



Principles of ISOL-type radioactive ion beam production

Simon Stegemann

CERN-ISOLDE

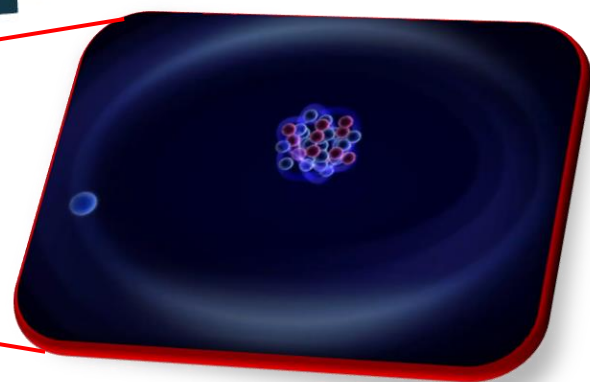
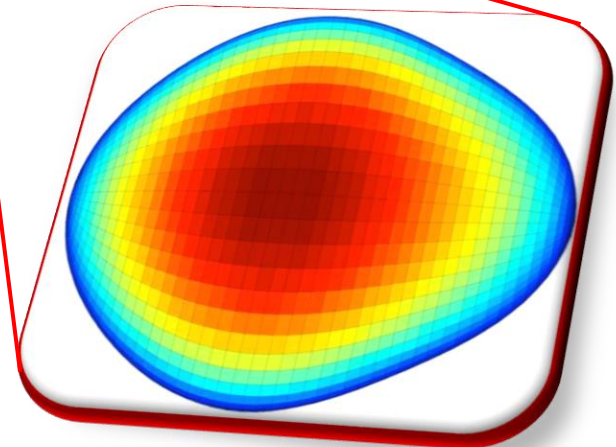
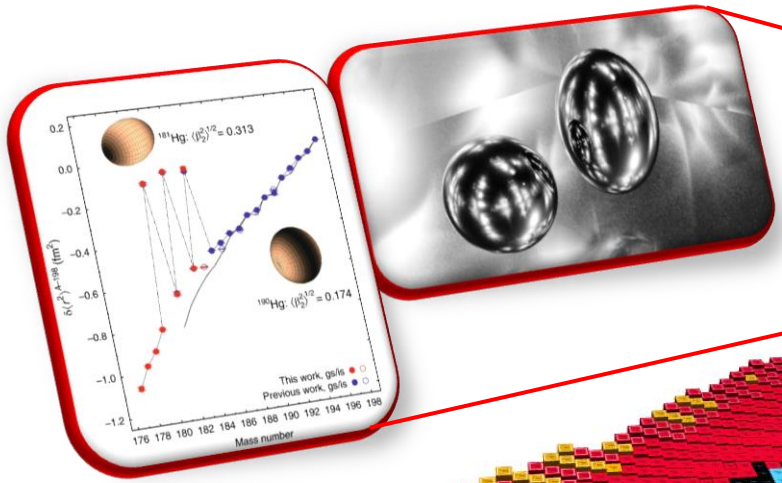
SY-STI-RBS

20/03/2023

ISOL-France workshop V, Bordeaux

Content

- 1. Why ISOL beams?, ISOL history**
- 2. ISOL challenges**
- 3. ISOL stages**
- 4. Use cases around the world**



Why radioactive ion beams?

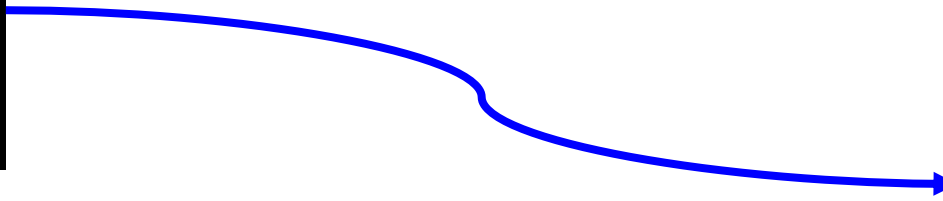
Production:

high radiation environment

primary
beam



target



Detection:

low radiation background



Transport methods:

- carry (“SRAFAP”)
- drive (*G.T. Seaborg and W.D. Loveland, The Elements beyond Uranium, John Wiley & Sons, 1990*)
- transport shuttle with pressurized air
- transport in gas-jet
- pump through vacuum system
- **send as ion beam**

Speed (approx.)

10 m/s

50 m/s

100 m/s

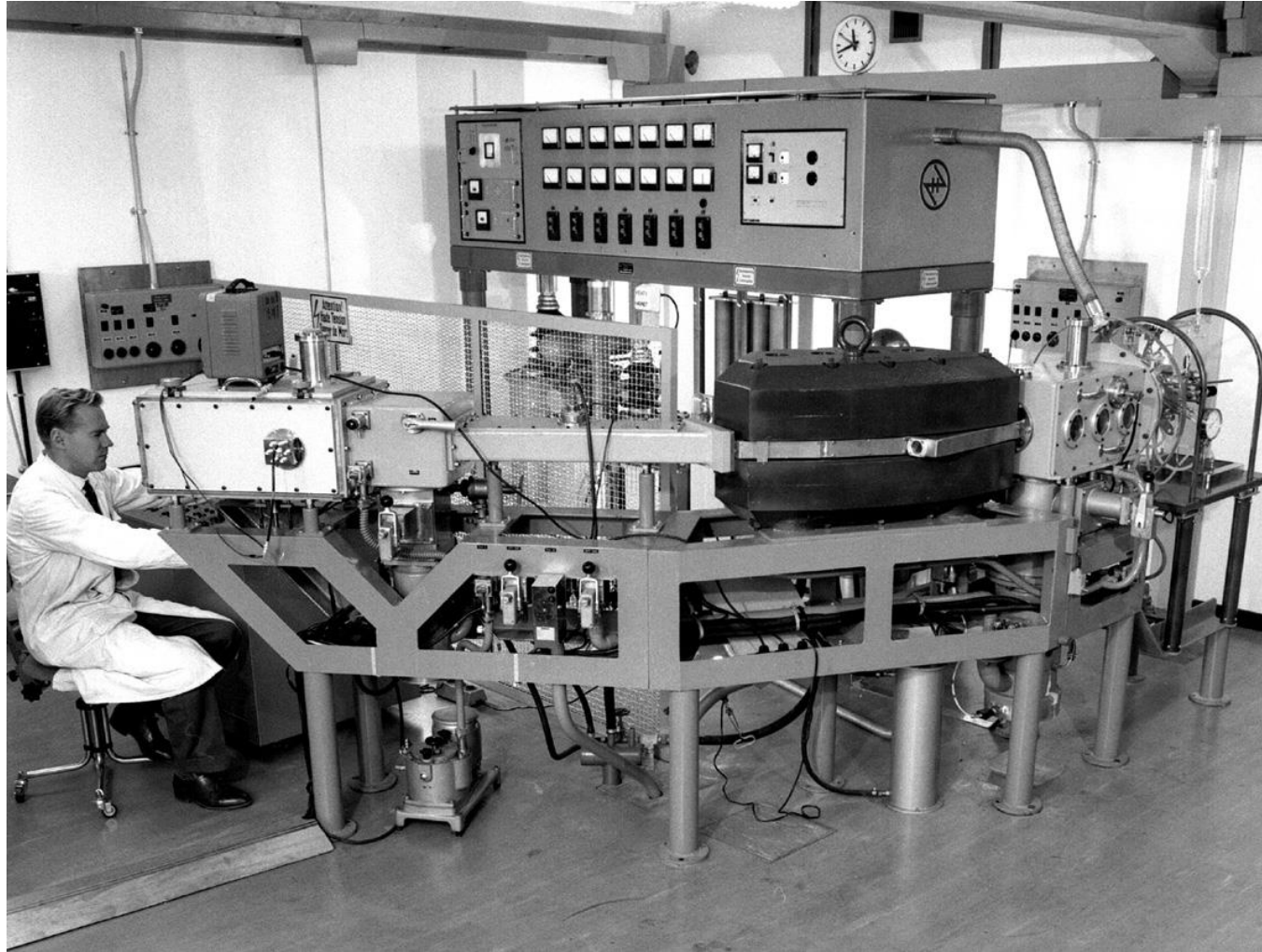
1'000 m/s

1'000'000 m/s

ISOL-history: irradiation of targets

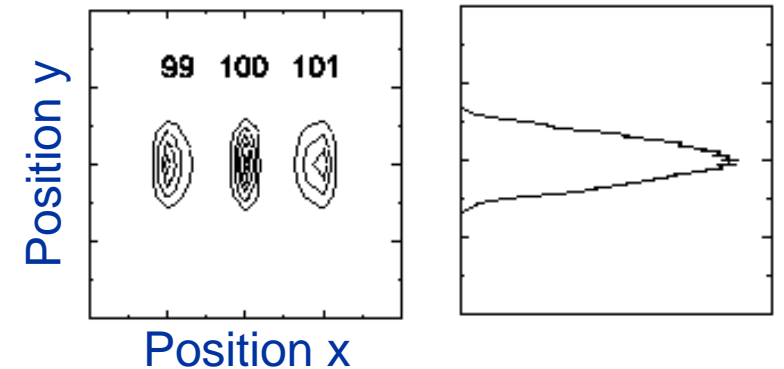
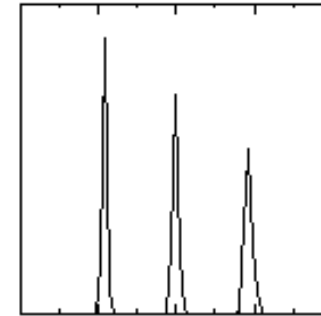


ISOL-history: off-line mass separation



- Ionization to typ. $q = 1+$
- Acceleration to e.g. 60 keV
- Mass selection by magnetic deflection (Lorentz force)

$$(B\rho = p/q \propto \sqrt{A})$$



ISOL-history: first ISOL experiment at Niels Bohr Institute

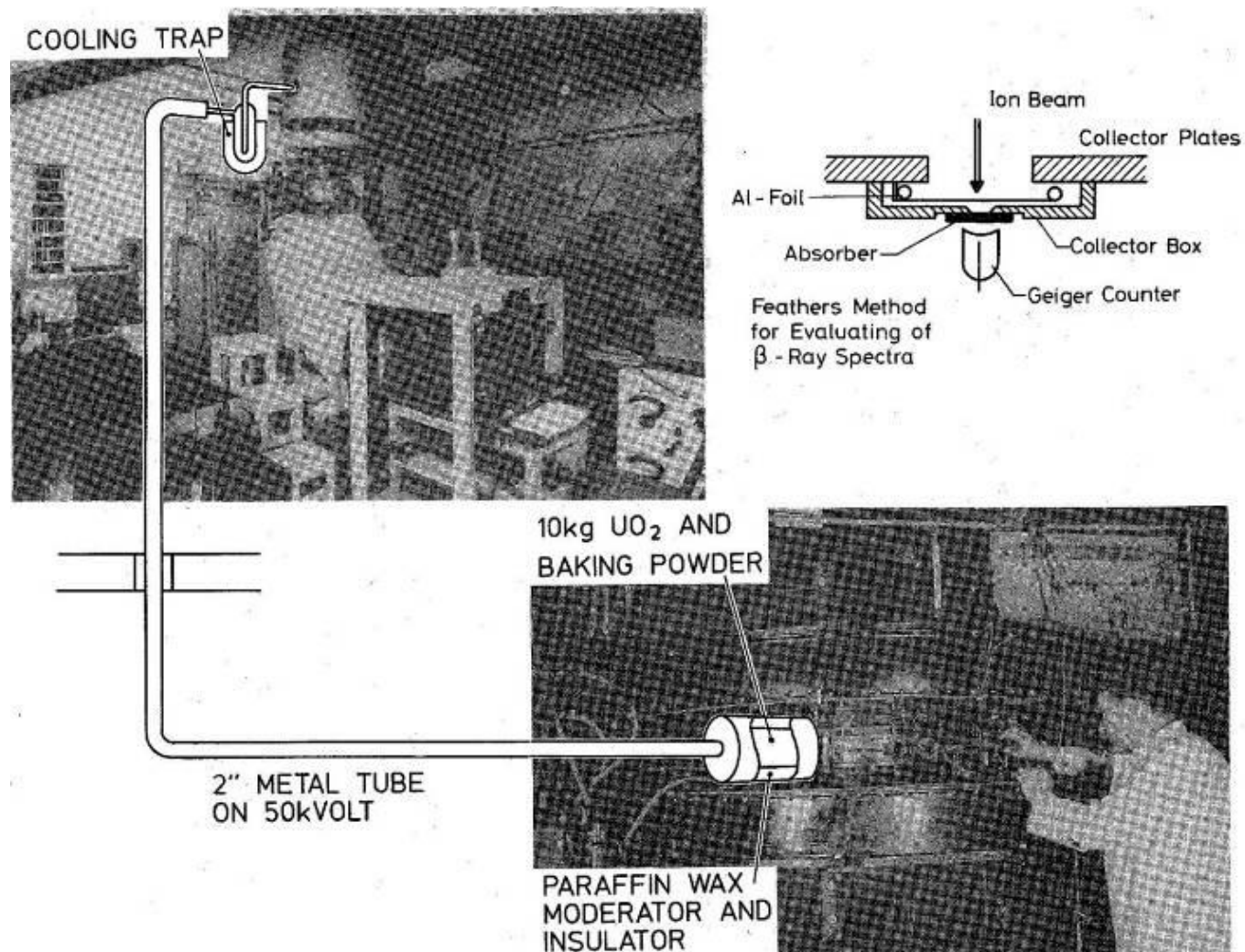
Short-Lived Krypton Isotopes and Their Daughter Substances

O. KOFOED-HANSEN AND K. O. NIELSEN

*Institute for Theoretical Physics, University of Copenhagen,
Copenhagen, Denmark*

(Received February 9, 1951)

THE isotopes Kr^{89} , Kr^{90} , Kr^{91} , and their daughter substances have been investigated. Krypton formed in fission of uranium was pumped through a 10-m long tube directly from the cyclotron into the ion source of the isotope separator. The cyclotron and the isotope separator were operated simultaneously, and the counting could begin immediately after the interruption of the separation. The rubidium and strontium daughter substances were separated chemically; strontium was precipitated as carbonate. Half-lives were measured and an absorption analysis of the radiations was carried out. The results are given in Table I.



ISOL beams of $^{89-93}\text{Kr}$

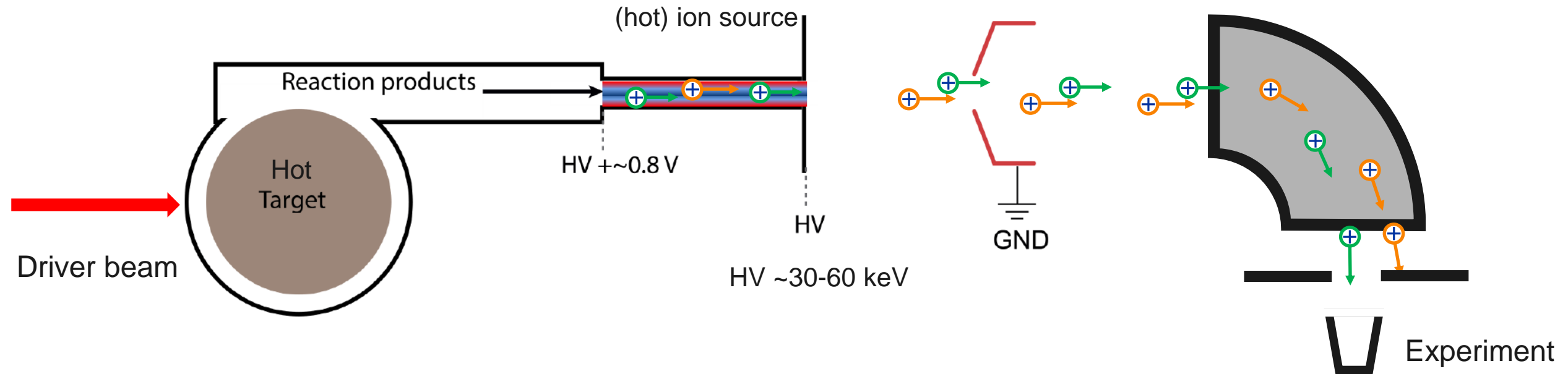
The isotope separation on-line (ISOL) method

(I) Production

(II) Ionization

Extraction
electrode

(III) Mass separation



Content

1. Why ISOL beams?, ISOL history

2. ISOL challenges

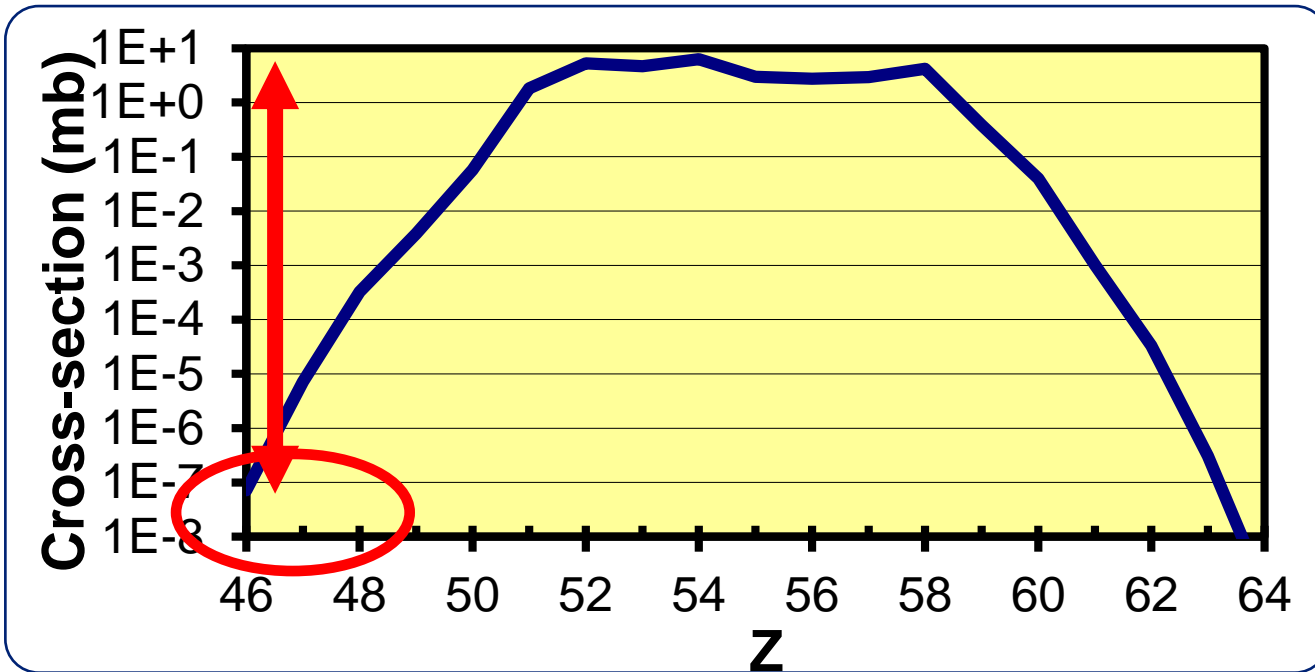
- **Challenges**
- **RIB optimization**

3. ISOL stages

4. Use cases around the world

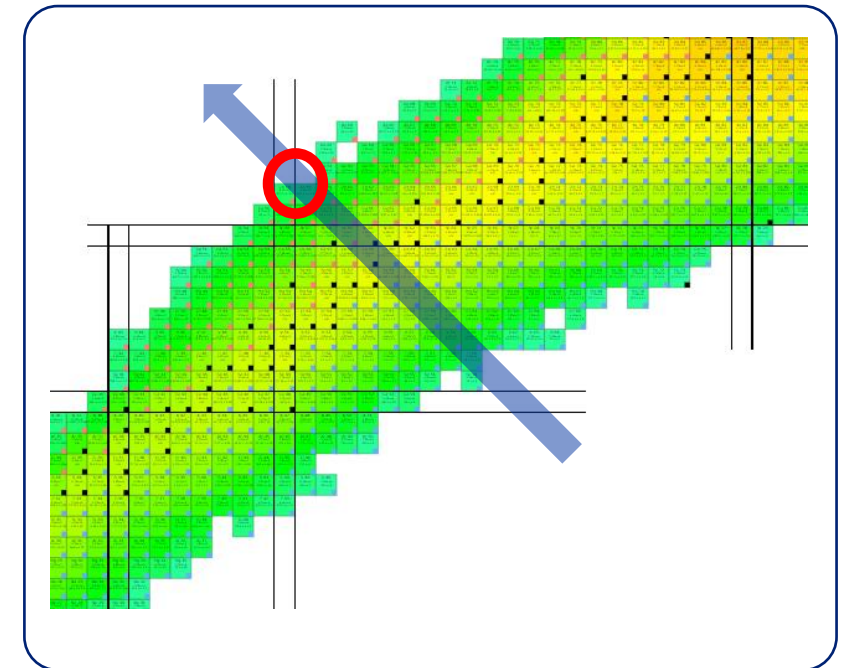
ISOL challenges

Isotope production



1. low cross-sections
2. enormous production of isobars
3. short half-lives

Isotope selection



- ⇒ optimize **efficiency**
- ⇒ optimize **selectivity**
- ⇒ optimize **rapidity**

ISOL: RIB optimization

Optimize event rate R of an experiment

$$\begin{aligned} & \text{In-target production} \quad \text{Efficiency} \\ & \underbrace{\hspace{10em}} \quad \underbrace{\hspace{5em}} \\ \mathbf{R} &= \mathbf{\Phi} \cdot \mathbf{\sigma} \cdot \mathbf{N}_t \cdot \mathbf{\varepsilon} \\ & \underbrace{\hspace{15em}} \\ \mathbf{\varepsilon} &= \mathbf{\varepsilon}_{\text{rel}} \cdot \mathbf{\varepsilon}_{\text{ion}} \cdot \mathbf{\varepsilon}_{\text{sep}} \cdot \mathbf{\varepsilon}_{\text{trans}} \\ & \underbrace{\hspace{10em}} \\ \mathbf{\varepsilon}_{\text{rel}} &= \mathbf{\varepsilon}_{\text{diff}} \cdot (\mathbf{\varepsilon}_f) \cdot \mathbf{\varepsilon}_{\text{eff}} \end{aligned}$$

To maximize event rate at experiment:

- Optimize in-target production
- Maximize efficiency cascade

Do for each beam!

σ : Cross section
 Φ : Pr. Particle flux
 N_t : Exposed target nuclei
 ε : Efficiency

ISOL: RIB optimization

Optimize event rate R of an experiment

$$R = \int_{E_f}^{E_i} dE \Phi \cdot \frac{\sigma(E)}{S(E)} \cdot N_t \cdot \varepsilon$$

In-target production **Efficiency**

$$\varepsilon = \varepsilon_{\text{rel}} \cdot \varepsilon_{\text{ion}} \cdot \varepsilon_{\text{sep}} \cdot \varepsilon_{\text{trans}}$$
$$\varepsilon_{\text{rel}} = \varepsilon_{\text{diff}} \cdot (\varepsilon_f) \cdot \varepsilon_{\text{eff}}$$

To maximize event rate at experiment:

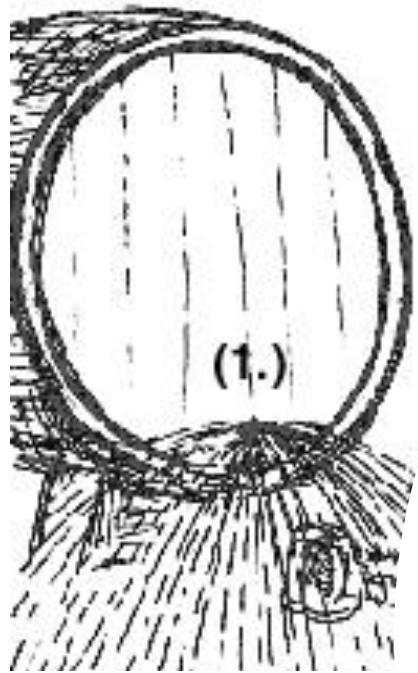
- Optimize in-target production
- Maximize efficiency cascade

Do for each beam!

σ : Cross section
 Φ : Pr. Particle flux
 N_t : Exposed target nuclei
 ε : Efficiency

ISOL: RIB optimization

All steps of the production-separation chain need to be



$$R = \Phi \cdot \sigma \cdot N_t \cdot \epsilon_{\text{rel}} \cdot \epsilon_{\text{ion}} \cdot \epsilon_{\text{sep}} \cdot \epsilon_{\text{trans}} \cdot \epsilon_{\text{det}}$$

powerful accelerator

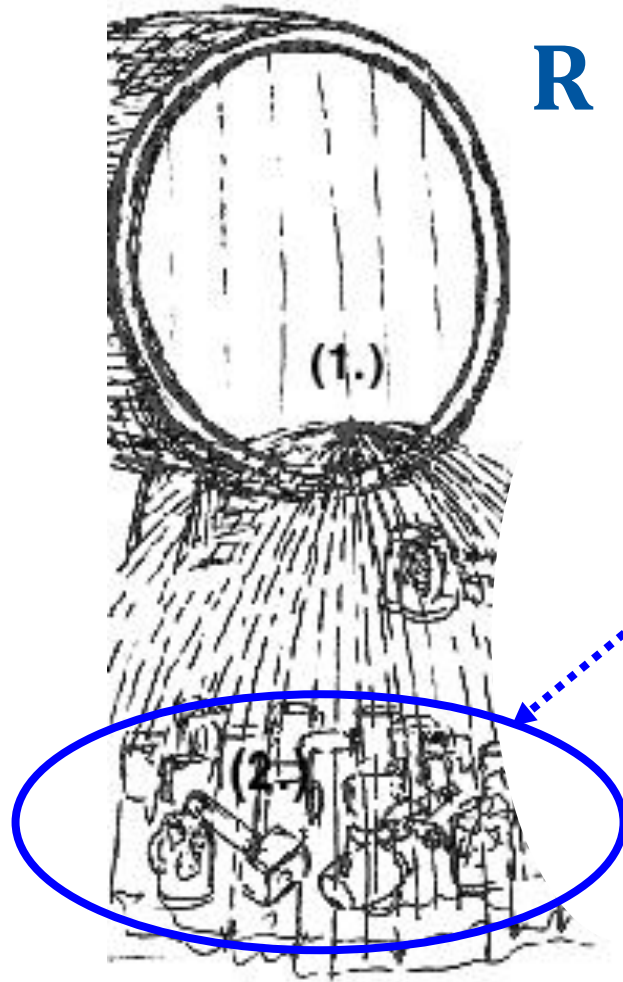
⇒ accelerator technology

σ : Cross section
 Φ : Pr. Particle flux
 N_t : Exposed target nuclei
 ϵ : Efficiency

ISOL: RIB optimization

All steps of the production-separation chain need to be optimized!

$$R = \Phi \cdot \sigma \cdot N_t \cdot \epsilon_{\text{rel}} \cdot \epsilon_{\text{ion}} \cdot \epsilon_{\text{sep}} \cdot \epsilon_{\text{trans}} \cdot \epsilon_{\text{det}}$$



high production cross-sections

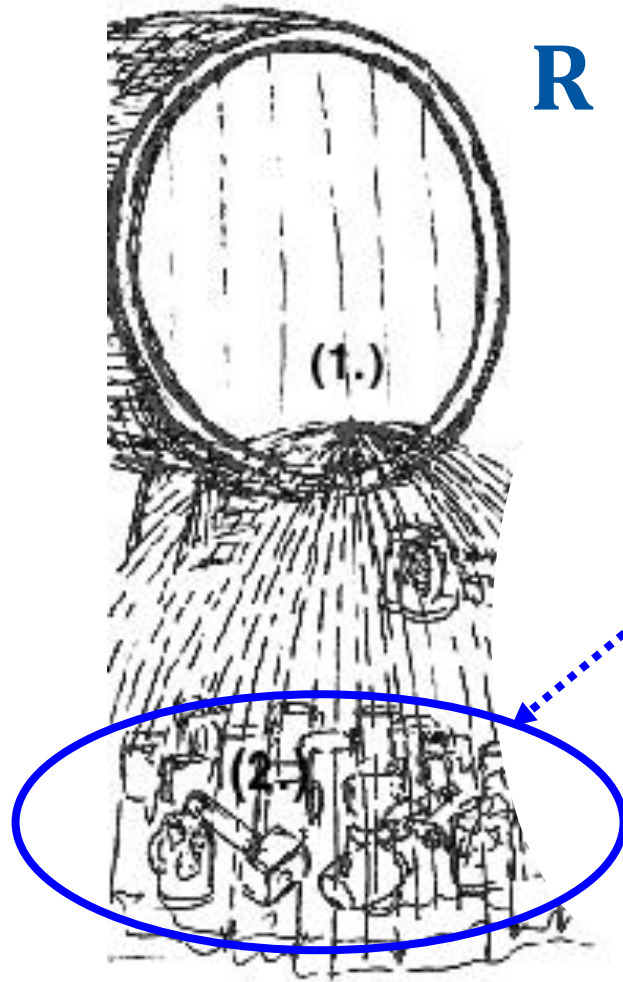
⇒ nuclear physics

σ : Cross section
 Φ : Pr. Particle flux
 N_t : Exposed target nuclei
 ϵ : Efficiency

ISOL: RIB optimization

All steps of the production-separation chain need to be optimized!

$$R = \Phi \cdot \sigma \cdot N_t \cdot \epsilon_{\text{rel}} \cdot \epsilon_{\text{ion}} \cdot \epsilon_{\text{sep}} \cdot \epsilon_{\text{trans}} \cdot \epsilon_{\text{det}}$$



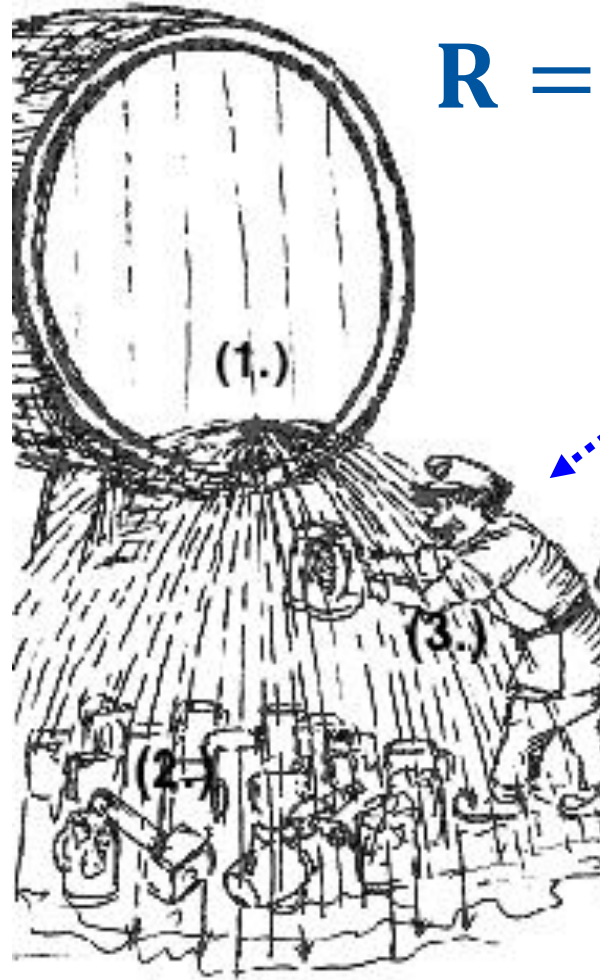
reliable “thick” targets

⇒ material science

σ : Cross section
 Φ : Pr. Particle flux
 N_t : Exposed target nuclei
 ϵ : Efficiency

ISOL: RIB optimization

All steps of the production-separation chain need to be optimized!



$$R = \Phi \cdot \sigma \cdot N_t \cdot \epsilon_{\text{rel}} \cdot \epsilon_{\text{ion}} \cdot \epsilon_{\text{sep}} \cdot \epsilon_{\text{trans}} \cdot \epsilon_{\text{det}}$$

Extraction efficiency from target determined by:

- diffusion

⇒ solid state physics

- surface desorption

⇒ surface chemistry

- effusion

⇒ gas phase chemistry

strongly element dependent!

σ : Cross section

Φ : Pr. Particle flux

N_t : Exposed target nuclei

ϵ : Efficiency

ISOL: RIB optimization

All steps of the production-separation chain need to be optimized!



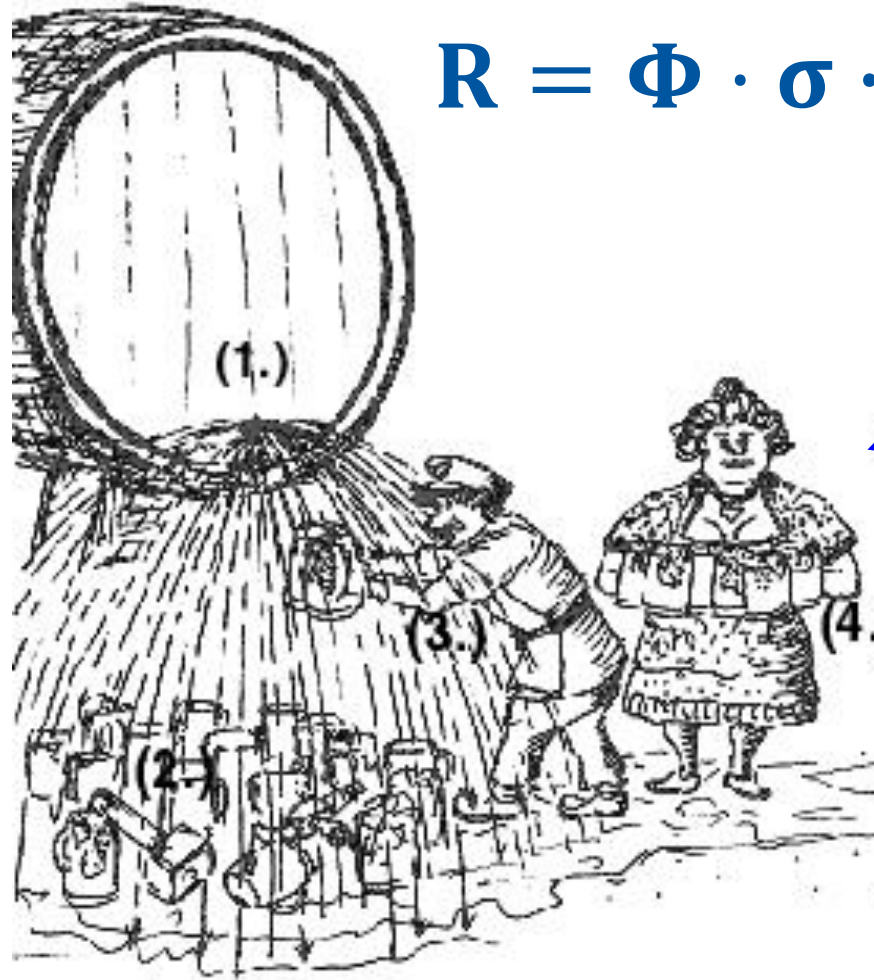
$$R = \Phi \cdot \sigma \cdot N_t \cdot \epsilon_{\text{rel}} \cdot \epsilon_{\text{ion}} \cdot \epsilon_{\text{sep}} \cdot \epsilon_{\text{trans}} \cdot \epsilon_{\text{det}}$$

high ionization and extraction efficiency

⇒ ion source technology

ISOL: RIB optimization

All steps of the production-separation chain need to be optimized!



$$R = \Phi \cdot \sigma \cdot N_t \cdot \epsilon_{\text{rel}} \cdot \epsilon_{\text{ion}} \cdot \epsilon_{\text{sep}} \cdot \epsilon_{\text{trans}} \cdot \epsilon_{\text{det}}$$

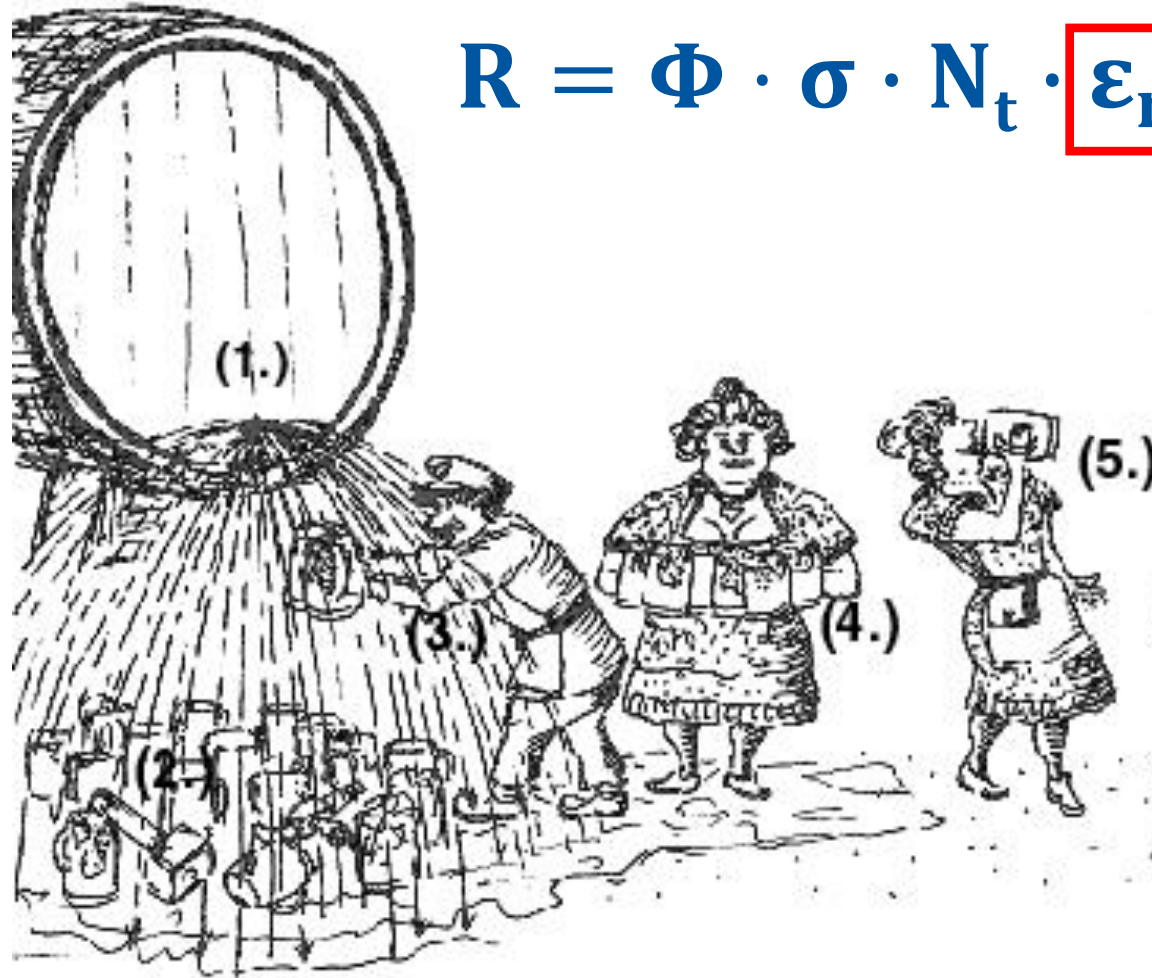
efficient separation and
transport of RIB

⇒ ion optics

σ : Cross section
 Φ : Pr. Particle flux
 N_t : Exposed target nuclei
 ϵ : Efficiency

ISOL: RIB optimization

All steps of the production-separation chain need to be optimized!



$$R = \Phi \cdot \sigma \cdot N_t \cdot \epsilon_{\text{rel}} \cdot \epsilon_{\text{ion}} \cdot \epsilon_{\text{sep}} \cdot \epsilon_{\text{trans}} \cdot \epsilon_{\text{det}}$$

Mind the decay losses during delays

⇒ efficiency strongly half-life dependent

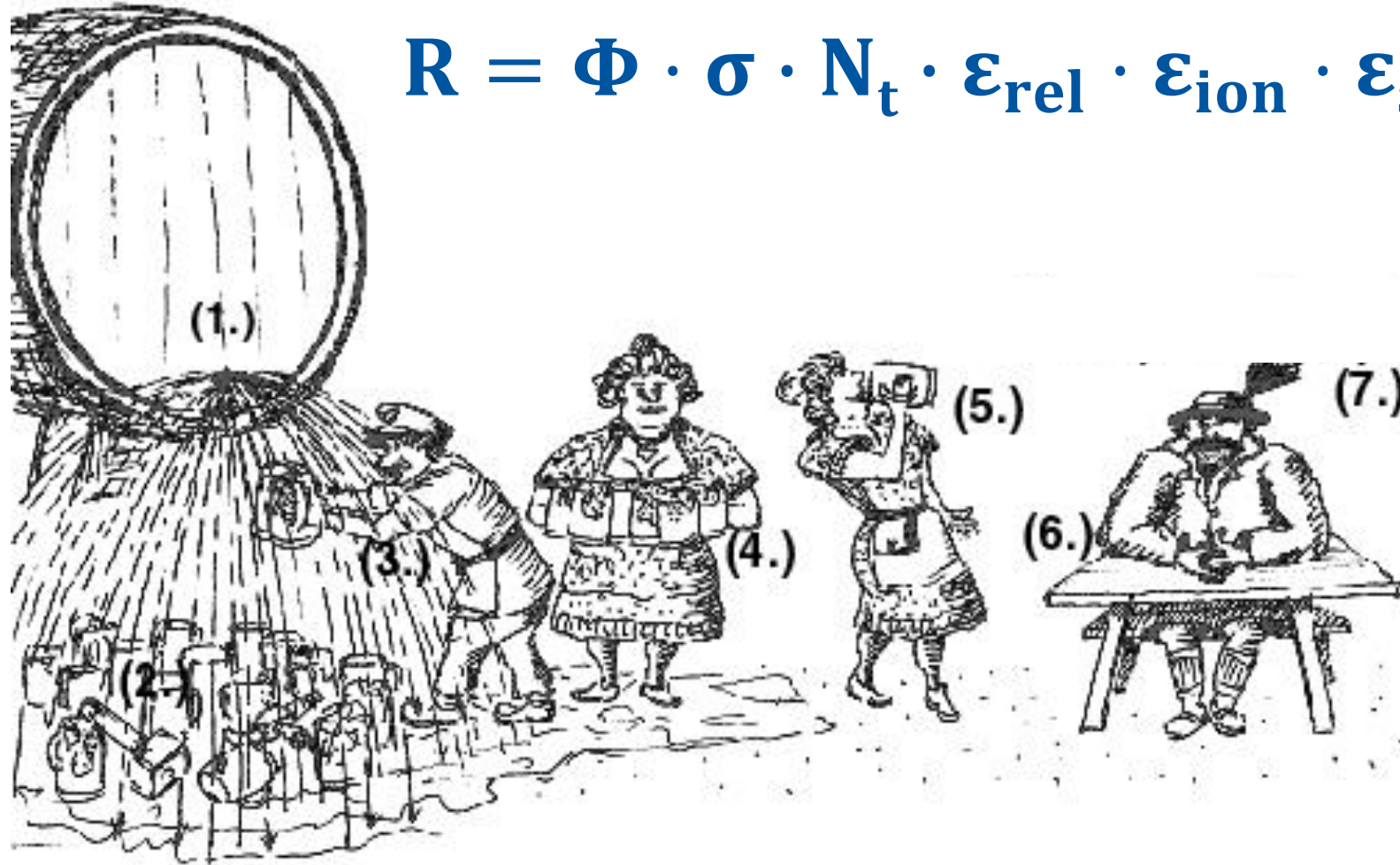
⇒ rapid extraction required!

σ : Cross section
 Φ : Pr. Particle flux
 N_t : Exposed target nuclei
 ϵ : Efficiency

ISOL: RIB optimization

All steps of the production-separation chain need to be optimized!

$$R = \Phi \cdot \sigma \cdot N_t \cdot \epsilon_{\text{rel}} \cdot \epsilon_{\text{ion}} \cdot \epsilon_{\text{sep}} \cdot \epsilon_{\text{trans}} \cdot \epsilon_{\text{det}}$$



Detection efficiency

σ : Cross section
 Φ : Pr. Particle flux
 N_t : Exposed target nuclei
 ϵ : Efficiency

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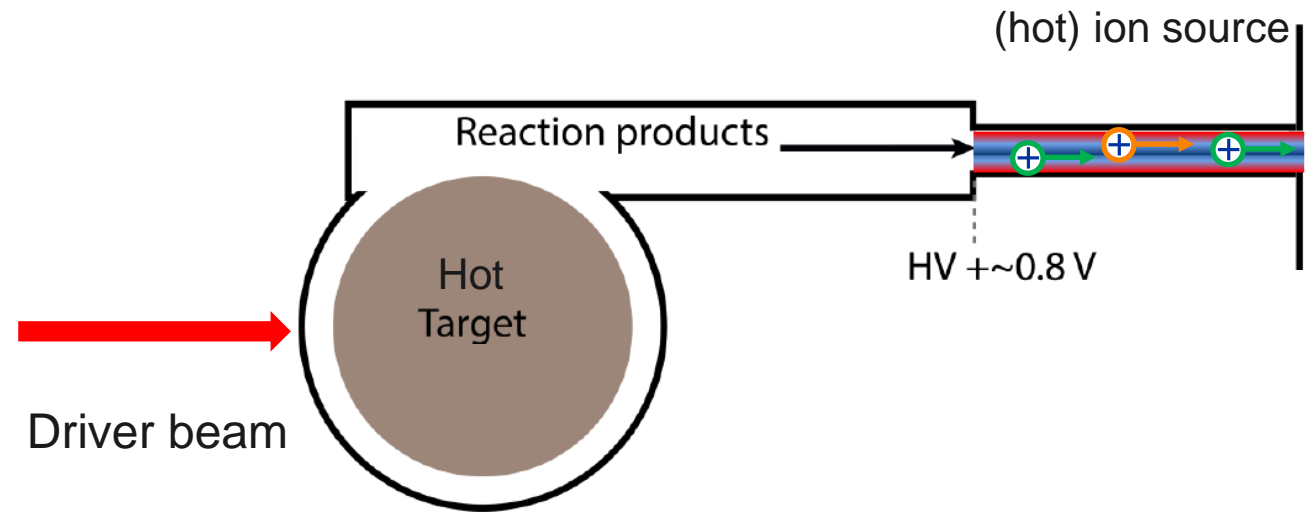
3. ISOL stages

- **Production**
- **Ionization**
- **Mass separation**

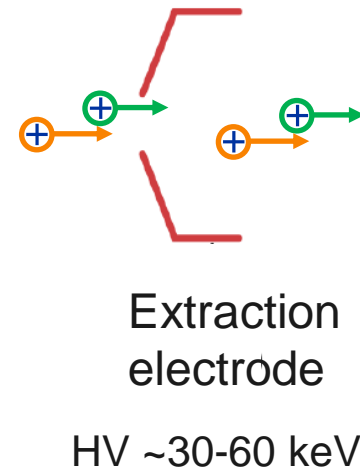
4. Use cases around the world

ISOL: stages

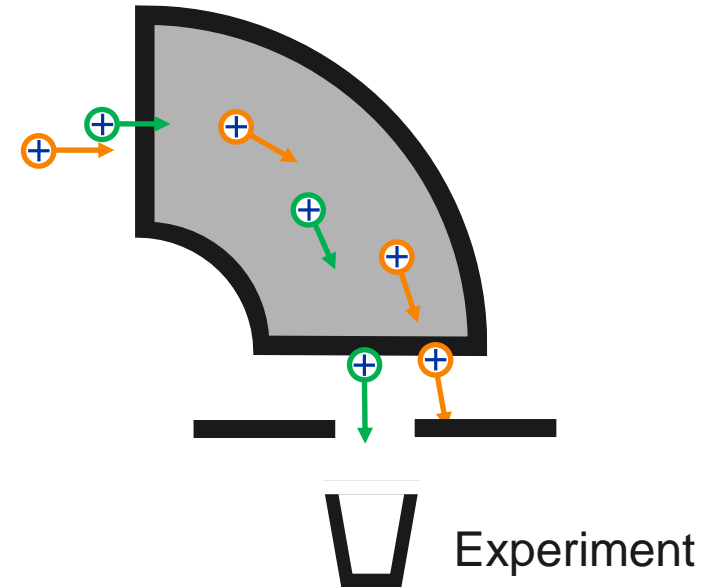
(I) Production



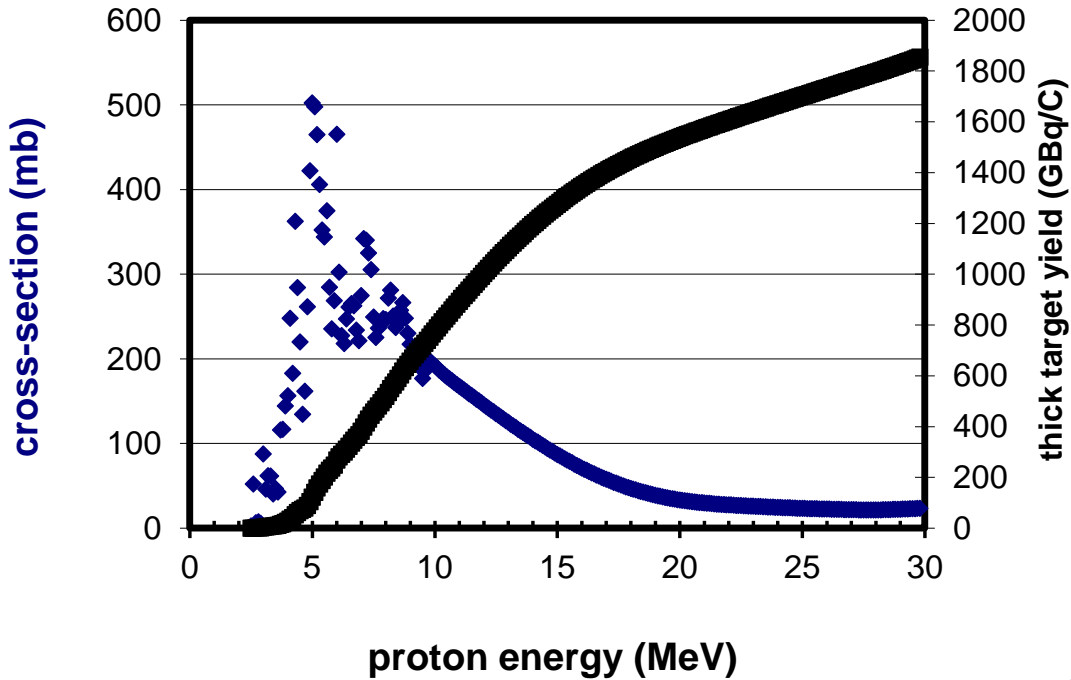
(II) Ionization



(III) Mass separation



(I) Isotope production: Direct reactions

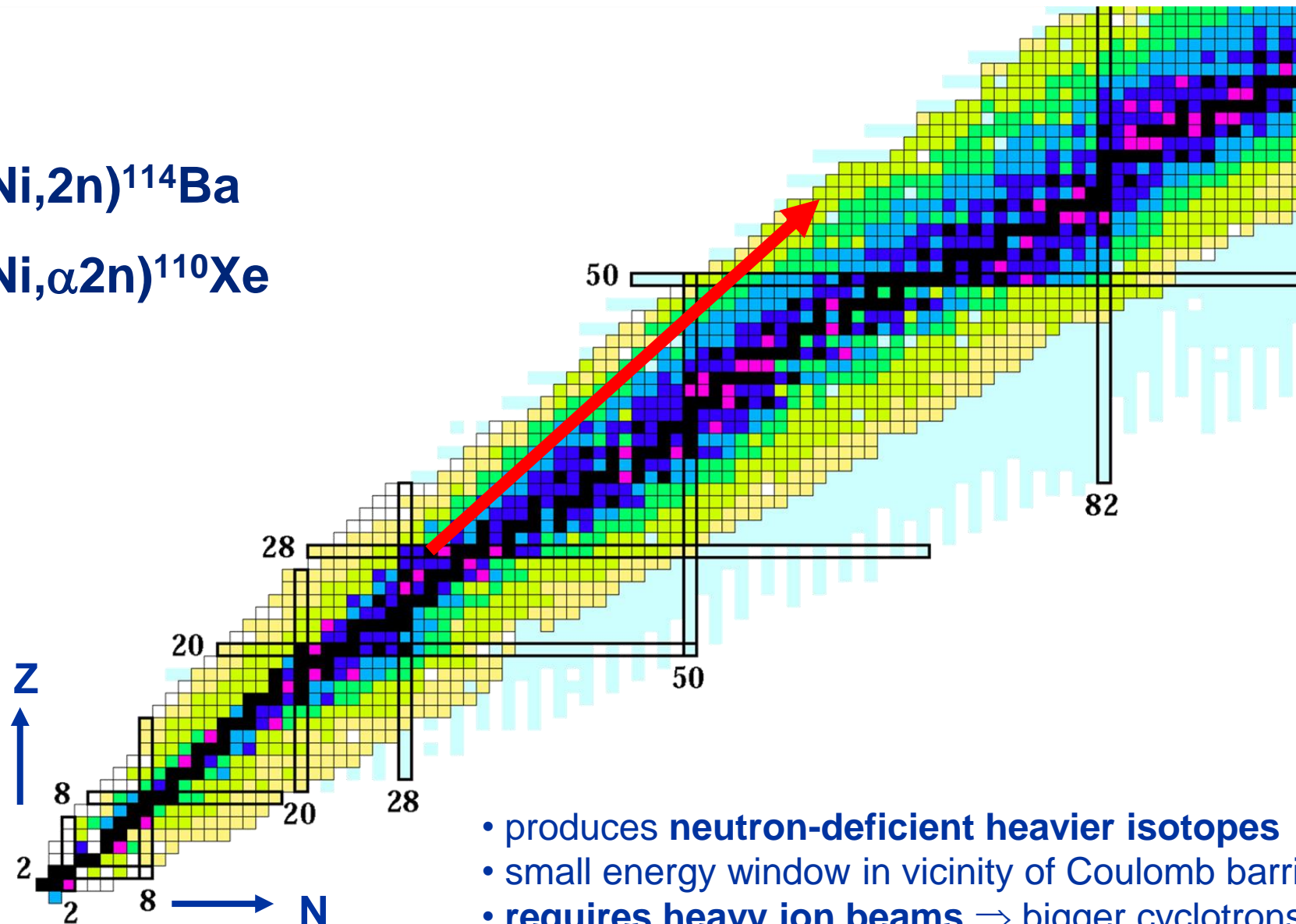


β^+ 8.0, 13.5... β_p 4.60, 3.80 5.12... $\beta\alpha$ 1.725... γ 495, 6129*...	β^+ 3.4... γ 1042...	β^+ 2.2... γ (110, 197 1357...)	σ 0.037	σ 0.666 $\sigma_{n,\alpha}$ 0.00018	σ 0.0527	β^- 4.4... γ 440, 1637...
F 16 40 keV $11 \cdot 10^{-21}$ s	F 17 64.49 s	F 18 109.728 m	F 19 100	F 20 11.0 s	F 21 4.158 s	F 22 4.23 s
β^+ 1.7... $\gamma?$	β^+ 0.634 no γ	σ 0.00951	β^- 5.4... γ 1634...	β^- 5.3, 5.7... γ 351, 1395...	β^- 5.5, 9.6... γ 1275, 2083... 2166, 4366...	
O 15 122.24 s	O 16 99.757	O 17 0.038	O 18 0.205	O 19 26.476 s	O 20 13.5 s	O 21 3.42 m
β^+ 1.732 no γ	σ 0.000173	σ 0.000540 $\sigma_{n,\alpha}$ 0.235	σ 0.000150	β^- 3.3, 4.8... γ 197, 1357...	β^- 2.8... γ 1057...	β^- 6.4... γ 1730, 3511... 280, 1787 1755...
N 14 99.636	N 15 0.364	N 16 5.25 μ s 7.13 s	N 17 4.173 s	N 18 0.619 ms	N 19 336 ms	N 20 136 m
σ 0.080, $\sigma_{n,p}$ 1.86	σ 0.000024	IT 120 β^- ...	β^- 4.3 10.4... γ 6129 7115... $\beta\alpha$ 1.76...	β^- 3.2, 8.7... βn 1.16, 0.39... γ 871, 2184 $\beta\alpha$ 1.25, 1.41	β^- 9.4, 11.9... γ 1982, 822 1652, 2473... $\beta\alpha$ 1.08, 2.28... βn 0.58, 2.44...	β^- 96, 1983*, 3851 1376, 2475... βn 1.054, 0.452 2.655...
β^- 1.08, 2.28... βn 0.58, 2.44...	β^- 96, 1983*, 3851 1376, 2475... βn 1.054, 0.452 2.655...					β^-, γ 1674, 9... 1376*... βn 2.071 1.098...
C 13 1.07	C 14 5730 a	C 15 2.449 s	C 16 0.747 s	C 17 193 ms	C 18 92 ms	C 19 46.2 m

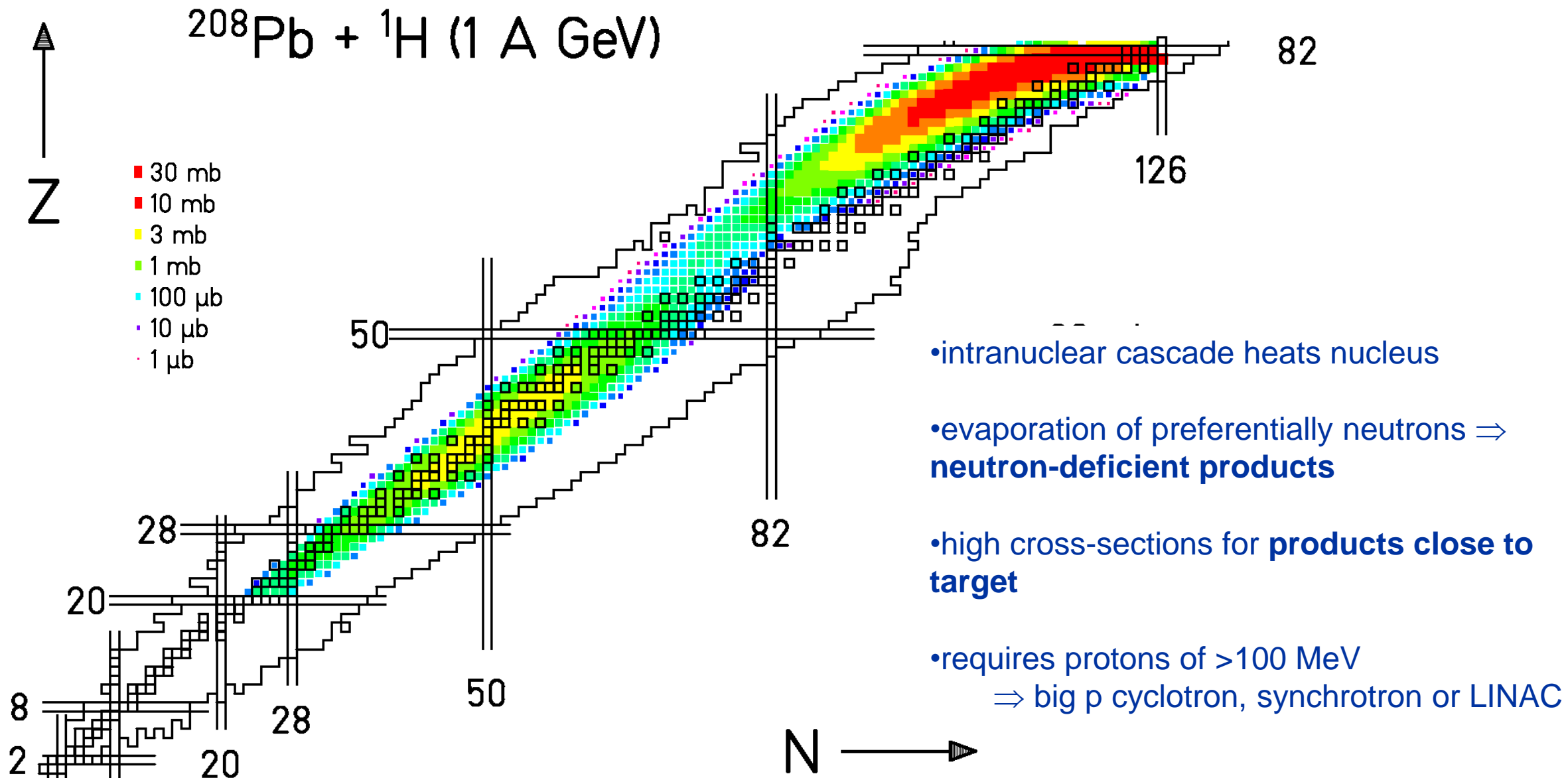
(p,n), (^3He ,n), (α ,n), (n, α), ...

- high cross-sections, **products relatively close to stability**
- driver beams from (low-cost) cyclotrons

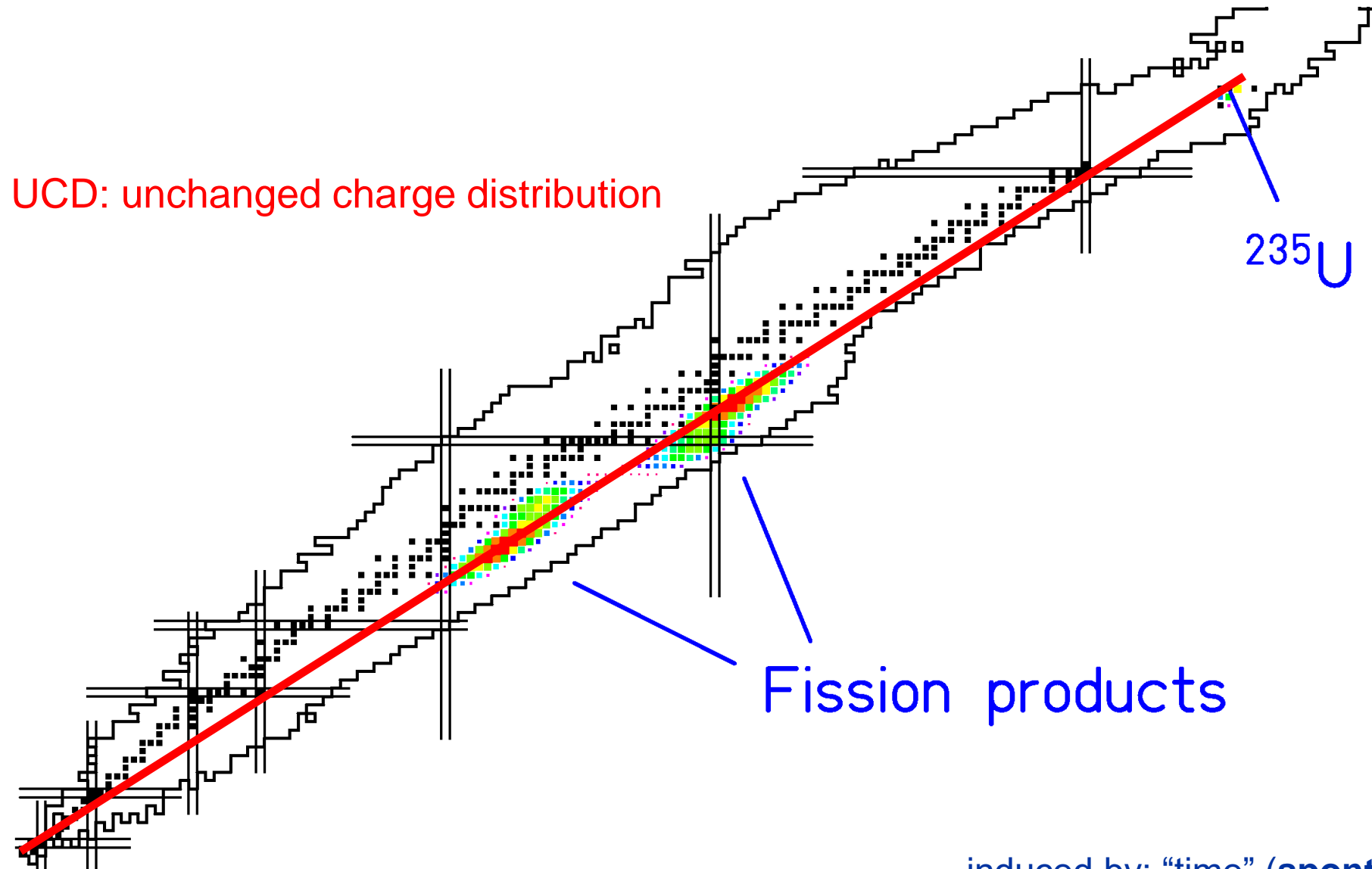
(I) Isotope production: Fusion-evaporation



(I) Isotope production: Spallation

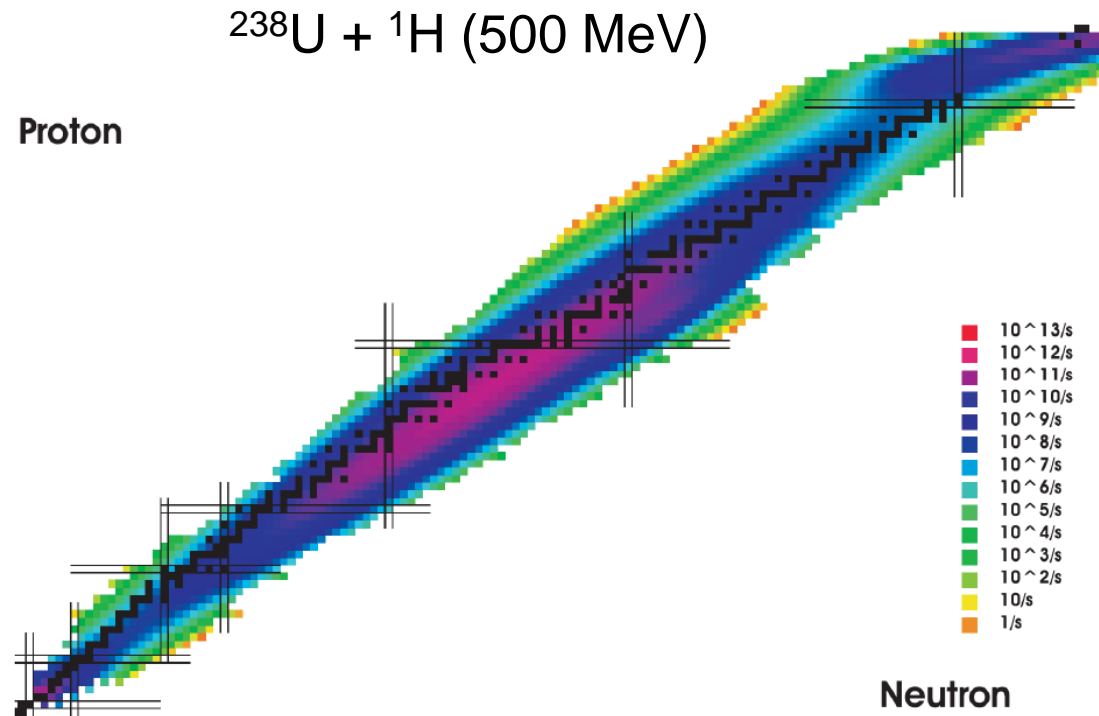


(I) Isotope production: Low energy fission



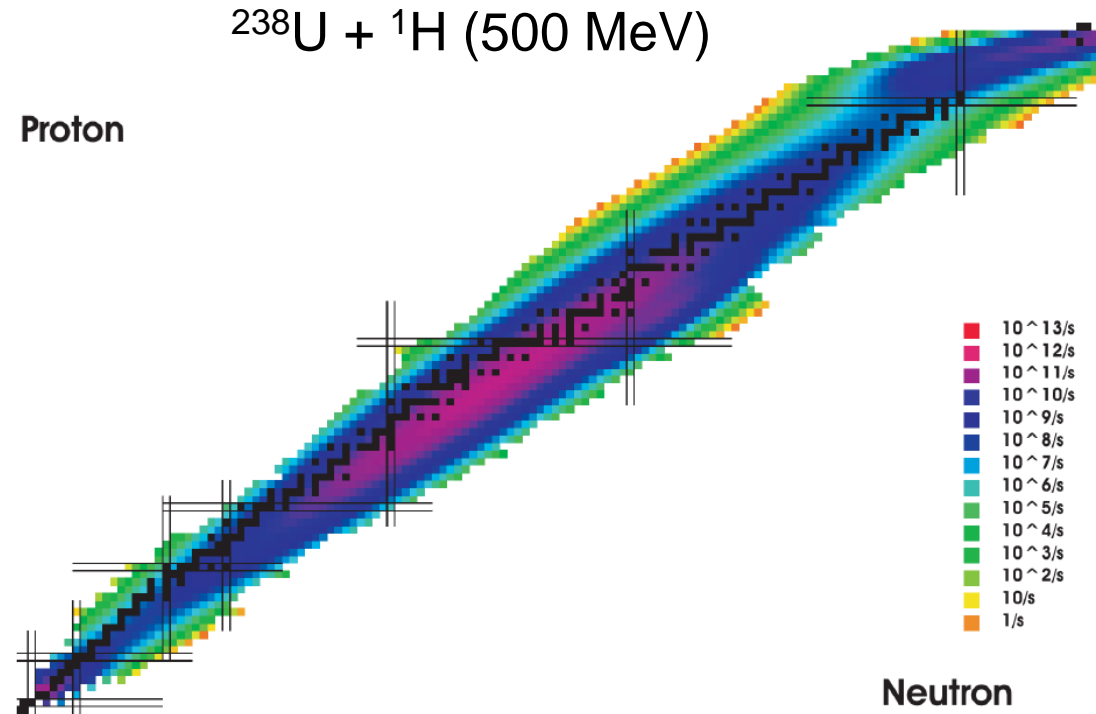
induced by: "time" (spontaneous)

(I) Isotope production: High energy fission



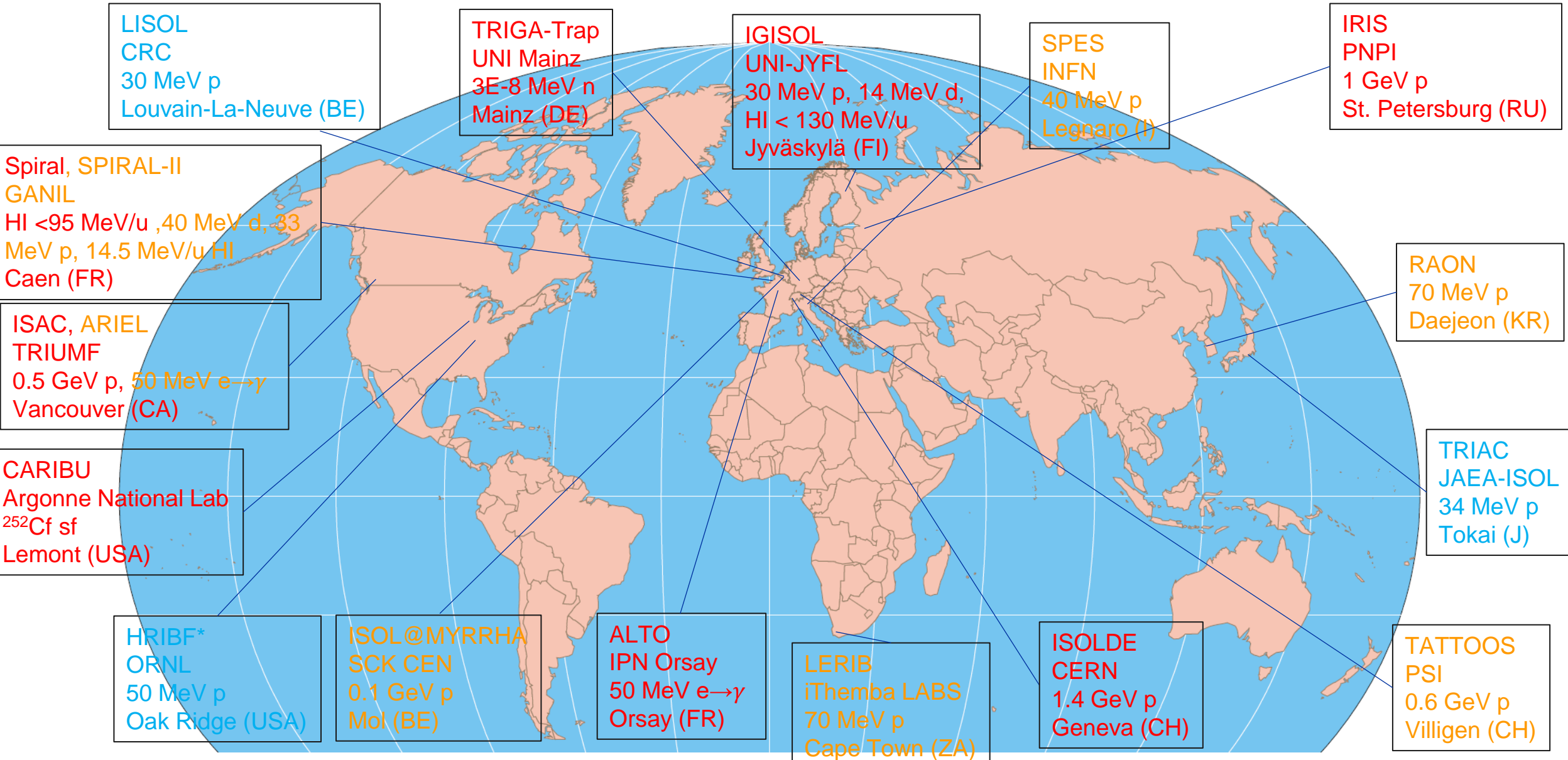
- induced by: **neutrons**, photons, **protons**, heavy ions, antiprotons, pions, post fusion-evaporation, beta-decay/EC
- with increasing excitation energy **symmetric and far asymmetric fission is favored**, but the products get in average less neutron-rich!
- driver accelerators: reactors, medium-energy (some MeV to tens MeV) deuterons from cyclotron or LINAC, microtron or LINAC for electron beams,...

(I) Isotope production: Fragmentation



- target fragmentation needs high energy protons
- Projectile fragmentation needs high energy heavy ions
- Lower-Z fragments $Z \sim \leq 23$ (U)
- Needs, big cyclotrons, synchrotron or LINAC

Variants of ISOL facilities (past and present, future, non exhaustive)



Variants of ISOL facilities (**past** and **present**, **future** non exhaustive)

1a **protons on thick (heavy) target: fragmentation, spallation, fission**

ISOLDE-CERN (1.4 GeV), IRIS-PNPI (1 GeV), ISAC-TRIUMF (0.5 GeV), TATTOOS (0.6 GeV), ISOL@MYRRHA (0.1(6) GeV) (SCK CEN), SPES (INFN), SPIRAL2 (GANIL), LERIB (iThemba LABS), RAON

1b **direct reactions** in thick target

CRC Louvain-la-Neuve, HRIBF Oak Ridge, TRIAC Tokai, SPES (INFN), LERIB (iThemba LABS)

1c (photo) **fission** in thick target

OSIRIS (Studsvik), HRIBF Oak Ridge, TRIAC Tokai, SPIRAL2 (GANIL), ARIEL (TRIUMF), ALTO

2 **projectile fragmentation** in **thick (carbon) target**

SPIRAL (GANIL), DRIBS (Dubna), EXCYT (LNS Catania)

3 **fusion-evap.** or multinucleon transfer in **thin target** plus **solid catcher**

GSI-ISOL, UNIRIB (ORNL), DOLIS (Daresbury), LISOL (Leuven), IMP Lanzhou, MASHA (Dubna), SPIRAL2 (GANIL)

4 **fusion-evap.**, **direct reaction** or **fission** in **thin target** plus **gas catcher** (Ion Guide ISOL =IGISOL)

IGISOL (Jyväskylä), LISOL (Leuven), ...

(I) Isotope production: target material

Example: ISOLDE

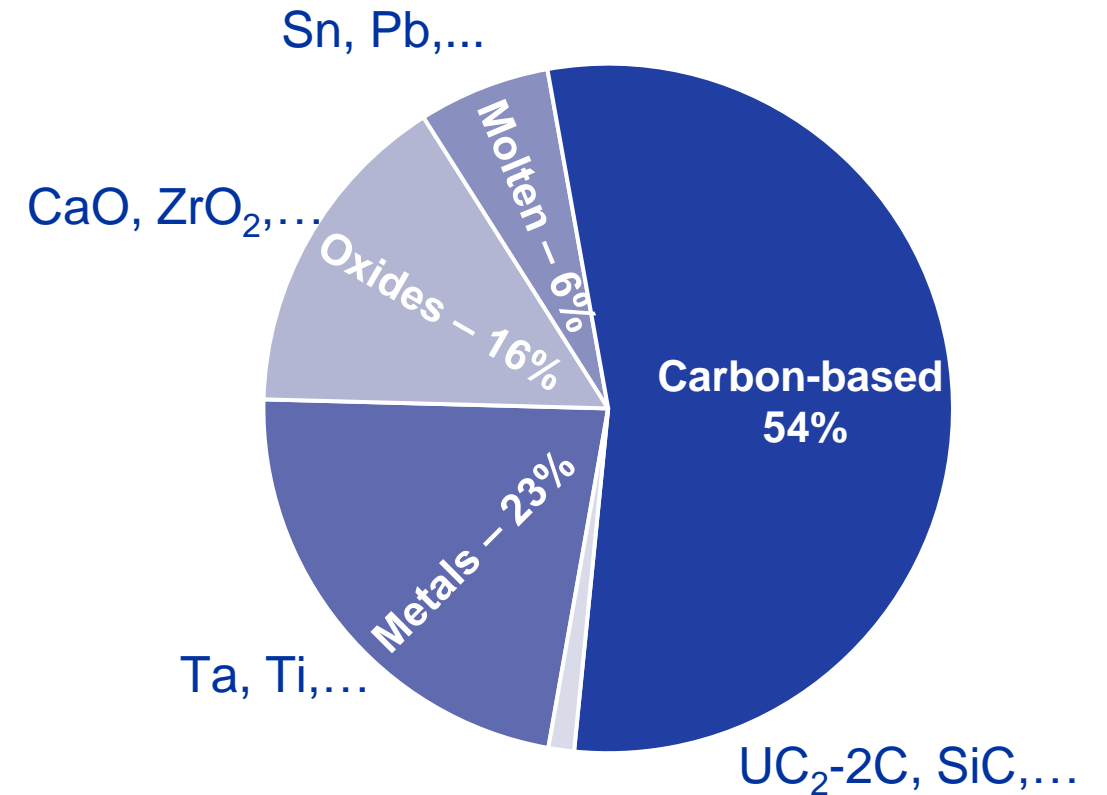
Material requirements

- High **production cross section** of the isotope(s) of interest
- Stability at **high temperatures**
- Chemically **stable and inert**
- Resistance to radiation damage
- Rapid **diffusion** and **effusion** rates of the element(s) of interest

Operation temperature limitations:

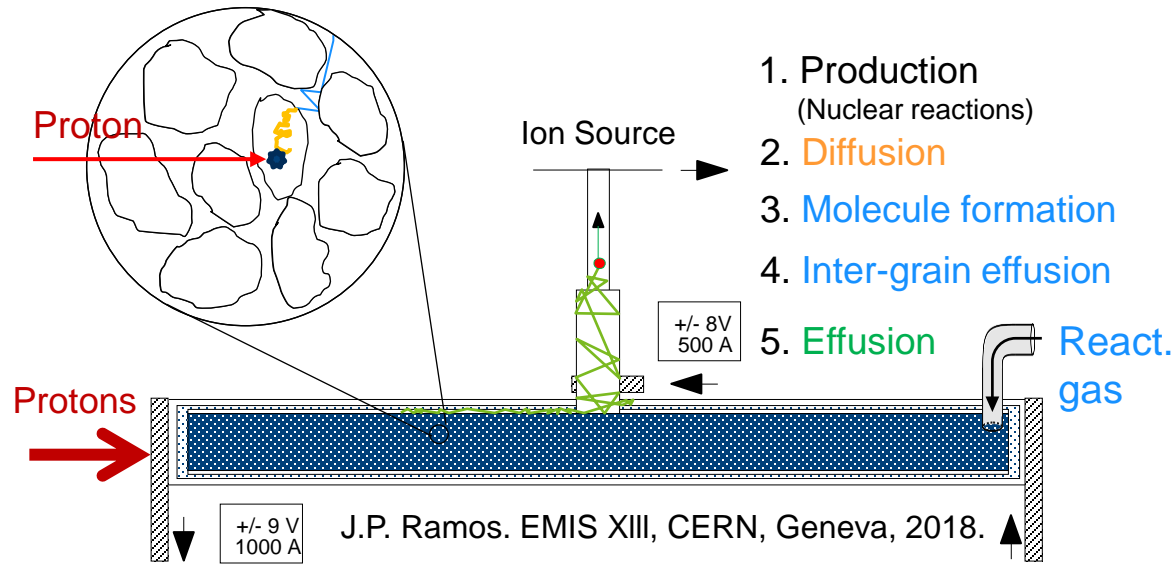
- **Sintering** (preserve target microstructure)
- Limited reactivity with surrounding materials
- Reduced stable beam contaminants (chemical impurities)
- Moderate equilibrium vapor pressure compatible with ion source ($\sim 10^{-4}$ mBar)

Target materials in the last 16 years



Powders, pellets or liquid form

(I) Isotope production: target material



$$R = \Phi \cdot \sigma \cdot N_t \cdot \epsilon_{\text{rel}} \cdot \epsilon_{\text{ion}} \cdot \epsilon_{\text{sep}} \cdot \epsilon_{\text{trans}} \cdot \epsilon_{\text{det}}$$

$$\epsilon_{\text{rel}} = \epsilon_{\text{diff}} \cdot (\epsilon_f) \cdot \epsilon_{\text{eff}}$$

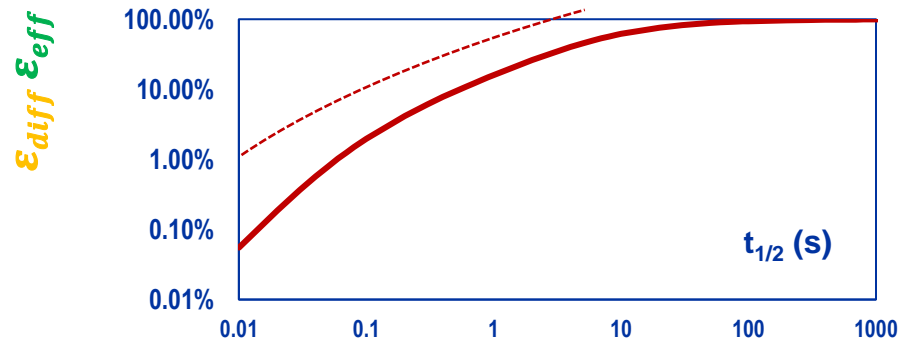
σ : \longleftrightarrow target-isotope chemistry, T-boundaries

N_t : High $\rho \rightarrow$ high production \longleftrightarrow low release

ϵ_{rel} : Optimize microstructure, stable at operation conditions

(I) Isotope production: target microstructure

In most cases $\epsilon_{diff}\epsilon_{eff}$ limit by far the yields.



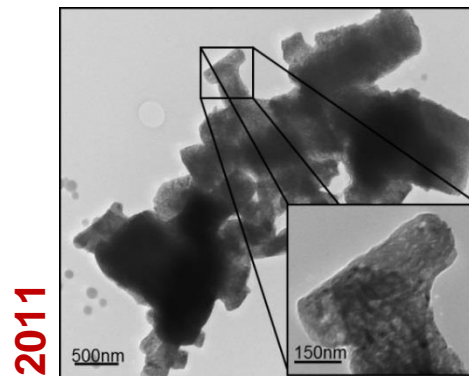
Diffusion limited release: $\lambda \leq 2\mu$

$$\epsilon_{diff} = \frac{3}{\pi} \sqrt{\frac{\mu}{\lambda}}, \mu = \frac{\pi^2 D}{r^2} \rightarrow r/10 \rightarrow \epsilon_{diff} \times 10$$

Same T!

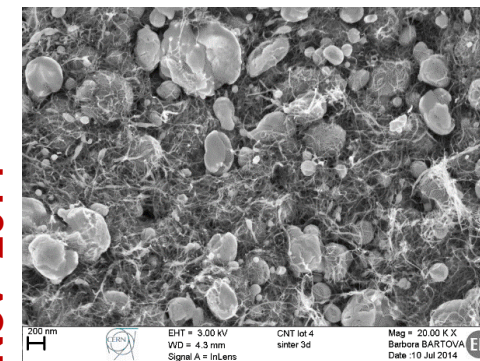
Operation T \longleftrightarrow Sintering - Grain size/Porosity

Specific surface area \longleftrightarrow Reactivity



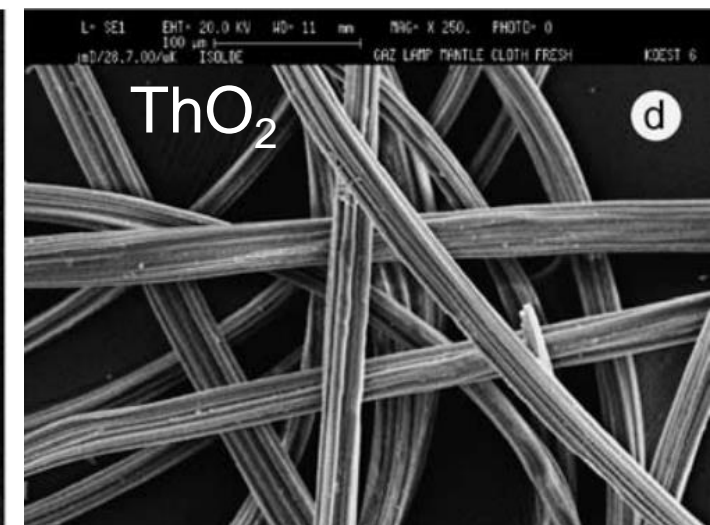
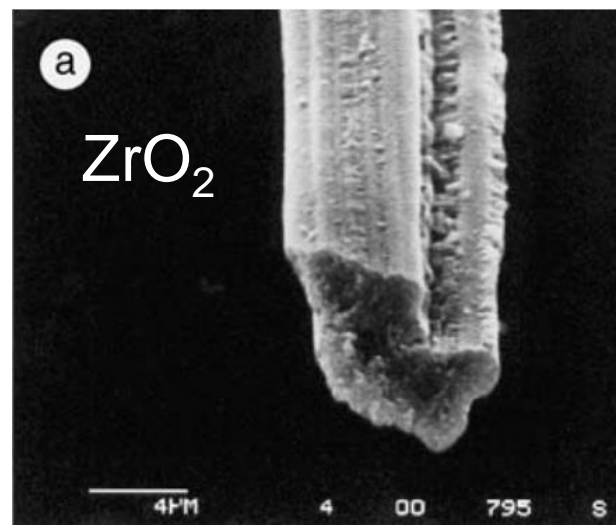
2011

CaO – J.P. Ramos, et al.

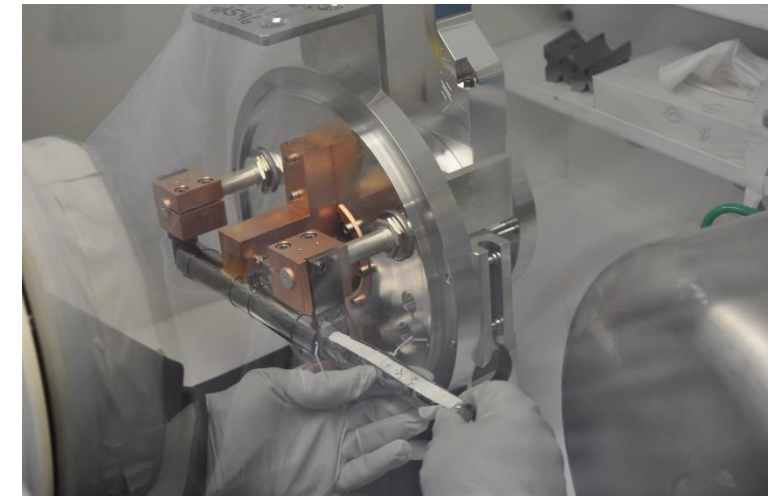
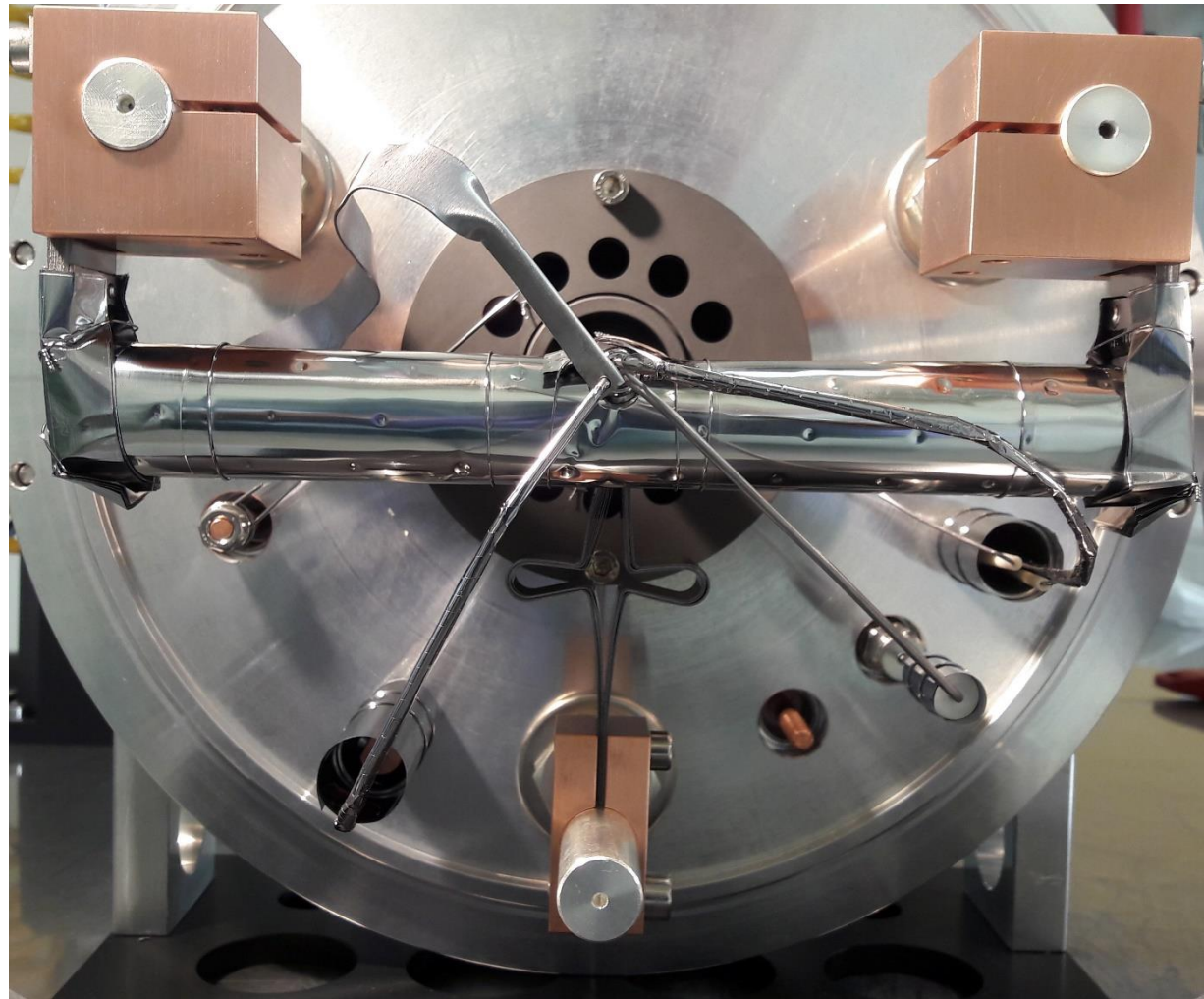
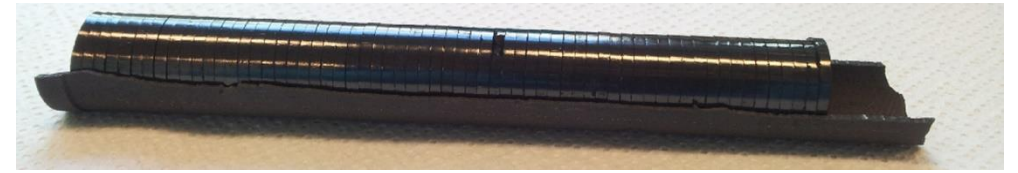


Nov-2014

LaC₂ + 2C – J. Guillot, et al.

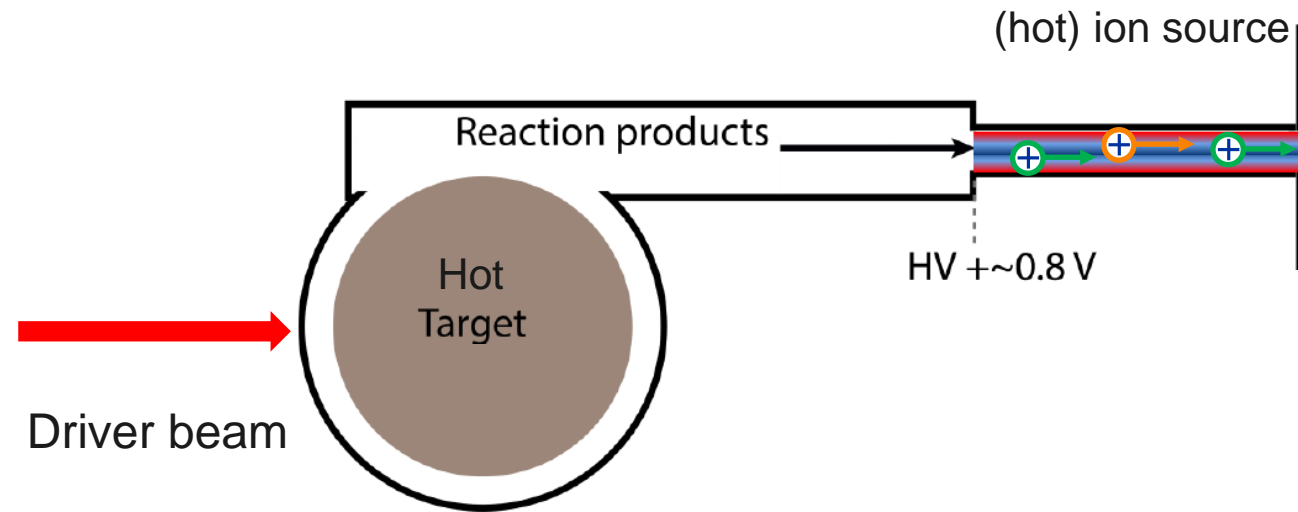


Example ISOLDE :Target and ion source unit

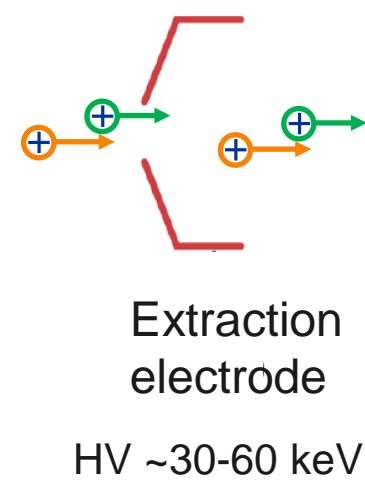


ISOL: stages

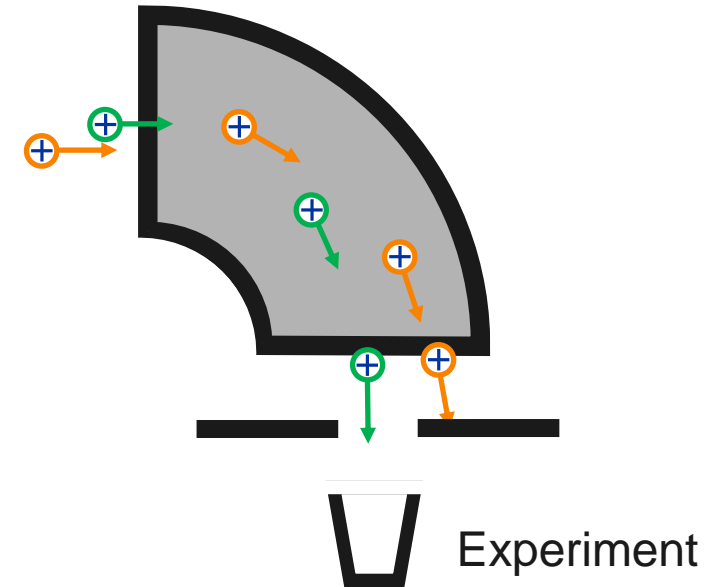
(I) Production



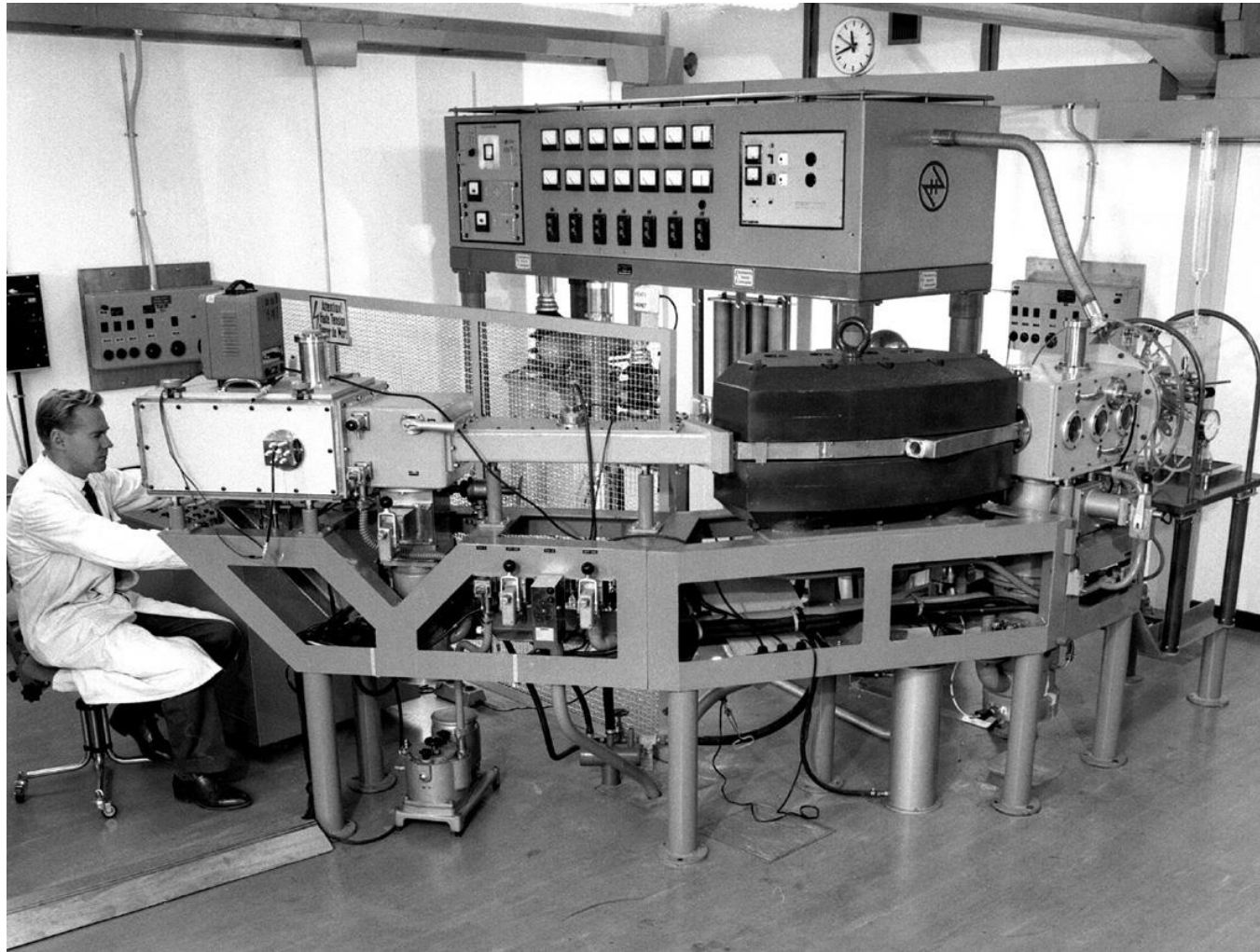
(II) Ionization



(III) Mass separation

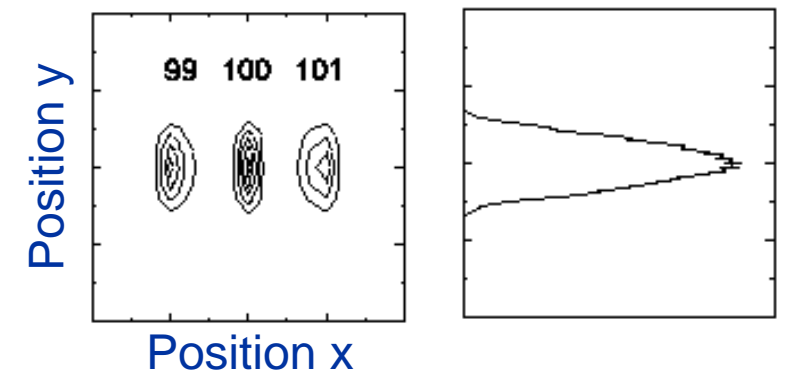
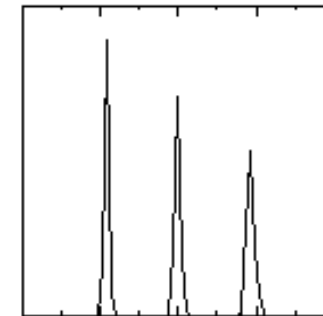


(III) Mass separation



- Ionization to typ. $q = 1+$
- Acceleration to e.g. 60 keV
- Mass selection by magnetic deflection (Lorentz force)

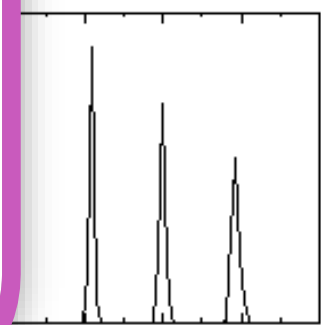
$$(B\rho = p/q \propto \sqrt{A})$$



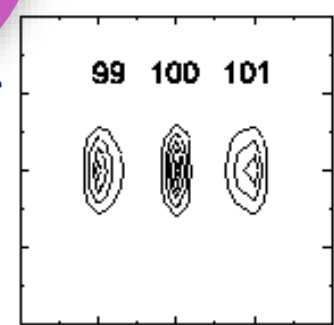
(III) Mass separation



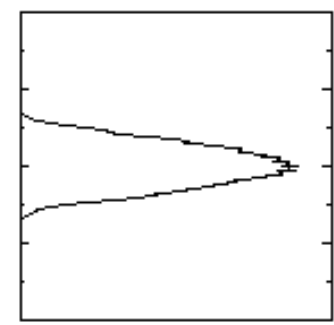
ionization to typ. $q = 1+$
 acceleration to e.g. 60 keV
 mass selection by magnetic deflection
 (Lorentz force)
 ($B\rho = p/q \propto \sqrt{A}$)



Position y



Position x



(II) Ionization

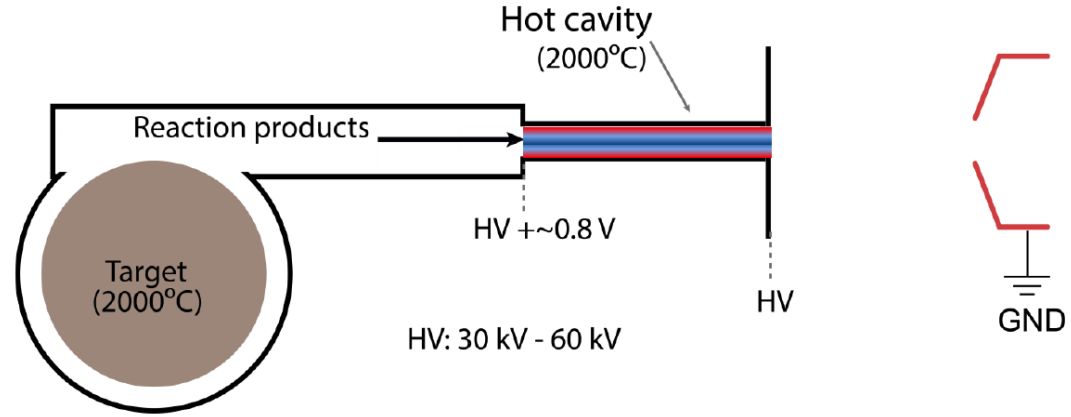
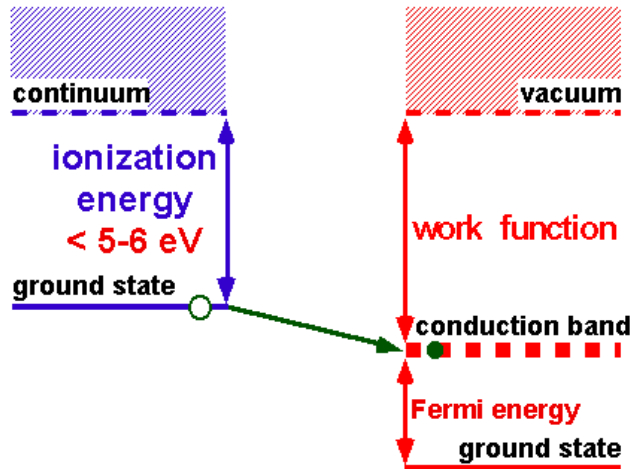
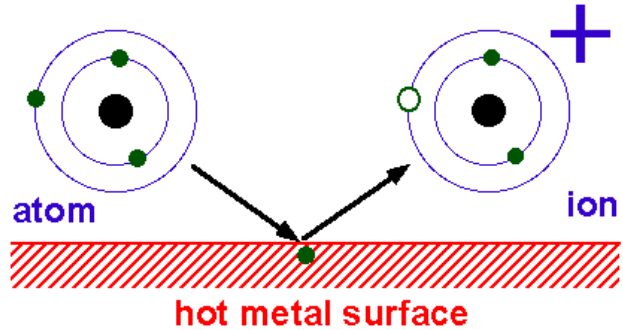
Surface Ionization

Laser Ionization

**Ionization by
electron impact**

(II) Ionization

Surface Ionization



$$\alpha = N \cdot \varepsilon \cdot e^{\frac{\phi - IP}{k_B T}}$$

$$\varepsilon_{ion} \approx \frac{\alpha}{1 + \alpha}$$

α : ionization degree
 N : atom-wall collision
 ε : ion survival probability

ϕ : work function
 IP : ionization potential
 $k_B T$: Boltzmann constant and temperature

Increasing the work function increases the ionization efficiency if $N \cdot \varepsilon$ are preserved
 → Sufficient thermal electron emission must be maintained

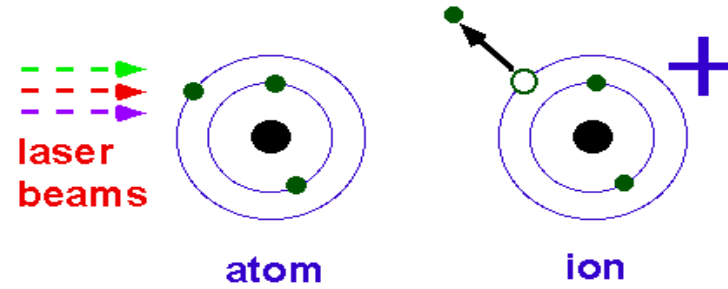
Ionization potentials of the elements

1 H	Ionization potential: < 5 eV																2 He
3 Li	4 Be	Ionization potential: 5.0 - 5.8 eV										5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg	Ionization potential: 5.8 - 6.5 eV										13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
55 Cs	56 Ba	57 La	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
87 Fr	88 Ra	89 Ac	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110	111	112						

58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr

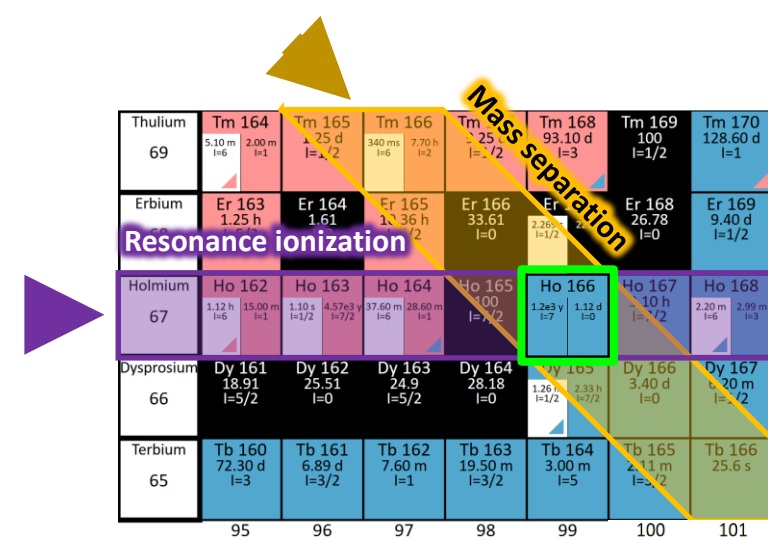
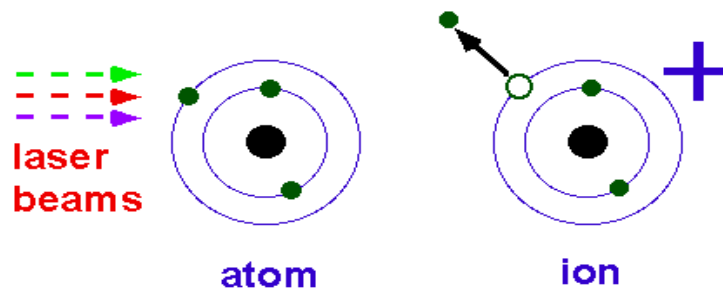
(II) Ionization

Laser Ionization

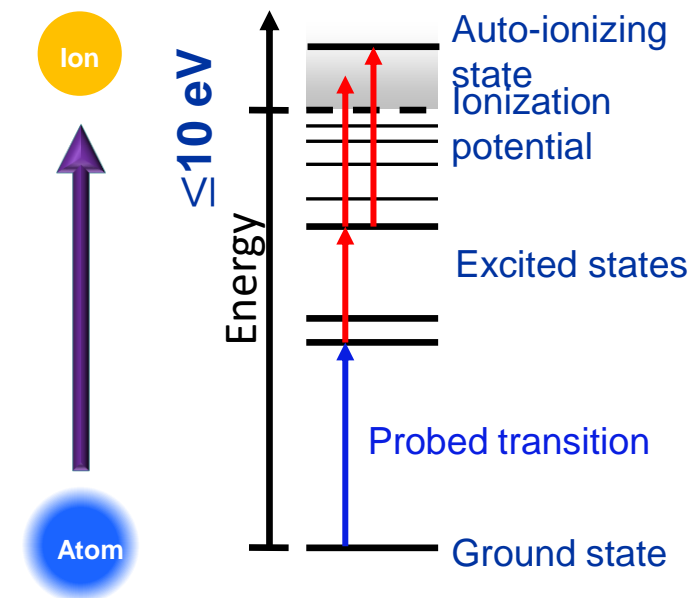
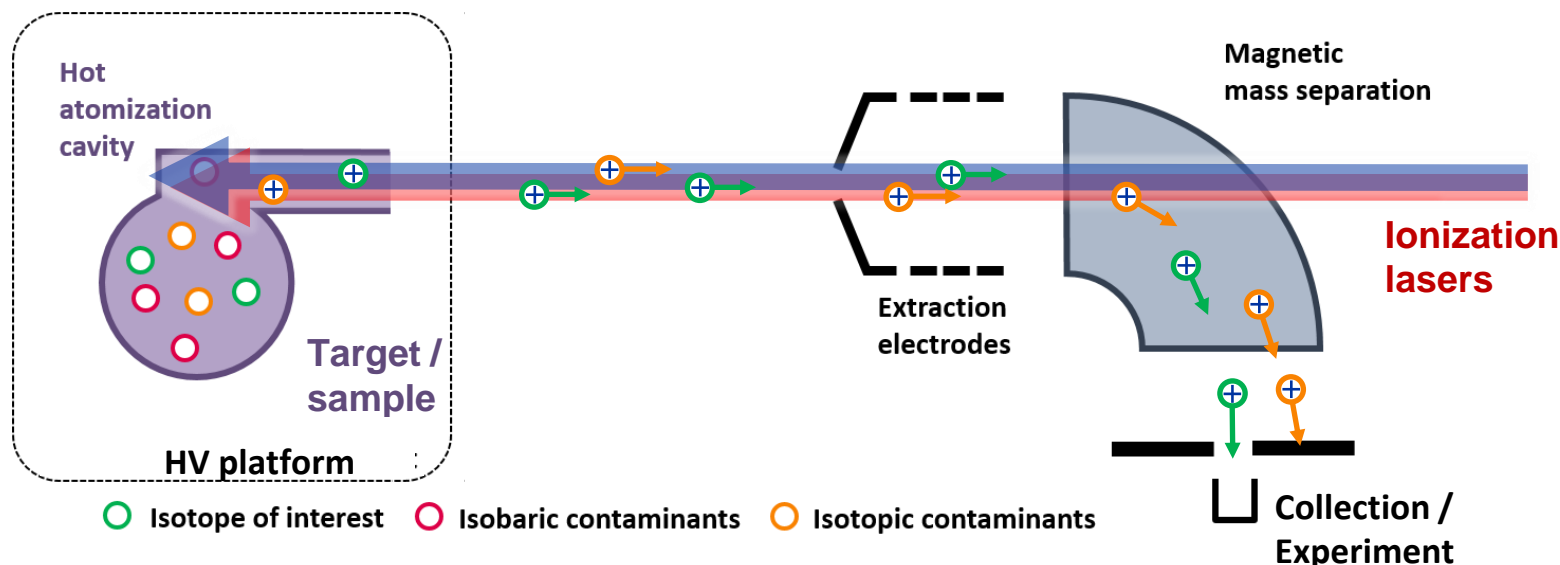


(II) Ionization

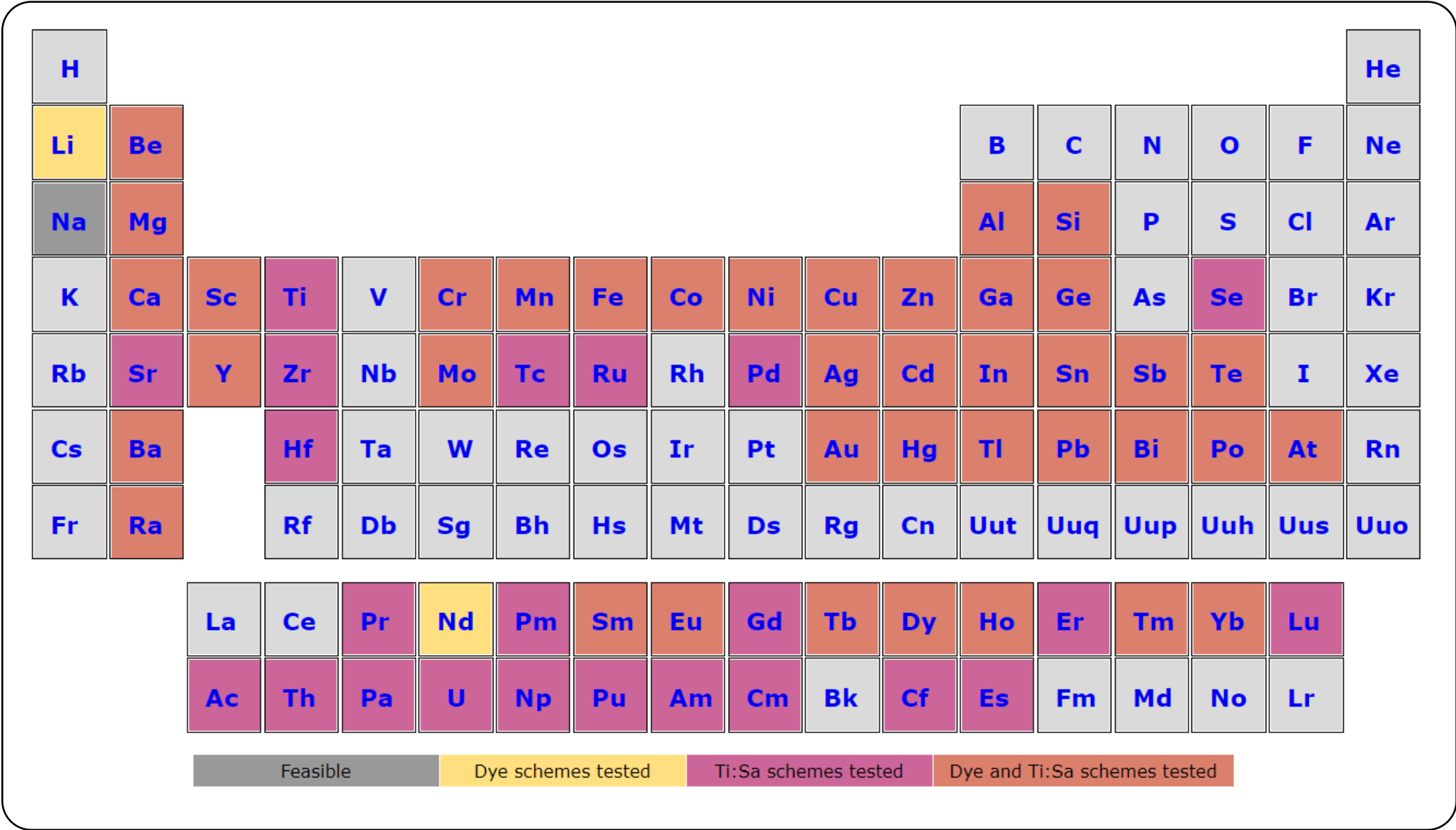
Laser Ionization



CERN-RILIS: Resonance Ionization Laser Ion source

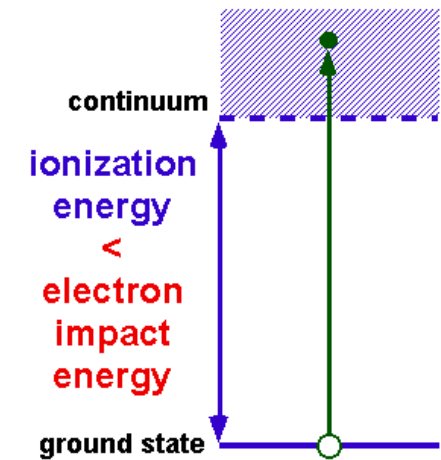
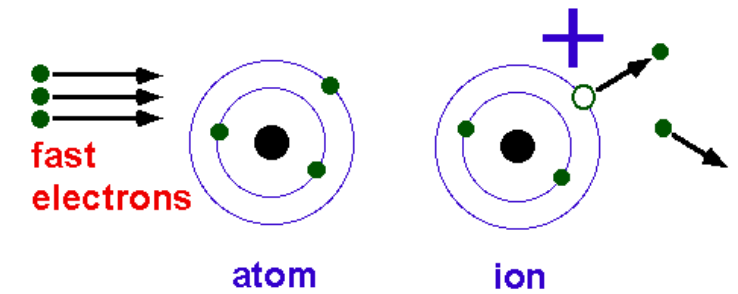


RILIS elements



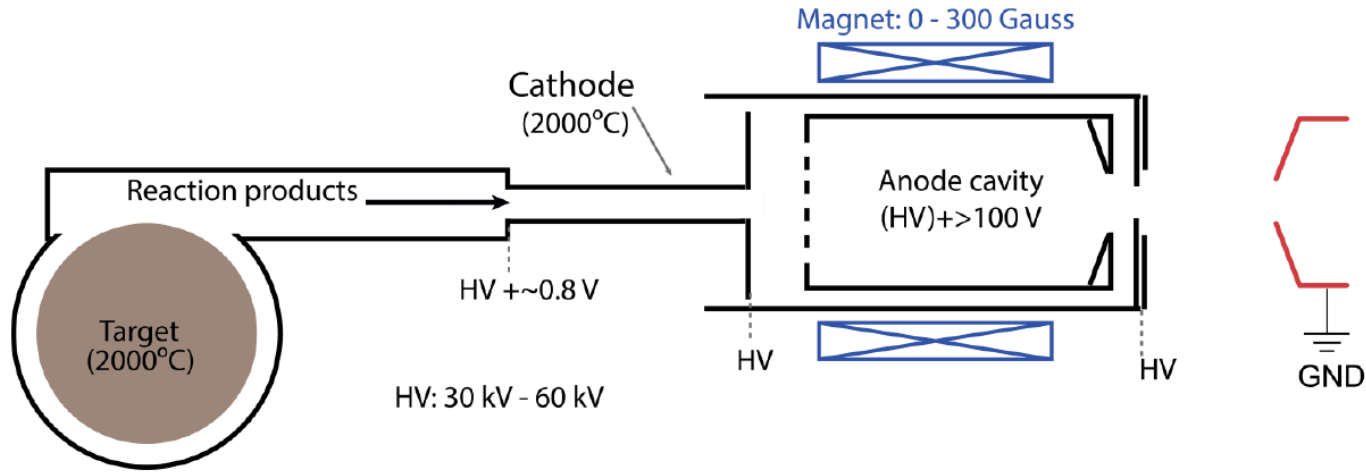
(II) Ionization

Ionization by electron impact

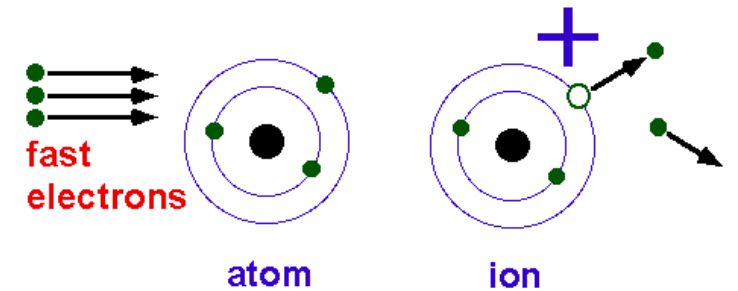


(II) Ionization:

Example: Forced Electron Beam Arc Discharge ion source

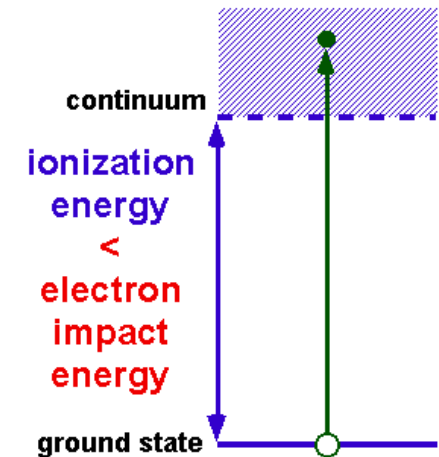


Ionization by electron impact



Properties

- + **universal ionization** (our “dirty” ion source)
- + **good efficiency for noble gasses and volatile species**
- + moderate emittance ($20 \pi \text{ mm mrad}$ at 15 kV, 95%)
- **Limited lifetime**
- + stable operation with little support gas (Pressures $5\text{E-}4$ to $3\text{E-}5$ mbar)
- + low ion current density ($1\text{-}20 \mu\text{A/mm}^2$)
- + low energy spread (2 eV)
- + volume as small as 1.3 cm^3 (6 ms intrinsic delay)



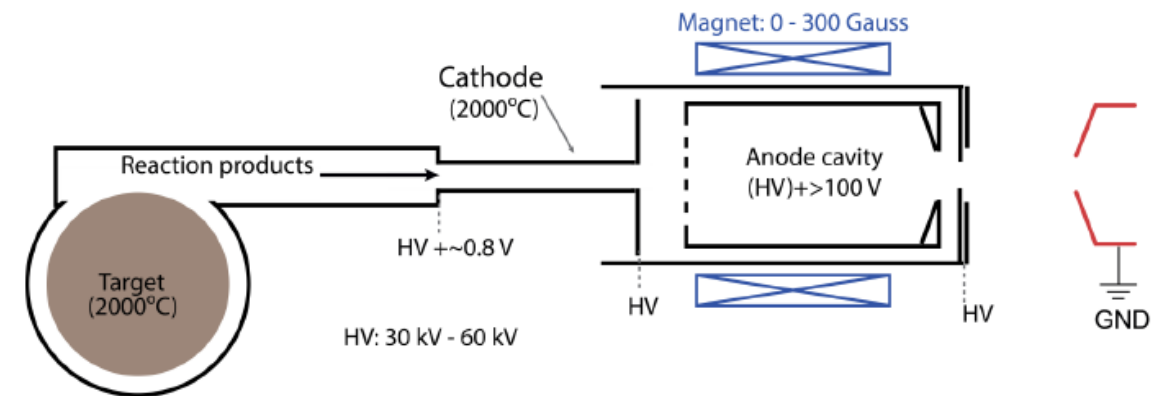
FEBIAD : Forced Electron Beam Arc Discharge ion source

1. Efficiency: ~universal typically 1-50 %
2. Extraction/ionization time: ~10-100 ms (neglecting sticking times)
3. Chemical selectivity: Introduced via transfer line development

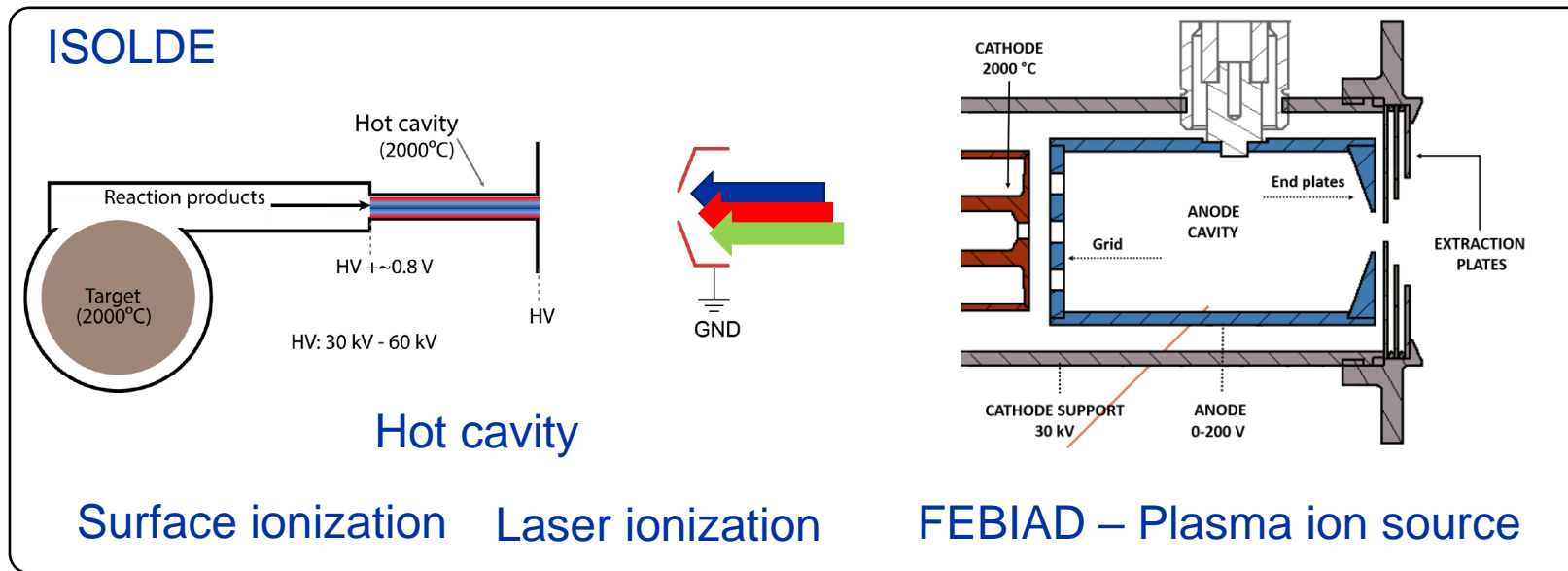
Selective operation for:

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
1	1 H																	2 He
2	3 Li	4 Be										5 B	6 C	7 N	8 O	9 F	10 Ne	
3	11 Na	12 Mg										13 Al	14 Si	15 P	16 S	17 Cl	18 Ar	
4	19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
5	37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
6	55 Cs	56 Ba		72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
7	87 Fr	88 Ra		104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Cn	113 Uut	114 Uuq	115 Uup	116 Uuh	117 Uus	118 Uuo
Lanthanides	57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu			
Actinides	89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr			

Dedicated arc discharge ion source for ISOL: operation over a range of pressures down to 10^{-5} mbar



(II) Ionization: overview



Hot cavity

Surface ionization

Laser ionization

FEBIAD – Plasma ion source

Efficiency



Universality



Selectivity



Simplicity

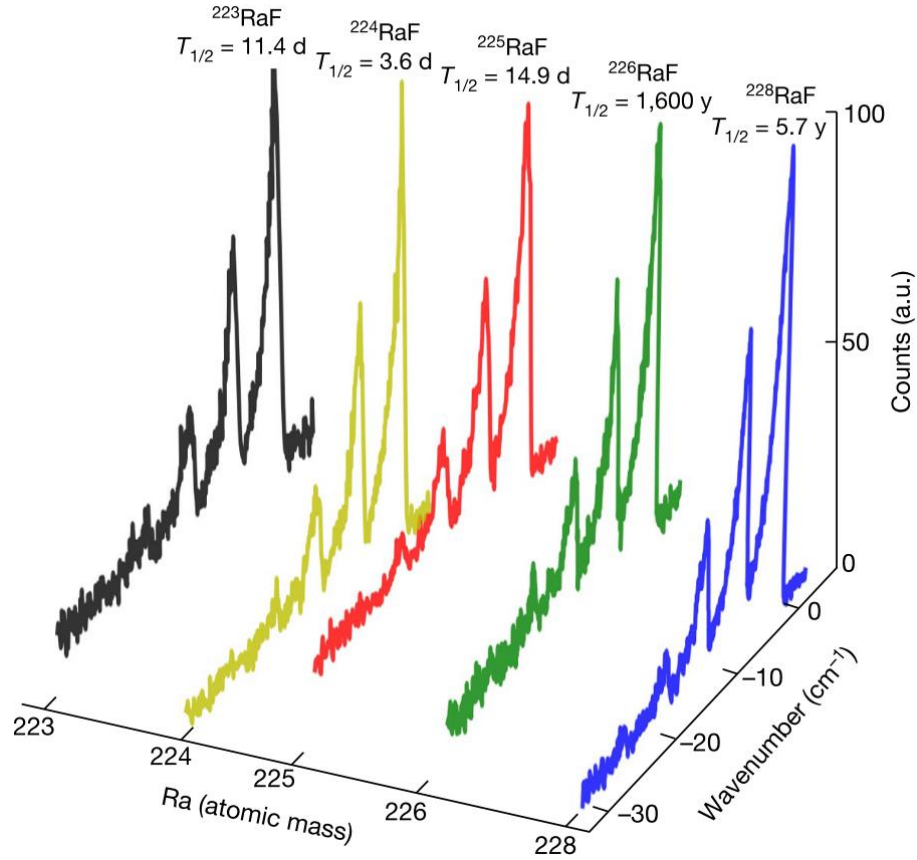
Reliability

Content

1. Why ISOL beams?, ISOL history
2. **ISOL challenges**
3. ISOL stages

4. Use cases around the world

Laser spectroscopy of radioactive molecules



- 1st laser spectroscopy of short-lived radioactive molecules
- 223-226, 228RaF
- Avenue for search of physics beyond the standard model



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Spectroscopy of short-lived radioactive molecules

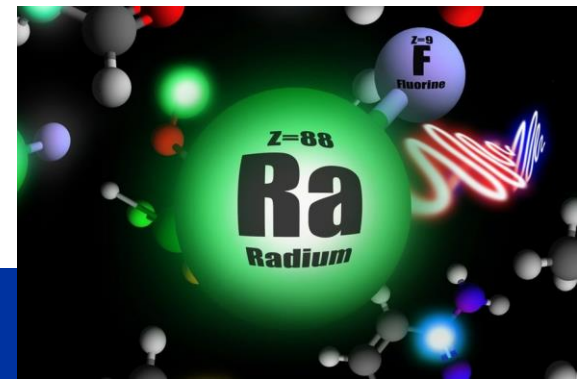
[R. F. Garcia Ruiz](#) , [R. Berger](#) , [J. Billowes](#), [C. L. Binnersley](#), [M. L. Bissell](#), [A. A. Breier](#), [A. J. Brinson](#), [K. Chrysalidis](#), [T. E. Cocolios](#), [B. S. Cooper](#), [K. T. Flanagan](#), [T. F. Giesen](#), [R. P. de Groot](#), [S. Franchoo](#), [F. P. Gustafsson](#), [T. A. Isaev](#), [Á. Koszorús](#), [G. Neyens](#), [H. A. Perrett](#), [C. M. Ricketts](#), [S. Rothe](#), [L. Schweikhard](#), [A. R. Vernon](#), [K. D. A. Wendt](#), [F. Wienholtz](#), [S. G. Wilkins](#) & [X. F. Yang](#) [Show fewer authors](#)

Nature **581**, 396–400 (2020) | [Cite this article](#)

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Abstract

Molecular spectroscopy offers opportunities for the exploration of the fundamental laws of nature and the search for new particle physics beyond the standard model^{1,2,3,4}. Radioactive molecules—in which one or more of the atoms possesses a radioactive nucleus—can contain heavy and deformed nuclei, offering high sensitivity for investigating parity- and time-reversal-violation effects^{5,6}. Radium monofluoride, RaF, is of particular interest because it is predicted to have an electronic structure appropriate for laser cooling⁶, thus paving the way

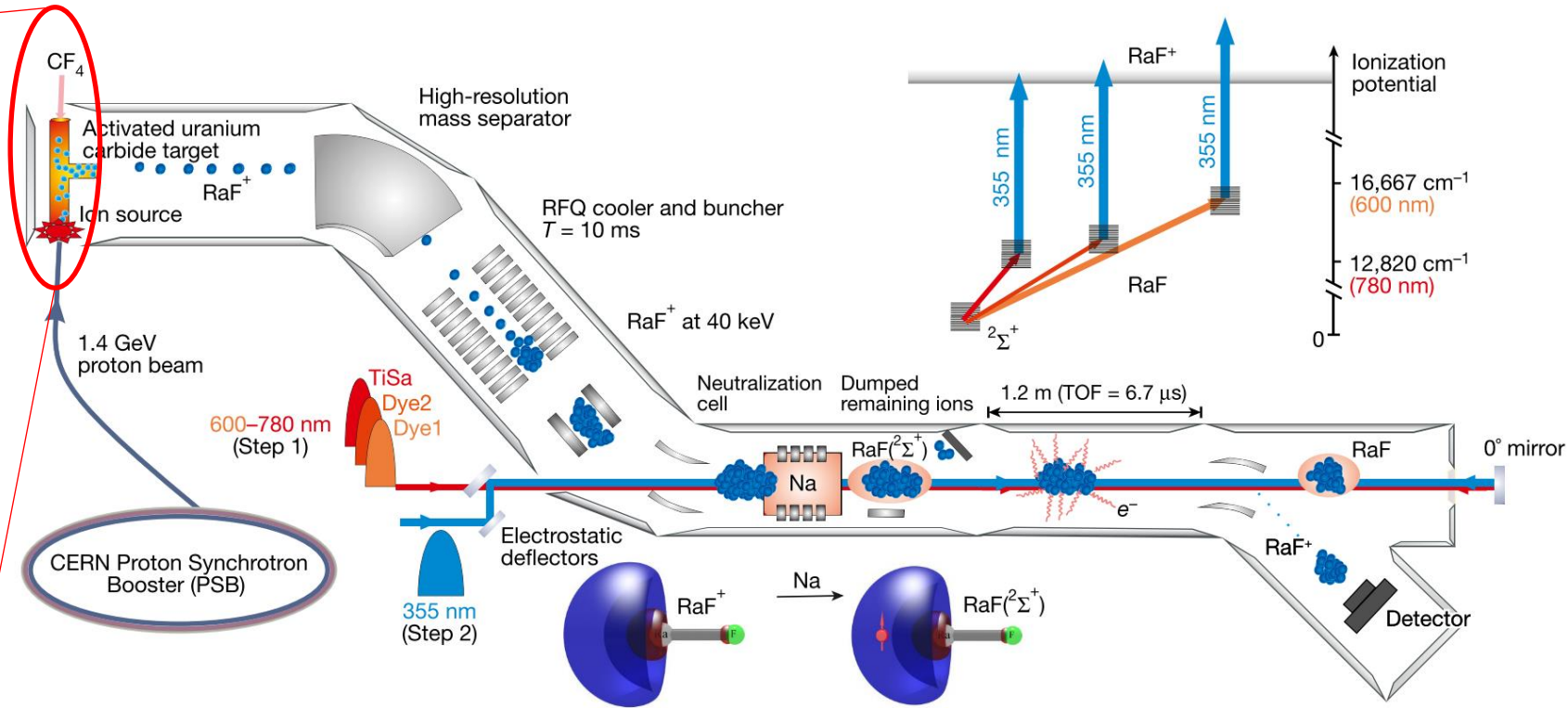
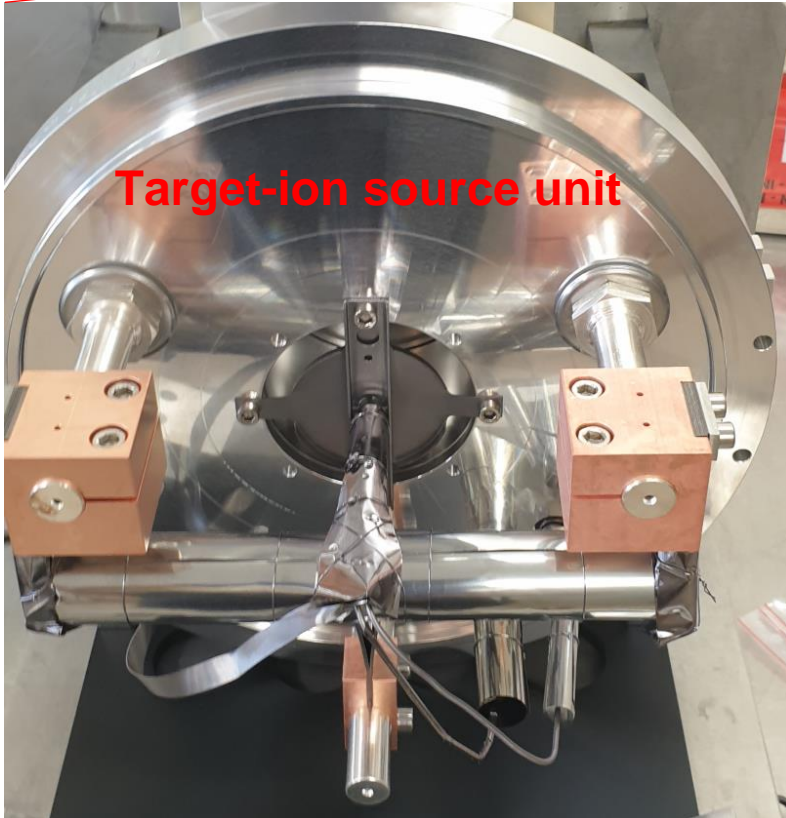


SY
Accelerator Systems



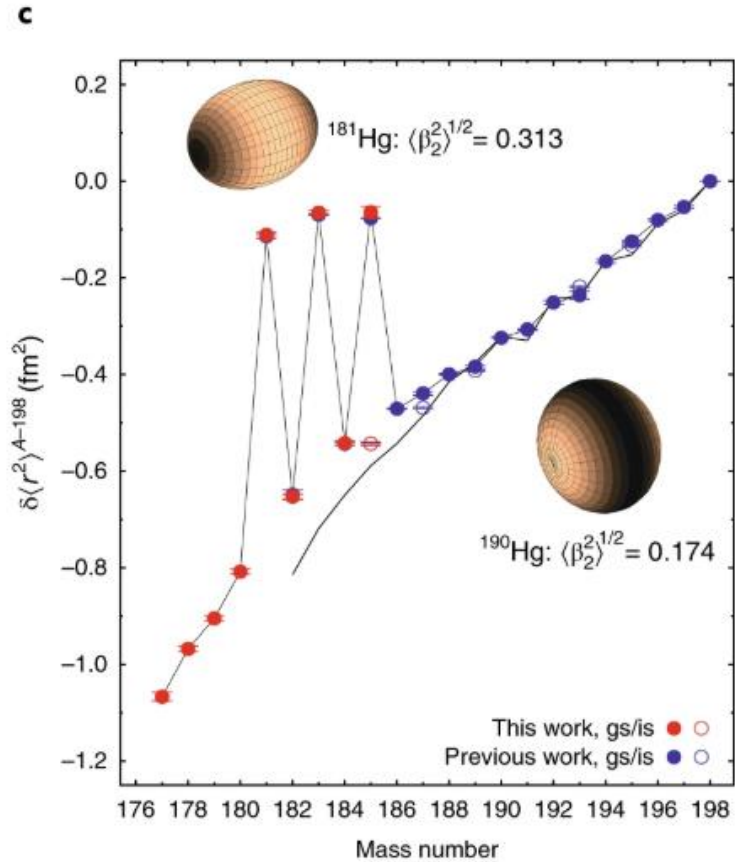
R.F. Garcia Ruiz et al. *Nature* 581 (2020). 10.1038/s41586-020-2299-4

Laser spectroscopy of radioactive molecules



- UC_x target with W-surface ion source with CF₄ injection
- Collinear Resonance Ionization Spectroscopy (CRIS) setup

Shape staggering in Hg-isotopes



- Odd-even shape staggering for $^{181-185}\text{Hg}$ isotopes
- Monopole and quadrupole interactions driving quantum phase transitions

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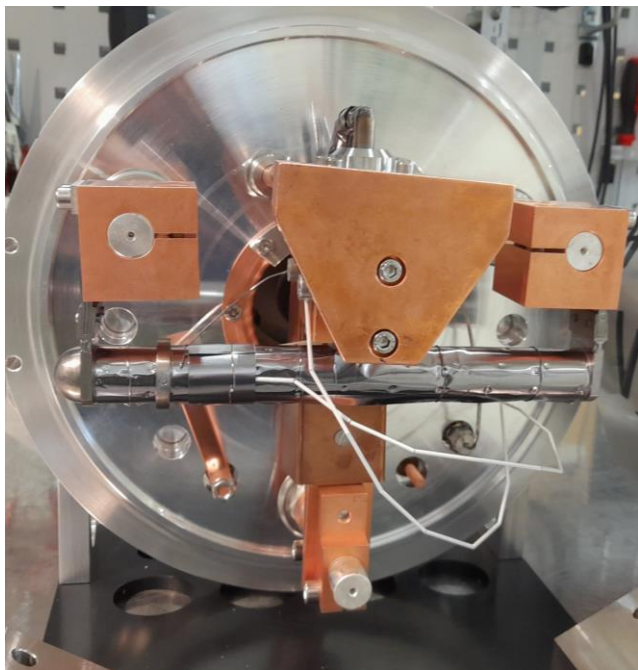
Characterization of the shape-staggering effect in mercury nuclei

[B. A. Marsh](#) ✉, [T. Day Goodacre](#), [S. Sels](#), [Y. Tsunoda](#), [B. Andel](#), [A. N. Andreyev](#), [N. A. Althubiti](#), [D. Atanasov](#), [A. E. Barzakh](#), [J. Billowes](#), [K. Blaum](#), [T. E. Cocolios](#), [J. G. Cubiss](#), [J. Dobaczewski](#), [G. J. Farooq-Smith](#), [D. V. Fedorov](#), [V. N. Fedosseev](#), [K. T. Flanagan](#), [L. P. Gaffney](#), [L. Ghys](#), [M. Huyse](#), [S. Kreim](#), [D. Lunney](#), [K. M. Lynch](#), [V. Manea](#), [Y. Martinez Palenzuela](#), [P. L. Molkanov](#), [T. Otsuka](#), [A. Pastore](#), [M. Rosenbusch](#), [R. E. Rossel](#), [S. Rothe](#), [L. Schweikhard](#), [M. D. Seliverstov](#), [P. Spagnoletti](#), [C. Van Beveren](#), [P. Van Duppen](#), [M. Veinhard](#), [E. Verstraelen](#), [A. Welker](#), [K. Wendt](#), [F. Wienholtz](#), [R. N. Wolf](#), [A. Zadvornaya](#) & [K. Zuber](#)

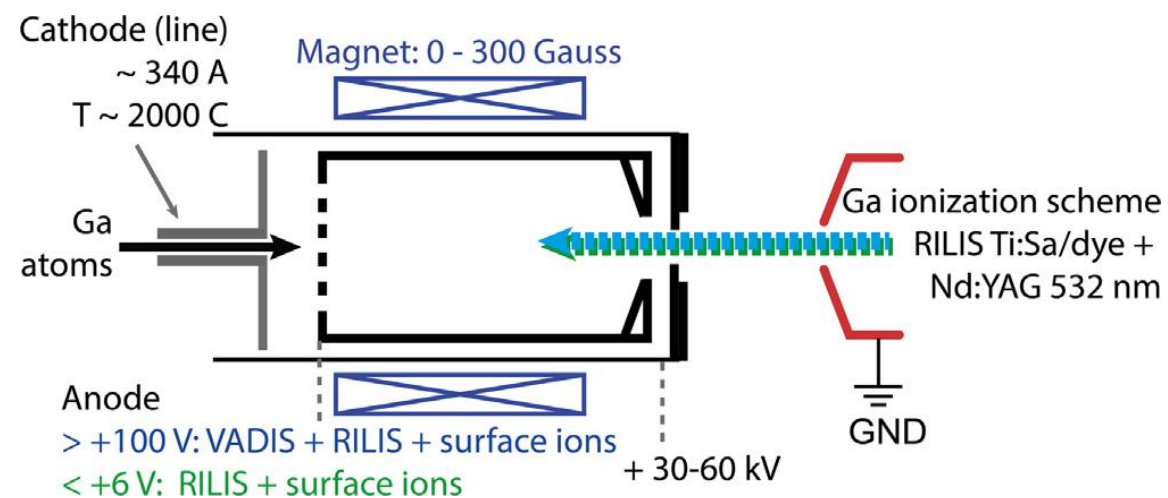
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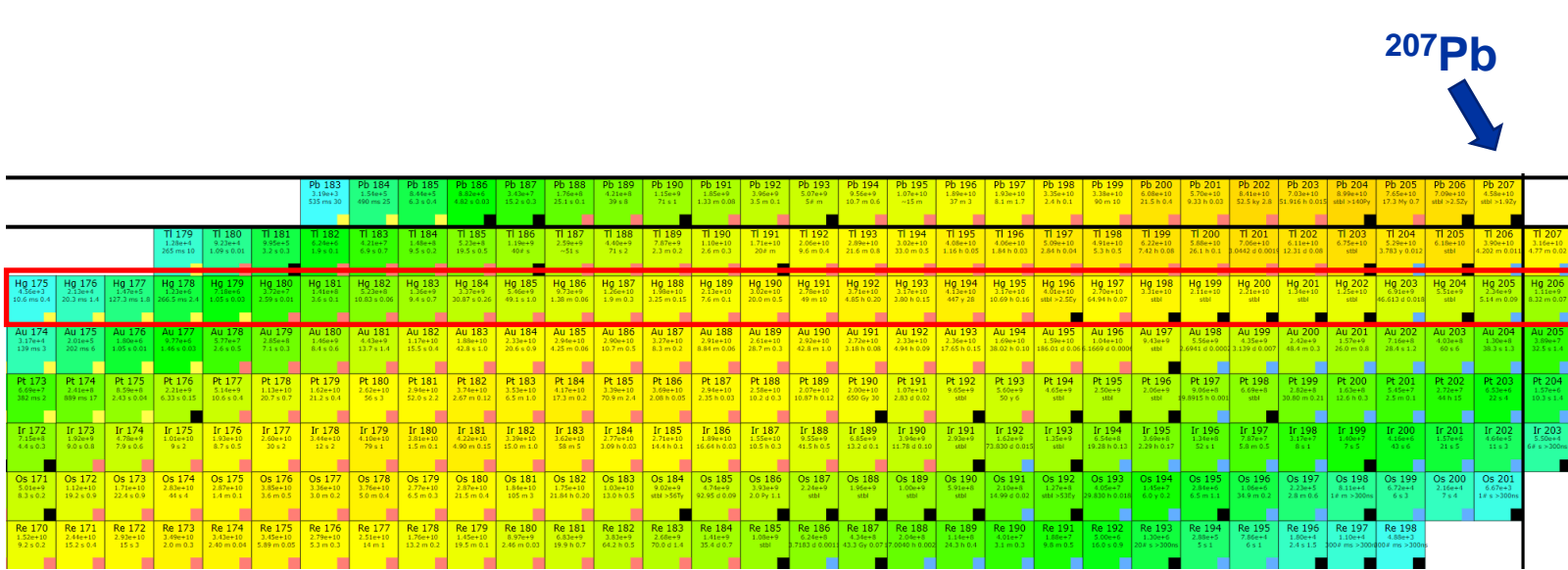
Shape staggering in Hg-isotopes



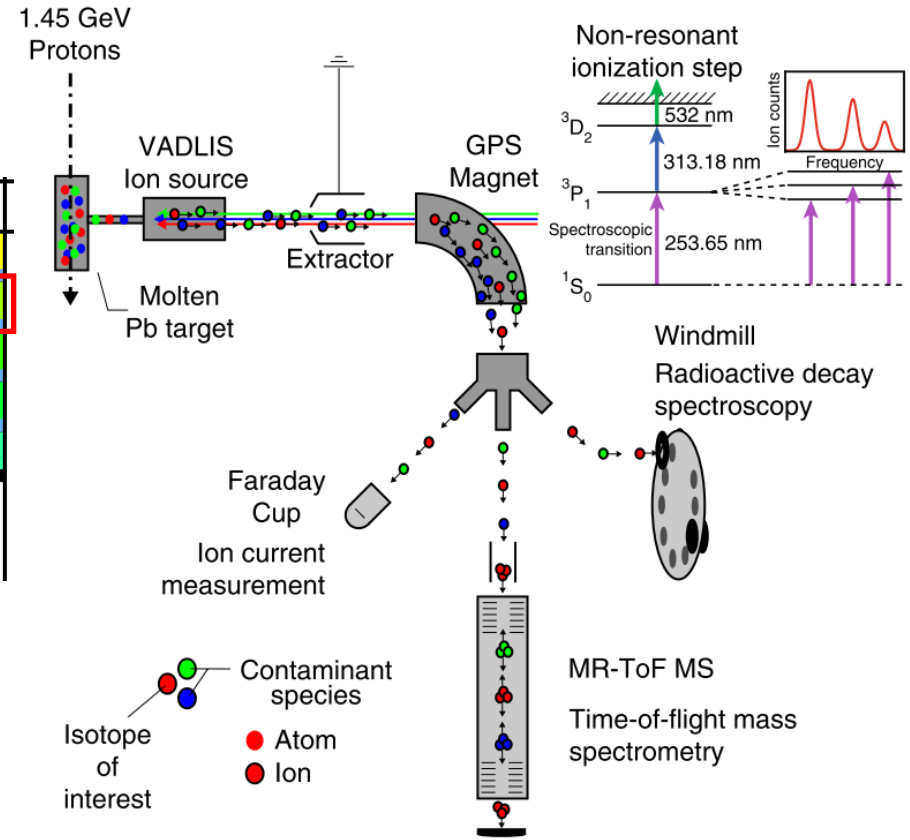
- Liquid Pb target
- Temperature-controlled chimney
 - Enable Hg-effusion
 - Suppress less volatile species (e.g. Pb)
- Special plasma-laser ion source (“VADLIS”)



Shape staggering in Hg-isotopes



- x Hg-isotopes from high-energy proton (1.4 GeV) induced spallation
- In-source laser spectroscopy to bridge to more n-deficient isotopes



Shape coexistence in $^{110,112}\text{Cd}$

PHYSICAL REVIEW LETTERS **123**, 142502 (2019)

Editors' Suggestion Featured in Physics

Multiple Shape Coexistence in $^{110,112}\text{Cd}$

P. E. Garrett,^{1,2} T. R. Rodríguez,³ A. Diaz Varela,¹ K. L. Green,¹ J. Bangay,¹ A. Finlay,¹ R. A. E. Austin,⁴ G. C. Ball,⁵ D. S. Bandyopadhyay,¹ V. Bildstein,¹ S. Colosimo,⁴ D. S. Cross,⁶ G. A. Demand,¹ P. Finlay,¹ A. B. Garnsworthy,⁵ G. F. Grinyer,⁷ G. Hackman,⁵ B. Jigmeddorj,¹ J. Jolie,⁸ W. D. Kulp,⁹ K. G. Leach,^{1,*} A. C. Morton,^{5,†} J. N. Orce,² C. J. Pearson,⁵ A. A. Phillips,¹ A. J. Radich,¹ E. T. Rand,^{1,‡} M. A. Schumaker,¹ C. E. Svensson,¹ C. Sumithrarachchi,^{1,†} S. Triambak,² N. Warr,⁸ J. Wong,¹ J. L. Wood,¹⁰ and S. W. Yates¹¹

¹Department of Physics, University of Guelph, Guelph, Ontario N1G2W1, Canada

²Department of Physics and Astronomy, University of the Western Cape, P/B X17, Bellville ZA-7535, South Africa

³Departamento de Física Teórica and CIAFF, Universidad Autónoma de Madrid, E-28049 Madrid, Spain

⁴Department of Physics and Astronomy, St. Mary's University, Halifax, Nova Scotia B3H3C3, Canada

⁵TRIUMF, 4004 Wesbrook Mall, Vancouver, British Columbia V6T2A3, Canada

⁶Department of Chemistry, Simon Fraser University, Burnaby, British Columbia V5A1S6, Canada

⁷Department of Physics, University of Regina, Regina, Saskatchewan S4S0A2, Canada

⁸Institut für Kernphysik, Universität zu Köln, Zùlpicherstrasse 77, D-50937 Köln, Germany

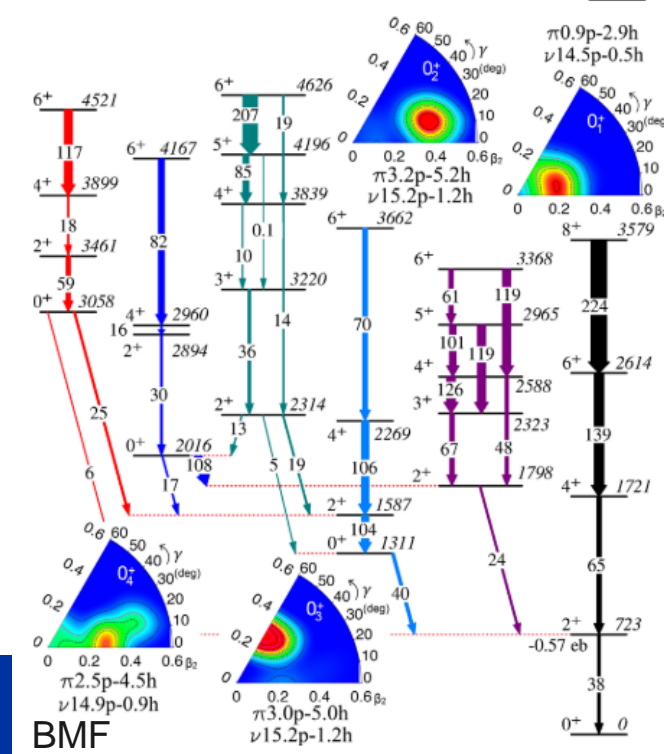
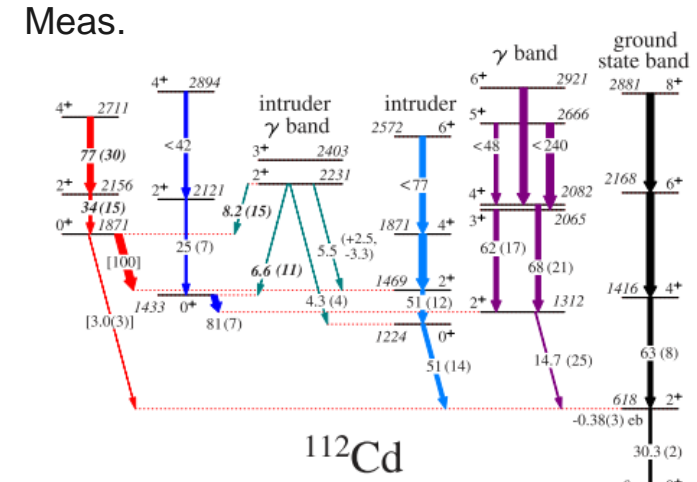
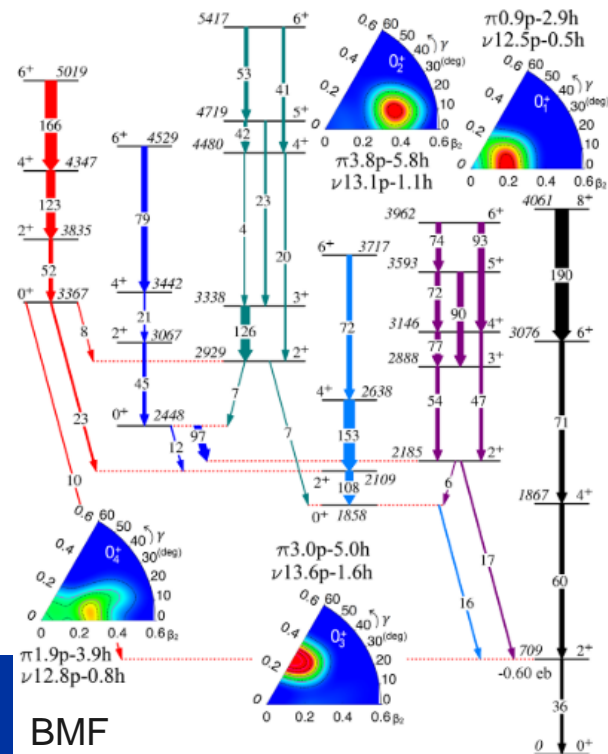
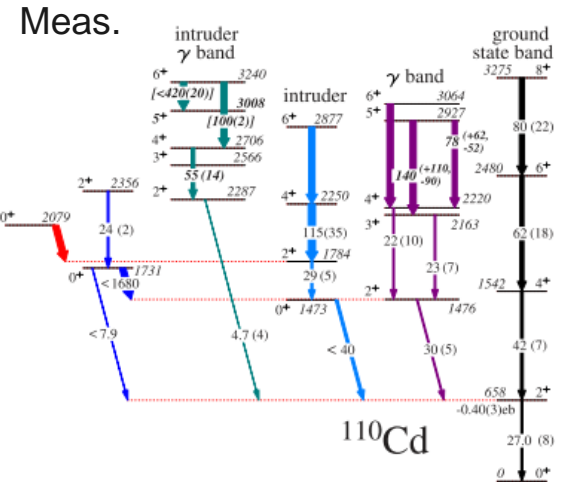
⁹Defense Threat Reduction Agency, 8725 John J Kingman Road, Fort Belvoir, Virginia 22060-6217, USA

¹⁰Department of Physics, Georgia Institute of Technology, Atlanta, Georgia 30332, USA

¹¹Departments of Chemistry and Physics and Astronomy, University of Kentucky, Lexington, Kentucky 40506-0055, USA

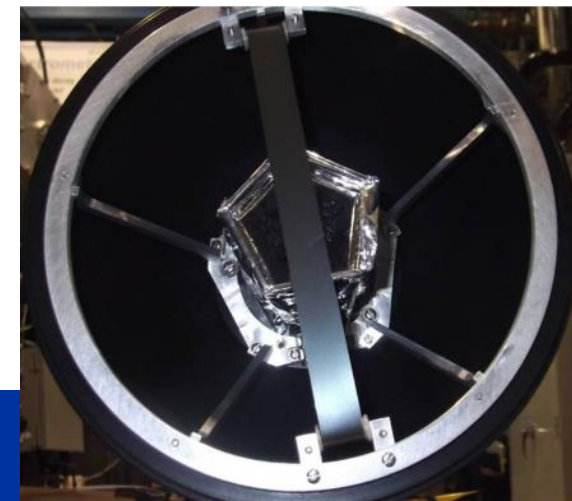
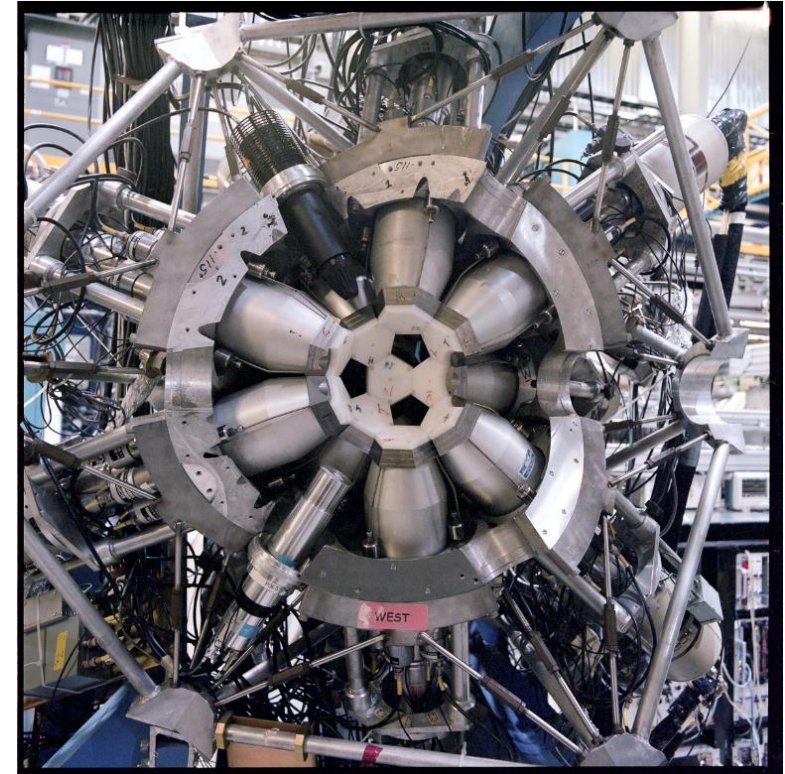
(Received 31 August 2018; revised manuscript received 29 June 2019; published 3 October 2019)

- $^{110,112}\text{Cd}$ decay spectroscopy
- Suggests: multiple shape coexistence (SC) in low-lying states
- Endorses: SC not as rare as thought
- Suggests: SC may begin immediately, even at closed-shell



Shape coexistence in $^{110,112}\text{Cd}$

- TRIUMF-ISAC 0.5 GeV, 40-65 μA protons
- Tantal target, Re-surface and TRILIS
- Primary RIBs: $^{110\text{g,m},112\text{g,m}}\text{In}$, ^{112}Ag ; rates $\sim\text{E6-E7 s}^{-1}$
- Implanted on Mylar tape
- Decay spectroscopy using 8π spectrometer
 - 20 HPGe-detector with BGO Compton-suppression

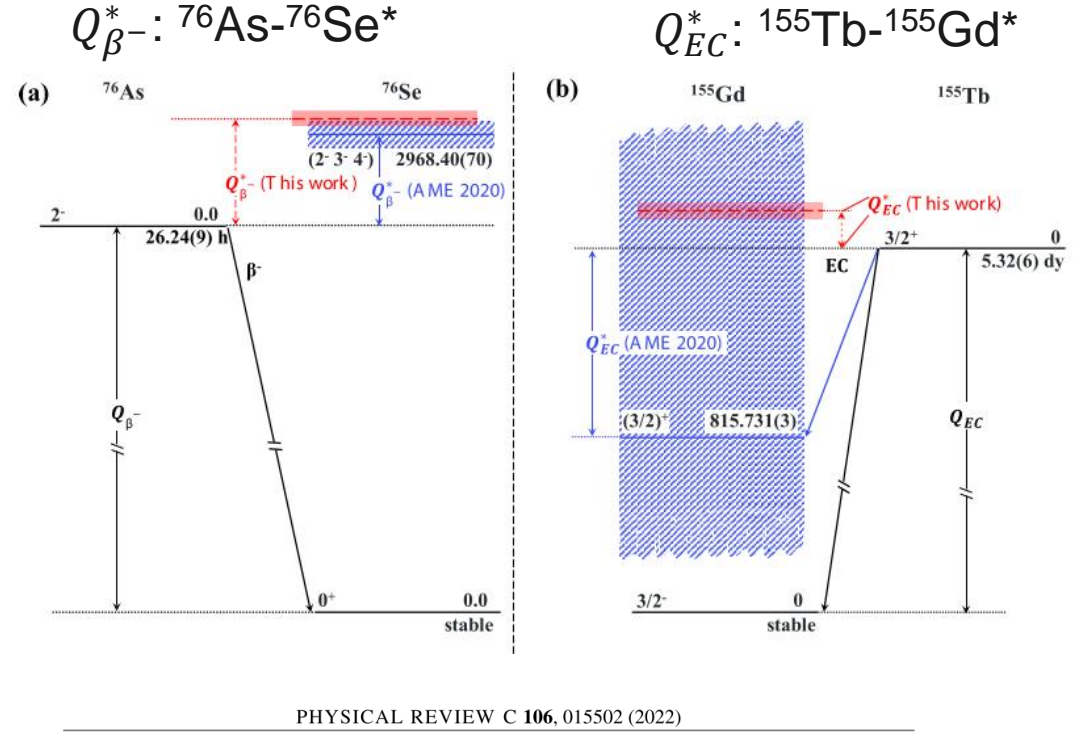
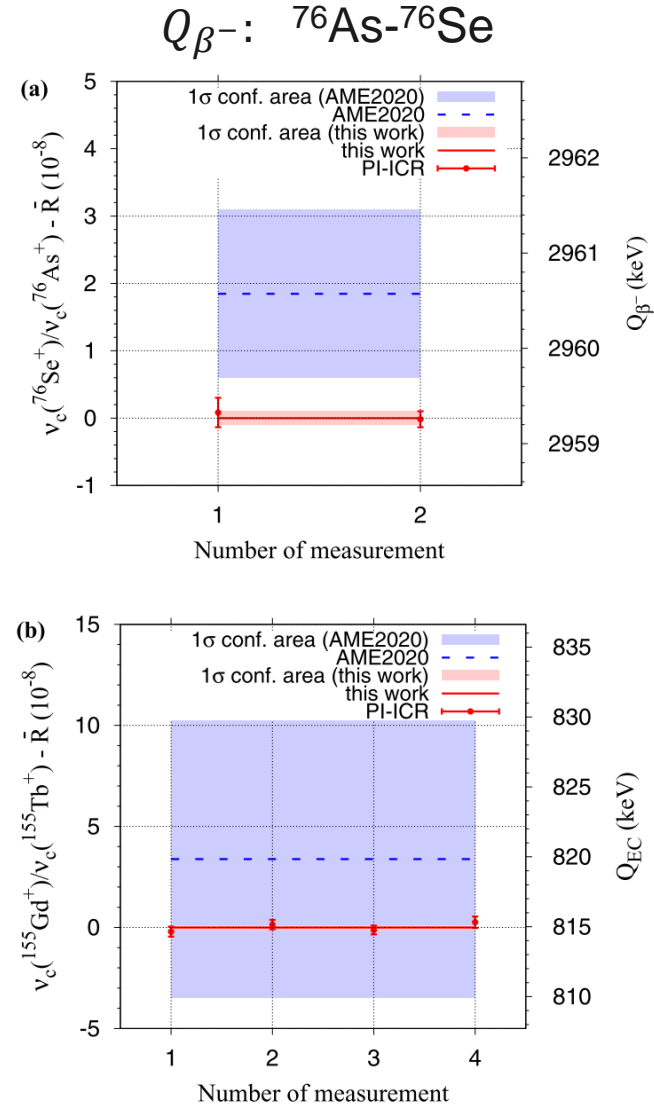


Atomic mass difference of ^{76}As - ^{76}Se and ^{155}Tb - ^{155}Gd



JYVÄSKYLÄN YLIOPISTO
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- Direct high-precision determination of Q_{β^-} , Q_{EC} using JYFLTRAP Penning-trap mass spectrometer
- Determined $Q_{\beta^-}^*$, Q_{EC}^* using tabulated energy-level data from γ -spectroscopy
- Energetically forbidden channels (negative)
- Unsuitable candidates for (anti)neutrino mass determination



Direct determination of the atomic mass difference of the pairs ^{76}As - ^{76}Se and ^{155}Tb - ^{155}Gd rules out ^{76}As and ^{155}Tb as possible candidates for electron (anti)neutrino mass measurements

Z. Ge^{1,*}, T. Eronen^{1,†}, A. de Roubin², J. Kostensalo³, J. Suhonen⁴, D. A. Nesterenko¹, O. Beliuskina¹, R. de Groot¹, C. Delafosse¹, S. Geldhof^{1,‡}, W. Gins¹, M. Hukkanen^{1,2}, A. Jokinen¹, A. Kankainen¹, J. Kotila^{1,4,5}, Á. Koszorús^{6,§}, I. D. Moore¹, A. Raggio¹, S. Rinta-Anttila¹, V. Virtanen¹, A. P. Weaver⁷ and A. Zadornaya^{1,†}

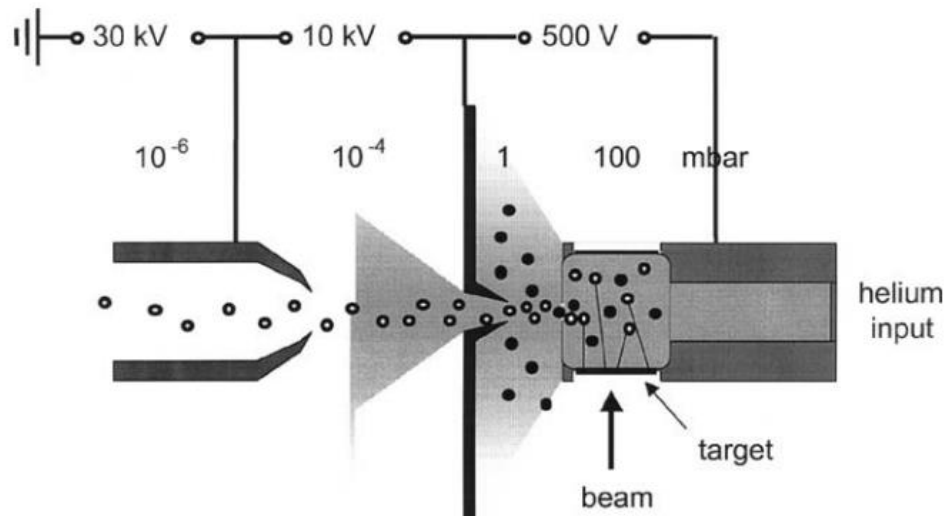
¹Department of Physics, University of Jyväskylä, P.O. Box 35, FI-40014 Jyväskylä, Finland
²Centre d'Etudes Nucléaires de Bordeaux Gradignan, UMR 5797 CNRS/IN2P3 - Université de Bordeaux, 19 Chemin du Solarium, CS 10120, F-33175 Gradignan Cedex, France
³Natural Resources Institute Finland, Yliopistokatu 6B, FI-80100 Joensuu, Finland
⁴Finnish Institute for Educational Research, University of Jyväskylä, P.O. Box 35, FI-40014 Jyväskylä, Finland
⁵Center for Theoretical Physics, Sloane Physics Laboratory Yale University, New Haven, Connecticut 06520-8120, USA
⁶Department of Physics, University of Liverpool, Liverpool L69 7ZE, United Kingdom
⁷School of Computing, Engineering and Mathematics, University of Brighton, Brighton BN2 4JG, United Kingdom

(Received 16 February 2022; accepted 21 June 2022; published 13 July 2022)

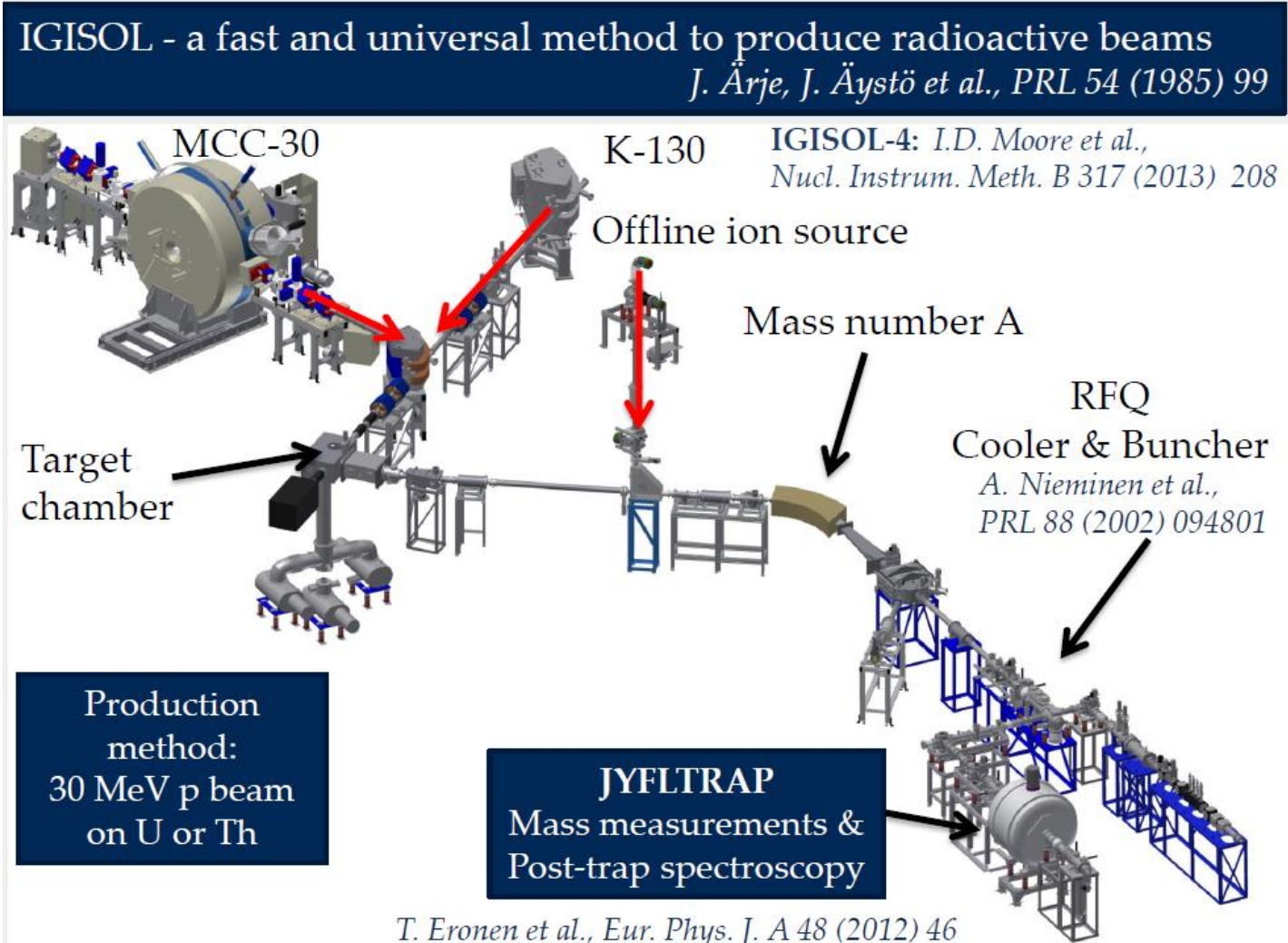
Atomic mass difference of ^{76}As - ^{76}Se and ^{155}Tb - ^{155}Gd



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UNIVERSITY OF JYVÄSKYLÄ



- IGISOL
- 2 mg/cm² Ge target - 9 MeV d ($^{76}\text{As}^+$, $^{76}\text{Se}^+$)
- 2 mg/cm² Gd target - 60 MeV p ($^{155}\text{Tb}^+$, $^{155}\text{Gd}^+$)
- Mass separation, cooled and bunched (RFQ)
- JYFLTRAP double Penning trap
- Separation of isobaric contaminants (1st trap)
- Measured cyclotron frequency via phase-imaging ion-cyclotron-resonance (PI-ICR)



Nuclear structure of $^{253-255}\text{Es}$ via laser spectroscopy

PHYSICAL REVIEW C **105**, L021302 (2022)

Letter

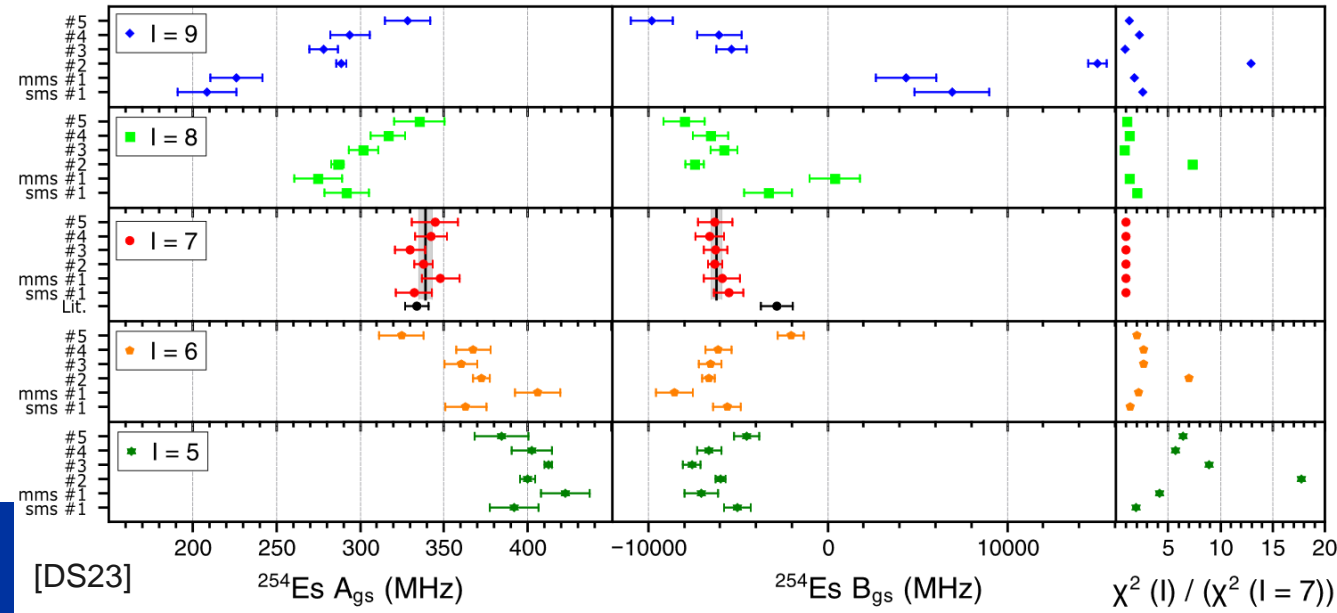
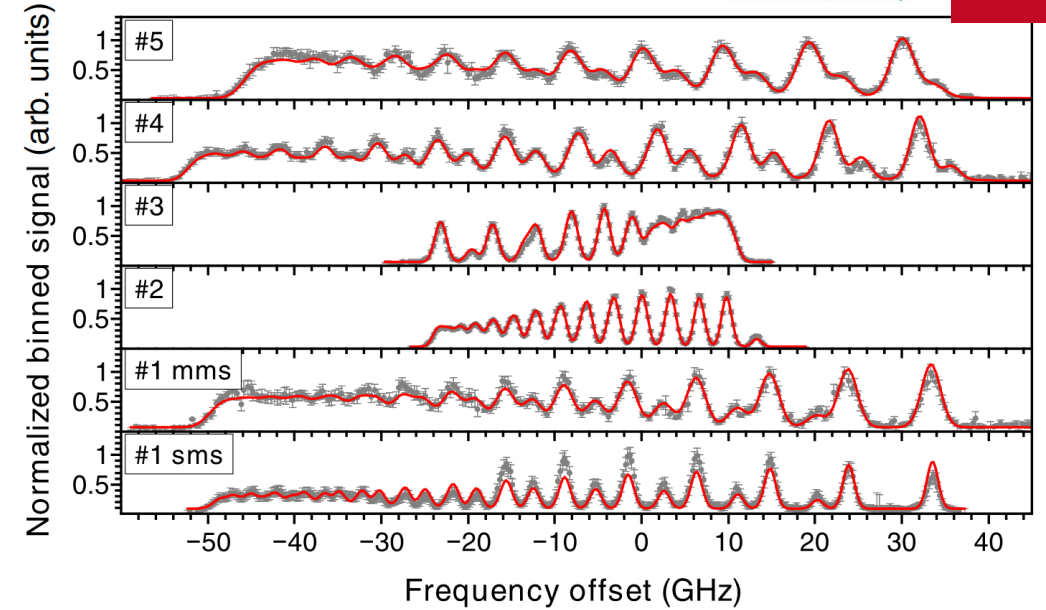
Nuclear structure investigations of $^{253-255}\text{Es}$ by laser spectroscopy

S. Nothhelfer^{1,2,3}, Th. E. Albrecht-Schönzart⁴, M. Block^{1,2,3}, P. Chhetri^{2,3},
Ch. E. Düllmann^{1,2,3}, J. G. Ezold⁵, V. Gadelshin^{1,6}, A. Gaiser⁴, F. Giacoppo^{2,3}, R. Heinke^{1,7}, T. Kieck^{1,2,3},
N. Kneip¹, M. Laatiaoui^{1,2,3}, Ch. Mokry^{1,2}, S. Raeder^{2,3}, J. Runke^{1,3}, F. Schneider^{1,2}, J. M. Sperling⁴, D. Studer¹,
P. Thörle-Pospiech^{1,2}, N. Trautmann¹, F. Weber¹, and K. Wendt¹

¹Johannes Gutenberg-Universität Mainz, 55099 Mainz, Germany
²Helmholtz-Institut Mainz, 55099 Mainz, Germany
³GSI Helmholtzzentrum für Schwerionenforschung GmbH, 64291 Darmstadt, Germany
⁴Florida State University, 32306 Tallahassee, Florida, USA
⁵Oak Ridge National Laboratory, Oak Ridge, 37831 Oak Ridge, Tennessee, USA
⁶Ural Federal University, 620002 Yekaterinburg, Russia

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- Hyperfine structures measured for 5 ground-state transitions in ^{254}Es
- Accurate determination of spin and A_{gs} , B_{gs} coupling constants through large dataset
- x100 atomic transitions recorded; scheme dev.

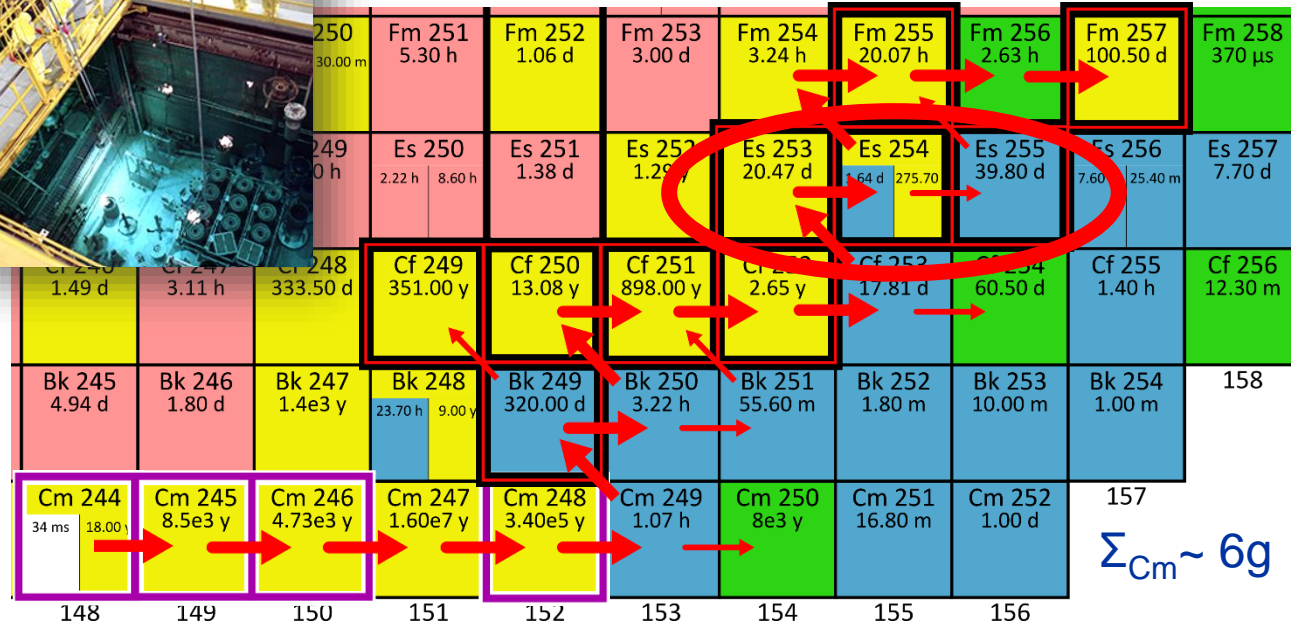


The isotopes used in this research were supplied by the U.S. Department of Energy, Office of Science, by the Isotope Program in the Office of Nuclear Physics. The $^{253,254,255}\text{Es}$ and $^{255,257}\text{Fm}$ were provided to Florida State University and the University of Mainz via the Isotope Development and Production for Research and Applications Program through the Radiochemical Engineering and Development Center at Oak Ridge National Laboratory.

Nuclear structure of $^{253-255}\text{Es}$ via laser spectroscopy



High Flux Isotope Reactor (HFIR) at ORNL



- Irradiation of 6g Cm at ORNL for ~5 months in 2018
- Chemical separation after ~4 months cooling
- Shipping of Es/Fm fraction to U. Mainz in Aug. 2019 (short-lived isotopes have decayed)

Specified sample contents Jan. 2019

^{257}Fm	~0.5 pg	~1e9 atoms
^{255}Es	~4 pg	~ 1e10 atoms
^{254}Es	~4 ng	~ 1e13 atoms
^{253}Es	~2 ng	~ 5e12 atoms
*Cf	n.d.	

Summary

- What are the stages of ISOL-type RIB production?
- What are general and stage-specific ISOL challenges?
- What reactions are employed and where?
- ISOL is powerful for nuclear physics research

Acknowledgements & References

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- **R. Heinke**
- **D. Studer**
- **J.P. Ramos**
- **F. Wenander**

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[RE23] RILIS Elements Website, <https://riliselements.web.cern.ch>

Thank you!



Questions, comments?

home.cern

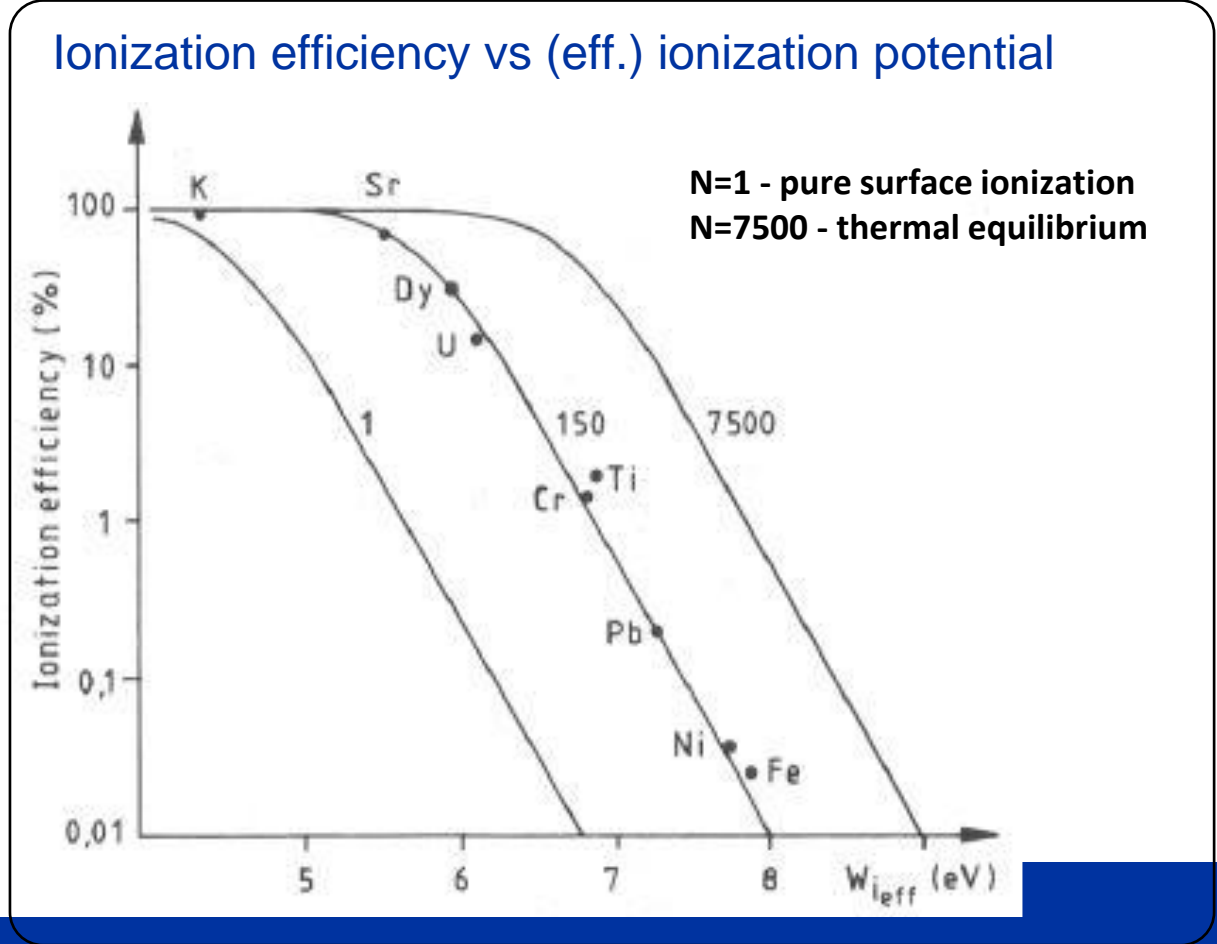
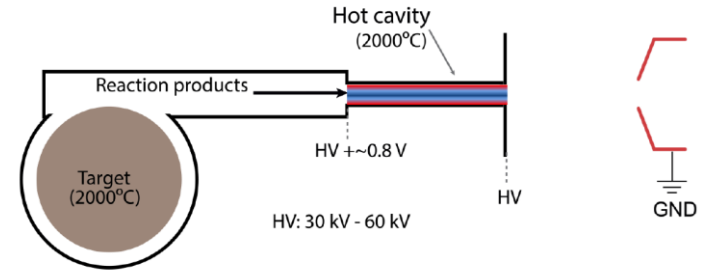
(positive) surface ionization source

Properties

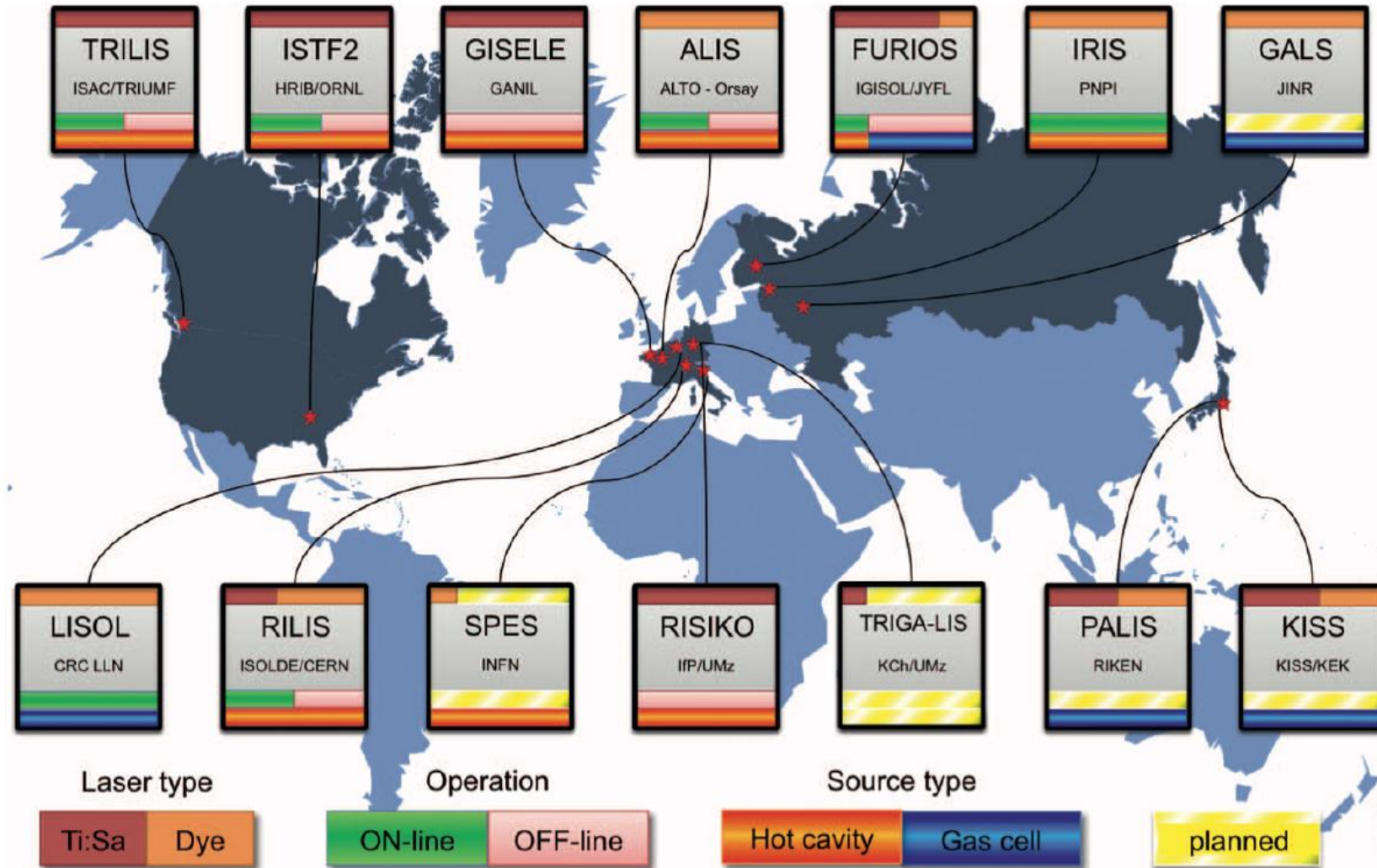
- * Ionisation efficiency 100% for $W_i < 5 \text{ eV}$, few % for $W_i = 6.5 \text{ eV}$
- * Used for alkalis, alkaline earths, rare earths, Ga, In and Tl
also molecules as BaF and SrF
- * Emittance $\sim 10 \pi \text{ mm mrad}$ (60 kV, 95%)
- * Energy spread $< 2 \text{ eV}$
- * max current $1 \mu\text{A/mm}^2$
- * Short delay time (half-lives as short as 10 ms)
small ionisation volume
operates at elevated temperatures
closely coupled to targets

Ionizer material

- * Ta, W, Re, Ir, Pt
- * temperatures up to 2800 K
- * e.g. tungsten with $\phi \sim 4.5 \text{ eV}$ at $2400 \text{ }^\circ\text{C}$
- * Work function depends on crystal orientation, temperature and cleanliness



Laser ion sources worldwide



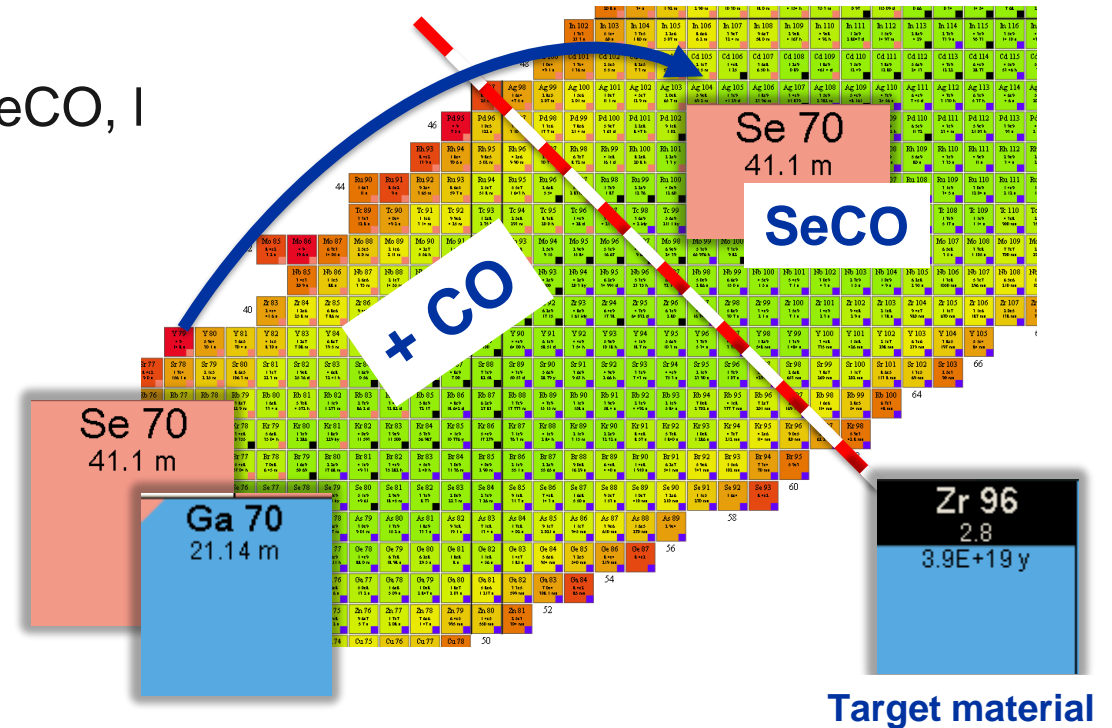
Molecular beams development

Beam purification

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

Beam extraction by *In-situ* volatilization

- Elements with very low volatility are not released
- Reactive elements can be chemically trapped

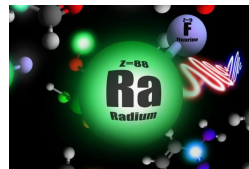


Physics with radioactive molecules

Article
Spectroscopy of short-lived radioactive molecules

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R. F. Garcia Ruiz^{1,2}, R. Berger^{1,3}, J. Billowes⁴, C. L. Binnersley⁴, M. L. Basselt⁴, A. A. Breier⁴, A. J. Brinson⁴, K. Chrysalidis⁴, T. E. Cocolos⁴, B. S. Cooper⁴, K. T. Flanagan⁴, T. F. Glavin⁴, R. P. de Groen⁴, S. Franchou⁴, F. P. Gustafsson⁴, T. A. Ivan⁴, A. Kozorov⁴, G. Nevejan⁴, H. A. Perrott⁴, C. M. Ricketts⁴, S. Roth⁴, L. Schwabhard⁴, A. R. Vernon⁴, K. D. A. Wendt⁴, F. Wienholtz⁴, S. G. Wilkins⁴ & X. F. Yang⁴



Garcia Ruiz, R., Berger, R., Billowes, J. et al. Spectroscopy of short-lived radioactive molecules. Nature 581, 396–400 (2020). <https://doi.org/10.1038/s41586-020-2299-4>



SY
Accelerator Systems



[JB17,RBB20,MIT20,LISA]



LISA – Laser Ionization and Spectroscopy of Actinides

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Filling the gaps - boiling points vs. ISOLDE yields

