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Basics of Radioactive Ion Beam production

Chair Roger Van Geen
Lecture 1 – 25 October 2021

Outline of this lecture

- Nuclear reactions for the production of radioisotopes
- Luminosity & cross sections
- Examples across the nuclear landscape
  - ➢Industrial production
  - ➢Towards ^{100}Sn
  - ➢Superheavy elements

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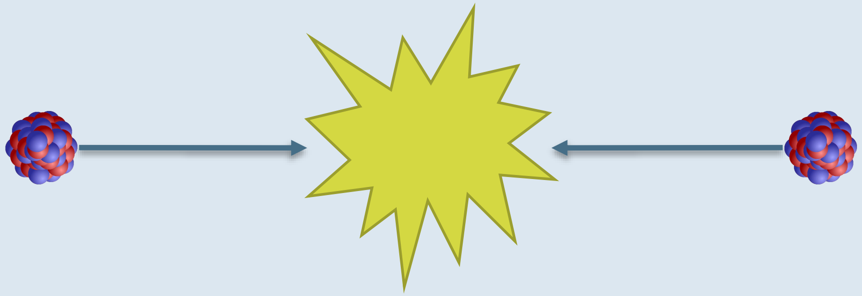

Nuclear reactions

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2 big families

Adding stuff

- Starting with any nucleus, you can add particles to it to make a new, heavier nucleus.
- Possible approaches are neutron capture, proton capture, etc. and particle transfers.
- Things do not always go as planned and those particles may either not stick or the produced nucleus may decide to shed away particles: fusion-evaporation.

Breaking stuff

- Starting with something rather big, it is possible to break it into smaller pieces.
- Depending on the process that triggers this breakdown, we speak about fragmentation, spallation, or fission.
- Each process leads to a different distribution of the end products.
- The different channels may be in competion with each other.

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What is best to induce nuclear reaction

Photon

Electron

Proton

Neutron

Heavy ion

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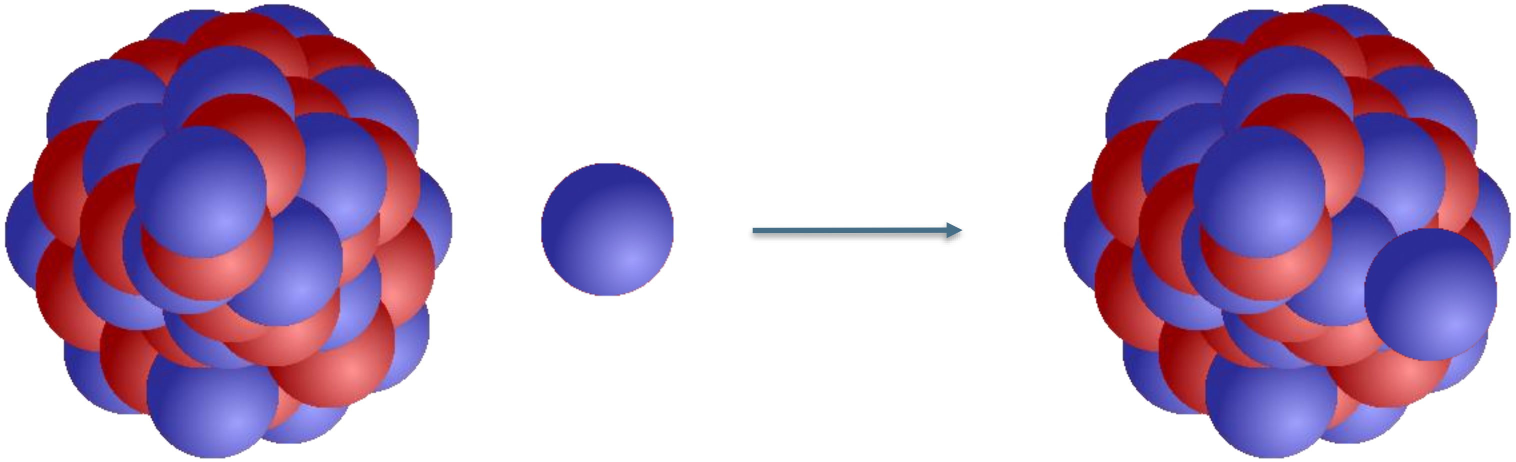
Neutron capture reactions

$^{A}X\ (n,\gamma)\ ^{A+1}X$



$^{A}X_{N}$ + neutron → $^{A+1}X_{N+1}$

<u>neutron capture:</u>

- Same element, new isotope
- Neutron has no charge and must not overcome the Coulomb barrier

➞Reaction can occur from low energy

What is the most efficient E?

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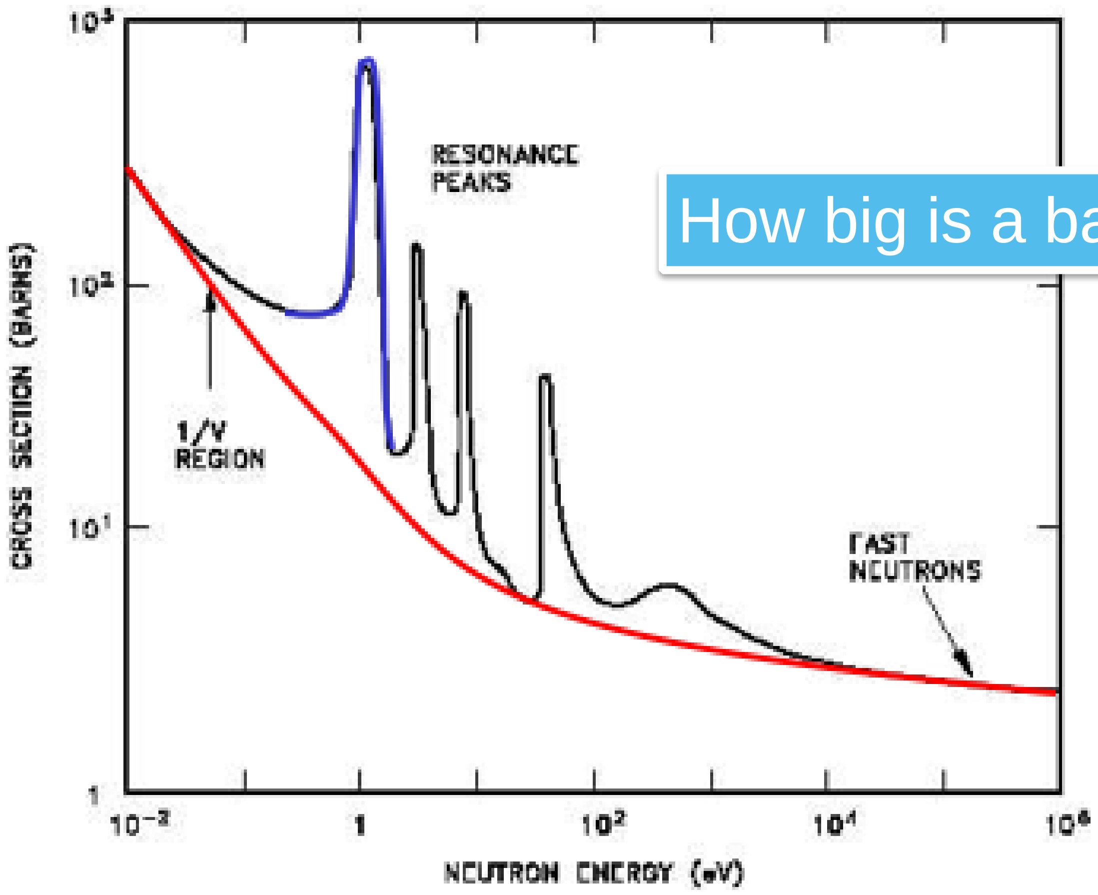
Neutron capture cross sections

- 2 types of reactions: non-resonant and resonant
- Non-resonant reactions scale as $1/v$, favouring slow neutrons over fast neutrons

➞100b → 1b

- Resonant reactions have a Lorentz profile centred around a specific energy, and having an associated width and intensity
- ~100b



How big is a barn!?

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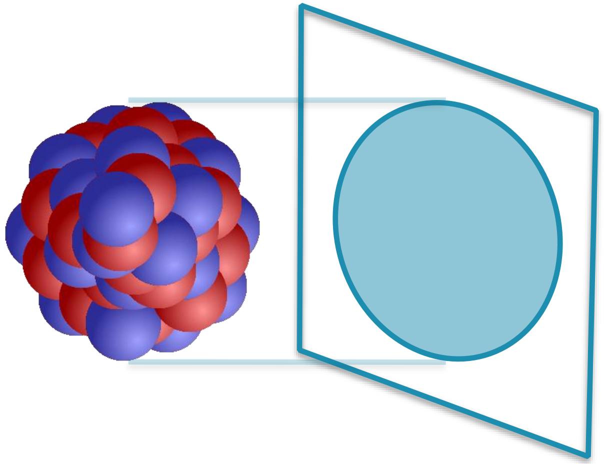
Reaction cross sections

- 1 barn = 10^{-24}cm^2
- 1 barn = 10^{-28}m^2 (SI)
- 1 barn = 100fm^2
- Defines the area that a particle may hit
- 1 barn ~ 2D projection of a nucleus of uranium, corresponding to a radius of 5.64fm
- It is now the measure of the probability of a reaction to occur (nuclear & particle physics)

He could not hit the broad side of a barn!



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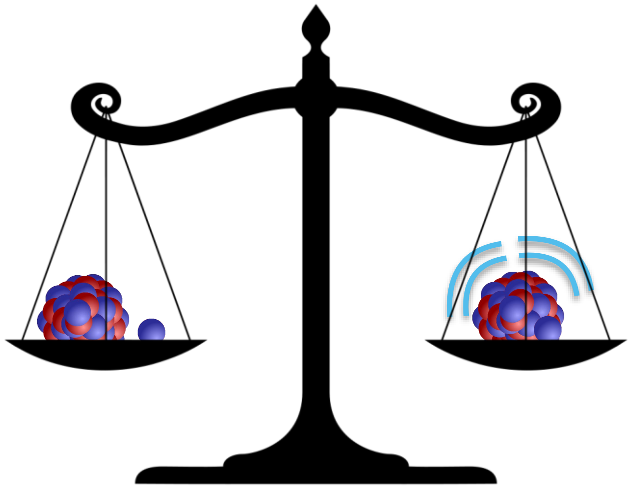
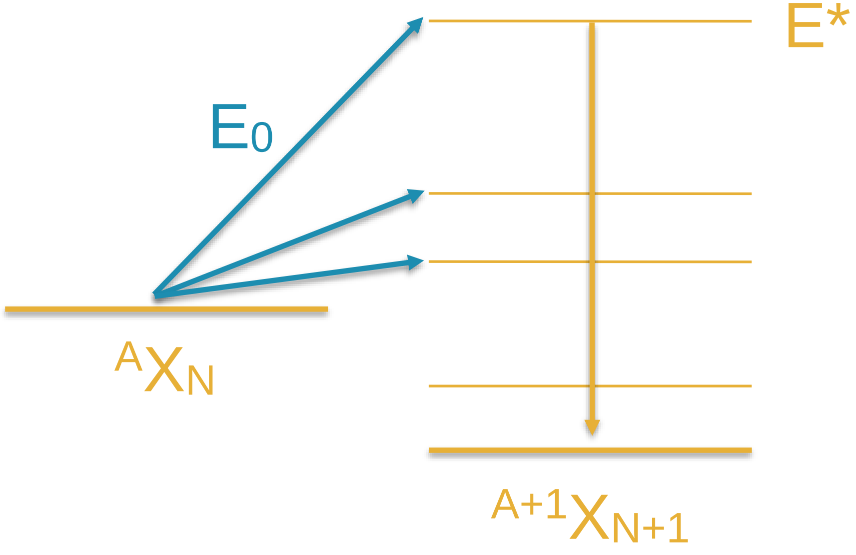
Resonant neutron capture

- The combined energy of the neutron and the nucleus corresponds to an excited state in the new nucleus
- The excited state decays away by any mode that is possible, according to its own properties. This may include γ decay to lower excited states down to the ground state.



$$E_0 + BE[{}^{A}X_N] = E^* + BE[{}^{A+1}X_{N+1}]$$



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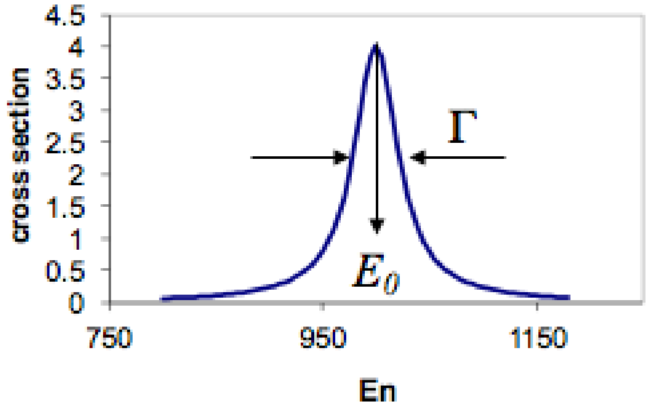
Resonant neutron capture cross section

- It follows the Breit-Wigner formula
- Γ is the width of the excited level
- The width scales with the half-life: $\Gamma = \frac{\ln(2)}{T_{1/2}}$
- $(E_n - E_0)$ is the neutron energy detuning
- The formula needs to account for all the possible decay channels from E^* besides γ-ray emission: particle emission, fission, …

$$\sigma \sim \frac{\Gamma^2}{(E_n - E_0)^2 + \Gamma^2/4}$$



$$\sigma \sim \frac{\Gamma\,\Gamma_\gamma}{(E - E_0)^2 + \Gamma^2/4}$$

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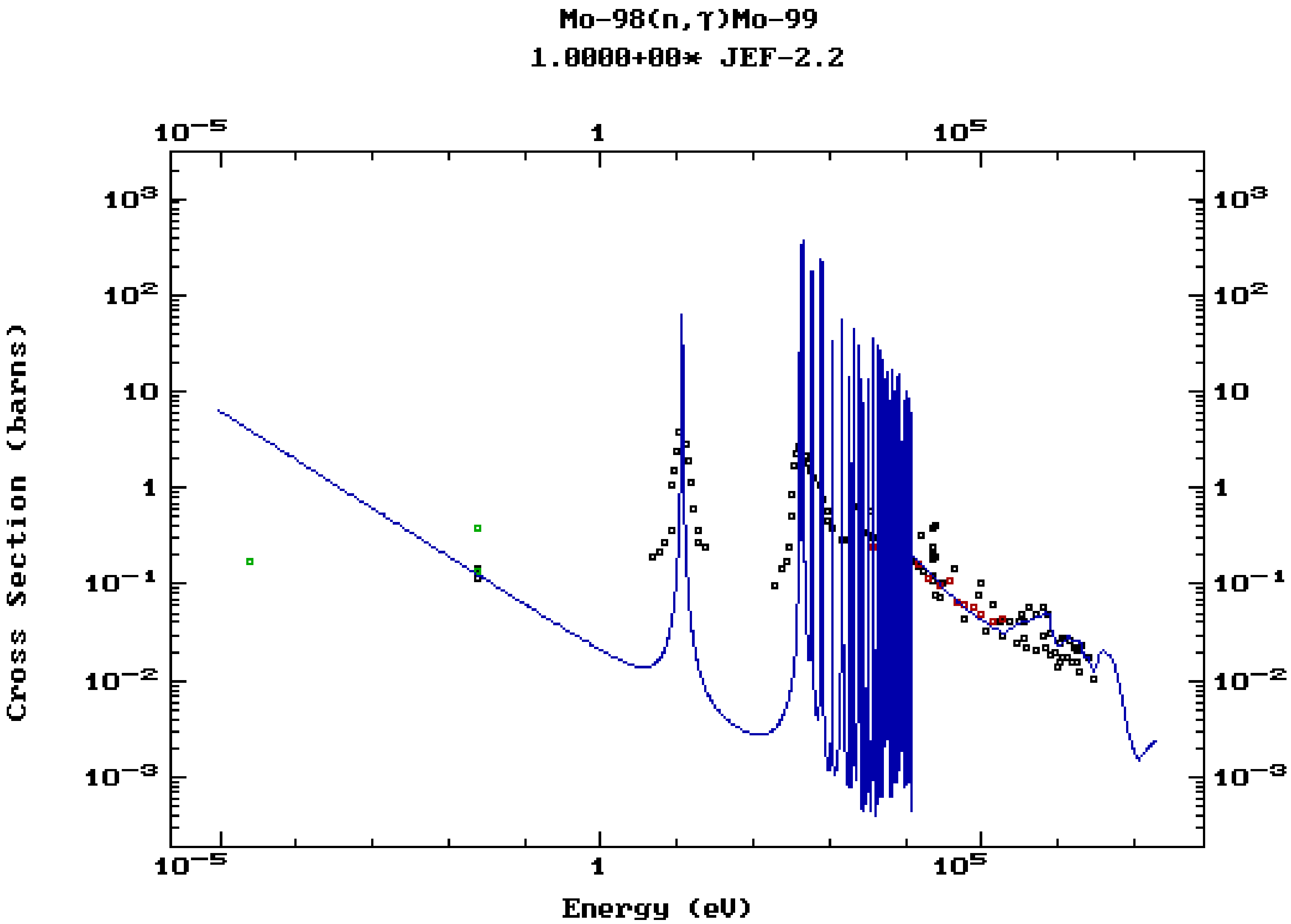
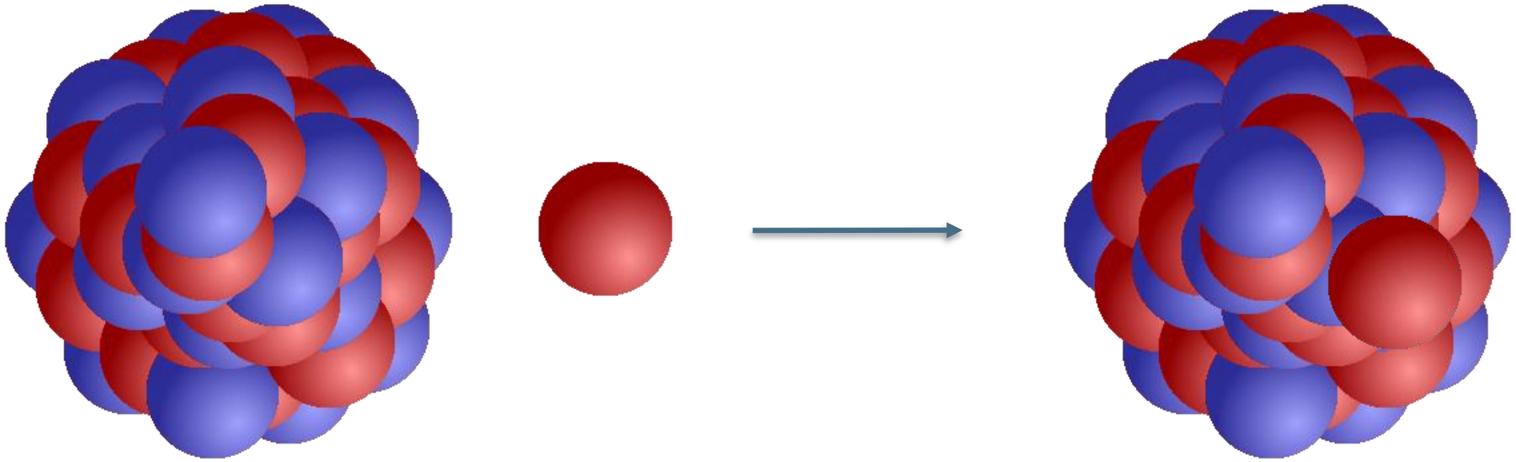
Neutron capture: ^{98}Mo (n,γ) ^{99}Mo



http://www-nds.iaea.org/ngatlas2

Proton capture reactions

$^{A}X\ (p,\gamma)\ ^{A+1}Y$



$^{A}_{Z}X_{N}$ + proton $\quad ^{A+1}_{Z+1}Y_{N}$

proton capture:

- Different element, different isotope
- Proton must overcome the Coulomb barrier

➞Reactions require a powerful driver

What happens if the driver is too powerful?

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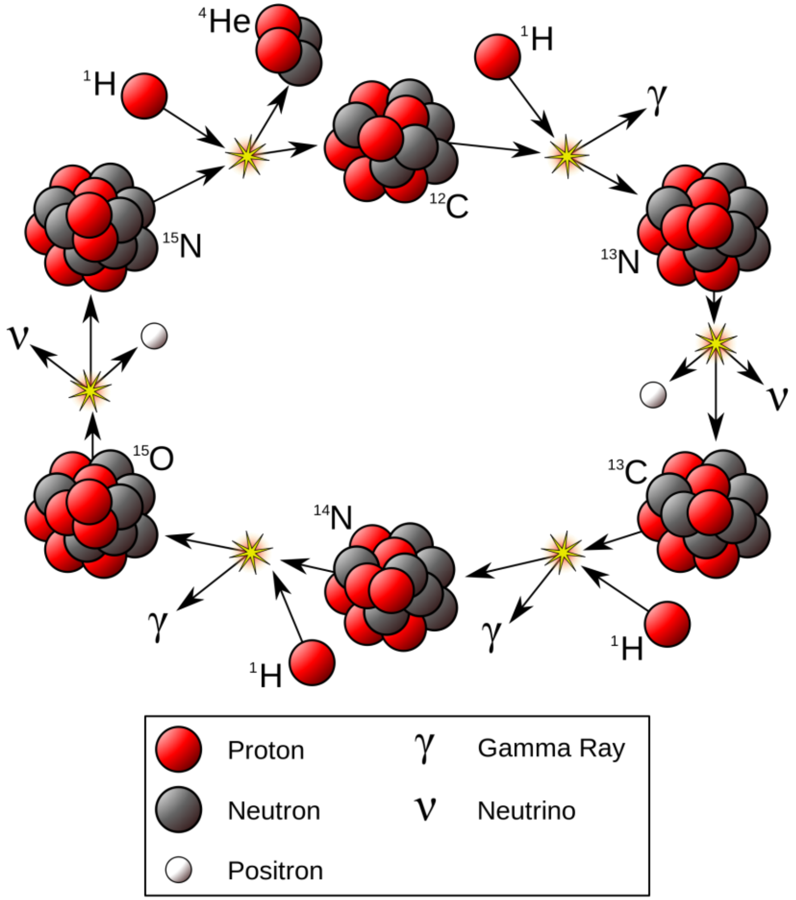
Resonant proton radiative capture

- Similar to resonant neutron capture, including the Coulomb barrier in the energy balance
- Only valid over a small number of states of well-defined energies
- Mostly interesting for astrophysical purpose to understand star burning processes such as the CNO cycle



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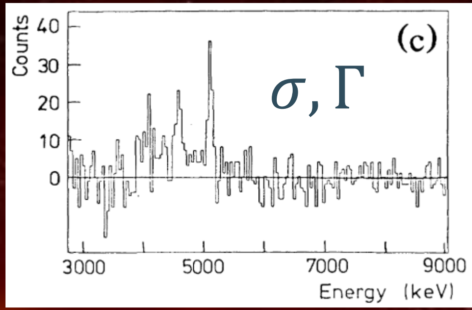
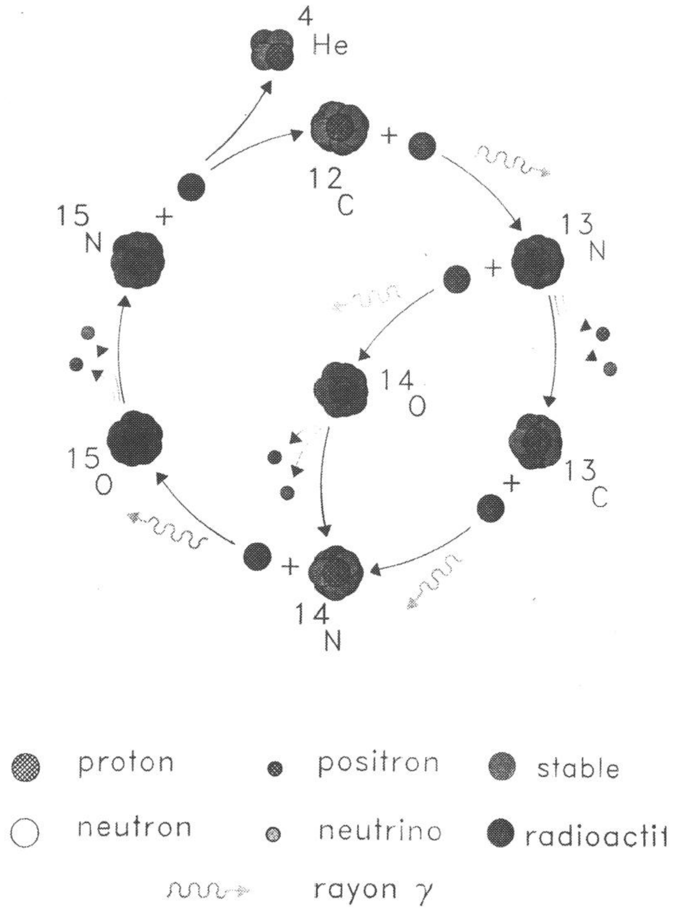
$^{13}N(p,\gamma)^{14}O$: the hot-CNO cycle



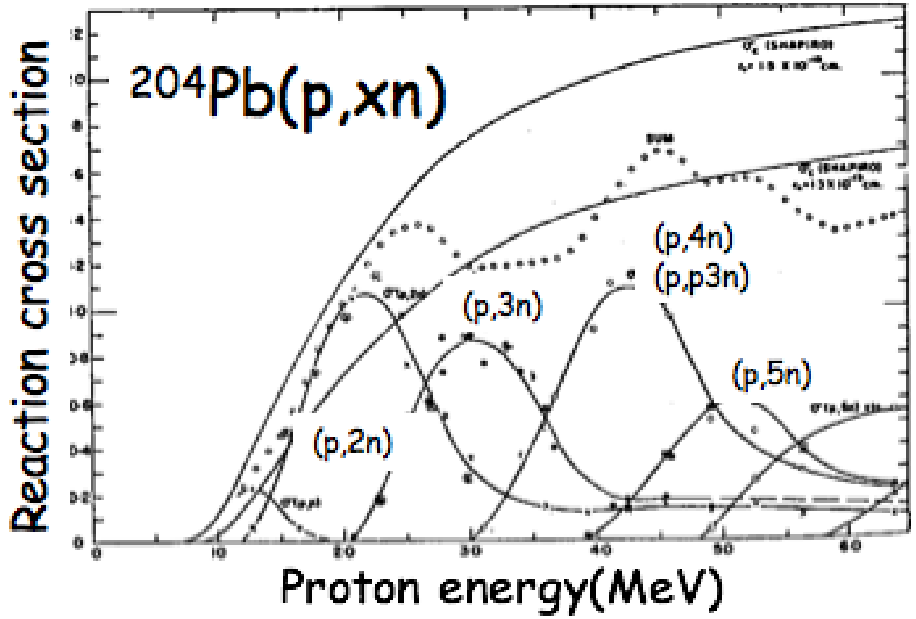


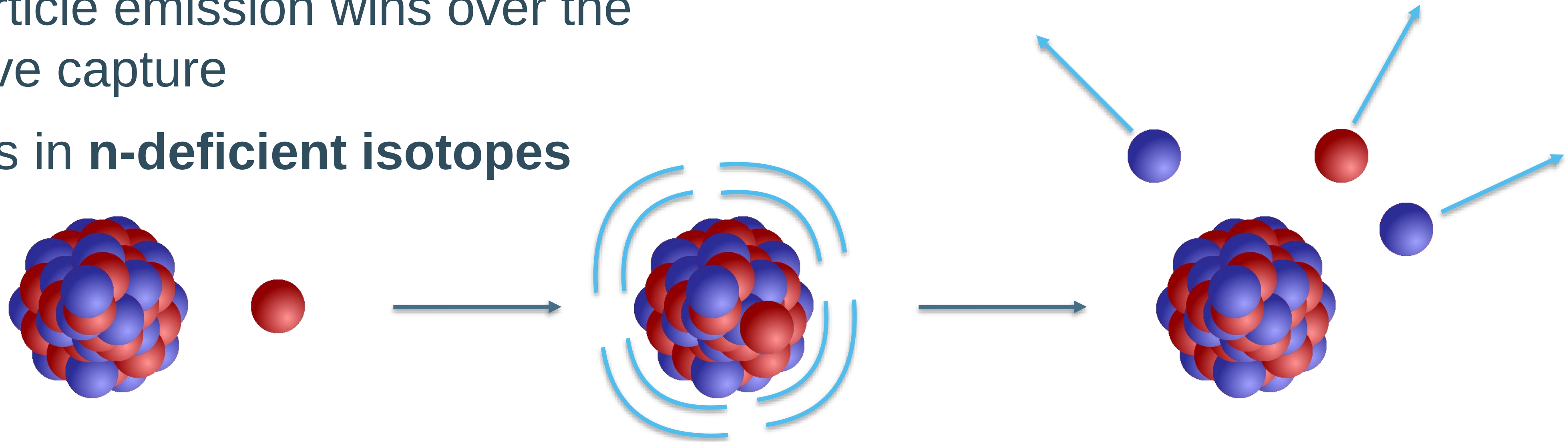
- The CNO-cycle is the astronomical process by which H is burned in C-containing stars.
- Depending on the temperature of the environment, the radioactive ^{13}N ($T_{1/2} = 10\text{min}$) may either decay or capture another proton.
- Measuring the direct reaction rate on the proton capture was a major challenge in the 1980s and gave rise to the first post-accelerated radioactive ion beam, in Belgium, at Louvain-La-Neuve.

P. Decrock et al., Determination of the $^{13}N(p,\gamma)^{14}O$ reaction cross section using a 13N radioactive ion beam, Physical Review Letters ***67*** *(1991) 808-811.*
M. Arnould, G. Paulus & A. Jorissen, Astrophysical implications of the direct measurement of the $^{13}N(p,\gamma)^{14}O$ cross section, Astronomy and Astrophysics ***254*** *(1992) L9-L12.*

Proton fusion-evaporation

- The proton brings so much energy that the compound nucleus is excited beyond the particle emission threshold
- The system is particle-unbound and the particle emission wins over the radiative capture
- Results in **n-deficient isotopes**





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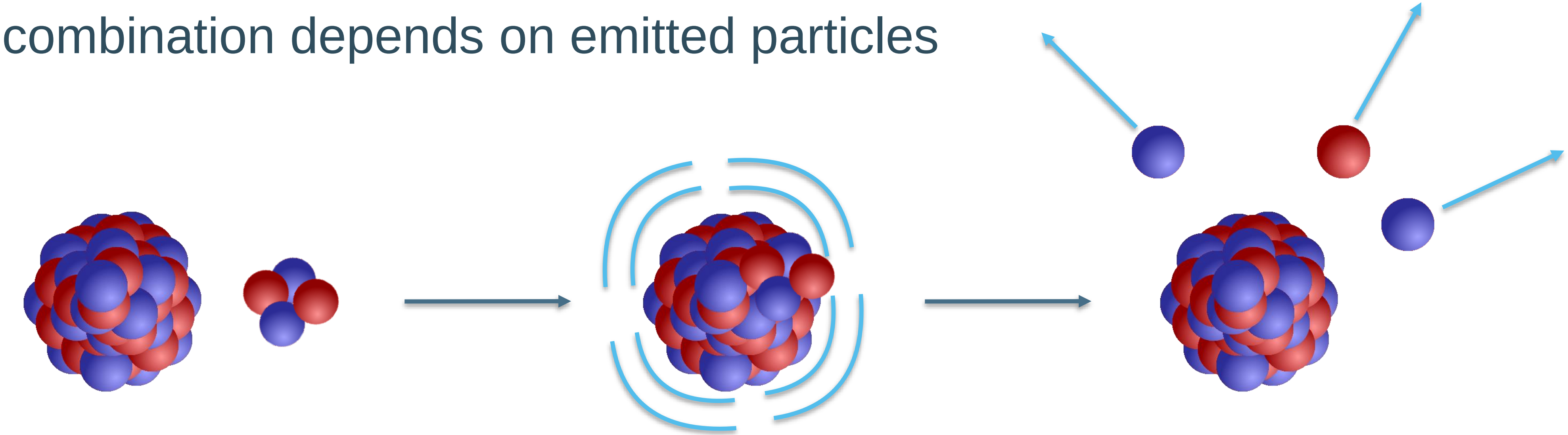
Heavy ion fusion-evaporation

$^{A}X\ (a,b)\ ^{A'}Y$

- Same concept as the proton fusion-evaporation reaction, but using larger projectiles, such as α particles or heavy ion beams
- Beam with Z_b, N_b; target with Z_t, N_t

➞Compound nucleus with $Z_c = Z_b + Z_t$; $N_c = N_b + N_t$

➞Final combination depends on emitted particles



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Fusion-evaporation formalism

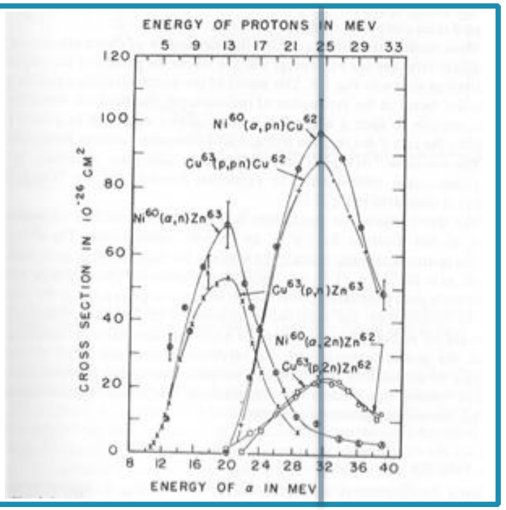
Energy considerations

$$E_{beam} = E_{Coulomb}\,(Z_b, Z_t) + X(\zeta n, \xi p)$$

- You need to break through the Coulomb barrier, which goes typically to ~3-4 MeV/u for heavy ion beams
- The number of particles ζ & ξ evaporated depends upon how much energy is brought in, with ~10 MeV per nucleon evaporated

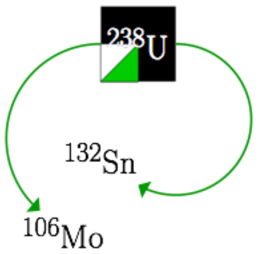
Cross section

- Each target/beam/energy combinations will yield to different outcomes
- Compare on the right the production of ^{62}Cu
  - ✦ $^{60}Ni(\alpha,pn)$: E_C ~ 3 x 4 = 12 MeV; X(1,1) ~ 20 MeV
  - ✦ $^{63}Cu(p,pn)$: E_C ~ 3 x 1 = 3 MeV; X(1,1) ~ 20 MeV
- Many things may be produced with a single combination

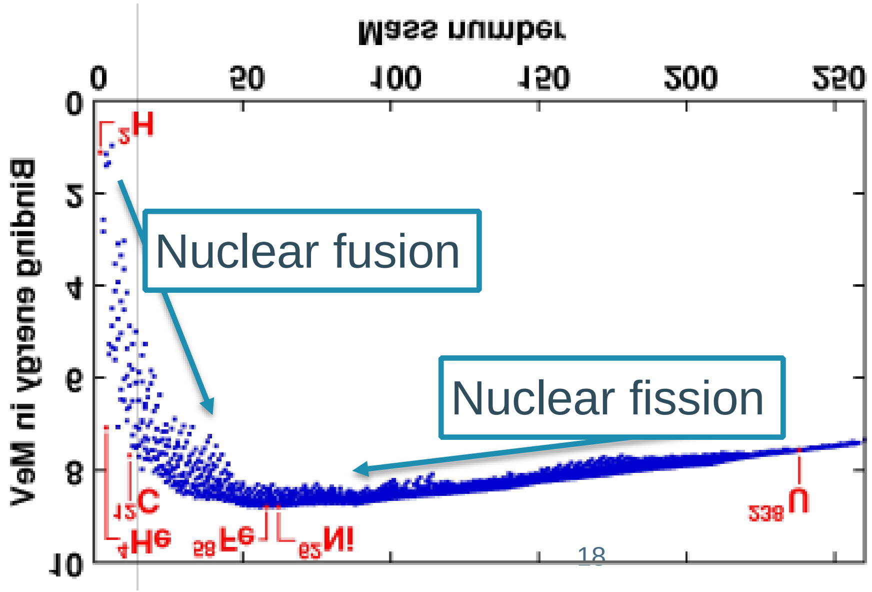


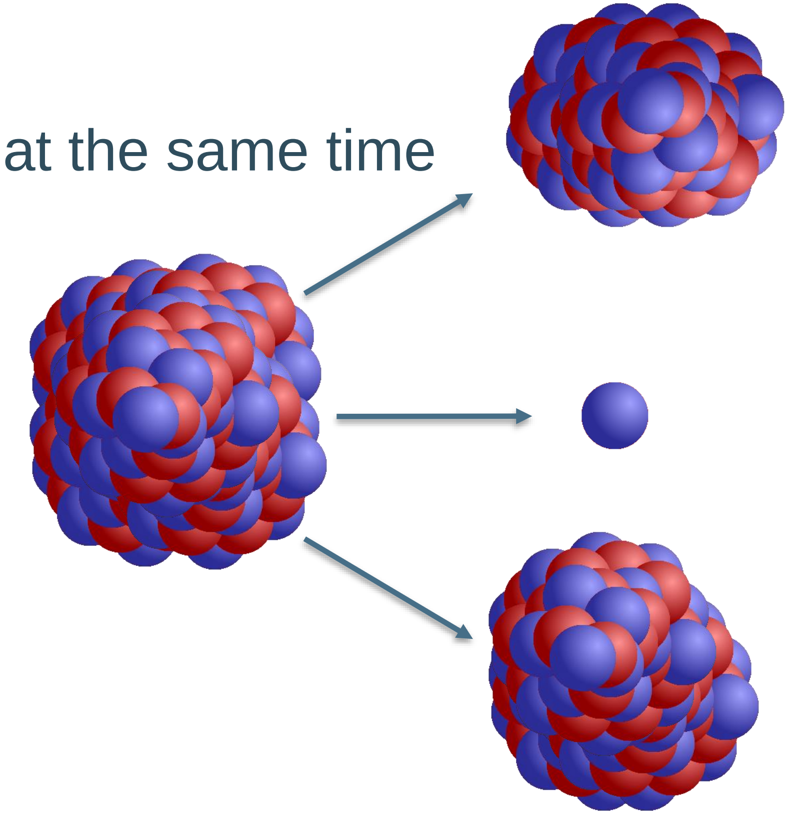
How do we guarantee the quality of the product?

Nuclear fission



- Nuclei always try to minimise energy
- Heavy nuclei may split into two smaller pieces
- Energy is released as those nuclei are more bound
- Additional particles (typically neutrons) can be emitted at the same time





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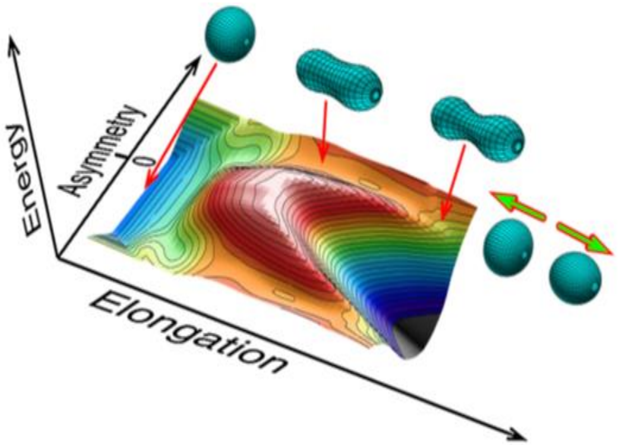
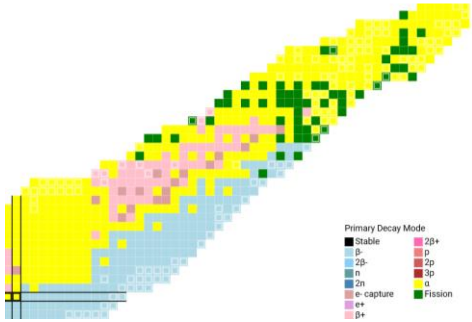
Nuclear fission

Fission barrier

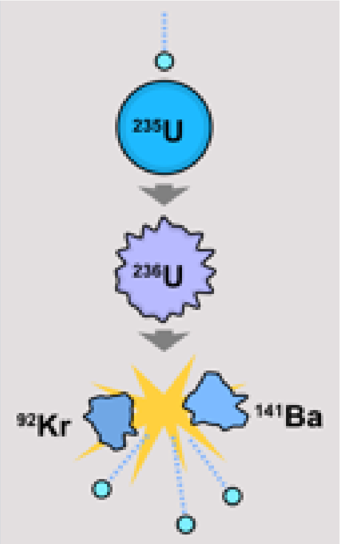
- Nuclei are bound to a local energetic minimum.
- To undergo fission, they have to overcome an energy wall beyond which more energy can be gained.
- Depending on the height of the barrier, fission can happen spontaneously, be induced by thermal neutrons, or require high energy bombardment.



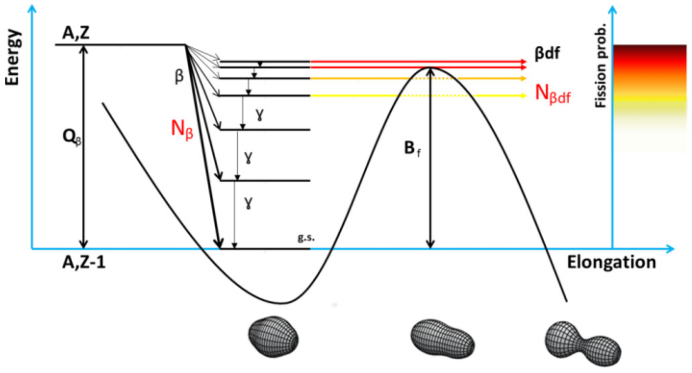
Spontaneous fission is found in the very & super heavy elements.



Particle-induced is most commonly used. Research with γ-induced fission has been done as well.



β-delayed fission can compete with γ decay



A.N. Andreyev et al, PRL ***105*** *(2010) 252502.*
L. Ghys et al, PRC ***91*** *(2015) 0443314.*

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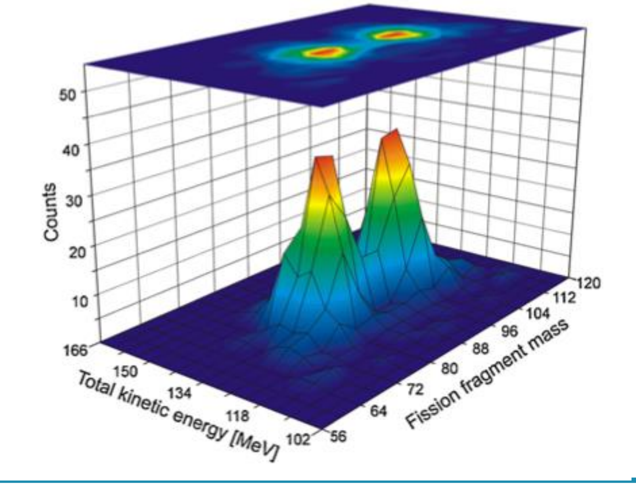
Fission fragments distribution

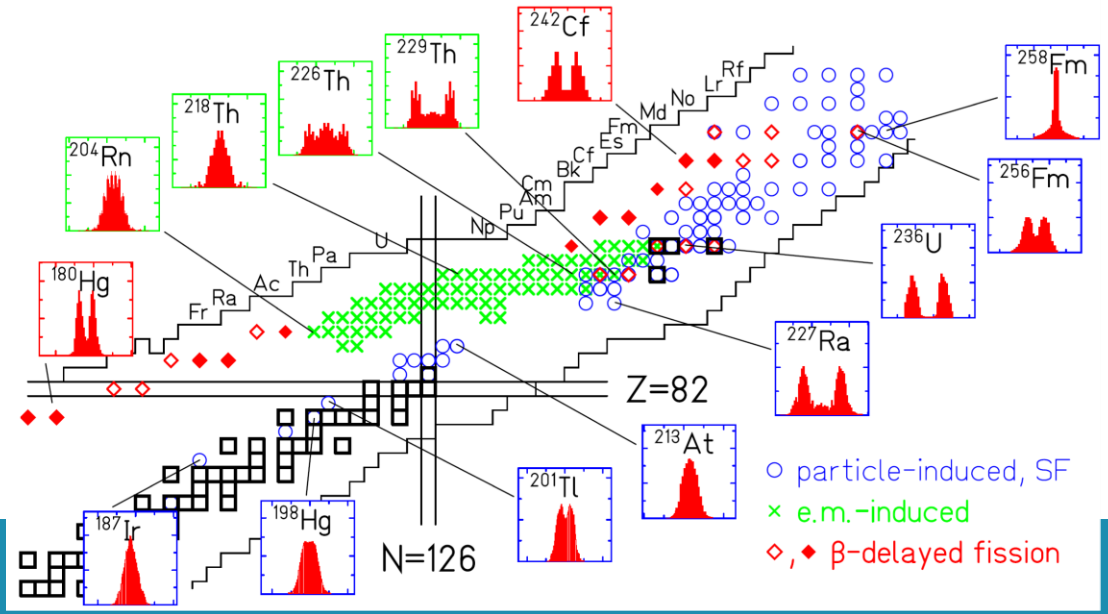
<u>Fission fragments</u>

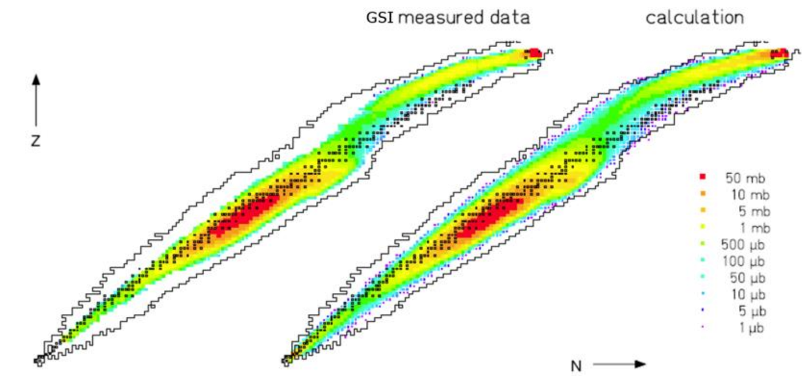
- They keep the same N/Z balance as the parent nucleus

➞Produces mostly **n-rich isotopes**

- The fission may be symmetric or asymmetric depending upon energy considerations (parent’s excitation energy, nuclear structure of the fragments)







1GeV protons on ^{238}U in inverse kinematics

S. Lukić et al, NIMA ***565*** *(2006) 784-800.*

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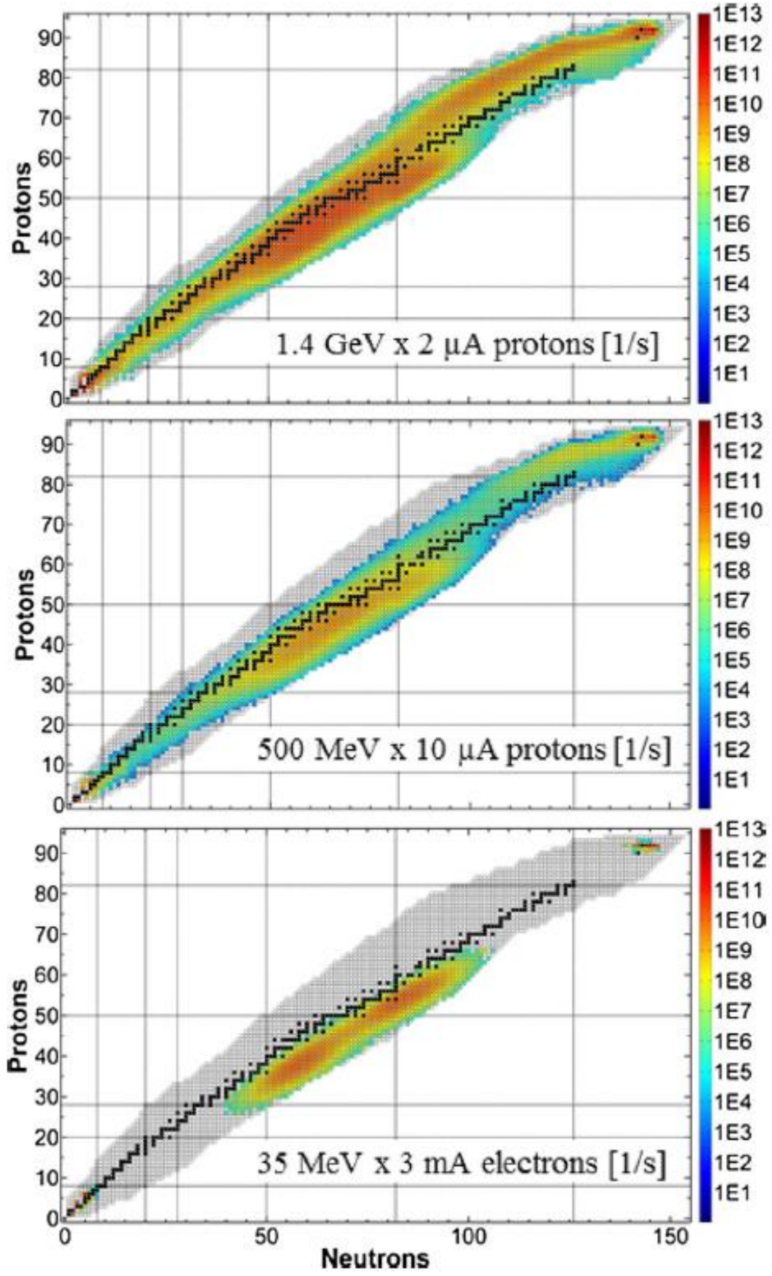
Fission of ^{238}U

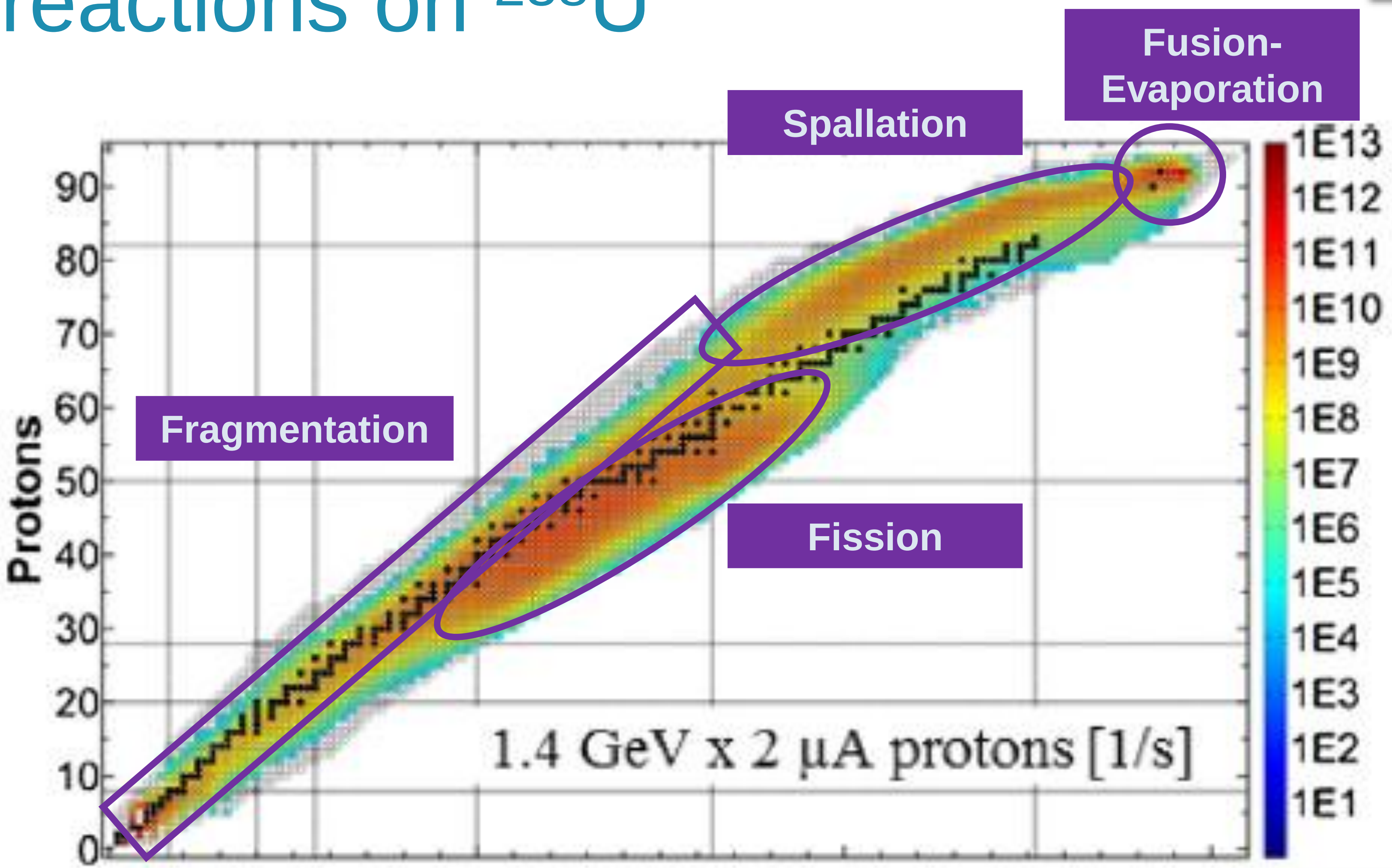
- ^{238}U is the most neutron-rich isotope that is easily available and with which to make a target. It is therefore the target of choice for the production of exotic radioisotopes.
- ^{252}Cf is a very interesting alternative, however it cannot be used with as high quantities as ^{238}U.
- Particle energy and type have a big impact on the fragment distribution.



 *A. Gottberg, NIMB **376** (2016) 8-15*

Other reactions on ^{238}U

^{238}U (p,x)



… and many more secondary reactions

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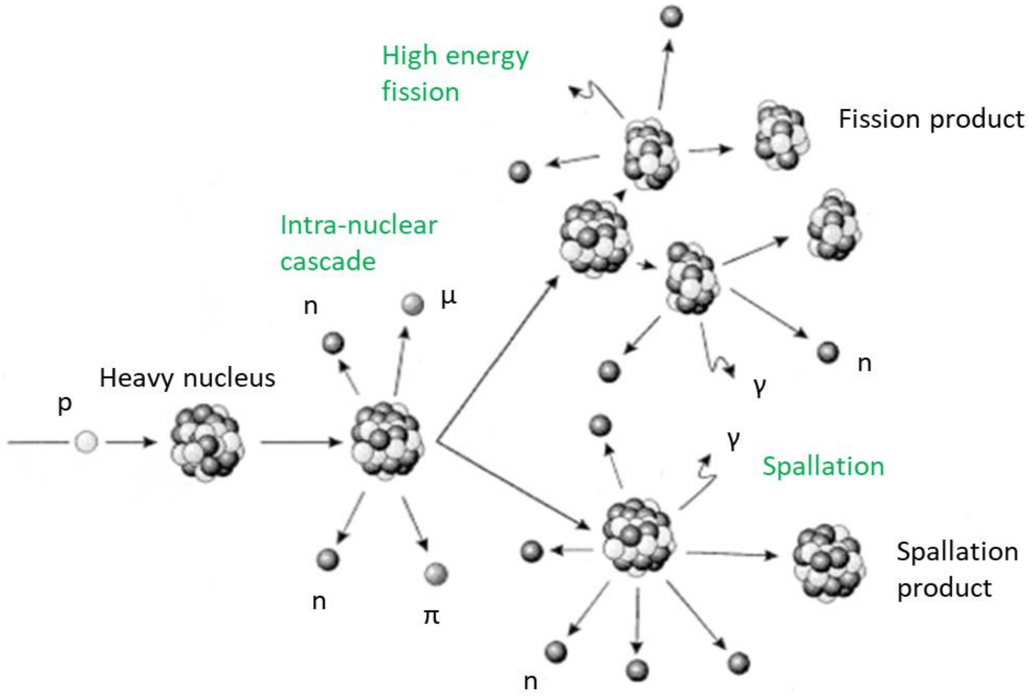
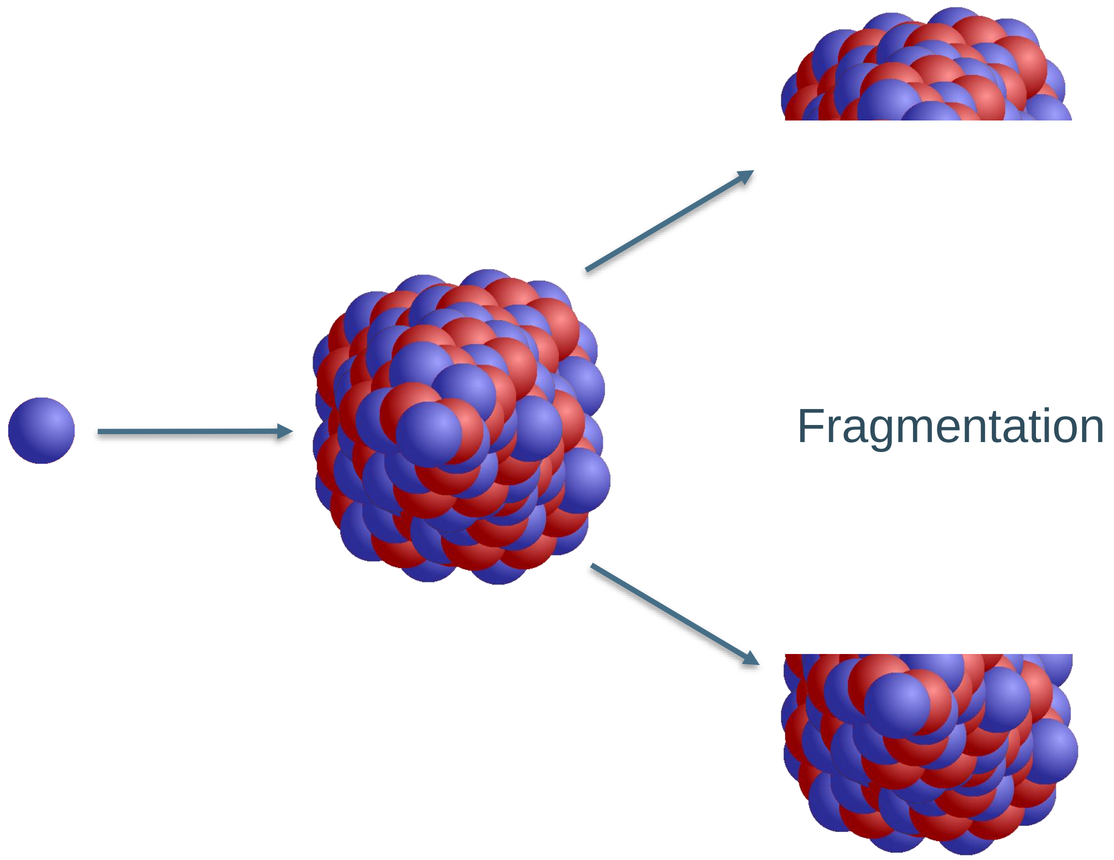
Multiple reaction channels





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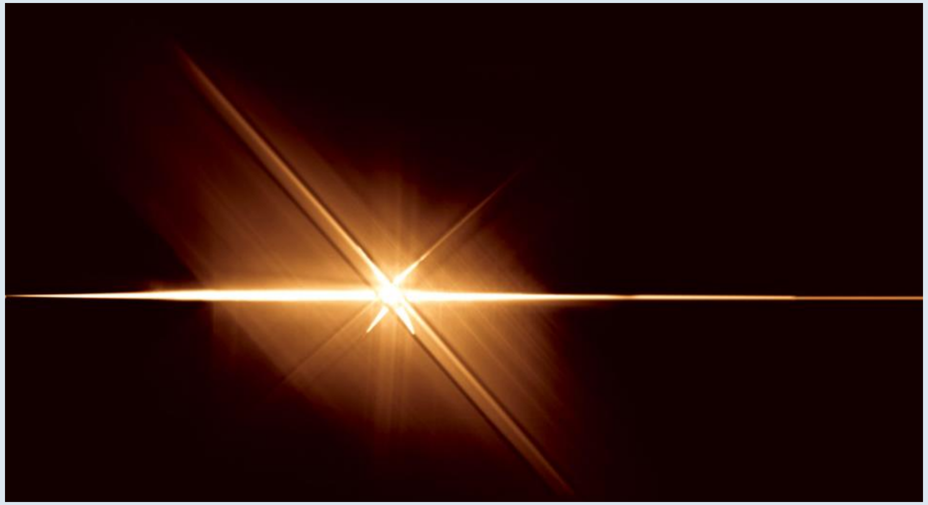
Luminosity & cross sections



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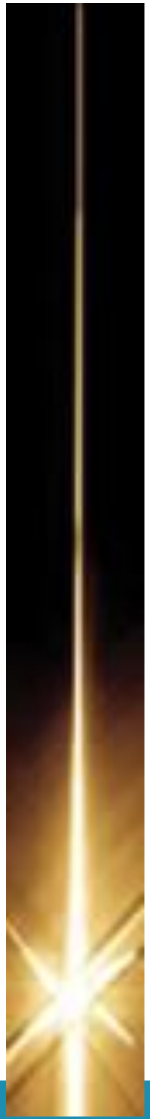
Luminosity in beam-induced reactions

- The luminosity is the unbiased measure of the reaction ingredients
  - number of incoming beam particles N_b
  - density of the target d_t
  - thickness of the target a_t
  - $d_t a_t$ is the number of target atoms per unit area

$$L = N_b \cdot d_t a_t$$



- The luminosity can also be determined in terms of more practical parameters
  - beam flux $\varphi = N_b \,/\, S$,where S is the beam spot size
  - $U = S d_t a_t$ is the number of target atoms within the beam spot

$$L = \varphi \cdot U$$

Beam-induced nuclear reactions

Production:
- Nuclear reaction scales with the cross section σ
- Nuclear reaction scales with the luminosity L

$$\frac{dn}{dt} = +L\sigma$$
$$= +N_b \cdot d_t a_t \cdot \sigma$$

Destruction:
- Radioactive decay is a net loss in the population
- Radioactive decay goes inverted to the decay half-life

$$\lambda = \frac{\ln 2}{T_{1/2}}$$
$$\frac{dn}{dt} = -\lambda n$$

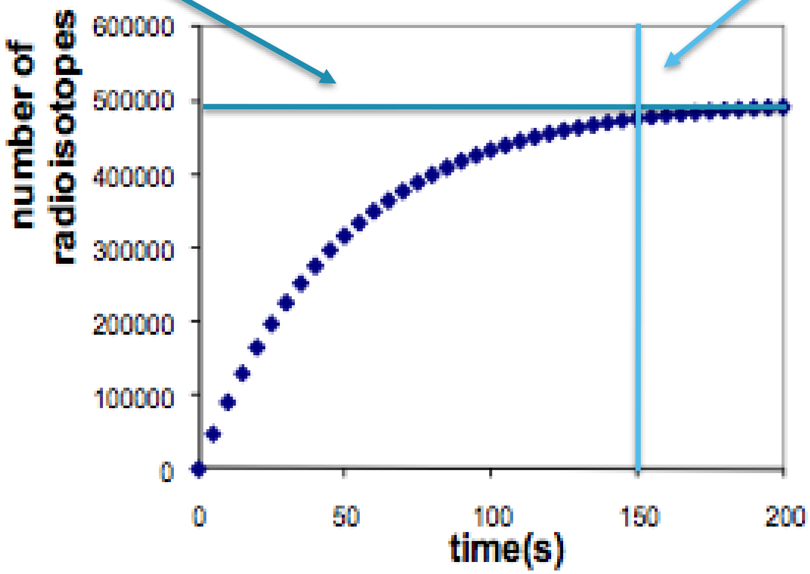
$$\frac{dn}{dt} = +N_b \cdot d_t a_t \cdot \sigma - \lambda n$$

Beam-induced nuclear reactions

$$n(t) = \frac{N_b \cdot d_t a_t \cdot \sigma}{\lambda}\left(1 - e^{-\lambda t}\right)$$

Saturation level depends on
- Beam intensity N_b
- Target atoms $d_t a_t$
- Cross section σ
- Half-life

Saturation time depends on
- Half-life



Saturation is reached after 3 lifetimes

$$T_S = 3\tau = \frac{3}{\lambda} = 3\,\frac{T_{1/2}}{\ln 2}$$

Activity

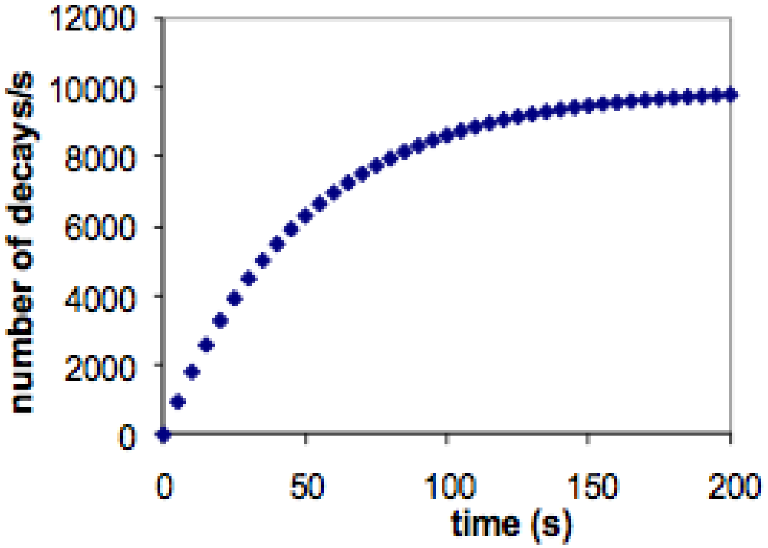
What assumptions are made for this solution?

<u>Nuclear activity:</u>

- Number of radioactive decays per unit time in Bq [decays/s]

$$A = \lambda n(t)$$
$$A = N_b \cdot d_t a_t \cdot \sigma\left(1 - e^{-\lambda t}\right)$$

- The activity saturates also after 3 lifetimes
- The maximum activity is independent of the half-life, and only depends on the cross section and luminosity
- The time it takes to reach this activity still depends on the half-life



Neutron-capture reactions

High neutron flux φ:

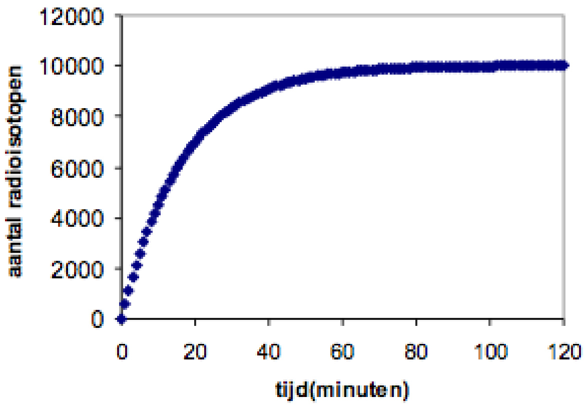
- If the neutron flux φ is high enough, the target material U is turned into something new, n, and disappears:

$$\frac{dU}{dt} = -\varphi \cdot \sigma \cdot U(t) \qquad U(t) = U_0 e^{-\varphi\sigma t}$$

Long half-life limit:

- Assuming that the half-life of the daughter is so long that we may neglect the decay:

$$n(t) = U_0(1 - e^{-\varphi\sigma t})$$



- The saturation level depends only on the number of target atoms → complete transmutation!
- Saturation time depends on flux and cross section
- Activity is proportional to λ, which depends on $T_{1/2}$

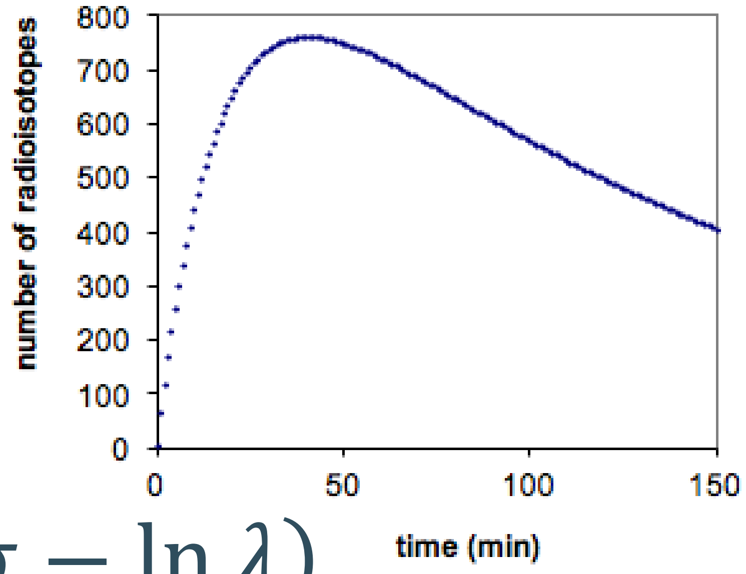
Neutron-capture reactions

Considering radioactive decay:

- The reaction differential equation becomes:

$$\frac{dn}{dt} = \varphi \cdot \sigma \cdot U(t) - \lambda n(t) = \varphi \cdot \sigma \cdot e^{-\varphi\sigma t} - \lambda n(t)$$

$$n(t) = \frac{\varphi\sigma U_0}{\varphi\sigma - \lambda}\left(e^{-\lambda t} - e^{-\varphi\sigma t}\right)$$



- The production does not saturate but instead reaches a maximum

$$\frac{dn}{dt}(t_{max}) = 0 \rightarrow t_{max} = \frac{1}{\varphi\sigma - \lambda}(\ln \varphi\sigma - \ln \lambda)$$

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Neutron capture & activity

Production

$$n(t) = \frac{\varphi\sigma U_0}{\varphi\sigma - \lambda}\left(e^{-\lambda t} - e^{-\varphi\sigma t}\right)$$

- builds up until t_{max}
- max value scales up with $T_{1/2}$
- for very long-lived isotopes, there can be as much as U_0

Activity

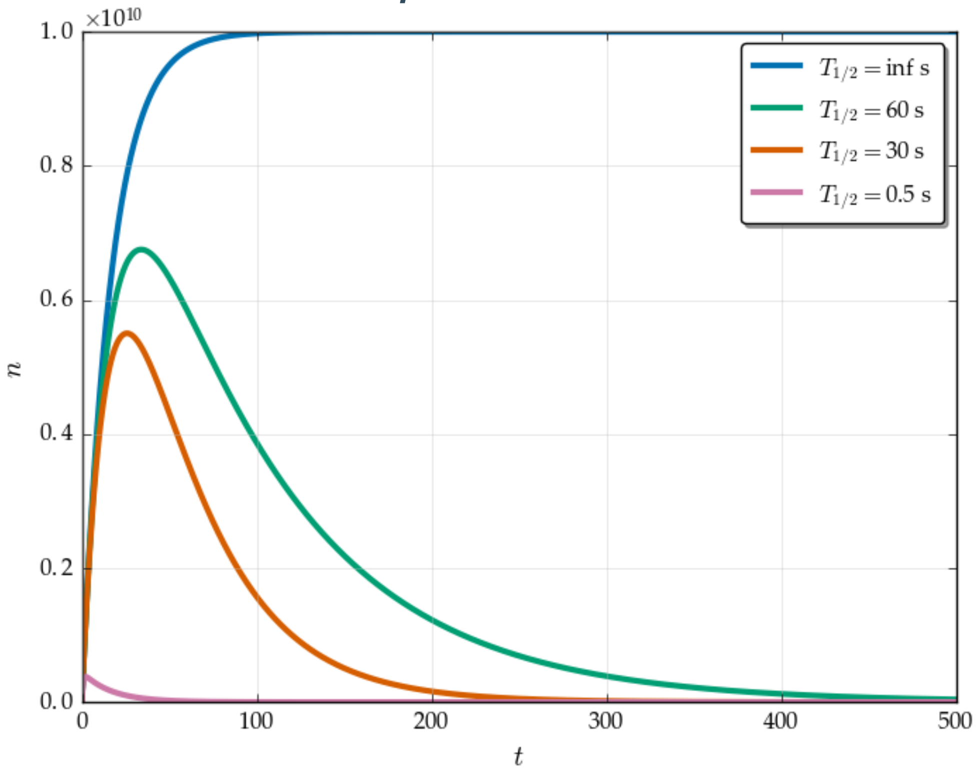
$$A(t) = \lambda\frac{\varphi\sigma U_0}{\varphi\sigma - \lambda}\left(e^{-\lambda t} - e^{-\varphi\sigma t}\right)$$

- max value at the same t_{max}
- max values scales down with $T_{1/2}$

Neutron capture & activity

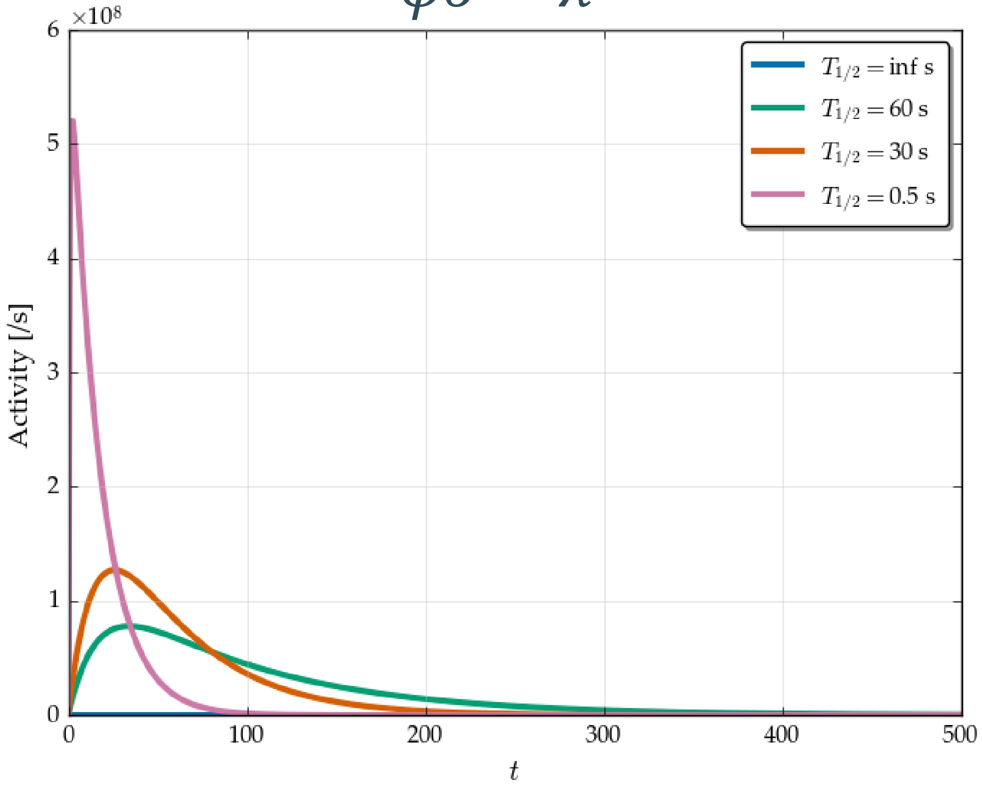
Production

$$n(t) = \frac{\varphi\sigma U_0}{\varphi\sigma - \lambda}\left(e^{-\lambda t} - e^{-\varphi\sigma t}\right)$$



Activity

$$A(t) = \lambda\frac{\varphi\sigma U_0}{\varphi\sigma - \lambda}\left(e^{-\lambda t} - e^{-\varphi\sigma t}\right)$$



BREAK

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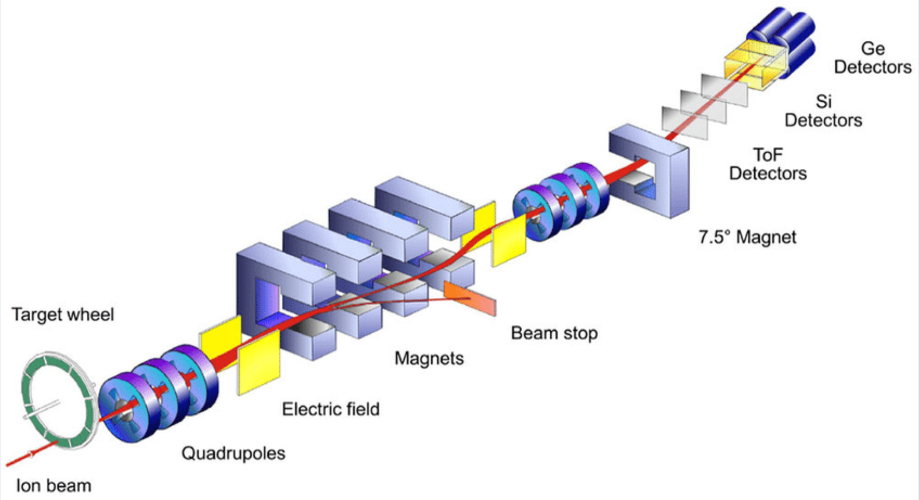

Specific examples

- Industrial production
- Towards ^{100}Sn
- Reaching out to the superheavy elements

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Industrial production

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Case study: ^{60}Co

- $^{59}Co\ (n,\gamma)\ ^{60}Co$ at nuclear reactors
- $T_{1/2}$ = 5.27 y vs full transmutation in a few months
  - $\lambda = 4.2 \times 10^{-9}$ Hz
  - $\varphi\sigma \sim 3 \times 10^{-7}$ Hz dominates
  - ➞ Specific activity is highest achievable on the account that the whole mass is radioactive. Samples can reach up to 900 Ci/g.
- Other reactions can happen such as $^{60}Co\ (n,\gamma)\ ^{61}Co$
  - $T_{1/2}$ = 1.6h → small waiting time can yield 100% purity
  - The specific activity is reduced accordingly

1 Ci = 3.7 10^{10} Bq = 37 GBq

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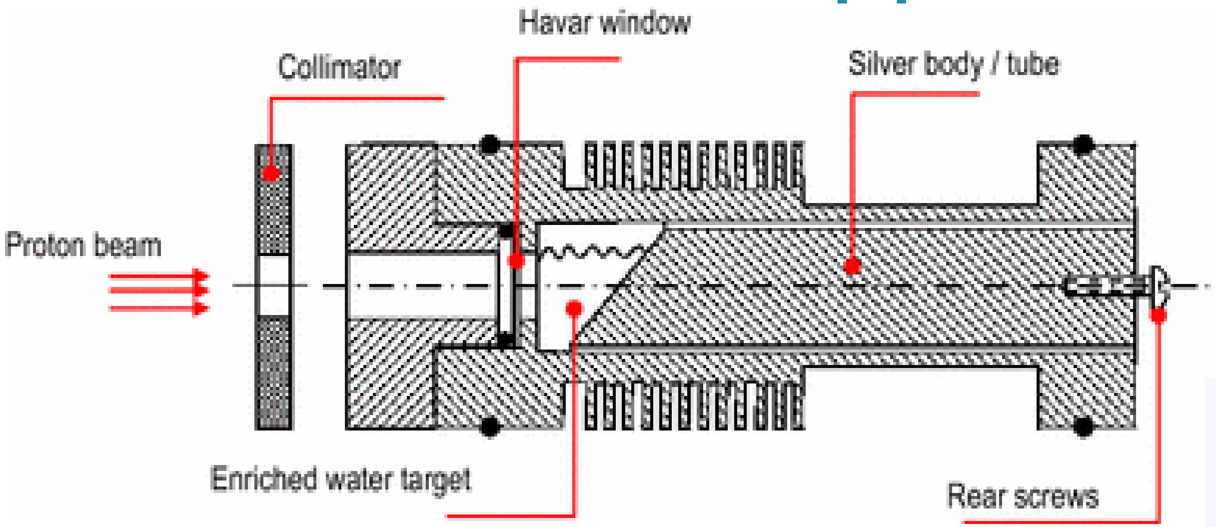
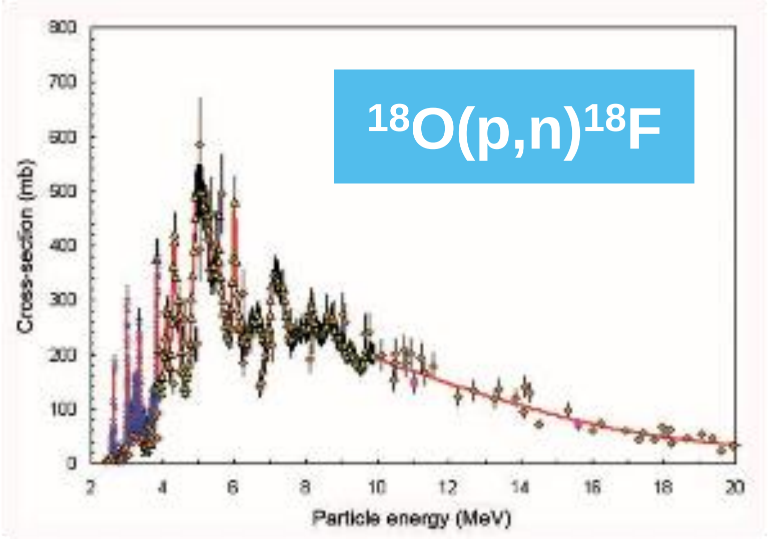
^{18}F for medical applications





I. Kambali, H. Suryanto, Parwanto, *Australasian Physical & Engineering Sciences* **39** (2016) 403-412.

Towards ^{100}Sn

- How it was produced so far
- Up and coming: S3 at GANIL

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^{100}Sn as seen in GSI

^{124}Xe fragmentation
0.16 pnA

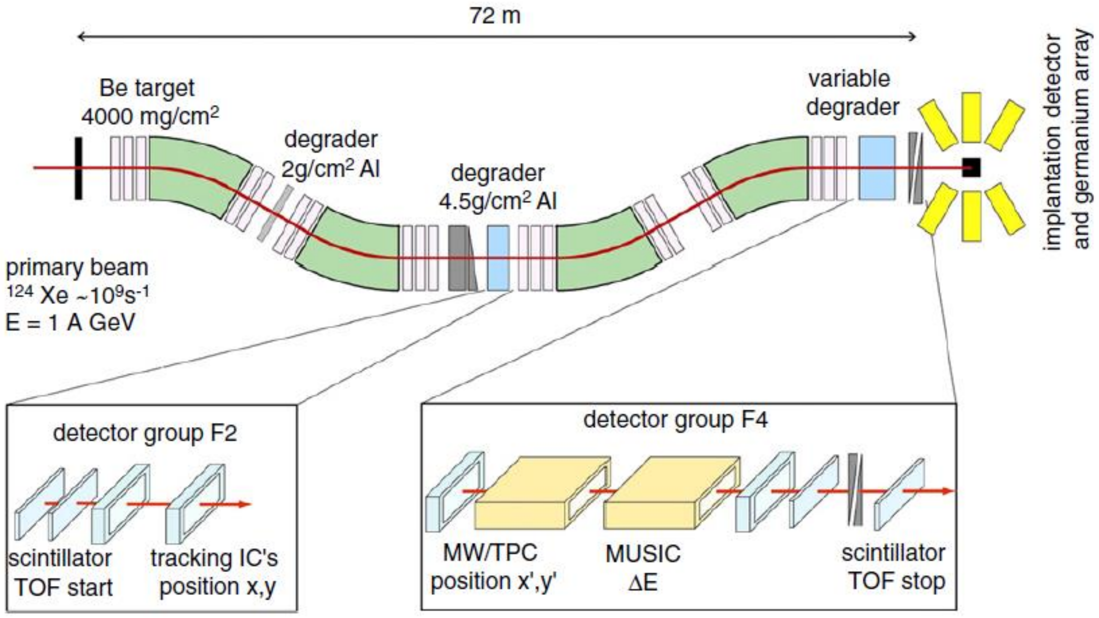


Fig. 2.4.1. The fragment separator FRS at GSI [97].

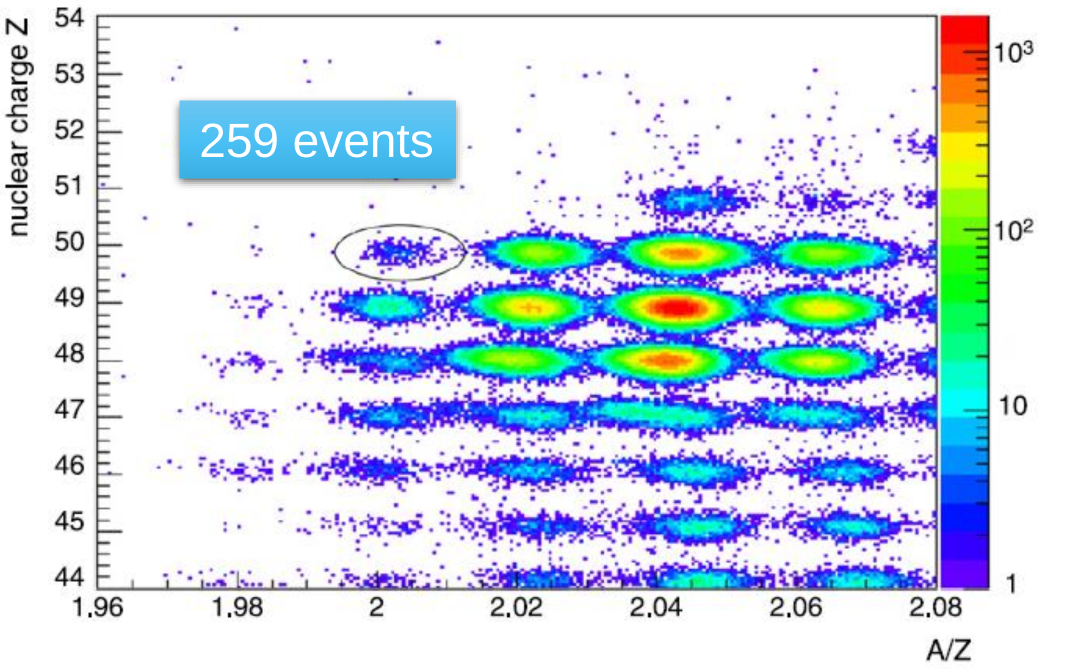


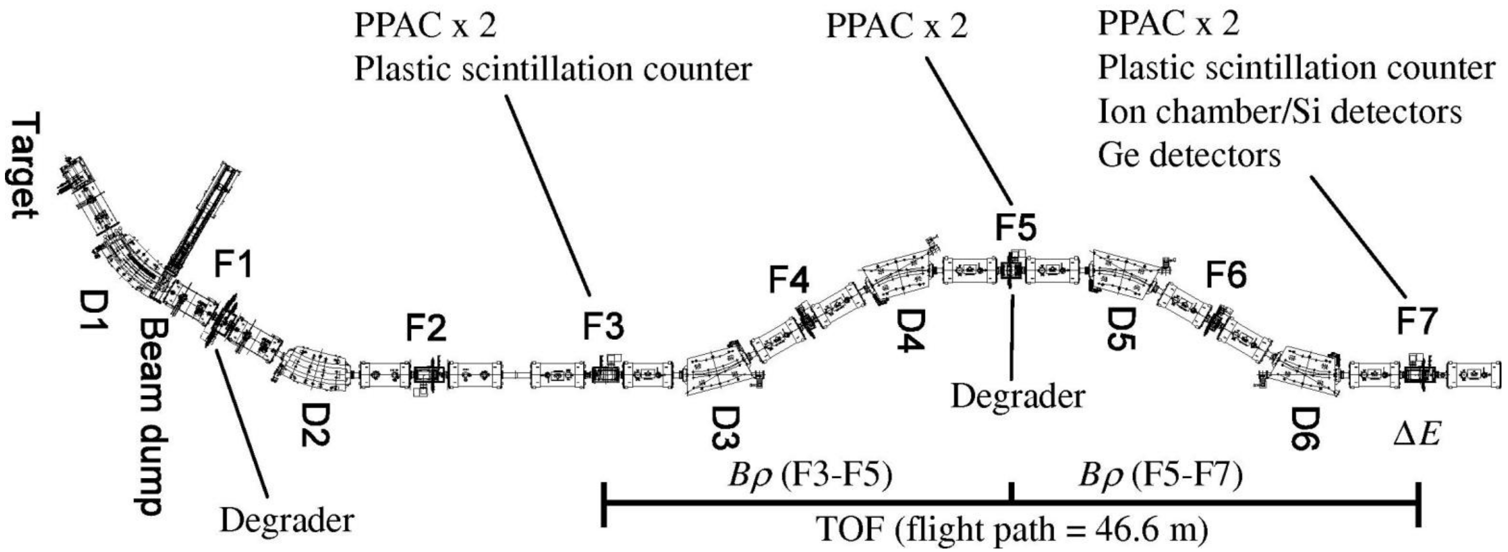
Fig. 2.4.2. Identification plot of ^{124}Xe fragmentation at GSI [33]. ^{100}Sn events are encircled.

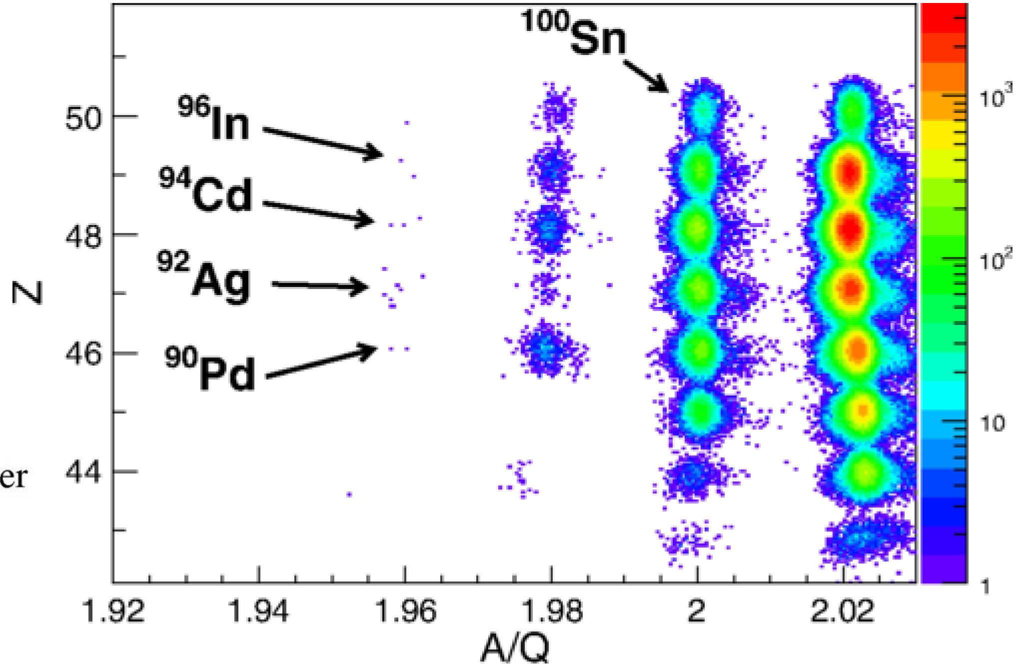
C.B. Hinke et al., *Superallowed Gammow-Teller decay of the doubly magic nucleus ^{100}Sn, Nature **486** (2012) 341-345.*
*T. Faestermann, M. Górska & H. Grawe, The structure of ^{100}Sn and neighbouring nuclei, Progress in Particle and Nuclear Physics **69** (2013) 85-130.*

^{100}Sn with EURICA & WAS3ABi at RIKEN

2500 events

^{124}Xe fragmentation
36 pnA





N. Fukuda et al., NIMB **317**(2013) 323-332.
I. Čelicović et al., PRL **116** (2016) 162501.
D. Lubos et al, PRL **122** (2019) 222502.

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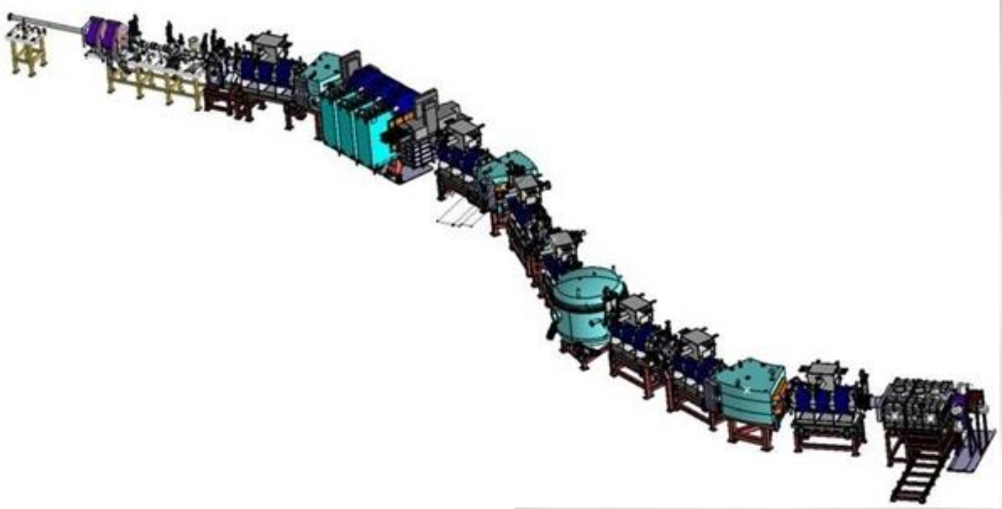
^{100}Sn tomorrow: S^3 at GANIL



1-20 ions/s at the focal plane

^{58}Ni(^{50}Cr,x)^{100}Sn fusion-evaporation
4.5 MeV/u
4000 pnA

Could match the existing statistics in just over 2 minutes!

 https://u.ganil-spiral2.eu/chartbeams/

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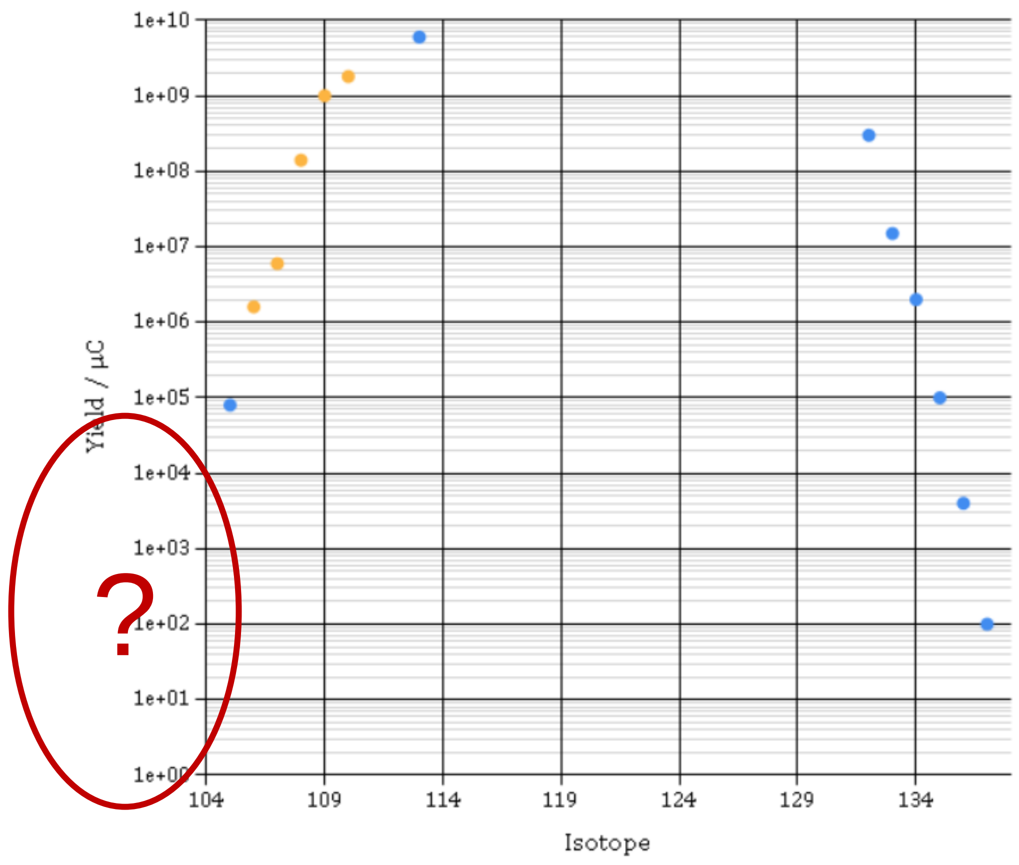
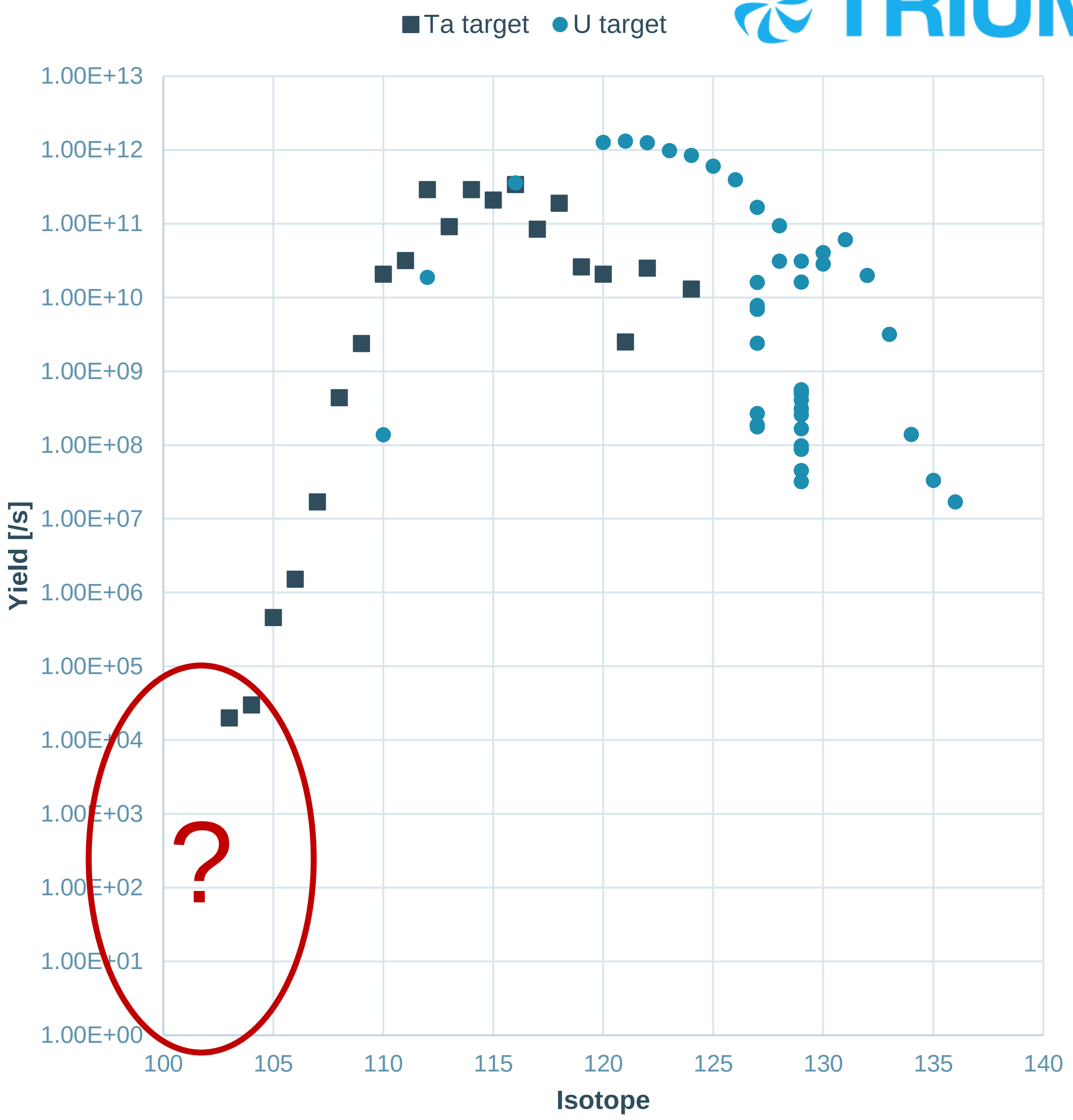
^{100}Sn at ISOL facilities?





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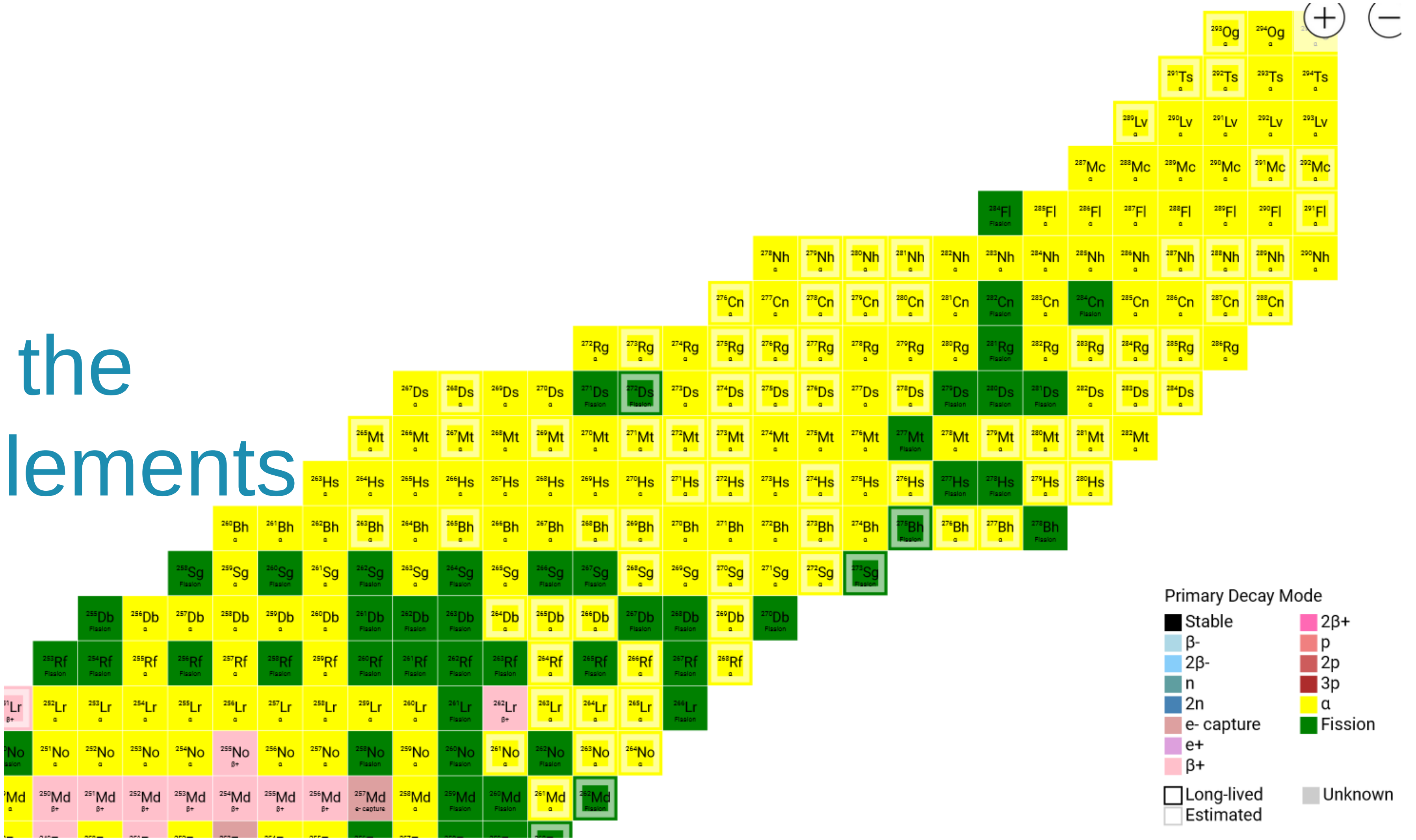
Reaching out the superheavy elements

- Cold fusion
- Hot fusion



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The Periodic Table of Elements

scientificgems.wordpress.com

1 H Hydrogen																	2 He Helium
3 Li Lithium	4 Be Beryllium											5 B Boron	6 C Carbon	7 N Nitrogen	8 O Oxygen	9 F Fluorine	10 Ne Neon
11 Na Sodium	12 Mg Magnesium											13 Al Aluminium	14 Si Silicon	15 P Phosphorus	16 S Sulfur	17 Cl Chlorine	18 Ar Argon
19 K Potassium	20 Ca Calcium	21 Sc Scandium	22 Ti Titanium	23 V Vanadium	24 Cr Chromium	25 Mn Manganese	26 Fe Iron	27 Co Cobalt	28 Ni Nickel	29 Cu Copper	30 Zn Zinc	31 Ga Gallium	32 Ge Germanium	33 As Arsenic	34 Se Selenium	35 Br Bromine	36 Kr Krypton
37 Rb Rubidium	38 Sr Strontium	39 Y Yttrium	40 Zr Zirconium	41 Nb Niobium	42 Mo Molybdenum	43 Tc Technetium	44 Ru Ruthenium	45 Rh Rhodium	46 Pd Palladium	47 Ag Silver	48 Cd Cadmium	49 In Indium	50 Sn Tin	51 Sb Antimony	52 Te Tellurium	53 I Iodine	54 Xe Xenon
55 Cs Cesium	56 Ba Barium	57–71 La–Lu Lanthanides	72	73	74	75	76	77	78	79 Au Gold	80 Hg Mercury	81 Tl Thallium	82 Pb Lead	83 Bi Bismuth	84 Po Polonium	85 At Astatine	86 Rn Radon
87 Fr Francium	88 Ra Radium	89–103 Ac–Lr Actinides	104 Rf Rutherfordium	105 Db Dubnium	106 Sg Seaborgium	107 Bh Bohrium	108 Hs Hassium	109 Mt Meitnerium	110 Ds Darmstadtium	111 Rg Roentgenium	112 Cn Copernicium	113 Nh Nihonium	114 Fl Flerovium	115 Mc Moscovium	116 Lv Livermorium	117 Ts Tennessine	118 Og Oganesson
		57 La Lanthanum	58 Ce Cerium	59 Pr Praseodymium	60 Nd Neodymium	61 Pm Promethium	62 Sm Samarium	63 Eu Europium	64 Gd Gadolinium	65 Tb Terbium	66 Dy Dysprosium	67 Ho Holmium	68 Er Erbium	69 Tm Thulium	70 Yb Ytterbium	71 Lu Lutetium	
		89 Ac Actinium	90 Th Thorium	91 Pa Protactinium	92 U Uranium	93 Np Neptunium	94 Pu Plutonium	95 Am Americium	96 Cm Curium	97 Bk Berkelium	98 Cf Californium	99 Es Einsteinium	100 Fm Fermium	101 Md Mendelevium	102 No Nobelium	103 Lr Lawrencium	

Superheavy elements: $Z \geq 104$

Actinides

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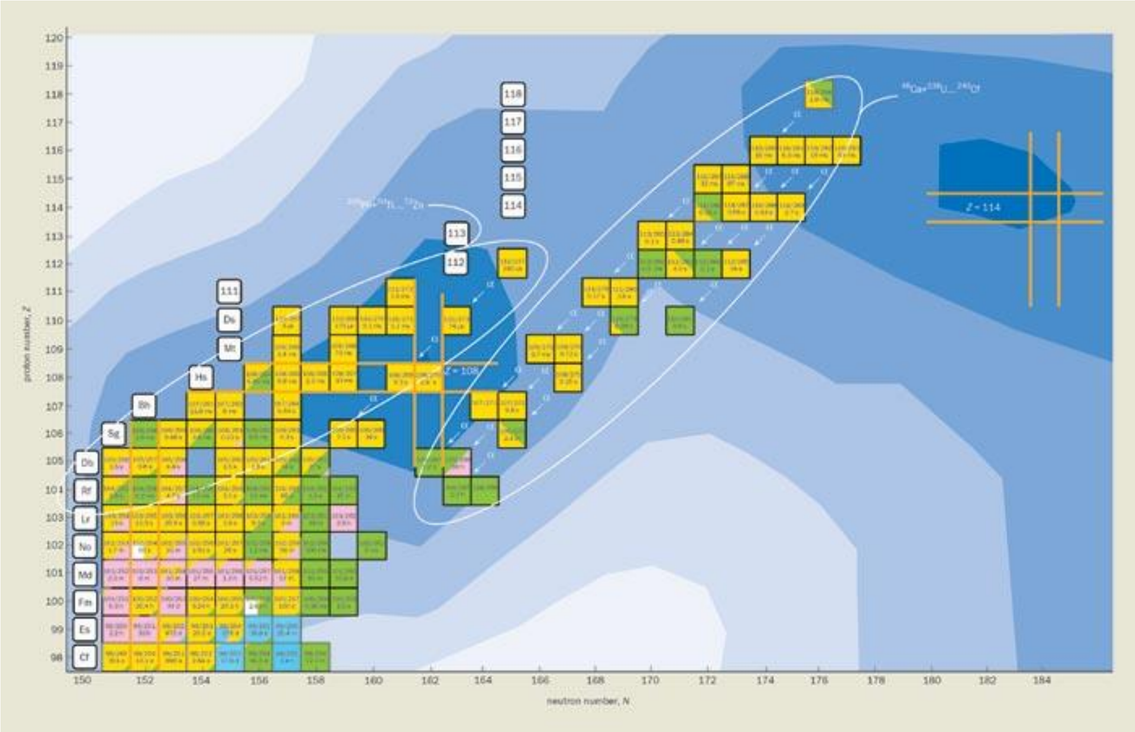
Producing superheavy elements



- Those can only be produced by ADDING matter together
  - Fusion(-evaporation) reactions
- Using stable beams on stable targets
  - Cold fusion, typically on ^{208}Pb targets, with e.g., ^{70}Zn beams
- Using stable beams on actinide targets
  - Hot fusion, e.g., with ^{48}Ca beams on a ^{252}Cf target

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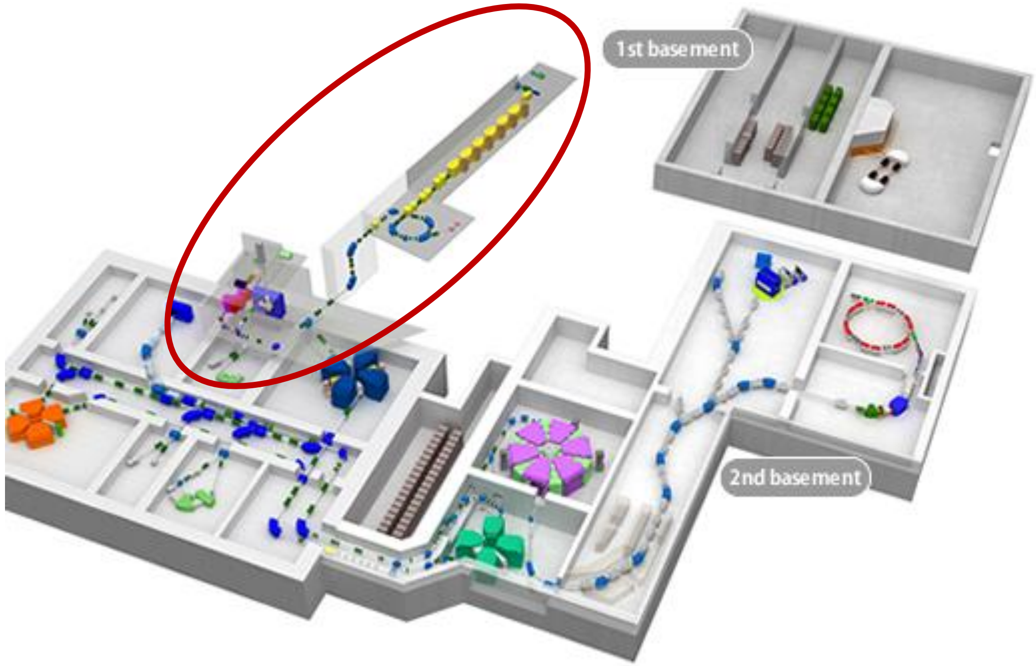
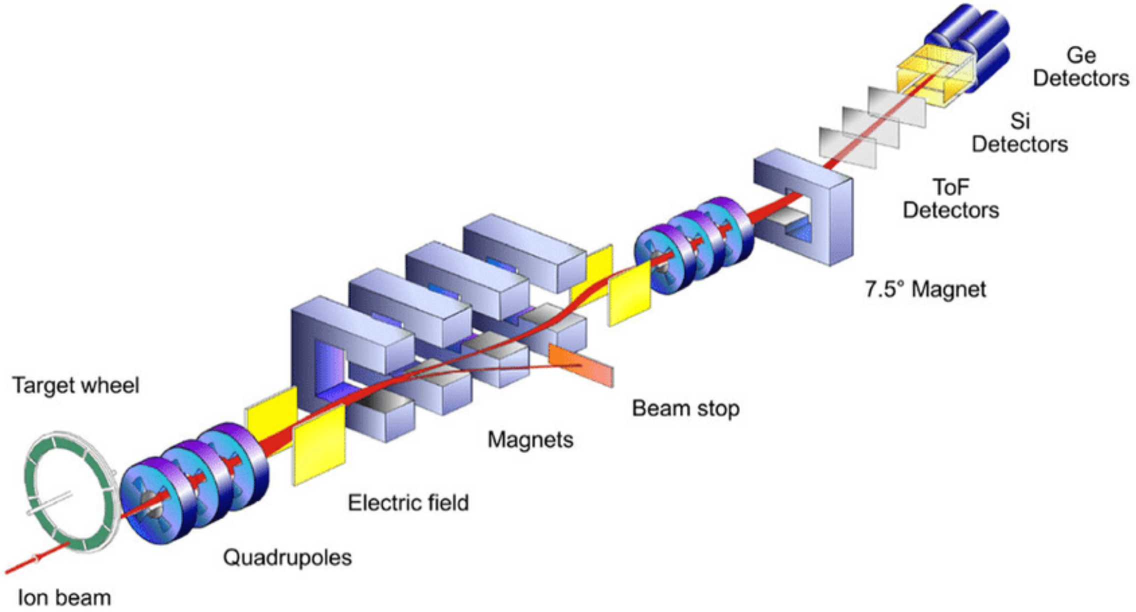
Many facilities around the world

Combining ^{48}Ca with actinide targets

RIKEN GARIS & GARIS-II



GSI SHIP



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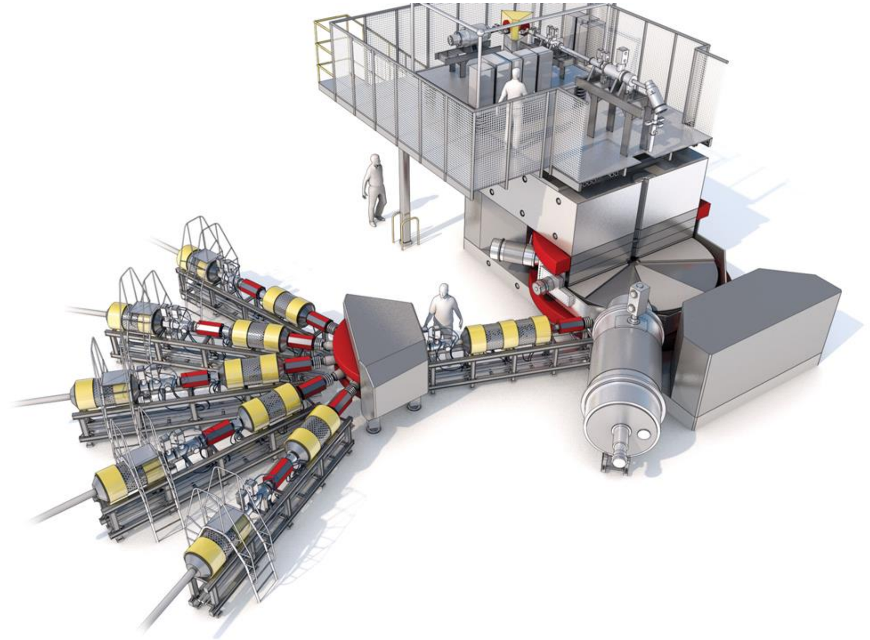
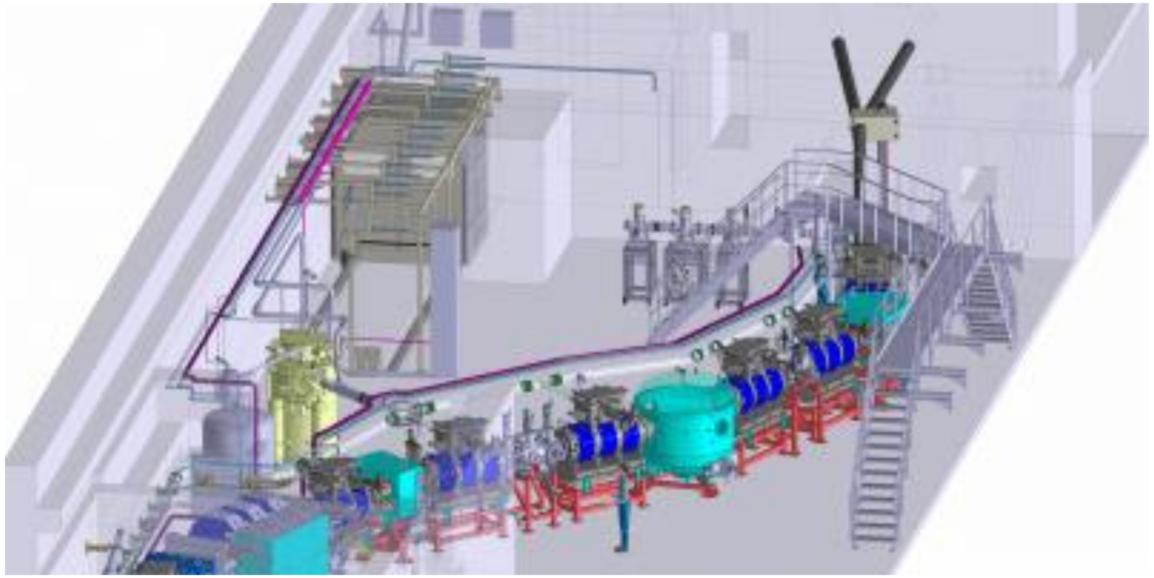
Many facilities upcoming around the world

Combining ^{48}Ca with actinide targets

Dubna SHEF



GANIL S^3



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- ✓ We have seen the different approaches to produce radioisotopes, first step towards radioactive beams.
- ✓ We can either add particles (neutron capture, fusion-evaporation, *multi-nucleon transfer*) or break things apart (fragmentation, spallation, fission).
- ✓ The production is a combination of the luminosity (beam and target content) and the reaction cross section. The half-life has also a substantial impact on what can be produced as well on the activity.

- ➢ How that is used for the production of radioisotopes for applications
- ➢ How ^{100}Sn has been produced but also what to look forward to
- ➢ The quest for superheavy elements

Recoil separators

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To be continued in the next lecture series

✓ 25 Oct 2021

1 – Basics of RIB production

- Nuclear reactions for the production of radioisotopes
- Luminosity & cross sections
- Examples across the nuclear landscape
  - Industrial production
  - Towards ^{100}Sn
  - Superheavy elements

8 Nov 2021

2 – State of the art in ISOL

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

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