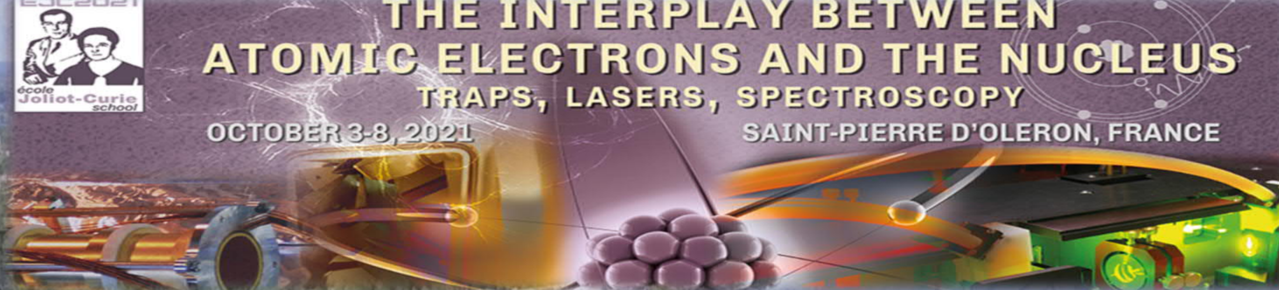

Laser spectroscopy for nuclear structure physics

Iain Moore (iain.d.moore@jyu.fi)
Department of Physics,
University of Jyväskylä, Finland

General remarks

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- Interrupt me any time to ask questions or clarifications. If I cannot answer, I will pass the question onto Ruben….☺
- Hopefully this will be relaxed...after all, I will use Comic Sans font instead of Times New Roman!
- A few topics I will discuss have already been presented. At a school it's good to hear things several times in different flavors:

  ``Repetitio est mater studiorum´´

  ``Practice makes perfect´´…

Additional reading for enthusiasts

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LASER SPECTROSCOPY

- W. Demtröder, Laser Spectroscopy Basic Concepts and Instrumentation, 3rd Edition Springer-Verlag (2003)

RESONANCE IONIZATION

- V.S. Letokhov, Laser Photoionization Spectroscopy, Ac. Press, (1987)

ISOTOPE SHIFTS IN ATOMIC SPECTRA

- W.H. King, Isotope shifts in atomic spectra, Plenum Press, (1984)

Review of field of laser spectroscopy for radioactive nuclei

- P. Campbell, I.D. Moore, M. Pearson, Prog. in Part. and Nucl. Phys. 86 (2016) 127

Electromagnetic moments for nuclear structure research

- G. Neyens, Rep. Prog. Phys. 66 (2003) 633

Recent progress in laser spectroscopy of the actinides

- M. Block, M. Laatiaoui, S. Raeder, Prog. in Part. and Nucl. Phys. 116 (2021) 103834

Future review paper to appear we hope in 2022 (Ruben and myself)?!?

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Outline of my two lectures

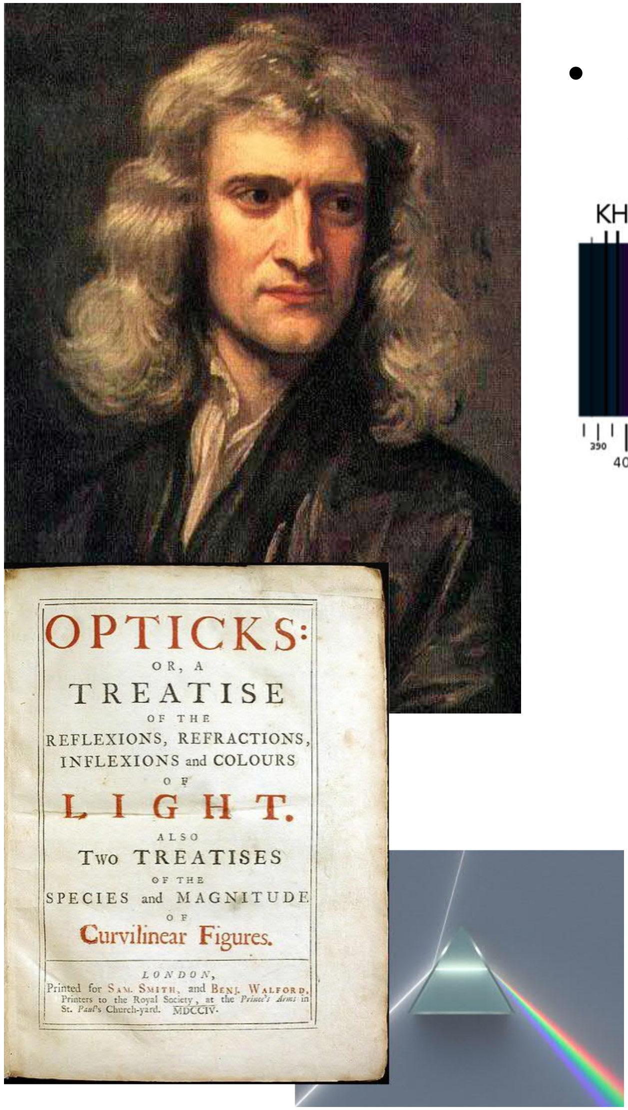
<u>Lecture 1:</u>

– A little history
– Nuclear fingerprints on atomic spectra (from a simple ”experimentalists” point of view)
– What can we learn from nuclear shapes and charge radii?

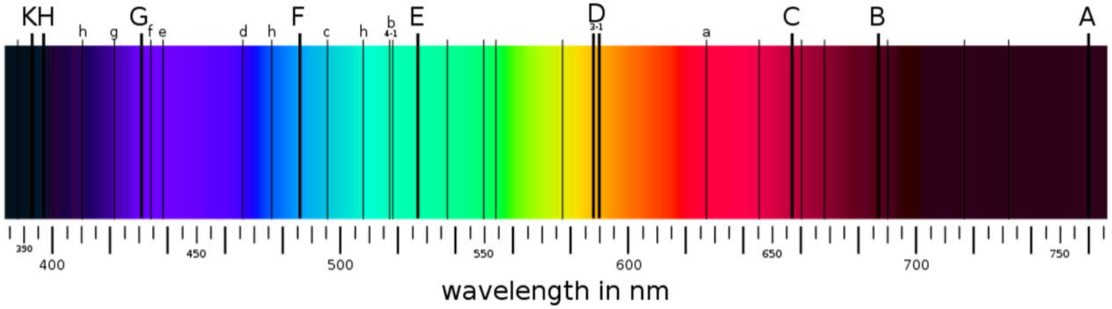
<u>Lecture 2:</u>

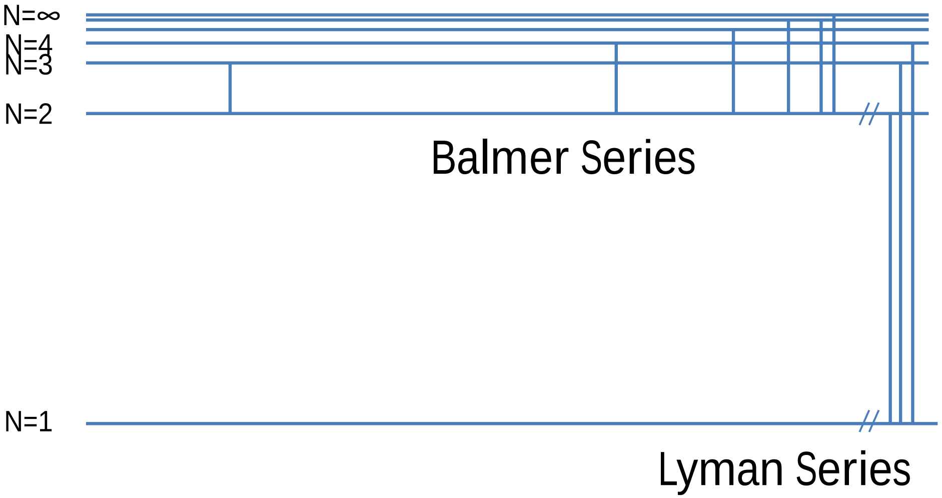
– A short introduction to radioactive ion beam production
– Laser resonance ionization
– Optical spectroscopy and the ”achilles tendon”
– Doppler-free approaches

A historical note on atomic spectroscopy


OPTICKS:
OR, A
TREATISE
OF THE
REFLEXIONS, REFRACTIONS,
INFLEXIONS and COLOURS
OF
LIGHT.
ALSO
Two TREATISES
OF THE
SPECIES and MAGNITUDE
OF
Curvilinear Figures.
LONDON,
Printed for SAM. SMITH, and BENJ. WALFORD,
Printers to the Royal Society, at the Prince's Arms in
St. Paul's Church-yard. MDCCIV.

- 1704: release of Newton's "Opticks".
  Sun's light can be dispersed into a "spectrum"

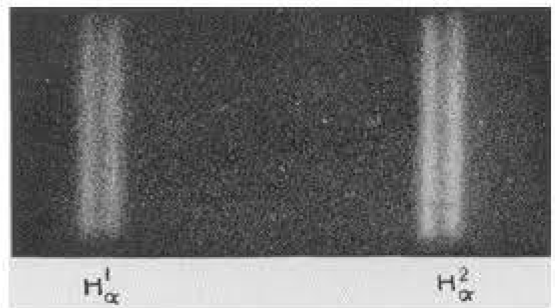


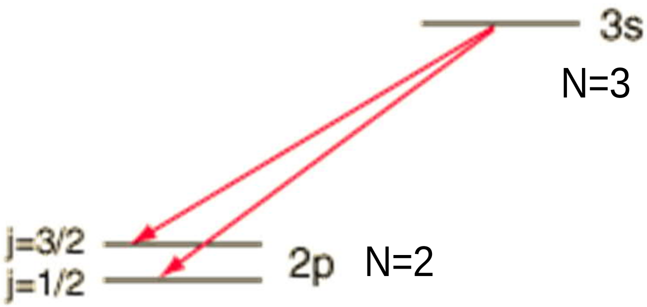


Fine structure

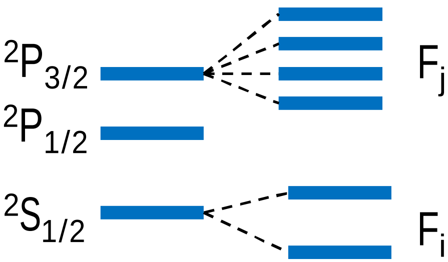
- Increasing the resolution by a factor of ~5000 reveals a fine structure splitting of hydrogen

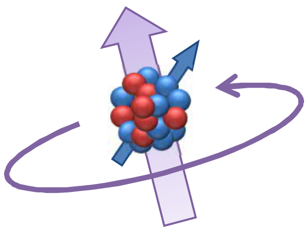
λ=656.279 nm (N=3 → N=2 in Balmer series)





- A further factor of 1000 in resolution reveals a finer splitting due to the coupling of the nucleus with the electronic orbital





F=J+I

→ Hyperfine structure (μeV perturbations)

Magnetic description
- Pauli 1924

Electric (quadrupole)
- Schuler & Schmidt 1934

Hyperfine interactions (in free atoms)

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Hyperfine interaction = the interaction of nuclear magnetic and electric moments with electromagnetic fields (which are produced at the nucleus by the orbiting electrons)

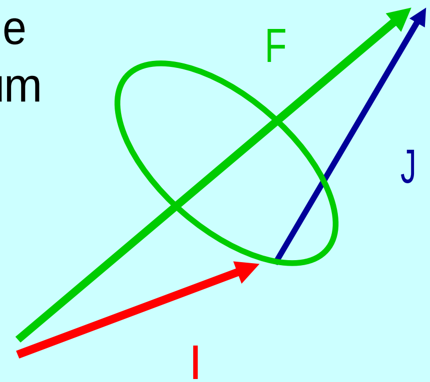
Lets consider the effect on an atomic orbit of spin J

The atomic and nuclear spins couple to form the total angular momentum

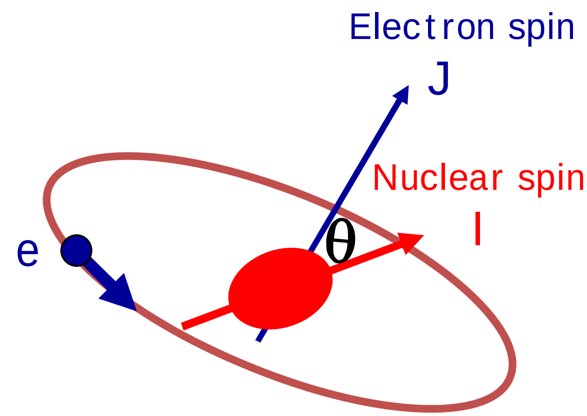
F = I + J

Each state J has several F-states:

$$\vec{F} = \vec{I} + \vec{J}$$

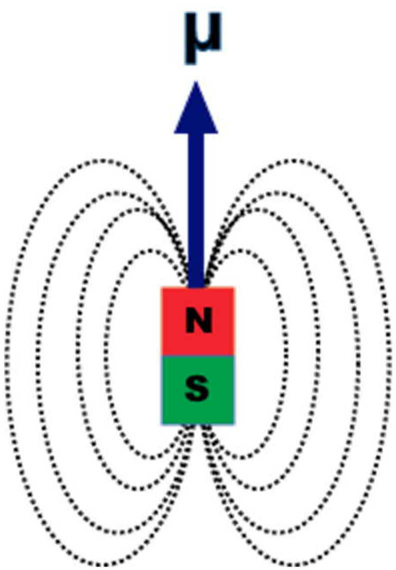


$$|I - J| \leq F \leq I + J$$



States of the same I and J but coupled to different angular momenta F have slightly different energies

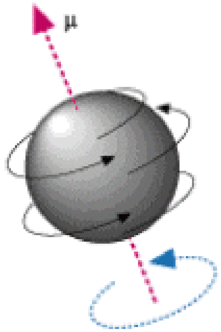
Nuclear magnetic dipole moment



$$\hat{\mu}_I = \sum_i (g_l^i \mathbf{l_i} + g_s^i \mathbf{s_i}) = g\mathbf{I}\mu_N$$

Contributions from orbiting charge and intrinsic spin

Protons:	$g_l = +1$	$g_s = +5.586$
Neutrons:	$g_l = 0$	$g_s = -3.826$



The magnetic dipole moment of a state of spin I = expectation value of the z-component of the dipole operator μ :

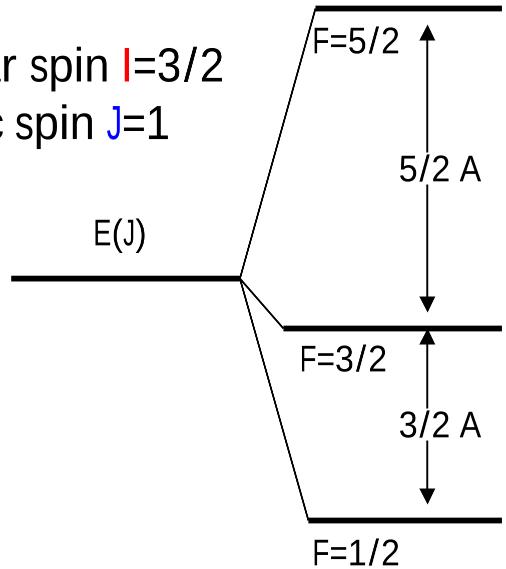
$$\mu(I) \equiv \left\langle I, m = I \middle| \hat{\mu}_z \middle| I, m = I \right\rangle = gI\mu_N$$

The magnetic moment (or g factor) therefore tells us about the valence nucleon orbits and couplings (tests of Shell Model).

The magnetic dipole interaction

^{201}Hg

Nuclear spin I=3/2
Atomic spin J=1



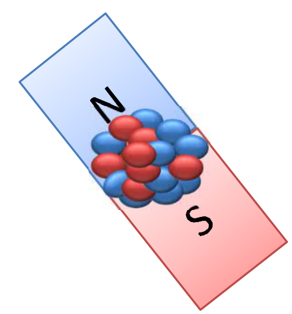
The interaction energy depends on angle θ

$$E = -\boldsymbol{\mu} \cdot \mathbf{B_e} = -\mu B_e \cos\theta$$

$$A = \frac{\mu_I B_e(0)}{I \cdot J},$$

$B_e(0)$ = magnetic field at nucleus

Access to nuclear spin I (number of hyperfine components) and μ_I



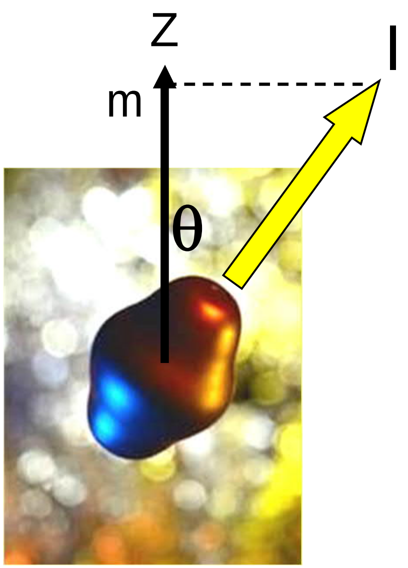
The original fine structure level E(J) is perturbed so that the final energy due to the magnetic hyperfine effect:

$$E(F) = \frac{A}{2} C$$

where $C = F(F+1) - I(I+1) - J(J+1)$

The electric quadrupole moment

The electric quadrupole moment provides a measure of the deviation of charge distribution from sphericity:



$$eQ = \int_0^\infty \rho_n(\mathbf{r})(3z^2 - r^2)\,\mathrm{d}\tau$$

Experiments measure the maximum "projection" of the intrinsic quadrupole moment along the quantization axis

Using angular momentum algebra, we get

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

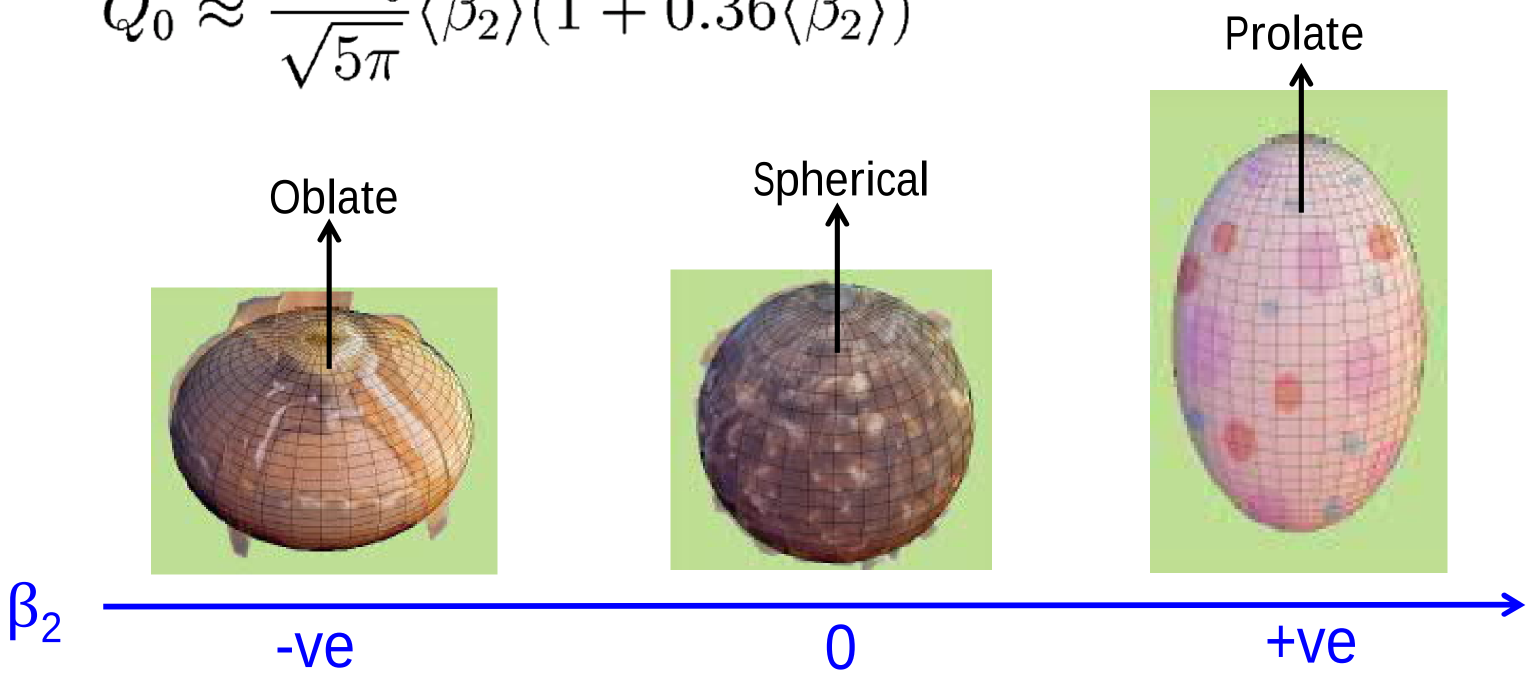
this assumes a well-defined deformation axis (not always a good approximation)

Note for nuclear spin I=0 and I=1/2 the spectroscopic quadrupole moment vanishes even if the intrinsic shape is deformed.

Quadrupole deformation

The intrinsic moment can in turn be related to the quadrupole deformation parameter β_2

$$Q_0 \approx \frac{3Zr_0^2}{\sqrt{5\pi}}\langle\beta_2\rangle(1+0.36\langle\beta_2\rangle)$$



How common is quadrupole deformation?

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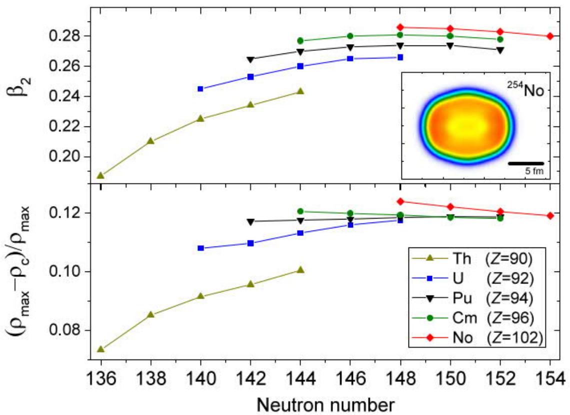
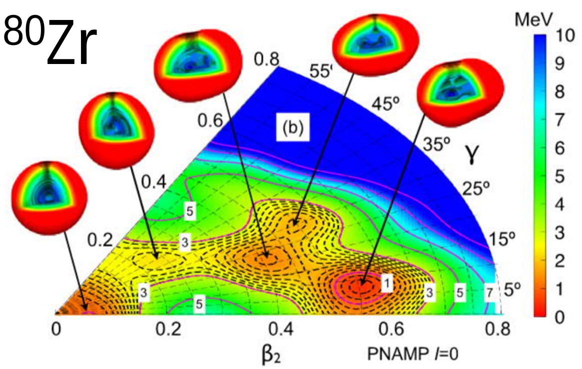
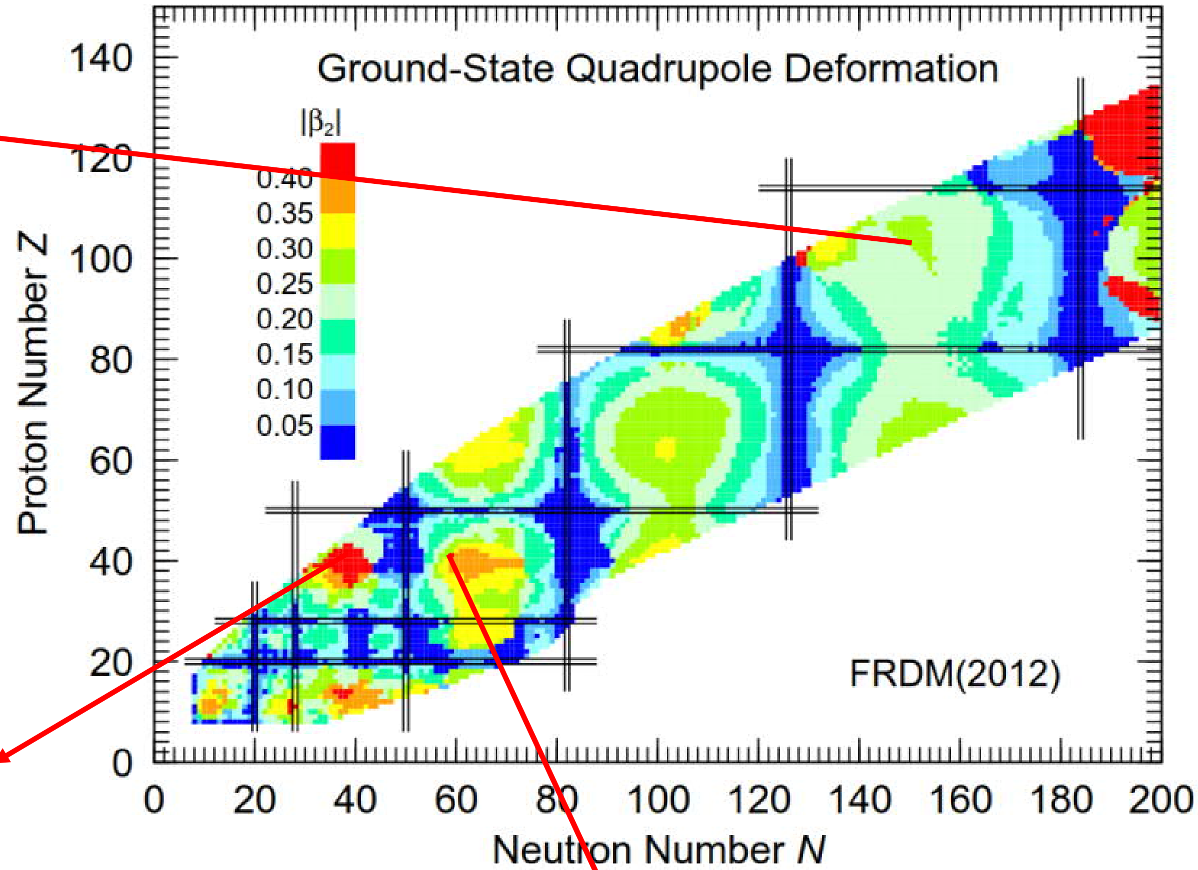
One might even ask how "uncommon" spherical nuclei are ☺?



S. Raeder et al., PRL 120 (2018) 232503



Rodriguez and Egido, PLB 705 (2011) 255



See later ☺

The electric quadrupole interaction

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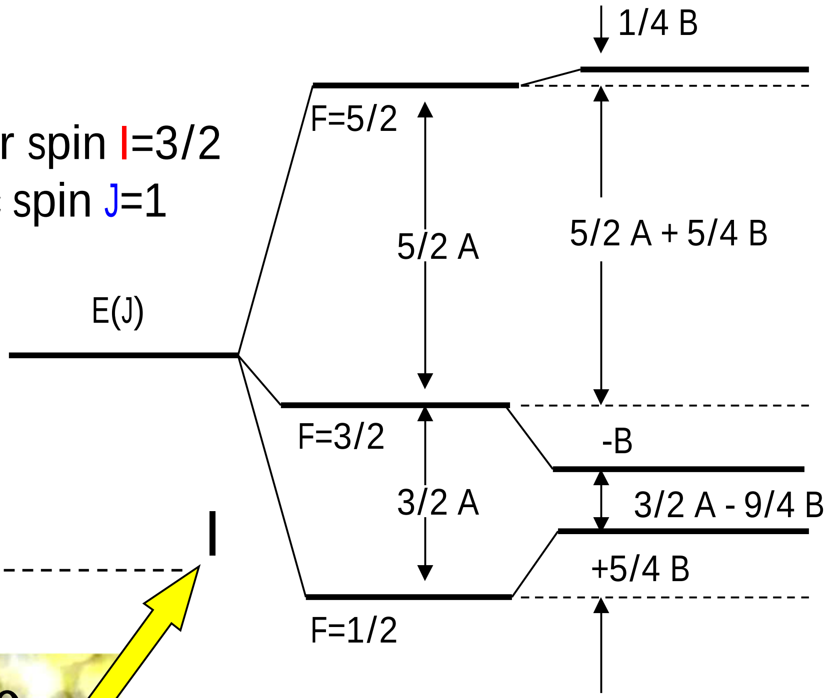
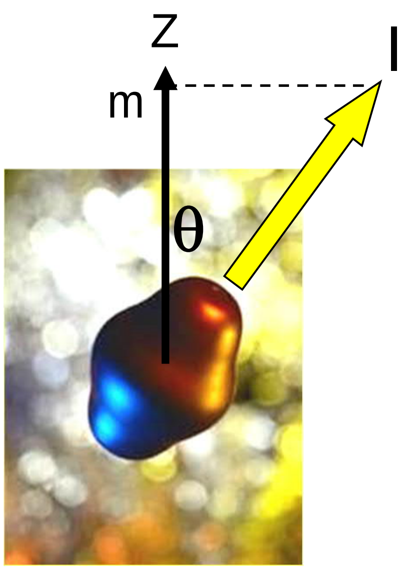
^{201}Hg

Nuclear spin I=3/2
Atomic spin J=1





$$A = \frac{\mu_I B_e(0)}{I \cdot J},$$

$B_e(0) =$ magnetic field at nucleus

Access to nuclear spin I (number of HF components) and μ_I

$$B = eQ_s V_{JJ}(0),$$

$V_{JJ}(0) =$ electric field gradient at nucleus

Access to Q_s

$$E(F) = \frac{A}{2}C + B\,\frac{\frac{3}{4}C(C+1) - I(I+1)J(J+1)}{2(2I-1)(2J-1)I \cdot J} \quad +$$

where $\quad C = F(F+1) - I(I+1) - J(J+1)$

Eye-watering higher orders……

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PHYSICAL REVIEW A **103**, 032826 (2021)

Magnetic octupole moment of ^{173}Yb using collinear laser spectroscopy

R. P. de Groote,[1,*] S. Kujanpää,[1] Á. Koszorús,[2] J. G. Li,[3] and I. D. Moore[1]
[1]*Department of Physics, University of Jyväskylä, PB 35(YFL) FIN-40351 Jyväskylä, Finland*
[2]*Department of Physics, University of Liverpool, Liverpool L69 7ZE, United Kingdom*
[3]*Institute of Applied Physics and Computational Mathematics, Beijing 100088, China*

Defining $K = F(F+1) - I(I+1) - J(J+1)$, this can be written as (truncated at the octupole ($k = 3$) term):

$$E_F^{(1)} = \frac{AK}{2} + \frac{3B}{4}\frac{K(K+1) - I(I+1)J(J+1)}{(2I(2I-1)J(2J-1))} + \frac{5C}{4}\frac{K^3 + 4K^2 + \frac{4}{5}K(-3I(I+1)J(J+1) + I(I+1) + J(J+1) + 3) - 4I(I+1)J(J+1)}{I(I-1)(2I-1)J(J-1)(2J-1))},$$

with hyperfine constants

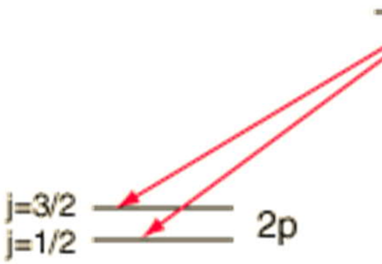
$$A = \tfrac{1}{IJ}\langle II|T_2^{(n)}|II\rangle\langle JJ|T_1^{(e)}|JJ\rangle = \frac{\mu_I}{IJ}\langle JJ|T_1^{(e)}|JJ\rangle,$$
$$B = 4\langle II|T_2^{(n)}|II\rangle\langle JJ|T_2^{(e)}|JJ\rangle = 2eQ\langle JJ|T_2^{(e)}|JJ\rangle,$$
$$C = \langle II|T_3^{(n)}|II\rangle\langle JJ|T_3^{(e)}|JJ\rangle = -\Omega\langle JJ|T_3^{(e)}|JJ\rangle.$$

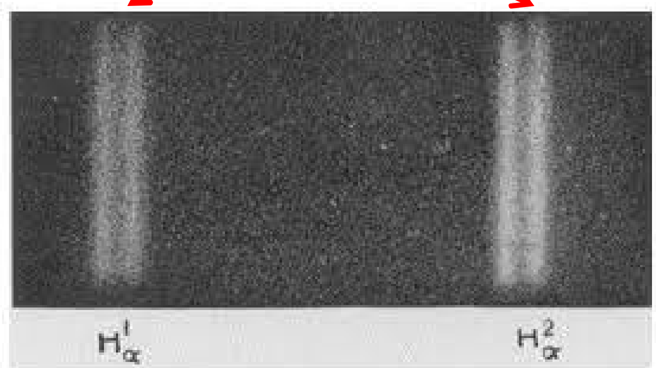
Measurements of the magnetic octupole constant C and moment Ω are scarce!


Brief pause, breathe, enjoy scenary.....

Let's return to the Balmer series

λ=656.279 nm (N=3 → N=2 in Balmer series)





Hydrogen   Deuterium

H. Urey (1932)

Neutron discovered (1932)

Chemistry Nobel Prize 1934 ("heavy" hydrogen)

H. Urey et al,. Phys. Rev. 39 (1932) 164

The isotope shift is the frequency difference in an electronic transition between two isotopes of mass A and A´

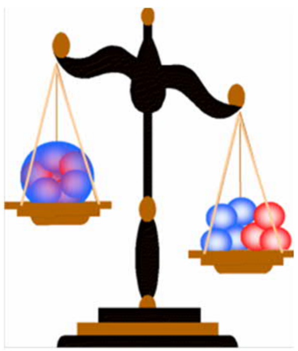
$$\delta\nu^{AA'} = \nu^{A'} - \nu^{A}$$

Isotopic shifts of electronic transitions

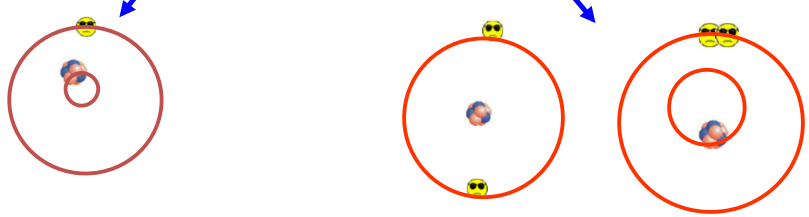
The shift in the atomic transition frequency between different isotopes of the same element arises due to changes in nuclear mass and size.

$$\delta\nu_{IS} = \delta\nu_{MS} + \delta\nu_{FS}$$

<u>Nuclear mass</u>



$$\delta\nu_{MS} = \delta\nu_{NMS} + \delta\nu_{SMS} = \left(\frac{A' - A}{AA'}\right)(N + S)$$



- Techniques of measuring the mass were discussed by Matthias!
- Adriana went into more detail regarding these two contributions to the mass shift

The nuclear volume effect (field shift)

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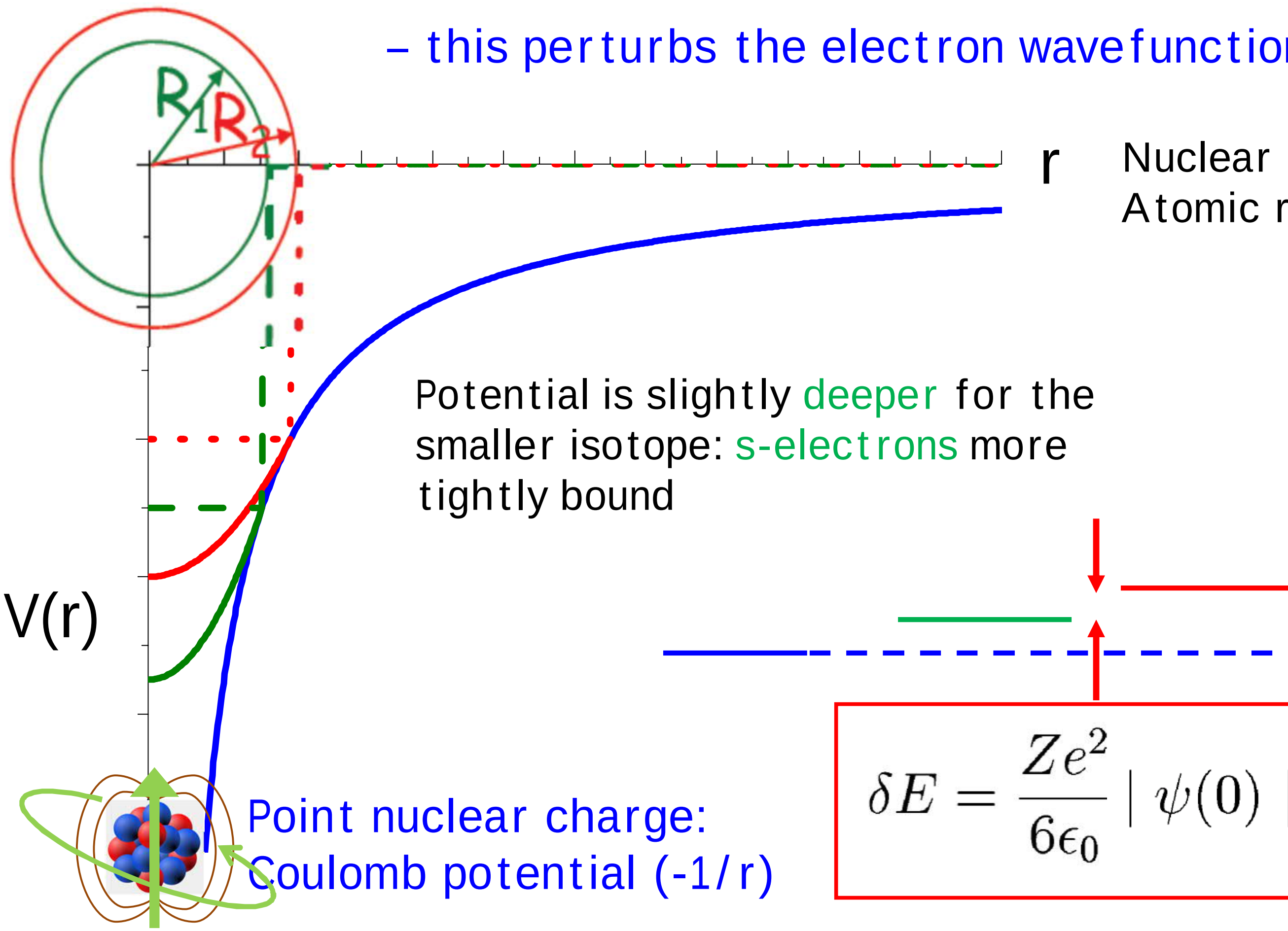
The finite spatial extent (volume) of the nucleus gives an electrostatic potential difference to that of the Coulomb potential

– this perturbs the electron wavefunction $\Psi_e(r)$



Nuclear radius: few × 10^{-15} m
Atomic radius: few × 10^{-10} m

Potential is slightly deeper for the smaller isotope: s-electrons more tightly bound

Isotope shift of atomic level

Point nuclear charge: Coulomb potential (-1/r)

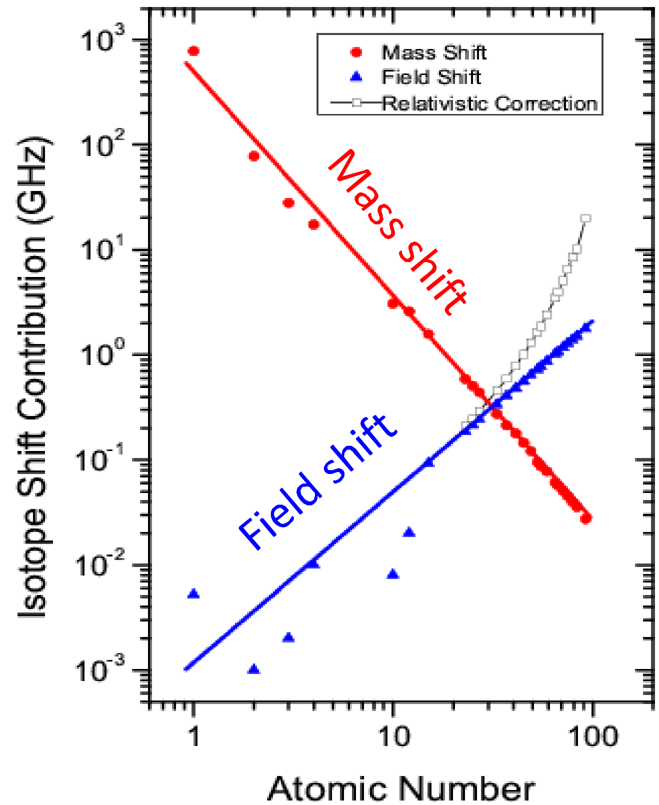
$$\delta E = \frac{Ze^2}{6\epsilon_0} \mid \psi(0) \mid^2 \delta\langle r^2 \rangle^{A,A'}$$

Isotopic shifts of electronic transitions

$$\delta\nu_{IS} = \delta\nu_{MS} + \delta\nu_{FS}$$

$$\delta\nu_{FS} = \frac{Ze^2}{6h\varepsilon_0}\Delta|\psi_e(0)|^2\,\delta\langle r^2\rangle$$

EXPERIMENT | THEORY



To evaluate IS data:

- mass data from Atomic Mass Evaluation (2021)
- SMS either calculated (ab-initio, MBPT, coupled cluster…) or evaluated via non-optical data (elastic e scattering, muonic atom X-rays)
- Field shift factor from non-optical, semi-empirical, atomic theory (accurate to ~10%)

- Anastasia discussed the role of relativistic corrections on the heaviest elements and nicely summarized computational methods!

I.D. Moore, EJC 2021

Using non-optical data to extract atomic factors

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When the mean-square charge radii has already been established between at least 3 isotopes, we can determine atomic factors for an optical transition:

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

We multiply our isotope shift by a modification factor, K, to remove the dependence on the nuclear masses:

$$K^{A,A'} = \frac{AA'}{A - A'} \frac{A_{ref} - A'_{ref}}{A_{ref} A'_{ref}} = \frac{AA'}{A - A'} \xi$$

$$\longrightarrow \quad K^{A,A'} \delta\nu_i^{AA'} = \frac{A_{ref} - A'_{ref}}{A_{ref} A'_{ref}} \times M_i + F_i K^{A,A'} \delta\langle r^2\rangle^{A,A'}$$

y = C + mx

W.H. King, Isotope shifts in atomic spectra, 1984 (Plenum Press)

Example using stable isotopes of Pu (Z=94)

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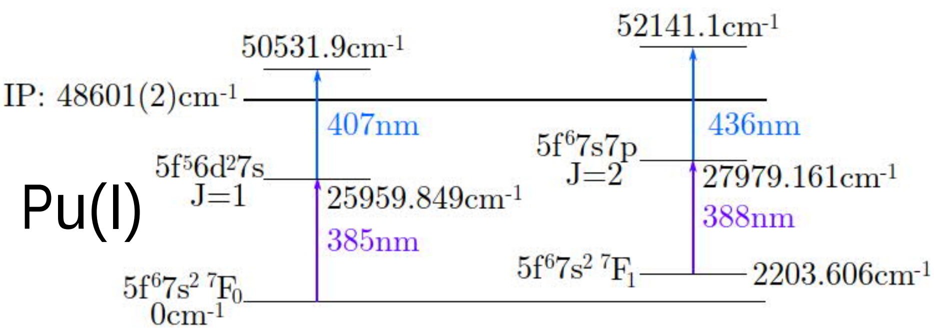
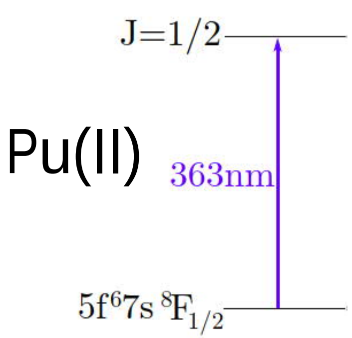
Transition	Lower configuration	Upper configuration	Field shift (GHz/fm^2)
388 nm	$5f^6 7s^2\ ^7F_1$	$5f^6 7s7p$	-22.8(23)
385 nm	$5f^6 7s^2\ ^7F_0$	$5f^5 6d^2\ 7s$	-7.1(7)
363 nm	$5f^6 5s\ ^8F_{1/2}$	J=1/2	+7.9(6)

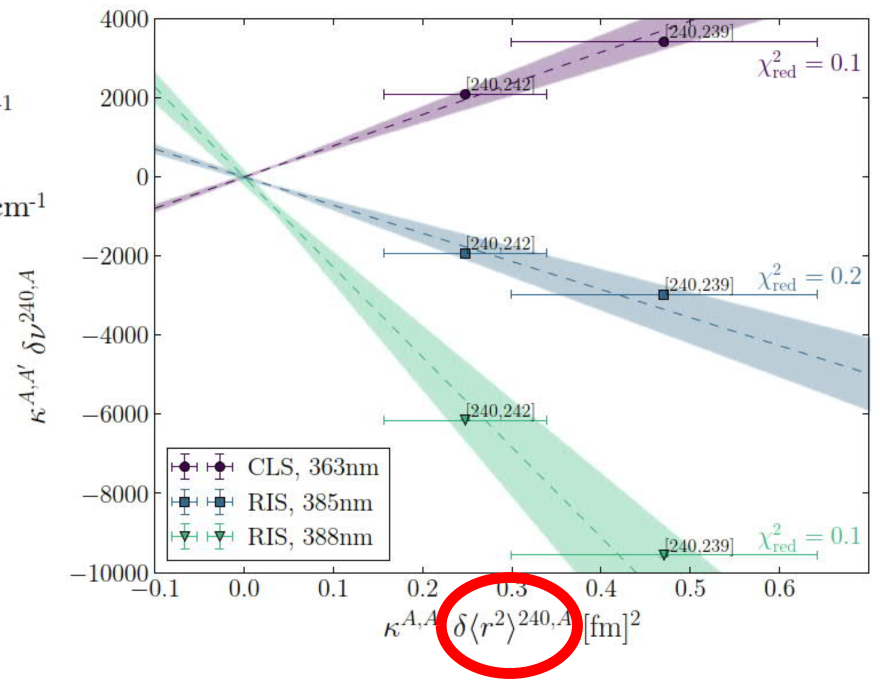
Gradient

The charge radii for all stable isotopes of Pu were measured using muonic atom X-ray spectroscopy affording a calibration of the atomic factors.





A. Voss et al., Phys. Rev. A 95 (2017) 032506

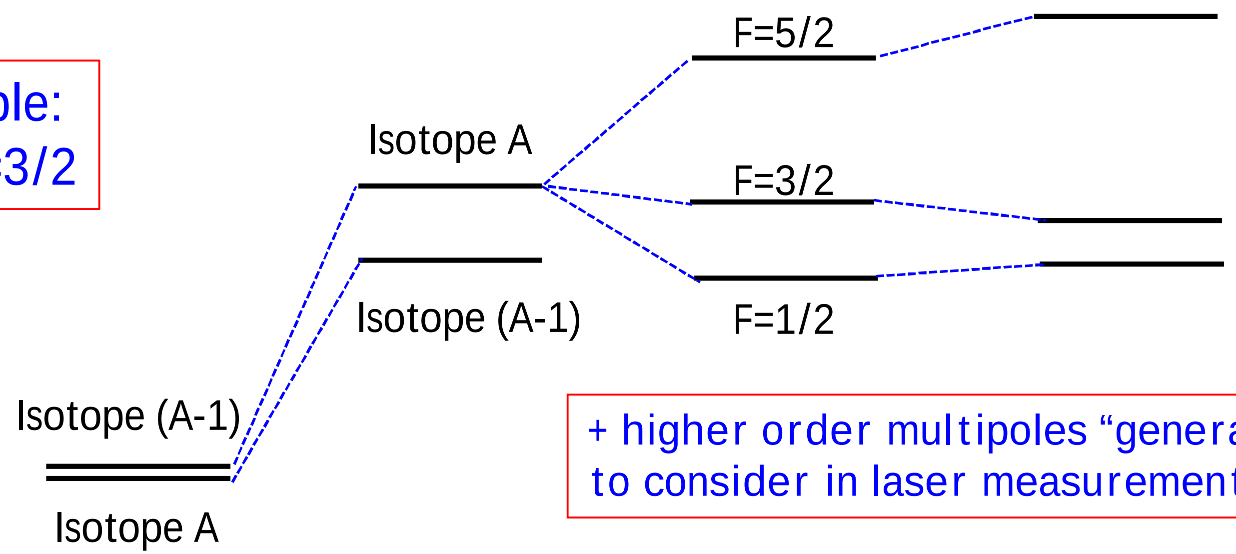


I.D. Moore, EJC 2021

A summary of our nuclear perturbations

Point nucleus + Finite size + Magnetic dipole + Electric quadrupole

Example:
J=1, I=3/2



+ higher order multipoles "generally" too small to consider in laser measurements

$$\text{Mass shift} + \text{Field shift} \quad -\mu B_e \cos\theta \quad + \frac{1}{4} e Q_0 V_{JJ} P_2(\cos\theta)$$

These energy shifts of may be only a few parts per million of the energy of an optical atomic transition. Optical techniques provide the sensitivity and precision required to measure these effects.


ALMA
Keep breathing, more scenary......

What can we learn from the charge radii?

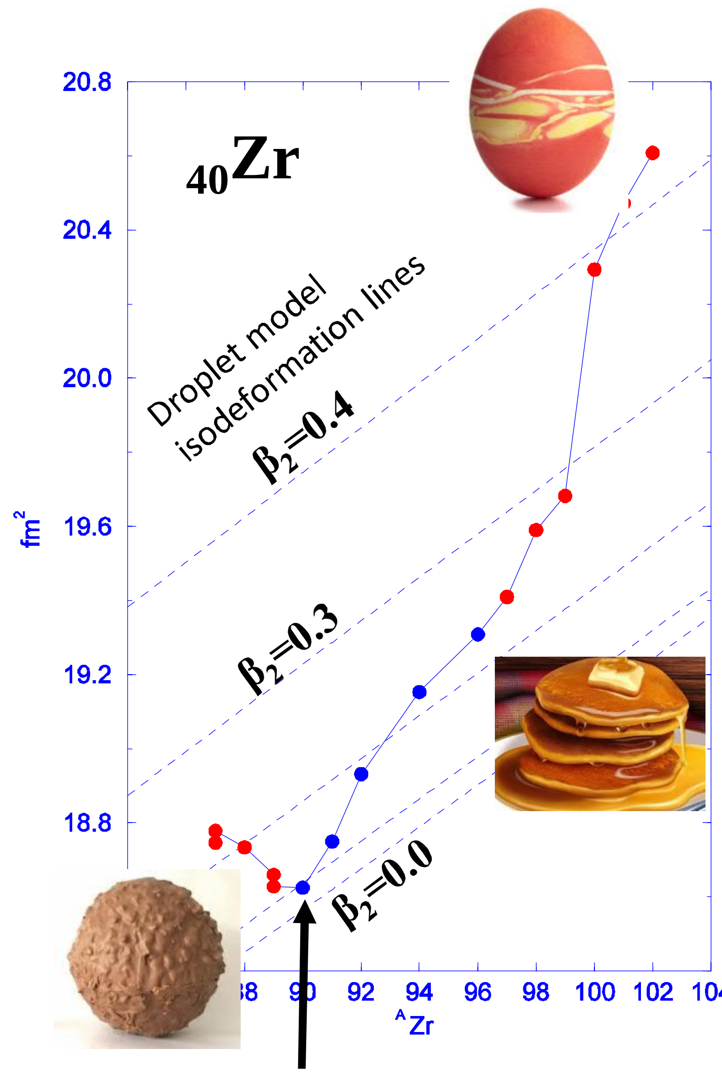
From a simple droplet model approach – we can expand a deformed charge distribution in terms of spherical harmonics.

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

Quadrupole deformation parameter (shape)

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

Radius of spherical nucleus of the same volume



Note: the sign of the deformation cannot be obtained!

N=50 shell closure

We can see trends in the raw data

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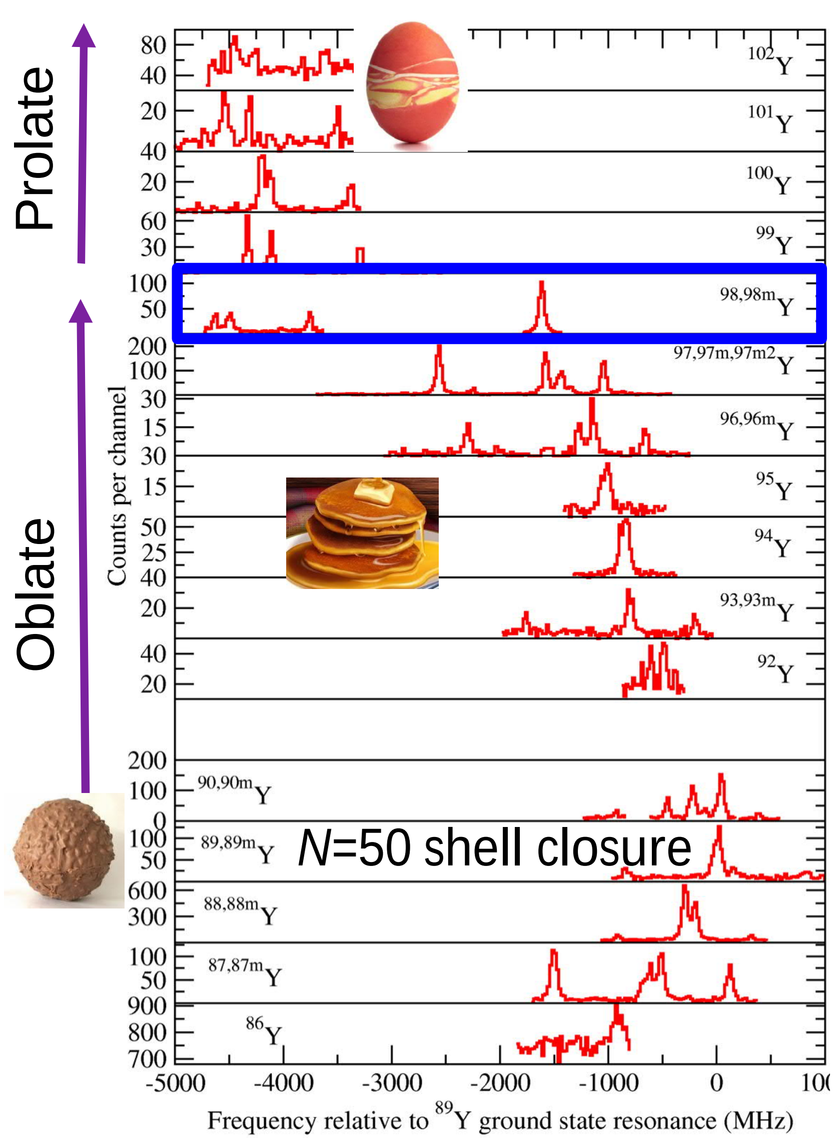
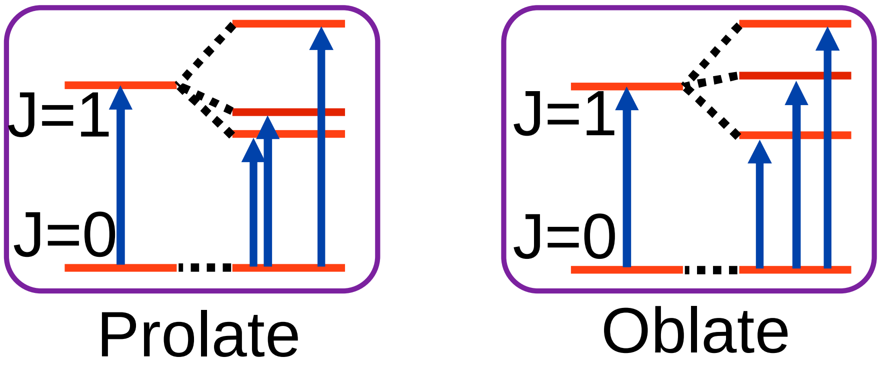


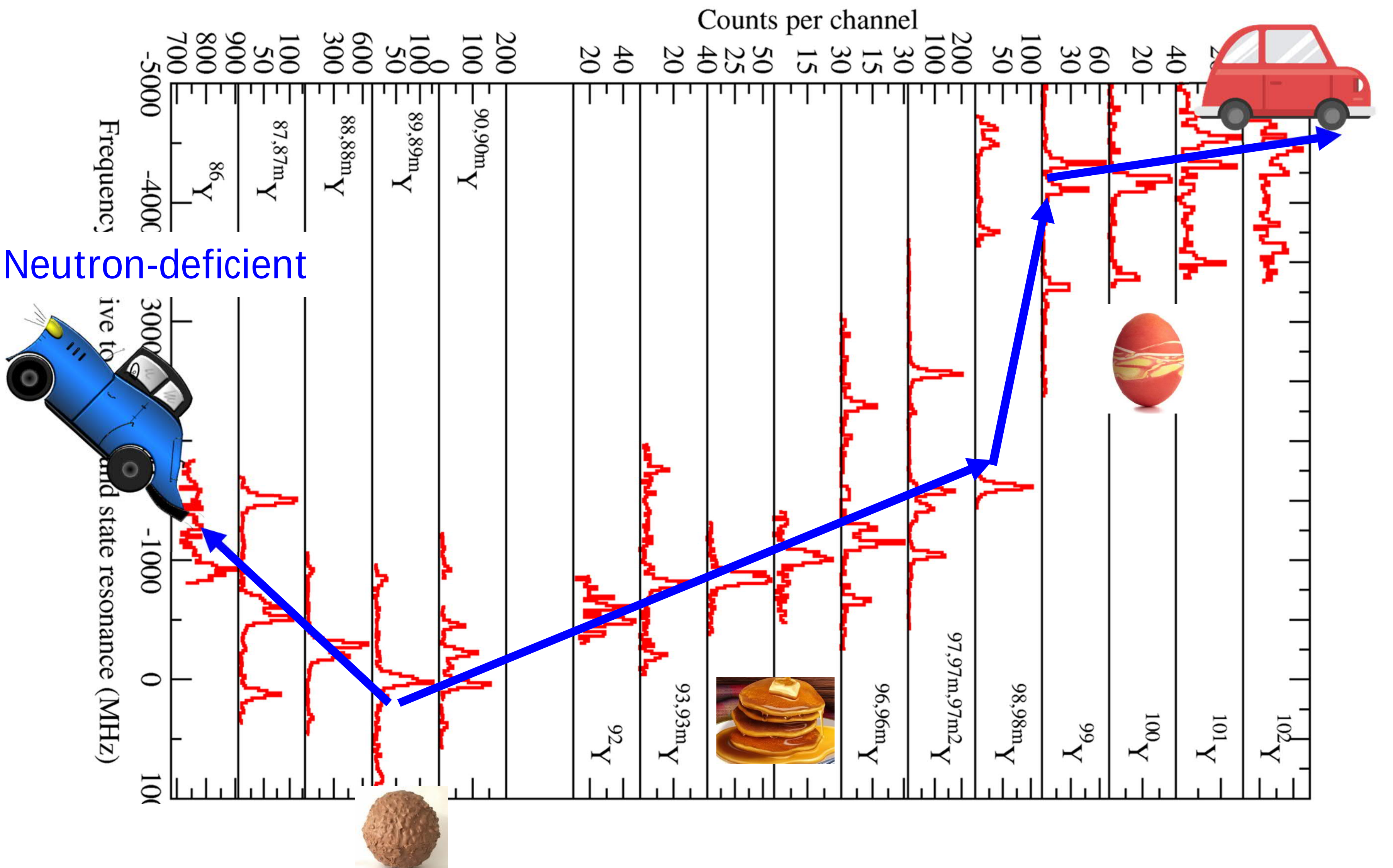


3 peaks maximum for each nuclear state

- Yttrium contains many isomeric (long-lived) nuclear states
- Note that laser spectroscopy can identify new states
- The ^{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 the ”simple way”



Charge radii systematics (Kr to Ru)

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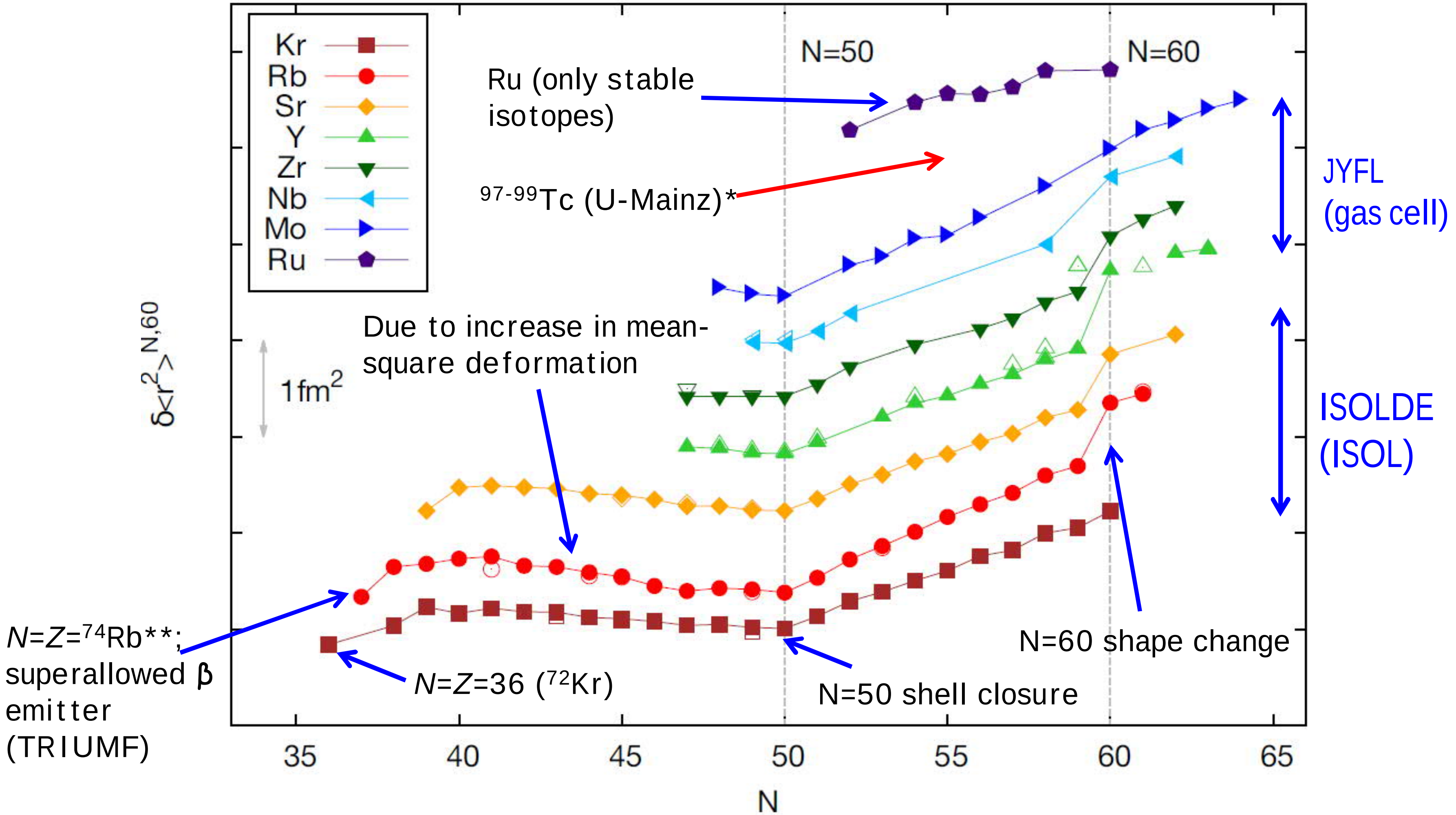
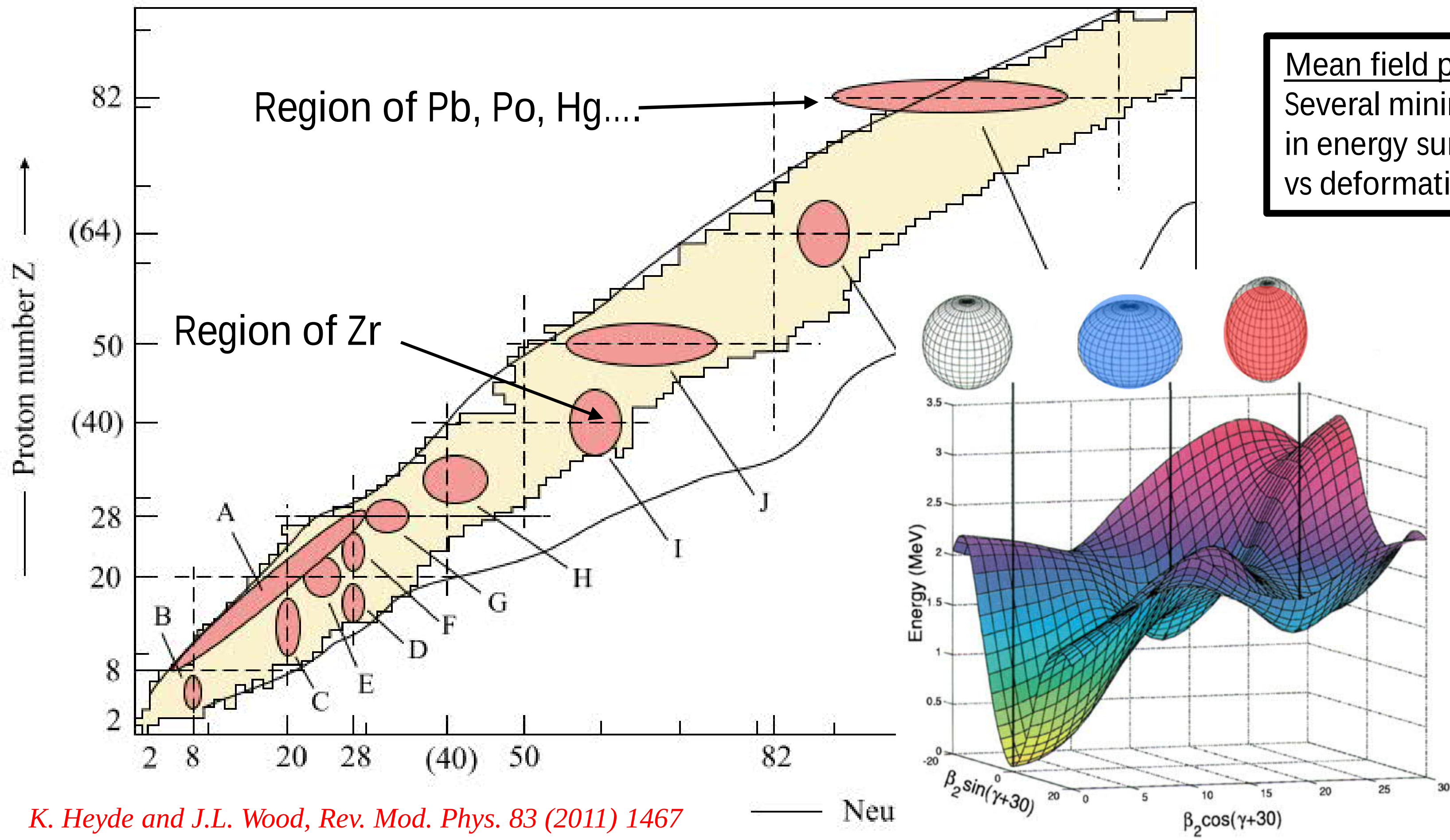


**E. Mane et al., PRL (2011) 212502*

**T. Kron et al., Phys. Rev. C 102 (2020) 034307*

Coexistence of nuclear shapes

- Shape coexistence appears to be unique in the realm of finite many-body quantum systems
- States with different shape/deformation at low energy
- Interplay between stabilizing effect of closed shells and mid-shells for proton-neutron interactions

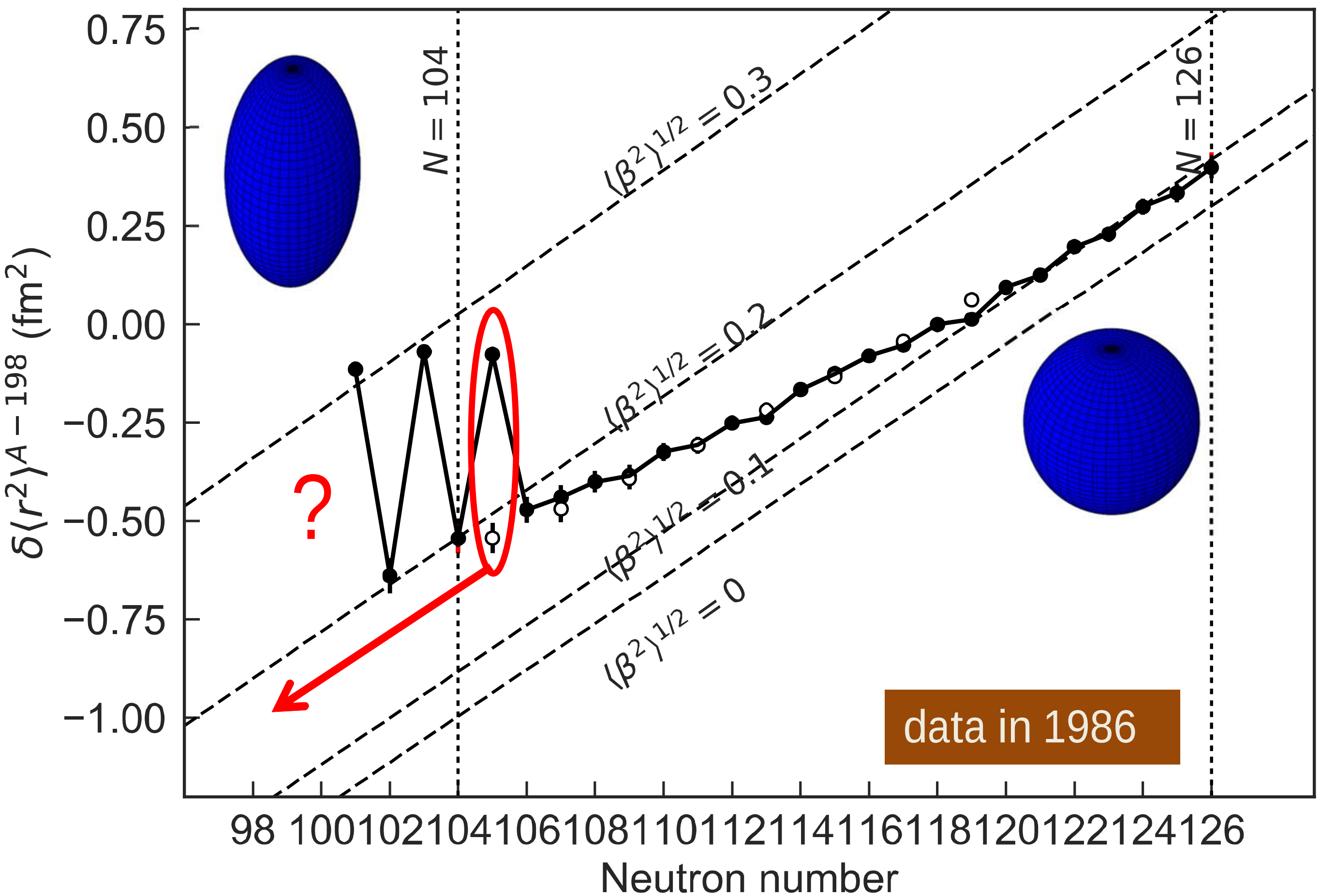


Mean field picture:
Several minima
in energy surface
vs deformation

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

Staggering in the charge radii of Hg isotopes

← *Neutron deficient* *Neutron rich* →



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

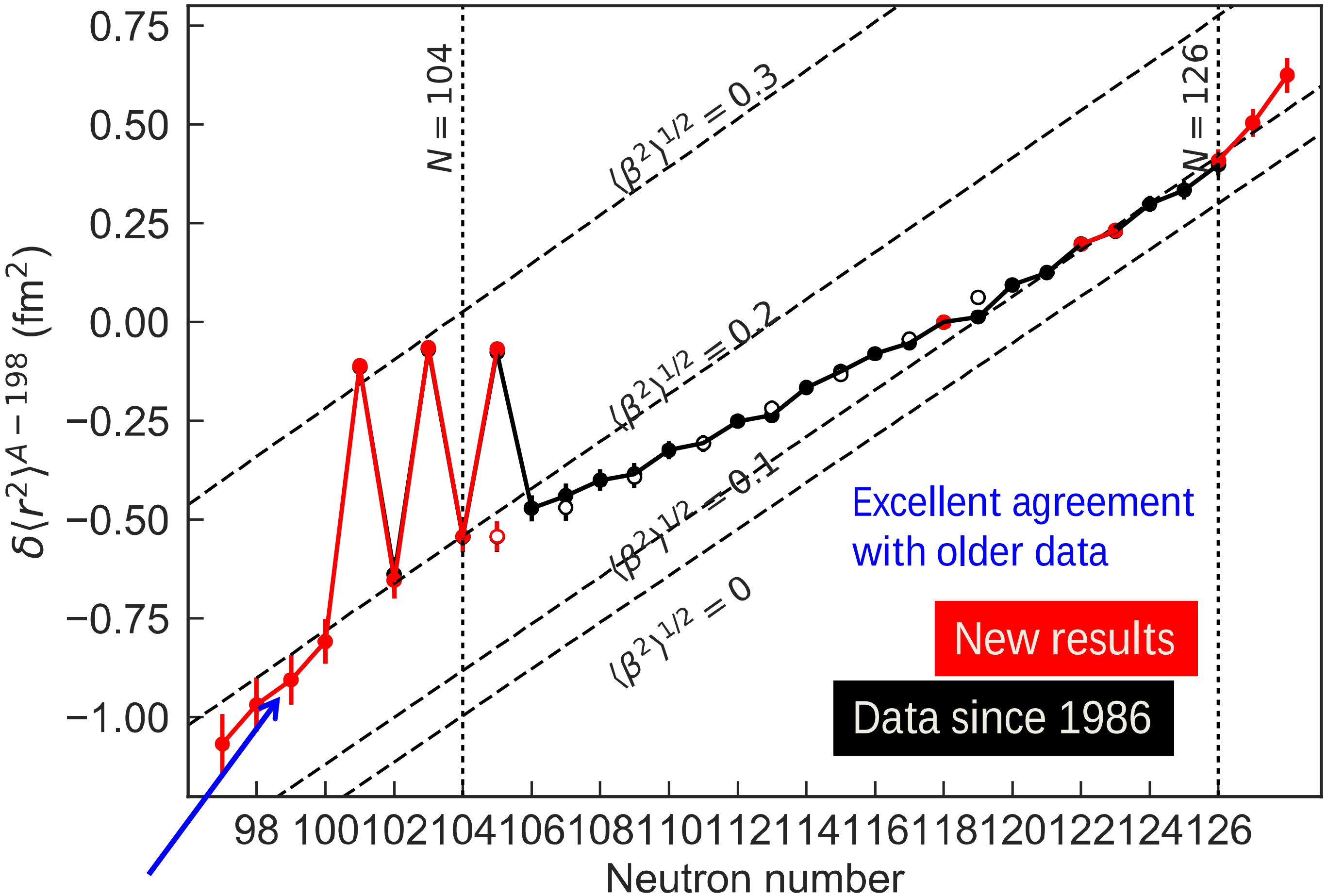
^{181,183,185}Hg

Shape coexistence established in ^{185}Hg!

J. Bonn et al., Phys. Lett. B 38 (1972) 308

G. Ulm et al., Z. Phys. A 325 (1986) 247

After 30 years of developments…

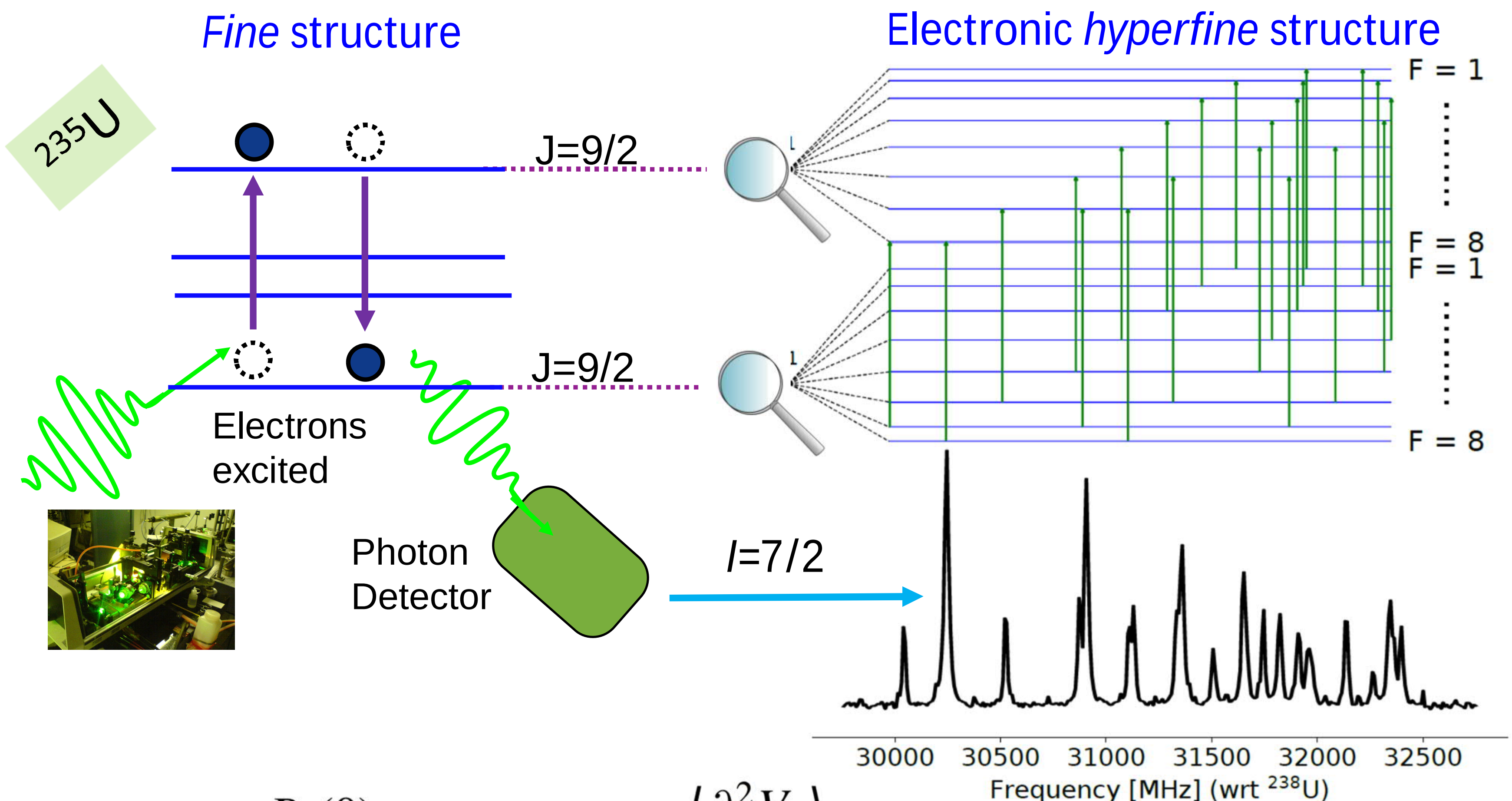


End-point of staggering observed, Hg isotopes return to more spherically-shaped trend.

Rich playground for testing theoretical calculations!

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

Take home message(s) from lecture 1



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

Magnetic dipole interaction

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

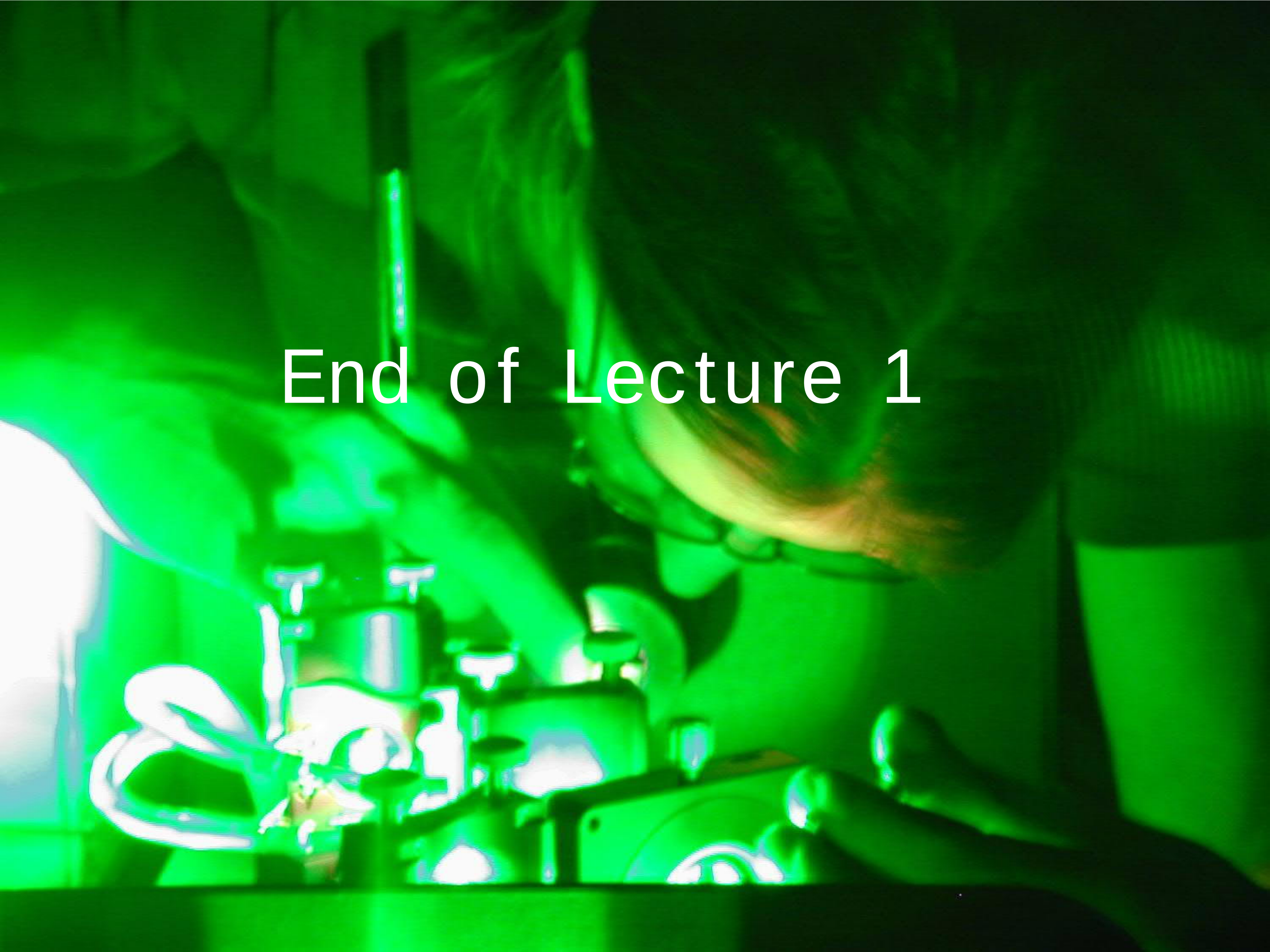
Electric quadrupole interaction

I

Nuclear spin

$\delta\langle r^2\rangle$

Mean-square charge radii

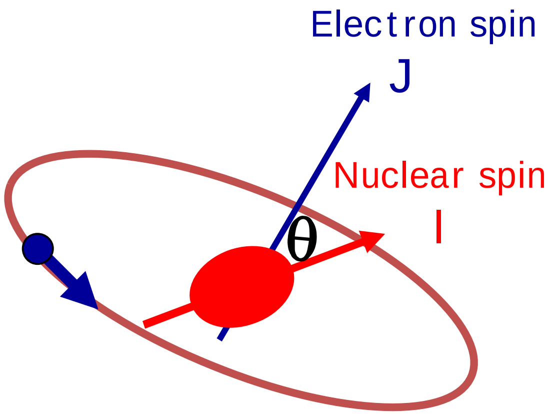

End of Lecture 1

Back up material for lecture 1

Magnetic dipole interaction

The interaction energy depends on angle θ

$$E = -\boldsymbol{\mu} \cdot \mathbf{B_e} = -\mu B_e \cos\theta$$



Since $\boldsymbol{\mu} = g\mathbf{I}\mu_N$ and $\mathbf{B}_e = -(\frac{B_e}{J})\mathbf{J}$

then the interaction Hamiltonian can be expressed as

$$H_m = (\frac{gB_e\mu_N}{J})\mathbf{I.J} = A\mathbf{I.J}$$

The different energy shifts of the different F-states are then

$$\Delta E = \langle IJF \mid H_m \mid IJF \rangle = A\langle \mathbf{I.J} \rangle$$

where

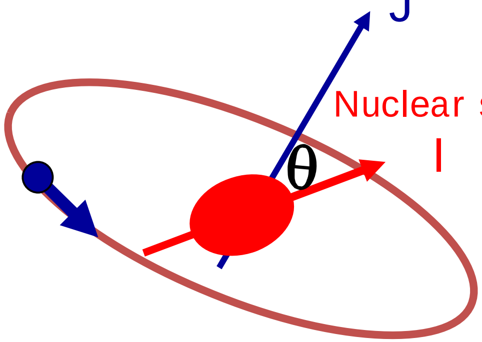
$$\langle \mathbf{I.J} \rangle = \frac{1}{2}\langle F^2 - I^2 - J^2 \rangle = \frac{1}{2}[F(F+1) - I(I+1) - J(J+1)]$$

B_e can be calibrated by measuring the energy shifts for an isotope of a known magnetic moment.

Electric quadrupole interaction

$$E = \tfrac{1}{4}\, e\, Q_0\, V_{JJ}\, P_2(\cos\theta)$$

Electric field gradient along J-direction due to atomic electrons.



Energy shifts of the F-states are then

$$\Delta E_Q = \frac{B}{4} \frac{\frac{3}{2}C(C+1) - 2I(I+1)J(J+1)}{I(2I-1)J(2J-1)}$$

where $C = [F(F+1) - I(I+1) - J(J+1)]$

The hyperfine factor "B" is measured by experiment

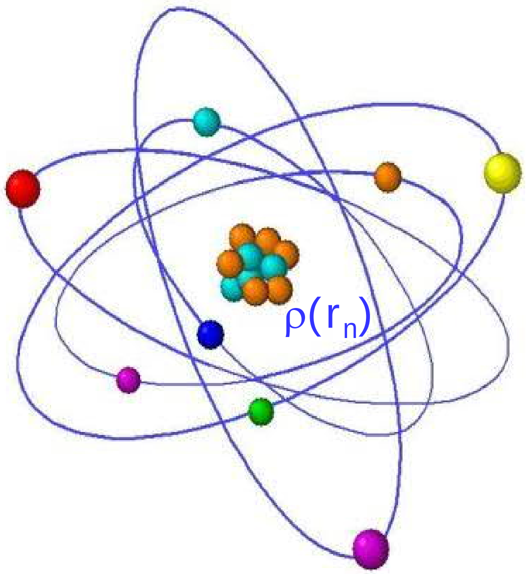
$$B = eQ_s \langle \frac{\partial^2 V}{\partial Z^2} \rangle = eQ_s V_{JJ}$$

The electric field gradient V_{JJ} may be calibrated with an isotope with known Q_s

The nuclear charge distribution

Expanding the charge distribution in multipoles:

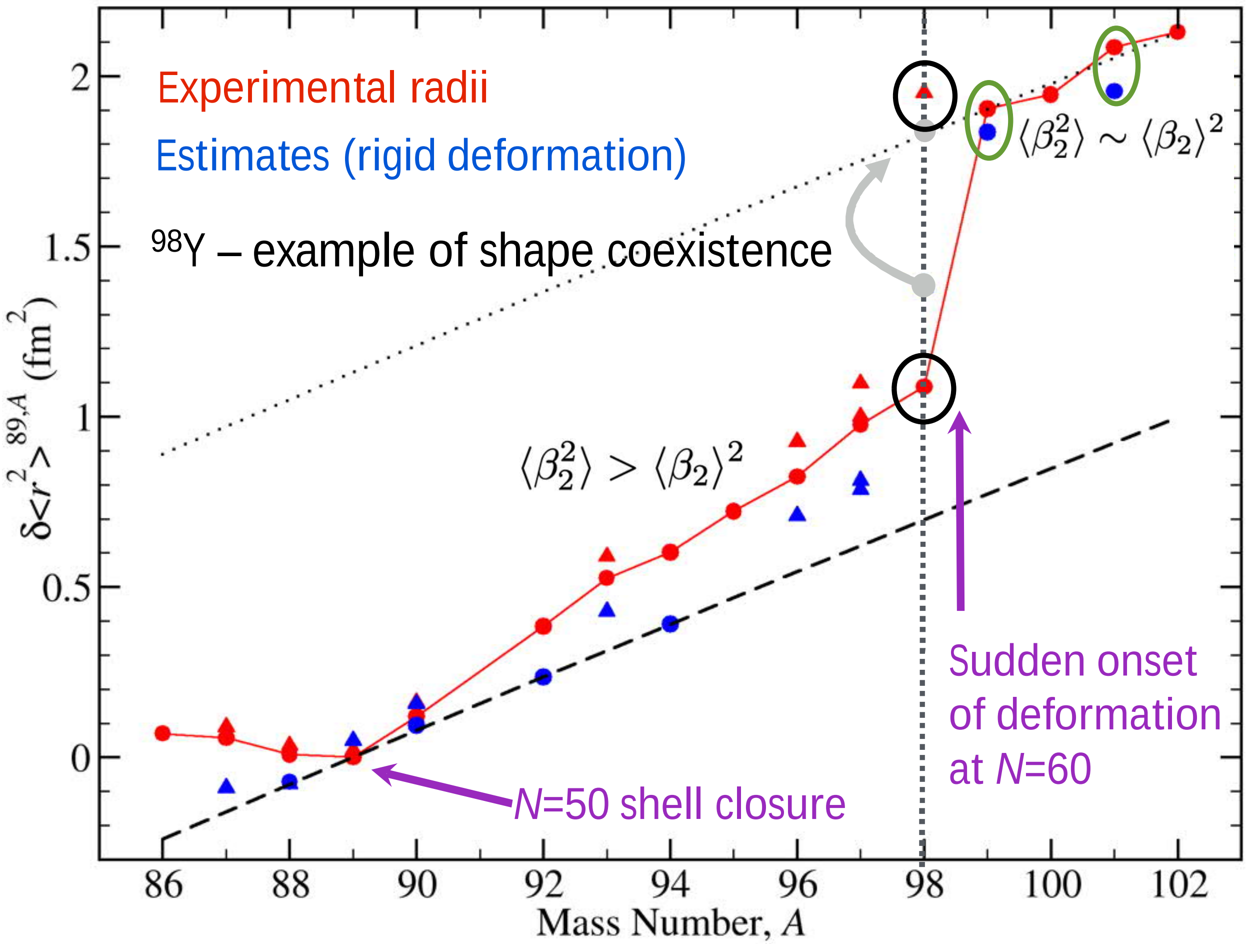
$$Q_q^n = eZ\sqrt{\frac{4\pi}{2n+1}}\langle I | r_n^n Y_q^n(\theta_n, \varphi_n) | I \rangle$$



- Electric monopole = $eZ\sqrt{4\pi}\langle I | Y_0^0 | I \rangle = eZ$
- Electric dipole = $eZ\sqrt{\frac{4\pi}{3}}\langle I | r\, Y_q^1 | I \rangle$ = 0
- Electric quadrupole: $Q_q^2 = eZ\sqrt{\frac{4\pi}{5}} r^2 Y_q^2$

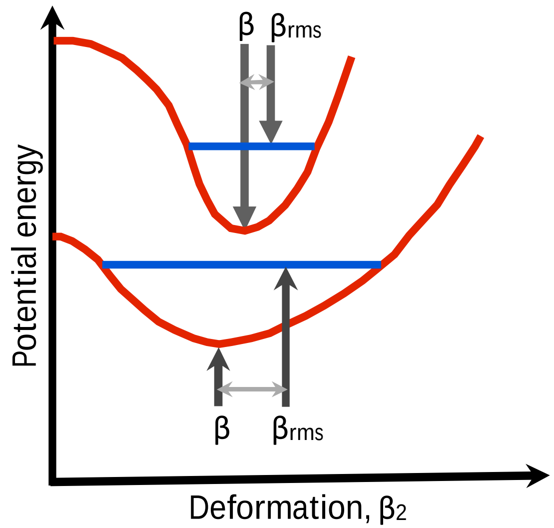
How ”soft” or ”rigid” are nuclei?

UNIVERSITY OF JYVÄSKYLÄ



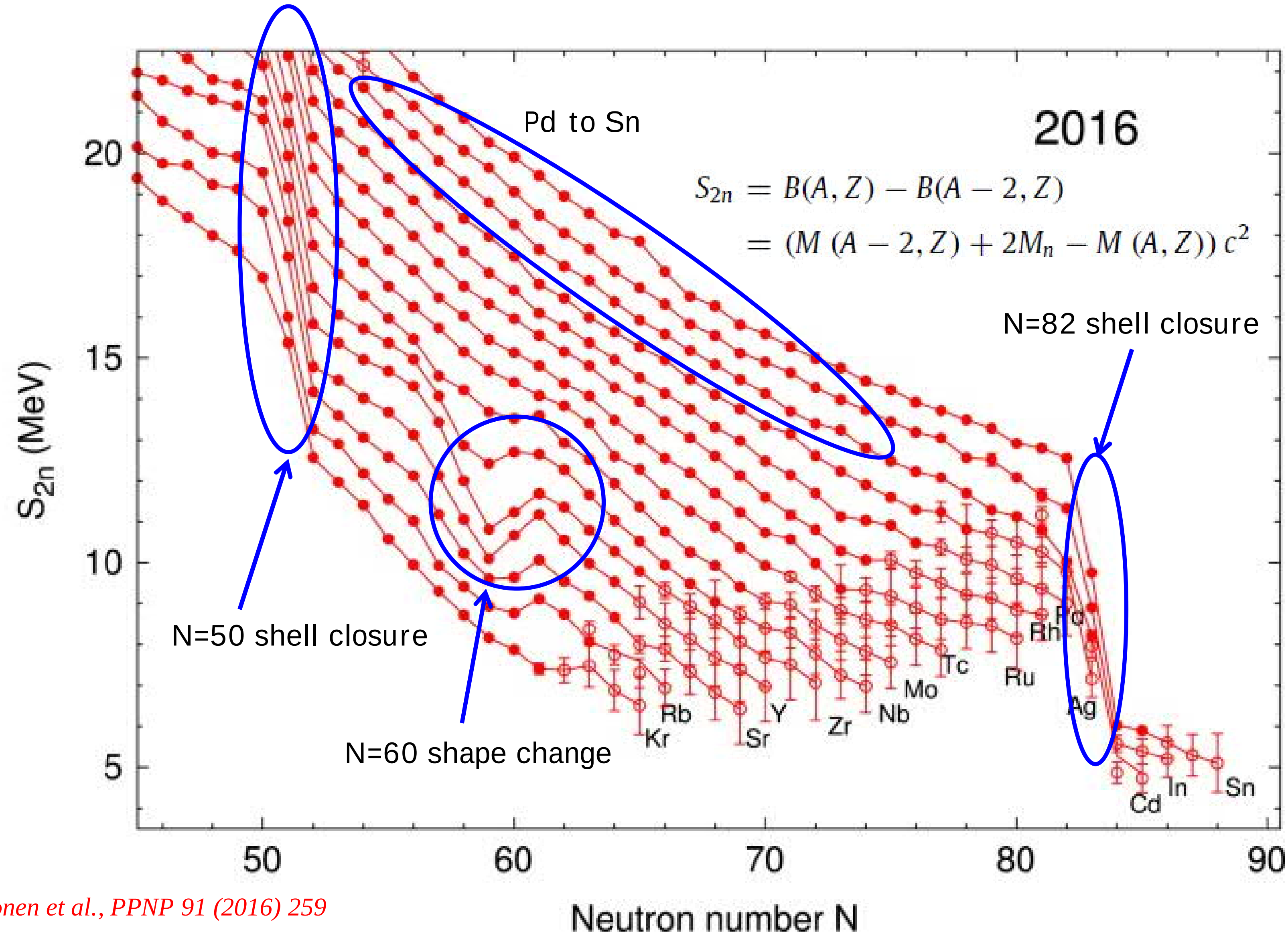
$$Q_s \rightarrow \langle \beta_2 \rangle$$

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



The difference between <β_2> and <β_2^2> gives the “softness” / “rigidity”.

Complementarity: the nuclear mass surface

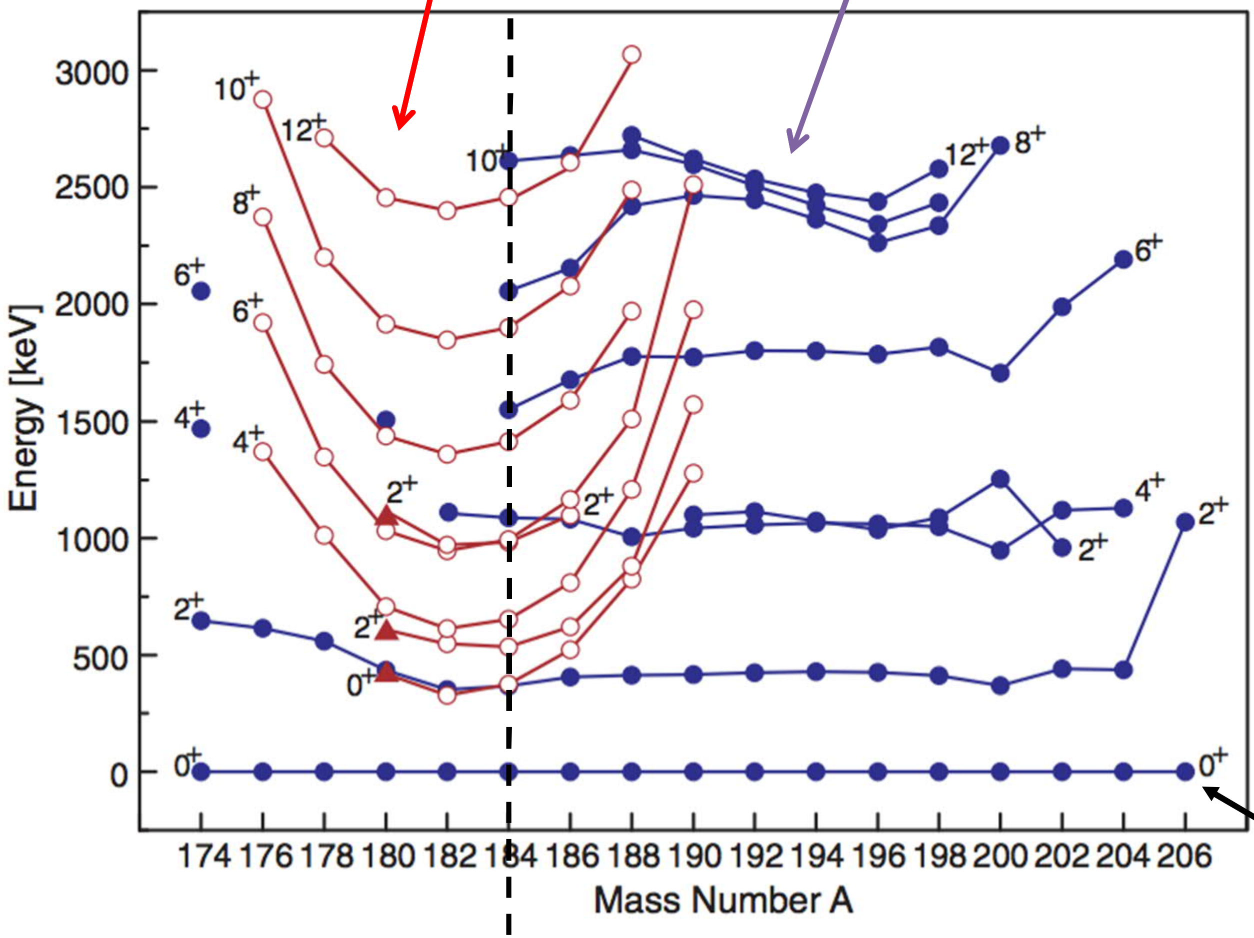


T. Eronen et al., PPNP 91 (2016) 259

Nuclear level systematics & coexistence

Prolate

Oblate



N=104 mid-shell

Coexistence of different bands in Hg isotopes

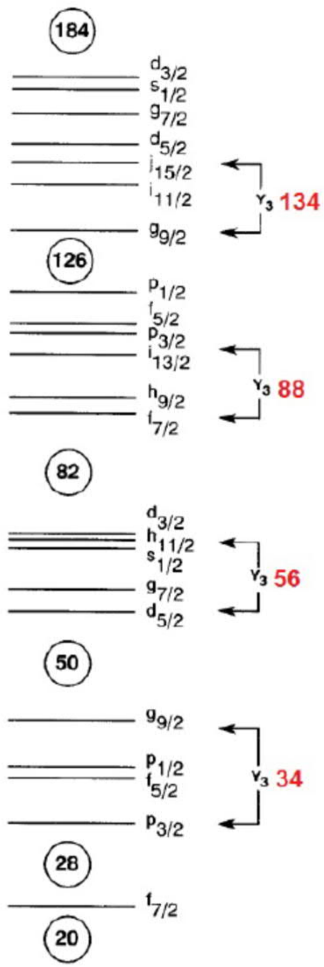
Prolate "intruder" states come down in energy towards minimum around *N=104* mid-shell region

Studied by many nuclear spectroscopy techniques

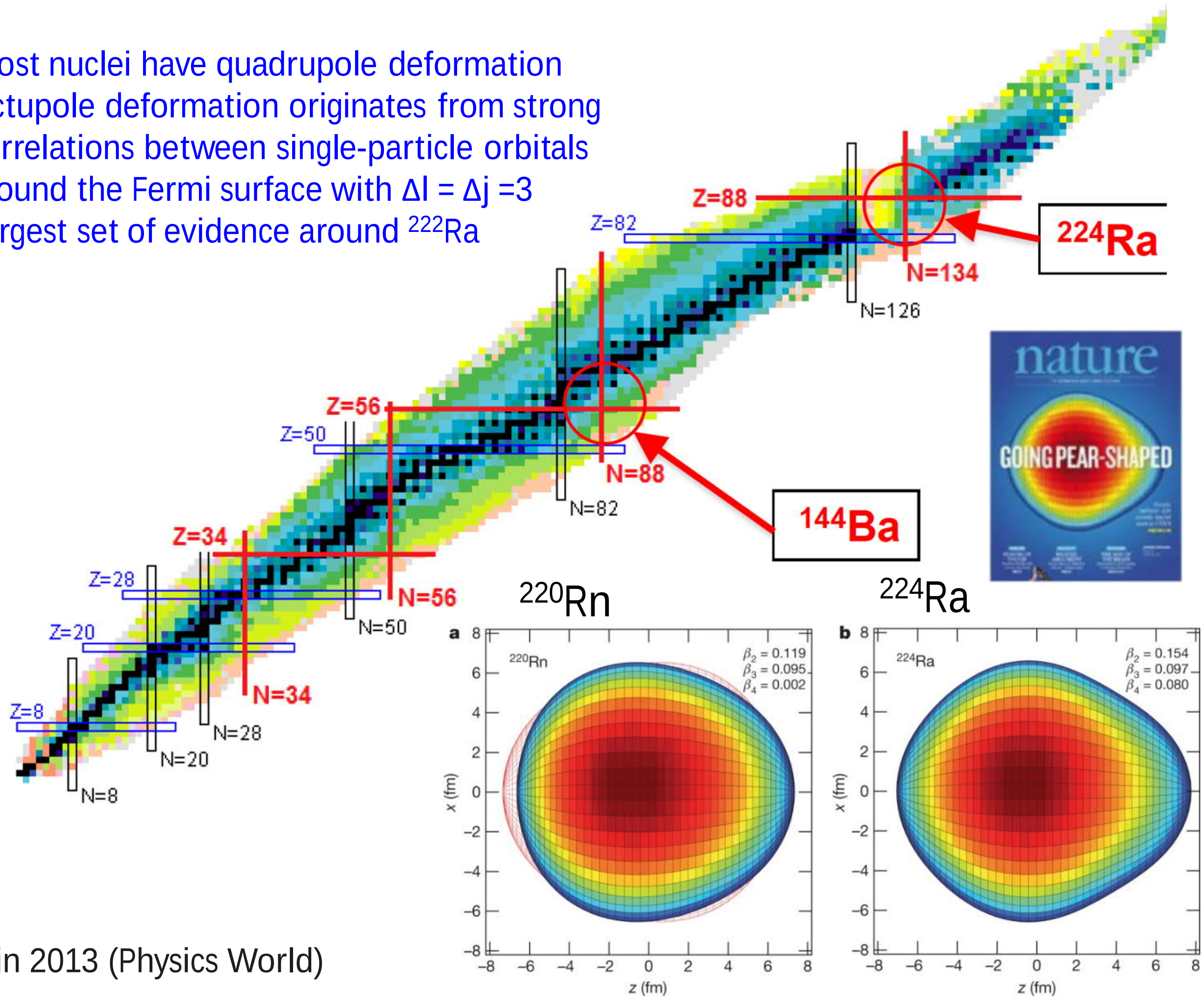
Ground state (probed by laser spectroscopy). Charge radius difference linked to the odd neutron driving deformation.

I.D. Moore, EJC 2021

Finally, even more exotic deformation



- Most nuclei have quadrupole deformation
- Octupole deformation originates from strong correlations between single-particle orbitals around the Fermi surface with Δl = Δj =3
- Largest set of evidence around ^{222}Ra

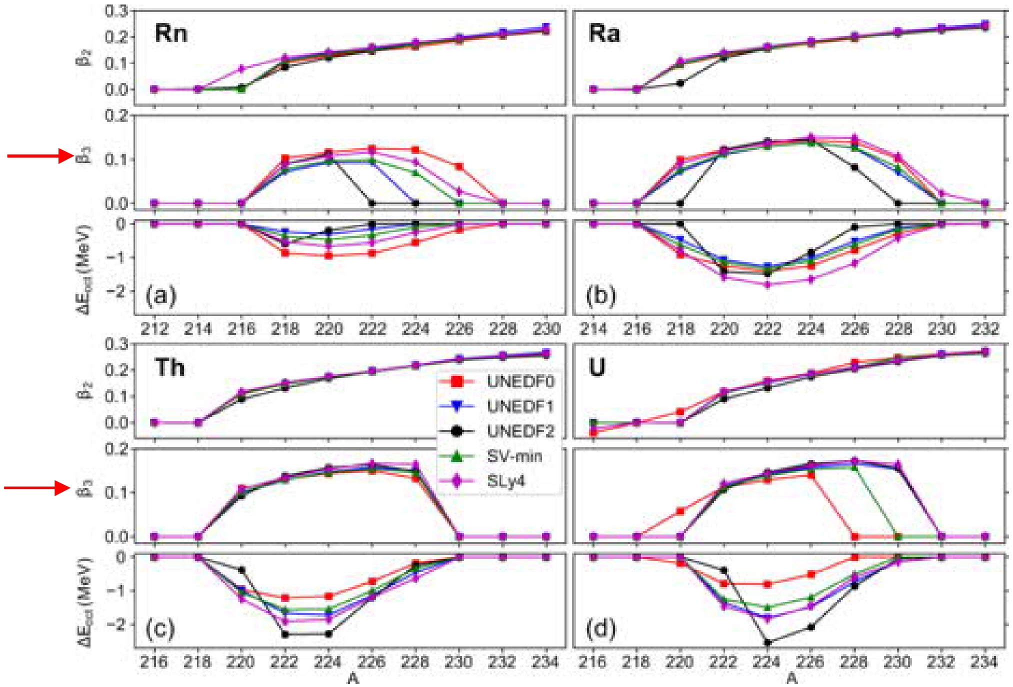


Top 10 breakthrough in physics in 2013 (Physics World)

"Pear-shaped nuclei discovery challenges time travel hopes"

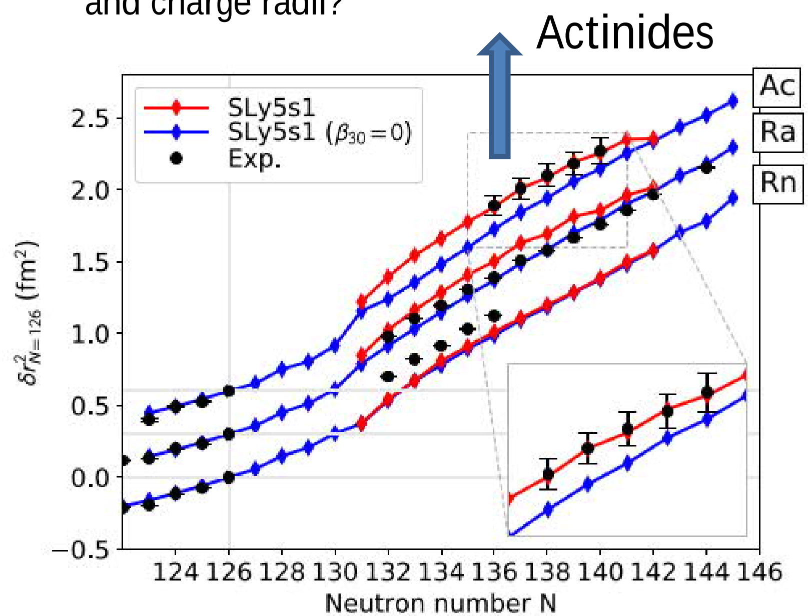
*L.P. Gaffney et al. Nature **497** (2013) 199*

A ”pear-shaped” actinide region?



Y. Cao et al., PRC 102 (2020) 024311

Is there a link between octupole deformation and charge radii?



E. Verstraelen et al., PRC 100 (2019) 044321

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

- Isotopes of Rn, Ra, Th and U are predicted to have the strongest octupolar ”correlations”
- Constraint of candidates for experimental studies of electric-dipole moment (EDM), and thus existence of physics Beyond the Standard Model

New experimental and theoretical efforts are required to systematically explore this question!