

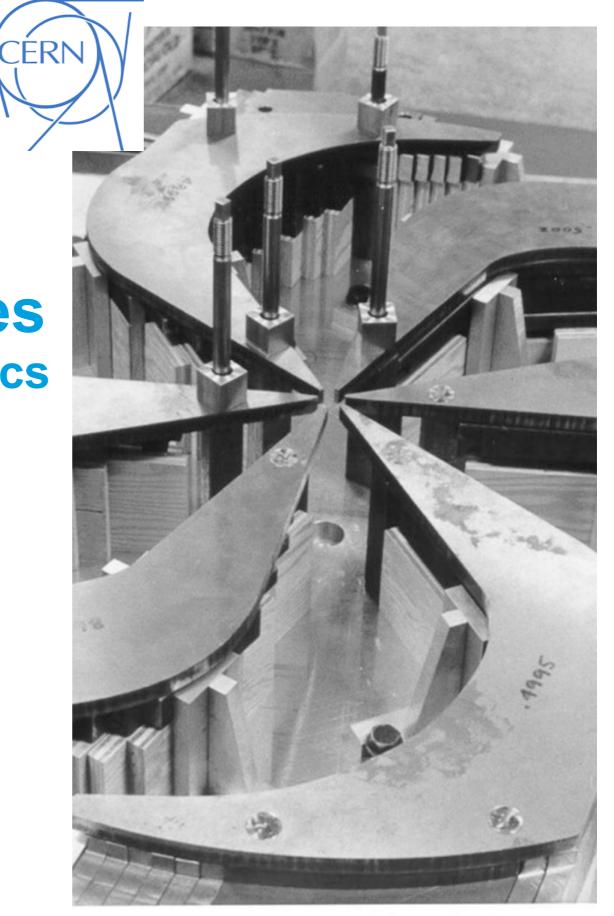




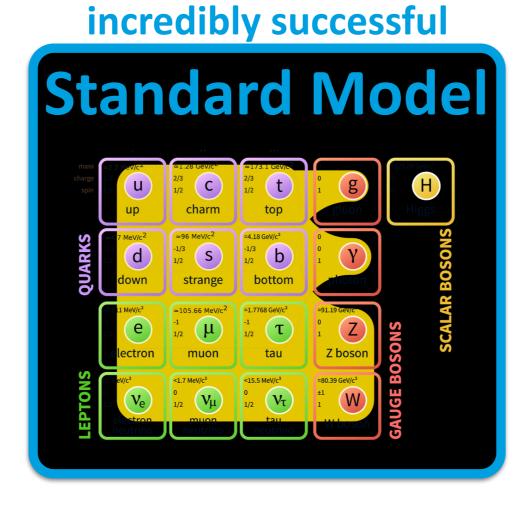
Radioactive Molecules Novel Probes for New Physics

Stephan Malbrunot-Ettenauer TRIUMF, University of Toronto, CERN

Humboldt Kolleg Kitzbühl 2022



Fundamental symmetries



Fundamental symmetries

matter-antimatter asymmetry

Antimatter

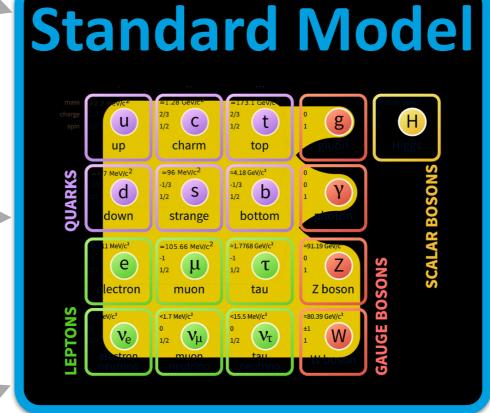
Antimatter

dark matter dark energy

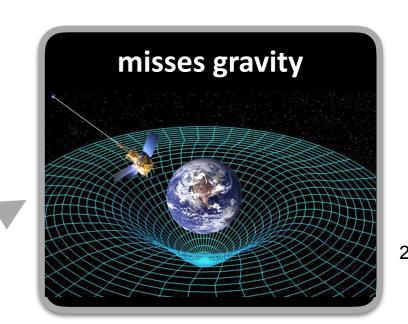
origin of neutrino masses

ve

incredibly successful



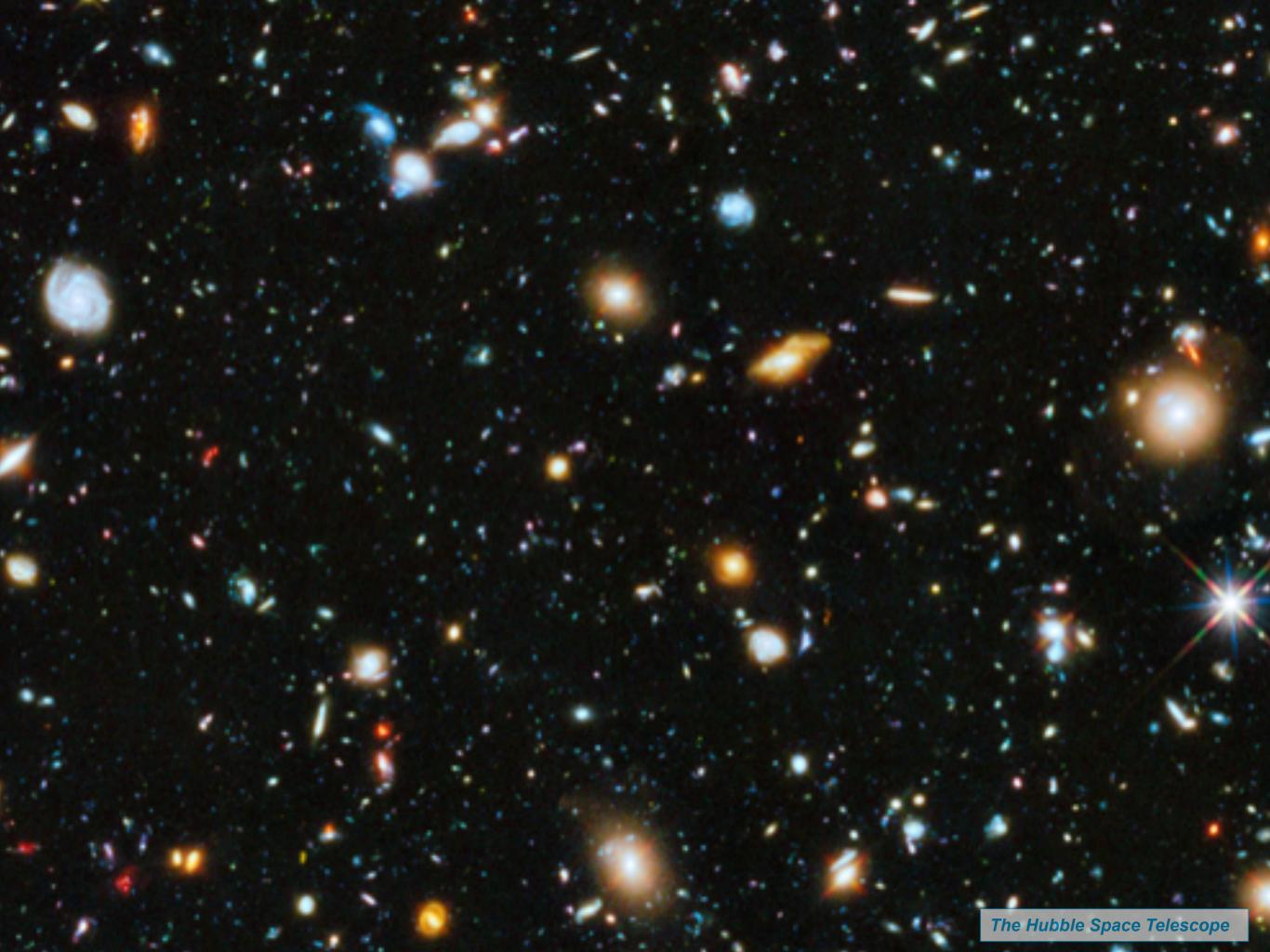
... yet incomplete



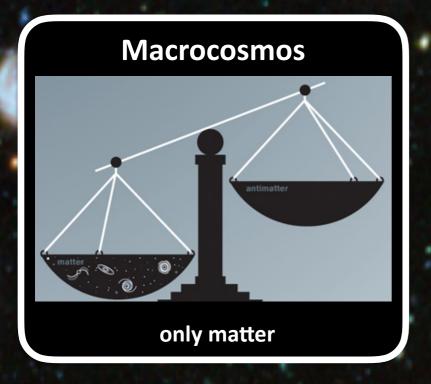
arbitrary constants:

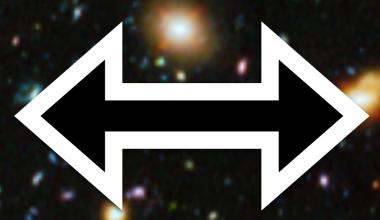
 m_e , m_μ , m_τ , m_u , m_d , m_s , m_c , m_b , m_t , m_H , θ_{12} , θ_{13} , θ_{23} , δ , g_1 , g_2 , g_3 , θ_{QCD} , V

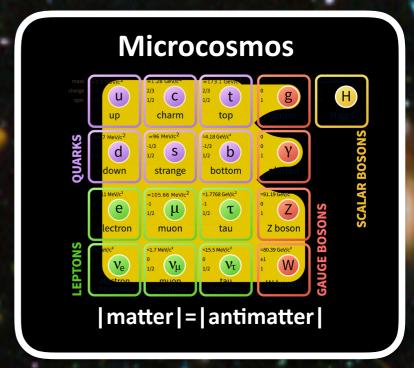
 $\mathbf{P}_{\theta_{12},\theta_{13},\theta_{23},\alpha_{1},\alpha_{2}}^{\mathbf{m}_{\nu_{e}},\mathbf{m}_{\nu_{\mu}},\mathbf{m}_{\nu_{\tau}}}$



where is all the antimatter?

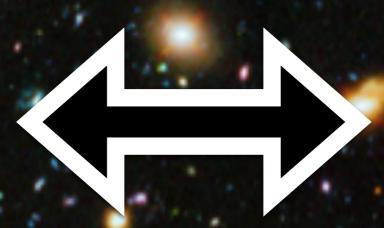


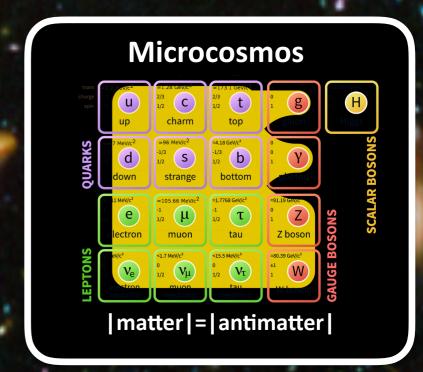




where is all the antimatter?

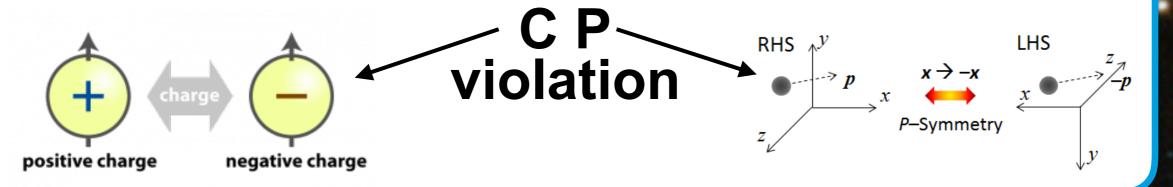






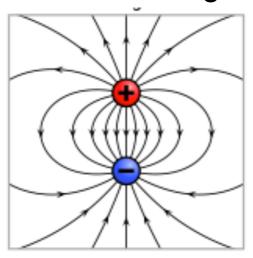
Sakharov, 1967

ingredient to resolve universe's matter-antimatter asymmetry:

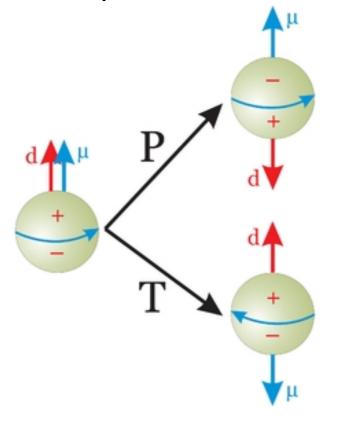


Permanent electric dipole moment

local separation of the electric charge along a particle's spin axis



• implies time-reversal (T) violation ⇒ violation of CP symmetry (assuming CPT)

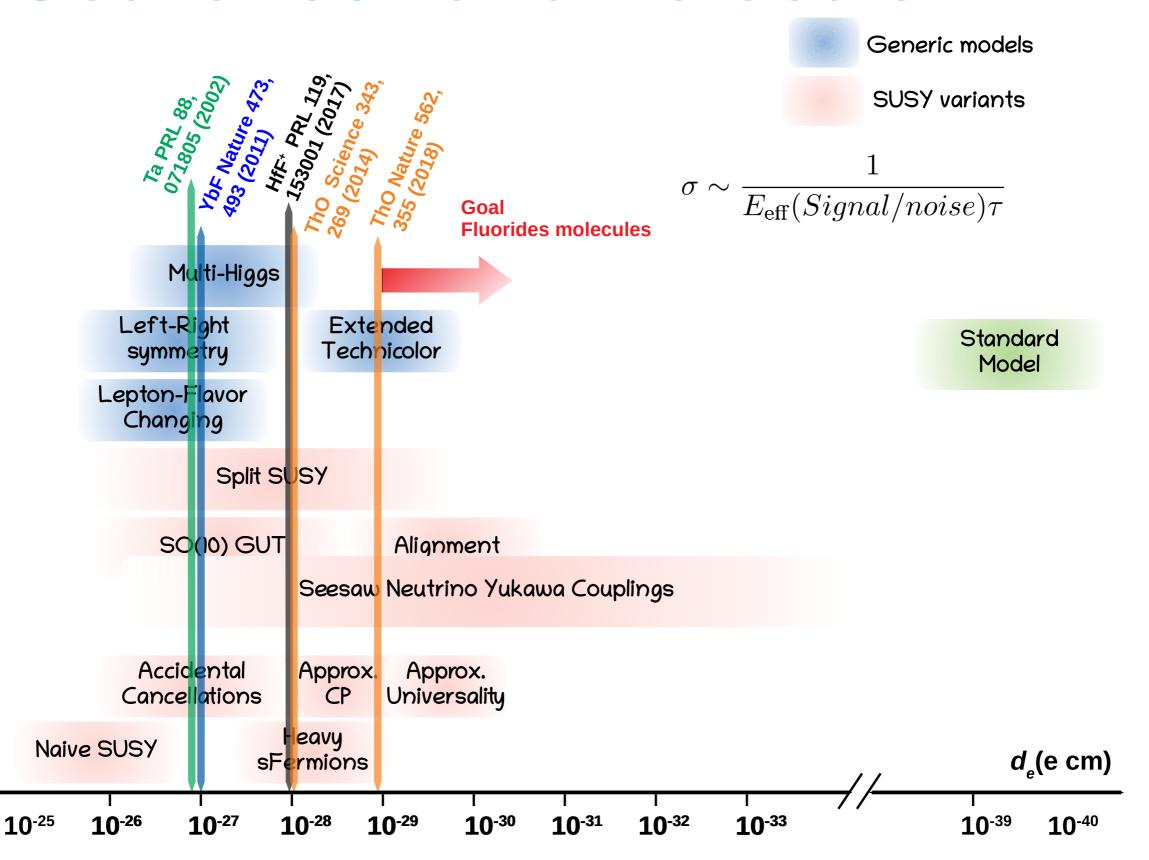




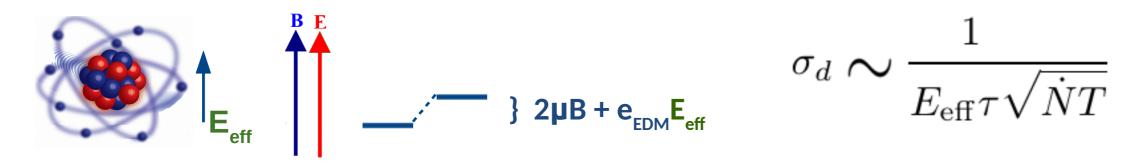
matter-antimatter asymmetry in the universe

1

Searches for an electron EDM

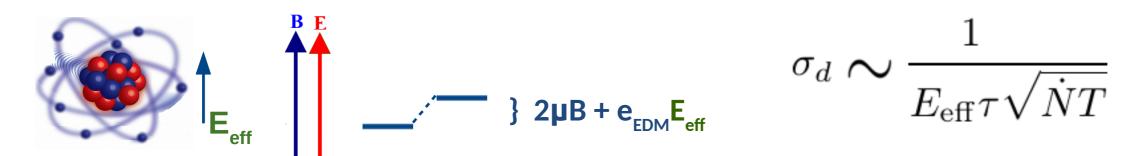


EDM searches in molecules

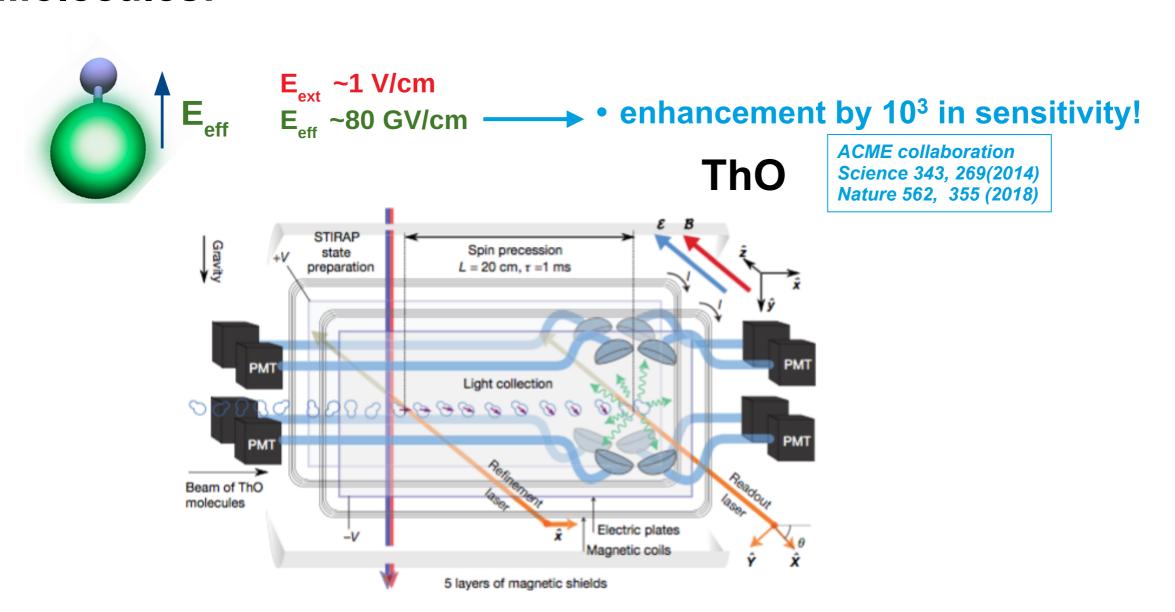


$$\sigma_d \sim \frac{1}{E_{\text{eff}} \tau \sqrt{\dot{N}T}}$$

EDM searches in molecules

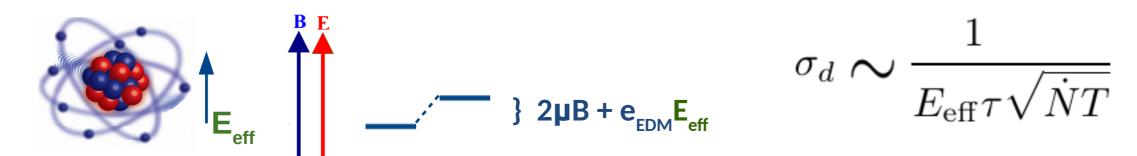


Molecules:

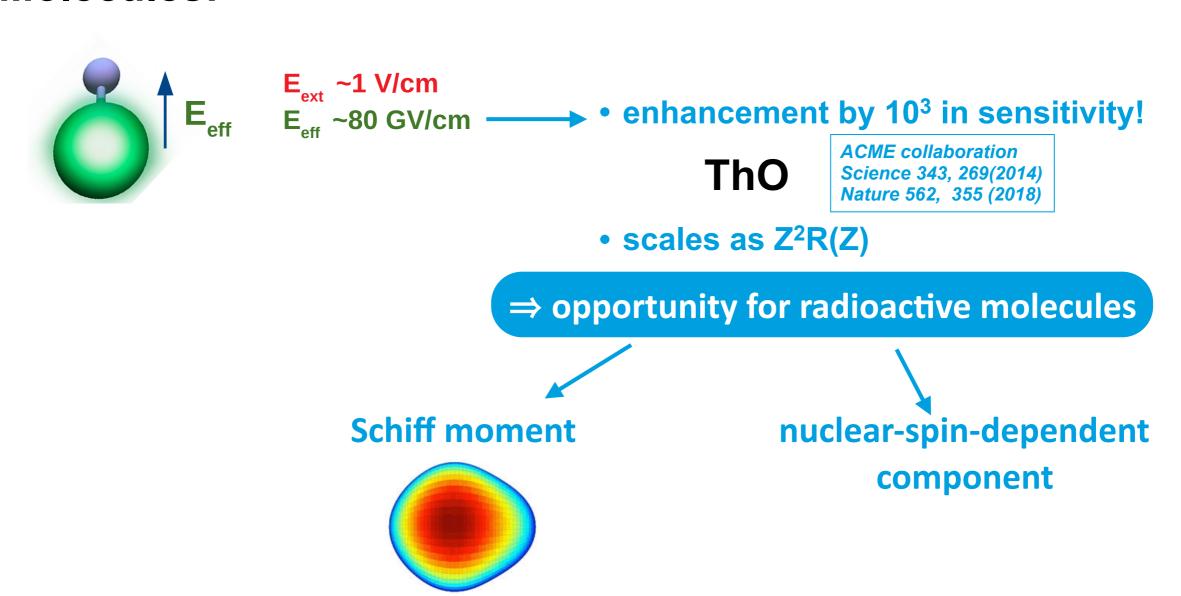


6

EDM searches in molecules



Molecules:



'Designer Molecules'

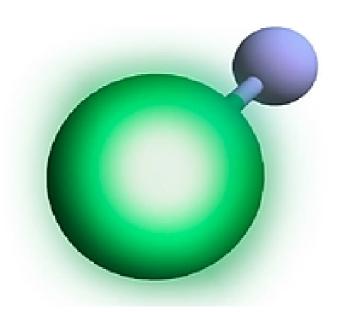
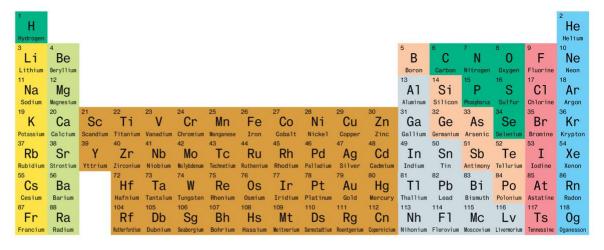
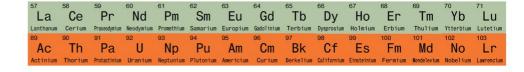


Table of Elements

80 chemical elements (with stable nuclides)



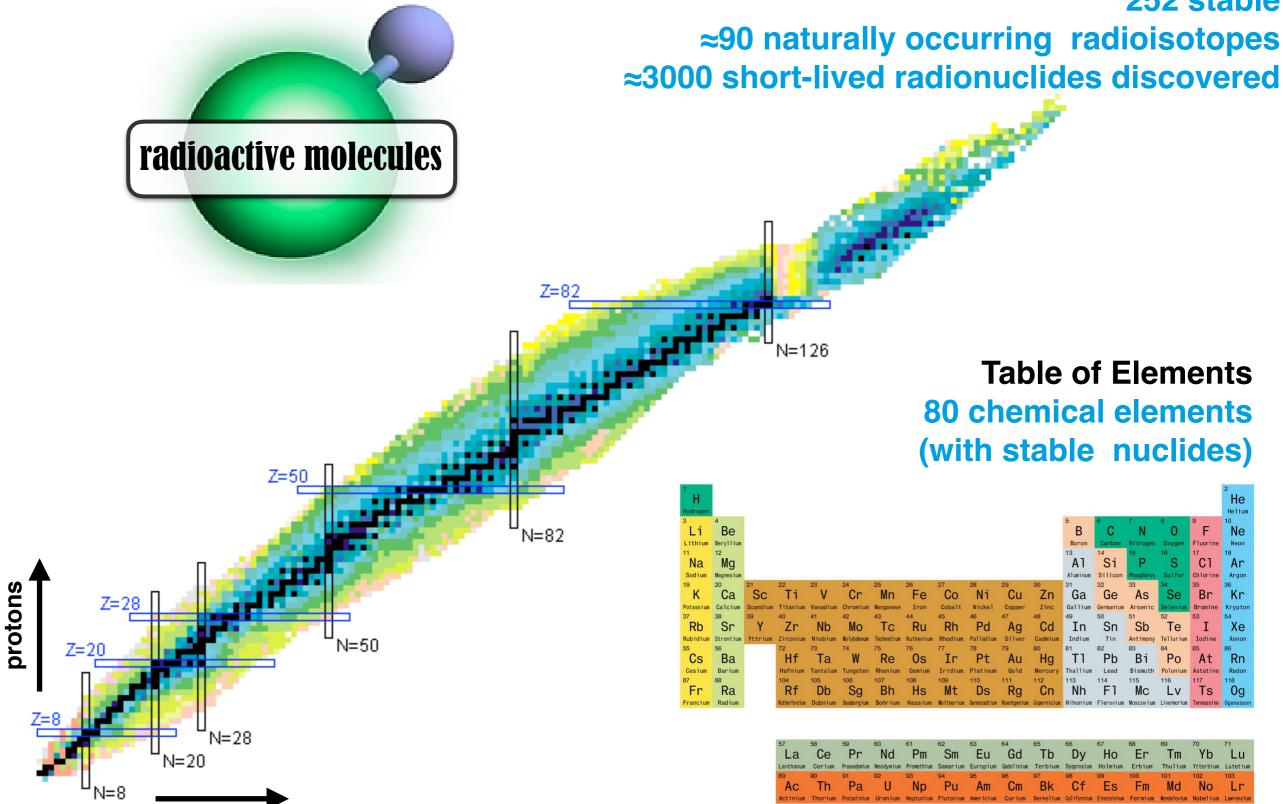


'Designer Molecules'

neutrons

Table of Isotopes

252 stable

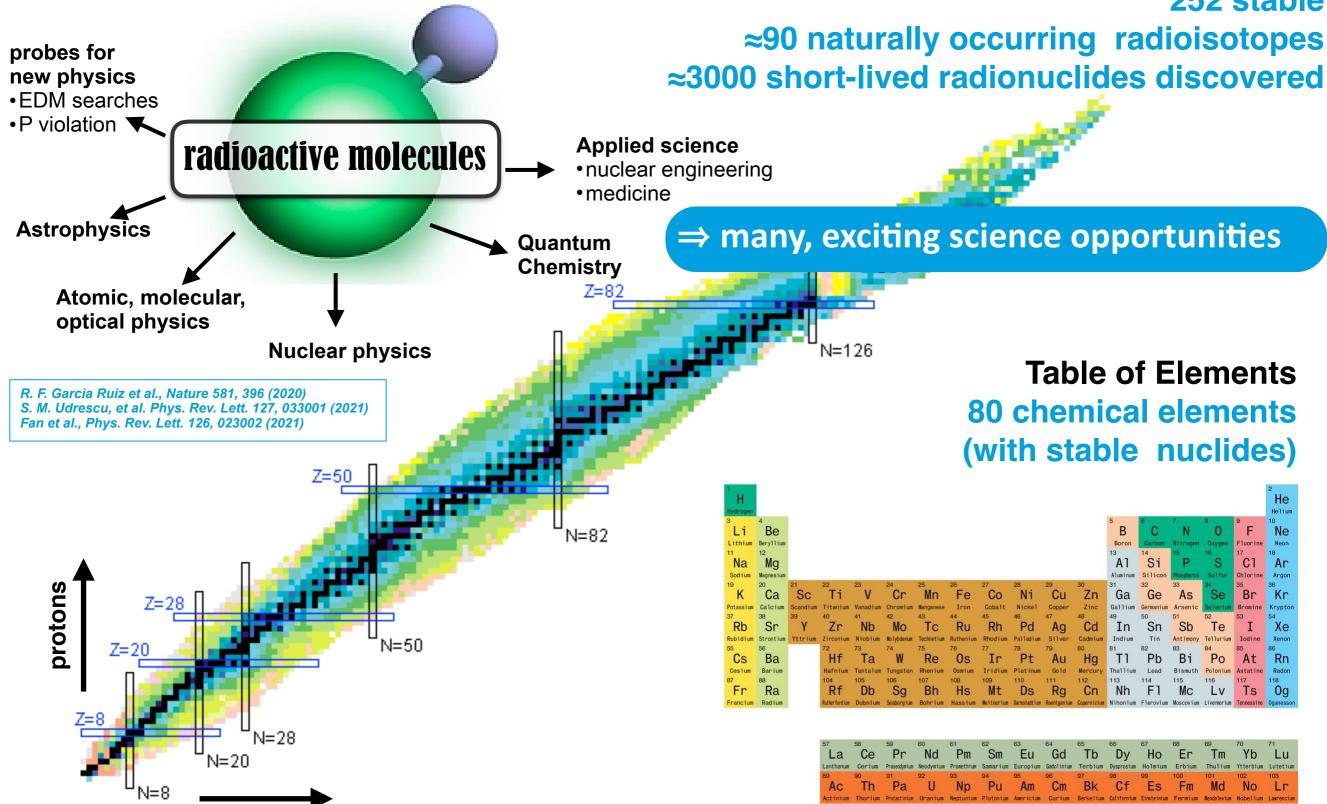


'Designer Molecules'

neutrons

Table of Isotopes

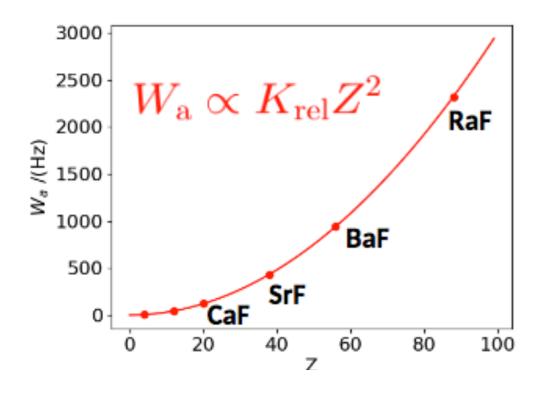
252 stable

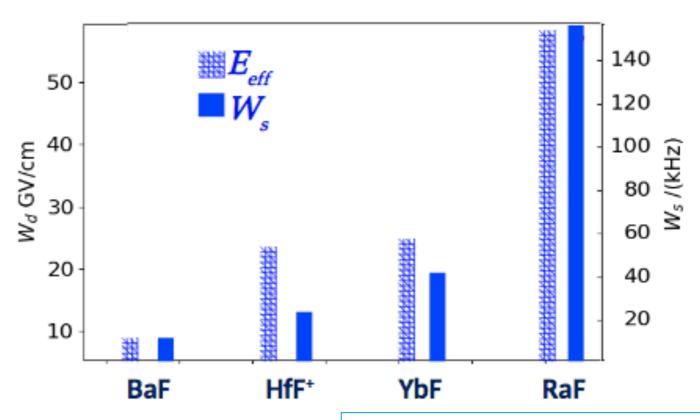


Sensitivity of Fluoride Molecules

$$\widehat{H}_{sr} = B\vec{\mathcal{N}}^2 + \gamma \, \vec{\mathbf{S}}^{eff} \cdot \vec{\mathcal{N}} + \vec{\mathbf{S}}^{eff} \cdot \widehat{\mathbf{A}} \cdot \vec{\mathbf{I}} + \vec{\mathcal{N}} \cdot \widehat{\mathbf{C}} \cdot \vec{\mathbf{I}} + \cdots
+ W_a (K_A/2) [\vec{\lambda} \times \vec{S}^{eff}] \cdot \vec{I} + (W_s k_s + E_{eff} d_e) \vec{\lambda} \cdot \vec{S}^{eff}$$

P-odd and P,T -odd effects





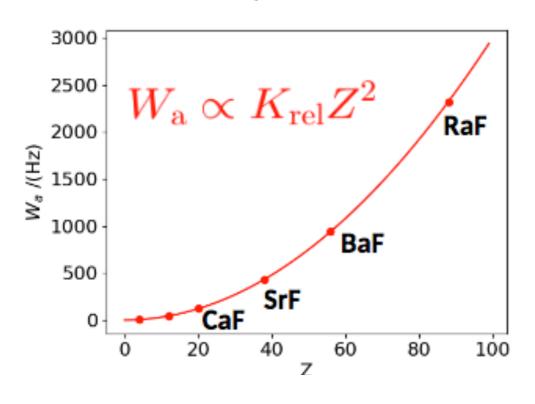
Gaul & Berger J. Chem. Phys 147, 014109(2017) Fleig. Phys. Rev. A 96, 040502 (2017)

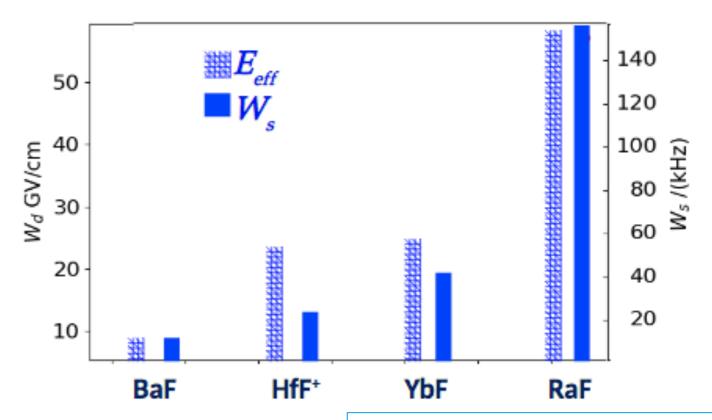
8

Sensitivity of Fluoride Molecules

$$\widehat{H}_{sr} = B\vec{\mathcal{N}}^2 + \gamma \, \vec{\mathbf{S}}^{eff} \cdot \vec{\mathcal{N}} + \vec{\mathbf{S}}^{eff} \cdot \widehat{\mathbf{A}} \cdot \vec{\mathbf{I}} + \vec{\mathcal{N}} \cdot \widehat{\mathbf{C}} \cdot \vec{\mathbf{I}} + \cdots
+ W_a (K_A/2) [\vec{\lambda} \times \vec{S}^{eff}] \cdot \vec{I} + (W_s k_s + E_{eff} d_e) \vec{\lambda} \cdot \vec{S}^{eff}$$

P-odd and P,T -odd effects





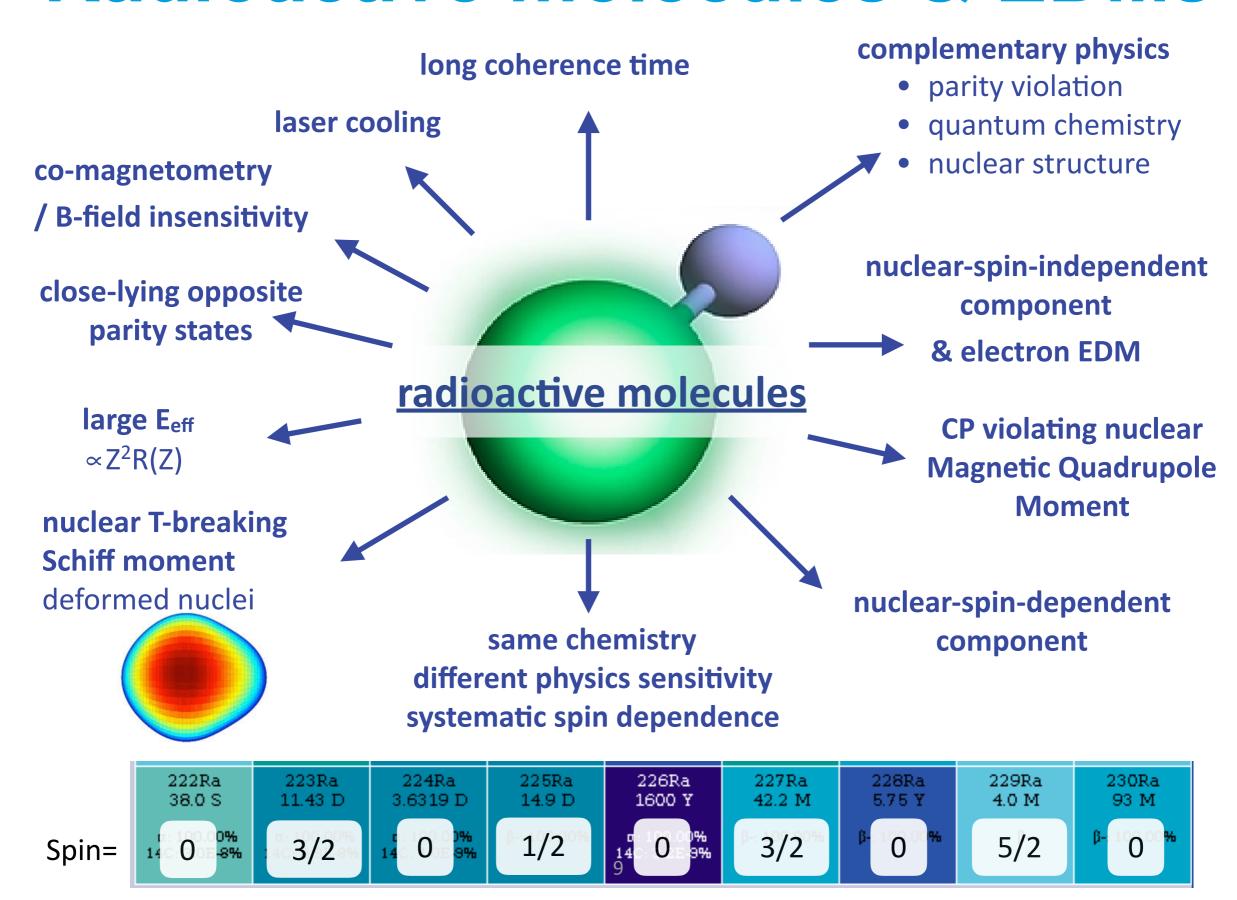
Nomen est omen: Radium is radioactive

Gaul & Berger J. Chem. Phys 147, 014109(2017) Fleig. Phys. Rev. A 96, 040502 (2017)

222Ra	223Ra	224Ra	225Ra	226Ra	227Ra	228Ra	229Ra	230Ra
38.0 S	11.43 D	3.6319 D	14.9 D	1600 Y	42.2 M	5.75 Y	4.0 M	93 M
α: 100.00% 14C: 3.0E-8%	α: 100.00% 14C: 8.9E-8%	α: 100.00% 14C: 4.0E-9%	β-: 100.00%	α: 100.00 % 14C: 3.2E-9 %	β-: 100.00%	β-: 100.00%	β-	β-: 100.00%

Radioactive molecules & EDMs

9



Examples of proposed radioactive molecules

Fundamental symmetries & new physics

RaF

Isaev et al., Phys. Rev. A 82, 052521 (2010)	Kudashov, Phys. Rev. A 90, 052513 (2014)	Garcia Ruiz et al., Nature 581, 396 (2020)
RaOH	Isaev et al., J. Phys. B: At. Mol. Opt. Phys. 50 225101 (2017)	Kozyryev et al., Phys. Rev. Lett. 119, 133002 (2017)
RaO	Flambaum, Phys Rev A 77, 024501 (2008).	
RaH	Fazil et al., Phys. Rev. A 99,052502 (2019).	
FrAg	PaF³⁺	C. Zülch et al., arXiv:2203.10333 (2022)

RaOCH₃⁺ Fan et al., Phys. Rev. Lett. 126, 023002 (2021)
Yu and Hutzler, Phys. Rev. Lett. 126, 023003 (2021)

short-lived ThO Flambaum, Phys. Rev. C 99 035501 (2019)

AcO⁺ Flambaum and V. A. Dzuba, Phys. Rev. A 101, 042504 (2020).

EuO⁺ Flambaum and V. A. Dzuba, Phys. Rev. A 101, 042504 (2020).

ThF⁺ C. F. Liang et al., Phys. Rev. C, 51:1199–1210, (1995)
N. J. Hammond et al., Phys. Rev. C, 65:064315 (2002)

D. N. Gresh et al., . J. Mol. Spectrosc., 319:1-9, (2016)

light nuclei for nuclear P-violation

Nuclear physics

KF- nuclear quadruple moments Teodoro et al., Phys. Rev. A 91, 032516 (2015)

CaCl- nuclear quadruple moments

nuclear anapole moments to study electroweak properties of short-lived nuclei

R. Garcia, personal communications

10

Astrophysics

Actinide molecules in nuclear engineering

26 T. Kamiński et al., Nat. Astron. 2, 778 (2018).

NpO, NpO₂

tal knowledge about radioactive molecules Examples of proposed radioactive mole

Fundamental symmetries & new physics

RaF

Isaev et al., Phys. Rev. A 82, 052521 (2010) Kudashov, Phys. Rev. A 90, 052513 (2014) Garcia Ruiz et al., Nature 581, 396 (2020)

Isaev et al., J. Phys. B: At. Mol. Opt. Phys. 50 225101 (2017) RaOH Kozyryev et al., Phys. Rev. Lett. 119, 133002 (2017)

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light nuclei for nuclear P-vi

Nuclear physics

KF- nuclear c

CaCl- pu re moments

moments to study electroweak properties of short-lived nuclei nu

R. Garcia, personal communications

Actinide molecules in nuclear engineering

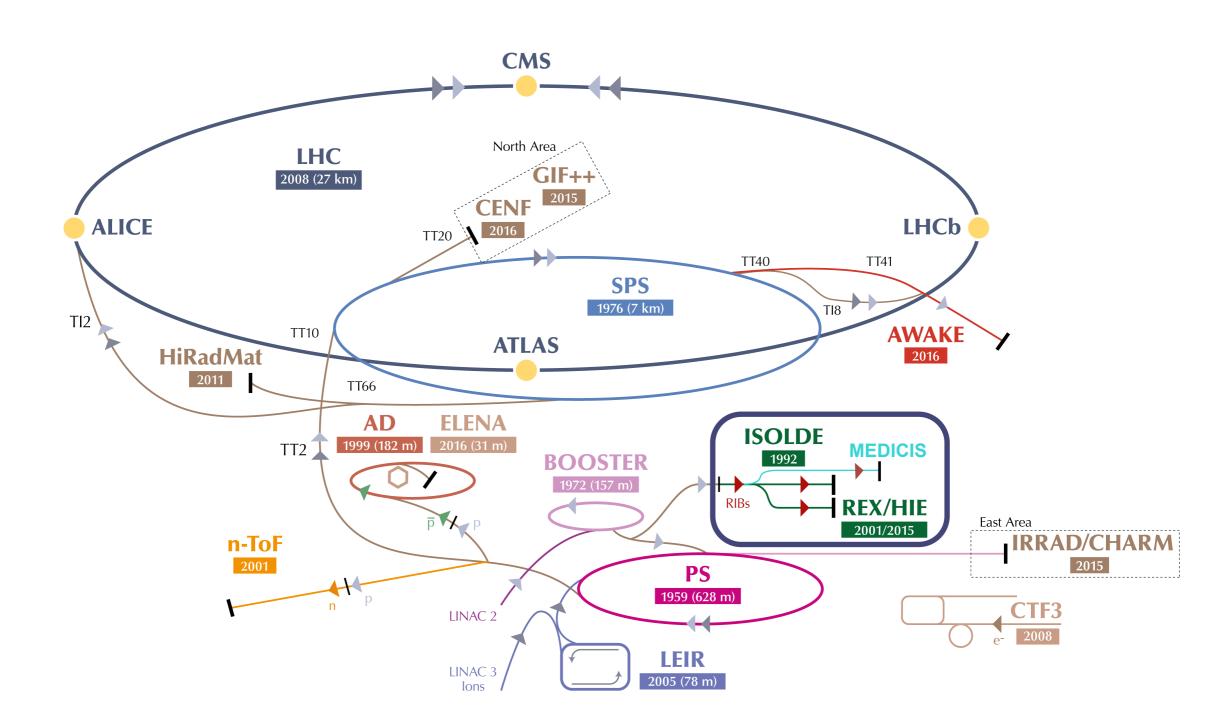
T. Kamiński et al., Nat. Astron. 2, 778 (2018).

NpO, NpO₂

10

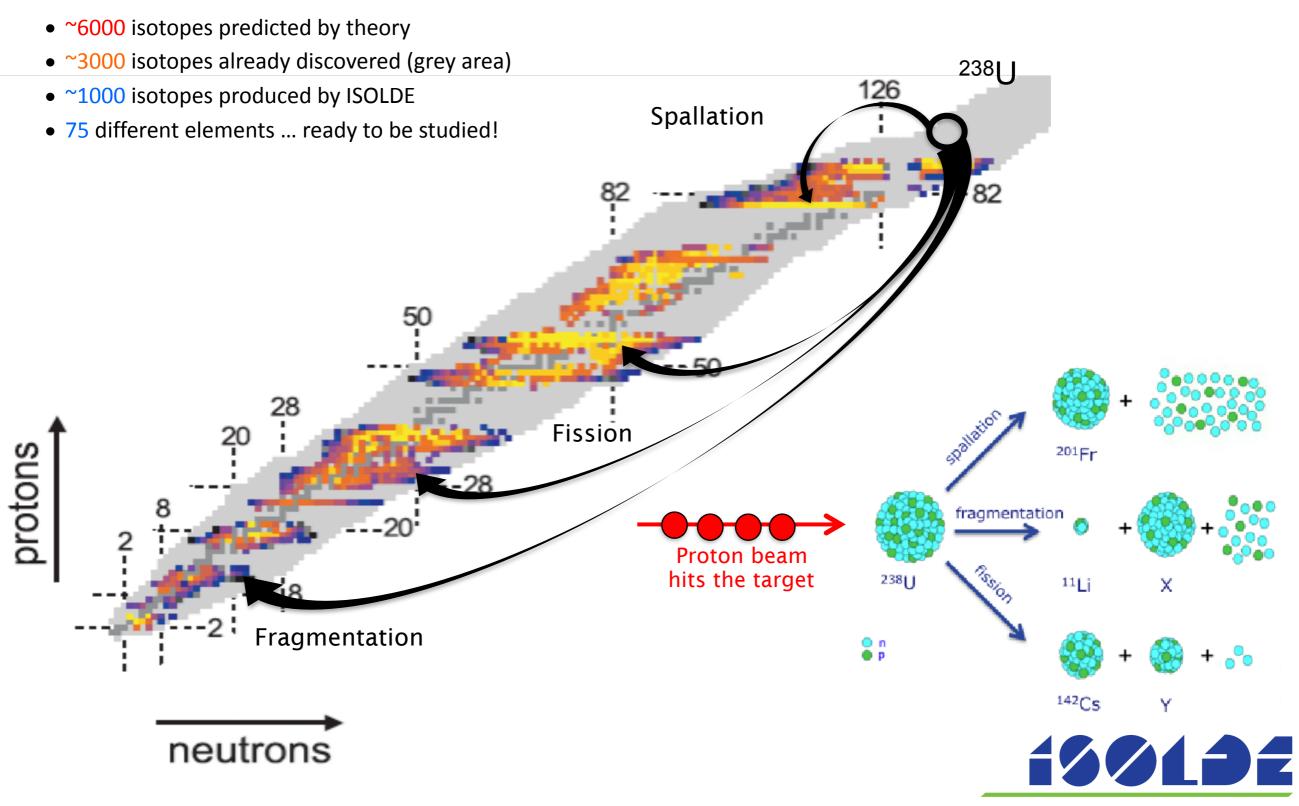
11

CERN accelerator complex

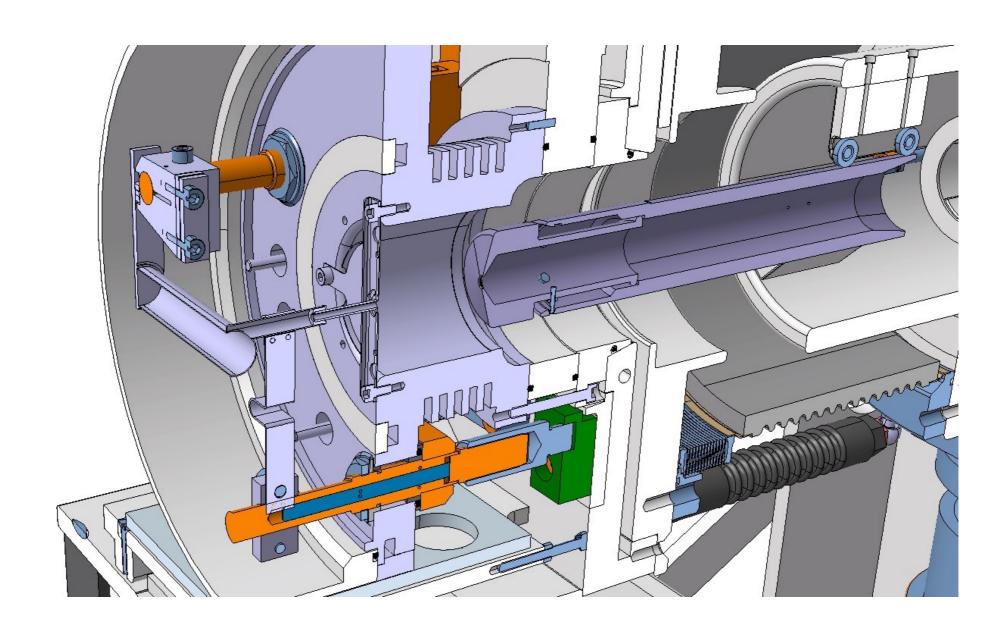


Isotope Production at ISOLDE

high energy (1.4 GeV) protons onto a thick target, e.g. ²³⁸U

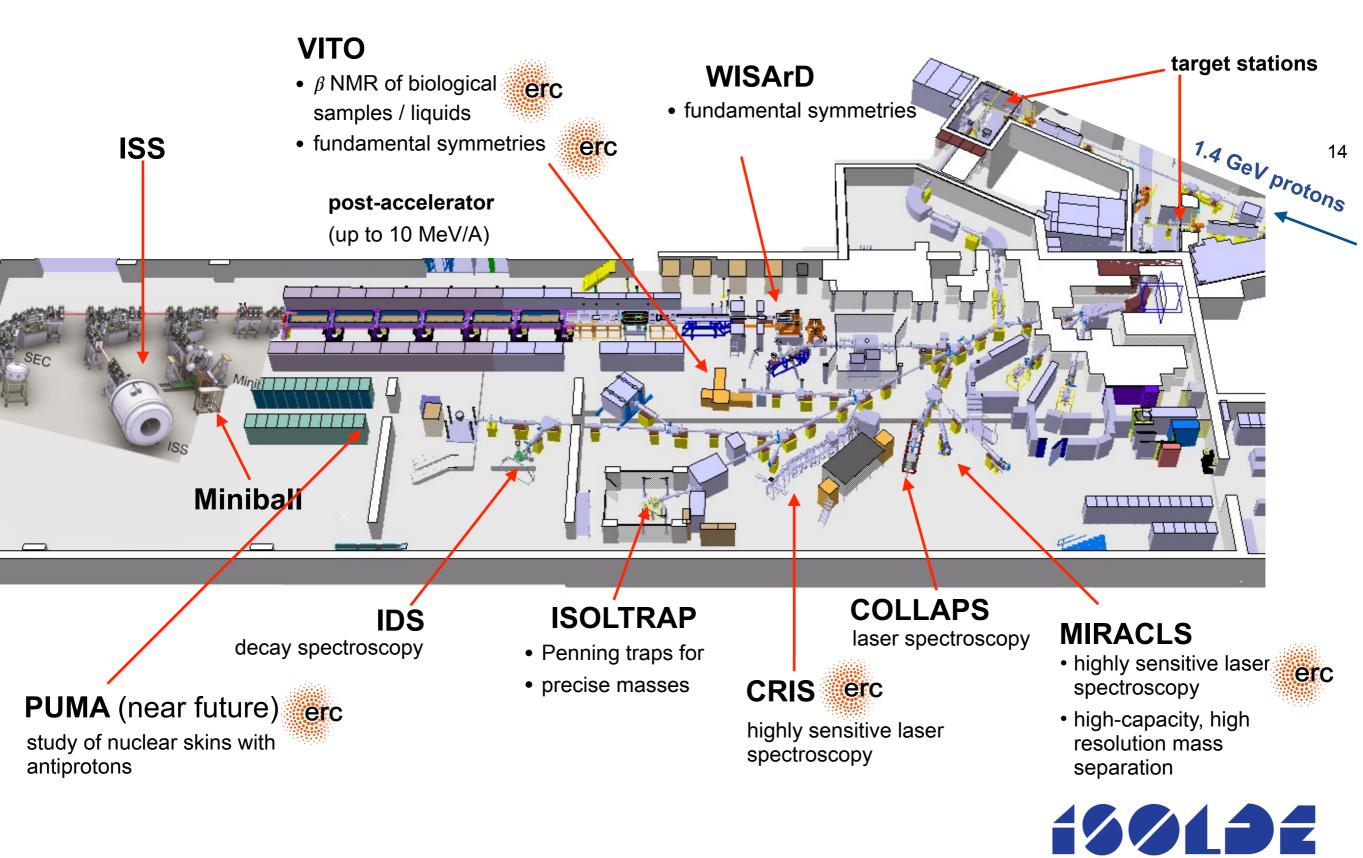


Rare Isotope Production

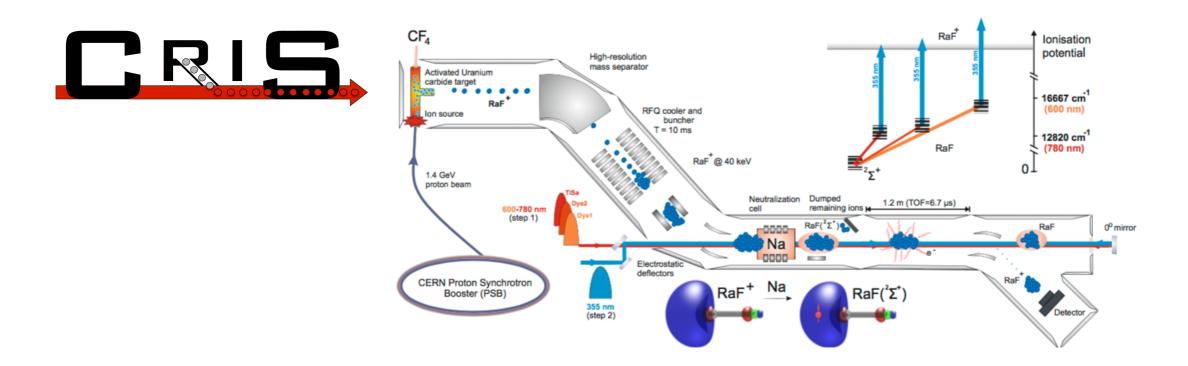


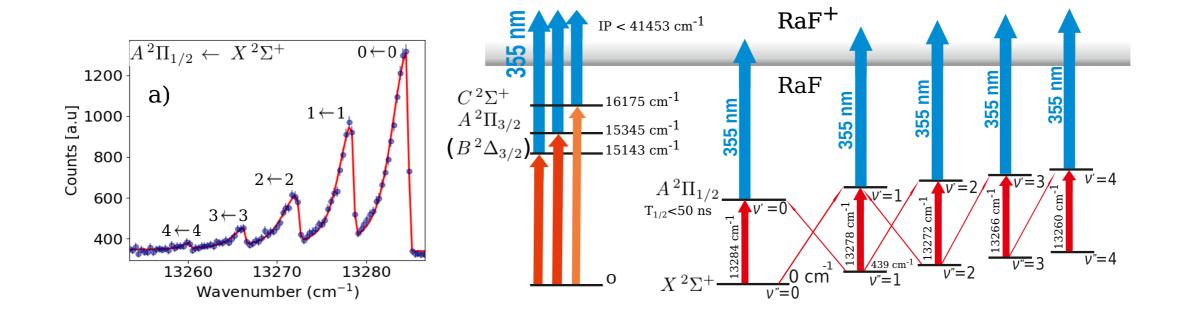


ISOLDE at CERN



First spectroscopy of radioactive molecules

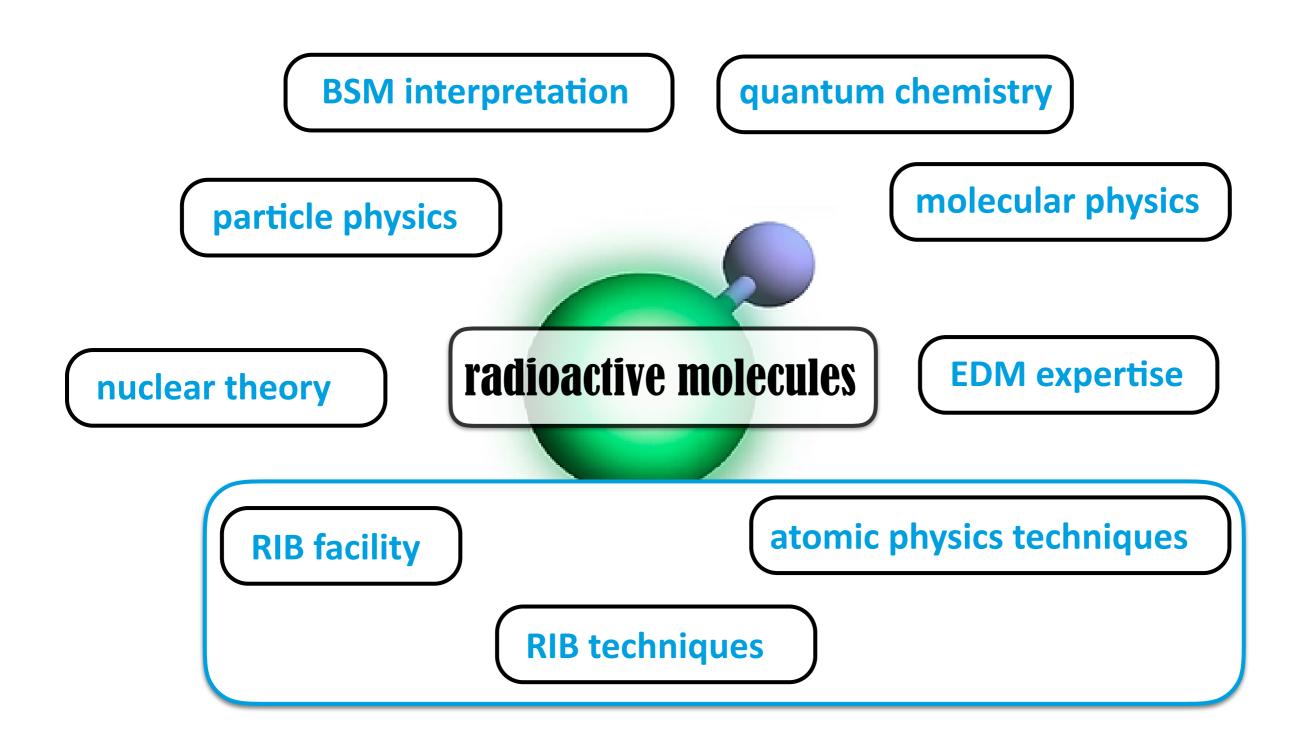




Multidisciplinary

quantum chemistry **BSM** interpretation molecular physics particle physics radioactive molecules **EDM** expertise nuclear theory atomic physics techniques **RIB** facility **RIB techniques**

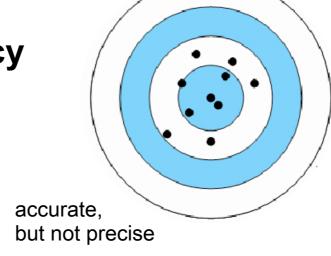
Multidisciplinary

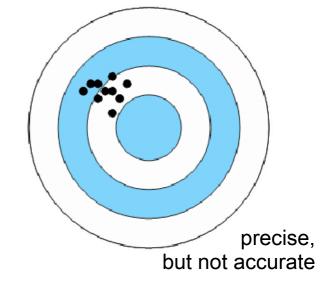


Atomic physics techniques at RIB facilities

high precision and accuracy

K. Blaum, et al., Phys. Scr. T152, 014017 (2013)
P. Campbell et al., Prog. Part. and Nucl. Phys. 86, 127-180 (2016)
J. Dilling et al., Annu. Rev. Nucl. Part. Sci. 68, 45 (2018)





ion traps

- masses
- RIB preparations
- mass separation
- in-trap decay

laser spectroscopy

- hyperfine structure
- isotope shifts
- optical pumping

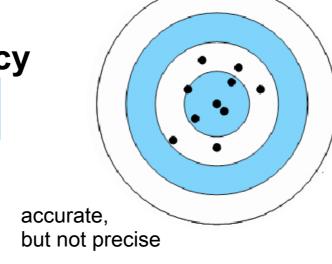
atom traps

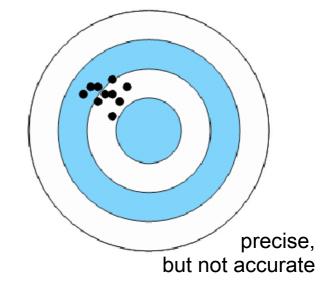
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- laser spectroscopy
- APV

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Challenges

short half-lives

 $T_{1/2} < 10 \text{ ms}$ ($\Delta m/m=6 \cdot 10^{-8}$)

M. Smith et al., PRL 101, 202501 (2008)

low intensity

masses: 0.5 ions / h

M. Block et al., Nature 463, 785 (2010) E. Minaya Ramirez et al., Science 337, 1207(2012)

temperature

buffer gas cooling (selected cases of laser cooling)

purity

R=m/ Δ m>5·10⁶ limited ion capacity

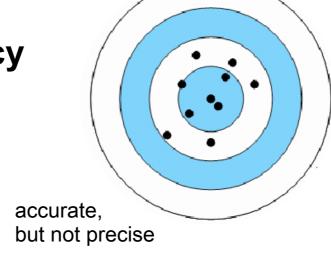
S. Eliseev et al., PRL 110, 082501 (2013)

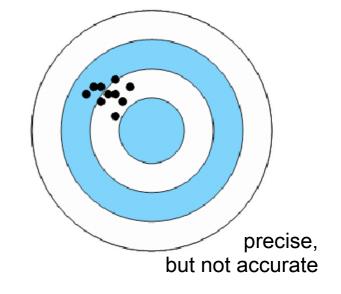
17

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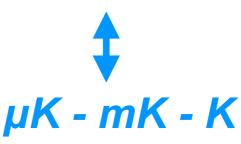
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temperature

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300 K



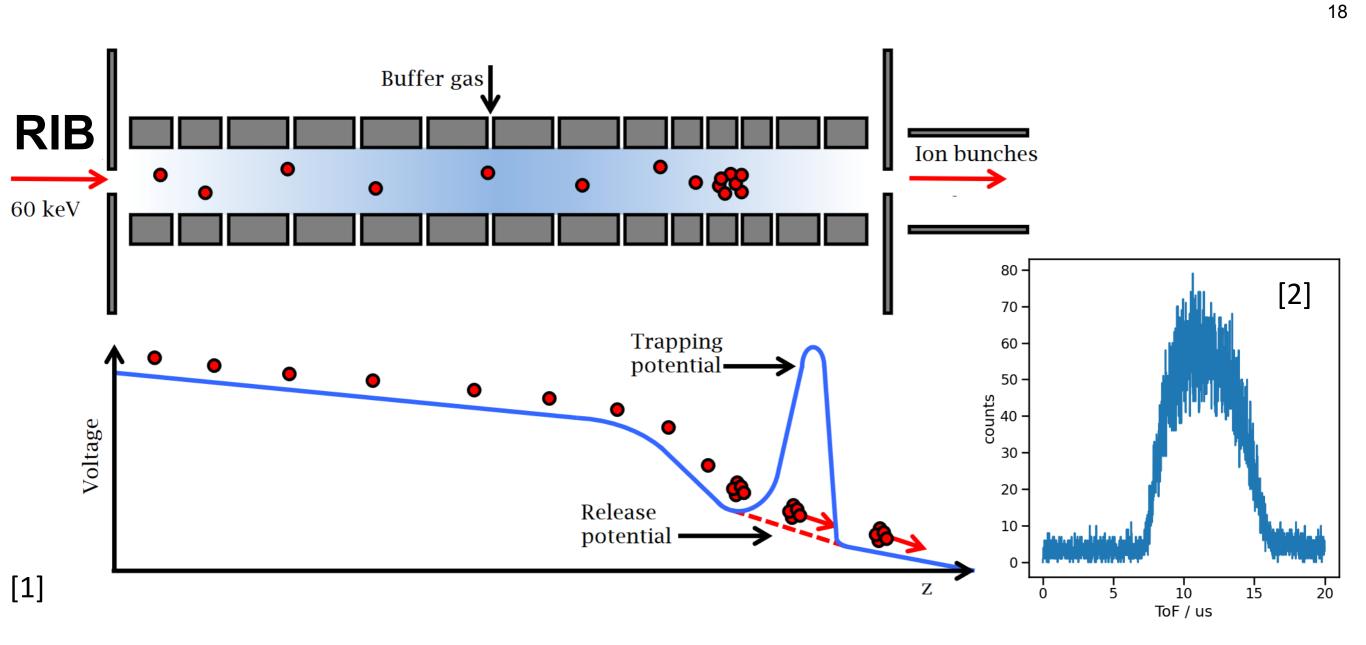
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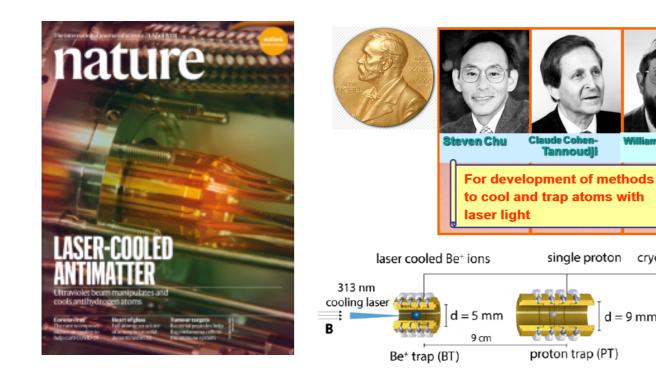
Standard buffer gas cooling

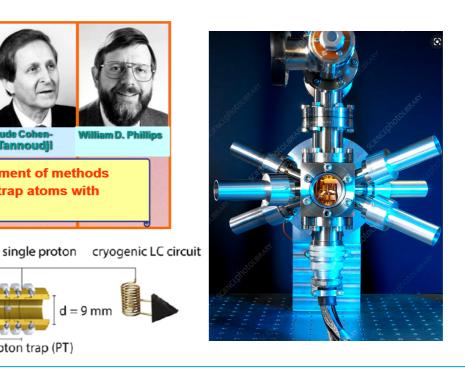
cooler and bunchers at RIB facilities , operated at 300 K buffer gas Cooling limit: 300 K



Doppler Cooling

- Powerful technique to reach sub-K atom and ion temperatures [1]
- Standard tool for high-precision measurements: atomic clocks [2], quantum information science [3], physics beyond the standard model [4]





[1] T. Haensch and A. Schawlow, Optics Communications 13, 68 (1975). D. J. Wineland and W. M. Itano, Phys. Rev. A 20, 1521 (1979). J. Eschner et al, J. Opt. Soc. Am. B20, 1003 (2003).

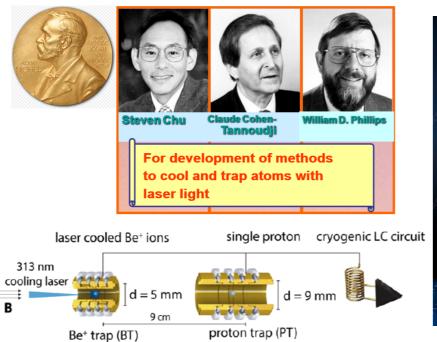
[2] D. Ludlow et al, Rev. Mod. Phys. 87, 637 (2015).

[3] C. D. Bruzewicz et al, Applied Physics Reviews 6, 021314 (2019). [4] M. S. Safronova et al, Rev. Mod. Phys. 90, 025008 (2018).

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J. Eschner et al, J. Opt. Soc. Am. B20, 1003 (2003).

[2] D. Ludlow et al, Rev. Mod. Phys. 87, 637 (2015).
[3] C. D. Bruzewicz et al, Applied Physics Reviews 6, 021314 (2019).
[4] M. S. Safronova et al, Rev. Mod. Phys. 90, 025008 (2018).

Specific applications with RIBs

G. D. Sprouse and L. A. Orozco, Annu. Rev. Nucl. Part. Sci.. 47, 429 (1997)
J. A. Behr et al., Phys. Rev. Lett. 79, 375 (1997).
M. Trinczek et al., Phys. Rev. Lett. 90, 012501 (2003).
L. B. Wang et al., Phys. Rev. Lett. 93, 142501 (2004).

P. A. Vetter et al., Phys. Rev. C 77, 035502 (2008).
J. R. A. Pitcairn et al., RRC 79, 015501 (2009)
A. Takamine et al., Phys. Rev. Lett. 112, 162502 (2014)
B. Fenker et al., Phys. Rev. Lett. 120, 062502 (2018)

• unexplored as cooling technique to deliver high quality (molecular) RIBs

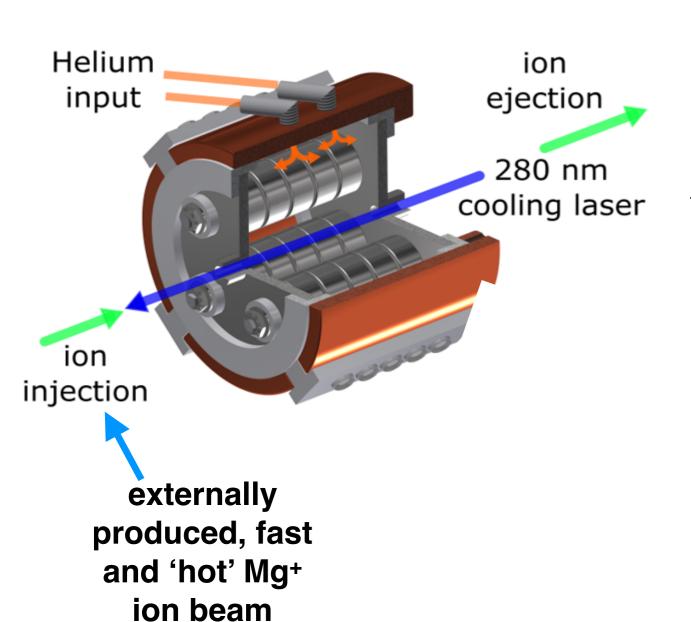
Goal: provide ultra-cold (molecular) RIBs

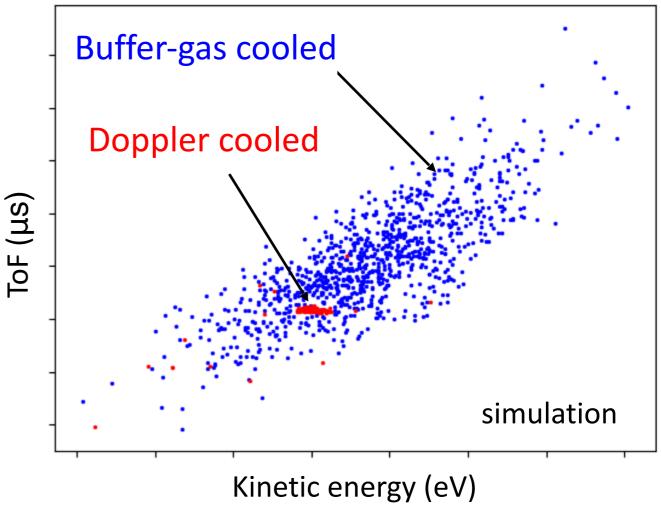
- ... compatible with short half-lives
- ... universally applicable (via sympathetic cooling)

Experimental Demonstration at MIRACLS



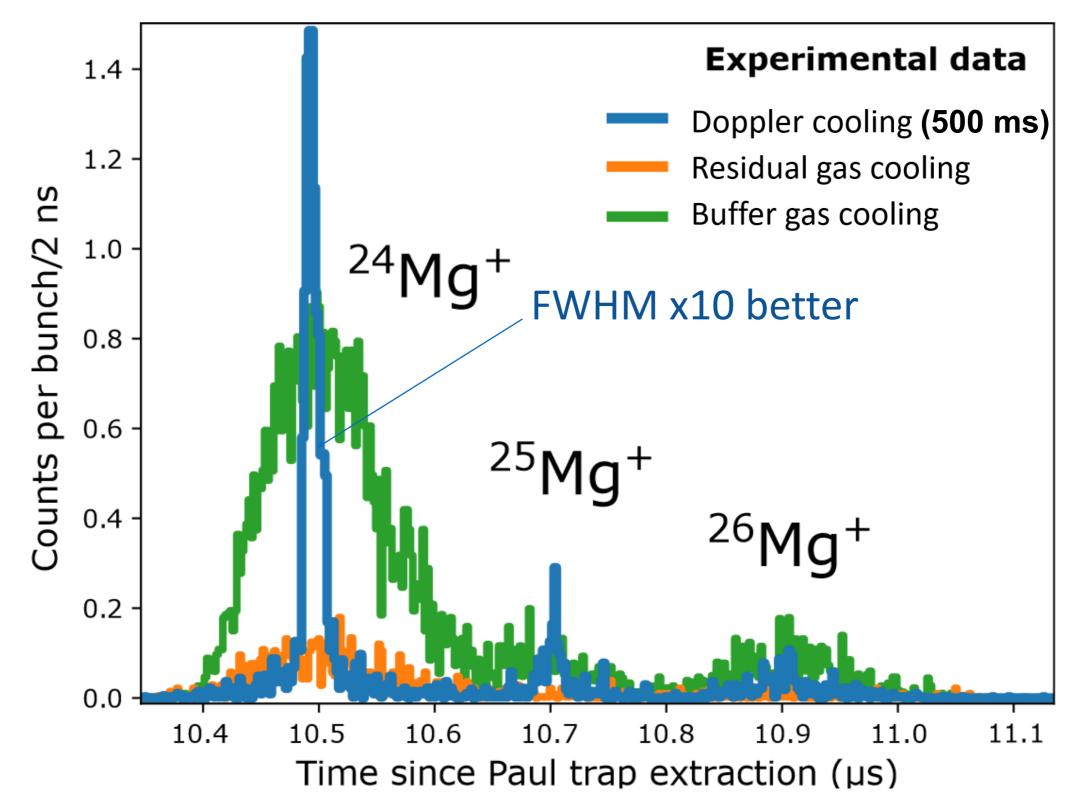
Paul trap:





Experimental results MIRACLS

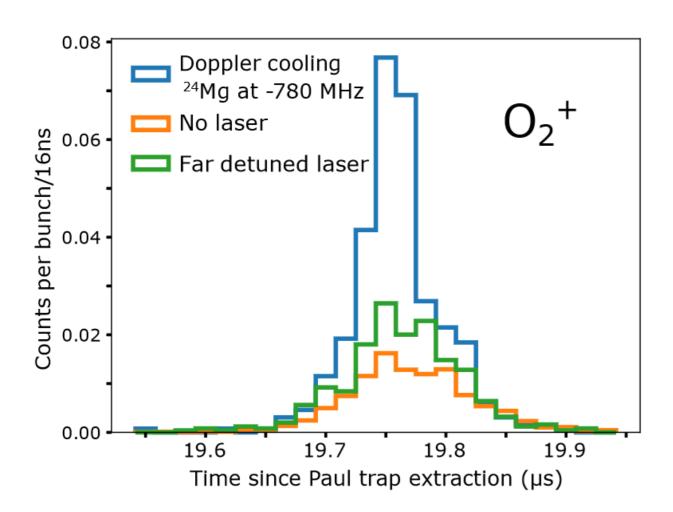




Sympathetic cooling MIRACLS



- 'universal' availability of cold ion ensembles
- · including ionic systems which cannot be directly laser-cooled



opportunity for cold molecular RIBs

	O ₂ ⁺		
Peak width residual-gas or buffer-gas cooling	113(5) ns		
Sympathetic cooling	58(4) ns		
Improvement in countrate	Factor 2.6		

S. Sels, F. Maier et al., accepted in Phys. Rev. Research

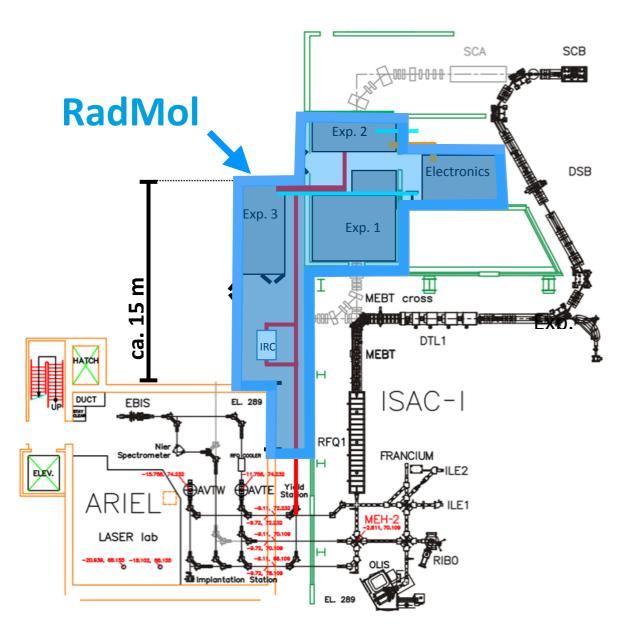
Can be done better analogous to existing work, e.g. [1],[2]

J. Wuebbena et al, Phys. Rev. A 85, 043412.2012. [2] M. Guggemos. New Journal of Physics *17*, *103001*, *2015*.

RadMol & TRIUMF



a radioactive molecule lab for fundamental physics



RadMol Collaboration















Goals:

world-wide unique laboratory

for radioactive molecules

searches for new physics

precision studies for

TRIUMF advantages

beams (RIB)

online facility

independent RIBs)

multi-station program

• large variety in radioactive ion

• high beamtime availability (3

existing laboratory space for large,

fast connection of RadMol lab to







u Ottawa





Summary

- Radioactive Molecules
 - entirely new science path
 - → intriguing&unexplored probes for New Physics



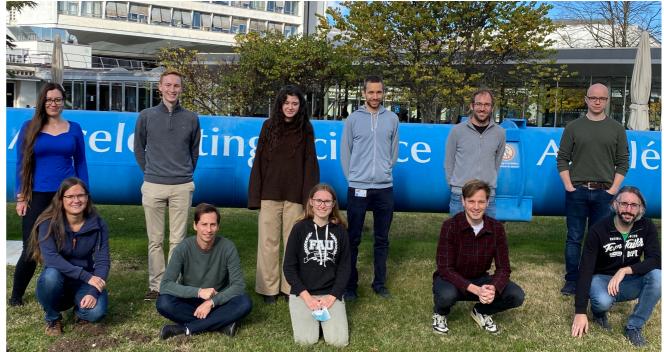
- RadMol
 - → dedicated laboratory for radioactive molecules & precision studies at TRIUMF
 - designed to master experimental challenges
- Cold radioactive, molecular beams
 - Doppler + sympathetic cooling





25





team members:

- I. Belosevic, L. Croquette, P. Fischer,
- C. Kanitz, F. Hummer, E. Leistenschneider,
- S. Lechner, F. Maier, P. Plattner, A. Roitman, M. Rosenbusch, S. Sels, F. Wienholtz,
- M. Vilen, R. Wolf, F. Buchinger, W. Nörtershäuser, L. Schweikhard, S. Malbrunot- Ettenauer

collaboration:







funding:









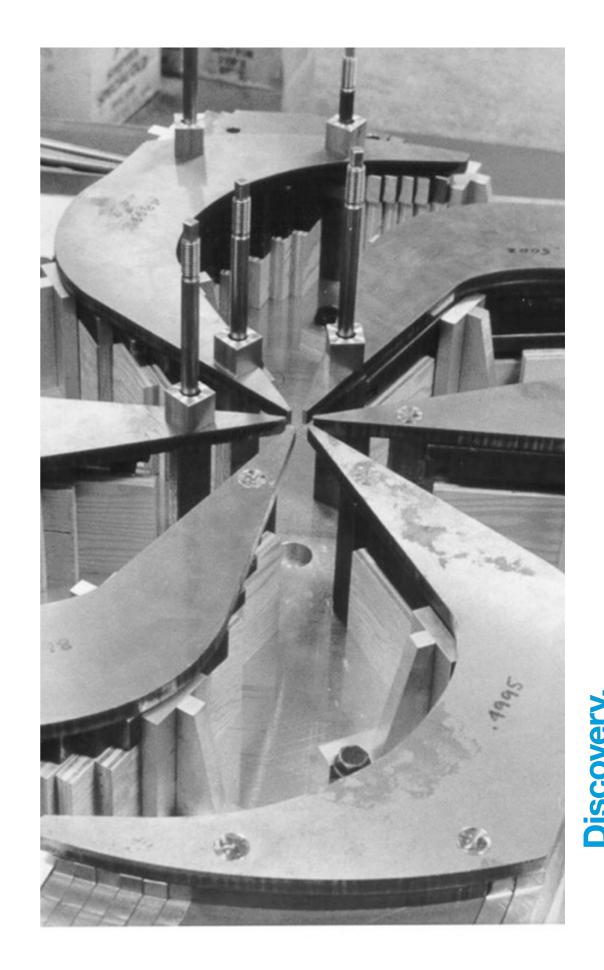
26



Thank you Merci

www.triumf.ca

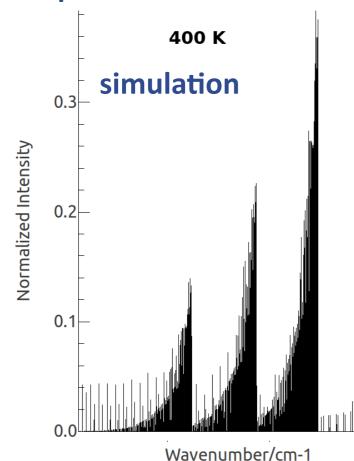
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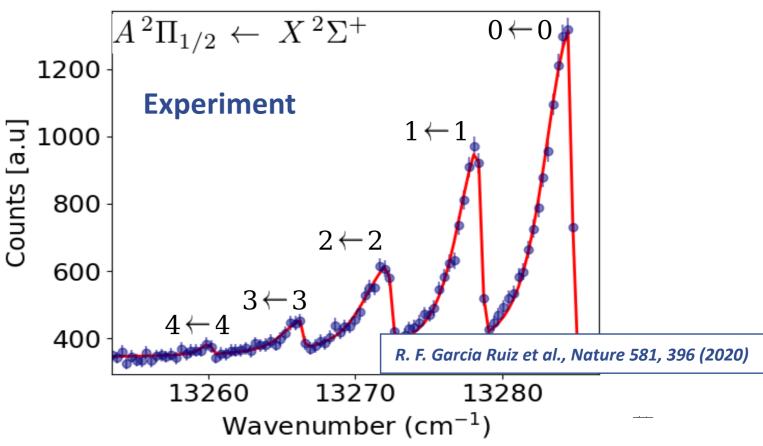


laser spectroscopy of molecules

- cooling of internal degrees of freedom
 - → higher population of the low-lying states
 - **⇒** simpler spectra ⇒ more easily identification
- buffer-gas cooling in cryogenic Paul trap:
 - → overall the gain could be more than x100 in scanning time.
 - ⇒ enables efficient initial state preparation for later EDM searches

RaF spectra





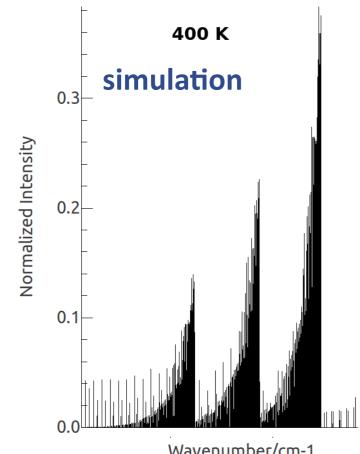
S. Malbrunot-Ettenauer: SMI 2022 simulation by R. F. Garcia Ruiz

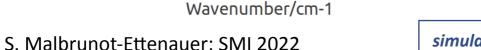


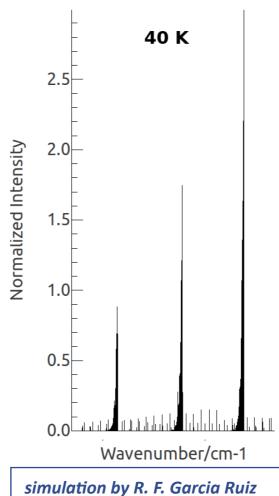
laser spectroscopy of molecules

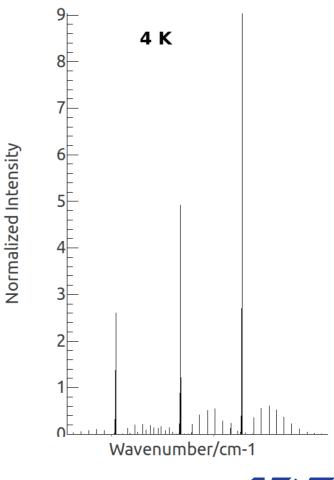
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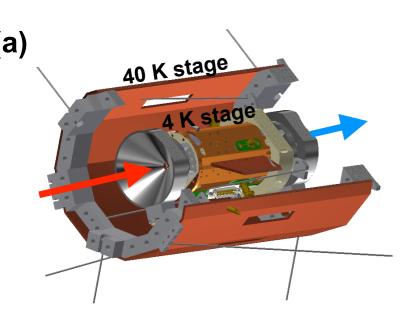


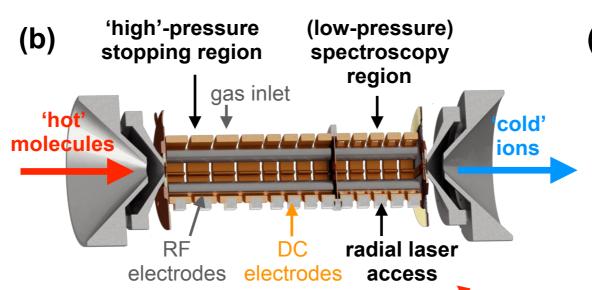


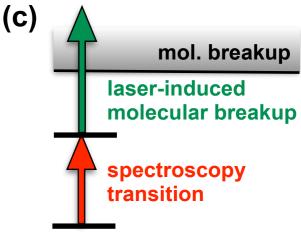


MIRACLS & rad. molecules

a cryogenic Paul trap for cooling and spectroscopy of of (ionic) radioactive molecules









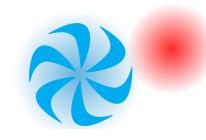
- higher population of the low-lying states
- → higher resolution (compared to RT-trap)
- high experimental sensitivity for trapped ions
- → MIRACLS' MR-ToF method for search for laser-cooling transitions in ionic molecules

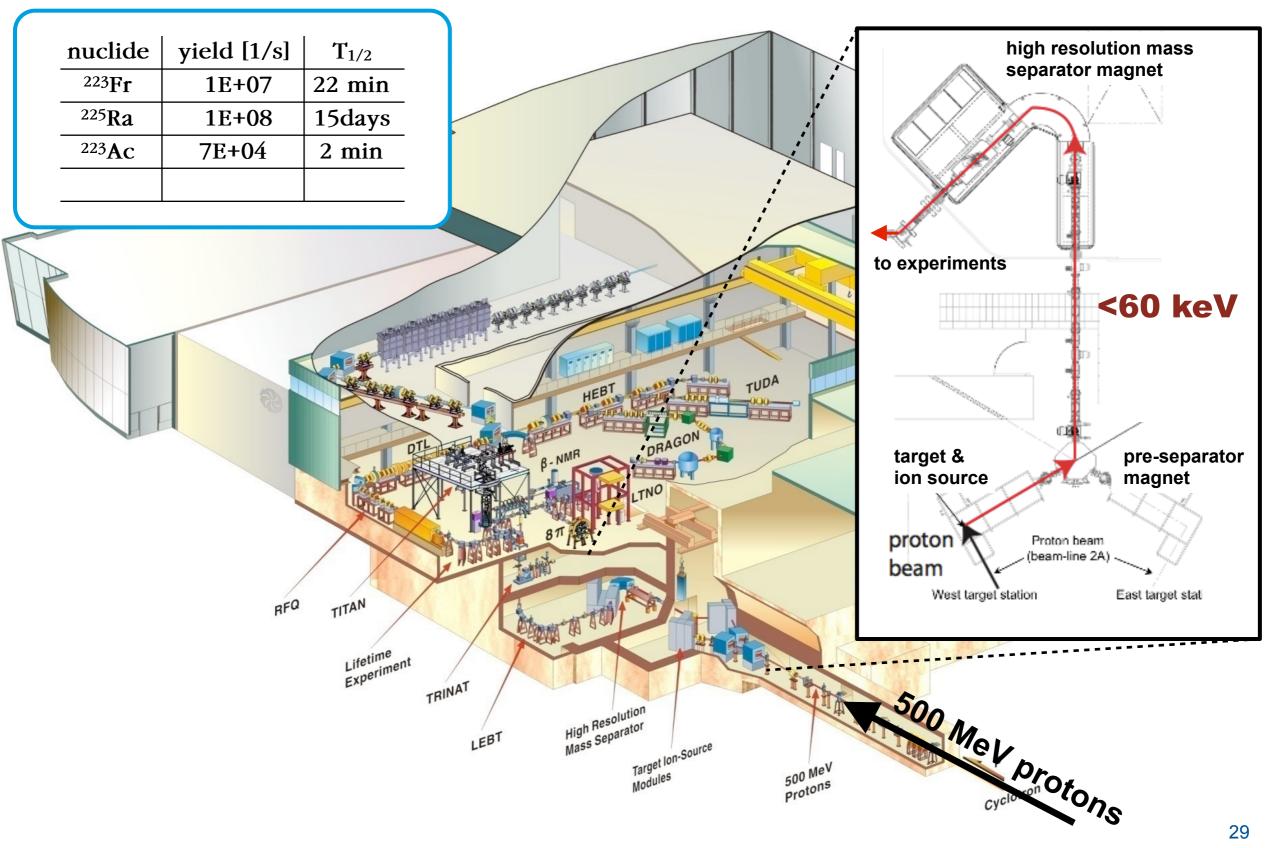
laser access for,
spectroscopy,
state preparations and
laser cooling

S. Sels., F. Maier et al., submitted



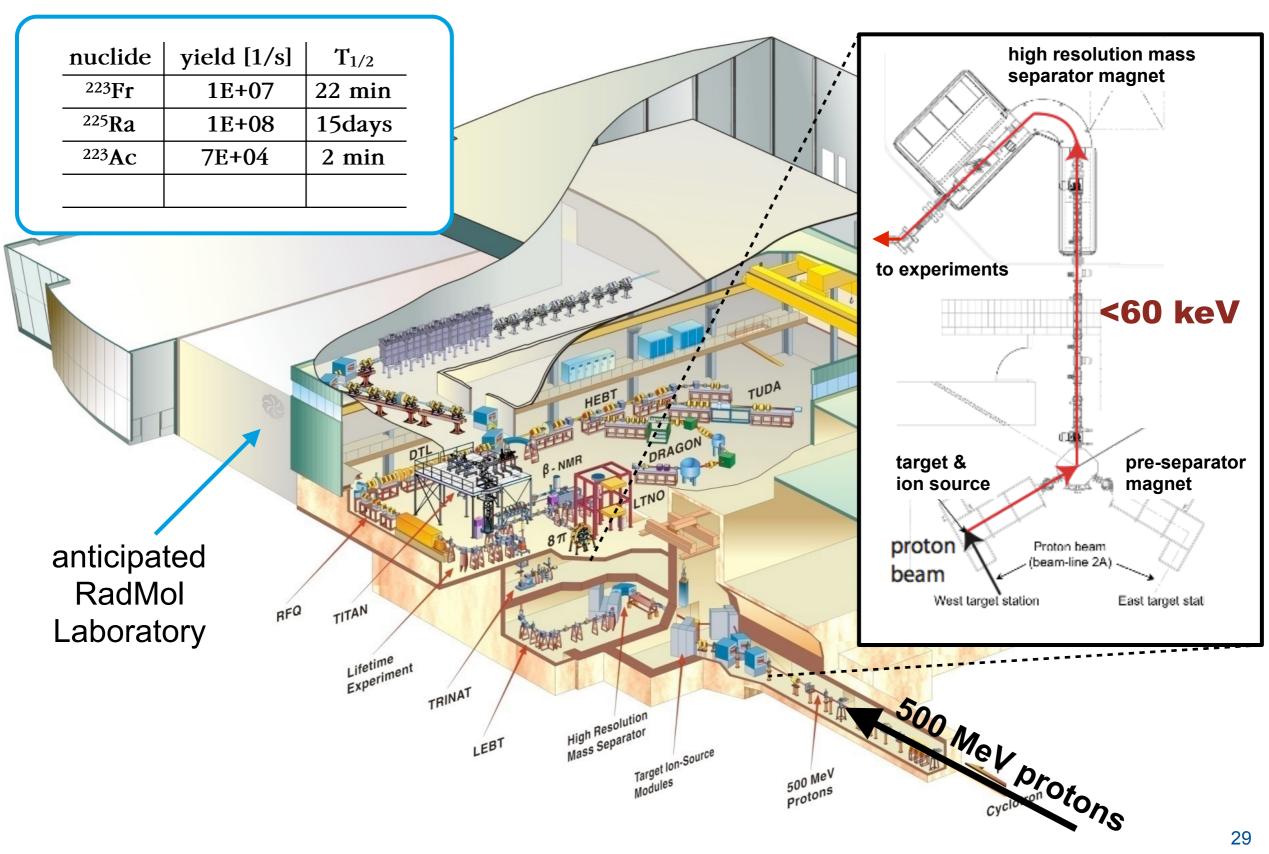
Radioactive Ions & TRIUMF





Radioactive Ions %TRIUMF

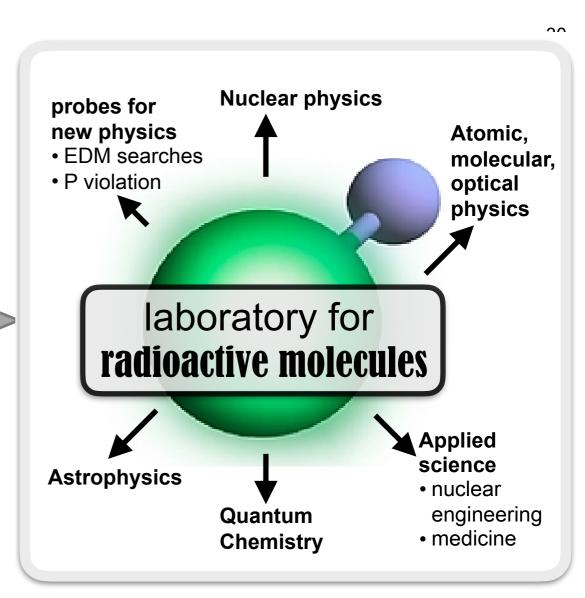




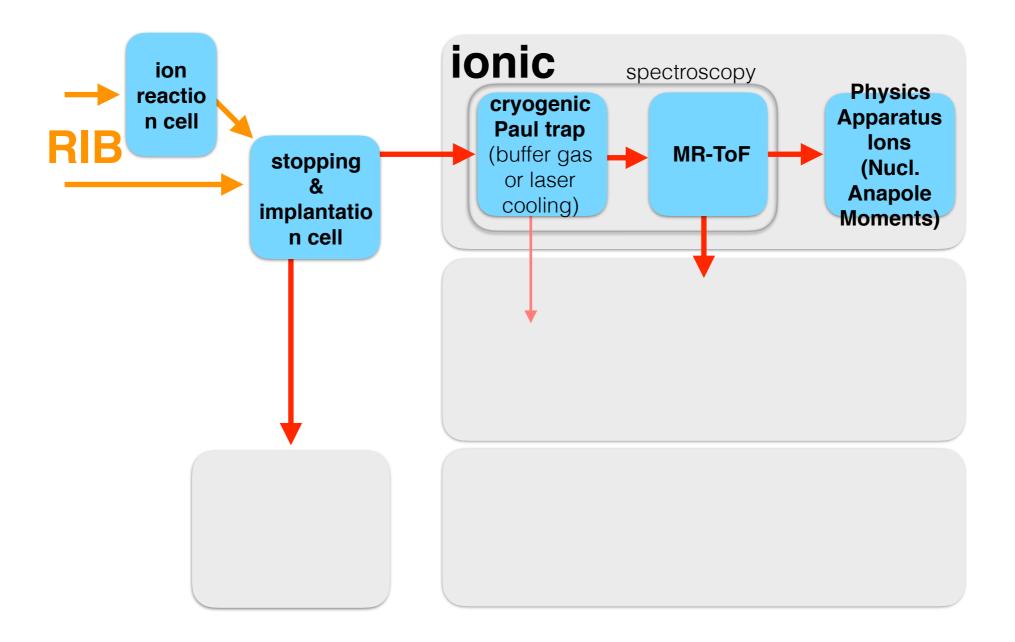
RadMol @ TRIUMF

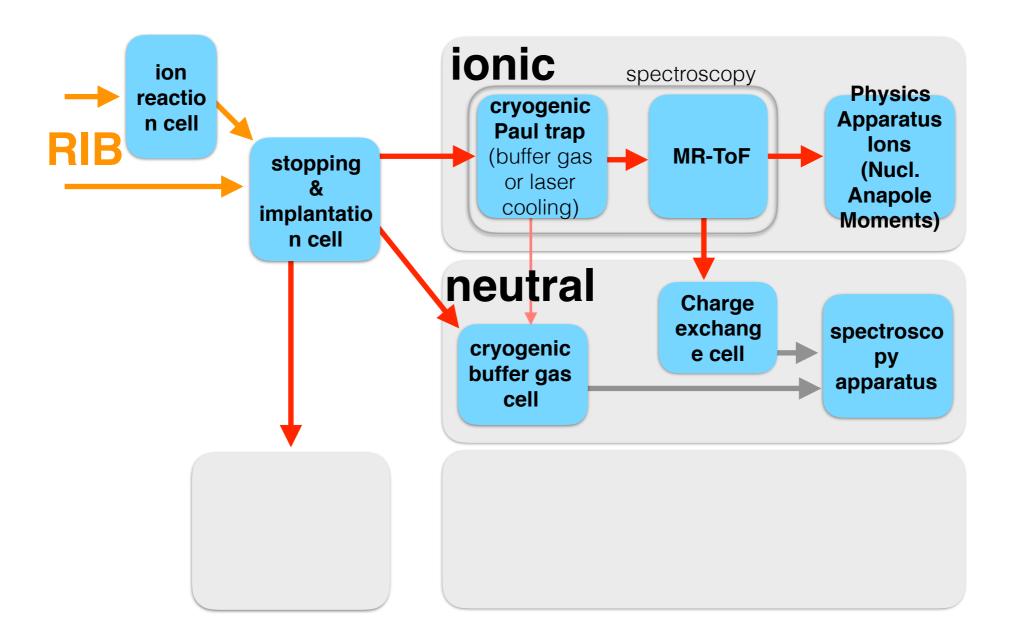


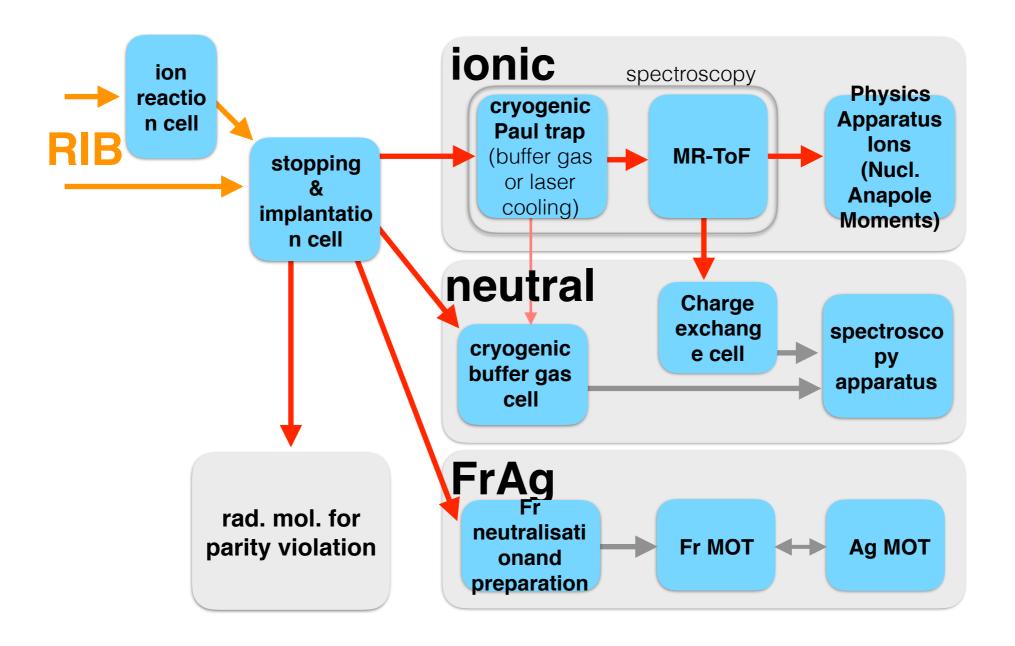
- Radioactive molecules as novel precision probes for fundamental physics
- Initial physics program:
 - octuple-deformed nuclei incorporated into polar molecules → Molecular EDM with unprecedented sensitivity to nuclear T-breaking
 - access nuclear anapole moments via diatomic molecules
- New laboratory for radioactive molecules @ TRIUMF:
 - dedicated laboratory for the study of radioactive molecules
 - to host 3 experimental stations
 - provision for expansions into other fields
- Current Canadian team: 12 faculty and staff physicists from UofToronto, TRIUMF, UBC, U. Manitoba, McGill, UofOttawa
- Current International team: MIT, Caltech, UoChicago,
 Manchester, KU Leuven, Marburg, Edinburgh, Kassel, etc.
- strengthens and builds on the initiative for a TRIUMF Precision&Quantum Centre

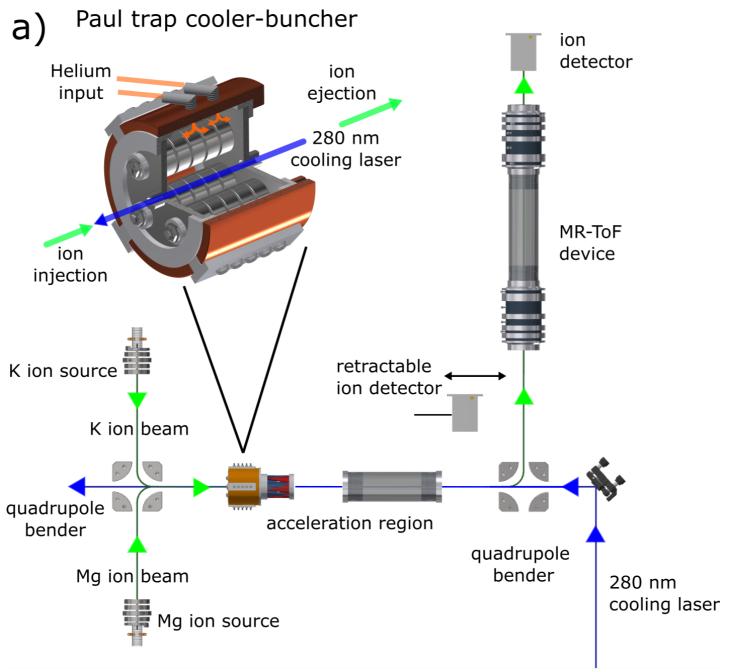


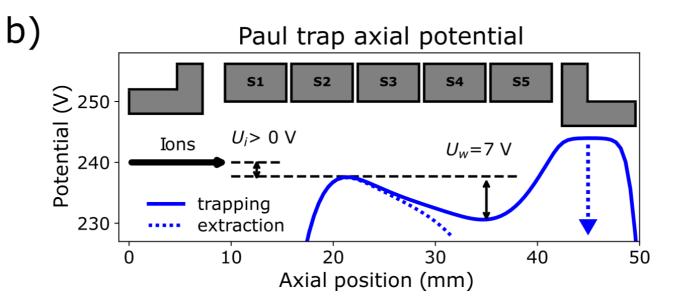
Two LOI's, ²²³FrAg and ^{227,229}ThF+, have two-five orders mag. discovery potential of nuclear time reversal compared to ¹⁹⁹Hg



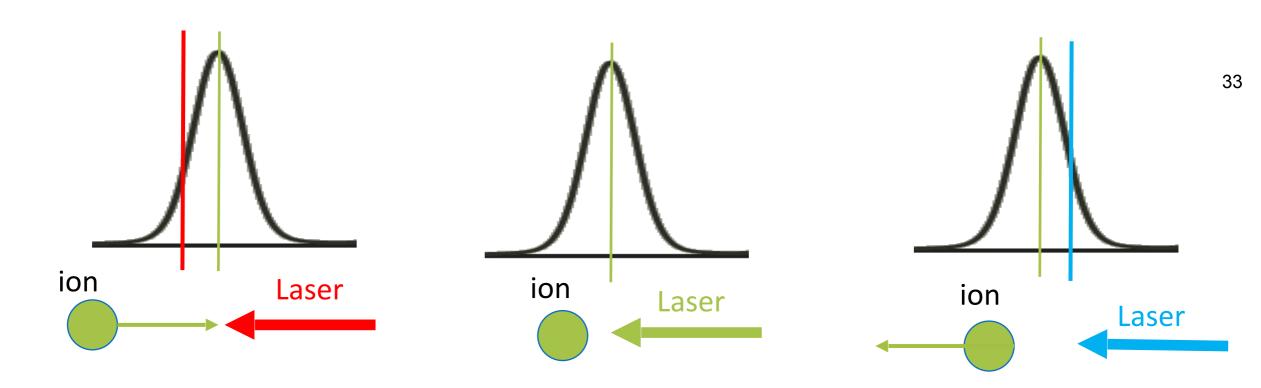








Doppler Cooling principle



- ➤ Moving ions observe Doppler shift in laser frequency
- ➤ Absorption of photon in one direction
- > Spontaneous emission of photon in random direction
- Net-cooling or heating effect since photon momentum is subtracted from/added to the Mg ion momentum

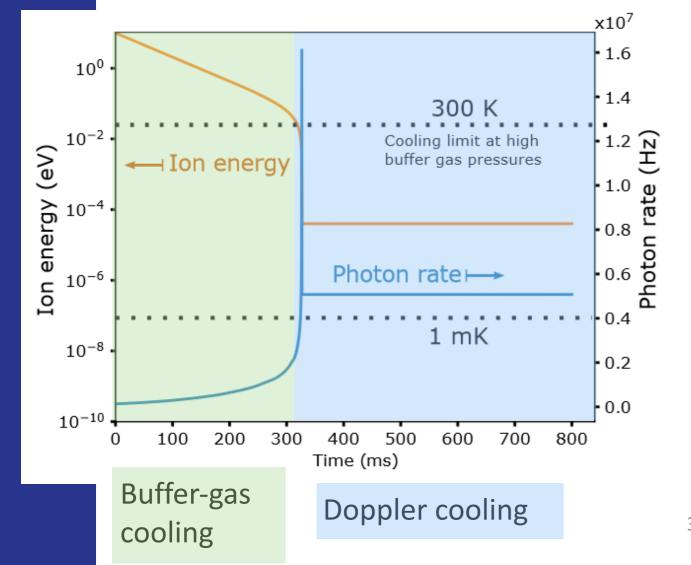
Red-detuning: cooling, blue detuning: heating

Needed cooling time

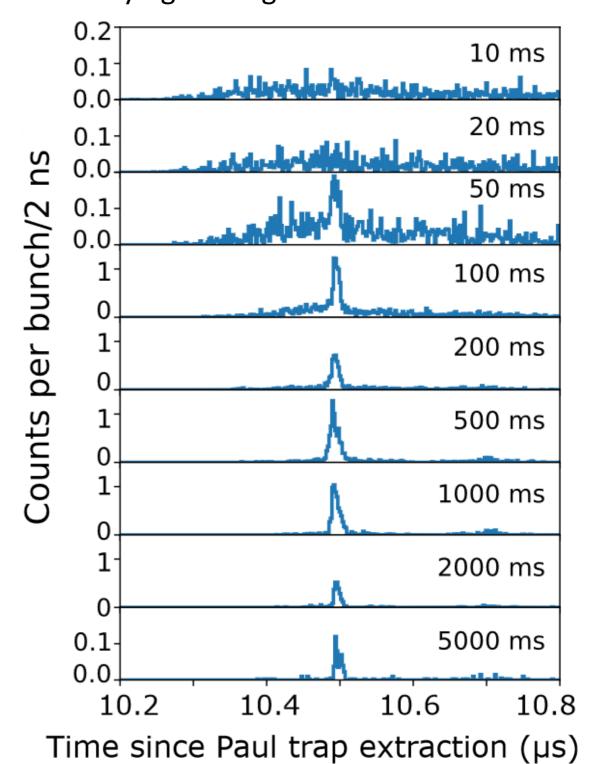
Experiment:

- ➤ 1e-5 to 1e-8 mbar residual gas present within Paul trap
- > cooling time ≈ 100 ms

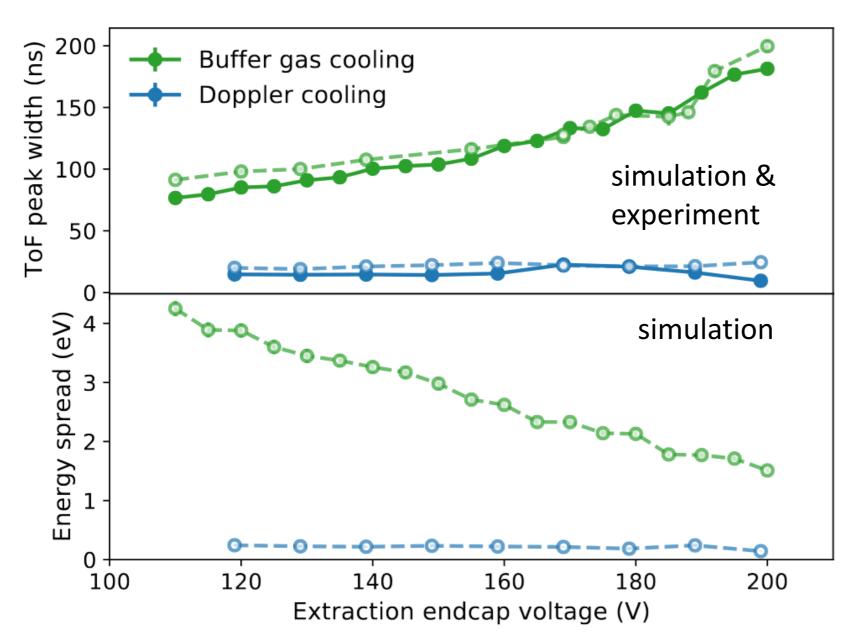
<u>Simulations + numerical model:</u> Presence of buffer gas speeds up the cooling

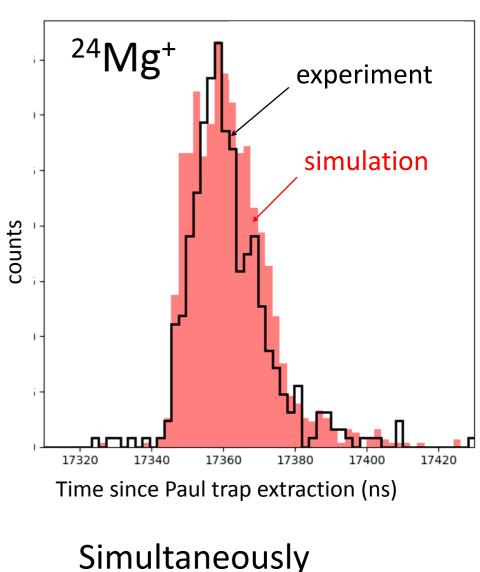


3 mW & -200 MHz detuning, varying cooling times:



Applications at RIB facilities





Simultaneously small Dt & DE

→ Improvements in precision and/or sensitivity for various experimental techniques such as collinear laser spectroscopy or mass spectrometry

