Lecture 2: Production of radioactive ion beams

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Outline of lecture 2

- Historical development of the nuclear chart
- Overview of our radioactive ion beam toolbox
- The ISOL method
- The in-flight method
- The ion guide/gas cell technique

Development of the nuclear chart

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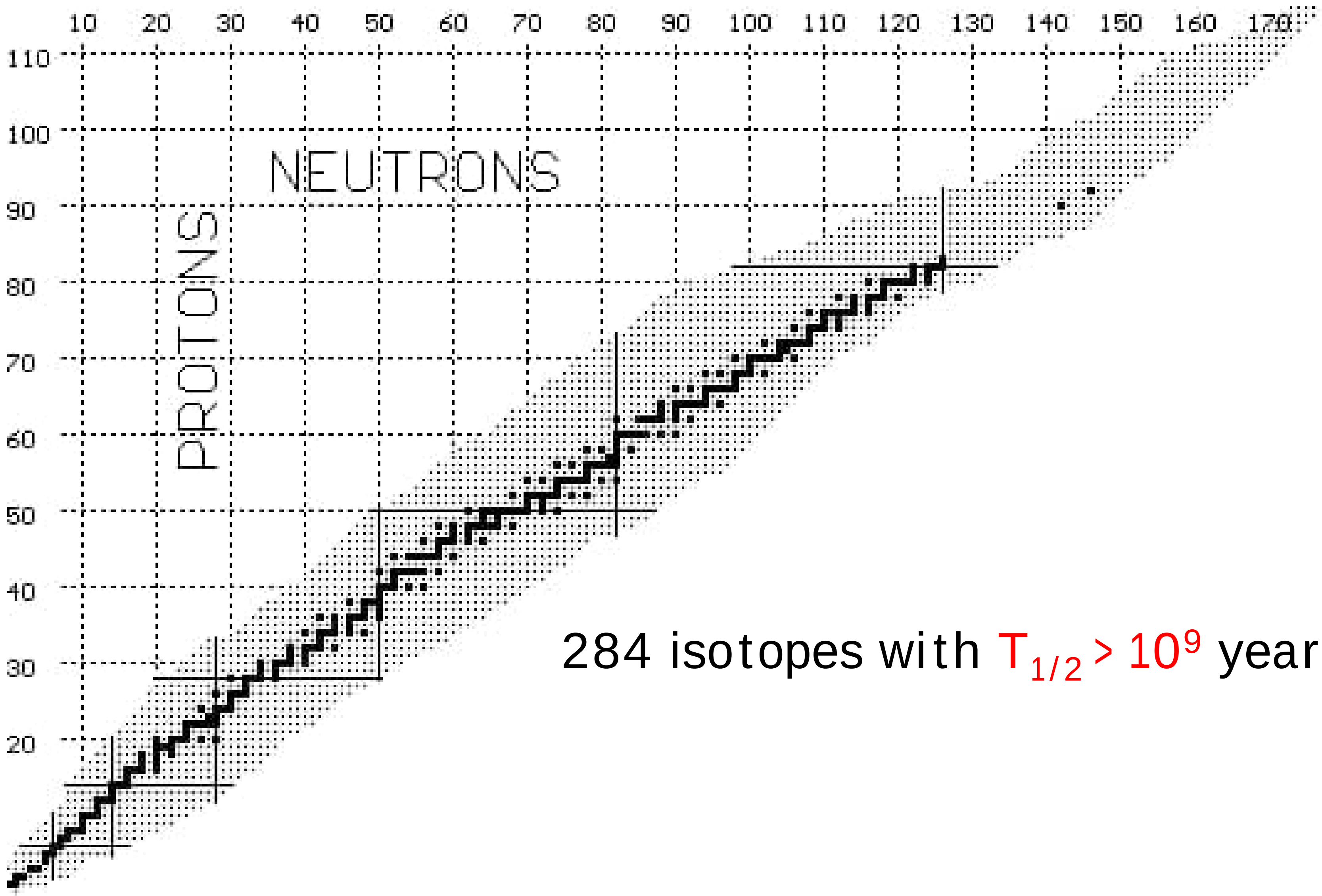
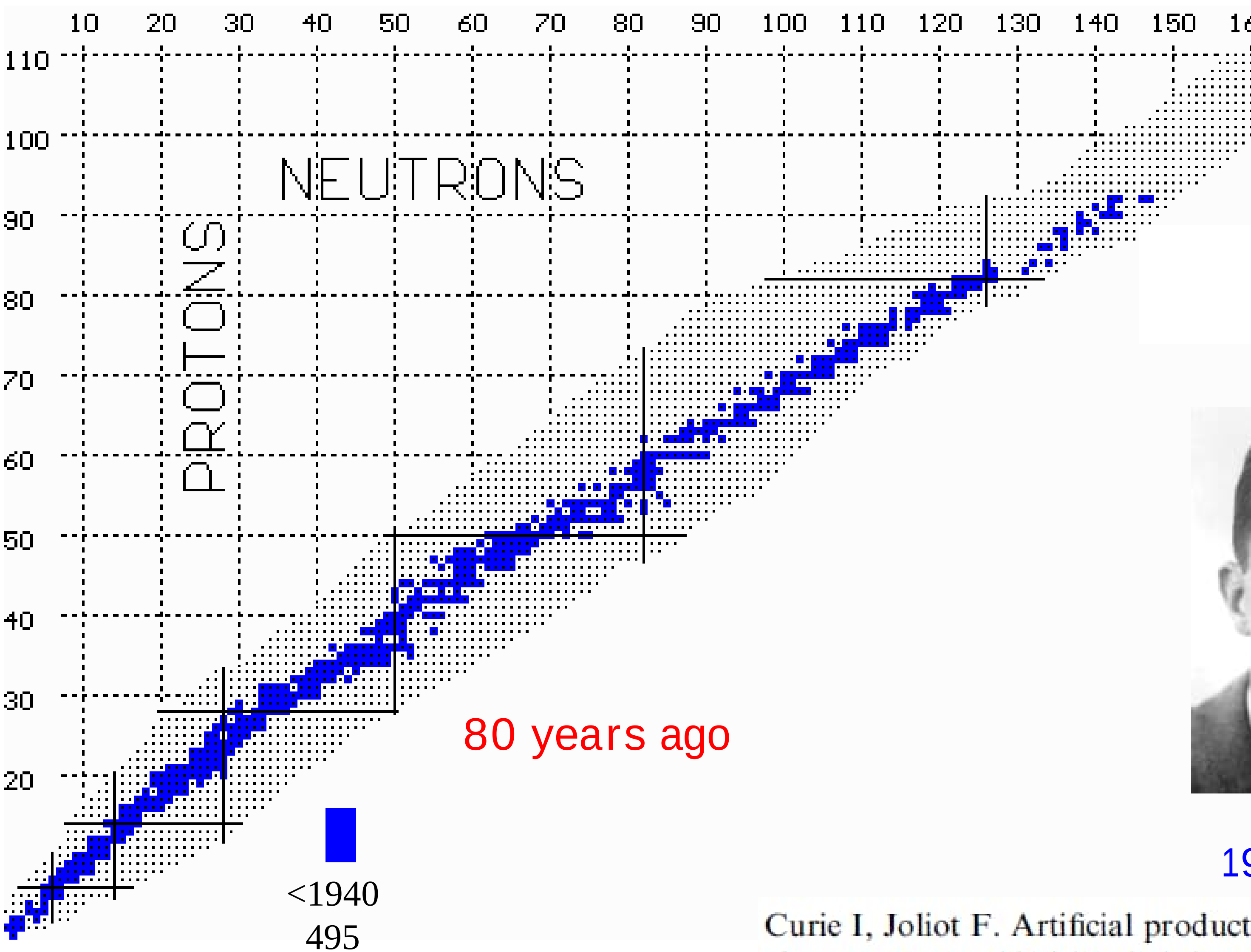


284 isotopes with $T_{1/2} > 10^9$ year

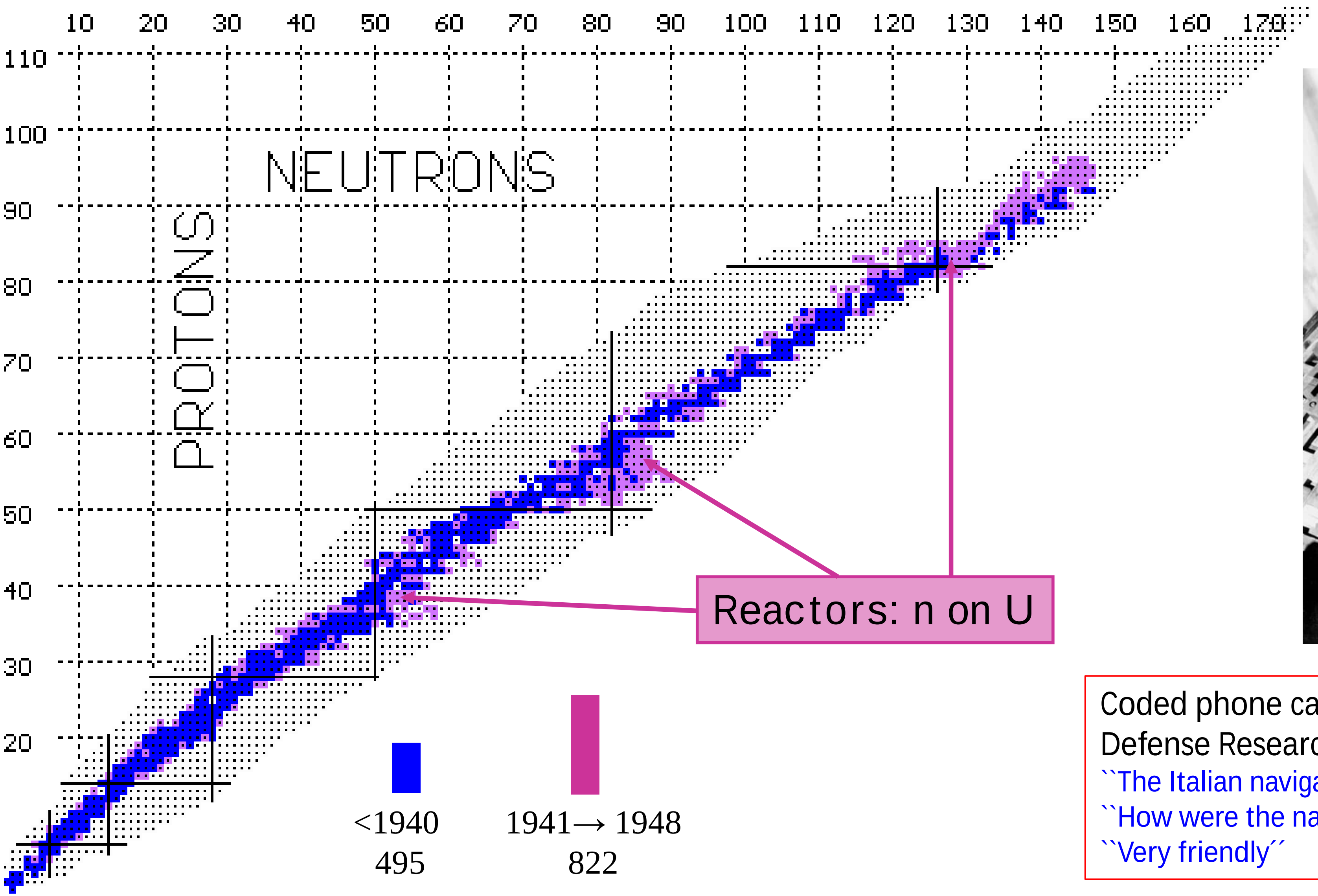
The discovery of radioactivity



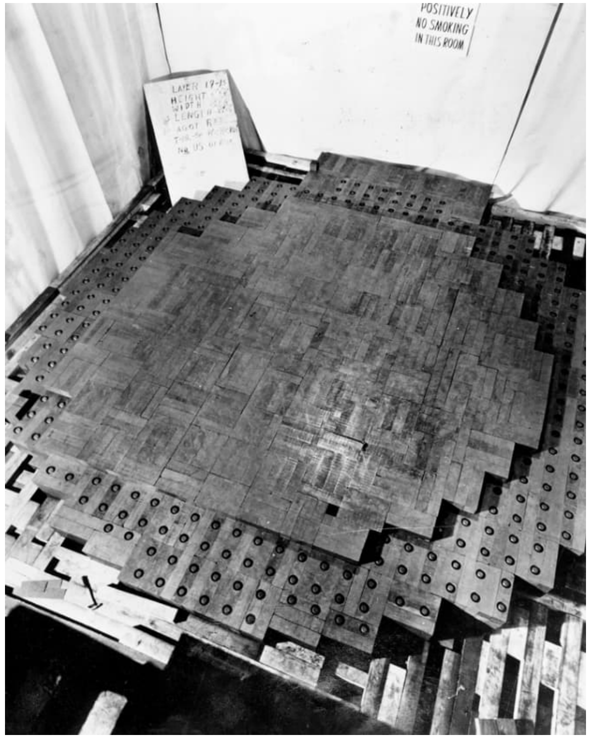
1935: Nobel Prize in Chemistry

Curie I, Joliot F. Artificial production of a new kind of radioactive element. Nature 1934;133:201–2.

+ the advent of nuclear reactors



Chicago Pile-1 (1942)



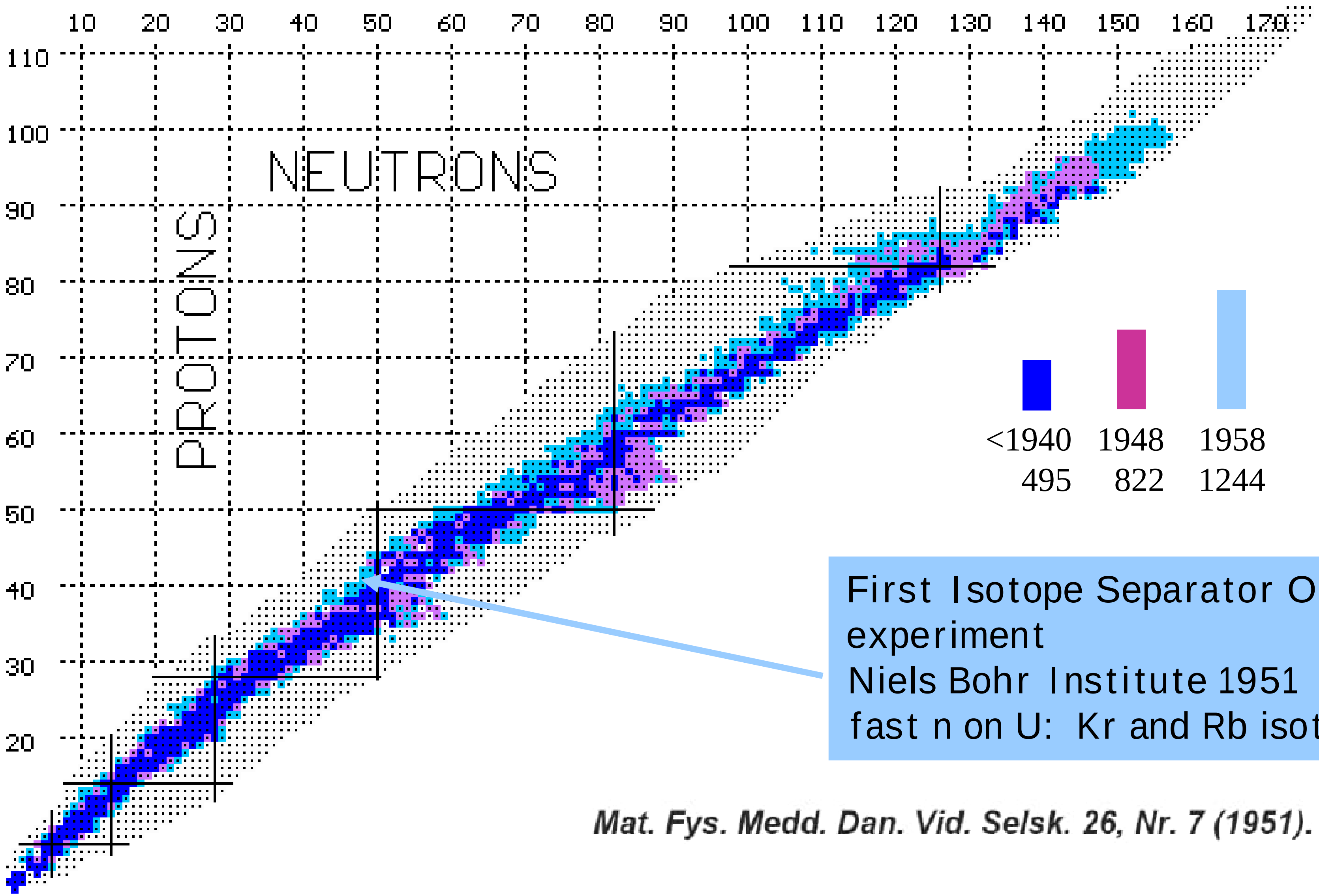
Coded phone call to the Chairman of the National Defense Research Committee:

``The Italian navigator has landed in the New World´´

``How were the natives?´´

``Very friendly´´

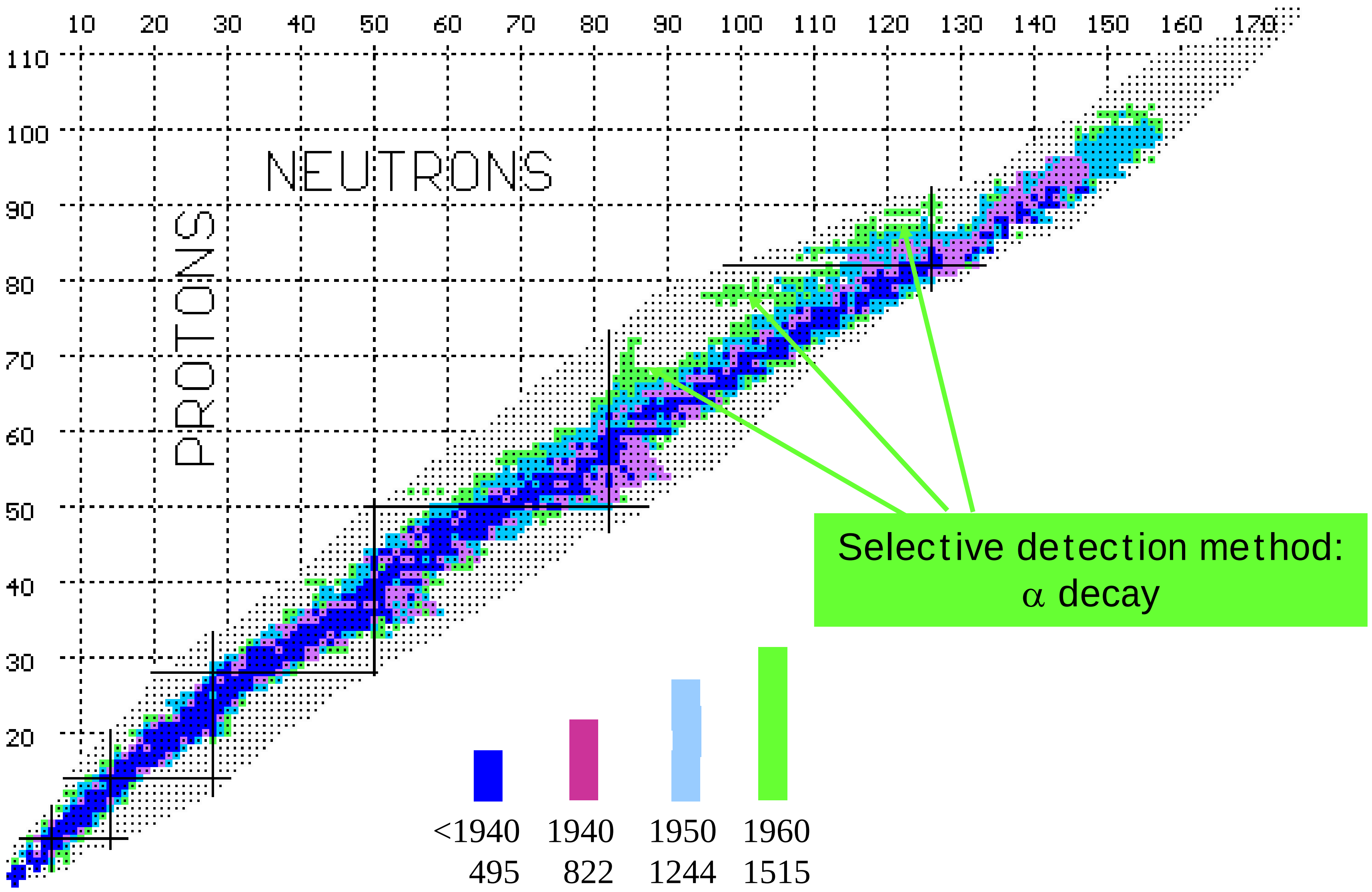
+ early Isotope Separator On-Line (ISOL) isotopes



First Isotope Separator On-Line (ISOL) experiment
Niels Bohr Institute 1951
fast n on U: Kr and Rb isotopes

Mat. Fys. Medd. Dan. Vid. Selsk. 26, Nr. 7 (1951).

+ sensitive detection methods



+ energy increases and driver beam upgrades

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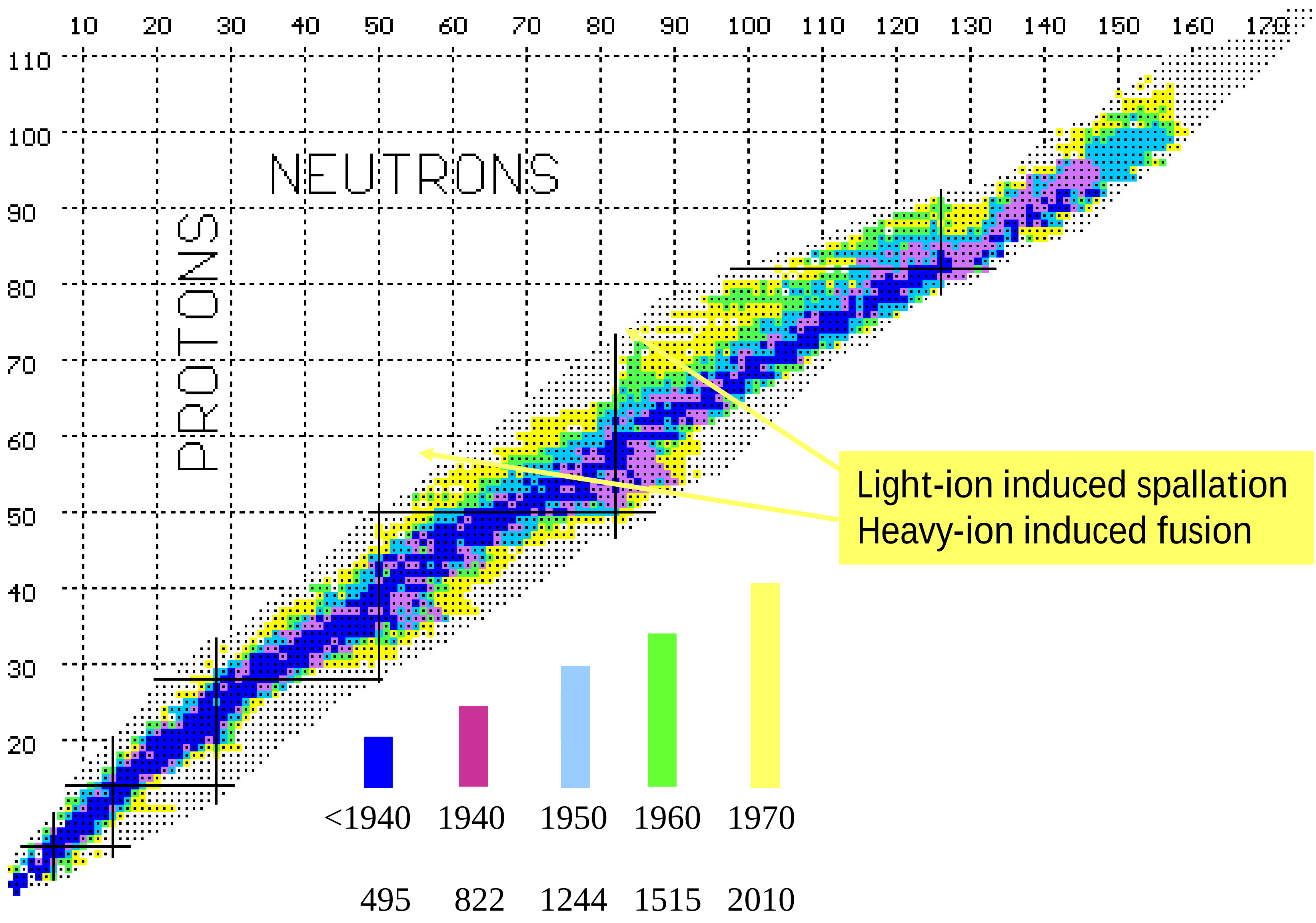

+ thin target and projectile fragmentation

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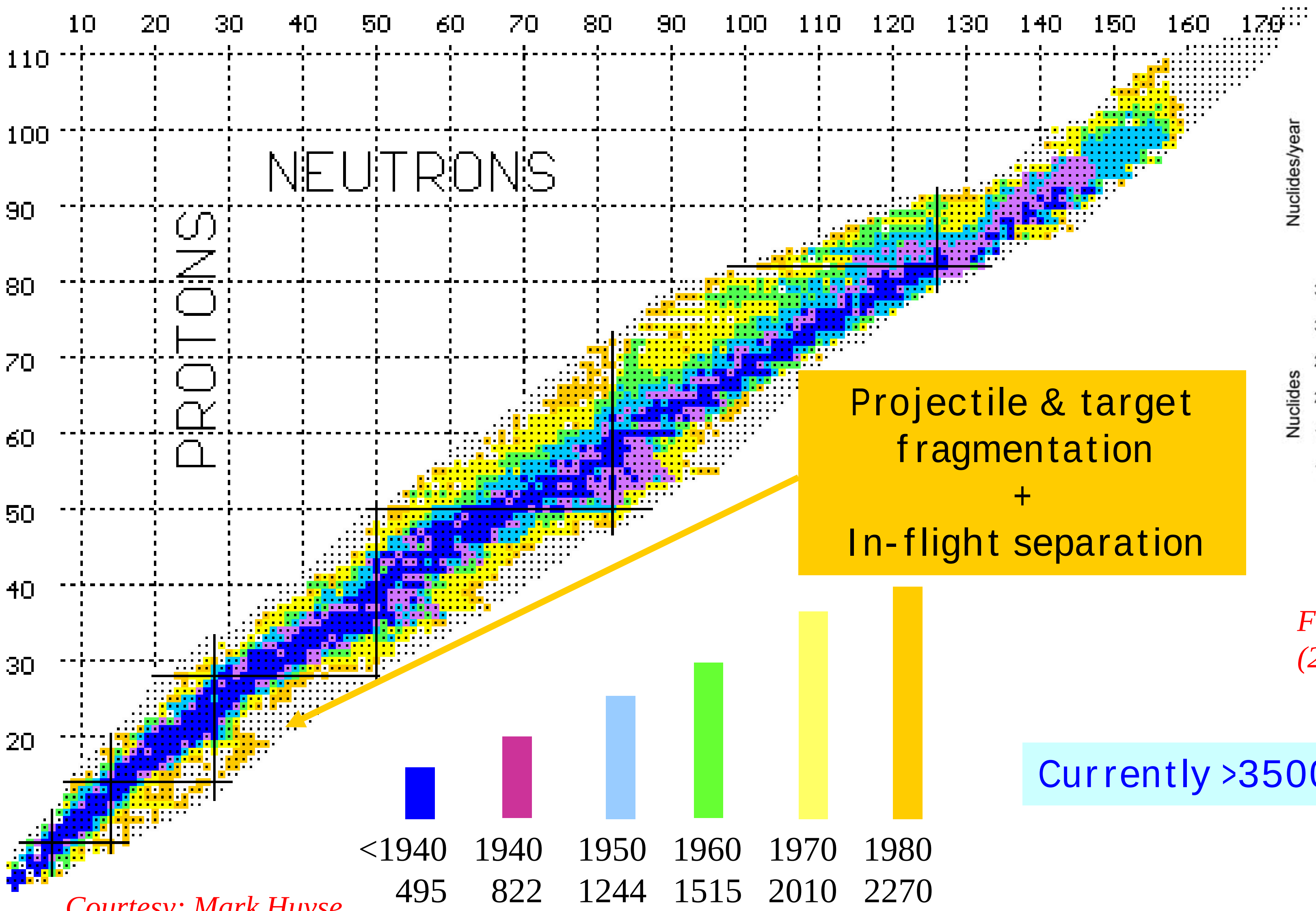

Courtesy: *Mark Huyse*

Projectile & target fragmentation
+
In-flight separation

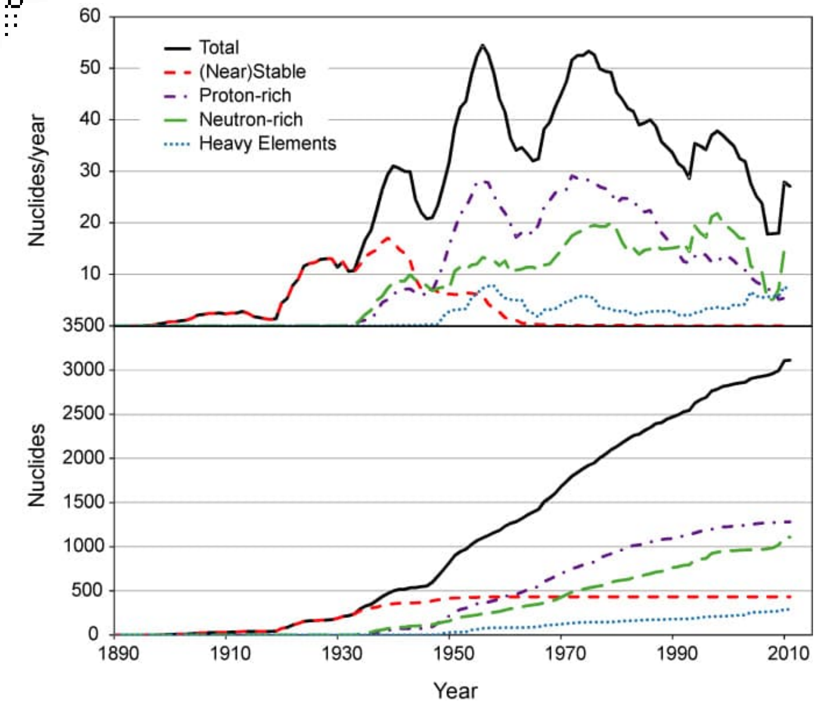

Figure from: M. Thoennessen, Rep. Prog. Phys. 76 (2013) 056301

Currently >3500 nuclei experimentally observed

A radioactive ion beam (RIB) toolbox

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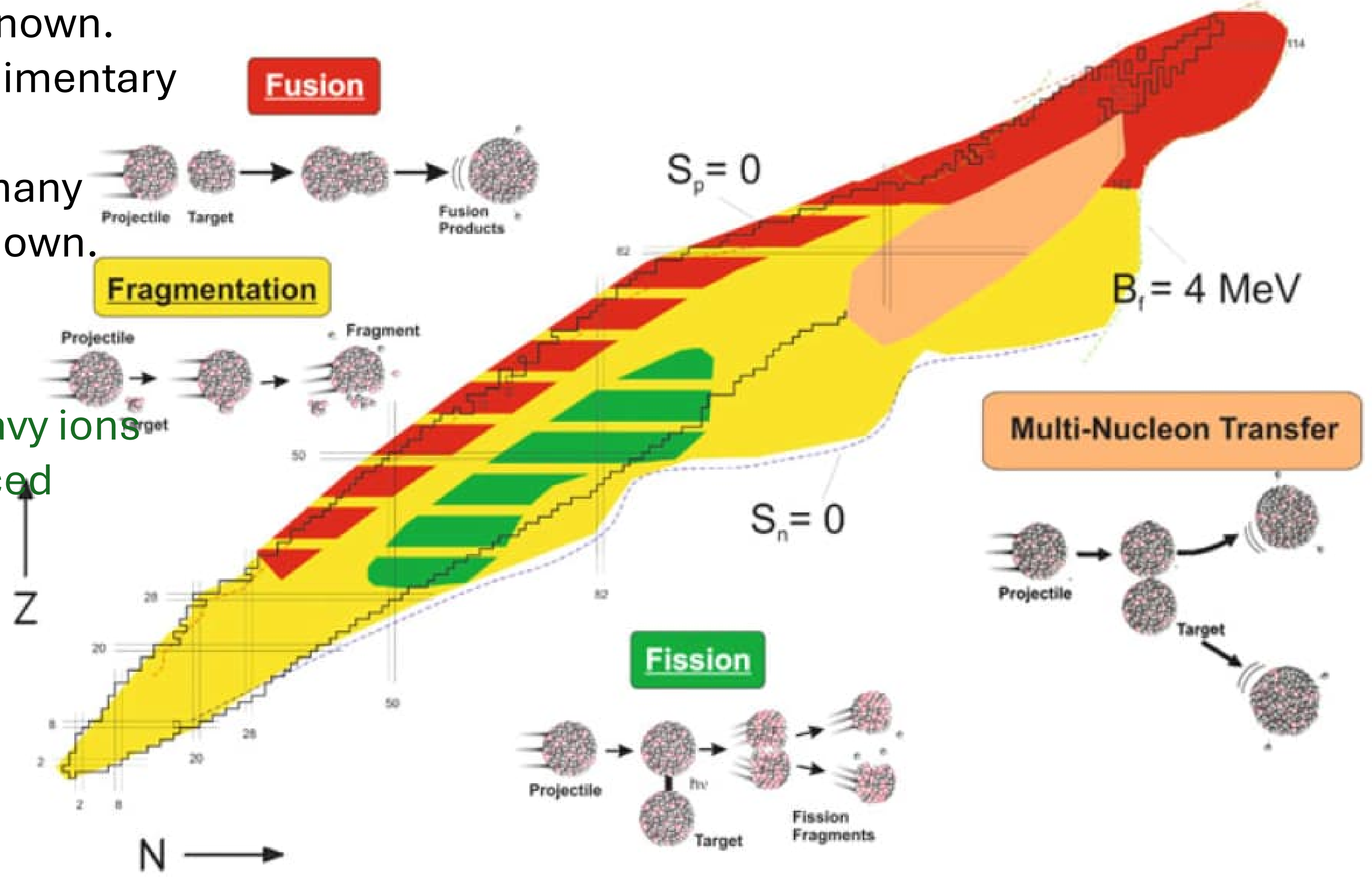
- Most isotopes predicted to exist are not known.
- Many known exotic isotopes have only rudimentary studies.
- The proton drip line has been reached in many cases; the neutron drip line is largely unknown.



Primary nuclear reaction:

- Fragmentation: high energy protons or heavy ions
- Fission: proton, neutron and photon induced
- Spallation: high energy protons
- Fusion: heavy- and light-ion induced

Lecture Notes on Physics: 651 (2004), 700 (2006), 764 (2008), Springer Verlag Berlin

- *"In-flight separation of projectile fragments", D.J. Morrissey & B.M. Sherrill*
- *"Isotope separation on line and post acceleration", P. Van Duppen*
- *"Spallation reactions…", J. Benlliure*

Figure from: Exotic Nuclei and their Separation, Electromagnetic Devices. H. Geissel and D.J. Morrissey, Handbook of Nuclear Physics, Springer Nature 2023.

Discussion pause: audience input please!!

Why do you think ”pure” radioactive ion beams might be important?

Production of pure radioactive beams

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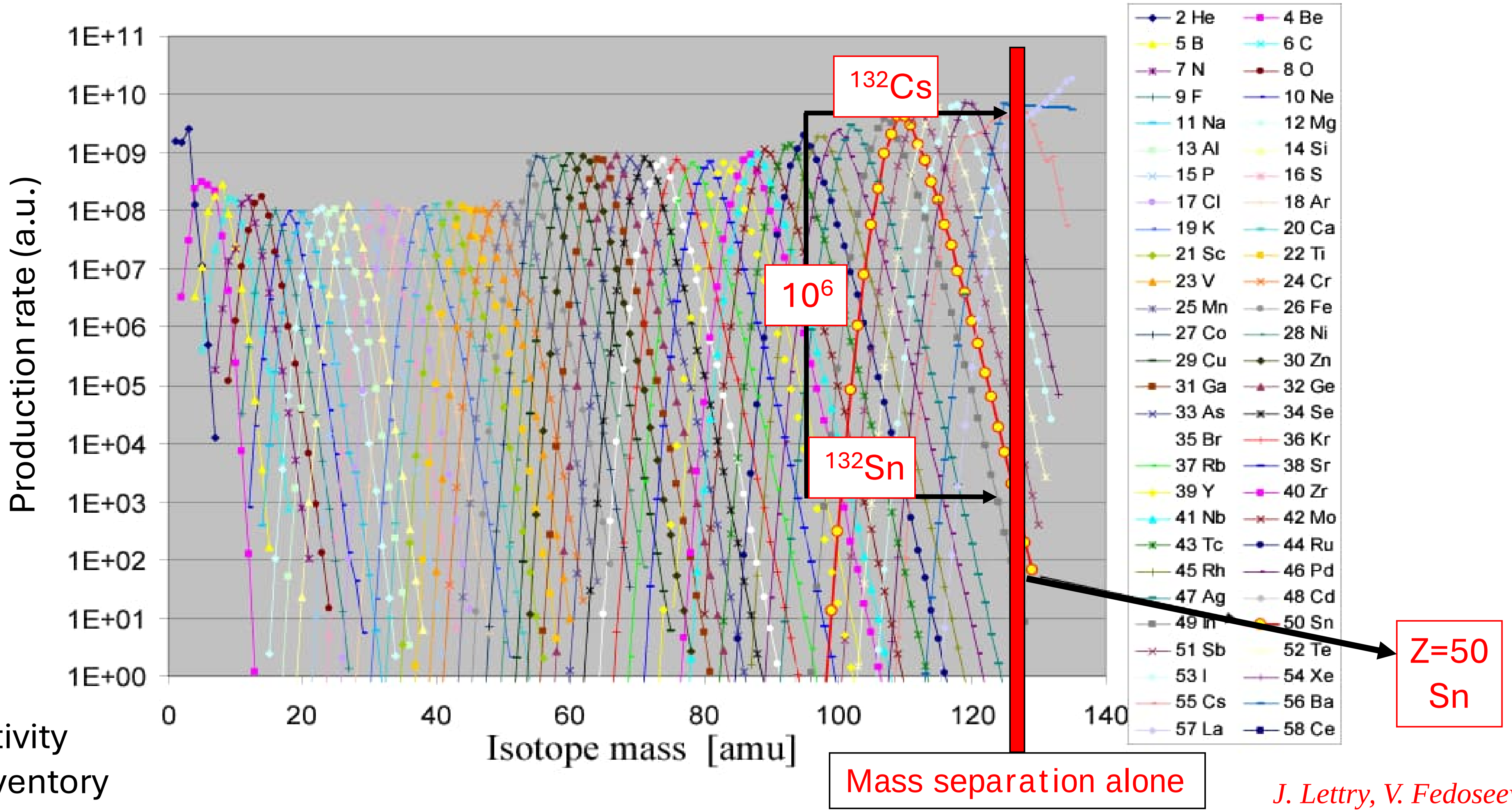
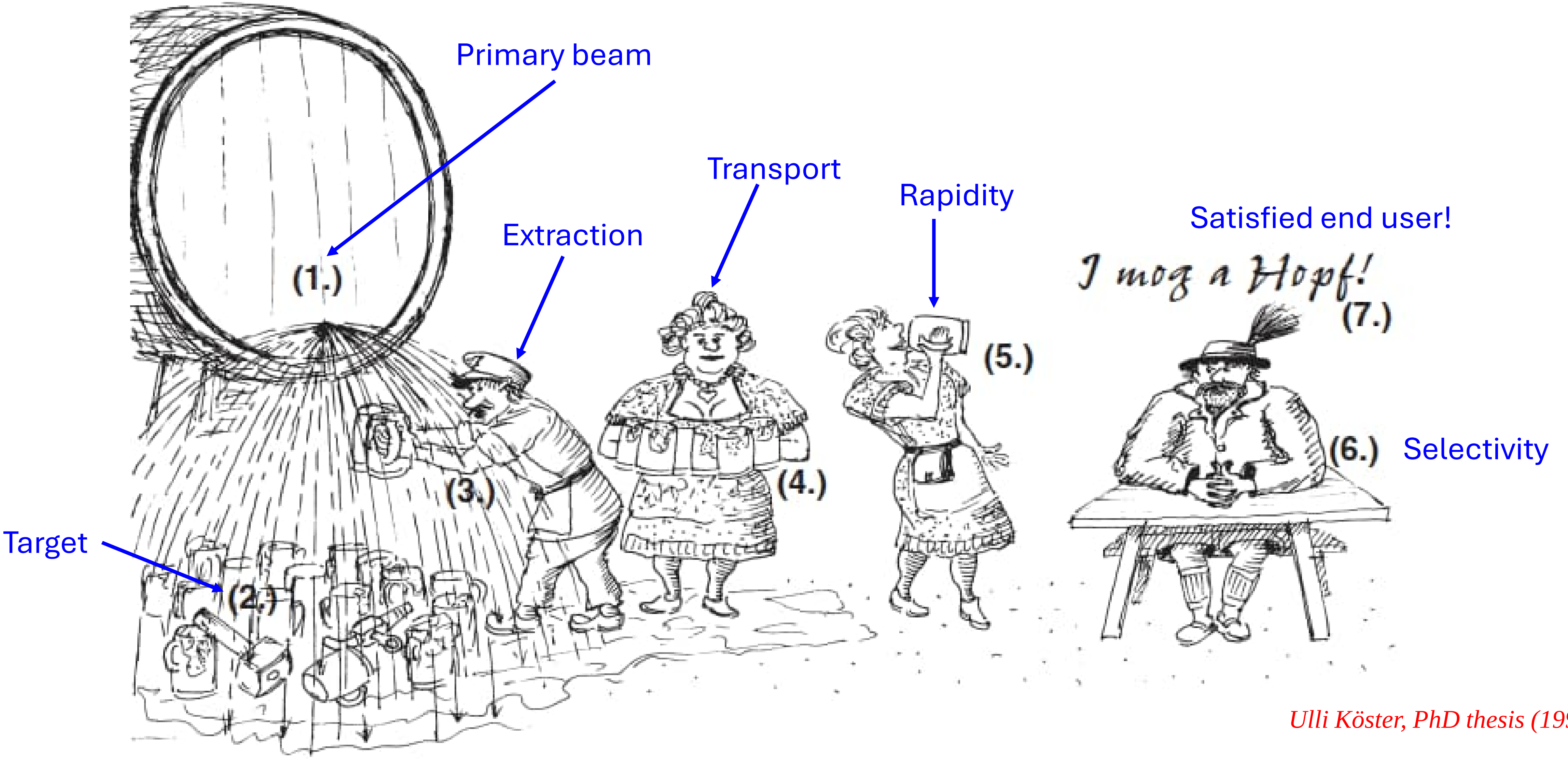
Isotope production for a 1 GeV p beam on a La target



- Element selectivity
- Radioactive inventory

J. Lettry, V. Fedoseev (CERN)

Schematic representation of a RIB facility



Ulli Köster, PhD thesis (1999)

Figures of Merit for RIB production

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High production rate:

- Cross section is given to us by Nature
- Optimize the beam/target combination
- Available beams (accelerators, reactors)
- Power deposition in targets – radioactive inventory

Efficient:

- Production rate of very exotic nuclei is always small

Fast:

- Exotic nuclei often have short half-lives

Selective:

- In most cases, unwanted contaminants are produced in copious amounts

We want to maximize

✓ Intensity

✓ Selectivity

✓ Sensitivity

The Isotope Separation On-Line (ISOL) method

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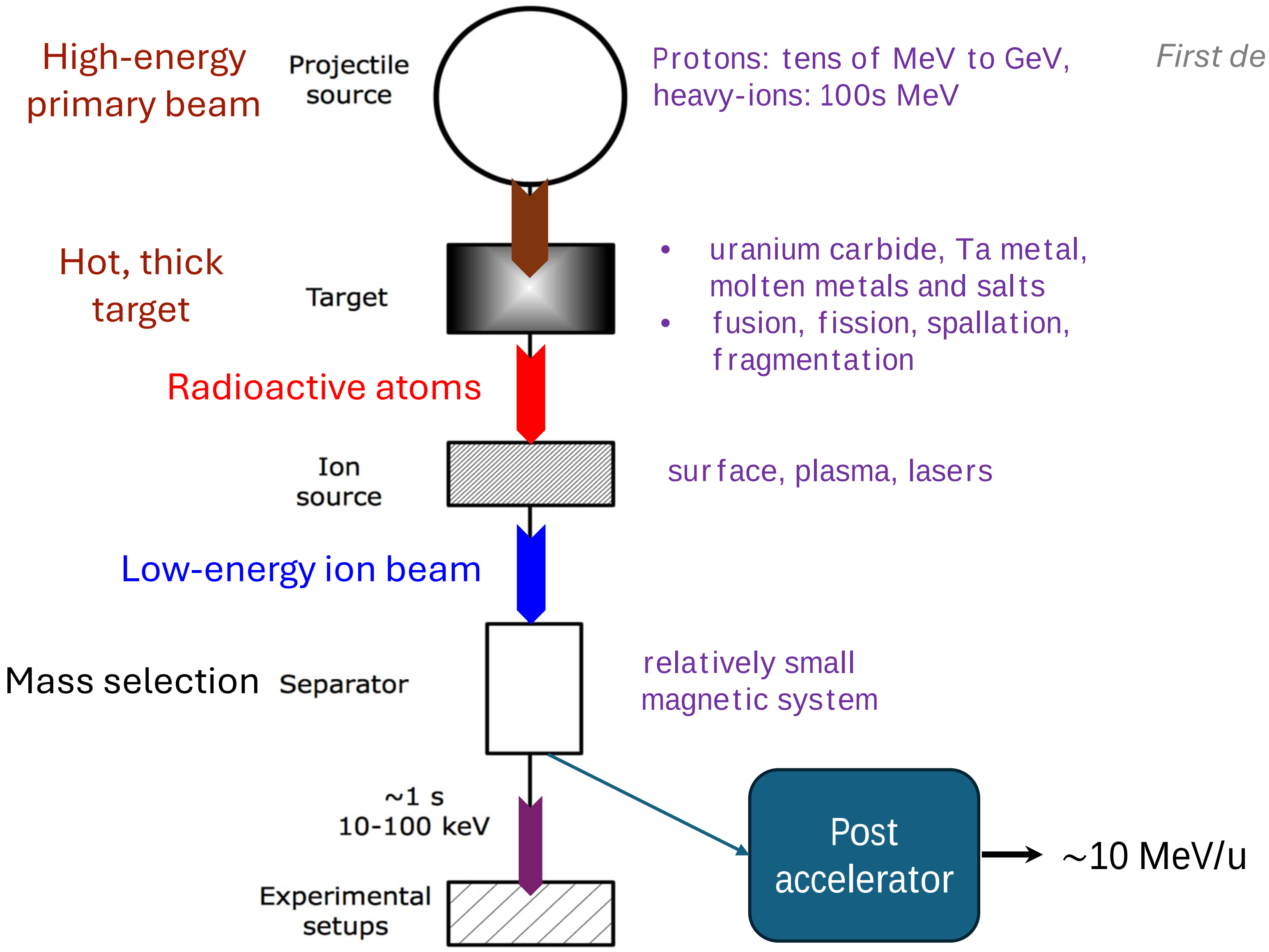


First developed in 1951, Niels Bohr Institute, Denmark

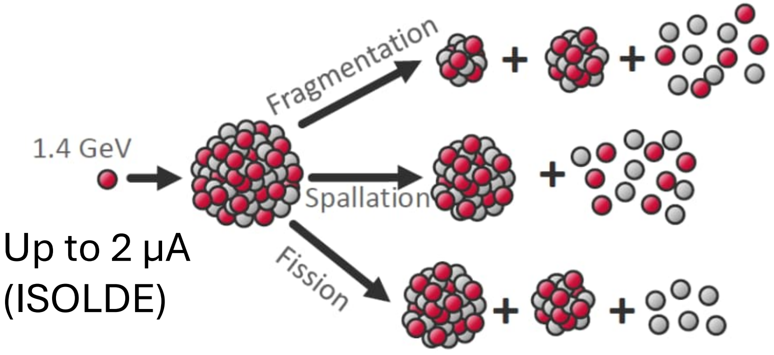
<u>Some key properties:</u>

- Excellent beam properties
- Chemical sensitivity (*Z* dependence)
- Half-lives > typically few ms

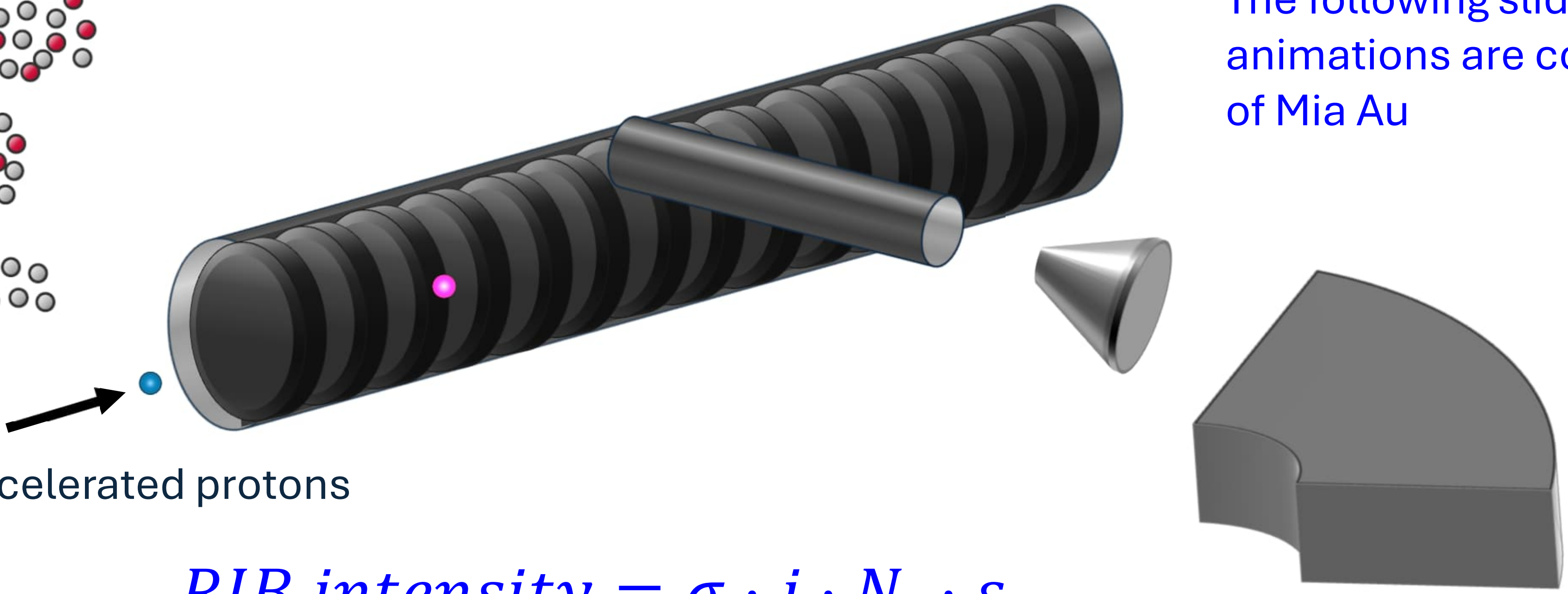
<u>Discovery potential</u>:
precision studies of exotic nuclei

<u>Example facilities</u>:
ISOLDE 1.4 GeV protons
TRIUMF-ISAC 600 MeV protons
INFN-SPES and RAON-Korea 70 MeV protons
SPIRAL/DESIR–GANIL heavy-ions 25 MeV/A (fusion etc.)

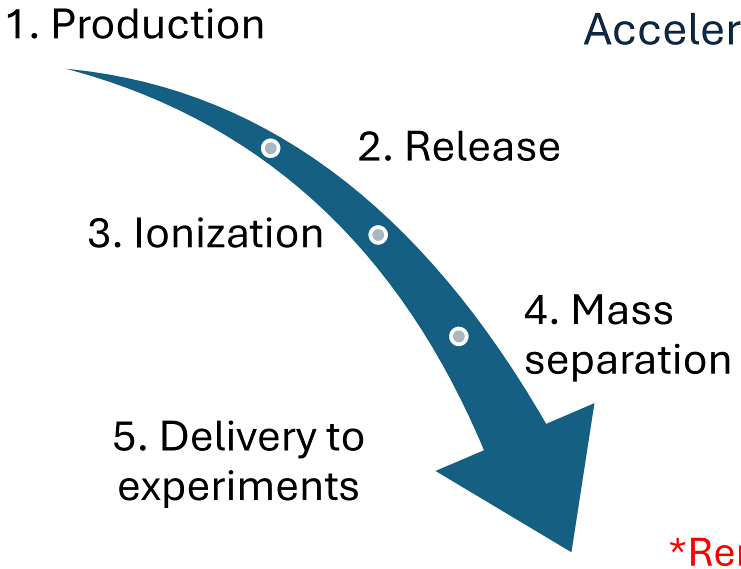
How does the ISOL method work?



Up to 2 μA (ISOLDE)

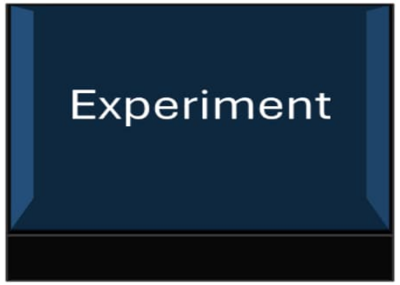


The following slide animations are courtesy of Mia Au



$$RIB\ intensity = \sigma \cdot j \cdot N_t \cdot \varepsilon$$

N_t – Number of target atoms
j – Proton flux [cm^{-2}]
σ – Cross section [b]*
ε – Efficiency [%]



*Remember 1 barn = 10^{-24} cm^2 (probability to produce a particular nucleus)

How does the ISOL method work?

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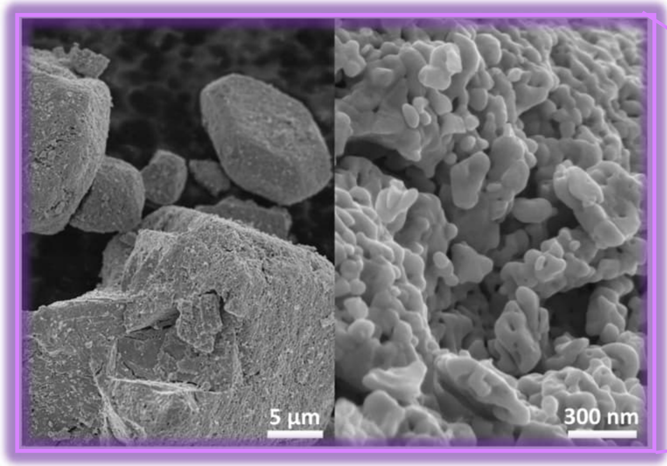

Microstructure of raw UO_2 material used at ISOLDE to produce UC_x targets

J.P. Ramos, Review of thick solid targets, NIMB 463 (2020) 201

Accelerated protons

1. Production
2. Release
3. Ionization
4. Mass separation
5. Delivery to experiments

$$RIB\ intensity = \sigma \cdot j \cdot N_t \cdot \varepsilon$$

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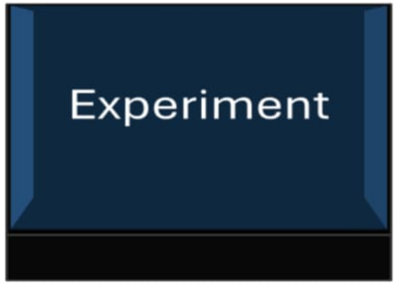

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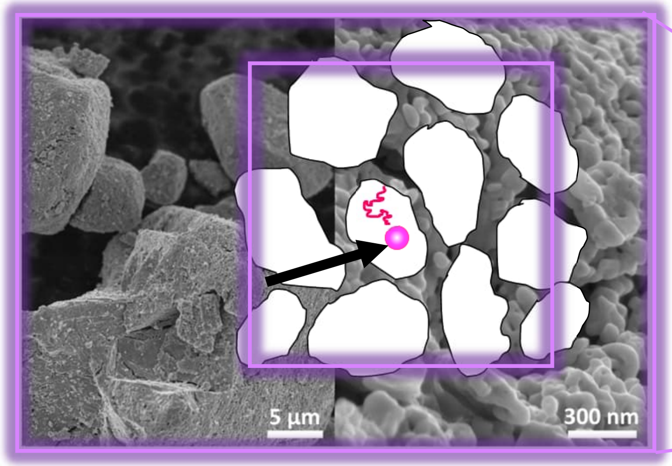

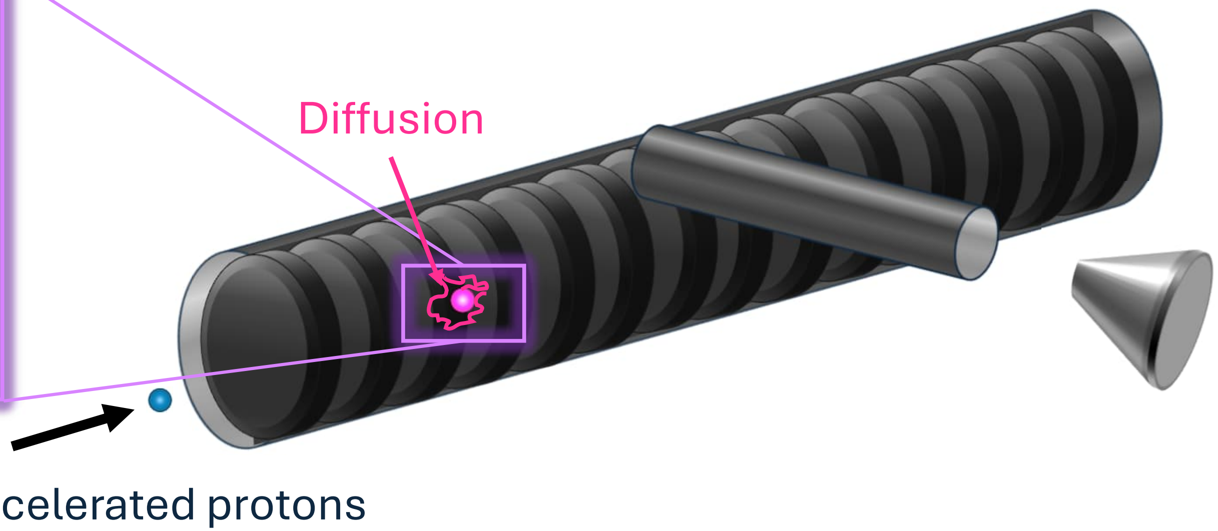

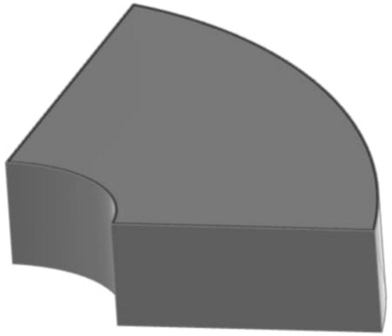

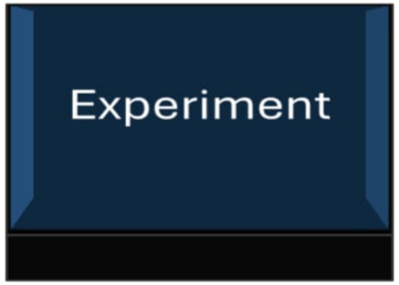

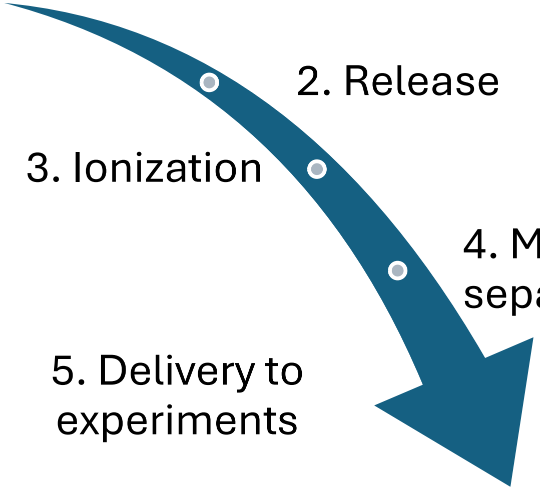

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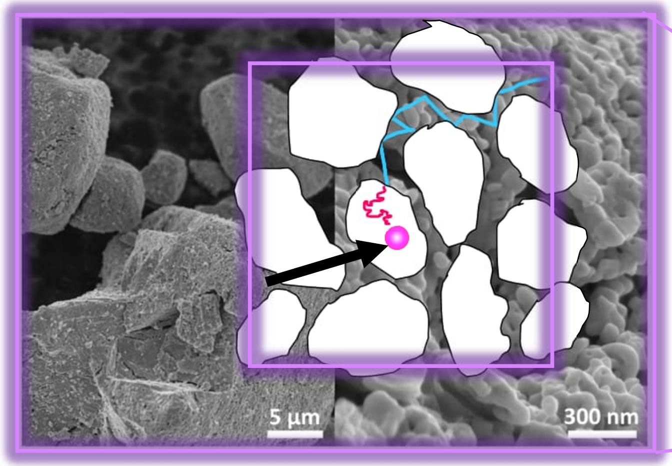
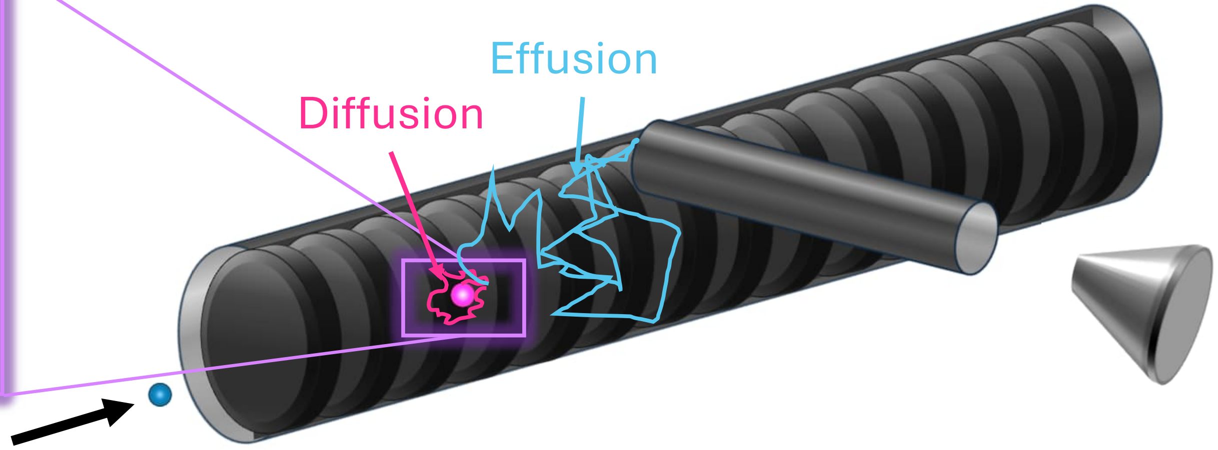
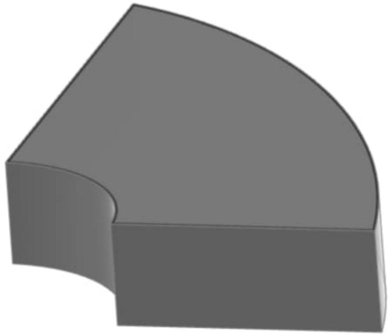
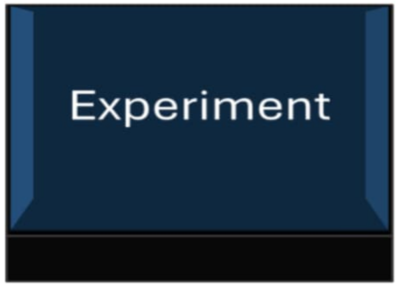




Accelerated protons





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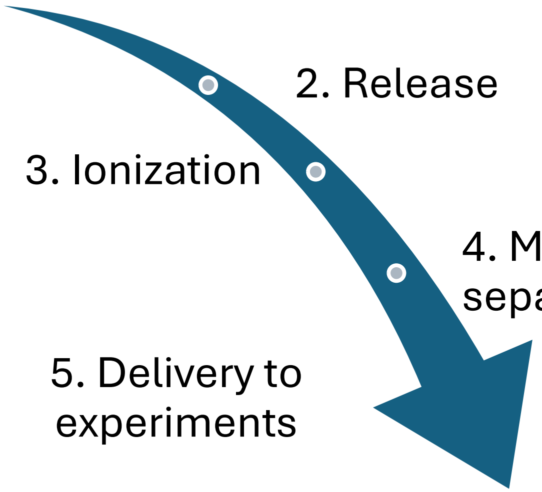
2. Release
3. Ionization
4. Mass separation
5. Delivery to experiments



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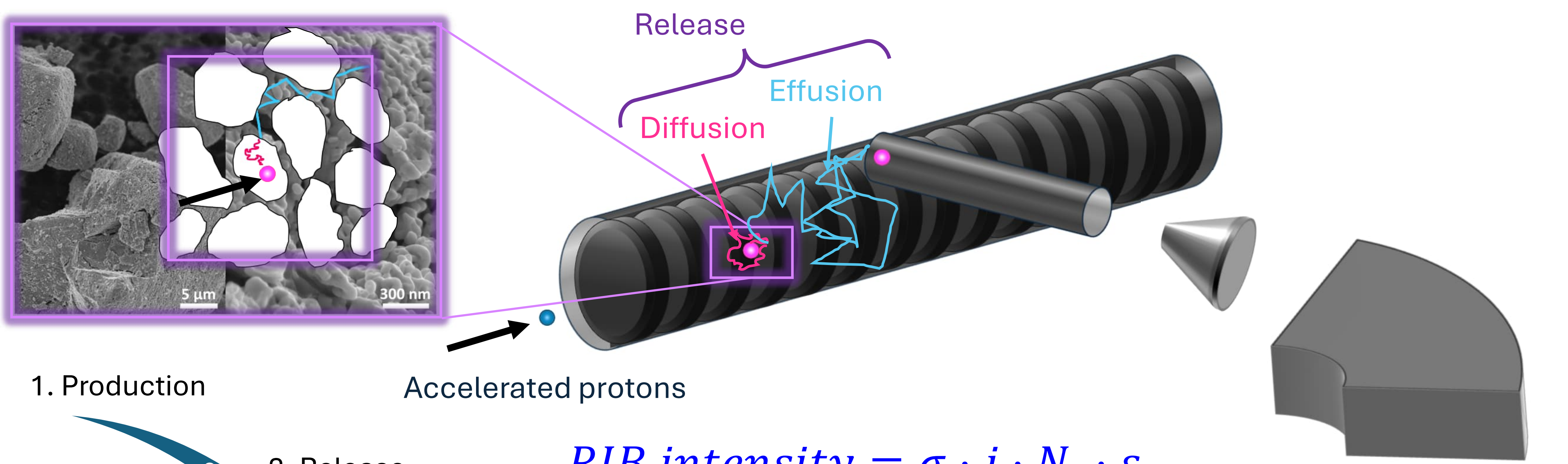

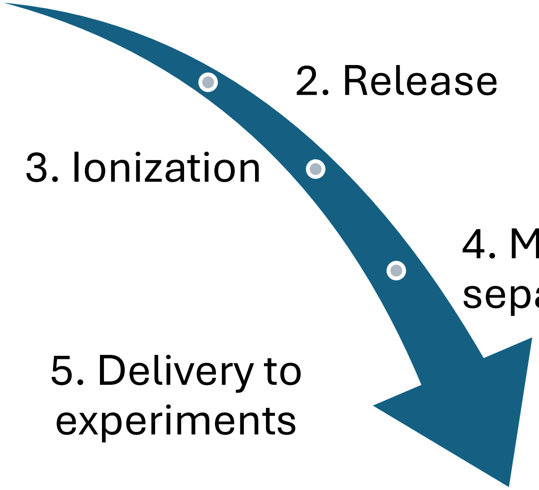

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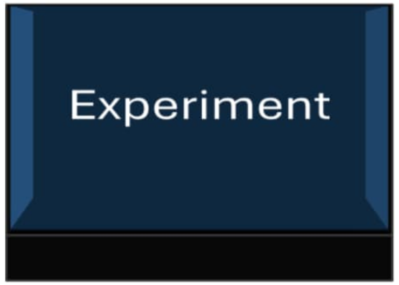
$$\varepsilon = \varepsilon_{diff}\varepsilon_{eff}$$



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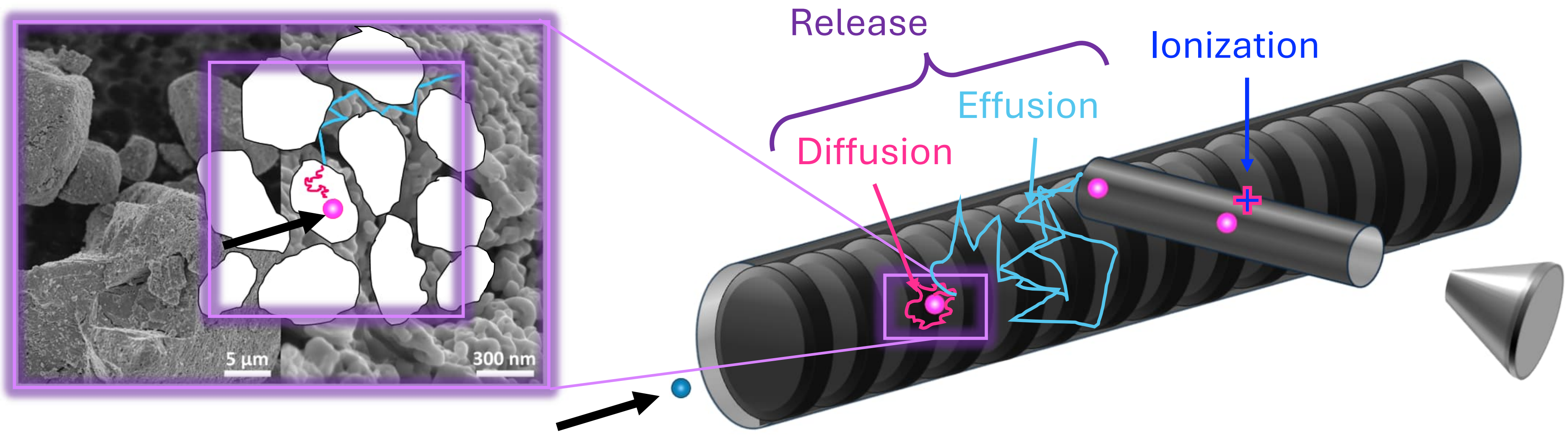

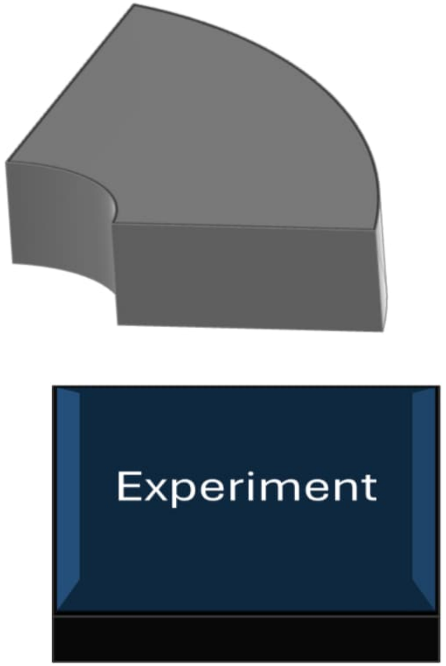

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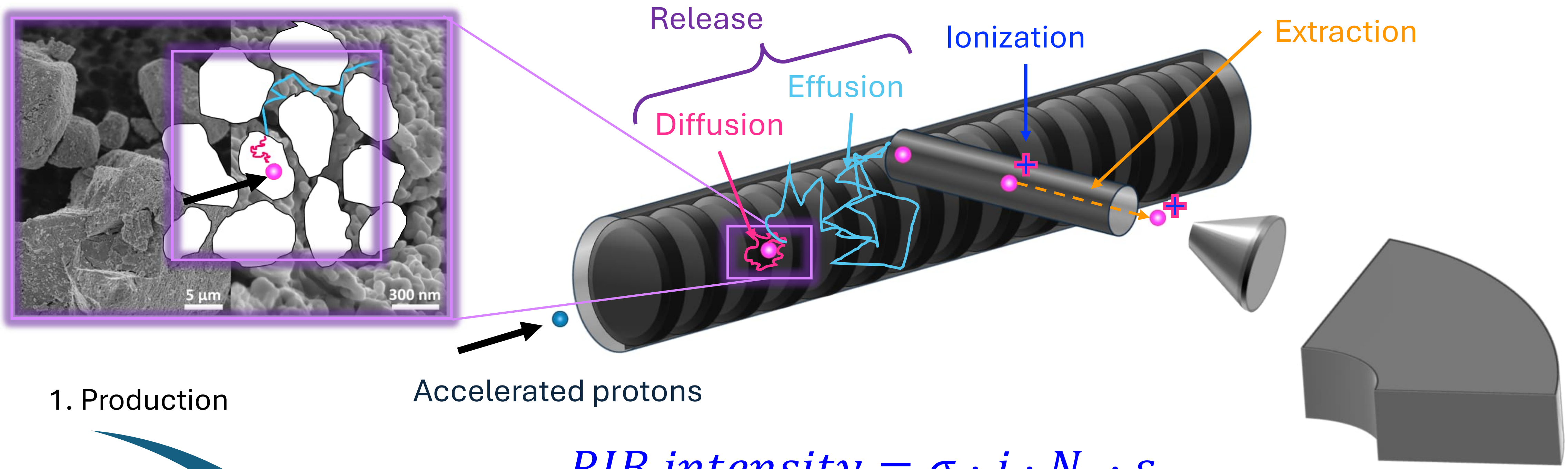
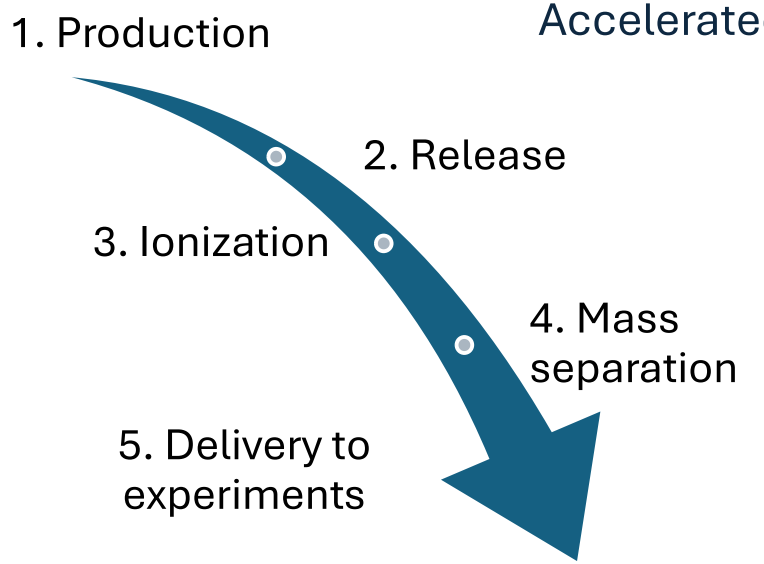
How does the ISOL method work?





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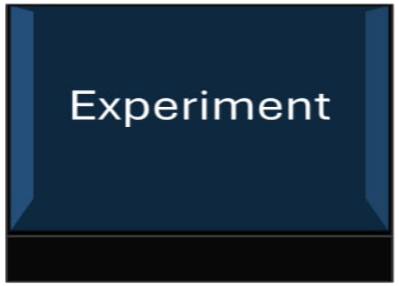
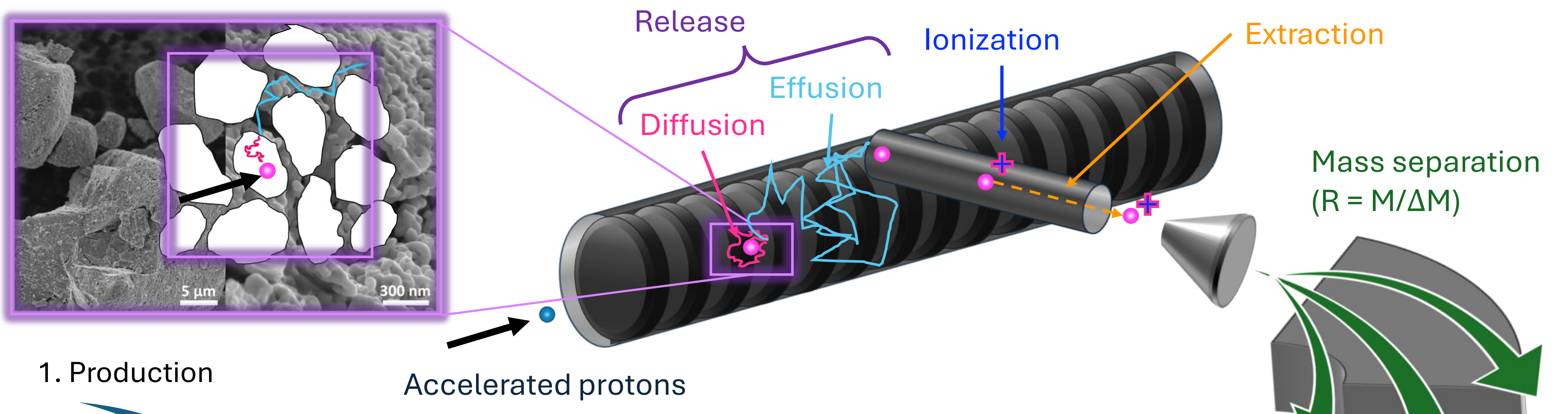
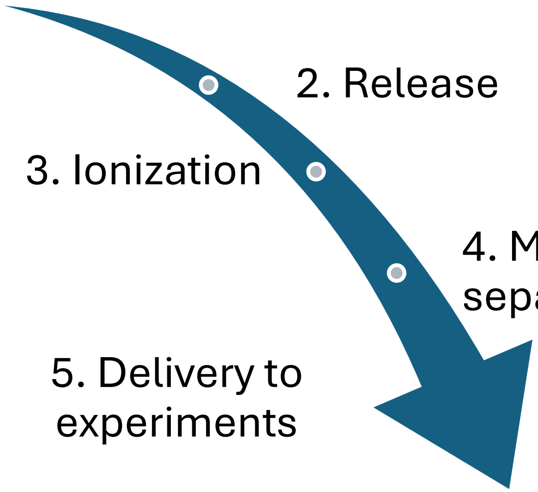


$$\varepsilon = \varepsilon_{diff}\varepsilon_{eff}\varepsilon_{is}\varepsilon_{ext}$$

How does the ISOL method work?





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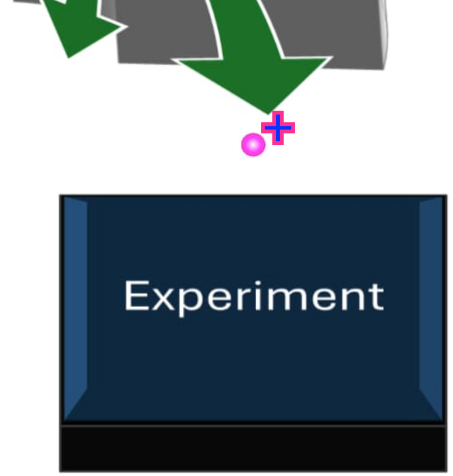
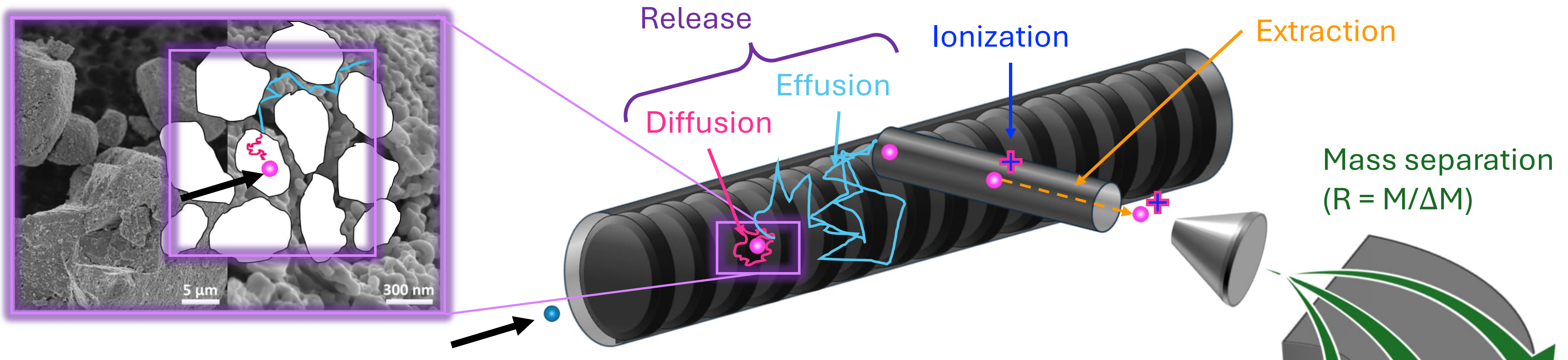
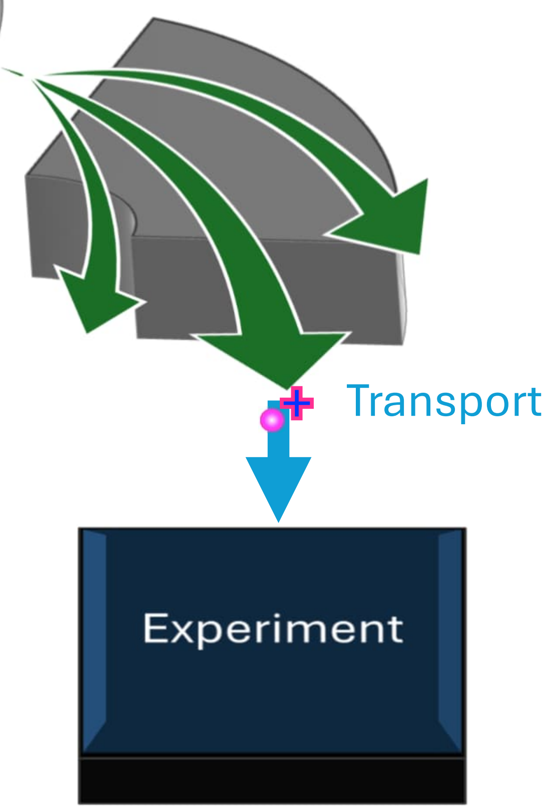
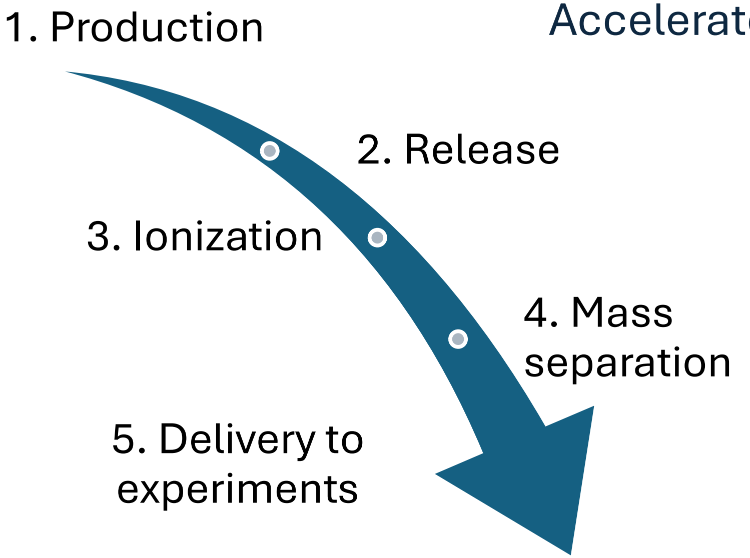


How does the ISOL method work?







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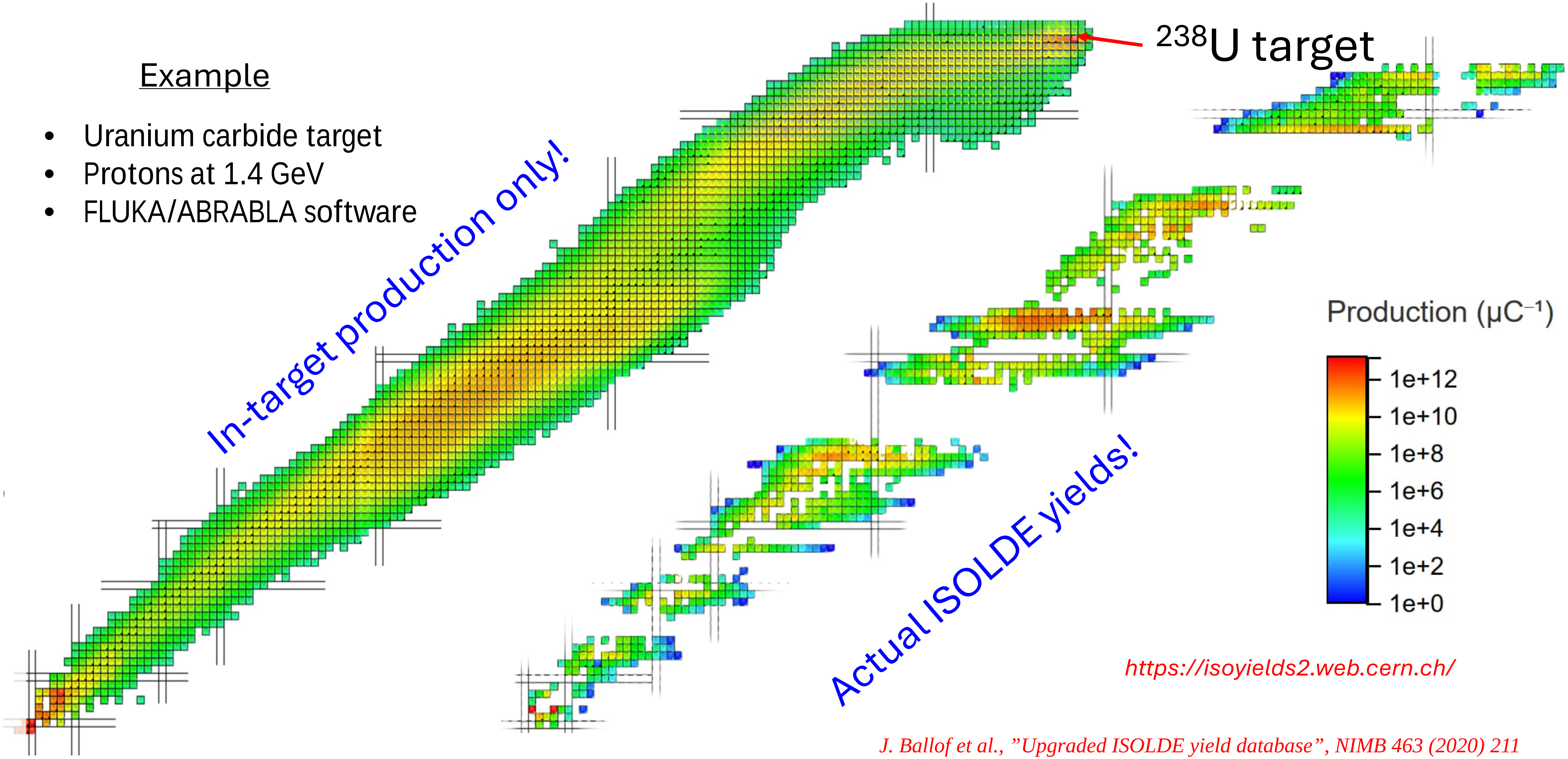
$$\varepsilon = \varepsilon_{diff}\varepsilon_{eff}\varepsilon_{is}\varepsilon_{ext}\varepsilon_{sep}\varepsilon_{trans}$$

ISOL step 1 - production

Example

- Uranium carbide target
- Protons at 1.4 GeV
- FLUKA/ABRABLA software



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

J. Ballof et al., "Upgraded ISOLDE yield database", NIMB 463 (2020) 211

Discussion pause: audience input please!!

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Why do you think there are gaps in the ISOL(DE) yields compared to the in-target production?

ISOL step 2 – release (element/Z dependency)

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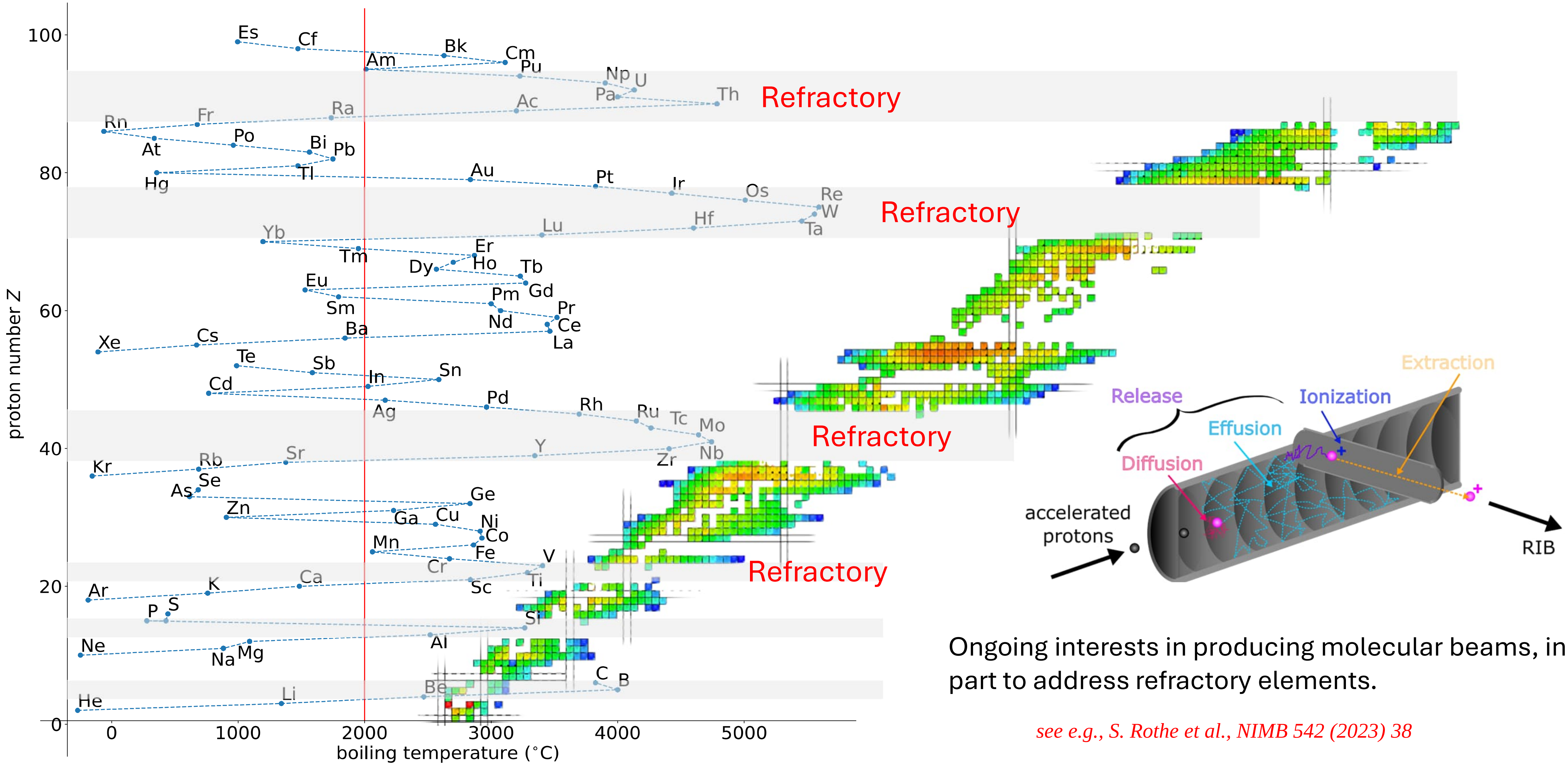

Ongoing interests in producing molecular beams, in part to address refractory elements.

see e.g., S. Rothe et al., NIMB 542 (2023) 38

ISOL step 3 - ionization

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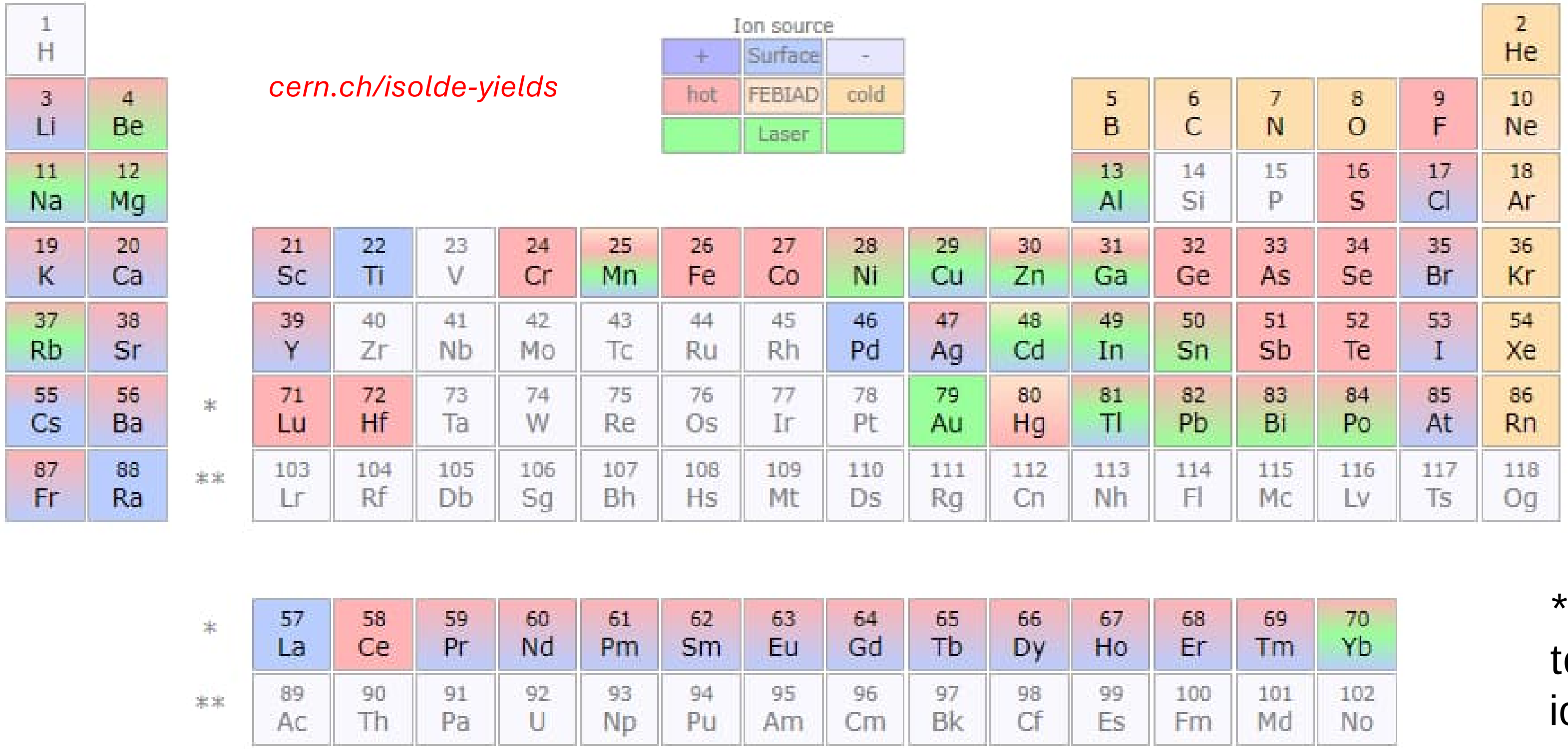
cern.ch/isolde-yields



J. Ballof et al., "The upgraded ISOLDE yield database…", NIMB 463 (2020) 211

Ion sources

- Surface ionization
- Plasma / electron impact ionization*
- Resonance laser ionization

*Note: electron impact ion sources include high-temperature gaseous discharge ion sources, ECR ion sources and EBIS.

These are generally very unselective!

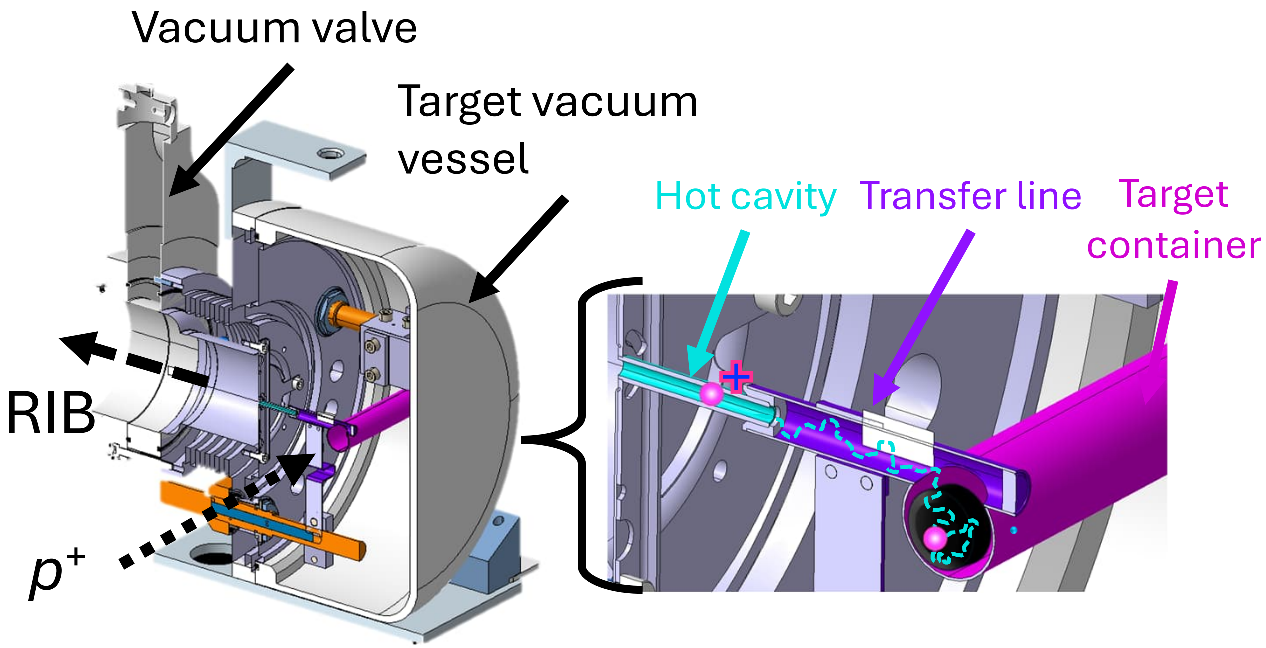
>1000 isotopes and isomers

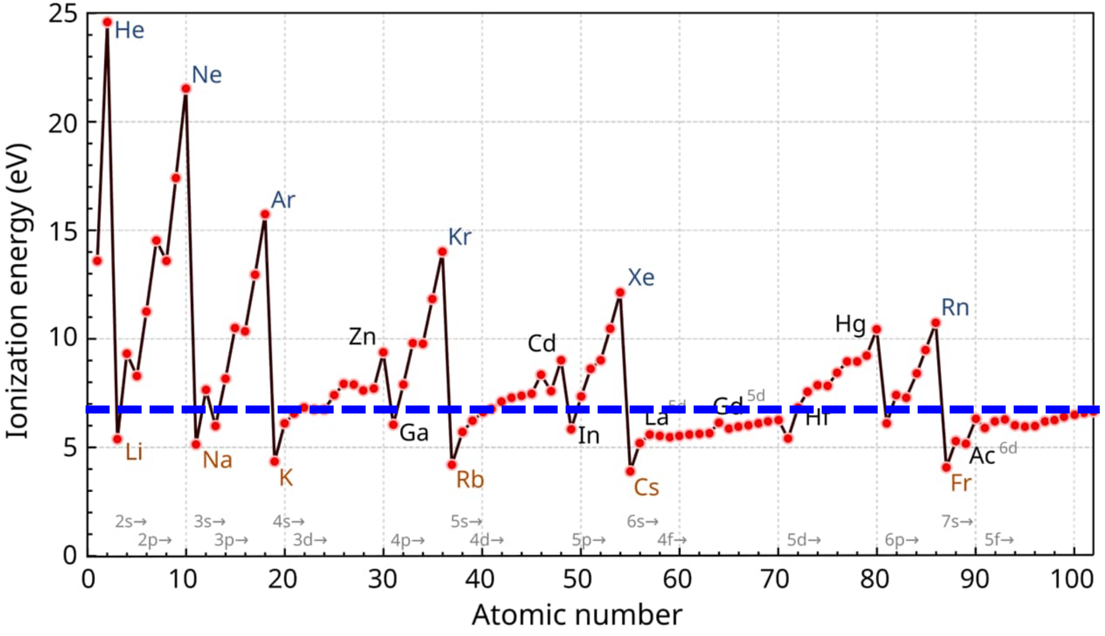
76 elements (ISOLDE)

- Ionization efficiency is usually defined for a specific isotope as the ratio of the number of ions extracted from the ion source to the number of atoms injected into the source.
- Radioactive decay losses treated separately.
- The ionization potential of the element of interest plays a critical role in ion source choice!

ISOL ion source types - 1

Surface ionization mechanism





Ionization efficiency depends on ionization potential, W_i, (and also the plasma potential in the hot cavity).
Φ = work function of the metal.

$$\frac{n_+}{n_a} = \frac{g_+}{g_a} exp\left(\frac{\Phi - W_i}{k_B T}\right) \qquad \epsilon_{ion} = {n_+}/{(n_+ + n_a)}$$

- (Positive) surface ion sources: simple, robust, reliable
- Very simple metal tube (Ta or W)
- Heated up to ~2400°C
- Low IP elements (W_i < 7eV) well surface ionized (e.g, alkali elements), however do become a source of contamination!

R. Kirchner, "On thermoionization in hot cavities", NIMA 292 (1990) 203

Surface ionization: a source of contamination

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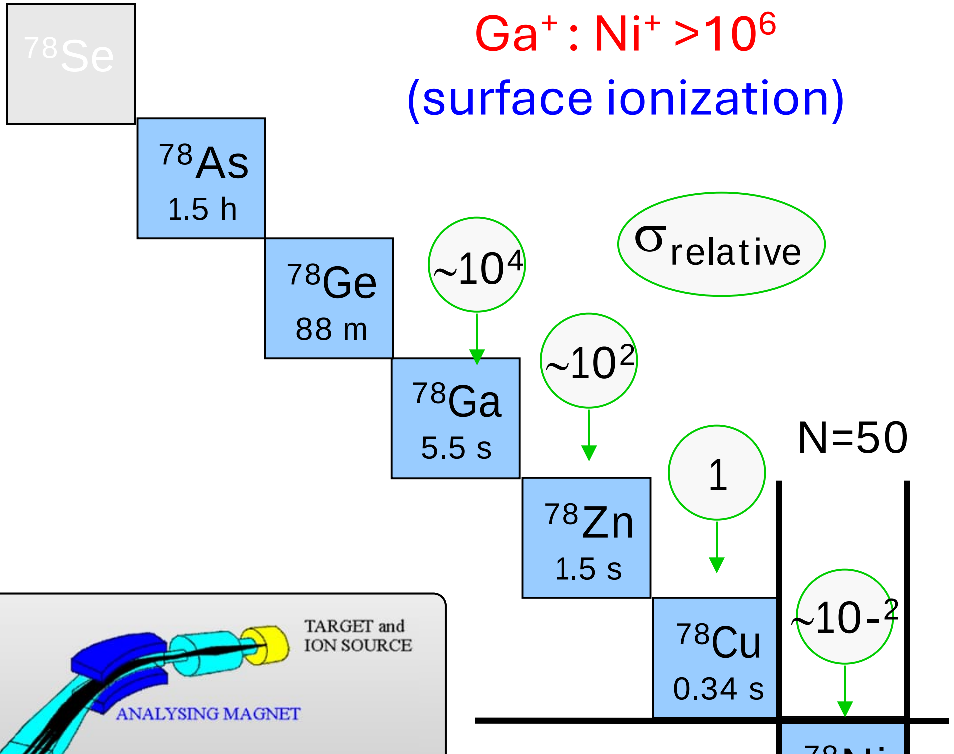
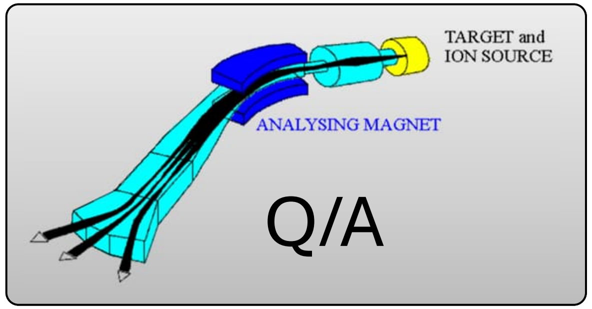
Fisson of U with 1 GeV protons (ISOLDE)

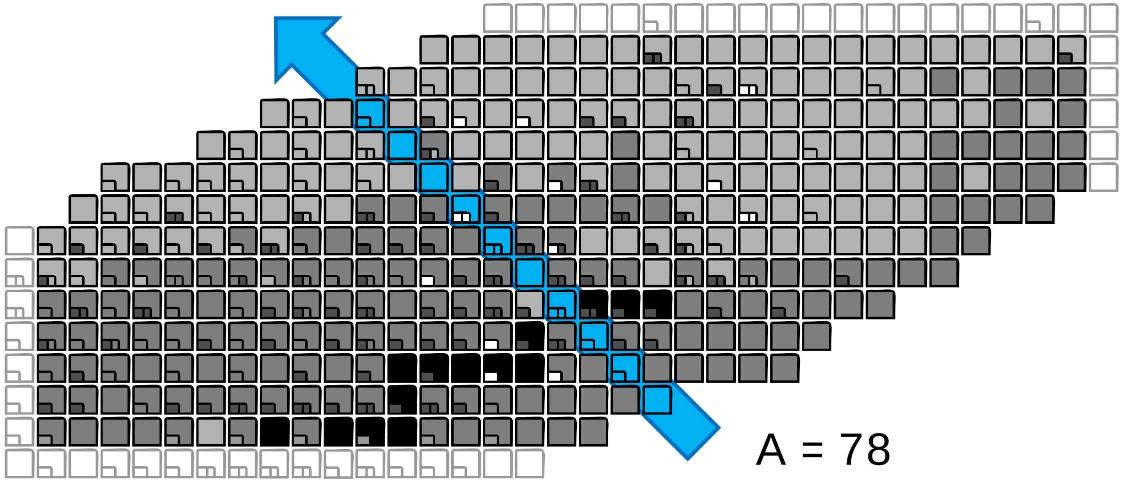
$Ga^+ : Ni^+ > 10^6$
(surface ionization)





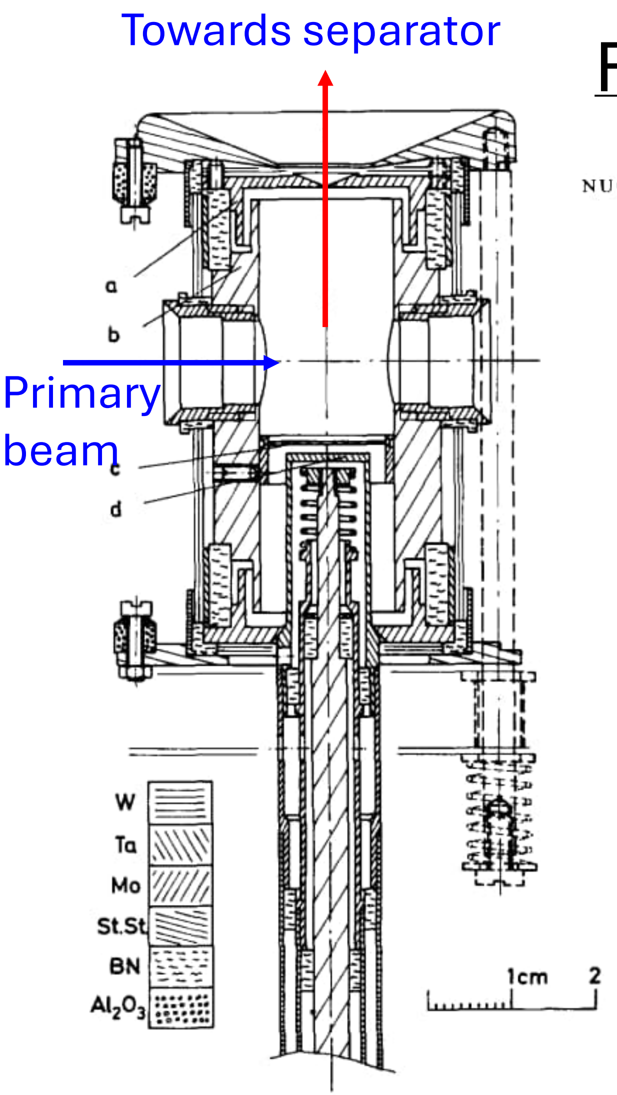
IP (Ga) = 5.99 eV IP (Ni) = 7.63 eV

$$\alpha = \frac{n_i}{n_0} = \frac{\omega_i}{\omega_0} \exp\left(\frac{\Phi - W_i}{kT}\right)$$



- Need to selectively increase Ni ionization efficiency and/or suppress other isobaric contaminants (Cu, Zn, Ga…)

ISOL ion source types - 2



FEBIAD source

(Forced Electron Beam Induced Arc Discharge)

NUCLEAR INSTRUMENTS AND METHODS 133 (1976) 187–204; © NORTH-HOLLAND PUBLISHING CO.

INVESTIGATION OF GASEOUS DISCHARGE ION SOURCES FOR ISOTOPE SEPARATION ON-LINE

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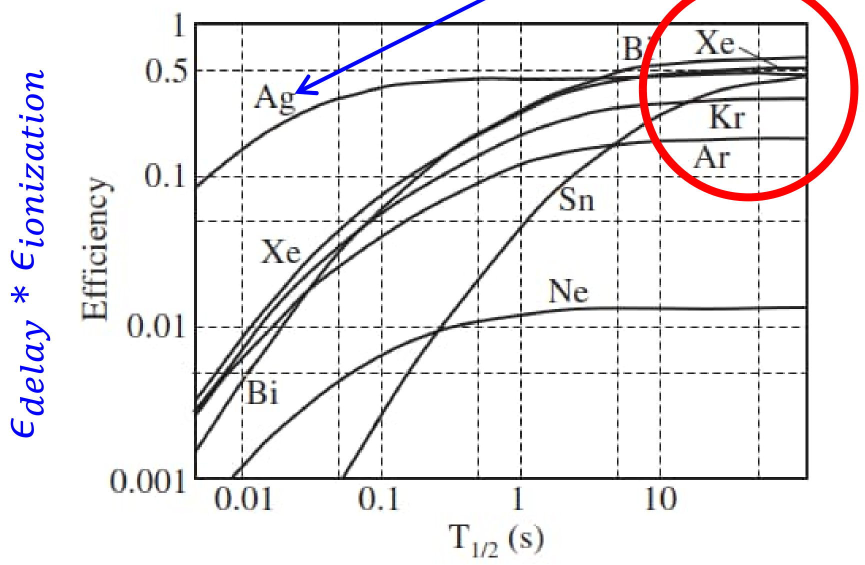
Received 20 November 1975

- FEBIAD ion source originally developed at the GSI on-line mass separator.
- Based on electron-impact ionization.
- Generally non-selective.
- Good for ionization of isotopes of elements with W_i > 7 eV.
- Many variants. Selectivity greatly increased by exploiting chemical properties of elements and materials.

Ag: fast and efficient!

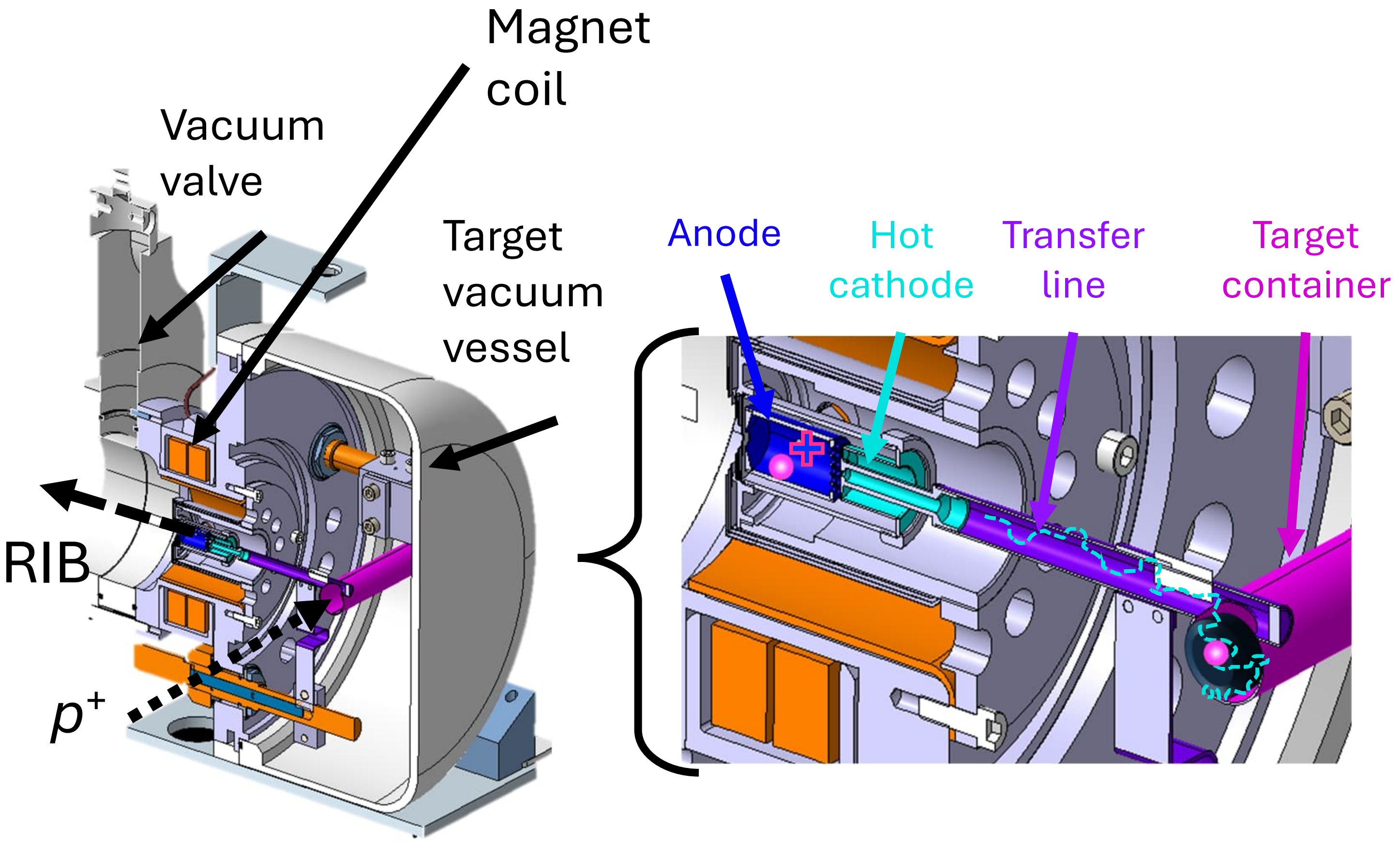


Half-life dependence of the separation efficiency for different catchers inside a FEBIAD ion source.

- Temp. (1700 – 2400 K)

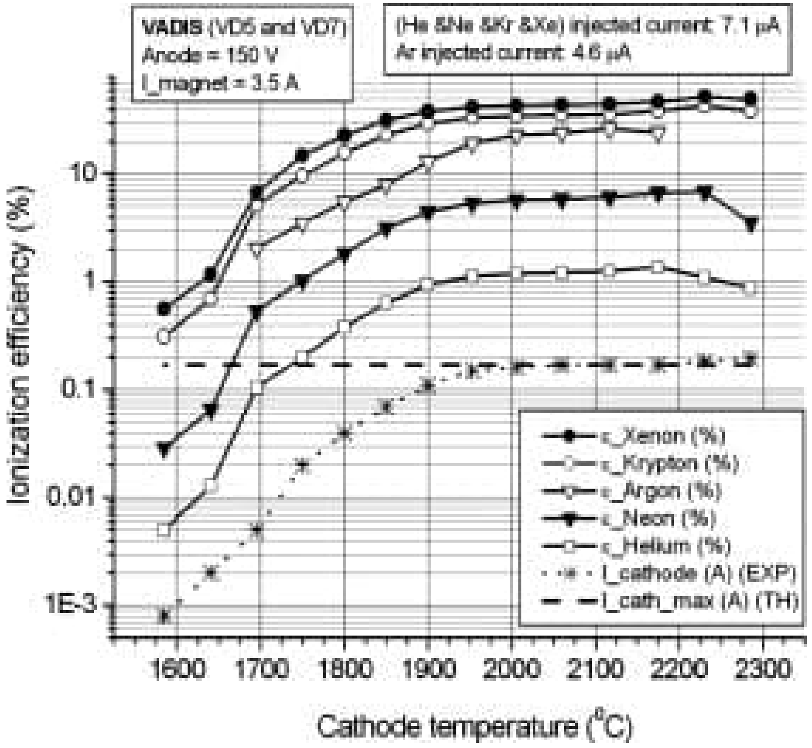
R. Kirchner et al., NIMB 70 (1992) 186

FEBIAD sources at ISOLDE

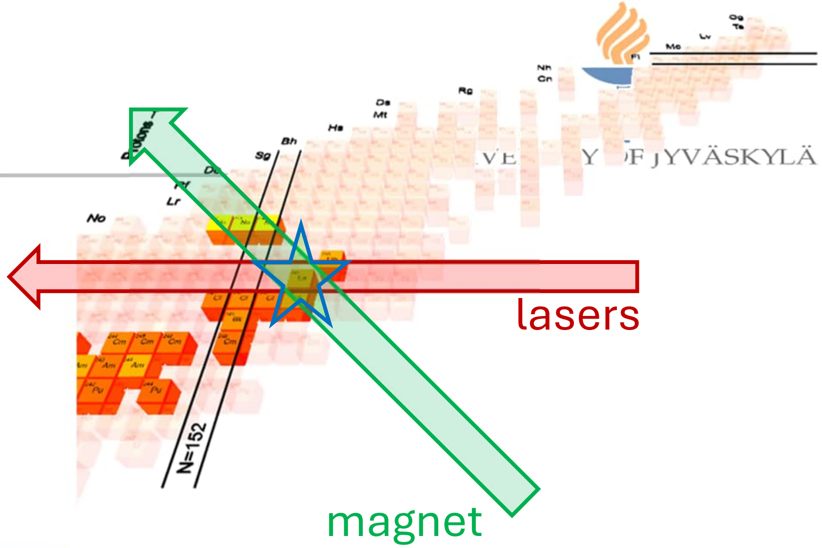


L. Penescu et al., "Development of the highly efficient VADIS source", Rev. Sci. Instrum. 81 (2010) 02A906

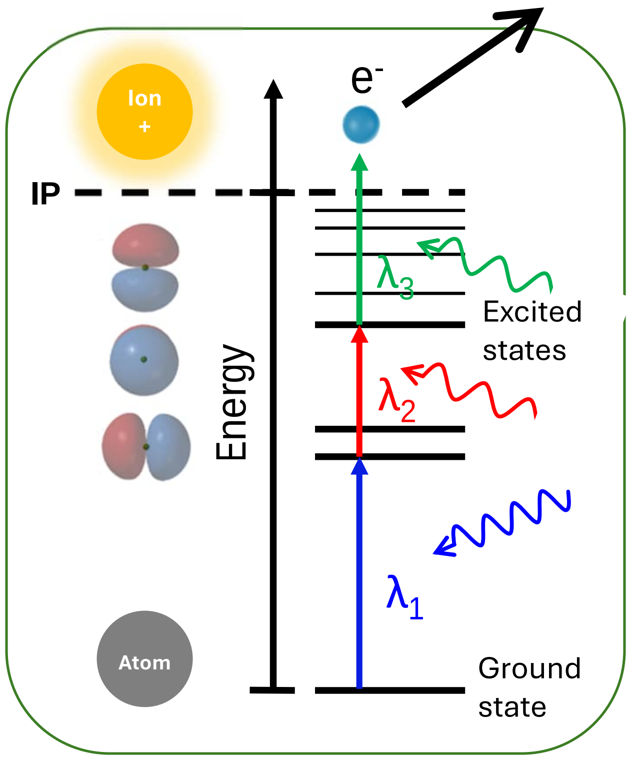
- At ISOLDE, a FEBIAD-type ion source is coupled with the target cylinder via a transfer tube.
- Later work led to the development of the VADIS ion source (Versatile Arc Discharge Ion Source), particularly promising for noble gas beams.

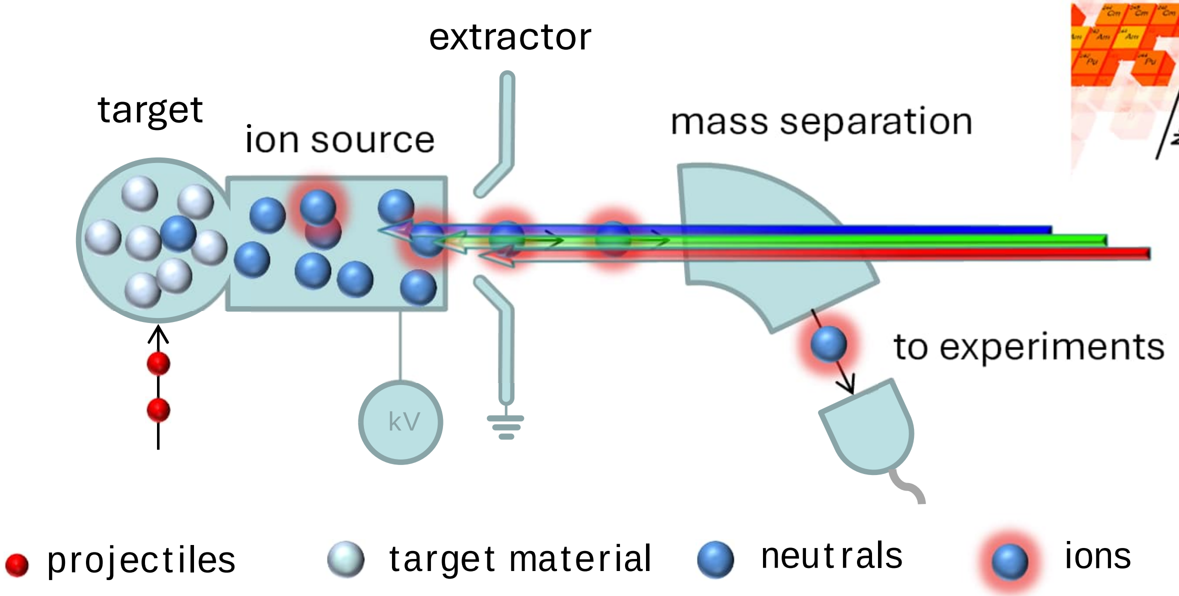


ISOL ion source types - 3



Resonance laser ionization ion source





SELECTIVITY & EFFICIENCY

- Effusion of reaction products as **hot atomic vapor** (> 2000°C)
- Suitable for elements with W_i ~4-9 eV
- Highly efficient laser ionization of **element of choice** (0.1 – 40%)
- Combined with mass separation, select only the isotope of interest.
- The ion source of choice for several RIB facilities!

How do we achieve efficient laser ionization?

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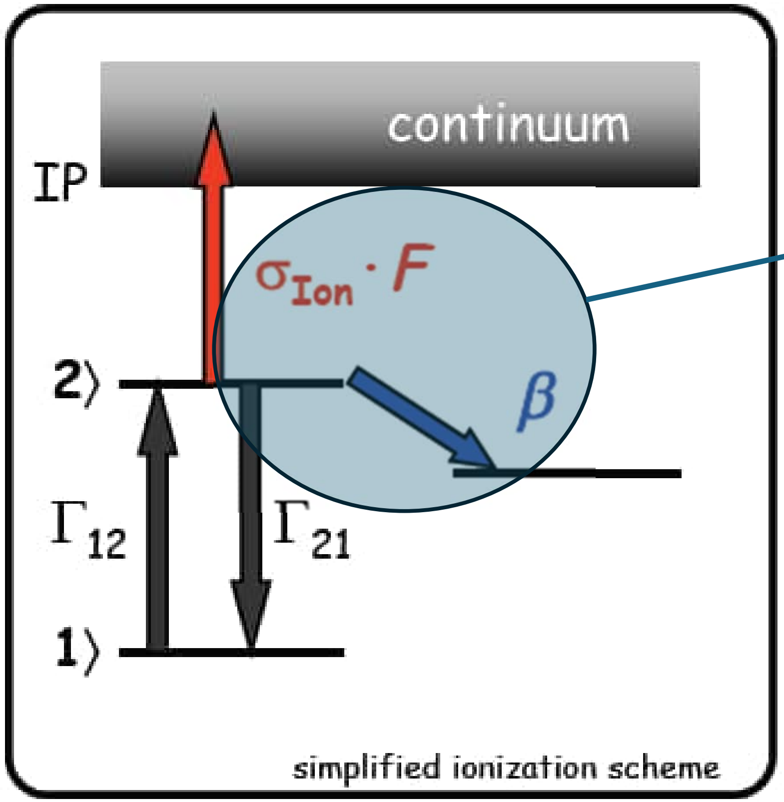
In order to efficiently ionize atoms irradiated by a laser we need to satisfy two conditions:



Flux condition

① $\sigma_{Ion} \cdot F \gg \beta$ → ionization rate » loss rate

Fluence condition

② $\sigma_{Ion} \cdot \varphi > 1$ → number of ionized atoms per laser interaction time (pulse)

σ_{Ion}	ionization cross section (cm^2)
β	loss rates to (metastable) states, state dependent (s^{-1})
F	photon flux ($cm^{-2}\ s^{-1}$)
φ	photon fluence (=photon flux · laser interaction time)

To be continuous or pulsed – that is the question!

Flux condition

(1) $\sigma_{Ion} \cdot F \gg \beta$ → ionization rate » loss rate

Fluence condition

(2) $\sigma_{Ion} \cdot \varphi > 1$ → number of ionized atoms per laser interaction time (pulse)

Typical values:

σ_{ion} → 10^{-17} cm^2

β → 10^6 s^{-1}

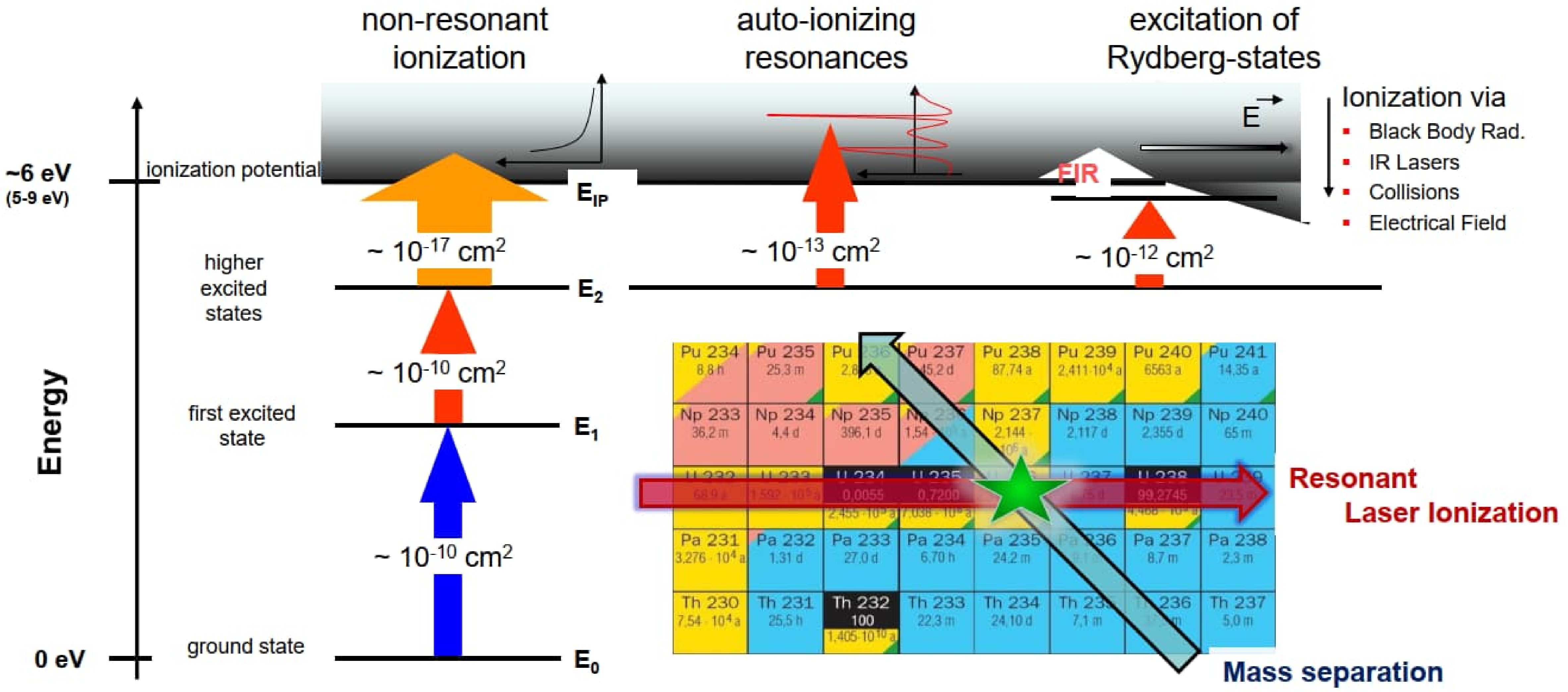
For simplicity and to have a safe margin, lets assume a laser beam area of 1 mm^2 and a photon energy of 3 eV.

From (1): Flux F >> 10^{23} cm^{-2}s^{-1}
→ # photons required >> 10^{21} /s → >> 500 W
Very challenging with continuous wave laser !!

But with a pulsed laser system:
Typical pulse length is 10 ns. → >> 5 µJ/pulse
No problem !!

Lets add in the Fluence (2) condition → > 0.5 mJ/pulse
(>5 W at 10kHz)

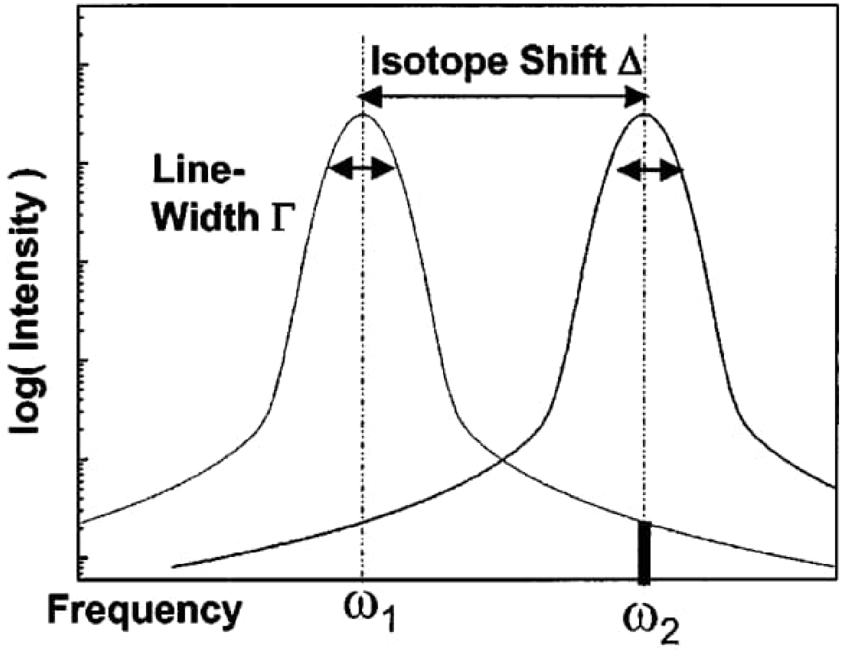
Pathways to ionization



+ mass separation → isotope selectivity

How do we quantify the optical selectivity?

Simplified model of two Lorentzian peaks:



When the laser is in resonance with a selected isotope and but far from other ` ` contaminating´ ´ elements or isotopes (Δ), the selectivity *S* is given by a simple quantitative estimate:

$$S \sim 4 \times \frac{\Delta^2}{\Gamma^2}$$

Pd isotopes

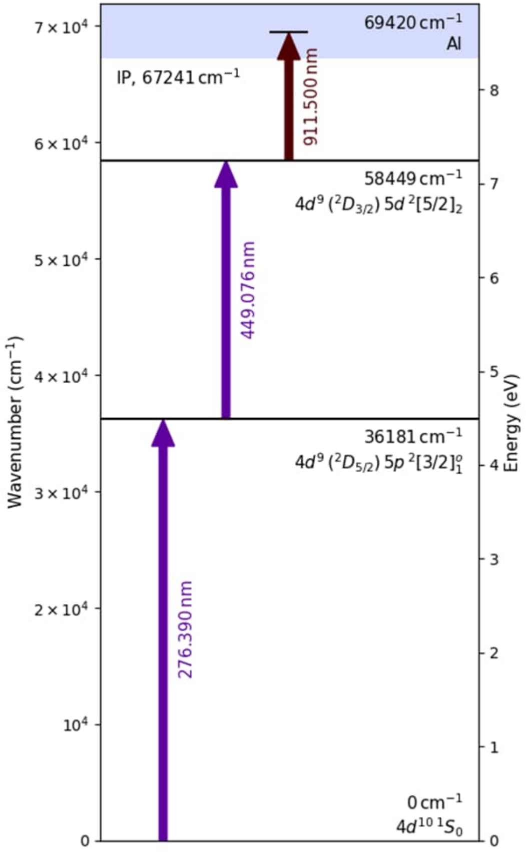
Γ ~3 MHz, Δ ~100 MHz (neighbouring isotopes):

S ~4000

Δ ~10^{15} Hz (palladium to silver):

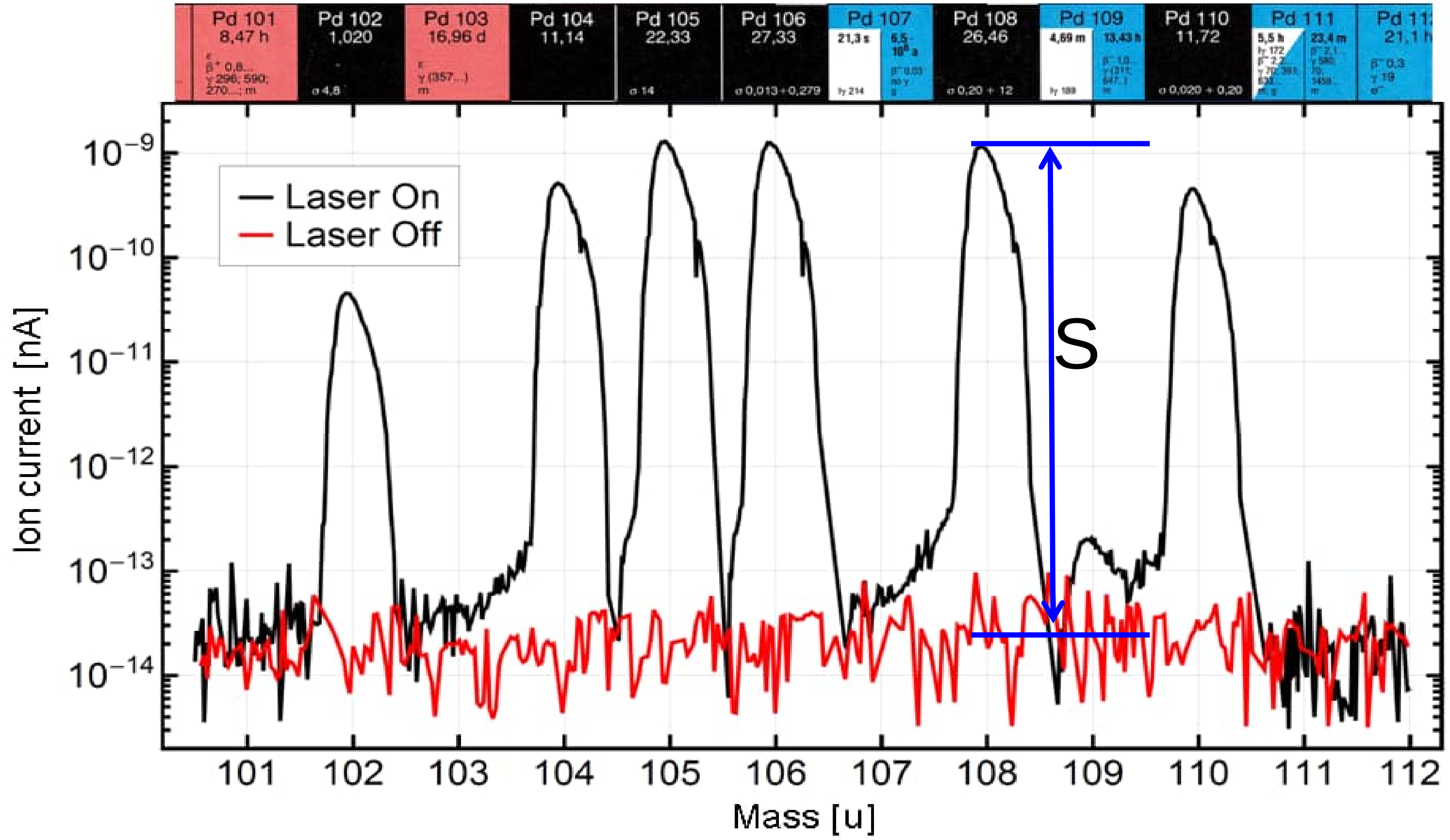
S ~10^{17} !!!

Multi-step excitation: $S = S_1 \cdot S_2 ... \cdot S_n$.



Three-step ionization scheme

Discussion pause...

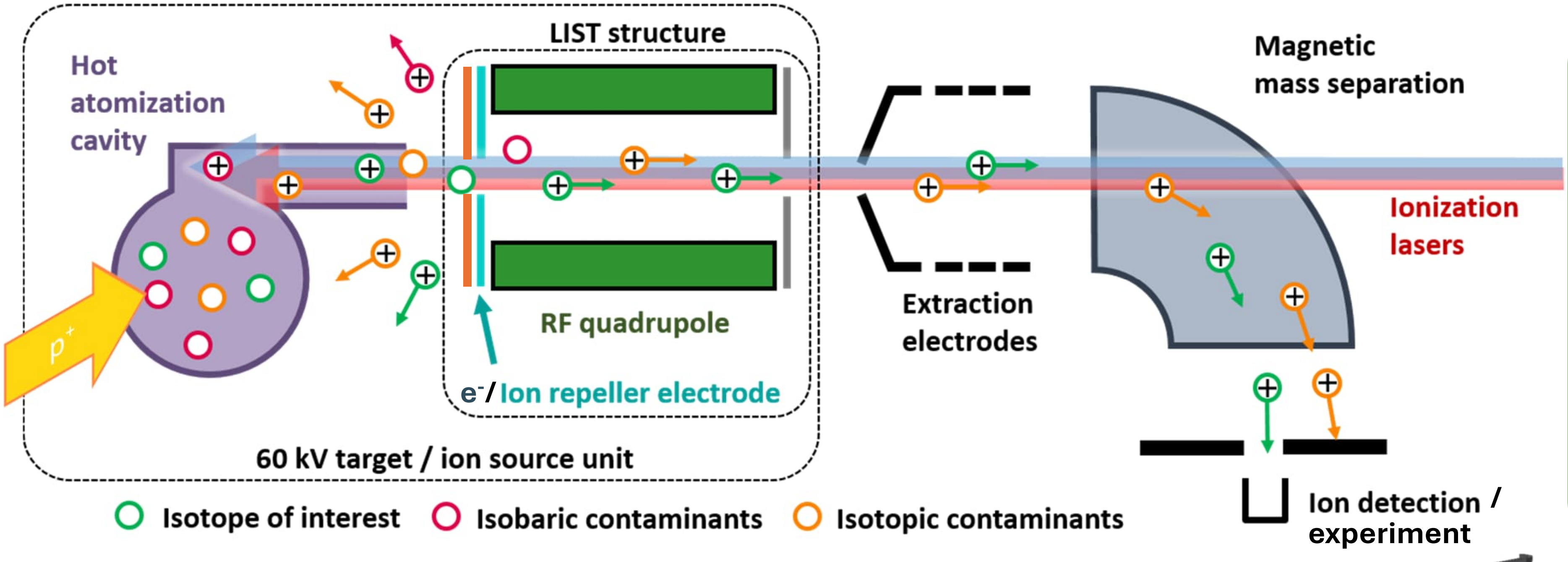


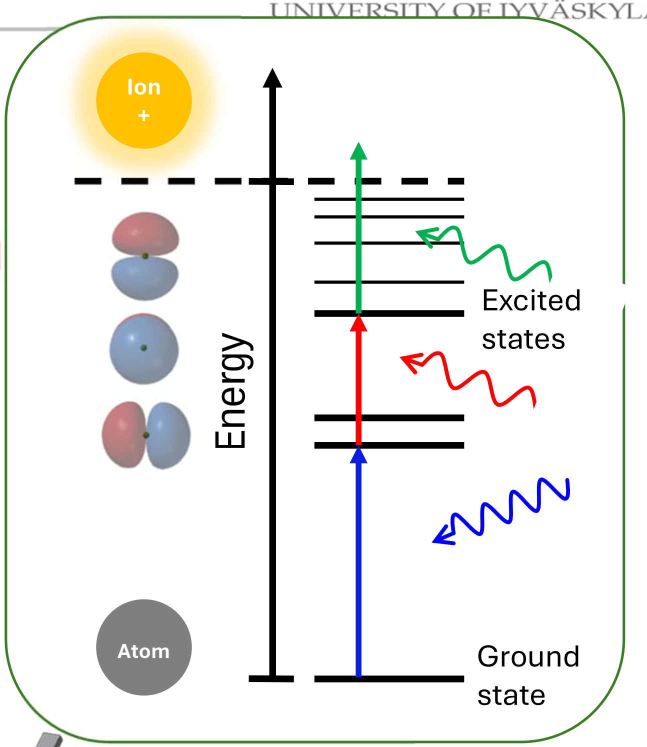
Mass scan over all stable Pd isotopes

Why do you think the selectivity as given here (using a three-step ionization scheme) is far from the simple estimate?

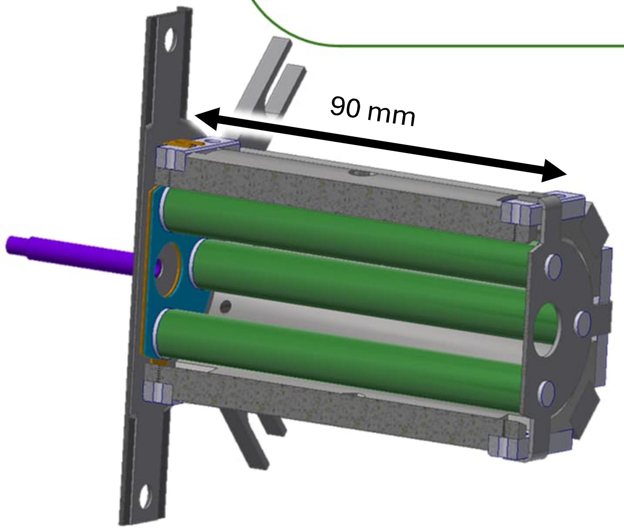
~10^{10} (est.) vs. 50000 (expt).

The Laser Ion Source and Trap (LIST)



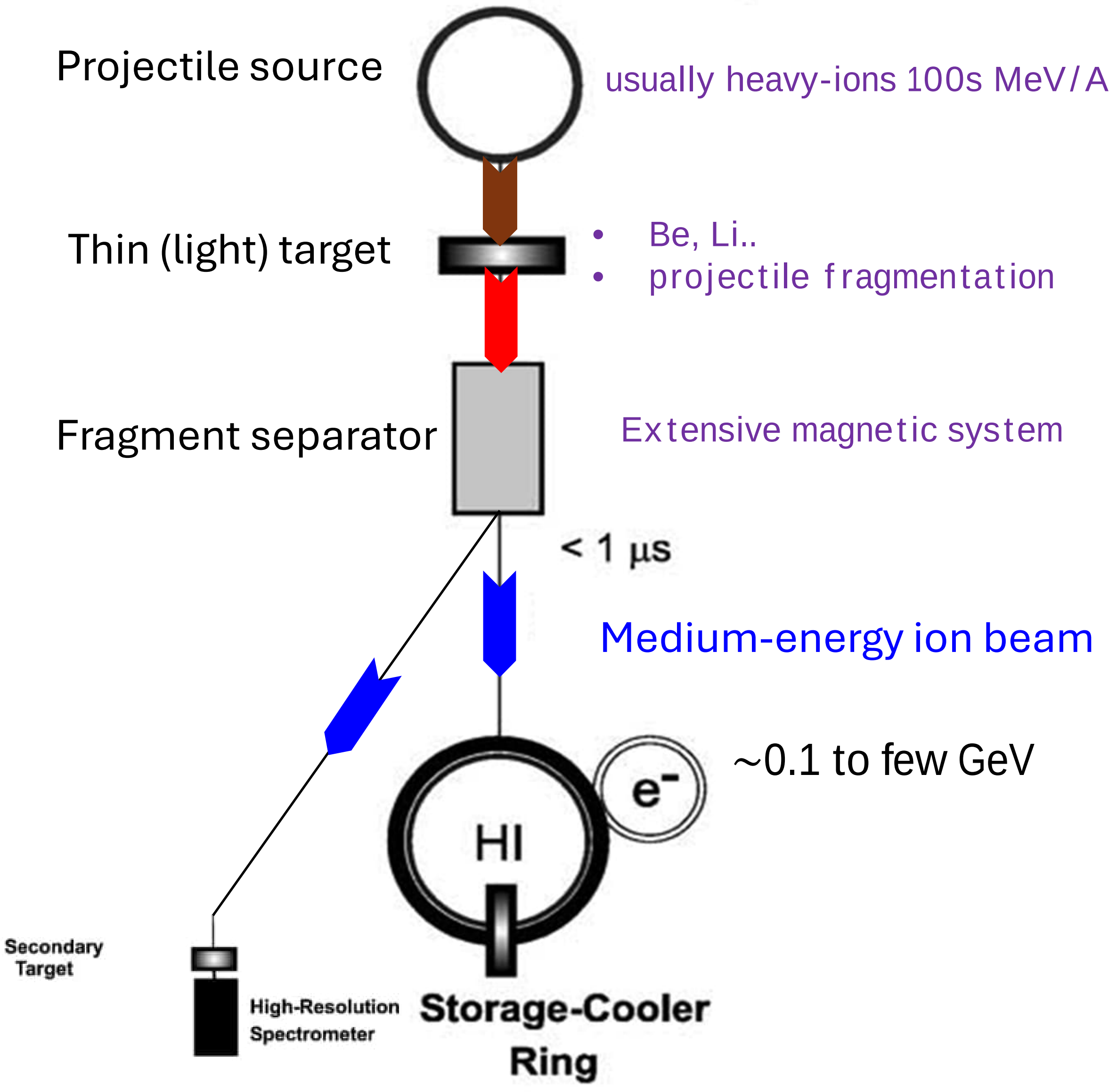


- **Spatial separation:** hot cavity ↔ laser ionization volume
- **Suppression** of surface ionized species
- **Pure laser ionization** inside **RF quadrupole** structure
- Factor up to 10^6 in suppression ↔ Factor 20-100 reduction in beam intensity
- Versatility: Mimic standard RILIS by extracting and guiding ions from hot cavity




Let’s pause here to breathe. I should find some new scenary!

The in-flight method



First in-flight separator, Oak Ridge, USA (1958)

Some key properties:

- Chemical insensitivity (beams of all elements)
- Half-lives << 1ms
- Beam properties fixed (often poor quality due to the large phase space)
- Precision experiments at low-energy not directly accessible

Discovery potential:
loosely-bound isotopes at the very edge of stability

Example facilities:
RIBF-RIKEN 350 MeV/A heavy ions
FRIB-USA 200-250 MeV/A heavy ions
FAIR-GERMANY 2 GeV/A heavy ions
GANIL 95 MeV/A

Fragment separators and isotope separation

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- Modern separators have large acceptances

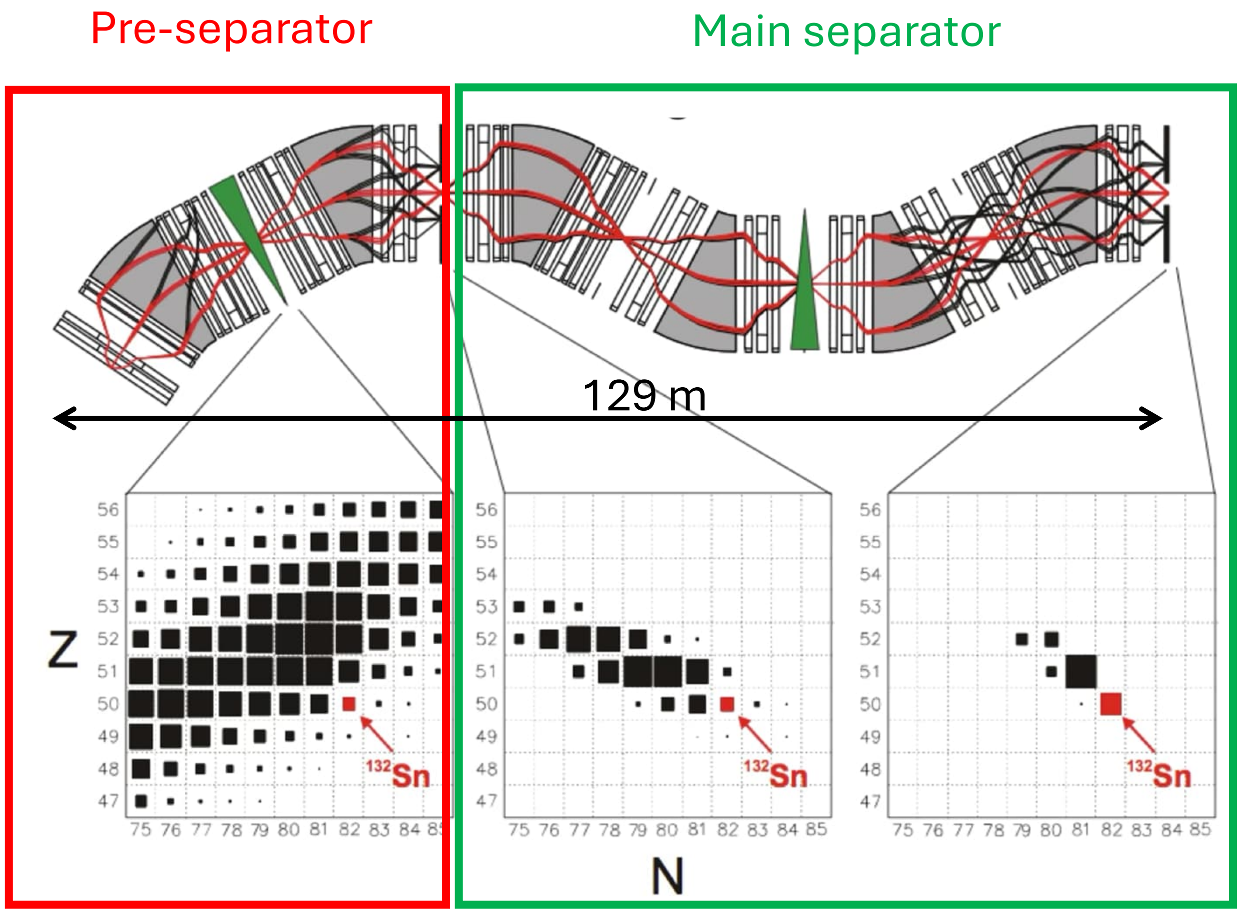
- can approach 100% of typical momentum distribution

- Often two-stage separator schemes are employed

- first stage for production & separation
- second stage may allow for delivery of “tagged” beams

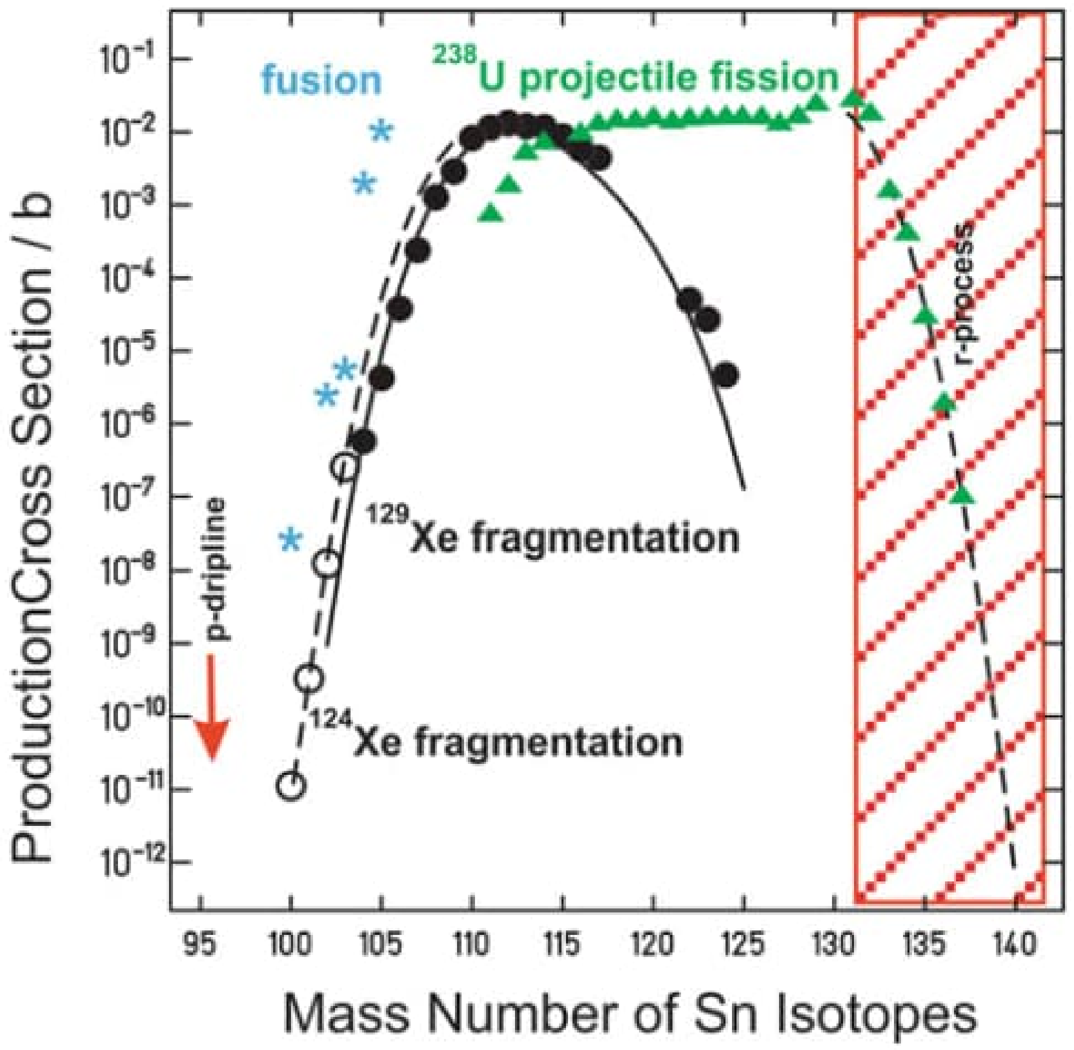
- 1.5 GeV/u ^{238}U beam on 4 g/cm^2 C target
- Using a combination of degraders, slits and detectors one can see the separation performance of the S-FRS for doubly-magic ^{132}Sn.
- Fragment intensities in main separator ~10^9 ions/s

Example of S-FRS separation performance



M. Winkler et al. DOI:10.5170/CERN-2003-004.7

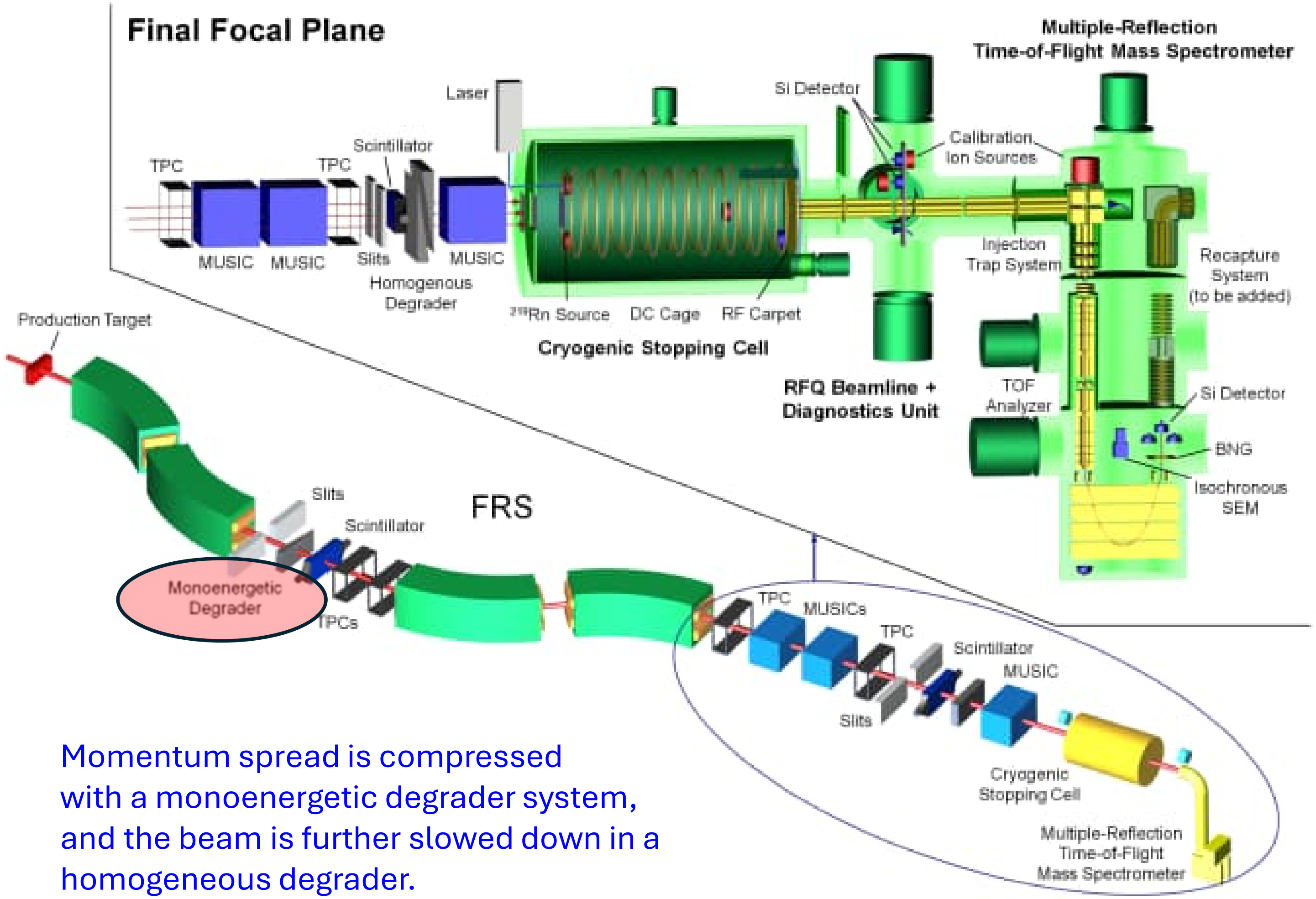
A comparison of production cross sections



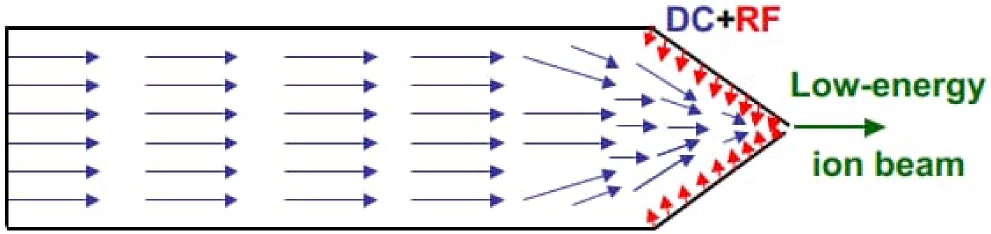
- Projectile (or target) fission of uranium favours the most neutron-rich Sn isotopes.
- Fragmentation of xenon isotopes or low-energy fusion-evaporation reactions favour the most neutron-deficient nuclei.
- We can be guided by the choice of facility depending on the species of interest but must consider the purity of the beam and type of experiment.
- Low-energy experimental techniques are not possible – further manipulation is required.

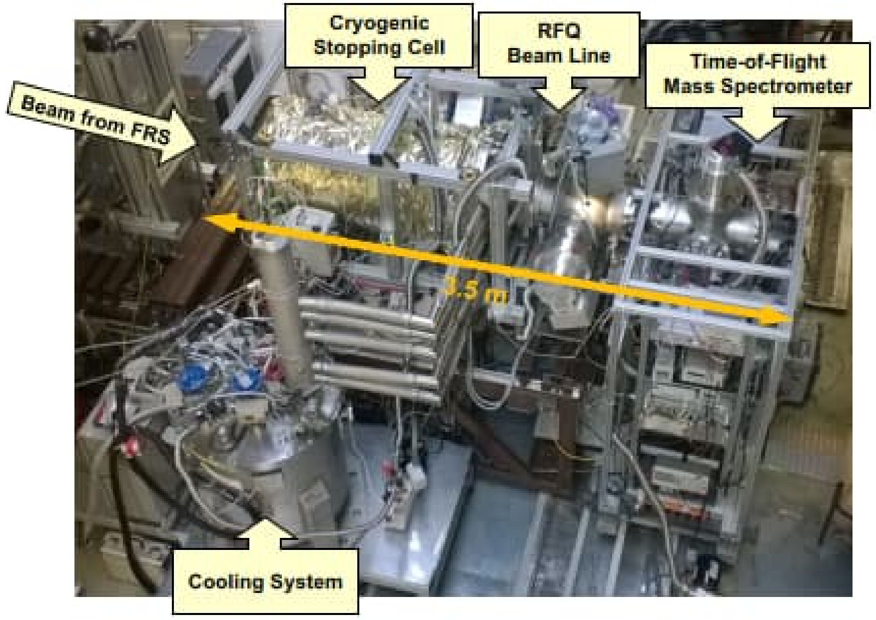
Exotic Nuclei and their Separation, Electromagnetic Devices. H. Geissel and D.J. Morrissey, Handbook of Nuclear Physics, Springer Nature 2023.

The Ion Catcher at the FRS, GSI



Momentum spread is compressed with a monoenergetic degrader system, and the beam is further slowed down in a homogeneous degrader.





TDR for the Cryogenic Stopping Cell of the Super-FRS at FAIR, Eur. Phys. J Special Topics (2025).

The IG(ISOL) / gas catcher (hybrid) method

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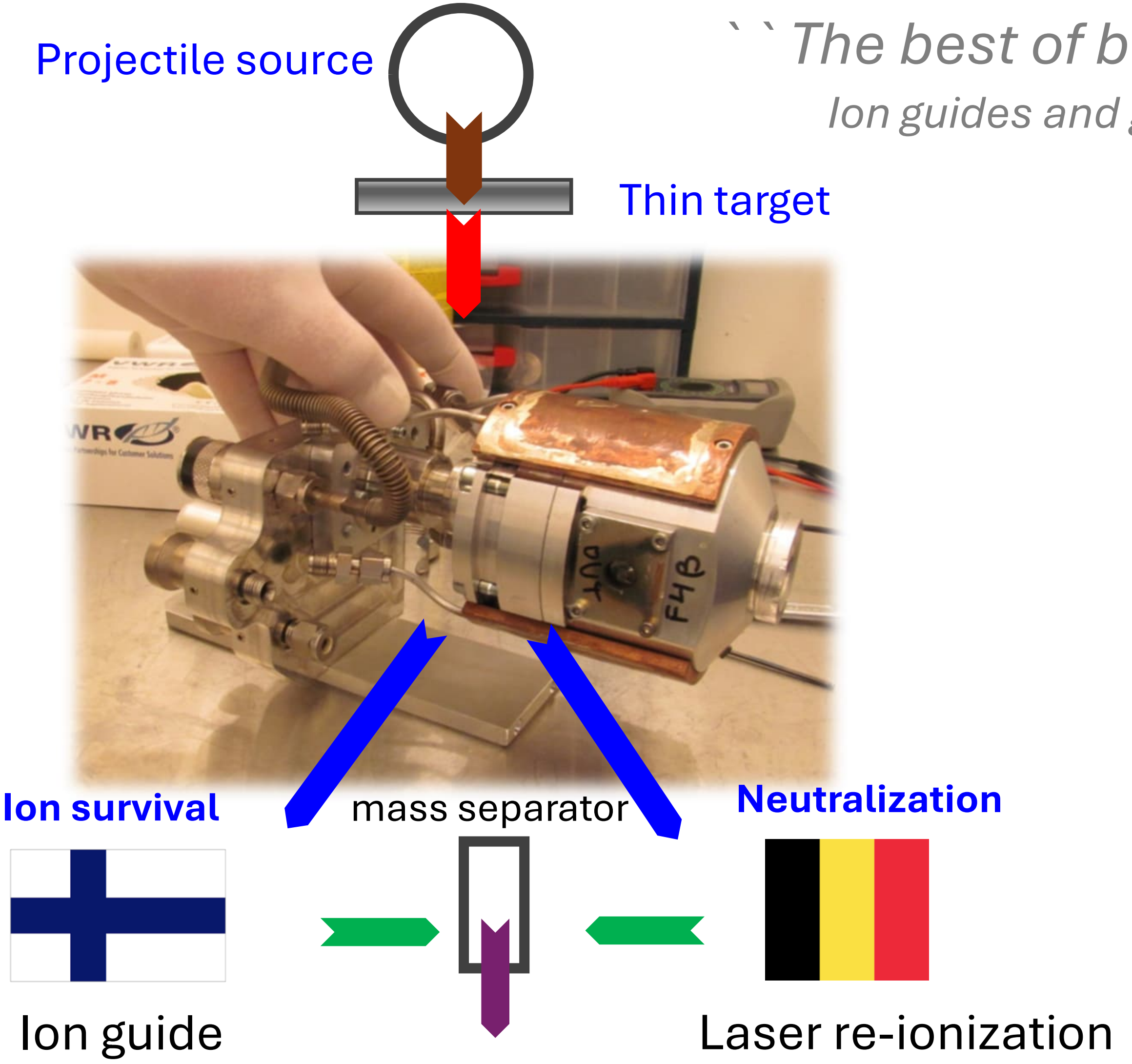
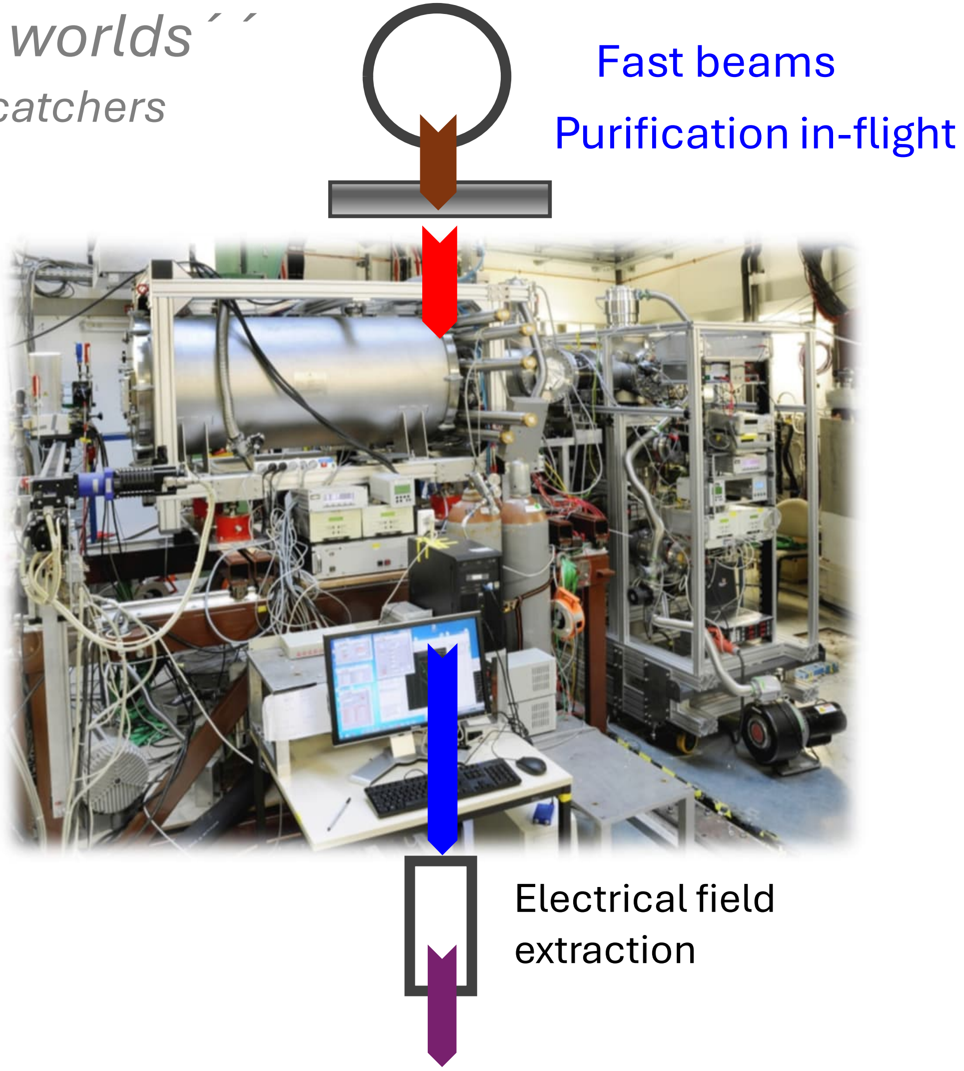
``The best of both worlds´´

Ion guides and gas catchers





Submillisecond On-Line Mass Separation

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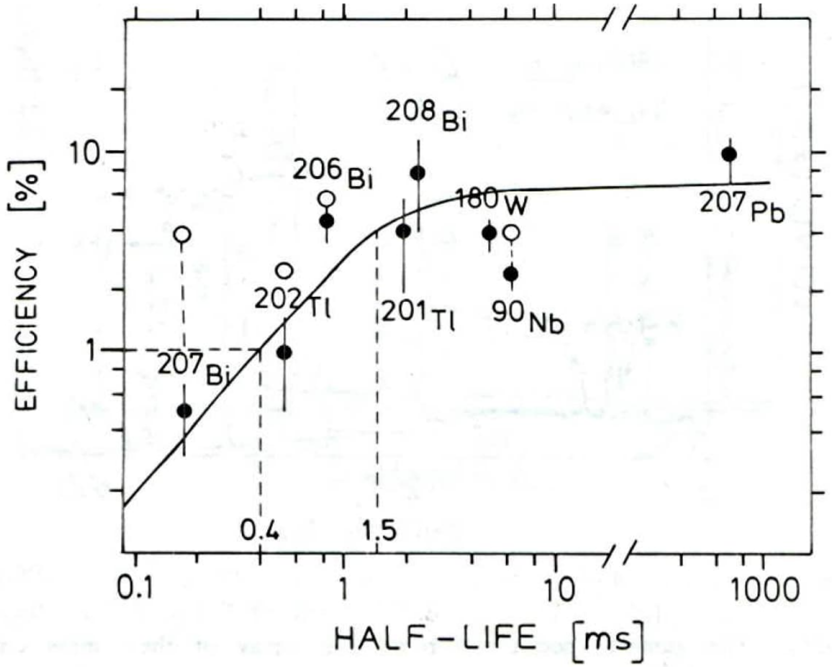
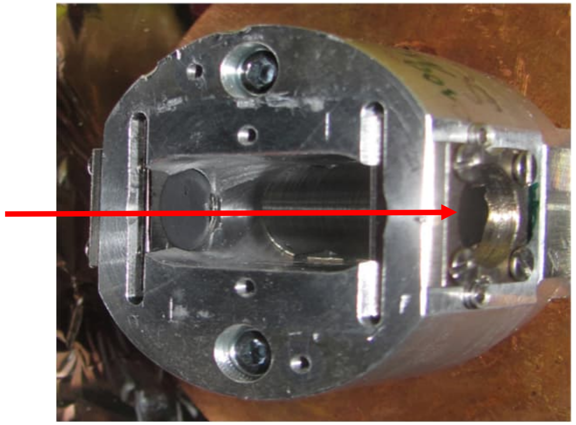
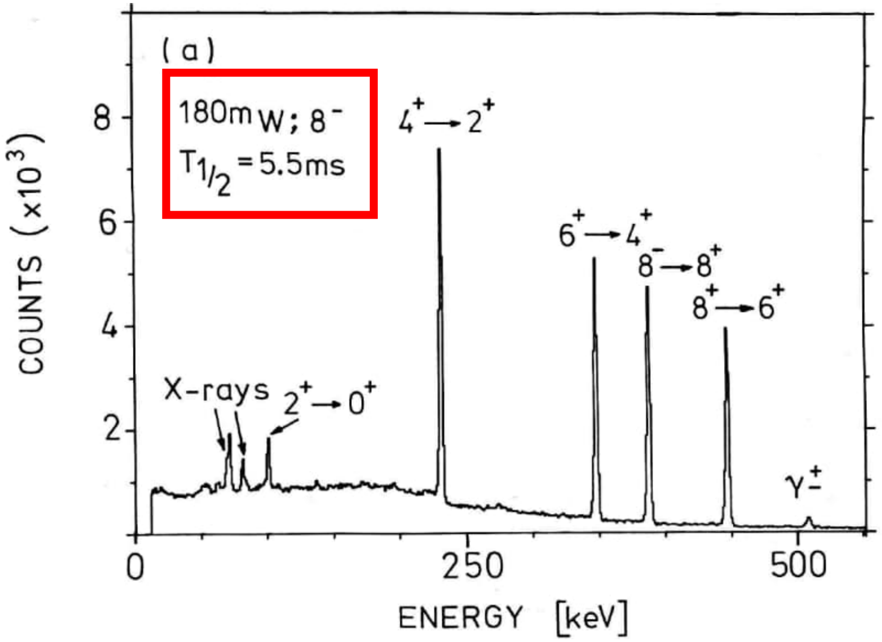
VOLUME 54, NUMBER 2 PHYSICAL REVIEW LETTERS 14 JANUARY 1985

Submillisecond On-Line Mass Separation of Nonvolatile Radioactive Elements: An Application of Charge Exchange and Thermalization Processes of Primary Recoil Ions in Helium



Small volume gas cell (few cm^3)





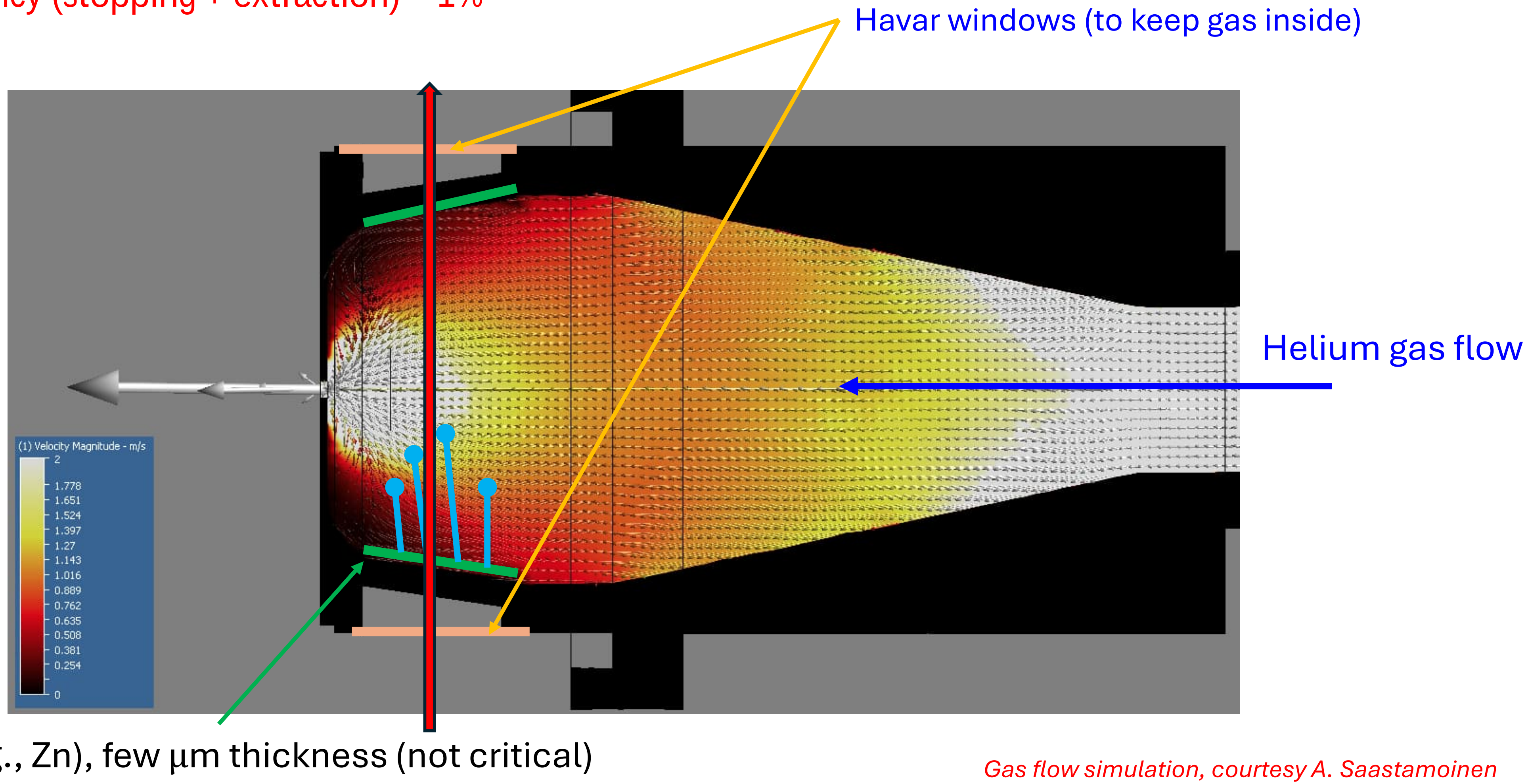
- Extraction times *can be* <1 ms; typical extraction efficiency ~10%
- Chemical independence (or independence of recoil volatility)
- Relatively low yields (~1 mg/cm^2) compared to ISOL (several g/cm^2)

Recall, tungsten is the most refractory element (melting point 3695 K, boiling point 6203 K)

J. Ärje, J. Äystö et al., Phys. Rev. Lett. 54 (1985) 99

Light-ion fusion-evaporation ion guide

- Goal: production of neutron-deficient nuclei (closer to stability)
- Light accelerated beams, e.g., protons, deuterons, $^{3,4}He$…., on (usually) stable metallic targets
- Ion guide efficiency (stopping + extraction) ~1%



Gas flow simulation, courtesy A. Saastamoinen

Light-ion induced fusion-evaporation reactions

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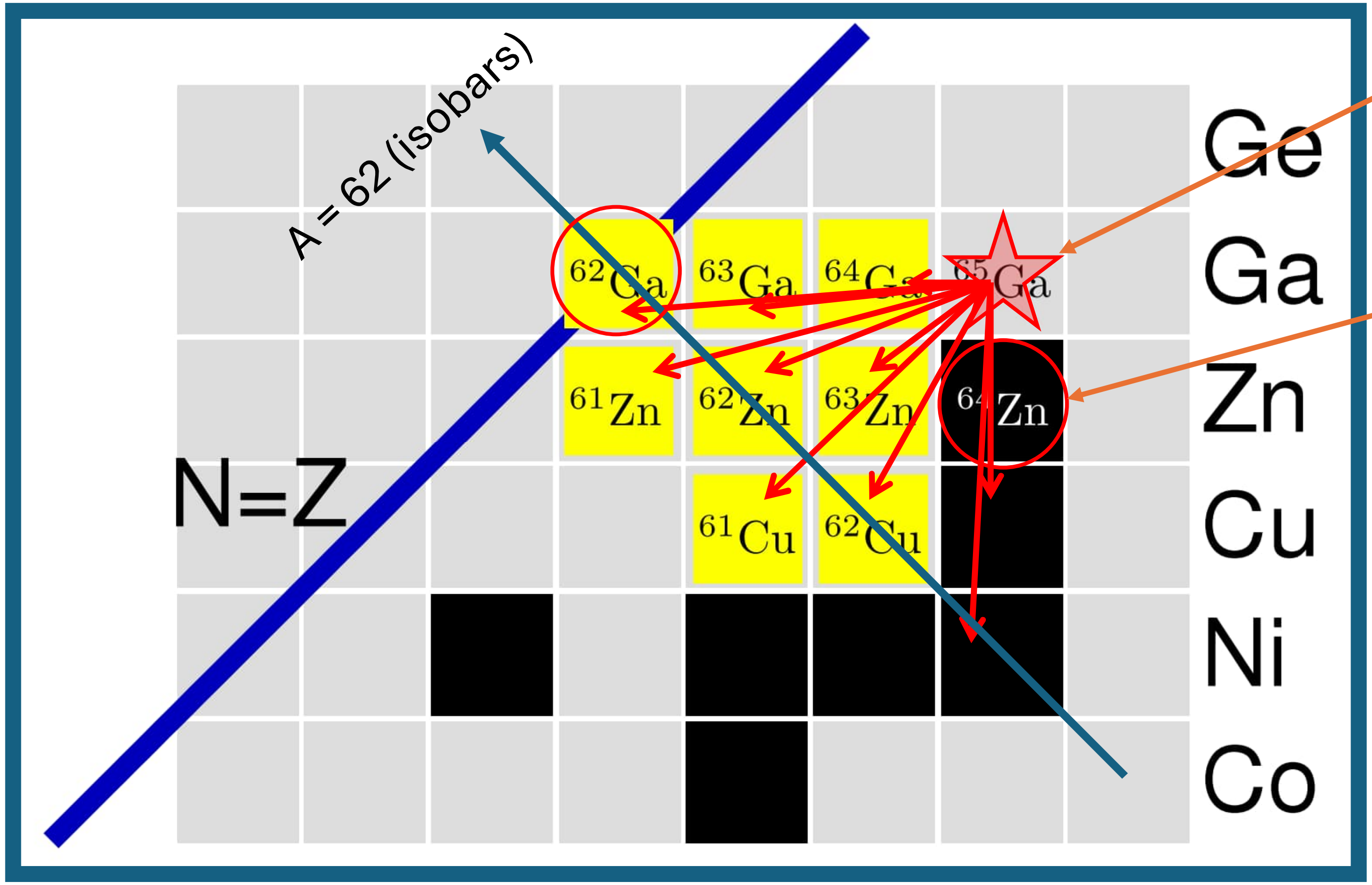
Goal: production of neutron-deficient nuclei (closer to stability)

^{62}Ga ($N = Z$ nucleus), half-life 116 ms



Example:

Protons + ^{64}Zn (48% isotopic abundance)

+ proton, compound nucleus (^{65}Ga)

Target (^{64}Zn)

- Several evaporation channels are "open"
- Production rates depend on:
  - Reaction cross section
  - # bombarding particles (protons)
  - Target thickness
  - Efficiency (from production to experiment)

Can we estimate the expected number of ``contaminants´´?

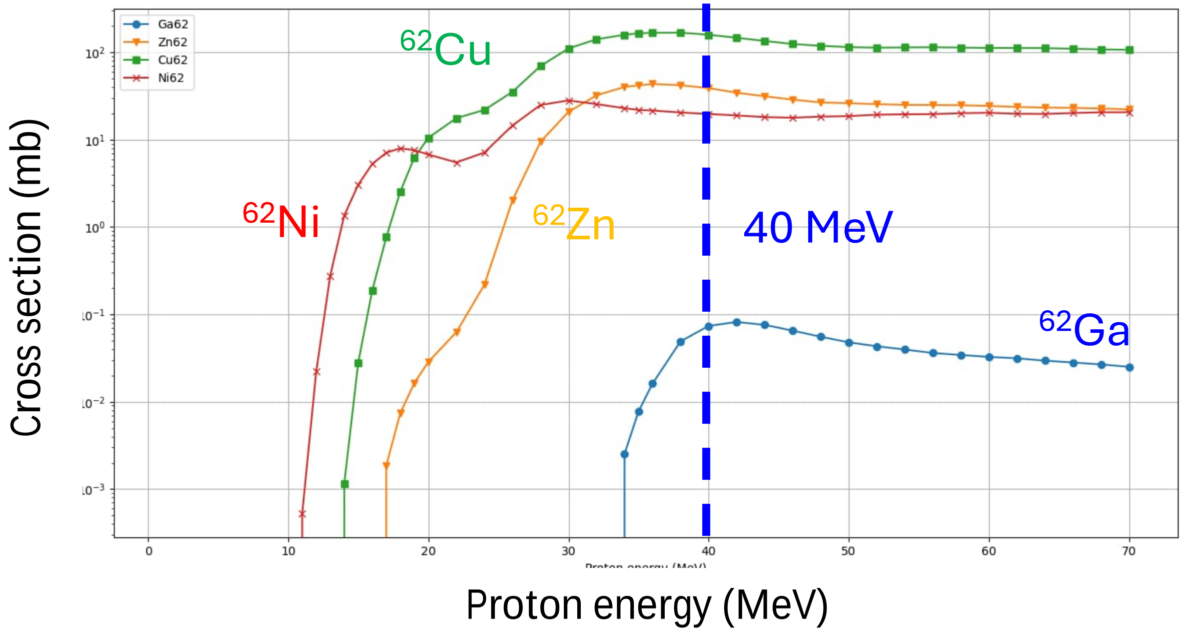
Reaction cross sections

- Reminder: the reaction cross section answers the question of how probable it is to have a reaction.
- Computer codes available, e.g.,
  - HIVAP, PACE4, TALYS
- Experimental cross sections compiled to Exfor database
  - https://www-nds.iaea.org/exfor/

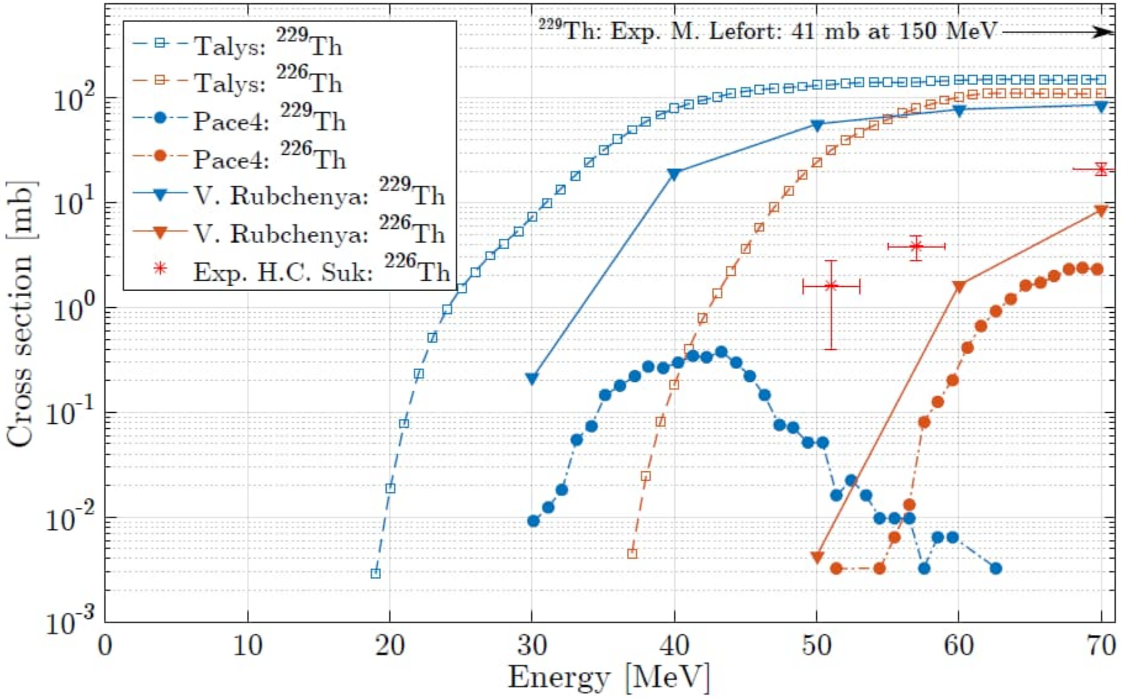
https://nds.iaea.org/relnsd/talys/talys.html

@ 40 MeV, σ ~0.1 mbarn (^{62}Ga via p,3n channel)
σ ~100 mbarn (^{62}Cu)

Protons on a ^{nat}Zn target (Talys)



Protons on a ^{232}Th target; (p,pxn) reactions



Take home messages

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Our beautiful radioactive ion beam production techniques

- The two main workhorses for radioactive ion beam production are the ISOL method and the in-flight (fragmentation) method. The ion guide/gas catcher technique is a compromise between these two.
- Depending on our region of interest / experiment of choice, we choose the appropriate facility to perform experiments. Target development, ion source type, are all important factors.
- The ion guide method of production is universal. This is a blessing and a curse.
- Element selectivity is critically important in RIB production (radioactive inventory) Laser ion sources are widely used/planned (ISOL).
- We will discuss some of the ion manipulation methods post- mass separator in Lecture 5.