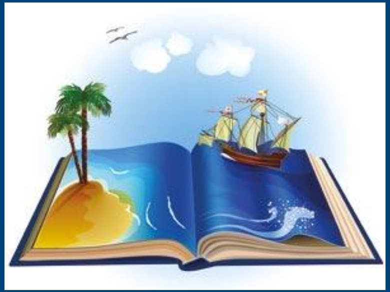
Ground-State Properties from Laser Spectroscopy
Part II: Methods and More



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Part II – Laser Spectroscopy for Nuclear Physics

- Requirements for spectroscopy
- Common laser spectroscopy methods applicable to radionuclides:
  - Collinear laser spectroscopy
  - Resonance ionization spectroscopy
- Production schemes for nuclides of interest
- Laser spectroscopy with gas cells

Hyperfine Splitting – Magnetic dipole part

Magnetic dipole Hamiltonian:

H_{hfs} = A I·J

Total angular momentum:

F = I (nuclear spin) + J (orbtial angular momentum L + spin S)

$F^2 = I^2 + J^2 + 2\, I \cdot J$

$$\mathbf{I}\cdot\mathbf{J} = \tfrac{1}{2}[F(F+1) - I(I+1) - J(J+1)]\hbar^2$$

Hyperfine Splitting - Magnetic dipole part

Insert into H_{hfs} : $E_F = \frac{A}{2}[F(F+1) - I(I+1) - J(J+1)] = \frac{A}{2}K$

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

allowed values: F = |I-J|, ..., I+J

Hyperfine Splitting - Electric Quadrupole Contribution

For $I > 1/2$ and $J > 1/2$:

$$E_Q = B \{ [3/4\ K(K+1) - I(I+1)J(J+1)] / [2I(2I-1)\ 2J(2J-1)] \} = B\ C$$

Total first-order energy:

$$E = (A/2)\ K + B\ C$$

Hyperfine Splitting – General Multipole Expansion

General HFS Hamiltonian: $$H_{\mathrm{hfs}} = \sum_k \mathbf{T}_e^{(k)} \cdot \mathbf{M}_n^{(k)}$$

First-order energy:

$E_F = \Sigma_k$ (-1)^(I+J+F) { I J F J I k } $\langle I||M^{(k)}||I\rangle$ $\langle J||T^{(k)}||J\rangle$

Multipoles:

k=1 Magnetic dipole → A
k=2 Electric quadrupole → B
k=3 Magnetic octupole → C
k=4 Electric hexadecapole → D

Hyperfine Splitting – General Multipole Expansion

General HFS Hamiltonian:

$$H_{\text{hfs}} = \sum_k \mathbf{T}_e^{(k)} \cdot \mathbf{M}_n^{(k)}$$

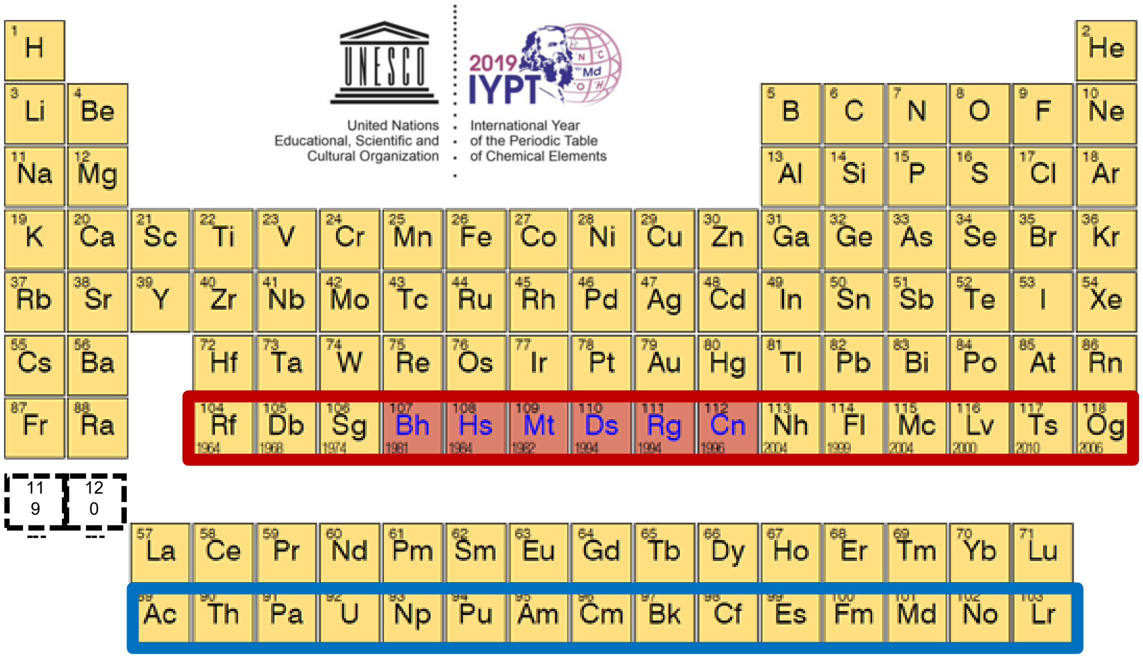
First-order energy:

$$E_F = \Sigma_k \, (-1)\text{^}(I+J+F) \quad \{\, I \; J \; F \; J \; I \; k \,\} \; \langle I||M^{(k)}||I\rangle \; \langle J||T^{(k)}||J\rangle$$

Multipoles:

For most stable nuclei, C and higher terms are **three to five orders of magnitude smaller than** A.

Today's Periodic Table of Elements



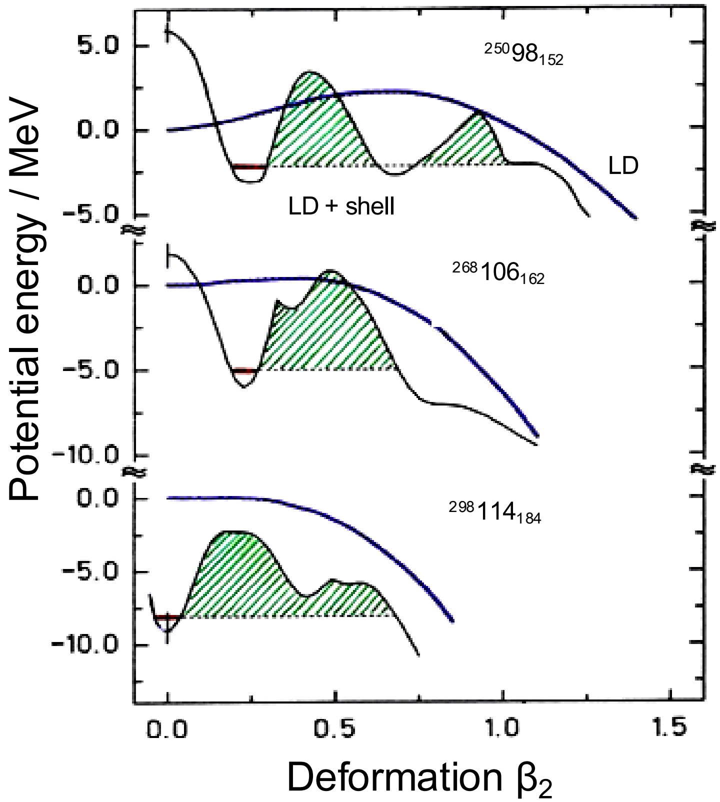
- established by Mendeleev, Meyer, and others about 150 years ago
- quantum mechanical description of atoms explains periodicity
- some deviations due to relativity

Transactinide Elements (Z>103)
= Superheavy Elements (SHE)

Actinide Elements

elements **Bh, Hs, Mt, Ds, Rg, Cn** with Z=107-112 discovered GSI Darmstadt, Germany

Stability of Superheavy Nuclei



- Existence of heavy nuclei depends on interplay of repulsive Coulomb and attractive nuclear force
- Heavy nuclei often deformed with fission barrier decreasing with increasing Z
- Nuclear shell effects stabilize superheavy nuclei (binding energy gain up to 10 MeV)

➢ **superheavy nuclei owe their very existence to nuclear shell effects**

Calculations by A. Sobiczewski, figure courtesy of S. Hofmann

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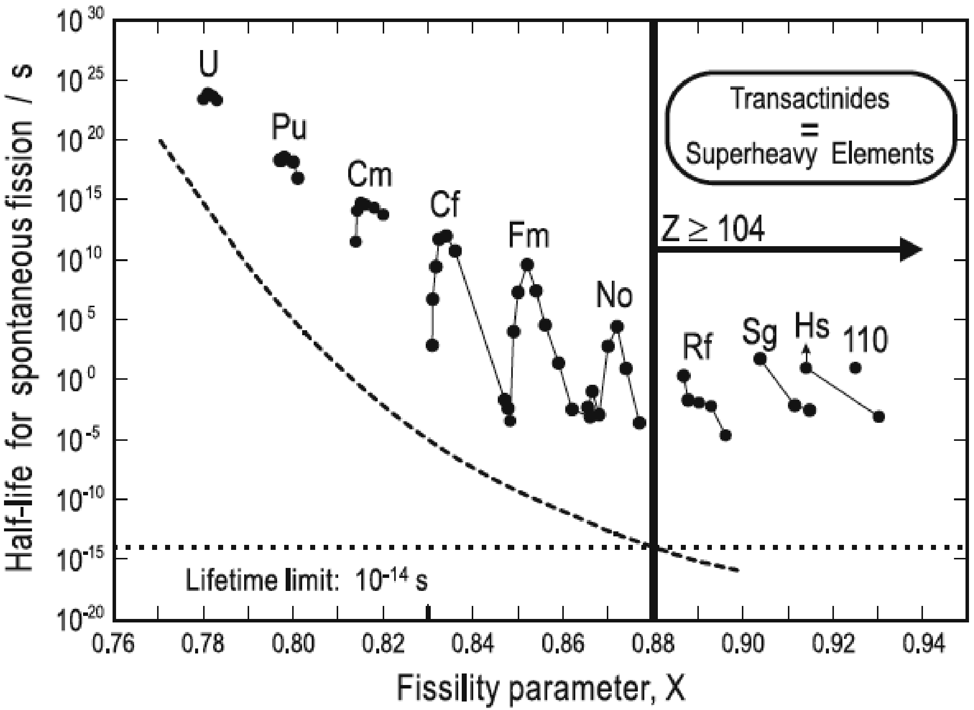
Definition of (Superheavy) Elements



- International Union of Pure and Applied Chemistry (IUPAC) requirement for an element's existence:

  *For the discovery of **new elements**, IUPAC also requires experimental evidence that a nuclide with a previously unknown atomic number exists for at least **10^{-14} seconds**, long enough for it to acquire an electron cloud and exhibit chemical identity.*

- According to liquid drop model, nuclei with atomic numbers Z > 104 will fission
- Nuclear shell effects stabilize SHN against disintegration by fission

Predictions in 1970s: Island of Stability

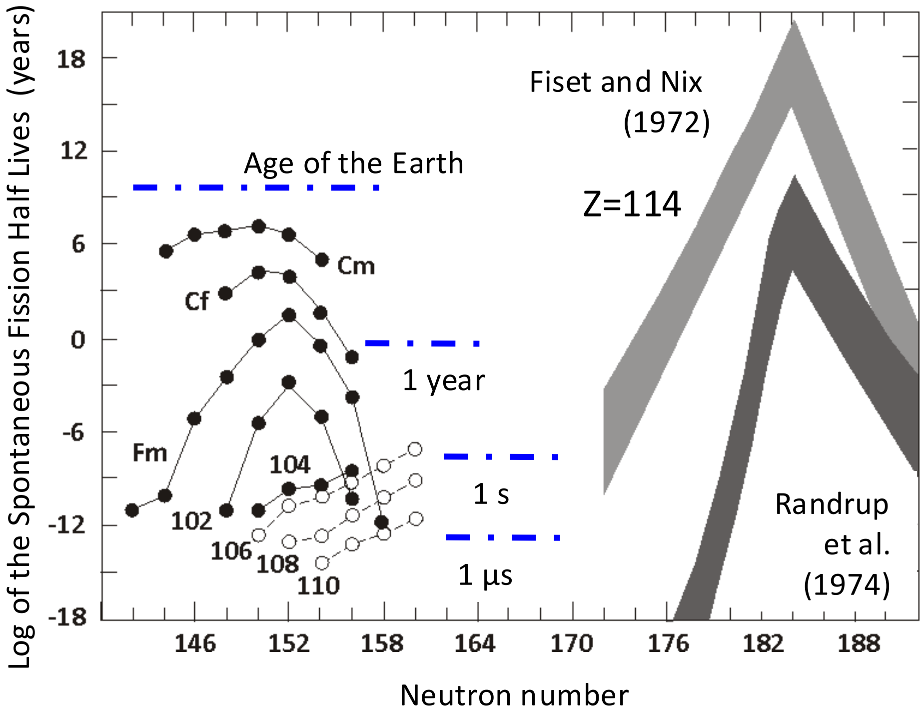
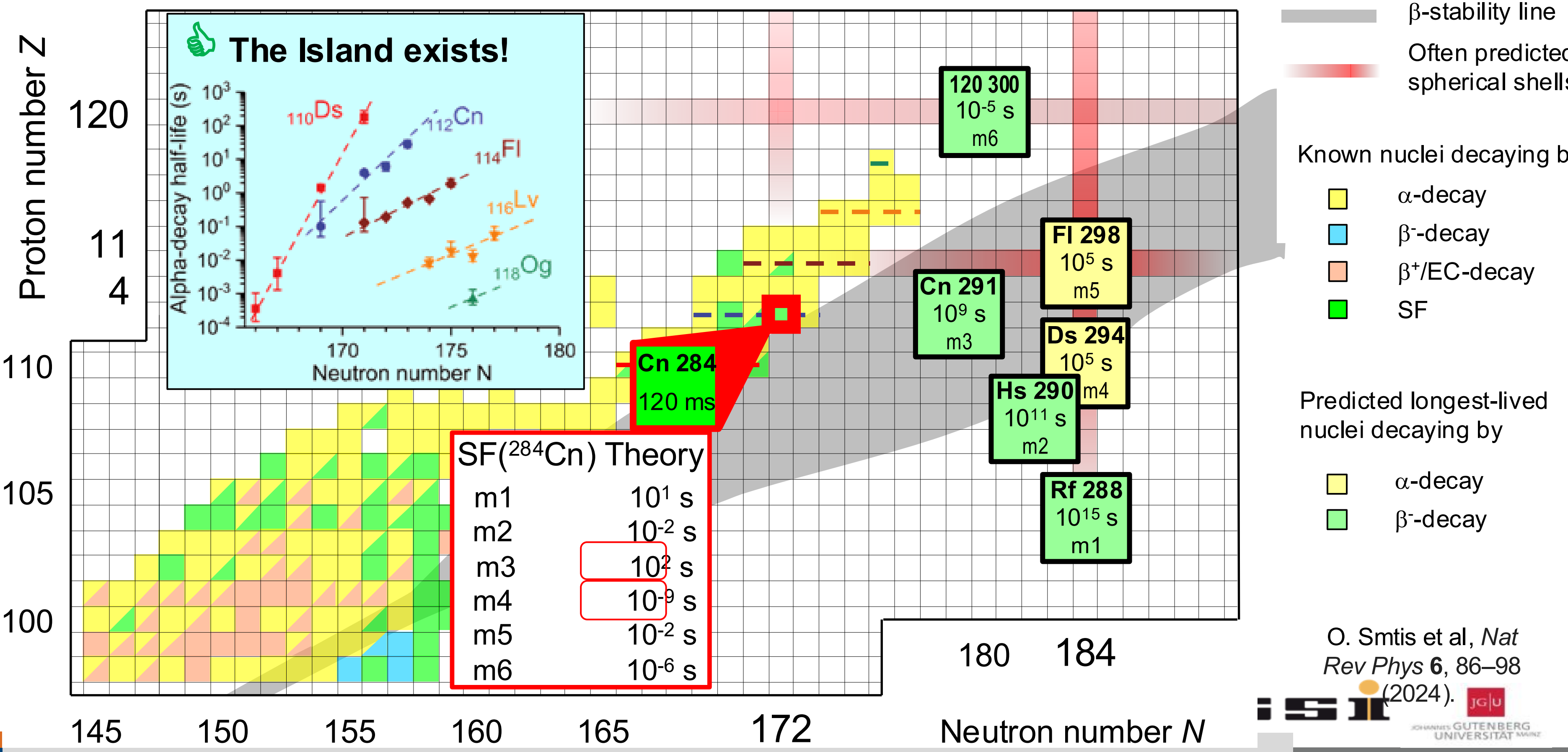


Figure Courtesy Yuri Oganessian

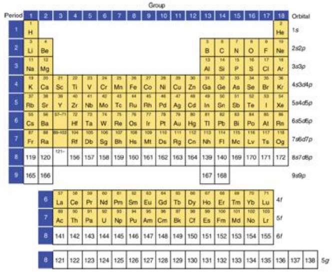
- superheavy nuclei with Z ≈ 114 and N ≈ 184 predicted to form **„Island of Stability“**
- theoretical models predict: $T_{1/2}(SF) > 10^9$ years
- initiate search for existence in nature
  – until now no evidence
- Today's models predict island of *enhanced* stability exists, but longest lifetime no more than weeks

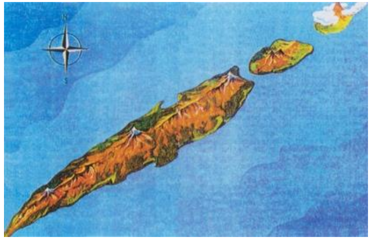
Island of Stability – Status Today



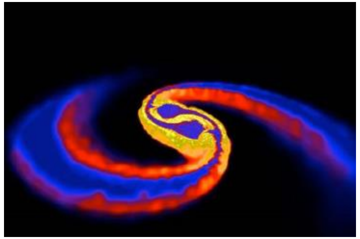
O. Smtis et al, *Nat Rev Phys* **6**, 86–98 (2024).

Superheavy Element Research – Open Questions





- Where is the end of the periodic table in atomic number and mass?
- What are the boundaries of the *island of enhanced stability* and what are the properties of nuclei there?
- How does relativity affect the architecture of the periodic table?
- Are there remnants of long-lived superheavy elements on earth?



See e.g. recent reviews by
O. Smits et al. Nat. Rev. Phys. (2024), https://doi.org/10.1038/s42254-023-00668-y
S.A. Giuliani et al., Rev. Mod. Phys. 91, 011001 (2019)
and special/topical issues on SHE in Nucl. Phys. A 944 (2015) and Eur. Phys. J. A

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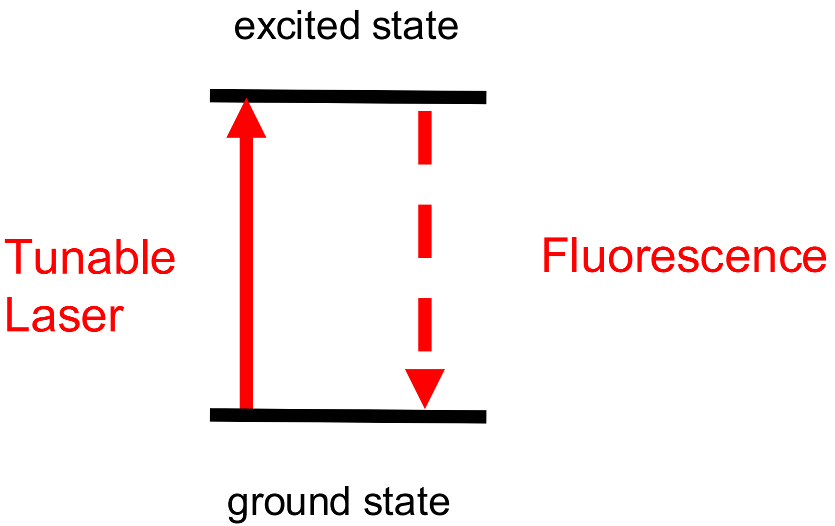
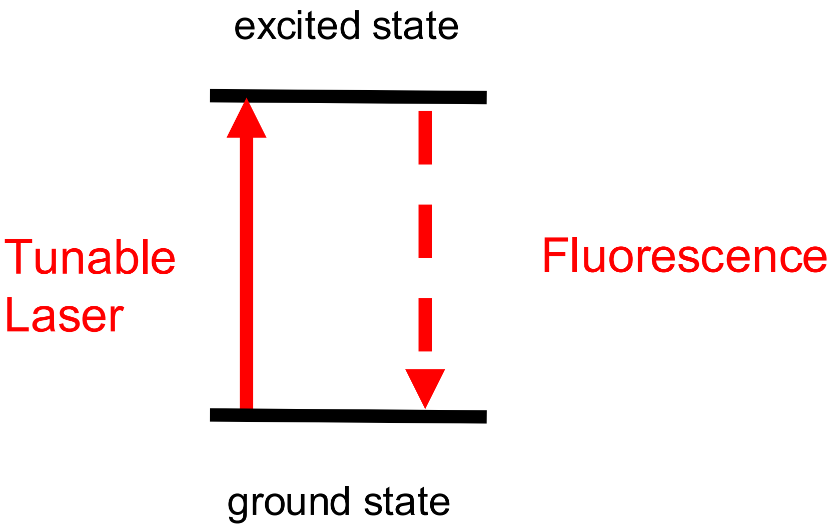
Considerations on Detection



- excite electron by tunable laser light
- deexcitation by photon emission
- typical lifetimes of atomic states on the order of 1-10 ns

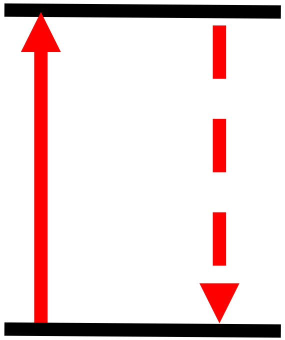
Basic Principles



estimate number of scatted photons:

- $1/\tau$ for $\tau=10$ ns: 10^8 photons / second
- detection of light limited by detector efficiency and solid angle coverage
- typical efficiencies: 10^{-3} - 10^{-4}
- corresponds to 10,000 photons / atom

Basic Principles



Challenges:

- background from scatted laser light
- needs suitable transitions
- typically start from neutral atoms
- transitions in ions often less accessible for practical reasons (wavelength)

Spectroscopy Requirements

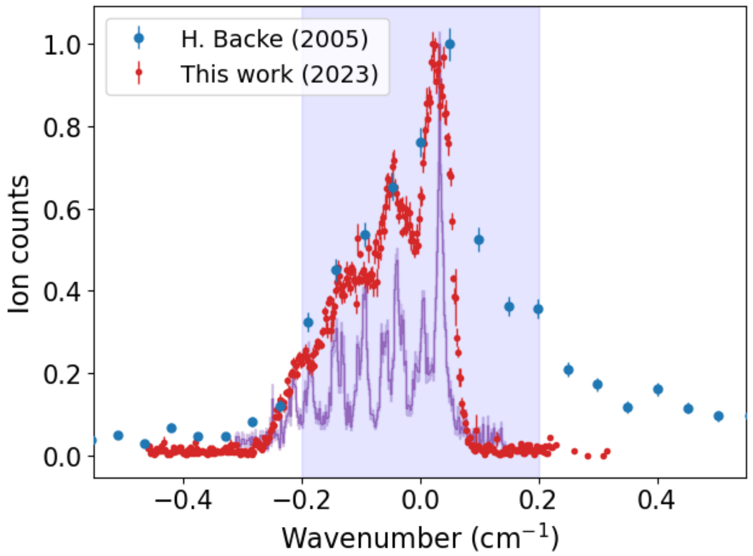
Obtain (nuclear) properties for hyperfine structure:

- Hyperfine splitting: of the order of 10–1000 MHz
- ionic (atomic) transitions in the visible frequency range (~10^{15} Hz)
- require very high spectroscopic resolution, better than 10^{-7}–10^{-8}.

Additional requirements for radionuclides

- provide low-energy ion beams with suitable parameters (energy spread, purity …)
- avoid/reduce broadening mechanisms
- high sensitivity and high efficiency

Example - ^{255}Fm hyperfine spectrum



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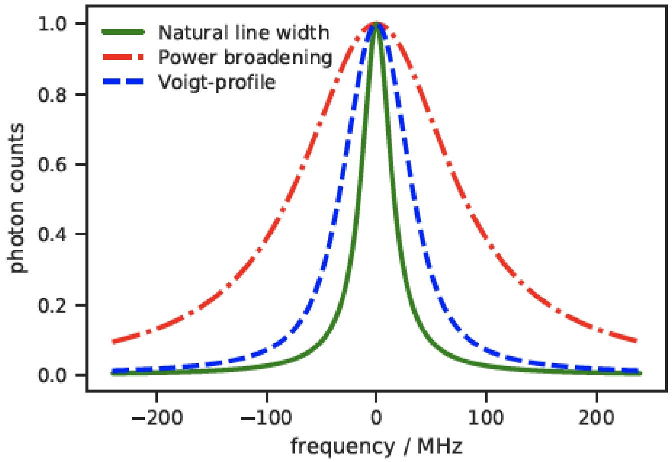
Spectral Resolution – Linewidth and Broadening



Natural linewidth: $$\delta\nu_{\text{natural}} = \frac{\Delta E_i}{h} = \frac{1}{2\pi\tau_i},$$

- nat .linewidth typical dipole transition 10s of MHz
- pulsed lasers have linewidth of 1-3 GHz
- cw lasers / seeded lasers linewidth about <100 MHz

- Doppler broadening (hot cavity)
- pressure broadening (gas cell)
- narrowband laser systems for high spectral resolution
- pulsed lasers for high power to saturate transitions

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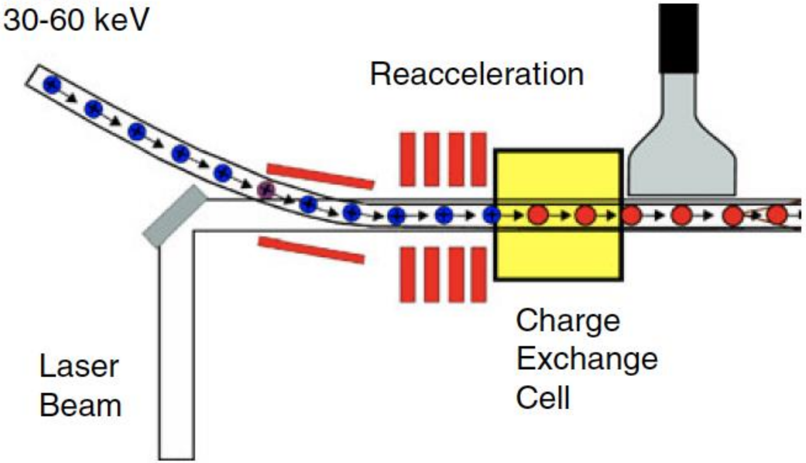
Collinear Laser Spectroscopy



Accelerate ion beam with energy spread dE

$\delta v = \delta E / \mathrm{sqrt}(2mE) = \delta E / \mathrm{sqrt}(2mqU)$

Transition frequency (collinear / anti-collinear)

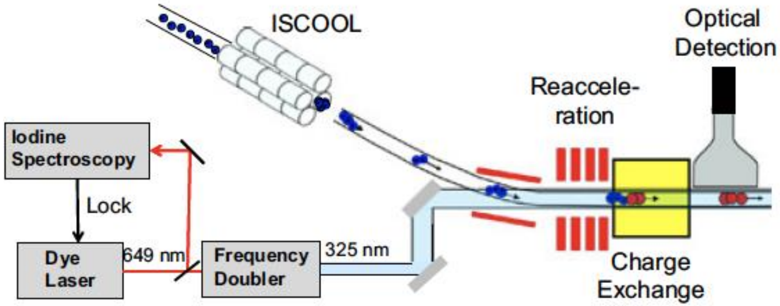
$$\nu_{c/a} = \nu_0 \frac{1}{\gamma(1 \mp \beta)} = \nu_0 \gamma (1 \pm \beta) .$$

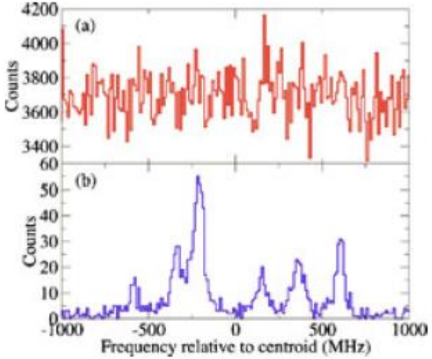
$$\nu_{c/a} = \nu_0 \left[1 + \frac{qU_{tot}}{mc^2} \left(1 \pm \sqrt{1 + 2\frac{mc^2}{qU_{tot}}} \right) \right]$$

- Neutralization of ion beam by charge exchange
- Excitation of ions in collinear geometry
- Observation of fluorescence (or radioactive decay)

S. L. Kaufman, Opt. Commun. **17**, 309 (1976); K. -R. Anton et al., Phys. Rev. Lett. **40**, 642 (1978)

Collinear Laser Spectroscopy - Bunched Beams





Non-gated

Gated on ion bunch

- reduce laser background by using bunched beams, introduced at JYFL, see A. Nieminen et al. Phys. Rev. Lett. **88**, 094801 (2002)
- benefit from radiofrequency beam cooler and buncher devices
- still requires large yield of 10^3-10^4/s

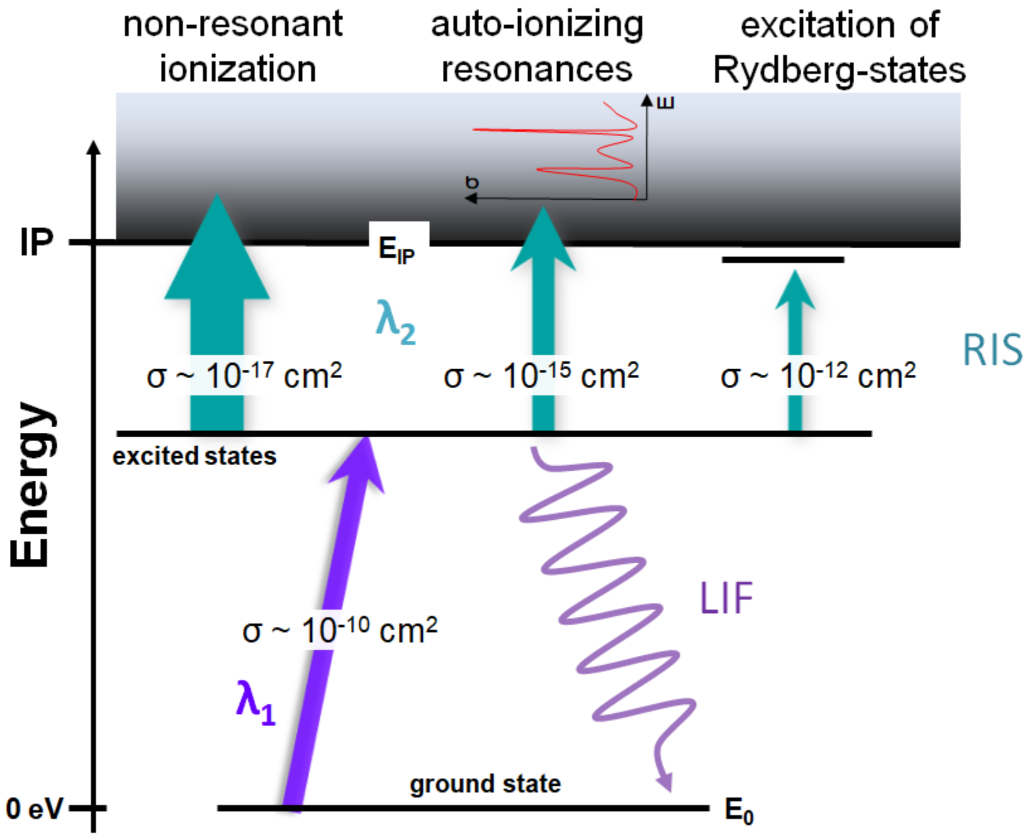
Advantages of CLS

- Reduced Doppler broadening.
- Long interaction region.
- Small beam divergence.
- High sensitivity and spectral resolution.

Problems:

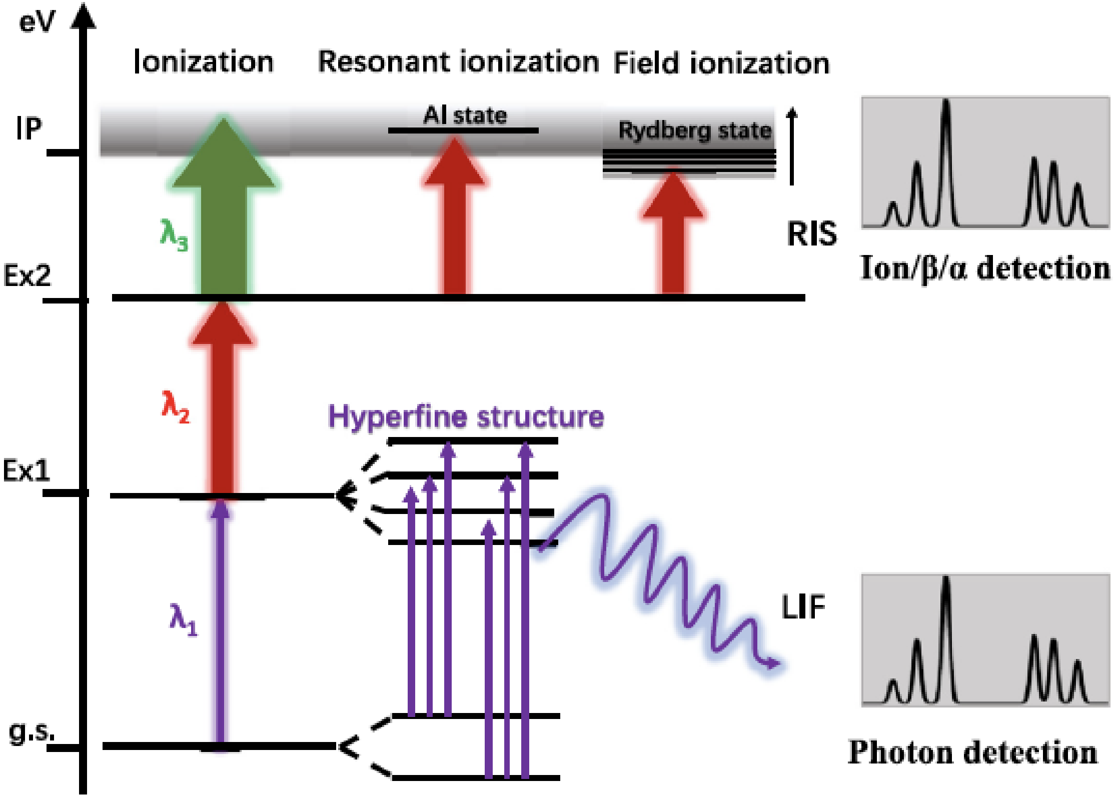
- background from scattered laser photons (fluorescence detection)
  - can be mitigated using ion-beam cooler bunchers
- and beam-energy (Voltage stability) and energy spread affect frequency measurement
  - can be mitigated with collinear/anti-collinear measurements

Principles of Resonant Ionization Laser Spectroscopy



- ion detection more efficient than fluorescence photon detection
- low-background conditions, particularly for detection via decay
- sensitive method applied in ultra-trace analysis and in laser-ion sources

Resonance Ionization Laser Spectroscopy (RIS)



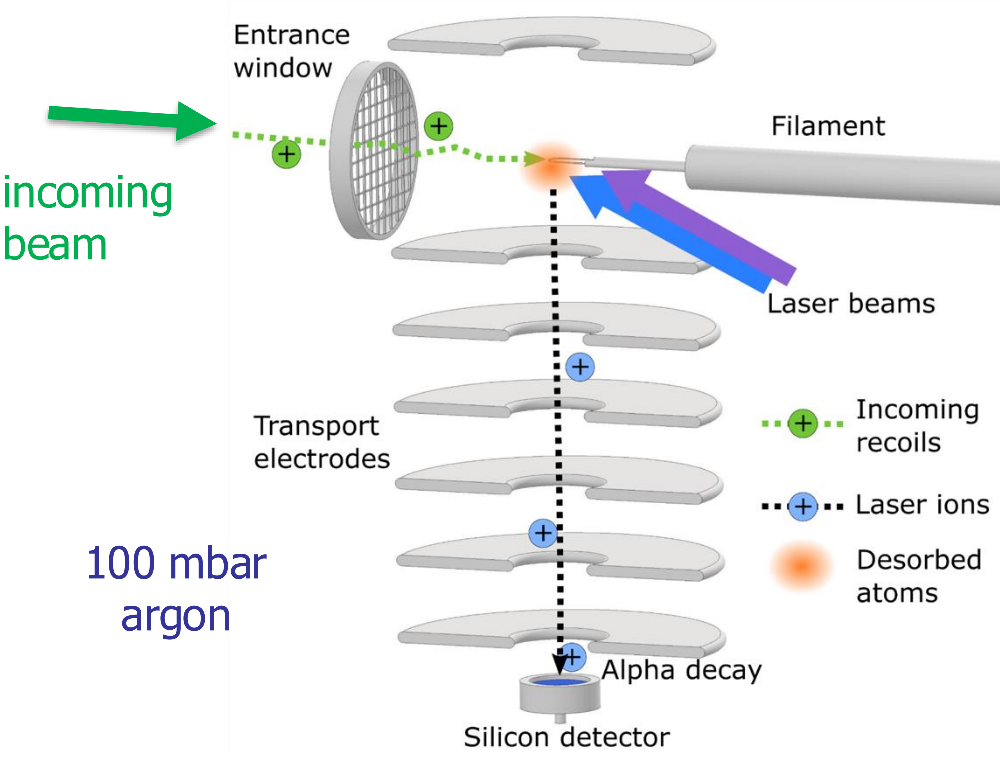
RIS method features:

- high sensitivity
- high selectivity
- high efficiency

challenges for (super)heavy elements:

- often no stable or long-lived nuclides (for developments)
- low yield and short half-life

Radiation Detected Resonance Ionization Spectroscopy



tailored method for measurements of actinide isotopes produced by fusion reactions with lowest rates:

- slow down ions in Ar gas
- neutralize on filament
- re-evaporate atoms
- two-step photo-ionization
- transport laser ions to detector and register radioactive decay

H. Backe et al. Eur. Phys. J. D, 45 (1) (2007), 99
F. Lautenschläger et al. Nucl. Instrum. Meth. B, 383 (2016),115
J. Warbinek et al., Atoms (2022)