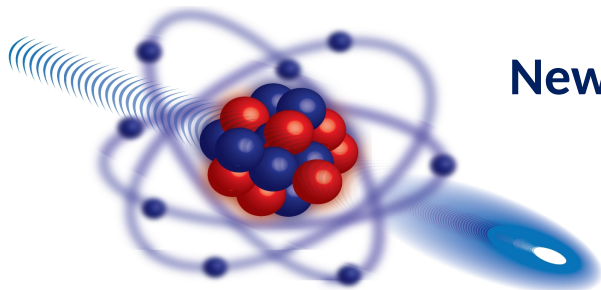




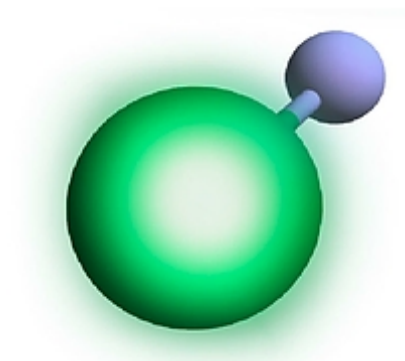
# Opportunities for New Physics searches with exotic atoms and molecules

Ronald Fernando Garcia Ruiz  
*CERN/MIT*



New Physics on the Low-Energy Precision Frontier

CERN, Geneva 2020





## CRIS Collaboration

**J. Billowes, C. Binnersley, A. Brinson, T.E. Cocolios, B. Cooper, K.T. Flanagan, S. Franchoo, V. Fedosseev, B.A. Marsh, M. Bissell, R.P. De Groote, R.F. Garcia Ruiz, A. Koszorus, G. Neyens, H. Perrett, F. Parnefjord Gustafsson, C. Ricketts, H.H. Stroke, S.M. Udrescu, A. Vernon, K. Wendt, S. Wilkins, X. Yang, .....**



## COLLAPS Collaboration

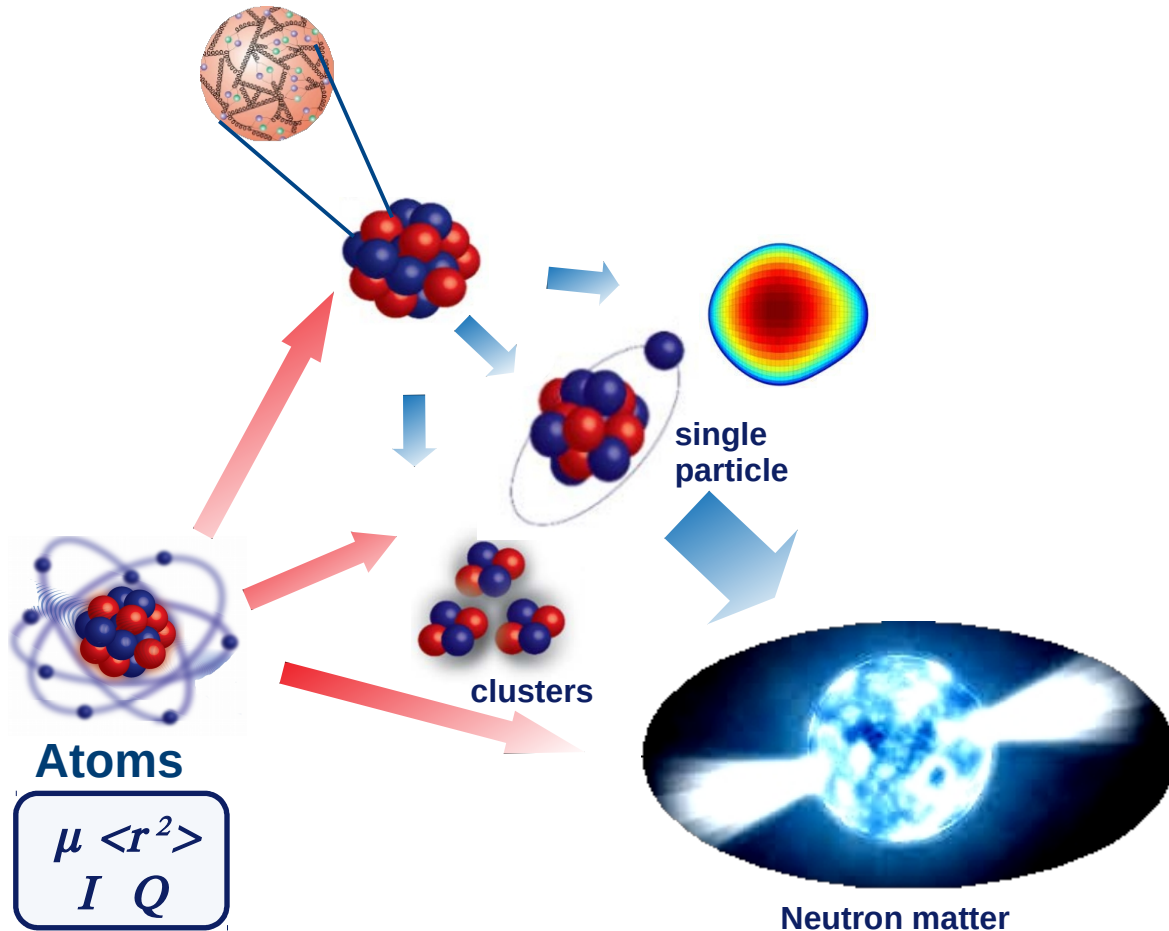
**ISOLTRAP (F. Wienholtz), RILIS (S. Wilkins, K. Chrysalidis)  
Target group (S. Rothe), ISOLDE Technical group**

***Nuclear theory: W. Nazarewicz (FRIB/MSU), P.-G. Reinhard (Erlangen-Nürnberg),  
G. Hagen (ORNL), J. Holt (TRIUMF), ...***

***Quantum chemistry: R. Berger (U. Marburg, Germany), T. Isaev (PNPI NRCKI, St. Petersburg)***

# Exotic Atoms & Molecules

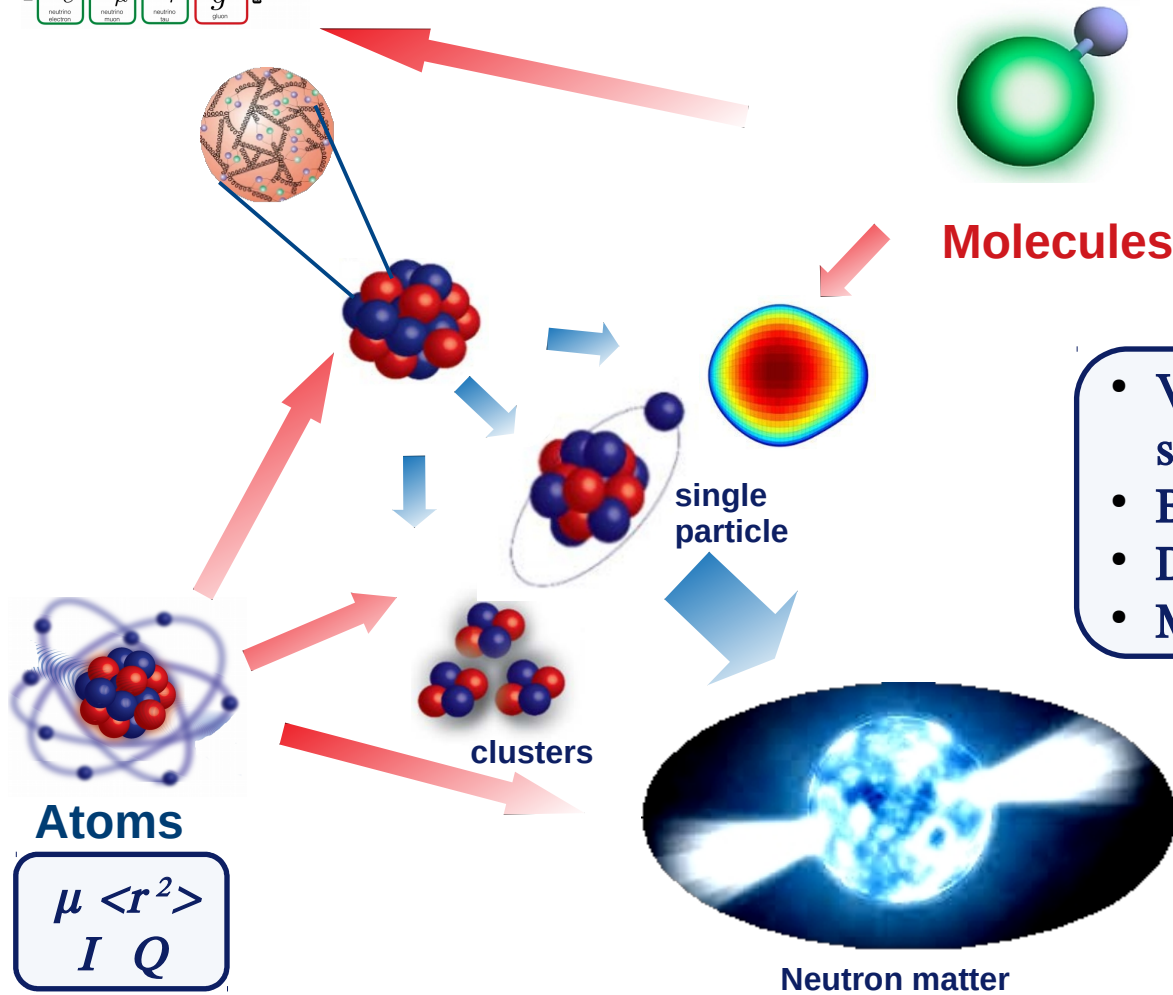
Quarks	$u$ up	$c$ charm	$t$ top	$\gamma$ photon	$H$ Higgs boson
	$d$ down	$s$ strange	$b$ bottom	$W^\pm$ W boson	
	$e$ electron	$\mu$ muon	$\tau$ tau	$Z^0$ Z boson	
Leptons	$\nu_e$ neutrino electron	$\nu_\mu$ neutrino muon	$\nu_\tau$ neutrino tau	$g$ gluon	Gauge bosons



- Nuclear force  $\rightarrow$  QCD
- Emergence of nuclear phenomena
- Understanding of nuclear matter

# Exotic Atoms & Molecules

Quarks	$u$ up	$c$ charm	$t$ top	$\gamma$ photon	$H$ Higgs boson
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Leptons	$\nu_e$ neutrino electron	$\nu_\mu$ neutrino muon	$\nu_\tau$ neutrino tau	$g$ gluon	
	Gauge bosons				



**Atoms**  
 $\mu \langle r^2 \rangle$   
 $I Q$

**Nuclear**  
 (Electromagnetic)

**Molecules**

- Violation of fundamental symmetries
- BSM searches
- Dark matter
- Matter / Antimatter asymmetry

- Nuclear force  $\rightarrow$  QCD
- Emergence of nuclear phenomena
- Understanding of nuclear matter

# Opportunities for New Physics searches with exotic atoms and molecules

## Contents

- Precision studies in atoms and molecules
- Nuclear structure / nuclear matter / new forces & particles
- Parity violation
- Parity and time-reversal violation
- First results: RaF
- Summary

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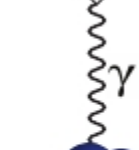
### Why exotic atoms/molecules?

- Large  $Z$ ,  $A$
- Nuclear spins
- Isotopic chains
- Octupole deformation

# Precision studies in atoms and molecules

Nuclear  
structure

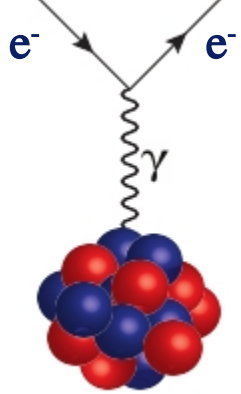
$e^-$   $e^-$



$$\mu \langle r^2 \rangle I Q$$

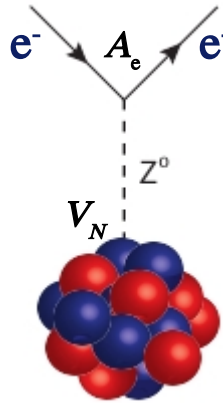
# Precision studies in atoms and molecules

## Nuclear structure

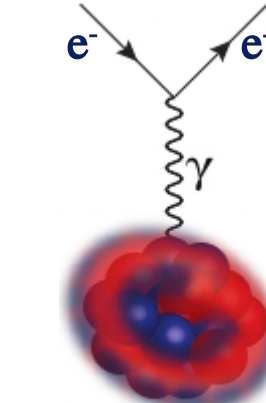
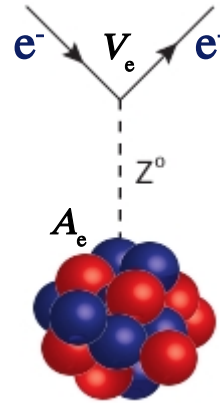


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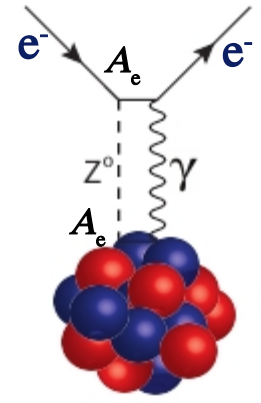
## P- violation



$$\sim Z^3 \times 10^N$$



$$W^+ Z^0 \sim Z^2 A^{2/3} R(Z) \times 10^5$$

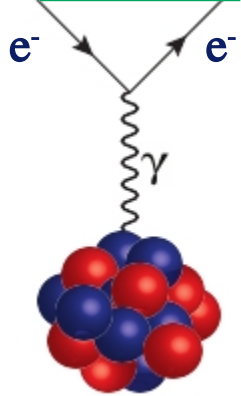


Atoms  
Molecules



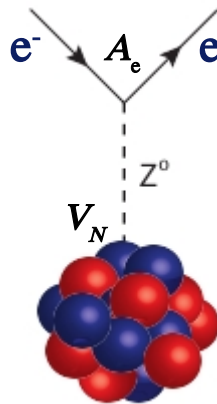
# Precision studies in atoms and molecules

## Nuclear structure

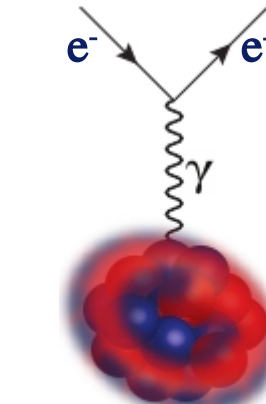
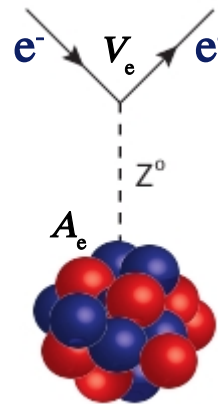


$$\mu \langle r^2 \rangle I Q$$

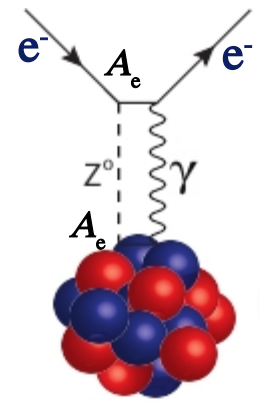
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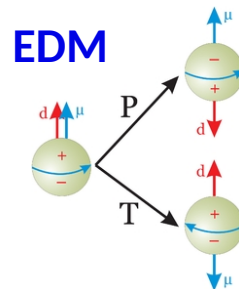


$$W^+ Z^0 \sim Z^2 A^{2/3} R(Z) \times 10^5$$

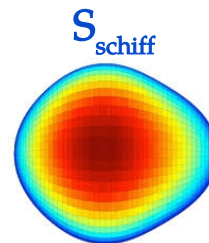


Atoms  
Molecules

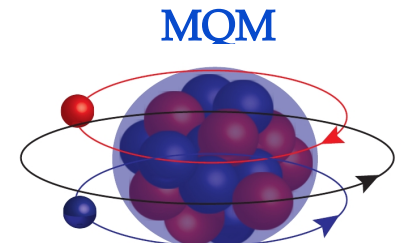
## P,T- violation



$$\sim Z^2 R(Z) > 10^3$$



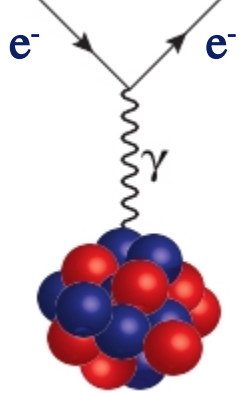
$$S \sim Q_2 Q_3 Z A^{2/3} / (E_+ - E_-) (> 10^5 \text{ Octupole})$$



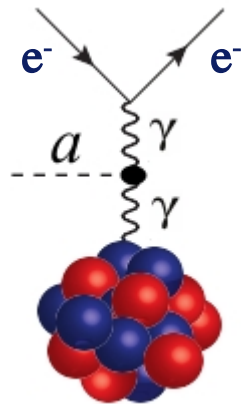
Atoms  
Molecules

# Precision studies in atoms and molecules

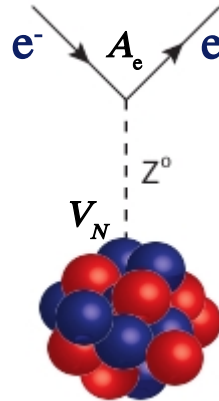
## Nuclear structure



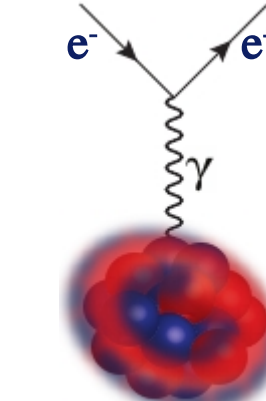
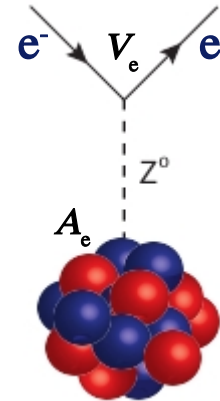
$$\mu \langle r^2 \rangle I Q$$



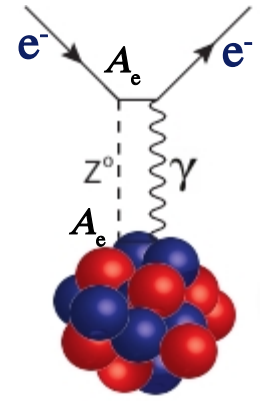
## P- violation



$$\sim Z^3 \times 10^N$$

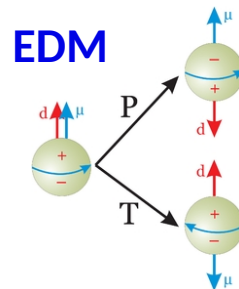


$$W^+ Z^0 \sim Z^2 A^{2/3} R(Z) \times 10^5$$

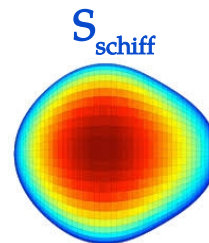


Atoms  
Molecules

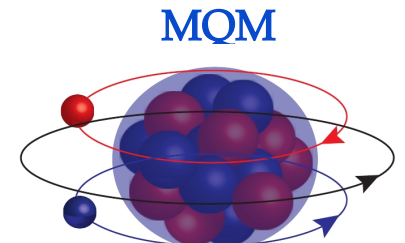
## P,T- violation



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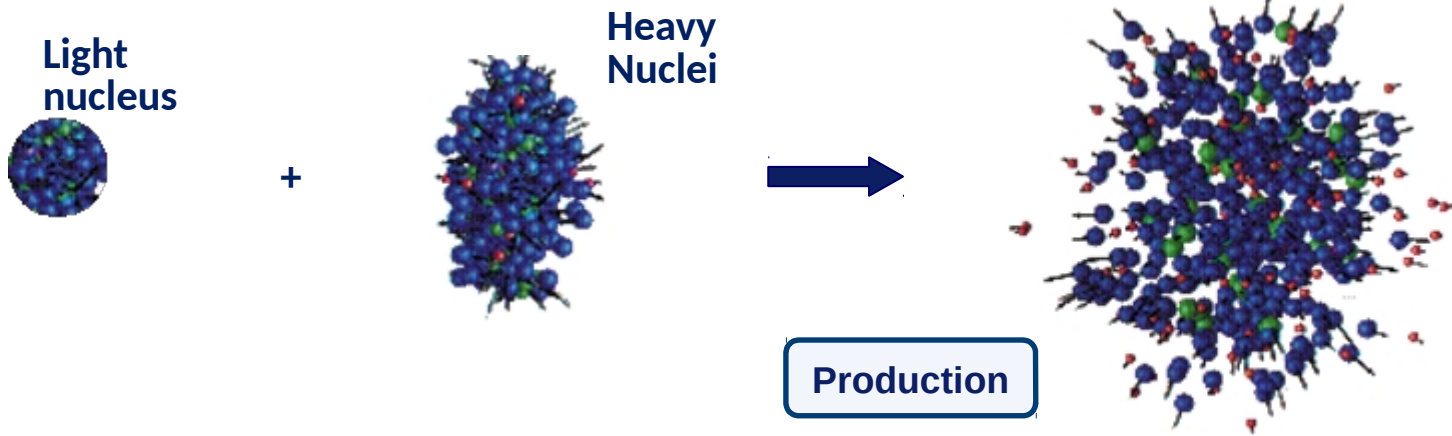
Atoms  
Molecules

Best of all worlds → Radioactive molecules containing heavy and octupole deformed nuclei

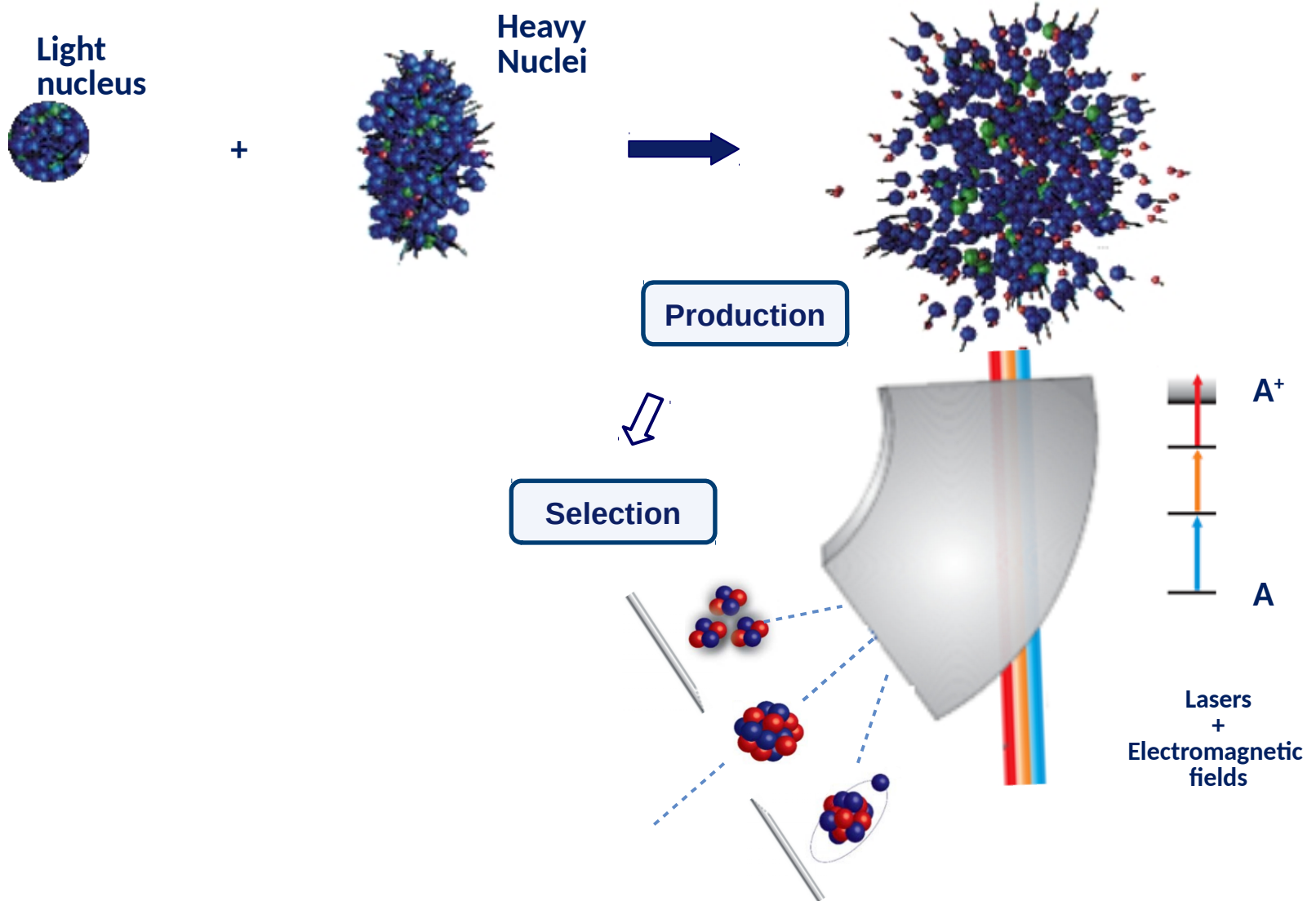
# Contents

- Precision studies in atoms and molecules
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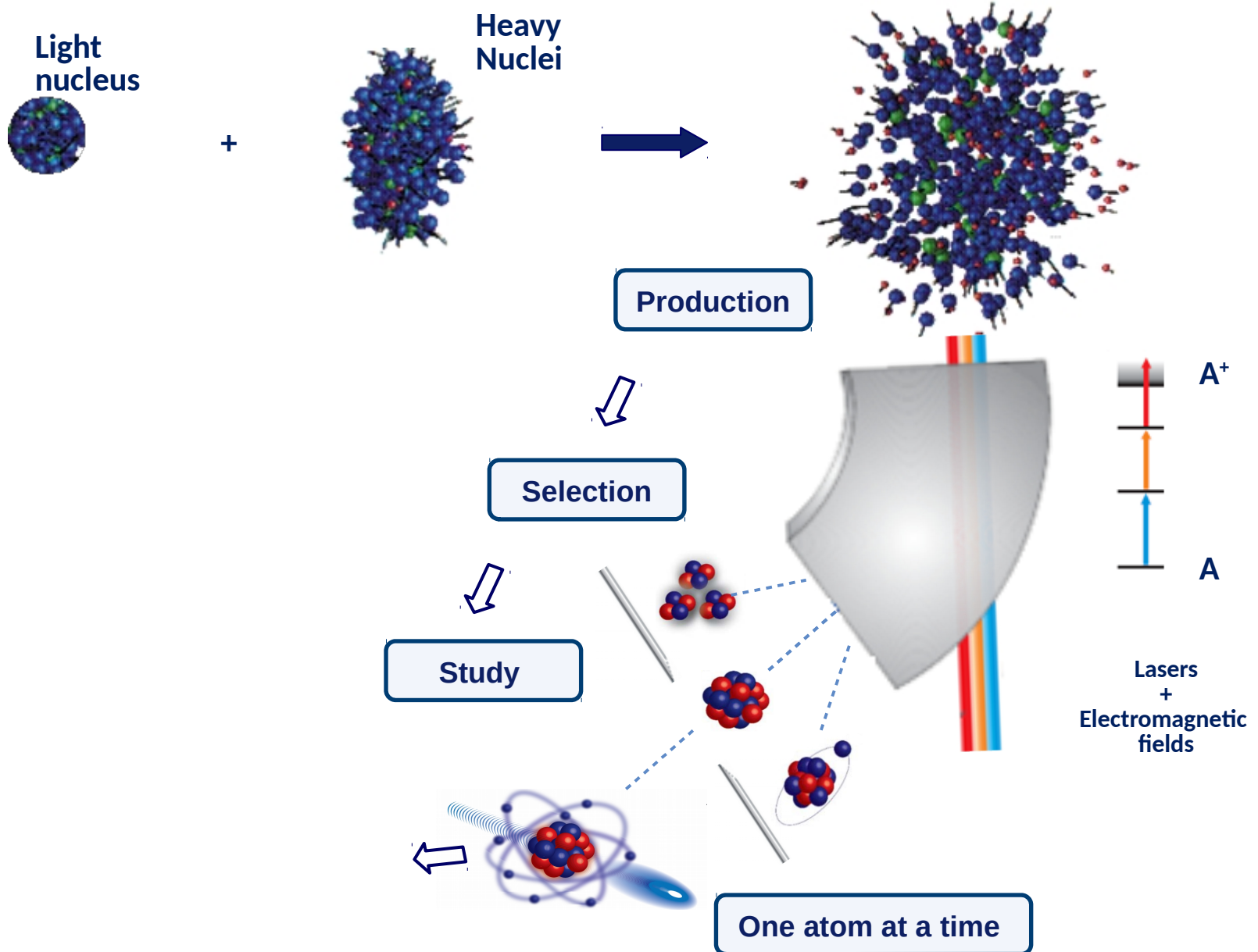
# How do we create and study exotic atoms/molecules?



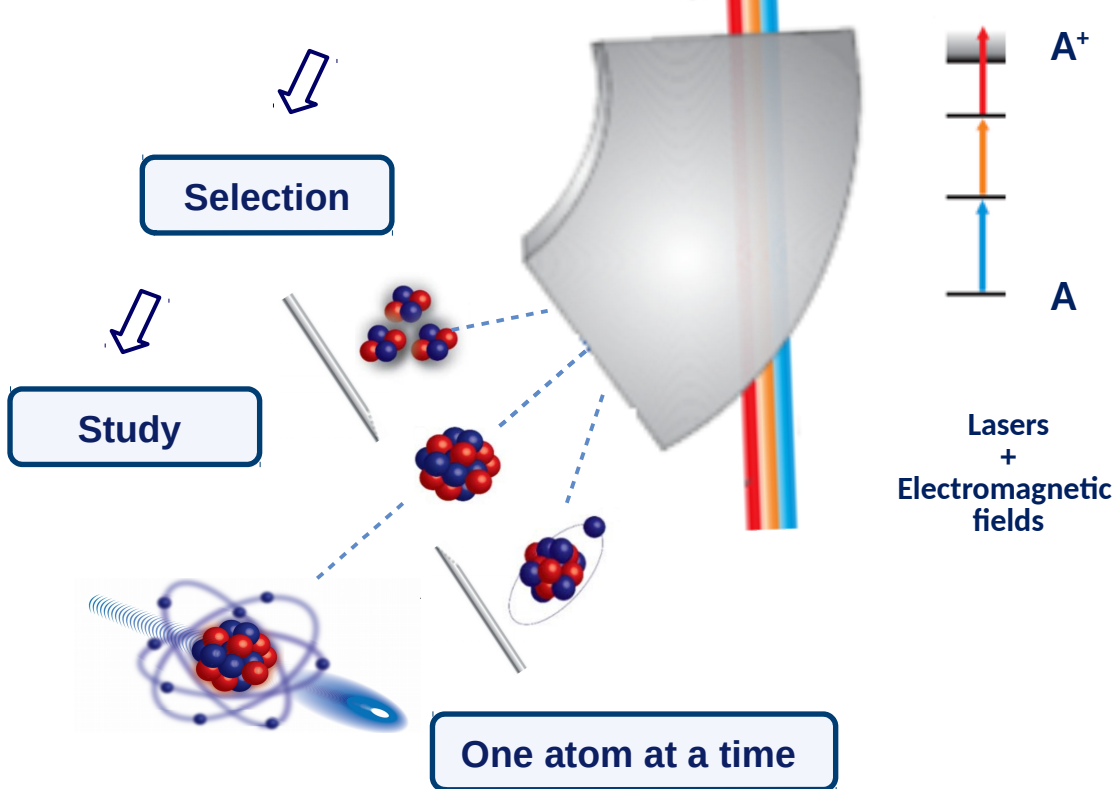
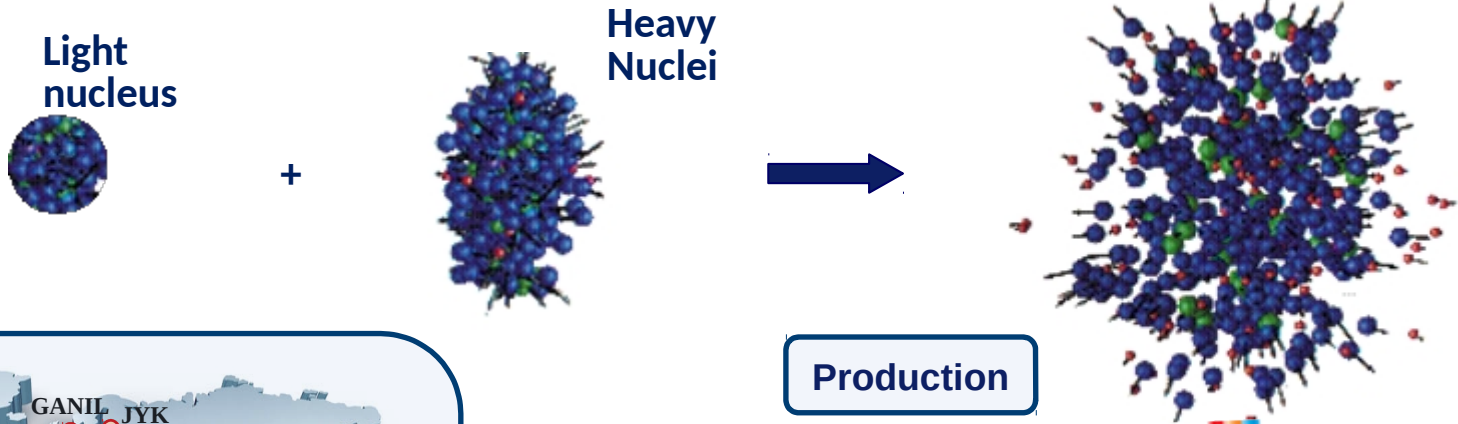
# How do we create and study exotic atoms/molecules?



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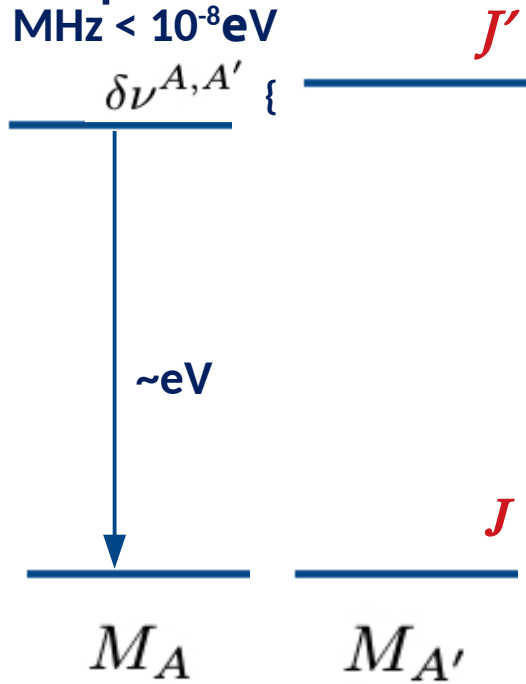


## Experimental challenges:

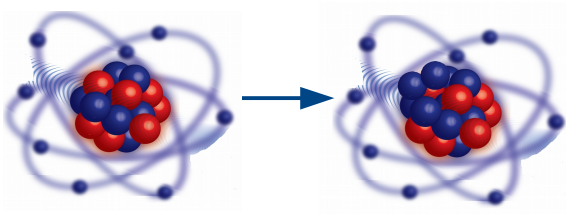
- High selectivity ( $>1/10^6$ )
- Short time scales ( $< 1$  s)
- High efficiency ( $<100$  ions/s)
- High precision/resolution ( $\sim$ MHz)

# Exotic atoms & Nuclear structure

Isotope shift  
MHz  $< 10^{-8}$  eV



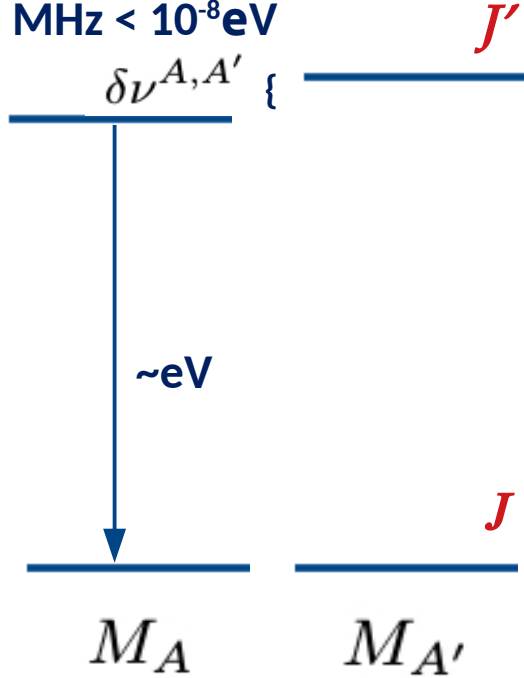
$$I = 0$$





# Exotic atoms & Nuclear structure

Isotope shift  
MHz <math> < 10^{-8} \text{eV}</math>



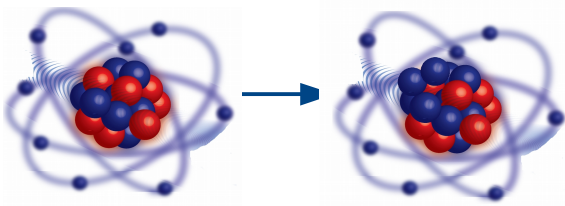
## Electromagnetic structure

- Rms charge radii:  $\langle r^2 \rangle$

Atomic  
Nuclear

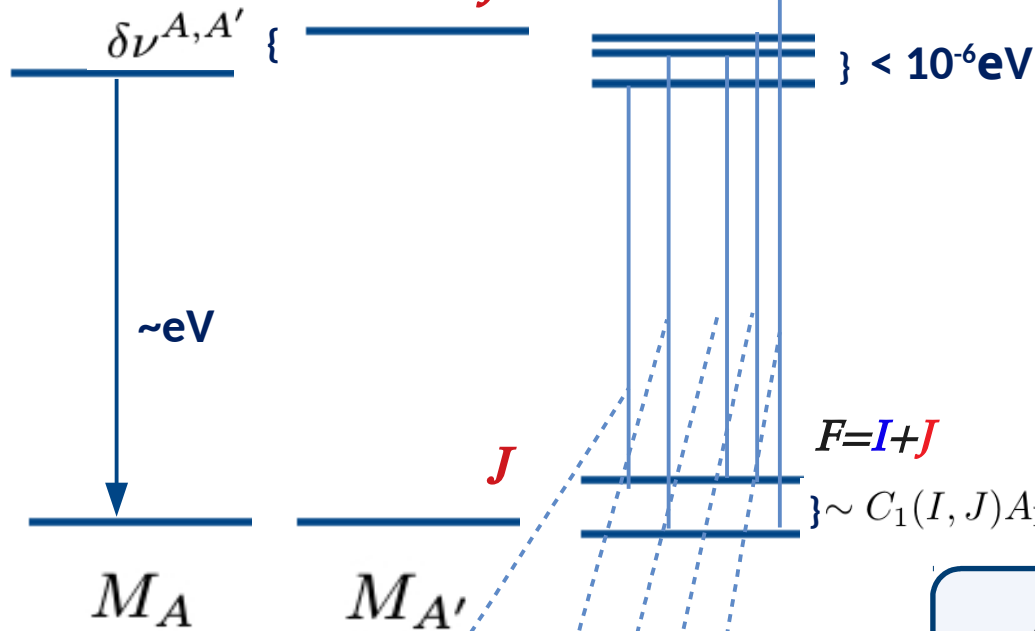
$I = 0$

$$\delta\nu^{A,A'} = K_{MS} \frac{M_{A'} - M_A}{M_{A'} M_A} + F \delta \langle r^2 \rangle^{A,A'}$$



# Exotic atoms & Nuclear structure

Isotope shift  
MHz <math>10^{-8}</math> eV



Atomic hyperfine structure

Electromagnetic structure

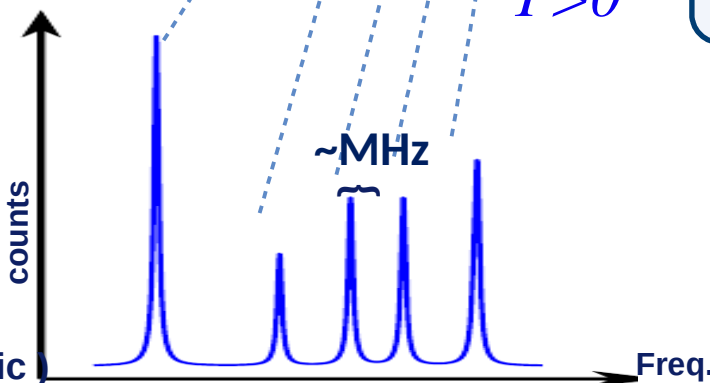
- Rms charge radii:  $\langle r^2 \rangle$
- Nuclear spin:  $I$
- Electromagnetic moments:  $\mu$
- Quadrupole moment :  $Q$

Atomic  
Nuclear

$$\delta\nu^{A,A'} = K_{MS} \frac{M_{A'} - M_A}{M_{A'} M_A} + F \delta \langle r^2 \rangle^{A,A'}$$

$$A_{\text{hfs}} = \frac{\mu B}{IJ}$$

$$B_{\text{hfs}} = eQ \frac{\partial^2 V}{\partial^2 z}$$



$\mu$   $\langle r^2 \rangle$   
 $I$   $Q$

Nuclear

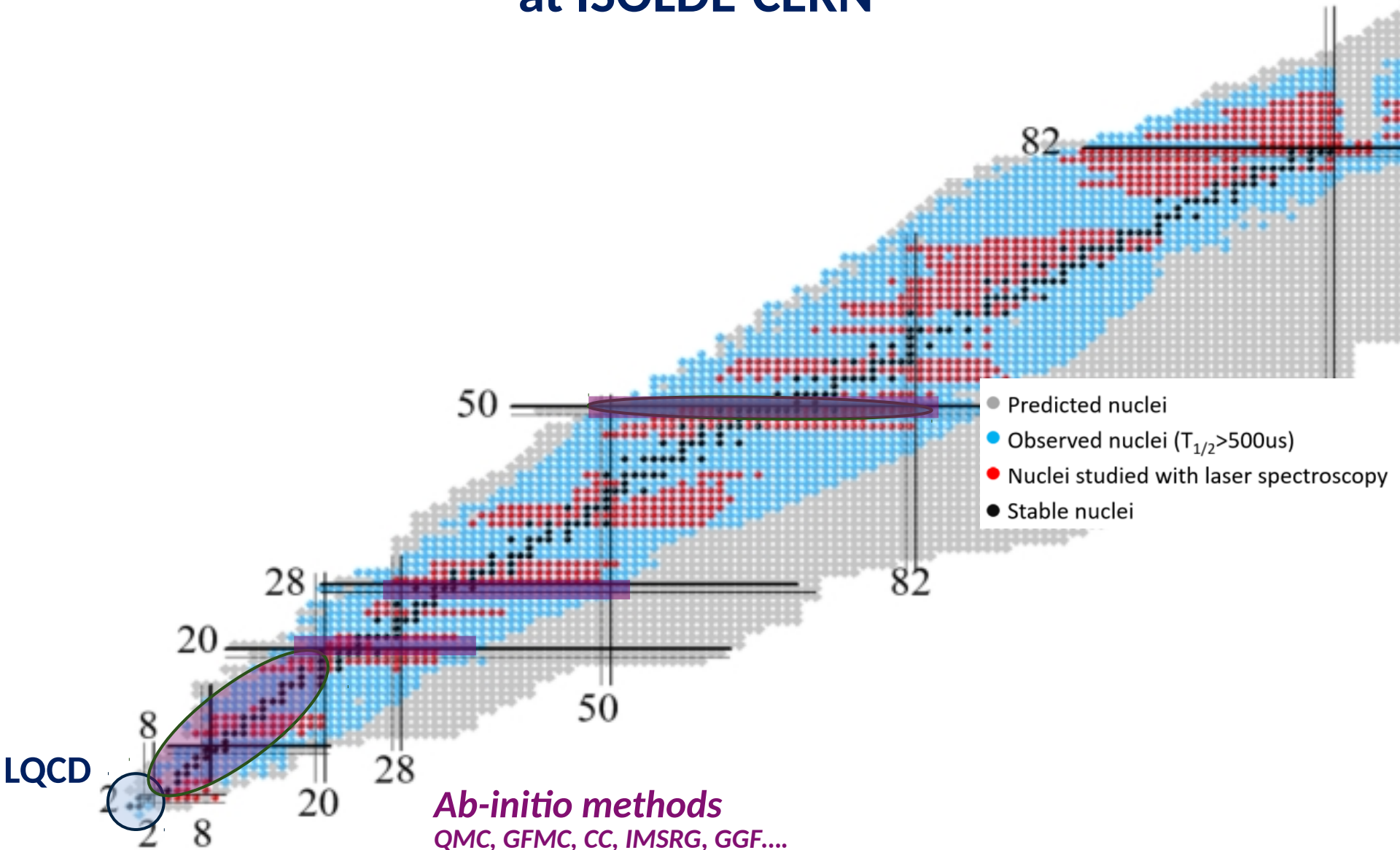
(Electromagnetic)

$I > 0$

$\sim$  MHz

Freq.

# Recent results from collinear laser spectroscopy at ISOLDE-CERN



# Recent results from collinear laser spectroscopy at ISOLDE-CERN

Published  
Unpublished

<sup>202-231</sup>Fr (Z=87) [Phys. Rev. Lett 115, 132501 (2015)]  
<sup>222-233</sup>Ra (Z=88) [Phys. Rev. X 4 (1), 011055 (2014)]  
 [Phys Rev. Lett. 111, 212502 (2013)]

<sup>100-130</sup>Cd (Z=48) [Phys Rev Lett 121, 102501 (2018)]  
 [Phys Rev Lett 116, 032501 (2016)] , ...

<sup>101-131</sup>In (Z=49) [Phys Rev X 8, 041005 (2018)]  
<sup>103-134</sup>Sn (Z=50) [Phys. Rev. Lett. 122, 192502 (2019)]  
<sup>112-134</sup>Sb (Z=51)

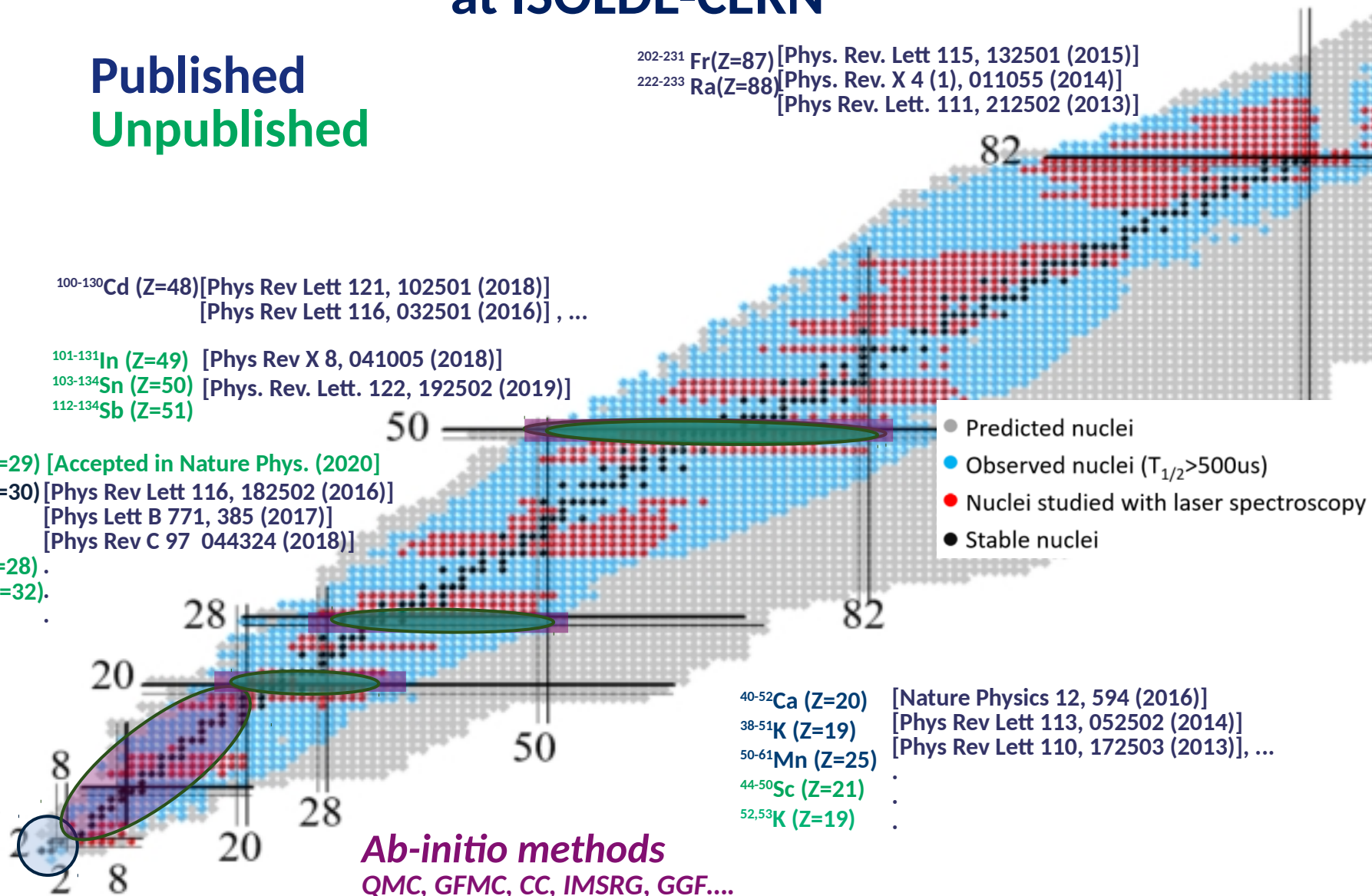
<sup>58-78</sup>Cu (Z=29) [Accepted in Nature Phys. (2020)]  
<sup>65-80</sup>Zn (Z=30) [Phys Rev Lett 116, 182502 (2016)]  
 [Phys Lett B 771, 385 (2017)]  
 [Phys Rev C 97 044324 (2018)]  
<sup>56-68</sup>Ni (Z=28) .  
<sup>68-74</sup>Ge (Z=32).

● Predicted nuclei  
 ● Observed nuclei ( $T_{1/2} > 500\mu\text{s}$ )  
 ● Nuclei studied with laser spectroscopy  
 ● Stable nuclei

<sup>40-52</sup>Ca (Z=20) [Nature Physics 12, 594 (2016)]  
<sup>38-51</sup>K (Z=19) [Phys Rev Lett 113, 052502 (2014)]  
<sup>50-61</sup>Mn (Z=25) [Phys Rev Lett 110, 172503 (2013)], ...  
<sup>44-50</sup>Sc (Z=21) .  
<sup>52,53</sup>K (Z=19) .

LQCD

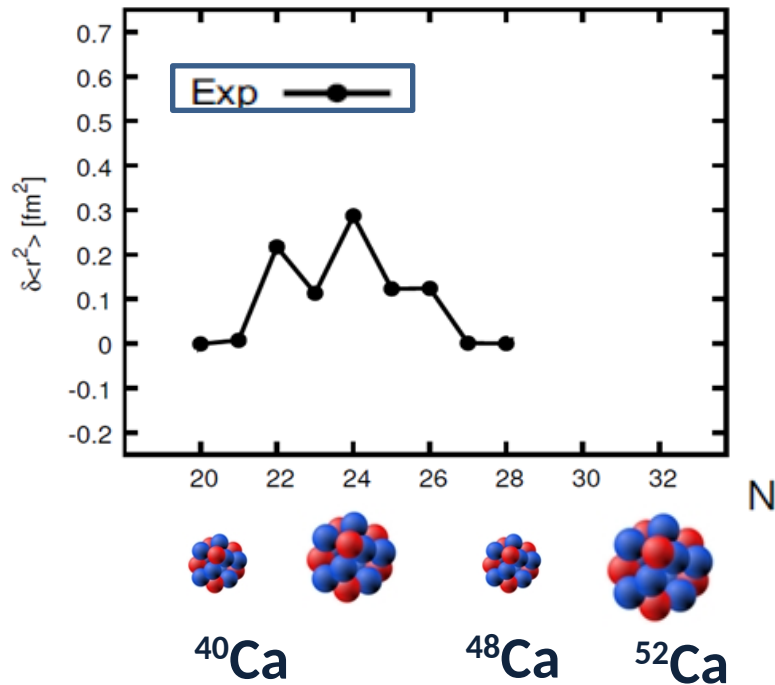
*Ab-initio methods*  
 QMC, GFMC, CC, IMSRG, GGF....



# An example: exotic calcium isotopes

[Ca:  $\langle r^2 \rangle$ : Garcia Ruiz et al. Nature Phys. 12, 594 (2016)]

$$\delta \nu^{A,A'} = K_{MS} \frac{M_{A'} - M_A}{M_{A'} M_A} + F \delta \langle r^2 \rangle^{A,A'}$$



$^{36}\text{Ca} \rightarrow \langle r^2 \rangle$  @NSCL

[Miller et al. Nature Phy, 15, 432 (2019)]

$^{52}\text{Ca} \rightarrow S_{2n}$  @CERN

[Wienholtz et al. Nature 498, 346 (2013)]

$^{54}\text{Ca} \rightarrow E(2^+)$  @RIKEN

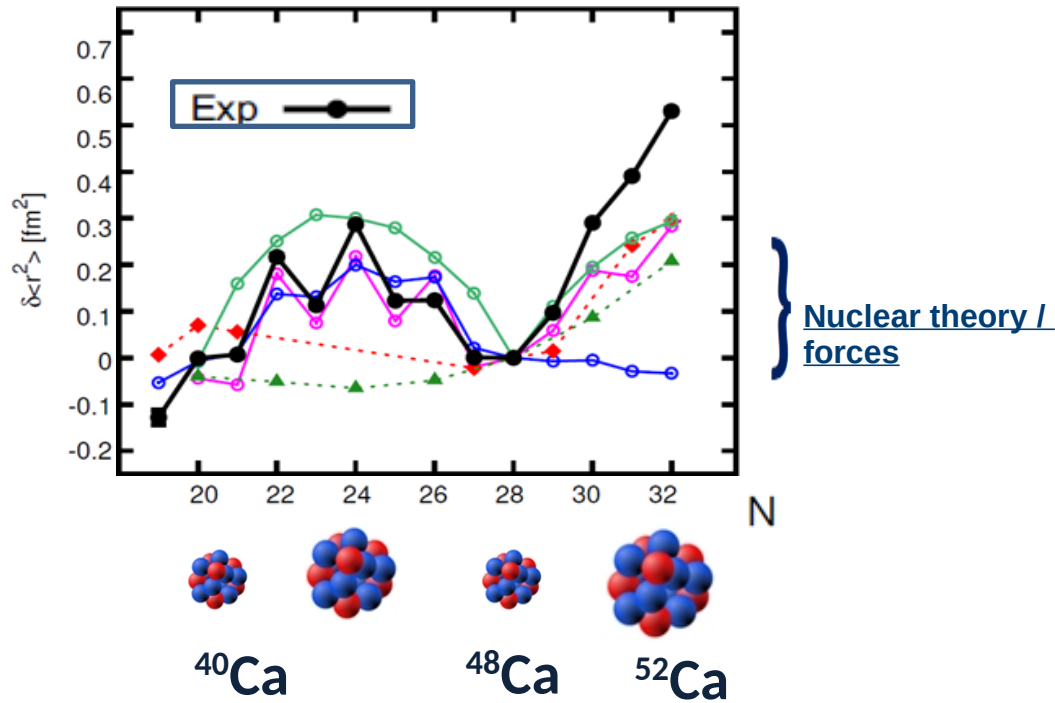
[Steppenbeck et al. Nature 502, 207(2013)]

# An example: exotic calcium isotopes

Atom  
Nucleus

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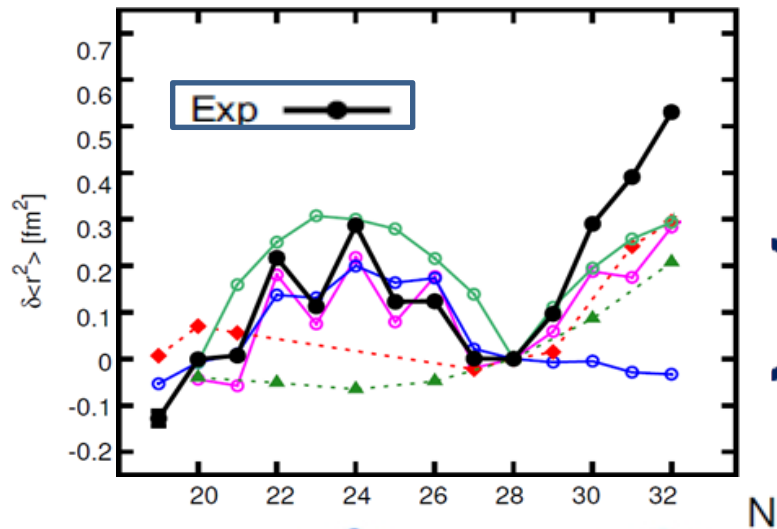


# An example: exotic calcium isotopes

Atom  
Nucleus

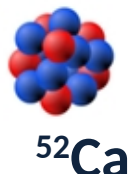
[Ca:  $\langle r^2 \rangle$ : Garcia Ruiz et al. Nature Phys. 12, 594 (2016)]

$$\delta \nu^{A,A'} = K_{MS} \frac{M_{A'} - M_A}{M_{A'} M_A} + F \delta \langle r^2 \rangle^{A,A'}$$



Nuclear theory /  
forces

Neutron-rich nuclei:  
“Quantum simulators” of neutron stars

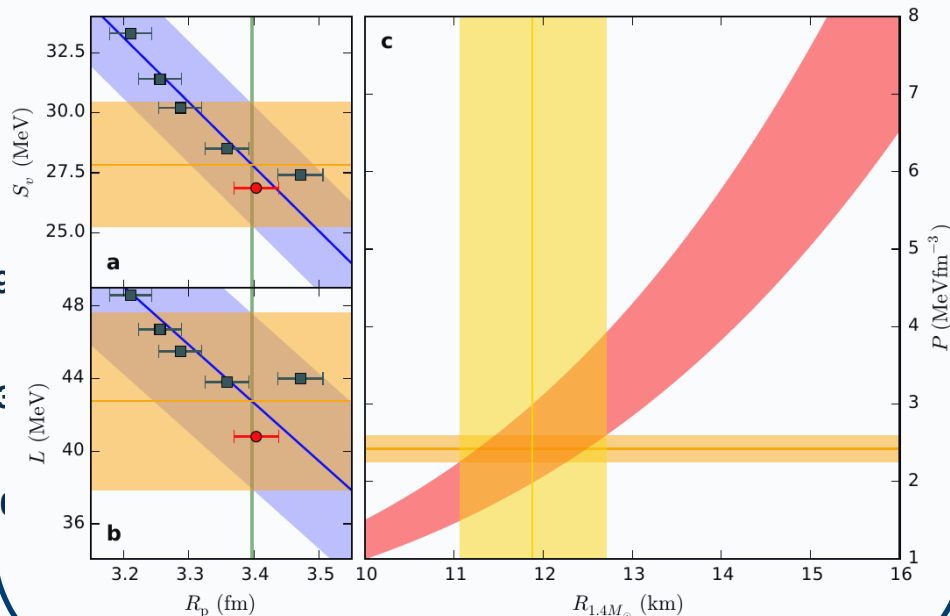


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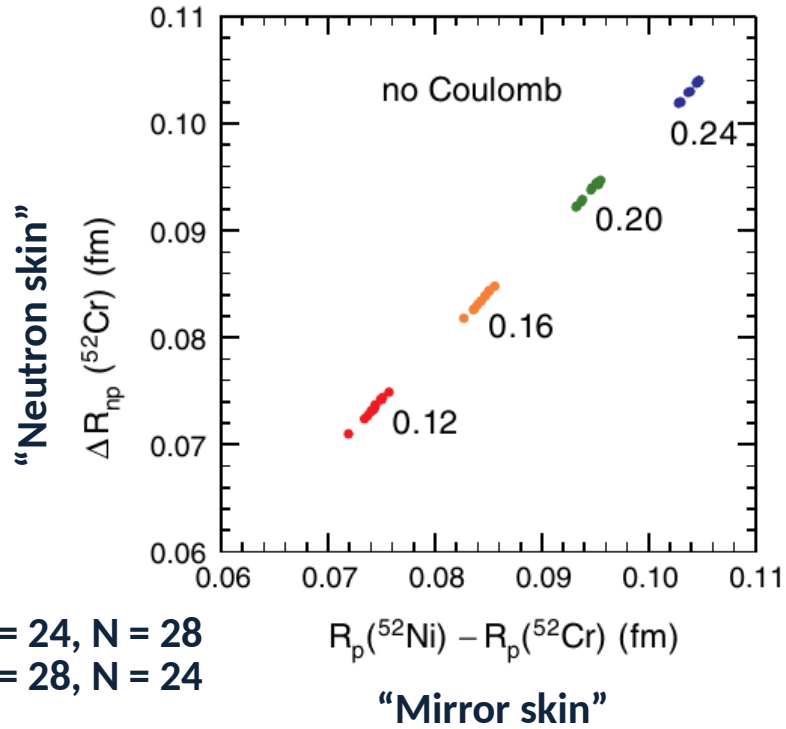
Nuclear charge radii ( $^{48}\text{Ca}$ ): Constrain to  
properties of nuclear matter



[Hagen et al., Nature Physics 12, 186 (2016)]

# Isotope shifts & “Skins”

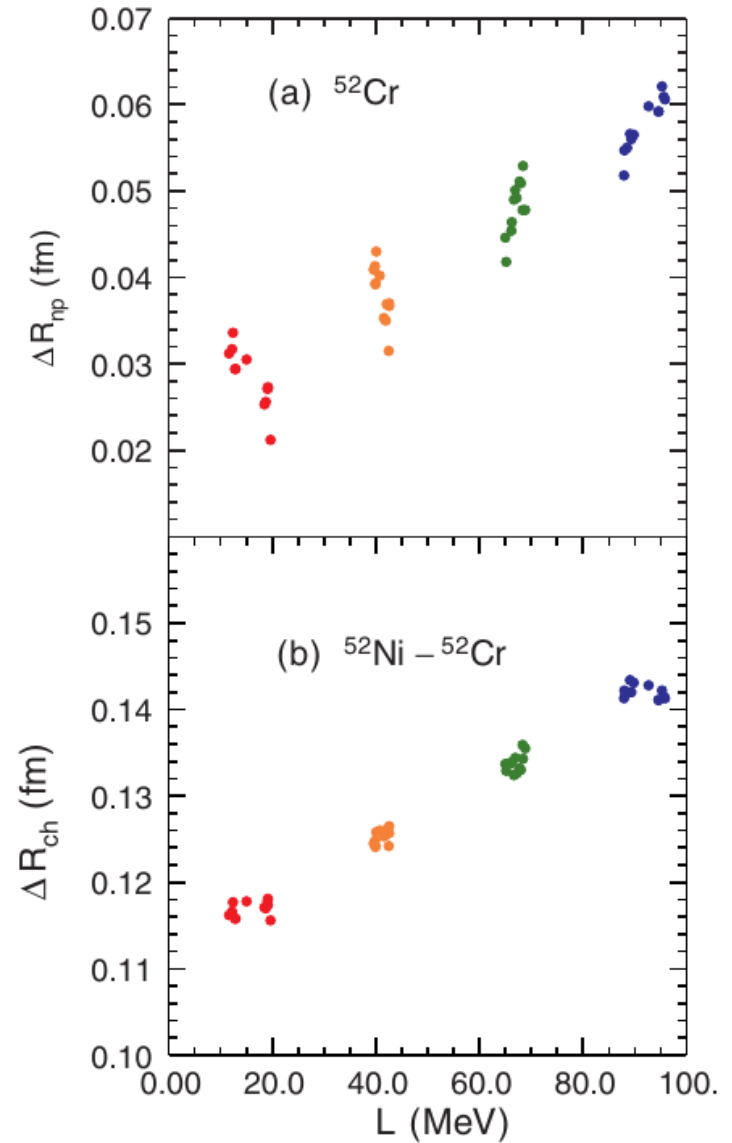
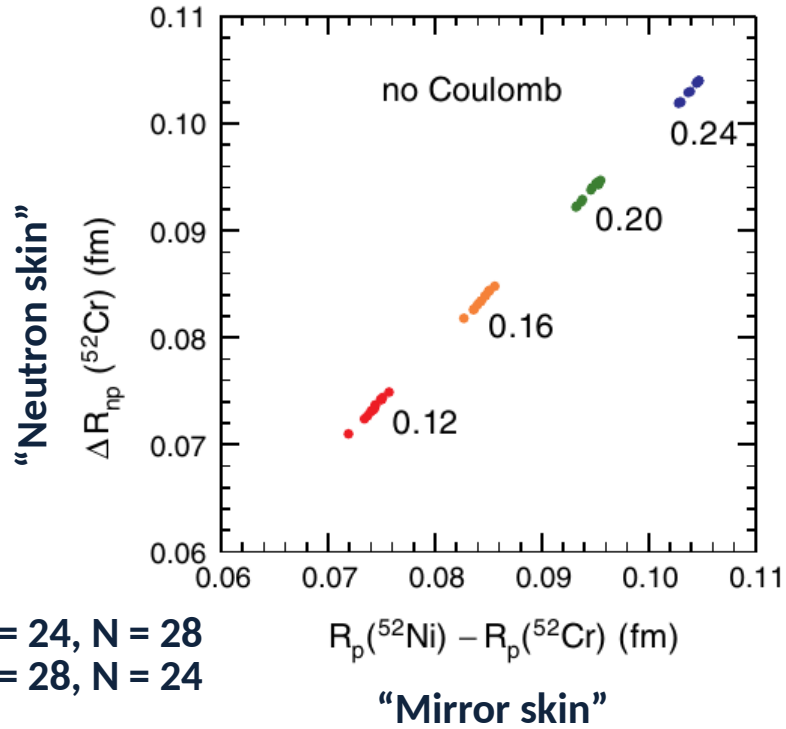
[Brown. PRL 119, 122502 (2017)]





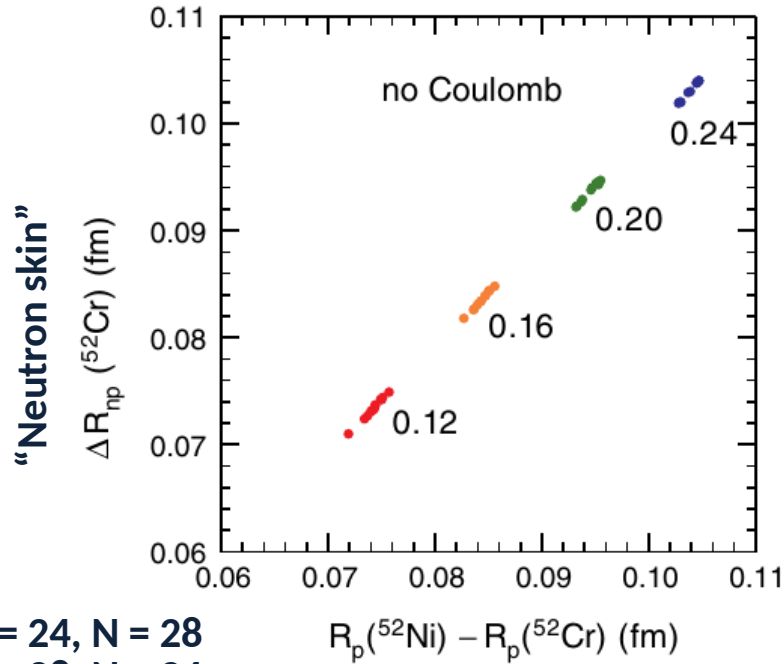
# Isotope shifts & “Skins”

[Brown. PRL 119, 122502 (2017)]



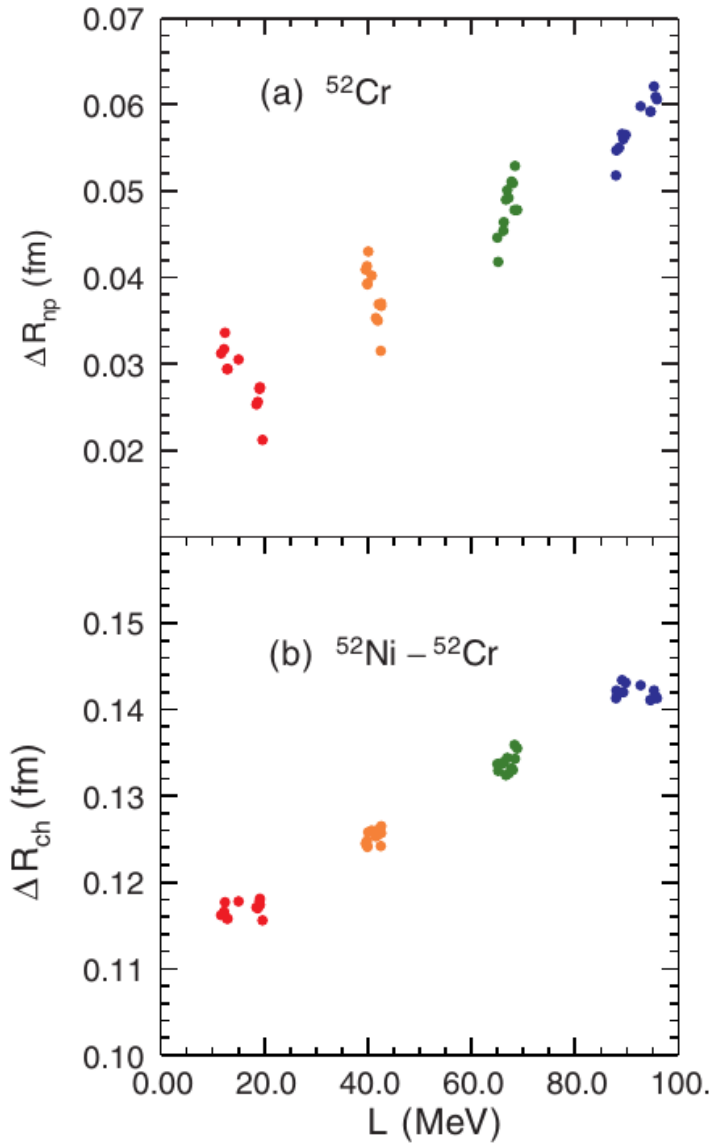
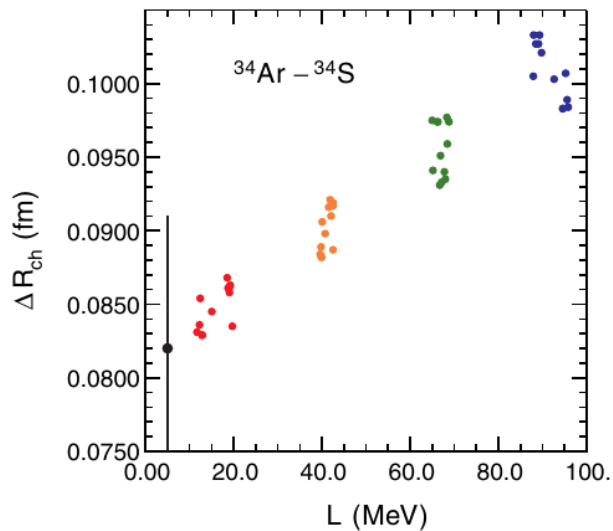
# Isotope shifts & “Skins”

[Brown. PRL 119, 122502 (2017)]



Z = 24, N = 28  
Z = 28, N = 24

“Mirror skin”

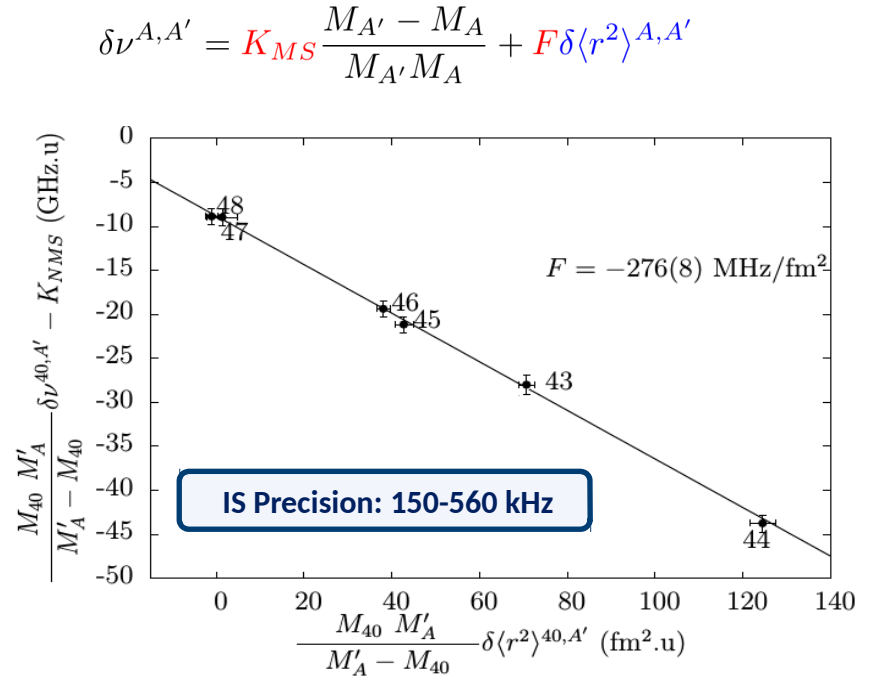
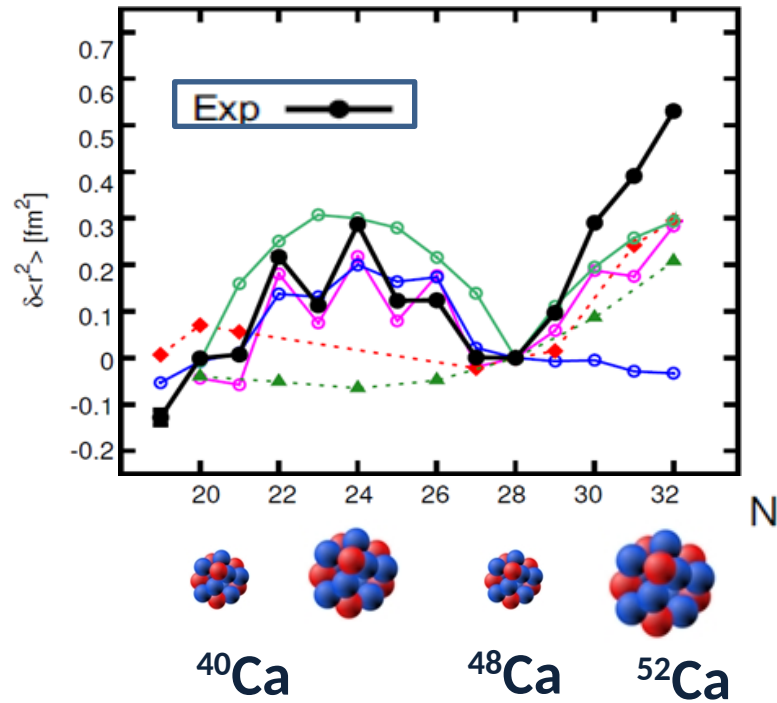


$^{32}\text{Si}$  (Z = 14, N = 18) &  $^{32}\text{Ar}$  (Z=18, N=14)

[Garcia Ruiz, Minamisono et al. NSCL PAC 20002 (2020)]

# Isotope shifts & New Physics

[Ca:  $\langle r^2 \rangle$ : Garcia Ruiz et al. Nature Phys. 12, 594 (2016)]

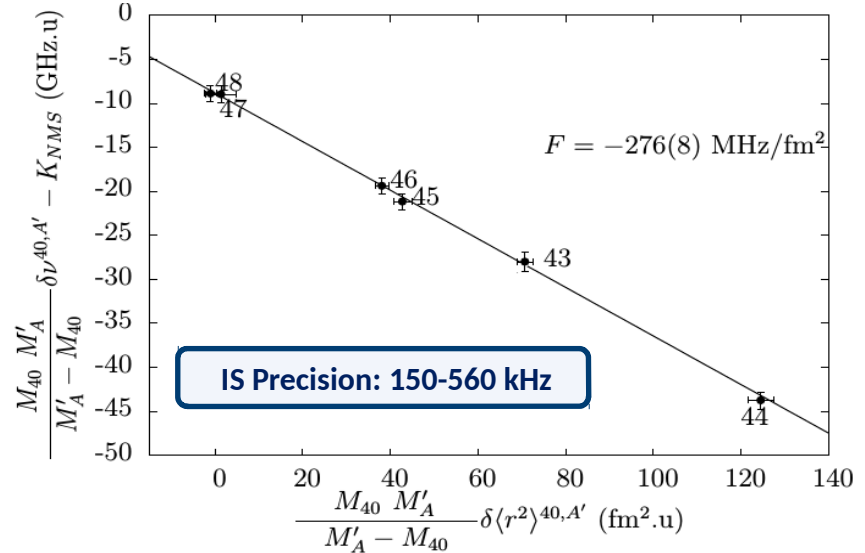


# Isotope shifts & New Physics

Atom  
Nucleus

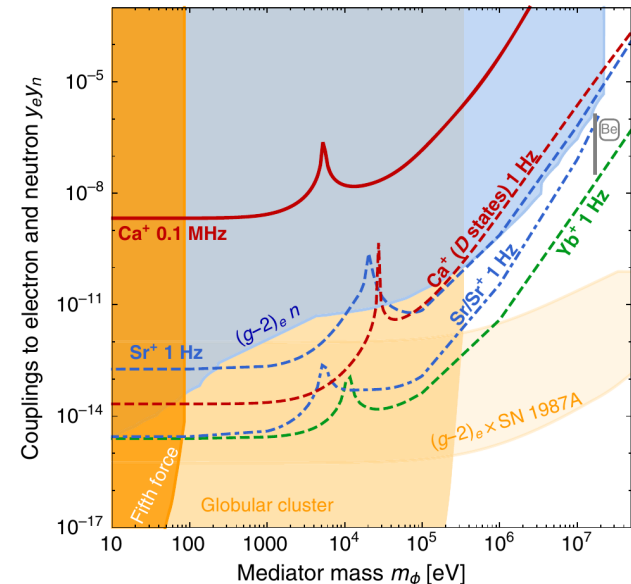
[Ca:  $\langle r^2 \rangle$ : Garcia Ruiz et al. Nature Phys. 12, 594 (2016)]

$$\delta\nu^{A,A'} = K_{MS} \frac{M_{A'} - M_A}{M_{A'} M_A} + F \delta \langle r^2 \rangle^{A,A'} + \alpha_{NP} X_i \gamma_{AA'}$$



A new force between electrons and nucleons will cause a “King plot” non-linearity

- [Berengut et al. Phys Rev Lett 120, 091801 (2018)]
- [Stadnik et al. Phys Rev Lett 120, 223202 (2018)]
- [Flambaum et al. Phys Rev A 97, 032510 (2018)]
- [Frugiuele et al. Phys Rev D 96, 015011 (2017)]

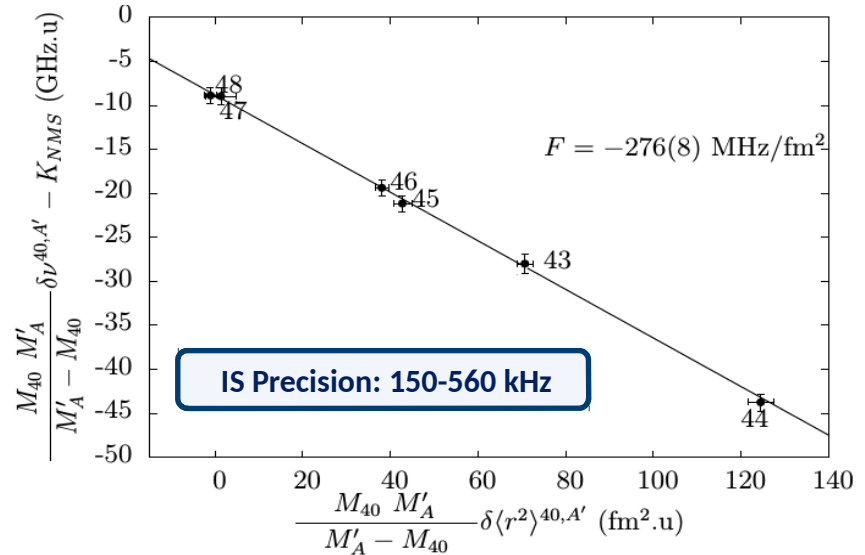


# Isotope shifts & New Physics

Atom  
Nucleus

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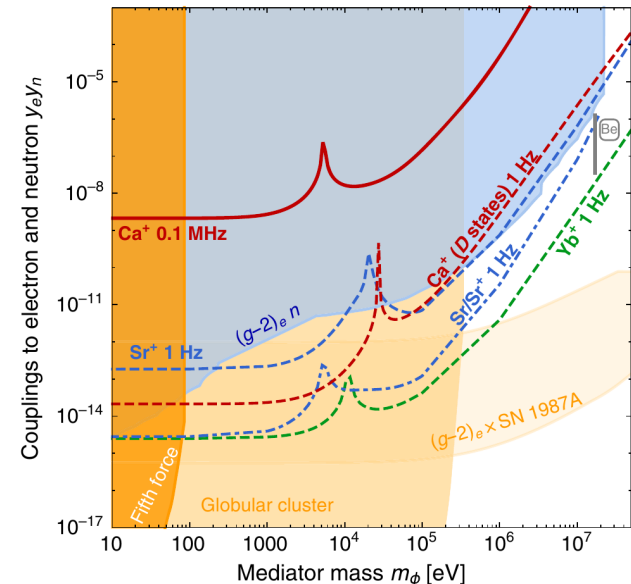
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Sr<sup>+</sup> → mHz (Weizmann Institute)

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Yb → < kHz (MIT, V. Vuletic group)



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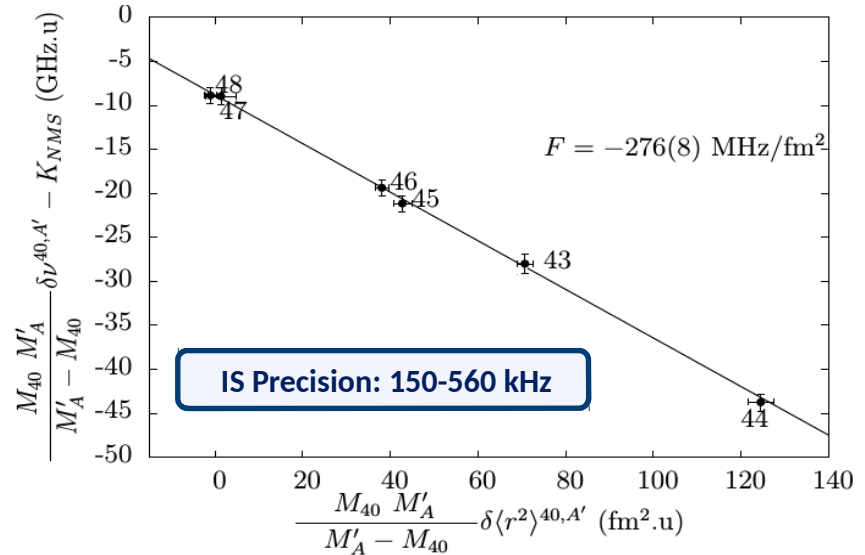
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## Opportunities with exotic atoms (?):

- ✓ Access to long isotopic chains
- ✓ Nuclear isomers ( $M'_A \sim M_A$ )
- ✓ Heavy nuclei



A new force between electrons and nucleons will cause a “King plot” non-linearity

[Berengut et al. Phys Rev Lett 120, 091801 (2018)]

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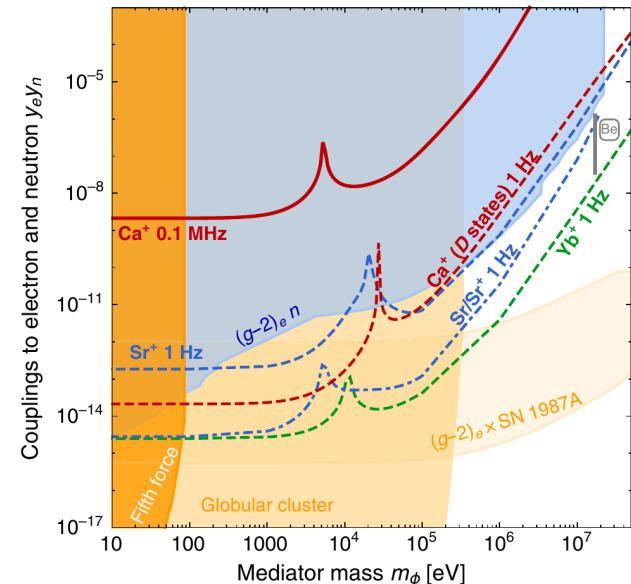
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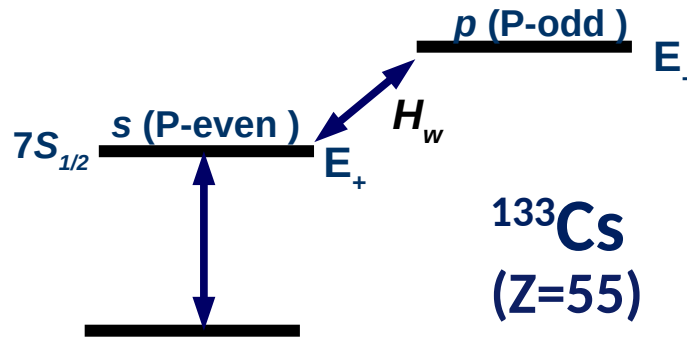
# Contents

- Precision studies in atoms and molecules
- Nuclear structure / nuclear matter / new forces & particles
- **Parity violation**
- Parity and time-reversal violation
- First results: RaF
- Summary

# Parity violation

## Atoms

PV → Mix states of different parity



$^{133}\text{Cs}$   
( $Z=55$ )

$$E_{PNC} \sim \frac{\langle \text{P-odd} | H_w | \text{P-even} \rangle}{E_- - E_+}$$

$$EI_{APV} = \langle 7\tilde{S}_{1/2} | D | 6\tilde{S}_{1/2} \rangle = k Q_w$$

measure atomic calculation

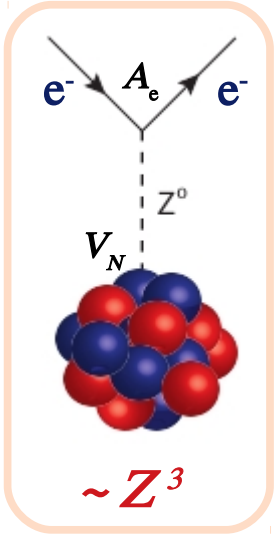
$$\text{Expt: } Q_w(^{133}\text{Cs}) = -72.06(28)_{\text{exp}}(34)_{\text{th}}$$

$$Q_w = -N + (1 - 4 \sin^2 \theta_w) Z$$

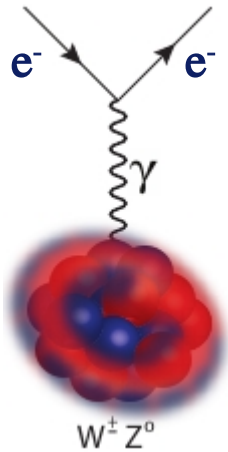
[ Wood et al. Science 275, 1759 (1997) ]

[ Porsev et al. PRL 102, 181601 (2009) ]

$$H_w = Q_w \frac{G_F}{\sqrt{8}} \gamma_5 \rho(r)$$



$\sim Z^3$



$W^\pm Z^0$

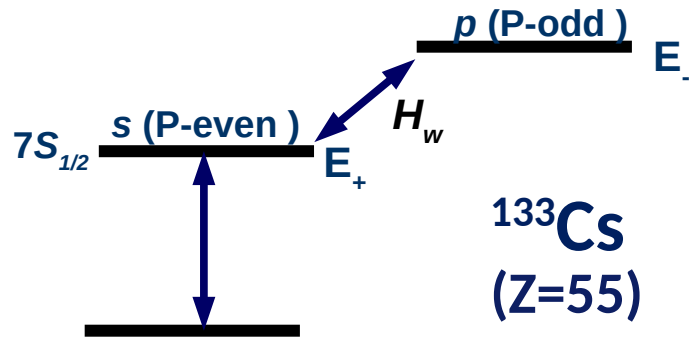
$\sim Z^2 A^{2/3} R(Z)$



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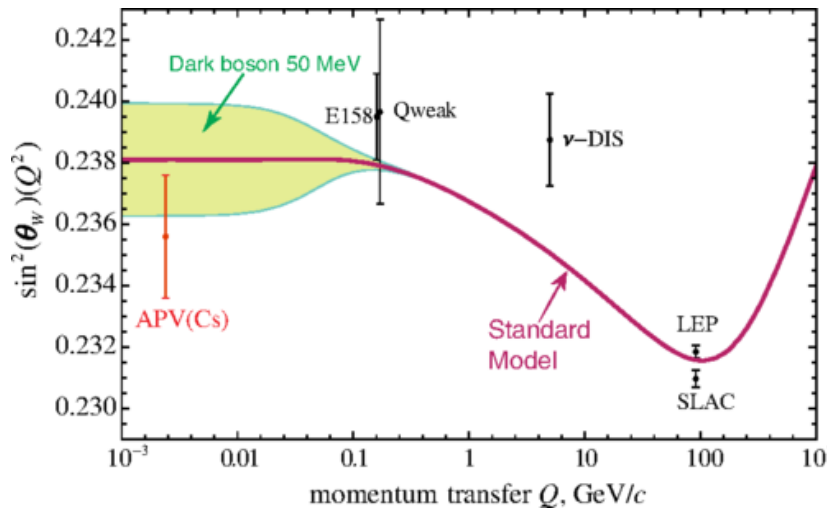
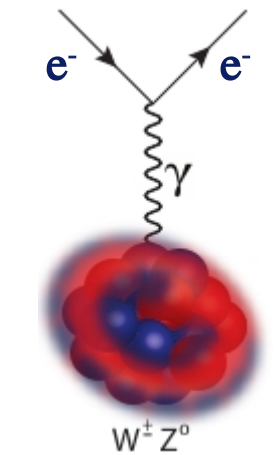
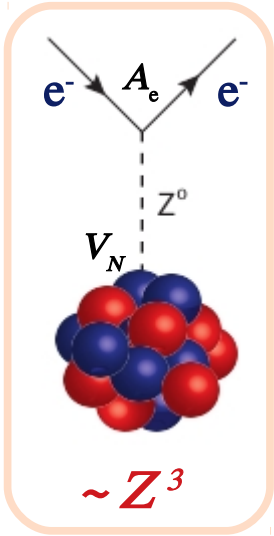
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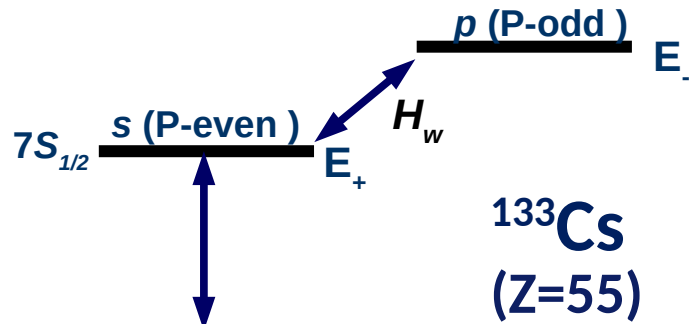
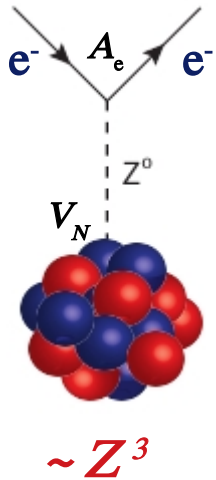
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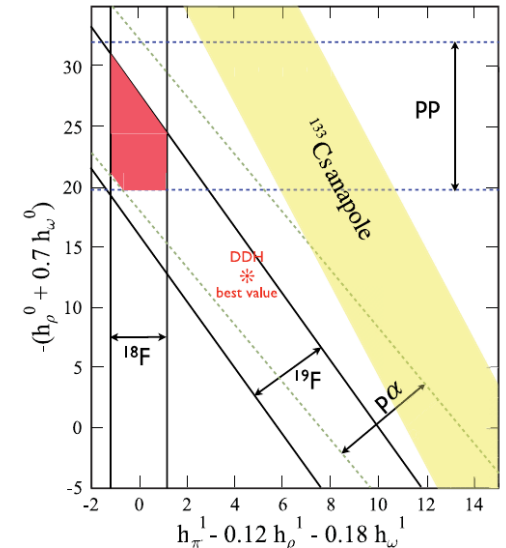
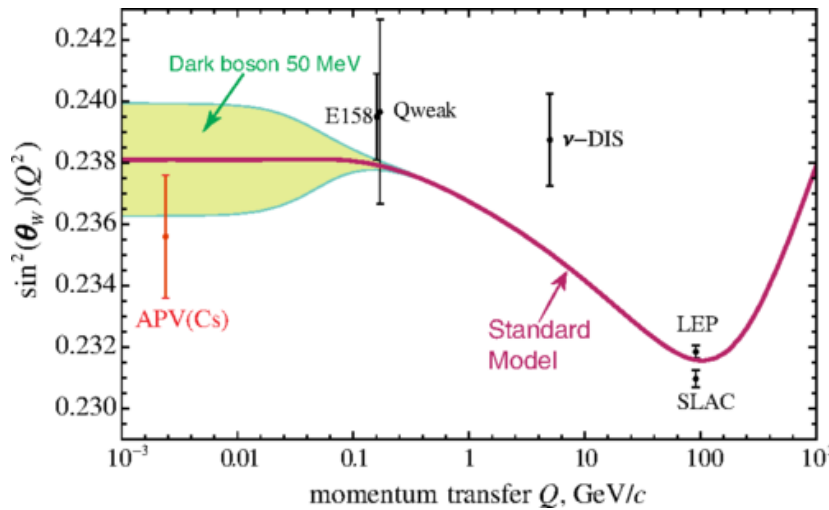
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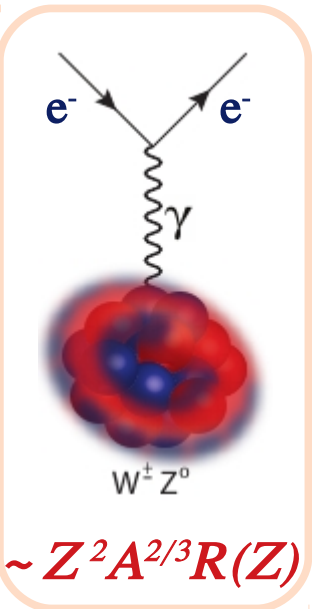
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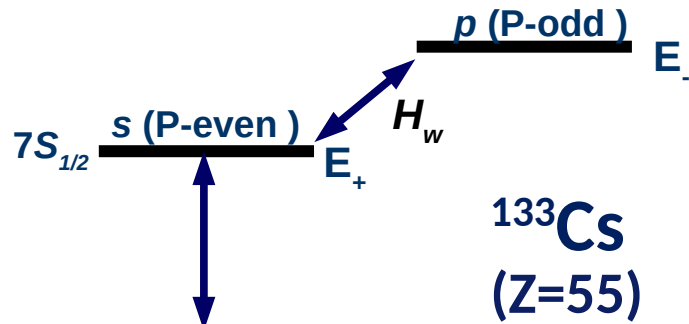
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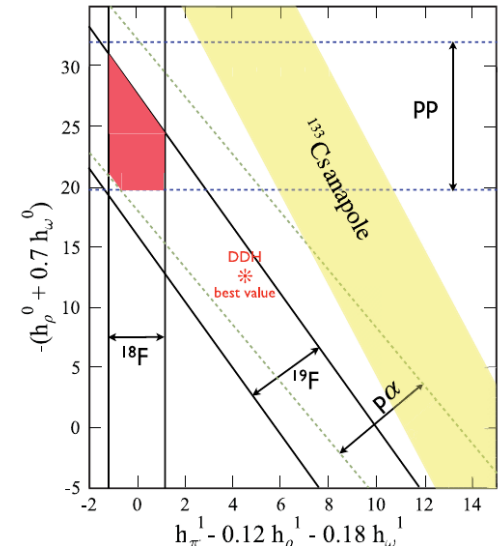
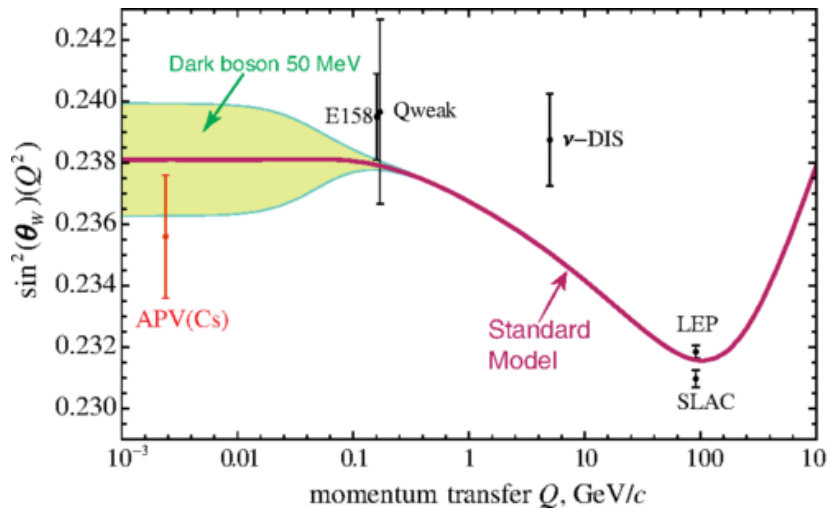
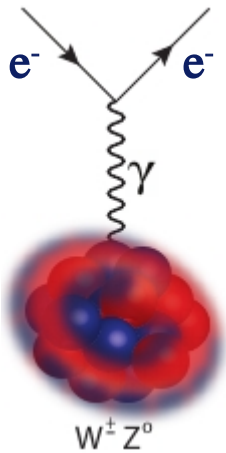
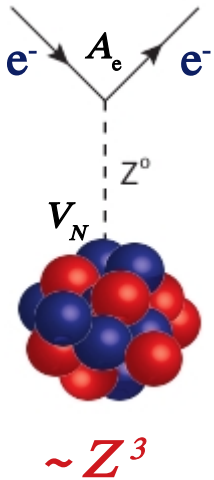
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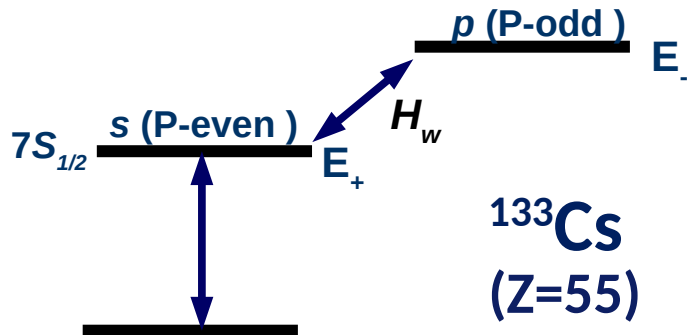
Yb ( $Z=70$ ) stable @ Mainz → Isotopic ratios [ Nature Phys. 15, 120 (2019) ]

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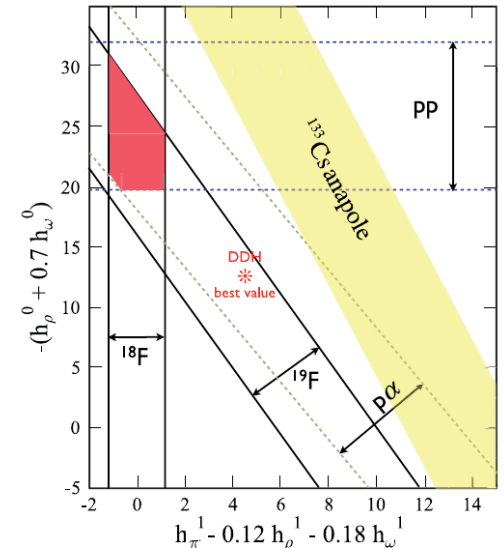
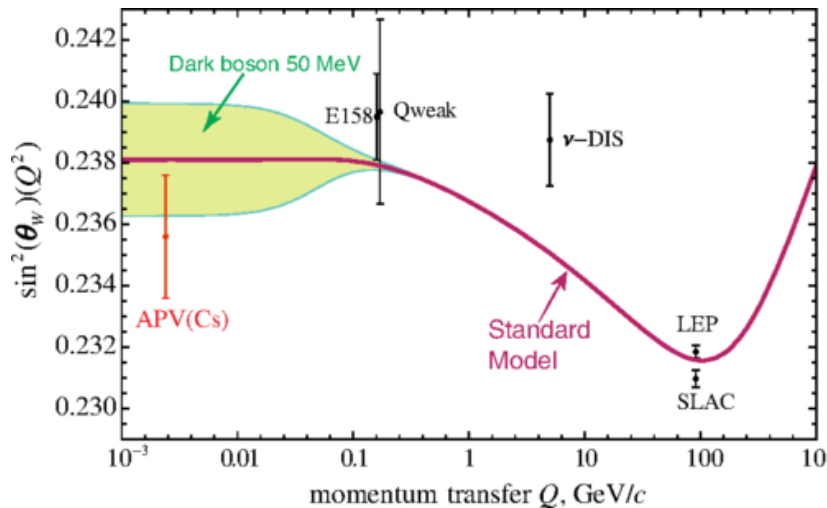
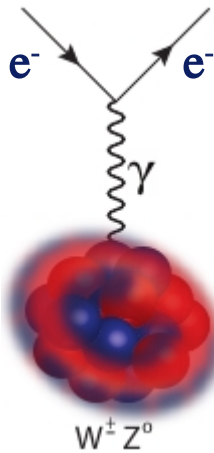
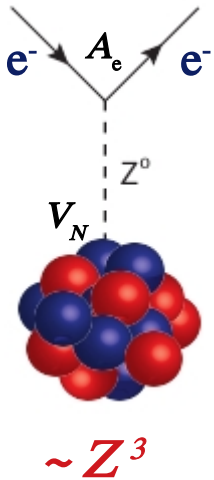
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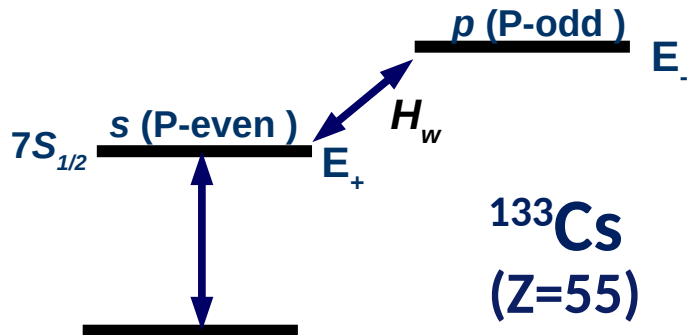
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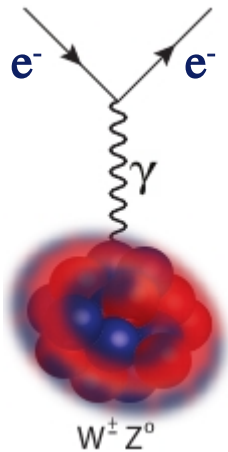
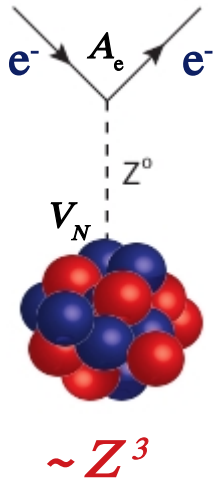
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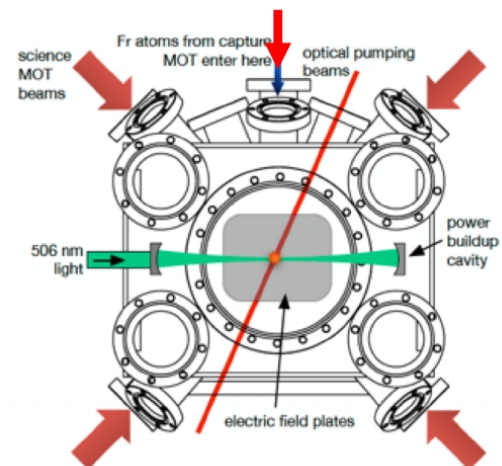
[ Porsev et al. PRL 102, 181601 (2009) ]



Fr (z=87) @ TRIUMF

[Zang et al. Phys. Rev. Lett. 115, 042501 (2015)]

[Kalita et al. Phys. Rev. A 97, 042507 (2018)]



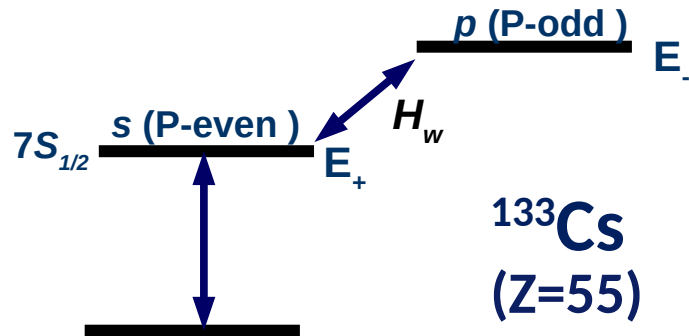
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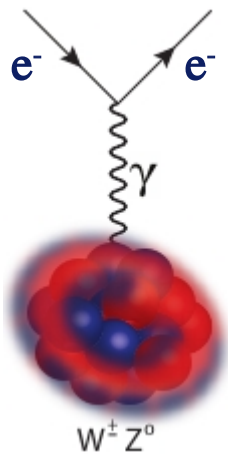
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Molecules

Why (exotic) molecules?



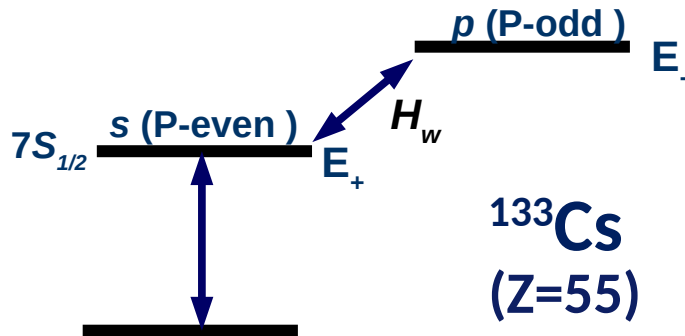
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# Parity violation

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## Molecules

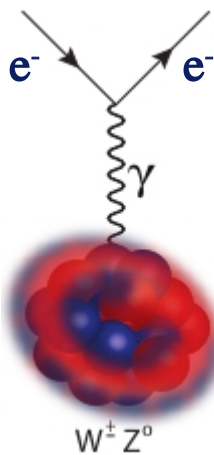
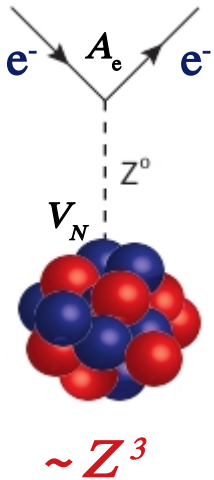
## Why (exotic) molecules?

- ✓  $(E_+ - E_-) \sim 0$  About  $10^3$  enhancement

[ Dzuba et al. Phys. Rev. Lett. 119, 223201 (2017) ]

- ✓ Nuclear-Spin-Dependent PV / Anapole moment (BaF)

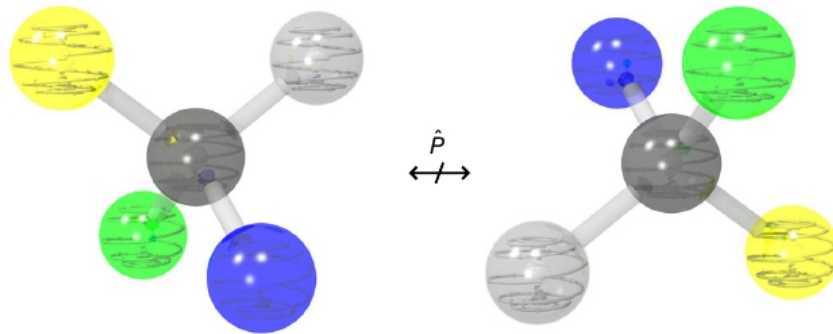
[ Altunas et al. Phys Rev Lett 120, 142501 (2018) ]



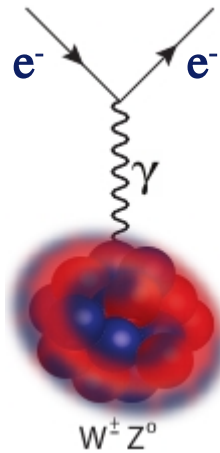
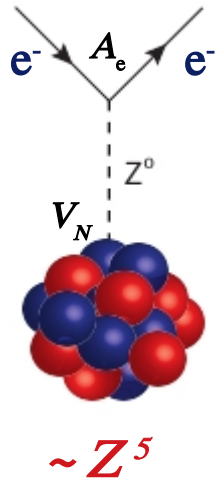
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# Parity violation

## Chiral molecules



[Berger & Stohner. WIREs Comput. Mol. Sci. e1396, 1 (2018)]  
 [Laerdahl et al. Phys. Rev. Lett. 84, 3811 (2000)]



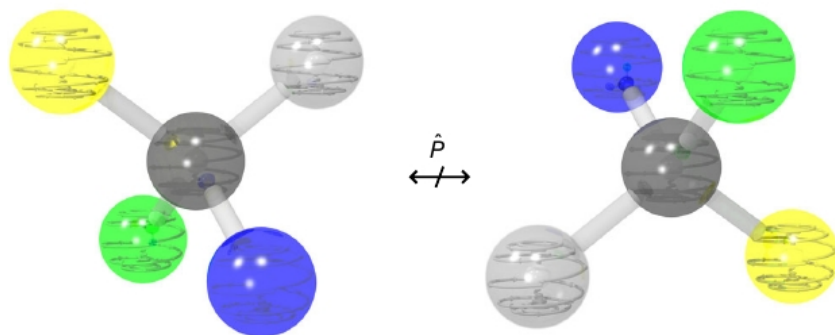
$$H_W = Q_W \frac{G_F}{\sqrt{8}} \gamma_5 \rho(r)$$

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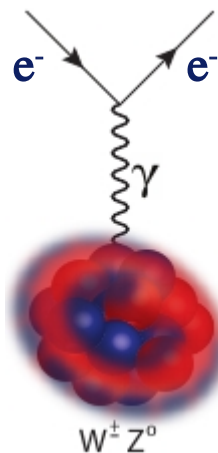
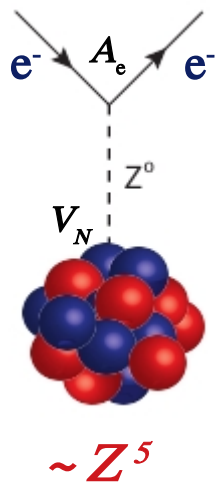
$\Delta\nu_{\text{pv}}/\nu$  ( $\times 10^{-16}$ )

Molecule	HF	B3LYP	B-LYP	LDA
CHBrClF	+0.72	+1.01	+0.93	+0.98
CHBrFI	+17.8	+22.9	+22.1	+24.1
CHClFI	+9.35	+8.96	+6.94	+7.38
CHAtFI	-1072	-1008	-850	-946

[Berger et al. Mol. Phys. 105, 41 (2007)]

$$H_W = Q_W \frac{G_F}{\sqrt{8}} \gamma_5 \rho(r)$$

$^{211}\text{At}$  ( $Z=85$ ,  $T_{1/2} \sim 7.2$  h)  $\rightarrow$  Freq. shift  $\sim 3$ -10 Hz



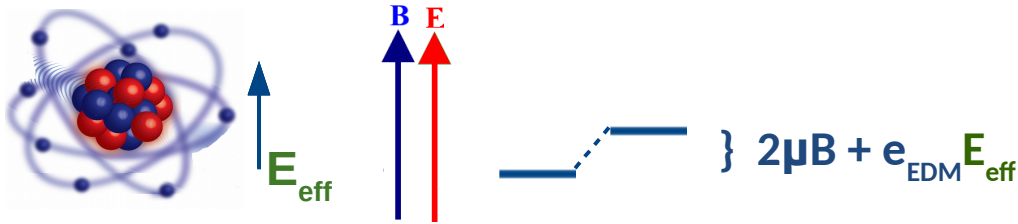
$\sim Z^2 A^{2/3} R(Z)$

# Contents

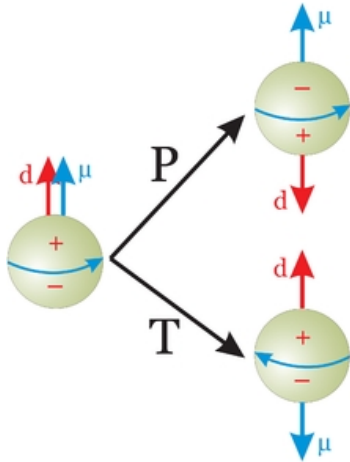
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- Nuclear structure / nuclear matter / new forces & particles
- Parity violation
- **Parity and time-reversal violation**
- First results: RaF
- Summary

# Parity and Time-reversal Violation

## Atoms

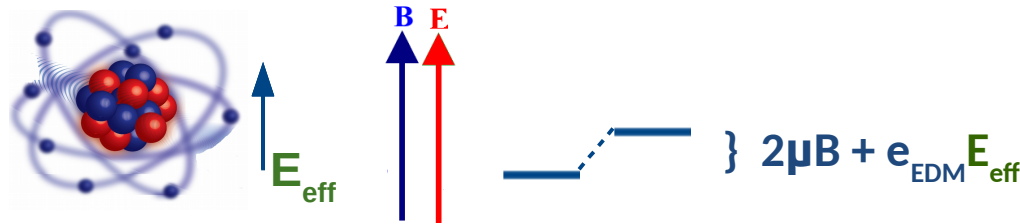


$$\sigma_d \sim \frac{1}{E_{\text{eff}} \tau \sqrt{NT}}$$



# Parity and Time-reversal Violation

## Atoms



$$\sigma_d \sim \frac{1}{E_{\text{eff}} \tau \sqrt{NT}}$$

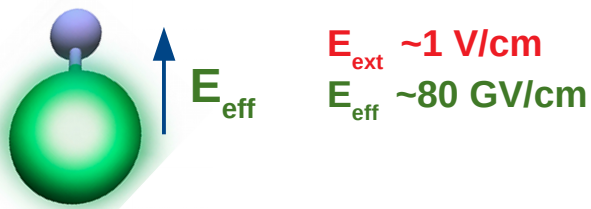
## Molecules

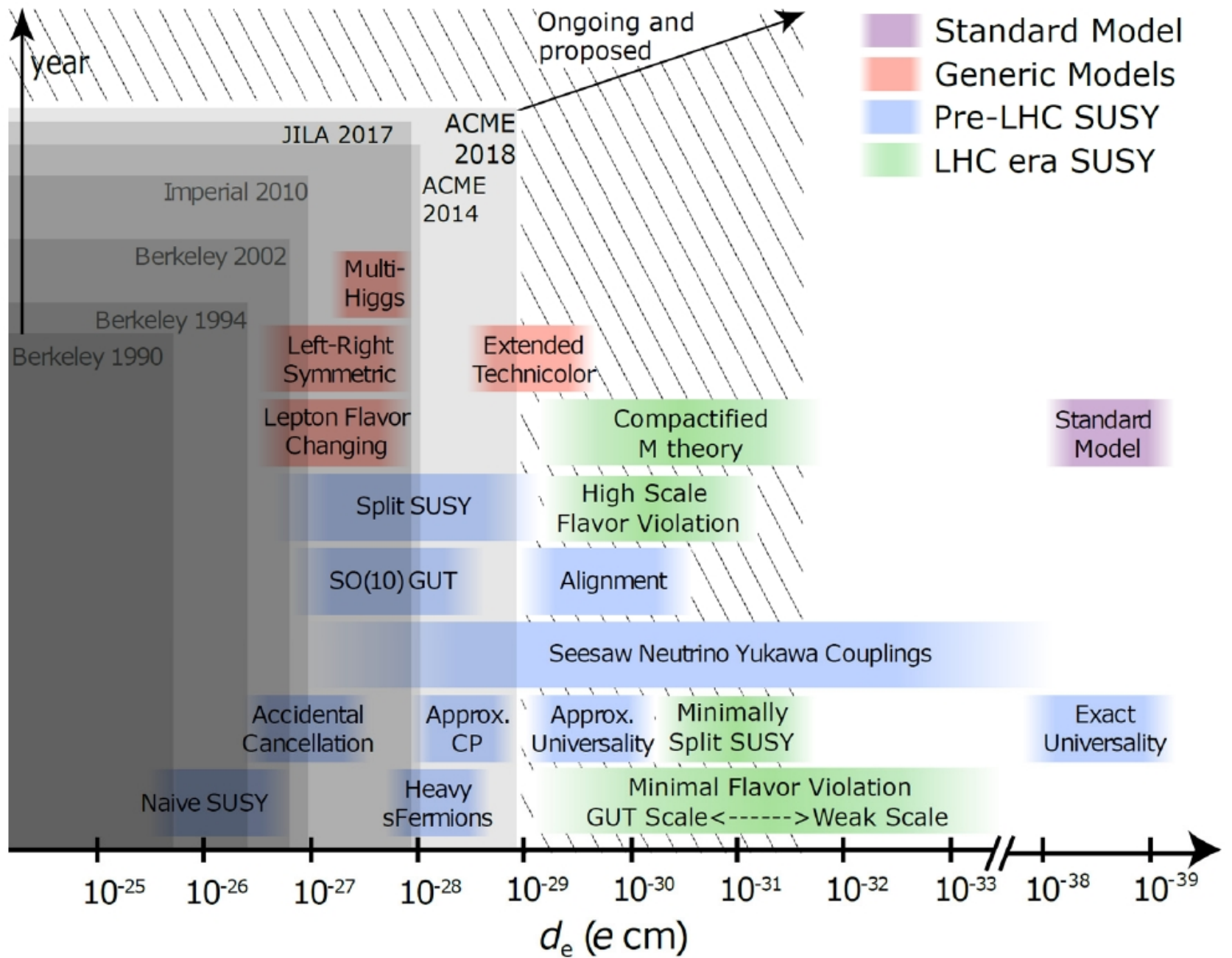
- ✓  $>10^3$  enhancement of  $E_{\text{eff}}$  for EDM measurements

[ACME, Nature 562, 355 (2018)]

[Baron et al. Science 343, 269 (2014)]

[Sandars Phys. Rev. Lett. 18, 1396 (1967)]





[Source: D. DeMille. Manipulating Quantum Systems: An Assessment of Atomic, Molecular, and Optical Physics in the United States (2019)]

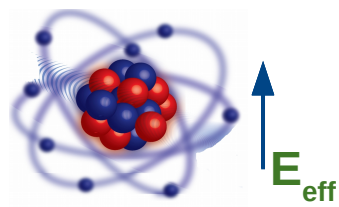
# Why radioactive nuclei?

- ✓ Large  $Z \rightarrow E_{\text{ef}} (>>)$
- ✓ Deformed nuclei ( $>10^2$ )
- ✓ Large enhancement of the MQM ( $>10^2$ )
- ✓ Study of NSD interactions ( $I>0$ )

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

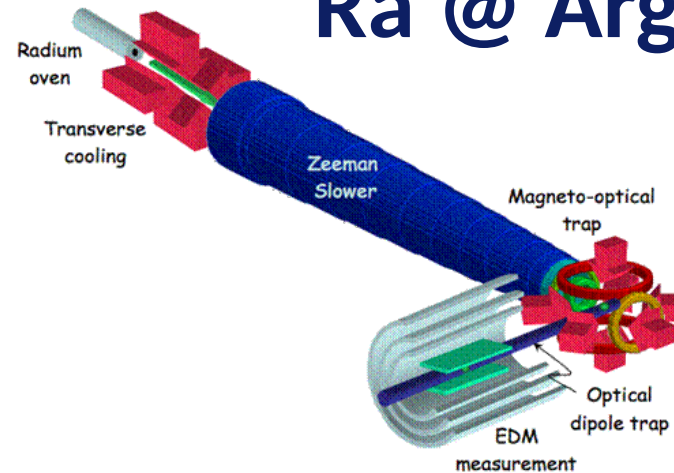
# Why radioactive nuclei?

Atoms



$$\sigma_d \sim \frac{1}{E_{\text{eff}} \tau \sqrt{NT}}$$

## Ra @ Argonne



$$|d(^{225}\text{Ra})| < 5.0 \times 10^{-22} \text{ e cm}$$

Phys. Rev. Lett. 114, 233002 (2015)

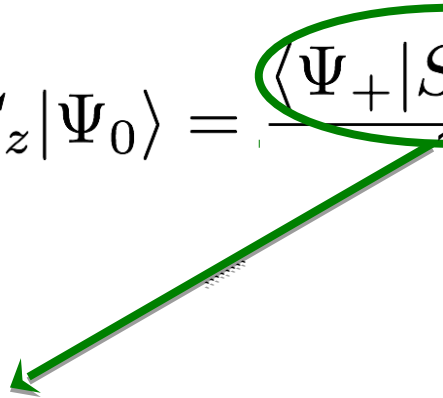
# Why radioactive nuclei?

P,T-odd nucleon-nucleon interaction

Schiff Moments

$$S = \langle \Psi_0 | S_z | \Psi_0 \rangle = \frac{\langle \Psi_+ | S_z | \Psi_- \rangle \langle \Psi_+ | V_{PT} | \Psi_- \rangle}{E_+ - E_-}$$

Nuclear structure



Experiment



$\Delta E$ : Energy splitting of opposite parity states



# Why radioactive nuclei?

P,T-odd nucleon-nucleon interaction

## Schiff Moments

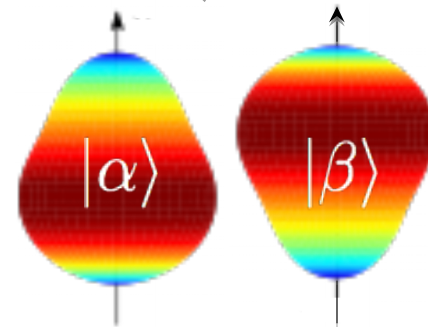
$$S = \langle \Psi_0 | S_z | \Psi_0 \rangle = \frac{\langle \Psi_+ | S_z | \Psi_- \rangle \langle \Psi_+ | V_{PT} | \Psi_- \rangle}{E_+ - E_-}$$

Nuclear structure

Theory + Experiment

Experiment

$\Delta E$ : Energy splitting of opposite parity states



$^{225}\text{Ra}$

$\Delta E = 55 \text{ keV}$



Slide from L. Gaffney

[Gaffney et al. Nature 497, 199 (2013)]

# Why radioactive nuclei?

P,T-odd nucleon-nucleon interaction

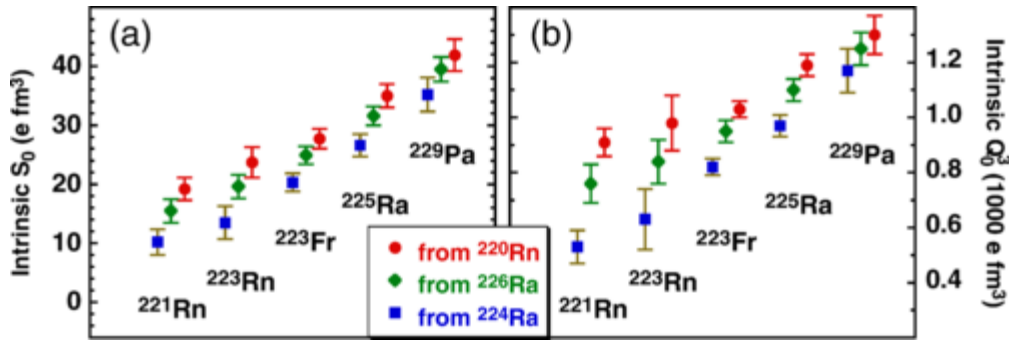
## Schiff Moments

$$S = \langle \Psi_0 | S_z | \Psi_0 \rangle = \frac{\langle \Psi_+ | S_z | \Psi_- \rangle \langle \Psi_+ | V_{PT} | \Psi_- \rangle}{E_+ - E_-}$$

Nuclear structure

Theory + Experiment

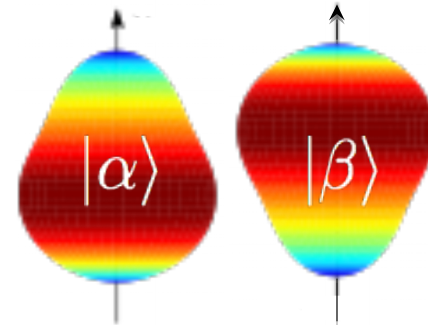
Experiment



Dobaczewski et al. Phys. Rev. Lett. **121**, 232501 (2018)  
 Chupp et al. Rev. Mod. Phys. **91**, 015001 (2019)

$$\hat{S}_0 = \frac{e}{10} \sqrt{\frac{4\pi}{3}} \sum_i \left( r_i^3 - \frac{5}{3} r_{ch}^2 r_i \right) Y_0^1(\Omega_i) + \dots$$

$\Delta E$ : Energy splitting of opposite parity states



$^{225}\text{Ra}$

$\Delta E = 55 \text{ keV}$



Slide from L. Gaffney

[Gaffney et al. Nature 497, 199 (2013)]

# Contents

- Precision studies in atoms and molecules
- Nuclear structure / nuclear matter / new forces & particles
- Parity violation
- Parity and time-reversal violation
- **First results: RaF**
- Summary

# RaF → Nuclear x Molecular Enhancement

- ✓ Large  $Z \rightarrow E_{ef} (>>)$
- ✓ Deformed nuclei ( $>10^2$ )
- ✓ Large enhancement of the MQM ( $>10^2$ )
- ✓ Study of NSD interactions ( $I>0$ )



## P- and P,T- odd effects

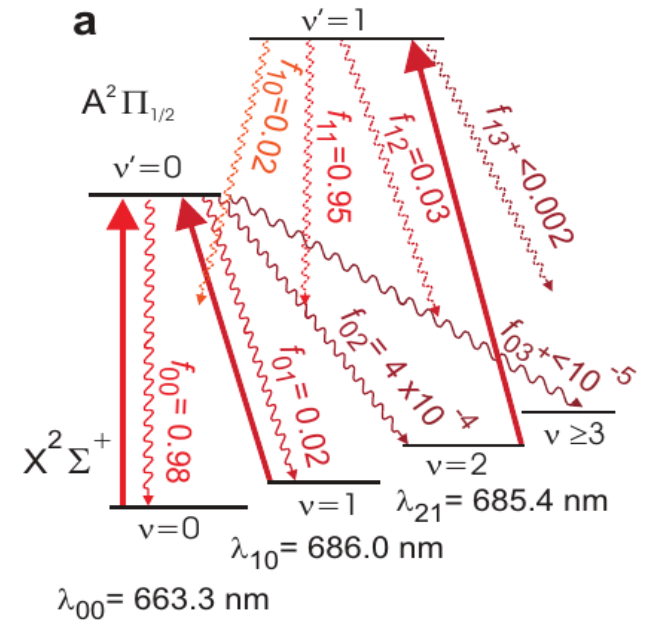
Enhanced sensitivity in radioactive molecules  
(composed of heavy and octupole deformed nuclei)

### Molecules: Electroweak structure

- **Anapole moment: AM**
- **Magnetic Quadrupole Moment: MQM**
- **Schiff Moment:  $S_{\text{schiff}}$**
- **eEDM, nEDM, ...**

# Fluoride molecules

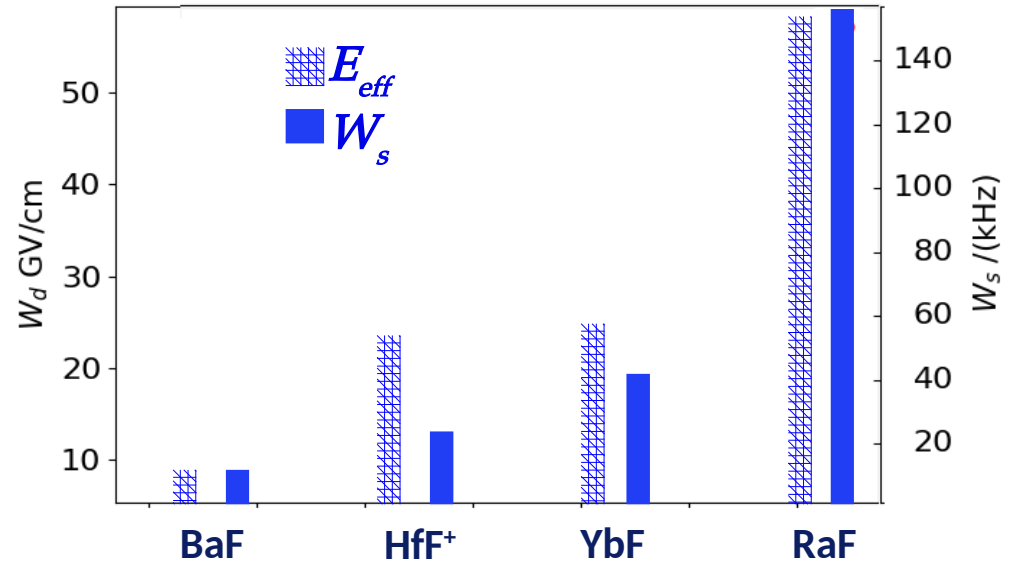
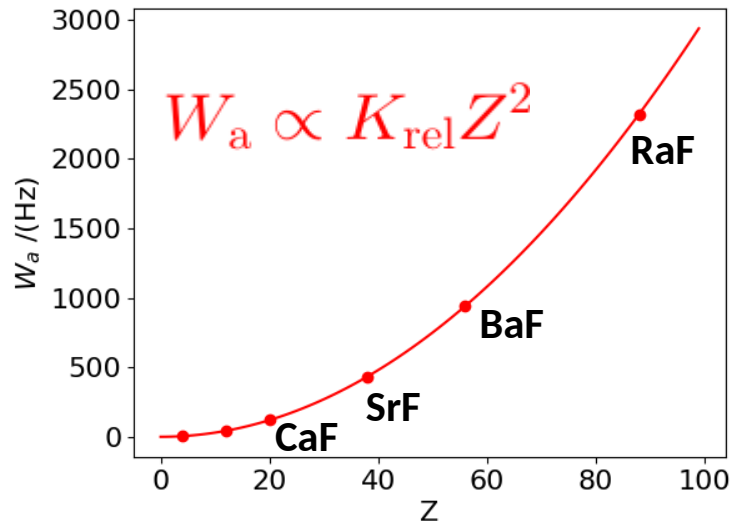
- SrF → First evidence of laser cooling [Nature 467, 820-823 (2010)]
- YbF → Nature 473, 493 (2011)
- .
- .
- .
- .
- .
- .
- .
- SrF → Nature Physics 13, 1173(2017)
- YbF → Phys. Rev. Lett. 120, 123201 (2018)
- CaF → Nature Physics 14, 890 (2018)  
Phys. Rev. Lett. 120, 163201 (2018)
- RaF → **Radioactive**  
*Coming soon ...*



# Radioactive Molecules: Ra(z=88)F Results

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

## P-odd and P,T -odd effects



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

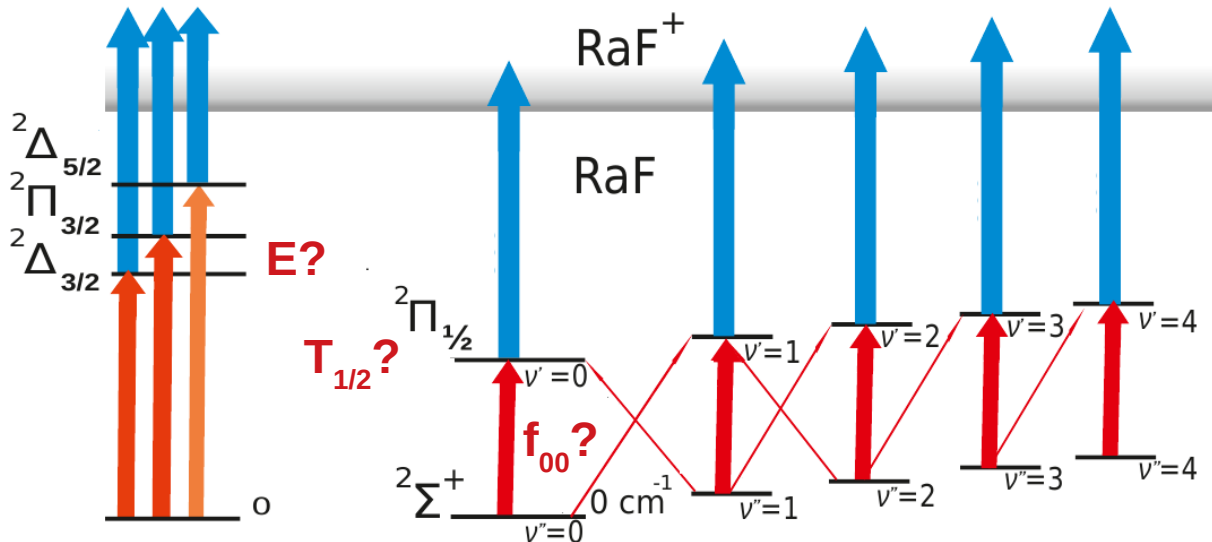
RaF → Superior sensitivity for both P- and P,T- odd effects

... BUT all parameters experimentally unknown!

# Results: Radium fluoride (RaF)

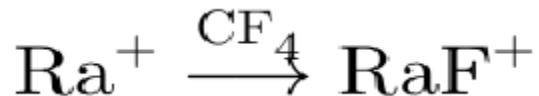
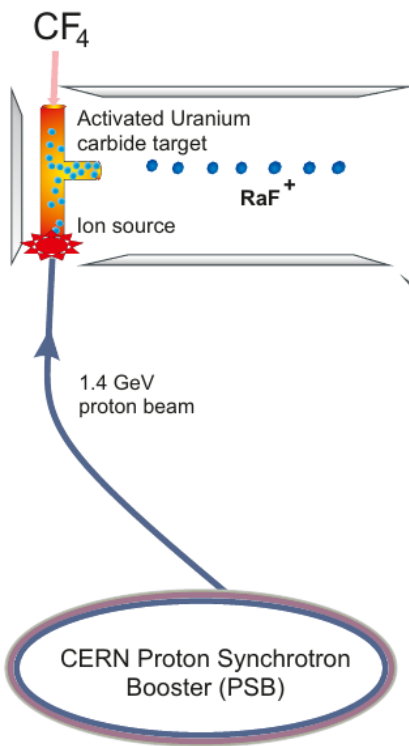
*Collinear resonance ionization spectroscopy of RaF molecules*  
[Garcia Ruiz, Berger et al. CERN-INTC-2018-017 (2018)]

- I. Low-lying structure?
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  1. Dominant  **$f_{00}$** ?
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# Results: Radium fluoride (RaF)

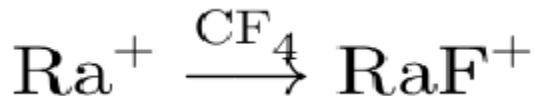
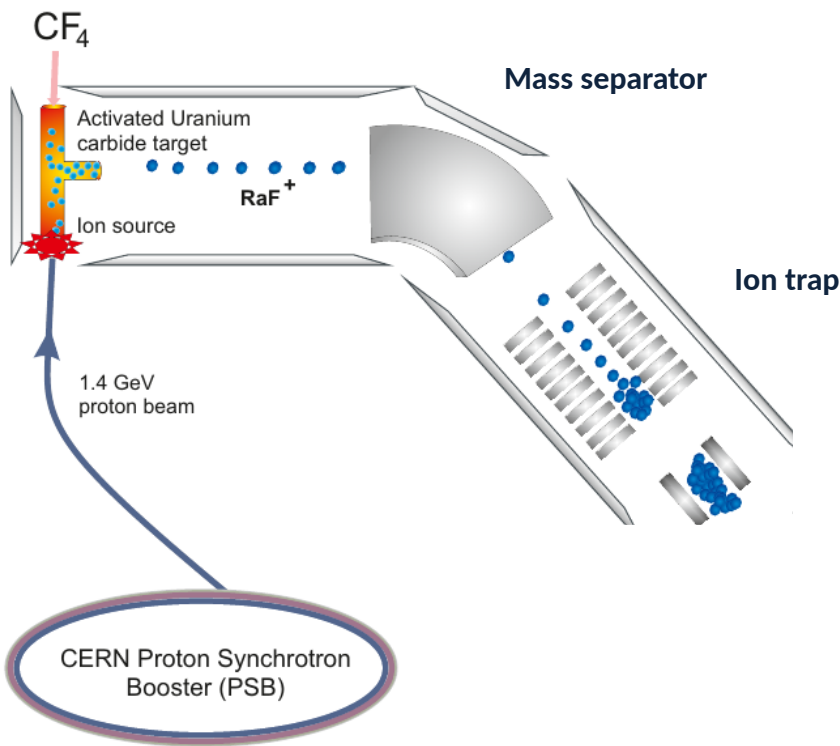
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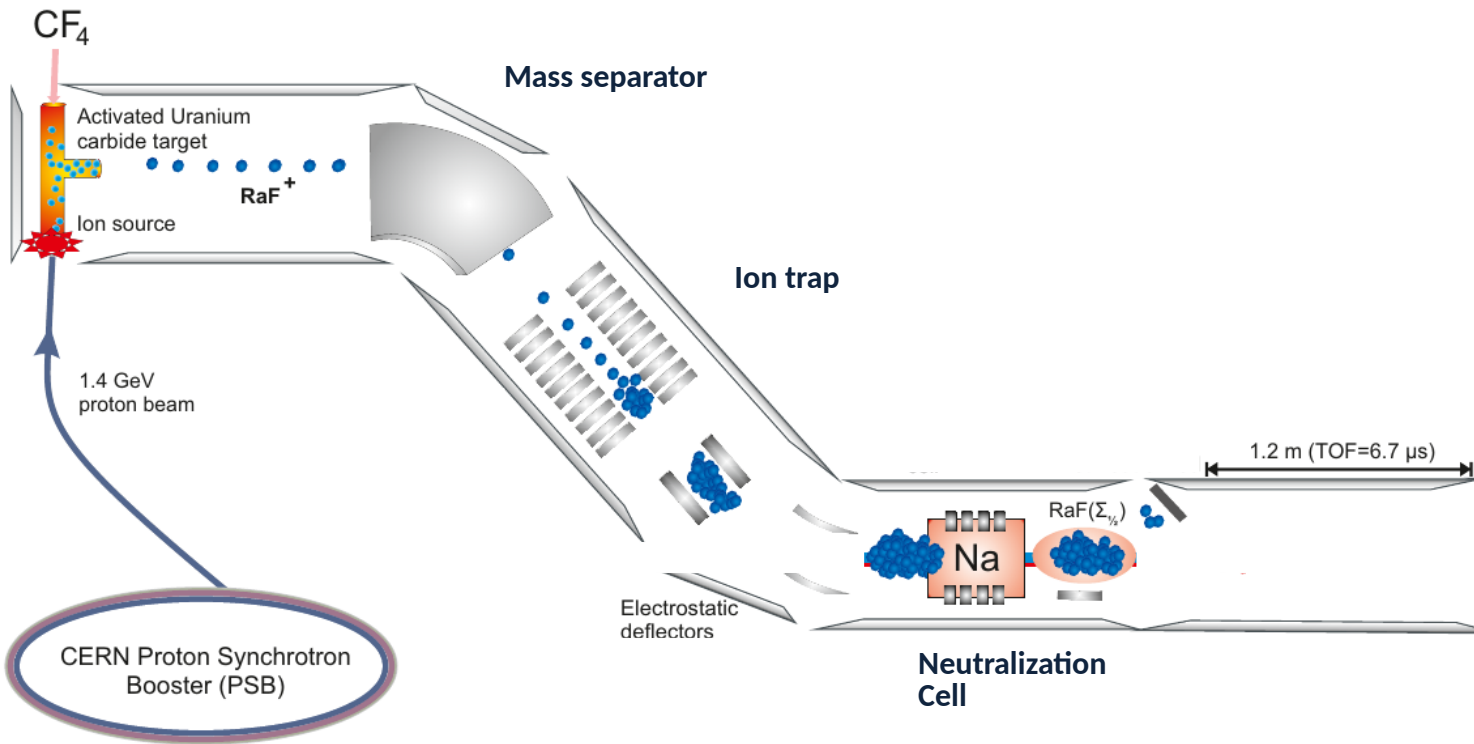
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*Collinear resonance ionization spectroscopy of RaF molecules*  
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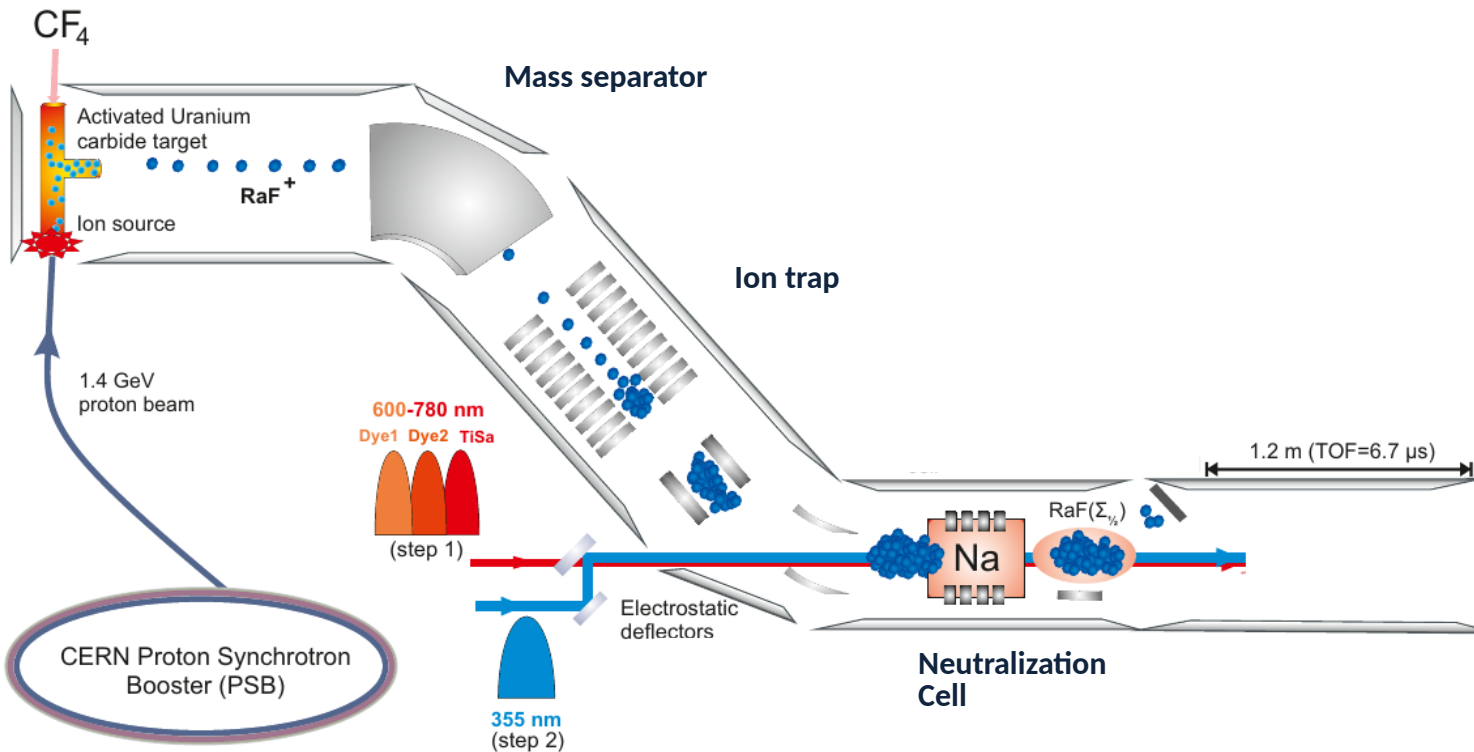
# Results: Radium fluoride (RaF)

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



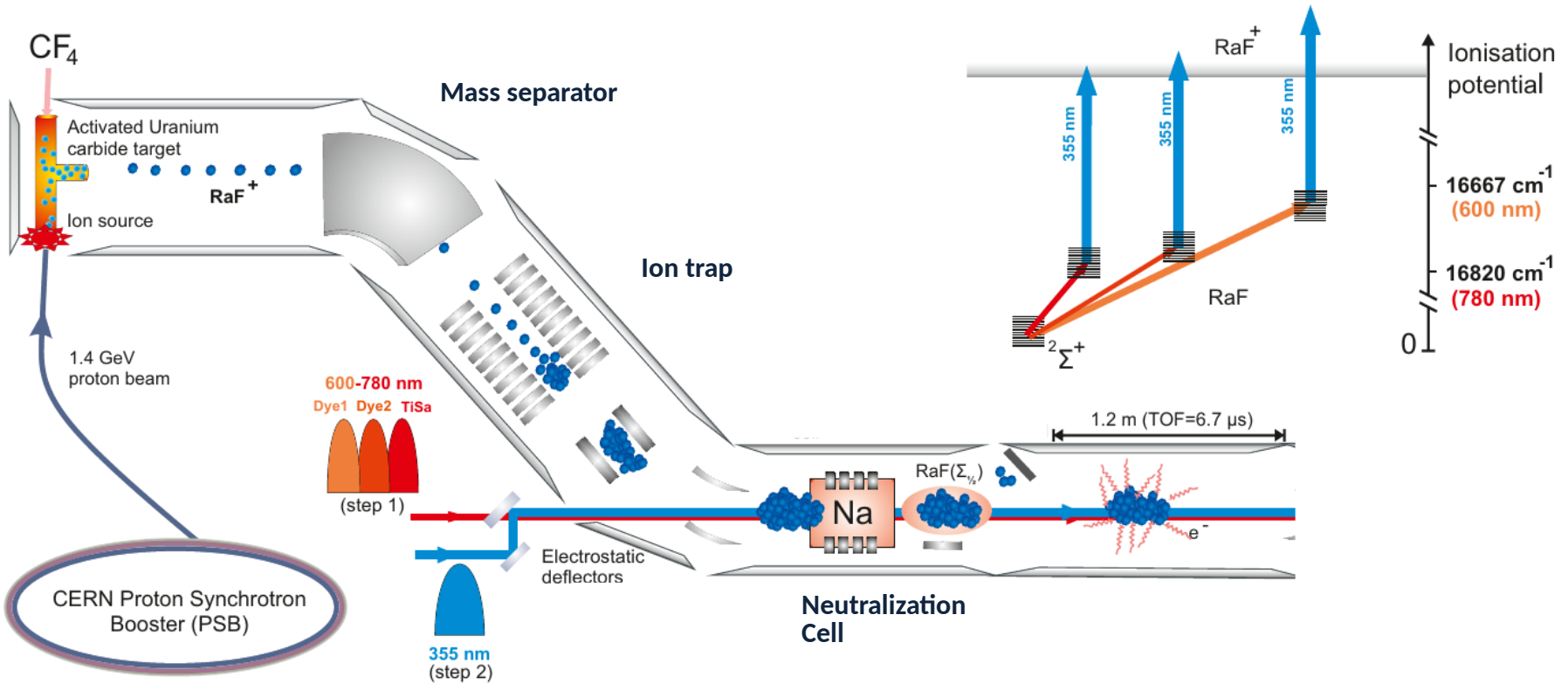
# Results: Radium fluoride (RaF)

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



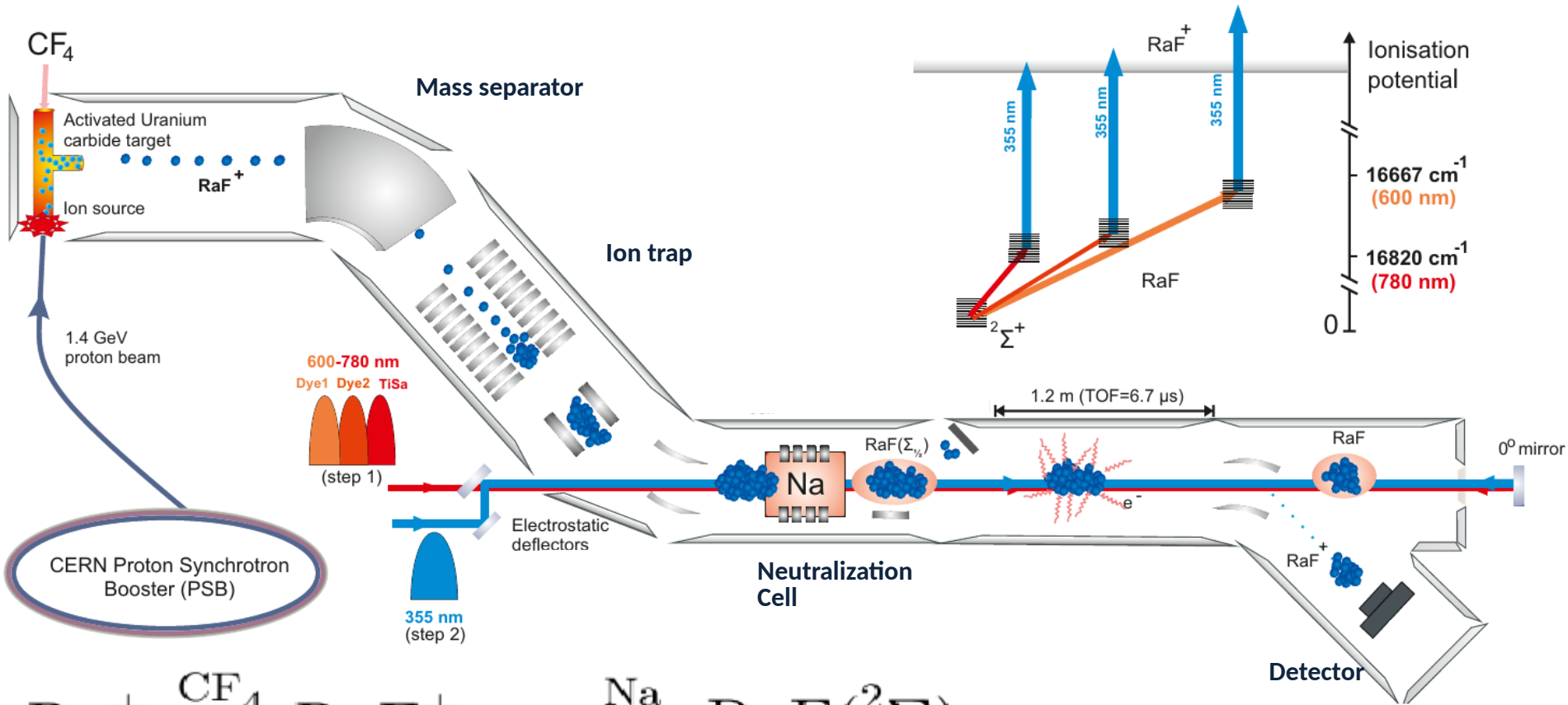
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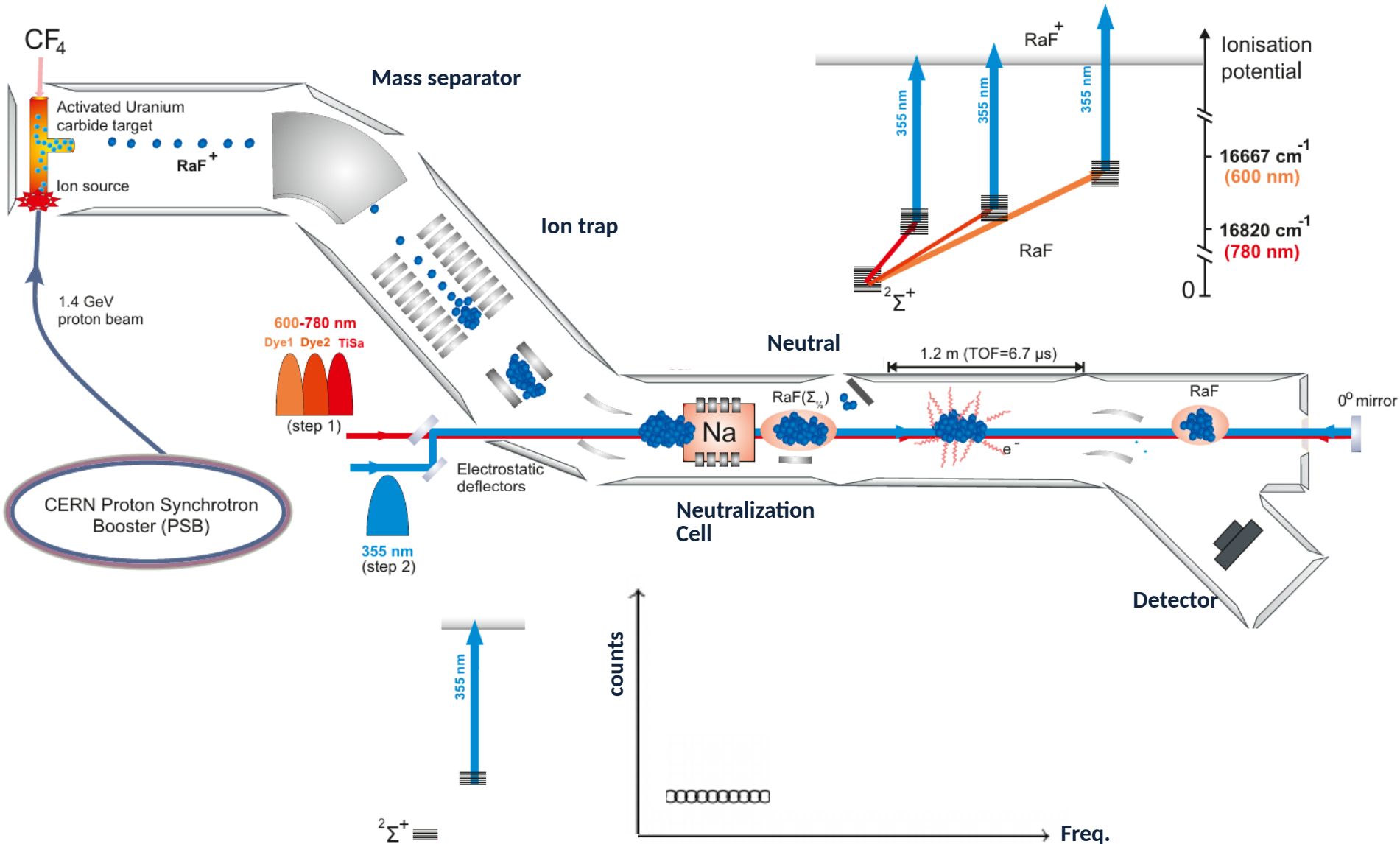
# Results: Radium fluoride (RaF)

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



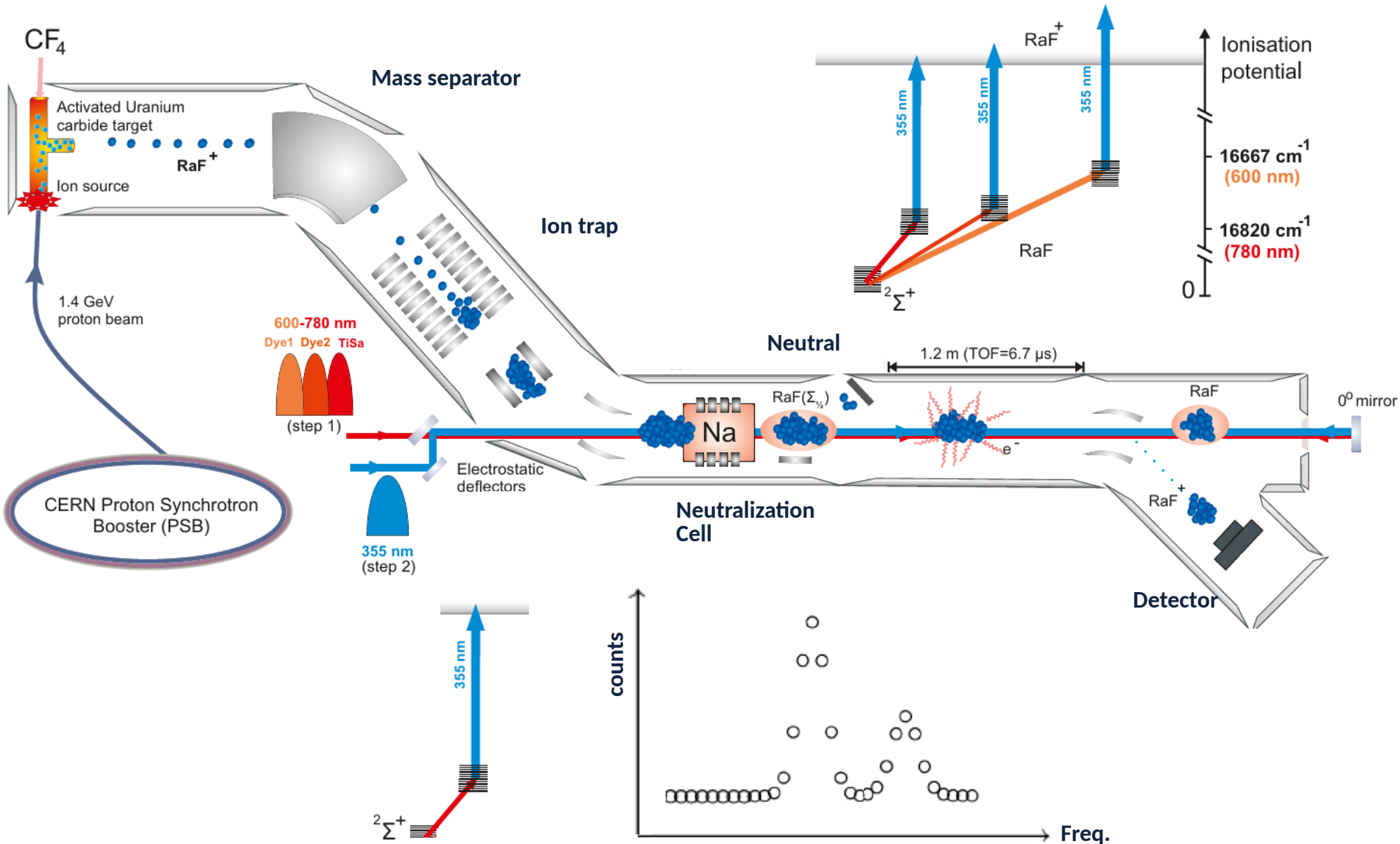
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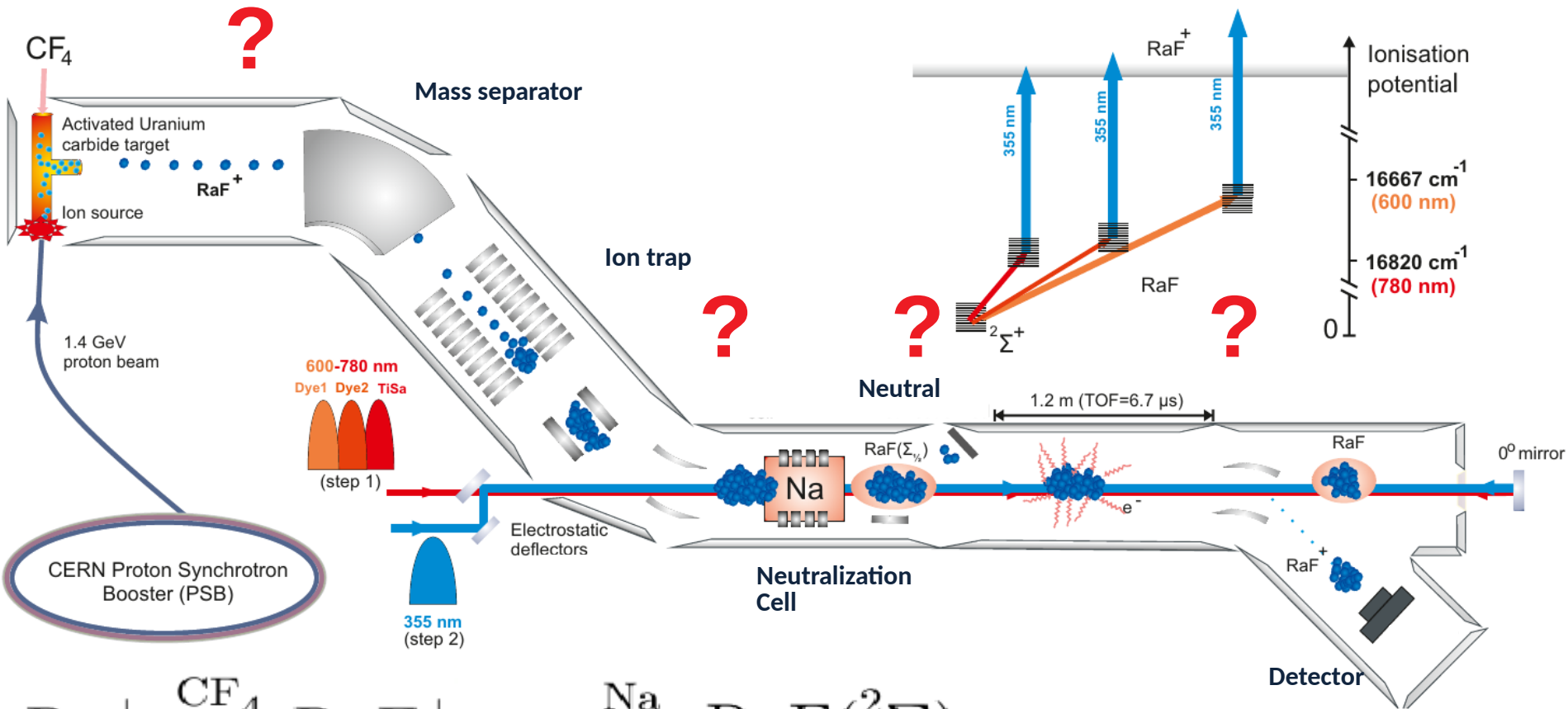
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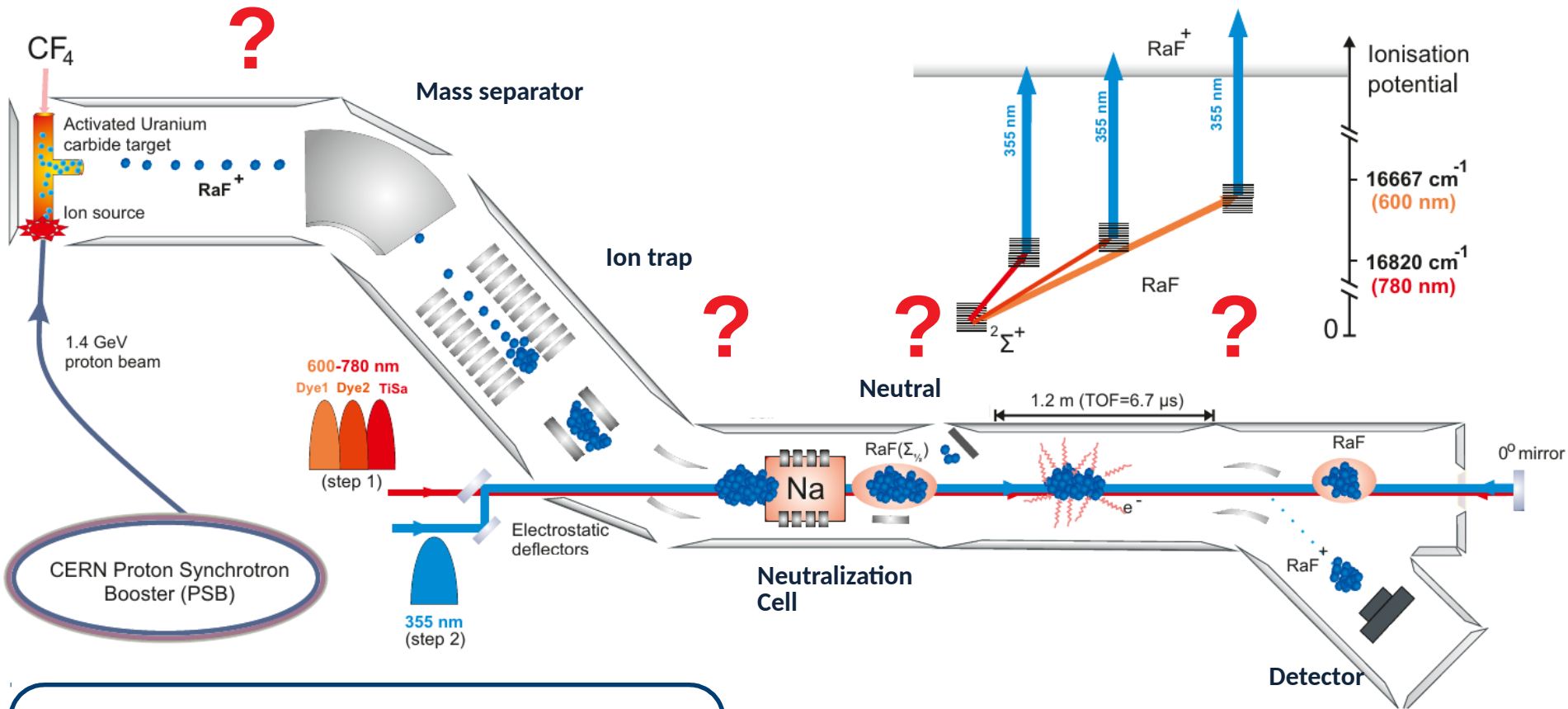
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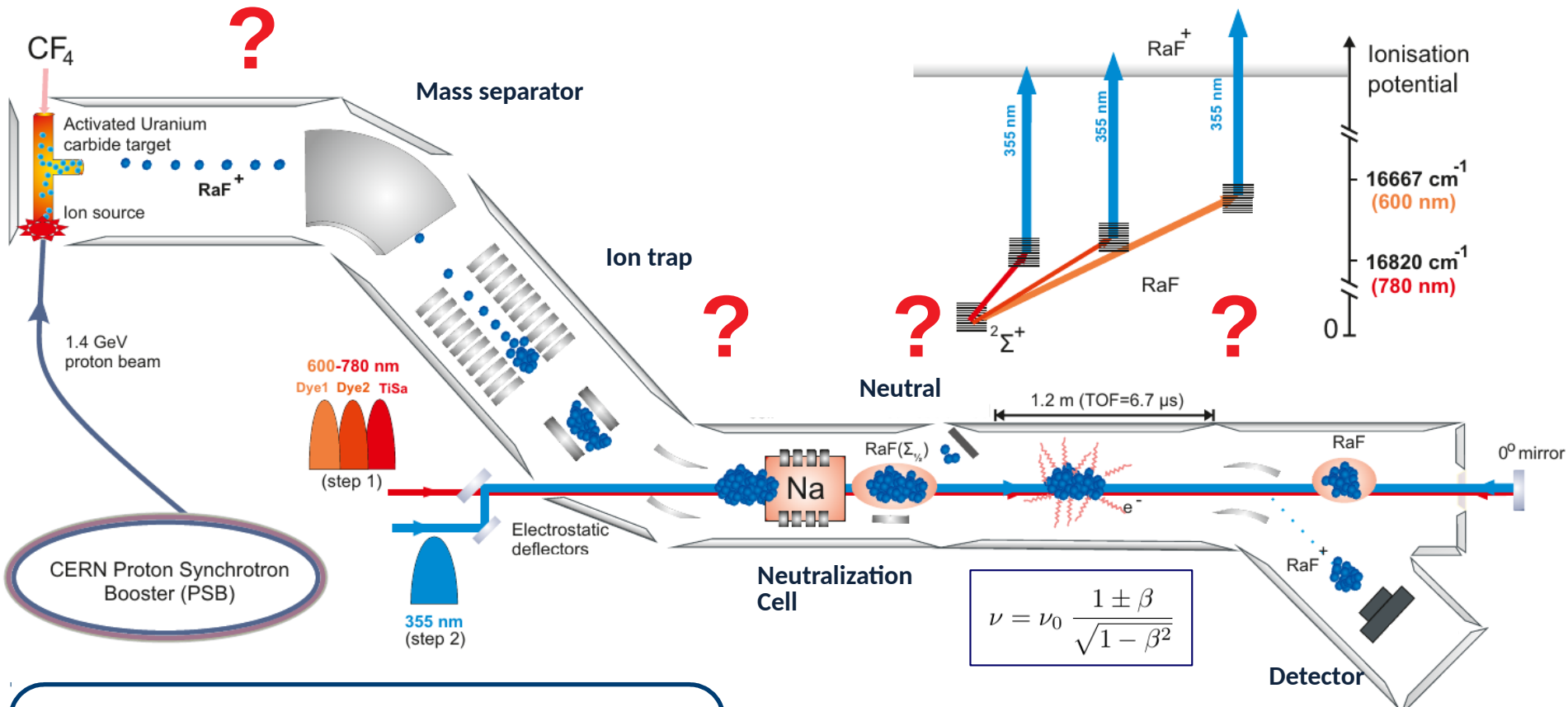
*Collinear resonance ionization spectroscopy of RaF molecules*  
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Molecules have complex structures  
→ More than  $10^4$  states can be populated  
*Vibrational* / *rotational* / *hyperfine*  
Impossible with a hot ( $> 300$  K) molecule?

# Results: Radium fluoride (RaF)

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

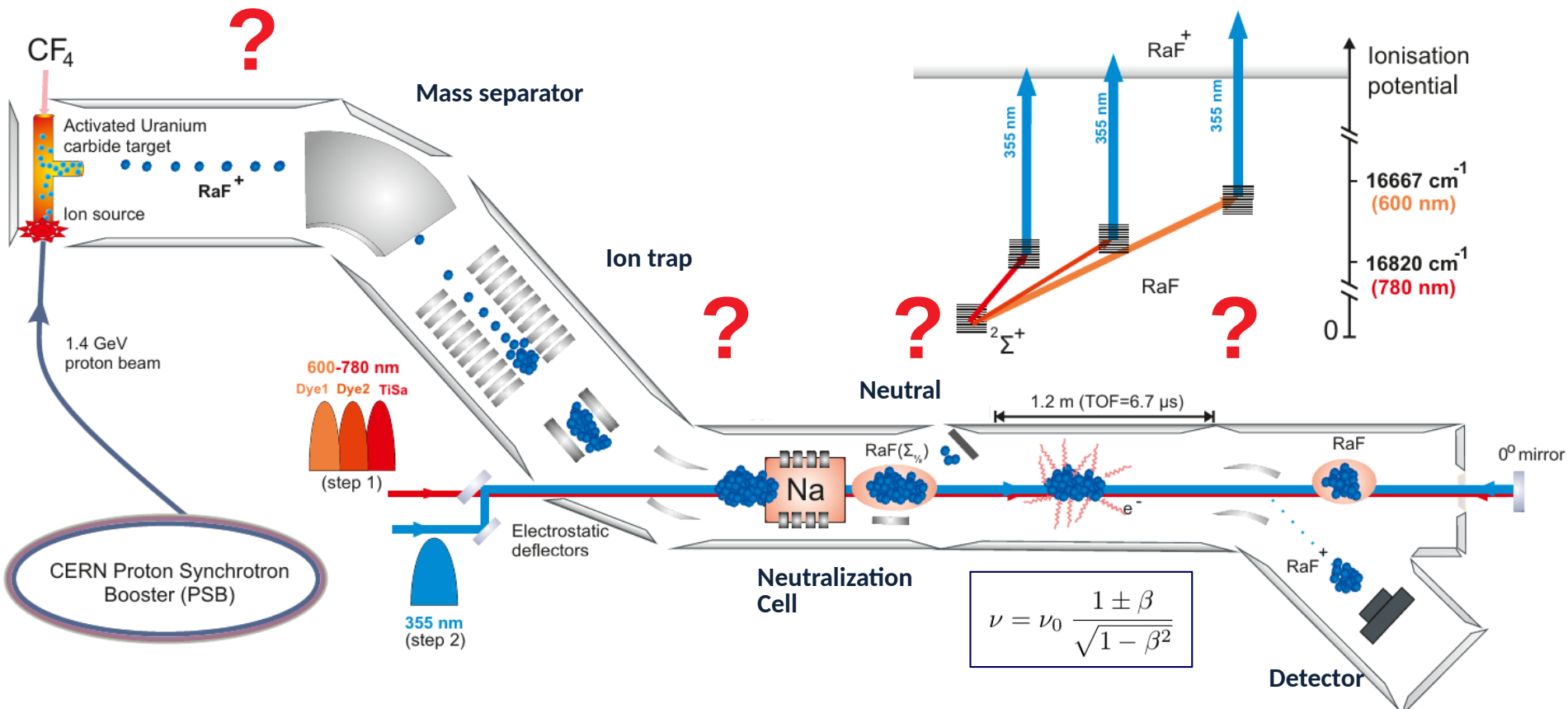


Molecules have complex structures  
 → More than  $10^4$  states can be populated  
*Vibrational / rotational / hyperfine*  
 Impossible with a hot (> 300 K) molecule?

Theory:  $13300(1000)\text{cm}^{-1}$   
 Scanning  $1000\text{cm}^{-1}$  at  $100\text{MHz/min}$  ( $1\text{cm}^{-1} = 30\text{GHz}$ )  
 → **208 days!!**

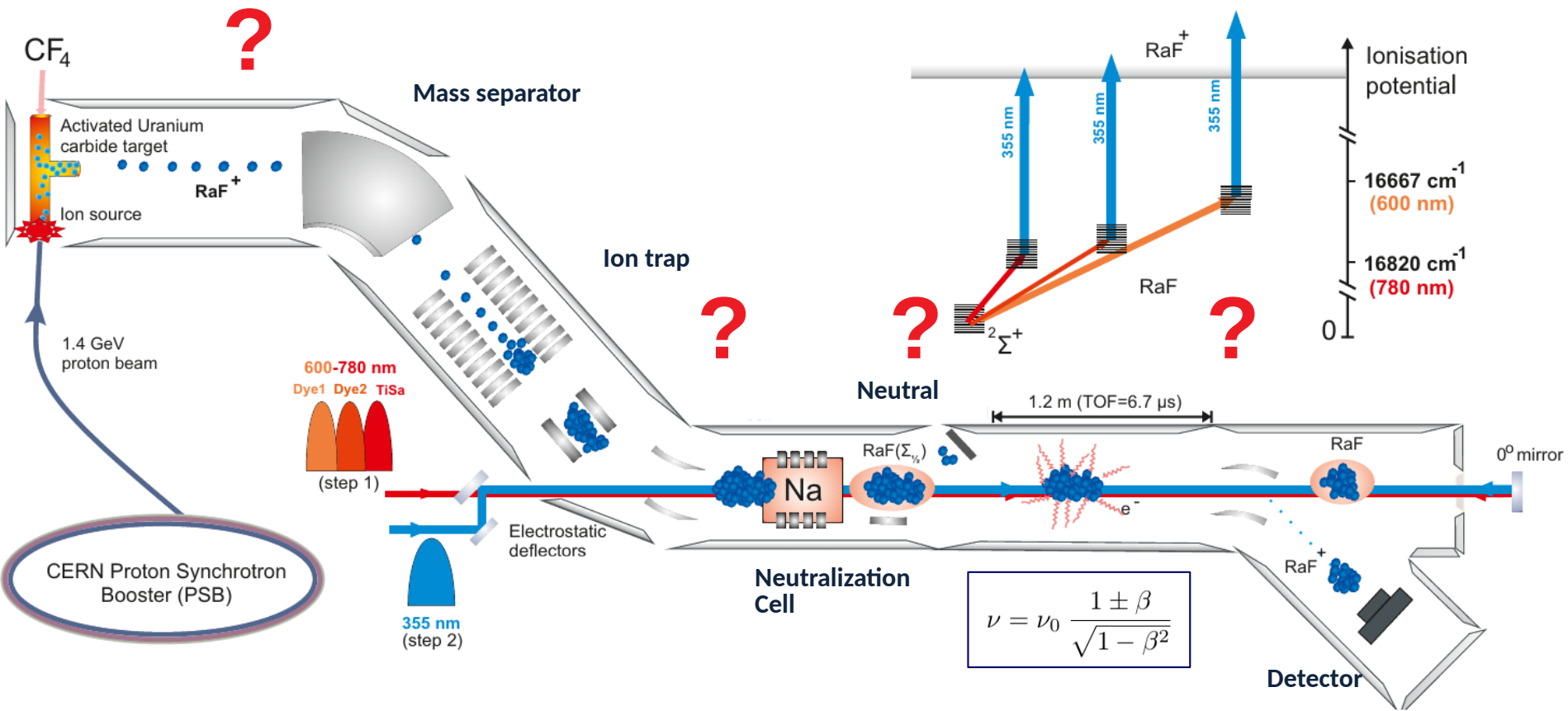
**Impossible with radioactive molecules (<  $10^6$  molecules/second)?**

# Results: Radium fluoride (RaF)



*"We shall send to the moon, 240,000 miles away from the control station in Houston, a giant rocket more than 300 feet tall, made of new metal alloys, some of which have not yet been invented, capable of standing heat and stresses several times more than have ever been experienced, on an untried mission, to an unknown celestial body, and then return it safely to Earth, reentering the atmosphere at speeds of over 25,000 miles per hour, causing heat about half that of the temperature of the sun, and do all this, and do it first before this decade is out—then we must be bold." J.F. Kennedy (1962).*

# Results: Radium fluoride (RaF)



ISOLDE mm CERN molecule

*"We shall send to the moon, 240,000 miles away from the control station in Houston, a giant rocket more than 300 feet tall, made of new metal alloys, some of which have not yet been invented, capable of standing heat and stresses several times more than have ever been experienced, on an untried mission, to an unknown gas-filled ion trap, and then return it safely to Earth, reentering the atmosphere at speeds of over 25,000 miles per hour, causing heat about half that of the temperature of the sun, and do all this, and do it first before this decade is out—then we must be bold."* J.F. Kennedy (1962).

fm

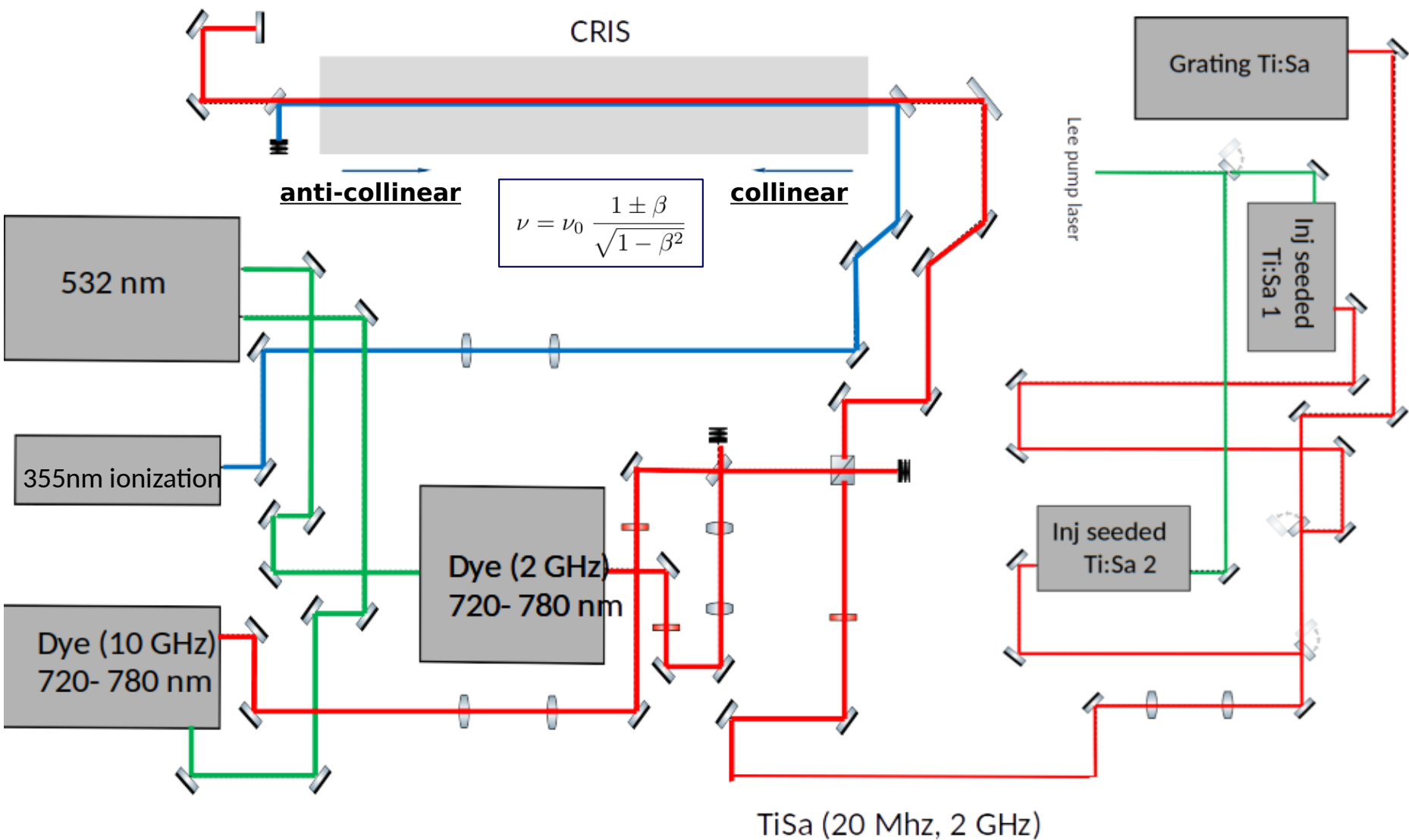
400,000

Long Shutdown 2

(neutral) neutralization cell

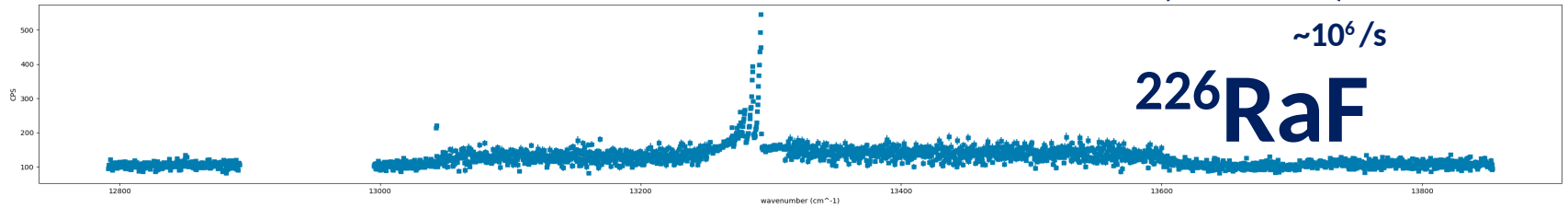
studied

# Results: Radium fluoride (RaF)



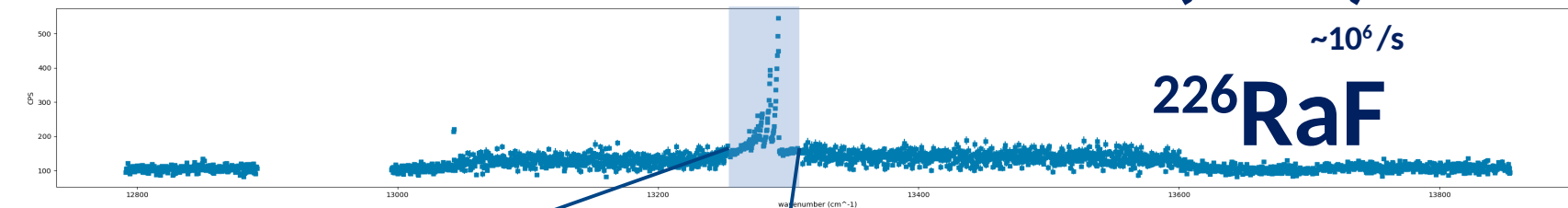
*About 20 laser systems  
(simplified scheme)*

# Results: Radium fluoride (RaF)

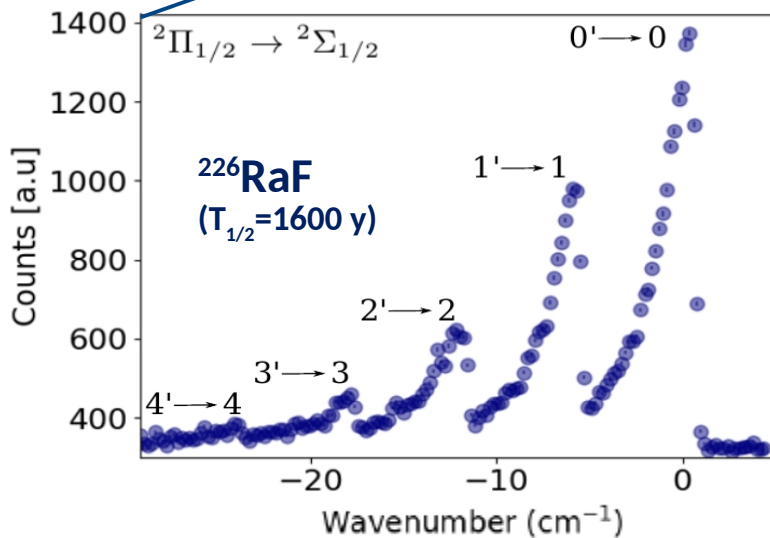


← ~1000 cm<sup>-1</sup> (30 THz) → only 4 hours!

# Results: Radium fluoride (RaF)

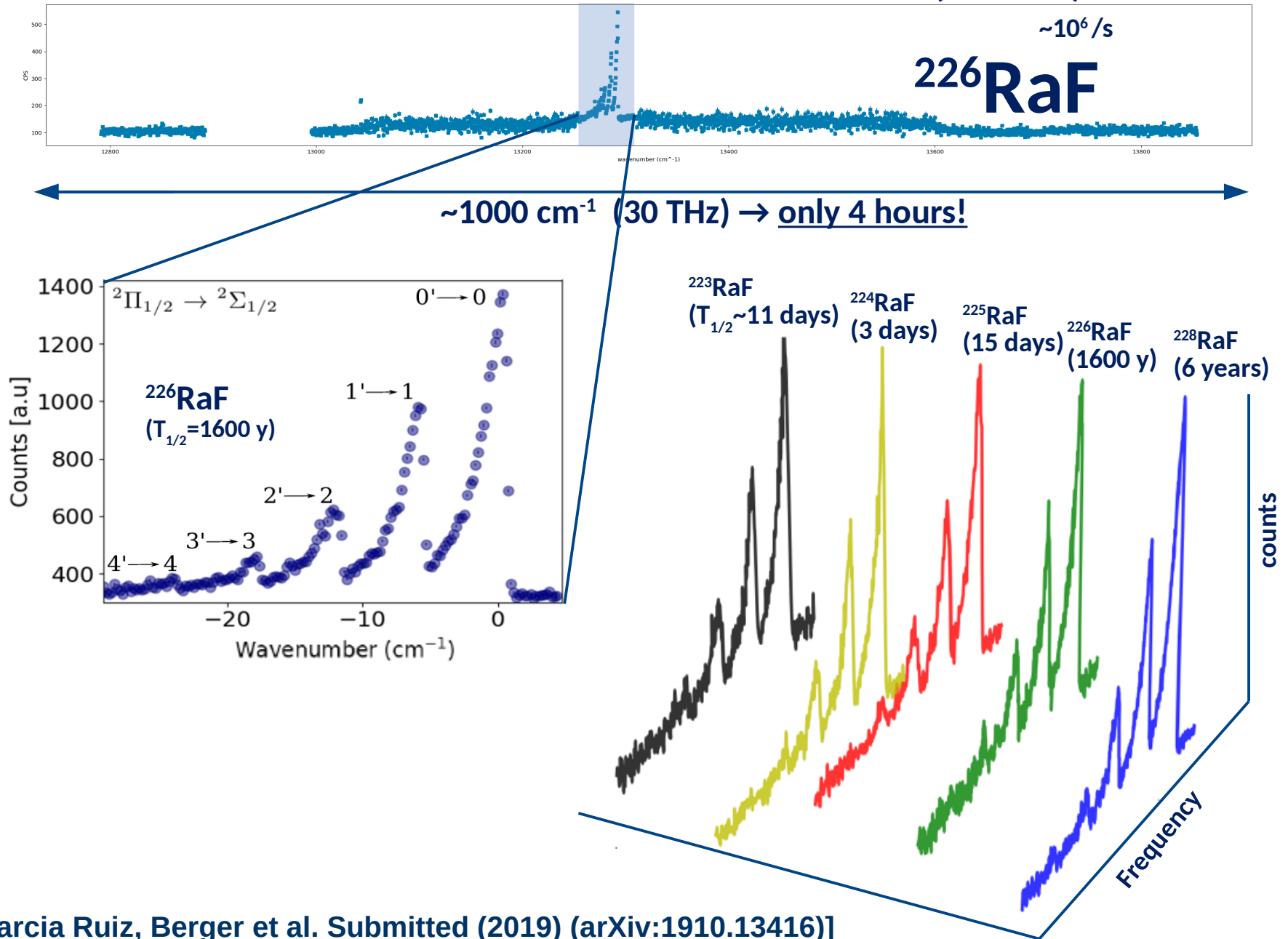


$\sim 1000 \text{ cm}^{-1}$  (30 THz)  $\rightarrow$  only 4 hours!





# Results: Radium fluoride (RaF)

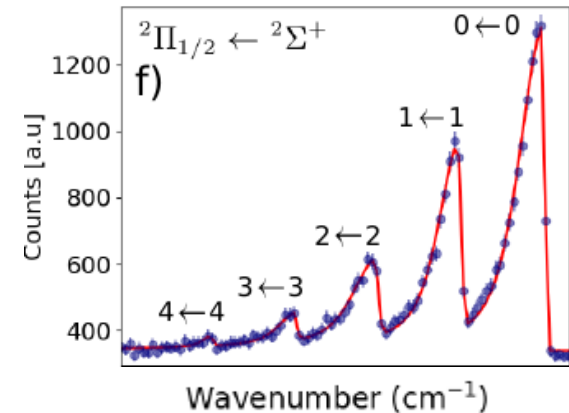
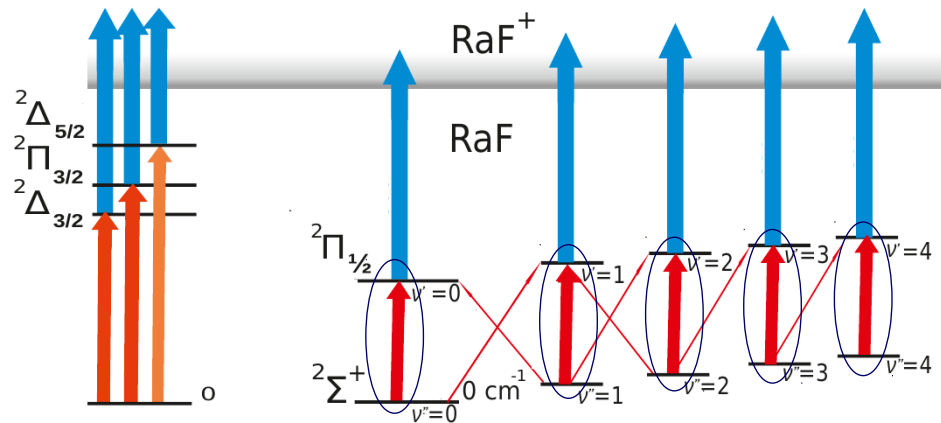


[Garcia Ruiz, Berger et al. Submitted (2019) (arXiv:1910.13416)]





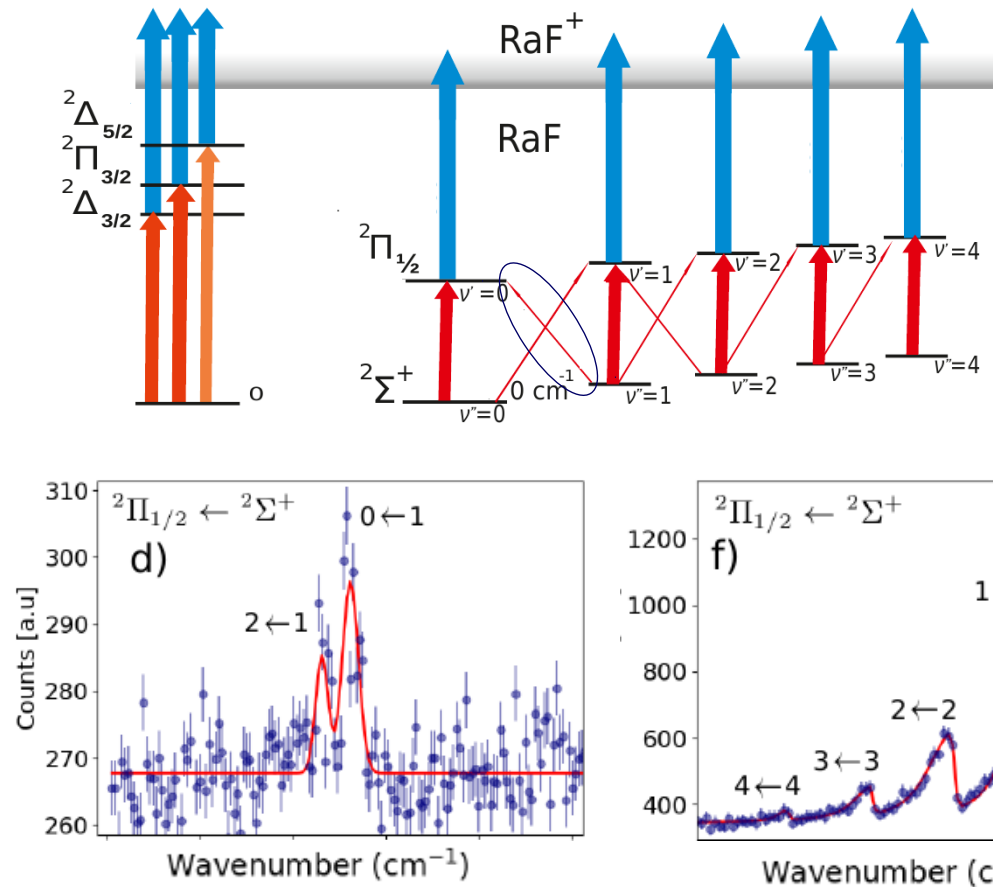
# Results: Radium fluoride (RaF)

- I. Low-lying structure ✓
- II. Feasibility of **laser cooling**?
  1. Dominant  $f_{00}$ ?
  2. Short-lived excited state ( $T_{1/2}$ )?
  3. Electronic states of lower energy (E)?



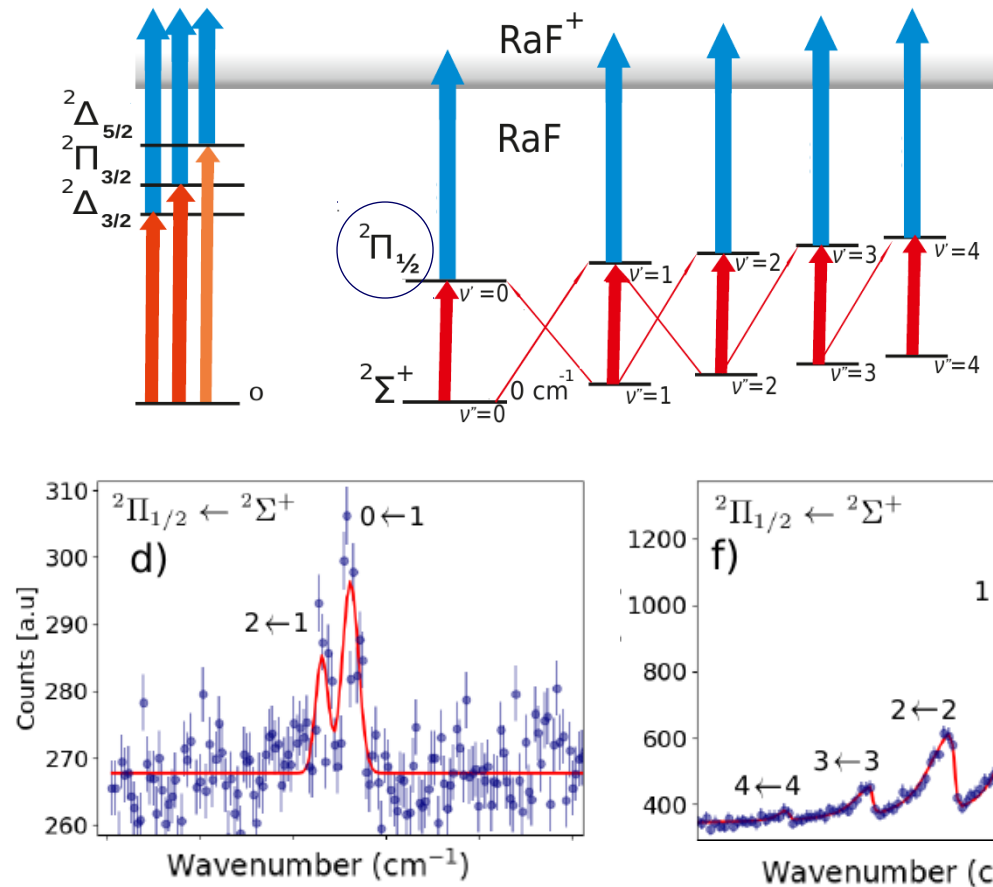
# Results: Radium fluoride (RaF)

- I. Low-lying structure 
- II. Feasibility of **laser cooling**?
  1. Dominant  $f_{00}$ ?  $\rightarrow f_{00}/f_{ij} > 0.97$  
  2. Short-lived excited state ( $T_{1/2}$ )?
  3. Electronic states of lower energy (E)?



# Results: Radium fluoride (RaF)

- I. Low-lying structure ✓
- II. Feasibility of laser cooling?
  1. Dominant  $f_{00}$ ?  $\rightarrow f_{00}/f_{ij} > 0.97$  ✓
  2. Short-lived excited state ( $T_{1/2}$ )?  $\rightarrow T_{1/2} < 50$  ns ✓
  3. Electronic states of lower energy (E)?



# Results: Radium fluoride (RaF)

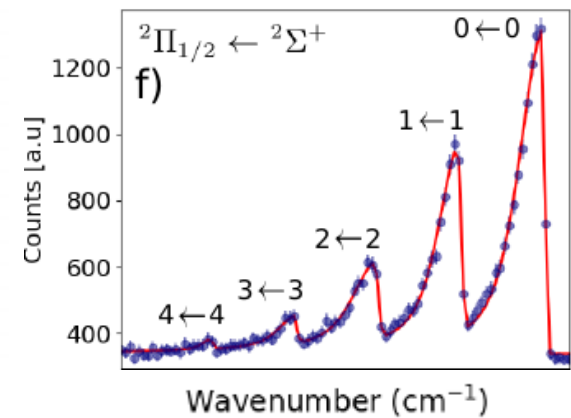
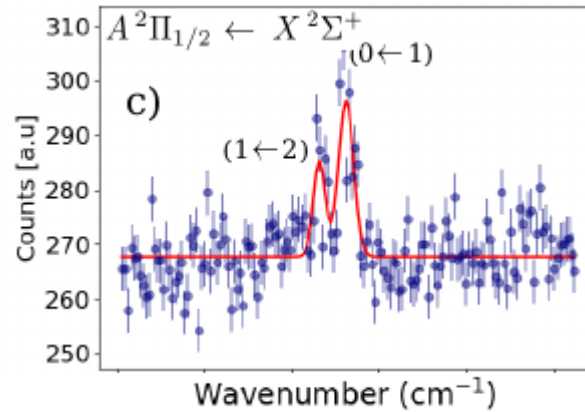
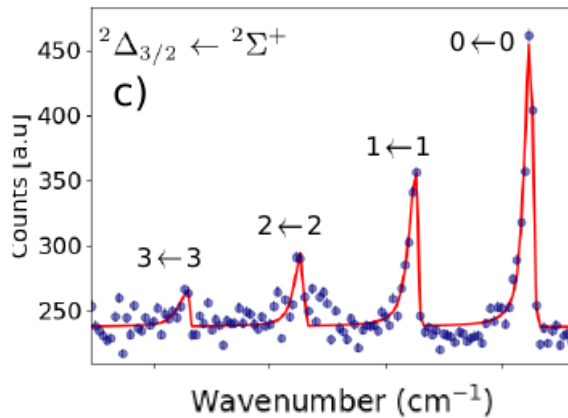
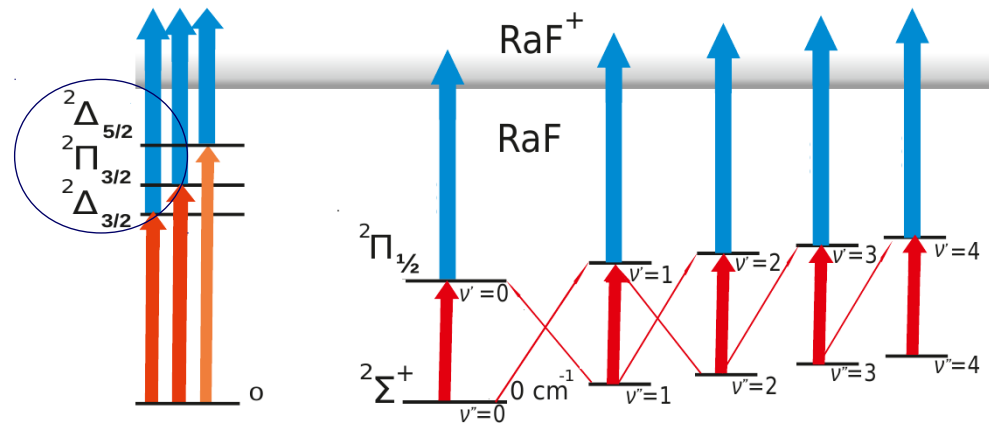
I. Low-lying structure ✓

II. Feasibility of laser cooling?

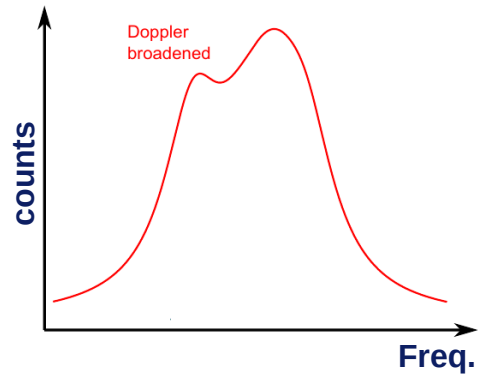
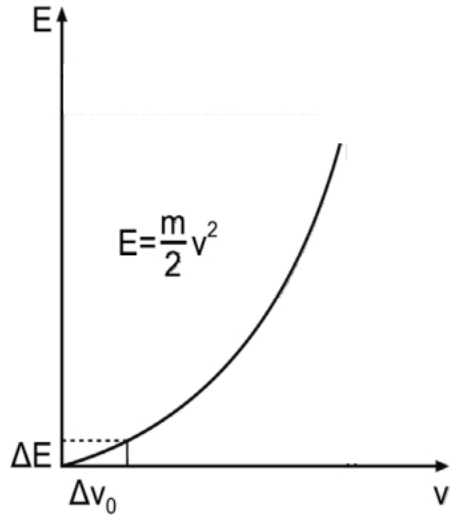
1. Dominant  $f_{00}$ ?  $\rightarrow f_{00}/f_{ij} > 0.97$  ✓

2. Short-lived excited state ( $T_{1/2}$ )?  $\rightarrow T_{1/2} < 50$  ns ✓

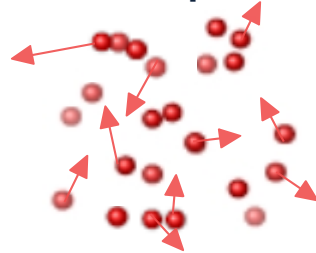
3. Electronic states of lower energy (E)?  $\rightarrow 2000$   $\text{cm}^{-1}$  above ✓



# Precision Laser Spectroscopy

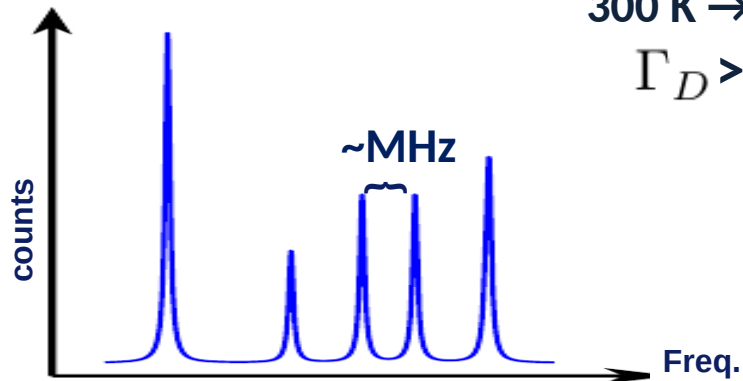


Room temperature

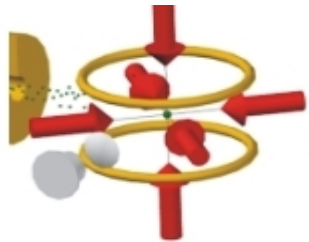
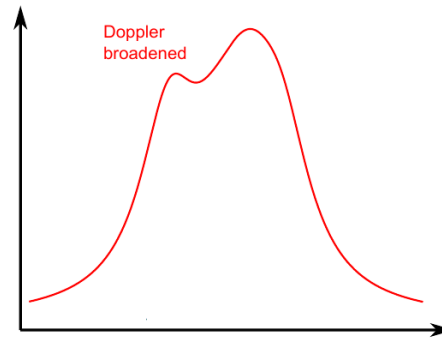
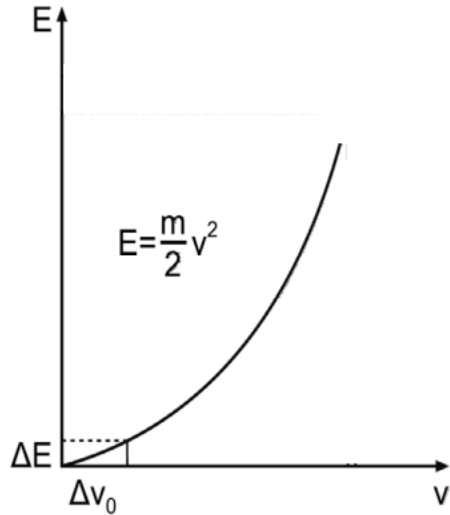


300 K  $\rightarrow$  25 meV

$\Gamma_D > \text{GHz}$



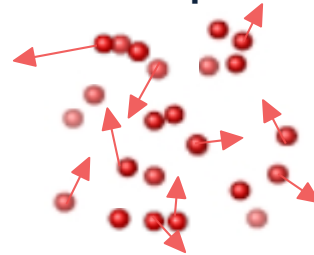
# Precision Laser Spectroscopy



$T < \mu\text{K}$   
 $\Gamma_D < \text{MHz}$



Room temperature



$300 \text{ K} \rightarrow 25 \text{ meV}$   
 $\Gamma_D > \text{GHz}$

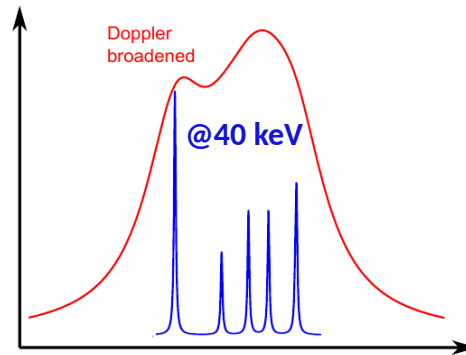
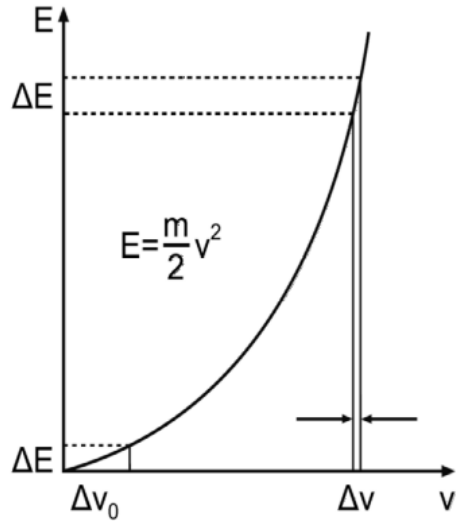
✓ High resolution ( $< \text{MHz}$ )

➤ High efficiency ( $< 100$  ions/s) ?

➤ High selectivity ( $> 1/10^6$ ) ?

➤ Short time scales ( $< 1 \text{ s}$ ) ?

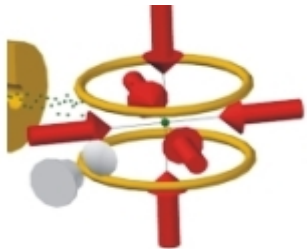
# Precision Laser Spectroscopy



Energy spread

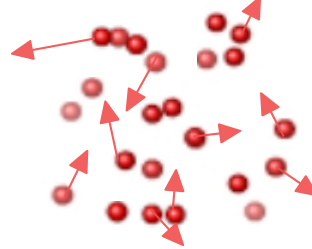
$$\Gamma_D = \nu_0 \frac{\delta E}{\sqrt{2eUmc^2}}$$

Ion beam energy



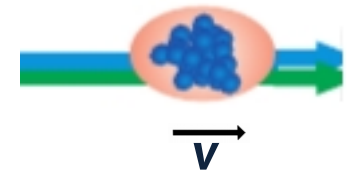
$T < \mu\text{K}$   
 $\Gamma_D < \text{MHz}$

Room temperature



300 K  $\rightarrow$  25 meV  
 $\Gamma_D > \text{GHz}$

Fast beam

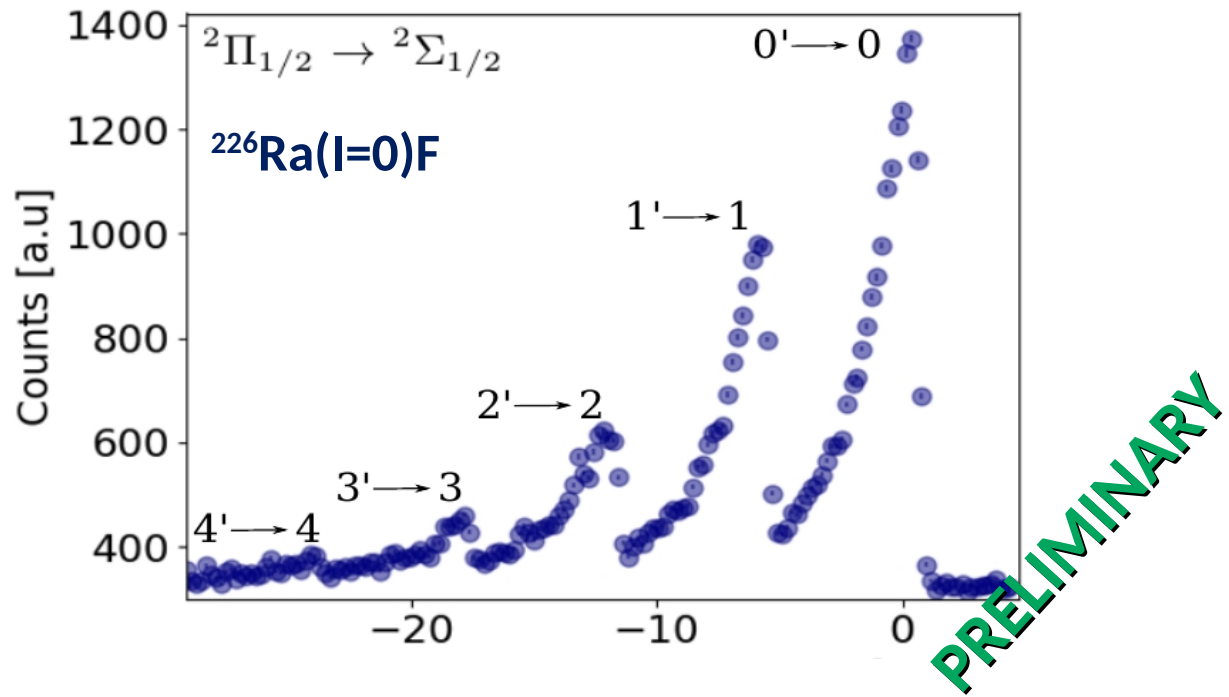


@ 40 keV  
 $\Gamma_D \sim \text{MHz}$

- ✓ High resolution (< MHz)
- High efficiency (< 100 ions/s) ?
- High selectivity (> 1/10<sup>6</sup>) ?
- Short time scales (< 1 s) ?

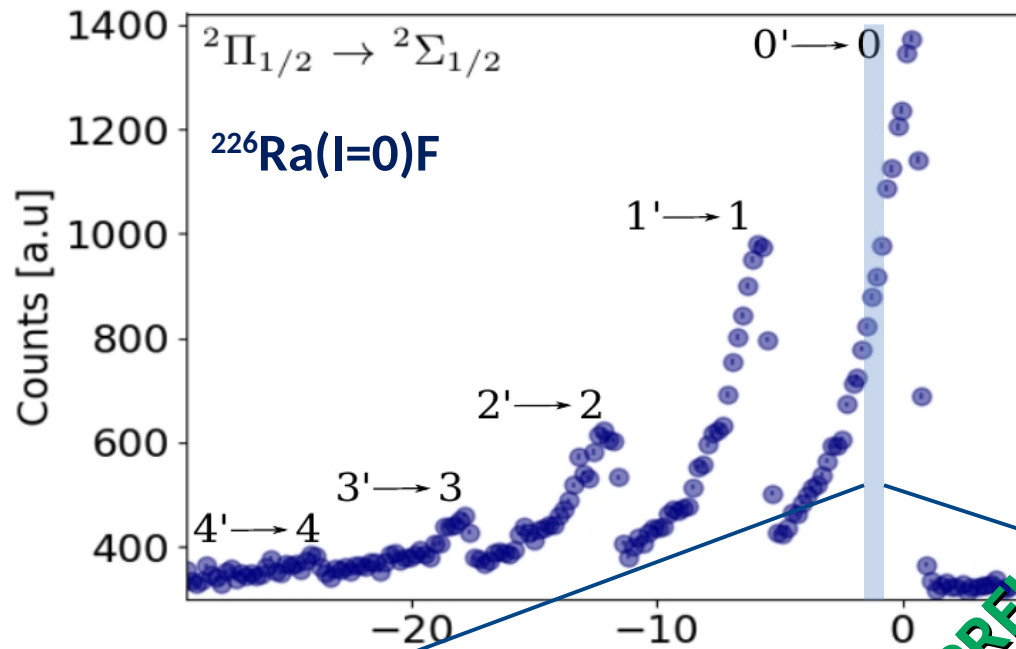
- ✓ High resolution (~MHz)
- ✓ High efficiency (< 100 ions/s)
- ✓ High selectivity (> 1/10<sup>6</sup>)
- ✓ Short time scales (< 1 s)

# RaF: Hyperfine Structure





# RaF: Hyperfine Structure

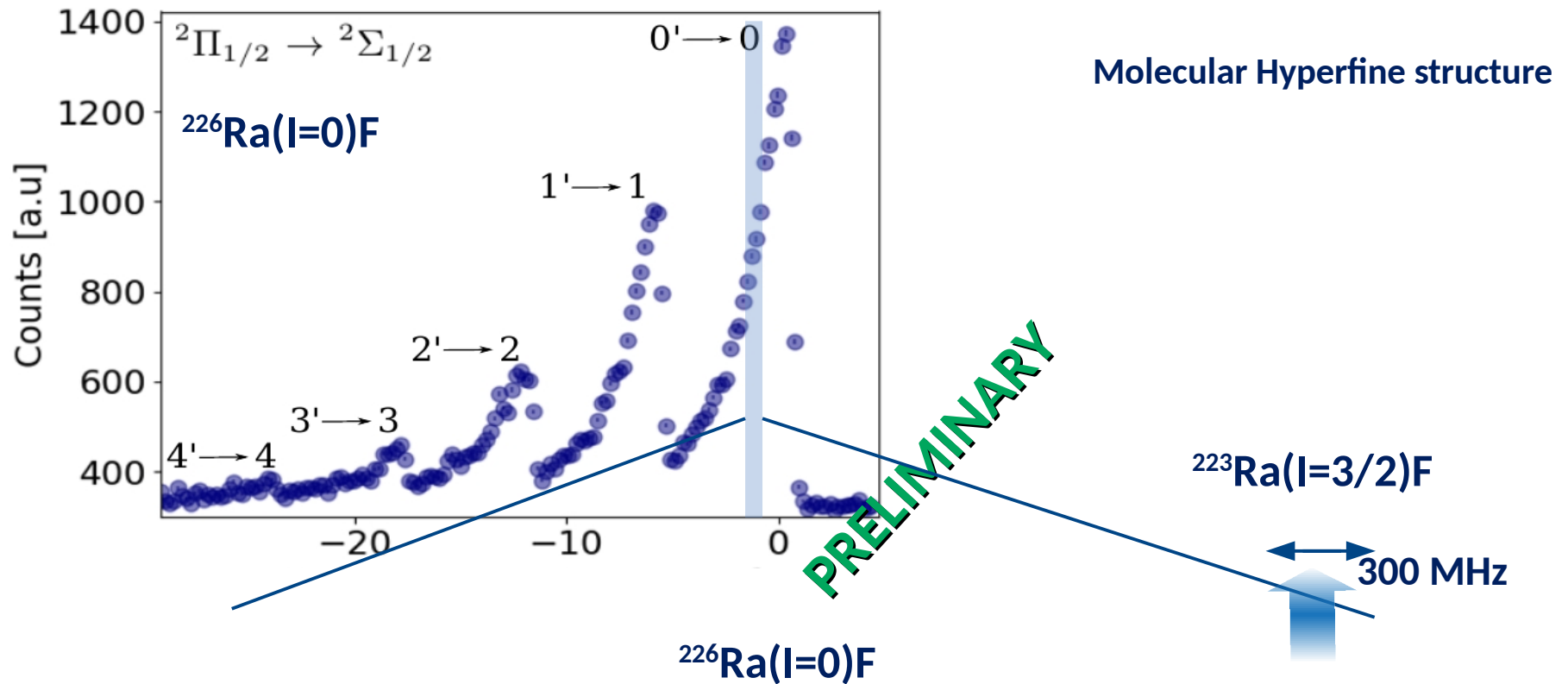


${}^{226}\text{Ra}(I=0)\text{F}$

Wavenumber ( $\text{cm}^{-1}$ )

~ 15 GHz

# RaF: Hyperfine Structure



~ 15 GHz

From THz to MHz

# Opportunities with radioactive molecules

- Axion dark matter produces oscillating  $S_{\text{schiff}}$ , MQM

Amplified in molecules with respect to atoms:

$$(M_{\text{mol}}/m_e)^2 \sim 10^6 - 10^8$$

→ AcF, RaO, PaN

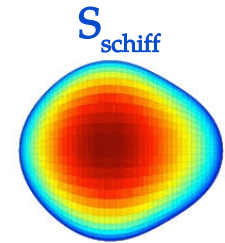
[Flambaum & Feldmeier . Phys. Rev. C 101, 015502 (2020)]

- Time reversal violating MQM

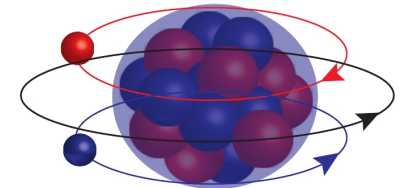
→  $^{229}\text{ThO}$  and  $^{229}\text{ThF}^+$  [Lackenby & Flambaum. Phys. Rev. D 98, 115019 (2018)]

[ Skripnikov, et al. Phys. Rev. Lett. 113, 263006 (2014)]

- Quantum chemistry, astrophysics, ....



MQM



**Astronomical detection of radioactive molecule**

$^{26}\text{AlF}$  in the remnant of an ancient explosion

[Kaminski et al. Nature Astronomy 2, 778 (2018)]

*“Spectroscopic laboratory studies of rare radioactive materials such as  $^{26}\text{AlF}$  would be very challenging”*

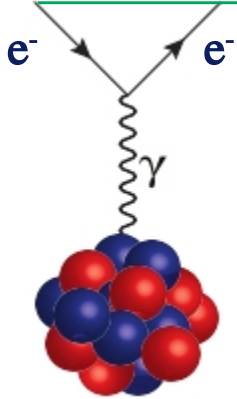
$$(T_{1/2} = 7.17 \cdot 10^5 \text{ years})$$

# Contents

- Precision studies in atoms and molecules
- Nuclear structure / nuclear matter / new forces & particles
- Parity violation
- Parity and time-reversal violation
- First results: RaF
- **Summary**

# Summary

## Nuclear structure

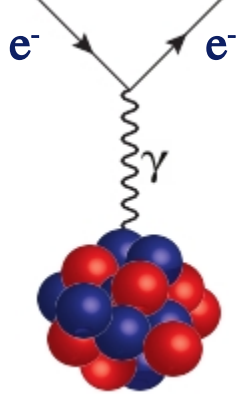


$$\mu \langle r^2 \rangle I Q$$

- Nuclear structure
- Nuclear matter
- New particle/forces

# Summary

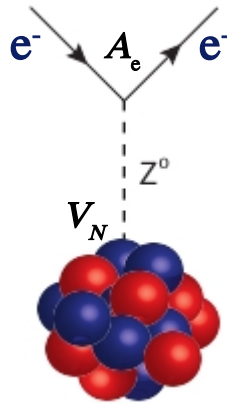
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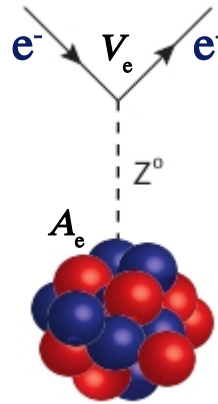
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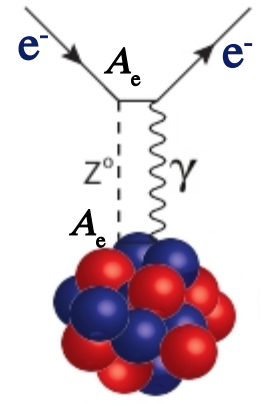
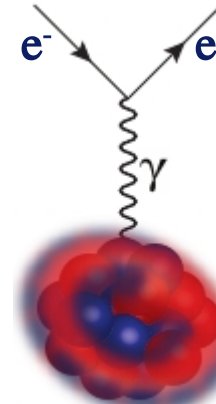
## P- violation



$$\sim Z^3 \times 10^N$$



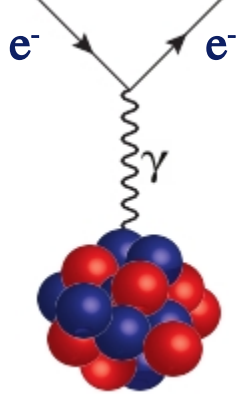
$$\sim Z^2 A^{2/3} R(Z) \times 10^5$$



Atoms  
Molecules

# Summary

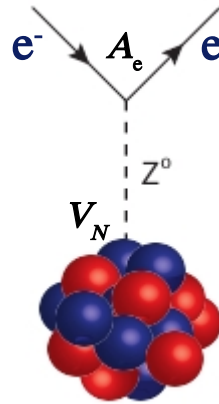
## Nuclear structure



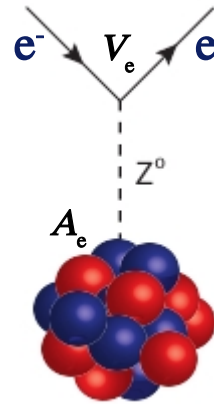
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- Nuclear structure
- Nuclear matter
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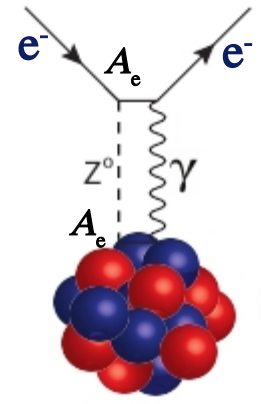
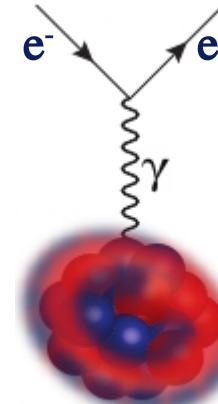
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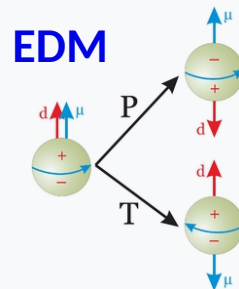


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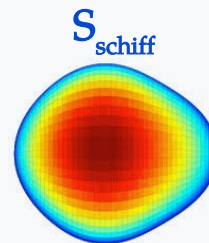


Atoms  
Molecules

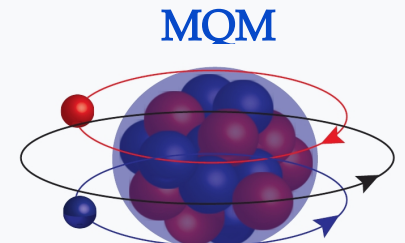
## P,T- violation



$$\sim Z^2 R(Z) > 10^3$$



$$S \sim Q_2 Q_3 Z A^{2/3} / (E_+ - E_-) (> 10^5 \text{ Octupole})$$



Atoms  
Molecules

# Summary

- Radioactive molecules → New window to study the atomic nucleus

## Molecules: Electroweak structure

- **Anapole moment:  $AM$**
- **Magnetic Quadrupole Moment: MQM**
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- **eEDM, nEDM, ...**





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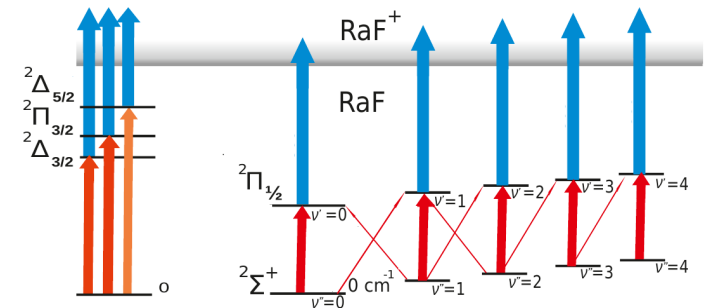
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- ✓ RaF → eEDM, Schiff moments, Anapole moments, MQM
- ✓  $^{223}\text{RaF}$ ,  $^{224}\text{RaF}$ ,  $^{225}\text{RaF}$ ,  $^{226}\text{RaF}$ ,  $^{228}\text{RaF}$
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- ✓ Precision spectroscopy  $^{223}\text{Ra}(I=3/2)\text{F}$ , lifetime, IP, ...



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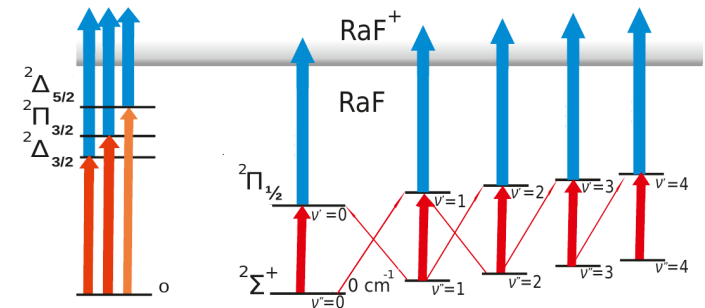
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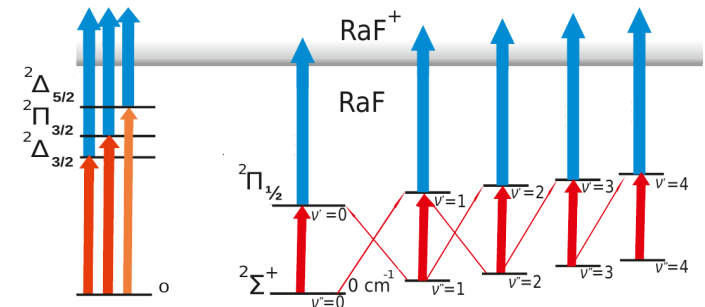
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... this is just the beginning!