

P,T-violation through laser spectroscopy of molecules

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From a classical point of view, there can be no average electric field at the nucleus unless some non-electric force is available to keep the nucleus from accelerating under the influence of this electric field.

Schiff, Physical Review 132 (1963)

1.1 What are we looking for: eEDM

P,T-violation exists in Standard Model

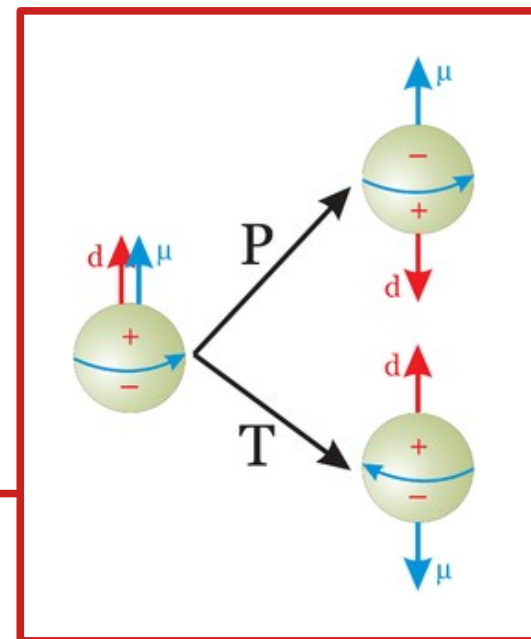
CP-odd weak force: complex phase in CKM

CP-odd strong force: θ vacuum

$eEDM(SM) \sim 10^{-38} e \cdot cm$ does not explain M-AM problem

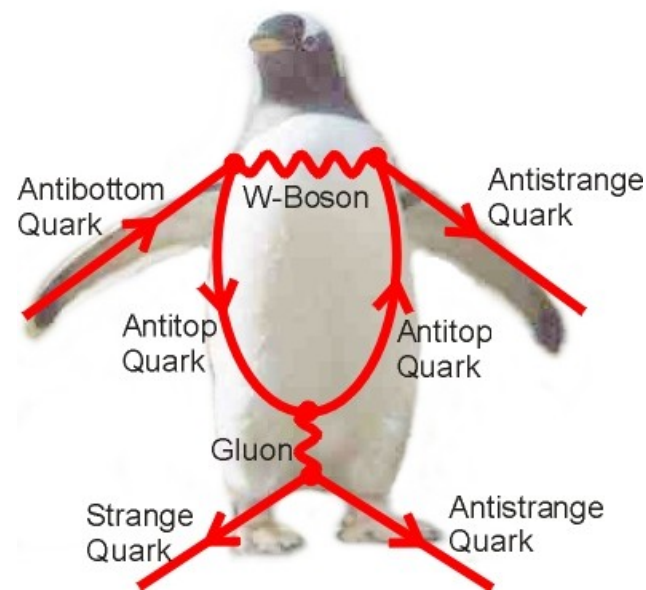
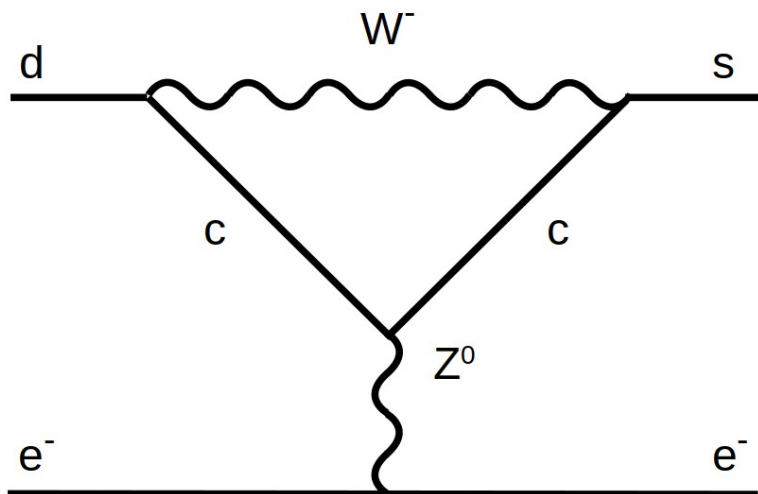
$eEDM(HfF^+) < 4 \times 10^{-30} e \cdot cm$ equivalent to $m > 10 \text{ TeV}/c^2$

Roussy et al, arXiv:2212.11841



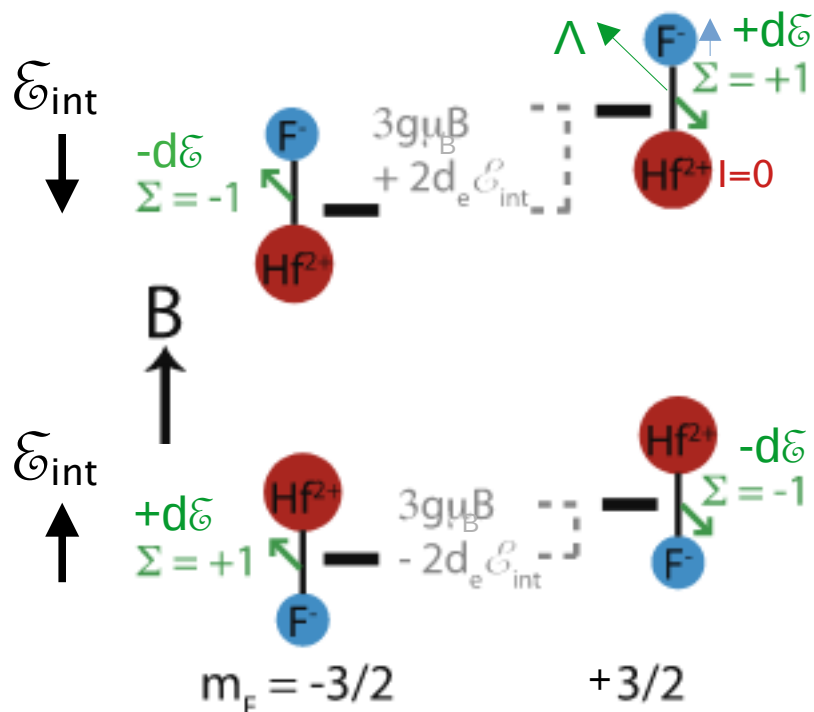
in β decay: $\langle e|W|\nu \rangle$

in laser spectroscopy: neutral weak current $\langle e|Z^0|e \rangle$



1.1 What are we looking for: eEDM

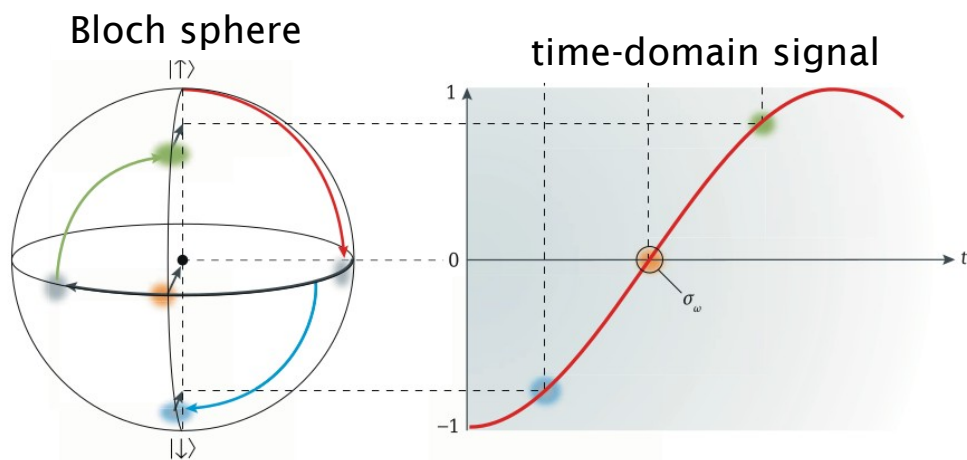
$$I=1/2, J=1, F=3/2$$



HfF⁺ ³Δ₁ molecular state: Λ=2, Σ=1, Ω=2-1=1
 projected on intranuclear axis F⁻→Hf²⁺
 whereas I+J=F=3/2 relative to magnetic field
 eEDM along Σ

ΔE(-3/2,+3/2) in applied magnetic field
 same when swapping the molecule...
 unless EDM in internal electric field!

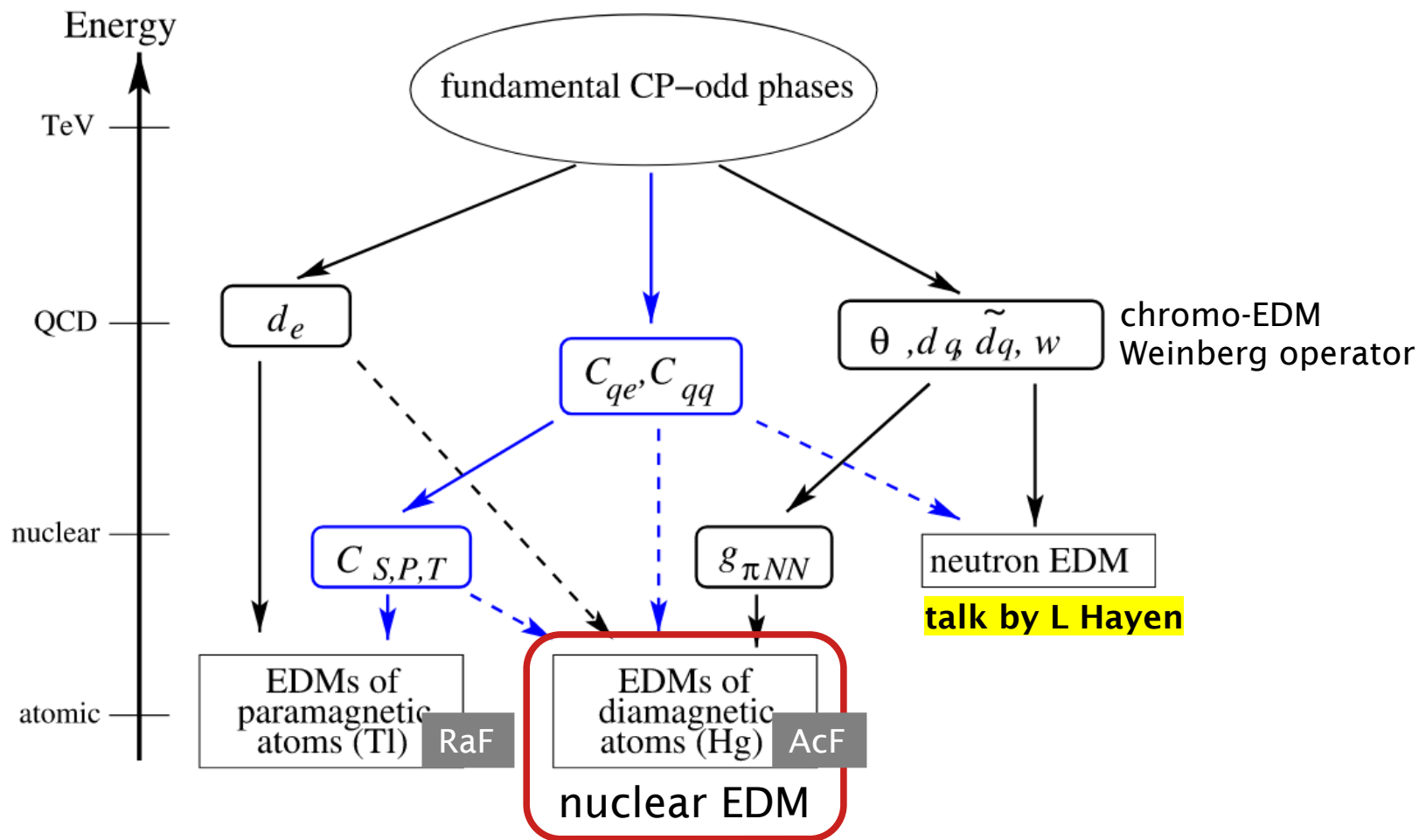
Cossel et al., Chemical Physics Letters 546, 1 (2012)
 Cairncross and Ye, Nature Reviews Physics 1, 510 (2019)



Ramsey spectroscopy
 $P = \cos^2((\omega - \omega_0)t/2)$

for unpaired electrons:
 electron paramagnetic resonance

1.2 What are we looking for: Schiff moment



Pospelov and Ritz, Annals of Physics 318 (2005)

**Schiff theorem: electron screening cancels nuclear EDM
but can be overcome by spin-magnetic & finite-size effects**

Schiff, Physical Review 132 (1963)

1.2 What are we looking for: Schiff moment

outlying p-electron falls back into s-orbital by screened EDM ie Schiff moment

P-odd EDM scatters core-penetrating s-electron into p-orbital

$$d_{\text{atom}} = 2 \sum_m \frac{\langle ns | -e\varphi(\mathbf{R}) | mp \rangle \langle mp | -e\mathbf{R} | ns \rangle}{E_{ns} - E_{mp}}$$

the closer the energy, the larger the contribution

$$\langle s | -e\varphi^{(1)} | p \rangle = 4\pi e \mathbf{S} \cdot (\nabla \psi_s^\dagger \psi_p)_{R \rightarrow 0}$$

$$\mathbf{S} = \frac{e}{10} \left[\langle r^2 \mathbf{r} \rangle - \frac{5}{3Z} \langle r^2 \rangle \langle \mathbf{r} \rangle \right]$$

P,T-odd deformation

EDM screening

Flambaum and Ginges, PRA 65 (2002)

P,T-odd octupole deformation **heavy nuclei**

$$S \propto \eta e \frac{\beta_2 \beta_3^2 Z A^{2/3} r_0^3}{E_+ - E_-}$$

coupling strength

parity doublets

Chupp, Fierlinger, Ramsey-Musolf & Singh, RMP 91 (2019)

1.2 What are we looking for: Schiff moment

in atoms

- relativistic s-electrons dive deep into heavy **Z** nucleus...
- ... and sense octupole deformation β_3 from within
 $\beta_3(^{229}\text{Th}) = 0.115$

Minkov and Palffy, Phys. Rev. Lett. 118 (2017)

$S(^{229}\text{Th}) \sim 600 \times S(\text{natTl})$ ie **2-3 orders of magnitude**

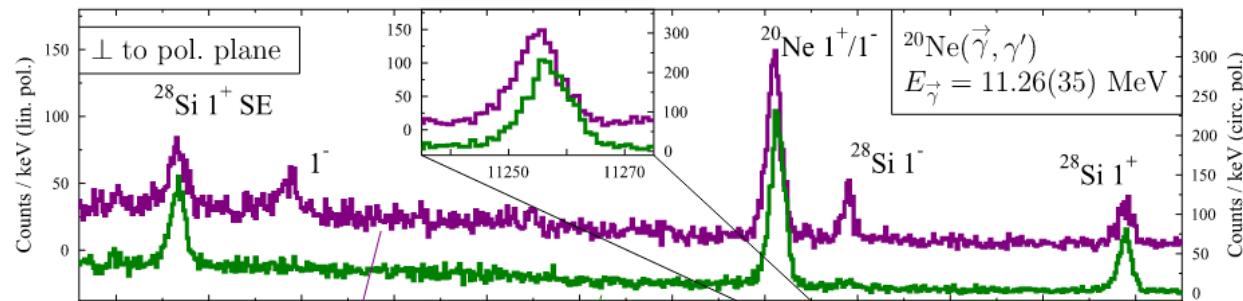
Flambaum, Phys. Rev. C 99 (2019)

in molecules

- $\Delta E(-m_F, m_F)$ amplified by **internal fields** as strong as 104 GV/cm in HgF
 Prasanna, Vutha, Abe, and Das, Phys. Rev. Lett. 114 (2015)
- energy shift in parity doublet $\Delta E(F^+, F^-) = \frac{D\mathcal{E}}{\sqrt{(\Delta/2)^2 + D^2\mathcal{E}^2}} \delta_{P,T}$

6-9 orders of magnitude smaller than in atoms

Safronova et al., Rev. Mod. Phys. 90 (2018)



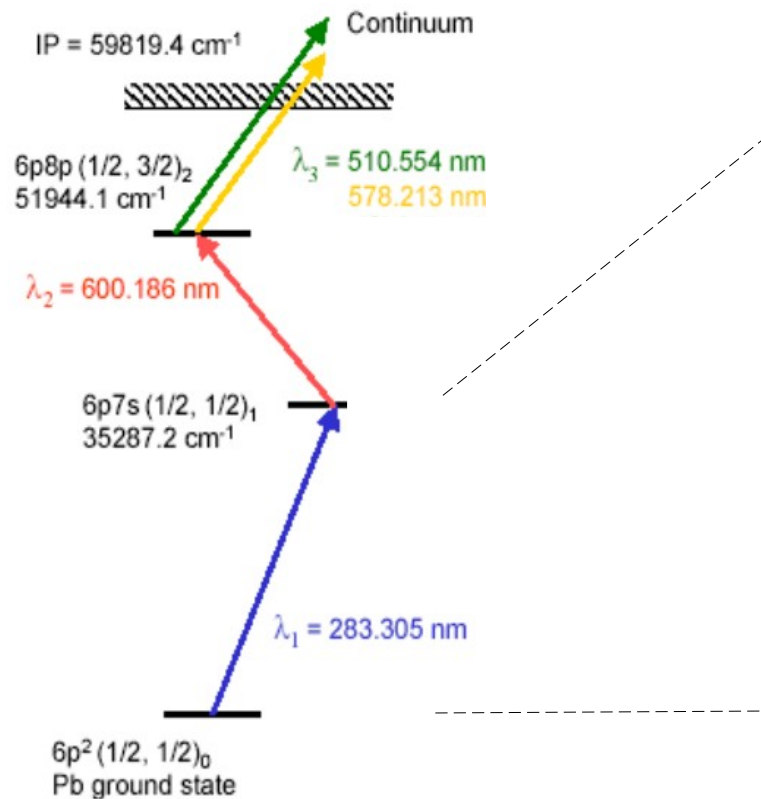
Beller et al, Physics Letters B 741 (2015) 128

in ^{20}Ne , $\Delta E = 3.2$ keV
 in atoms, \sim eV
 in ThO, \sim neV !

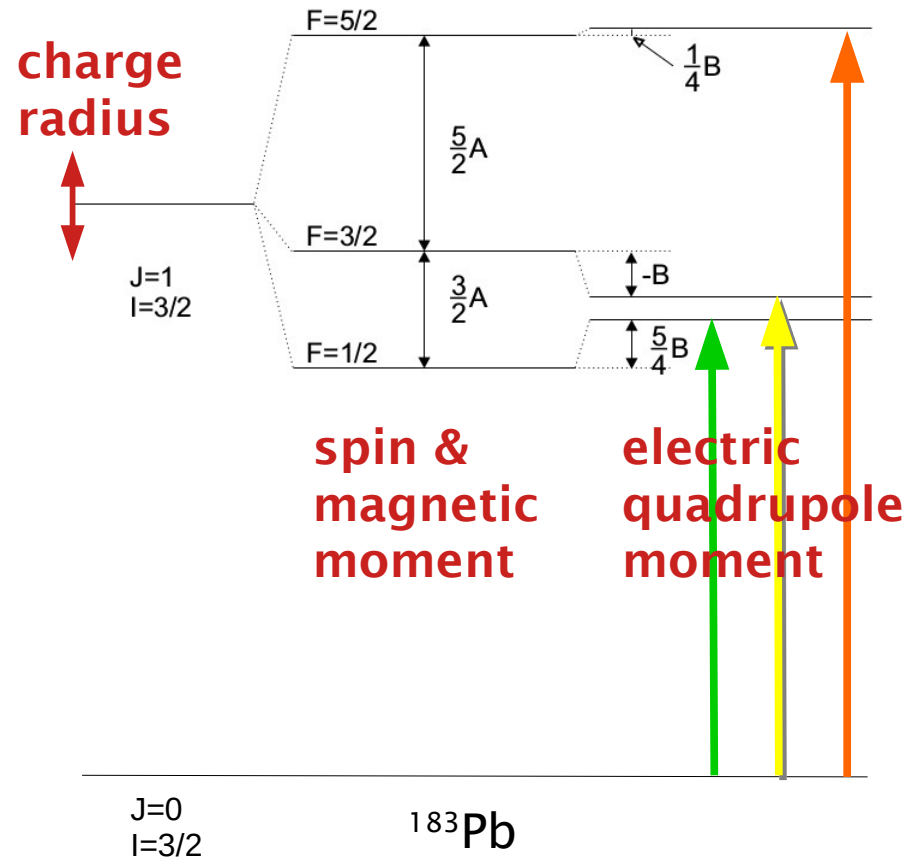
in radioactive molecules, combine heavy nucleus with molecular enhancement:
 laser spectroscopy of RaF and AcF at Isolde

1.3 Laser spectroscopy

laser ion source:
broadband lasers (~10 GHz)



laser spectroscopy:
narrowband lasers (<100 MHz)



- resonant laser ionisation selects one element within many
- probing hyperfine interaction gives access to nuclear structure
- intrasource > intrajet > collinear spectroscopy for increased resolution
- Collaps = fluorescence vs Cris = resonant ionisation

2.1 at Isolde

RaF: 10^3 available states...

Theory predicts $13300(1000) \text{ cm}^{-1}$ ($\lambda\nu=c$, 1.65 eV, 752 nm)

R Berger, priv. comm.

Scanning $1000 \text{ cm}^{-1} = 30 \text{ THz}$ at 10 MHz/min

... 2080 days!

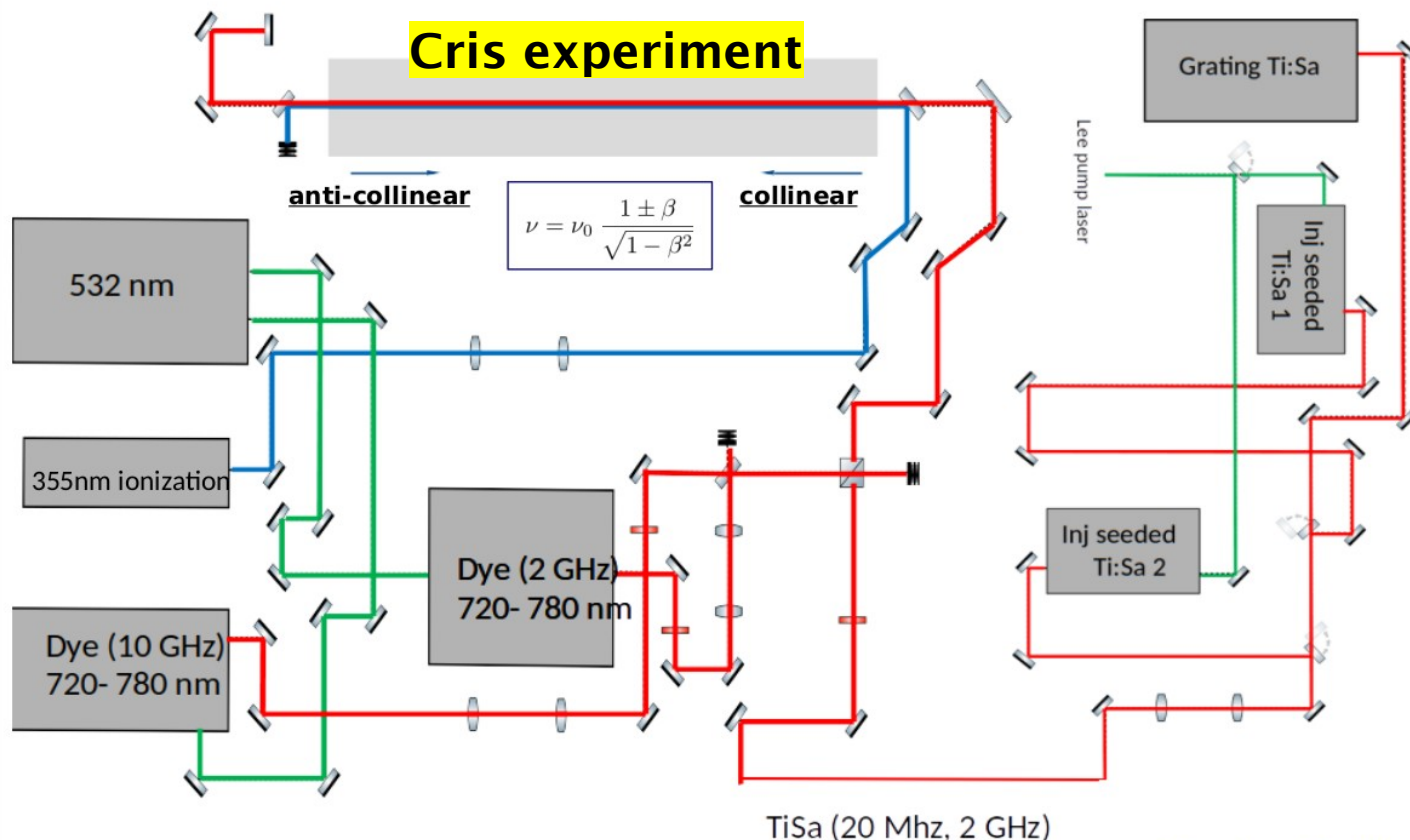
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Simplified laser setup



- many lasers
- cooling in RFQ-B

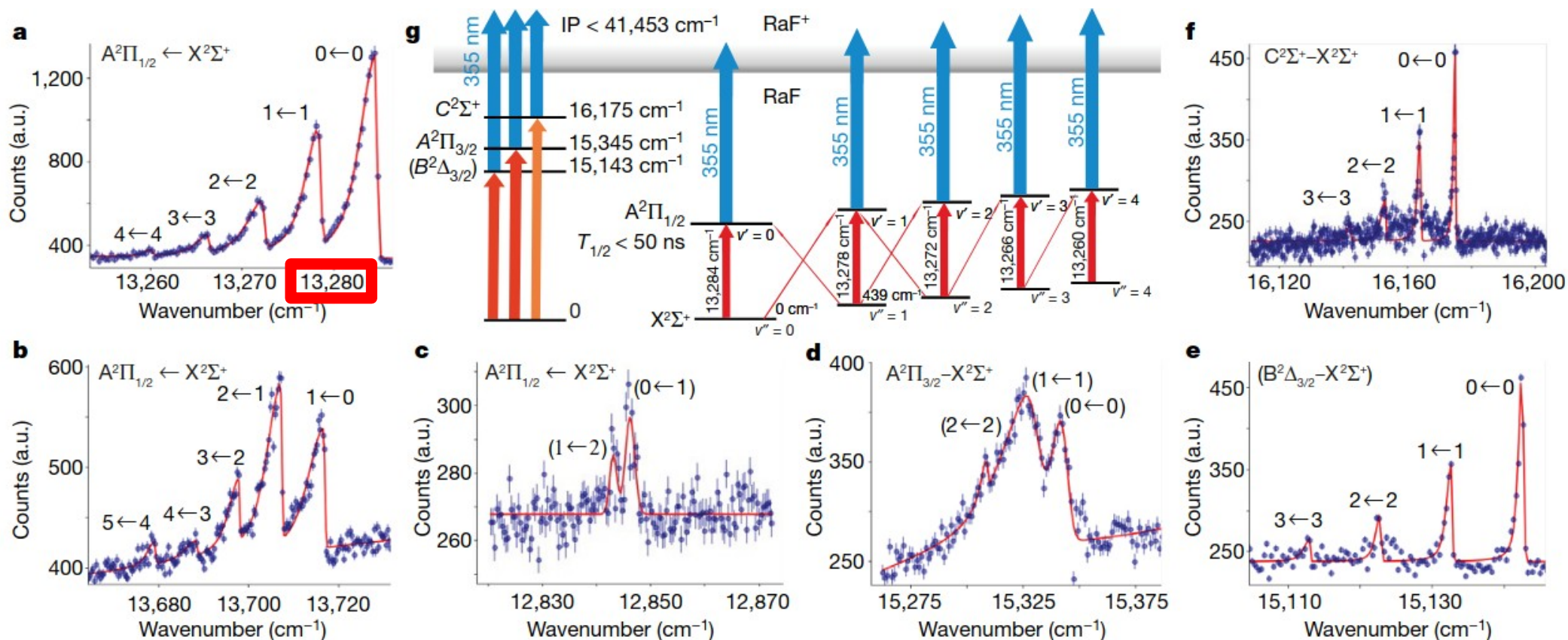
... signal already within 4 hours!

Figure from Fredrik Parnefjord Gustafsson

2.1 at Isolde

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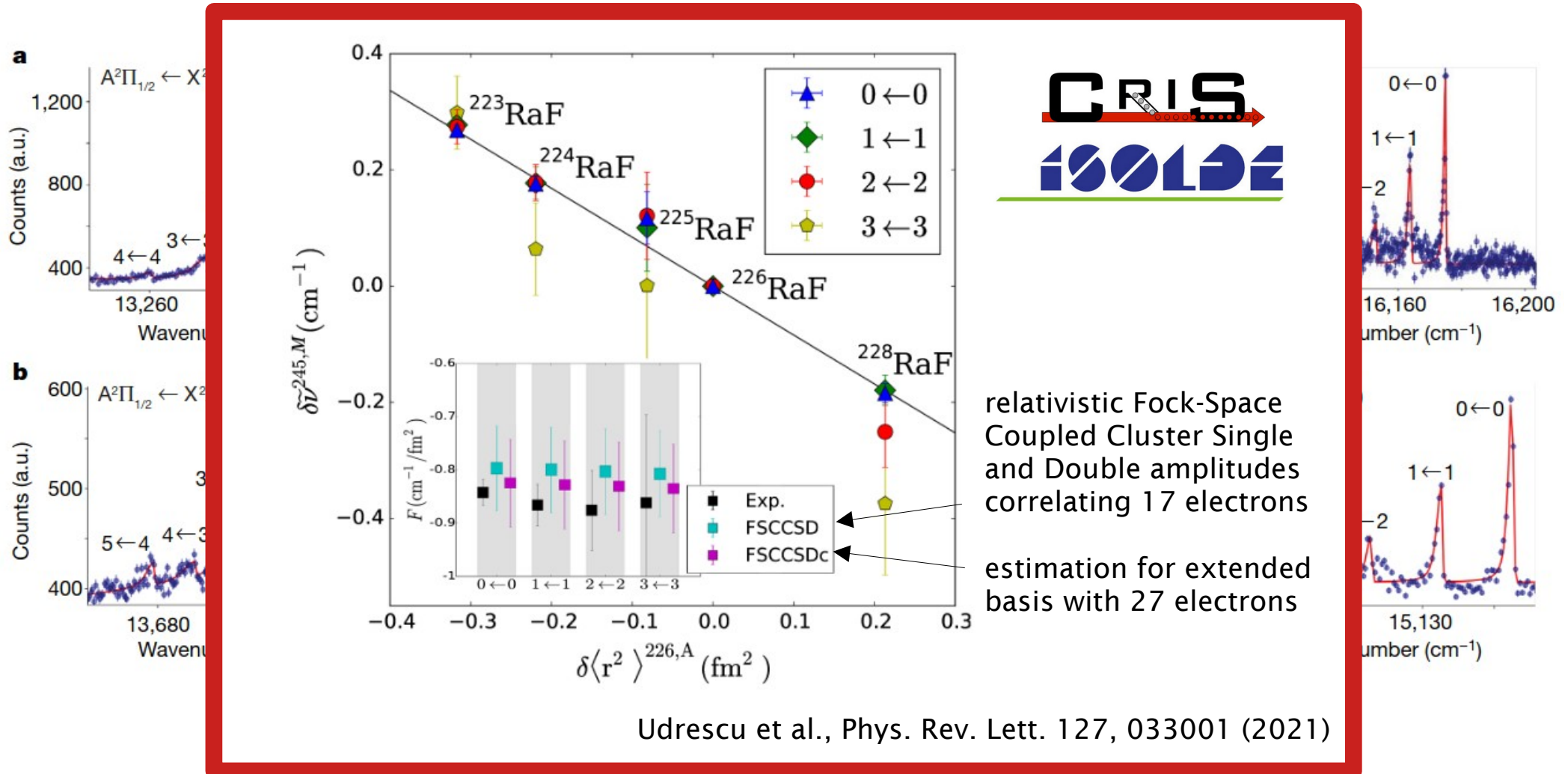


- low-lying structure & Ionisation potential
- laser cooling scheme
- ^{223}RaF , ^{224}RaF , ^{225}RaF , ^{226}RaF , ^{228}RaF

2.1 at Isolde

RaF: 10^3 available states...

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• laser cooling scheme

• ^{223}RaF , ^{224}RaF , ^{225}RaF , ^{226}RaF , ^{228}RaF

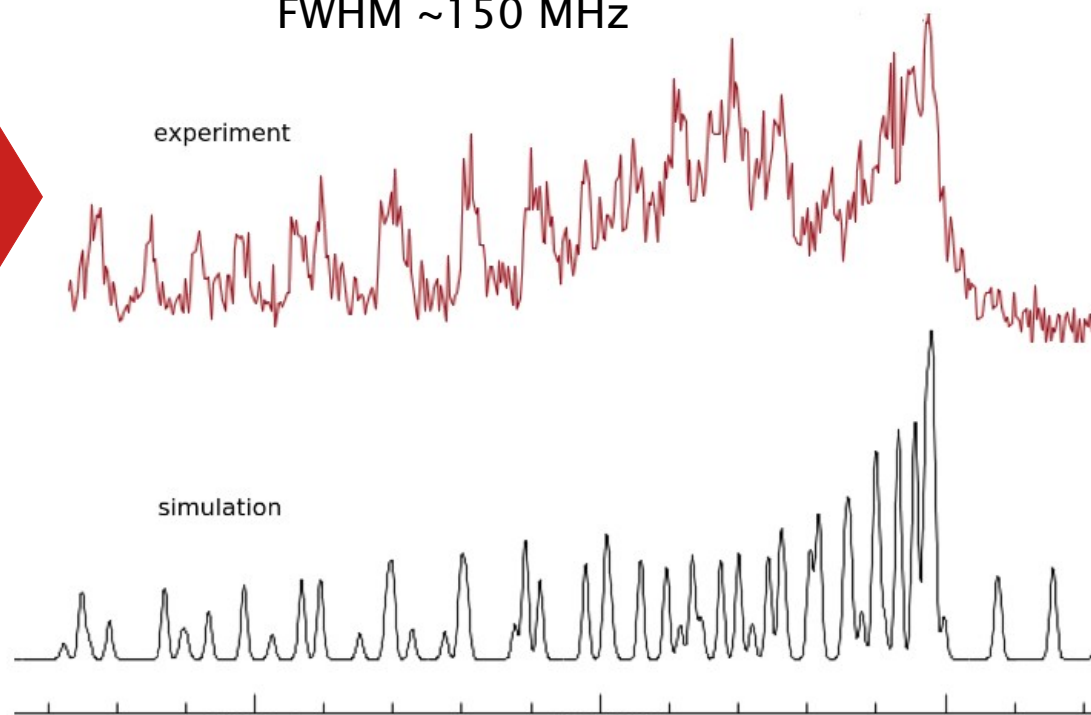
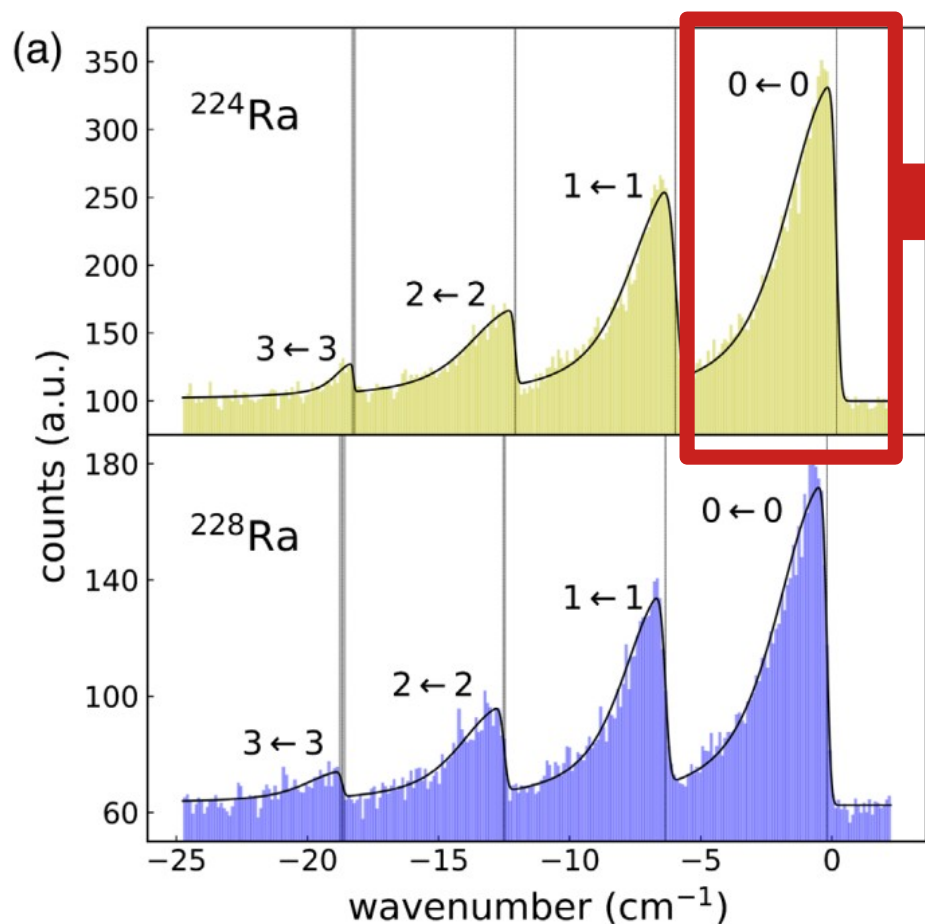
Garcia Ruiz et al., Nature 581, 396 (2020)

RaF low resolution

$A^2\Pi \leftarrow X^2\Sigma$ isovibrational bands
FWHM ~ 10 GHz

RaF high resolution

rotational spectrum
within one band
FWHM ~ 150 MHz



**rotational & hyperfine parameters needed
for accessing fundamental physics**

extract isotope shift

2.1 at Isolde

why?

search for symmetry breaking in relativistic heavy nuclei
molecular enhancement by several orders of magnitude

how?

probe tiny anomalies in hyperfine structure
need large number of particles in small number of states
cooling to room temperature in RFQB yields $v=v'$

2.2 at S3

set-up for laser spectroscopy in gas jet... can be used rightaway!

supersonic expansion cools to 20 K

implement gas mixture for molecule formation

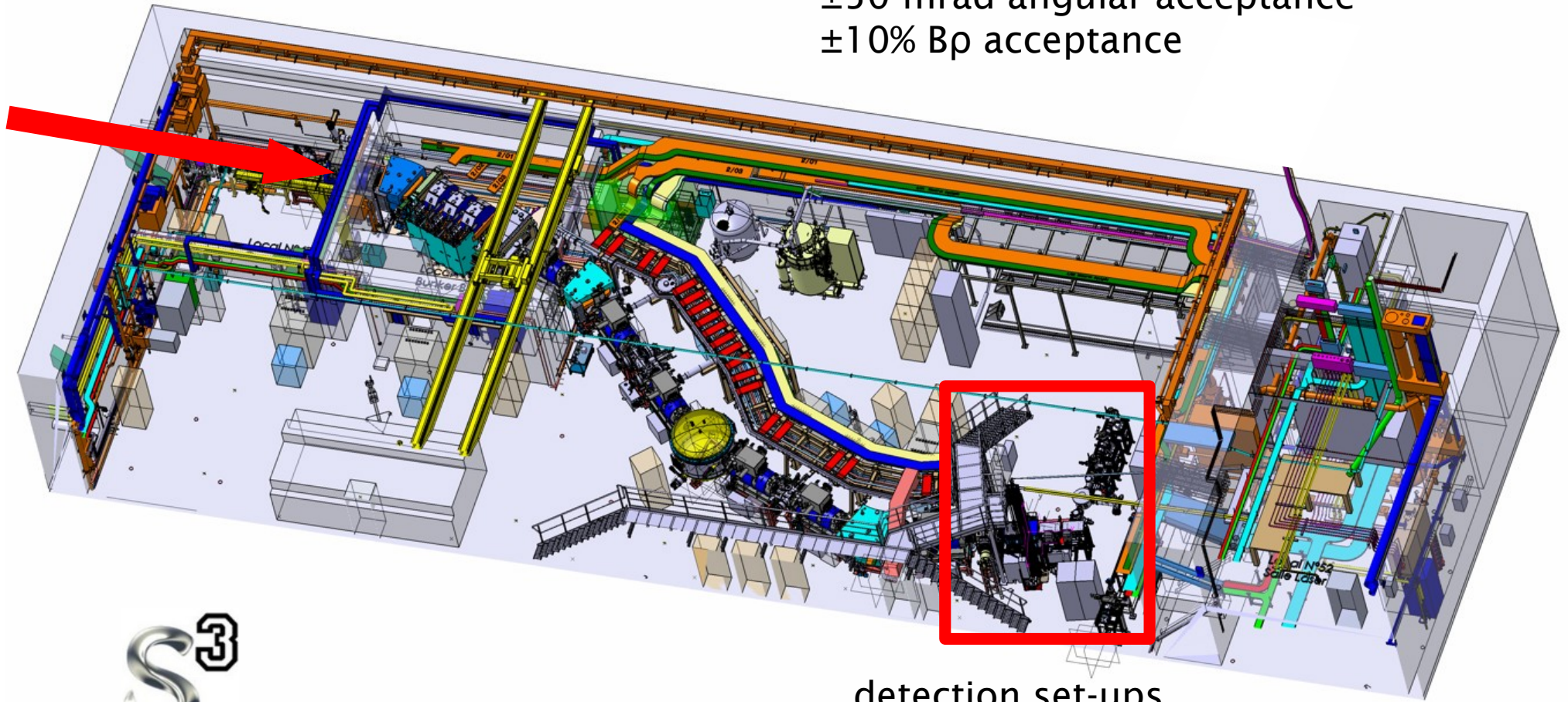
look for scheme with scanning laser + 355 nm

reduce background with intermediate step + 1064 nm

(trans)actinides are major physics case at S3

Spiral-2 S³ spectrometer

10 μA = 10^{14} pps beam on target
 10^{13} beam rejection
1/300 mass resolution
 ± 50 mrad angular acceptance
 $\pm 10\%$ Bp acceptance



detection set-ups

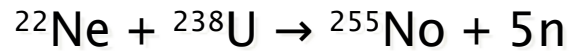
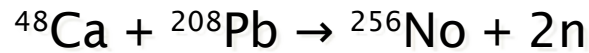
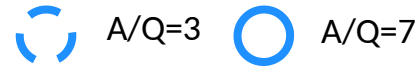


Heavy & Very Heavy Nuclei

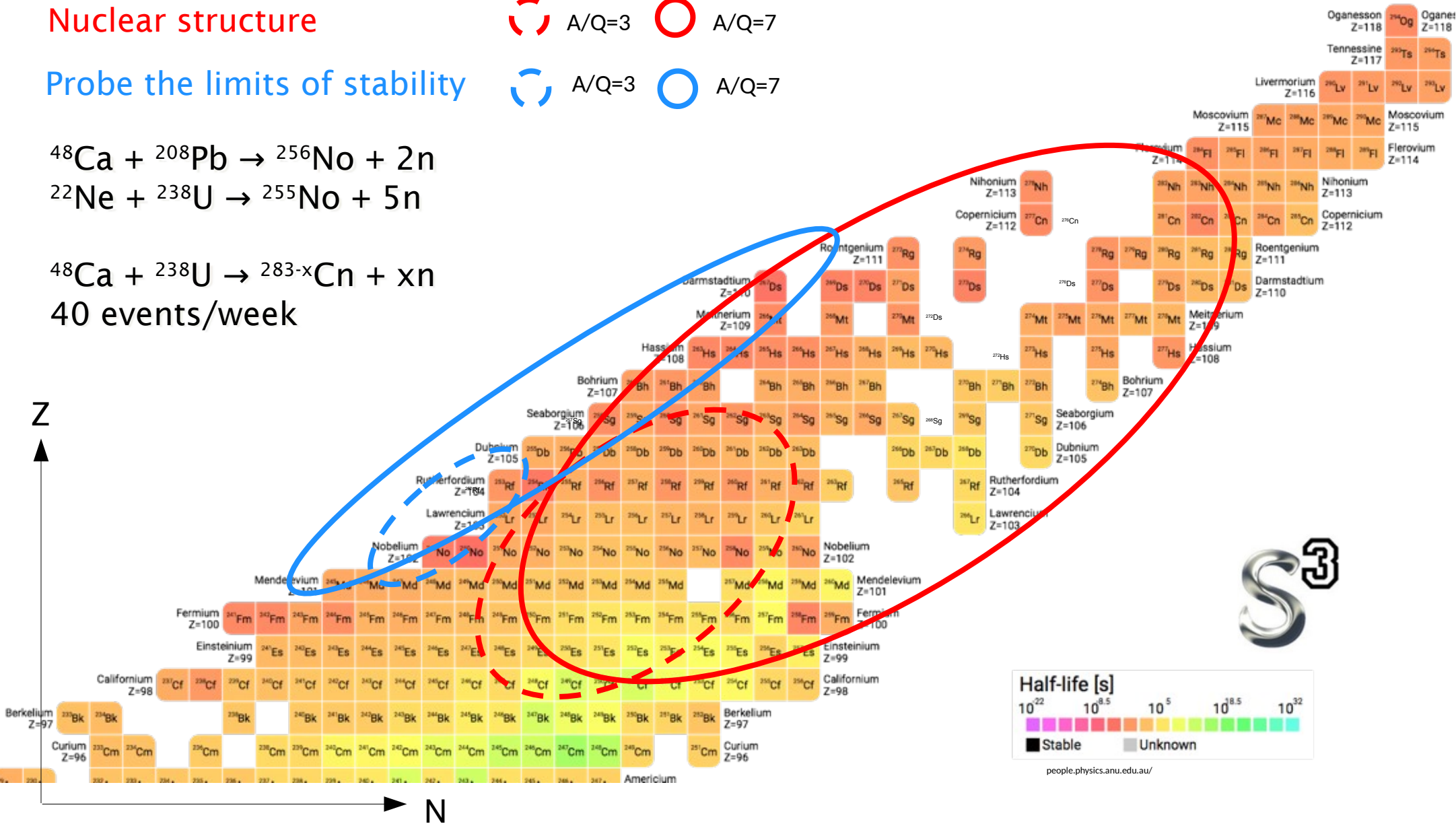
Nuclear structure

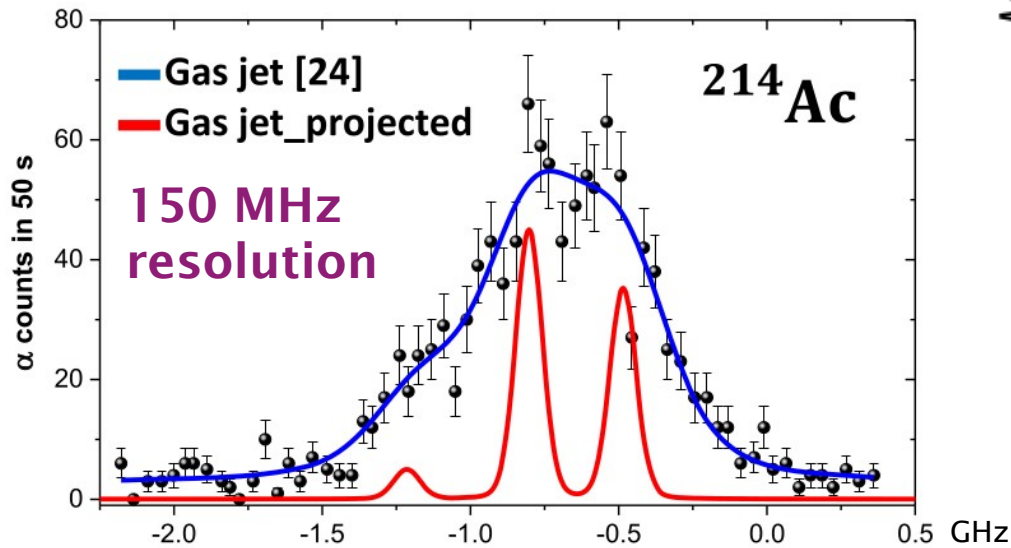
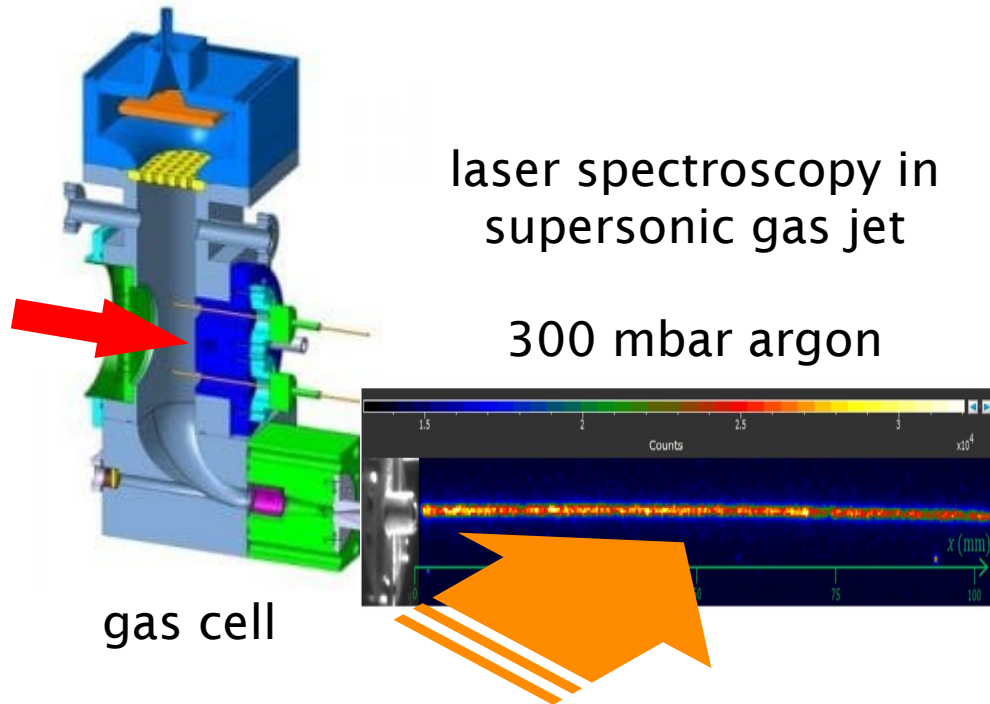


Probe the limits of stability

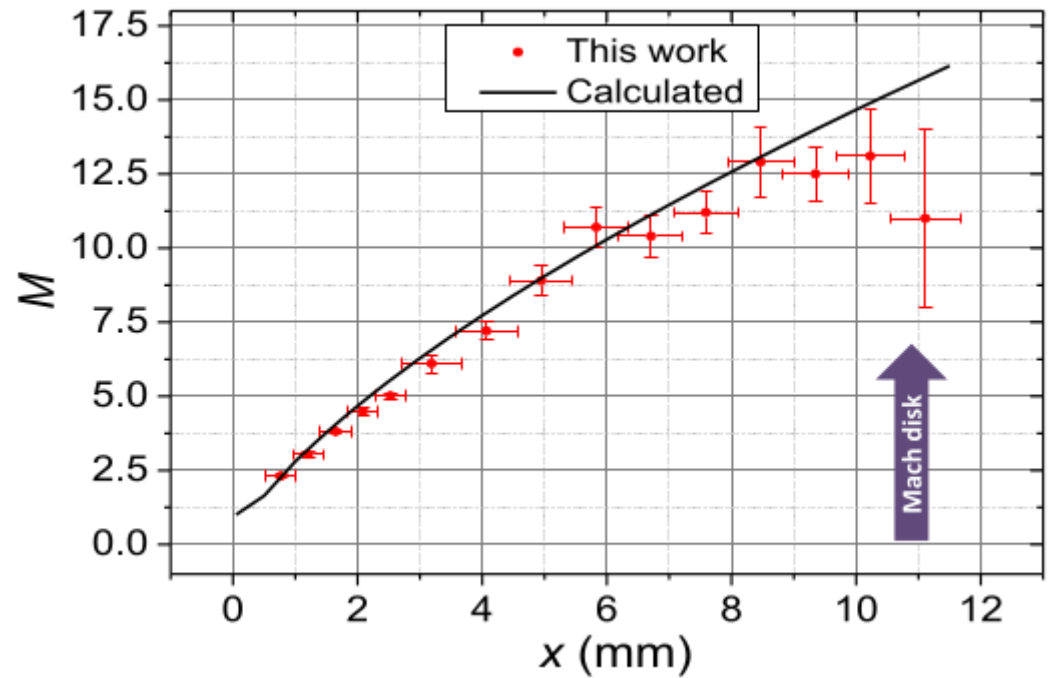
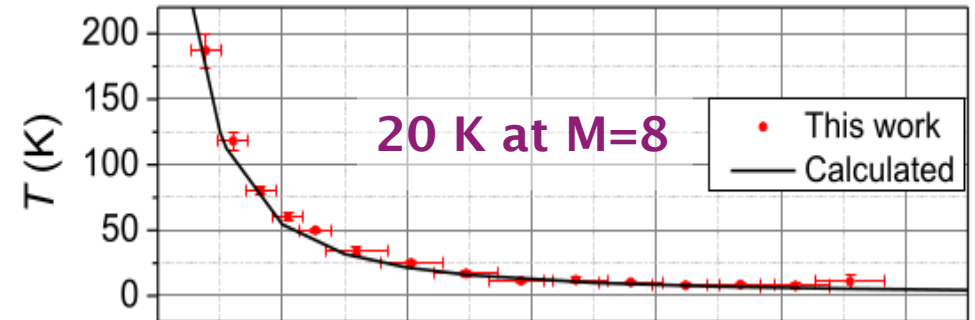


40 events/week





Ferrer et al., Nat. Comm. 8 (2017)



Zadvornaya et al., Phys. Rev. X 8, 041008 (2018)

off-line commissioning
at LPC Caen 2020-2023

mr-tof mass
spectrometer

RFQ-B

QMF

S-RFQ

gas cell



off-line commissioning
at LPC Caen 2020-2023

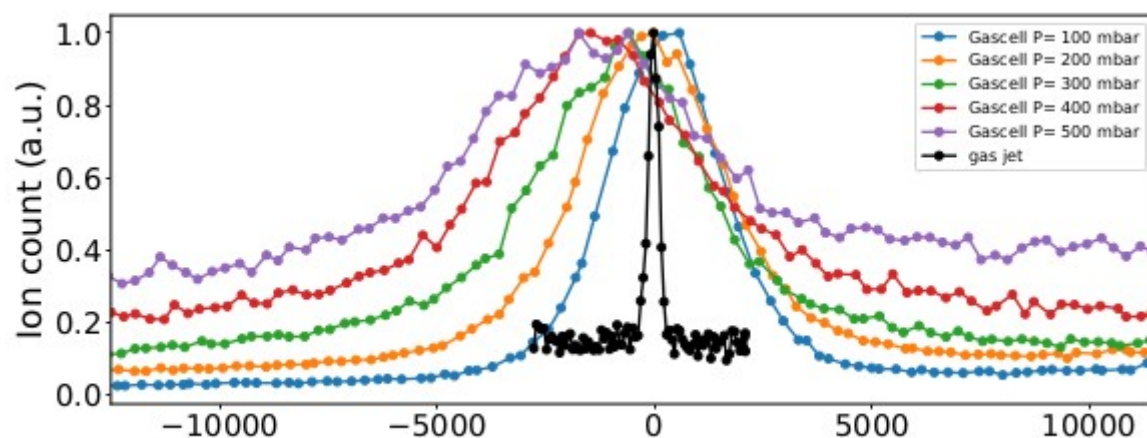
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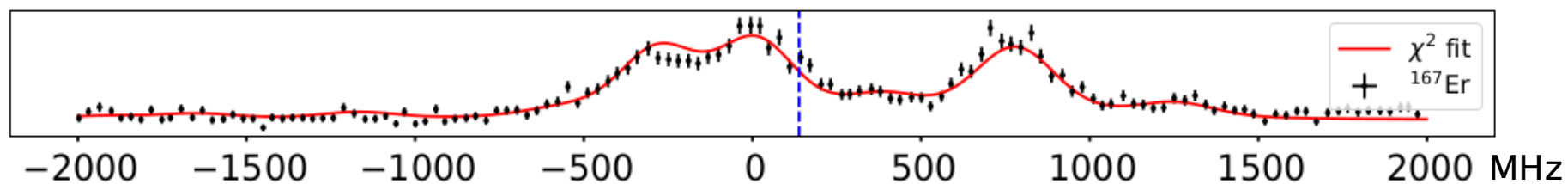
QMF

S-RFQ

gas cell



S³



8 orders of magnitude between Standard Model and experiment

Schiff theorem is overcome in heavy, octupole-deformed nuclei;
enormous enhancement from parity doublets in molecules

laser spectroscopy at Cris limited by $T = 300$ K

laser spectroscopy at S3 down to $T = 20$ K

genuine precision experiments at Desir or Orsay?

need nuclear spectroscopy of parity doublets

need theoretical guidance for S, MQM...

to be continued...

