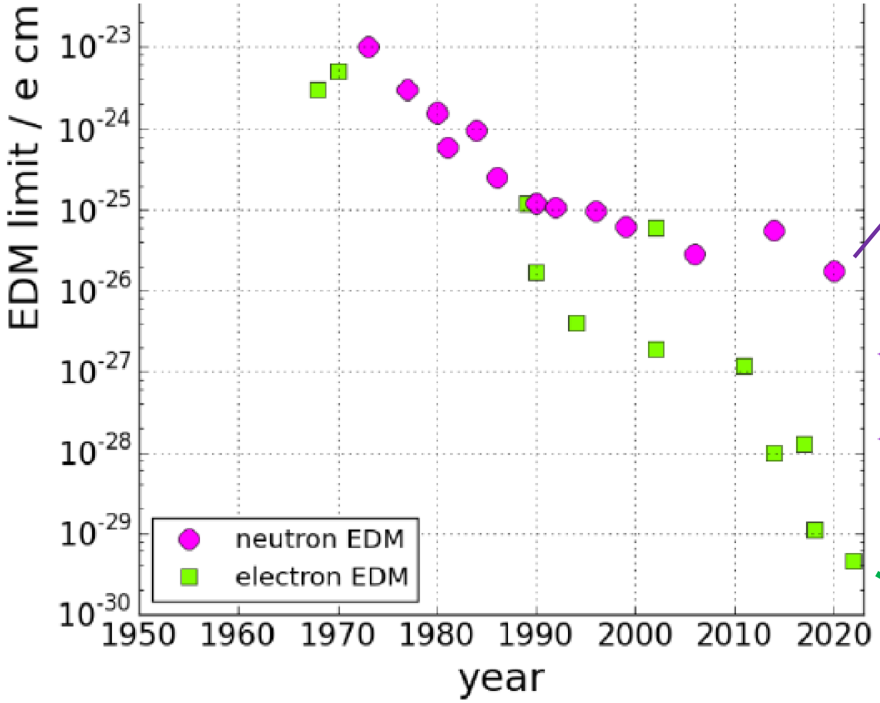
Phase 1 n2EDM target sensitivity $d_n \approx 10^{-27} e\ cm$

- ***Improved neutron statistics:*** *larger neutron storage volume, improved source, etc.*
- ***Improved systematics:*** *Better magnetometry and magnetic field control*

[PSI nEDM limit] doi:10.1103/PhysRevLett.124.081803 (2020)

Evolution of LIMITS on nEDM and eEDM compared to their Standard Model values

Measurements started in the 1950s (nEDM) and 1960s (eEDM)



Current best limit: nEDM experiment at PSI (PRL 124 (2020) 081803) ~ 10^{-26}

Design sensitivity of 4 new experiments:

n2EDM@PSI + panEDM@ILL + LANL + TUCAN@TRIUMF

Design sensitivity cryogenic nEDM@SNS

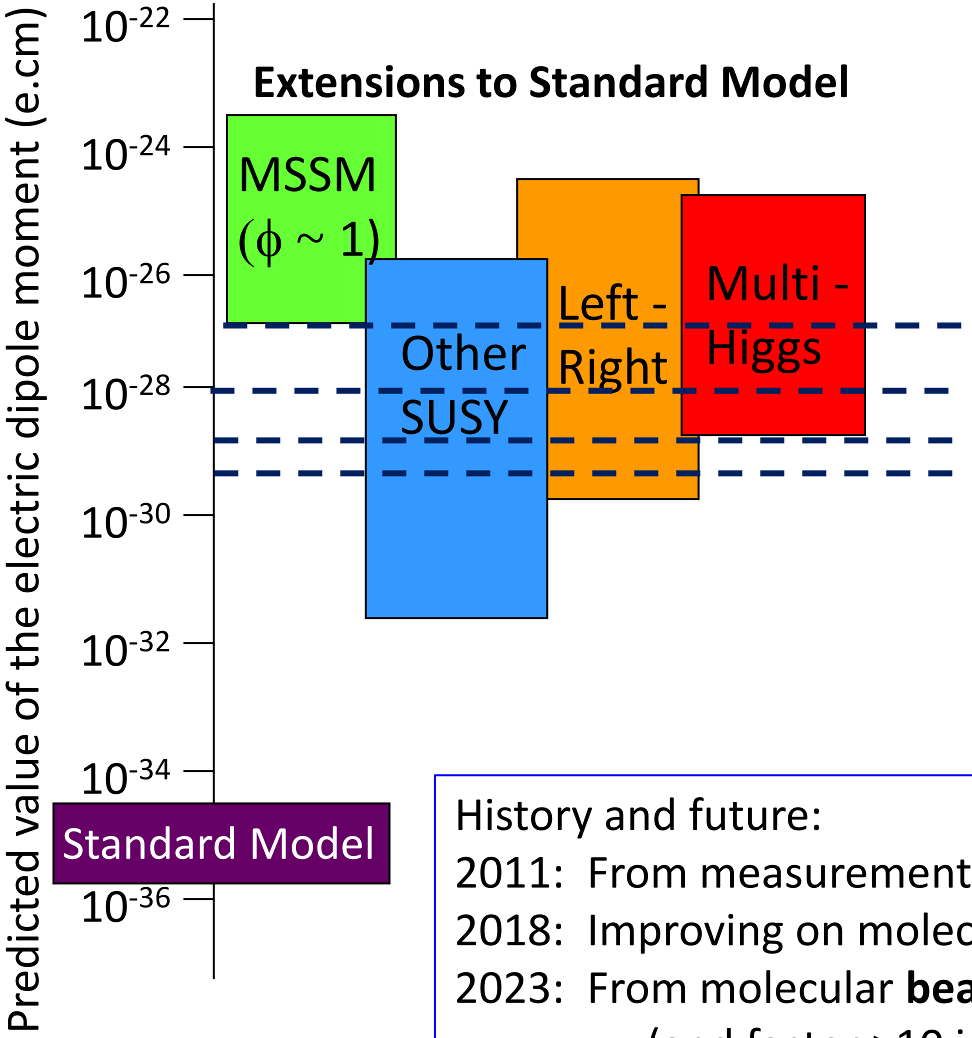
Current best limit eEDM: trapped HfF^{+} ionic molecules ~ 4.10^{-30} (Science 381 (2023) 84)

10^{-32} STANDARD MODEL nEDM

10^{-35} STANDARD MODEL eEDM

Phys. Rev. Lett. **129**, 231801(2022)

Electron EDM predictions – and experiments



Best eEDM limits come from molecules !

YbF beam (Imperial, 2011, [1]):| d_e | < 1.0×10^{-27} e.cm

ThO beam (ACME, 2014, [2]): | d_e | < 9.4×10^{-29} e.cm

ThO beam (ACME, 2018, [3]): | d_e | < 1.1×10^{-29} e.cm

Trapped HfF^+ (JILA, 2023, [4]): | d_e | < 4.1×10^{-30} e.cm

Sensitive to new particles with masses up to 30 TeV

[1] Nature 473, 493 (2011)
[2] Science 343, 269 (2014)
[3] Nature 562, 355 (2018)
[4] Science 381, 46 (2023)

History and future:

2011: From measurements on **atoms to molecules**: gain factor 100

2018: Improving on molecular beam experiments: gain another factor 10-100

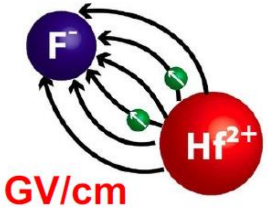
2023: From molecular **beams to trapped** molecules (quantum control): gain factor of 2 (and factor >10 in next decade expected)

Next step: From **stable to radioactive molecules**: gain factor >1000 in next 2 decades
➔ reaching PeV scales sensitivity for new particles

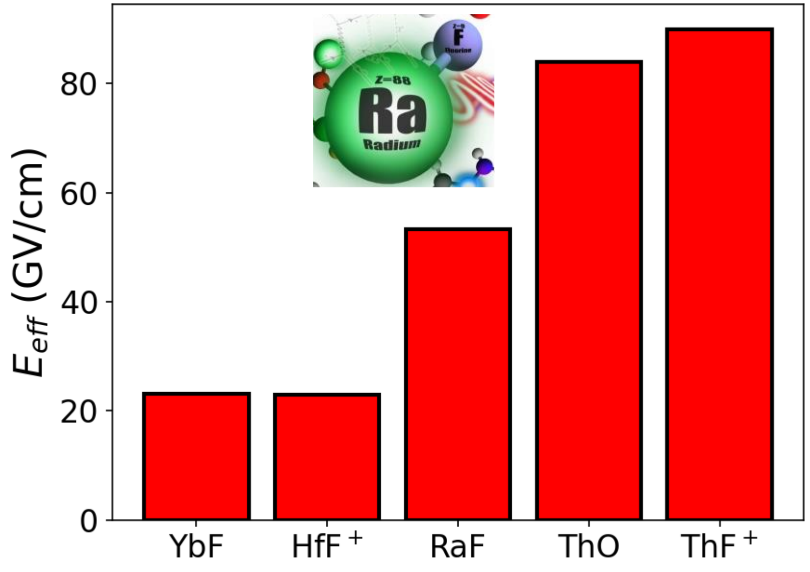
Modified from M. Tarbutt, Imperial College London, 2024 ECT* workshop on EDM studies

Why radioactive molecules ?

- Sensitivity to eEDM is enhanced in molecules due to the large electric field (E_{eff}) in polar molecules

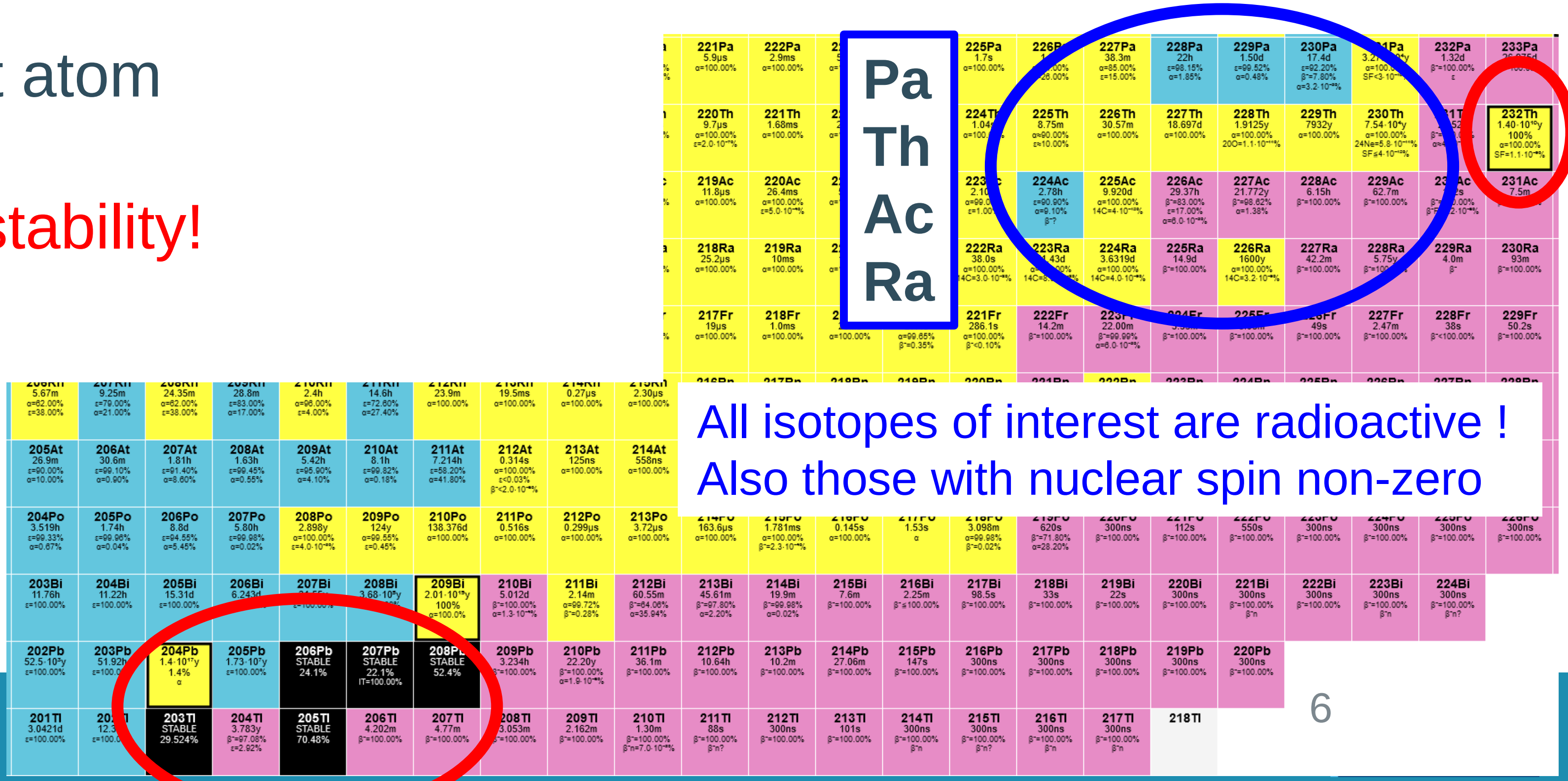


E_{eff}~20 GV/cm



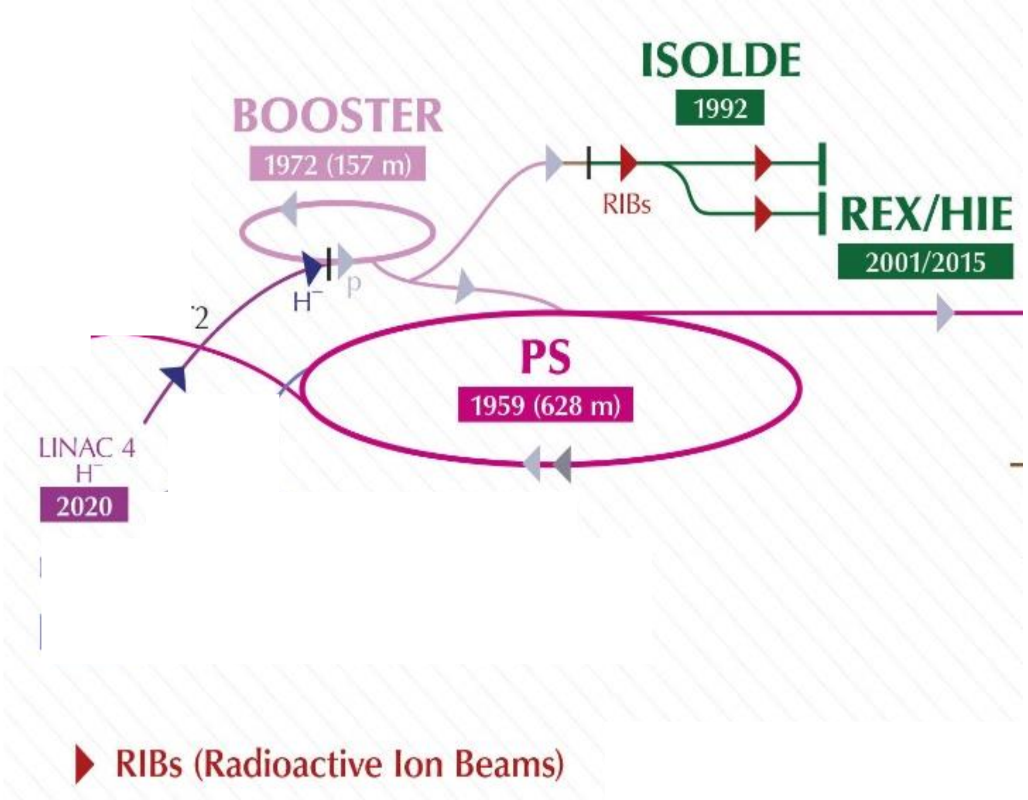
➔ $E_{\rm eff}$ scales with Z^3 of heaviest atom

BUT: there is a limit to nuclear stability!



All isotopes of interest are radioactive !
Also those with nuclear spin non-zero

ISOLDE = CERN's Radioactive Ion Beam (RIB) facility



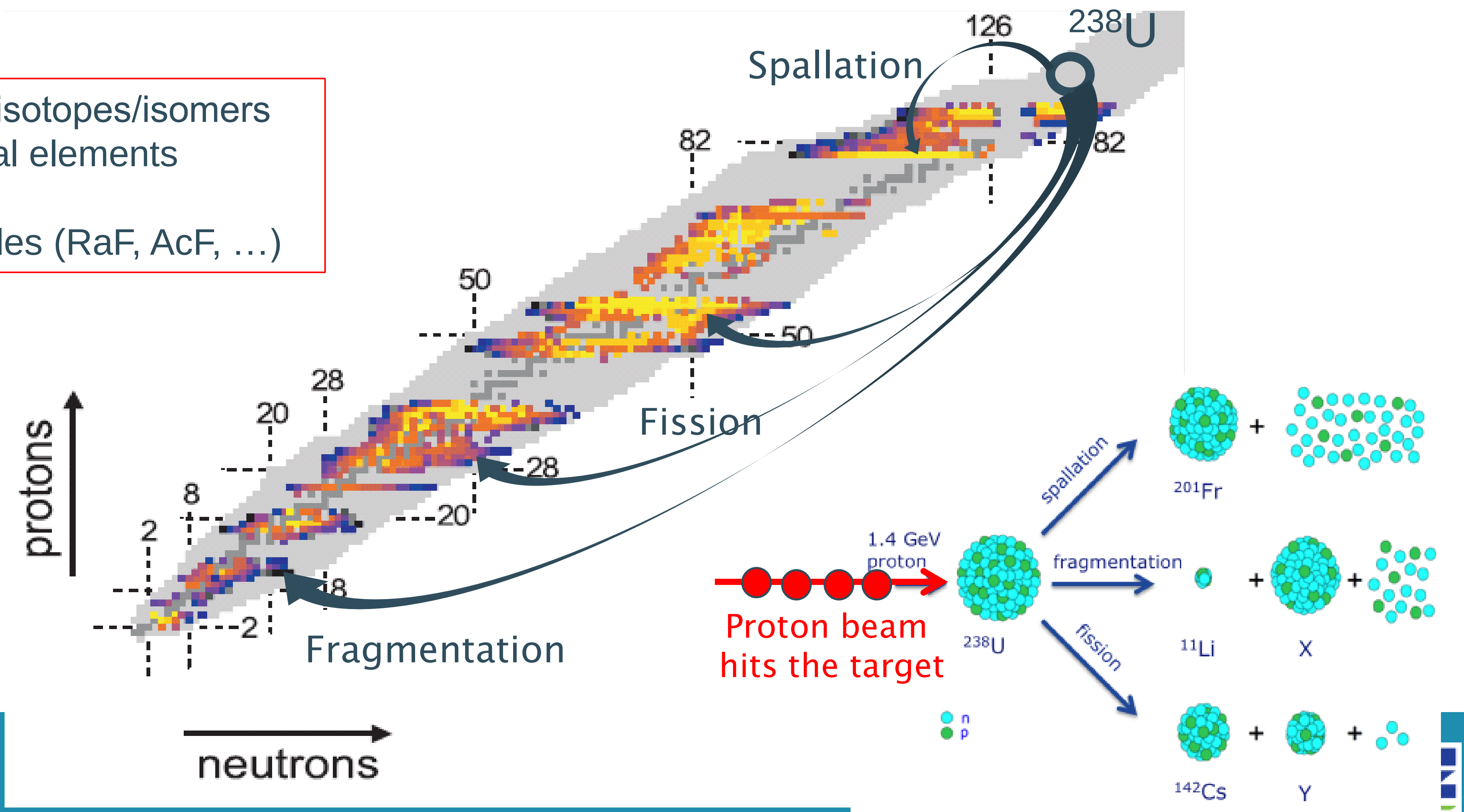
- >50% of Booster protons are sent to ISOLDE
- E_p = 1.4 GeV

ISOLDE = Isotope **S**eparation **O**n-**L**ine **D**evice
→ ISOL is a method to produce and select **short-lived radioactive isotopes** ($t_{1/2}$ down to tens of milliseconds)

Production of RIB's at ISOLDE

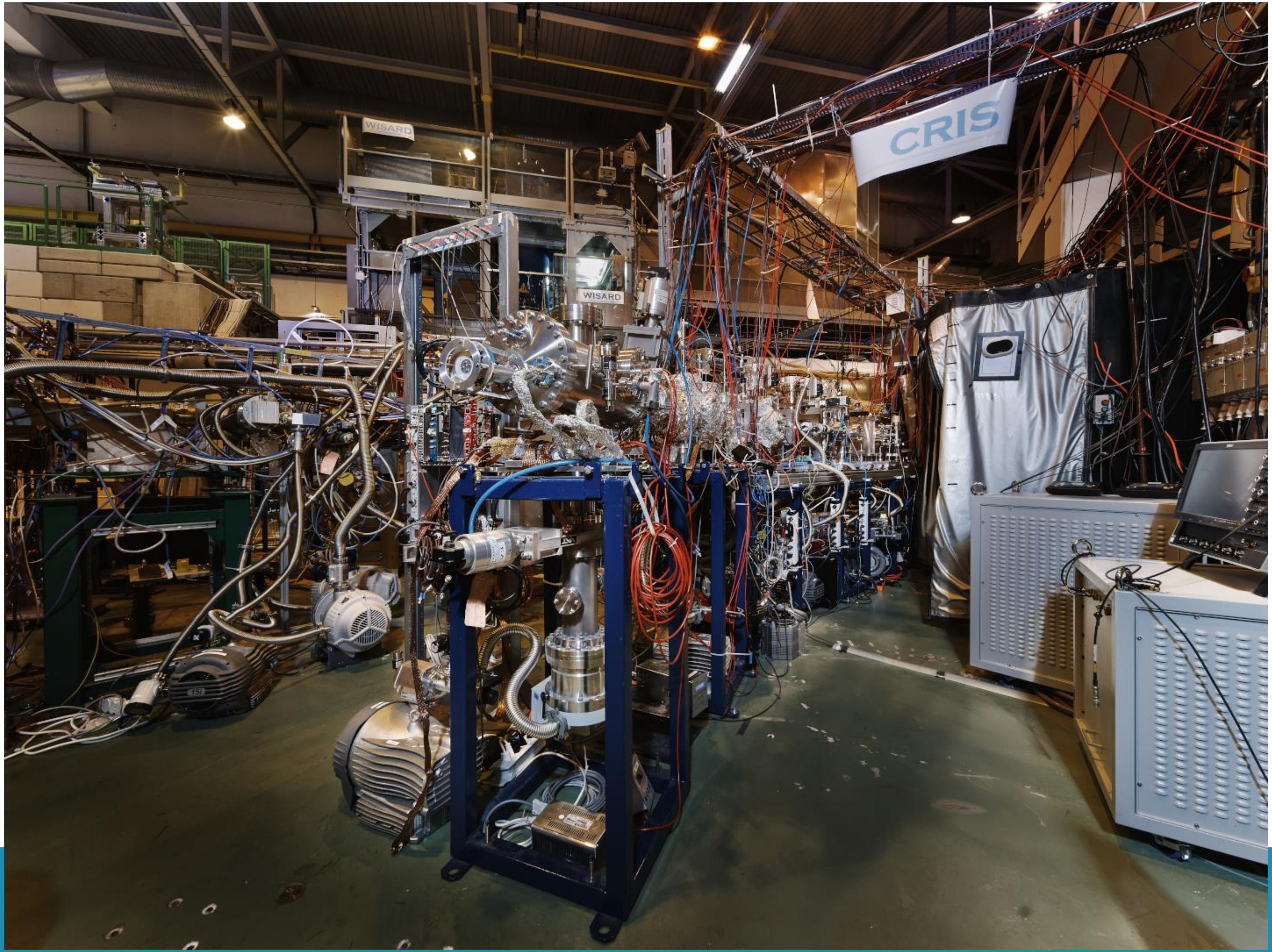
- Unique worldwide: high energy protons (1.4 GeV) on thick targets
- The (heavy) target nucleus splits in smaller nuclei through

> ➔ More than 1300 'exotic' isotopes/isomers of more than 74 chemical elements
> ➔ Half-life > 10 ms
> ➔ Since 2018 also molecules (RaF, AcF, …)



Radioactive molecules

CRIS experiment at ISOLDE



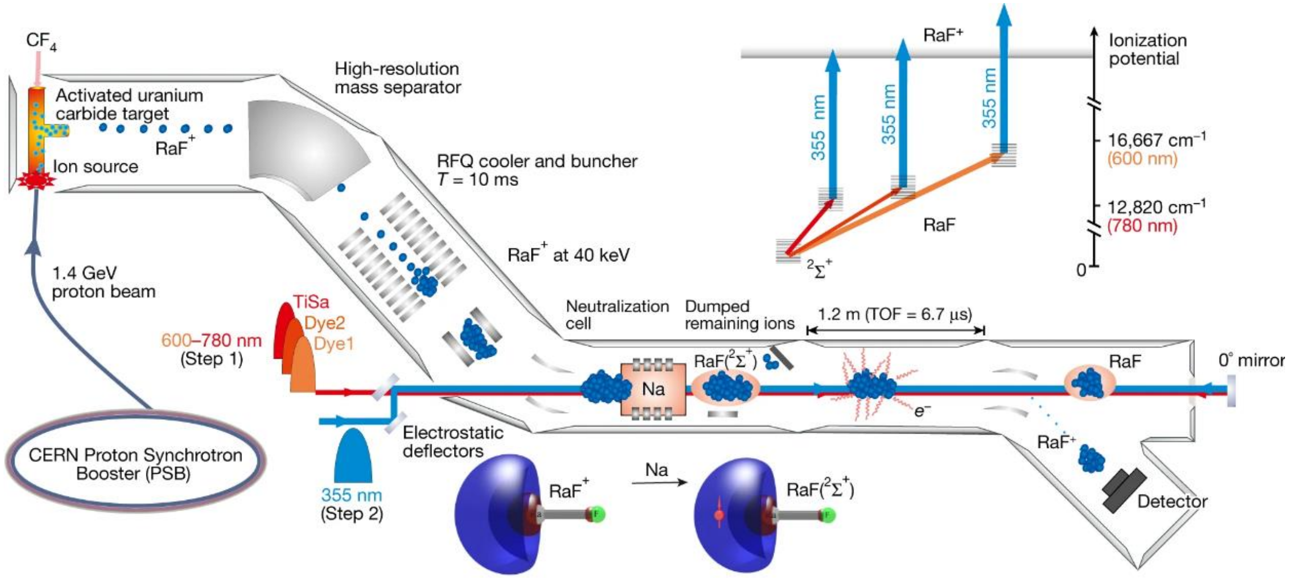
Collinear resonance ionization spectroscopy (CRIS) experiment at ISOLDE-CERN

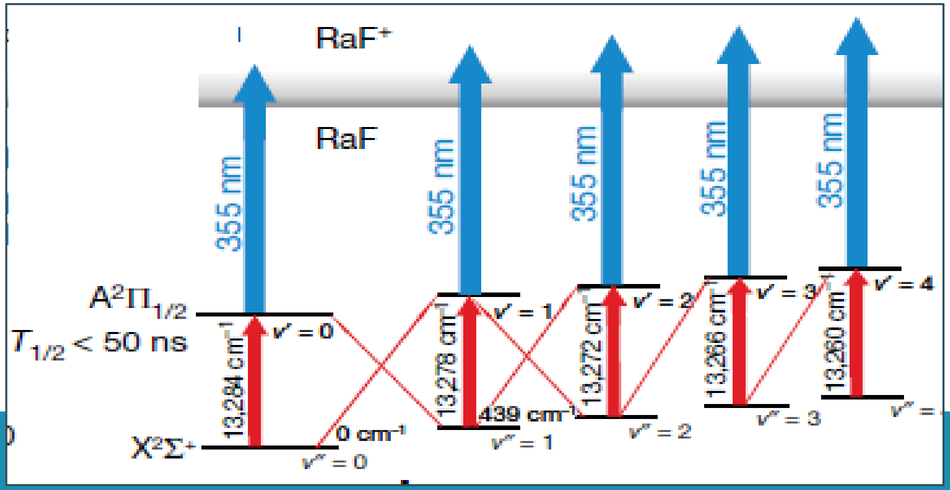
(KU Leuven is founding member)

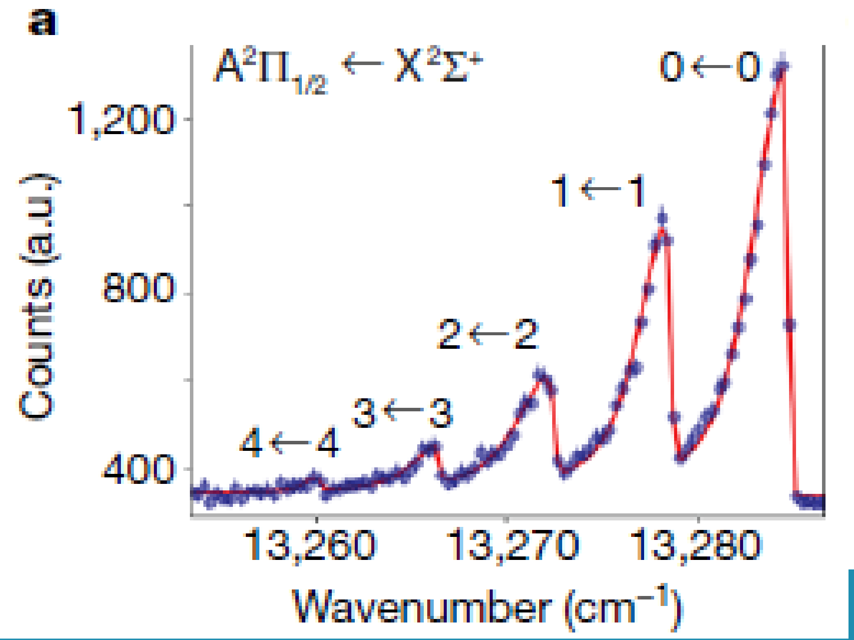
First radioactive molecules spectroscopy on RaF:

Garcia Ruiz et al., Nature **581** (2020)

Collinear resonance ionization spectroscopy (CRIS) experiment at ISOLDE-CERN
(KU Leuven is founding member)





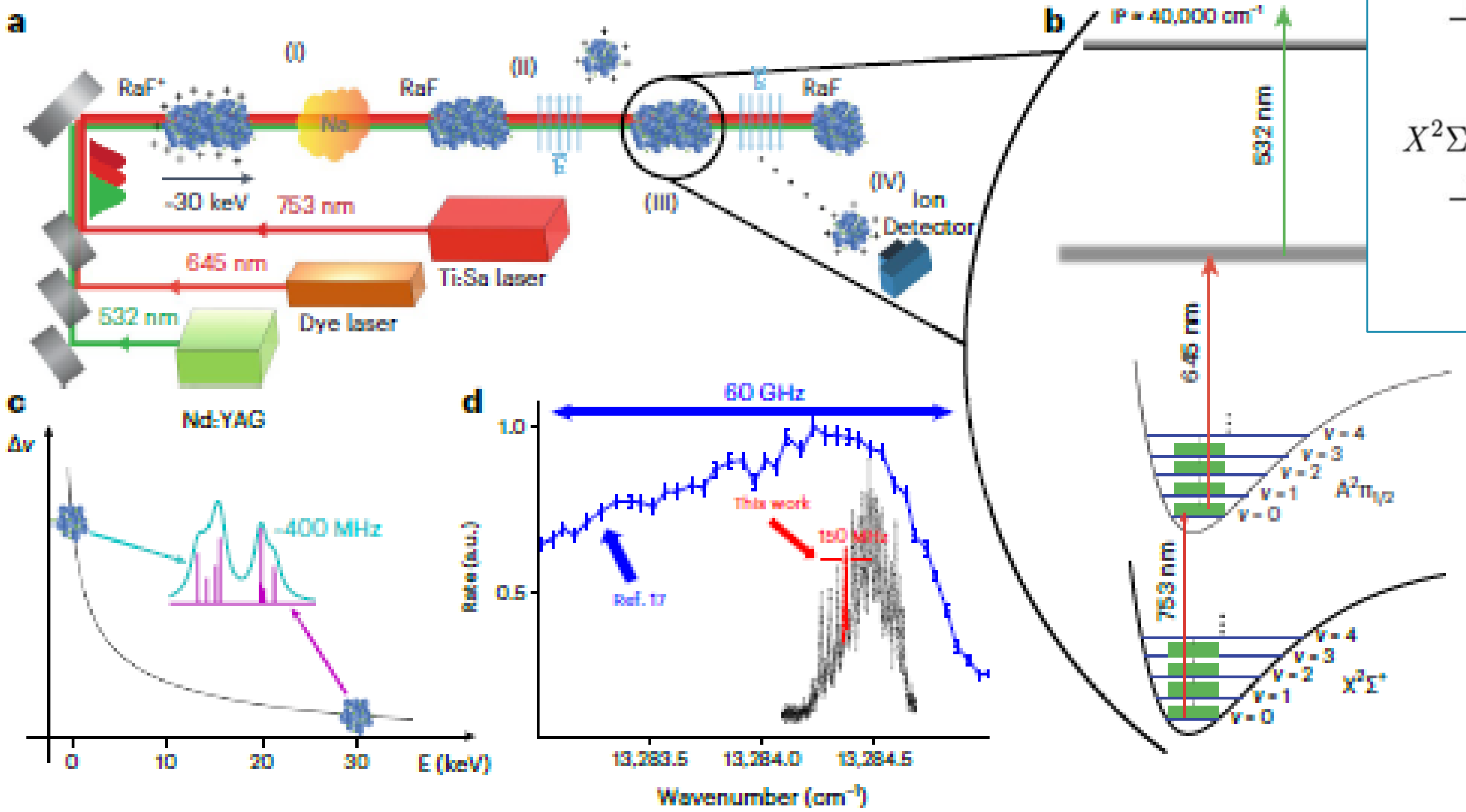


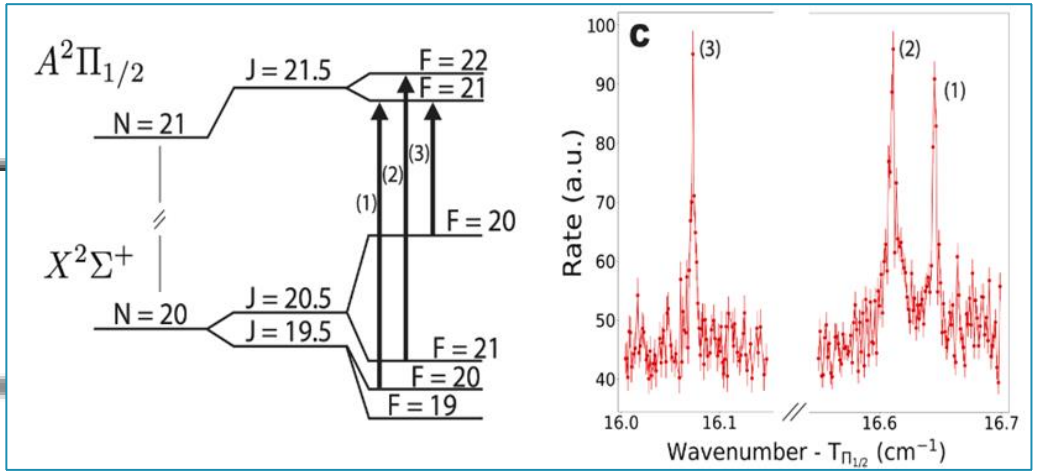
Precision spectroscopy and laser-cooling scheme of a radium-containing molecule:

first step towards an eEDM measurement

S. Udrescu et al., Nature Physics 20, 202 (2024)

Improved resolution from
60 GHz to 150 MHz

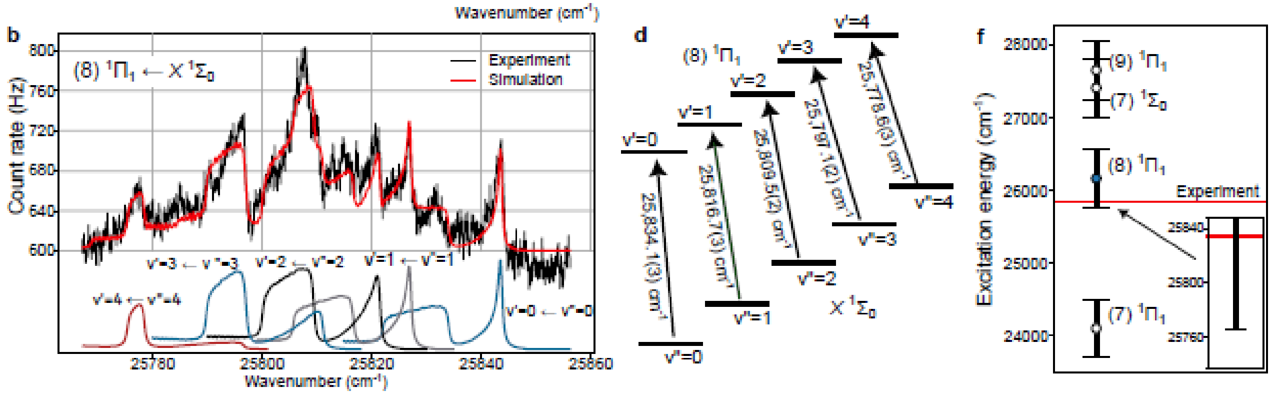




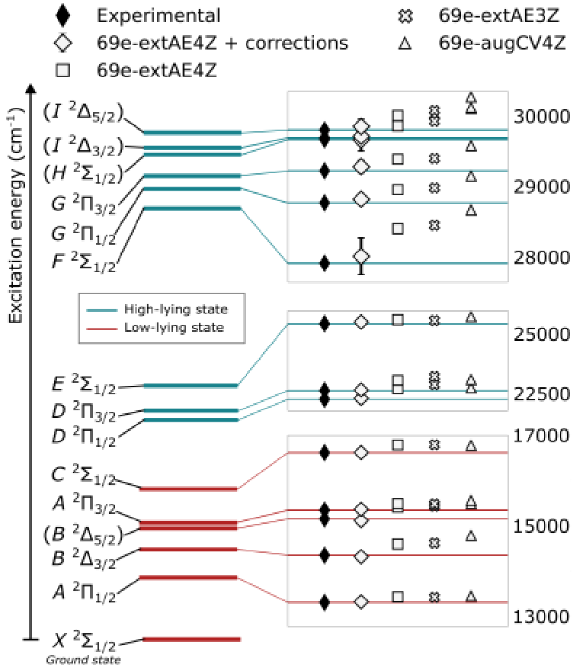
Benchmark molecular theory – needed as input prior to precision studies

M. Athanasakis-Kaklamanakis et al., "*Pinning down electron correlations in RaF via spectroscopy of excited states and high-accuracy relativistic quantum chemistry*" Nature Communications (2025) accepted

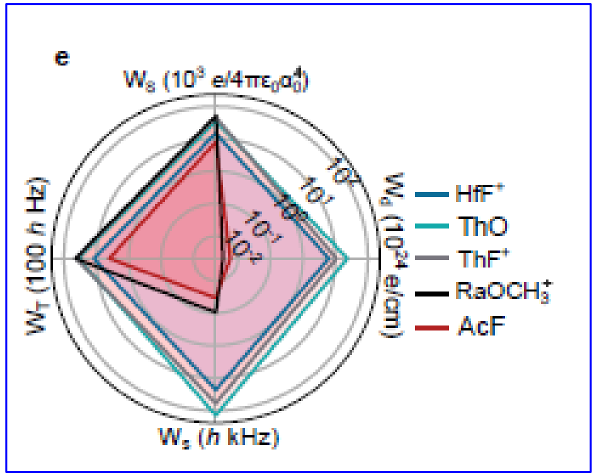
M. Athanasakis-Kaklamanakis et al., "*Laser spectroscopy of AcF as a sensitive probe for CP violation*", in preparation



14 excited states in RaF: 99,9% agreement with theory, after including correlations and QCD corrections



AcF: highest sensitivity to several BSM observables



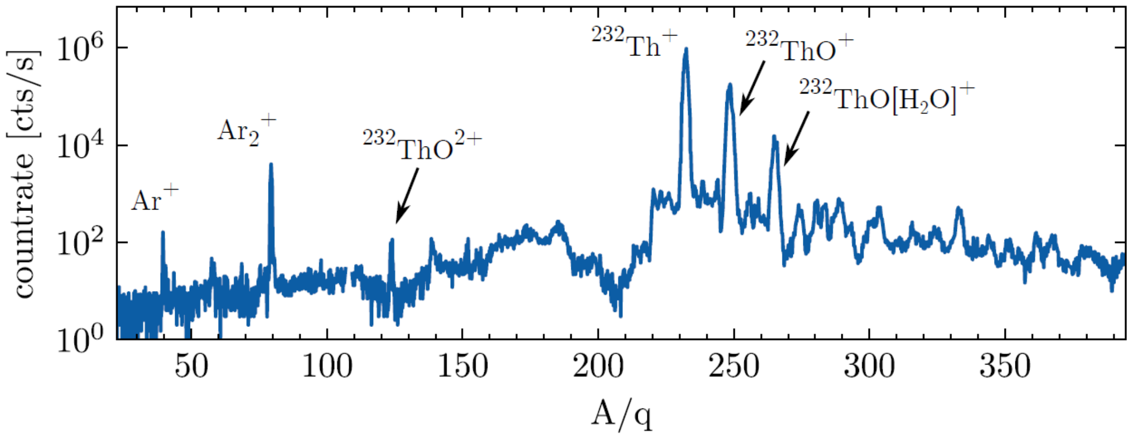
Michail Athanasakis-Kaklamanakis, PhD KU Leuven (2023)

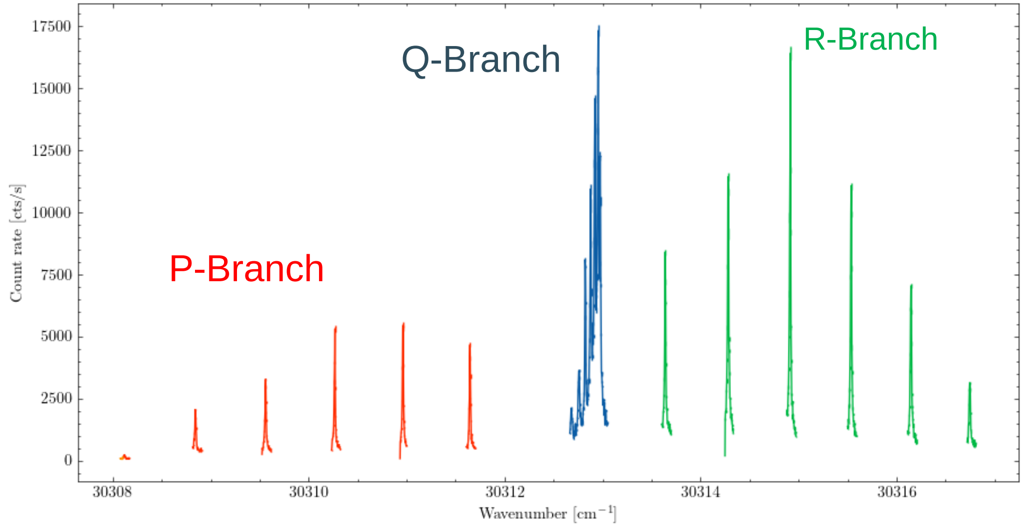
Cooling of molecules using hypersonic nozzles at IKS

Gas jet cools translational temperature, i.e., spread of velocity

- ^{63}Cu: T = 12.5(10) K (M = 8.7(3)) using RIS Flow mapping
- Molecule, internal temperature
  - ➢ **we have ThO available from ablation ion source**

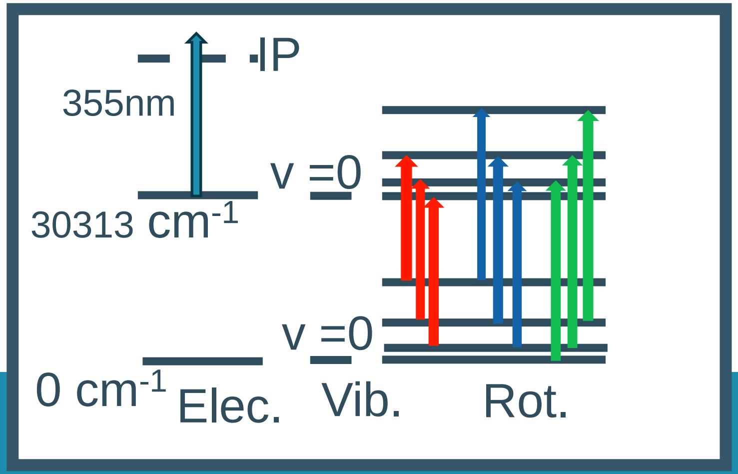
Mass spectrum of molecules from ablation ion source





High-resolution laser spectroscopy of rotational lines in a cooled gas-jet

➔ rotational temperature = 7 K

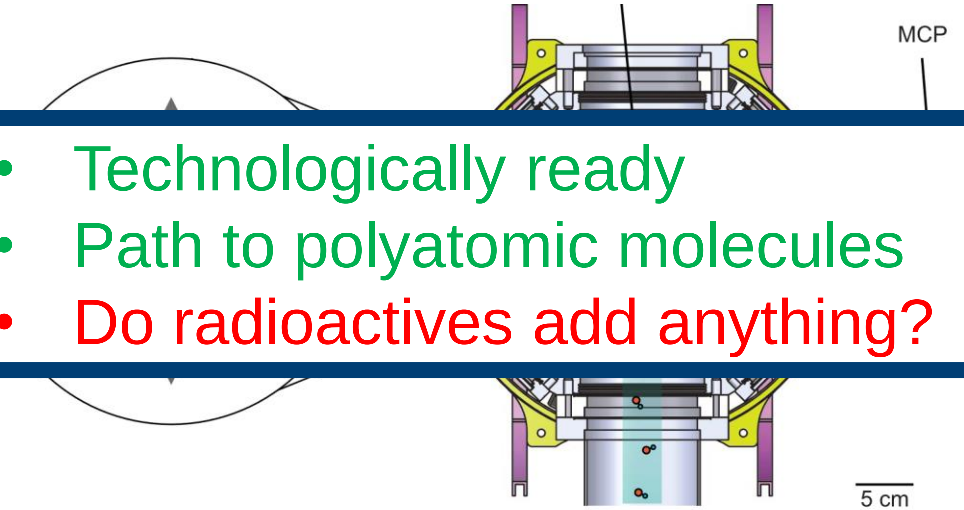


A. Claessens, PhD dissertation, KU Leuven (2024)

Precision experiments: Where do we go from here?

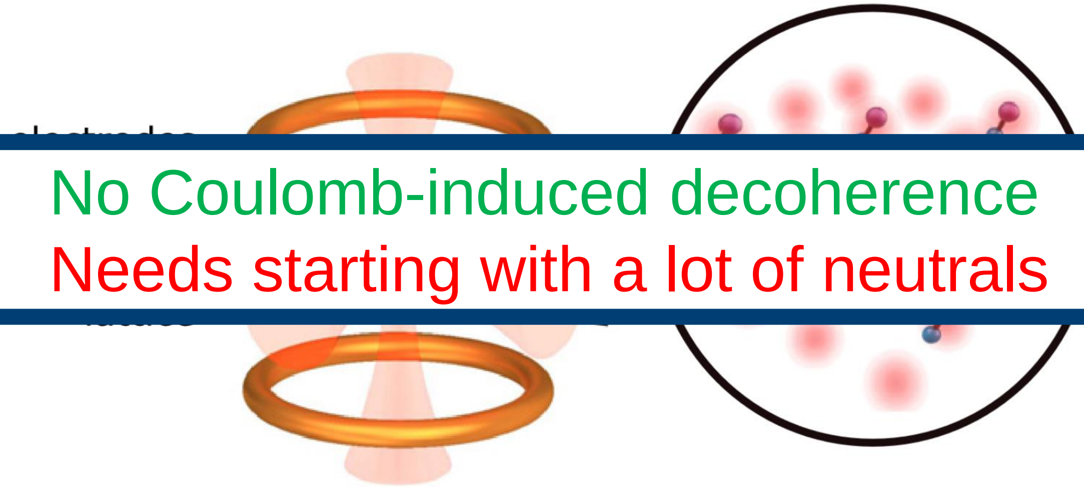
- Realistically: not compatible with beam experiments (one exception?)
- Need TRAPPED molecules - challenging

Ion trapping



- Technologically ready
- Path to polyatomic molecules
- Do radioactives add anything?

MOT → Optical lattice



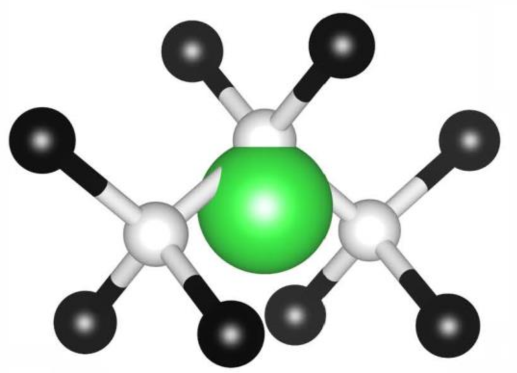
- No Coulomb-induced decoherence
- Needs starting with a lot of neutrals

Roussy et al., Science 381, 46–50 (2023)

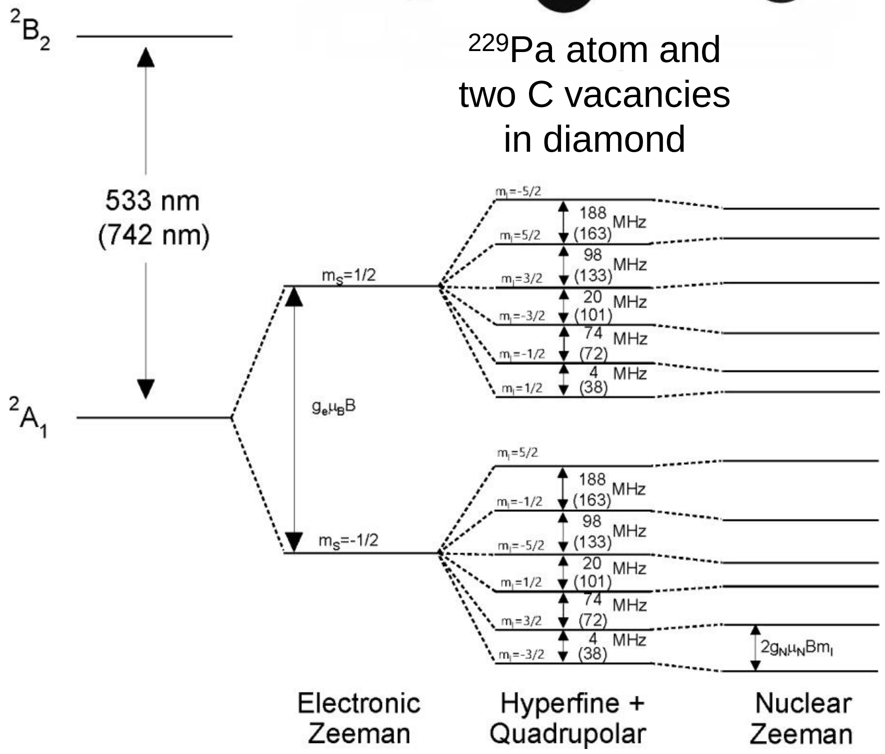
Fitch et al., Q.Sci.T. 6, 014006 (2021)

Alternative *solid-state* approach

- **polar defects in diamond as alternative to polar molecules**
- similar physics/approach, but based on *optical transitions between electronic states of the (optically active) polar defects containing octupole-deformed nuclei*
- candidate isotopes (large Schiff moments): **^{229}Pa, ^{229}Th, …**
- **advantages: large ensembles, without need for complex electromagnetic trapping** (“trapping in a solid”), …
- **ongoing proof-of-principle experiments by KU Leuven team at ISOLDE-CERN (experiment L281) based on ion implantation of Pa, Th and Ra isotopes into diamond** building on expertise on quantum defects in diamond (e.g. [1,2])



^{229}Pa atom and two C vacancies in diamond



Phil. Trans. R. Soc. A 382, 20230169 (2023)

[1] *Phys. Rev. Lett.* 125, 045301 (2020)
[2] *Mater. Quantum. Technol.* 4, 025101 (2024)

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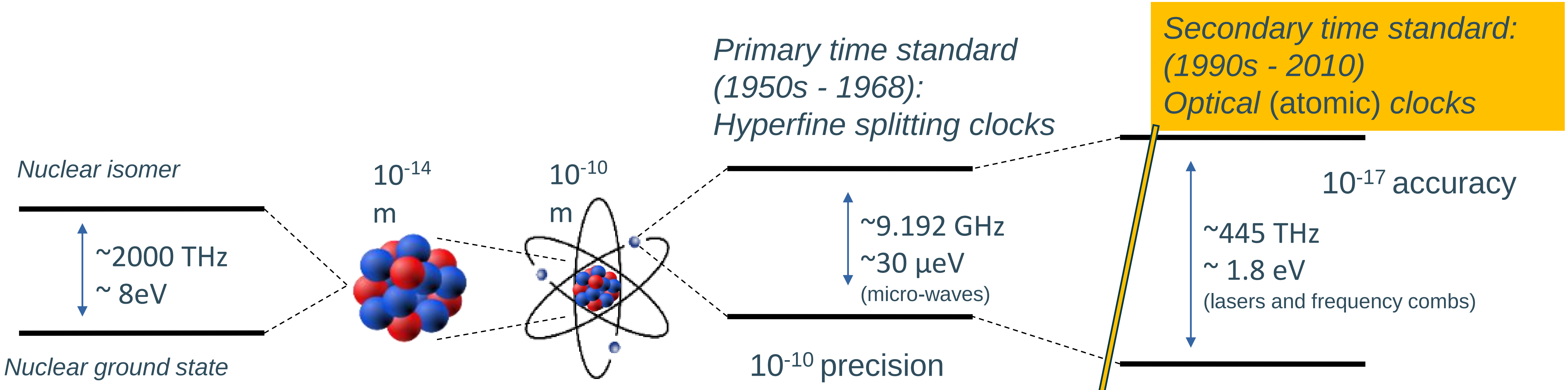
Necessary input from theory

- Prediction of **electronic transition energies** and transition dipole moments
- Ionization and dissociation energies
- Excited state lifetimes and **metastable states**
- Chemical reaction modeling, charge exchange simulations

The ^{229m}Th nuclear clock

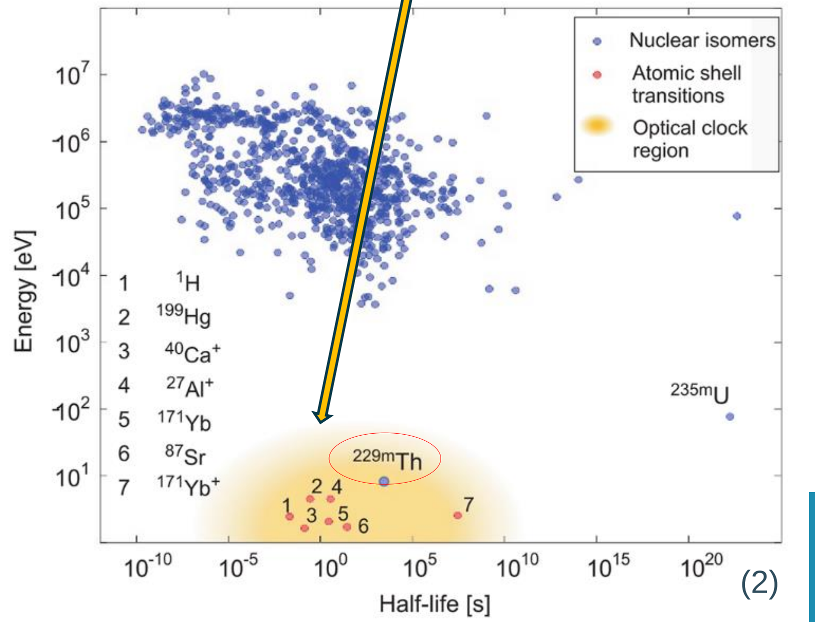
^{229m}Th: a unique isomer in the nuclear landscape



Peik and Tamm., (2003) (1)

➔ Towards a ^{229}Th nuclear clock ?
ultimate precision !

2016: After 50 years indirect evidence: **first direct observation** of the isomer ! (2)



(2)

(1) Peik and Tamm., *Europhys. Lett.* **61** 2 (2003)
(2) Von der Wense et al., Nature 533 7601 (2016)

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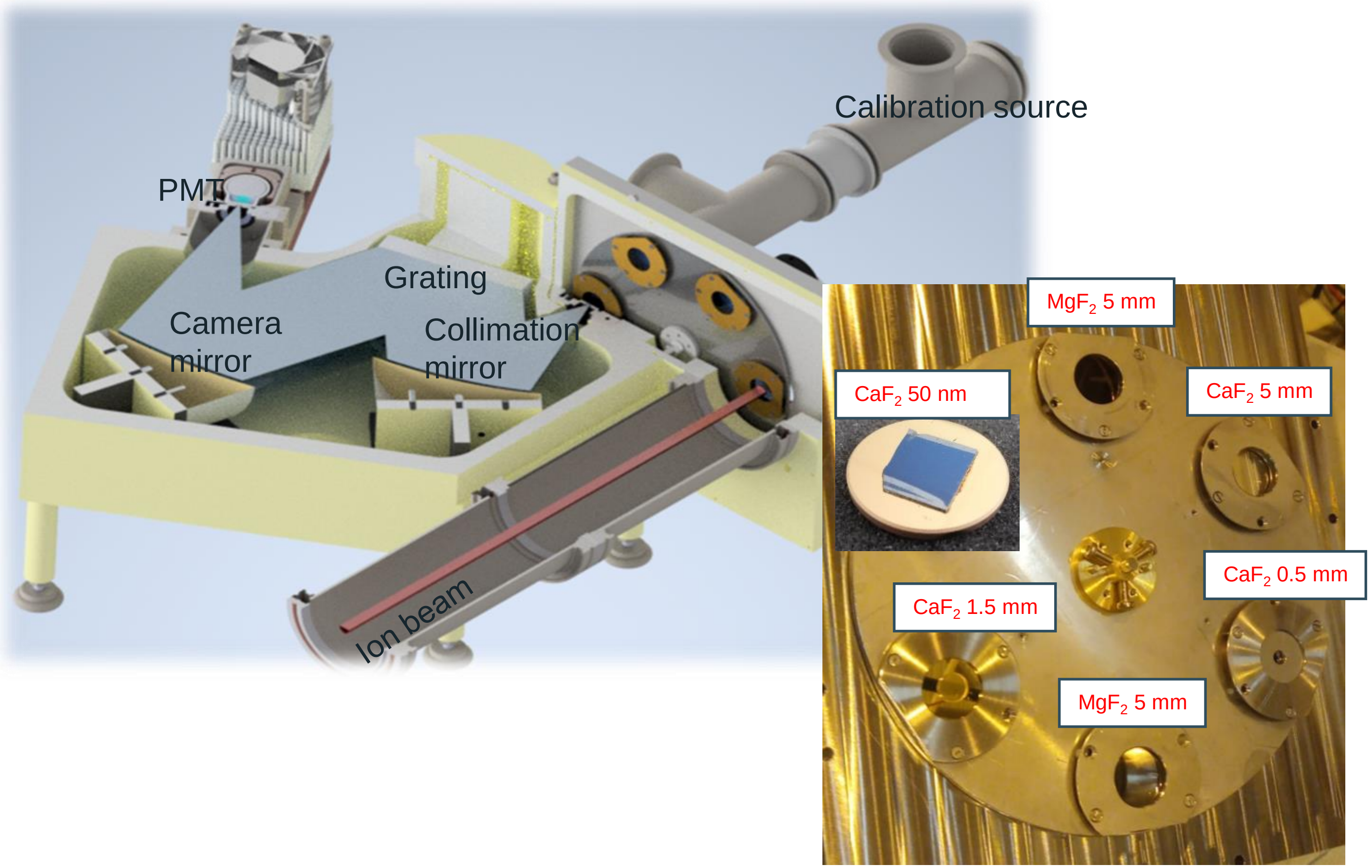
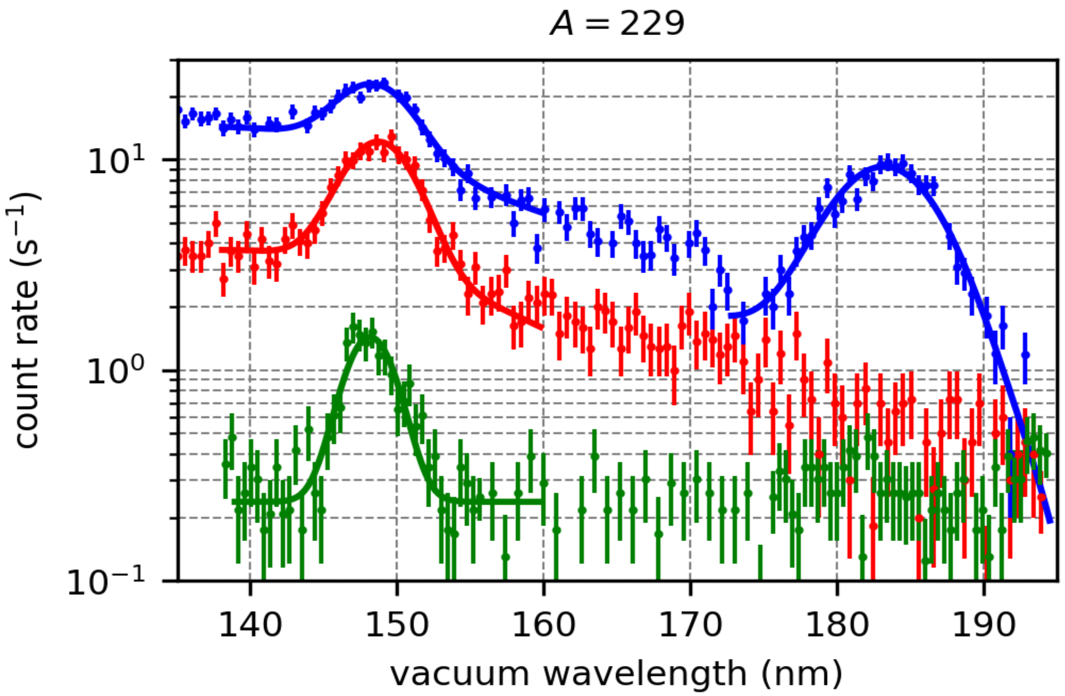
^{229}Th nuclear clock transition

VUV-spectroscopy on radioactive ^{229}Ac (62 min)/^{229}Th implanted in different optically transparent crystals at ISOLDE



→ 2023: first detection of the photon decay of ^{229m}Th at ISOLDE:
E = 8.338(24) eV, ν=148.7(4) nm



→ 2024: first laser excitation of ^{229m}Th at JILA

transition frequency precision improved by 10^6

^{229}Th nuclear clock: fundamental physics opportunities

Probe: measure oscillations of ^{229m}Th transition relative to another clock transition

Nuclear transition: EM + QCD coupling

Electron shell transition (atoms or molecules): EM coupling only

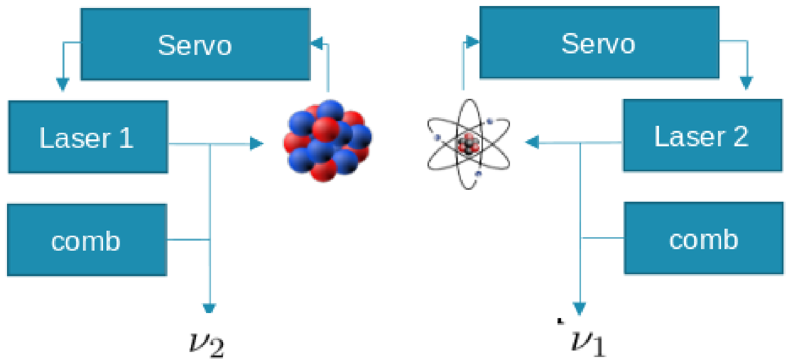
Time variations of coupling constants(3) such as α

$$\frac{\partial}{\partial t}\ln\frac{\nu_2}{\nu_1} = (K_2 - K_1)\frac{1}{\alpha}\frac{\partial \alpha}{\partial t}$$

$K_1 \approx 1$

$K_2 > 10$

4



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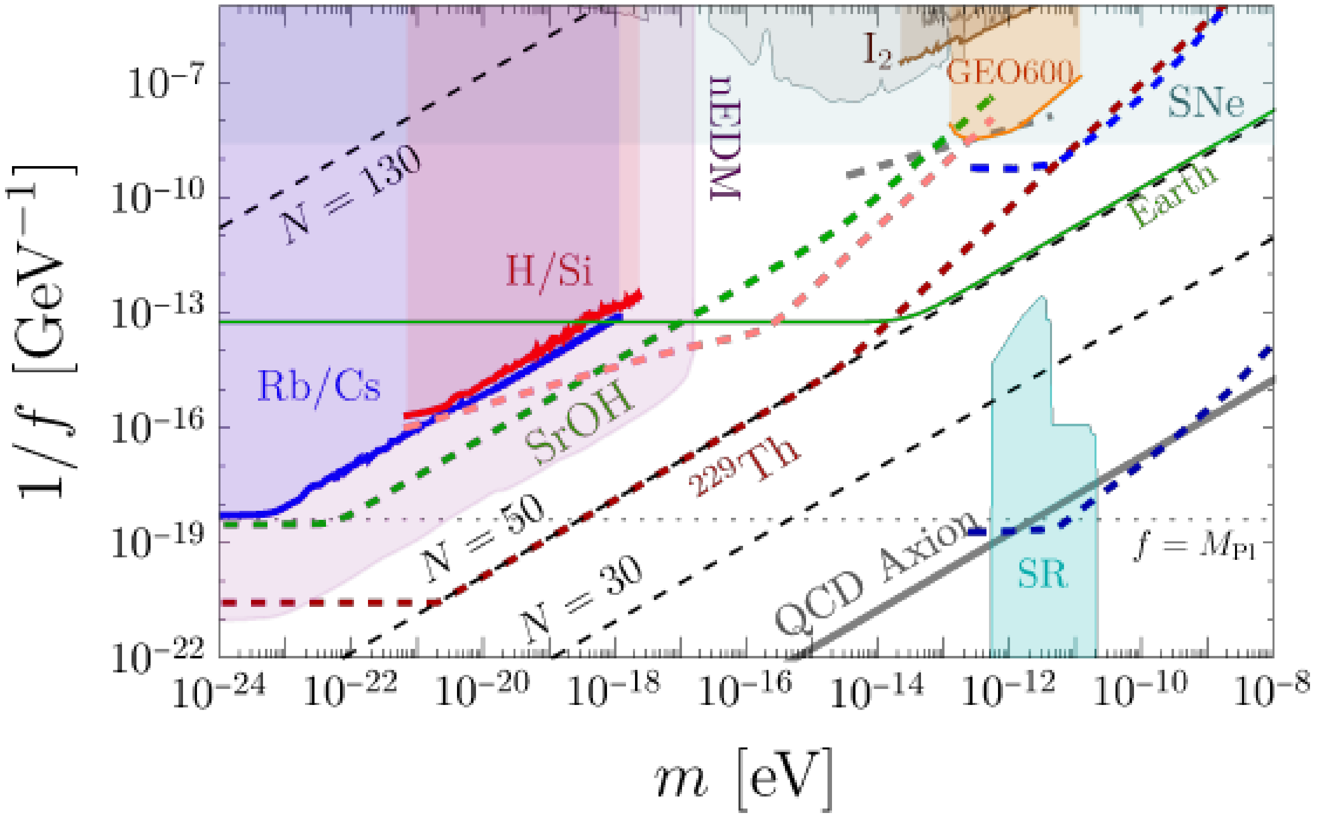
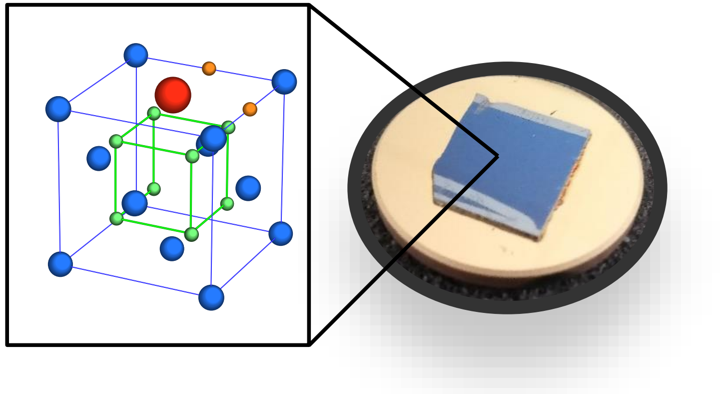
Searches for ultralight dark matter(4)

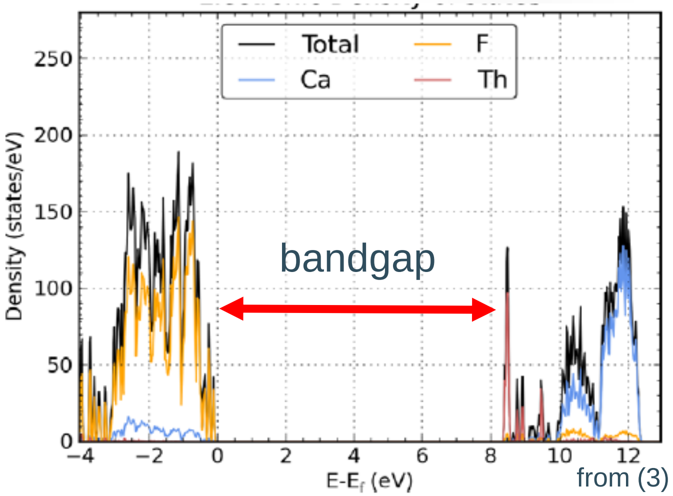
Constraints on axion-gluon coupling



^{229}Th nuclear clock

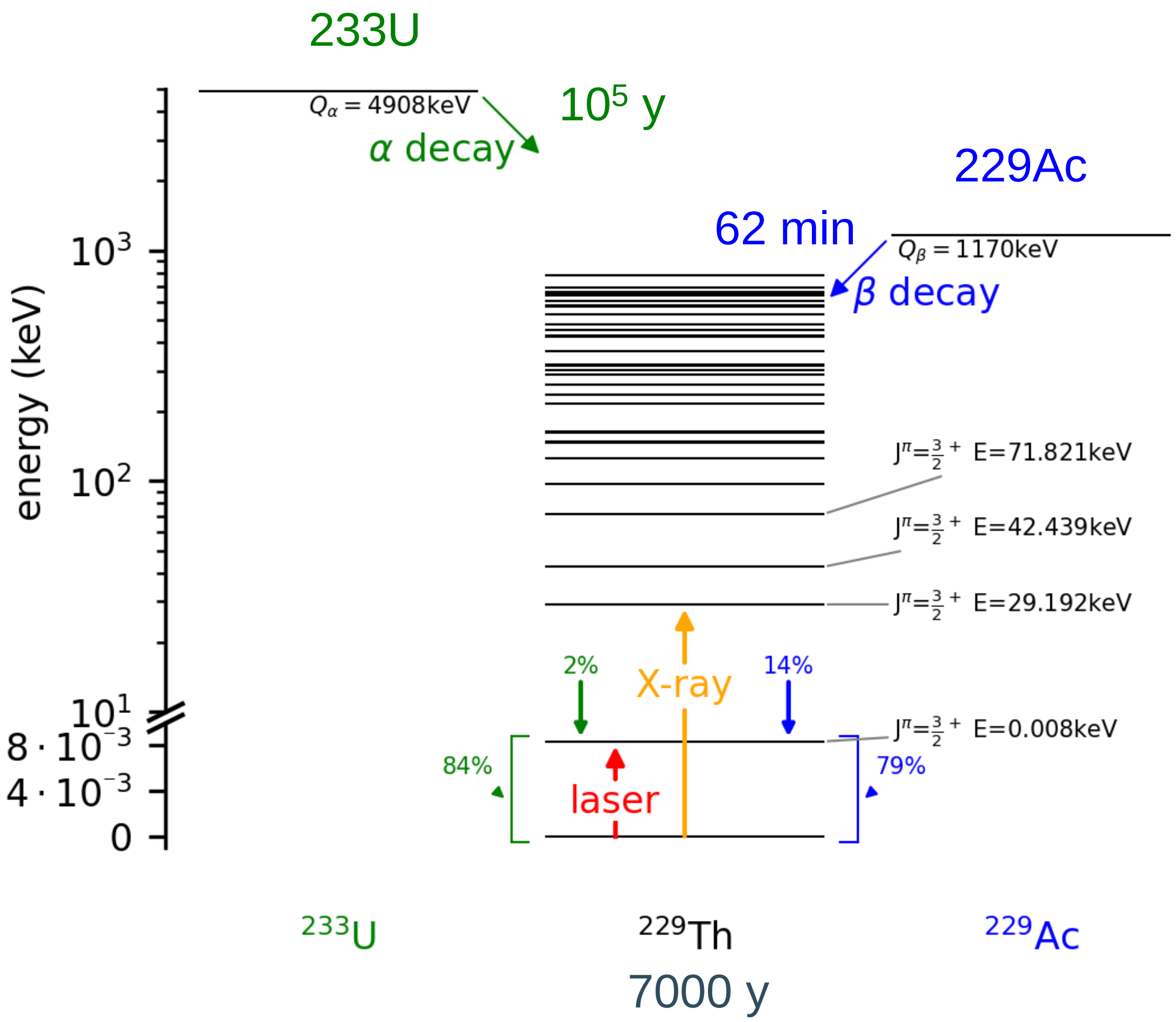
Solid-state approach: High density compensating for small laser excitation cross section





from (3)

Population of nuclear states in ^{229}Th



Dessovic. et al. *J. Phys. Condens. Matter* **26** 10 (2014)
L. Pereira, A. Vantomme, P. Van Duppen, S. Kraemer (LoI 281, Implantation of 229Ac and 229Th in diamonds, ISOLDE)

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Isotope shifts to search for a 5th force

Fifth force searches with isotope shift spectroscopy

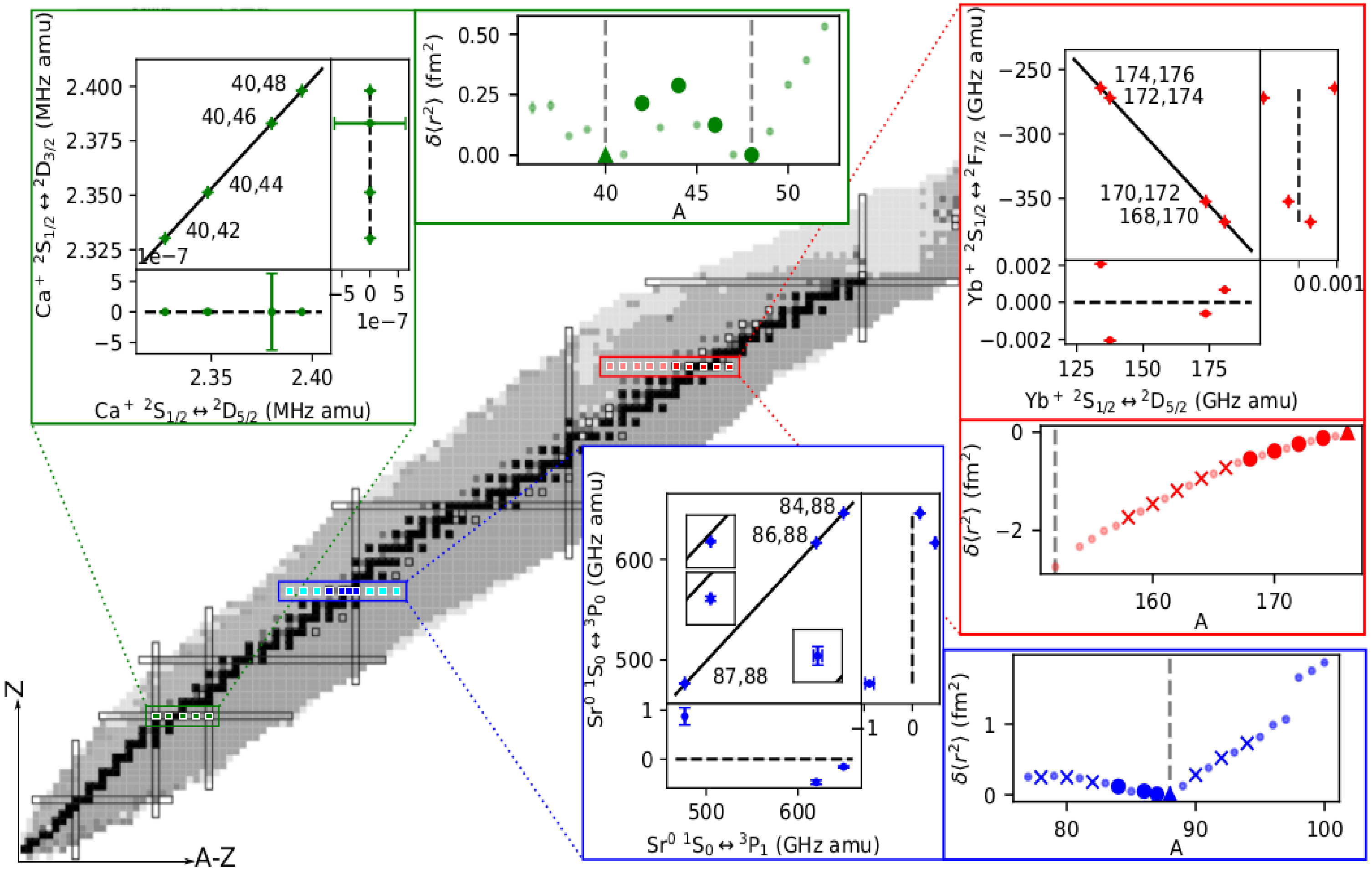
Probe: compare transition frequencies in atoms and ions
King plot analysis (comparison of isotope shifts between two isotopes)

Intensive field of research in the past 5 years.
Currently only on stable isotopes of Ca (5), Sr (4), Yb (5)

→ By extending to radioactive isotopes, the king-plots can be extended to include more points

→ Better test of non-linearity

→ Verify potential origin of non-linearity (nuclear polarization ? nuclear structure ?)



Fifth force searches with isotope shift spectroscopy

New interaction that couples neutrons to electrons

Yb^0/Yb^+

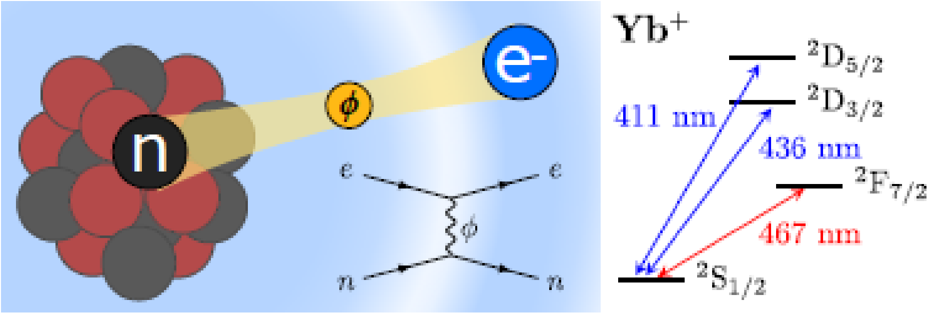
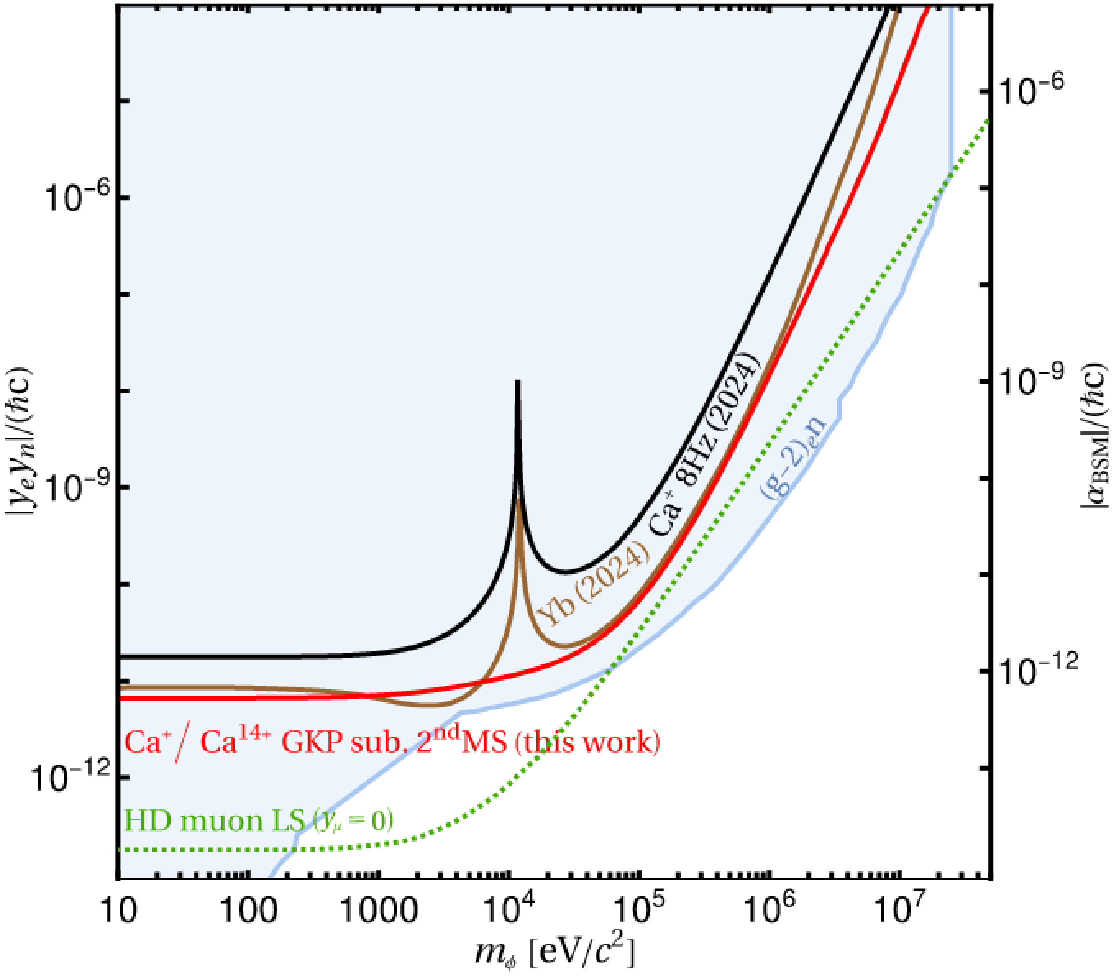


FIG. 1. New interatomic force arising from the exchange of a hypothetical ϕ boson between neutrons and electrons (left), and simplified Yb^+ level structure (right). In this work, the highly forbidden octupole transition at 467 nm is measured, and compared to the quadrupole transitions at 411 nm and 436 nm that were probed in Ref. [18].

arXiv:2201.03578v1

Ca^+/Ca^{14+} (2)



Hur et al., Phys. Rev. Lett. 128, 163201 (2022)

Wilzewski et al, arXiv:2412.10277 (2024)

Outlook: radioactive beam laboratories around the world

ISOLDE is leading
BUT: Current ISOLDE hall is NOT suited for ultra-high precision studies !
NEW ISOLDE target stations and exp hall ~ 300-400 Meuro
(EPIC project, presented ESPP Granada meeting 2018)

> Multiple laboratories around the world have now initiated Radioactive Molecules activities

ISOL@Myrrha (PTF): a green field facility at SCK CEN!

MINERVA (Myrrha phase 1) funded with 600 Meuro

- Accelerator 100 MeV protons at 2 mA (~ 450 MEuro)
- Coupled to a **Proton Target Facility (~150 Meuro)**
- Design finalized
- Dec 2022: formal approval to start construction (ongoing)
- 2029: isotopes from PTF
- **3-4 experiments for ultra-high precision studies that require long beamtimes !**

Outlook

The field of **ultra-high precision spectroscopy** using high-resolution lasers, rf-fields, optical frequency combs, trapped atoms and molecules, … for testing the Standard Model (and to search for BSM physics) is maturing !

We can bring something unique to this: species that are orders of magnitude more sensitive to find new physics !

BUT: they are RADIOACTIVE
Ongoing developments at KU Leuven to develop these high-precision methods and to make them applicable to radioactive species (led by Ruben de Groote, Agi Koszorus, Lino Pereira, GN)
 * initially for nuclear structure studies
 * later for BSM physics

Thanks to ..

Elise Wursten
KU Leuven PhD 2021
Post-doc Imperial College London

neutron EDM studies

Michail Athanasakis-Kaklamanakis
KU Leuven PhD 2023
Post-doc Imperial College London

radioactive molecules for EDM and Shiff moment studies

Sandro Kraemer
KU Leuven PhD 2023
Post-doc KU Leuven

229Th nuclear clock and isotope shifts King plots

Arno Claessens
KU Leuven PhD 2024
Post-doc

Gas-jet spectroscopy of ThO

Nathal Severijns
Prof. KU Leuven (IKS)

Neutron EDM and other CP-violating measurements

Lino Pereira
Associate prof. KU Leuven (QSP)

Chip-scale nuclear/molecular clocks (Th, Pa, …)

Ruben de Groote
Assistant prof. KU Leuven (IKS)

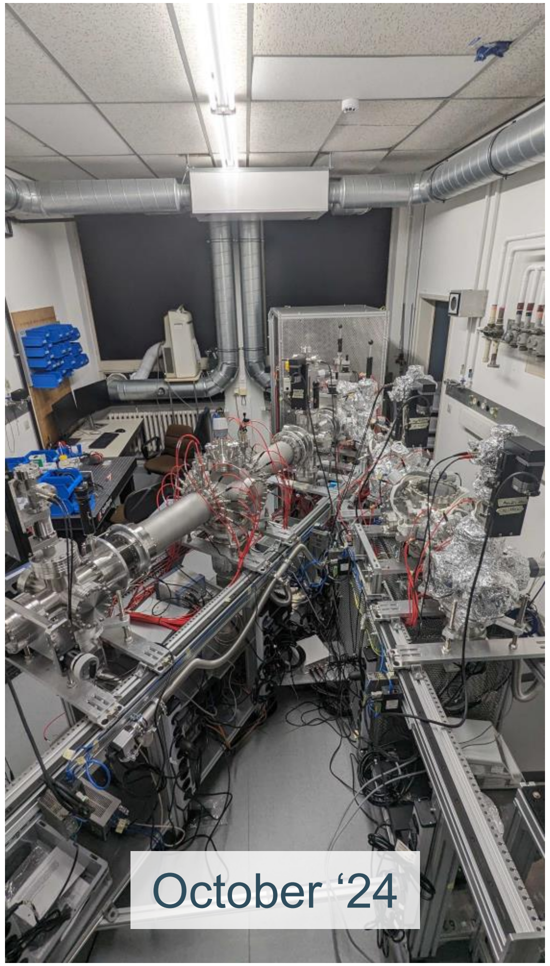
towards high-precision studies on radioactive species (ionic crystals)

Agi Koszorus
Assistant prof. KU Leuven (IKS) and SCK CEN

towards high-precision studies on radioactive species (beam preparation)

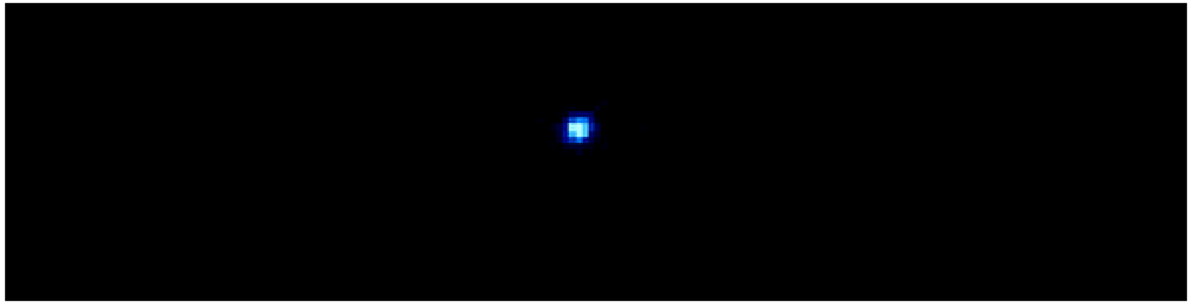
Supporting slides

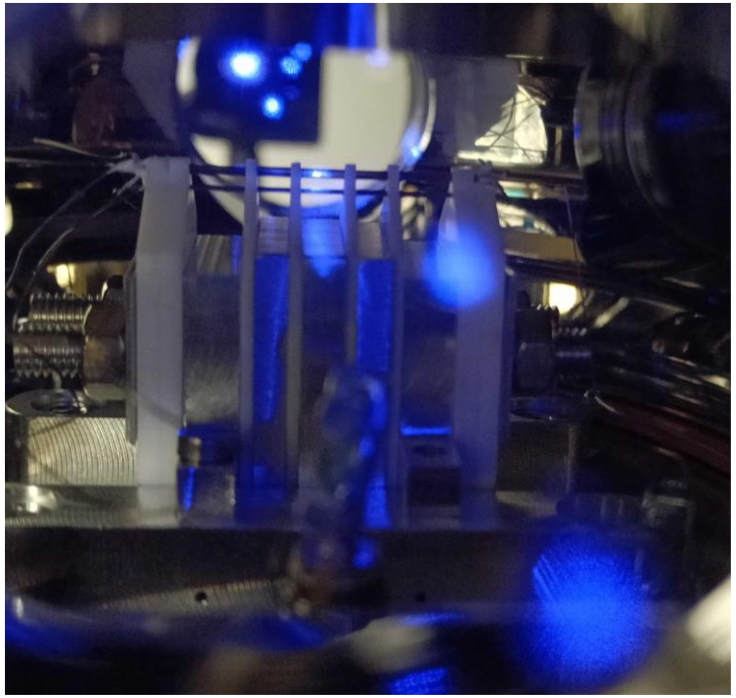
Construction progress IKS labs during 2024



Led by Agi Koszorus

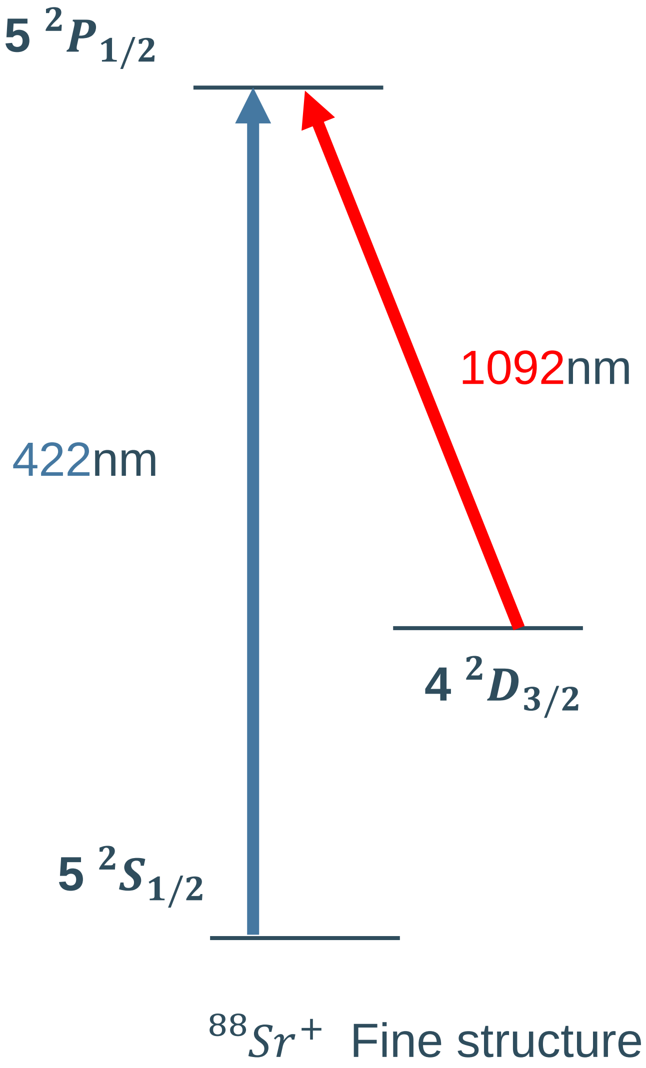
Trapped Sr ions: an ion crystal





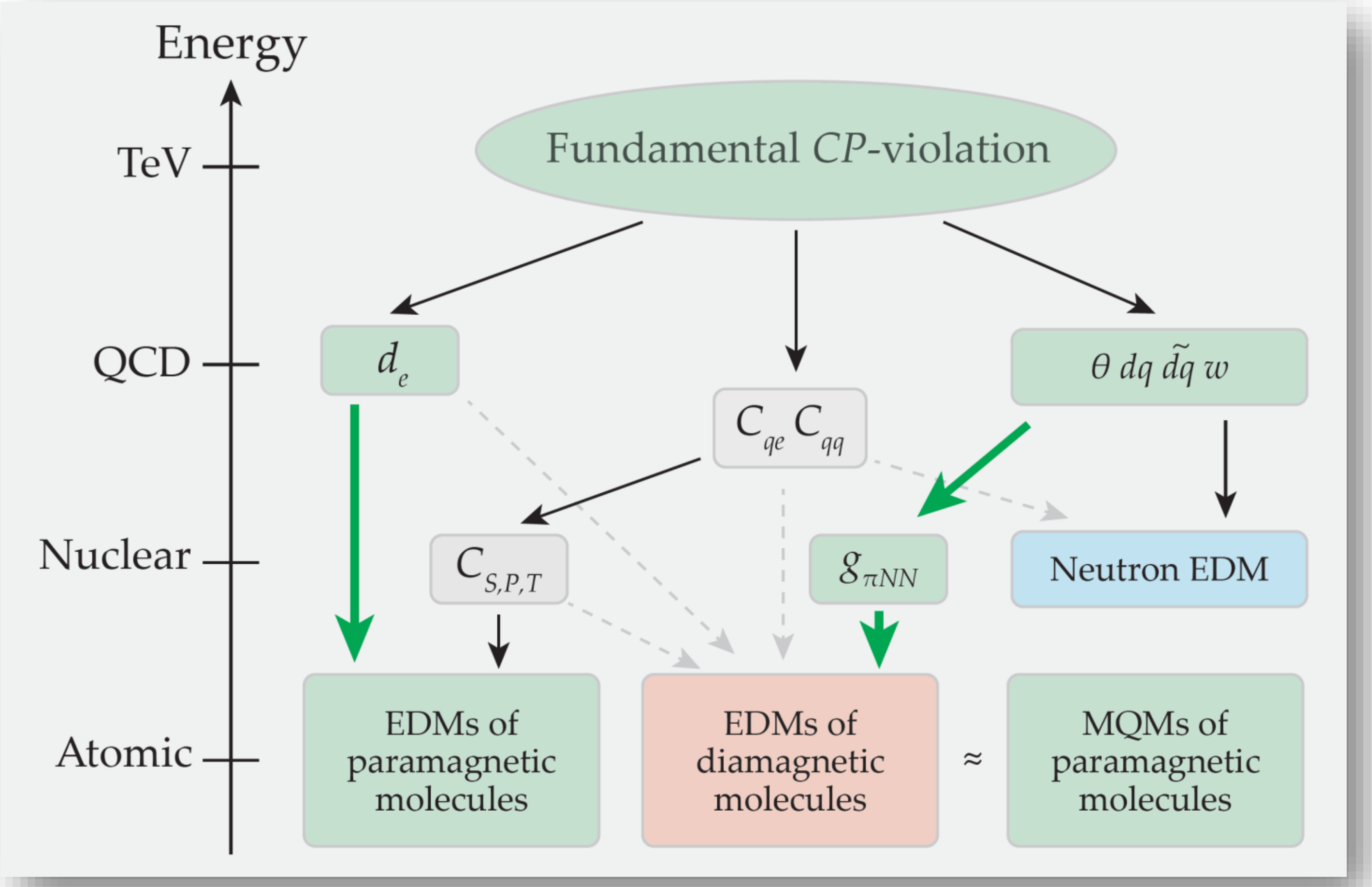
T ~ **10 mK**
→ ions form a ‘coulomb crystal’
Storage time **~10s** for single ions

→ mostly limited by base pressure of 1E-8mbar

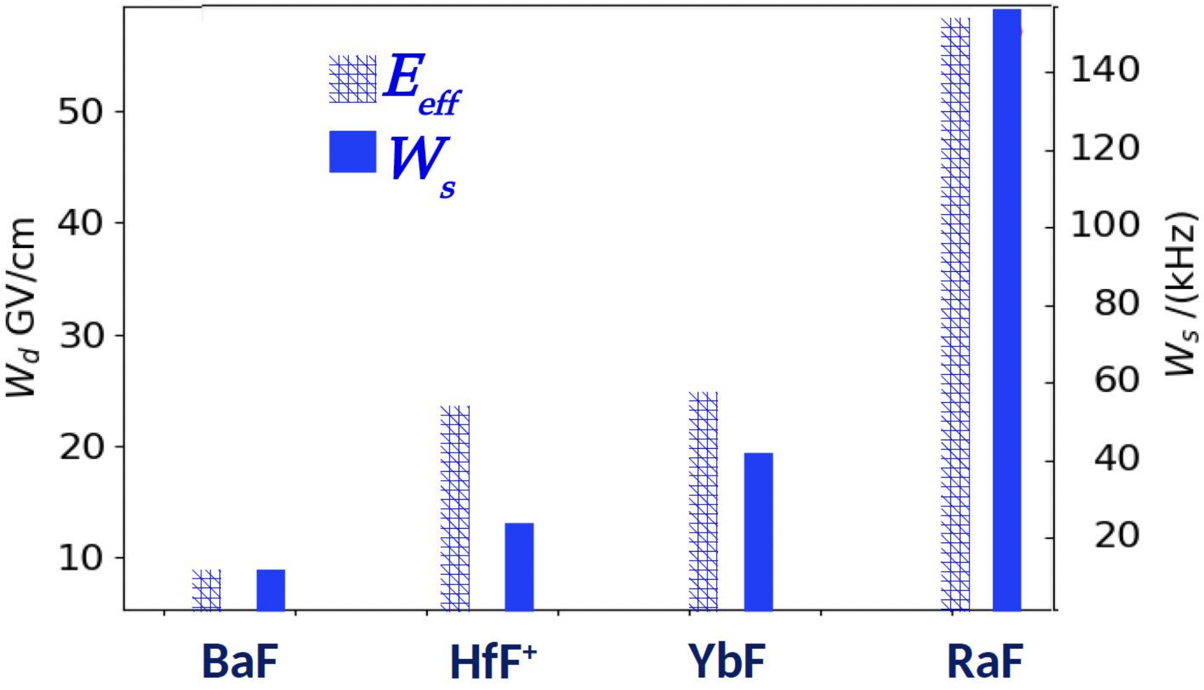


eEDM roadmap

- In the past 10 years
  - 250 times improvement on eEDM
  - YbF, ThO, HfF^+
  - 10 TeV energy scale
- In the next decade
  - Another 2-orders of magnitude
  - **Toward PeV energy scale**
  - **Far beyond LHC energy**
  - Cross-verifications
    - Species
    - Platforms
- Hadronic sector of the Standard Model



RaF is a highly promising system!

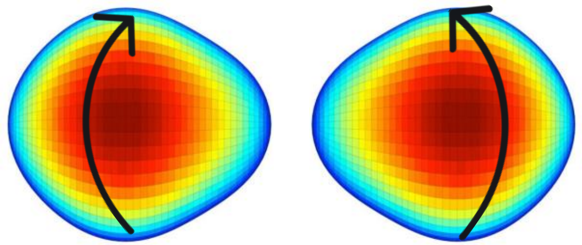


Plot courtesy of R. F. Garcia Ruiz and S. G. Wilkins (MIT)

Signatures of *P,T*-violating moments
>10^5 times than in atoms!

Heavy & deformed nucleus

Butler et al., PRL **124** (2020)



Gaffney et al., Nature **497** (2013)

close-lying opposite-parity rotational doublet

$$|J^{\pm}\rangle = \frac{1}{\sqrt{2}}\left(|\Omega\rangle \pm |-\Omega\rangle \right)$$

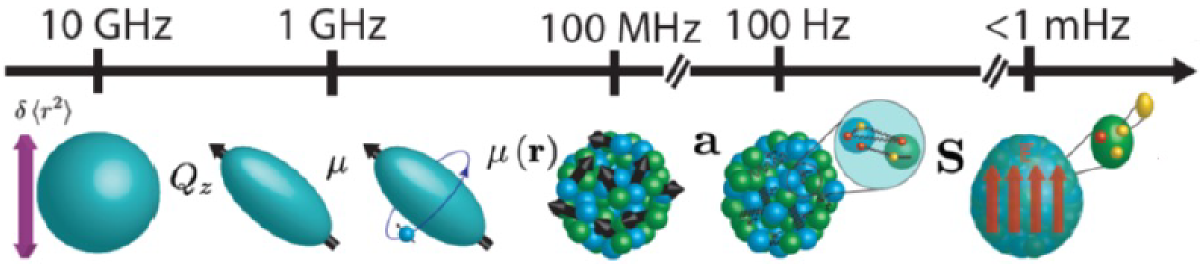
stronger P,T-odd interaction mixing

$$\alpha = \frac{\langle J^{+}|\widehat{W}|J^{-}\rangle}{E^{+} - E^{-}}$$

Enhanced symmetry-violating nuclear moments in Ra

Why atoms (Radioactive) molecules?

Molecules with heavy, exotic nuclei: Ra(Z=88)



Nuclear spin > 0

$$H_{mol} = H_e + H_{vib} + H_{rot} + \dots + H_{hfs} + H_{PV} + H_{PTV} + d_e E_{eff}$$

[1 eV=241.8 THz]

eV	~ 2	10^{-2}	10^{-5}	10^{-8}	$<10^{-12}$	$<10^{-18}$	$<10^{-20}$

$\sim O_{Nucl}\, E_{mol}$

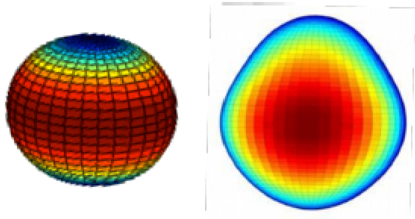
Nuclear

$\sim Z A^{2/3} \beta_2 \beta_3^2 / (E^N_+ - E^N_-)$

protons · nuclear mass · nuclear deformation · nuclear levels

Molecule

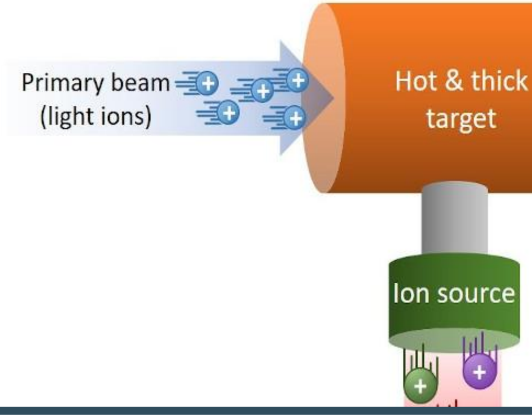
$\sim Z^3 / (E^e_+ - E^e_-)$



$\beta_2 > 0$ $\beta_3 > 0$

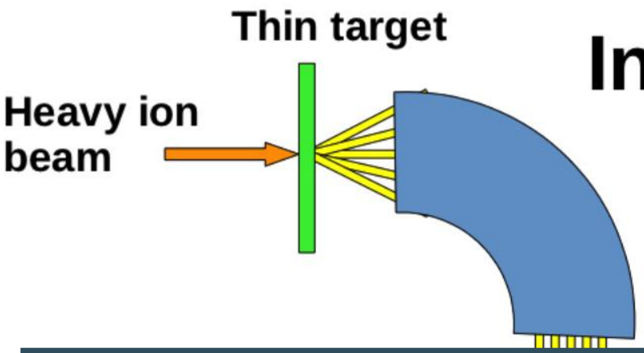
R.F. Garcia Ruiz (MIT), ICAP Conference, 2024

Radioactive isotope production: 2 approaches



ISOL method

- Well-behaved ion beams
- All isotopes produced simultaneously
- Chemistry crucial
- Basis for **precision** studies



In-Flight method

- Messy ion beams
- Beam-target choice dictates production
- Largely chemistry-insensitive
- Basis for **discovery** studies

Figs from ISOLPHARM and Maria Borge

Proposed radioactive molecules and their isotopes

- *RaF*
- AcF
- AcF^+
- AcO^+
- PaF^{3+}

- *RaOH*
- *$RaOCH_3$*
- $RaOCH_3^+$
- *$AcOH^+$*

Open-shell molecule
Closed-shell molecule
Laser-coolable

Nuclide	Energy [keV]	J^π	$T_{1/2}$ or Width Abund. [mole fract.]
$^{223}_{88}Ra_{135}$	0.0	3/2+	11.43 d *5*

Nuclide	Energy [keV]	J^π	$T_{1/2}$ or Width Abund. [mole fract.]
$^{225}_{88}Ra_{137}$	0.0	1/2+	14.9 d *2*

Nuclide	Energy [keV]	J^π	$T_{1/2}$ or Width Abund. [mole fract.]
$^{226}_{88}Ra_{138}$	0.0	0+	1600 y *7*

Nuclide	Energy [keV]	J^π	$T_{1/2}$ or Width Abund. [mole fract.]
$^{227}_{89}Ac_{138}$	0.0	3/2-	21.772 y *3*

Isotope sensitivity to nuclear Shiff moment

- <u>*RaF*</u>
- AcF
- AcF^+
- AcO^+
- PaF^{3+}

- <u>*RaOH*</u>
- <u>*$RaOCH_3$*</u>
- $RaOCH_3^+$
- <u>*$AcOH^+$*</u>

Isotope	Schiff moment compared to ^{225}Ra
^{221}Fr	0.14
^{223}Fr	1.6
^{225}Ac	3
^{227}Ac	6
^{229}Th	$\lesssim 2$
^{229}Pa[a]	40

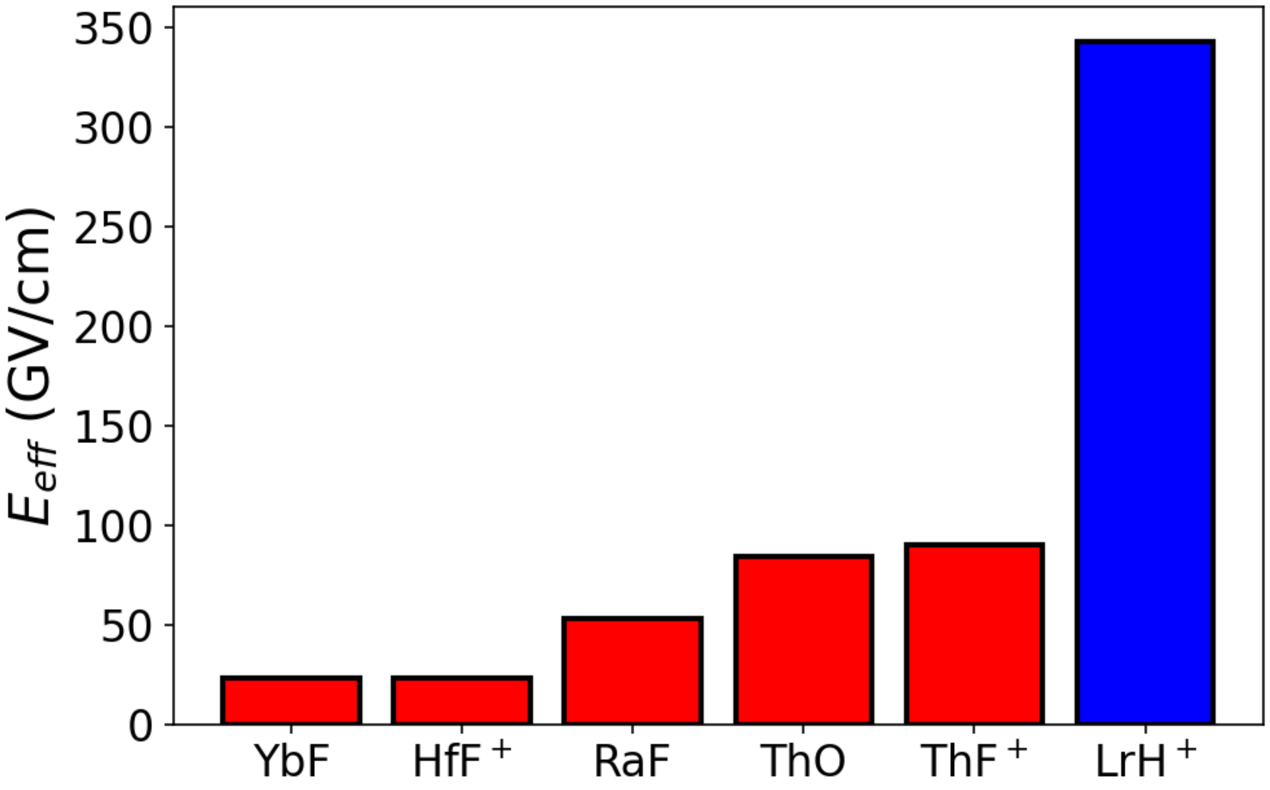
Open-shell molecule
Closed-shell molecule
<u>*Laser-coolable*</u>

Flambaum and Dzuba, PRA 101, 042504 (2020)

Effective electric field in the molecules

- *RaF*
- AcF
- AcF^+
- AcO^+
- PaF^{3+}
- *RaOH*
- *$RaOCH_3$*
- $RaOCH_3^+$
- *$AcOH^+$*

Open-shell molecule
Closed-shell molecule
Laser-coolable



PRA 104, 062801 (2021)

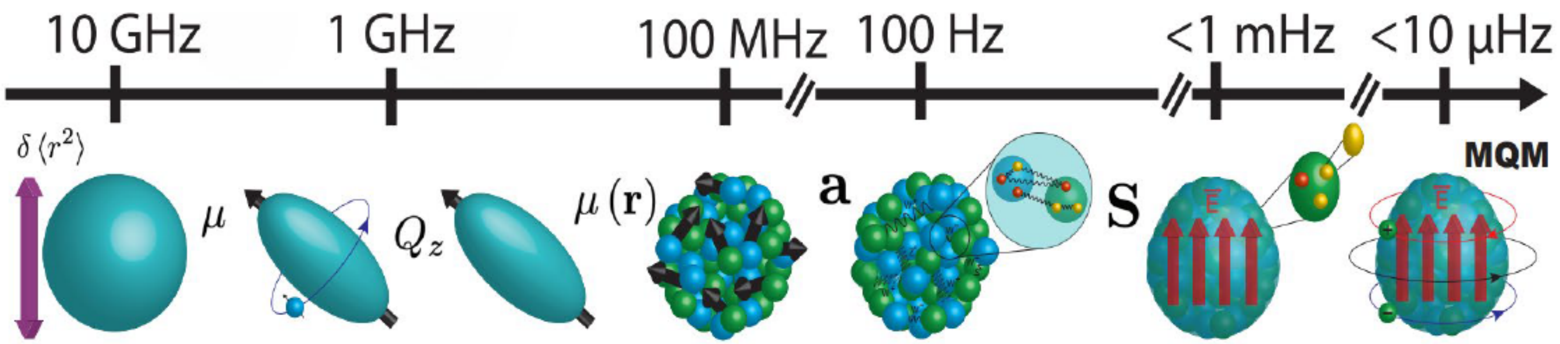
Unique opportunities at ISOL@MYRRHA

Phase 1
Exploiting long beamtimes for precision studies

- Beams which are easily produced
- Tailor-made techniques (which we will master in our offline lab)
- **Double resonance technique** (combine laser and RF)
- Moments, Nuclear magnetization radius
- $^{37-45}$K, Sc, Ca, …

Phase 2
Exploiting long beamtimes for very exotic cases

- Fission fragments provide an exceptional opportunity to extend the measurements on the neutron-rich side
- CRIS experiment combined with MR-ToF
- Measurements of nuclear spins, moments, radii and binding energies beyond *N*=50,82
- Cu, Ag …



KU LEUVEN