Study, Development and Optimization of Laser Resonant Photo-Ionization processes applied to species of interest for the Isolpharm-SPES project

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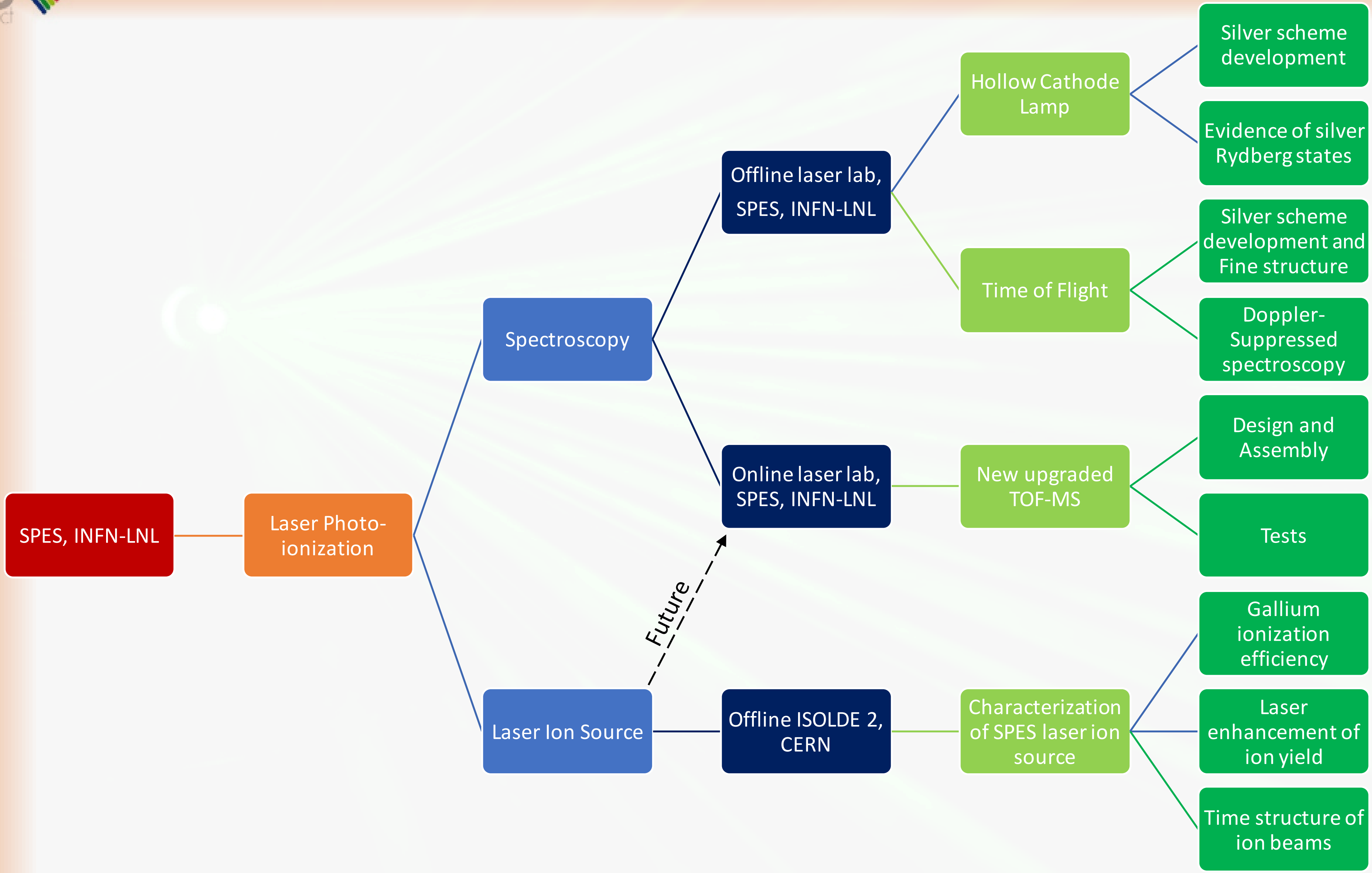
INFN-LNL

UNISI

A very strong collaboration on-going for 10+ years.



- SPES, INFN-LNL
  - Laser Photo-ionization
    - Spectroscopy
      - Offline laser lab, SPES, INFN-LNL
        - Hollow Cathode Lamp
          - Silver scheme development
          - Evidence of silver Rydberg states
        - Time of Flight
          - Silver scheme development and Fine structure
          - Doppler-Suppressed spectroscopy
      - Online laser lab, SPES, INFN-LNL
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          - Design and Assembly
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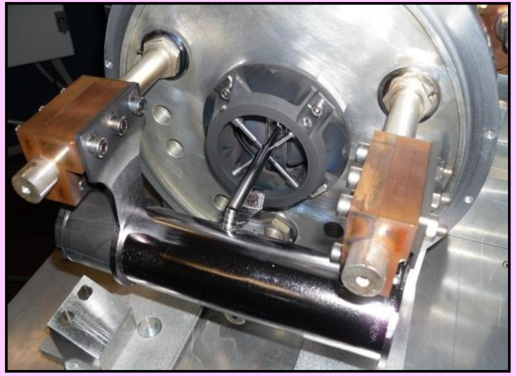
SPES @ INFN-LNL

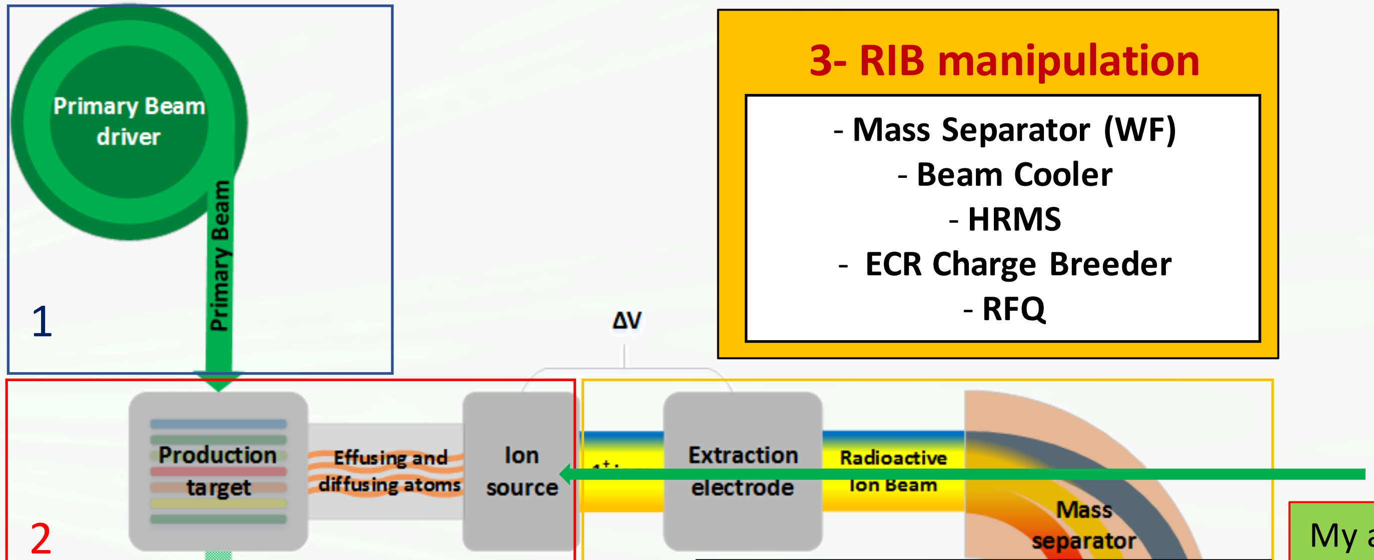
1 - Driver

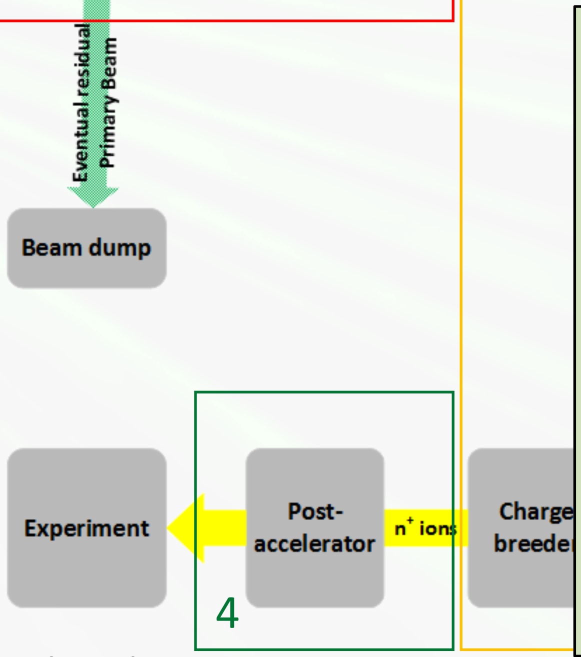
70 MeV cyclotron

2 – Target-Ion Source Complex



40 MeV 200μA protons on $UC_x = 10^{13}$ fission/s.





My activities

3- RIB manipulation

- Mass Separator (WF)
- Beam Cooler
- HRMS
- ECR Charge Breeder
- RFQ

4 - Post Accelerator:

ALPI existing complex

The ISOLPHARM method

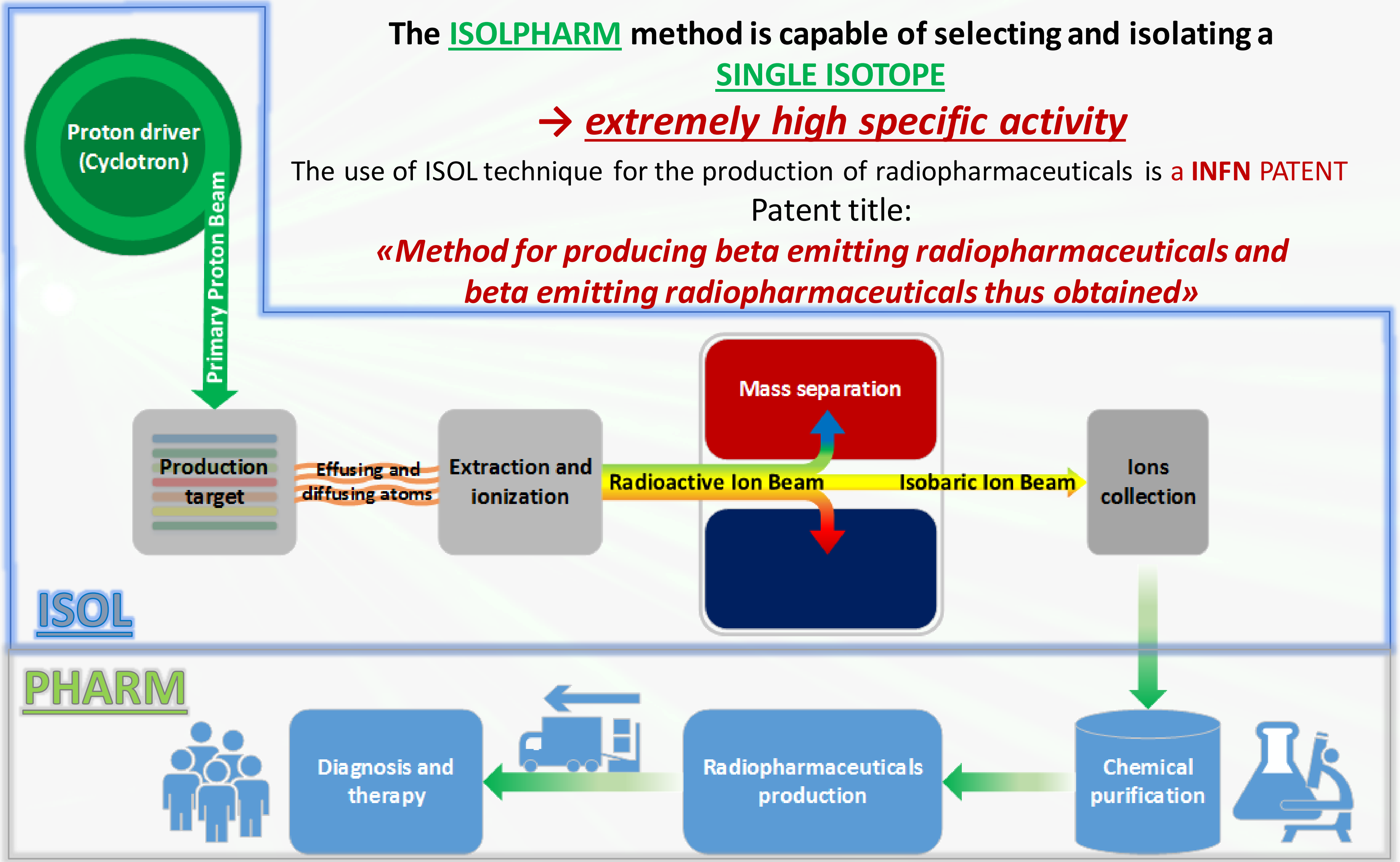
The ISOLPHARM method is capable of selecting and isolating a SINGLE ISOTOPE

→ *extremely high specific activity*

The use of ISOL technique for the production of radiopharmaceuticals is a **INFN** PATENT

Patent title:

«Method for producing beta emitting radiopharmaceuticals and beta emitting radiopharmaceuticals thus obtained»



Proton driver (Cyclotron)

Primary Proton Beam

Production target

Effusing and diffusing atoms

Extraction and ionization

Radioactive Ion Beam

Mass separation

Isobaric Ion Beam

Ions collection

ISOL

PHARM

Chemical purification

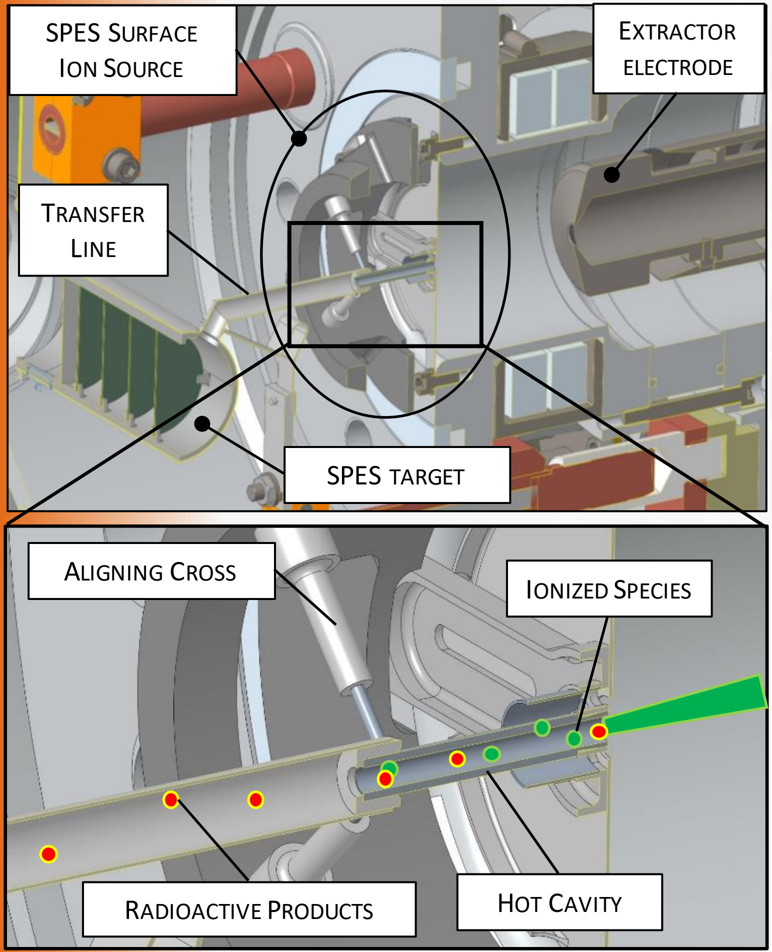
Radiopharmaceuticals production

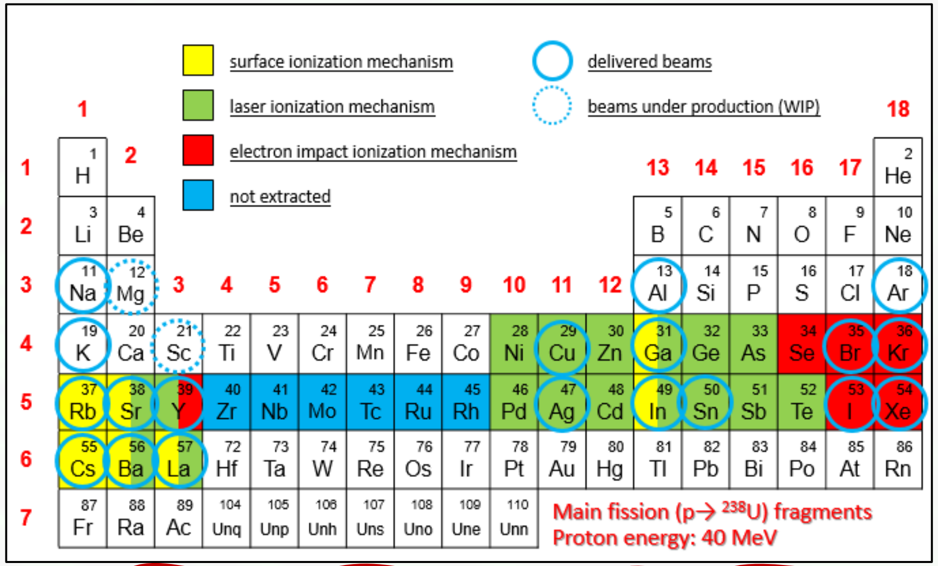
Diagnosis and therapy

Nuclide production can be extremely flexible

The SPES Ion Sources

The SPES Surface Ion Source (SIS)



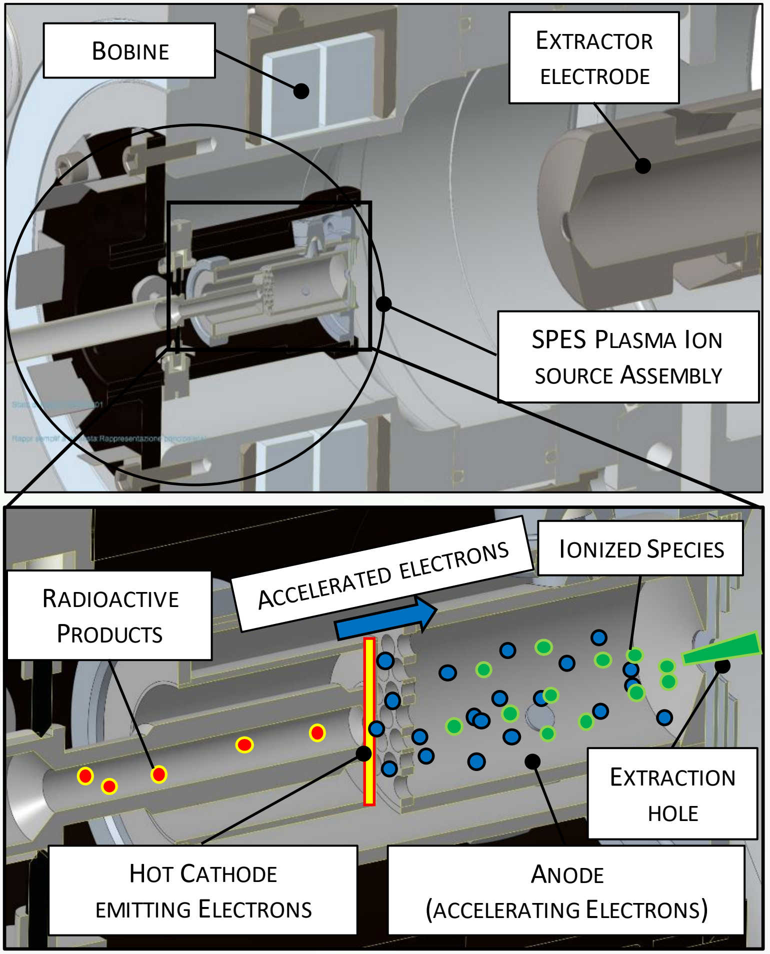


🟨 surface ionization mechanism
🟩 laser ionization mechanism
🟥 electron impact ionization mechanism
🟦 not extracted
○ delivered beams
◌ beams under production (WIP)

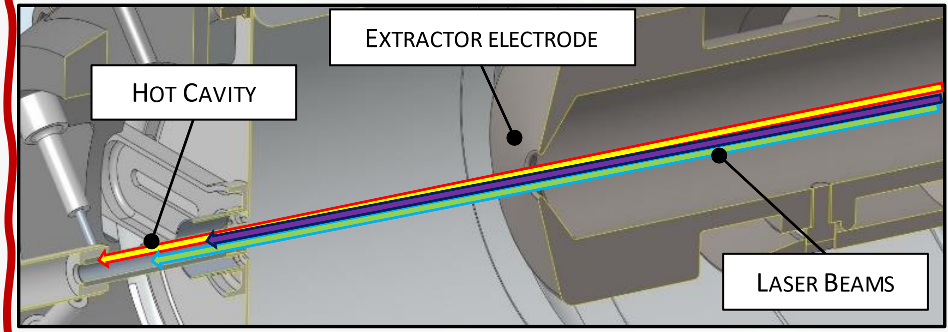
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
1	1 H																	2 He
2	3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
3	11 Na ○	12 Mg ◌											13 Al ○	14 Si	15 P	16 S	17 Cl	18 Ar ○
4	19 K ○	20 Ca	21 Sc ◌	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni 🟩	29 Cu 🟩 ○	30 Zn 🟩	31 Ga 🟨🟩 ○	32 Ge 🟩	33 As 🟩	34 Se 🟥	35 Br 🟥 ○	36 Kr 🟥 ○
5	37 Rb 🟨 ○	38 Sr 🟨 ○	39 Y 🟩🟥 ○	40 Zr 🟦	41 Nb 🟦	42 Mo 🟦	43 Tc 🟦	44 Ru 🟦	45 Rh 🟦	46 Pd 🟩	47 Ag 🟩 ○	48 Cd 🟩	49 In 🟨🟩 ○	50 Sn 🟩 ○	51 Sb 🟩	52 Te 🟩	53 I 🟥 ○	54 Xe 🟥 ○
6	55 Cs 🟨 ○	56 Ba 🟨 ○	57 La 🟨🟩 ○	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
7	87 Fr	88 Ra	89 Ac	104 Unq	105 Unp	106 Unh	107 Uns	108 Uno	109 Une	110 Unn								

Main fission (p→ ^{238}U) fragments
Proton energy: 40 MeV

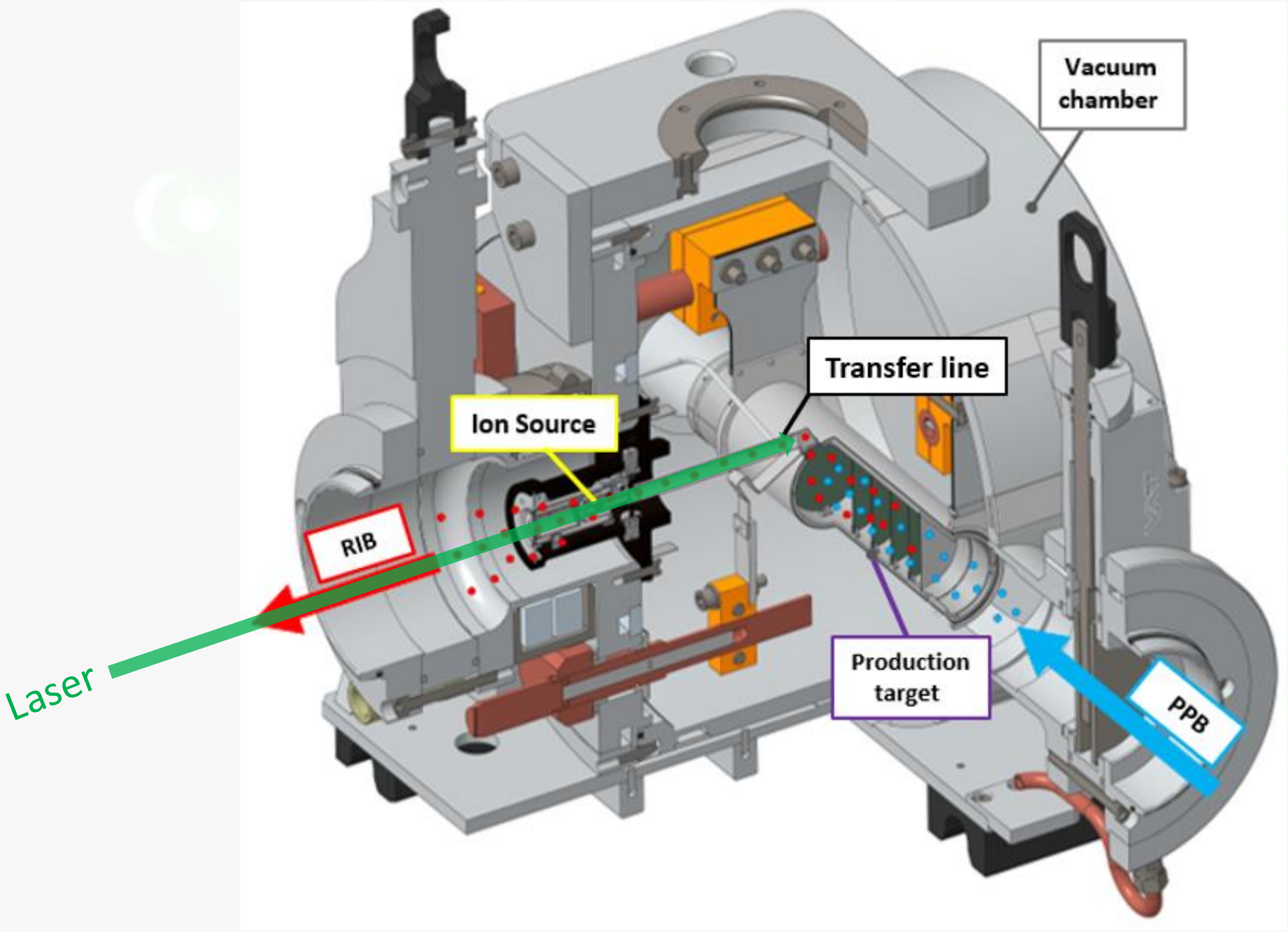
The SPES Plasma Ion Source



The SPES Laser Ion Source
(SIS + laser beams)



SPES Front-End and Laser Ion Source

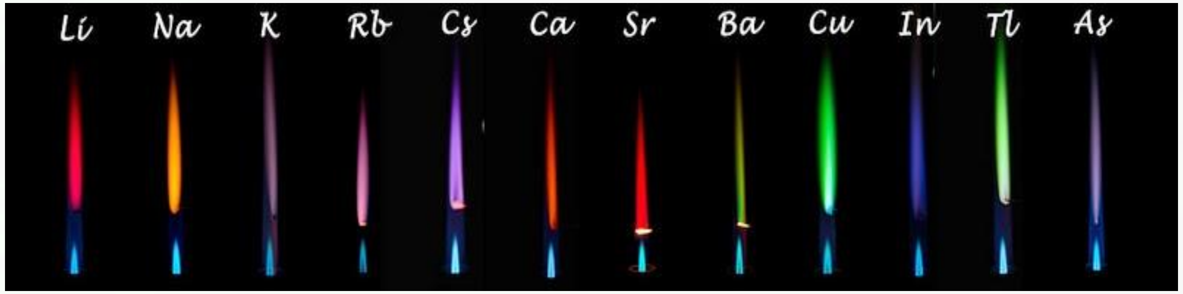


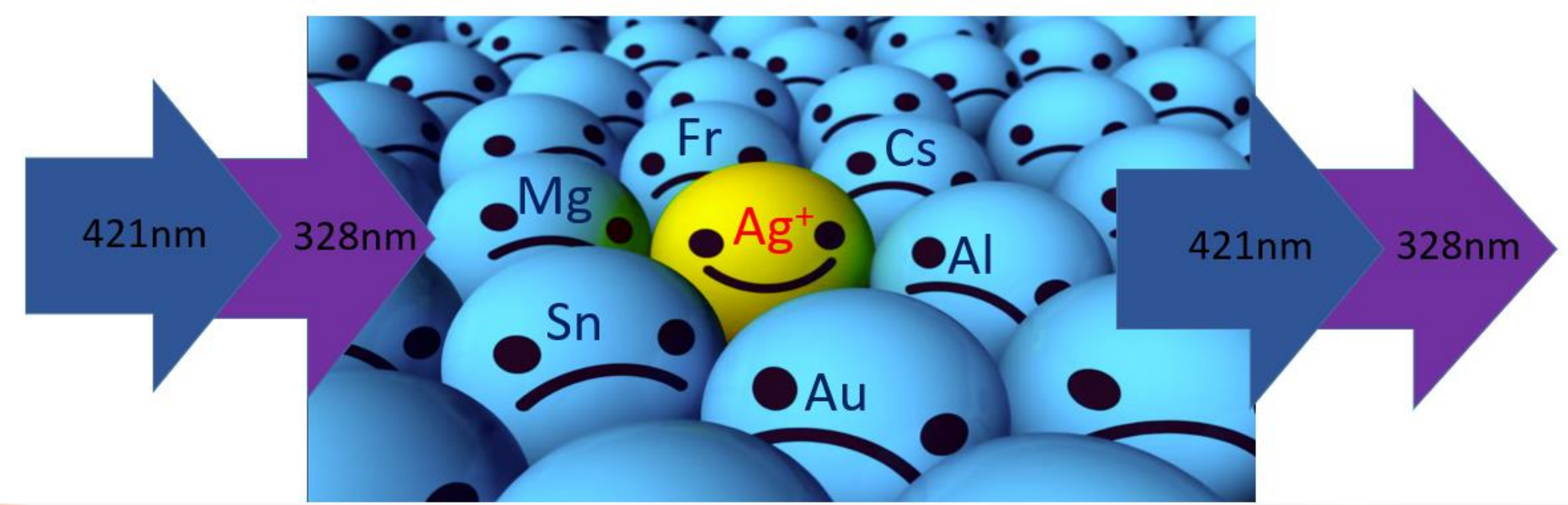
- ✓ **PPB** stands for Primary Proton beam
- ✓ **Production target** is a set of UC_x discs
- ✓ **RIB** stands for Radioactive Ion Beam

Laser Photo-ionization: Introduction

Flame test:

- Different elements "respond" with different colors
- The color depends on internal electrons energy level structure



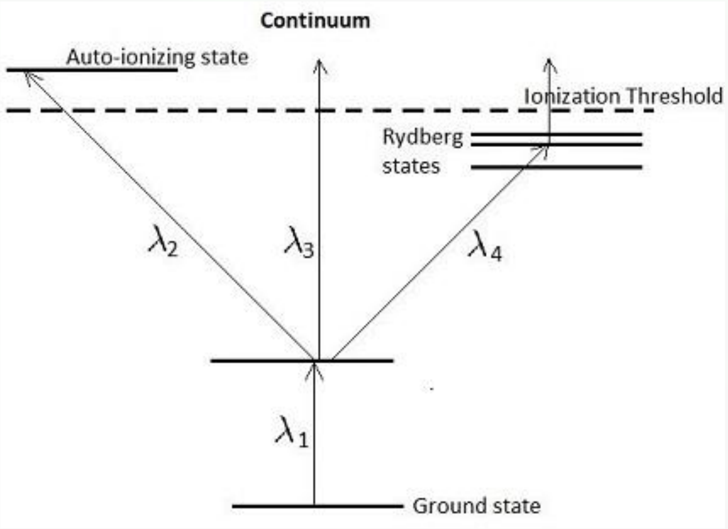


What separates the Laser Ion Source from Surface and Plasma Ion Source?

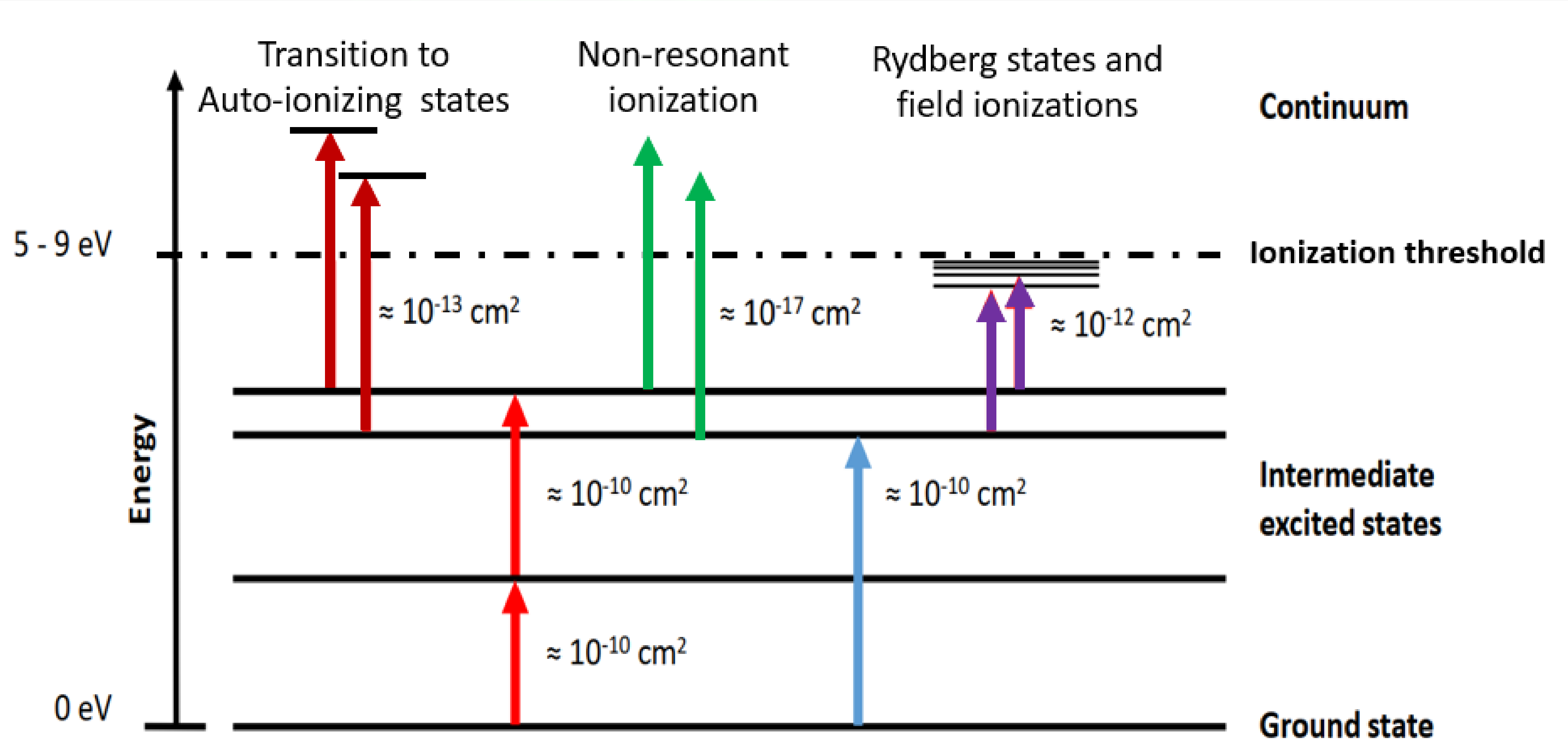
- ✓ Surface and plasma ion sources looks essentially at the **ionization threshold** as one unit/limit that needs to be crossed at one step.
- ✓ Laser ion source breaks down this process **into several steps** combined. Each step is unique to the electronic configuration of the element considered.
- ✓ And so called the “**Resonant Ionization Laser Ion Source (RILIS)**”.

Advantages of a laser ion source

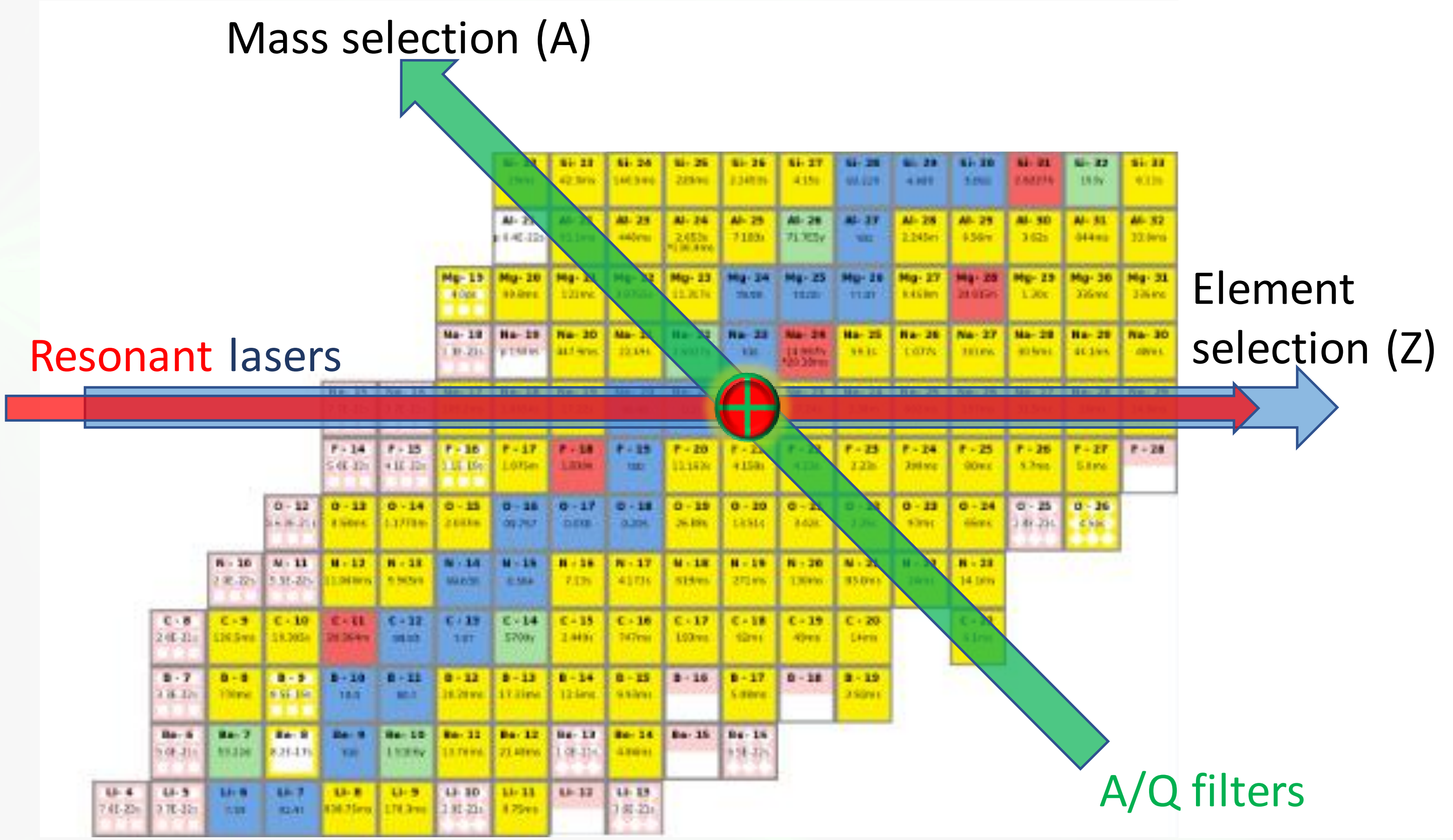
- ✓ Element selectivity
- ✓ Reduced isobaric contamination
- ✓ Beam diagnostics (switch the laser on and off)
- ✓ High efficiency (tens of percent)
- ✓ The ionization region is well-defined



Cross-sections of the different pathways



Production of Isotopic beam with RILIS technique



Mass selection (A)

Element selection (Z)

Resonant lasers

A/Q filters

The Laser Resonant Ionization @ SPES

Offline lab: Spectroscopy

- 2 Dye Lasers @ 10 Hz rep. rate

Online lab: RIB prod.

- 3 TiSa Laser @ 10 kHz rep. rate

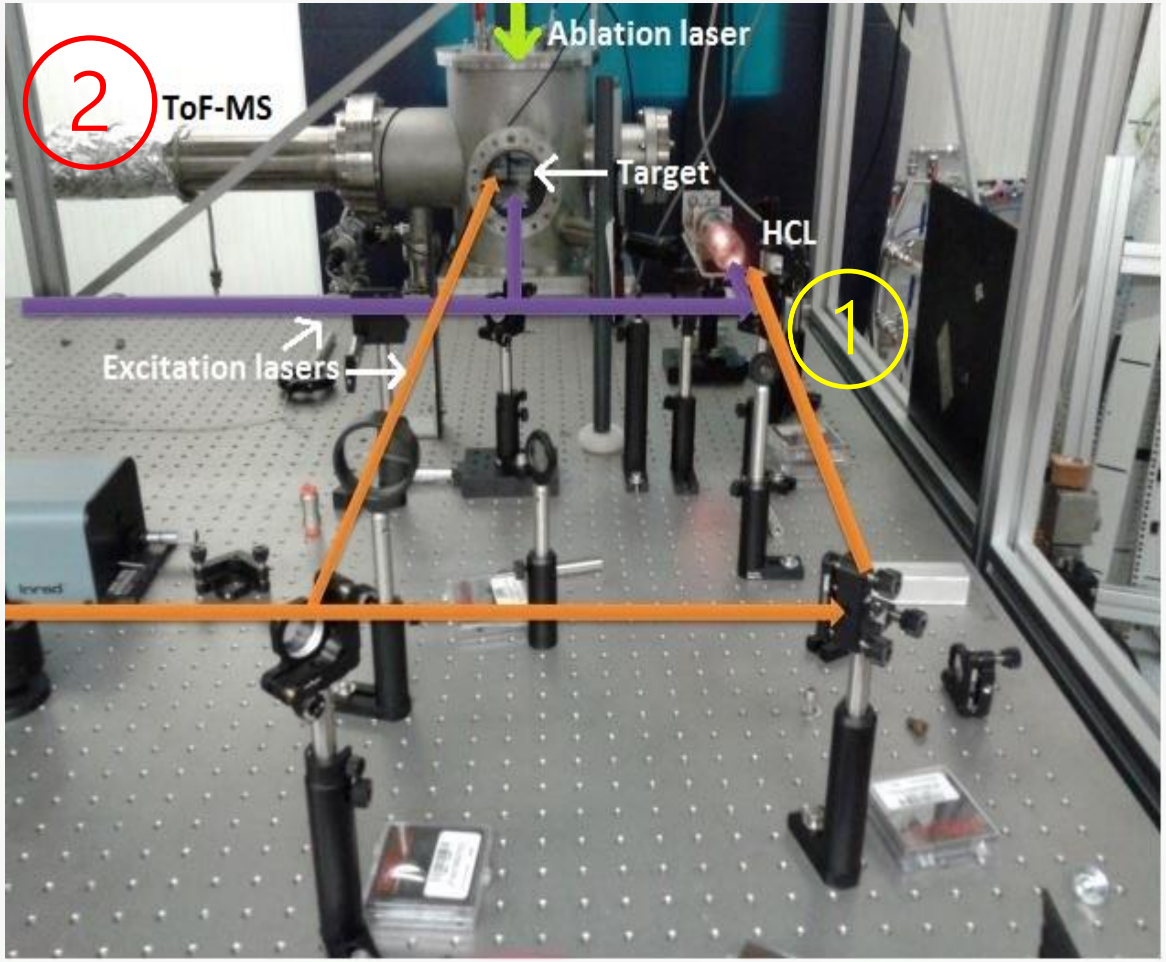
Methods of study in the offline laser lab

1. Using Hollow Cathode Lamp (HCL)

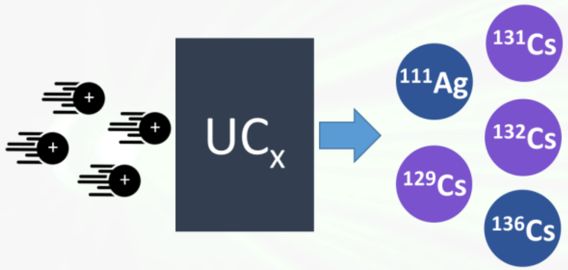
- Analysis of the Opto-galvanic effect (OGE)

2. Using ToF-MS

- Ablation source of atoms
- MCP detectors



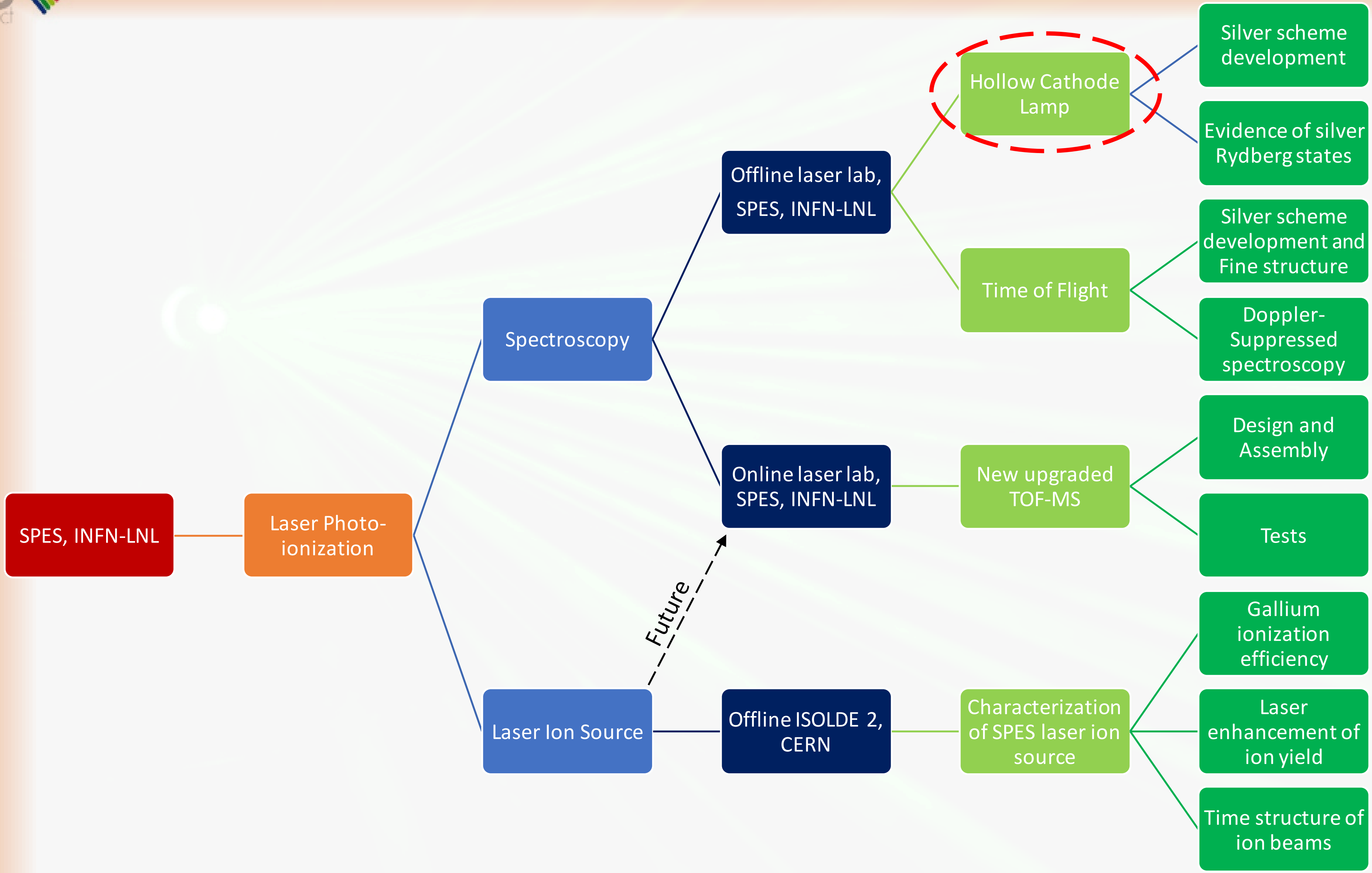
First topic: Photo-Ionization of silver



^{111}Ag properties

- ✓ **β^- emitter** (average energy **360 keV**)
- ✓ Medium half-life (**7.45 days**)
- ✓ Medium tissue penetration **(1.8 mm)**
- ✓ Low energy γ rays SPECT

We need to verify existing photo-ionization schemes of silver and/or, find new ones.



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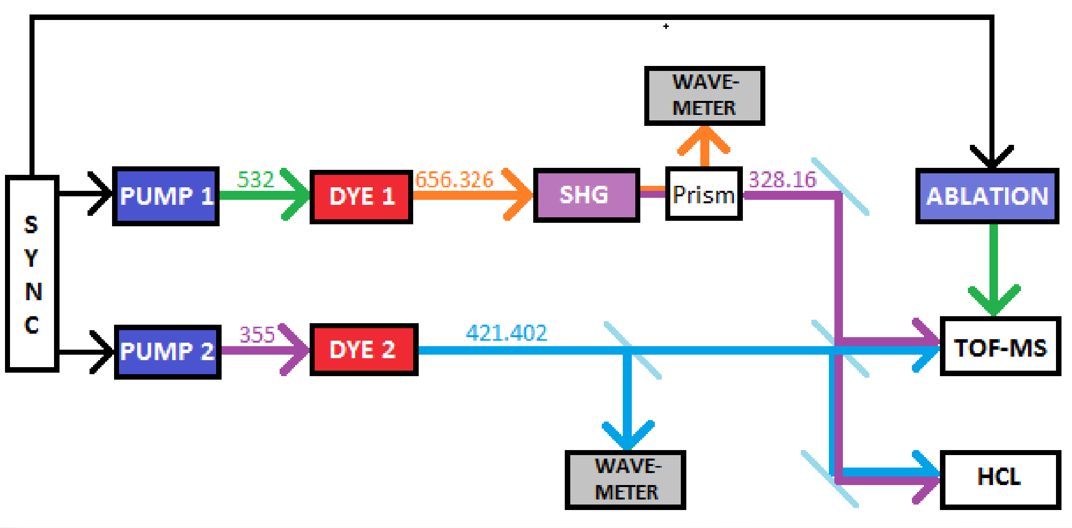
Future

Scheme development for the photo-ionization of silver

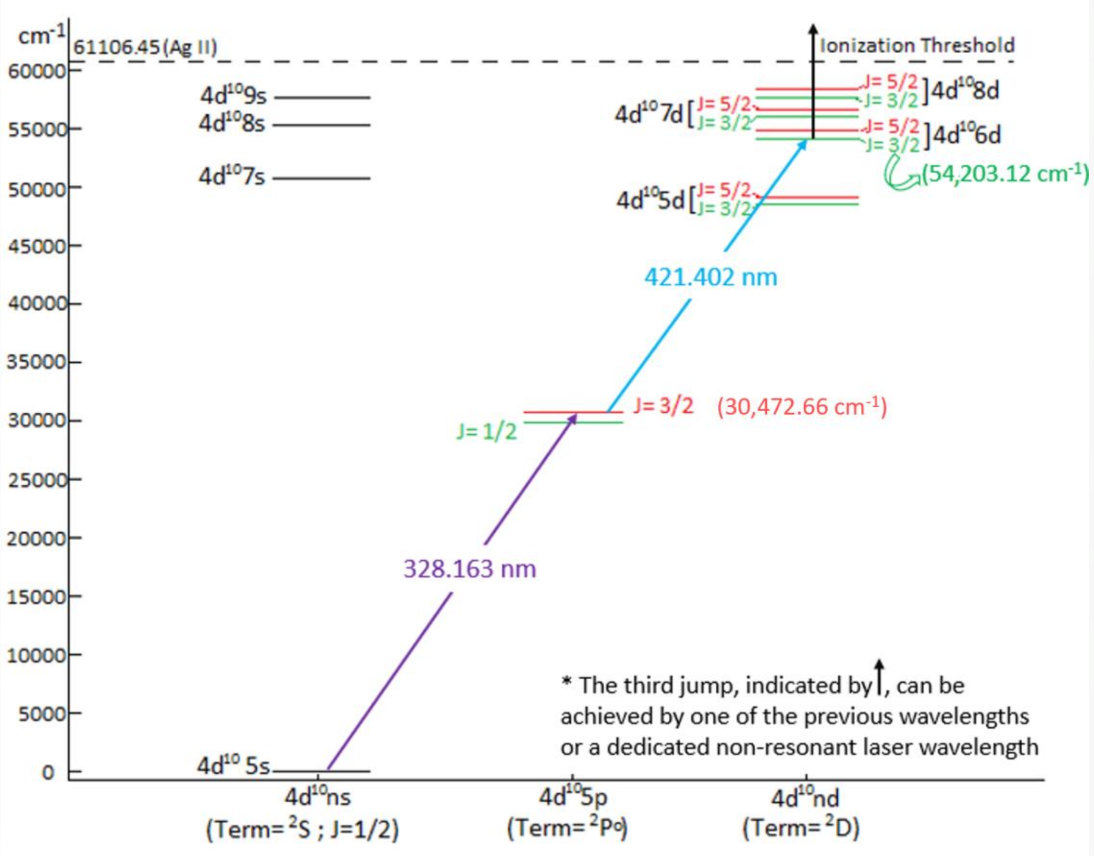
Experimental Set-up



Laser Parameters

Laser	Fundamental			SHG			Power	Pulse length
	λ (nm)	$\Delta\lambda$ (pm)	$\Delta\nu$ (GHz)	λ (nm)	$\Delta\lambda$ (pm)	$\Delta\nu$ (GHz)	(μW)	(ns)
TDL50	656.326	3-3.5	2.1-2.4	328.163	1.1-1.2	3.0-3.3	20-30	20
FL2002	421.402	1-1.1	1.7-1.9	-	-	-	500-550	20

Scheme

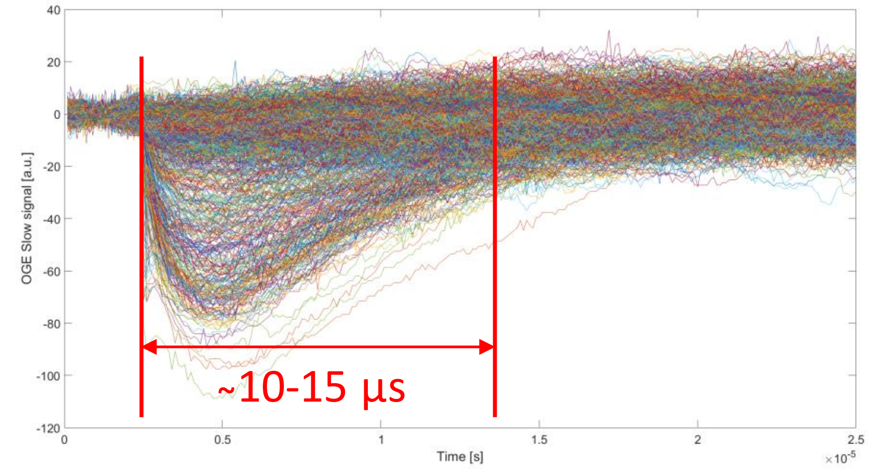


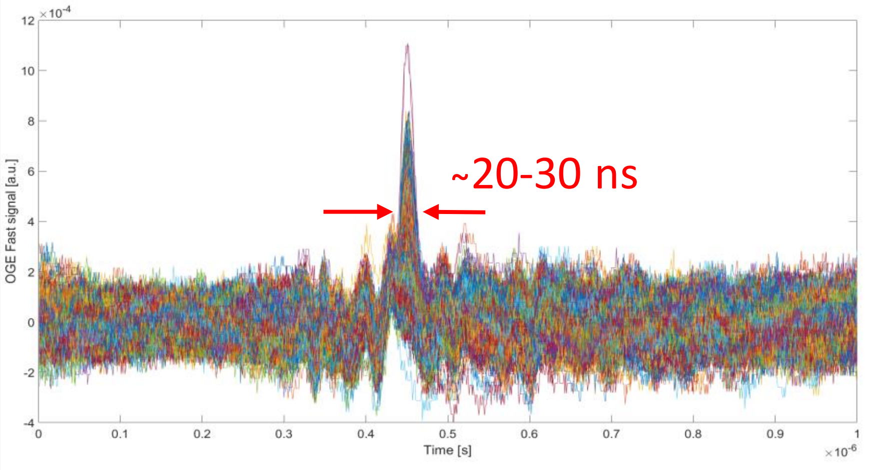
* The third jump, indicated by ↑, can be achieved by one of the previous wavelengths or a dedicated non-resonant laser wavelength

Hollow Cathode Lamps (HCLs): Opto-Galvanic Signals

SLOW Opto-Galvanic Signal:

- ✓ The absorption of laser radiation in the discharge results in a **change in the steady-state population** of bound atomic or molecular levels.
- ✓ Since different levels have different ionization cross-sections, a perturbation to the steady-state situation results in **a net change in the discharge current or equivalently a change in the discharge impedance**.
- ✓ The electric signal detected is the slow signal, negative and lasting µs.



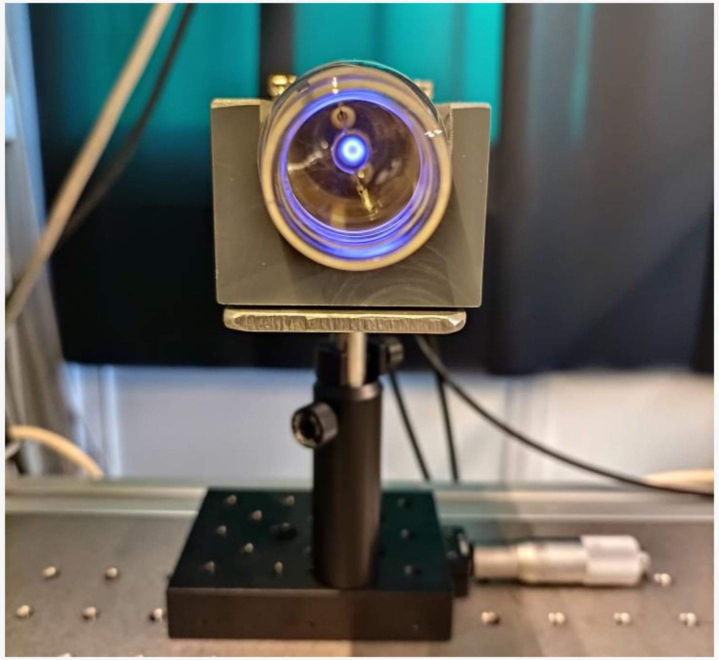


FAST Opto-Galvanic Signal:

- ✓ It is **a direct ionization process** during laser pulse. The laser radiation brings the selectively excited atoms directly to ionization.
- ✓ Electrons are immediately available as carriers.
- ✓ This effect produces a fast-electric signal. It was found that **this fast signal follows the laser pulse temporal behavior** (ns) [2].

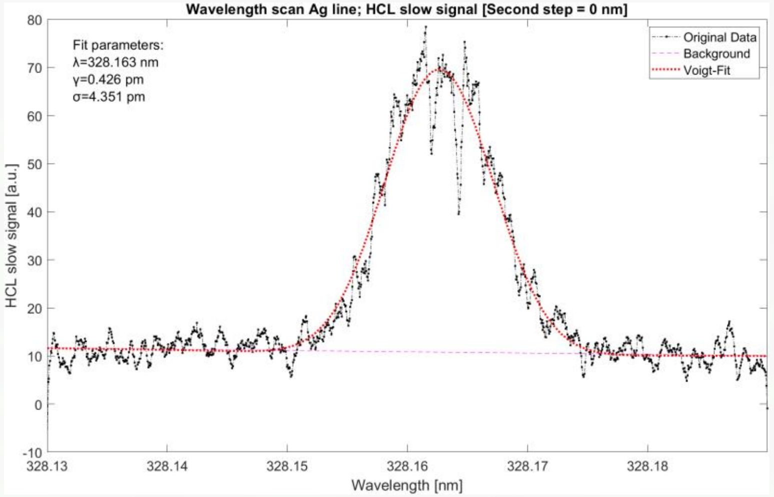
[2] M.Broglia, F.Catoni, P.Zampetti: "Temporal behaviour of the optogalvanic signal in a hollow cathode lamp", Journal de Physique, Colloque C7, supplement au n° 11, Tome 44, novembre 1983

Silver HCL: Scheme test with Slow OGE signals

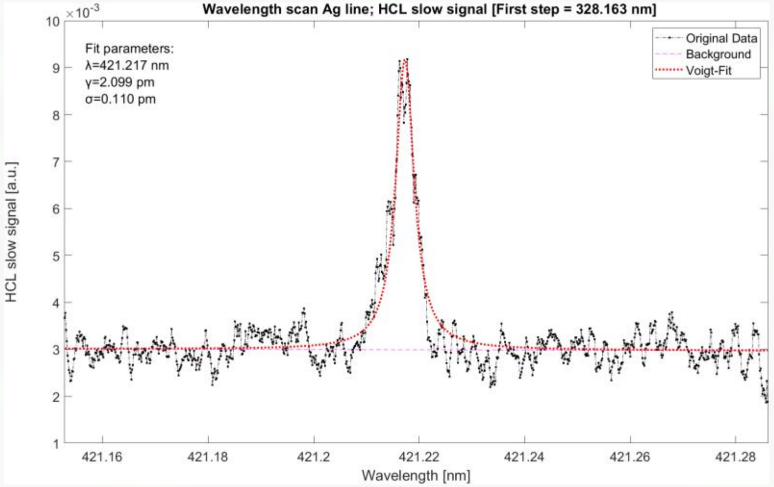


- ✓ Cathode element: Silver
- ✓ Buffer gas: Argon
- ✓ Maximum current: 4 mA
- ✓ Primary emission line: 328.163 nm

First step



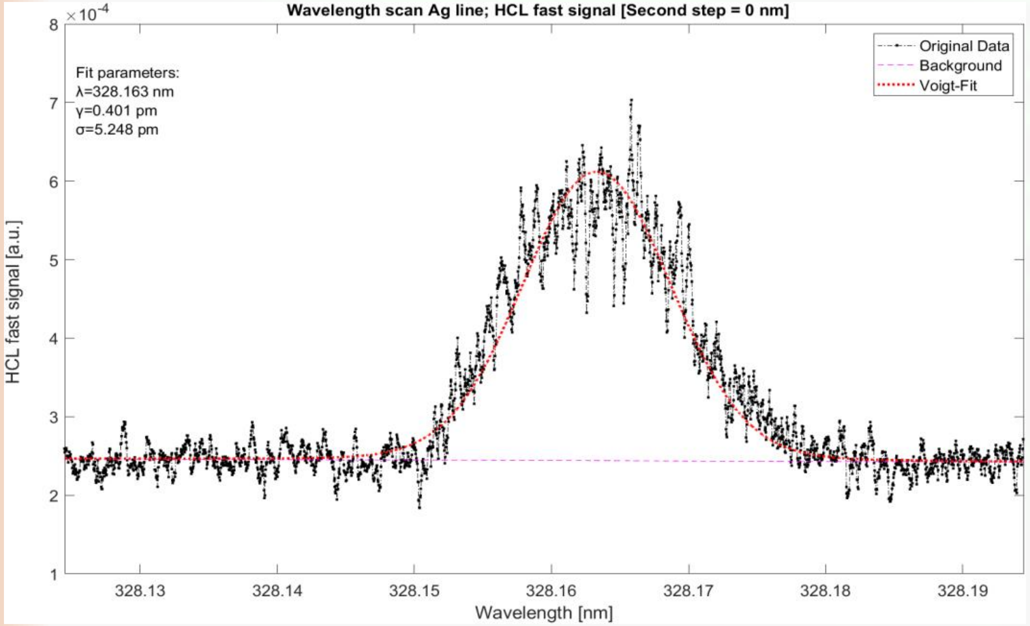
Second step

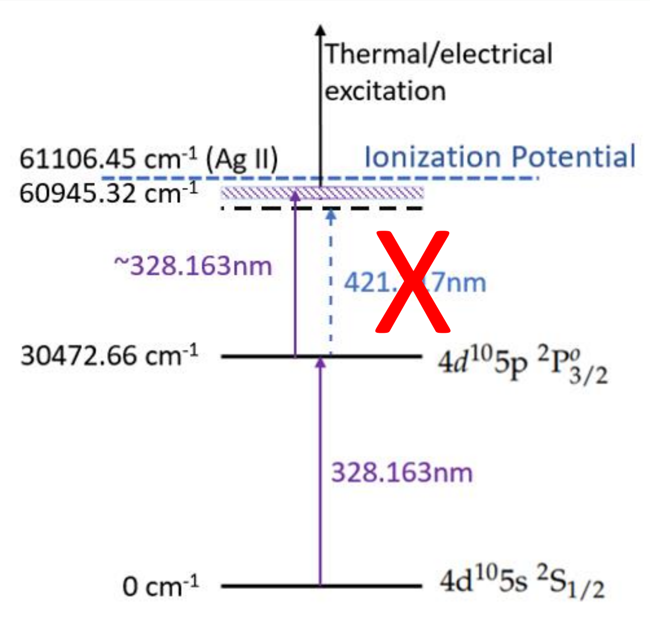


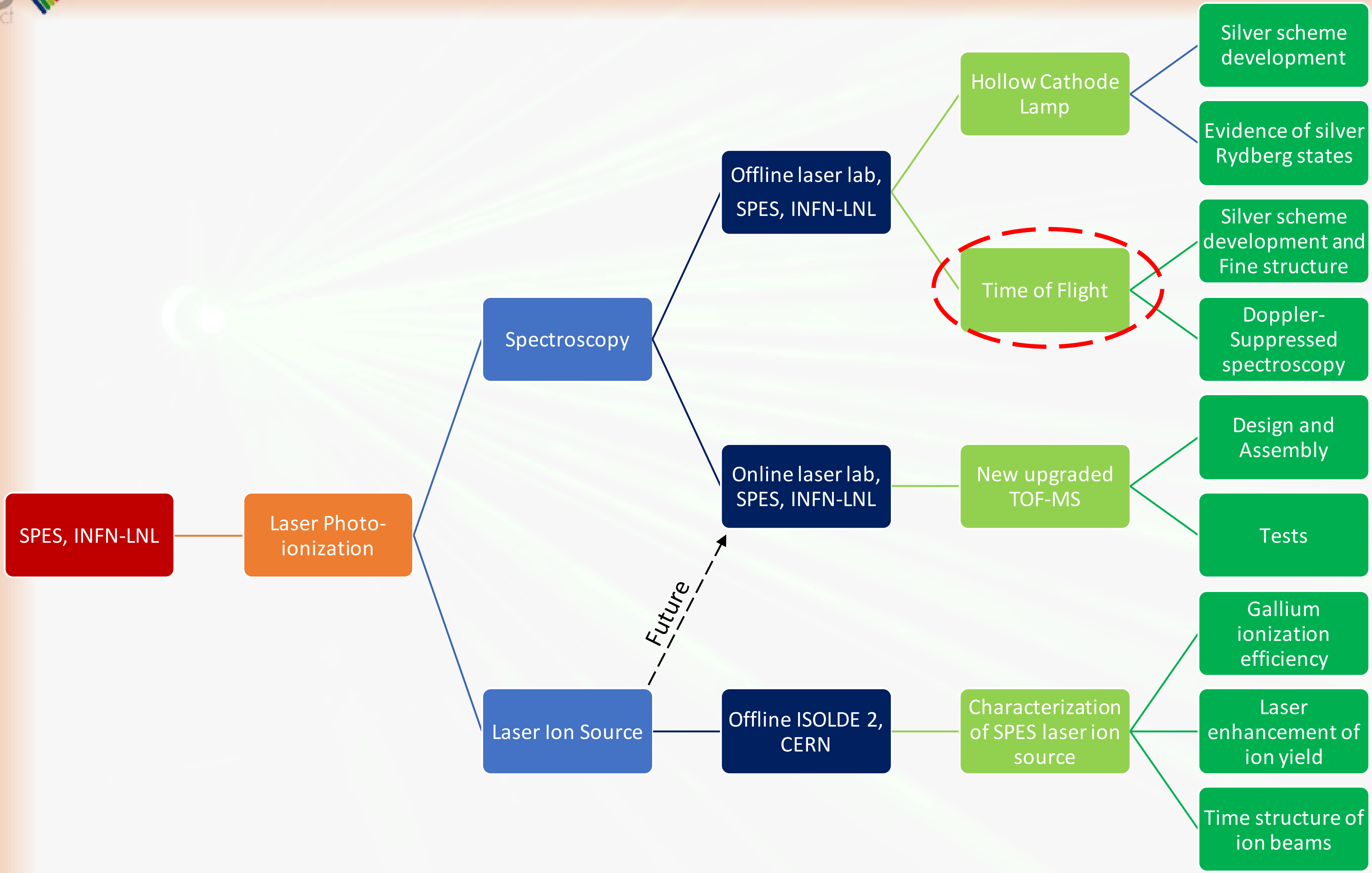
Silver HCL: Rydberg states with Fast OGE signals

- ✓ Fast signals are observed only when there is direct ionization.
- ✓ Only 328.163 nm is injected into the HCL.
- ✓ Two photons of 328.163 nm would only take the electrons to roughly 60945.32 cm^{-1}, not beyond the ionization potential 61106.45cm^{-1}.

- ✓ Shows the presence of high-lying Rydberg states around the energy value of 60945.32 cm^{-1}.







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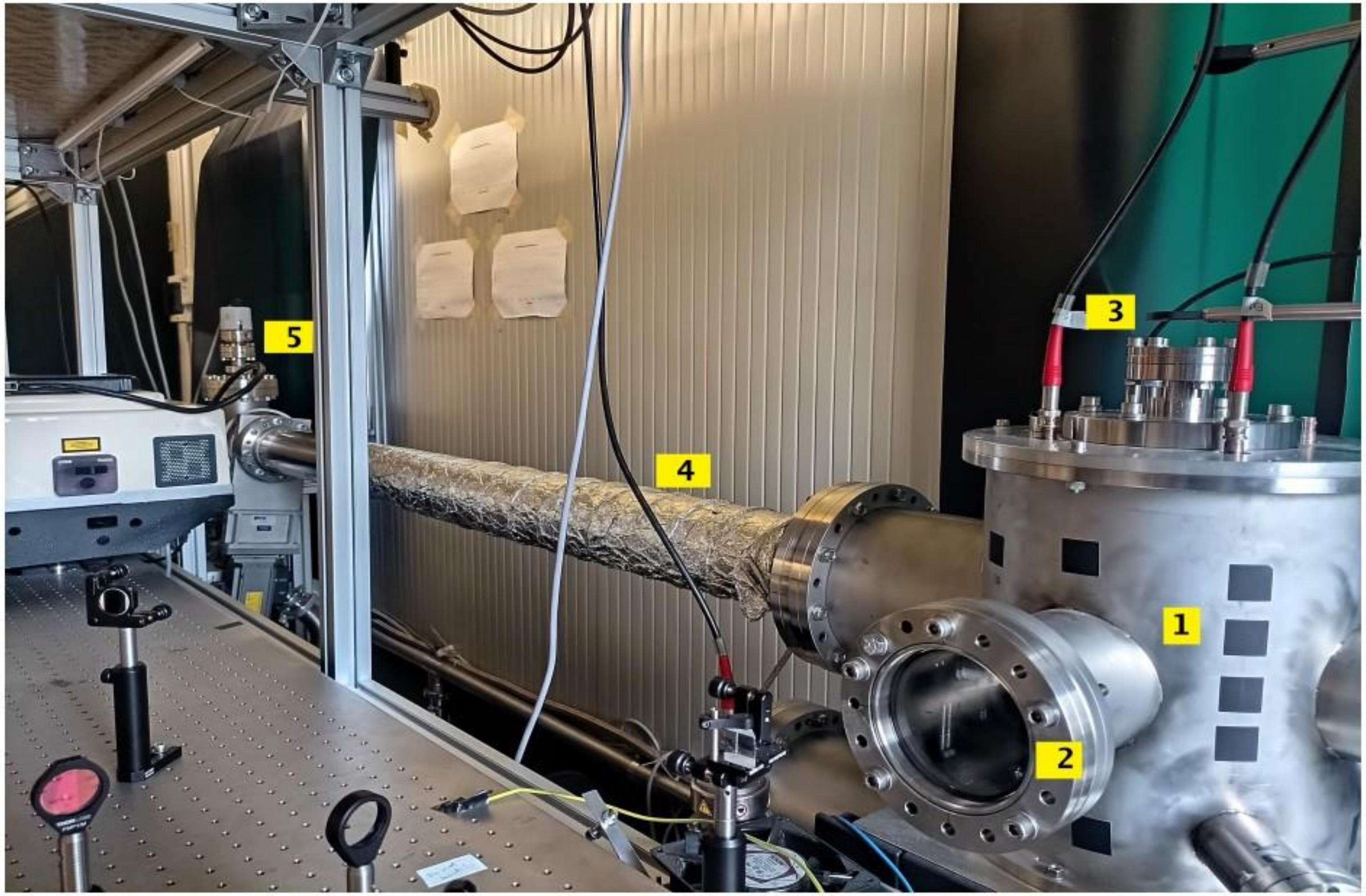
Future

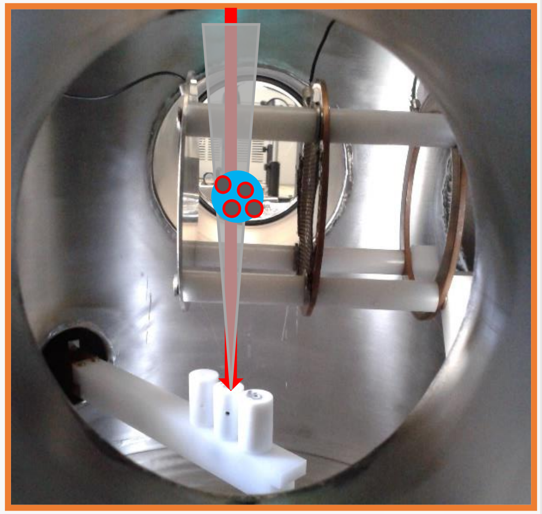
Home-made ToF (using ablation source)



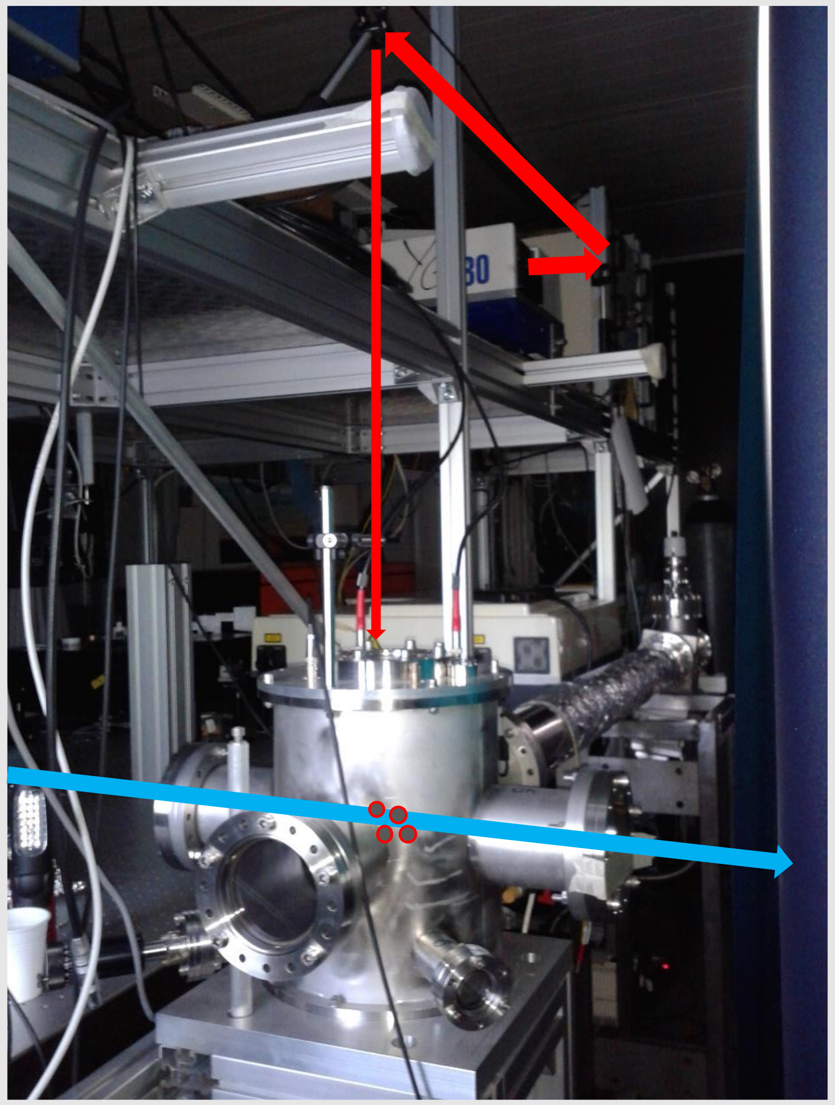
1. Ionization chamber (10^{-7} mbar)
2. Excitation laser(s) entrance window
3. Ablation laser (1064 nm) entrance window
4. Ion flight tube (1.90 meters)
5. Micro Channel Plate (MCP) detector

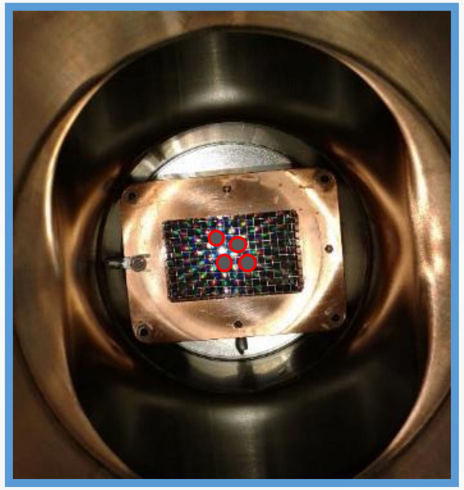
Home-made ToF (using ablation source)

Top view



Front view
(as seen by the ionization laser)





1) Ablation (1064 nm, 10 Hz, 20 ns, 2J)
2) Plume Expansion (roughly 30-35 µs)
3) Photoionization
4) Ion creation and flight (26-27 µs)
5) Collection

Time of Flight measurements

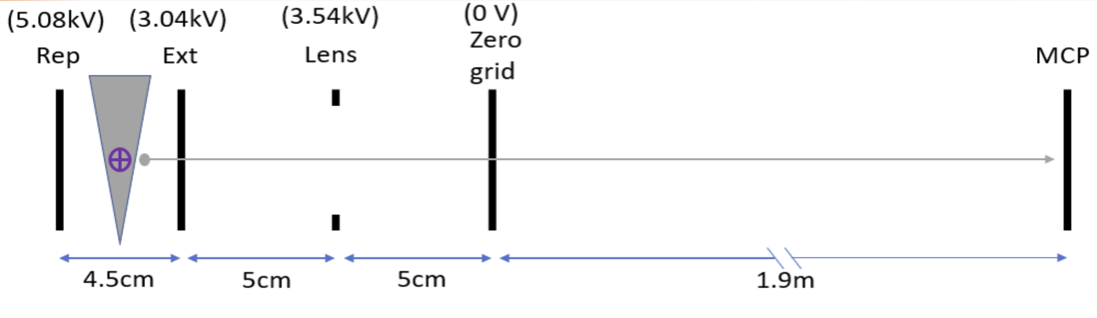
1. Photo-ionization scheme of silver: 328.163 + 421.402 nm
    - ✓ Synchronize the laser pulses in time
    - ✓ Overlap in space
    - ✓ Frequency scan of one at a time

2. Fine structure of the silver level: $4d^{10}6d\ ^2D$ (J=5/2 and J=3/2)
    - ✓ A large frequency scan of roughly 560 GHz in the second step

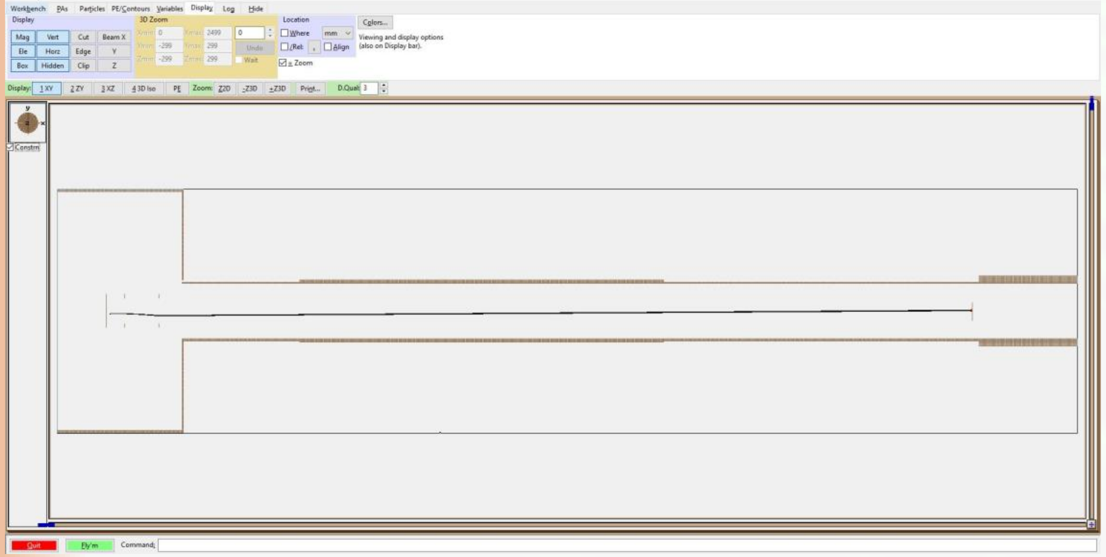
3. Doppler suppressed spectroscopy in the hot ablation plume
    - ✓ Reducing the atom-laser interaction volume
    - ✓ Probe different velocity components for the different excitation laser

Detection and Identification of isotopes

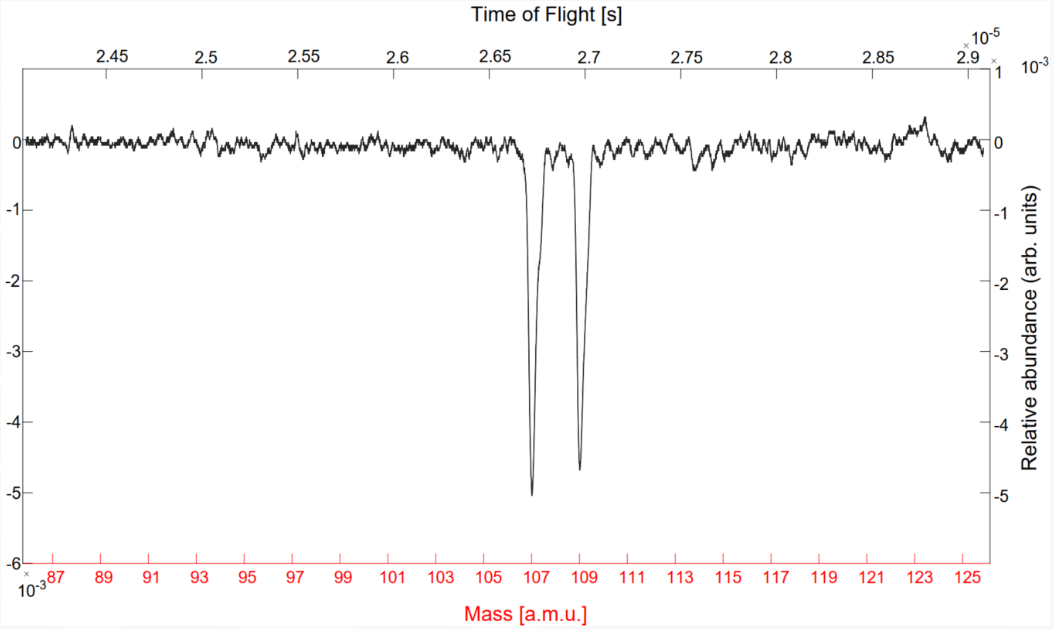
Ion optics and applied voltages



SIMION simulation

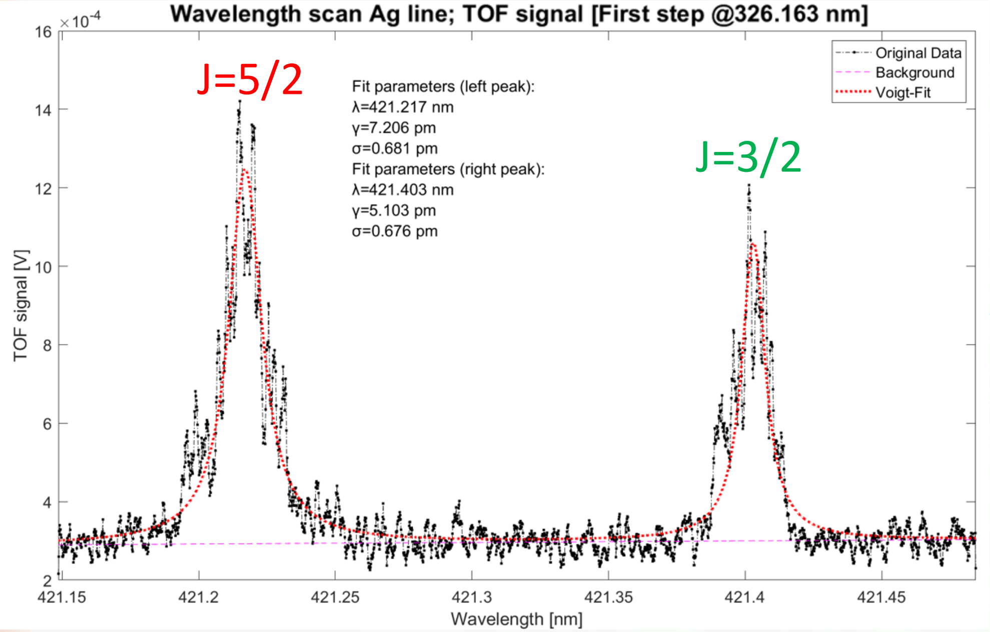


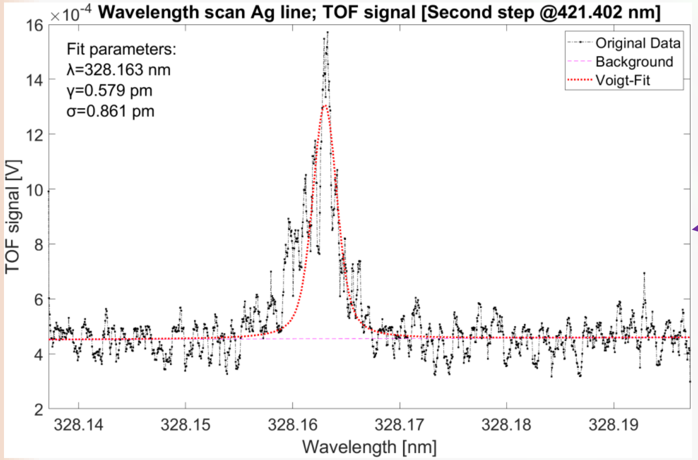
Collected silver isotopes

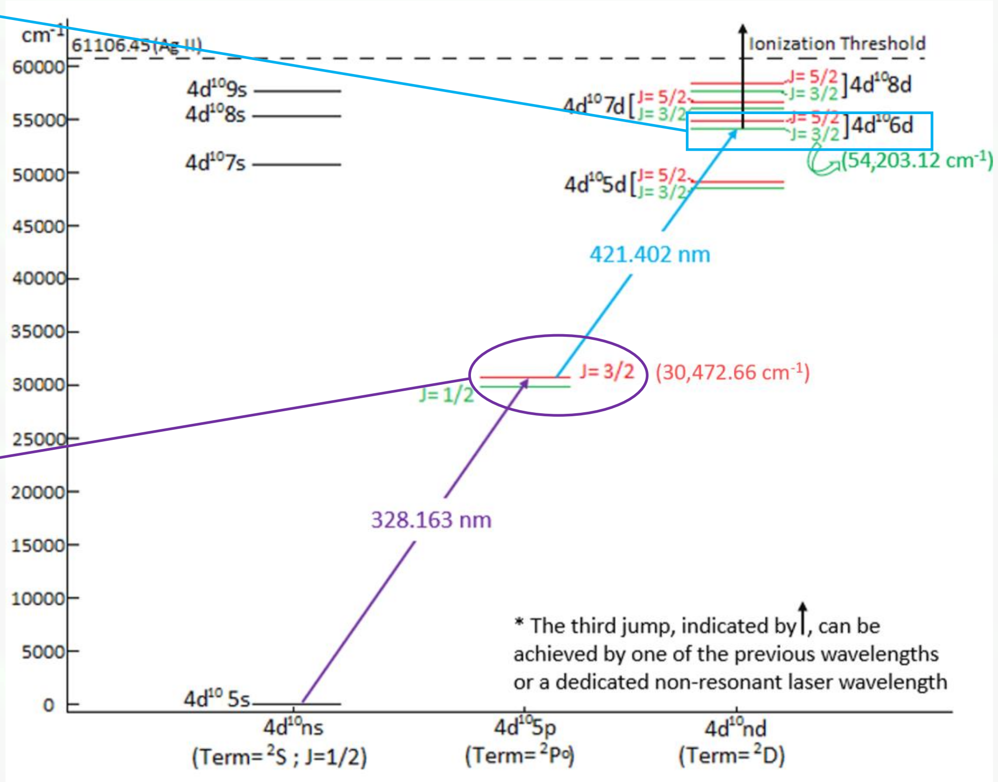


A SIMION simulation with the applied electrode voltages and the desired ion (isotope) gives the approximated time of flight for the species to arrive to the detector after the flight, immediately after it is formed.

Scheme test and Fine structure







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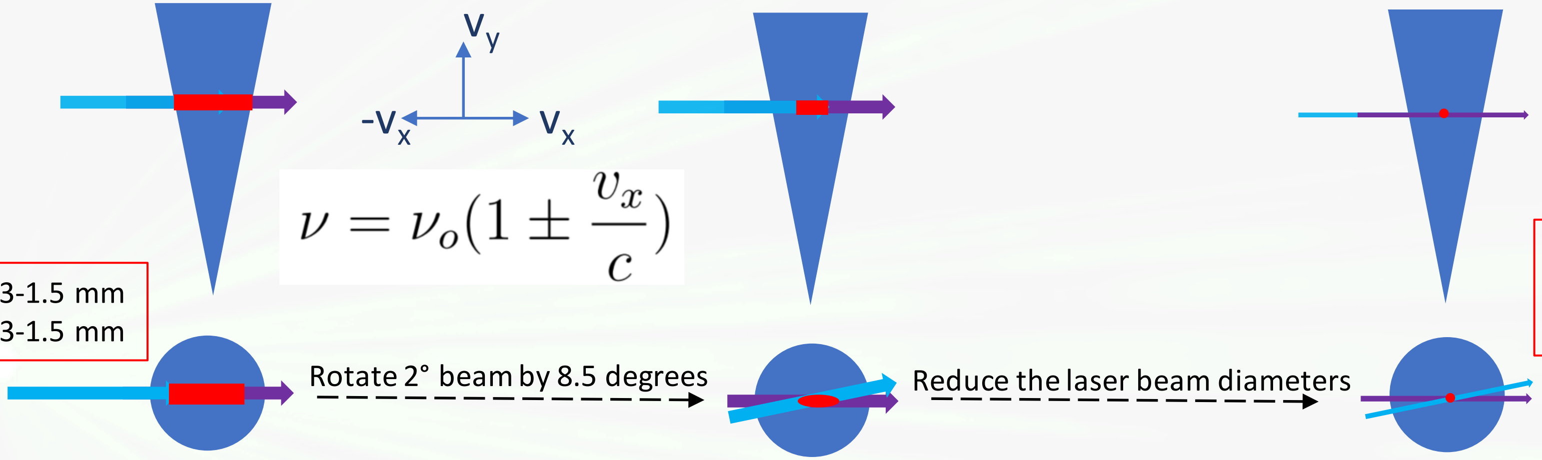
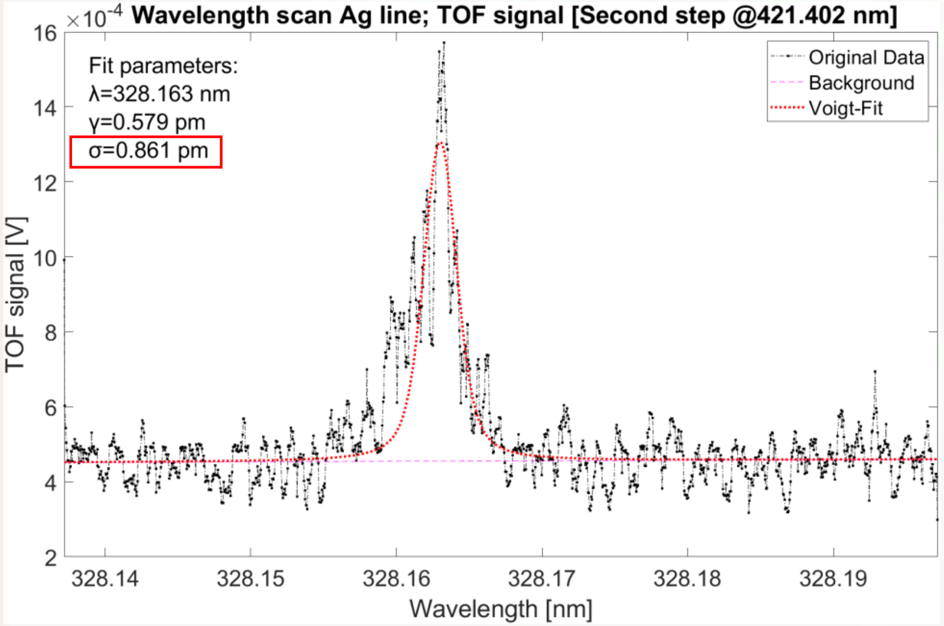
Doppler broadening and suppression

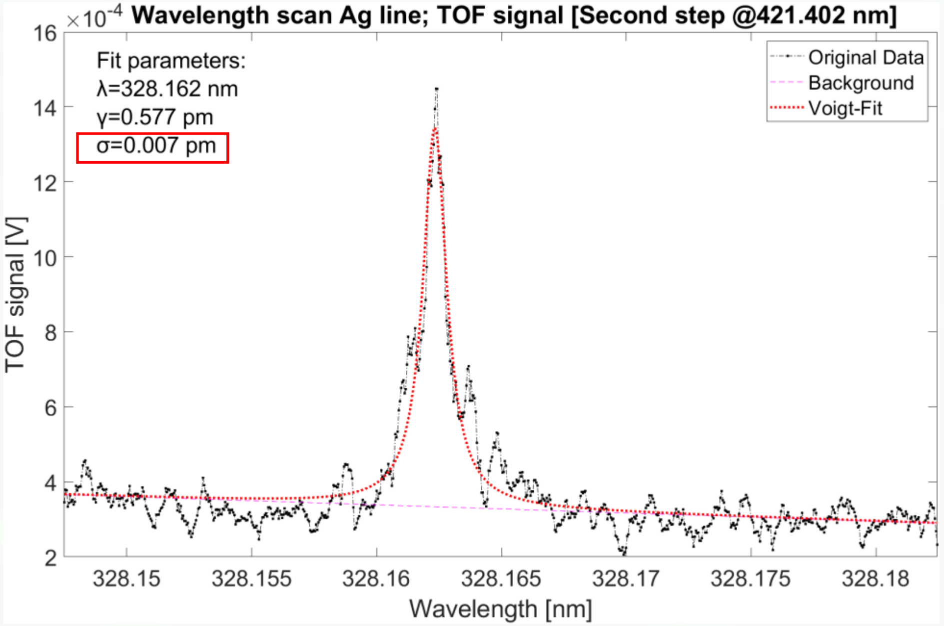


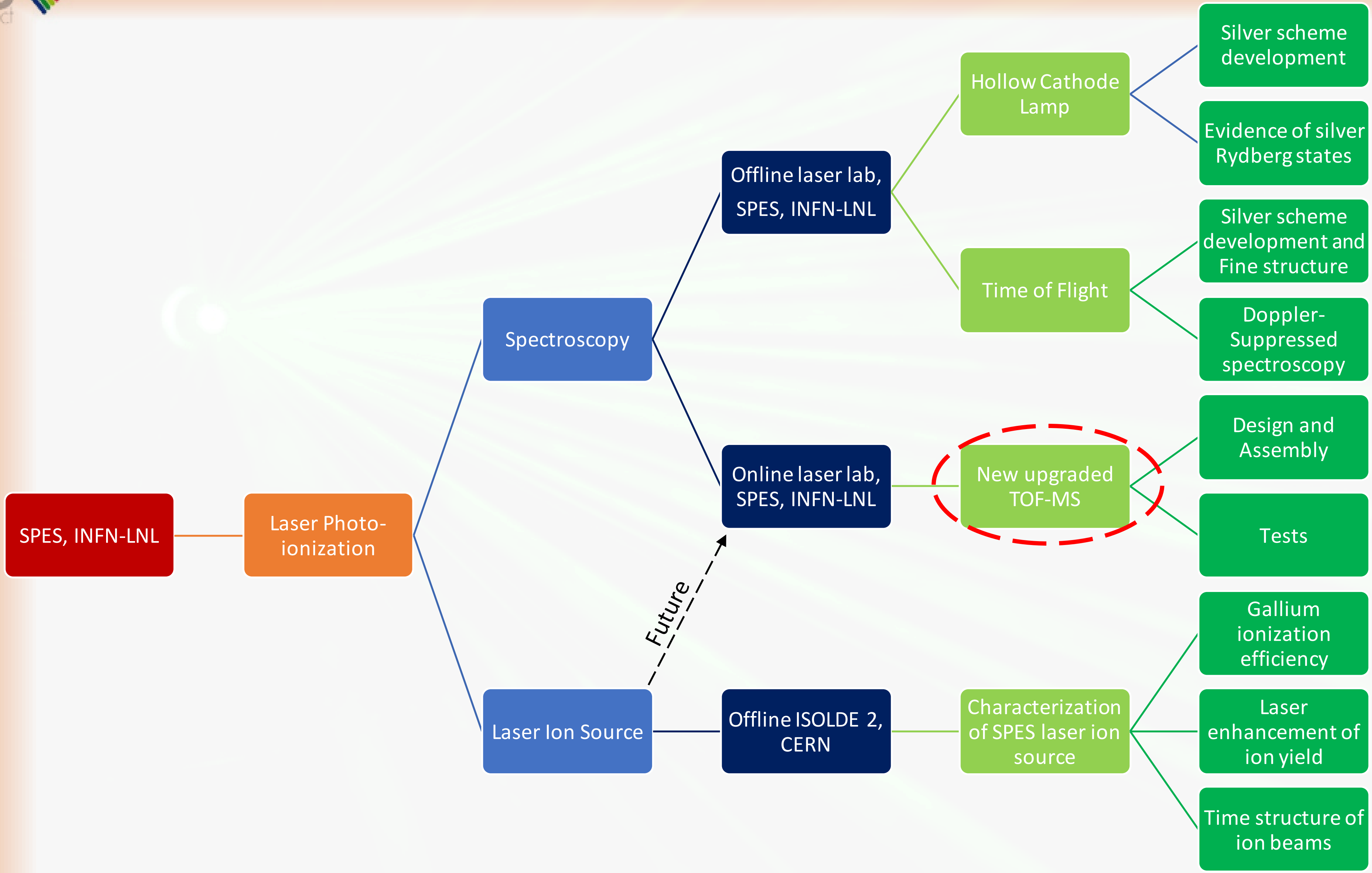
$$\nu = \nu_o(1 \pm \frac{v_x}{c})$$

1° laser beam = 1.3-1.5 mm
2° laser beam = 1.3-1.5 mm

1° laser beam = 0.6-0.8 mm
2° laser beam = 1.0-1.2 mm
Final interaction volume < 0.5 mm^3







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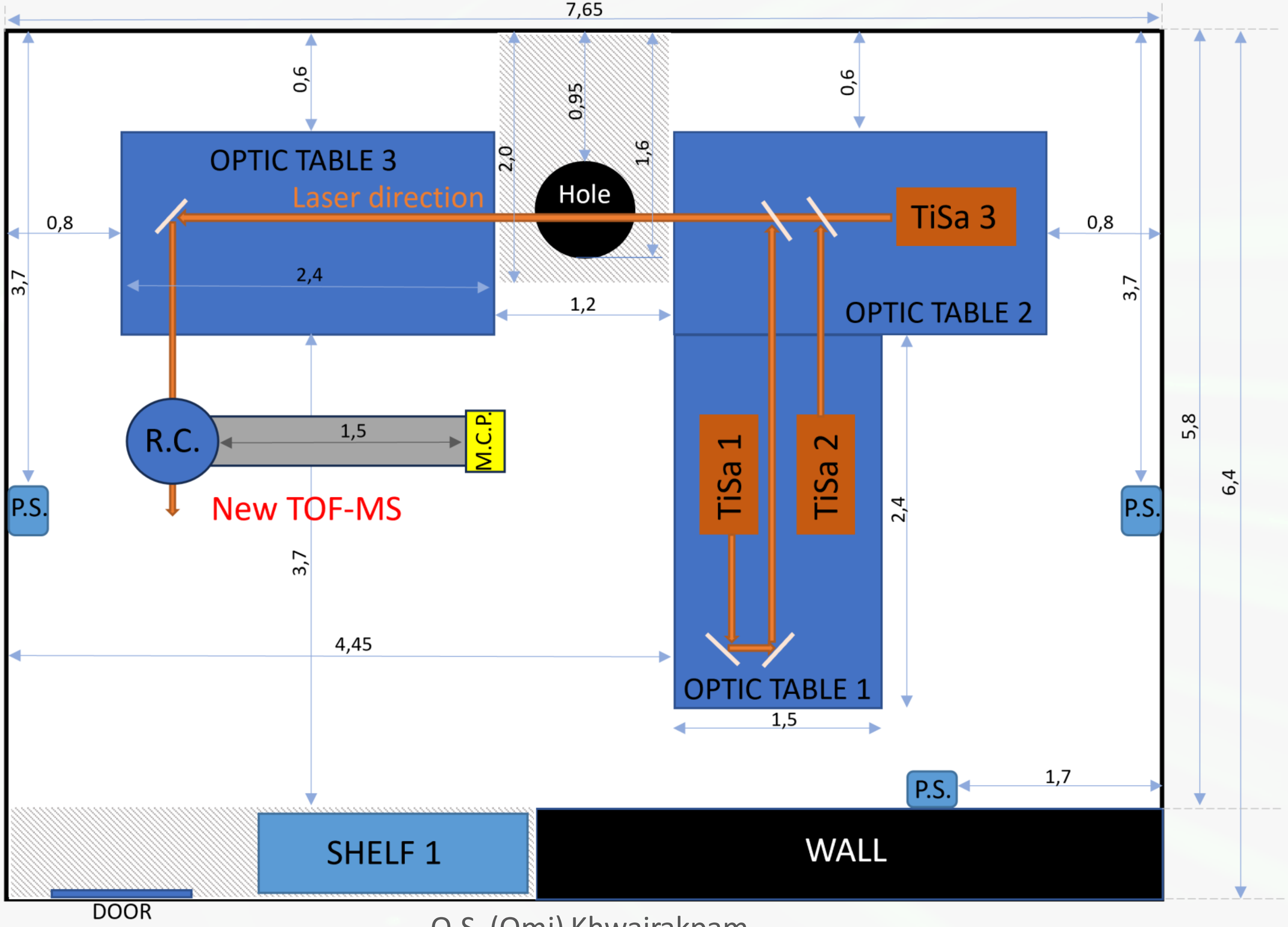
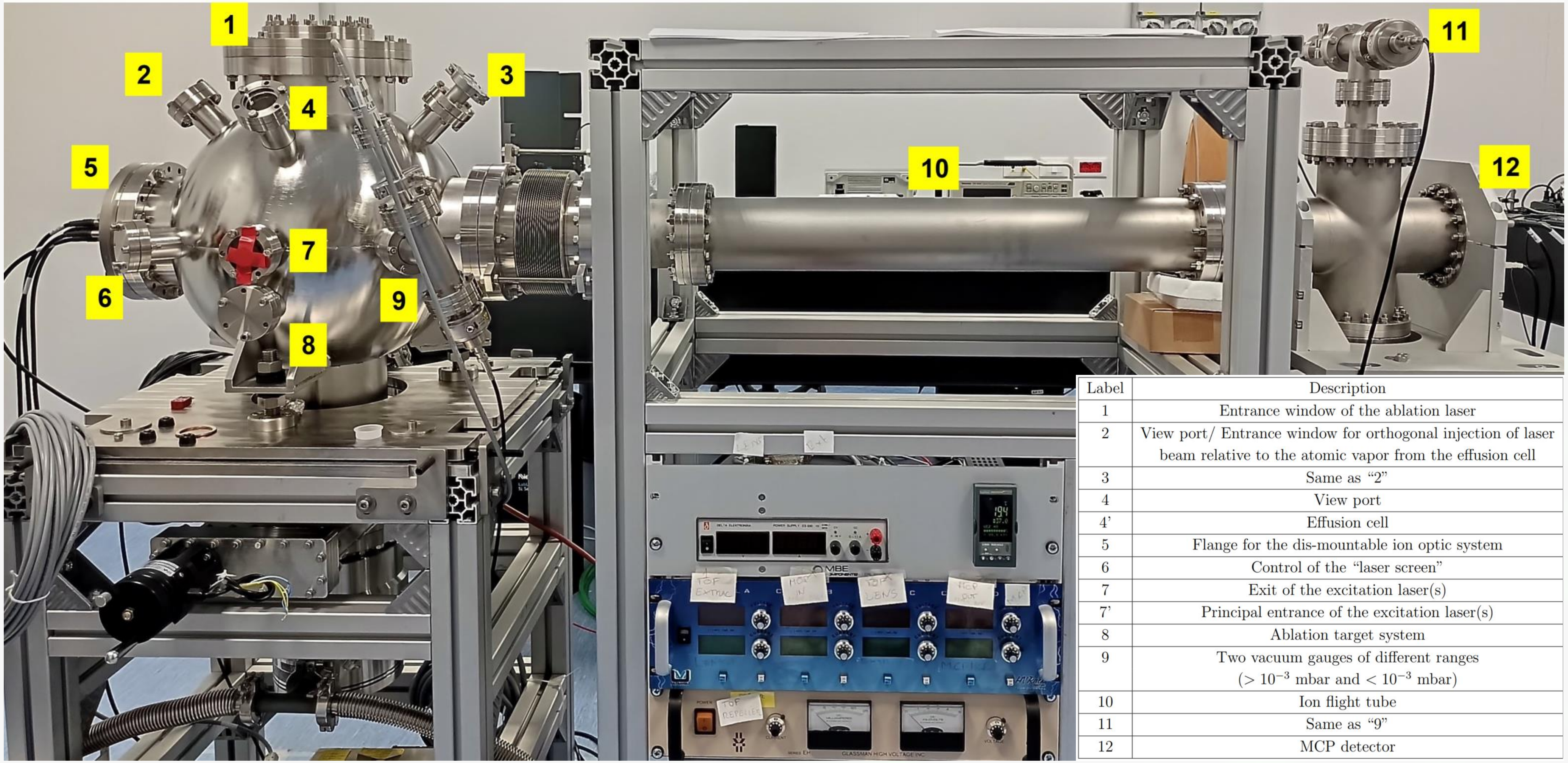
Next Topic: New TOF-MS in the SPES online laser lab

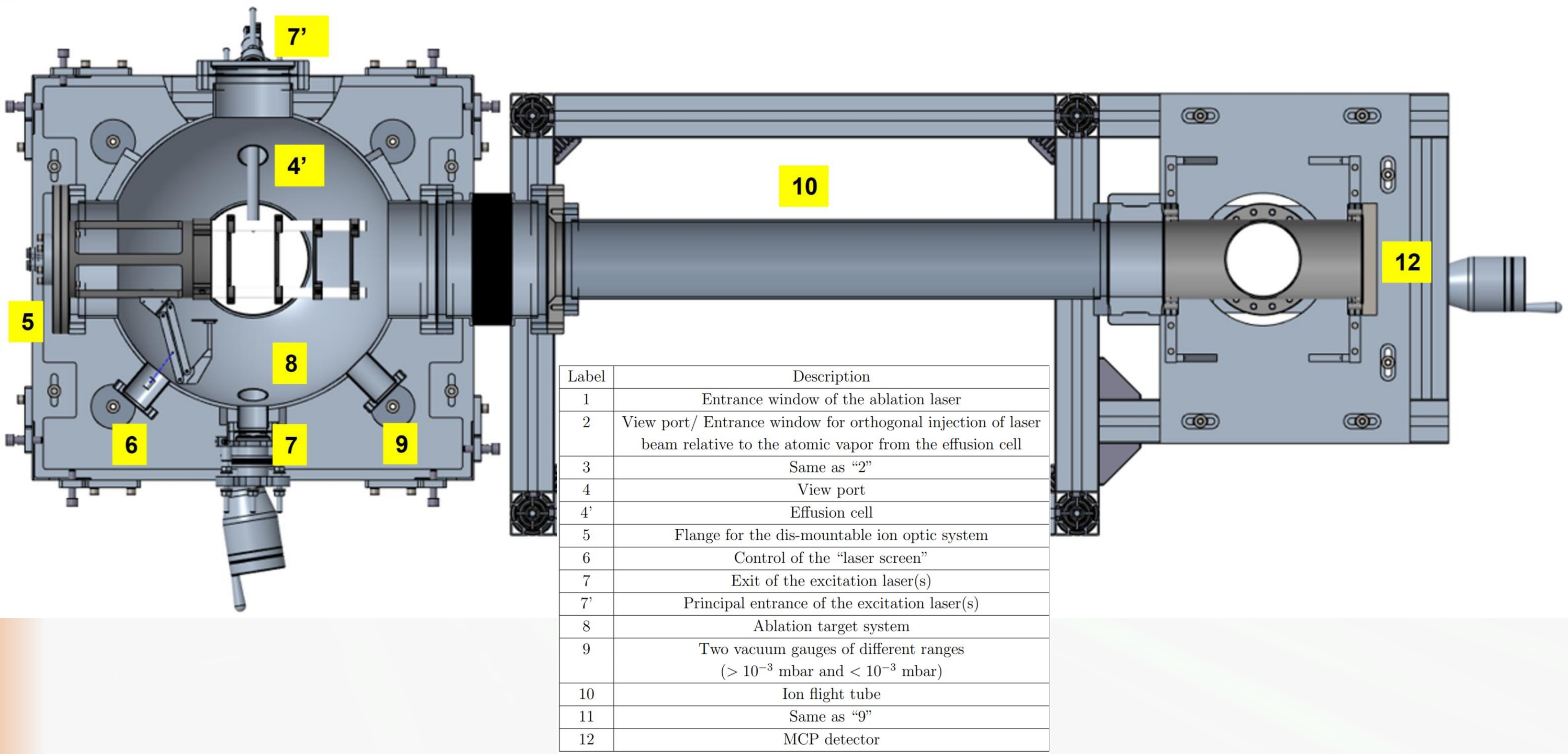


Assembled TOF- MS



Label	Description
1	Entrance window of the ablation laser
2	View port/ Entrance window for orthogonal injection of laser beam relative to the atomic vapor from the effusion cell
3	Same as “2”
4	View port
4’	Effusion cell
5	Flange for the dis-mountable ion optic system
6	Control of the “laser screen”
7	Exit of the excitation laser(s)
7’	Principal entrance of the excitation laser(s)
8	Ablation target system
9	Two vacuum gauges of different ranges ($> 10^{-3}$ mbar and $< 10^{-3}$ mbar)
10	Ion flight tube
11	Same as “9”
12	MCP detector

Sectioned Top View: TOF- MS



Label	Description
1	Entrance window of the ablation laser
2	View port/ Entrance window for orthogonal injection of laser beam relative to the atomic vapor from the effusion cell
3	Same as “2”
4	View port
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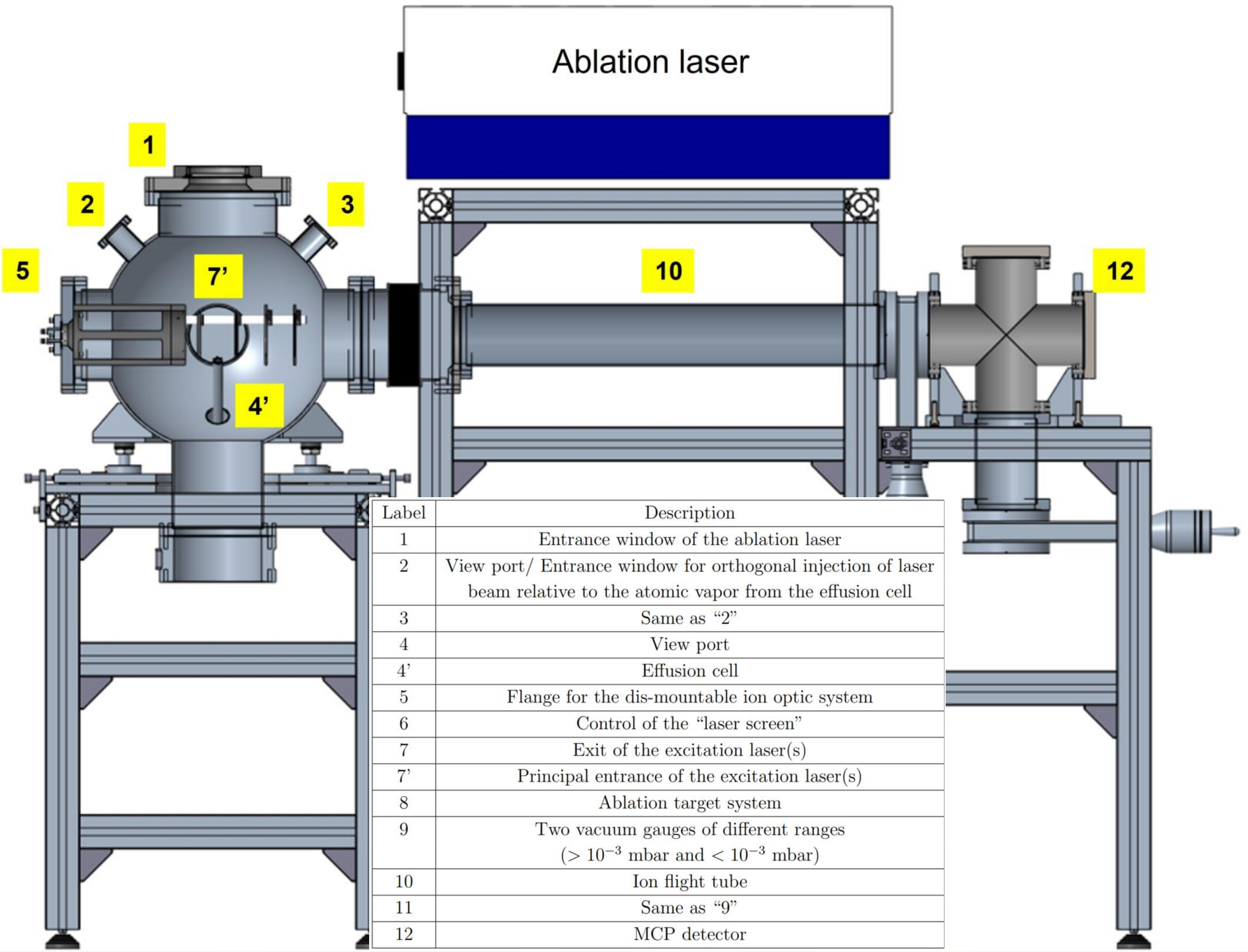
5	Flange for the dis-mountable ion optic system
6	Control of the “laser screen”
7	Exit of the excitation laser(s)
7’	Principal entrance of the excitation laser(s)
8	Ablation target system
9	Two vacuum gauges of different ranges ($> 10^{-3}$ mbar and $< 10^{-3}$ mbar)
10	Ion flight tube
11	Same as “9”
12	MCP detector

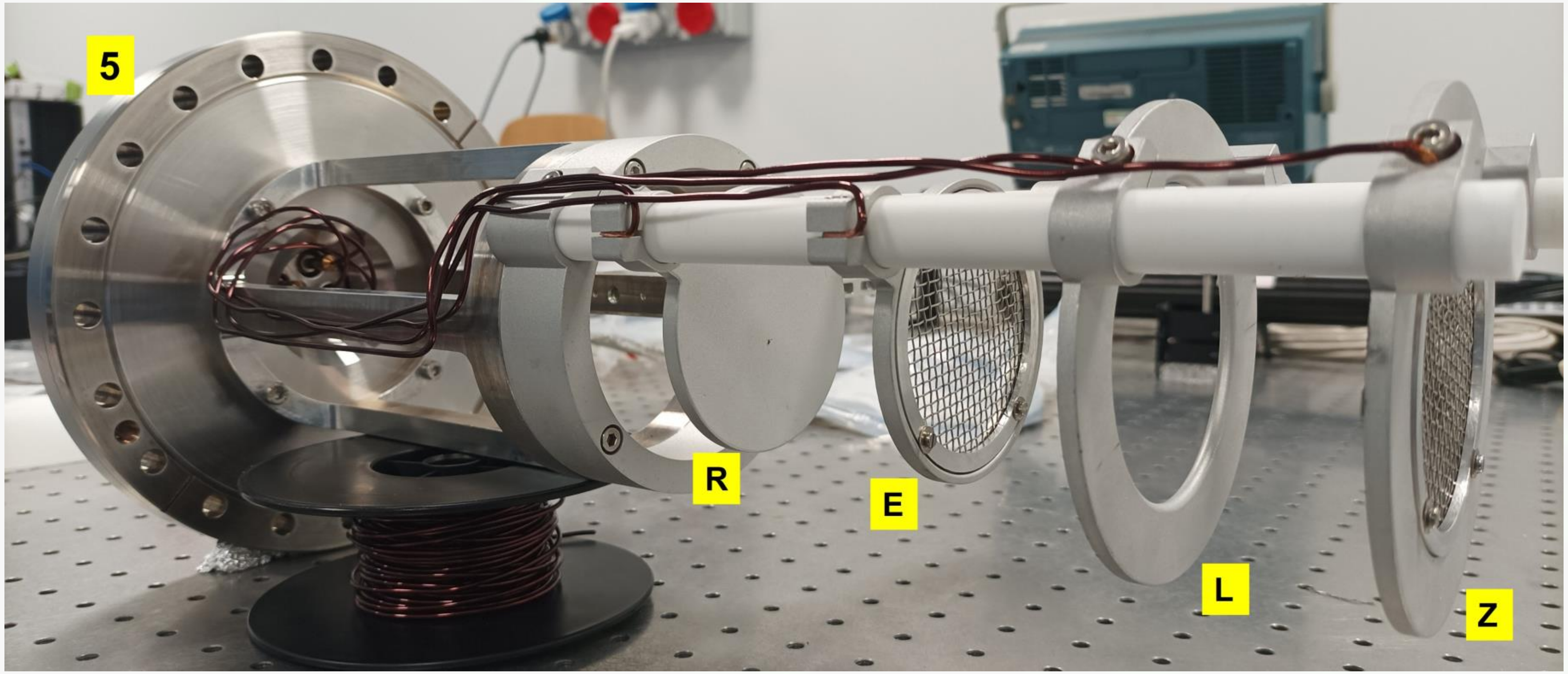
Sectioned Side View: TOF- MS



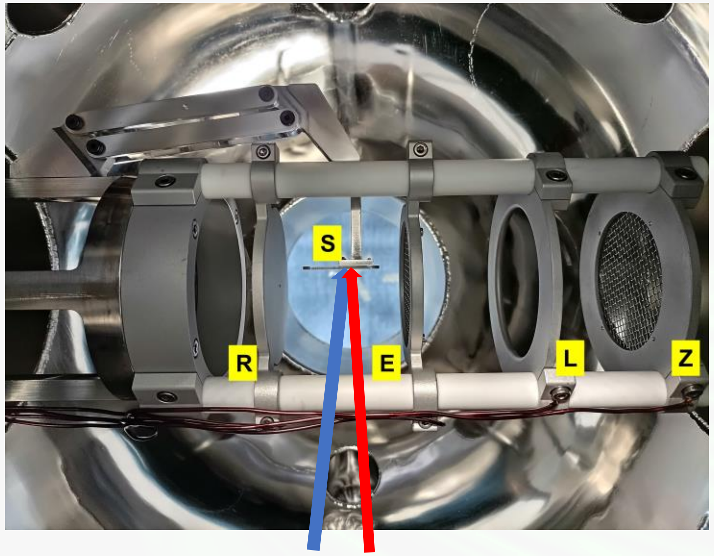
Label	Description
1	Entrance window of the ablation laser
2	View port/ Entrance window for orthogonal injection of laser beam relative to the atomic vapor from the effusion cell
3	Same as "2"
4	View port
4'	Effusion cell
5	Flange for the dis-mountable ion optic system
6	Control of the "laser screen"
7	Exit of the excitation laser(s)
7'	Principal entrance of the excitation laser(s)
8	Ablation target system
9	Two vacuum gauges of different ranges ($> 10^{-3}$ mbar and $< 10^{-3}$ mbar)
10	Ion flight tube
11	Same as "9"
12	MCP detector

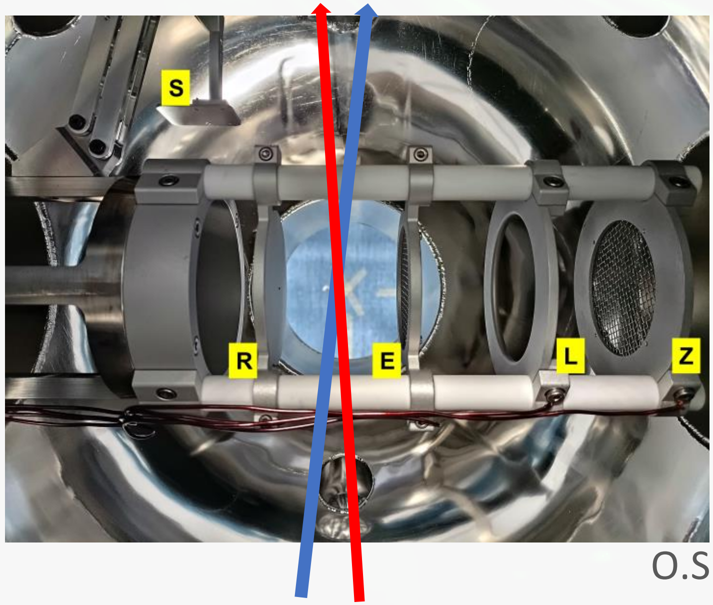
O.S. (Omi) Khwairakpam

Completely dismountable ion optic system



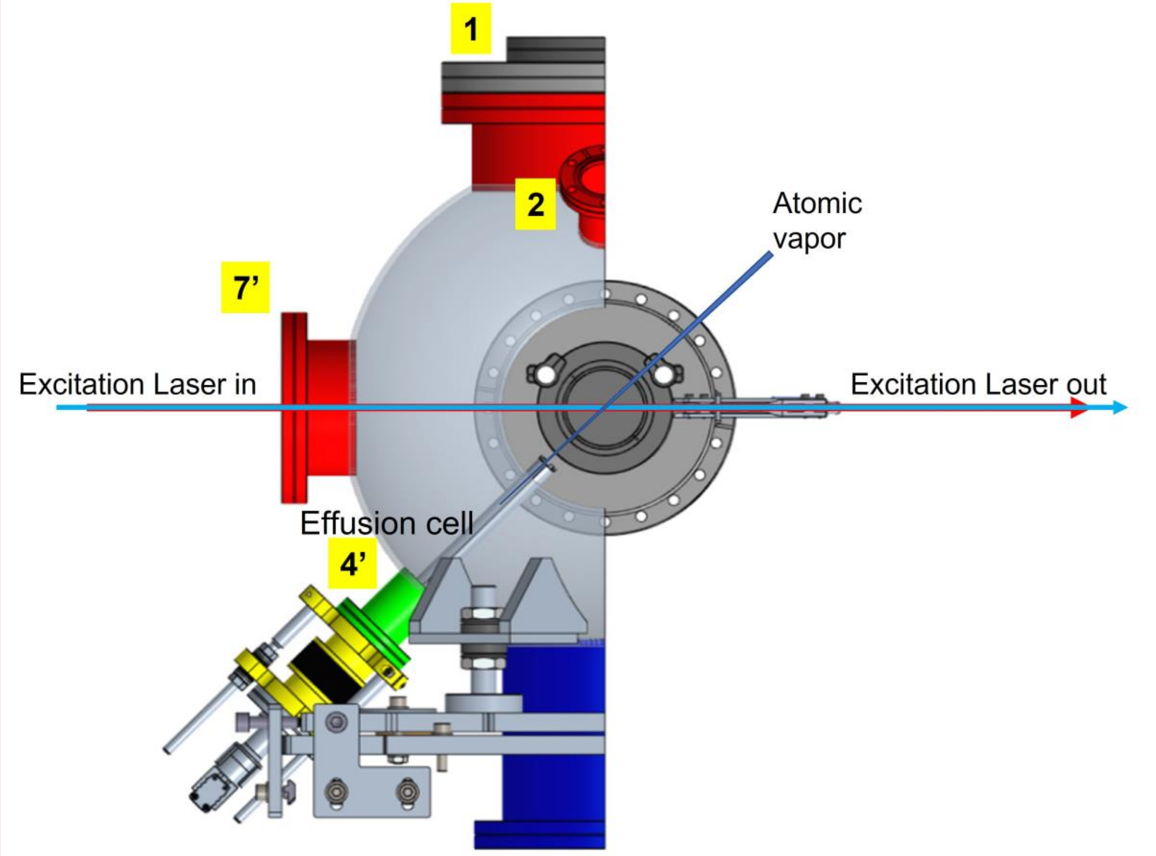
Laser Screen: To visualize overlap of laser beams



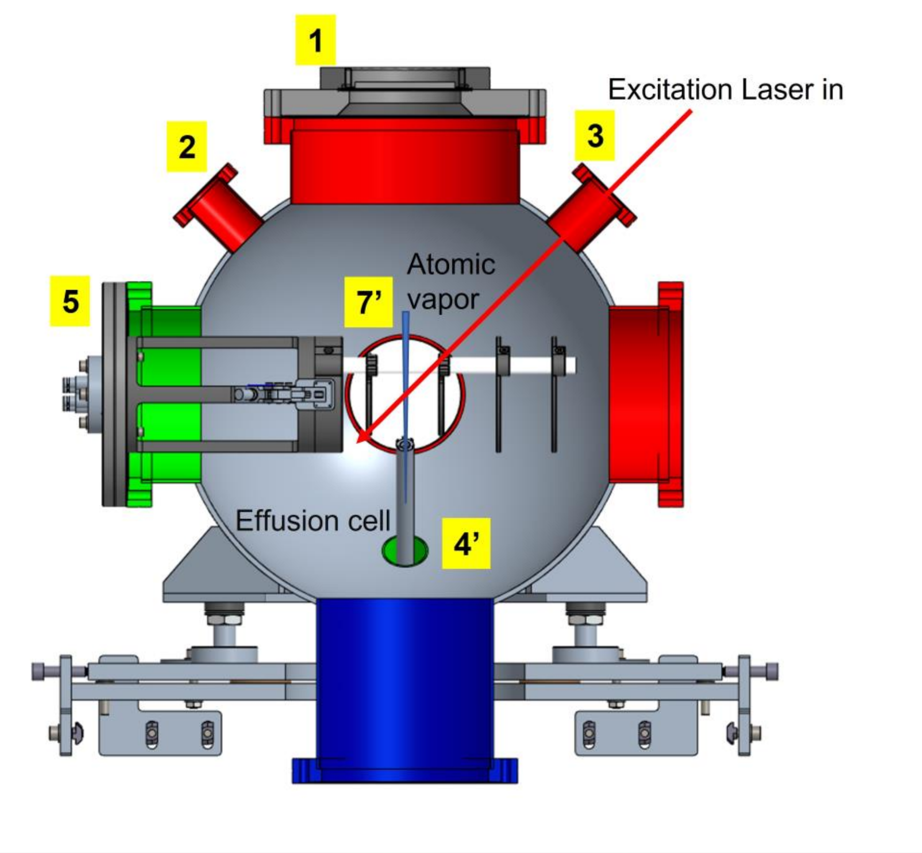


Laser-Atom Interaction
(Geometry to suppress Doppler broadening)

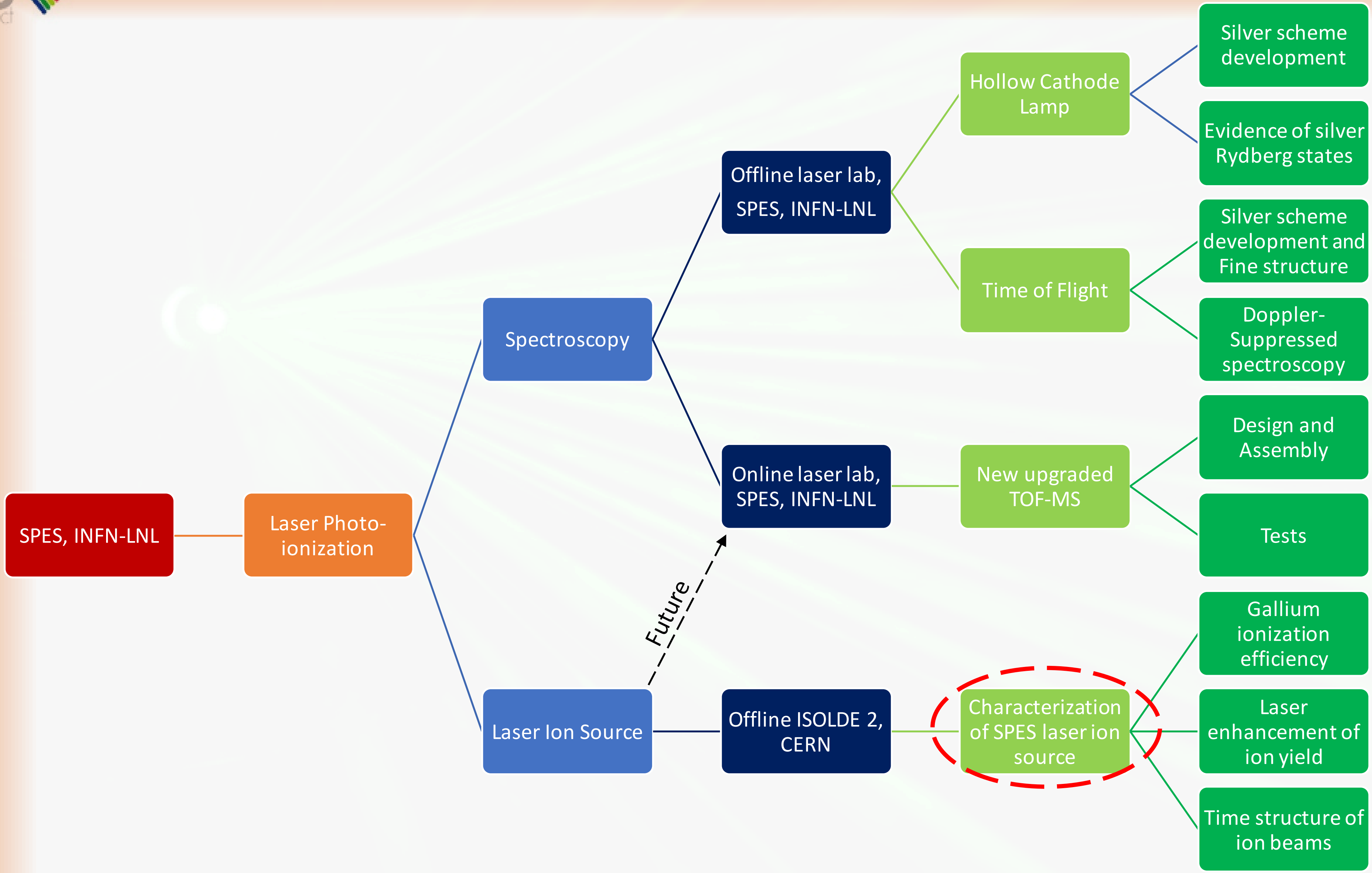
Laser beams longitudinal to each other



Laser beams orthogonal to each other



>> Should allow Doppler suppressed laser spectroscopy



SPES, INFN-LNL

Laser Photo-ionization

Spectroscopy

Laser Ion Source

Offline laser lab, SPES, INFN-LNL

Online laser lab, SPES, INFN-LNL

Offline ISOLDE 2, CERN

Future

Hollow Cathode Lamp

Time of Flight

New upgraded TOF-MS

Characterization of SPES laser ion source

Silver scheme development

Evidence of silver Rydberg states

Silver scheme development and Fine structure

Doppler-Suppressed spectroscopy

Design and Assembly

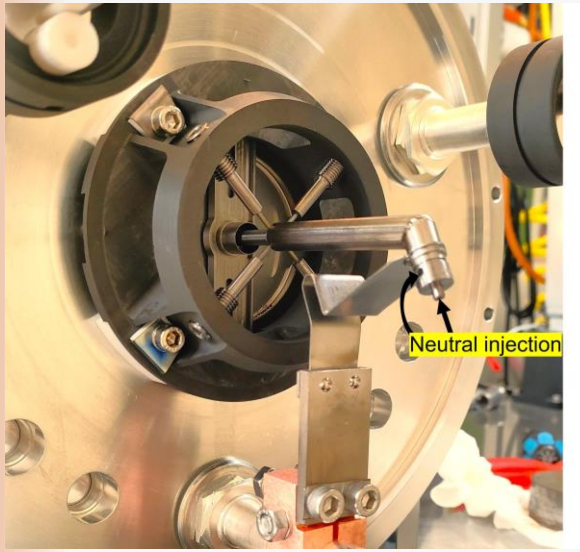
Tests

Gallium ionization efficiency

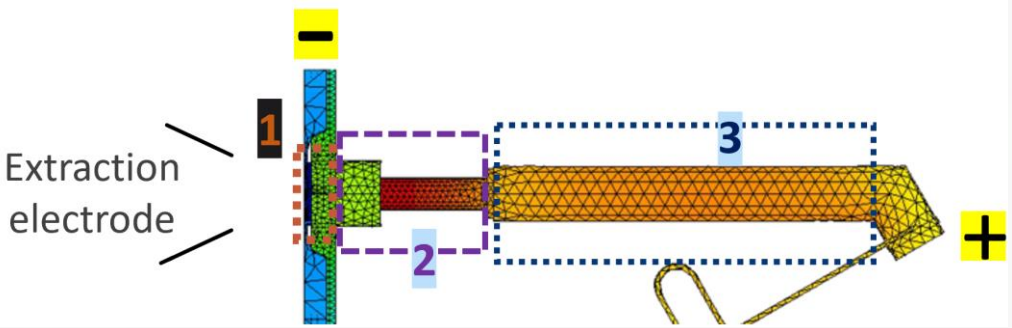
Laser enhancement of ion yield

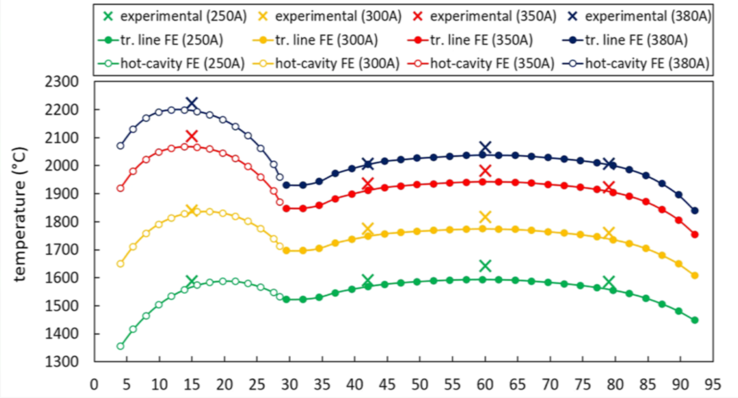
Time structure of ion beams

Next topic: SPES Laser Ion Source



- ✓ **Material: Tantalum**
  Work function = 4.28 eV
- ✓ **Region 1: Orifice of the ion source**
- ✓ **Region 2: Hot cavity**
  Length: 33 mm
  Internal diameter: 3.1 mm
  External diameter: 5.1 mm
- ✓ **Region 3: Transfer line**
  Length: 70 mm
  Internal diameter: 8 mm
  External diameter: 8.8 mm





M. Manzolaro et al. http://dx.doi.org/10.1063/1.4998246

The SPES laser ion source is essentially a hot cavity laser ion source.

- ✓ Optimized thermal profile.
- ✓ The principle is to introduce tunable laser beams into the hot cavity.

Requirements of a good Laser Ion Source (LIS)

- ✓ Maximum interaction of the lasers with the atoms, spatially and temporally.
- ✓ **Laser-atom interaction** is the key. We would like that there are more atoms than ions for the particular element.
- ✓ In other words, it is essential that **the surface ionization** of this element in the hot cavity is **low.**
- ✓ Also, **minimal high temperature** needs to be maintained to avoid “sitting” of the neutrals on the walls of the ion source.
- ✓ This temperature is required to maintain **good confinement** of the laser ions inside the hot cavity.

Confinement of Ions inside the Hot cavity

Q. What happens if we lose all the ions we produce via collision with the wall?

✓ A **thermal plasma** is formed inside the hot cavity. This plasma has **negative potential (in terms of a few volts)** respect to the wall of the cavity, and which **confines the ions inside the volume**. This potential is :

Sum of all the surface positive ions

Confining potential $\phi_p = \frac{k_B T}{2e} ln \frac{\boxed{n_{is}}}{n_{es}}$

M. Huyse et al. https://doi.org/10.1016/0167-5087(83)91284-X

Ø is the work function of the cavity material

$$\boxed{n_{\mathrm{es}} = 2\left(\frac{2\pi mkT}{h^2}\right)^{3/2} \exp(-\phi/kT)}$$

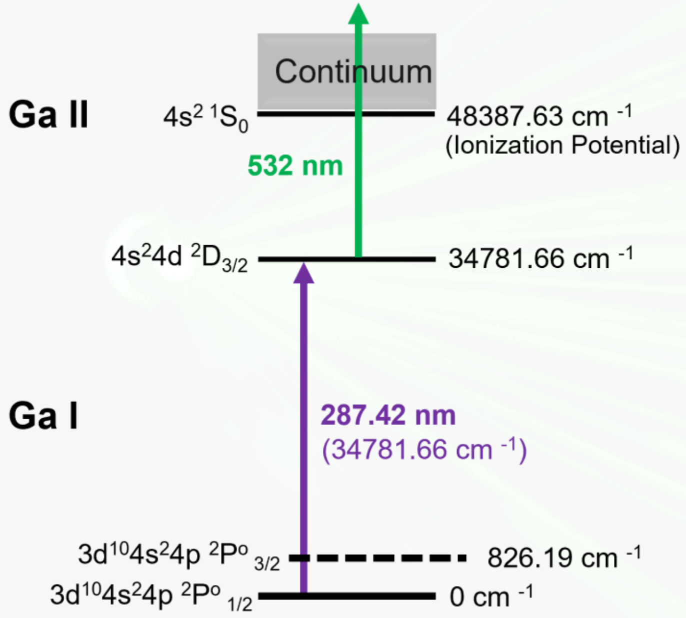
Electron density at the wall

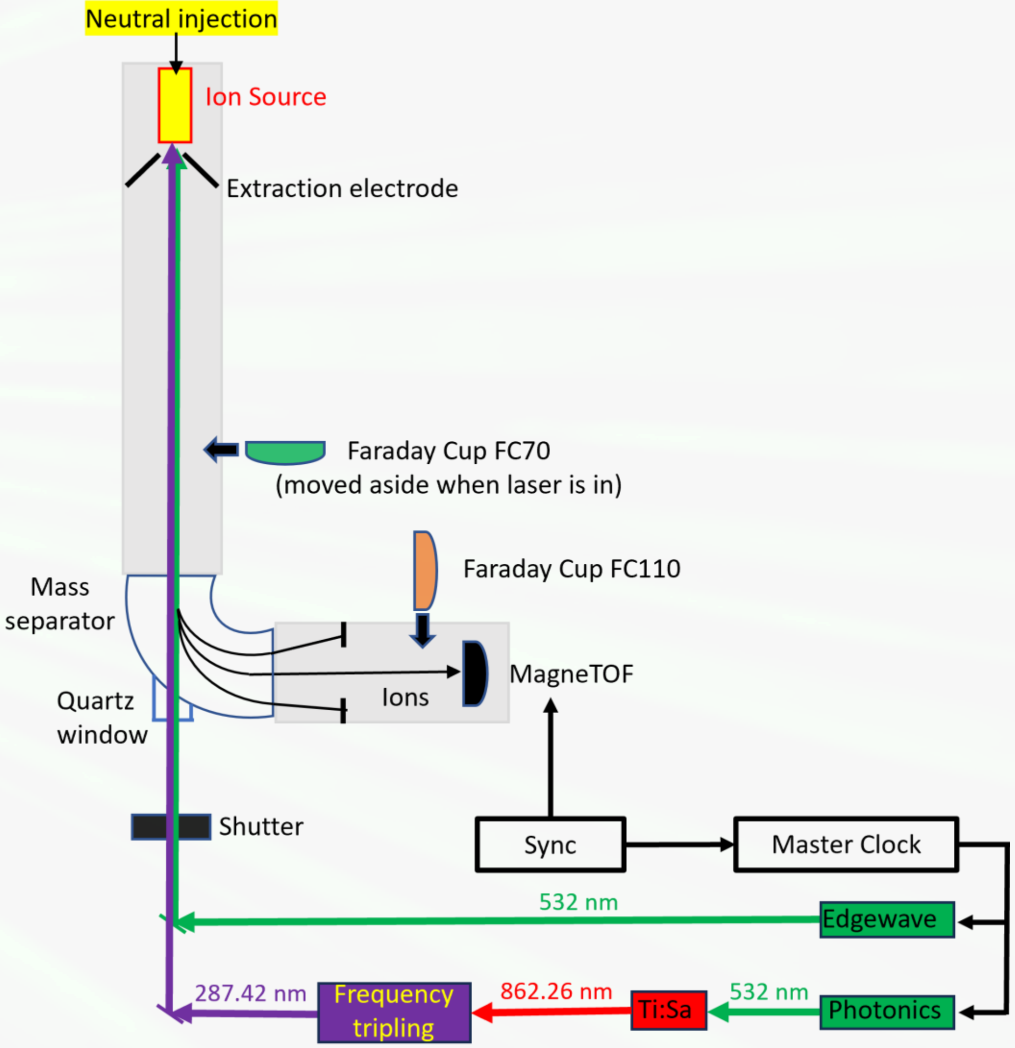
V.I. Mishin et al. https://doi.org/10.1016/0168-583X(93)95839-W

“Far more advantageous is **the increase of neutral density** though quite high densities are required. The **efficiency can be increased** by a factor of approximately 5 if the mean free part of ions is reduced to the order of the cavity dimensions.”

R. Kirchner et al. https://doi.org/10.1016/0168-9002(90)90377-I

Experimental Set-Up at ISOLDE Offline 2, CERN





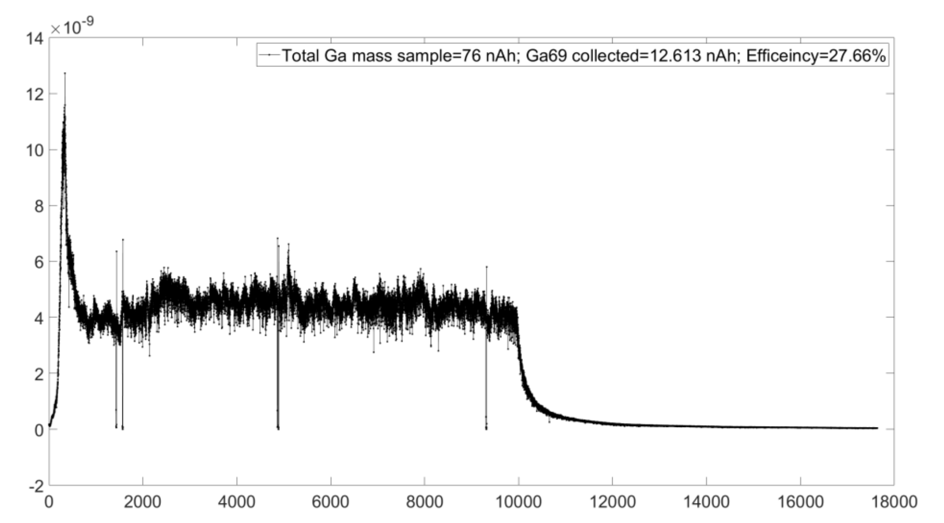
Parameters of the laser system:

- ✓ Repetition rate = 10 kHz
- ✓ UV (287.42 nm) power = 100 mW
- ✓ Green (532 nm) power = 20 W

Ionization Efficiency of Gallium

How is the measurement performed?

- ✓ We take a precise sample of gallium and heat it up to release the neutrals.
- ✓ We collect the gallium ions produced.
- ✓ Efficiency $= \frac{Number\ of\ collected\ ions}{Number\ of\ injected\ neutals}$



Test no.	Type	Efficiency(%)	Mean Efficiency(%)
1	Surface	0.49 ±0.04	0.49 ±0.04
2	Laser	27.66 ±2.07	27.18 ±1.18
3	Laser	27.64 ±2.07	
4	Laser	26.23 ±1.97	

No.	LERs	Mean LER
1	56.1	55.5 (4.5)
2	48.8	
3	54.8	
4	61.5	
5	57.5	
6	55.2	
7	60.5	
8	52.8	
9	57.5	
10	51.2	
11	47.7	
12	60.2	
13	61.4	
14	54.2	

0.49 x 55.5 = 27.12

Laser enhancement of the ion yield

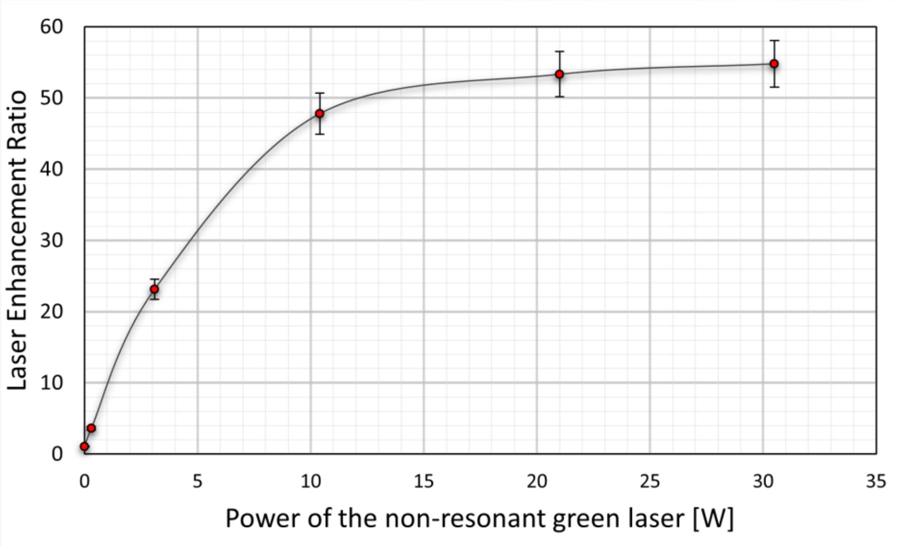
What is Laser Enhancement Ratio?

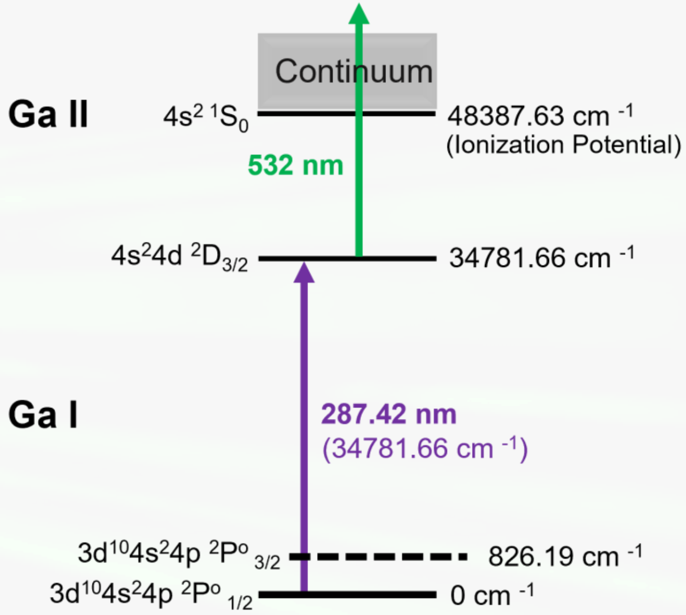
- ✓ In a hot cavity, the production of surface ions is inevitable.
- ✓ We need to understand the selectivity of the laser ion source.

$$\text{Laser Enhancement Ratio, LER} = \frac{\textit{Ion current with Laser ON}}{\textit{Ion current with Laser OFF}}$$

- ✓ Basically, gives us the comparison between the laser ions and the surface ions in the hot cavity.
- ✓ Currents in the range of pA.

LER against the non-resonant laser power (532 nm)

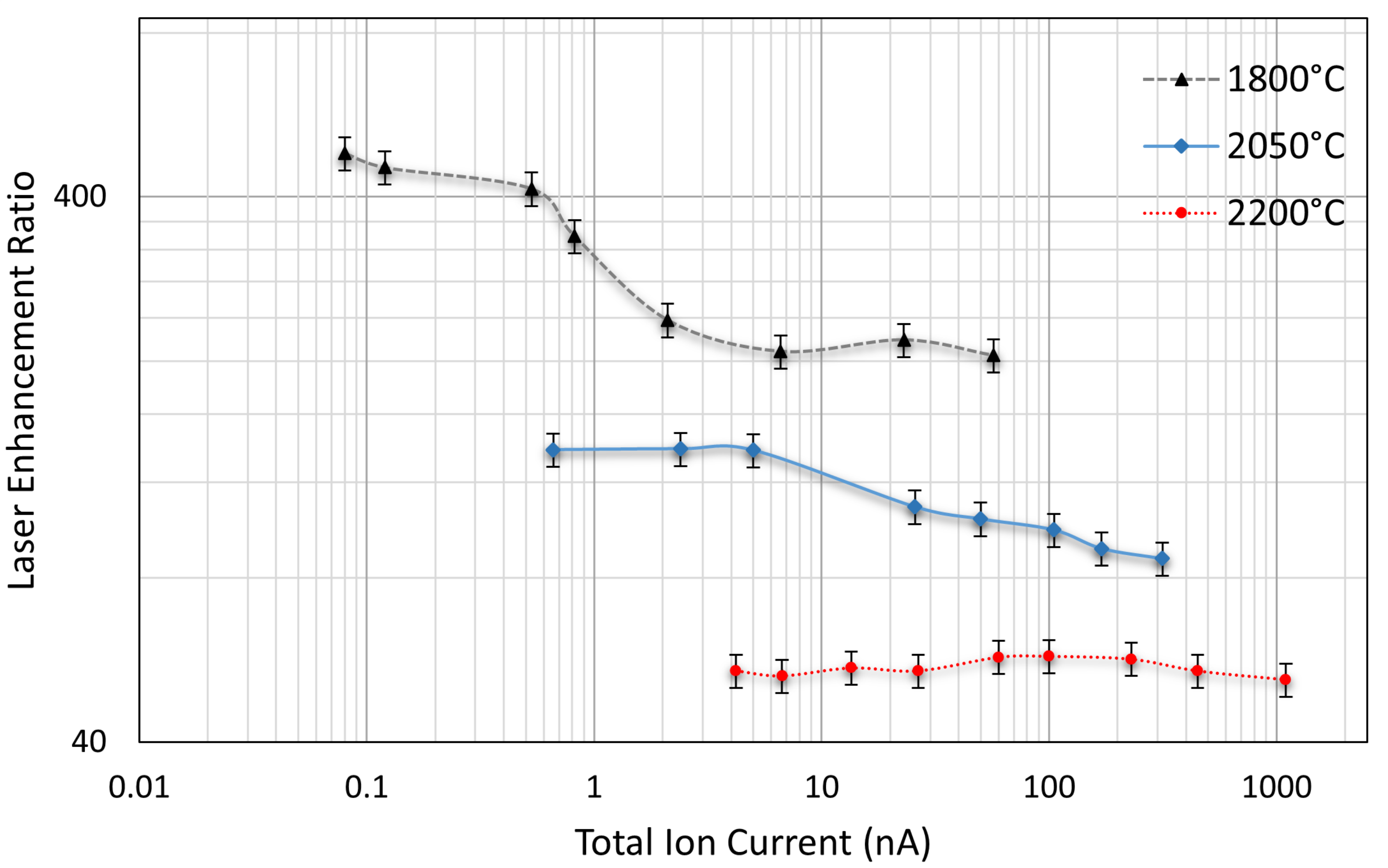




Observance of **false** saturation: Why?

- ✓ Deposition on the laser entrance window from the previous failures of the mass separation magnet.
- ✓ Thermal lensing effect is caused when these deposits absorb high laser power.

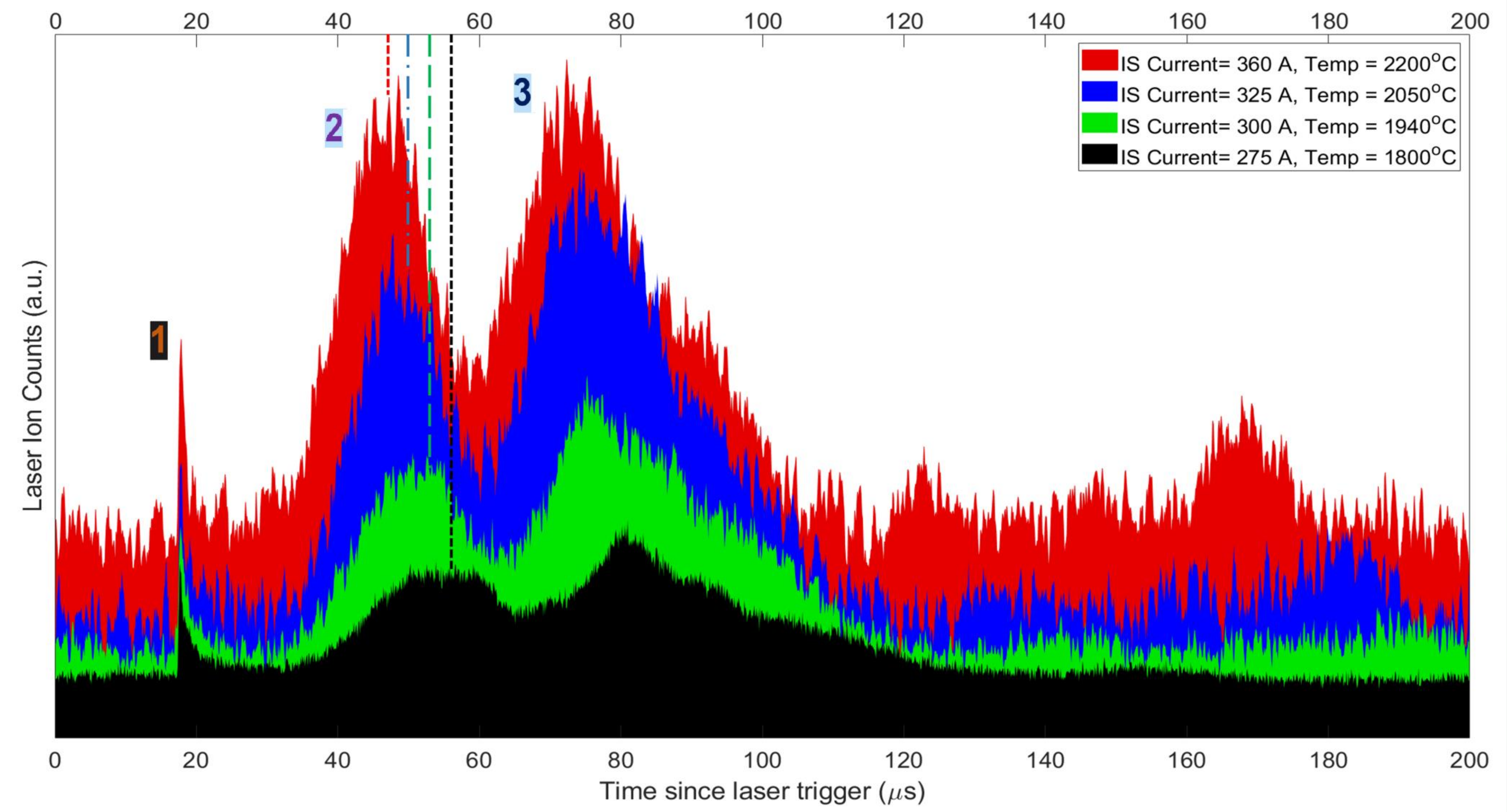
LER against ion source temp. and ion load



Time structure of the ion beam

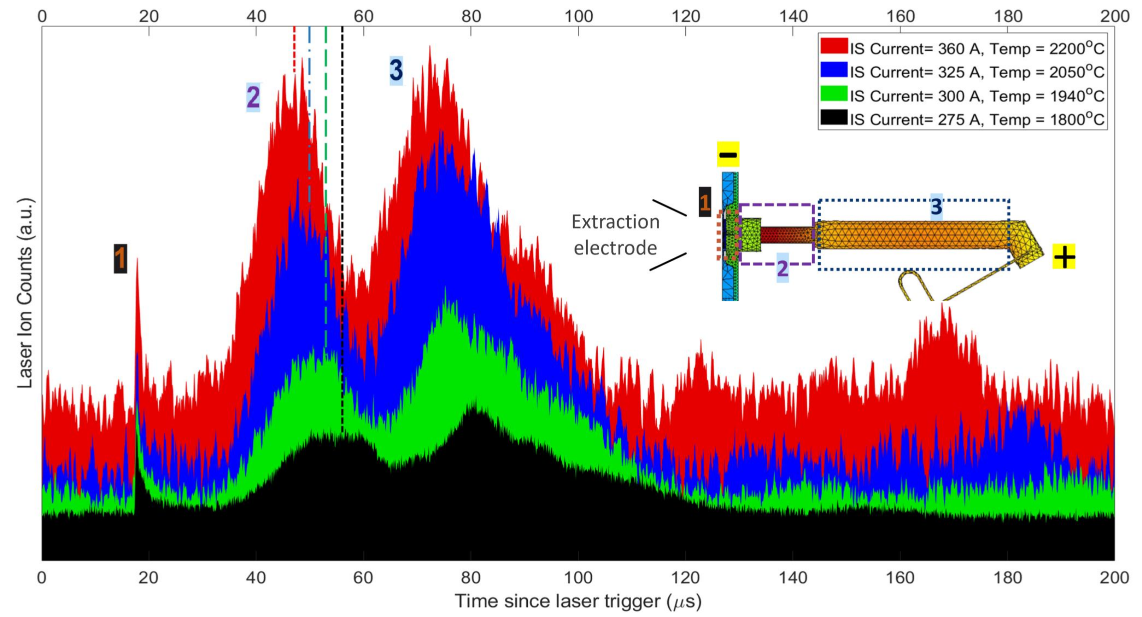
- ✓ Time structure is a **single ion counting** method of measurement.
- ✓ The ion count is performed inside a time window **synchronized with the laser pulses @5 kHz** (200 µs in time)
- ✓ Basically, putting **a time-stamp on each ion** arriving at the magnetof detector with reference to the pulses of the laser and making **a histogram over thousands of laser shots**.
- ✓ Measurement performed with the **range of fA** isotope current on the detector.
- ✓ It provides information regarding the ion source environment.

Measured Time structures at different temperatures of the Ion Source

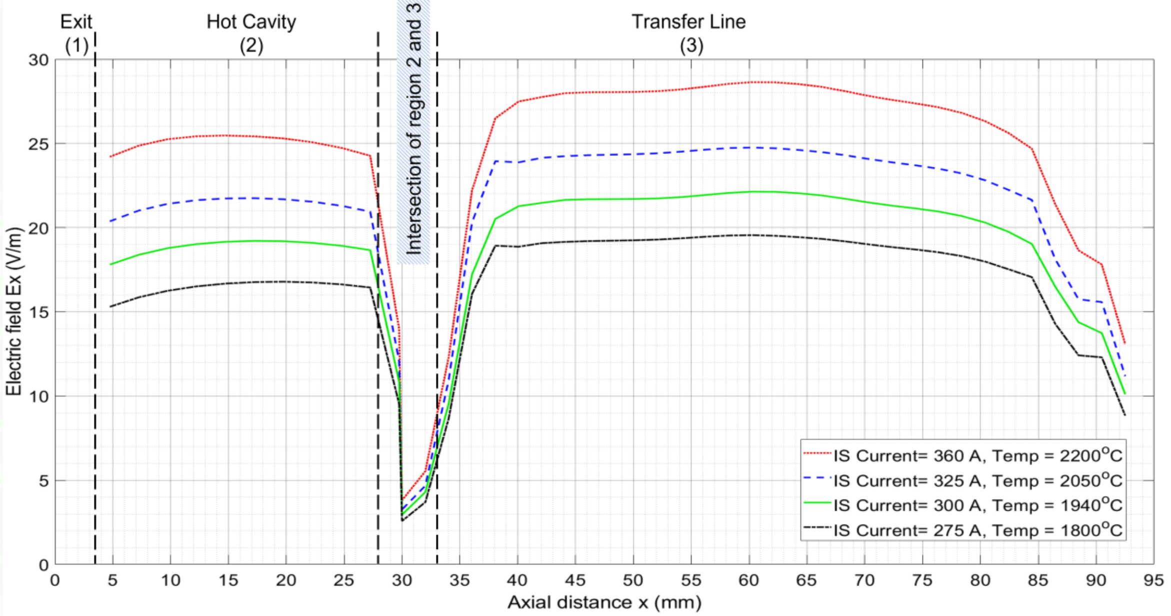


Axial movement of the laser ions

Time structure at different ion source temperatures



Finite Model Electric field simulation



Simple kinematics $a = \frac{qE_x}{m}$ $L_{21} = \frac{1}{2}aT_{21}^2$

Table 1: Computation of the parameters related to the second bunch of ions in the time structure for different temperatures.

Temp. (°C)	$T_1(\mu s)$	$T_2(\mu s)$	$T_{21}(\mu s)$	Av. E_x (V/m)	L_{21_calc} (mm)	L_{10} (mm)	L_{20} (mm)
2200	17.84 ±0.05	46.08 ±0.15	28.24 ±0.20	24.99	13.53 ±0.19	3.10	16.63 ±0.19
2050		47.72 ±0.16	29.88 ±0.21	21.34	12.93 ±0.18		16.03 ±0.18
1940		49.23 ±0.20	31.39 ±0.25	18.82	12.59 ±0.20		15.69 ±0.20
1800		54.34 ±0.53	36.50 ±0.58	16.40	14.83 ±0.47		17.93 ±0.47