JYVÄSKYLÄN YLIOPISTO
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Laser spectroscopy in radioactive ion beam facilities - Recent highlights

ISOL-FRANCE V, 21.3.2023

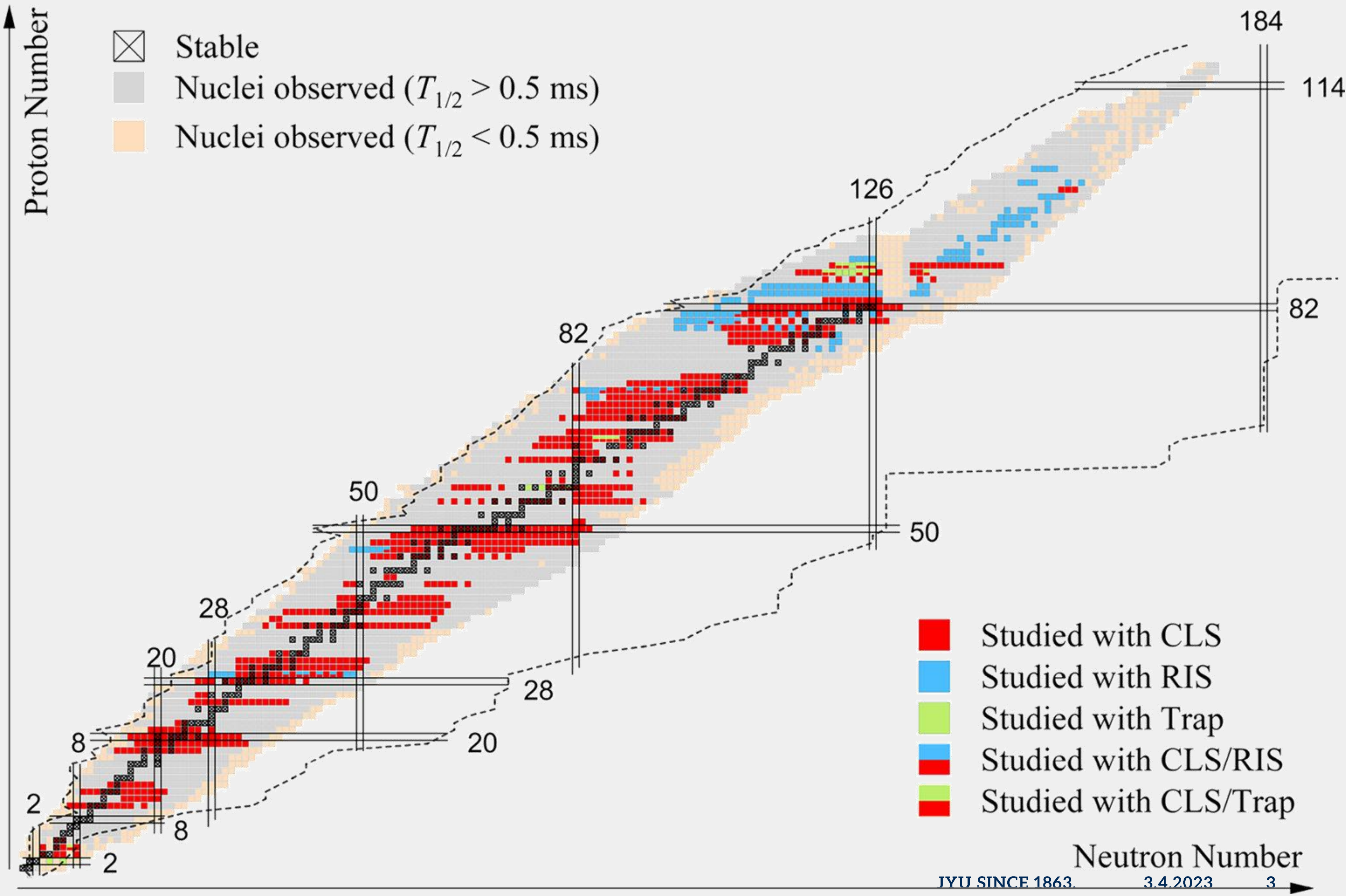
Mikael Reponen

Outline

- Laser spectroscopy status overview
- RIB Facilities with (planned) laser infrastructure
- Laser spectroscopy
  - Possibilities
  - Approaches, techniques
- Testing nuclear theories with optical measurements
- Near future outlook
  - New frontiers in the heavy region
  - Towards higher-resolution in-source RIS
  - Ultra-high precision frontier and radioactive molecules
- Final remarks

Status

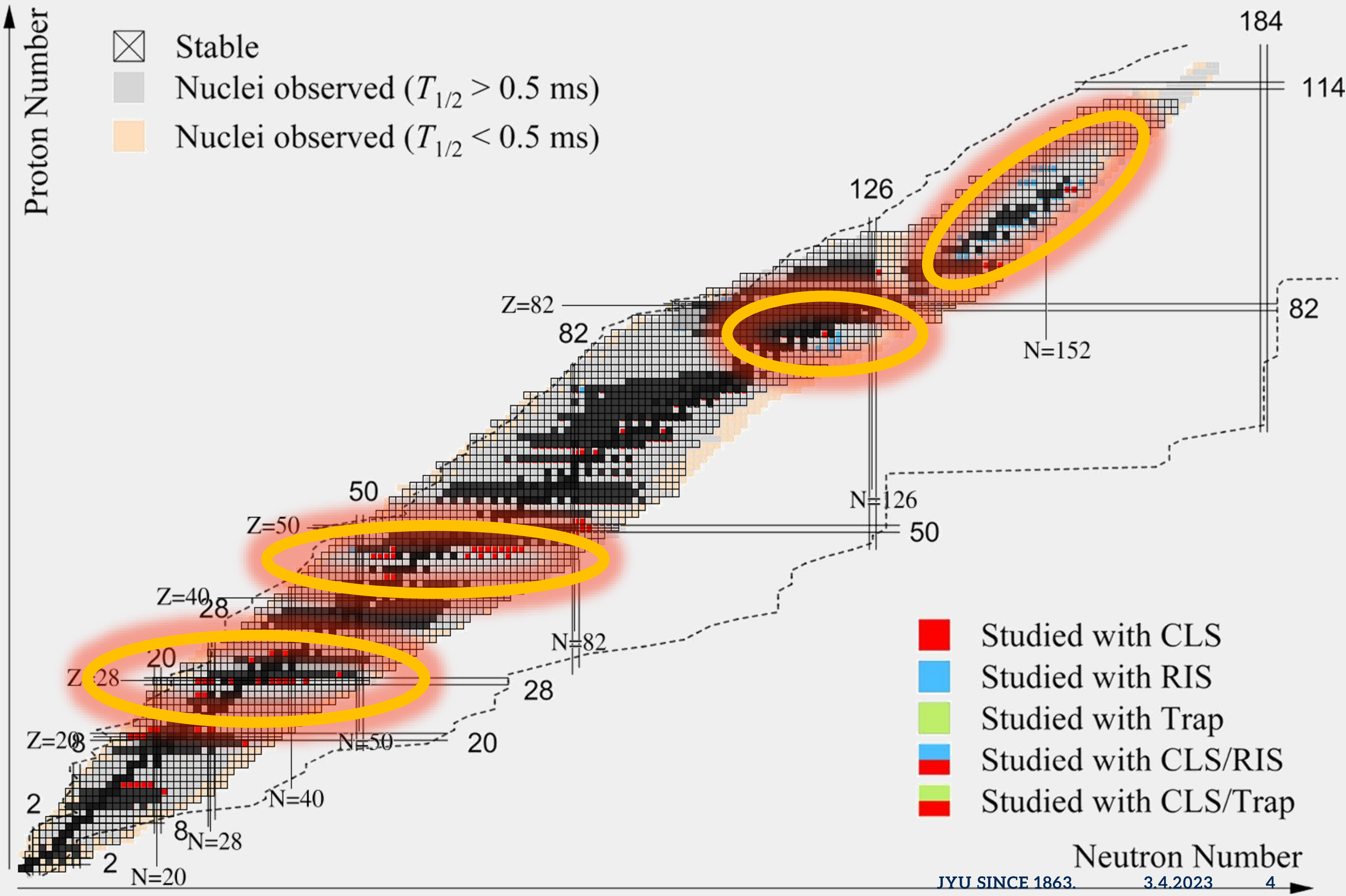
- Much progress since the 2016 review article:
  - Collinear RIS established
  - Laser spectroscopy of heavy elements and actinides
  - Testing nuclear theory with optical results
  - Radioactive molecules
  - Emerging on ultra-high precision and sensitivity techniques.



X.F. Yang et al., Prog. Part. Nuc. Phys, ***129****, (2023), 104005*

Status

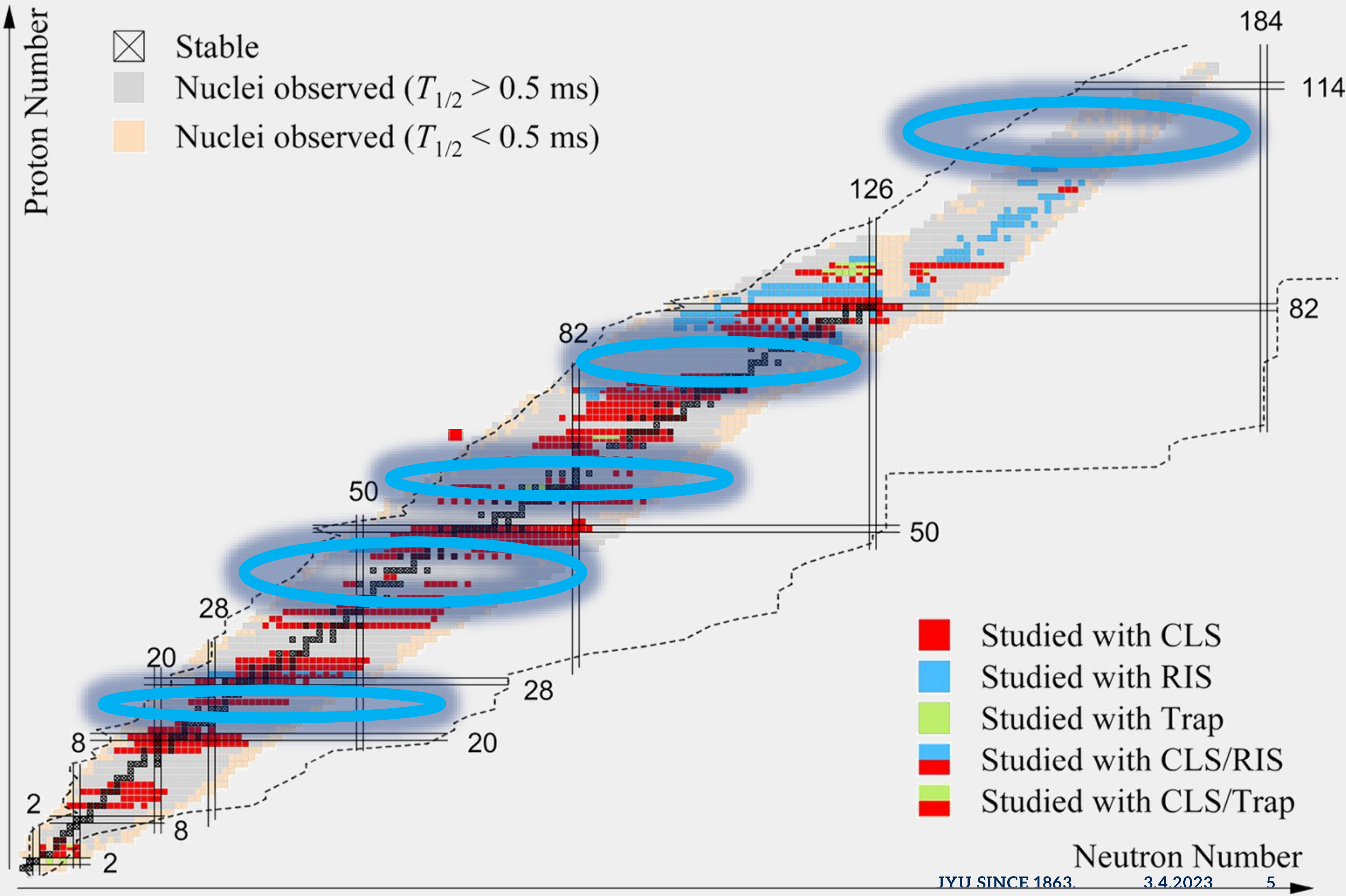
- Much progress since the 2016 review article:
  - Pd-Sn region
    - Access to *N*=50 and *N*=82
  - Ca and K chains
  - Work towards *N*=126
  - Actinides
    - Towards heavy elements



X.F. Yang et al., Prog. Part. Nuc. Phys, **129**, (2023), 104005 P. Campbell et al., Prog. Part. Nuc. Phys, **86**, (2016)

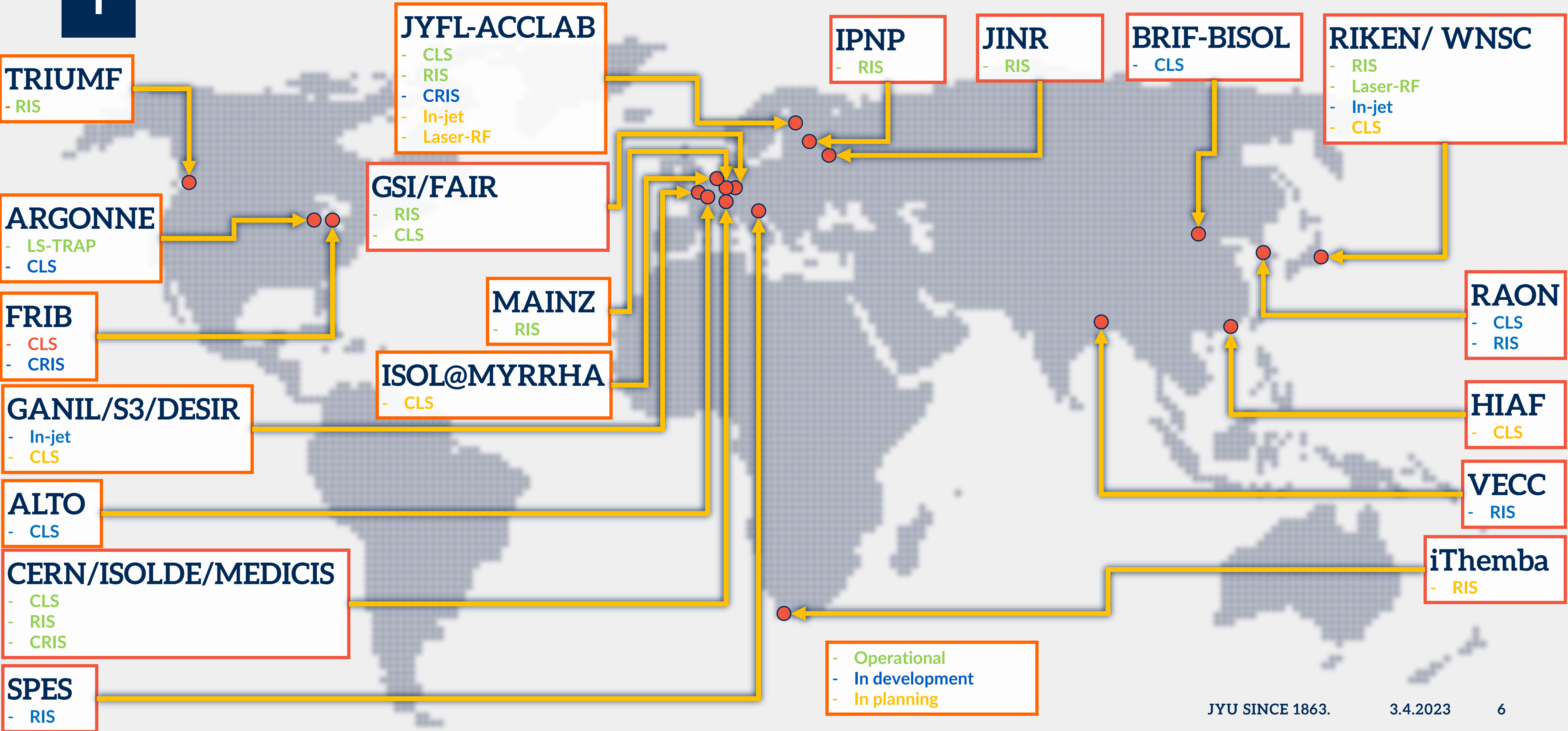
Status

- Gaps still persist in the nuclear chart
  - Below Ni (*Z*=28)
  - Tc (*Z*=43) to Pd (*Z*=46)
  - Rare –earth elements
  - Ir-Pt region
  - Superheavy elements

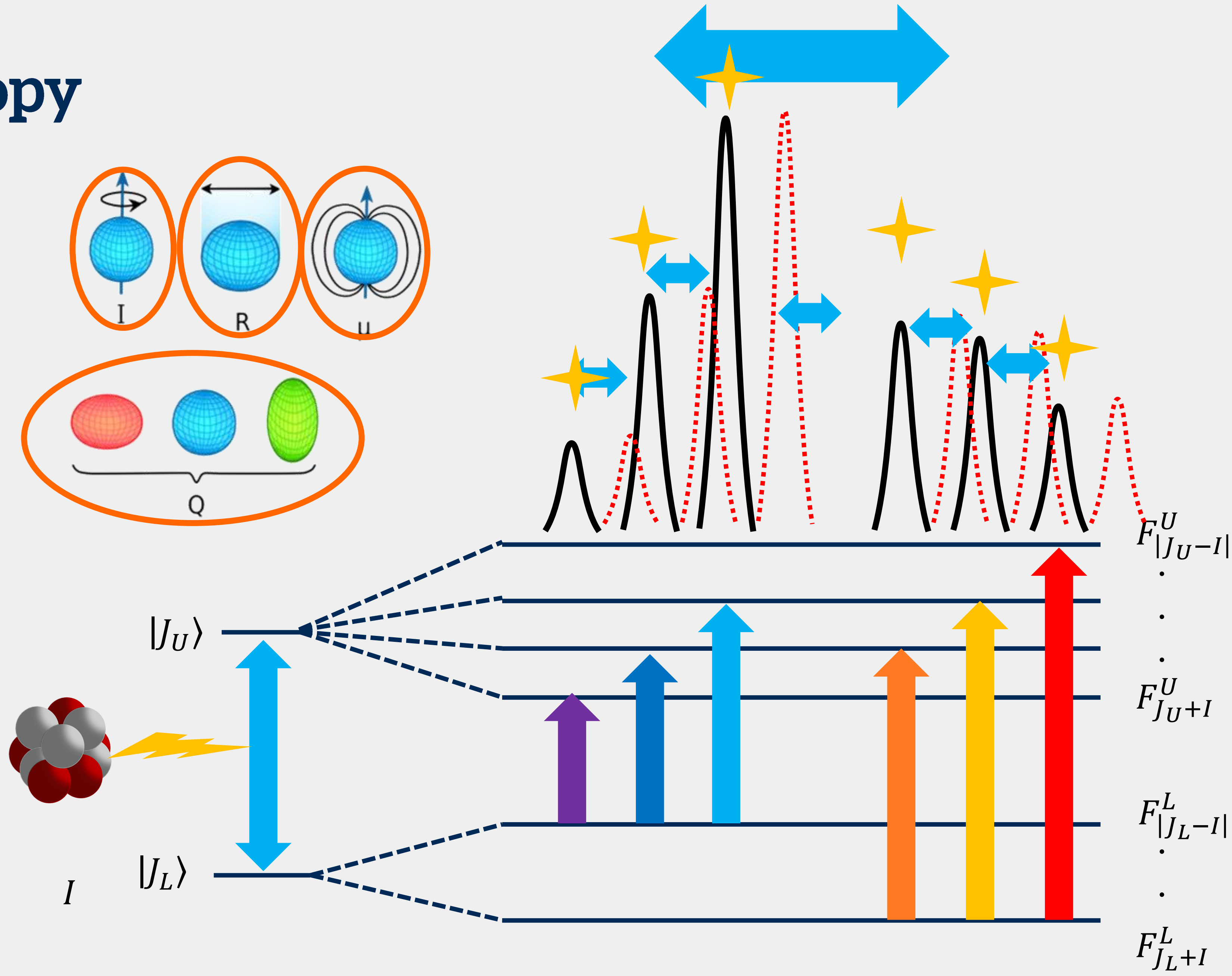


Y. F. Yang et al., Prog. Part. Nuc. Phys. **129** (2023) 104005

RIB Facilities with (planned) laser infrastructure



Laser spectroscopy



- Nuclear properties are mapped to the atomic structure.
  - 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
- **Spin, radii, moments be extracted without assuming a nuclear model!**

Laser spectroscopy
Atomic physics

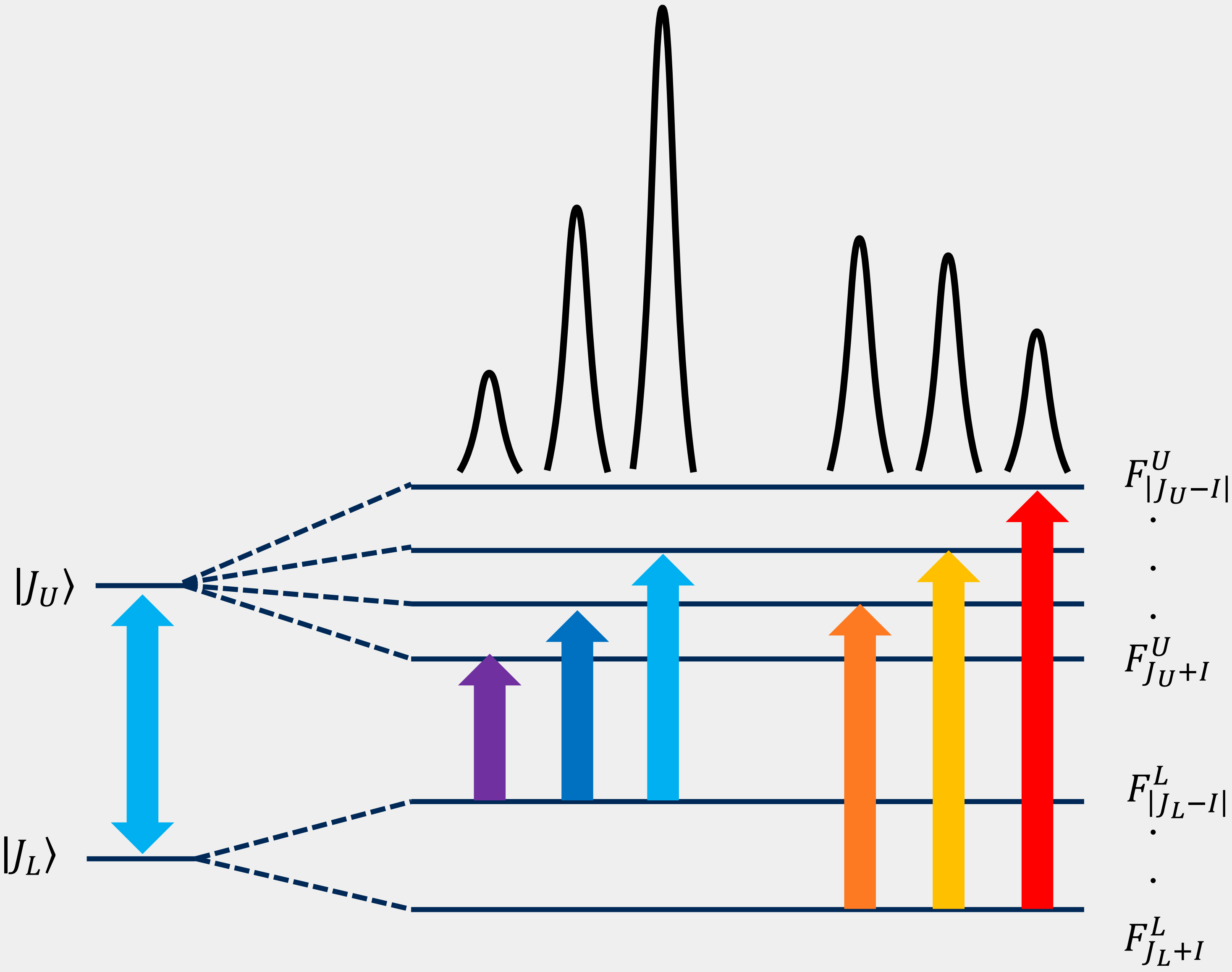
$$W^{(1)}_{F,J} \simeq W^{M1}_{F,J} + W^{E2}_{F,J},$$

$$W^{M1}_{F,J} = A_{hf}\mathbf{I}.\mathbf{J},$$

$$W^{E2}_{F,J} = B_{hf}\frac{3(\mathbf{I}.\mathbf{J})^2 + \frac{3}{2}(\mathbf{I}.\mathbf{J}) - I(I+1)J(J+1)}{2I(2I-1)J(2J-1)}$$

$$A_{hf} = g_I\mu_N\frac{\langle J||T^{(1)}_e||J\rangle}{\sqrt{J(J+1)(2J+1)}},$$

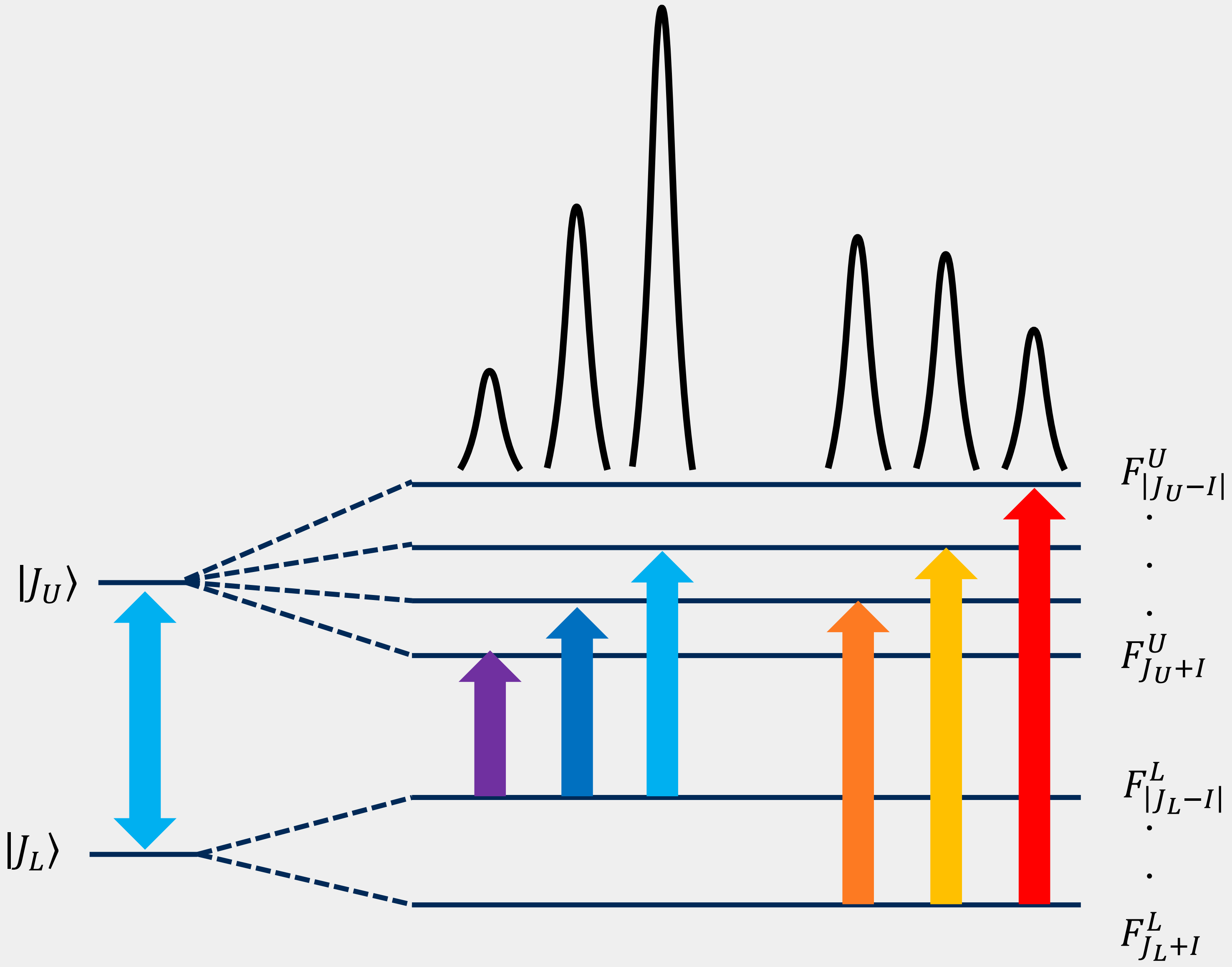
$$B_{hf} = 2Q\left[\frac{J(2J-1)}{(J+1)(2J+1)(2J+3)}\right]^{1/2}\langle J||T^{(2)}_e||J\rangle$$



Courtesy of K. Flanagan

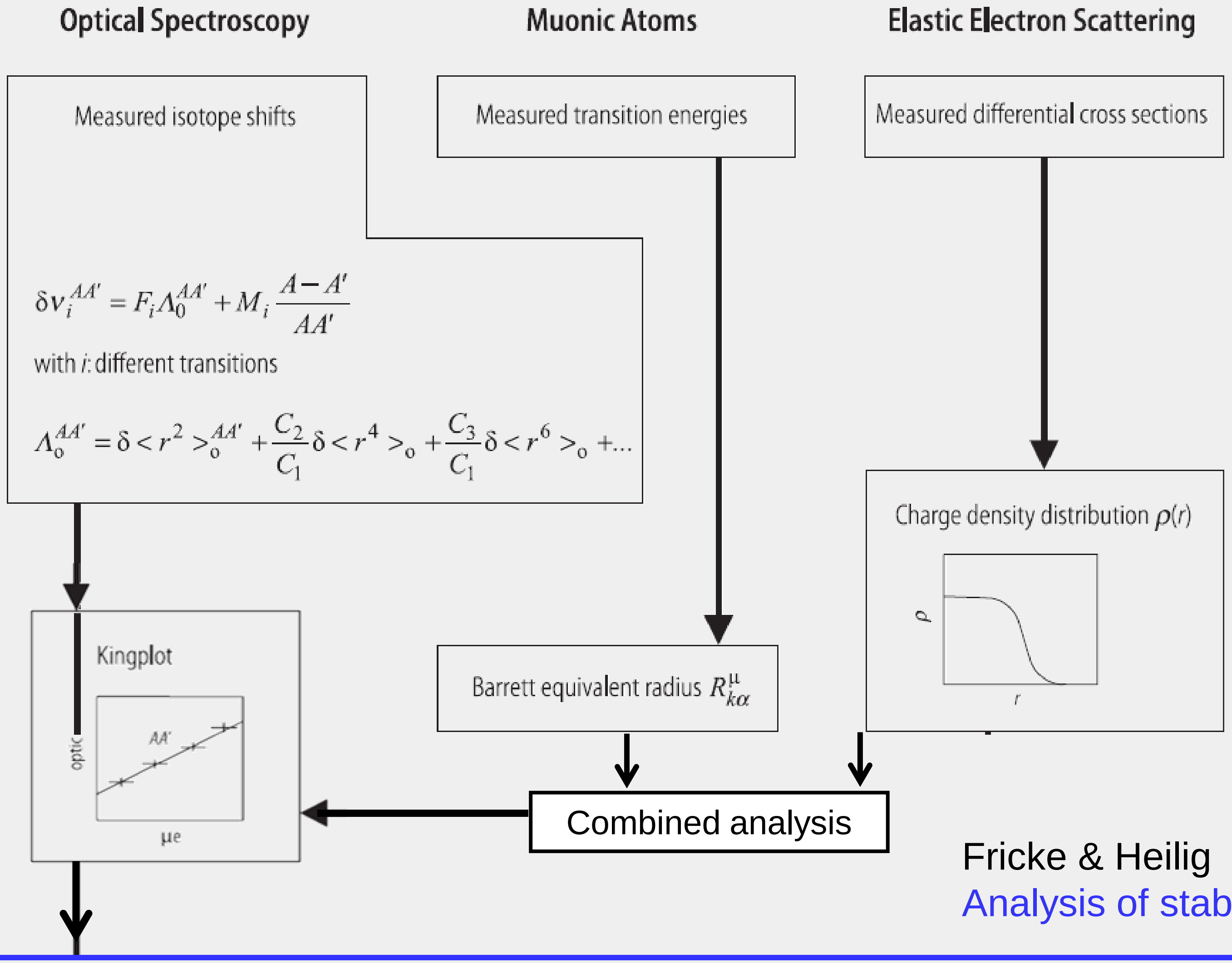
Laser spectroscopy technical

- Laser spectroscopy: Drive a transition and monitor a fluorescence/ion signal/etc. as a function of laser frequency
- Transition wavelengths: 10-2000 nm
  - Practically range: 200-1000 nm
- Isotope shifts, hyperfine splitting :
  - MHz to many GHz
- Transition @400 nm = 750 THz
- Laser spectroscopy inherits the richness of atomic structure.
  - Efficiency, applicability, etc. are element-specific.



Charge radii from isotope shift measurements

General approach



Fricke & Heilig *Nuclear Charge Radii* (Springer 2004)

Analysis of stable isotopes

Result: F_i and M_i providing $\delta<r^2>$ for all isotopes (including radioactive)

Laser Spectroscopy possibilities

Exotic nuclei at the limits of stability

Expected yields <<1 atom/second
Lifetimes <1s
Relatively large isobar contamination

Resolution:
Very little known => low resolution ok
As low as 10 MHz / precision ~ 100 kHz possible.

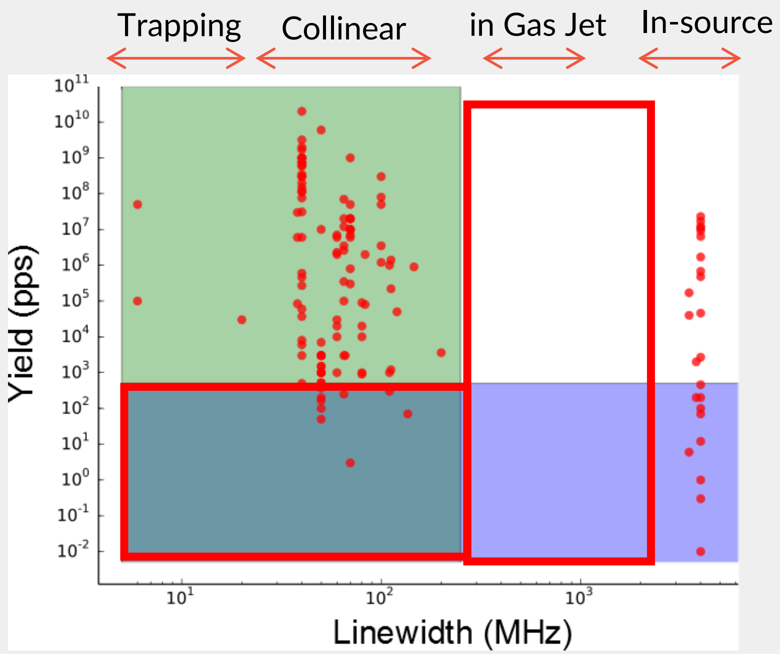
Technique :
Fast due to short half-lives
Highly selective due to isobars
Low yield requires a high sensitivity
Lower resolution can be acceptable

Near Stability Nuclei

Expected yields $>10^8$ atom/second
Lifetimes >>1s
High purity

Resolution/precision frontier:
kHz/mHz

Technique :
New physics requires high resolution sensitivity is not critical
The method can be slow



(Typical) Laser spectroscopy approaches

- In-source laser spectroscopy
  - Hot cavity
  - Gas cell
- Collinear laser spectroscopy
  - Fluorescence detection
  - Resonance ionization
  - Beta-NMR/collisional ionization/state-selective CEC
- Methods tailed to specific cases
  - Spectroscopy on trapped ion/atom
  - Techniques superseded by more modern ones…

Very efficient, low production rates
Low resolution, limited element choice
(New techniques to improve resolution)

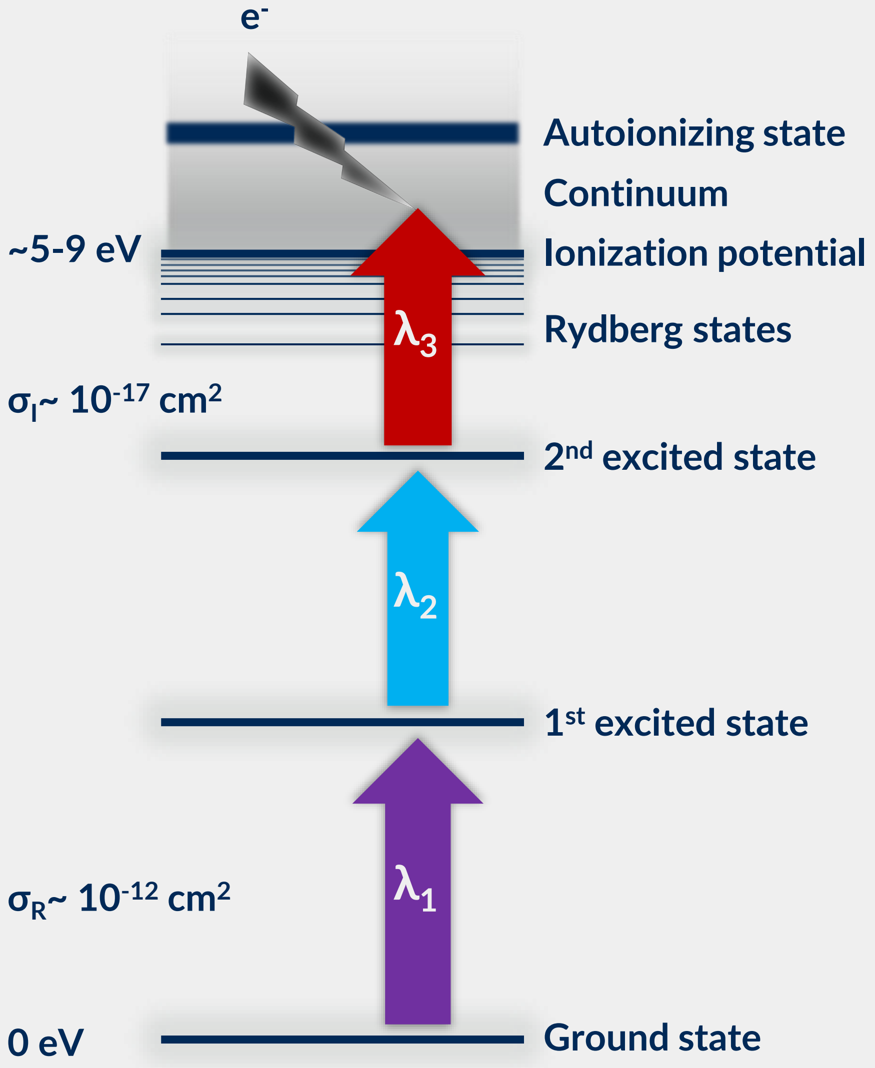
A bit less efficient
High resolution, wide element choice

More niche, specific cases/applications
Search for new physics

Courtesy of R. P. de Groote

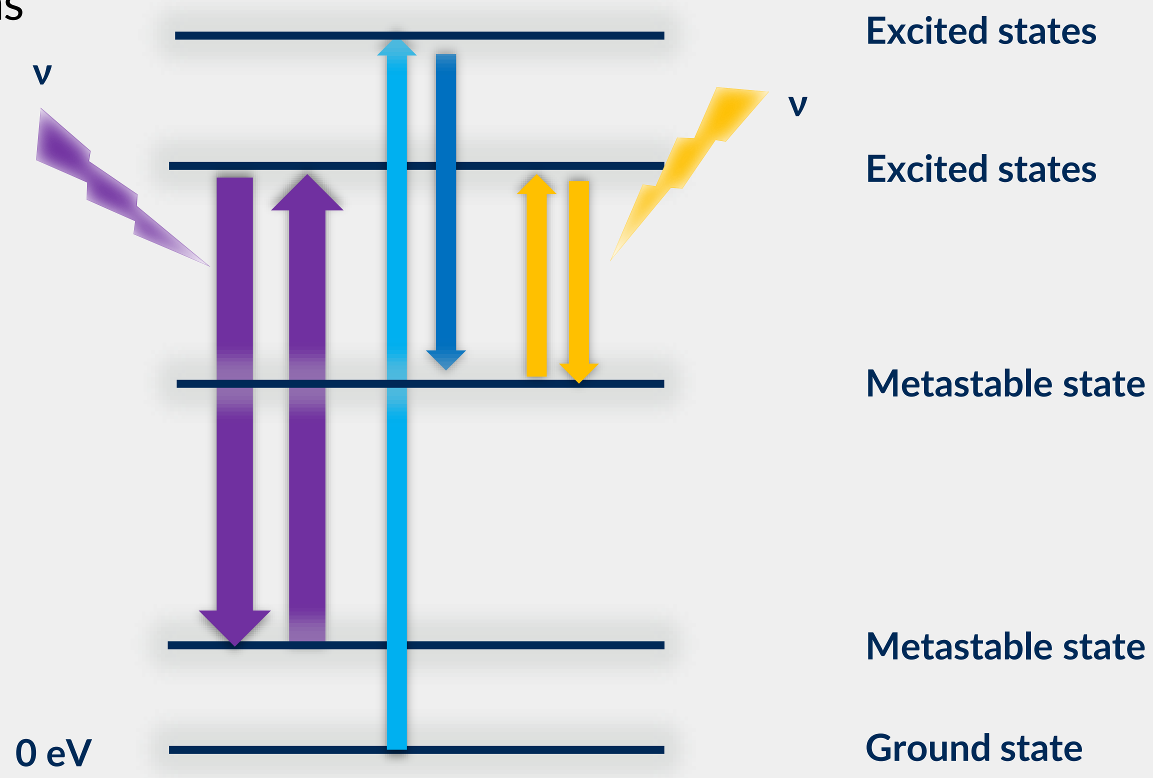
Method: Laser resonance ionization

- Each element have their unique atomic structure - “fingerprint”.
- Multiple laser beams overlapped with atoms to stepwise excite and ionize
  - Efficient! As high as >50%, typically a few %
- A great method for sensitive laser spectroscopy
  - Resolution depends highly environment dependent.



Method: Collinear laser spectroscopy

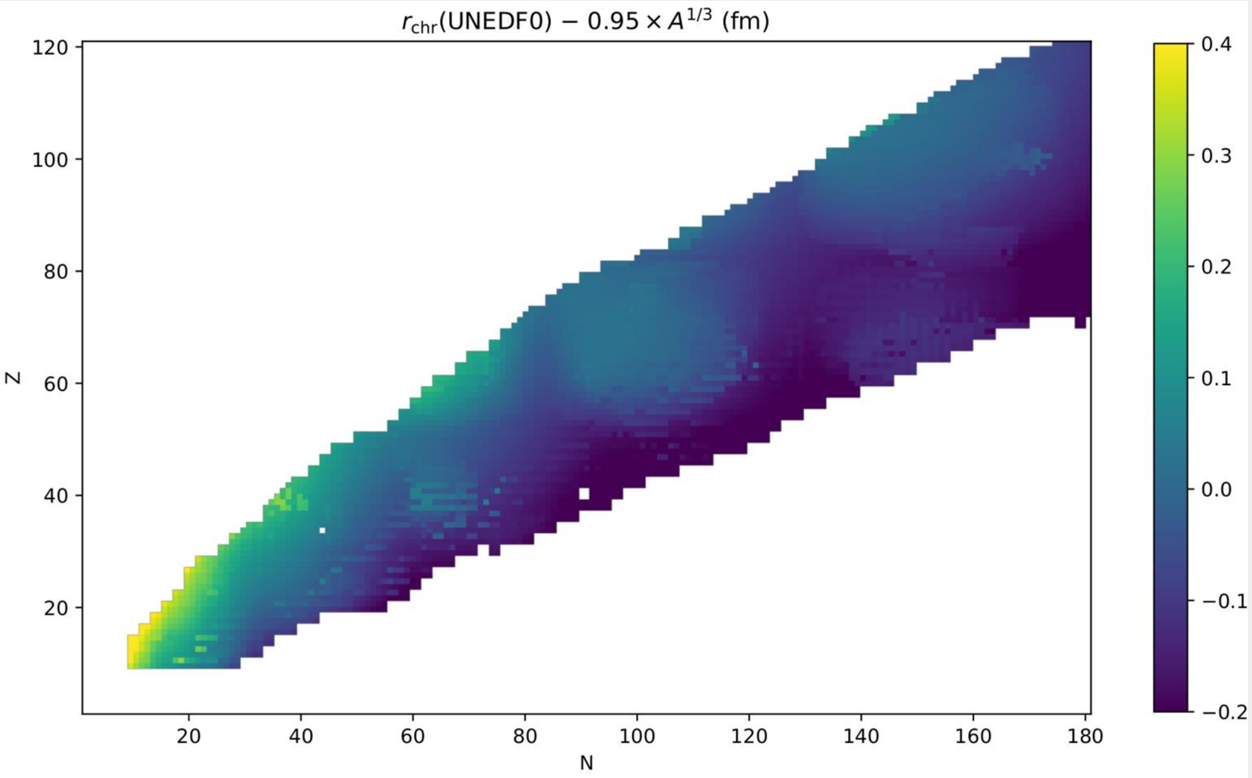
- In-flight laser spectroscopy with fast atomic or ionic beams
  - Typically, ~20-40 keV => Doppler compression!
  - CLS of confined ions
    - MR-ToF
    - Relativistic CLS in storage rings.
- Resolution down to the natural linewidth
- Typically, fluorescence detection
  - Either from a ground state or metastable state
    - Optical pumping to prepare a suitable state!
- Collinear RIS + ion detection a recent breakthrough.



Experiments + theory
Benchmarking models with optical measurements

Testing and developing nuclear theory
Nuclear charge radius from isotope shift measurements

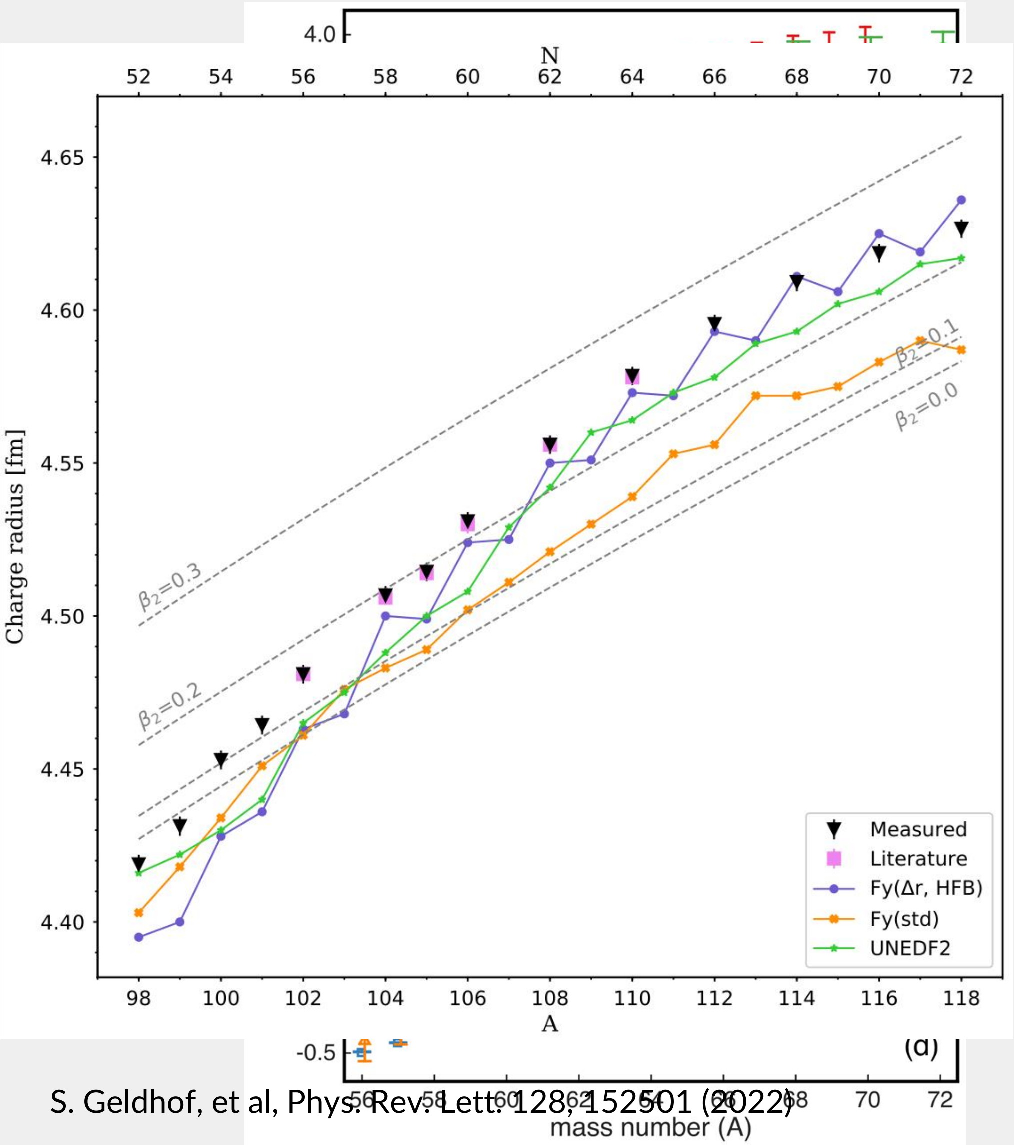
- Local variation in charge radii requires precise nuclear structure calculations
- Microscopical effects impact the radius:
  - shell structure
  - deformation
  - pairing
  - proximity to continuum, etc.
- For example: A sudden change in the systematics of the radius can signal a transition between, e.g., deformed and spherical systems.



Courtesy of M. Kortelainen

Recent DFT test cases
DFT: Nuclear charge radius

- Compared to Skyrme EDF, Fayans EDF has a more complicated structure.
  - Includes density dependence and gradient term.
- Recent Fayans functionals add gradient term on pairing energy density.
  - To reproduces Ca chain radii OES
- Recently applied for example in
  - Ni
  - Pd
- Few more charge-radii cases to follow…



S. Geldhof, et al, Phys. Rev. Lett. 128, 152501 (2022)

S. Malbrunot-Ettenauer et al, Phys. Rev. Lett. 128, 022502 (2022)

S. A. Fayans et a *Nucl. Phys. A*, vol. 676, no. 1, pp. 49–119, Aug. 2000,
P.-G. Reinhard and W. Nazarewicz, *Phys. Rev. C*, vol. 93, no. 5, p. 051303, May 2016,

Courtesy of M. Kortelainen

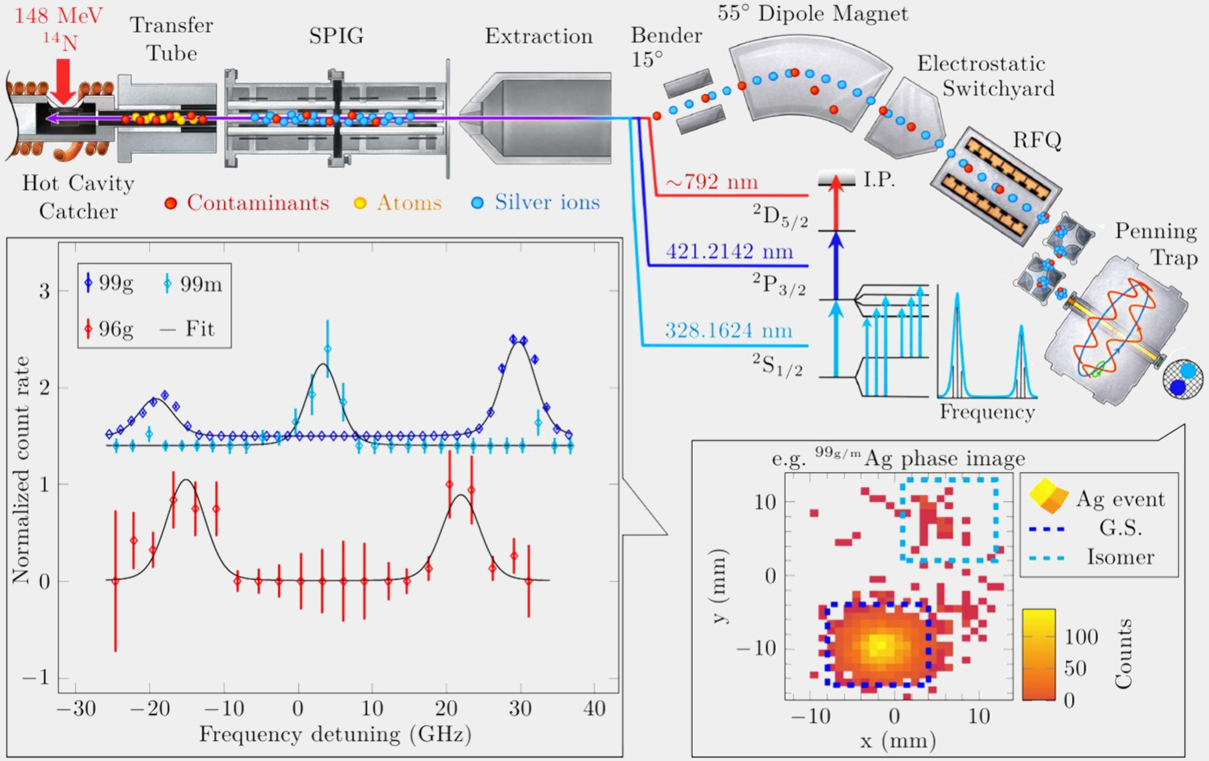
Challenging nuclear theory
Little explored regions of the nuclear chart

- In-source RIS in the immediate vicinity of ^{100}Sn
  - Crossing the *N*=50 in the
  - Approaching *N*=Z line
- Signal rates of 1 per 5 minutes

Lit. Optical Data: Low-res. | High-res.

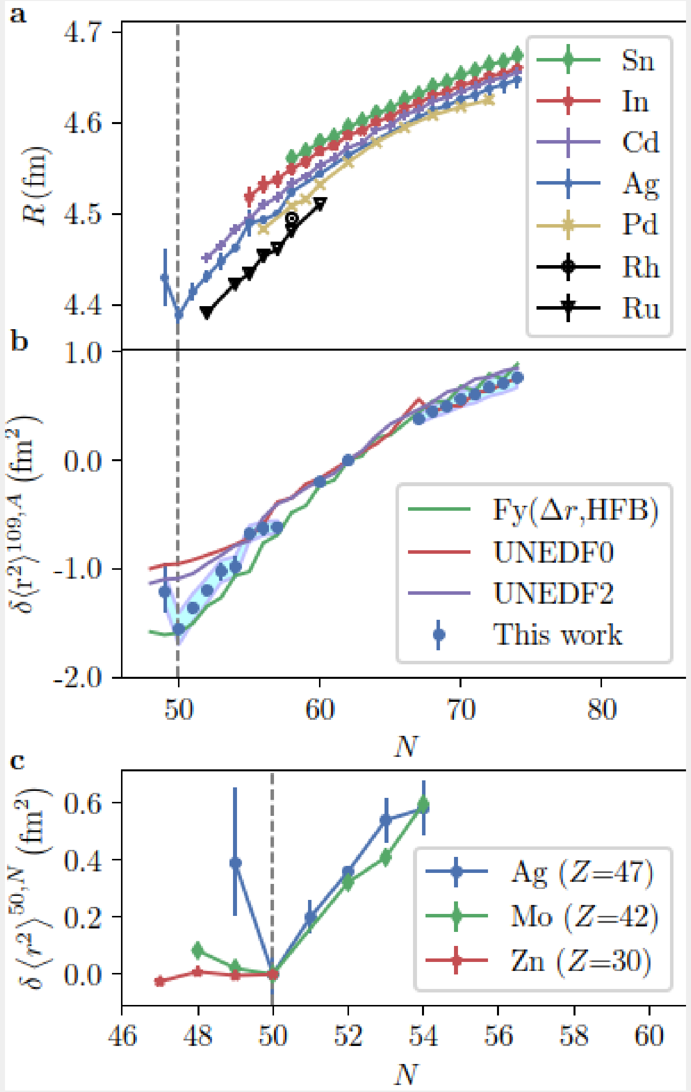
				Z=50	Sn 99	Sn 100 1.16 s	Sn 101 1.97 s	Sn 102 3.8 s	Sn 103 7.0 s	Sn 104 20.8 s	Sn 105 34 s
			In 96	In 97	In 98 37 ms	In 99 3.1 s	In 100 5.83 s	In 101 15.1 s	In 102 23.3 s	In 103 60 s	In 104 1.80 m
		Cd 94	Cd 95	Cd 96 880 ms	Cd 97 1.10 s	Cd 98 9.2 s	Cd 99 16 s	Cd 100 49.1 s	Cd 101 1.36 m	Cd 102 5.5 m	Cd 103 7.3 m
	Ag 92	Ag 93	Ag 94 37 ms	Ag 95 1.76 s	Ag 96 4.44 s	Ag 97 25.5 s	Ag 98 47.5 s	Ag 99 2.07 m	Ag 100 2.01 m	Ag 101 11.1 m	Ag 102 12.9 m
	Pd 91	Pd 92 1.1 s	Pd 93 1.15 s	Pd 94 9.0 s	Pd 95 7.5 s	Pd 96 122 s	Pd 97 3.10 m	Pd 98 17.7 m	Pd 99 21.4 m	Pd 100 3.63 d	Pd 101 8.47 h

N=50



M. Reponen et al., Nat Comm, **12**, (2021), 4596

Challenging nuclear theory
Little explored regions of the nuclear chart



- Spectra for $^{104\text{-}96}$Ag obtained using ^{14}N(^{92}Mo, 2pxn) Ag
  - Data ^{96,95}Ag using ^{40}Ca($^{58/60}$Ni, pxn) Ag
- *Virtually* a background-free measurements
- Very sharp kink observed at *N*=50 – beyond current DFT models. More data needed to refine error bar.
  - Points towards a need for symmetry-restored multi-reference EDF
- Magnetic moments near *N*=*Z* an important test for theory

M. Reponen et al., Nat Comm, 12, (2021), 4596

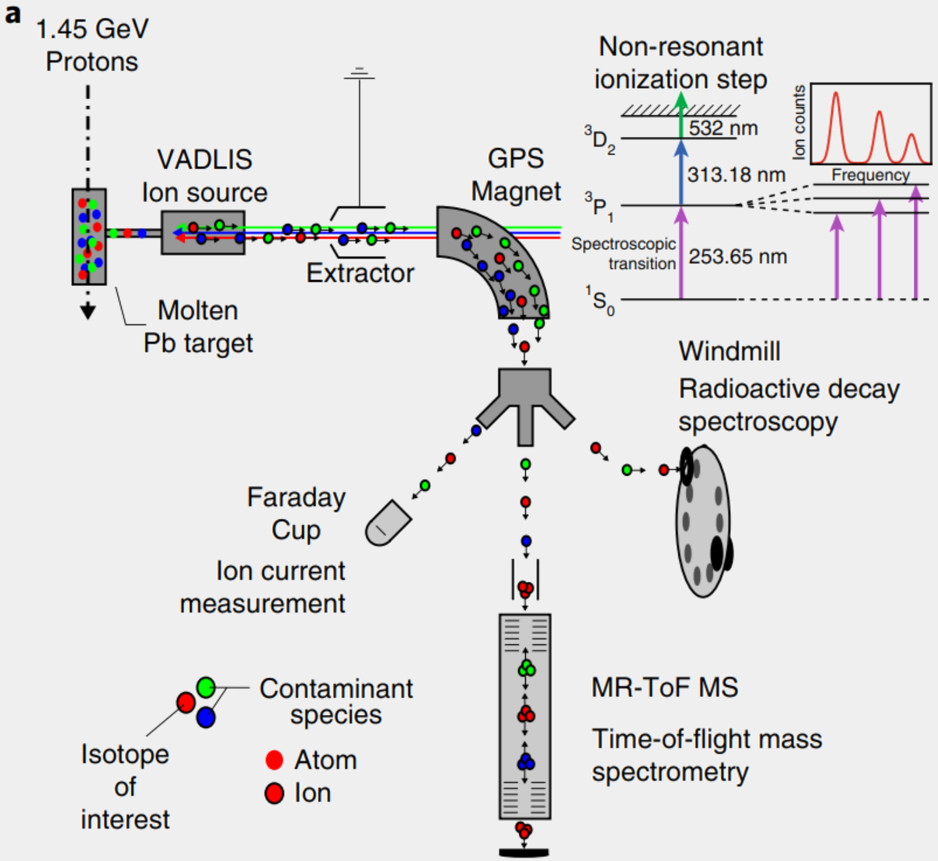
Challenging nuclear theory
Exotic isotopes with minute yields

RILIS:

- 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

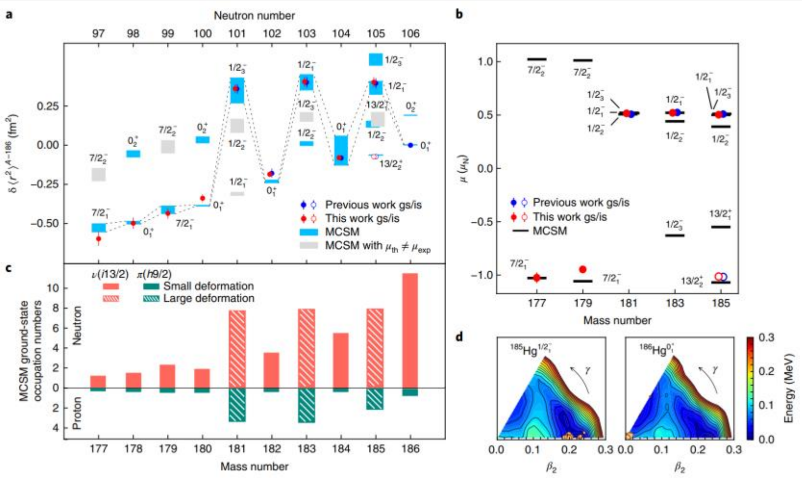
  - **FLEXIBILITY!** Tailor the detection to the isotope and beam at hand

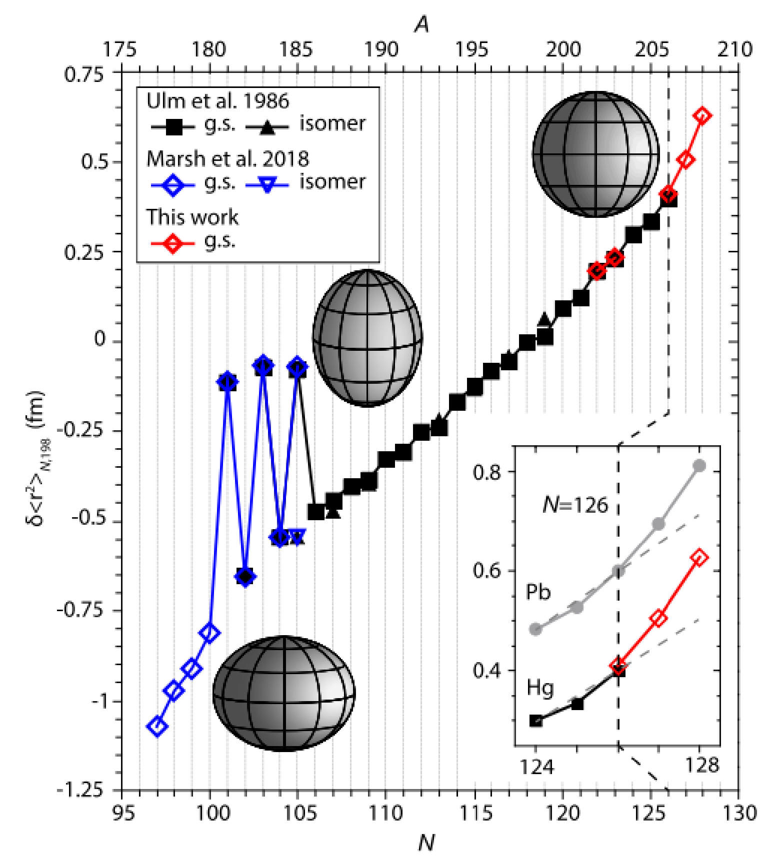


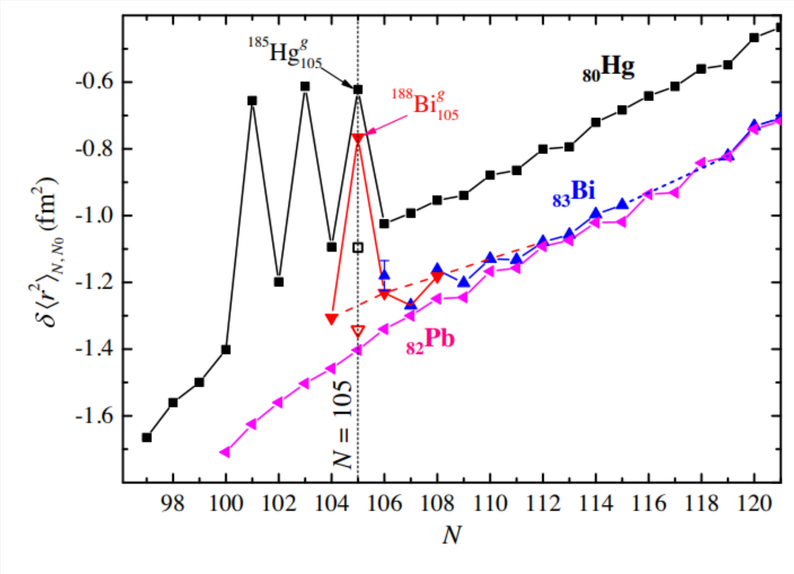
Marsh, B.A., Day Goodacre, T., Sels, S. *et al.* Characterization of the shape-staggering effect in mercury nuclei. *Nature Phys* **14,** 1163–1167 (2018)
Barzakh, A., *et al.* Large Shape Staggering in Neutron-Deficient Bi isotopes, Phys. Rev. Lett. 127, 192501 (2021)

Challenging nuclear theory
Exotic isotopes with minute yields

- Shape staggering in **Hg** and **Bi**
  - Significant challenge for nuclear theory
  - Magnetic moments are key to pin down nuclear configuration to aid the interpretation!







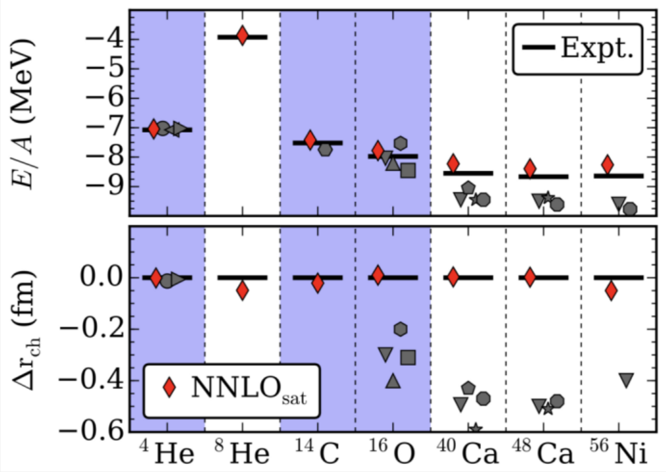
Marsh, B.A., Day Goodacre, T., Sels, S. *et al.* Characterization of the shape-staggering effect in mercury nuclei. *Nat. Phys* **14,** 1163–1167 (2018)
T. Day Goodacre, PRL **126**, 032502 (2021)
Barzakh, A., *et al.* Large Shape Staggering in Neutron-Deficient Bi isotopes, Phys. Rev. Lett. **127**, 192501 (2021)

Development of nuclear interactions

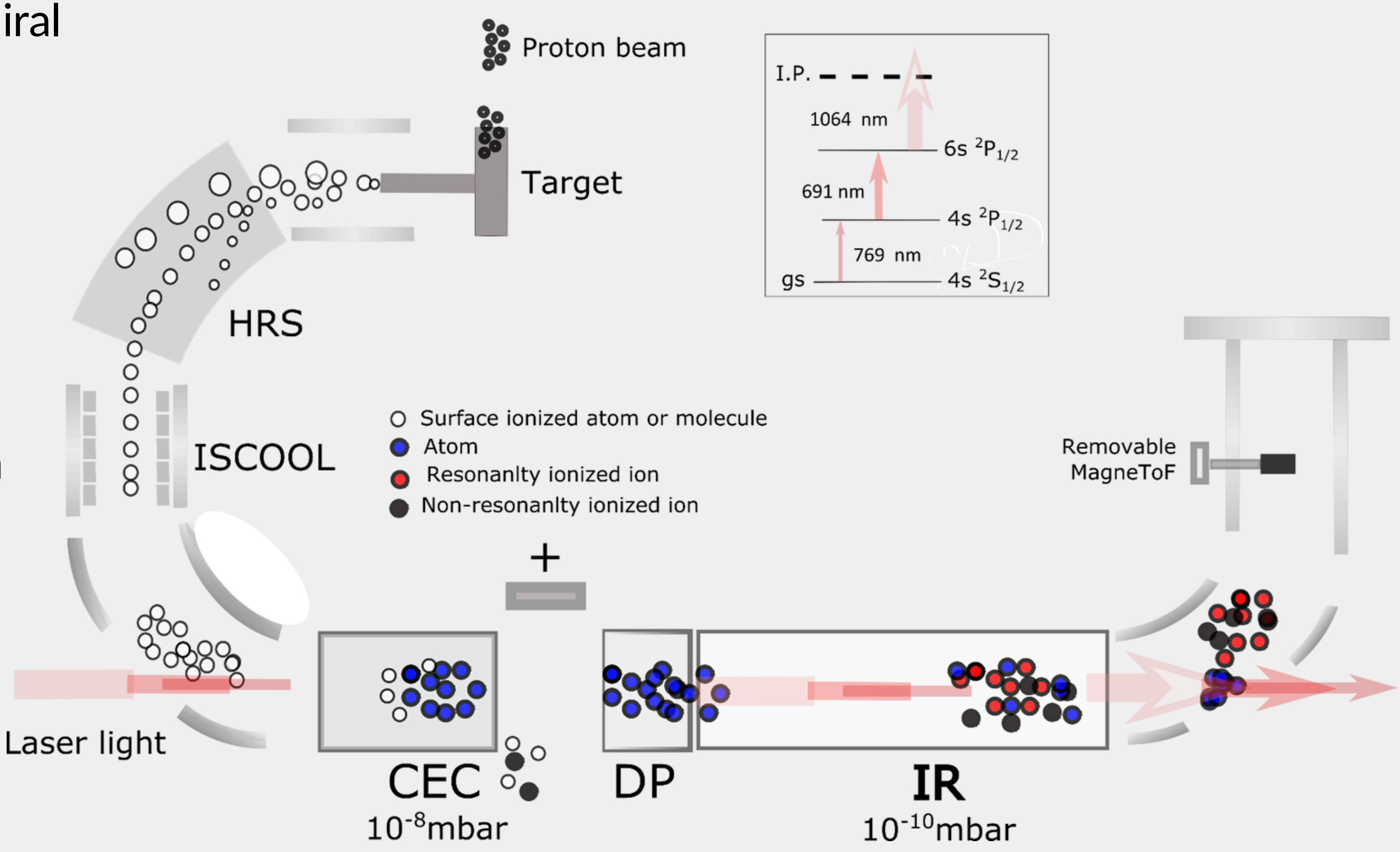
Pushing methods to short lifetimes and low yields with collinear methods

Study of K as a test to DFT and $NNLO_{sat/go}$ Chiral Effective Field Theory

- CRIS at ISOLDE:
  - Collinear = doppler-free
  - RIS = versatile, efficient
- Challenges:
  - Low yields, Large isobaric contamination



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

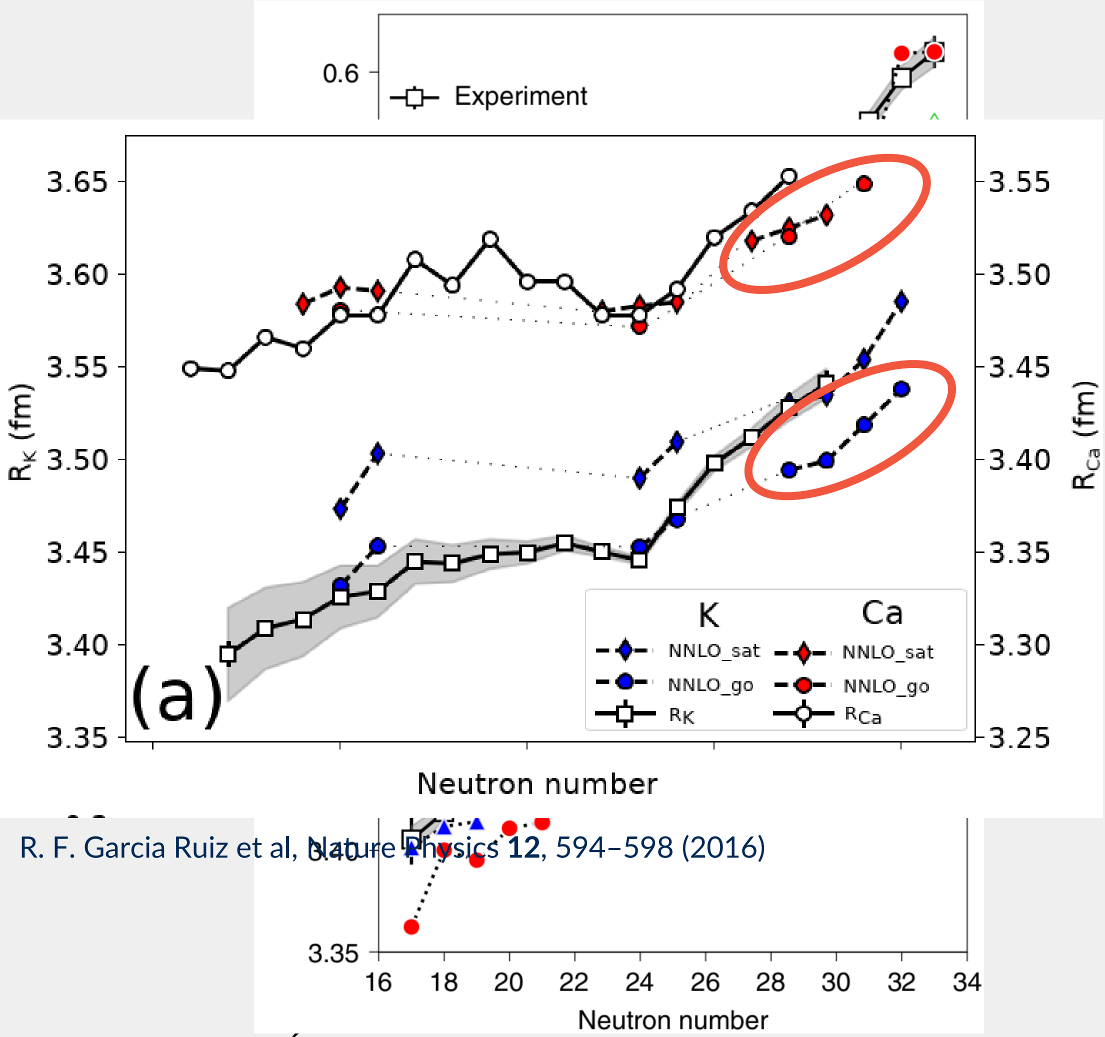


Courtesy of R. P. de Groote

Development of nuclear interactions

Pushing methods to short lifetimes and low yields with collinear methods

- Theoretical calculations
  - DFT framework (Fy(Δr,HFB) EDF),
  - ab-initio coupled-cluster calculations with $NNLO_{sat}$ and $\Delta NNLO_{GO}$
- Fy(Δr,HFB) reproduces the kink and trend after N = 28
  - However, too strong odd-even effect.
- Ab-initio results show more realistic odd-even effect, but the trend after N = 28 is not so well reproduced
  - No signature of a magic shell gap at N = 32
- $NNLO_{sat}$ and newer $NNLO_{GO}$ may work better close to stability
  - Also observed in Ca (CLS measurement)



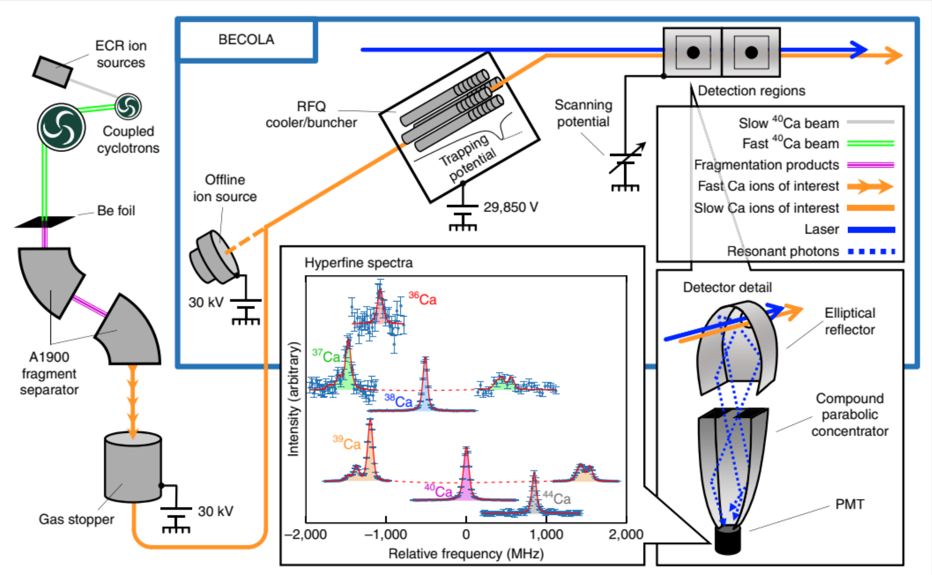
R. F. Garcia Ruiz et al, Nature Physics 12, 594–598 (2016)

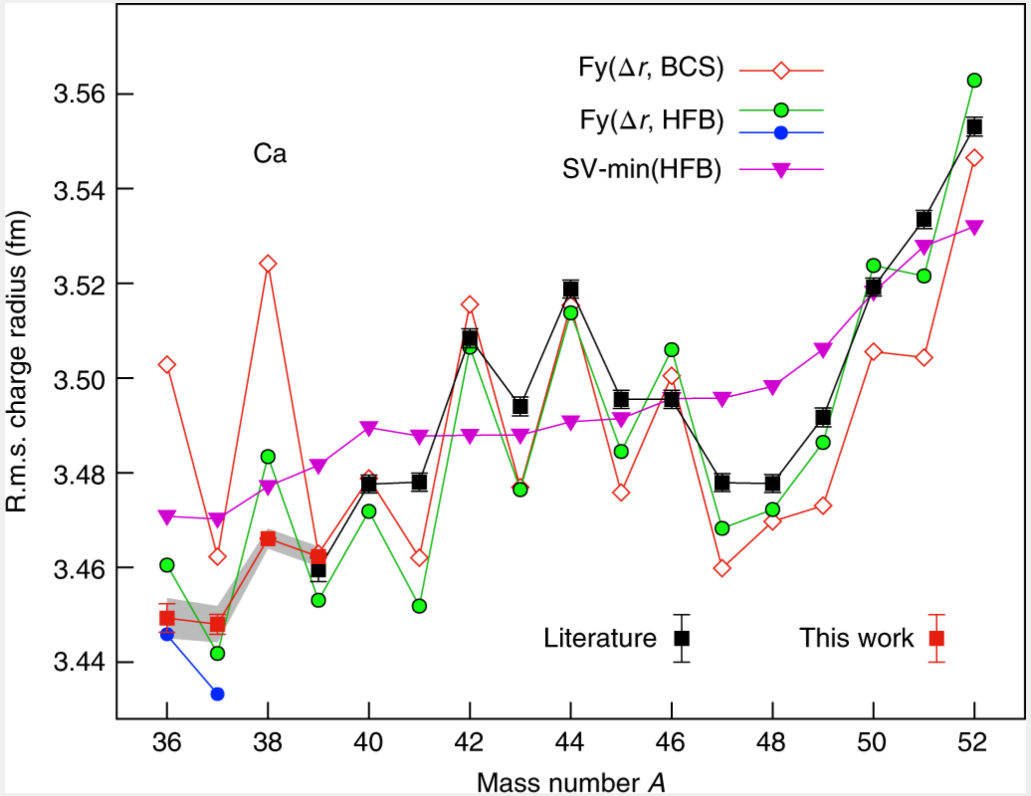
Á. Koszorús et al, Nature Phys. 17, 439 (2021)

Courtesy of R. P. de Groote

Nuclei at the limits of existence
Continuum effects

- Proton-rich Ca:
  - Treatment of proton superfluid correlations in the presence of low-lying continuum states is vital.
  - Charge radii of very proton-rich nuclei
    - Important for pinning down pairing interactions

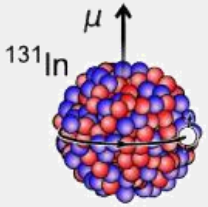


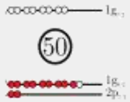


A. J. Miller et al, Nature Physics **15**, 432–436 (2019)

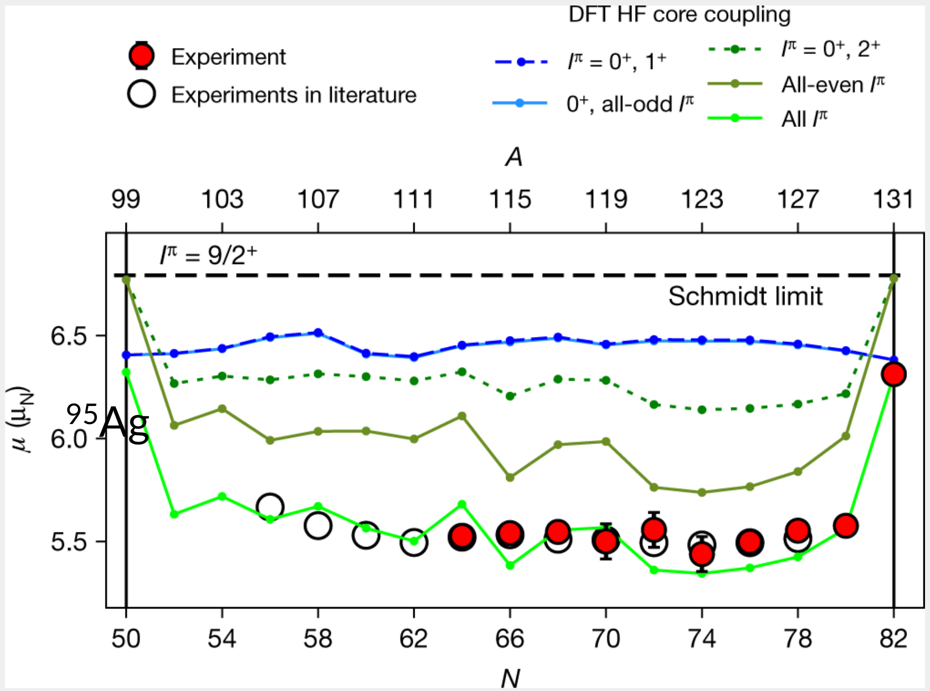
Testing nuclear theory
Magnetic moments

- Magnetic moments are a very sensitive probe to nuclear structure
- Indium isotopes were a textbook case on single particle behavior.
- An abrupt change observed at *N*=48,
  - Time-symmetry-breaking mean fields is essential in DFT
  - No need for effective
- More data required near the indium chain
  - Data on silver isotopes under analysis past the *N*=50 shell closure





Odd-*N* Ag



A. Vernon, Nature **607**, 260–265 (2022)

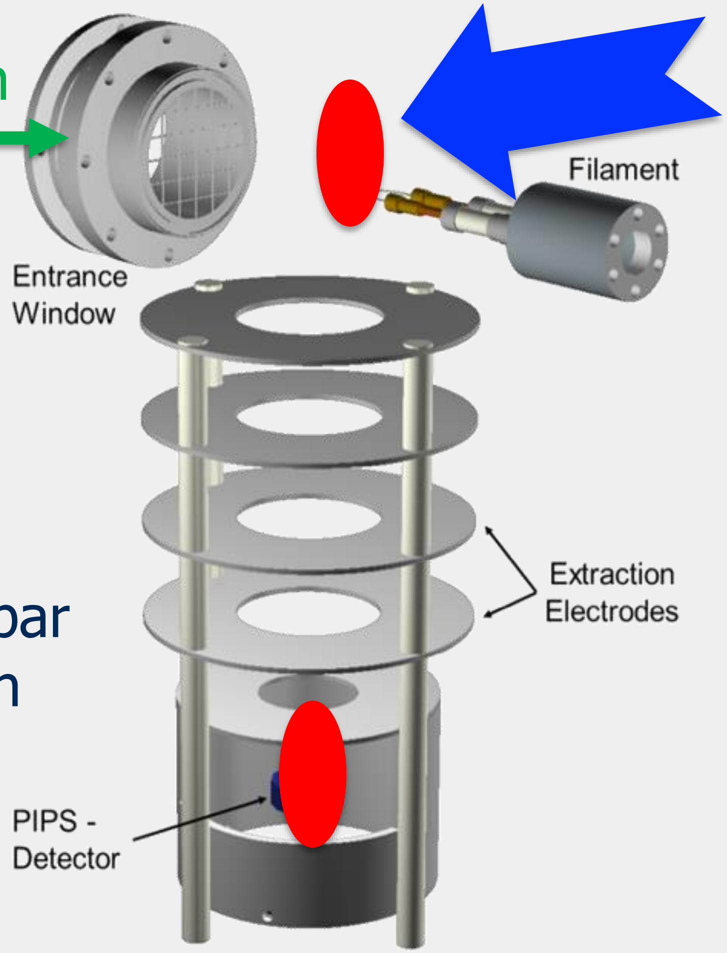
Near future outlook:
New frontiers in the heavy region

Reaching new frontiers
Laser Spectroscopy in heavy elements

RADRIS method tailored to actinides produced by fusion with lowest rates:

- Nobelium first laser spectroscopy beyond *Z*=100
  - yield as low as 0.05 atoms / second
- Isotope shift allowed determining changes in mean-square charge radii around *N* = 152
- Magnetic dipole and electric quadrupole moment of ^{253,255}No obtained from hyperfine splitting

Incoming beam



100 mbar argon

LASER IONISATION AND SPECTROSCOPY OF ACTINIDES

H. Backe et al. Eur. Phys. J. D, 45 (1) (2007), 99
F. Lautenschläger et al. Nucl. Instrum. Meth. B, 383 (2016),115

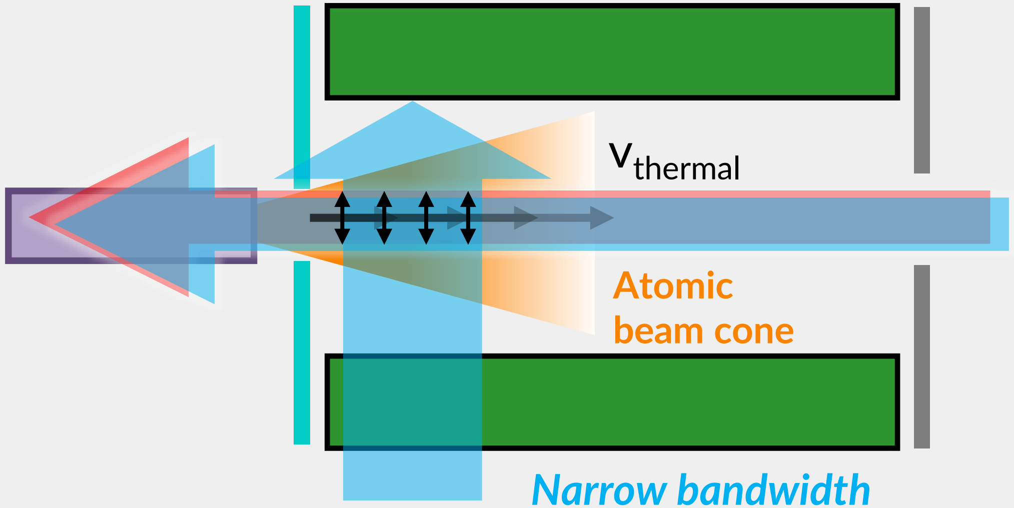
Courtesy of M. Block

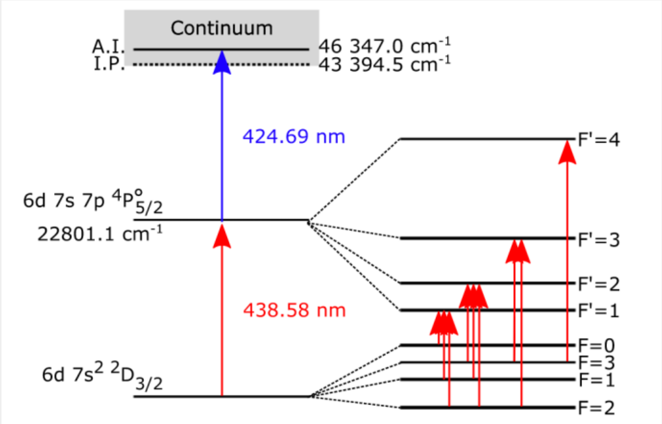
Near future outlook:

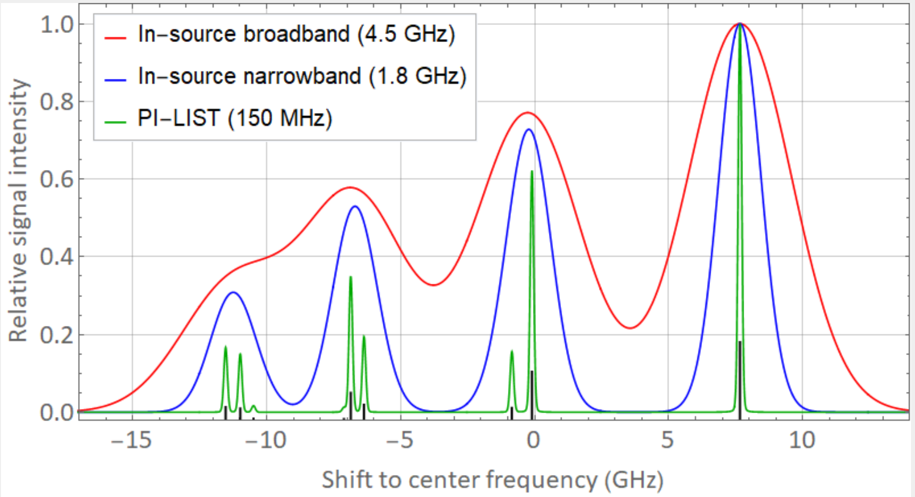
Towards higher-resolution in-source RIS

PI-LIST

Perpendicularly Illuminated Laser Ion Source and Trap







"Sub-Doppler" hot cavity in-source spectroscopy

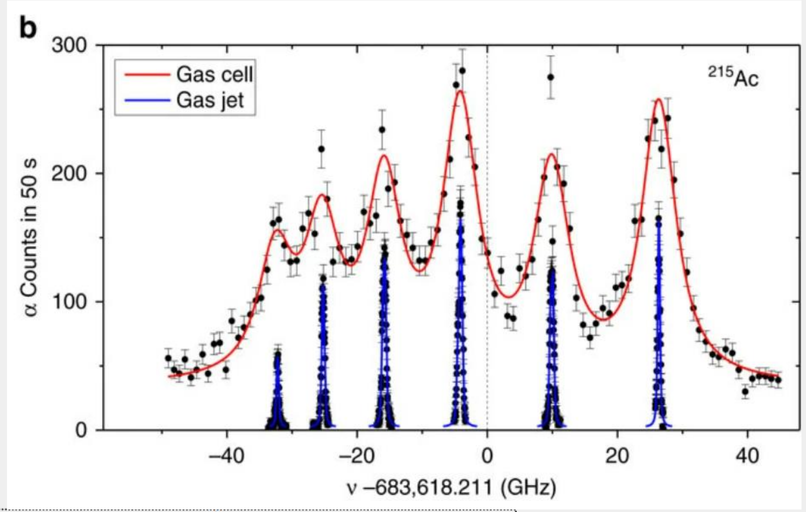
- Crossed atom beam / laser geometry in LIST structure
- Selection of reduced Doppler ensemble in laser intersection volume
- Suitable narrow-band laser
- Resolution improvement by >1 order of magnitude

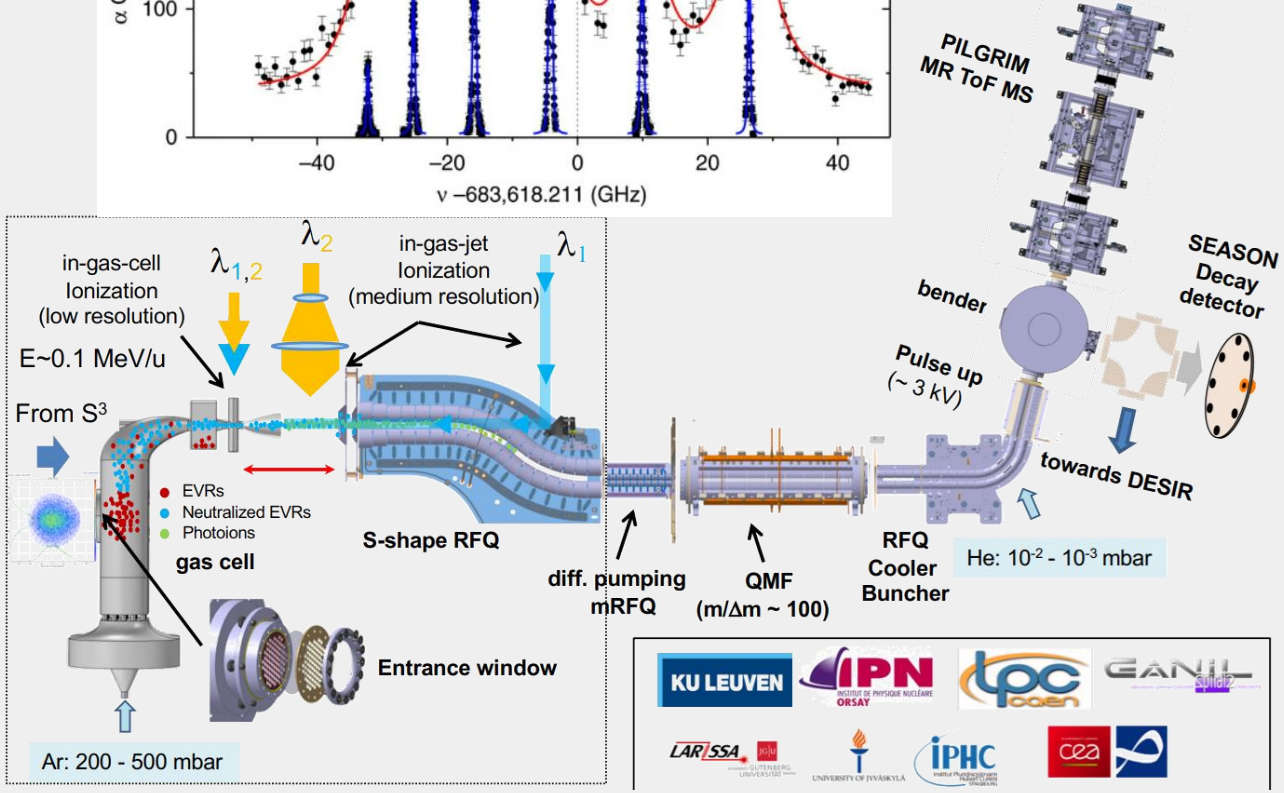
Courtesy of R. Heinke

In-jet RIS

High resolution with in-source efficiency

- 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





Ferrer, et al. Nat Commun **8**, 14520 (2017)

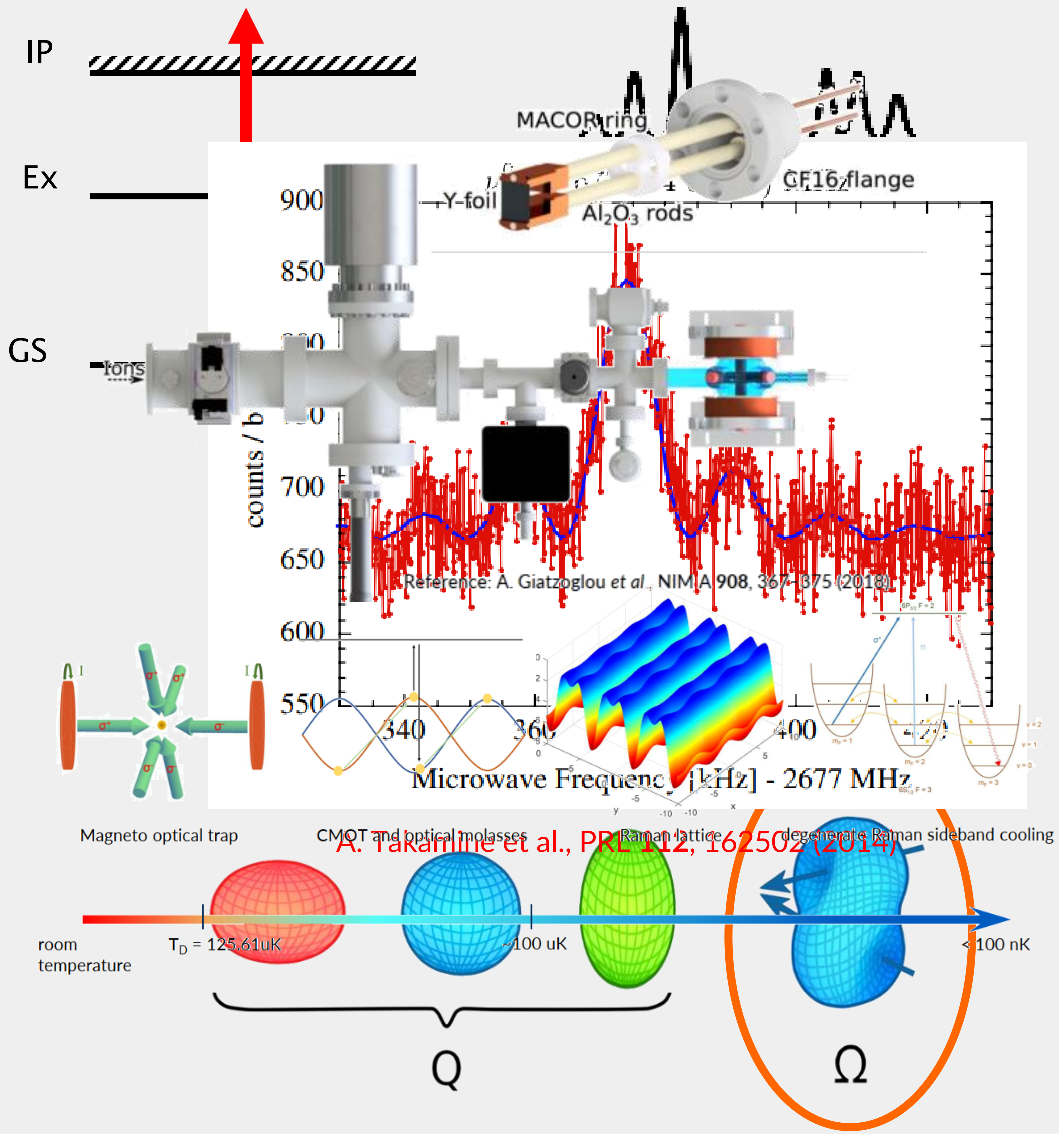
Courtesy of R. P. de Groote

Near future outlook:

Ultra-high precision frontier and radioactive molecules

Research cases

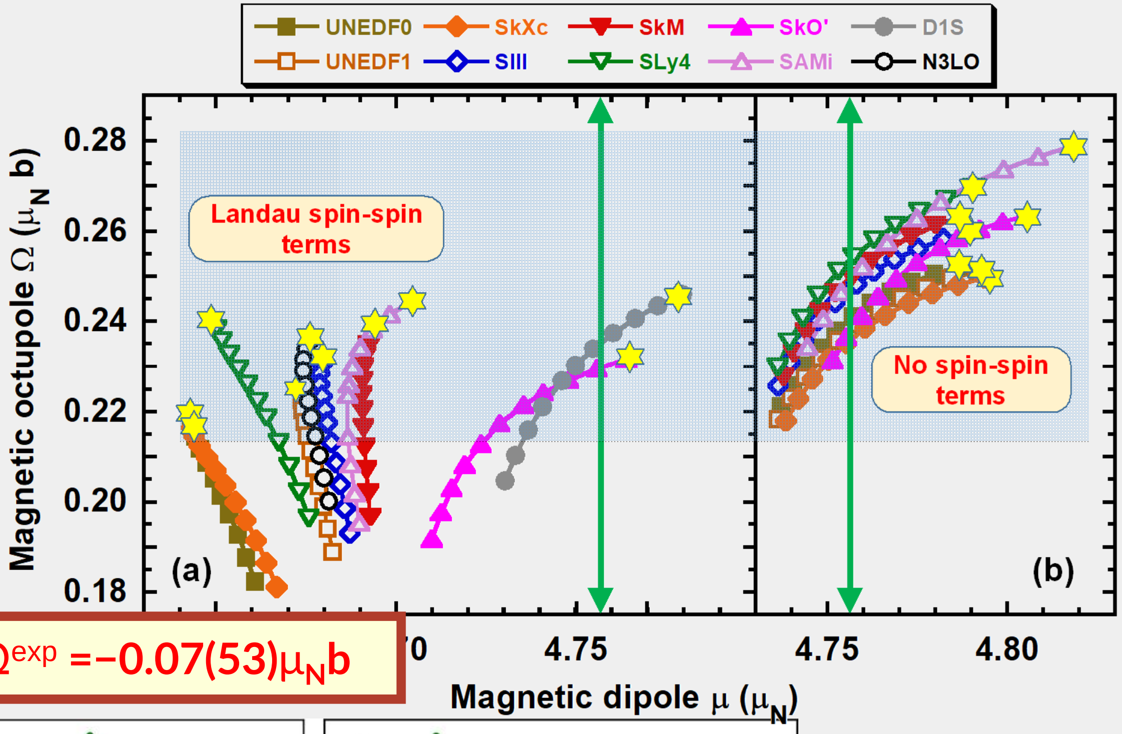
- Overview of hyperfine structure was a simplification.
  - Hyperfine anomaly: ratio of atomic *A* and the nuclear moment is ***not*** a
  - Higher-order nuclear moments: there is physics *beyond* the quadrupole
- Measuring these effects typically requires new precisions techniques
  - Laser-RF double resonance is a competitive option
    - Radiofrequency excitation within ground-state hyperfine manifold
  - Other is spectroscopy of ultra-cold atoms in a MOT



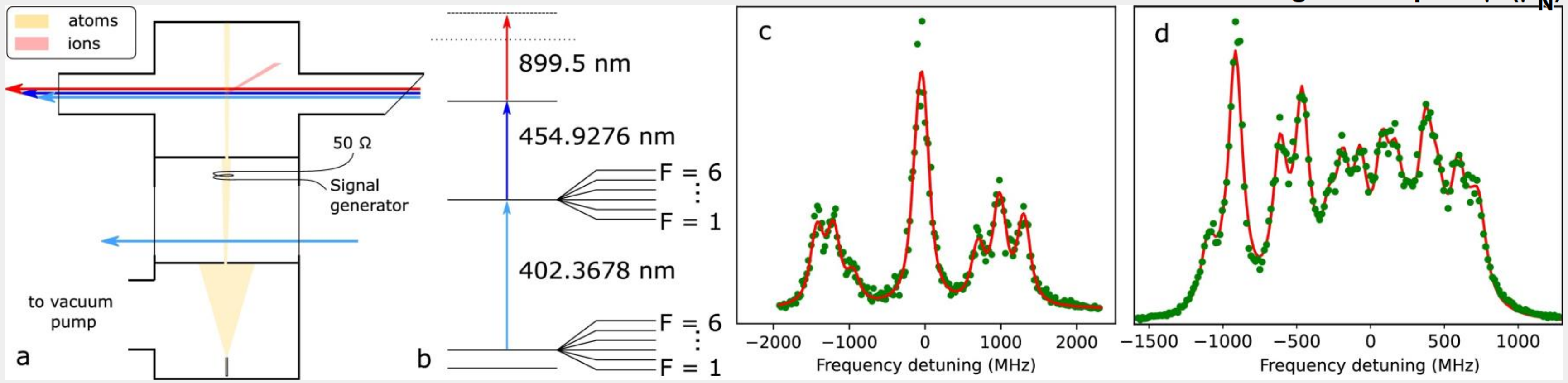
Courtesy of R. P. de Groote, W. Rattanasakuldilok

Technique
Laser-RF double resonance

- Optical resonance is not scanned
  - Radiofrequency excitation within ground-state hyperfine manifold
  - Optical 'amplifier' of RF resonance
- **Magnetic moments in ^{45}Sc**



$\Omega^{exp} = -0.07(53)\mu_N b$



R. P de Groote et al., Phys. Lett **B** 827, 10 2022, 136930

Courtesy of J. Dobaczewski

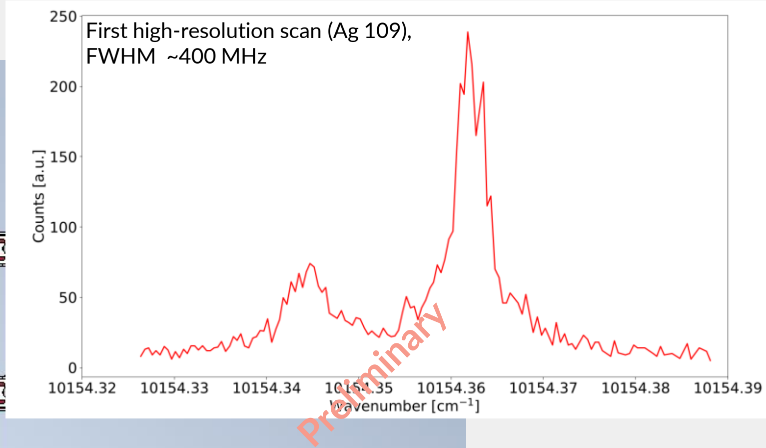
RAPTOR
“RIS And Purification Traps for Optimised spectroscopy”

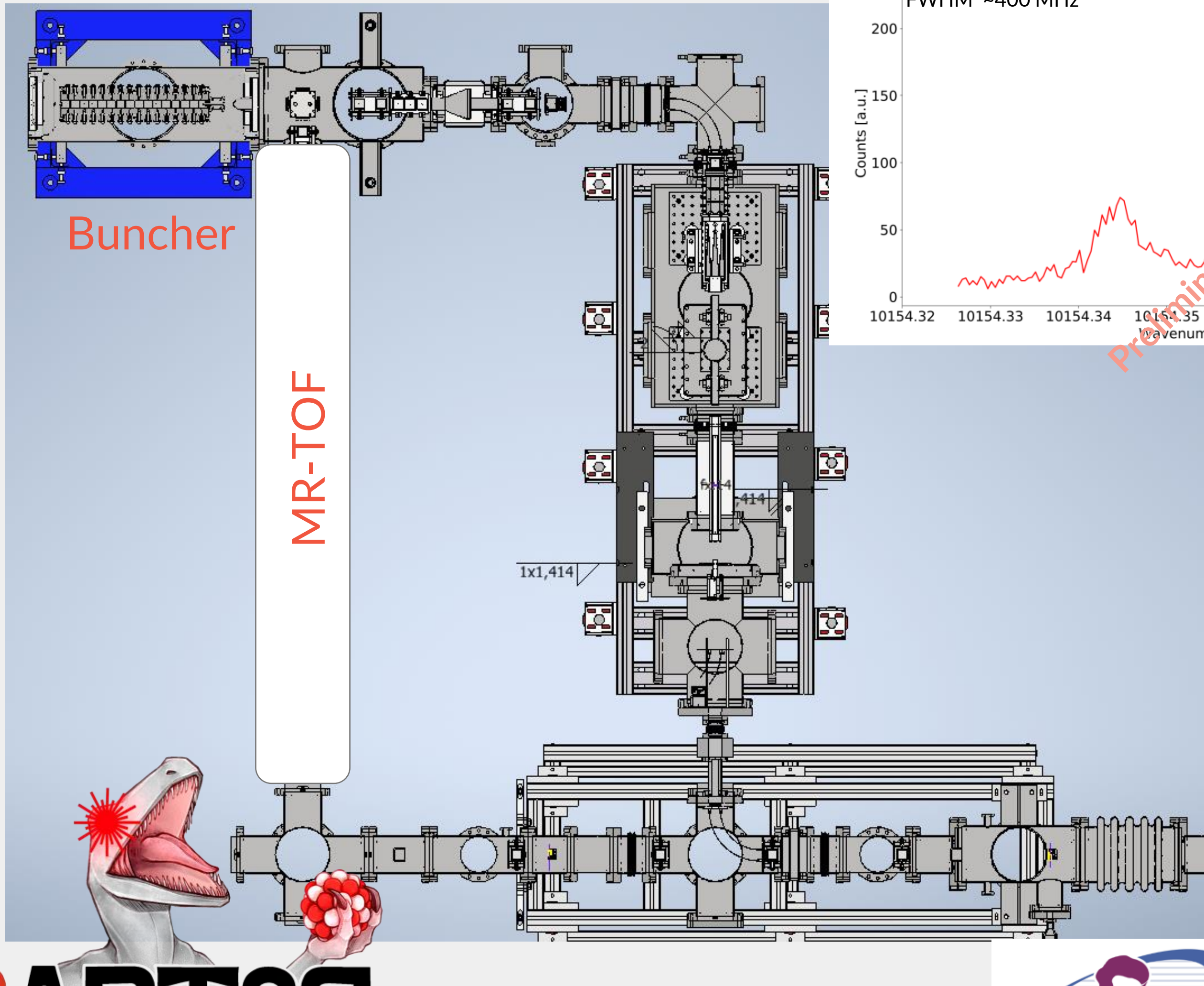
- “RIS And Purification Traps for Optimised spectroscopy”
- Collinear laser resonance ionization spectroscopy
  - Collinear geometry at a few kV beam energy yields ~300 MHz linewidth
  - Laser resonance ionization provides high efficiency
  - Using Penning trap as ultimate background-removal device

Laser-RF double resonance capability!

Bi or In possible cases.

First High-resolution resonances!!





RAPTOR

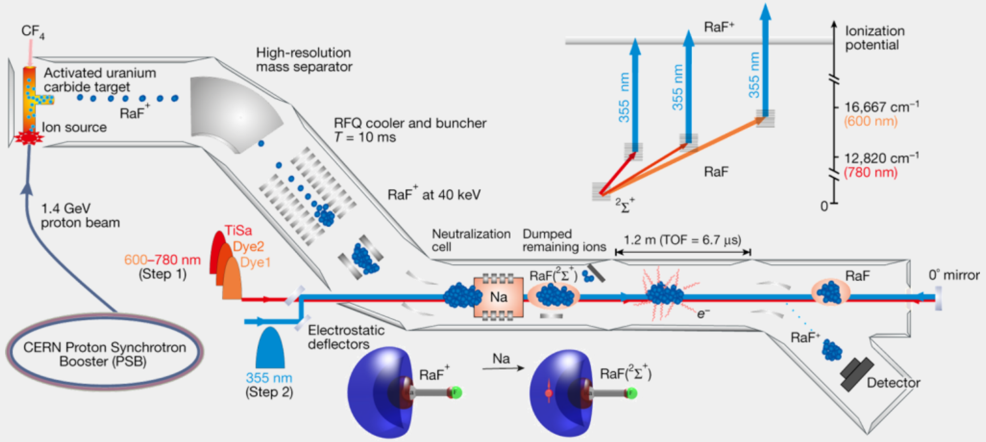
MARIE CURIE ACTIONS

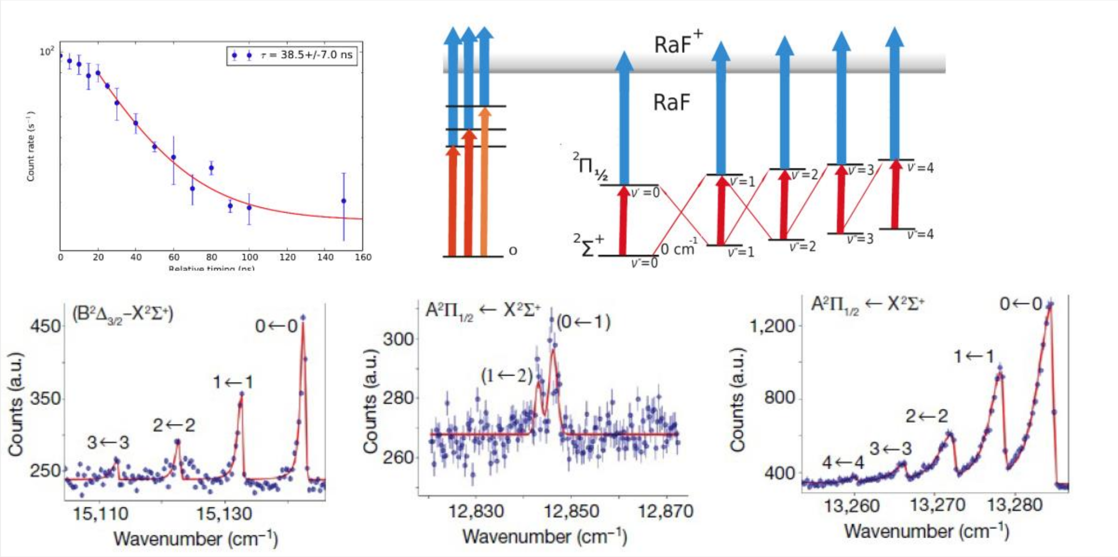
Courtesy of J. Dobaczewski, R. P de Groote

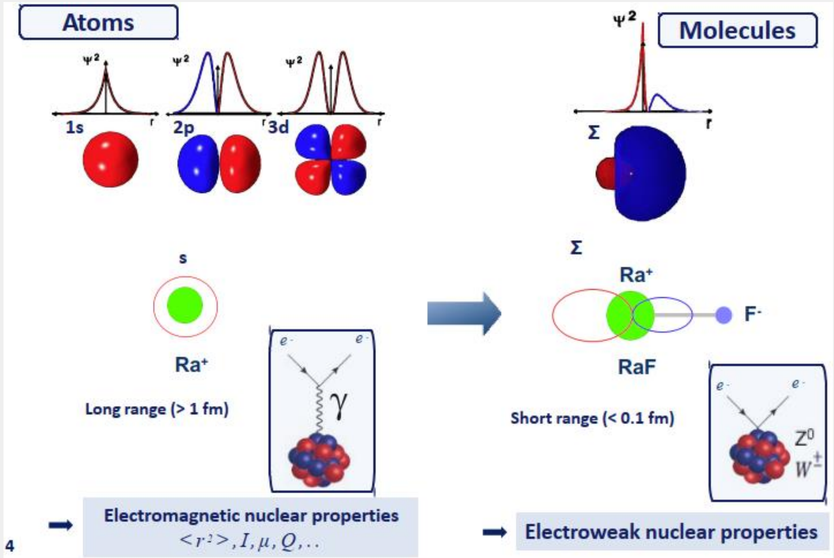
Radioactive Molecules

New Era of Precision (Atomic, Molecular, Nuclear) Physics

- A promise of sensitivity not only to nuclear structure, but also to fundamental symmetries
- 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







R. F. Garcia Ruiz Nature volume 581, 396–400 (2020)

Courtesy of R. P. de Groote, R F Garcia Ruiz

Final remarks

- Nuclear chart has been explored extensively with optical techniques, but **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**
- Collinear RIS has been established as vital tool for probing nuclear structure at the **limits of existence**
  - And it has fostered new avenues such as **radioactive molecules** and **precisions techniques**
- Collaboration between experimentalists and theorists has been extremely fruitful - It is *vital* to expand this collaboration
- Sharing technical expertise is very productive
  - Collinear-RIS, gas cell and gas jet techniques, Laser techniques such as injection-locking .
- Exploration of different facilities opens new frontiers
  - e.g. IGISOL@JYFL and KISS@RIKEN: access refractory isotopes, MNT reactions
  - e.g. NSCL/FRIB: fragmentation reactions
  - e.g. GANIL/ S3LEB: High yields near ^{100}Sn
- Embracing new frontiers, with precision comes additional information!
  - Measurement schemes and setups tailored to specific goals are very worthwhile
  - Information extracted via Radioactive molecules can perhaps compete with collider data?

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