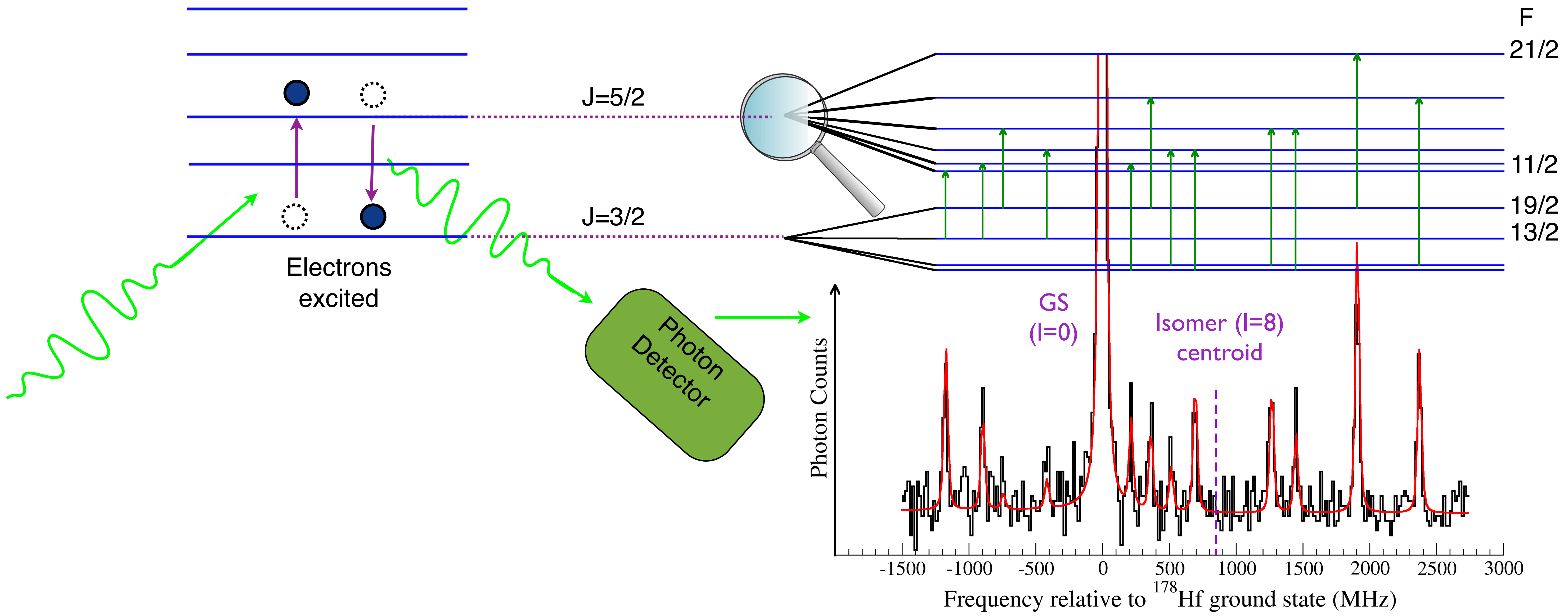
# Measuring nuclear moments using laser spectroscopy

Bradley Cheal
University of Liverpool, UK

# Nuclear properties from laser spectroscopy




Electronic *hyperfine* structure

- Isotope shifts
  - ➔ Charge radius
- Hyperfine splitting
  - ➔ Nuclear spin (measurement)
  - ➔ Magnetic dipole moment
  - ➔ Electric quadrupole moment

➔ Sensitive probe of nuclear wave functions

➔ Single particle level migration

➔ Collectivity and deformation

➔ Existence of a nuclear state at all

Electric dipole selection rule: $\Delta F = 0, \pm 1$ (but not $0 \rightarrow 0$)

Get $\mu, Q_s, \delta\langle r^2\rangle$ from $A, B, \delta\nu$ respectively.

Generally ground and isomeric states, from milliseconds to stability, measured at Radioactive Ion Beam Facilities

# Nuclear properties from optical spectra

Hyperfine energy splittings given by:

$$\Delta E/h = \underbrace{\boxed{\frac{K}{2}}}_{\alpha} A + \underbrace{\boxed{\frac{3K(K+1) - 4I(I+1)J(J+1)}{8I(2I-1)J(2J-1)}}}_{\beta} B$$

where

$$K = F(F+1) - I(I+1) - J(J+1)$$

meaning the hyperfine peak positions (frequency) are given by:

$$\gamma = \nu + \alpha_u A_u + \beta_u B_u - \alpha_l A_l - \beta_l B_l$$

centroid

from which the hyperfine coefficients can be deduced:

$$A = \frac{\mu_I B_e(0)}{IJ}$$

$$B = eQ_s \left\langle \frac{\partial^2 V_e}{\partial z^2} \right\rangle$$

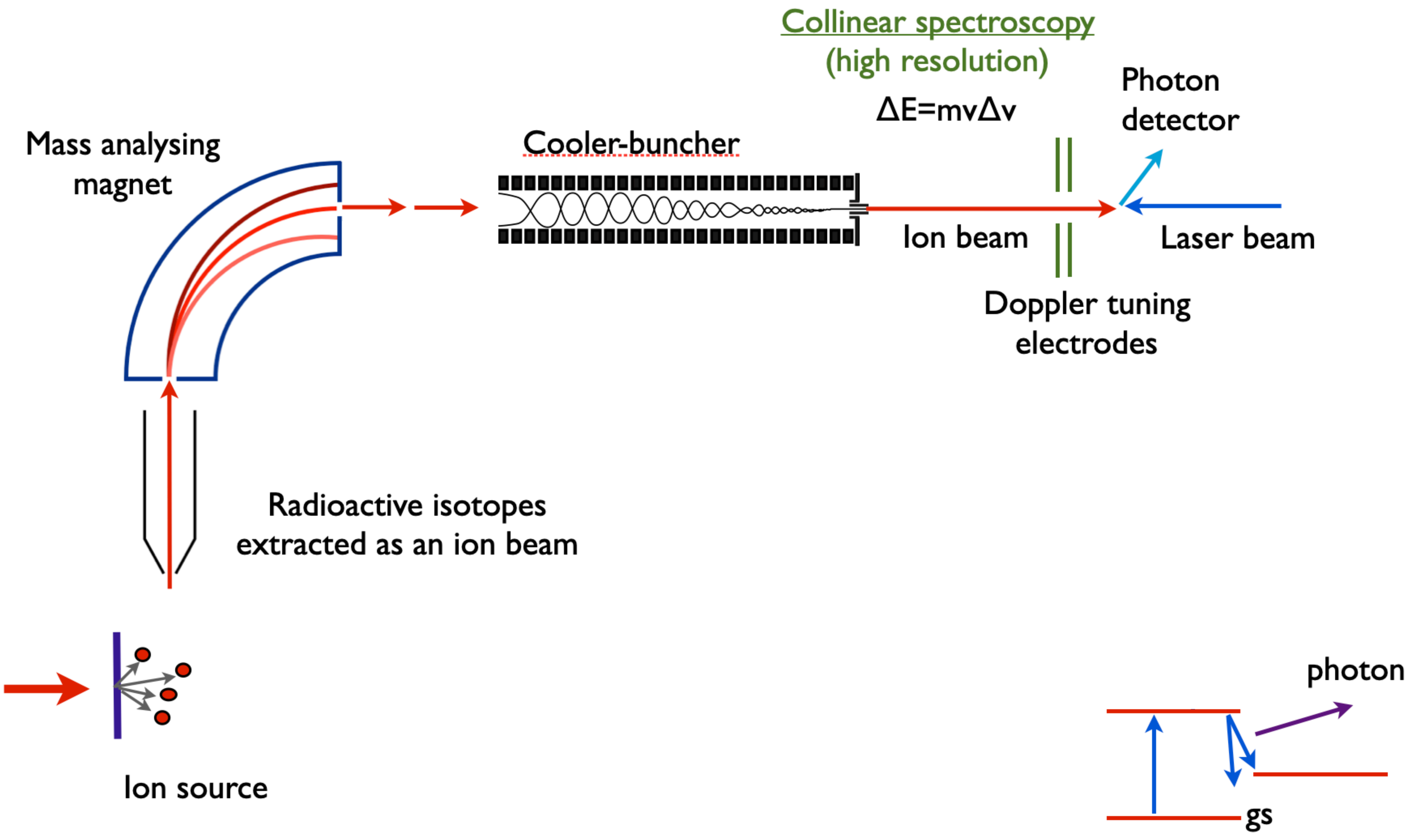
This assumes:

- No hyperfine mixing (eg. seen in Ta due to levels 9 cm$^{-1}$ and 20 cm$^{-1}$ away)
- No hyperfine anomaly (ratios $A_u/A_l$ and $B_u/B_l$ sometimes assumed constant)

Typically, reference values for moments are used to calibrate atomic constants

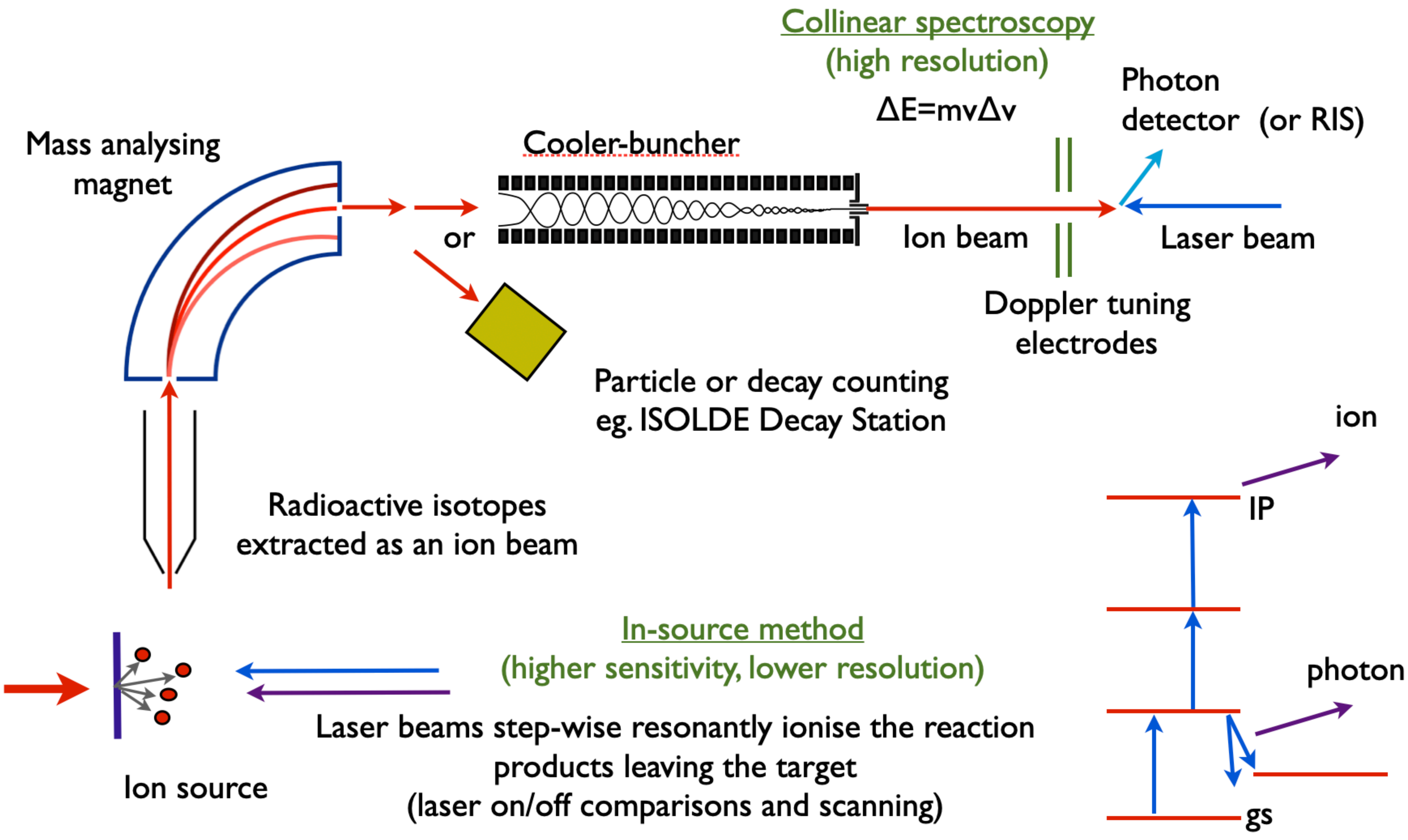
The correct extraction of nuclear properties requires the correct nuclear spin value!

# Laser spectroscopy at RIB facilities




Cheal, Cocolios and Fritzsche, Phys. Rev. A **86**, 042501

# Laser spectroscopy at RIB facilities



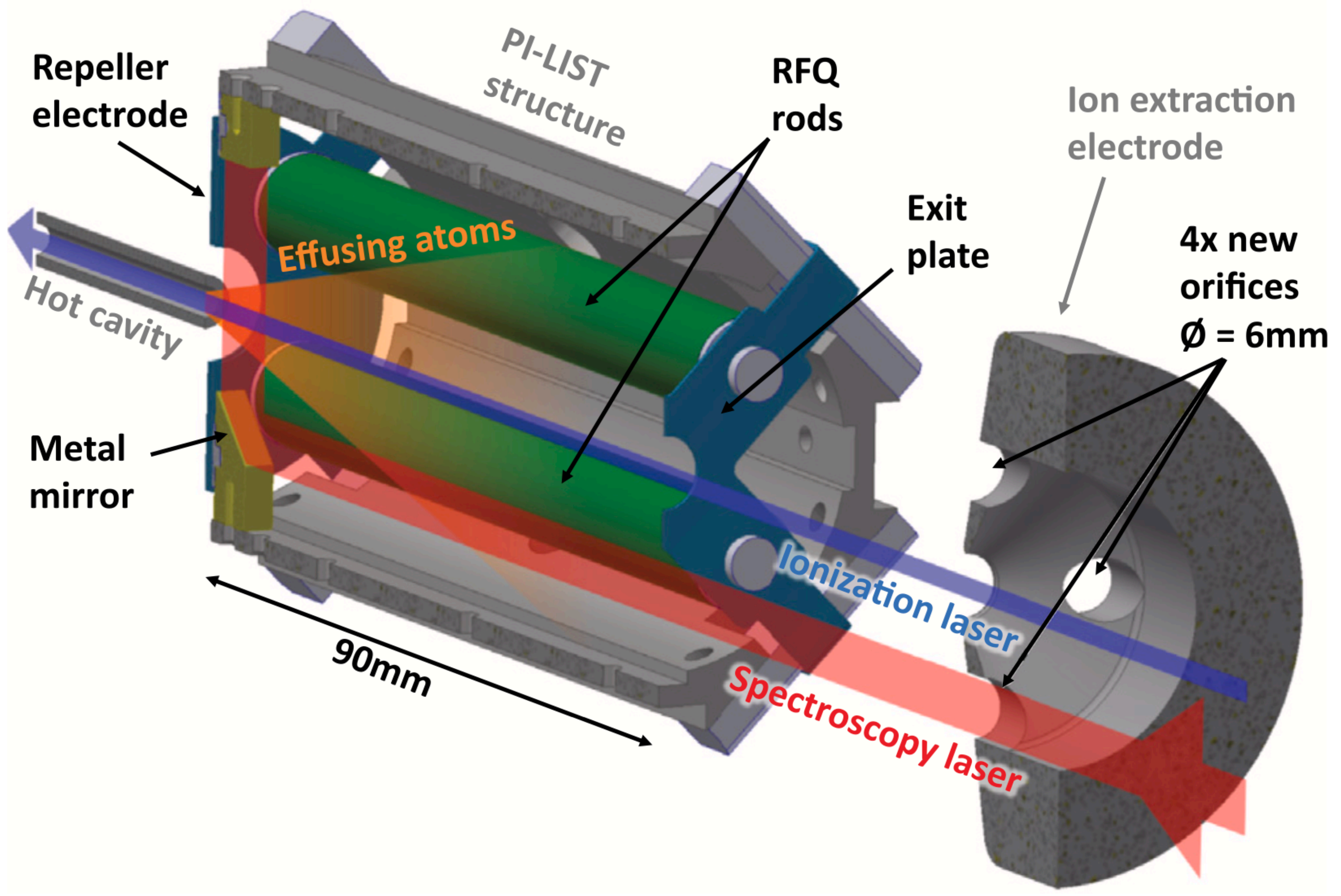

Cheal, Cocolios and Fritzsche, Phys. Rev. A **86**, 042501

# Variants

## Perpendicularly Illuminated Laser Ion Source Trap (PI-LIST)

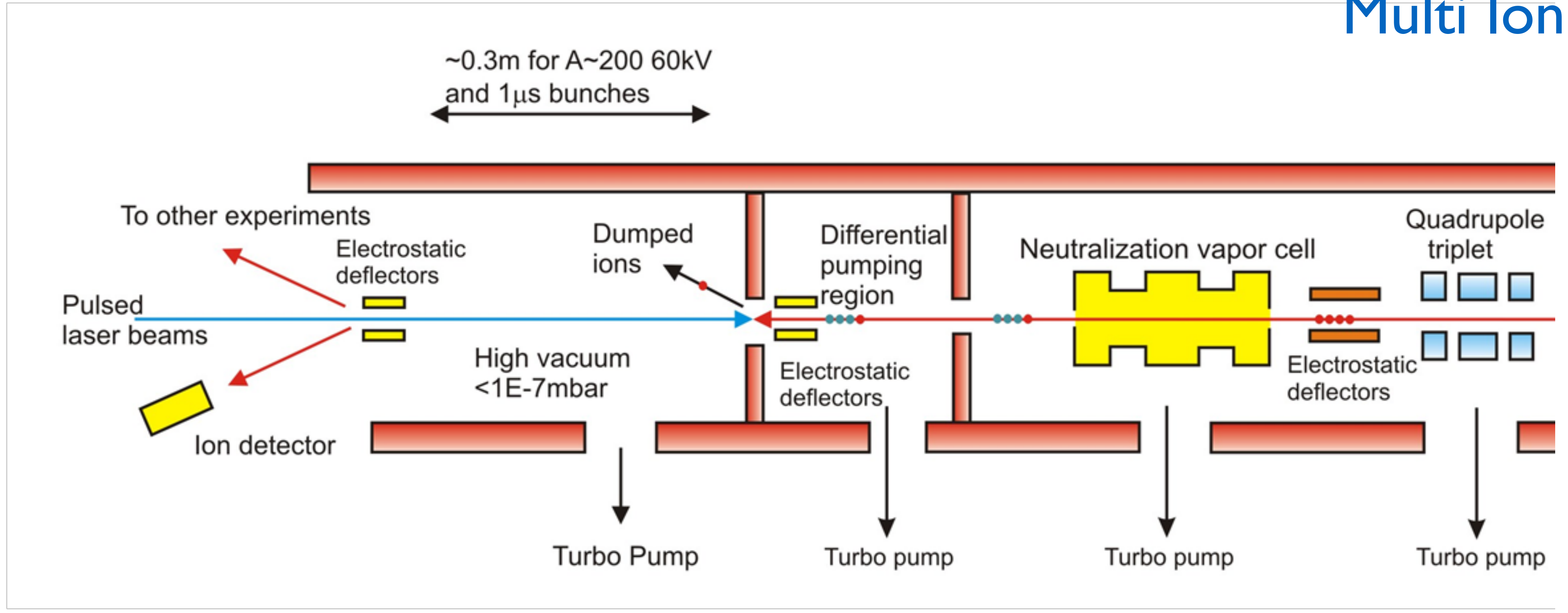
- Development of the insource technique (specifically a further development of LIST)
- Resolution improved to a few 100 MHz

Heinke *et al.*, Nucl. Instrum. Meth. Phys Res. B **541**, 8 (2023)




## Collinear Resonance Ionisation Spectroscopy (CRIS)

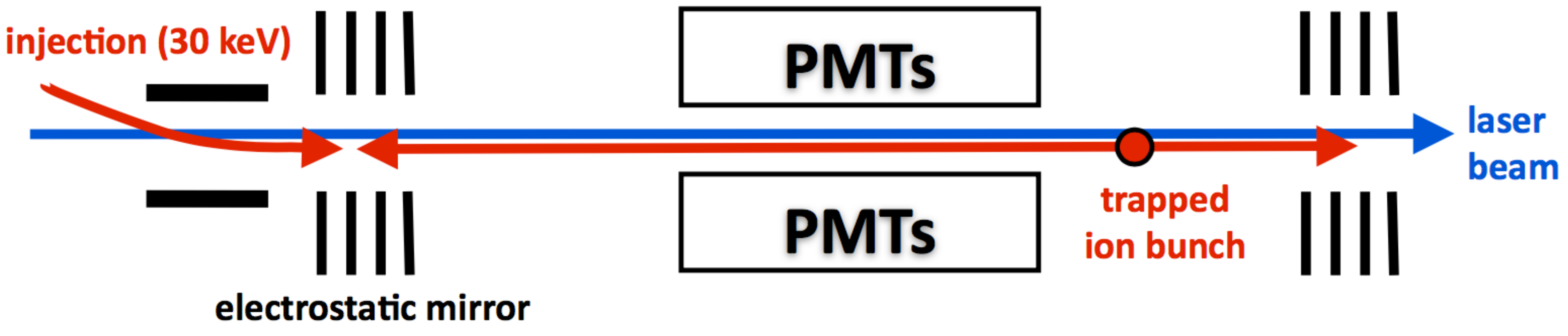
Attempts to combine CLS and particle detection




Vernon *et al.*, Nature **607**, 260 (2022)

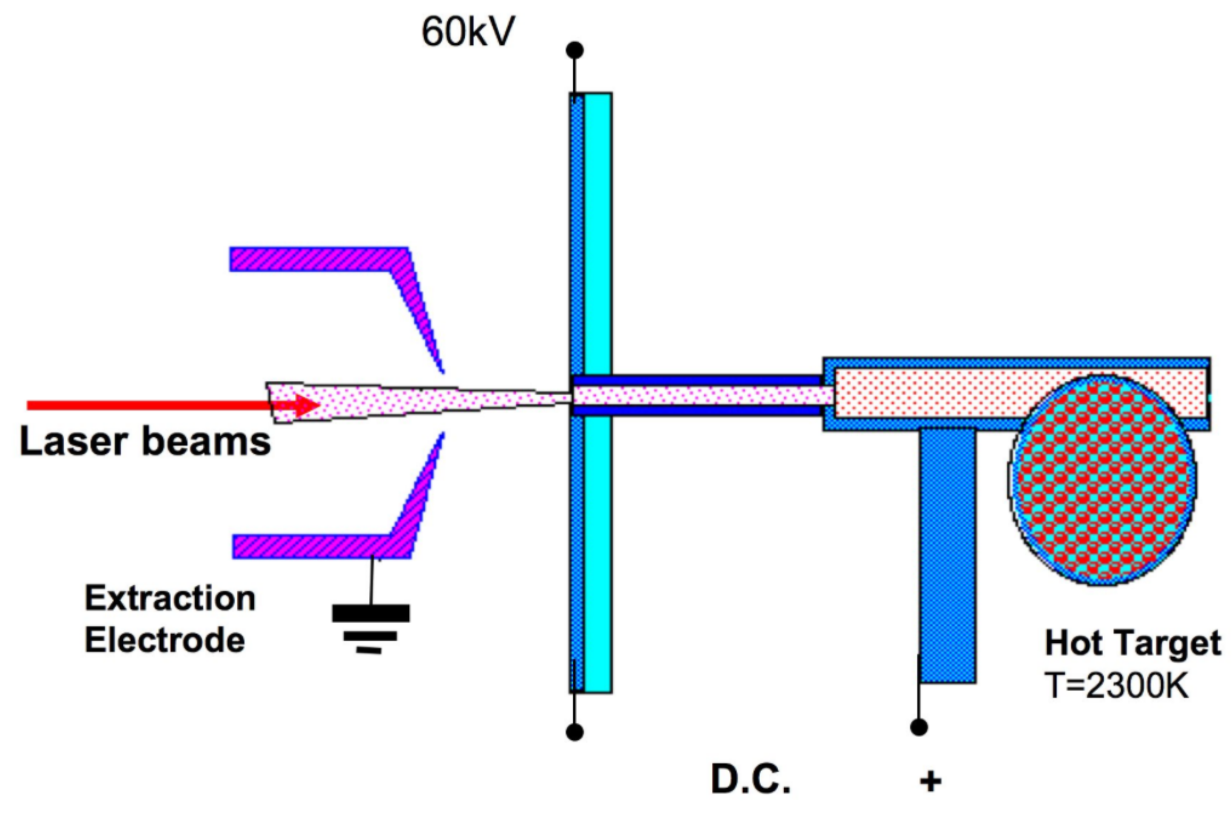
## Multi Ion Reflection Apparatus for Collinear Laser Spectroscopy (MIRACLS)

A development of CLS with multiple passes in a trap



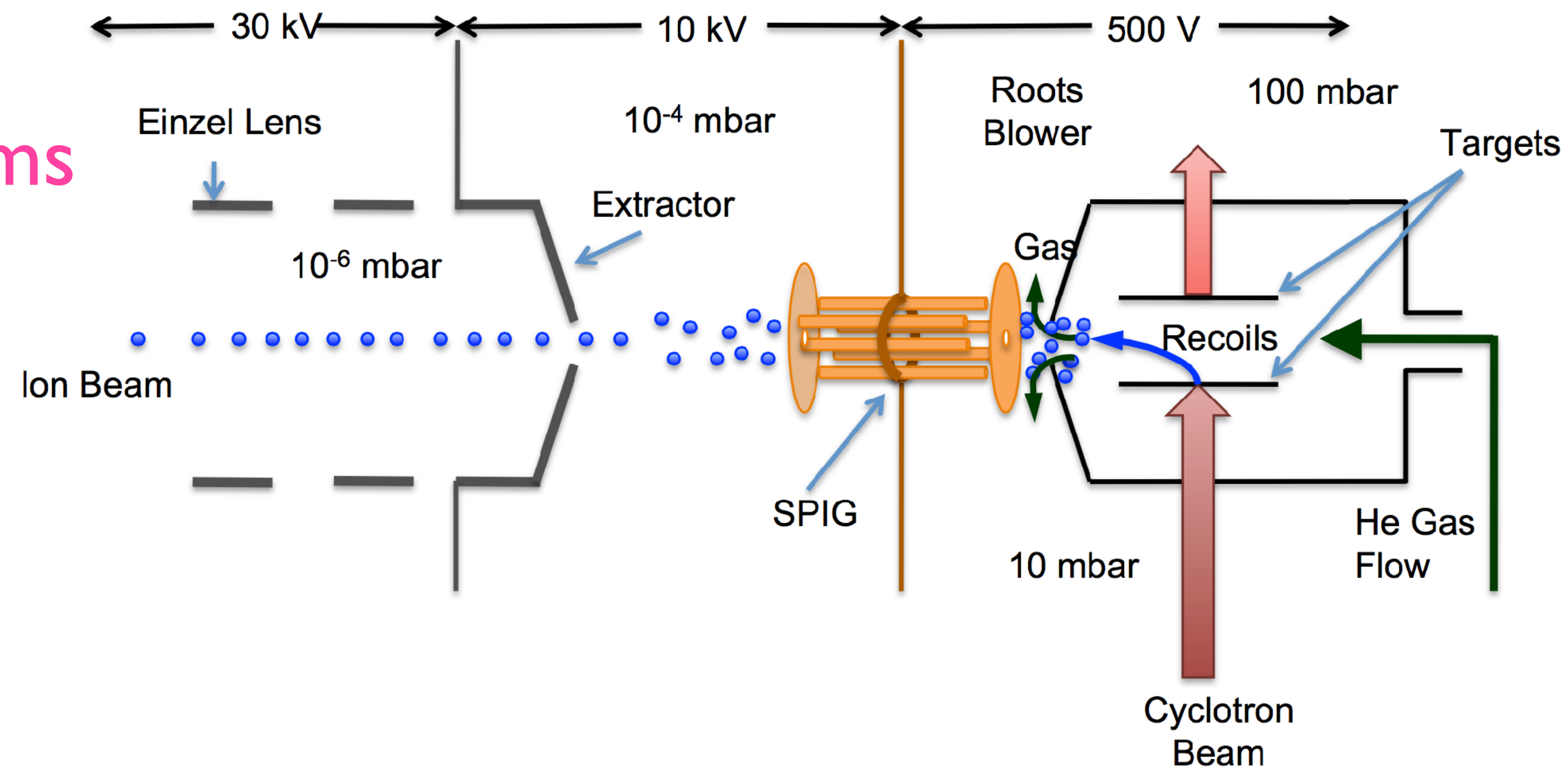

Maier *et al.*, Nucl. Instrum. Meth. Phys Res. A **1075**, 170365 (2025)

# Main Facilities for Laser Spectroscopy




ISOLDE, CERN

- RILIS / LIST / PI-LIST
- COLLAPS
  - Collinear laser fluorescence spectroscopy of ions and atoms
  - State-selective charge exchange followed by radiation detection (ROC)
  - State-selective re-ionisation
- CRIS — Resonance ionisation of atoms to ions in a collinear geometry
- MIRACLS — Collinear laser fluorescence spectroscopy of ions between electrostatic mirrors




JYFL, Finland

- Collinear laser fluorescence spectroscopy of ions and atoms (plus use of optical pumping in the cooler)
- In source spectroscopy for hot cavity work (followed by detection using a Penning trap)

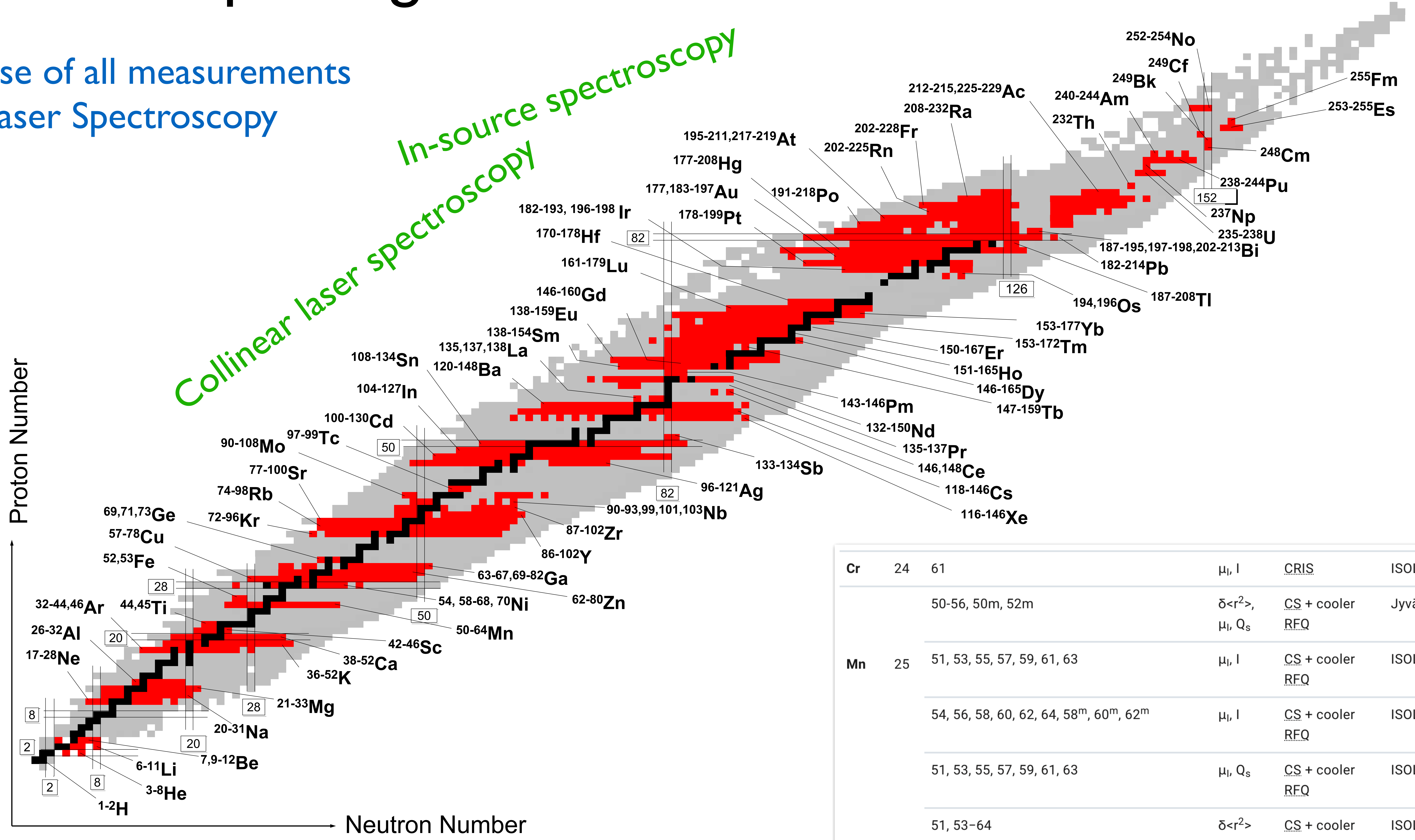
BECOLA, MSU — eg. collinear laser fluorescence spectroscopy

CARIBU, ANL — eg. collinear laser fluorescence spectroscopy

… and others in operation / planned

# Exploring the nuclear chart with lasers

Database of all measurements using Laser Spectroscopy

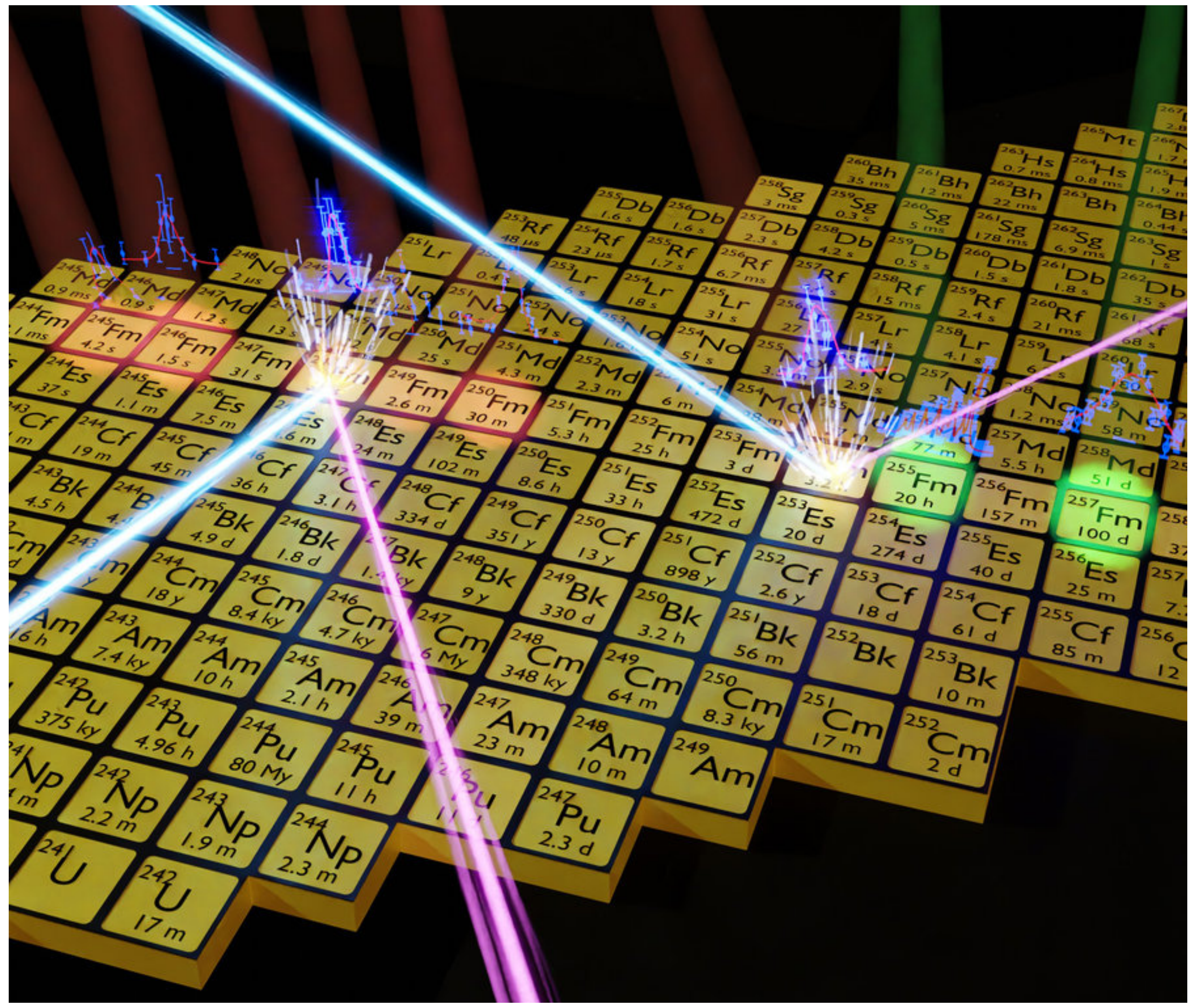



RADRIS & JetRIS

| | | | | | | |
|---|---|---|---|---|---|---|
| Cr | 24 | 61 | $\mu_I$, I | CRIS | ISOLDE | [→ 251] |
| Mn | 25 | 50-56, 50m, 52m | $\delta\langle r^2\rangle$, $\mu_I$, $Q_s$ | CS + cooler RFQ | Jyväskylä | [→ 99] |
| | | 51, 53, 55, 57, 59, 61, 63 | $\mu_I$, I | CS + cooler RFQ | ISOLDE | [→ 148] |
| | | 54, 56, 58, 60, 62, 64, $58^m$, $60^m$, $62^m$ | $\mu_I$, I | CS + cooler RFQ | ISOLDE | [→ 149] |
| | | 51, 53, 55, 57, 59, 61, 63 | $\mu_I$, $Q_s$ | CS + cooler RFQ | ISOLDE | [→ 157] |
| | | 51, 53-64 | $\delta\langle r^2\rangle$ | CS + cooler RFQ + SPOP | ISOLDE | [→ 163] |

TU Darmstadt, Laser Spectroscopy of Radioactive Isotopes - Survey

# RADRIS at GSI, Darmstadt

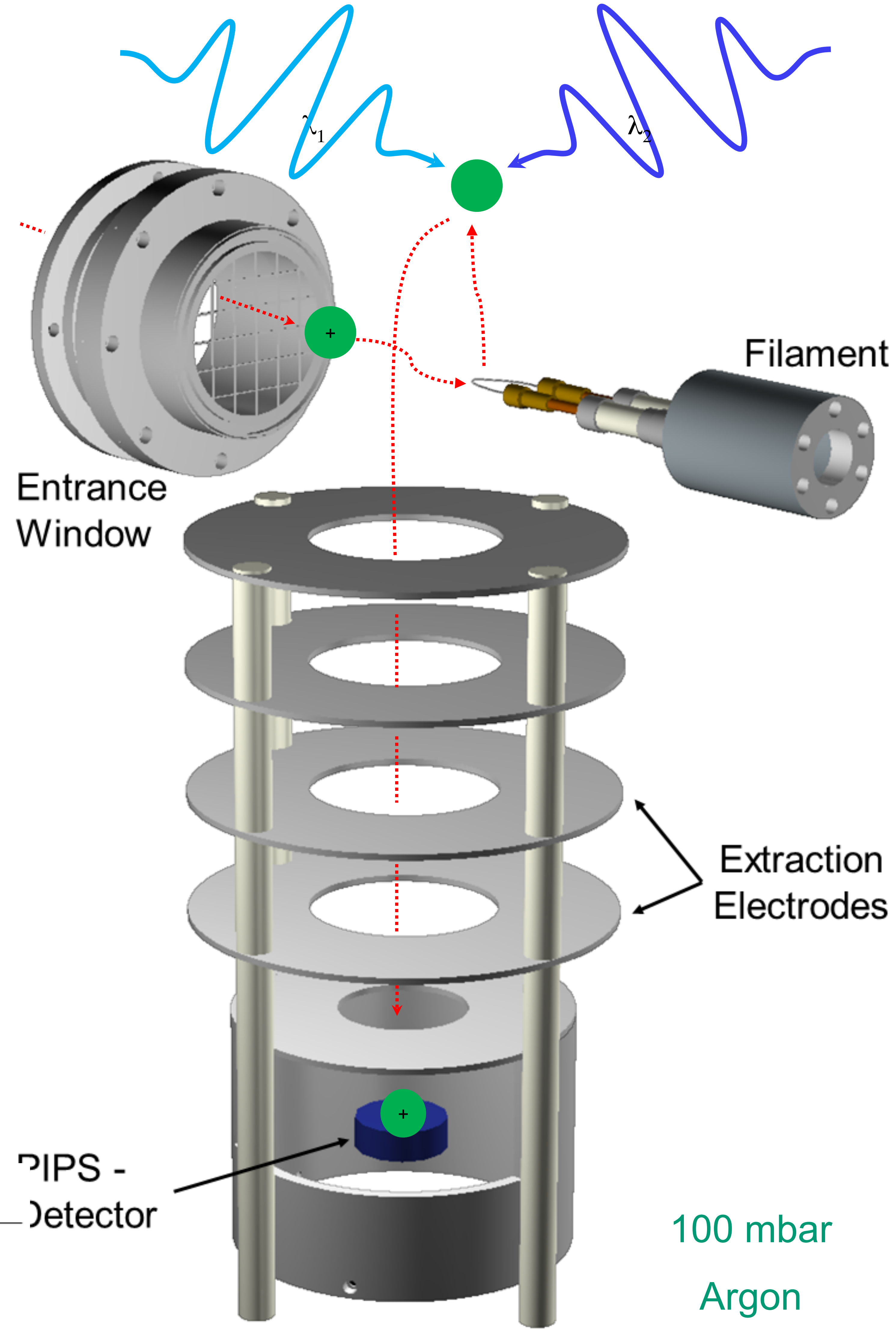


First resonance in nobelium

…leading to moments

Raeder *et al.* Phys. Rev. Lett. **120**, 232503 (2018)

Editors' Suggestion | Featured in Physics




## Atom-at-a-time laser resonance ionization spectroscopy of nobelium

Mustapha Laatiaoui[1,2], Werner Lauth[3], Hartmut Backe[3], Michael Block[1,2,4], Dieter Ackermann[2]†, Bradley Cheal[5], Premaditya Chhetri[6], Christoph Emanuel Düllmann[1,2,4], Piet Van Duppen[7], Julia Even[1]†, Rafael Ferrer[7], Francesca Giacoppo[1,2], Stefan Götz[1,2,4], Fritz Peter Heßberger[1,2], Mark Huyse[7], Oliver Kaleja[2,8], Jadambaa Khuyagbaatar[1,2], Peter Kunz[9], Felix Lautenschläger[6], Andrew Kishor Mistry[1,2], Sebastian Raeder[1,2,7], Enrique Minaya Ramirez[1]†, Thomas Walther[6], Calvin Wraith[5] & Alexander Yakushev[1,2]

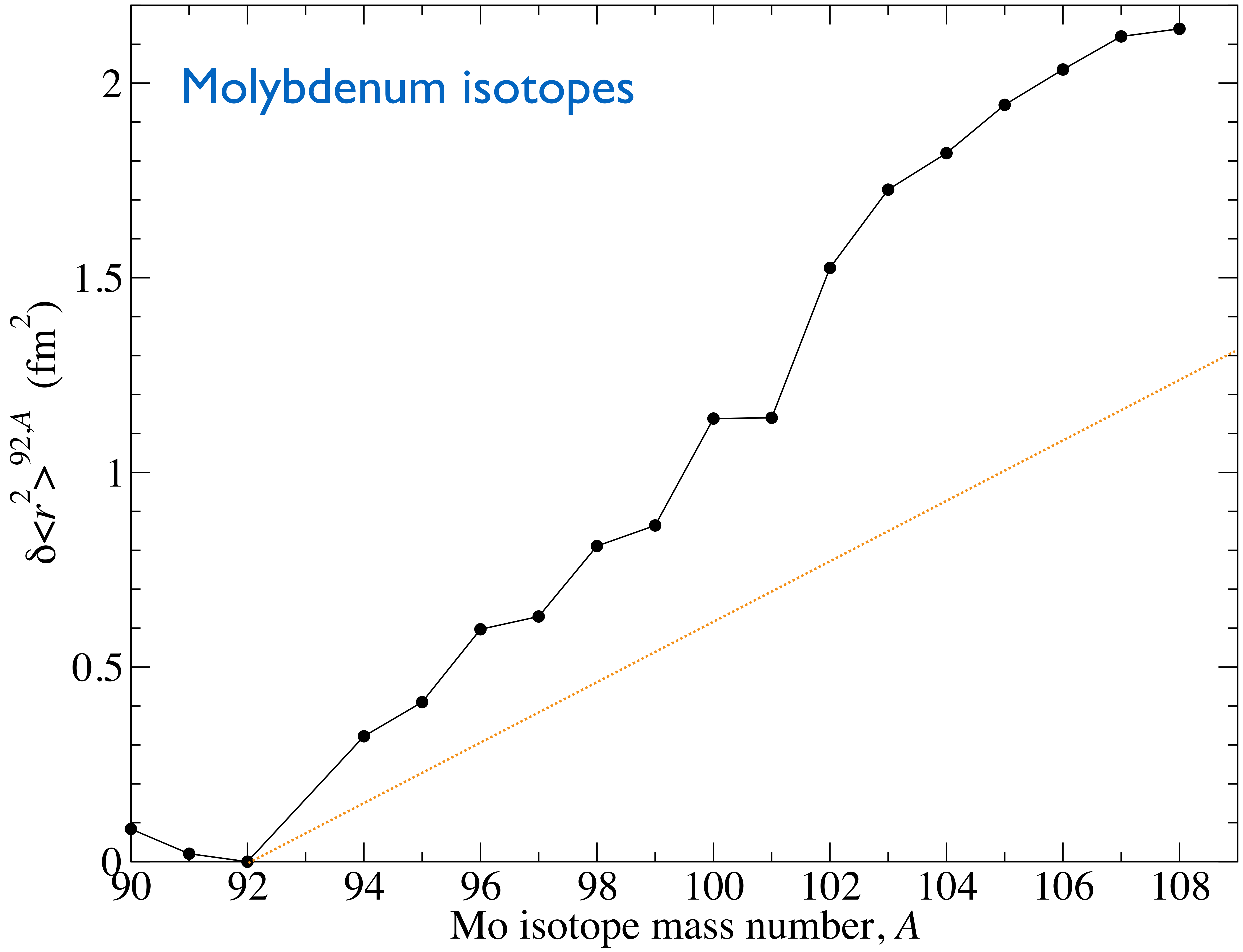
Laatiaoui *et al.*, Nature **538**, 495 (2016)

Warbinek *et al.*, Nature **634**, 1075 (2024)

**Article**

## Smooth trends in fermium charge radii and the impact of shell effects

# Measuring charge-radii




Isotope shifts

$$\delta\nu_i^{A,A'} = M_i \frac{A' - A}{AA'} + F_i \delta\langle r^2\rangle^{A,A'}$$

Constants

Charge radii

B. Cheal, T.E. Cocolios and S. Fritzsche Phys. Rev. A 86 042501 (2012)

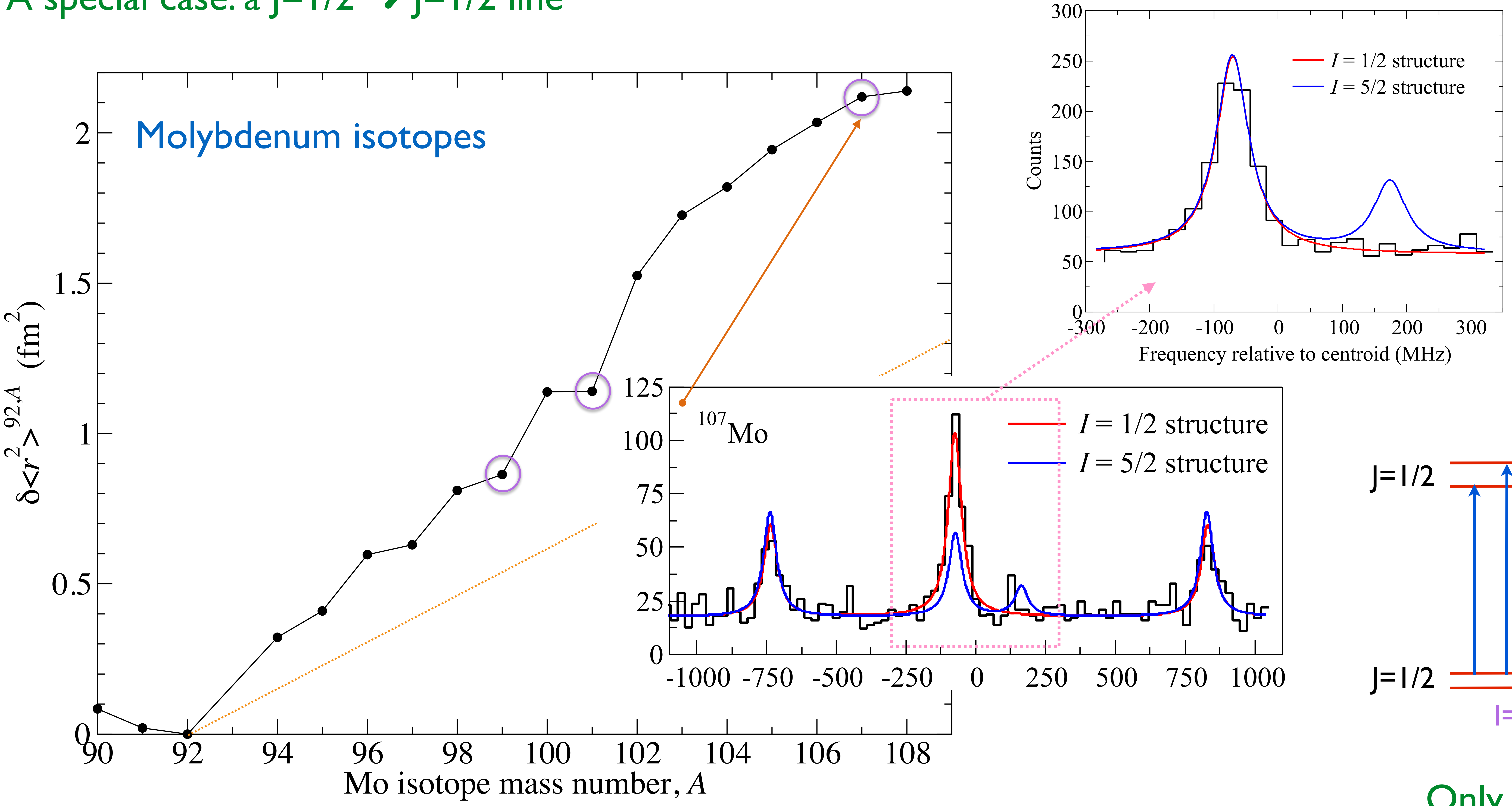
Tables of all data (recalibrated) compiled

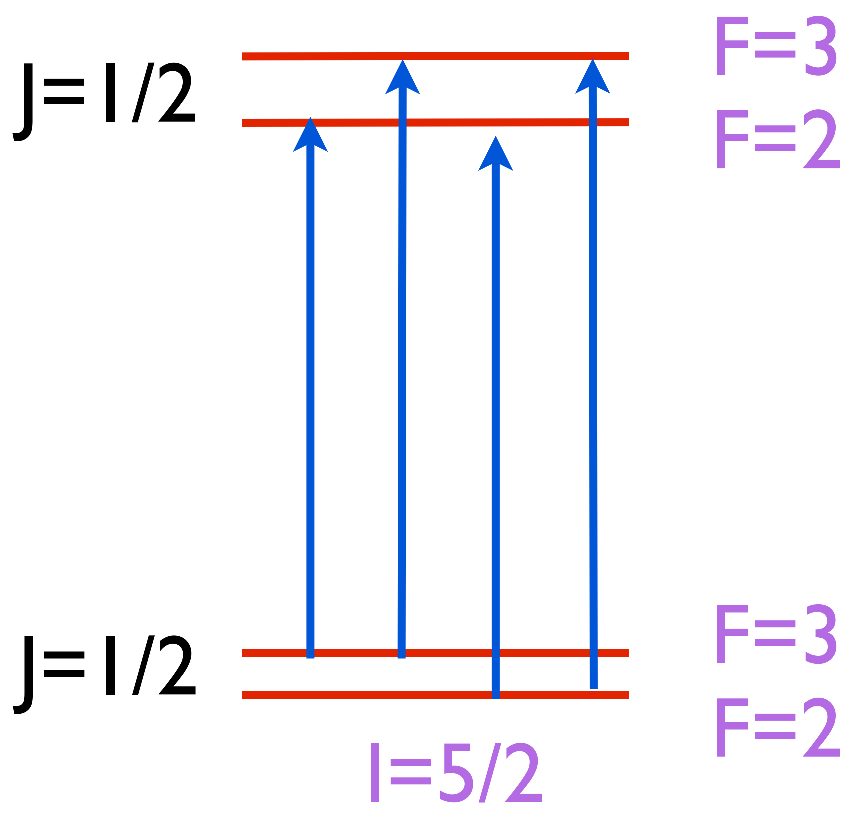
I. Angeli and K. P. Marinova, At. Data Nucl. Data Tables 99, 69 (2013).

G. Fricke and K. Heilig, Nuclear Charge Radii (Landolt-Börnstein/Springer, Berlin, 2004).

# How can we determine the nuclear spin?

A special case: a J=1/2 ➜ J=1/2 line

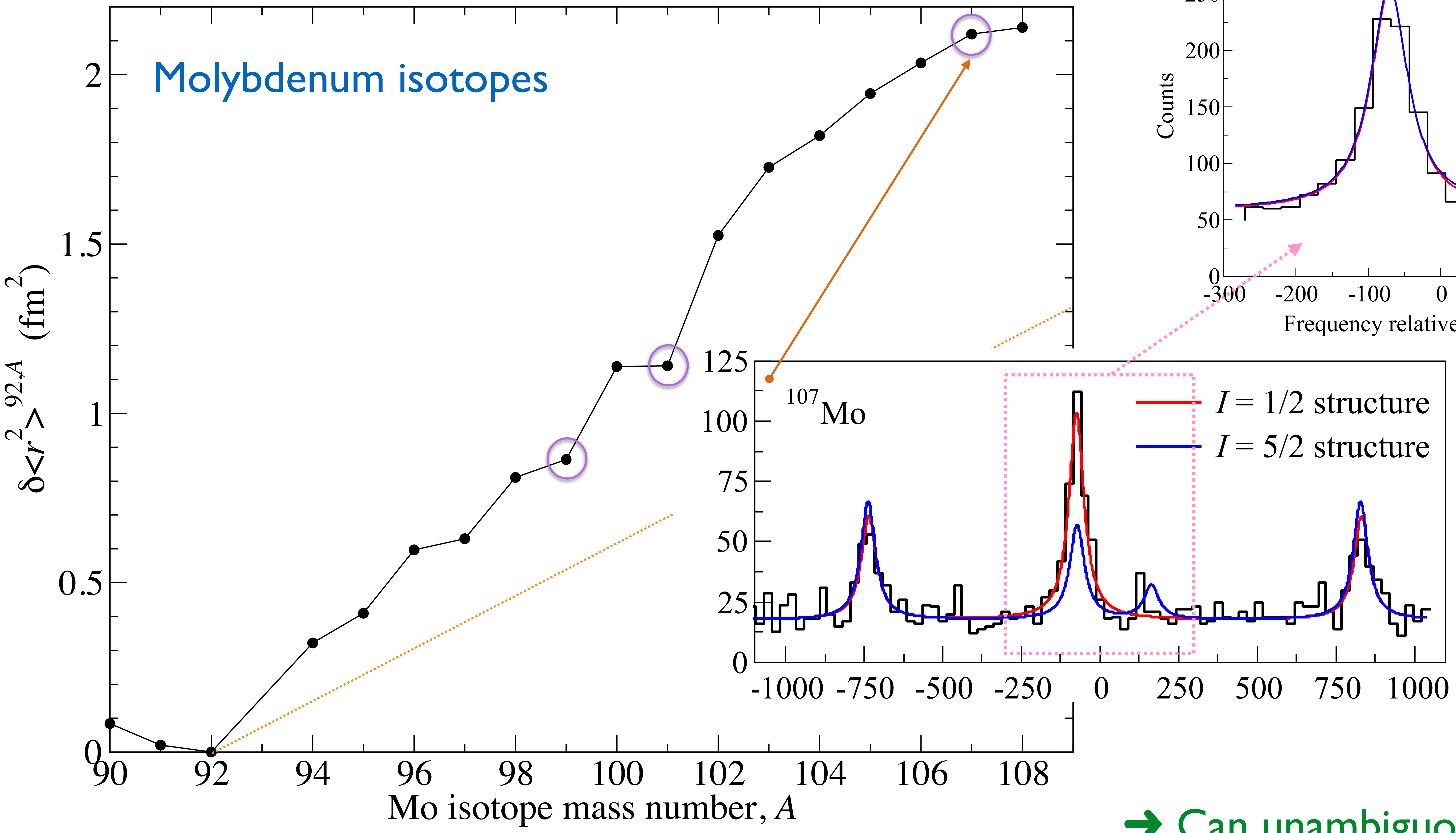


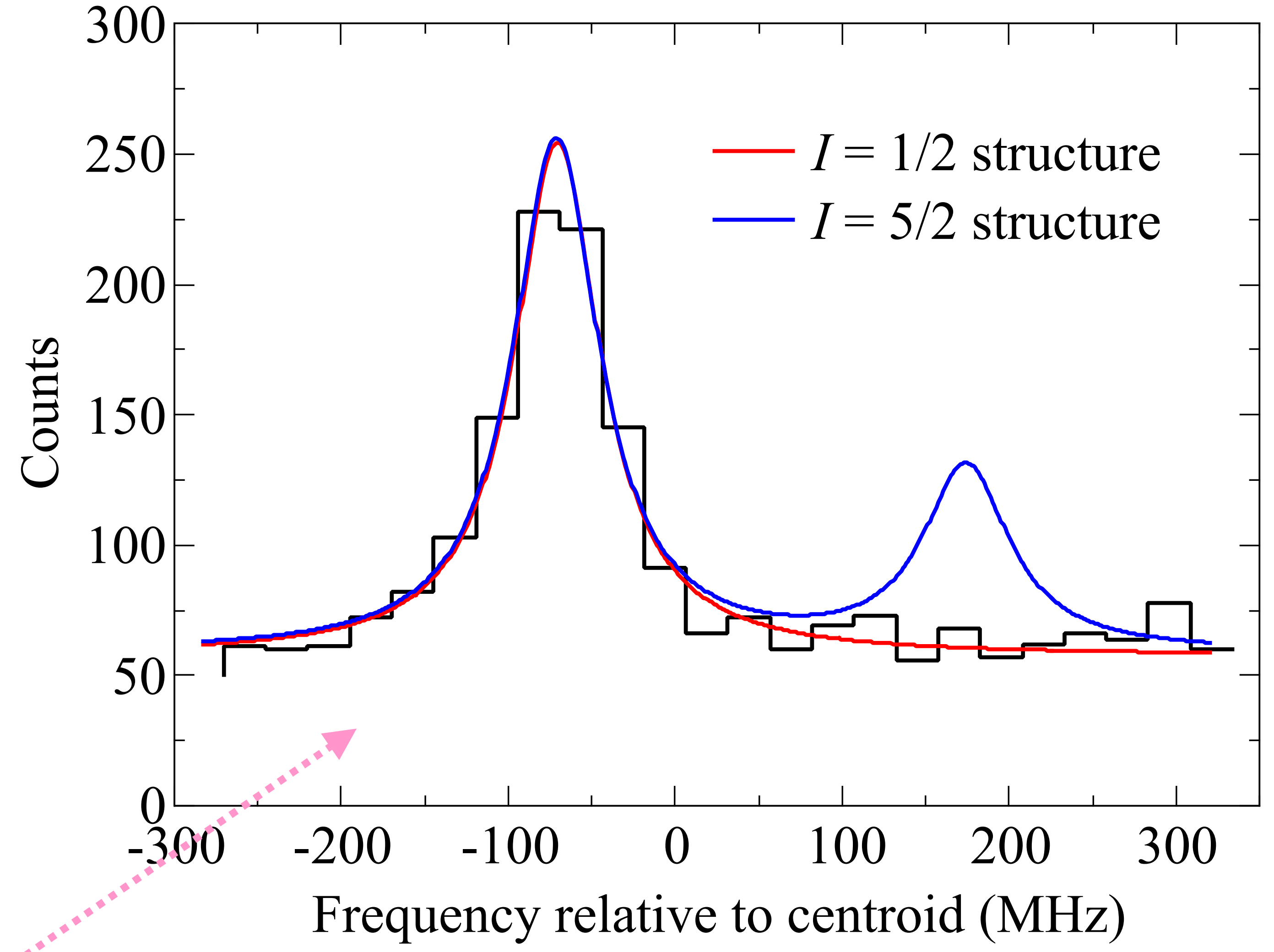



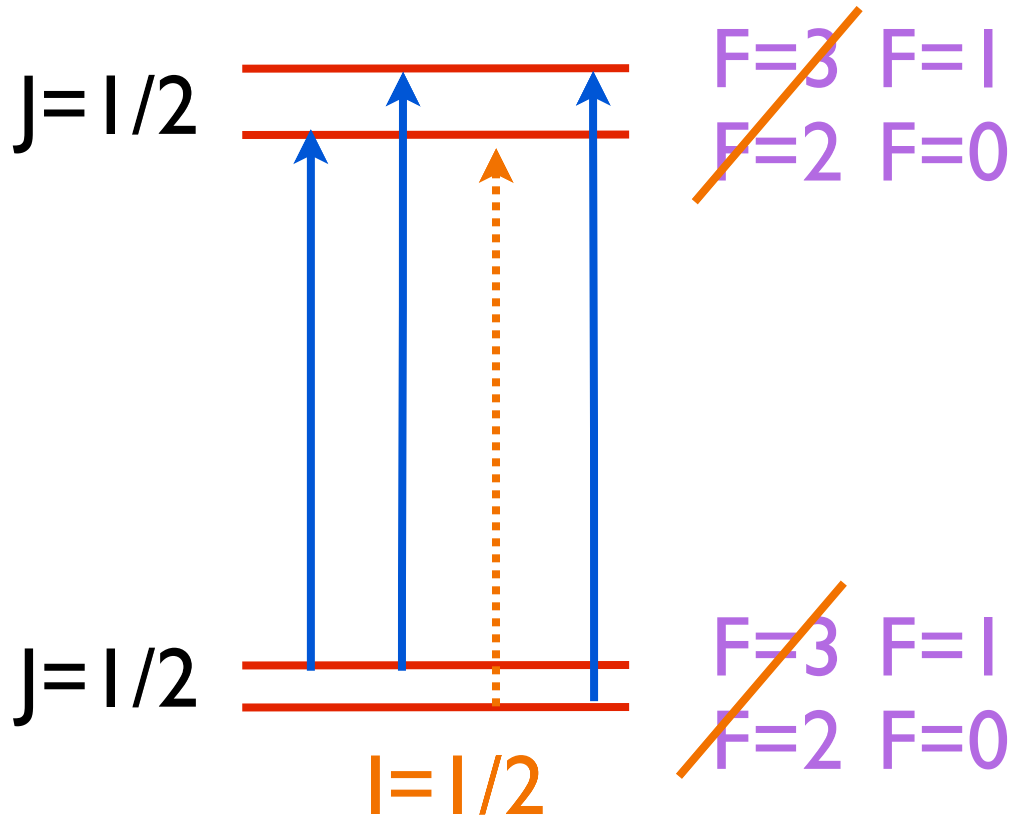

Only 3 peaks!

# How can we determine the nuclear spin?

A special case: a J=1/2 ➜ J=1/2 line



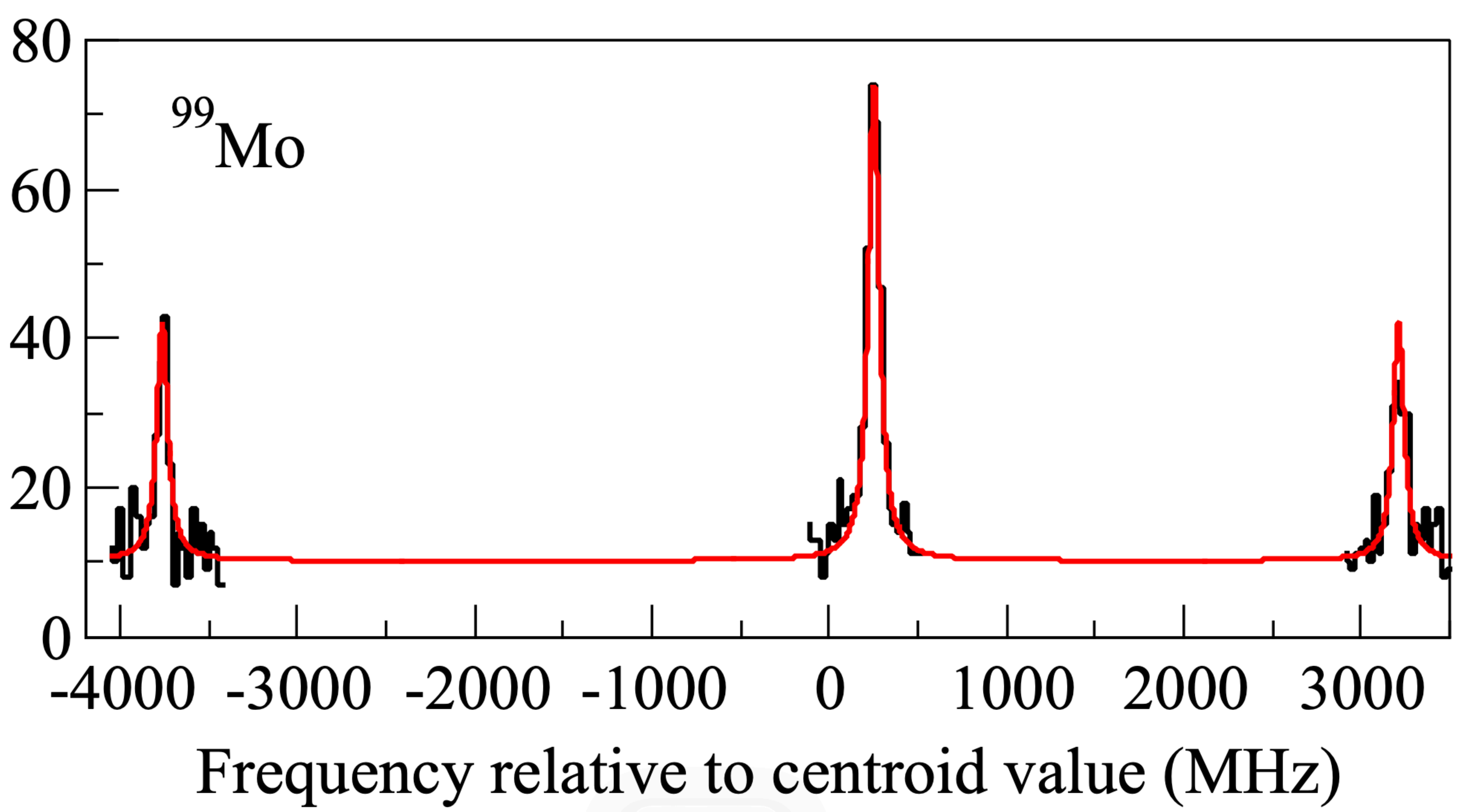



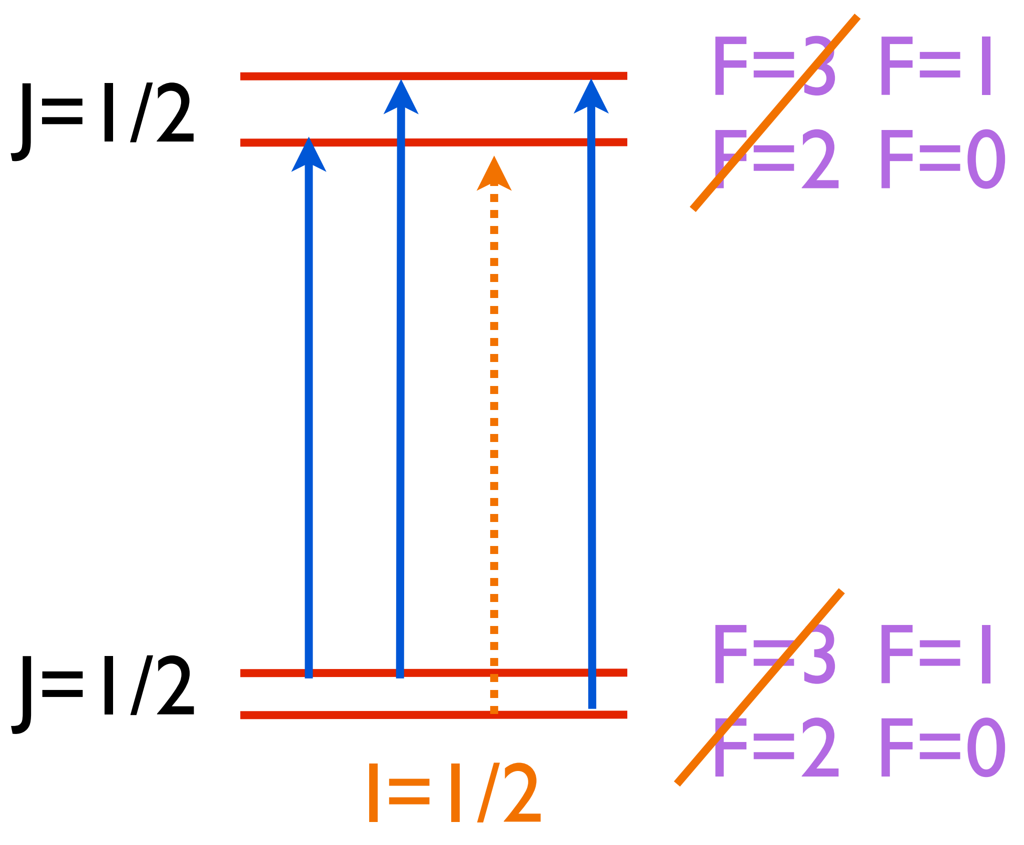




➜ Can unambiguously assign as spin *I* = 1/2.

# Another example: $^{99}$Mo

Again, clear that we can unambiguously assign as spin $I$ = 1/2, in agreement with literature.




But happened to notice the magnetic moment is $\mu = -0.794(6)\,\mu_N$

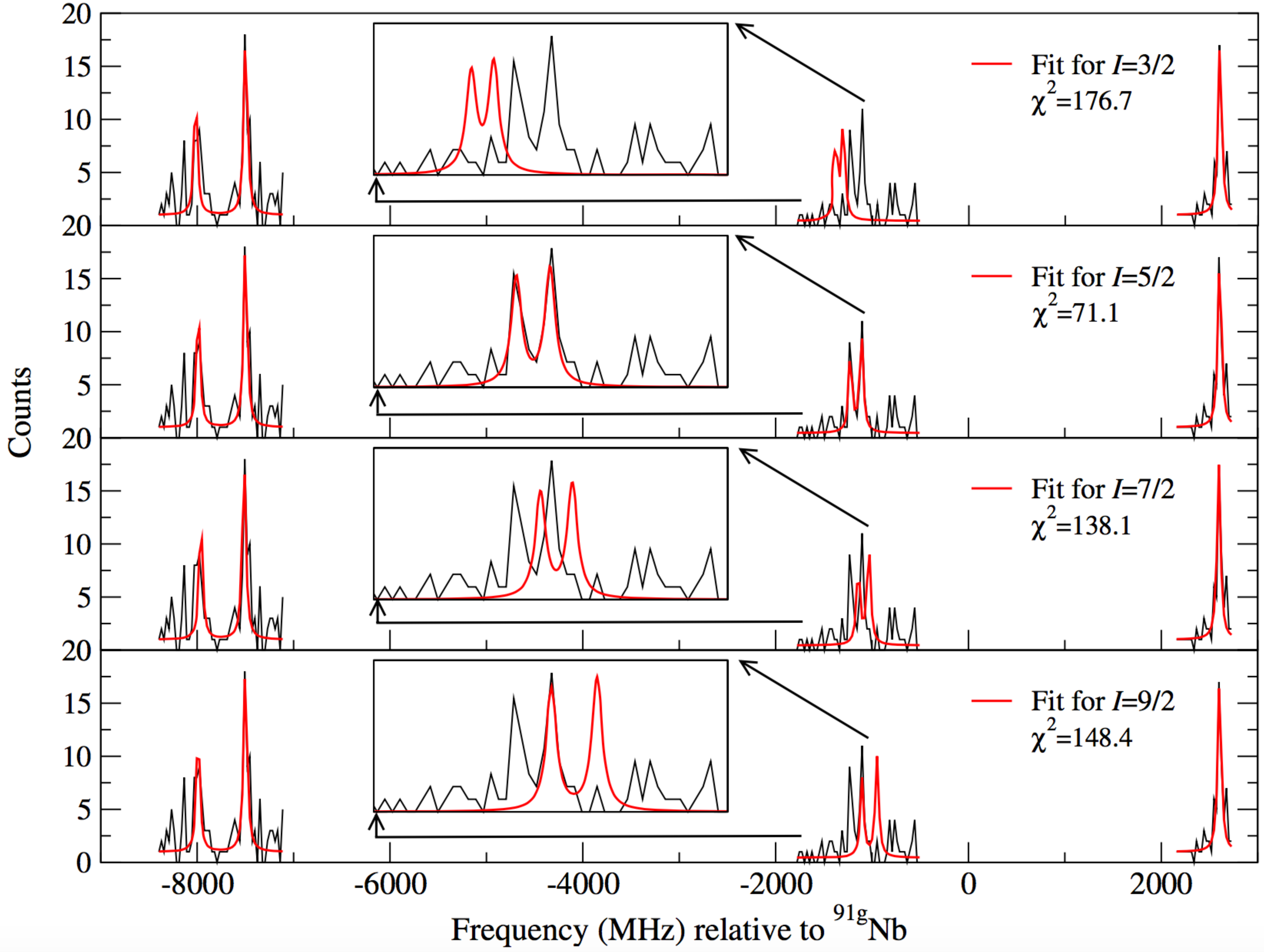
Literature value (ABMR) is $\mu = 0.374(3)\,\mu_N$

(What can be wrong? Can literature values be reanalysed and corrected?)

# Measuring the nuclear spin (general case)

eg.
$^{101}$Nb

J=1 ➜ J=1 line

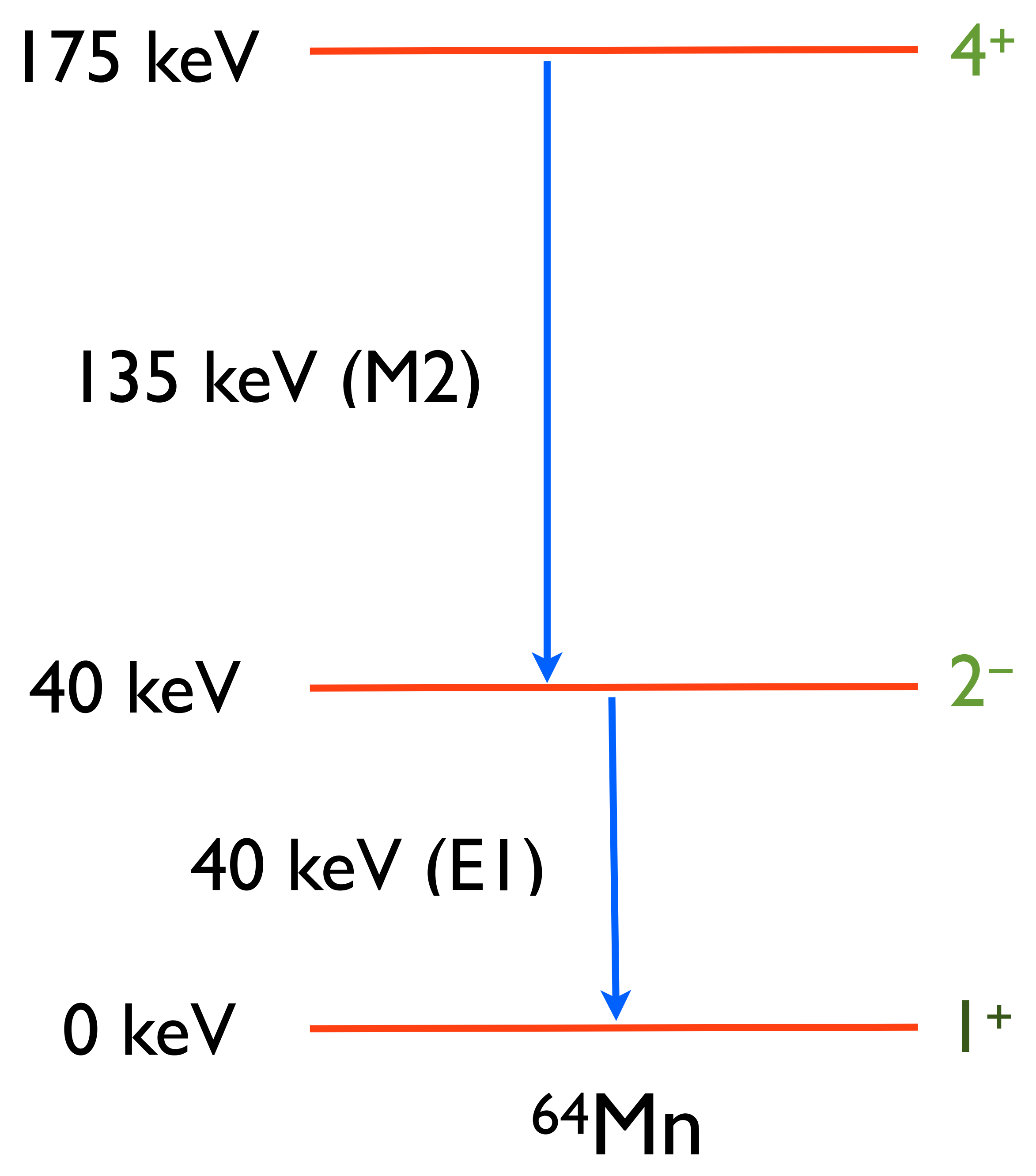



➜ $I$ = 5/2

The frequency pattern of the hyperfine peaks gives the spin unambiguously.

# Measuring the nuclear spin

From gamma spectroscopy

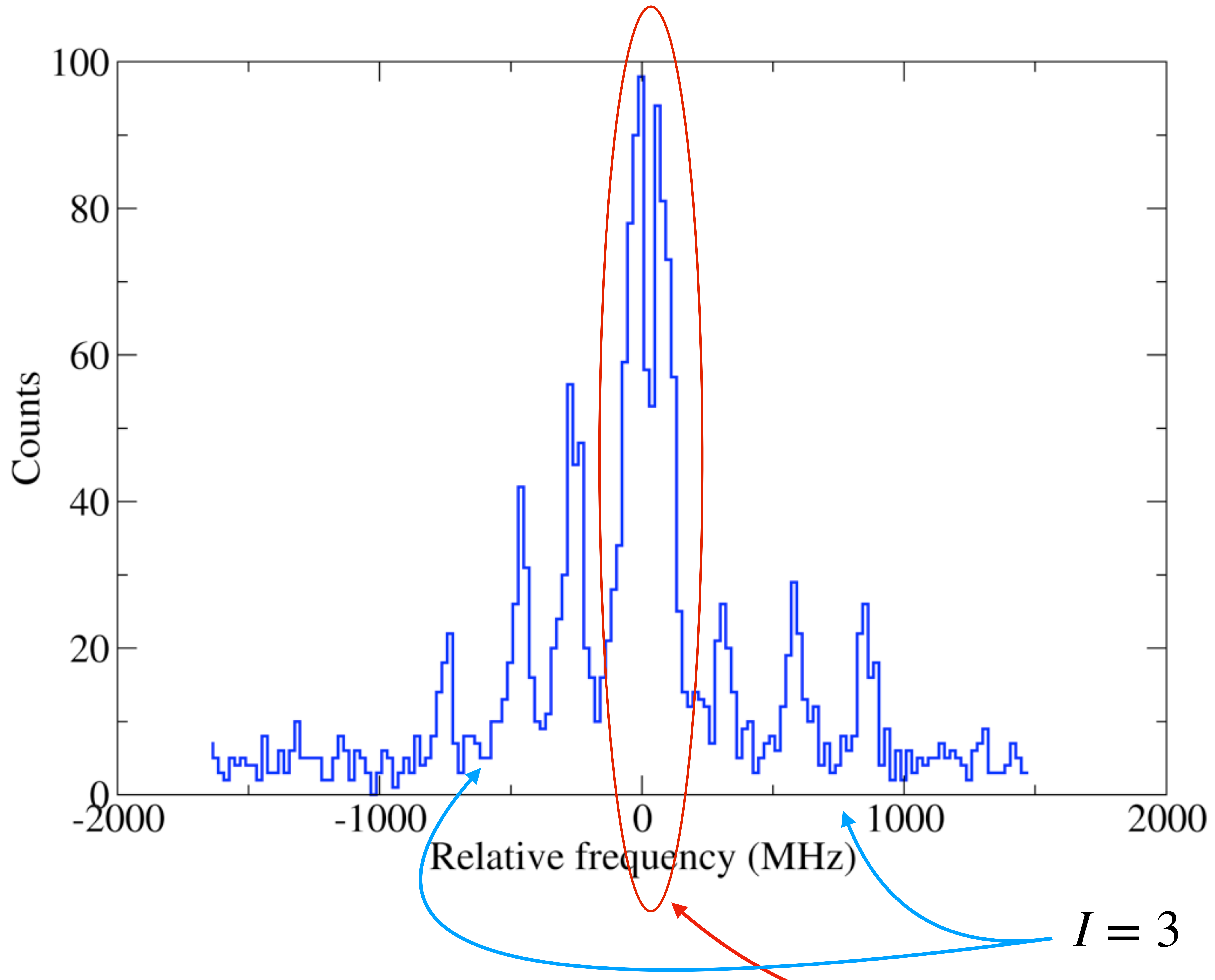



Decay spectroscopy may be able to assign spins to exotic high lying states and bands.

But the assignments are often relative.

These measurements then underpin assignments to higher states

# Discovery of states




This time we have too many peaks!

An example: an isomer in $^{80}Ga$:

- Could be too long lived for some decay methods
- Half-life similar to gs
- Too low-lying
  - very similar mass

$I = 3$

$I = (6)$ predicted to be the gs. (SM calculations)

Cheal *et al.* Phys. Rev. C **82**, 051302(R)

# Discovery of states

**Recommended† magnetic dipole moments**

| Isotope | Energy [keV] | $t_{1/2}$ | Spin/Parity | μ [nm] | Method | Recommended Tables |
|---|---|---|---|---|---|---|
| $^{80}Ga$ | 0 | 1.9 s | 6(-) | +0.036(4) | CLS | INDC(NDS)-0915 |
| | 22.4 | 1.3 s | 3(-) | -1.425(5) | CLS | INDC(NDS)-0794 |

Spins have (correctly?) lost their parentheses

**Recommended† electric quadrupole moments**

| Isotope | Energy [keV] | $t_{1/2}$ | Spin/Parity | Q [b] | Method | Recommended Tables |
|---|---|---|---|---|---|---|
| $^{80}Ga$ | 0 | 1.9 s | (3-) | +0.38(2) | CLS | INDC(NDS)-0833 |
| | 22.4 | 1.3 s | (6-) | +0.48(3) | CLS | INDC(NDS)-0833 |

Confusion regarding states
(but moments and spins correspond ✔)

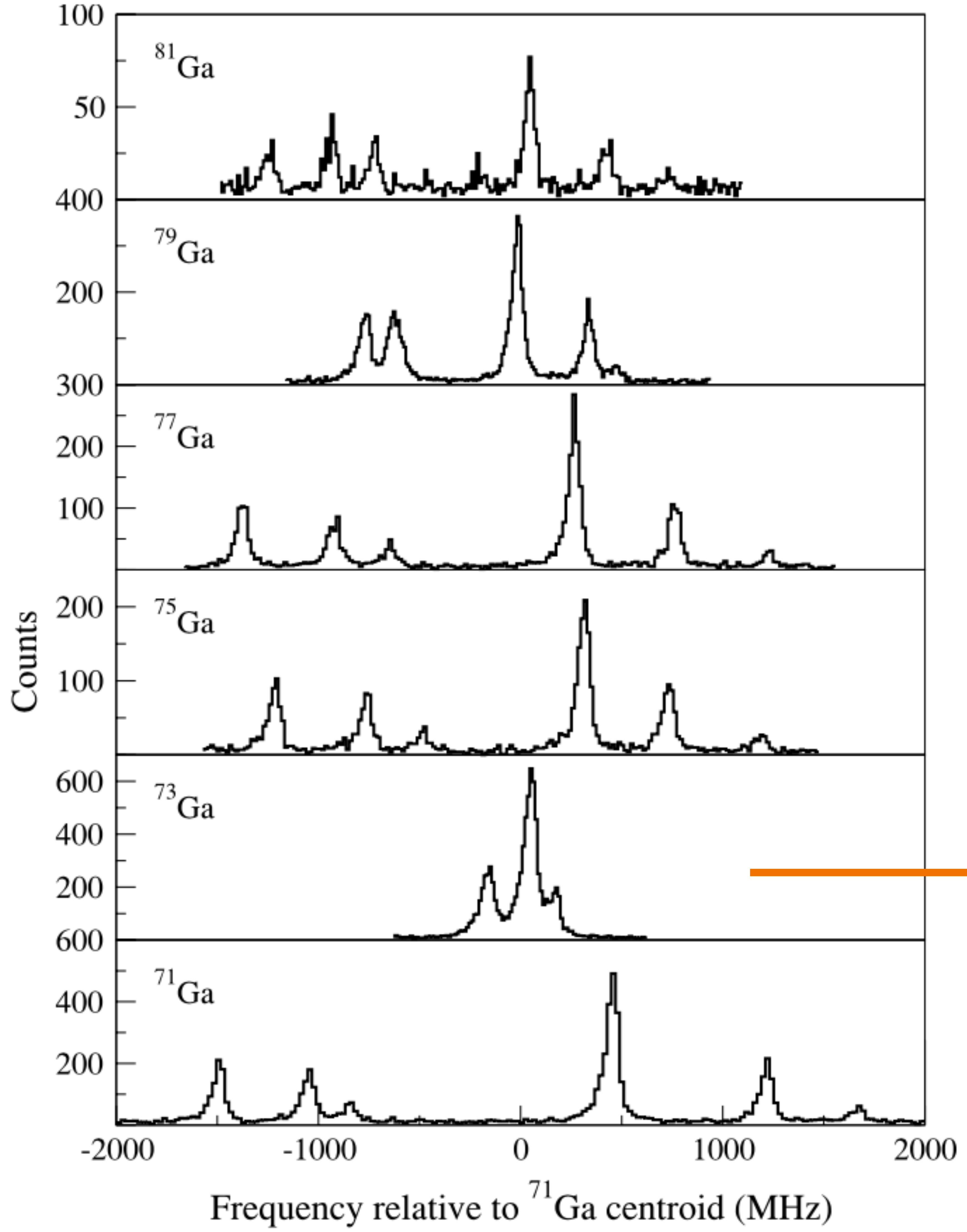
**Compiled‡ magnetic dipole and electric quadrupole moments** from published articles

| Isotope | Energy [keV] | $t_{1/2}$ | Spin/Parity | μ [nm] | Q [b] | Ref. Std | Method | NSR keyword | doi |
|---|---|---|---|---|---|---|---|---|---|
| $^{80}Ga$ | 0 | 1.9 s | ($6^-$) | +0.036(4) | | [$^{71}Ga$] | CLS | 2010CH50 | 10.1103/PhysRevC.82.051302 |
| | | | | | +0.48(3) | [$^{71}Ga$] | CLS | 2010CH50 | 10.1103/PhysRevC.82.051302 |
| | 22.4 | 1.3 s | ($3^-$) | -1.425(5) | | [$^{71}Ga$] | CLS | 2010CH50 | 10.1103/PhysRevC.82.051302 |
| | | | | | +0.38(2) | [$^{71}Ga$] | CLS | 2010CH50 | 10.1103/PhysRevC.82.051302 |

Laser measurements alone say nothing directly about energy ordering or parity.
(But can <u>sometimes</u> roughly estimate half-lives, or correlate through production yields with eg. masses)

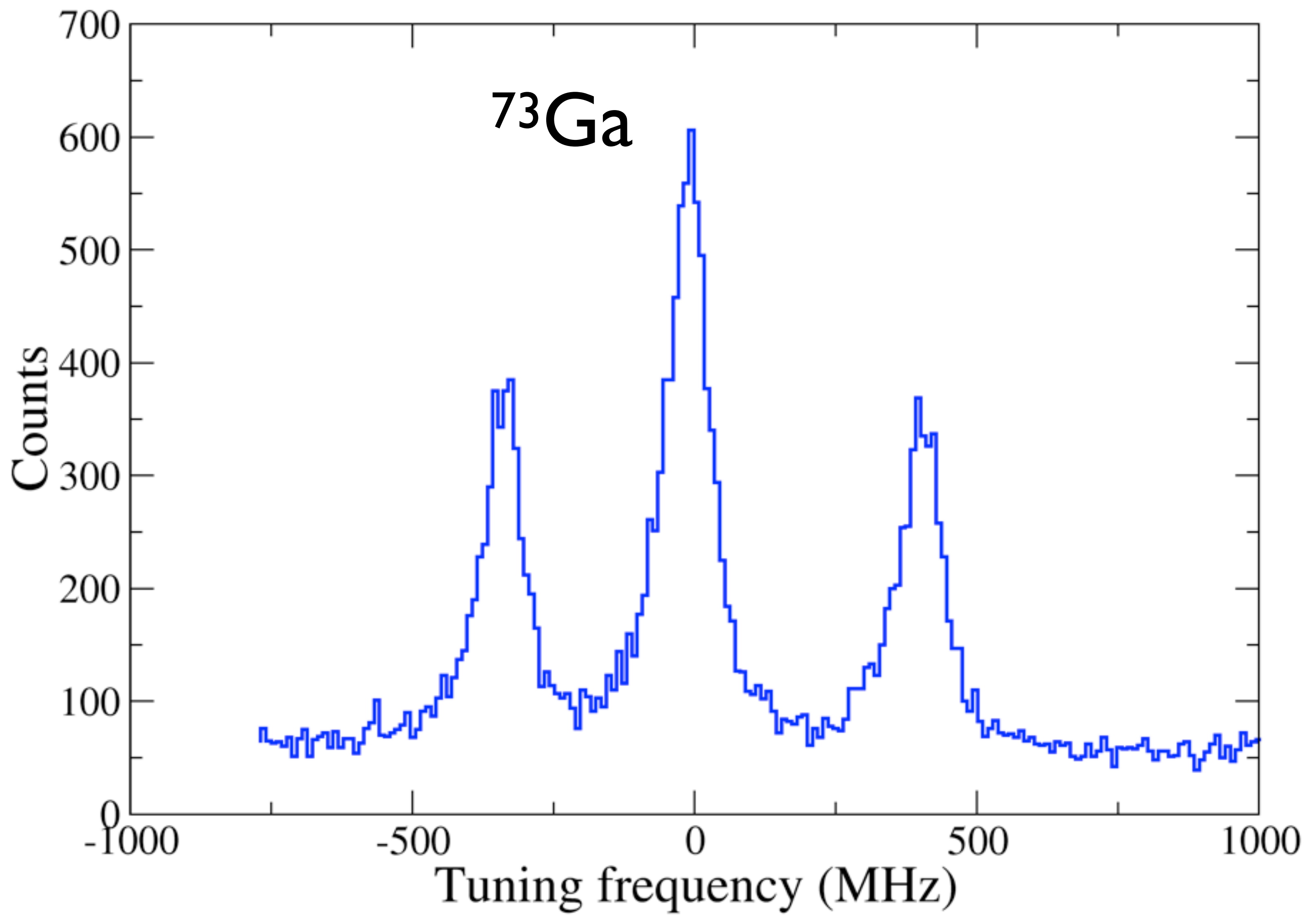
# Sometimes we even discover the ground state

Ga: J=3/2 → J=1/2




The ground states of $^{71,73,75,77}$Ga were all firmly assigned as *I* = 3/2

Remeasure on a J=1/2 → J=1/2 line




But it is clearly *I* = 1/2.

Cheal *et al.* Phys. Rev. Lett. **104**, 252502

# Sometimes we even discover the ground state

## NUCLEAR ELECTROMAGNETIC MOMENTS

The present compilation includes experimental information on nuclear magnetic dipole and electric quadrupole moments found in print compilations (such as IND
journals, international conferences and other resources. The online interface was created by Theo J. Mertzimekis under the IAEA auspices.

**New**: Recommended magnetic dipole moments for long-lived states (INDC(NDS)-0794) and short-lived states (INDC(NDS)-0816), and recommended electric qu
recommended tables have been produced by N.J. Stone. Detailed discussion with Karel Jackowski (diamagnetism), Pekka Pyykko (electric field gradient compu
acknowledged.

Periodic Table | Z-Helix | Elementary Particles | CSV file with recommended nuclear dipole moments | CSV file with recommended nuclear quadrupole moments | CSV file

**Gallium (Z=31)**

$^{63}Ga$ $^{66}Ga$ $^{67}Ga$ $^{68}Ga$ $^{69}Ga$ $^{70}Ga$ $^{71}Ga$ $^{72}Ga$ $^{73}Ga$ $^{74}Ga$ $^{75}Ga$ $^{76}Ga$ $^{77}Ga$ $^{78}Ga$ $^{79}Ga$ $^{80}Ga$ $^{81}Ga$

### Recommended† magnetic dipole moments

| Isotope | Energy [keV] | $t_{1/2}$ | Spin/Parity | μ [nm] | Method | Recommended Tables |
|---|---|---|---|---|---|---|
| $^{73}Ga$ | 0 | 4.86 h | 3/2- | +0.209(2) | CLS | INDC(NDS)-0794 |

### Recommended† electric quadrupole moments

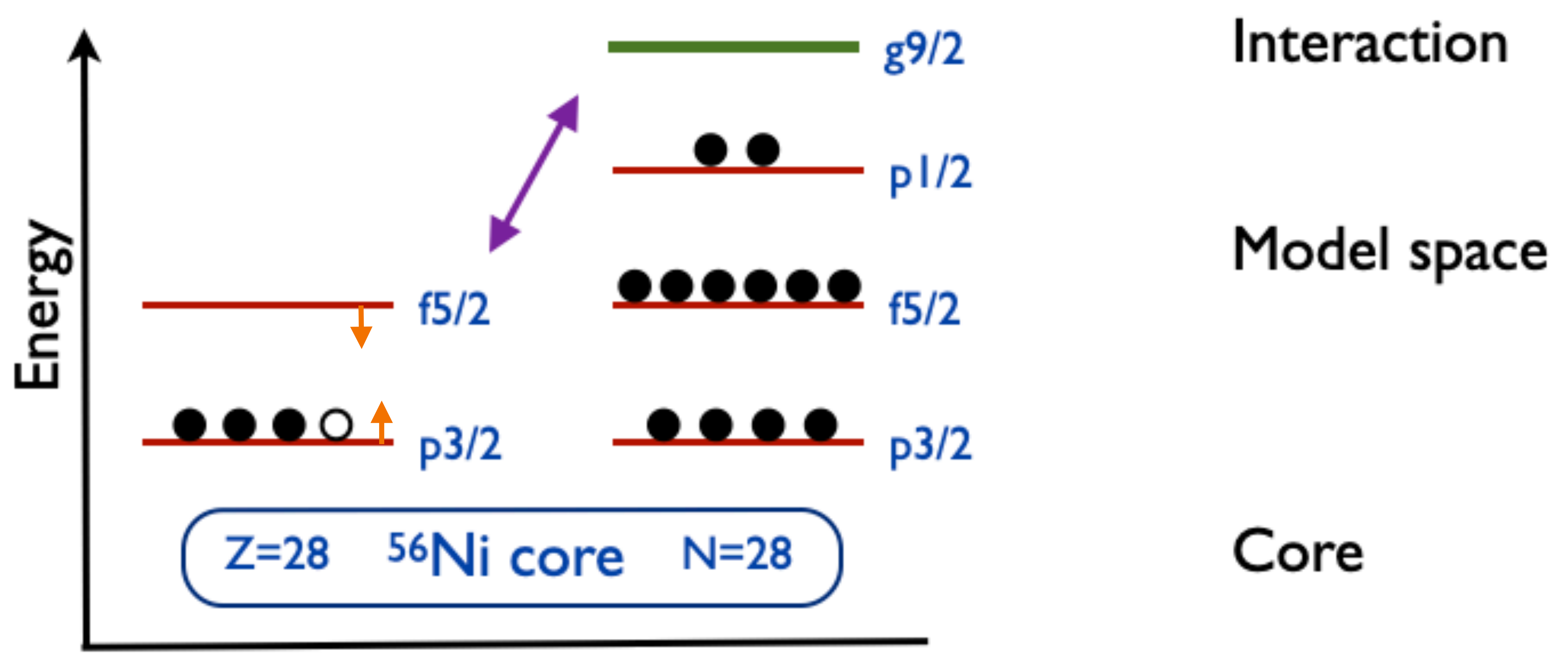
| Isotope | Energy [keV] | $t_{1/2}$ | Spin/Parity | Q [b] | Method | Recommended Tables |
|---|---|---|---|---|---|---|
| $^{73}Ga$ | 0 | 4.86 h | 3/2- | +0.209(2) | CLS | INDC(NDS)-0833 |

### Compiled‡ magnetic dipole and electric quadrupole moments from published articles

| Isotope | Energy [keV] | $t_{1/2}$ | Spin/Parity | μ [nm] | Q [b] | Ref. Std | Method | NSR keyword | doi |
|---|---|---|---|---|---|---|---|---|---|
| $^{73}Ga$ | 0. | 4.86 h | $3/2^-$ | +0.209(2) | | [$^{71}Ga$] | CLS | 2010CH16 2013STZZ | 10.1103/PhysRevLett.104.252502 |

# Testing shell model calculations

Testing shell model calculations in (copper and) gallium
How do nuclear properties change moving away from the valley of stability?
Proton level migrations as we increase the neutrons excess...



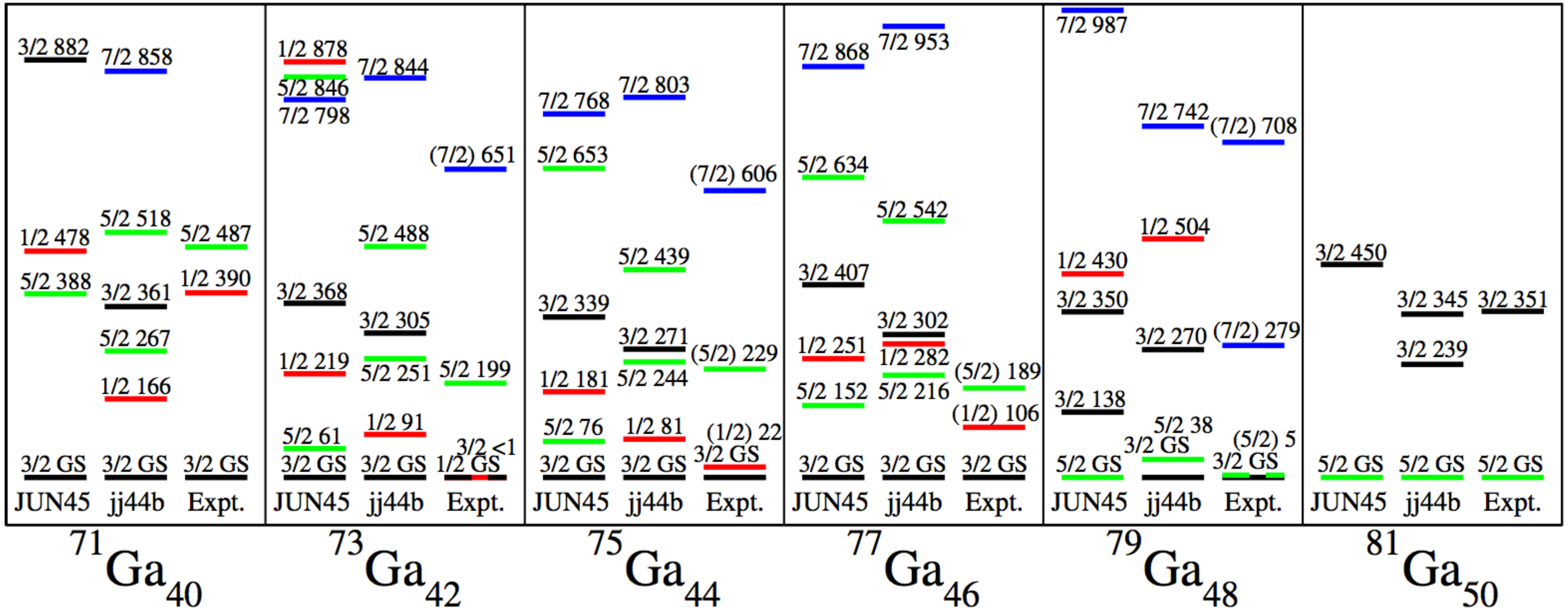

g9/2 "attracts" f5/2,
"repulses" p3/2

"Inversion" of ground state spin seen?

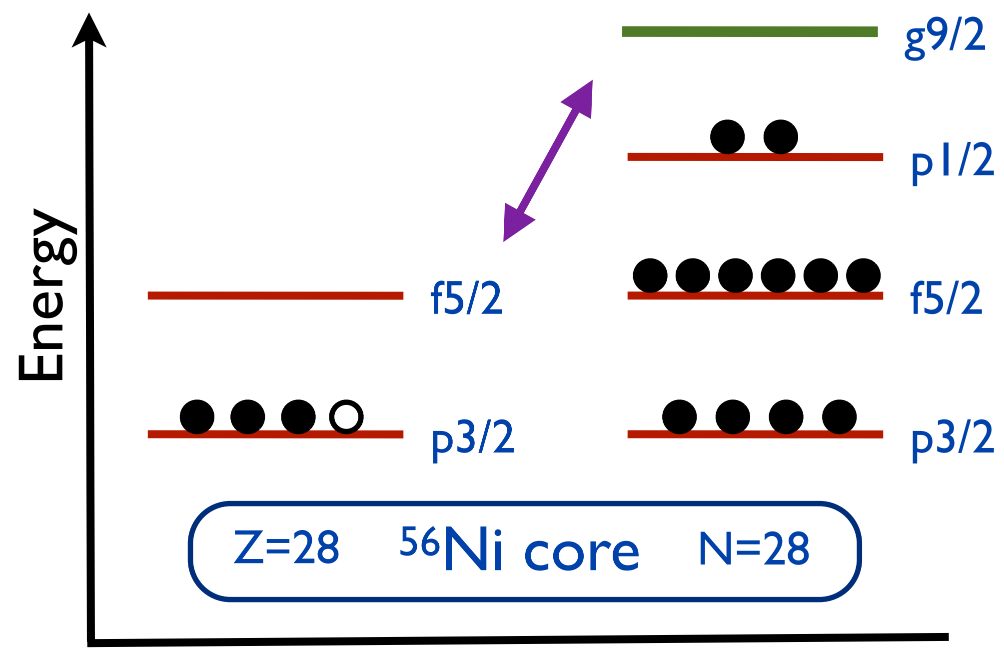
Otsuka *et al.* Phys. Rev. Lett. **95**, 232502

# Testing shell model calculations

Yes, an "Inversion" of ground state spin is indeed seen




I=3/2 I=3/2 I=3/2 I=5/2



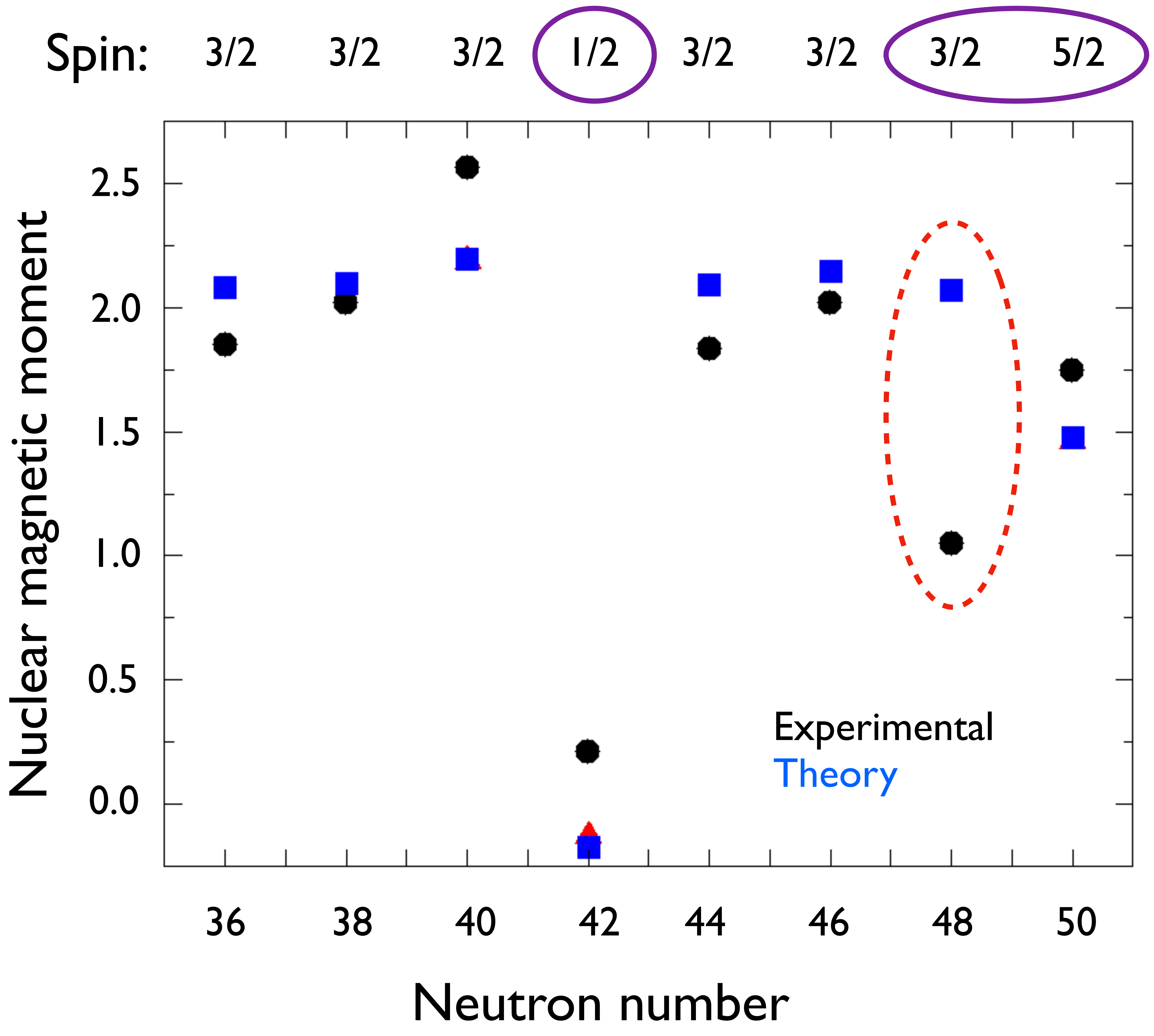

…and apparently predicted by one theory (jj44b interaction)…

Cheal *et al.* Phys. Rev. Lett. **104**, 252502

# What can we learn from the moments?

"Inversion" of ground state spin seen

But is it "correctly" predicted?




Cheal *et al.* Phys. Rev. Lett. **104**, 252502

# What can we learn from the moments?

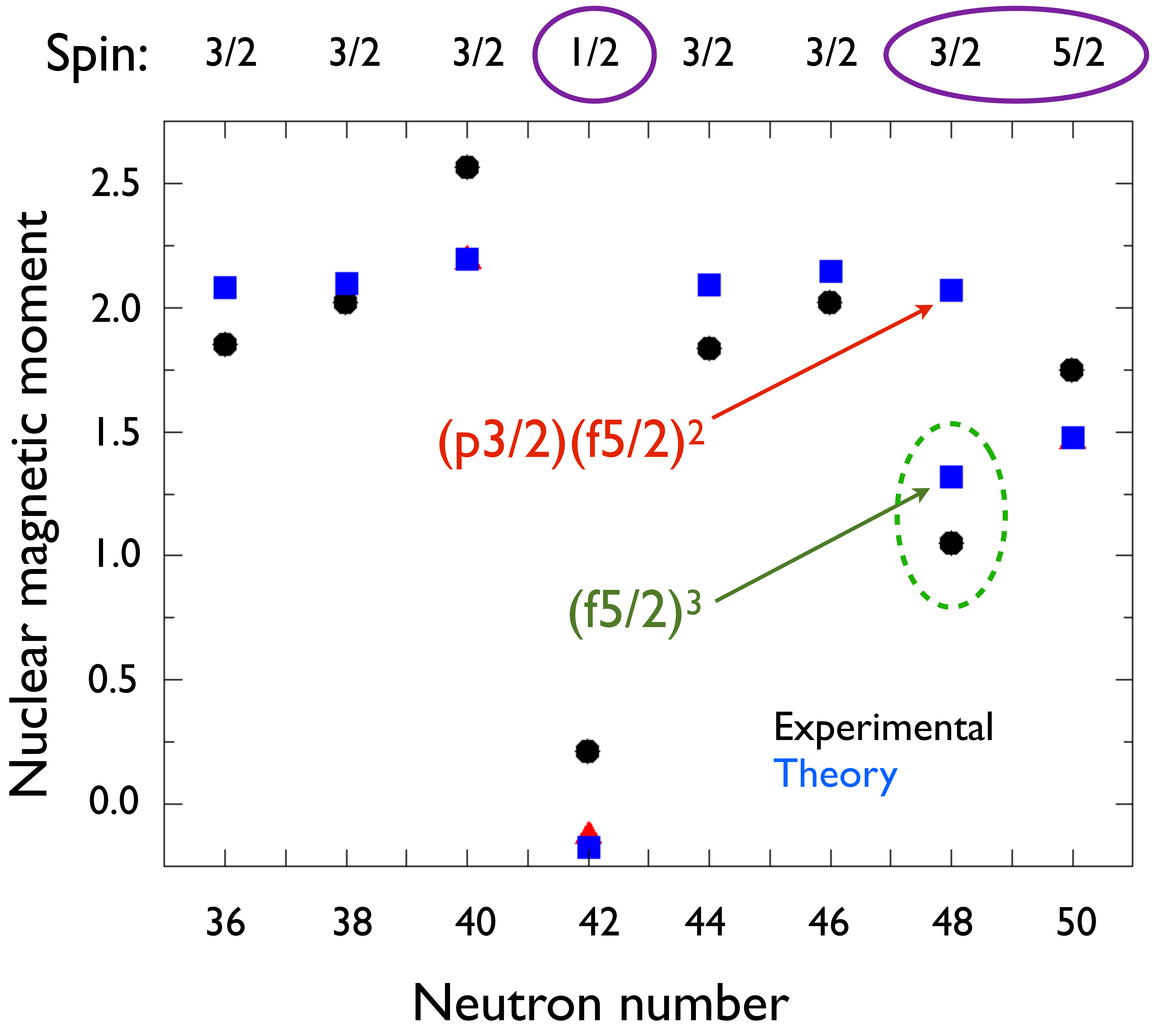
"Inversion" of ground state spin seen

- Predict (p3/2)(f5/2)$^2$ as gs configuration
- Moments suggest configuration is

$\pi$[(f5/2)$^3$]3/2

(thought to be the first excited 3/2 state)

Moments are a sensitive probe of $\psi$
SP occupation % → energy migration

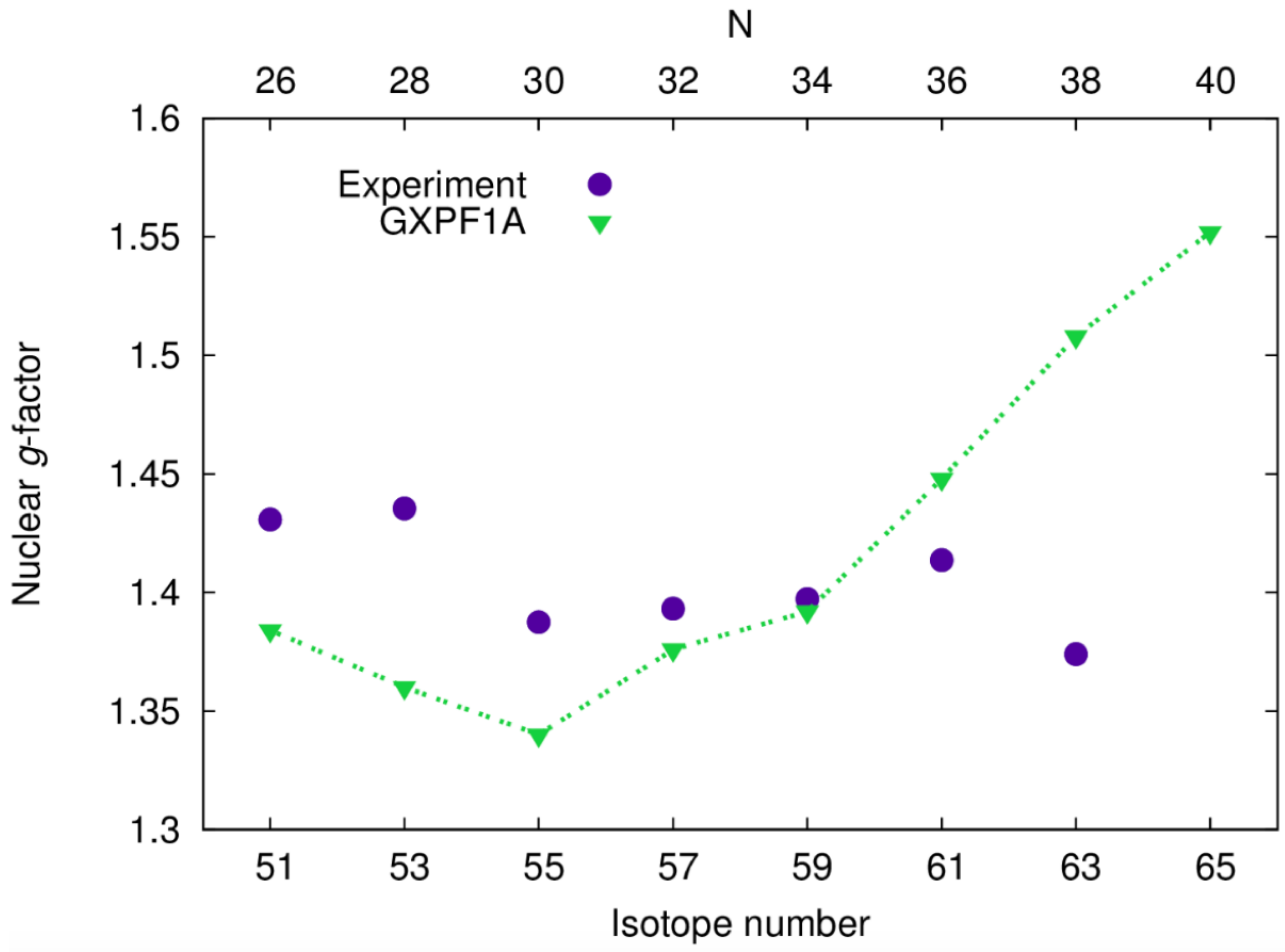


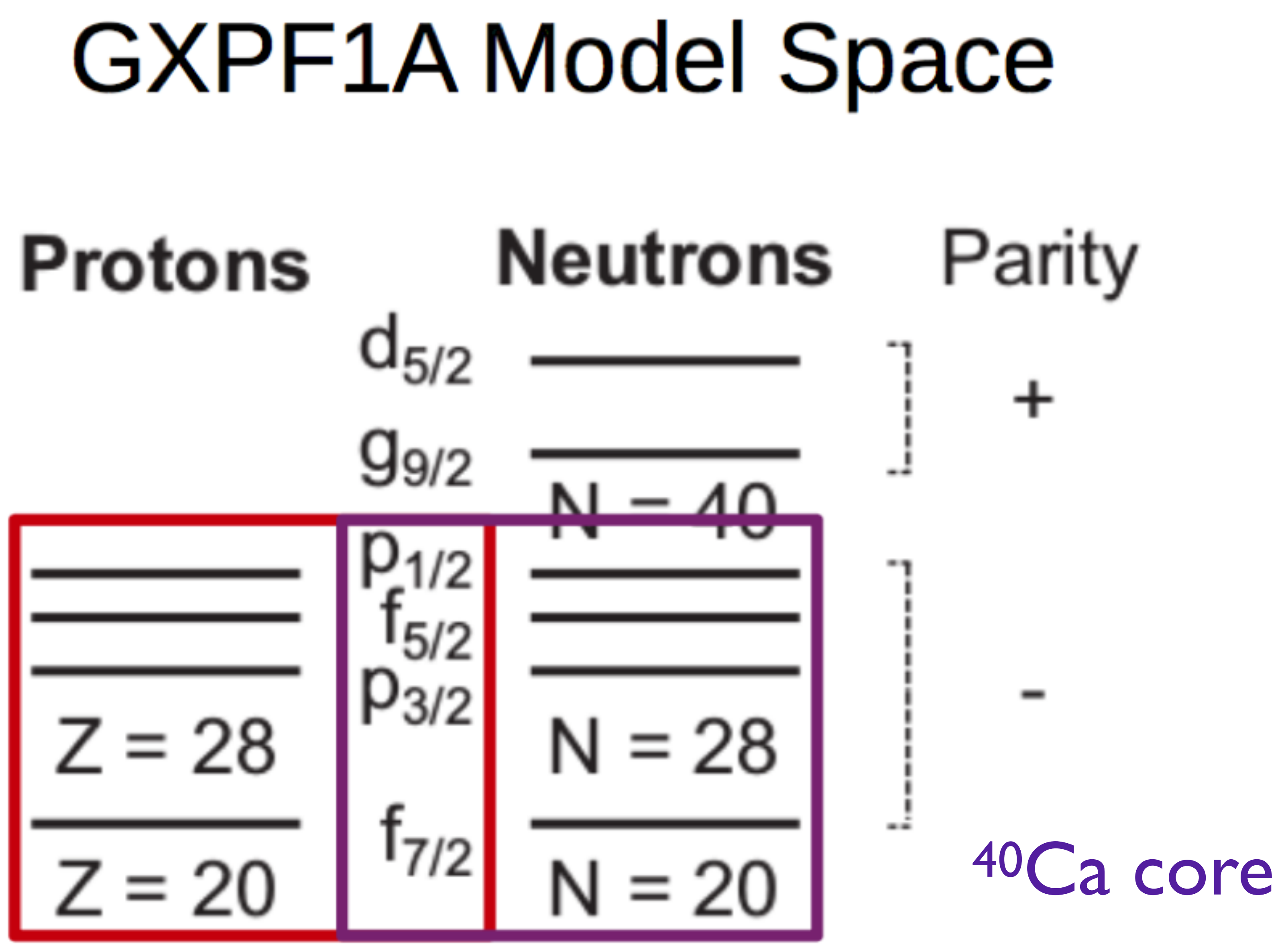

Cheal *et al.* Phys. Rev. Lett. **104**, 252502

# When the model space is not correct

Suggestions of an onset of collectivity towards N=40 for Cr, Fe…

➔ Perform laser spectroscopy of the intermediate chain of manganese



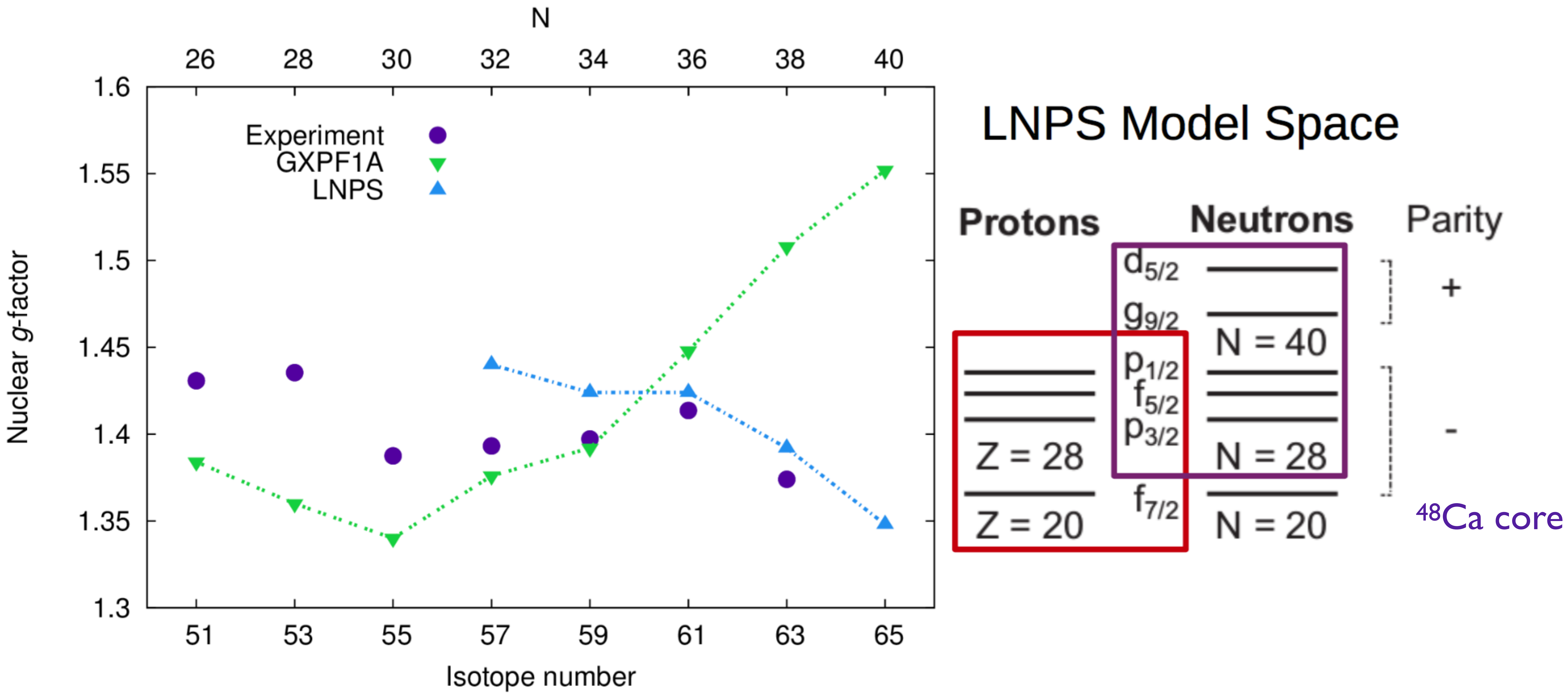




➔ Magnetic moments cannot be understood from using a pf space alone

C. Babcock, H. Heylen et al. PLB 750 176 (2015)

# When the model space is not correct

"Extend" the model space to higher neutron orbitals...

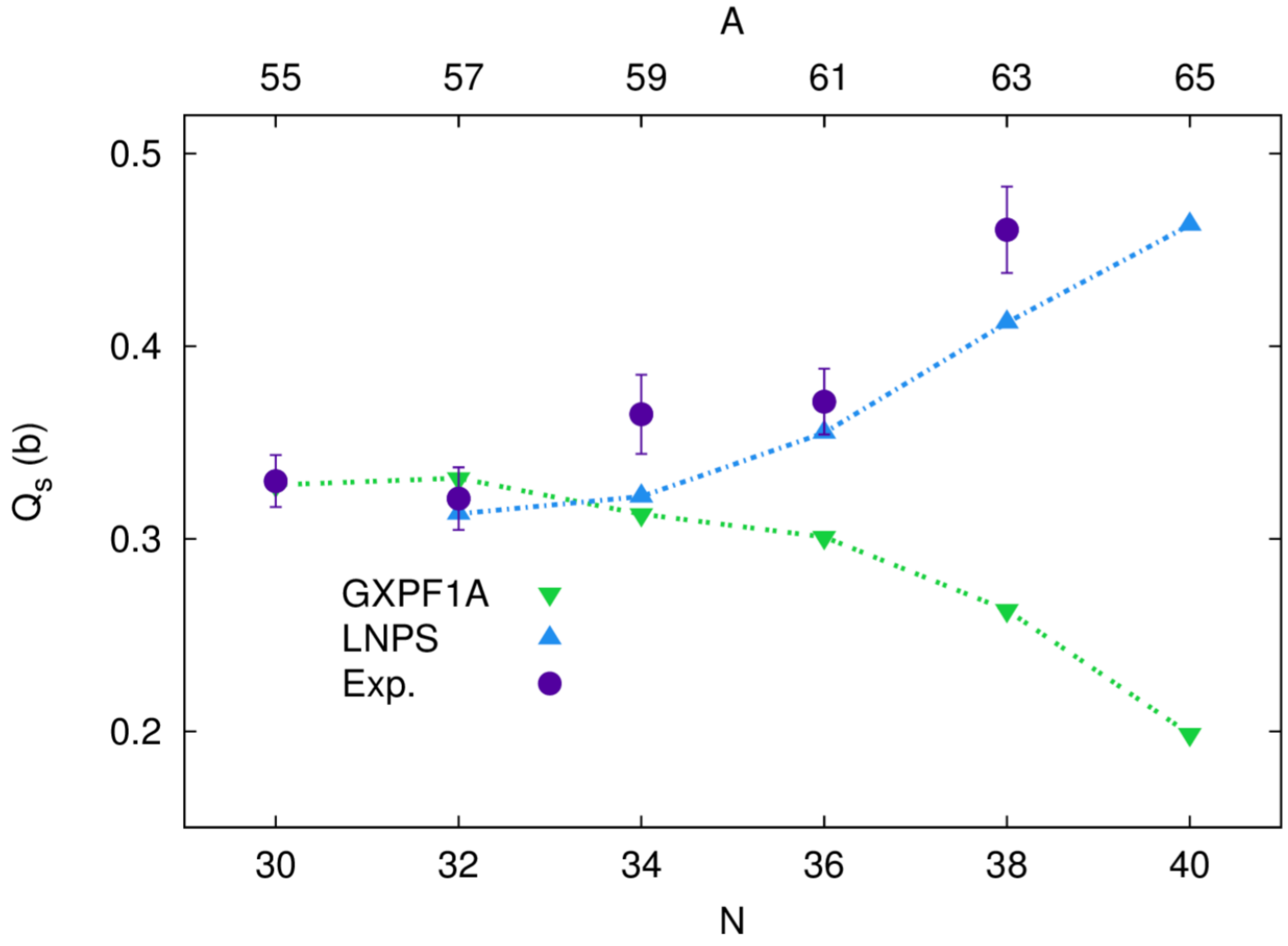


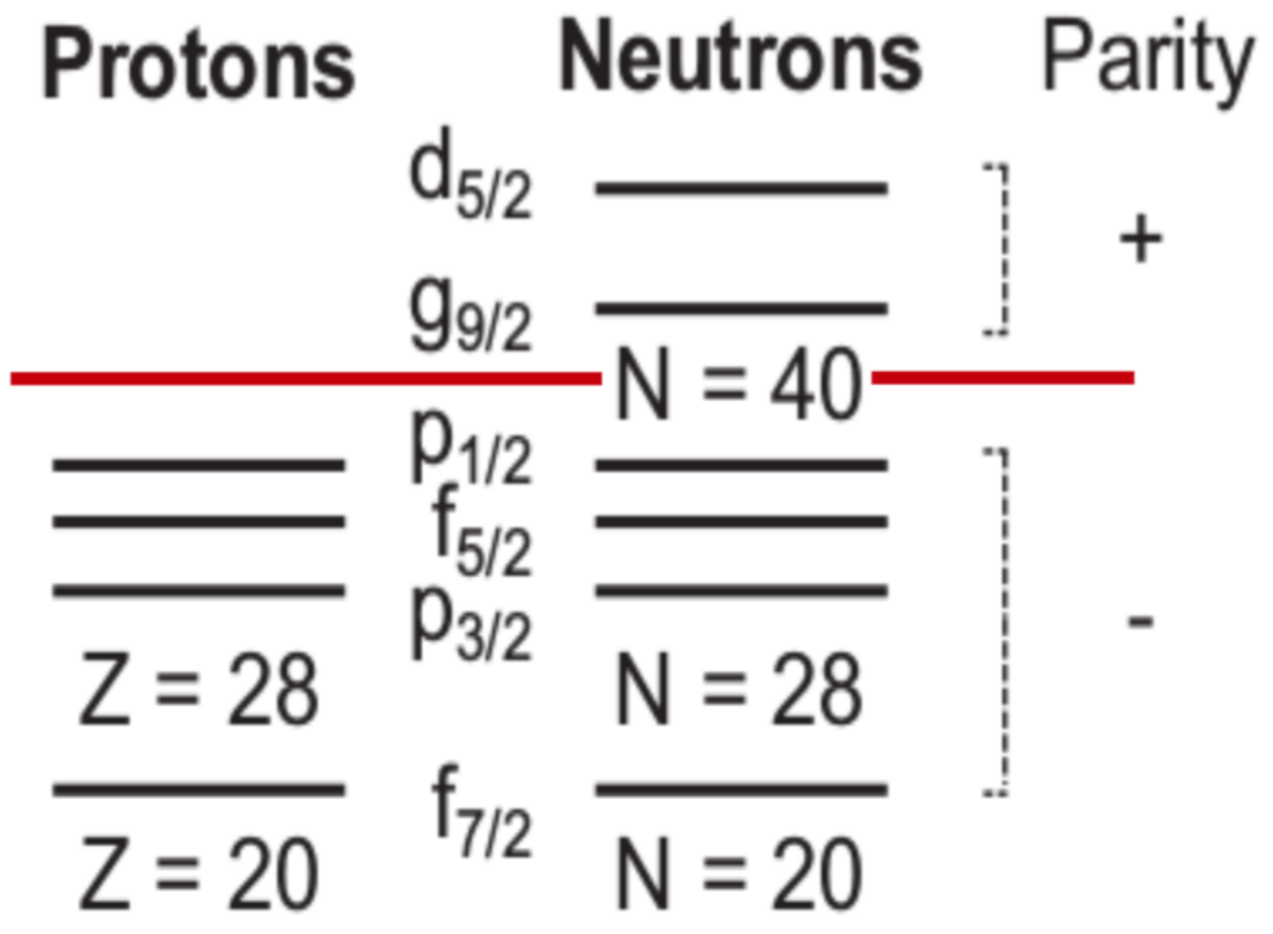

➔ Excitations indeed take place long before N=40

C. Babcock, H. Heylen et al. PLB 750 176 (2015)

# What about the quadrupole moments?

GXPF1A uses full pf space, LNPS adds the νg9/2 and νd5/2 orbitals,

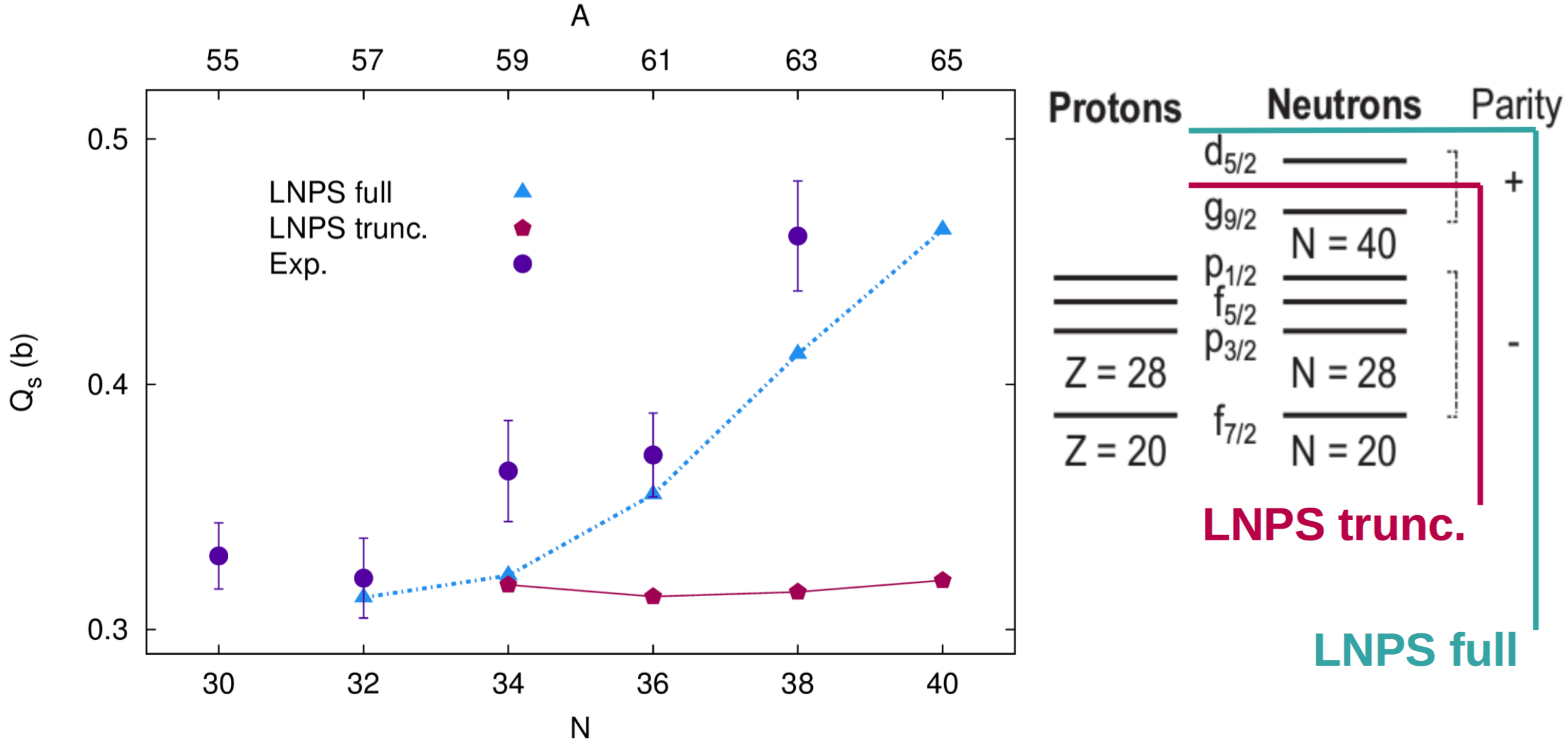



→ Again this proves to be essential, but would the g9/2 alone be sufficient?

C. Babcock, H. Heylen et al. PLB 750 176 (2015)

# What about the quadrupole moments?




Conclusion: Including the neutron d5/2 specifically is essential

C. Babcock, H. Heylen et al. PLB 750 176 (2015)

# Changes in mean-square charge radii

Size (Droplet model)

Shape (Quadrupole term)

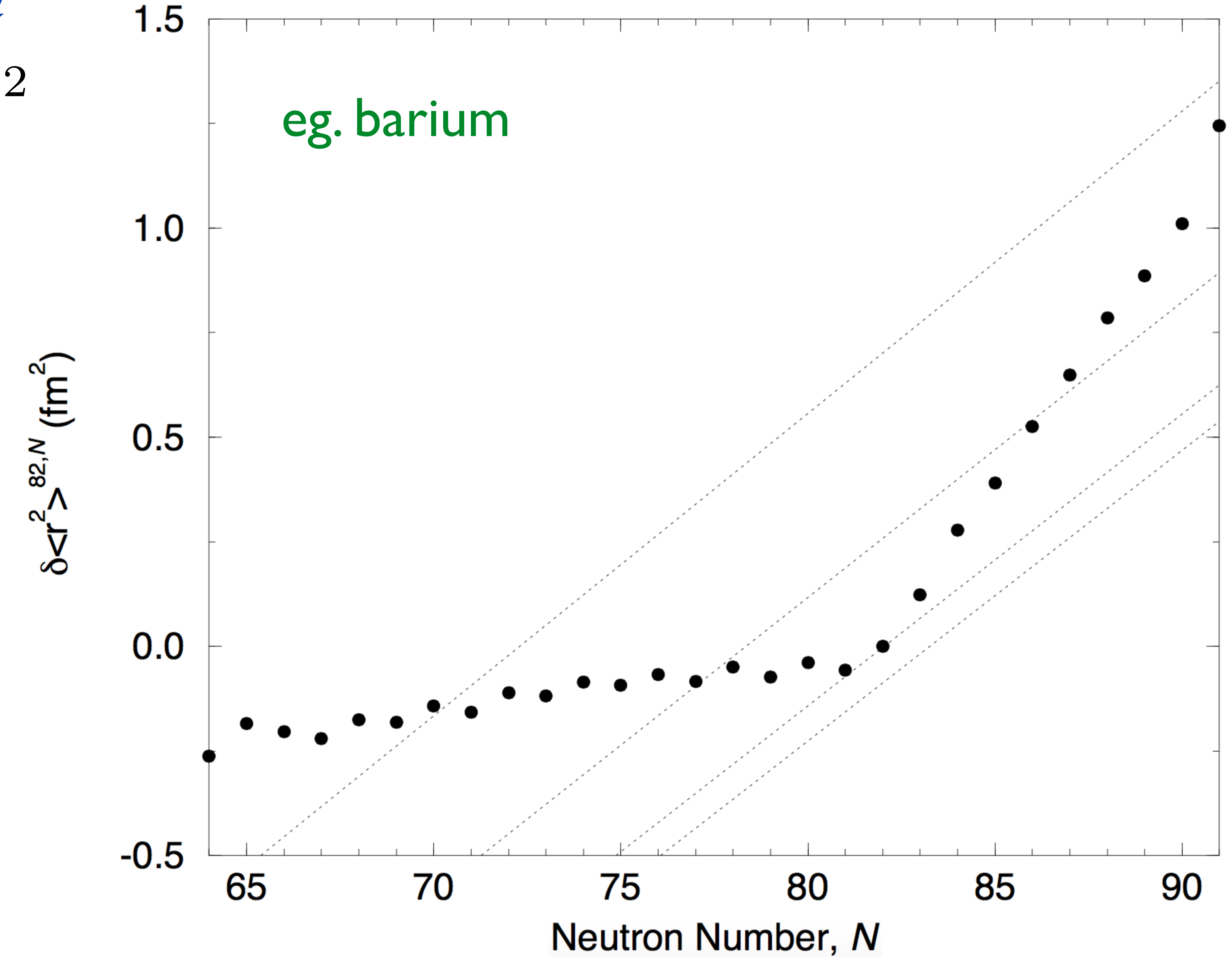
Diffuseness (assumed constant)

$$\langle r^2 \rangle = \langle r^2 \rangle_{\rm sph} \left( 1 + \frac{5}{4\pi} (\langle \beta_2^2 \rangle + \ldots) \right) + 3\sigma^2$$

Complementary probe of deformation

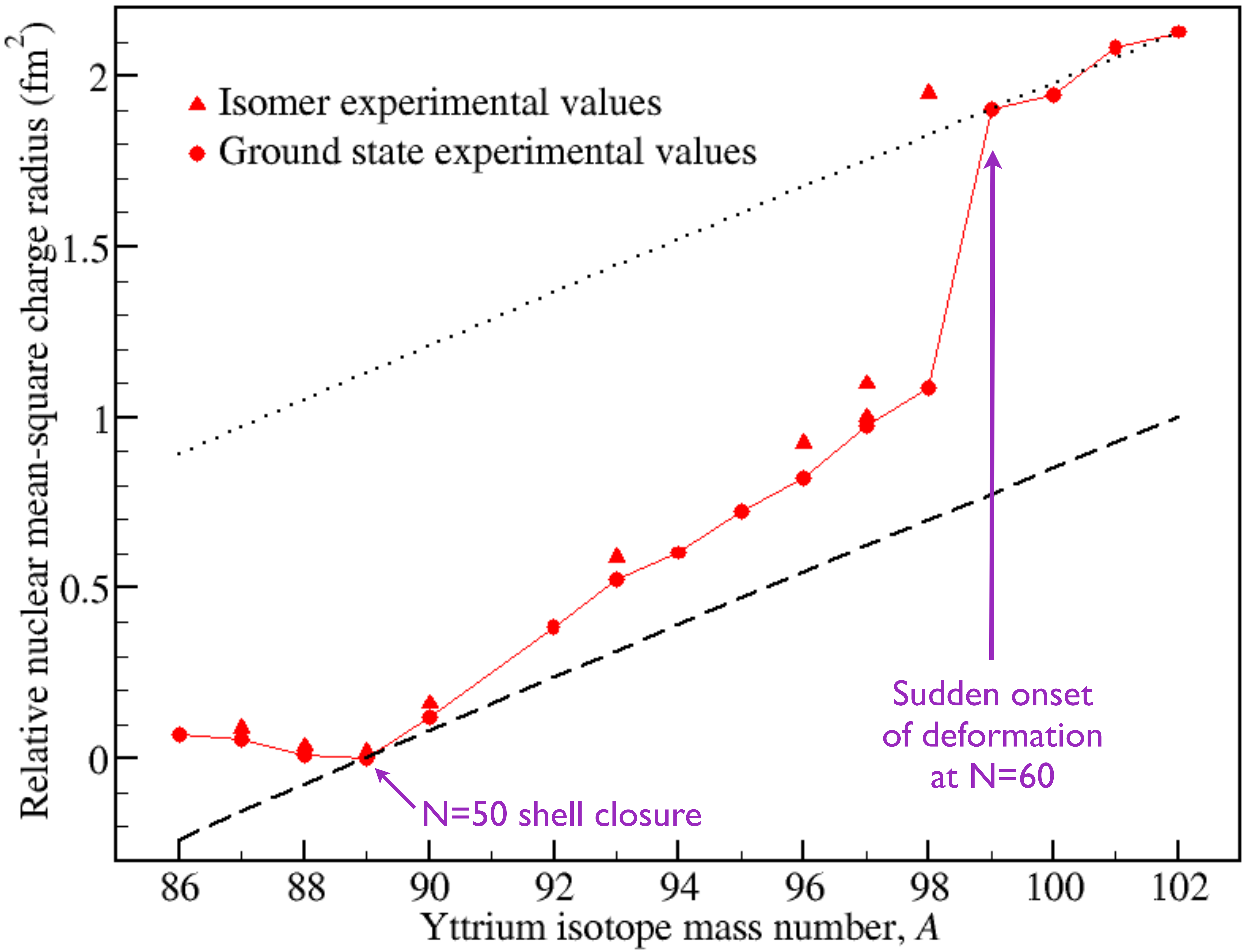
Pairing…

Magicity…



# Charge radii as a probe of deformation

Probes the deformation of ALL isotopes / states.

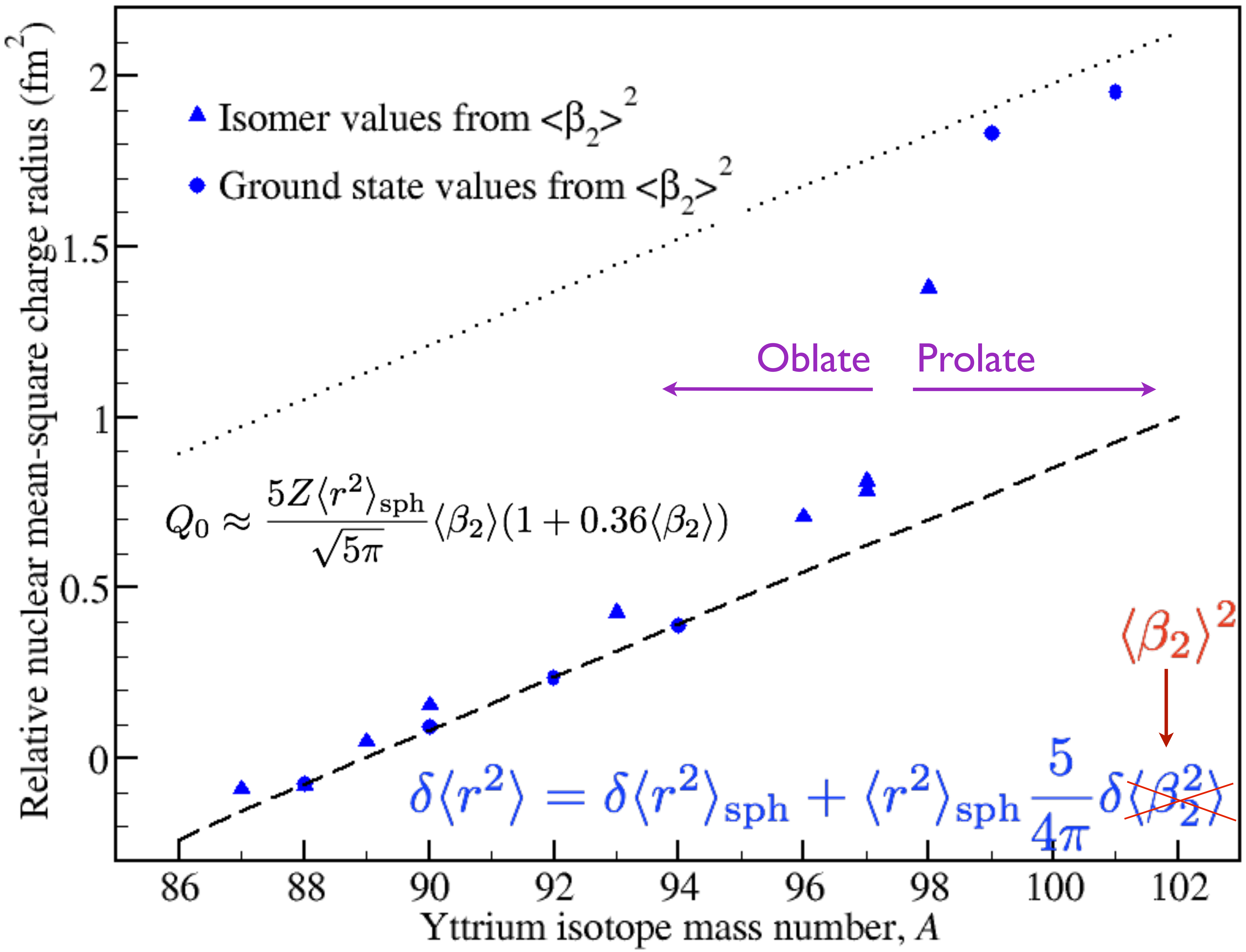



Cheal *et al.* Phys. Lett. B **645**, 133 (2007)

# Deformation from the quadrupole moments

Only probes $I > 1/2$ nuclei but tells us the sign.

But here we plot the estimated effect on the radii.

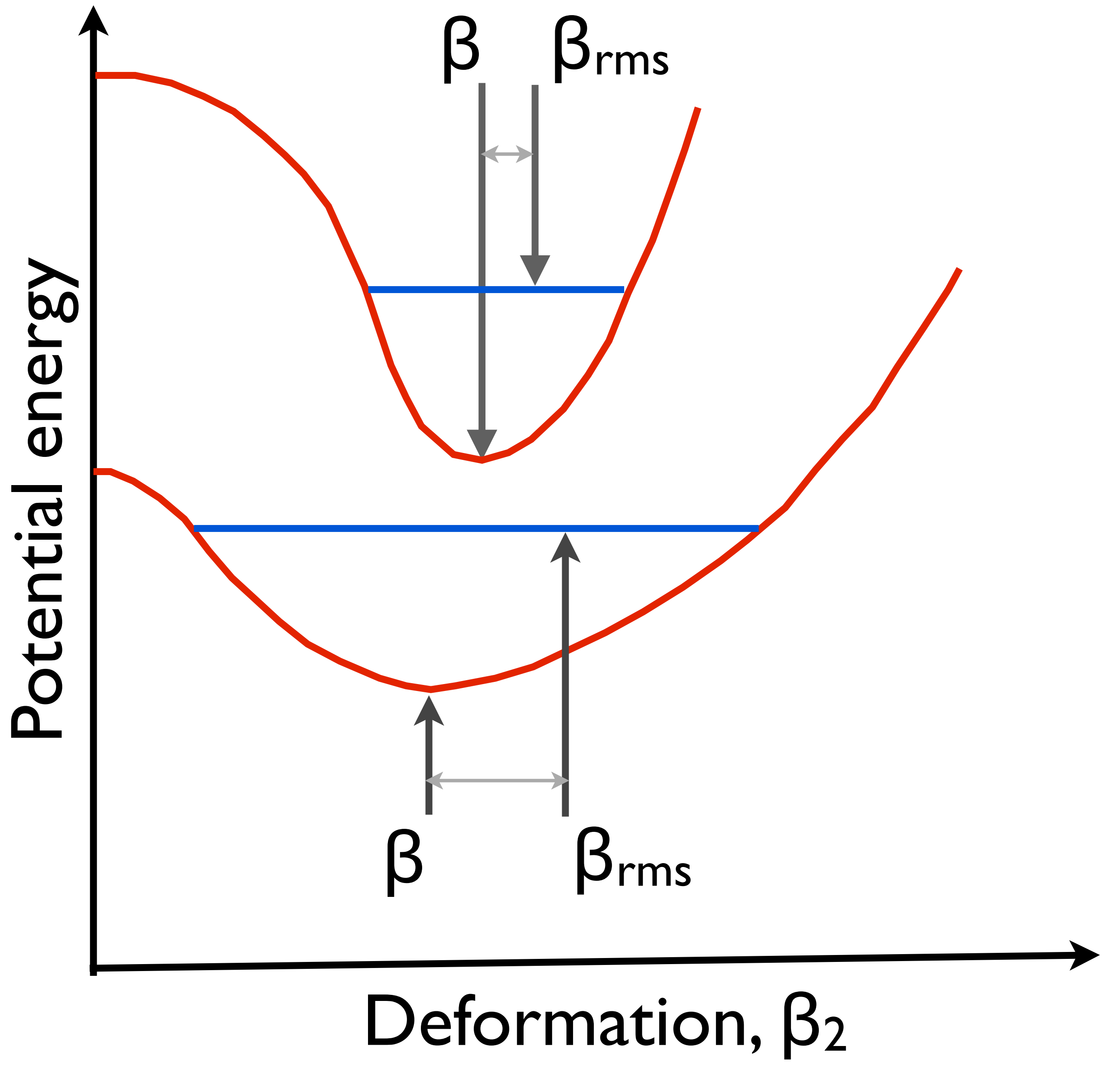


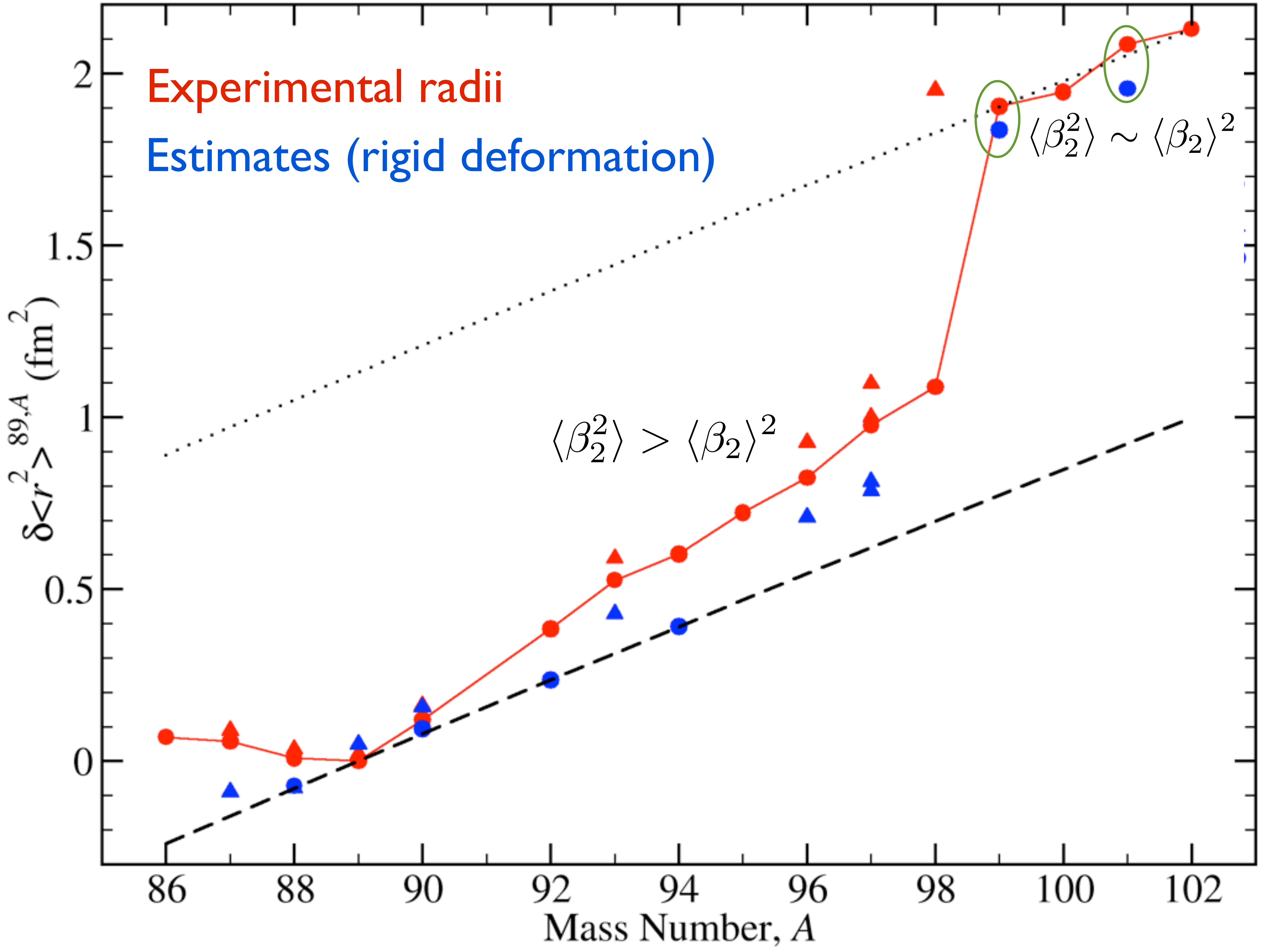

Cheal *et al.* Phys. Lett. B **645**, 133 (2007)

# Complementarity: Nuclear rigidity

$$Q_s \rightarrow \langle\beta_2\rangle$$

$$\delta\langle r^2\rangle \rightarrow \delta\langle\beta_2^2\rangle$$

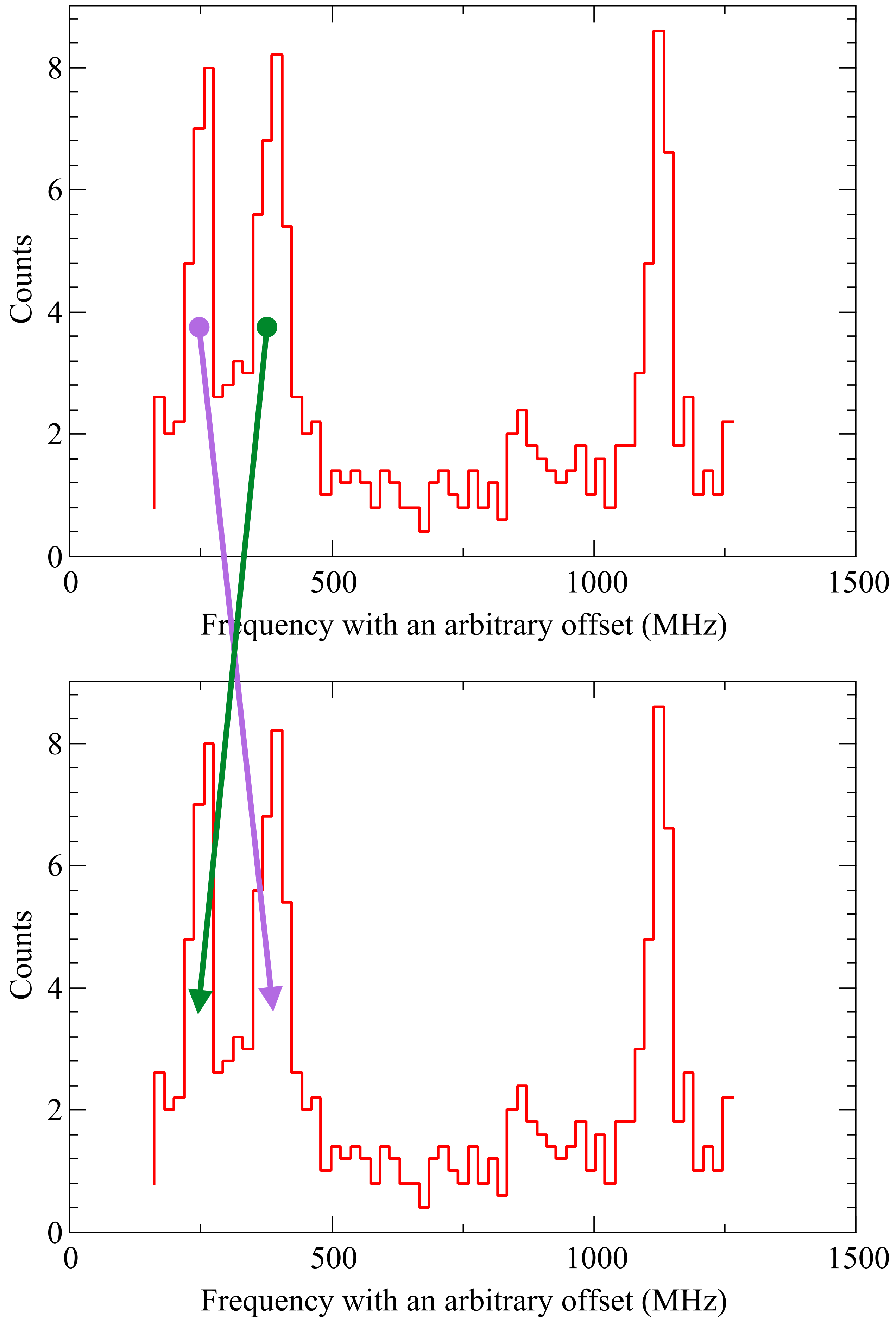



The difference gives the "softness" / "rigidity" of the deformation.

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

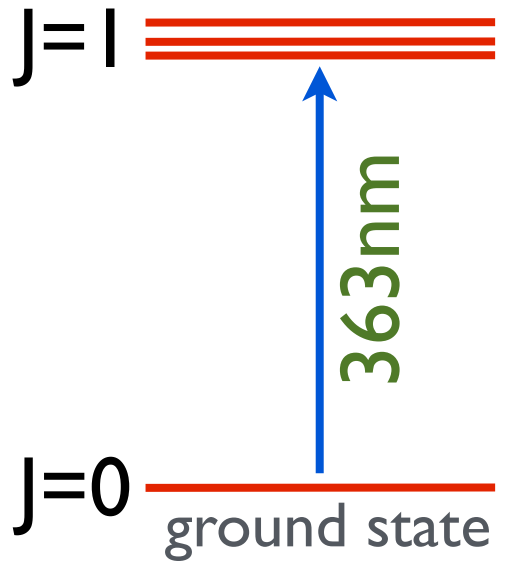
# $^{98}$Y — a shape coexistence at the critical point




The problems of J=0 ➜ J=1 lines…

Each nuclear spin fits equally well…

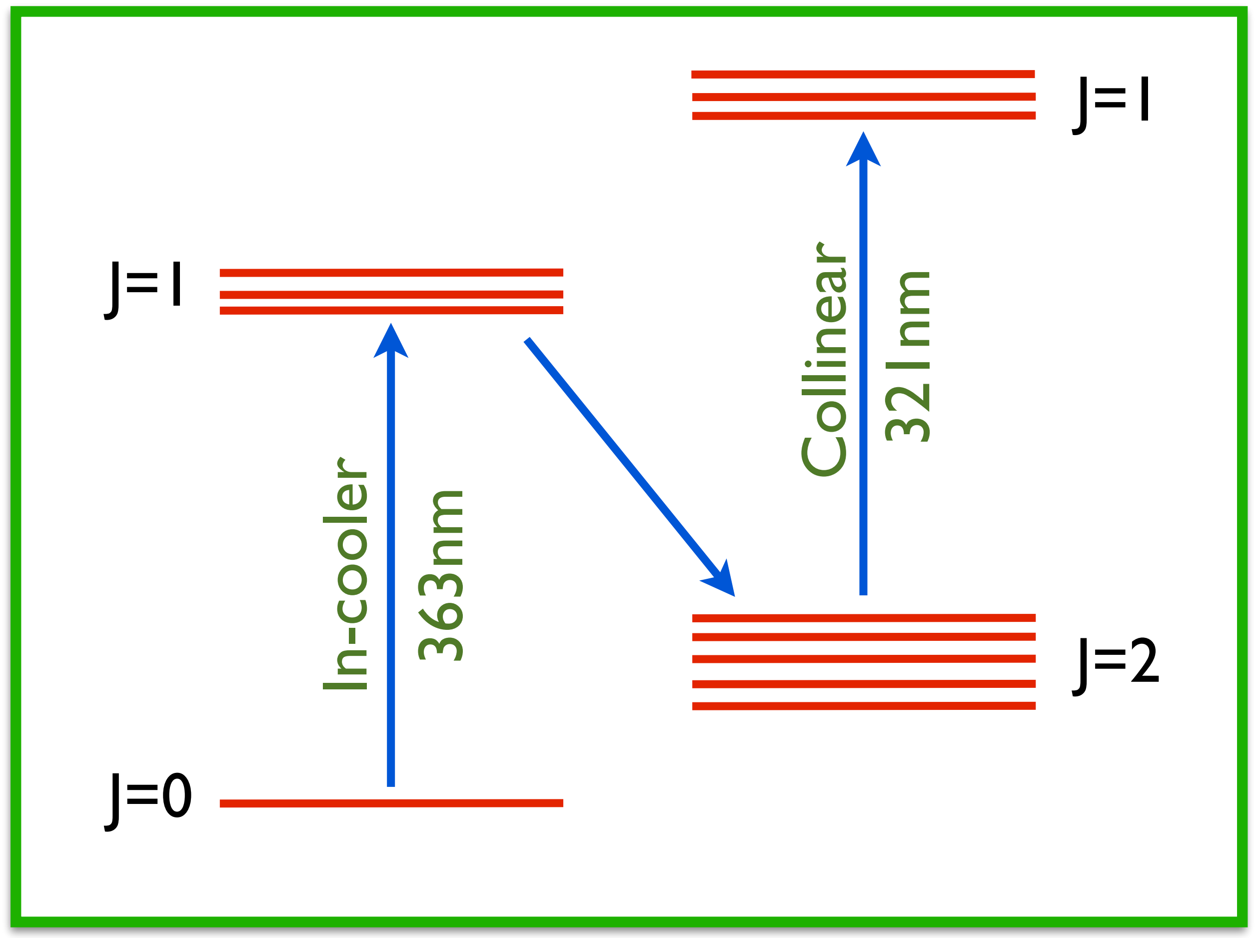
3 peaks ➜ tells us 3 unknowns (only)




| $A$ | $I^{\pi}$ | $A$ (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) |

| $A$ | $I^{\pi}$ | $\mu$ ($\mu_N$) | $Q_s$ (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 |

…and the peaks are indistinguishable.



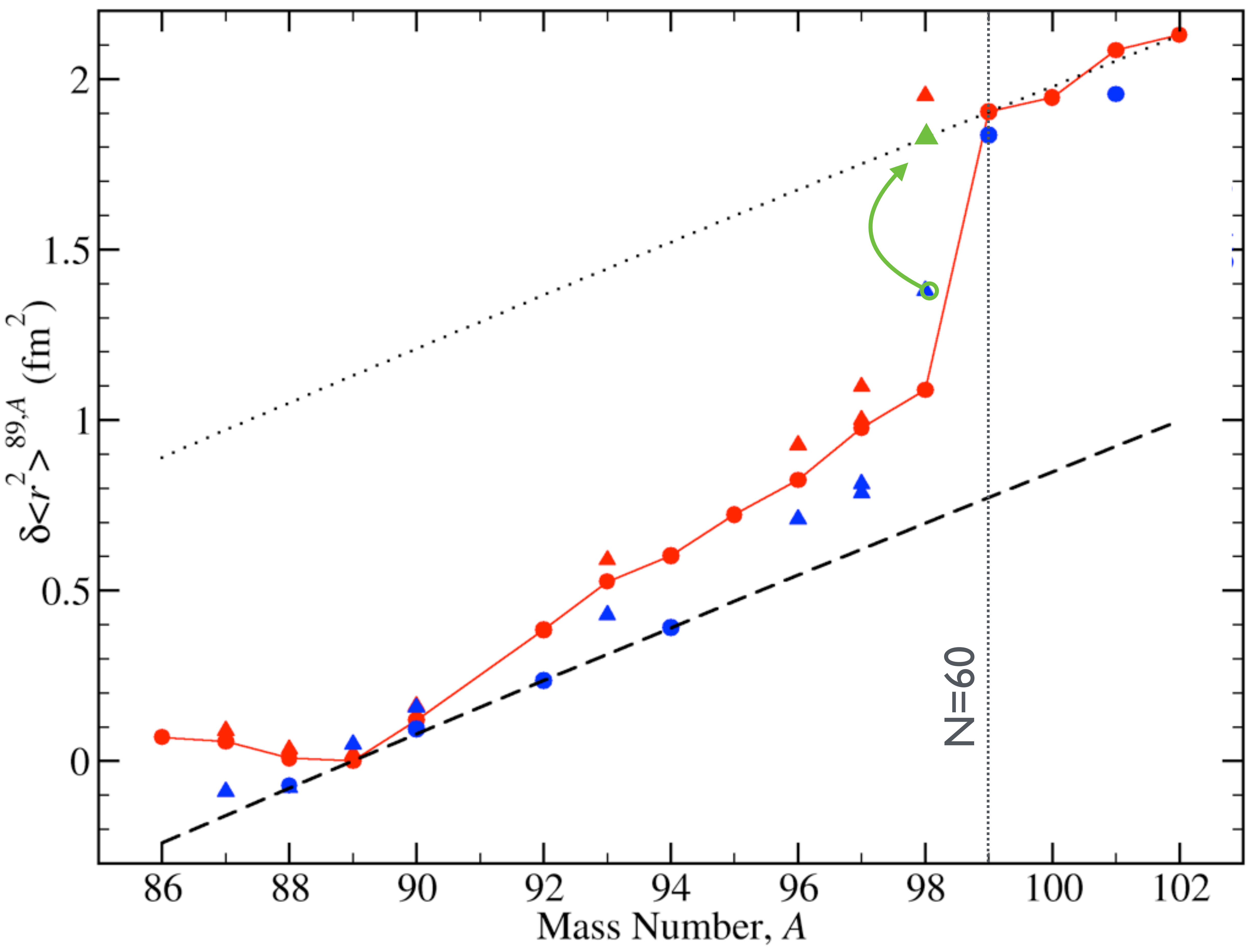

➜ Measuring on a line with more peaks now confirms that the spin of the isomer is actually much higher: $I$ = 7.

# What does this tell us?

$^{98}$Y marks the centre of the onset of deformation

Additionally, laser measurements tell us:-

- The spin, radii and moments
- The changing deformation
  both size (all isotopes) and sign
- The softness/rigidity of the deformation?



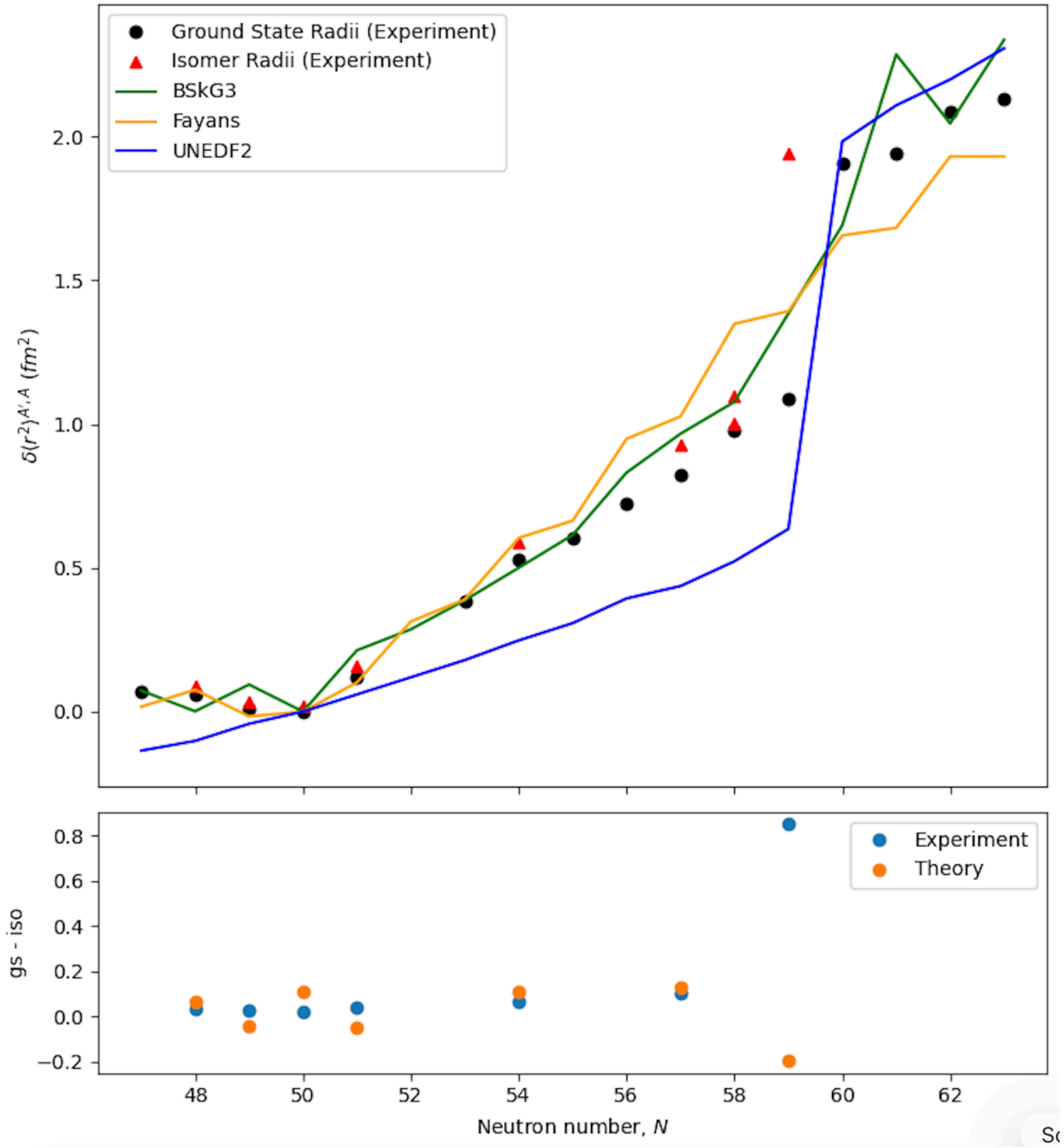

The isomer has a much higher quadrupole moment than previously thought

➜ with a strong (prolate) deformation that is very rigid

# Softness? Triaxiality? Latest DFT calculations…

Analysis in progress…

Important to benchmark calculations using quadrupole moments also

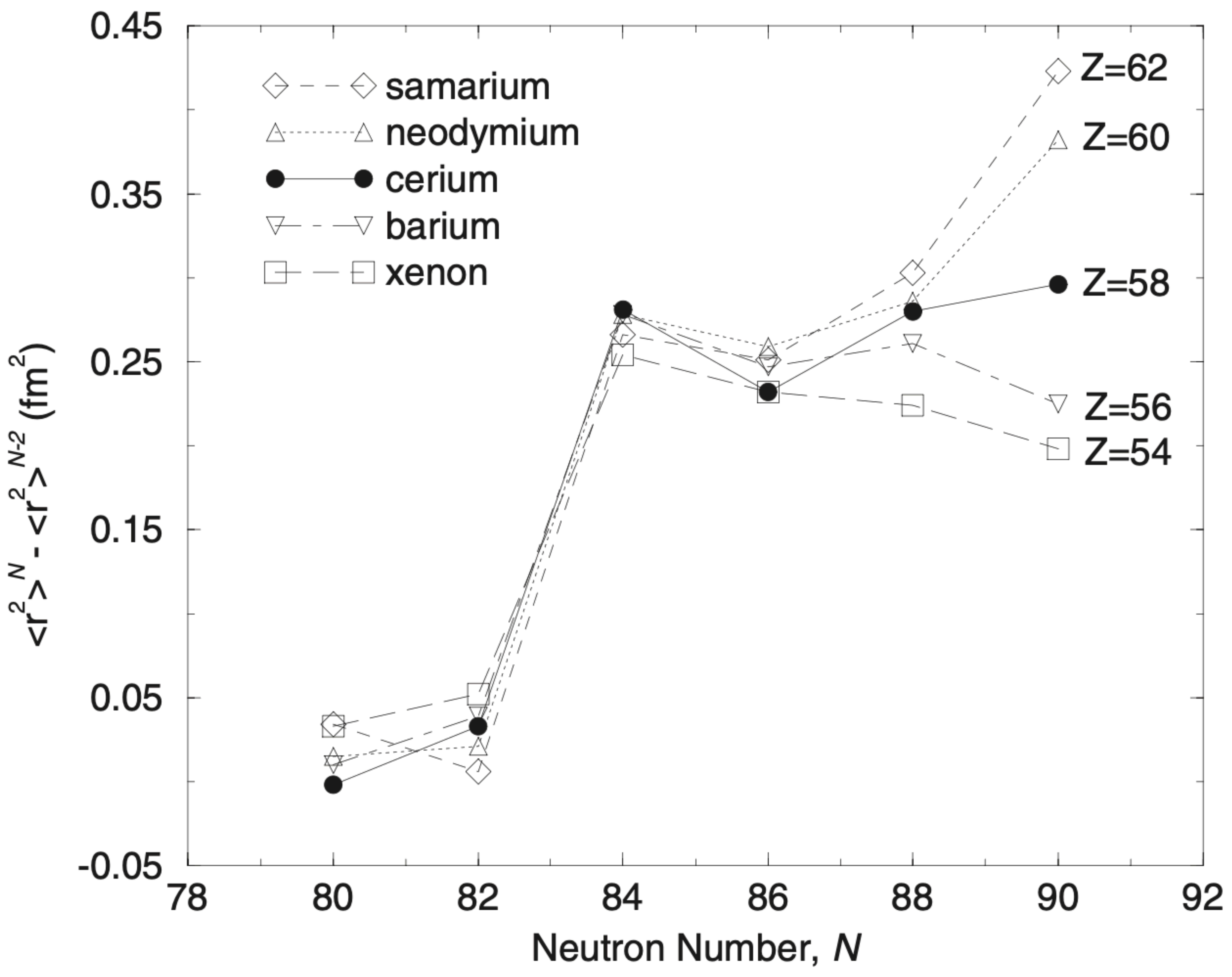


# Then from N=60 to N=88

Wish to return to cerium…

and measure odd-$A$ nuclei

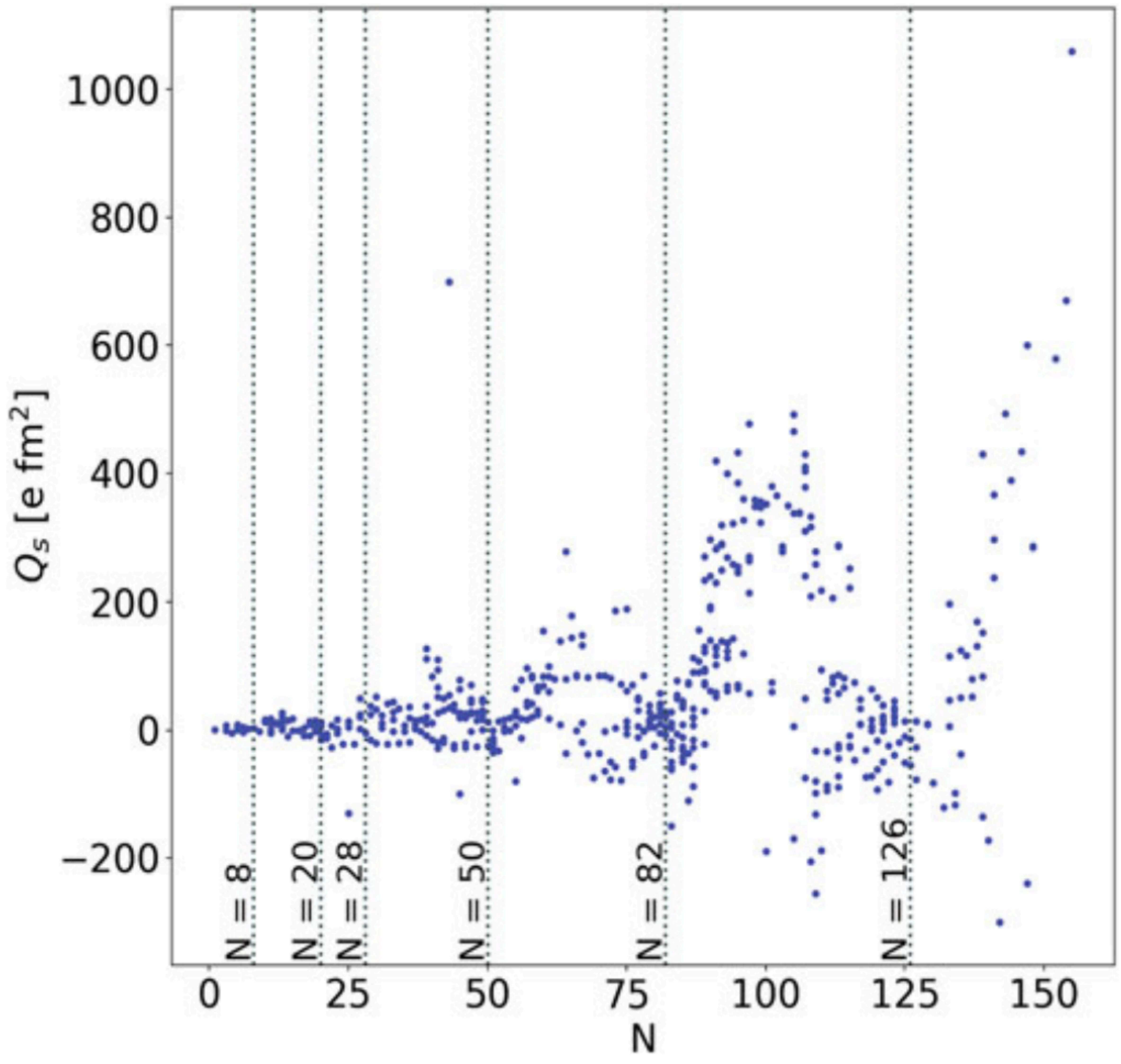
No reference value for $Q_S$




**Compiled‡ magnetic dipole and electric quadrupole moments** from published articles

| Isotope | Energy [keV] | $t_{1/2}$ | Spin/Parity | μ [nm] | Q [b] | Ref. Std | Method | NSR keyword | doi |
|---|---|---|---|---|---|---|---|---|---|
| $^{131}$Ce | 162 | 88 ns | 9/2$^-$ | -0.85(3) | | | TDPAD | 1998IO01 | 10.1016/S0375-9474(98)00157-2 |
| | | | | | 0.92(10) | [$^{138}$Ce 3538.] | TDPAD | 1998IO01 | 10.1016/S0375-9474(98)00157-2 |

# Not just a reference for <u>individual</u> values




Database to <u>explore</u> general trends

and <u>globally</u> benchmark / tune latest theories

Great that it is an electronic database!

de Groote and Neyens, Spins and Electromagnetic Moments of Nuclei,
in Handbook of Nuclear Physics, ed. Tanihata (2022)

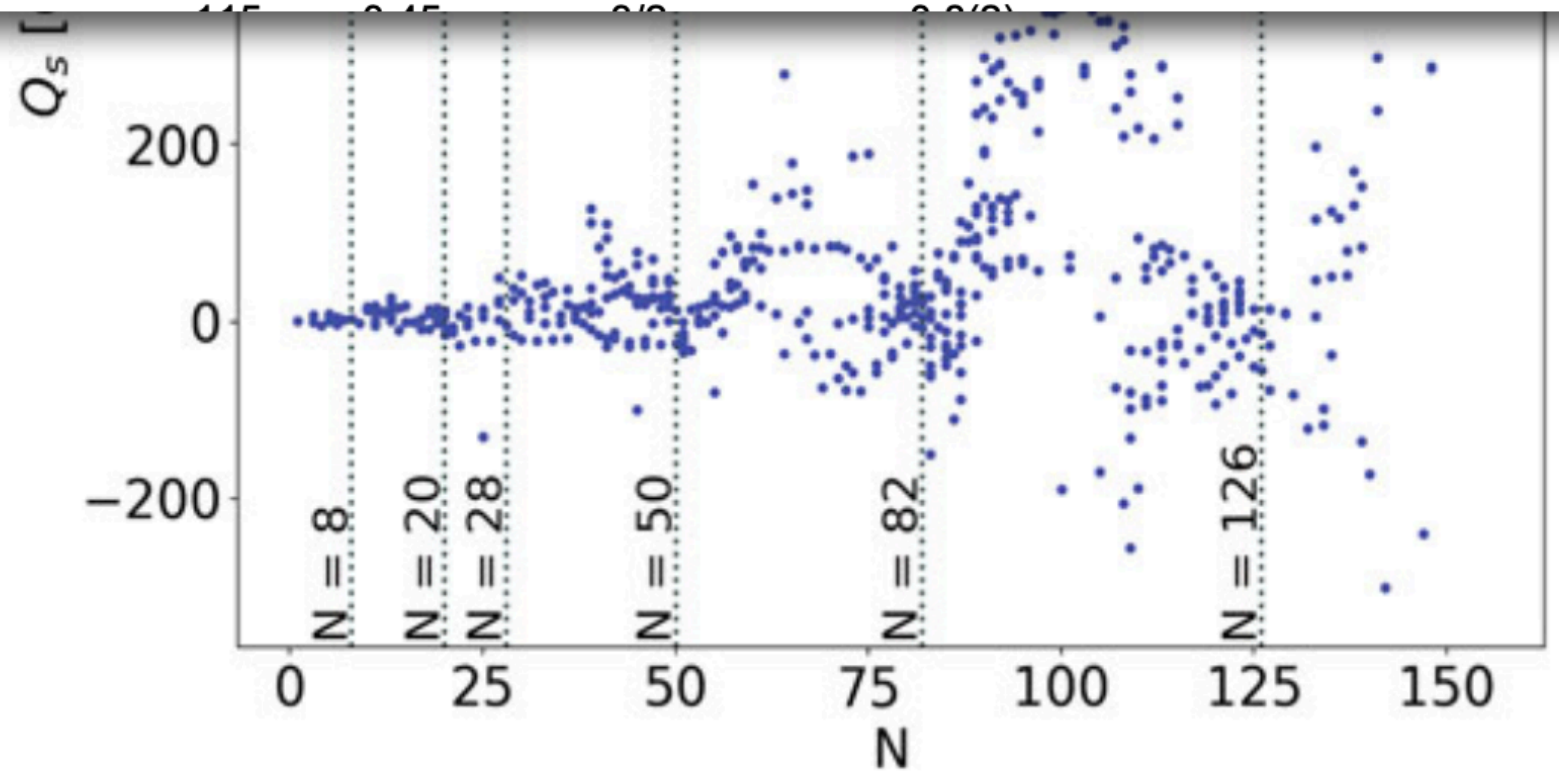
# Not just a reference for <u>individual</u> values

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| 30 Zn 65 | 0 | 244.1 d | 5/2- | +0.7690(2)[l] | | [$^{67}$Zn] | OD | PR 134 A47(64) |
| | | | | | −0.023(2)[f,u] | [$^{67}$Zn] | OD | PR 134 A47(64) |
| | | | | | −0.03(2)[u] | [$^{67}$Zn] | NO/S,R | HFI 22 19(85) |

oments.pdf
ge 35 of 148

Search

| | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| 30 Zn 65 | 0 | 244.1 d | 5/2- | +0.7690(2) | | [67Zn] | OD | 1964By01 | PR 134 A47 (64) |
| | | | | | -0.023(2) | [67Zn] | OD | 1964By01 | PR 134 A47 (64) |
| | | | | | -0.3(2) | [67Zn] | NO/S, R | 1985Ha41 | HFI 22 19 (85) |




Great that it is an electronic database!

Need help from the NP community to keep correct and up to date.

de Groote and Neyens, Spins and Electromagnetic Moments of Nuclei, in Handbook of Nuclear Physics, ed. Tanihata (2022)

# Summary

- Laser spectroscopy is quite unique in being able to *measure* nuclear spins.
  (But if the spins remain tentative, the moments are only correct for the correct spin)

- If an additional structure is observed there must exist another state.
  (a “long-lived” state - but these are the most elusive to decay spectroscopists)

- Nuclear moments are a sensitive probe of the wave function configuration.

- Laser Spectroscopy gives a measure of quadrupole deformation via:-
  - The quadrupole moment
  - The mean-square charge radius

- These complementary (and model-independent) probes are an essential benchmark and guide of nuclear theory (ab-initio, shell model, DFT…)

- Recent measurements on Y, Nb, Mo, Rh, Tm, Fe, Co, Pd, Sc, Ca, Cr; Pb, Te, Tl to be published