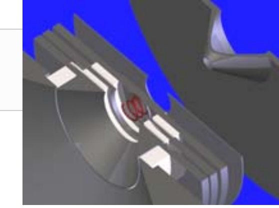
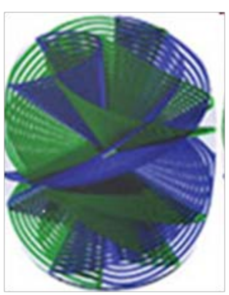
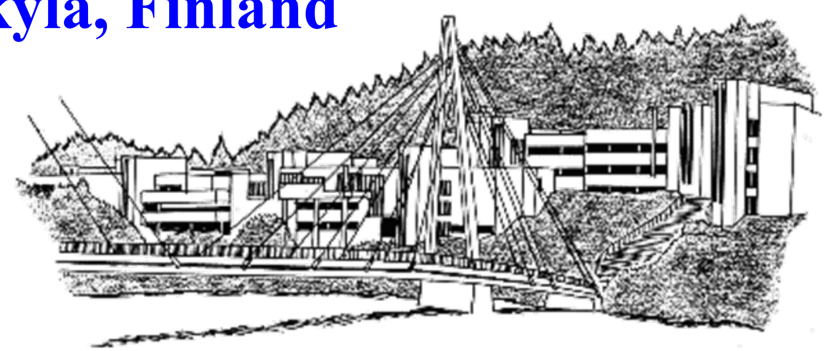


Towards the study of heavy elements with resonance ionization spectroscopy at IGISOL

Iain Moore

University of Jyväskylä, Finland

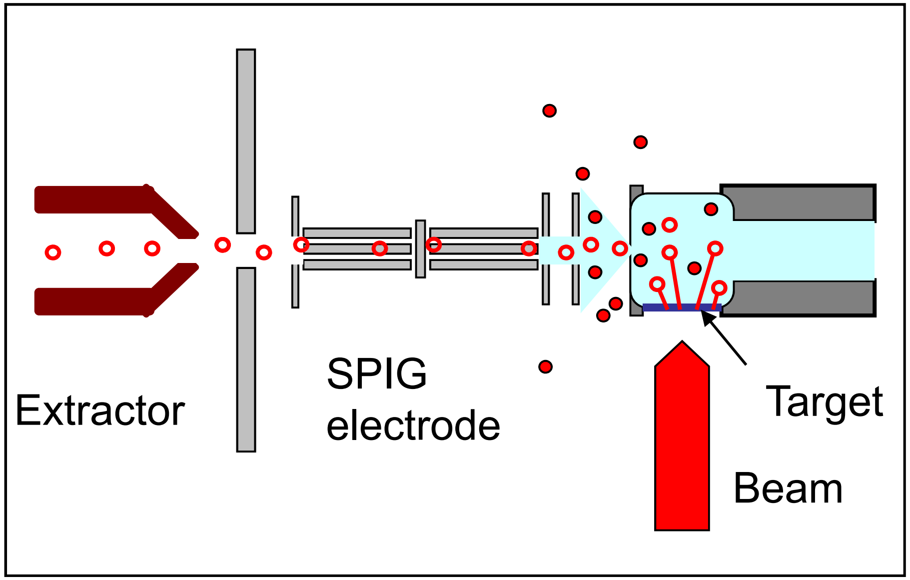




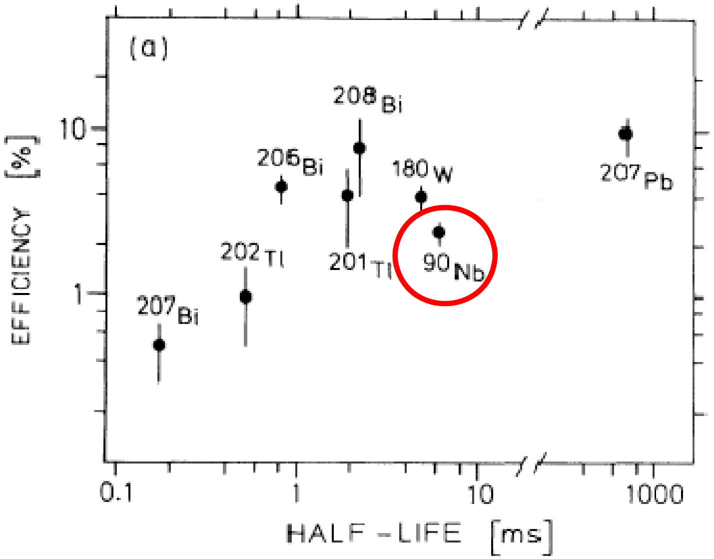
Outline

- IGISOL – a brief introduction
- Motivation: spectroscopy of bismuth and uranium (thorium)
- Experimental efforts
- Outlook

Ion guide principle: a universal production method



- Based on the survival of primary ions in helium buffer gas
- Charge state concentration: (0), +1 (+2)
- Fast gas flow required to prevent neutralization
- Produces ions of any element

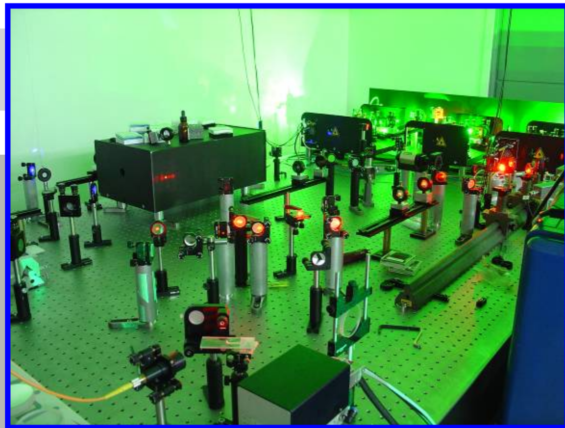


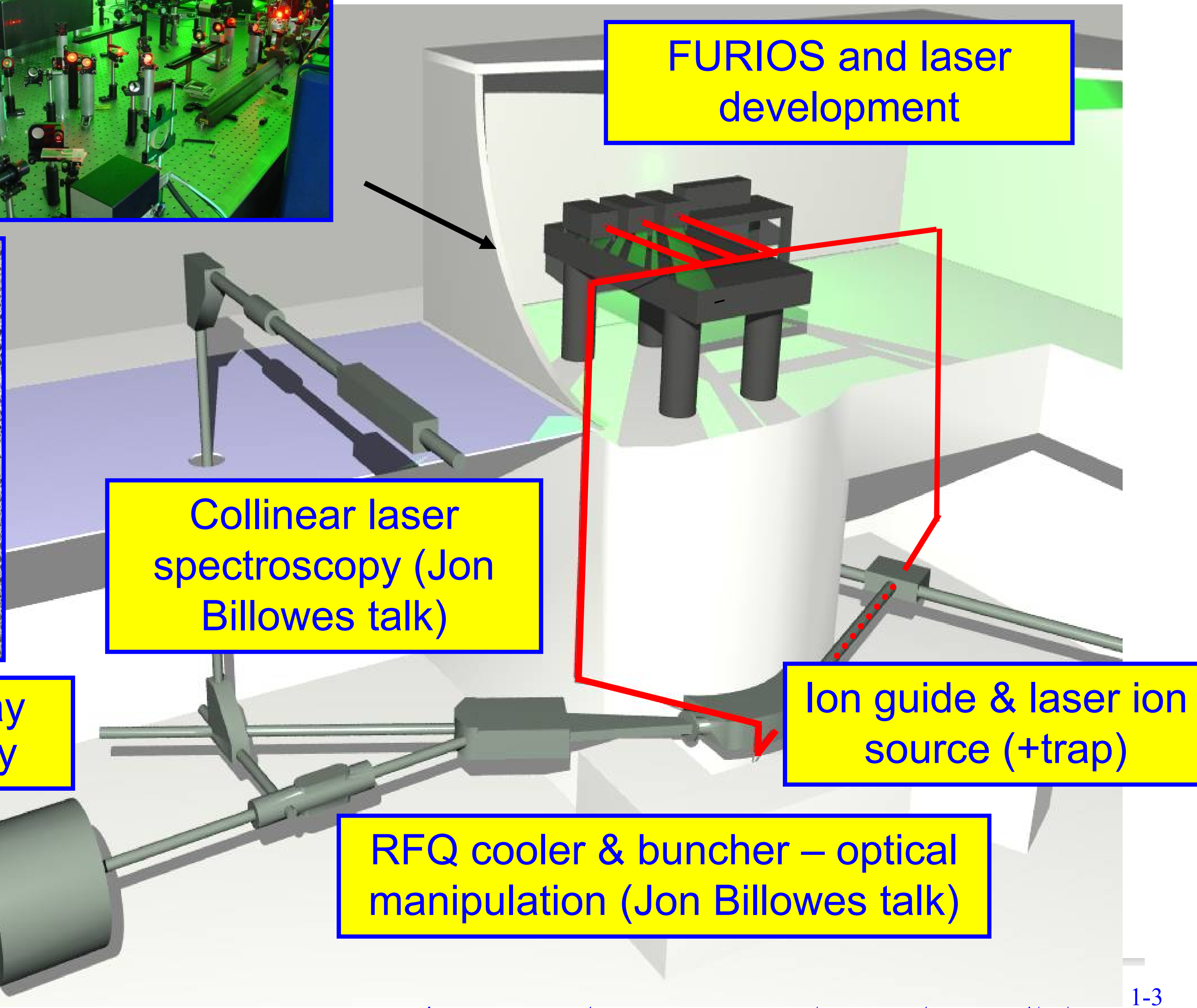
J. Ärje *et al.*, Phys. Rev. Lett. 54 (1985) 99

More advanced gas catchers
☺ dc fields
☺ rf carpets, funnels and walls
☺ cryogenic temperatures
☺ selective laser ionization

☹ molecular formation
☹ space charge, recombination

The IGISOL facility at JYFL





Outline

- IGISOL – a brief introduction
- **Motivation: spectroscopy of bismuth and uranium (thorium)**
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Nuclear ground state properties…

…by atomic spectroscopy

Isotope Shift (IS)

↓

Mean Square Charge Radii

$$\delta\langle r^2\rangle^{AA'}$$

Hyperfine Structure (HFS)

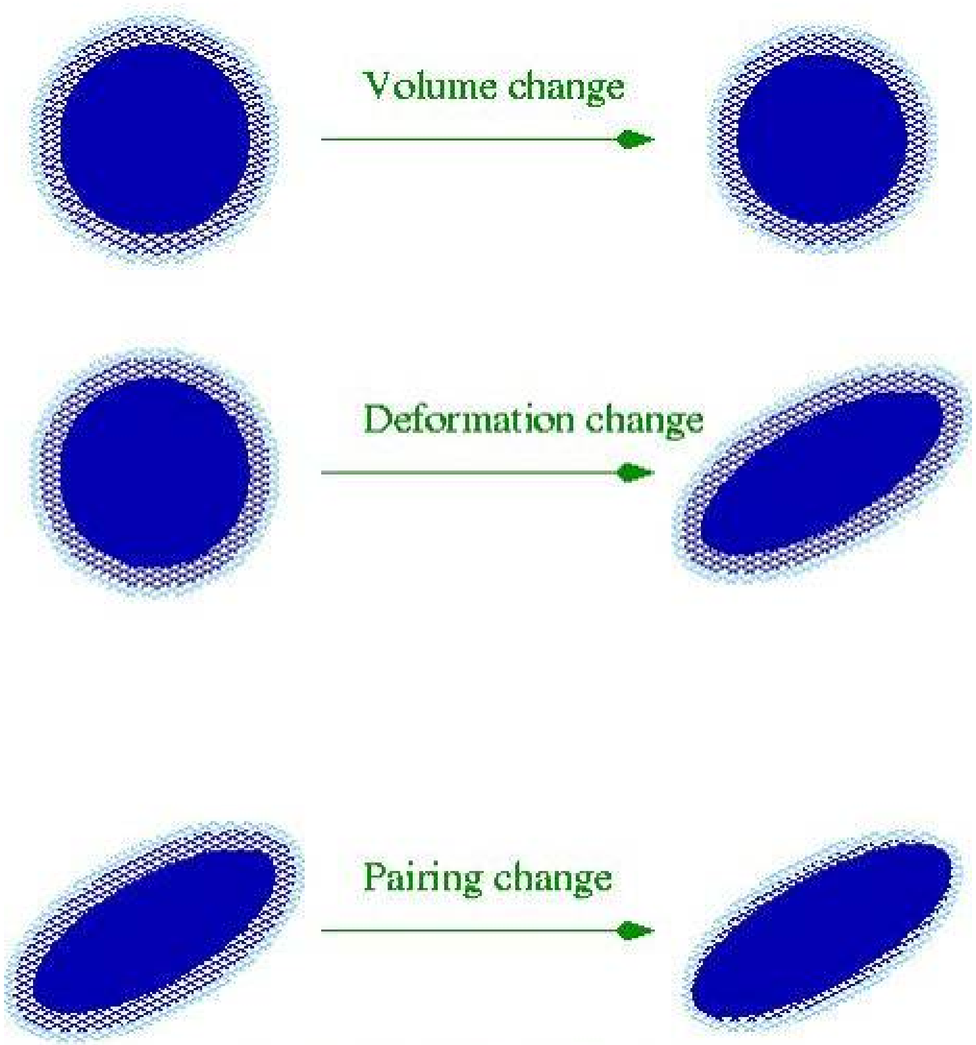
↓

Nuclear Spin I
Magnetic Dipole Moment μ_I
Electric Quadrupole Moment Q_s
Hyperfine Anomaly

Sample preparation is crucial.
Nuclear reaction products must be slowed and thermalized quickly, efficiently, universally and selectively.

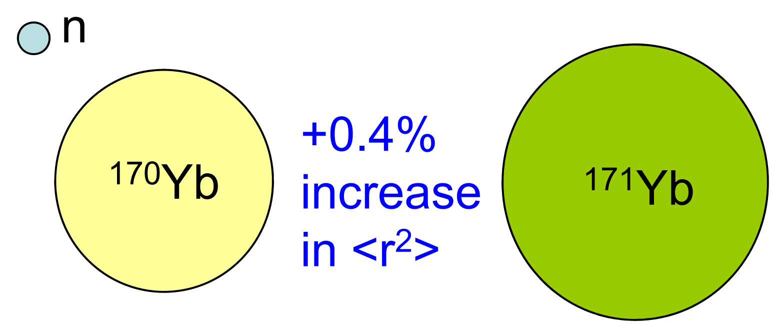
Special nuclear interest in the heavier systems…

Factors controlling δ<r²>



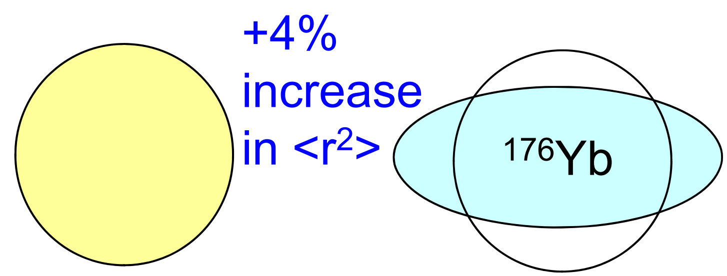
(change in surface diffuseness)

Volume change



(small but easily detectable
with laser spectroscopy)

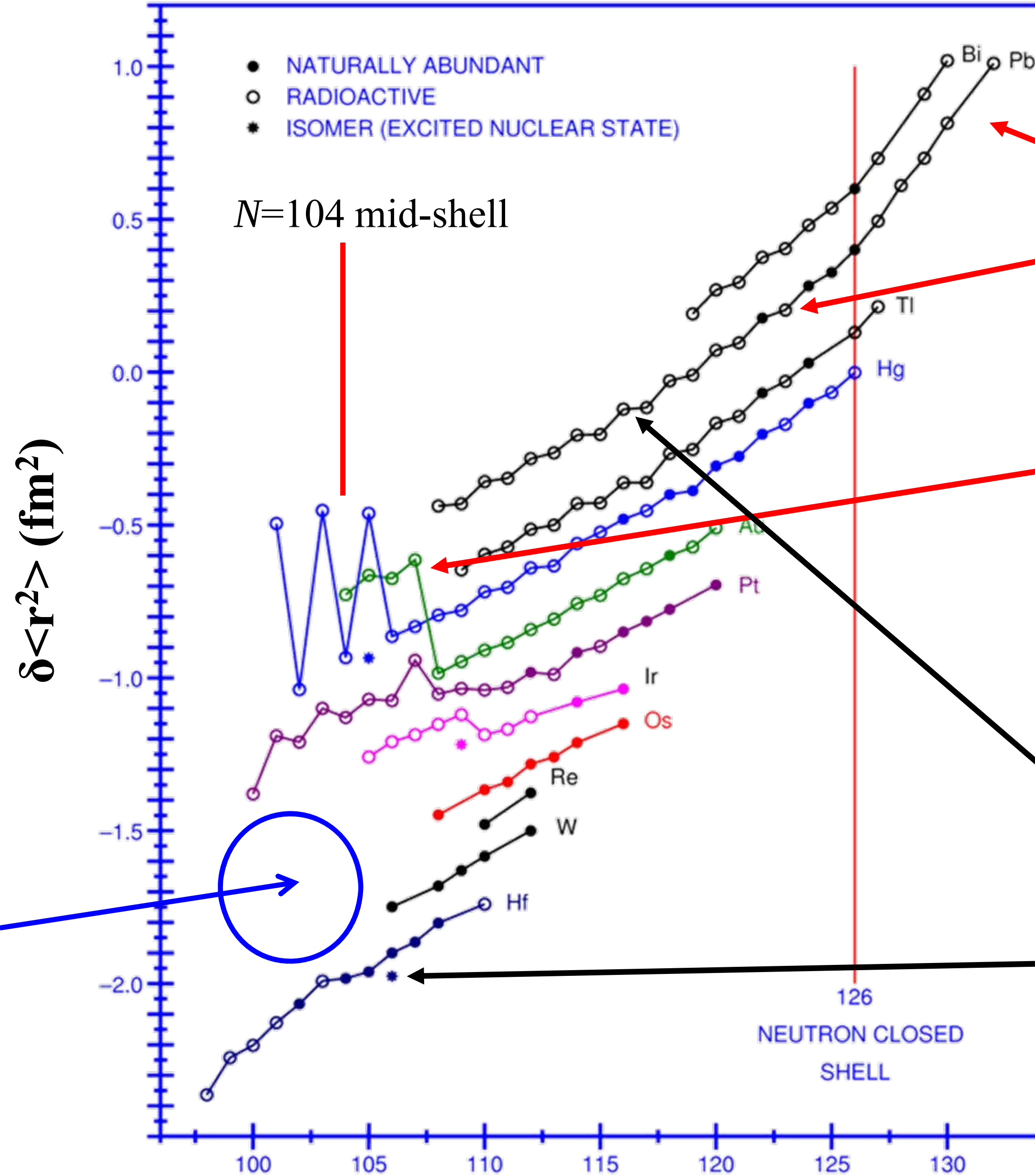
Shape change



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

contributions may be STATIC (nuclear shape)
or DYNAMIC (fluctuations on the nuclear surface)

Nuclear physics: the $\delta\langle r^2\rangle$ landscape



No studies yet in this refractory region.

Static effects

Volume

Deformation

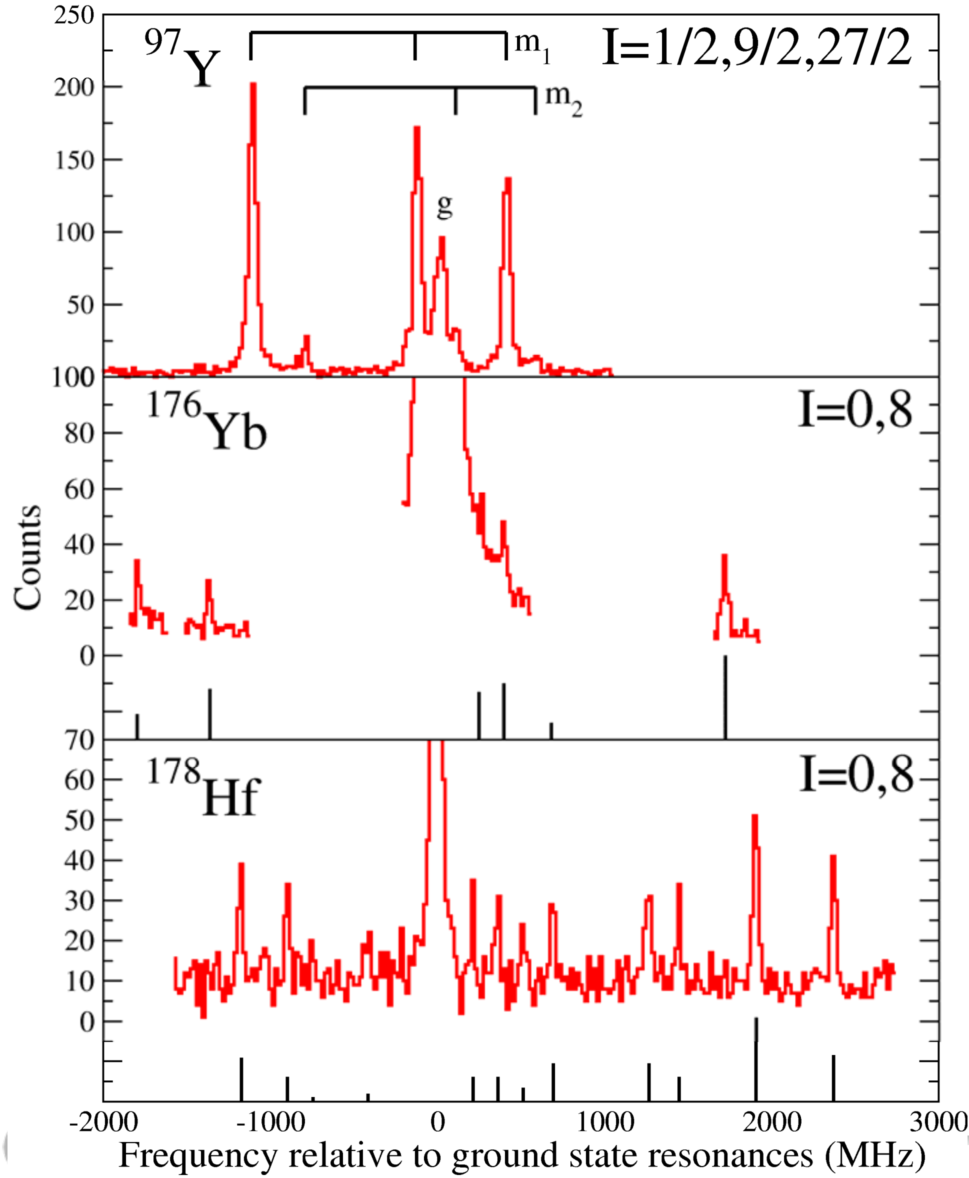
Dynamical effects

Odd-even staggering

Isomer shifts

Laser spectroscopy of multi-quasiparticle isomers

M.L. Bissel *et al.*, Phys. Lett. B 645 (2007) 330



A\Y	I	μ (μ_N)	Q_s (b)	Q_0 (b)	$\delta\langle r^2\rangle^{97m1,A}$
97^{m1}	$\frac{9}{2}$	+5.88(2)	−0.76(8)	−1.40(15)	
97^{m2}	$(\frac{27}{2})$	+5.64(3)	−1.21(14)	−1.50(17)	−0.098(1)
A\Cs	I	μ (μ_N)	Q_s (b)	Q_0 (b)	$\delta\langle r^2\rangle^{135g,A}$
135^g	$\frac{7}{2}$	+2.73(1)	+0.03(2)	∼ 0	
135^m	$\frac{19}{2}$	+2.18(1)	+0.89(7)	+1.20(9)	−0.0081(13)
A\Ba	I	μ (μ_N)	Q_s (b)	Q_0 (b)	$\delta\langle r^2\rangle^{130g,A}$
130^g	0	—	—	+3.419(24)	
130^m	8	−0.043(28)	+2.77(30)	+3.95(43)	−0.0473(30)
A\Yb	I	μ (μ_N)	Q_s (b)	Q_0 (b)	$\delta\langle r^2\rangle^{176g,A}$
176^g	0	—	—	+7.30(13)	
176^m	8	−0.151(15)	+5.30(8)	+7.55(11)	−0.0224(1)
A\Lu	I	μ (μ_N)	Q_s(b)	Q_0 (b)	$\delta\langle r^2\rangle^{177g,A}$
177^g	$\frac{7}{2}$	+2.239(7)	+3.39(3)	+7.26(6)	
177^m	$\frac{23}{2}$	+2.308(11)	+5.71(5)	+7.33(6)	−0.035(<1)
A\Hf	I	μ (μ_N)	Q_s (b)	Q_0 (b)	$\delta\langle r^2\rangle^{178g,A}$
178^g	0	—	—	+6.961(43)	
178^{m1}	8	+3.10(1)	+4.99(4)	+7.11(6)	−0.0384(1)
178^{m2}	16	+8.16(4)	+6.00(7)	+7.20(8)	−0.0873(20)

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

$$Q_0=\frac{5Z\langle r^2\rangle_s}{\sqrt{5\pi}}\langle\beta_2\rangle\left(1+0.36\langle\beta_2\rangle\right)$$

Only 6 such isomeric systems have been measured so far. All show decreases in mean-square charge radii despite increases in Q_0.

Dynamic

$$\beta_{rms}^2 = \beta_{static}^2 + (\langle\beta^2\rangle - \langle\beta\rangle^2) \text{ reduction ?}$$

IS, B(E2) $\quad Q_s$

Why?

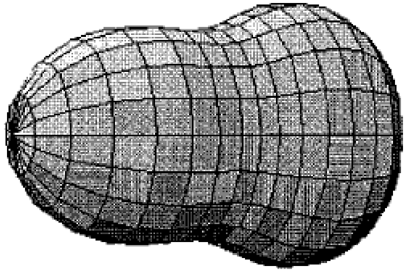
Reduction in surface diffuseness σ (loss of pairing) ?

One possibility is to measure spherical multi-quasiparticle isomers

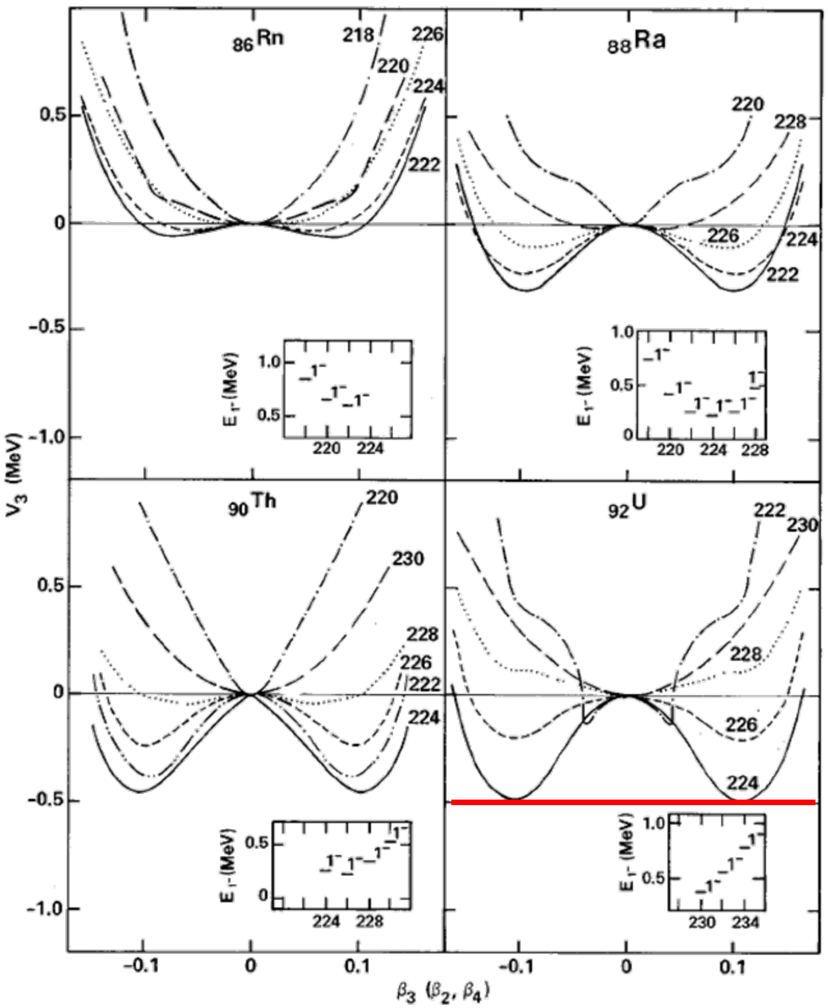
^{207}Bi ($21/2^+$, 182 μs)
^{204}Bi (10^-, 13 ms)
^{204}Bi (17^+, 1.07 ms)

The lifetimes of these systems become challenging to collinear laser spectroscopy, move to in-source spectroscopy.

Spectroscopy on n-deficient U isotopes

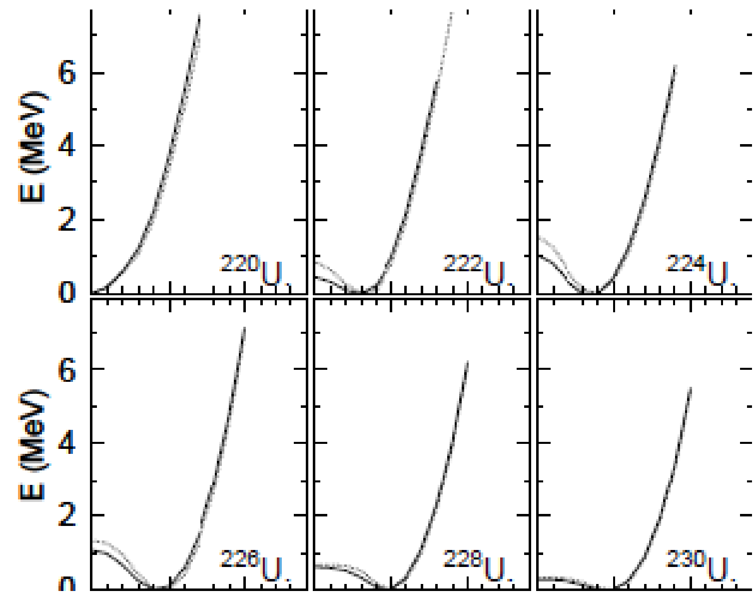


Motivation:



- Uranium isotopes with *N*=132,134 should have deepest PE surface minimum for non-zero octupole deformation.

0.5 MeV

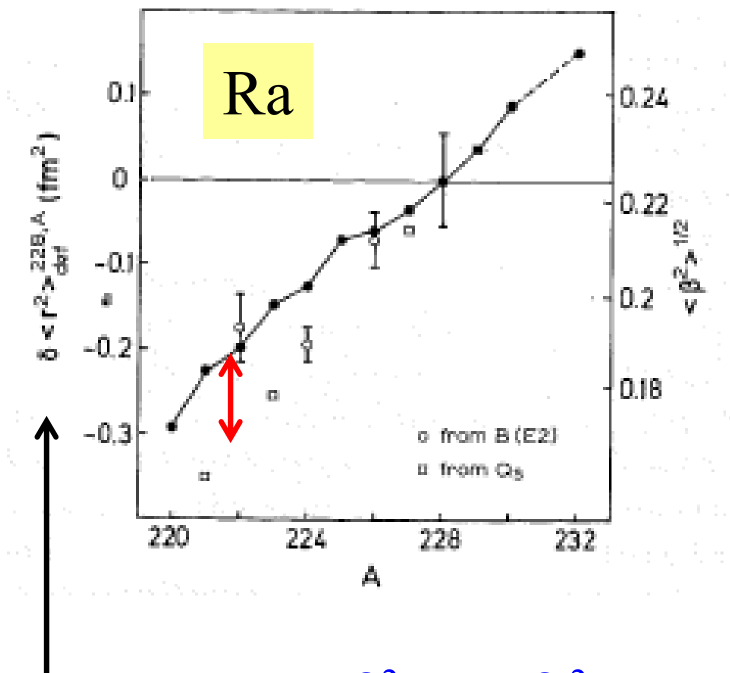


P.A. Butler and W. Nazarewicz, Nucl. Phys. A533 (1991) 249

Figure kindly provided by L. Robledo, Madrid

Earlier studies on Rn, Ra at ISOLDE (1980's)

$$< r^2 > = < r^2 >_{sph} \left(1 + \frac{5}{4\pi}\left(< \beta_2^2 > + < \beta_3^2 > + \dots\right)\right)$$



Represents $<\beta^2> = \Sigma_i <\beta_i^2>$

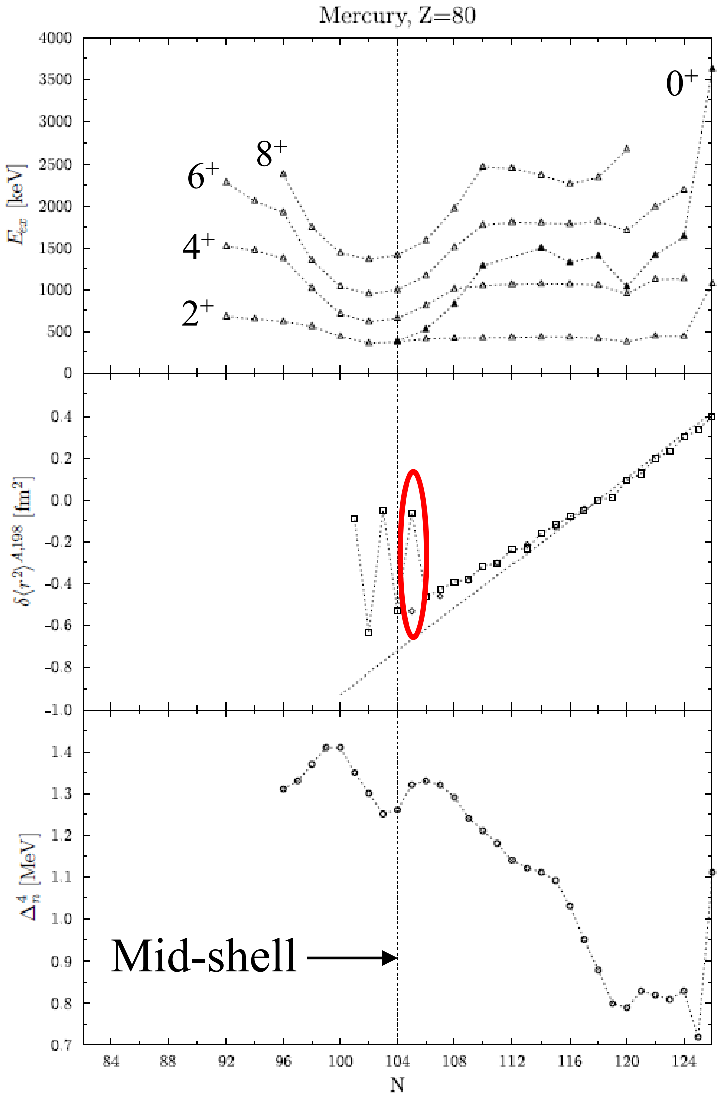
- Inclusion of octupole deformation effects improves consistency of $\delta<r^2>$
- Inversion of odd-even staggering (OES) (very unusual) between *A*=220-226. Connected to octupole degree of freedom suggested – where reflection-asymmetric shapes are predicted!

☺ ^{225}Ra is an attractive case for searches of the atomic EDM due to the enhancement effect from the nuclear octupole deformation.

S.A. Ahmad *et al.*, Nuc. Phys. A483 (1988) 244 (Ra)

W. Borchers *et al.*, Hyp. Int. 34 (1987) 25 (Rn)   W. Kälber *et al.*, Z. Phys. A 334 (1989) 103 (Th)

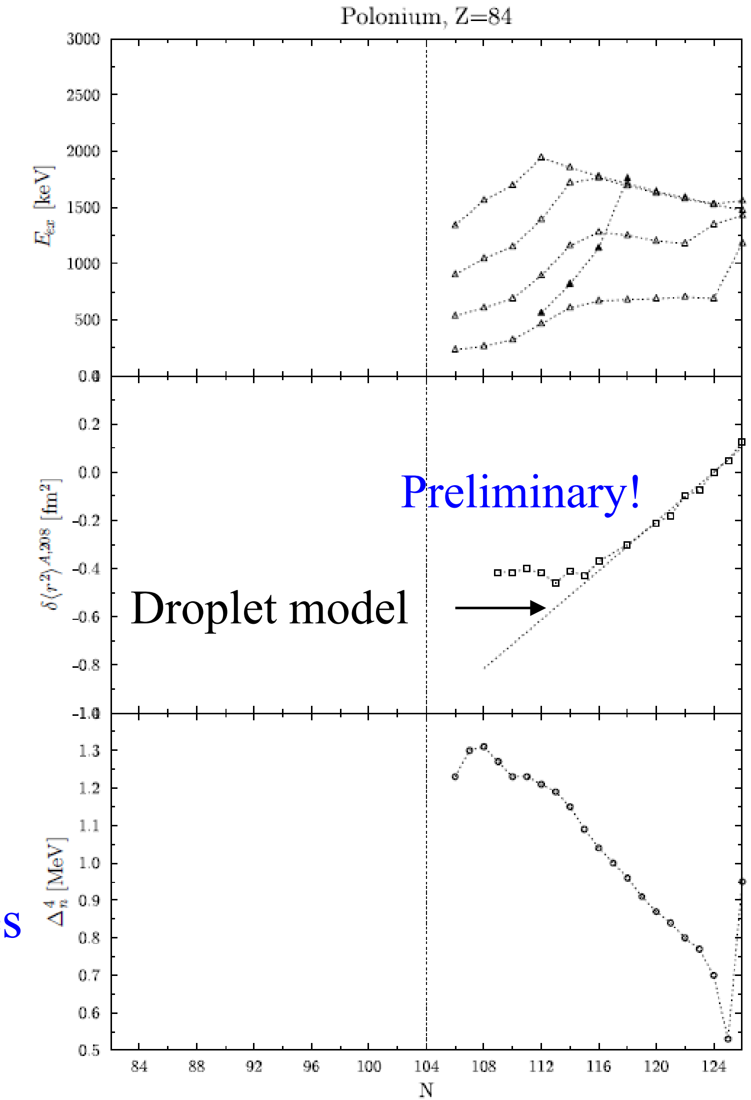
"Complete" spectroscopy is needed



Excited states

Charge radii

Masses/
Binding energies



Figures reproduced with kind permission from P. Rahkila

Outline

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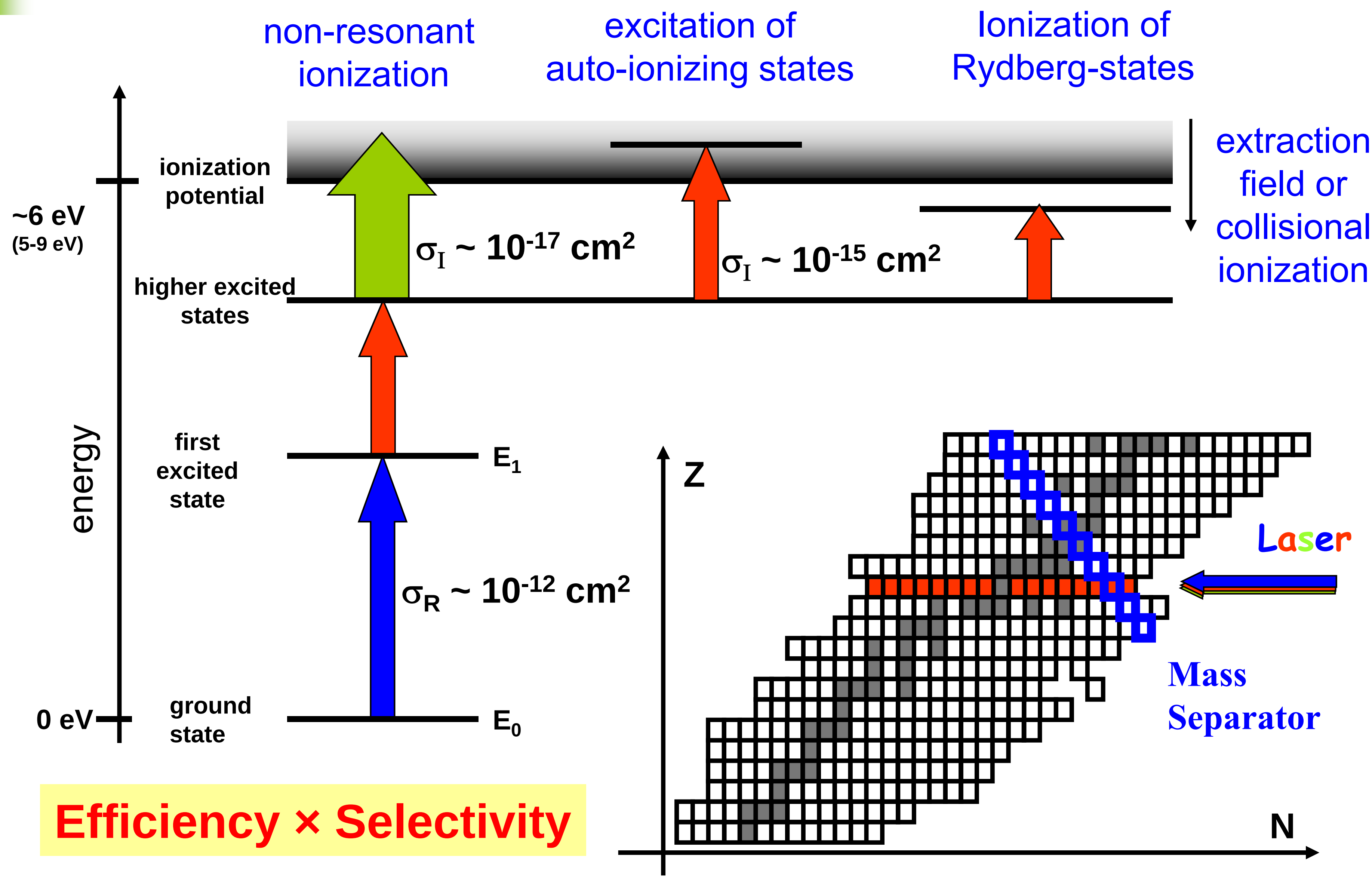
Principle of resonant laser ionization



Efficiency × Selectivity

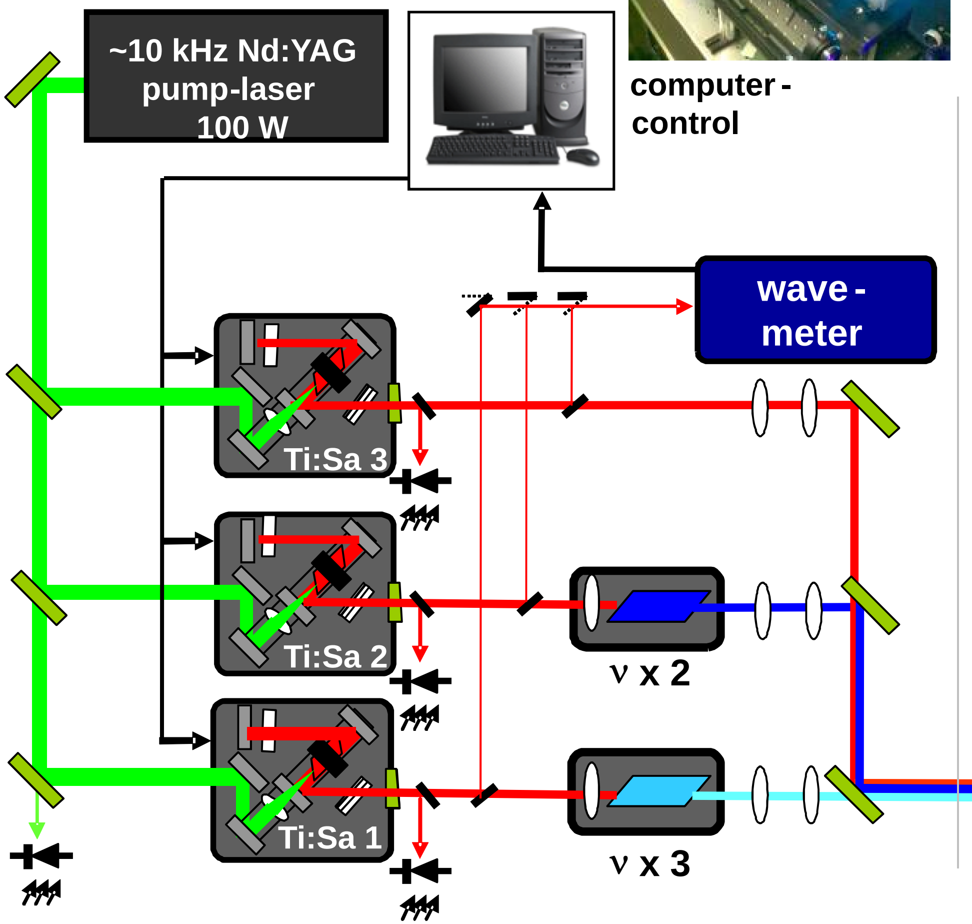
C. Geppert, EMIS conference, 2007.

All solid-state laser system

JOHANNES GUTENBERG UNIVERSITÄT MAINZ

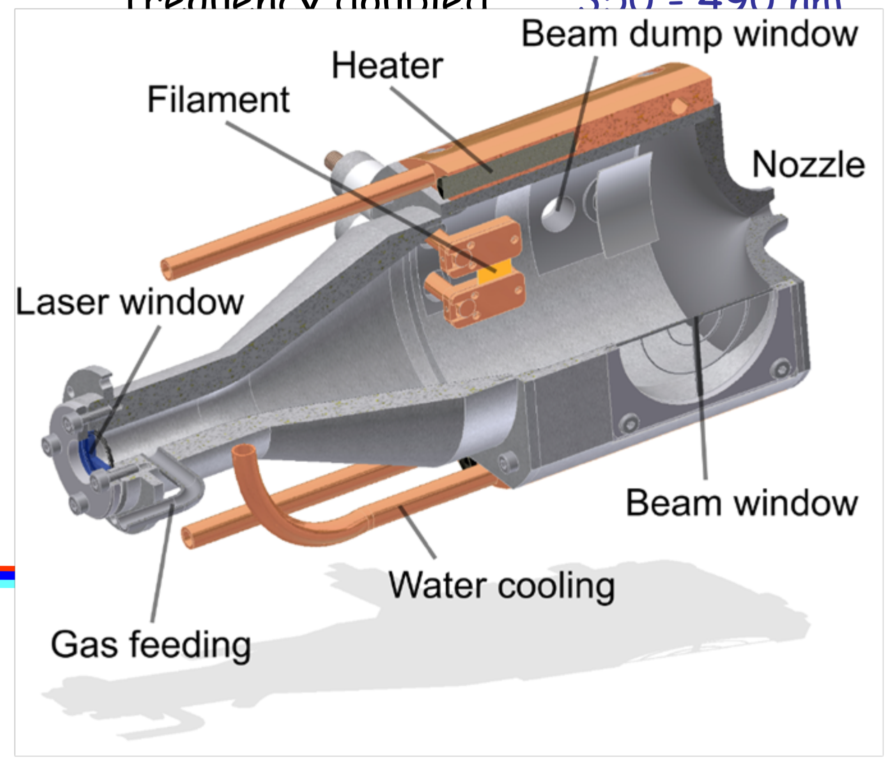
LARISSA

Setup:



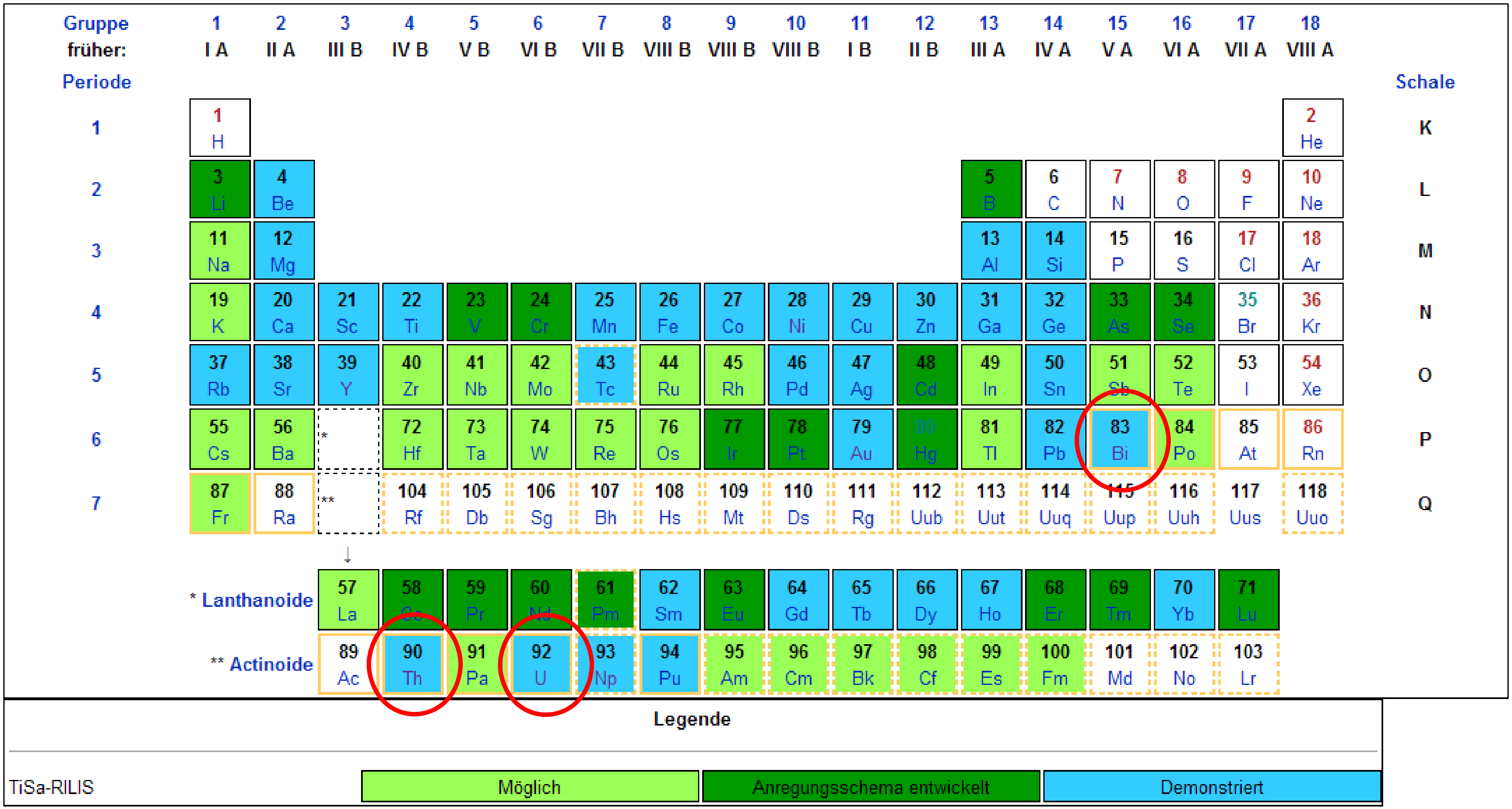
Specifications:

- repetition rate: 5 - 12 kHz
- tuning range:
  - fundamental 700 - 980 nm
  - frequency doubled 350 - 490 nm



Operational at JYFL, Mainz and Triumf, Vancouver

Versatility of solid-state laser system



Possible

Developed scheme

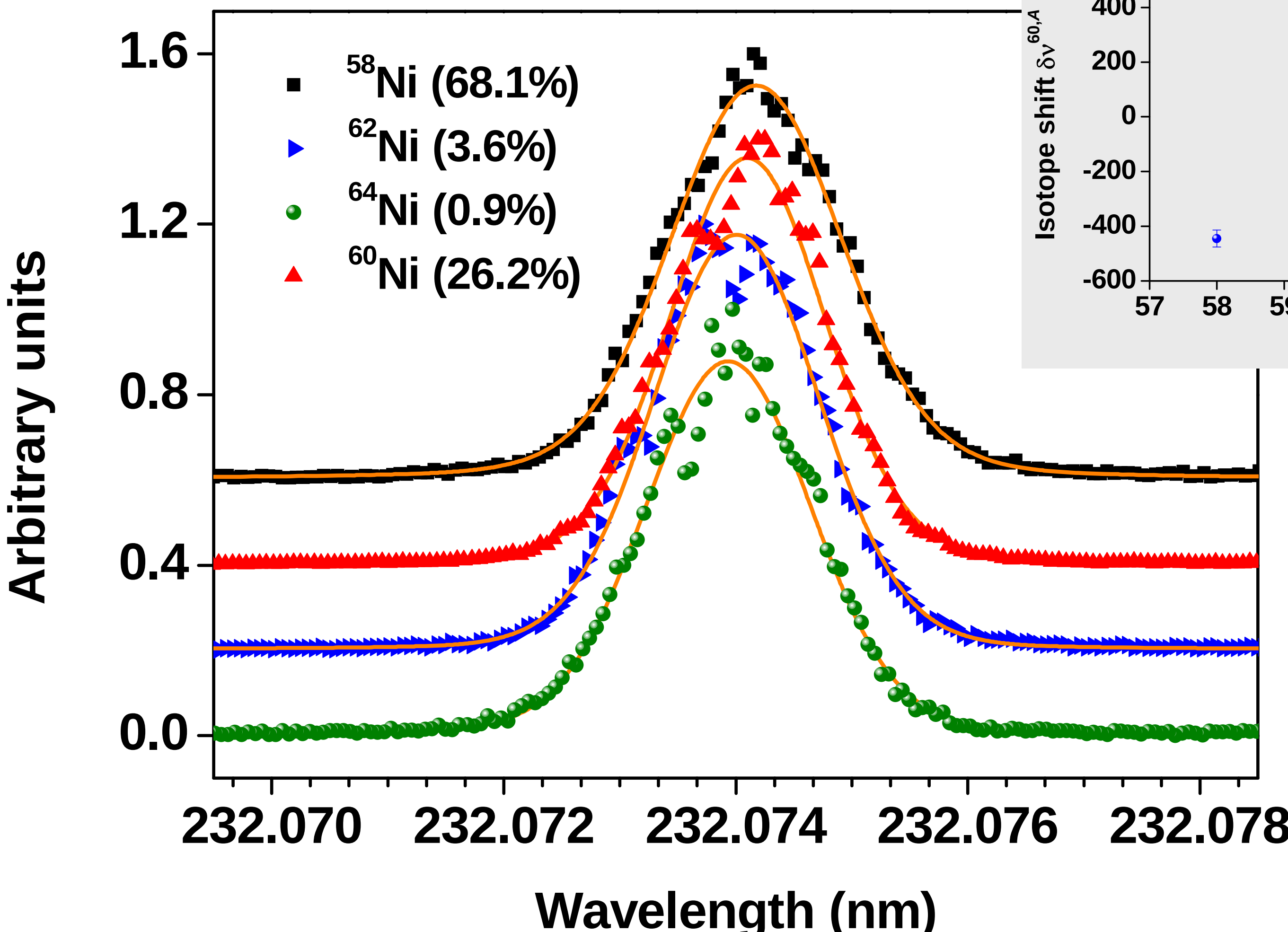
Demonstrated

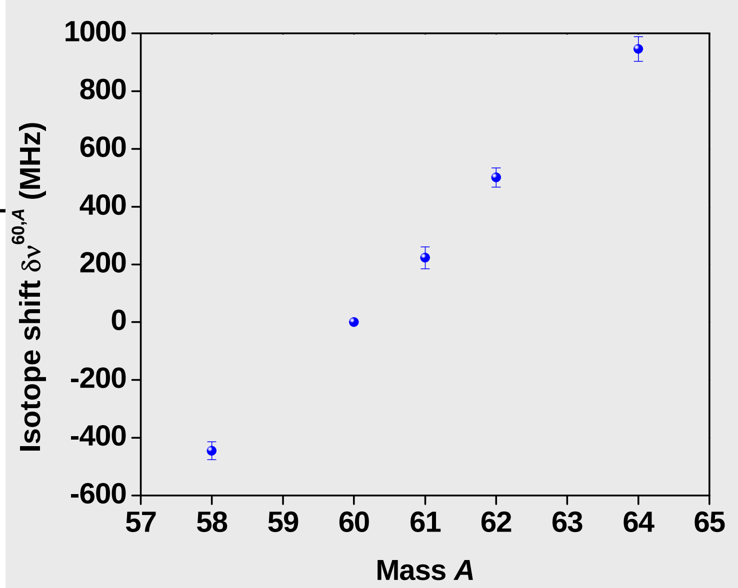
Klaus Wendt

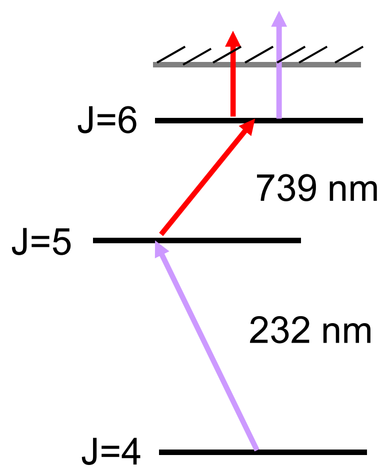
In operation since 2002

First tests on stable Ni using RIS in a gas cell (2009)

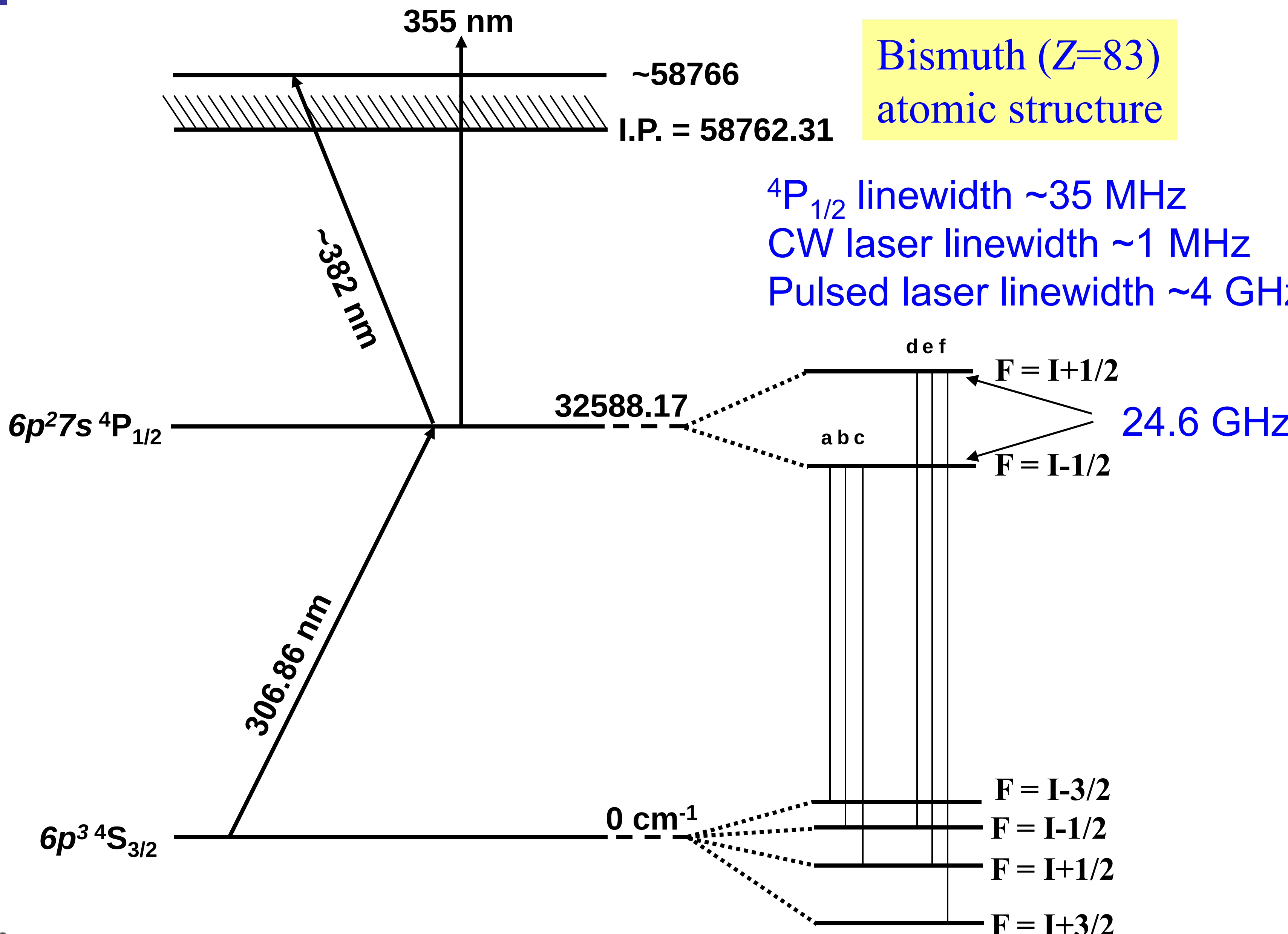
Mass shift dominates over field shift.
Uncertainty in IS < 50 MHz







Bismuth: illustrating the challenges in-source

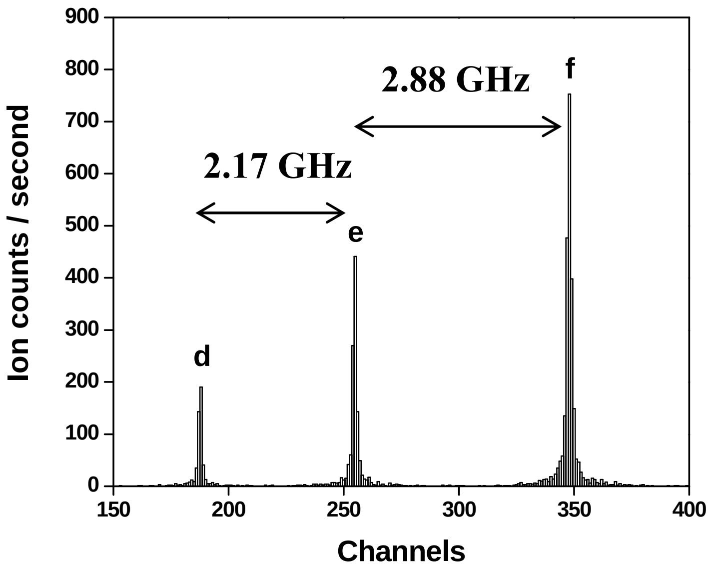


Bismuth (Z=83) atomic structure

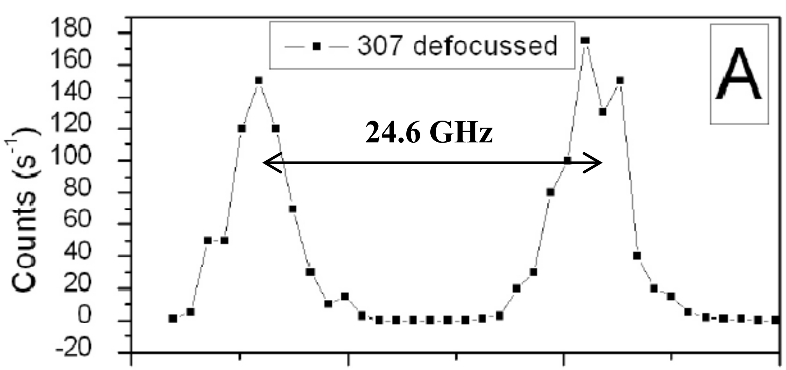
$^4P_{1/2}$ linewidth ~35 MHz
CW laser linewidth ~1 MHz
Pulsed laser linewidth ~4 GHz

Resolution limitations

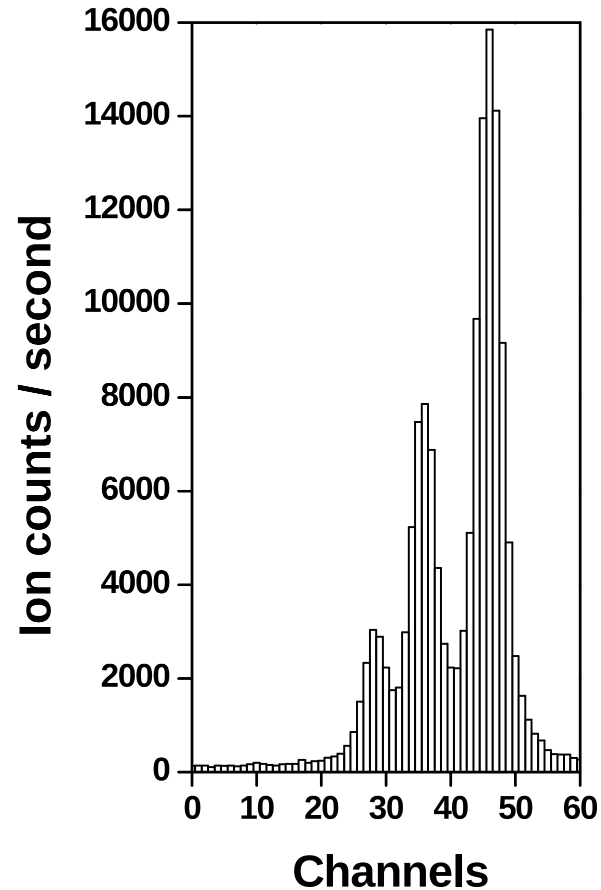
Ideal case: Doppler-free spectroscopy in vacuum (CW first step)



Pulsed dye laser first step (4 GHz linewidth)



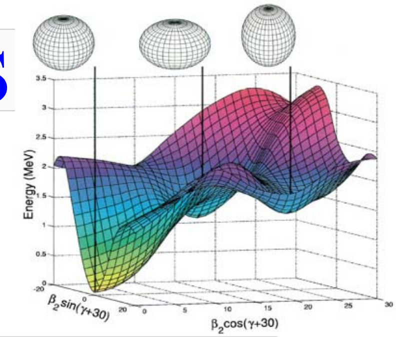
In the ion guide: 100 mbar He gas doppler broadening ~800 MHz, pressure broadening ~500 MHz



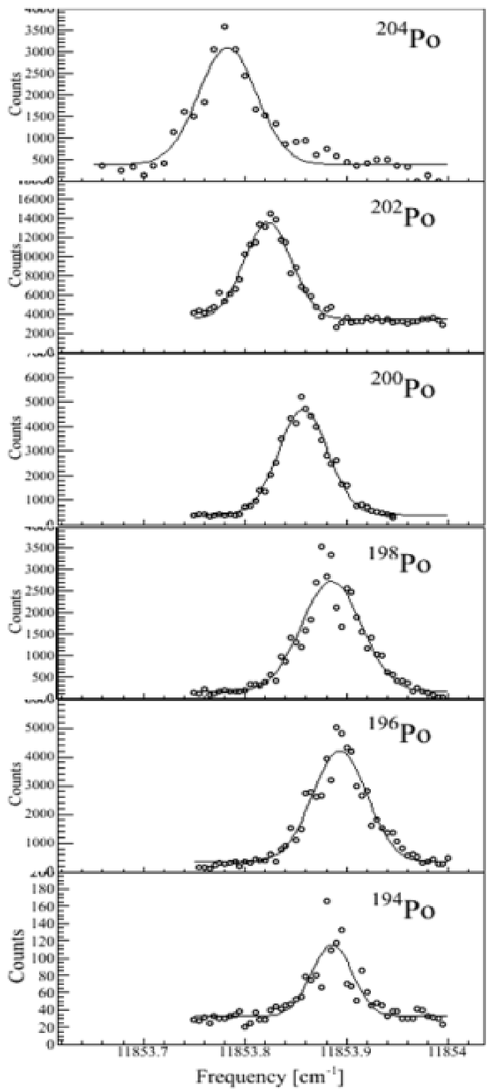
I.D. Moore et al., Hyp. Int. 171 (2007) 135

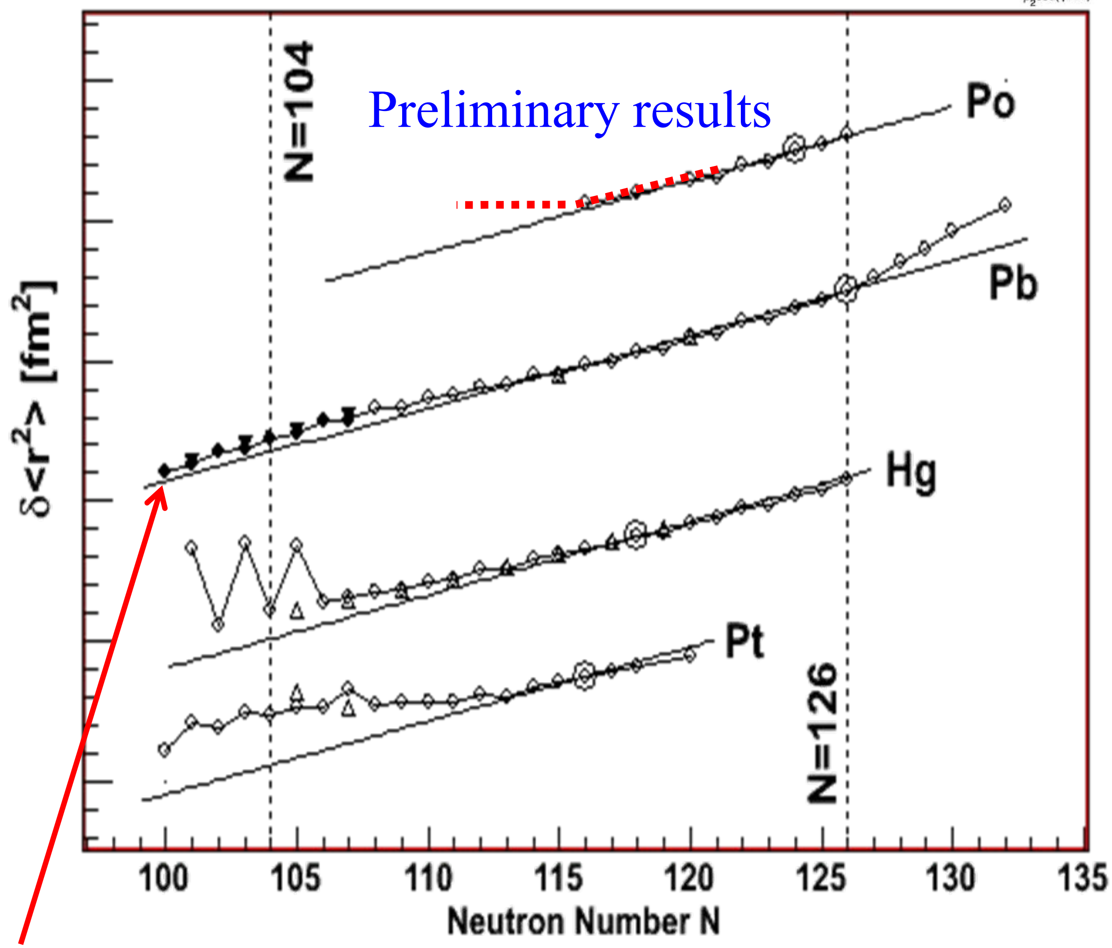
B. Tordoff, PhD Thesis, University of Manchester (2007).

ISOLDE: demonstration of hot cavity RIS



T. Cocolios, under analysis (Po)

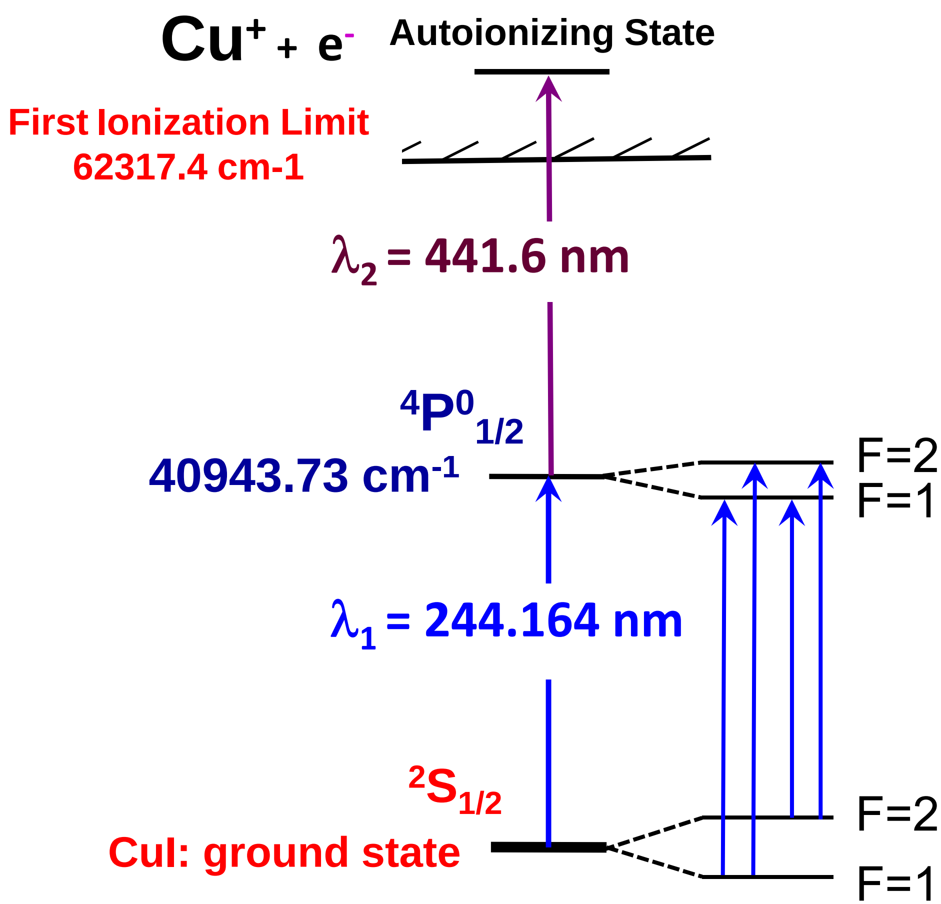


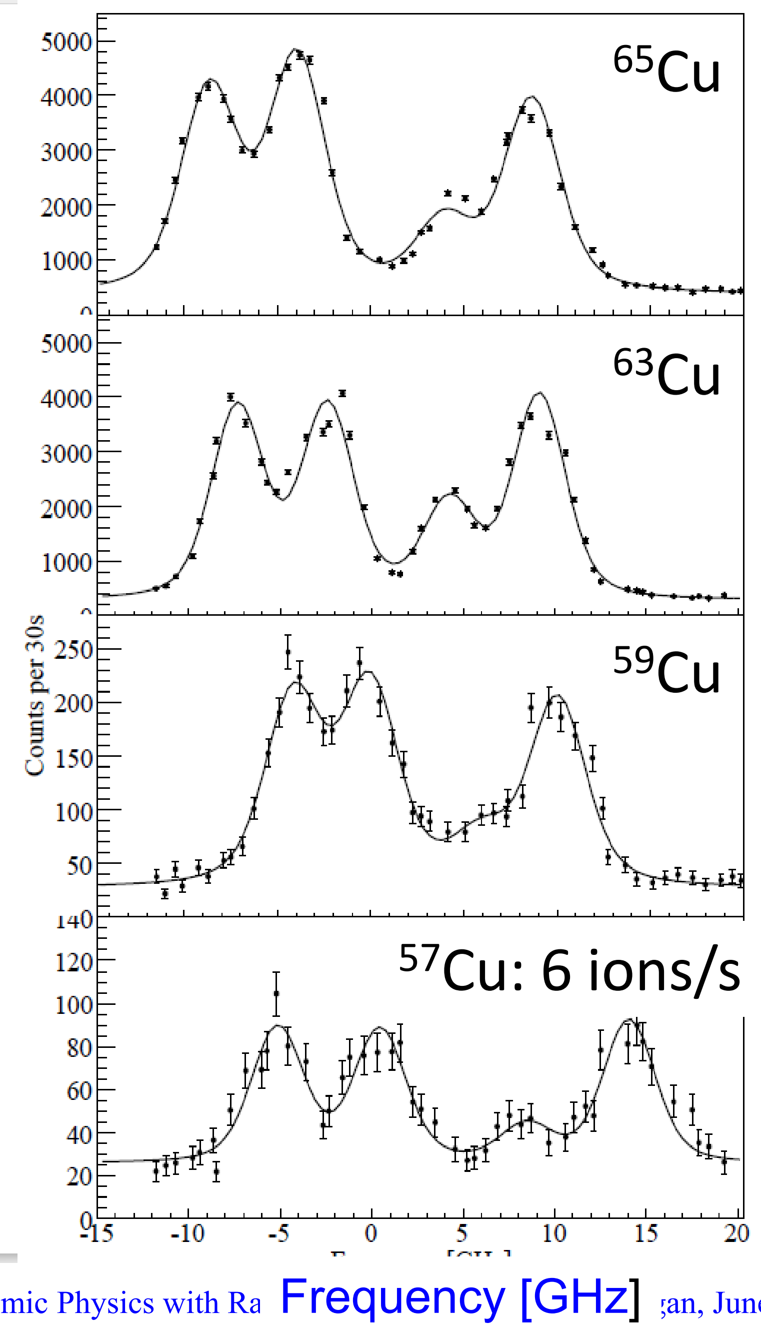


New down to ^{182}Pb ($T_{1/2}$ = 55 ms)

H. De Witte et al., Phys. Rev. Lett. 98 (2007) 112502

In-gas cell spectroscopy of ^{57,59}Cu at LISOL

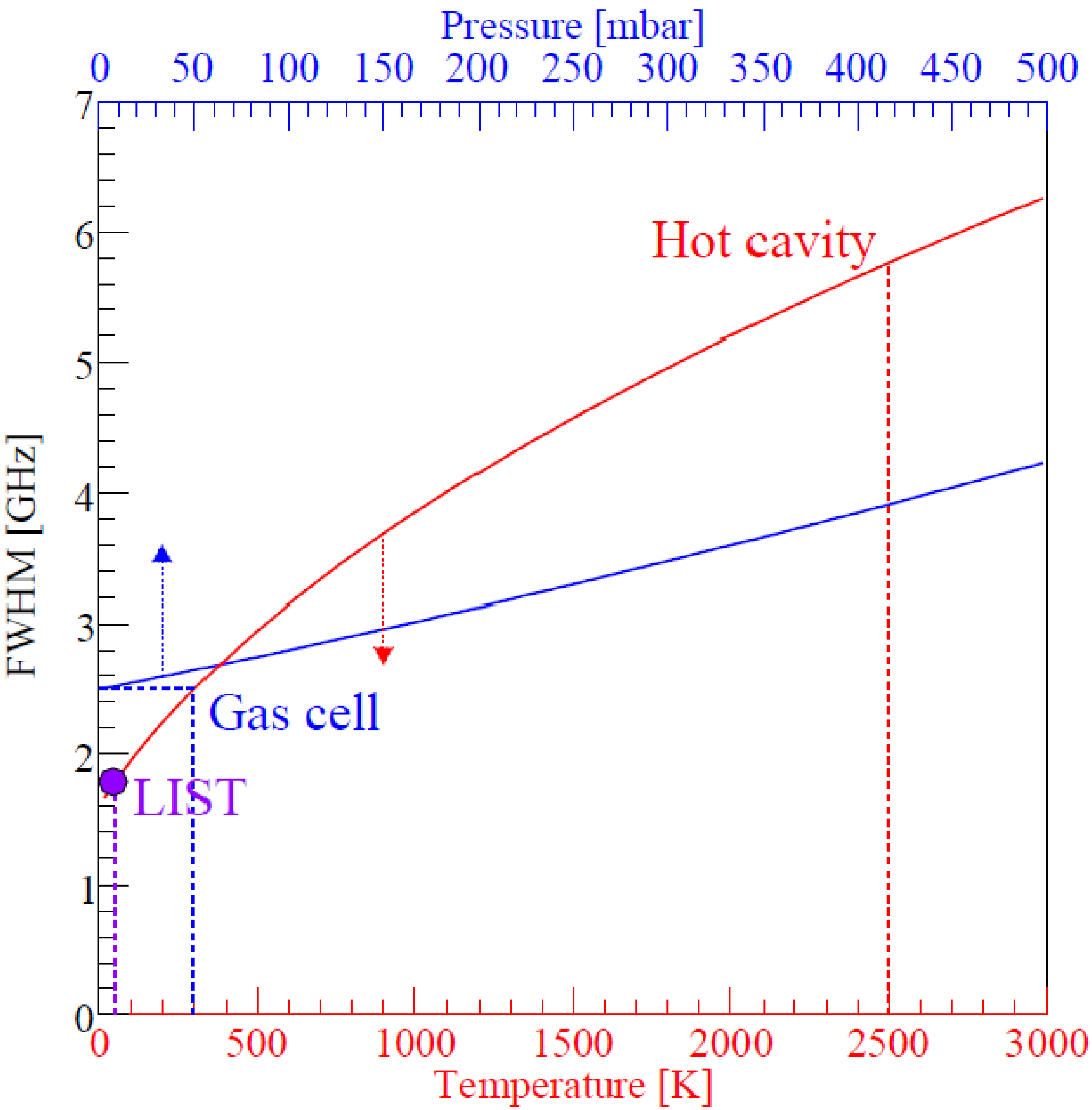




$$\mu({}^{A}Cu) = \frac{A_{hf}({}^{A}Cu)}{A_{hf}({}^{63}Cu)}\,\mu({}^{63}Cu)$$

P. Van Duppen, RNB conference (2009)

Hot cavity vs gas cell: in-source spectroscopy



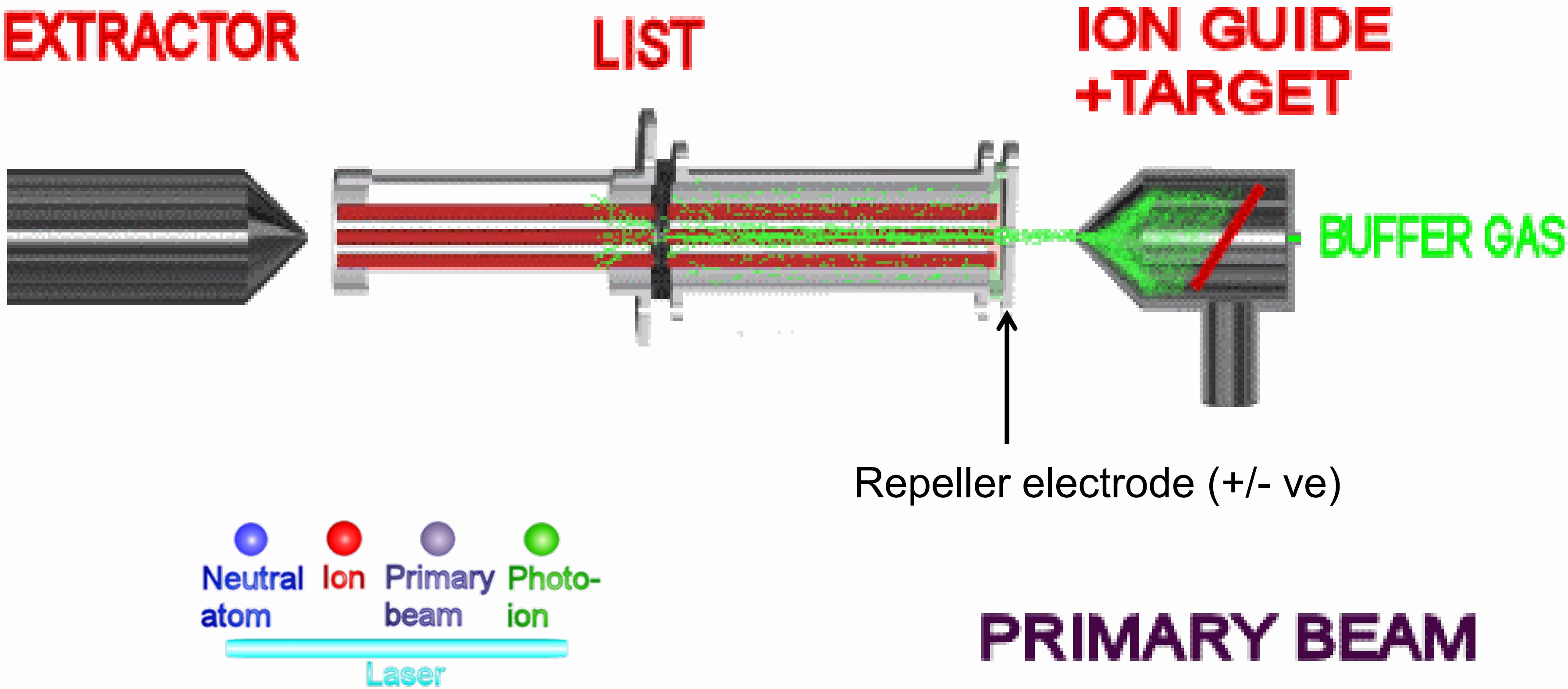
Simulated resonance linewidth of copper transition (244 nm)

- Laser bandwidth 1.6 GHz
- Hot cavity temperature 2500 K
- Gas cell at room temperature
- Pressure broadening important
- LIST mode is promising!

T. Sonoda *et al.*, arXiv:0904.3716 (April 2009)

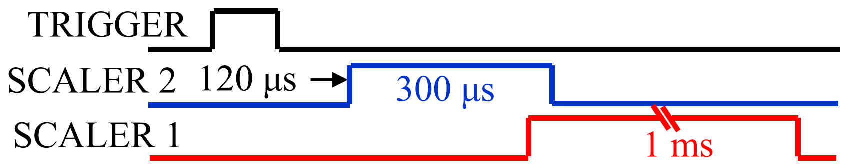
Principle of LIST method (Laser Ion Source Trap)

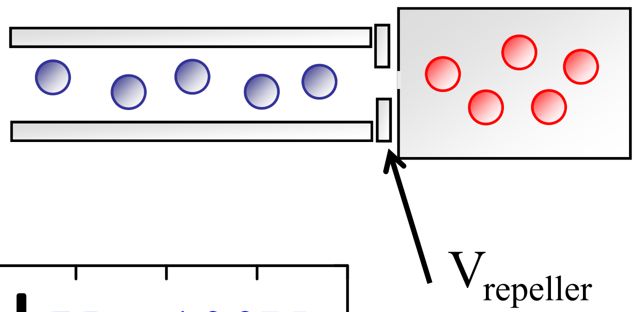
MOTIVATION:
"to achieve the highest selectivity for radioactive ion beam production"

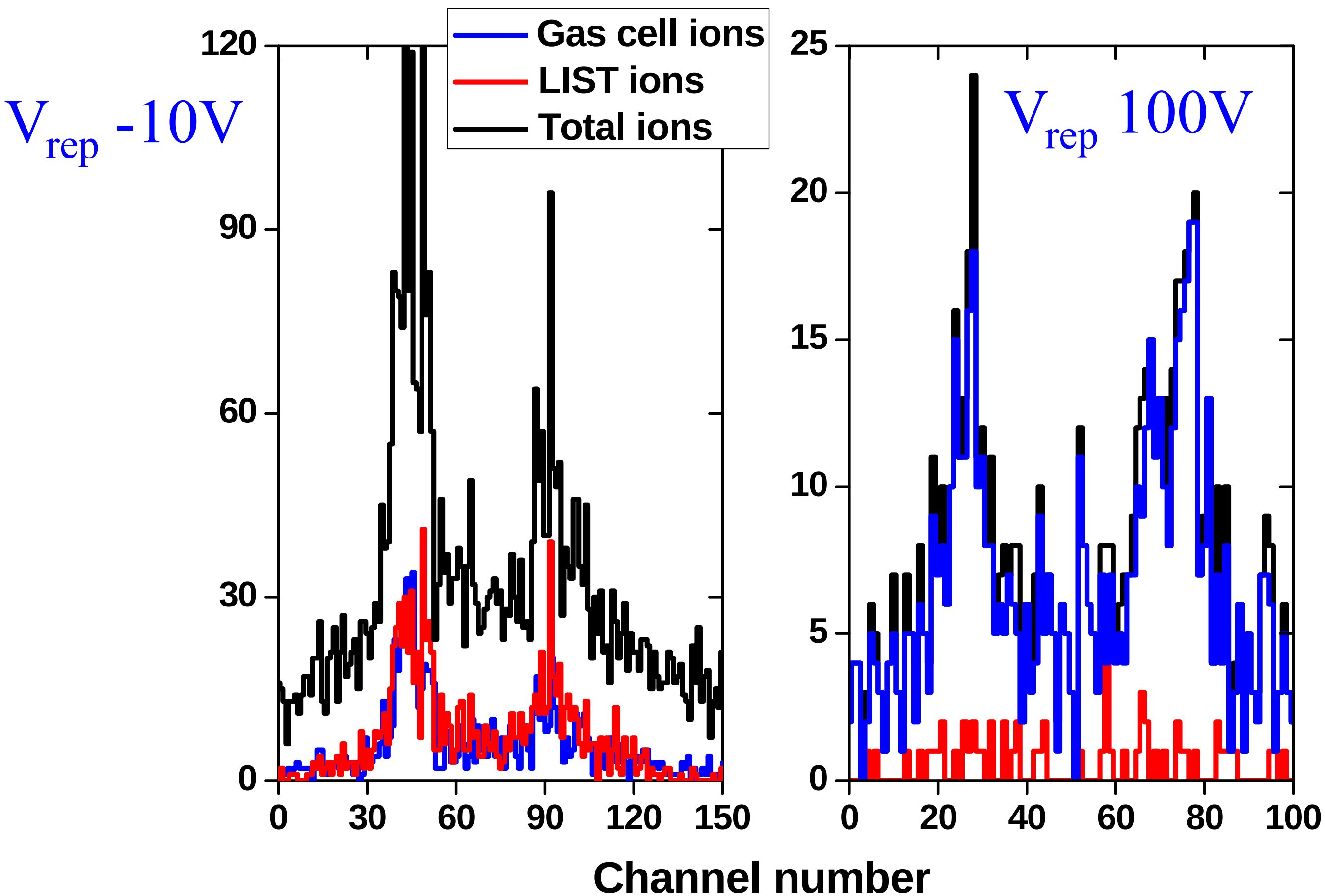


LIST principle also to be applied at ISOLDE and the hot cavity ion source

Demonstration of RIS in LIST mode – ^{209}Bi

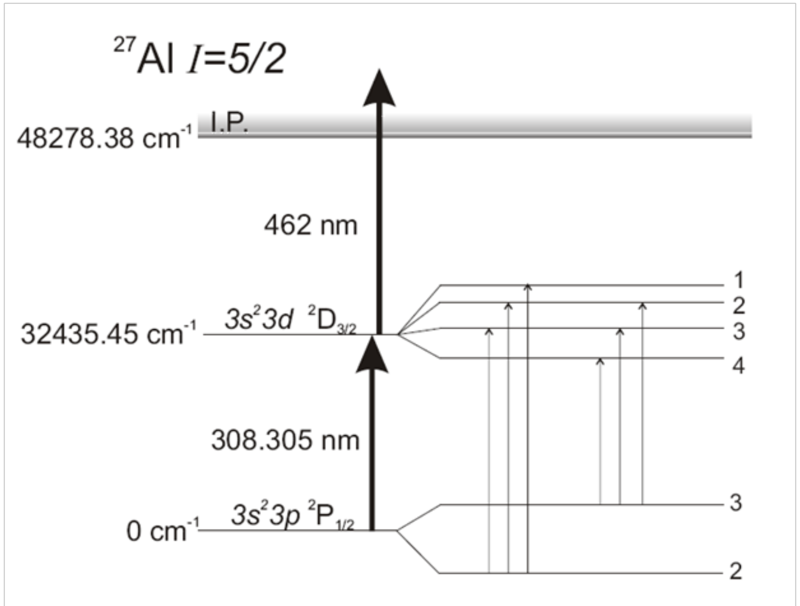




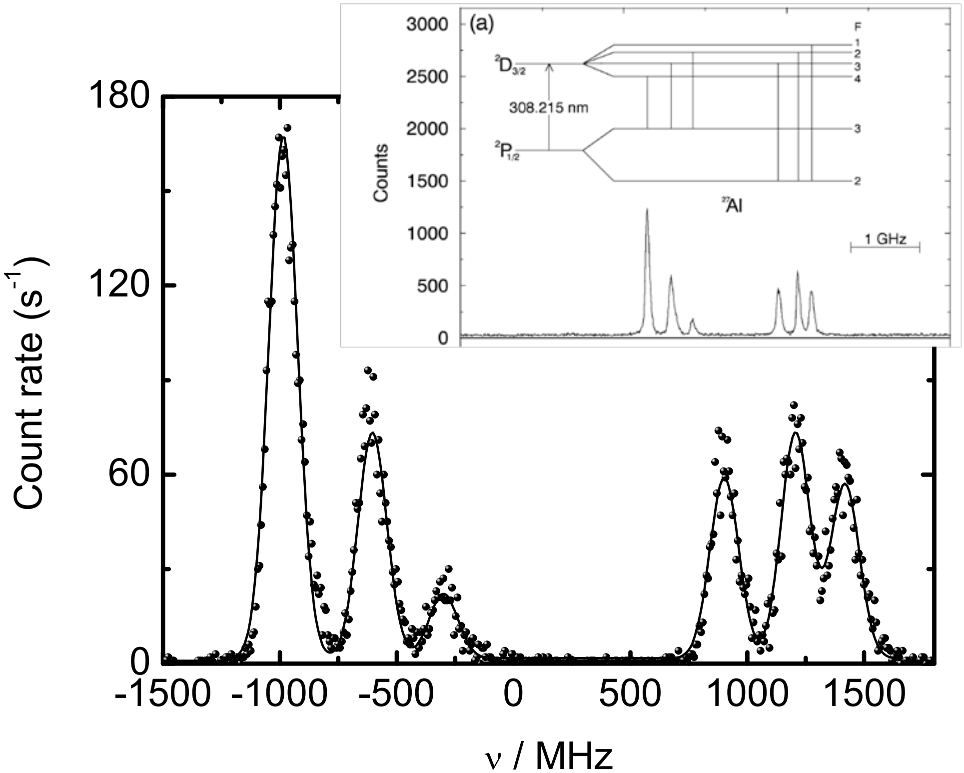


Further development: narrow linewidth pulsed Ti:Sa

Injection seeding of a pulsed Ti:Sapphire laser.
Results in a linewidth reduction from ~4 GHz to ~20 MHz



(a) Hyperfine structure of ^{27}Al

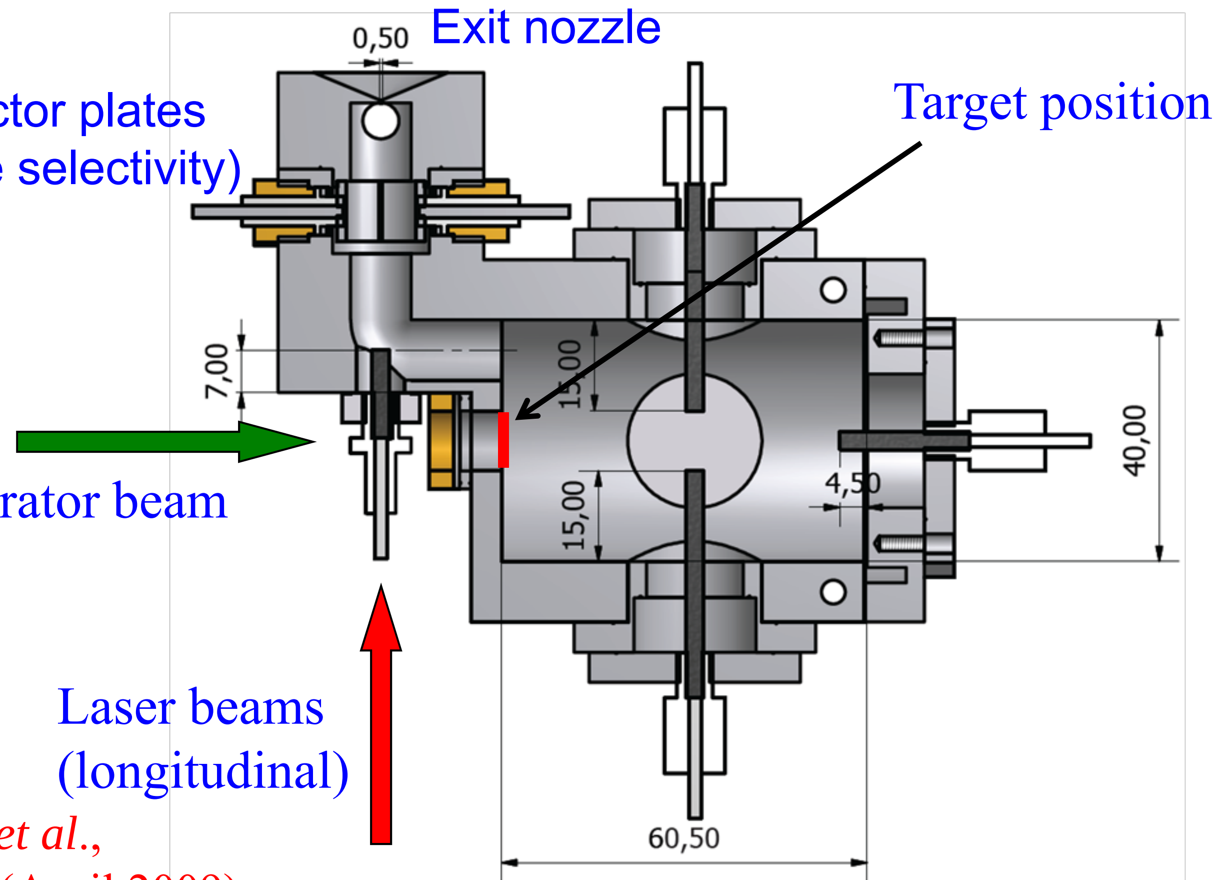


Residual FWHM of 145 MHz of the hyperfine components explained by a combination of Doppler (~100 MHz) and power boadening 33 MHz.

T. Kessler *et al.*, Laser Physics 18 (2008) 1.

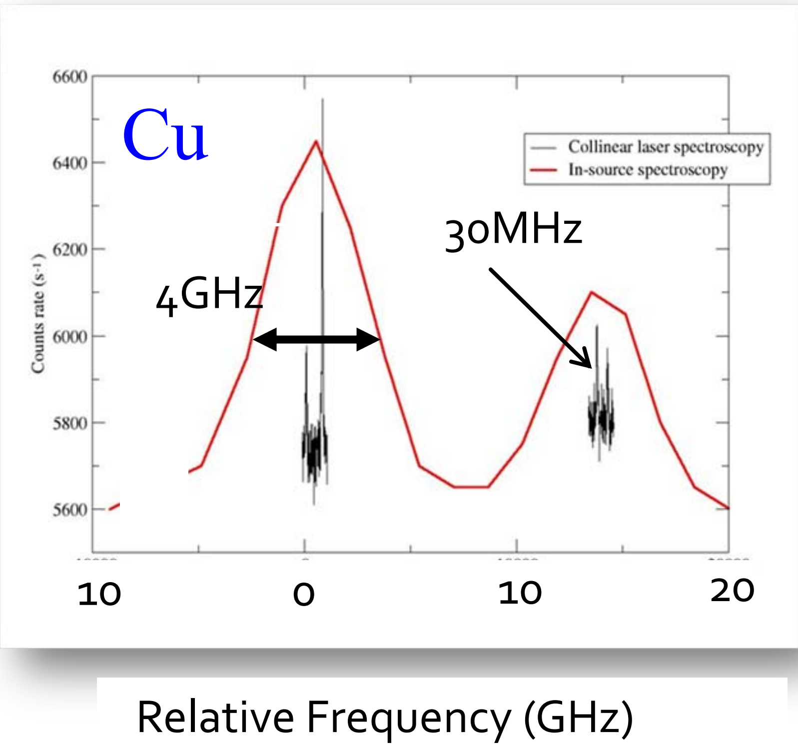
Production and RIS of uranium

- Rare ^{231}Pa ($t_{1/2}$~3×10^4 a) targets exist. High cross sections (100's mb) for (p,xn), (d,xn) reactions
- Utilize new shadow gas cell concept (Leuven development)



Yu. Kudryavstev *et al.*, arXiv:0904.3635 (April 2009)

- Combining the high resolution nature of the collinear beams method with the high sensitivity of the in-source technique.
- Extraction of B factors and hence quadrupole moments – search for a deviation which may indicate octupole deformation.



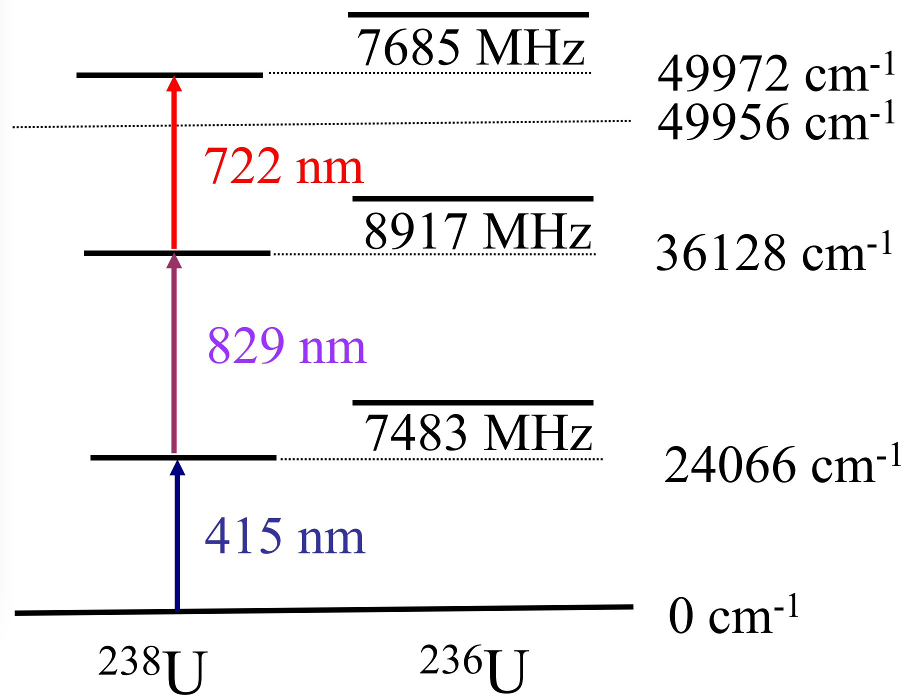
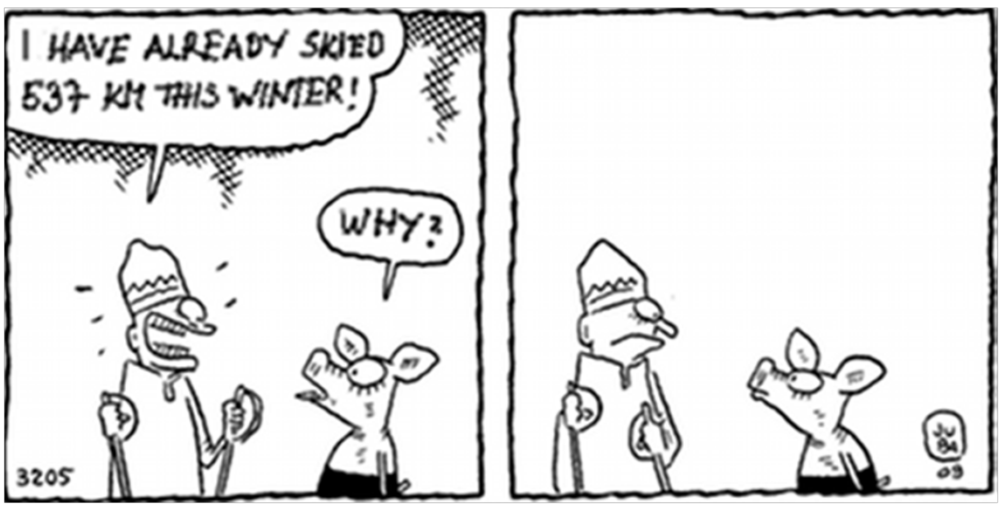


Figure kindly provided by K. Flanagan

Outlook

- We have many open questions to be answered that lie at the borders of atomic and nuclear physics.
- A *complete* approach should be taken to view the problem from different angles.
- Developing new tools and techniques take time but are rewarding.

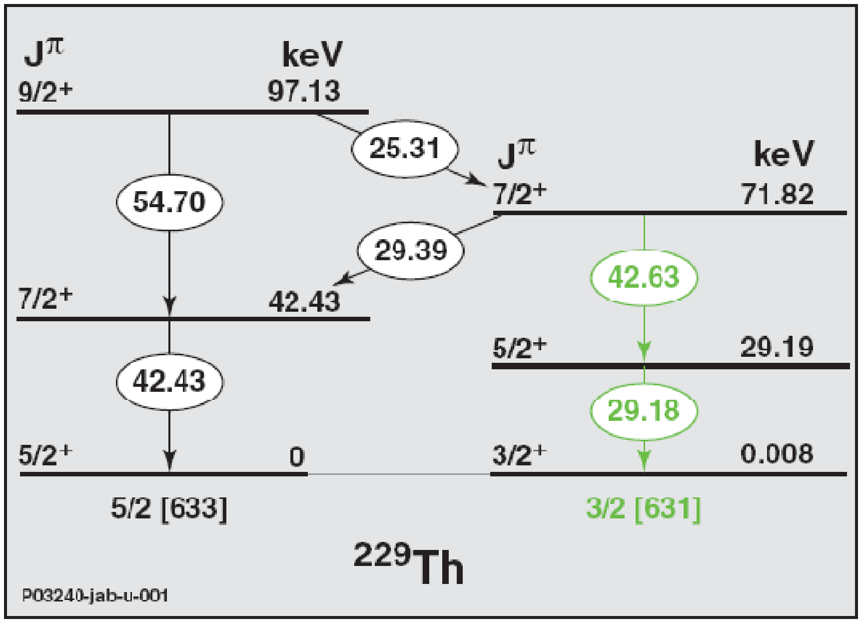


THANK YOU FOR YOUR ATTENTION !

SPARE SLIDES

Detection of the illusive isomeric state in ^{229}Th

Motivation: detection of the lowest lying isomeric state which has yet to be confirmed following 3 decades of experiment and theory.



B.R. Beck *et al*., PRL 98 (2007) 142501

- NASA/EBIT x-ray microcalorimeter spectrometer: E (^{229}Th^{m}) = 7.6±0.5 eV
- Most recent half-life range: 1 min $\leq T_{1/2}^{m} \leq$ 3 min (PRC 79 (2009) 034313)
- Experimental studies include:
  - Closed cycle of γ ray energies
  - Direct reaction work, ^{230}Th(d,t)^{229}Th
  - Detection via optical measurements
  - Radiochemical techniques
  - Feasibility studies of NEET process

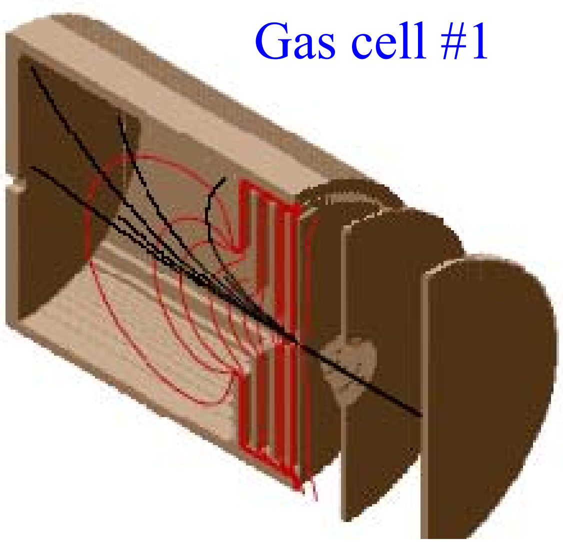
Why is there such an interest?

- A unique system to investigate atomic – nuclear couplings
- An optical clock based on a nuclear transition:
  general relativity tests; the variability of physical constants
  (Flambaum, PRL 97 (2006) 092502)
  (G. Wade *et al*., arXiv:0905.2230, 14th May 2009)
  (E. Litvinova *et al*., arXiv:0901:1240, January 2009)
- A solid-state nuclear frequency standard
  (E. Peik *et al*., arXiv:0802.3548, December 2008)
- Tests of the effect of the chemical environment on nuclear decay rates
- Novel ways to achieve stimulated nuclear excitation

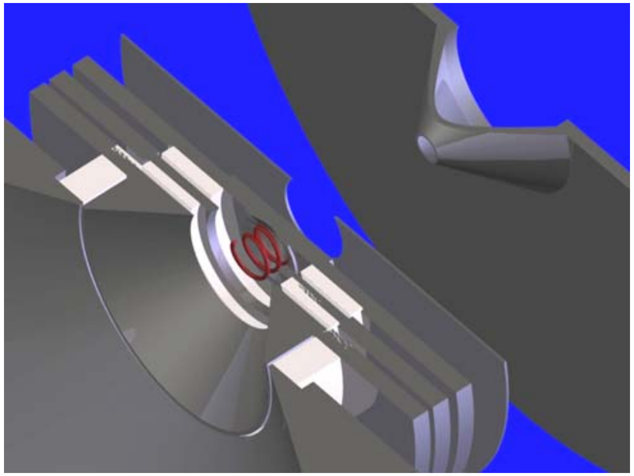
Production and detection of $^{229}Th^m$

- ^{233}U electroplated on stainless steel strips, ~10^5 recoils/s
- Stopped in 50 mbar He gas, guided to exit hole
- Collinear laser spectroscopy or in-source RIS for HFS

Gas cell #1



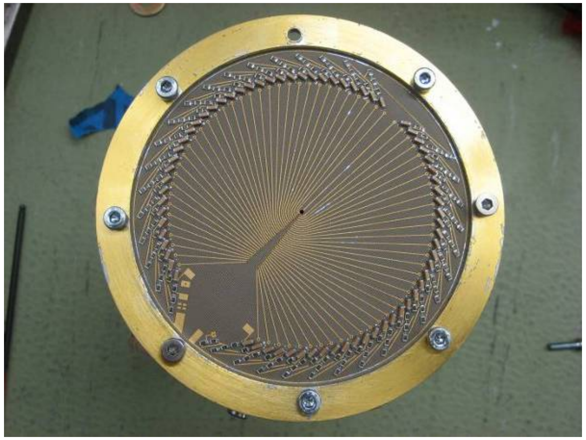
Electron emitter in gas cell #2



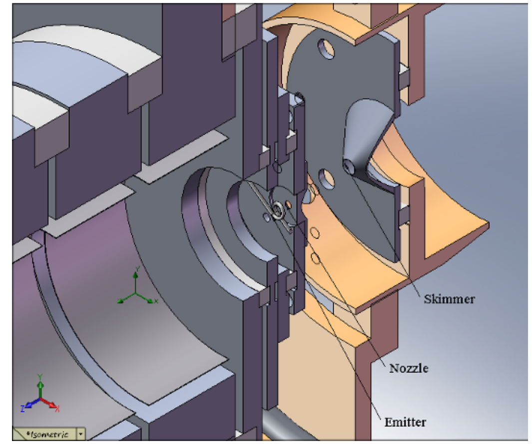
Ion guide efficiency of $^{221}Fr^+$ ($\tau_{1/2}$=4.9 min) and $^{217}At^+$ (32.3 ms) was 6%, $^{229}Th^+$ 0.06%. Missing efficiency in molecular formation, doubly-charged ions and unknown neutral fraction.

B. Tordoff *et al.*, NIMB 252 (2006) 347

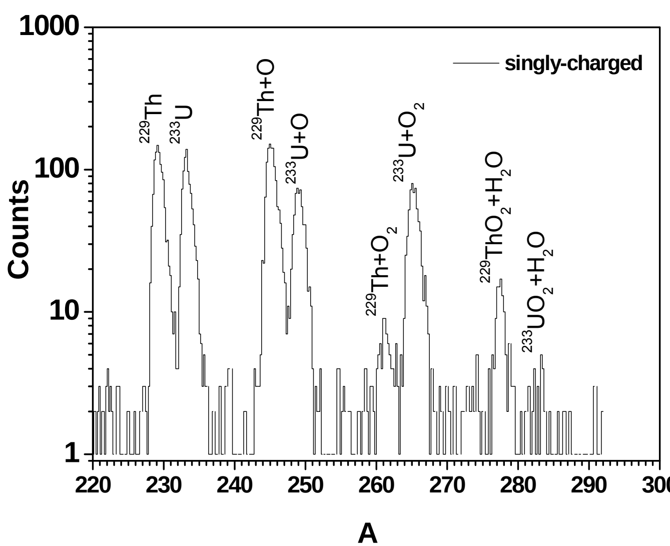
RF carpet + gas cell #3

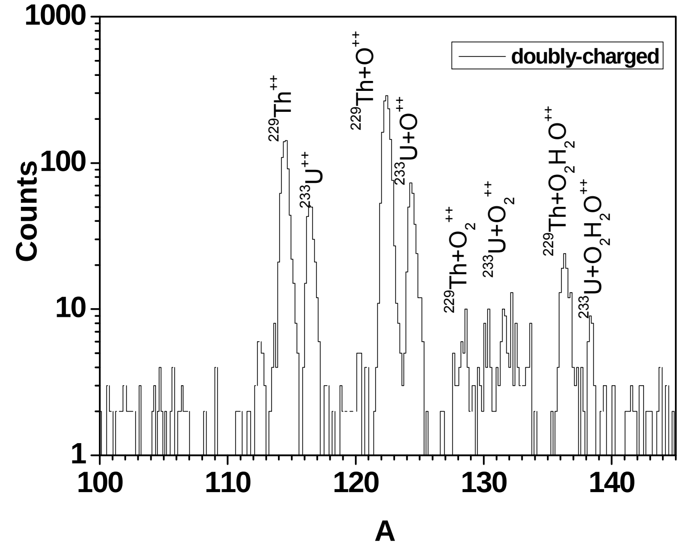


DC and electron emitter in gas cell #4



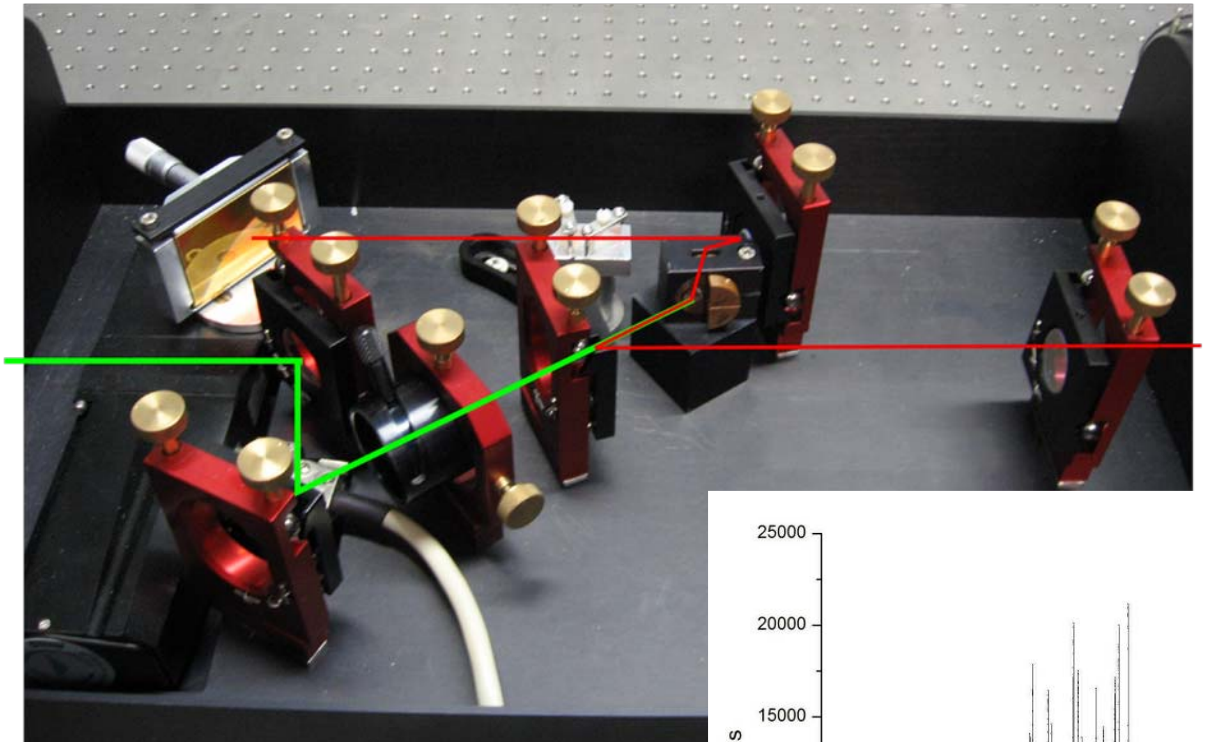
Gas cell #3 cryogenically cooled. Overall efficiency ($^{221}Fr^{+}$) 0.6% but $^{229}Th^{+}$ extraction efficiency 0.36%. Gas cell #4, $^{221}Fr^{+}$ ~16% , $^{229}Th^{+}$ 1.6% (JYFLTRAP).

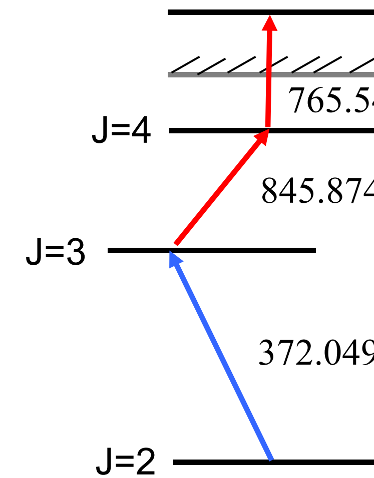


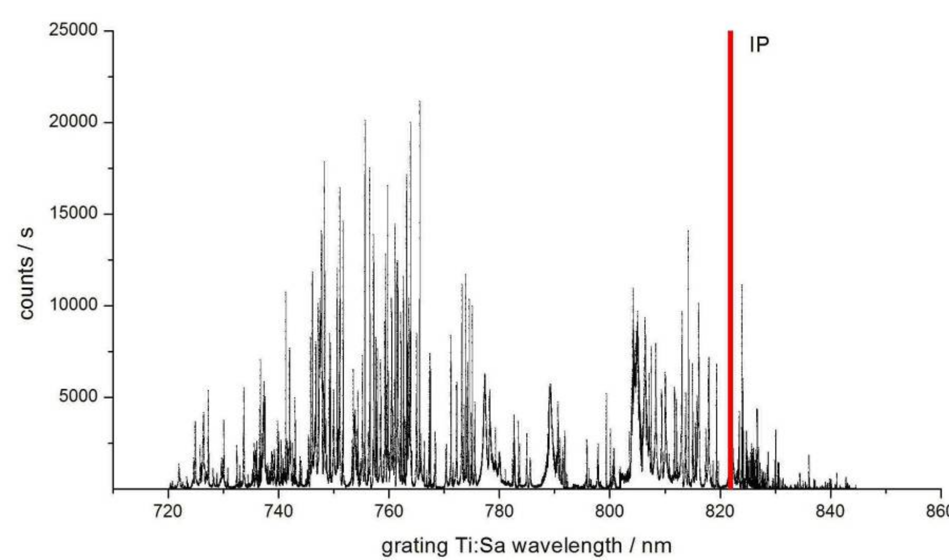


RIS scheme development in Mainz (2009)

Development of a wide-tunable Ti:Sapphire laser

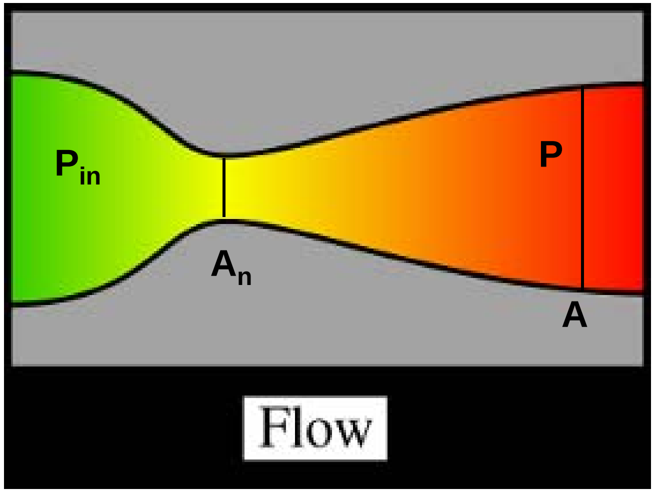




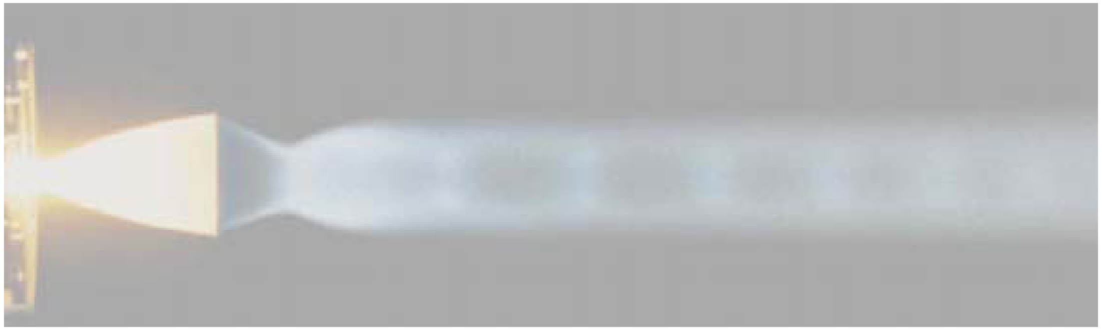


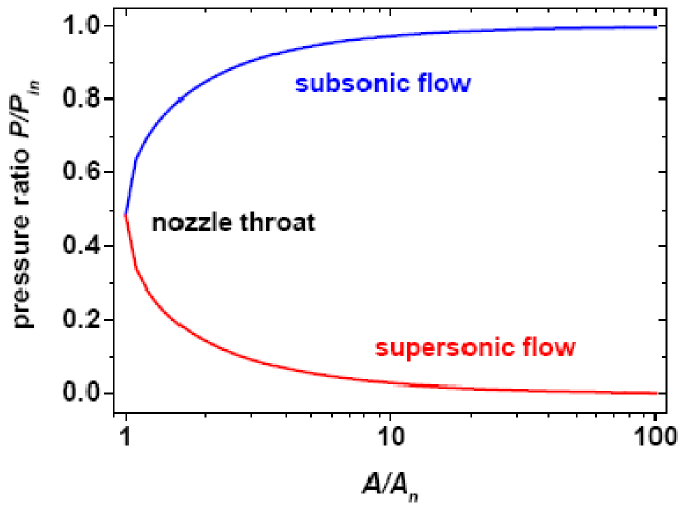
300 autoionizing states found in thorium

Improve LIST efficiency - shape the gas jet

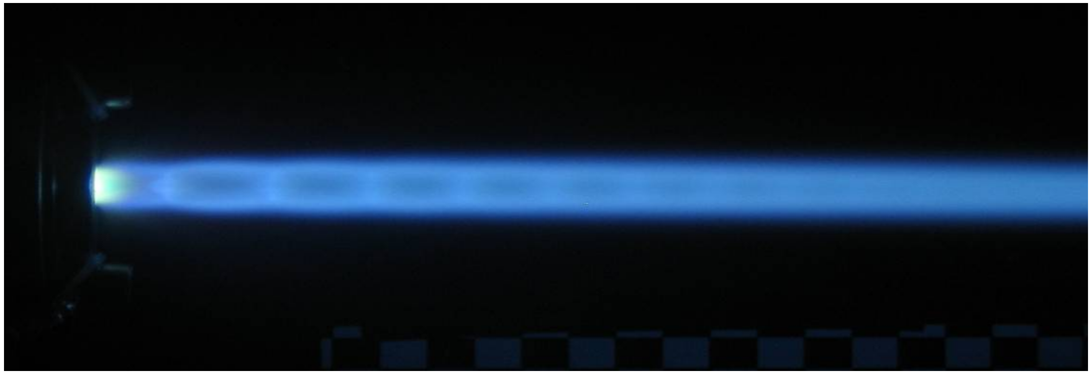


A rocket scientist approach – the de Laval nozzle

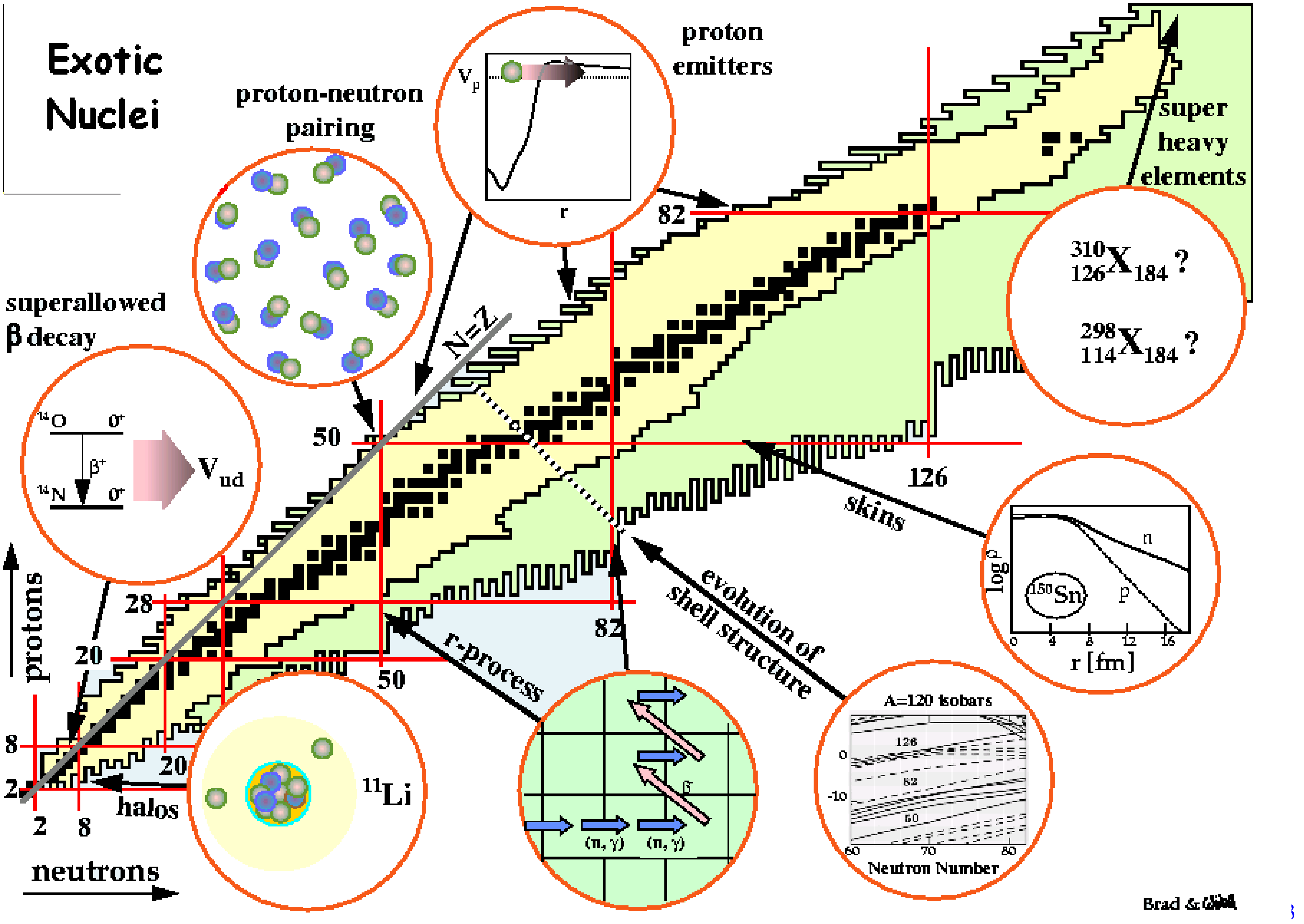




A supersonic jet ”engine” at IGISOL

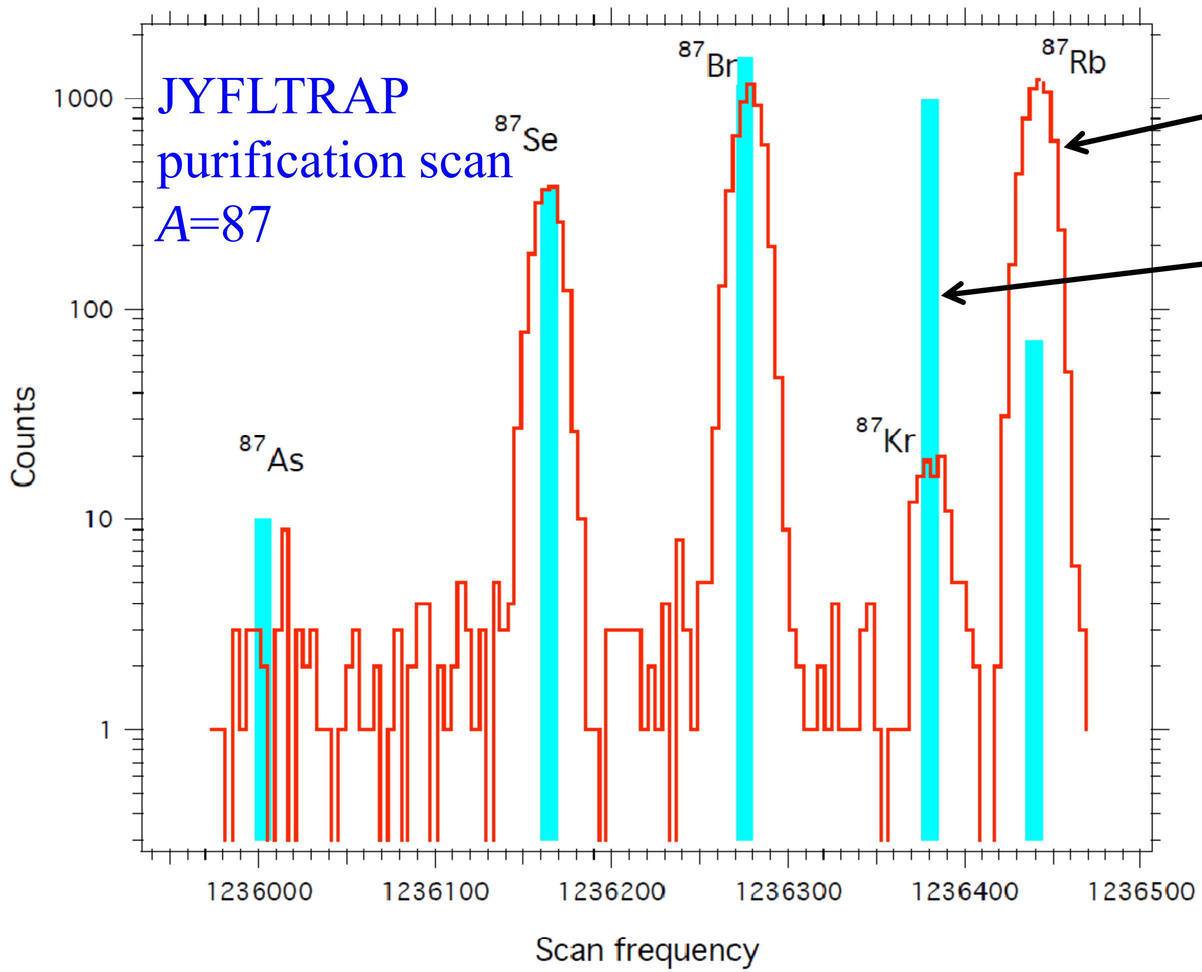


The nuclear landscape and modern topics



The argument for chemical independence

”All species have a much lower ionization potential than He, therefore they remain ionized during extraction from the gas cell”.



Fission yield measurements

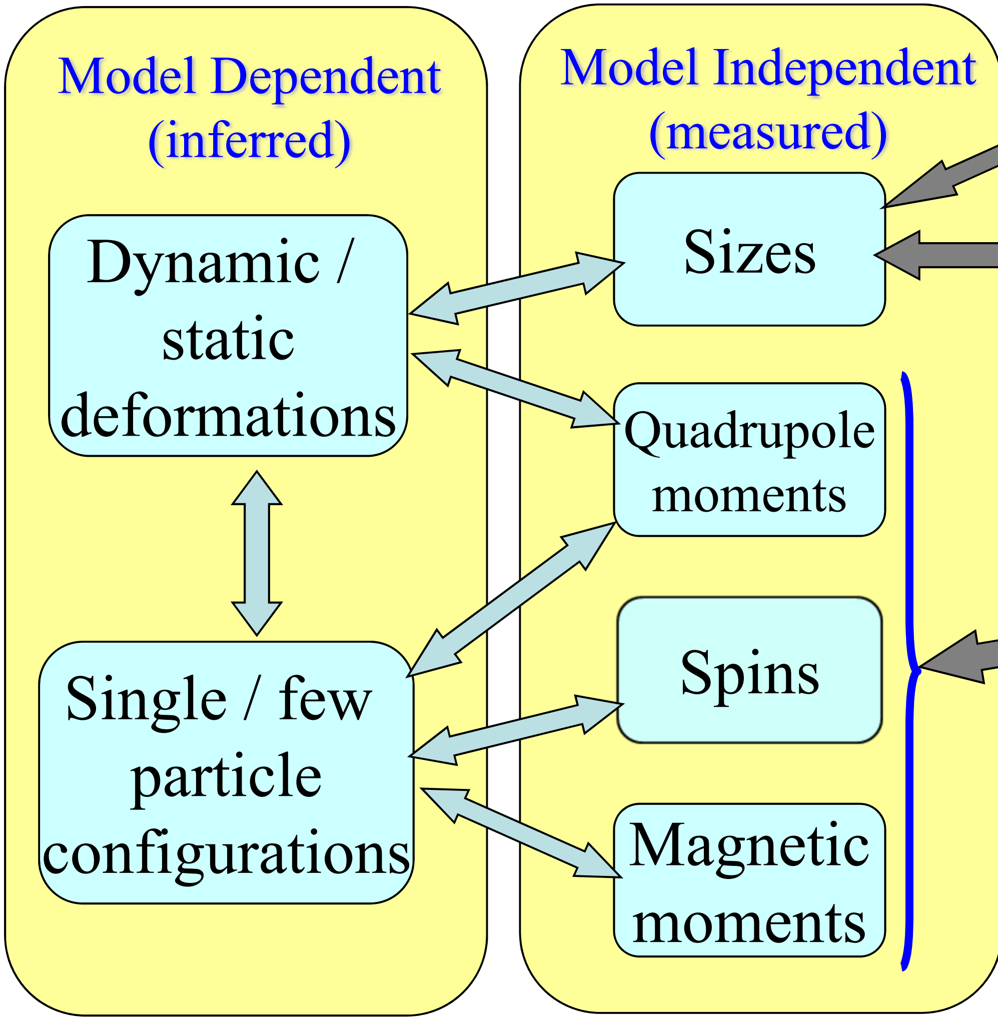
Fission yield predictions

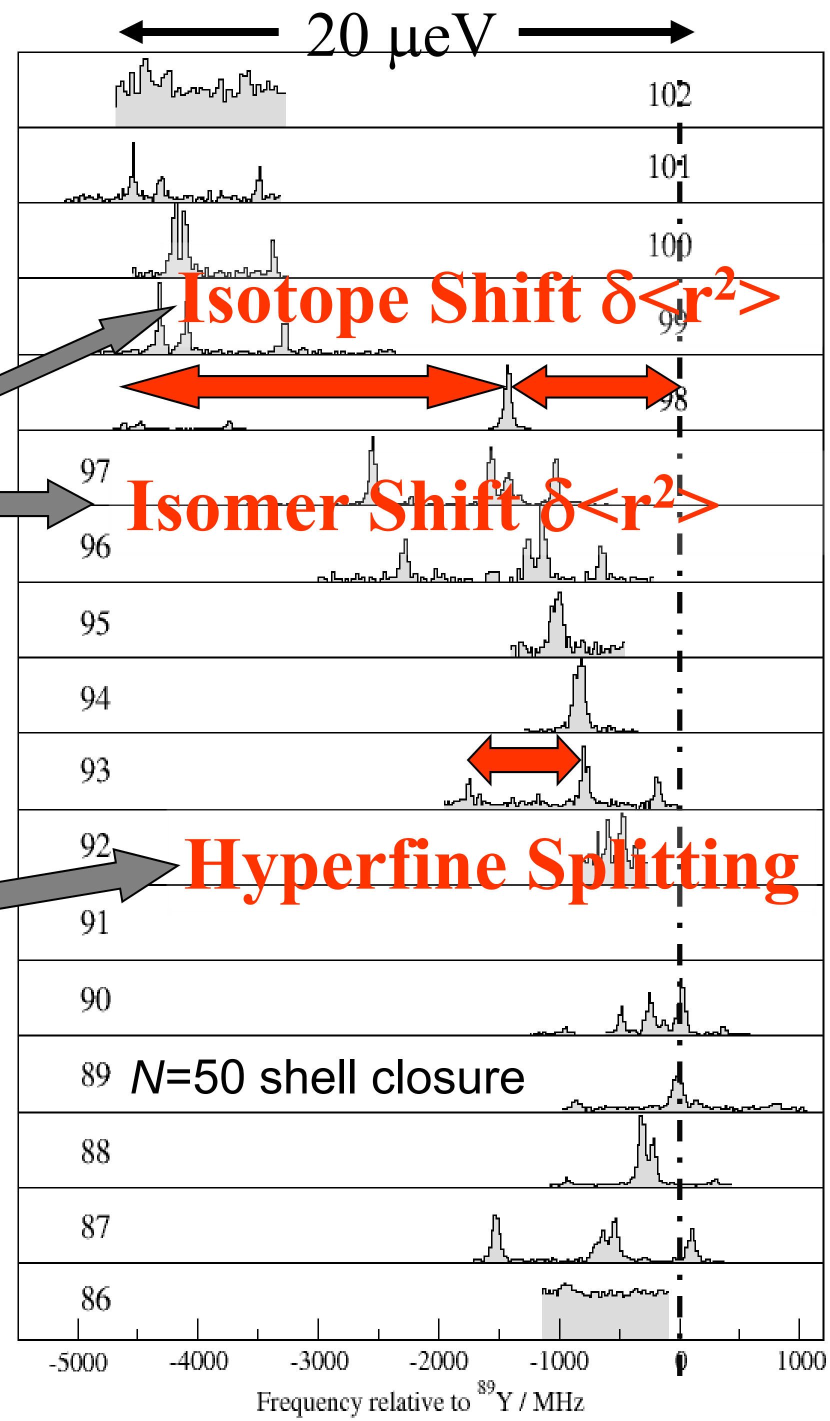
^{87}Rb stable isotope: overproduced (fission+ elsewhere).

^{87}Kr radioactive isotope underproduced (lost via charge exchange)

Laser spectroscopy

Measurement of optical spectra





Properties obtained and inferred

Isotope Shift (IS) → Mean Square Charge Radii

$$\delta\langle r^2\rangle^{AA'}$$

Hyperfine Structure (HFS) →

Nuclear Spin I

Magnetic Dipole Moment μ_I

Electric Quadrupole Moment Q_s

Size (droplet model) — Shape (quadrupole term) — Diffuseness (assumed constant)

$$<r^2>=<r^2>_{sph}(1+\frac{5}{4\pi}(<\beta_2^2>+\ldots)+3\sigma^2)$$

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

$$<\beta_2^2>\neq<\beta_2>^2$$

$$\beta_{rms}^2=<\beta_2>^2+(<\beta_2^2>-<\beta_2>^2)=\beta_{static}^2+\beta_{dynamic}^2$$

From atomic to nuclear physics

Isotope Shift

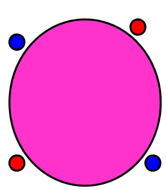
Atomic factors to calibrate

$$\delta\nu^{A,A'} = M_i \frac{A - A'}{AA'} + F_i \delta < r^2 >^{A,A'}$$

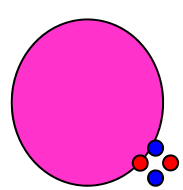
MASS SHIFT

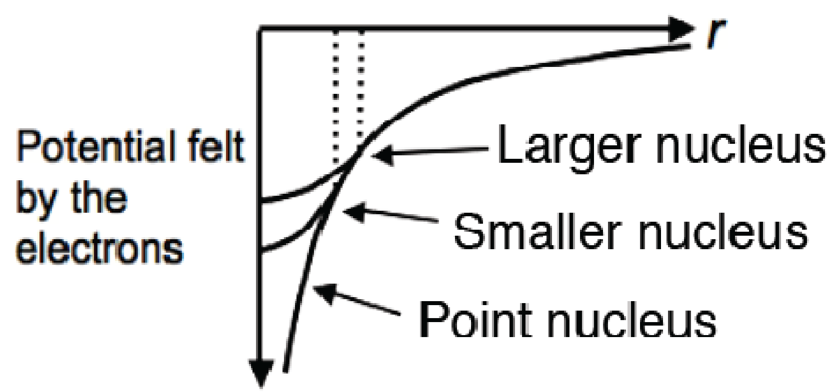
FIELD SHIFT

Mass shift due to change in the nucleus recoil kinetic energy (partly related to change in electron reduced mass)

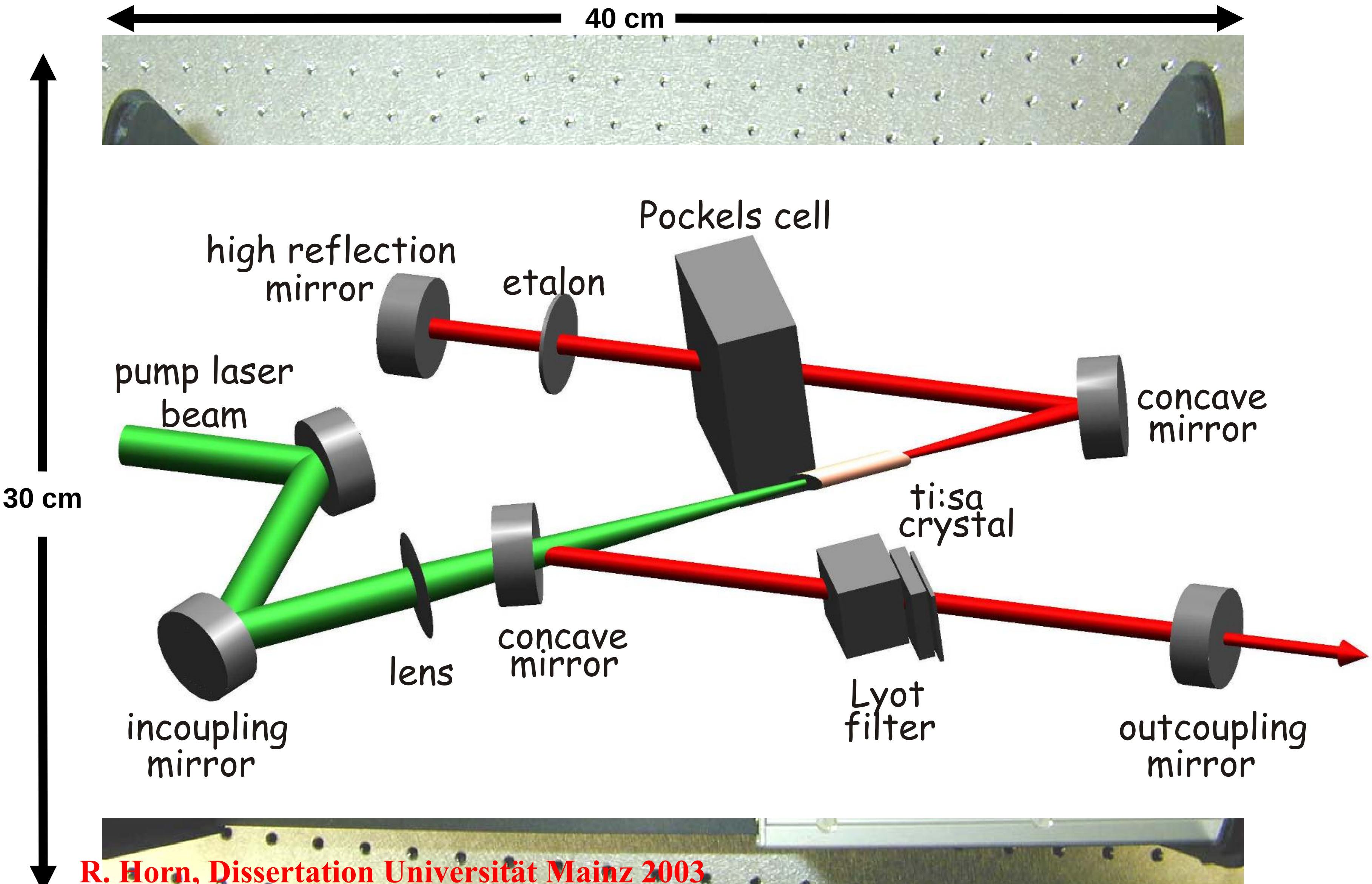


or



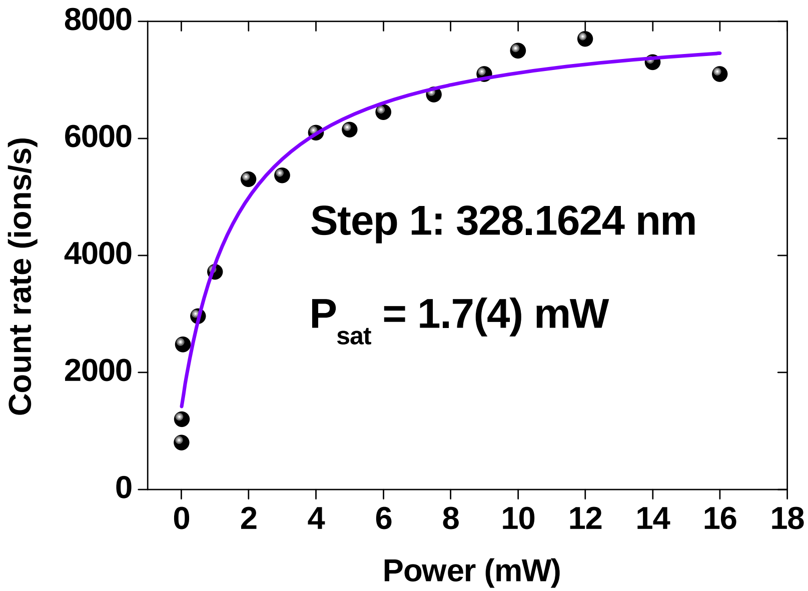


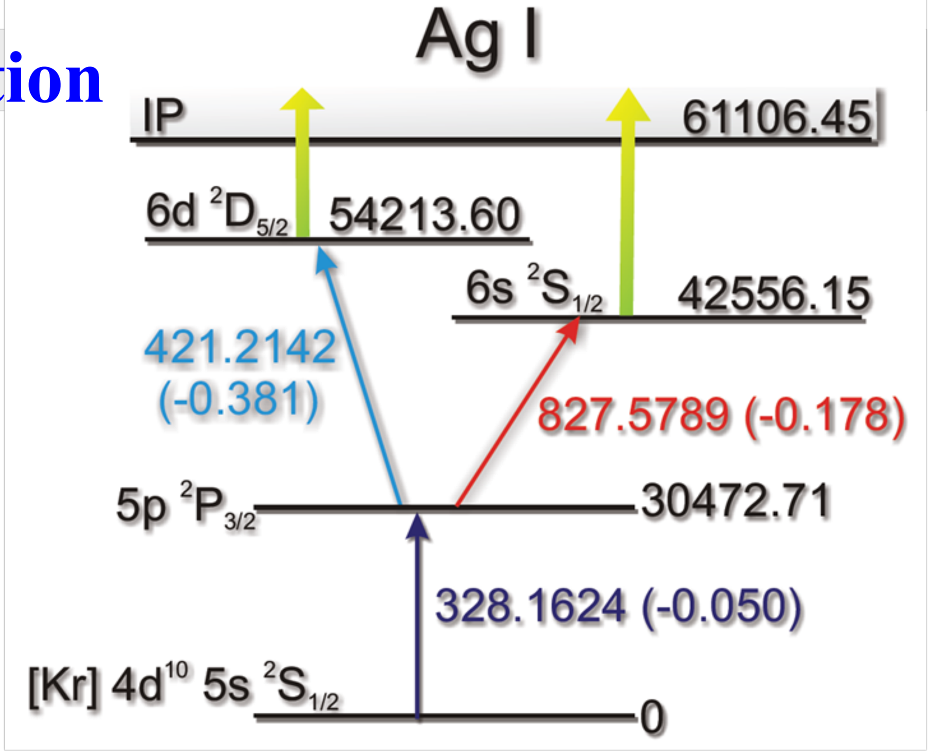
Mainz Titanium:Sapphire Laser

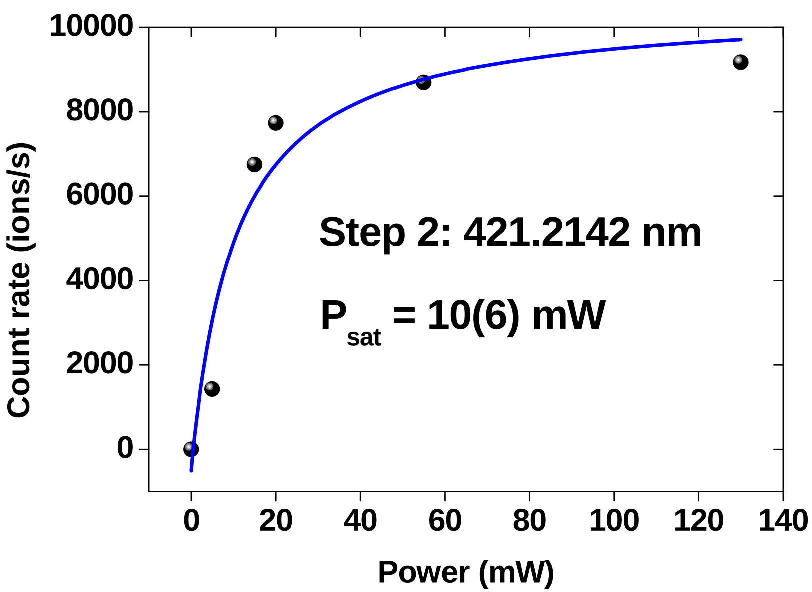


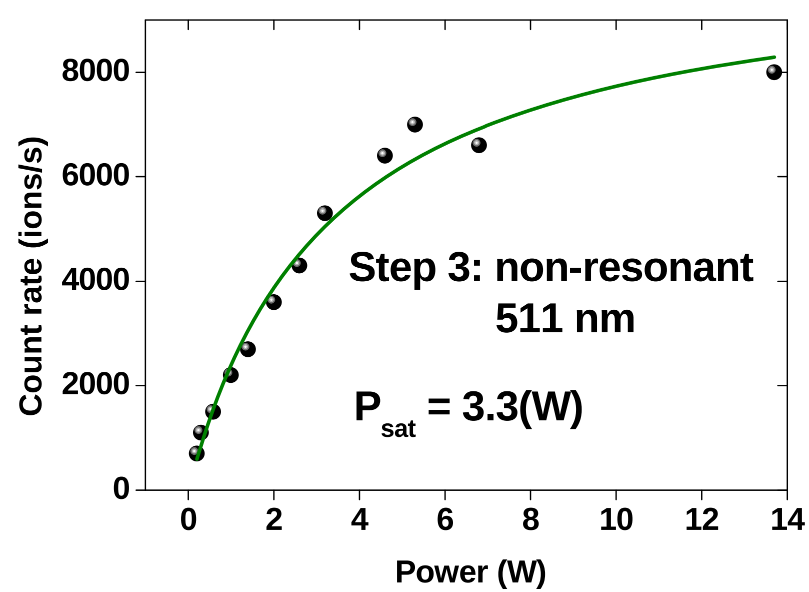
R. Horn, Dissertation Universität Mainz 2003

Non-resonant laser ionization

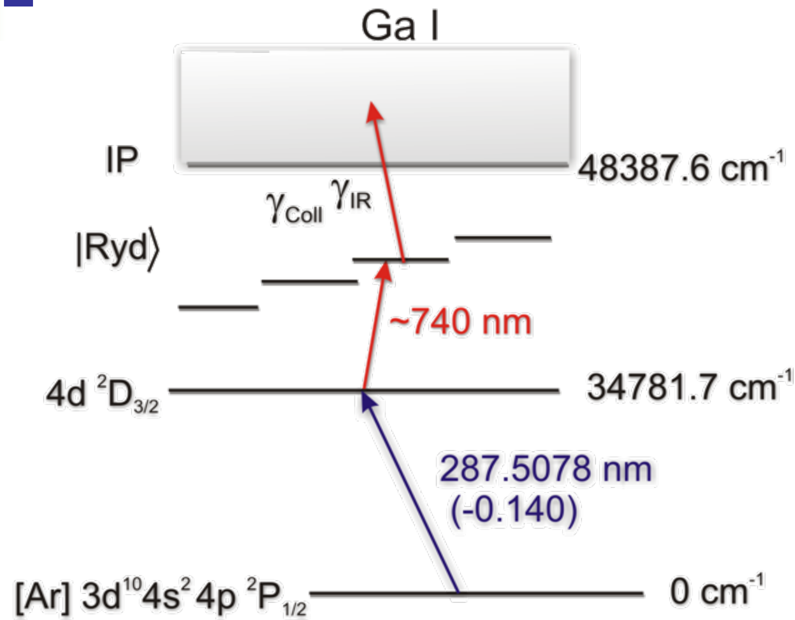


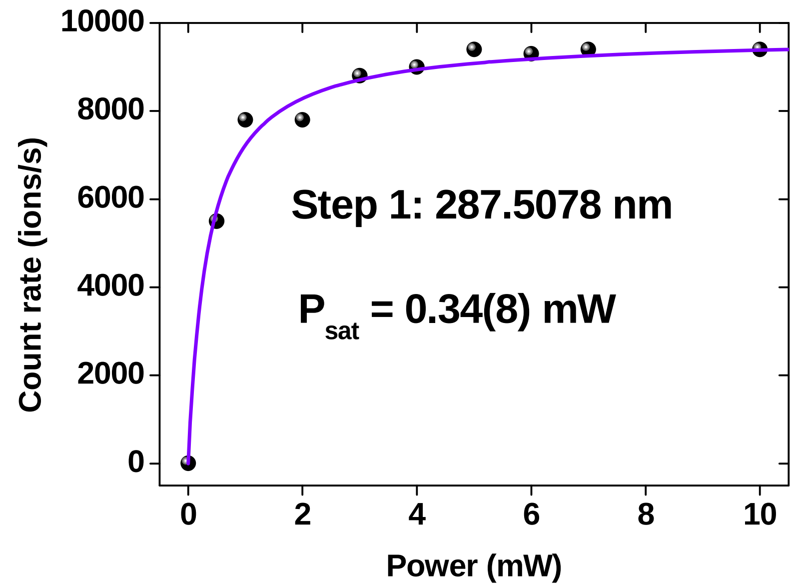


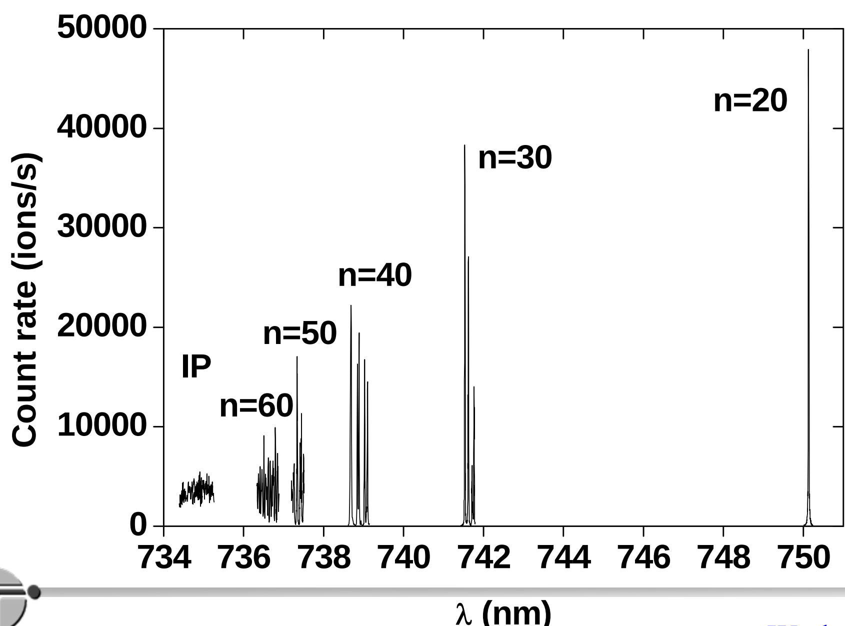


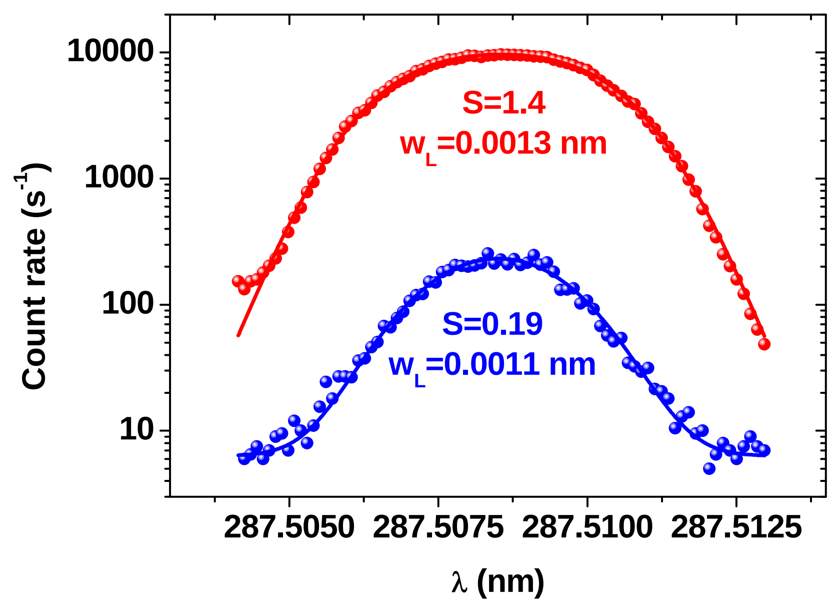


Ionization via Rydberg states - Gallium

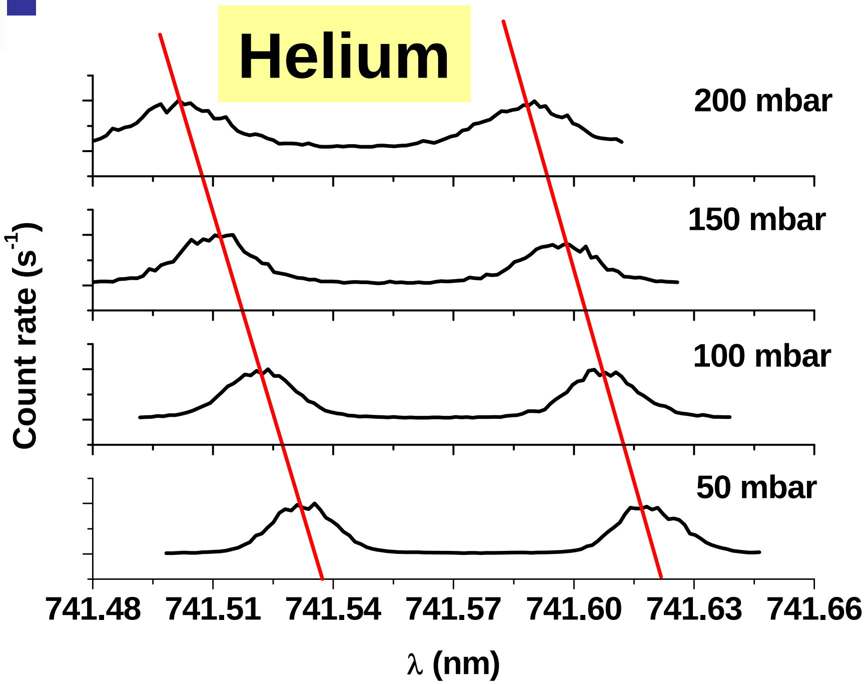


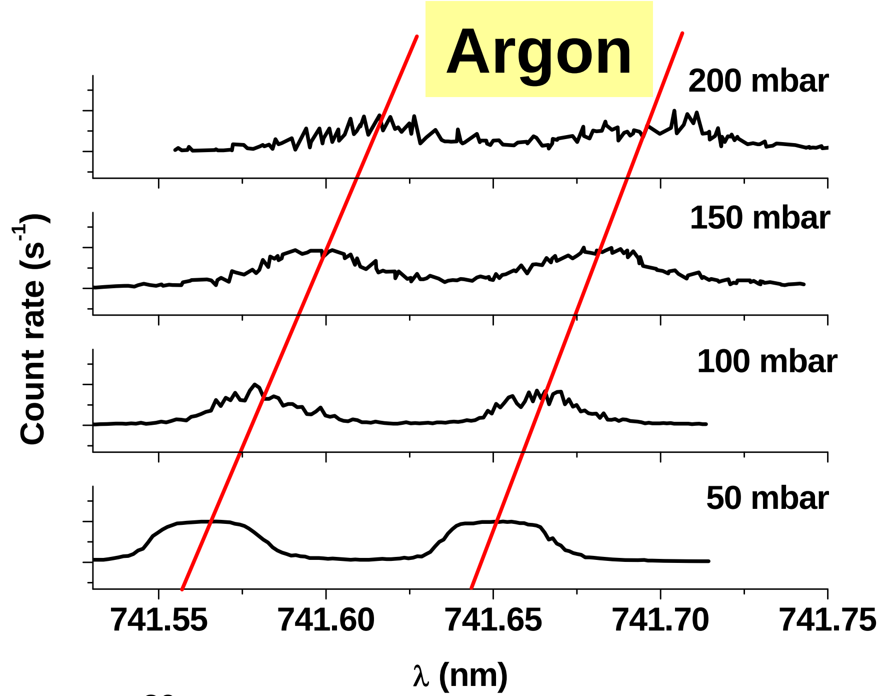






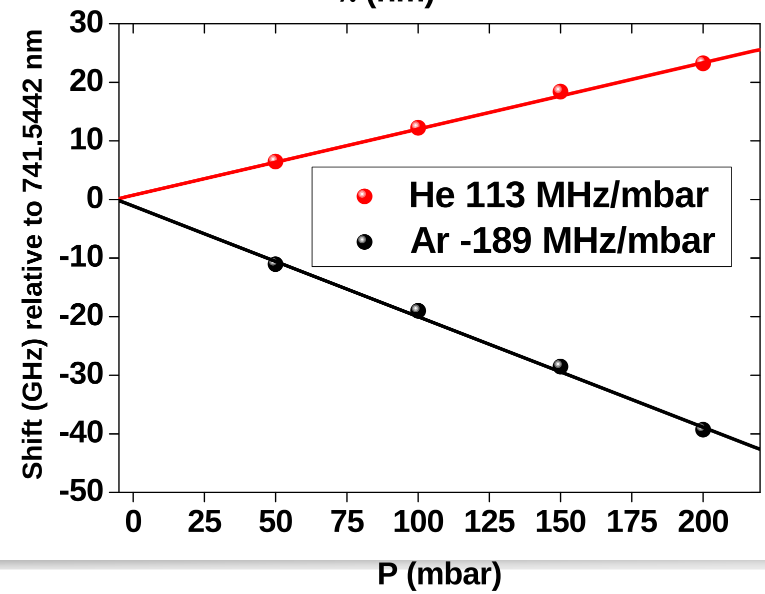
Pressure shifts of n=30 state in He and Ar





Plotting the centroid shifts vs pressure provides extrapolated ”unperturbed” value for Rydberg transition.

Scattering length of He and Ar has different signs (He = 1.19, Ar = -1.7) hence the shifts have opposite signs.



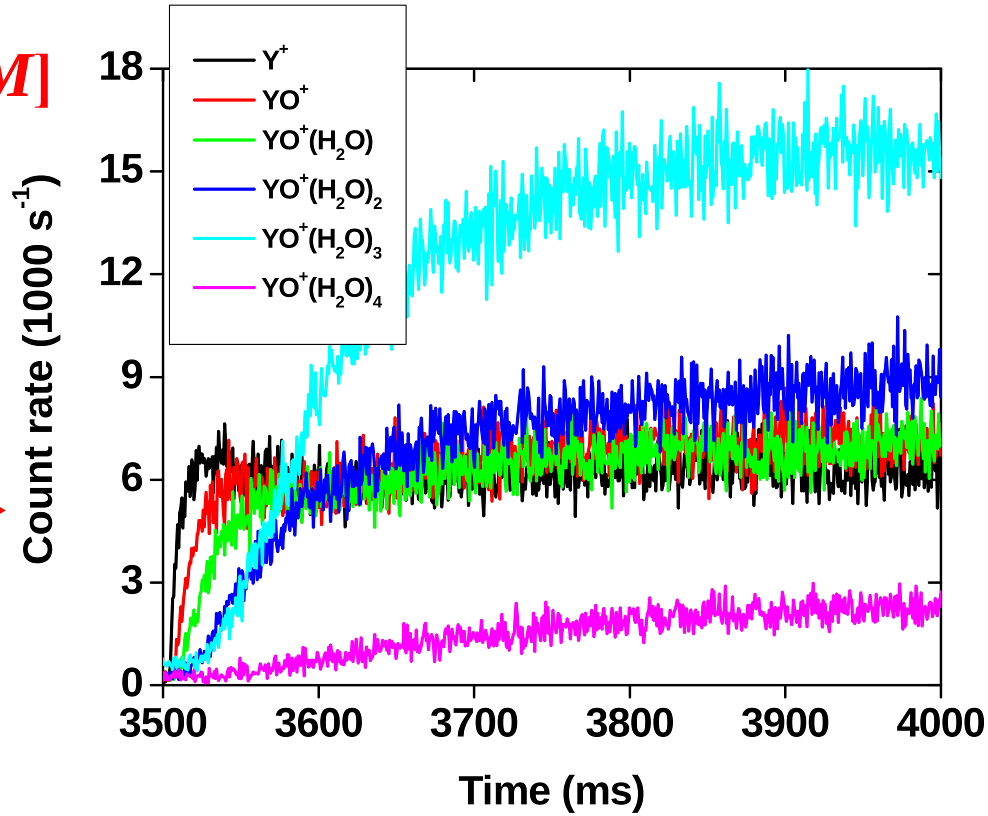
The issue of gas purity control

Molecular ion formation:
$X^+ + M \rightarrow XM^+$

$dn/dt = -kn[M] \longrightarrow \tau = 1/k[M]$

Impurities at 0.1 ppm level

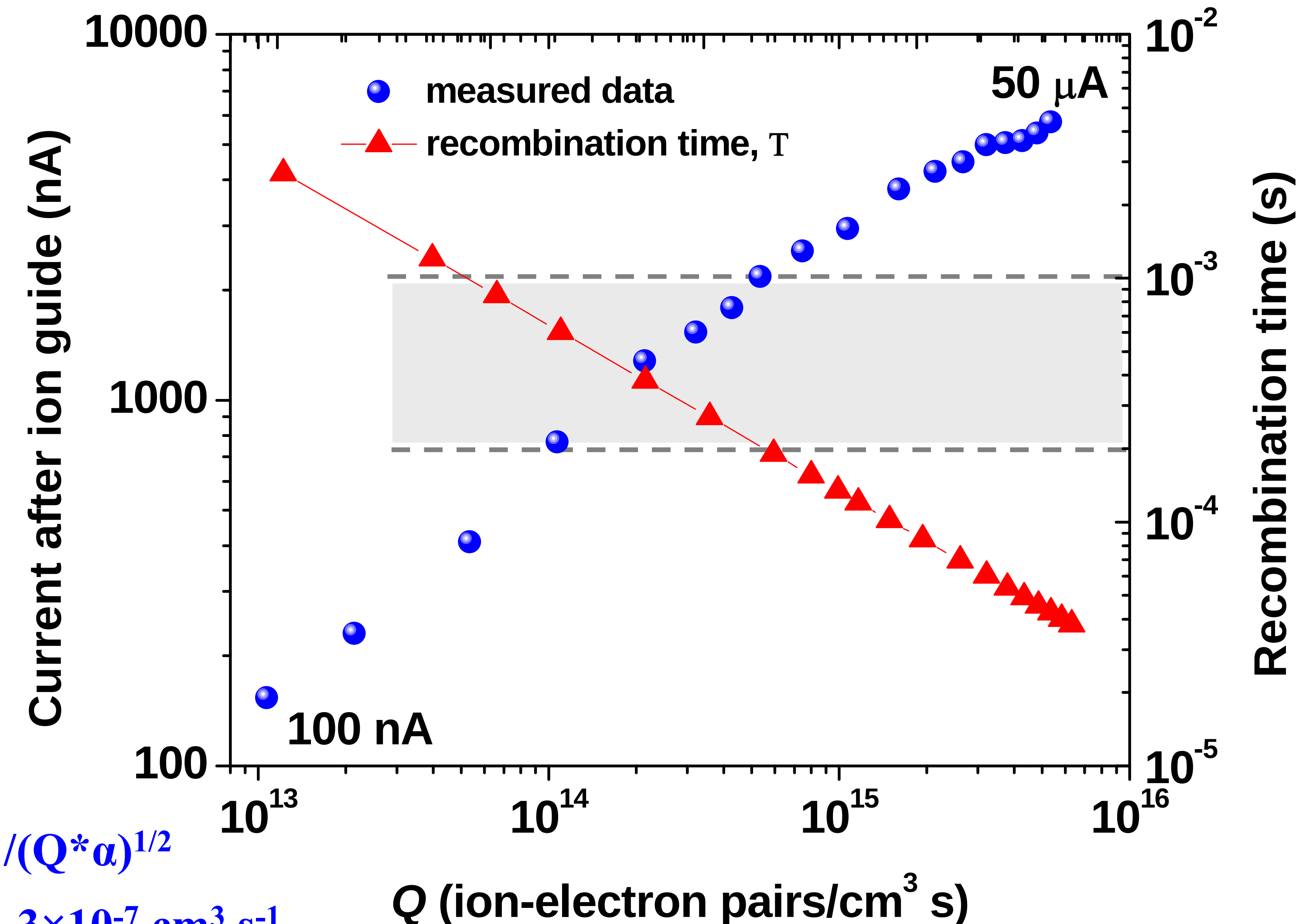
Reaction	Rate constant k (cm^3s^{-1})
$Mo^+ + O_2$	7.5×10^{-11}
$Ru^+ + O_2$	1.7×10^{-13}
$Rh^+ + O_2$	9.2×10^{-14}
$Ti^+ + H_20$	6.1×10^{-11}
$Ti^+ + O_2$	4.6×10^{-10}
✓ $Y^+ + O_2$	4.1×10^{-10}
✓ $Th^+ + O_2$	6.0×10^{-10}
✓ $U^+ + O_2$	8.5×10^{-10}
✓ $Zr^+ + O_2$	5.0×10^{-10}
$Ag^+ + O_2$	1.0×10^{-13}



150 mbar He. Impurity level ~0.1 ppm. The reaction time for yttrium reacting with oxygen and forming a molecule is ~5 ms. Total evac time ~500 ms.

T. Kessler, I.D. Moore *et al.*, Nucl. Instr. And Meth. B 266 (2008) 681

Are there limitations to the ion guide production?



$T = 1/(Q*\alpha)^{1/2}$

$\alpha = 1.3\times10^{-7}$ cm^3 s^{-1}

P. Karvonen, I.D. Moore *et al.*, Nucl. Instr. and Meth. B 266 (2008) 4794

First on-line studies (April 2009)

30 MeV α beam, 1 μA, on 12.5 μm Ni window. 200 mbar Ar.