



HÜBNER Photonics



UNIVERSITY OF GOTHENBURG

# Laser development towards resonant laser ionization spectroscopy of actinides

LISA Conference, CERN  
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# Motivation

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## Laser Ionization and Spectroscopy of Actinides

Actinides are complicated to study.

- Provide atomic and nuclear structures
- For the heaviest elements, limited by [1]
  - Low production rates
  - Scarce information on atomic levels



[1] M. Block, et al., *Progress in Particle and Nuclear Physics* 116, 103,834 (2021).



# Motivation

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## Laser Ionization and Spectroscopy of Actinides

Lasers need pulses with enough power density, stability, and narrowband operation.

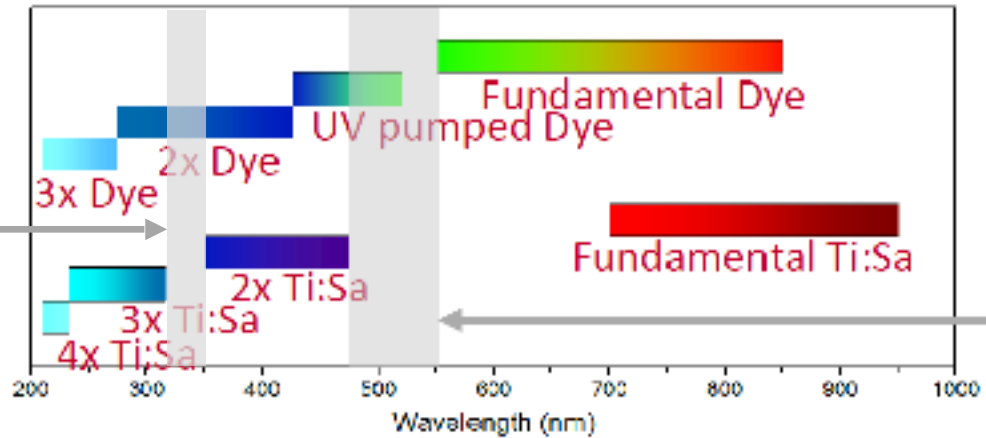
Challenges:

- Produce *difficult* wavelengths
- Optical linewidth  $\ll$  GHz
- Frequency stability



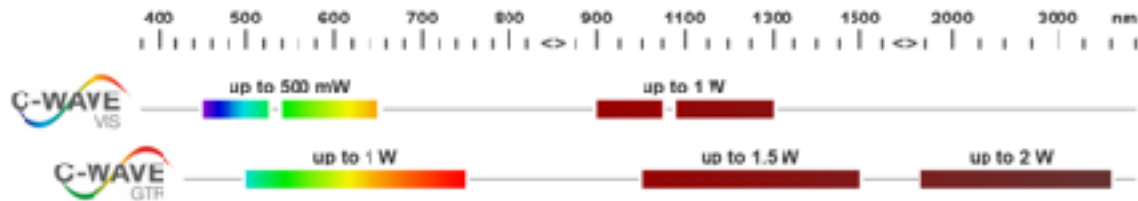
# Spectral coverage at RILIS [5]

Am 28009  $\text{cm}^{-1}$  [2] 357 nm  
 Fm 28185  $\text{cm}^{-1}$  [3], 354.8 nm  
 No 29961  $\text{cm}^{-1}$  [4], 333.8 nm



Dyes are 'complicated'

cw → pulsed amplification → SHG

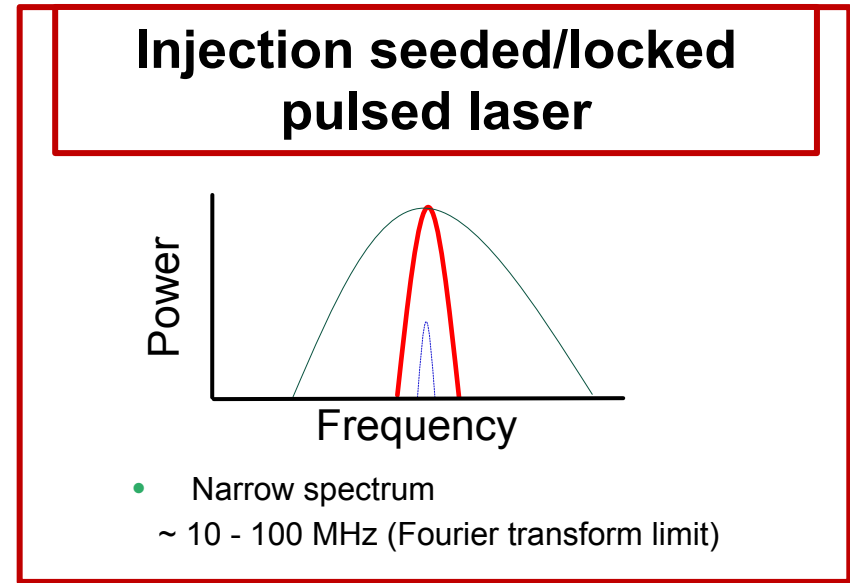
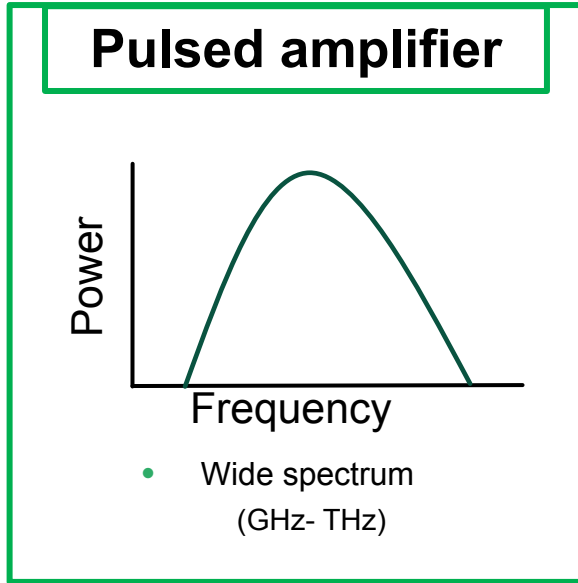
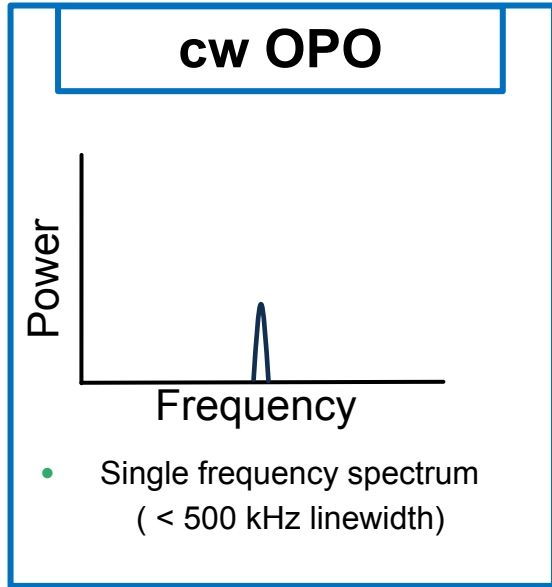


470 - 540 nm is difficult or impossible to access

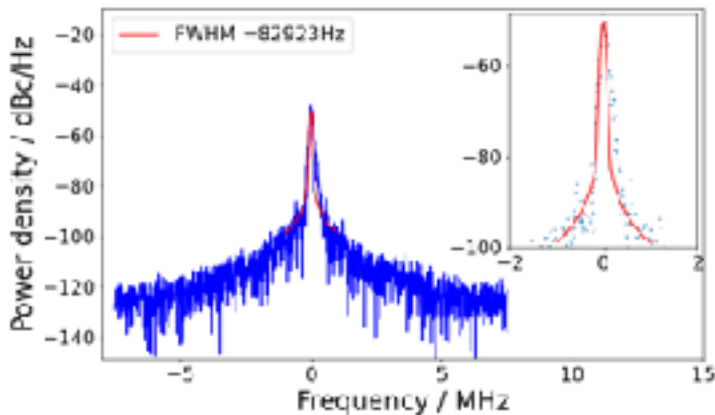
- [2] Fred et al., *J. opt. Soc. Am.*, **47**, 1076-87 (1957).
- [3] Allehabi et al., *JQSRT* **253**, 107137 (2020)
- [4] Laatiaoui et al., *Nature* **538**, 495–498 (2016)
- [5] Marsh, *Leuven Pb-U workshop* (2019)



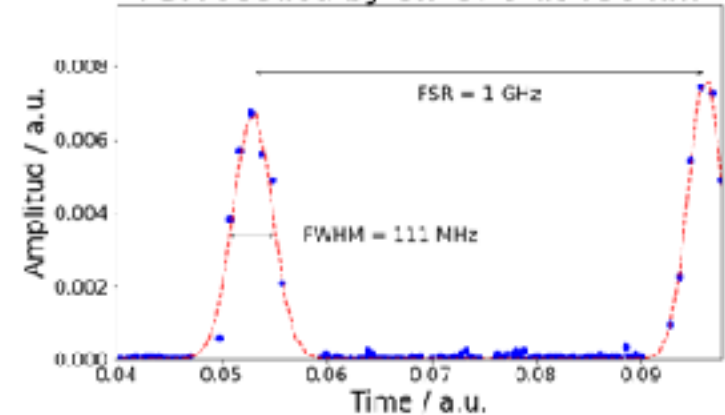
# Laser linewidth at FWHM



OPO linewidth at 656 nm



PDA seeded by cw-OPO at 656 nm

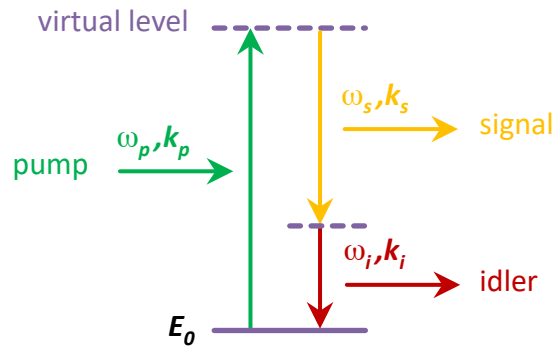


- Dye amplifier
- Ti:Sa cavity
- OPA



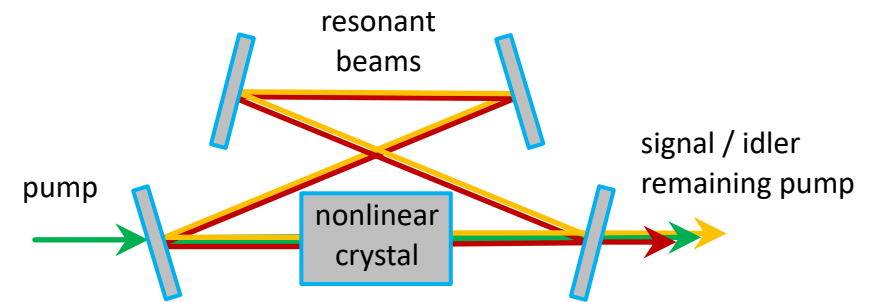
# OPO basics

## PRINCIPLE



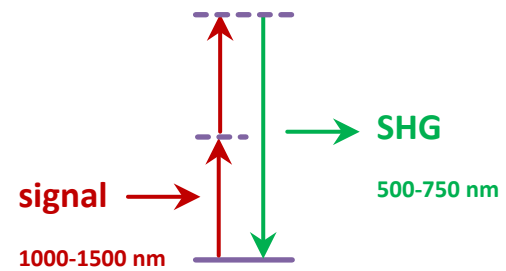
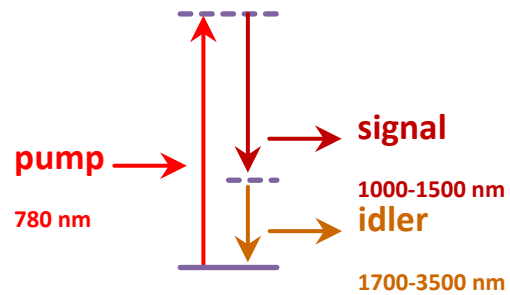
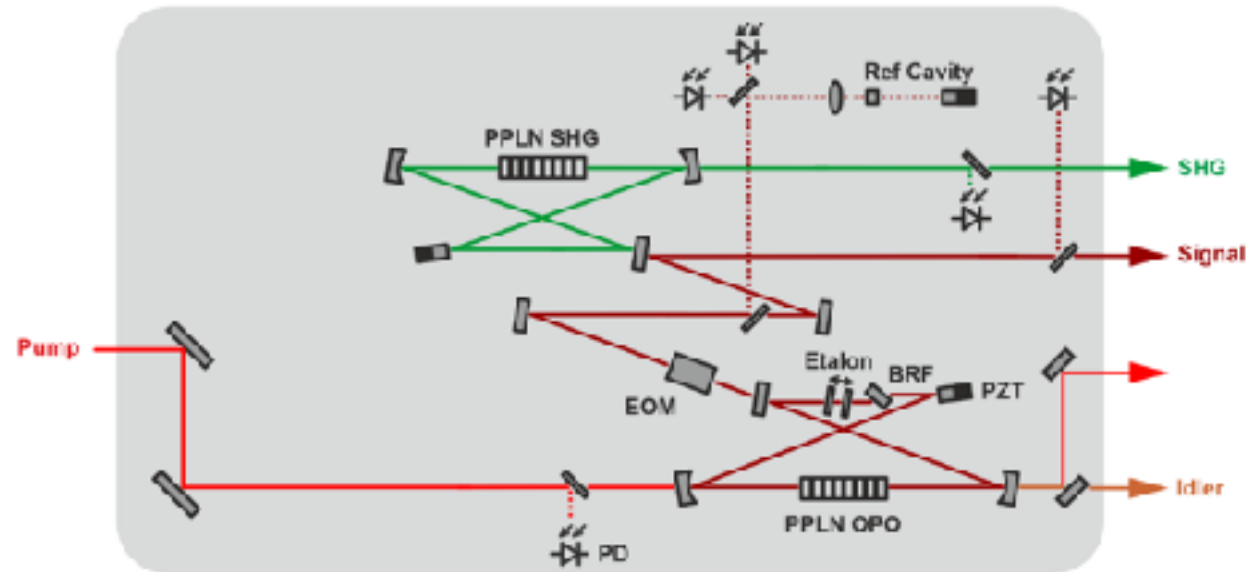
- Three-wave-mixing of pump, signal, and idler
- $\omega_p = \omega_s + \omega_i$
- $k_p = k_s + k_i + \Delta k$

## REALIZATION

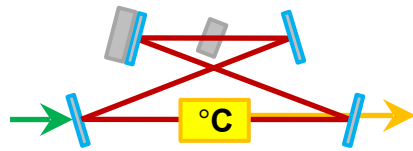


- Source of coherent pump light
- $\chi^{(2)}$  nonlinear medium
- Bow-tie optical resonator

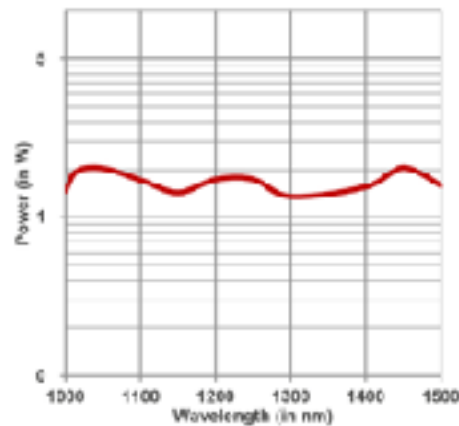
# Beam Path Step-by-Step



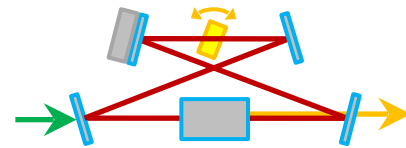
# Wavelength Tuning Mechanisms



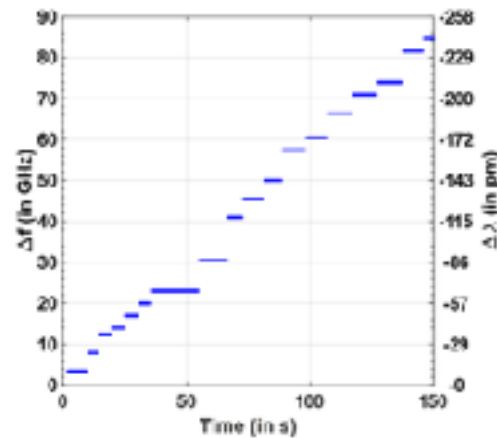
**Coarse tuning**



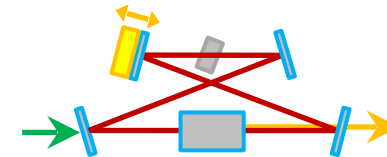
A few nm per centigrade,



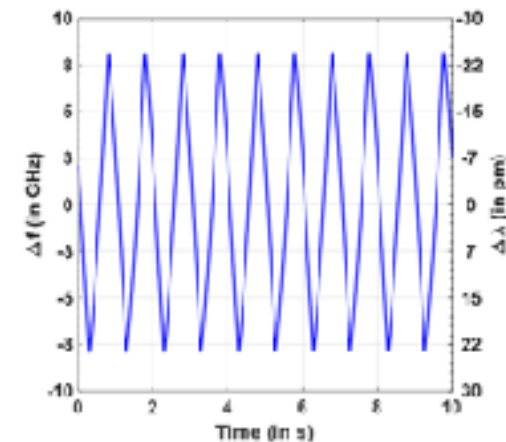
**Stepwise tuning**



up to 100 GHz,



**Continuous tuning**



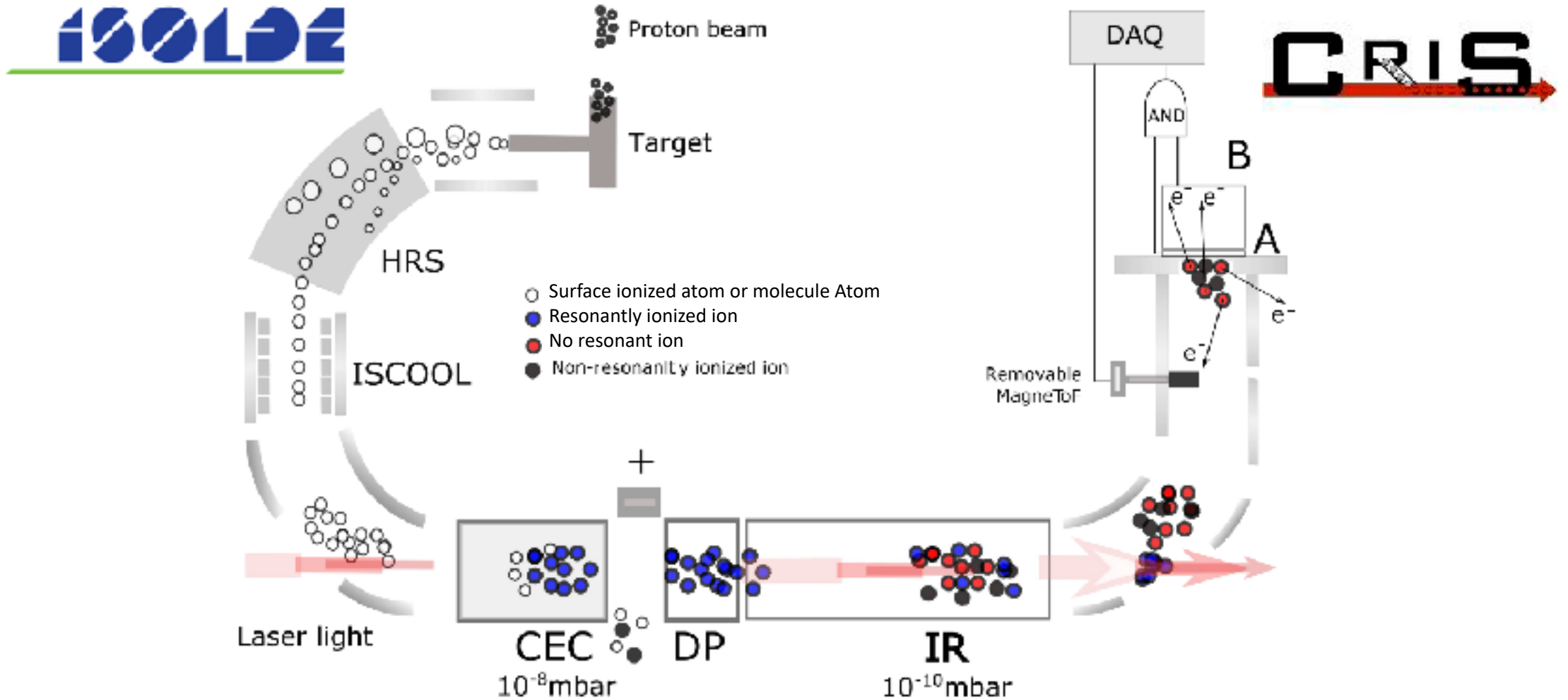
up to 20 GHz per scan.



# First experiment

Benchmark evaluation of an OPO seeded system with silver @ 328 nm

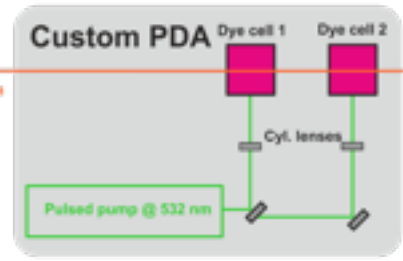
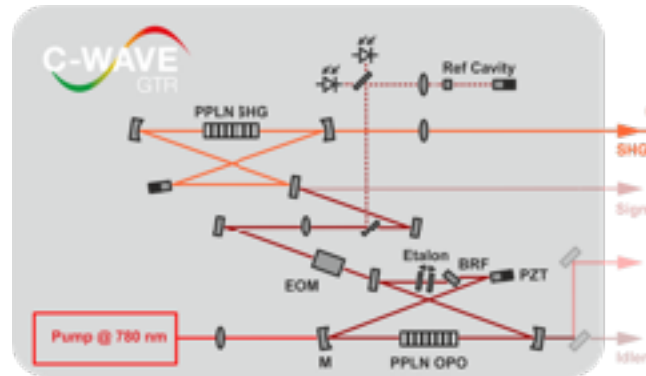
# CRIS setup at ISOLDE, CERN [5]



[5] Koszorús et al., *Nat. Phys.* **17**, 539 (2021).

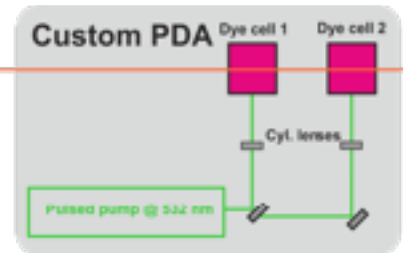
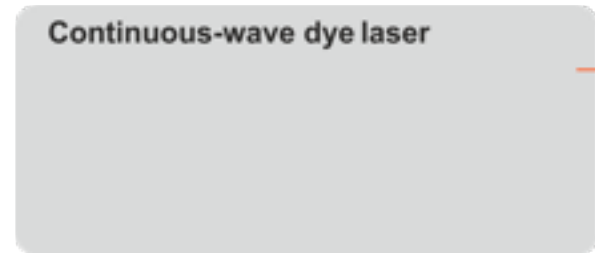
# Laser systems to generate 328 nm pulses

- OPO



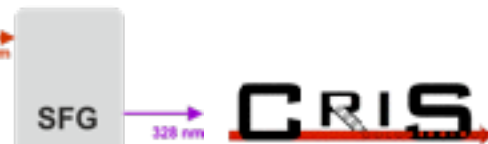
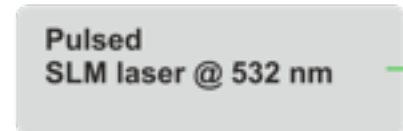
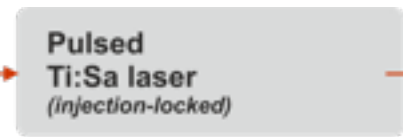
**CRIS**

- Dye

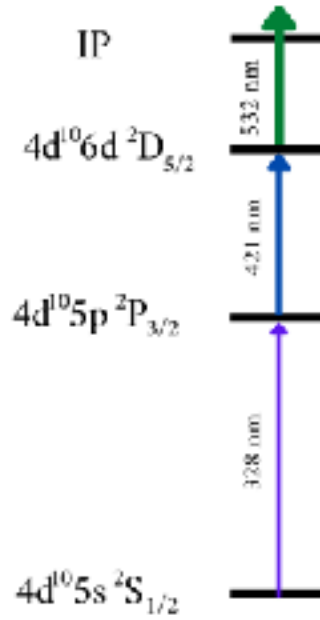


**CRIS**

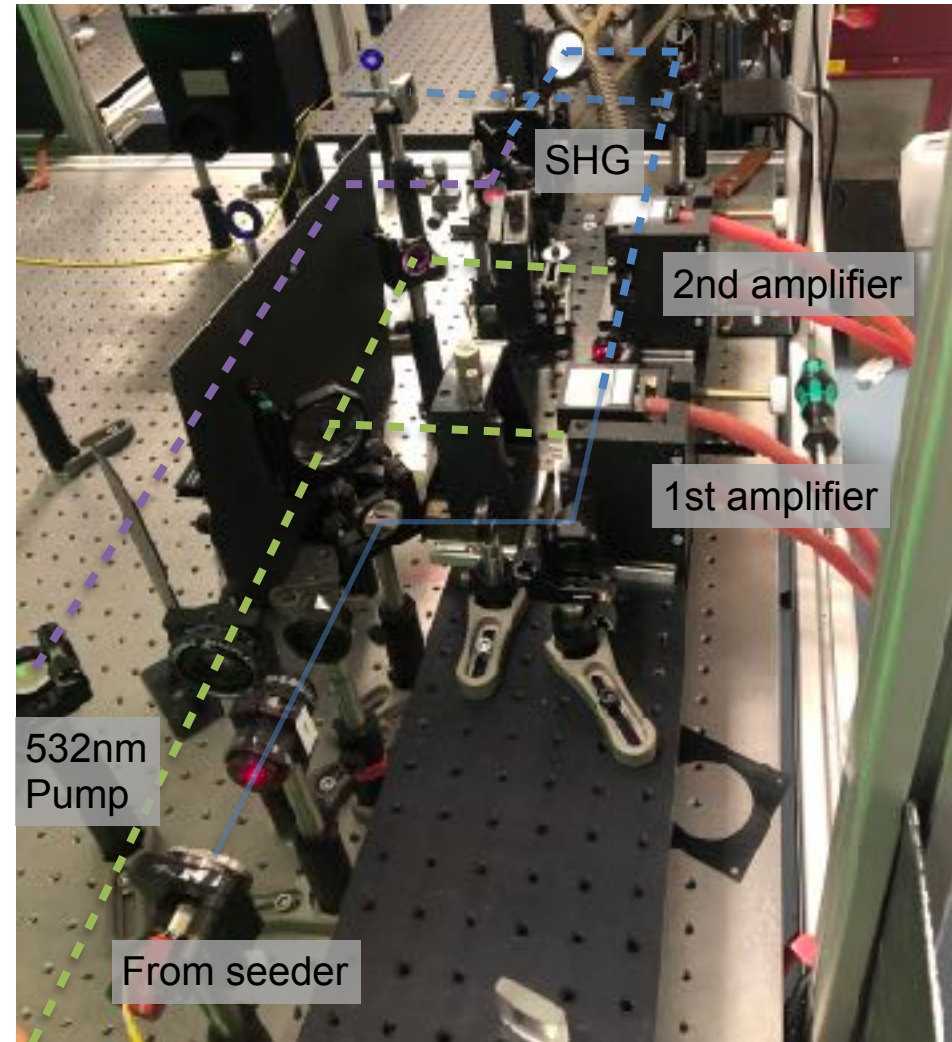
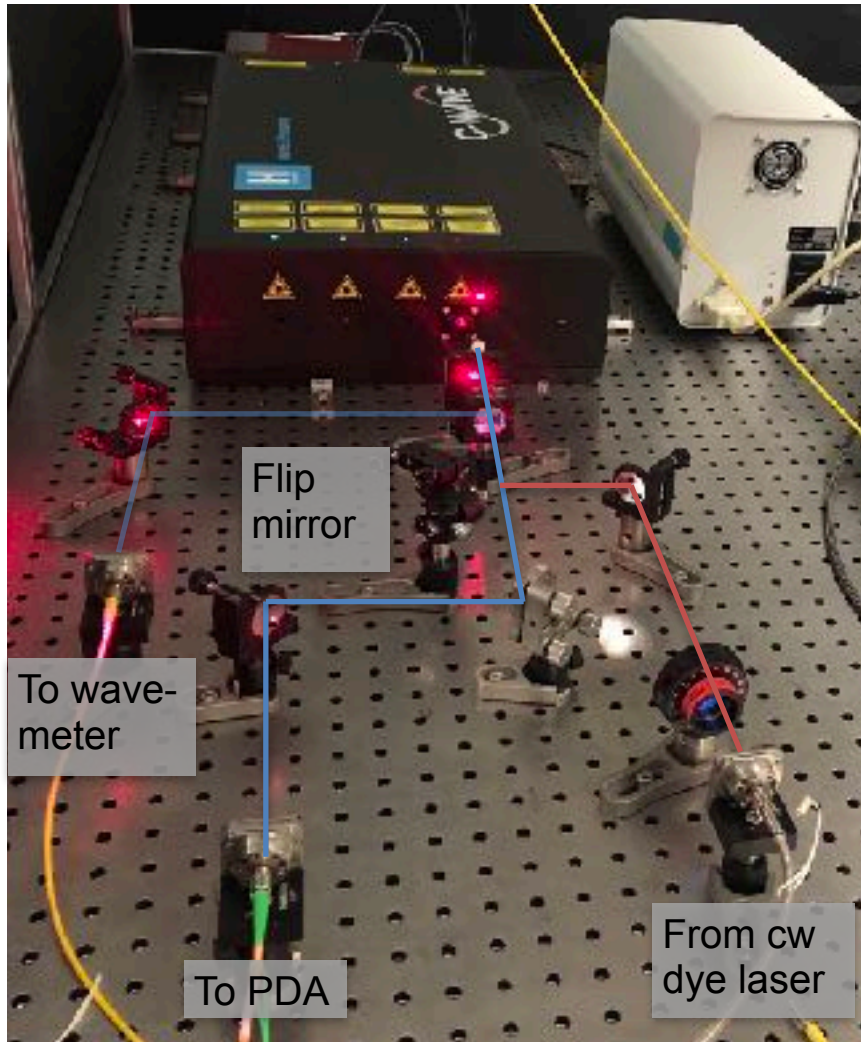
- SFG



**CRIS**

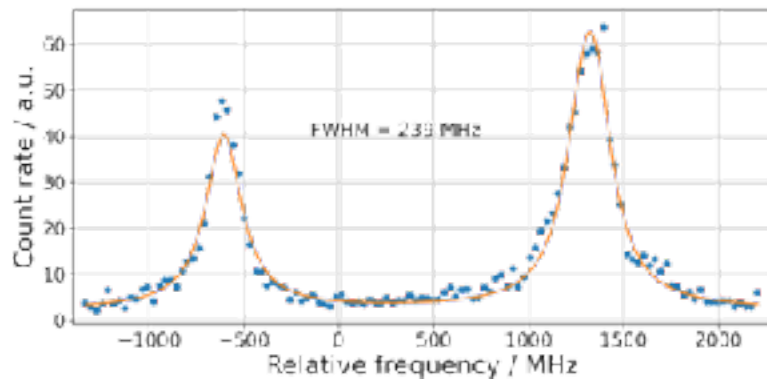


# Experimental setup

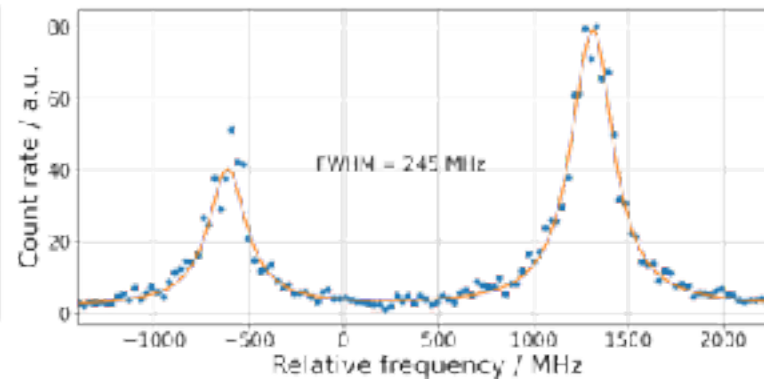


# Benchmarking the OPO system for laser spectroscopy

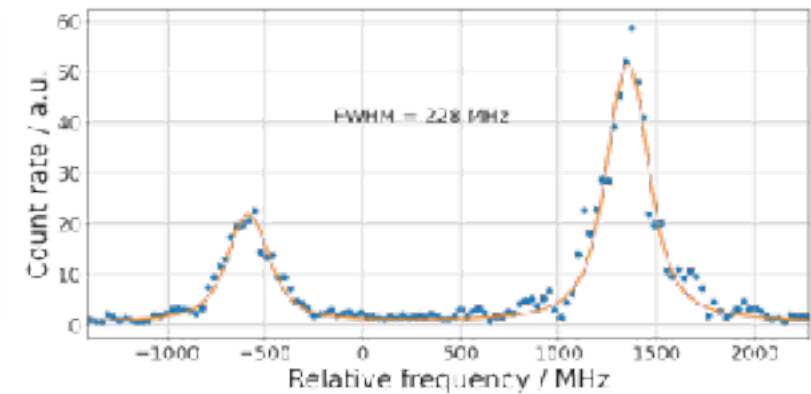
- Compare  $^{109}\text{Ag}$  with three systems
  - Optical and transition linewidth
  - Handling and maintenance



a) HFS with OPO.



b) HFS with Dye.



c) HFS with SFG.

**Optical LW**                      156(17) MHz  
**Transition LW**                239(14) MHz

140(17) MHz  
 245(10) MHz

141(14) MHz  
 228(15) MHz



## Results

Transition linewidth, hf splitting, dipole moment, IS, and change in the mean-squared charge radii

A	FWHM				
109	239(14)	<b>1.959(21)<sup>a</sup></b>	Ref. b	0	0
111	221(5)	2.153(8)	<b>-0.144(2)<sup>c</sup></b>	-436(5)	0.181(27)
111m	188(10)	38.684(8)	4.52(5)	-357(4)	0.162(26)
117	232(17)	2.615(17)	-0.174(3)	<b>-1198(10)<sup>d</sup></b>	<b>0.578(94)<sup>e</sup></b>
117m	<b>173(9)</b>	37.960(2)	4.43(5)	-1113(2)	0.558(91)

## Literature values

a 1976.932075(17) MHz [6]

b  $-0.1306906(2) \mu_N$  [7]

c  $-0.146(2) \mu_N$  [8]

d  $-1181(6)$  MHz [9]

e  $0.568(73) \text{ fm}^2$  [9]

[6] Dahmen et al., *Z. Physik* **200**, 456–466 (1967)

[7] Sahm and Schwenk, *Z. Naturforsch. A* **29**, 1763 (1974)

[8] Woodgate and Hellwarth, *Proc. Phys. Soc. A* **69**, 581 (1956)

[9] Reponen et al., *Nat Commun* **12**, 4596 (2021)

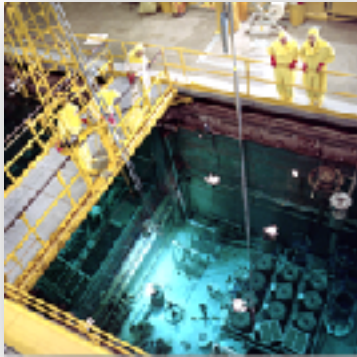
# Second experiment

## High-resolution spectroscopy of Fm-255

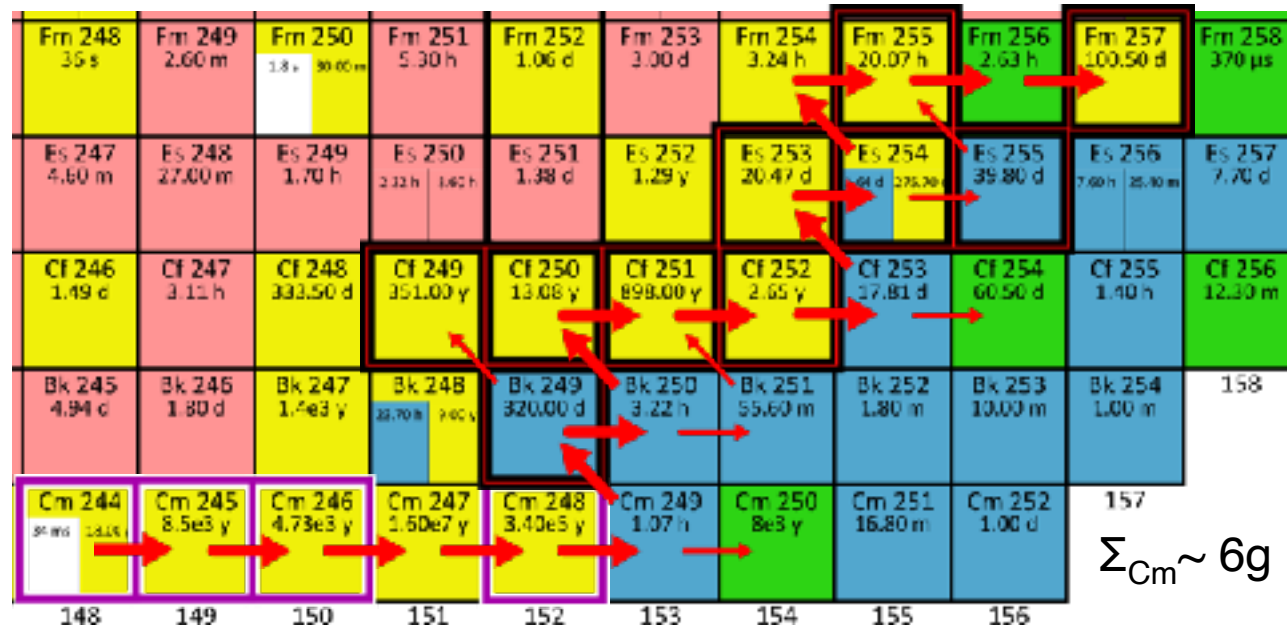
(Attempted) evaluation an OPO injection-locked Ti:sa system with fermium @ 355 nm

# Production of fermium-255

High Flux Isotope Reactor (HFIR) at ORNL



See:  
- Jessica Warbinek  
Tomorrow 10:15



$$\Sigma_{Cm} \sim 6g$$

— Initial nuclides  
— Produced nuclides

High Flux Reactor at ILL

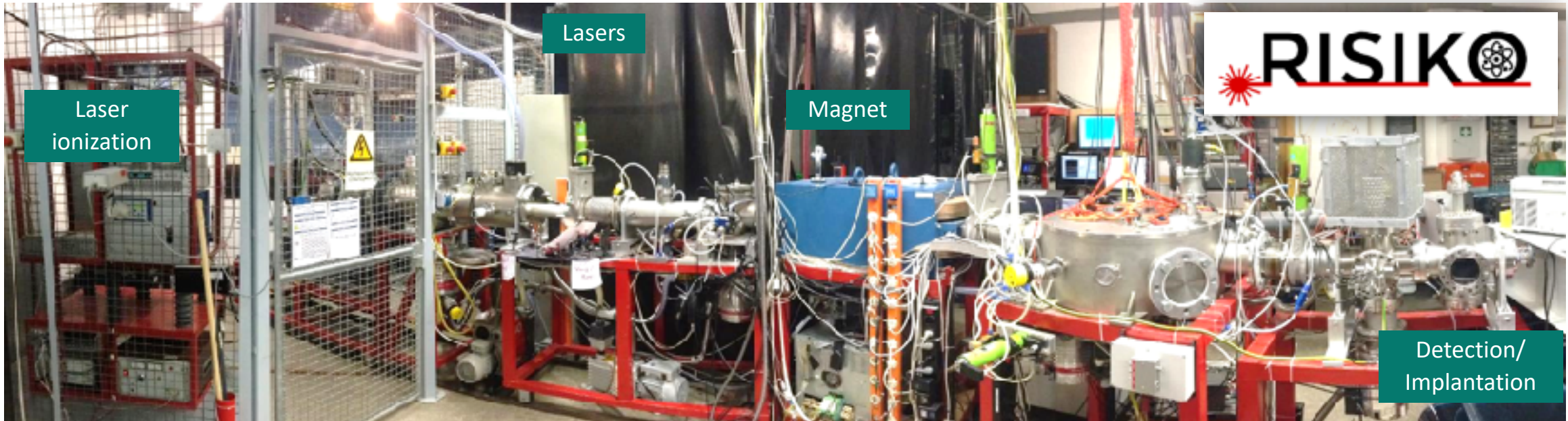


The isotopes used in this research were supplied by the U.S. Department of Energy, Office of Science, by the Isotope Program in the Office of Nuclear Physics. The <sup>253,254,255</sup>Es and <sup>255,257</sup>Fm were provided to Florida State University and the University of Mainz via the Isotope Development and Production for Research and Applications Program through the Radiochemical Engineering and Development Center at Oak Ridge National Laboratory.

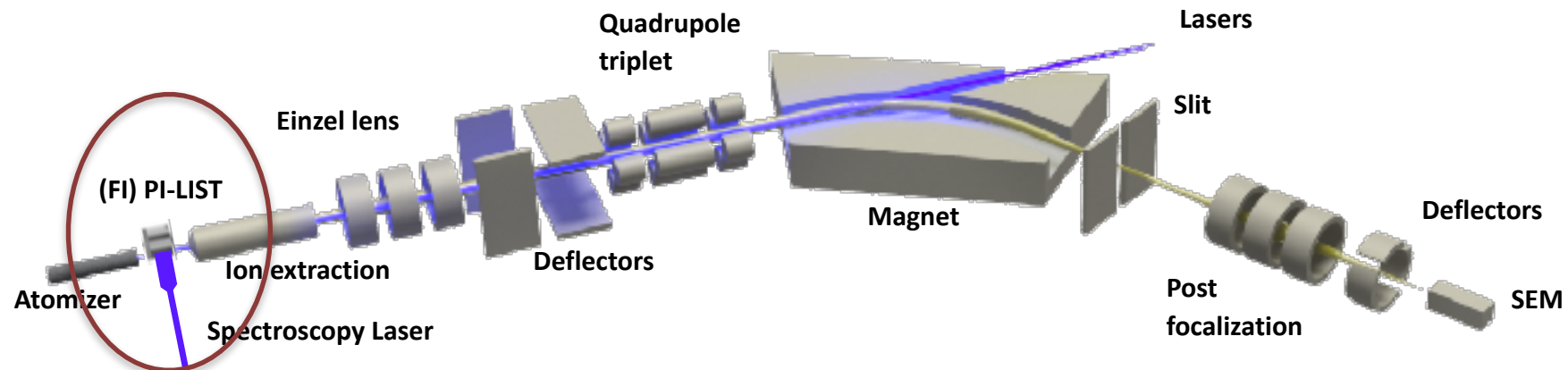




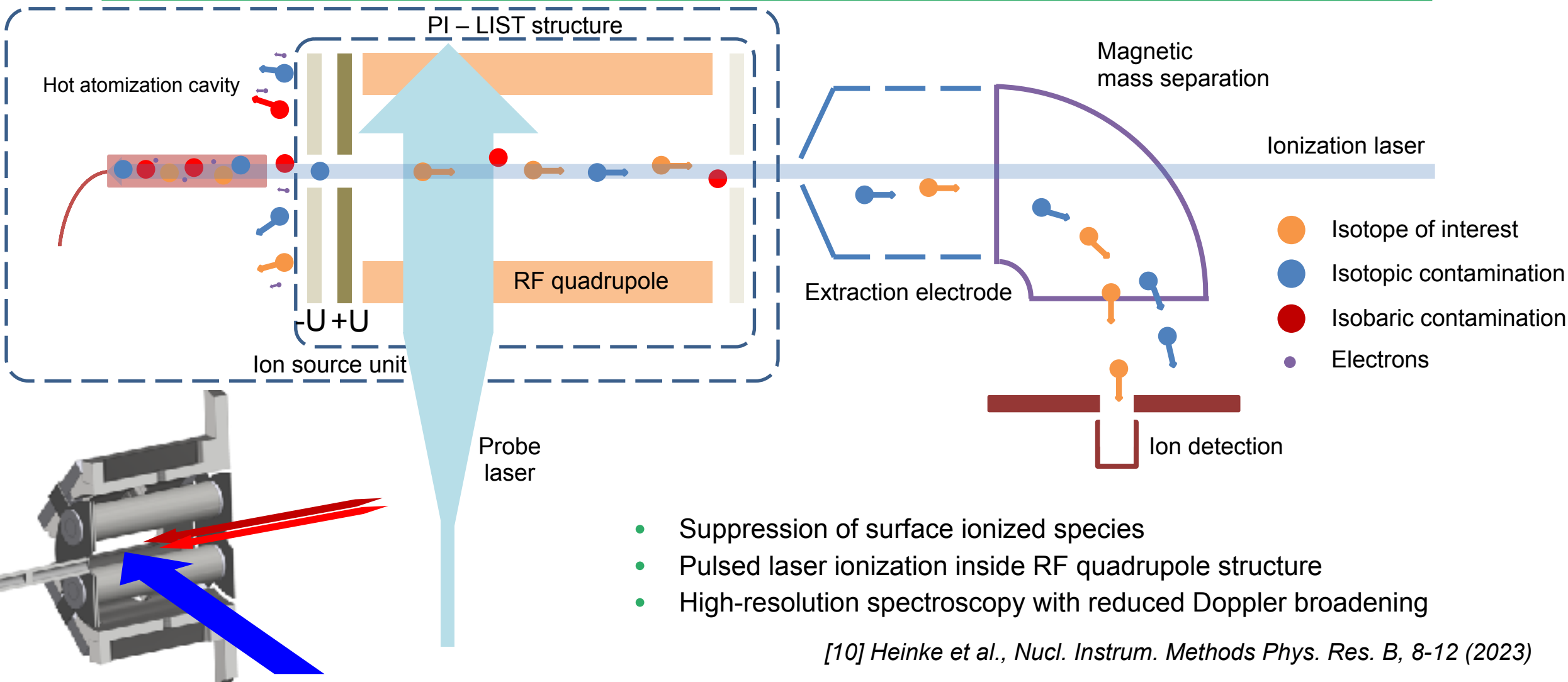
# RISIKO mass separator



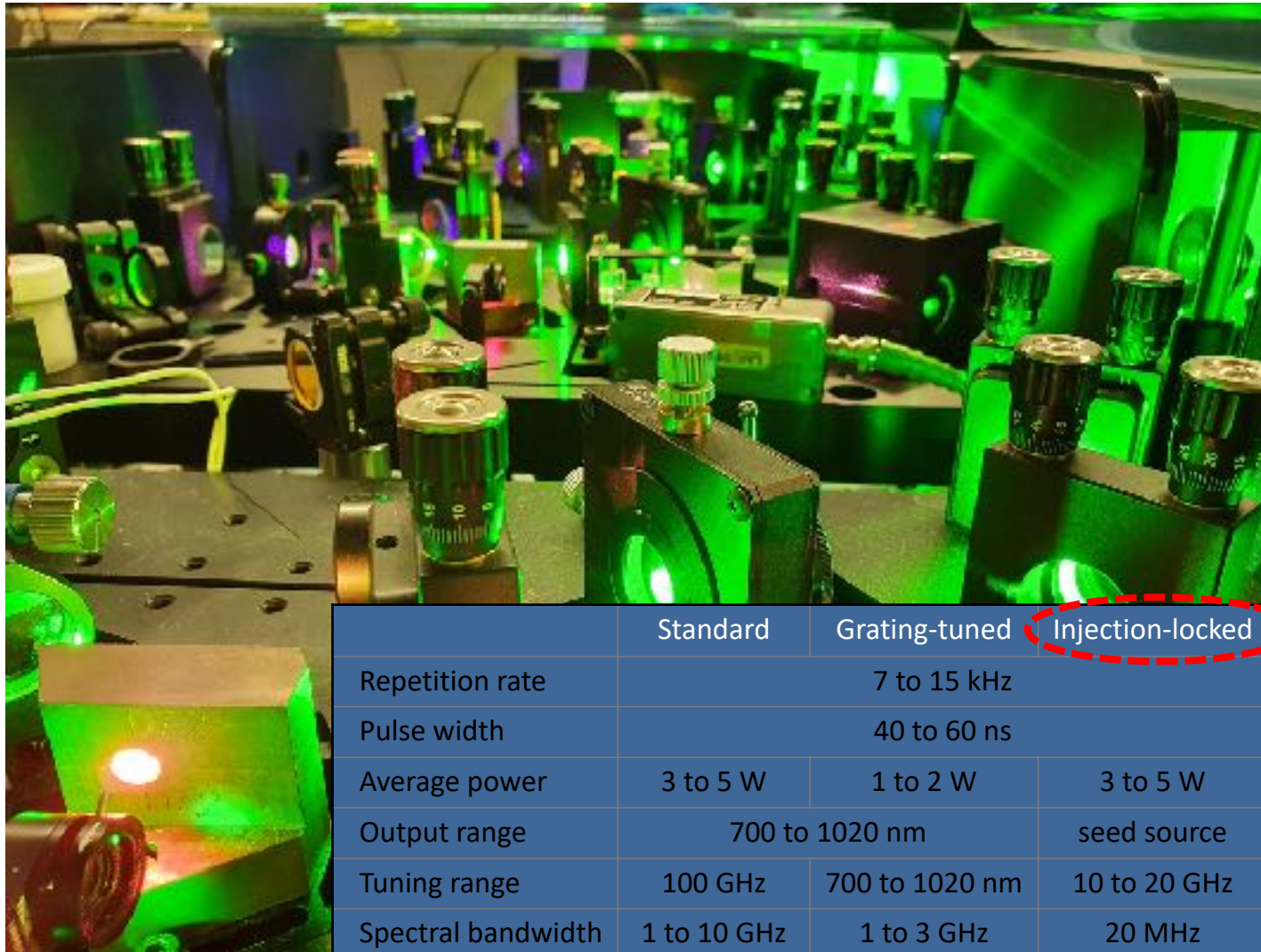
- Hot cavity laser ion source
- Standard in-source ionization
- Perpendicular laser interaction geometry for high-resolution spec.



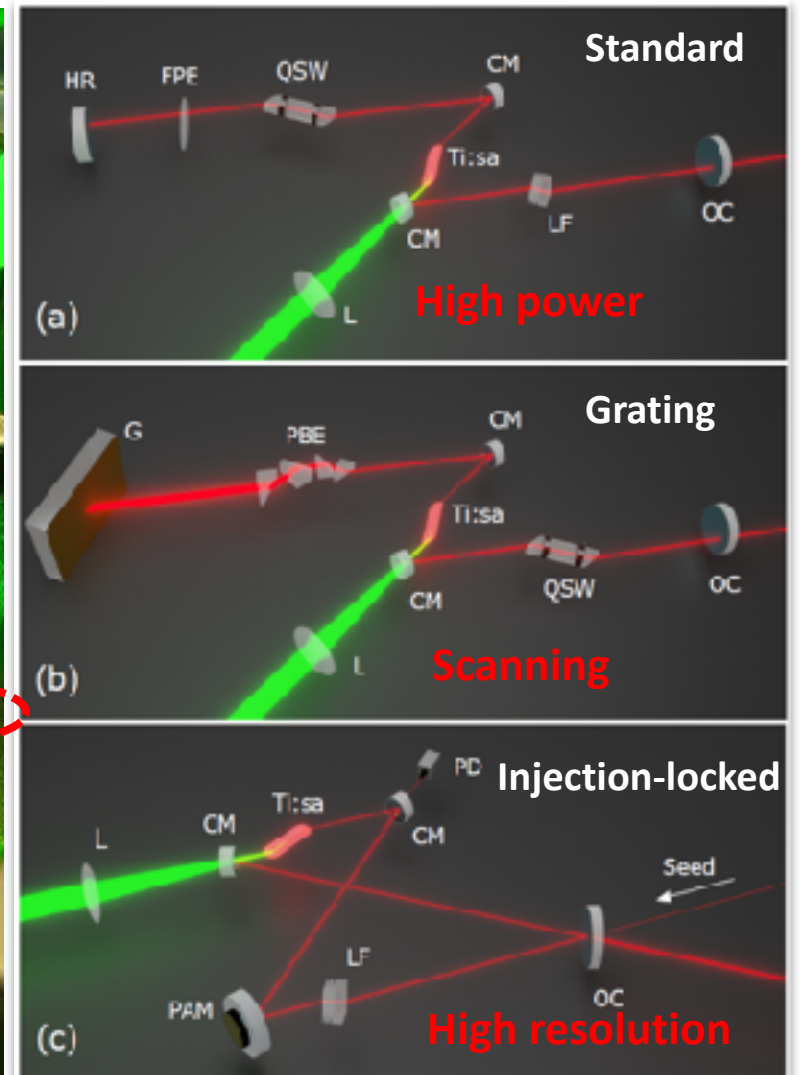
# PI-LIST Perpendicularly Illuminated - Laser Ion Source and Trap [10]



# Laser systems at Mainz

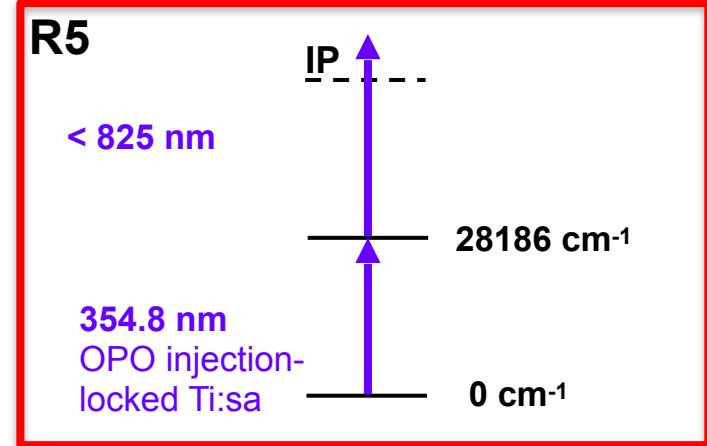
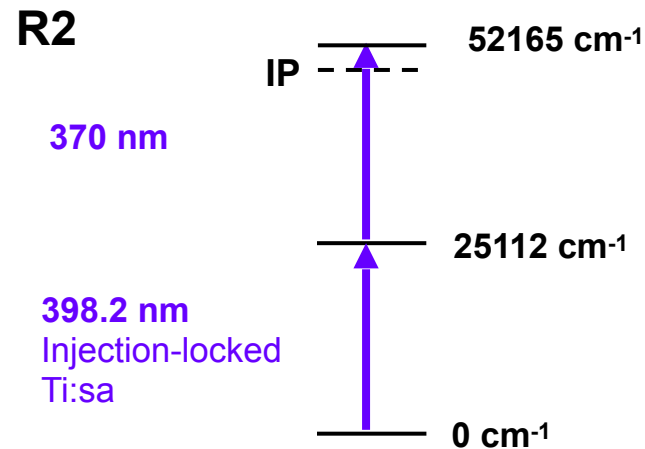
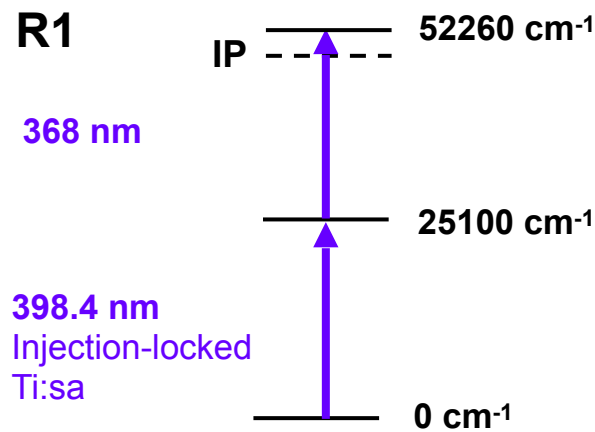


	Standard	Grating-tuned	Injection-locked
Repetition rate		7 to 15 kHz	
Pulse width		40 to 60 ns	
Average power	3 to 5 W	1 to 2 W	3 to 5 W
Output range	700 to 1020 nm		seed source
Tuning range	100 GHz	700 to 1020 nm	10 to 20 GHz
Spectral bandwidth	1 to 10 GHz	1 to 3 GHz	20 MHz



# Ionization schemes [11]

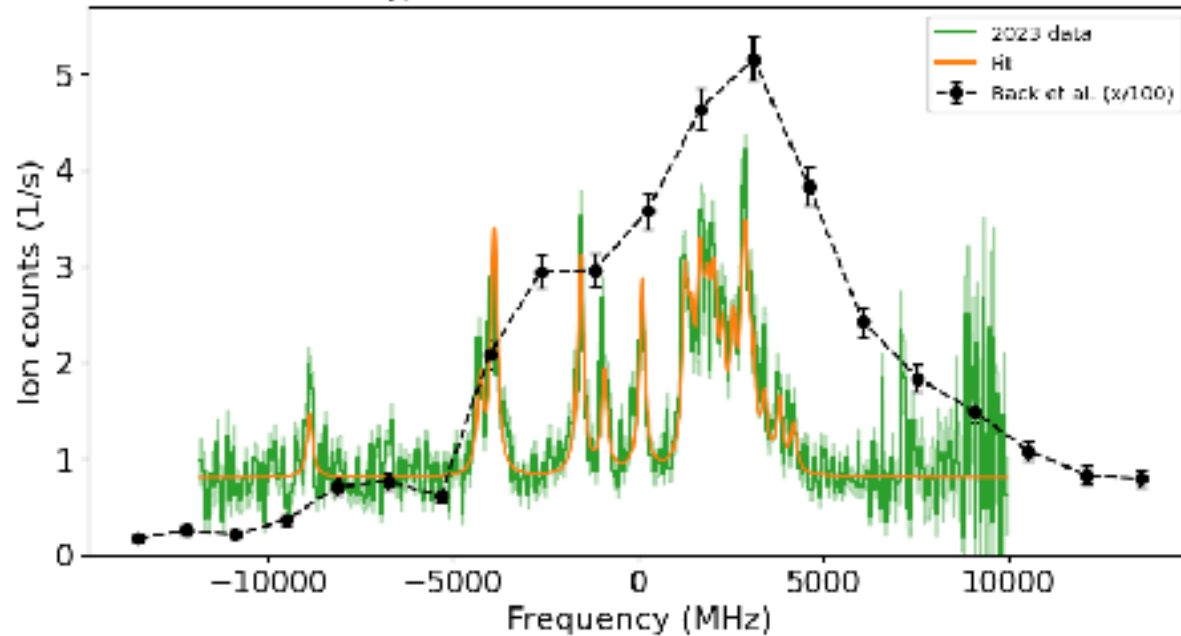
No.	WN (cm <sup>-1</sup> )	FWHM (cm <sup>-1</sup> )	WN fund. (cm <sup>-1</sup> )	WL fund. (nm)	WN to IP (cm <sup>-1</sup> )	WN to IP fund. (cm <sup>-1</sup> )	WL to IP fund. (nm)
R1	25,089.80 pm 0.2		12,549.90	796.8	27,300.2	13650.1	732.6
R2	25,111.80 pm 0.2	-	12,555.90	798.4	27,288.2	13644.1	732.9
R3	27,389 pm 1.5	0.85 pm 0.16	13,694.5	730.2	25,011	12505.5	799.6
R4	27,466 pm 1.5	1.34 pm 0.09	13,733	728.2	24,934	12467	802.1
R5	28,185 pm 1.5	1.08 pm 0.05	14,092.5	709.6	24,215	12107.5	825.9
R6	28,377 pm 1.5	0.75 pm 0.05	14,188.5	704.8	24,023	12011.5	832.5
R7	28,391 pm 1.5	0.81 pm 0.03	14,195.5	704.4	24,009	12004.5	833.0



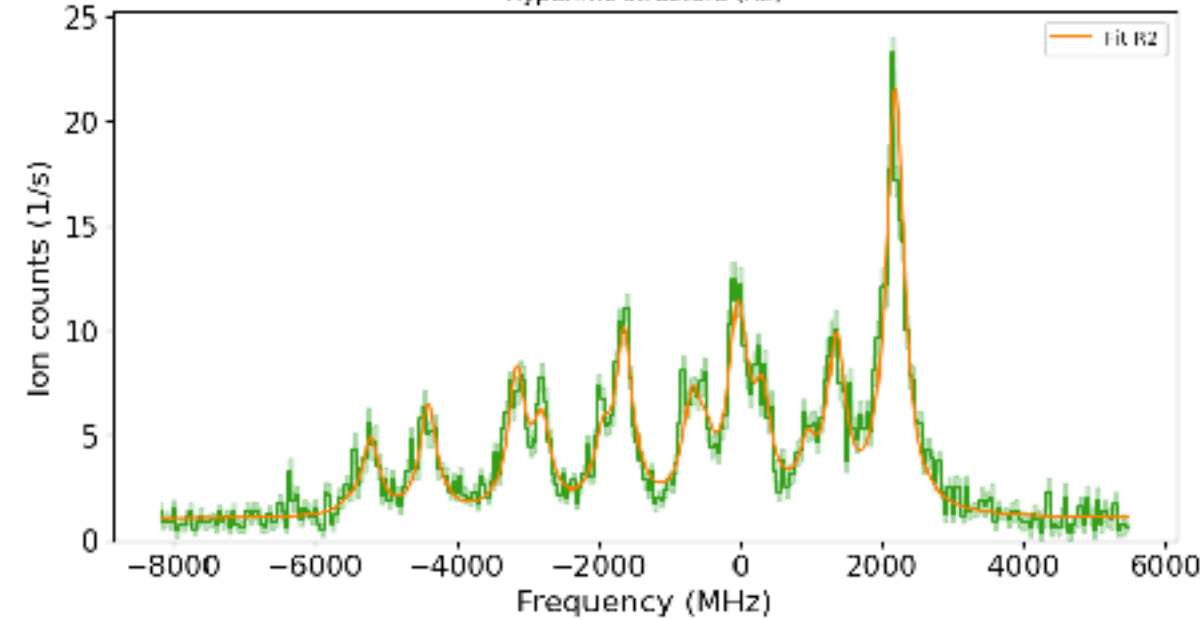
[11] H. Backe, et al., *Hyperfine interactions* 162(1-4), 3–14 (2005)

## PI-LIST measurements

Hyperfine structure (R1) with 50 MHz bin size



Hyperfine structure (R2)



	J	E (cm <sup>-1</sup> ) [12]	A (MHz)	B (MHz)
Ground state	6	0	-149 (5)	-10454 (100)
Excited state R1	6	25100	-309 (5)	-12821 (100)
Excited state R2	5	25112	-9 (5)	-13529 (100)

[12] H. Backe, et al., *Hyperfine interactions* 162(1-4), 3–14 (2005)

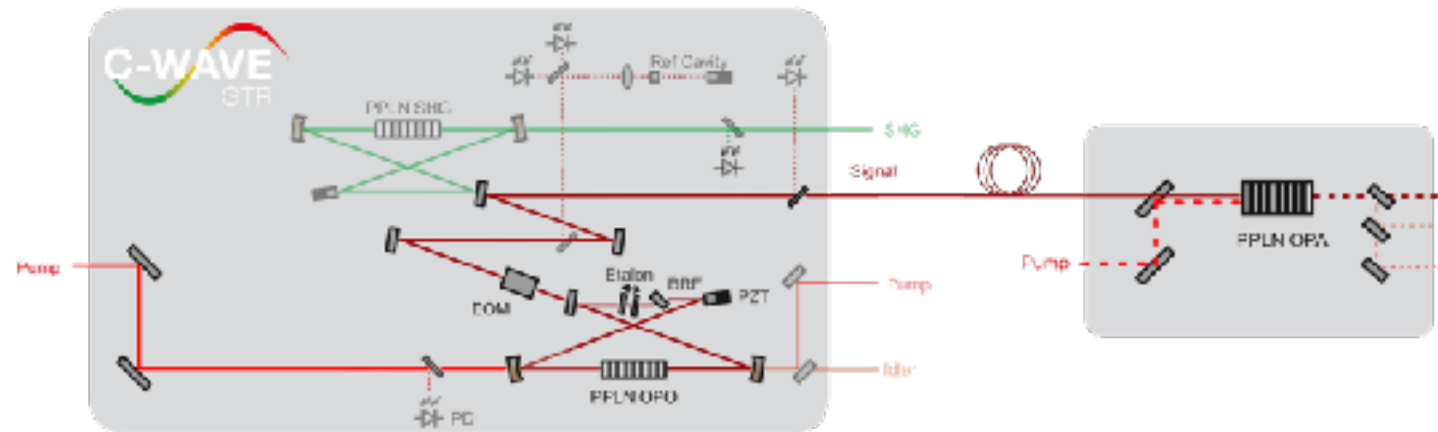
# Conclusions

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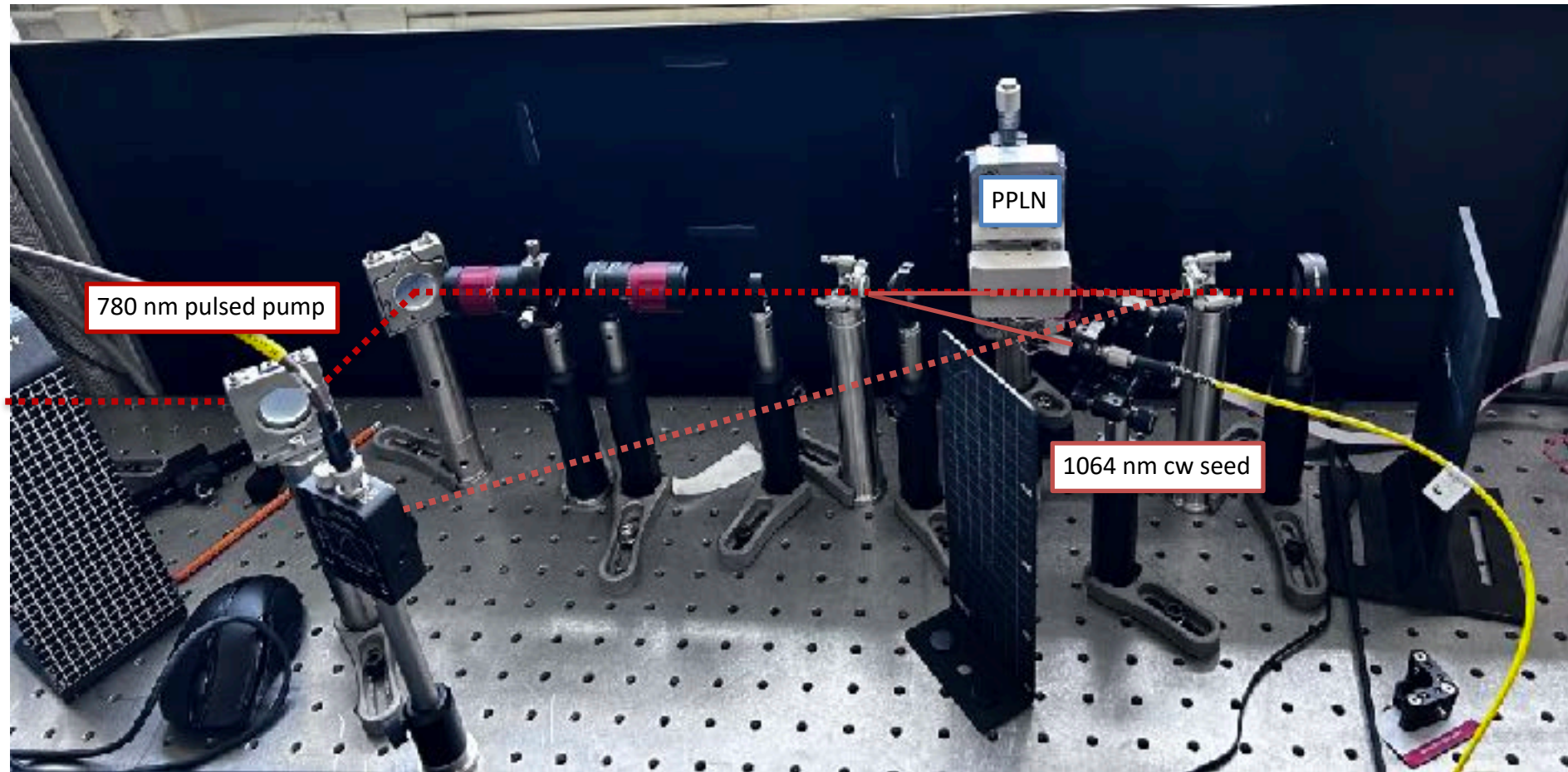
- OPO injection-seeded PDA is suitable for high resolution spectroscopy
  - FWHM  $\leq 173$  MHz
  - Known values are in good agreement with literature
  - New measurements for some silver isotopes
- First attempt OPO injection-locked Ti:Sa limited by pump availability
  - Good data from other two excitation schemes

# Current work

- OPO/OPA system, all solid state
  - Uses same crystal as the OPO
  - 780 nm pulsed pump (Ti:Sa)
  - No cavity needed
  - Amplified pulses in the OPO fundamental (1000nm - 1500nm)
- Further frequency doubled or tripled stages needed



# Experimental setup

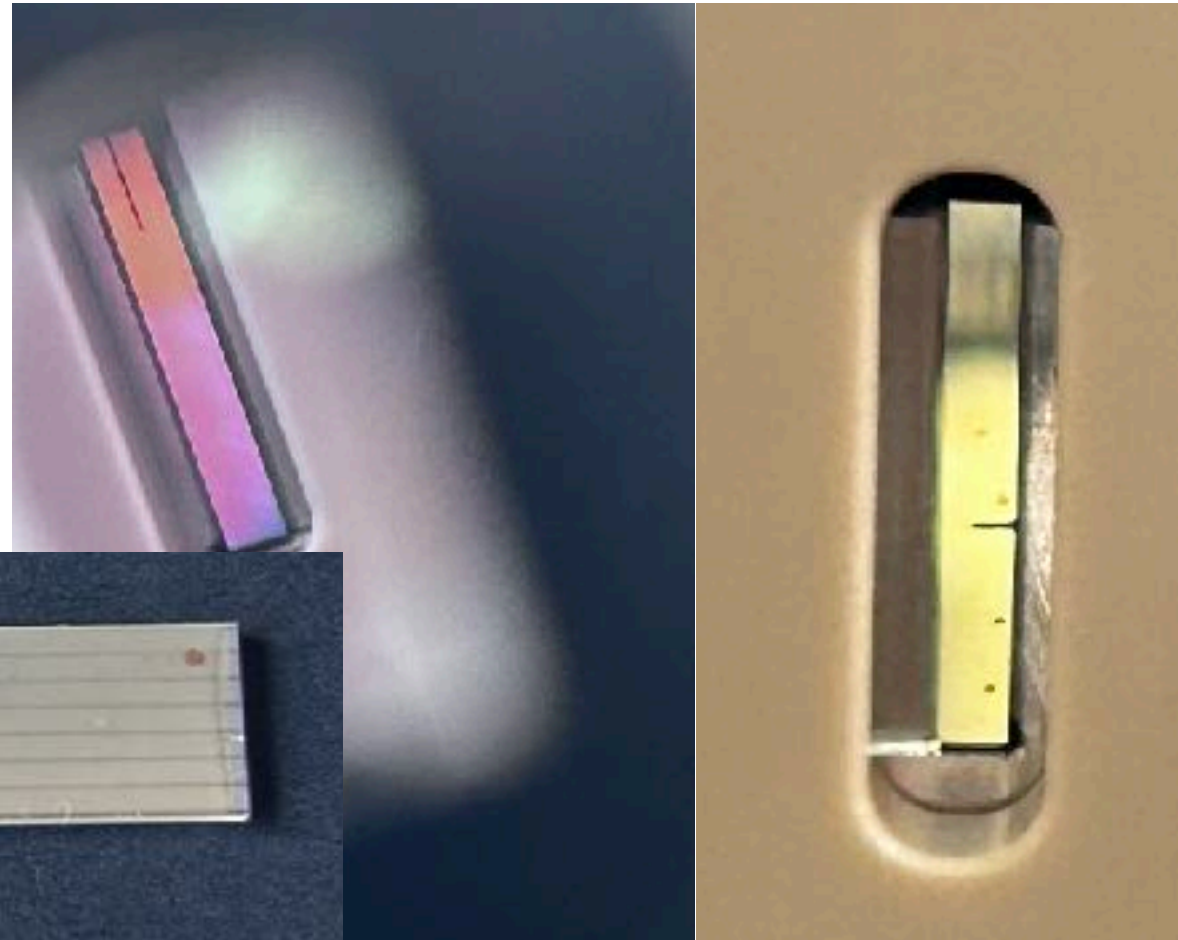




# Current setbacks

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- 140mW pump light at 1kHz:
- Unseeded
- No optimization
  - 12% conversion rate



# Acknowledgements silver experiment



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# The fermium collaboration



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Andrea T. Loria Basto  
Christoph Mokry  
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Danny Münzberg  
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Elisabeth Rickert  
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Kristian Myhre  
Samantha Schrell  
Shelley Van Cleve

## TU Darmstadt

Thomas Walther

## Nagoya University

Hideki Tomita

## CERN

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S. Karpuk

### ILL Grenoble

ILL reactor team and health physics

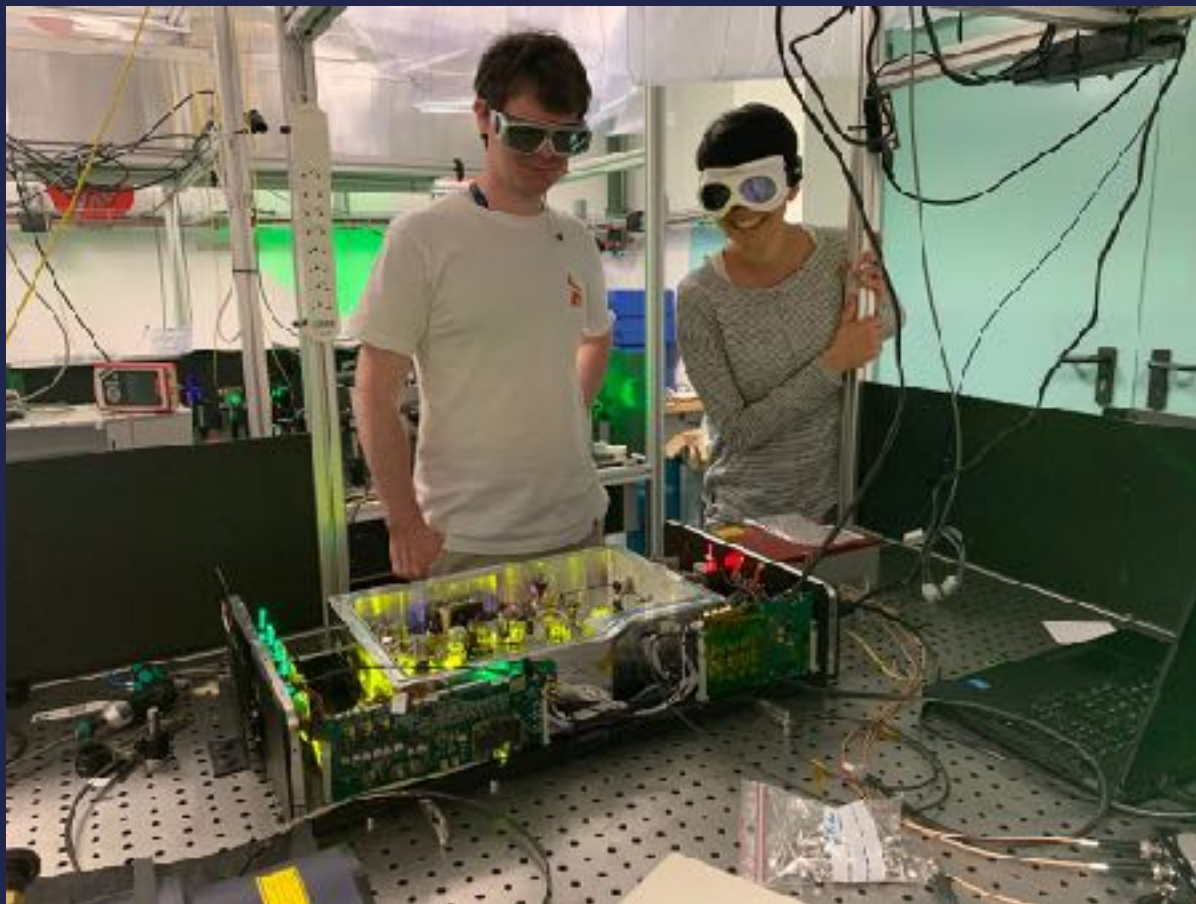
### Oak Ridge National Laboratory

Nate Sims, Radioisotope Laboratory Technician  
Nonreactor Nuclear Facilities Division Hot Cell Staff

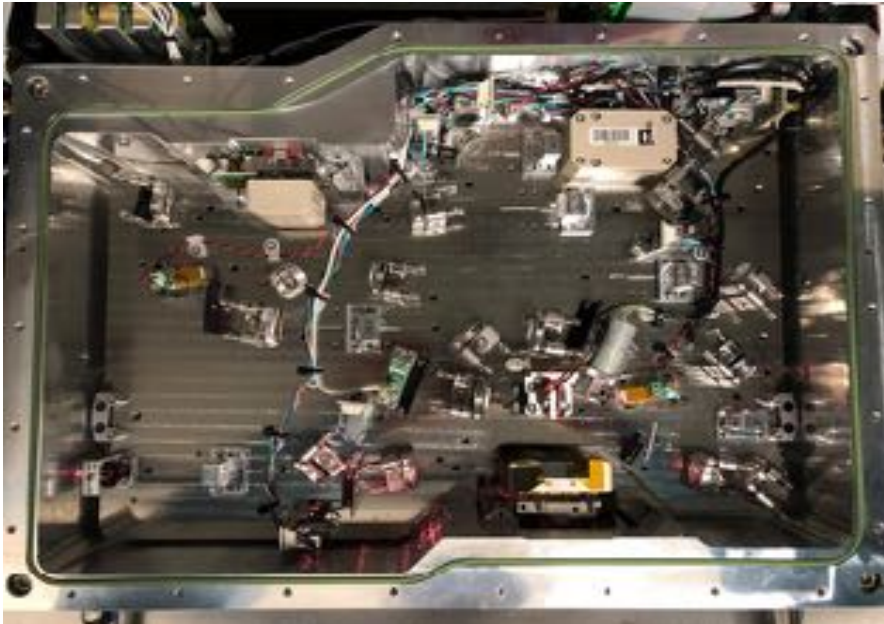
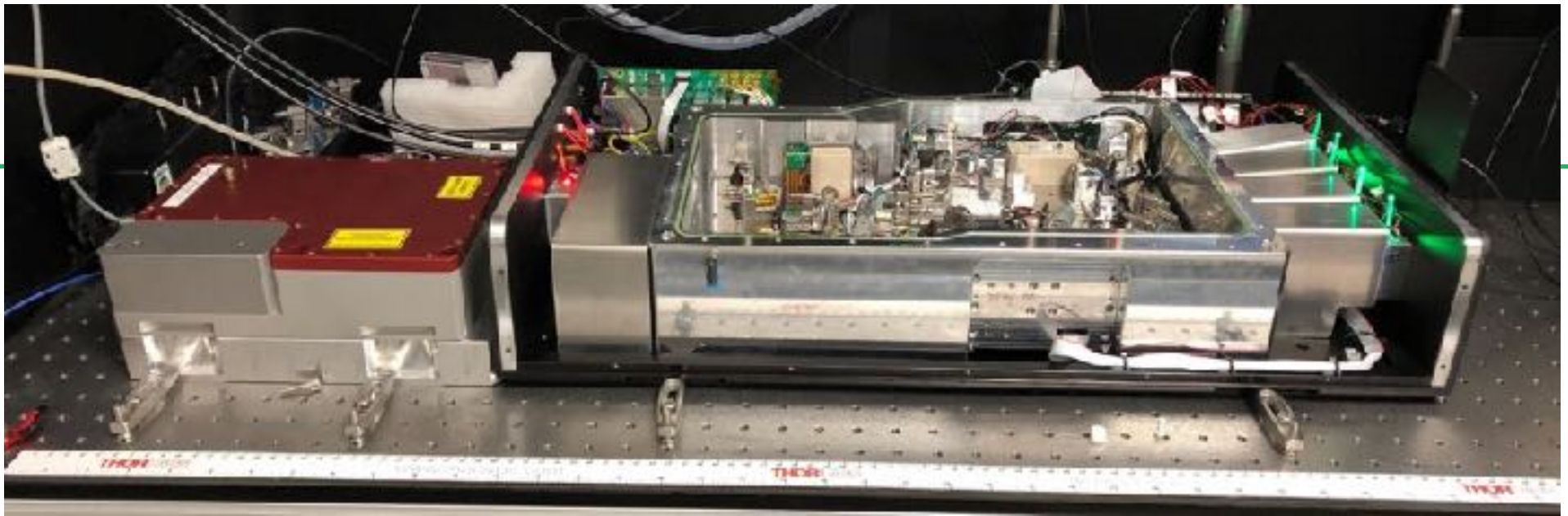
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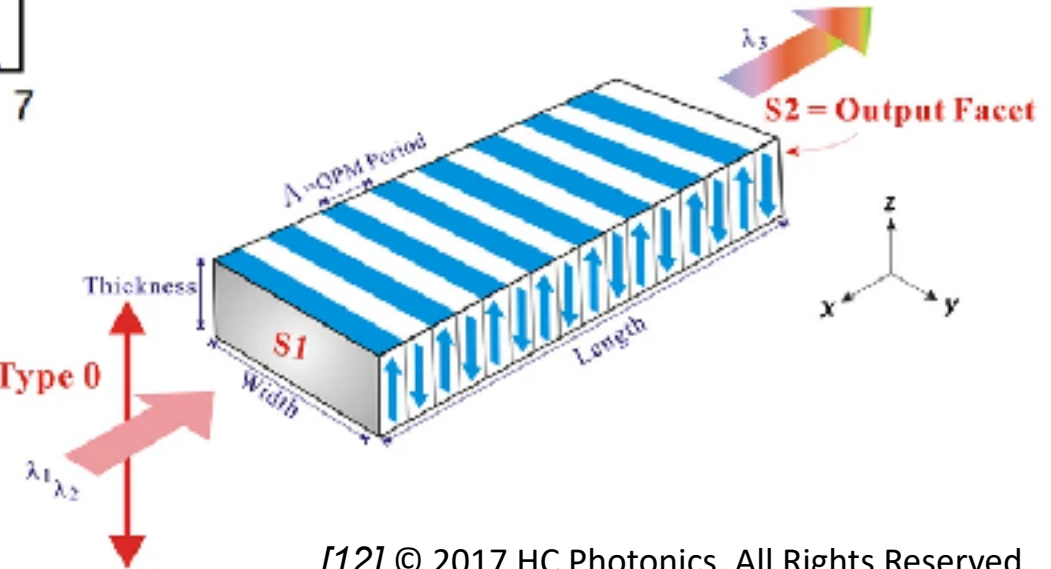
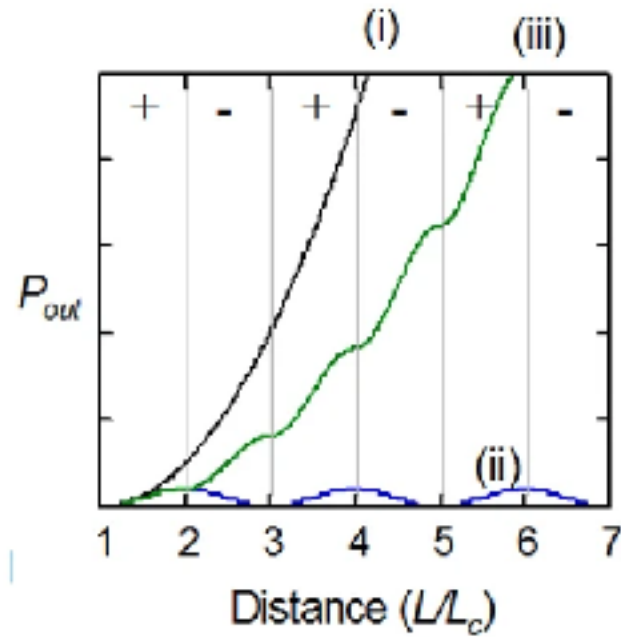
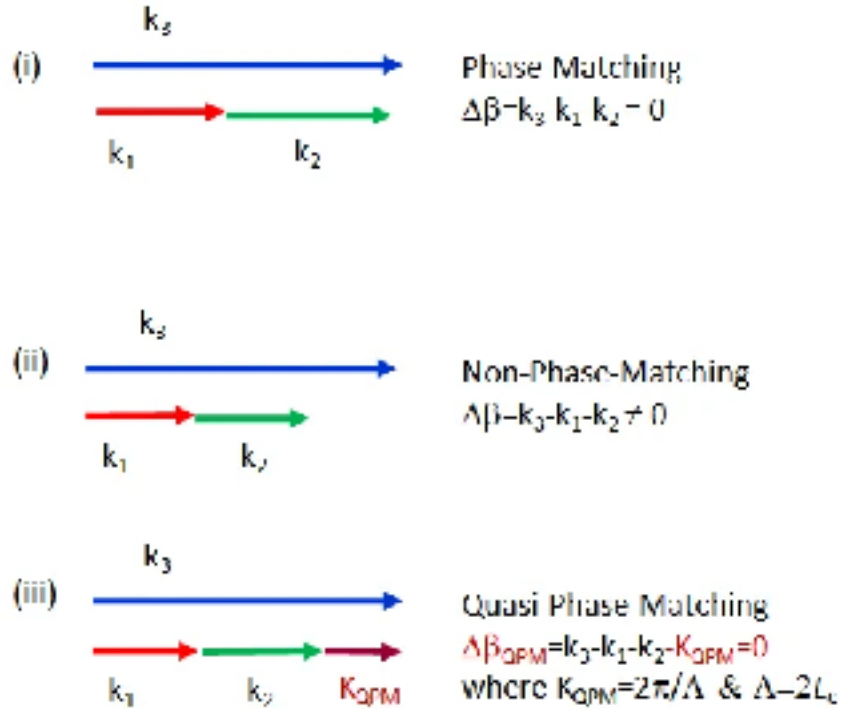
# Thanks for your attention!



Back-up slides



# PPLN crystal



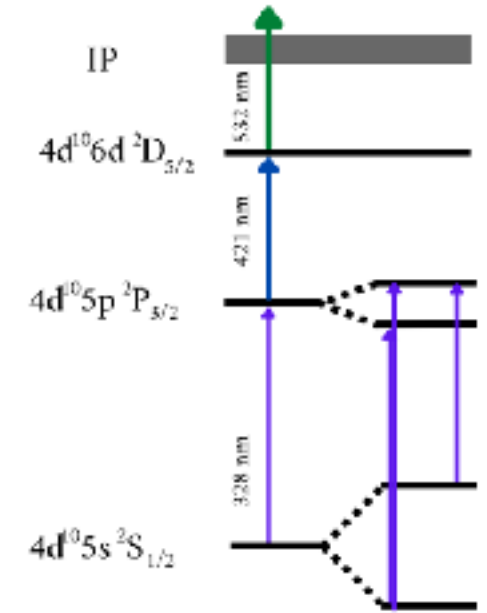
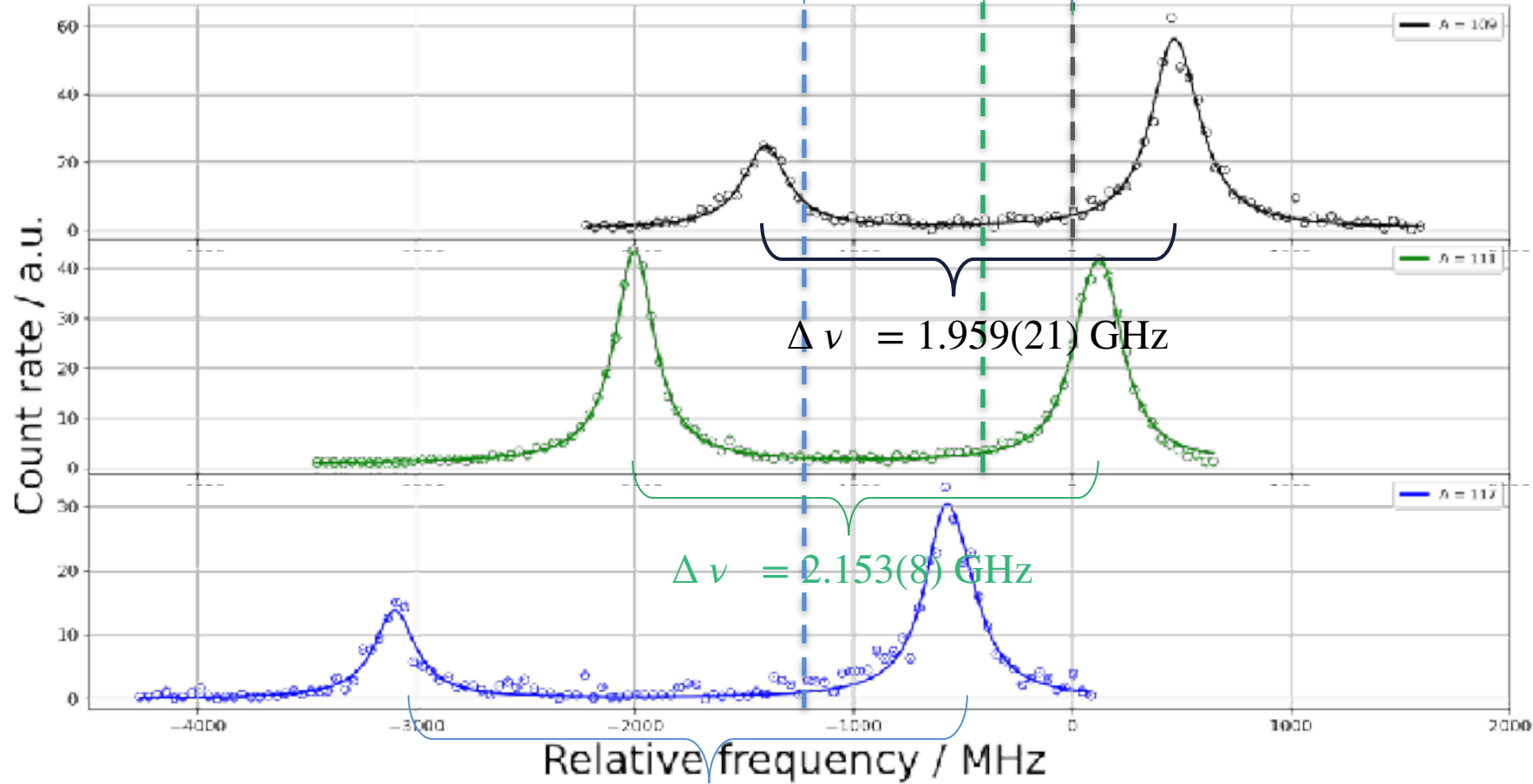
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# HFS 109,111,117Ag

$$\delta\nu_{IS}^{109,117} = -1198(10) \text{ MHz}$$

$$\delta\nu_{IS}^{109,111} = -436(5) \text{ MHz}$$

$$\delta\nu_{IS}^{109,A} = \text{COG}_A - \text{COG}_{109}$$



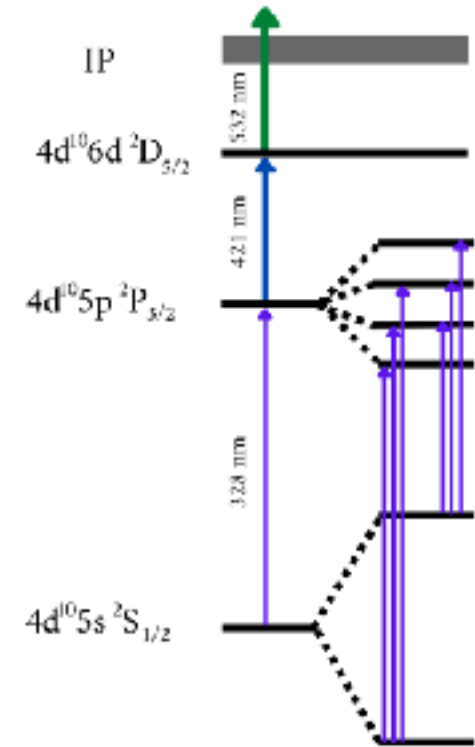
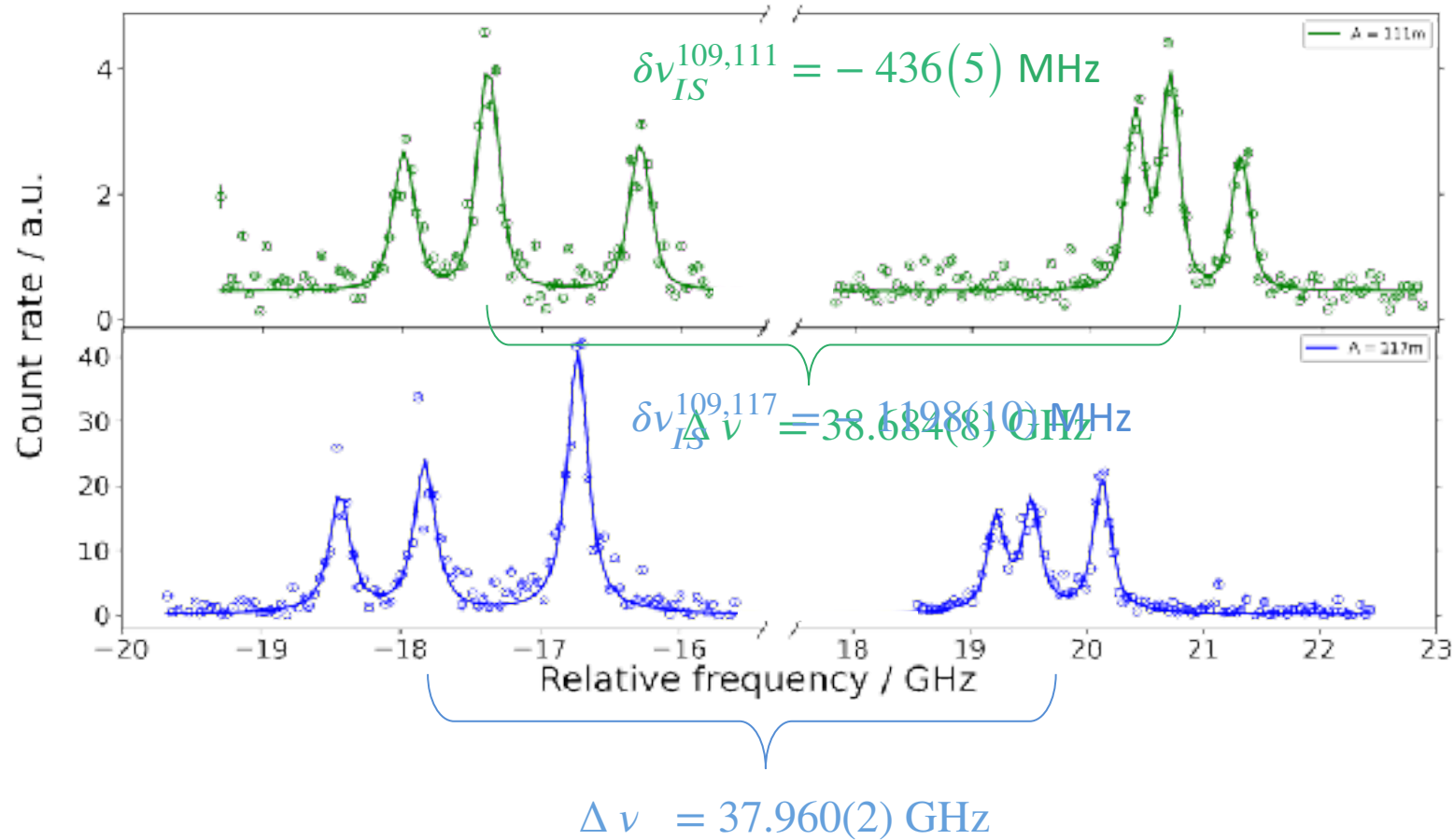
For  $J = \frac{1}{2}$  and  $I = \frac{1}{2} \Rightarrow \Delta\nu =$

$$\Delta\nu = 2.615(17) \text{ GHz}$$





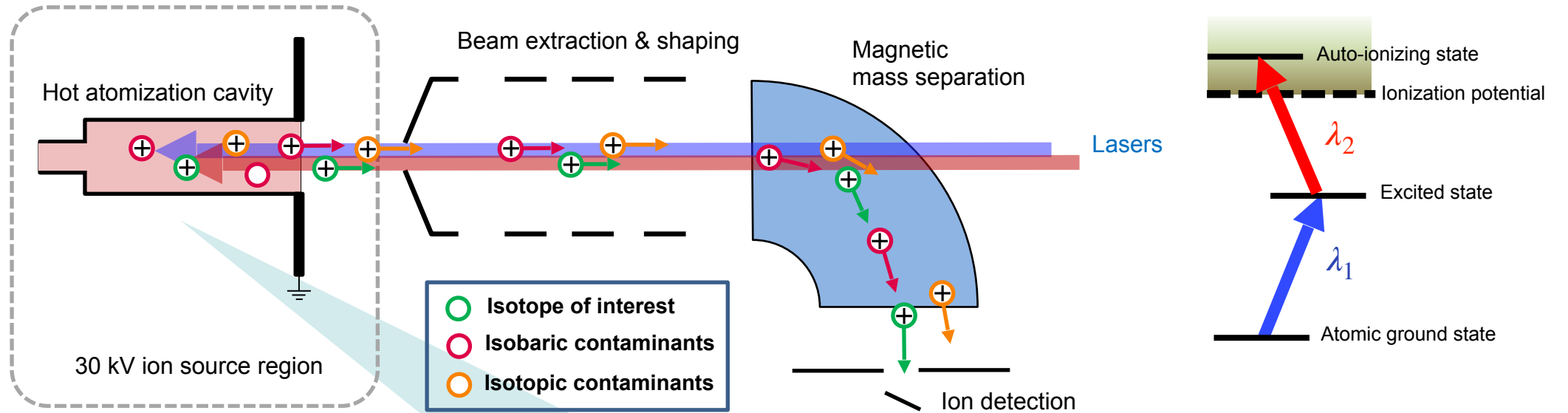
# HFS 111m,117mAg



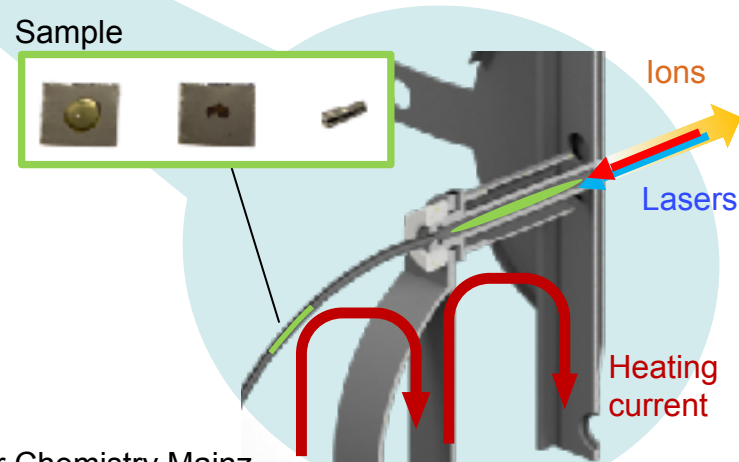
For  $J = \frac{1}{2}$  and  $I = \frac{7}{2} \Rightarrow \Delta\nu =$

4A

# RISIKO mass separator



- Chemical sample preparation\*
- Sample evaporation in hot cavity
- Multi-step photoionization by pulsed lasers
- Mass separation in dipole magnet  $m/\Delta m \approx 800$
- Single ion detection



\*in collaboration with Dpt. of Nuclear Chemistry Mainz

**RISIKO**

# Medium resolution scans

- R1, R2, R5
- Seeded laser insource
- Influence of AI studied
- New AIS for R1, R2

