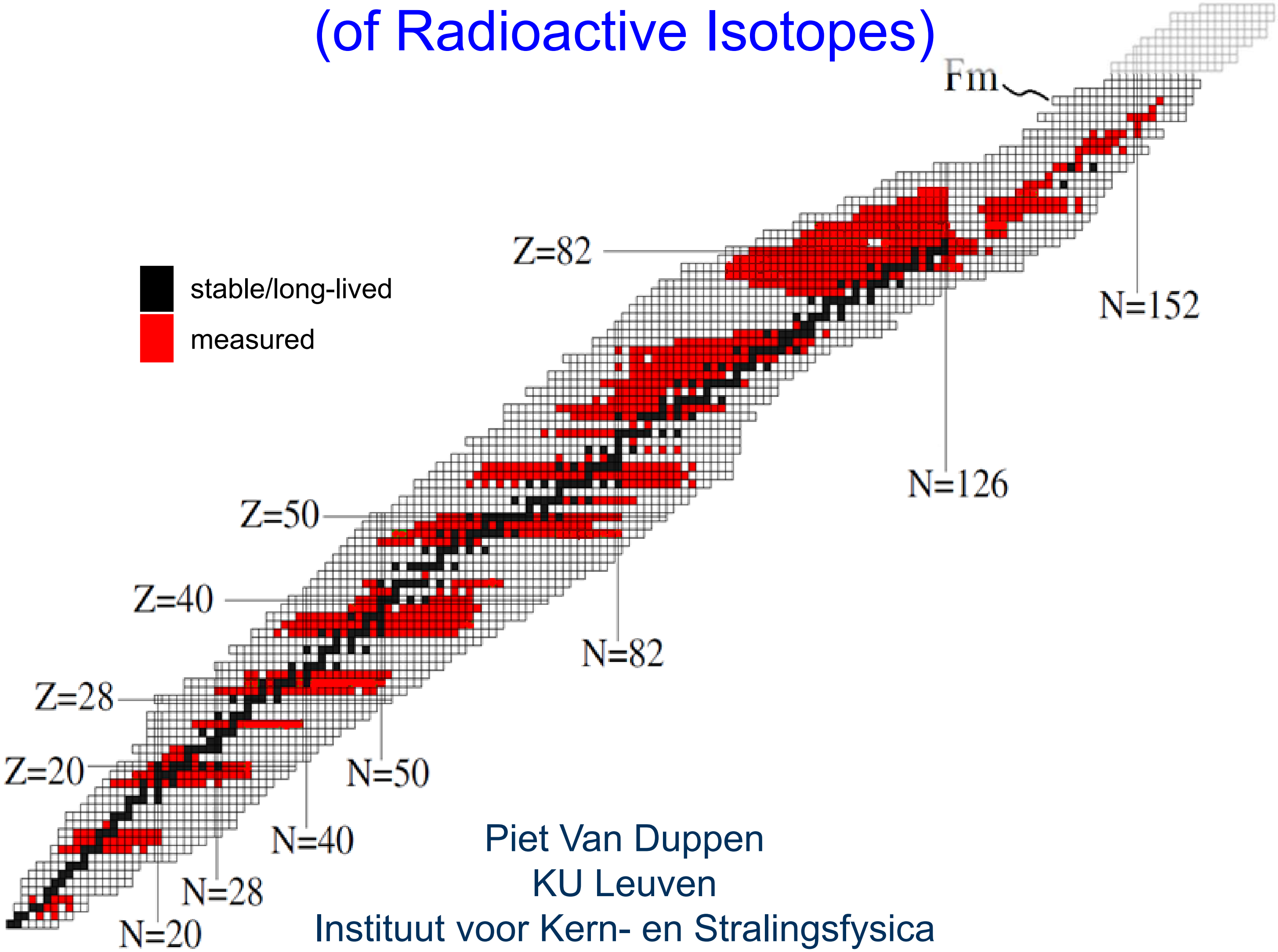
# Introduction to Laser Spectroscopy
# (of Radioactive Isotopes)




Piet Van Duppen
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Instituut voor Kern- en Stralingsfysica
Belgium

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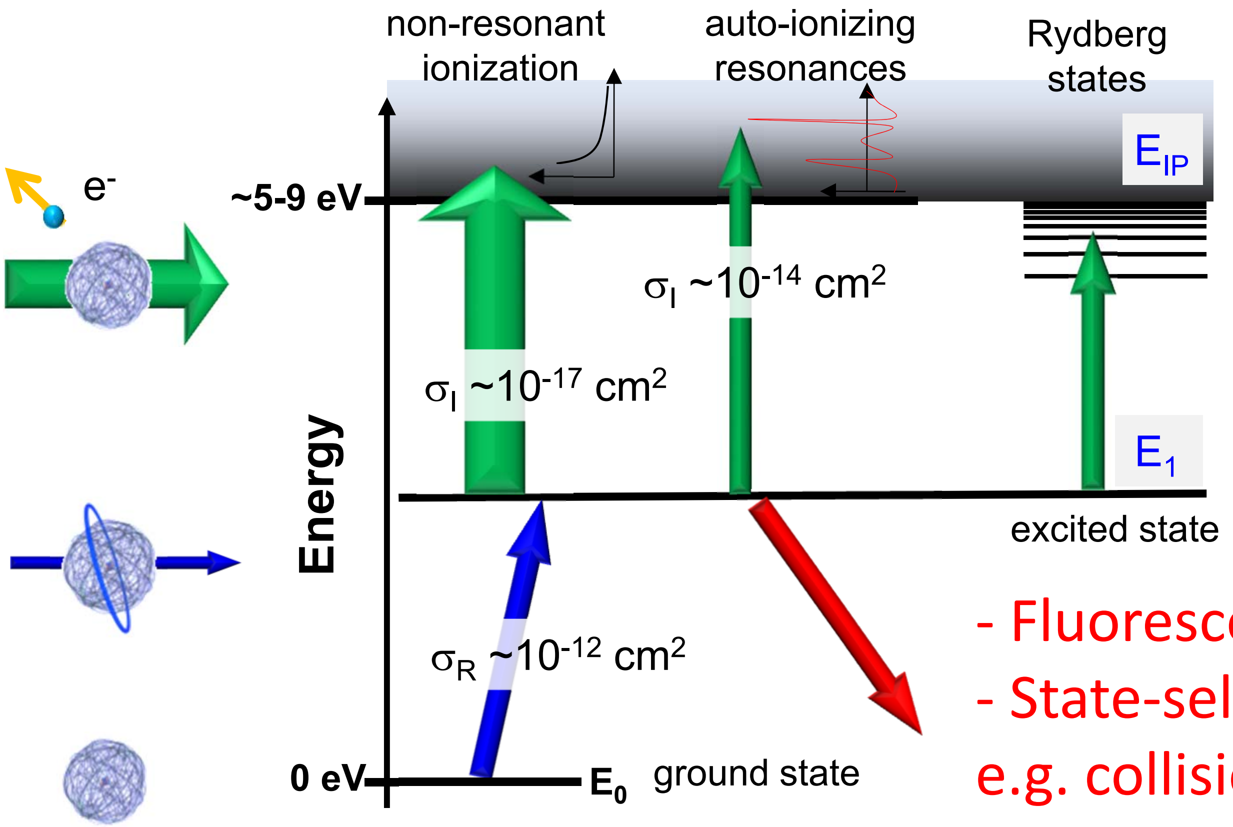
# Outline

- Laser spectroscopy: basics and observables
- Implementation in radioactive ion beam research
- Impact in:
  - Atomic Physics
  - Nuclear Physics
  - Fundamental Interaction Studies
- Developments to improve resolution, efficiency, selectivity and delay losses
- Conclusion and Outlook

Atomic physics techniques for studying nuclear ground state properties, fundamental interactions and symmetries: status and perspectives

H.-J. Kluge Hyperfine Interact (2010) 196:295–337

# Laser Spectroscopy: basics




- Ionization
  - Ion counting
  - Radioactive decay

- Fluorescence
- State-selective process
e.g. collisional re-ionization
- Laser-RF double resonance
- …



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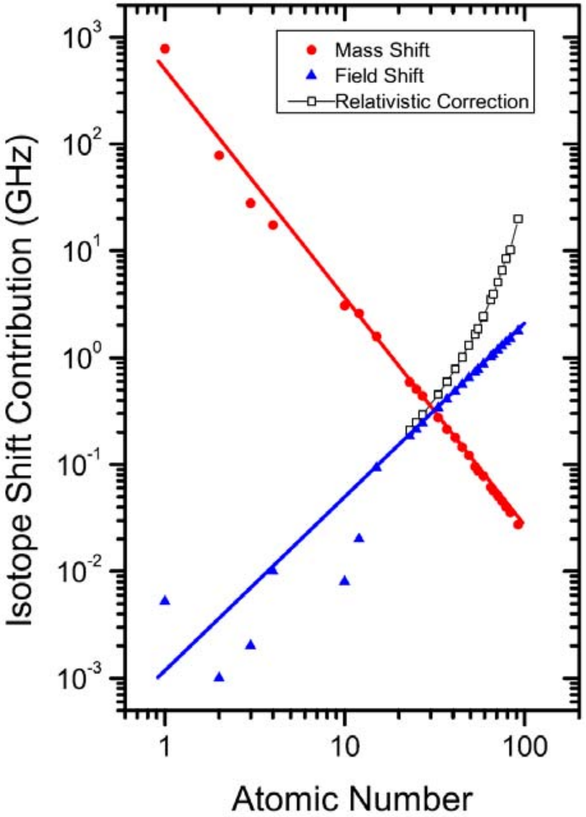
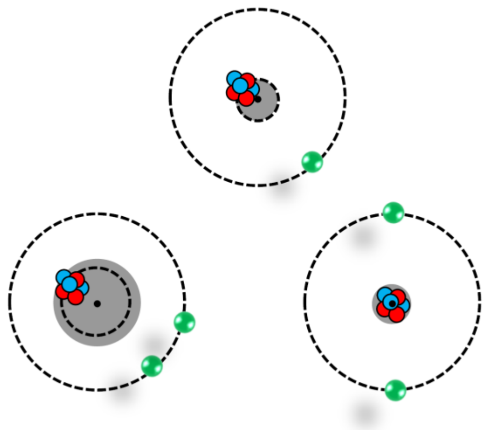
# Laser Spectroscopy: observables

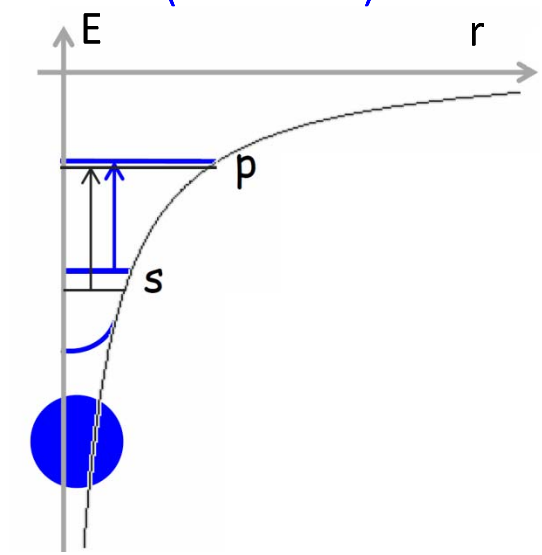

Isotope Shift

$$\delta\nu_{\mathrm{IS}}^{AA'} = K_{\mathrm{MS}} \cdot \frac{M_{A'} - M_A}{M_A M_{A'}} + \frac{2\pi Z e}{3} \Delta \left|\Psi(0)\right|^2 \ \delta \left\langle r^2 \right\rangle^{AA'}$$

Mass shift
(center of mass motion)



Field shift
(finite size)

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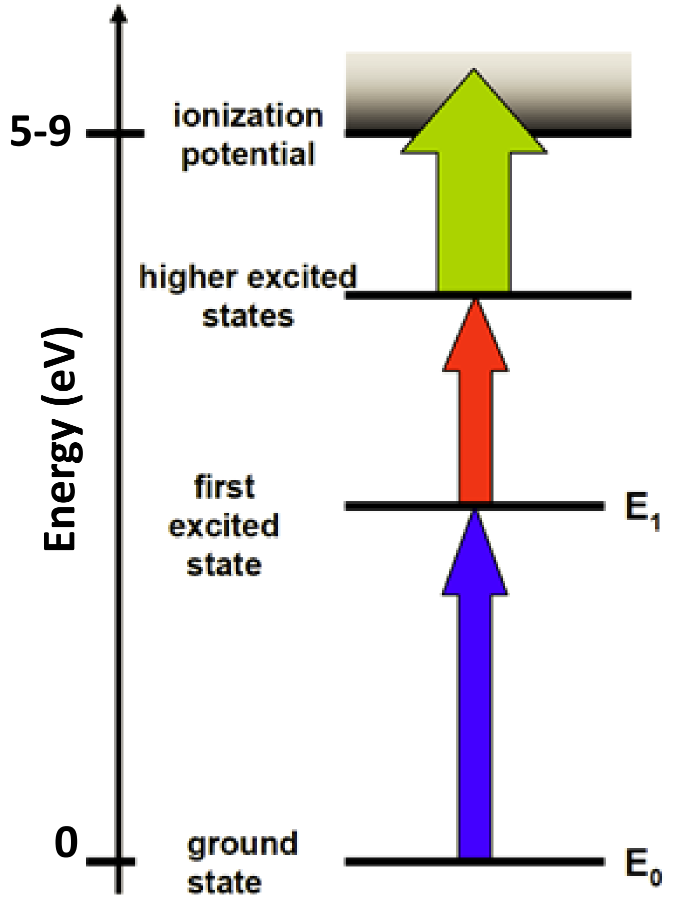
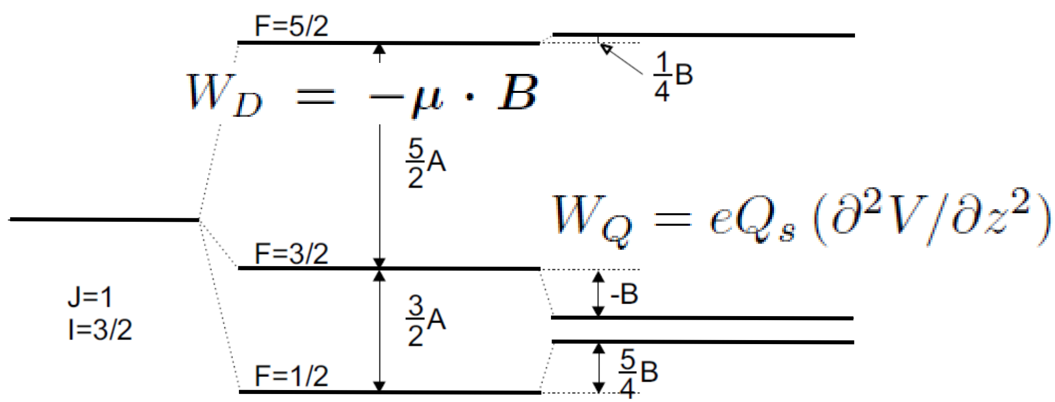
# Laser Spectroscopy: observables




$$\vec{F} = \vec{I} + \vec{J} \quad \left( |I - J| \le F \le I + J \right)$$

## Hyperfine Splitting




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

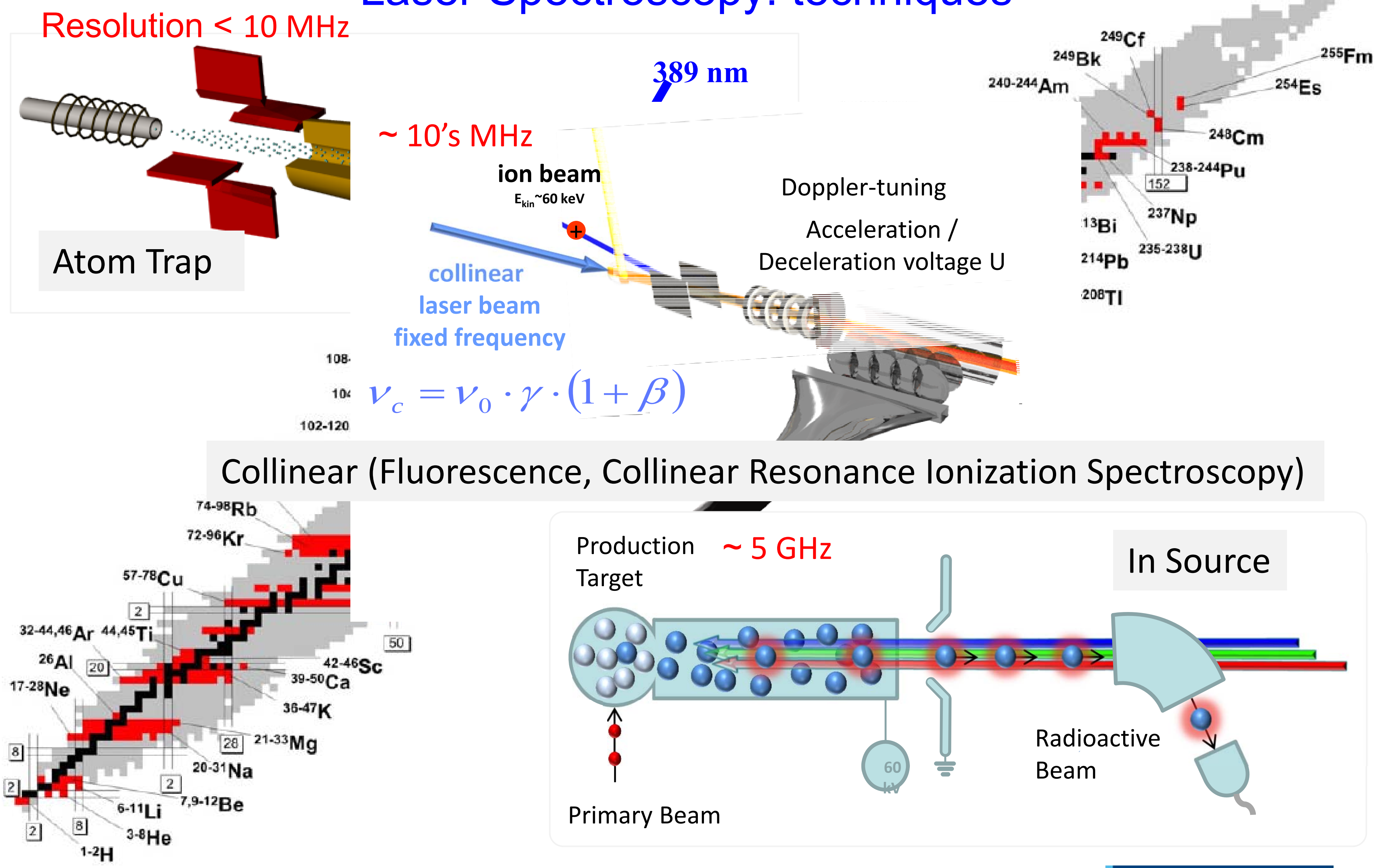
$$C = F(F+1) - I(I+1) - J(J+1).$$

$$A = \mu_I B_e(0) / (IJ)$$

$$B = eQ_S V_{ZZ}(0)$$

courtesy I. Moore - G. Neyens and R. Neugart “The Euroschool Lectures on Physics with Exotic Beams LNP 700 (2006)

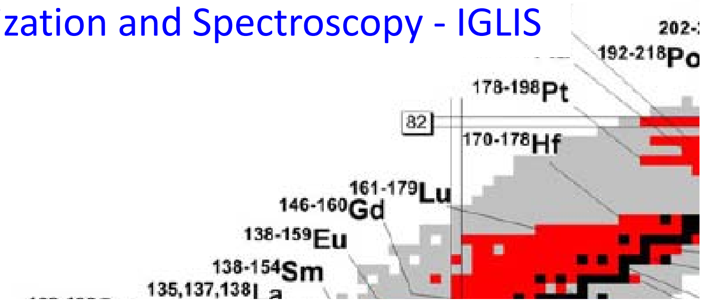
# Laser Spectroscopy: techniques

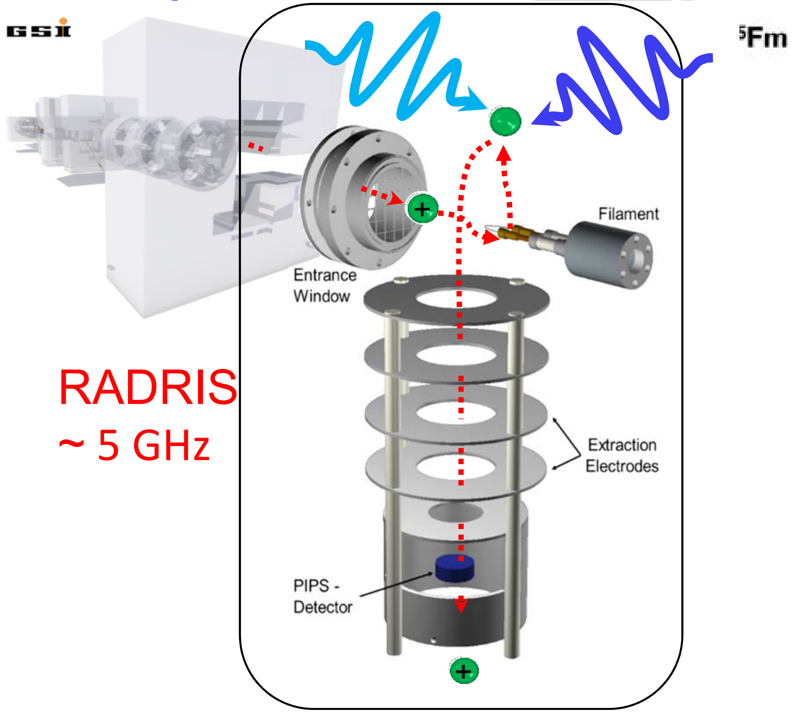


# Laser Spectroscopy: techniques

## In Gas Cell / Gas Jet

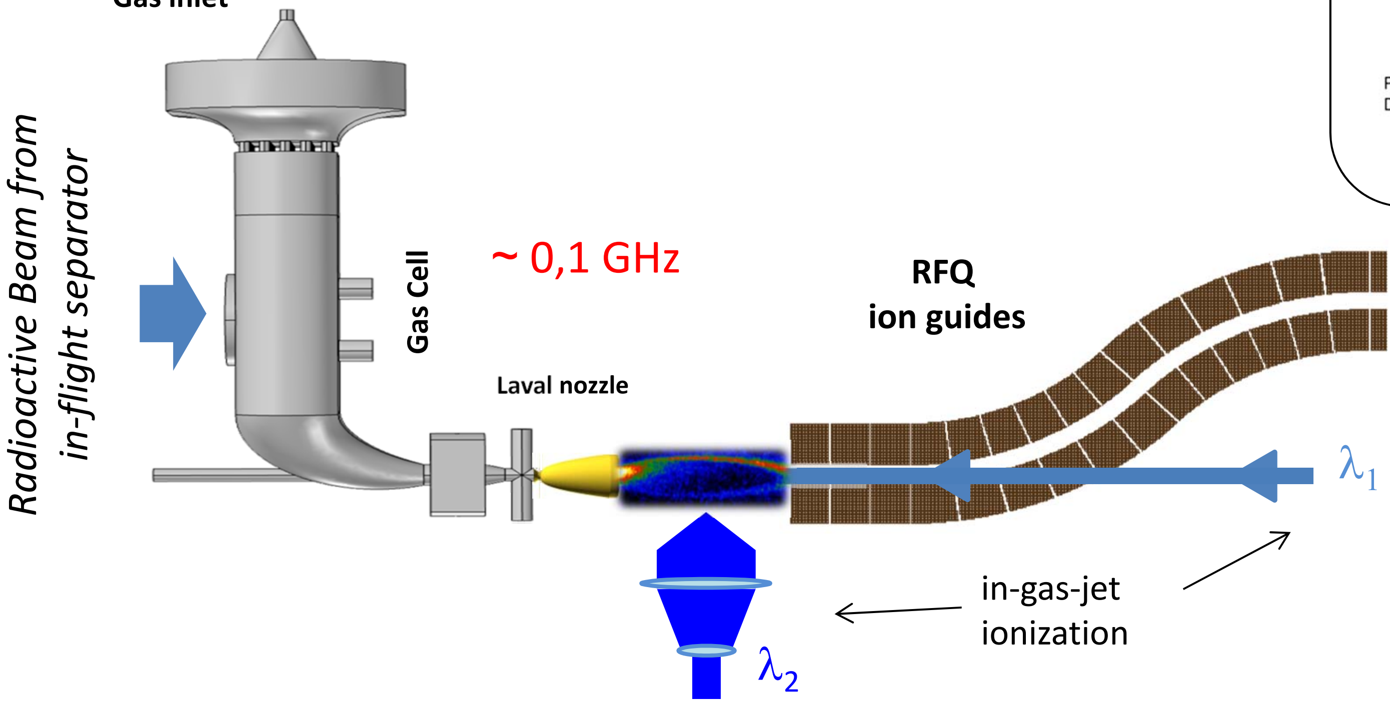
- In-Gas Laser Ionization and Spectroscopy - IGLIS




RADRIS
~ 5 GHz

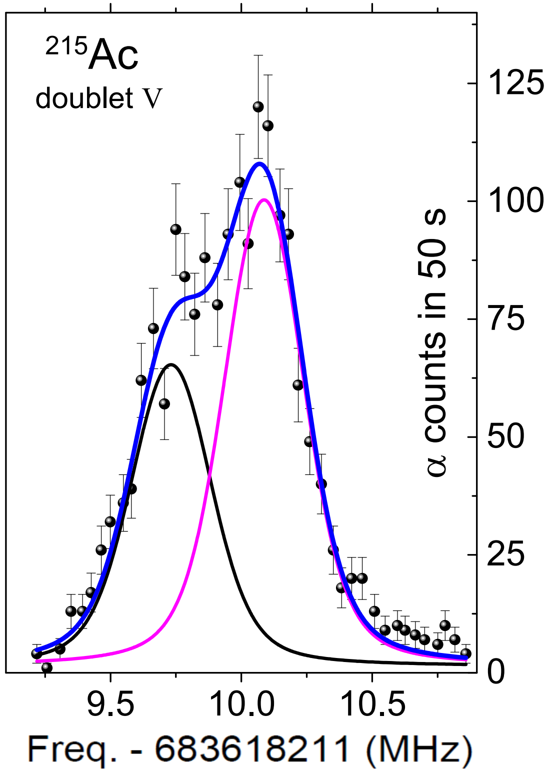
~ 0,1 GHz

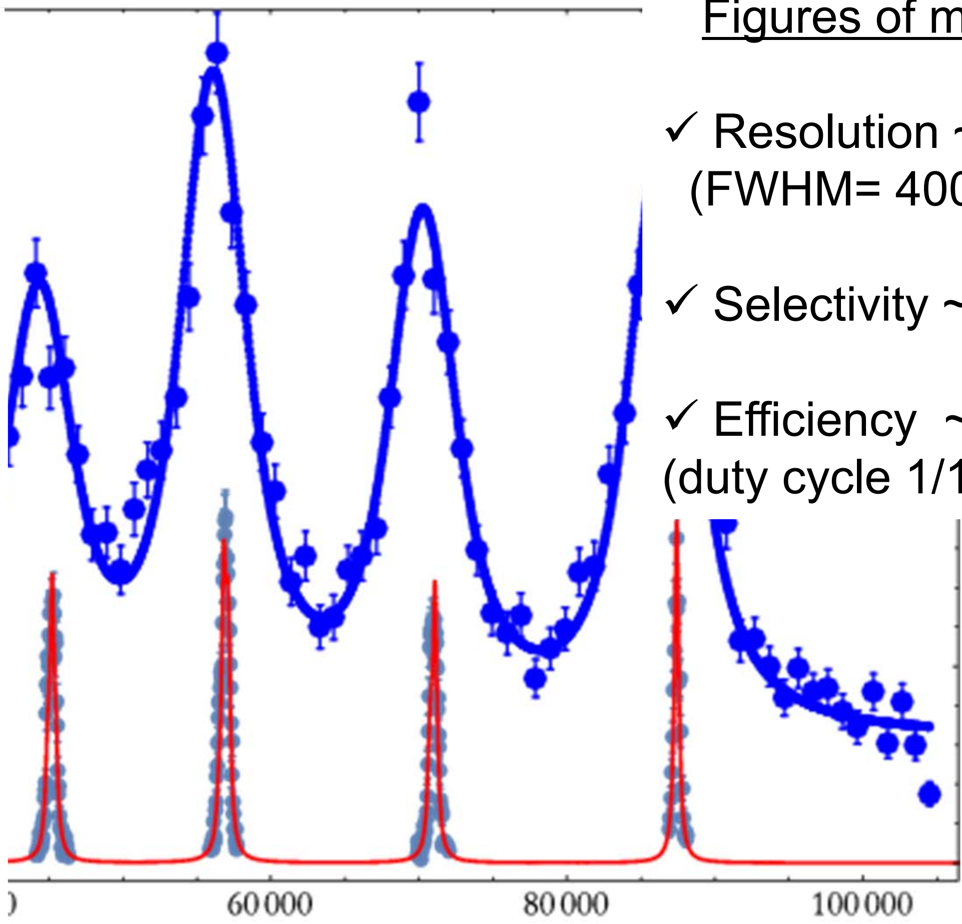


# LISOL's famous last action (*)

## * 1/5/1974 - † 6/12/2014

$^{215}$Ac $T_{1/2}$ = 0.17 s $J_\pi = (9/2^-)$




Figures of merit:

- ✓ Resolution ~ 5 $10^{-7}$ (FWHM= 400 MHz)
- ✓ Selectivity ~ 200
- ✓ Efficiency ~ 0.5% (duty cycle 1/10)

**- in gas cell** **- in gas jet**

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(*) courtesy Mark Huyse R. Ferrer,- Nature Comm. 8 (2017), Yu. Kudryavtsev et al, NIMB61 (2016)

# Laser Spectroscopy Observables - impact

- Atomic physics
  - Especially in the (super) heavy element region – ionization potential, atomic levels, transition strengths
  - Ex.: actinium, nobelium

  (M. Bissel, D. Hanstrop, M. Laatiaoui, D. Yordanov)

- Nuclear physics
  - $\delta<r^2>$, $\mu$, $Q_s$ and $I^\pi$
  - Nuclear-model independent provided atomic physics is known
  - Ex. : density profile in nobelium, shape coexistence in Hg isotopes, Ca charge radii

  (K. Flanagan, K. Minamisono, I. Moore, P. Müller, G. Neyens, W. Nörtershäuser, D. Yordanov, M. Bissell)

- Fundamental physics
  - Use of the high (ultimate) precision
  - Molecular ions

  (R. Garcia Ruiz, I. Moore , P. Müller)

- Instrumentation developments
  - Obtaining high efficiency (RIB), improved precision, sensitivity

  (K. Flanagan, R. Ferrer, N. Lecesne, D. Verney, W. Nörtershäuser)

# Nuclear Moments of $Ac^{227}$†

MARK FRED AND FRANK S. TOMKINS, *Chemistry Division, Argonne National Laboratory, Lemont, Illinois*

AND

WILLIAM F. MEGGERS, *National Bureau of Standards, Washington, D. C.*

(Received April 11, 1955) Phys. Rev. 98, 1514

The values derived for the moments from the conventional treatment of hfs in intermediate coupling are $+1.1$ nm and $-1.7\times10^{-24}$ cm². The experimental error is believed to be less than 10 percent, but it is difficult to estimate the total error because of the configuration interaction and the large relativity corrections. No correction for closed shell distortion was made.

It is hoped that improved values can be obtained, but meanwhile it appears useful to offer the present results. We should like to acknowledge helpful discussions with Dieter Kurath and R. E. Trees.

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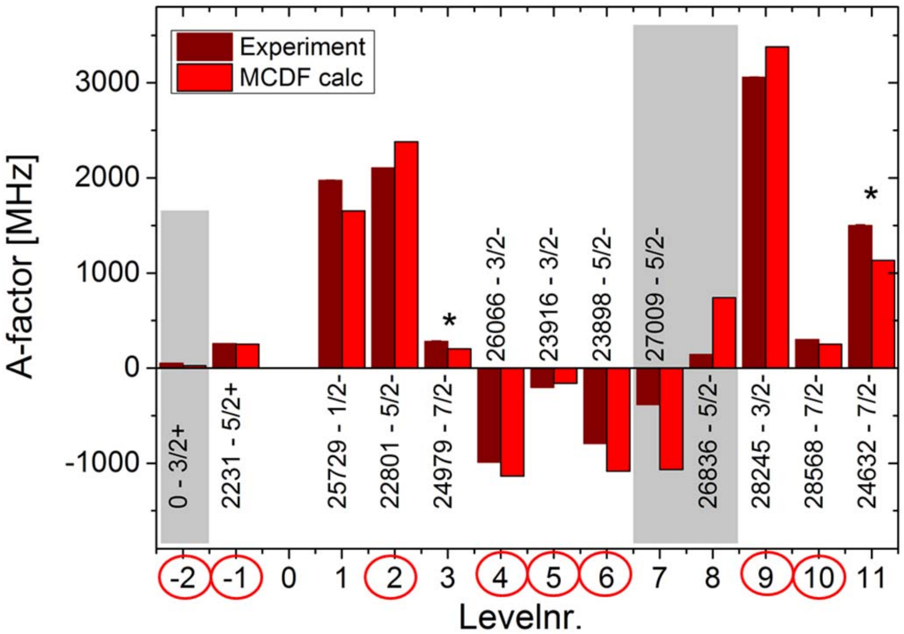
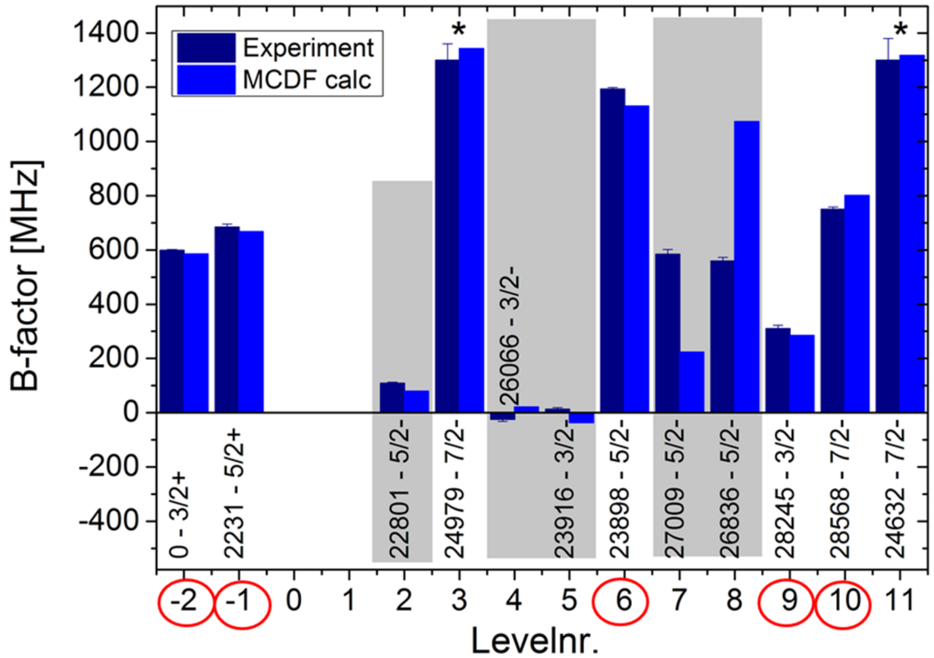
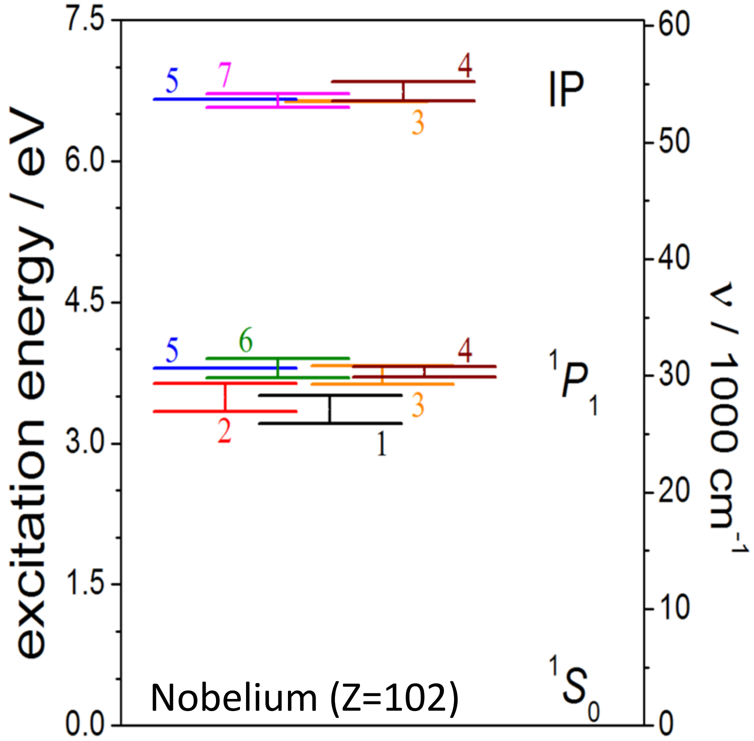
# Multi-Monfiguration Dirac Fock atomic physics calculations: $^{227}$Ac




| Fred,- Phys. Rev. 98 (1955) | MCDF calculations + experimental data on $^{227}$Ac |
|---|---|
| $\mu_{lit.} = 1.1(1)\ \mu_N$ | $\mu_{calc.} = 1.07(18)\ \mu_N$ |
| $Q_{lit.} = 1.7(2)$ eb | $Q_{calc.} = 1.74(10)$ eb |

# Atomic levels and Ionization potential: nobelium (Z=102)




Atomic ground state: [Rn]$5f^{14}7s^2$ $^1S_0$

- $Z\alpha \rightarrow 1$: large QED contribution
  & relativistic effects
  in the electronic structure
- Strong electron correlations
- Benchmark predictive power of atomic theory
- Ionization potential (IP) → chemical properties
- Determination of nuclear properties

**Model calculations**

**1, 2 (MCDF)**: S.Fritzsche, Eur. Phys. J. D 33 (2005) 15

**3 (IHFSCC)**: A.Borschevsky et al., Phys. Rev. A 75 (2007) 042514

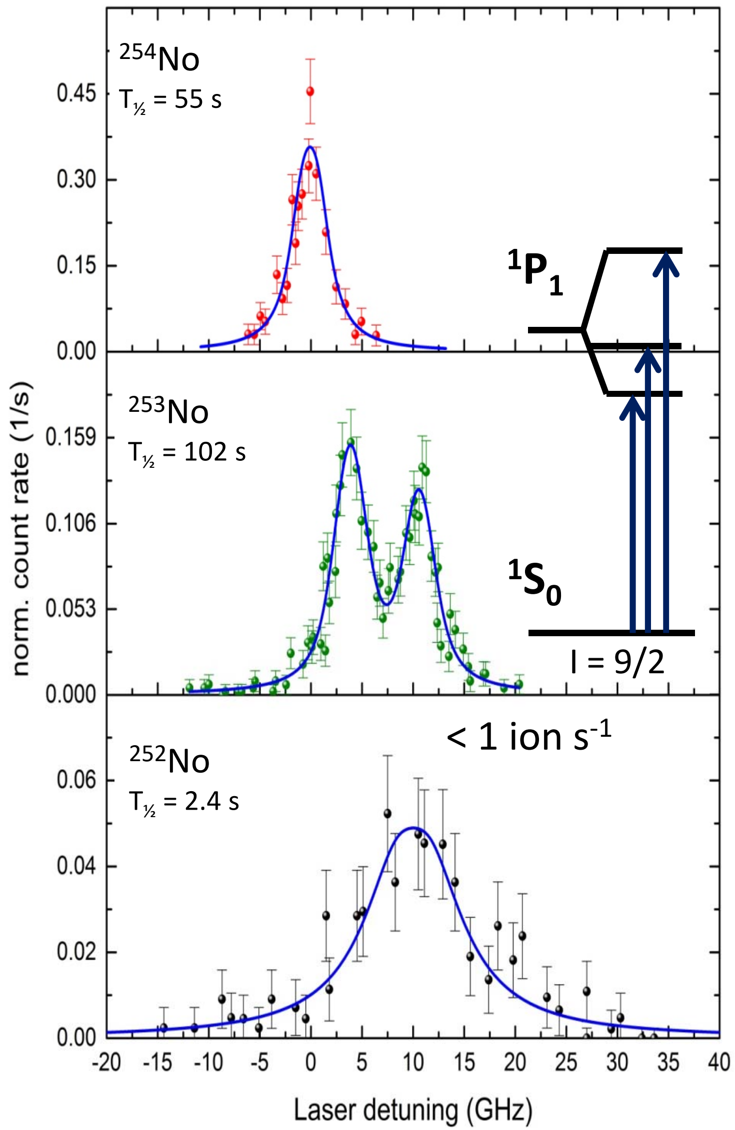
**4 (RCC)**: V.A.Dzuba et al., Phys. Rev. A 90 (2014) 012504

**5 (MCDF)**: Y.Liu et al., Phys. Rev. A 76 (2007) 062503

**6 (MCDF)**: P.Indelicato et al., Eur. Phys. J. D 45 (2007) 155

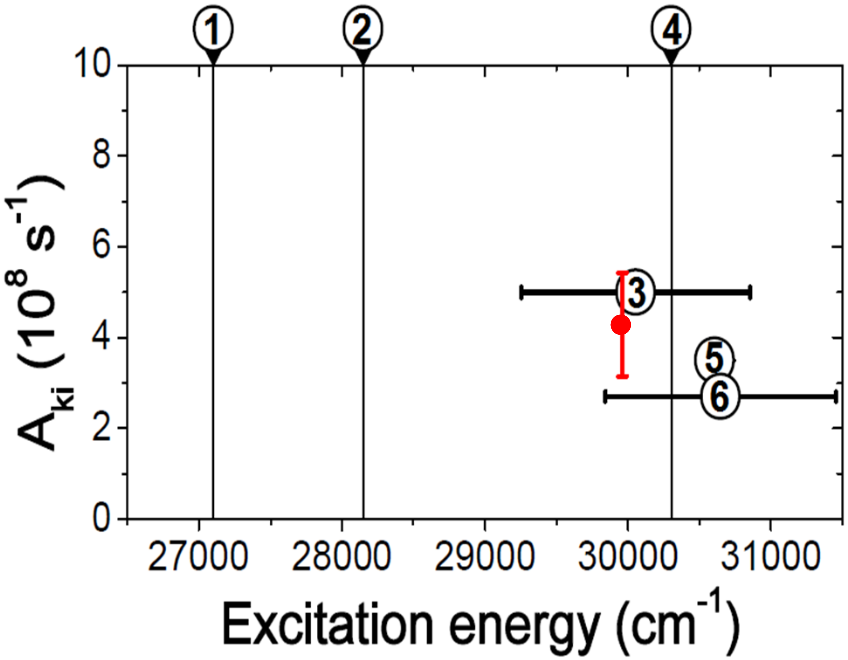
**7 (extrapolation)**: J.Sugar, J. Chem. Phys. 60 (1974) 4103

# Laser ionization spectroscopy of $^{252,253,254}$No




- Theory-guided search for the atomic transition

| | $\nu_1$ (cm$^{-1}$) | $A_{ki}$ (s$^{-1}$) x 10$^{8}$ |
|---|---|---|
| Experiment | 29,961.457(7)$_{stat}$ | 4.2 (2.6)$_{stat}$ |
| IHFSCC [3] | 30,100(800) | 5.0 |
| MCDF [6] | 30,650(800) | 2.7 |

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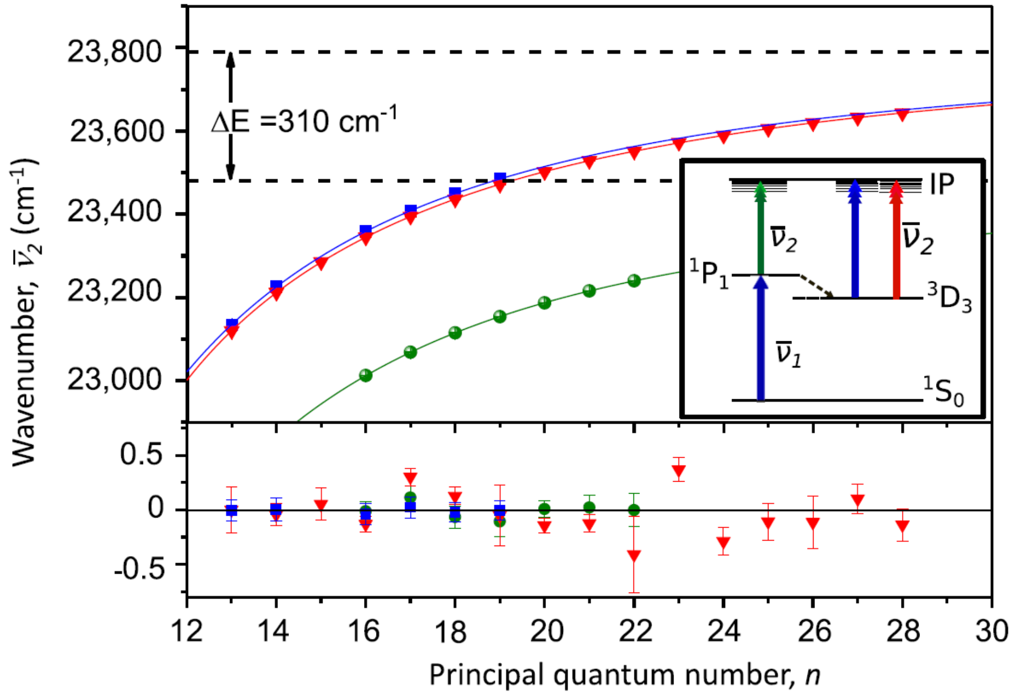
M. Laatiaoui et al., Nature 538 (2016), S. Raeder et al., PRL120 (2018), P. Chhetri et al., PRL120 (2018)

# Ionization Limits & Ionization Potential




- Series fitted with Rydberg-Ritz formula:

$$E_n = E_{\mathrm{IP}} - \frac{R_\mu}{[n - \delta(n)]^2}.$$

| | IP ($cm^{-1}$) | $^3D_3$ |
|---|---|---|
| Experiment | 53444,0 (4) | 29652 (+8/-1) |
| IHFSCC [1] | 53489 (800) | 29897 (800) |

*[1] A. Borschevsky et al., Phys. Rev. A **75** (2007) 042514*



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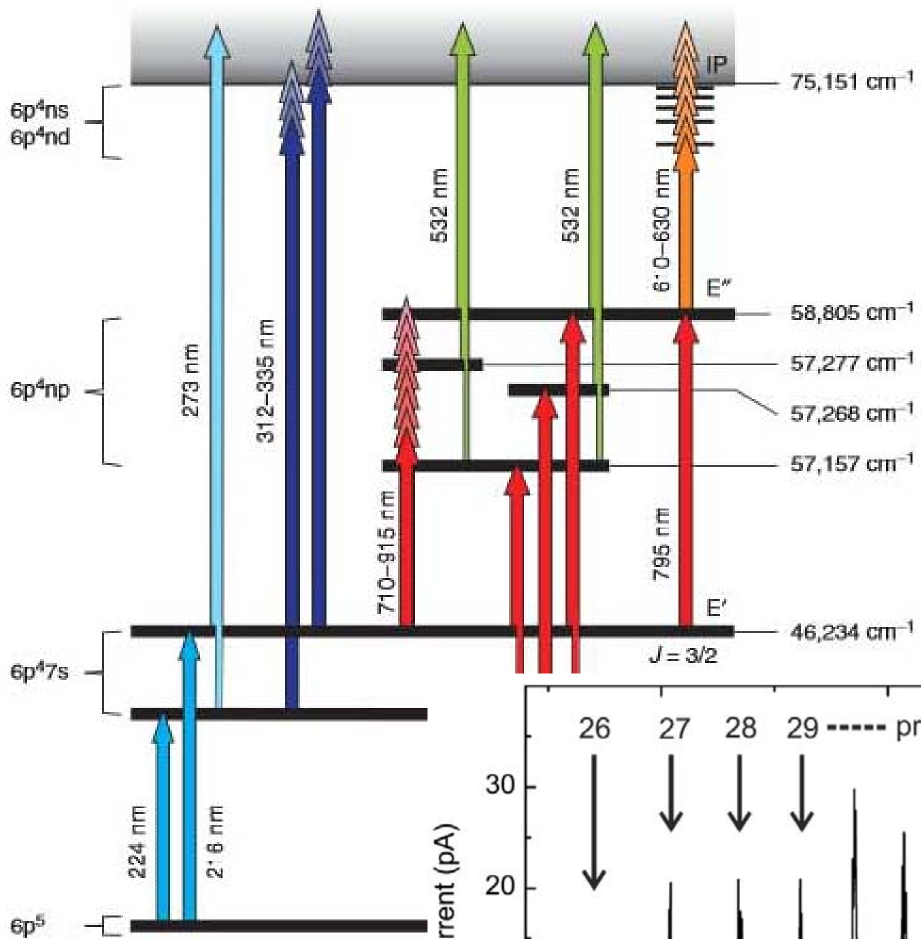
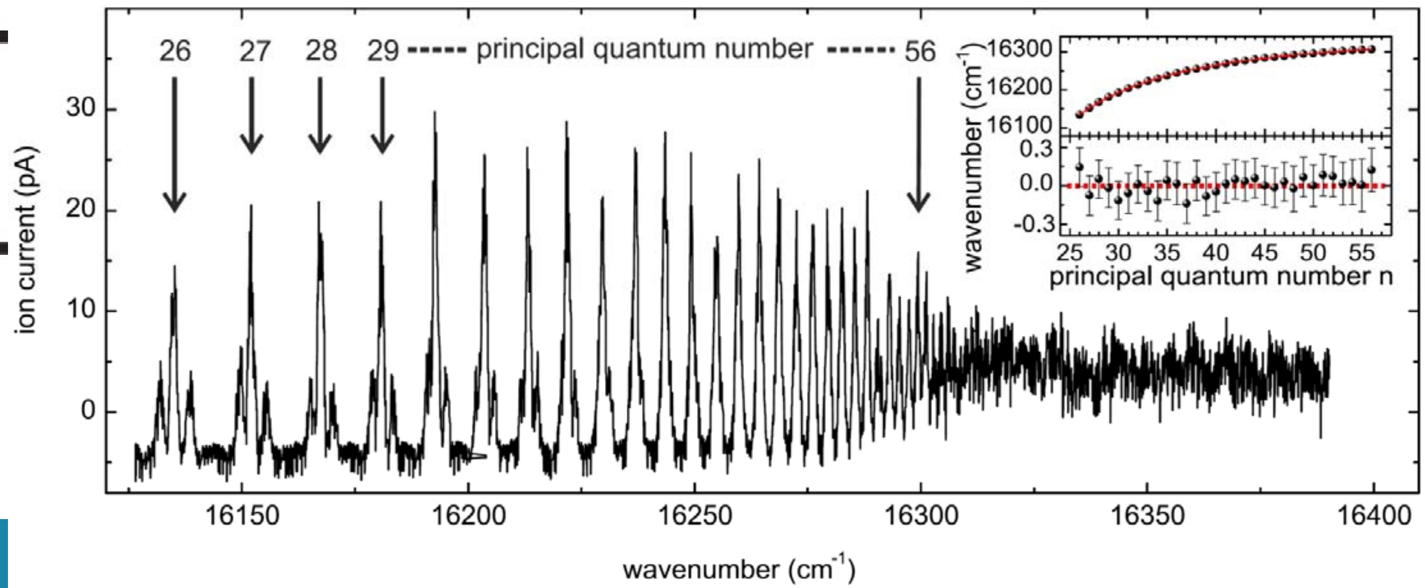
# The Ionization Scheme of Astatine




- ISOLDE (CERN) and TRIUMF (Canada)
- IP(At) = 9.31751(8) eV
- Atomic theory
  - MCDF: 9.24 (15) eV (Fritzsche)
  - CCSD: 9.307 (25) eV (Pershina)

Rothe,- Nature Comm. (2013)



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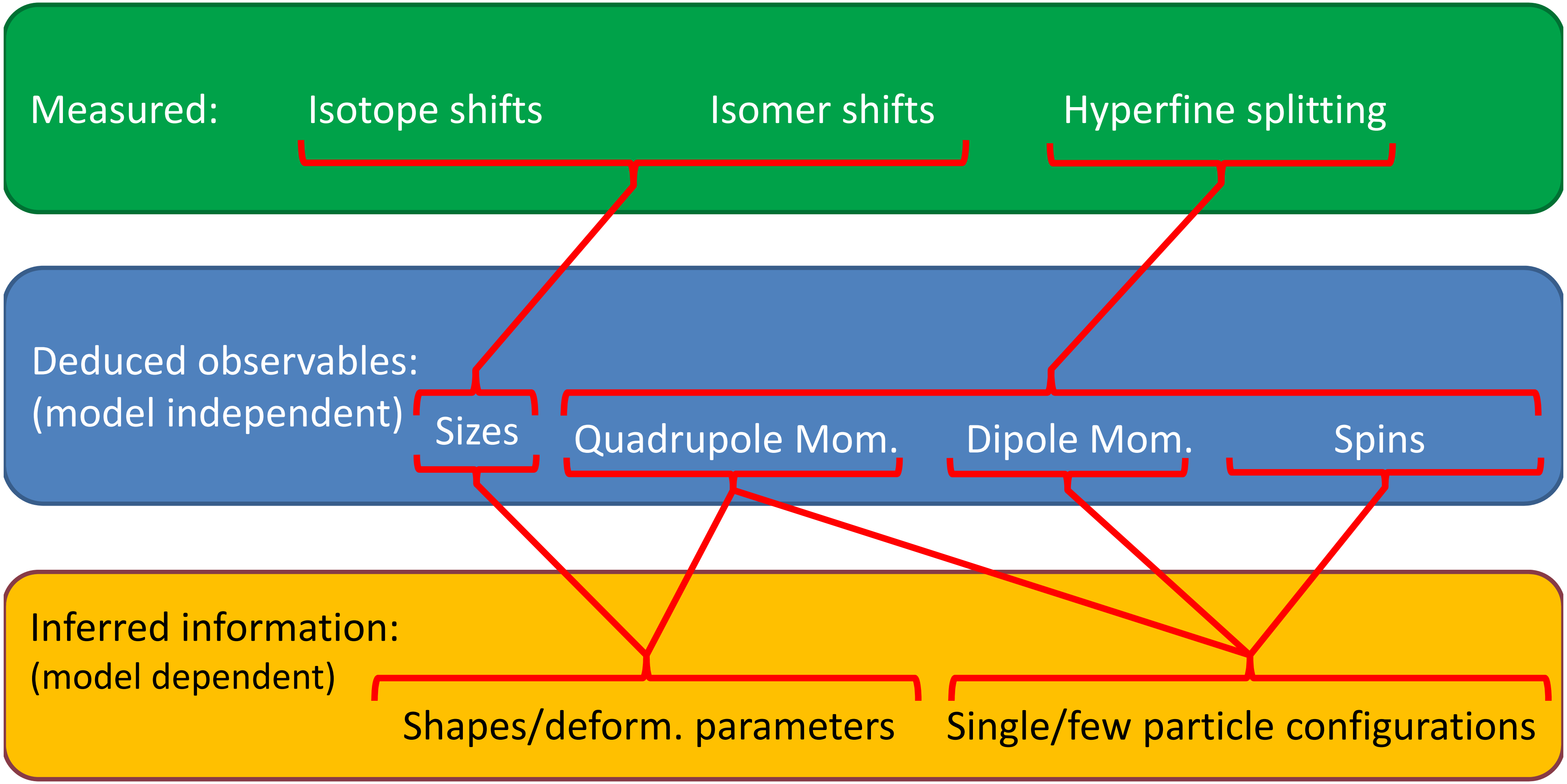
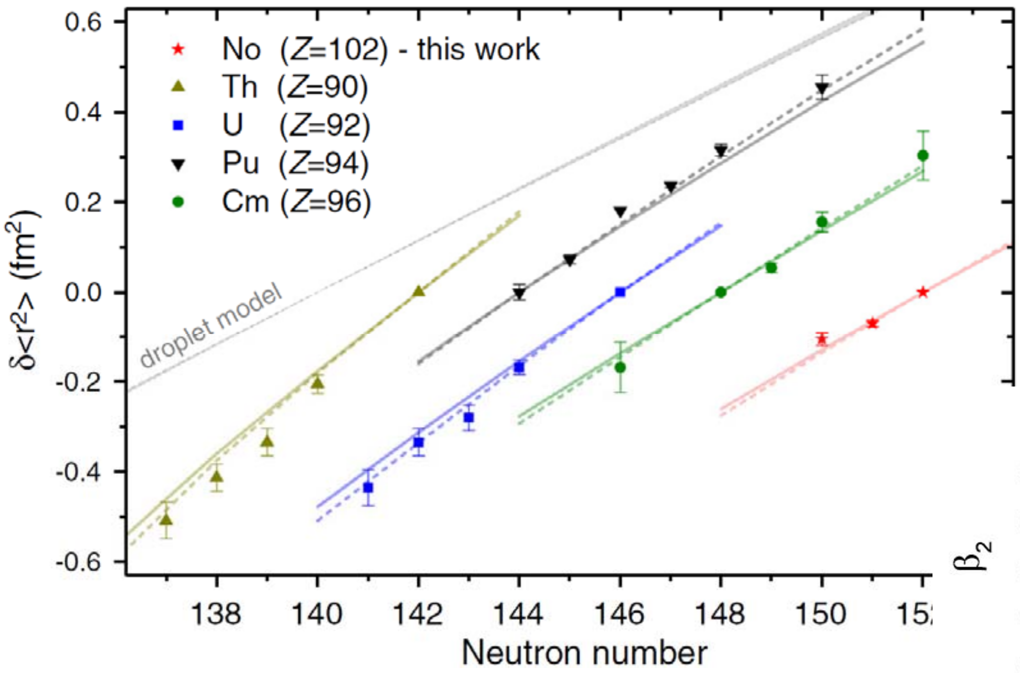
# Laser Spectroscopy Observables - impact

- Nuclear – structure physics
  - independent of nuclear models provided the atomic physics is known

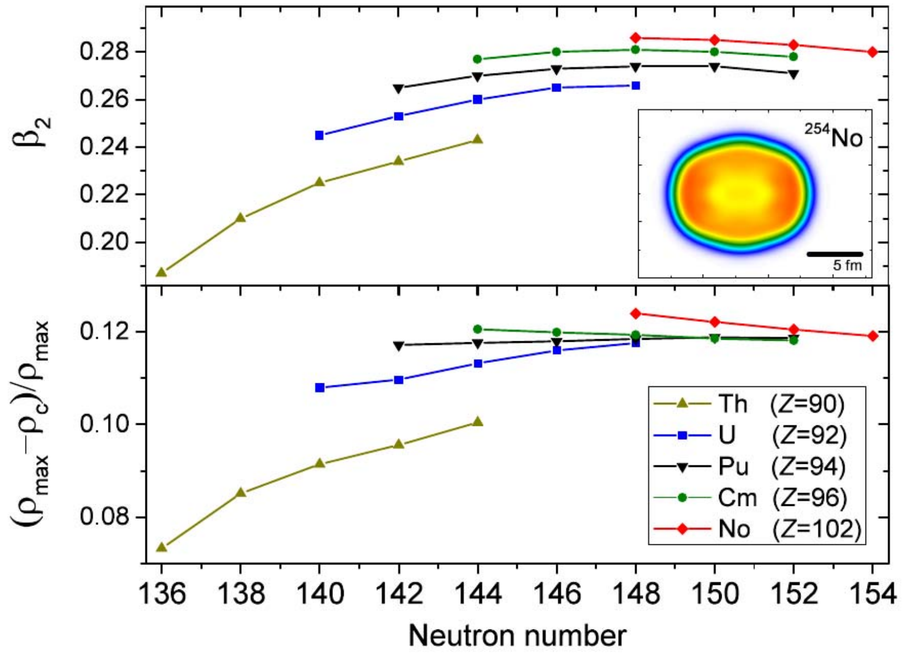


# Laser ionization spectroscopy of $^{252,253,254}No$

- Isotope shift for $^{252\text{-}254}No$ measured
- Change in charge radii: Input from atomic theory




Deformation from DFT calculations



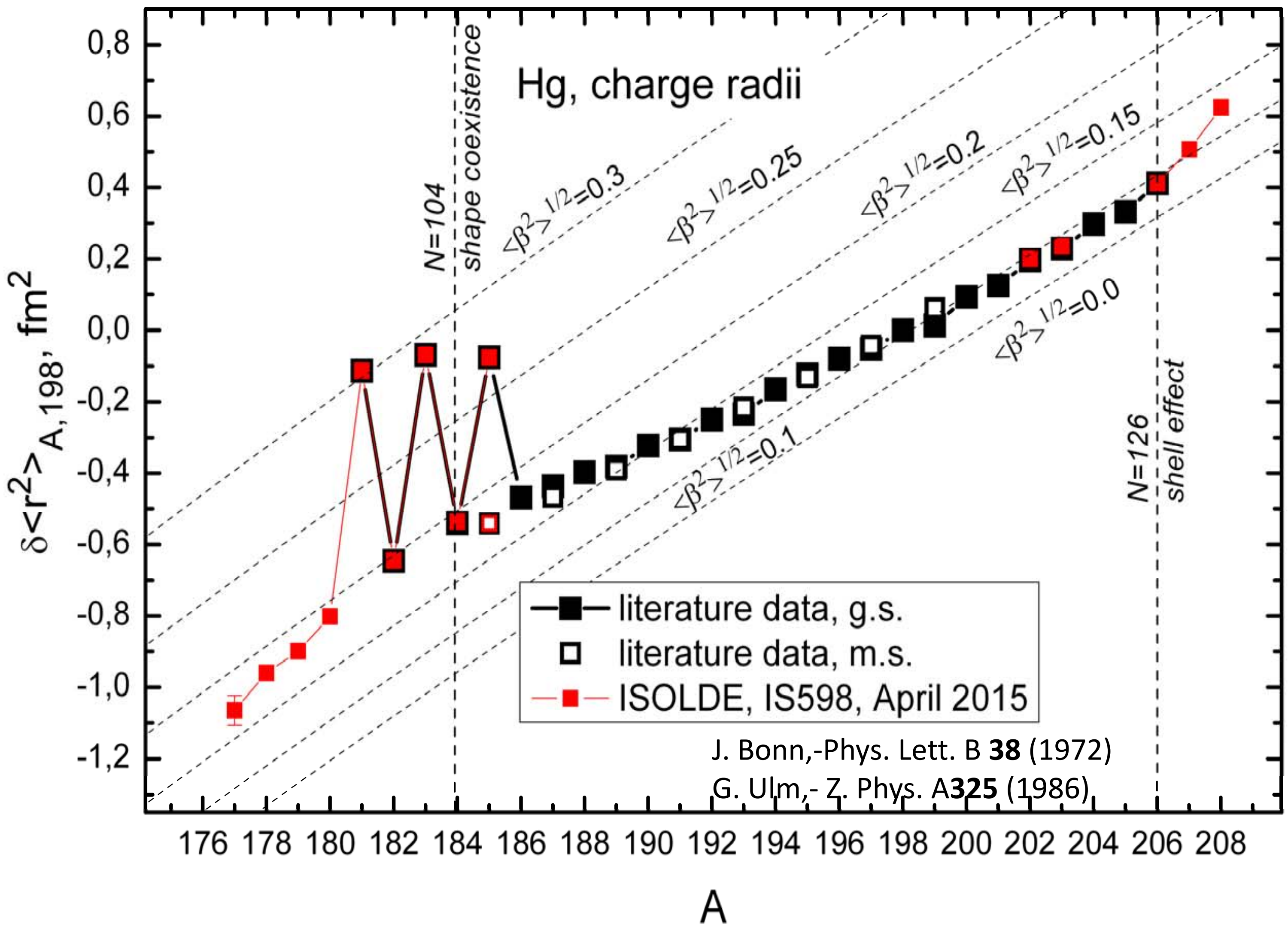

- Nuclear moments of $^{253}No$

| | $\mu$ ($\mu_N$) | $Q_s$ (eb) |
|---|---|---|
| Laser spec. (this work) | -0.527(33)[75] | 5.8(14)[8] |

statistics atomic theory

M. Laatiaoui et al., Nature 538 (2016), S. Raeder et al., PRL120 (2018), P. Chhetri et al., PRL120 (2018)

- Charge radii of mercury isotopes



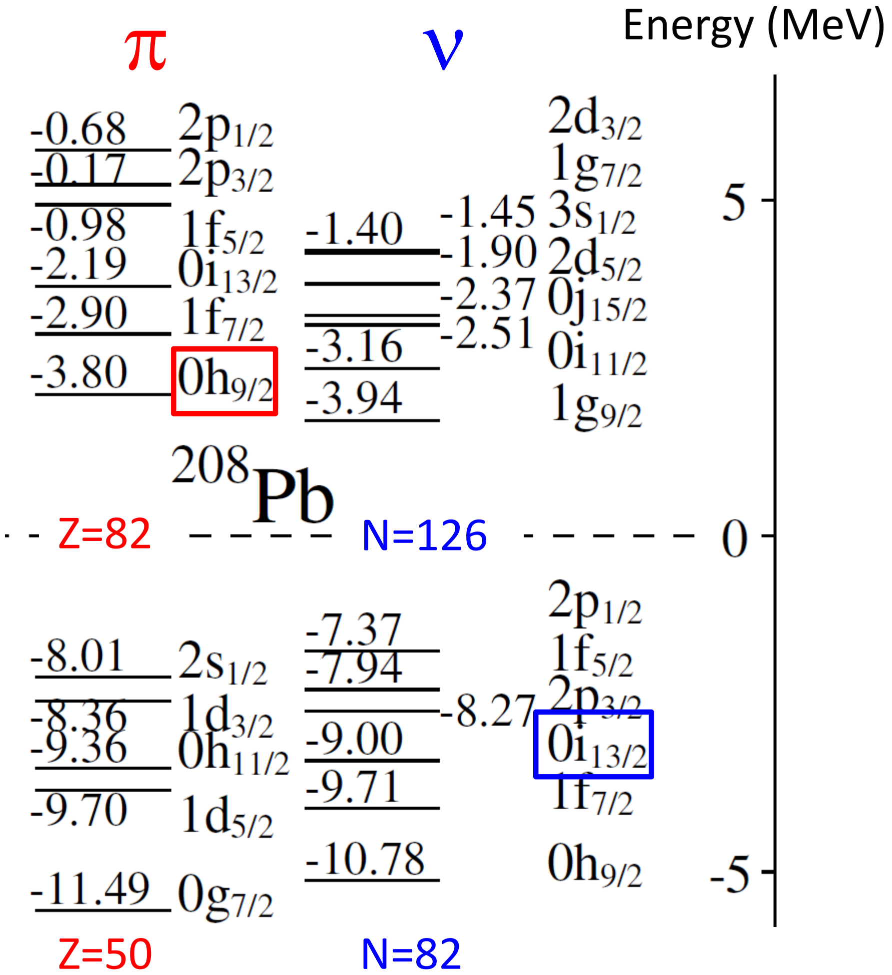

J. Bonn,-Phys. Lett. B **38** (1972)

G. Ulm,- Z. Phys. A**325** (1986)



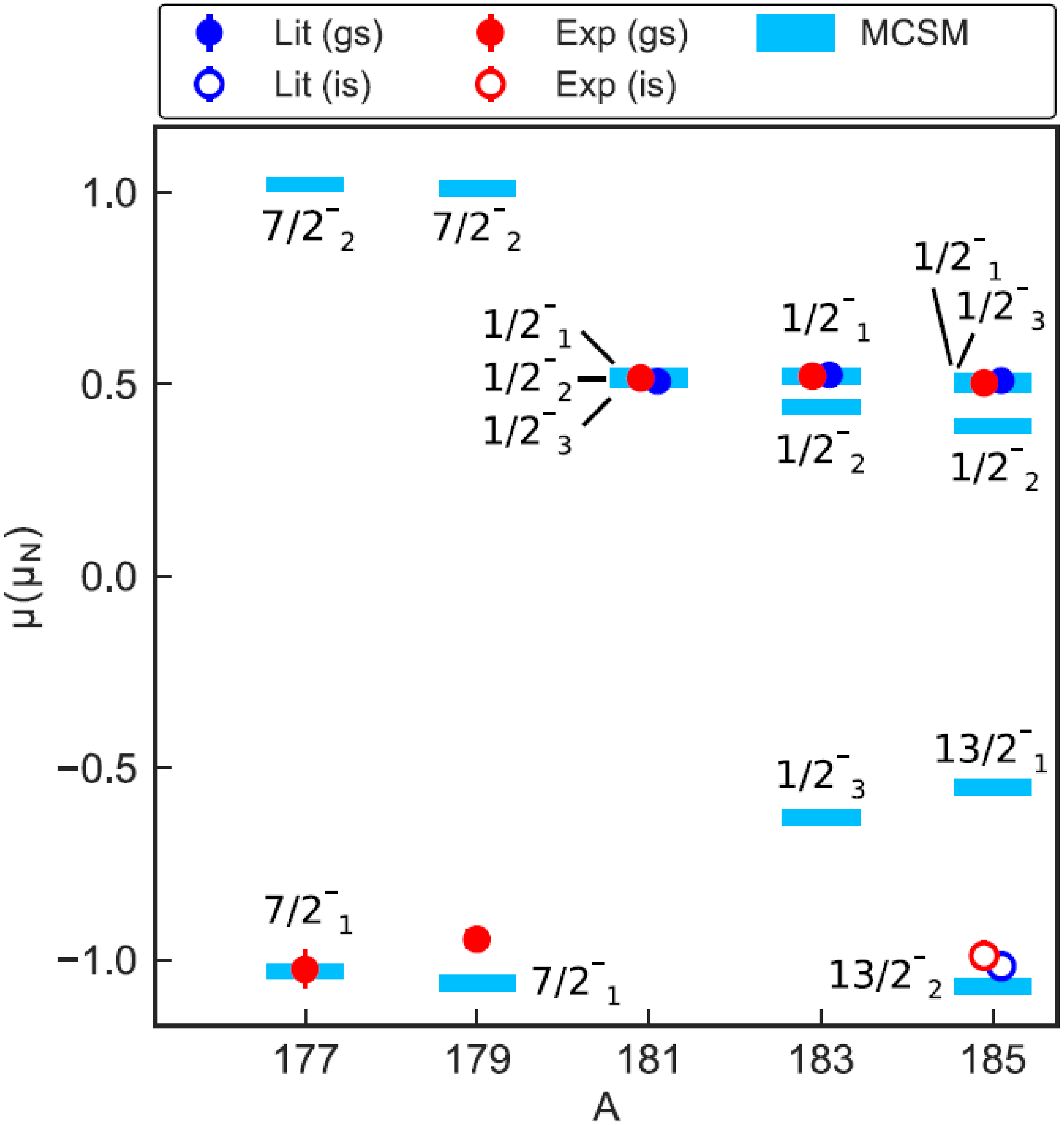
B. Marsh,- Nat. Physics (2018), S. Sels,- PRC 99 (2019)

- Monte-Carlo Shell Model calculation (Y. Tsunoda & T. Otsuka)
  - $^{132}$Sn core – NN, PP[1], PN [2]
  - eff. ch. $\pi$ = 1.6e / eff. ch. $\nu$ = 0.6e / spin quenching = 0.9
  - $\pi$: $1g_{7/2}$ → $1i_{13/2}$ (11 proton orbitals) / $\nu$: $1h_{9/2}$ → $1j_{15/2}$ (13 neutron orbitals)






[1] B.A. Brown, Phys. Rev. Lett. 85, 5300 (2000).
[2] T. Otsuka et al., Phys. Rev. Lett. 104, 012501 (2010).


Lit (gs)
Lit (is)
Exp (gs)
Exp (is)
MCSM
1.0
0.5
0.0
−0.5
−1.0
μ(μN)
177
179
181
183
185
A
7/2⁻2
7/2⁻2
1/2⁻1
1/2⁻2
1/2⁻3
1/2⁻1
1/2⁻2
1/2⁻3
1/2⁻1
1/2⁻3
1/2⁻2
13/2⁻1
7/2⁻1
7/2⁻1
13/2⁻2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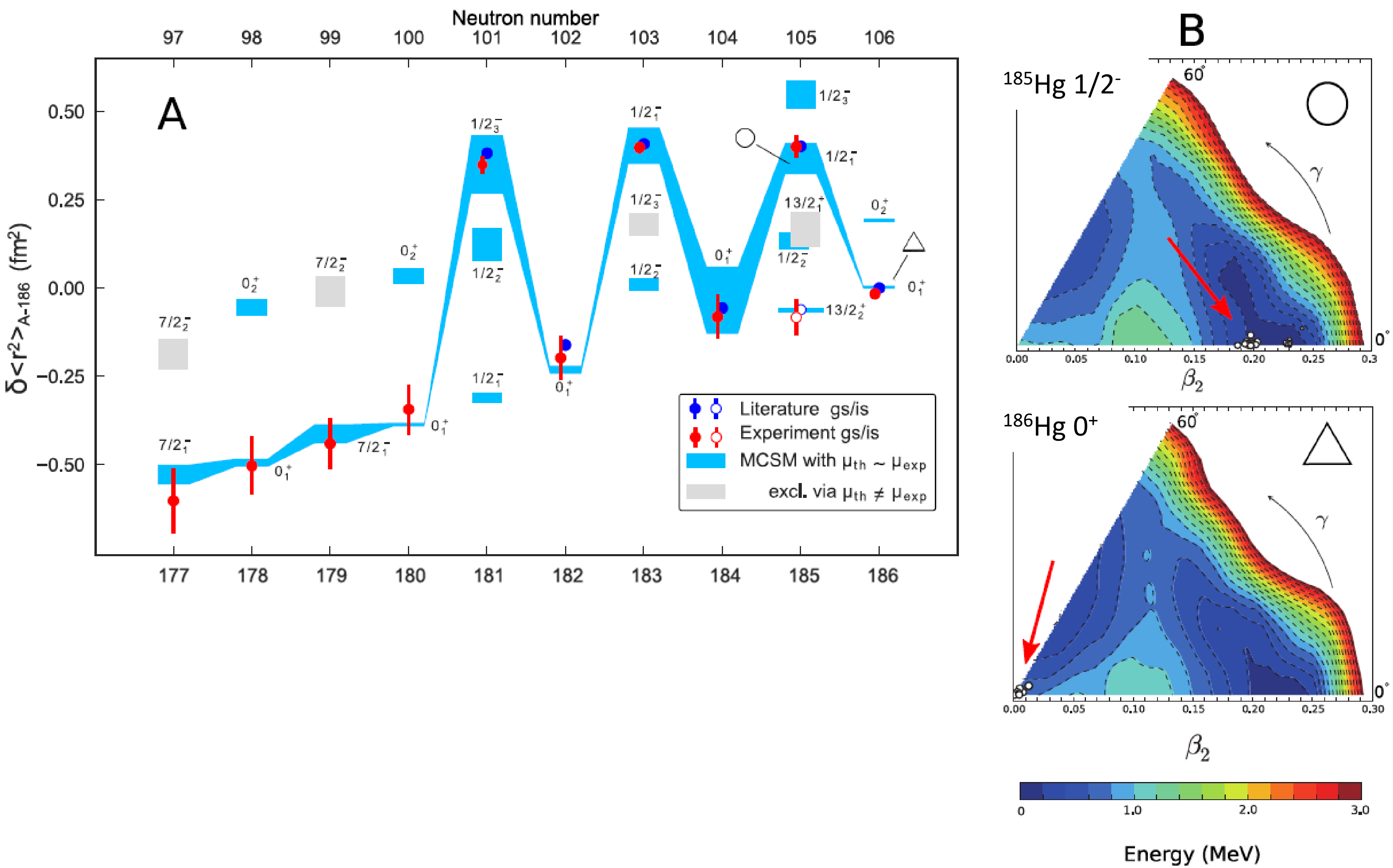



- proton / neutron occupancy!

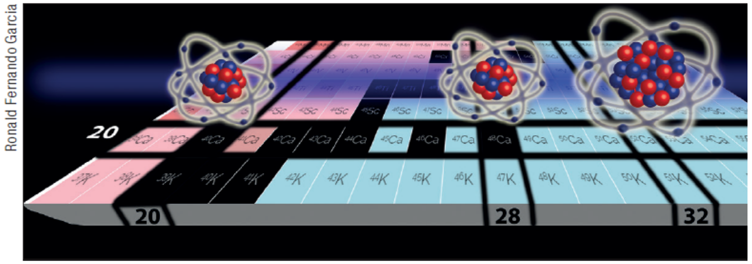
Mass number

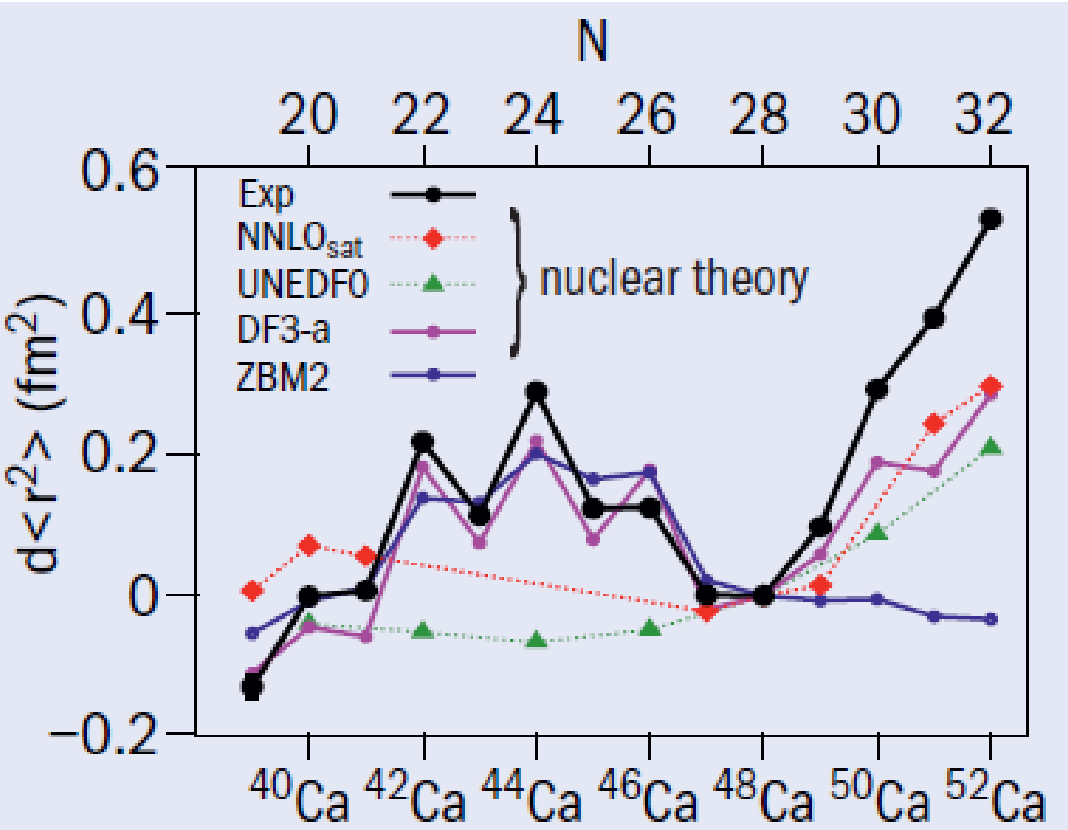
$$Q_0 = \frac{3Z}{\sqrt{5\pi}} < r^2 >< \beta_2 > (1 + 0.36 < \beta_2 >)$$

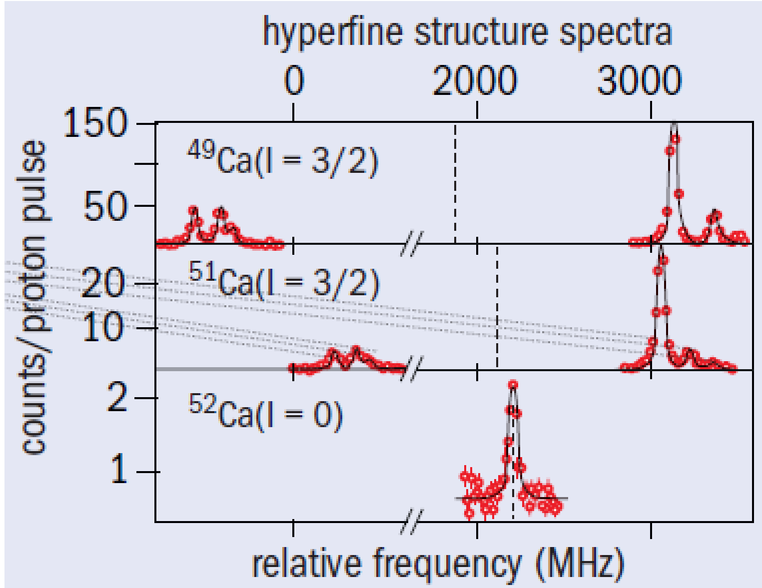
$$< r^2 > = < r^2 >_{DM} \left(1 + \frac{5}{4\pi} < \beta_2 >^2\right)$$

# Charge radii: sensitive to shell gaps and deformation

Ca (Z=20) closed proton shell




Large charge radius of $^{52}Ca$:

- No signature for shell closure at N=32

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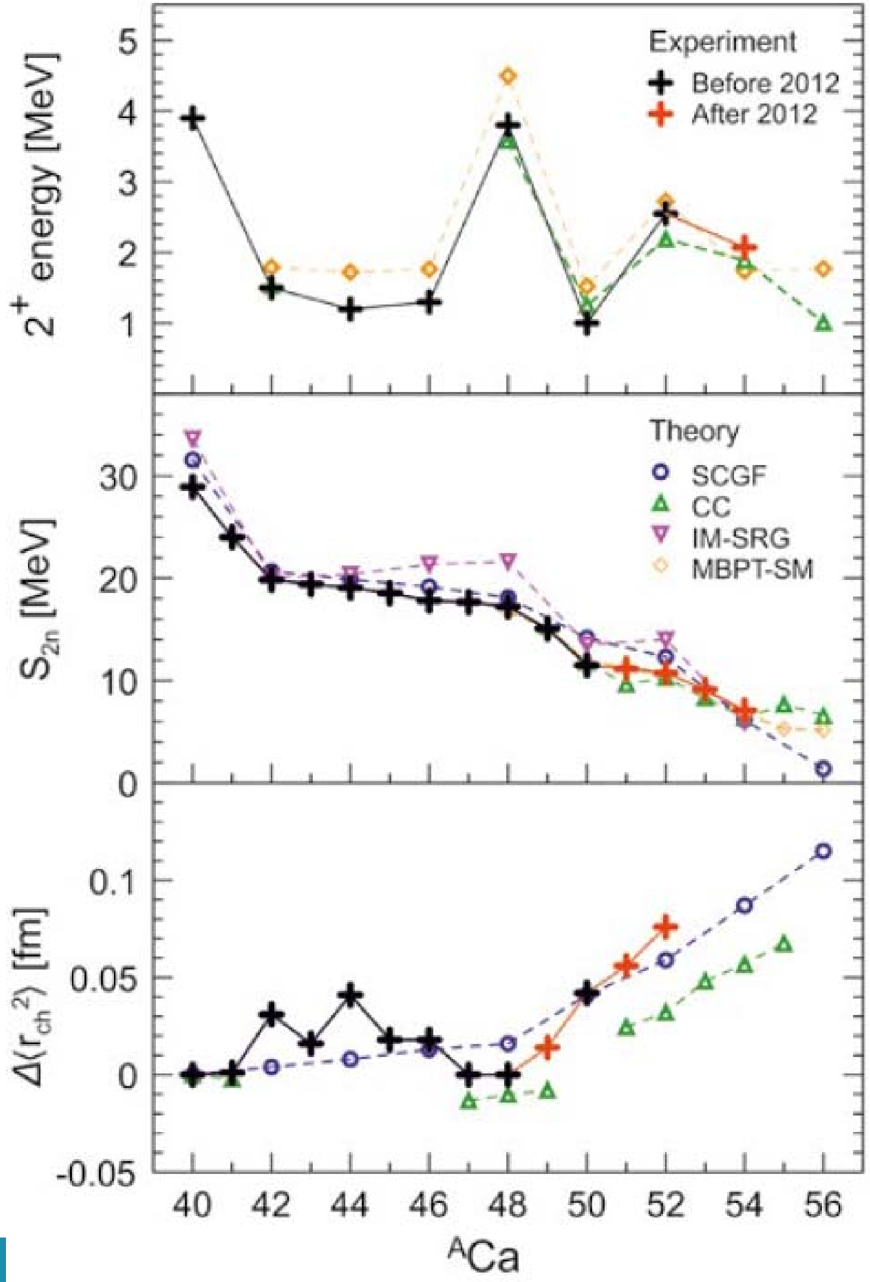
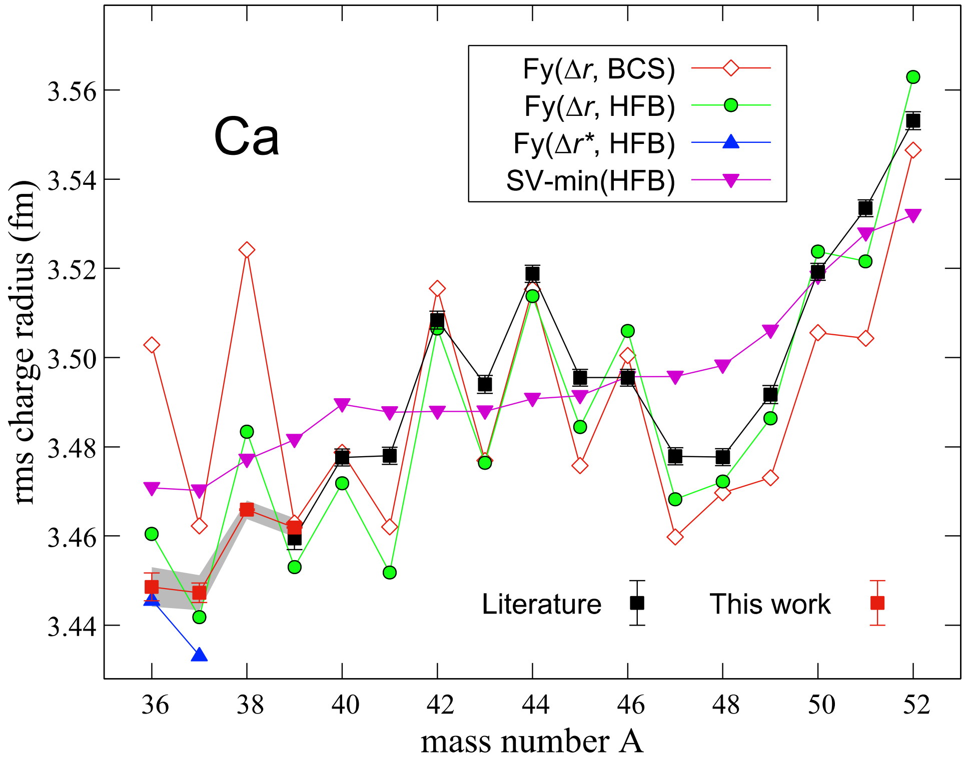
Courtesy: G. Neyens - R.F. Garcia Ruiz et al., Nature Physics 12 (2016) 594

# Charge radii: sensitive to shell gaps and deformation




Fayans Energy Density Functionals
P.G. Reinhard, W. Nazarewicz Phys. Rev. C 95 (2017)

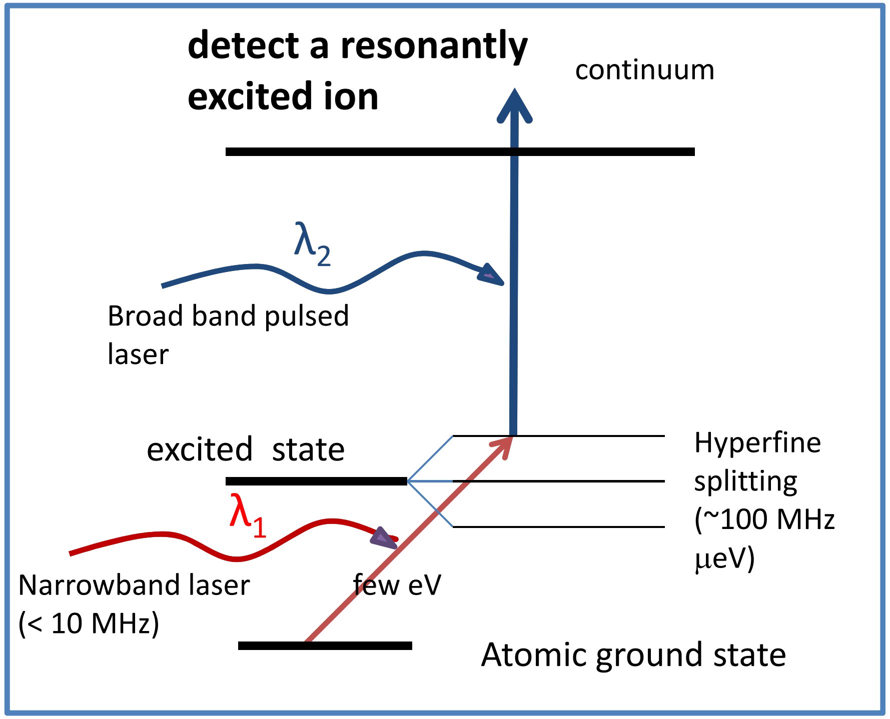



A.J. Miller,- Nature Physics 15 (2019)

# Collinear Laser Spectroscopy

Since 2008 → sensitivity improved by factor 100.000 using CRIS on bunched beams!

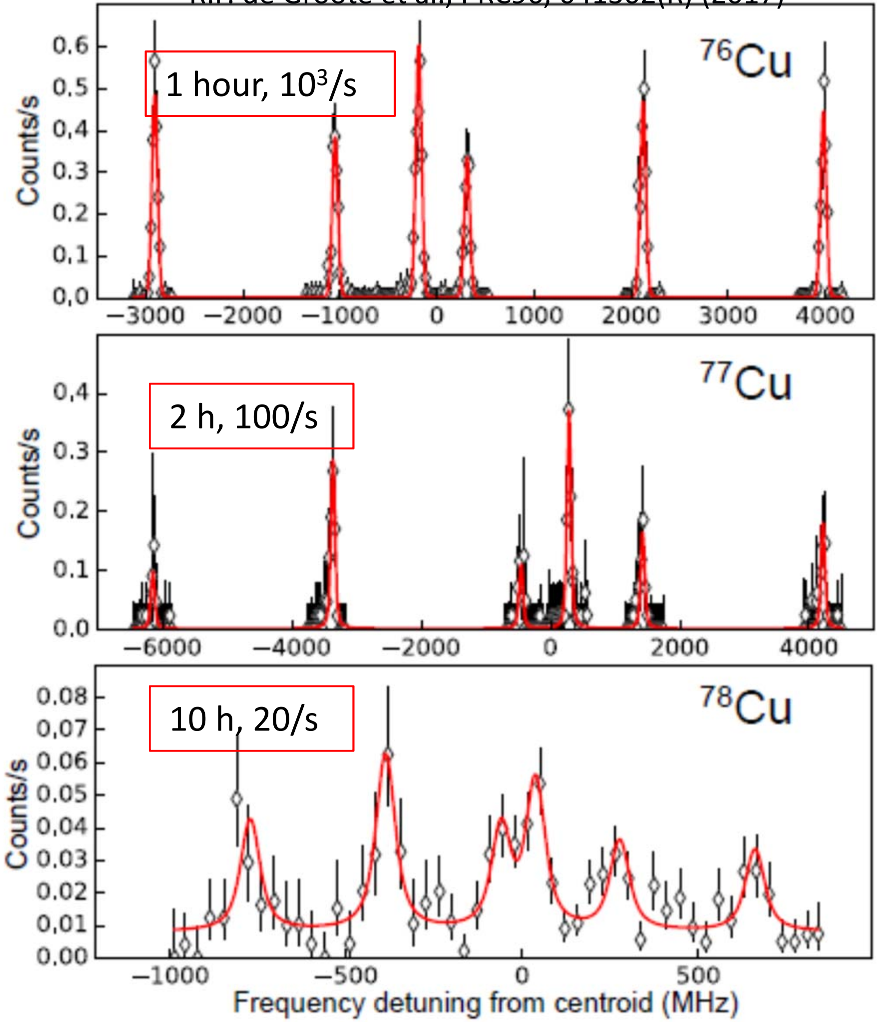
## Collinear **Resonance Ionization** Spectroscopy



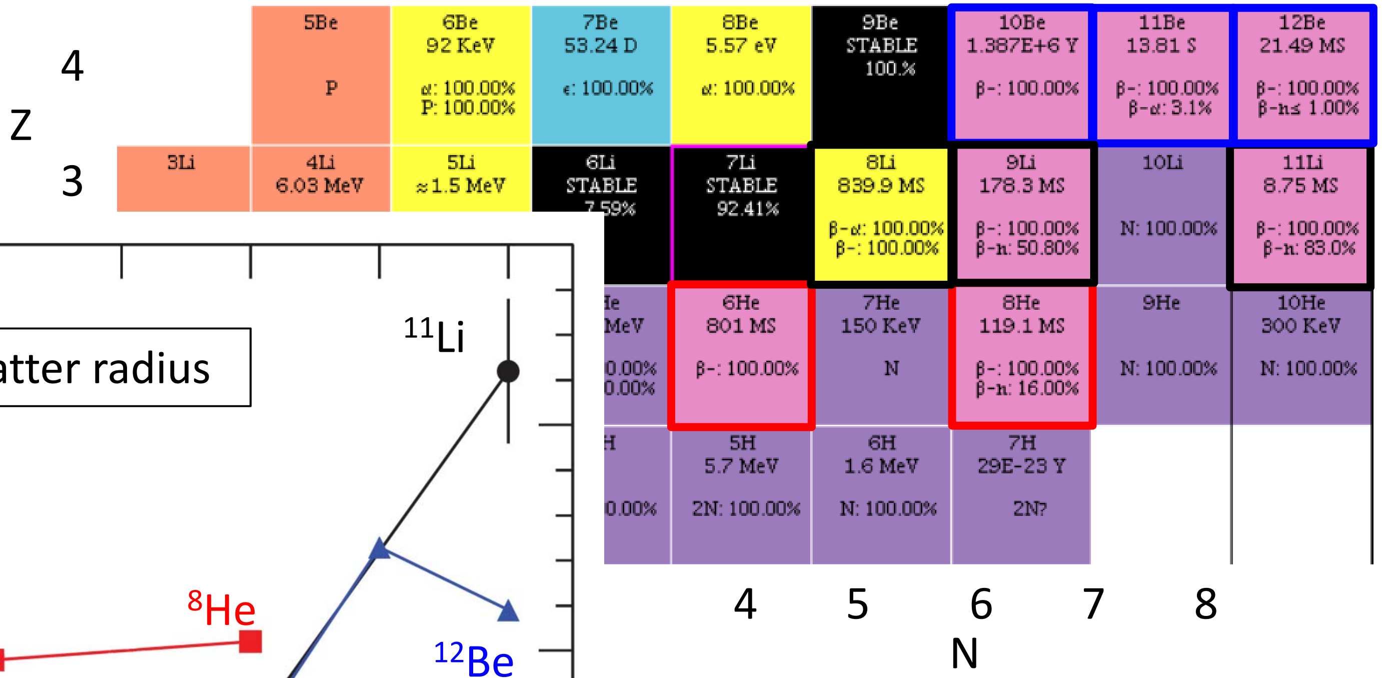

- ultra-low background ( 1 event /10 min)
- high efficiency (~1-5 %)
- high resolution (~ 20-80 MHz)
- current sensitivity 20 ions/s

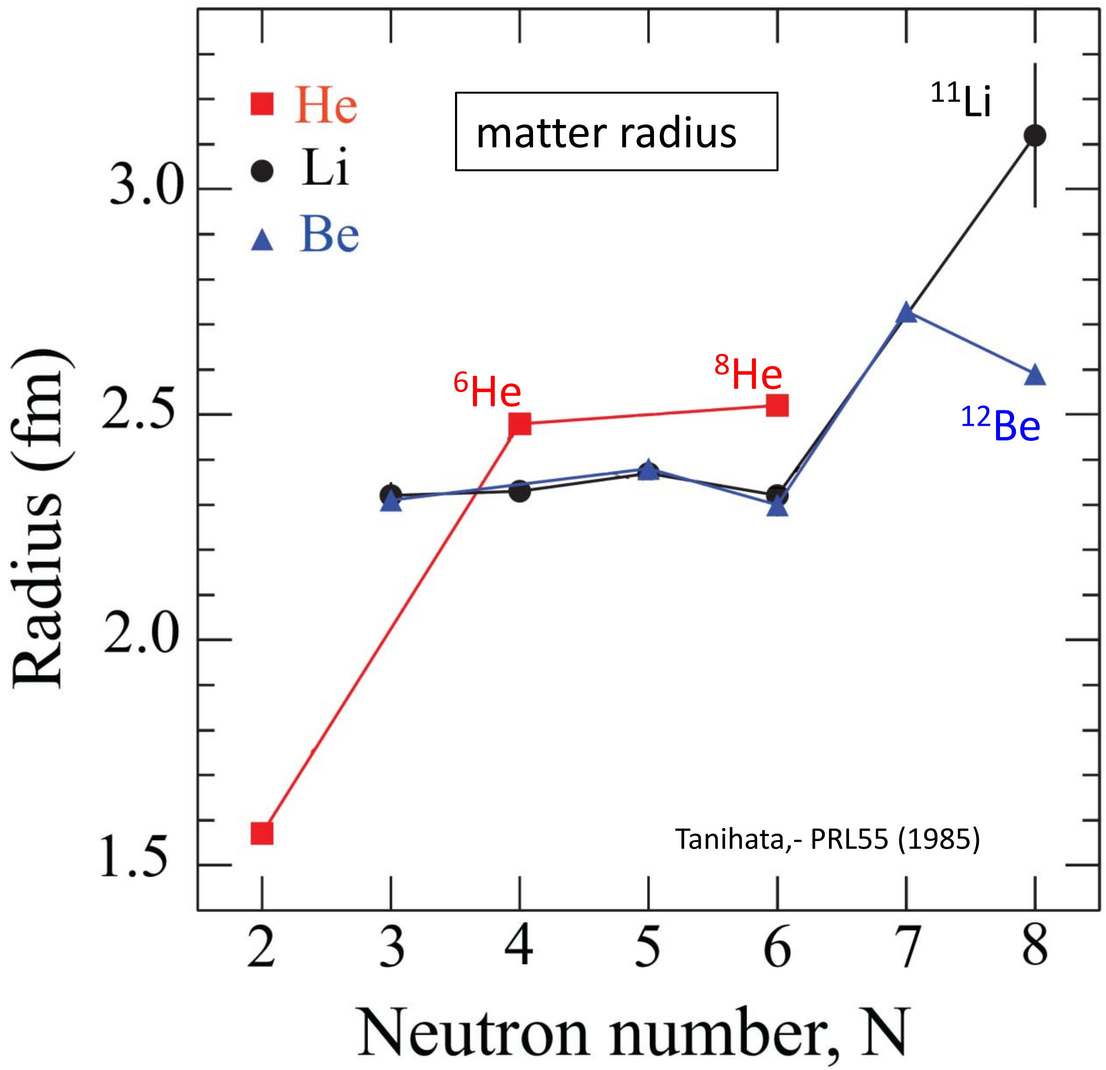
HFS of 63-78Cu

R.P. de Groote et al., PRC96, 041302(R) (2017)



- Halo Nuclei




Jonson and Riisager, Nuclear Physics News 24 (2014) 18

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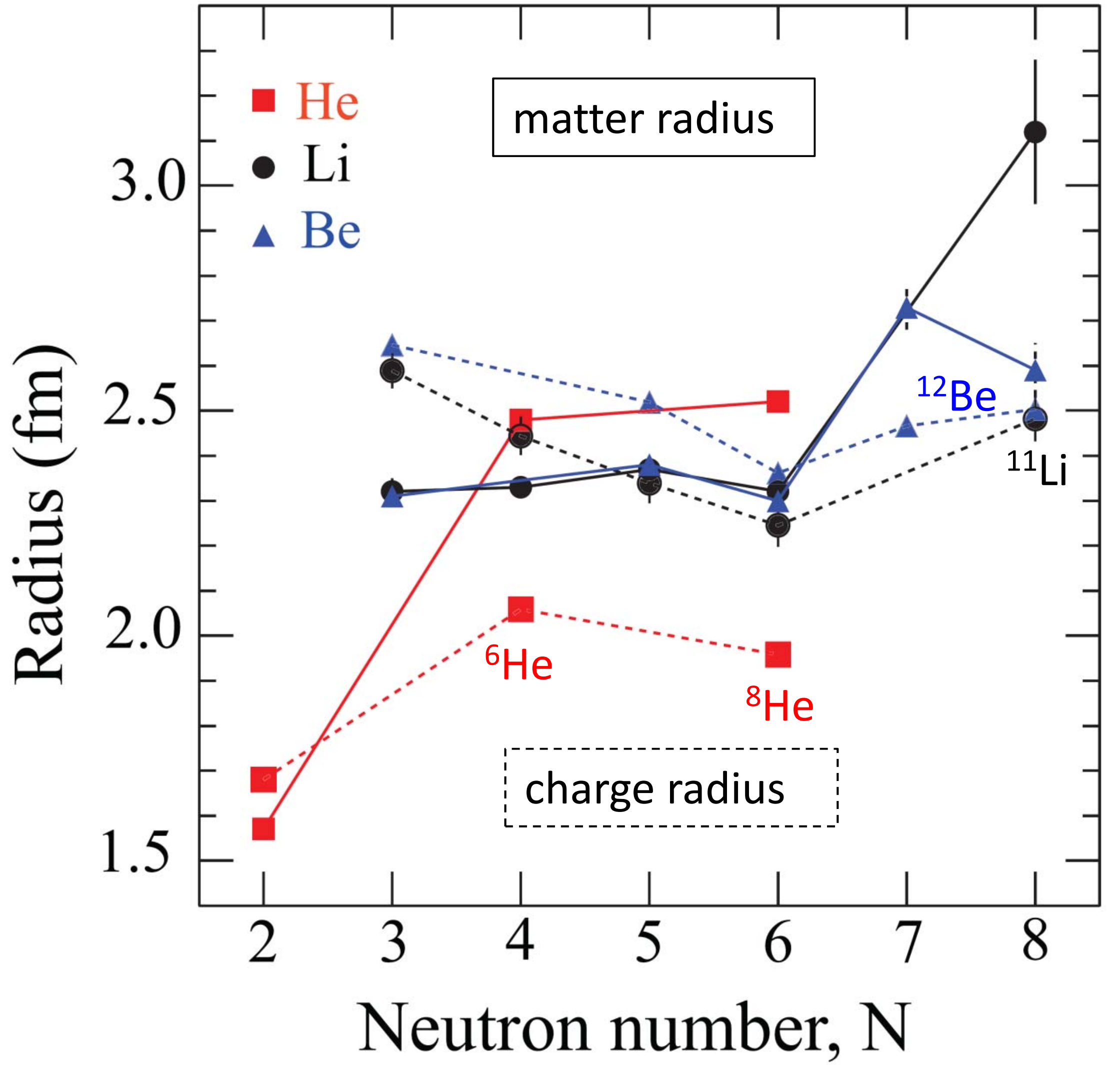
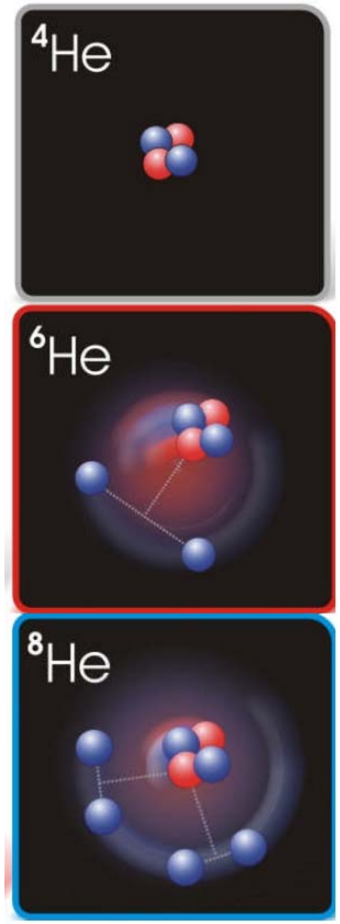
- Halo Nuclei



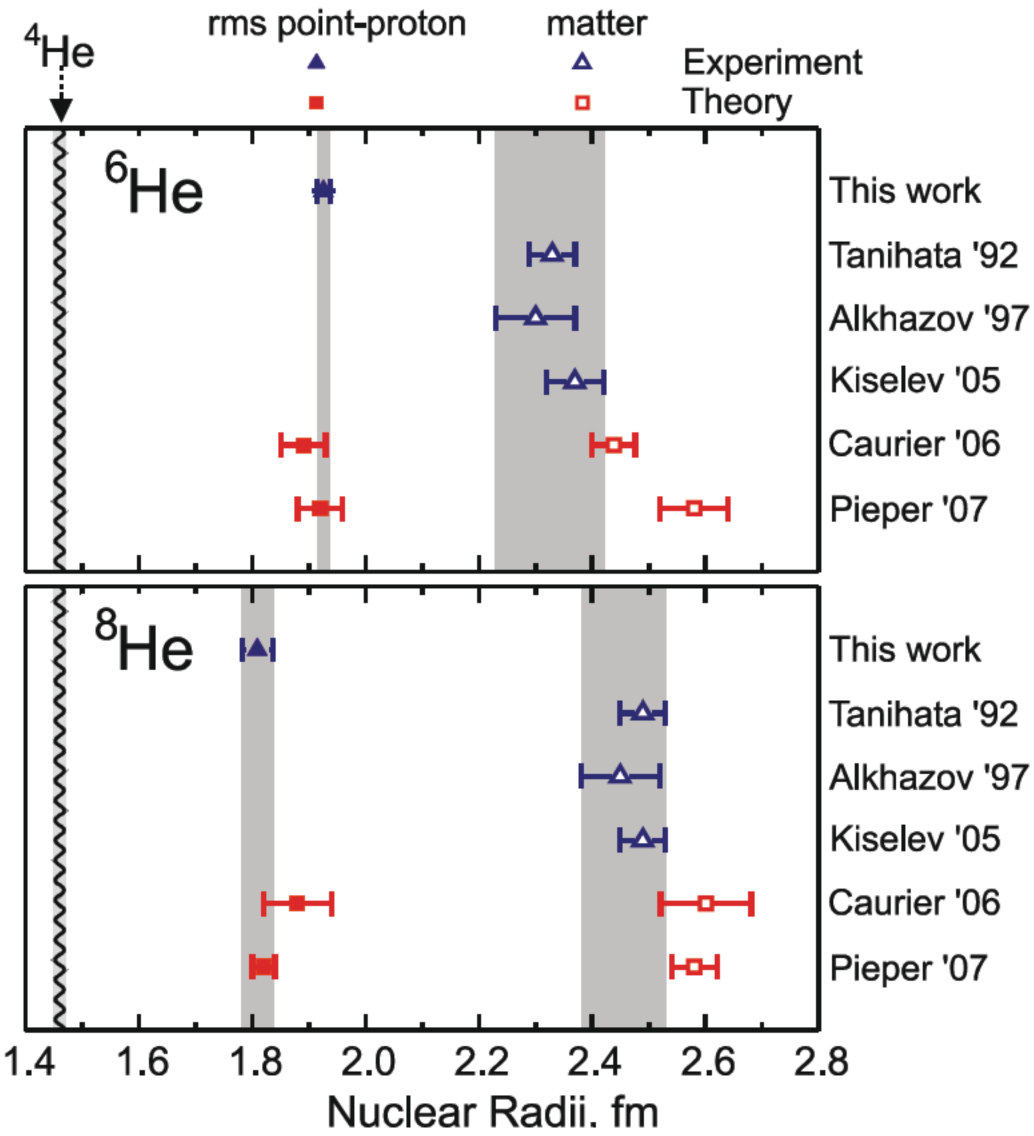



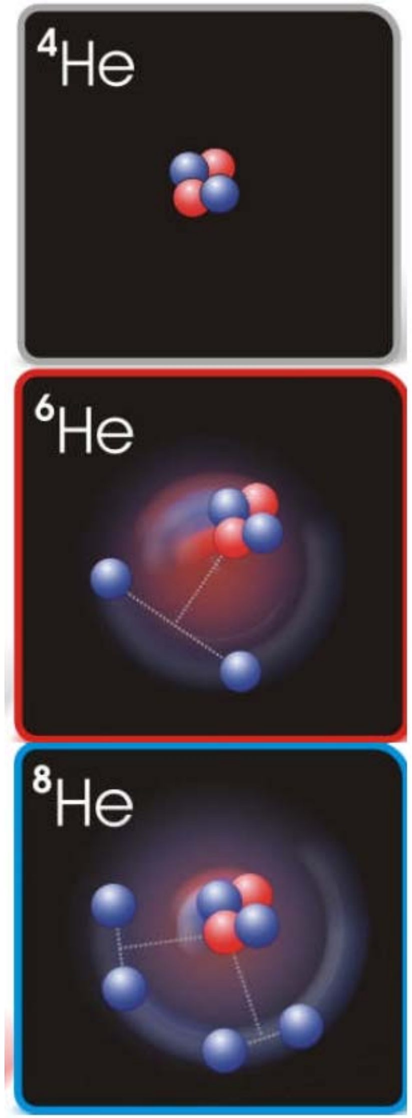

Argonne National Laboratory - GANIL

Atom trap

Wang,- PRL 93 (2004) – He-6

Mueller,- PRL 99 (2007) – He-8



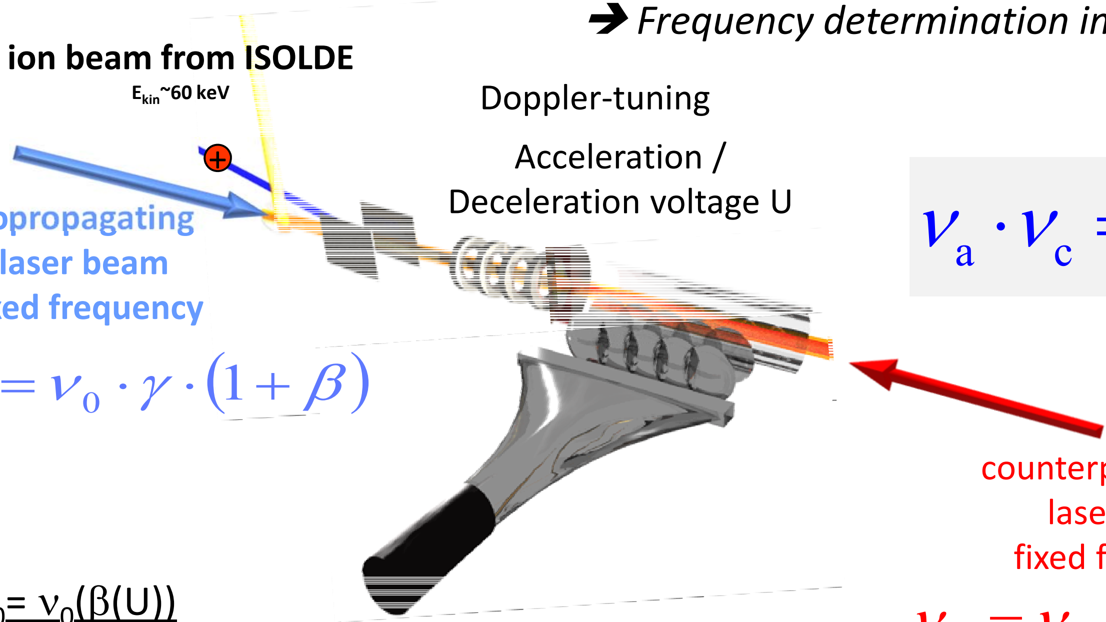




L.B. Wang *et al.*, PRL **93**, 142501 (2004) – He-6

P. Mueller *et al.*, PRL **99**, 252501 (2007) – He-8

- Laser spectroscopy of short-lived Be isotopes:
  Copropagating and Counterpropagating Laser Beams.

➔ *Frequency determination independent of acceleration voltage*




$$\nu_c = \nu_0 \cdot \gamma \cdot (1+\beta)$$

$$\nu_a = \nu_0 \cdot \gamma \cdot (1-\beta)$$

$$\nu_a \cdot \nu_c = \nu_0^2 \cdot \gamma^2 \cdot \left(1-\beta^2\right) = \nu_0^2$$

$\nu_0 = \nu_0(\beta(U))$

- acceleration voltage $\Delta U/U = 10^{-4}$
- $\delta\nu_{IS}(^{10}Be, ^{11}Be) = 18$ MHz

Requirements:

- Measure absolute frequencies: Accuracy: $\Delta\nu/\nu < 10^{-9}$
- Dedicated Laser System for absolute Frequency Measurements: laser frequency Comb
- High sensitivity: 8000 ions per second $^{12}Be(T_{1/2} = 21ms)$

Courtesy: W. Nörtershäuser

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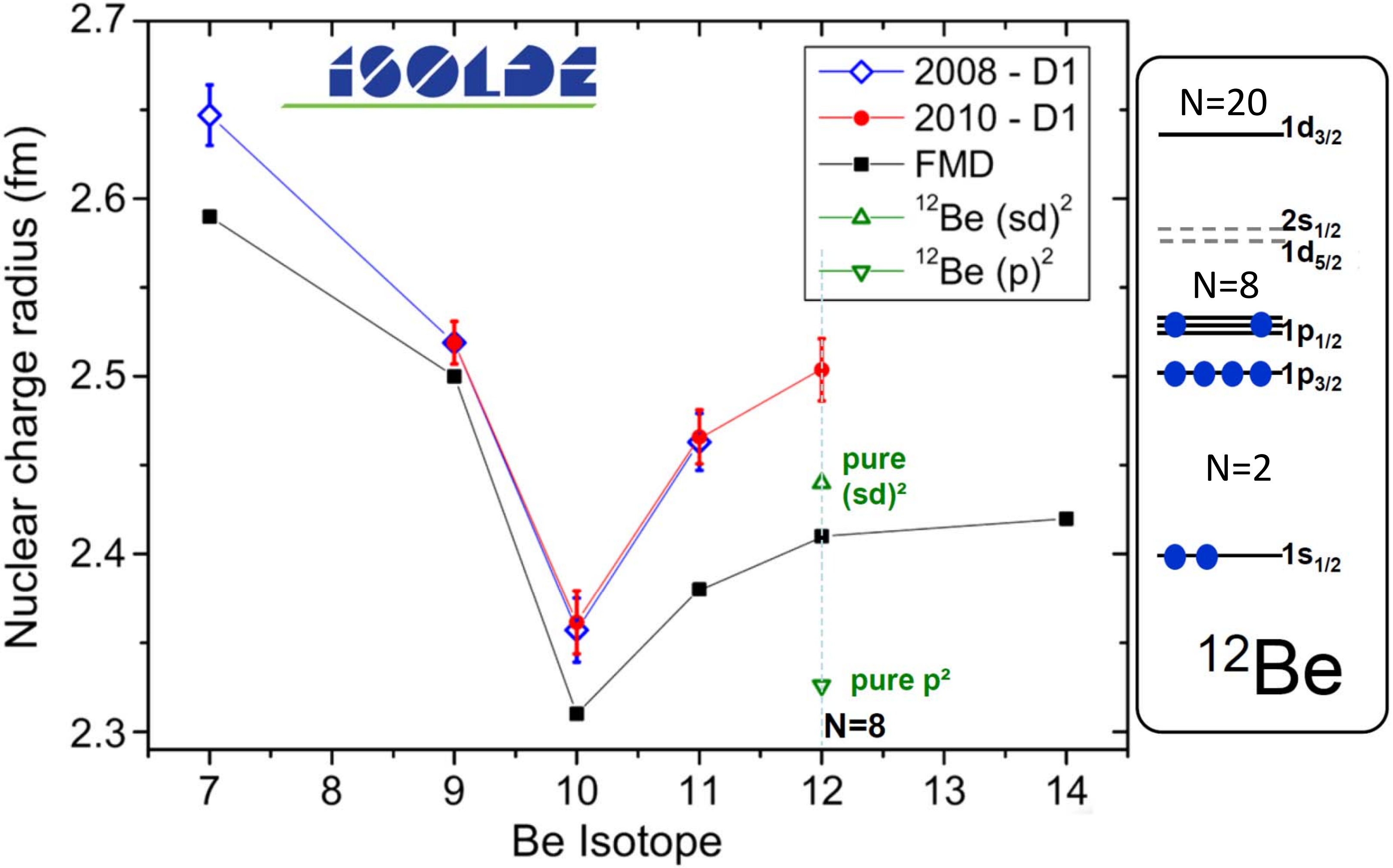



Nörtershäuser,- PRL 102 (2009)
Krieger,- PRL 108 (2012)

The $(sd)^2$ configuration in $^{12}$Be is very different from the picture in $^{11}$Be where a very small neutron separation energy favors the appearance of an $s$-wave halo.
This is also reflected in the matter radius that is significantly smaller in the $^{12}$Be$(sd)^2$ configuration than in $^{11}$Be.

# Laser Spectroscopy Observables - impact

- Atomic physics
  - Especially in the (super) heavy element region – ionization potential, atomic levels, transition strengths
  - Ex.: actinium, nobelium

  (M. Bissel, D. Hanstrop, M. Laatiaoui, D. Yordanov)

- Nuclear physics
  - $\delta<r^2>$, $\mu$, $Q_s$ and $I^\pi$
  - Nuclear-model independent provided atomic physics is known
  - Ex. : density profile in nobelium, shape coexistence in Hg isotopes, Ca charge radii

  (K. Flanagan, K. Minamisono, I. Moore, P. Müller, G. Neyens, W. Nörtershäuser, D. Yordanov, M. Bissell)

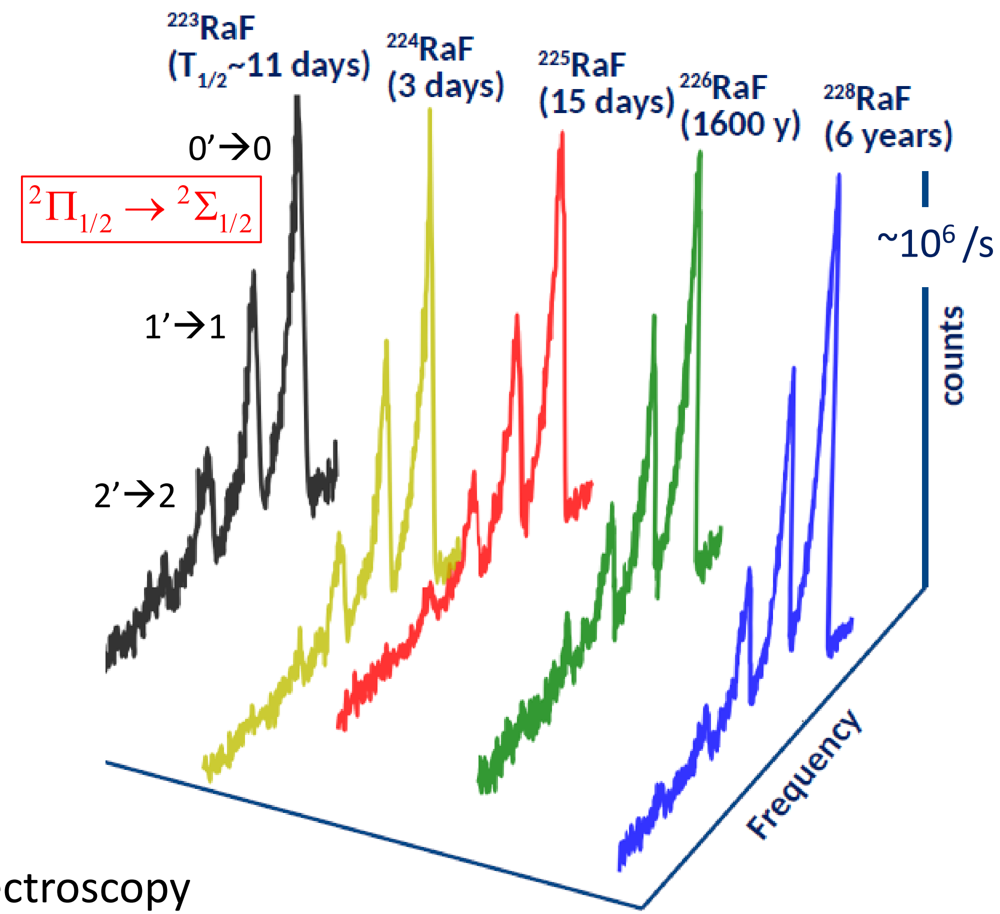
- Fundamental physics
  - Use of the high (ultimate) precision
  - Molecular ions,…

  (R. Garcia Ruiz, I. Moore , P. Müller)

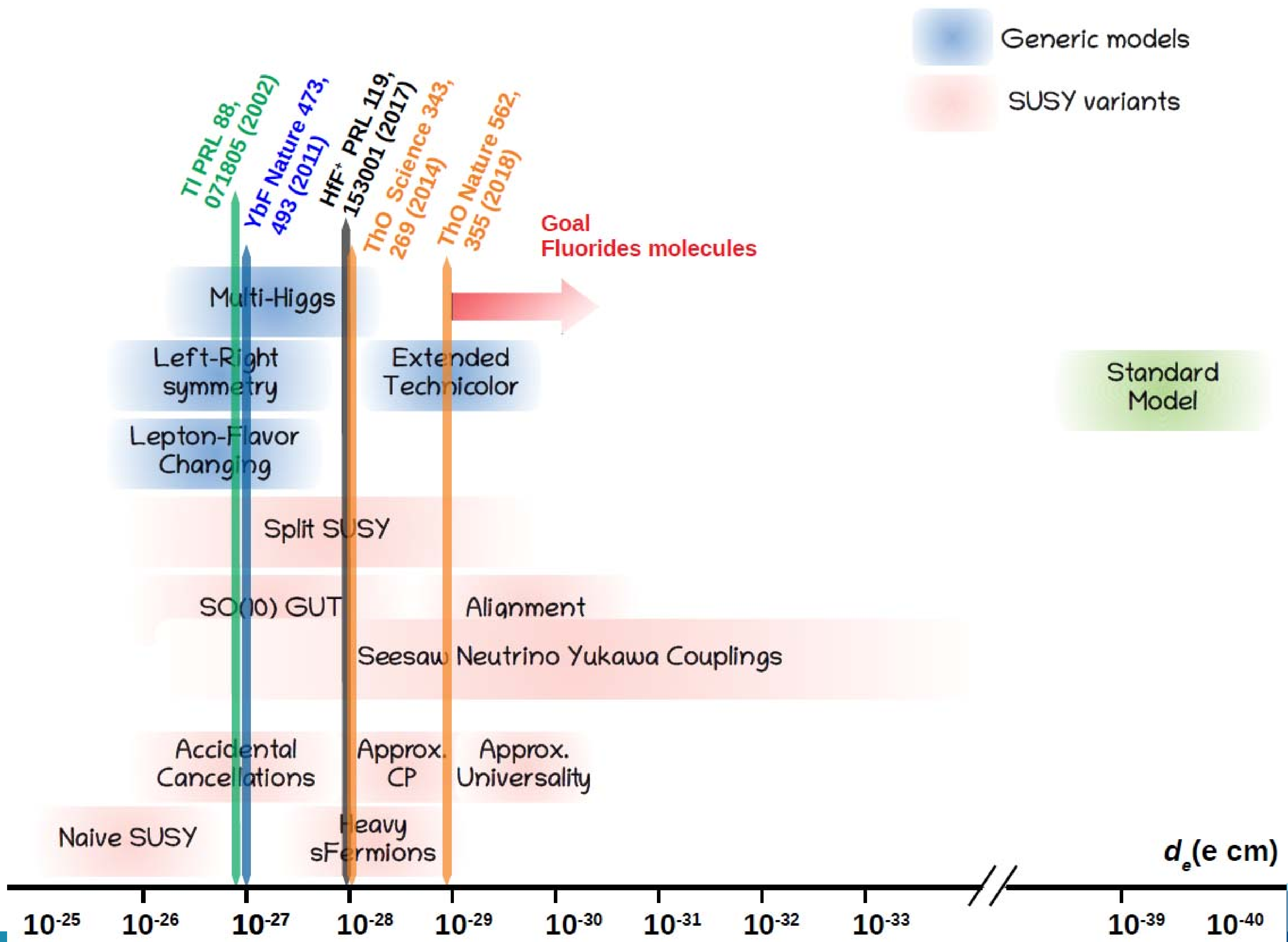
- Instrumentation developments
  - Obtaining high efficiency (RIB), improved precision, sensitivity

  (K. Flanagan, R. Ferrer, N. Lecesne, D. Verney, W. Nörtershäuser)

# Radium fluoride (RaF) spectroscopy




First Experimental Spectroscopy
➔ Transitions observed
➔ Suitable scheme for laser cooling established

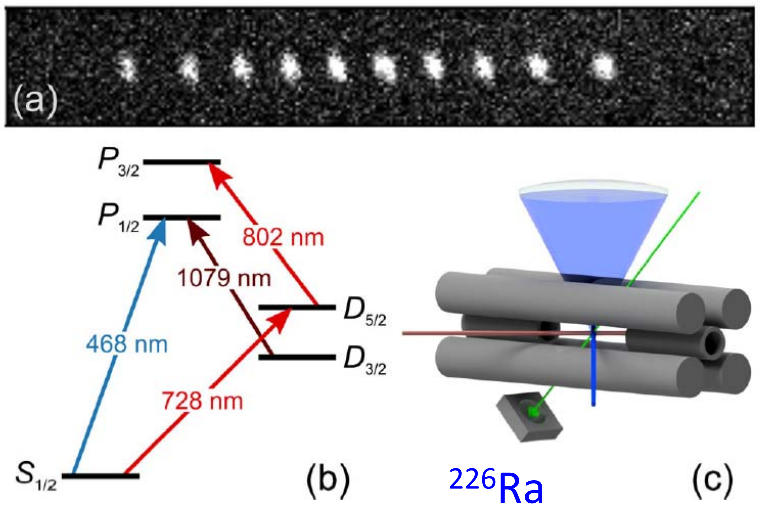


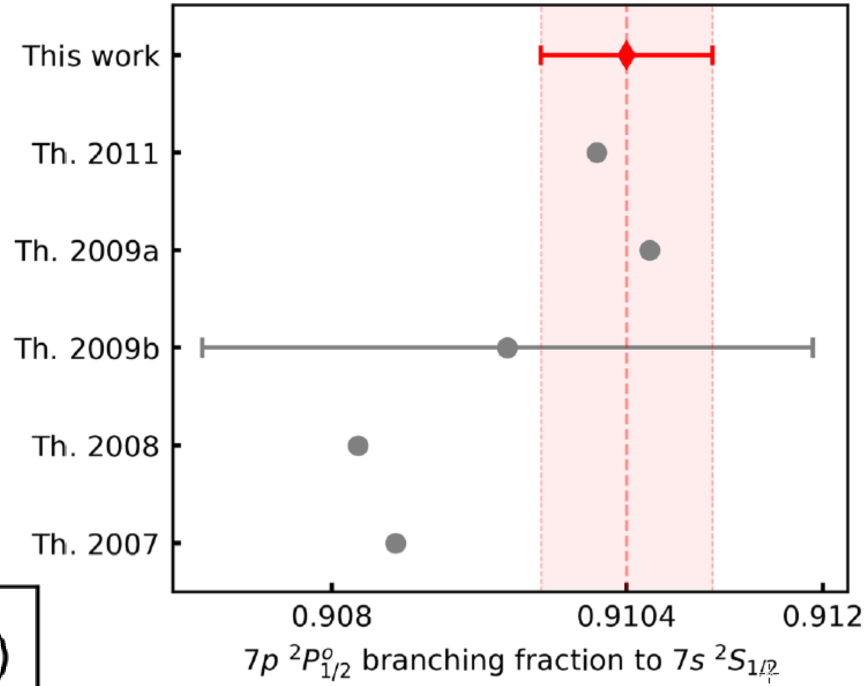

Courtesy: R. Garcia Ruiz - Garcia Ruiz et al. submitted to Nature (2019)

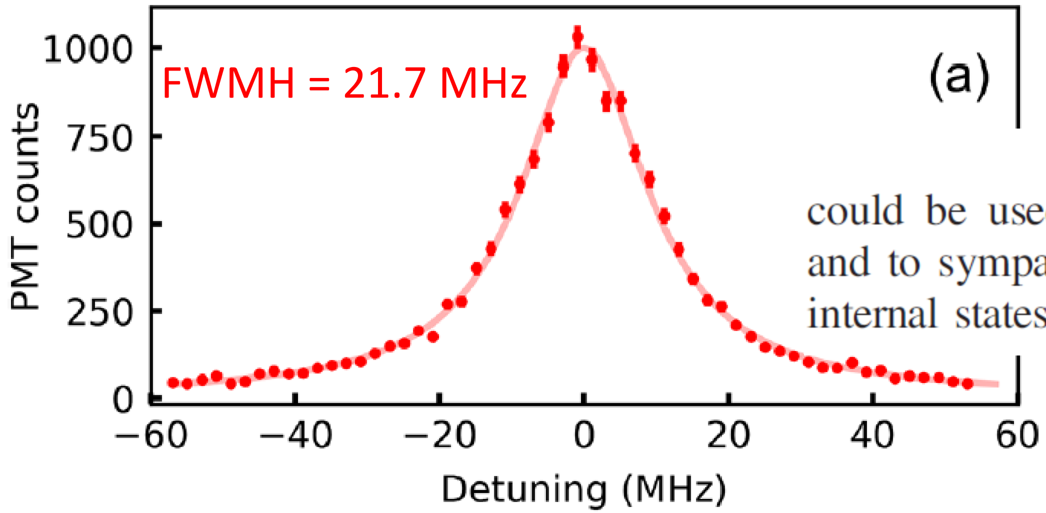
# Laser Cooling of Radium Ions

M. Fan, C. A. Holliman, A. L. Wang, and A. M. Jayich

PHYSICAL REVIEW LETTERS **122**, 223001 (2019)




Cold $Ra^+$ could be used to make molecular ions such as $RaOH^+$, and to sympathetically cool their motion and control their internal states with quantum logic spectroscopy [42].

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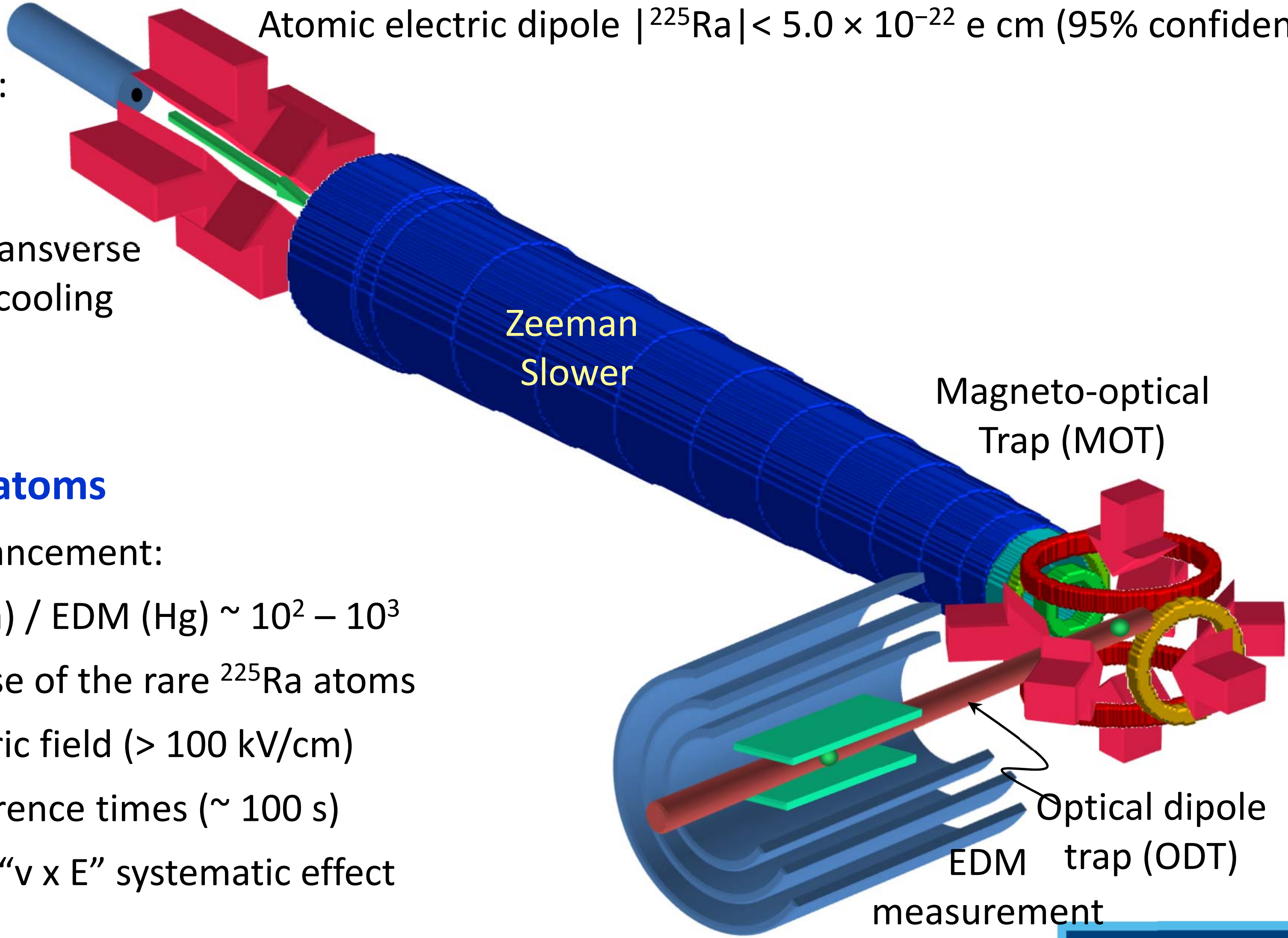
# EDM measurement on $^{225}$Ra

P. Müller, ANL

Atomic electric dipole $|^{225}\mathrm{Ra}| < 5.0 \times 10^{-22}$ e cm (95% confidence).




## Trap $^{225}$Ra atoms

- Large enhancement:

  EDM (Ra) / EDM (Hg) ~ $10^2 - 10^3$
- Efficient use of the rare $^{225}$Ra atoms
- High electric field (> 100 kV/cm)
- Long coherence times (~ 100 s)
- Negligible "v x E" systematic effect

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R.H. Parker, …, P. Müller,- PRL (2015)

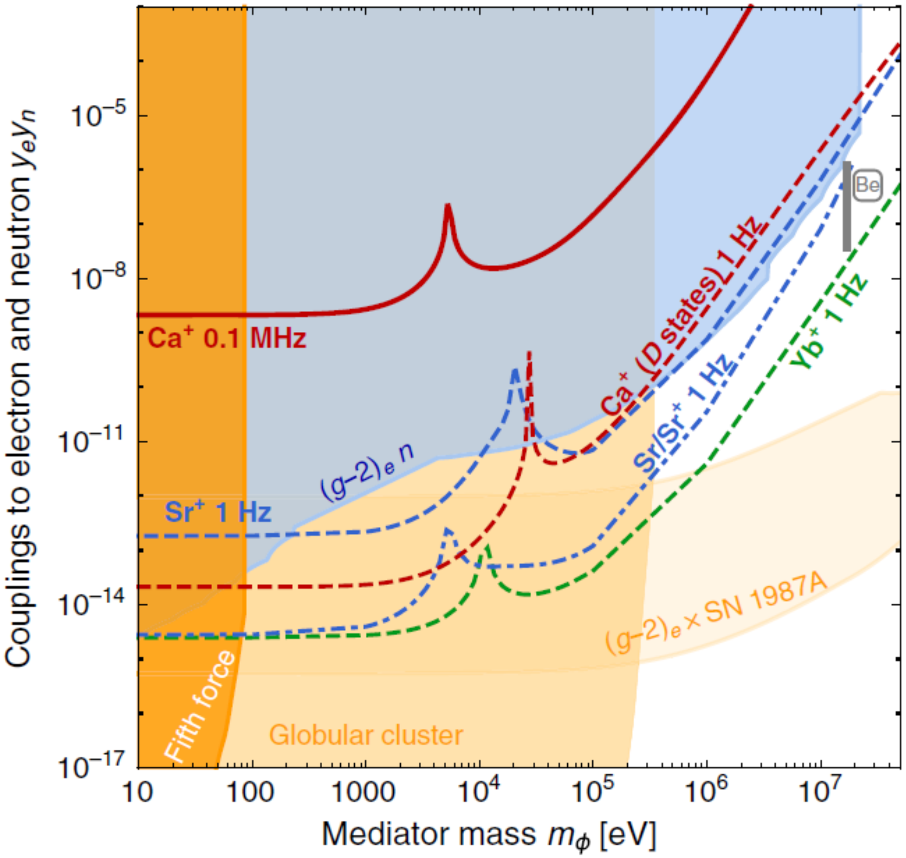
- The road to high (ultimate) resolution/precision

**Probing New Long-Range Interactions by Isotope Shift Spectroscopy**

Julian C. Berengut,[1,*] Dmitry Budker,[2,3,4,†] Cédric Delaunay,[5,‡] Victor V. Flambaum,[1,§] Claudia Frugiuele,[6,∥] Elina Fuchs,[6,¶] Christophe Grojean,[7,8,**] Roni Harnik,[9,††] Roee Ozeri,[10,‡‡] Gilad Perez,[6,§§] and Yotam Soreq[11,∥∥]

Physical Review Letters 120 (2018)

FIG. 1. Limits on the electron and neutron couplings ($y_e y_n$) of a new boson of mass $m_\phi$ (for the experimental accuracies $\sigma_i$ specified on the labels). Constraint from existing IS data: $Ca^+$ (397 vs 866 nm [19], the solid red line). IS projections (the dashed lines) for $Ca^+$ ($S \to D$ transitions), $Sr^+$, $Sr/Sr^+$, and $Yb^+$. For comparison, existing constraints from other experiments are shown as shaded areas: fifth force (dark orange) [20,21], $(g-2)_e$ [22,23] combined with neutron scattering (light blue) [24–27] or SN 1987A (light orange) [28], and star cooling in globular clusters (orange) [29–33]. The gray line at 17 MeV indicates the $y_e y_n$ values required to accommodate the Be anomaly [34,35].




See also:

- T. Manovitz,- arXiv:1906,05770v
- C. Delaunay,- PRD 96 (2017)
- C. Frugiuele,- PRD 96 (2017)

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# Laser Spectroscopy Observables - impact

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  (Mark Bissel, Dag Hanstrop, Mustapha Laatiaoui, Deyan Yordanov)

- Nuclear physics
  - $\delta\langle r^2\rangle$, $\mu$, $Q_s$ and $I^\pi$
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  - Ex. : shape coexistence in Hg isotopes,

  (Kieran Flanagan, Kei Minamisono, Iain Moore, Peter Müller, Gerda Neyens, Wilfried Nörtershäuser, Deyan Yordanov, Mark Bissell)

- Fundamental physics
  - Use of the high (ultimate) precision
  - Molecular ions

  (Gerda Neyens, Iain Moore)

- Instrumentation developments
  - Obtaining high efficiency (RIB), improved precision, sensitivity, selectivity (isomer)

  (Kieran Flanagan, Rafael Ferrer, Nathalie Lecesne, David Verney, Wilfried Nörtershäuser)

# Laser Spectroscopy: experimental requirement

- High spectral resolution
- High efficiency/sensitivity (weak production)
- Fast measurement cycle (short-lived radioactive isotopes)
- High accuracy

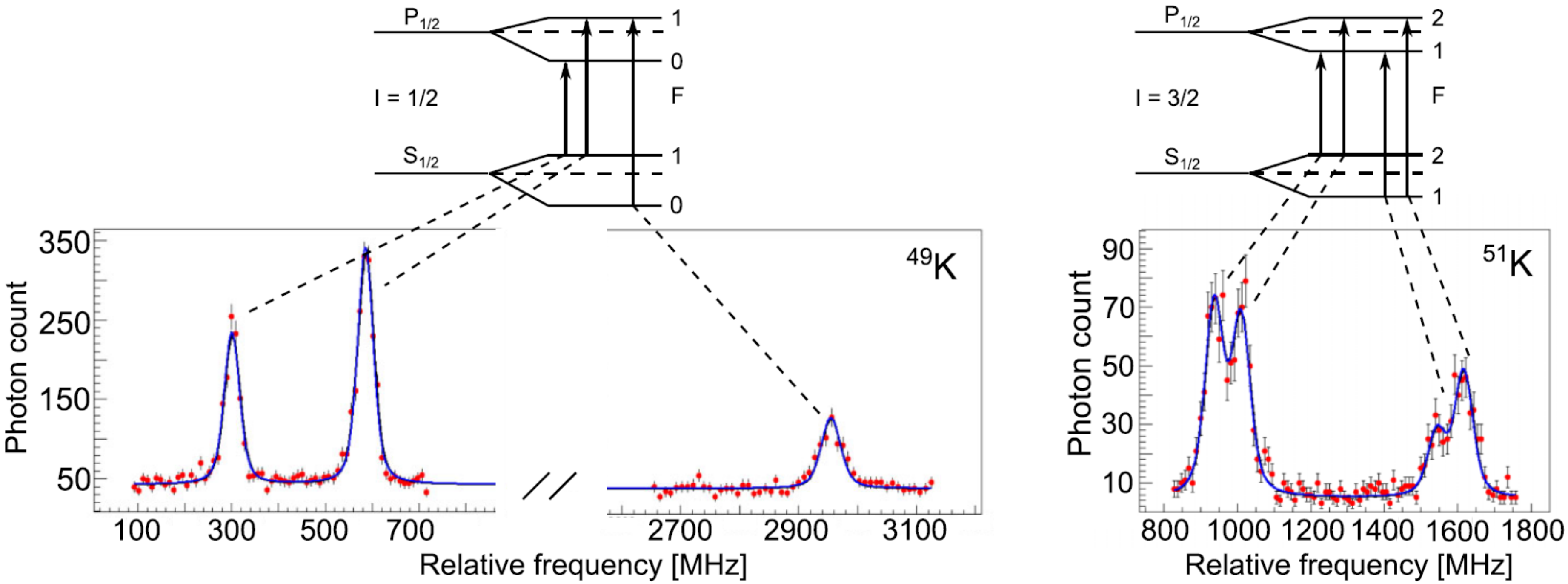
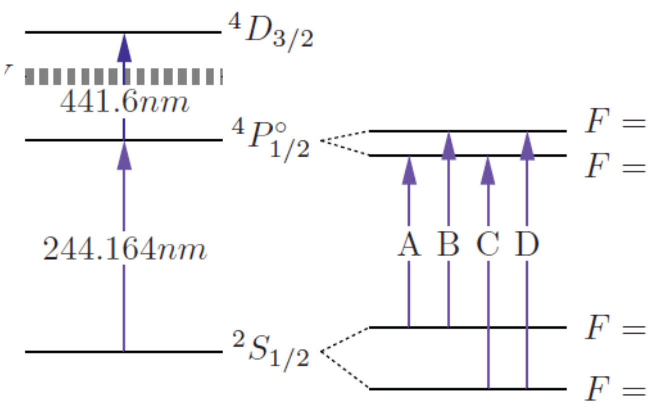
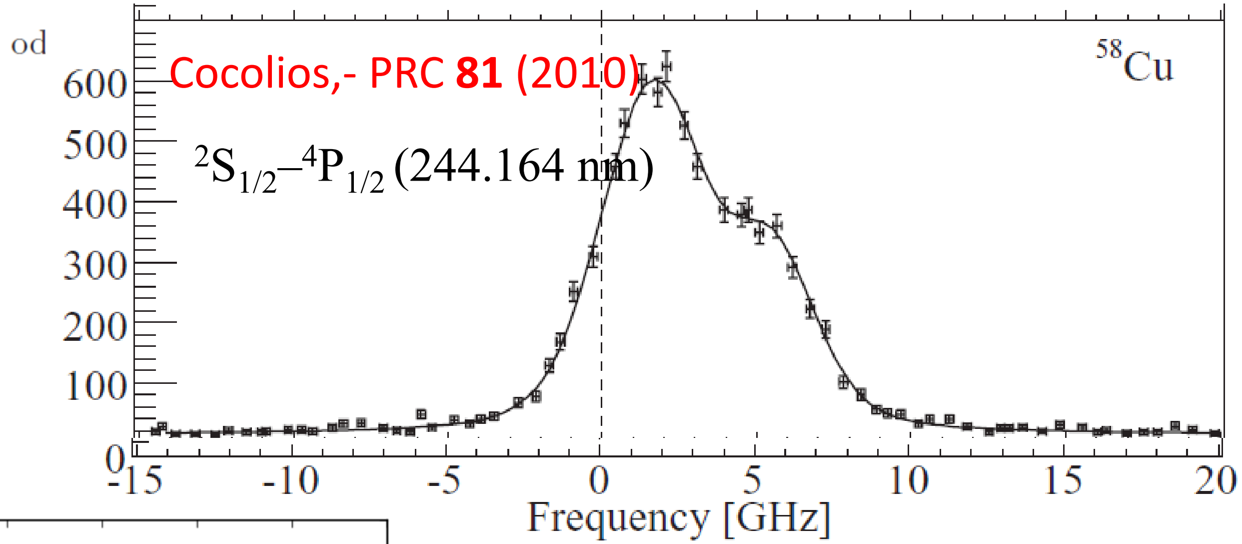


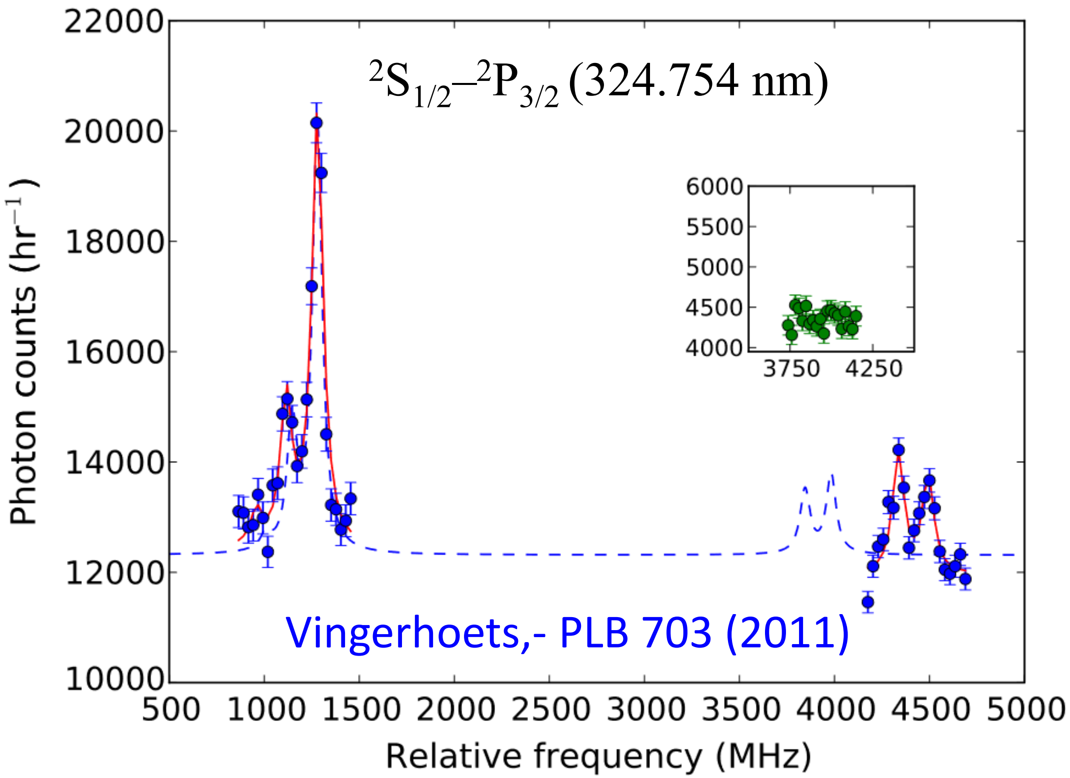

TABLE I. Fitted hyperfine parameters for the studied isotopes (assuming different spins for $^{51}$K).

| Isotope | $I$ | $A(^2S_{1/2})$ (MHz) | $A(^2P_{1/2})$ (MHz) |
|---|---|---|---|
| $^{49}$K | 1/2 | +2368.2 (14) | +285.6 (7) |
| $^{51}$K | 3/2 | +302.5 (13) | +36.6 (9) |

J. Papuga,- PRL110, 172503 (2013)




| $A$ | $I$ | $A_{hf:exp:gs}$ (GHz) |
|---|---|---|
| 58 | $1^+$ | 1.891(52) |

$A(^2S_{1/2})/A(^4P_{1/2}) = 2.4$

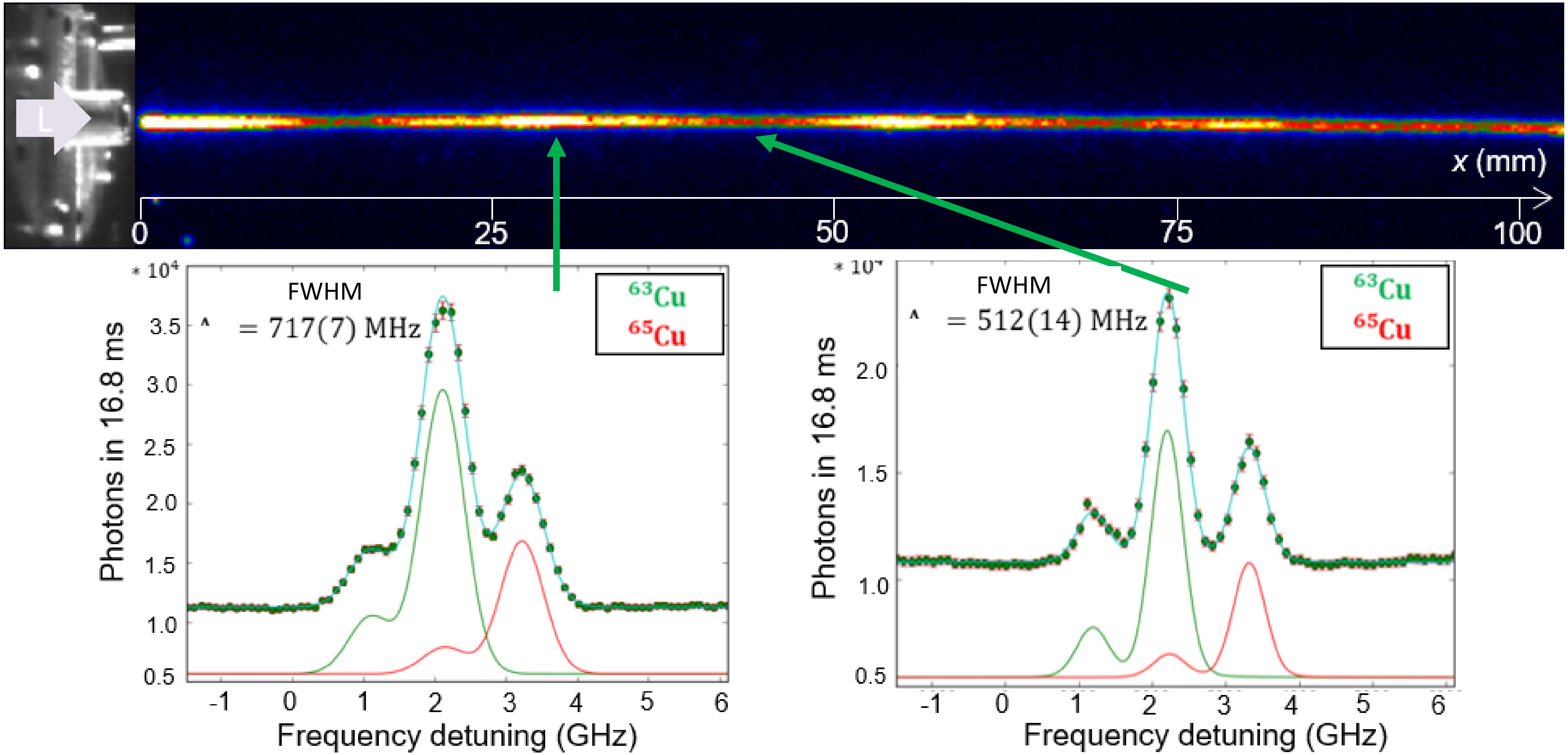
| Isotope | $I^\pi$ | $A(^2S_{1/2})$ (MHz) |
|---|---|---|
| $^{58}$Cu | $1^+$ | +2257(9) |

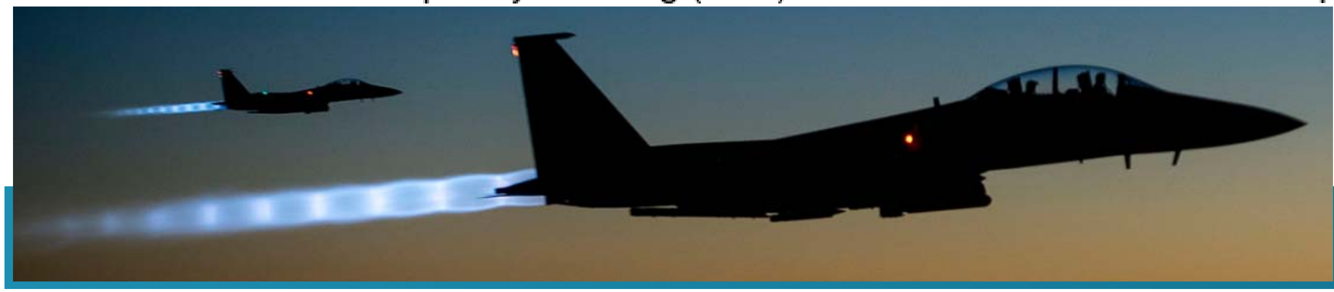
$A(^2S_{1/2})/A(^2P_{3/2}) = 29.3$

# Jets formation with a de Laval nozzle

Narrowband PLIF-spectroscopy of $^{63,65}$Cu

Central line of underexpanded jet ($P_{bg} < P_{opt}$)



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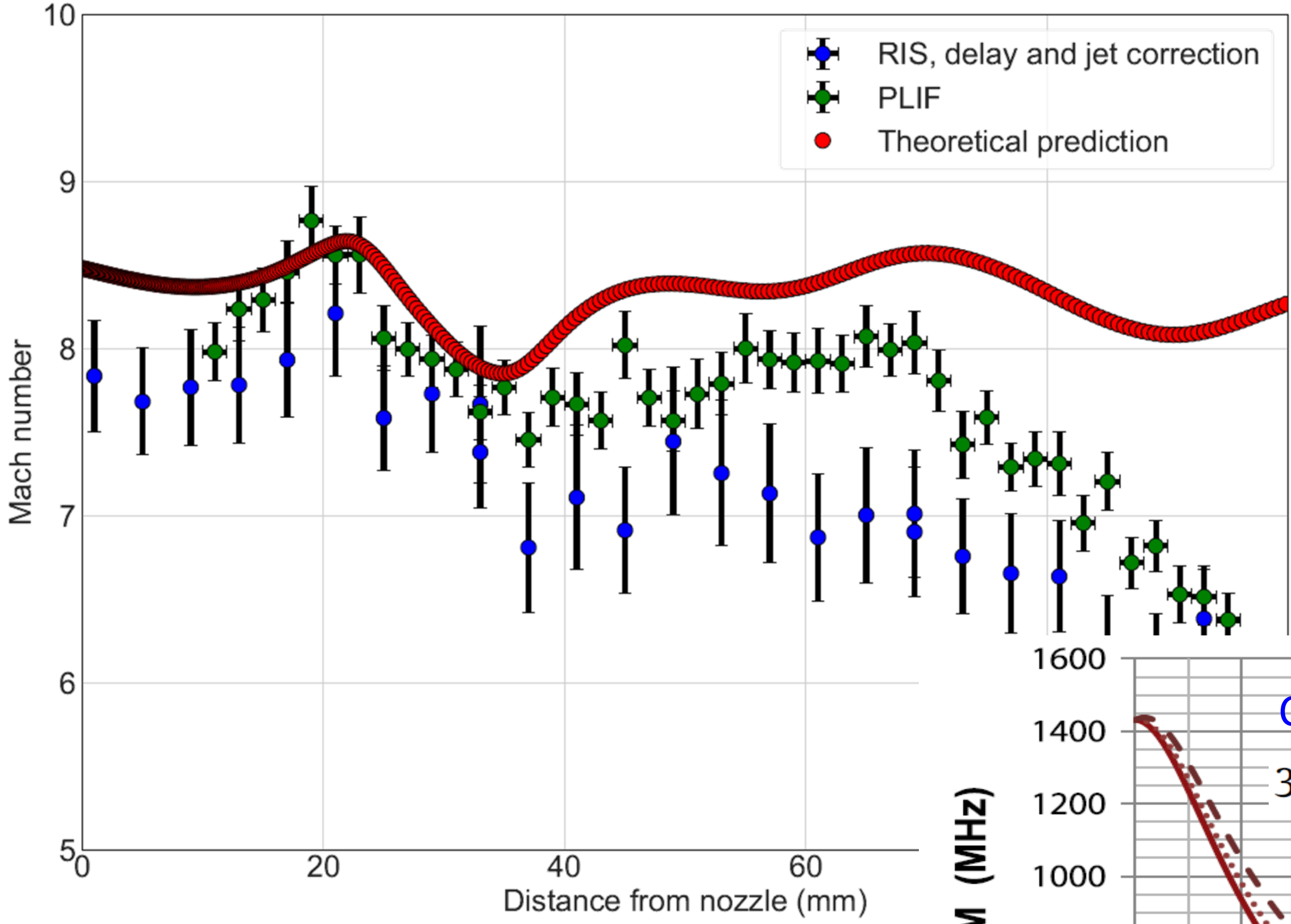
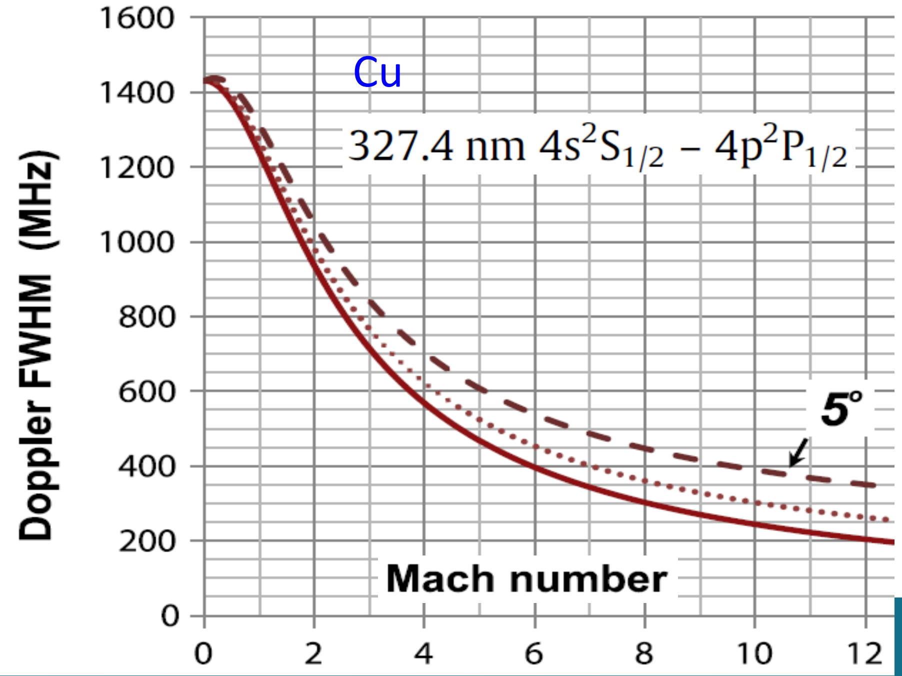
# Gas jet developments



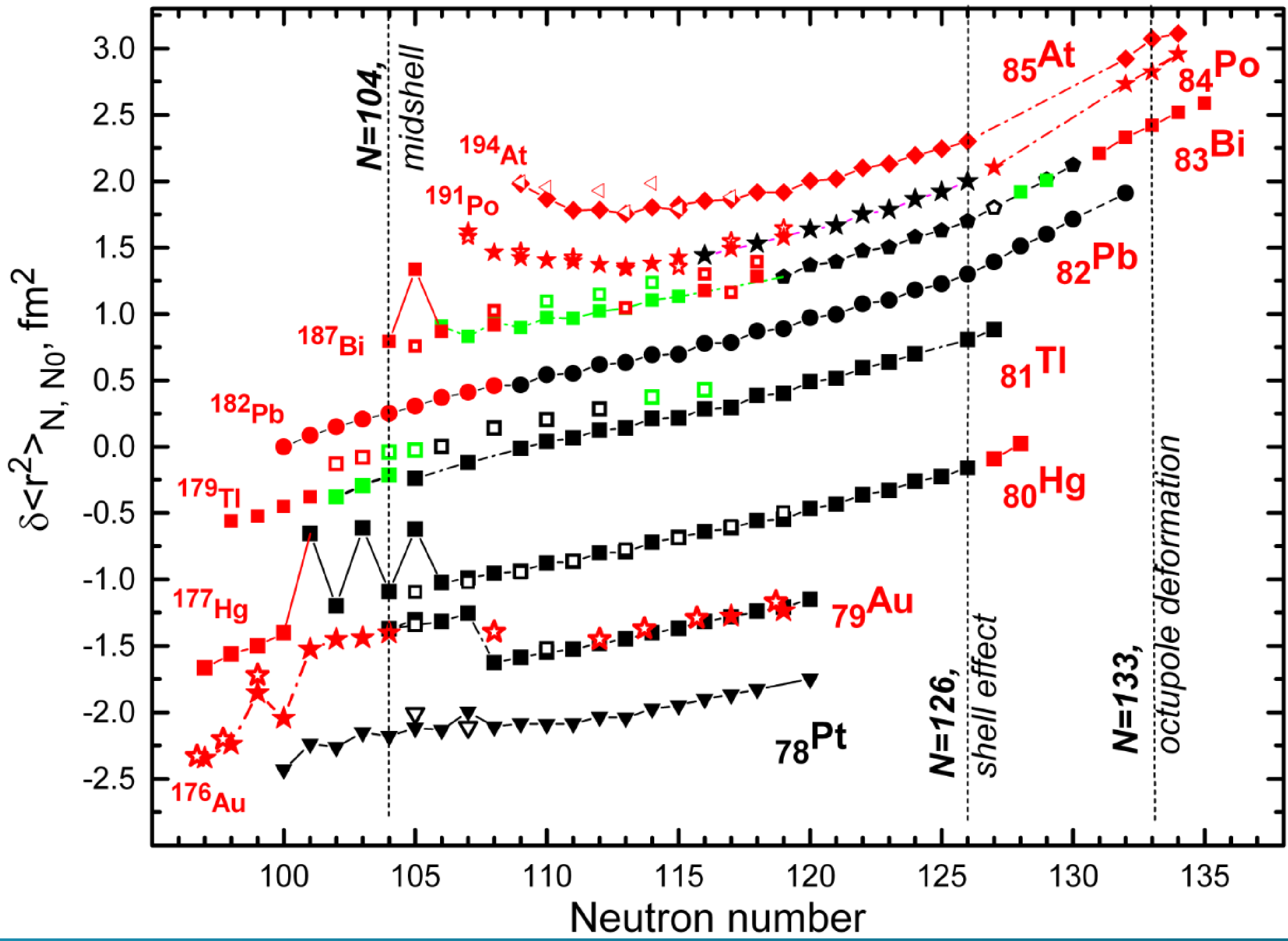

Kudryavtsev,- NIMB 297 (2013)




- Calculations of Mach number compared to experimental data (RIS and PLIF)

See: R. Ferrer

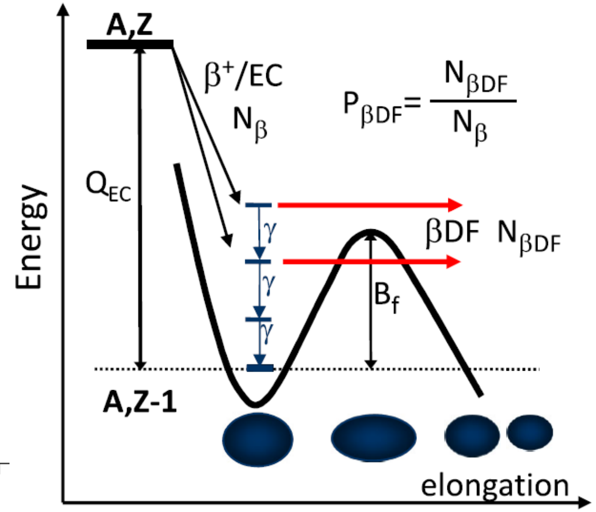
# Laser Ionization: pure / isomeric beams

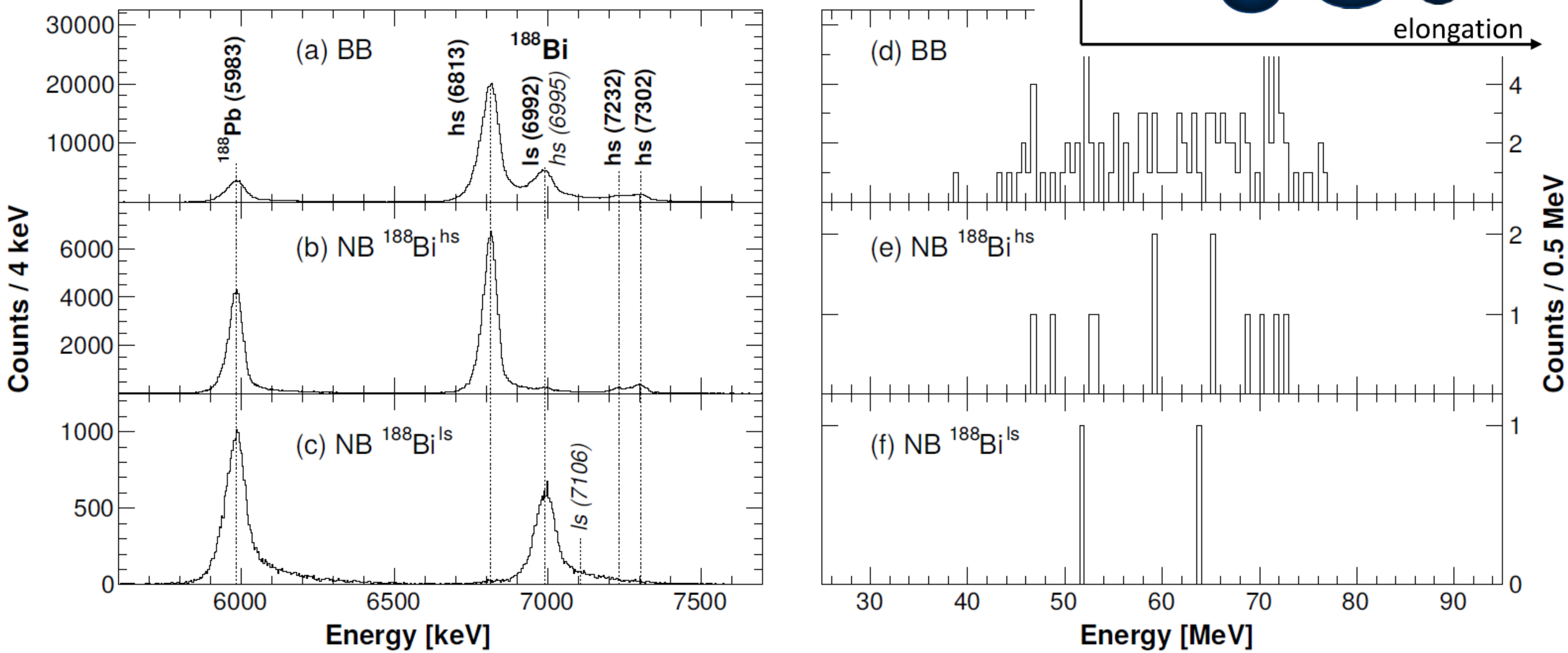


# Laser Ionization: pure / isomeric beams

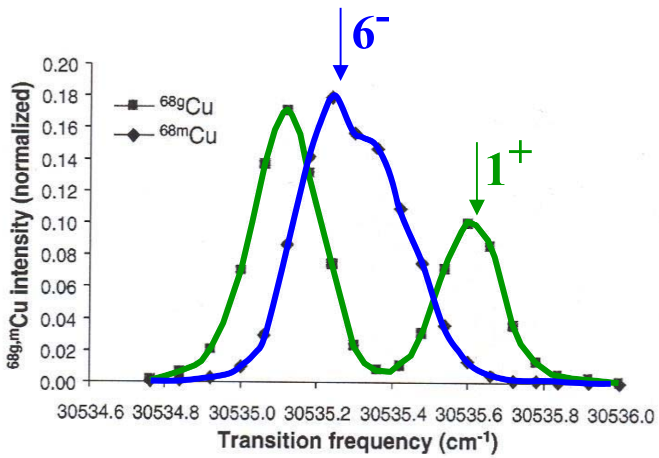
- Beta-delayed fission of $^{188m,g}Bi$

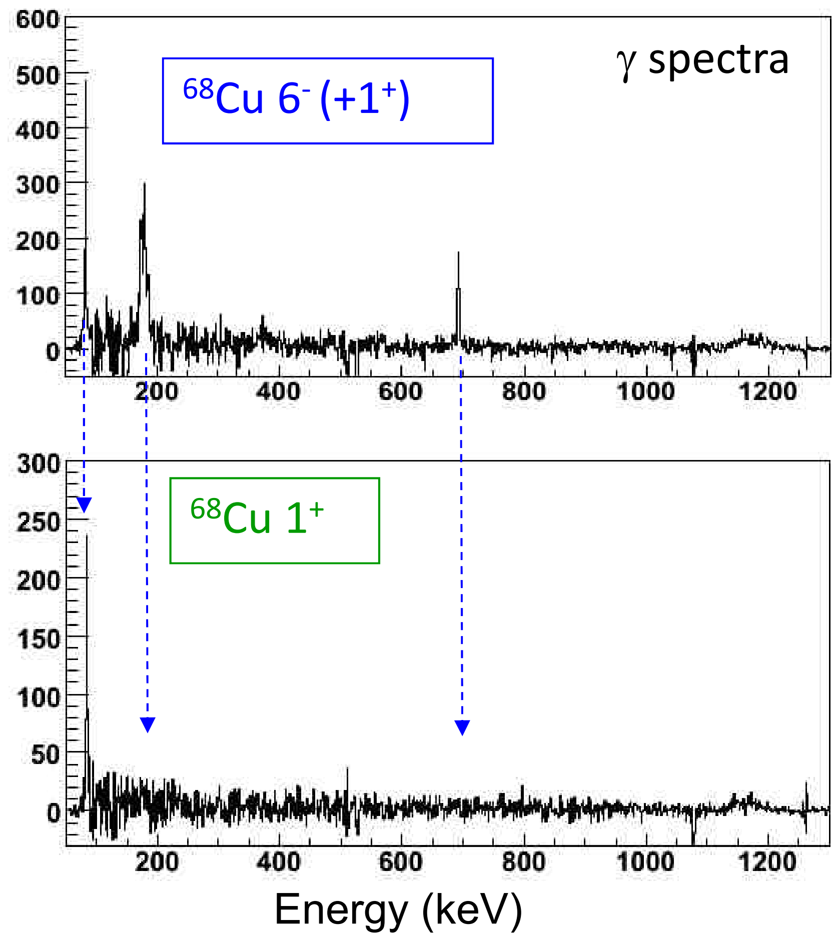
| $J^{\pi}$ | Isomer | $T_{1/2p,\beta DF}$ [s] |
|---|---|---|
| $10^{-}$ | $^{188}Bi^{hs}$ | $5.6(8)\times10^{3}$ |
| $1^{+}$ | $^{188}Bi^{ls}$ | $1.7(6)\times10^{3}$ |



- Isomeric beams for Coulomb excitation

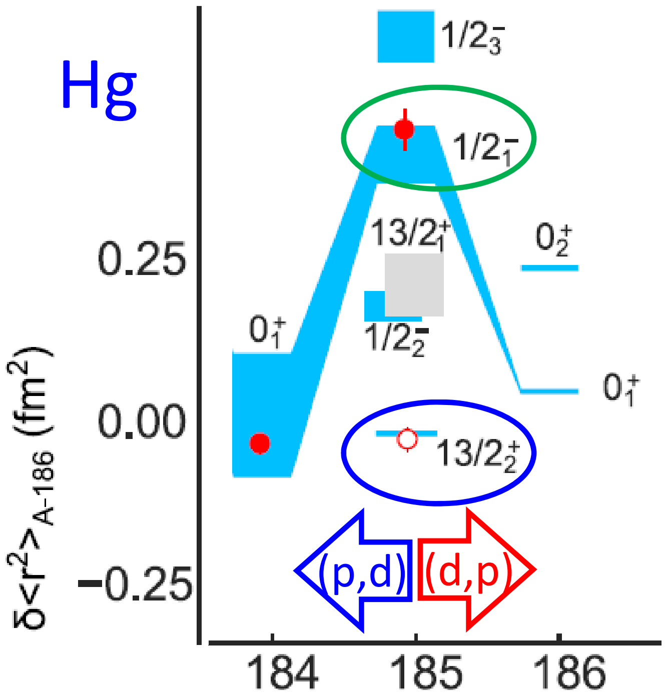


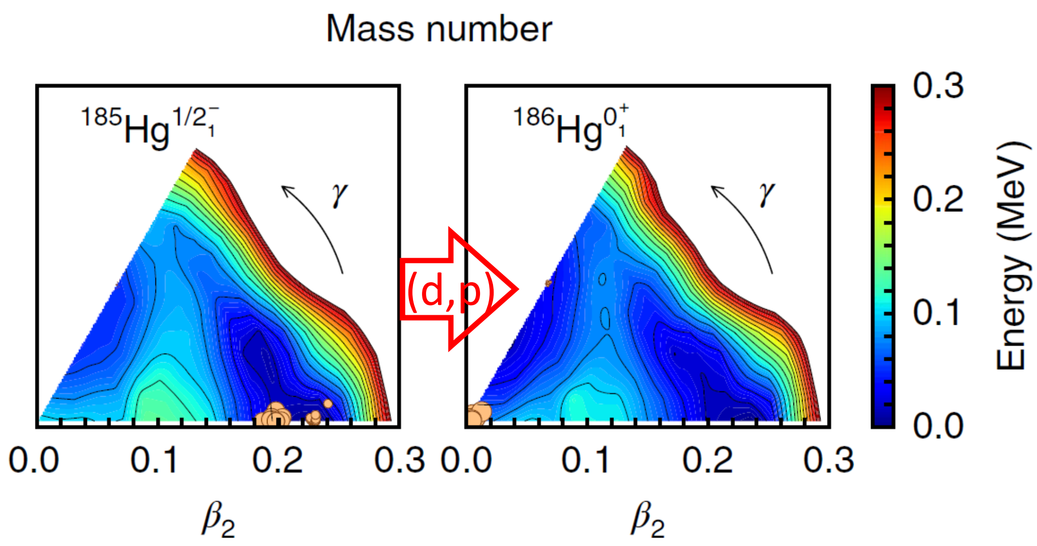



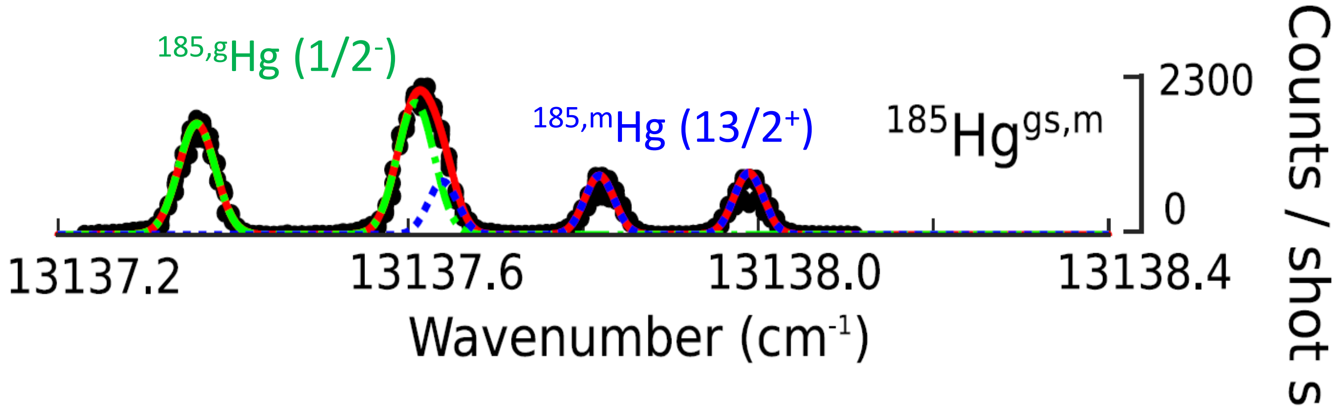

➢ $^{68m,g}$Cu (2.86 MeV/u, 3 $10^5$ pps, 74% pure) @ $^{120}$Sn (2.3 mg/cm$^2$)

I. Stefanescu,- PRL 98 (2007)

- One/two-nucleon transfer reaction: $^{185}$Hg



# Conclusion and outlook

- Laser spectroscopy of RIB is a powerful tool to provide accurate information on a number of observables of interest to atomic physics, nuclear physics and fundamental interaction studies – should be combined with information from other observables to get a global picture. “The results stay in the literature …” (J. Kluge)

- Laser ionization is an excellent tool to produce pure, intense radioactive ion beams (including isomeric beams) for physics studies

- Developments to implement laser spectroscopy focus on improving the efficiency (low production rate), sensitivity (to specific aspects), speed (short-lived isotopes) and spectral resolution

- There is a long tradition and expertise in laser-spectroscopy studies of radioactive isotopes (see e.g. EU sponsored networks and USA and Canadian efforts). New accelerators (or upgrades) are coming on-line, promising increased RIB intensities and capabilities. The know-how on laser spectroscopy of RIB should be consolidated and expanded to fullfill the interest from different fields of research.

- Thanks to G. Neyens, R. Garcia Ruiz, M. Laatiaoui, S. Raeder, W. Nörtershäuser,
K. Blaum, I. Moore

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