A cold electron-impact ion source driven by laser-induced electron emission

New opportunities for radioactive molecular beams?

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GSI

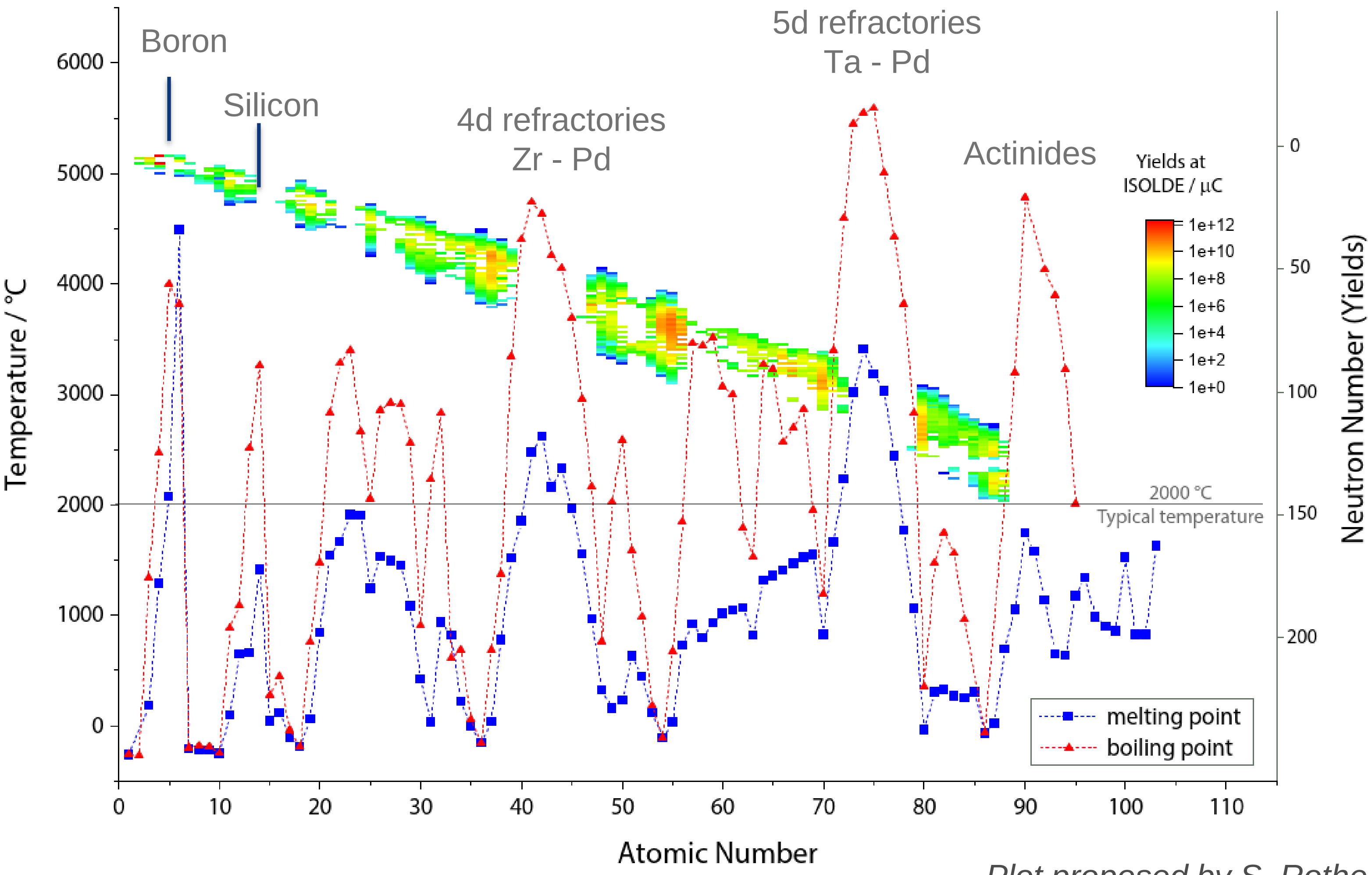
ICIS'21

ISOLDE
and available beams

(Un)available beams at ISOLDE

- **Available beams**
  - more than 1000 radioisotopes
  - 74 chemical elements
  - lightest element: Helium
  - heaviest element: Uranium
  - half-lives down to ~ ms

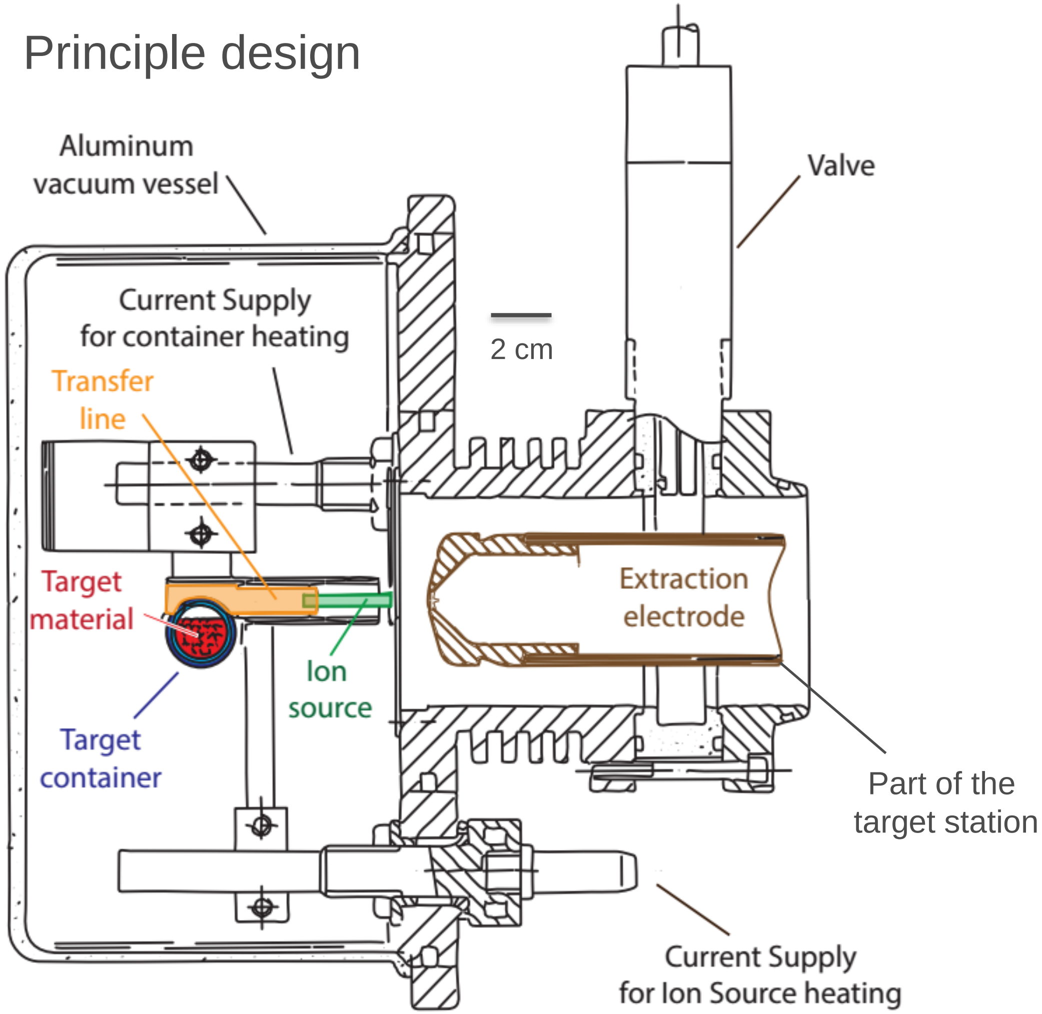
- **Unavailable beams**
  - Many refractory elements

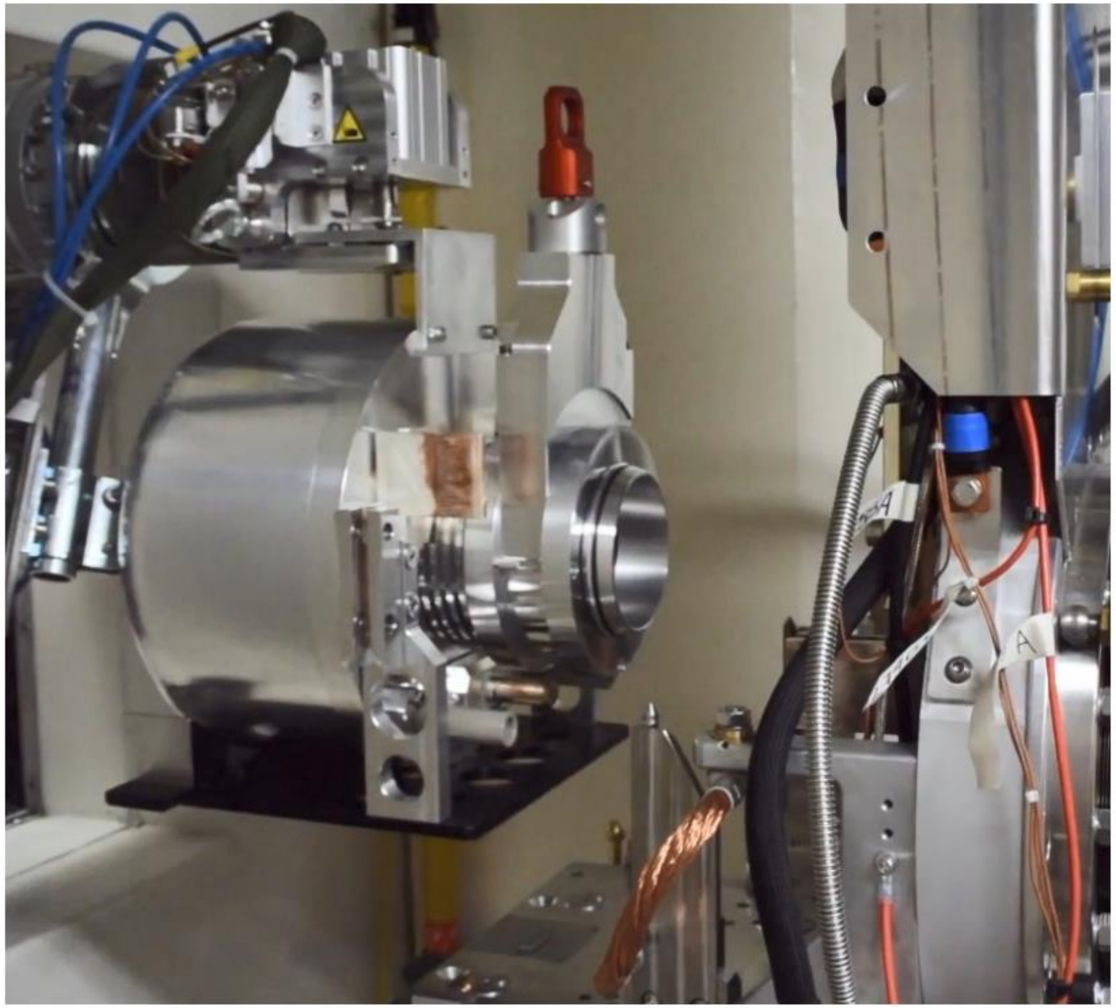


Plot proposed by S. Rothe

ISOLDE target and ion source unit

Principle design



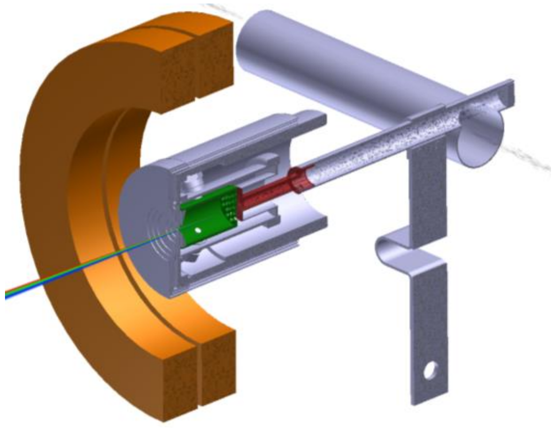


Ion sources at ISOLDE

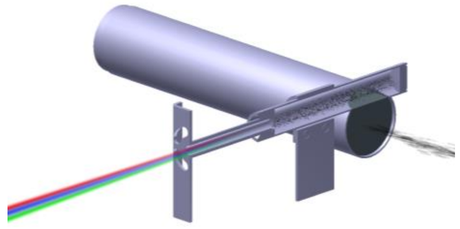
Ion source requirements

1. **Compact and radiation hard**
   - Integrated in the target unit
   - handling by robot required
2. **Withstand pulsed gas loads**
   - pulsed primary beam
3. **Efficient**
   - production of radio isotopes is limited
4. **Rapid**
   - low residence and ionization times
   - decay of radio isotopes
5. **Chemical selectivity (desired)**
   - No (or less efficient) ionization of radiogenic impurities

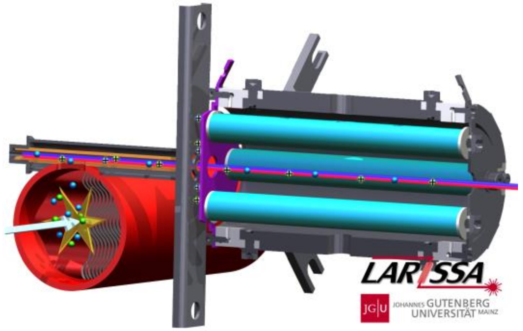
Frequently used ion sources (1+)



VADIS
electron impact ionization
thermionic electron emission



hot cavity ionizer (with RILIS)
surface or resonant laser ionization

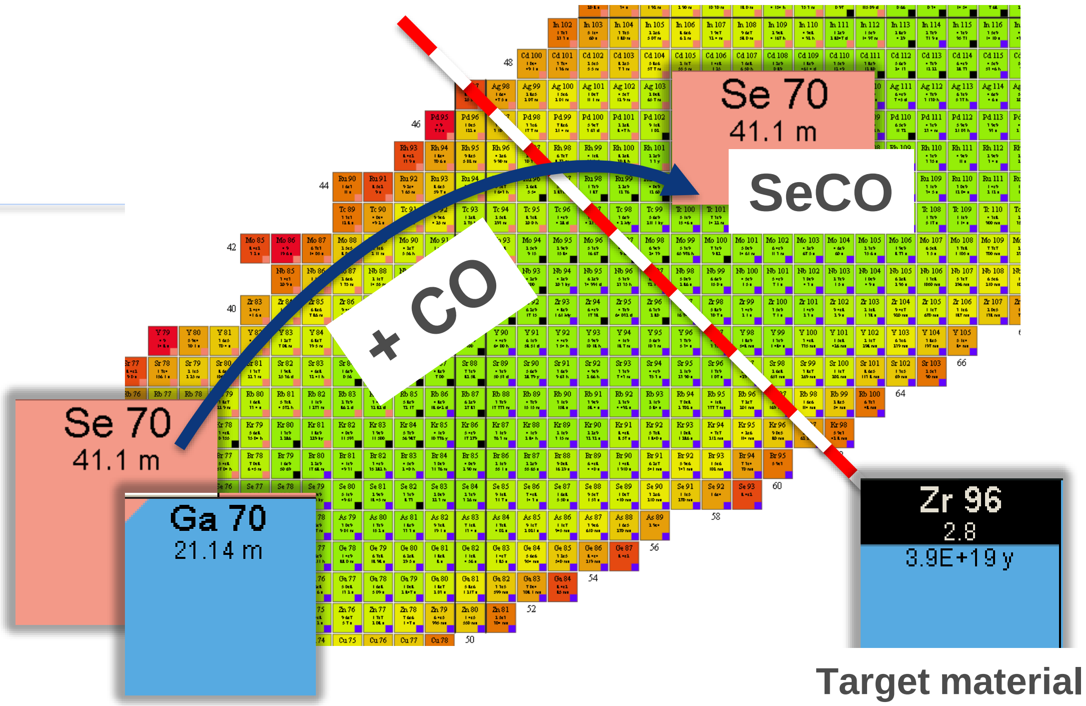


LIST ion source:
Hot cavity, repeller, RFQ ion guide

graphics by Y. Martinez-Palenzuela

Molecular beams

Molecular ISOL beams



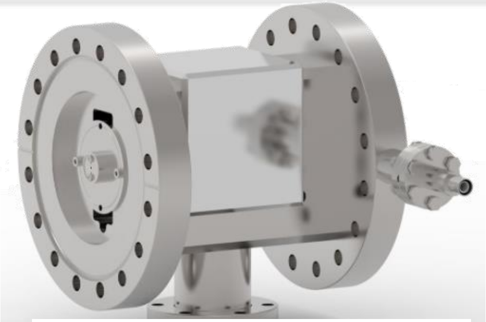
Target material

- **Beam purification**
  - Shift the mass region to a higher mass to avoid isobaric contaminants. e.g. ^{66}GeS, ^{133}SnS, ^{70}SeCO
- **Beam extraction by *In-situ* volatilization**
  - Elements with very low volatility are not released
  - Reactive elements can be chemically trapped e.g. $^{8}BF_2$, ^{15}CO
- **To study the radioactive molecules**
  - Fundamental properties, e.g. ^{223}RaF

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

Molecule formation in RFQ structures

- Ion traps for molecular formation/dissociation
  - Development for gas injection into RFQ for in-trap chemistry
  - Development of mass spectrometer and ToF detection for identification after RFQ
- In-source laser ionization of molecules



DREEBIT *Wien filter*

Mia Au

Offline 2 RFQ

M. Fan et al, *Optical mass spectrometry of cold RaOH+ and RaOCH3+*, PRL **126**, 23002 **(**2021)

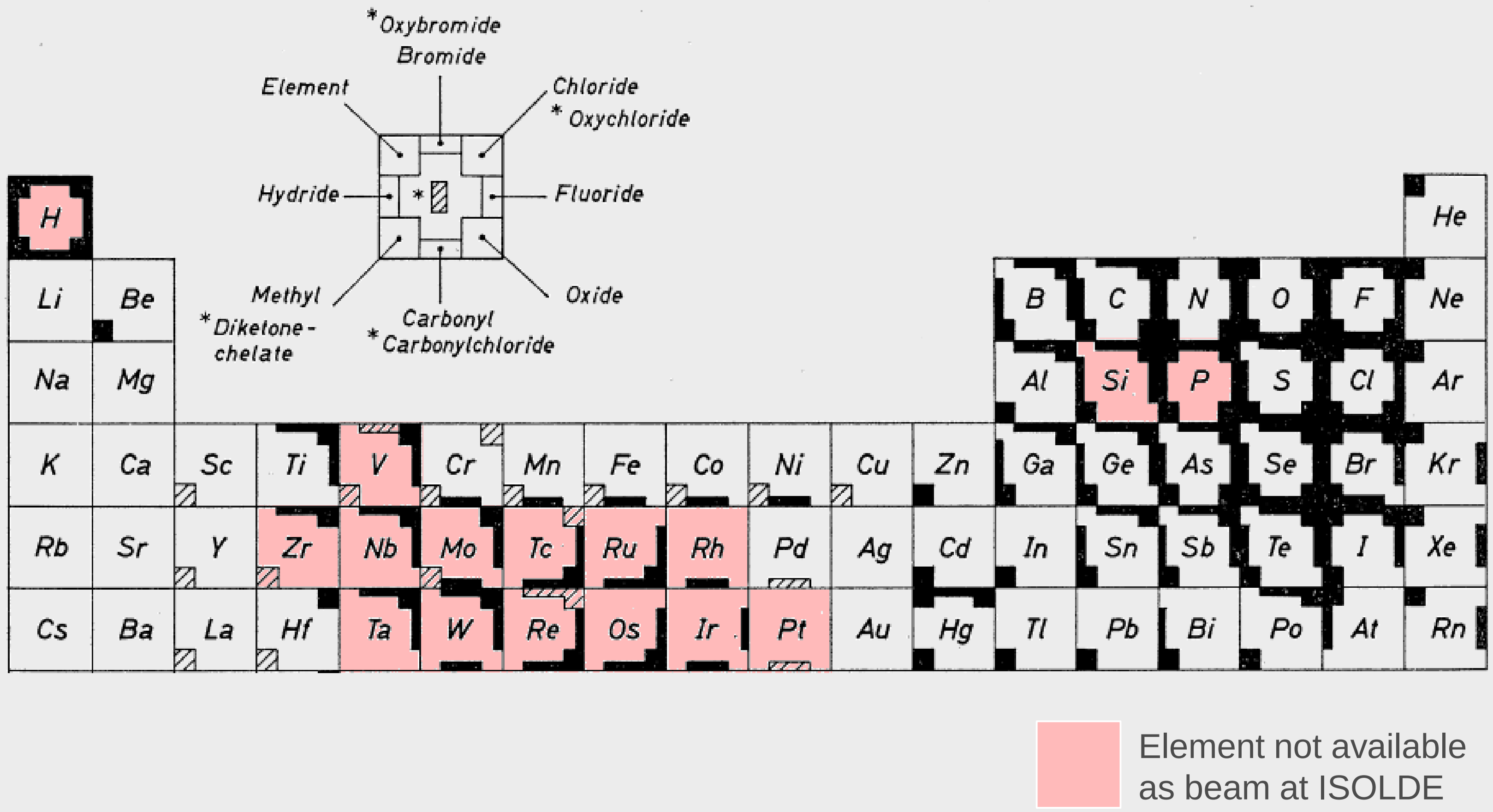
A. Ringvall Moberg et al, *Time-of-Flight study of molecular beams extracted from the ISOLDE RFQ cooler and buncher* NIMB **463**, 522 (2020)

Volatile carrier molecules

- **Volatile carriers known for all reactive elements**
- **Many are not compatible with high-temperature conditions**

U. Köster, (Im-)possible ISOL beams, Eur. Phys. J. Special Topics ***150****, 285 (2007)*

Volatile carrier compounds



G. Herrmann, Ark. Fys. ***36****, 111 (1967)*

Do we have to operate at the highest temperatures?

Cold targets?

- Nano-sctructured target material (CaO) operated at ambient temperature
  - fast diffusion in nano-materials and possible release by recoil effect

Target nanomaterials at CERN-ISOLDE: synthesis and release data

J.P. Ramos [a,b,*], A. Gottberg [a,1], R.S. Augusto [a,c], T.M. Mendonca [a], K. Riisager [d], C. Seiffert [a,e], P. Bowen [b], A.M.R. Senos [f], T. Stora [a,*]

*J.P. Ramos et al, NIMB **376**, 81 (2016)*

- Foil target at ambient temperature, thermalizing recoils in gas atmosphere
  - no diffusion required

A concept for the extraction of the most refractory elements at CERN-ISOLDE as carbonyl complex ions

J. Ballof[1,2 a], K. Chrysalidis[1], Ch.E. Düllmann[2,3,4], V. Fedosseev[1], E. Granados[1], D. Leimbach[1,5], B.A. Marsh[1], J.P. Ramos[1 b], A. Ringvall-Moberg[1,7], S. Rothe[1], T. Stora[1 c], S.G. Wilkins[1,8], and A. Yakushev[3,4]

J. Ballof et al., preprint, https://arxiv.org/abs/2108.01745

Cold ion sources?

- Cold RF-heated plasma ion sources at ISOLDE:
  - COMIC / Helicon *A. Kronenberg et al, NIMB 266, 19 (2008)*
  - For delicate molecules: favour breakup over ionization

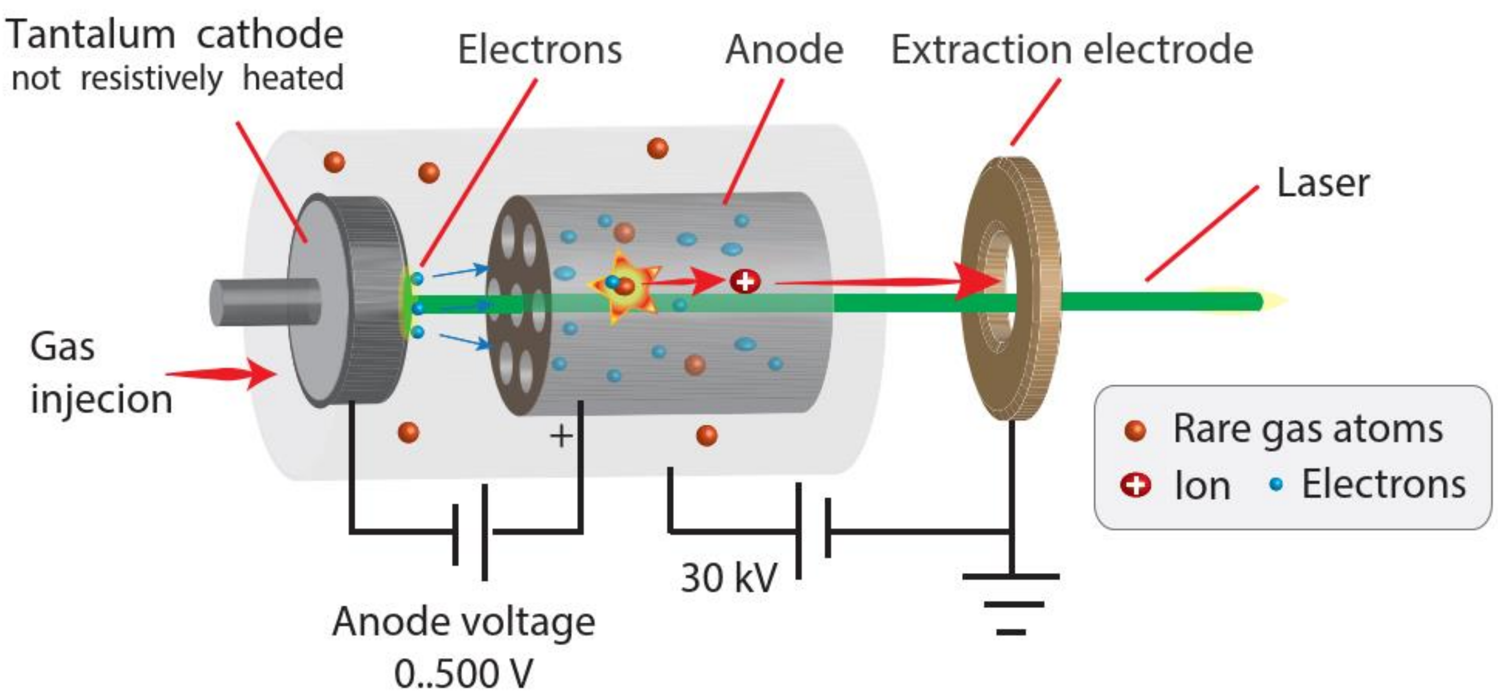
Ionization chamber of Helicon plasma source after operation with $Mo(CO)_6$

- Ion source development required

The photo-cathode electron-impact source

Decoupling electron-emission and temperature

Proof-of-principle setup 1: Ta cathode

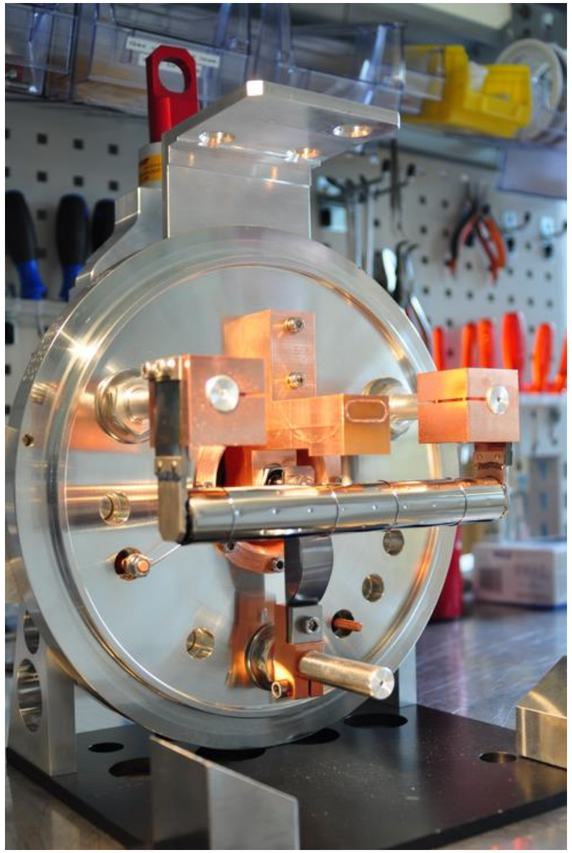


Laser	
Pulse length	265 fs
Power	~ 4.5 W
Wavelength	343 nm
Rep. Rate	50 kHz
Beam diameter	~ 6 mm
Beam spot on cathode	1.5 mm

Cathode	
Material	tantalum
Anode-cathode gap	1.5 mm

Proof-of-principle setup 1: Ta cathode

Target assembly



Before operation / during assembly

Anode assembly

Ta-cathode

Extraction side

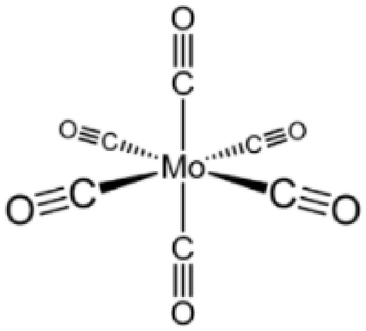
After operation

Laser-induced craters on the cathode surface during focusing attempts

Traces from laser-alignment on the extraction side

Proof-of-principle setup 1: Ta cathode

Fragile carrier compound $Mo(CO)_6$



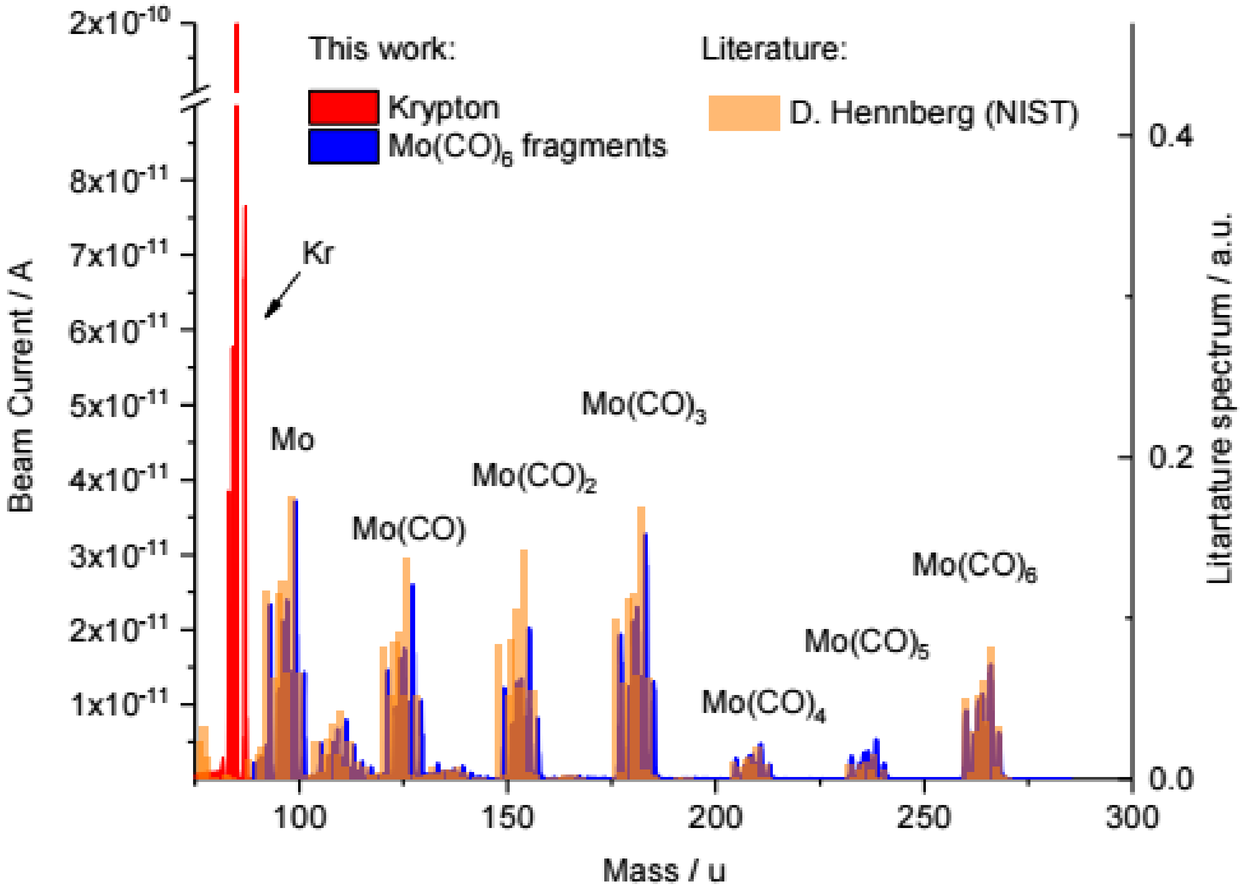
FBDE: 1.7 eV
IP : 8.3 eV

Vapour pressure:
25 Pa (ambient temp.)

Measured efficiencies

Nuclide	Current	Efficiency
^{98}Mo	33 pA	1.5×10^{-5}
^{84}Kr	200 pA	2.2×10^{-5}

Measured mass spectrum



Proof-of-principle setup 1: Ta cathode

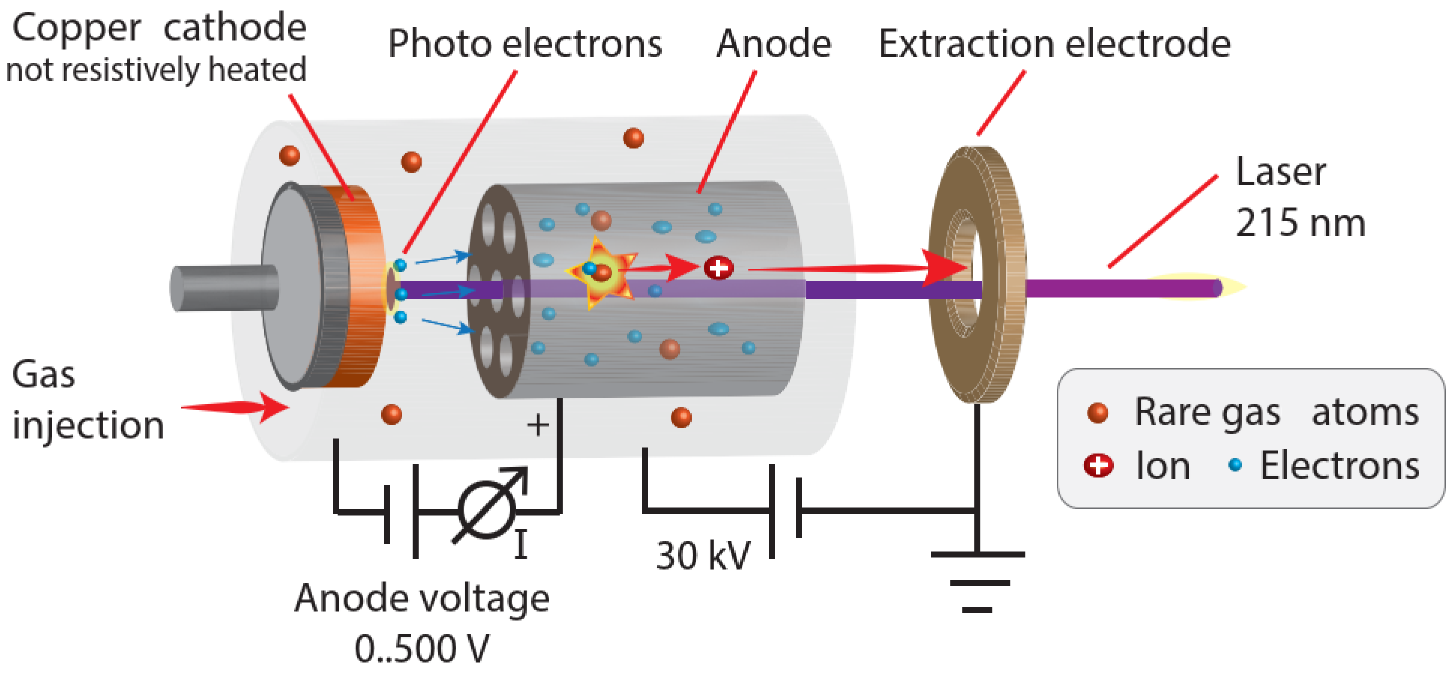
- **Results from first operation**
  - Fragile compound $Mo(CO)_6$ could be ionized in the setup
  - Efficiency of Mo(CO)6 is in the same order of magnitude as Kr
  - Mechanism of electron production unclear: Photo-electric effect or thermionic emission?
  - Efficiency is very likely limited by electron current:

    ~ 100 nA instead of ~ 100 mA with a hot VADIS

- **Further focusing attempts**
  - focusing the beam diameter from ~ 6 mm to below 1.5 mm caused cathode damage

Proof-of-principle setup 2: Cu cathode

- **Setup optimized for photo-electric effect**



Laser	
Pulse length	50 ns
Power	~ 0.01 W
Wavelength	215 nm
Rep. Rate	10 kHz
Beam diameter	~ 1.5 mm

Cathode	
Material	copper
Anode-cathode gap	~ 3 mm

Proof-of-principle setup 2: Cu cathode

Target assembly

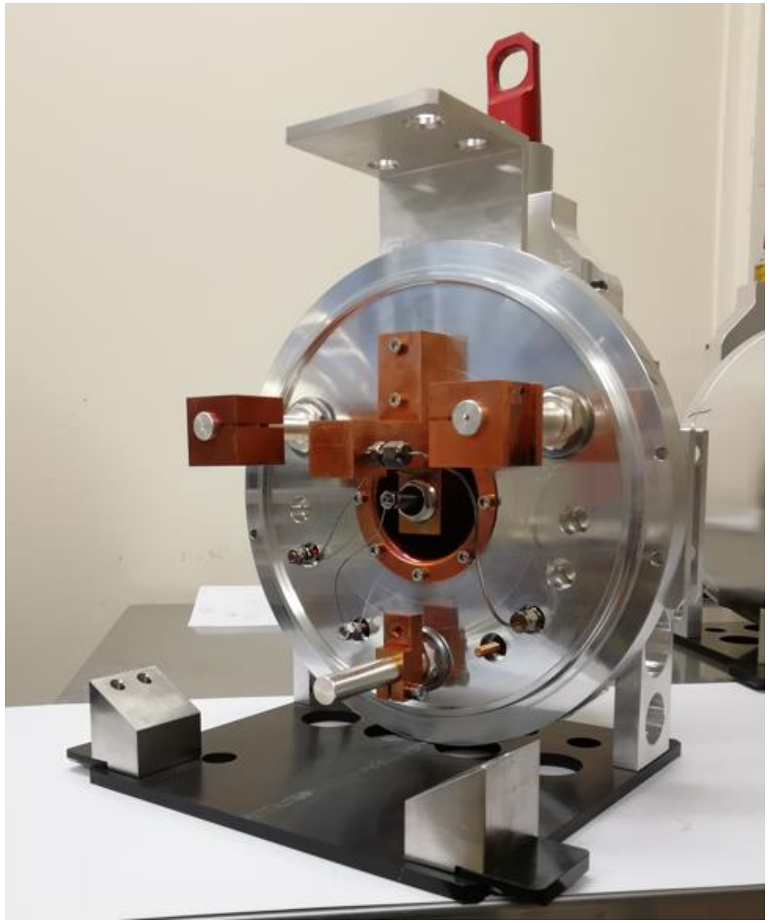
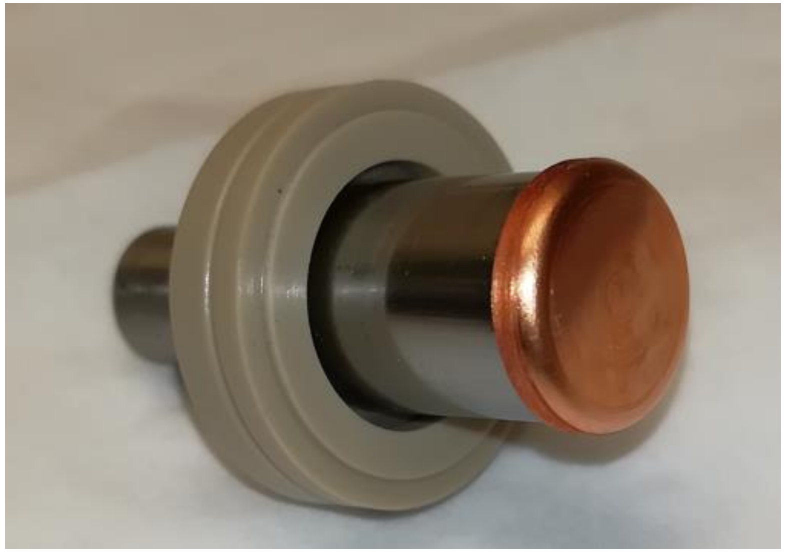
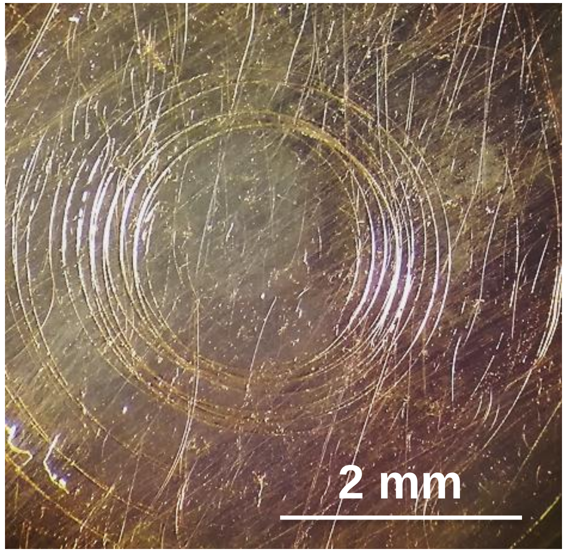


Photo-cathode after operation

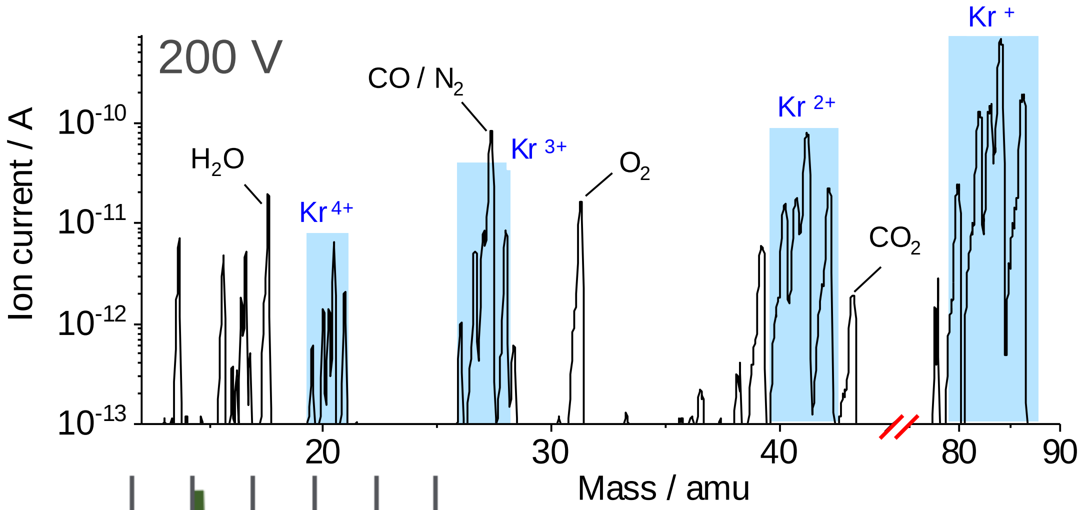


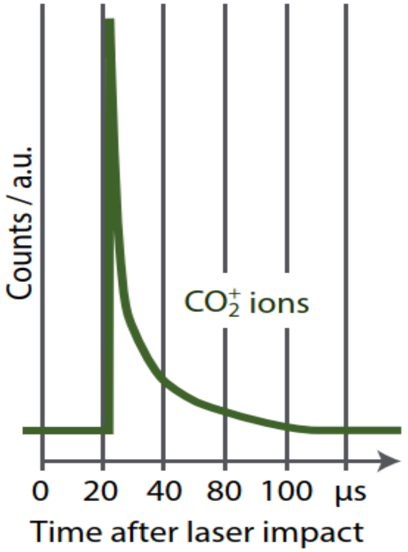


- **Cap made of oxygen free copper foil (OFHC)**
- **PEEK insulator**
- **No visible difference before and after operation**
- **No polishing, nor special cleaning or bake out**

Proof-of-principle setup 2: Cu cathode

- **Results from Cu-cathode operation**
  - Source operated for 6 days (24/7)
  - **Kr** and **CO_2** can be ionized with a photo-cathode electron-impact ion source
  - No electron emission with 440 mW of blue light (430 nm) confirms photo-electric effect
  - Source magnet increased efficiency by factor ~8
  - Estimated quantum efficiency: 3 x 10^{-4}
  - **Decomposition of CO_2 degrades the photo-cathode**.
    - Estimated decrease of 65% in 66 hours, pressure ca. 1 x 10^{-4} mbar
    - Typical hot VADIS: 1 x 10^{-6} mbar (injected gas)





Injected gas	Ionization efficiency
Kr	4 x 10^{-5}
CO_2	2 x 10^{-5}

How to increase the efficiency?

- Increase of electron current required
- First approximation: ionization efficiency increases proportionally with electron current

Parameter	VADIS Thermionic emission	Photo-cathode prototype
Electron current	~ 100 mA	~ 0.1 to 1 μA
Ionization efficiency Kr	30 %	0.004 %
Diameter electron emitter	1.2 cm	0.15 cm

Ionization efficiency

L. Penescu *et al*, RSI 81, 02A906

$$\varepsilon \propto n_e \times \sigma \times f$$

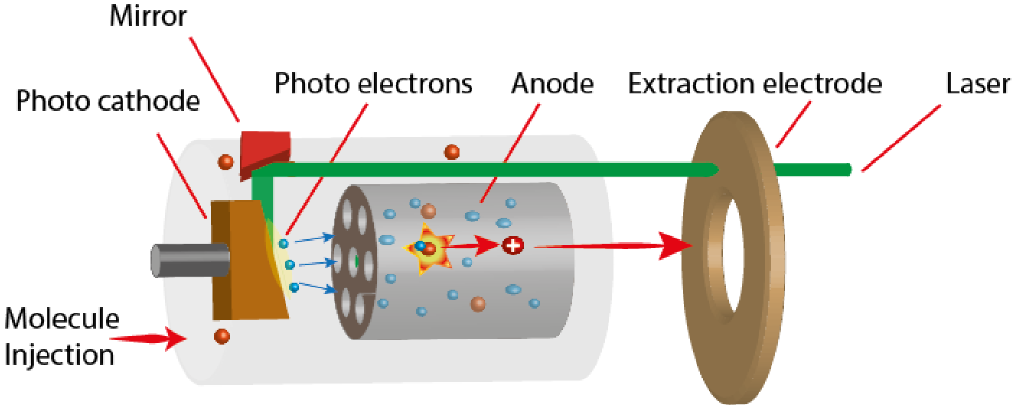
electron density

ionization cross section

- ion extraction
- electron focusing
- other effects

Next Prototype?

- Proposal for a future prototype: perpendicular illumination
  - Resolves space charge limitations
  - Beam path developed for the PI-LIST ion source
    R. Heinke *et al*, Hyperfine Interactions **238**, (2017)
- Efficiency estimate by scaling of space-charge limited electron currents



Estimated parameters to reach 1% ionization efficiency:

Parameter	Next prototype	Tested prototype
Electron emitter diameter	12 mm	1.5 mm
Anode-cathode distance	3 mm	1.5 mm
Repetition rate	2 MHz	50 kHz
Wavelength	257 nm	343 nm
Power	3.7 W	4.5 W
Mean electron current	90 µA	90 nA
Ionization efficiency $Mo(CO)_6$	1%	0.001%

J. Ballof et al., preprint, https://arxiv.org/abs/2108.01745

Conclusion

- Electron-impact ion source can be driven by a photo-cathode
- Photo-cathode can cope with typical ISOL conditions and gas-loads (~ 10^{-6} mbar)
- Ionization efficiency (proof-of-concept experiment) 0.001% for $Mo(CO)_6$
- Upgrading laser-system and source geometry could yield efficiencies of at least 1%

Impact on molecular beams

- Decoupling of electron production from ion source temperature
- First ISOL ion source for delicate molecules
- Could facilitate refractory beam extraction and extract molecules for fundamental physics research

Thank you for
your attention!