Off-line and On-line Applications of High Resolution Laser Spectroscopy on Exotic Species – Collinear and Alternative Approaches

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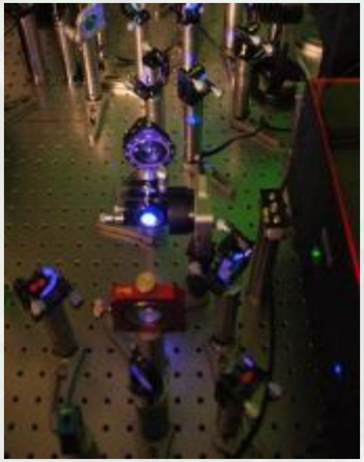
Outline

- **The Study of Exotic Nuclides –** from the Periodic Table to the Nuclear Chart

- **Scientific Findings and Data** from laser spectroscopy



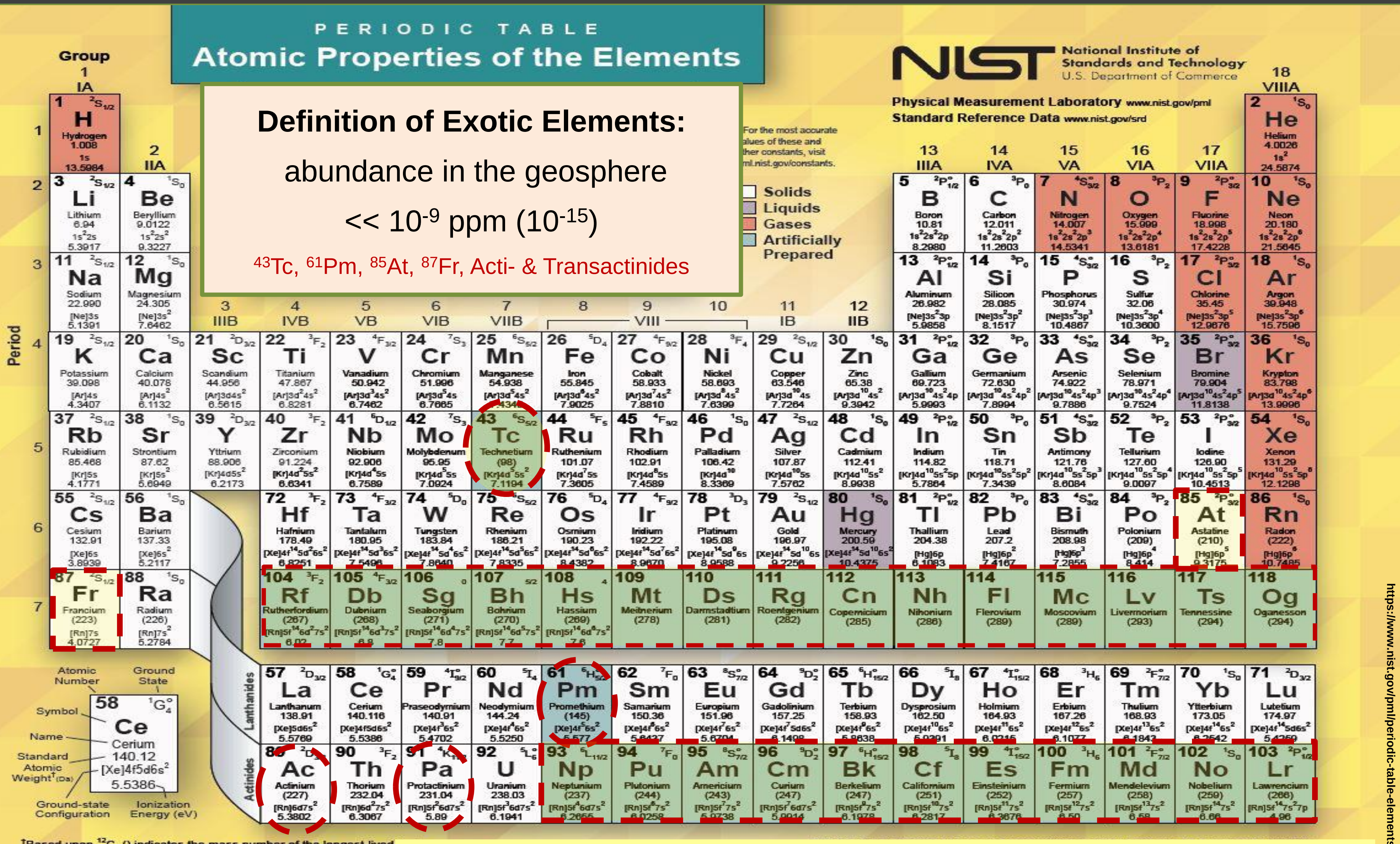
  - experimental prerequisites and techniques
  - high resolution laser spectroscopy
  - line broadenings
  - collinear versus other experimental techniques

- **Some Specific Results** on most **Exotic Elements** and **Isotopes**

- Summary and Outlook

UNESCO
United Nations Educational, Scientific and Cultural Organization
2019 IYPT
International Year of the Periodic Table of Chemical Elements

The Periodic System 2019 – with todays Exotic Elements



Definition of Exotic Elements:

abundance in the geosphere

<< 10^{-9} ppm (10^{-15})

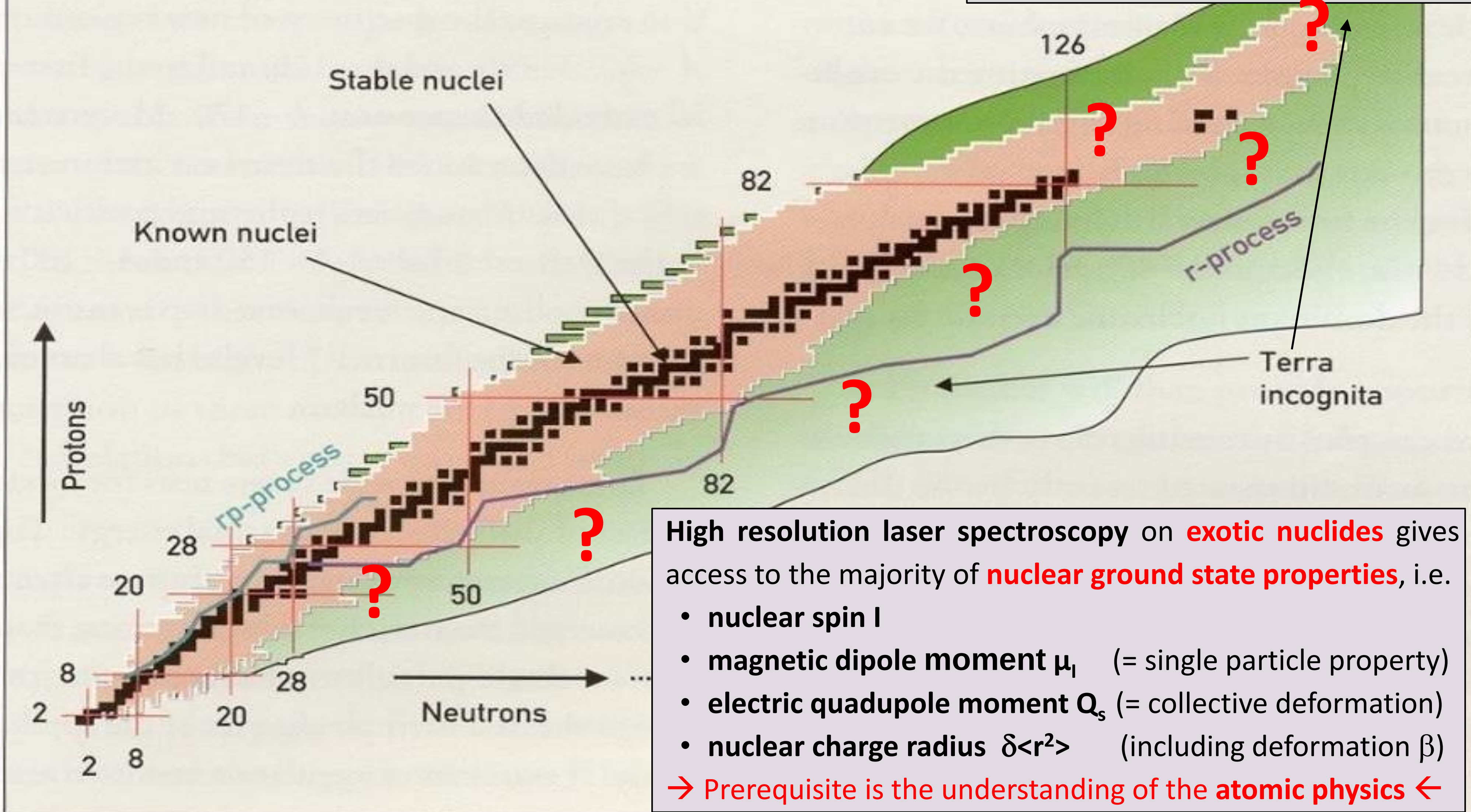
^{43}Tc, ^{61}Pm, ^{85}At, ^{87}Fr, Acti- & Transactinides

…going on-line – heading for exotic **isotopes** far off stability…

From the Periodic System to the Nuclear Chart…

… or from **Exotic Elements** to **Exotic Nuclides** …

The nuclear chart is tracking the pathway from the Big Bang to the world around us…

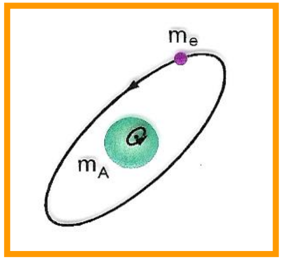


High resolution laser spectroscopy on **exotic nuclides** gives access to the majority of **nuclear ground state properties**, i.e.

- **nuclear spin I**
- **magnetic dipole moment μ_I** (= single particle property)
- **electric quadupole moment Q_s** (= collective deformation)
- **nuclear charge radius $\delta\langle r^2\rangle$** (including deformation β)

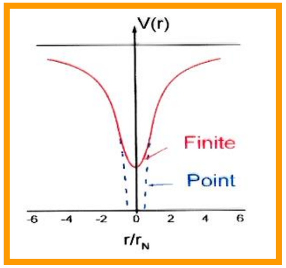
→ Prerequisite is the understanding of the **atomic physics** ←

The Influence of the Nucleus on the Atomic Structure

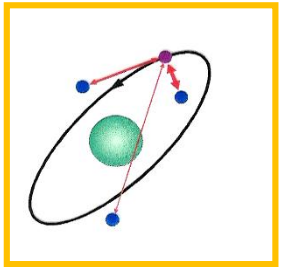


$$H\Psi = E\Psi$$

$$\sum_{i=1}^{N} \frac{-\hbar^2}{2m}\nabla^2 \longleftarrow U + V \longrightarrow V(\vec{r}_1\vec{s}_1, \vec{r}_2\vec{s}_2 \ldots \vec{r}_n\vec{s}_n)$$



$$\frac{\vec{p}^{\,2}}{2\mu} = \sum_i \frac{\vec{p}_i^{\,2}}{2\mu} + \sum_i \sum_{j>i} \frac{\vec{p}_i \cdot \vec{p}_j}{\mu}$$



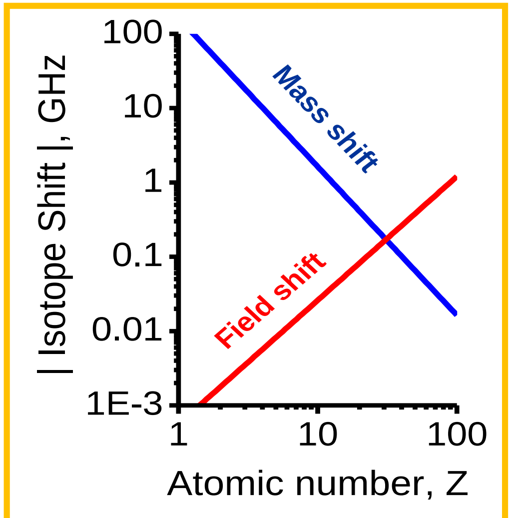
$$V_{Ze} + V_{Is} + V_{I\ell} + \underbrace{V_{ee} + V_{s\ell} + V_{ss} + V_{ll}}_{\text{pure electronic}}$$

Normal Mass Shift **Specific Mass Shift** **Field Shift** **Hyperfine Structure** →

$$\Delta\nu_{ij}^{AA'} = \nu_{ij}^{0}\frac{m_e}{m_p}\frac{(A-A')}{AA'} + C_{ij}\nu_{ij}^{0}\frac{(A-A')}{AA'} + \frac{\pi a_0^3}{Z}\Delta|\Psi(0)|^2 f(Z)\left[\delta\langle r^2\rangle^{AA'} + C_1\delta\langle r^4\rangle^{AA'} + \ldots\right]$$

$$\Delta\nu_{IS} = (M_N + M_S)\frac{(A-A')}{AA'} + F\delta\langle r^2\rangle^{AA'}$$

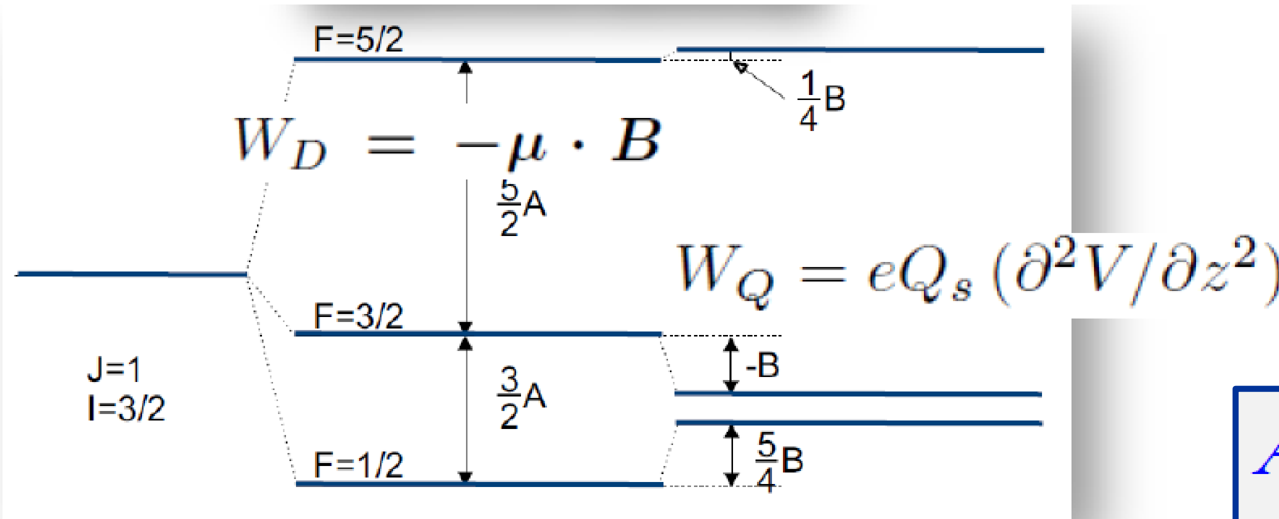
→ Mean squared nuclear charge radius difference and deformation β



From Hyperfine Structure to the Nuclear Moments μ_I and Q_s

Atomic Level Splittings by Coupling of Electron Angular Momentum J and Nuclear Spin I via the Moments μ_I and Q_s

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



Nuclear Magnetic Dipole Moment μ_I

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

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

Nuclear Electric Quadrupole Moment Q_s

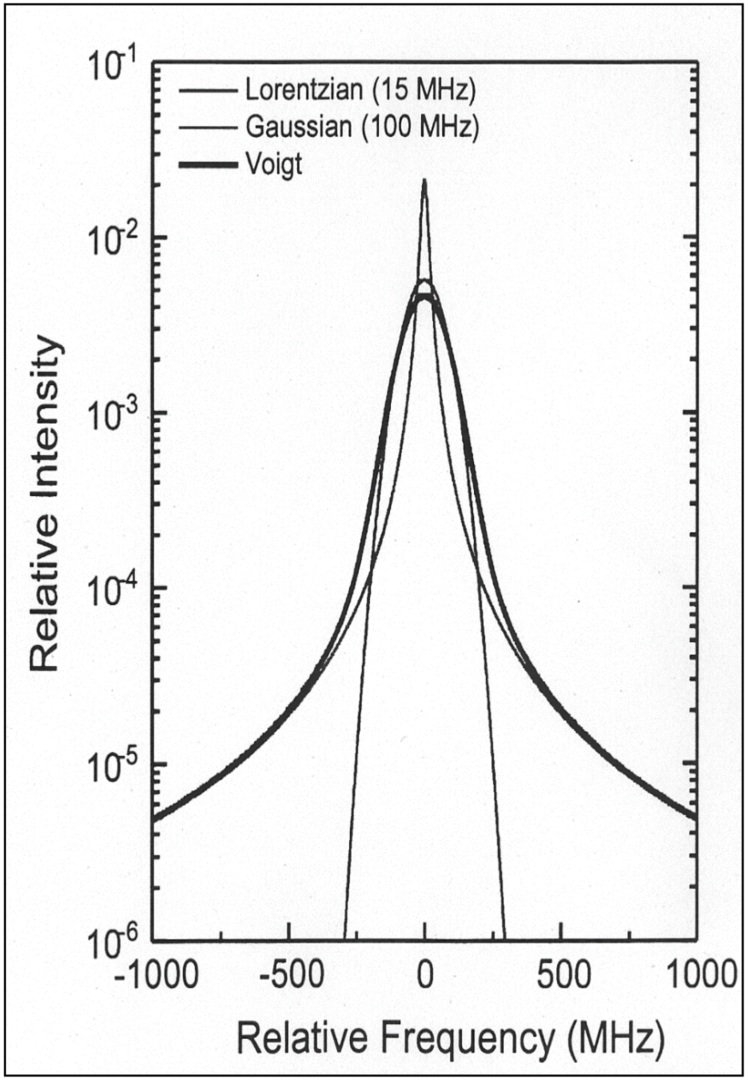
$$W_F = \frac{1}{2}AC + B\,\frac{\frac{3}{4}C(C+1) - I(I+1)J(J+1)}{2I(2I-1)J(2J-1)}$$

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

Spectral Considerations for Narrowband Laser Excitation

Resolution in laser spectroscopy determined by

- Single free atom at rest → natural linewidth = **Lorentzian of ~10 MHz**
- Ensemble of thermally moving atoms in vapor → **Gaussian 500 MHz – 5 GHz**
- Additional contribution from laser line width → **Gaussian 1 MHz(cw) – 5 GHz(pulsed)**
- Strong linewidth suppression down to natural linewidth by fast beam **Doppler** compression in **collinear laser spectroscopy** (on ions, atoms or molecules)



Lorentzian (homogeneous)

$$L(\nu) = \frac{1}{\pi[w_L + (\nu_0 - \nu)^2 / w_L]}$$

$$w_L = HWHM = \frac{1}{4\pi\tau}$$

Gaussian (inhomogeneous)

$$G(\nu) = \frac{1}{w_G\sqrt{\pi}} \exp\left[-(\nu_0 - \nu)^2 / w_G^2\right]$$

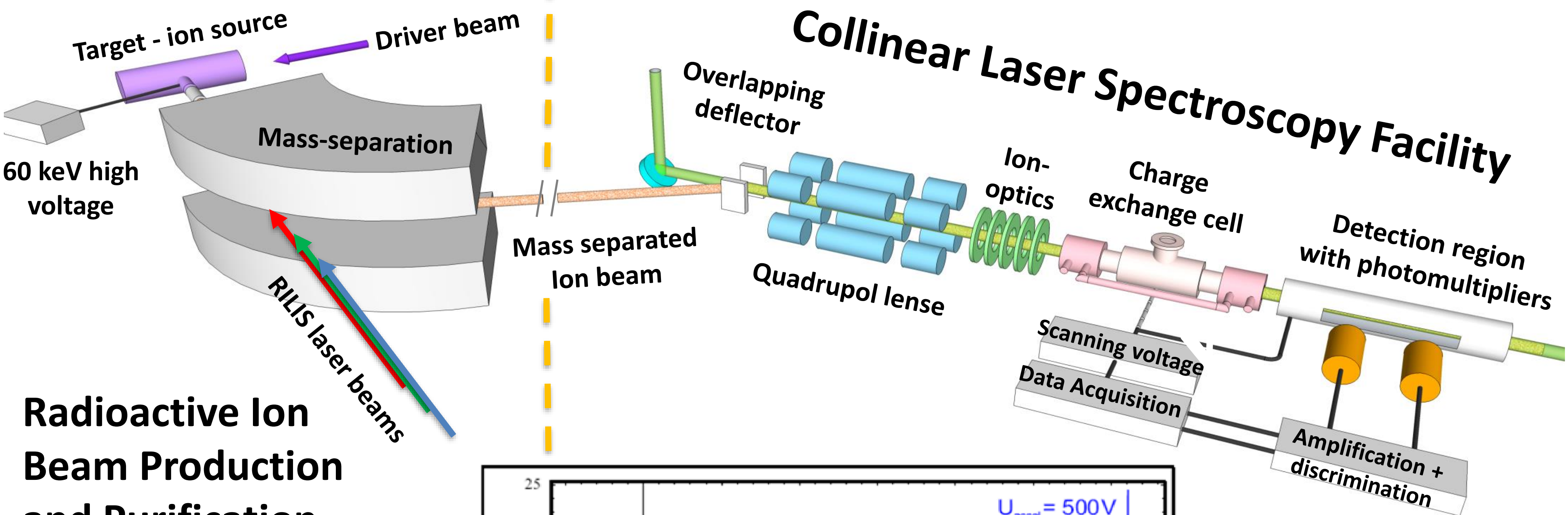
$$w_G = HW_{\frac{1}{e}} = 4.3\mathrm{x}10^{-7}\,\nu_0\sqrt{T/M}$$

Voigt (convolution)

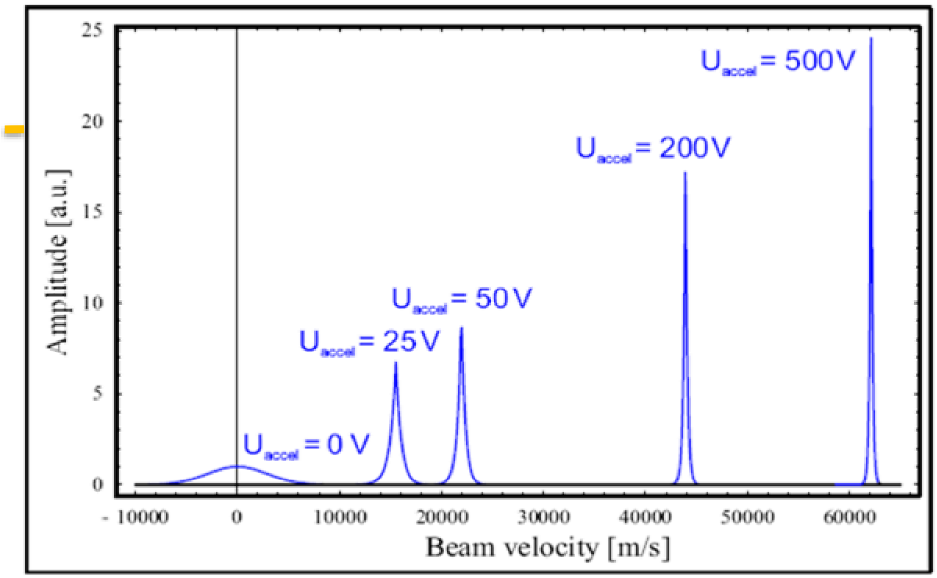
$$V(\nu) = \int_{-\infty}^{\infty} G(\nu')L(\nu - \nu')\,d\nu'$$

Klaus D.A. Wendt

Principles of Collinear Laser Spectroscopy



Radioactive Ion Beam Production and Purification



$$\nu_{Ion} = \nu_{Laser} \cdot \gamma \cdot \left(1 - \frac{\upsilon}{c}\right)$$

$$E = eU = 1/2\, m\upsilon^2$$

$$\delta E = m\upsilon\, \delta\upsilon$$

The Cradle of Collinear Laserspectroscopy…

at the **MAFIA** on-line separator of the TRIGA Research Reactor of UMz in 1978

AN ON-LINE MASS SEPARATOR FOR FISSION-PRODUCED ALKALI ISOTOPES

L. VON REISKY *, J. BONN, S.L. KAUFMAN **, L. KUGLER, E.-W. OTTEN, J.-M. RODRIGUEZ
K.P.C. SPATH and D. WESKOTT
Institut für Ph

Collinear Laser Spectroscopy on Fast Atomic Beams

K.-R. Anton, S. L. Kaufman,[a] W. Klempt, G. Moruzzi,[b] R. Neugart, E.-W. Otten, and B. Schinzler
Institut für Physik, Johannes Gutenberg–Universität, D-6500 Mainz, Federal Republic of Germany

Phys. Rev. Lett. 40 (1978) 642

Z. Physik A 289, 227–228 (1979)

Short Note

Hyperfine Structure and Isotope Shifts of Neutron-Rich $^{138-142}$Cs

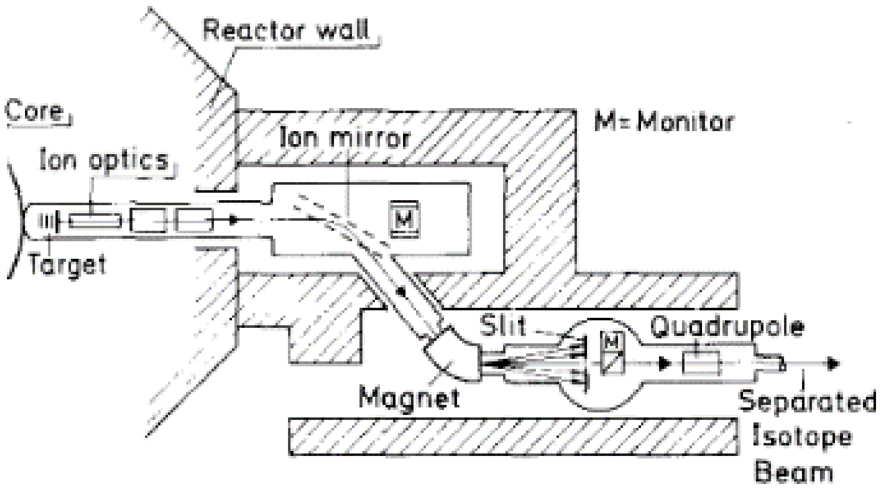
J. Bonn, W. Klempt, R. Neugart, E.-W. Otten, and B. Schinzler
Institut für Physik, Johannes Gutenberg Universität, Jakob-Welder-Weg
Mainz,
Federal Republic of Germany

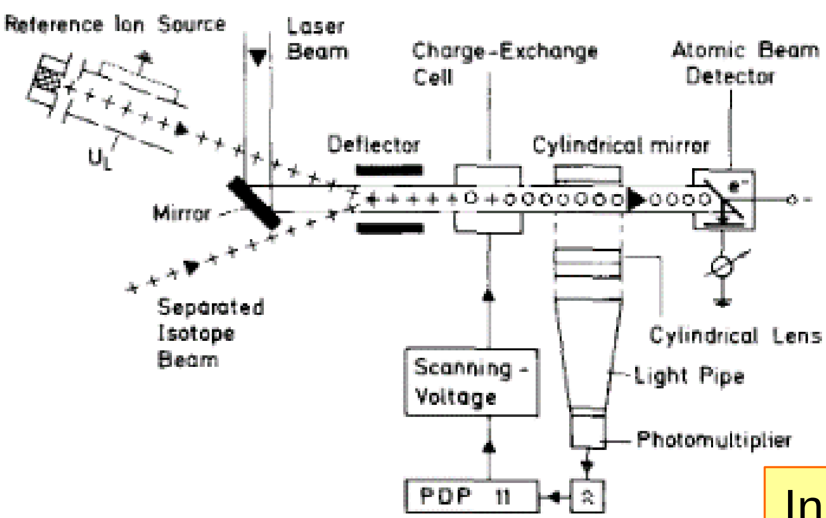
Volume 79B, number 3 PHYSICS LETTERS 20 November 1978

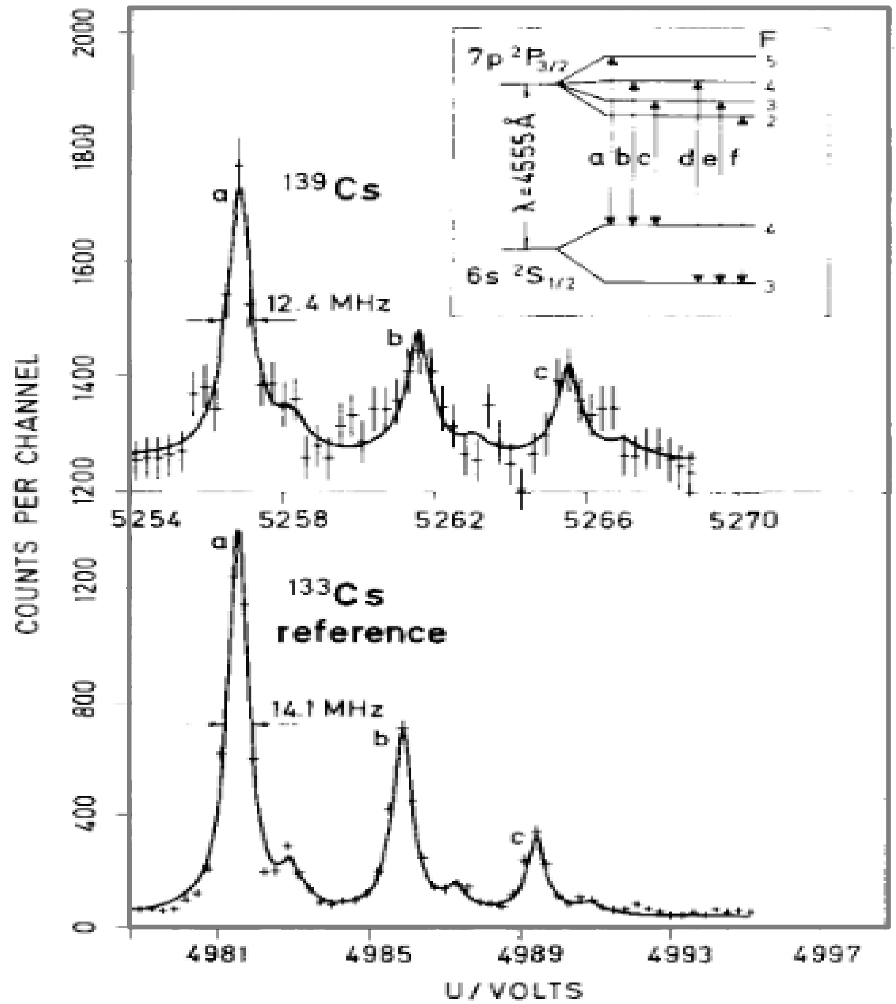
COLLINEAR LASER SPECTROSCOPY OF NEUTRON-RICH Cs ISOTOPES AT AN ON-LINE MASS SEPARATOR

B. SCHINZLER, W. KLEMPT, S.L. KAUFMAN [1], H. LOCHMANN, G. MORUZZI [2], R. NEUGART, E.-W. OTTEN, J. BONN, L. VON REISKY, K.P.C. SPATH, J. STEINACHER and D. WESKOTT
Institut für Physik, Universität Mainz, Germany

The first Hyperfine Structure Spectra in Radiocesium







In reactor core ion source limited to alkaline elements
→ Transfer of CLS to ISOLDE CERN in 1980

The Prosperous First 10 Years of CLS at ISOLDE/CERN

1982

0 **Fast-Beam Laser Spectroscopy on Metastable Atoms applied to Neutron-Deficient Ytterbium Isotopes**
F. Buchinger, A.C. Mueller, B. Schinzler, K. Wendt,
C. Ekström, W. Klempt, and R. Neugart,
Nuclear Instruments and Methods ***202,*** *159-165 (1982)*

Yb

1983

1 **Spins, Moments and Charge Radii of Barium Isotopes in the Range of $^{122\text{-}146}$Ba determined by Collinear Fast-Beam Laser Spectroscopy**
A.C. Mueller, F. Buchinger, W. Klempt and E.W. Otten, R. Neugart, C. Ekström,
J. Heinemeier and The ISOLDE Collaboration,
Nuclear Physics A403 (1983) 234-262

Ba

2 **Nuclear Moments and Charge Radii of Rare-Earth Isotopes studied by Collinear Fast-Beam Laser Spectroscopy**
R. Neugart, K. Wendt, S.A. Ahmad, W. Klempt, and C. Ekström,
Hyperfine Interactions ***15/16****, 181-186 (1983)*

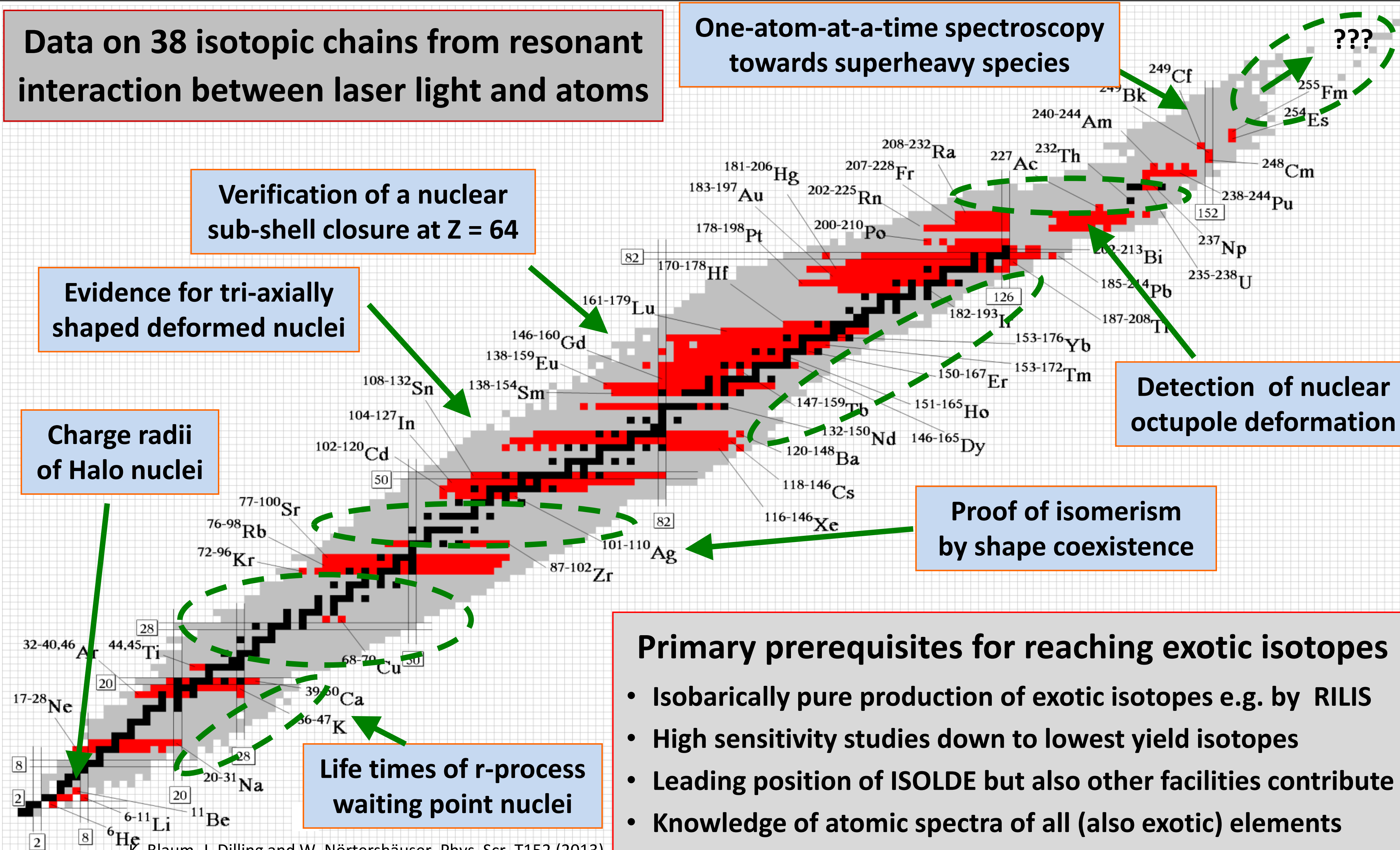
Dy

3 **Determination of Nuclear Spins and Moments in a Series of Radium Isotopes**
S.A. Ahmad, W. Klempt, R. Neugart, E.W. Otten, K. Wendt, and C. Ekström,
Phys. Lett. ***133B****, 47-52 (1983)*

Ra

1984

Nuclear Structure from on-line High Res Laser Spectroscopy



Data on 38 isotopic chains from resonant interaction between laser light and atoms

One-atom-at-a-time spectroscopy towards superheavy species

Verification of a nuclear sub-shell closure at Z = 64

Evidence for tri-axially shaped deformed nuclei

Detection of nuclear octupole deformation

Charge radii of Halo nuclei

Proof of isomerism by shape coexistence

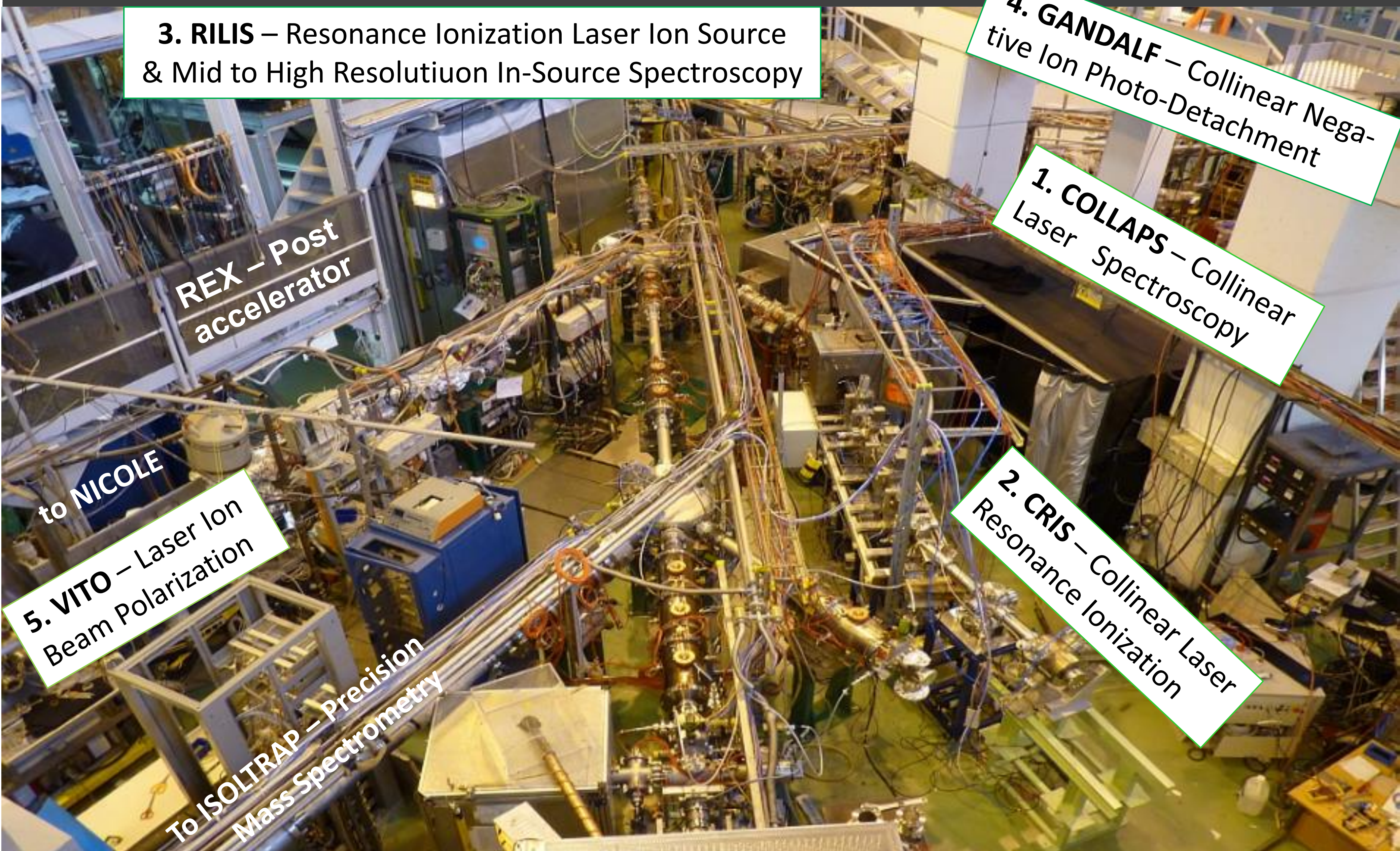
Life times of r-process waiting point nuclei

Primary prerequisites for reaching exotic isotopes

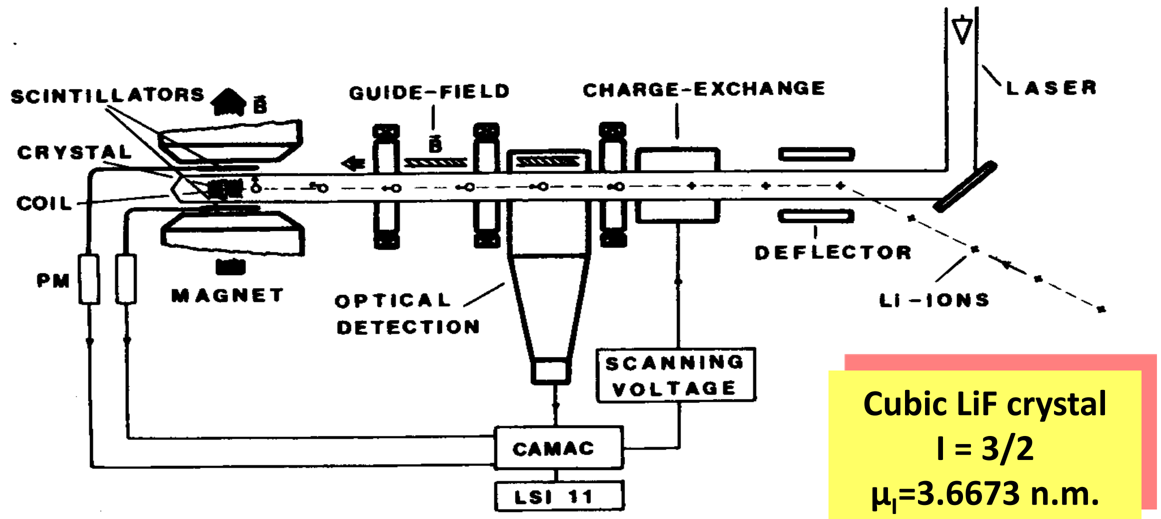
- Isobarically pure production of exotic isotopes e.g. by RILIS
- High sensitivity studies down to lowest yield isotopes
- Leading position of ISOLDE but also other facilities contribute
- Knowledge of atomic spectra of all (also exotic) elements

K. Blaum, J. Dilling and W. Nörtershäuser, Phys. Scr. T152 (2013)

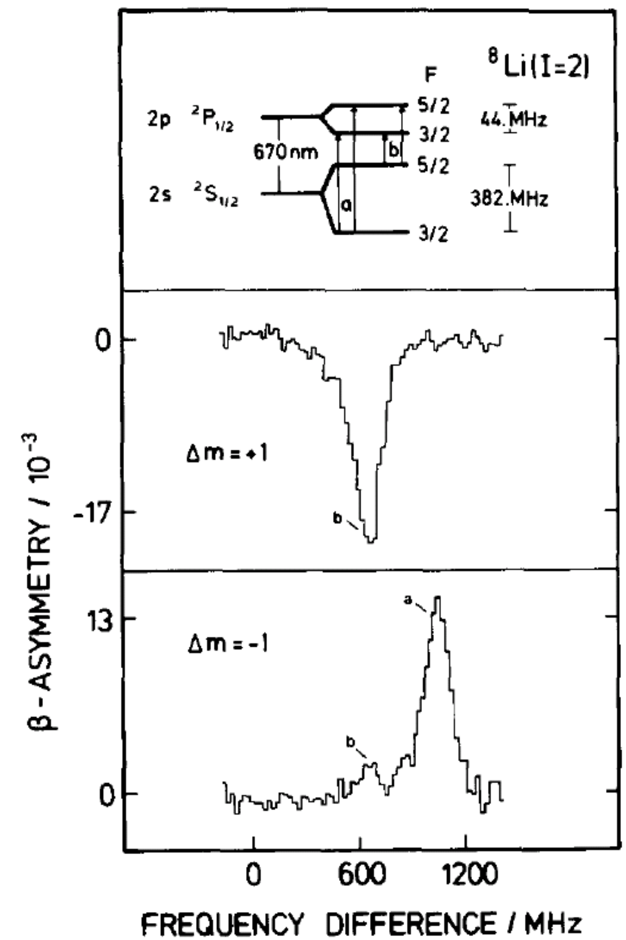
ISOLDE Experimental Hall with Laser & Mass Spectrometry



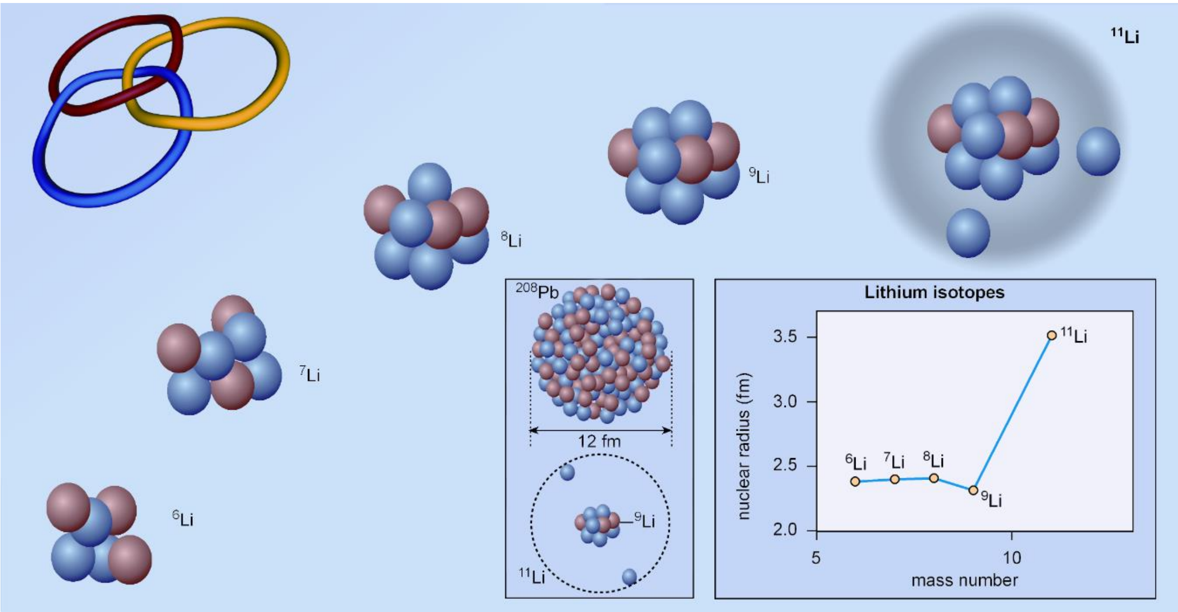
Nuclear Polarization and β-NMR Detection in CLS



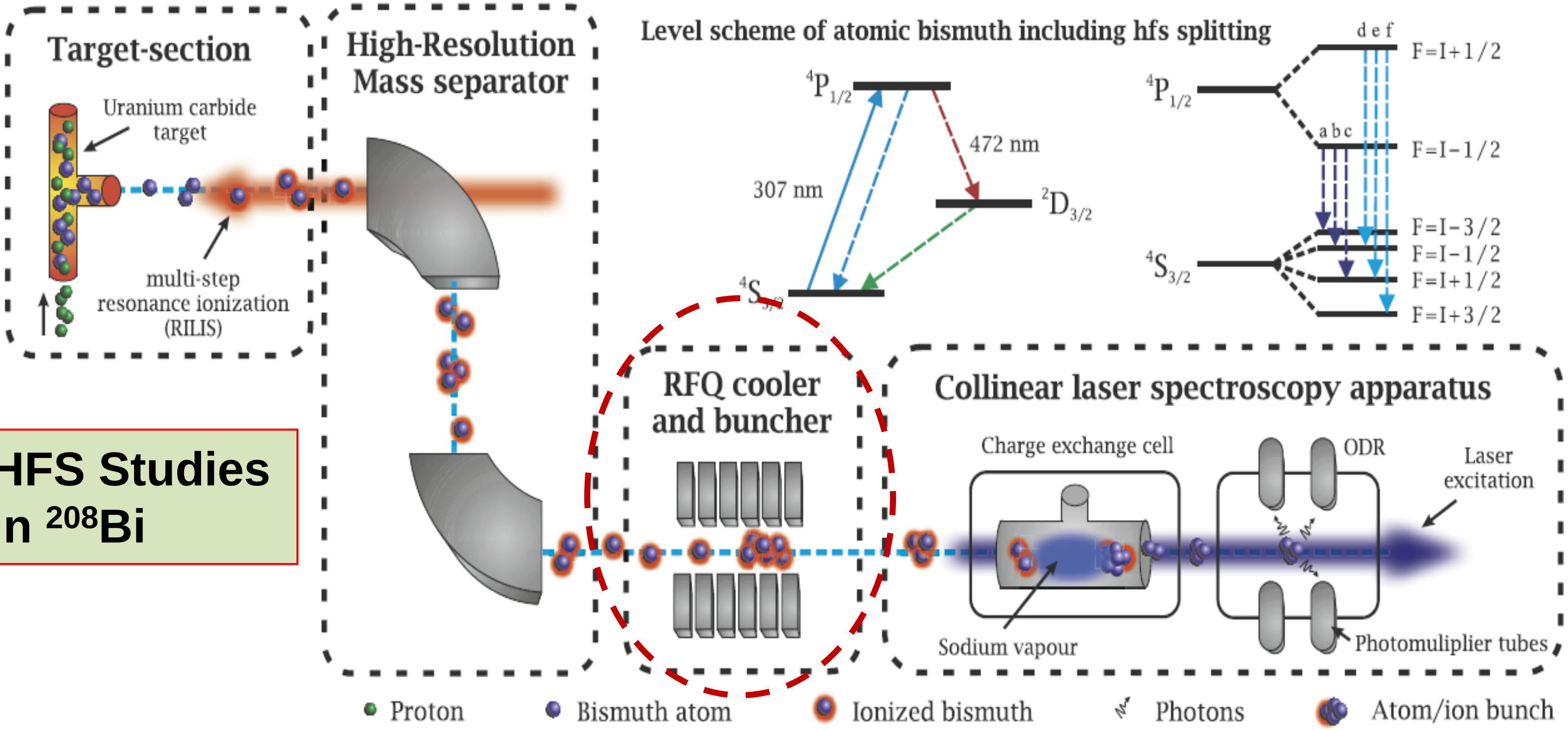
Cubic LiF crystal
I = 3/2
μ_I=3.6673 n.m.

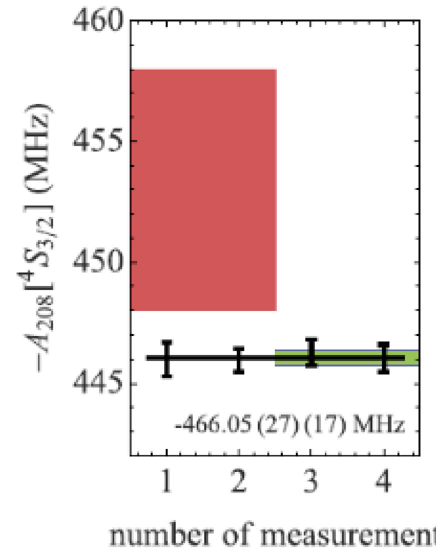


NUCLEAR SPIN AND MAGNETIC MOMENT OF ^{11}Li
E. Arnold et al., Phys. Lett B 197, 311 (1987)

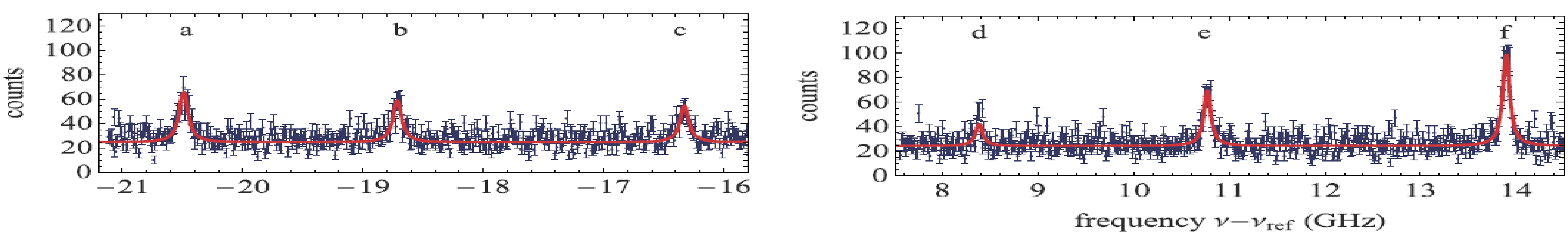


Upgrading of Collaps by Ion Beam Pulsing



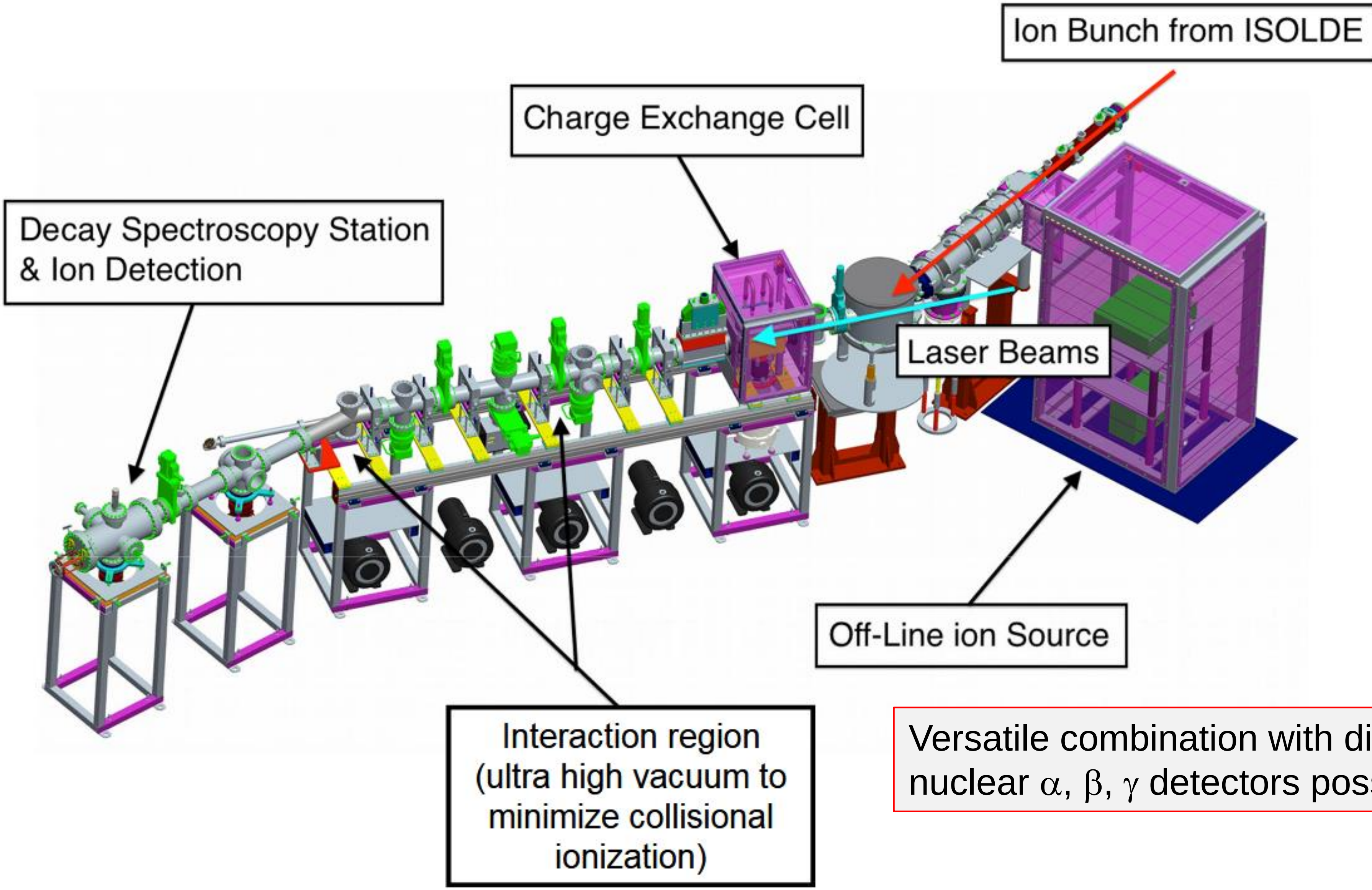


HFS Studies in ^{208}Bi



The nuclear magnetic moment of ^{208}Bi and its relevance for a test of bound-state strong-field QED, S.Schmidt et al., *Physics Letters B 779 (2018) 324–330*

Resonance Ionization Detection in CLS



Versatile combination with different nuclear α, β, γ detectors possible

The CRIS Collaboration

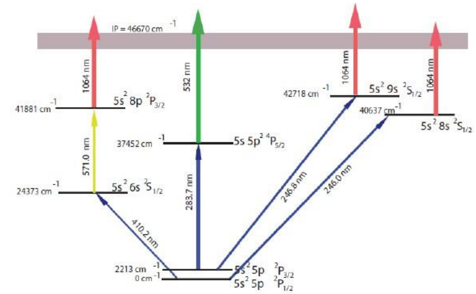
J. Billowes, T.E. Cocolios, K.T. Flanagan, T.J. Procter, A. Smith, I. Strashnov, K.M. Lynch, S. Franchoo, V. Fedosseev, B. Marsh, G. Simpson, M. Bissell, I. Budincevic, R.P. De Groote, S. De Schepper, R.F. Garcia Ruiz, H. Heylen, J. Papuga, G. Neyens, H.H. Stroke, R.E. Rossel, S. Rothe, K. Wendt

Recent CRIS on-line Experiments

Indium:

→ Laser spectroscopy up to ^{101}In (Z=49,N=52)

Yields ~ 100 ions/s

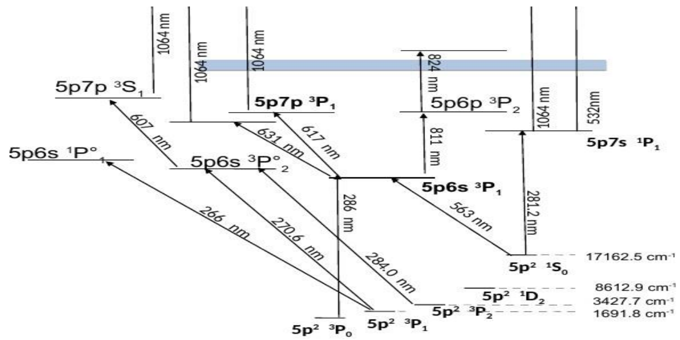


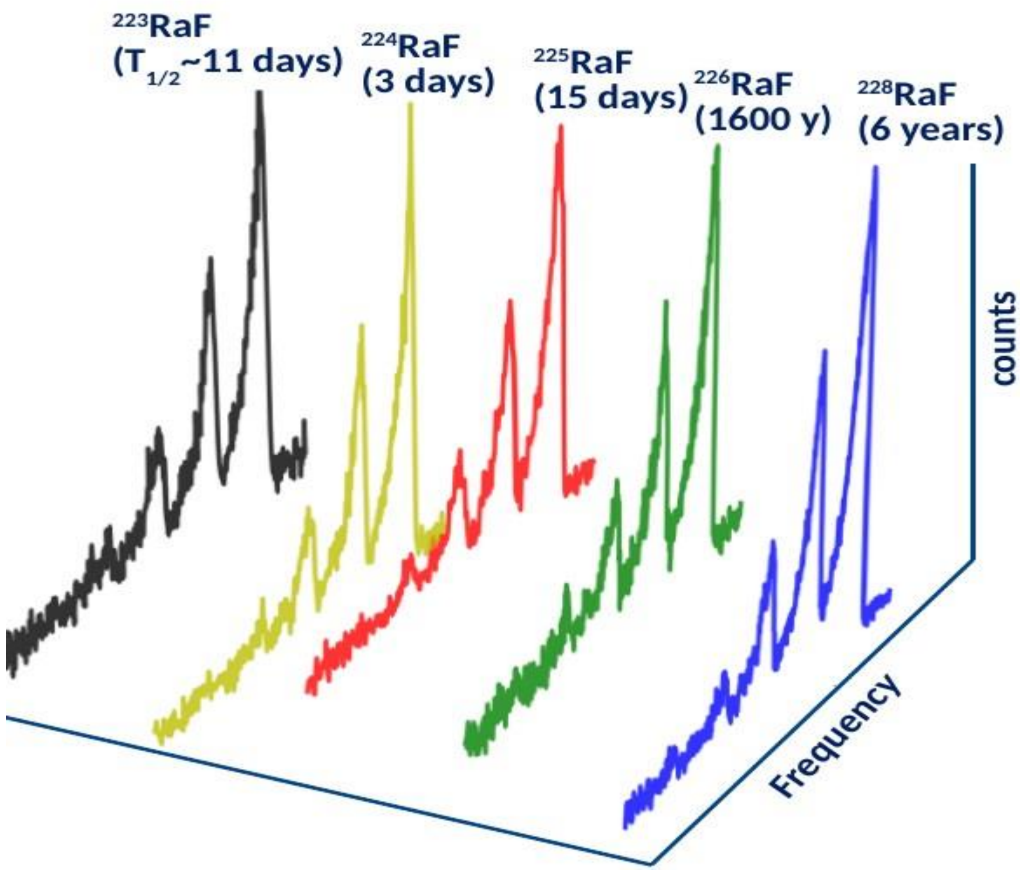
Potassium:

→ Laser spectroscopy of ^{52}K (Z=19, N=33)

Tin:

→ Laser spectroscopy of $^{103-122}$Sn





First on-line Spectroscopy on molecules

→ RaFluoride

Excitation energy of low-lying levels of ^{226}RaF

The Need of a full Collection of Lasers

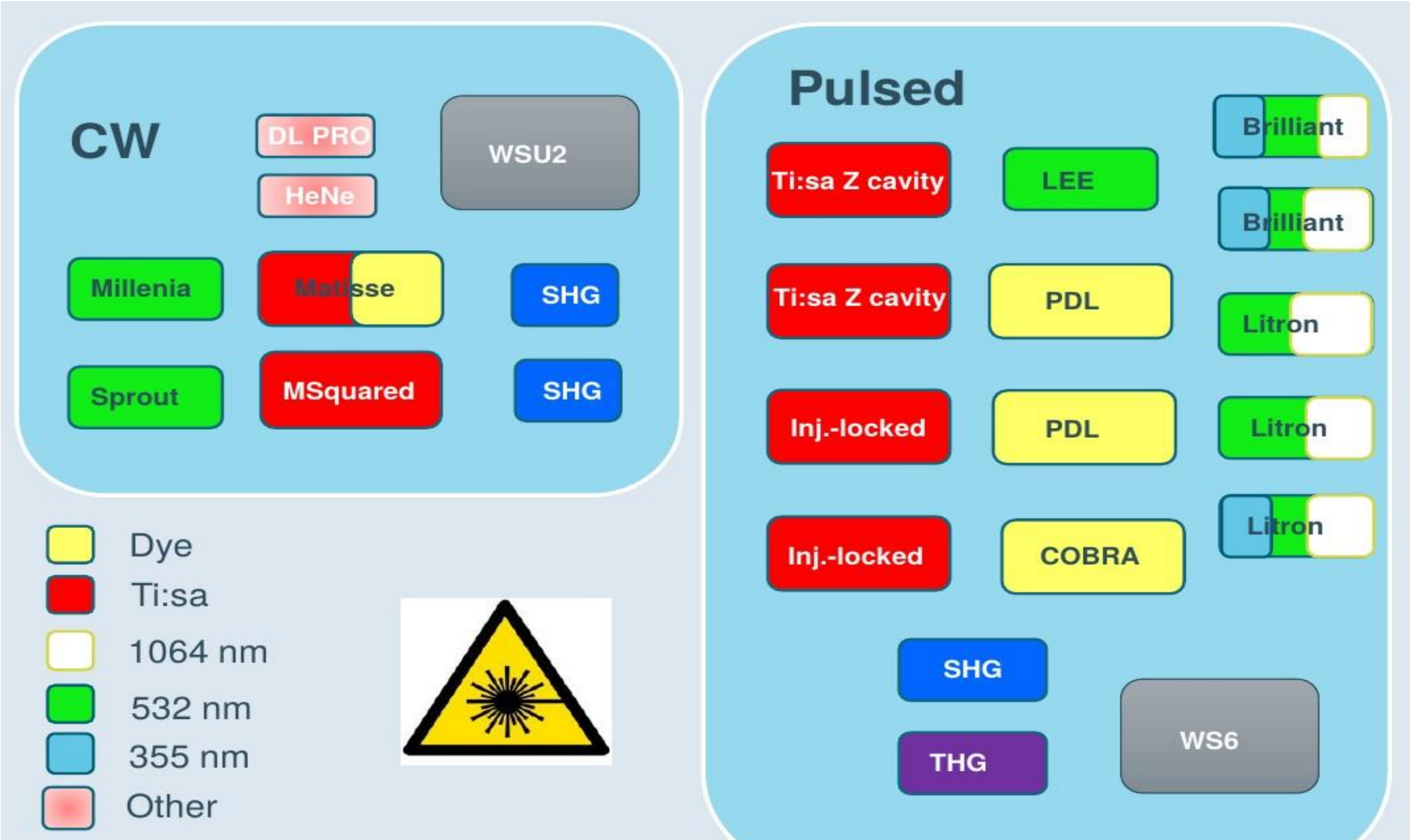
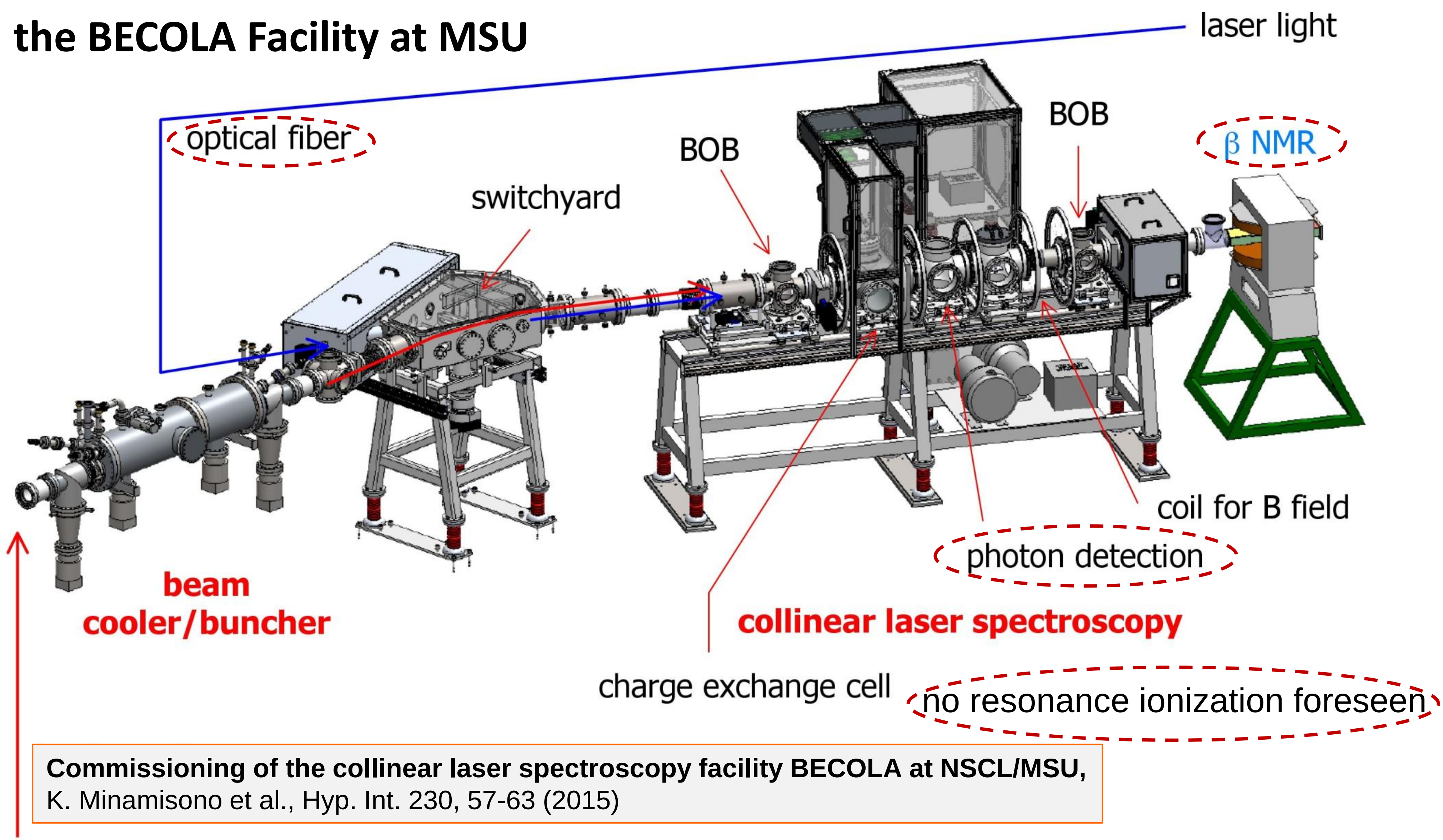


Figure by Agi Koszorus, CERN and KUL

Collinear Laser Spectroscopy up to date

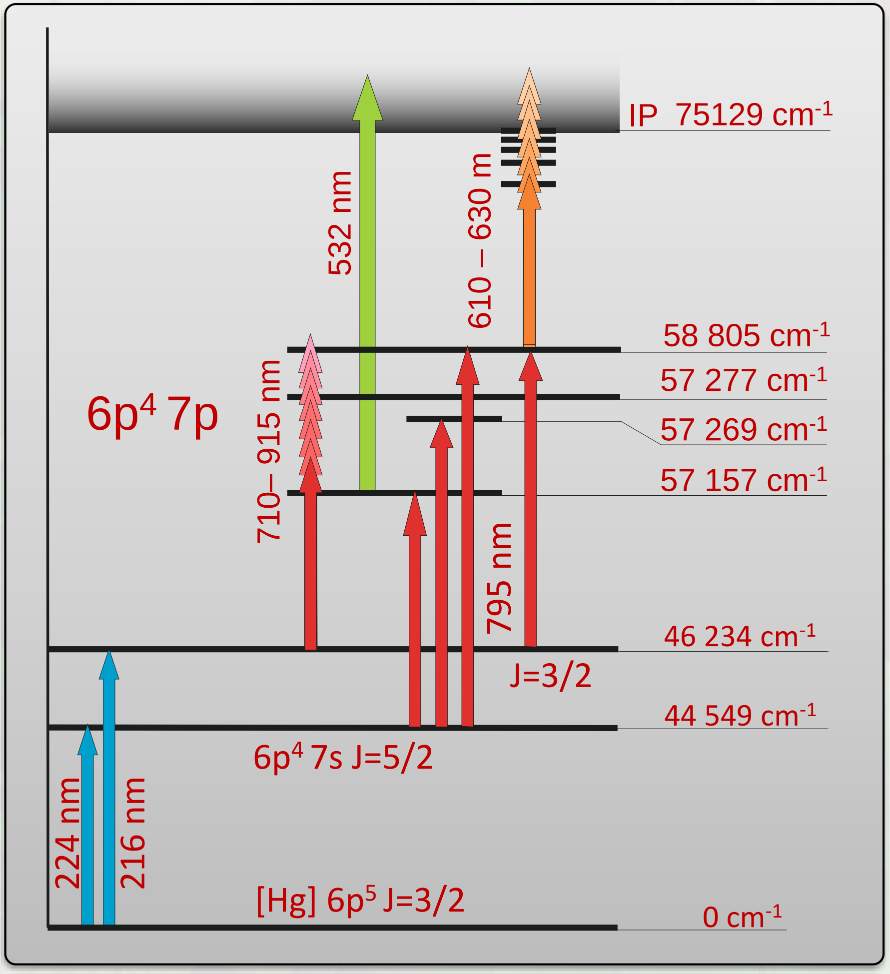
the BECOLA Facility at MSU

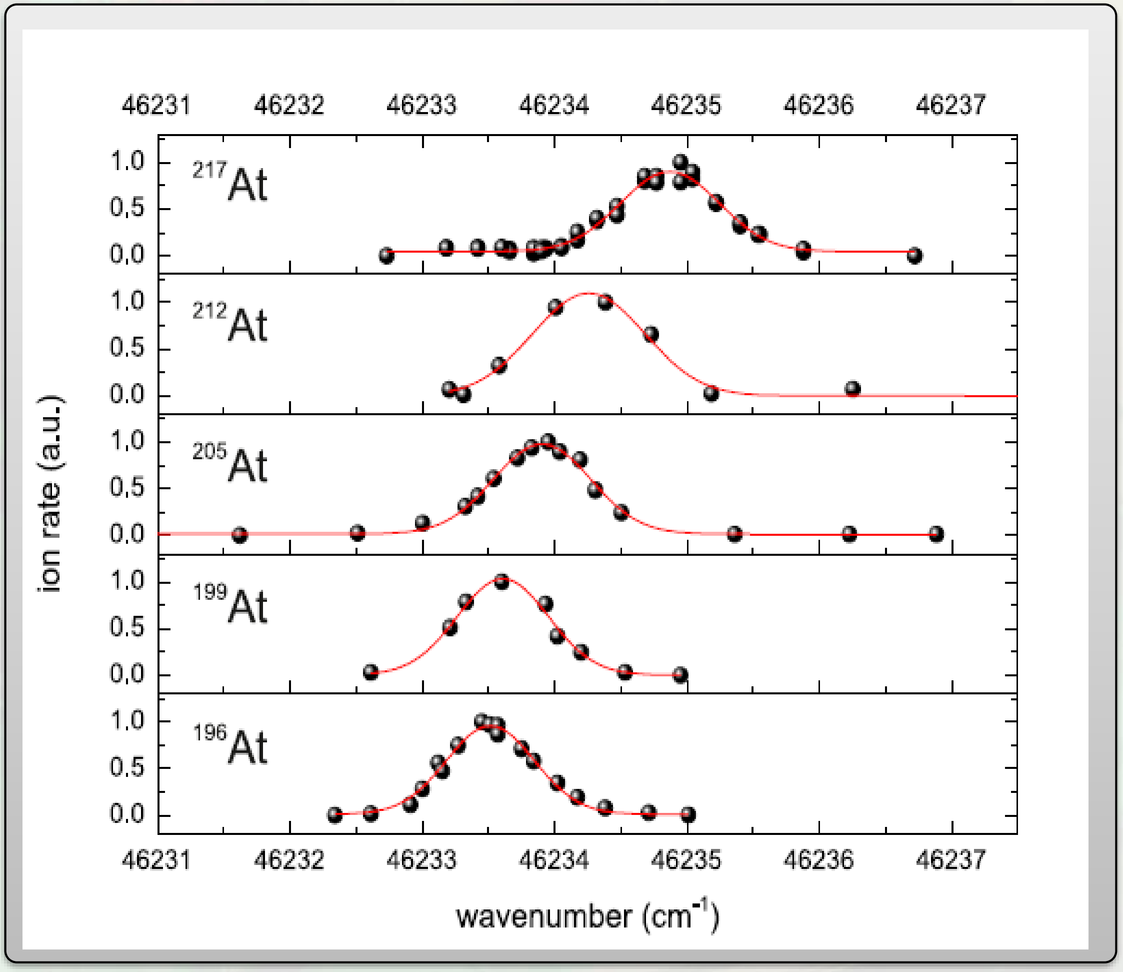


Commissioning of the collinear laser spectroscopy facility BECOLA at NSCL/MSU,
K. Minamisono et al., Hyp. Int. 230, 57-63 (2015)

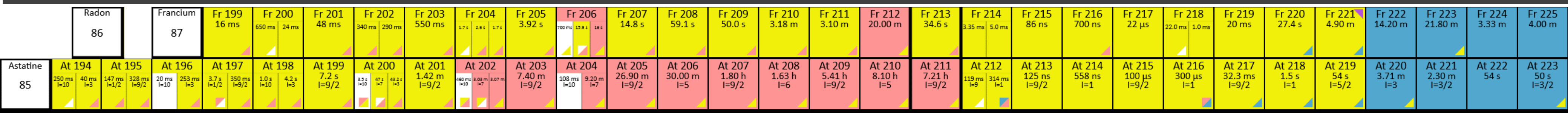
Alternative Approaches? Mid-Resolution In-Source Spec.

- Resolution limited by **Doppler broadening** in the hot ion source and lasers **(FWHM ≈ 15 GHz)**
- **In-RILIS Spectroscopy** for **heavy elements** with large isotope shift & hyperfine structure
- Direct in-source laser spectroscopy on hyperfine structure & isotope shift of ^{196,199,205,212,217}**At**

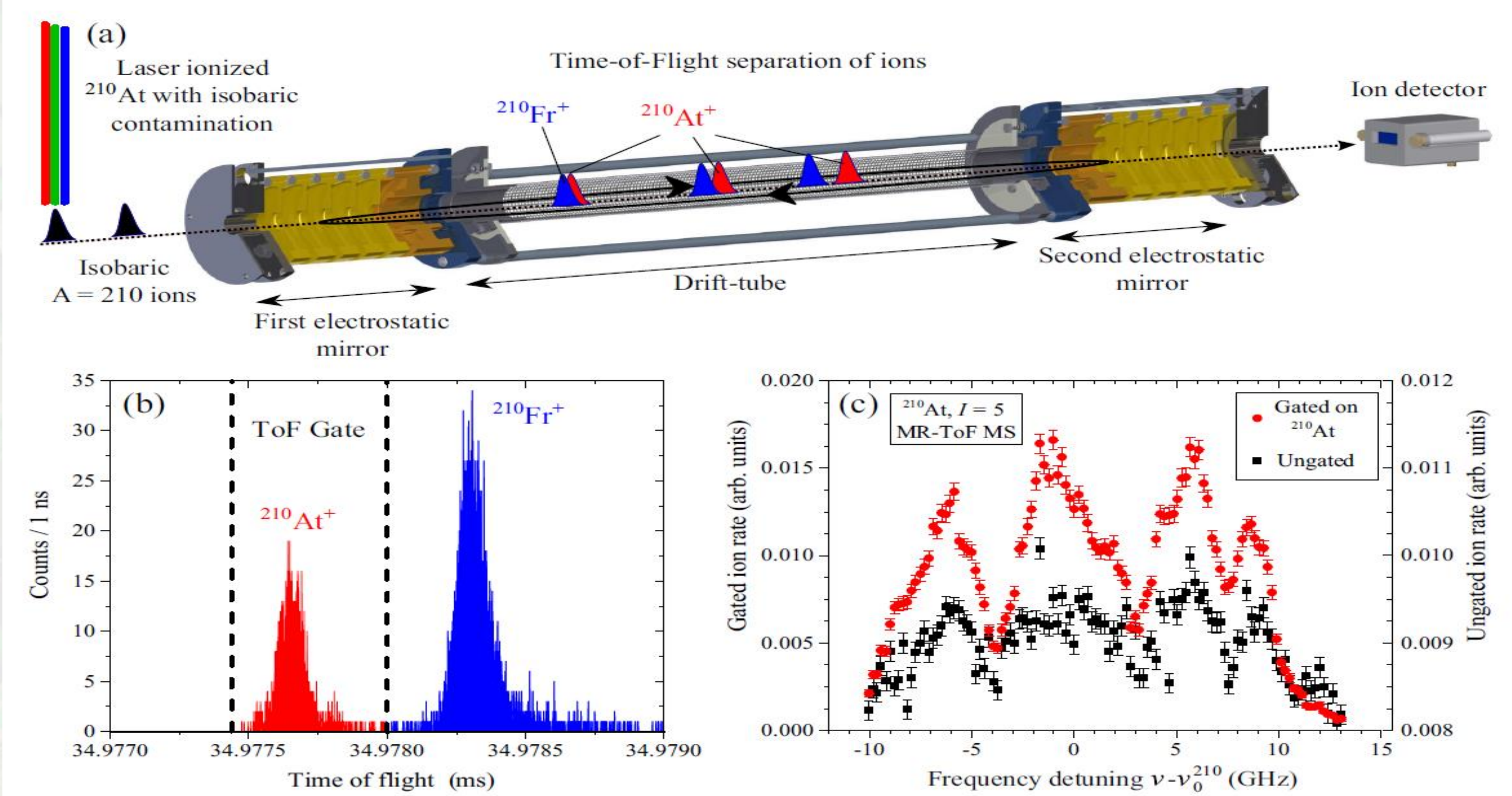




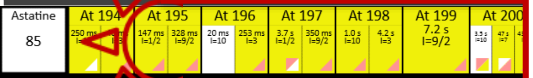
Suppression of Contaminations in In-Source Spectroscopy



J. G. CUBISS *et al.* PHYSICAL REVIEW C **97**, 054327 (2018)



Nuclear Ground State Properties of At from In-RILIS Spec



CHARGE RADII AND ELECTROMAGNE

TABLE I. Measured values of the hyper the fitting of the hfs data are given in round of A-constants ratio is added. The values of

Nucleus	I^π	$T_{1/2}$
^{195}At^g	$(1/2^+)$	290(20) ms
^{195}At^m	$(7/2^-)$	143(3) ms
^{196}At	(3^+)	387(14) ms
^{197}At^g	$(9/2^-)$	381(6) ms
^{197}At^m	$(1/2^+)$	2.0(2) s
^{198}At^g	(3^+)	4.1(3) s
^{198}At^m	(10^-)	1.03(15) s
^{199}At^g	$(9/2^-)$	6.92(13) s
^{199}At^m	$(1/2^+)$	310(80) ms
^{200}At^g	(3^+)	43(1) s
^{200}At^{m1}	(7^+)	47(1) s
^{200}At^{m2}	(10^-)	3.5(1) s
^{201}At	$(9/2^-)$	83(2) s
^{202}At^g	(3^+)	184(1) s
^{202}At^m	(7^+)	182(2) s
^{203}At	$9/2^-$	7.4(2) min
^{204}At	7^+	9.22(13) min
^{205}At	$9/2^-$	26.9(8) min
^{206}At	(6^+)[a]	30.6(8) min
^{207}At	$9/2^-$	1.80(4) h
^{208}At	6^+	1.63(3) h
^{209}At	$9/2^-$	5.41(5) h
^{210}At	$(5)^+$	8.1(4) h
^{211}At	$9/2^-$	7.214(7) h

[a]For $I = 5$, $\delta\nu^{205}_{795} = -117(75)$ MHz, $A = -$ … MHz, and $B = -$ … MHz. Spin assignment for … At is discussed in Sec. V A 3.

TABLE II. Extracted values of the change in mean-square charge radii, the mean-square deformation extracted from the IS data (see Sec. IV C), and the magnetic and quadrupole moments for astatine isotopes. Errors due to the statistical uncertainties in the extracted hyperfine parameters in Table I are given in round brackets. Systematic uncertainties are given in curly brackets, in $\delta\langle r^2\rangle$, stemming from the theoretical indeterminacy of the F and M factors for $\delta\langle r^2\rangle_{A,205}$; in μ, due to the uncertainty in $\mu_{\rm ref}$ and the HFA indeterimnacy; and in Q_S, resulting from the uncertainty in the theoretical B_0/Q_S, and the experimental B_1/B_0 ratios.

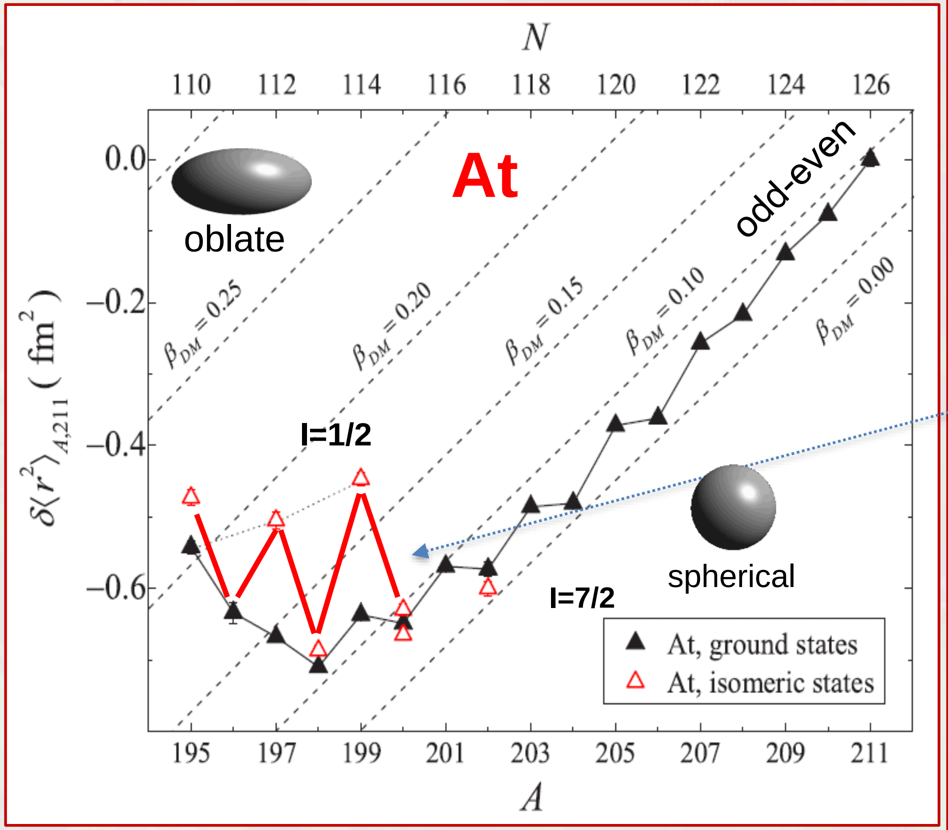
Nucleus	N	I^π	$\delta\langle r^2\rangle_{A,205}$ (fm^2)	β_{DM}	μ (μ_N)	Q_S (b)
^{195}At^g	110	$(1/2^+)$	−0.171(7){9}	0.21(2)	1.611(25){39}	
^{195}At^m	110	$(7/2^-)$	−0.101(7){5}	0.22(2)	3.714(97){90}	−2.04(25){100}
^{196}At	111	(3^+)	−0.262(10){13}	0.17(2)	3.739(110){90}	−0.64(13){35}
^{197}At^g	112	$(9/2^-)$	−0.296(7){15}	0.15(3)	3.849(45){54}	−1.15(8){60}
^{197}At^m	112	$(1/2^+)$	−0.133(7){7}	0.19(2)	1.546(13){37}	
^{198}At^g	113	(3^+)	−0.338(7){17}	0.11(4)	4.037(94){97}	−0.59(15){30}
^{198}At^m	113	(10^+)	−0.315(7){16}	0.12(3)	2.554(81){62}	0.44(25){25}
^{199}At^g	114	$(9/2^-)$	−0.265(7){13}	0.12(3)	3.955(45){56}	−0.95(8){50}
^{199}At^m	114	$(1/2^+)$	−0.075(10){4}	0.17(2)	1.595(38){39}	
^{200}At^g	115	(3^+)	−0.293(7){15}	0.08(4)	4.279(96){110}	−0.50(8){50}
^{200}At^{m1}	115	(7^+)	−0.277(7){14}	0.09(5)	4.74(13){12}	−0.96(12){50}
^{200}At^{m2}	115	(10^-)	−0.258(9){13}	0.10(4)	2.694(82){65}	0.54(25){30}
^{201}At	116	$(9/2^-)$	−0.197(7){10}	0.10(4)	4.025(45){57}	−0.96(15){50}
^{202}At^g	117	(3^+)	−0.229(10){11}	0.04(9)	4.16(12){10}	−0.54(13){30}
^{202}At^m	117	(7^+)	−0.201(10){10}	0.06(6)	4.54(16){11}	−0.65(13){30}
^{203}At	118	$9/2^-$	−0.115(7){6}	0.08(5)	4.021(45){57}	−0.73(8){35}
^{204}At	119	7^+	−0.109(7){5}	0.05(8)	4.84(13){12}	−0.62(8){30}
^{205}At	120	$9/2^-$	0	0.08(4)	4.111(34){58}	−0.61(8){30}
^{206}At	121	(6^+)[a]	0.020(7){1}	0.06(6)	4.39(13){11}	−0.42(10){20}
^{207}At	122	$9/2^-$	0.115(7){6}	0.08(4)	4.150(45){59}	−0.45(8){25}
^{208}At	123	6^+	0.155(7){8}	0.07(4)	4.48(14){11}	−0.40(25){20}
^{209}At	124	$9/2^-$	0.240(7){12}	0.09(3)	4.141(45){59}	−0.40(8){20}
^{210}At	125	$(5)^+$	0.295(7){15}	0.09(3)	4.74(12){11}	−0.42(12){20}
^{211}At	126	$9/2^-$	0.372(9){19}	0.09(2)	4.139(37)[b]	−0.33(12){20}

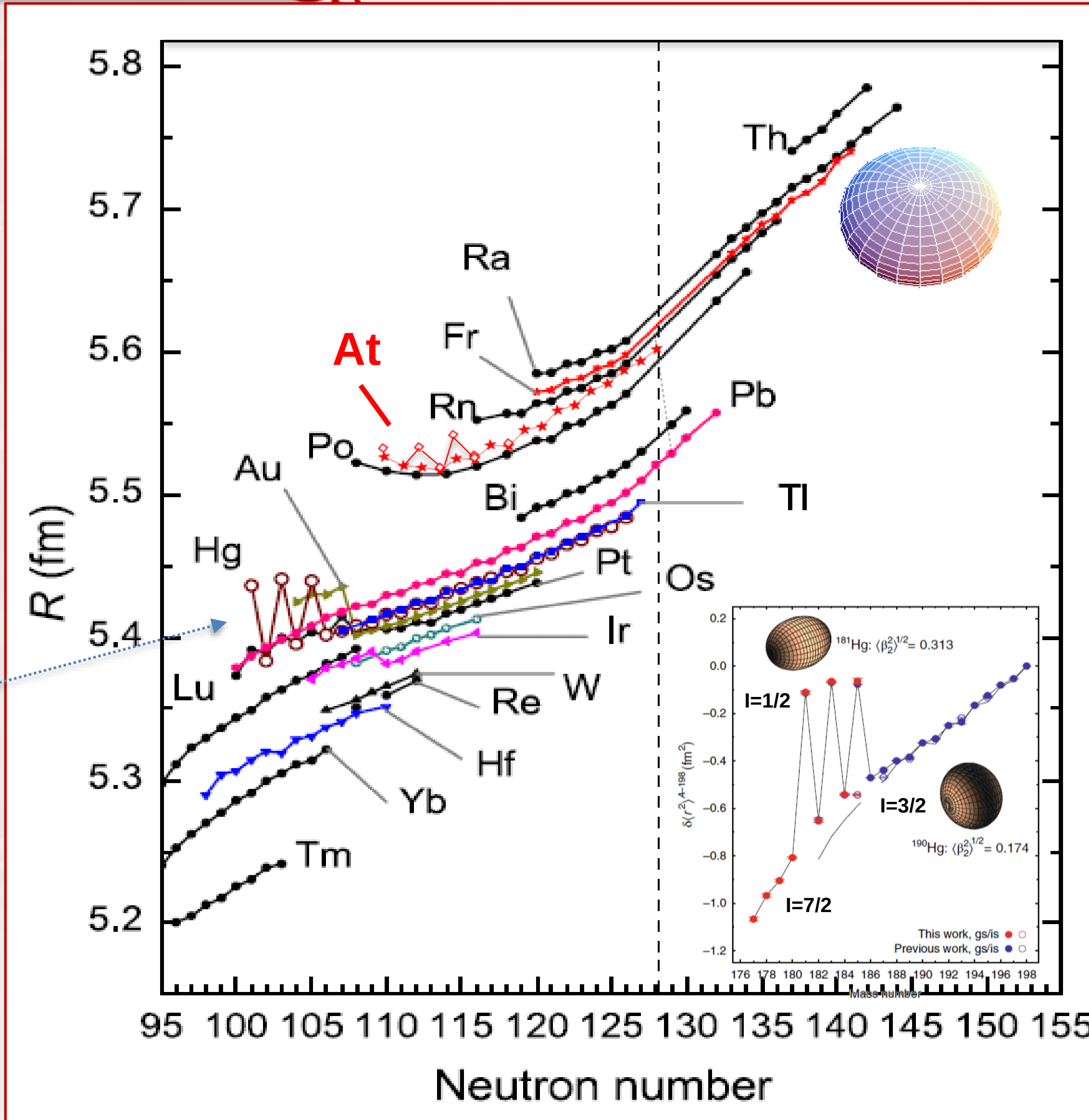
[a]For $I = 5$, $\delta\langle r^2\rangle_{A,205} = 0.009(7)\{1\}$ fm^2, $\mu = 4.34(12)\{11\}\,\mu_N$, $Q_S = -0.29(10)\{15\}$. Spin assignment for ^{206}At is discussed in Sec. V A 3.
[b]Reference value.

Frequency detuning ν-ν_0^{211} (GHz)

Charge Radii and Deformations in Astatine

- Ground state & isomer nuclear charge radii – almost a renaissance of the Hg shape staggering?
- Decrease in odd-even staggering points to onset of octupolar deformed nuclei around N = 145

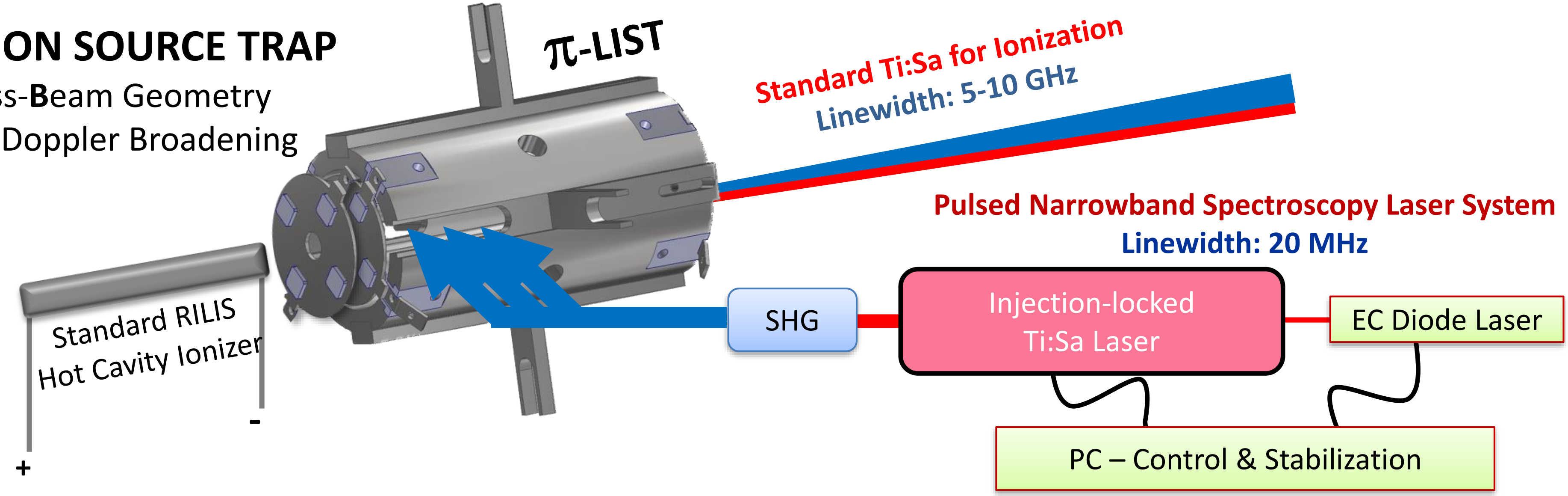




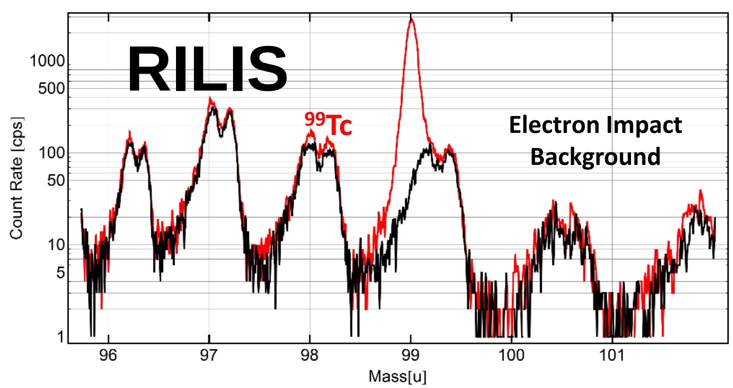
Cubiss, et al., Phys. Rev C 97, 054327 (2018)

Next Generation – High-Resolution In-RILIS/LIST Spec.

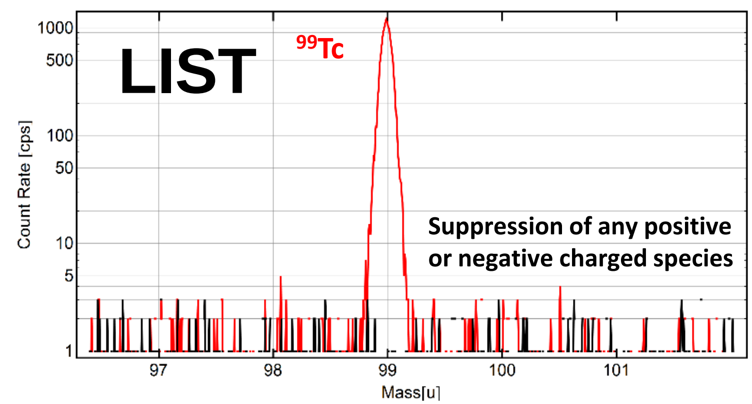
LASER ION SOURCE TRAP
with **C**ross-**B**eam Geometry
prevents Doppler Broadening



Ultimate Background Suppression by Double Repeller LIST Design

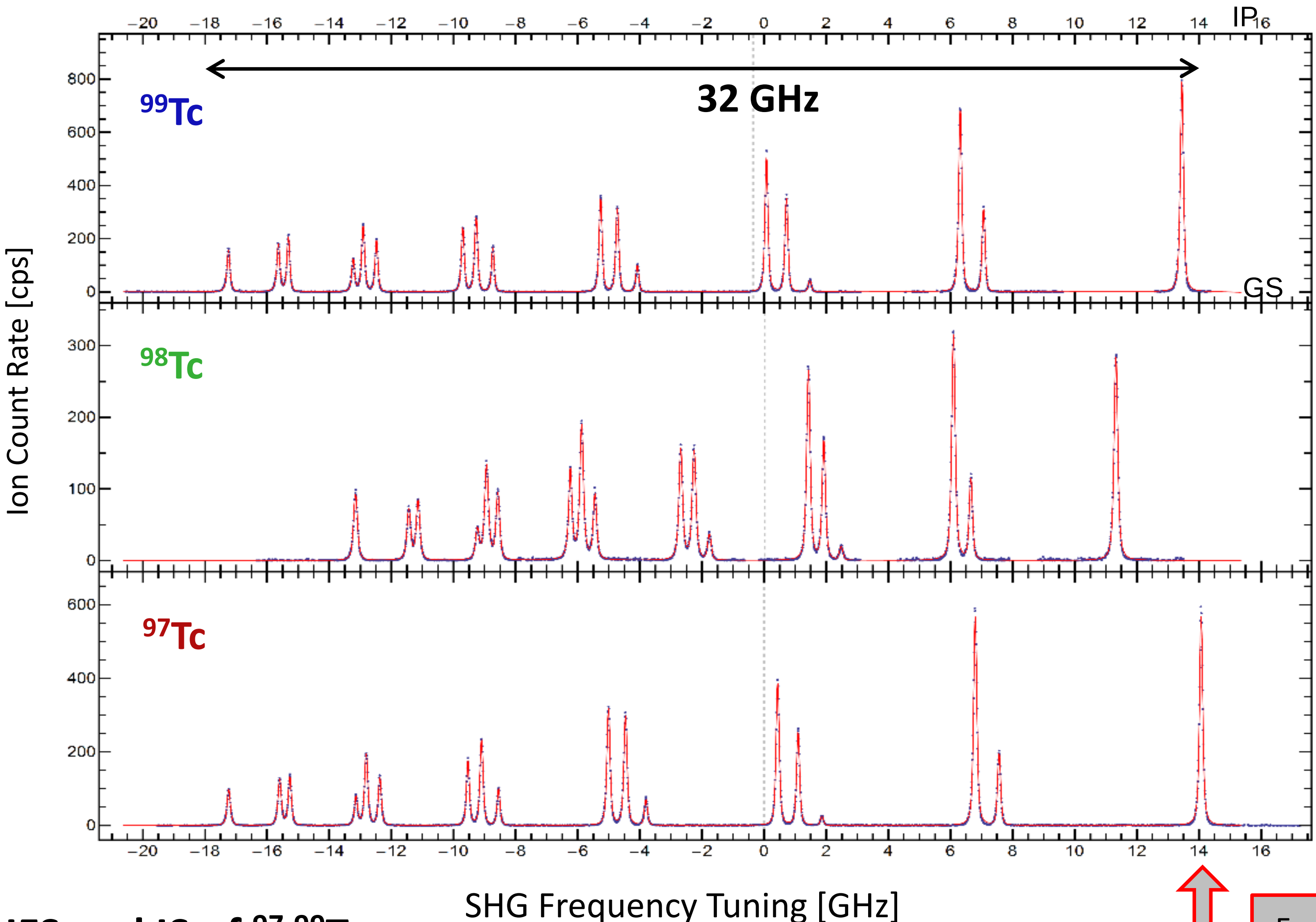


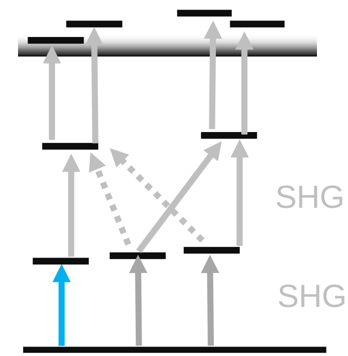
„Background-free“ mass spectrum



π-LIST efficiency estimated to 0.1 - 1 %

High Resolution In-LIST Spectroscopy





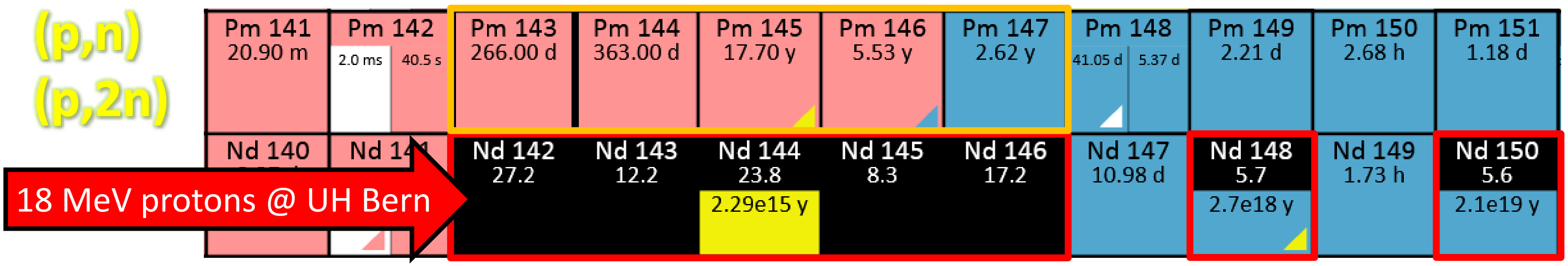
Lowest back-ground < 1 cps

Experimental linewidth ~ 50 MHz

HFS and IS of $^{97-99}$Tc

measured off-line at the RISIKO RIB facility at JGU Mz on samples < 10^{11} atoms in the π-LIST

Off-line High-Resolution Laser Spec on Pm



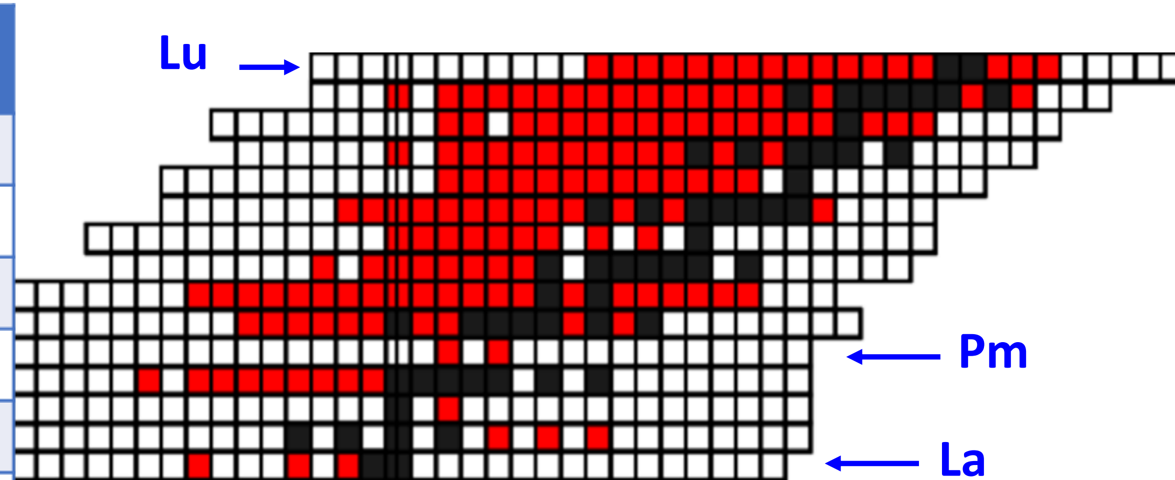
Sample specs from γ spectroscopy

Ratio Pm/Nd ≈ 1/100 after radiochemistry

Isotope	Handling limit (kBq)	A total (kBq)	total atom number
Pm-143	1000	56	2.04 e12
Pm-144	1000	85	3.83 e12
Pm-145	10000	? (no γ)	?
Pm-146	1000	8.5	1.87 e12
Pm-147	10000	? (no γ)	?
Pm-148m	1000	21	1.25 e12

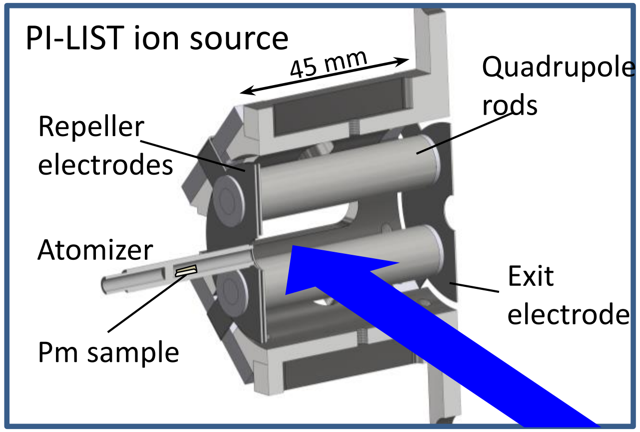
The Nuclear Chart

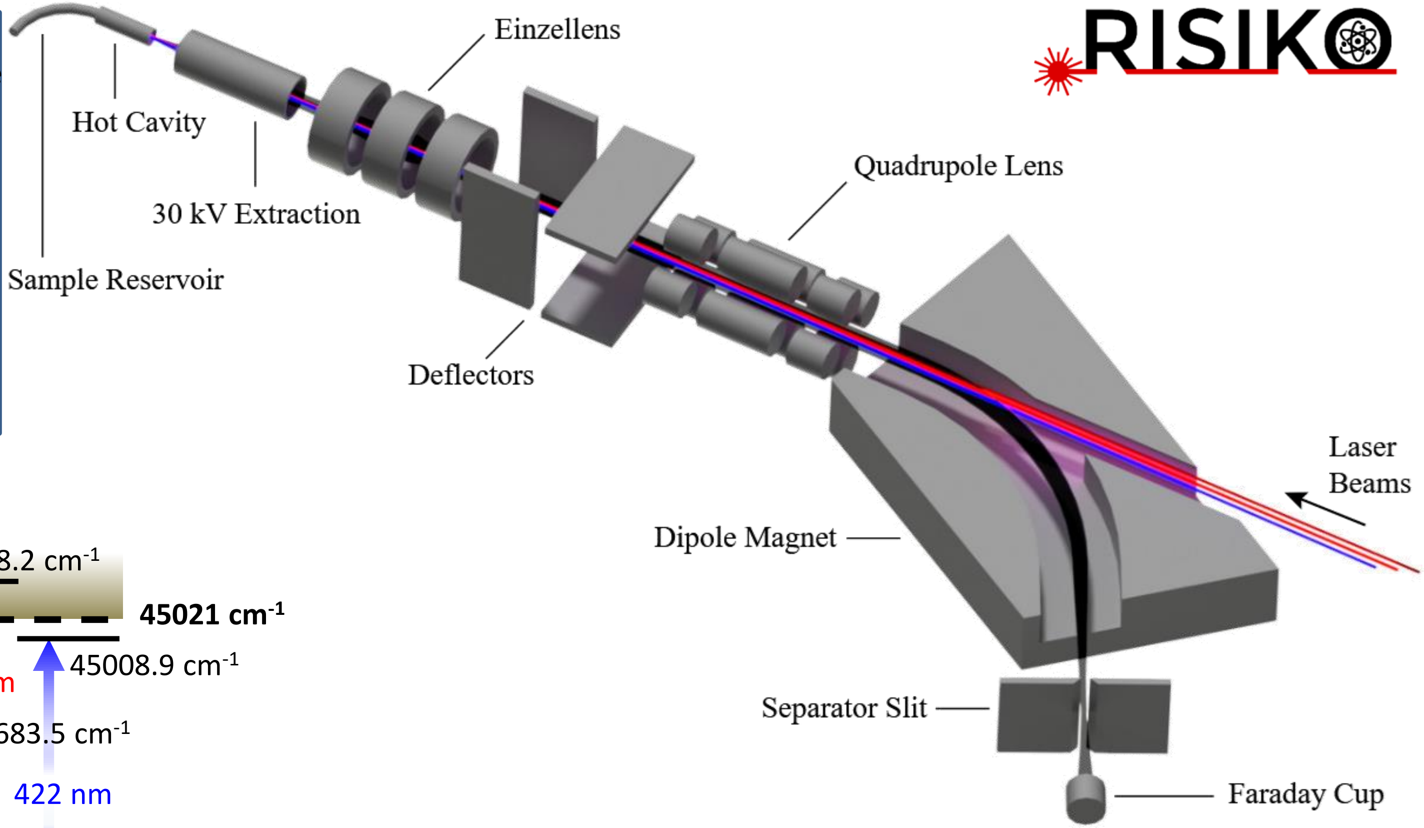
Cutout of the Lanthanides

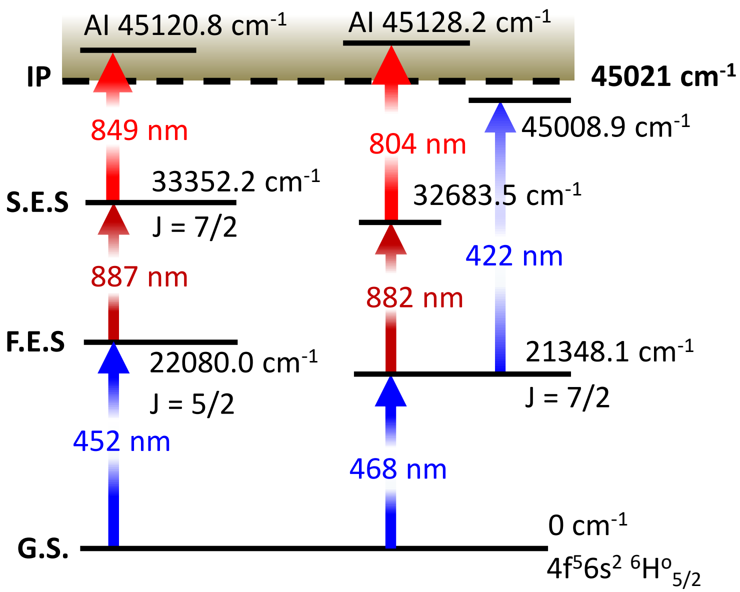


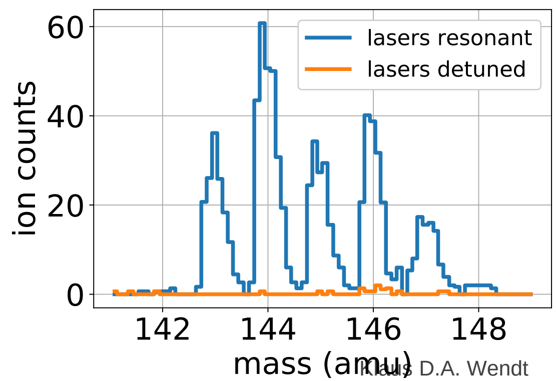
^{145,147}Pm (II): *G.D. Alkhazov et al., J. Phys. B* ***25*** *(1992) 571-576*

High Resolution In-Source Laser Spectroscopy

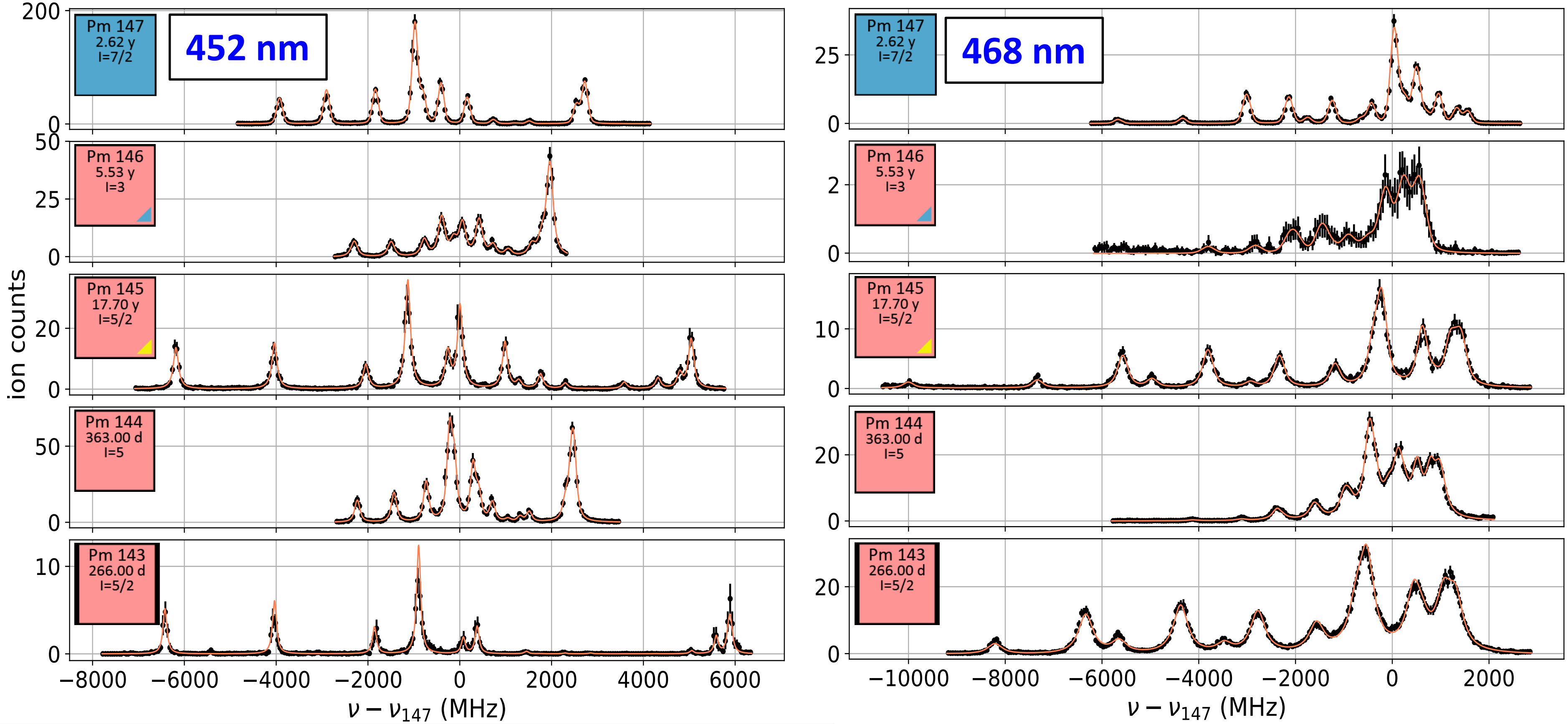








HFS spectra of $^{143\text{-}147}$Pm



Isotope	$\nu - \nu_{147}$ (MHz)	A_{GS} (MHz)	A_{FES} (MHz)	B_{GS} (MHz)	B_{FES} (MHz)
147	0(6)	619.9(2)	499.6(2)	-409(2)	-119(3)
146	225(6)	429.0(3)	345.7(4)	5(3)	1.5(10)
145	347(5)	1255.5(2)	1011.9(1)	-139(2)	-40(1)
144	596(6)	329.0(2)	265.1(2)	127(3)	37(1)
143	736(7)	1368.8(7)	1103.2(6)	-66(6)	-19(2)

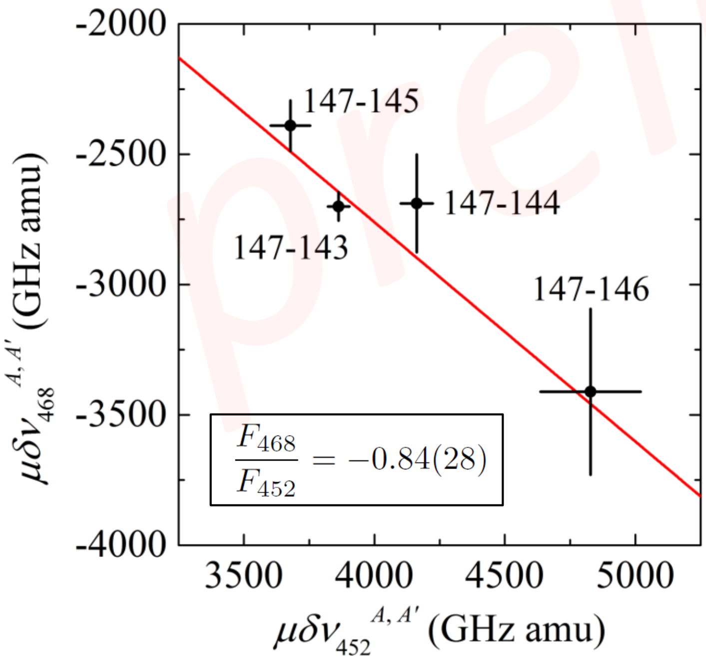
preliminary

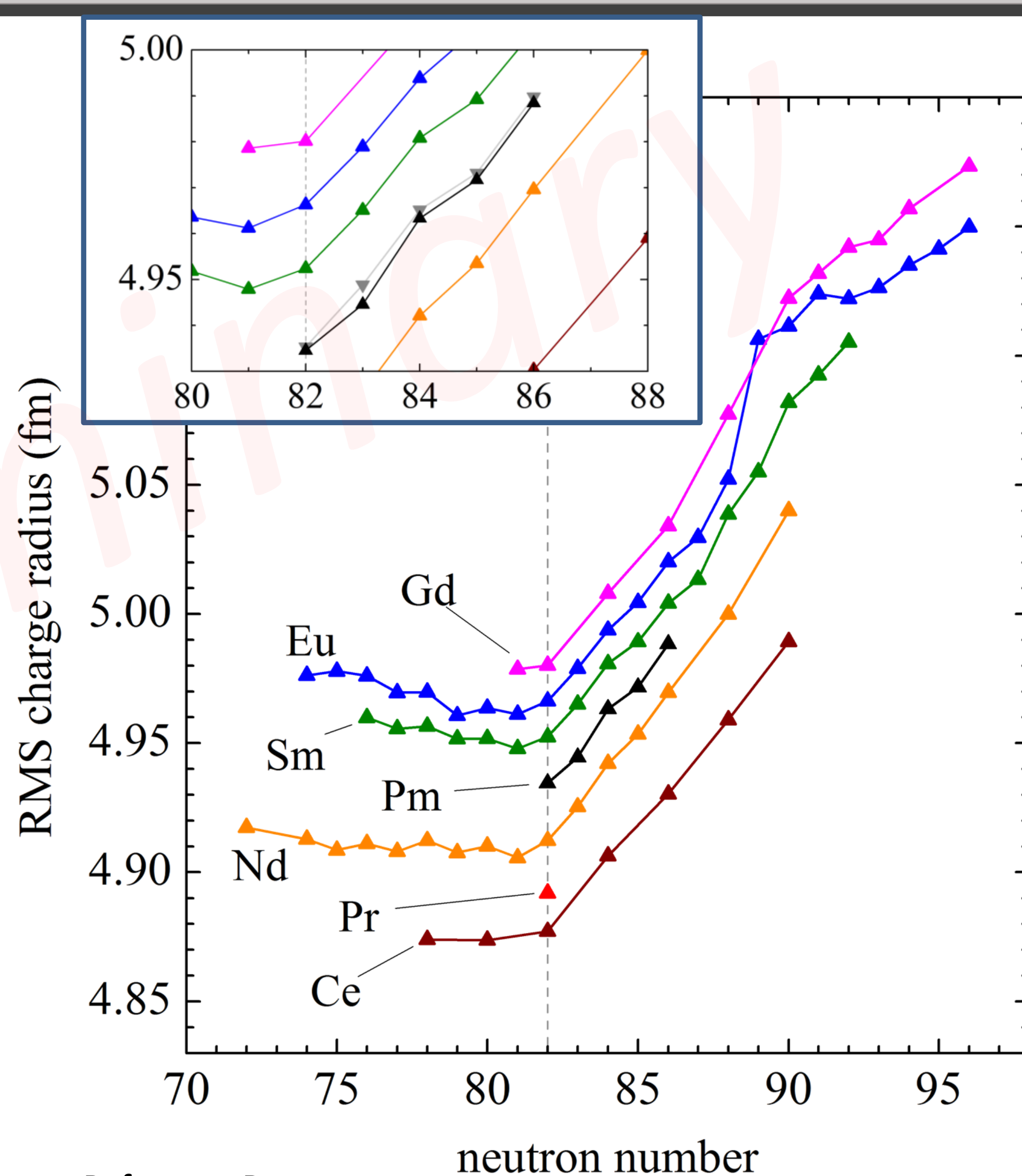
452 nm transition

- exp. linewidth FWHM ~**100 MHz**
- extracted A and B parameters agree for 452 nm and 468 nm transition

Nuclear Charge Radii in Pm

- Electronic factor ratio from King-Plot
- **SMS** neglected, ***F*** interpolated from neighboring elements
- Indication of small odd-even staggering





Reference Data:
I. Angeli, K.P. Marinova / Atomic Data and Nuclear Data Tables 99 (2013) 69–95

Conclusion and Outlook

- **High resolution laser spectroscopy** delivers most valuable **nuclear ground state properties** as well as **atomic physics information** within long isotopic chains up to the most exotic, short-lived isotopes
  - Experimental resolution in the order of natural atomic line widths needed

- Collinear Laser Spectroscopy still is the most versatile technique

- Various sensitive detection techniques are in use
  - **Standard fluorescence detection** still very prosperous after 40 years of use
  - **Nuclear polarization** and **β-NMR detection**
  - **Resonance ionization** enables efficient detection of ions or nuclear decays
  - **Cooler and buncher** combinations for background reduction
  - On-line Collinear **Photodetachment Spectroscopy** for study on exotic negative ions

- **Alternative techniques** found in high resolution **in-source** spectroscopy

Acknowledgements

Thank you for your attention – and these gentlemen and their actual colleagues for their work

Ultra Trace Analysis by Collinear Fast Beam Laser Spectroscopy

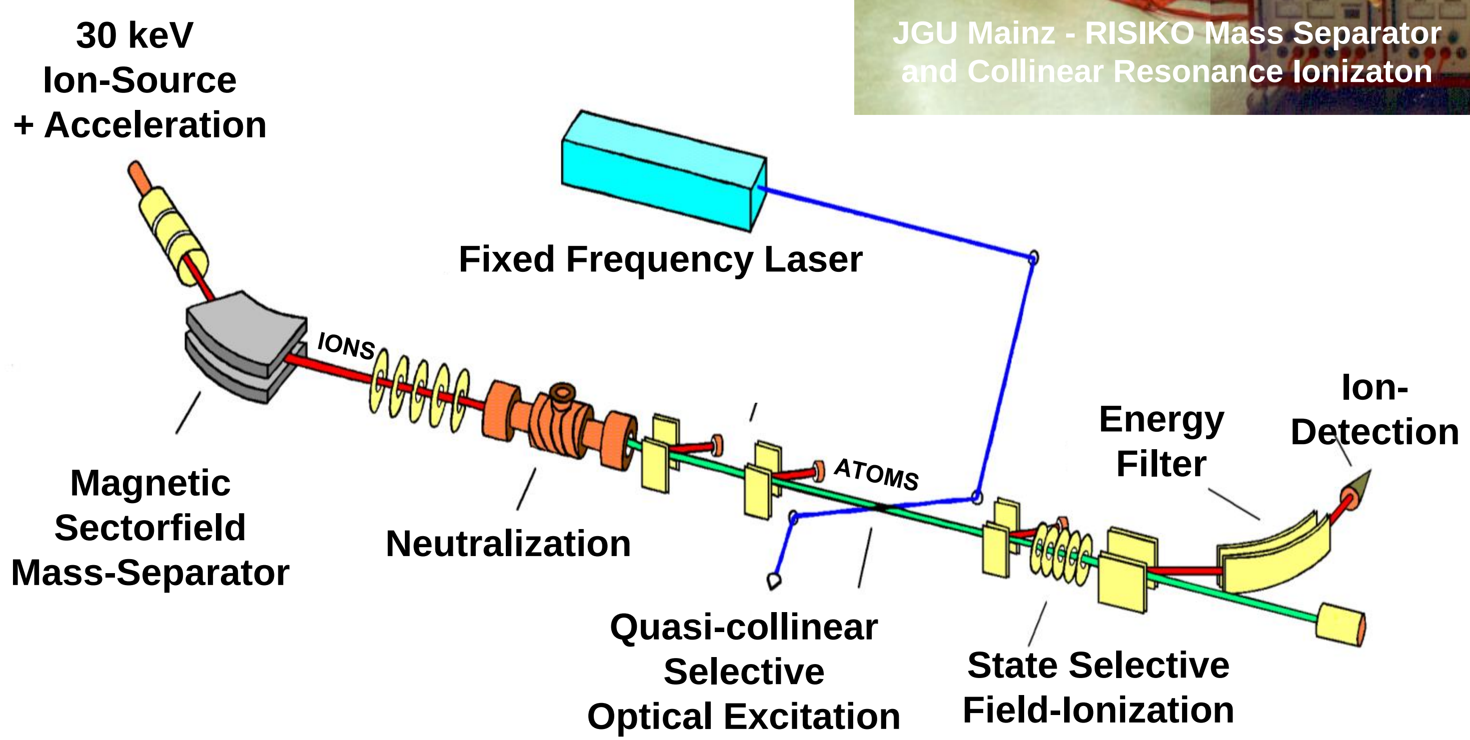
^{89,90}Sr Ultra Trace Analysis by RIMS

Isotopic Selectivity: $\geq 10^{10}$
(background limited)

Overall Efficiency : $\sim 10^{-5}$
(laserpower limited)

Detection Limit : $\sim$ **3 x 10^{6} atoms ^{90}Sr per sample (~ 2 mBq)**

JGU Mainz - RISIKO Mass Separator and Collinear Resonance Ionizaton

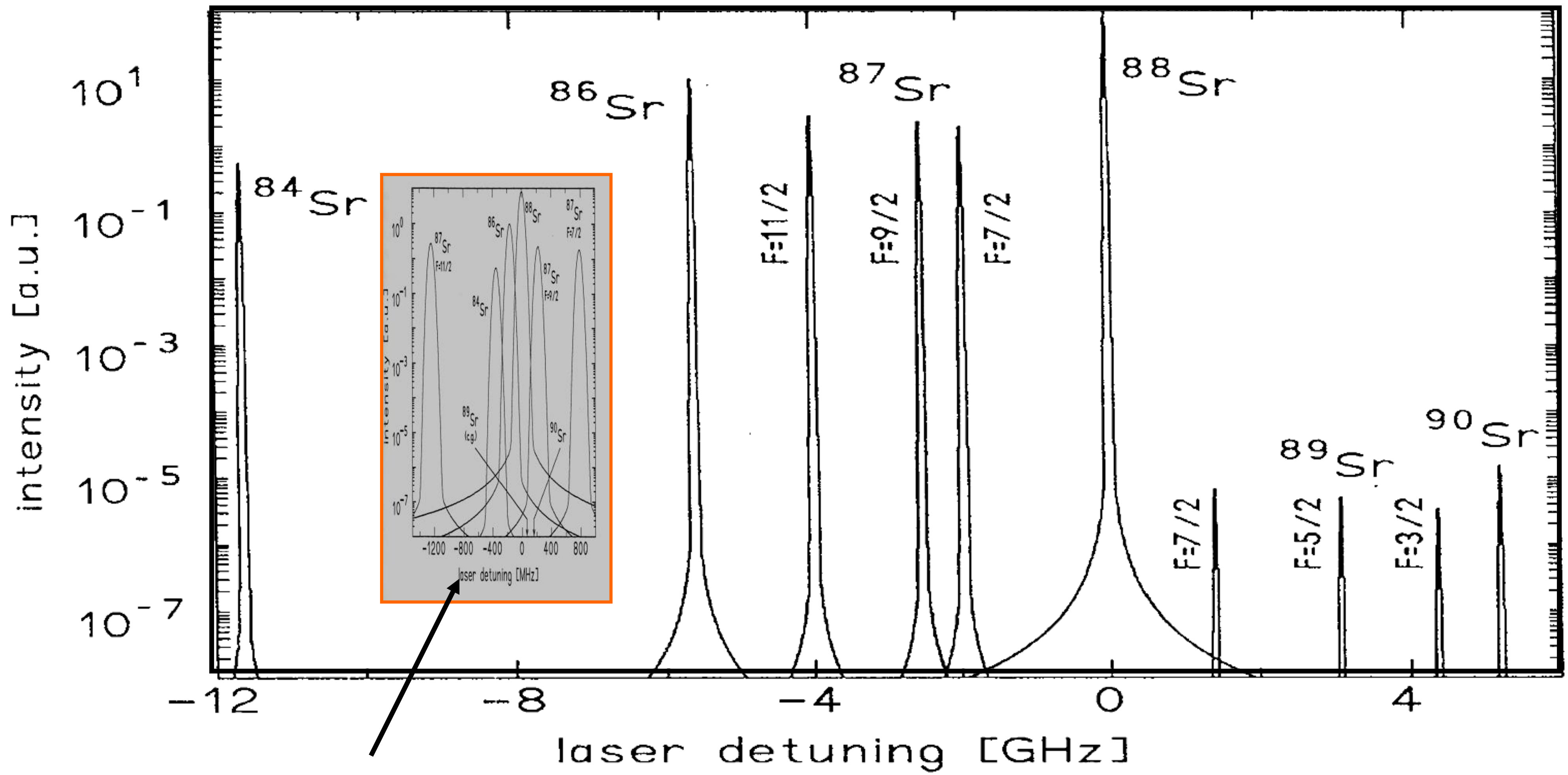


1-Step Quasi-collinear RIS

5s 4d $^3D_J \rightarrow$ 5s 23f ^3F_J (363.8 nm)
+
17 keV field ionization

Klaus D.A. Wendt

Isotope Selection by Collinear Fast Beam Laser Spectroscopy



No isotope resolution - no selectivity

Fast atomic beam (energy 50 keV):

Selectivity > 10^9 per excitation step