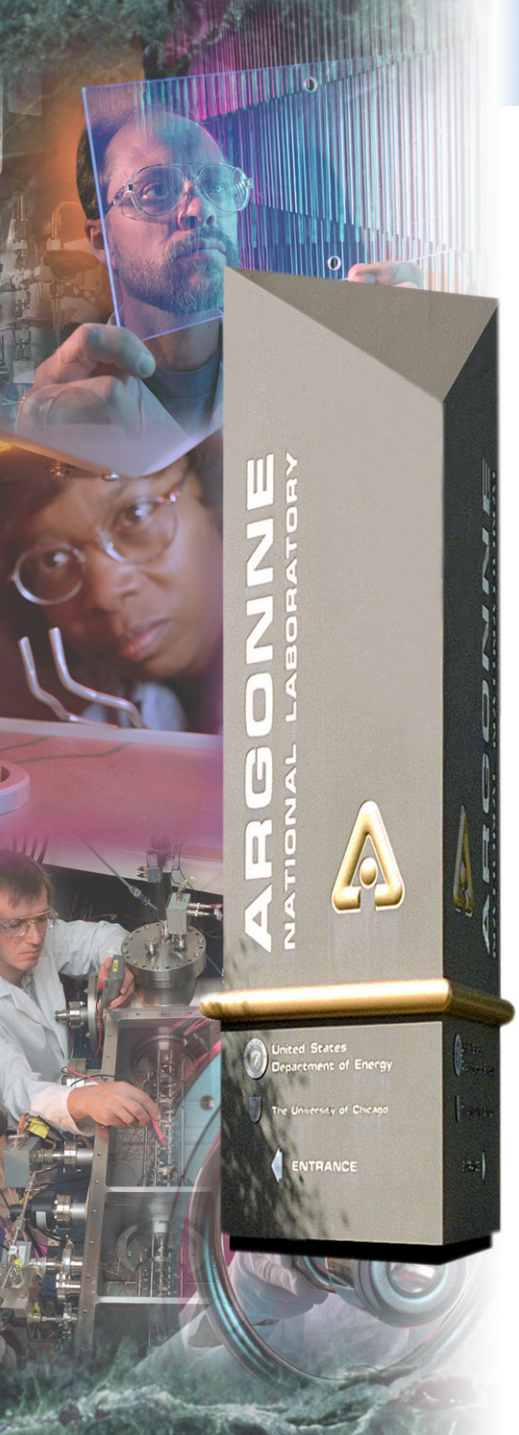


Isotope harvesting at high power radioactive beam facilities

Jerry Nolen

Physics Division

Argonne National Laboratory

[Helge Ravn, CERN/ISOLDE]

2nd High-Power Targetry Workshop

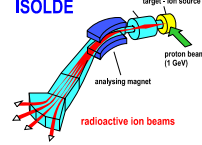
Oak Ridge National Laboratory

October 10-14, 2005

Argonne National Laboratory is managed by
The University of Chicago for the U.S. Department of Energy

Introduction

- General overview of isotope separation methods and potential for practical applications of isotope harvesting at high power RIB facilities
- Presentation based mainly on contributions of Helge Ravn
- Ideas are generally applicable for parasitic operation at future facilities such as RIA and EURISOL

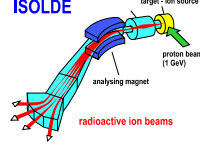


The role of isotope separation in future medical isotope production programs

H. L. Ravn/CERN, PH

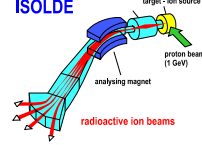
http://cern.ch/Helge.Ravn/files/The Role of Isotope Sep.ppt /

Overview



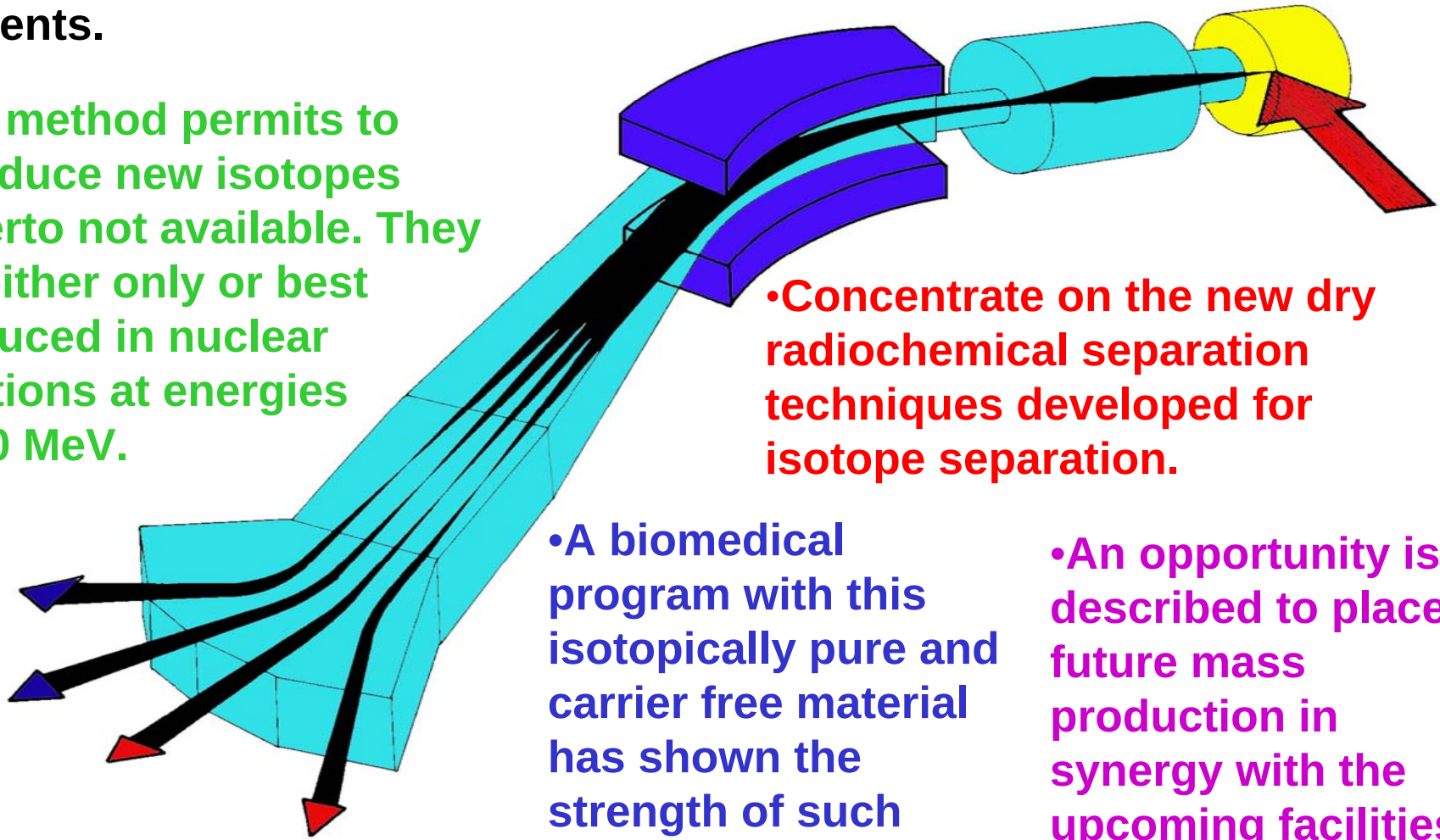
- The mass separation in the Radioactive Ion beam Facilities (RIB)
- A biomedical program with carrier free radionuclides has shown the strength of such high quality preparations.
- Concentrate on the new dry radiochemical separation techniques using isotope separation
- The nuclear production reactions at energies > 100 MeV
- Ion sources for efficient and selective ionization
- Efficiency of the mass separation process
- Describe the new production methods that may be derived from the ISOL target and ion-source techniques
- The method makes almost all existing radionuclides available and allows to introduce new ones in medicine
- The opportunity for parasitic medical isotope production in collaboration with future high power accelerator driven projects
- Conclusion and outlook
- Acknowledgement and links

Overview



•**Use of the mass separation methods from Radioactive Ion beam Facilities (RIB) allows to produce carrier free radioisotopes of all elements.**

•**The method permits to introduce new isotopes hitherto not available. They are either only or best produced in nuclear reactions at energies > 100 MeV.**

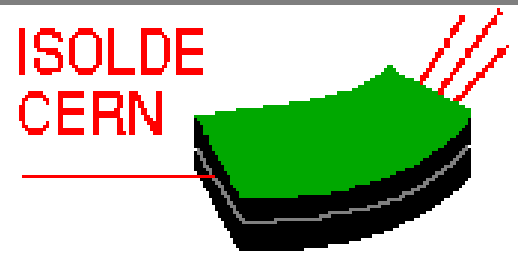


•**Concentrate on the new dry radiochemical separation techniques developed for isotope separation.**

•**A biomedical program with this isotopically pure and carrier free material has shown the strength of such preparations.**

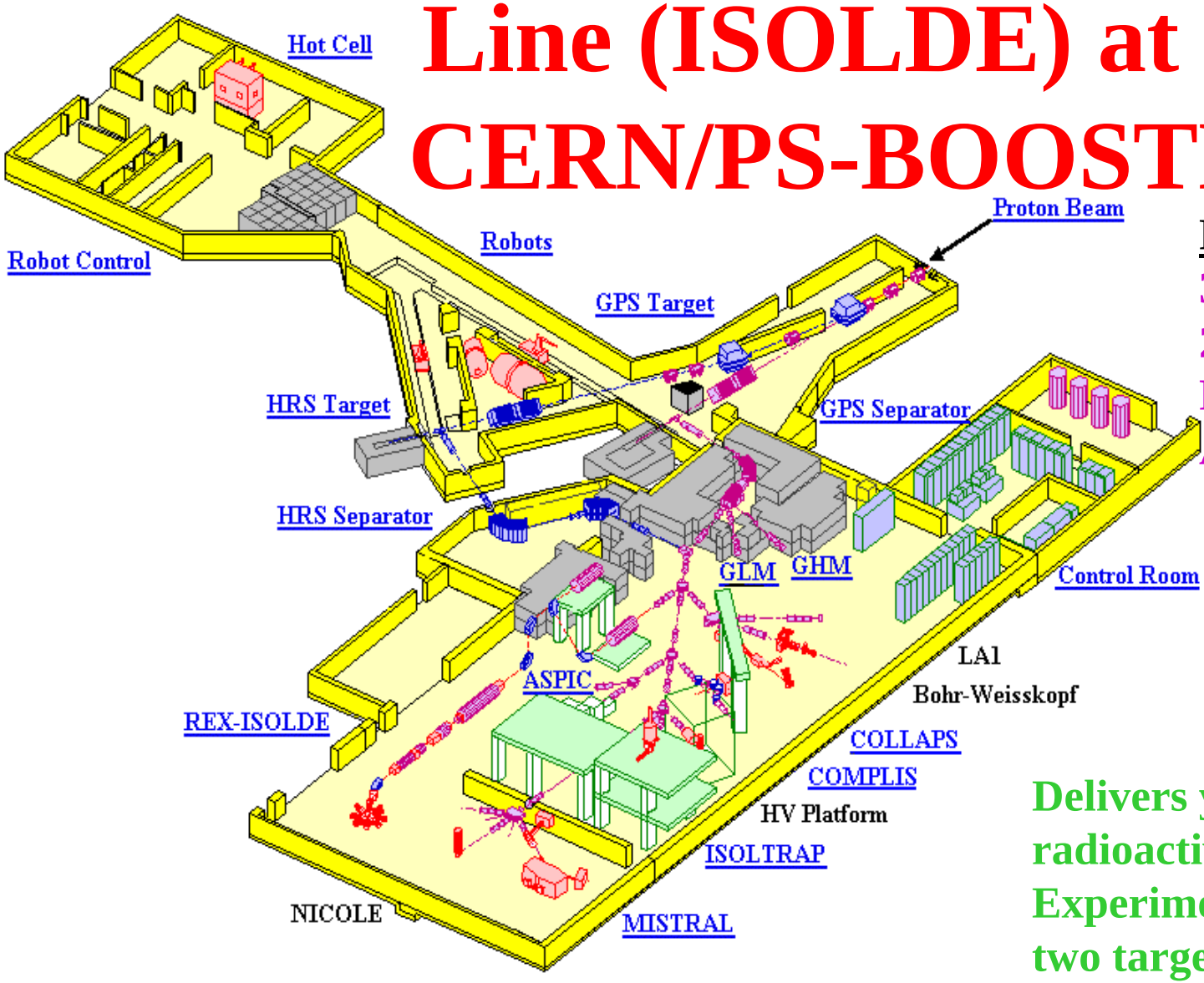
•**An opportunity is described to place future mass production in synergy with the upcoming facilities using MW targets.**

- Ionisation to q = 1+
- Acceleration to 60 keV
- Mass selection by magnetic deflection
- $B\rho = p/q \propto \sqrt{A}$



Z selection by chemically selective step

The Isotope Separator On-Line (ISOLDE) at the CERN/PS-BOOSTER



Proton beam:

3E13 per pulse

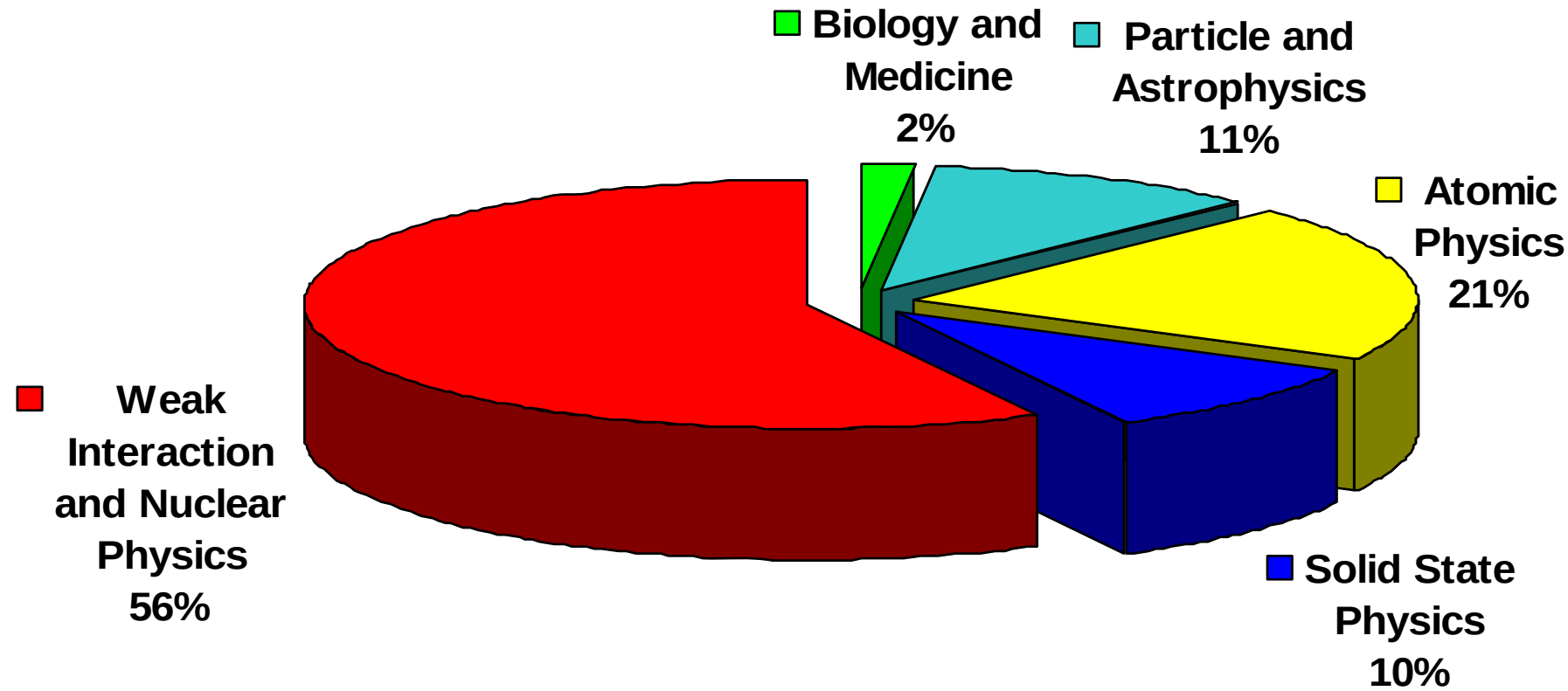
2.4 μs pulse length

Rep. Rate 0.5 Hz

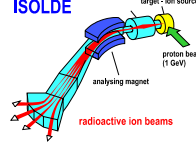
Av. current 2 μA

Delivers yearly 3200 h of radioactive ion-beam to 30 Experiments by means of two target stations

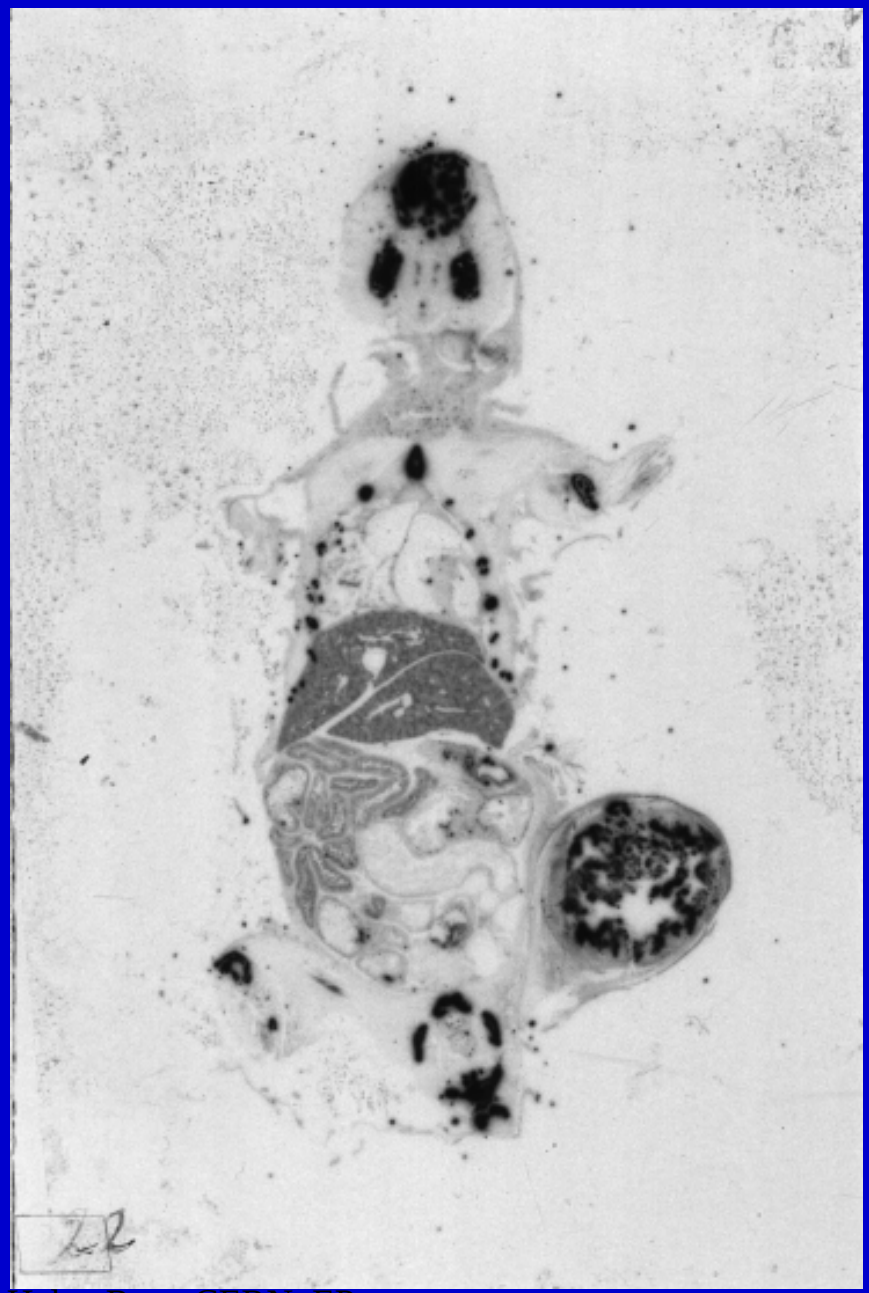
ISOLDE 2004: 2560 hours radioactive ion beam



Biomedical program at at the CERN ISOLDE facility



- Almost all nuclides of the nuclidic chart available in one laboratory
- Unprecendented purity chemical (carrierfree) since mass separated
- 30 years experiences in complex bio-medical studies using ISOLDE produced radionuclides
- Implementation of isotope separation technique into the medical isotope programs.
- First patient studies 1978 MED Dresden planar scintigraphy of lymphoma patients using ^{167}Tm Citrate
- Over more than 2 decades collected data set on the bio-kinetik behaviour of radio-lanthanides in tumor bearing animals
- Lanthanides are used today in tumor seeking radiotracers
- Exotic positron emitters are available
- First in-vivo study with the partially alpha emitting ^{149}Tb
- Already today patient doses available
- Materials: Collections in the on-line mass separator, Waste – recycling, radiochemically processed, partially at CERN, later in Rossendorf and Geneva Hospital



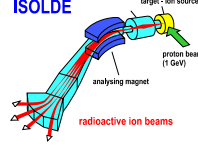
Autoradiogram

of a whole body sagittal slice of a tumor bearing mouse 24 hours after injection of 0.4 MBq of ^{167}Tm-Citrate

Lanthanides are unspecific tumor seeking tracers

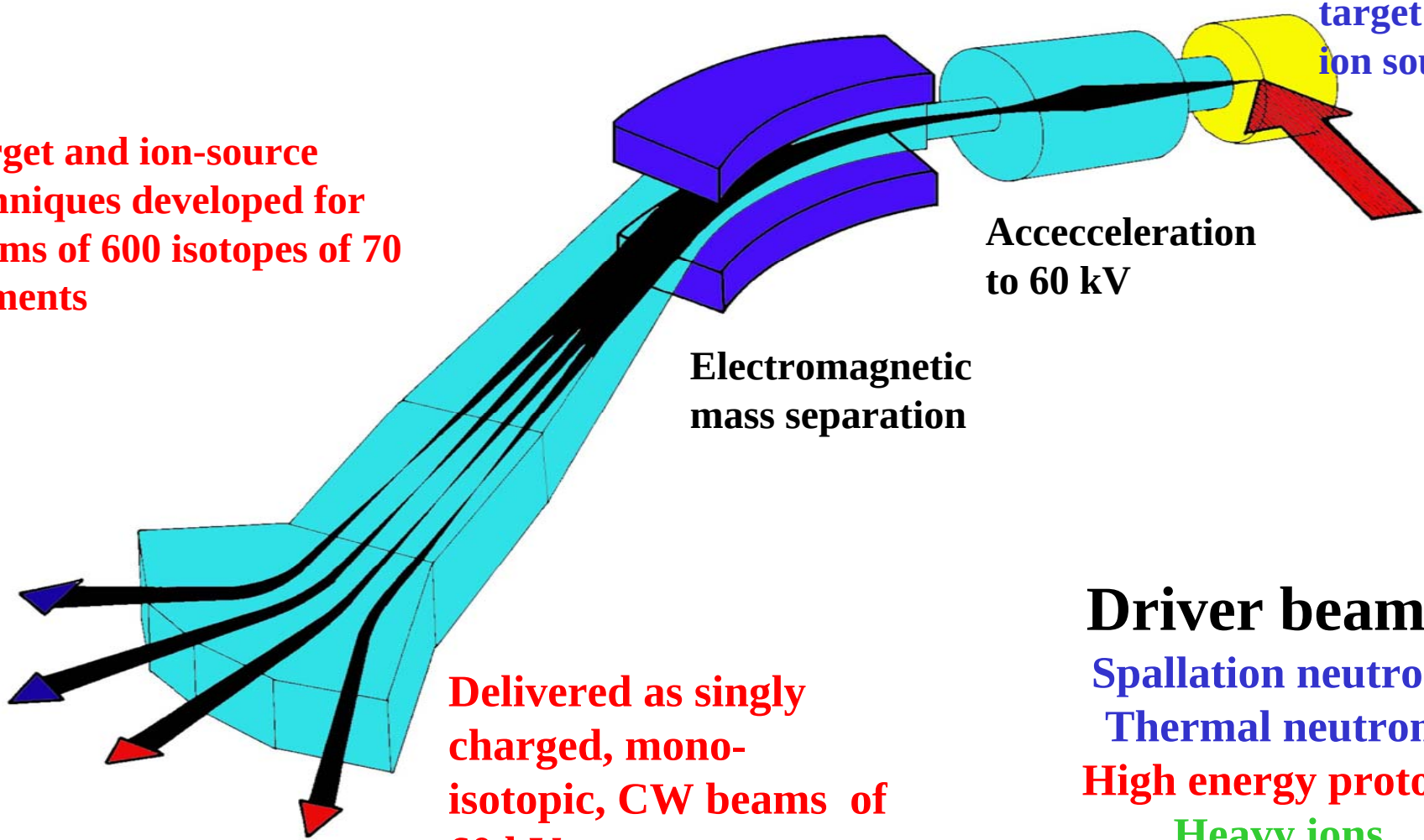
G.J.Beyer, R.Münze et al., in: "Medical Radionuclide Imaging 1980" IAEA Vienna (1981) Vol.1 p. 587

The Isotope Separator On-Line (ISOL)



Integrated target and ion source

Target and ion-source techniques developed for beams of 600 isotopes of 70 elements



Acceceleration to 60 kV

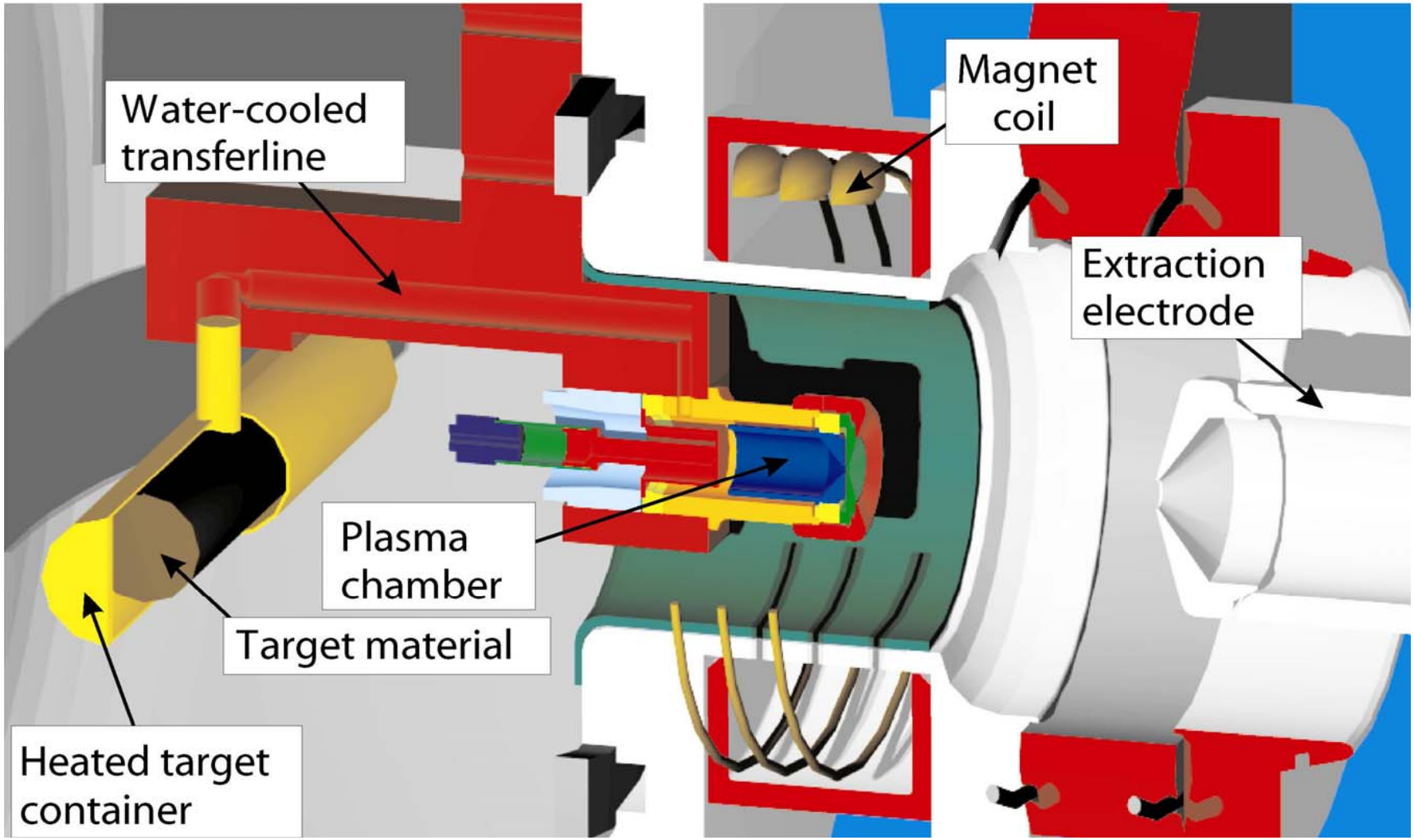
Electromagnetic mass separation

Delivered as singly charged, mono-isotopic, CW beams of 60 kV energy

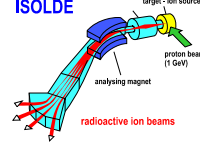
Driver beams:

Spallation neutrons
Thermal neutrons
High energy protons
Heavy ions

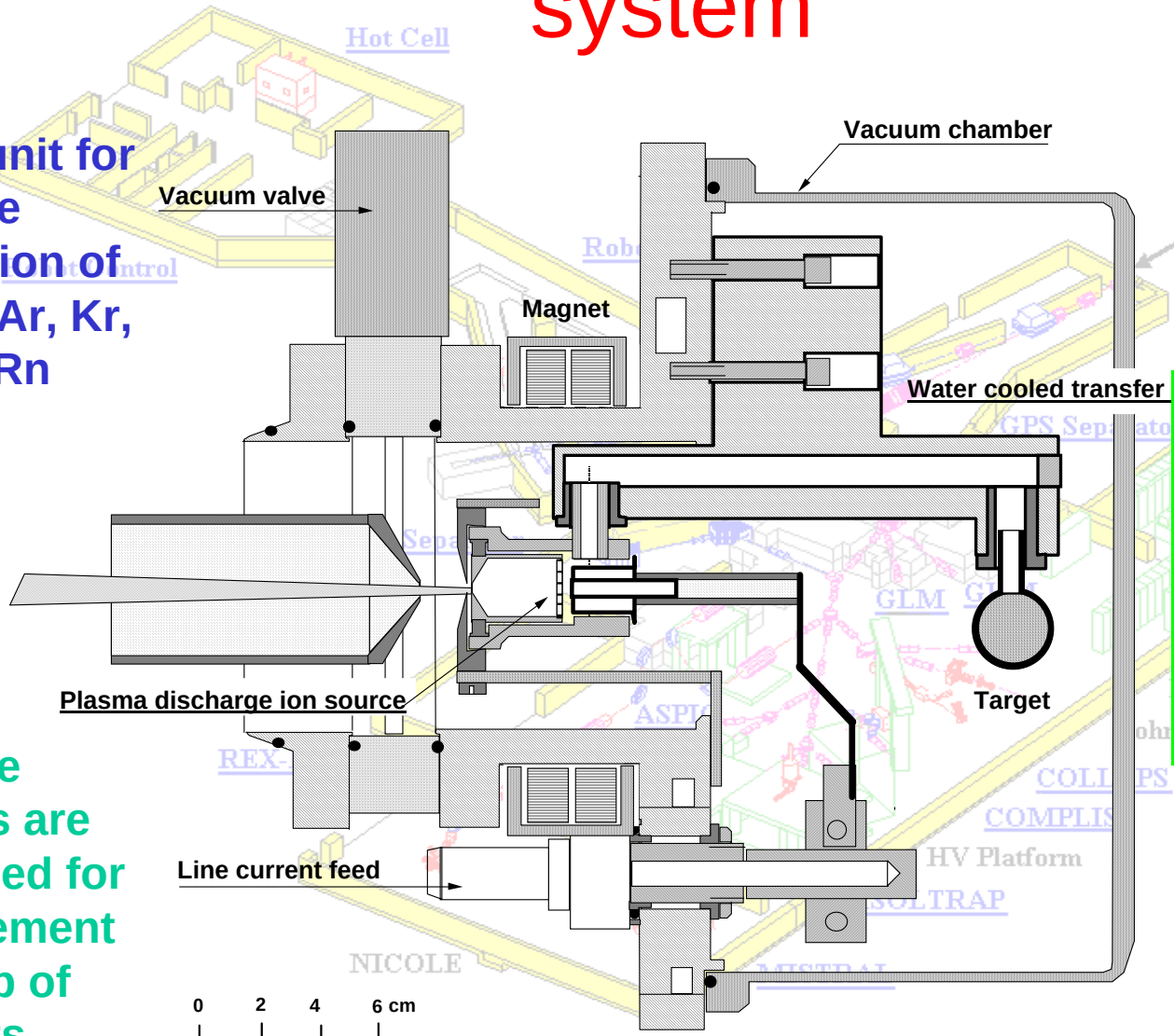
An ISOLDE target and ion source unit



The ISOLDE target and ion-source system



Target unit for selective production of He, Ne, Ar, Kr, Xe and Rn beams

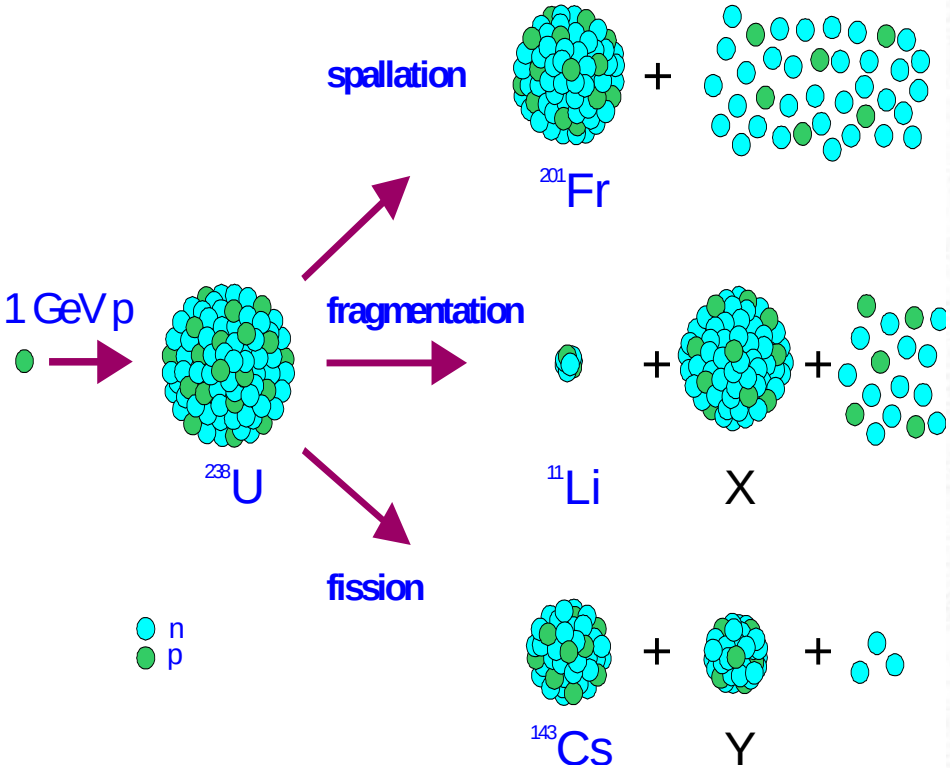


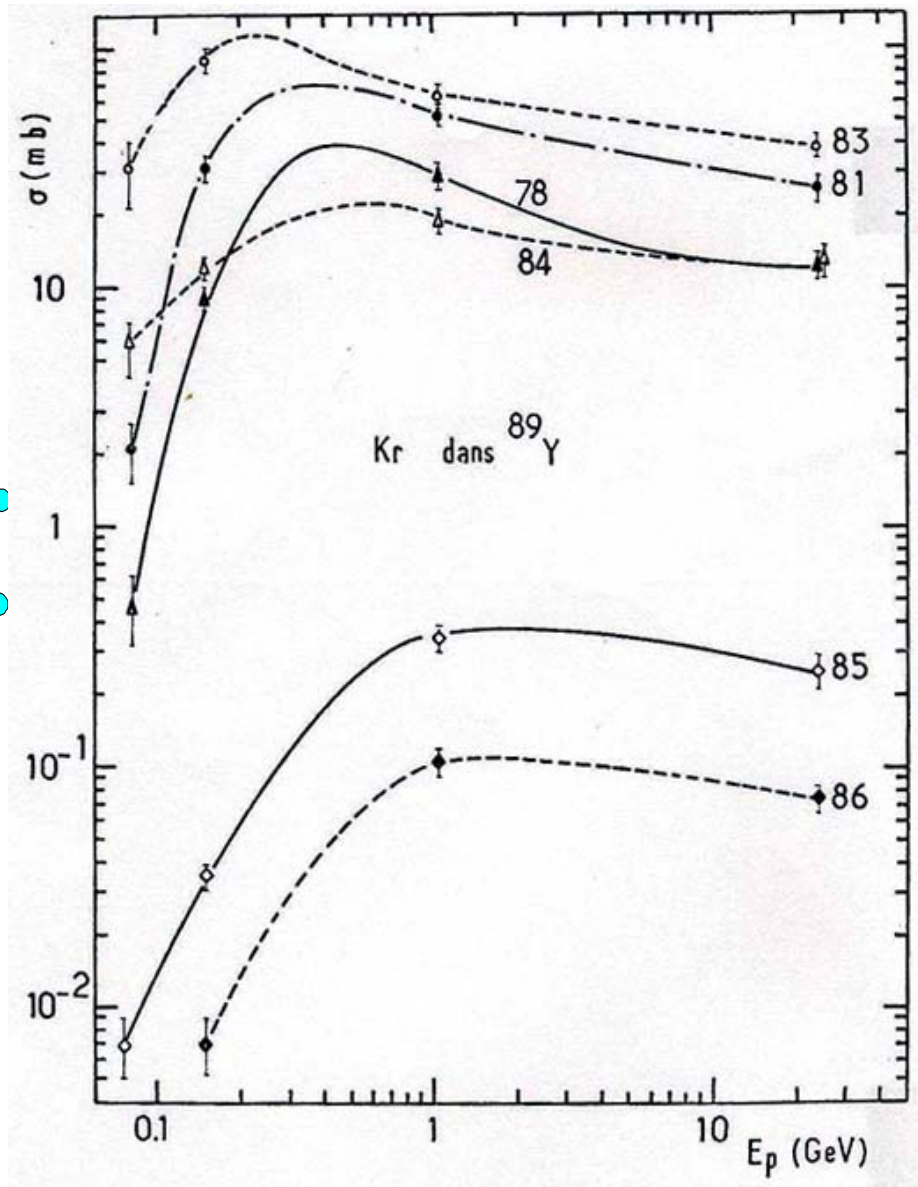
Lifetime about one month due to optimisation for short-lived nuclei

Long-lived reusable targets may be developed for production of long lived nuclei only

Separate systems are developed for each element or group of elements

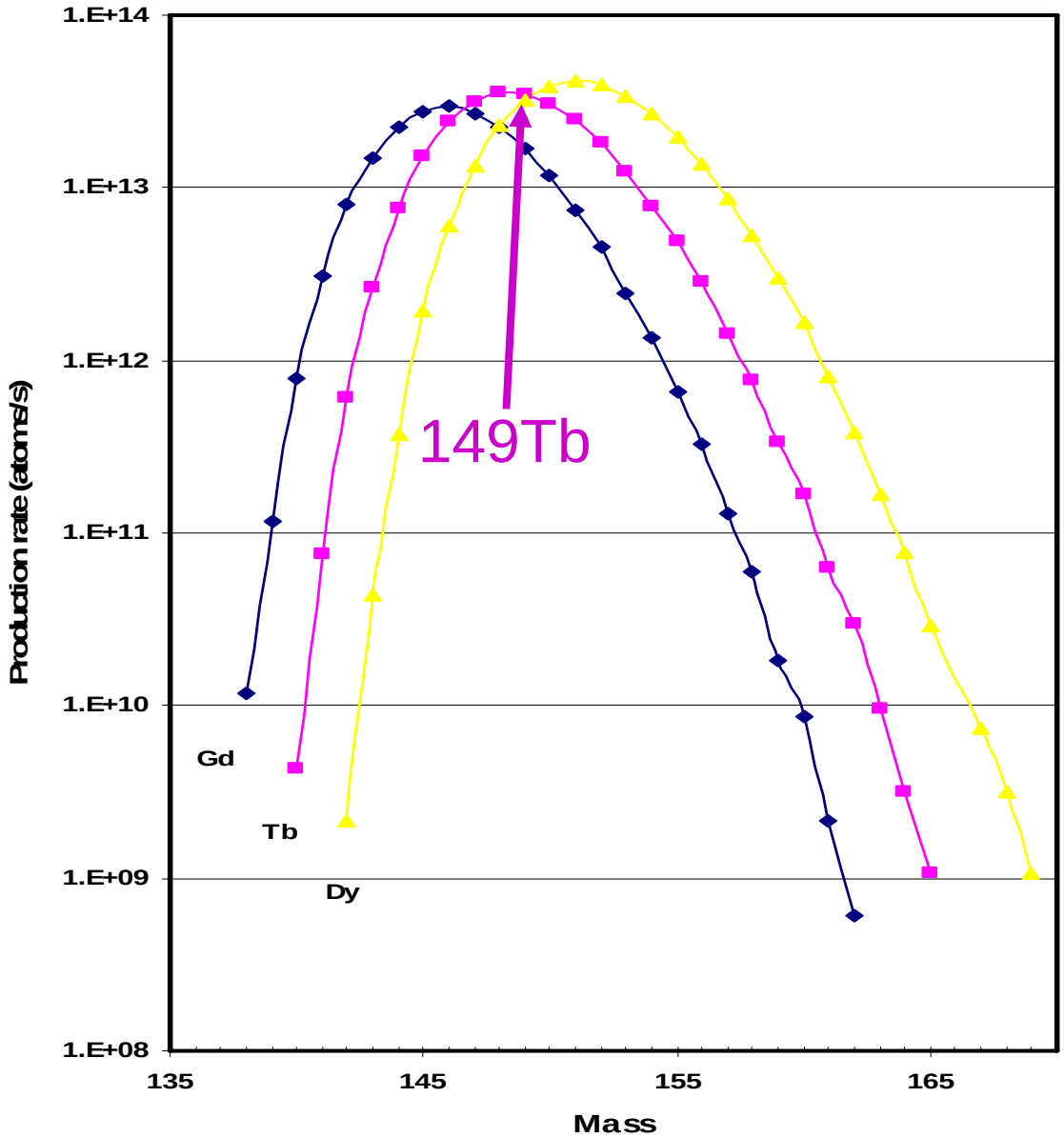
Reaction mechanisms





The need for selectivity

Calculated production rate of some rare earths in a Hg target irradiated with a 5mA , 1 GeV proton beam.



Production rate in the target

$$A = \Phi \ \sigma \ N$$

σ ***REACTION CROSS SECTIONS***

Tens of milibarns

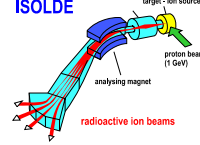
N ***TARGET THICKNESS***

Very thick targets mol/cm^2

Φ ***DRIVER BEAM INTENSITY***

Driver beam intensity presently 1 to 100 μA

$$A = 10^{13} \text{ atoms/s}$$



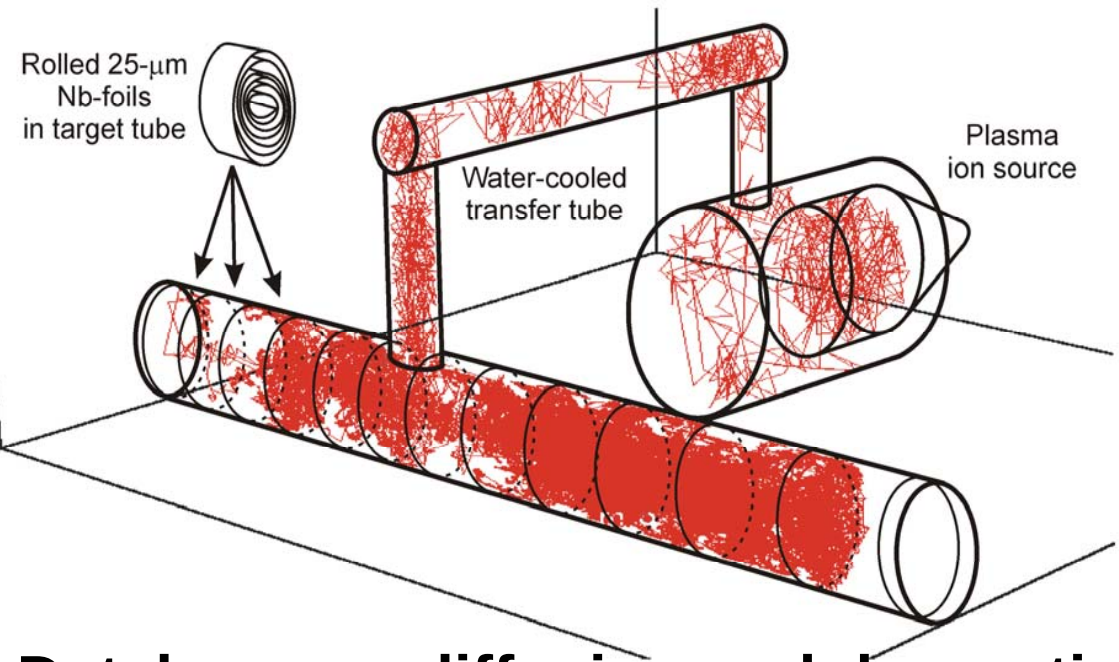
Mass transport efficiency

- $A = \Phi \sigma N \varepsilon_1 \varepsilon_2 \varepsilon_3$

- **How to get the products out and transferred into an ion beam for separation and acceleration.**
- **Decay losses due to diffusion and effusion from the target to ion source ε_1 ε_2 = 100% for nuclei with $t_{1/2}$>1h**
- **Ionisation efficiency ε_3 = 10-25% for long-lived nuclei**

- **$A = 10^{12}$ atoms/s**

Release models



C.J. Densham et al., Nucl. Instr. Meth. B126 (1997) 154.

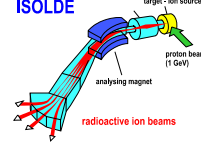
B. Mustapha and J.A. Nolen, Nucl. Instr. Meth. B204 (2003) 286.

Mario Santana-Leitner, PhD, 2005 (http://www.ganil.fr/eurisol/)

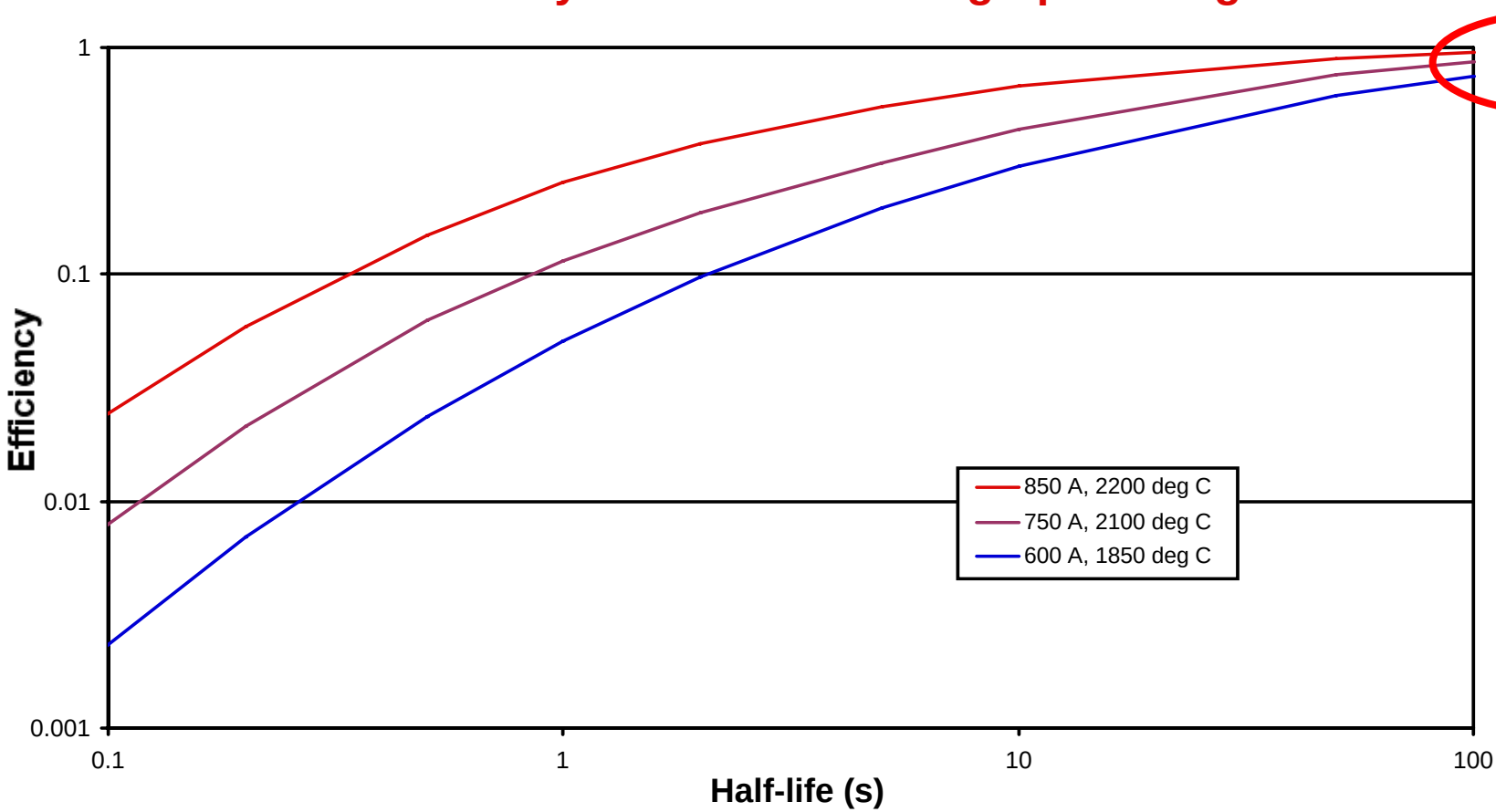
Database on diffusion and desorption properties is maintained and further studied made in the EU FP 5 program TARGISOL

http://www.targisol.csic.es/

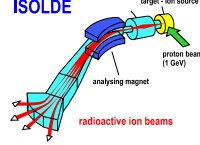
Release efficiency ε_1 ε_2 determined by the decay losses



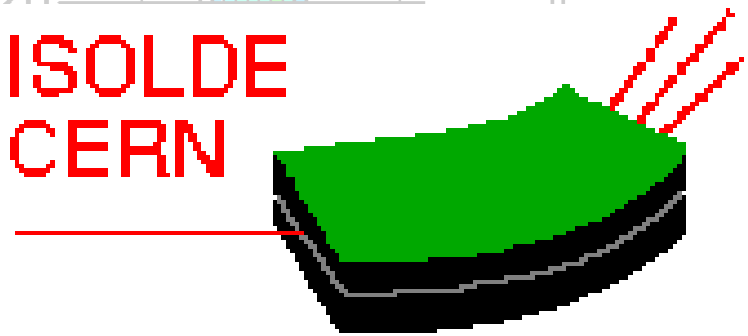
Release efficiency of tin from a UCx/graphite target



Isotope harvesting: general considerations



1. **High energy reactions provide a wide spectrum of reaction products**
   ⇒ Universality !
2. **Mass separated beams are intrinsically carrier-free !**
3. **Simultaneous extraction of several isotopes !**
4. **Ion implantation facilitates the molecule labelling !**
5. **Ion source unit and target materials reusable many times !**
6. **No liquid waste !**
7. **Production presently only allows to supply research quantities !**
8. **Future industrial production sites for therapeutical amounts ?**

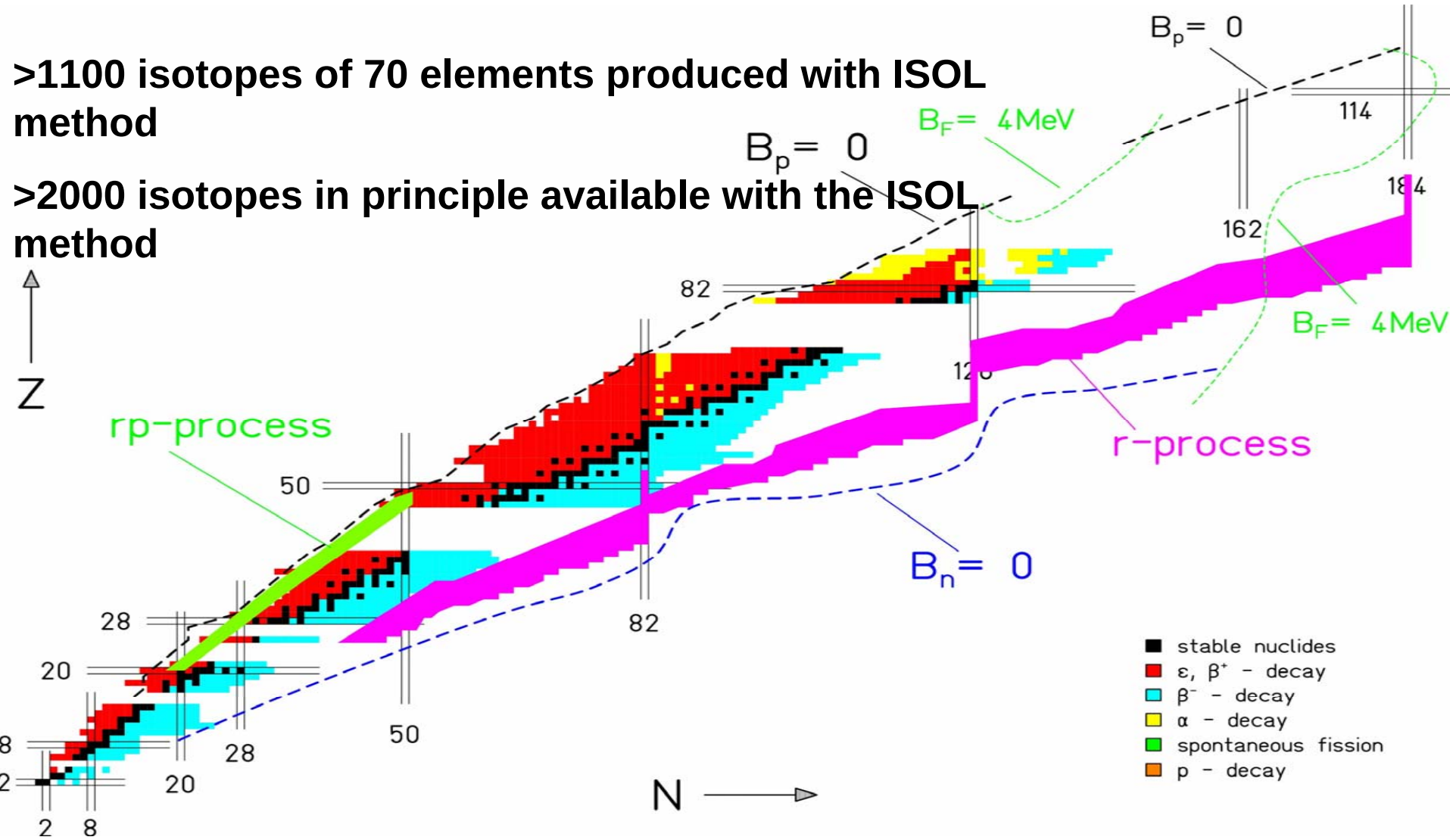


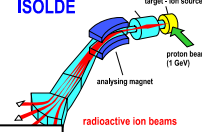
EURISOL

Nuclear chart at ISOLDE

>1100 isotopes of 70 elements produced with ISOL method

>2000 isotopes in principle available with the ISOL method





Existing ISOL beams

Isotopes with $T_{1/2} < 0.1$ s separated

Isotopes with $T_{1/2} < 10$ s separated

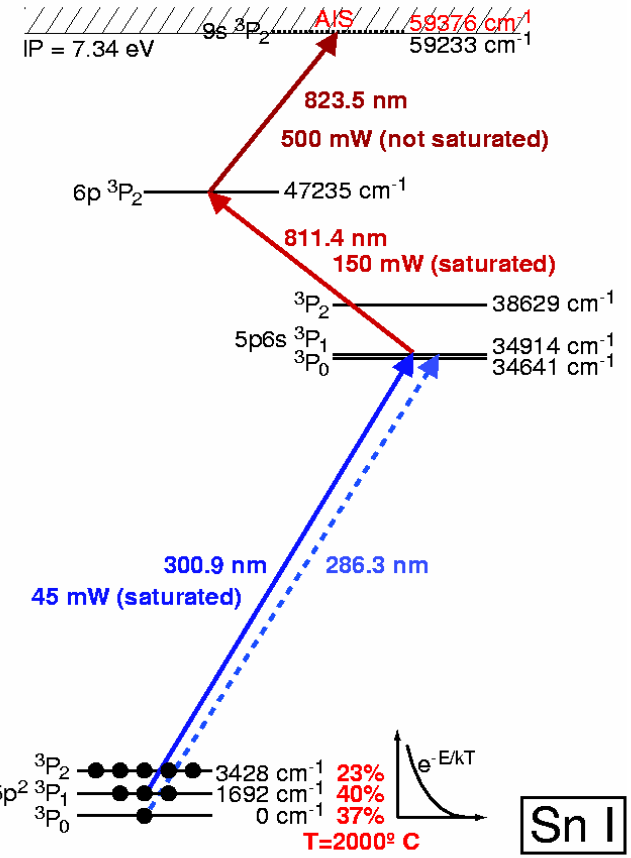
Isotopes with $T_{1/2} > 10$ s separated

1 H																2 He
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg											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						

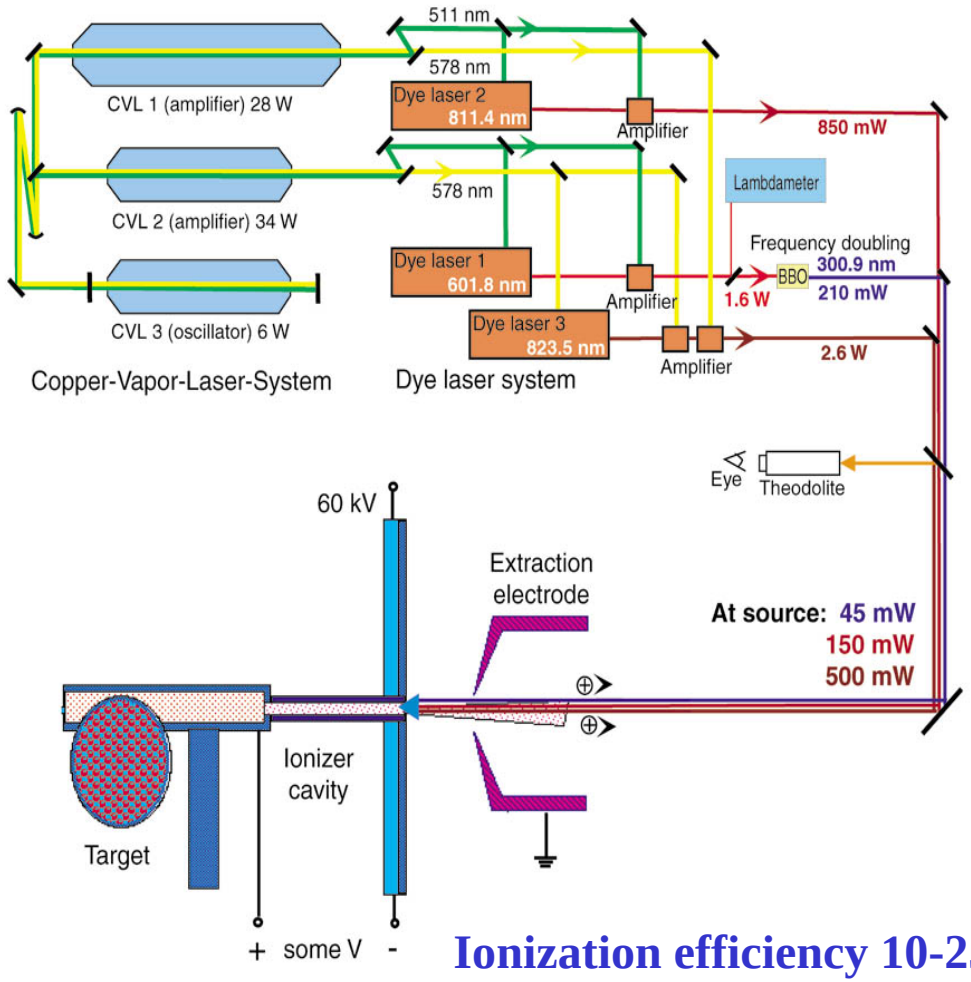
58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr

Stepwise resonant laser ionization of Tin

Ionization scheme for Tin

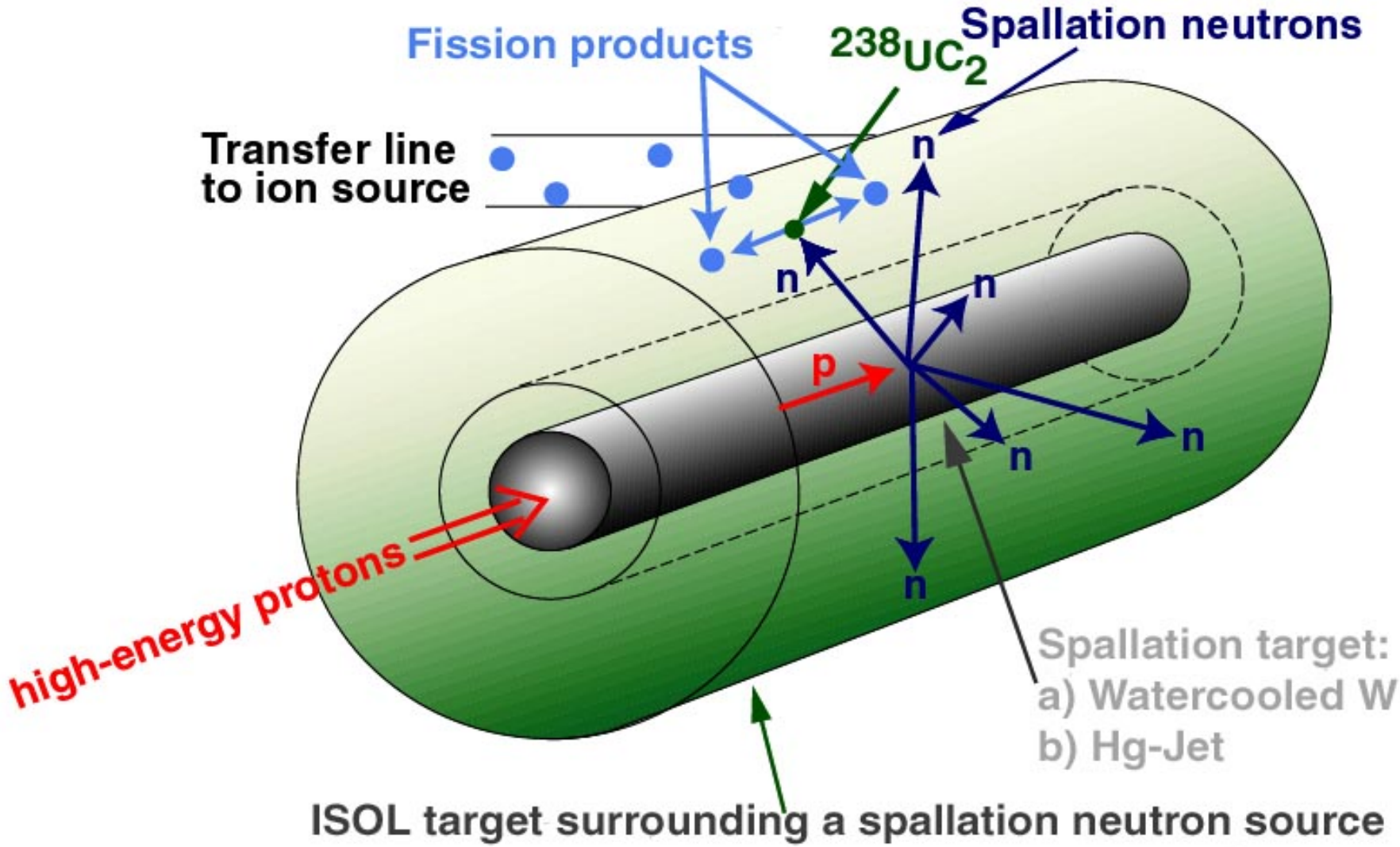


Three fine structure components of the ground state are thermally populated, but only one can be excited at a time. A second UV laser (dotted line) could roughly double the efficiency.

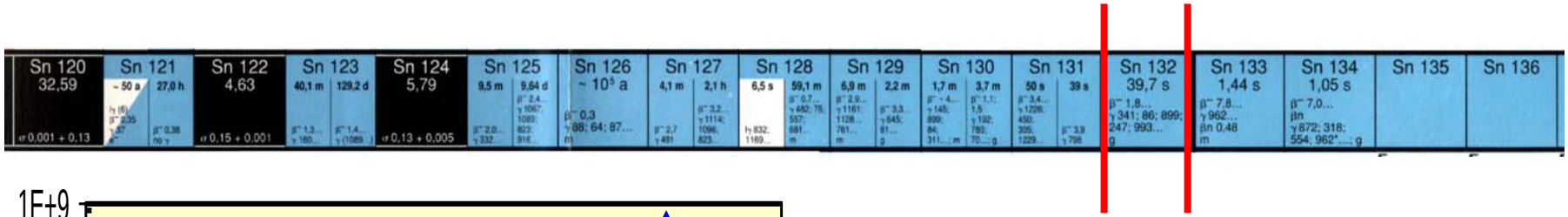


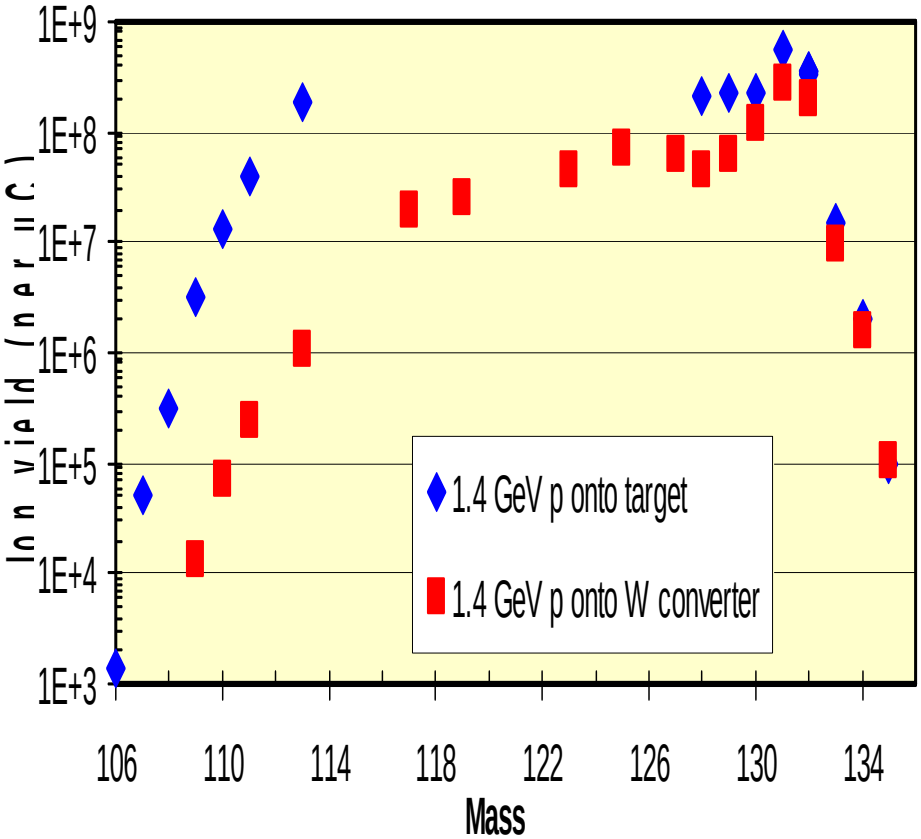
Ionization efficiency 10-25%

1 MW target for 10^{15} fissions/s



Sn yields from a UC target





Today at ISOLDE:

$^{132}Sn^{+}$ intensity: **5.0E8 per s**

with 2.5 μA of 1.4 GeV protons

onto 12.7 mm W converter

(release efficiency about 80%)

EURISOL:

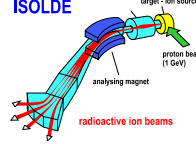
1.0 GeV protons instead of 1.4 GeV	**0.7**
1 mA protons	**400**
cylindrical target	**10**
RILIS improvement	**5**
Total	**14000**

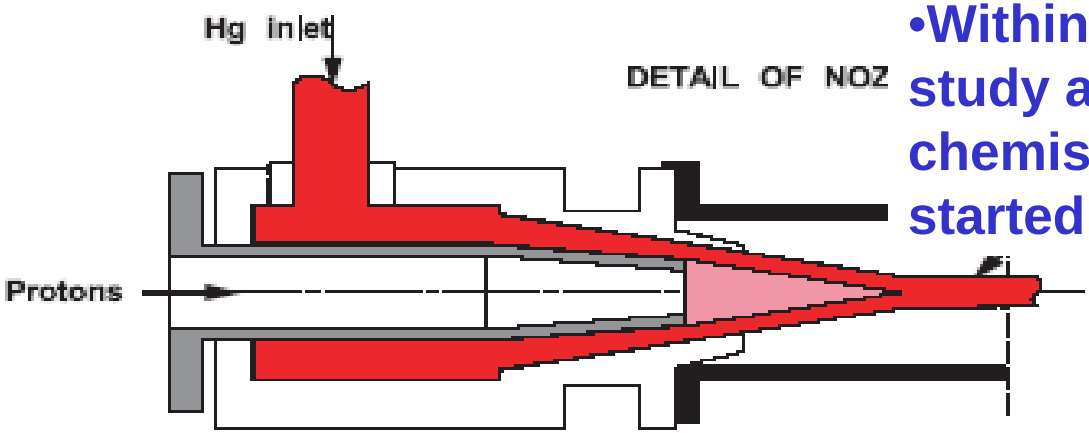
Expected intensity:

7E12 per s = 1 μA $^{132}Sn^{+}$

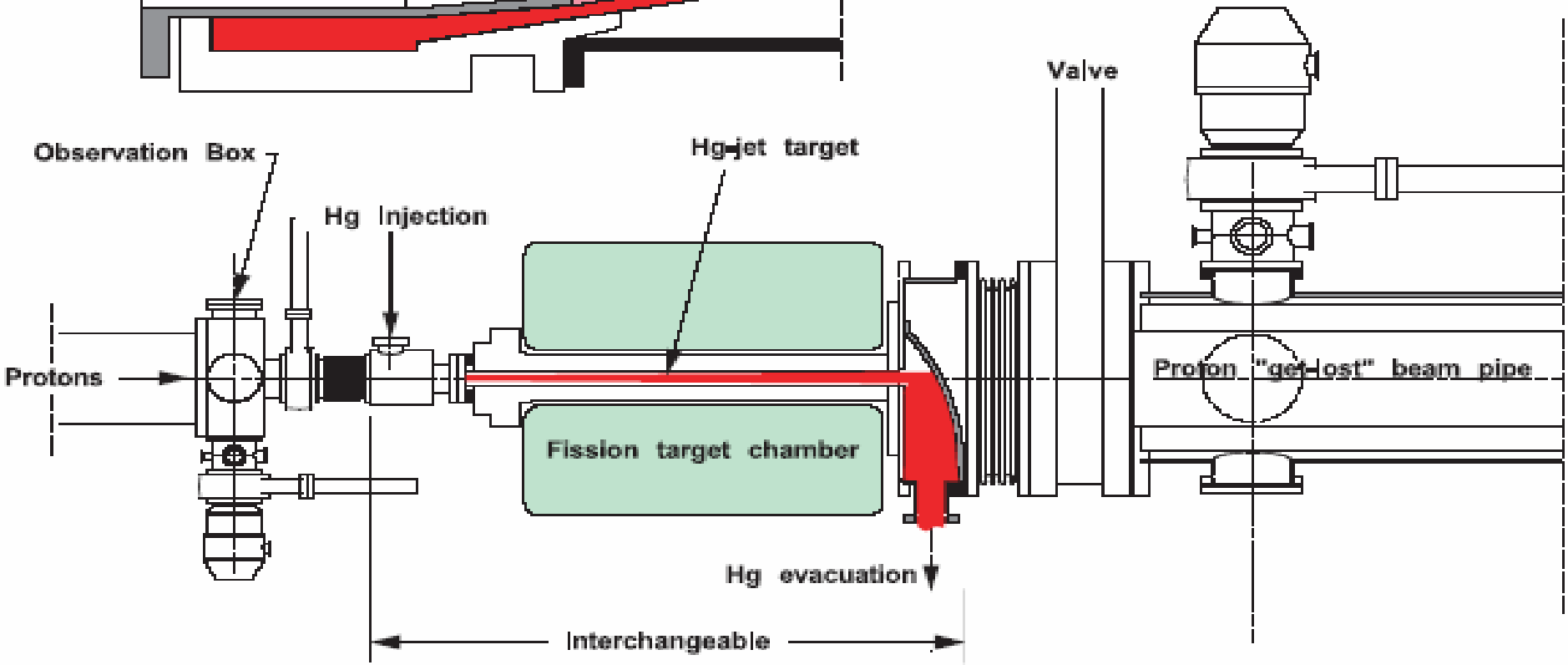
Ref. Ulli Koester

Hg-jet p-converter target





- **Within the EURISOL-EU, FP6 design study a program of reaction product chemistry in a Hg flow is being started.**



Synergies with targets for SNS, Superbeam, ν-factory, μ-collider,...

EURISOL

1-2.2 GeV, multi-MW proton driver (CERN SPL)

Several direct target stations (ca. 100 kW)

One Hg spallation + fission target station (>1 MW, i.e. 1E15 fissions/s)

Multiple user operation in parallel

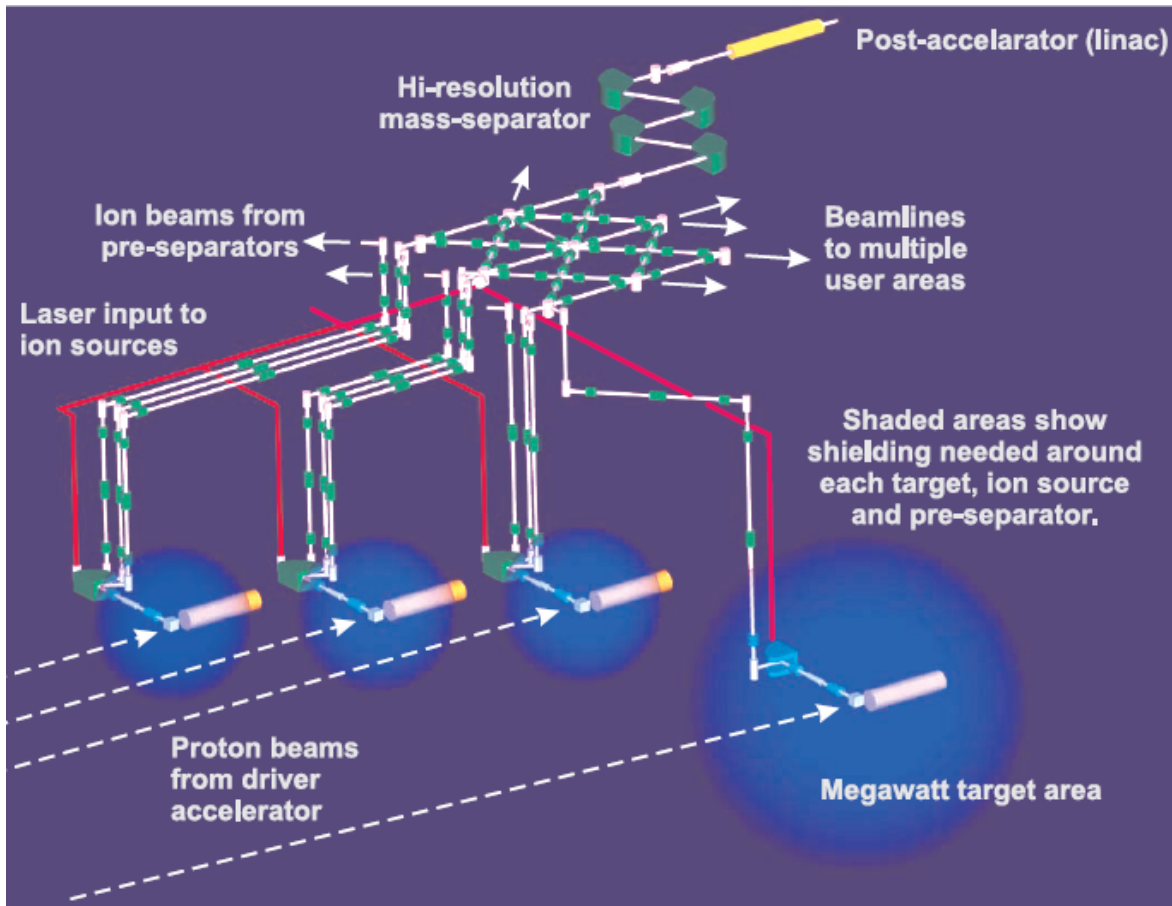
Low-energy beam area

Post-acceleration with
LINAC up to ca. 10 A.MeV

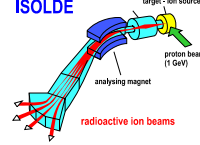
Post-acceleration to
ca. 100 A.MeV
with LINAC or cyclotron

Fragmentation of
post-accelerated RIBs

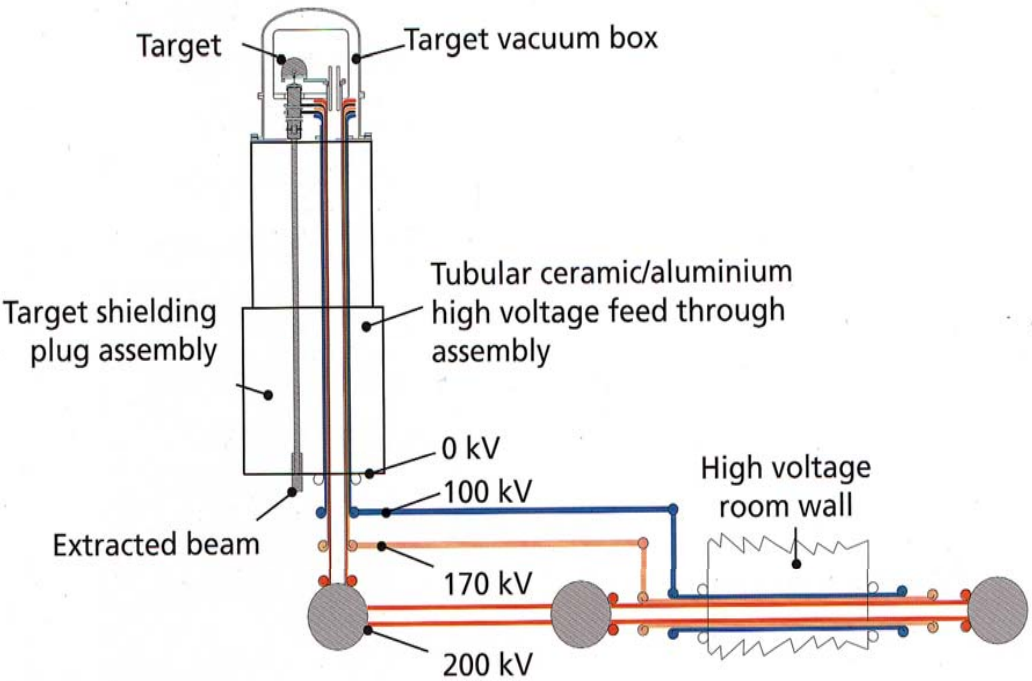
Commissioning: 2015?

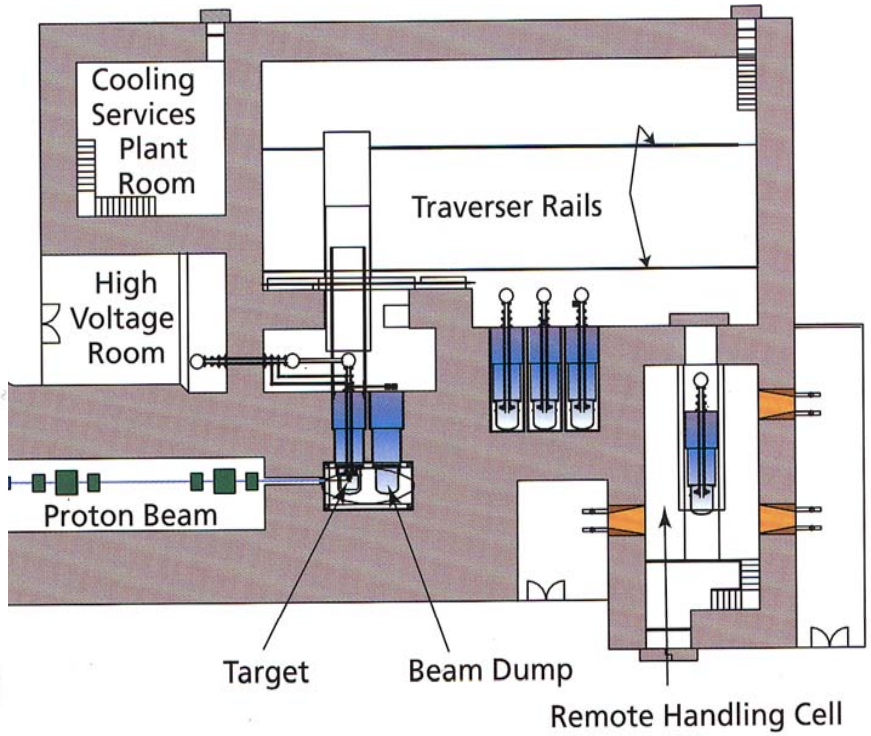


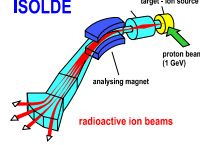
EURISOL target layout



Very similar to the target stations of the ISOL facility ISAC in operation at TRIUMF



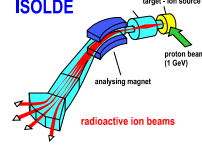




Production rates in the EURISOL Mercury target

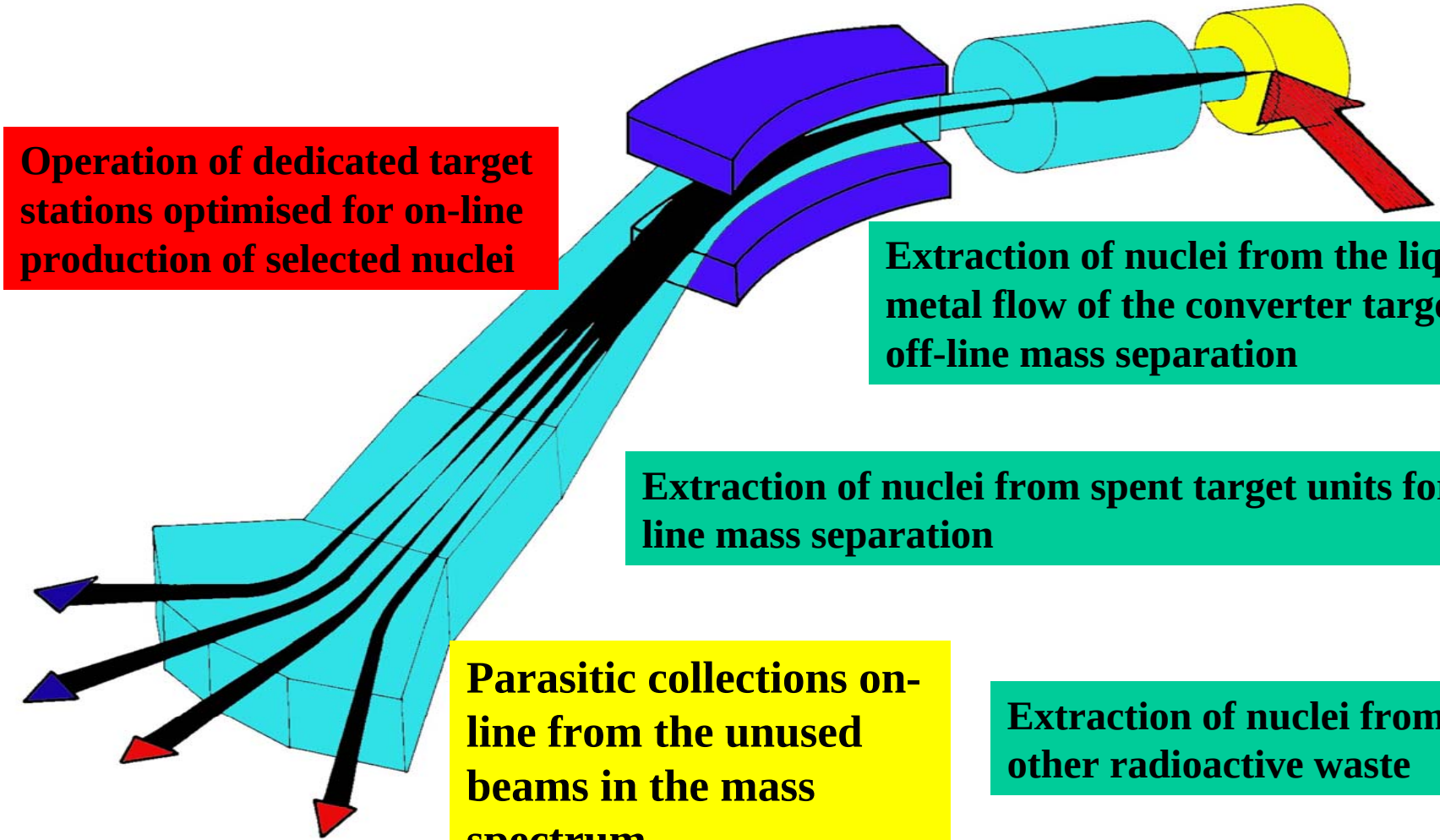
Radio-isotope	Half-life $T_{1/2}$	X-section (mb)	Production rate (per s)	Alternative production processes		Applications
192-Ir	74 d	2.58E+00	**1.0E+14**	(n,γ)	reactor	Sealed sources for industry and cancer therapy
188-W/Re	69 d	6.90E-02	**2.7E+12**	(2n,γ)	HFR	Radio-immuno-therapy with 188-Re
178-W/Ta	22 d	8.08E+00	**3.1E+14**	(p,4n)	accelerator	Generator with potential in PET
177-Lu	6.7 d	6.31E-02	**2.4E+12**	(n,γ)	reactor	Therapy with labelled antibodies and peptides
166-Ho	25.8 h	5.30E-03	**2.0E+11**	(n,γ)	reactor	Therapy with labelled antibodies and peptides
149-Tb	4.12 h	9.21E-01	**3.5E+13**			Targeted Alpha Therapy, single cancer cell targeting
148-Gd	74.6a	5.31E-01	**2.1E+13**	spallation	accelerator	Low-energy alpha sources
153-Sm	46.75 h	1.41E-03	**0.6E+11**	(n,γ)	reactor	Therapy of bone metastases
127-Xe	76.4 d	9.22E-02	**3.5E+12**	(p,x...)	accelerator	SPECT, lung ventilation and brain perfusion
117m-Sn	13.6 d	1.78E-01	**0.7E+13**	(n,γ)	HFR	Systemic radionuclide therapy
99-Mo/99m-Tc	66 h	2.78E-01	**0.6E+13**	(n, f)	reactor	Most important radionuclide for nuclear medical imaging
89-Sr	50.5 d	5.39E-01	**2.1E+13**	(n,γ), (n,p)	reactor	Palliative therapy of bone metastases
82-Sr/Rb	25.5 d	1.36E-01	**0.5E+13**	(p,4n)	accelerator	Generator, PET, myocardial perfusion
68-Ge/Ga	288 d	9.38E-02	**3.6E+12**	(p,2n), spall.	accelerator	Different PET imaging procedures, calibration of PET
67-Cu	61.9 h	3.83E-01	**1.5E+13**	(p,γ)	accelerator	Therapy with labelled antibodies and peptides
44-Ti/Sc	47.3 y	1.77E-03	**0.7E+11**	spallation	accelerator	Generator, great potential for PET
32-Si	101 y	3.03E-02	**1.2E+12**			Important isotope for R&D and technical application
26-Al	7.16e5 y	6.05E-03	**2.3E+11**	(p,n)	cyclotron	Important isotope for R&D and technical application
28-Mg	20.9 h	1.45E-02	**0.6E+12**			Important isotope for R&D

Options for harvesting radioisotopes



Beam dump irradiations of targets for off-line mass separation

Operation of dedicated target stations optimised for on-line production of selected nuclei



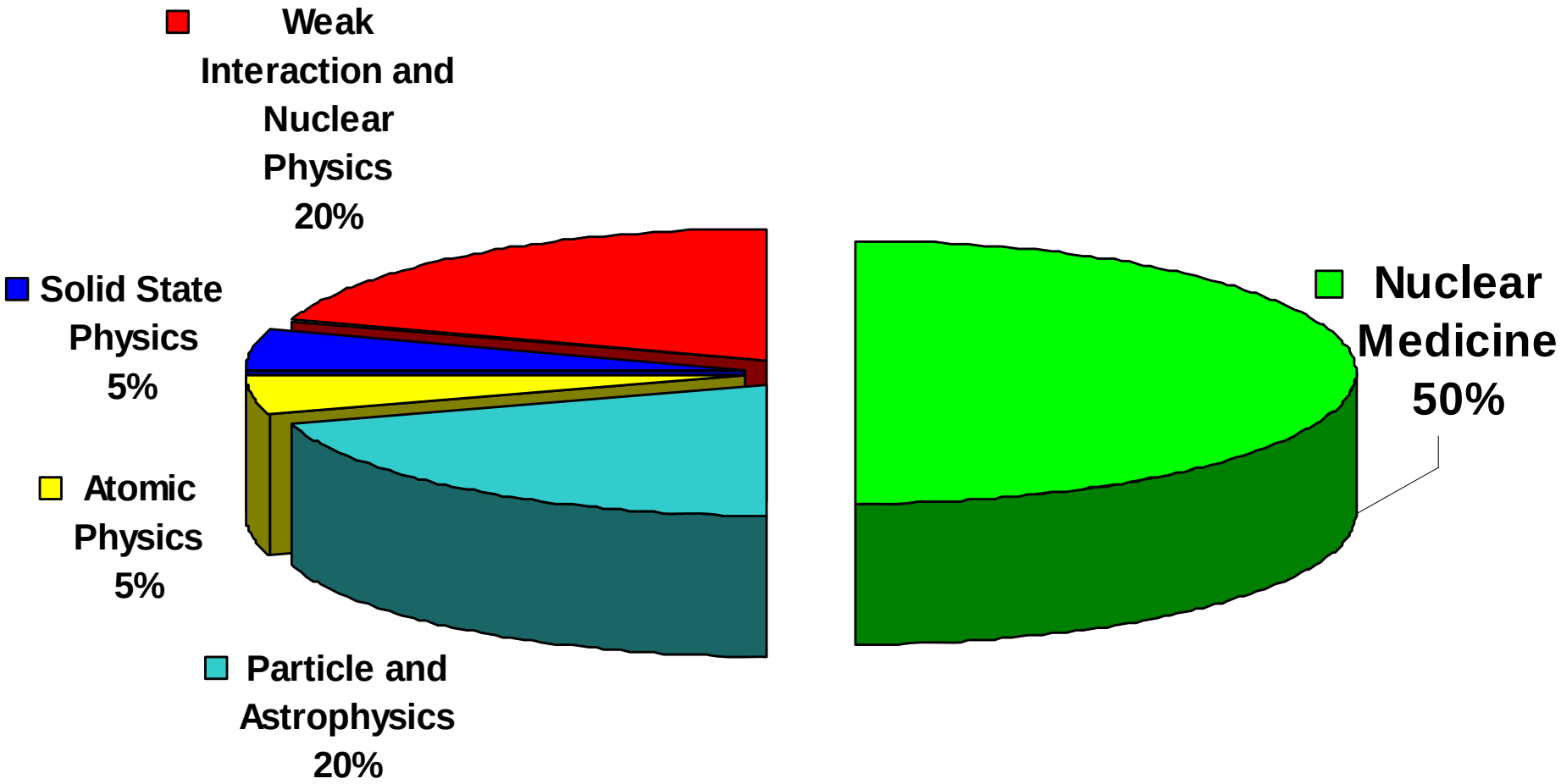
Extraction of nuclei from the liquid metal flow of the converter target for off-line mass separation

Extraction of nuclei from spent target units for off-line mass separation

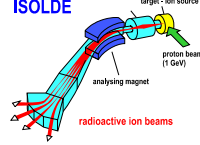
Parasitic collections on-line from the unused beams in the mass spectrum

Extraction of nuclei from other radioactive waste

EURISOL 2020(?) 2×7200 hours Of radioactive ion beam



Conclusion and outlook



- A large amount of data has shown the strength of medical application of carrier free lanthanides.
- It is timely to introduces mass separation as a chemical unit operation in the industrial isotope production line for quality improvement.
- Present research facilities can cover the upcoming need of carrier free rare isotopes in amounts that allows the development and testing new radiopharmaceutical products.
- A number of new exotic nuclide can be made available for medical research.
- Large scale industrial production can be made by harvesting the nuclei of interest from spent targets in present and future physics research facilities.
- Variants of the ISOLDE target and ion-source techniques lend itself to this kind of industrial isotope production.
- A program of validation securing of these new production methods should be started now so that they are ready in time for future marketing and mass production.
- A joint technology transfer enterprise between industry and CERN/ISOLDE during a 10 y period has been proposed.

References and acknowledgements

- The ISOLDE collaboration at CERN, Switzerland:
  http://isolde.web.cern.ch/ISOLDE/frames/isoframe.html
- The European EURISOL project
  http://www.ganil.fr/eurisol/index.html
- TARGISOL
  http://www.targisol.csic.es
- TRIUMF Canada
- http://www.triumf.ca/
- The US Rare Isotope Accelerator project RIA:
- http://www.orau.org/ria/
- MEGAPIE Collaboration PSI, Switzerland