Infrastructure for radioactive molecule production at CERN-ISOLDE

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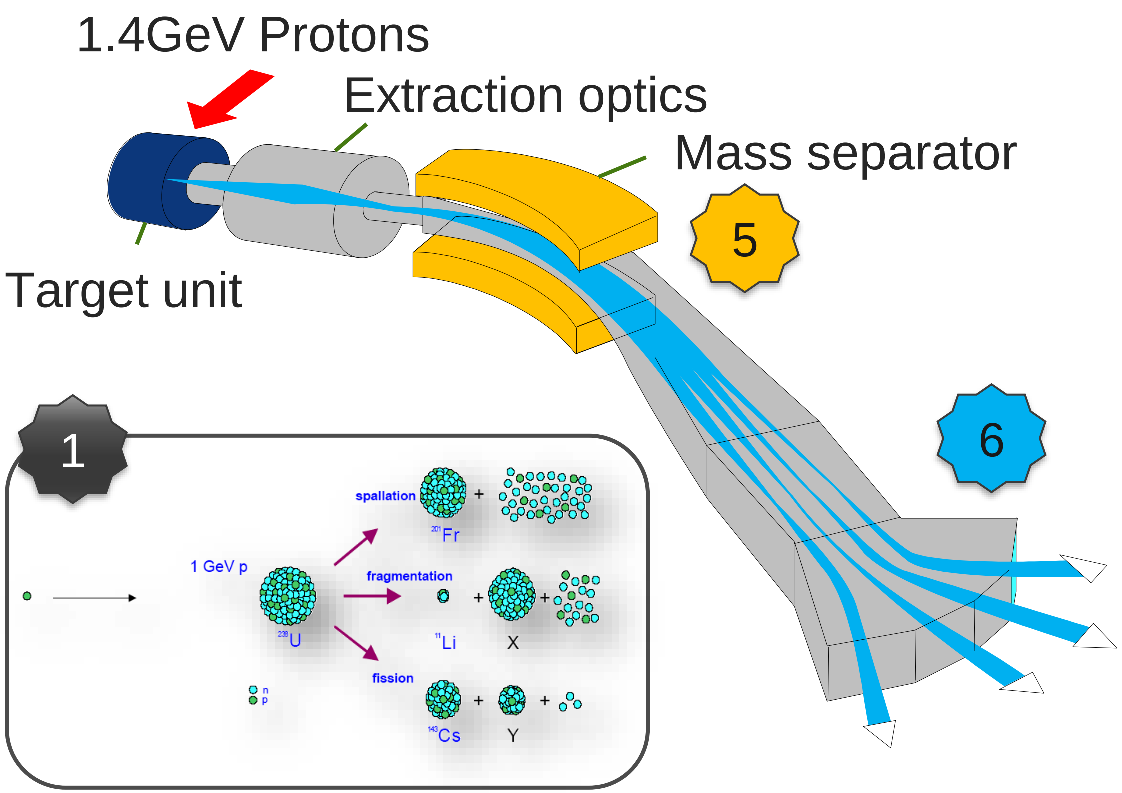
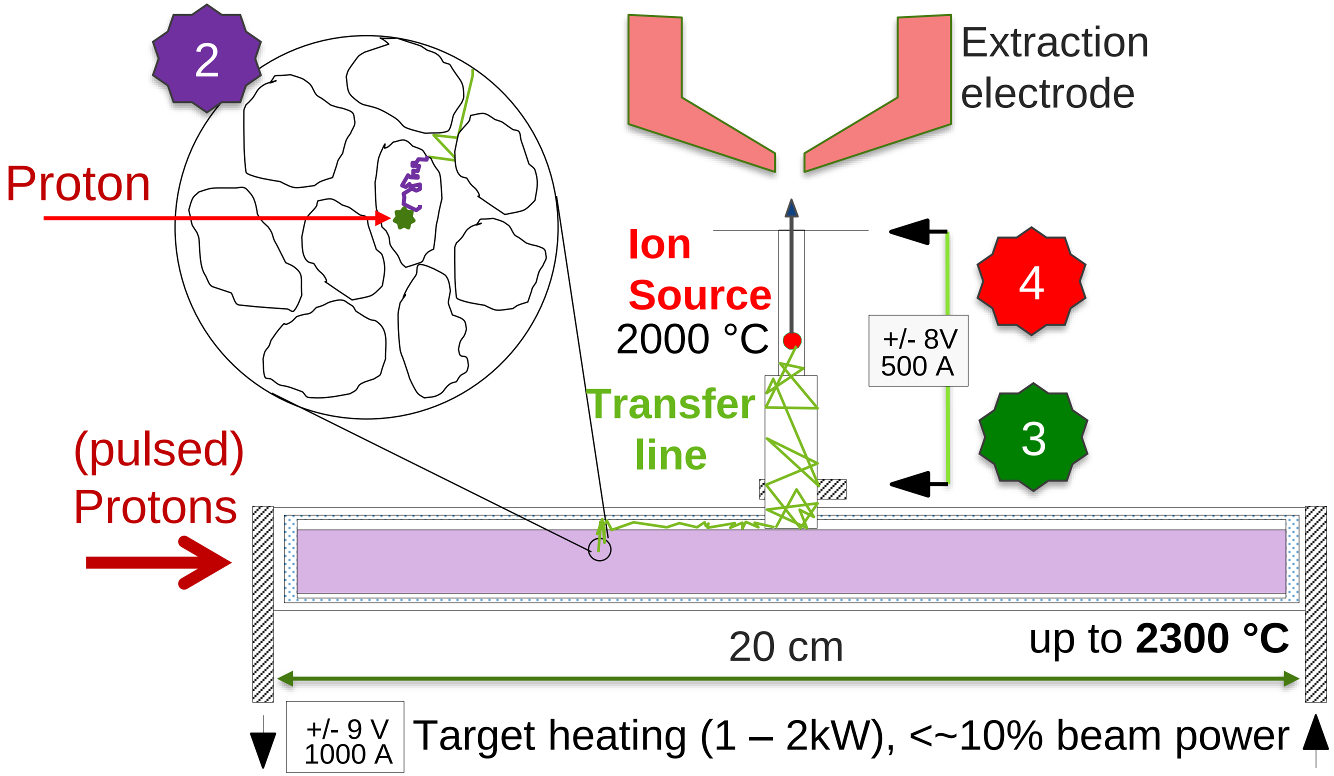
ISOLDE: Isotope Separation On Line DEvice

R. Catherall *et al* 2017 *J. Phys. G: Nucl. Part. Phys.* **44** 094002

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



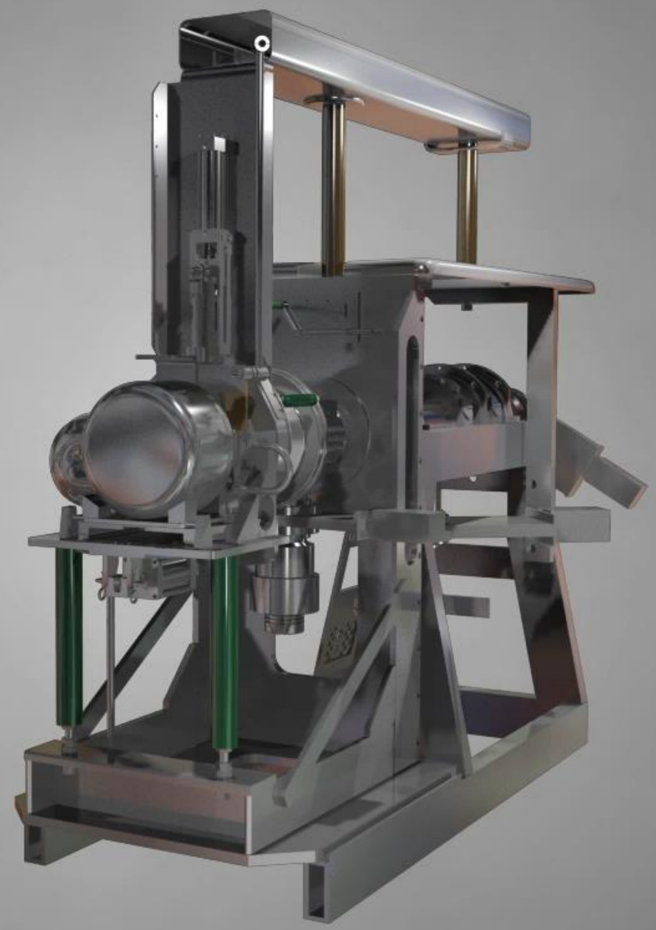


1. Production
2. Diffusion
3. Effusion
4. Ionization
5. Mass Separation
6. Transport

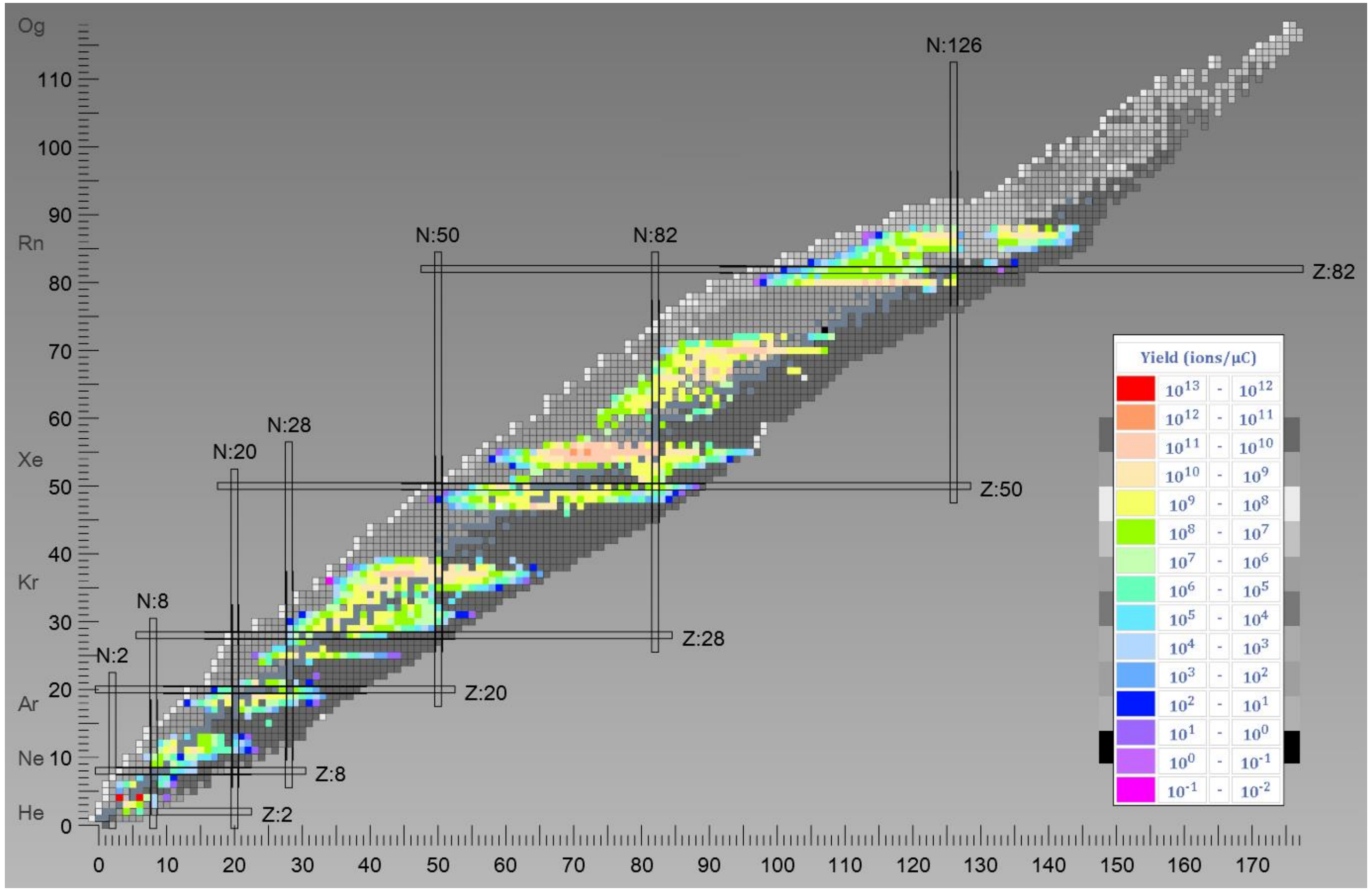
$$Beam\ Intensity = \sigma . j . N_t . \varepsilon$$

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

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

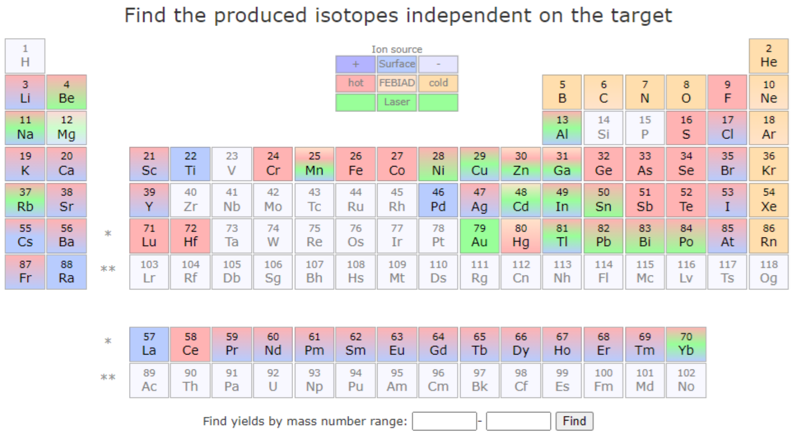


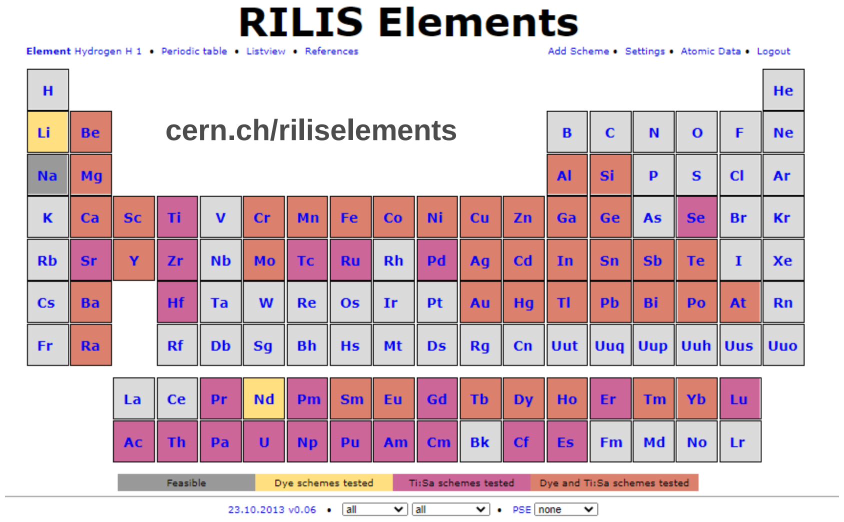
ISOLDE Radioactive beams



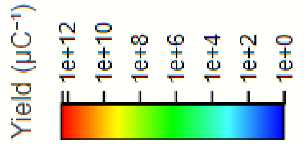
https://isoyields2.web.cern.ch/IsoldeYieldChart.aspx

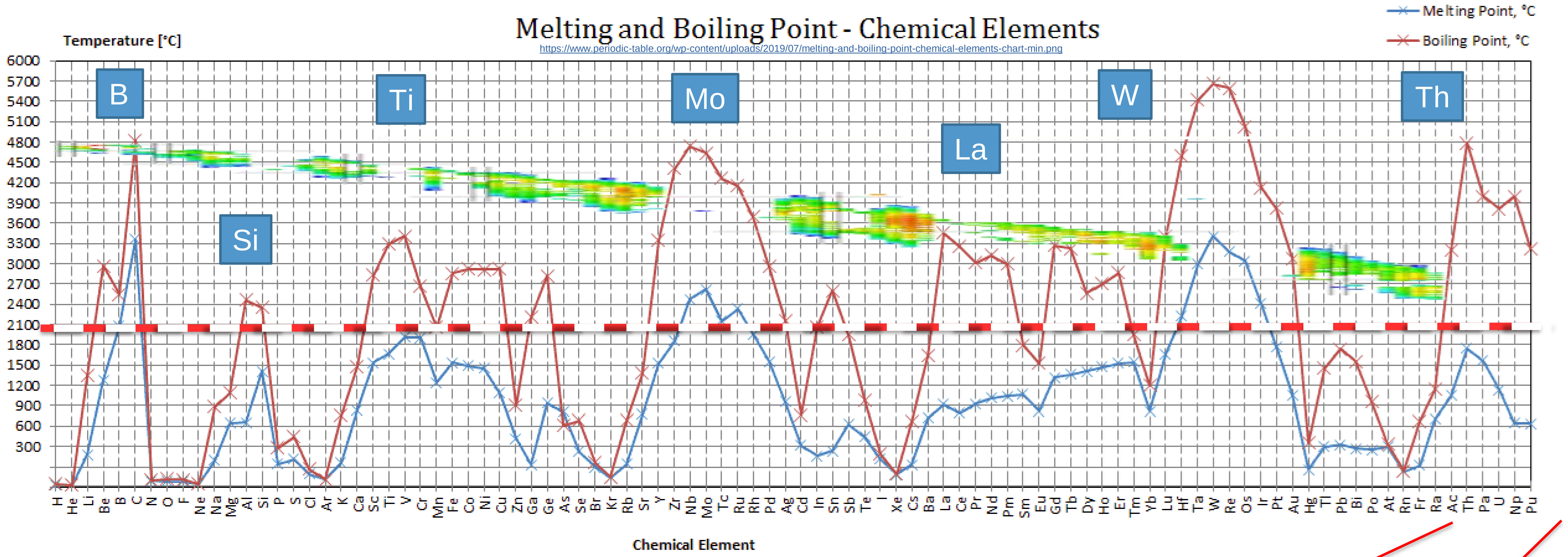
J.Ballof *et.al*, Nuclear Inst. and Methods in Physics Research B 463 (2020) 211





boiling/melting points vs. ISOLDE Yields





actinides

Target materials operated at < 2200 C

S.Rothe | LISA Kickoff CERN | ESR3 - Molecular Actinide Beams

Molecular Beams – Why?

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

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

- **Physics with radioactive molecules**

Article

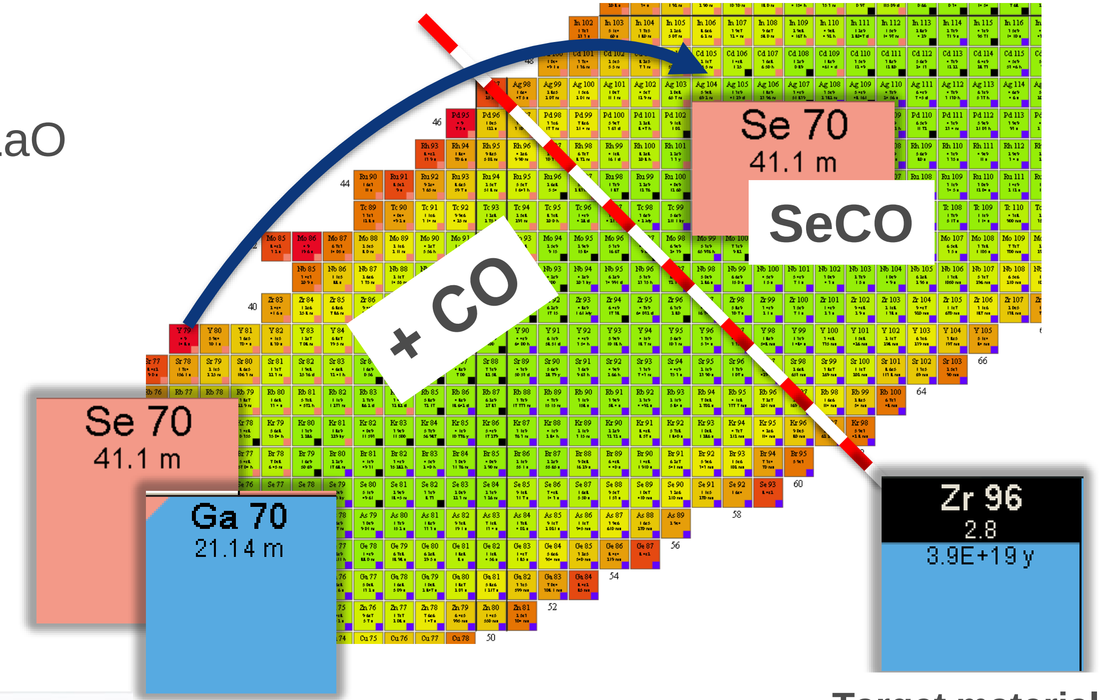
Spectroscopy of short-lived radioactive molecules

https://doi.org/10.1038/s41586-020-2299-4
Received: 24 July 2019
Accepted: 13 March 2020
Published online: 27 May 2020

R. F. Garcia Ruiz[1,2], R. Berger[3], J. Billowes[4], C. L. Binnersley[4], M. L. Bissell[4], A. A. Breier[5], A. J. Brinson[2], K. Chrysalidis[1], T. E. Cocolios[6], B. S. Cooper[4], K. T. Flanagan[4,7], T. F. Giesen[5], R. P. de Groote[8], S. Franchoo[9], F. P. Gustafsson[6], T. A. Isaev[10], Á. Koszorús[6], G. Neyens[1,6], H. A. Perrett[4], C. M. Ricketts[4], S. Rothe[1], L. Schweikhard[11], A. R. Vernon[4], K. D. A. Wendt[12], F. Wienholtz[1,11], S. G. Wilkins[1] & X. F. Yang[13]

(biased example)

https://web.mit.edu/radiomolecules/



Target material

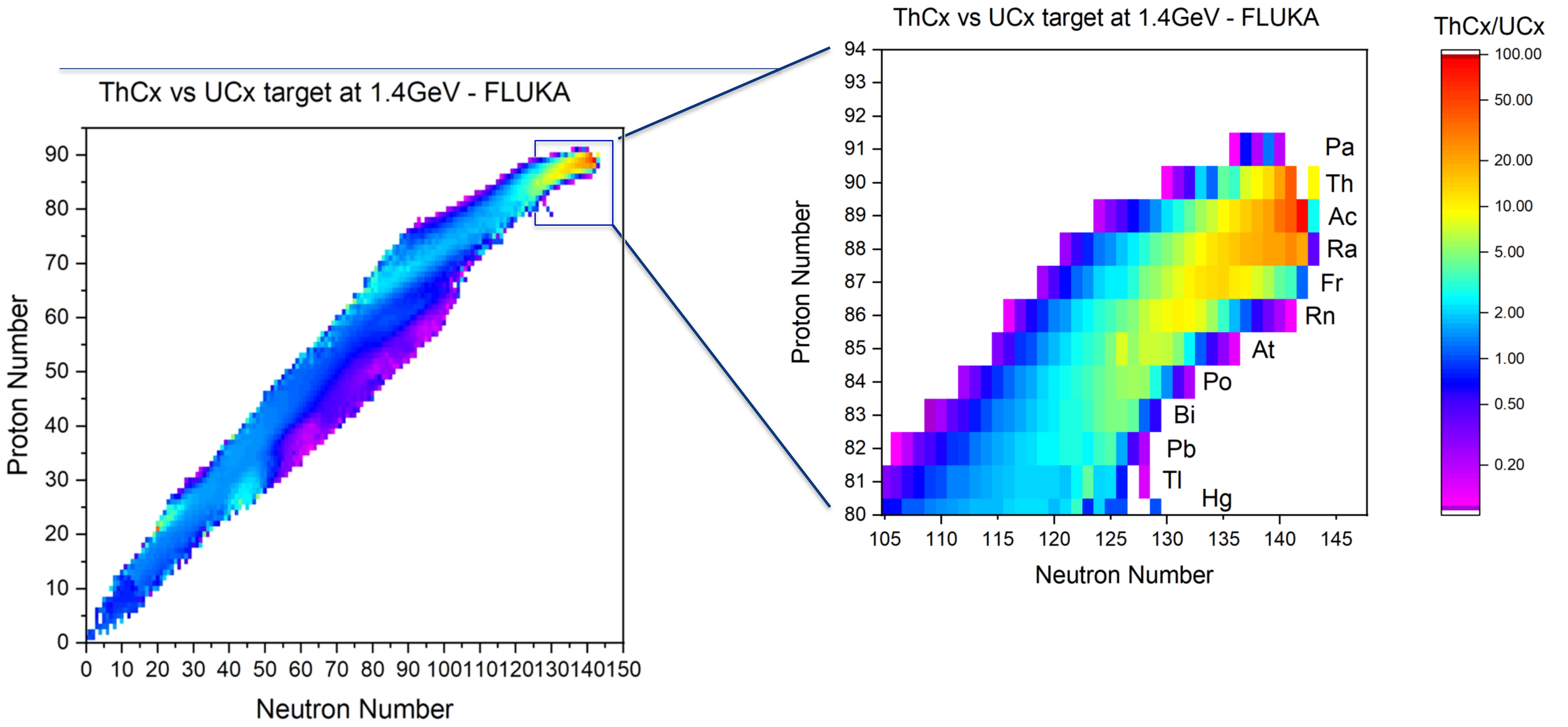
Jochen Ballof | ISOLDE Workshop | 5.DEC.2017 (modified)

Optimize reaction conditions

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

In-target production

Thorium vs Uranium

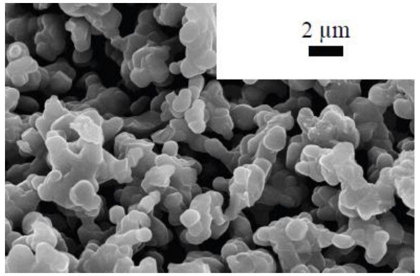


FLUKA Simulations: : Joao Pedro Ramos

Nanomaterials

SiC - S. Fernandes, et al.

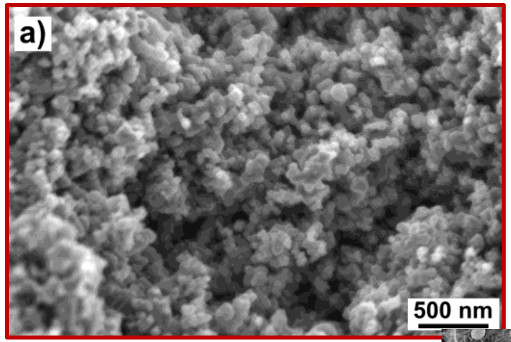
2010



***submicron**

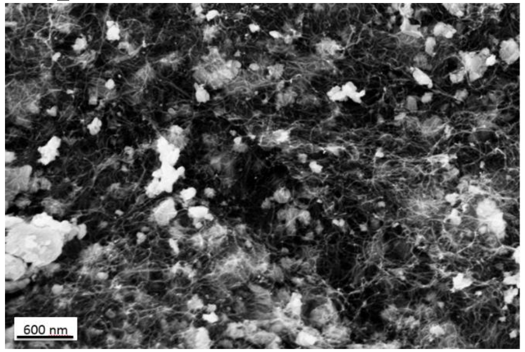
TiC+CB – J.P. Ramos, et al.

Nov- 2014



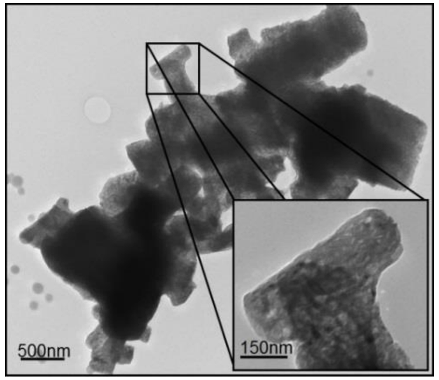
UC_2 + 2C – A. Gottberg, et al.

Nov - 2014



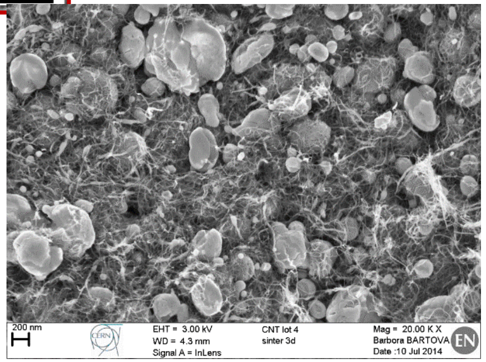
***1st nanomaterial at ISOLDE**

2011



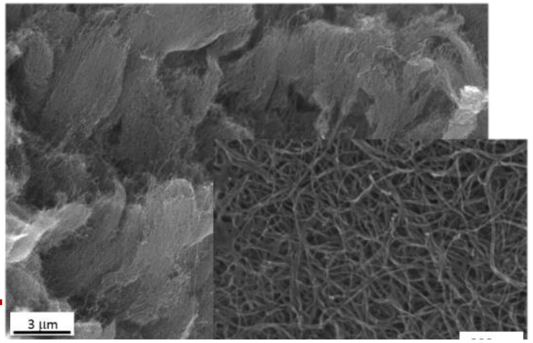
CaO – J.P. Ramos, et al.

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

Ionization / molecular formation techniques

- Surface ->In target molecular, BaF, RaF
- Plasma (FEBIAD/VADIS) -> BeF_2, TiF_3, SeCO
- E-impact : $Mo(CO)_6$
- Direct laser -> ionization LaF, RaF (to be tested YOL2, GPS)

See talk by S.Wilkins

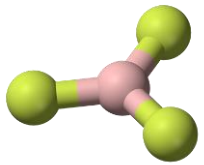
- Photochemistry -> hints seen with Ac/AcO, U/UO
  - Isotope selective ?
- In-trap formation :
  - U+ -> UO+ seen in ISOLTRAP's RFQ.
  - RFQcb@YOL2 , ISCOOL@ISOLDE
    - Addition of reactive gas to promote molecular species

c.f.: M. Fan et al, 2021, *Optical mass spectrometry of cold RaOH+ and RaOCH3+*

    - laser access, study breakup / photochemistry

Example: Boron beams at ISOLDE

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



Boron

Low Volatility (m.p. 2076 °C)

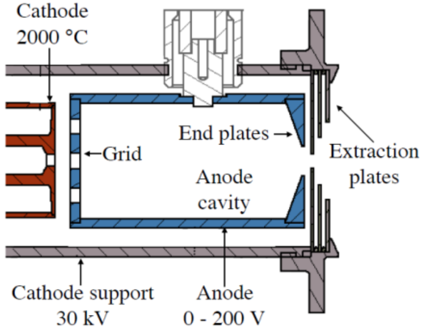
reactive with many metals

Boron trifluoride

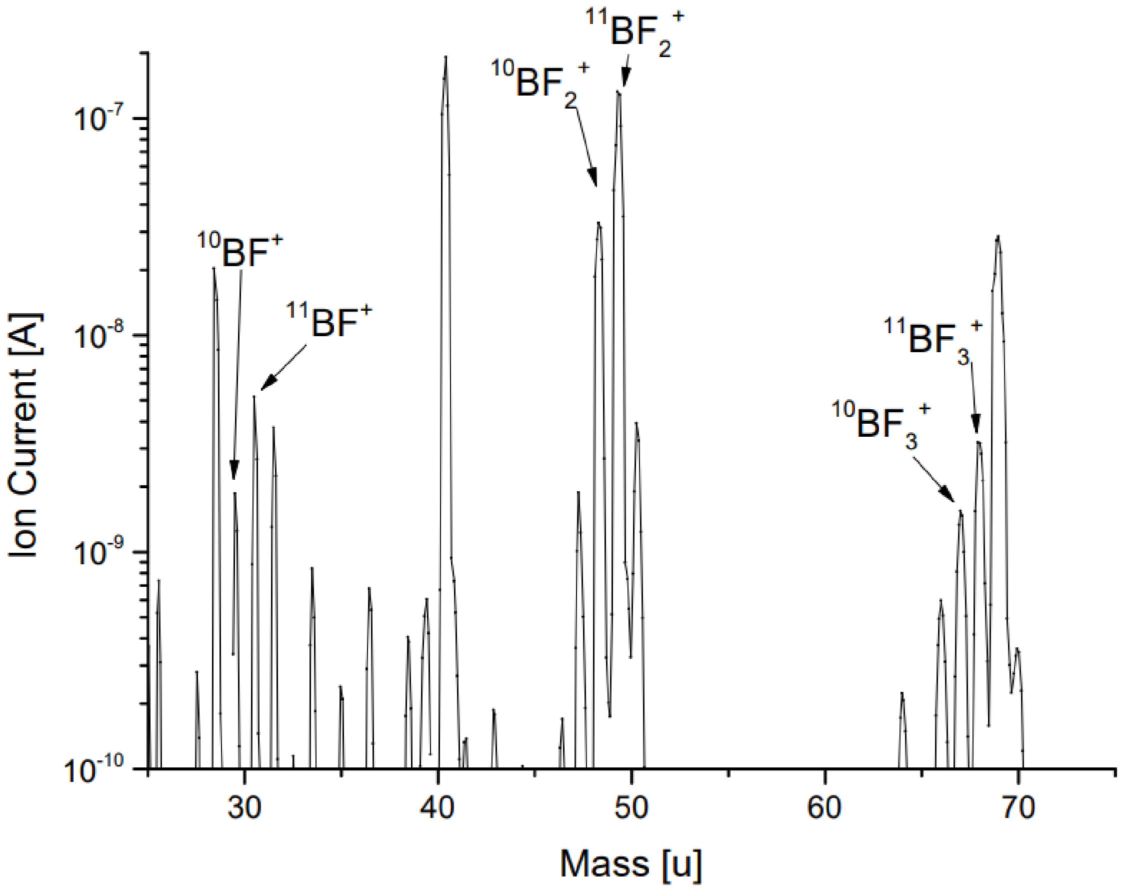
gaseous even at RT

very stable

VADIS / FEBIAD ion source



Y. Martinez Palenzuela, Thesis (https://lirias.kuleuven.be/handle/123456789/636675)



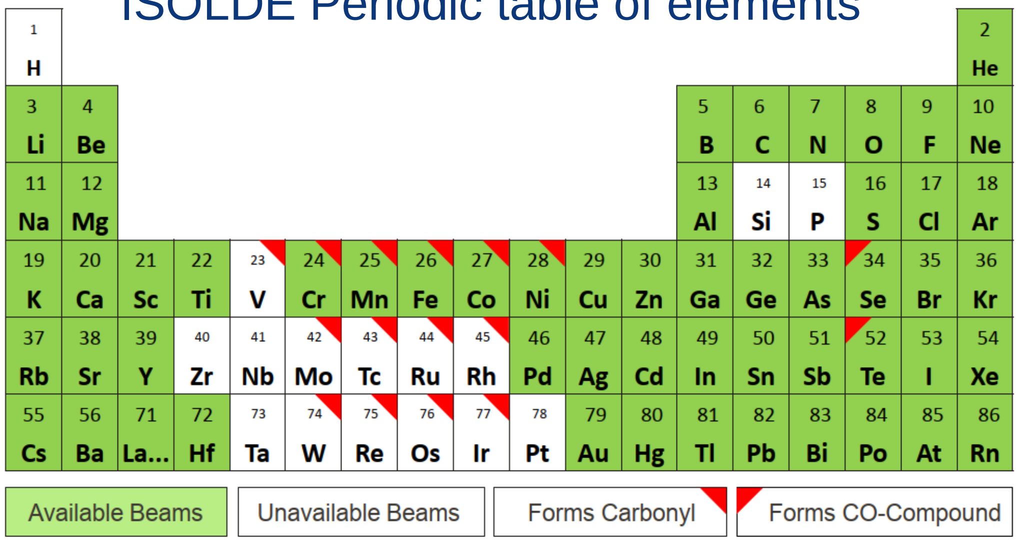
J. Ballof, 2019, *Radioactive boron beams produced by isotope mass separation at CERN-ISOLDE*, *Eur. Phys. J. A 55 (2019) 65*

Refractory Metal Carbonyl beams

Extraction as complex compounds

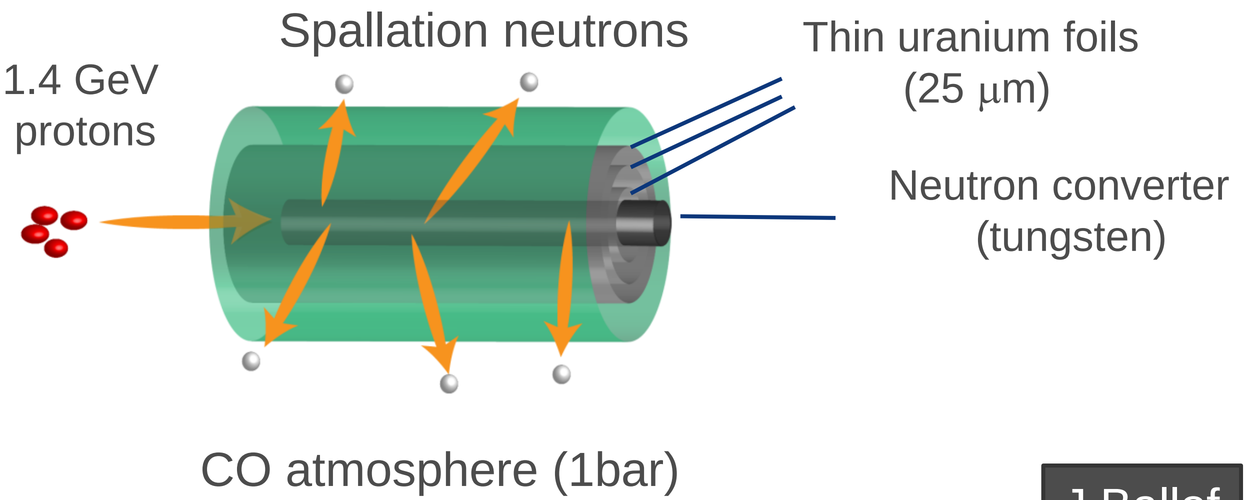
e.g. Mo + 6 CO → $Mo(CO)_6$

ISOLDE Periodic table of elements



$Mo(CO)_6$ is fragile:

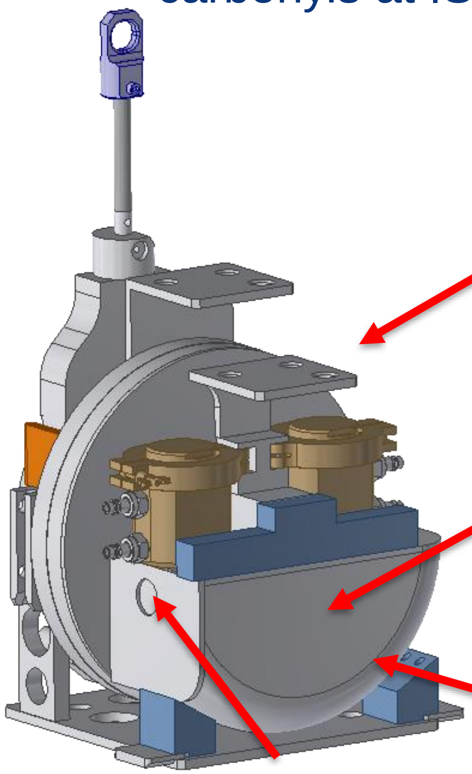
- Decomposition in beam induced plasmas
- Using a neutron converter to reduce charged particle fluence

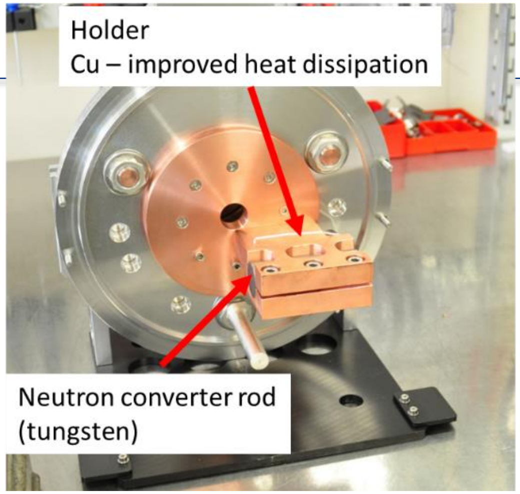


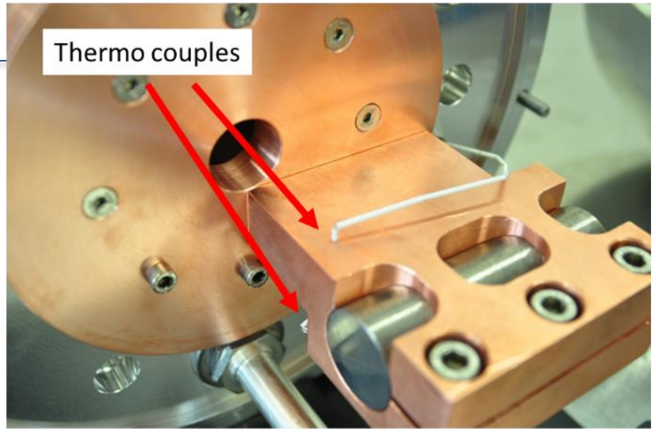
J.Ballof

Refractory Metal Carbonyl beams

Dedicated Experiment to study formation and decomposition of carbonyls at ISOLDE

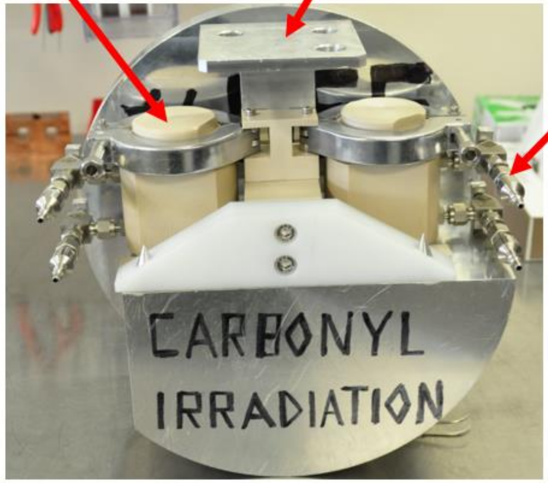




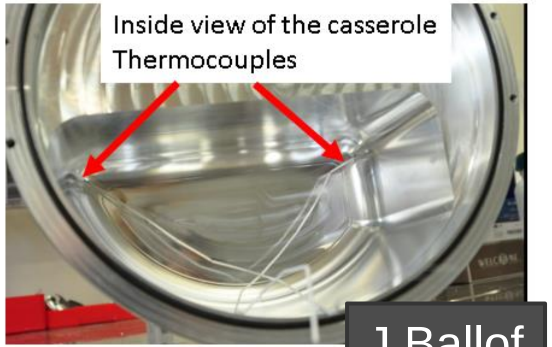


PEEK gas containers with KF clamping

Handle for the sample assembly



Swagelok Angle Valves (radiation hard) and Stäubli RBE03 quick connectors



J.Ballof

Photocathode source

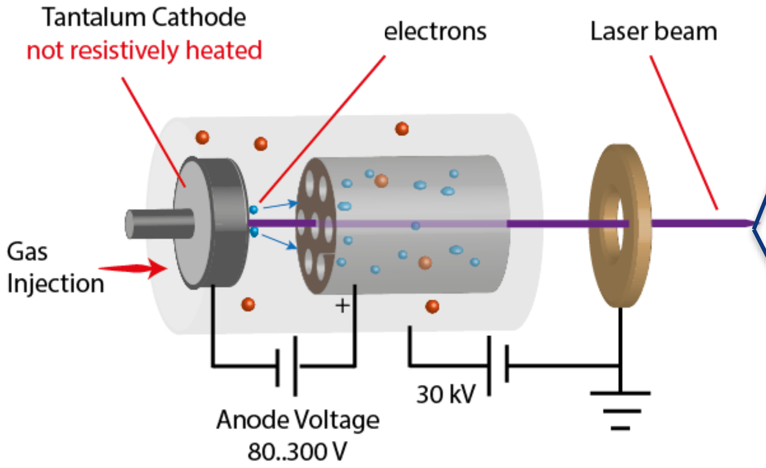
VADIS source at ambient temperature

Photon-induced electron generation

Motivation

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

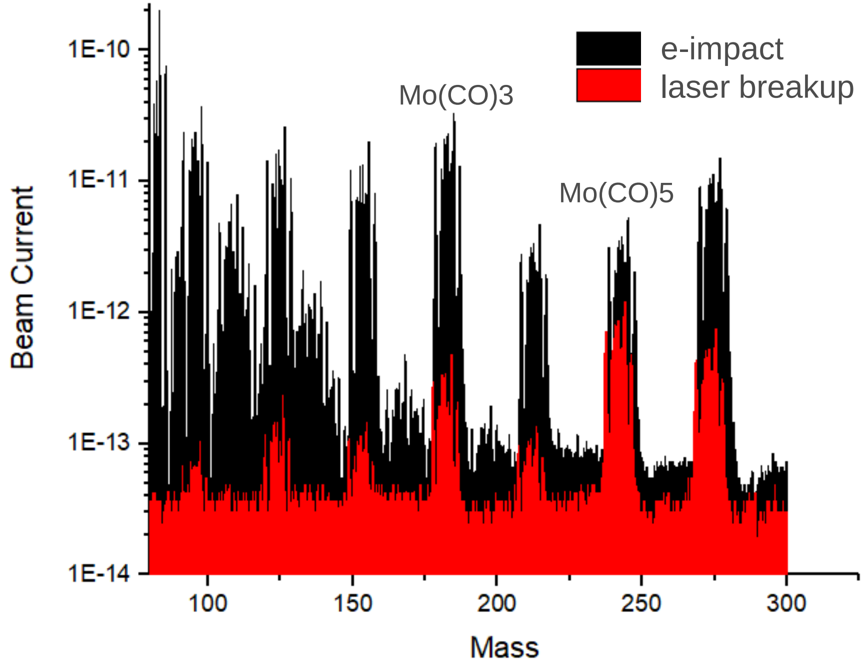
Set up



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

Studying molecular beam formation

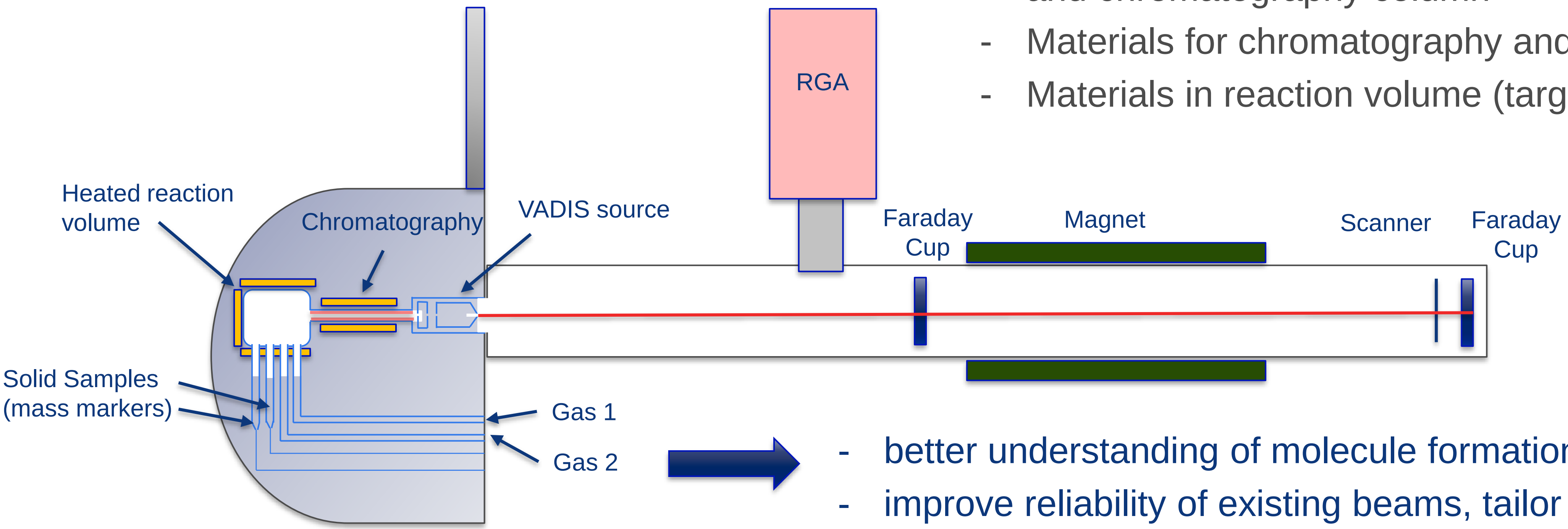
Concept for a dedicated development unit for molecular beams

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)

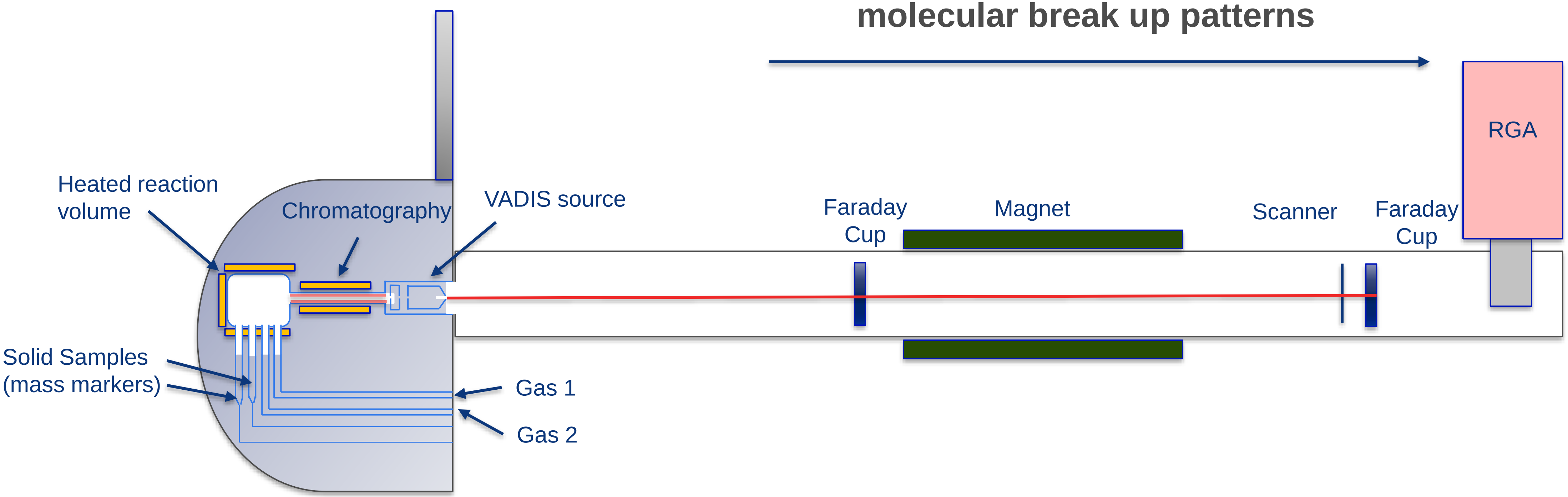


- better understanding of molecule formation
- improve reliability of existing beams, tailor new beams

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**

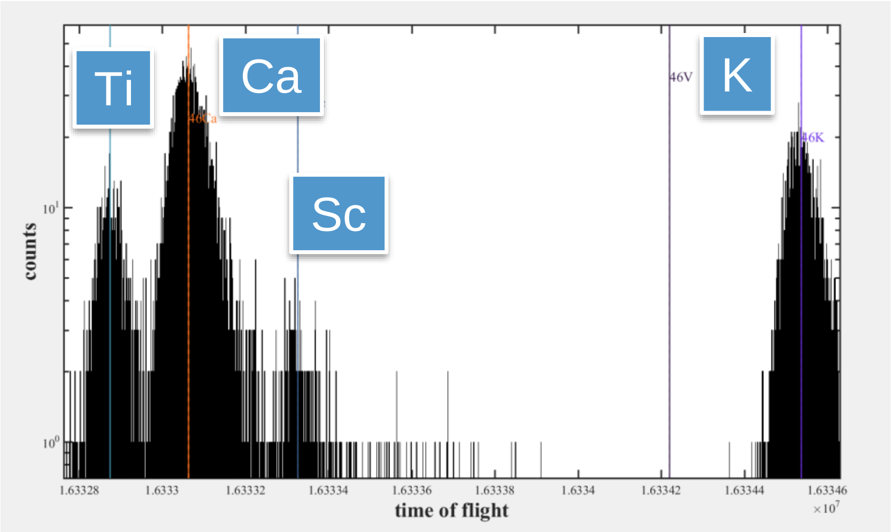


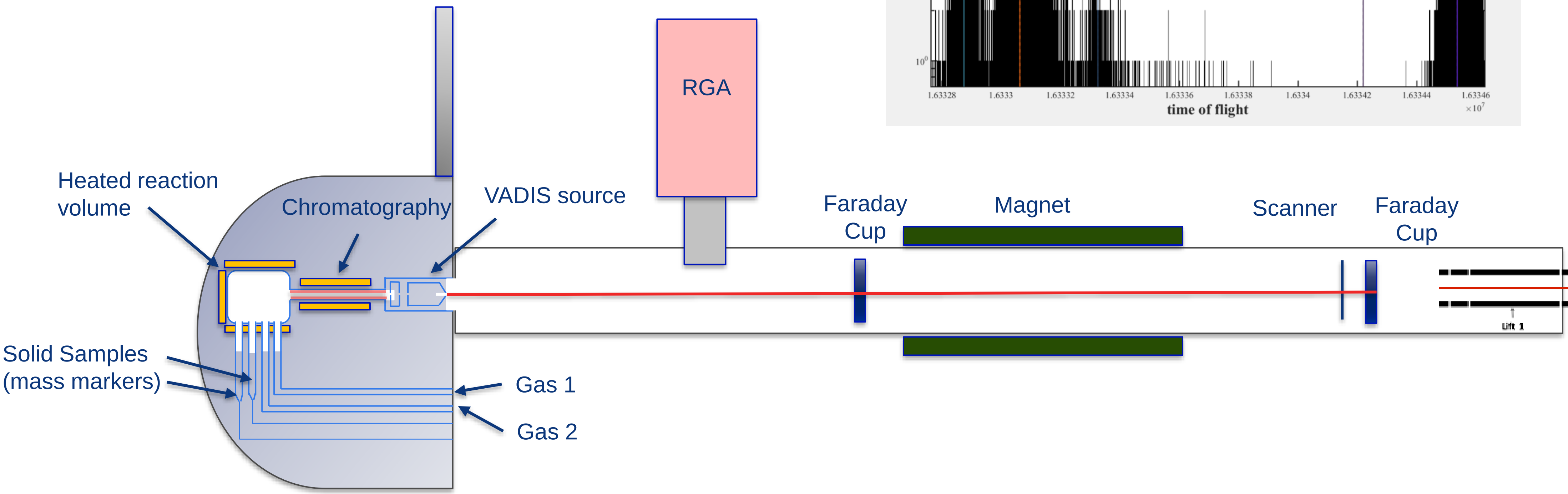
Studying molecular beam formation

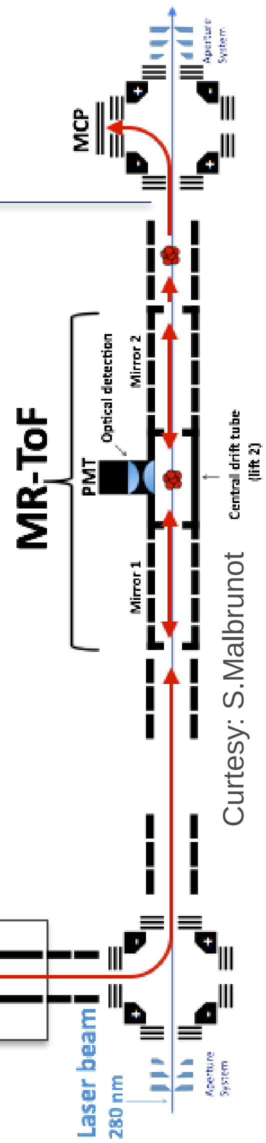
Concept for a dedicated development unit for molecular beams

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

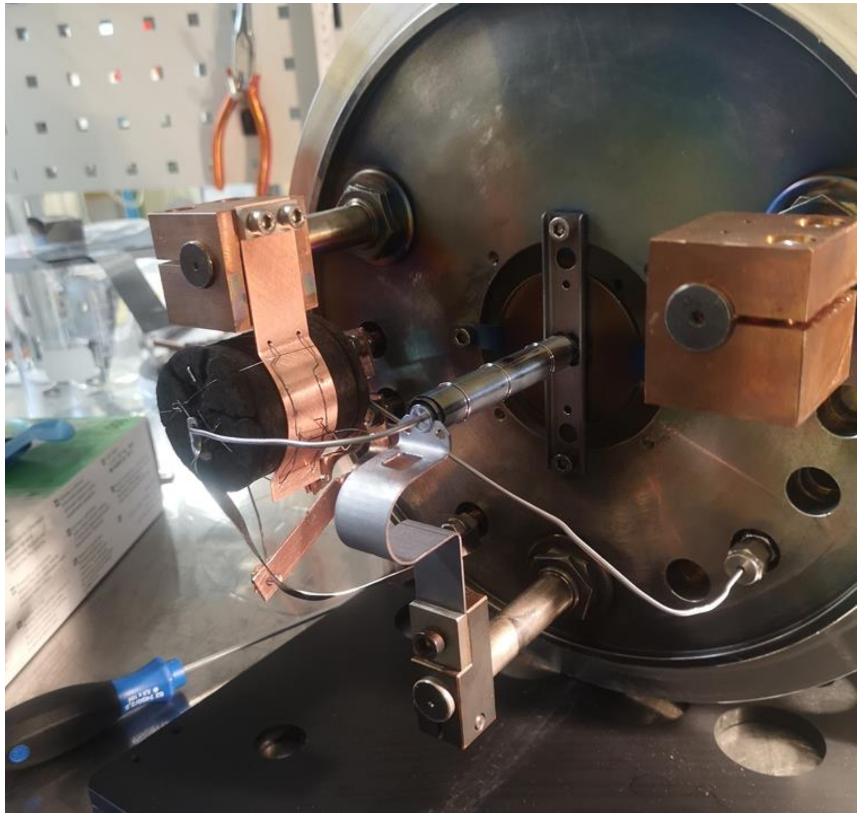
Example TOF spectrum on mass 46



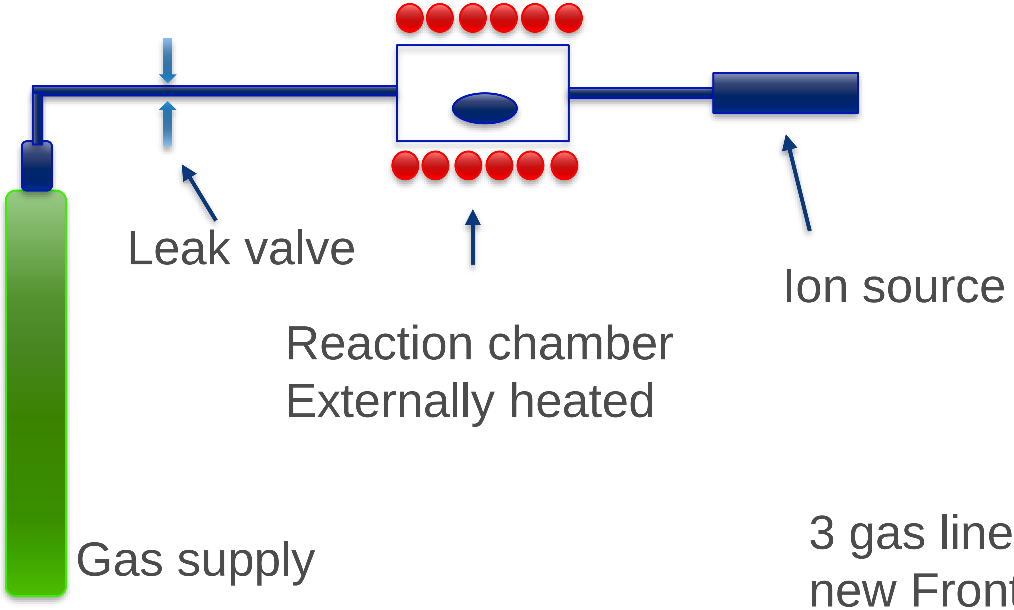




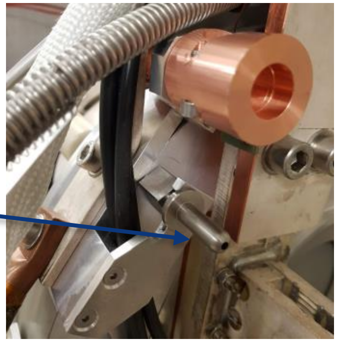
First prototype for reaction chamber tested at YOL1



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



3 gas lines to
new Frontends

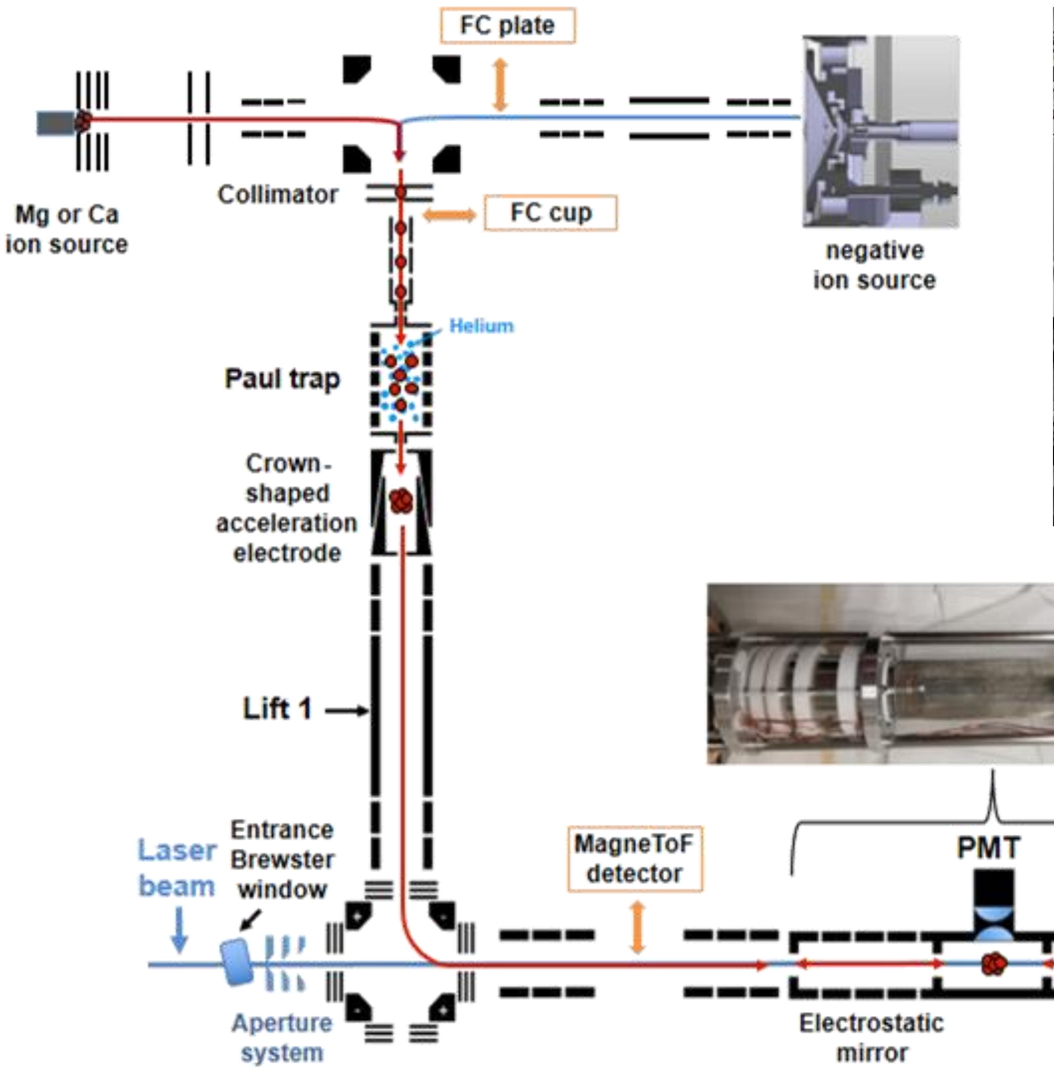


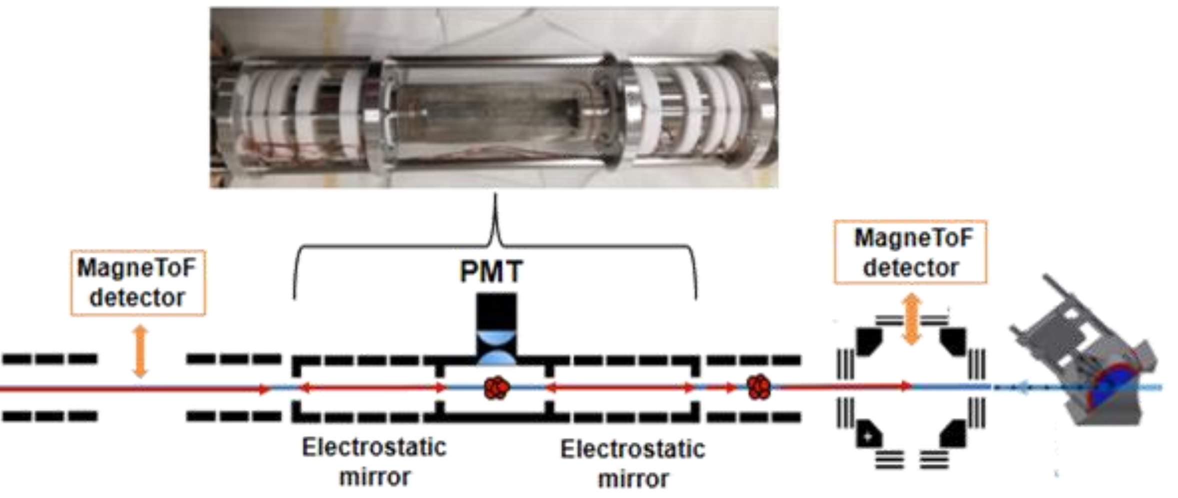
1 reacting gas
1 sample supply
Surface ionization

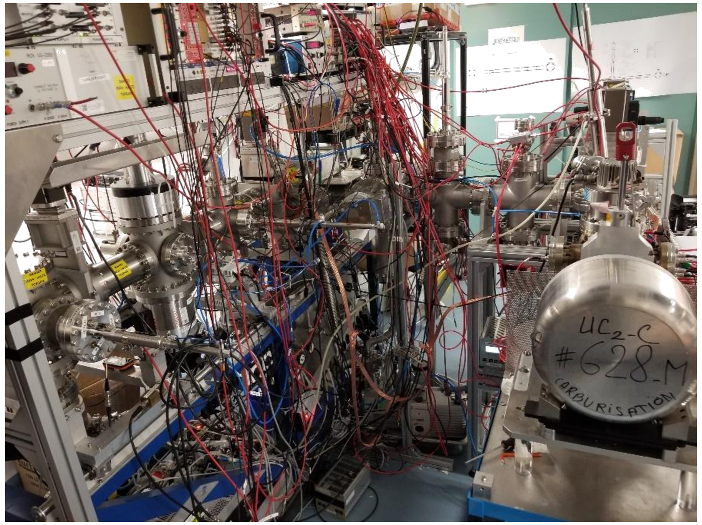
Multiple gasses
Plasma sources, Negative ions

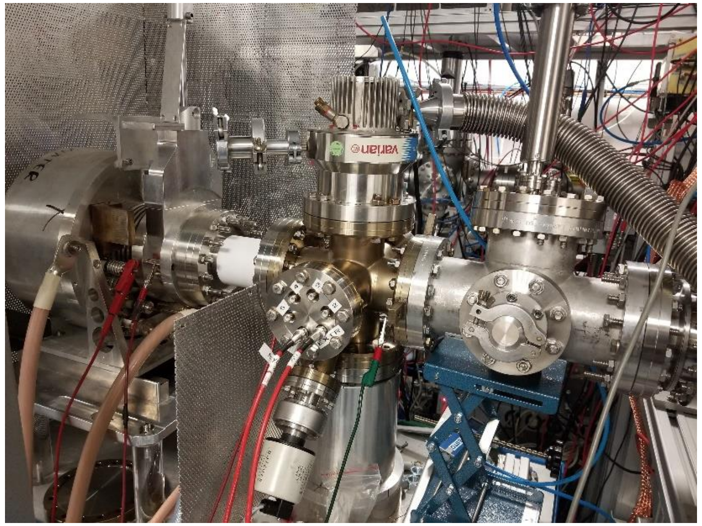
V.Samothrakis, M.Ballan, J.Ballof, D.Leimbach, B.Crepieux, S.Rothe et al.

ISOLDE target at MIRACLS PoP





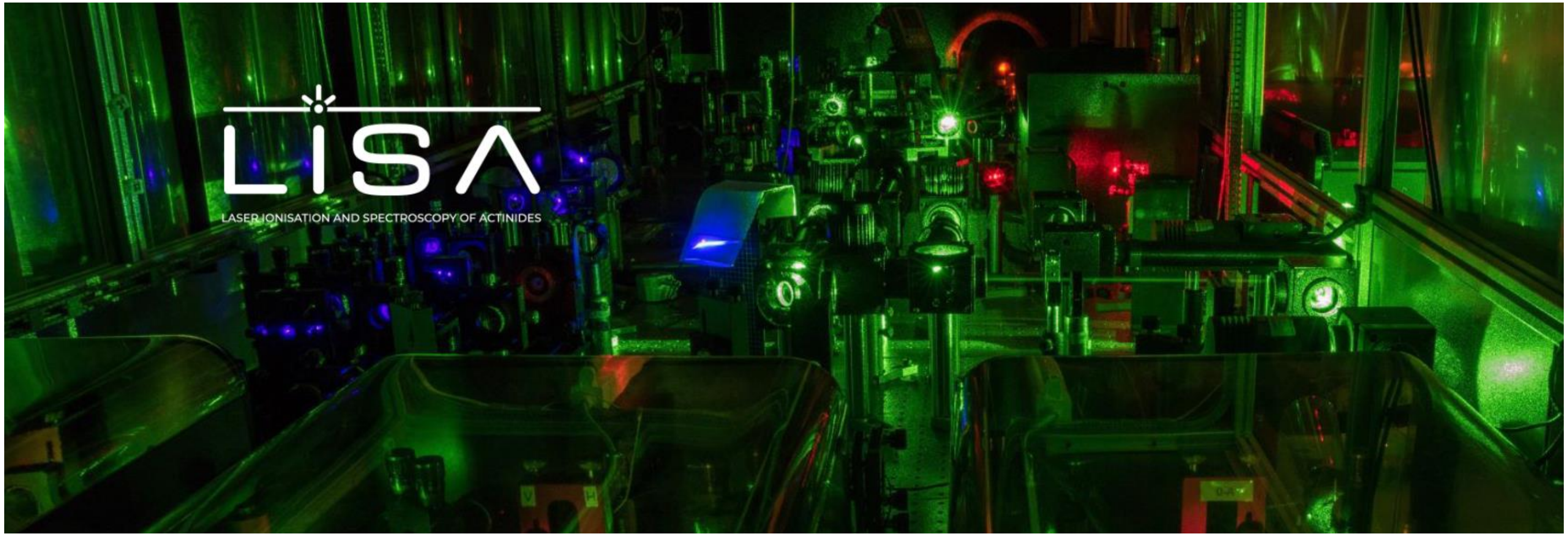




- Ion sources, compatible to ISOLDE target
- Paul trap
- MR-ToF
- Laser access and PMT

D.Leimbach (PhD. Thesis)
F. Maier et al, 2019, Hyperfine Interact (2019) 240: 54

LISA – Laser Ionization and Spectroscopy of Actinides



ESRs at ISOLDE-CERN

Bianca Reich

ESR 02

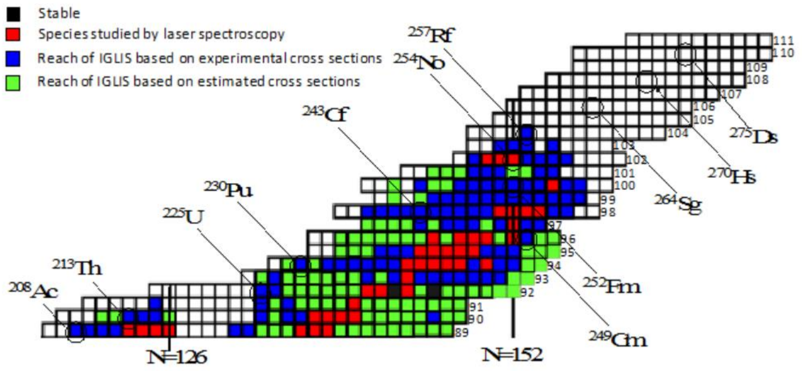
Development of high-resolution in-source hot-cavity RILIS methods for actinides.

Mia Au

ESR 03

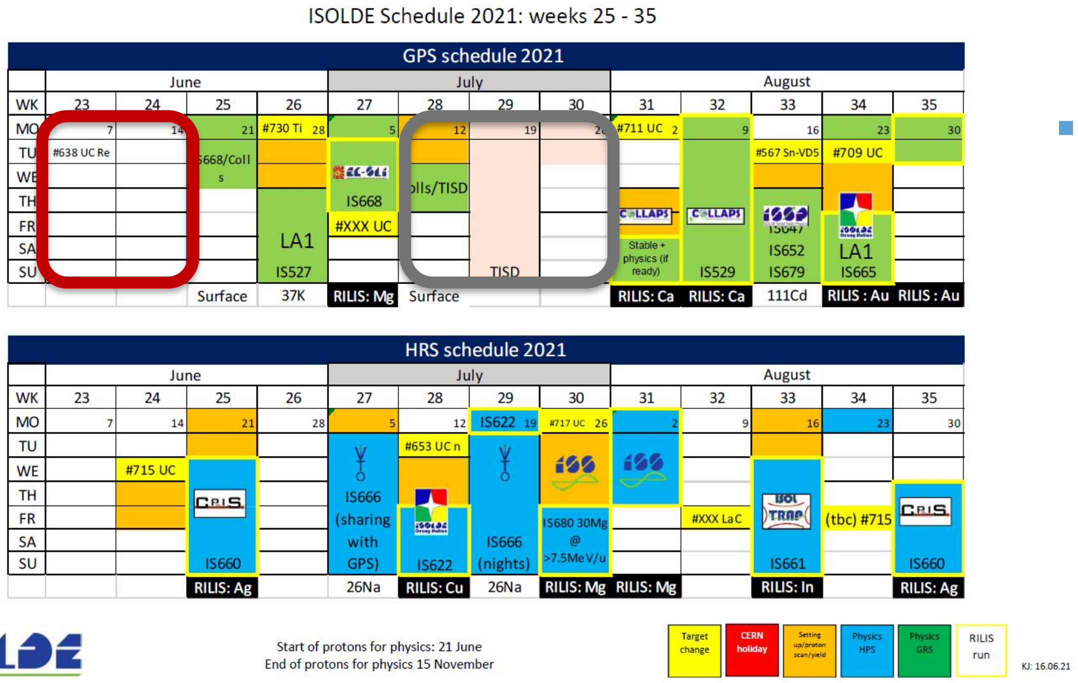
Target developments for extraction of actinides from thick ISOL targets followed by laser-induced molecular break-up and/or ionization.

https://lisa-itn.web.cern.ch



LISA – Laser Ionization and Spectroscopy of Actinides.
This Marie Sklodowska-Curie Action (MSCA) Innovative Training Networks (ITN) receives funding from the European Union’s H2020 Framework Programme under grant agreement no. 861198

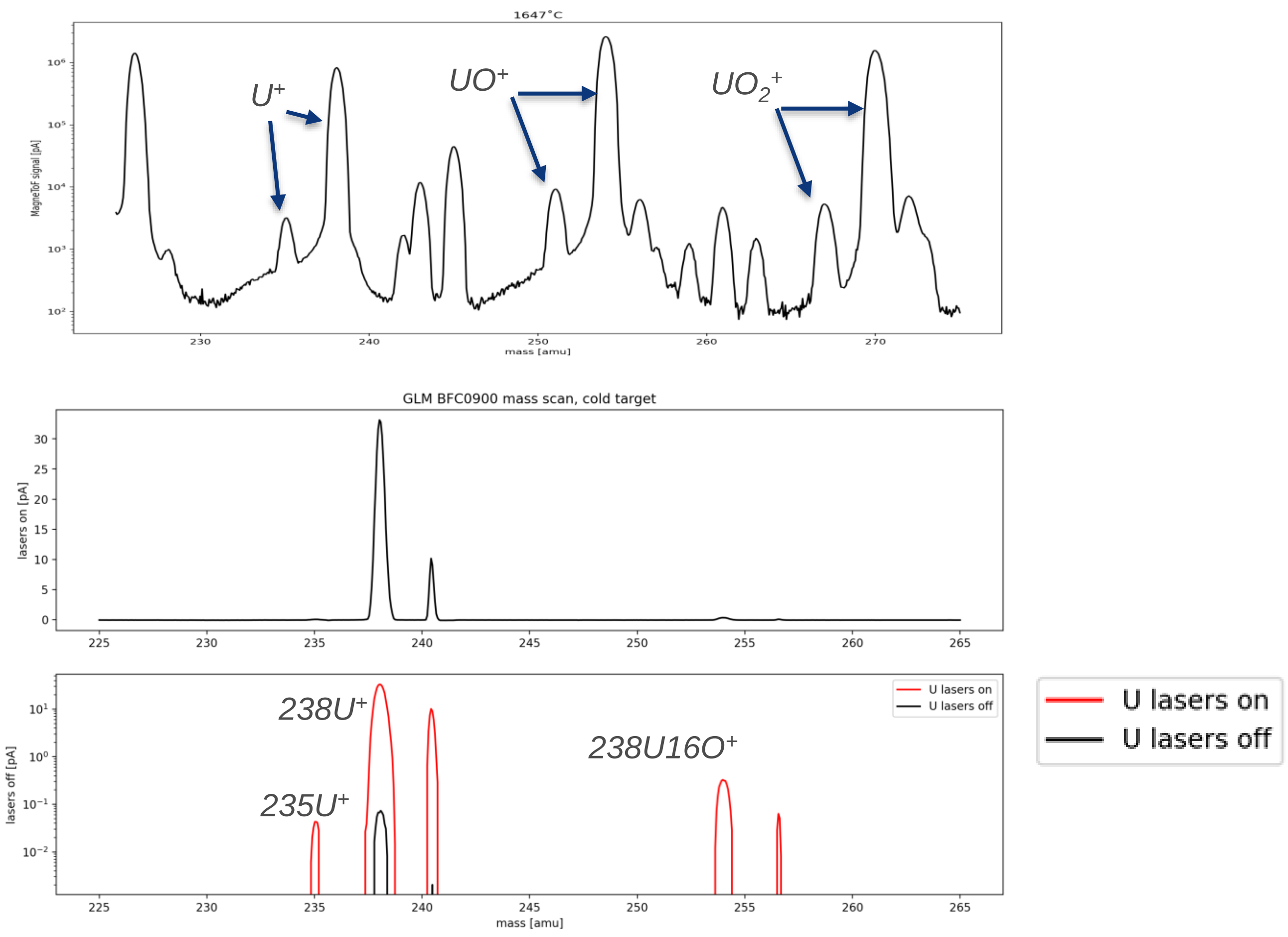
Pre-irradiated targets (June 2021), **M.Au** et al.

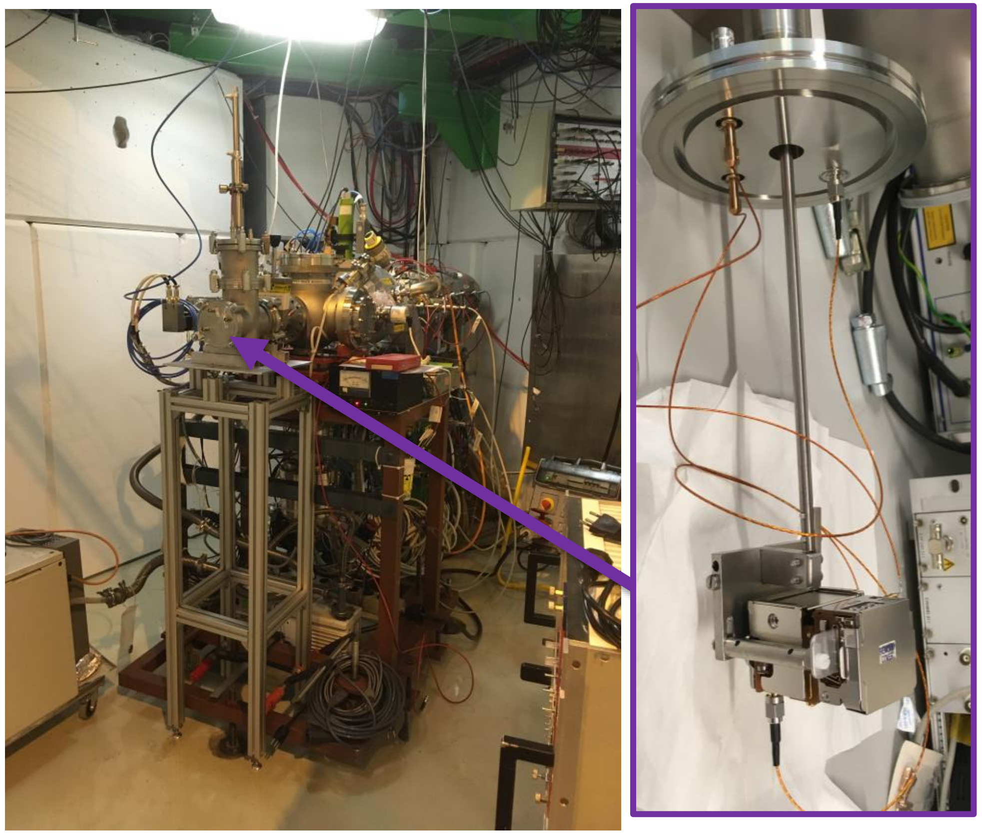


- <u>1. #638-UC-Re:</u>
  - Target irradiated end of run2 at ISOLDE (2018) before LHC long shutdown 2 (LS2)
  - Preparation of July TISD beam time
  - Test single ion detectors
  - Tune to ISOLTRAP MR-ToF

Letter of Intent: **Radioactive molecules at ISOLDE**:
NTC I-227 (http://cds.cern.ch/record/2748712)

Radioactive molecules at ISOLDE (June 2021)





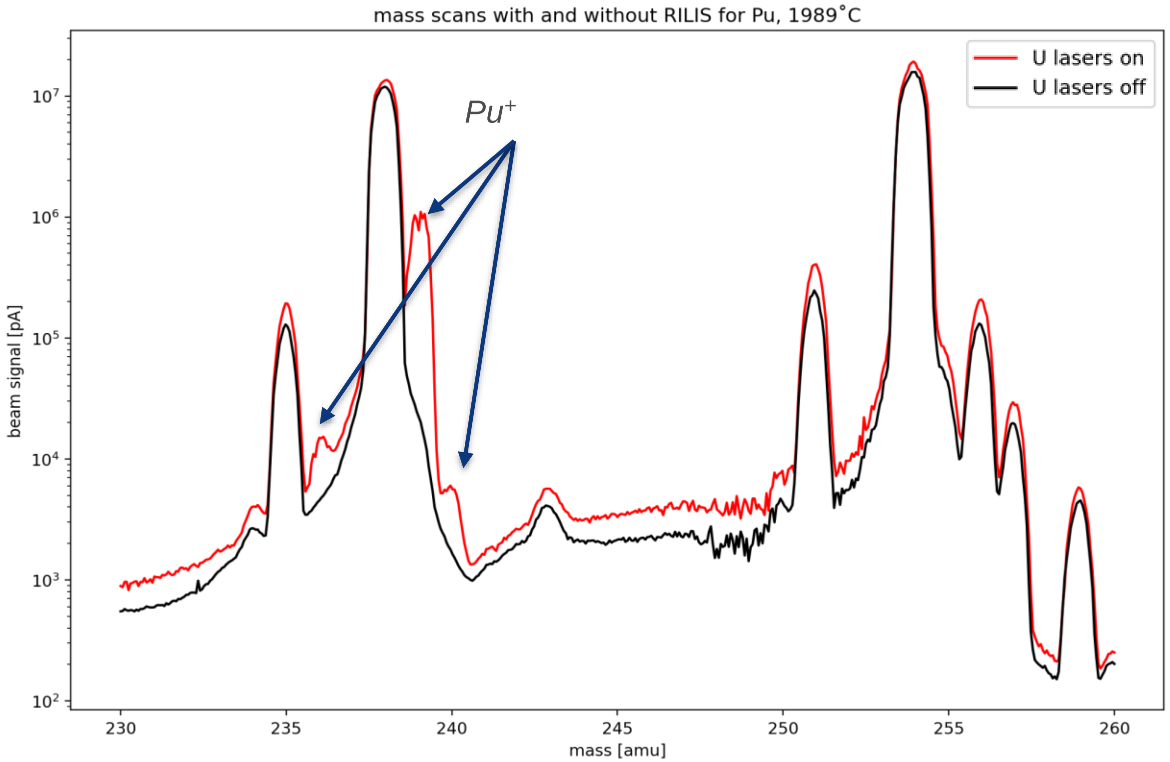
Installation of TISD chamber with MagneToF detector and signal plate at ISOLDE GLM

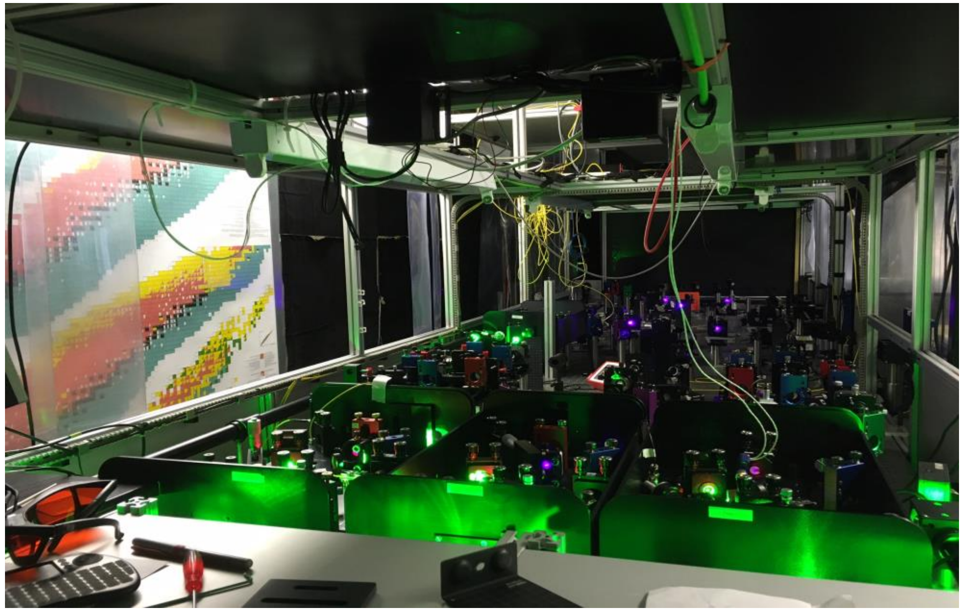
Photochemistry ?, tbc.

M.Au et.al

Radioactive molecules at ISOLDE (June 2021)

- Atomic actinide beams (transuraniums : Np, Pu)
- **Identified at ISOLTRAP MR-ToF**



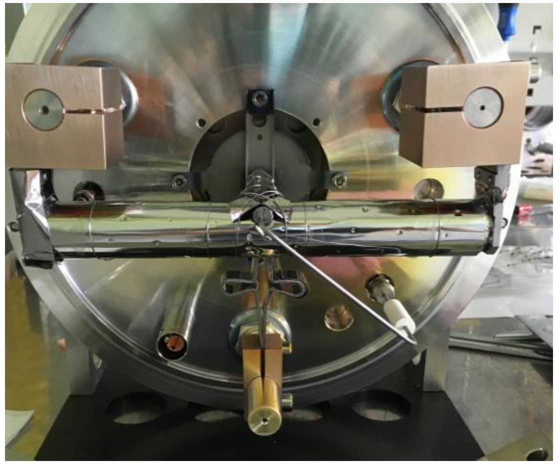


RILIS lab at ISOLDE set up for fast switching between two-step ionization schemes for actinides

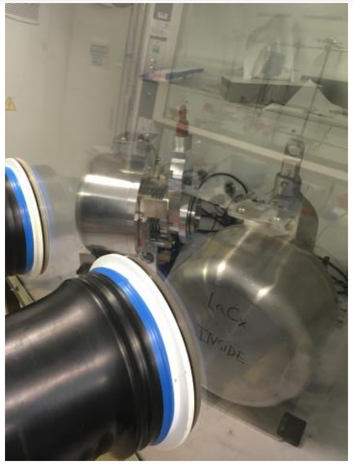
M.Au et.al

Offline molecular beam development (YOL1)

- Targets and material samples
- Gas injection: NF_3, CF_4, SF_6
- Ion sources: surface, plasma, RILIS
  - Direct ionization
  - Dissociation ionization
  - Thermal dissociation
- Reaction condition studies

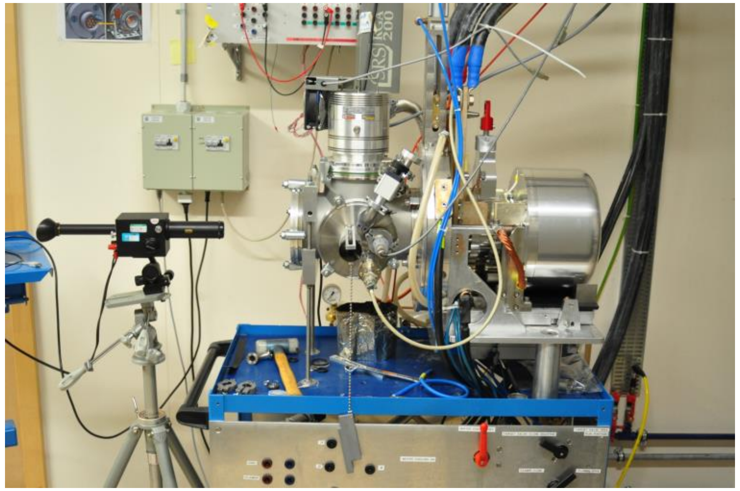


An ISOLDE target and ion source unit with gas injection



Two target units in a glove box for sample exchange

LaC is pyrophoric !



ISOLDE PUMP STAND

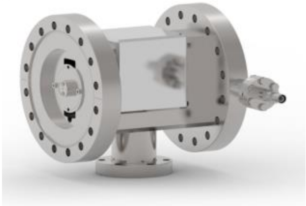
ISOLDE OFFLINE 1 (YOL1)

Offline molecular beam developments (YOL2)

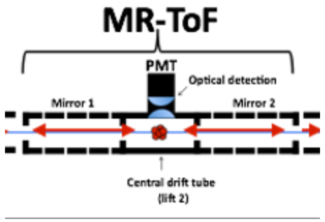
- 1) In-source laser spectroscopy of stable molecules
  - Laser lab is ready
  - magneToF installed
  - LaF target ready #712
- 2) In-trap molecular formation/dissociation
  - **Laser access to trapped region**
  - **gas system upgrade** underway
  - ! detection and identification after creation in RFQcb
    - Design ongoing
    - **Seed funding received !**

2021

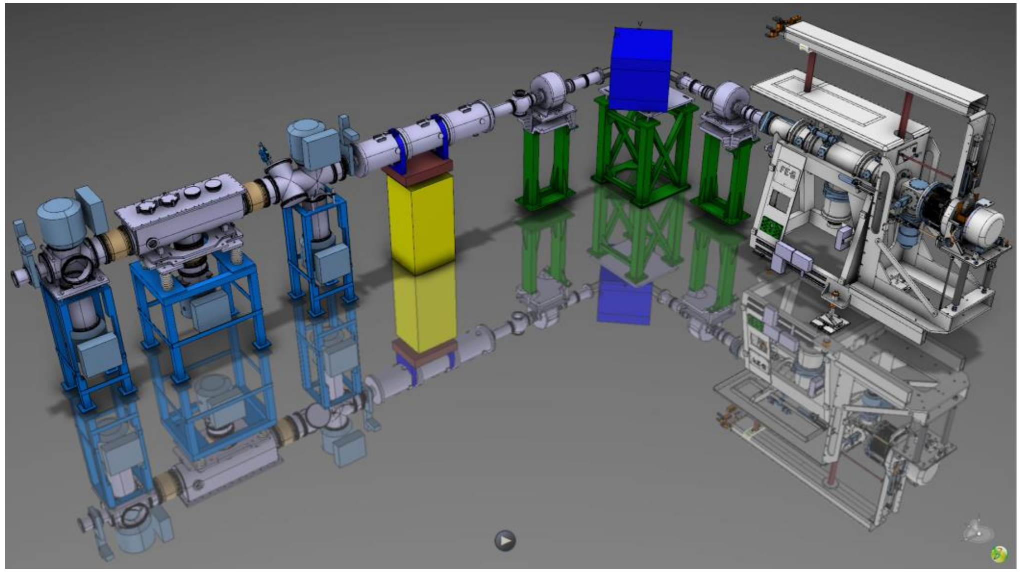
~Wien filter



>2023

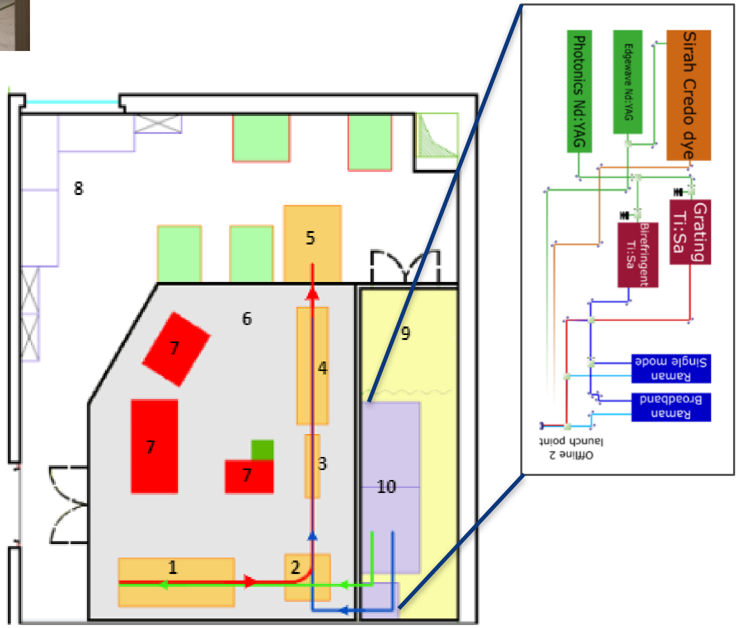


ISOLDE OFFLINE 2 (YOL2)



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

Laser Room @ YOL2

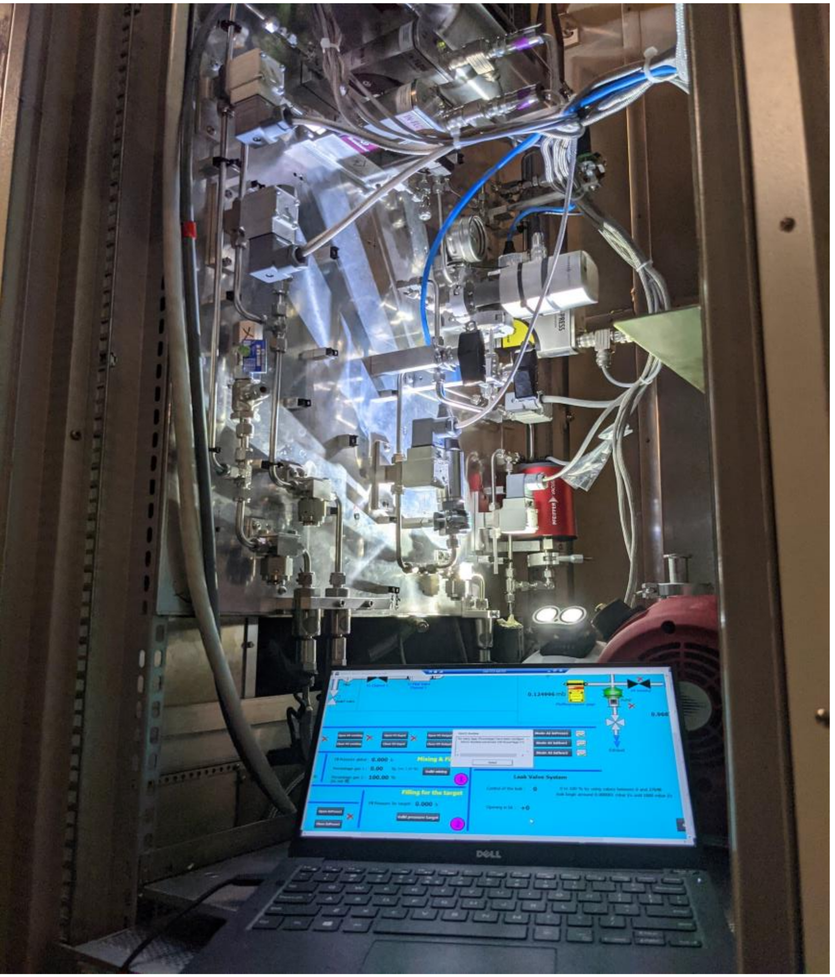


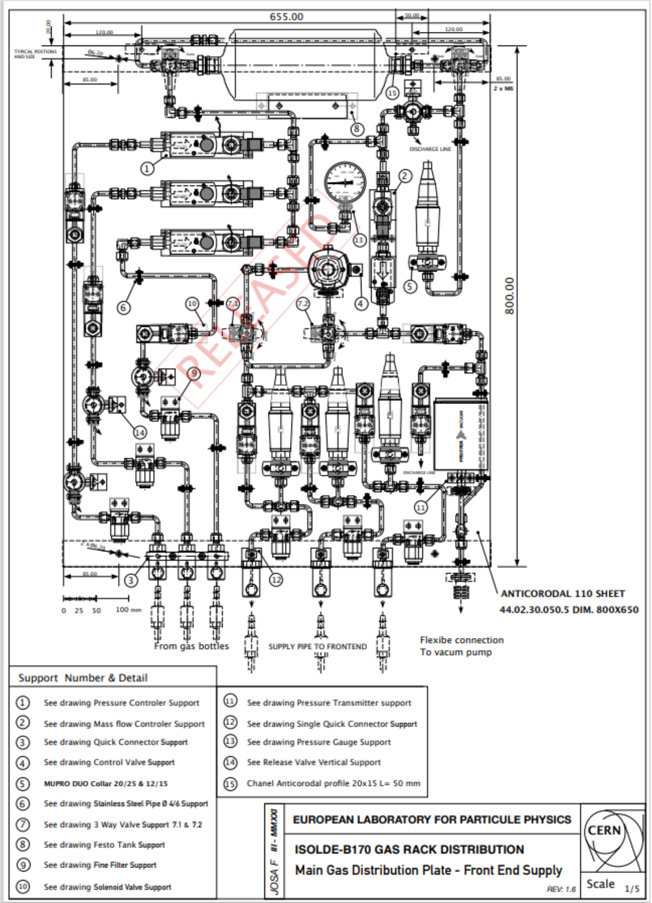
From: A.Ringvall *et.al* , (to be published)

M.Athanasakis-Kaklamanakis , M.Au et.al

M. Fan et al, 2021, *Optical mass spectrometry of cold RaOH+ and RaOCH3+*
A. Ringvall Moberg et al, 2020, *Time-of-Flight study of molecular beams extracted from the ISOLDE RFQ cooler and buncher*

HRS, GPS, YOL2 gas systems

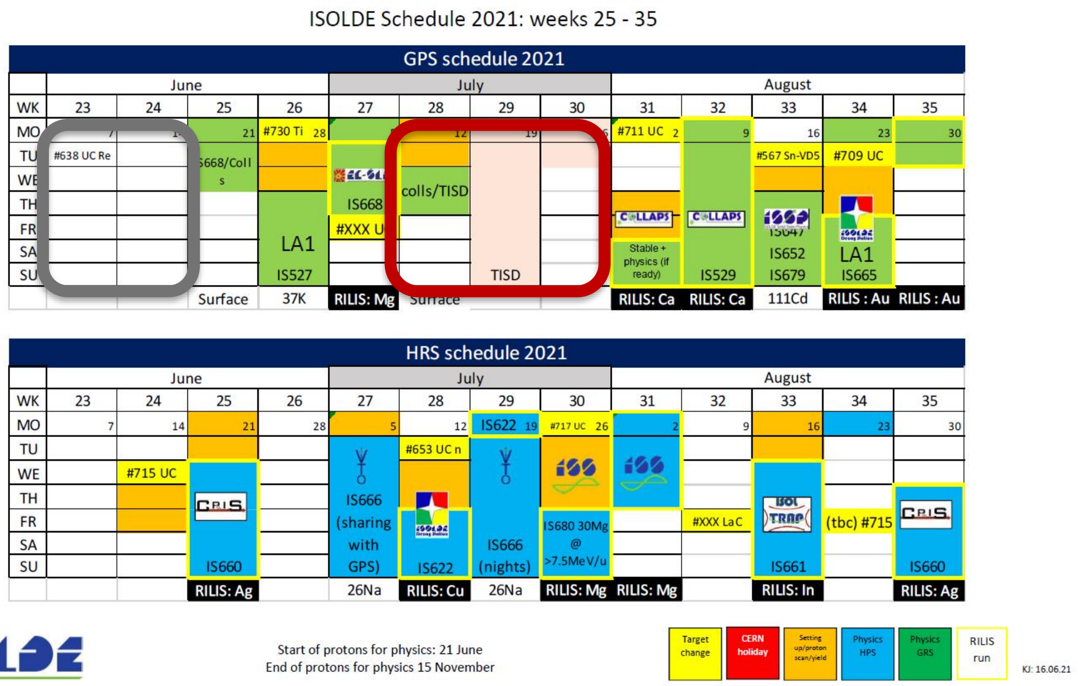




- In-place gas mixing
- Auto filling / flushing

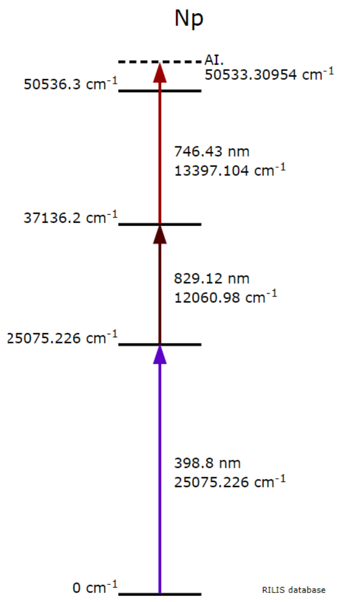
- **Hardware installed and tested, expert CTRL ready.**

Radioactive molecules at ISOLDE: target and ion source development (TISD) campaign (**M.Au *et al.***)

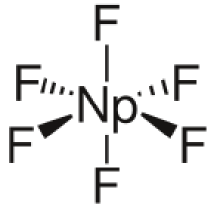


See talk by S.Wilkins

1. #637-UC-W:
   - RaF **in-source** laser spectroscopy
2. #713-UC-VD5:
   - **Actinide fluoride** production, identification and laser manipulation
   - CF4 injection
   - **Identification** via decay station or ISOLTRAP **MR-ToF**
   - **Single ion detection** at GLM beamline
   - Possible **laser ionization**, probing of dissociation
   - Check for 229Pa ! ☺



e.g. NpF6
BP: 55C



Letter of Intent: **Radioactive molecules at ISOLDE**: NTC I-227 (http://cds.cern.ch/record/2748712)

ISOLDE first radioactive beams after LS2 - June 2021

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