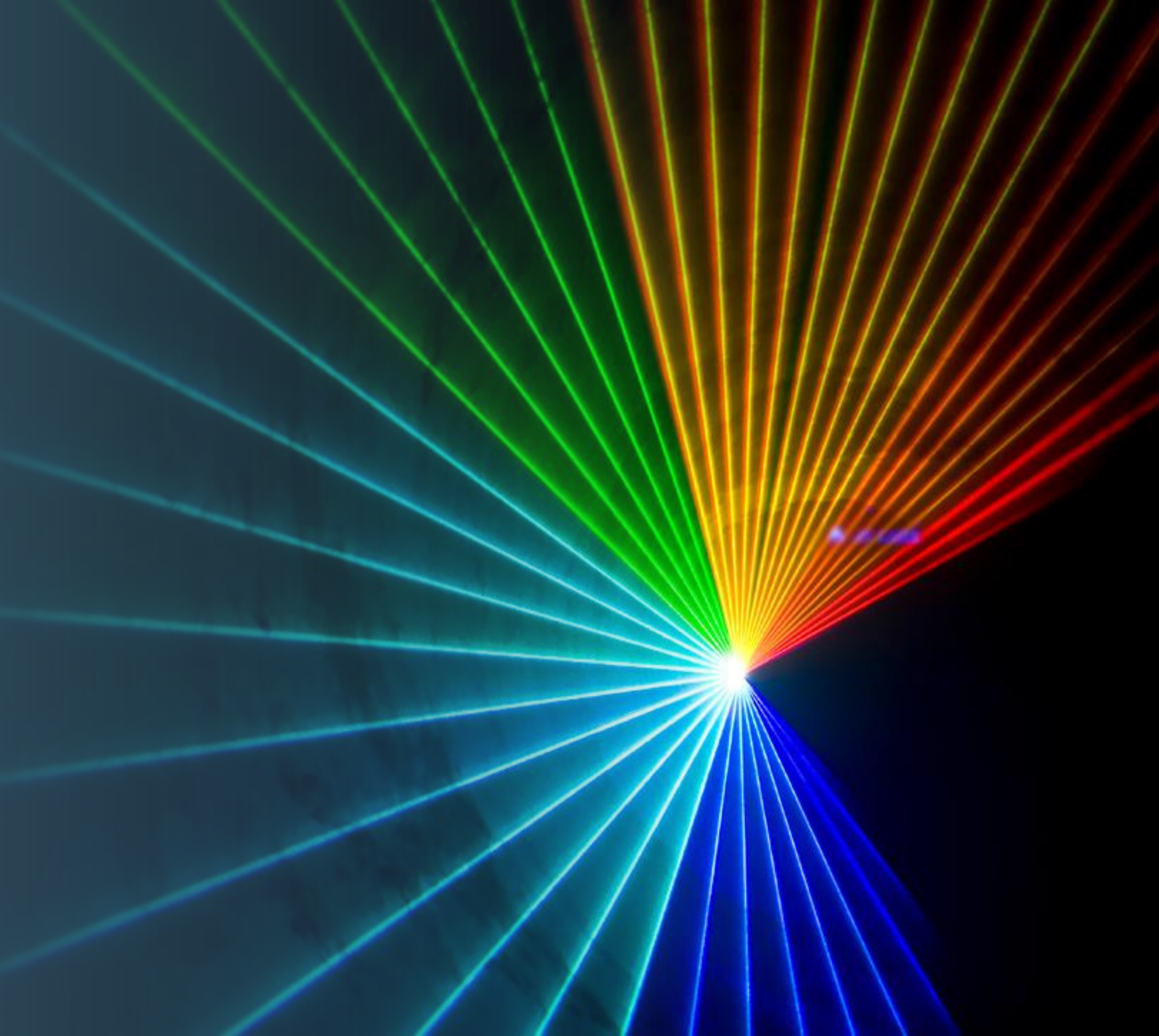
Laser spectroscopy of radioactive isotopes

Recent highlights and trends in the field

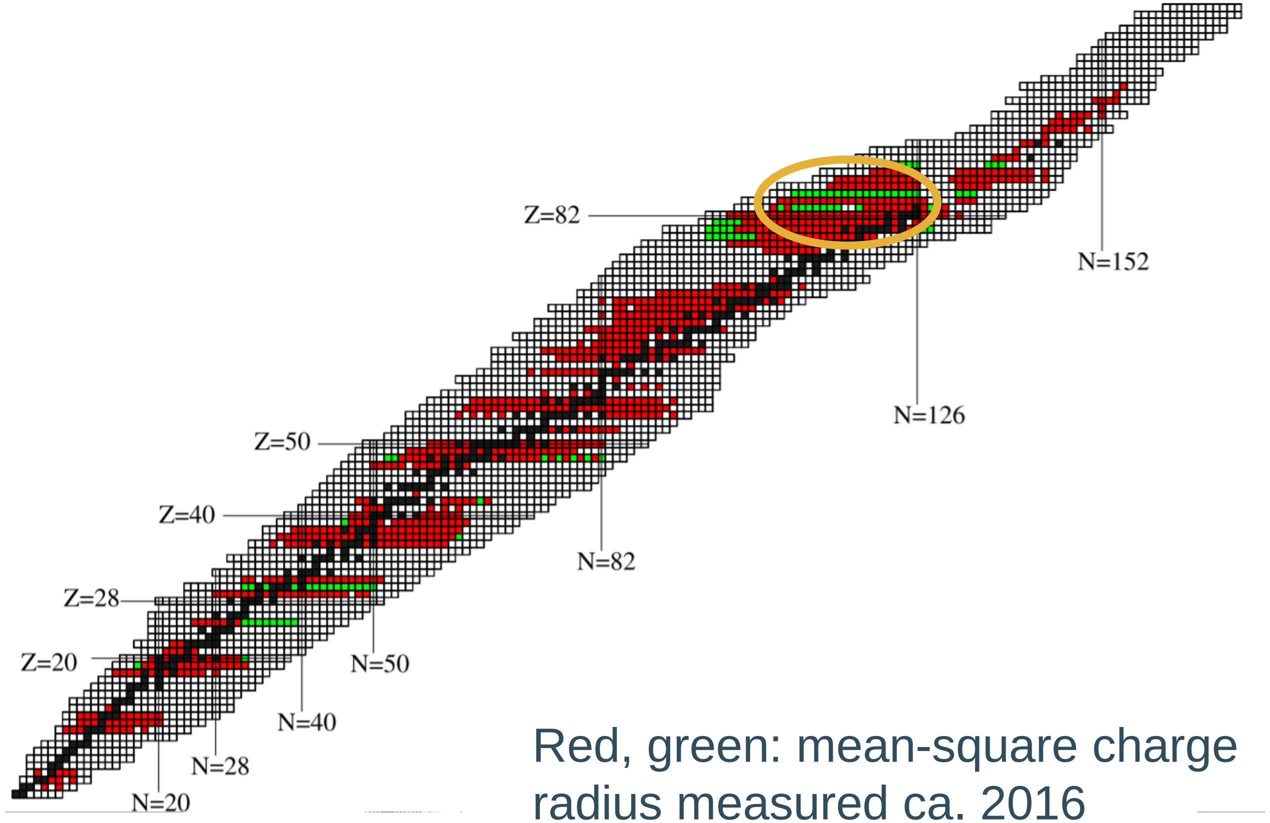


Disclaimer

- Overview of laser spectroscopy activities … an impossible task.
- I have picked some of the most recent results from different labs around the world.
- Many great results have been omitted!
- I hope they showcase the diversity of the science, the ever-expanding reach of the techniques, and the steady technical progress that is made worldwide
- (Even if I have left out the measurements closest to your heart, sorry! Let's discuss it in the question time after the talk!)

Outline

- **Exotic (shape) phenomena**
  - In-source laser spectroscopy of Hg, Bi
- Superheavy elements
- Test of nuclear interactions near shell closures
  - ^{52}K, 52Ca (CRIS&COLLAPS, ISOLDE)
  - ^{36}Ca (BECOLA, NSCL)
  - In-source laser spectroscopy of Ag
- Precision measurements
  - Trapped-atom and ion experiments
- Beyond atomic spectroscopy
  - First spectroscopy of radioactive molecules



Red, green: mean-square charge radius measured ca. 2016

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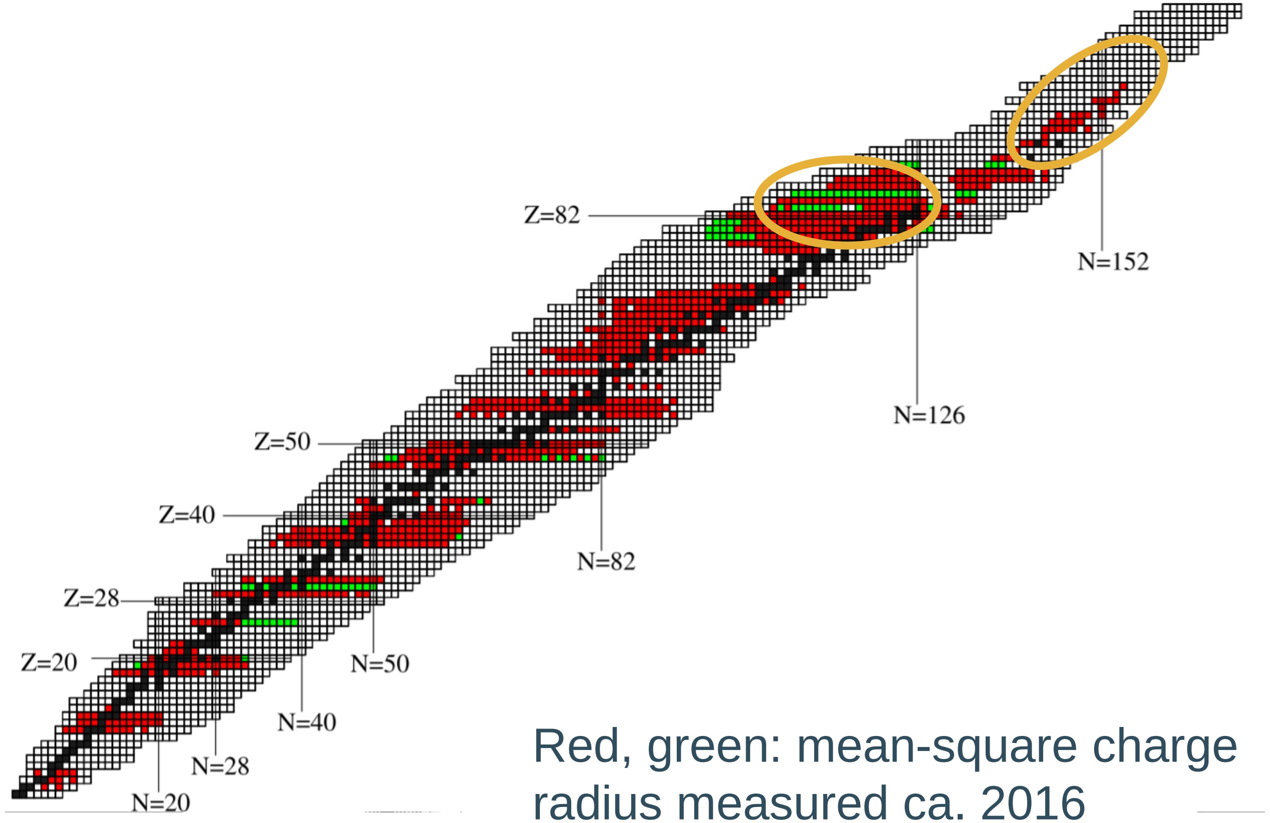

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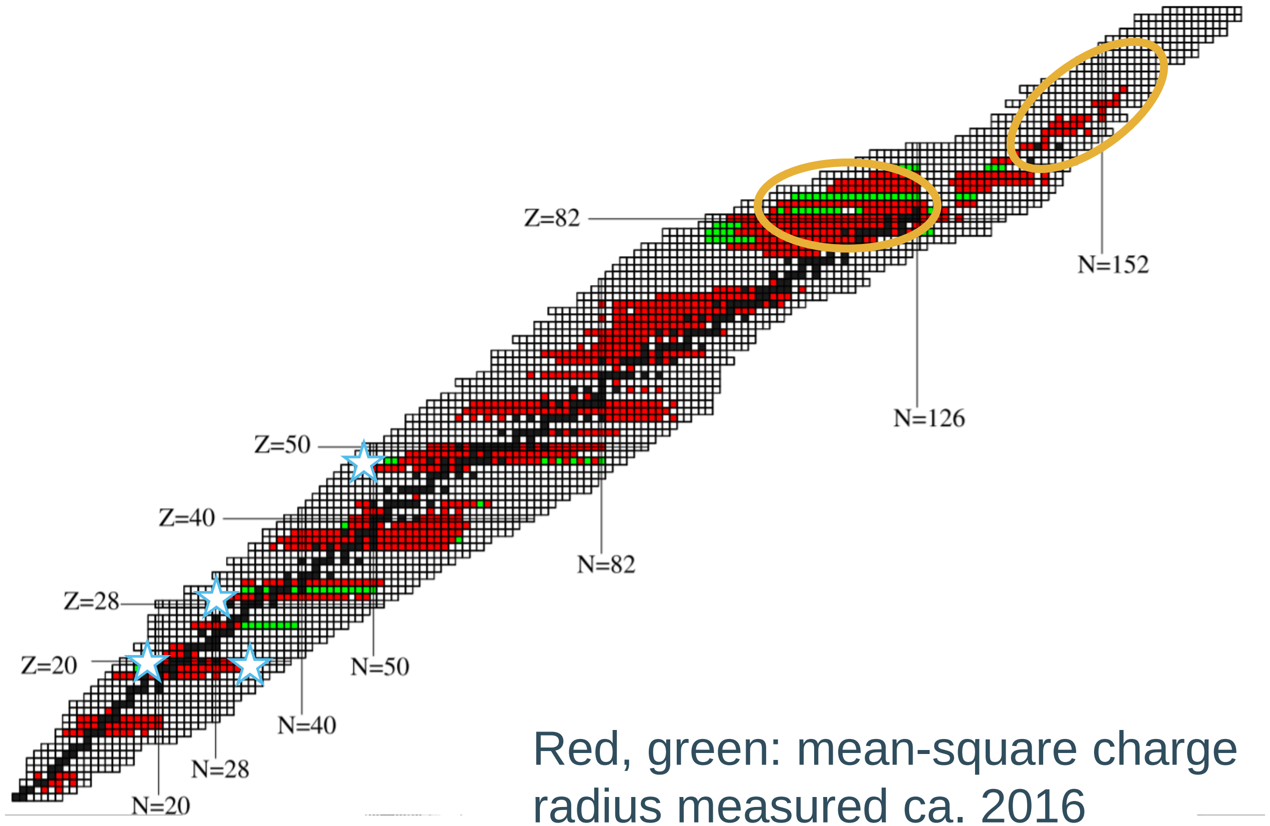

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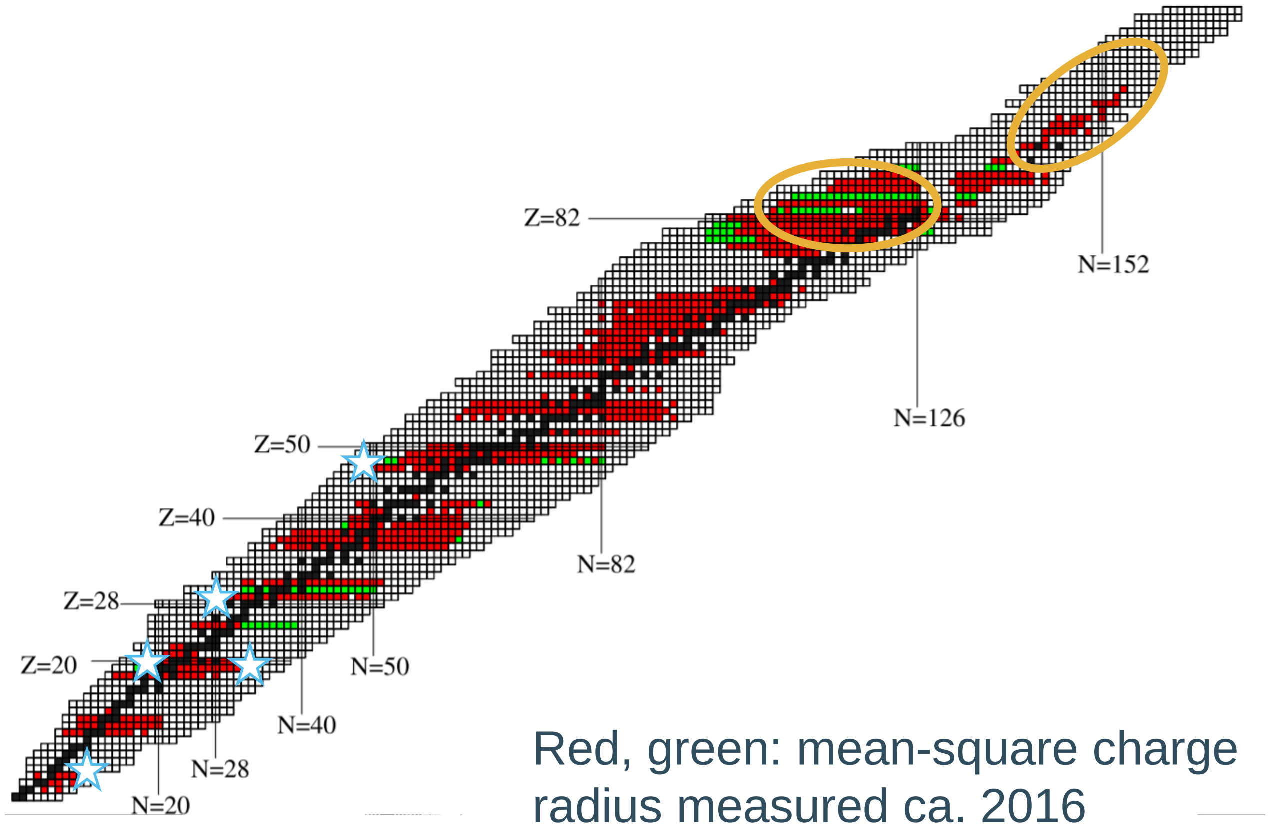

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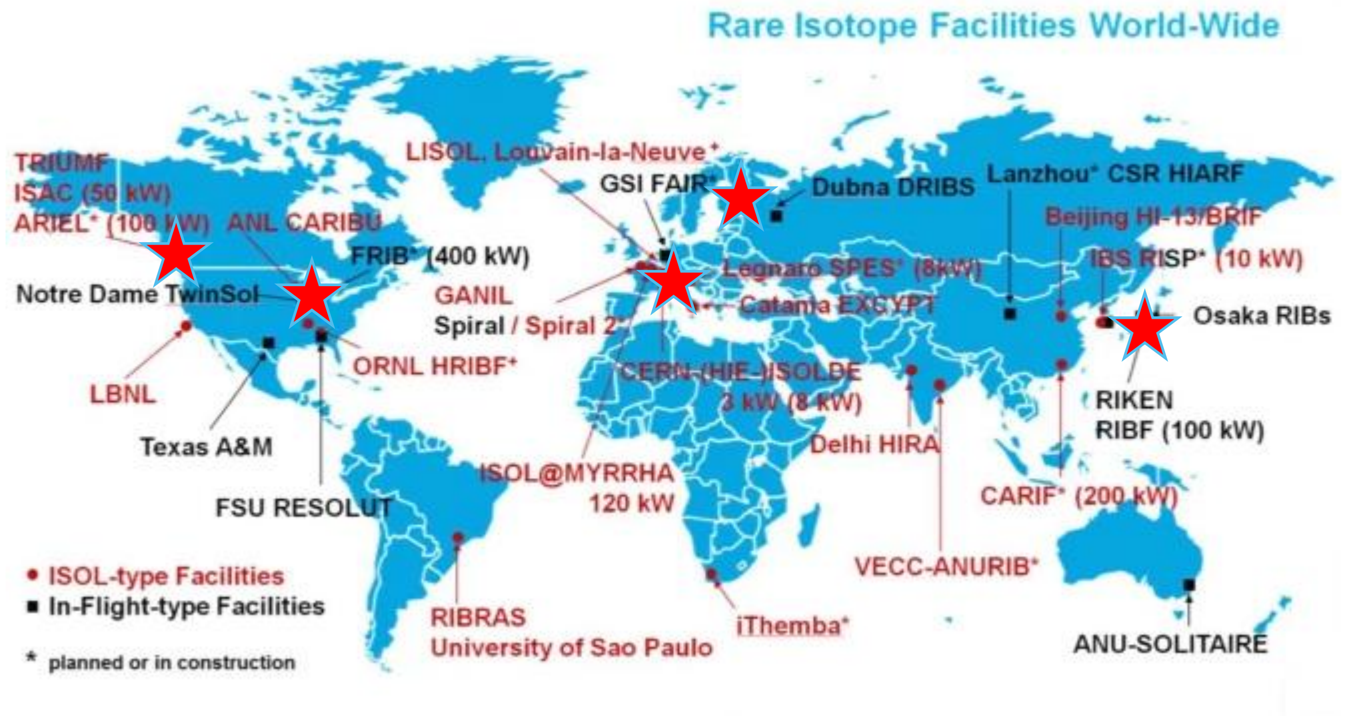

Each facility brings its own challenges and advantages.

Steady improvements in technology and methodology have allowed for this vigorous exploration of the nuclear chart!

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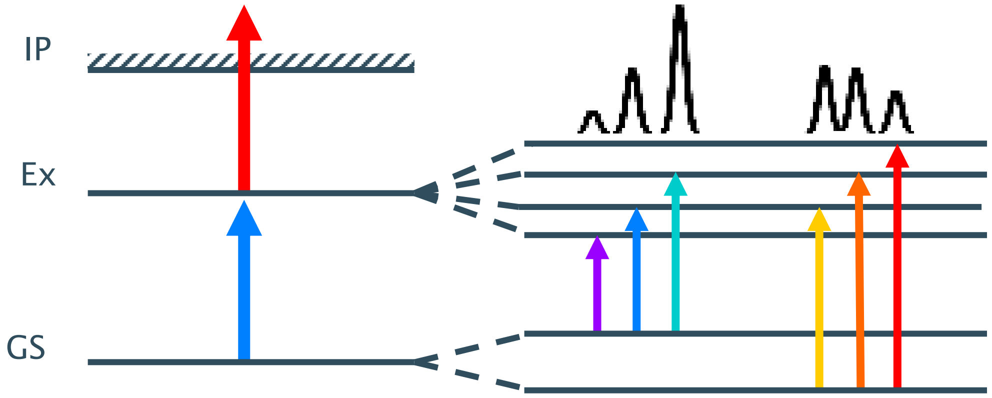
- In-source laser spectroscopy
- Collinear laser spectroscopy
- Methods tailed to specific cases

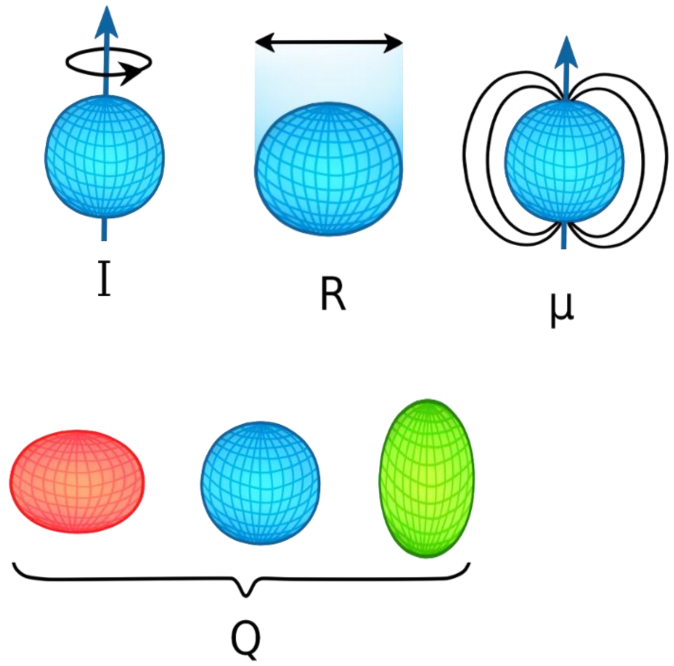
Each technique brings its own challenges and advantages.

Steady improvements in technology and methodology have allowed for this vigorous exploration of the nuclear chart!

Laser spectroscopy: atom and nucleus

- Electromagnetic interaction between orbiting electrons and the nucleus
  - Isotopes have a **finite size** leading to isotope shifts
    ➔ nuclear size
  - Orbiting charges and intrinsic nucleon moments lead to **nuclear magnetic moments**
    ➔ valence configuration, wf purity
  - Charge can assume non-spherical charge distribution, and thus **electric moments**
    ➔ deformation, shape, collectivity





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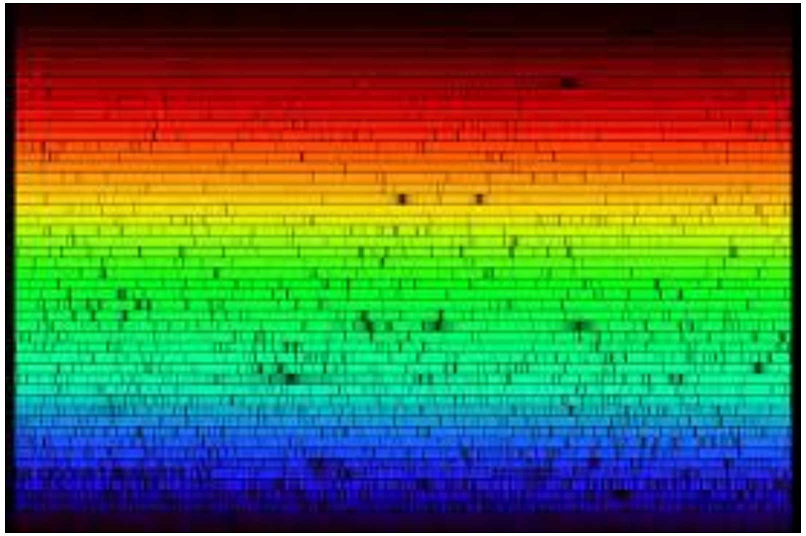
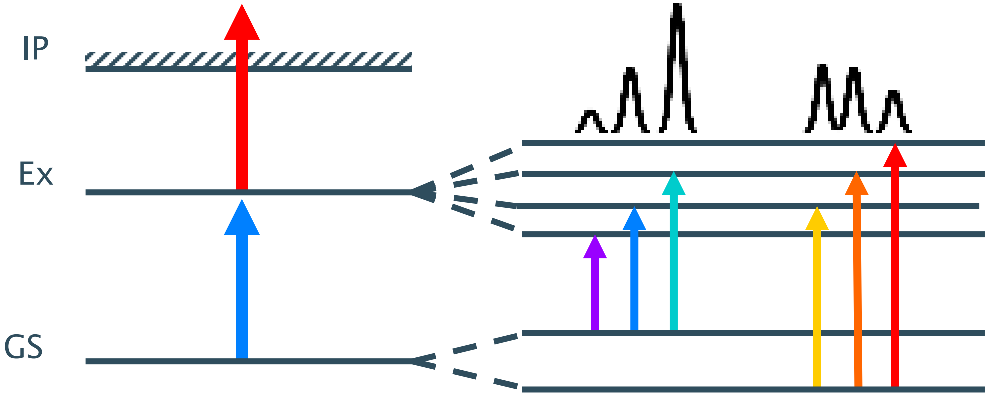
Some orders of magnitude…





- Laser spectroscopy: excite the electron, monitor fluorescence/ion signal/… as you change the laser wavelength

- Transition wavelengths: 10-2000 nm
  Practically useful range: 200-1000 nm (with gaps and caveats)

- Isotope shifts, hyperfine splitting : MHz – many GHz
- Transition @400 nm = 750 THz

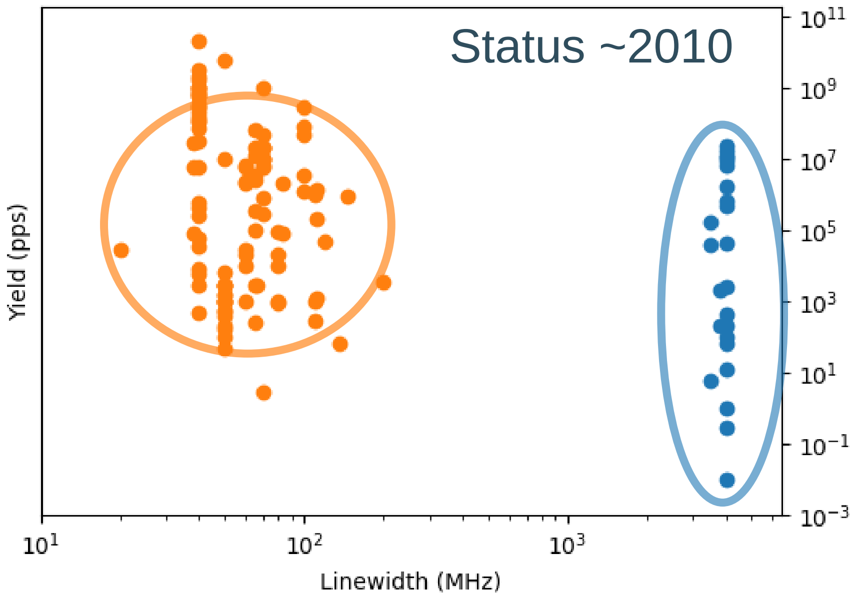
- Laser spectroscopy inherits the *richness* of atomic structure, but any statements on efficiency, applicability, … are thus *element-specific.*

Laser spectroscopy approaches

- In-source laser spectroscopy
  - Hot cavity
  - Gas cell

- Collinear laser spectroscopy
  - Fluorescence detection
  - Resonance ionization
  - Beta-NMR/collisional ionization/state-selective charge exchange/…

- Methods tailed to specific cases
  - Spectroscopy on trapped ion/atom
  - Techniques superseded by more modern ones… Worth reading up on!



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Laser spectroscopy approaches

- In-source laser spectroscopy
  - Hot cavity
  - Gas cell

Very efficient, low production rates
Low resolution, limited element choice

- Collinear laser spectroscopy
  - Fluorescence detection
  - Resonance ionization
  - Beta-NMR/collisional ionization/state-selective charge exchange/…

A bit less efficient
High resolution, wide element choice

- Methods tailed to specific cases
  - Spectroscopy on trapped ion/atom
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More niche, specific cases/applications

Laser spectroscopy approaches

- In-source laser spectroscopy
  - Hot cavity
  - Gas cell

- Collinear laser spectroscopy
  - Fluorescence detection
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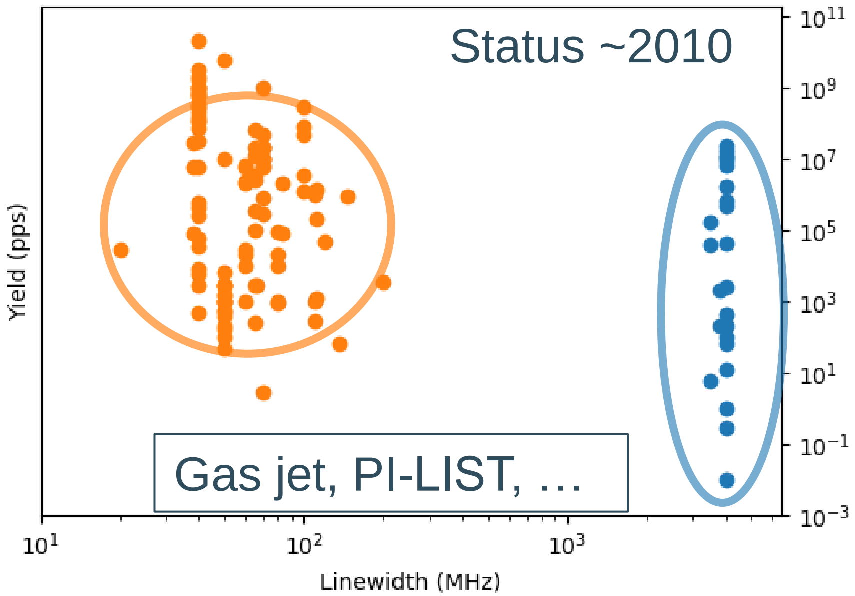

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What is possible?

Typically,

- Requirement: quality beam, low energy spread

- Production rates: as low as 0,01/s (in-source), 10/s (collinear (RIS))

- Resolution: as low as 10 MHz / precision ~ 100 kHz

- Most elements (exceptions: reactive non-metals – top-right periodic table)

But: not always, at the same, time for every element!

Depends on beam purity, atomic structure, …

Exotic (shape) phenomena

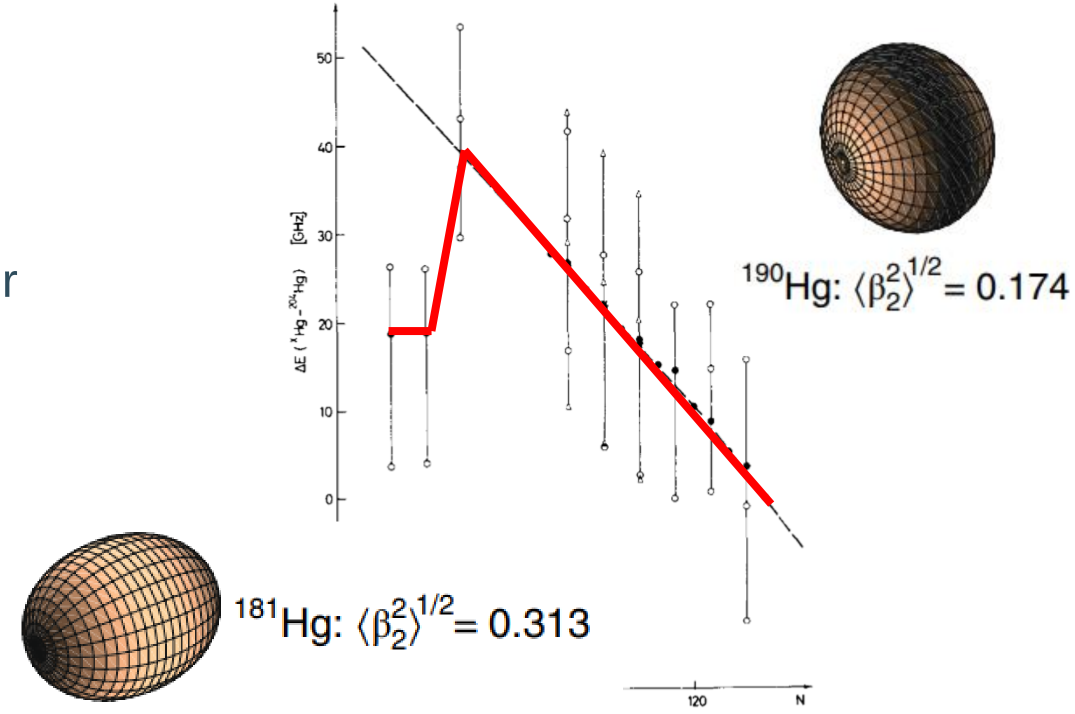
In-source laser spectroscopy

Exotic (shape) phenomena

- Exploration of nuclear structure far from stability sometimes brings unexpected phenomena

- Classic example: shapes in the Hg chain
  - Strong prolate deformation close to magic number Z=82!?
  - Sudden change in shape with single nucleon removal

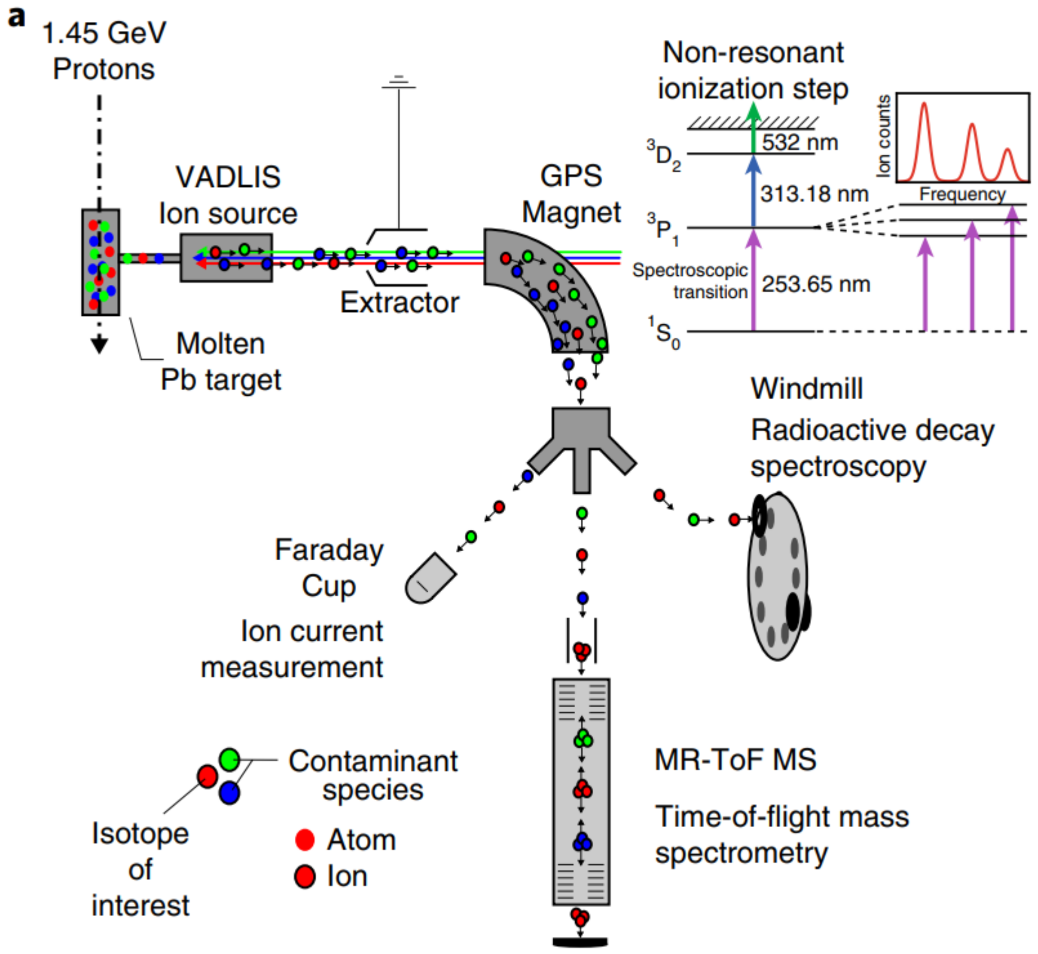
  - 40+ years of developments culminating in…



Bonn, J., Huber, G., Kluge, H.-J., Kugler, L. & Otten, E. Sudden change in the nuclear charge distribution of very light mercury isotopes. Phys. Lett. B 38, 308–311 (1972).

Technique: in-source laser spectroscopy

- Radioactive isotopes produced through a nuclear reaction
- Target is heated to very high temperature (2000+ K) to enable diffusion towards the ion source region
- Multiple laser beams overlapped with atoms to stepwise excite and ionize
  - Spectroscopy in the ion source
  - Efficient! As high as 50%, typically a few %



Marsh, B.A., Day Goodacre, T., Sels, S. *et al.* Characterization of the shape-staggering effect in mercury nuclei. *Nature Phys* **14,** 1163–1167 (2018)

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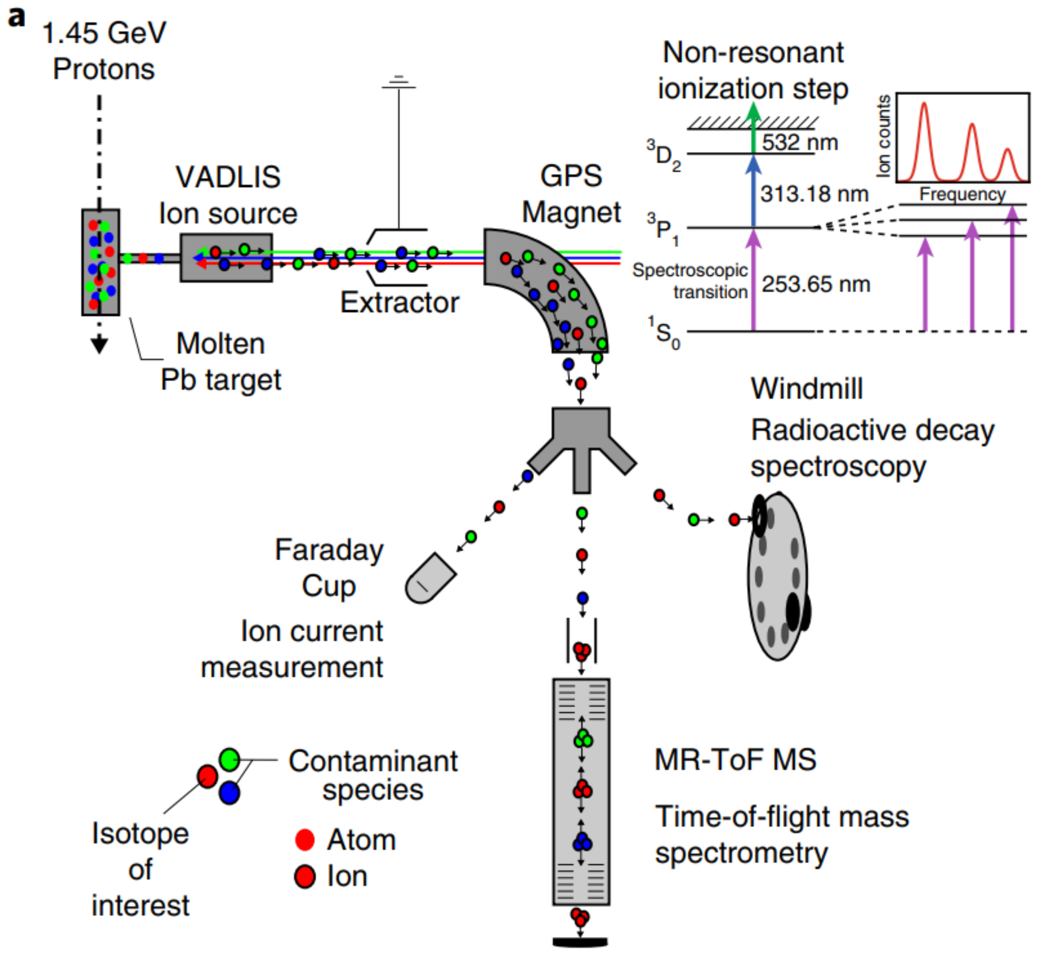
Technique: in-source laser spectroscopy

- Linewidth: order of GHz
  - Source of broadening: Doppler effect due to high temperature
  - Spectral properties of the laser are matched to this linewidth

- Ions transported to one of several possible detection stations
  - Decay spectroscopy: tag on characteristic radiation
  - Mass spectrometry: single out one isotope from other isobars using its mass

  - **FLEXIBILITY!** Tailor the detection to the isotope and beam at hand



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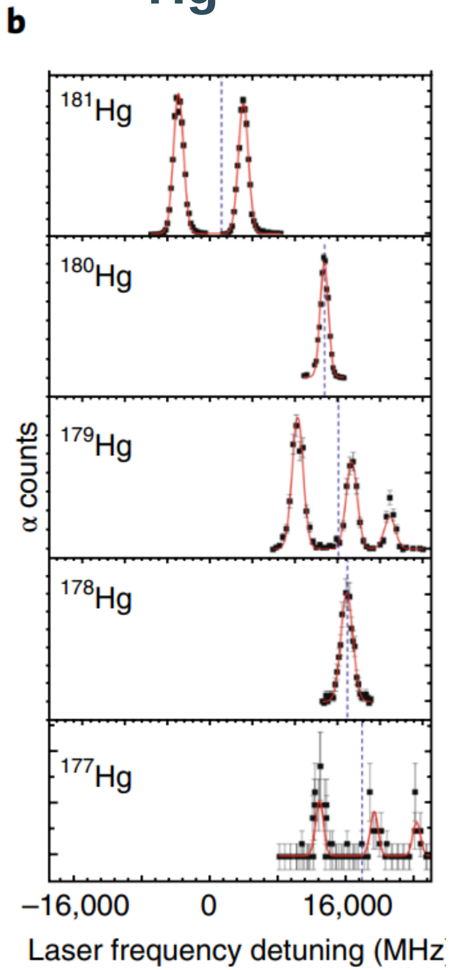
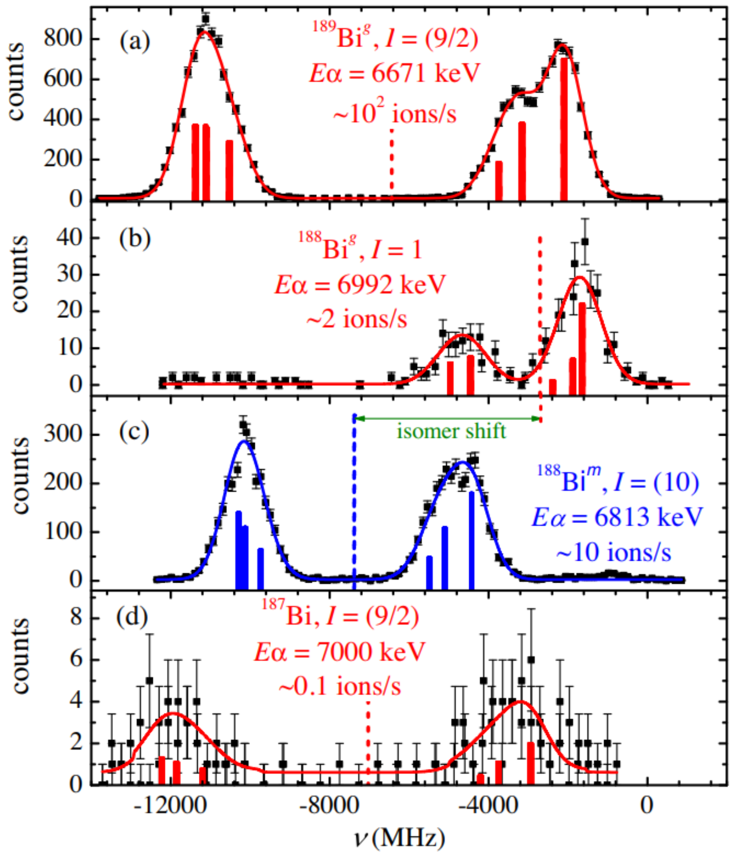
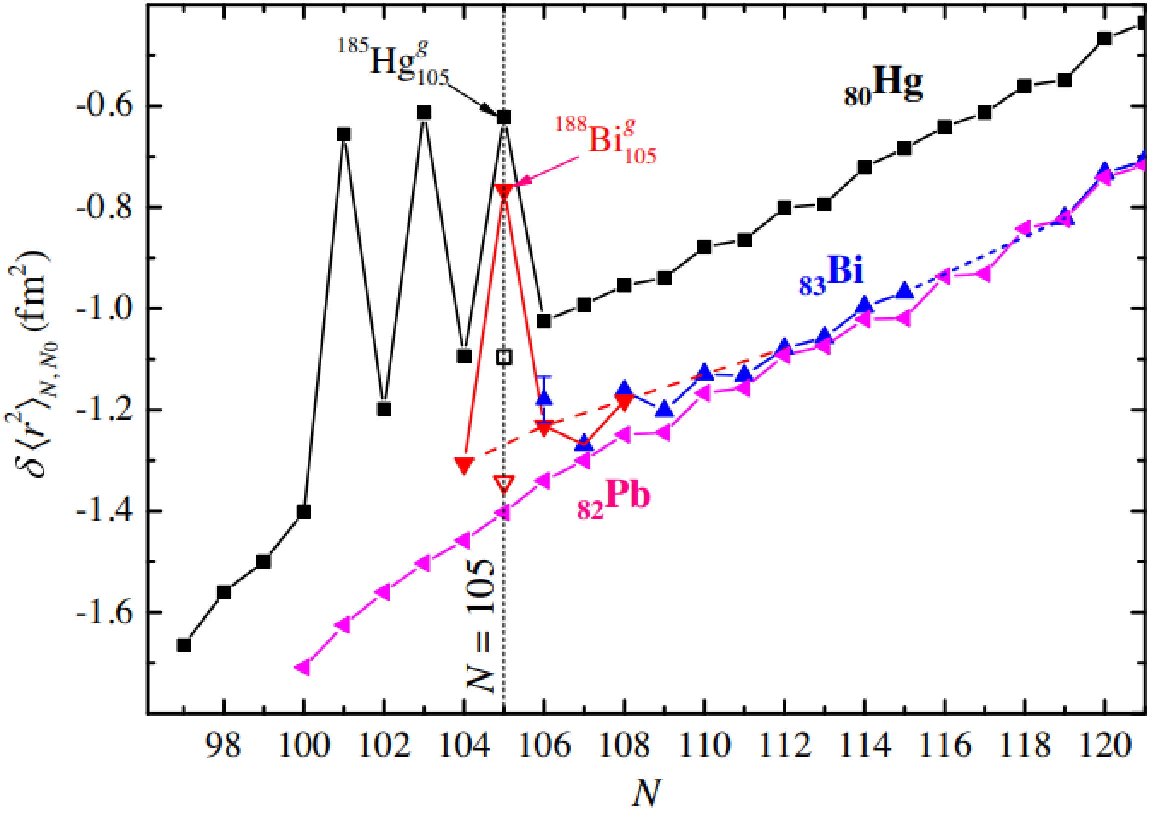
Results

Hg



Bi





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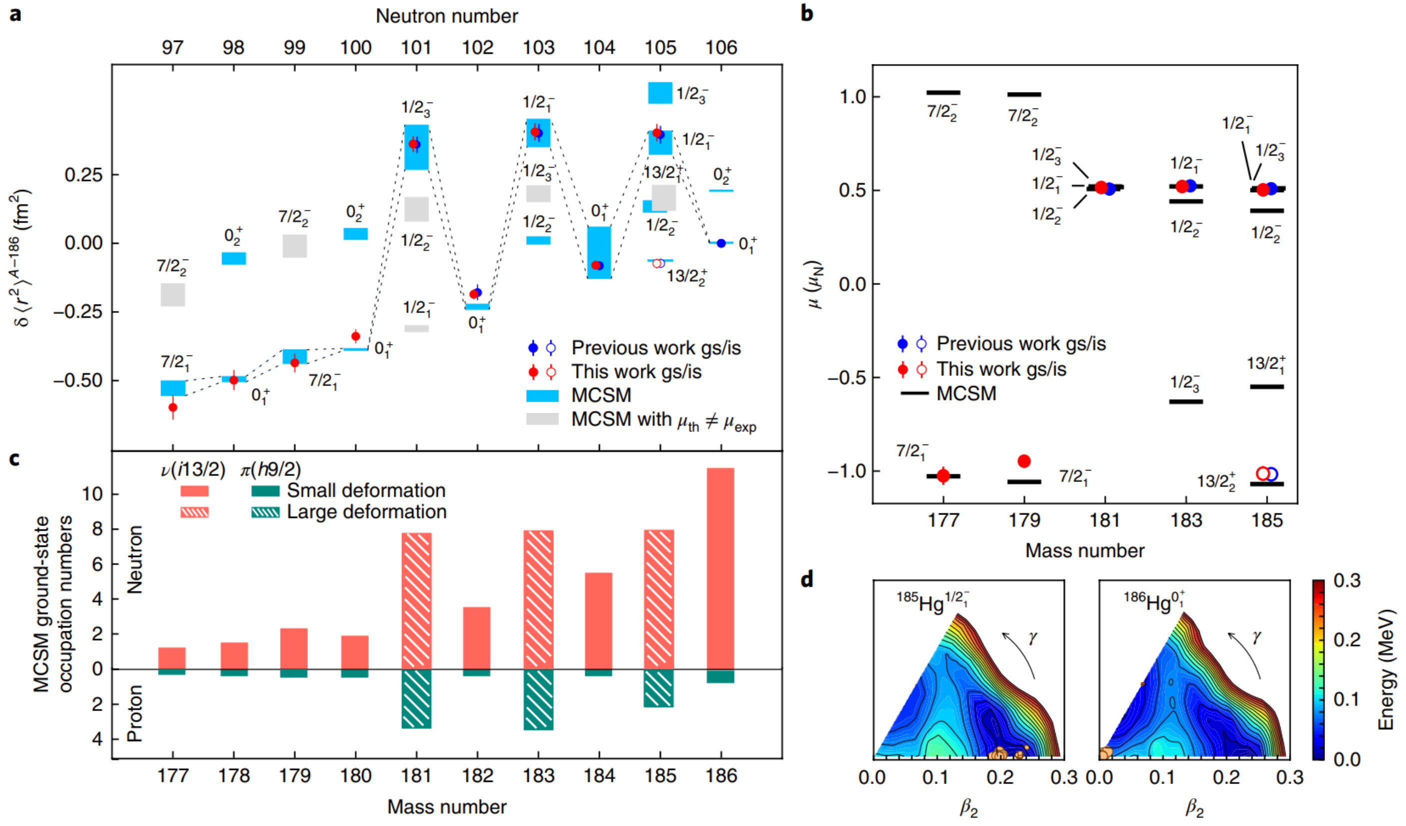

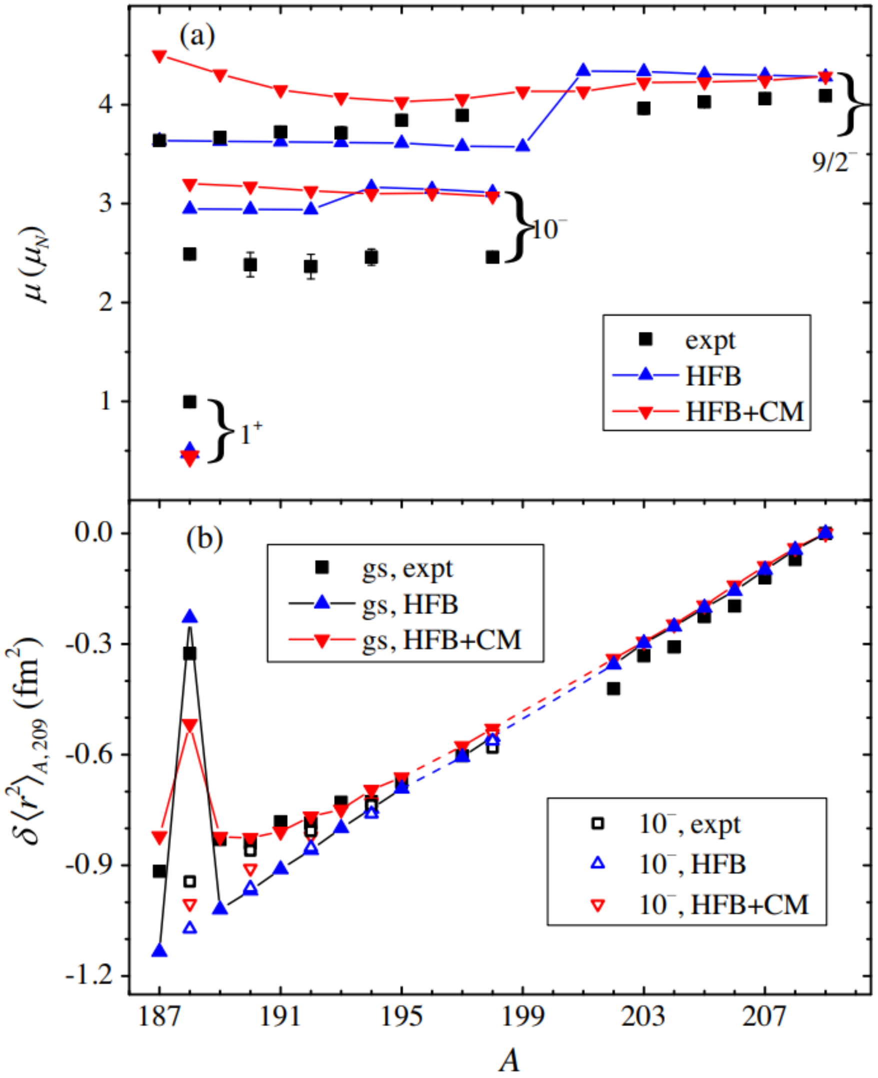

- Significant challenge for nuclear theory
- Magnetic moments are key to pin down nuclear configuration to aid the interpretation!

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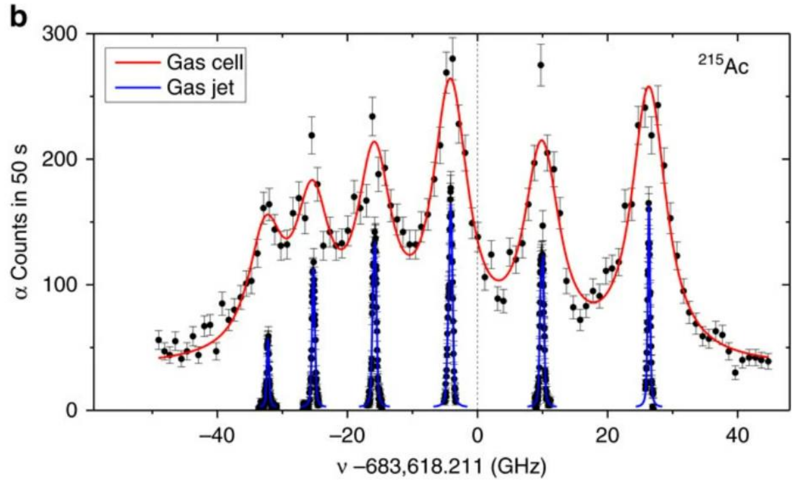
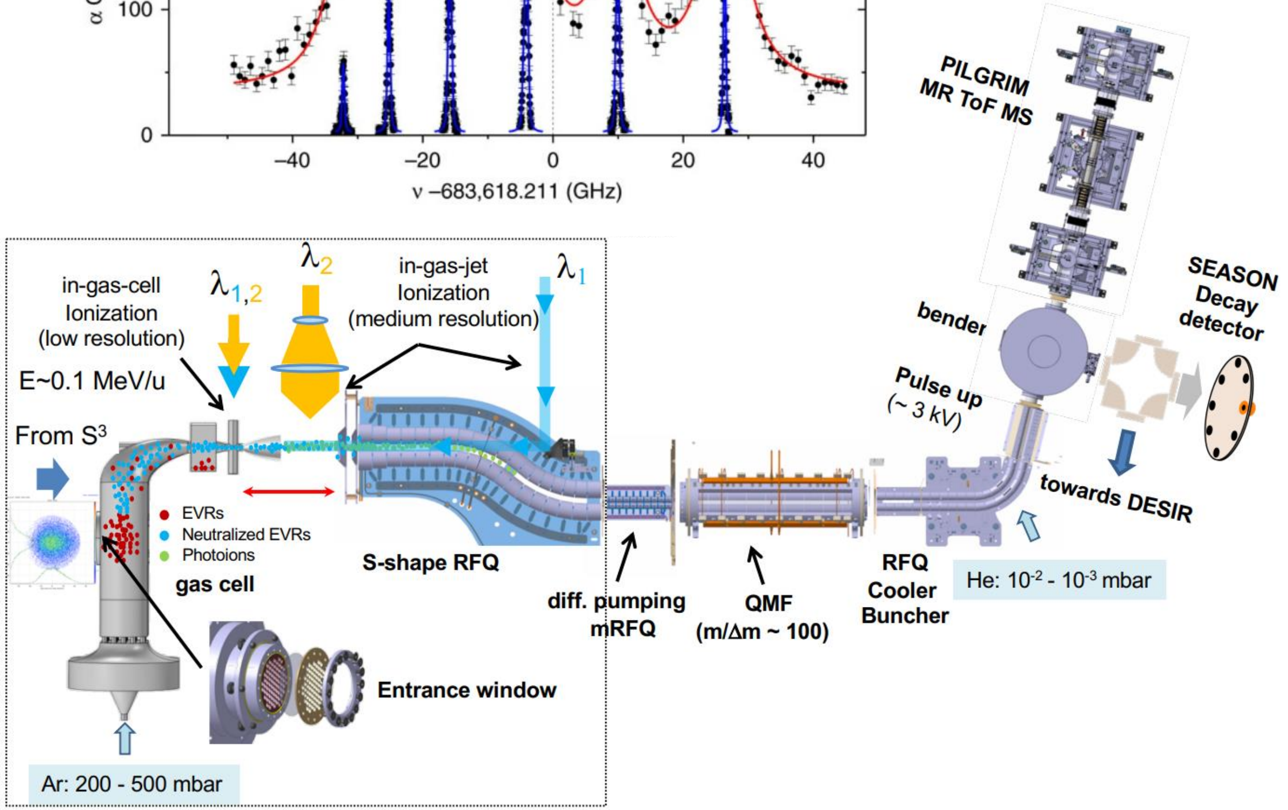
Wider context

- In-source laser spectroscopy provides the **ultimate sensitivity, at the cost of resolution**
  - **Exception:** in-gas jet laser spectroscopy promises high resolution with in-source efficiency
  - Take ions out of the high-pressure environment, cool through supersonic expansion

- Combination with ultra-selective and efficient detection techniques provides **access to the most exotic isotopes**
  - *Make every ion count!*





Ferrer, et al. Towards high-resolution laser ionization spectroscopy of the heaviest elements in supersonic gas jet expansion. Nat Commun 8, 14520 (2017)

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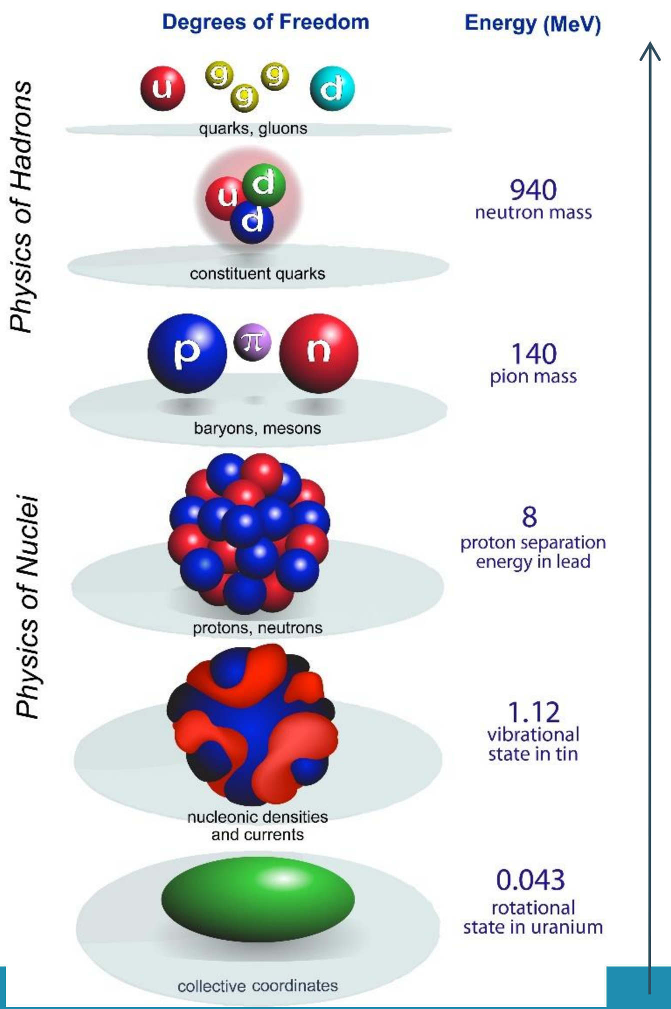
Development of nuclear interactions

Pushing methods to short lifetimes and low yields with collinear methods

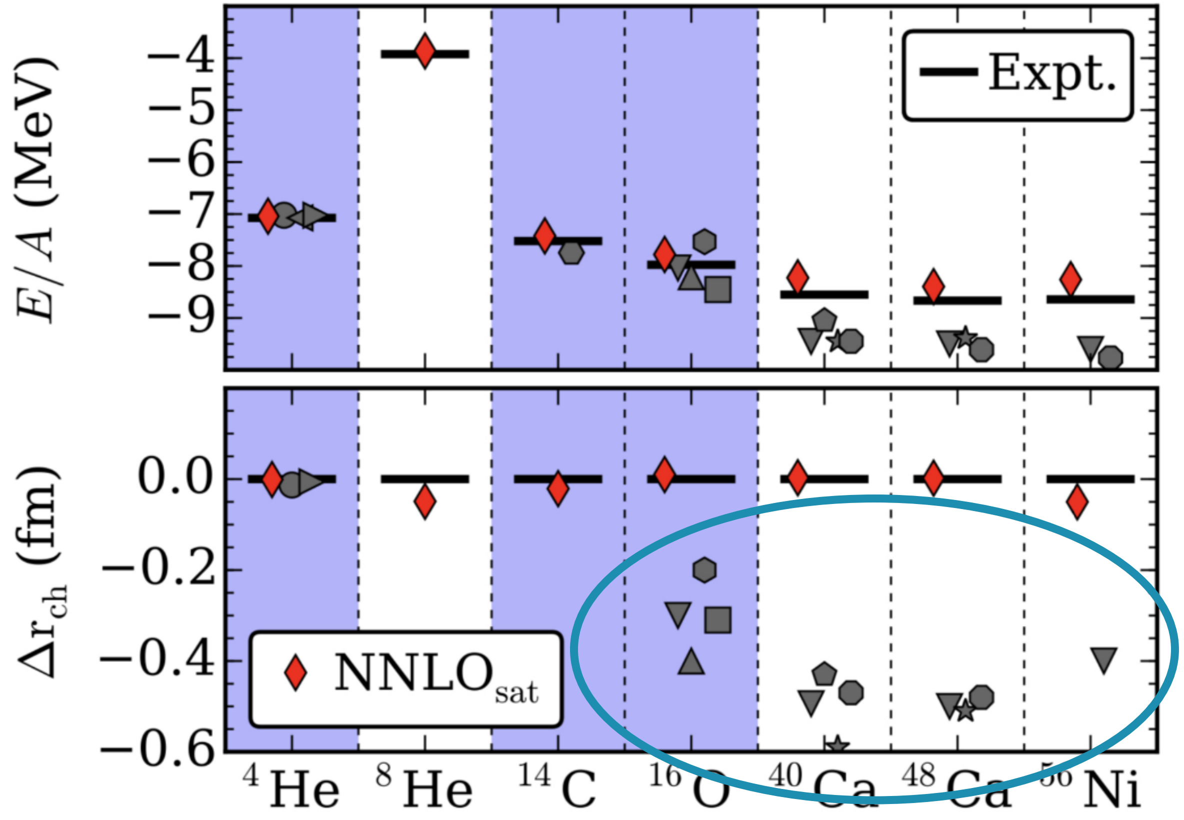
Scientific case



Energy or resolution

Chiral Effective Field Theory

A. Ekström, PRC **91**, 051301(R) 2015



Constrained to provide an accurate description of the A=2 and A=3 systems.

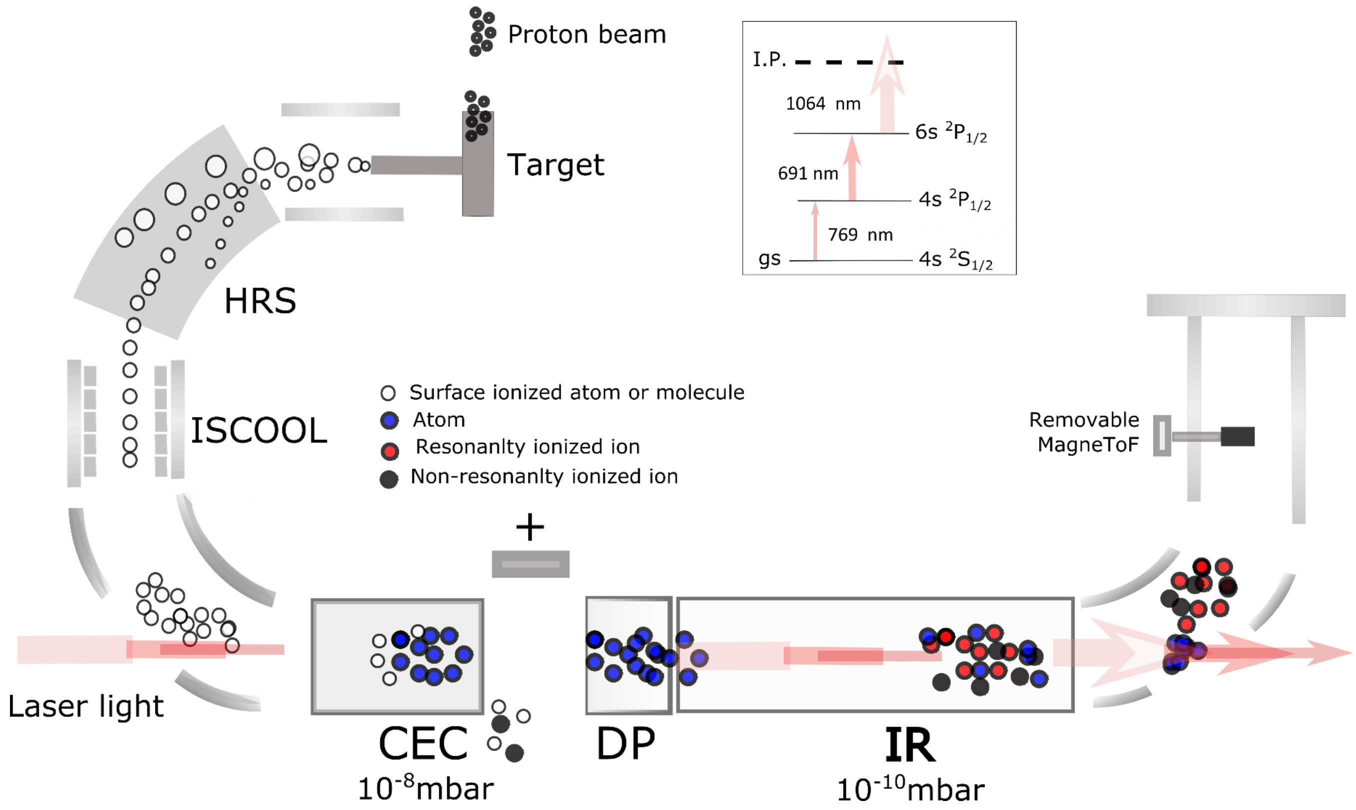
Predictions for nuclear structure and dynamic (A>3).

Why is there the mismatch?
Why does NNLO$_{sat}$ work?

An open problem…

Fig.: Bertsch, Dean, Nazarewicz, SciDAC review (2007)

Study of the Ca region
Spectroscopy of K



CRIS:

- Collinear = doppler-free
- RIS = versatile, efficient

Challenges:

- Large isobaric contamination
- Low yields
- Changing proton current
  → changing production and background

At mass 52:

- **>10^6 pps contamination**
- 300 pps ^{52}K

- Beta-detection: remove stable contaminants

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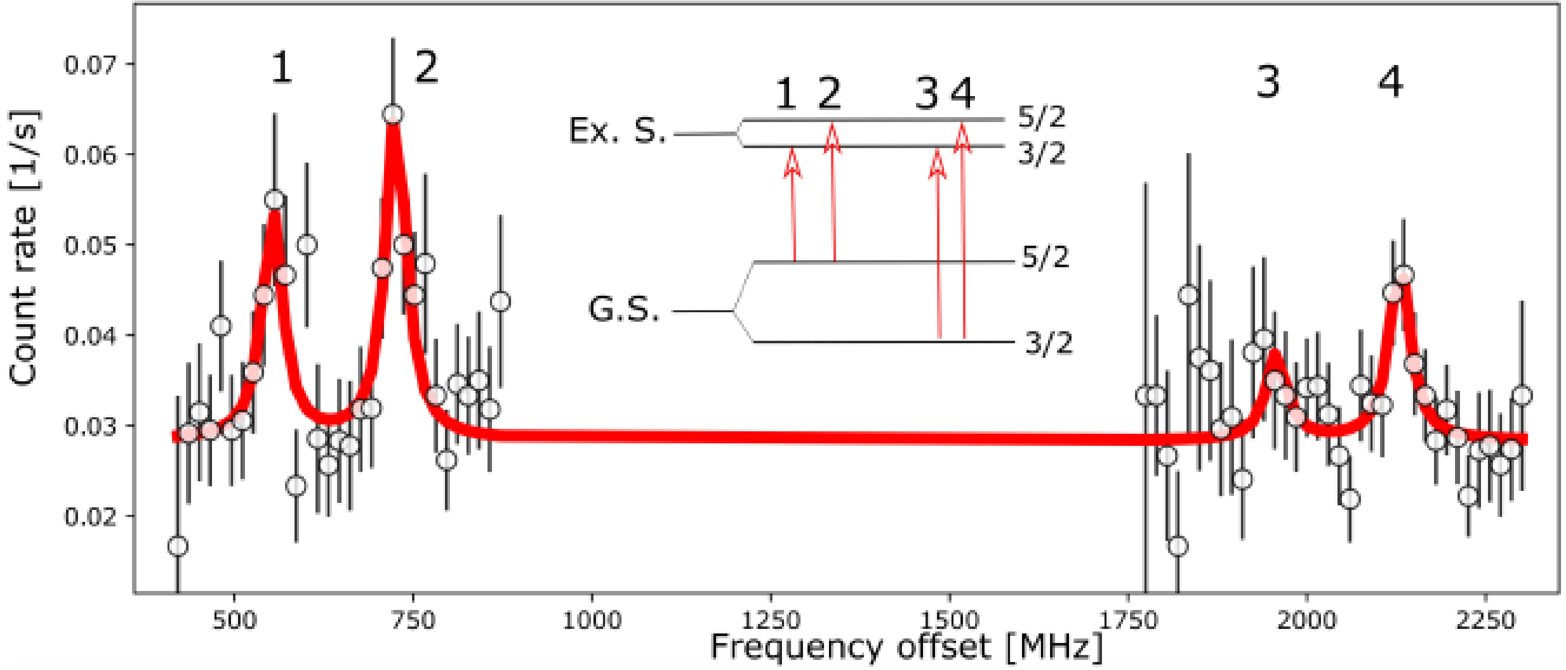
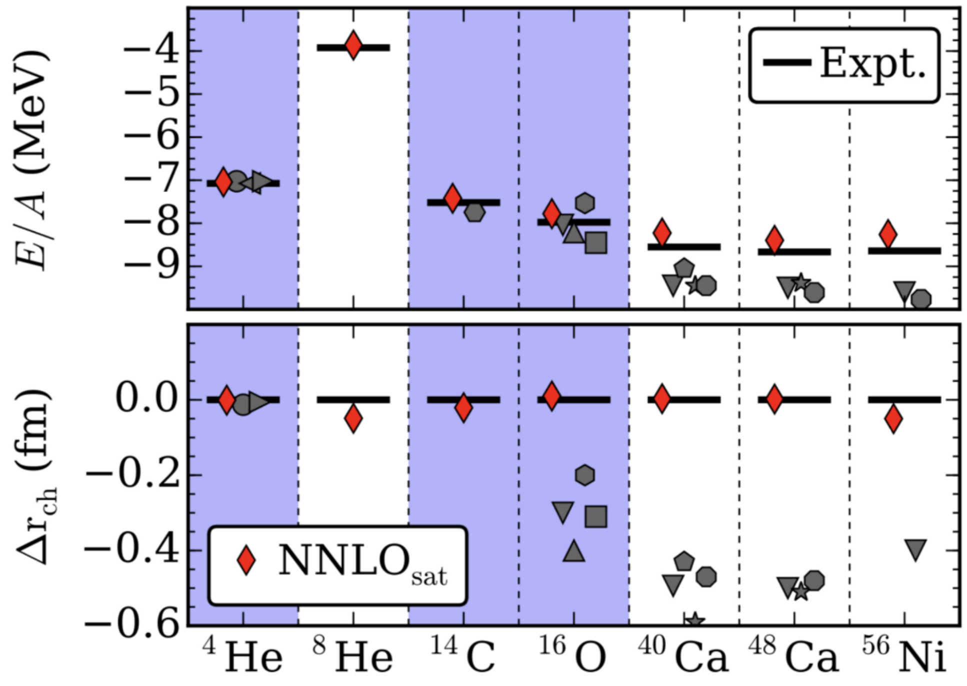
Results



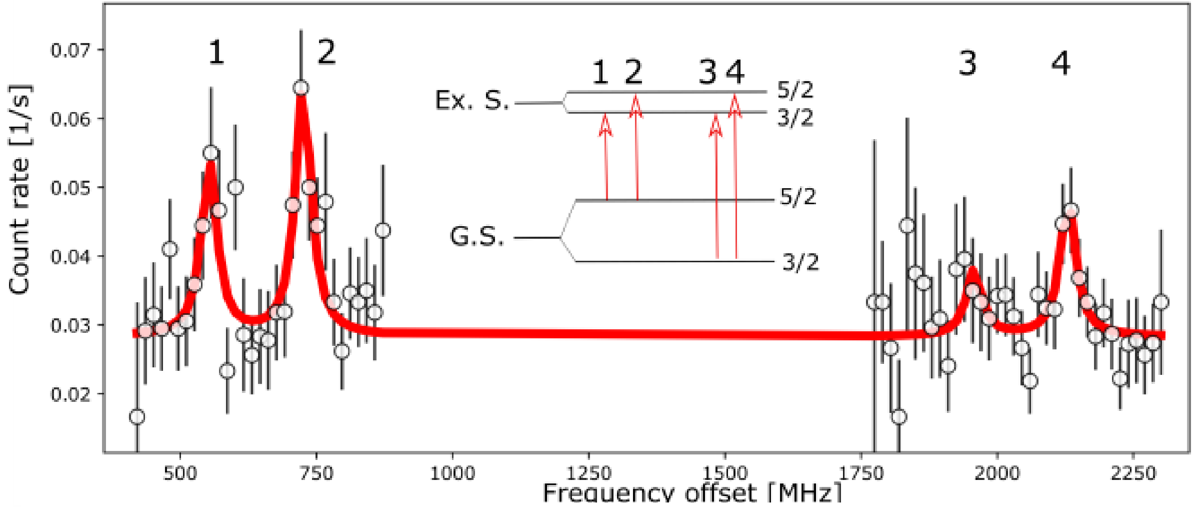
- 300 pps ^{52}K
- < 1 day of data

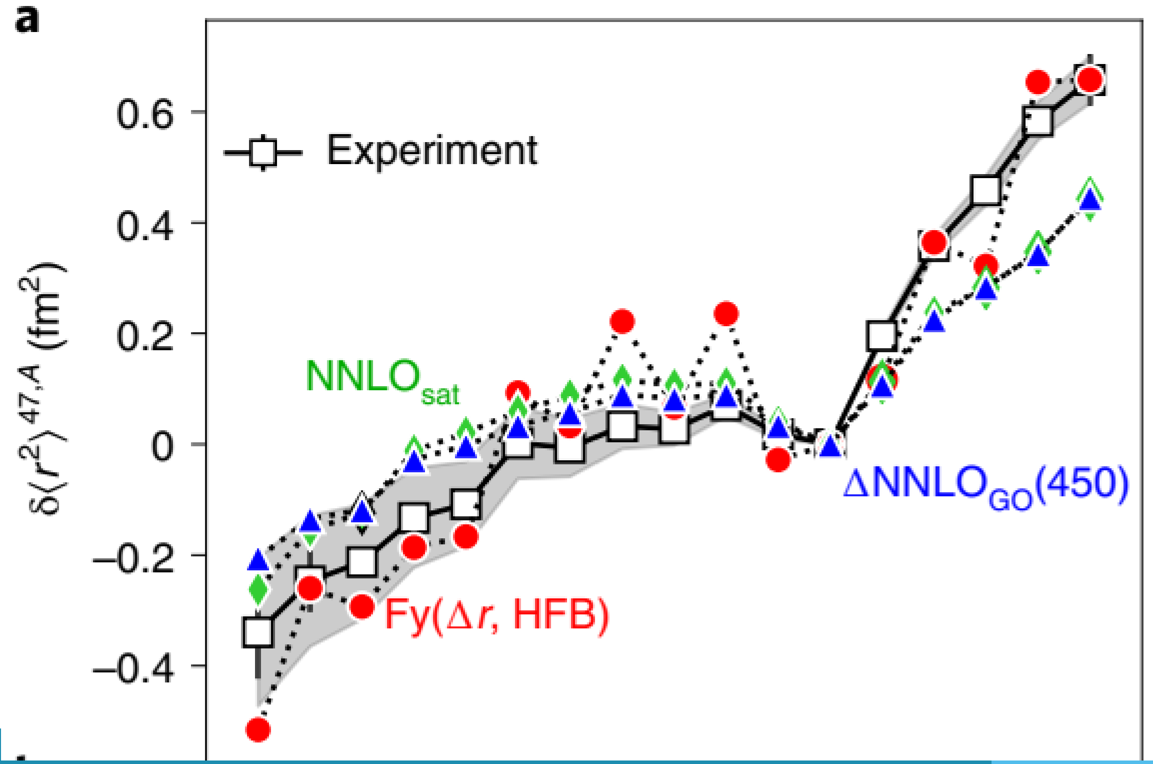
Á. Koszorús, X. F. Yang et al., Nature Physics **17**, 439–443 (2021)

Results



- NNLO$_{sat}$ and newer NNLO$_{Go}$ may work better close to stability, but neutron-rich isotopes are not well modeled…
- What is missing still? We don't know.





K: Á. Koszorús, X. F. Yang et al., Nature Physics 17, 439–443 (2021)
Ca: R. F. Garcia Ruiz et al, Nature Physics **12**, 594–598 (2016)

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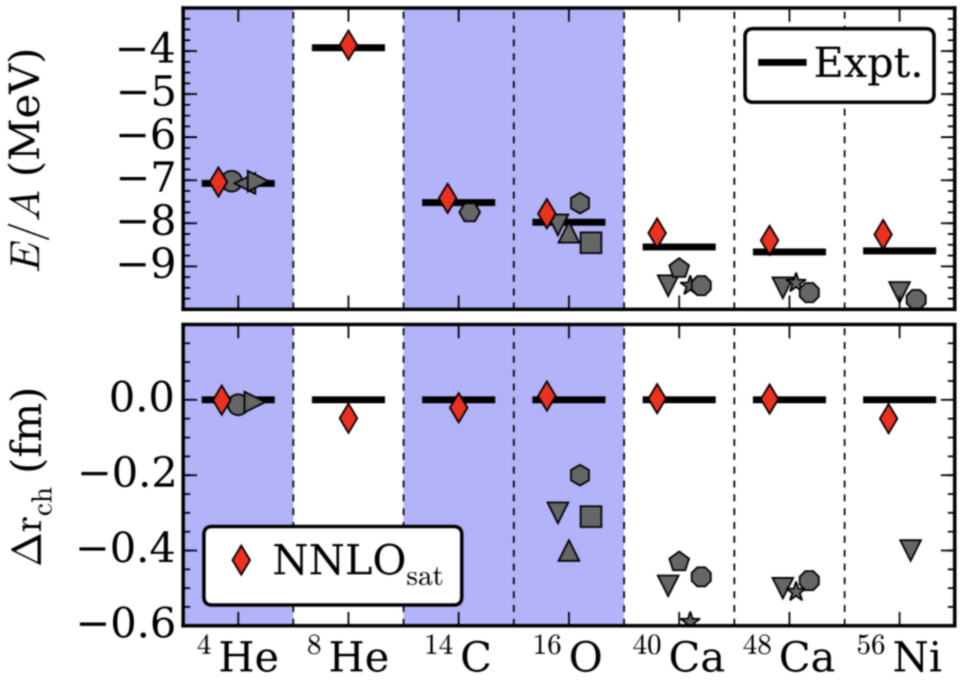

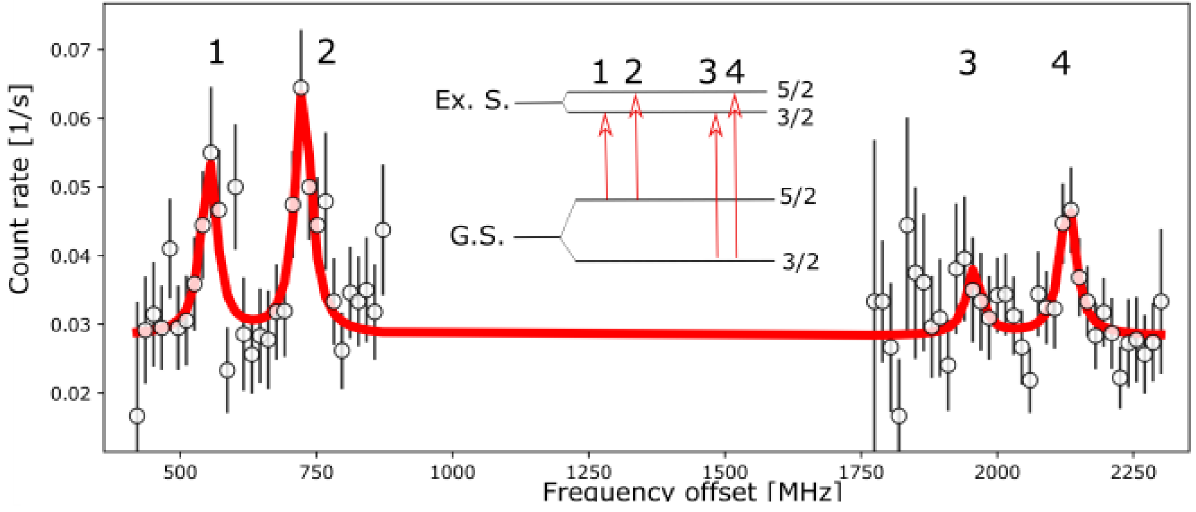

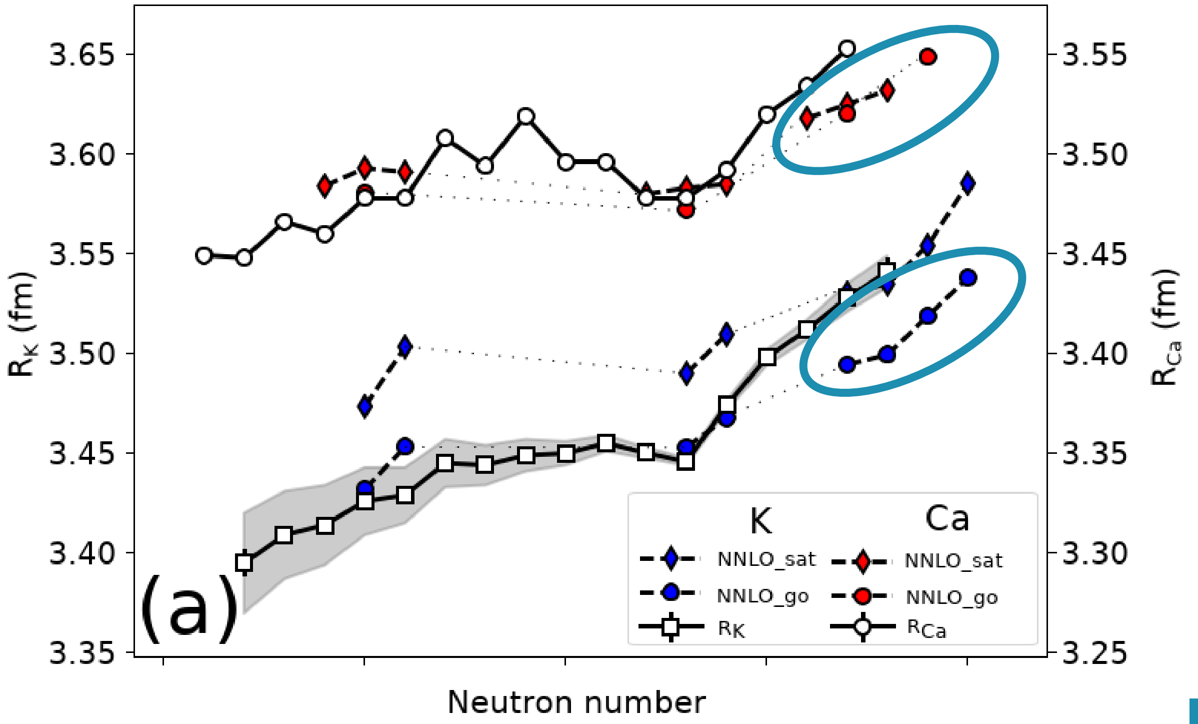

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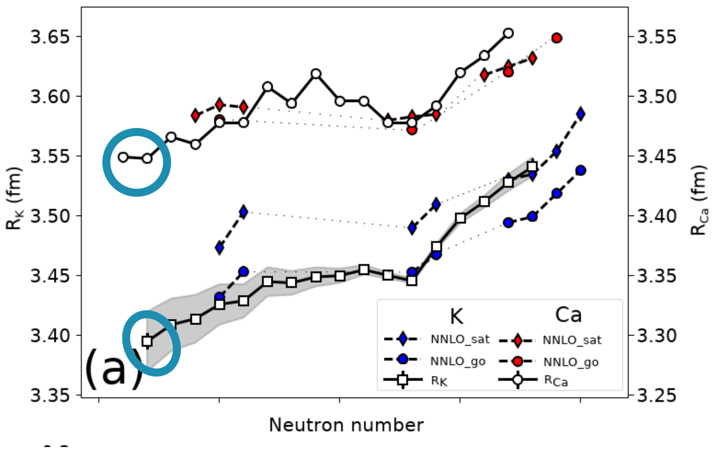
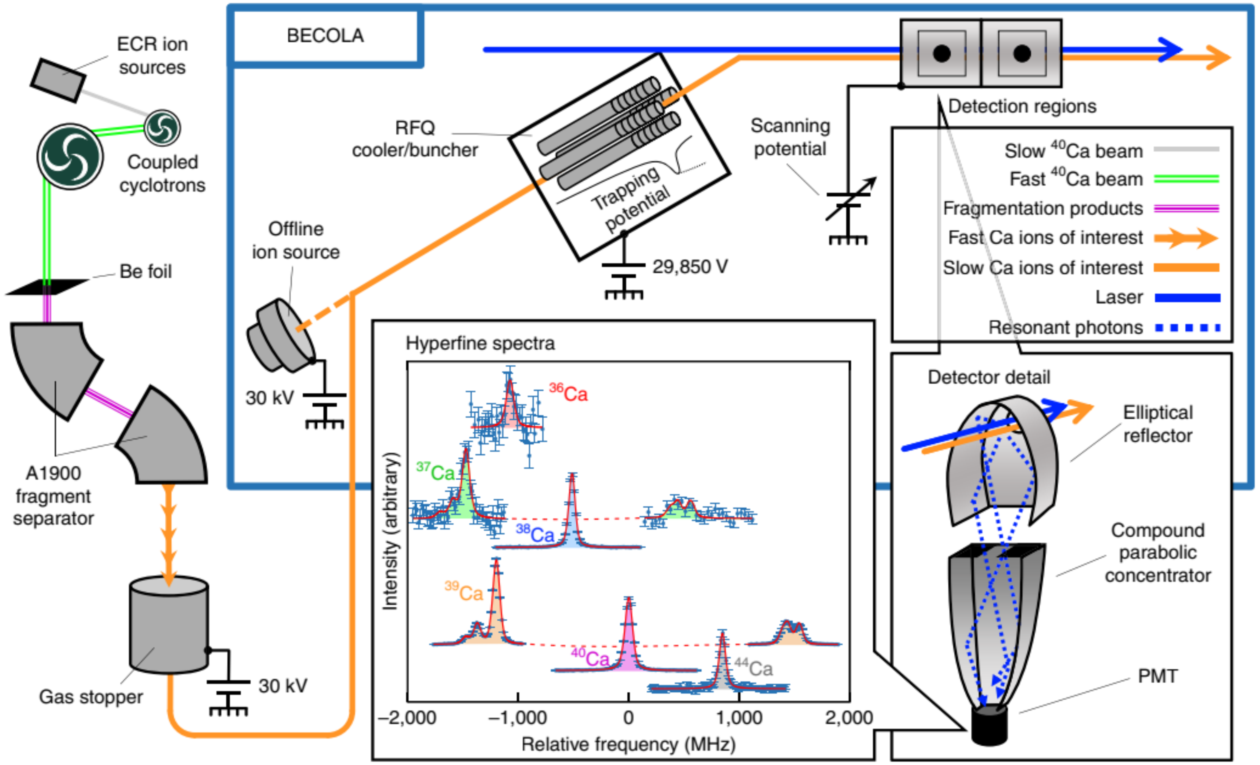
Results

- Where were these neutron-deficient cases measured?
- NSCL!

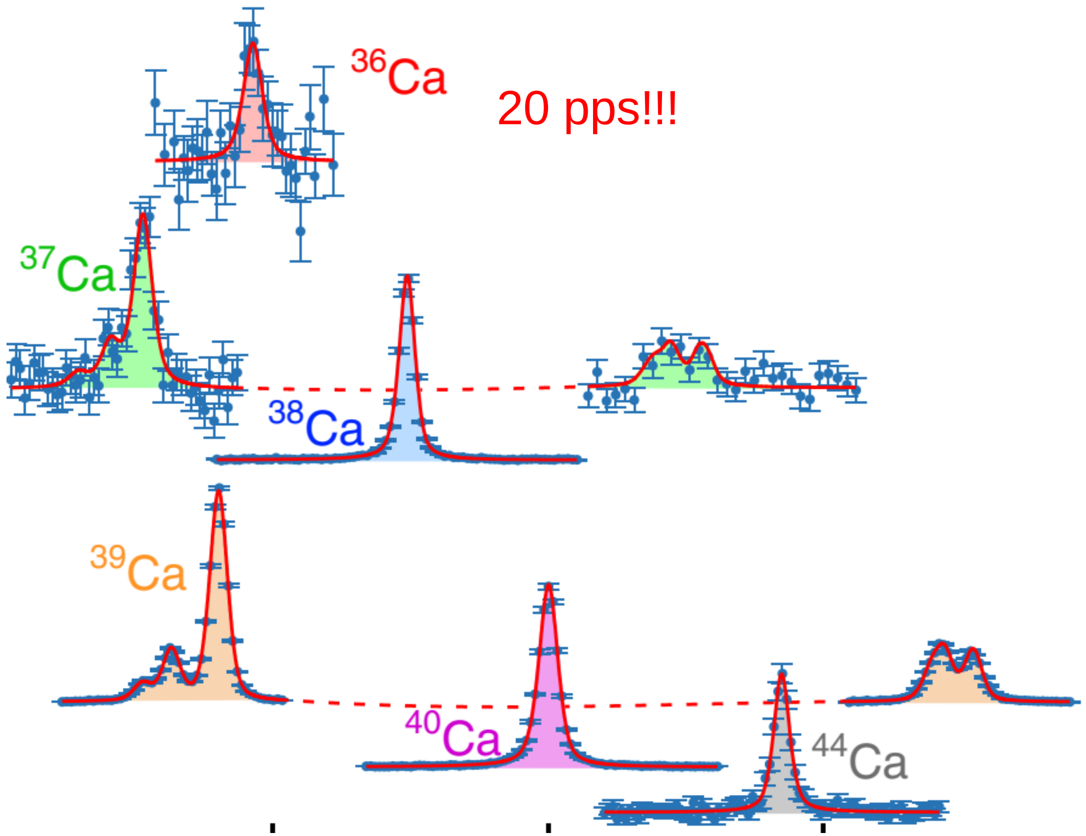
- ^{40}Ca (140 MeV/u) with Be foil
- Separator delivers beam into large-volume gas stopper cell
- Beam extracted, cooled, bunched
- Fluorescence collinear laser spectroscopy
- **CRIS implementation underway!**



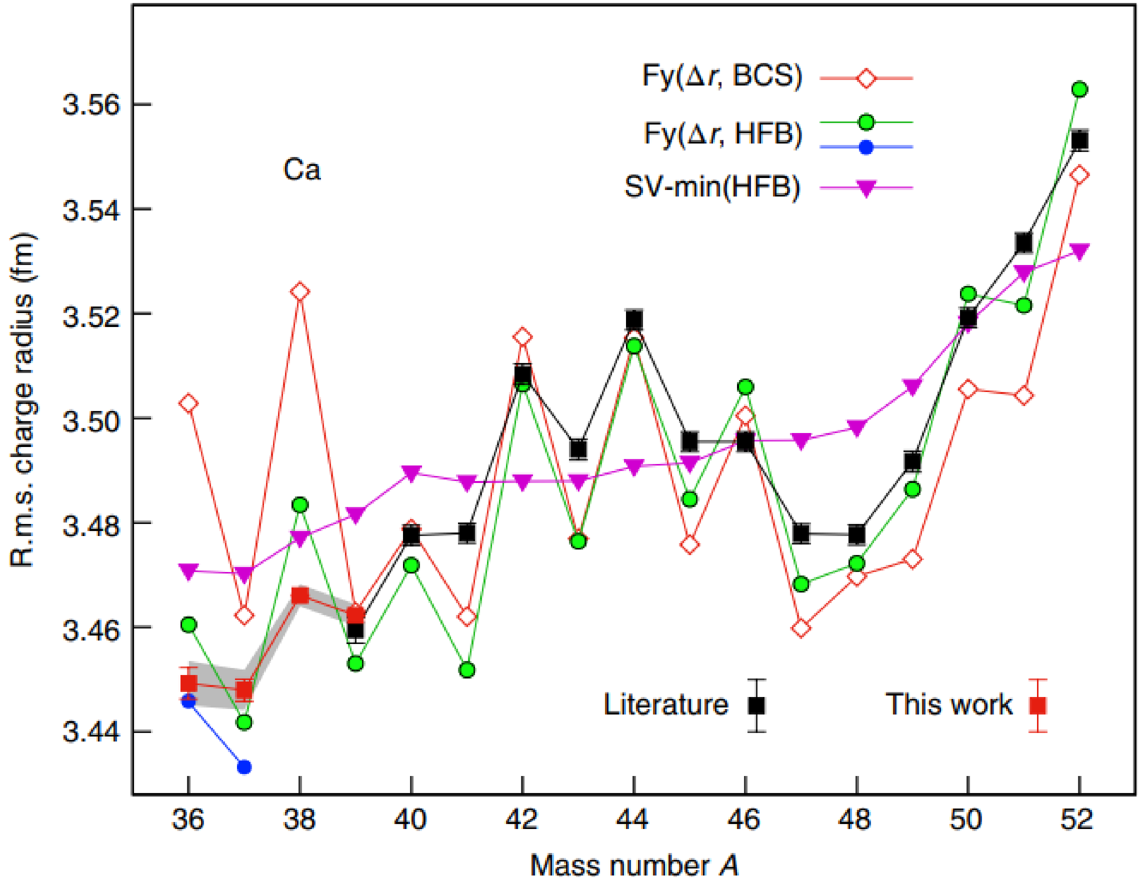


A. J. Miller et al, Nature Physics **15**, 432–436 (2019)

Results



Correct inclusion of continuum states is essential to predict good radii for proton-rich Ca



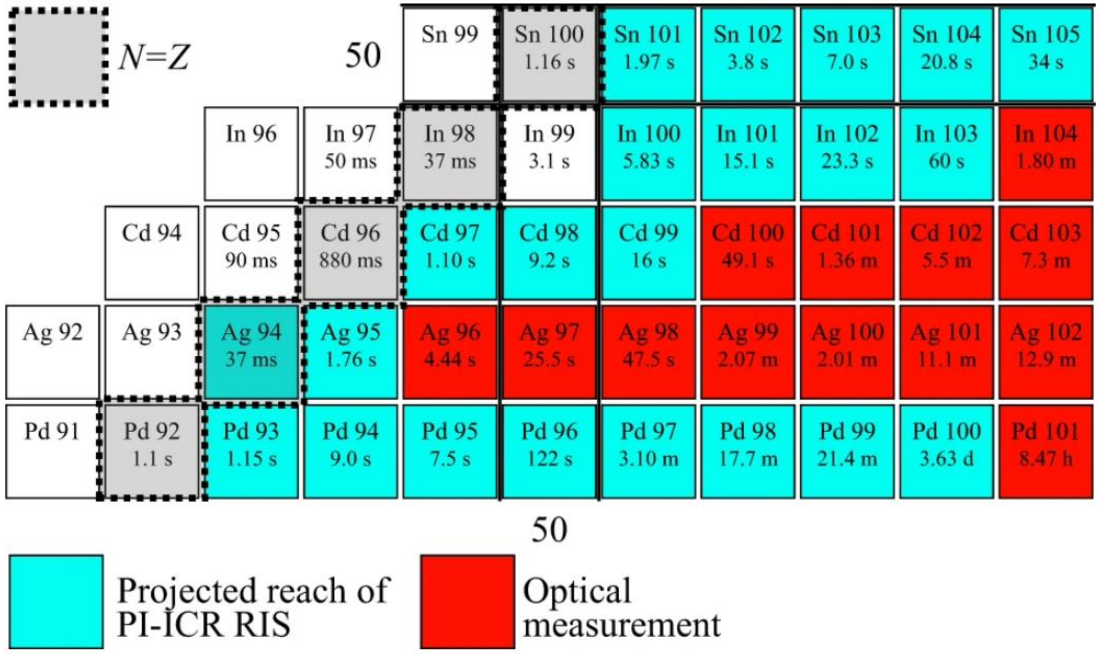
A. J. Miller et al, Nature Physics **15**, 432–436 (2019)

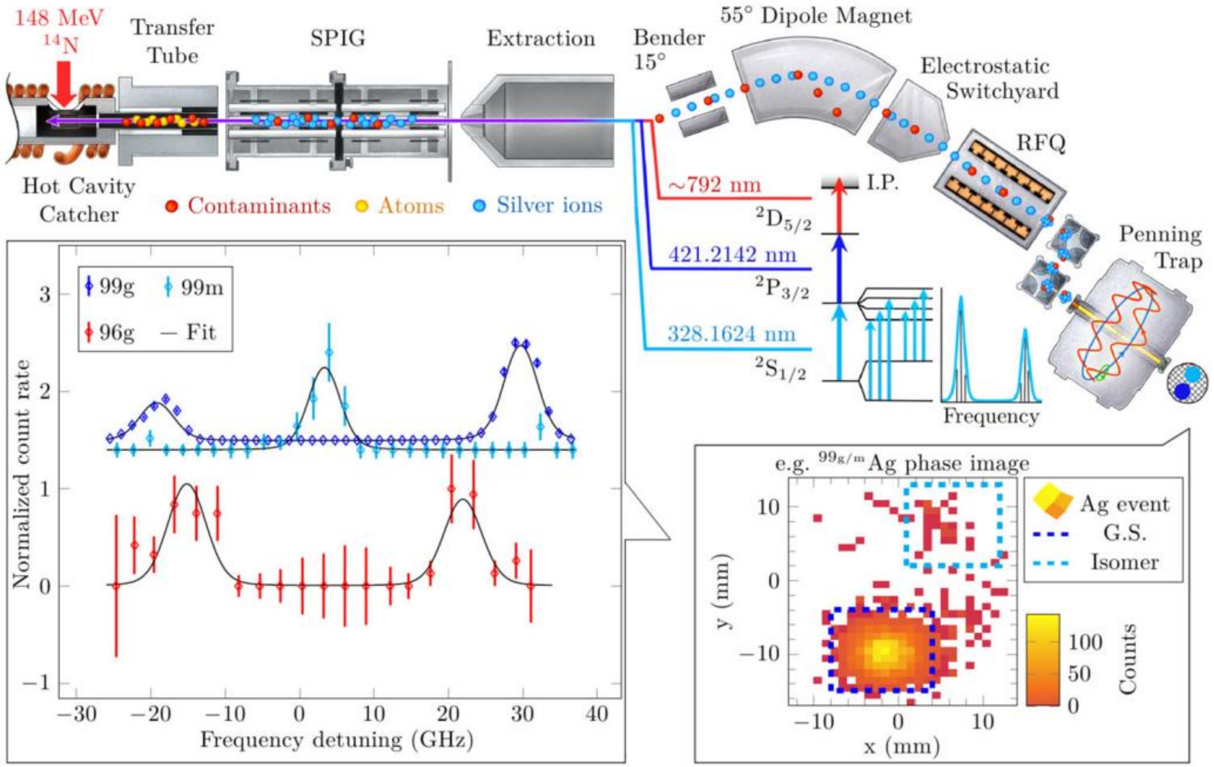
Development of nuclear interactions

Pushing methods to short lifetimes and low yields with in-source laser spectroscopy

Technique: in-source laser spectroscopy

- Push towards ^{94}Ag
- Crossing N=50 shell closure

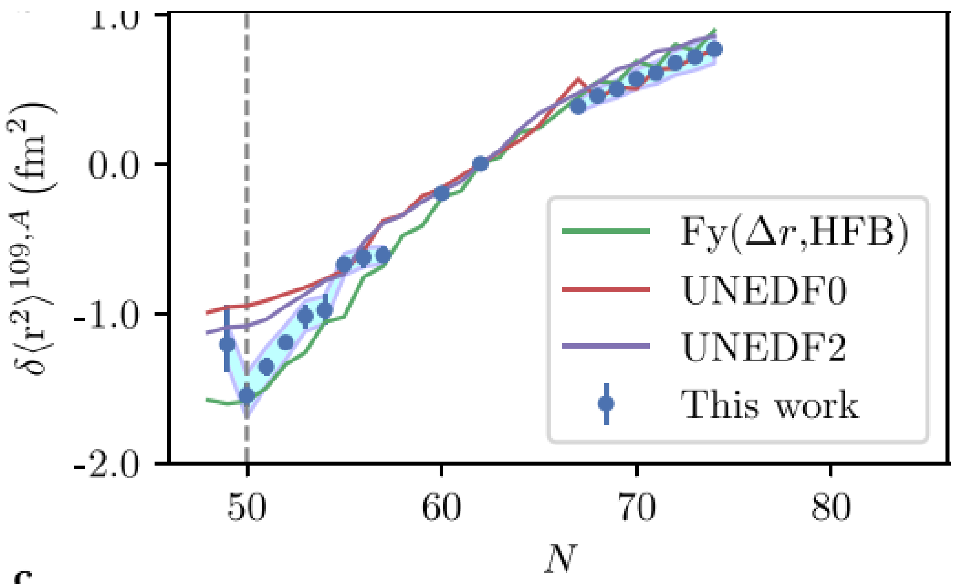


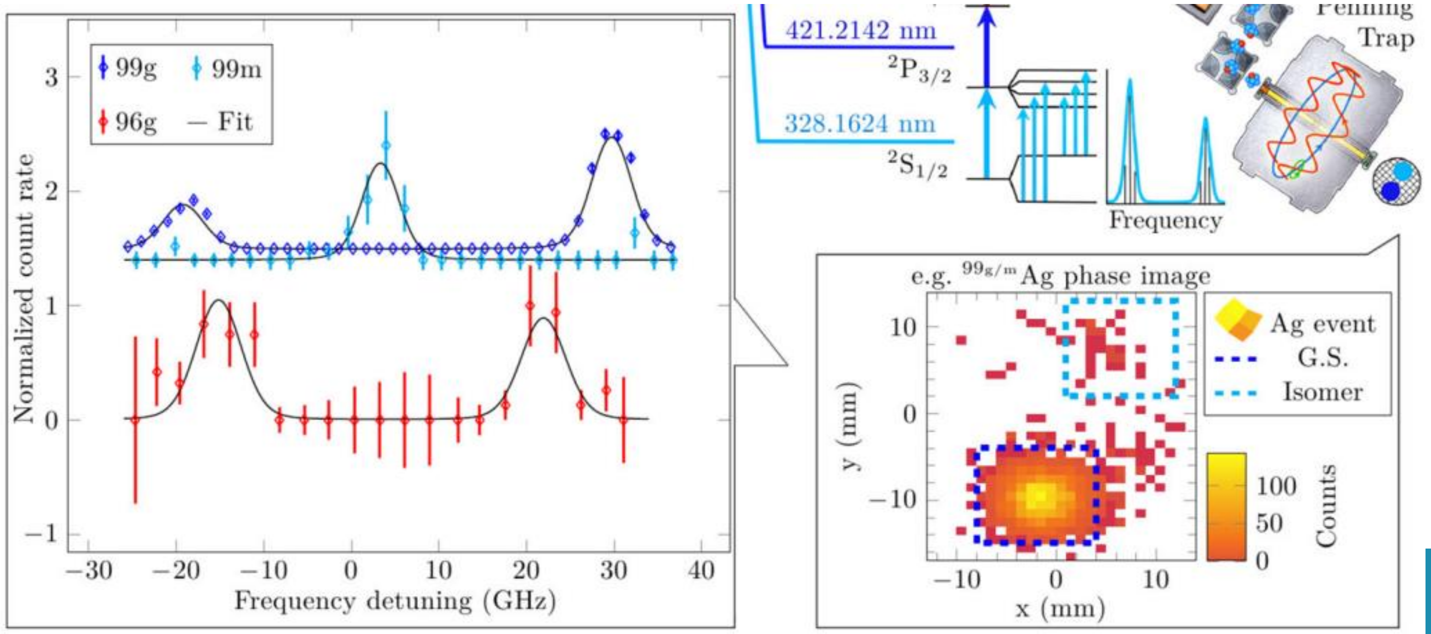


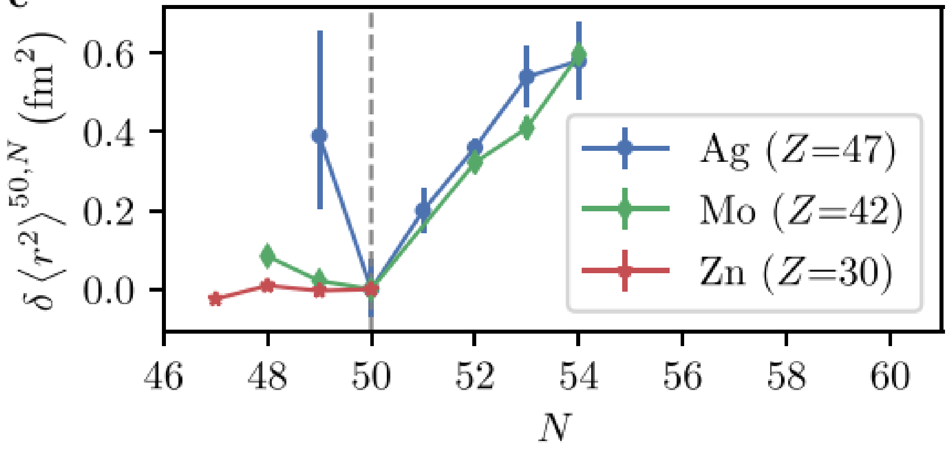
Reponen, M., de Groote, R.P., et al. Evidence of a sudden increase in the nuclear size of proton-rich silver-96. Nat Commun 12, 4596 (2021)

Results

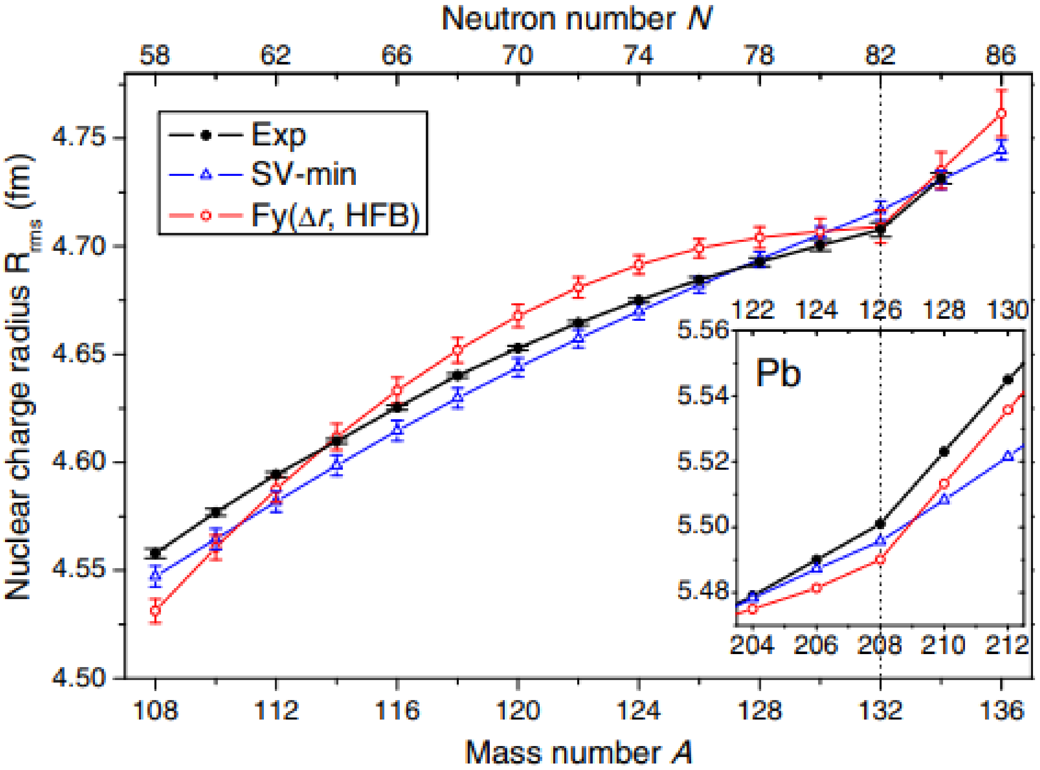
- Spectra obtained for $^{104\text{-}96}$Ag using ^{14}N(^{92}Mo, 2pxn) Ag
- *Totally* background-free measurements
- Very sharp kink observed at N=50 – beyond current DFT models. More data needed to refine error bar.

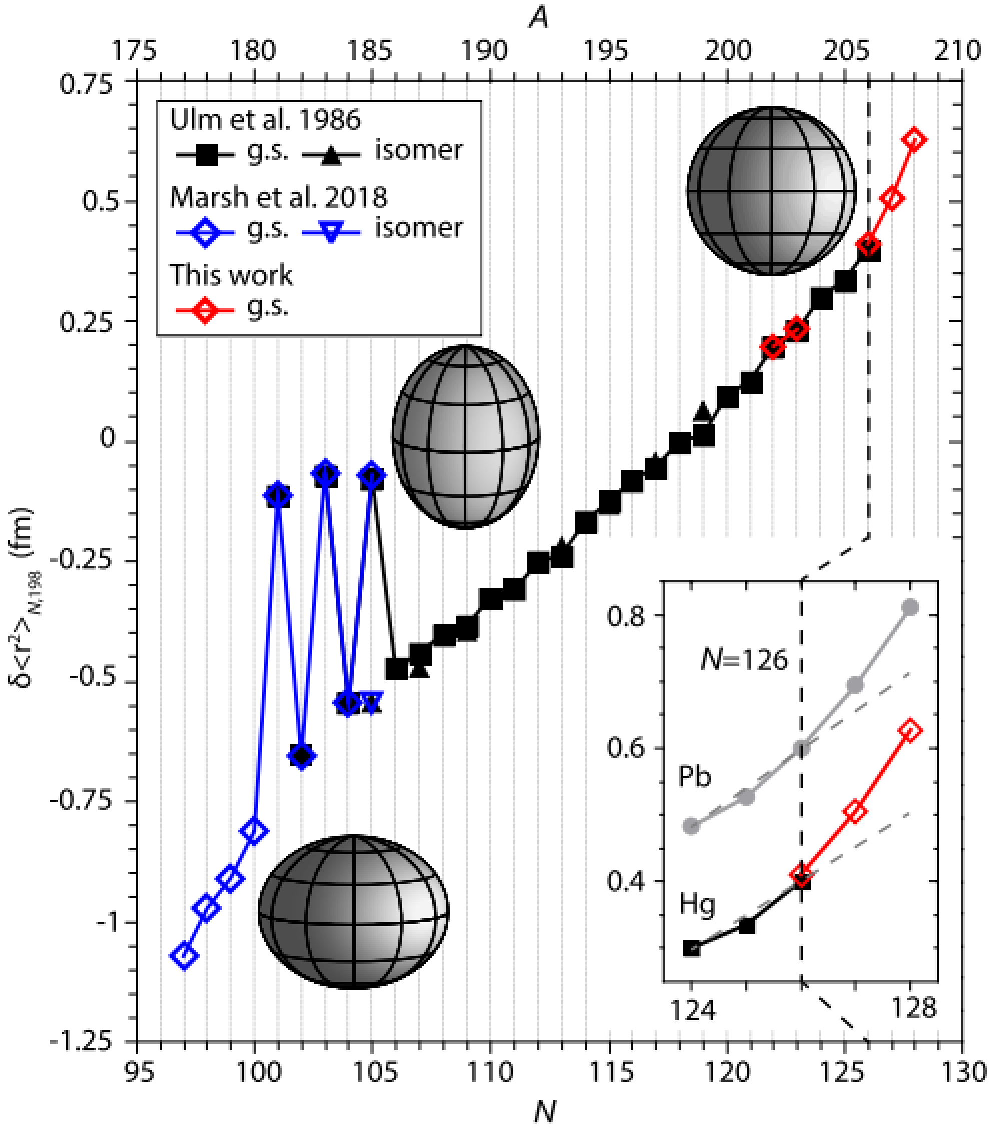






Other recent studies near shell closures…





C. Gorges, L. V. Rodríguez, PRL **122**, 192502 (2019)
T. Day Goodacre, PRL **126**, 032502 (2021)

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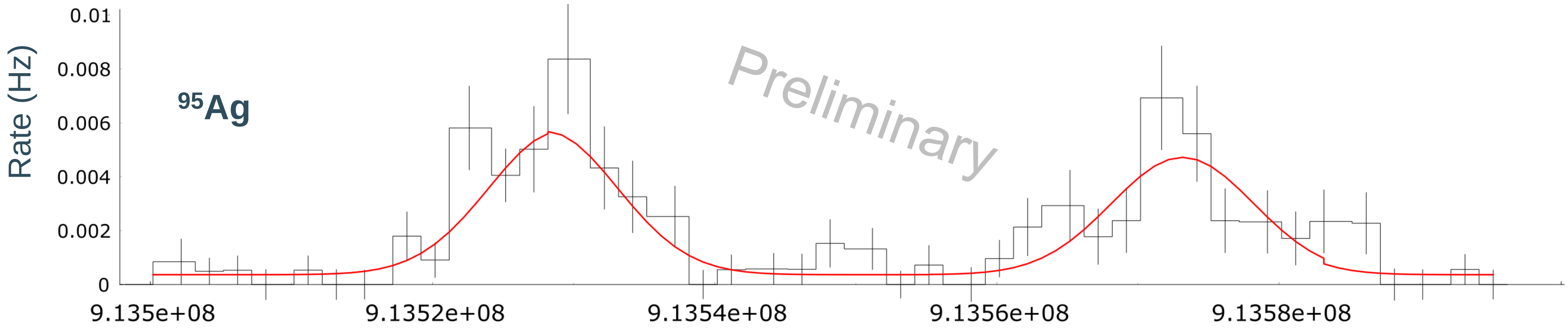
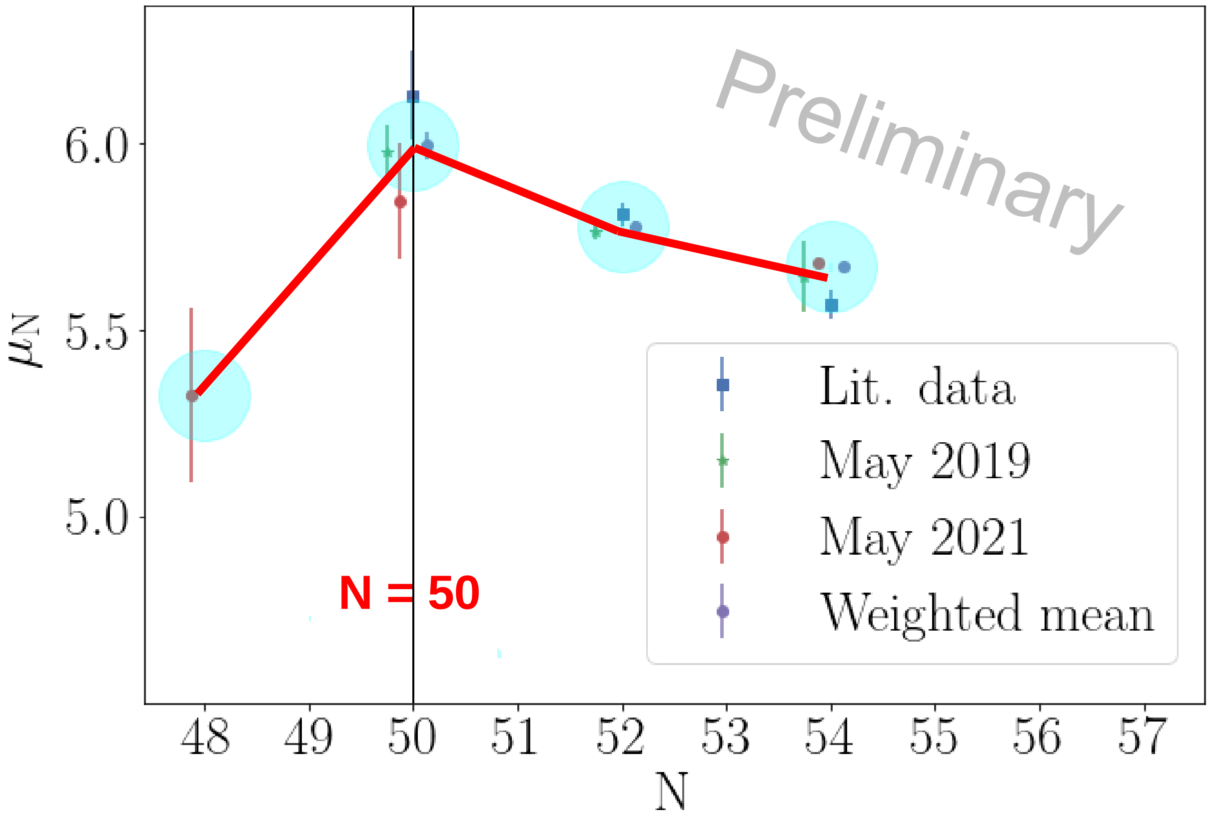
- **Data taken for ^{96,95}Ag!! using ^{40}Ca($^{58/60}$Ni, pxn) Ag**
- Isotope shift under analysis

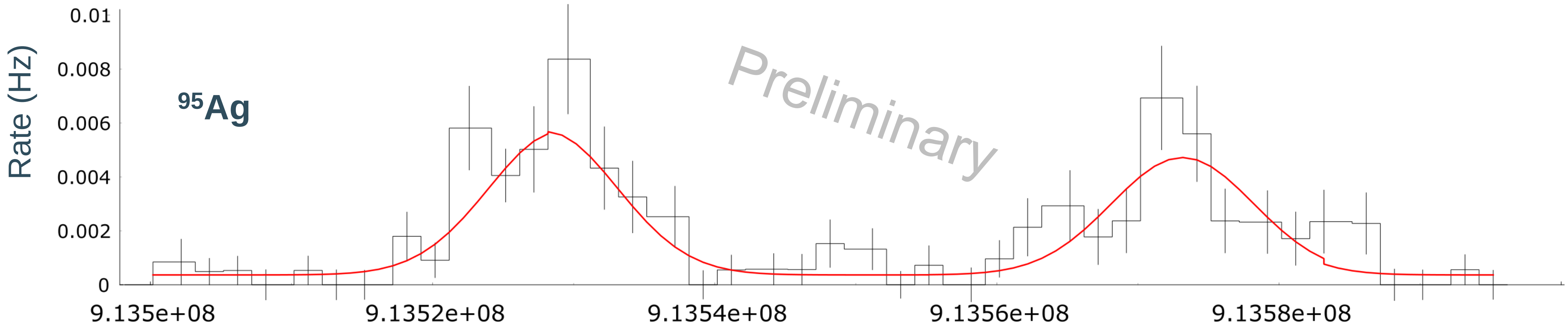


Results

- Spectra obtained for $^{104-96}$Ag using ^{14}N(^{92}Mo, 2pxn) Ag
- *Totally* background-free measurements
- Very sharp kink observed at N=50 – beyond current DFT models. More data needed to refine error bar.

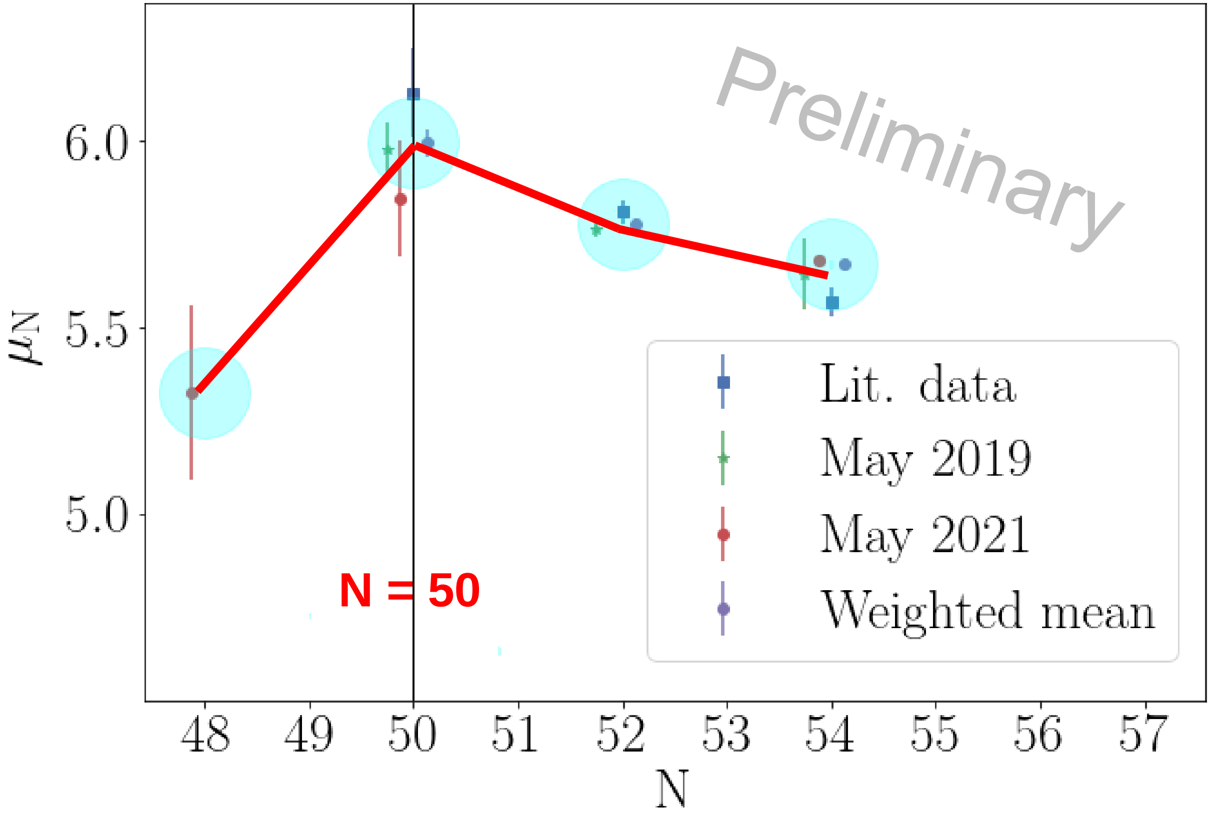
- **Data taken for ^{96,95}Ag!! using ^{40}Ca($^{58/60}$Ni, pxn) Ag**
- Isotope shift under analysis



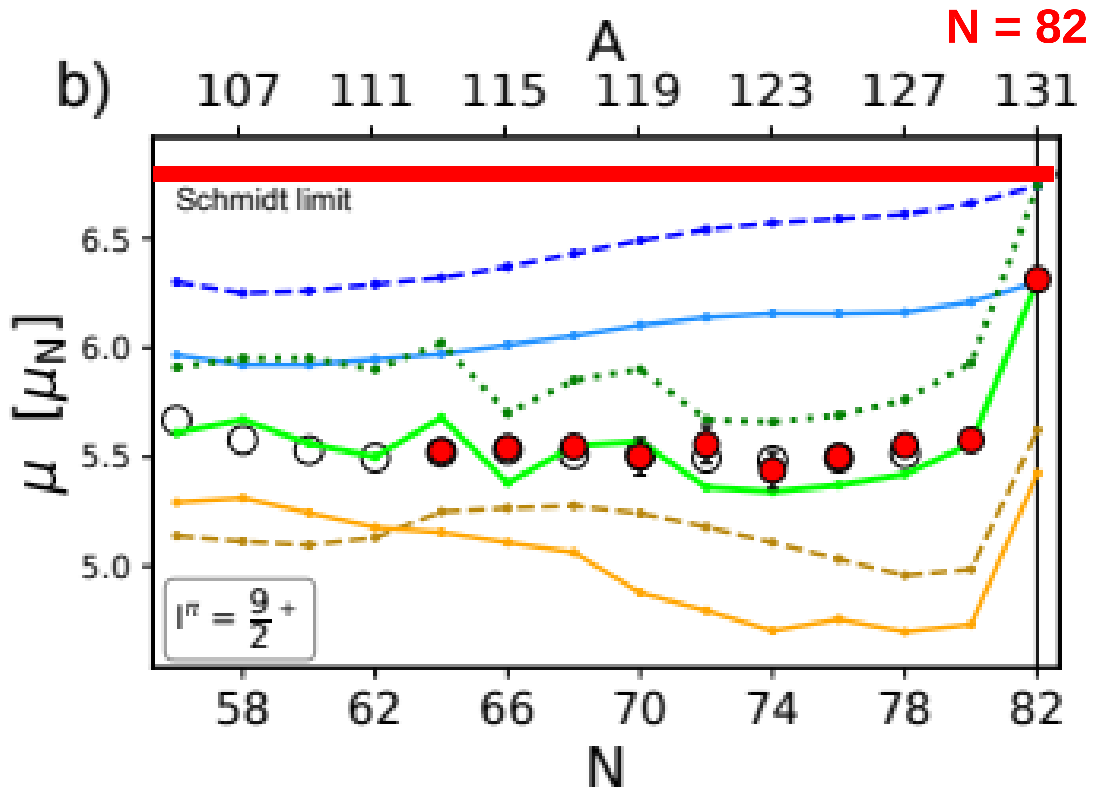


Lots more data to consider...

Silver (LISOL, JYU)



Indium, CRIS (ISOLDE)



M. Reponen et al, in preparation

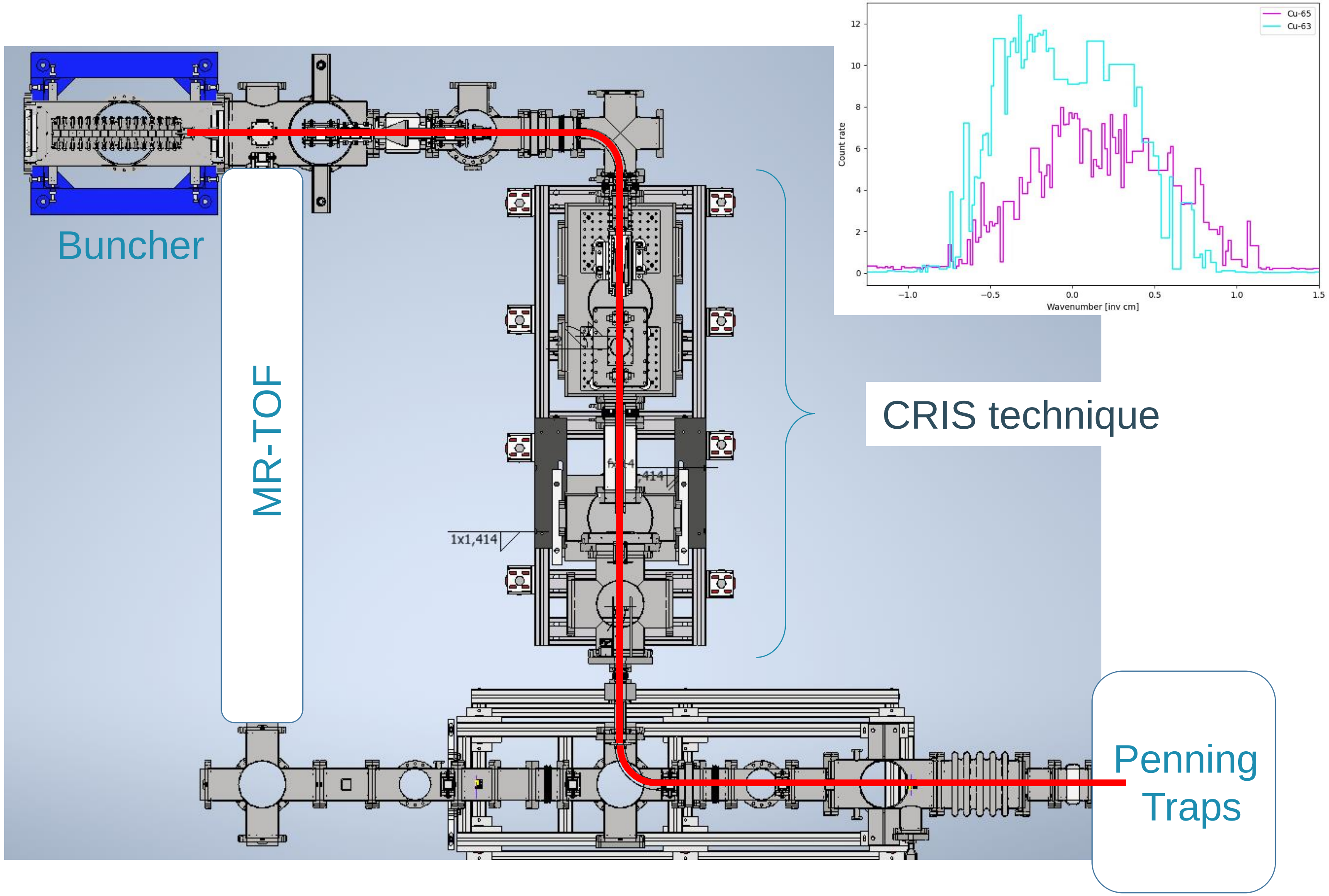
A. R. Vernon et al, under review

RAPTOR

- “RIS And Purification Traps for Optimised spectroscopy”

- Collinear laser resonance ionization spectroscopy
  - Collinear geometry at a few kV beam energy yields ~100 MHz linewidth
  - Laser resonance ionization provides high efficiency
  - Using Penning trap as ultimate background-removal device

First resonances!!



MARIE CURIE ACTIONS

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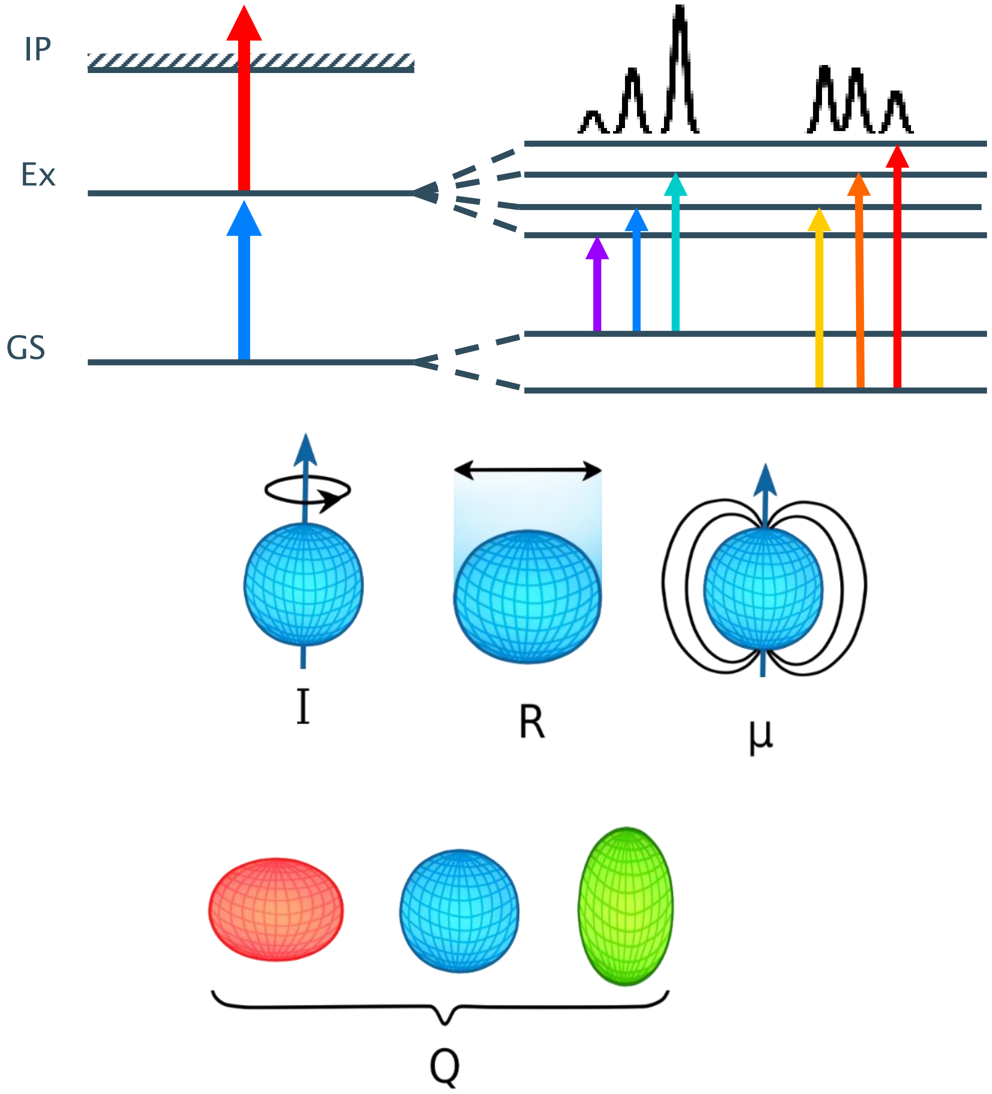
Precision measurements

Pushing beyond doppler-free optical spectroscopy

Scientific case

- Overview of hyperfine structure shown in the start was a simplification.
  - Hyperfine anomaly: ratio of atomic *A* and the nuclear moment is ***not*** a constant over a chain of isotopes
  - Reason: as the number of nucleons changes, so does the distribution of magnetization
  - Higher-order nuclear moments: there is physics *beyond* the quadrupole
- Measuring these effects typically requires precisions beyond the techniques shown so far
  - Hyperfine anomaly: measurable in *typically* the heavier cases (some exceptions)



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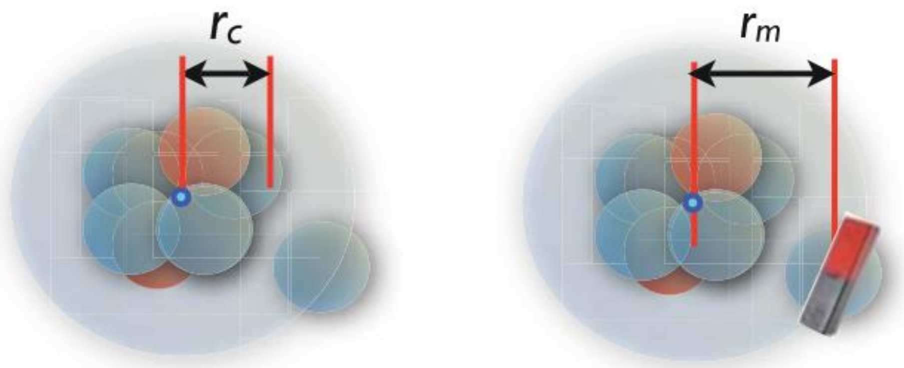
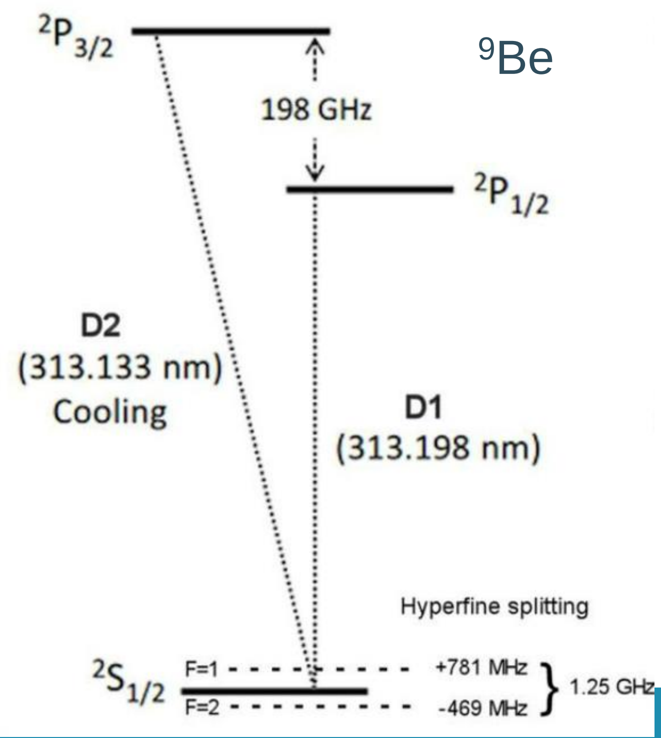
Scientific case

- Hyperfine anomaly of a neutron halo?
  - The *charge radius* is only a measure of where the protons are
  - The *magnetization radius* is affected by the neutrons as well!

- ^{11}Be:
  - neutron halo
  - Has a simple atomic structure conducive to precision spectroscopy





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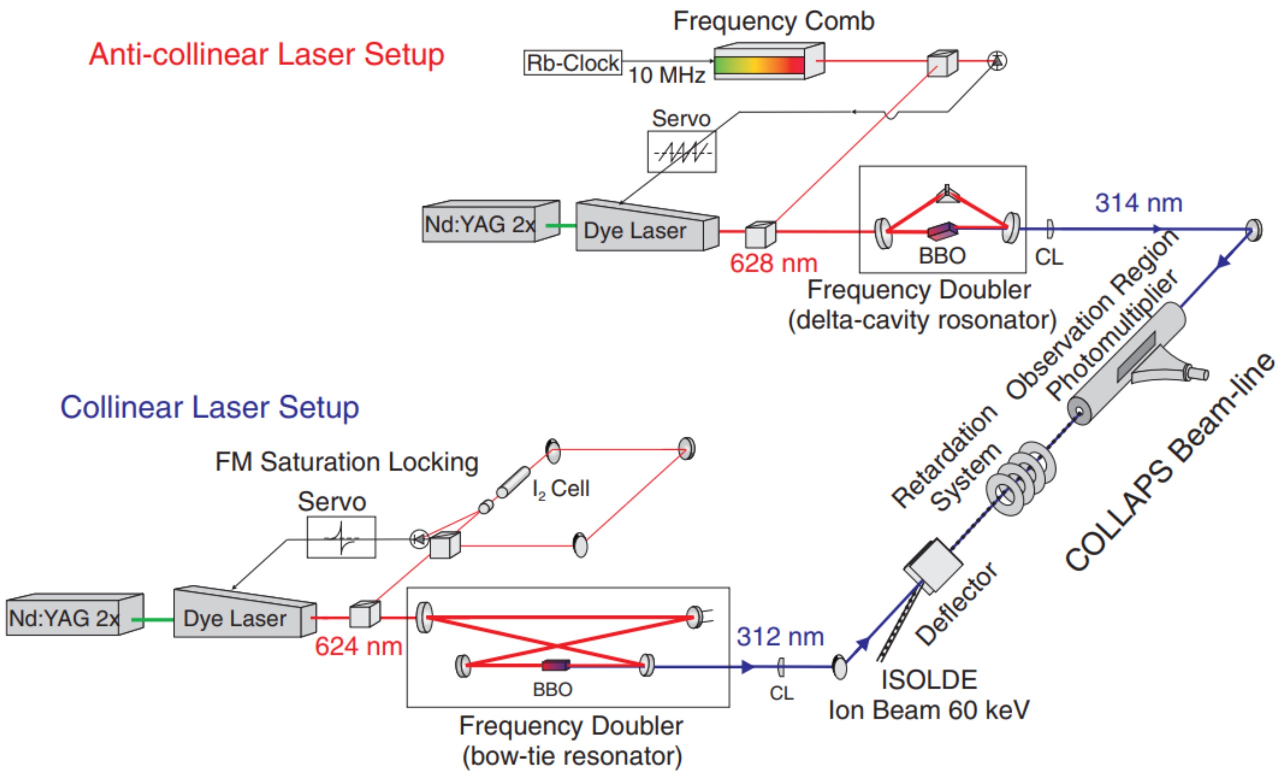
Results – charge radius

- Precision collinear laser spectroscopy of Be
- Be is very light
  ➔ Atomic field shift factor is small
- Problem with collinear laser spectroscopy: exact velocity of the ions needs to be determined
- Accuracy and precision of 100 kHz is required

$$\nu_c = \nu_0 \cdot \gamma \cdot (1 + \beta),$$

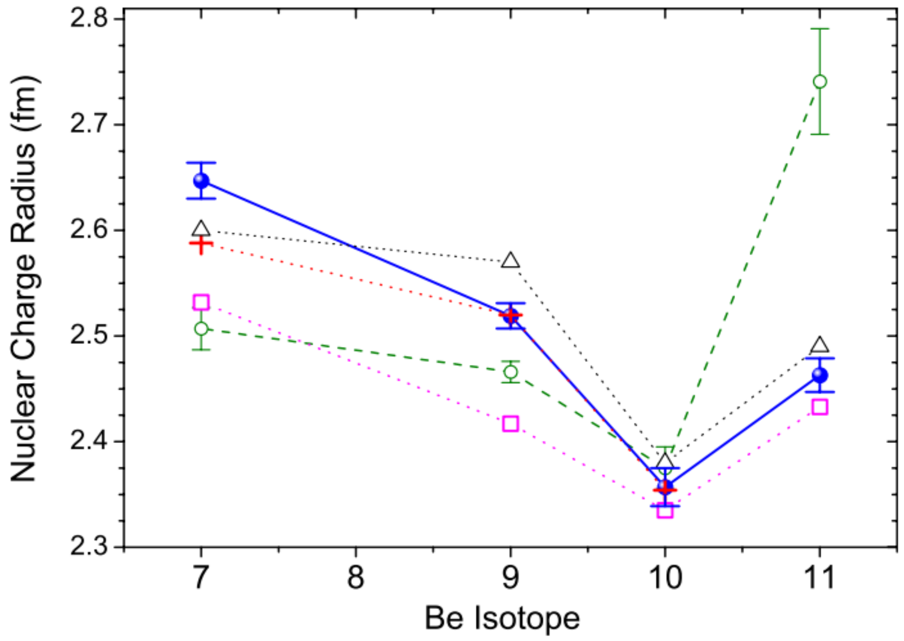
$$\nu_a = \nu_0 \cdot \gamma \cdot (1 - \beta),$$

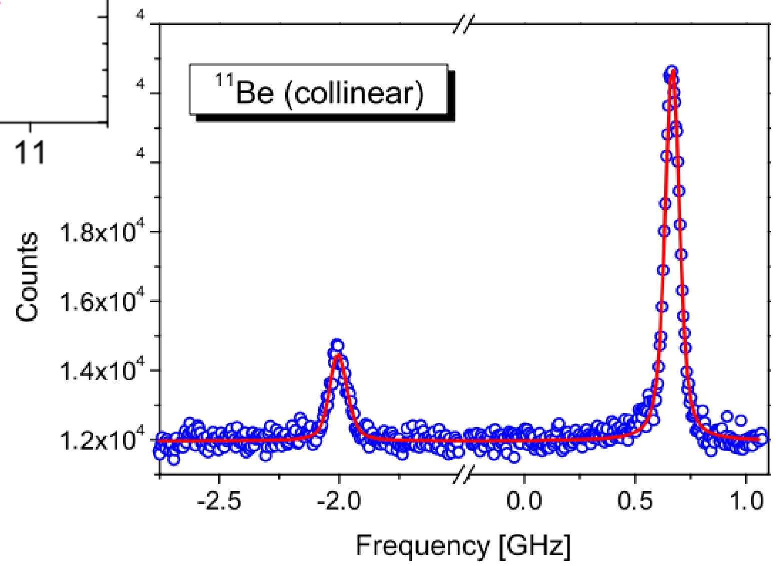
$$\nu_0^2 = \nu_c \cdot \nu_a$$

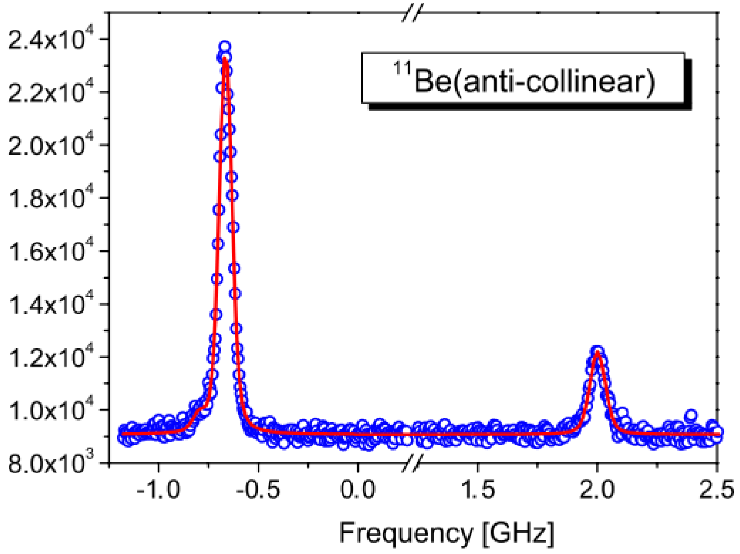


M. Žáková et al., J. Phys. G: Nucl. Part. Phys. 37 (2010) 055107

Results – charge radius



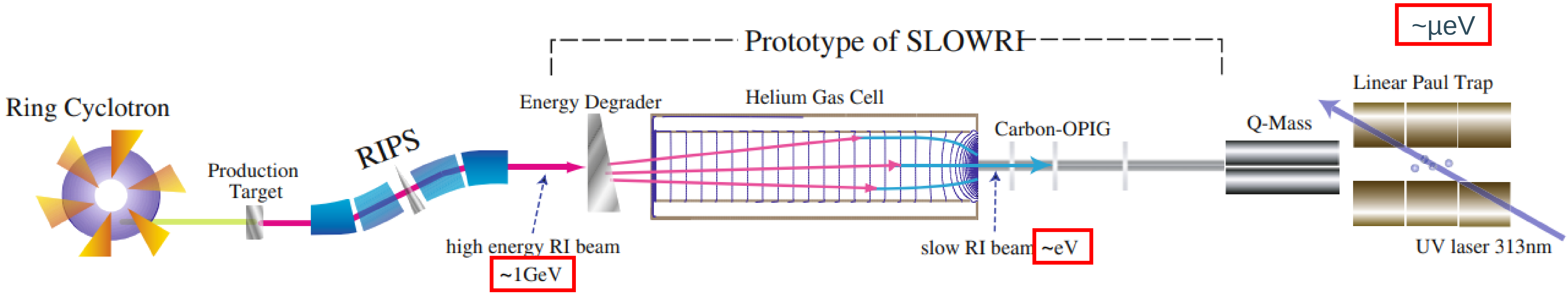




M. Žáková et al., J. Phys. G: Nucl. Part. Phys. 37 (2010) 055107
W. Nörtershäuser et al., Phys. Rev. Lett. 102, 062503 (2009)

Results – hyperfine anomaly

- Charge distribution ✓
- Measuring hyperfine anomaly:
  - Measure the g-factor *independently* *(not done yet!)*
  - **Measure the hyperfine A-constant**



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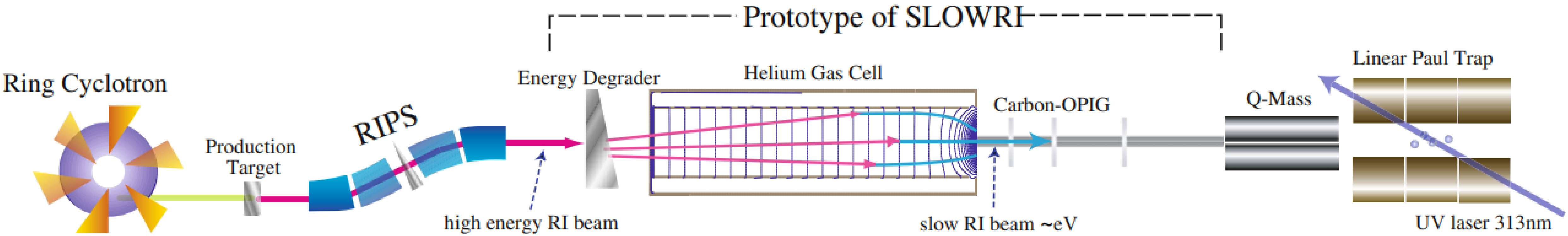
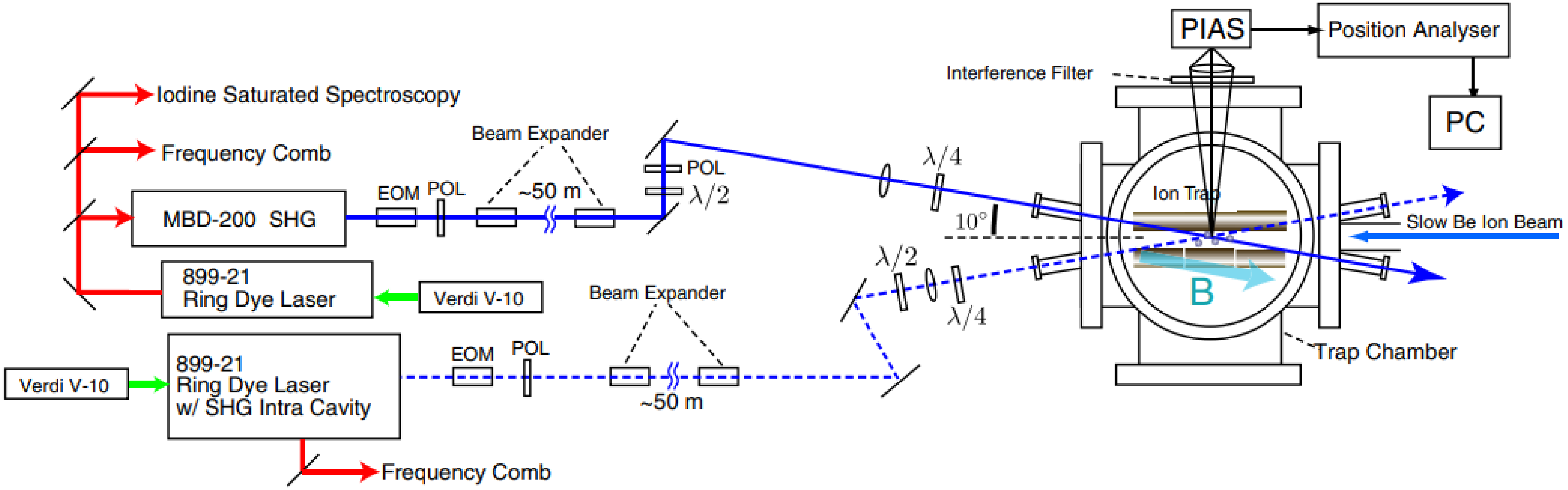
Technique





A. Takamine et al., Eur. Phys. J. A 42, 369–373 (2009)

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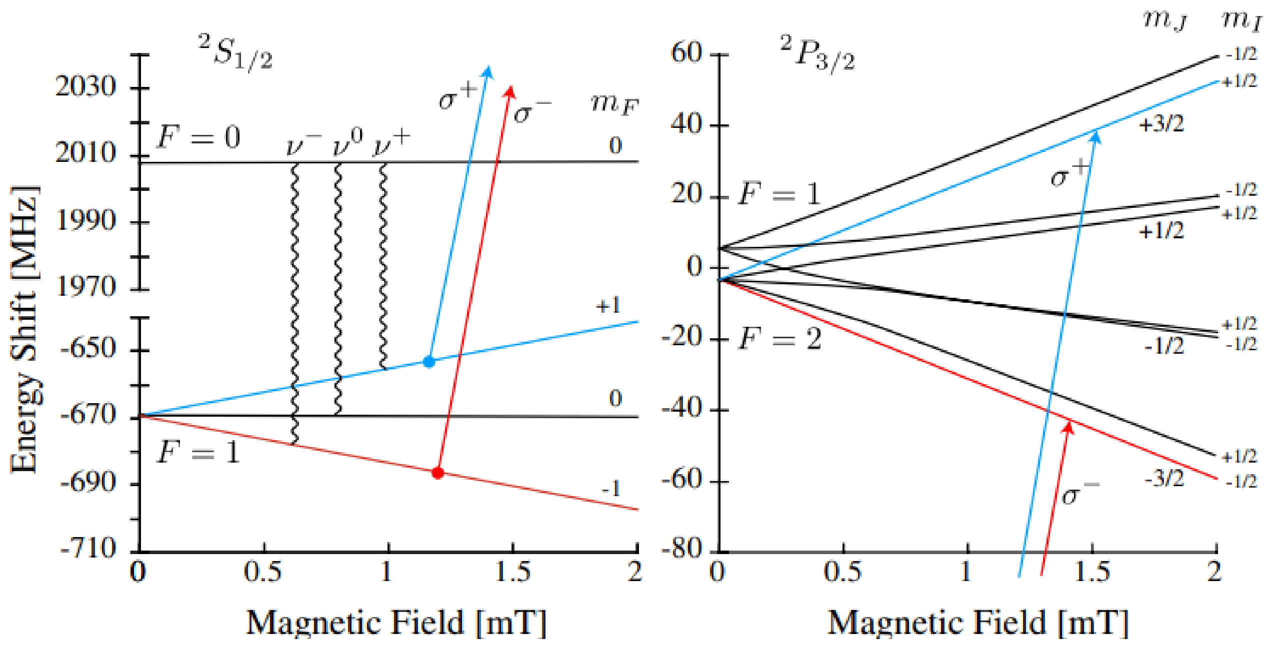
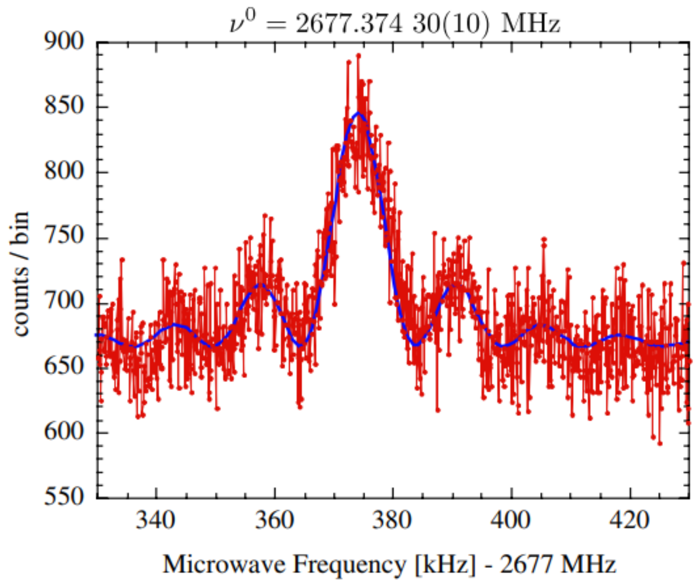
Technique

- Optical resonance is not scanned
- Radiofrequency excitation within ground-state hyperfine manifold
- Optical ‘amplifier’ of RF resonance

- Precision: ppb…
- Awaits independent g-factor measurement to evaluate HFA



Isotope	A [MHz]
^{7}Be	−742.772 28(43)[c]
^{9}Be	−625.008 837 048(10)[d]
^{11}Be	−2677.302 988(72)[b]

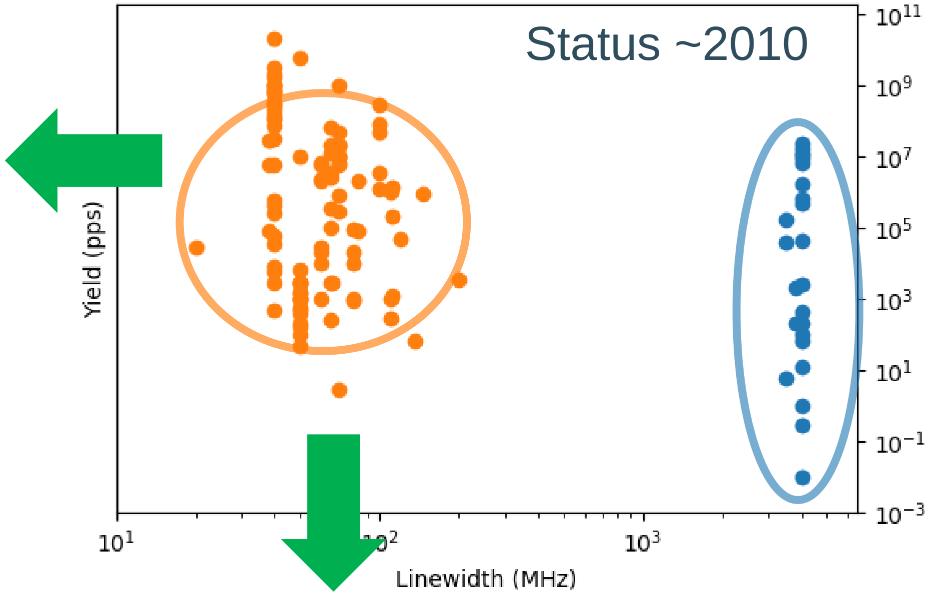


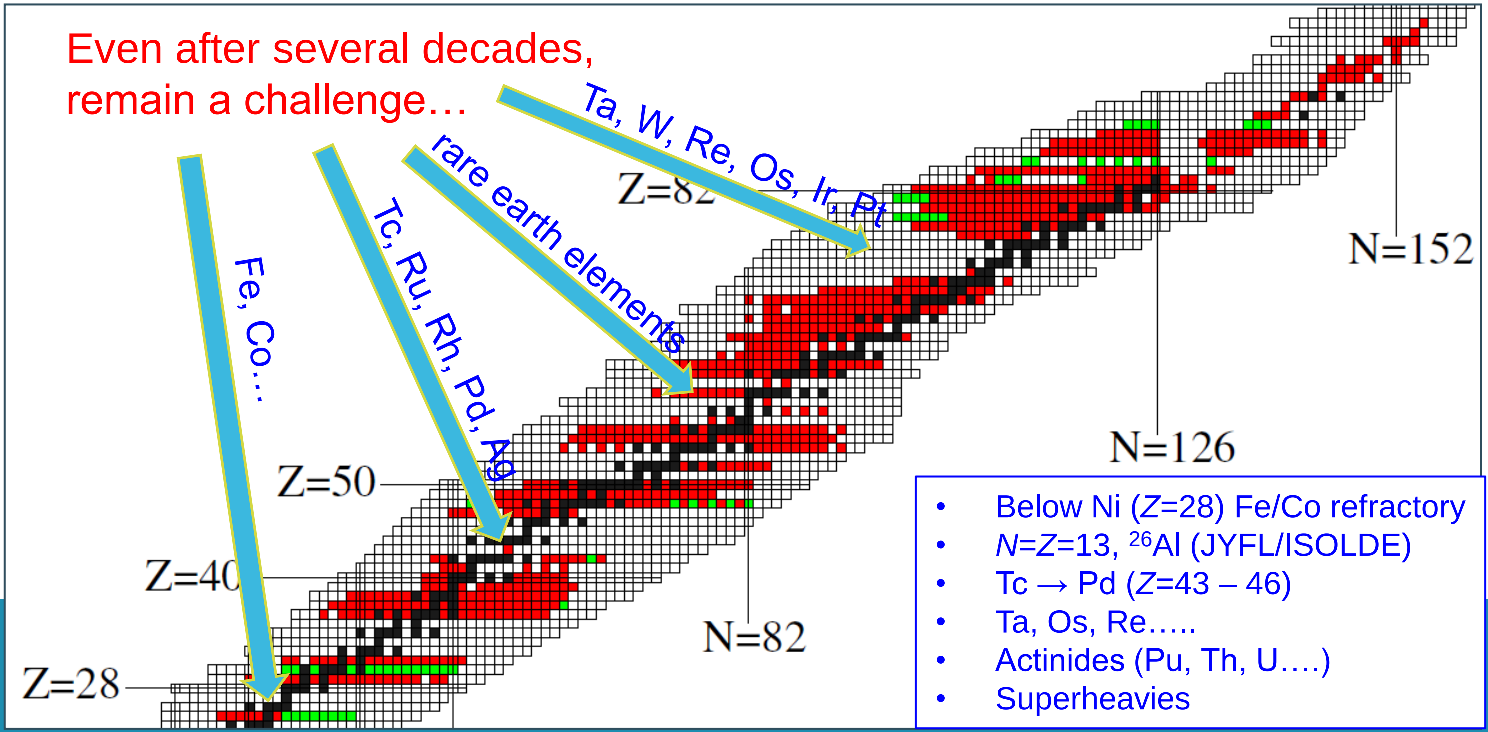
A. Takamine et al., PRL **112**, 162502 (2014)

Conclusions

Some concluding remarks

- Nuclear chart has been successfully explored extensively with optical techniques
  - **Challenges remain**: the most exotic, most short-lived, challenging chemistry, complex or unknown atomic structure, …
- Combination of RIS with ultra-selective and efficient detection techniques provides **access to the most exotic isotopes**
  - Collaboration between different specialists of different low energy beam techniques is *vital*
  - Use of diverse techniques is *vital*
- Exploration of different facilities opens new frontiers
  - e.g. IGISOL: access refractory isotopes
  - e.g. NSCL/FRIB: fragmentation reactions
  - e.g. GANIL/S3LEB
- With precision comes additional information!
  - Measurement schemes and setups tailored to specific goals are very worthwhile





- Below Ni (Z=28) Fe/Co refractory
- N=Z=13, ^{26}Al (JYFL/ISOLDE)
- Tc → Pd (Z=43 – 46)
- Ta, Os, Re…..
- Actinides (Pu, Th, U….)
- Superheavies

Some concluding remarks

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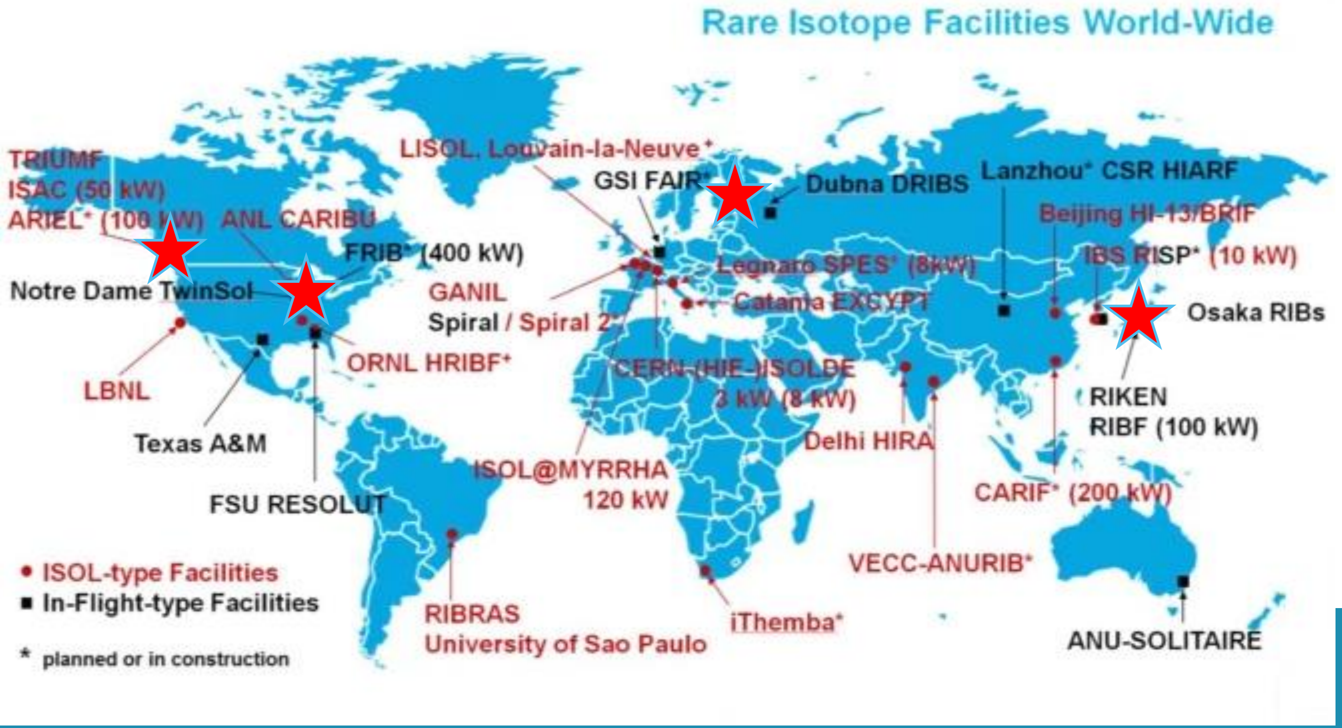
- With precision comes additional information!
  - Measurement schemes and setups tailored to specific goals are very worthwhile

- In-source laser spectroscopy
- Collinear laser spectroscopy
- Methods tailed to specific cases



Innovation and developments takes many many years, but it is a worthwhile investment.

Beyond atomic spectroscopy

Laser spectroscopy of molecules

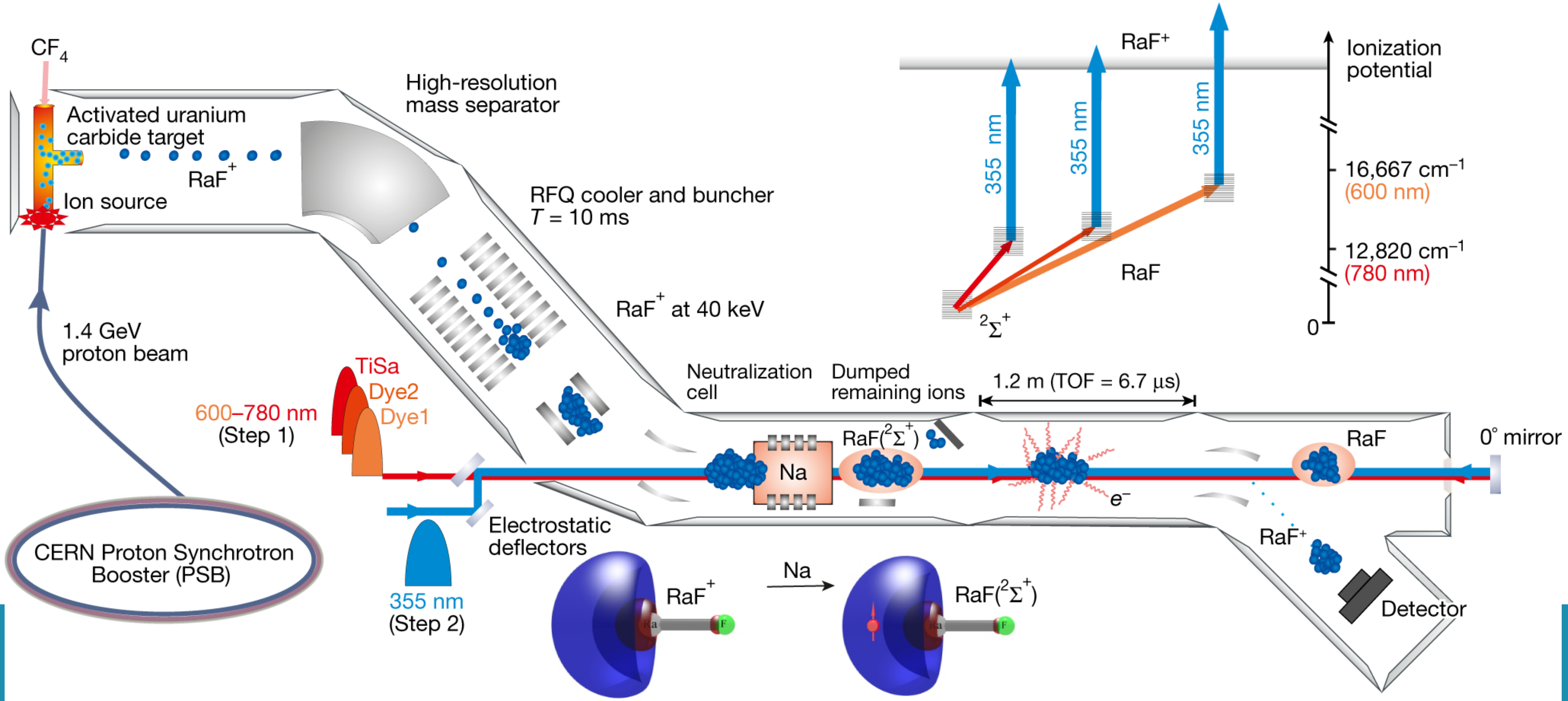
Scientific case

- Heavy radioactive molecules are potential probes for physics beyond the standard model
  - Parity and time reversal violating effects are significantly enhanced in molecules compared to atomic systems
  - (Octupole) deformation further enhances some of these effects
- Any precision experiment first needs reliable spectroscopic data.

- Note: molecular hyperfine structure also provided access to nuclear properties!
  - Route to explore nuclei with difficult chemistry?

Technique

- Collinear resonance ionization spectroscopy
  - Technical point: more flexibility in laser wavelengths, no wavelength-dependent detection
  - Use of multi-step laser excitation permits finding highly excited states as well



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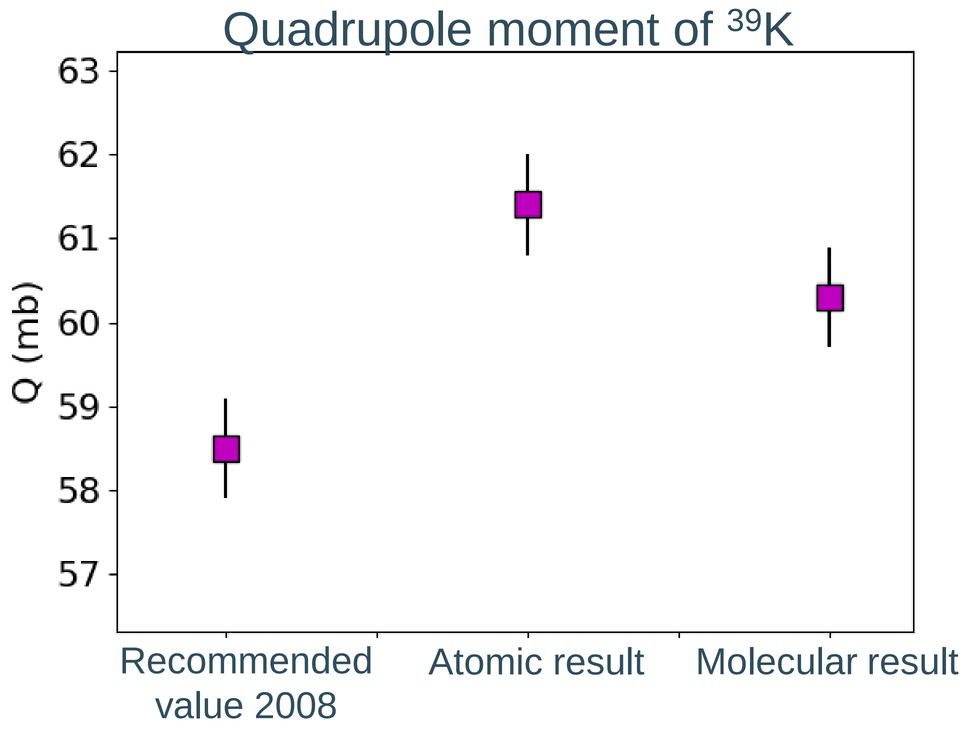
Results

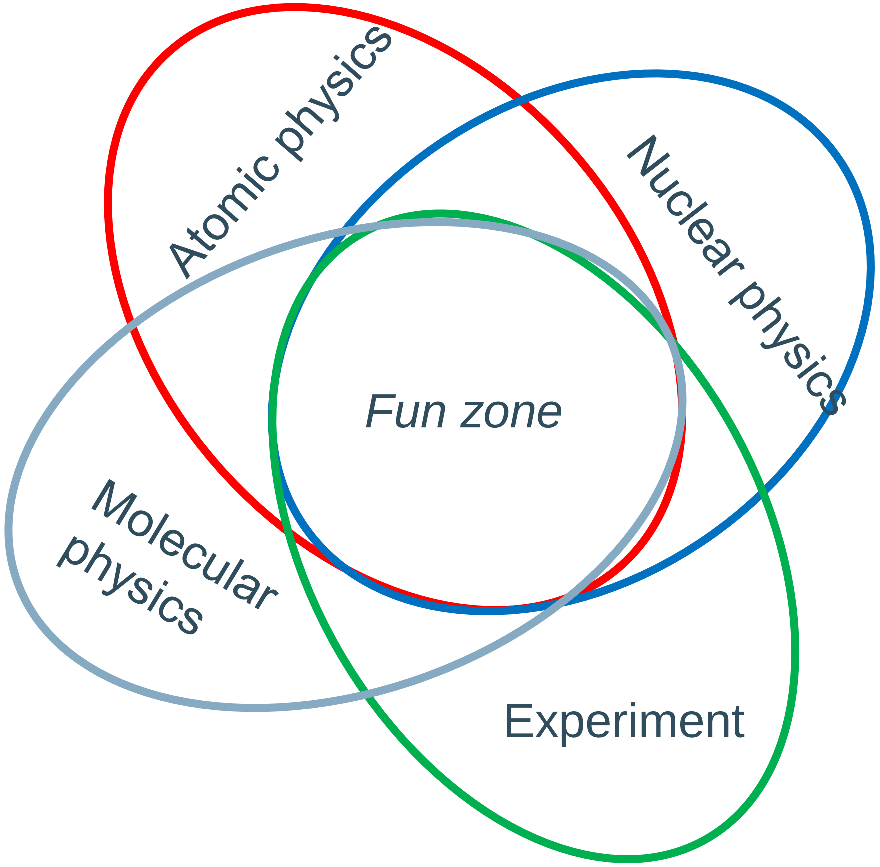
What's next in the region?

- Using **molecules** to access the nuclear quadrupole moment candidates: KI, KBr, KCl, KF – measured spectra!
- LOI accepted by ISOLDE INTC for KF

Theoretical calculations are crucial for quadrupole moments

Similar precision for both atomic and molecular calculations





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