ESR3 – WP2 – D2.4

Target developments for extraction of actinides from thick ISOL targets followed by laser-induced molecular break-up and/or ionization.

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ESR3 – WP2 – D2.4

ESR3	CERN	Y	Start: M6	Duration: 36	D2.4
Project title: Target developments for extraction of actinides from thick ISOL targets followed by laser-induced molecular break-up and/or ionization. (**WP2**: Novel techniques and technologies for actinide research)					
Objectives: Study and optimize the reaction conditions required to create volatile molecular species of refractory elements in general and actinides in particular. Develop a dissociation scheme for the provision of atomic species suitable for efficient laser ionization or in-source laser spectroscopy.					
Expected results: Extraction of radiogenic actinide elements from an ISOL target and delivered in atomic form to the users. Determine the production yield and purity of the beams and report the new beam availability to the community.					
Planned secondment(s): JGU (Christoph E. Düllmann) – M8-10 – working on the sample preparation to be used for the actinide molecular release studies; TRIUMF (Thomas Day Goodacre, Peter Kunz) – M15-17 – Extraction of actinides from thick targets at the ISAC facility. Participation in activities around radioactive ion beam development involving actinide beams and molecular beam extraction. Gaining experience in radioactive detection techniques used to assess the quantity and quality of the produced isotopes.					
Enrolment in Doctoral degree: JGU in the Department of Chemistry under the supervision of Prof Christoph E. Düllmann					

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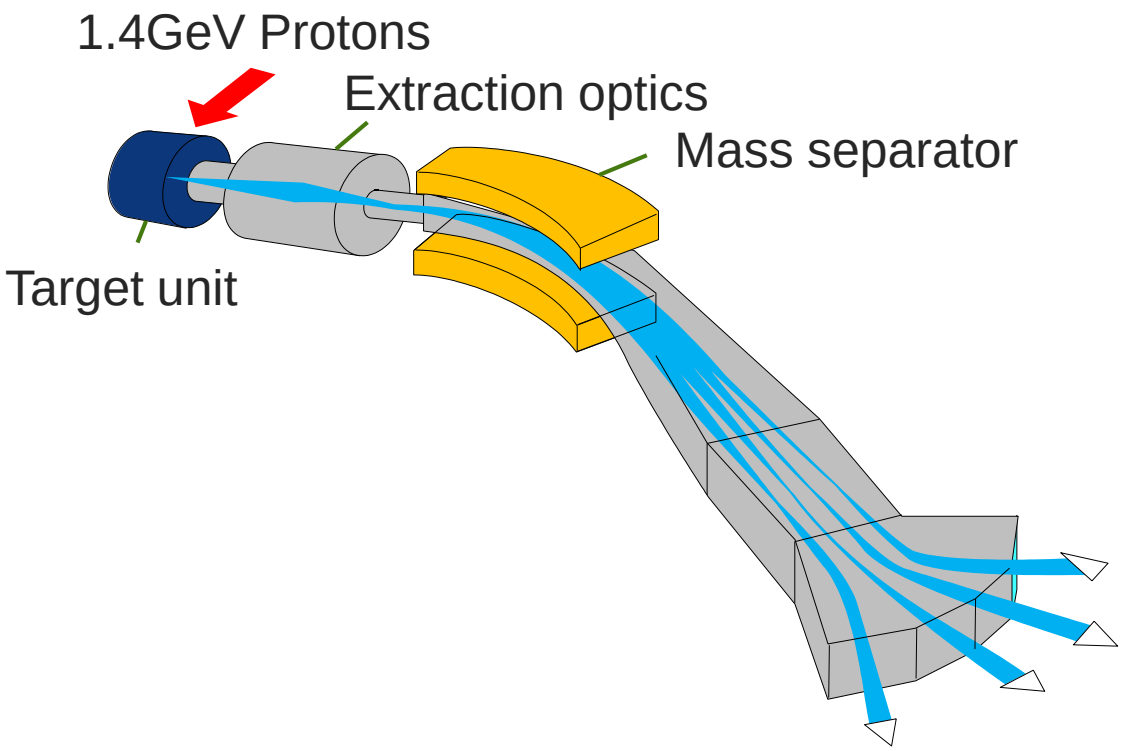
Outline

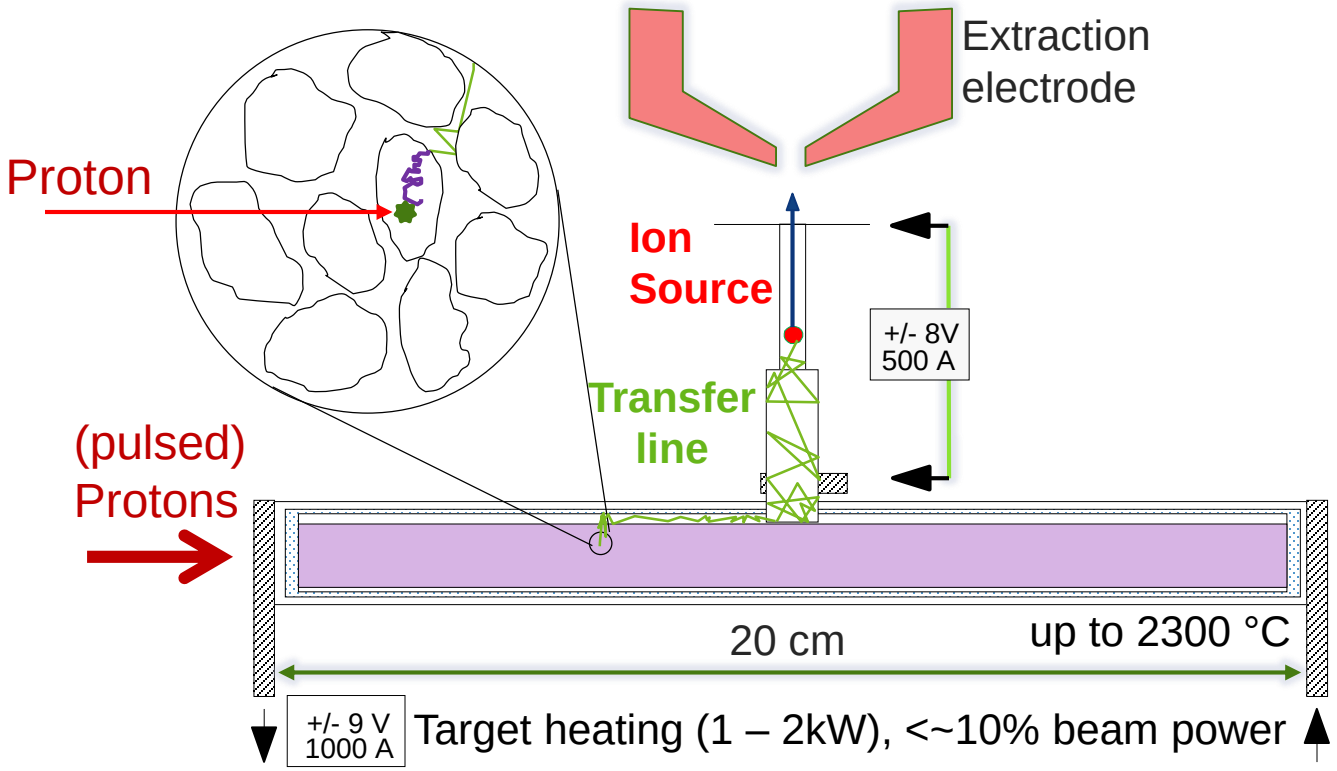
- Motivation for molecular beams

- Optimization of reaction conditions
- Develop a dissociation / ionization scheme
- Determine production yield

ISOLDE: Isotope Separation On Line

Adapted from
J. P. Ramos | 17/09/2018
EMIS2018





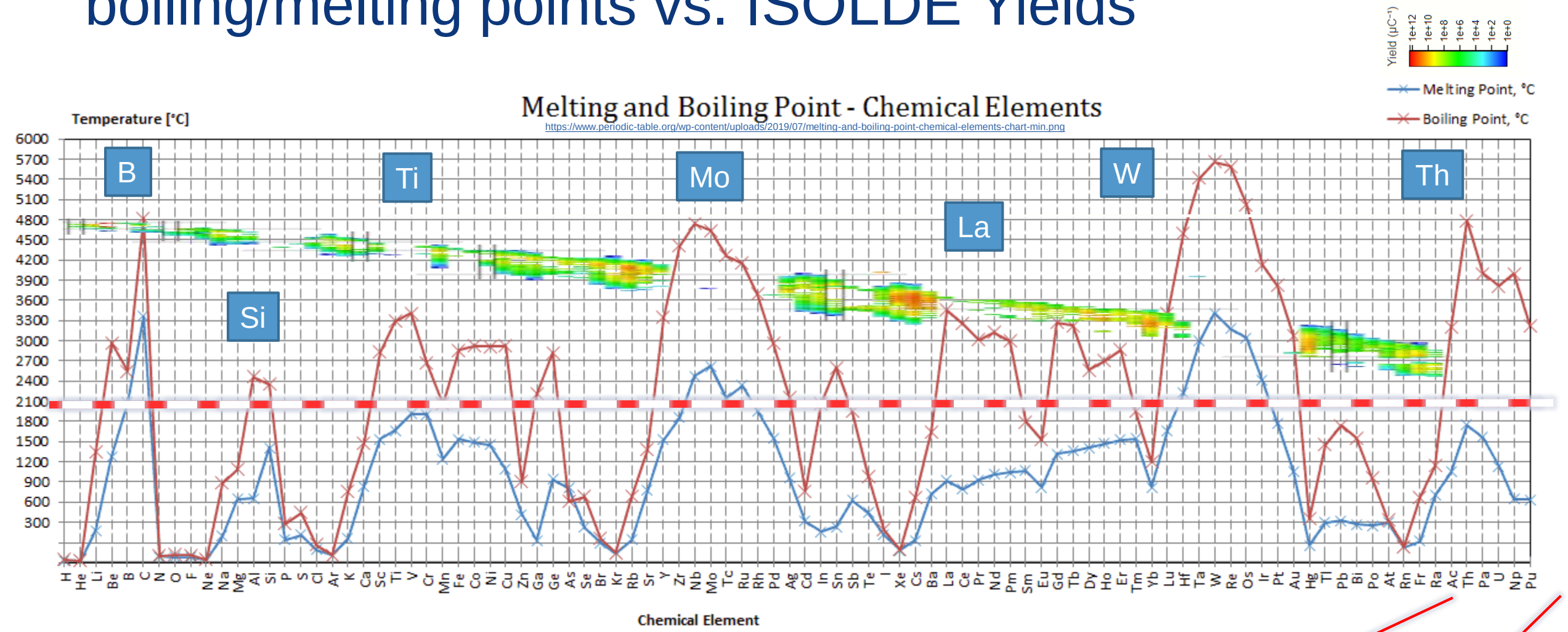
1. Production
2. Diffusion
3. Effusion
4. Ionization
5. Mass Separation
6. Transport

$$Beam\ Intensity = \sigma . j . N_t . \varepsilon$$

$$\varepsilon = \varepsilon_{diff} \varepsilon_{eff} \varepsilon_{is} \varepsilon_{sep} \varepsilon_{trans}$$

N_t – Nr of exposed atoms [dim]
j – Proton flux [cm^{-2}]
σ – Cross section [mb]
ε – Efficiency [%]

boiling/melting points vs. ISOLDE Yields

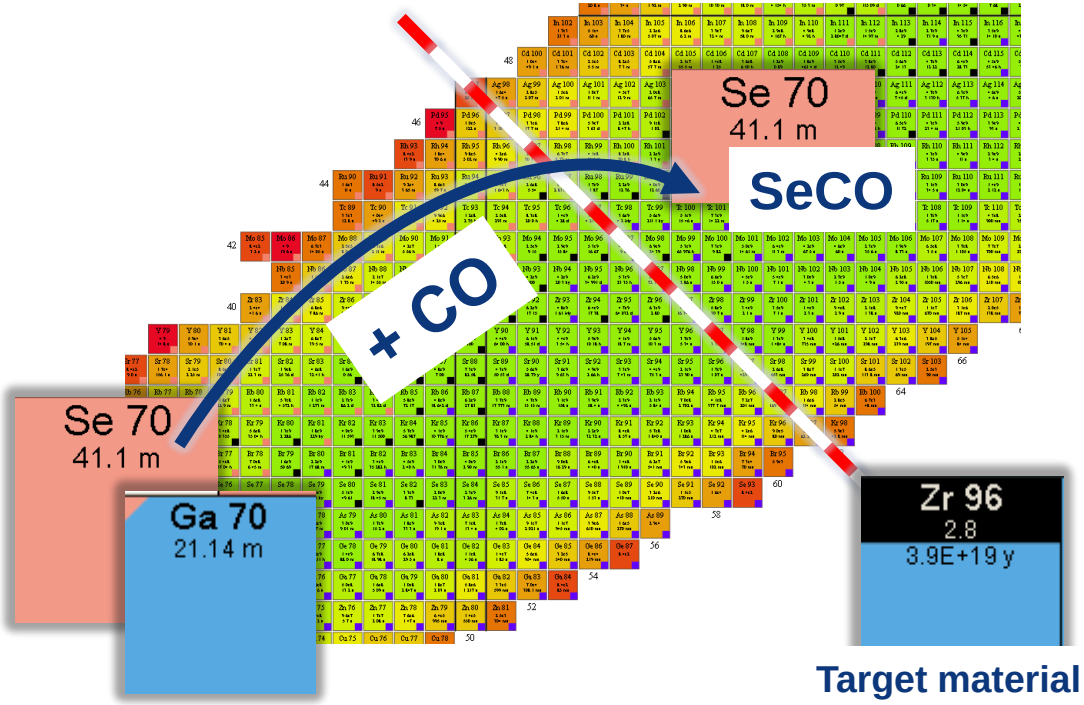


Target materials operated at < 2200 C

actinides

Molecular Beams – Why?

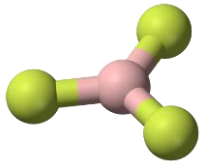
- **Beam purification**
  - Shift the mass region to a higher mass to avoid isobaric contaminants. e.g. GeS, SnS, SeCO, LnO



Target material

- **Beam extraction by *In-situ* volatilization**
  - Elements with very low volatility are not released
  - Reactive elements can be chemically trapped

$$^{8}B \xrightarrow{SF_6\ (g)} {}^{8}BF_3$$

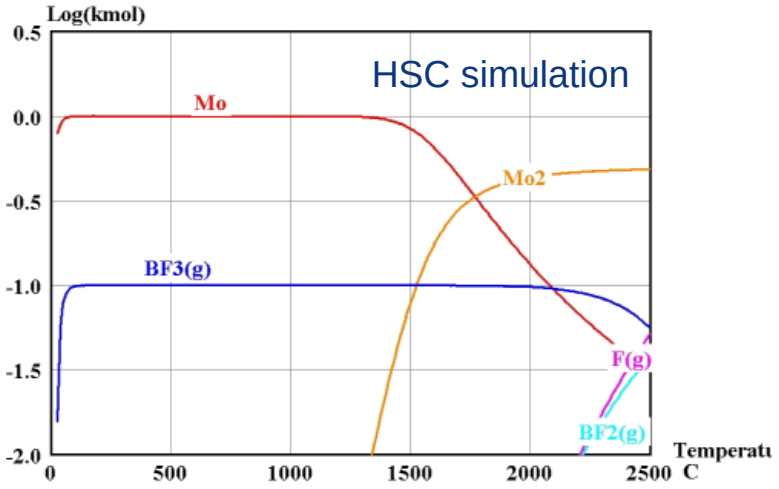


Boron

Low Volatility (m.p. 2076 °C)
reactive with many metals

Boron trifluoride

gaseous even at RT
very stable



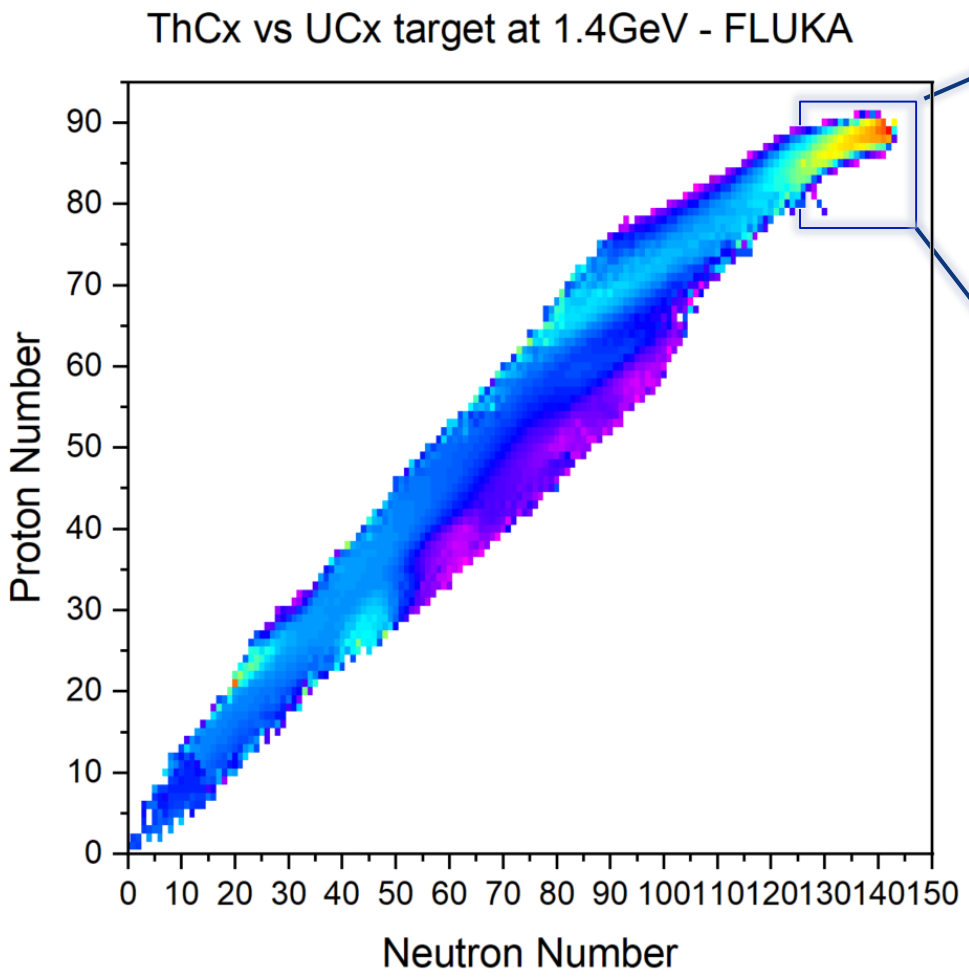
Jochen Ballof | ISOLDE Workshop | 5.DEC.2017

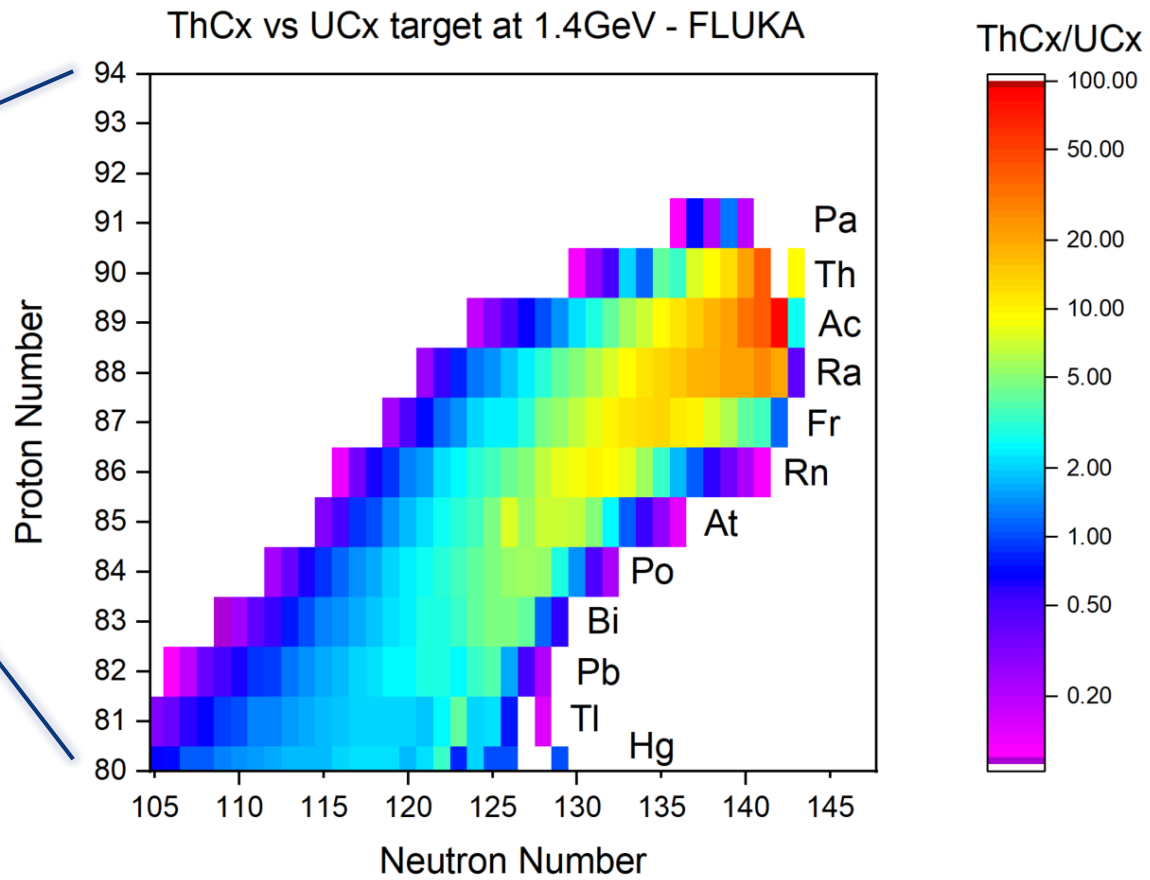
Optimize reaction conditions

- target material
  - U vs. Th
  - Metal vs. Carbide vs. Oxide
- target microstructure
  - Investigate nano materials , stabilize nanostructure
- Reactive gas
  - (O, F, S, …)
- Reaction conditions
  - Concentrations, temperatures

In-target production

Thorium vs Uranium





! Release properties to be studied

! Simulated results require validation

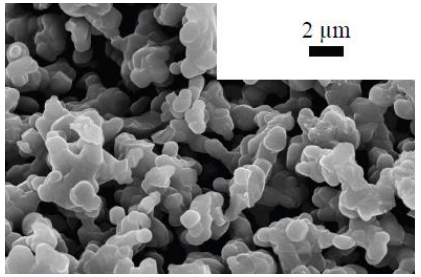
- ThC is a standard target material at ISOLDE
- Nano ThC possible with new nano lab
  - co-development with SCK*CEN possible

FLUKA Simulations: : Joao Pedro Ramos

Nanomaterials

SiC - S. Fernandes, et al.

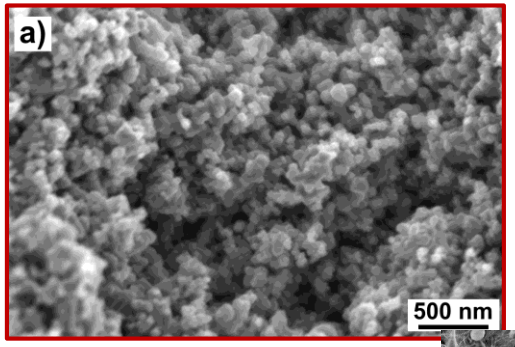
2010



*submicron

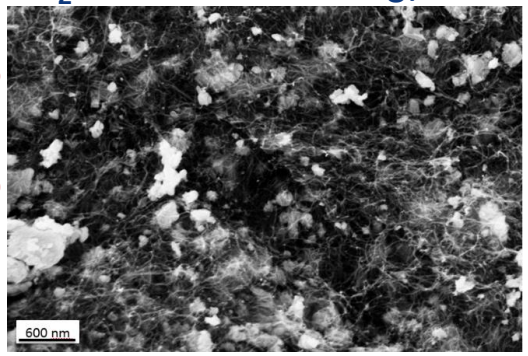
TiC+CB – J.P. Ramos, et al.

Nov- 2014



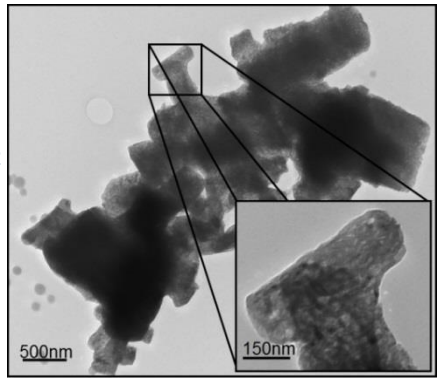
UC_2 + 2C – A. Gottberg, et al.

Nov - 2014



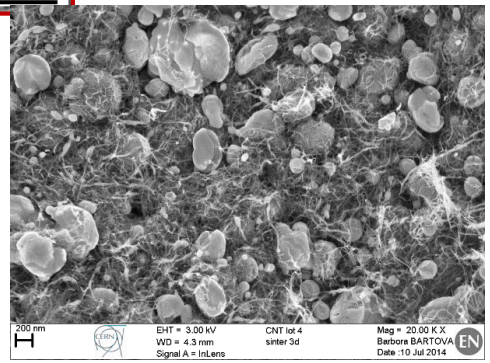
*1st nanomaterial at ISOLDE

2011



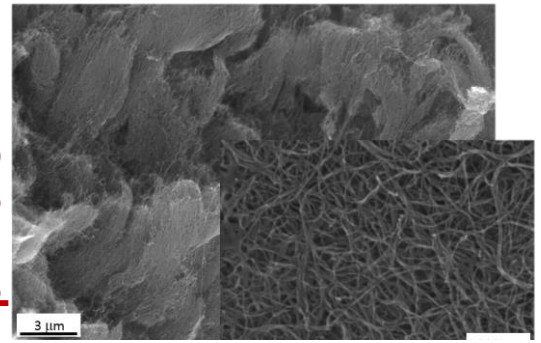
CaO – J.P. Ramos, et al.

Nov- 2014



LaC_2 + 2C – J. Guillot, et al.

Apr- 2015



MWCNT – C. Seiffert, et al.

Study effects of reactive gas to actinide nano materials

Adopted from

João Pedro Ramos | 07/09/2017

MEDICIS-Promed Specialized Training on Radioisotope Production

Molecular Beams – The classical approach

Formation of molecules under low pressure conditions and high temperatures

Selected molecular sidebands

Halides | Al-X | Sulfides | X-CO | Oxides

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
1 H																	2 He
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
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
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
55 Cs	56 Ba	71 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
87 Fr	88 Ra	71 Ac...	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Cn	113 Nh	114 Fl	115 Mc	116 Lv	117 Ts	118 Og

What are the best sidebands for actinide release ?
1st experiments planned with irradiated targets at MEDICIS

U. Köster et al.
NIMB 266 (2008), 4229
EPJ-ST (2007), 285

Slide adapted from: Jochen Ballof | ISOLDE Workshop | 5.DEC.2017

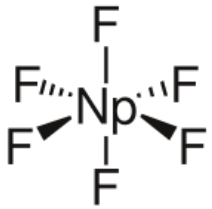
Beyond Uranium

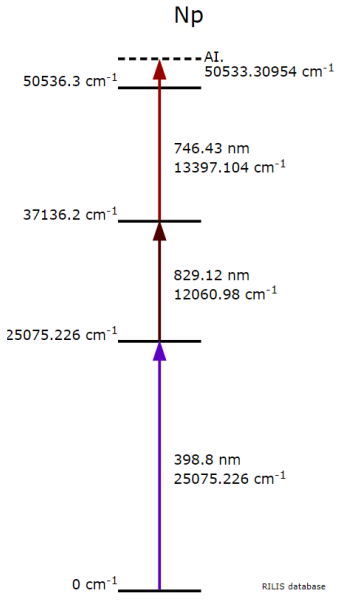
Pu 232 2.29e+5 33.7 m 0.5	Pu 233 7.70e+5 20.9 m 0.4	Pu 234 1.39e+6 8.8 h 0.1	Pu 235 4.31e+6 25.3 m 0.5	Pu 236 8.97e+6 2.858 y 0.008	Pu 237 2.55e+7 45.64 d 0.04	Pu 238 5.14e+7 87.7 y 0.1	Pu 239 2.92e+7 24.11 ky 0.03	Pu 240 1.23e+7 6.561 ky 0.007	Pu 241 5.41e+5 14.329 y 0.029	
Np 231 8.61e+6 48.8 m 0.2	Np 232 3.24e+7 14.7 m 0.3	Np 233 5.65e+7 36.2 m 0.1	Np 234 1.03e+8 4.4 d 0.1	Np 235 6.70e+8 396.1 d 1.2	Np 236 6.91e+8 153 ky 5	Np 237 1.08e+9 2.144 My 0.007	Np 238 4.08e+8 2.099 d 0.002	Np 239 9.86e+7 2.356 d 0.003	Np 240 1.41e+7 61.9 m 0.2	Np 241 3.74e+5 13.9 m 0.2
U 230	U 231	U 232	U 233	U 234	U 235	U 236	U 237	U 238	U 239	U 240

UCx , 1.4GeV, Fluka

Neptunium

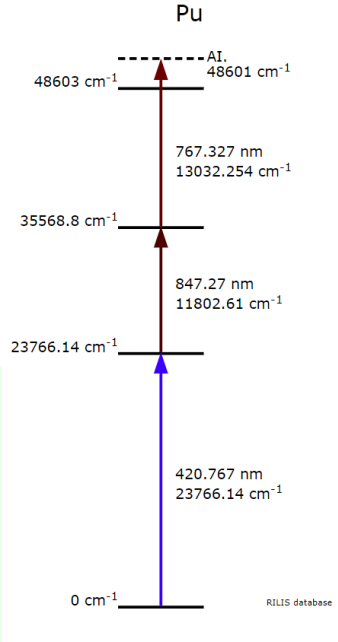
NpF6
BP: 55C





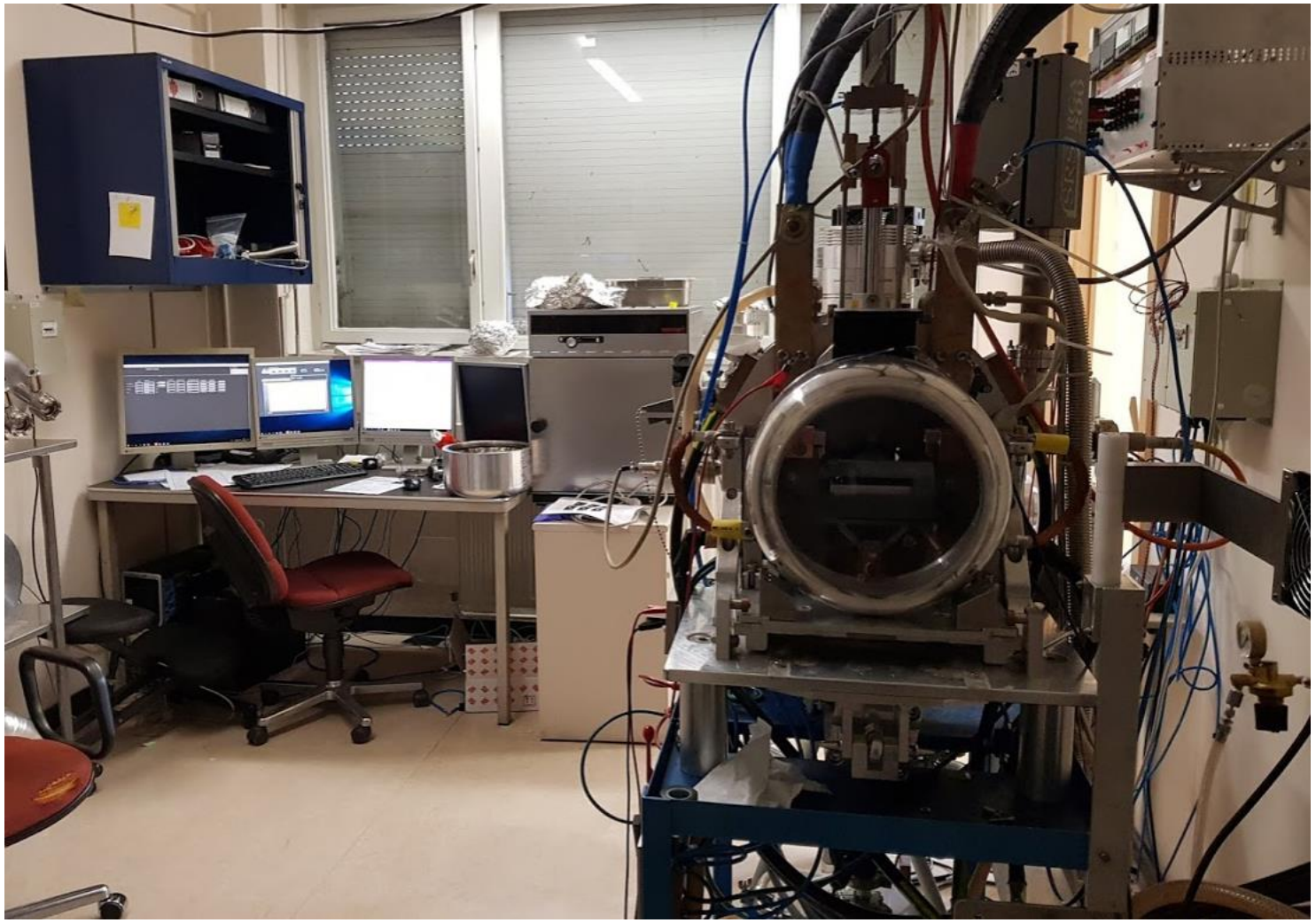
Plutonium

TRIUMF 239Pu: 1E7 /s

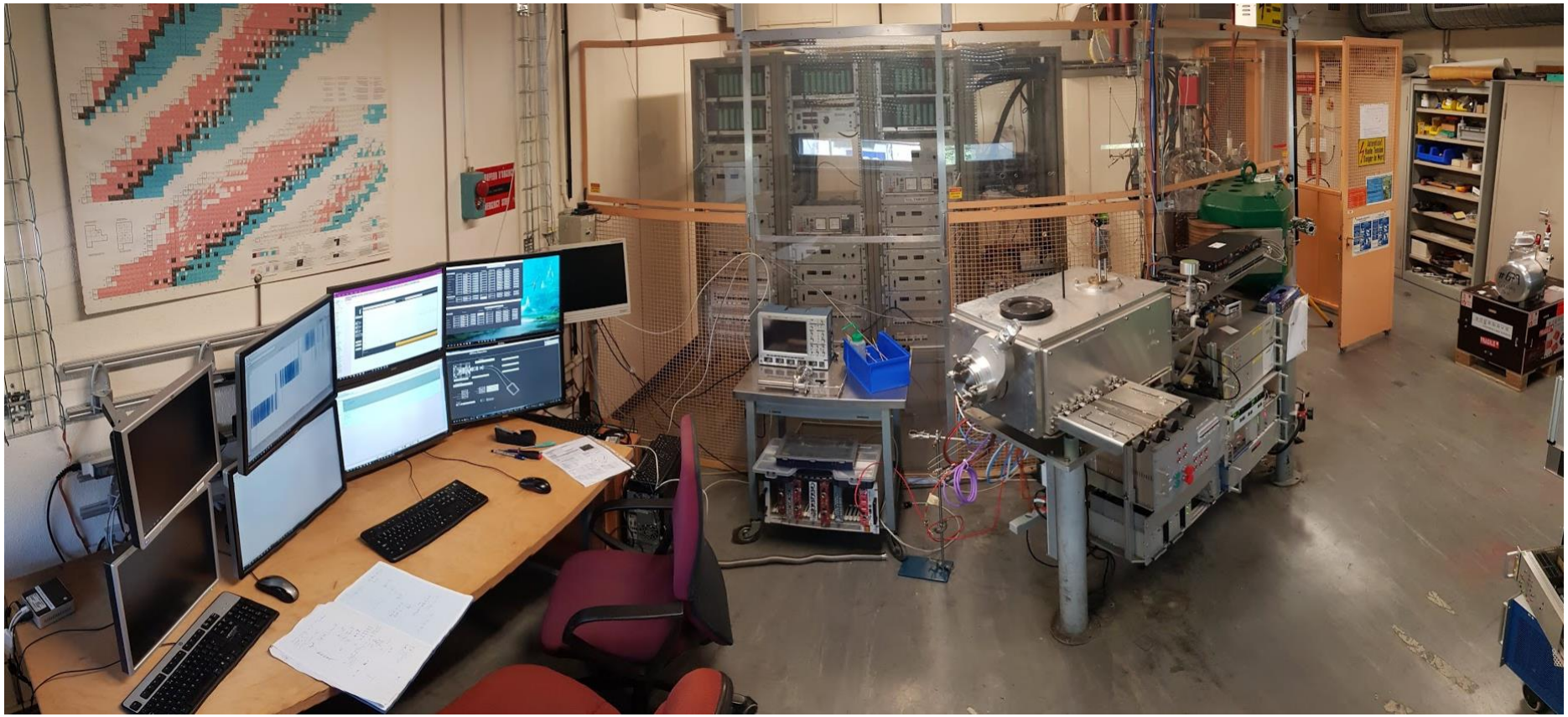


Study NpF6 release

Pump stand (YPS1)



Offline 1 (YOL1)



Offline 2 (YOL2)

Frontend, Separator magnet

RFQ-CB

https://home.cern/news/news/experiments/isoldes-new-offline-2-source-nears-completion

Studying molecular beam formation

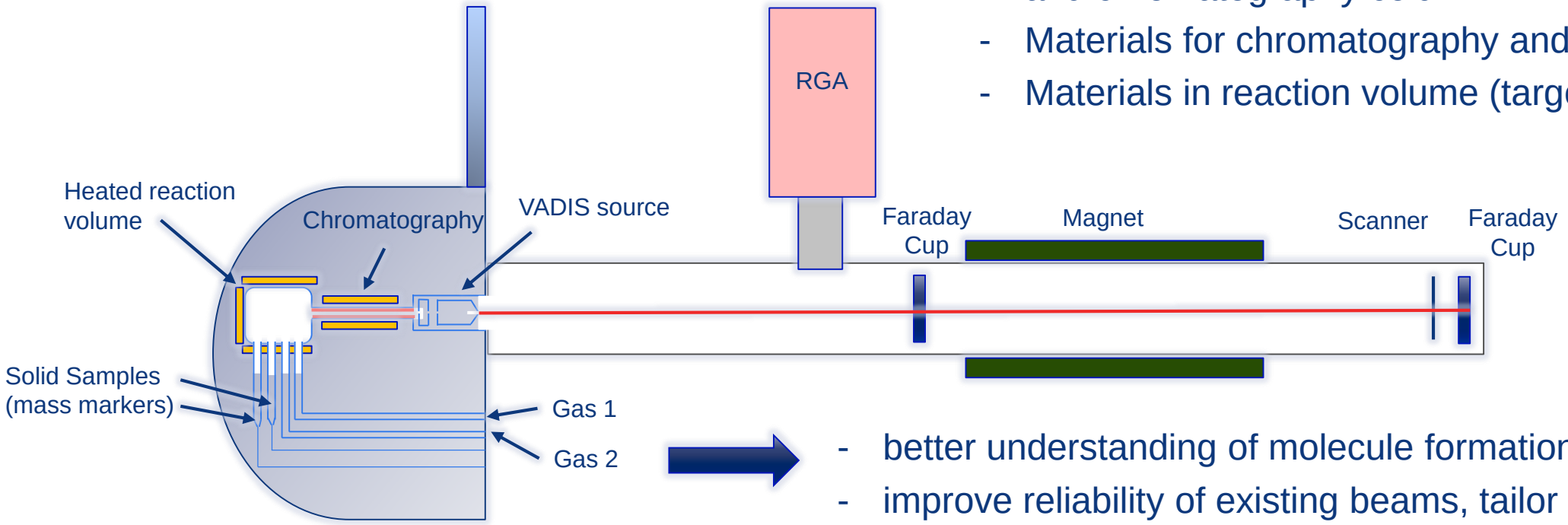
Concept for a dedicated development unit for molecular beams

Study chemical reactions

- Injection of gases and vapor of solid samples into reaction volume
- Suppression by quartz and other materials

Parameters

- 2 gases, controllable flow rates
- 2 mass markers
- Controllable temperatures in reaction volume and chromatography column
- Materials for chromatography and
- Materials in reaction volume (target matrix)

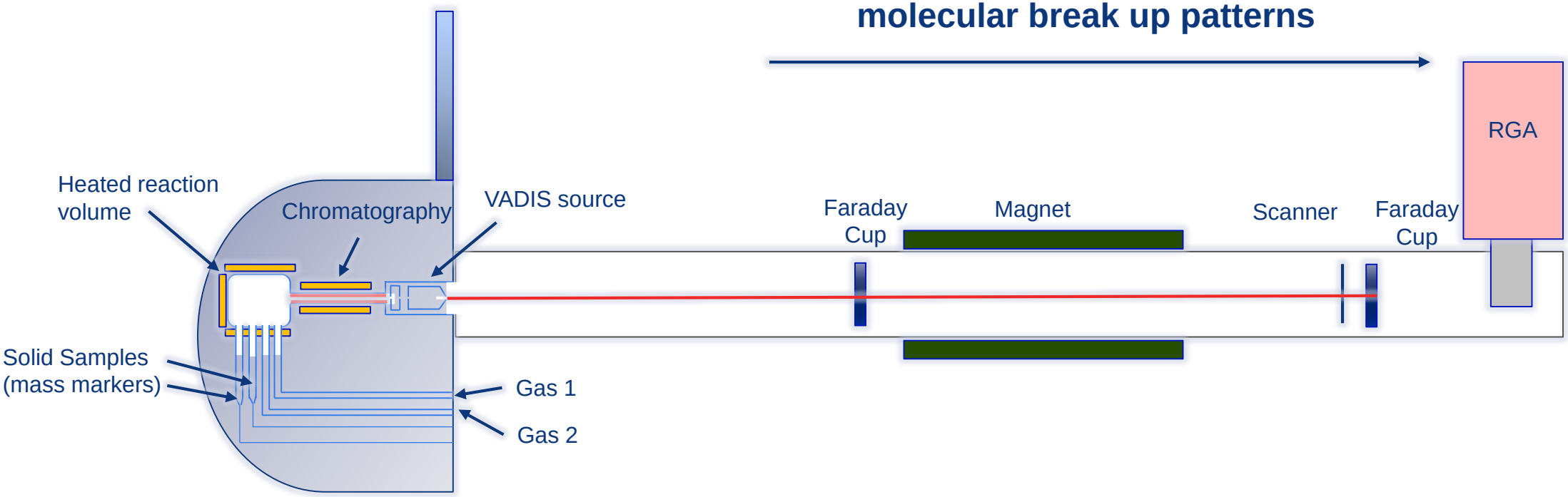


- better understanding of molecule formation
- improve reliability of existing beams, tailor new beams

Studying molecular beam formation

Concept for a dedicated development unit for molecular beams

- Move residual gas analyzer to identify separated beam composition through **molecular break up patterns**

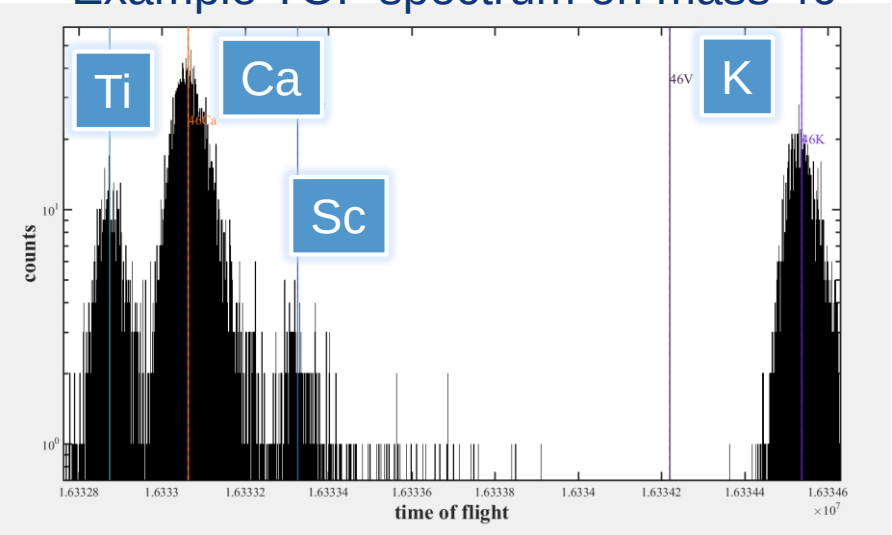


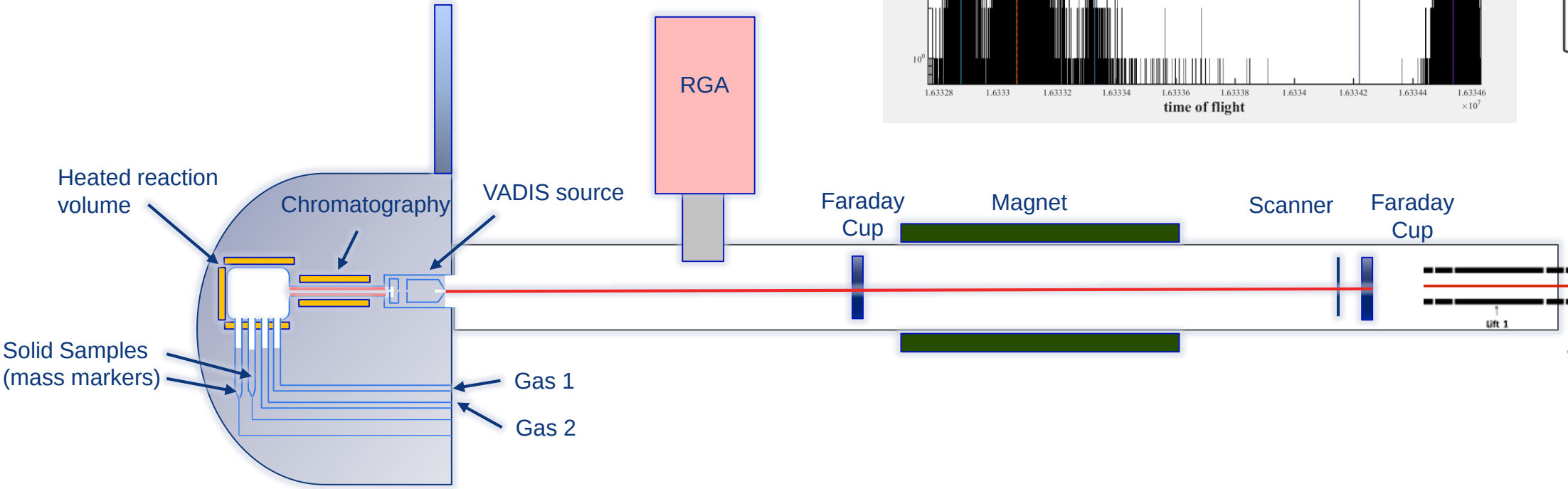
Studying molecular beam formation

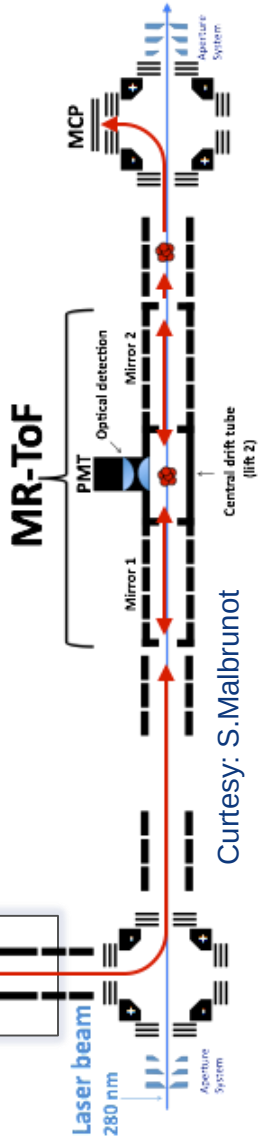
Concept for a dedicated development unit for molecular beams

- Add **Multi Reflection Time of Flight (MR-ToF)** mass spectrometer: allows ISOBAR separation.
- Collaboration with MIRACLS experiment launched

Example TOF spectrum on mass 46

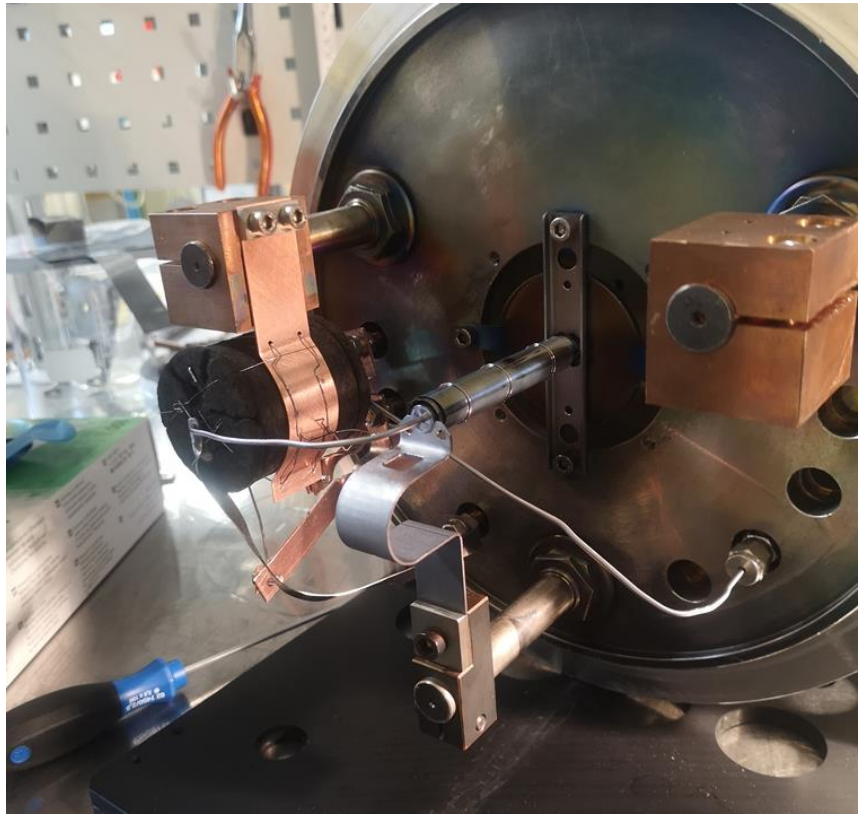


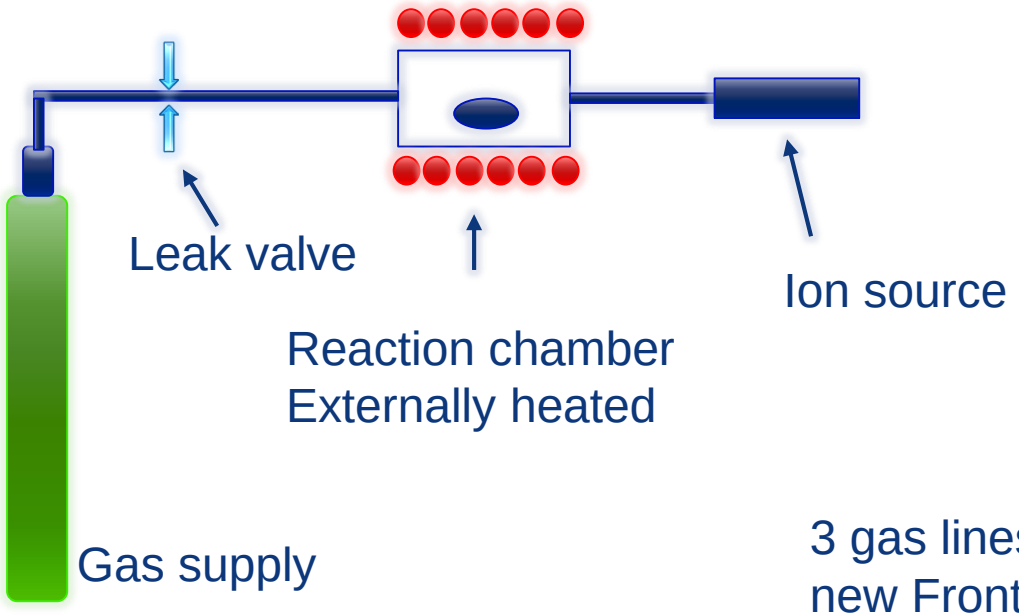


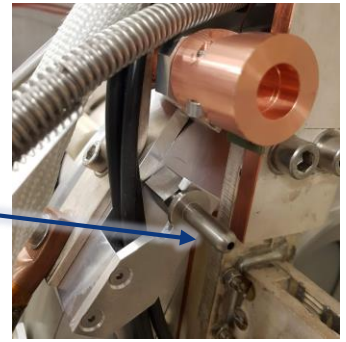


Curtesy: S.Malbrunot

First prototype for reaction chamber tested at YOL1







3 gas lines to
new Frontends

1 reacting gas
1 sample supply
Surface ionization

Multiple gasses
Plasma sources, Negative ions

BaO + CF4 -> BaF2 + x -> BaF+

V.Samothrakis, M.Ballan, J.Ballof, D.Leimbach, B.Crepieux, S.Rothe et al.

Photocathode source

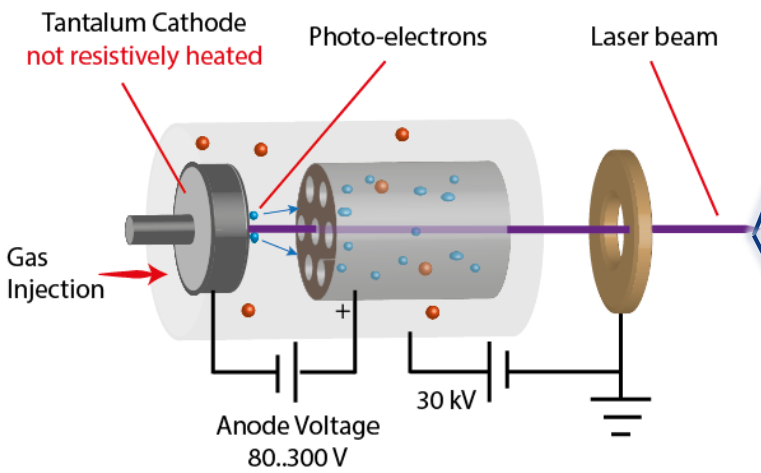
VADIS source at ambient temperature

Electron generation by laser, not thermal evaporation

Motivation

- Ionization of fragile molecules
- No decomposition on hot surfaces
- Diagnostic tool to measure ionization properties

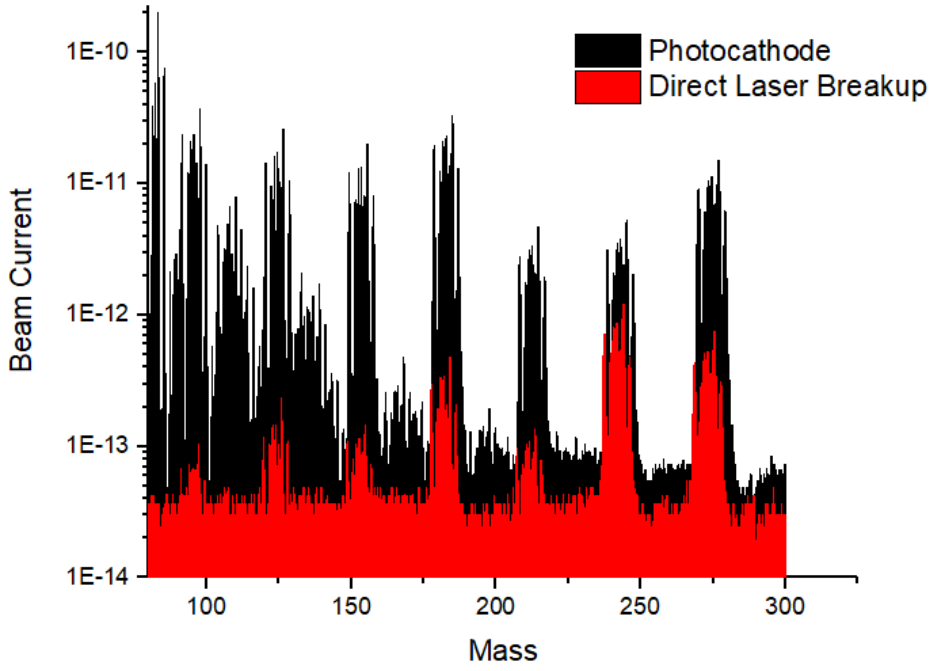
Set up



Laser properties (PHARROS)	
Pulse length	265 fs
Power	4.5 W
Wavelength	343 nm
Rep. Rate	50 kHz

J. Ballof, D. Leimbach, B.A. Marsh, A. Ringvall-Moberg, S. Rothe, T. Stora, S. Wilkins

First Results:
Mass spectrum of $Mo(CO)_6$ + Kr



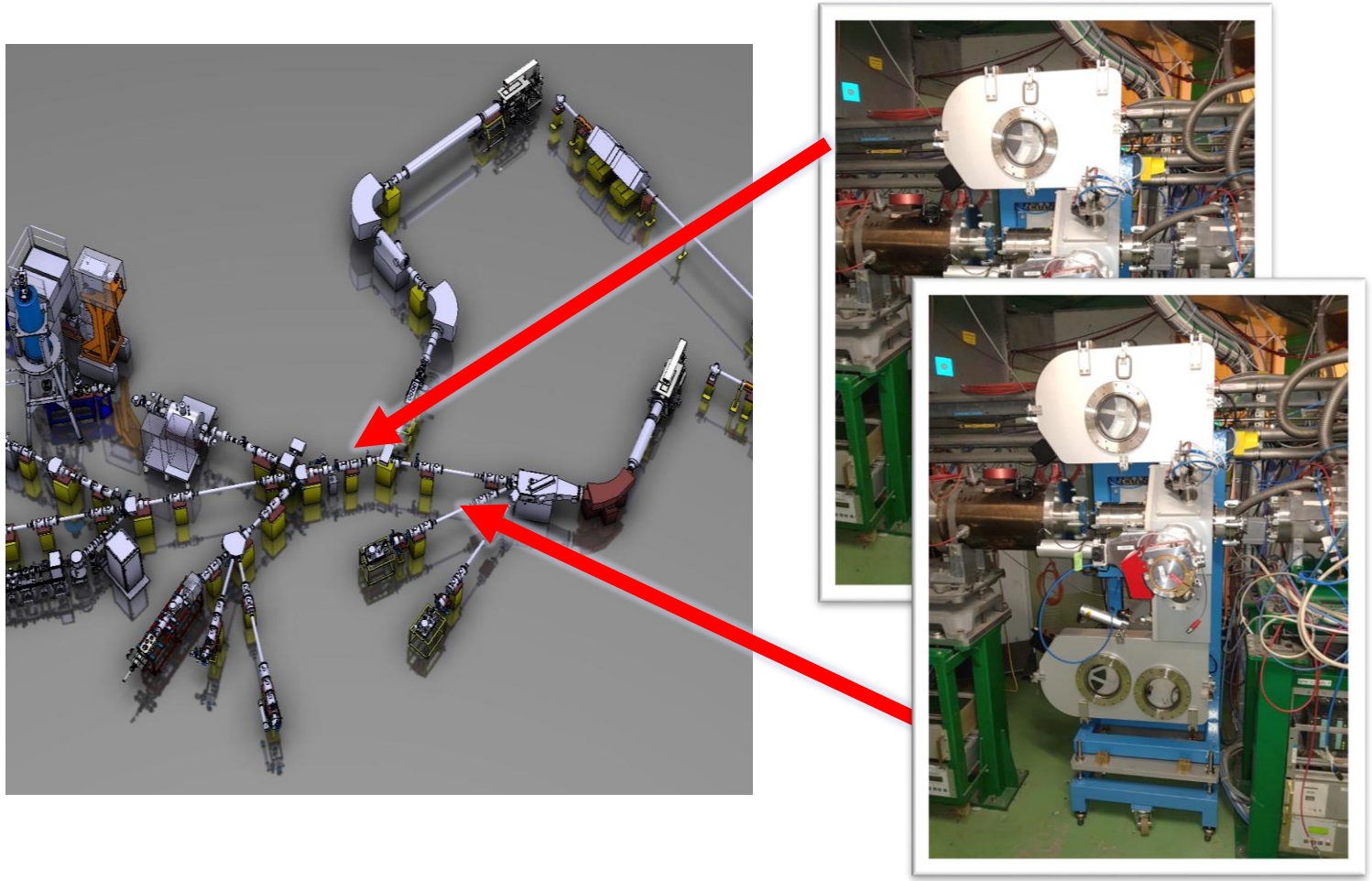
Two operation modes found

Photo cathode	Direct laser breakup
Anode biased	Anode off
Magnet 6A	Magnet off
Krypton ionized	Krypton not ionized
Mo(CO)3 predominant	Mo(CO)5 predominant

THE ISOLDE Yield station (YYS)

Proposal: install spare YYS2 at GLM

- Used for beam development
- Can be used in p.sharing mode with minimal impact to ISOLDE physics
- Opportunistic development, in-source laser spectroscopy
- Upgrade (LS3…): integrate MR-ToF-MS



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