



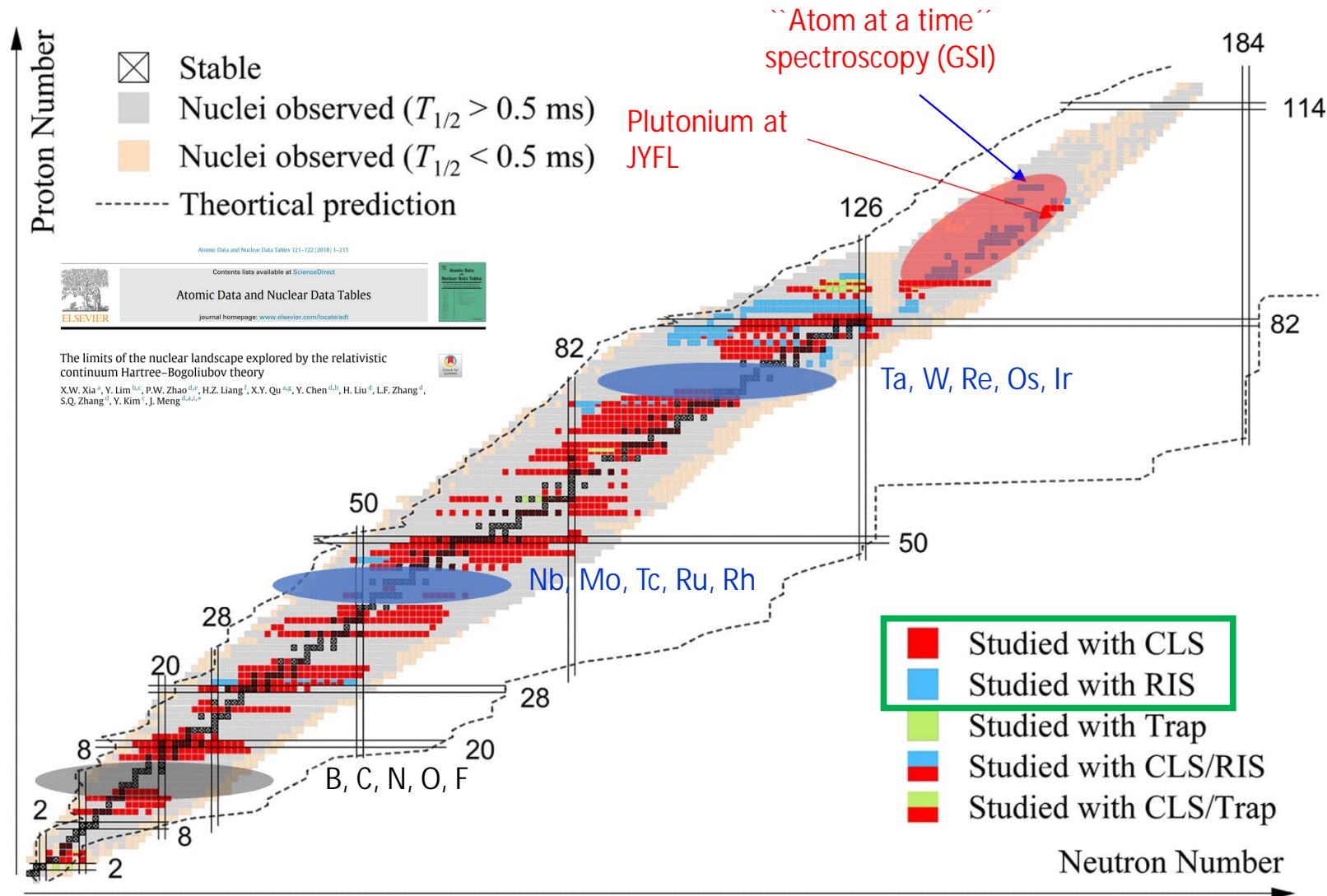
Ground-state properties and techniques

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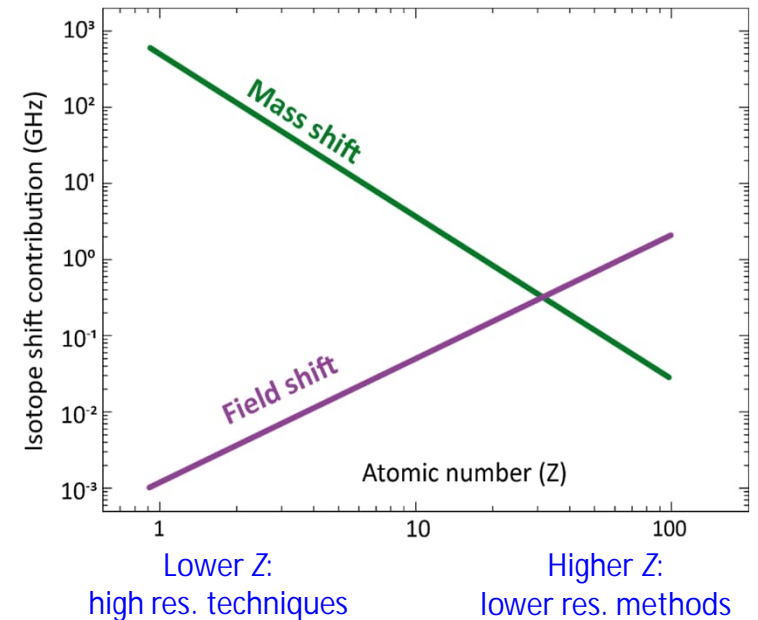


Chart of nuclides from optical spectroscopy



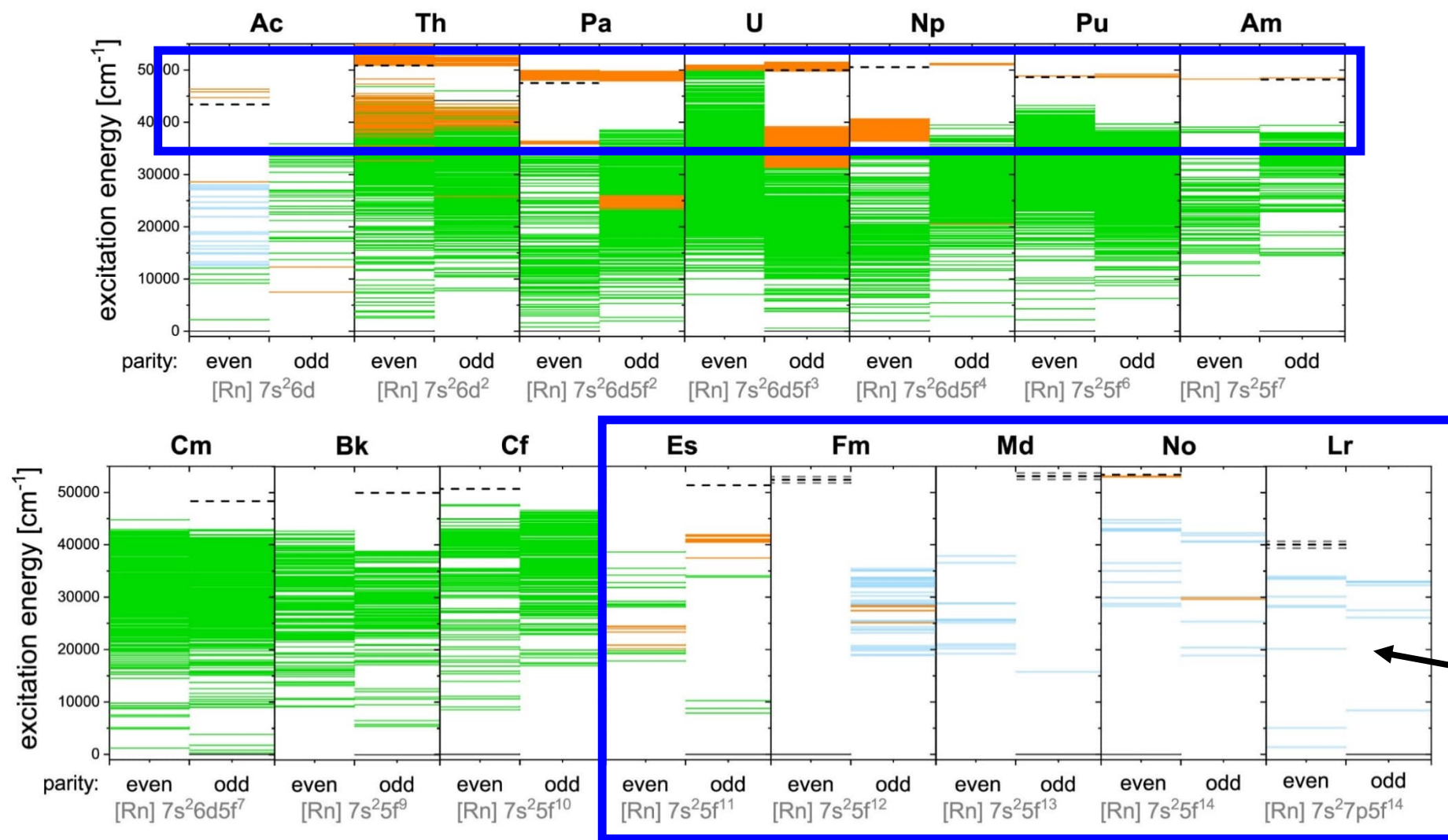
Limitations due to production and atomic properties

- Actinide region (low production)
- Refractory elements (target/ion source developments needed)
- Light elements are very reactive (molecular compounds). Also require laser developments



Latest review: X.F. Yang et al., PPNP 129 (2023) 104005

(Lack of) atomic levels: the heaviest elements....



- Blaise & Wyrat compilation
- Recent additions
- Theoretical predictions

The hunt for the first atomic level in Lr (Z=103) is (still) ongoing at GSI.

Only theoretical calculations!

Figure from: M. Block, M. Laatiaoui and S. Raeder, PNP 116 (2021) 103834

Laser spectroscopy setups worldwide



Operational

Commissioning

Planning

Nuclear properties

Laser ion source

Application

Outline of my two lectures

Lecture 1: Nuclear ground state properties

- The atomic mass, nuclear binding energy and nuclear structure
- Nuclear fingerprints on atomic lines – the isotope shift
- Hyperfine structure

Lecture 2: Techniques of optical spectroscopy and selected results

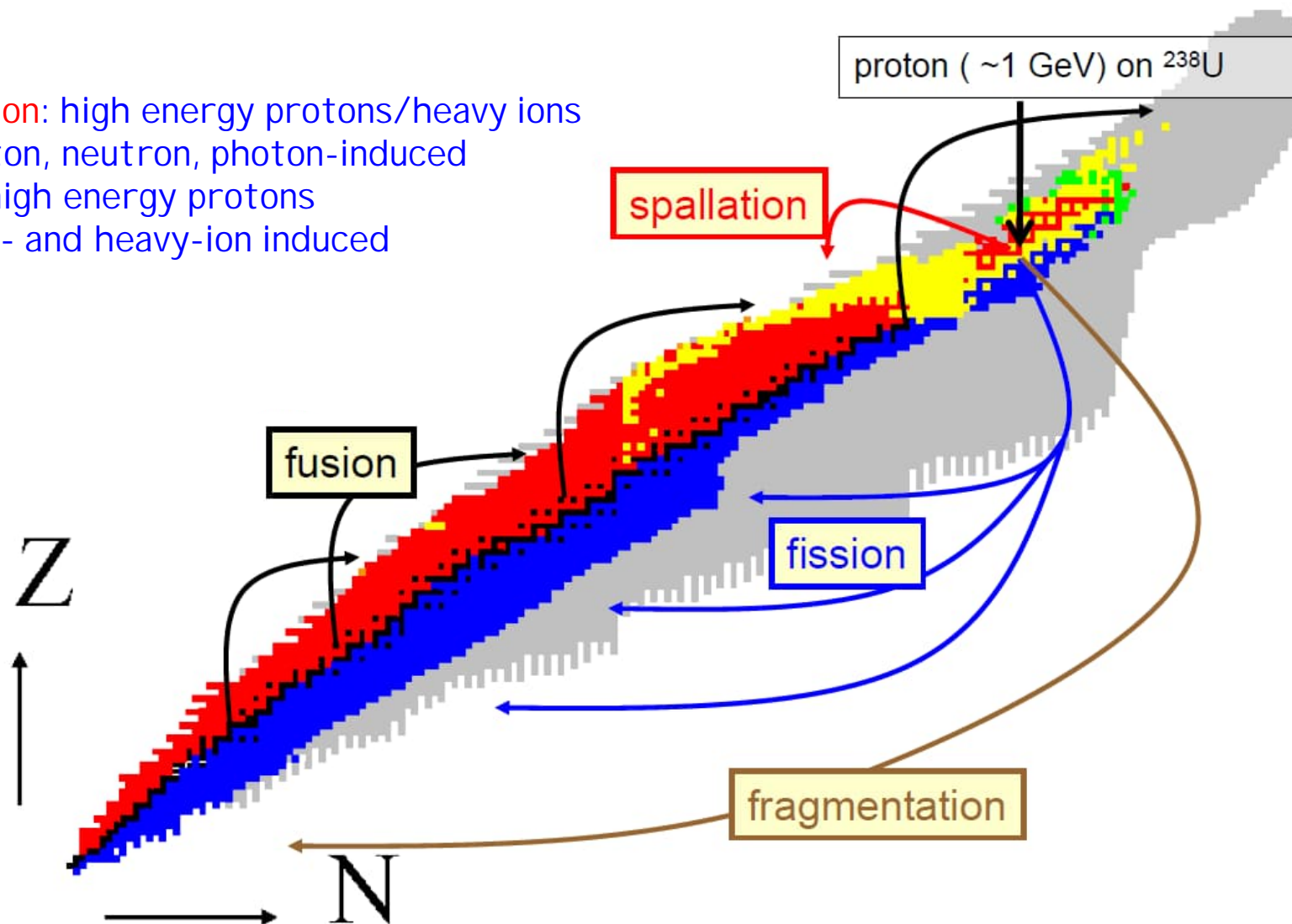
- A short introduction to radioactive ion beam production
- Laser resonance ionization
- Doppler-free laser spectroscopy
- What can we learn from charge radii, nuclear moments and spins?



A radioactive ion beam toolbox

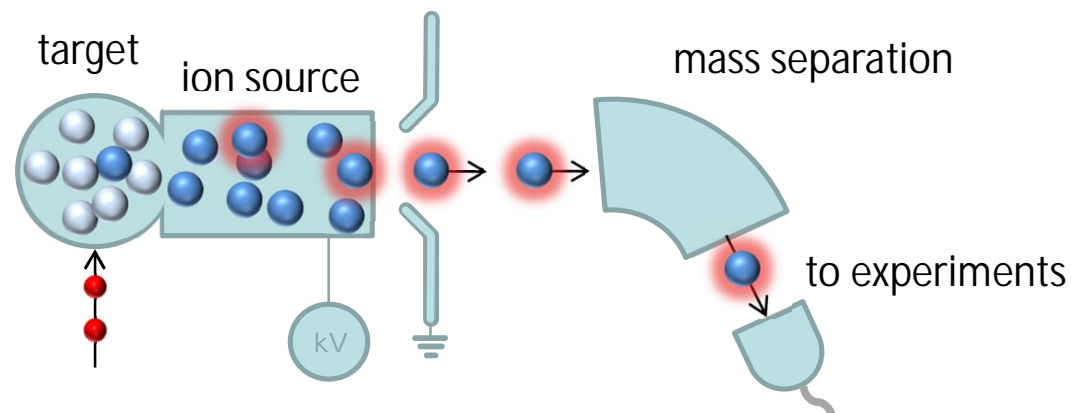
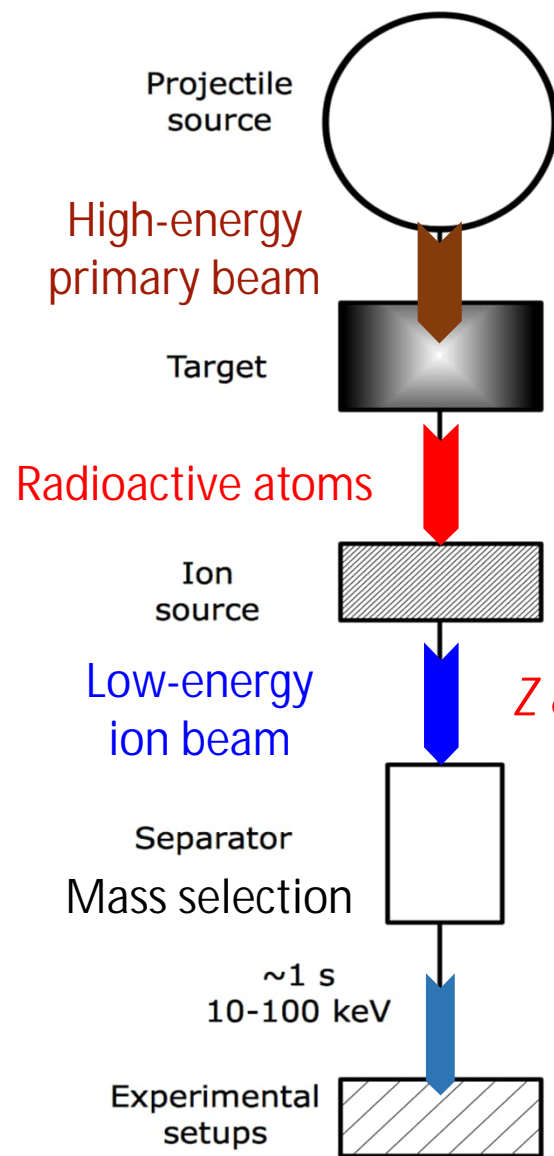


- Fragmentation: high energy protons/heavy ions
- Fission: proton, neutron, photon-induced
- Spallation: high energy protons
- Fusion: light- and heavy-ion induced



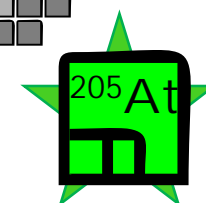
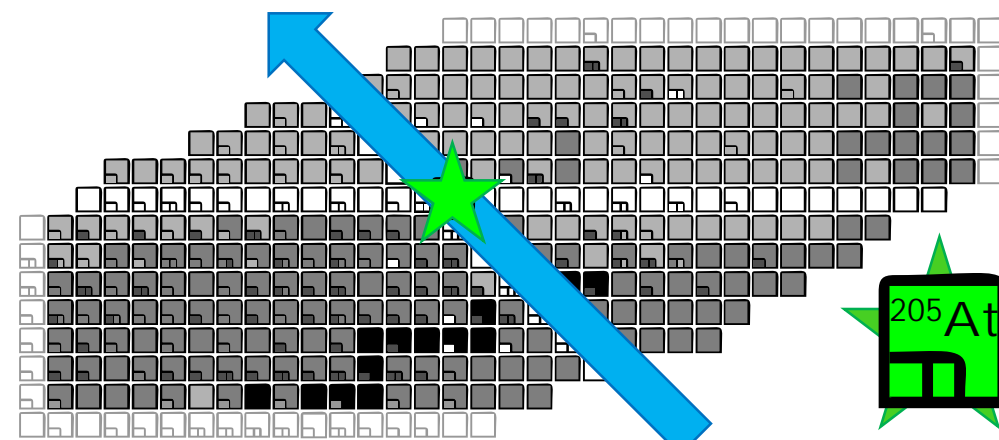
The Isotope Separation On-Line method

First developed in 1951, Niels Bohr Institute



● projectiles ● target material ● neutrals ● ions

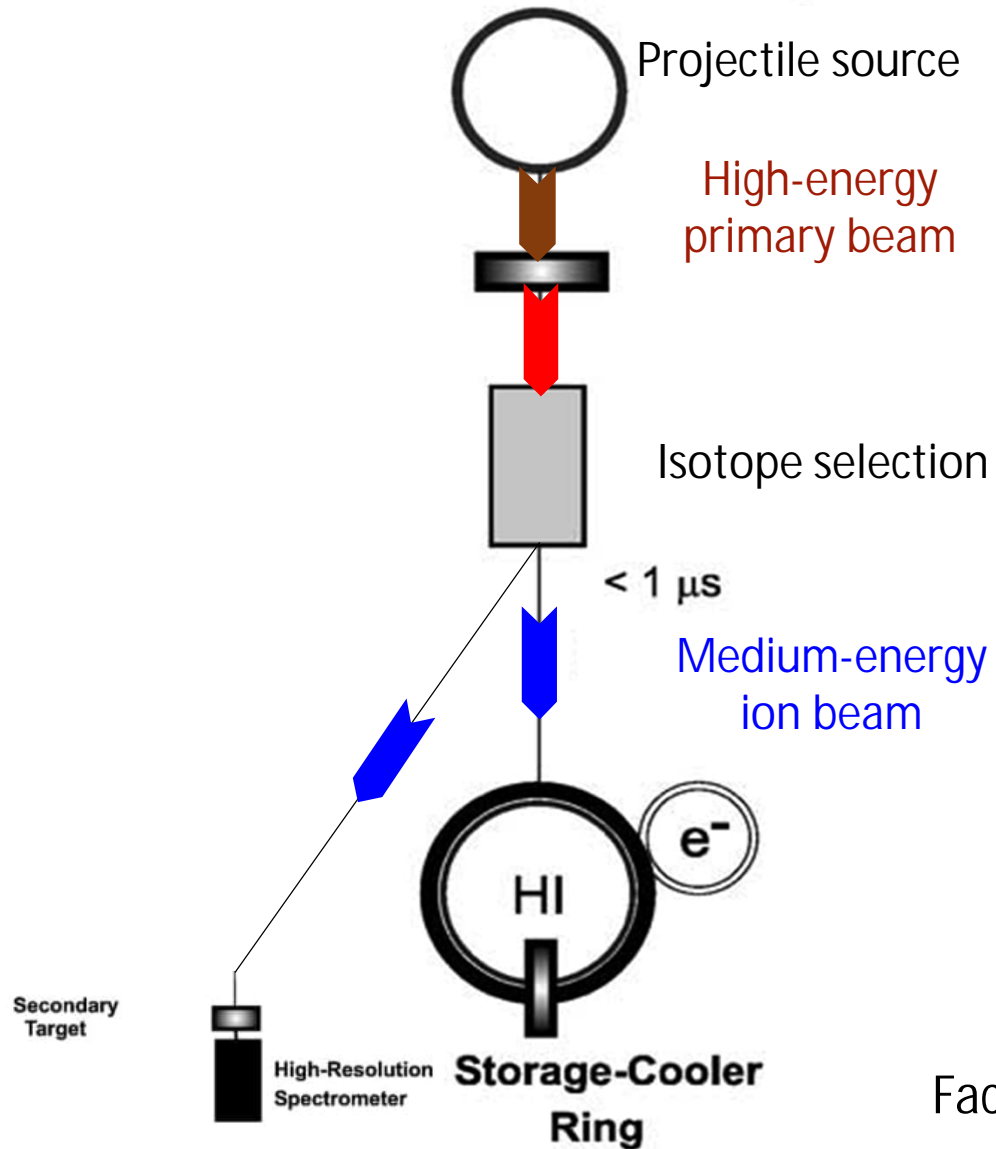
Z & $T_{1/2}$ dependence



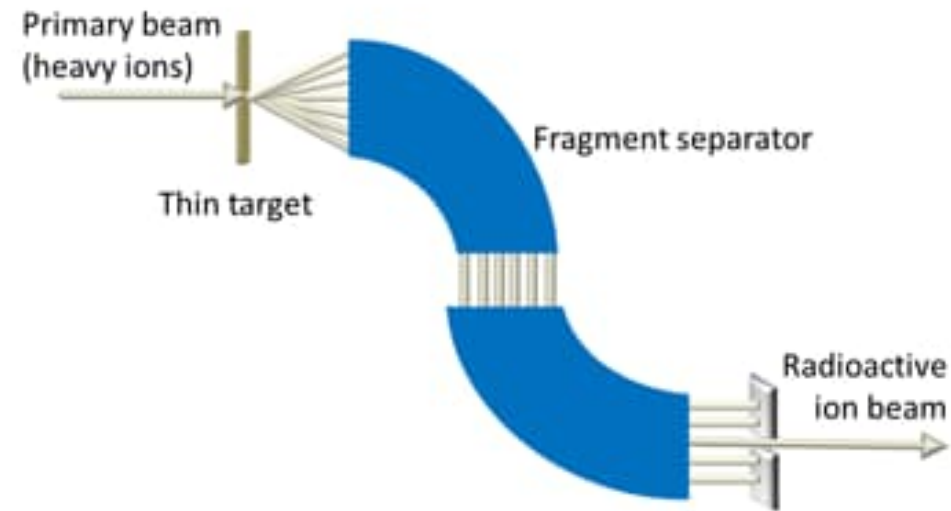
+ ^{205}Pb , Bi, Po, Rn, Fr...

e.g., ISOLDE, TRIUMF, GANIL...

The in-flight method



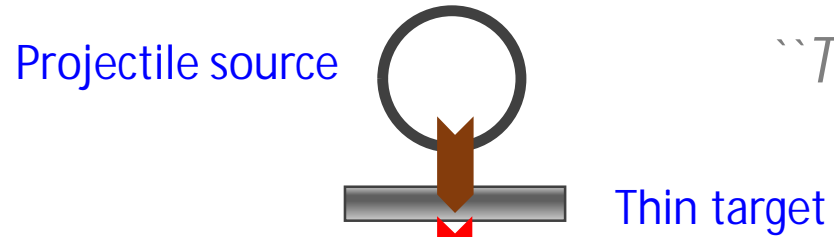
First in-flight separator, Oak Ridge (1958)



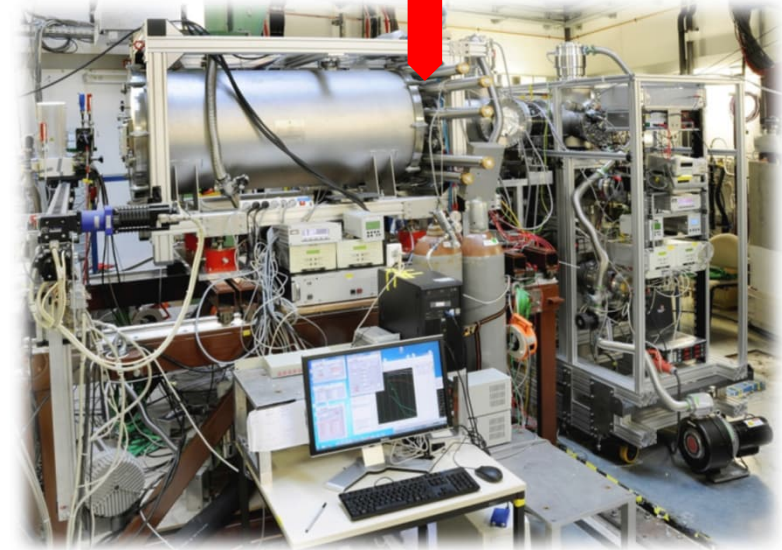
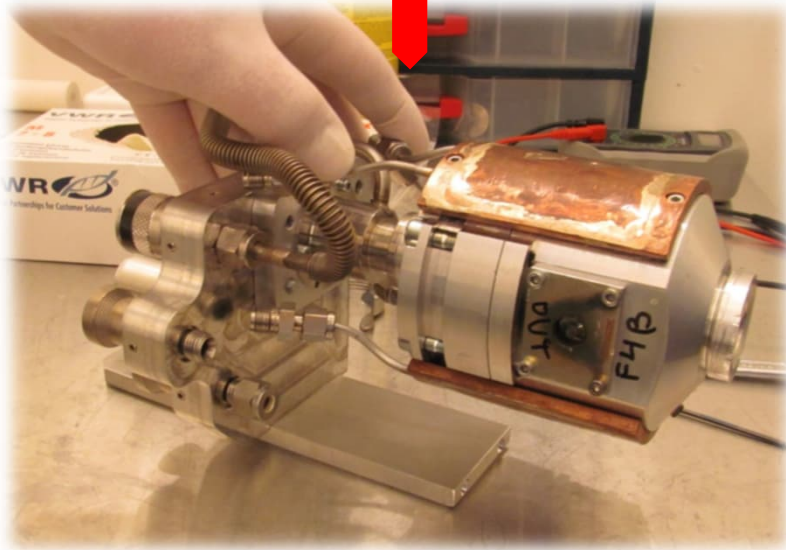
- Very fast separation, access to μs half-lives and beams of ALL elements
- Often poor beam quality (due to large phase space)
- Precision experiments at low-energy not directly accessible

Facilities include e.g., GANIL, GSI, NSCL/MSU, RIBF @RIKEN...

The IG(ISOL) / gas catcher (hybrid) method



“The best of both worlds”



- Extraction of ions in gas flow (ion guide), or electrical fields (gas catcher)
- For the IGISOL method the (stopping) efficiency is relatively low, poor selectivity
- Universal method of radioactive ion beam production

Universality – advantages and drawbacks

Your favorite exotic nucleus



Gas catcher, Argonne National Lab

=

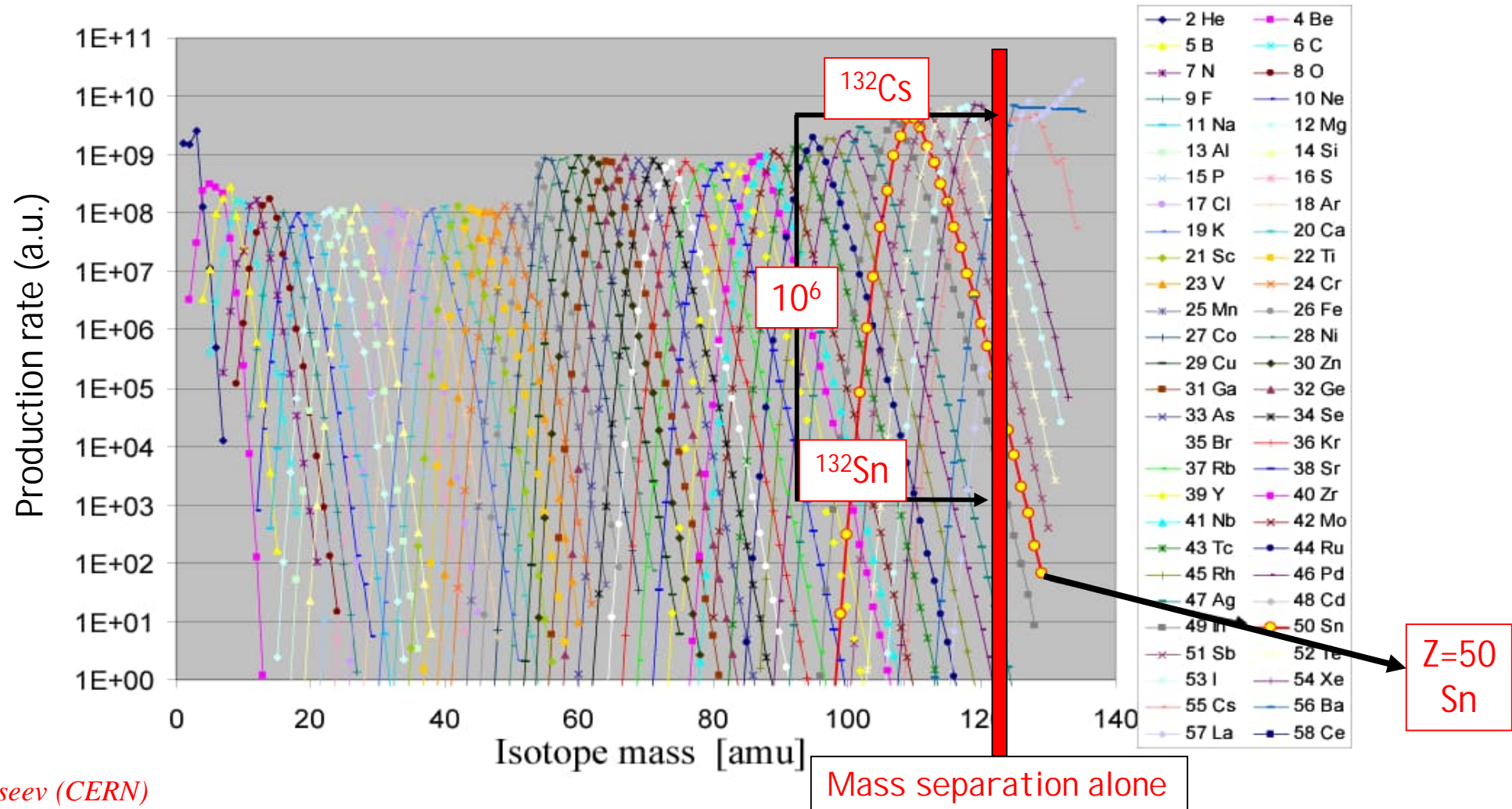


+



Why do we care about selectivity?

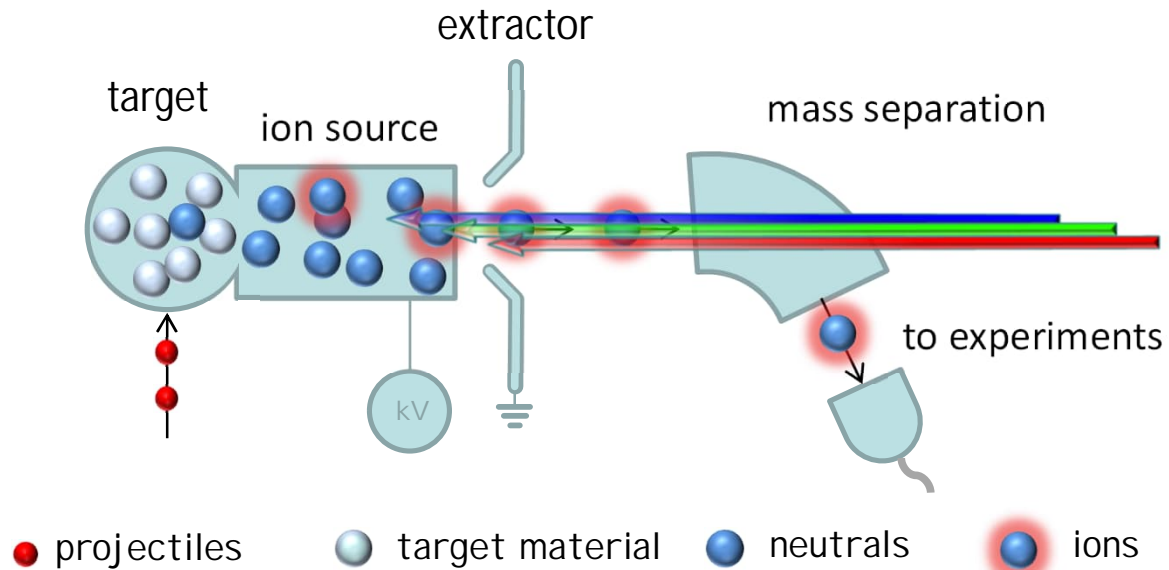
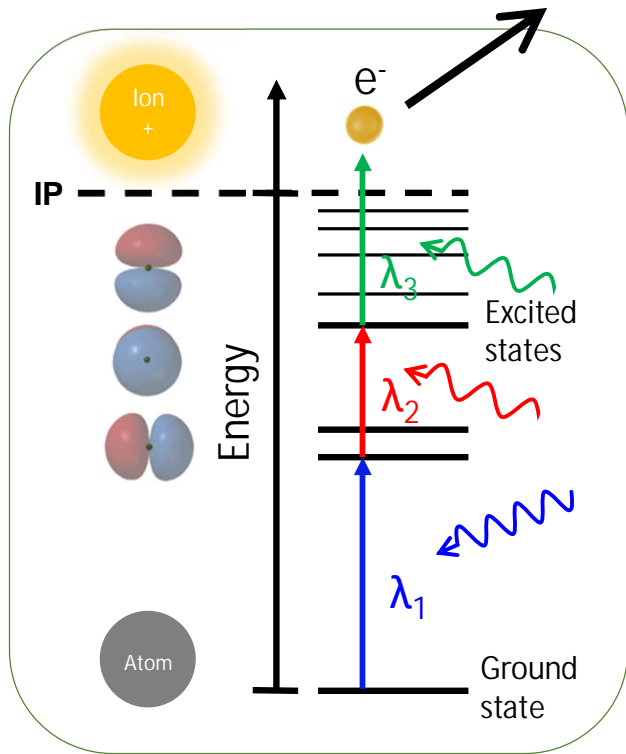
Isotope production for a 1 GeV p beam on a La target



J. Lettry, V. Fedoseev (CERN)

Using the atomic fingerprint – a resonant process

We transport the laser light directly into the ion source.



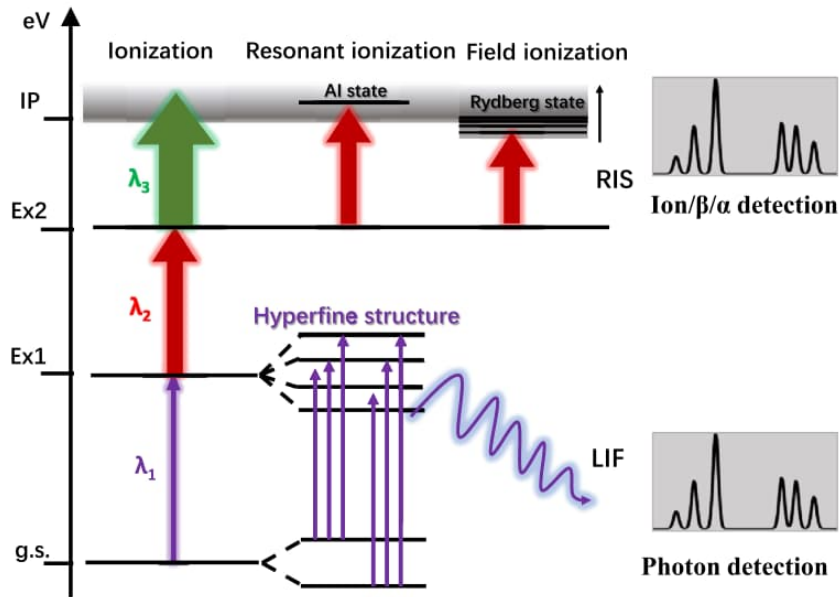
SELECTIVITY & EFFICIENCY

- Laser ionization and mass separation is used to only select the isotope of interest, and reduce the number of unwanted isotopes, molecules,... in the beam
- Laser ionization is a useful tool in the RIB toolbox
- The ion source of choice for several RIB facilities!

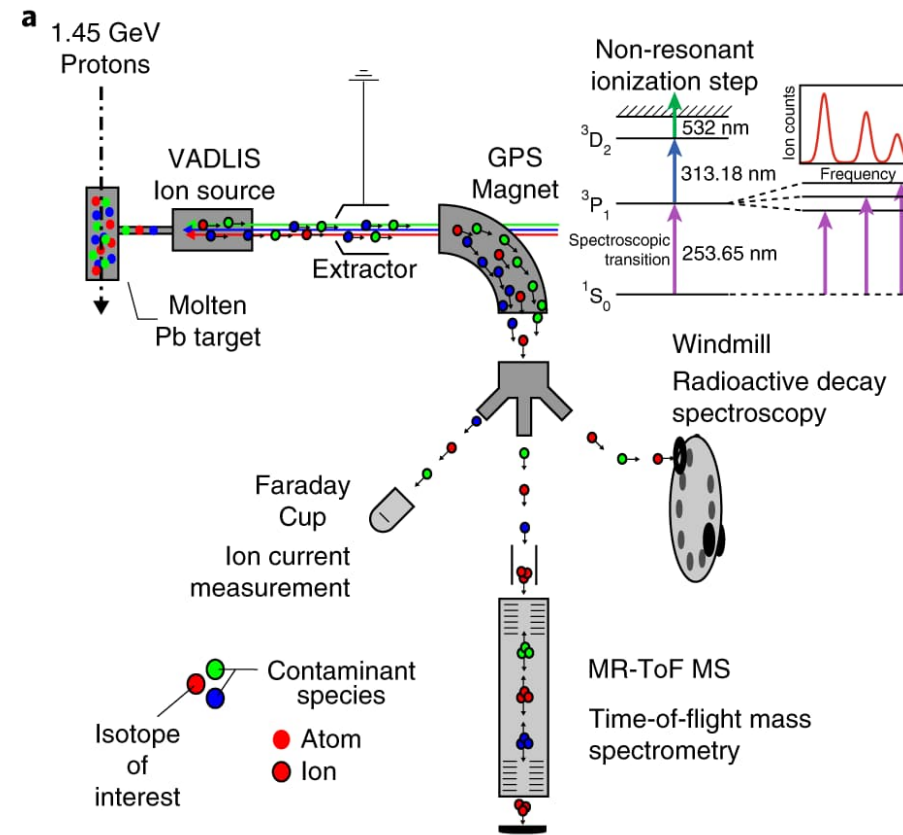


Resonance ionization spectroscopy (RIS)

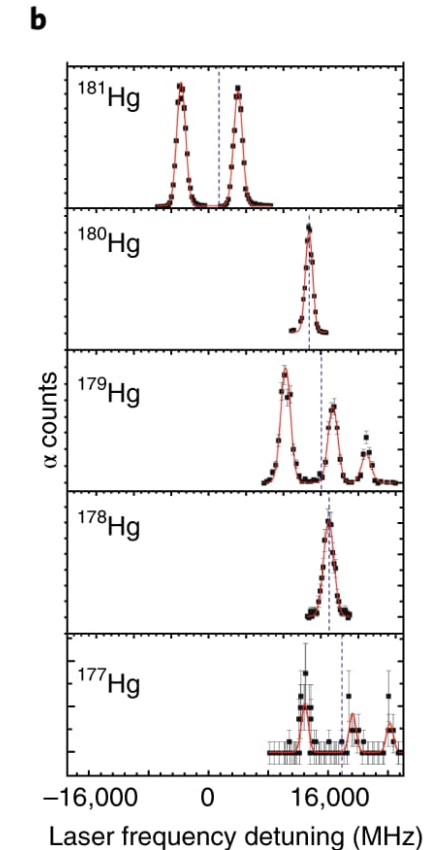
"In-source" RIS is a close variant to laser ionization for radioactive ion beam production.



- Selective process (laser "on" vs "off")
- Efficient process ($\gg 1\%$ is routine)
- "Short" lifetimes
- Low yields (< 1 ion/s)
- High detection efficiency (ions/ α decays...)



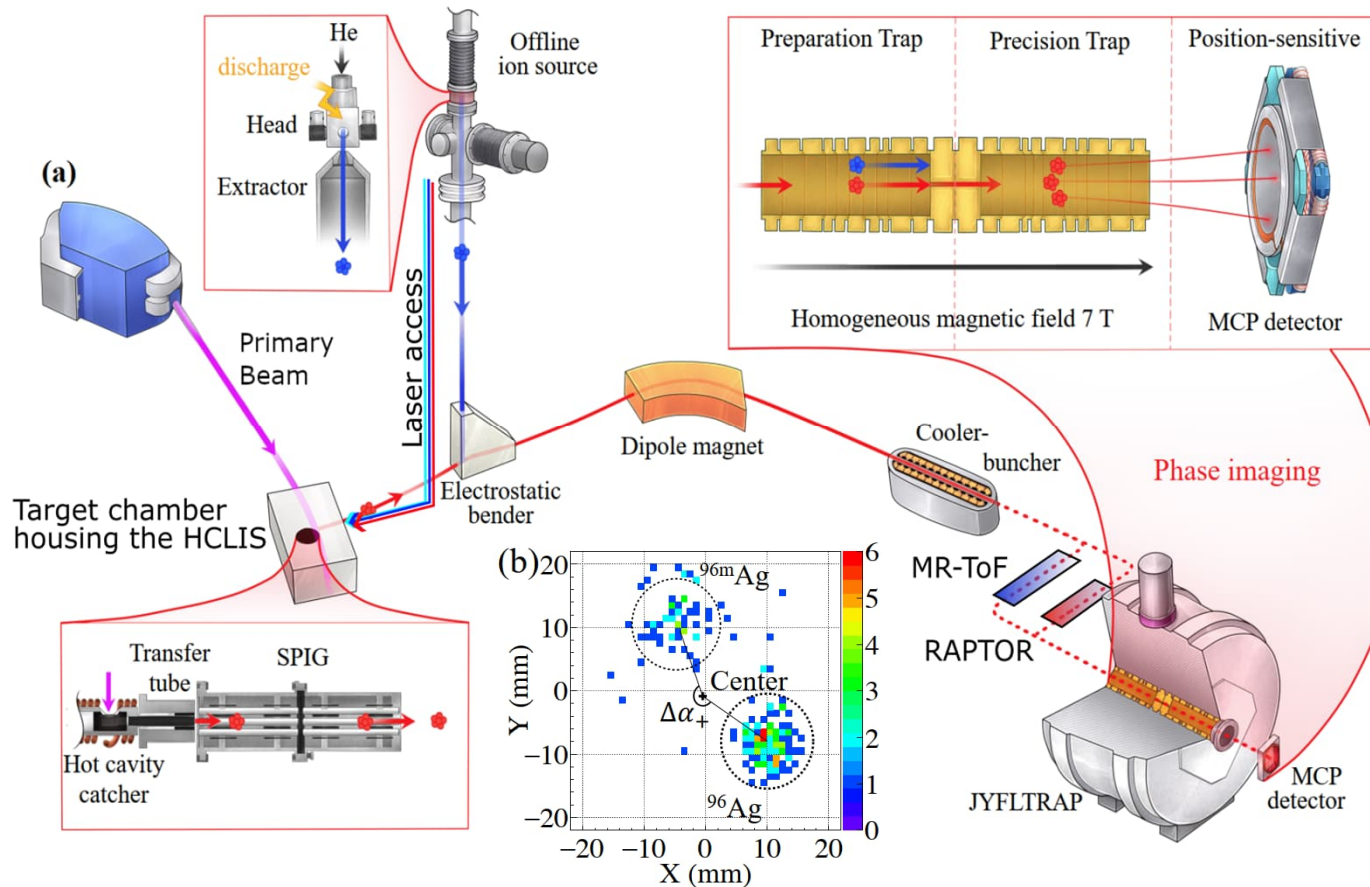
ISOLDE facility, CERN



B. Marsh et al., Nature Phys. 14 (2018) 1163

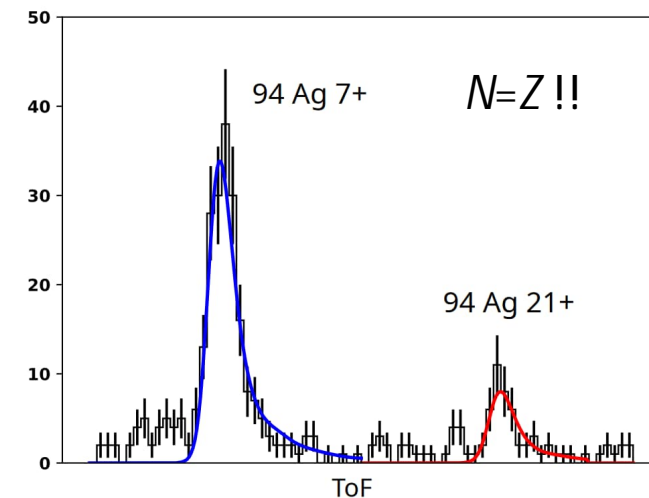
Pushing the sensitivity of in-source RIS

Recent efforts to combining methods from ion manipulation and trapping techniques with laser resonance ionization have enabled almost a complete suppression of background, opening a window to the most exotic nuclei.



- Three-step RIS of silver in a hot cavity ion source
- Heavy-ion fusion-evaporation reactions produce neutron-deficient Ag isotopes ($^{96-104}\text{Ag}$)
- Few % total efficiency
- Penning trap used for detection of ions
- On resonance rate of $^{96}\text{Ag} \sim 0.005/\text{s}$
- Excitation energy of ^{96m}Ag determined to be 115.0(17) keV*

* PRL accepted 2024



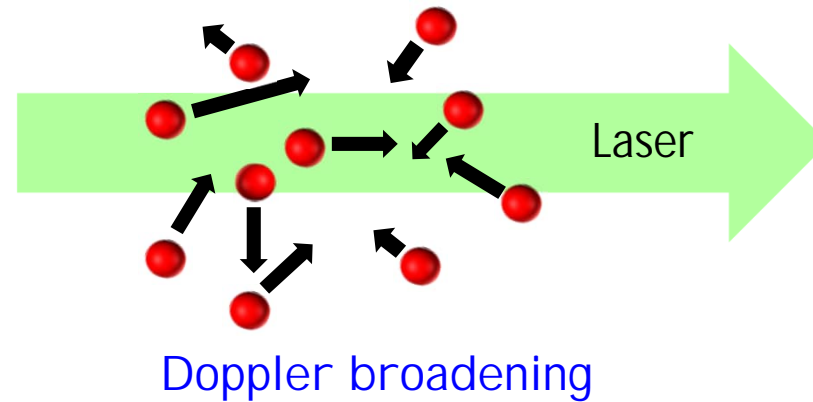
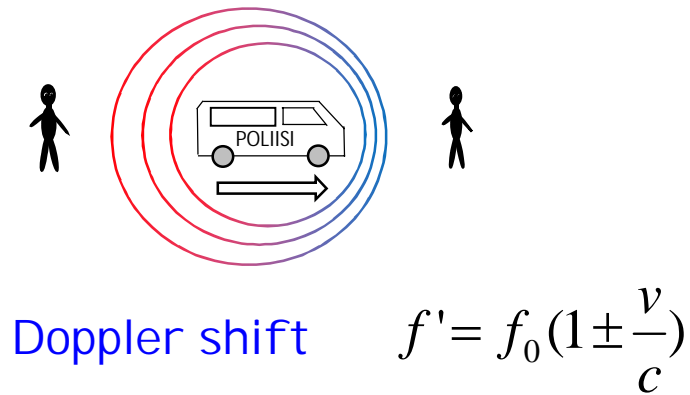
(2023) MR-TOF mass spectrometer for ^{94}Ag (0.15/min)

Technique described in: M. Reponen et al., Nature Comm. 12 (2021) 4596

The drawback of the in-source RIS approach



The observed transition linewidth can be broadened by Doppler effects due to the high temperatures involved (hot cavity), or collisions (if in gas):

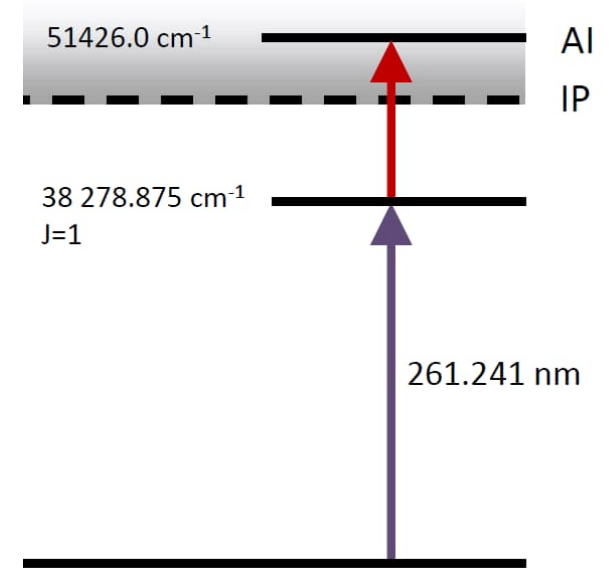
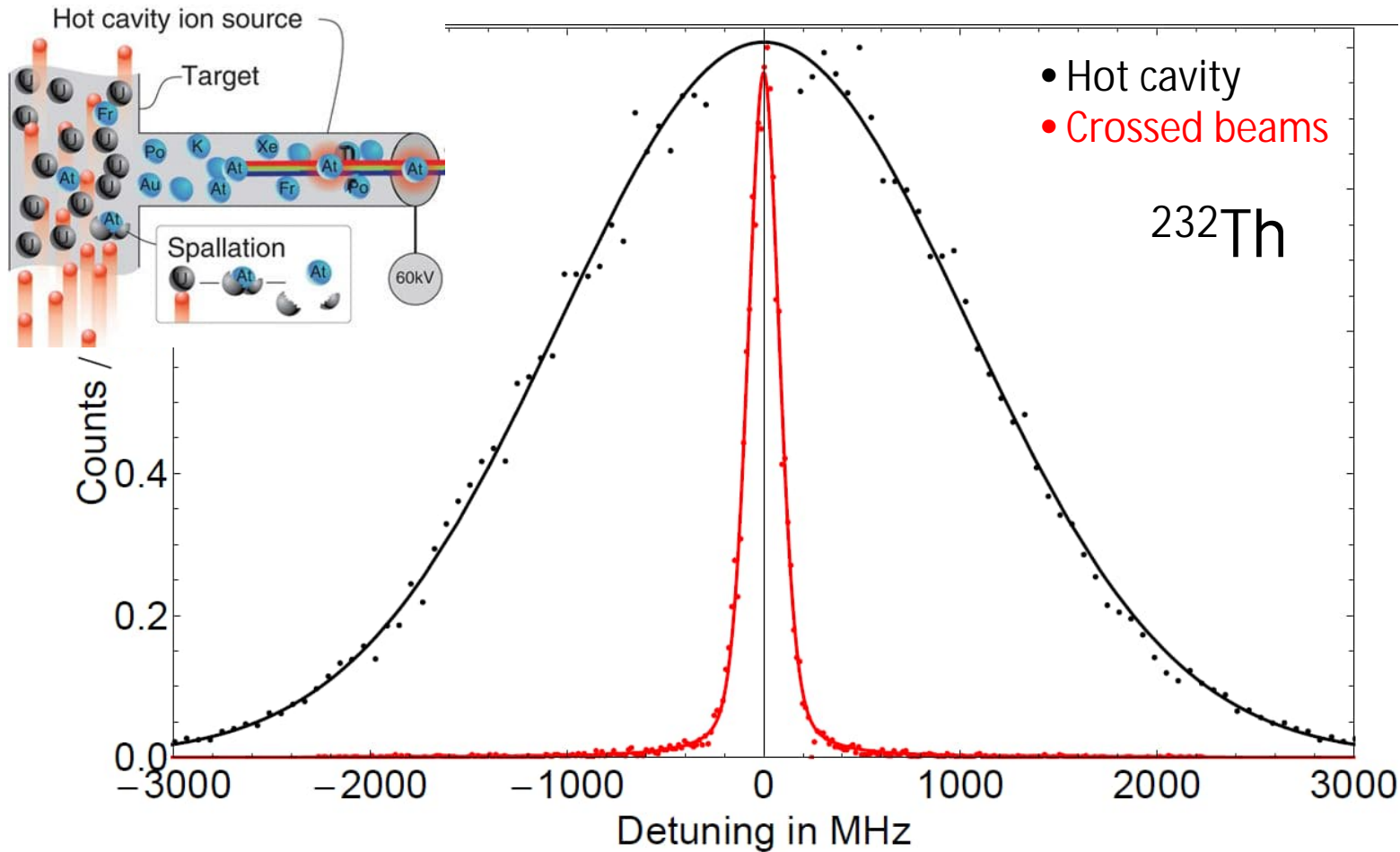


Thermal motion is a Maxwell-Boltzmann probability distribution. Causes a spread of frequencies observed by atoms:

$$P(f)df \propto \exp\left(-\frac{mc^2(f - f_0)^2}{2k_b T f_0^2}\right)df$$

$$\Delta_{FWHM} = f_0 \sqrt{\frac{8k_b T \ln 2}{mc^2}}$$

Doppler broadening from a hot oven



“Low” resolution allows us to measure:

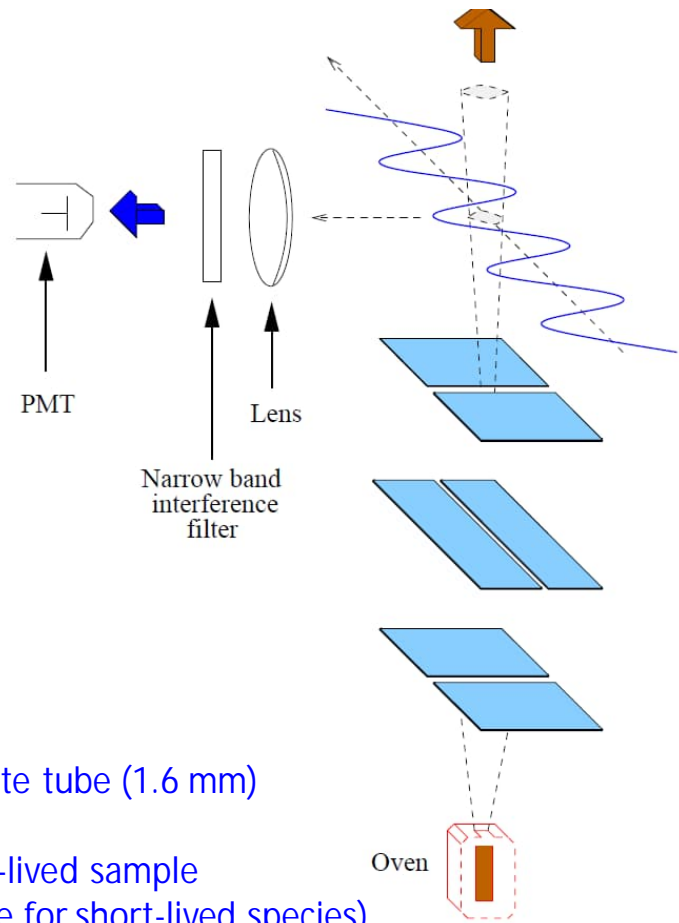
- Hyperfine A factor → magnetic moments
- Isotope shift → charge radii

Natural linewidth 35 MHz; spectral linewidth 2.4 GHz (in oven), 170 MHz (crossed-beams configuration).
 The Doppler broadening is often comparable or greater than HFS or IS!

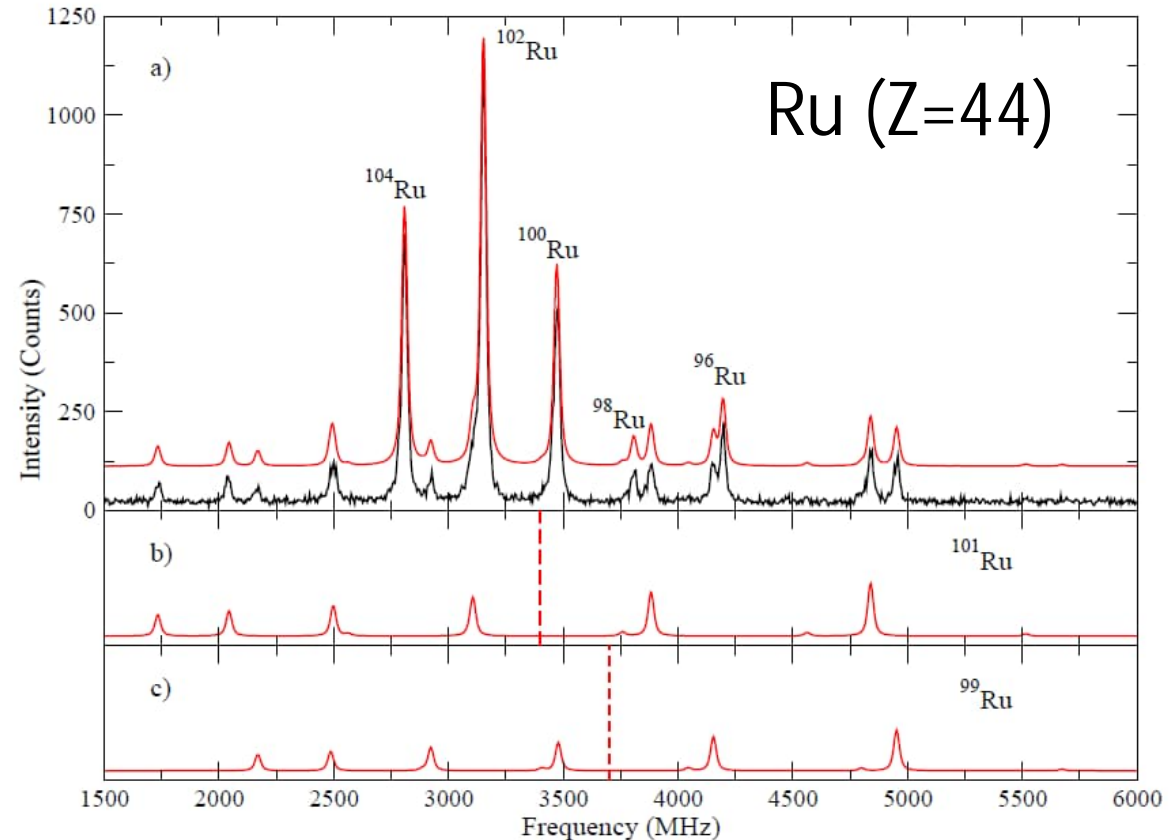
Methods used to mitigate the broadening



“Crossed beams” atomic beam spectroscopy. Incident laser beam(s) interact perpendicularly with a collimated beam of atoms. Photons or ions are detected orthogonally.



Ta or graphite tube (1.6 mm)
T ~ 2500K
Stable/long-lived sample
(not suitable for short-lived species)

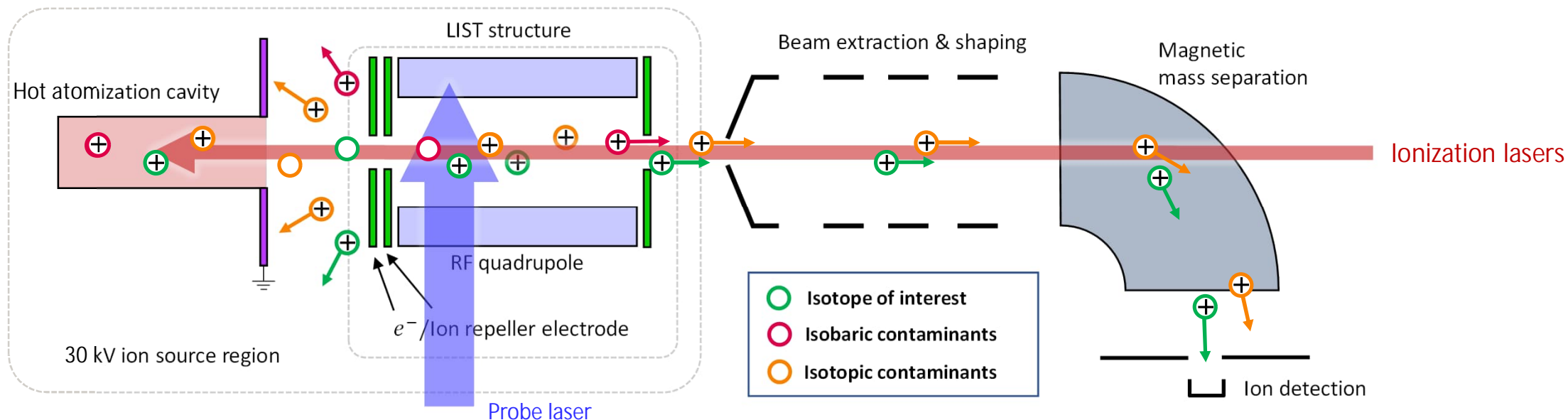


- 5 even-even isotopes
- 2 odd-A isotopes (15 HF components each)

D.H. Forest et al., J. Phys. G 41 (2014) 025106

Perpendicularly-Illuminated Laser Ion Source (& Trap)

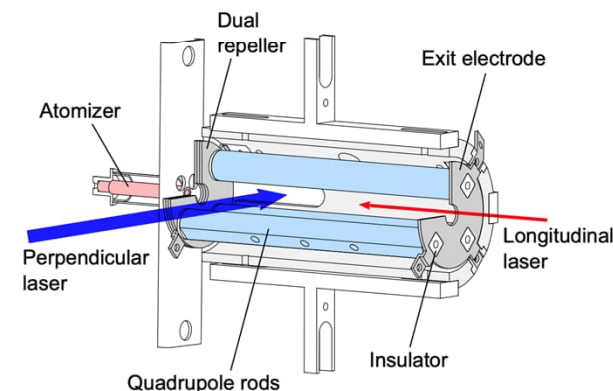
A variation to in-source RIS recently applied online at ISOLDE, CERN. Based on developments in U-Mainz.



Crossed atom beam / laser geometry in LIST structure:

- Electrodes provide a full suppression of surface ionized isobars
- Probe a reduced Doppler ensemble of atoms
- Suitable narrow-band laser must be used

T. Kron et al., PRC 102 (2020) 034307
R. Heinke et al., Hyp. Int. 238 (2017) 6

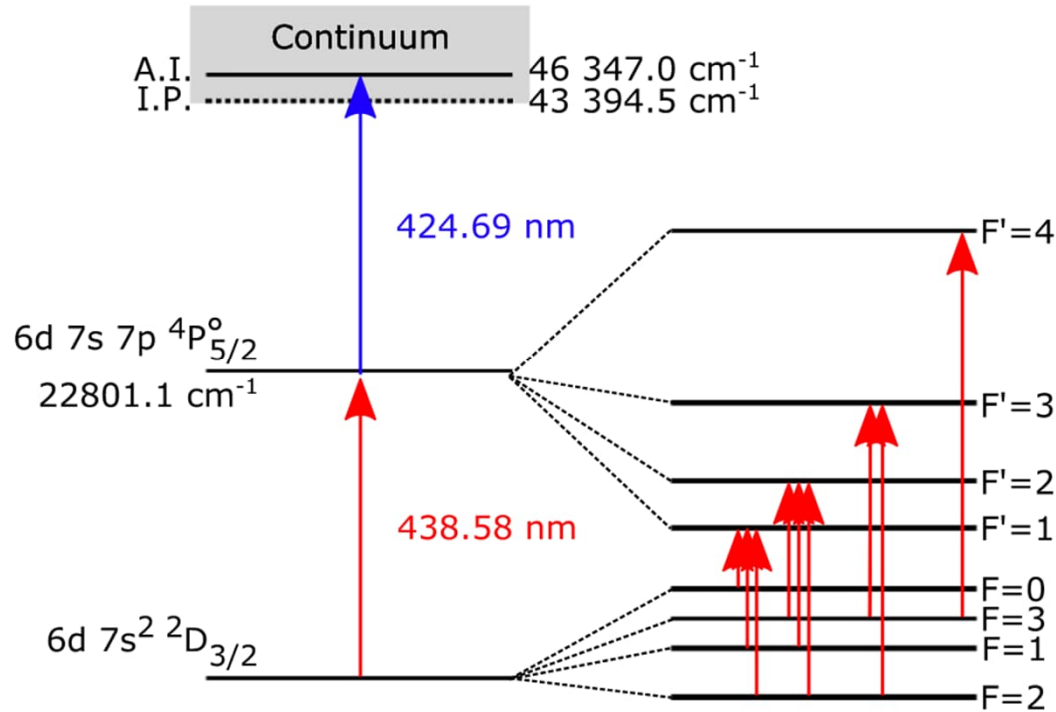


Perpendicularly-Illuminated Laser Ion Source

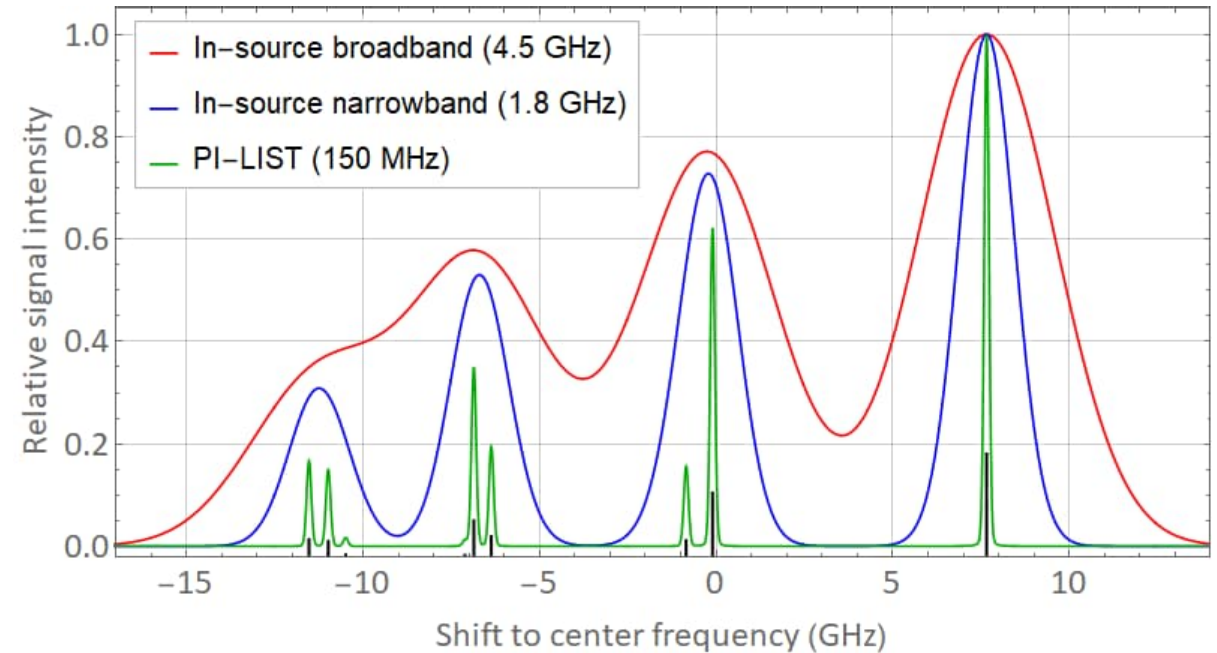


Ac laser ionization scheme

(used at several online facilities – ISOL & gas cell)



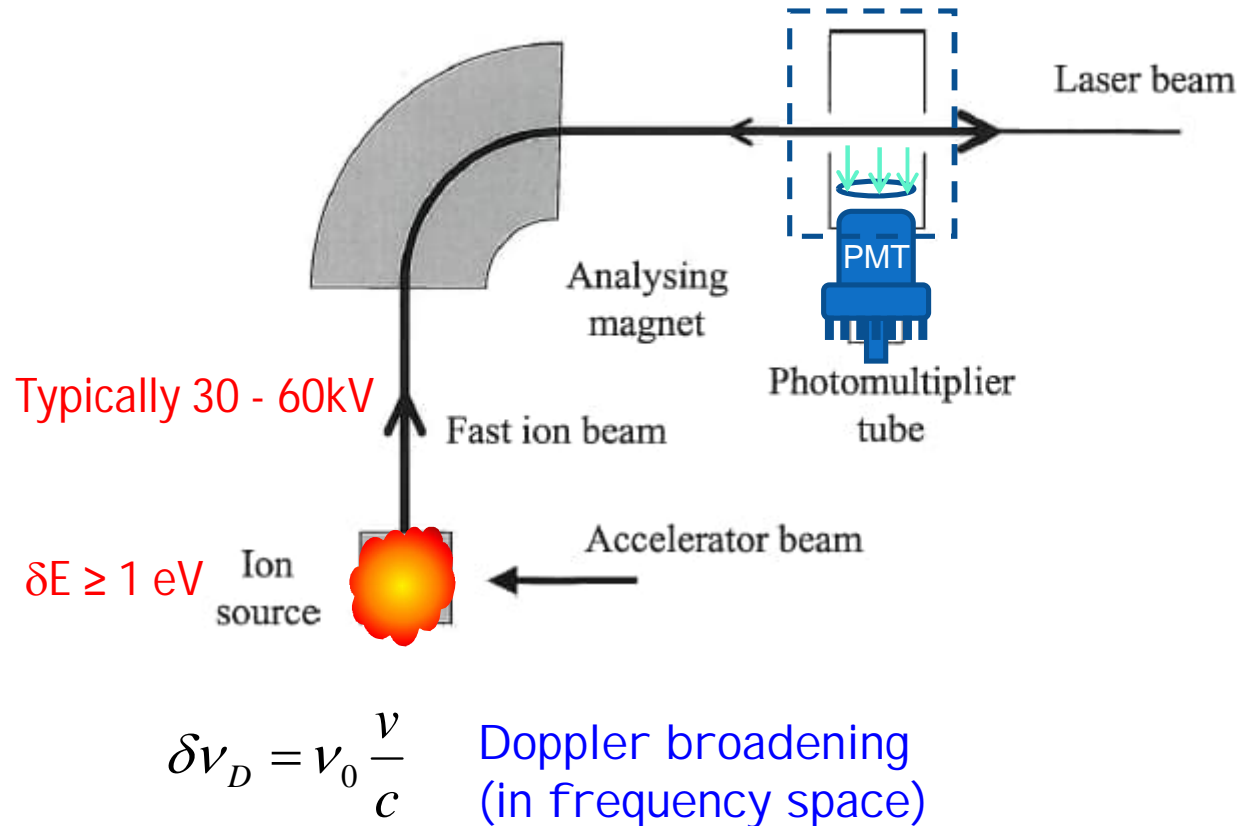
“High” resolution allows us to measure hyperfine B factor → Q moment



- Strong reduction in Doppler broadening seen
- Sensitivity <1 ion/s; resolution ~200 MHz
- Sacrifice in efficiency (10^2 to 10^4 compared to standard in-source resonance laser ionization, ~100 to “LIST”)

(Fast beams) collinear laser spectroscopy

In a collinear geometry, light, whether co- or counter propagating with the ion beam, interacts with accelerated ionic ensembles.



1. Accelerate all ions to energy E

$$E = eV = \frac{1}{2}mv^2$$

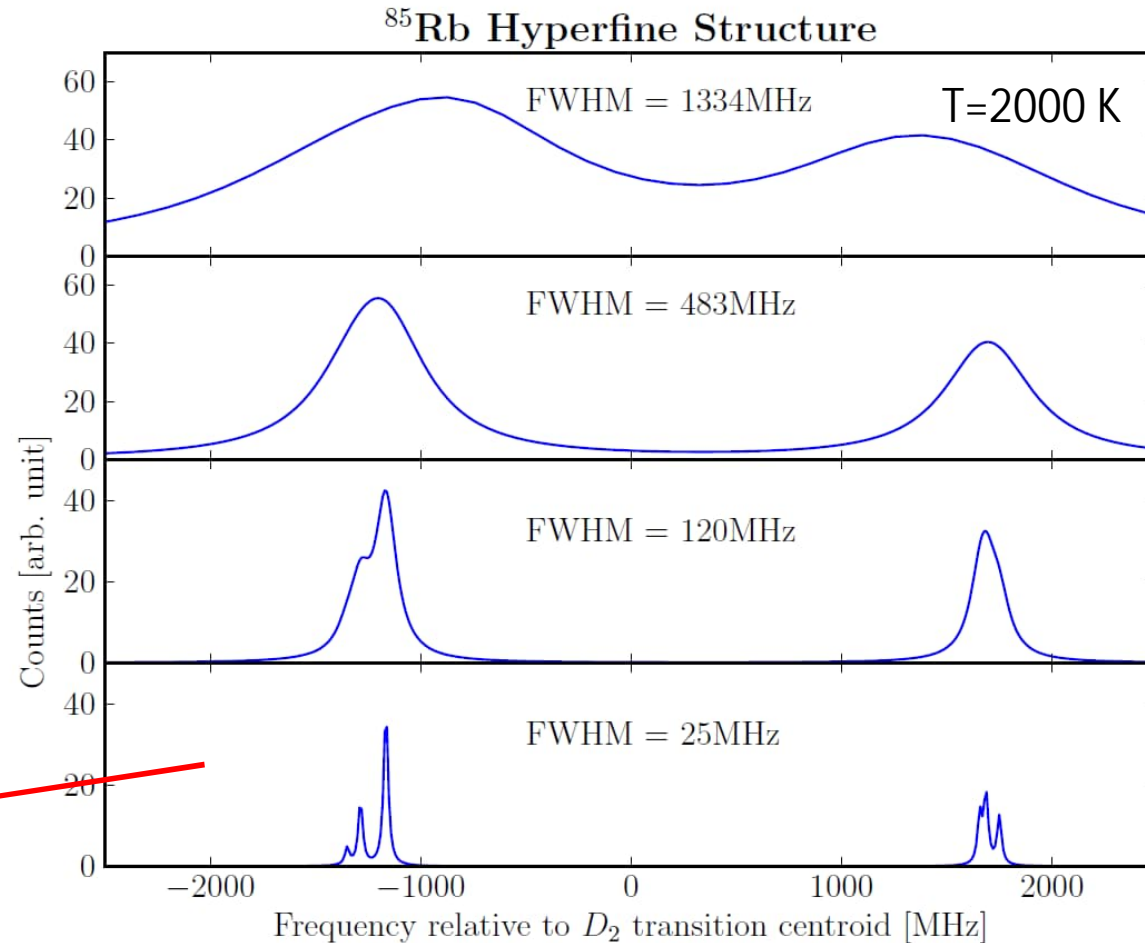
2. The energy spread δE (from source) remains constant

$$\delta E = \delta\left(\frac{mv^2}{2}\right) = mv\delta v = \text{const.}$$

3. The corresponding velocity spread is decreased. We obtain the Doppler width (in frequency):

$$\delta \nu_D = \nu_0 \frac{\delta E}{\sqrt{2eVm\dot{c}^2}}$$

The effect of the velocity compression

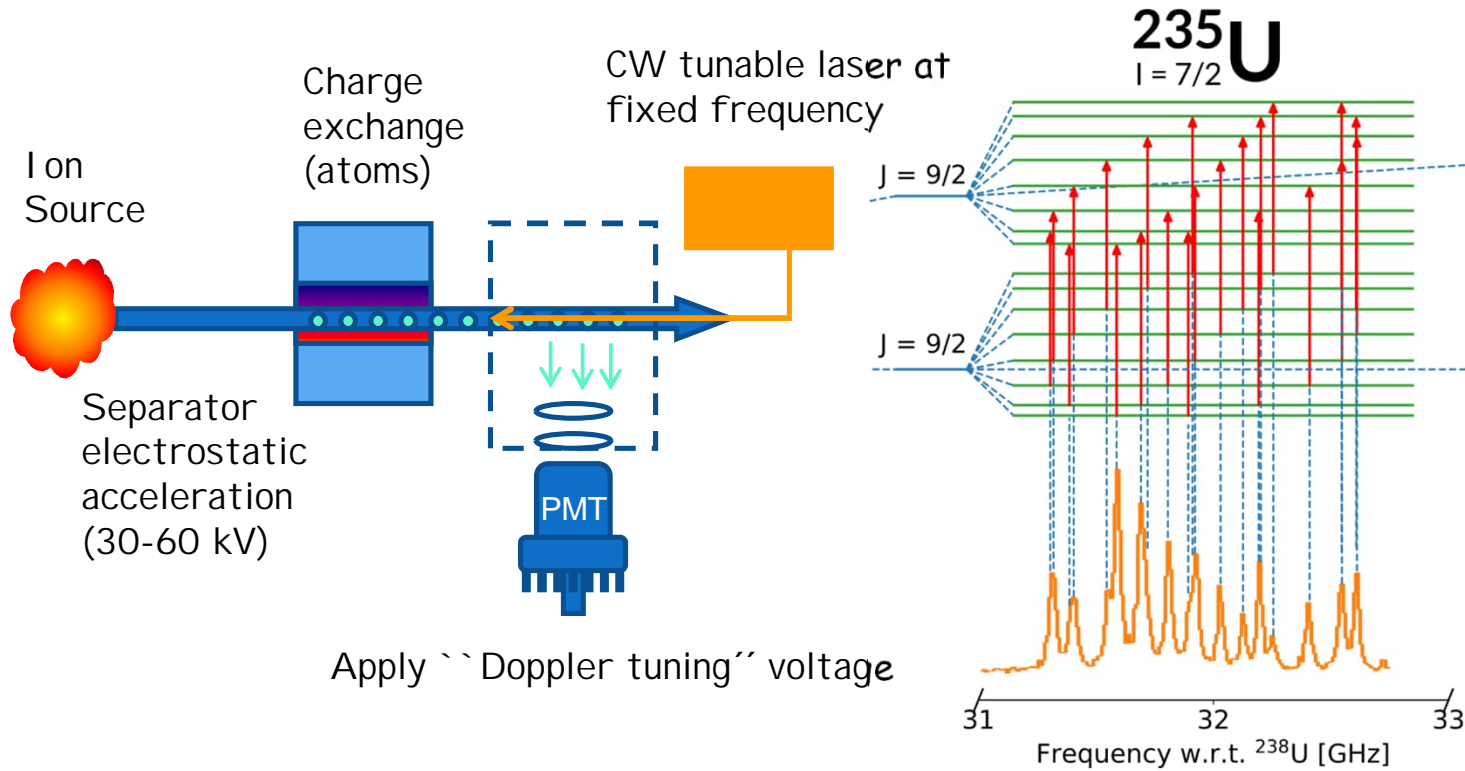


reduction in
Doppler
broadening

Natural linewidth
achieved!

Typical ion source energy spreads are ~ 1 eV. Acceleration of medium-mass nuclei to 30 keV produces a 3 order of magnitude velocity compression. Unresolved hyperfine structure becomes visible!

Doppler tuning the ion/atom beam



The collinear beams technique has high sensitivity. All ions/atoms pass through the laser beam and contribute to the fluorescent signal.

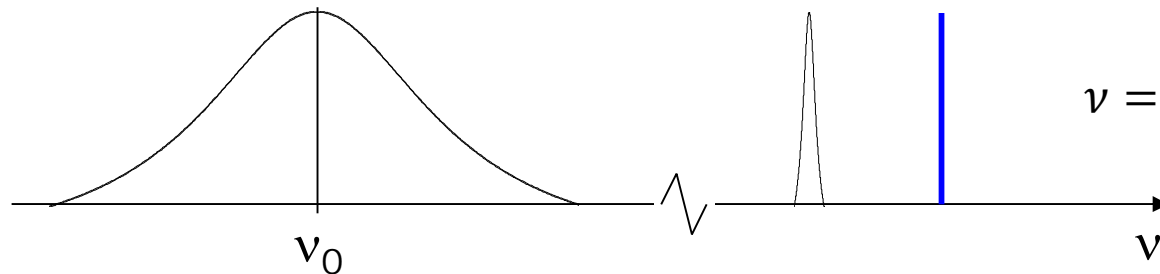
However.....

Signal (laser on resonance) = 1 photon detected per 1,000 ions in beam

Background (laser light scatter) = 200 photons / sec
(per mW of laser light)

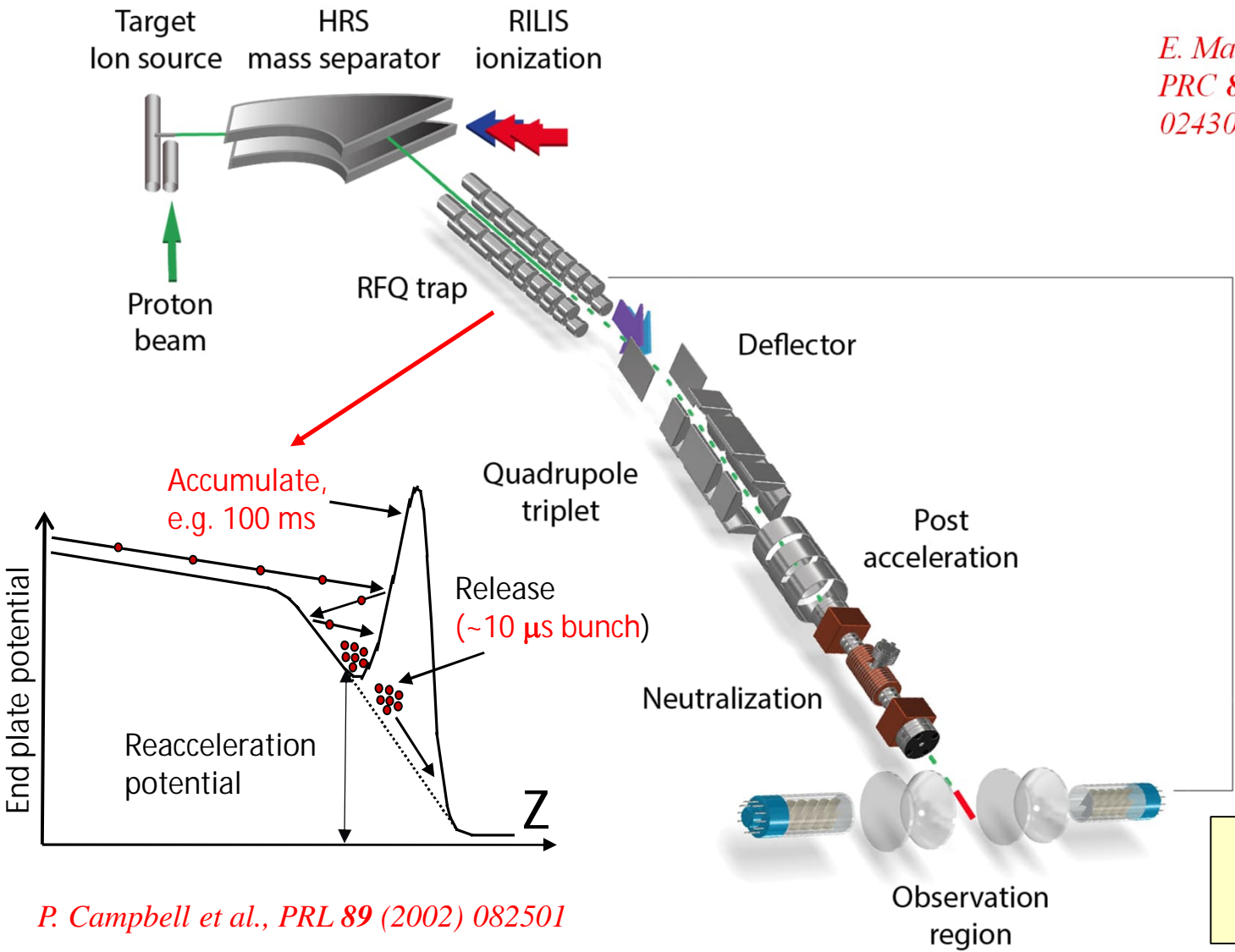
Low-flux beams (1,000 ions / sec): background must be suppressed to see signal.

Doppler-shifted (relative) frequency



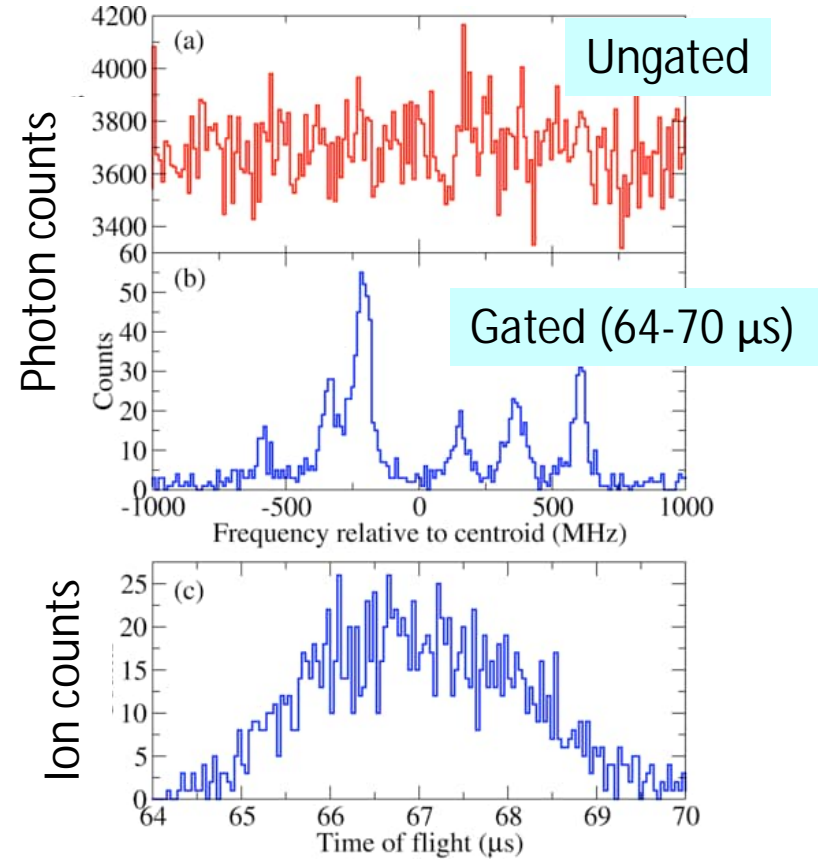
$$\nu = \nu_0 \frac{1 - \beta}{\sqrt{1 - \beta^2}}, \beta = \frac{v}{c}$$

From continuous to bunched beams



*E. Mané et al.,
PRC 84 (2011)
024303*

P. Campbell et al., PRL 89 (2002) 082501



Accept photons in a time window during which the bunched beam passes.

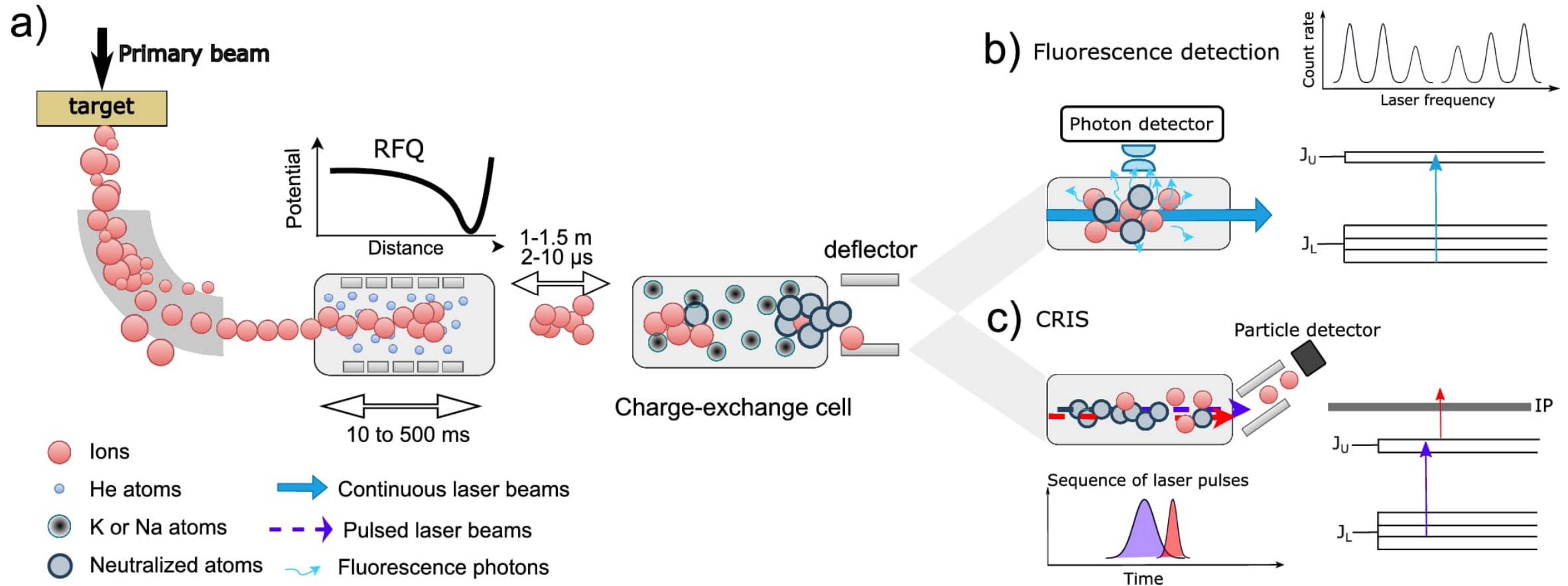
$$\text{Background suppression} = \frac{\text{e.g. 100ms accumulation}}{10\mu\text{s gate width}} \sim 10^4$$

From photon detection to ion detection

Production

Manipulation

Spectroscopy & detection



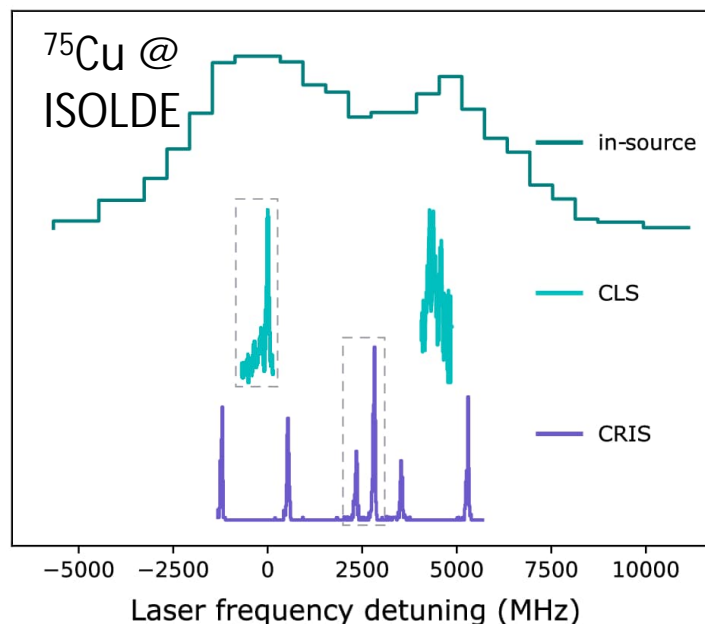
Note: the application of ion detection (via CRIS) requires charge exchange

Figure from A. Koszorus et al., EPJ A 60 (2024) 20

Summary of main laser spectroscopy techniques

The most suitable technique can be determined based on the scientific questions, spectroscopic properties and ion beam properties. The techniques are complementary to each other.

The most suitable facility can be determined based on the production yields and the isobaric contamination.

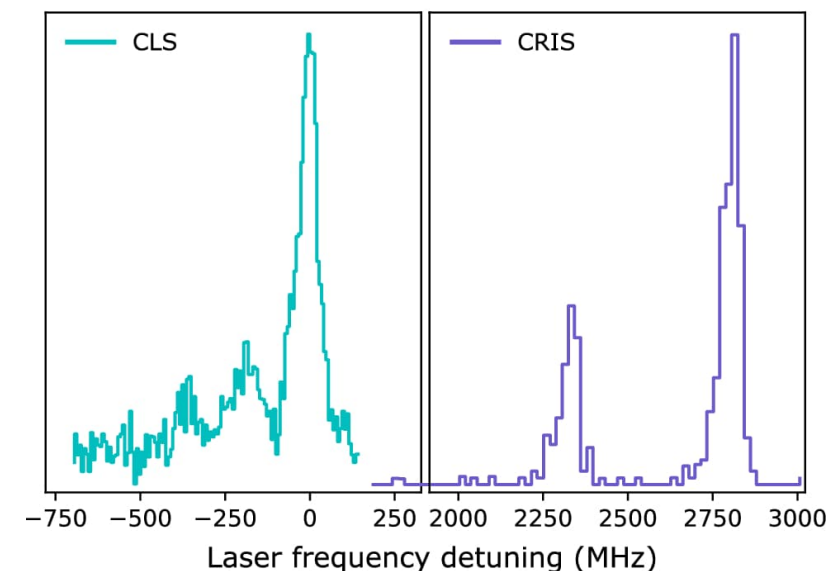


IN-SOURCE (RIS)

- Selective process
- Short lifetimes, low yields (<1 ion/s)
- High detection efficiency
- Poor resolution (100-1000× < CLS)

CLS/CRIS

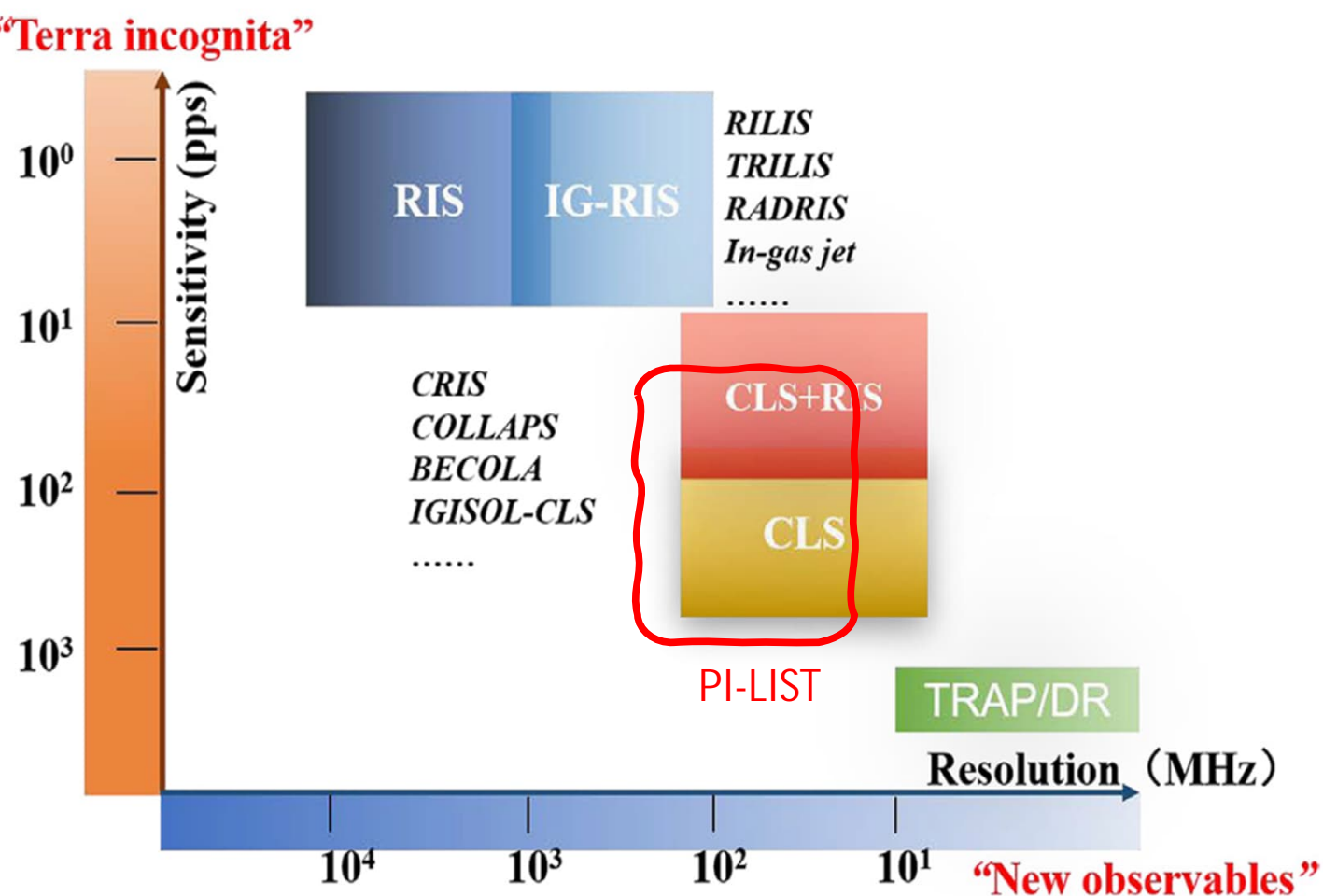
- High resolution: (~10-100 MHz)
- Beams of some $10^3 - 10^4$ ions/s (CLS)
- Few tens of ions/s (CRIS)



In-source data: U. Köster et al., PRC 84 (2011) 034320; Collinear fluorescence data: K.T. Flanagan et al., PRL 103 (2009) 142501; CRIS data: R.P. de Groote et al., PRC 96 (2017) 041302

Sensitivity vs resolution and when does it apply?

“Terra incognita”



GHz

- Magnetic dipole moments
- Electrical quadrupole moments and charge radii
- Hyperfine anomaly
 - / Distribution of magnetization inside nuclear volume
- Higher-order moments
 - / Magnetic octupole...
- Higher-order moments of the charge radii
 - / e.g., $\langle r^4 \rangle$ - surface thickness of nuclear density
- Beyond-standard model physics from Hz-level isotope shift spectroscopy

sub-Hz

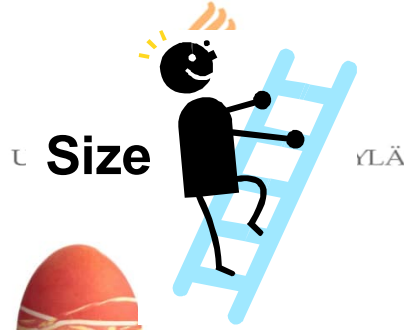
And we work in a typical lifetime range from ms to stable nuclei

Figure modified from: X.F. Yang et al., PPNP 129 (2023) 104005

A photograph of a laser laboratory setup. The scene is filled with various optical components, including mirrors, lenses, and beam splitters, all mounted on metal stands. Several bright light sources, likely lasers, are visible, casting a strong glow on the equipment. The overall atmosphere is technical and scientific.

Let's have a brief pause and enjoy this beautiful laser scenery!

What can we learn from the charge radii?



Reminder: $\delta v_i^{A,A'} = K_i \frac{m_{A'} - m_A}{m_A m_{A'}} + F_i \delta \langle r^2 \rangle^{A,A'}$

From a simple liquid droplet model approach – we can expand a deformed charge distribution in terms of spherical harmonics.

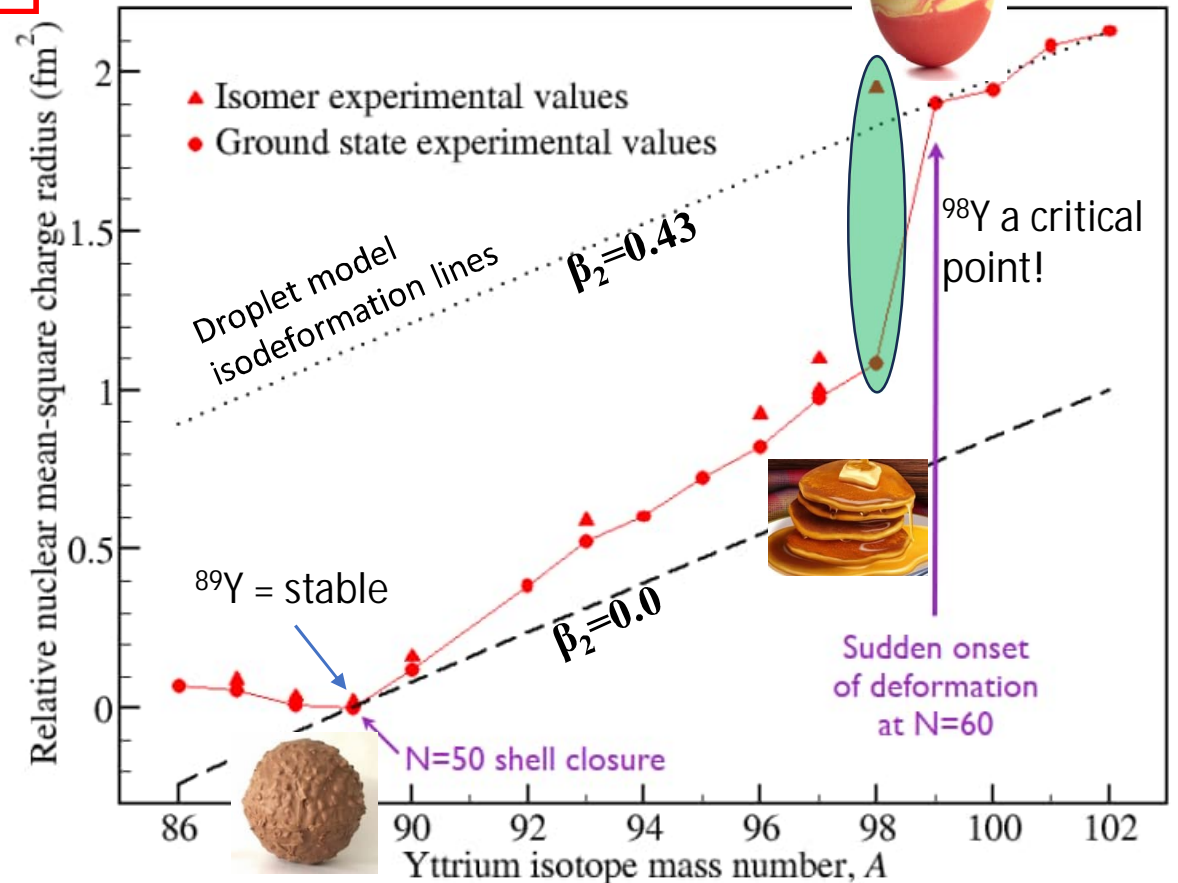
$$\langle r^2 \rangle = \langle r^2 \rangle_0 \left(1 + \frac{5}{4\pi} \sum_{i=2}^{\infty} \langle \beta_i^2 \rangle \right)$$

Quadrupole deformation parameter (shape)

$$\langle r^2 \rangle = \langle r^2 \rangle_0 \left(1 + \frac{5}{4\pi} (\langle \beta_2^2 \rangle + \langle \beta_3^2 \rangle + \dots) \right)$$

Radius of spherical nucleus of the same volume

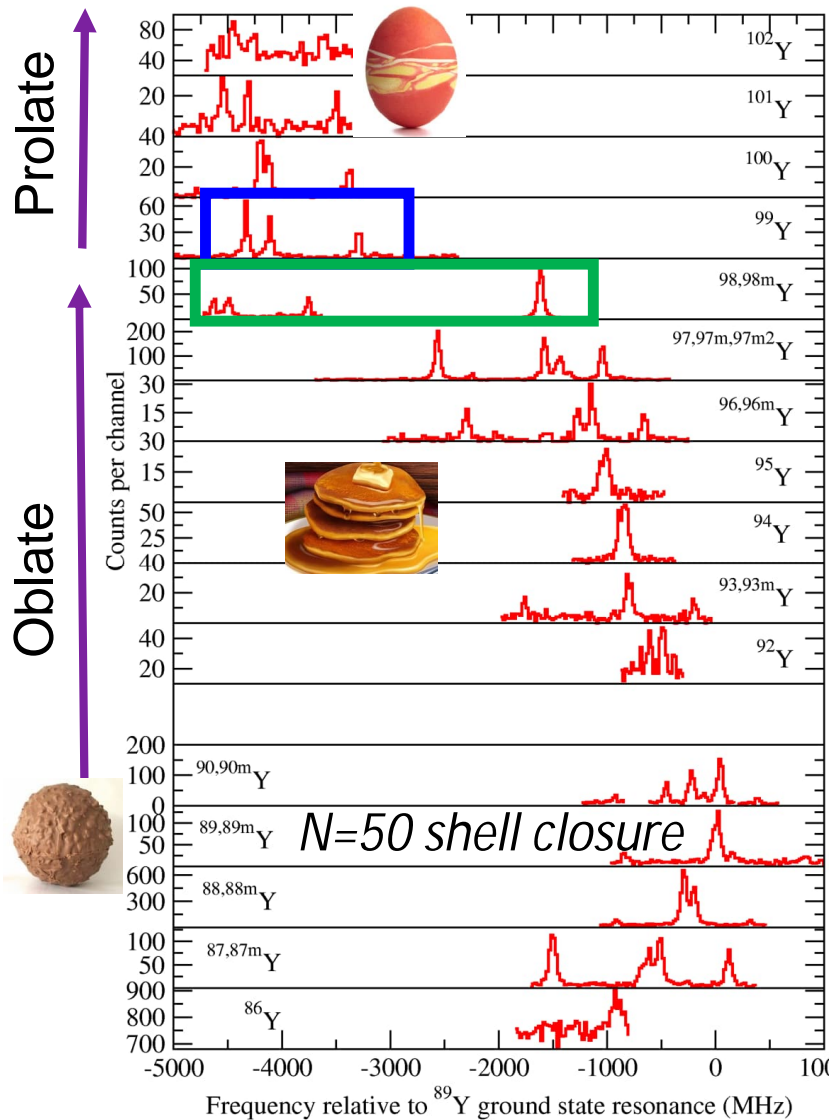
³⁹Yttrium



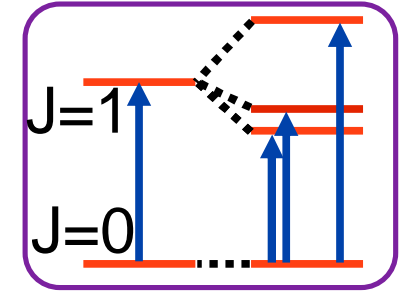
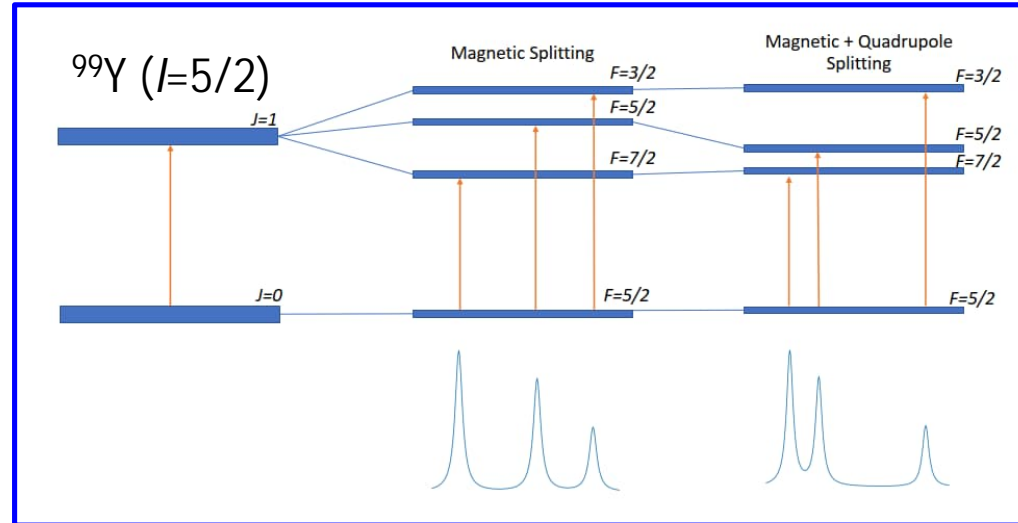
Note: charge radius is probing the deformation of ALL isotopes/states!

B. Cheal et al., Phys. Lett. B 645 (2007) 133

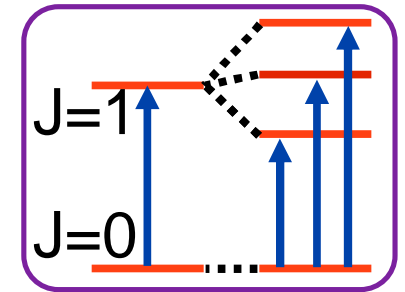
We can already identify trends in the raw data



Yttrium contains many isomeric (long-lived) nuclear states



Prolate

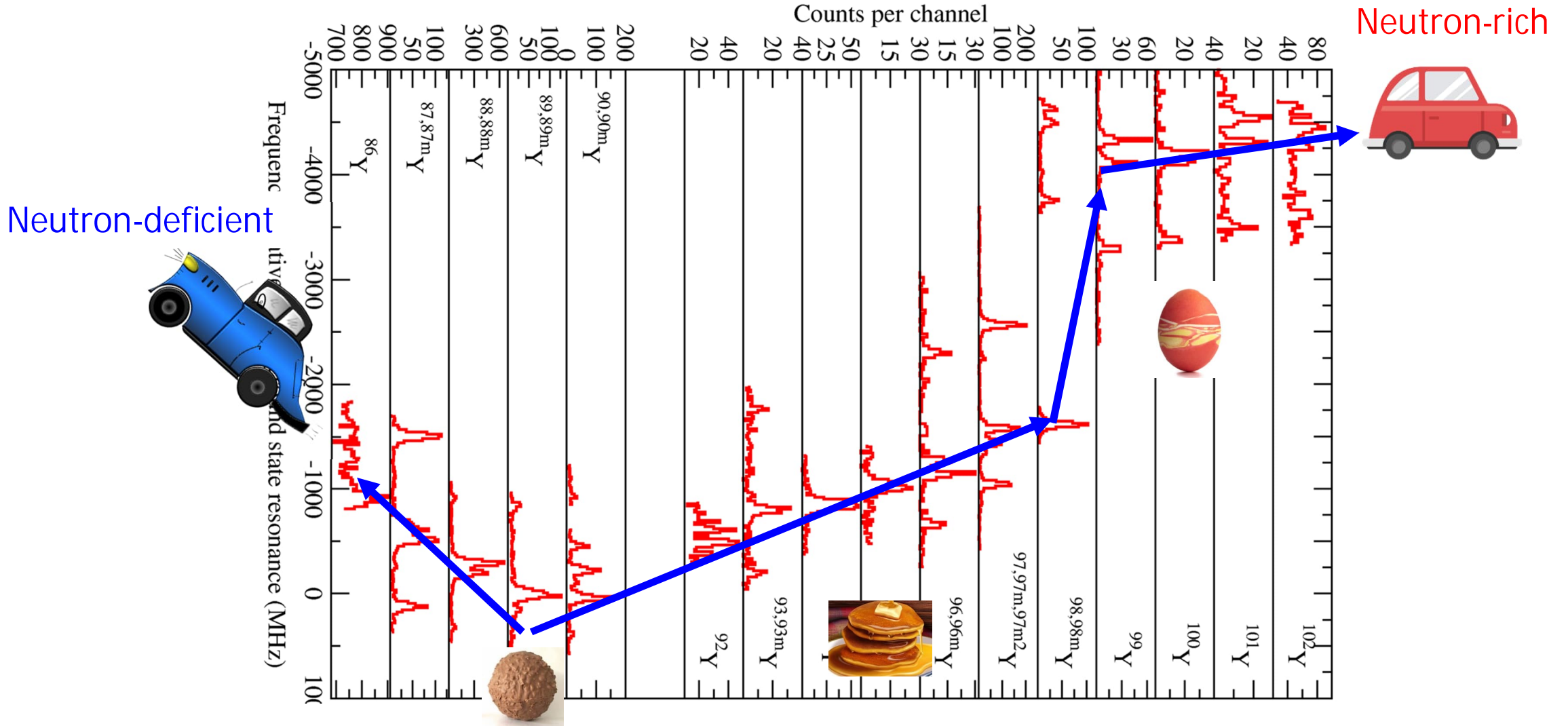


Oblate

- ^{98}Y is at a "critical point" whereby the ground state exhibits a weakly oblate shape; the isomer a rigid prolate shape – a "coexistence of shapes" in one nucleus

B. Cheal et al., Phys. Lett. B 645 (2007) 133

Isotope shifts to charge radii – a “simple” way



Deformation from the quadrupole moments



- If the nuclear spin has been assigned, e.g., via laser spectroscopy or non-optical measurements (γ -ray spec), the intrinsic quadrupole moment can be calculated:

$$Q_s = Q_0 \frac{3K^2 - I(I + 1)}{(I + 1)(2I + 3)}$$

- We then extract the quadrupole deformation parameter, β_2 , from:

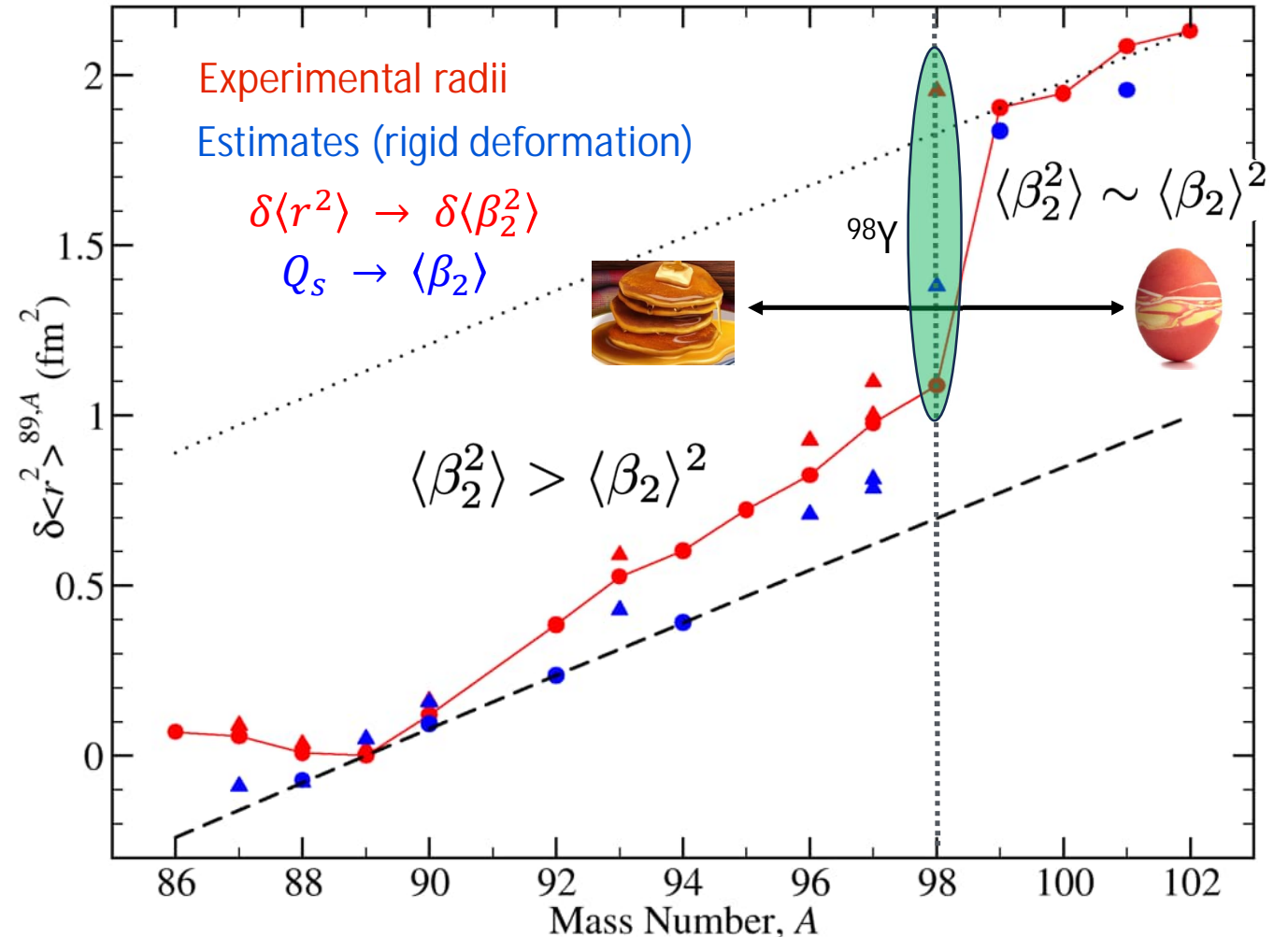
$$Q_0 \approx \frac{5Z \langle r^2 \rangle_{\text{sph}}}{\sqrt{5\pi}} \langle \beta_2 \rangle (1 + 0.36 \langle \beta_2 \rangle)$$

- The mean-square quadrupole deformation parameter, $\langle \beta_2^2 \rangle$, is extracted from our mean-square charge radius:

$$\delta \langle r^2 \rangle = \delta \langle r^2 \rangle_{\text{sph}} + \langle r^2 \rangle_{\text{sph}} \frac{5}{4\pi} \delta \langle \beta_2^2 \rangle$$

- Compare $\langle \beta_2^2 \rangle$ with $\langle \beta_2 \rangle^2$

Yttrium isotopes (Z=39)

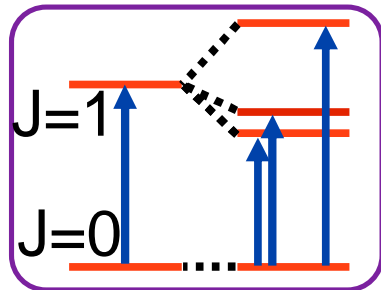


^{98}Y - Shape coexistence at the critical point

In our original spectroscopy, we used the ionic ground-state transition $5s^2\ ^1S_0 \rightarrow 4d5p\ ^1P_1$ (363 nm) for all Y isotopes*.

The problem of $J = 0 \rightarrow J = 1$ lines – each nuclear spin fits equally well.

*B. Cheal et al., *Phys. Lett. B* 645 (2007) 133



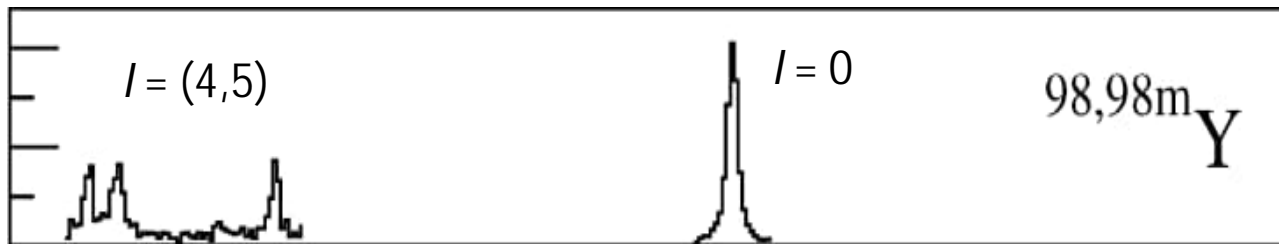
Prolate

3 peaks maximum for each nuclear state

A	I^π	A (MHz)	B (MHz)	$\delta\nu$ (MHz)
98m	(4)	-88.3(0.6)	+324.7(4.2)	-2746(3)
98m	(5)	-73.7(0.4)	+339.1(4.2)	-2735(3)

A	I^π	μ (μ_N)	Q_s (b)	$\delta\langle r^2 \rangle$ (fm^2)
98m	(4)	+2.98(2)	+1.73(19)	+0.863
98m	(5)	+3.11(2)	+1.80(20)	+0.860

- Three peaks tells us 3 unknowns (only)



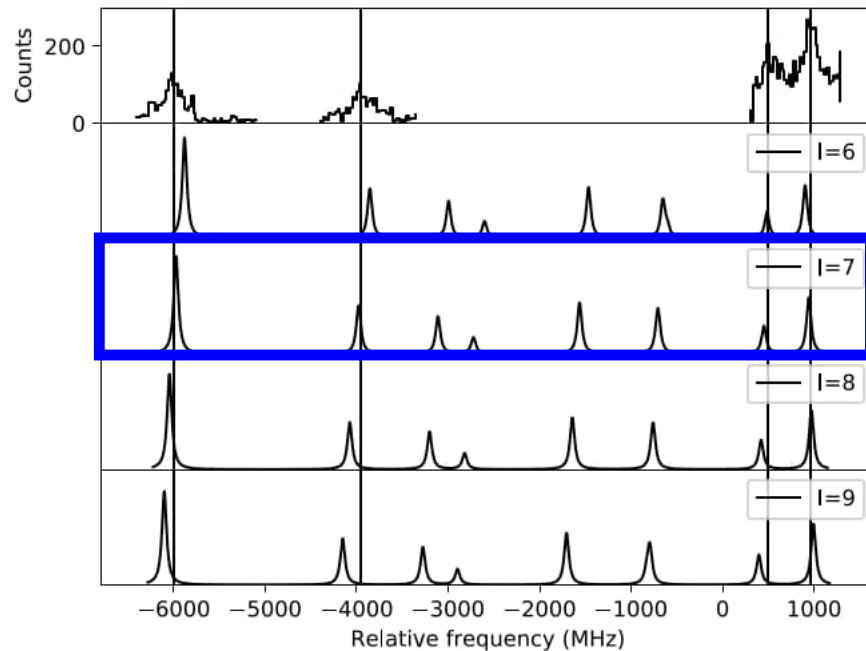
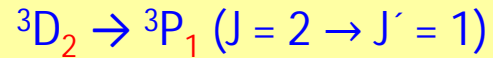
- The nuclear spin was assumed to be either $I = 4$ or $I = 5$
- **GAMMASPHERE gamma-ray spectroscopy following β decay of ^{98m}Y indicated the spin was likely to be $I = (6,7)^+$

**W. Urban et al., *PRC* 96 (2017) 044333

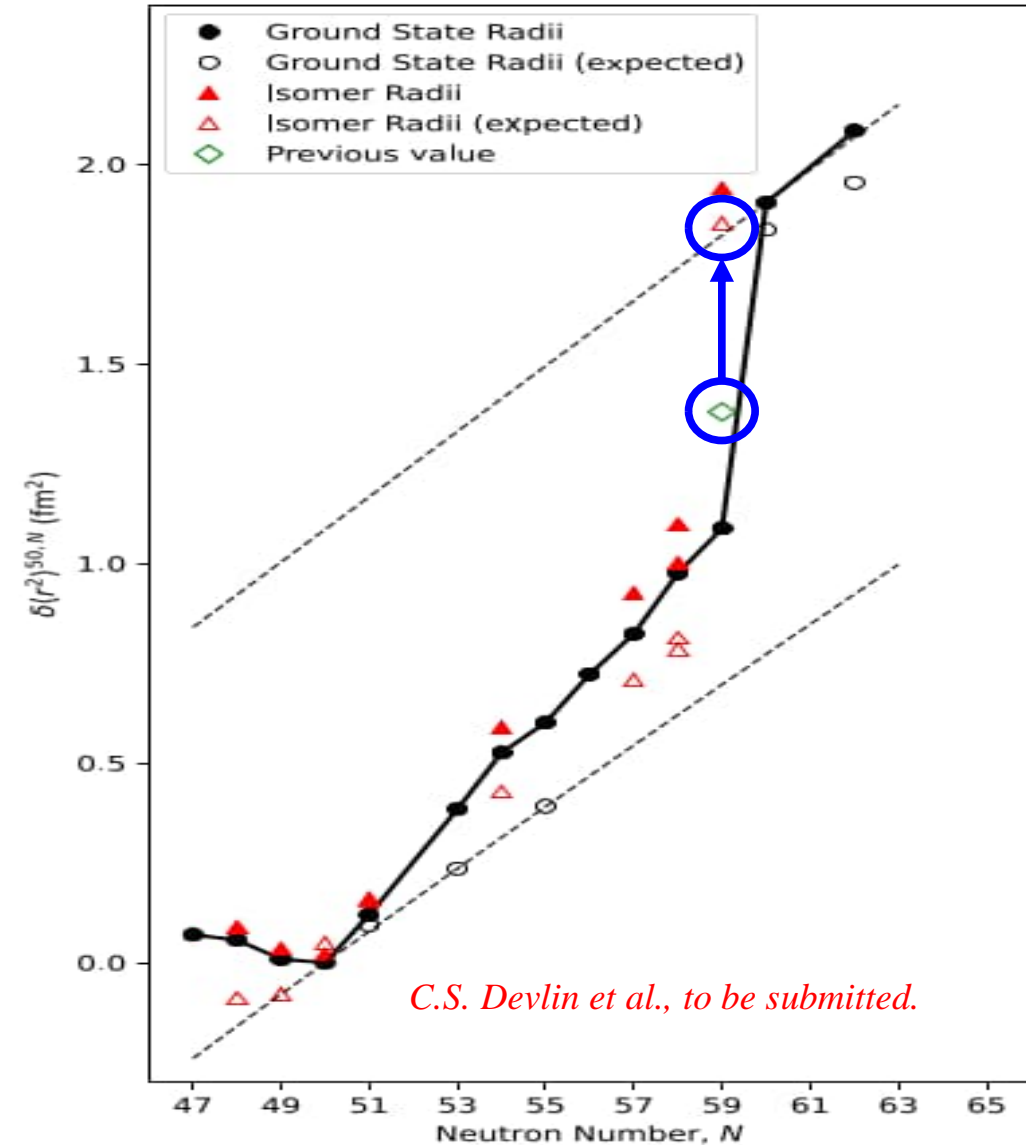
Revisiting ^{98}Y – illustrating the sensitivity of spin I



Now we measure on a transition with higher J values

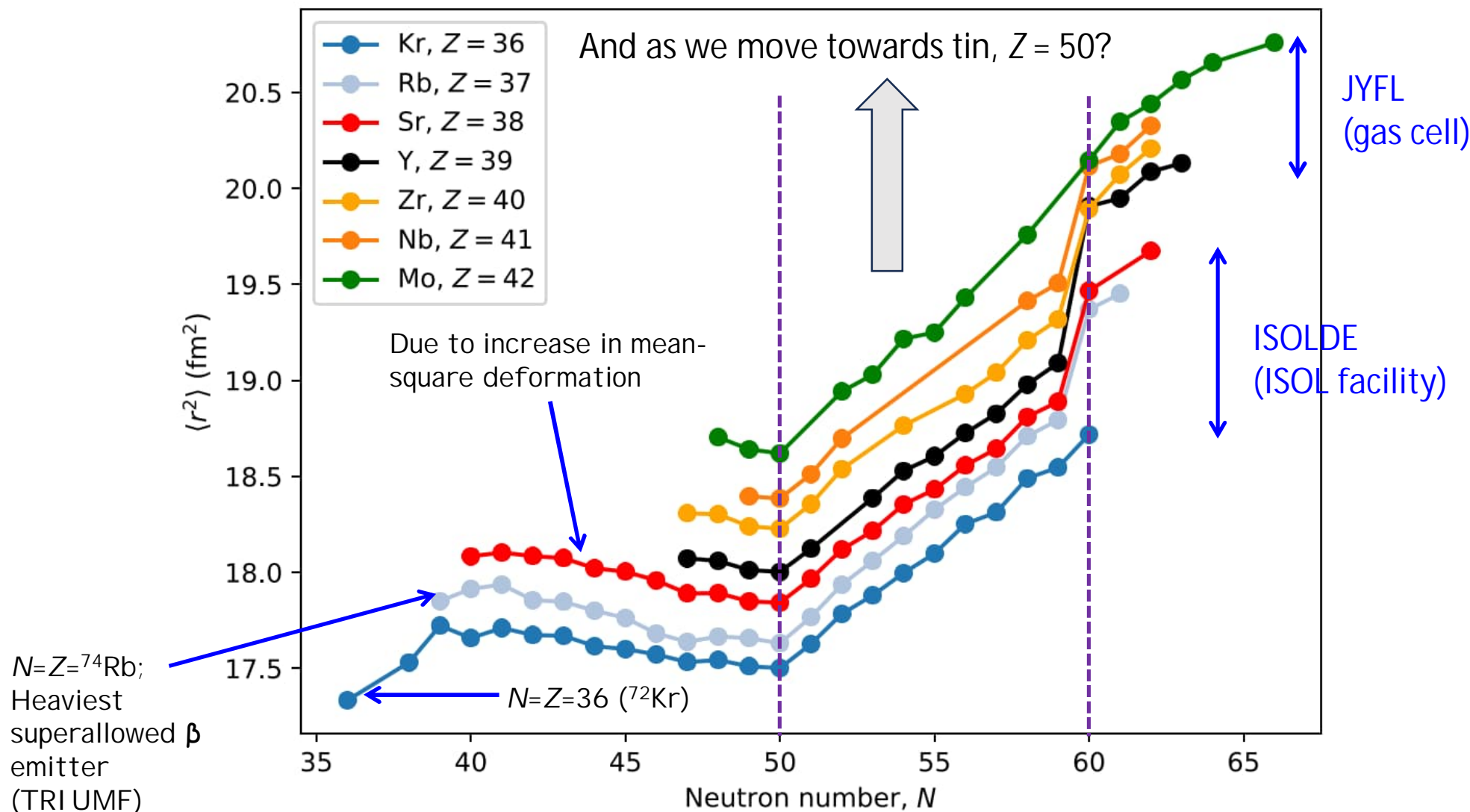


- Firm spin assignment, $I = 7$, allowing for revised hyperfine A and B parameters, thus nuclear moments.
- The isomer has a much higher quadrupole moment than previously thought – a strong (prolate) deformation that is very rigid.



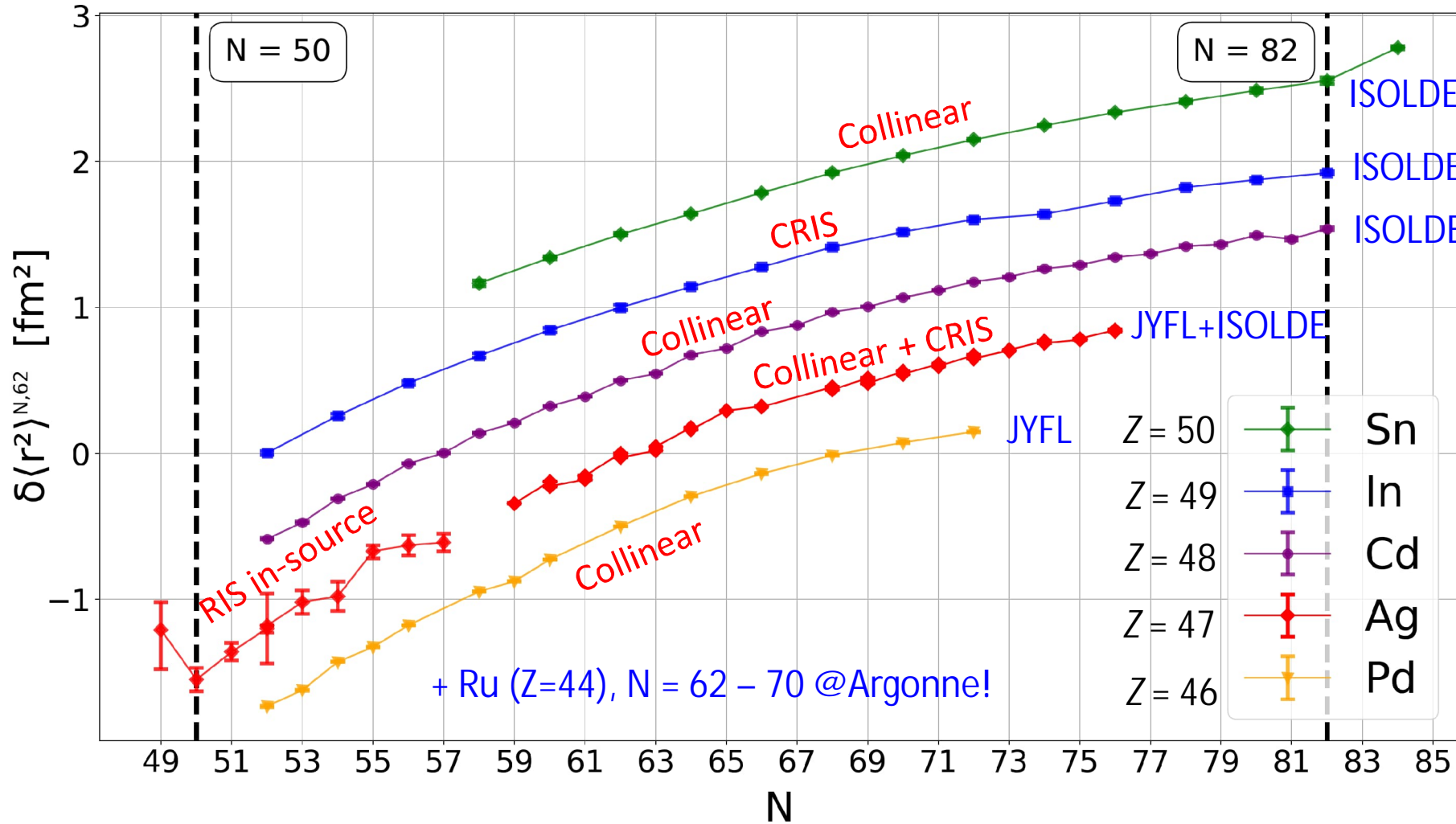
C.S. Devlin et al., to be submitted.

Charge radii systematics (shape change region)

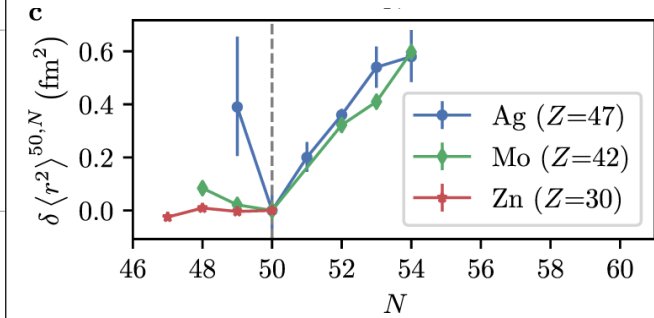


Overview figure: Nörtershäuser & Moore, Handbook of Nuclear Physics (2023)

Charge radii systematics in the tin region

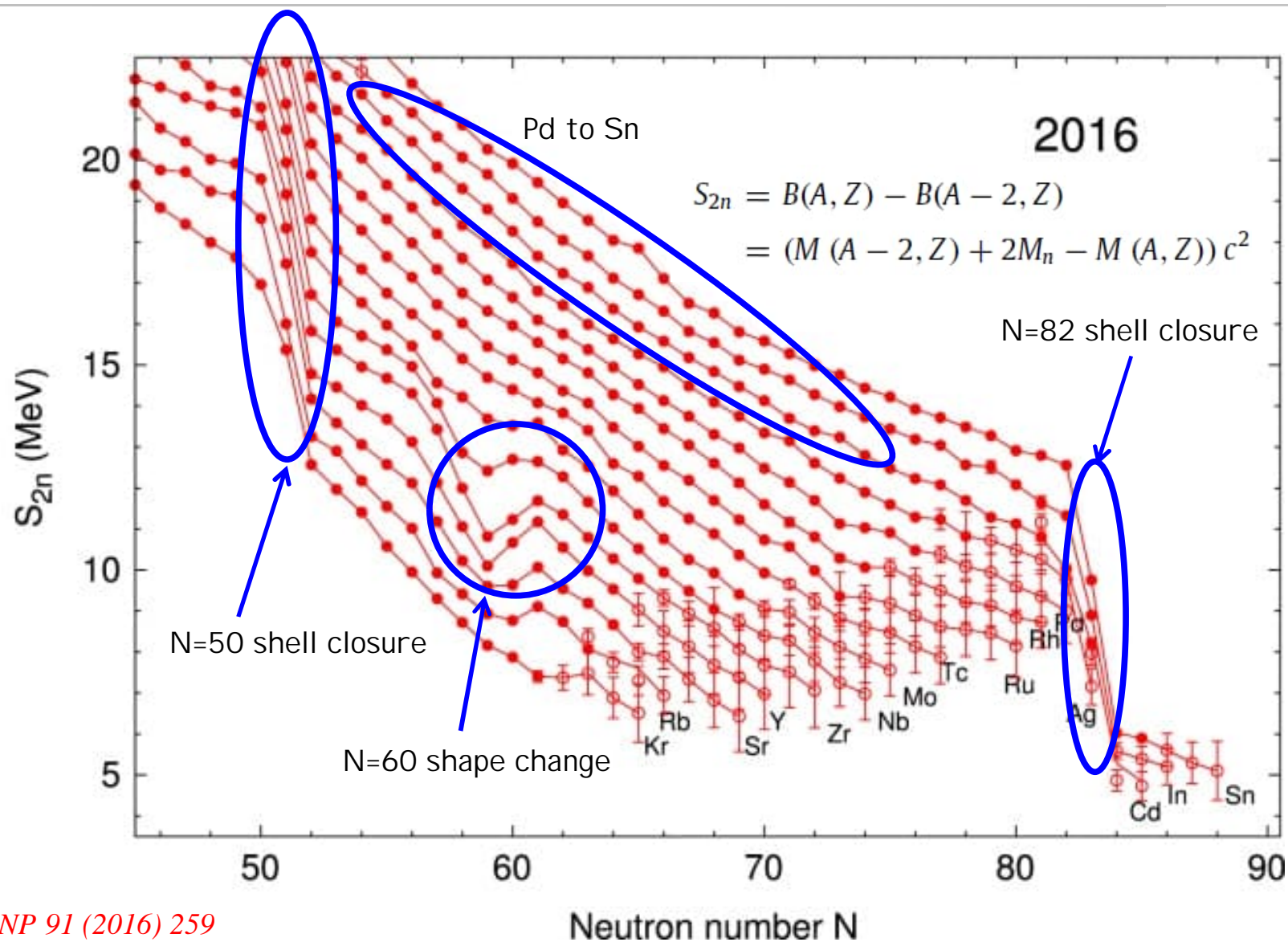


- Sn: *C. Gorges et al., PRL 122 (2019) 192502*
- In: *J. Karthein et al., arXiv.2310.15093 (2023)*
- Cd: *M. Hammen et al., PRL 121 (2018) 102501*
- Ag: *M. Reponen et al., Nat. Comm. 12 (2021) 4596 & to be submitted*
- Pd: *S. Geldhof et al, PRL 128 (2022) 152501*



Possible trend in the "kink" at $N = 50$?

Complementarity with the nuclear mass surface



T. Eronen et al., PNP 91 (2016) 259

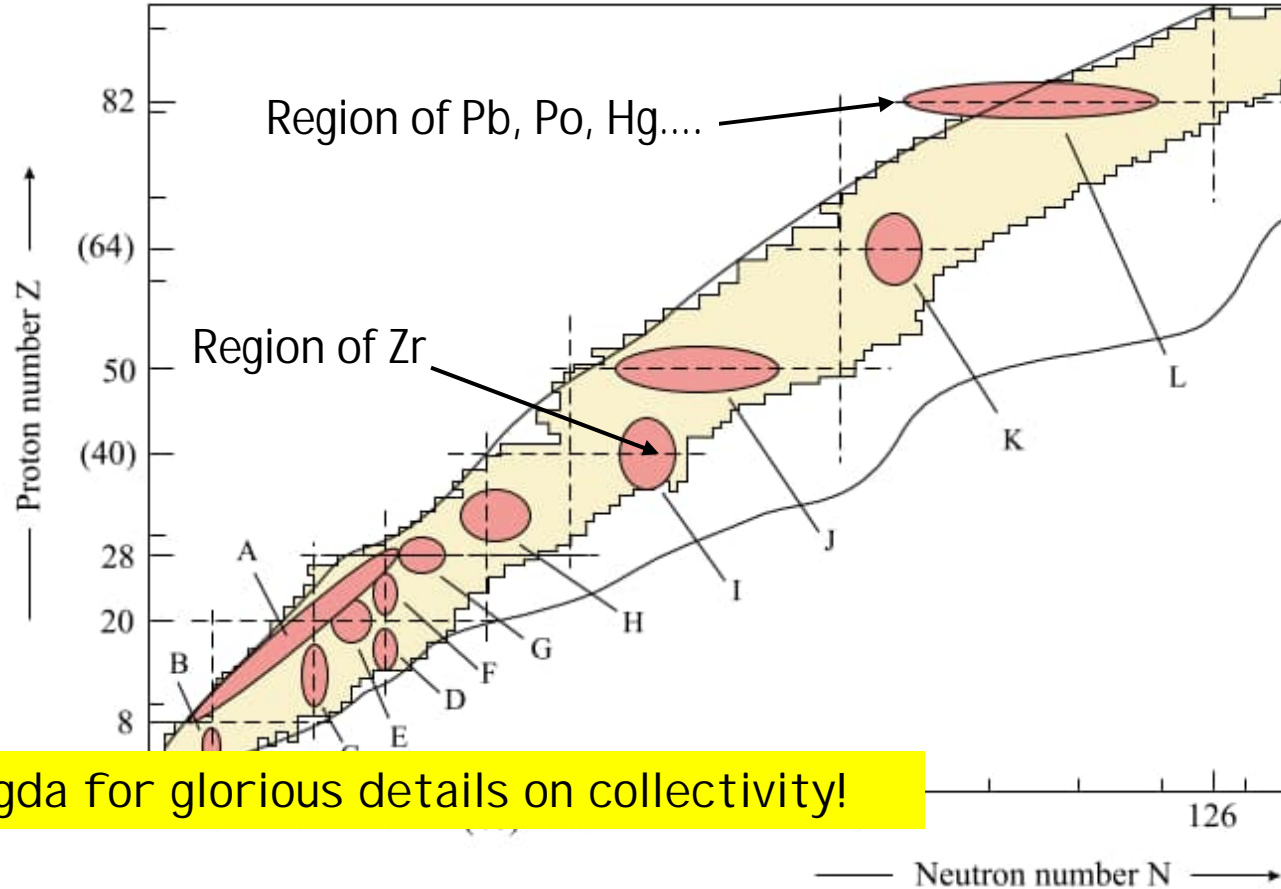
Let's finish with a little more shape coexistence



- Shape coexistence appears to be unique in the realm of finite many-body quantum systems.
- Different “shapes” coexist at similar excitation energies.

Shell Model picture:
Coexistence of “normal” and “intruder” structures

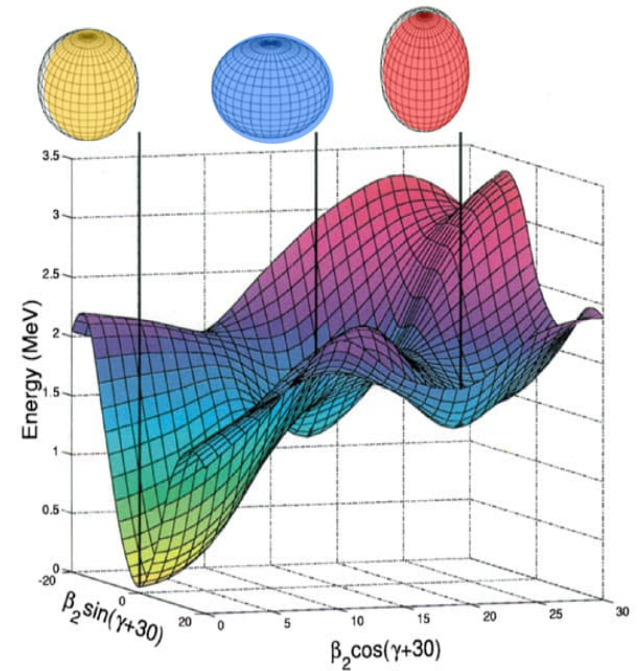
Mean field picture:
Several minima in energy surface vs deformation



See Magda for glorious details on collectivity!

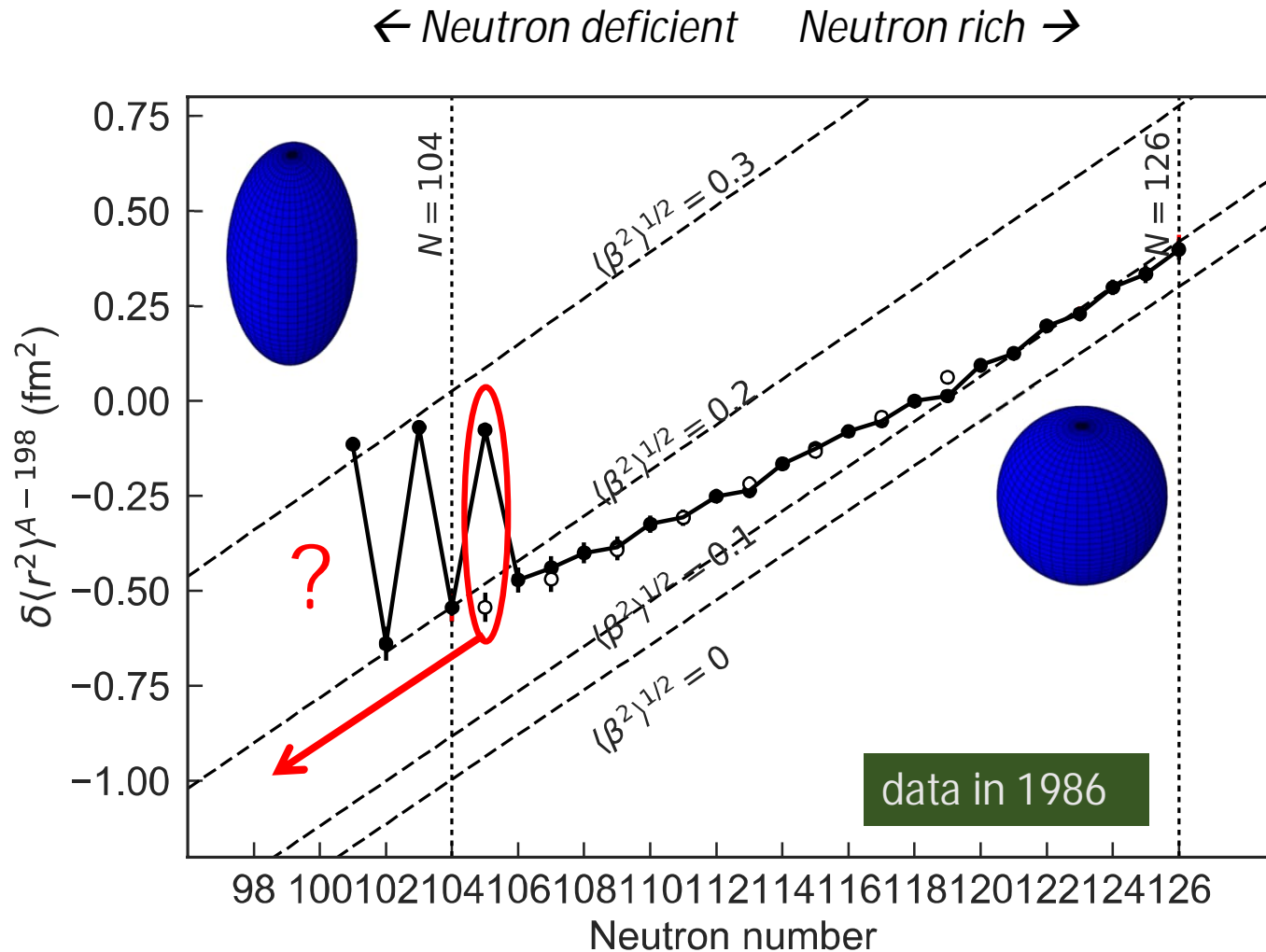
K. Heyde and J.L. Wood, Rev. Mod. Phys. 83 (2011) 1467

P.E. Garret, M. Zielinska & E. Clement, PPNP 124 (2022) 103931



A. Andreyev et al. Nature 405 (2000) 430

Shape staggering in the charge radii of Hg isotopes

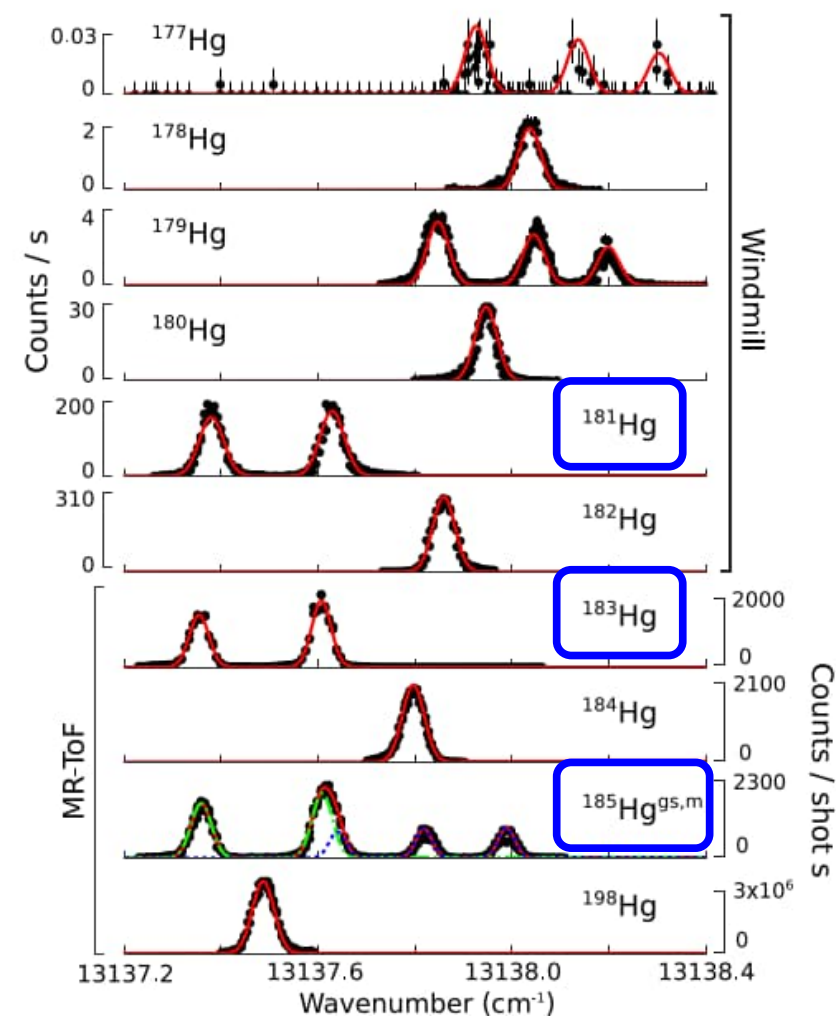
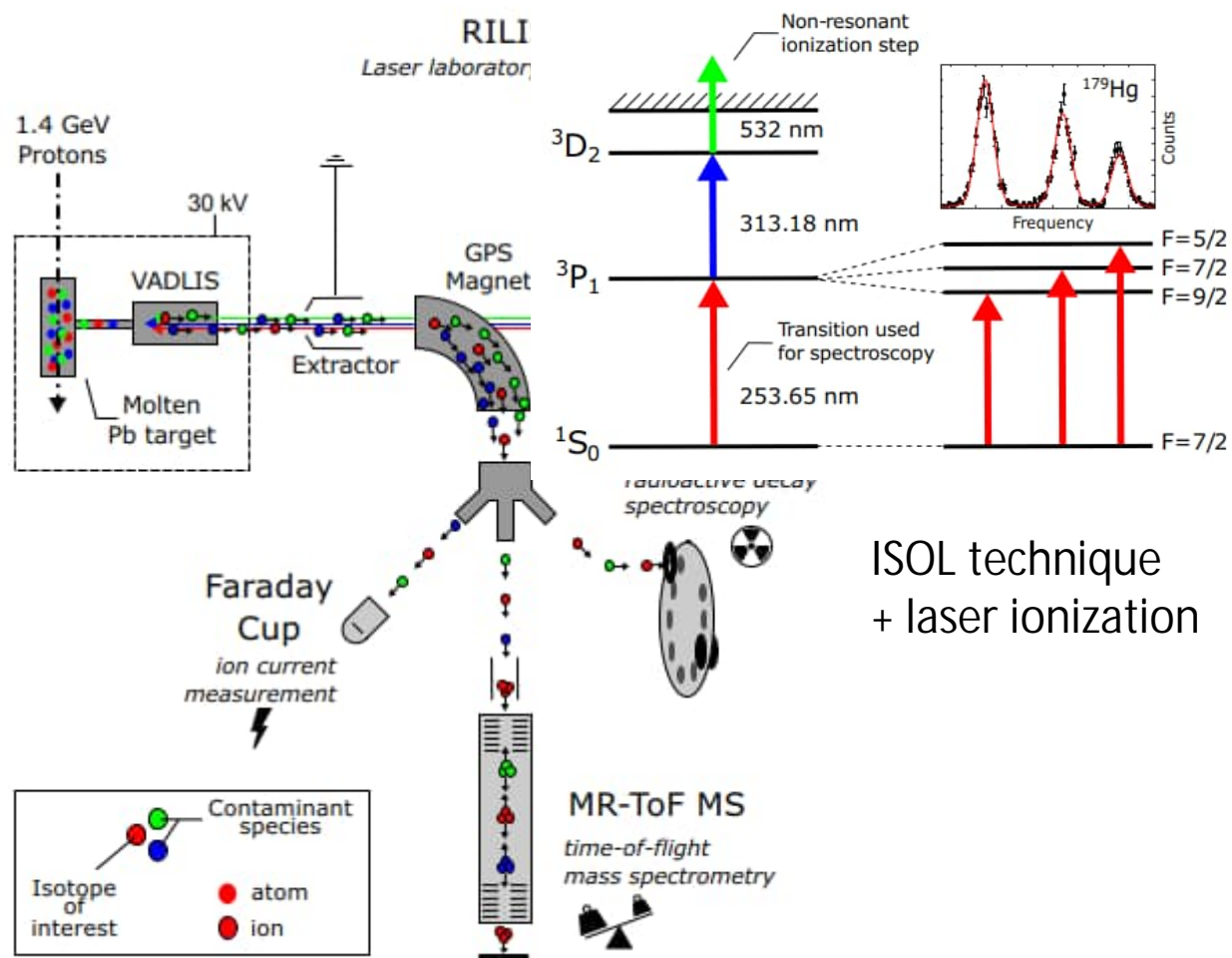


Huge increase in charge radius around the neutron mid-shell ($N=104$);

$^{181,183,185}\text{Hg}$
($N=101,103,105$)

Shape coexistence established in ^{185}Hg !

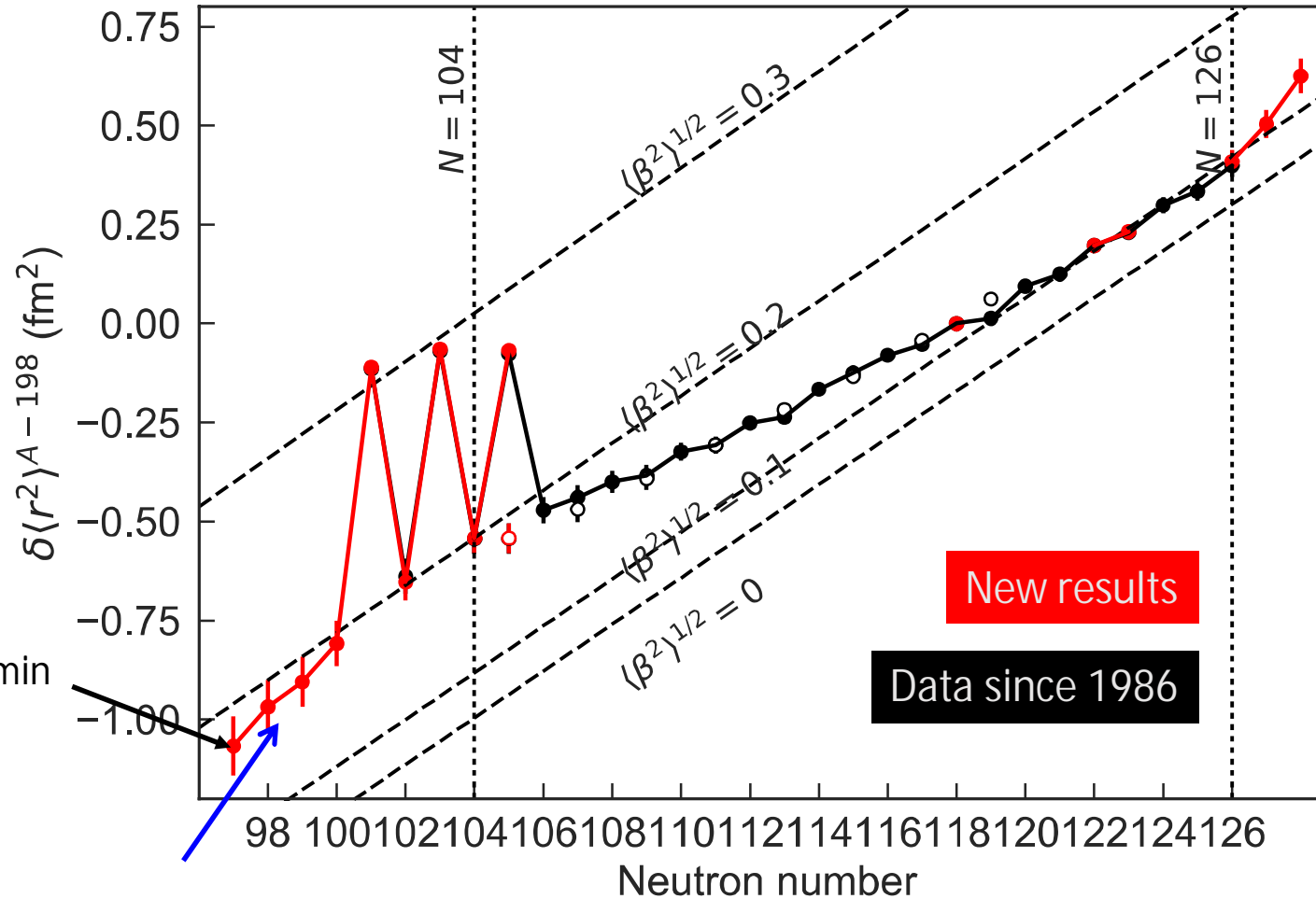
When does the staggering end?



Combining detection in three different experimental stations

B. Marsh et al., Nature Phys. 14 (2018) 1163, S. Sels et al., Phys. Rev. C 99 (2019) 044306

After 30 years of developments...



^{177}Hg ($N=97$)
Production few ions/min
 $T_{1/2} = 127$ ms.

- Excellent agreement with older data.
- Combination of large-scale Monte Carlo shell-model and DFT calculations performed.

End-point of staggering observed; Hg isotopes return to more spherically-shaped trend.

B. Marsh et al., Nature Phys. 14 (2018) 1163, S. Sels et al., Phys. Rev. C 99 (2019) 044306

The neighboring chains

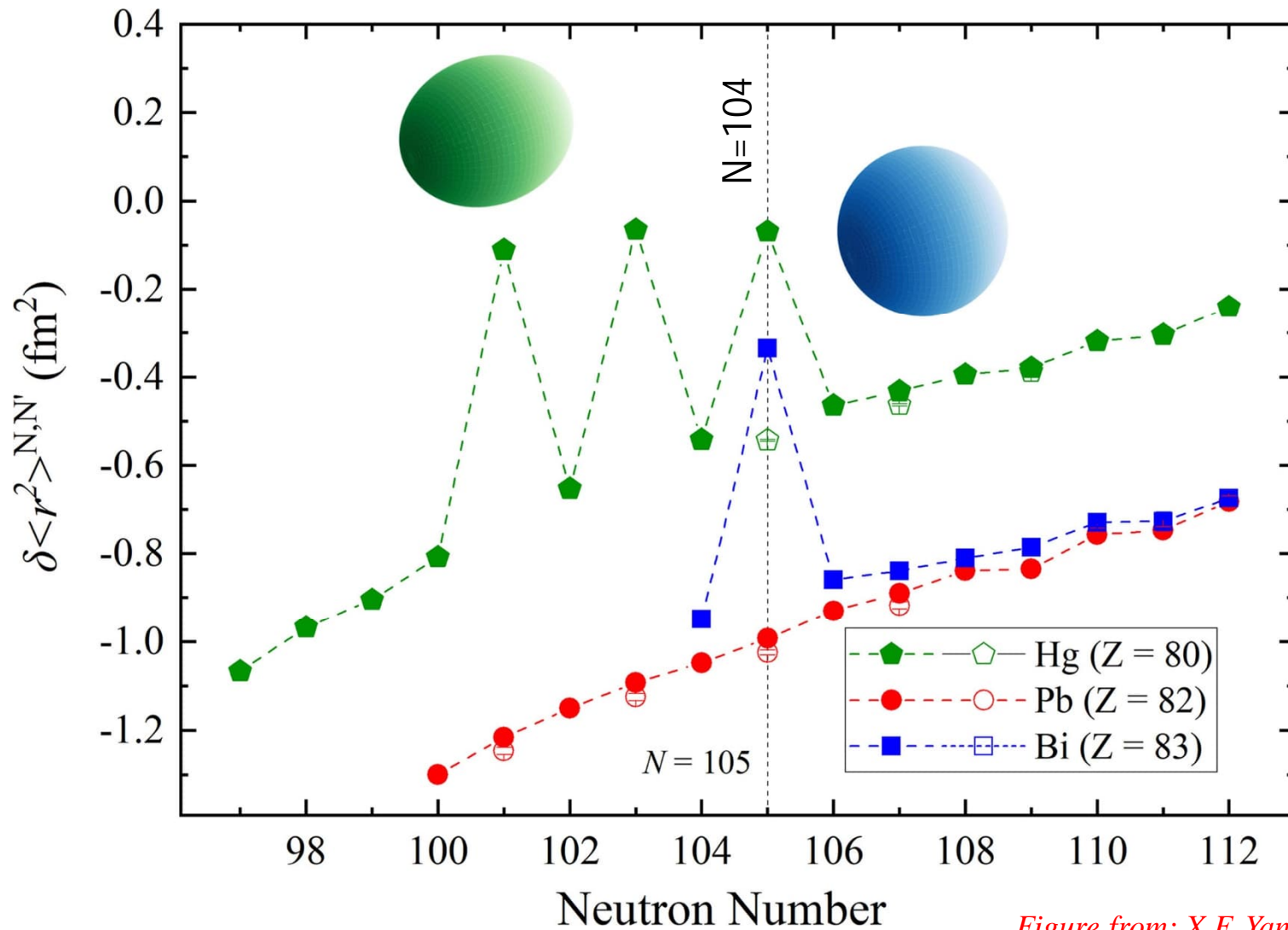


Figure from: X.F. Yang et al., PPNP 129 (2023) 104005.

Take home messages and summary



Size



**Electromagnetic
property**



Shape



Is it excited or not?

- Laser spectroscopy of radioactive nuclei features (or is planned) at almost all online radioactive ion beam facilities.
- Our spectroscopy can be performed at both ISOL (traditional) as well as fragmentation facilities (via gas catcher developments).
- Element selectivity is critically important in RIB production and laser ion sources are widely used/planned (ISOL).
- Lower-resolution in-source methods are complementary to high-resolution techniques and are often used “together”.

Take home messages and summary



Size



**Electromagnetic
property**



Shape



Is it excited or not?

- Laser spectroscopy is quite unique in being able to *measure* nuclear spins.
- If additional structure is observed there must exist another state (``long-lived``) which can be elusive to decay spectroscopy.
- Laser spectroscopy gives a measure of quadrupole deformation via the quadrupole moment & the mean-square charge radius. The difference gives an indication of ``softness`` / ``rigidity`` of the deformation.
- Not discussed: nuclear moments are a sensitive probe of the wave function configuration.
- These complementary (and model-independent) probes are an essential benchmark and guide for nuclear theory (ab-initio, shell model, DFT...)

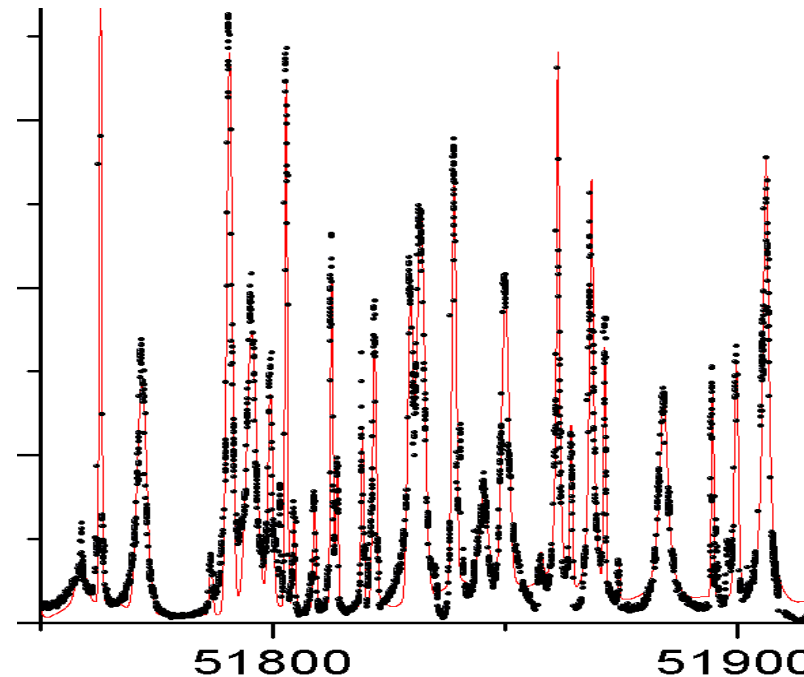
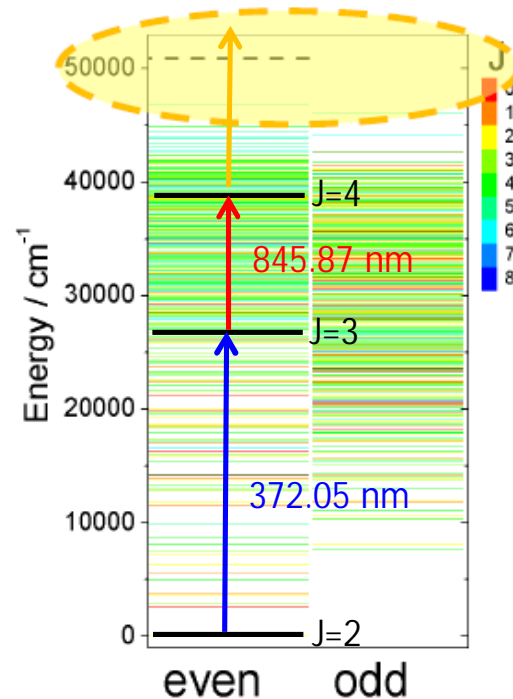
Back up material for lecture 2

How do we develop the laser ionization scheme?

Literature Search

On-line atomic spectral line databases, published spectroscopy work.

- R.L. Kurucz' CD-ROM 23 Atomic Line Database: <http://www.pmp.uni-hannover.de/cgi-bin/ssi/test/kurucz/sekur.html>
- NIST atomic spectral line database: <http://www.nist.gov/pml/data/asd.cfm>
- Blaise and Wyart (actinides): <http://web2.lac.u-psud.fr/lac/Database/Contents.html>



V. Sonnenschein, I.D. Moore et al., EPJ A 48 (2012) 52

How do we quantify the optical selectivity?



The excitation probability of an atom in a laser beam whose frequency is tuned near resonance:

$$P \propto \frac{1}{\delta^2 + \frac{\Gamma^2}{4}}$$

$$\delta = \omega_L - \omega_0,$$

Γ is the interaction linewidth

When the laser is in resonance with a selected isotope and but far from other "contaminating" elements or isotopes (Δ), the selectivity S

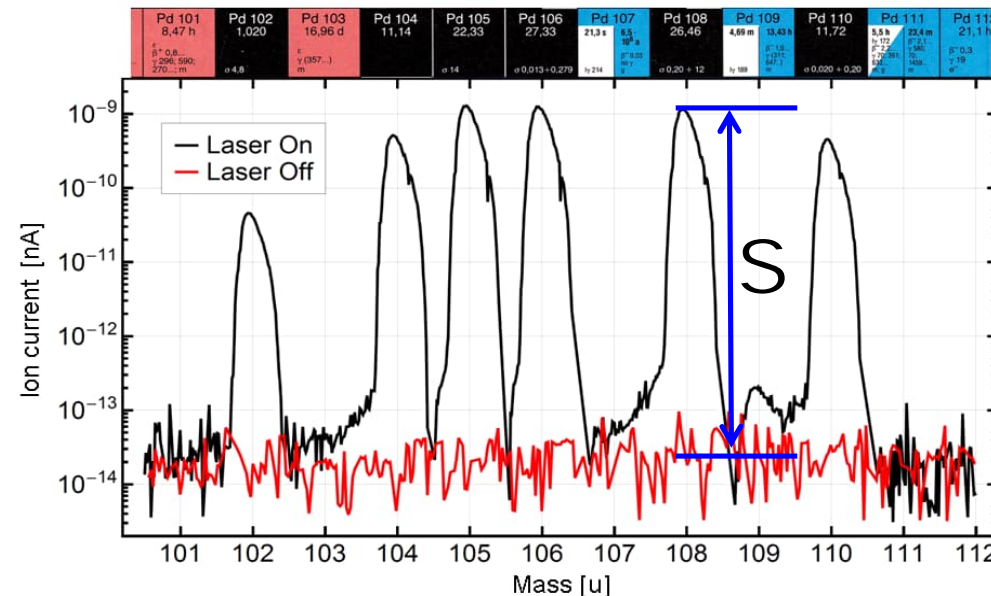
$$S \sim 4 \times \frac{\Delta^2}{\Gamma^2}$$

Pd isotopes

$\Gamma \sim 3$ MHz, $\Delta \sim 100$ MHz (neighbouring isotopes):
 $S \sim 4000$

$\Delta \sim 10^{15}$ Hz (palladium to silver):
 $S \sim 10^{17}$!!!

Multi-step excitation: $S = S_1 \cdot S_2 \dots \cdot S_n$.

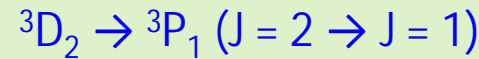
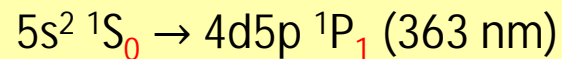
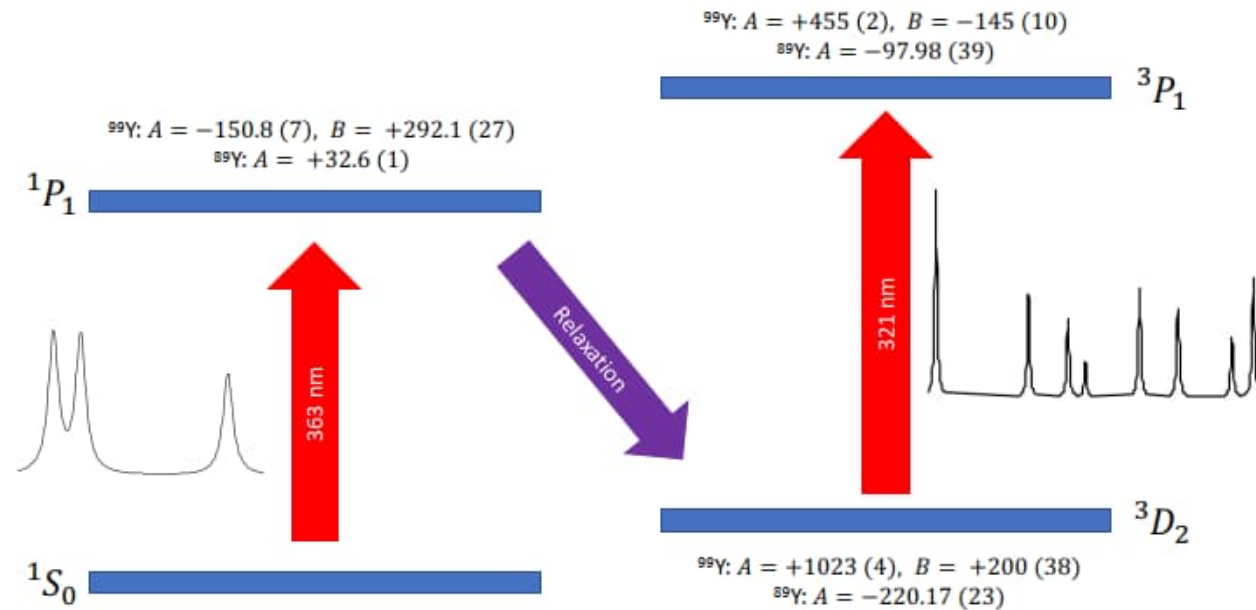


Mass scan over all stable Pd isotopes

Preparing to revisit ^{98}Y



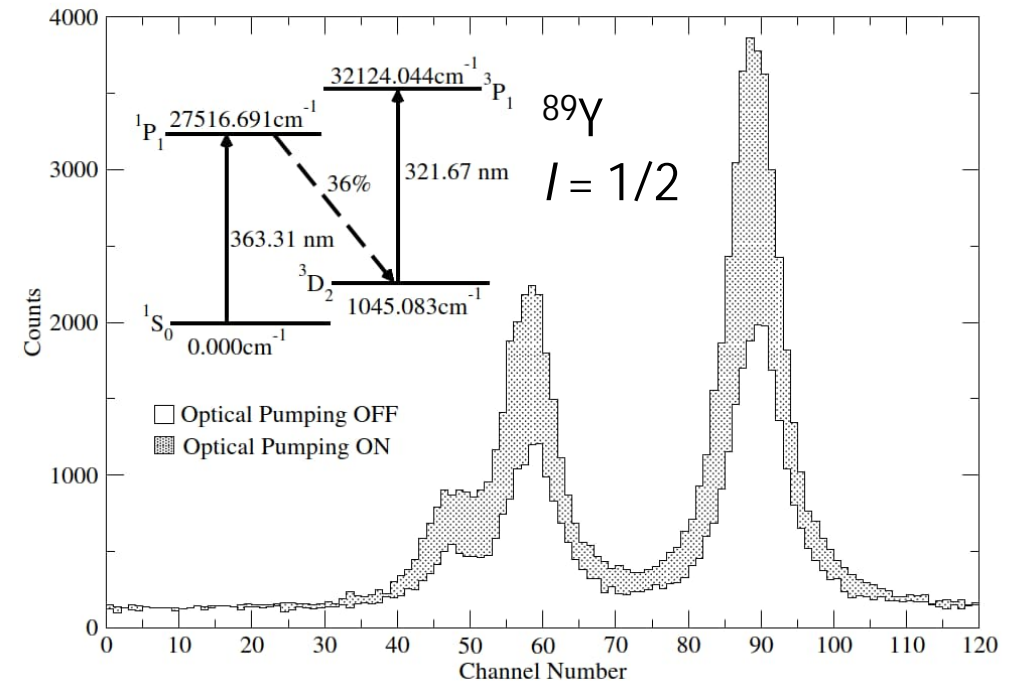
Perform laser spectroscopy on a transition with higher J values



To boost the natural population of the low-lying metastable state ($3D_2$) at 1045 cm^{-1} , we optically pumped population into the state from the ground state. This increased the efficiency for online measurements.

Nuclear spin determination of ^{100m}Y by collinear laser spectroscopy of optically pumped ions

K Baczynska¹, J Billowes², P Campbell², F C Charlwood², B Cheal², T Eronen³, D H Forest¹, A Jokinen³, T Kessler³, I D Moore³, M Ruffer¹, G Tungate¹ and J Äystö³



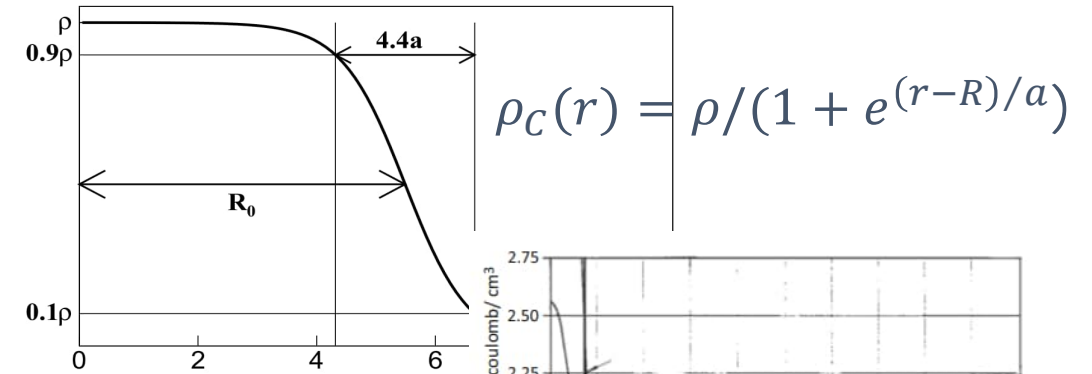
K. Baczynska et al., J. Phys. G 37 (2010) 105103

Charge radii: back to basics

The majority of modern day experiments apply electromagnetic probes to obtain (**absolute**) **nuclear radius** information – preferably point-like, e.g., **electrons and muons**.

The mean-square charge radius can be defined as:

$$\langle r^2 \rangle = \frac{\int r^2 \rho_{ch}(r) dV}{\int \rho_{ch}(r) dV} \quad \text{where } \rho_{ch}(r) \text{ is the nuclear charge density distribution.}$$



Extensive studies of **stable nuclei** with electron scattering experiments revealed the charge density to be nearly constant in the nuclear volume.

The trend of the mean-square charge radii has the form:

$$\langle r^2 \rangle = \frac{3}{5} (r_0 A^{\frac{1}{3}})^2 \quad \text{Thus the rms charge radius } R = \sqrt{\langle r^2 \rangle} \text{ was seen to scale with } A^{\frac{1}{3}}$$

Great! So we can all go home.....however....

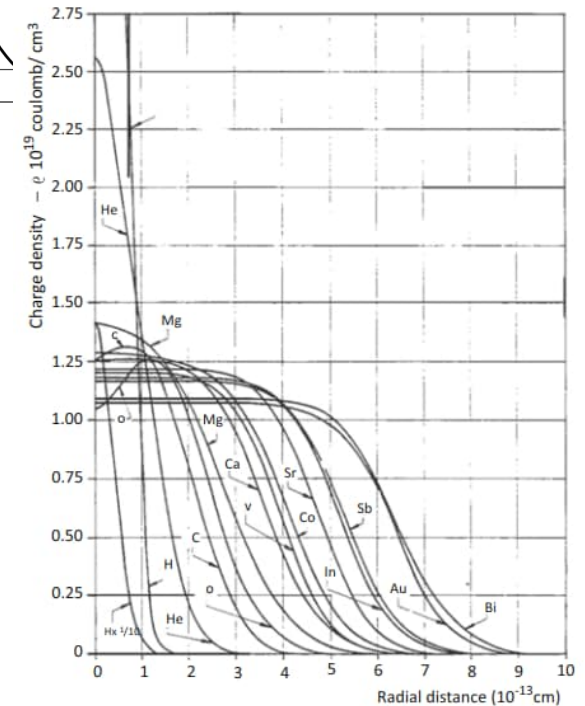
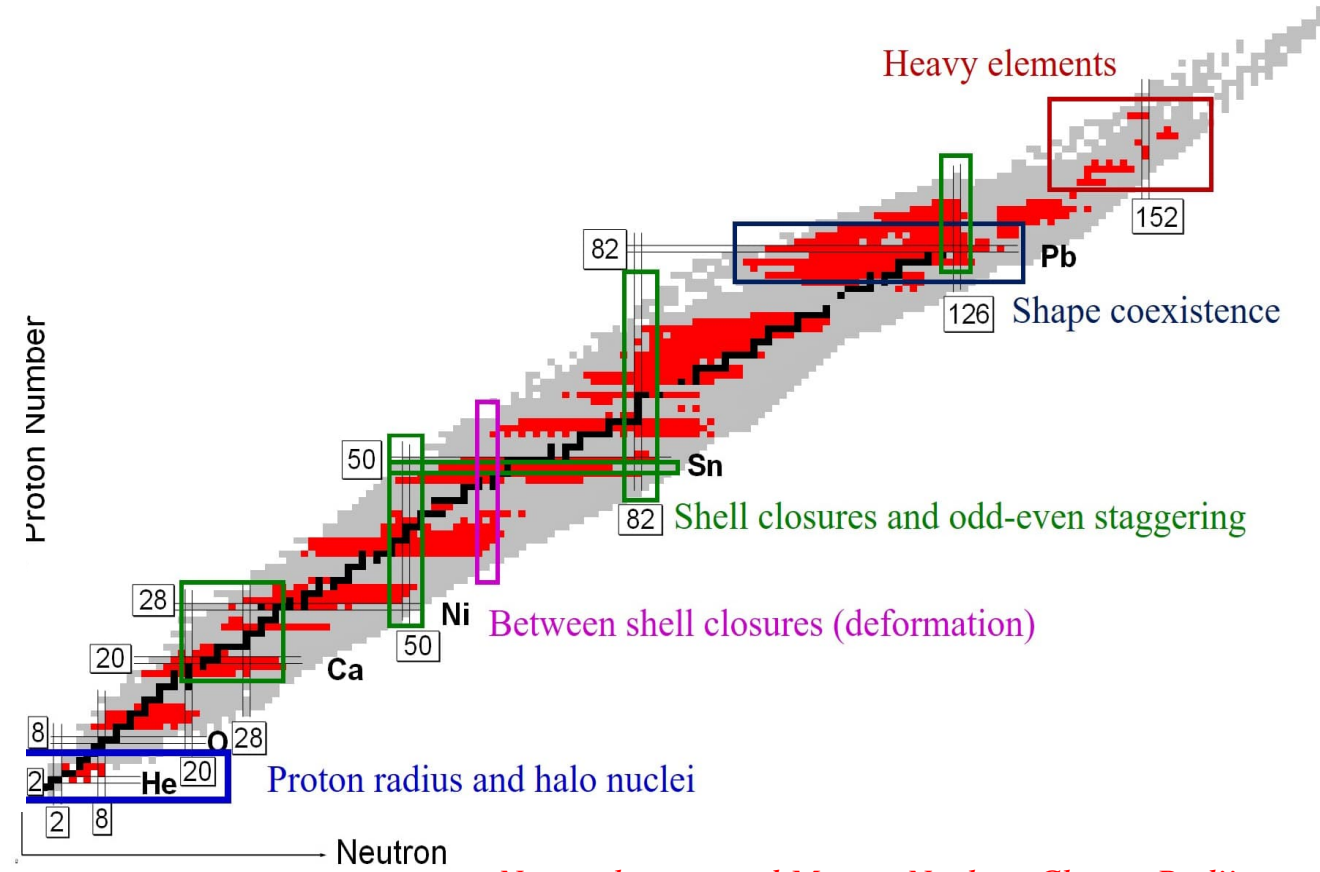
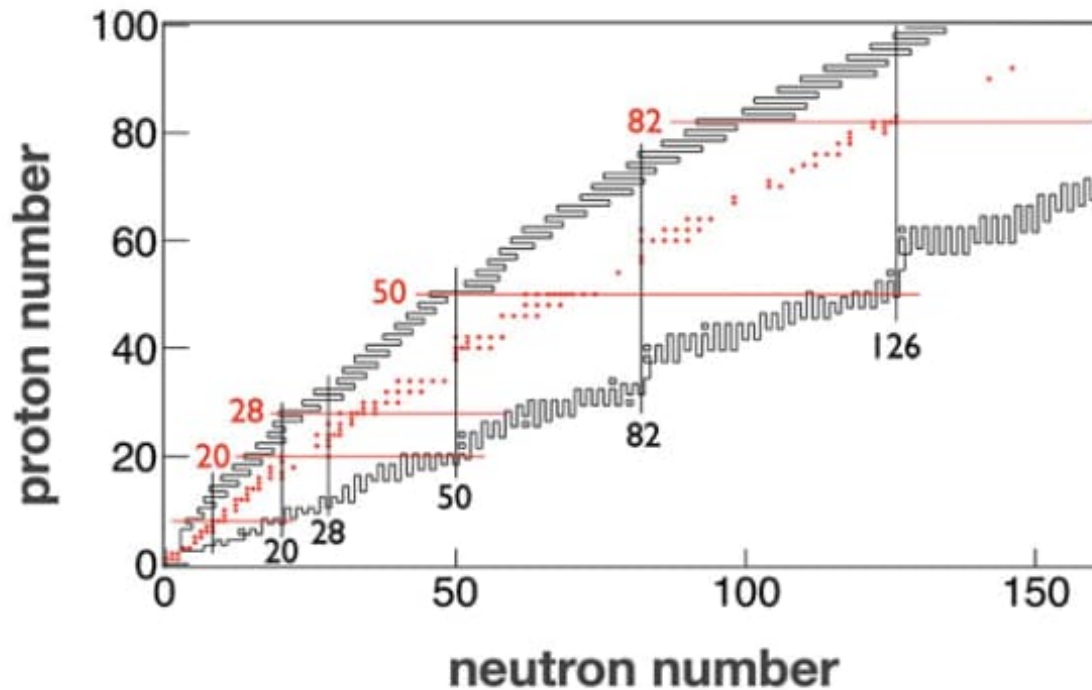


Figure from Hofstadter Nobel Prize lecture, 1961.

Electron scattering vs optical spectroscopy

Unfortunately, scattering experiments (generally) cannot be applied to exotic radioactive nuclei. Optical spectroscopy however provides us with the required sensitivity to probe changes in mean-square charge radii across long chains of isotopes far from stability.



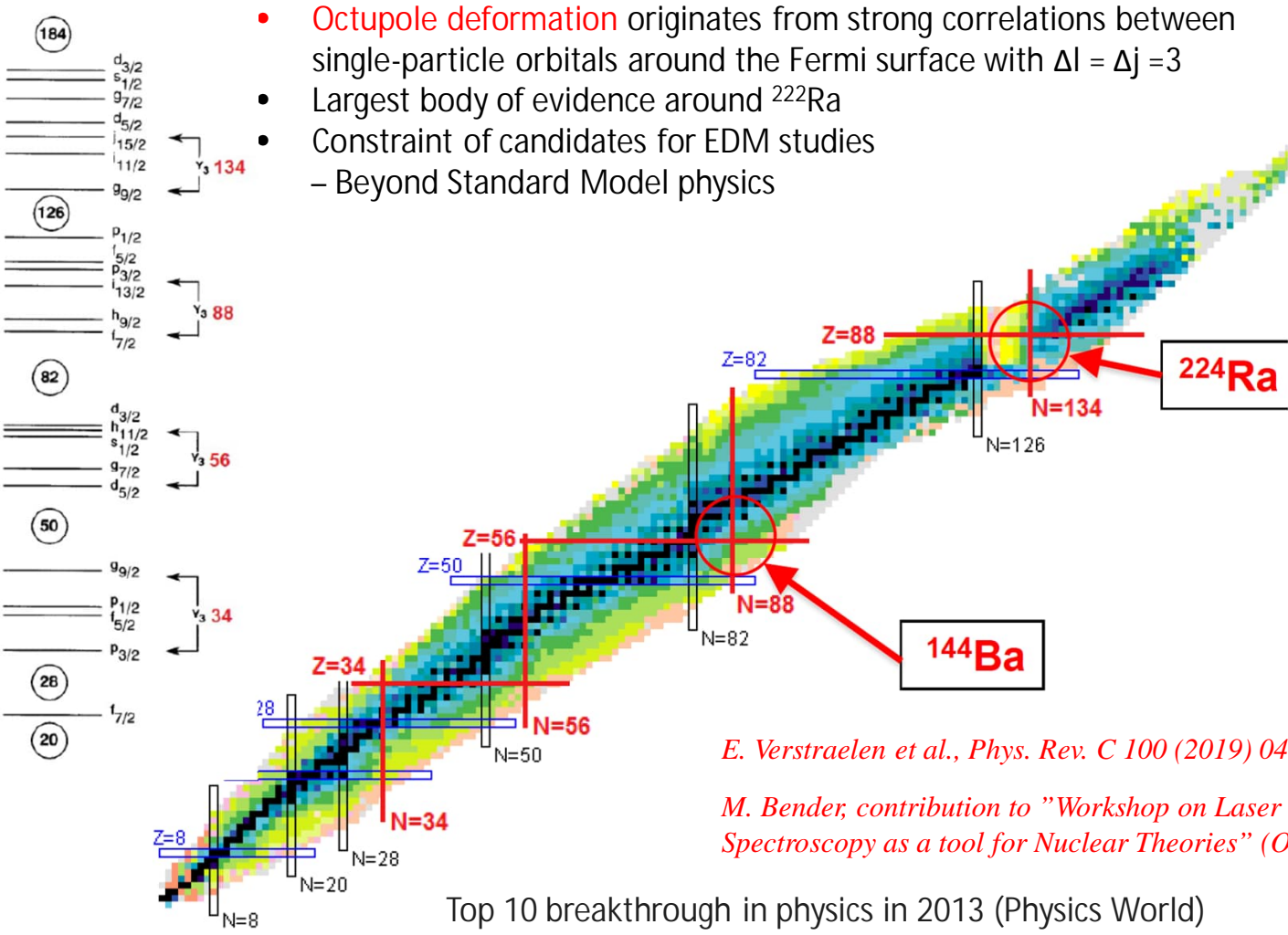
T. Suda, Electron scattering off stable and unstable nuclei, Handbook of Nuclear Physics (2023)

Nörtershäuser and Moore, Nuclear Charge Radii, Handbook of Nuclear Physics (2023)

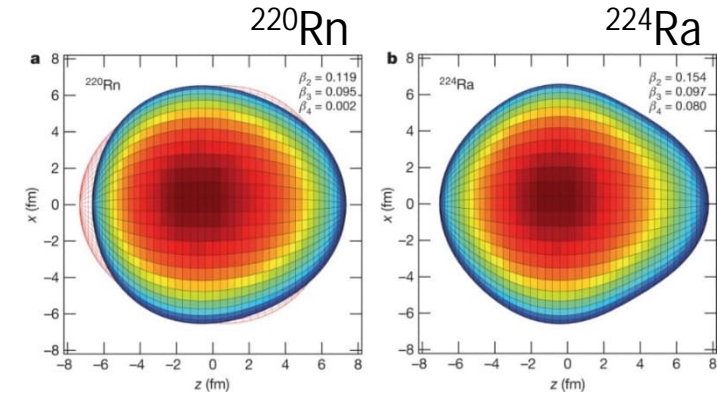
New territory - exotic forms of deformation



UNIVERSITY

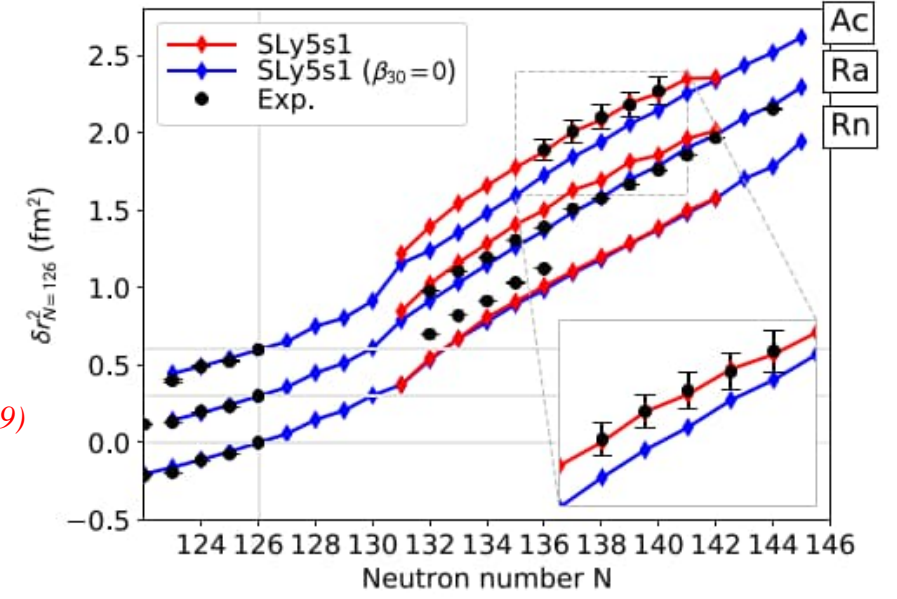


- **Octupole deformation** originates from strong correlations between single-particle orbitals around the Fermi surface with $\Delta l = \Delta j = 3$
- Largest body of evidence around ^{222}Ra
- Constraint of candidates for EDM studies – Beyond Standard Model physics



L.P. Gaffney et al. Nature 497 (2013) 199

Charge radii sensitivity?



E. Verstraelen et al., Phys. Rev. C 100 (2019) 044321

M. Bender, contribution to "Workshop on Laser Spectroscopy as a tool for Nuclear Theories" (Oct. 2019)

Top 10 breakthrough in physics in 2013 (Physics World)

"Pear-shaped nuclei discovery challenges time travel hopes"