Lecture 3: Techniques of laser spectroscopy for radioactive nuclei

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Outline of Lecture 3

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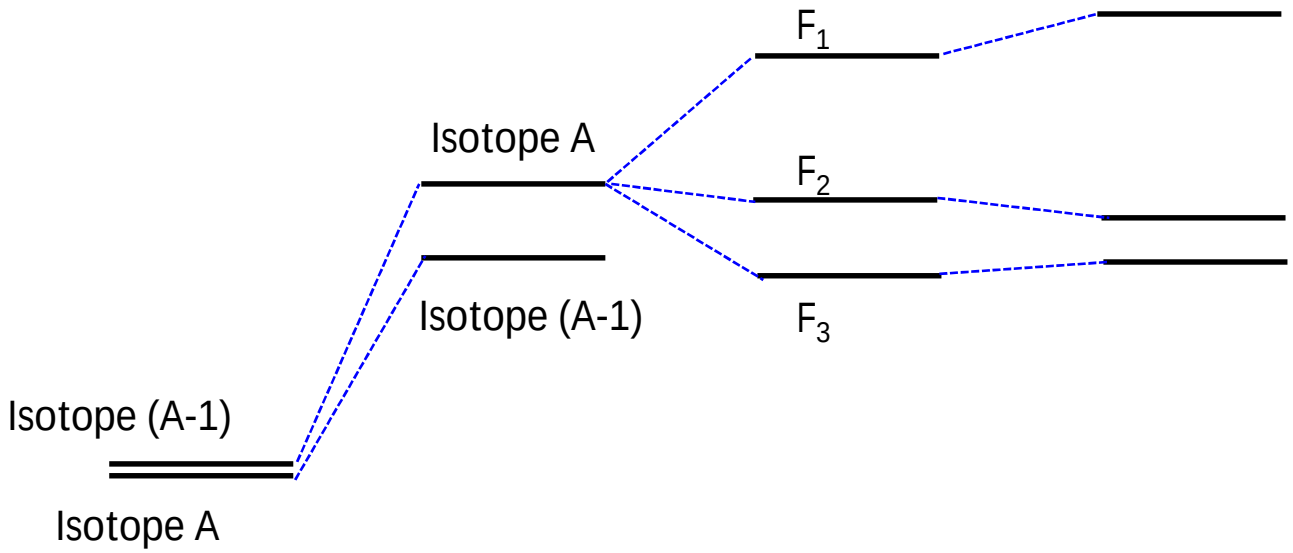
- Brief introduction to optical spectroscopy
- Resonance ionization spectroscopy (``in source´´)
- Environmental limitations
- Towards higher-resolution studies ``near´´ the ion source
- Traditional collinear laser spectroscopy
- Sensitivity vs. resolution

*Links to the discussion paper: Raggio *et al.*, Scientific Reports 14 (2024) 14

Reminder from lecture 1

We discussed atomic structure and the effect of perturbations between the nucleus and the electrons. This led to the isotope-dependent hyperfine structure of the atom + the nucleus!

Point nucleus + Finite size + Magnetic dipole + Electric quadrupole



+ higher order perturbative HFS
+ higher multipole order HFS
+ higher order radial moment isotope shift
+ Zeeman effect
+ Bohr Weisskopf effect
+ Breit-Rosenthal effect
+ ...

Mass shift + Field shift $-\mu B_e \cos\theta + \frac{1}{4} eQ_0 V_{JJ} P_2(\cos\theta)$

All of this is *not 'new'* physics. It is (in practice often poorly!) known.

Let’s mention something about energy scales...

Optical transitions: 100’s of THz, a few eV, 10000s of cm^{-1}, 100s of nm

Hyperfine splitting: 100’s of MHz, a few μeV, ~0.01 cm^{-1}

Optical linewidth: 100’s of MHz, a few μeV, ~0.01 cm^{-1}

1 cm^{-1}: ~ 30000 MHz

1eV: ~ 8000 cm^{-1}

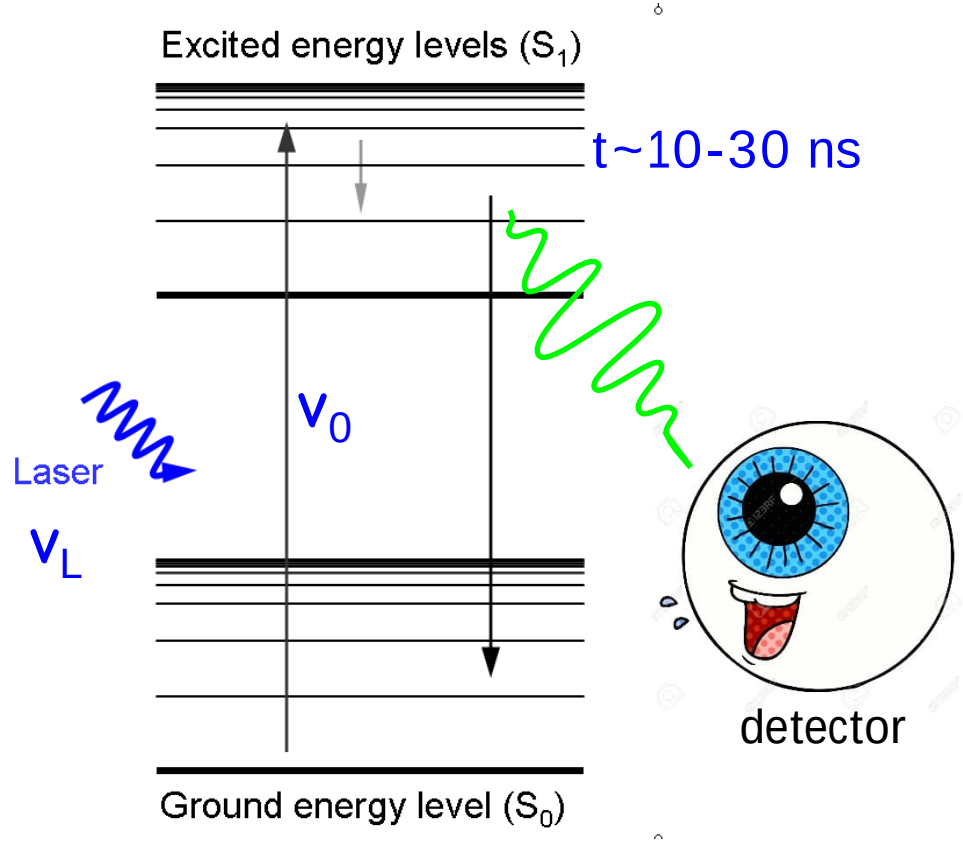
How do we measure these effects in practice?

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

We use lasers to induce transitions in the atom, and record the response of the atom as a function of laser frequency

= Laser spectroscopy

What is laser spectroscopy?

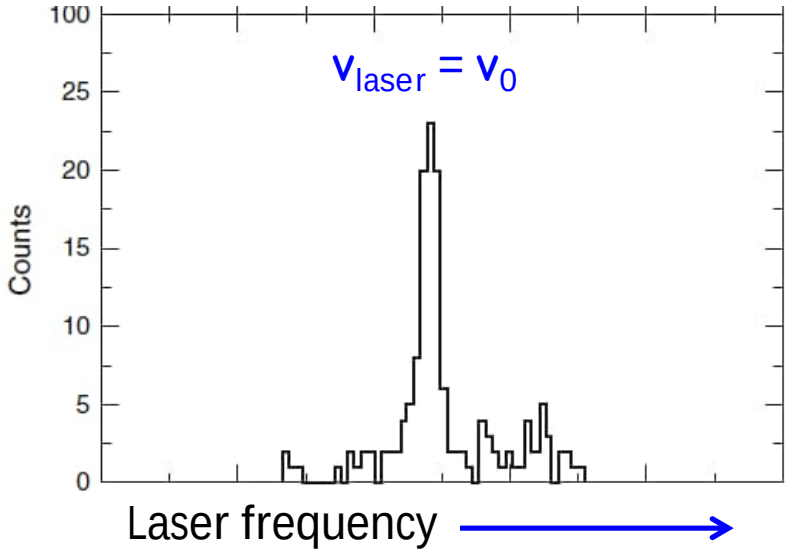


- Detect the photons the atom emits when it relaxes back to the ground (or other) state

= Fluorescence detection

The absorption cross section of a photon by an atom and subsequent relaxation is given by a Lorentzian function:

$$\sigma = \frac{\lambda^2}{2\pi}\left\{\frac{1}{1+[4\pi\tau(\nu-\nu_0)]^2}\right\}$$

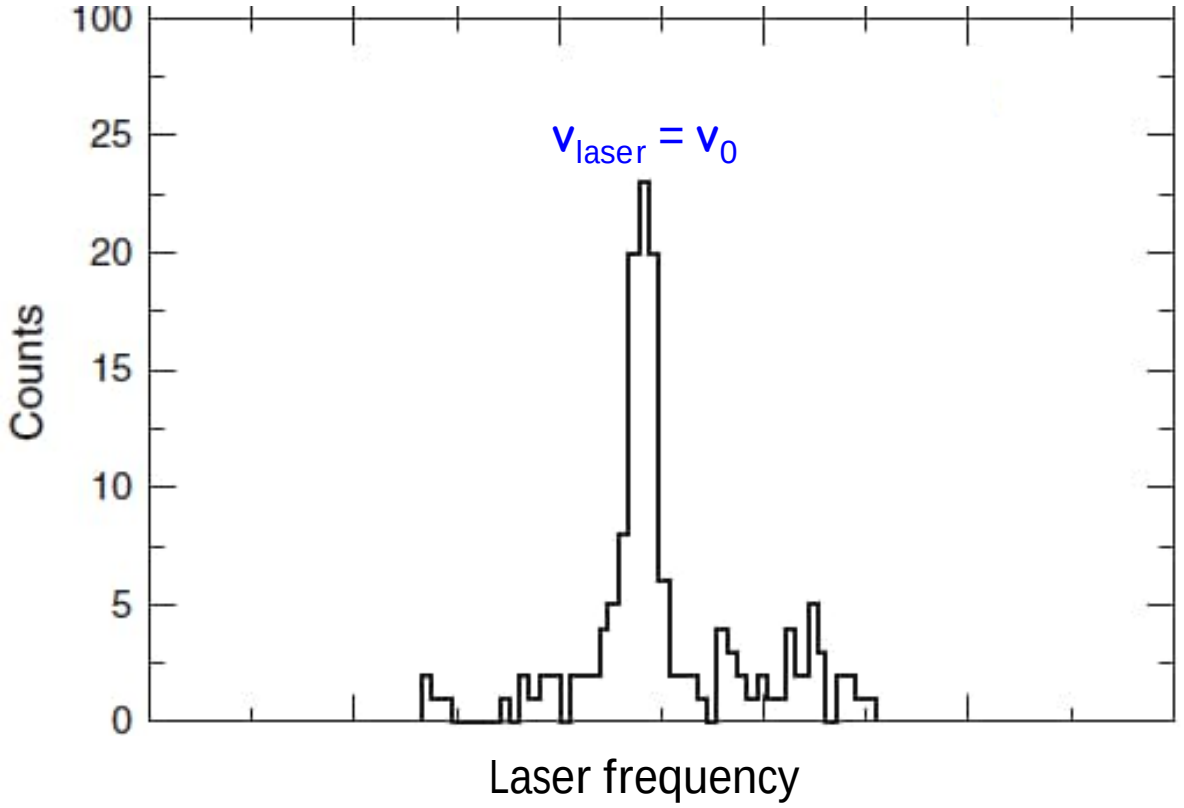


When $\nu_L = \nu_0$, resonant absorption happens.

$$\sigma \approx \lambda^2 / 2\pi$$

Discussion pause...

The absorption cross section of a photon by an atom and subsequent relaxation is given by a Lorentzian function:



What determines the width of these peaks?

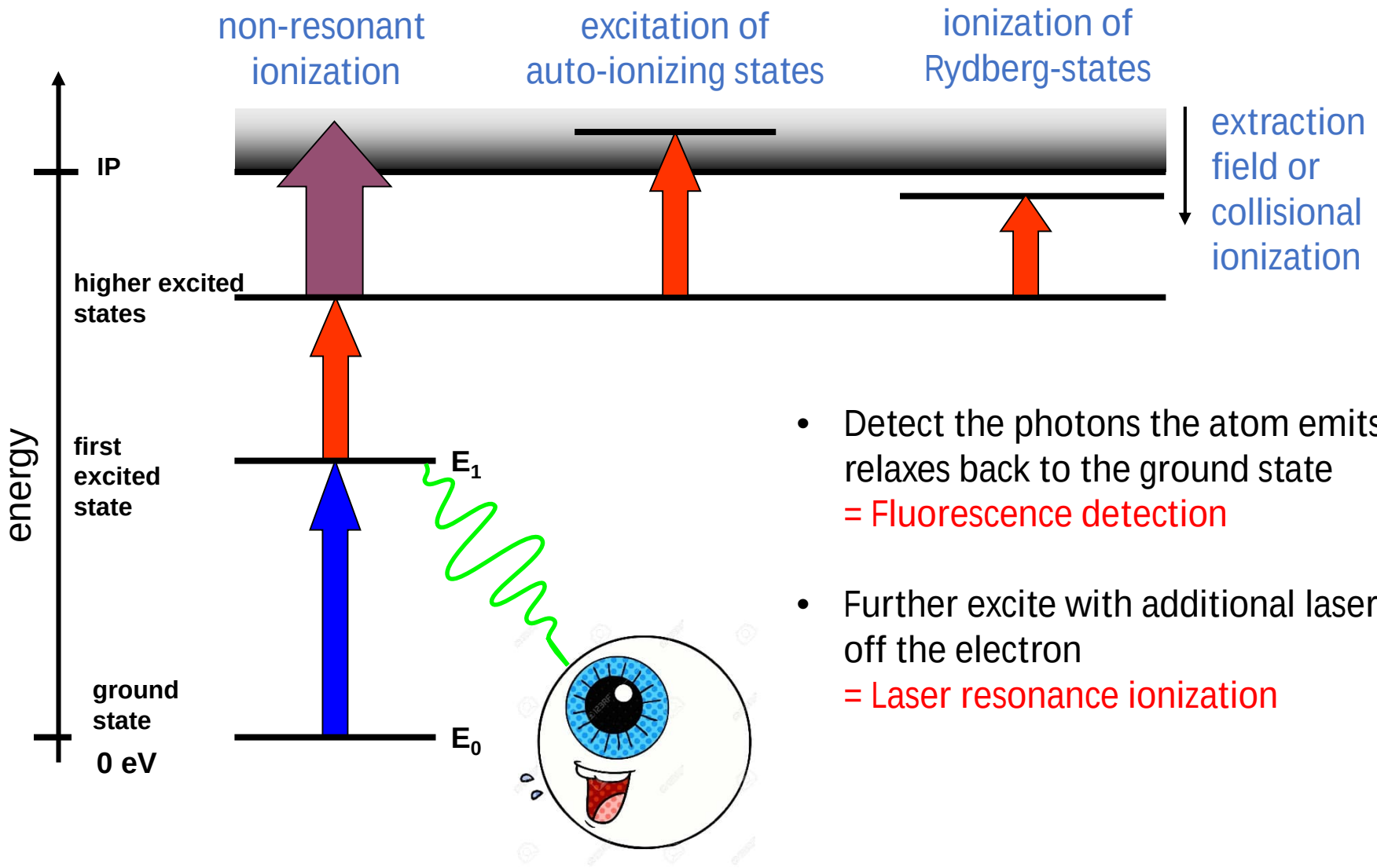
Answer: Natural line broadening:

$$\Delta t \, \Delta E \geq \frac{\hbar}{2}$$

The finite lifetime means the energy spread cannot be zero.

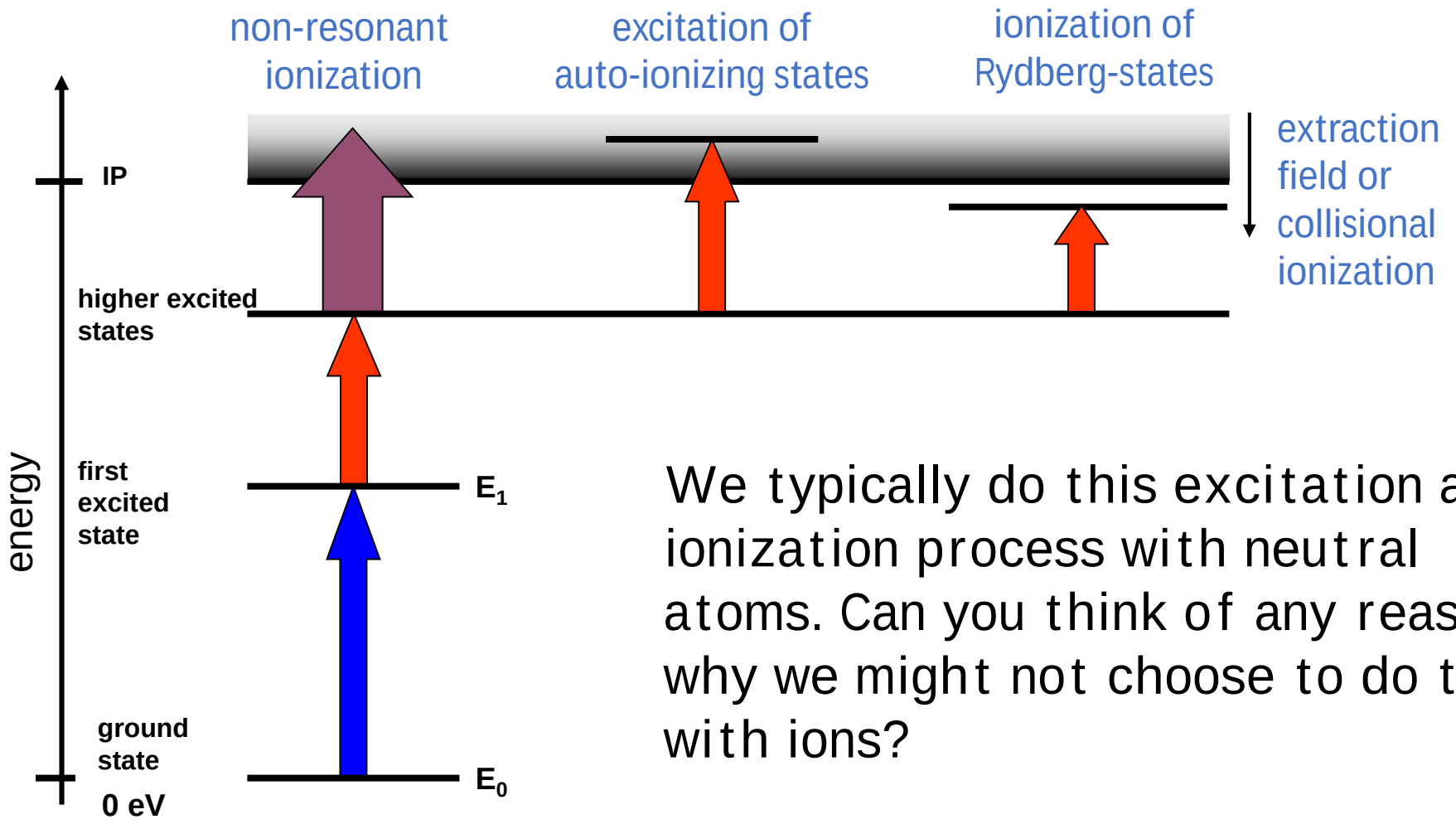
Dipole-allowed transitions: ~10 MHz

What is laser spectroscopy?



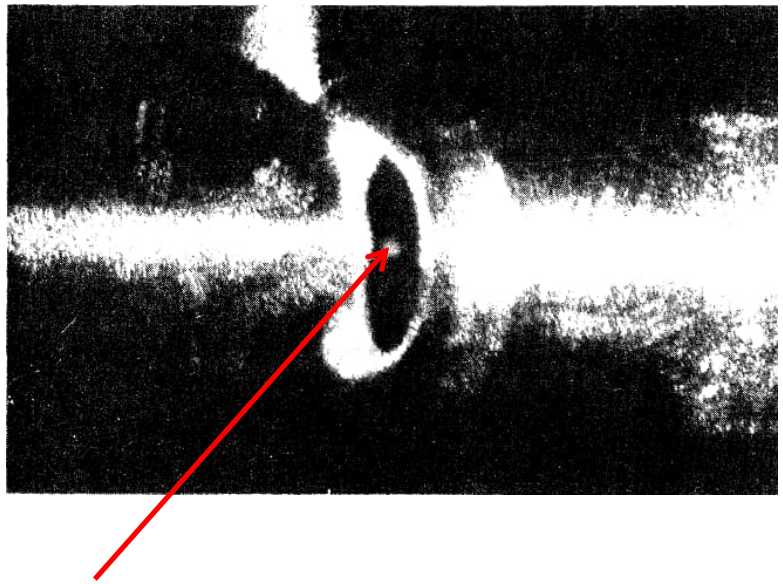
- Detect the photons the atom emits when it relaxes back to the ground state
  = Fluorescence detection
- Further excite with additional lasers and peel off the electron
  = Laser resonance ionization

Discussion pause...



We typically do this excitation and ionization process with neutral atoms. Can you think of any reasons why we might not choose to do this with ions?

Detection of single trapped ions



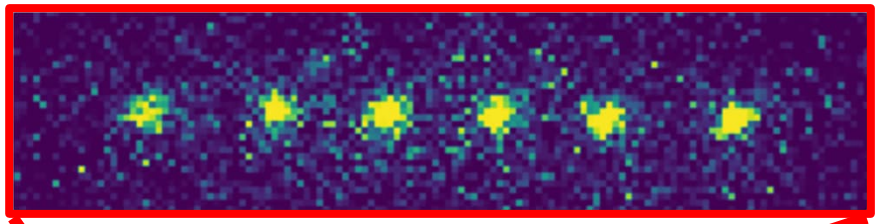
A trapped Ba^+ ion cloud with estimated number <50 ions in the cloud

W. Neuhauser et al, PRL 41 (1978) 233

Recent work from KU Leuven – trapping Sr^+ ions:

- Each ion is emitting a few 10^7 photons/s
- ~40000 photons/s/ion are detected.

~ 15 µm



Courtesy: Ruben de Groote, KU Leuven

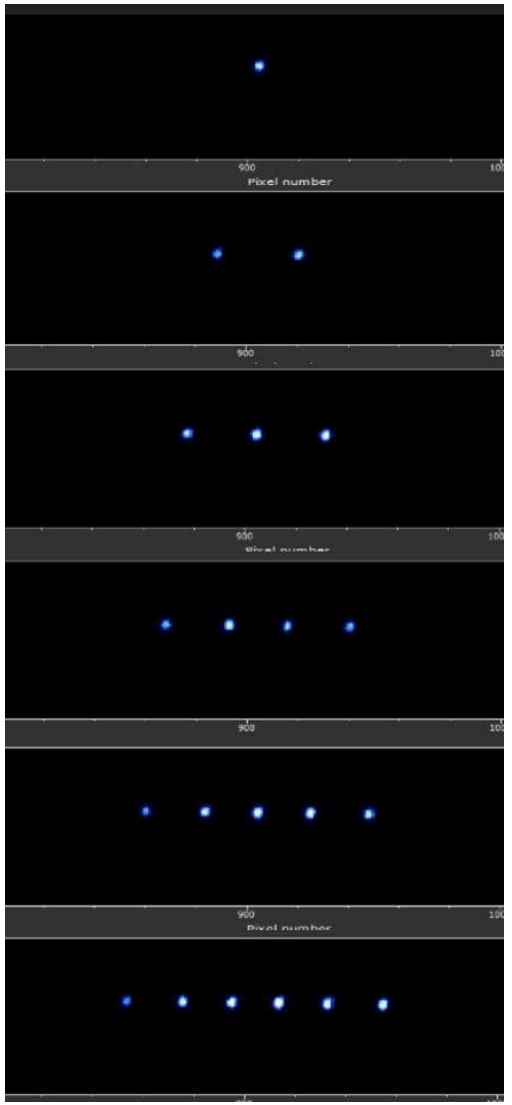
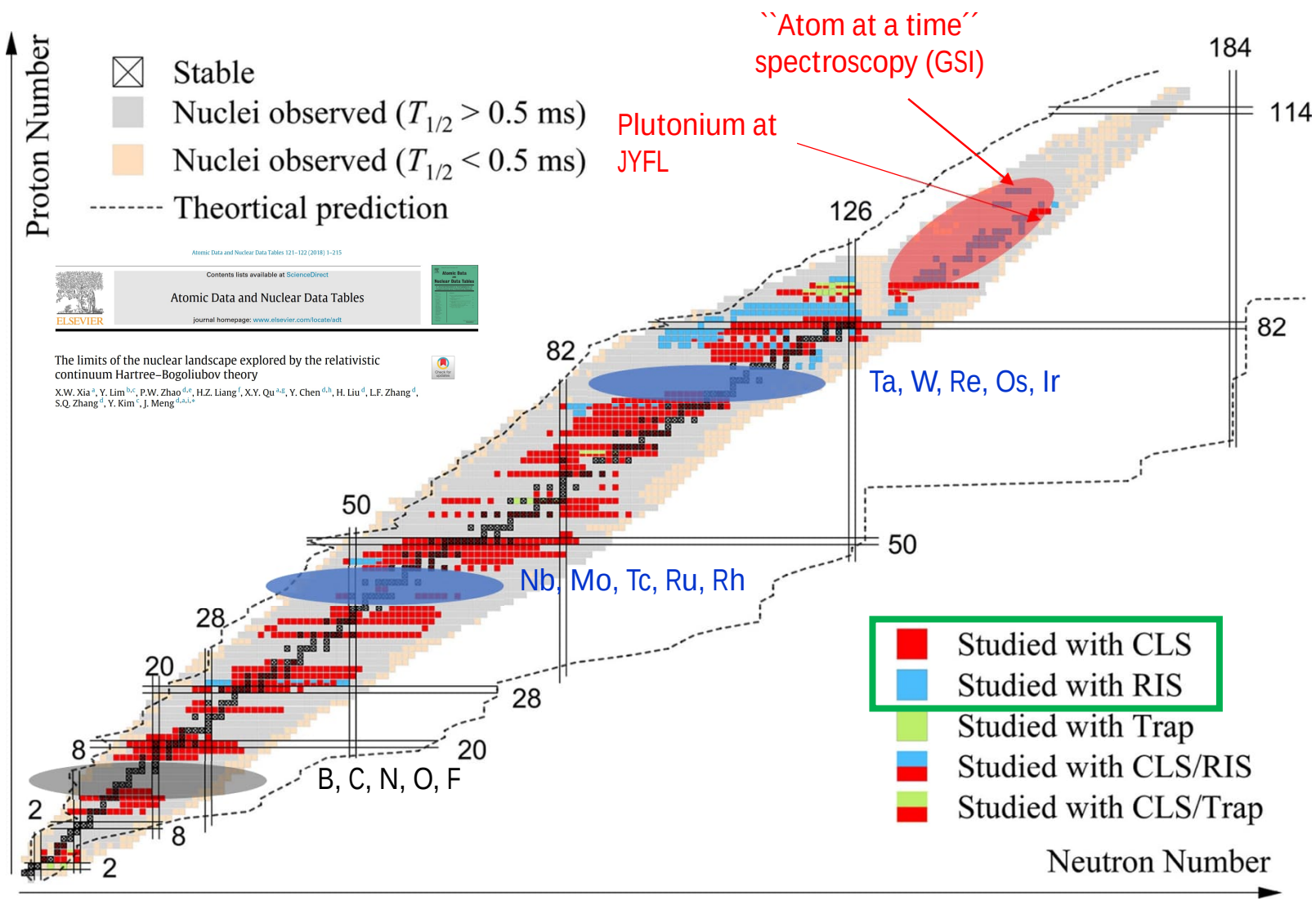


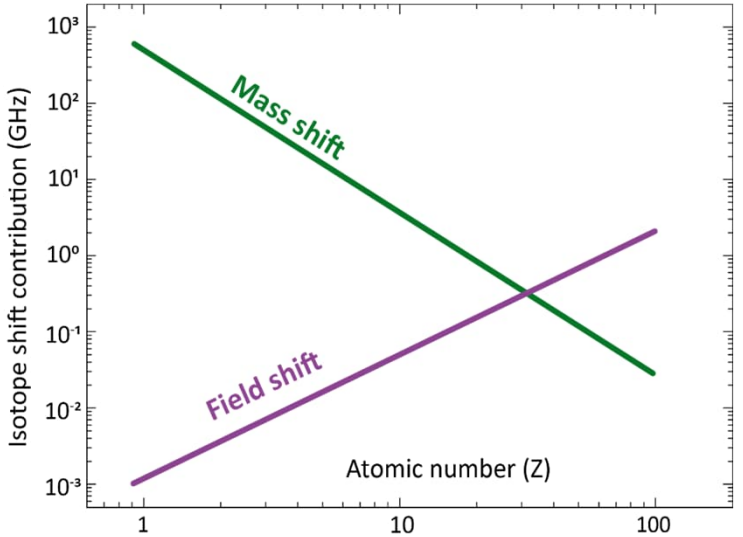
Chart of nuclides from optical spectroscopy



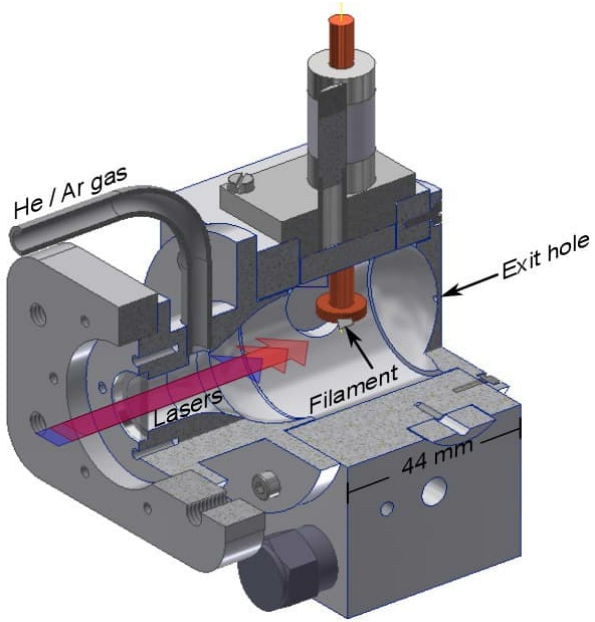
Latest review: X.F. Yang et al., PPNP 129 (2023) 104005

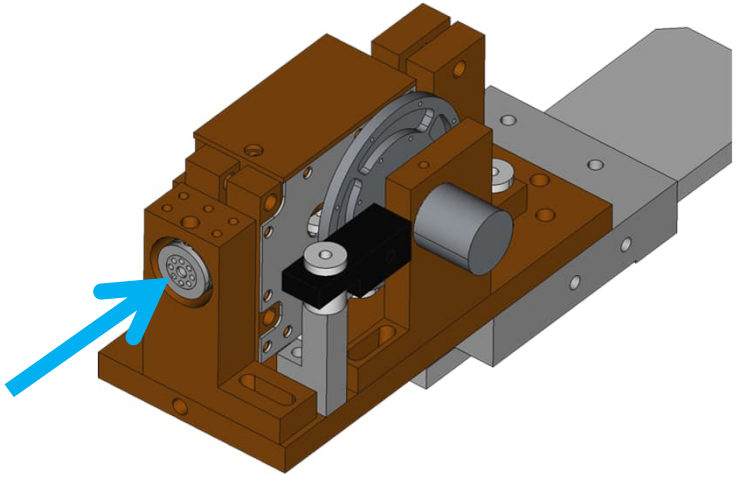
Limitations due to production and atomic properties

- Actinide region (low production)
- Refractory elements (target/ion source developments needed)
- Light elements are very reactive (molecular compounds). Also require laser developments



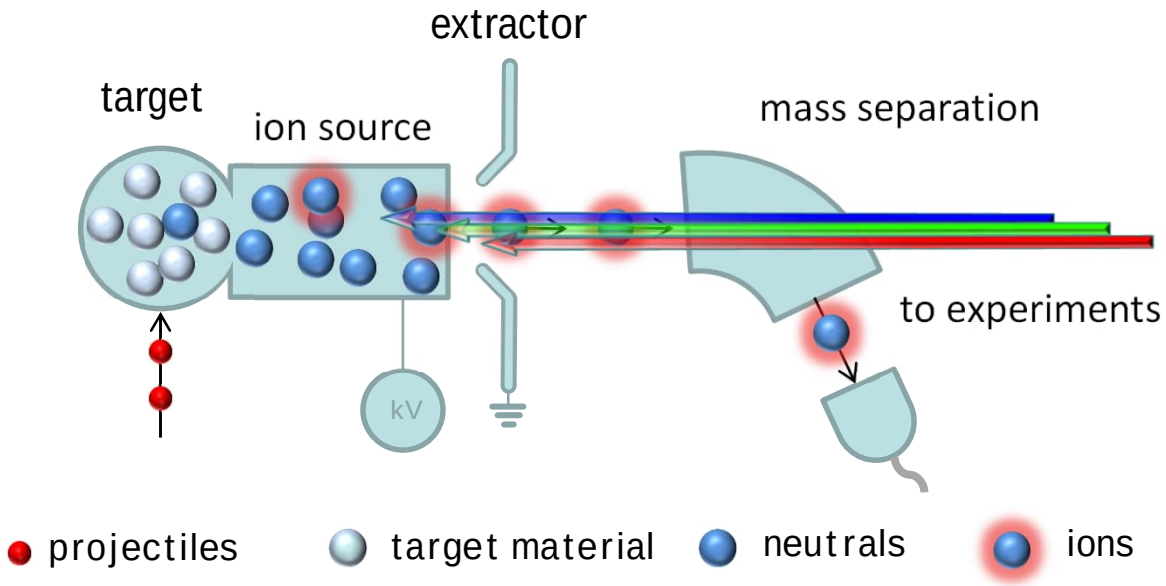
In-source laser spectroscopy

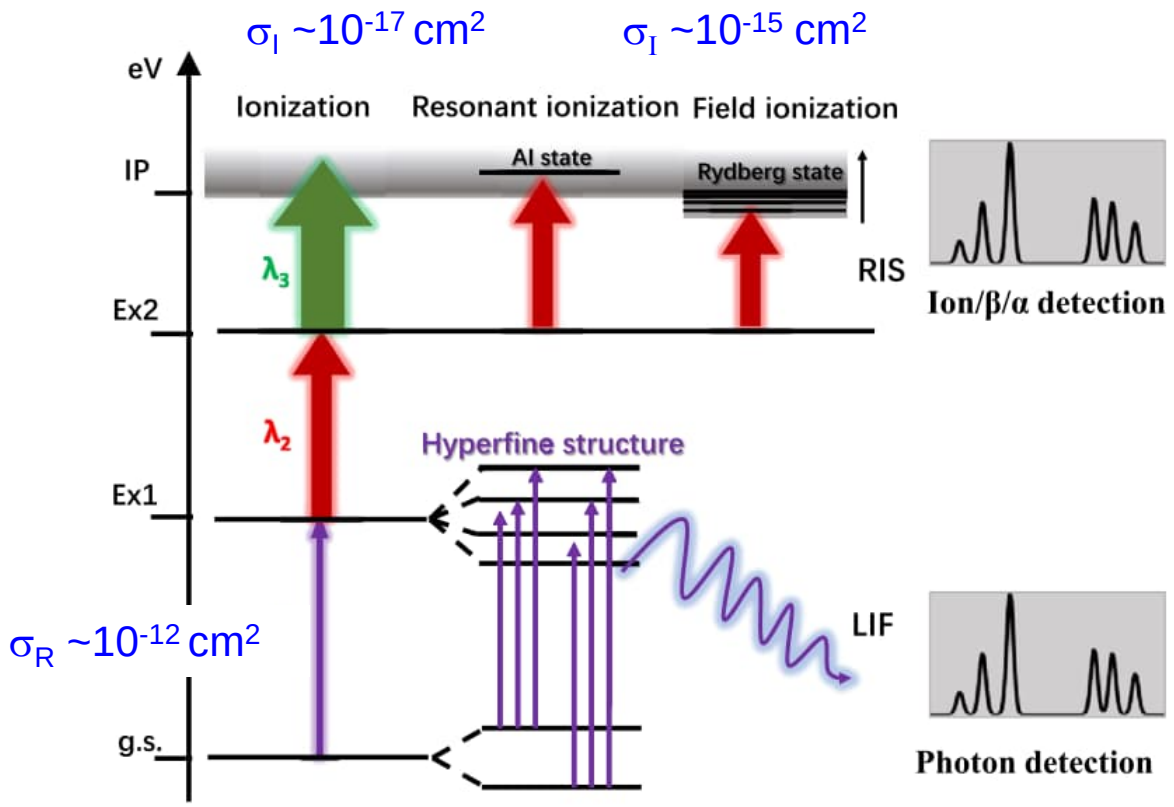




Resonance ionization spectroscopy (RIS)

Resonance Ionization Spectroscopy (RIS) is a close variant to the use of laser ionization for selective and efficient radioactive ion beam production. This is the most direct form of laser spectroscopy at a RIB facility.

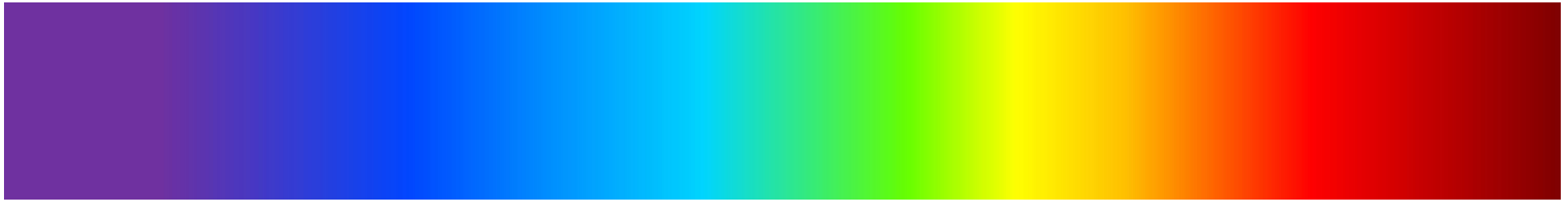




- Selective process (laser ``on´´ vs ``off´´)
- Efficient process (>>1% is routine)
- ``Short´´ lifetimes
- Access to low yields (<<1 ion/s)
- High detection efficiency (ions/α decays…)

Remember from L2: we need pulsed lasers to efficiently ionize!

Laser tunability is very important!

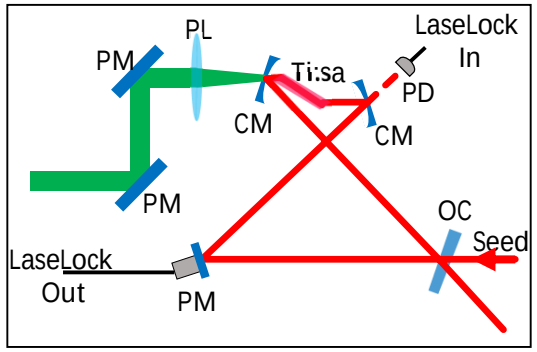


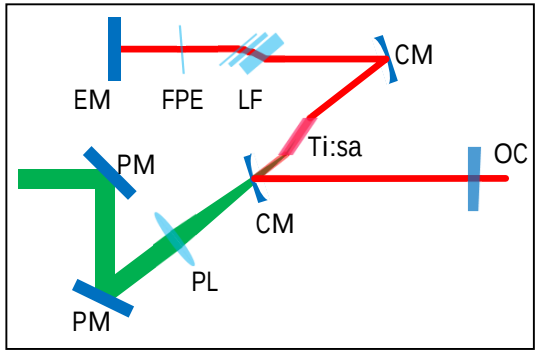
A community-driven laser scheme database

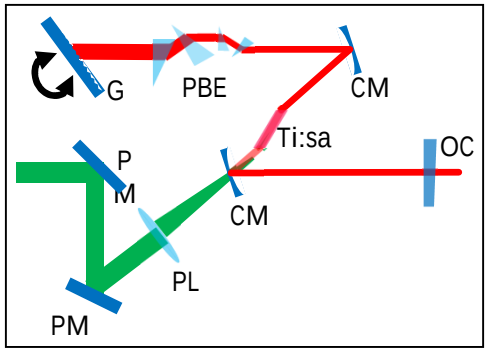
https://rims-code.github.io

Courtesy, Reinhard Heinke, ISOLDE/Manchester

The family of the Mainz Ti:sapphire lasers

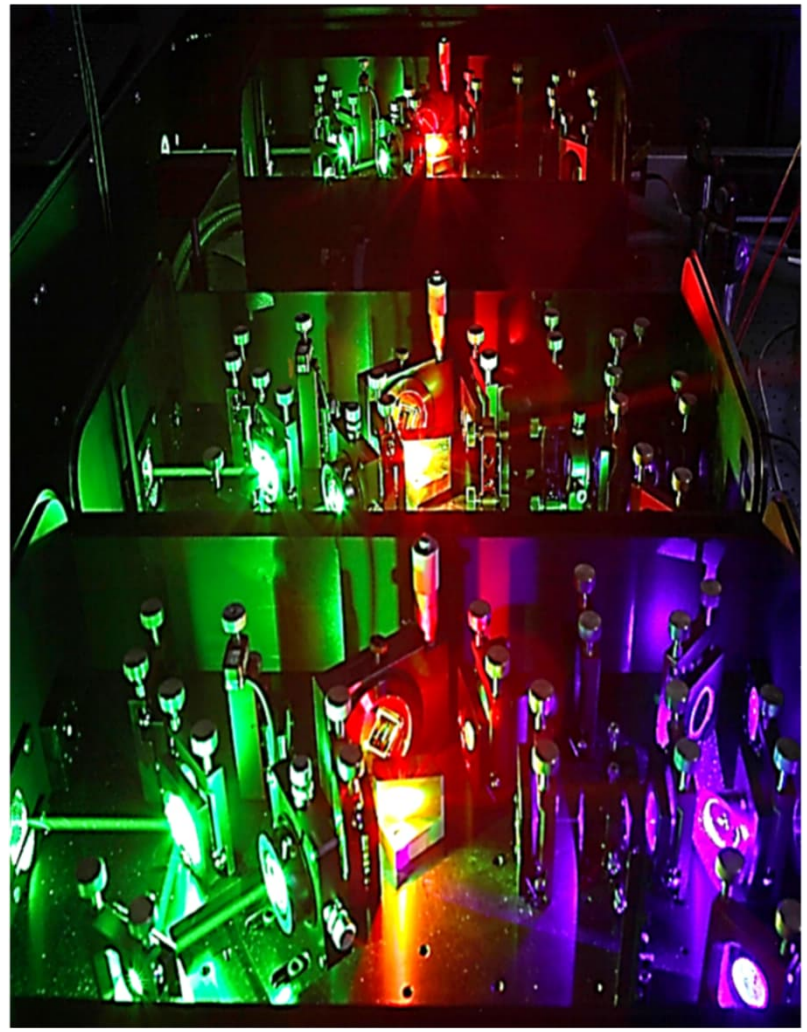






Specifications	Injection-locked laser	Standard laser	Grating laser
Repetition rate	10 kHz	10 kHz	10kHz
Power fund.	2 W	3 - 5 (10) W	1 - 2.5 W
Power SHG	100 mW	2 W	1 W
Pulse length	30 - 50 ns	30 - 50 ns	30 - 50 ns
Spectral width	≈ 10 MHz	2 - 10 GHz	1 - 2.5 GHz
Tuning range	10 - 20 GHz	100 GHz	690 - 1000 nm

In use at many facilities including RILIS (ISOLDE), TRILIS (TRIUMF), JYFL, GANIL...
(safer and more robust than carcinogenic dyes!!)

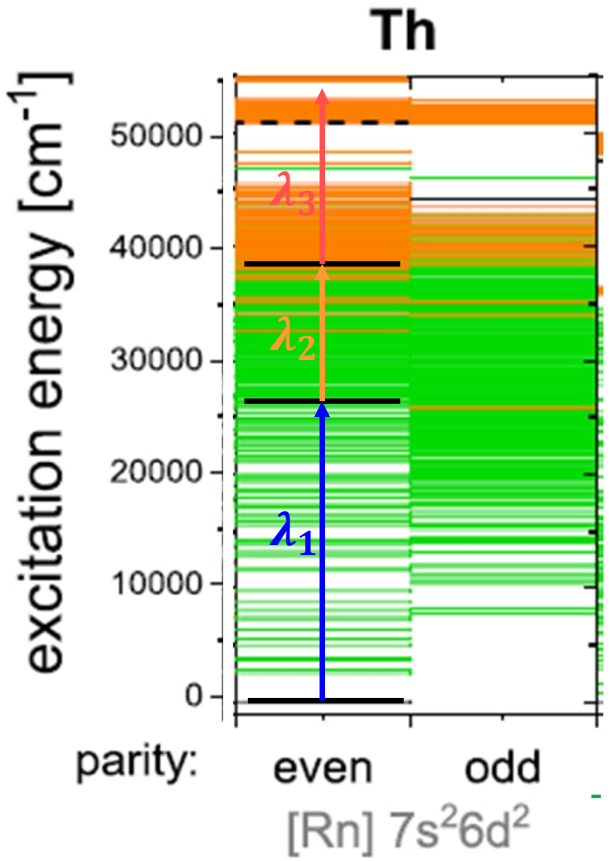


Courtesy, Klaus Wendt (Mainz)

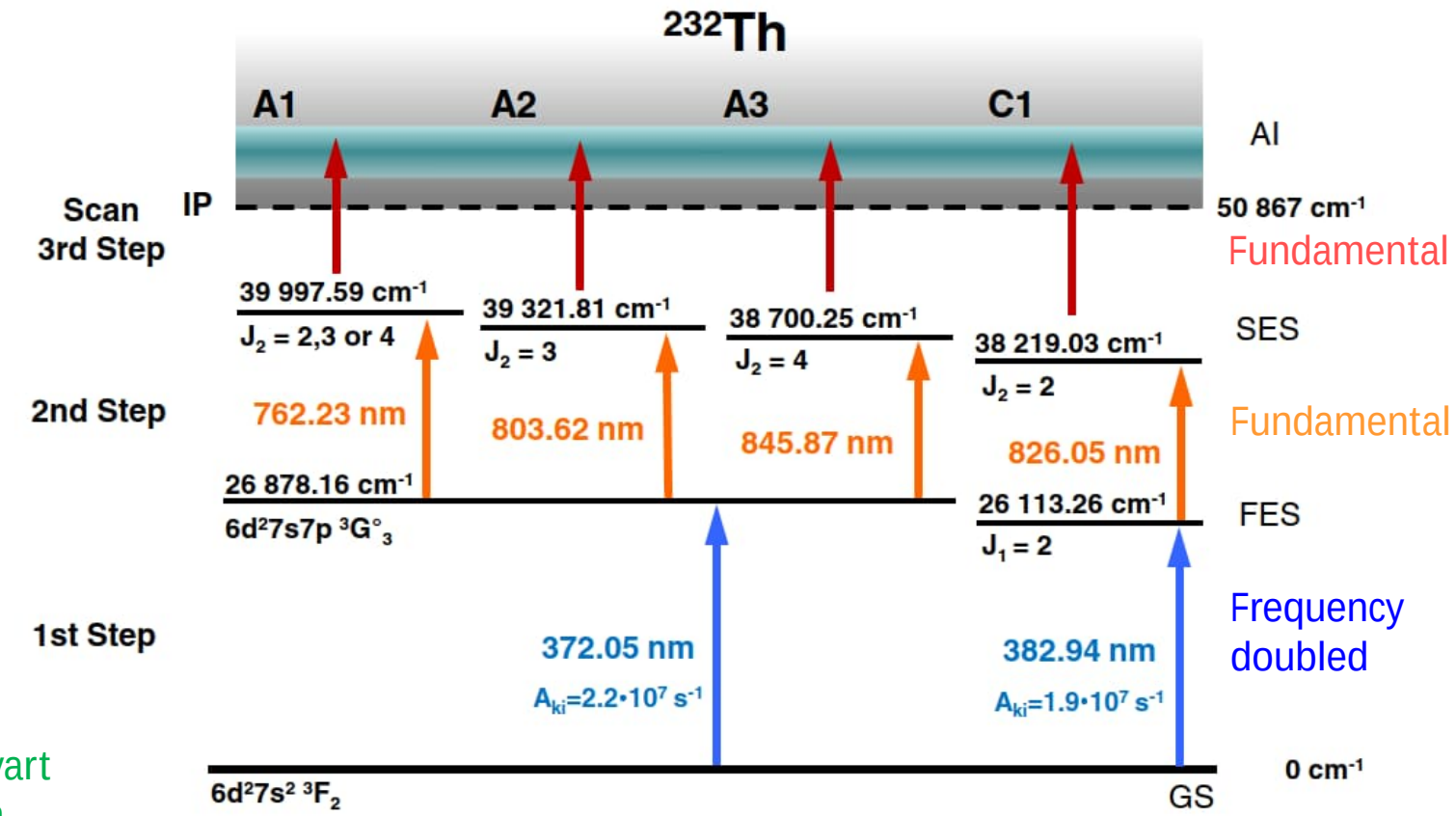
How do we develop the laser ionization scheme?

We start with a literature search in on-line atomic spectral line databases (and we try to remember what wavelengths our lasers can provide!! ☺)

- NIST atomic spectral line database
- Blaise and Wyart (actinides)

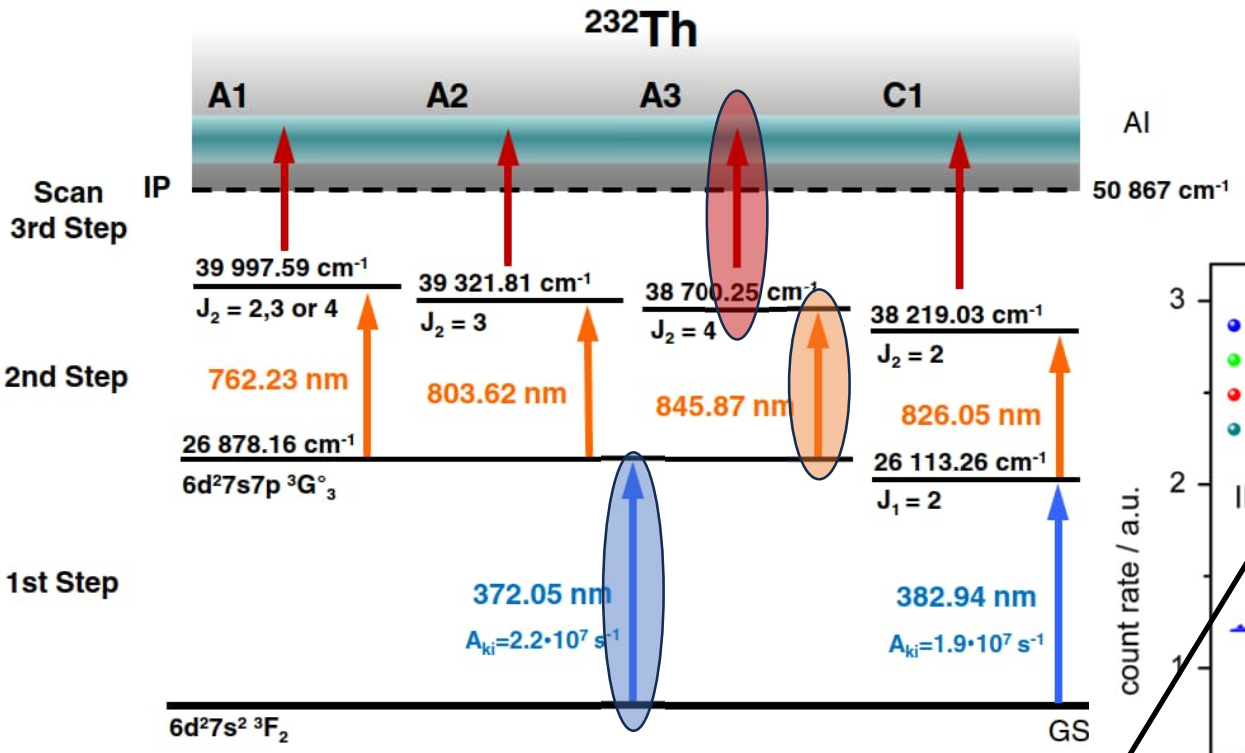


- Blaise & Wyart compilation
- Recent additions

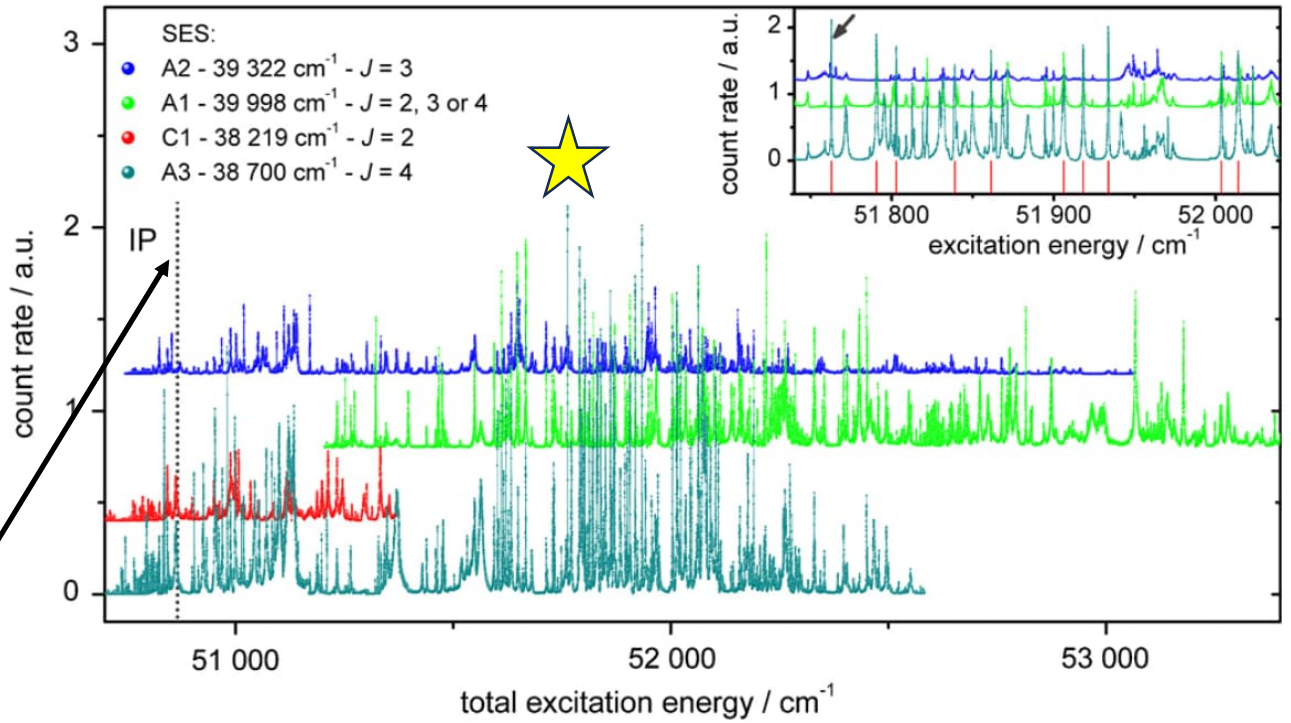


S. Raeder et al., J. Phys. B 44(2011) 165005.

Searching in the continuum!



- Basically we are looking for the peaks with the highest count rates – most efficient scheme.
- Very often, these studies are done in offline laboratories using stable samples of elements



The ionization potential is here (@ 50867 cm^{-1})

Note the high level density, complex structures, different peak widths.

S. Raeder et al., J. Phys. B 44(2011) 165005.

A short remark about atomic transitions

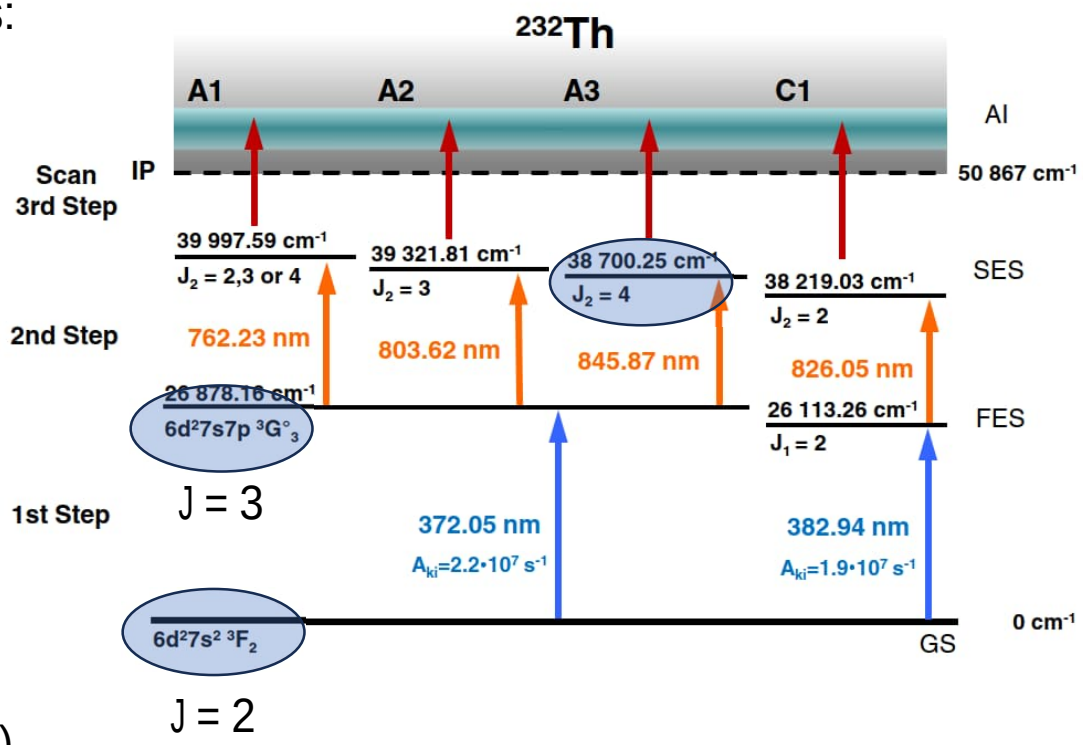
In Lecture 1 we briefly mentioned the atomic configurations:

Electron angular momenta couple:

$$J = L+S, L+S-1, \ldots, |L-S|$$

Giving the configuration for a state: $^{2S+1}L_J$

e.g., atomic level 5d 6s^2 has a configuration $^2D_{5/2}$

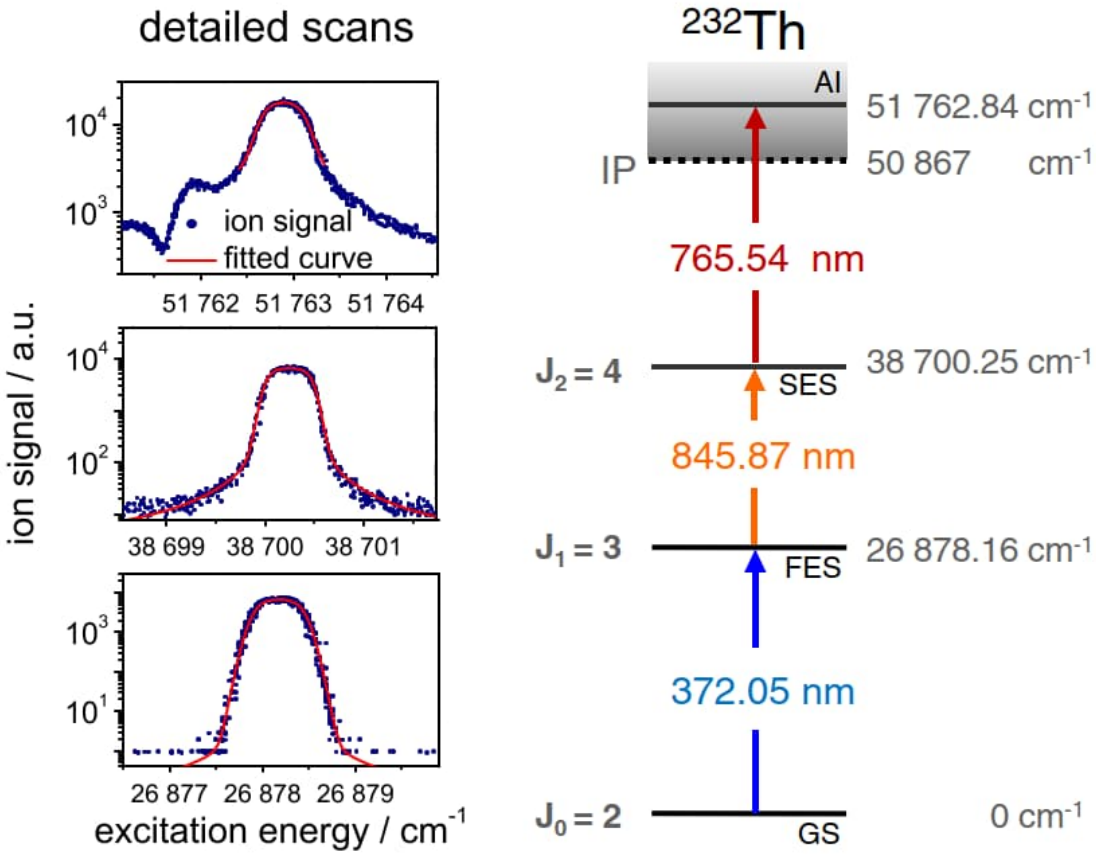


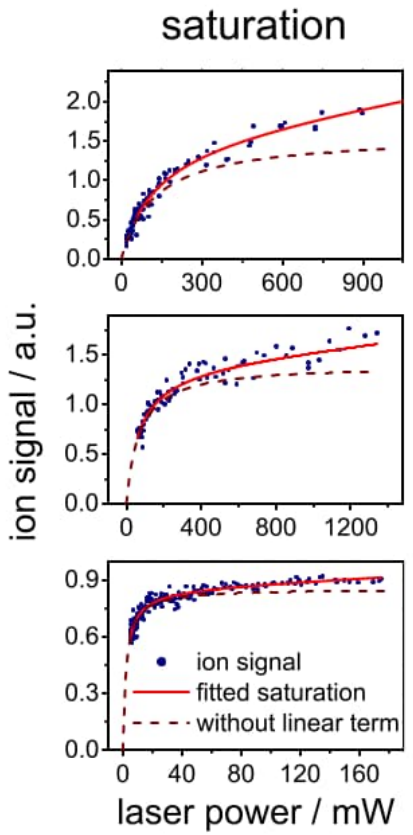
- Most probable transitions are electric dipole in nature (E1)
- Selection rules for J are: $\Delta J = 0, \pm 1$ and $J = 0 \not\rightarrow J = 0$
- Transitions are typically stronger when J increases
- The spontaneous emission rate $A_i = {}^1/_\tau$, where τ is the state lifetime

- Very often configurations of higher-lying states are unknown
- Transition strengths often unknown (and labeled according to Weak, Medium, Strong etc)!

Let's perform saturation scans

Once we have chosen the ionization scheme to be employed at the RIB facility, we also often check what laser power is required to saturate a transition…





$$f(P) = I_0 + m \cdot P + A \cdot \frac{P/P_S}{1 + P/P_S}$$

I_0 accounts for non-resonant ionization, linear increase with power (slope m) = deviations from basic saturation, P = laser power and P_S = the saturation power (related to population of the excited level).

- P_s (λ_1) = 2.2 mW
- P_s (λ_2) = 68 mW
- P_s (λ_3) = 125 mW

``Easy´´ to saturate with our laser system!

S. Raeder et al., J. Phys. B 44(2011) 165005.

Trade off between efficiency & resolution in RIS

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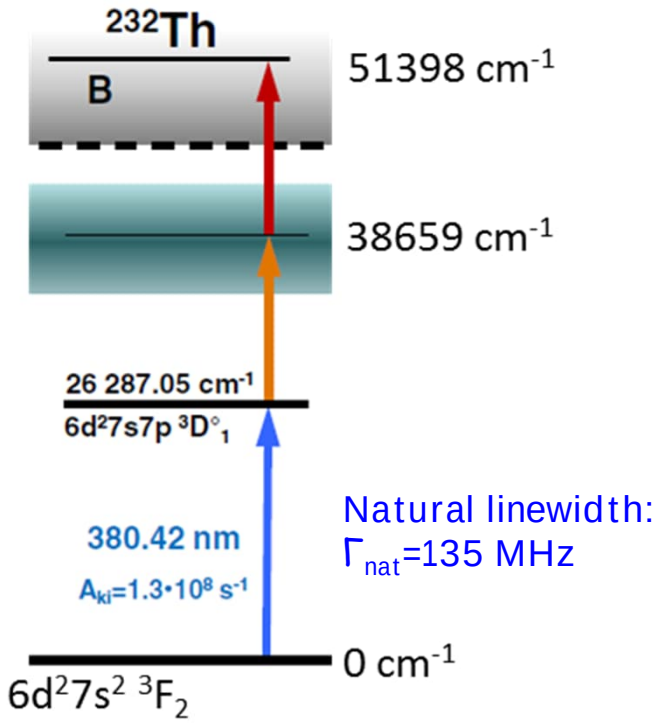
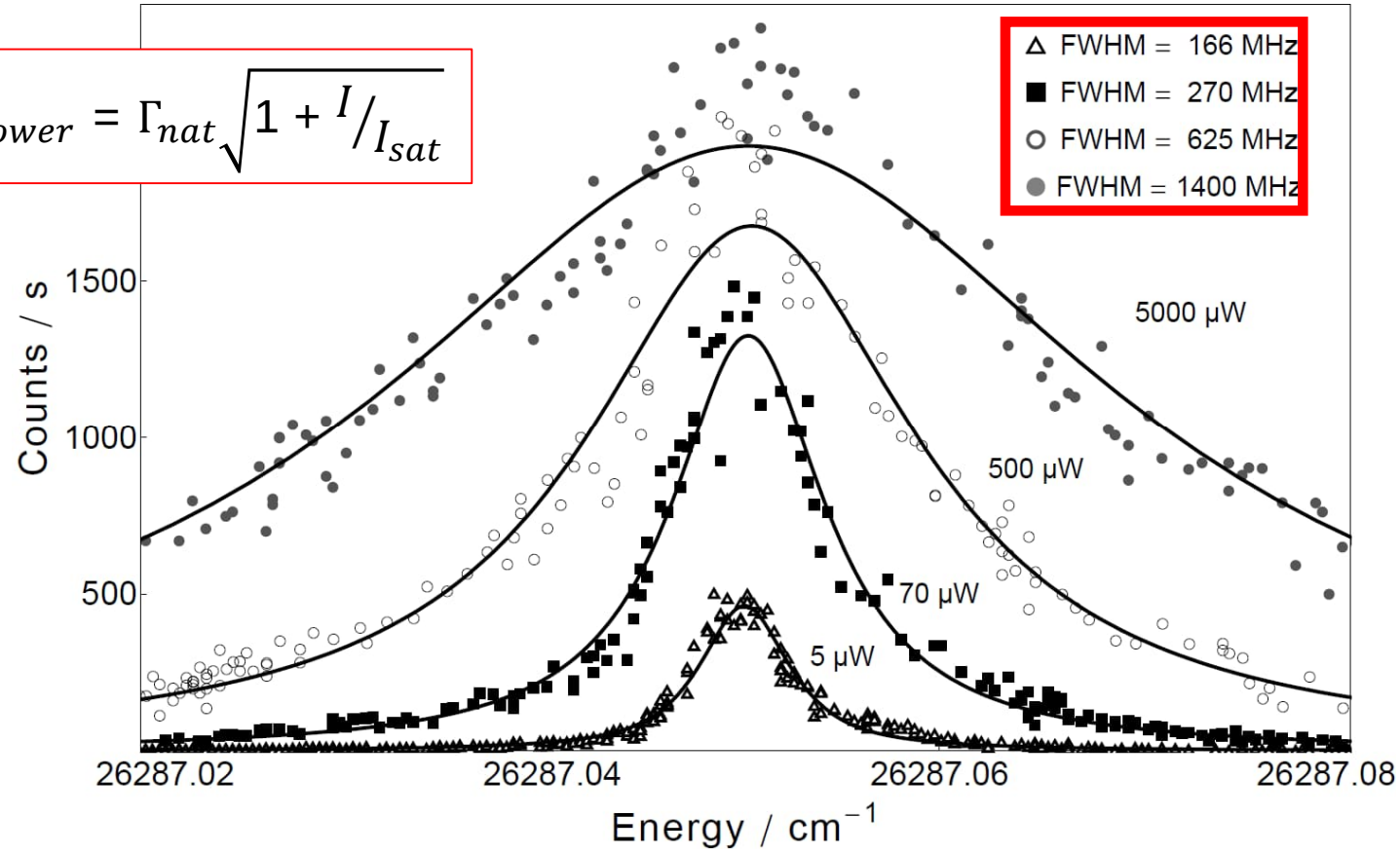
There is a trade off between using (high power) pulsed lasers for efficient production of radioactive ion beams and the spectroscopic resolution required for resonance ionization spectroscopy (in-source). Here we can directly observe the effect of spectral power broadening of atomic lines due to high power pulsed lasers.



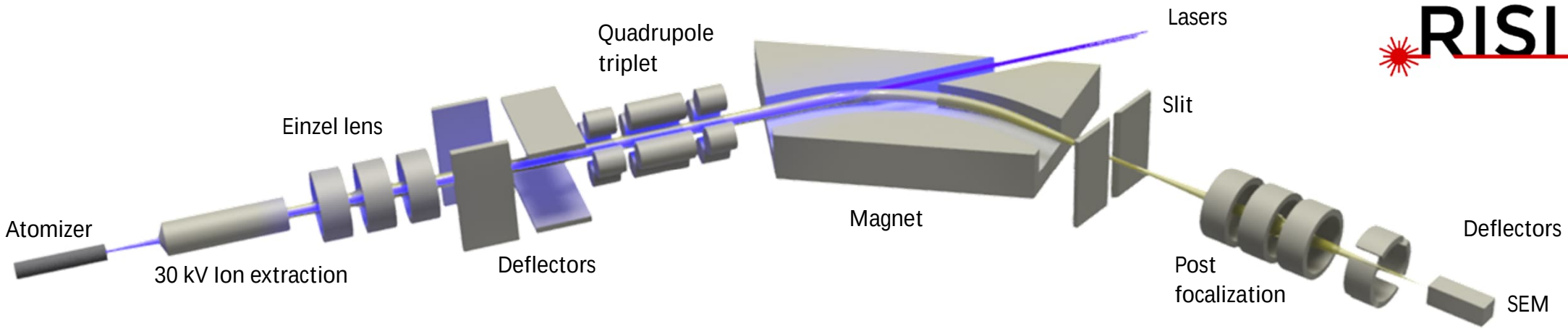
Natural linewidth:
Γ_{nat}=135 MHz

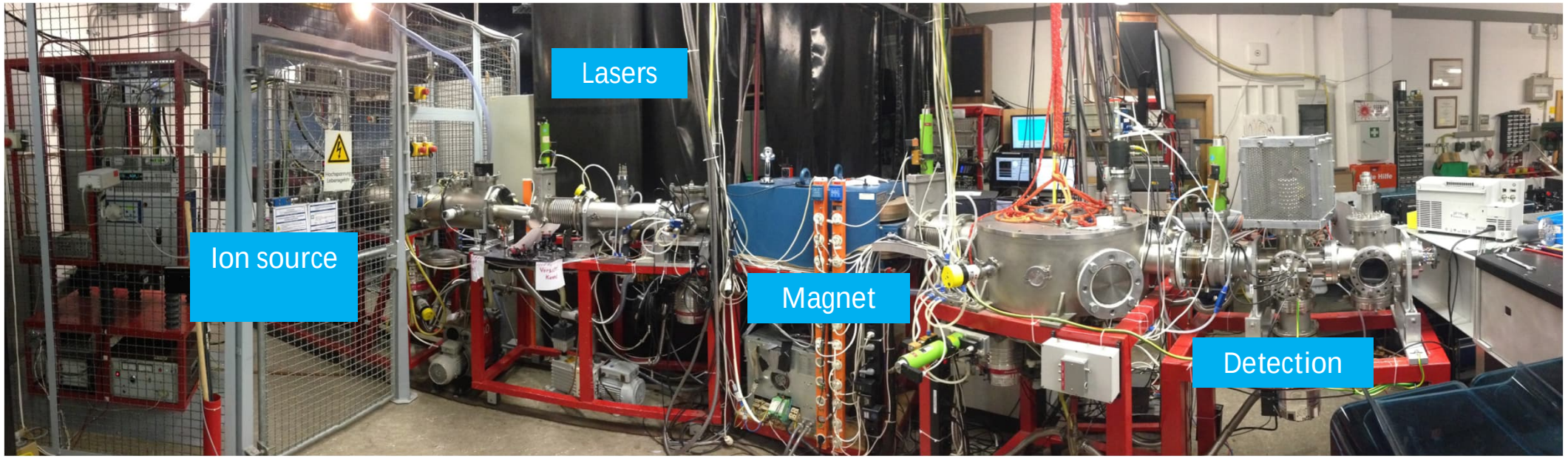
$$\Gamma_{power} = \Gamma_{nat}\sqrt{1 + {}^{I}/_{I_{sat}}}$$



V. Sonnenschein, I.D. Moore et al., EPJA 48 (2012) 52

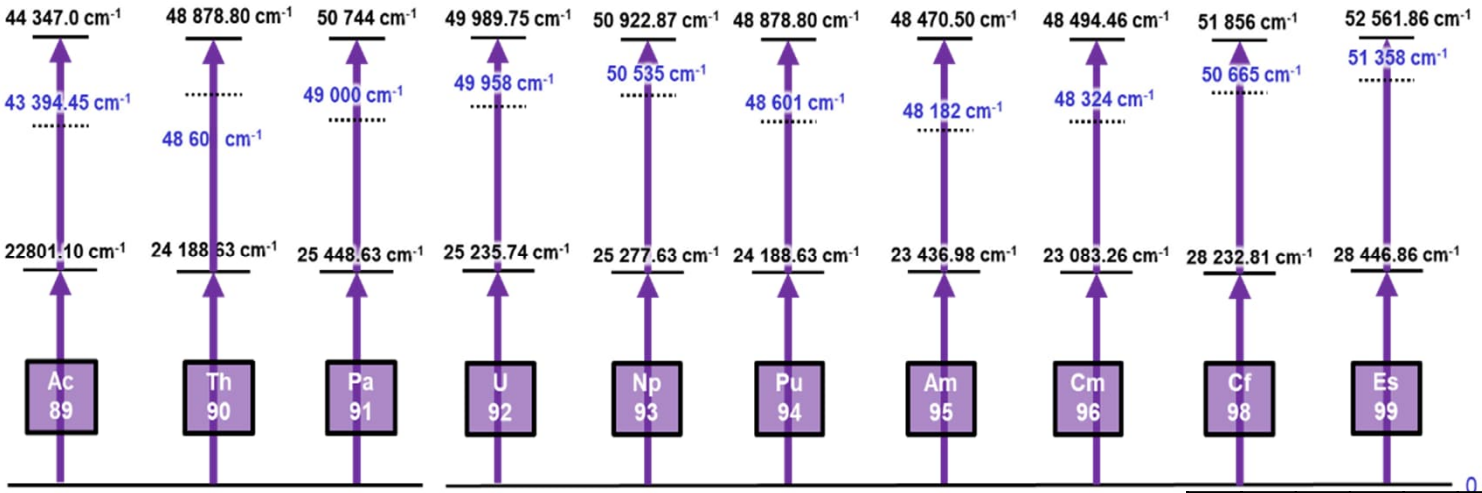
Example of an offline RIS laboratory





Courtesy S. Raeder & K. Wendt

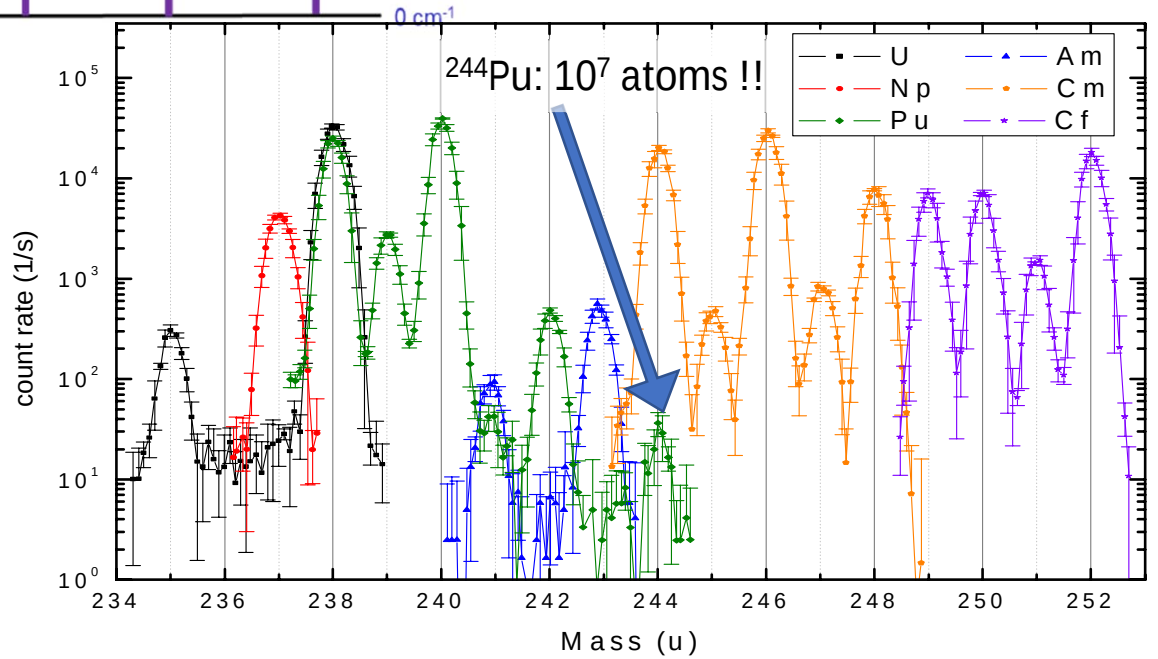
Example of versatility: multi-element analysis



- Simple & efficient two-step RIS schemes
- Combined with mass separation
- RIMS

- Rapid characterization of unknown compositions
- Isobar-free, low-background isotope ratios
- Selective laser spectroscopy in mixed samples
- For ultra-trace analysis & fundamental studies

Exclusive feed stock solution from high flux reactor breeding at ORNL (J. Etzold)



Courtesy S. Raeder & K. Wendt

Laser resonance ionization + sensitive detection

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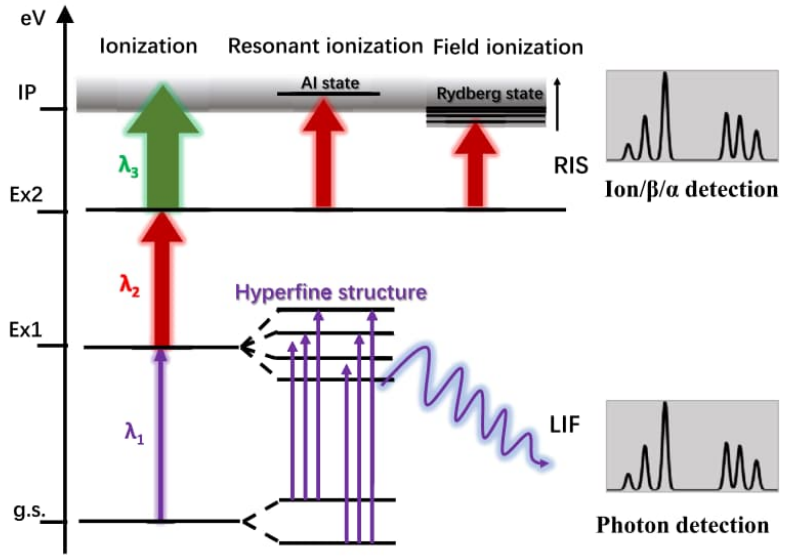
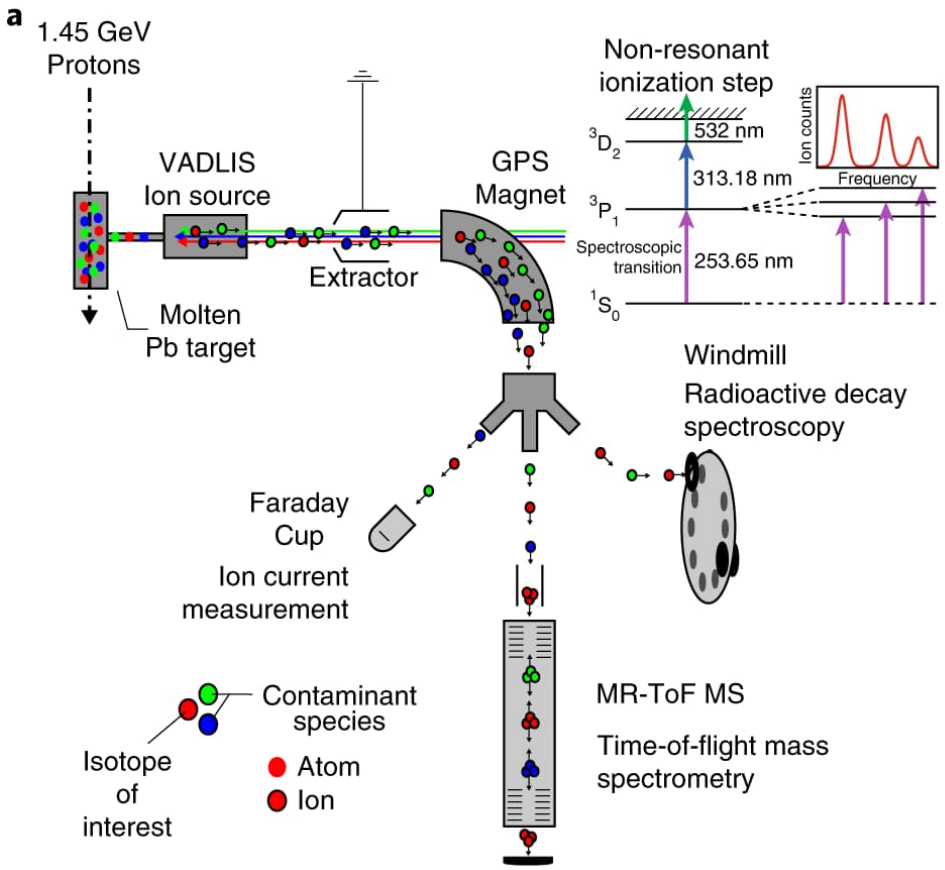
At RIB facilities, our laser-produced ions can be transported to one of several possible detection stations:

- Decay spectroscopy:
  - *tag on characteristic radiation*
- Mass spectrometry:
  - *single out one isotope from other isobars using its mass*





ISOLDE facility, CERN

B. Marsh et al., Nature Phys. 14 (2018) 1163

Provides flexibility as we will see in the next lecture ☺!

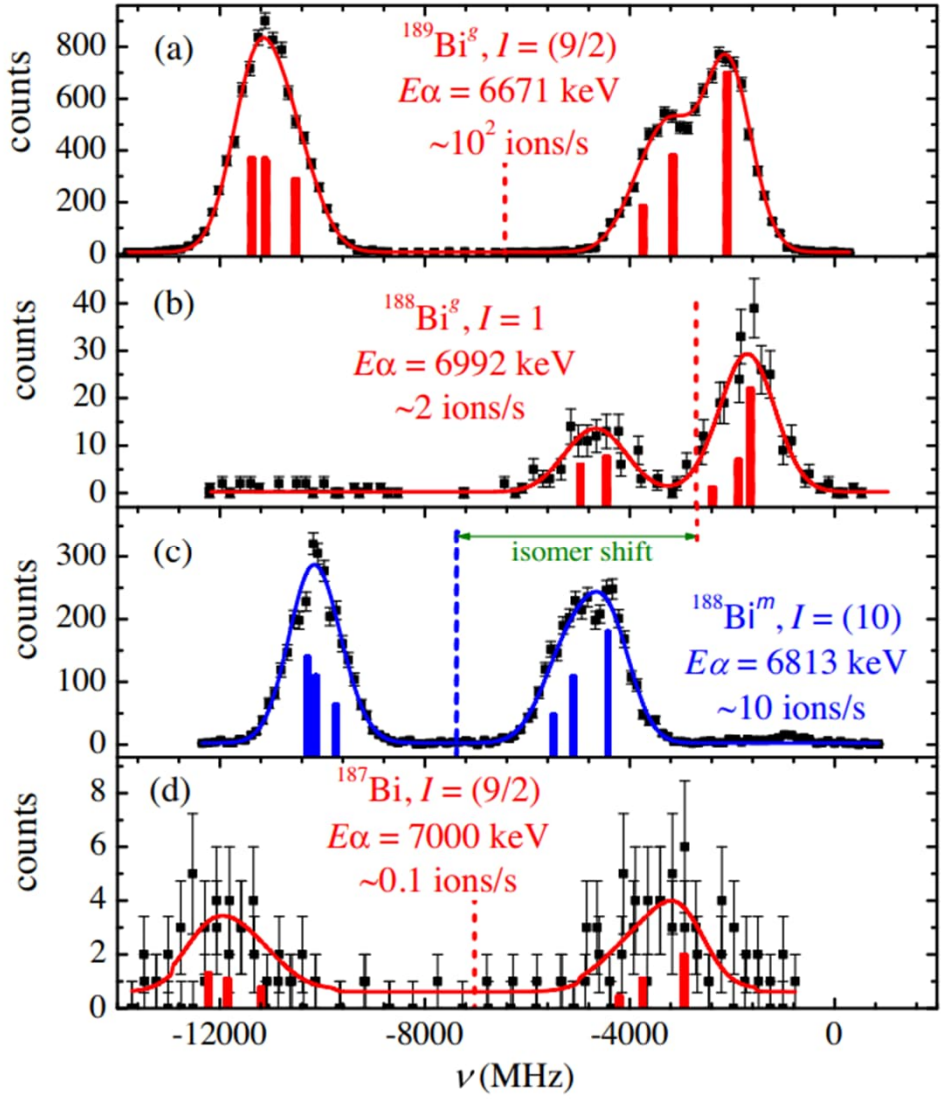
Discussion pause...

$^{189}Bi^{g}$

$^{188}Bi^{g}$

$^{188}Bi^{m}$

^{187}Bi



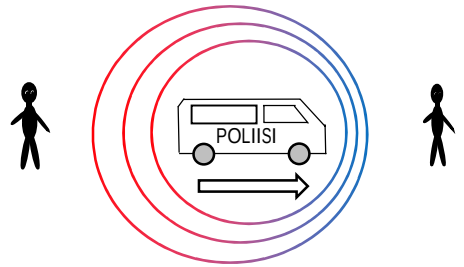
- Examples of in-source RIS at ISOLDE.
- Yields measured with alpha-decay spectroscopy, down to the level of < 1 ion/s !

Given we are using the ISOL method of RIB production combined with RIS, what do you think are the limitations in this method?

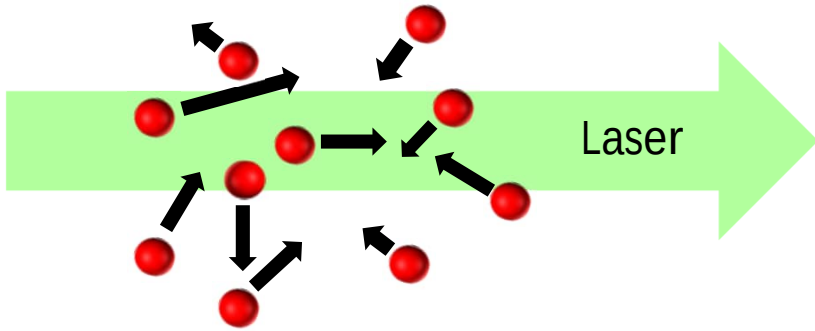
A. Barzakh et al., Phys. Rev. Lett. 127 (2021) 192501

The drawback of the in-source RIS approach

The observed transition linewidth can be broadened by Doppler effects due to the high temperatures involved (hot cavity), or collisions (if in gas):



Doppler shift $f' = f_0(1 \pm \frac{v}{c})$



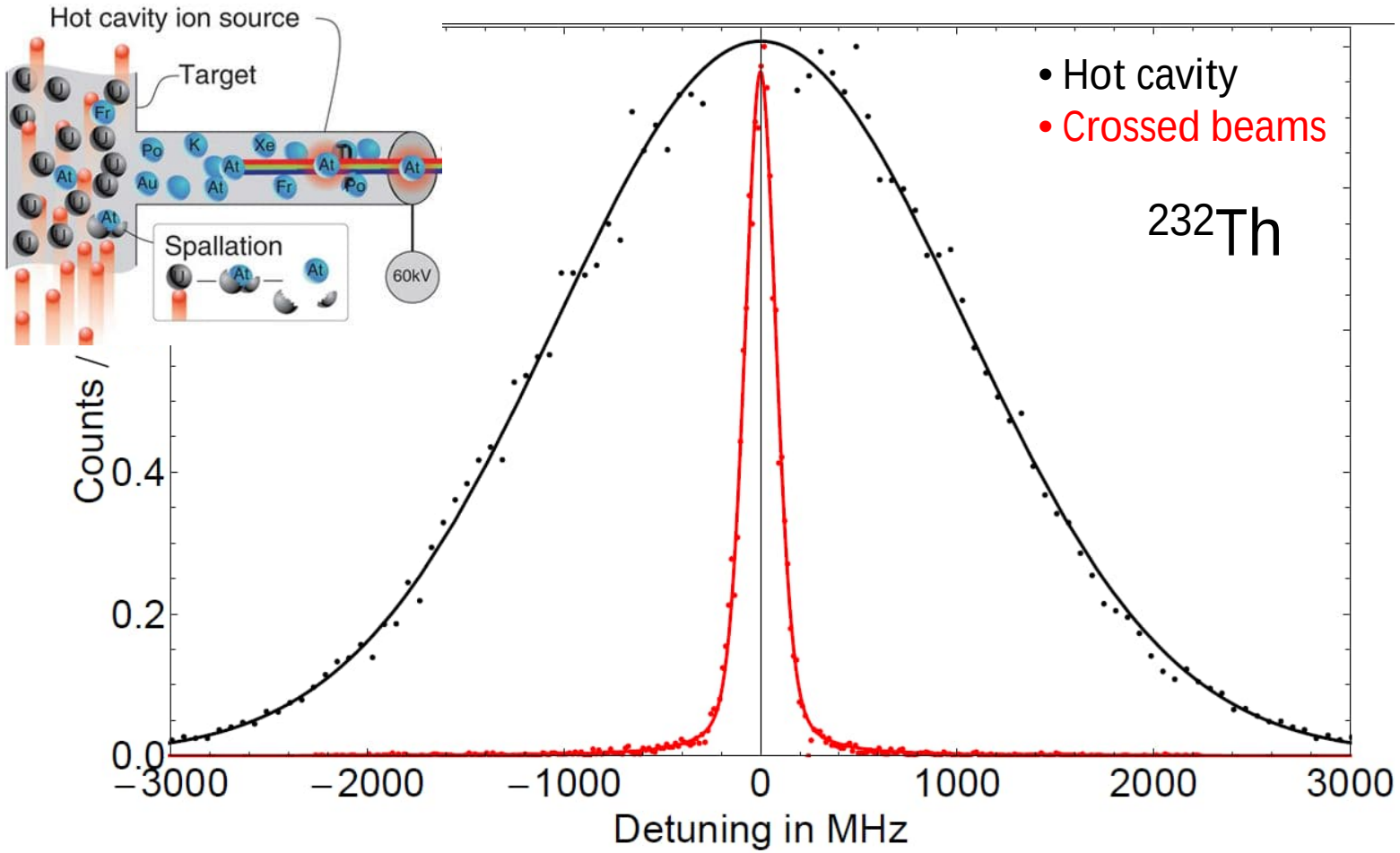
Doppler broadening

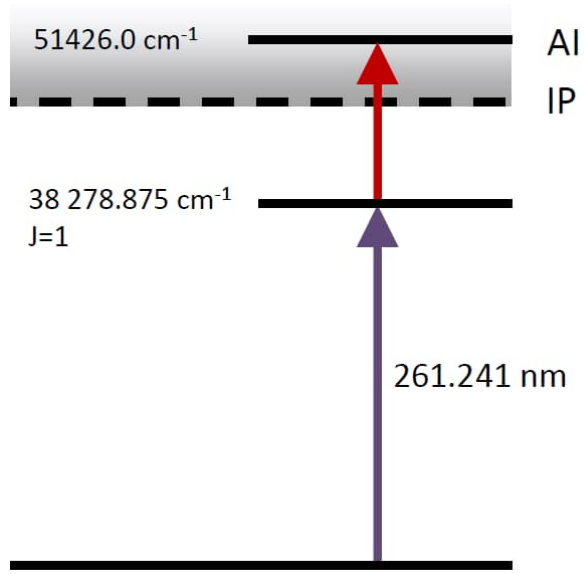
Thermal motion is a Maxwell-Boltzmann probability distribution. Causes a spread of frequencies observed by atoms:

$$P(f)df \propto \exp(-\frac{mc^2(f-f_0)^2}{2k_bTf_0^2})df$$

$$\Delta_{FWHM} = f_0\sqrt{\frac{8k_bT\ln 2}{mc^2}}$$

Doppler broadening from a hot oven



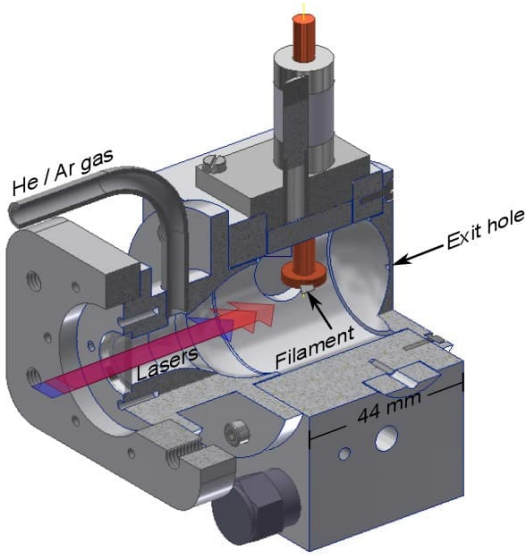


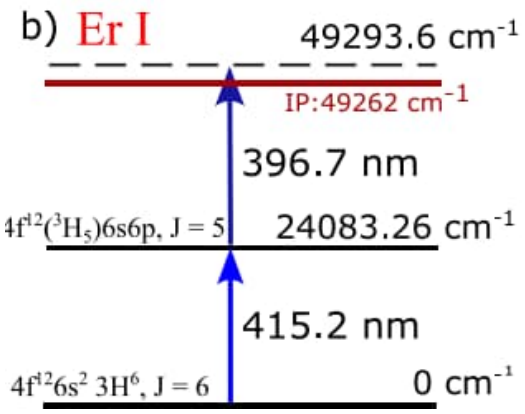
``Low´´ resolution allows us to measure:

- Hyperfine A factor → magnetic moments
- Isotope shift → charge radii

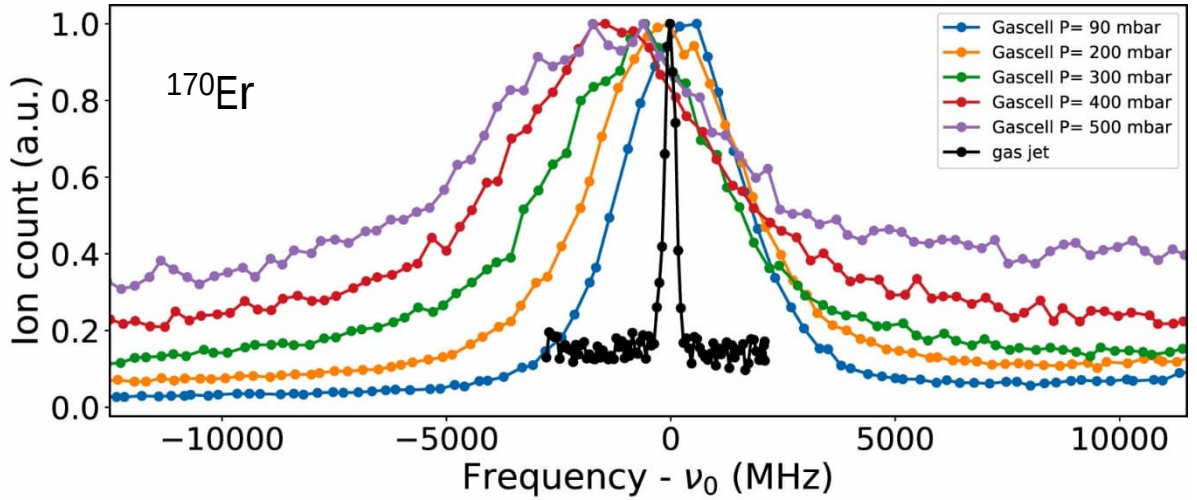
Natural linewidth 35 MHz; spectral linewidth 2.4 GHz (in oven), 170 MHz (crossed-beams configuration).
The Doppler broadening is often comparable or greater than HFS or IS!

Collisional broadening in a gas cell



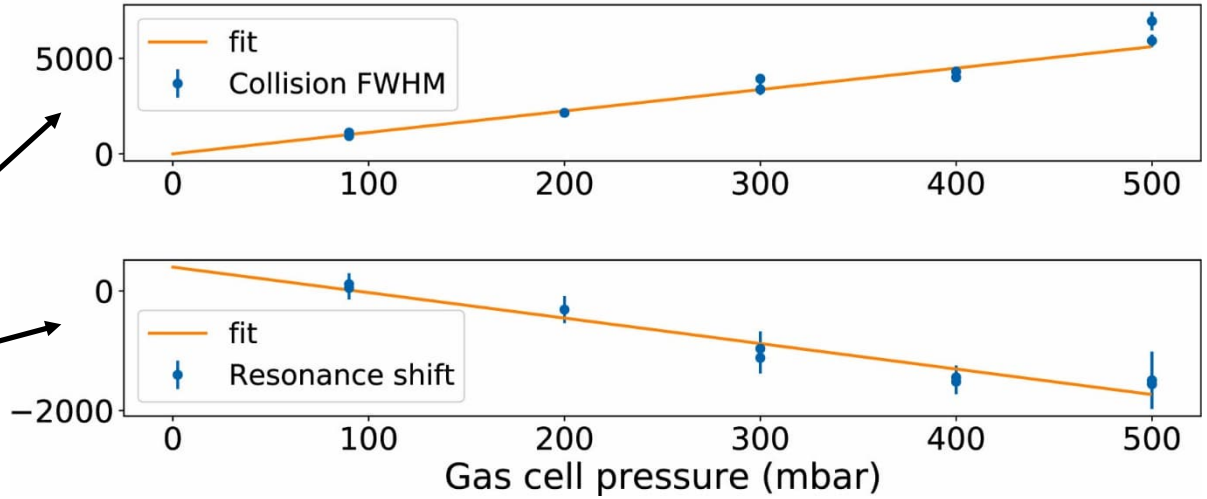


Frequency scan of the first step transition



- In a gas cell, atomic spectral lines ``suffer´´ from collisions with buffer gas atoms. Many collisions perturb the energy levels
- Leads to spectral line broadening & energy shifts

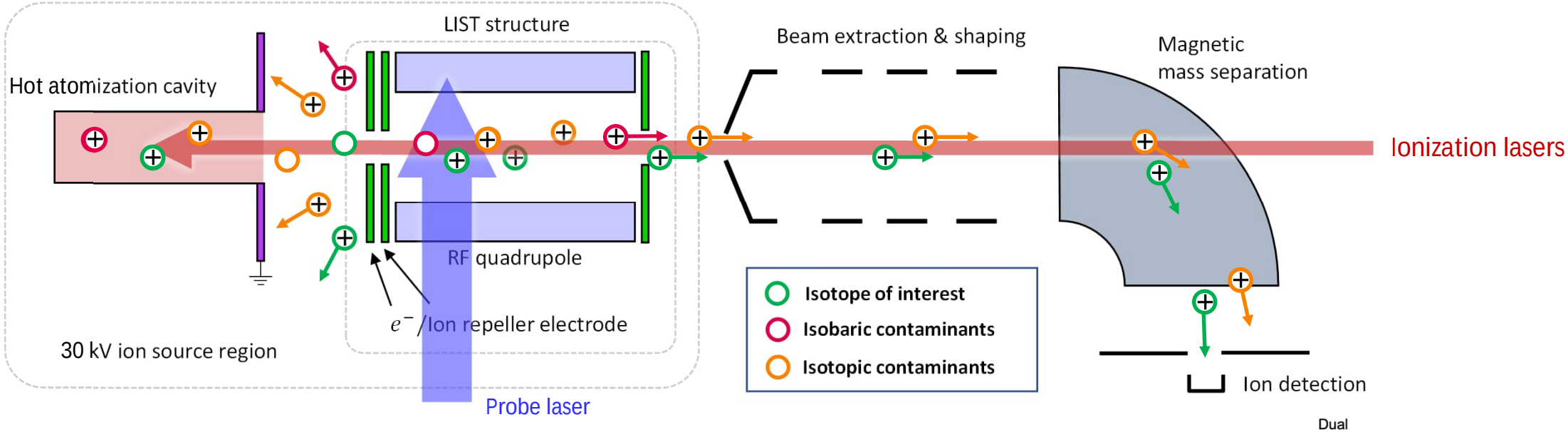
- Resonance peak FWHM, Γ_{coll} = 11(1) MHz/mbar
- Resonance peak centroid, Γ_{shift} = -4(1) MHz/mbar

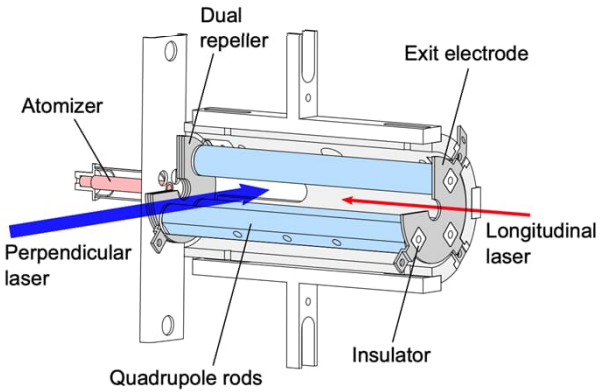


A. Ajayakumar et al., NIMB 539 (2023) 102

Methods used to mitigate the broadening

A variation to in-source RIS recently applied online at ISOLDE, CERN. Based on developments in U-Mainz.





Crossed atom beam / laser geometry in LIST structure:

- Electrodes provide a full suppression of surface ionized isobars
- Probe a reduced Doppler ensemble of atoms
- Suitable narrow-band laser must be used

T. Kron et al., PRC 102 (2020) 034307
R. Heinke et al., Hyp. Int. 238 (2017) 6

Perpendicularly-Illuminated Laser Ion Source

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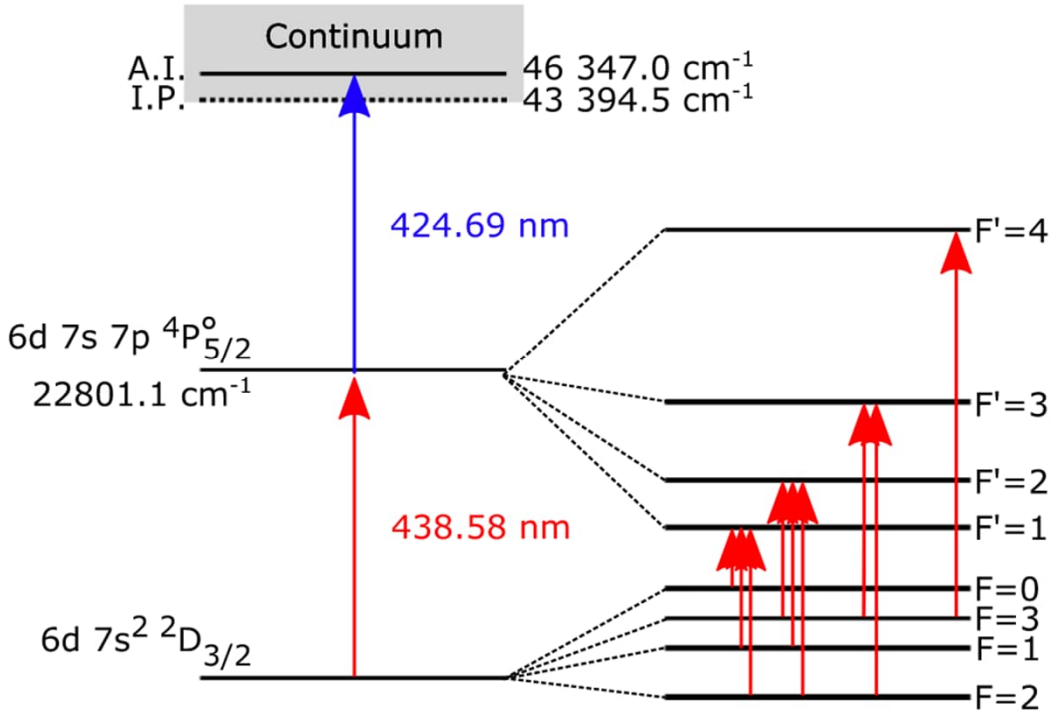
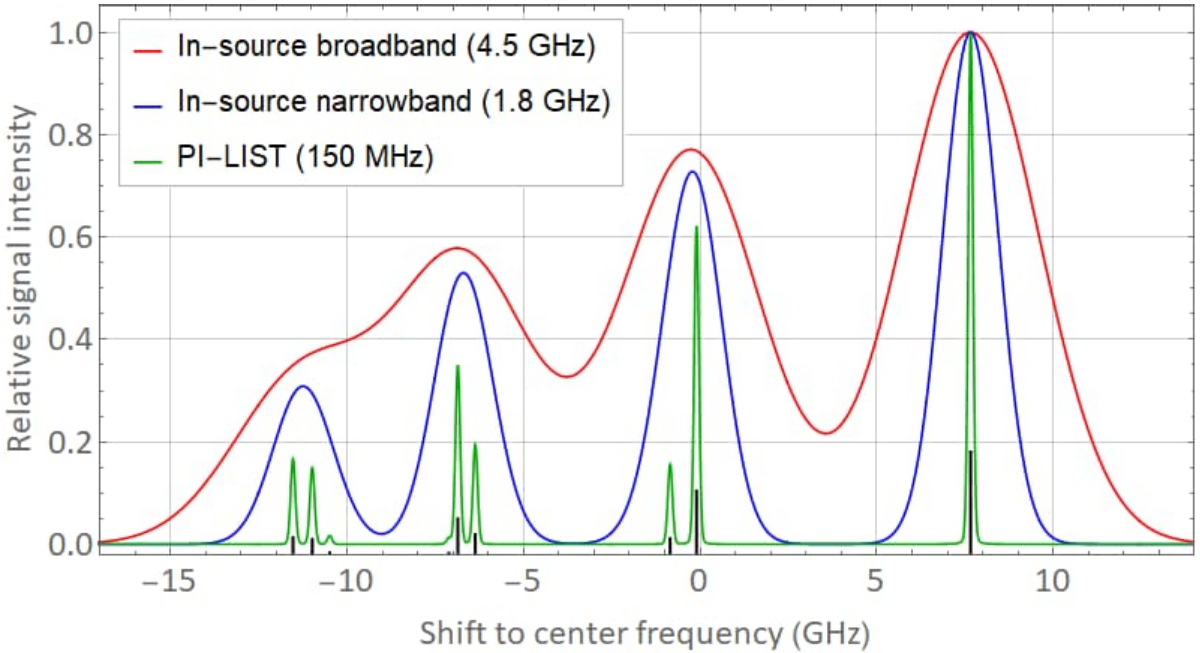
Ac laser ionization scheme

(used at several online facilities – ISOL & gas cell)

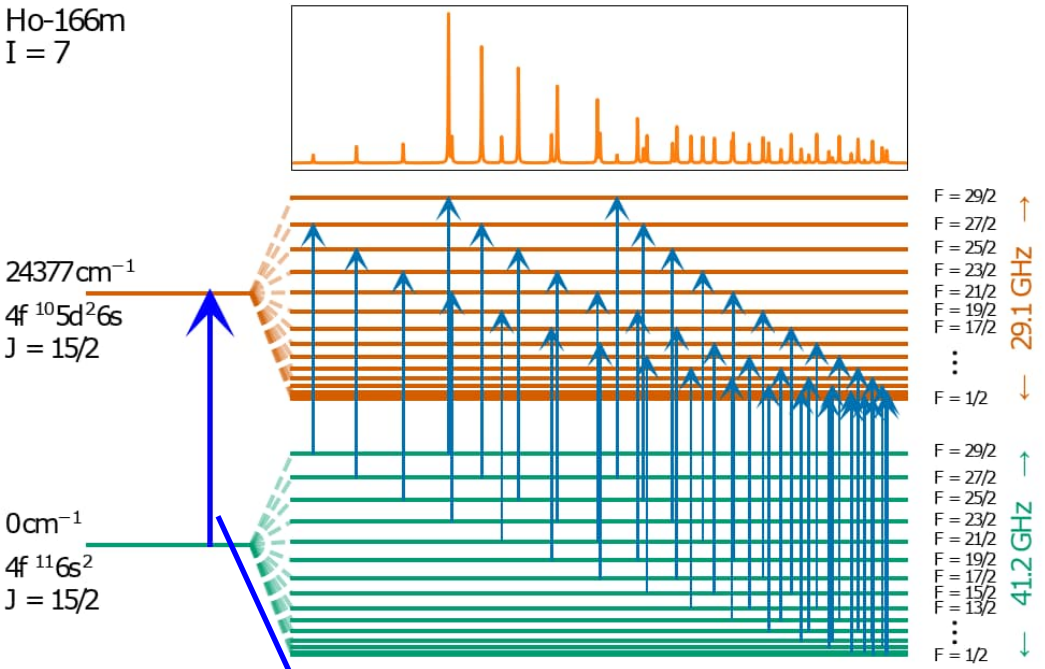


``High´´ resolution allows us to measure hyperfine B factor → Q moment



- Strong reduction in Doppler broadening seen
- Sensitivity <1 ion/s; resolution ~200 MHz
- Sacrifice in efficiency (10^2 to 10^4 compared to standard in-source resonance laser ionization, ~100 to ``LIST´´)

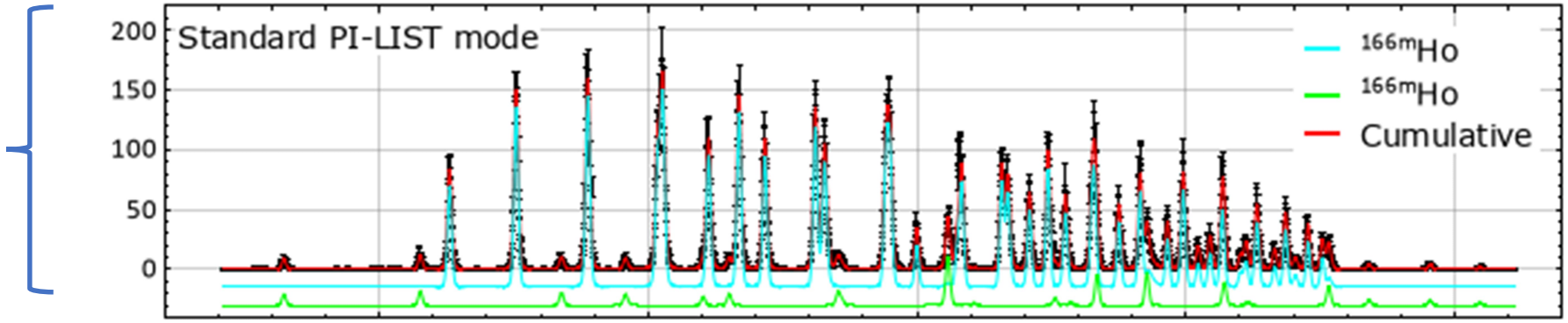
Offline results from PI-LIST at U-Mainz



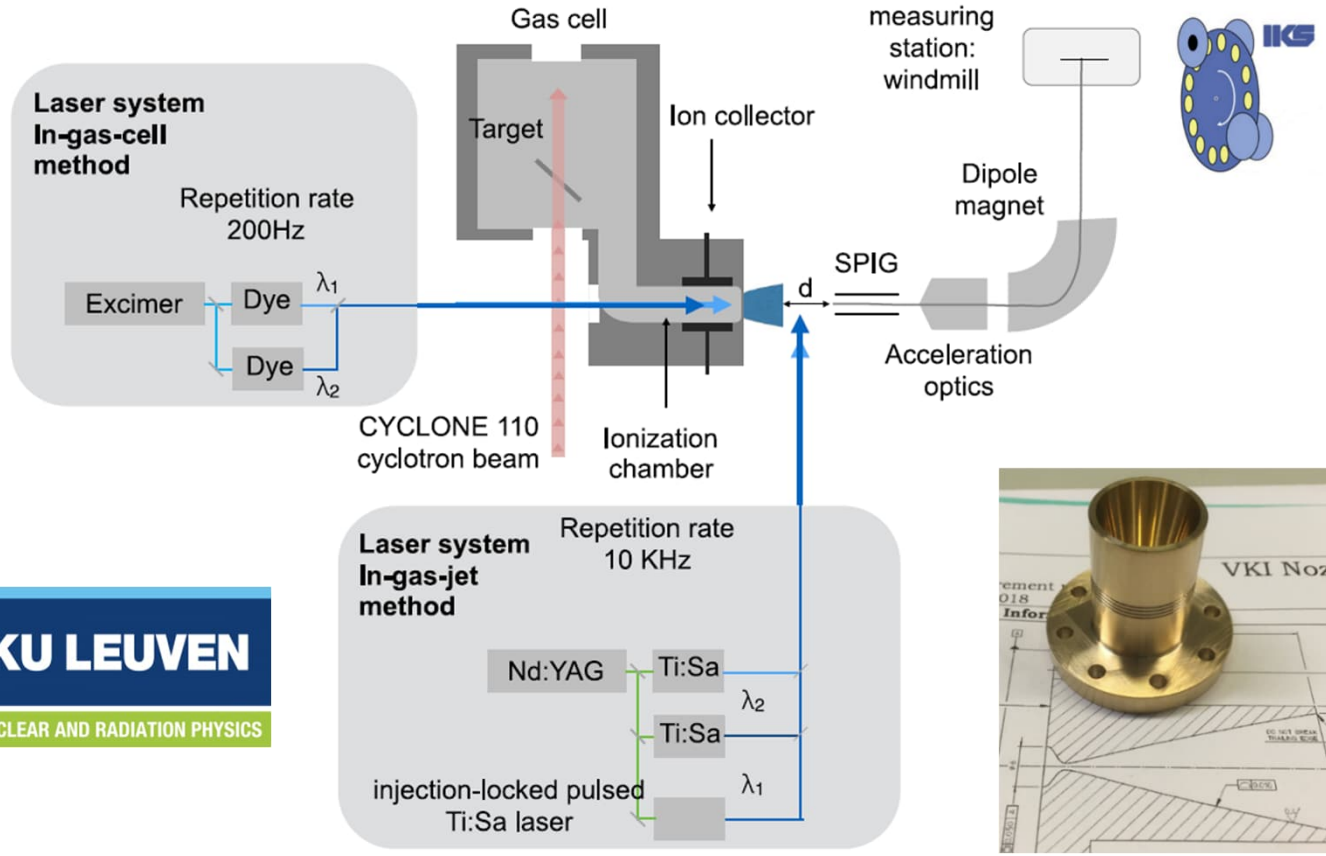
A cautionary note on high angular momenta!

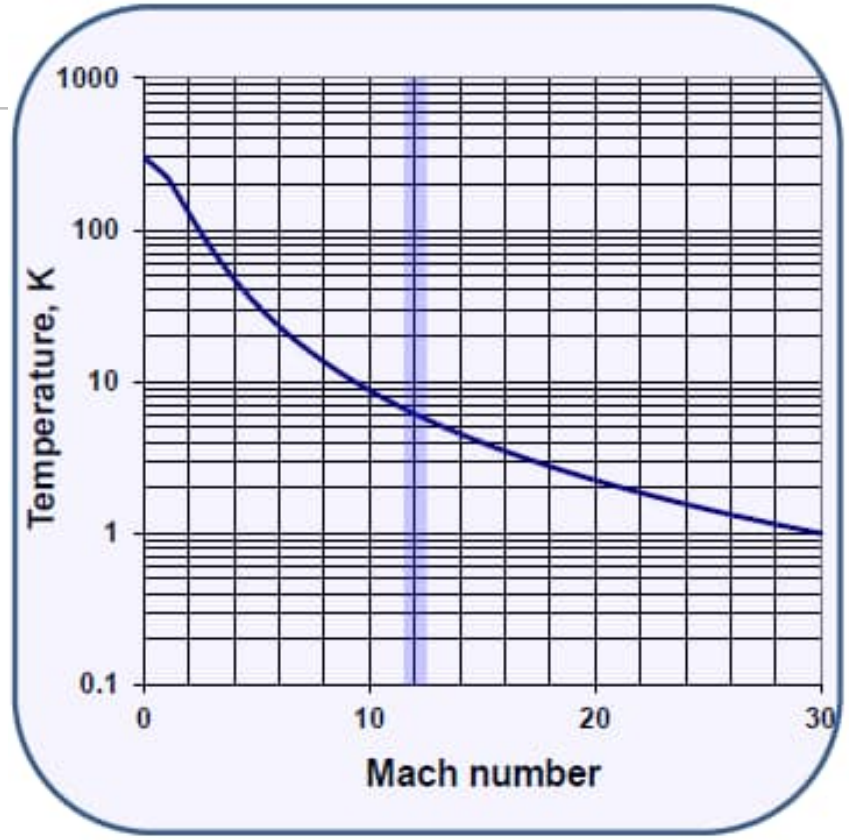
- A transition involving high angular momenta is good for sensitivity to spins and moments
- In this example, altogether 43 resonances are present!!
- But it comes at a cost! Signal strength is spread over many peaks.
- Even with super high production rates, these kinds of measurements on radioactive isotopes are simply not feasible.

R. Heinke, PhD thesis, University of Mainz (2019)



From the gas cell to the gas jet

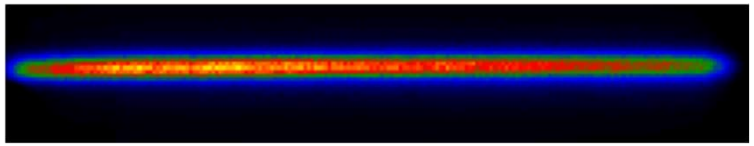




M= 12, gas jet temp T=6K

- Specially designed (Laval) nozzles
- After the nozzle the gas expands and goes supersonic.
- The Mach number *M* is the ratio of the stream velocity *u* to the local speed of sound *a*.
- Temperature drops, gas density drops.

Gas jet studies:



Yu. Kudryavtsev et al., NIMB 297 (2013) 7

A. Zadvornaya et al., PRX 8 (2018) 041008

A (final) pioneering experiment at the LISOL facility

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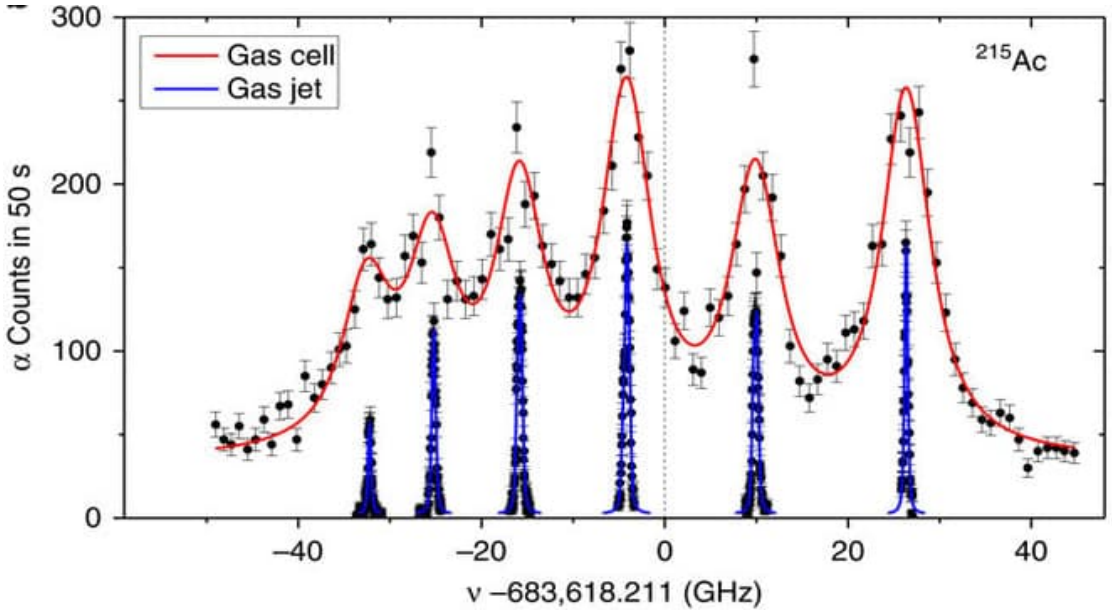
ARTICLE

Received 25 Aug 2016 | Accepted 9 Jan 2017 | Published 22 Feb 2017 DOI: 10.1038/ncomms14520 OPEN

Towards high-resolution laser ionization spectroscopy of the heaviest elements in supersonic gas jet expansion

^{215}Ac ($\tau_{1/2}$=170 ms)



- in gas cell

Resolution: 5800(300) MHz

- in gas jet

Resolution: ~400 MHz

*1/5/1974 - †6/12/2014

Celebrating the 40th anniversary of LISOL (Louvain-la-Neuve)

R. Ferrer et al., Nat. Comm. 8 (2017) 14520

Gas-jet exploited for heavy element studies at GSI

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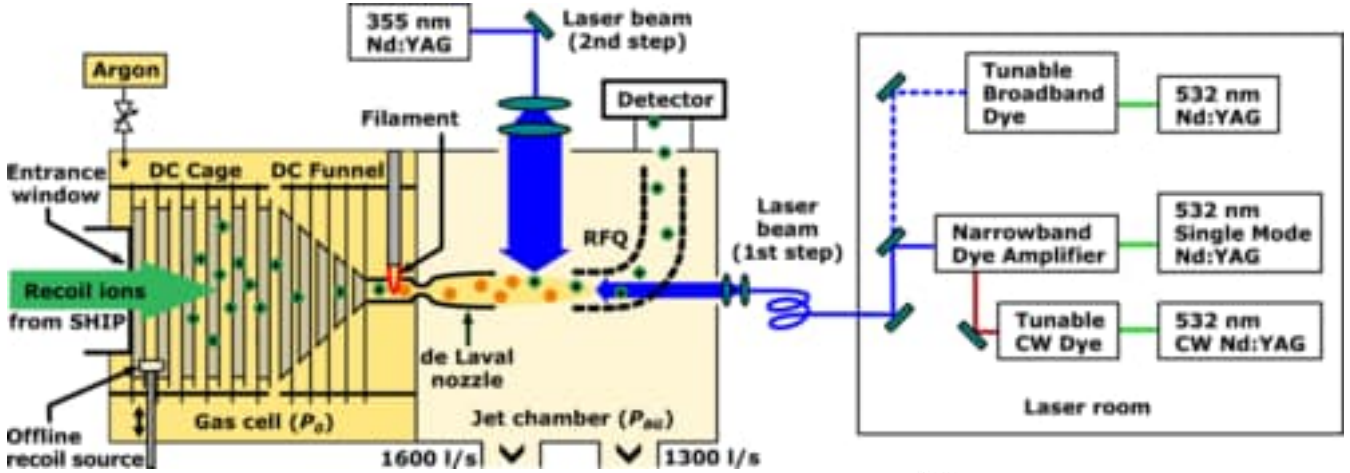
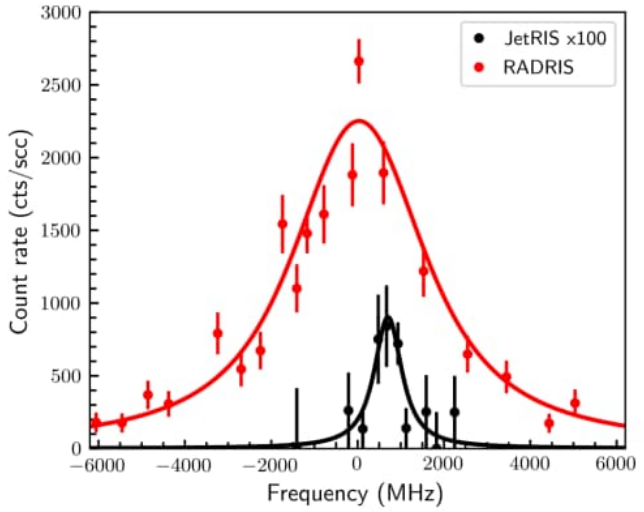
In-gas-jet laser spectroscopy of ^{254}No using the JetRIS apparatus at GSI, Germany.

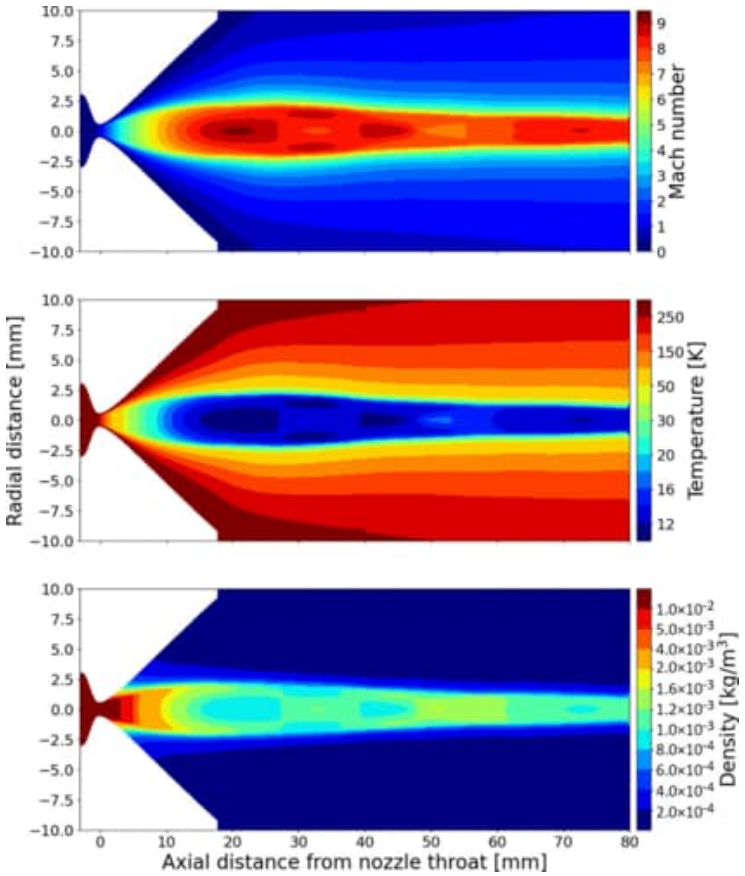
$^{208}Pb(^{48}Ca, 2n)^{254}No$



- First successful heavy actinide gas-jet study
- 4 ions/s of ^{254}No at entrance to gas cell
- Overall efficiency 0.010(3) % (2 oom < RADRIS)
- 5-fold improvement in resolution over in-gas cell spectroscopy.



2D plots of Mach number, T, and density distributions.



J. Lantis et al., Phys. Rev. Res. 6 (2024) 023318.

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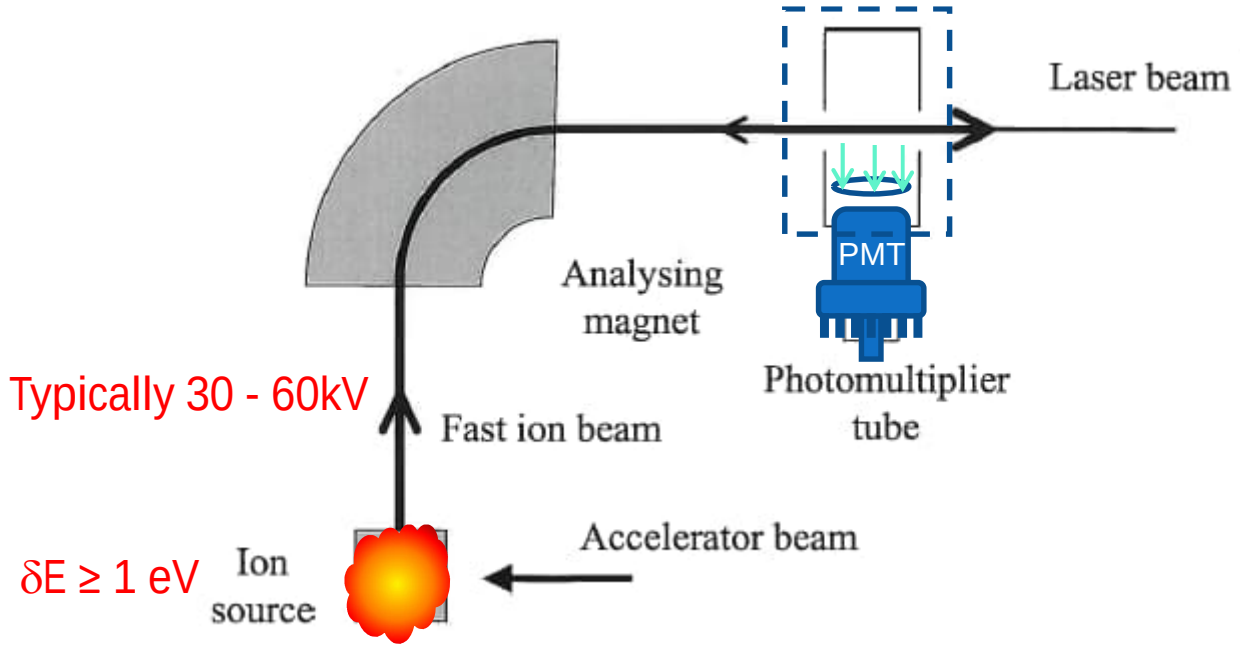
Collinear laser spectroscopy

(Fast beams) collinear laser spectroscopy

In a collinear geometry, light, whether co- or counter propagating with the ion beam, interacts with accelerated ionic ensembles.



$$\delta\nu_D = \nu_0 \frac{v}{c}$$ Doppler broadening (in frequency space)

1. Accelerate all ions to energy E

$$E = eV = \tfrac{1}{2}mv^2$$

2. The energy spread δE (from source) remains constant

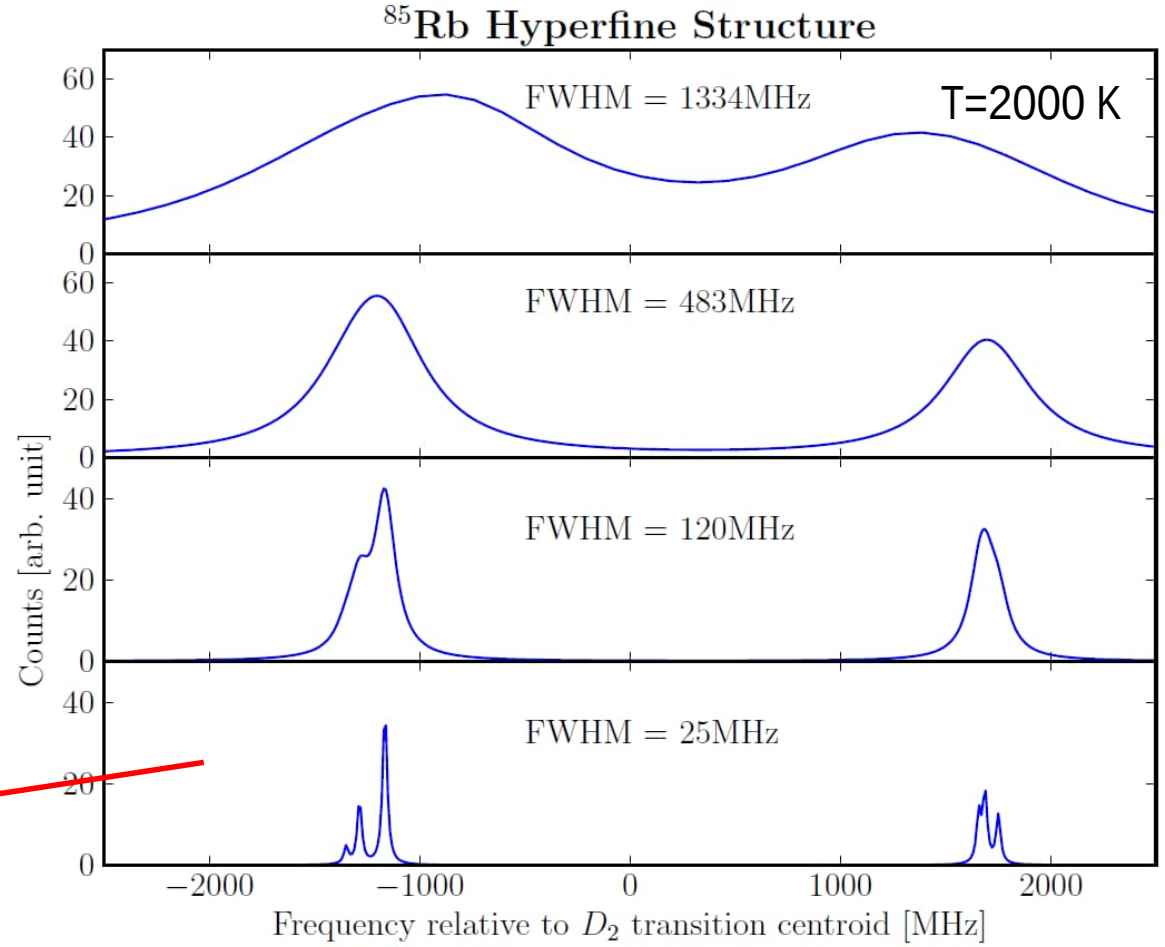
$$\delta E = \delta\left(\frac{mv^2}{2}\right) = mv\delta v = const.$$

3. The corresponding velocity spread is decreased. We obtain the Doppler width (in frequency):

$$\delta\nu_D = \nu_0 \frac{\delta E}{\sqrt{2eVmc^2}}$$

S.L. Kaufmann, Opt. Comm. ***17*** *(1976) 309* *W.H. Wing et al., PRL* ***36*** *(1976) 1488*

The effect of the velocity compression

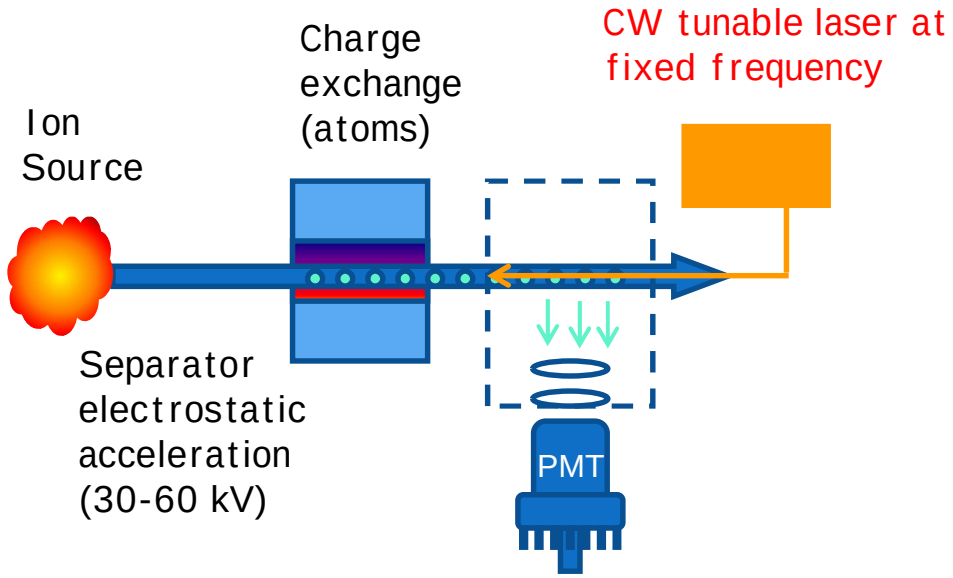


reduction in Doppler broadening

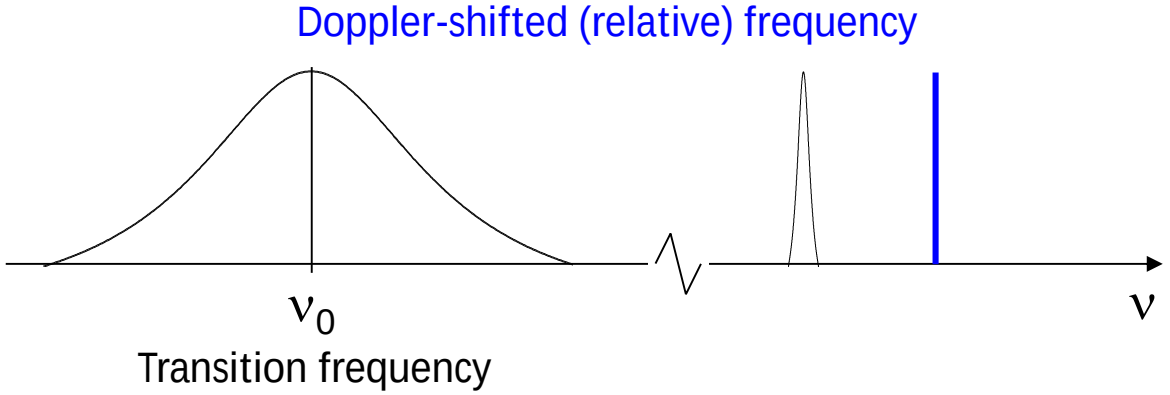
Natural linewidth achieved!

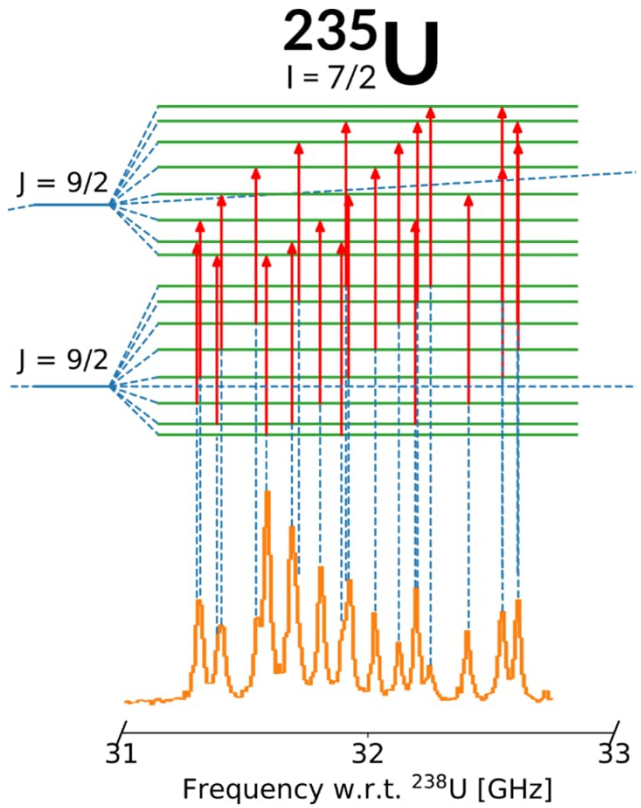
Typical ion source energy spreads are ~1 eV. Acceleration of medium-mass nuclei to 30 keV produces a 3 order of magnitude velocity compression. Unresolved hyperfine structure becomes visible!

Doppler tuning the ion beam



Apply ``Doppler tuning´´ voltage

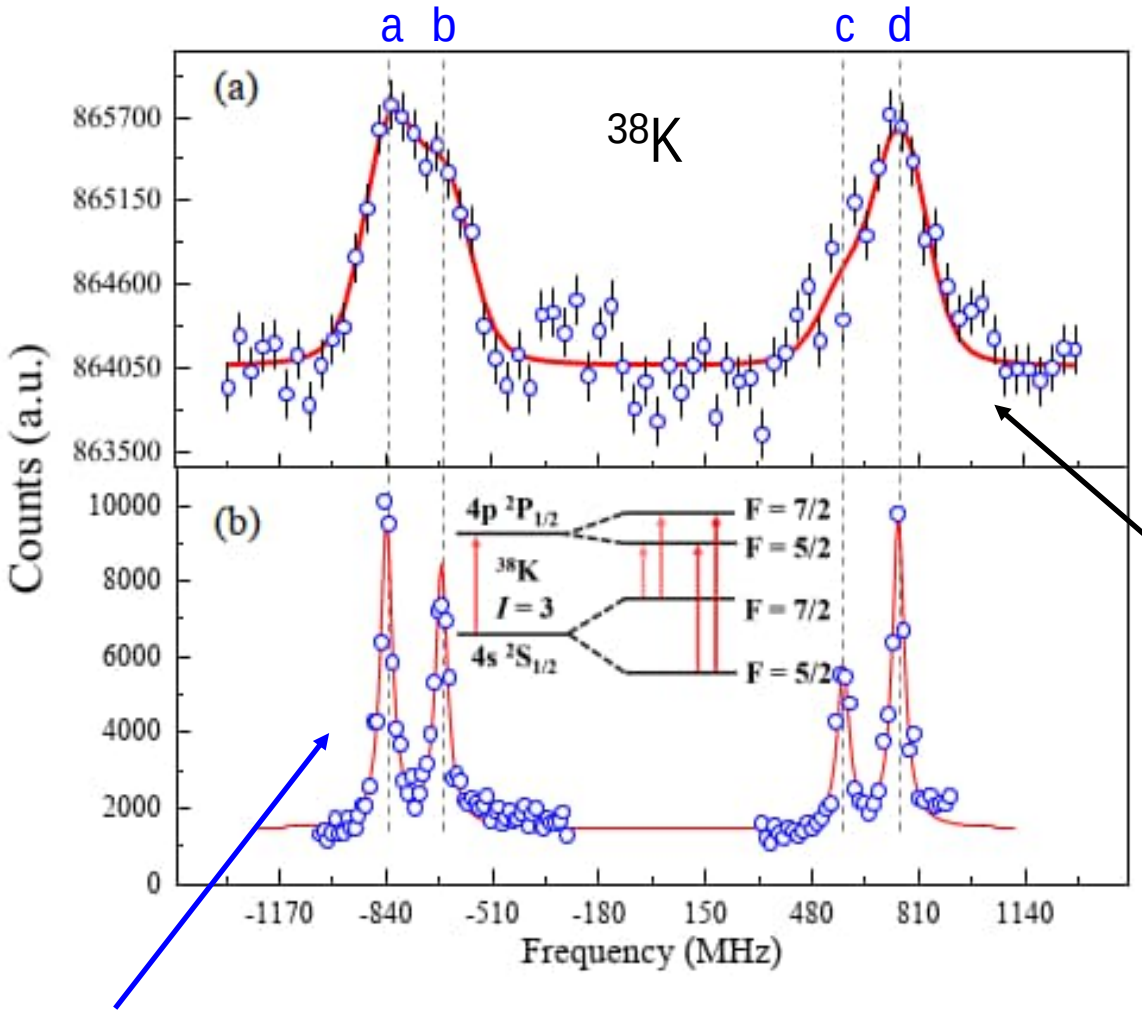




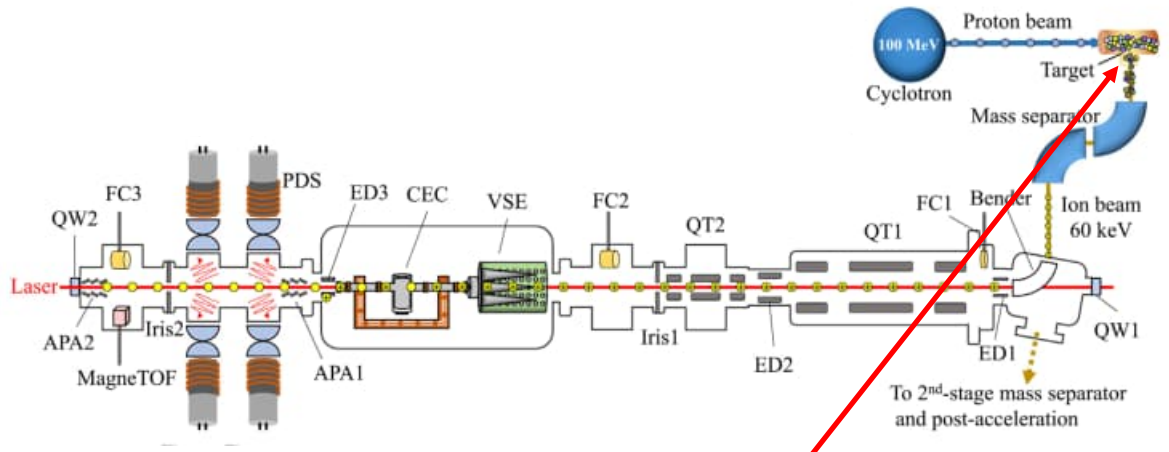
$$\nu_{\pm} = \nu_0 \frac{1 \pm \beta}{\sqrt{1 \mp \beta^2}}, \beta = \frac{v}{c}$$

(Exact collinear ``+´´; exact anticollinear ``-´´)

Illustrating the effect of the (source) energy spread



Clear impact with a reduced energy spread (CRIS@ISOLDE)



- Ion source energy spread ~20 eV from the Beijing (ISOL) RIB facility.
- Leads to a Doppler "imprint" of ~125 MHz on the spectral transition(s).

$$\Gamma_D = \nu_0 \delta E / \sqrt{2Emc^2}$$

Note the difference in the background!!

S.J. Wang et al., NIMA 1032 (2022) 166622

Sometimes we need to work with fast atom beams

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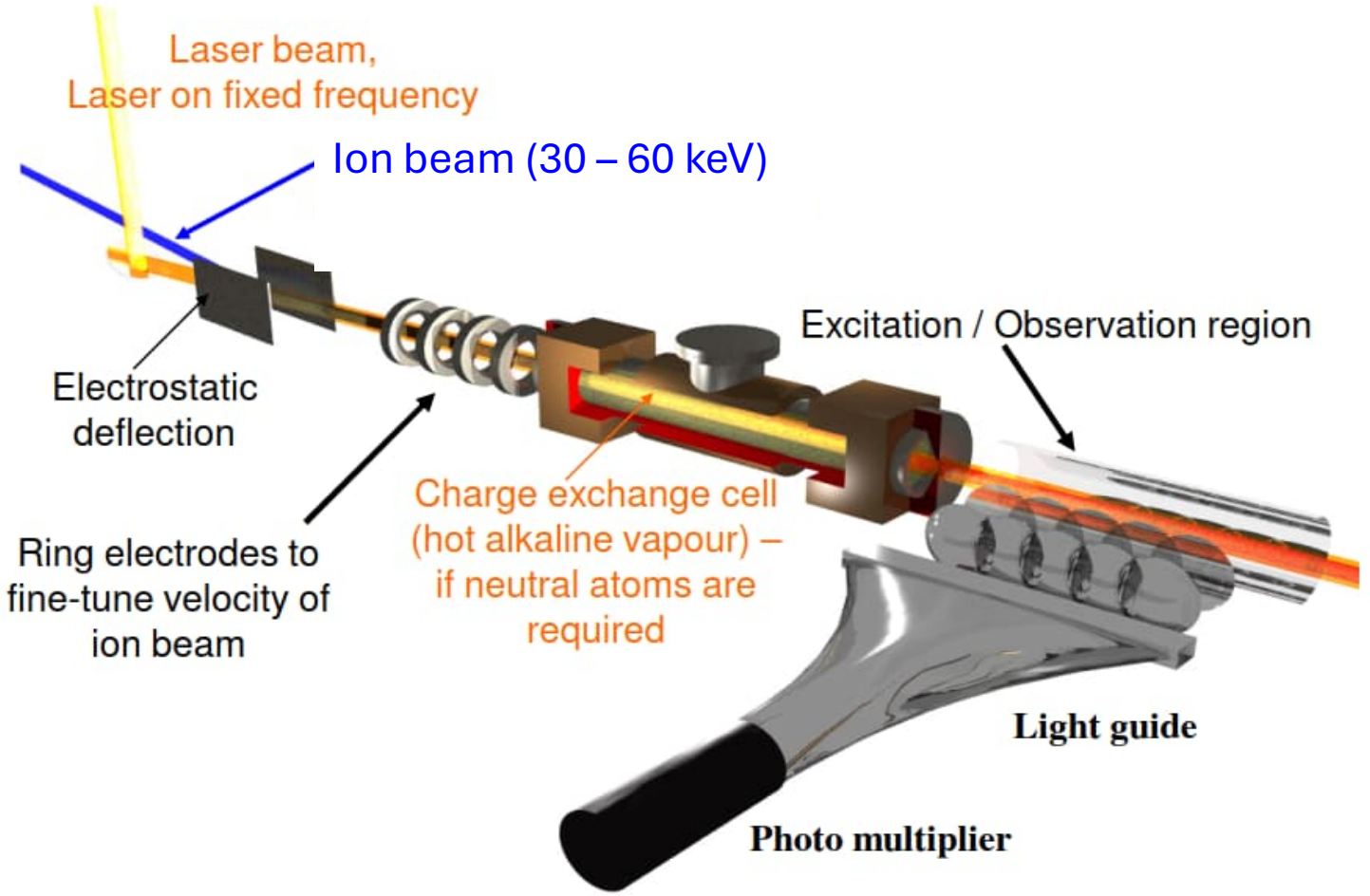
- Transitions from the ionic ground state might be in the UV or VUV – so are not directly accessible for our laser systems.
- Alkaline metal ions, ions of rare gases, most non-metal ions and many metals.
- Pass the ion beam thrugh a charge-exchange cell (CEC) with low pressure alkaline vapor. Sodium or potassium are most commonly used.
- Doppler tuning performed by applying voltage to the CEC.



Recall the example of Pd (from King plot discussion)

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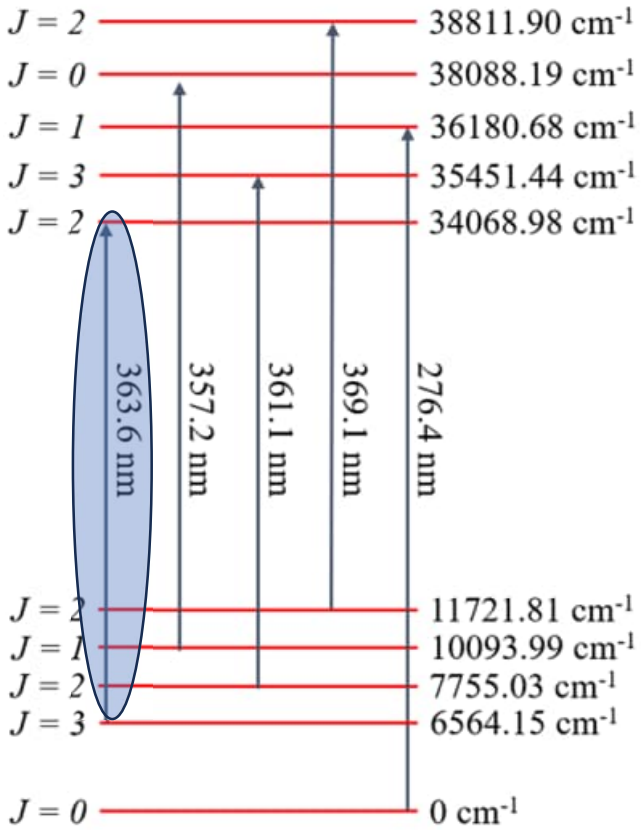
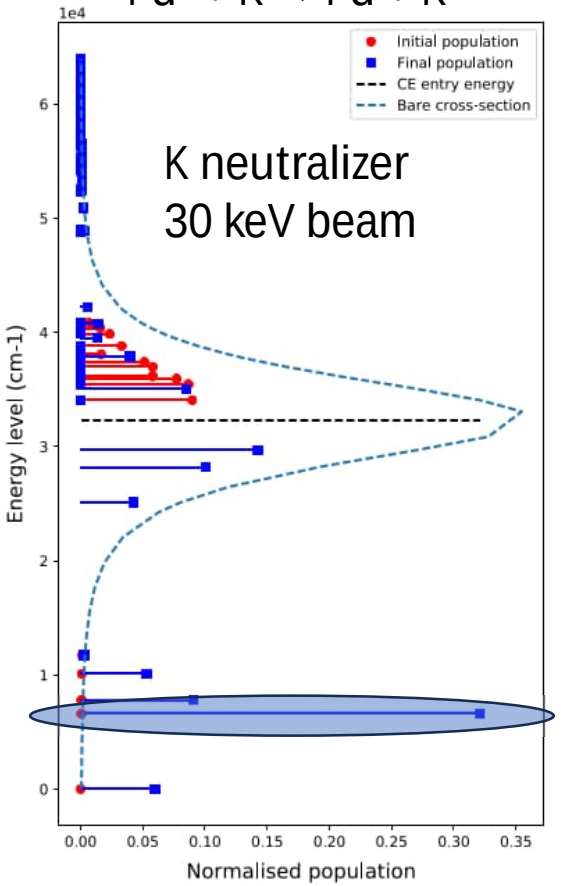
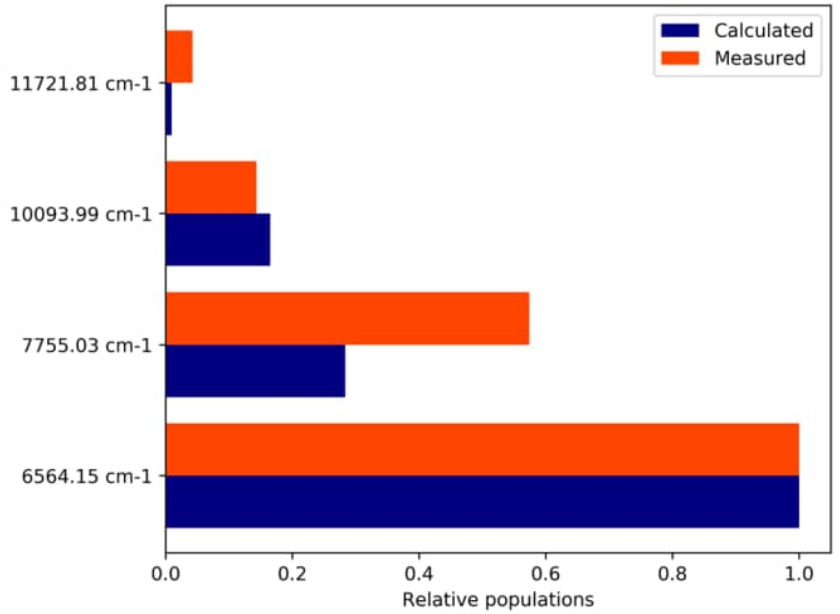
- Strong transitions in Pd^+ ion are inaccessible to standard CW laser systems (very short wavelengths).



$Pd^+ + K \rightarrow Pd + K^+$





- Total neutralization eff. ~40%
- The majority of the population goes into the 6564.15 cm^{-1} level.
- 363.6-nm transition: ~1 photon per 7000 ions.

A.R. Vernon et al., Spectrochim. Acta B 153 (2019) 61

S. Geldhof et al., Hyp. Int. 241 (2020) 41.

Discussion pause...

In the previous slide, I stated that our measured ``spectroscopic efficiency´´ using the 363 nm transition of Pd was 1 photon detected per 7000 ions. This can be thought of as the combined excitation and detection efficiency.

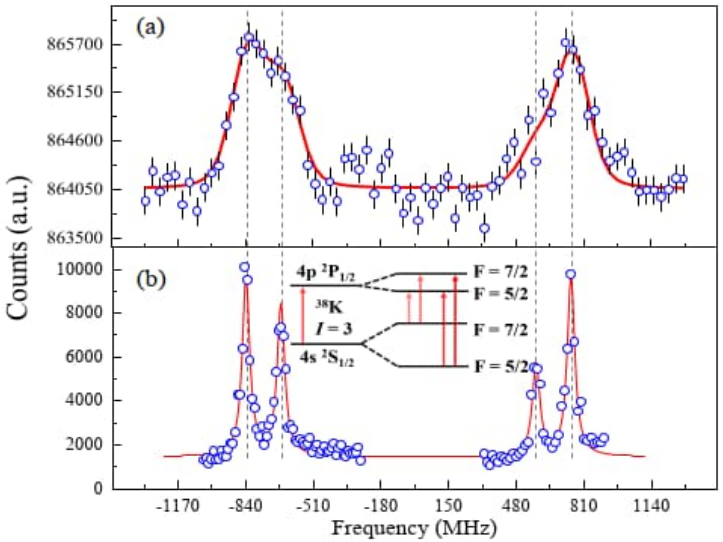
Any thoughts on why we don't measure 1 photon per 1 incoming ion?

Fluorescence light collected by optics and focused to a PMT detector. Solid angle coverage of the light collection system and quantum efficiency of detectors constitute the main limitations of this method.

Counting rates for low flux ion beams

The collinear beams technique has high sensitivity. All ions/atoms pass through the laser beam and contribute to the fluorescent signal.

However……here are some typical numbers:



Signal (laser on resonance) =
1 photon detected per 1,000 ions in beam

Background (laser light scatter) = 200 photons / sec
(per mW of laser light)

Low-flux beams (1,000 ions / sec): background must be suppressed to see signal.

Addressing the scattered background (old method)

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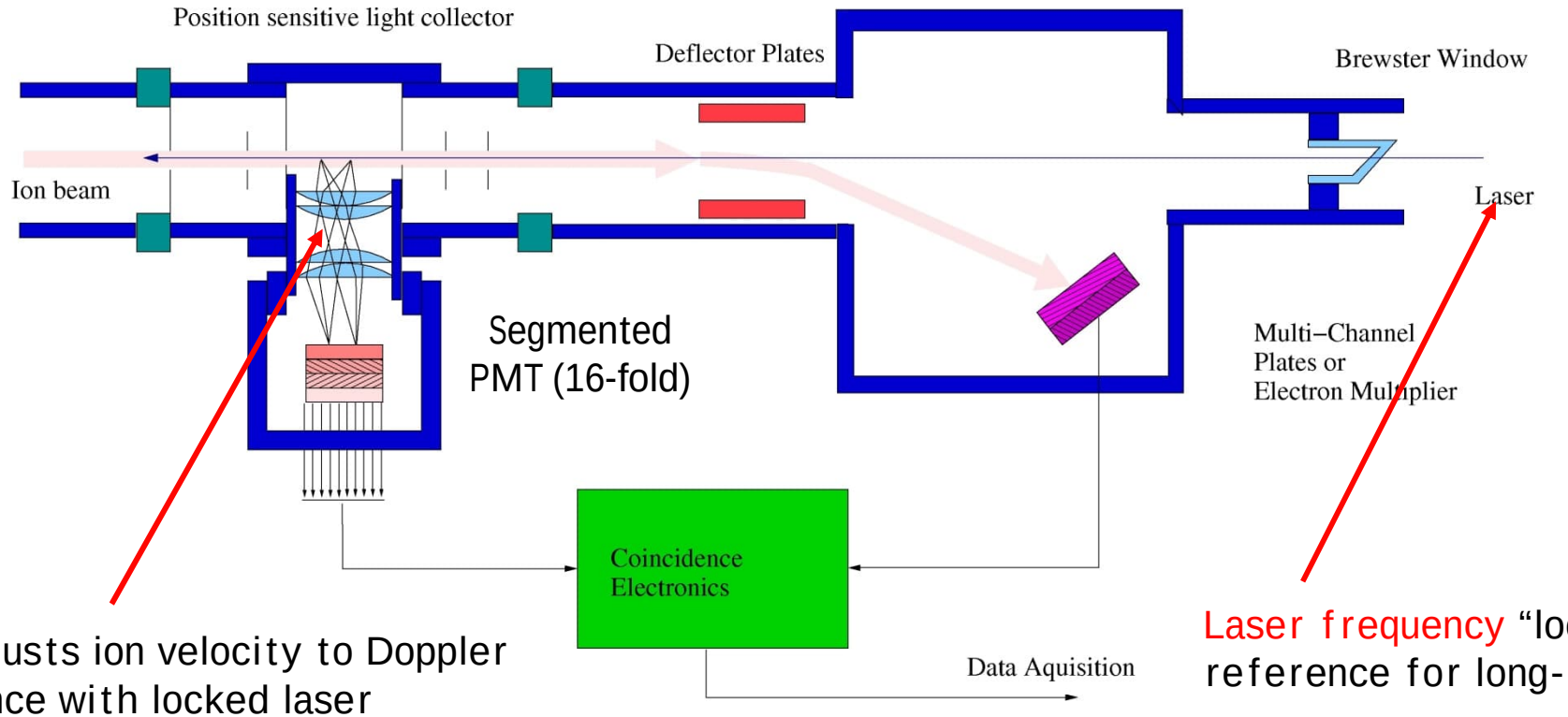
Photon-ion coincidence detection



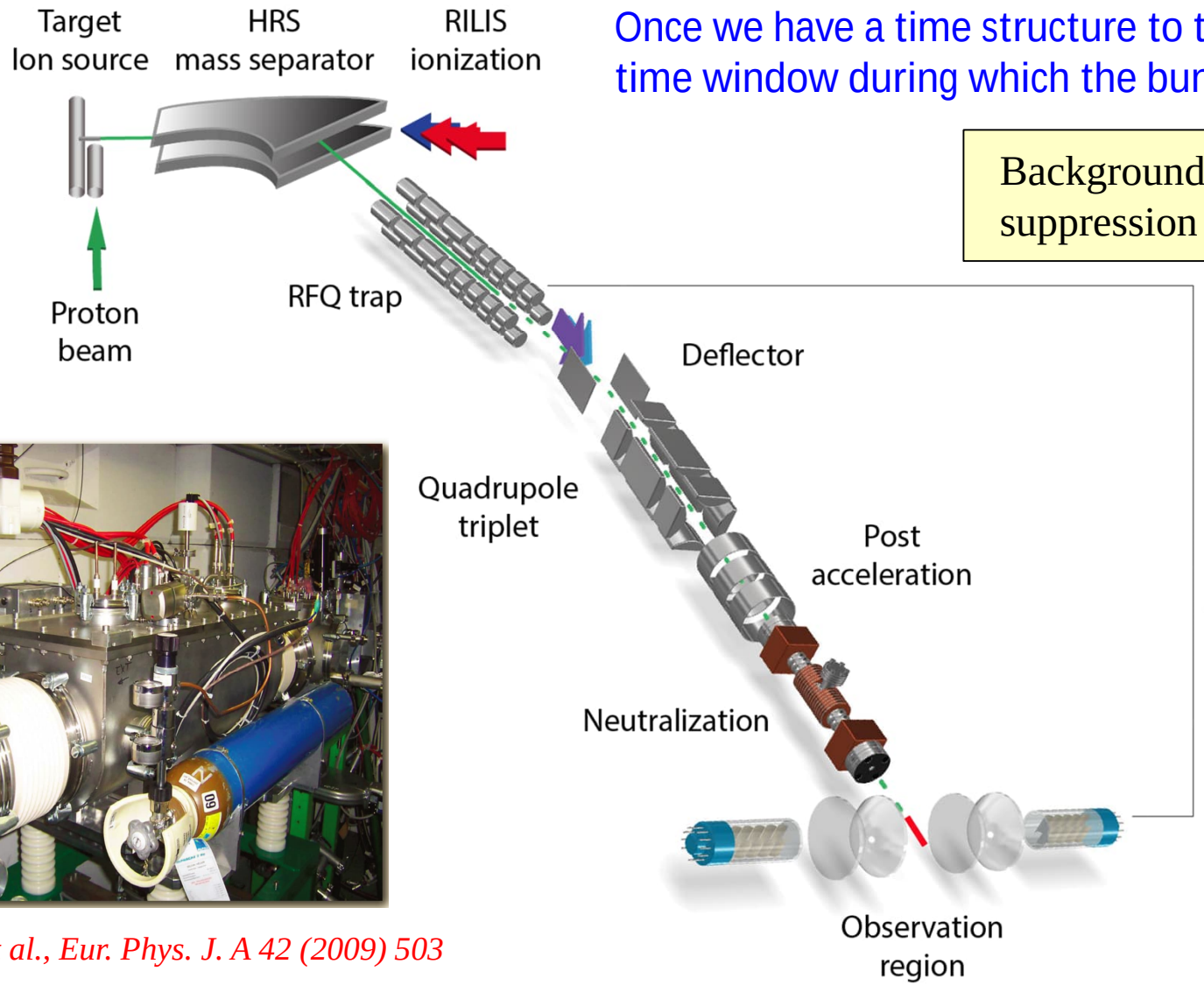
Tuning voltage adjusts ion velocity to Doppler shift it to resonance with locked laser

Laser frequency "locked" to a reference for long-term stability

Accept photons in delayed coincidence with the corresponding ion (or atom). Position sensitivity along the detection region can enhance the time resolution (to ~20 ns). Good for beams with little isobaric contamination.

D.A. Eastham et al., Opt. Commun. ***82 (1-2)*** *(1991) 23*

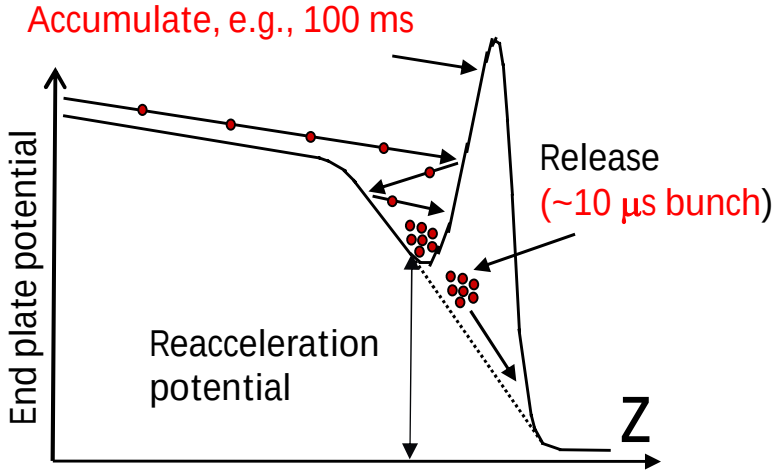
From continuous to bunched beams



Once we have a time structure to the beam we can accept photons in a time window during which the bunched beam passes

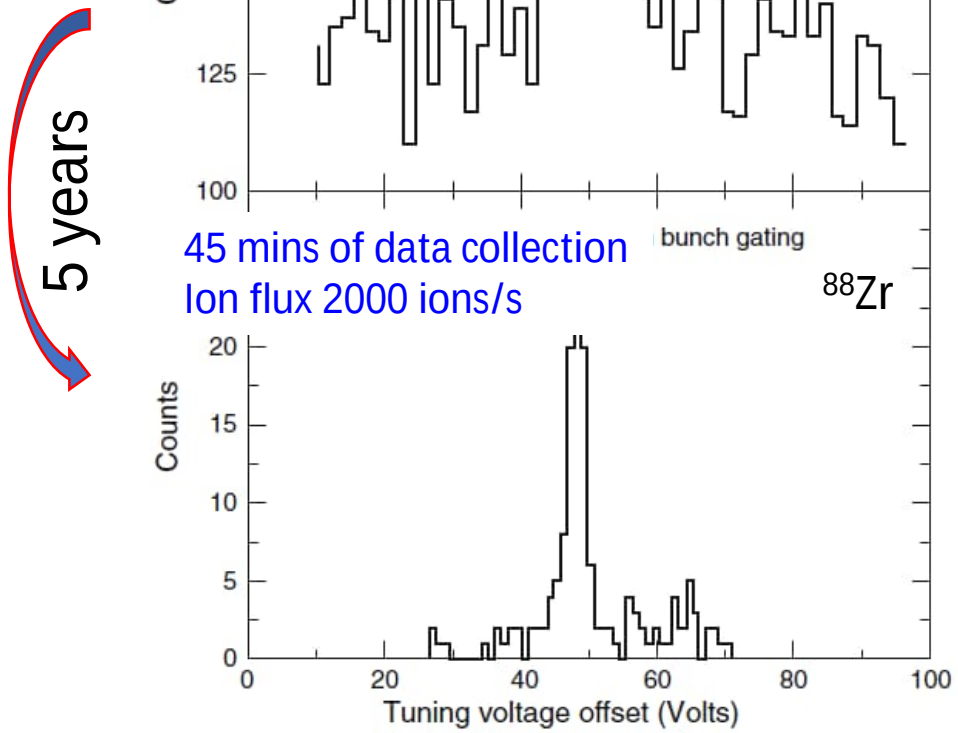
$$\text{Background suppression} = \frac{\text{e.g., 100ms accumulation}}{\text{10µs gate width}} \sim 10^4$$

E. Mane et al., Eur. Phys. J. A 42 (2009) 503

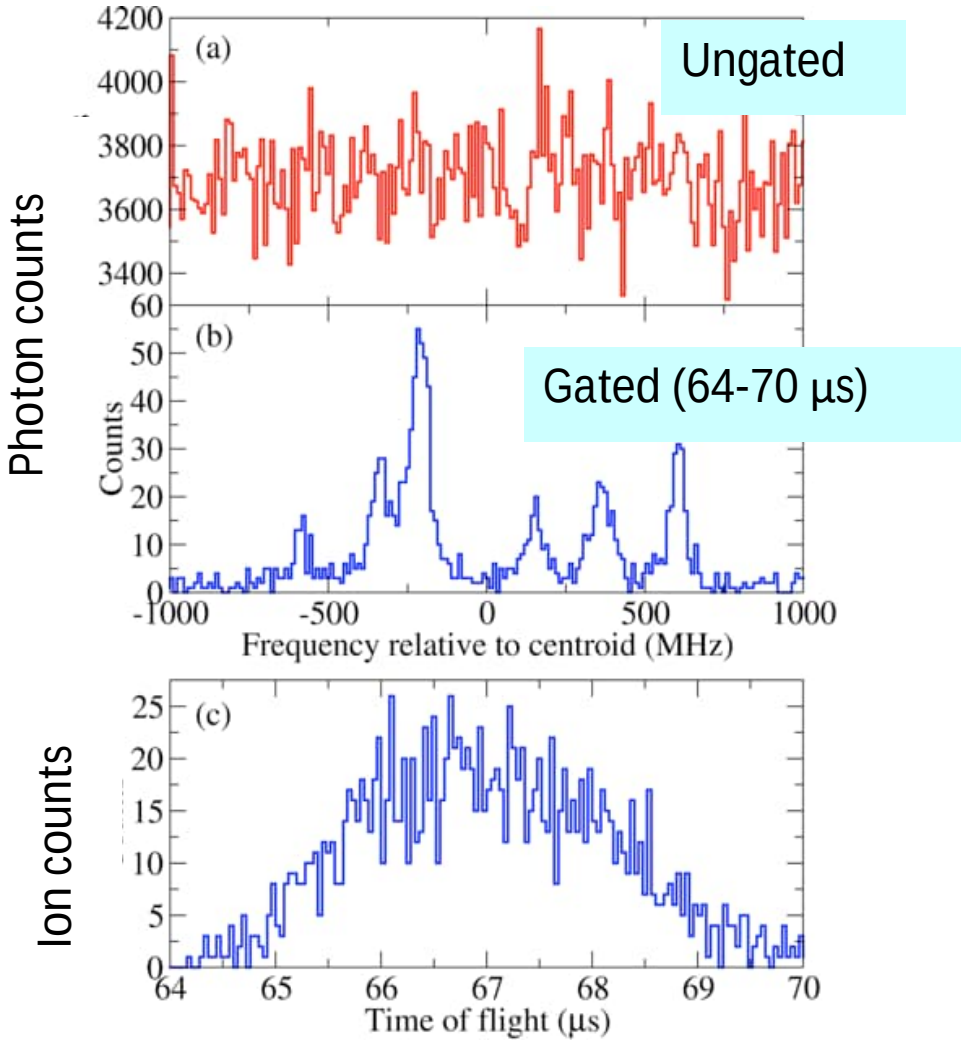


*P. Campbell et al., PRL **89** (2002) 082501*

Photon-ion coincidence vs. bunched beams



B. Cheal and D.H. Forest, Hyp. Int. ***223*** *(2014) 63*



E. Mané et al., PRC ***84*** *(2011) 024303*

From photon detection to ion detection

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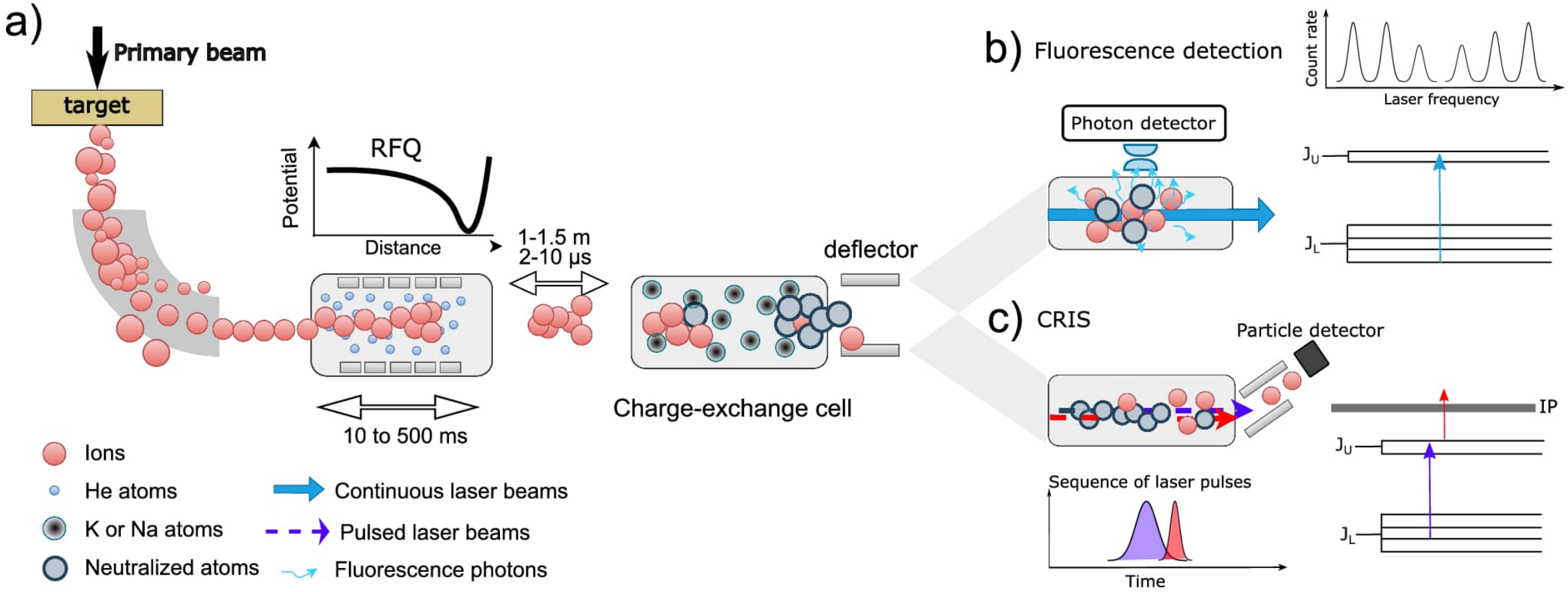
Production | Manipulation | Spectroscopy & detection



Note: the application of ion detection (via CRIS) requires charge exchange

Figure from A. Koszorus et al., EPJ A 60 (2024) 20

Complementarity in our techniques

The most suitable technique can be determined based on the scientific questions, spectroscopic properties and ion beam properties.

IN-SOURCE (RIS)

- Selective process
- Short lifetimes, low yields (<1 ion/s)
- High detection efficiency
- Poor resolution (100-1000× < CLS)

CLS (photons) / CRIS (ions/decays)

- High resolution: (~10-100 MHz)
- Beams of some 10^3 - 10^4 ions/s (CLS)
- Few tens of ions/s (CRIS)

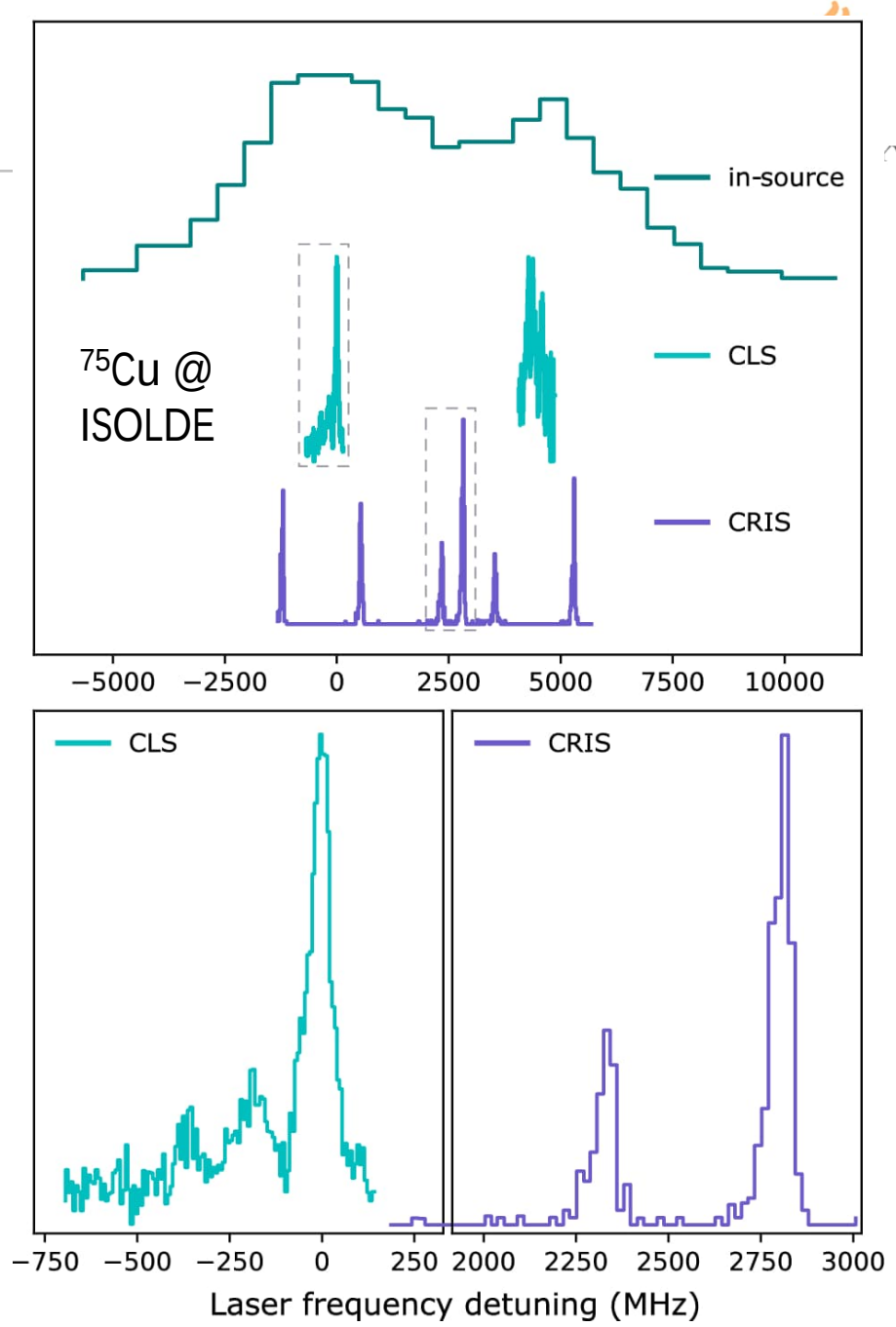
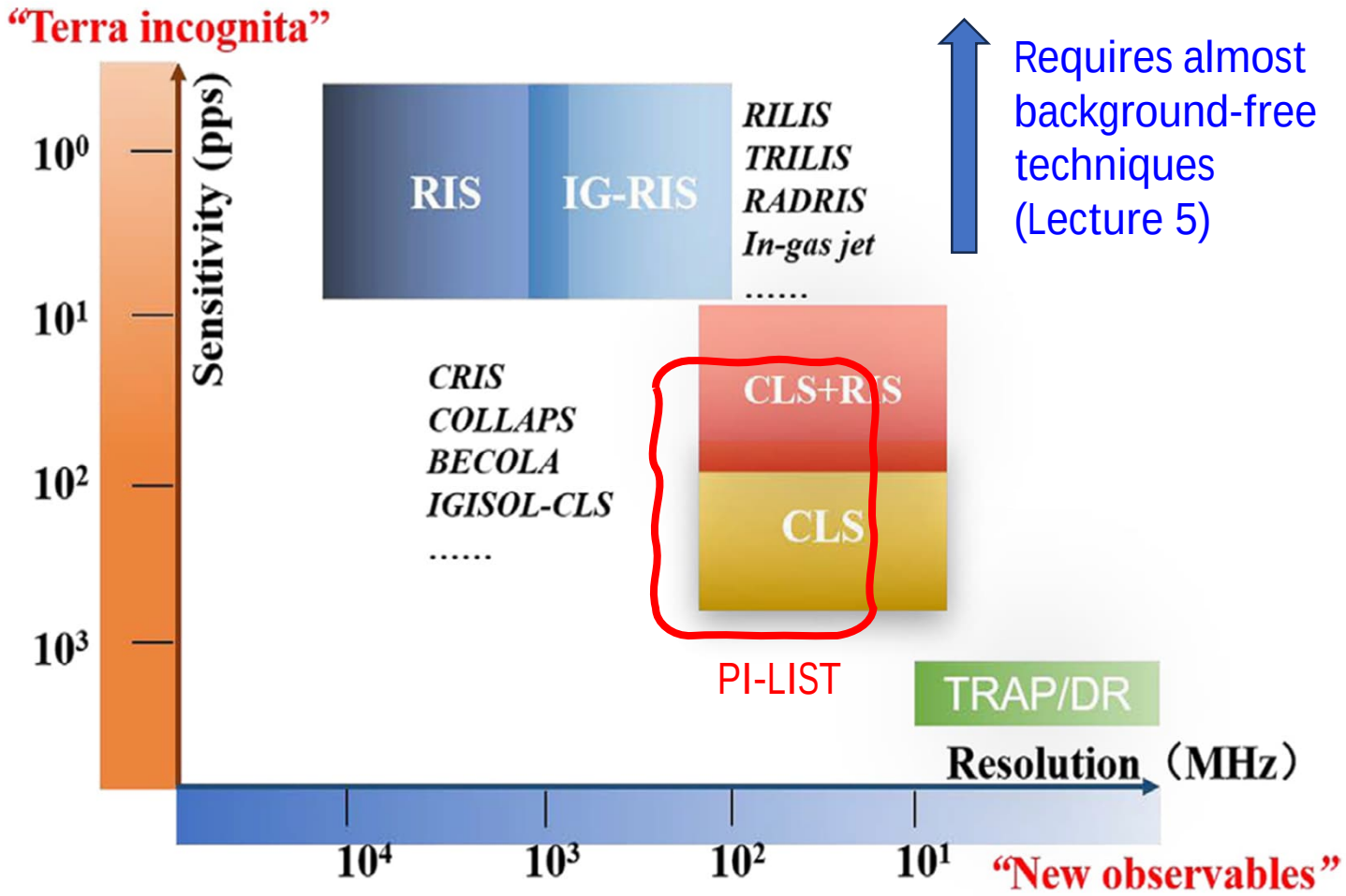


Figure from: A. Koszorus et al., Eur. Phys. J. A 60 (2024) 20

Sensitivity vs resolution



Requires almost background-free techniques (Lecture 5)

GHz

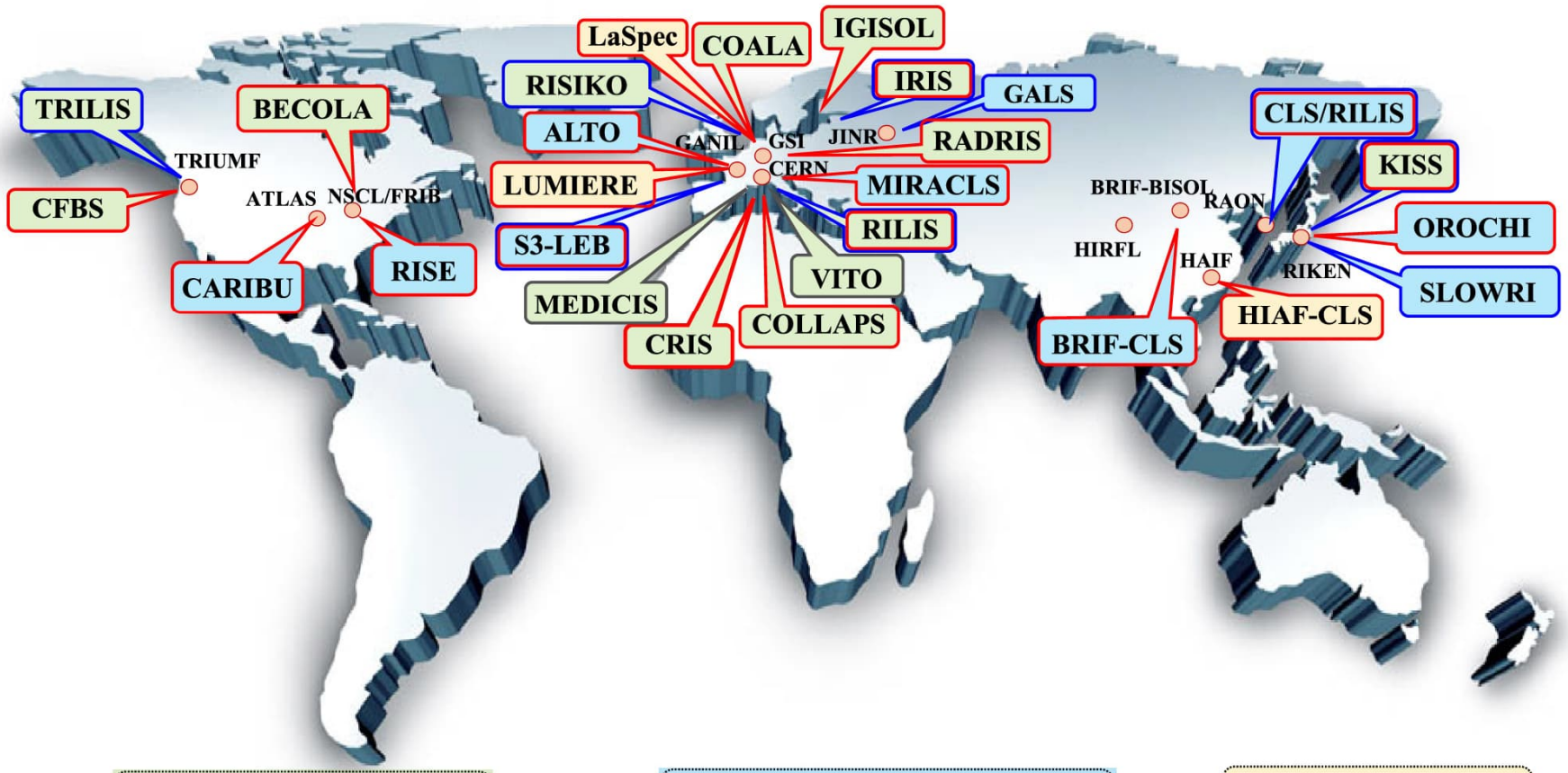
- Magnetic dipole moments
- Electrical quadrupole moments and charge radii
- Hyperfine anomaly
  - Distribution of magnetization inside nuclear volume
- Higher-order moments
  - Magnetic octupole…
- Higher-order moments of the charge radii
  - e.g., $\langle r^4 \rangle$ - surface thickness of nuclear density
- Beyond-standard model physics from Hz-level isotope shift spectroscopy

sub-Hz

And we work in a typical lifetime range from ms to stable nuclei

Figure modified from: X.F. Yang et al., PPNP 129 (2023) 104005

Laser spectroscopy setups worldwide



X.F. Yang et al., PPNP 129 (2023) 104005

Take home messages and summary

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Our beautiful techniques

- Laser spectroscopy of radioactive nuclei features (or is planned) at almost all online radioactive ion beam facilities.
- Our spectroscopy can be performed at both ISOL and IGISOL (traditional) as well as fragmentation facilities (via gas catcher developments).
- Lower-resolution in-source methods are complementary to high-resolution techniques and are often used ``together´´. The choice of technique is determined by the observables we wish to measure and the yield of our exotic nuclei.