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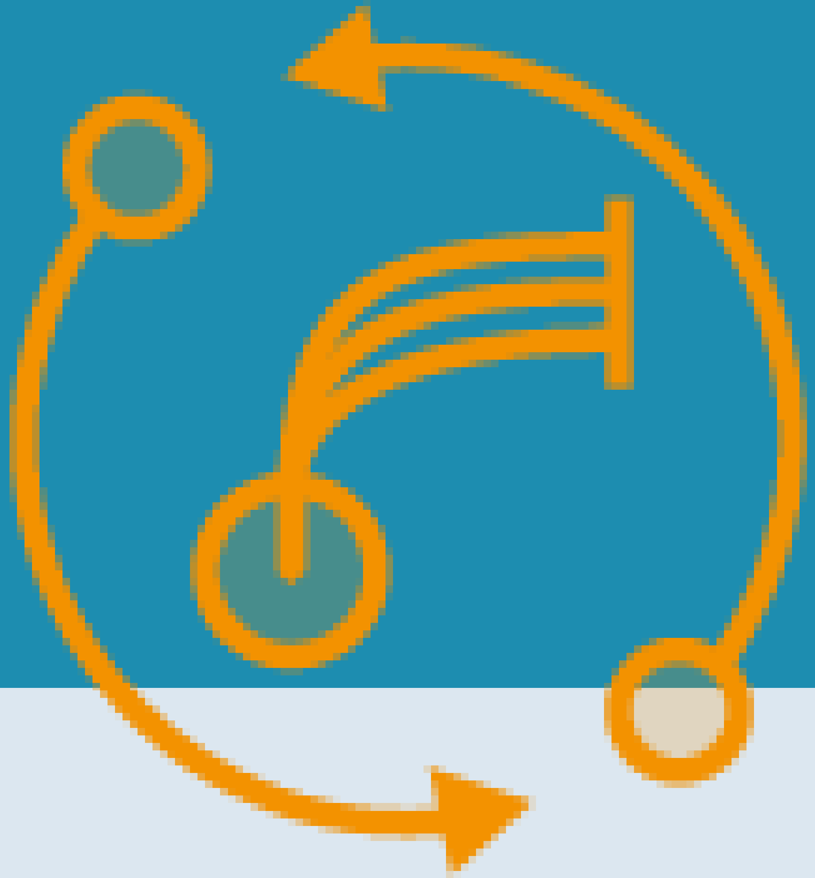
State of the art in the isotope separation online technique



Chair Roger Van Geen

Lecture 2 – 8 November 2021

Radioisotope production

Luminosity

- More primary beam
- More target

$$\frac{dn}{dt} = +L\sigma$$

Cross section

- What beam
- On which target
- At what energy

What about purity!?

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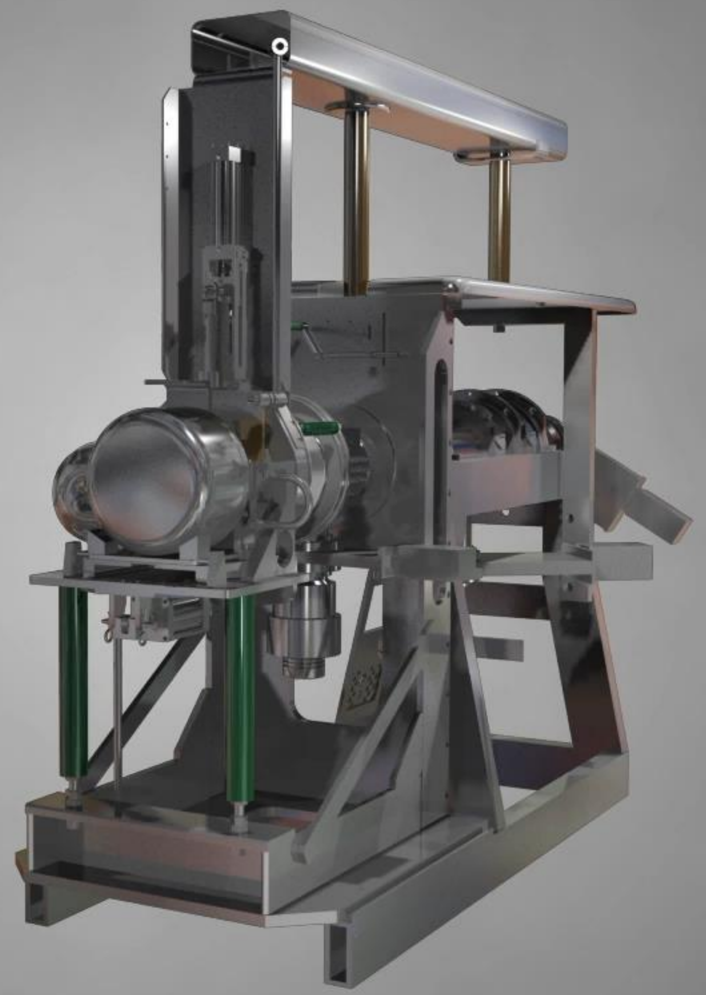

Video animation available here: https://videos.cern.ch/record/2289929

Outline of this lecture

- Target materials
  - Basic concepts
  - Innovative concepts
- Ion sources
  - Ultra pure beams
  - High intensity beams
- Beam manipulation
- Beam post-acceleration

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ISOL Targets

Beyond maximizing luminosity

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Isotope release

Diffusion:

$D = D_0 \cdot e^{-\frac{E}{RT}}$ (Arrhenius eq.)

$\varepsilon_{diff} = \frac{3}{\pi}\sqrt{\frac{\mu}{\lambda}}$ $\mu = \frac{\pi^2 \cdot D}{G^2}$

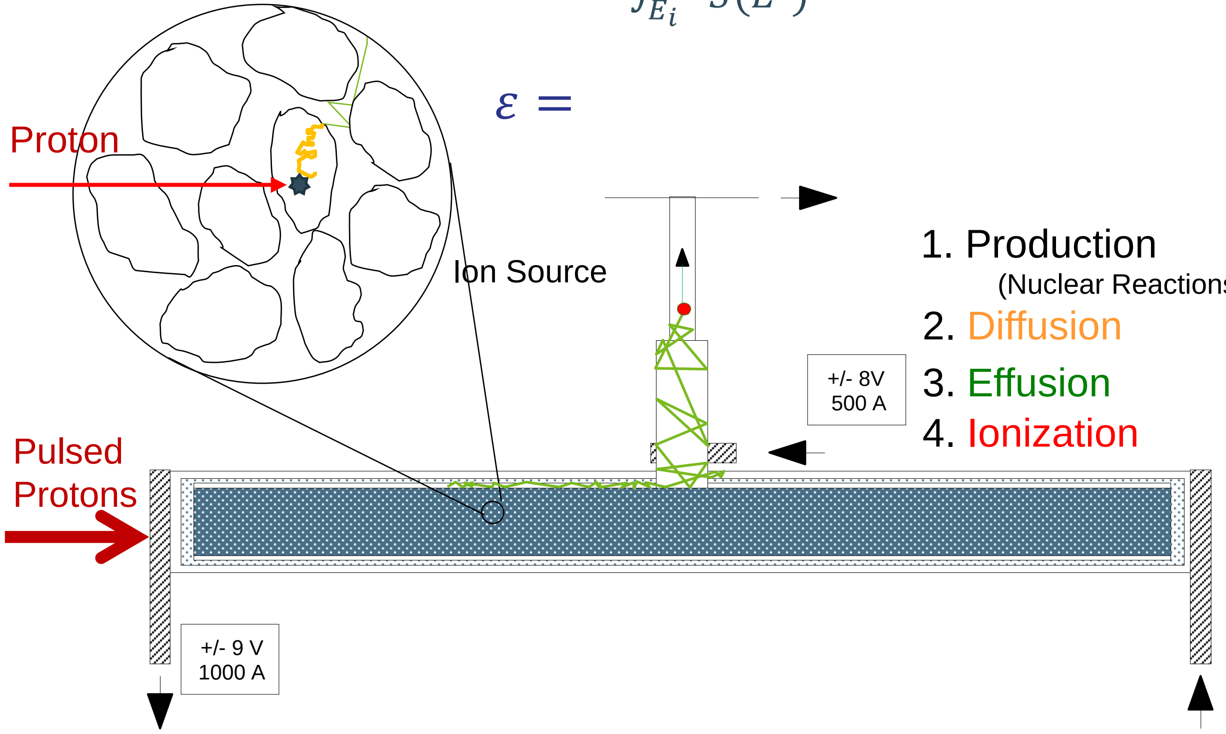
$\Rightarrow \varepsilon_{diff} \propto \frac{1}{G}$

D: Diffusion coefficient
μ : Diffusion time
λ : Decay constant
G: Grain size

Control microstructure to enhance release properties

$Y = I \cdot N_t \cdot \int_{E_i}^{E_f} \frac{\sigma(E')}{S(E')} \cdot dE' \cdot \varepsilon$

$\varepsilon =$



1. Production
(Nuclear Reactions)
2. Diffusion
3. Effusion
4. Ionization

 Slide courtesy of J.P. Ramos, EMIS Conference Sept 2018

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Metalic targets

- The simplest form of a target material is a pure metal:
  - ➢It can reach high density
  - ➢The reaction channels are simple: one beam on one target
  - ➢It is easy to handle

- There are challenges:
  - ? How do you release the recoils from the target material?
  - ? ... while keeping the integrity of the material!

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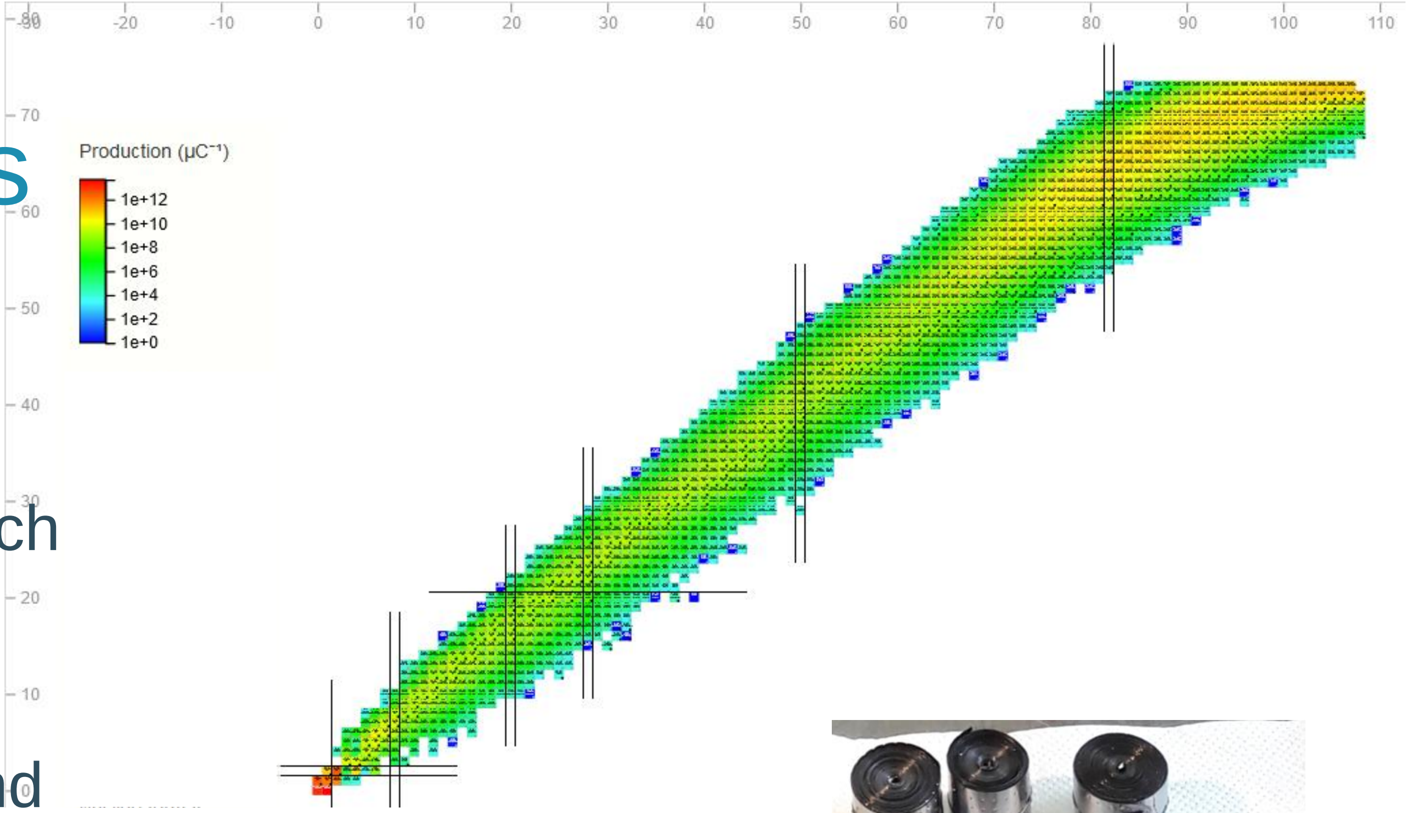
Typical metalic targets



Rolled Ta foils

- ^{180}Ta is a heavy material from which many isotopes can be produced.
- As a refractory material, it can sustain very high temperatures and can operate at >2000°C as typical from ISOL facilities.
- 20 μm thick foils are rolled to allow for efficient release while maintaining the luminosity

https://isolde.cern

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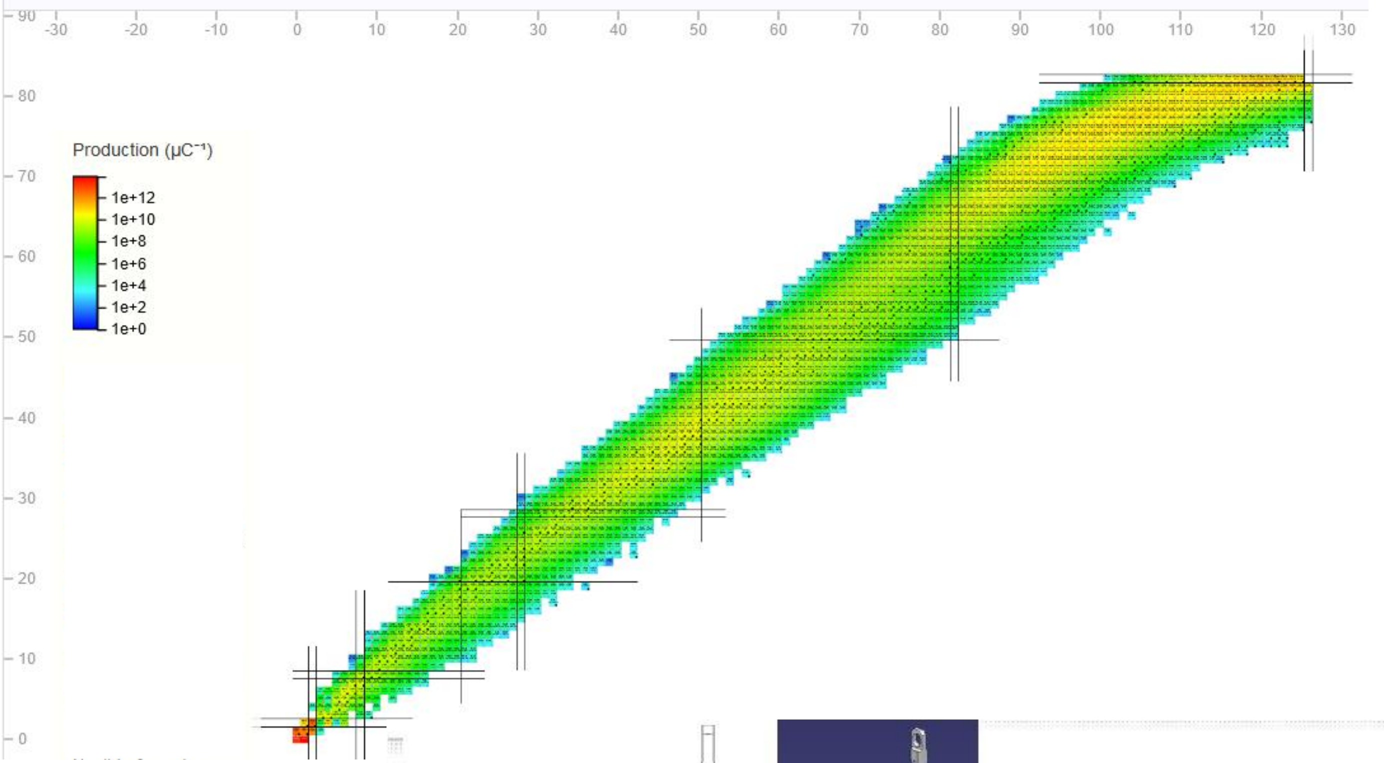
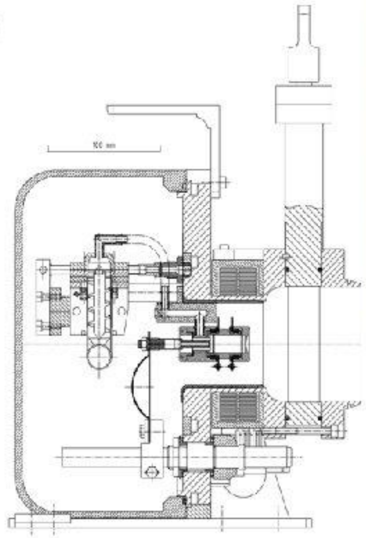
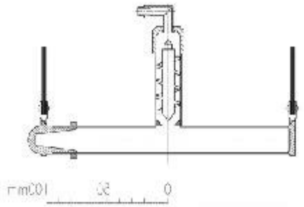
Exotic metalic targets



Molten lead target

- ^{208}Pb is also a heavy element from which a lot of radioisotopes can be produced.
- However, as a soft metal, it cannot sustain the high operating temperatures.
- Instead of taking any risk, you operate directly with a molten target appropriate for the high temperature!





Pb/Bi264 container
after dismantling in 2007

4

https://isolde.cern
Slide courtesy of Thierry Stora

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Overview metalic targets at ISOLDE

Metal foils

- Titanium
- Tantalum

Molten metals

- Tin
- Lanthanum
- Lead

How can we get thinner materials to release faster?

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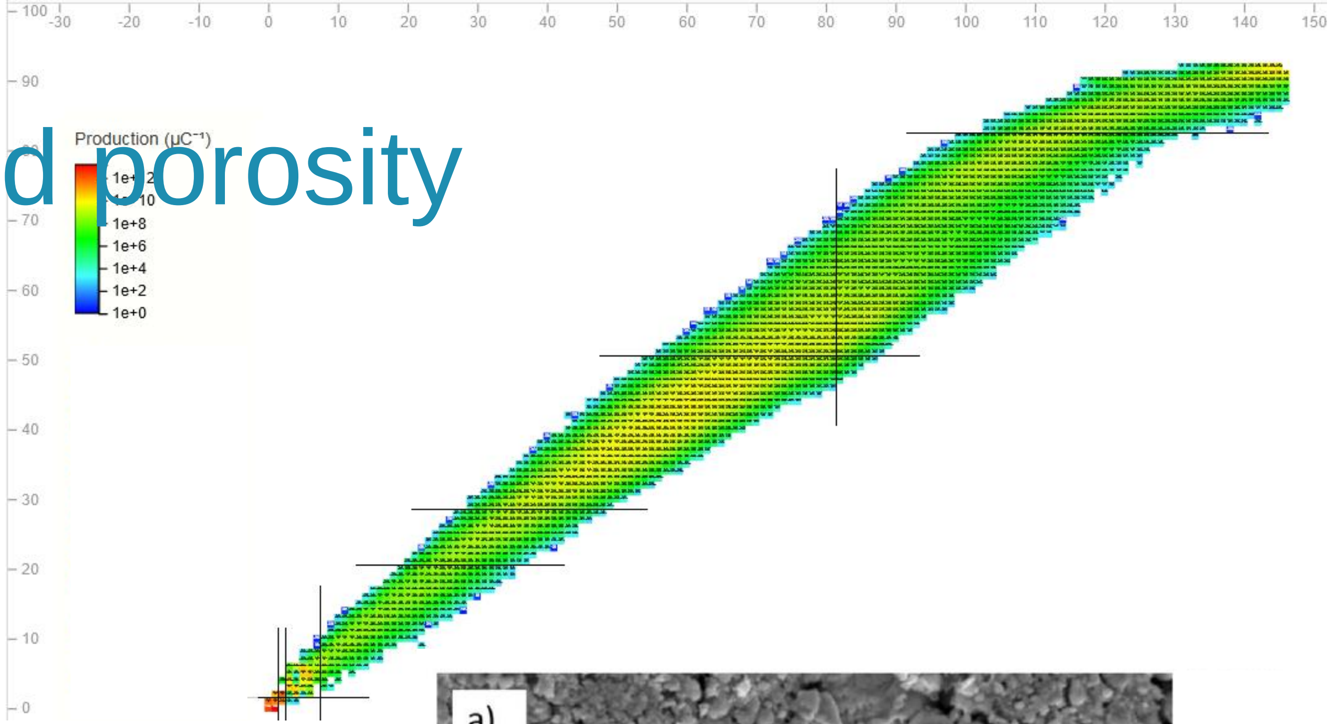
Ceramics with controled porosity



Uranium carbide

- ^{238}U is the most neutron-rich heavy isotope that can be found in large quantities on earth.
  - $N/Z = 146/92 = 1.59$
- ^{238}U is also fissile and can produce isotopes with the same N/Z, e.g., $^{129\text{-}130}$Sn.
- Depleted uranium is actually enriched in ^{238}U!

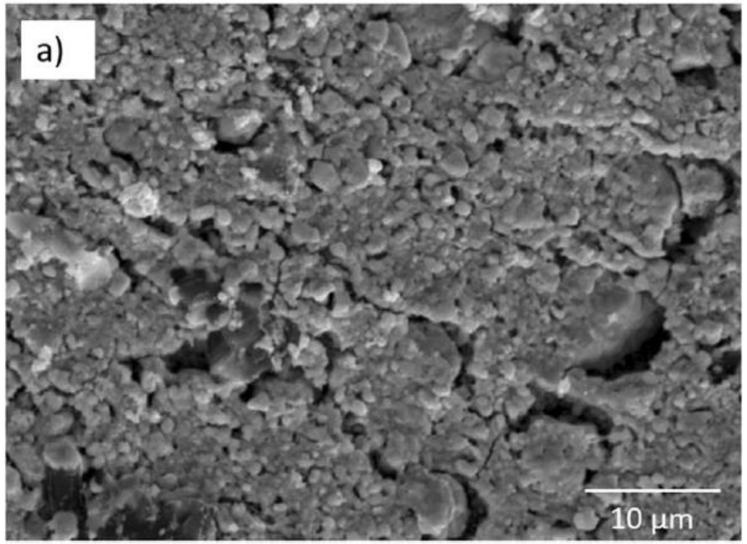


Figure 2. SEM images of the cross-section of UC_x-graphite (**a**)

https://isolde.cern
*L. Biasetto et al., ... synthesis of uranium carbide for isotope production targets, Scientific Reports **8** (2018) 8272.*

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Uranium carbide: tough chemistry

- Uranium is not stable in carbide form:
  - Supply is typically as uranium oxide.
  - Carburation is performed in the laboratory (whether at ISOLDE or TRIUMF).
  - The target material must then be kept under Ar atmosphere to prevent oxidation!
- The exact form of uranium carbide is poorly known:
  - Depends on the C concentration and the caburation and operation temperatures
  - Assumed as UC_x, with $1 \leq x \leq 2$

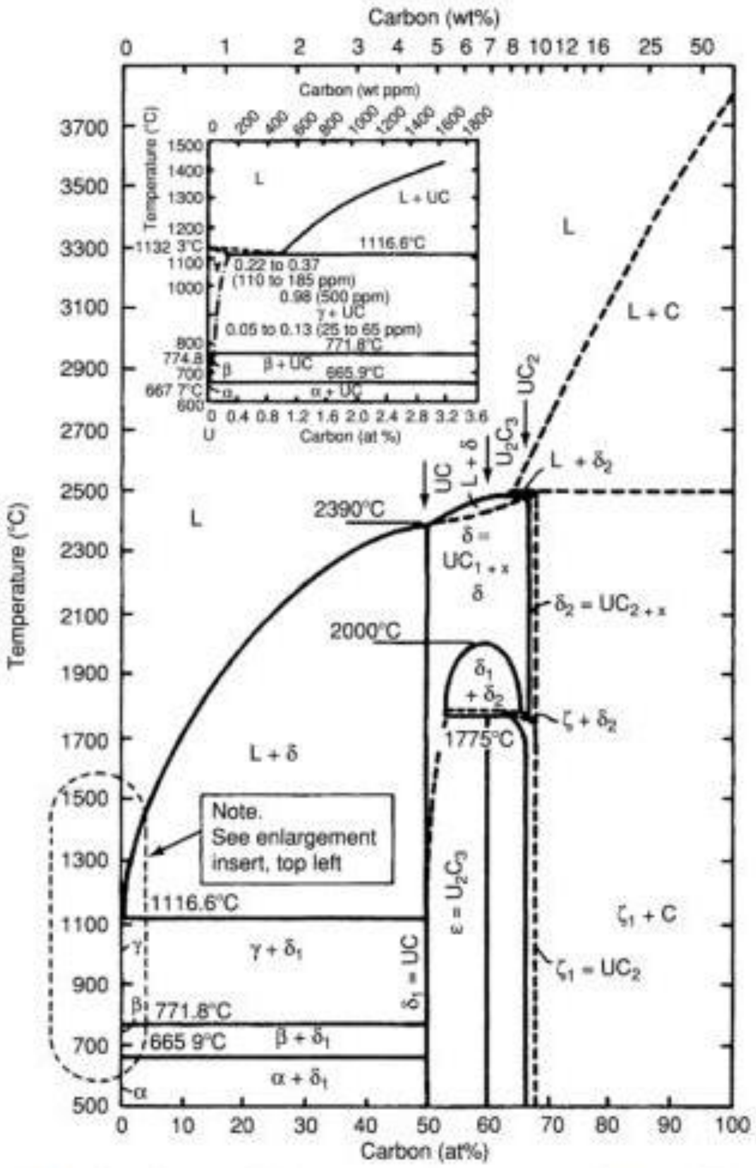

Fig. 5.29 *Phase diagram of the uranium–carbon system (Wilkinson, 1962).*

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The limits of uranium carbide

- Other elements form very stable carbides:
  - ❖None of the rare-earth elements can be easily extracted from a UC_x target as they form LnC_x compounds themselves!
  - ➔ Use metalic Ta targets.
- Oxygen and carbon always come out as CO^+ beams.
- The grain size depends on the starting material – typically µm-sized grains – and of the sintering of the target during preparation and operation
  - ➢Start with nanometric material!

$$\boxed{\varepsilon_{diff} \propto \frac{1}{G}}$$

  - ➢Work at low temperatures!

$$\boxed{D = D_0 \cdot e^{-\frac{E}{RT}}}$$

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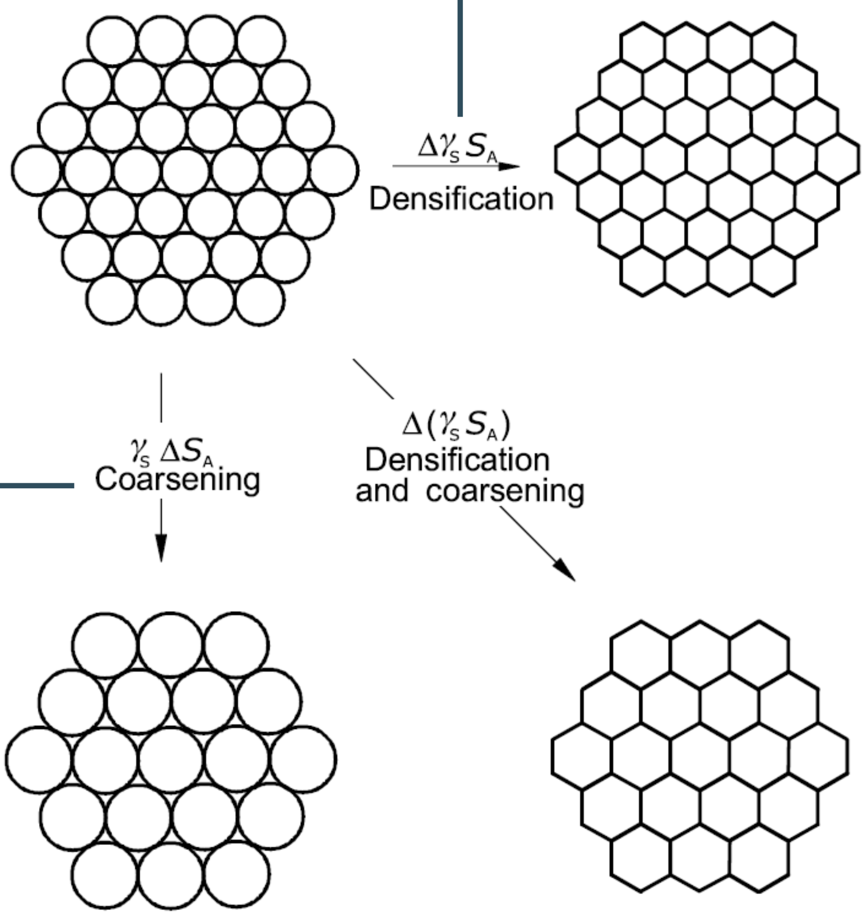
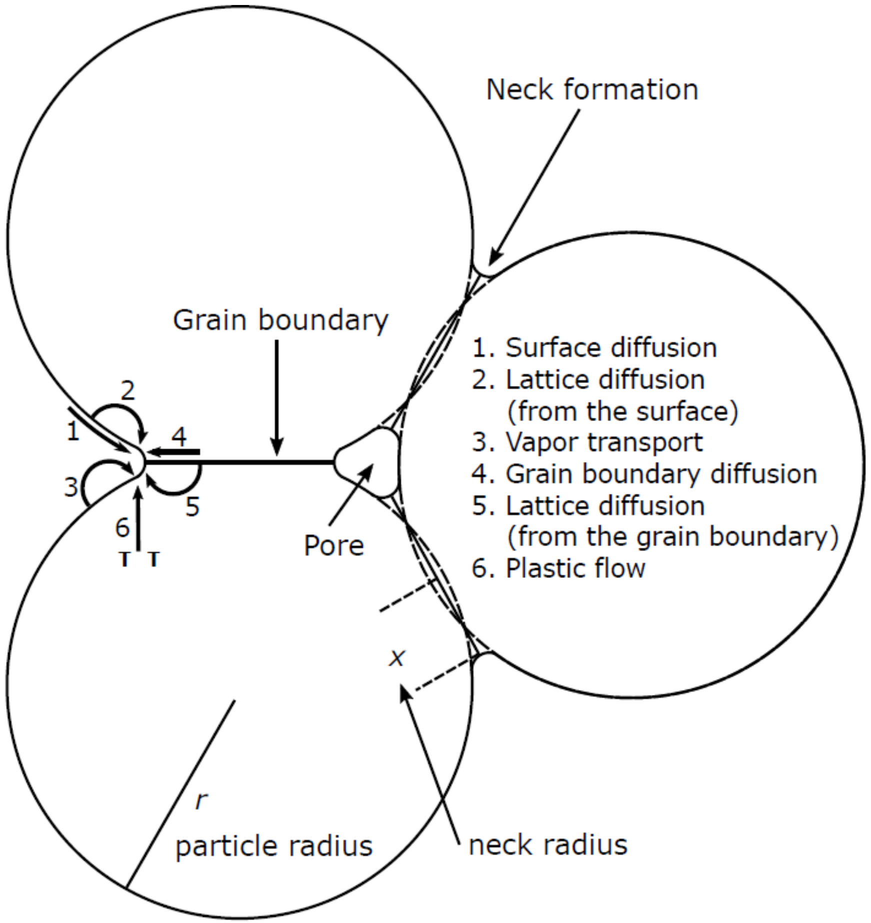
Sintering

Pore closure → Poorer effusion

Grain growth → Poorer diffusion





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Designing the best uranium carbide target

Enhanced release

- Start from nanomaterial to keep diffusion fast
- Run at high temperature to keep fast diffusion
- Inject reactive gases like CF_4 or O_2 to promote the release of simple molecules

Operation limits

- Don't work with nanomaterial to minimize sintering
- Run at low temperature to minimize sintering
- Run in as pure a vacuum as possible to prevent the degradation of the UC_x material

How can we reconcile those two considerations?

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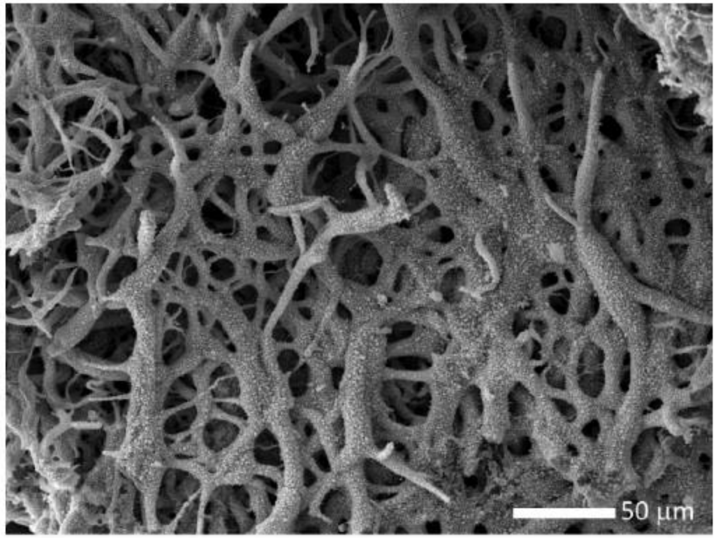
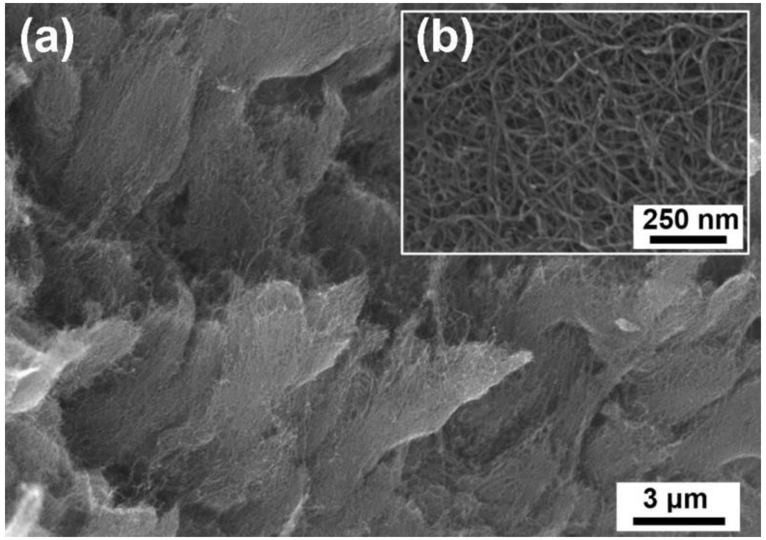
Stabilizing the UC_x material

Advanced nanotechnology

- Embedding the UC_x nanomaterial in a nanotube matrix to prevent sintering.
- Processing uranium-nanofibres.





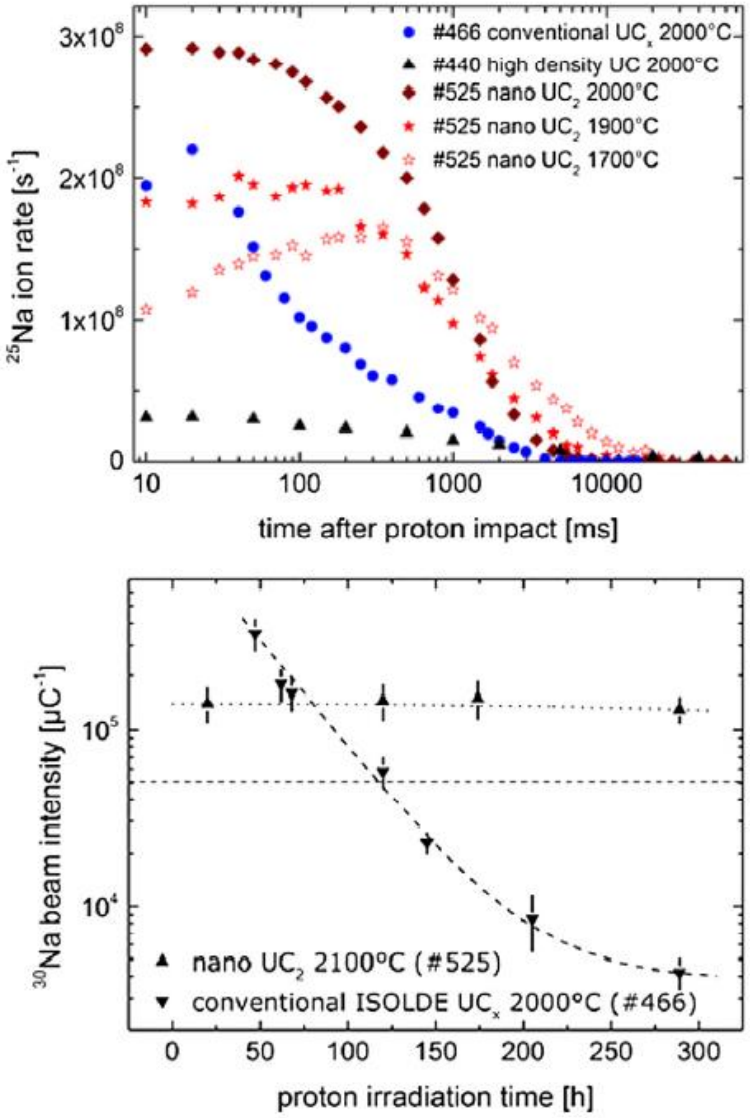


Fig. 3. Release time structure of three different UC_x target materials at different operation temperatures (top). Comparison of long-term release stability under high-energy proton irradiation and high temperature (bottom).

A. Gottberg, Target materials for exotic ISOL beams, NIMB ***376*** *(2016) 8-15.*
J.P. Ramos et al., Target nanomaterials at CERN ISOLDE..., NIMB ***376*** *(2016) 81-85*
S. Chowdhury et al., Uranium carbide fibers ... for ISOL targets, Nanomaterials ***10*** *(2020) 2458*

Overview of the ceramic targets at ISOLDE

✓UC_x is everybody's favorite, as it produces almost the entire nuclear chart and it can yield very neutron-rich nuclei.

✓LaC_x is a target that can yield very intense beam around ^{100}Sn.

✓SiC is a target suited for the production of light beams.

- But you may also use oxides!
  - ➢ThO_2 is an alternative to UC_x, especially for the production of oxide sidebands.
  - ➢CaO is also a very good target for light beams, which has yielded the most intense Ar^+ beams at ISOLDE and can be produced from nanograins.

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ISOL Targets

2018 operation

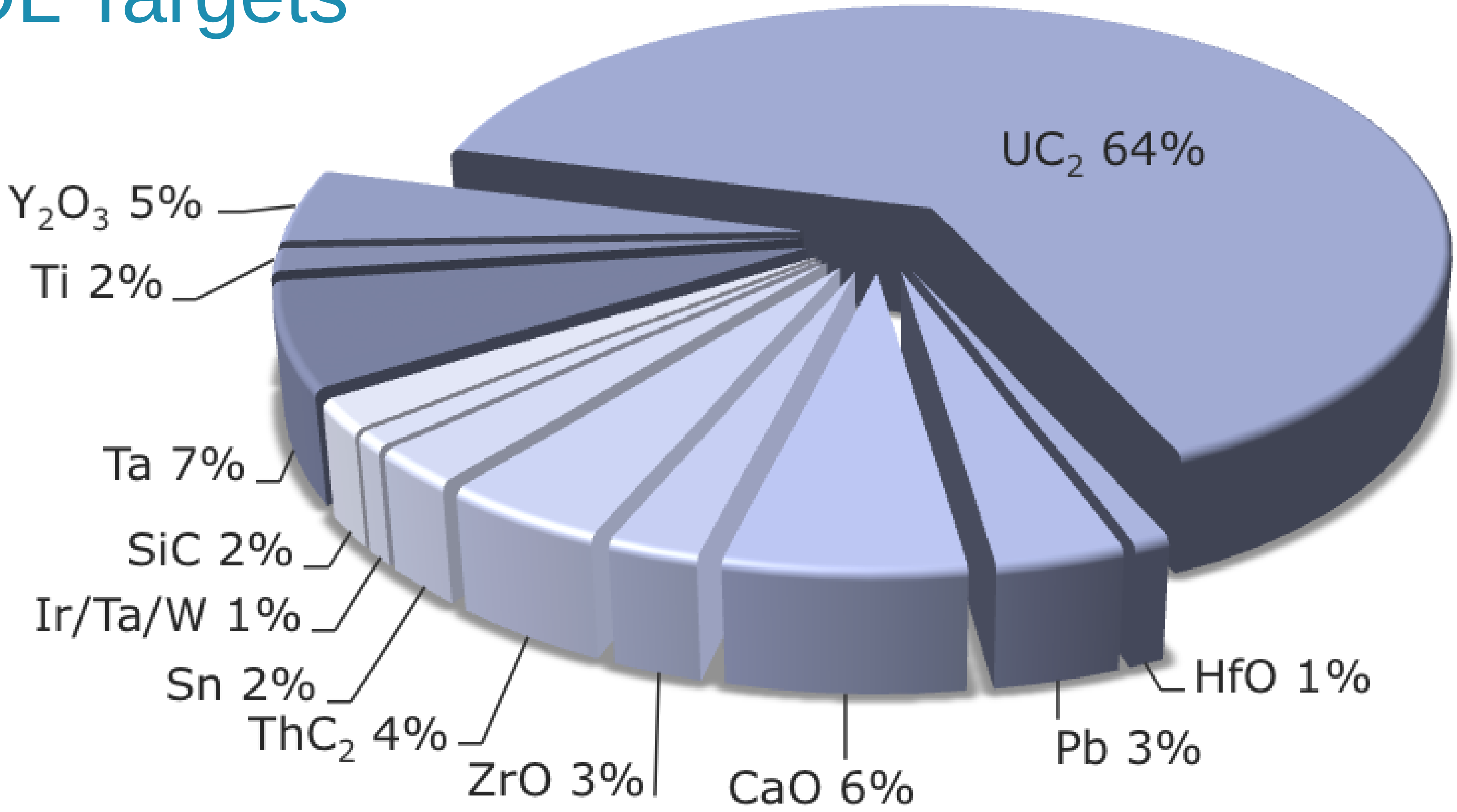

Figure courtesy of Karl Johnston, ISOLDE Scientific Coordinator.

Ion sources

Who let the BEAMS out!!!

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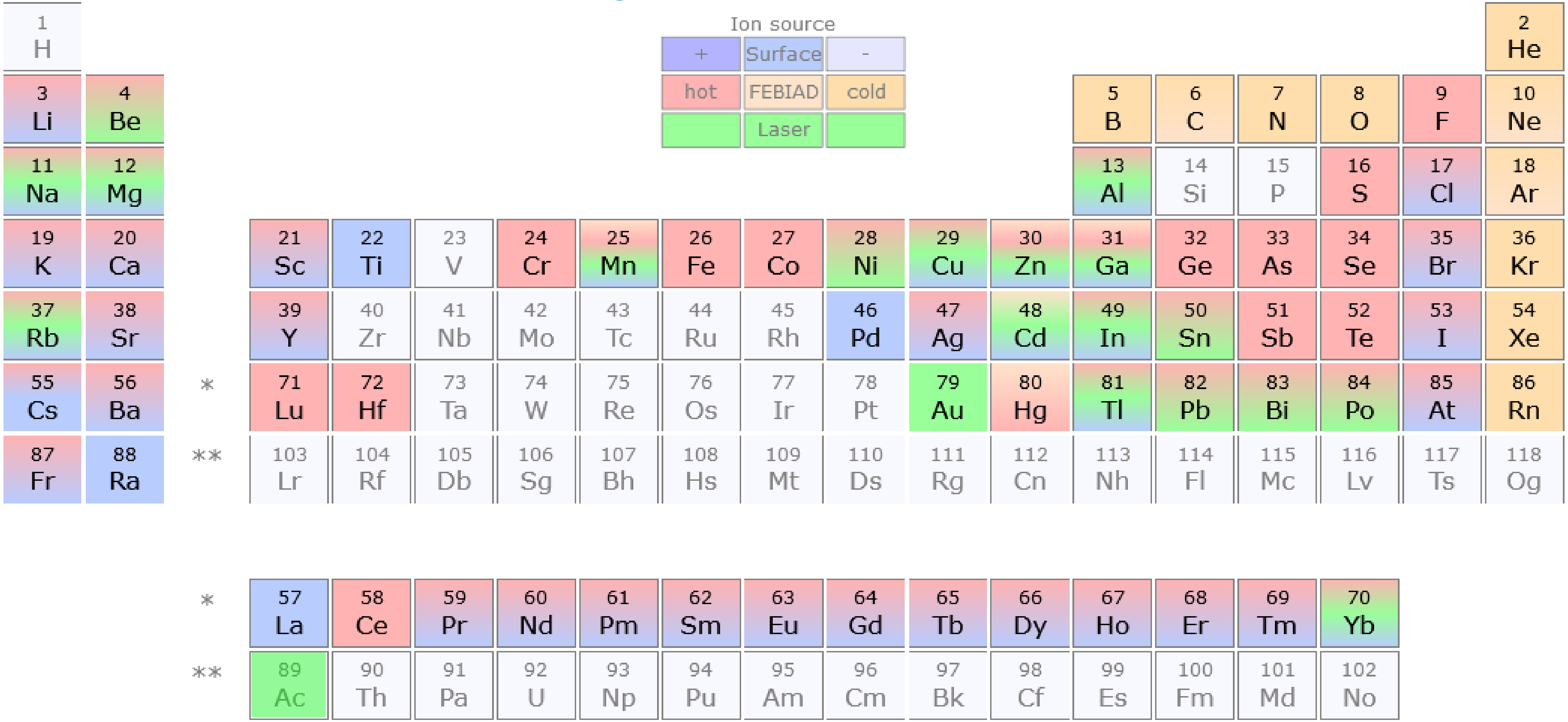
Back to ion sourcery



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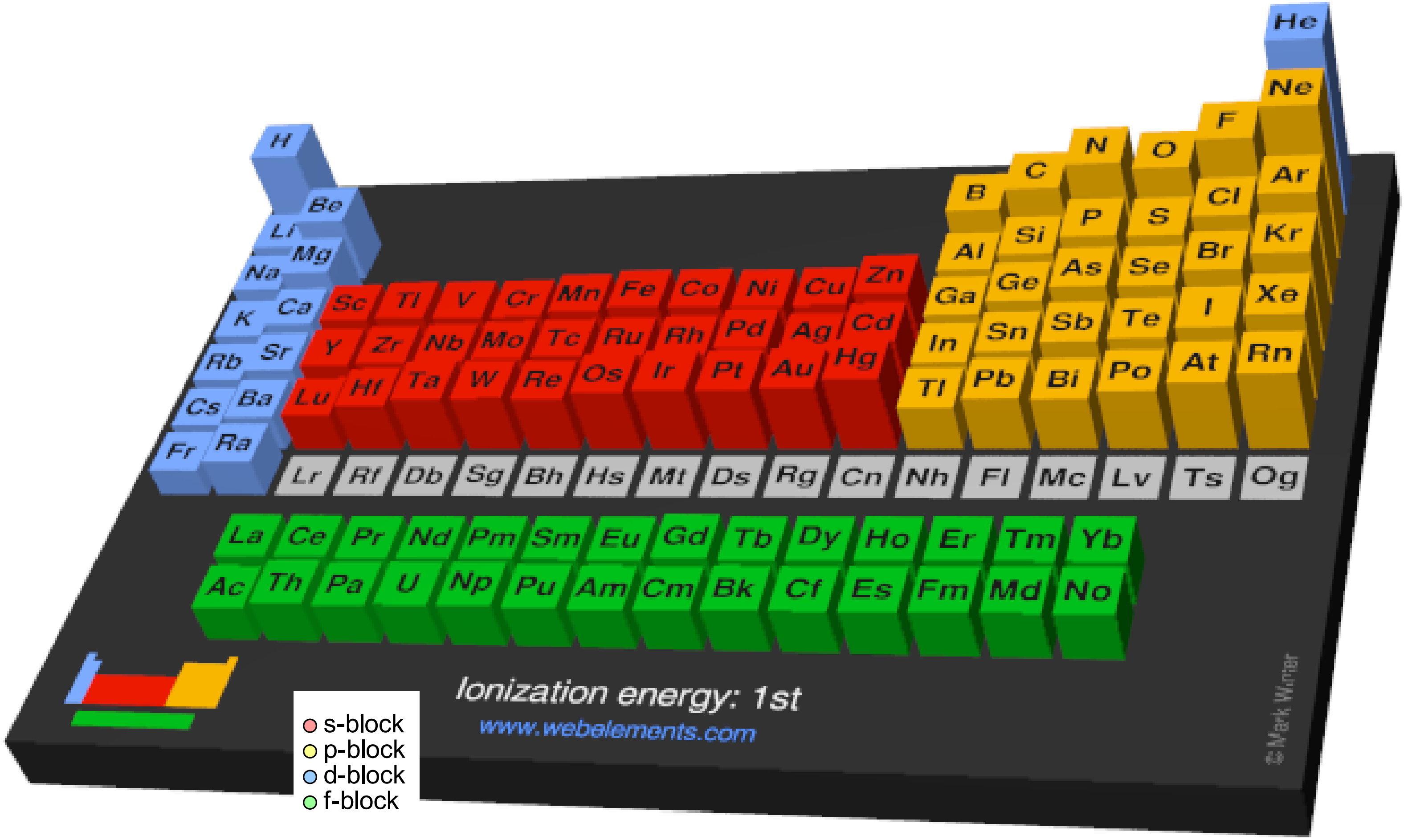
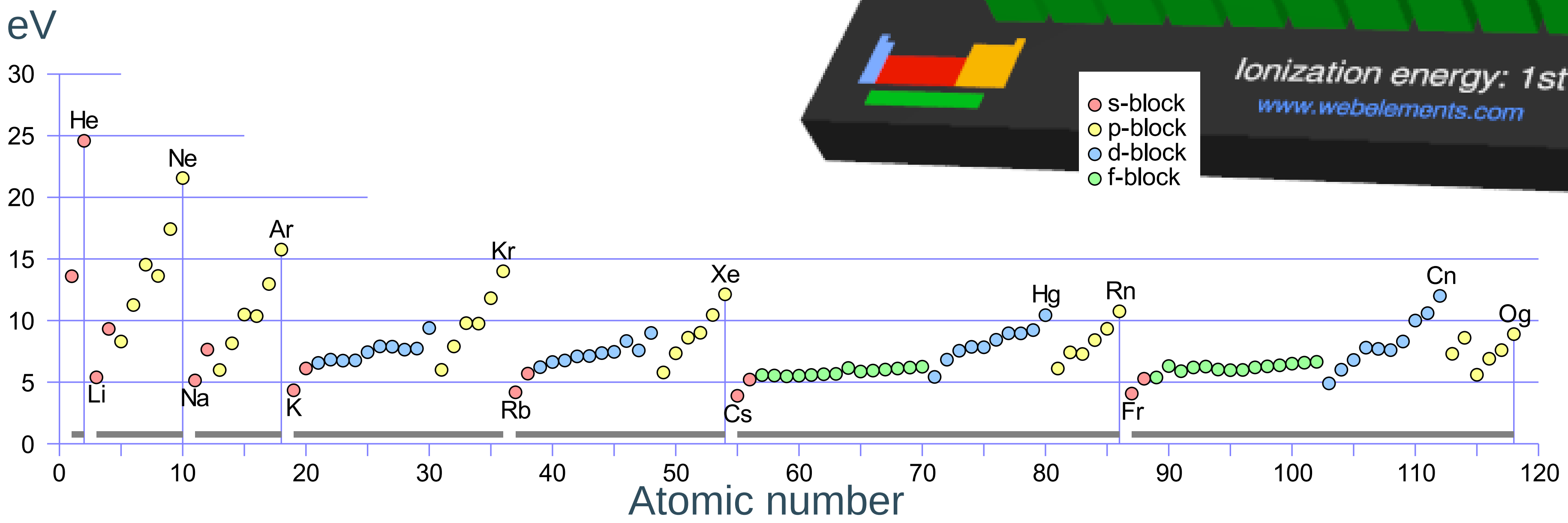
Ionization energy





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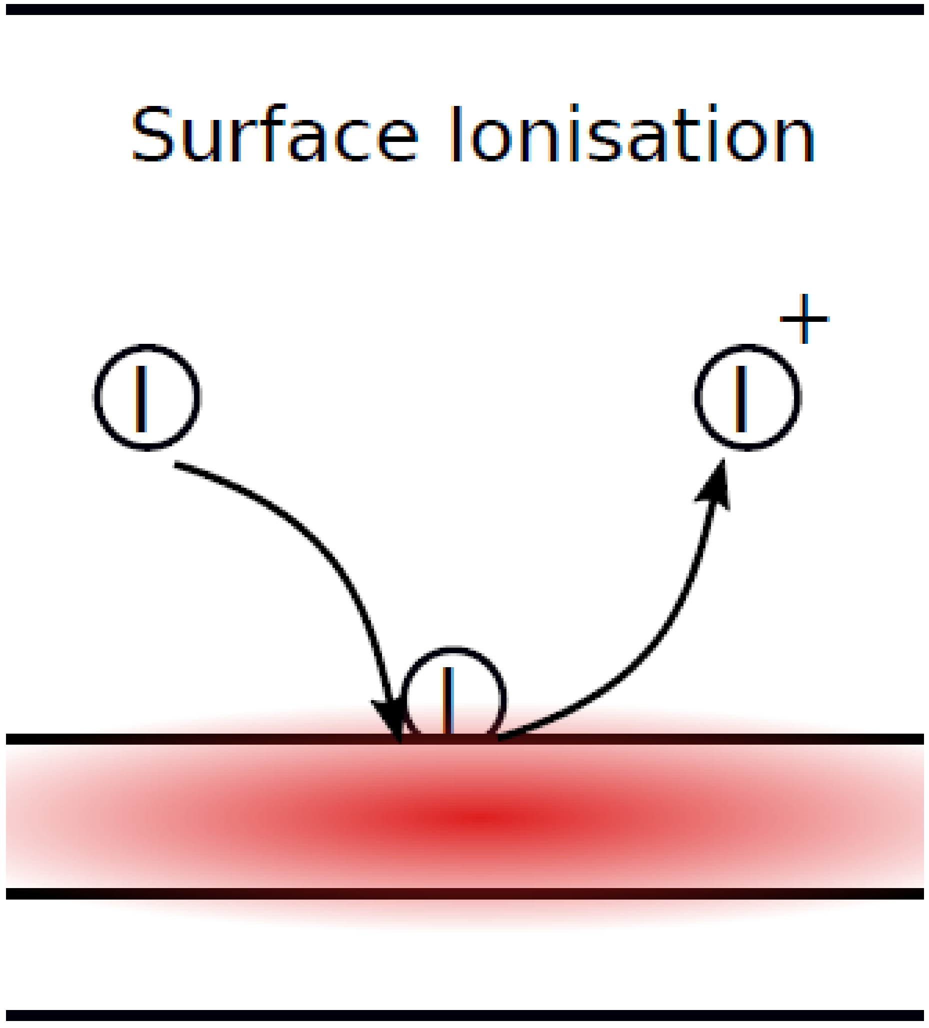
Surface ionization



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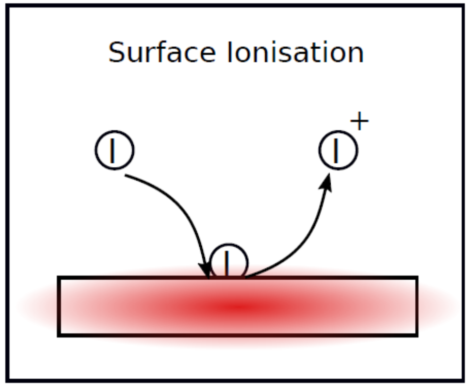
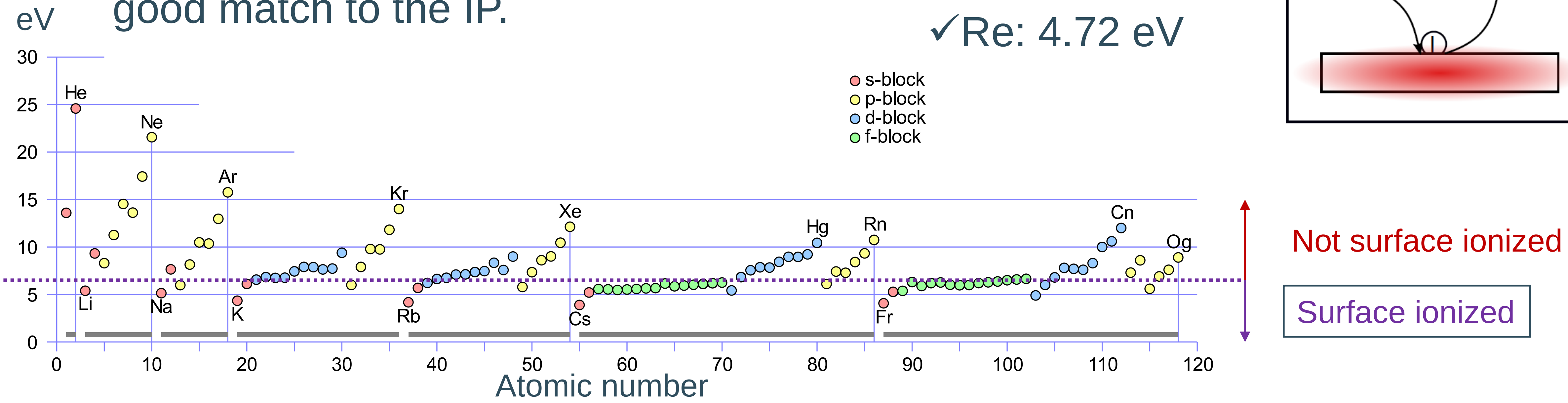
Surface ionization

- A loosely bound electron can be removed from an atom when coming in contact with a hot surface if the work function of the material is a good match to the IP.

- It must be a material that holds its integrity at the high operating temperature ≥ 2000°C
- Workfunctions of typical surface ionization materials:
  - ✓W: 5.22 eV
  - ✓Ta: 4.8 eV
  - ✓Re: 4.72 eV





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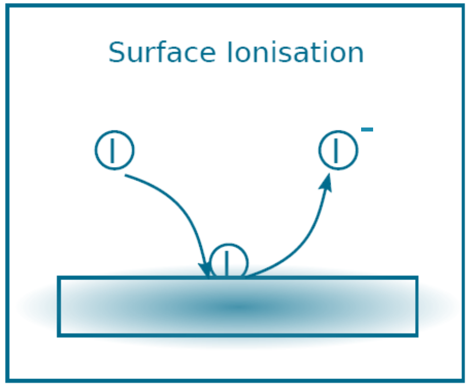
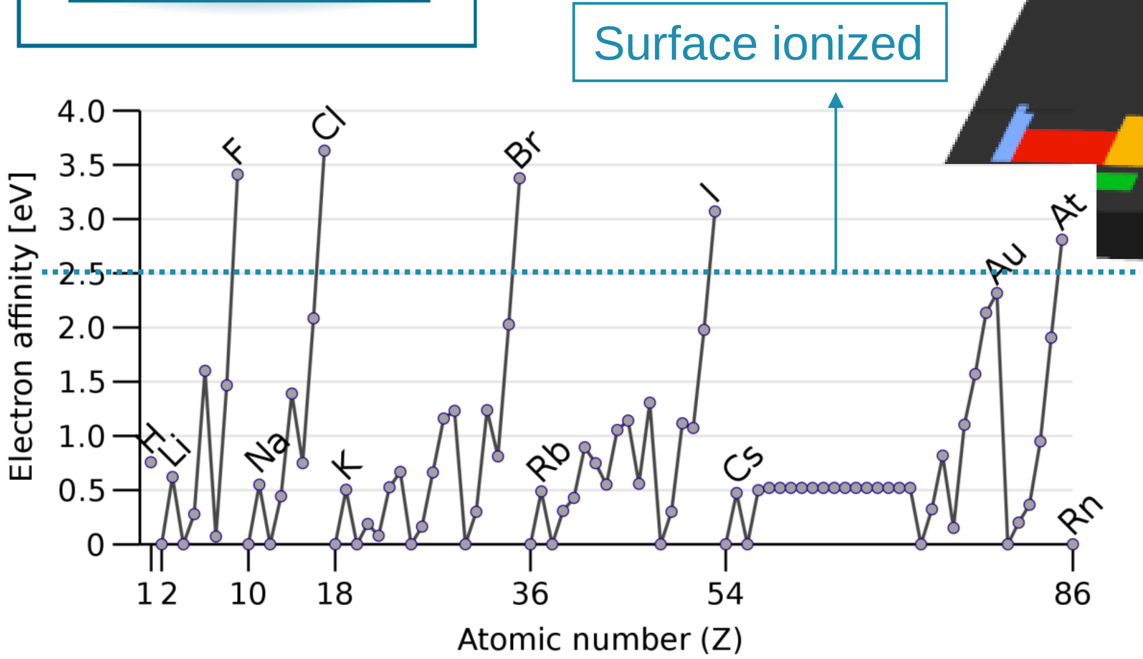
Negative surface ionization





➢Requires a surface that can easily give away an electron to an element that can easily receive it.

➢Typically only applies to halogenes.

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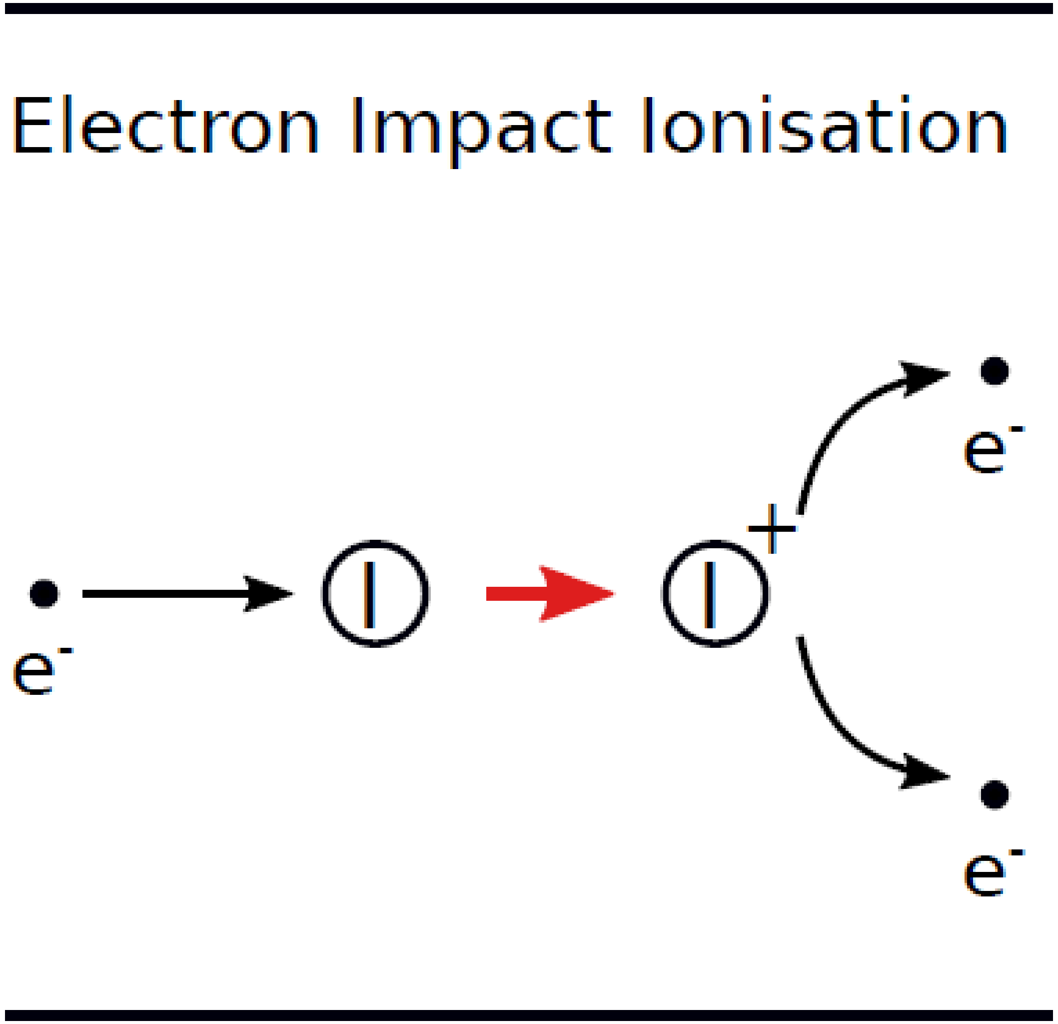
Electron impact ionization



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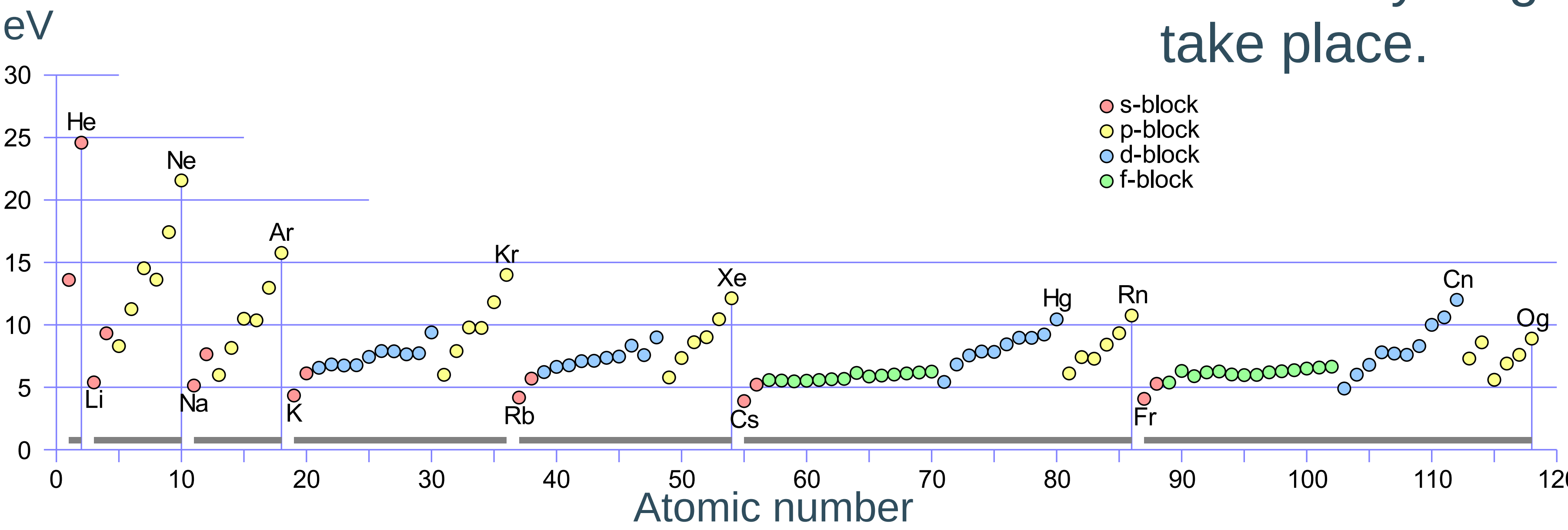
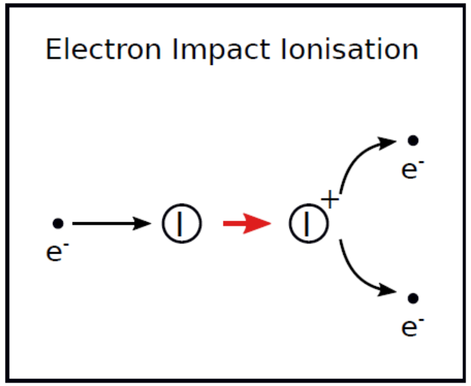
Electron impact ionization

- A stream of electrons is accelerated to sufficient energy to induce ionization upon collision with any atom.

- There must be a source of electrons and those must be overlapped with the atoms.
- It thus requires an interaction volume where both can be held sufficiently long for the interaction to take place.





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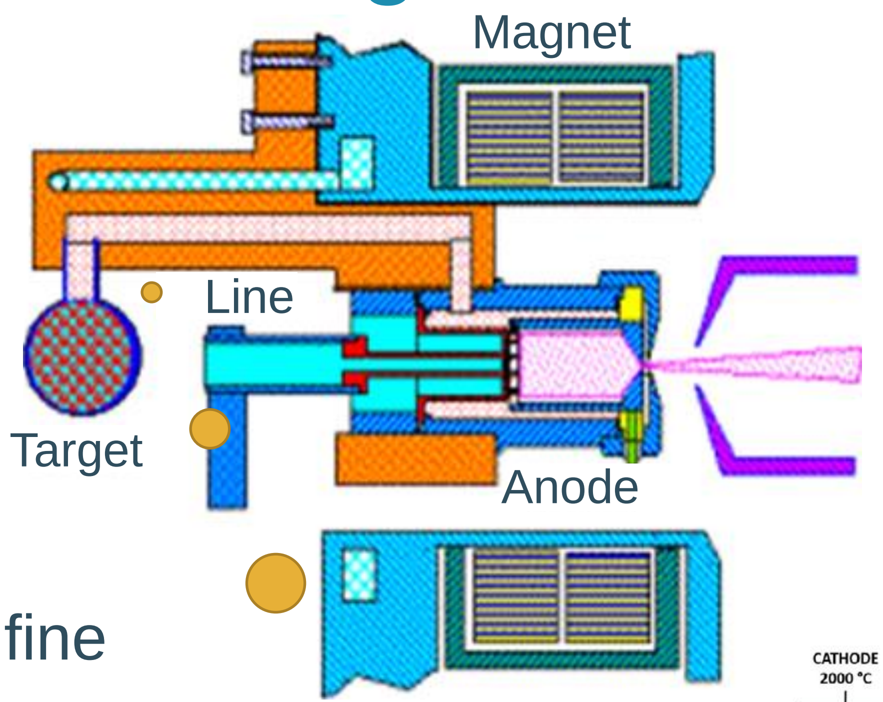
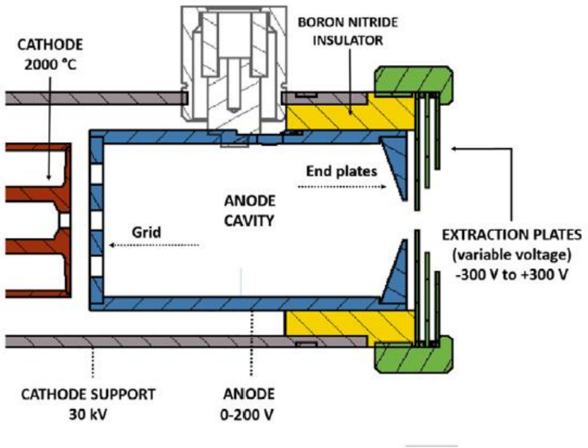
VADIS: Versatile Arc Discharge Ion Source

- The ISOLDE electron impact ion source is the VADIS.
- A cathode is heated to produce electrons and accelerated to an anode with a grid that lets most enter the anode volume.
- A magnetic field is applied to confine the electrons within the anode volume.
- A buffer gas is introduced to help containing the atoms.

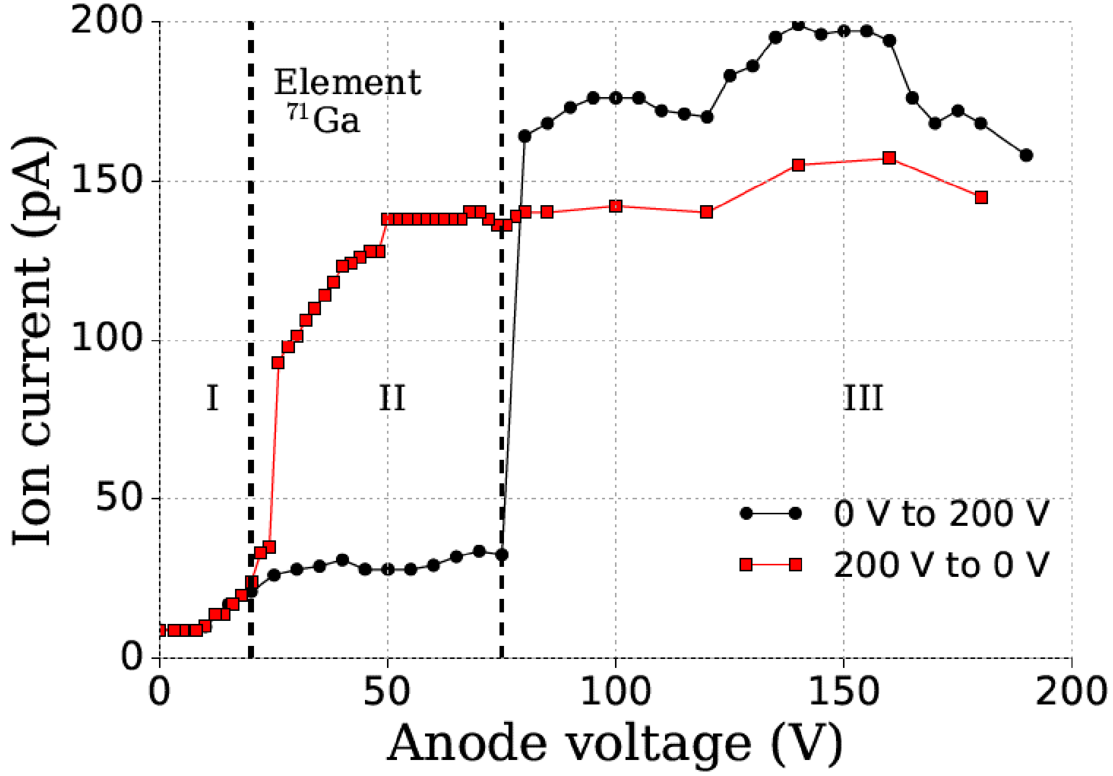


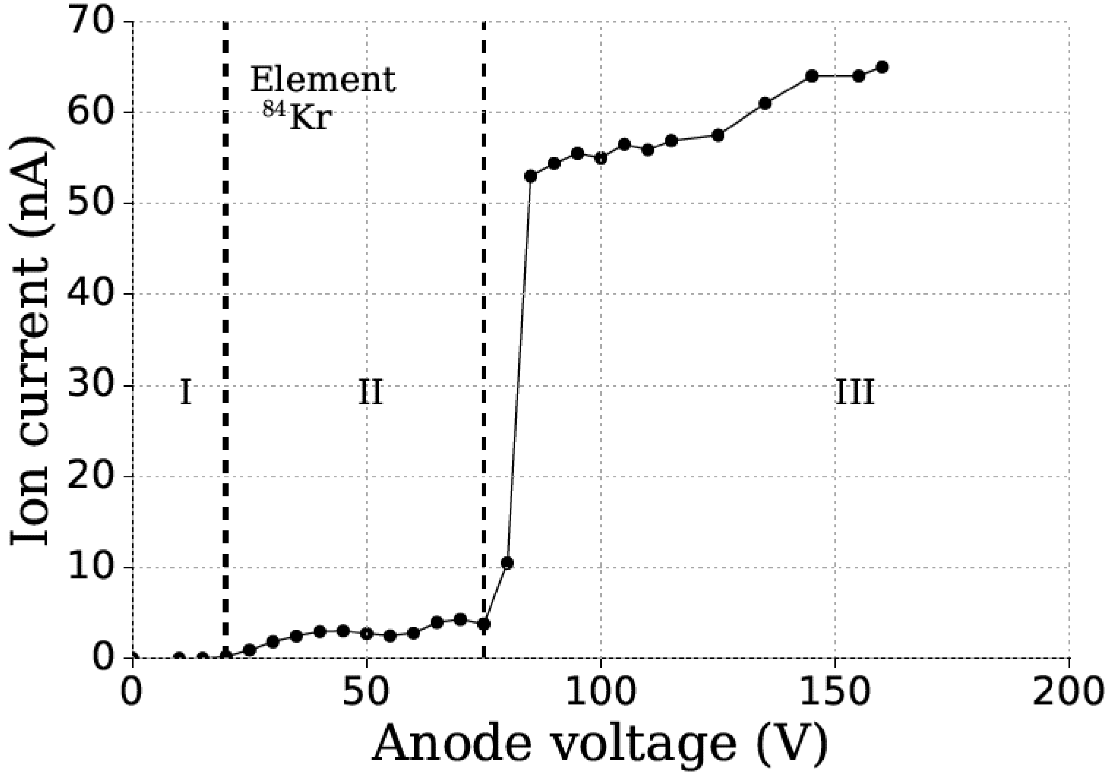


A cooled transfer line can be used to select only the most volatile elements!

 Y. Martinez Palenzuela et al., *Enhancing the extraction ... from an arc discharge ion source*, NIMB **431** (2018) 59-66.

VADIS: anode voltage





 Y. Martinez Palenzuela, PhD Thesis, KU Leuven (2019).

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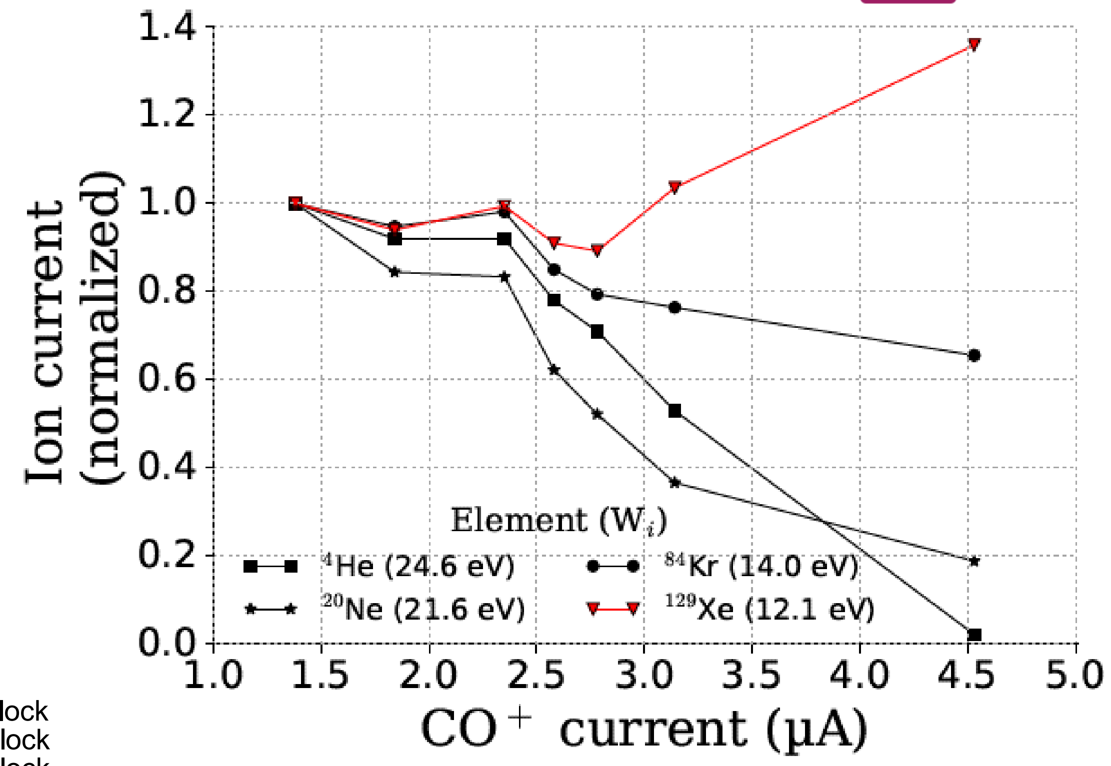
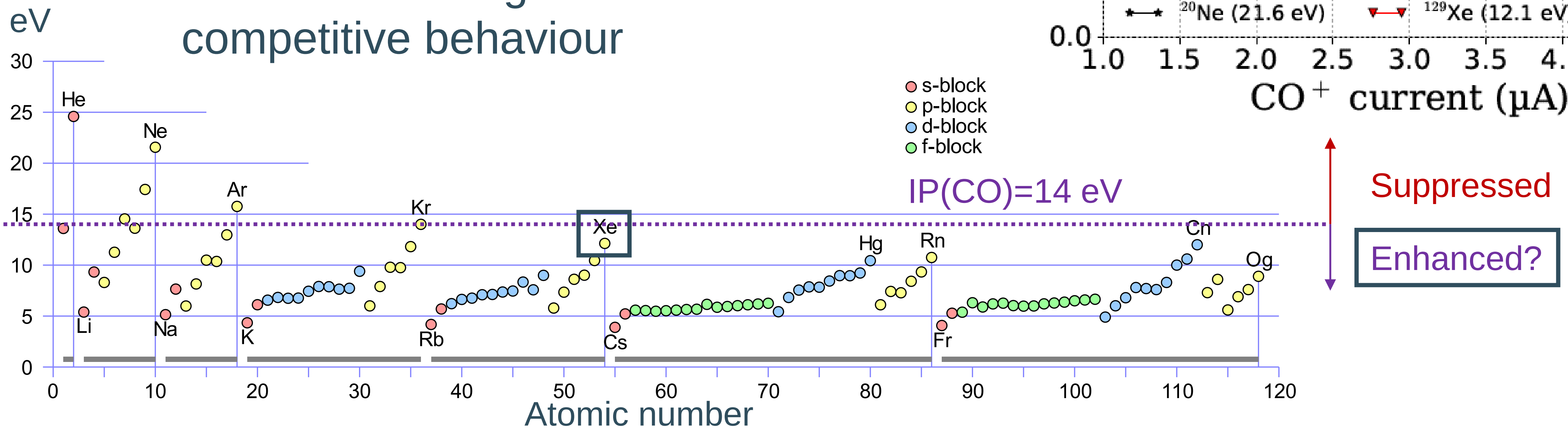
Load limitations

- The ion source can only cope with so much!
  - ➢If the ion load is too high, the efficiency drops significantly.
  - ➢Other effects might show competitive behaviour





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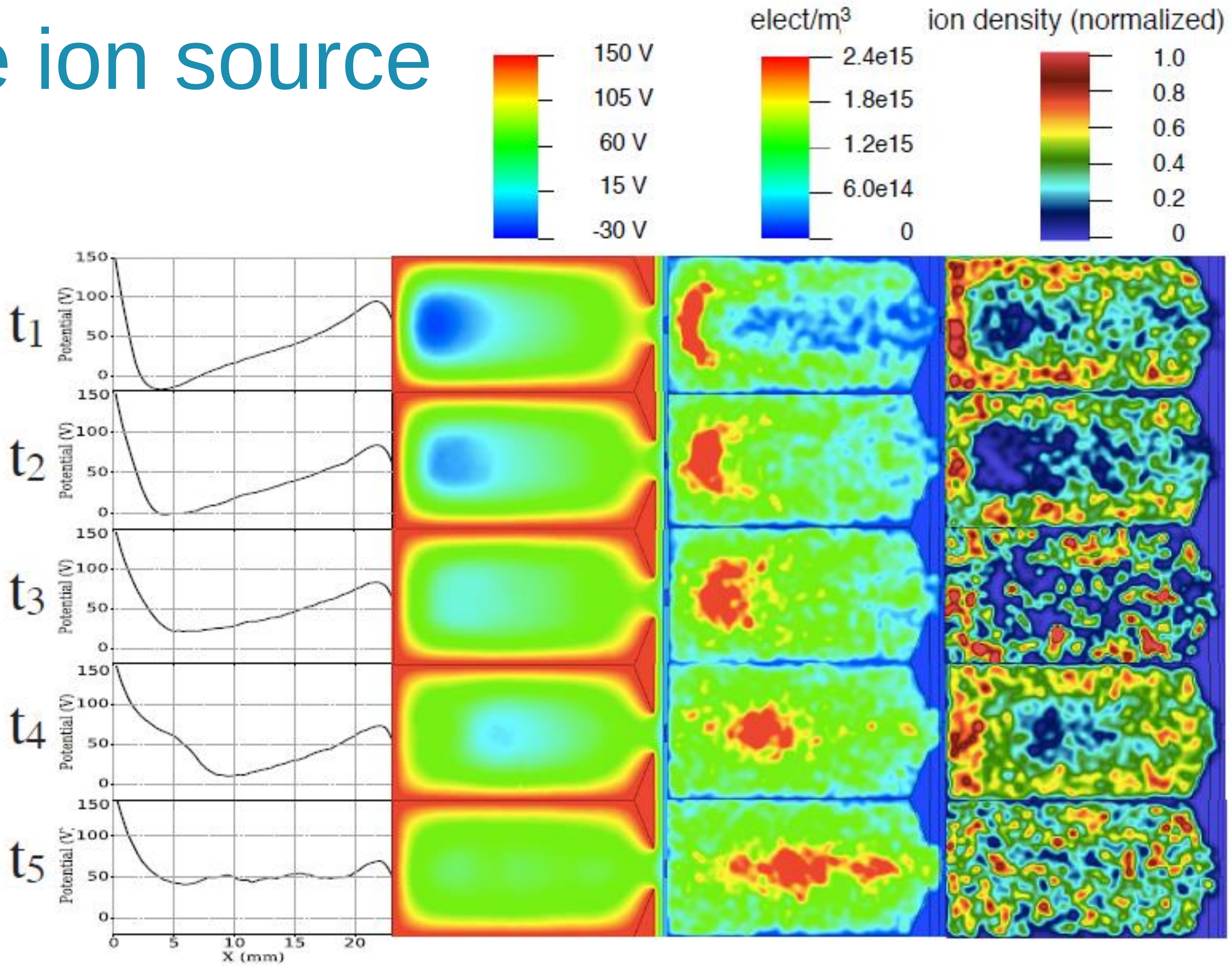
Understanding the ion source

- Advanced simulations may be used to study the dynamics of the ion source.
- One of the key aspects is the electrostatic field distribution within the anode volume, which determines the ‘active’ volume that may be extracted.
- Those fields are affected by the electron and ion densities in the anode volume.



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BREAK

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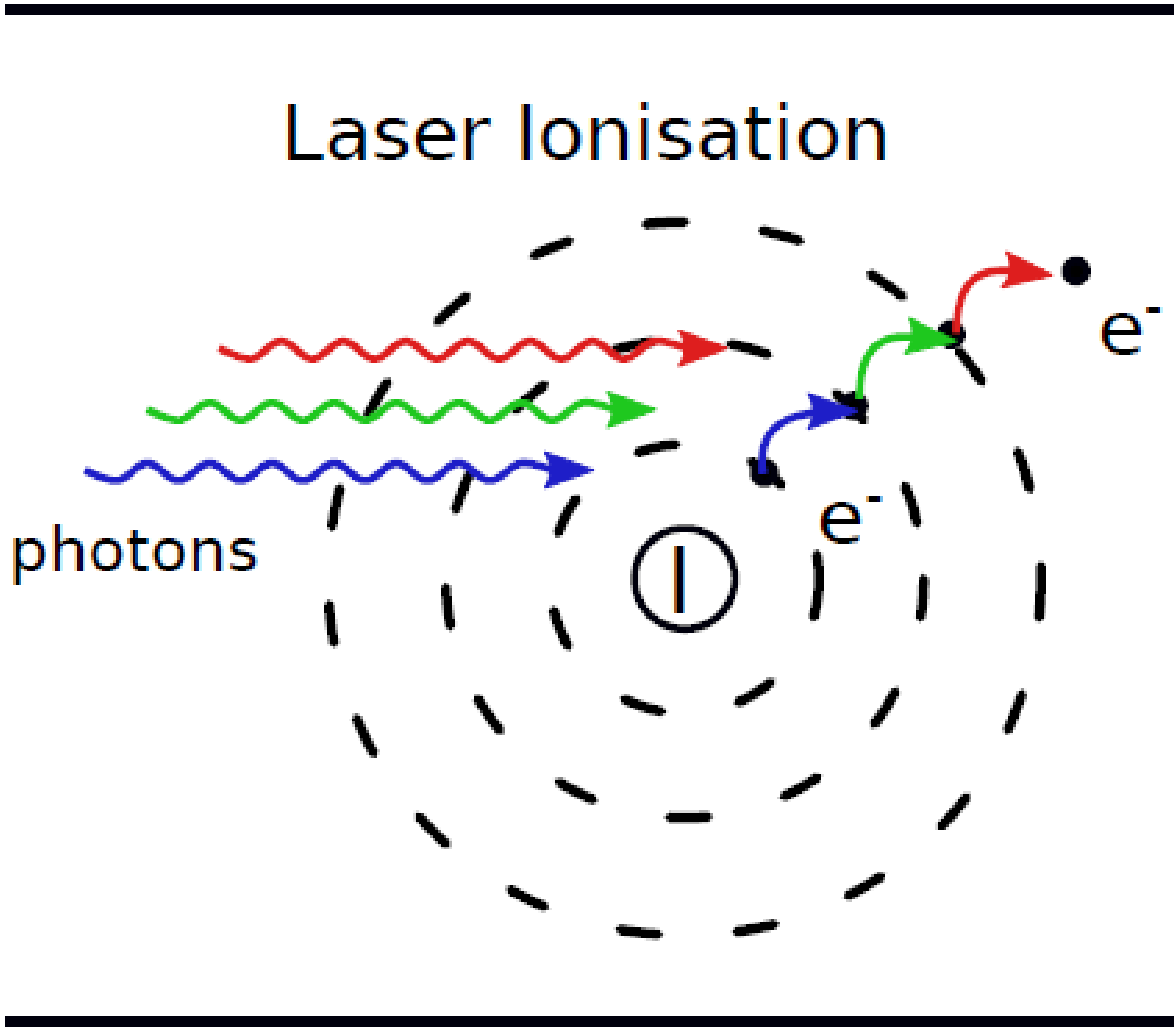
Resonant ionization



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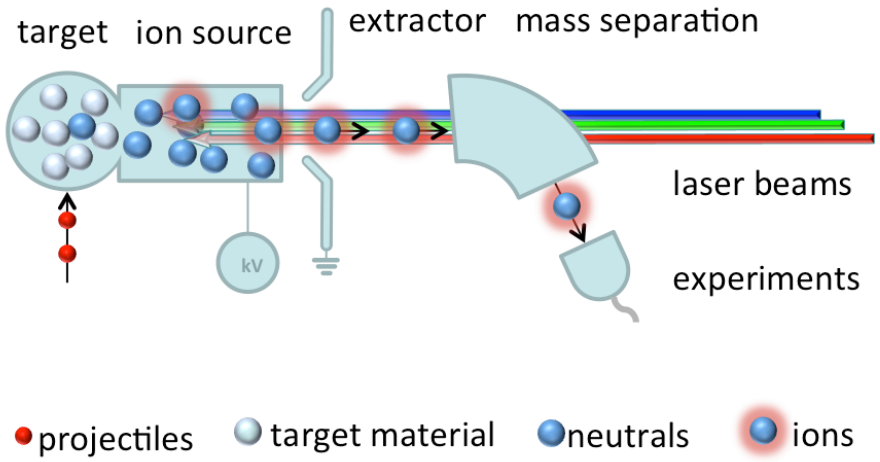
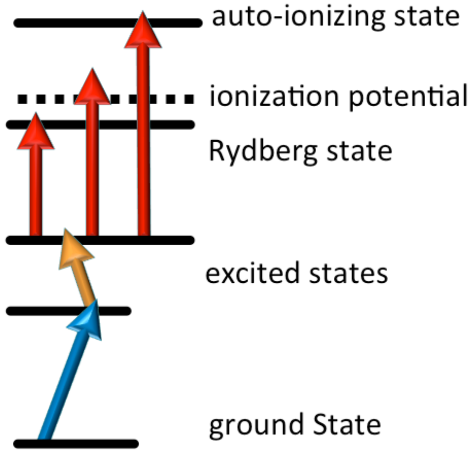
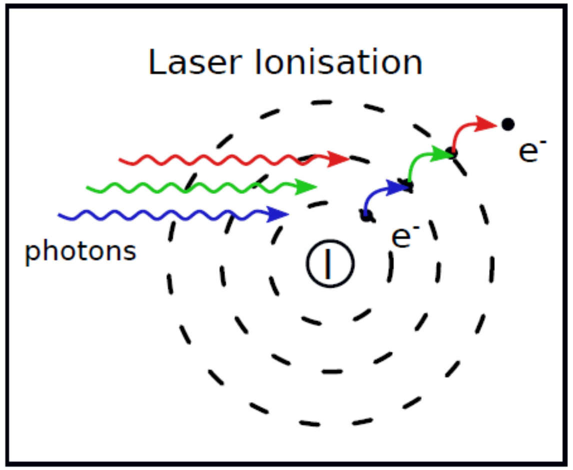
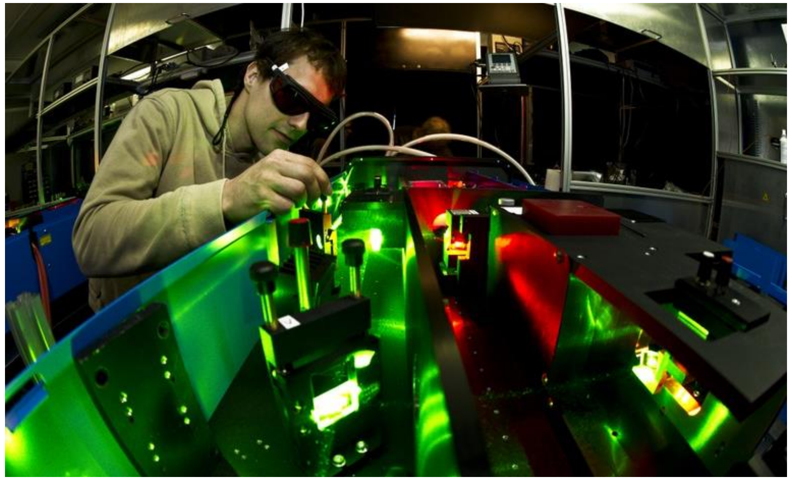
Shining light on the elements

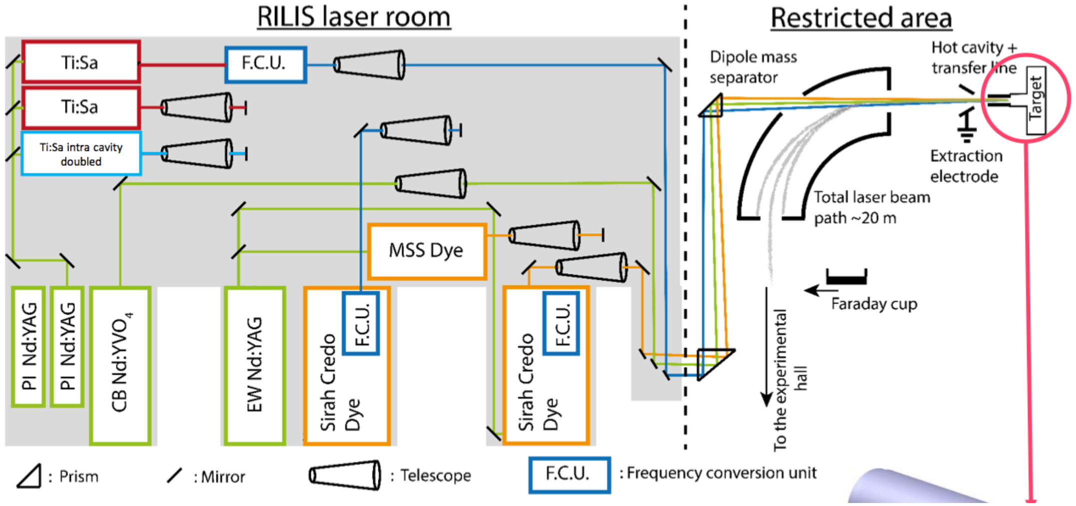
Resonance
Ionisation
Laser
Ion
Source











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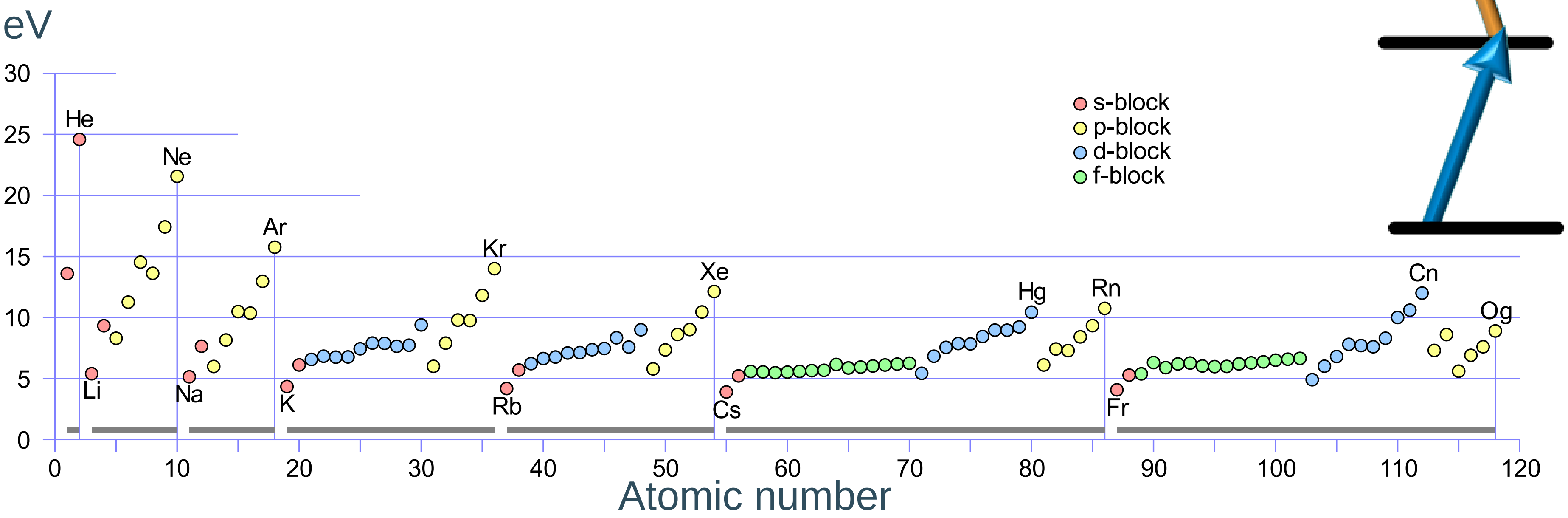
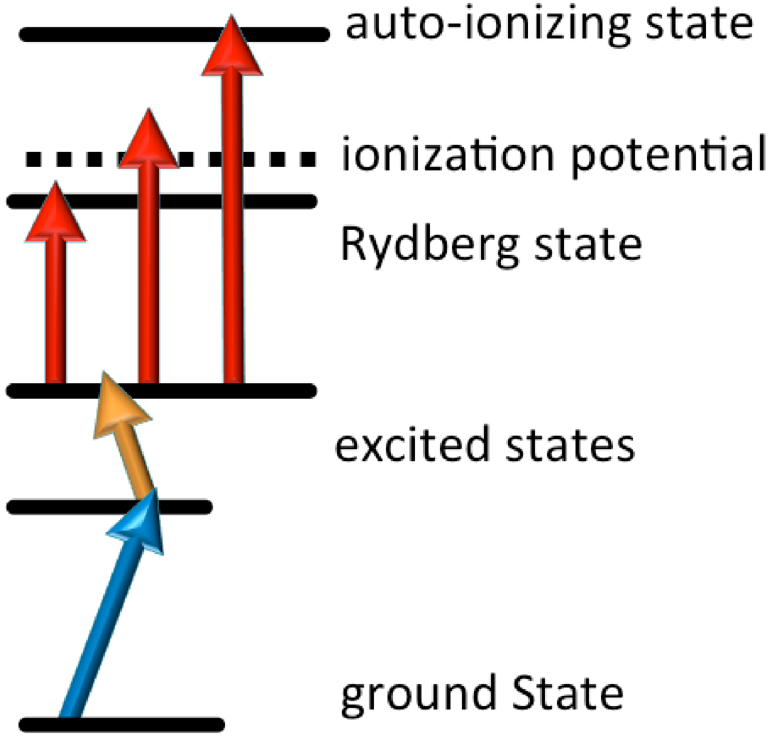
Resonant ionization concepts

- The **transitions** must be short enough in frequency to prevent the possible single-step ionization of any other element.
  - ➢Cs → Cs^+ is 3.89 eV = 940 THz = 318 nm





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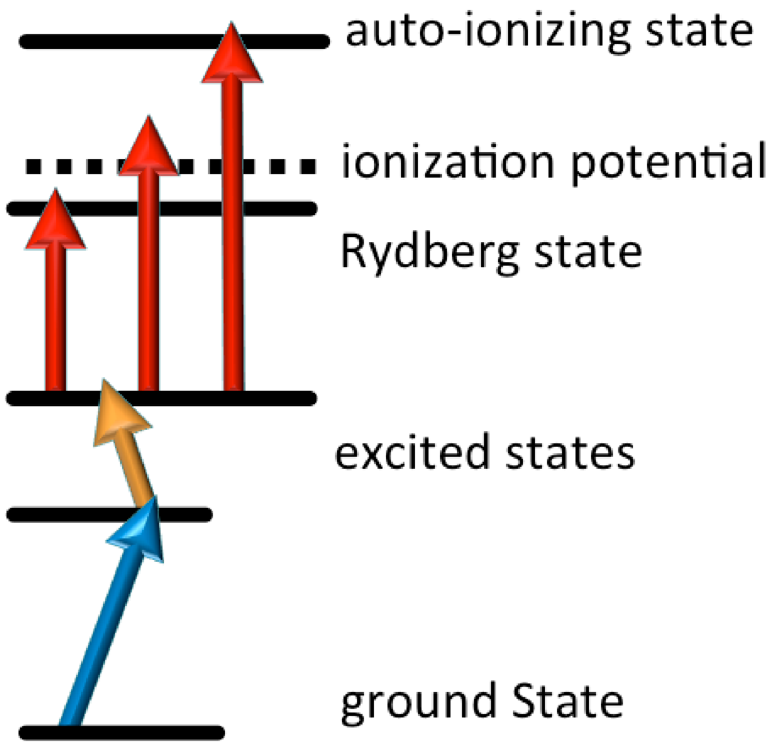
Resonant ionization concepts

- The **power** depends on the nature of the transition:
  - ➢Resonant transitions saturate with little power of a few mW.
  - ➢Transitions to auto-ionizing states might require a bit more to saturate as they are also broader.
  - ➢Non-resonant transitions require several W to saturate and are rarely saturated.
    - ❖Too much power may also induce 2-photon absorbtion, hereby open ionization of other elements
- Importance of **power density** ➔ Pulsed lasers



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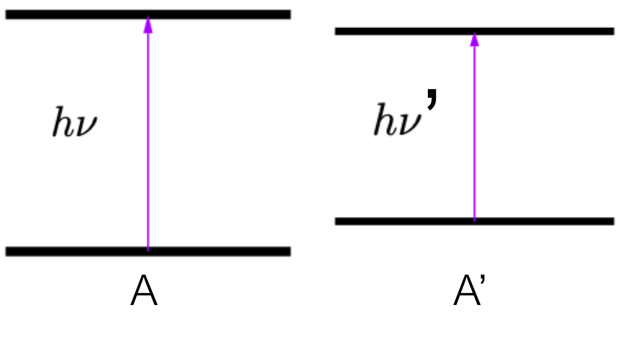
Nuclear component to resonant ionization

Key aspect for the next lecture!

Isotope shift

- The finite size of the charge distribution of the nucleus induces a perturbation of 1:10^6 on atomic levels.



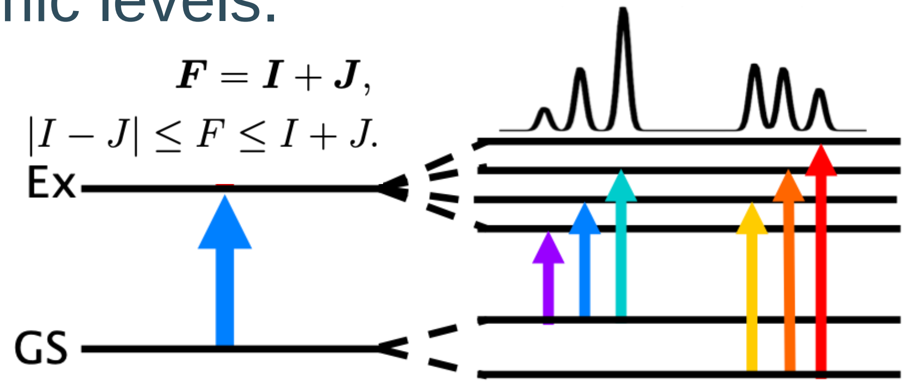
$$\delta\nu^{AA'} = \frac{A'-A}{AA'}\left(m_e\nu + M_{SMS}\right) + F\delta\langle r^2\rangle^{AA'}$$

Hyperfine structure

$$A = \frac{\mu B_0}{IJ} \qquad B = \frac{eQ}{4}\frac{\partial^2 V}{\partial z^2}$$

- The electromagnetic moments of the nucleus induce a split and shift in the atomic levels.

$$\boldsymbol{F} = \boldsymbol{I} + \boldsymbol{J},$$
$$|I-J| \leq F \leq I+J.$$



$$\Delta E = \frac{A}{2}K + \frac{B}{2}\frac{3K(K+1) - 2I(I+1)2J(J+1)}{2I(2I-1)2J(2J-1)}$$

$$K = F(F+1) - I(I+1) - J(J+1)$$

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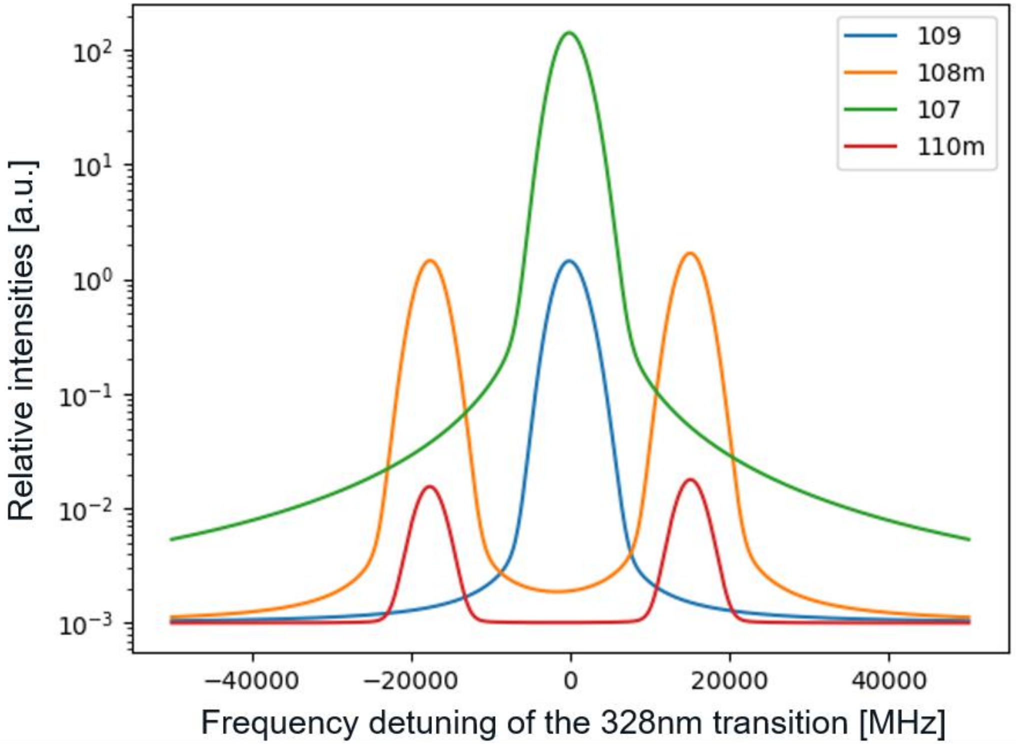
Impact of IS & HFS on RILIS?

Isotope separation with the lasers!

- The balance comes in the comparison of the resonance linewidth vs the IS & HFS effects.
  - ➢ The high operation temperature induces a large Doppler broadening of 3-5 GHz.
  - ➢ The high power of the lasers may also induce further broadening of ~1 GHz.
  - ➢ The lasers are typically operated at 10 GHz linewidth but may be reduced to 1.2 GHz if needed.
- The IS and HFS are element and even transition specific!
  - ➢ Cu, Ag, and Au have particularly large ground-state HFS.
  - ➢ Coupled with a odd-even staggering in the moments due to the coupling of the neutron with the proton.

Ag ionization



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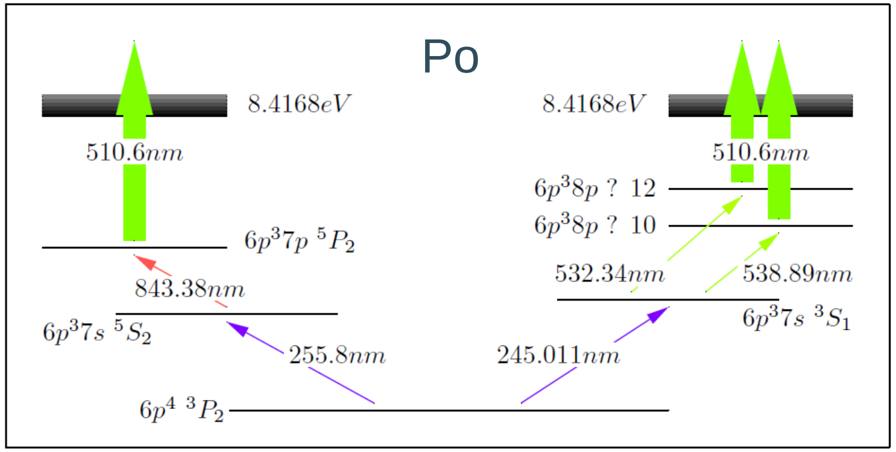
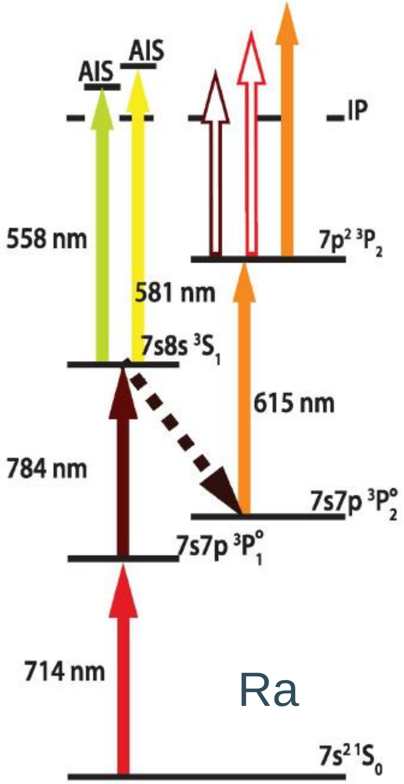
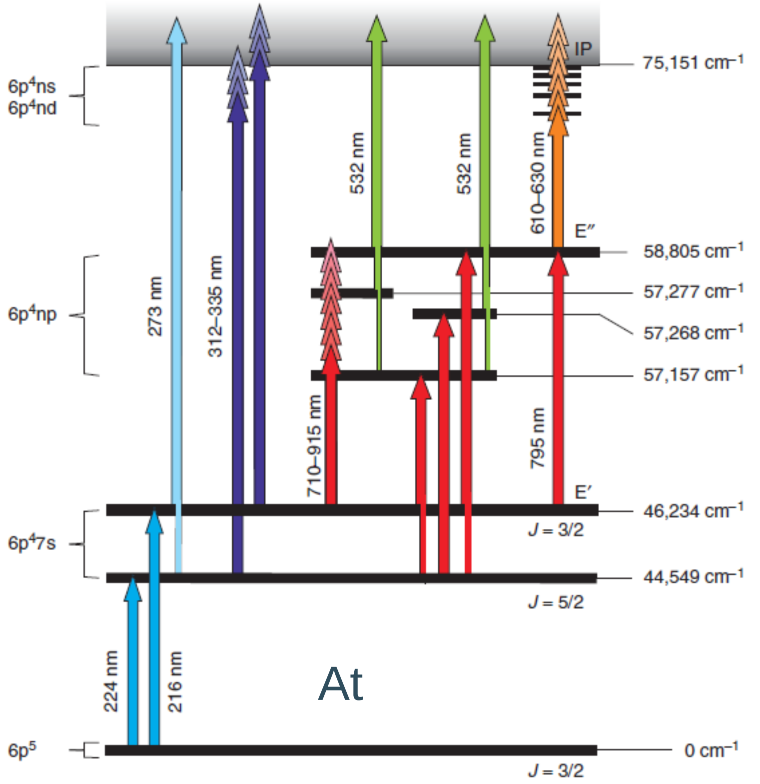
Trial and error

- For each element, dedicated investigations are needed to identify the most appropriate ionization scheme.
- The population might also be split between different low excitation states due to the high temperature.
- The literature is a good starting point but not always sufficient: e.g. Po, At, Ac, ...

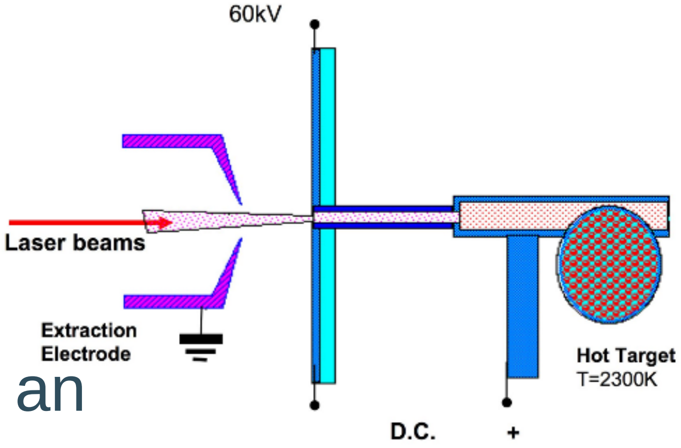


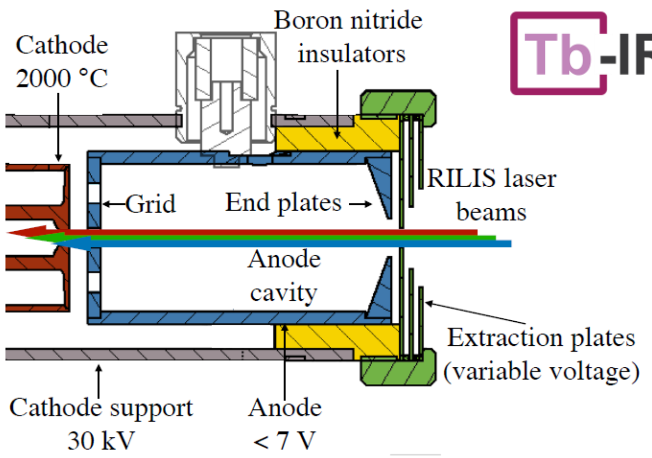




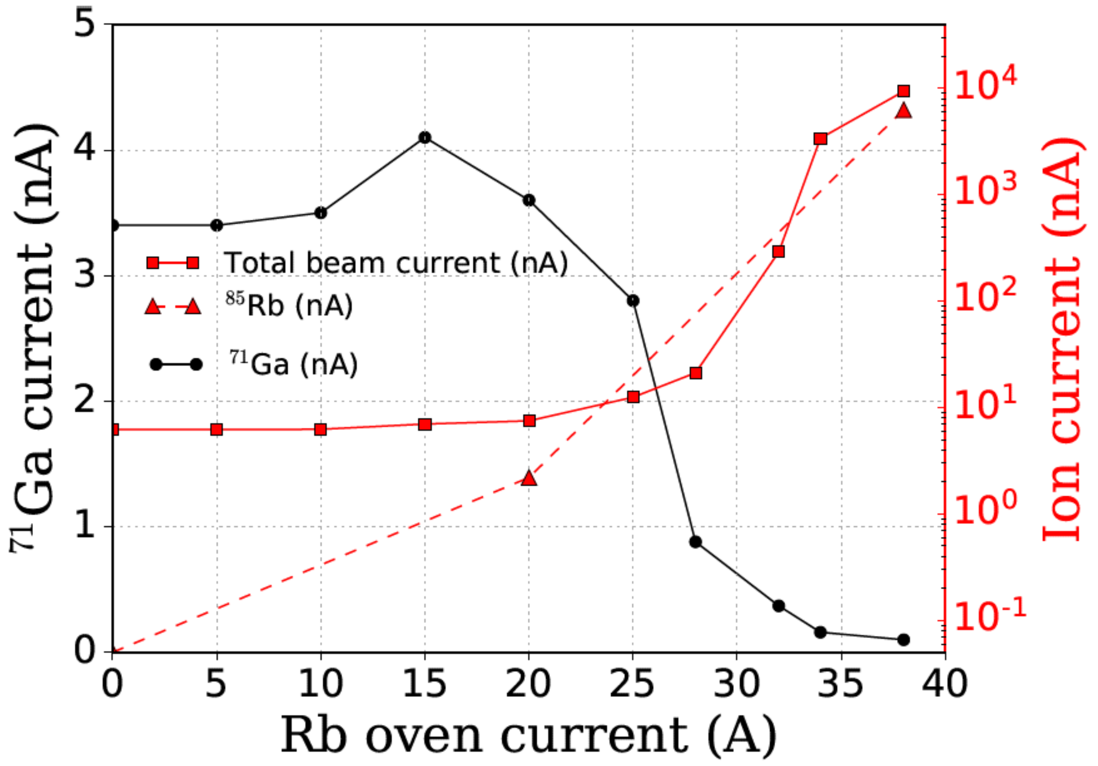
*T.E. Cocolios et al., Resonant laser ionization of polonium ..., NIMB **266** (2008) 4403-4406.*
*S. Rothe et al., Measurement of ... astatine by laser ionization spectroscopy, Nature Communications **4** (2013) 1835.*
*T. Day Goodacre et al., Radium ionization scheme development..., Spectrochemica Acta B **150** (2018) 99-104.*

Load limitation





- A laser ion source requires an atomizer:
  - ➢A hot tube ➔ surface ion source
  - ➢A large volume ➔ VAD(L)IS
- Competing ionization mechanisms cannot be avoided.
- The ion source may easily become saturated by other ions and dramatically affect the ionization efficiency.



 Y. Martinez Palenzuela, PhD Thesis, KU Leuven (2019).

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Beam manipulation

- Time structure
- Mass separation

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Time structure of the ion beam

Primary beam time structure

- (Pseudo-)Continuous drivers
  - ➢ The cyclotron at TRIUMF delivers a pulsed beam at high repetition rate that flies such a long distance that the bunches overlap and the target receives a slightly modulated modulated proton beam.
- Pulsed drivers
  - ➢ The PSB at CERN has a period of 1.2s and delivers bunches to ISOLDE with an irregular pattern of bunches separated by an integer number of periods.
  - ➢ Pulsed drivers have a high instantaneous power deposition, even if the average power is limited!

Radioactive ion beam time structure

- The diffusion and effusion processes are inducing delays upon the release of the radioactive ions from the target.
  - ➢ Depends on the target material (grain size, porosity);
  - ➢ Depends on the target temperature;
  - ➢ Depends on the element (target chemistry)
- The full time structure will be a convolution of the primary beam, release, and half-life of the radioisotope.
  - Short-lived isotopes may decay during the release process and not make it to the ion source!

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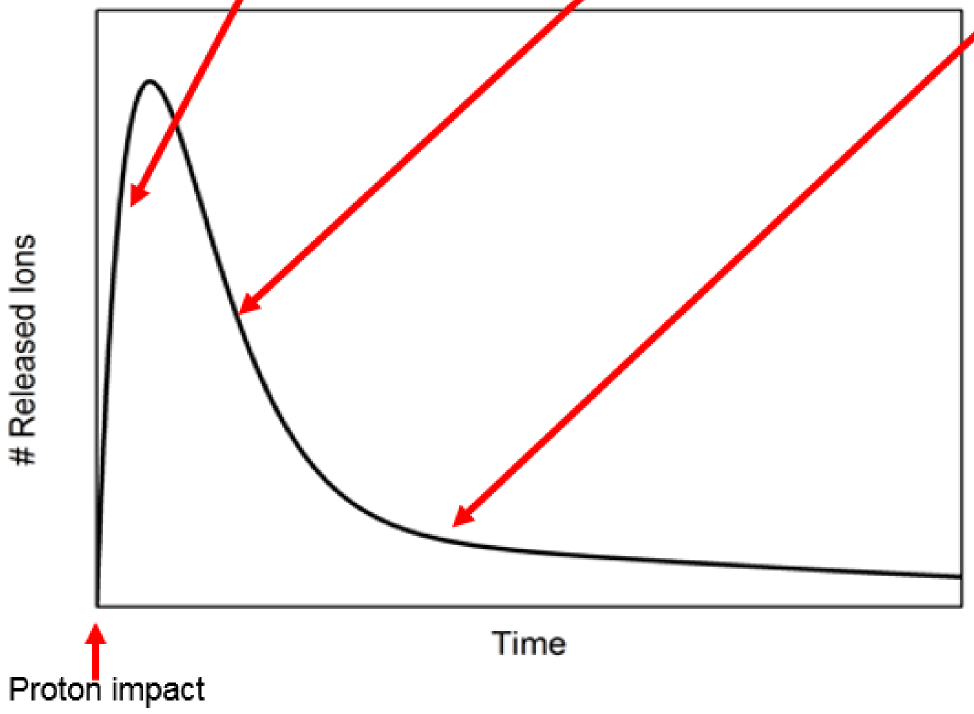
Isotope release

Fast component

Slow component

$$P_i(t,\lambda_i) = \exp(-\lambda_i t)\cdot\frac{[1-\exp(-\lambda_r t)]\cdot[\alpha\cdot\exp(-\lambda_f t)+(1-\alpha)\cdot\exp(-\lambda_s t)]}{\text{Normalisation factor}}$$

To convolute with the half-life



Slide courtesy of Kristof Dockx.

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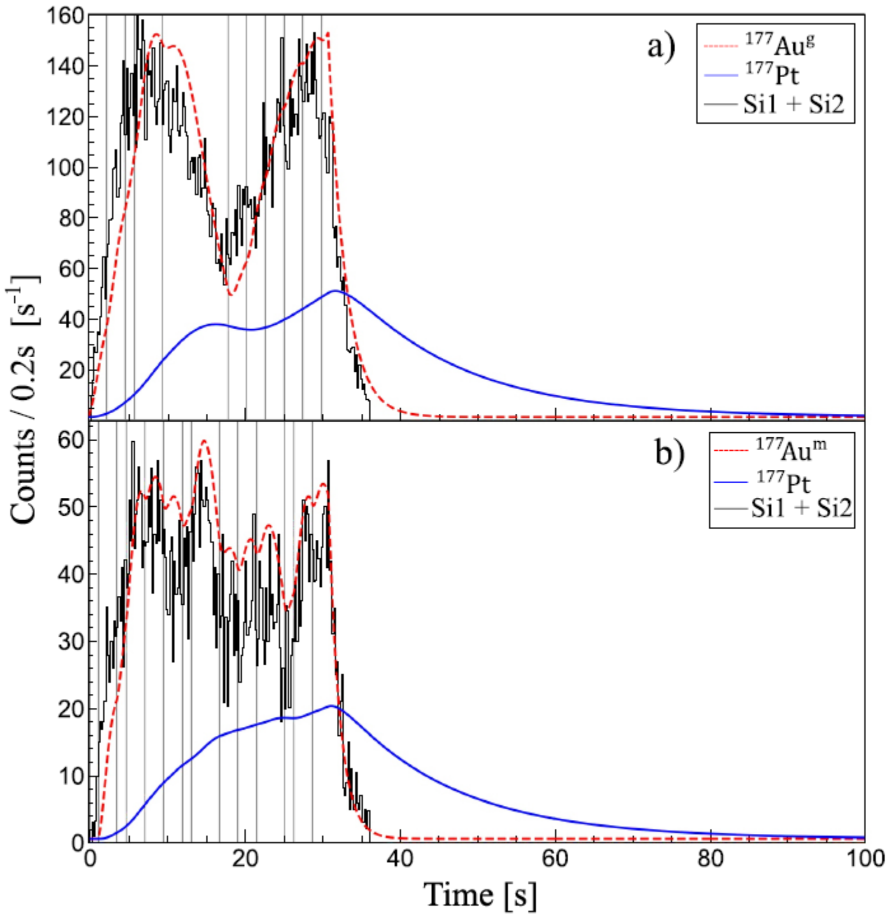
A practical illustration: producing Au

- Showing the alpha decay rate of ^{177g}Au ($T_{1/2} = 1.193\text{s}$) and ^{177m}Au ($T_{1/2} = 1.501\text{s}$) isotopes from direct production.
  - Beam delivery is stopped at $t = 30.89\text{s}$
  - Observation is concluded after $t = 36\text{s}$
  - These simulations are used to estimate the missed observation of ^{177}Au and ^{177}Pt ($T_{1/2} = 10\text{s}$) from beta decay of Au.
- Each vertical gray line is a proton pulse.
- The red-dotted line is a simulation of the time structure where the only free parameter is the total number of alphas.



J.D. Johnson et al., Supplementary material to...
*R.D. Harding et al., Laser assisted nuclear decay spectroscopy of ^{176,177,179}Au, PRC **104** (2021) 024326.*

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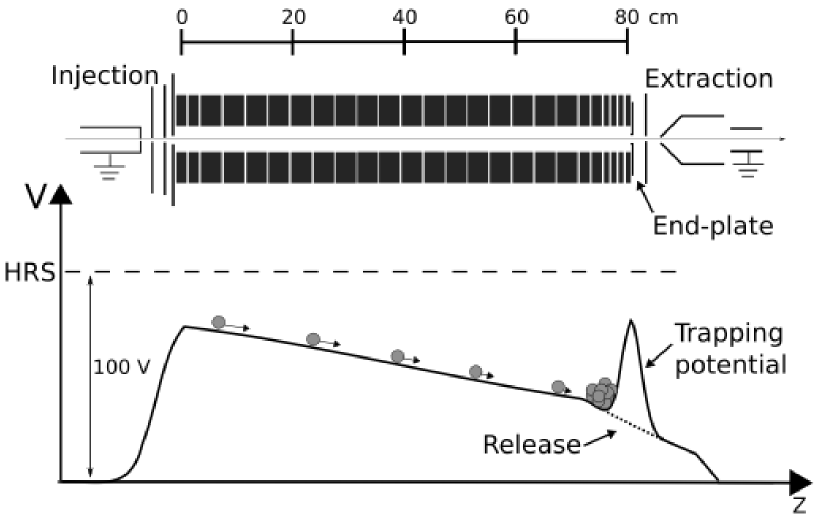
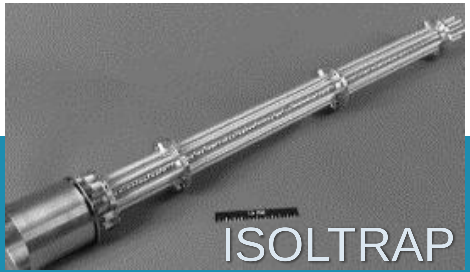
RFQ Cooler Buncher



- Accelerators can go positive or negative
  - Controlled beam deceleration
- The electric field gradient can be shaped to control the beam delivery
  - Trapping
  - Bunching
- The use of a buffer gas rather than an evacuated volume enhances the collisions, hereby reducing the beam straggling until all particles are nearly at rest
  - Beam cooling
  - Best possible emittance
- Those properties are ideal for the handling of low-energy beams, enhancing the time definition and beam quality
  - Injection into Penning traps for high-resolution mass measurement
  - Bunched-beam laser spectroscopy



Key aspect for the next lecture!

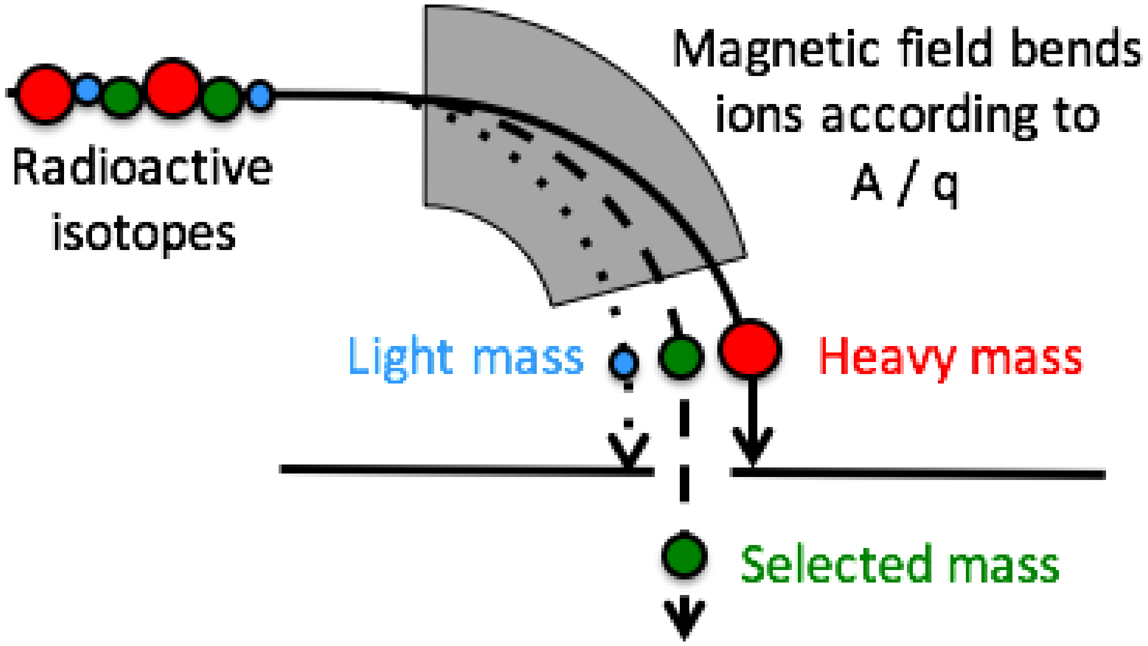
*F. Herfurth et al, NIMA **469** (2001) 254-275.*
*E. Mané et al, EPJA **42** (2009) 503-507.*

Mass separation

$$E = \frac{1}{2}mv^2 \Rightarrow v = \sqrt{\frac{2E}{m}}$$

$$F = qv \times B = ma$$

- A mono-energetic ion beam at energy 30-60 keV is extracted, shaped, and transported to a dipole magnet.
- The magnetic rigidity (bending radius) of each element is dependent upon the magnetic field and the mass-to-charge ratio ${}^{m}/_{q}$.
- Tuning the magnetic field allows to select a given ${}^{m}/_{q}$, though beams typically have $q = 1$, resulting in mass separation.
- Resolution varies from $R = {}^{m}/_{\Delta m}$ 500 to 20,000 for the most ambitious. 500 corresponds to single mass separation, while 20,000 is enough to separate molecules from single-element ions.



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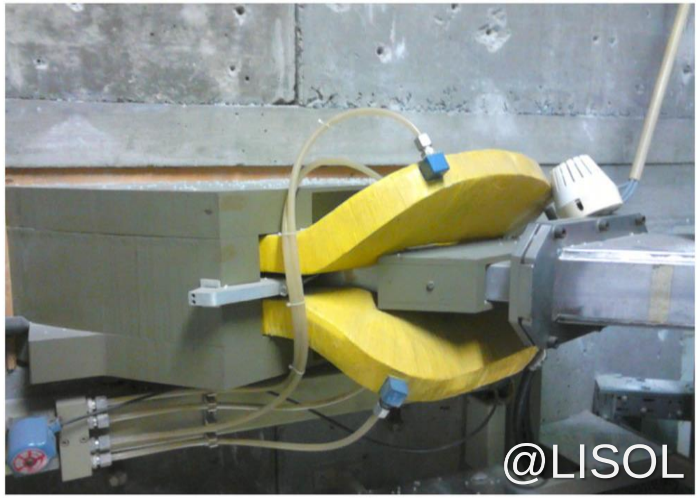
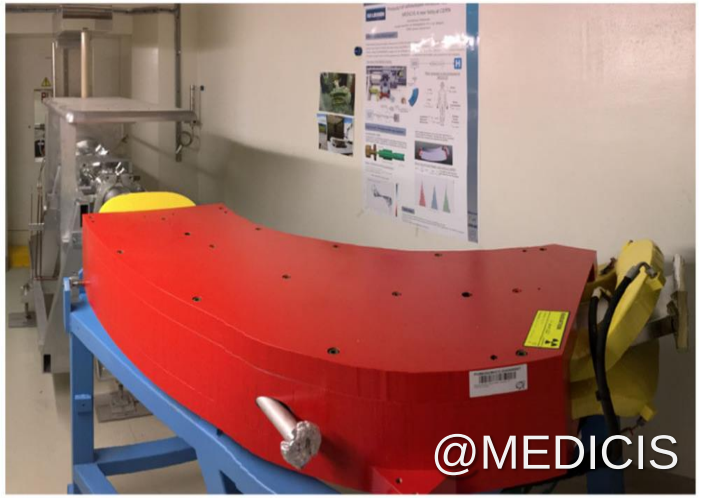
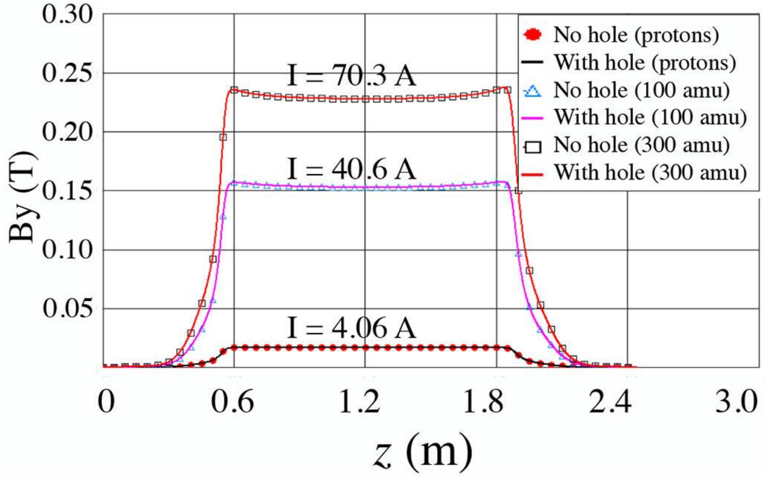
Not just a magnet...

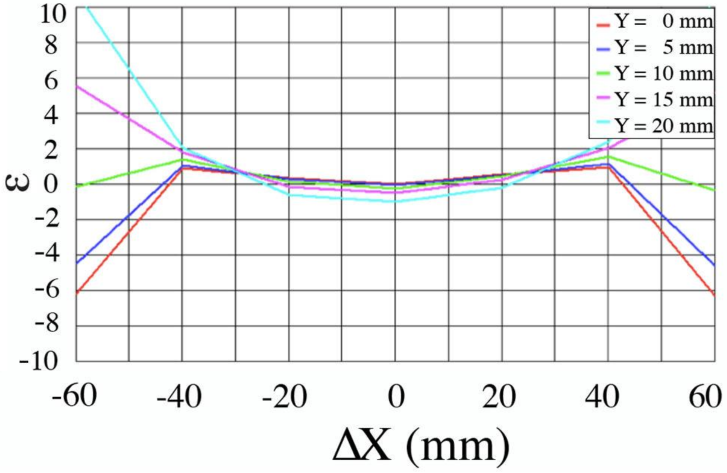
- The homogeneity of the magnetic field is crucial to the resolving power of the dipole magnet.
- Extensive ion beam and magnetic simulations, and accurate field mapping are required to achieve the expected resolving power.

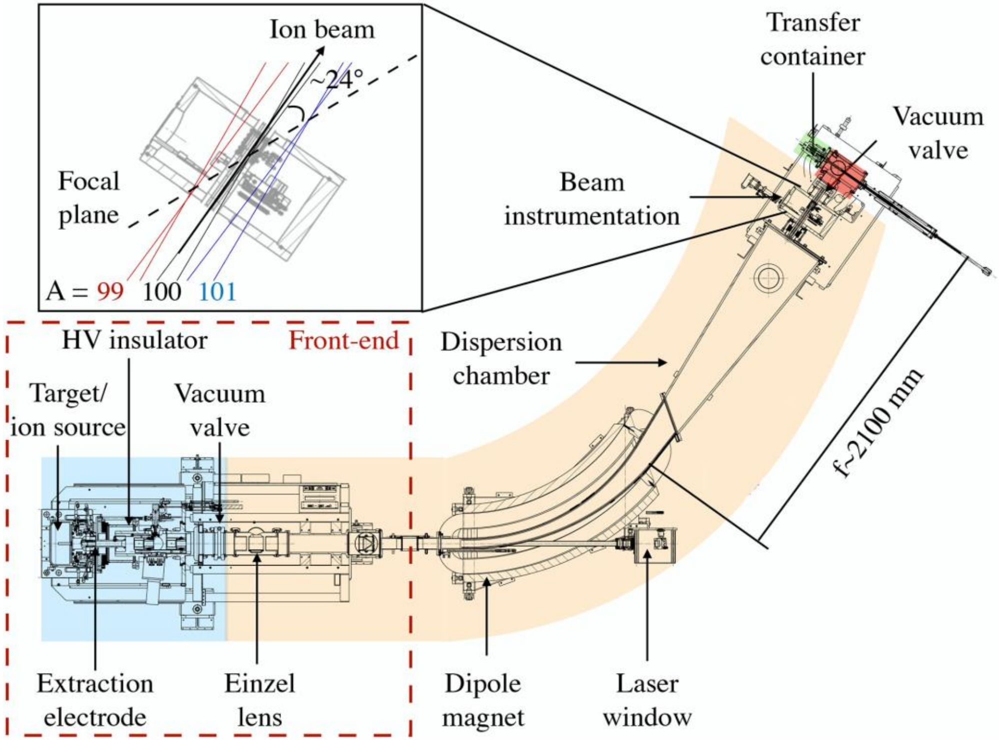




Simulation of the impact of the laser window







Y. Martinez Palenzuela et al., The CERN MEDICIS isotope separator beamline, Frontiers in Medicine ***8*** *(2021) 689281.*

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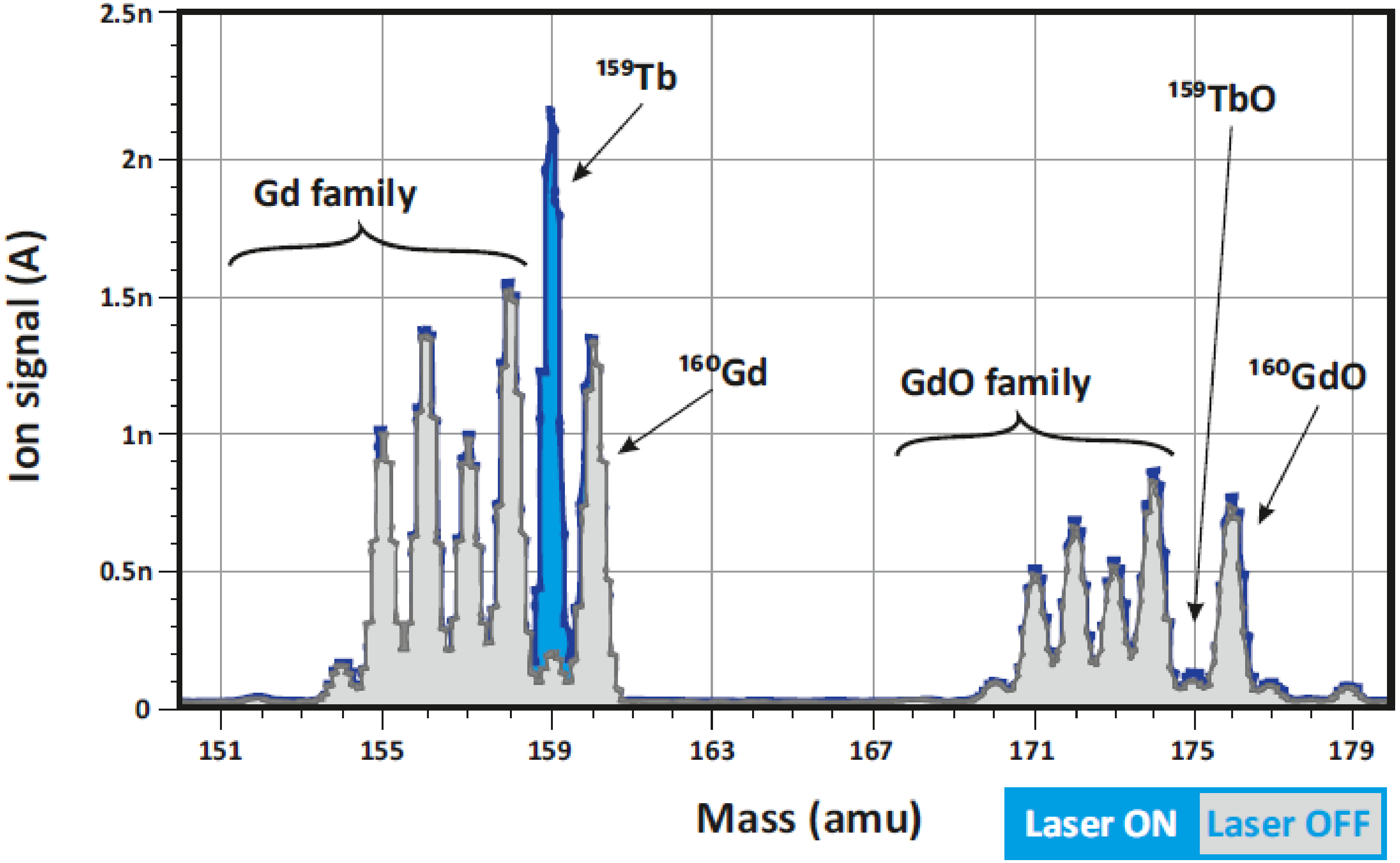
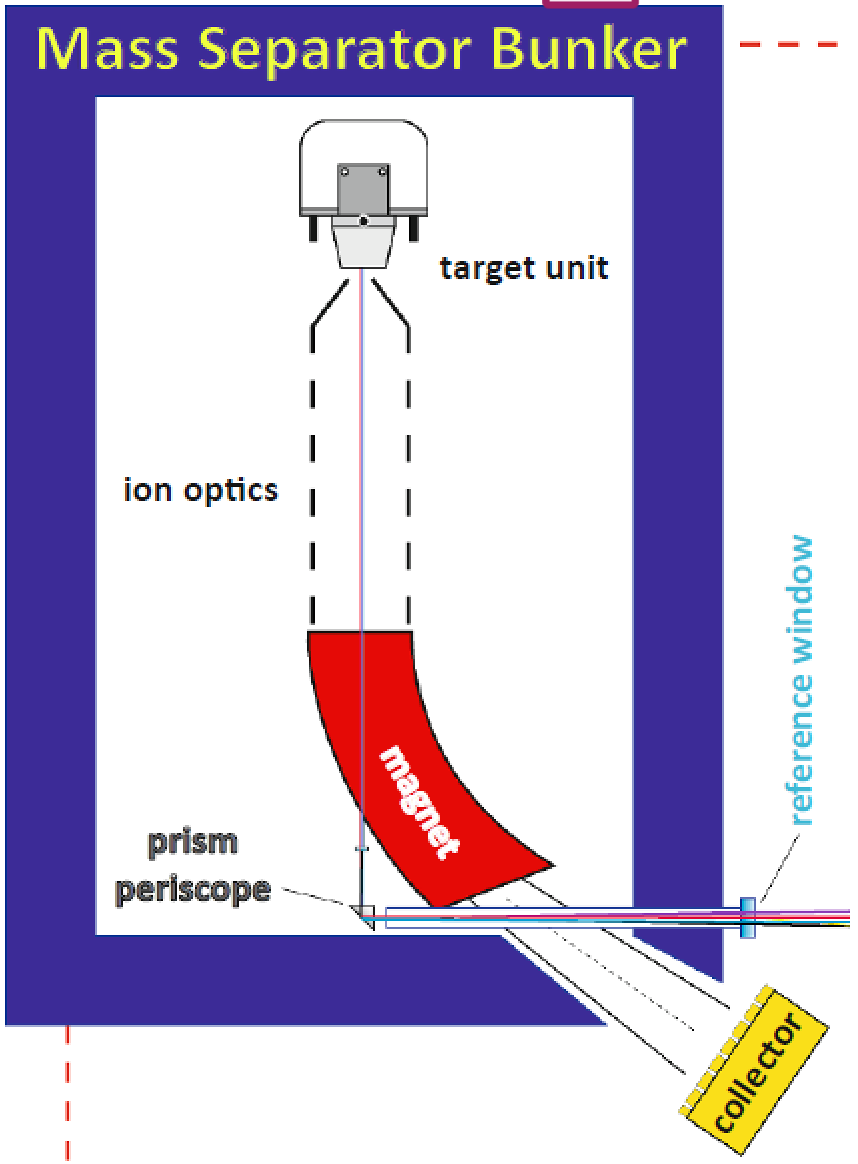
Mass scan





V.M. Gadelshin et al., MELISSA: Laser ion source setup at CERN-MEDICIS..., NIMB **463** (2020) 460-463.
V.M. Gadelshin et al., First laser ions at the CERN-MEDICIS facility, Hyperfine Interactions **241** (2020) 55.

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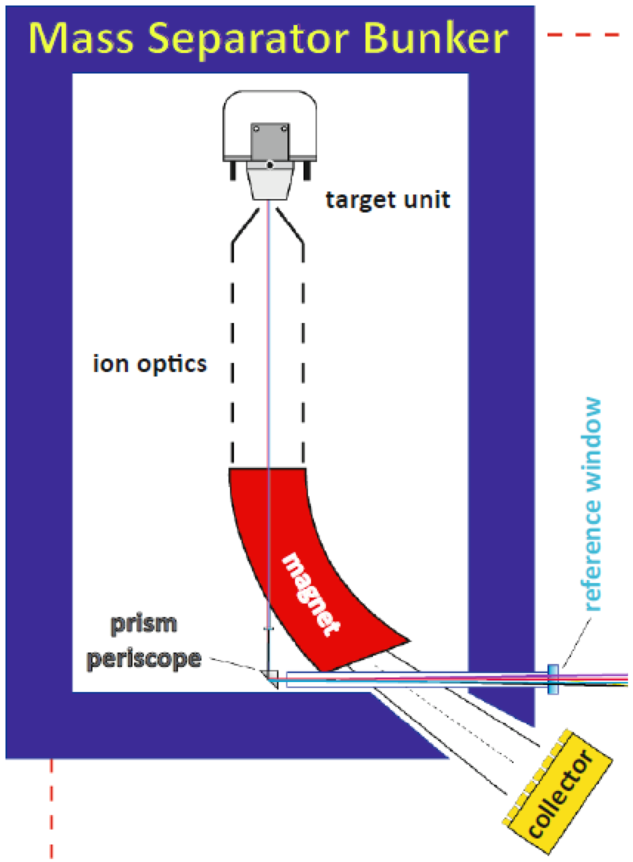
Mass scan



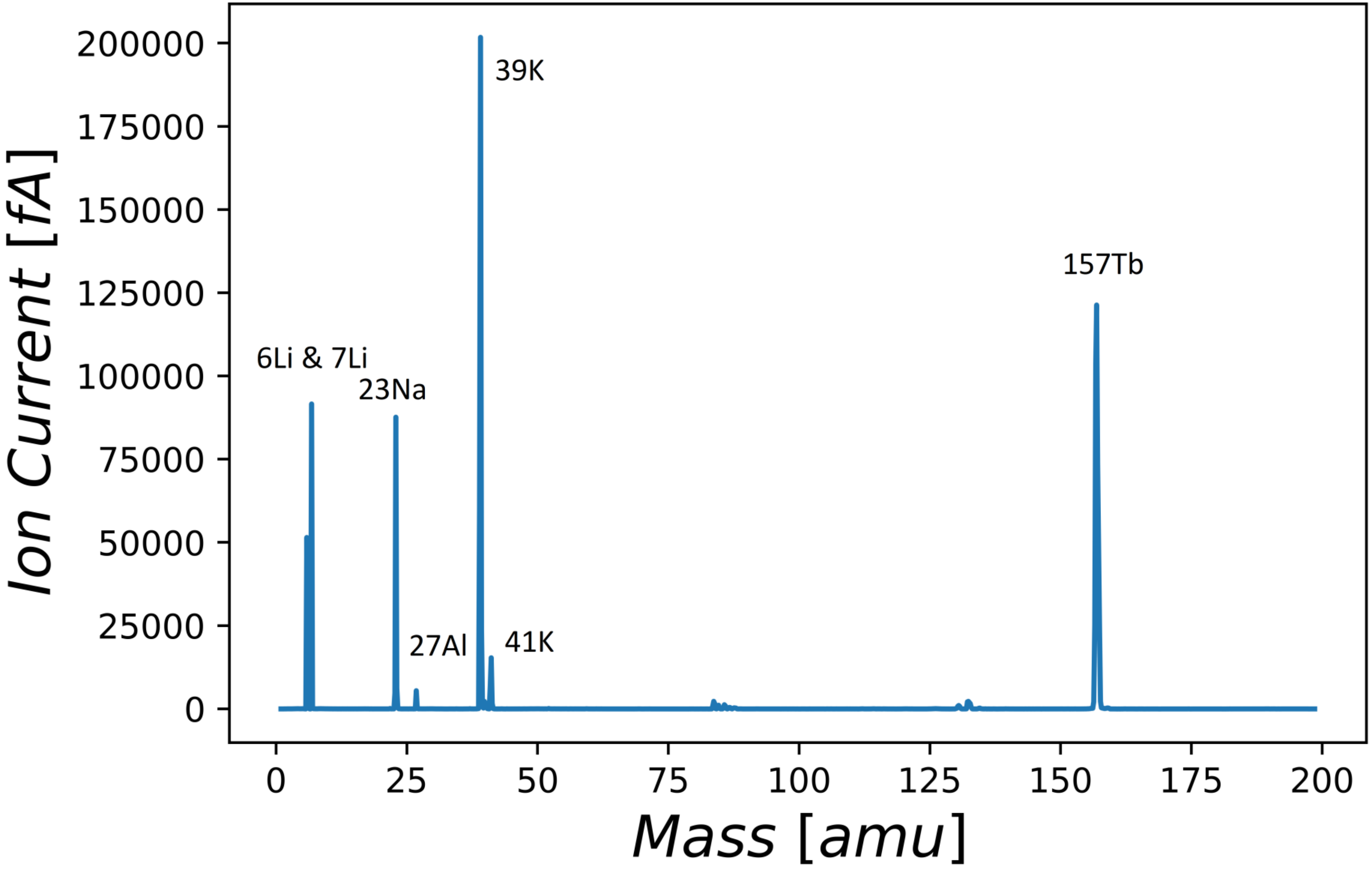


Figure courtesy of Willemijn Goossens, KU Leuven, for MED020.

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Back to efficiencies

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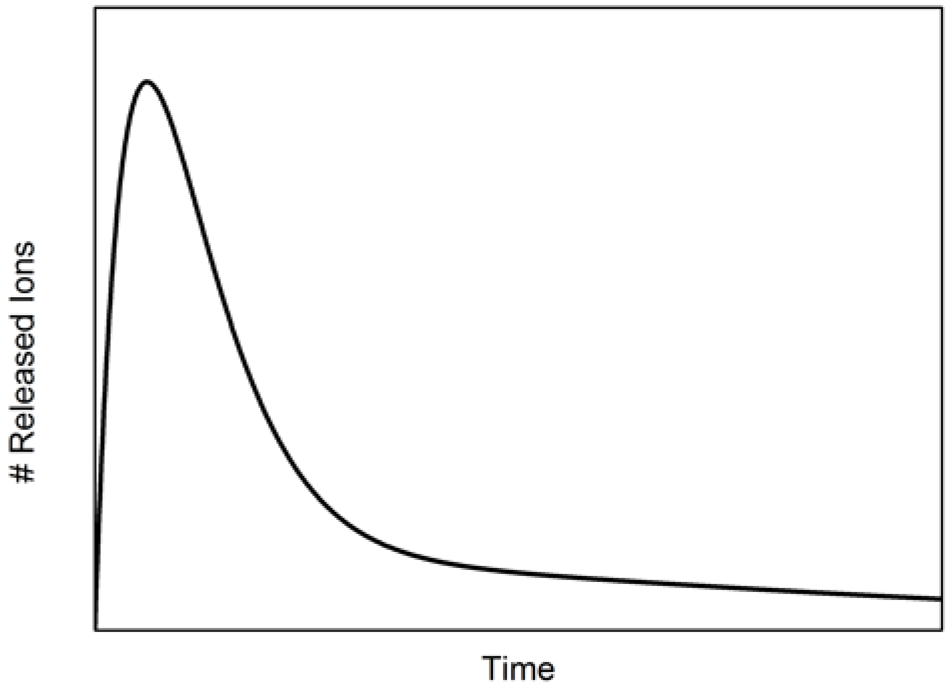
Isotope release

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

Limiting factors

- Diffusion & effusion
  - Partial or total
  - Delays and decay losses
- Ionization efficiency
  - Elemental property
  - Depends on the ion source load
- Separation and transport are well under control and are typically factored out

Time structure vs half-life



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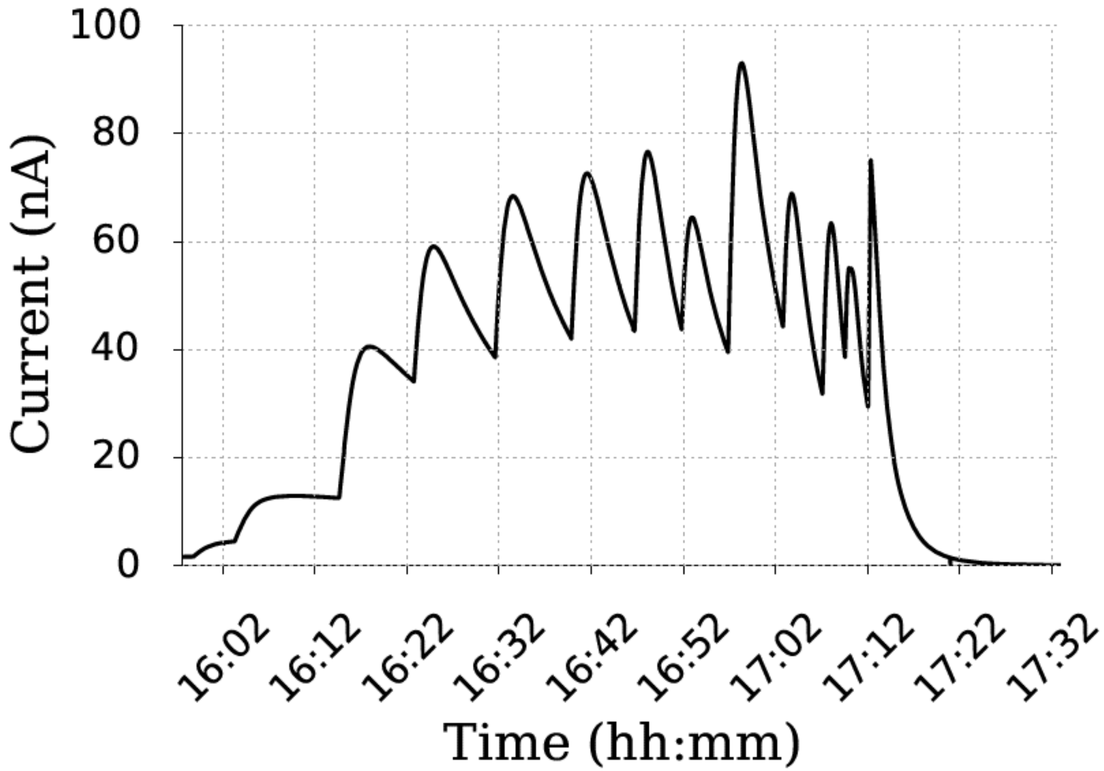
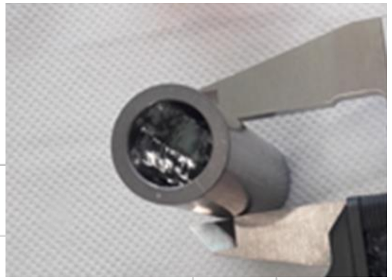
$\varepsilon = \varepsilon_{ion}$

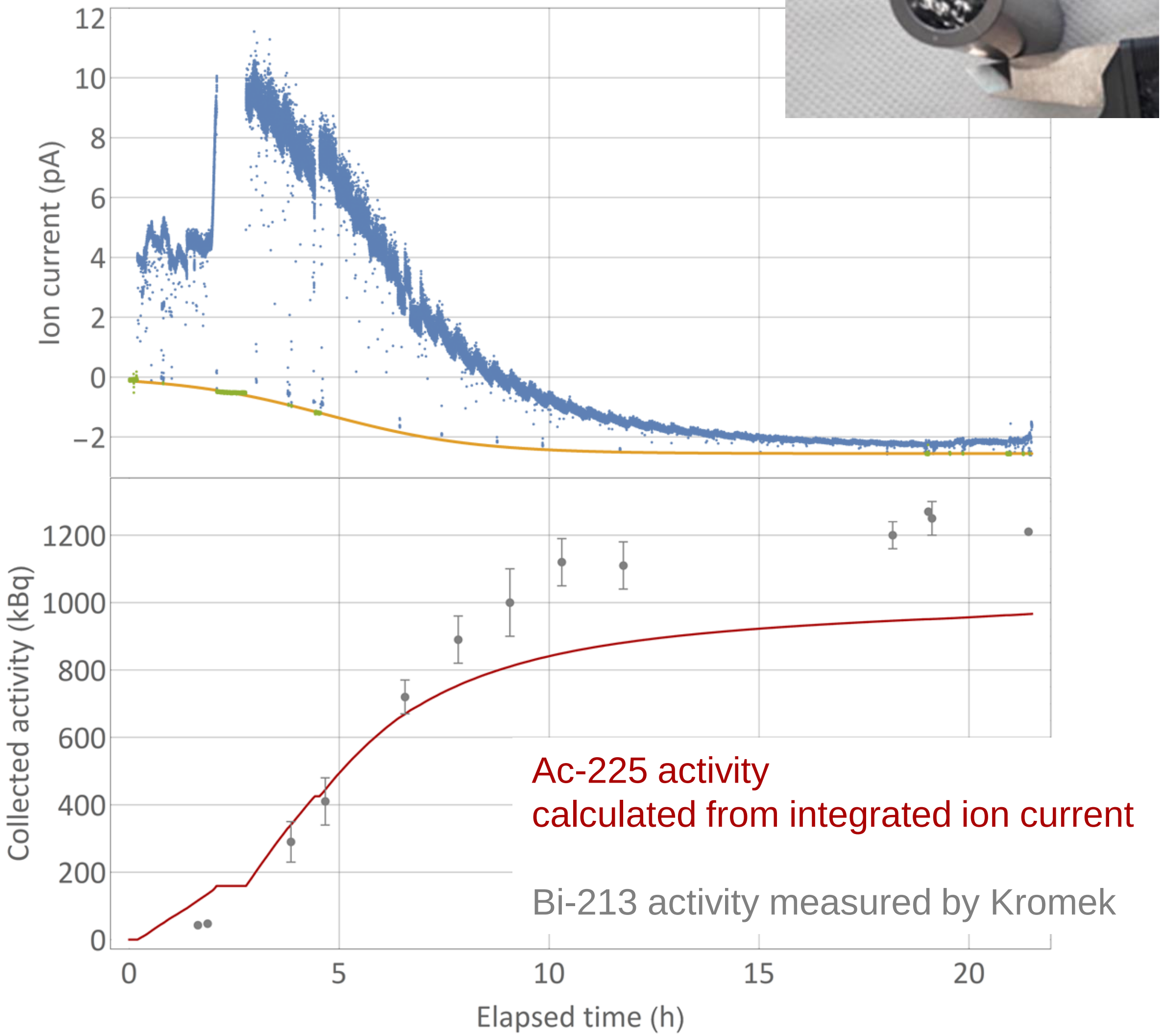
Ionization efficiency studies

Hg in VADIS



Ac in RILIS





Hg: Y. Martinez Palenzuela, PhD Thesis, KU Leuven (2019).
Ac: courtesy Jake Johnson, KU Leuven.

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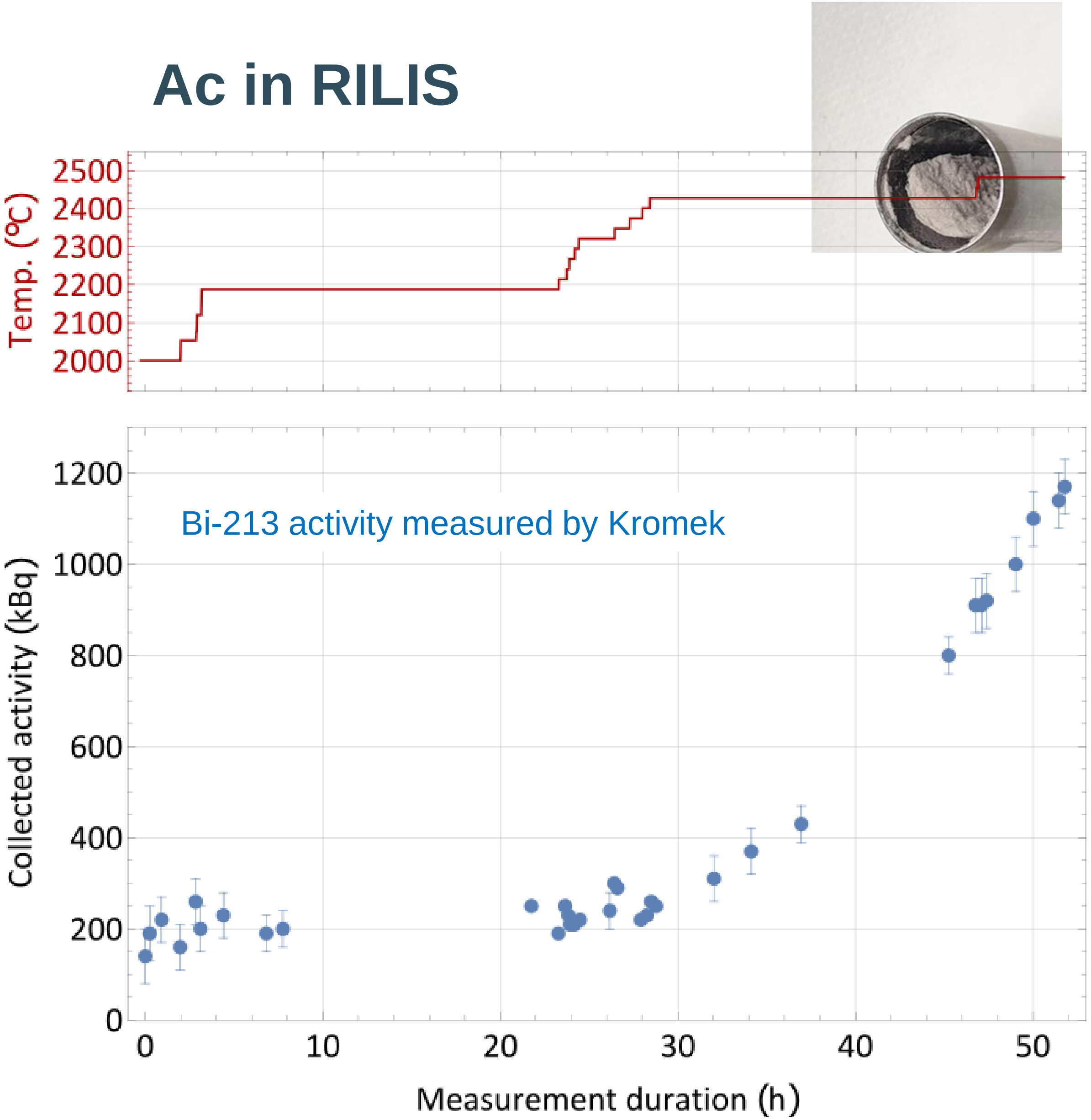
$\varepsilon = \varepsilon_{eff}\varepsilon_{ion}$

Effusion studies

- There are two types of effusion:
  - Through the pores of the target material
  - Through the target and transfer line volume to the ion source
- Effusion is based on a sequence of landing on a hot surface followed by re-emission in a random direction.
- The delays are a question of how long the element sticks to the material it lands on and how far it can travel before landing again.

Ac in RILIS



 Ac: courtesy Jake Johnson, KU Leuven.

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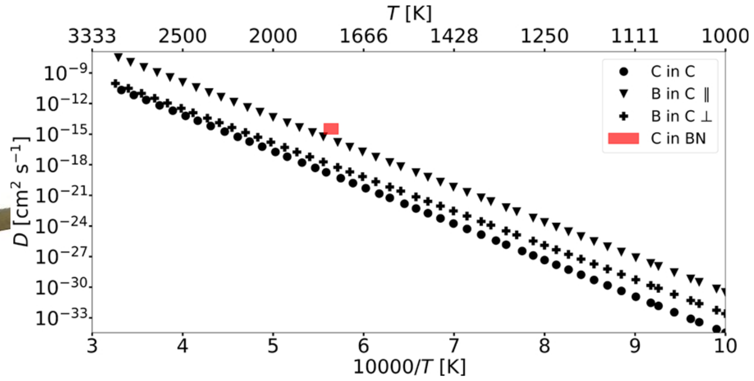
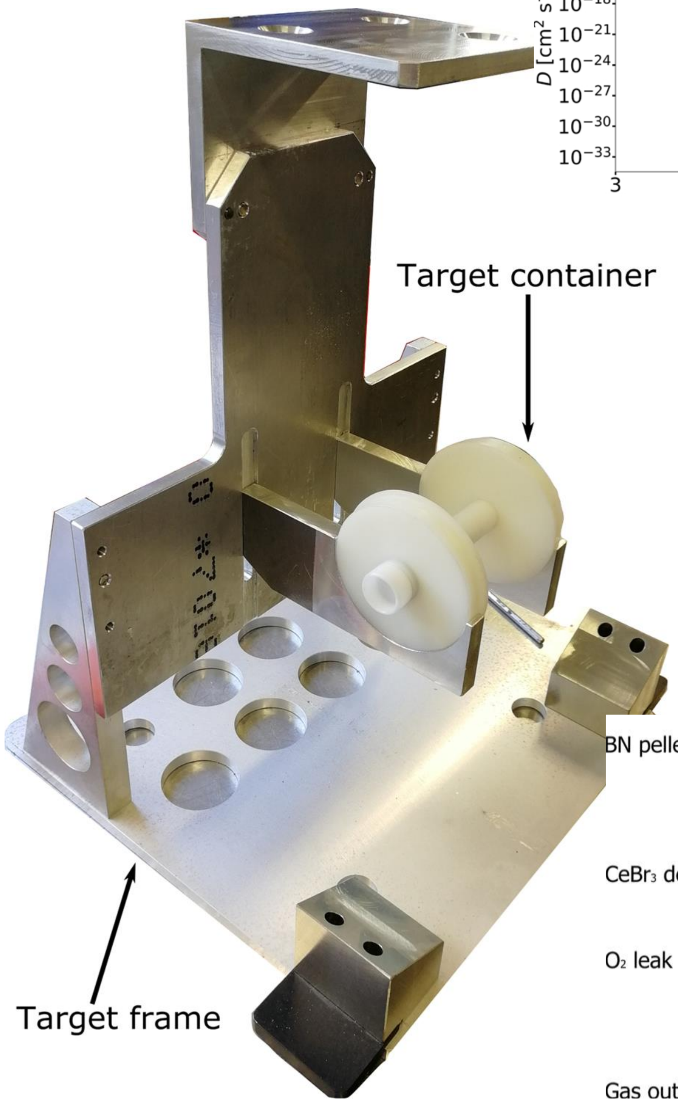
$\varepsilon = \varepsilon_{diff}$

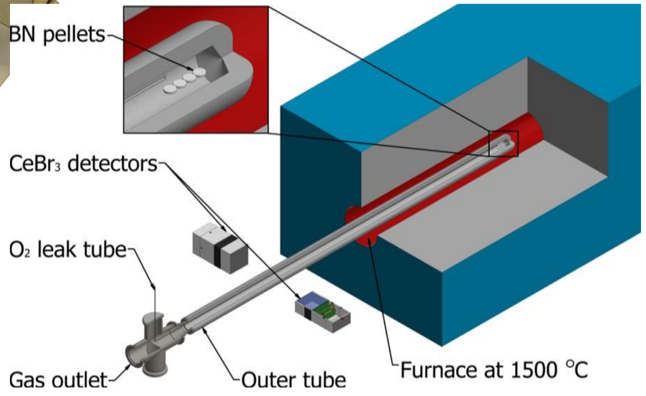
Diffusion studies

- Step 1: produce a known quantity of radioisotopes within the target material
  - ❑Based on simulation codes like FLUKA
  - ❑By measuring the inventory right after irradiation
- Step 2: perform a heat treatment of the target
- Step 3: control what has been released
  - ❑By direct measurement of the exhaust
  - ❑By post-treatment measurement of the inventory

CO out of BN





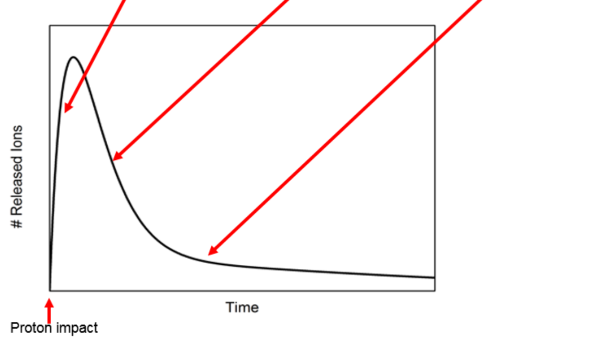


S. Stegemann et al., ... for the production and release of radioactive 11C, Journal of the European Ceramic Society ***41*** *(2021)*

$$\varepsilon = \varepsilon_{diff}\varepsilon_{eff}\varepsilon_{ion}$$

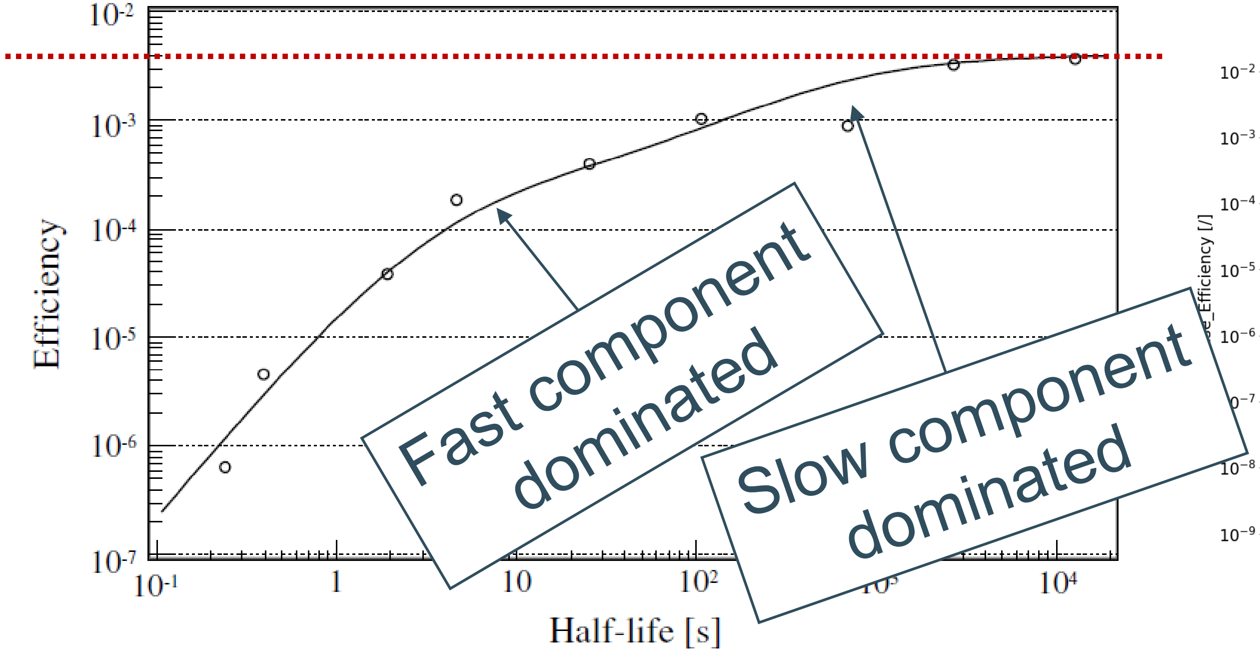
Full analysis of isotope release

$$P_i(t,\lambda_i) = \exp(-\lambda_i t) \cdot \frac{[1-\exp(-\lambda_r t)] \cdot [\alpha \cdot \exp(-\lambda_f t) + (1-\alpha) \cdot \exp(-\lambda_s t)]}{\text{Normalisation factor}}$$

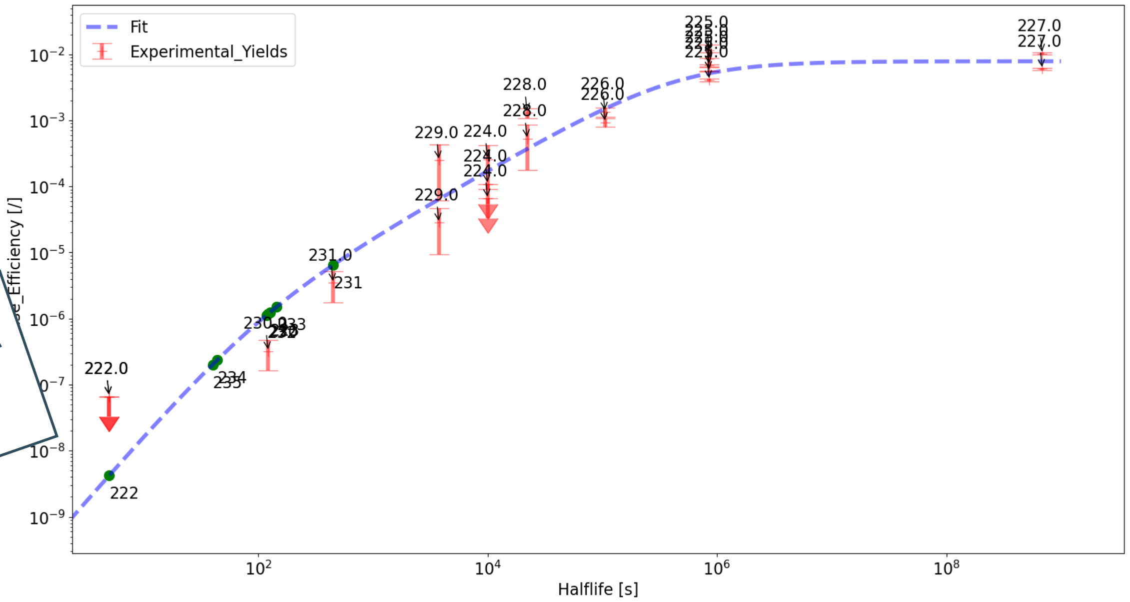


Polonium release

Asymptotic to ionization efficiency



Actinium release



Po: T.E. Cocolios et al., *Resonant laser ionization of polonium ..., NIMB **266** (2008) 4403-4406.*
Ac: courtesy Kristof Dockx, KU Leuven.

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- ✓ We have discussed what sort of targets are used in ISOL facilities and how they are taylored and developped. There are so many, it would be impossible to list them all in this short lecture!
- ✓ We have discussed ion sources and how these are also adapted to each element of interest. In particular, resonant ionization gives access to high selectivity. Each ion source requires also extensive investigation, which are ongoing, sometime closer than we think!
- ✓ The beam distribution is then determined by the different parameters from the primary beam, the target material, the ion source, the element of interest, the isotope half-life, ...
- ✓ The beam can be manipulated upon to deliver the best solution to each experiment in terms of time structure, beam emittance or beam energy.

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To be continued in the lecture series

15 Nov 2021

3 – Fundamental research with RIB

- Studying the nuclear forces by challenging nuclear models
  - Nuclear structure
  - Physics beyond the Standard Model
- Exploring the limits of existence
  - ^{28}O, the unbound doubly magic nucleus
  - Proton-unbound systems
  - The path to superheavy elements
- Nuclear physics in the stars

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