JYVÄSKYLÄN YLIOPISTO
UNIVERSITY OF JYVÄSKYLÄ

New frontiers in optical spectroscopy of radioactive nuclei

Ruben de Groote, University of Jyväskylä, Finland

TRIUMF seminar 11/06/2020

Outline

- Collinear resonance ionization spectroscopy of copper
- First spectroscopy of RaF molecules
- High-precision measurements: magnetic octupole moments

- Looking ahead

Many collaborators involved from all over the world!

Nuclear theory:
J. Dobacewski (York), J. Holt (TRIUMF), W. Nazarewicz (MSU), P.-G. Reinhard (Erlangen), C. Yuan (Beijing)

Atomic theory:
B. K. Sahoo (PRL), J. Li (Beijing)

Quantum Chemistry:
R. Berger (U. Marburg)

Experimentalists:
Too many to name... (Leuven, JYU, Mainz , Manchester, ...)

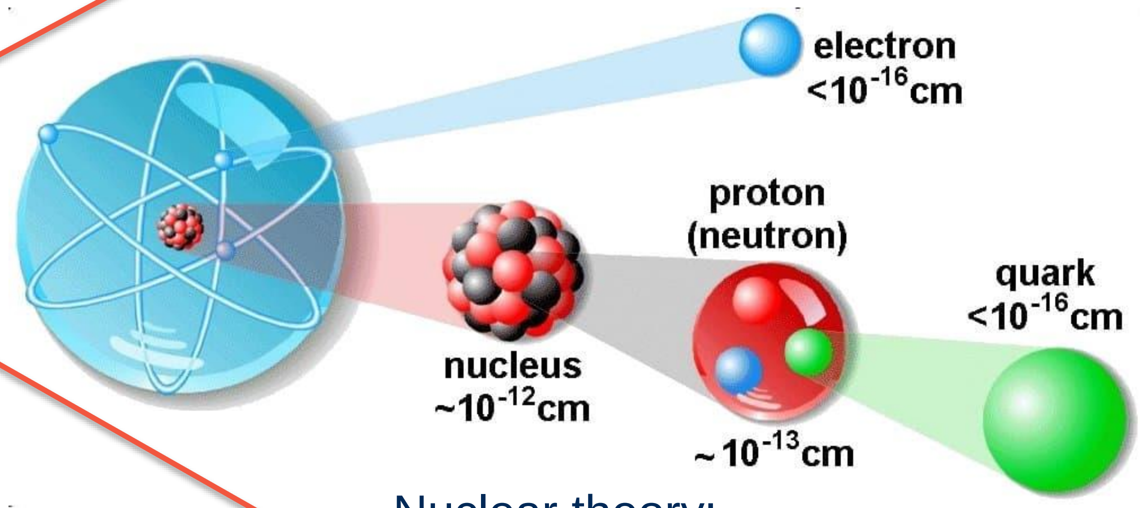
Studying the atomic nucleus

- Astrophysics
- Neutrino physics, dark matter searches
- Fundamental symmetries
- BSM physics, ...

Nuclear quantum-many body problem

Many different types of experiments

This seminar: atomic spectroscopy:
- μ, Q, charge radii



Nuclear theory:
- Config. interaction
- DFT
- ab-initio
- Collective models

QCD

Optical spectroscopy for nuclear structure

- Hyperfine interaction: interaction between nuclear moments and bound electrons

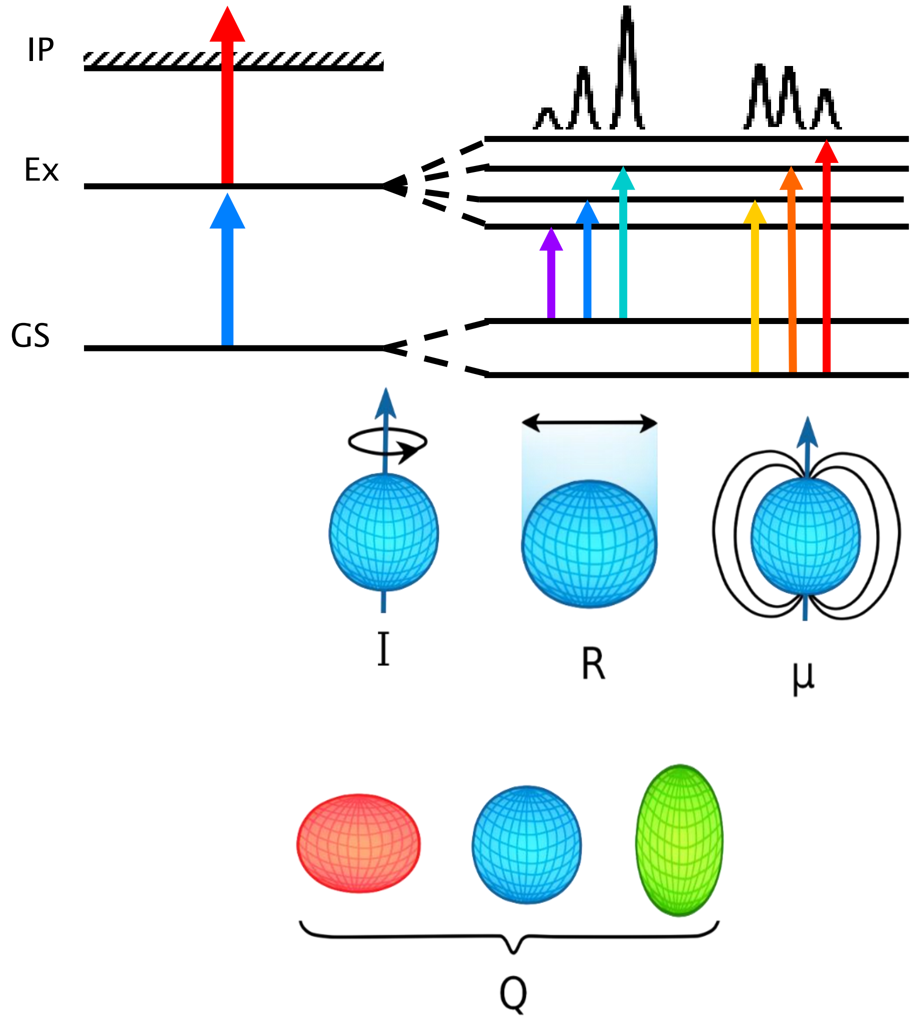
$$h\nu \sim \nu_0 + A\frac{C}{2} + B\frac{1}{4}\frac{(3/2)C(C+1)-2I(I+1)J(J+1)}{I(2I-1)J(2J-1)}$$

- Model-independent extraction of:

$\delta\langle r^2\rangle$ $A=\frac{\mu_I B_J}{IJ}$ $B=eQV_{zz}$

provided the atomic parameters are known (e.g. knowing mass and field shift is required)

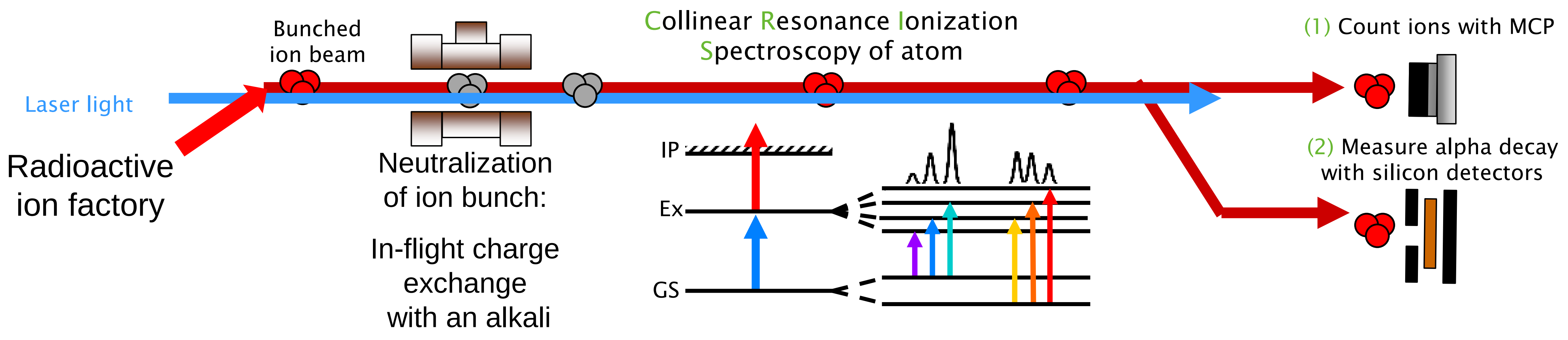
- Electromagnetic moments teach us about nuclear configurations, sizes and shapes
- Good tests of nuclear theory



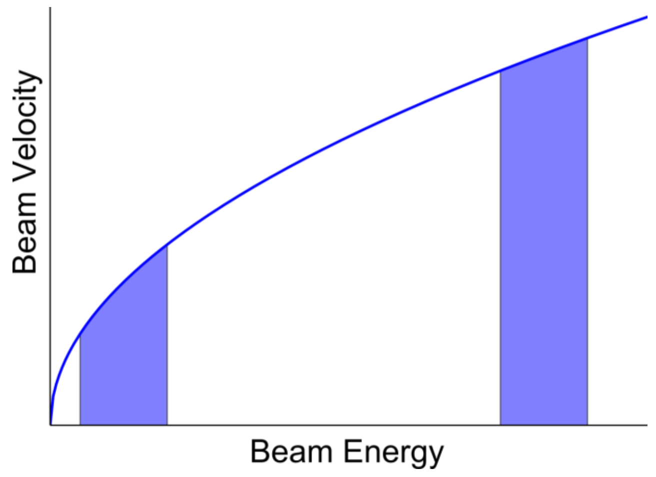
Results

- Collinear resonance ionization spectroscopy of copper

Collinear resonance ionization spectroscopy

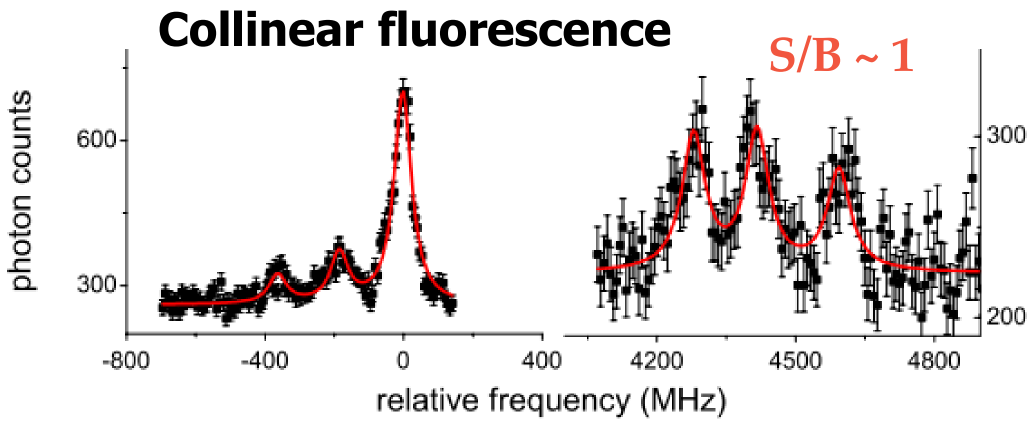


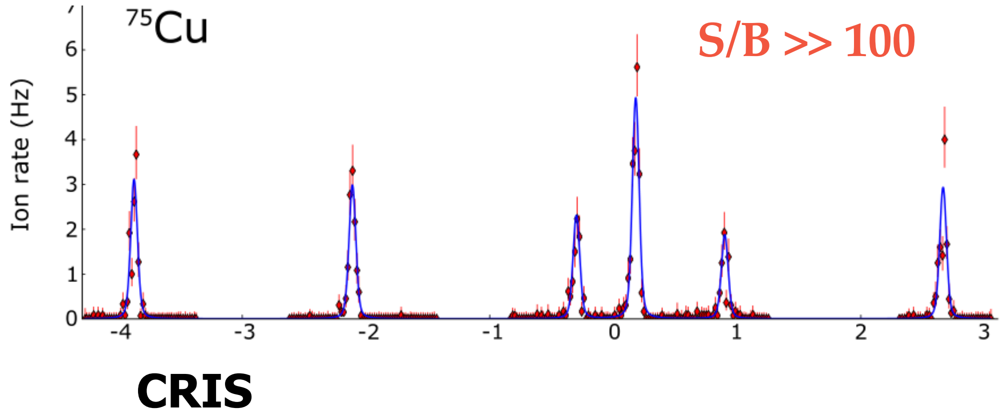
- Ions are produced in a hot environment and are typically only 'cooled' to room temperature using gas-filled RFQ
- High resolution is achieved not by reducing ΔE, but by reducing Δv through acceleration



CRIS of copper isotopes

- Neutron-rich isotopes $^{76\text{-}78}$Cu studied for the first time in high-resolution
  - Three additional isotopes in reach due to superior efficiency and S/B ratio
  - Background-free measurements achieved for ^{75,76,77}Cu





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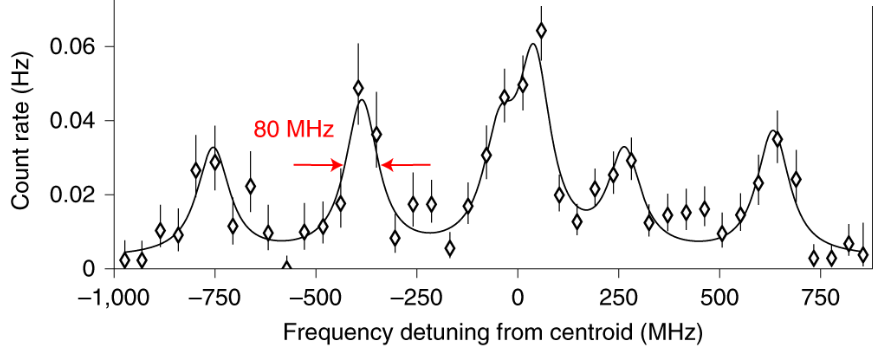
- ^{78}Cu could be studied (half life 330 ms)
  - Production: 20 nuclei per second, among >10^7 other unwanted atoms
    total 'sample' used: 200 000 ions
  - Shell-model calculations using interactions derived from chiral effective field theory reproduce results very well!
  - With 29 protons and 49 neutrons, this is a major computational challenge

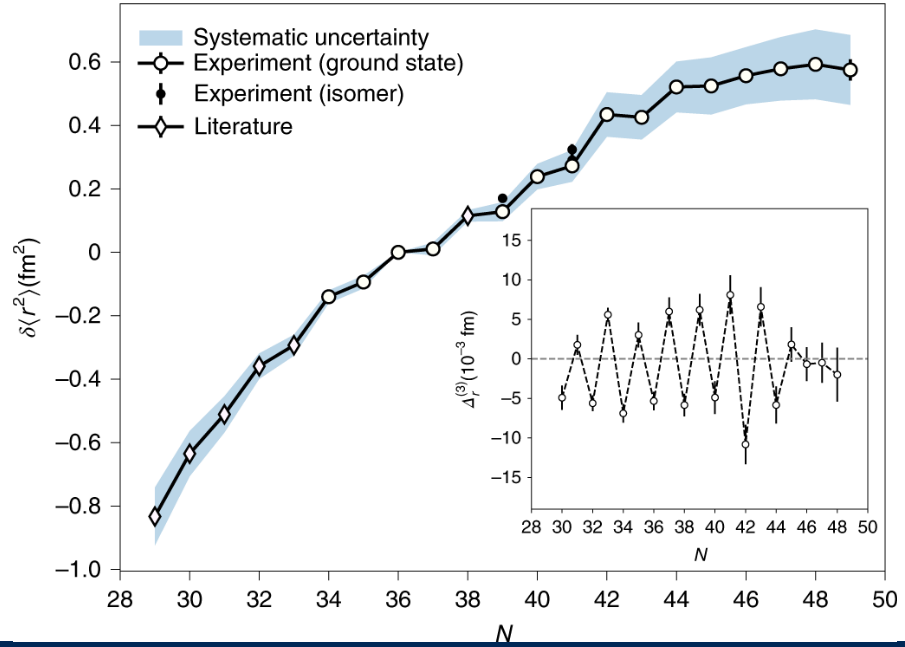
nature physics

Letter | Open Access | Published: 13 April 2020

Measurement and microscopic description of odd–even staggering of charge radii of exotic copper isotopes

R. P. de Groote, J. Billowes, [...] X. F. Yang





Copper charge radii as sensitive tests of state-of-the-art nuclear theory

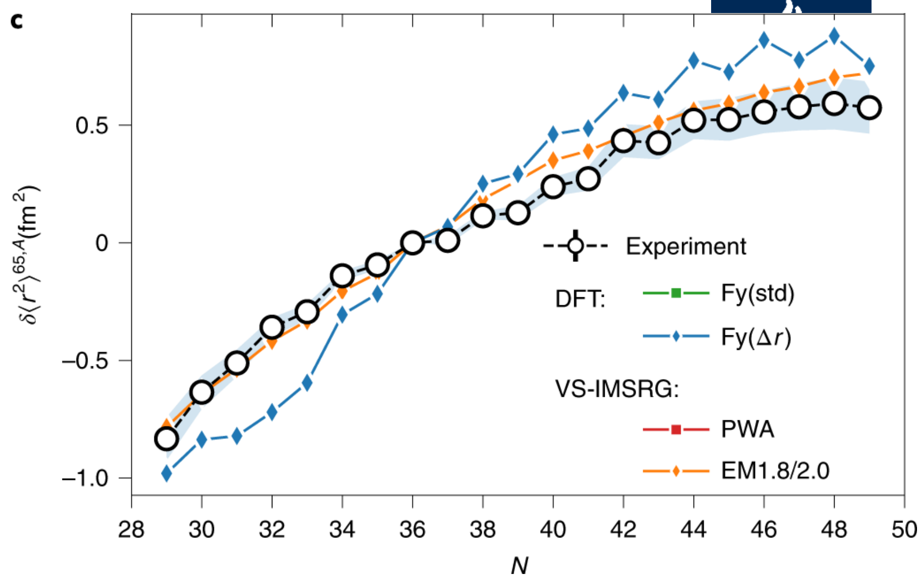
Staggering* of binding energies

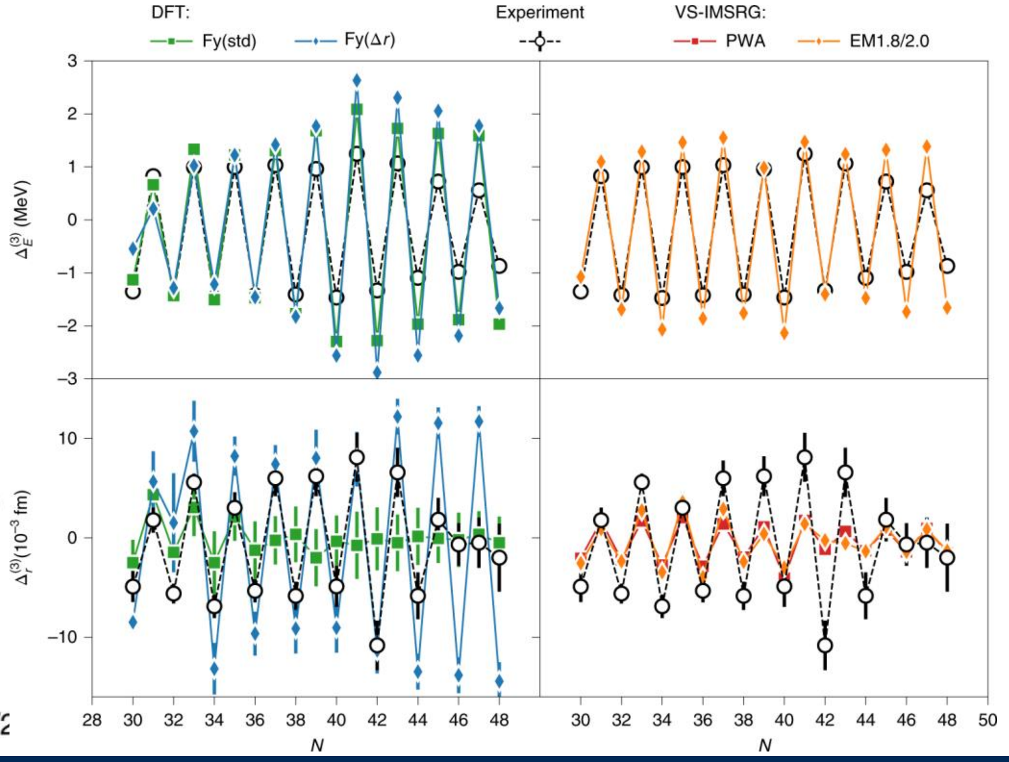
- good agreement with Fayans DFT and VS-IMSRG calculations
- Not unexpected, especially for DFT

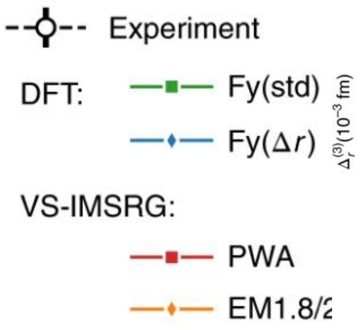
Staggering nuclear charge radii:

- good agreement for with Fayans DFT
  - / Due to introducing radii into the fitting procedure and adding terms proportional to density gradients
  - / Slight overestimation of radius OES, as observed also in e.g. Sn, Cd
- very good agreement with VS-IMSRG
  - / **Origins of OES are well-captured by fitting interactions to A<4 data!**

Small-scale variations well-captured by VS-IMSRG, bulk properties better reproduced with Fayans DFT





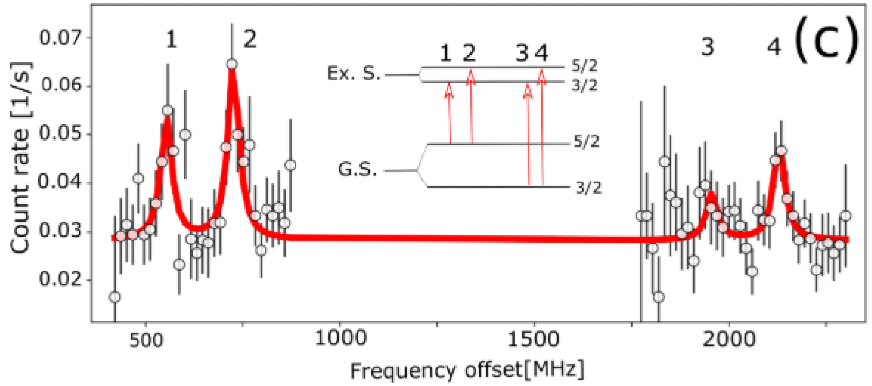


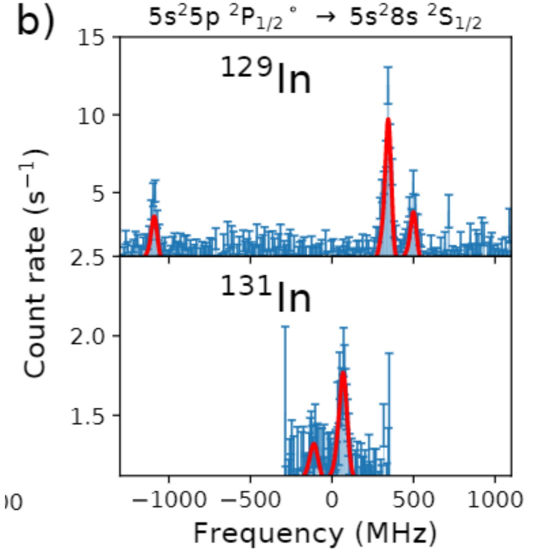
* Defined as three-point difference ½ $(x_{A+1} + x_{A-1} - 2x_A)$

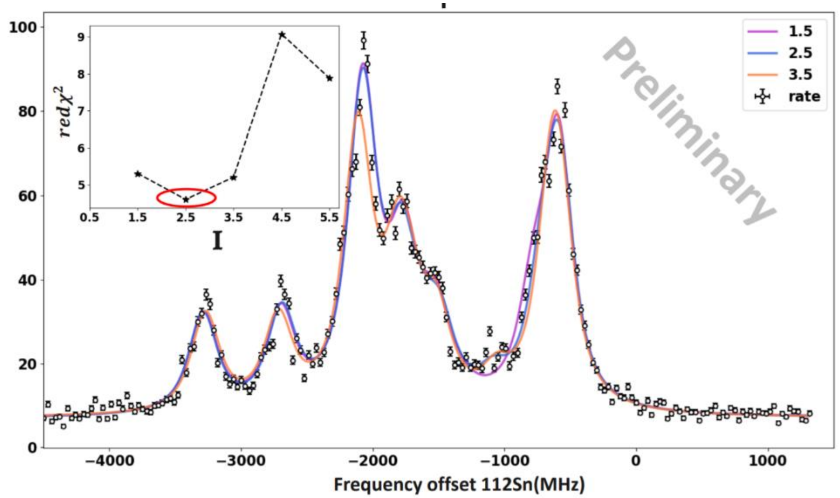
R. P. de Groote, et al. Nat. Phys. 16, 620–624 - 2020

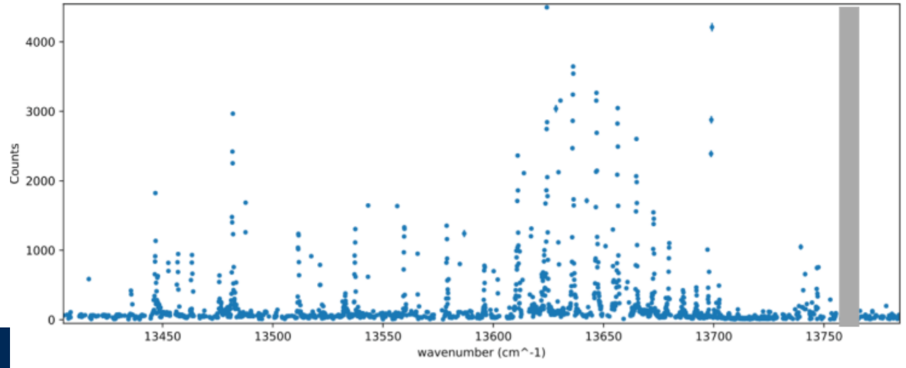
… and much, much more…

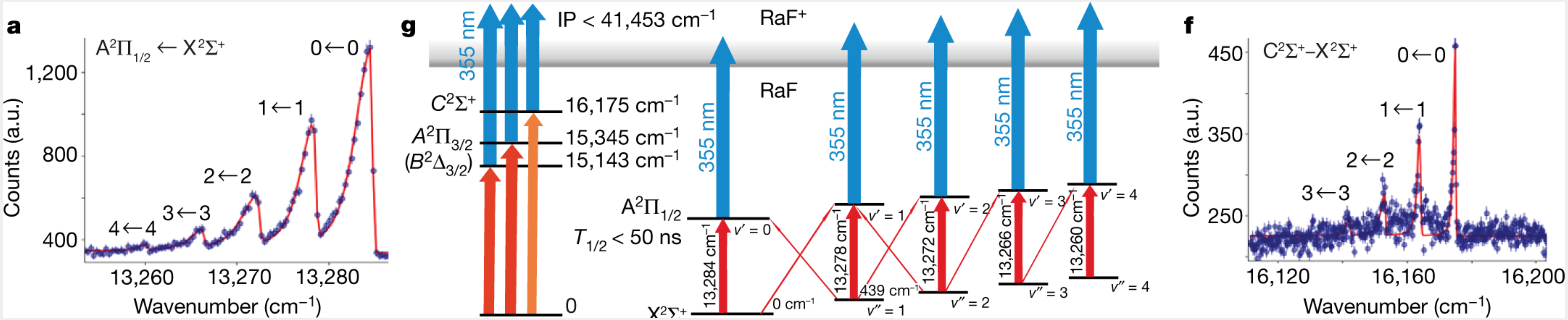
- Crossing N=32 with potassium ^{52}K
- Pushing towards N=50,82 with $^{101-131}In$
- Shell structure near N=Z=50 with $^{104-120}Sn$
- Many ongoing developments (field ionization, ion ionization, …)











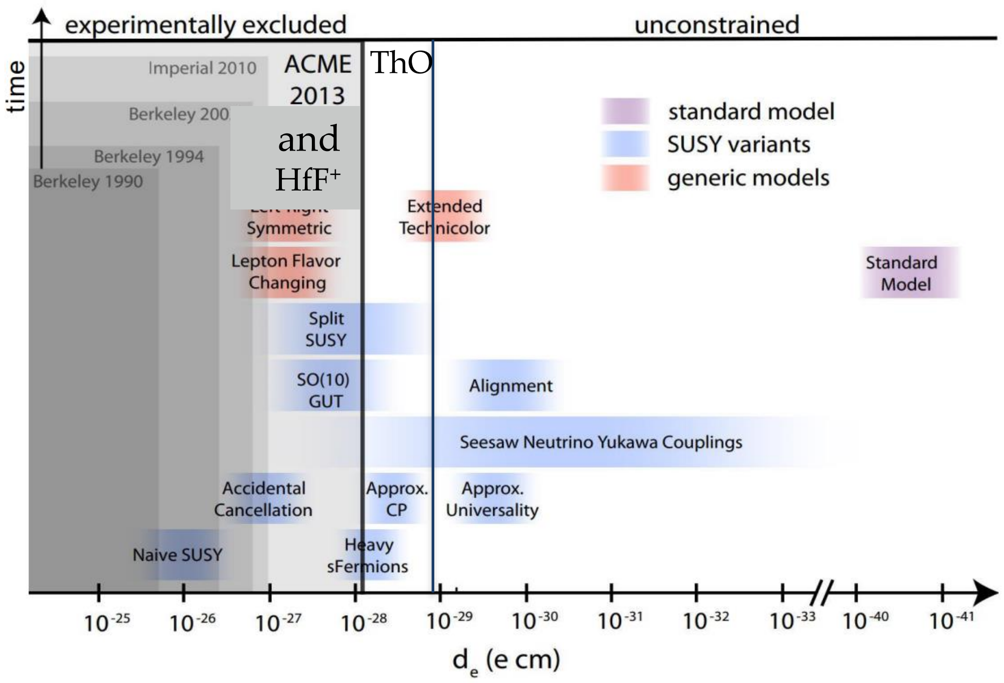
Results

- **First spectroscopy of RaF molecules**

Molecular probes for BSM physics

Molecules are very good probes for physics beyond the standard model

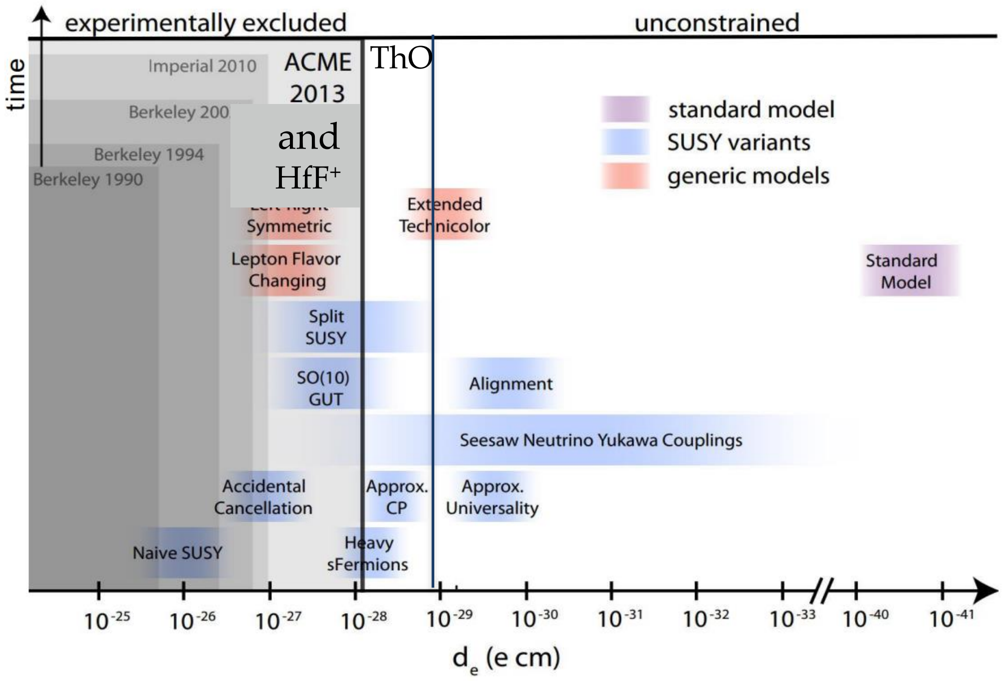
- eEDM: current limits set by molecular systems
  - Ion trap work: HfF^{+}, molecular beams (YbF, ThO)



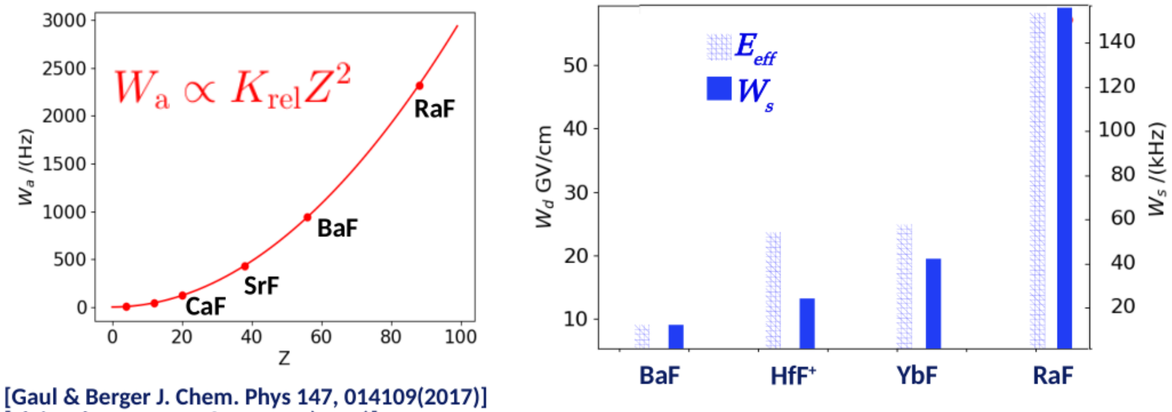
Molecular probes for BSM physics

Molecules are very good probes for physics beyond the standard model

- eEDM: current limits set by molecular systems
- P-odd and P,T-odd effects are enhanced strongly in well-chosen molecules



Example for RaF:



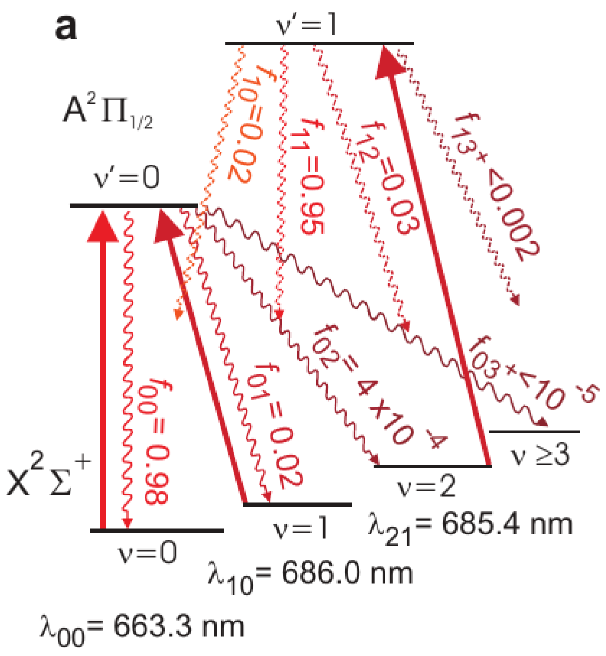
[Gaul & Berger J. Chem. Phys 147, 014109(2017)]
[Fleig. Phys. Rev. A 96, 040502 (2017)]

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

Molecular probes for BSM physics

Molecules are very good probes for physics beyond the standard model

- eEDM: current limits set by molecular systems
- P-odd and P,T-odd effects are enhanced strongly in well-chosen molecules
- Fluoride molecules: **can be laser cooled**!
    - SrF → Nature 467, 820-823 (2010), Nature Physics 13, 1173(2017)
    - YbF → Nature 473, 493 (2011), Phys. Rev. Lett. 120, 123201 (2018)
    - CaF → Nature Physics 14, 890 (2018), Phys. Rev. Lett. 120, 163201 (2018)
    - RaF …..



Radioactive molecule: *zero* spectroscopic info known…

- Can we do spectroscopy on radioactive molecules?
- Can we find out if RaF can be laser-cooled?

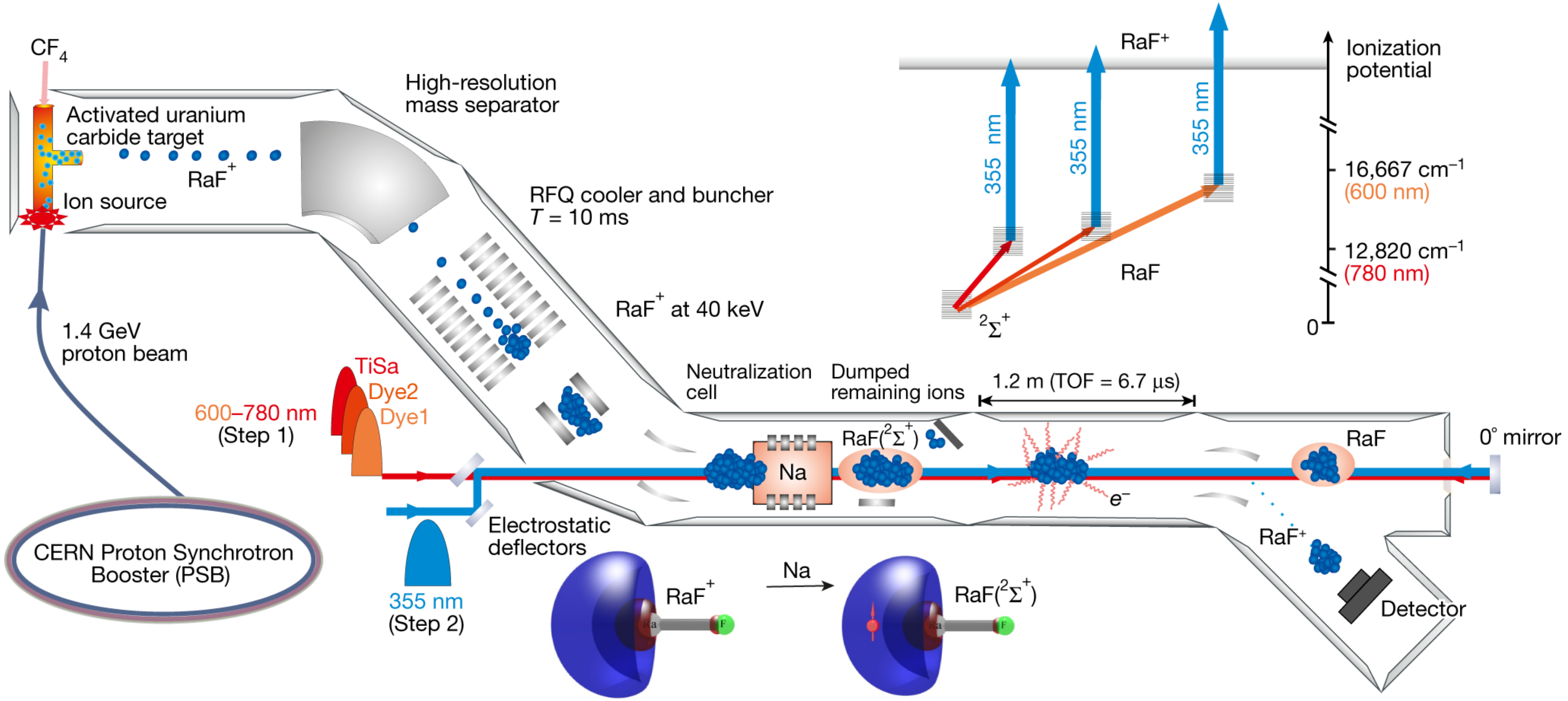
Measuring the first spectra of RaF

nature

Article | Open Access | Published: 27 May 2020

Spectroscopy of short-lived radioactive molecules

R. F. Garcia Ruiz, R. Berger, […] X. F. Yang



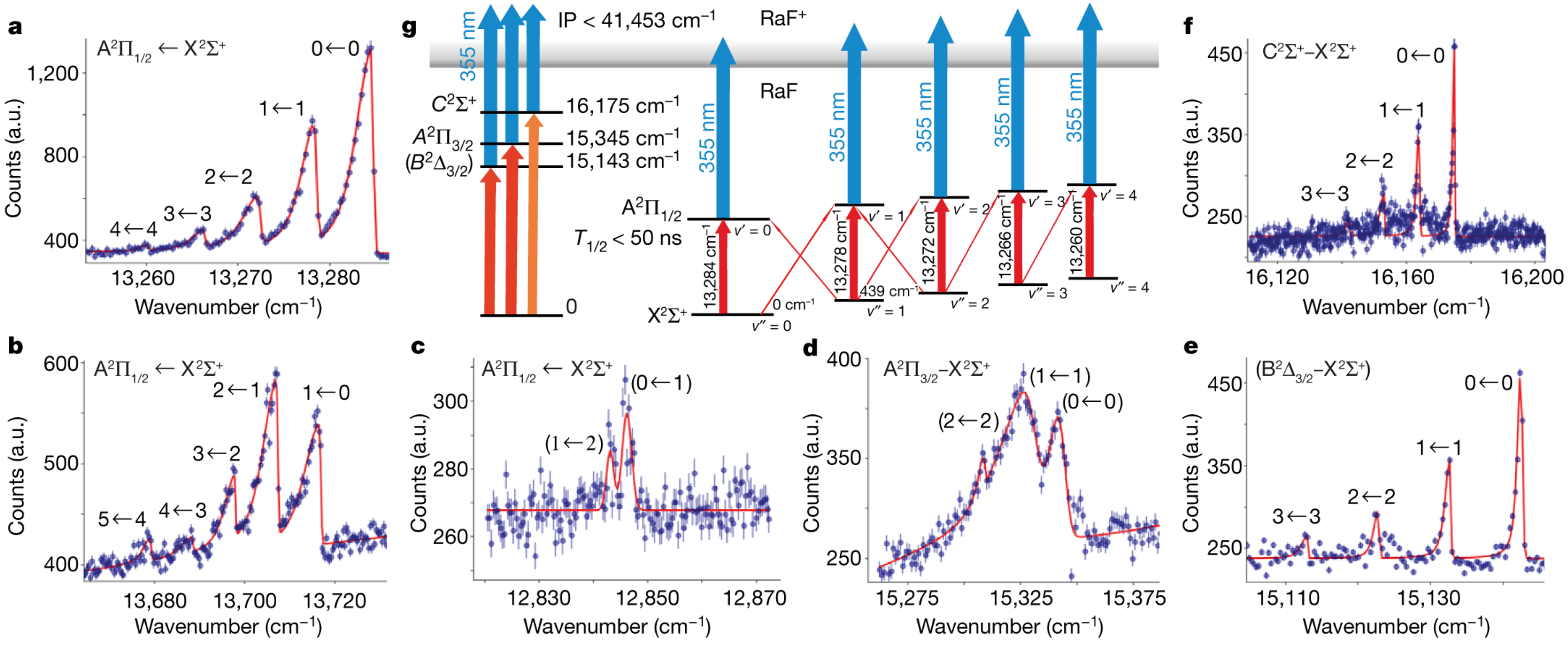
No lack of challenges…

- Production rate? Target had been dismounted for a while…
- Fluorination efficiency?
- Breakup in cooler?
- Breakup in charge exchange?
- Spread of population after charge exchange?
- Ionization potential too high to ionize?
- Non-resonant laser ionization by accident?
- Molecular breakup rather than ionization?
- Predictions ~1000 cm^{-1} precision: required scanning time too large?
- …….

Measuring the first spectra of RaF

Many results!

- Excitation energy of first few states
- Vibronic structure constants established
- Established structure is suitable for laser cooling
- Ionization potential measured
- Two- and three-step laser ionization schemes found
- High-resolution measurements of molecular hyperfine structure obtained

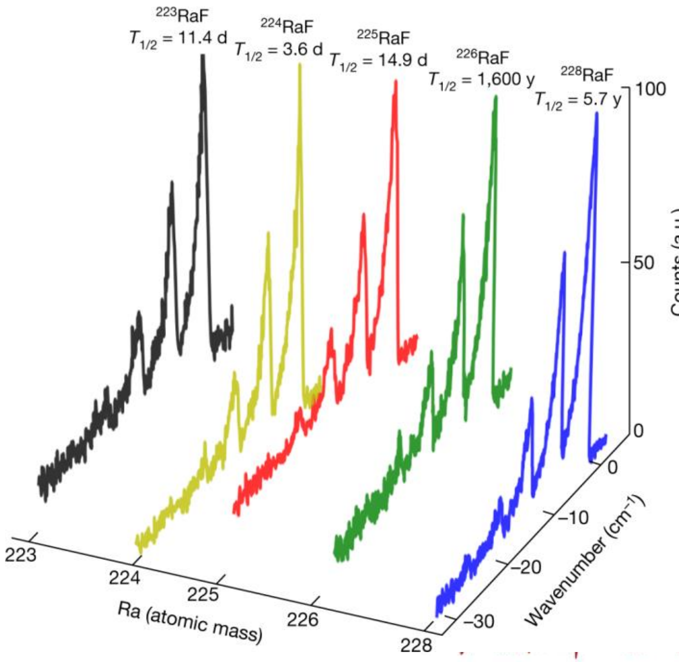


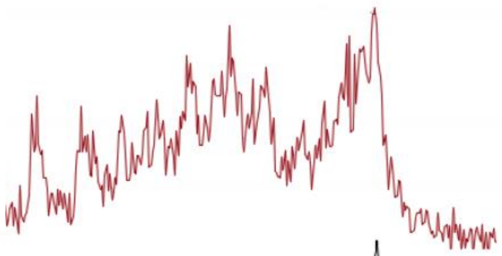
Future directions?

- Further spectroscopic studies of RaF to fully characterise the rotational and hyperfine structure
  - Demonstrated even short-lived, weakly produced isotopologues are in reach
  - Tests Quantum Chemistry calculations which are required to extract eEDM from measurements
  - ISOLDE proposal INTC-P-555

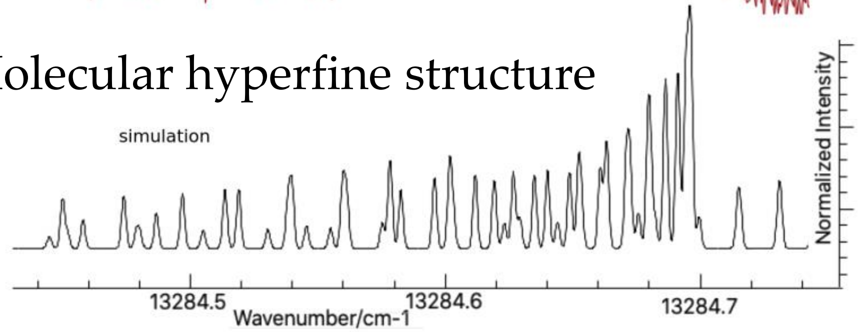
- Technical developments towards characterization of molecules and measurements of BSM effects
  - TRIUMF LOI: S2068-LOI

- Molecules may provide a more sensitive probe for nuclear structure as well
  - E.g. KF^+ could be used to measure extremely precise Q-moments of potassium, RbCl for Chlorine
    (both not possible with current laser spectroscopy methods)





Molecular hyperfine structure



Nuclear moment ratio

$Q(^{41}K)/Q(^{39}K)$ 1.217699 (0.000055)

Paquette, G., et al. "The hyperfine spectrum of KF." *Journal of Molecular Structure* 190 (1988): 143-148.

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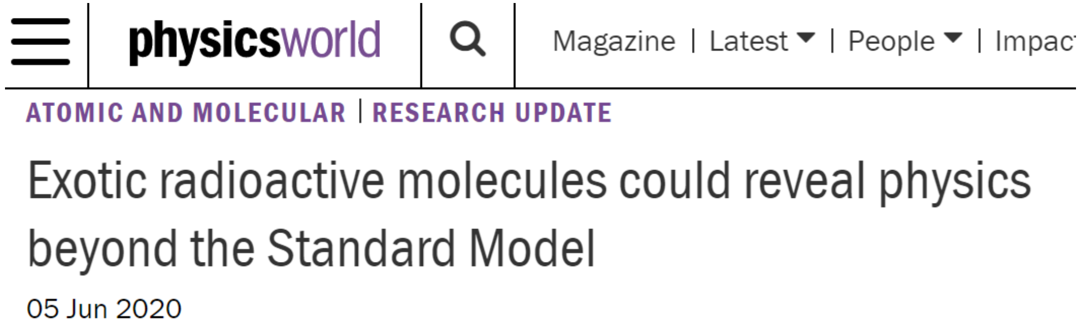
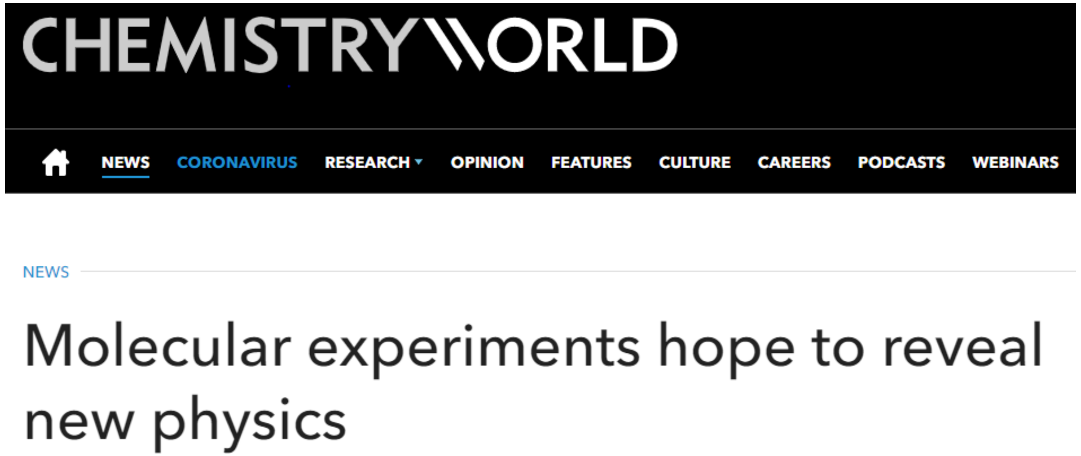
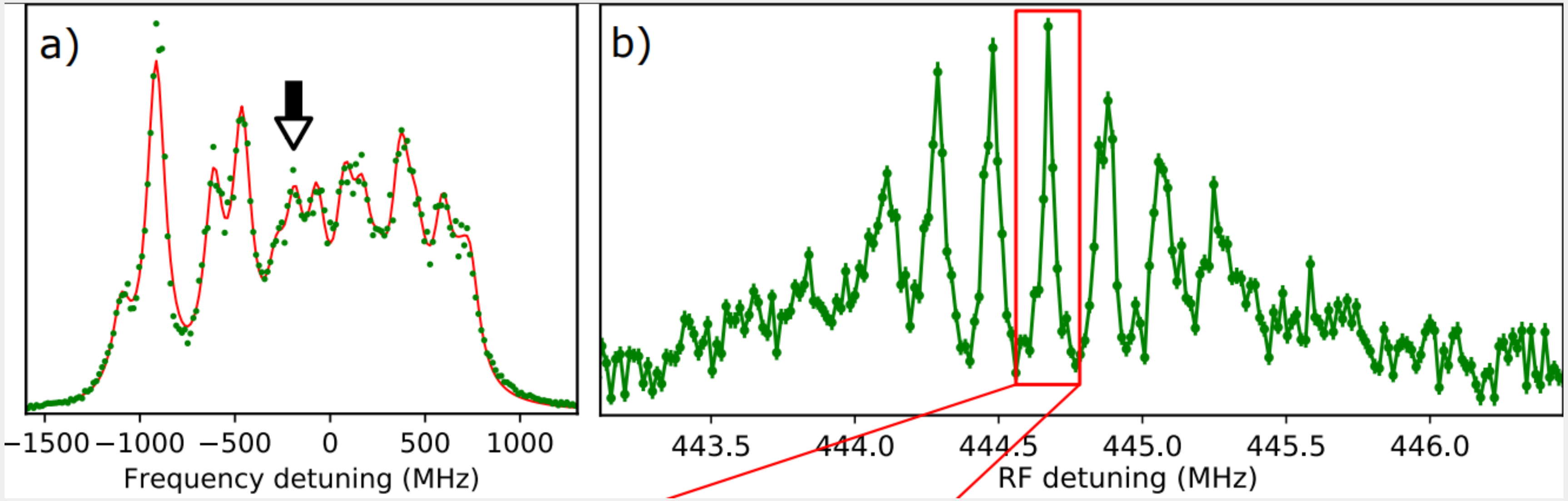






Results

- High-precision measurements: magnetic octupole moments

The precision frontier for optical spectroscopy

- Magnetic dipole moments
- Electrical quadrupole moments and charge radii
- Hyperfine anomaly [1]
  - Relates to the distribution of magnetization inside nuclear volume
- Higher-order moments
  - **Magnetic octupole**, electric hexadecupole, …
- Higher-order moments of the charge radii
  - E.g. $\langle r^4 \rangle$ relates to surface thickness of nuclear density
- BSM physics from Hz-level isotope shift spectroscopy, molecular probes, …

GHz

In-source optical spectroscopy

Collinear, in-gas-jet, …

Current state-of-the-art

Laser-rf methods on fast ion beams

Feasible on RIB

Coming soon?

Atom/ion traps + laser cooling + ultra-narrow lines

Currently only on stable species

BSM searches

sub-Hz

[1] J. Zhang (张颉颃), et al. (FrPNC collaboration) Phys. Rev. Lett. 115, 042501 – 2015

Optical spectroscopy for nuclear structure

- Hyperfine interaction: interaction between nuclear moments and bound electrons

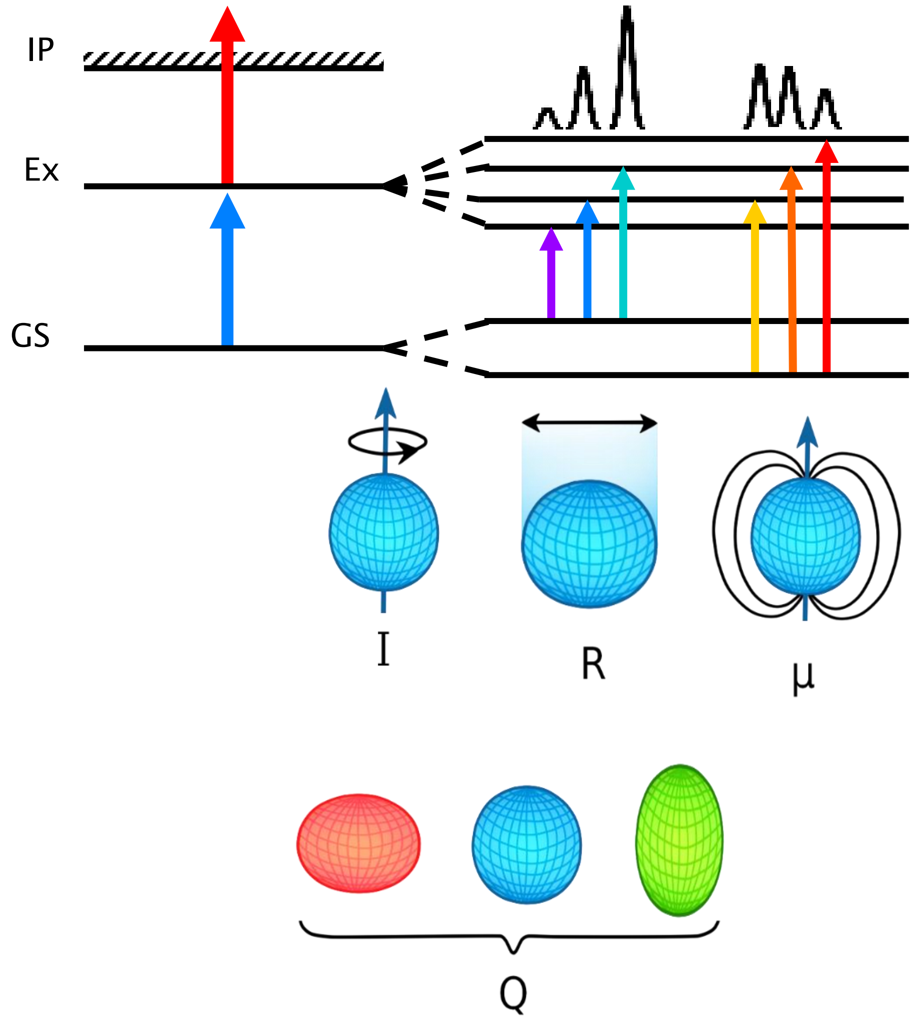
$$h\nu \sim \nu_0 + A\frac{C}{2} + B\frac{1}{4}\frac{(3/2)C(C+1) - 2I(I+1)J(J+1)}{I(2I-1)J(2J-1)}$$

- Model-independent extraction of:

$\delta\langle r^2\rangle$ $\quad A = \frac{\mu_I B_J}{IJ}$ $\quad B = eQV_{zz}$

provided the atomic parameters are known (e.g. knowing mass and field shift is required)

- Electromagnetic moments teach us about nuclear configurations, sizes and shapes
- Good tests of nuclear theory



Magnetic octupole moments?

- Hyperfine interaction: interaction between nuclear moments and bound electrons

$$h\nu \sim \nu_0 + A\frac{C}{2} + B\frac{1}{4}\frac{(3/2)C(C+1)-2I(I+1)J(J+1)}{I(2I-1)J(2J-1)} + C_{HFS}\, f(I,J,F)$$

- Model-independent extraction of:

$\delta\langle r^2\rangle$ $A=\frac{\mu_I B_J}{IJ}$ $B=eQV_{zz}$ $C = -\Omega\langle JJ|T_3^{(e)}|JJ\rangle$

provided the atomic parameters are known (e.g. knowing mass and field shift is required)

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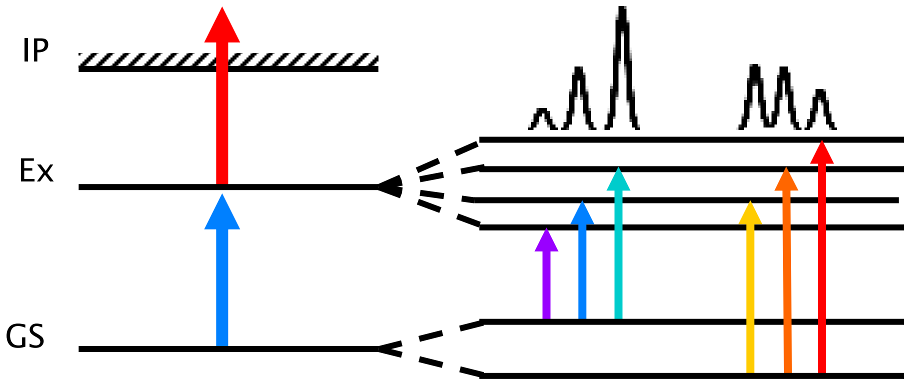
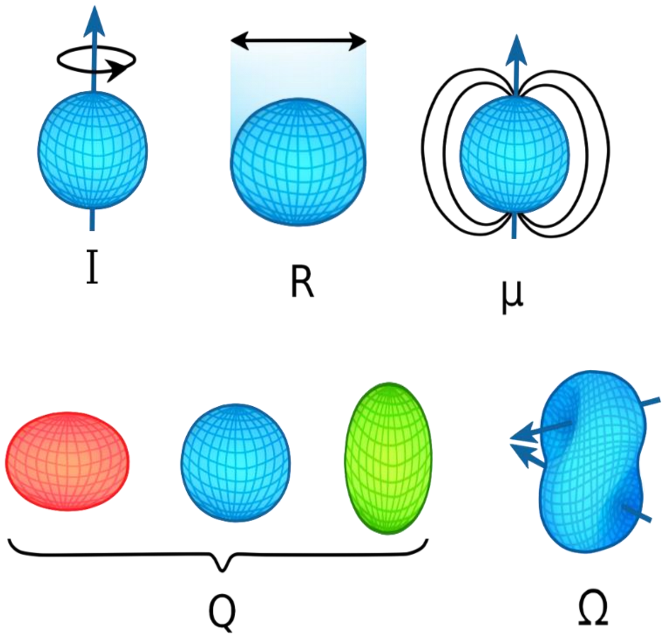
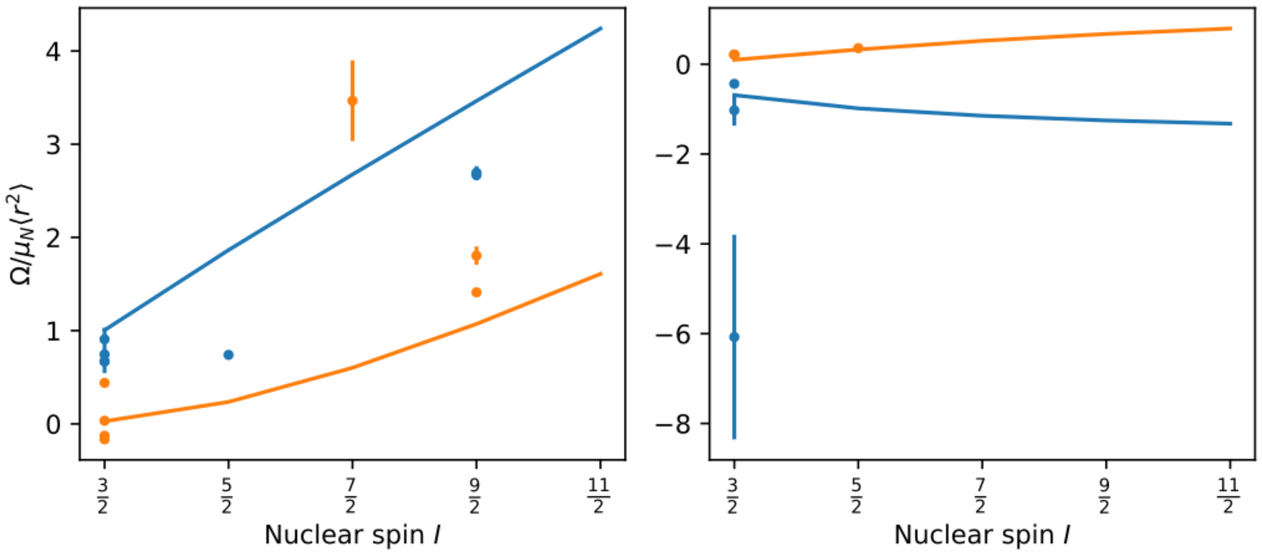




Magnetic octupole moments

- Pen-and-paper prediction for magnetic octupole moments: *Schwartz* lines
- Very few experimental numbers exist (~20)
- All experimental data points lie close to these lines assuming the same spin g-factor quenching
  - However, deviations are larger than for dipole moments

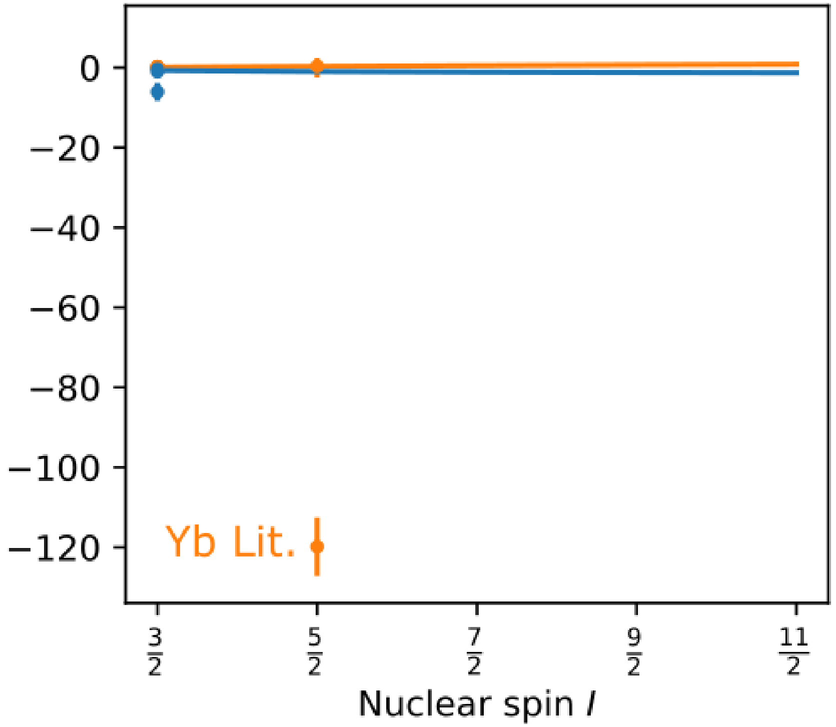
- Can we learn more about the **nuclear magnetization distribution** (neutron skin)? Origins of **quenching** and **MEC**? Is the simple shell model as good as it is for dipole moments?



$$\Omega/\mu_N\langle r^2\rangle = \frac{3}{2}\frac{2I-1}{(2I+4)(2I+2)} \times \begin{cases}(I+2)[(I-\frac{3}{2})g_l+g_s], & I=l+\frac{1}{2}\\ (I-1)[(I+\frac{5}{2})g_l-g_s], & I=l-\frac{1}{2}\end{cases}$$

Magnetic octupole moment of ytterbium: what conventional spectroscopy can do

- Extremely large magnetic octupole moment measured for ^{173}Yb (stable)



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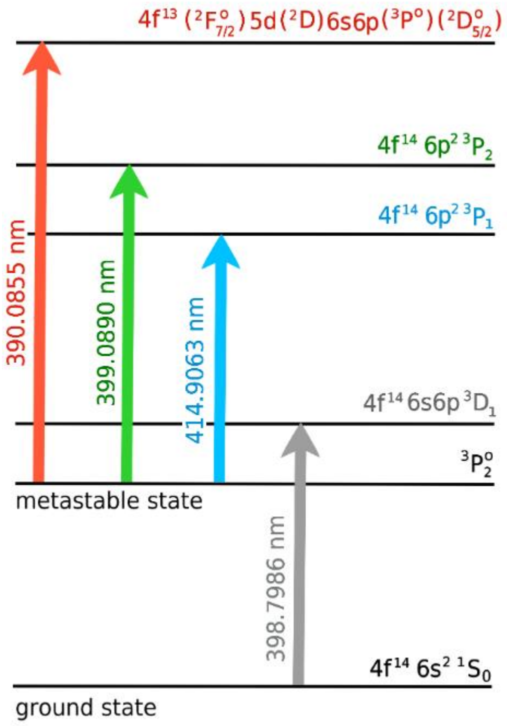
- Hyperfine octupole constant so large it is measurable even with conventional collinear laser spectroscopy



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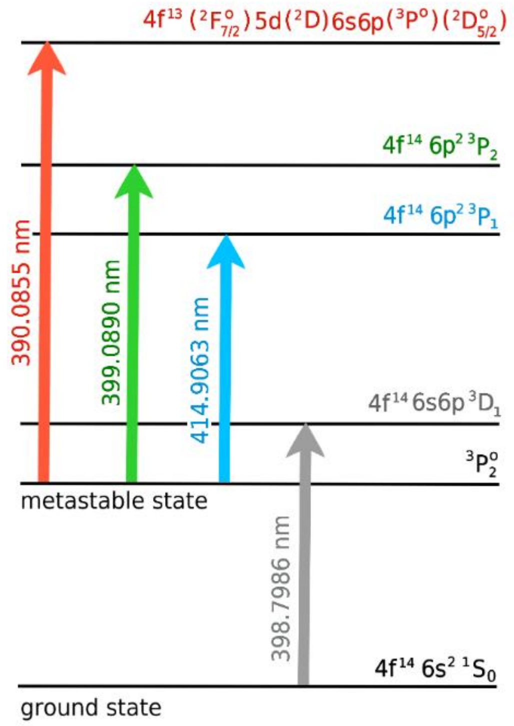
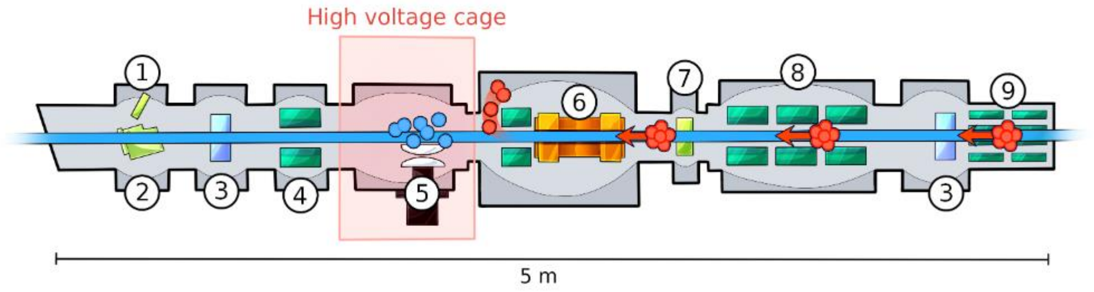
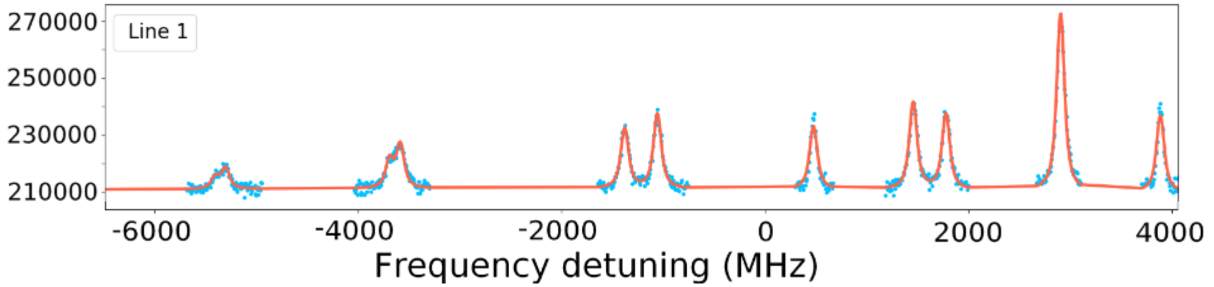


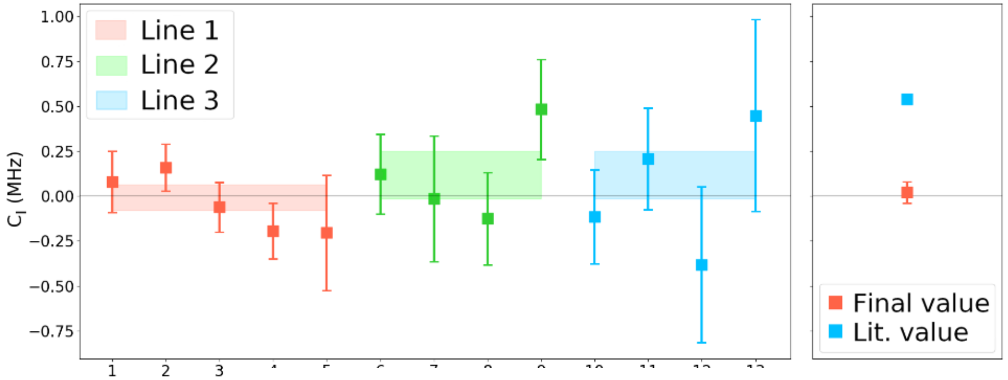


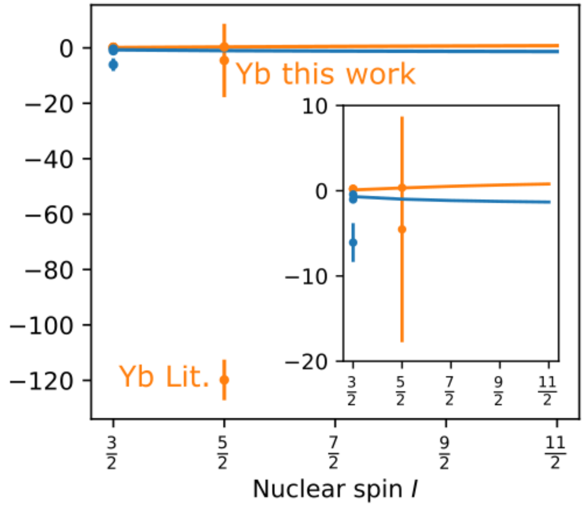


Alok K. Singh, et al. Phys. Rev. A 87, 012512 – 2013

Magnetic octupole moment of ytterbium: what conventional spectroscopy can do

- Extremely large magnetic octupole moment measured for ^{173}Yb (stable)
- Hyperfine octupole constant so large it is measurable even with conventional collinear laser spectroscopy
  - Our value restores agreement with expectations from single-particle estimates
  - Future work will require higher resolution to further reduce uncertainties

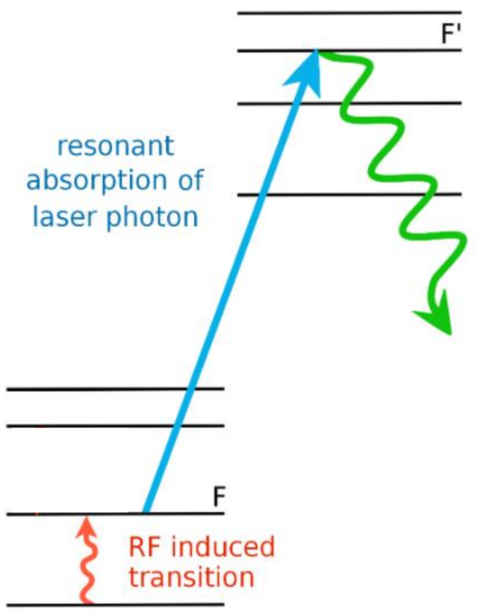


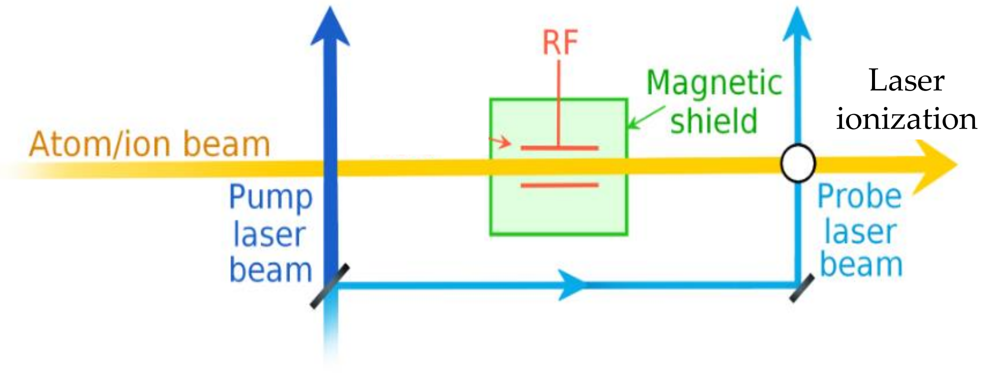


Magnetic octupole moment of scandium: using laser-rf techniques instead

- Direct radiofrequency spectroscopy of hyperfine levels
  - Linewidth ~ 1 / interaction time ~ 10kHz

- We developed a combination of laser-rf spectroscopy and laser resonance ionization spectroscopy
  - Using ionization scheme developed at TRIUMF ☺

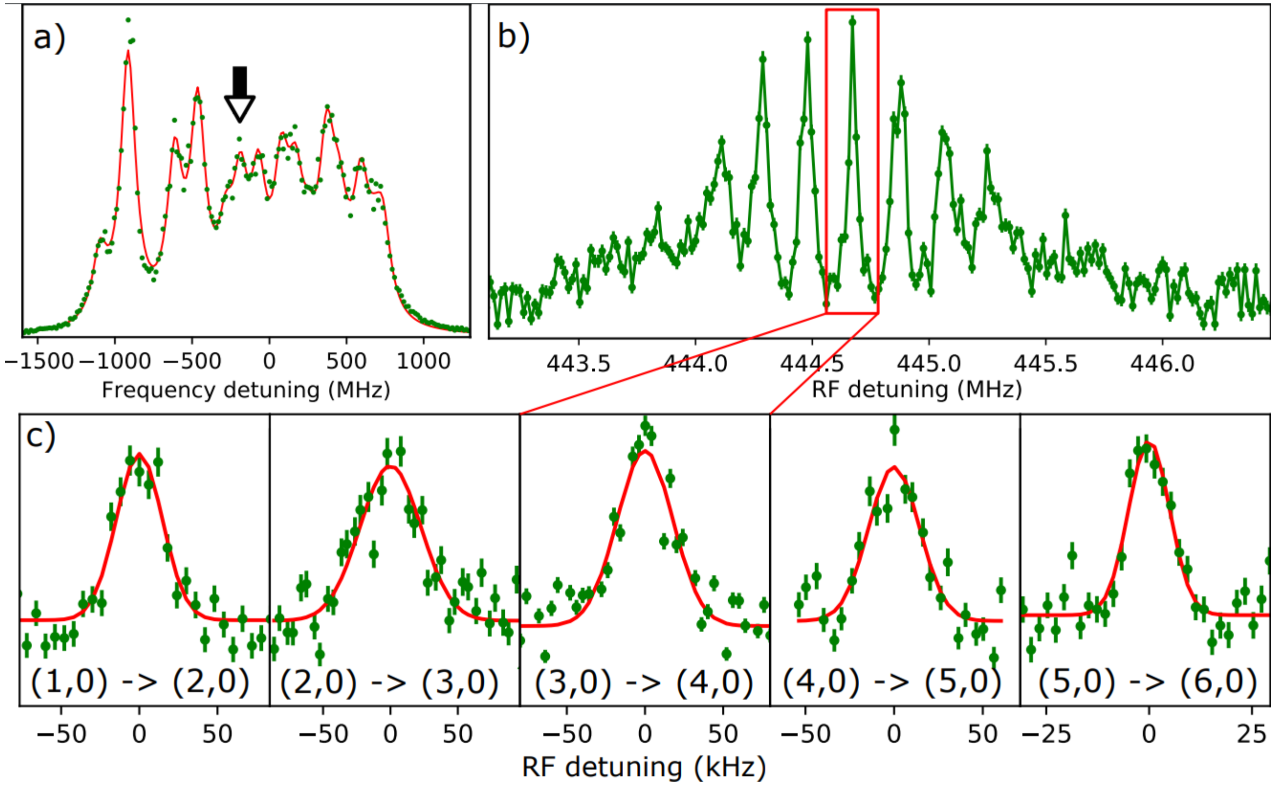




Magnetic octupole moment of scandium: using laser-rf techniques instead

Optical spectrum

Several zeeman components



B-field-insensitive transitions

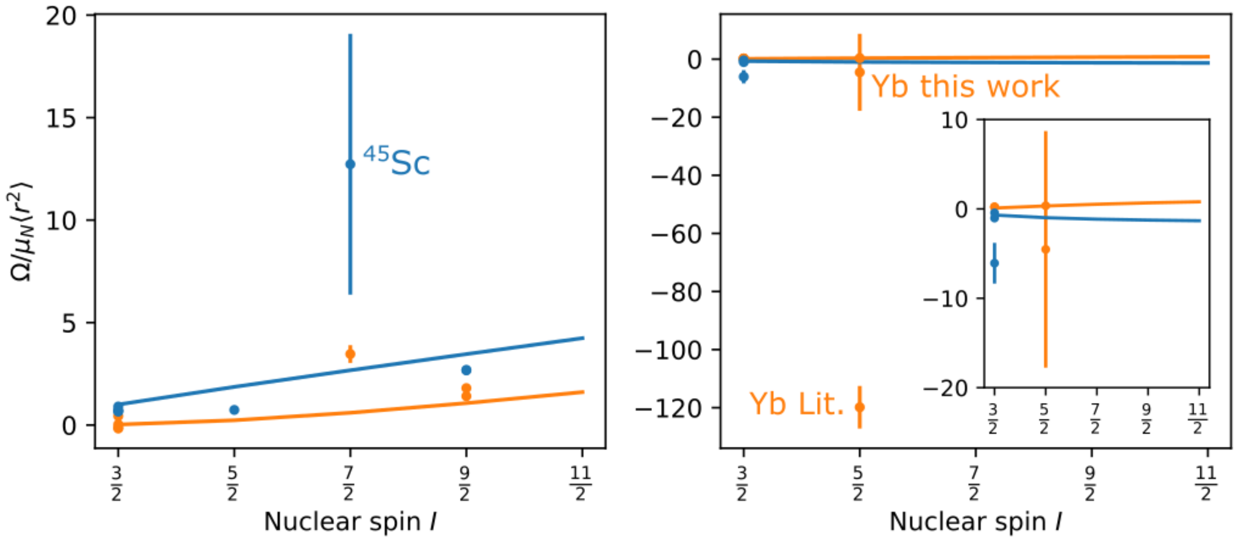
R. P. de Groote et al., submitted to Phys. Rev. Lett.

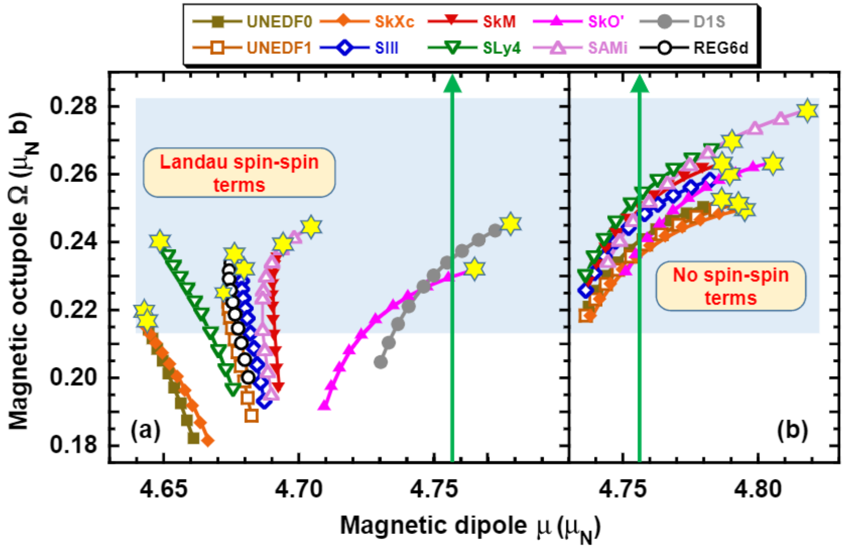
Magnetic octupole moment using laser-rf techniques in

- Resulting precision is orders of magnitude better than conventional spectroscopy

	Theory	Corrected	
	This work	Ref. [17]	This work
A [MHz]	108(2)	109.033(1)	109.0329(1)
B [MHz]	-37.7(5)	-37.373(15)	-37.371(1)
C [kHz]		1.5(12)	-0.25(12)
Ω [μ_Nb]		-9.4(75)	+1.6(8)

- First steps to interpret this unusual observable made using shell-model and nuclear DFT
  - Experimental value is larger than theoretical models can account for
  - Further theoretical and experimental work is definitely needed!



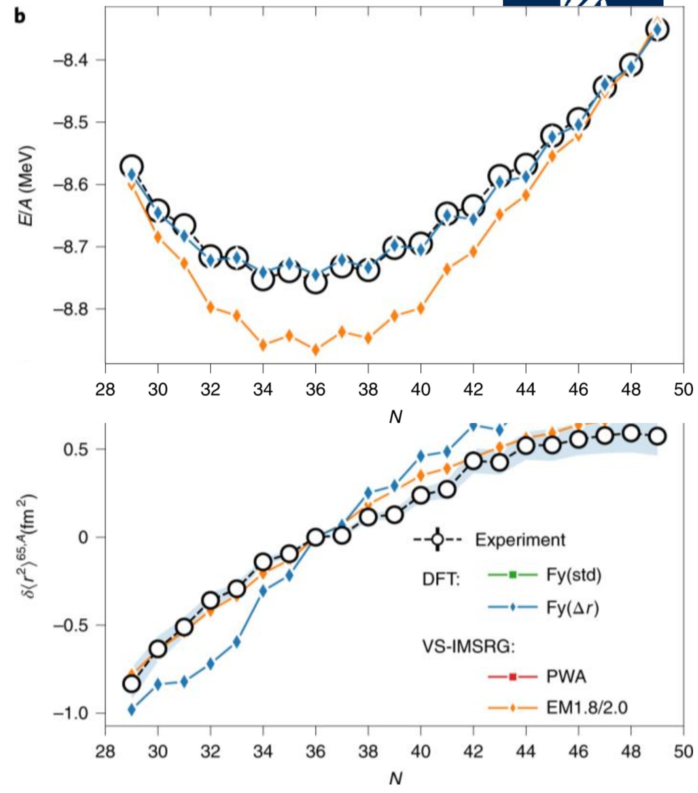


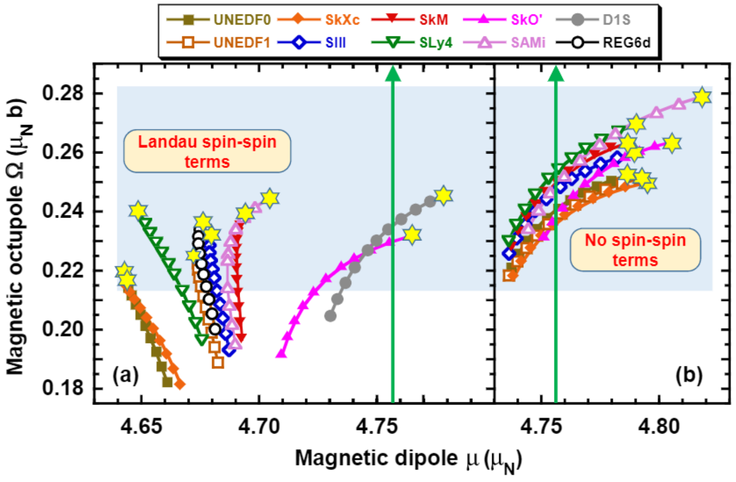
Looking ahead

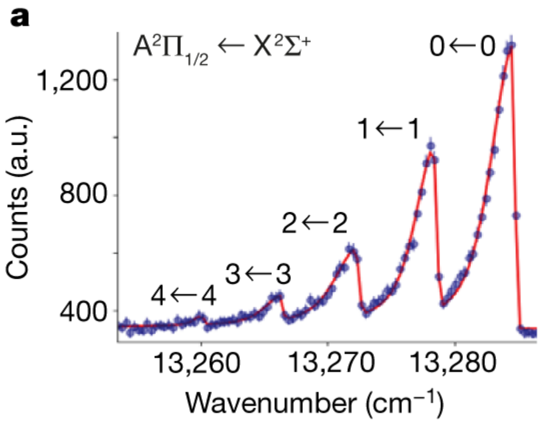
Radioactive probes for beyond standard model physics

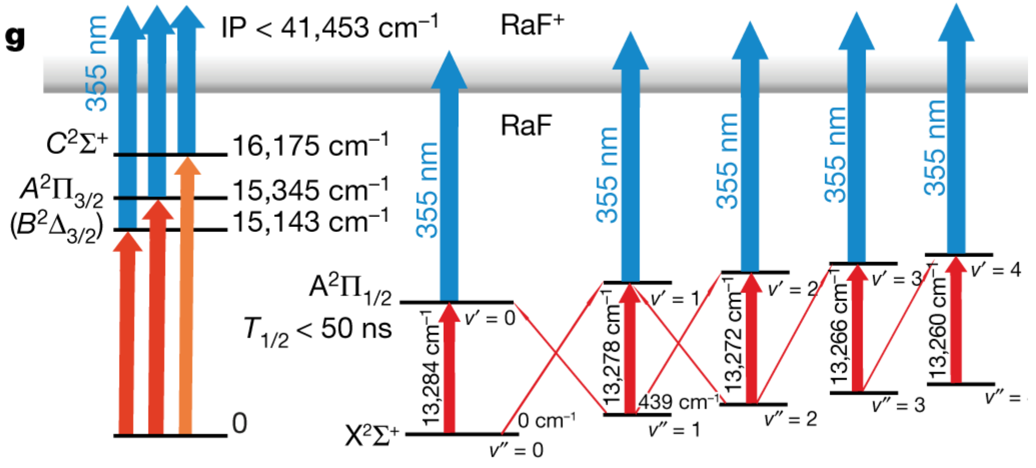
In summary

- (Optical) spectroscopy techniques keep marching on
  - Nuclear structure: charge radii of copper isotopes
  - Molecular structure and future BSM work: RaF molecules
  - High-precision spectroscopy for novel observables: scandium





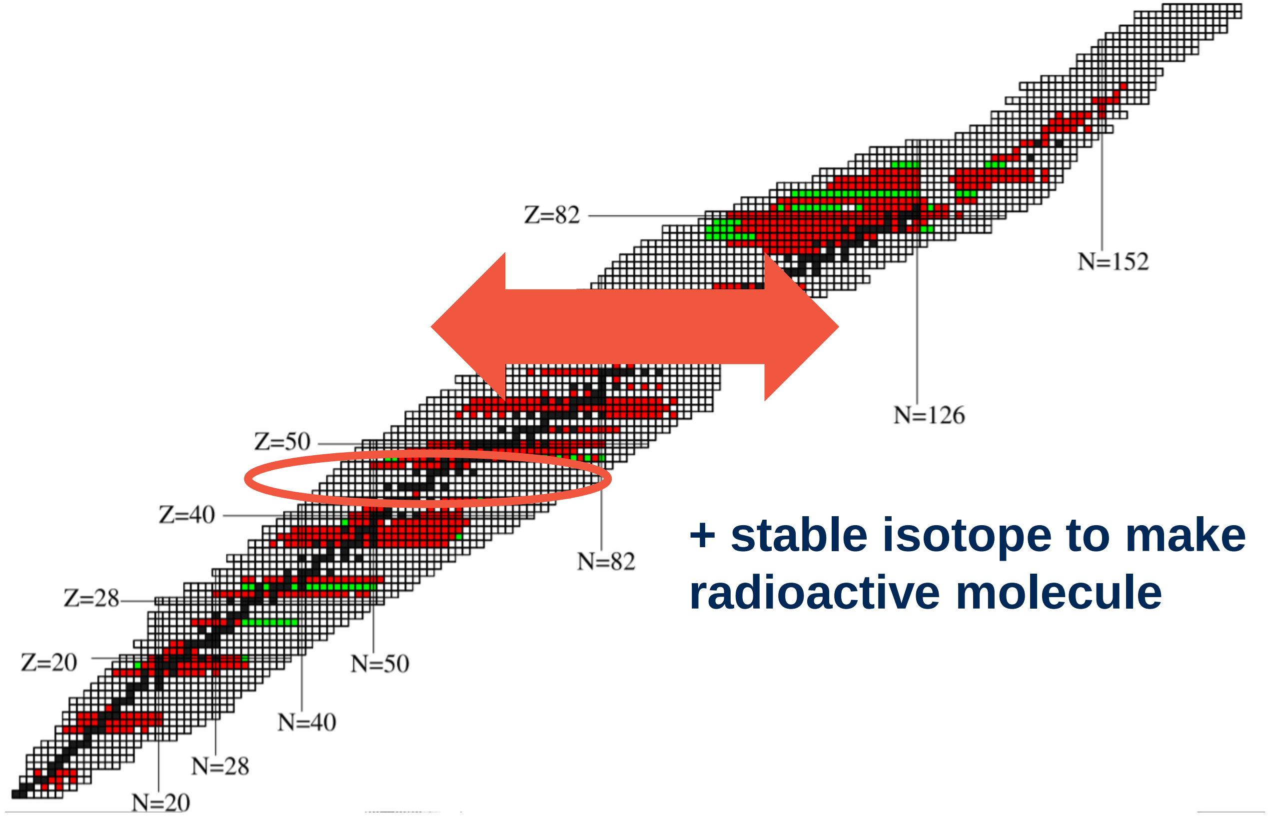




The nuclear chart from the point of view of optical spectroscopy

Instrumentation is improving across the board

- Efficiency frontier: the effort to push further and further from the valley of stability towards e.g. ^{78}Ni, ^{100}Sn, …
  - **Charge radii of neutron-rich copper**
- 'Accessibility' frontier: how can we reach isotopes that are hard to produce and/or have atomic structure complications
  - Key role for facilities like JYFL, NSCL/FRIB, RIKEN… Not featured in this seminar
- Precision frontier
  - Light elements
  - $\delta\langle r^4 \rangle$, hyperfine anomalies, **magnetic octupole**, electric hexadecapole moments …



+ stable isotope to make radioactive molecule

Copper charge radii as sensitive tests of state-of-the-art nuclear theory

nature physics

Letter | Open Access | Published: 13 April 2020

Measurement and microscopic description of odd–even staggering of charge radii of exotic copper isotopes

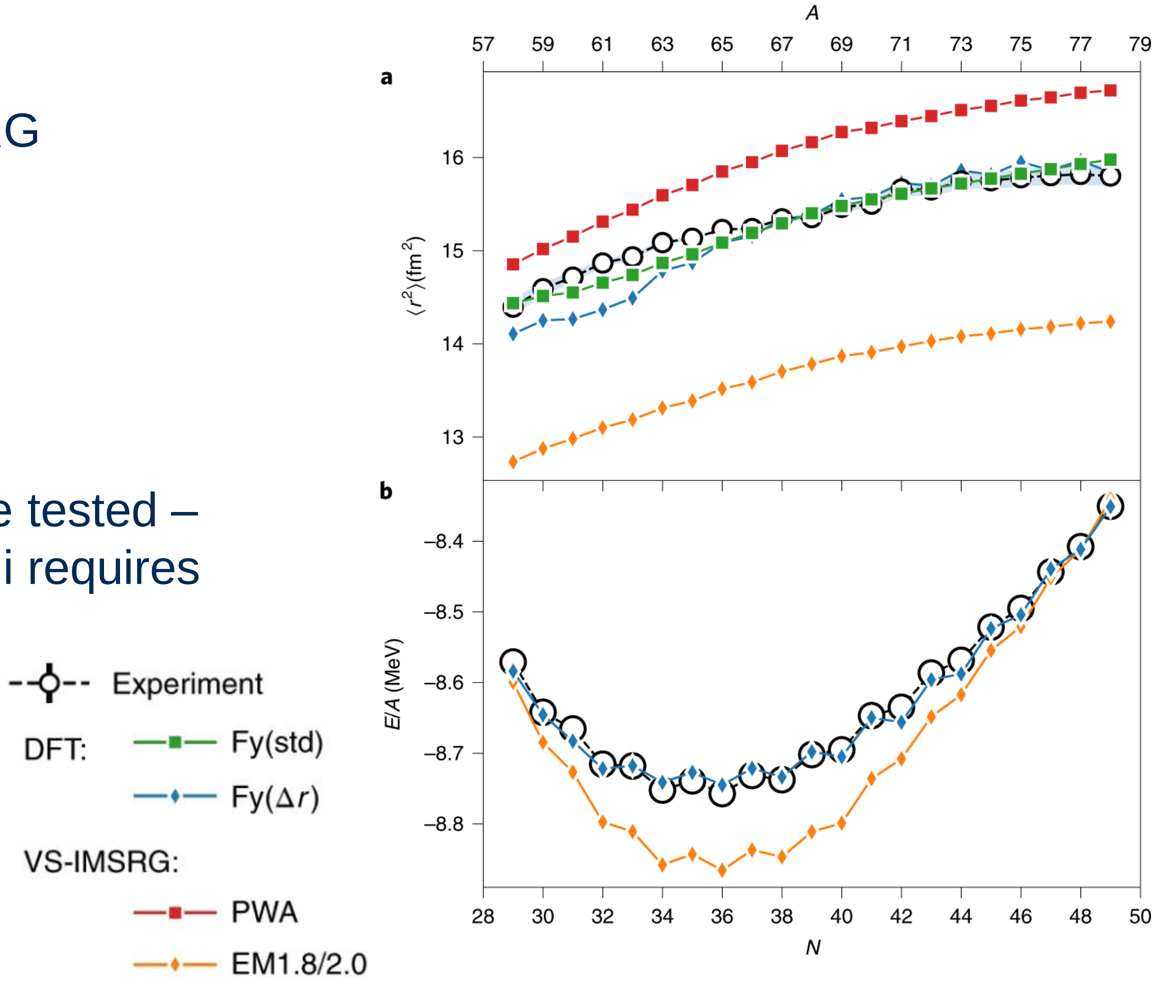
R. P. de Groote, J. Billowes, [...] X. F. Yang

Total binding energies

- good agreement with Fayans DFT and VS-IMSRG calculations

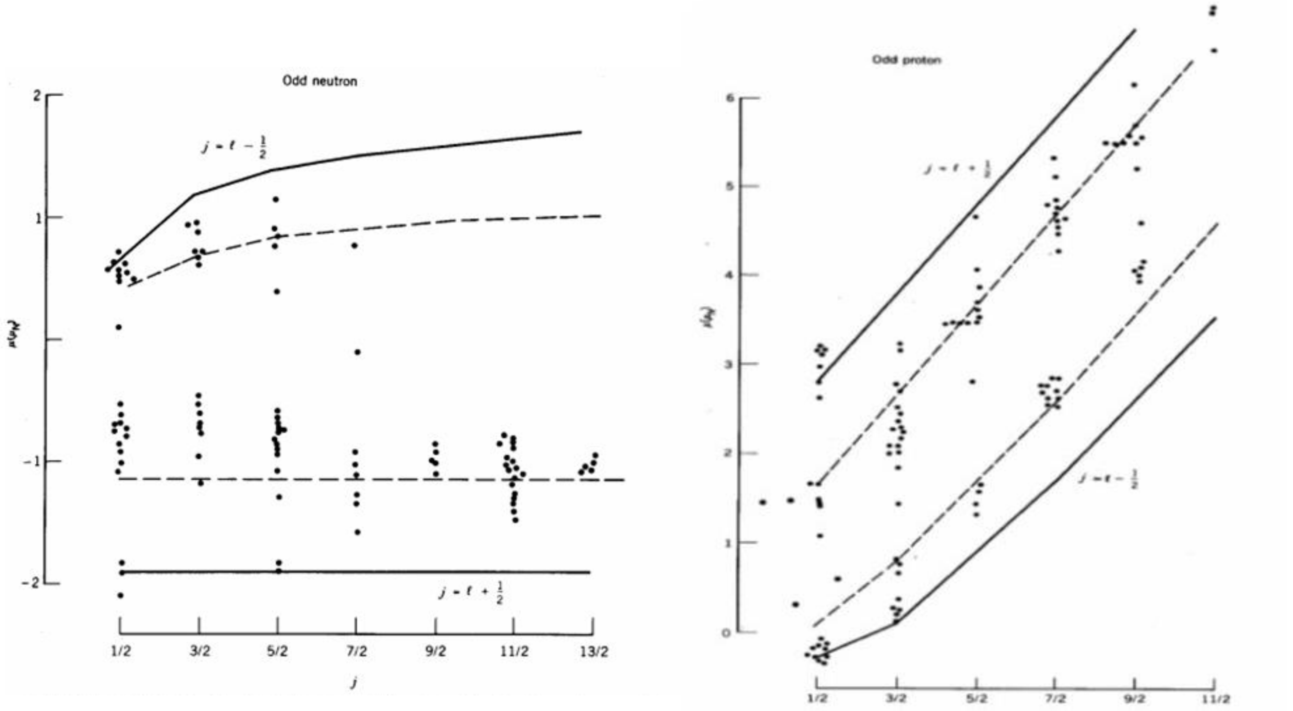
Total nuclear charge radii:

- good agreement for with Fayans DFT
- VS-IMSRG off the mark with both interactions we tested – matches prior observations that reproducing radii requires reproducing saturation density



Some textbook physics on magnetic dipoles…

- Pen-and-paper prediction for magnetic dipole moments: Schmidt lines
- By now, hundreds of experimental moments have been measured
- All experimental data points lie close to these lines assuming some spin g-factor quenching
  - Configuration mixing, MEC, …
- Magnetic moment is a fingerprint of the nuclear configuration
- Comparison to more advanced models: investigate purity of configuration, shell evolution, …



$$\langle\mu\rangle_{j=l+1/2} = \mu_{\rm N}\left[g_l(j-\tfrac{1}{2})+\tfrac{1}{2}g_s\right]$$

$$\langle\mu\rangle_{j=l-1/2} = \mu_{\rm N}\left[g_l\frac{j(j+\frac{3}{2})}{(j+1)} - \frac{g_s}{2}\frac{j}{(j+1)}\right]$$