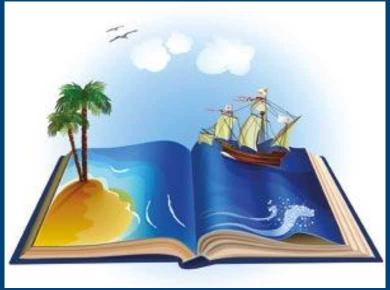
Ground-State Properties from Laser Spectroscopy
Part I: Scientific Motivation and Challenges



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Plan for My Lectures

- **Part I**: scientific motivation and main challenges for studying radioactive nuclei and (super)heavy elements
- **Part II**: basic principles of laser spectroscopy and commonly employed technique (RIS, CLS, …)
- **Part III**: specific examples from select recent experiments

Related information in lectures by Jackie Gates, Helena Albers
Mass measurements / ion traps will be covered by Vladimir Manea

Part I – Laser Spectroscopy for Nuclear Physics

- Nuclear (ground-state) properties we obtain via laser spectroscopy
  - ➢ Mean square charge radii from isotope shifts
  - ➢ Quadrupole moment from hyperfine spectroscopy
- What do need to know on the atomic structure ?
- impact of many-body effects, and for heavier systems:
  special relativity and quantum electrodynamics (QED)

Related Literature – Review Articles

Progress in Particle and Nuclear Physics 116 (2021) 103834

Contents lists available at ScienceDirect

Progress in Particle and Nuclear Physics

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Review

Recent progress in laser spectroscopy of the actinides

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ABSTRACT

The interest to perform laser spectroscopy in the heaviest elements arises from the strong impact of relativistic effects, electron correlations and quantum electrodynamics on their atomic structure. Once this atomic structure is well understood, laser spectroscopy also provides access to nuclear properties such as spins, mean-square charge radii and electromagnetic moments in a nuclear-model independent way. This is of particular interest for the heaviest actinides around $N = 152$, a region of shell-stabilized deformed nuclei. The experimental progress of laser spectroscopy in this region benefitted from continuous methodological and technical developments such as the introduction of buffer-gas-stopping techniques that enabled the access to ever more exotic nuclei far-off stability. The key challenges faced in this endeavor are small yields, nuclides with rather short half-lives and the need to search for atomic transitions in a wide spectral range guided by theoretical predictions. This paper describes the basics of the most common experimental methods and discusses selected recent results on the atomic and nuclear properties of the actinides up to nobelium where pioneering experiments were performed at the GSI Helmholtzzentrum für Schwerionenforschung in Darmstadt, Germany.

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Review

Laser spectroscopy for the study of exotic nuclei

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ABSTRACT

Investigation into the properties and structure of unstable nuclei far from stability is a key avenue of research in modern nuclear physics. These efforts are motivated by the continual observation of unexpected structure phenomena in nuclei with unusual proton-to-neutron ratios. In recent decades, laser spectroscopy techniques have made significant contributions in our understanding of exotic nuclei in different mass regions encompassing almost the entire nuclear chart. This is achieved through determining multiple fundamental properties of nuclear ground and isomeric states, such as nuclear spins, magnetic dipole and electric quadrupole moments and charge radii, via the measurement of hyperfine structures and isotope shifts in the atomic or ionic spectra of the nuclei of interest. These properties offer prominent tests of recently developed state-of-the-art nuclear theory and help to stimulate new developments in improving the many-body methods and nucleon–nucleon interactions at the core of these models. With the aim of exploring more exotic short-lived nuclei located ever closer to the proton and neutron driplines, laser spectroscopy techniques, with their continuous technological developments towards higher resolution and higher sensitivity, are extensively employed at current- and next-generation radioactive ion beam facilities worldwide. Ongoing efforts in parallel promise to improve the availability of these even more exotic species at next-generation facilities. Very recently, an innovative application of laser spectroscopy on molecules containing short-lived nuclei has been demonstrated offering additional opportunities for several fields of research, e.g. fundamental symmetry studies and astrophysics. In this review, the basic nuclear properties measurable with laser spectroscopy will be introduced. How these observables are associated with nuclear structure and nucleon–nucleon interactions will be discussed. Following this, a general overview of different laser spectroscopy methods will be given with particular emphasis on technical advancements reported in recent years. The main focus of this article is to review the numerous highlights that have resulted from studying exotic nuclei in different mass regions with laser spectroscopy techniques since the last edition in this series. Finally, the challenges facing the field in addition to future opportunities will be discussed.

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2. Nuclear properties from atomic spectroscopy ... 5
   2.1. Magnetic dipole and electric quadrupole hyperfine interactions ... 5

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- basics on laser spectroscopy are discussed in numerous textbooks (see next slide)
- several recent review articles address common techniques applicable to (heavy) radio-nuclides that are often short-lived and can typically be produced in low quantities

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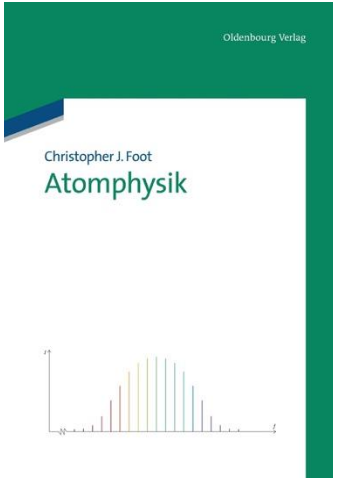
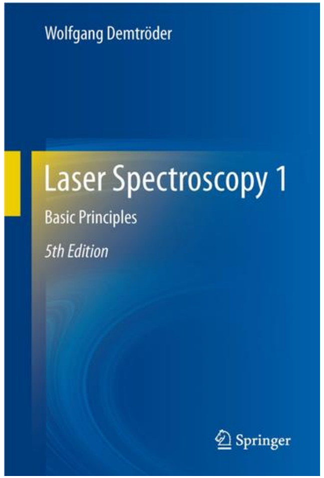
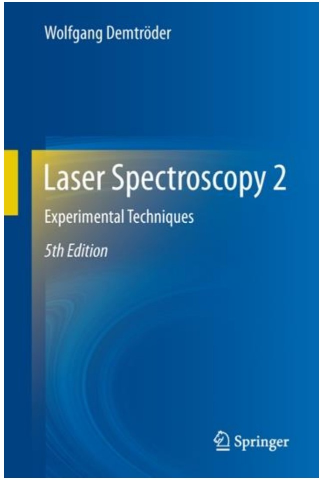
Related Literature – Textbooks

Atomic physics
Christopher Foot

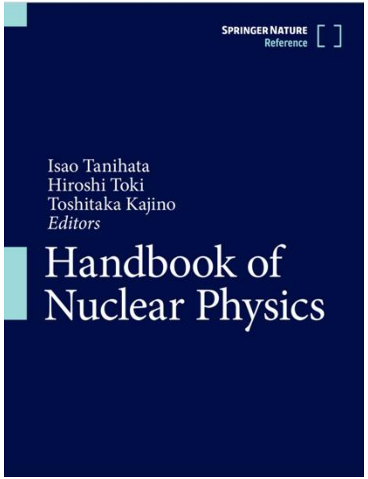


Laser Spectroscopy 1&2
Wolfgang Demtröder





Handbook of
Nuclear Physics



Facilties Worldwide

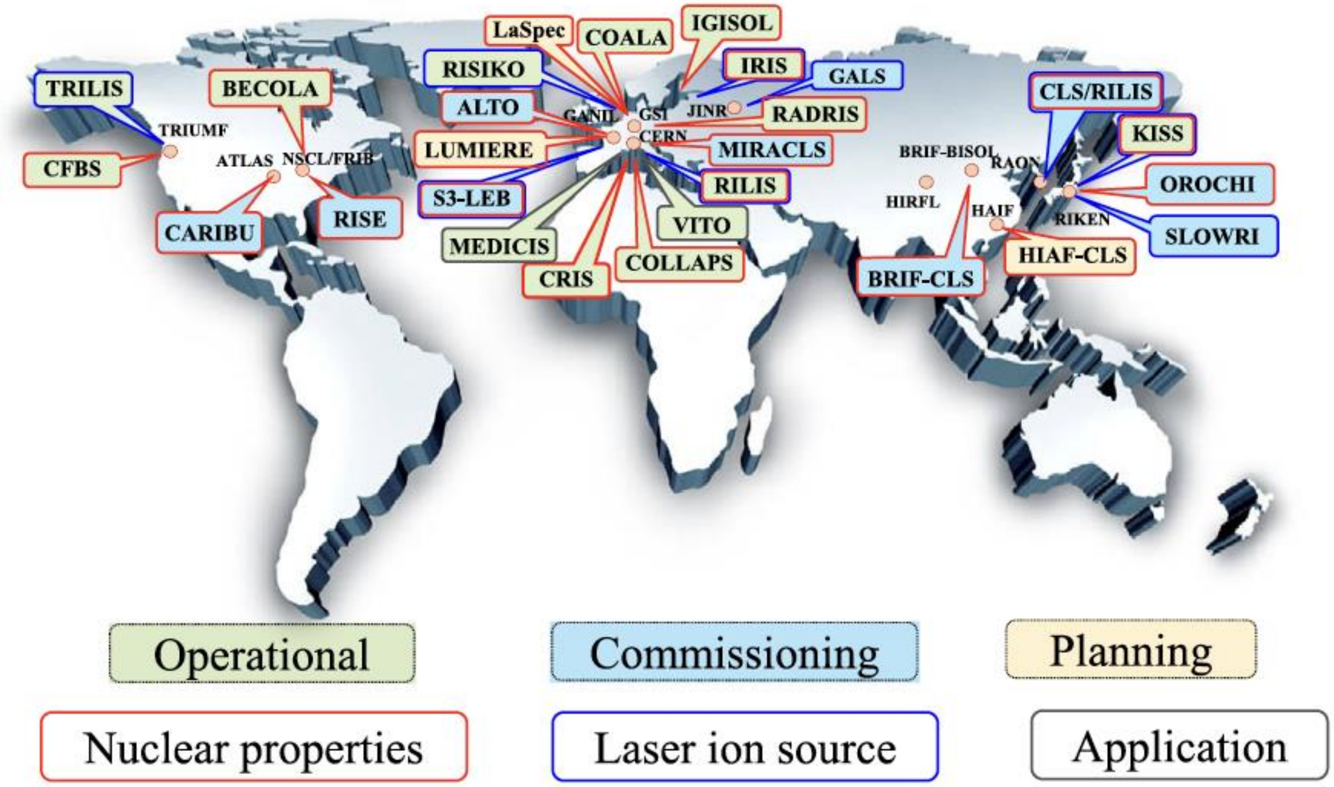
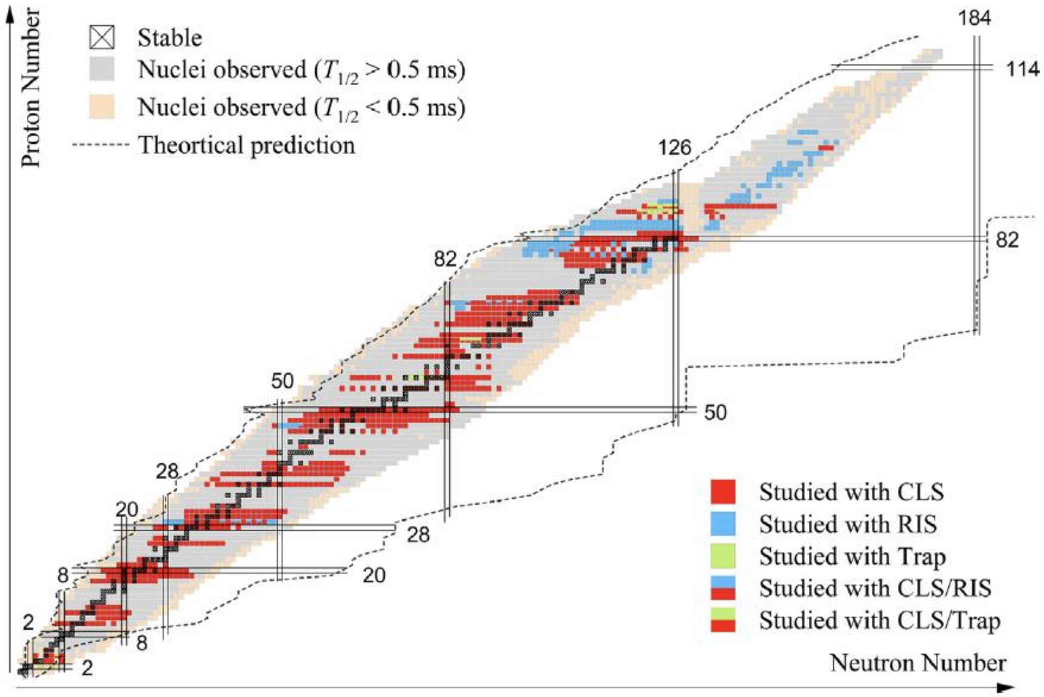


Fig. from X.F. Yang et al., Prog. Part. Nucl. Phys. 129 (2023) 104005

- RIB facilities are discussed in lecture by Helena Albers
- Typically, two classes: **ISOL-type** and **in-flight-type** facilities
- employing gas-cell based approaches: nowadays nuclides from essentially all production schemes available

Laser Spectroscopy Landscape



CLS: Collinear Laser Spectroscopy

RIS: Resonant Laser Ionization Spectroscopy

- numerous nuclides (and isomers) have been studied by laser spectroscopy to reveal nuclear structure evolution:
  location of shell closes, onset of deformation, nuclear spins, …
- nowadays, essentially all elements accessible (with few exceptions)
- applied techniques often tailored to production schemes (ISOL, in-flight, gas cell, …)

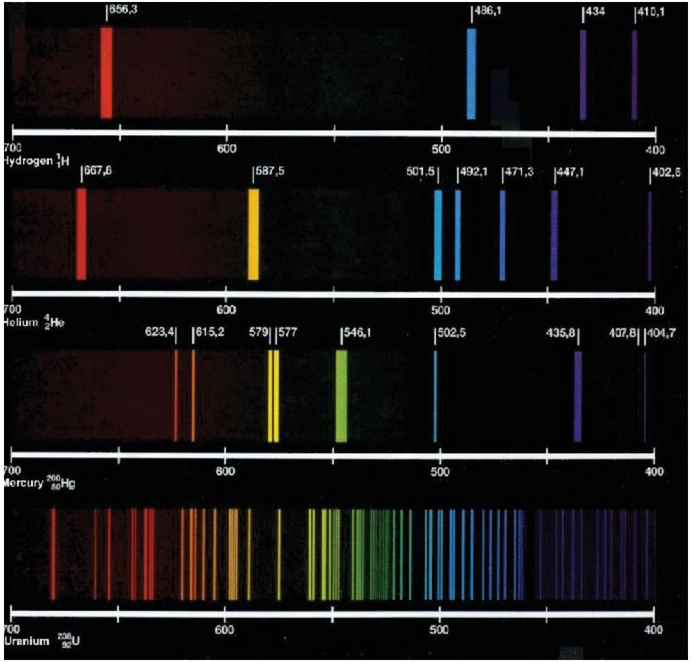
Characteristic Emission Lines

Hydrogen

Helium

Mercury

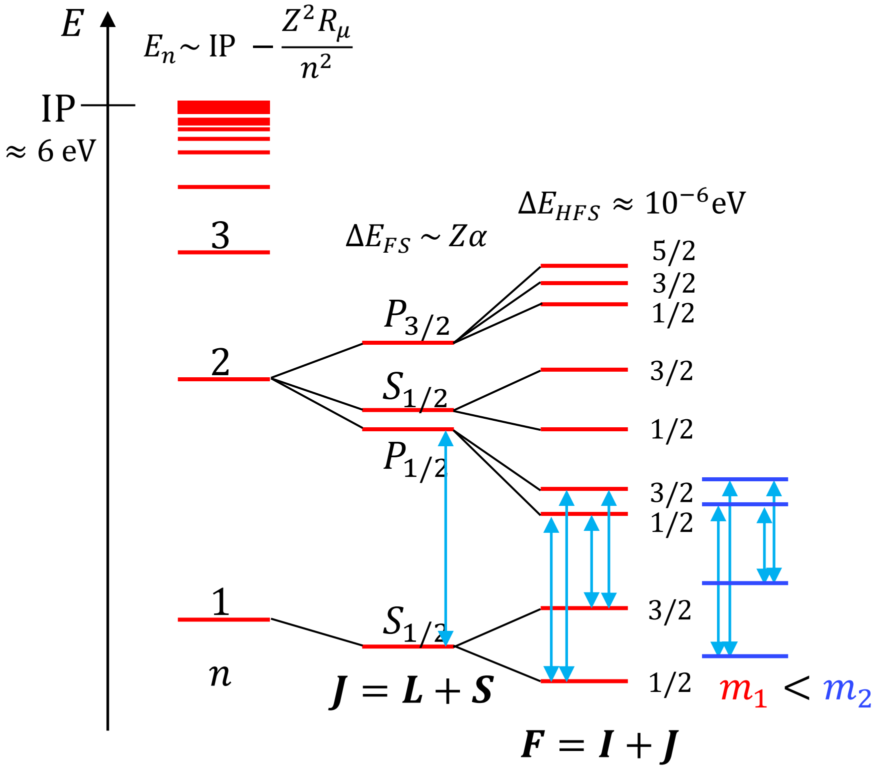
Uranium



Wavelength / nm

- emission spectra of element provide specific fingerprint
- line structure reflects atomic level structure
- with highest resolution nuclear effects are accessible

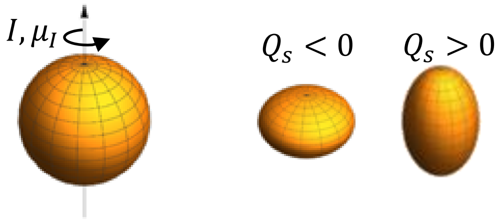
Electronic Structure of Atoms and Nuclear Properties



$\Delta E_{FS} \approx 10^{-1} \dots 10^{-3}$ eV

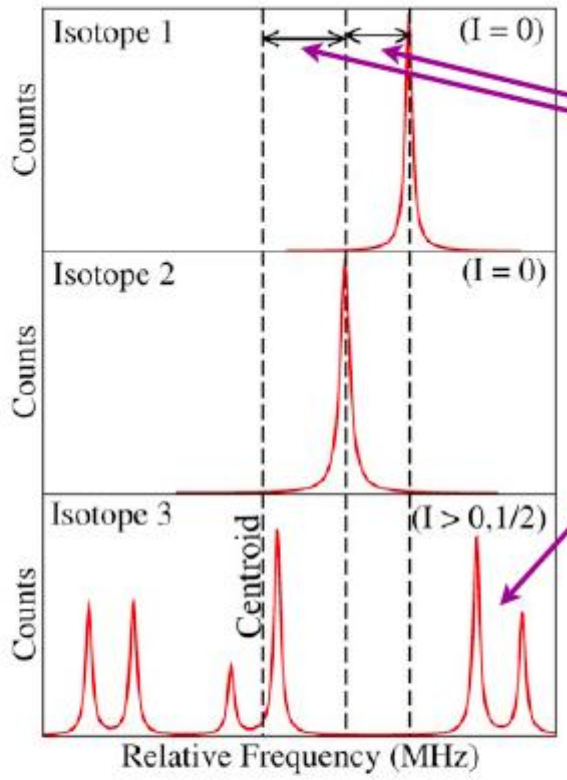
$$A = \frac{\mu_I \overline{H(0)}}{IJ} \qquad B = eQ_s \left\langle \frac{\partial^2 \phi}{\partial z^2} \right\rangle_{r=0}$$

Atomic shell
Nucleus



- Atomic level structure probed by laser spectroscopy with (optical) transitions
- ionization potential can be determined from convergence of Rydberg series
- Nuclear spins and electromagnetic moments from hyperfine splitting

Laser Spectroscopy of Radionuclides



Information on nuclear properties from laser spectroscopy:

- isotope shift yields information on changes in mean-square charge radii from which we infer nuclear size

$$\delta\langle r^2\rangle^{AA'} = \left(\Delta\nu^{AA'} - \frac{A - A'}{AA'} M\right)\frac{1}{F}$$

Experiment Theory

- hyperfine spectroscopy yields parameters linked to magnetic dipole moment and spectroscopic quadrupole moment

I, μ_I

$Q_s < 0$ $Q_s > 0$

$$A = \mu \frac{B_e(0)}{IJ} \quad B = eQ_s \left\langle \frac{\delta^2 V}{\delta z^2} \right\rangle$$

Nuclear Atomic

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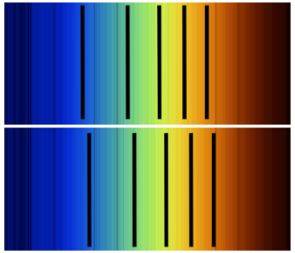
Isotopic Shift of Atomic Transitions - Basics

Isotope 1

Isotope 2



Isotope shift of atomic transition = Frequency difference of the same atomic transition in two different isotopes

Harold C. Urey (1932):

- Discovery of the splitting of the H_β line in the spectrum of natural hydrogen
- Identification of the isotope deuterium ("heavy hydrogen")

Nobel Prize for Chemistry 1934

$$\delta\nu^{A,A'} = \nu_{A'} - \nu_{A} = F\,\delta\langle r^2\rangle^{A,A'} + M\,\frac{m_{A'} - m_{A}}{m_{A'} m_{A}}$$

field shift mass shift

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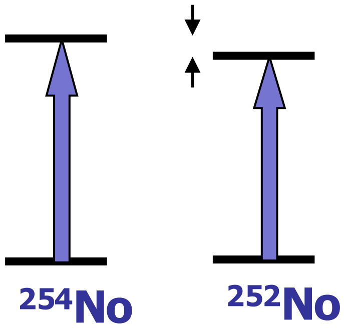
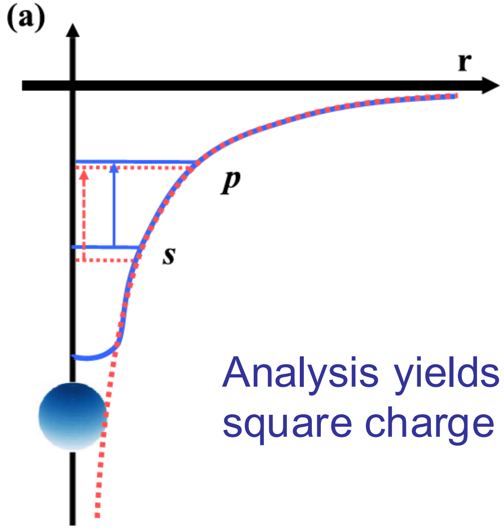
Isotopic Shift of Atomic Transitions



Isotope shift of atomic transition = Frequency difference of the same atomic transition in two different isotopes



Cause:

- charge (proton) distribution in nucleus changes for different isotopes
- shift of energy levels
- stronger effect for orbitals with better overlsp with nucleus

Analysis yields change of mean-square charge radius **$\delta\langle r^2\rangle$**

Isotope Shift – Field Shift

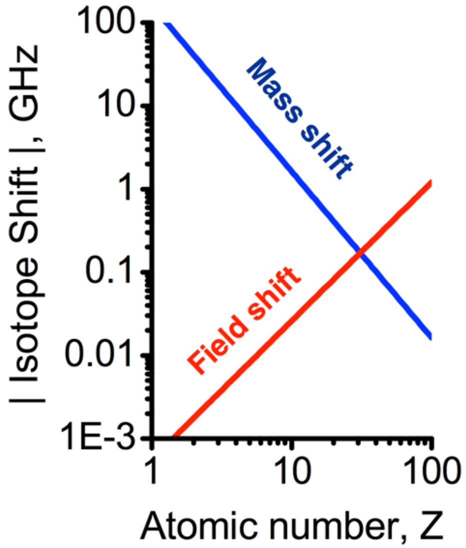
$$\Delta\nu_{FS} = \frac{2\pi Z}{3}\Delta|\psi(0)|^2\,\delta\langle r^2\rangle$$

$$FS \propto \frac{Z^2}{\sqrt[3]{A}}$$

$$\delta\langle r^2\rangle = R_2^2 - R_1^2$$

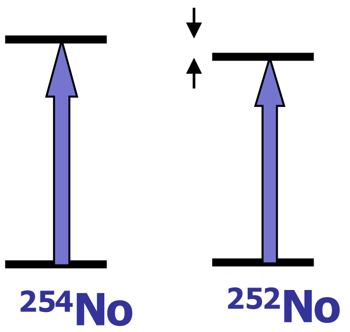
Probability density of electrons at nucleus (r=0, electronic wave function)

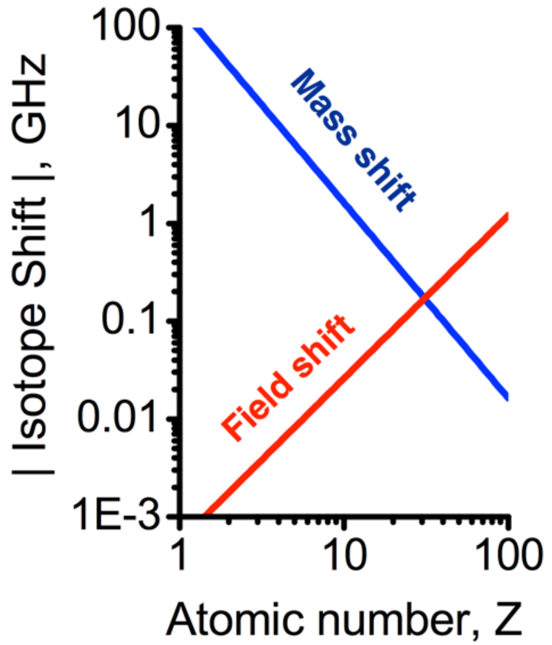
Difference of mean-square charge radii (nuclear part)



due to the Z^2 dependence, FS easier accessible in heavy nuclei

Nuclear Properties and Isotope Shift





For axially symmetric nucleus (volume constant):

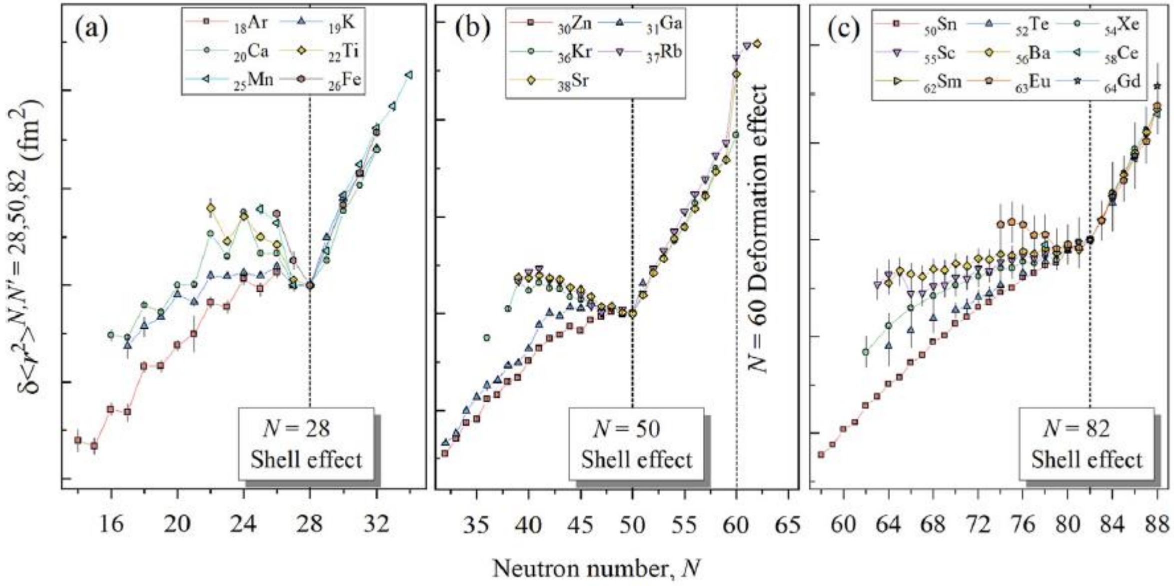
$$\delta\langle r^2\rangle = \delta\langle r^2\rangle_{\rm sph} + \langle r^2\rangle_{\rm sph}\frac{5}{4\pi}\delta\langle\beta_2^2\rangle$$

Volume Deformation

mean-square charge radius depends quadratically on β_2 – no distinction between oblate / prolate

Nuclear Properties – Changes in Charge Radii

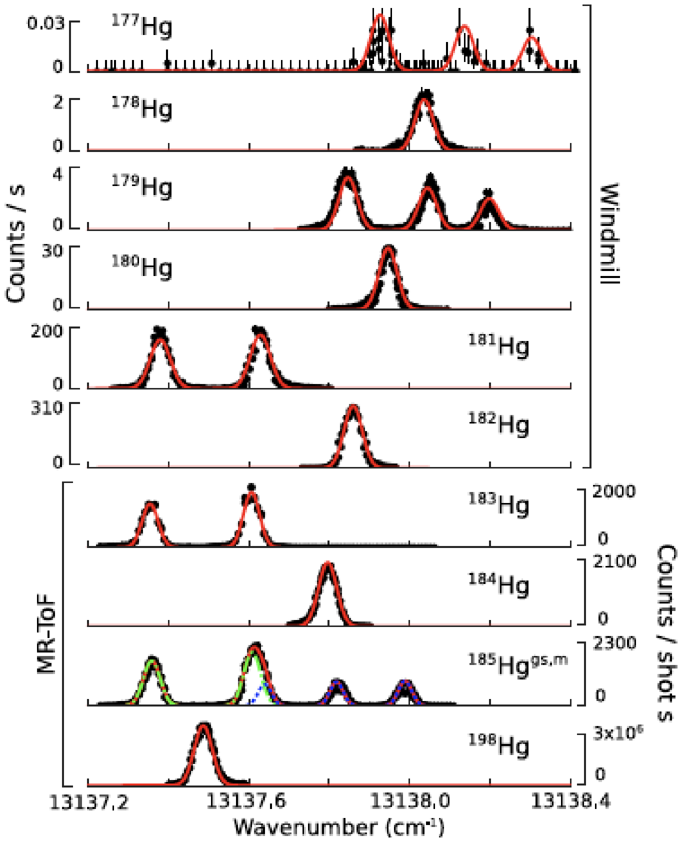
Changes in charge radii of isotopic chains

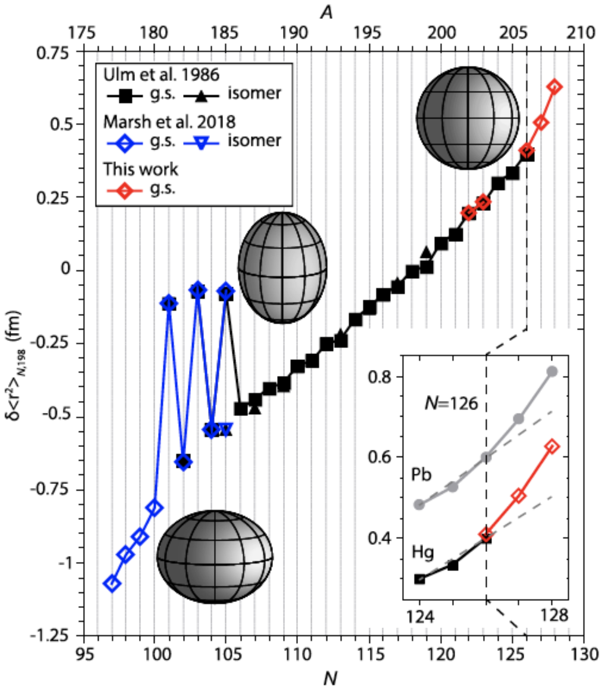


- major shell closures are reflected in charge radii of isotopic chains as "kinks"
- kinks are well established for major shell such as N = 28, 50, 82
- situation for other regions and deformed shell gaps less clear

Fig. from X.F. Yang et al., Prog. Part. Nucl. Phys. 129 (2023) 104005

Laser Spectroscopy of Hg Isotopes at ISOLDE





T. Day Goodacre et al., Phys. Rev. Lett. 126, 032502 (2021)

See also: B. A. Marsh et al.,, Nat. Phys. 14, 1163 (2018).

S. Sels et al. Phys. Rev. C 99, 044306 (2019).

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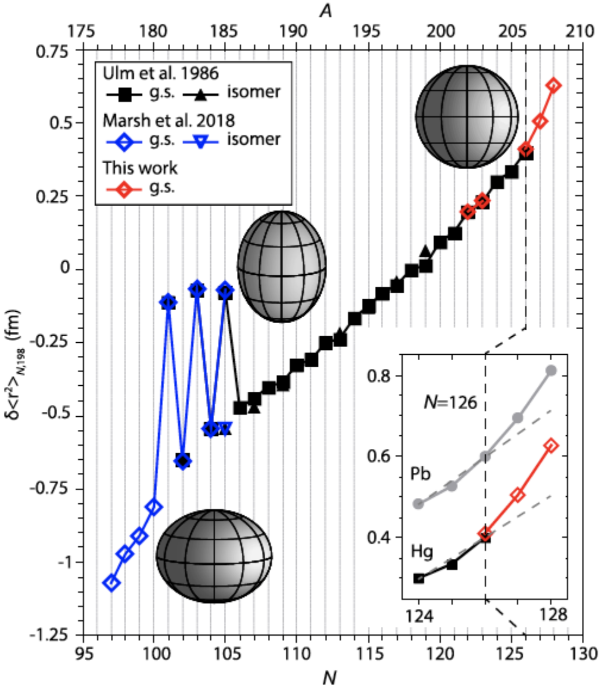
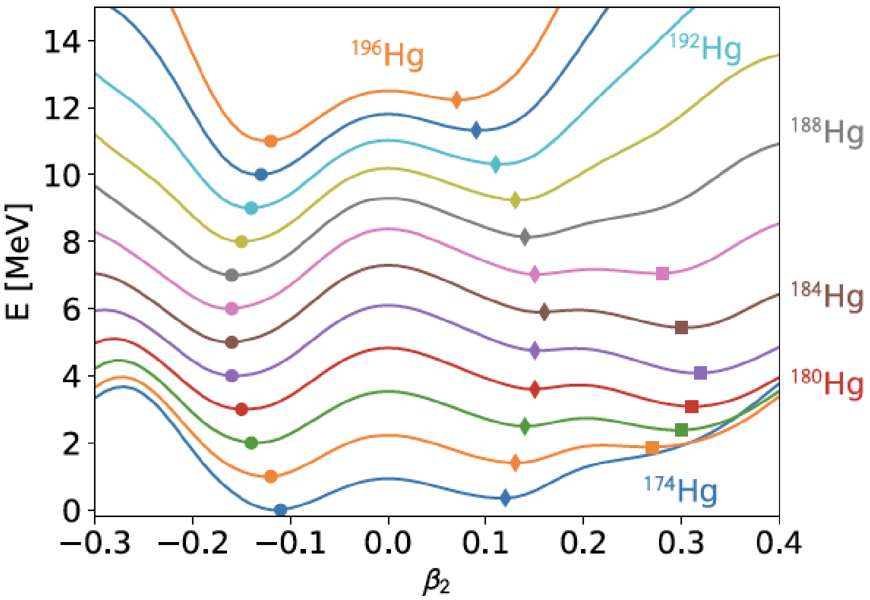
Laser Spectroscopy of Hg Isotopes at ISOLDE



Calculated potential energies for Hg isotopes



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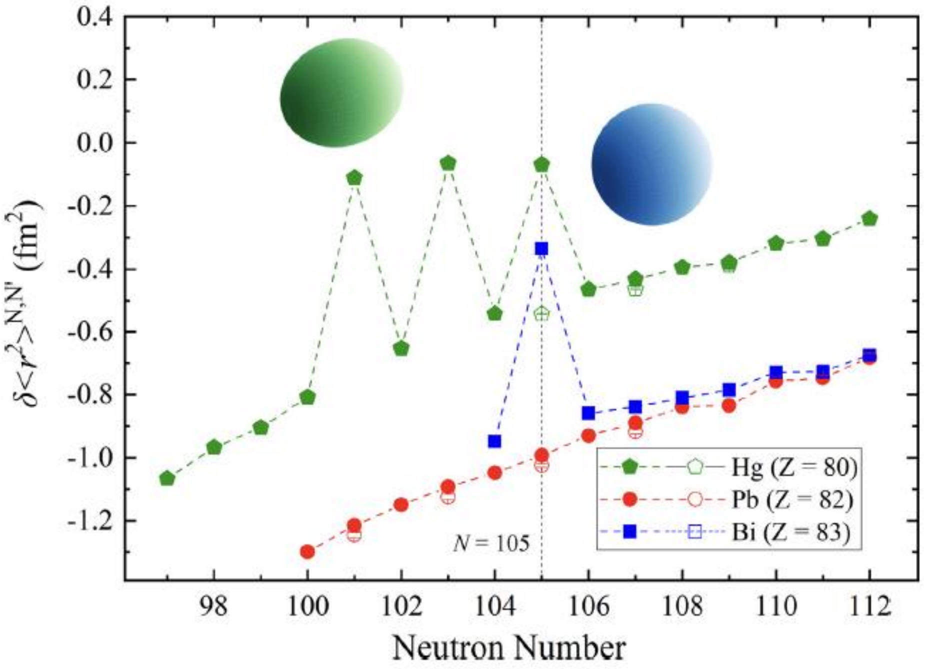
Nuclear Properties

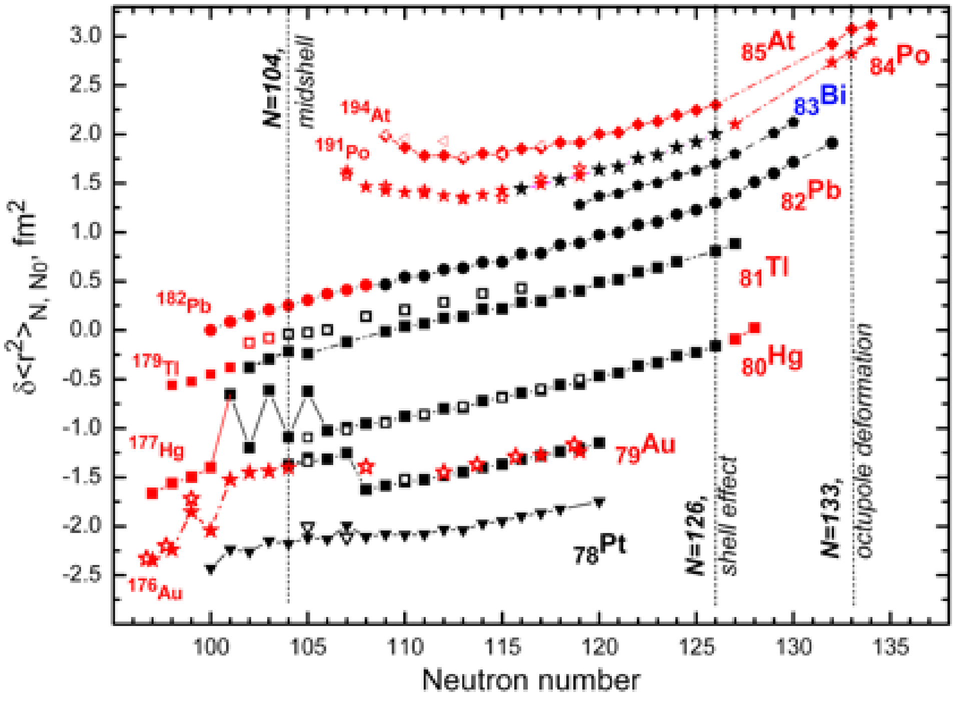


- Shape staggering / coexistence has been observed for neutron-deficient Hg (and Bi, Au) isotopes
- Early observations in Hg at ISOLDE already in the 19xxs (without lasers)
- triggered lots of experiments, today good data for many isotopic chains from N=98-126 below lead

Fig. from X.F. Yang et al., Prog. Part. Nucl. Phys. 129 (2023) 104005

Charge Radii N=104-132

Data obtained at ISOLDE / CERN



general features commonly observed:

- smooth trend in between shell closures
- rms charge radii show kinks at neutron shell closures
- also, odd-even staggering (OES), odd-N isotope has usually smaller charge radius average of two even-N neighbors

Nuclear Density Distributions

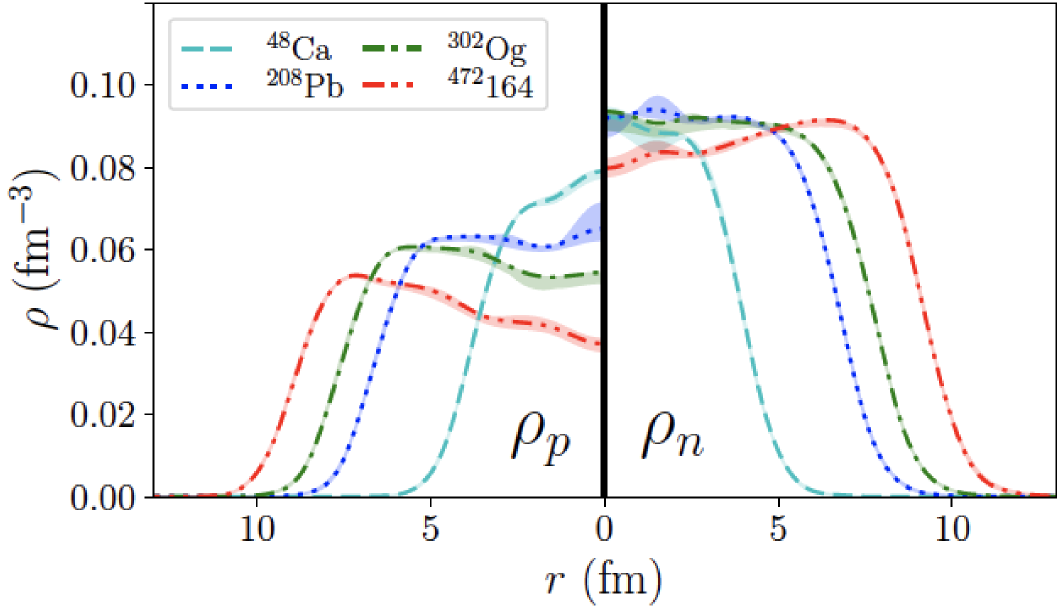


Fig. 5 Radial proton (left) and neutron (right) densities of doubly-magic nuclei ^{48}Ca, ^{208}Pb, ^{302}Og, and ^{472}164 obtained in nuclear DFT with three different energy density functionals. The shaded areas indicate the spread of DFT predictions. (Modified from [125].)

Figure courtesy W. Nazarewicz

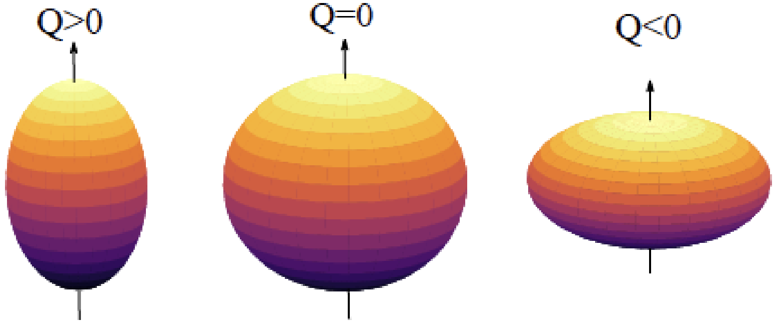
Many nuclear models predict:

- radial proton distribution of (super)heavy nuclei shows a central depression
- around Z=102, this effect is predicted to be about 10%
- direct measurements of this feature are not feasible

Mean-Square Charge Radius and Nuclear Deformation

Assumptions:

- constant nuclear volume
- uniform charge distribution
- small quadrupole deformation ($\beta_2 \ll 1$)



prolate $\beta_2>0$ | spherical $\beta_2=0$ | oblate $\beta_2<0$

Mean-square charge radius:

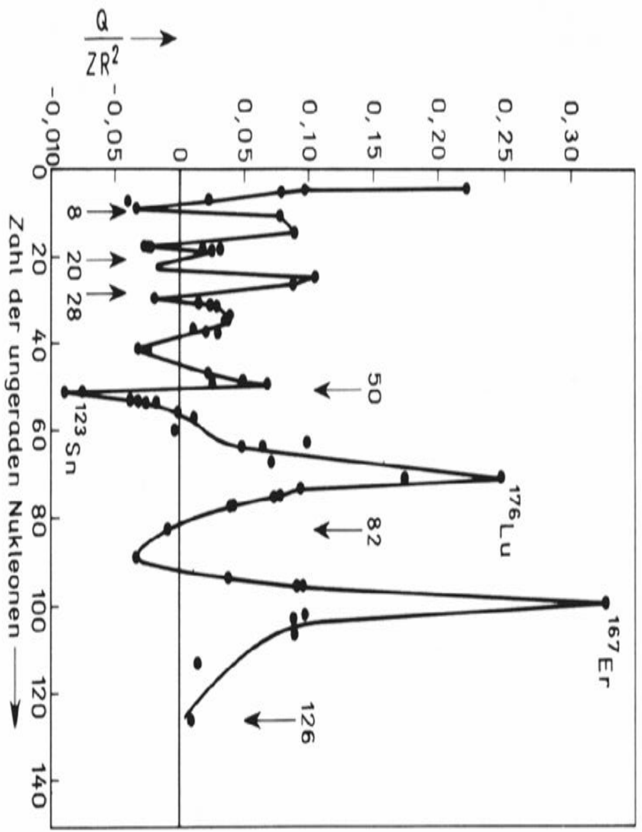
$$\langle r^2 \rangle \approx \langle r^2 \rangle_0 \,(1 + 5\beta_2^2 / 4\pi)$$

Change between isotopes:

$$\delta\langle r^2 \rangle = \delta\langle r^2 \rangle_{sph} + (3R_0^2 / 4\pi)\, \delta(\beta_2^2)$$

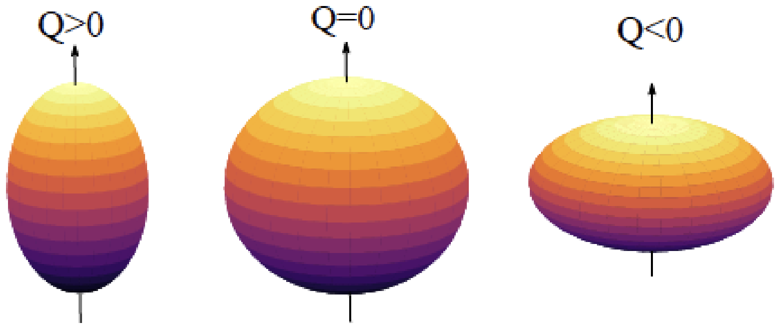
- deformation increases the rms charge radius.
- radius depends on β_2^2, thus prolate and oblate shapes produce the same increase. $\beta_2<0$
- in laser spectroscopy, changes in β_2 dominate the deformation contribution to isotope shifts.

Quadrupole Moments



- Q is zero close to magic (nucleon) numbers
- nuclei with half-filled shell are typically deformed
- oblate nuclei (Q<0) are less abundant than prolate ones (Q>0)
- deformation of known nuclei:
  - about 45% are spherical
  - about 40% are prolate
  - about 10% are oblate
  - about 5% are triaxial

Electromagnetic Nuclear Properties



prolate spherical oblate

Deformed nuclei feature non-zero electric quadrupole moment

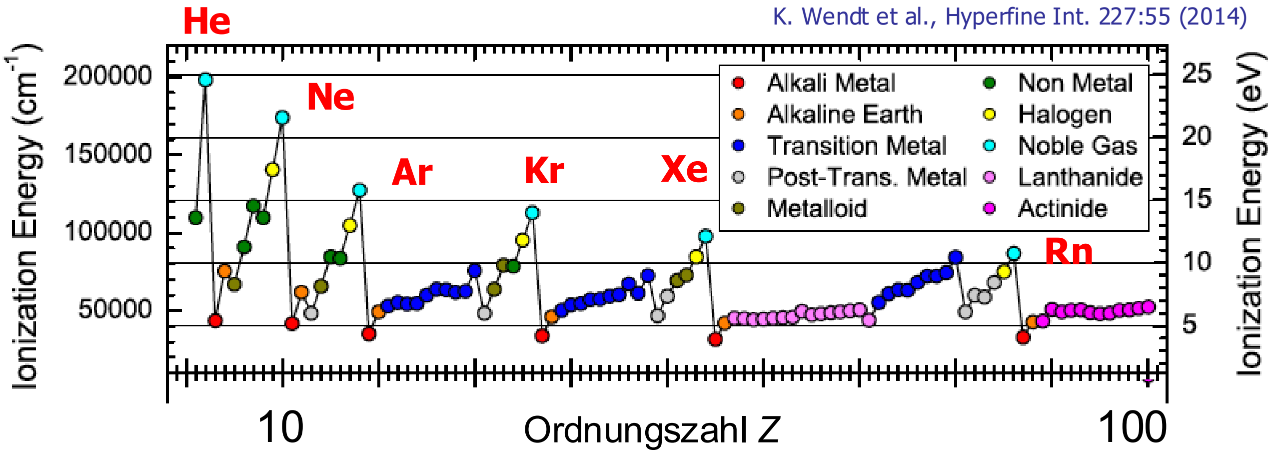
spectroscopic Quadrupole moment

$$Q_s = \langle I, m = I | \hat{Q}_2 | I, m = I \rangle = \sqrt{\frac{I(2I-1)}{(2I-1)(2I+3)(I+1)}} \langle I || \hat{Q}_z || I \rangle$$

intrinsic Quadrupole moment

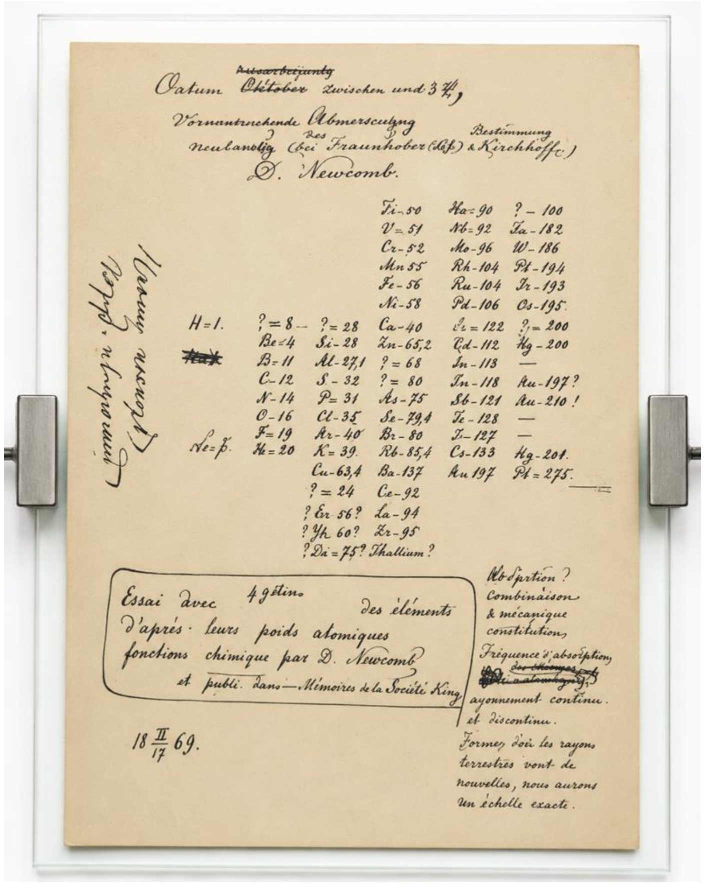
$$Q_0 = \frac{(I+1)(2I+3)}{3K^2 - I(I+1)} Q_s$$

First Ionisation Potential



- Architecture des Periodic Table of Elements reflects electronic shell structure: closed-shell elements (noble gases) have high IP
- relativistic effects gain in importance with increasing Z

Mendeleyev's Table in 1869



Mendeleyev's key achievements:

- Sorting elements according to their (assumed) weight
- Grouping elements with similar properties results in periodicity
- Based on this, he predicts / postulates existence of then undiscovered elements that were later found, for example Gallium, Germanium, Scandium

Mendeleev's Draft – 17/02/1869 jul.

(Mendeleev-Museum, St. Petersburg)

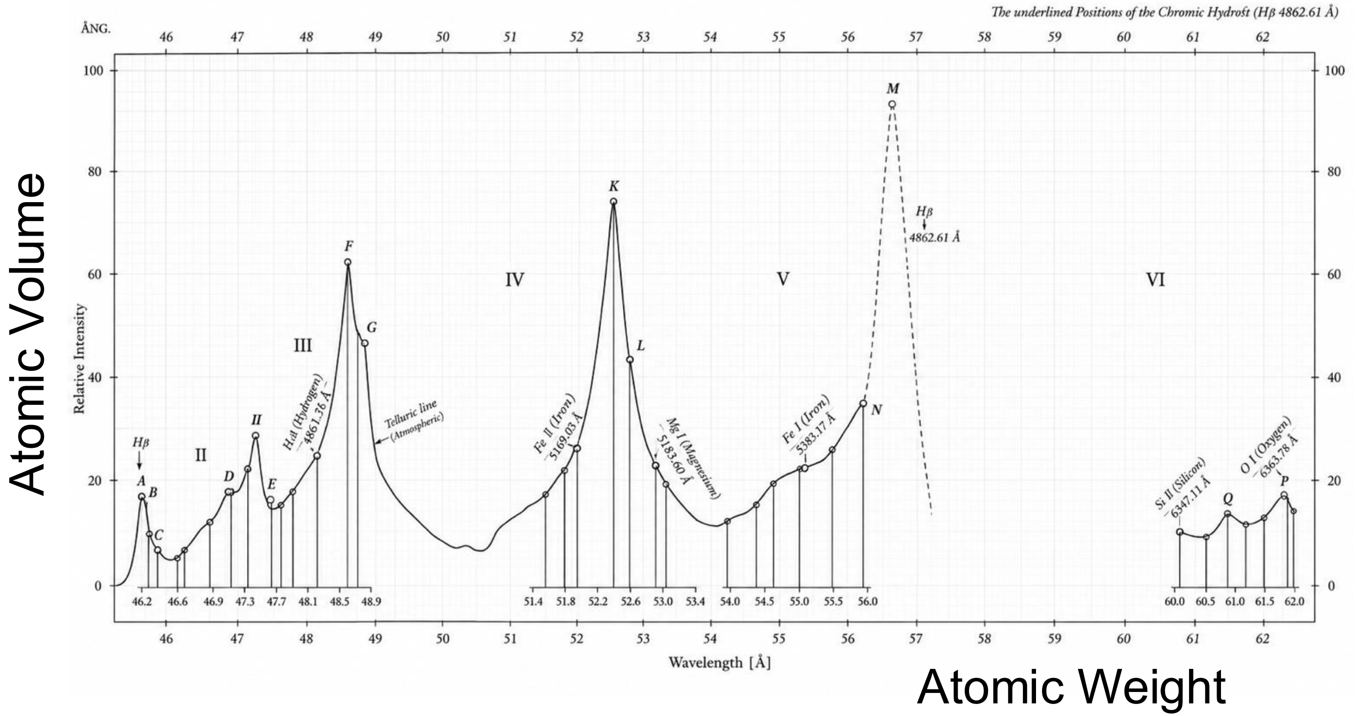
Meyer's first Table (*Modern Theories* 1864)

	4 werthig	3 werthig	2 werthig	1 werthig	1 werthig	2 werthig
	—	—	—	—	Li = 7,03	(Be = 9,3 ?)
Differenz =	—	—	—	—	16,02	(14,7)
	C = 12,0	N = 14,04	O = 16,00	Fl = 19,0	Na = 23,05	Mg = 24,0
Differenz =	16,5	16,96	16,07	16,46	16,08	16,0
	Si = 28,5	P = 31,0	S = 32,07	Cl = 35,46	K = 39,13	Ca = 40,0
Differenz =	$\frac{89,1}{2}$ = 44,55	44,0	46,7	44,51	46,3	47,6
	—	As = 75,0	Se = 78,8	Br = 79,97	Rb = 85,4	Sr = 87,6
Differenz =	$\frac{89,1}{2}$ = 44,55	45,6 !	49,5	46,8	47,6	49,5
	Sn = 117,6	Sb = 120,6	Te = 128,3	J = 126,8	Cs = 133,0	Ba = 137,1
Differenz =	89,4 = 2.44,7	87,4 = 2.43,7	—	—	(71 = 2.35,5)	—
	Pb = 207,0	Bi = 208,0	—	—	(Tl = 204 ?)	—

Moderne Theorien (1864); hier nach K. Seubert (1895)

Courtesy G. Boeck

Atomic volume and atomic weight



L. Meyer:

Atomic volume = quotient of atomic weight and density

Shows periodic behavior

L. Meyer, Die Natur der chemischen Elemente als Function ihrer Atomgewichte, *Annalen der Chemie und Pharmacie*. VII. Supplementband **1870**, 354-364 (Fig. from *Moderne Theorien*, 1872)

Courtesy G. Boeck

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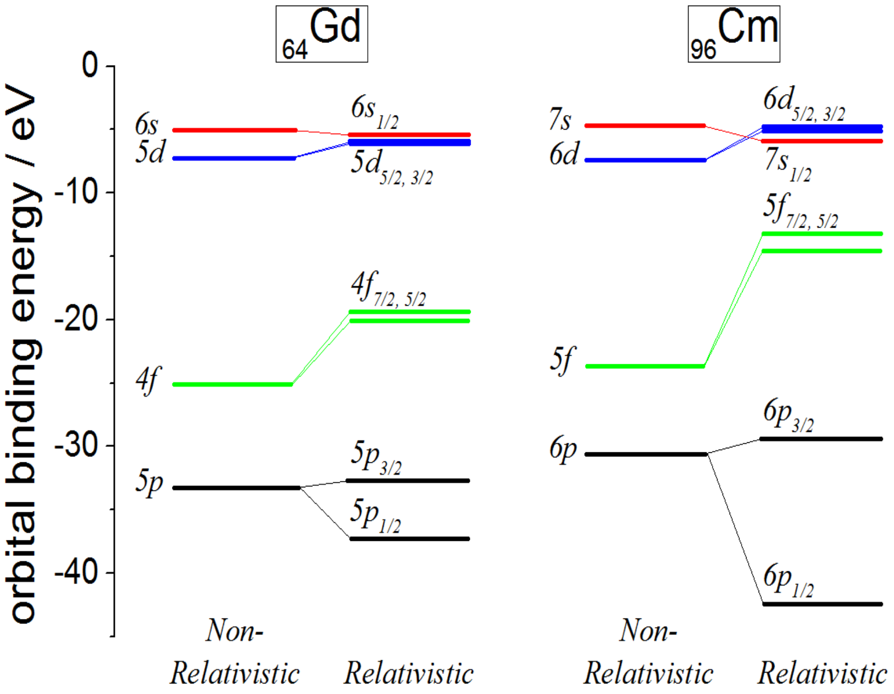
Relativistic Effects



J.P. Desclaux, At. Data Nucl. Data Tables **12**, 311 (1973)

Relativistic effects

- increase binding of $s_{1/2}$ and $p_{1/2}$ orbitals
- decrease binding of $p_{3/2}$, $d_{3/2}$, ... orbitals
- large Spin orbit splitting for high-*Z* elements

electron velocity according to Bohr's model: $v = Z\alpha/hc$

Gd: v = 0.5 c
Cm v = 0.7 c

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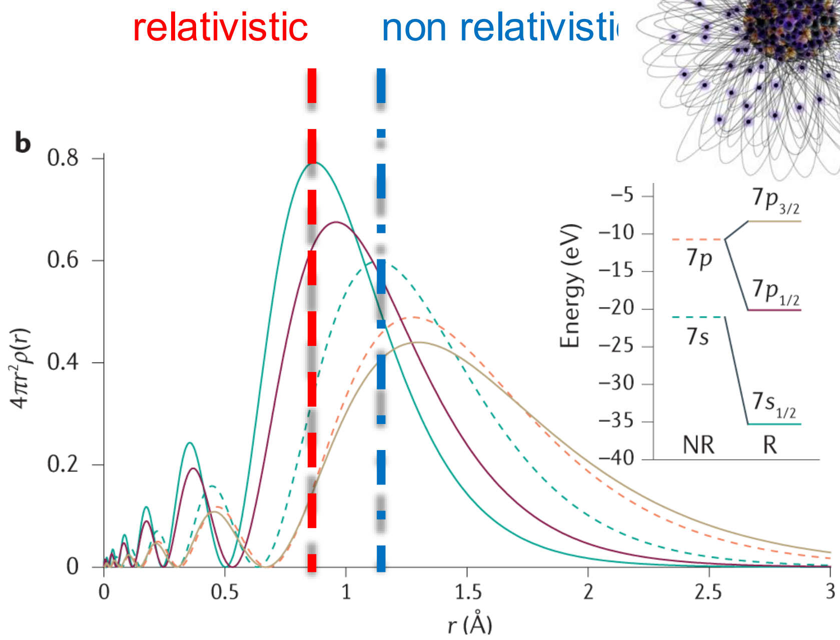
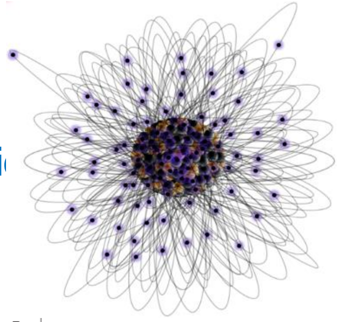
Atomic Structure of Heaviest Elements





atomic structure of heavy elements strongly influenced by relativistic effects

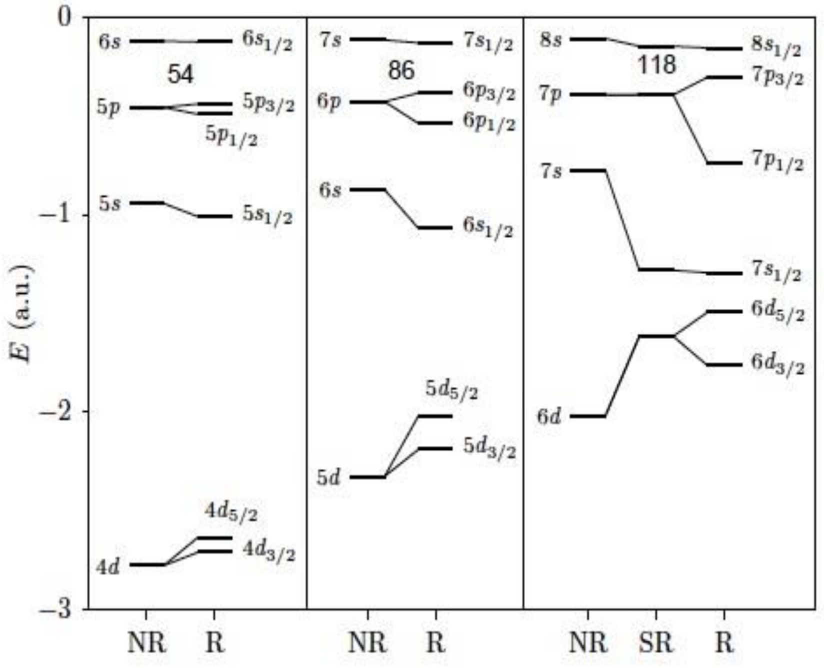
$Z\alpha \approx 1$: quantum electrodynamics (non-perturbative regime) gains in importance

- quantum many-body problem: significant influence of electron correlations
- all different effects intertwined
- accurate theoretical descriptions challenging

➢ benchmark theoretical predictions with comprehensive experimental data

O. Smits et al,. Phys. Rep. 1035 (2023) 1

Atomic Structure of Heaviest Elements



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- All different effects intertwined
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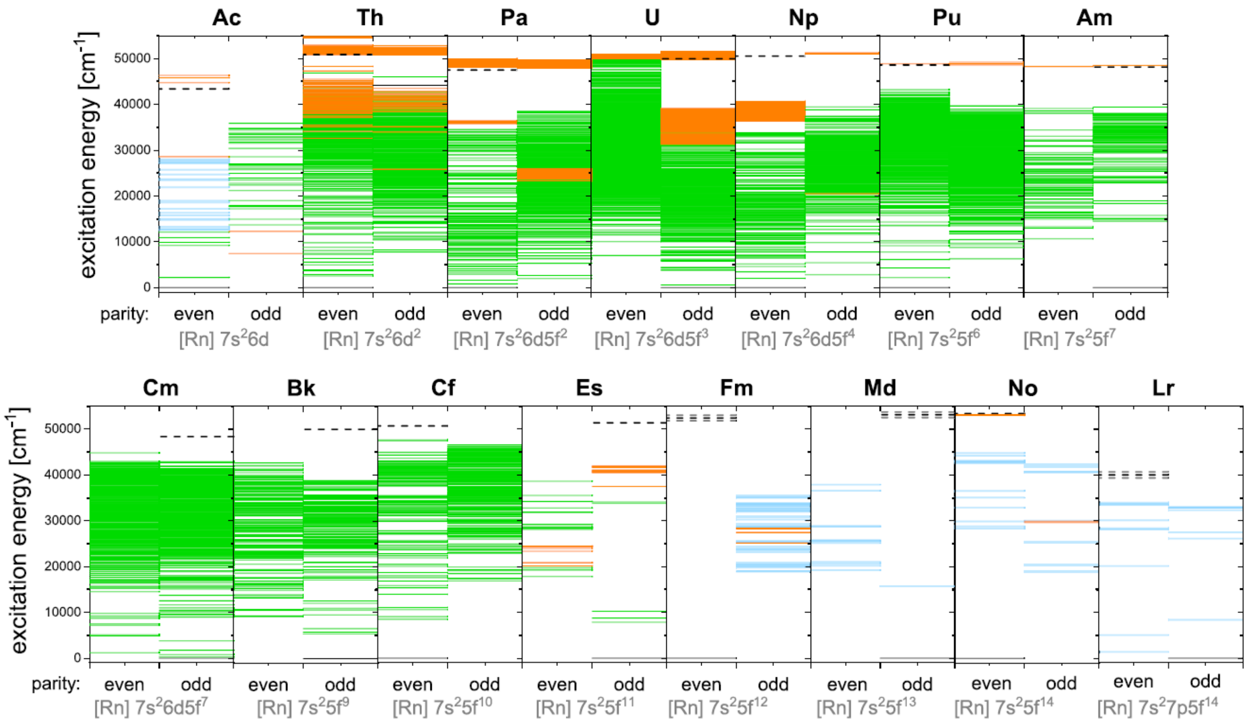
➢ Benchmark theoretical predictions with comprehensive experimental data

P. Jerabek et al., PRL **120,** 053001 *(2018)*

Atomic Level Data of Actinides



- complex atomic structure with many close-lying levels and open-shell systems
- limited experiment data for many actinide elements
- for heaviest actinides mainly theoretical predictions
- theoretical uncertainties about ≈ 300 cm^{-1}

Blaise 1992

Experimental data > 1992

Theory – calculated levels

MB et al., Prog. Nucl. Part. Phys. 116 (2021) 103834

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