Solid State Physics and Perturbed Angular Correlations at ISOLDE

new tools, new ideas, new people

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Universität des Saarlandes -> Universität Duisburg Essen & ISOLDE-CERN

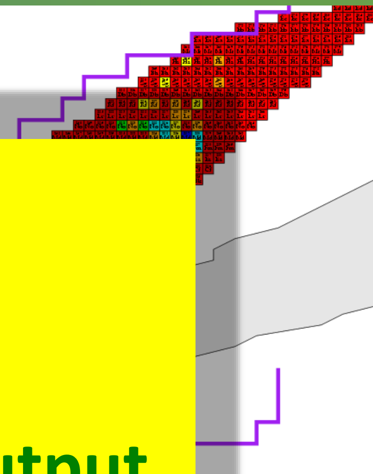
SSP and OF EXOTIC RADIOACTIVE BEAMS

Presenting a review of today's offer at:

- **Nuclear physics and Astrophysics**
- **Material's science**
- **Bio-physics**
- **Life sciences**
- What is **WORKING** and **NEW**...
- What is **USEFUL** and ... **USABLE**
- Who are the **USERS** ...
- **Where** ...

... aiming to work better in 10 – 15 years !

KEY Features for Sustainability of RIB Applications



ADVANTAGES ➔ SENSITIVITY / TRACEABILITY:
Very low concentrations of radioactive impurity atoms in **materials,**
surfaces or i

ADVANTAGE
Element tra
decay add ch
spectroscopy
photolumine
deep level tr
spectroscopy

SUSTAINABILITY !!!!!!

...Yes but under conditions ...

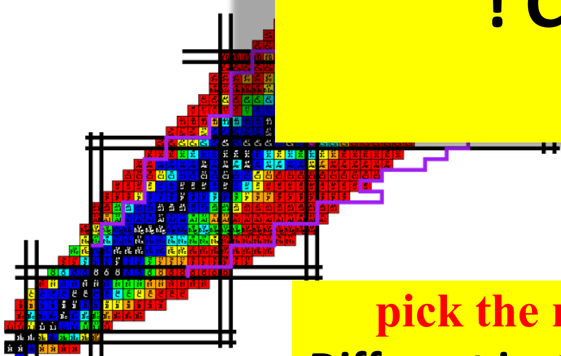
Originality & Uniqueness of Science output

Large panoply of Elements and isotopes
Variety, Intensity and Purity

! CONCENTRATION OF EFFORTS !

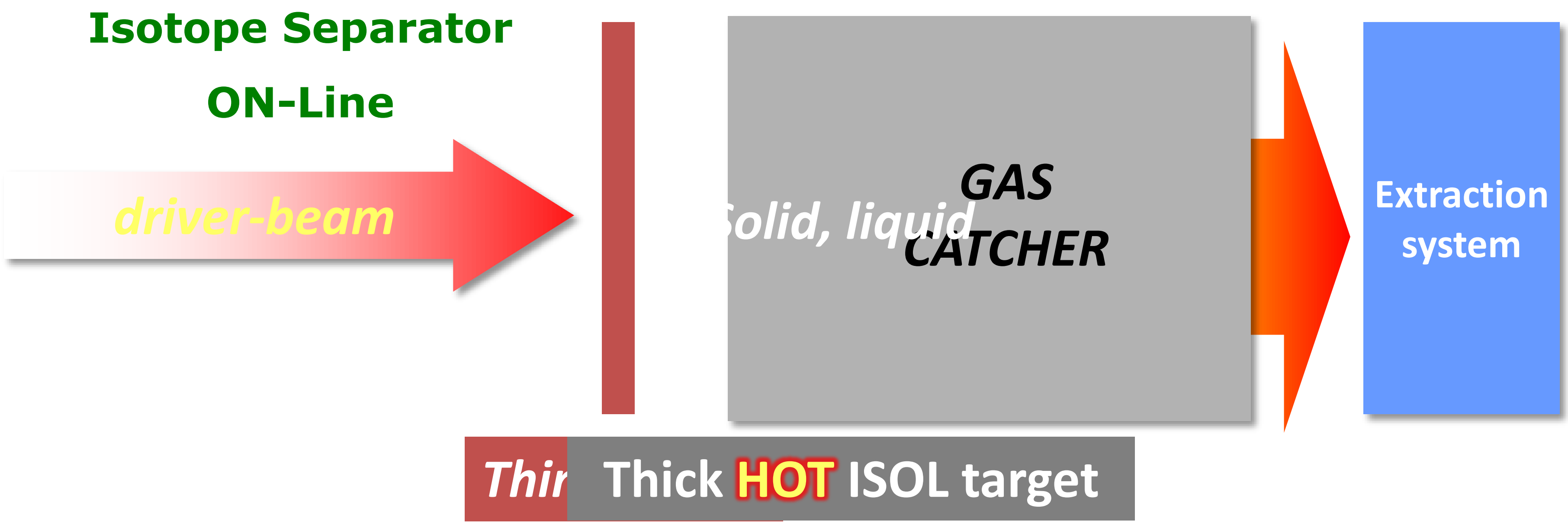
ABILITY
d Purity
isotopes

MATION:
β-NMR)
borhood
e-, β-,β+, α Emission Channeling
direct and precise lattice site location.



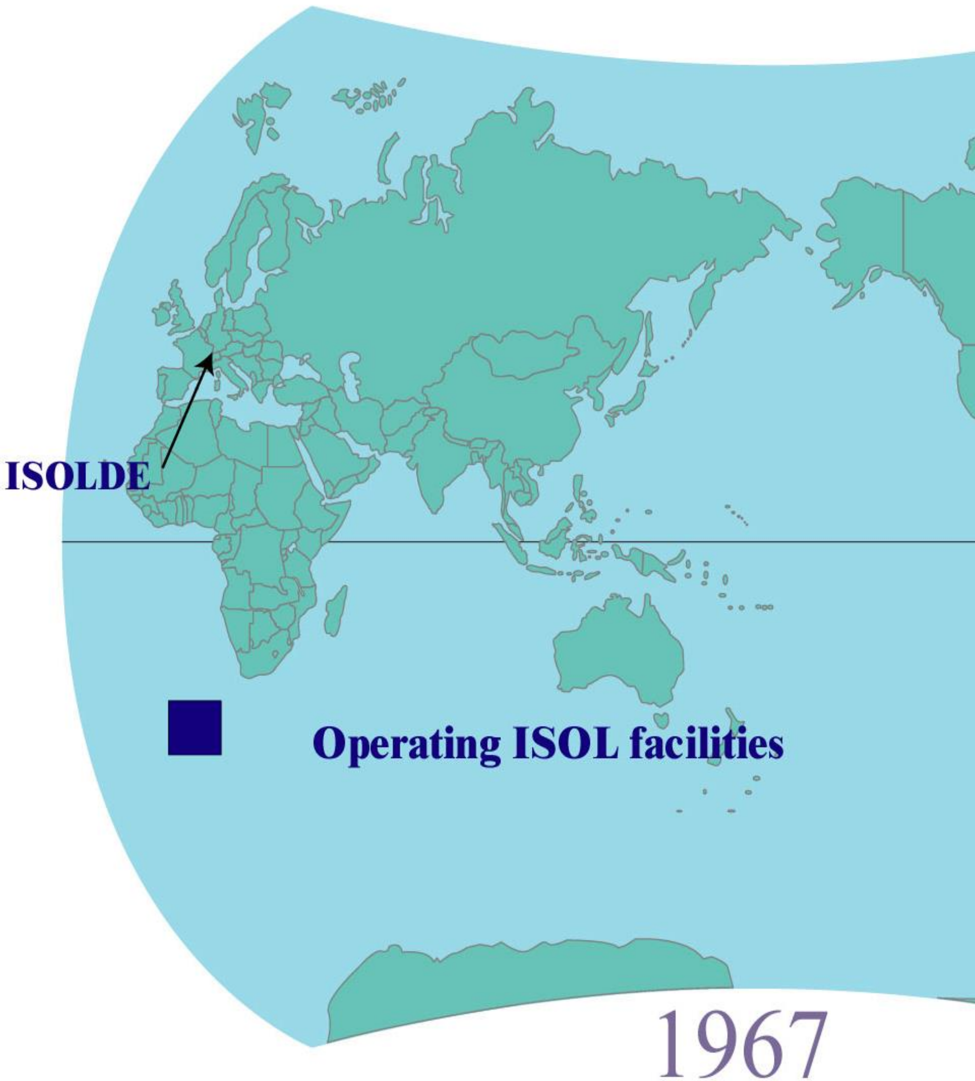
pick the right isotope to have TUNEABLE RANGE OF ENERGY DEPOSITION :
Different isotopes and decay particles can adapt to tumor or tumor cell killing.

ISOL (ISOLDE EURISOL)



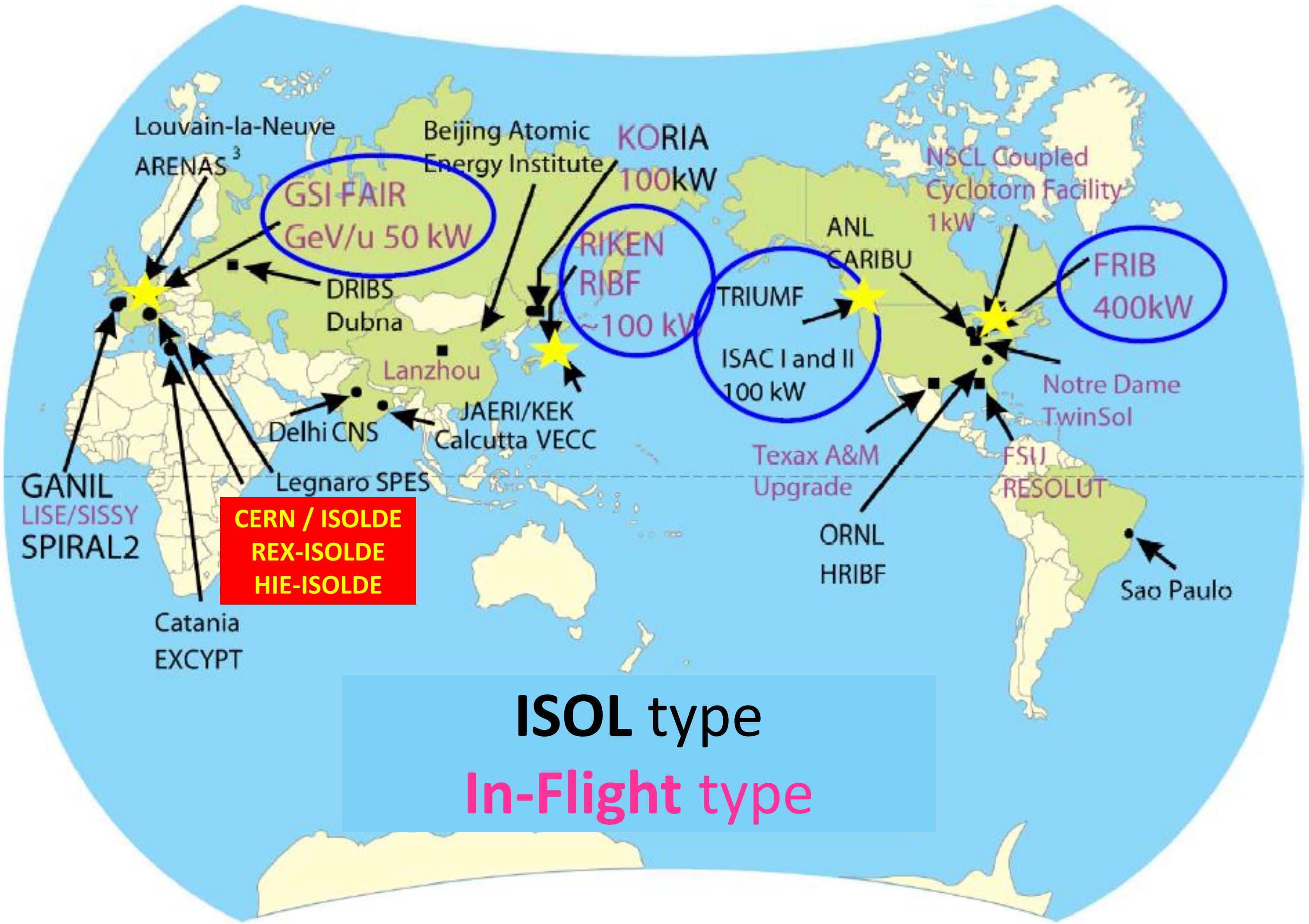
Isotope Separator ON-LINE – ISOL:
Such an instrument is essentially a target, ion source and an electromagnetic mass analyzer coupled in series. The apparatus is aid to be on-line when the material analyzed is directly the target of a nuclear bombardment, where reaction products of interest formed during the irradiation are slowed down and stopped in the system.
H. Ravn and B.Allardyce, 1989, Treatise on heavy ion science

Isotope Separator On-Line
ISOL - facilities 1967



Radioactive Ion Beam – facilities Worldwide 2013

- Existing and in preparation

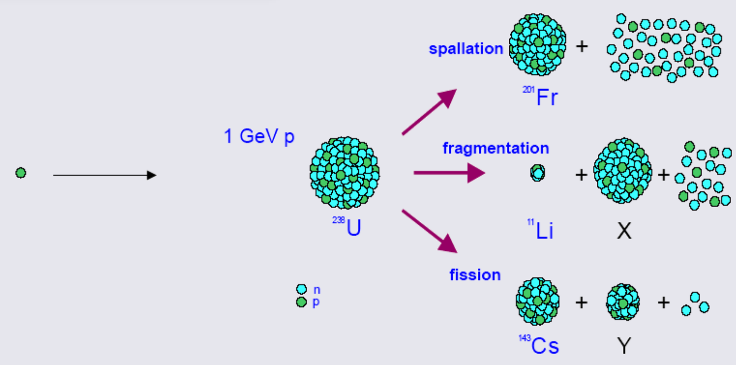


- After B. Sherill

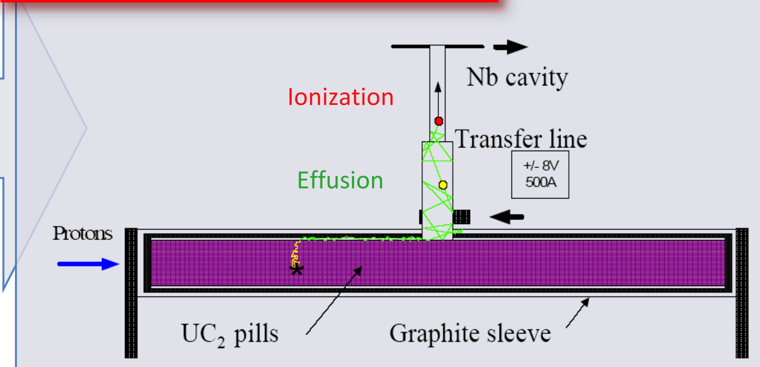
ISOLDE: CERN's longest running facility

The ISOL Process

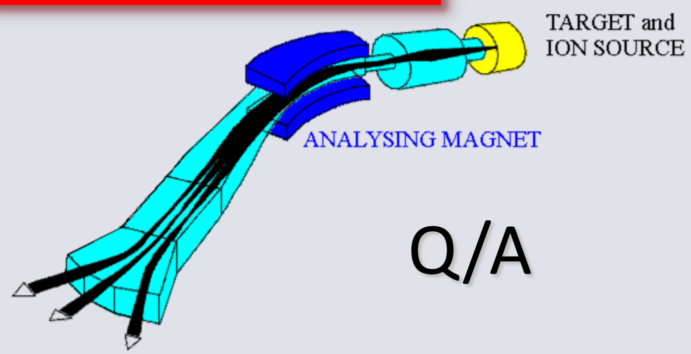
Production



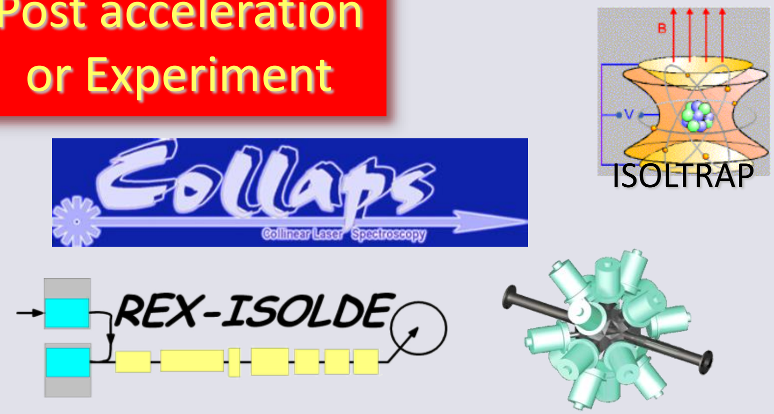
Extraction/Ionization



Isotope Separation

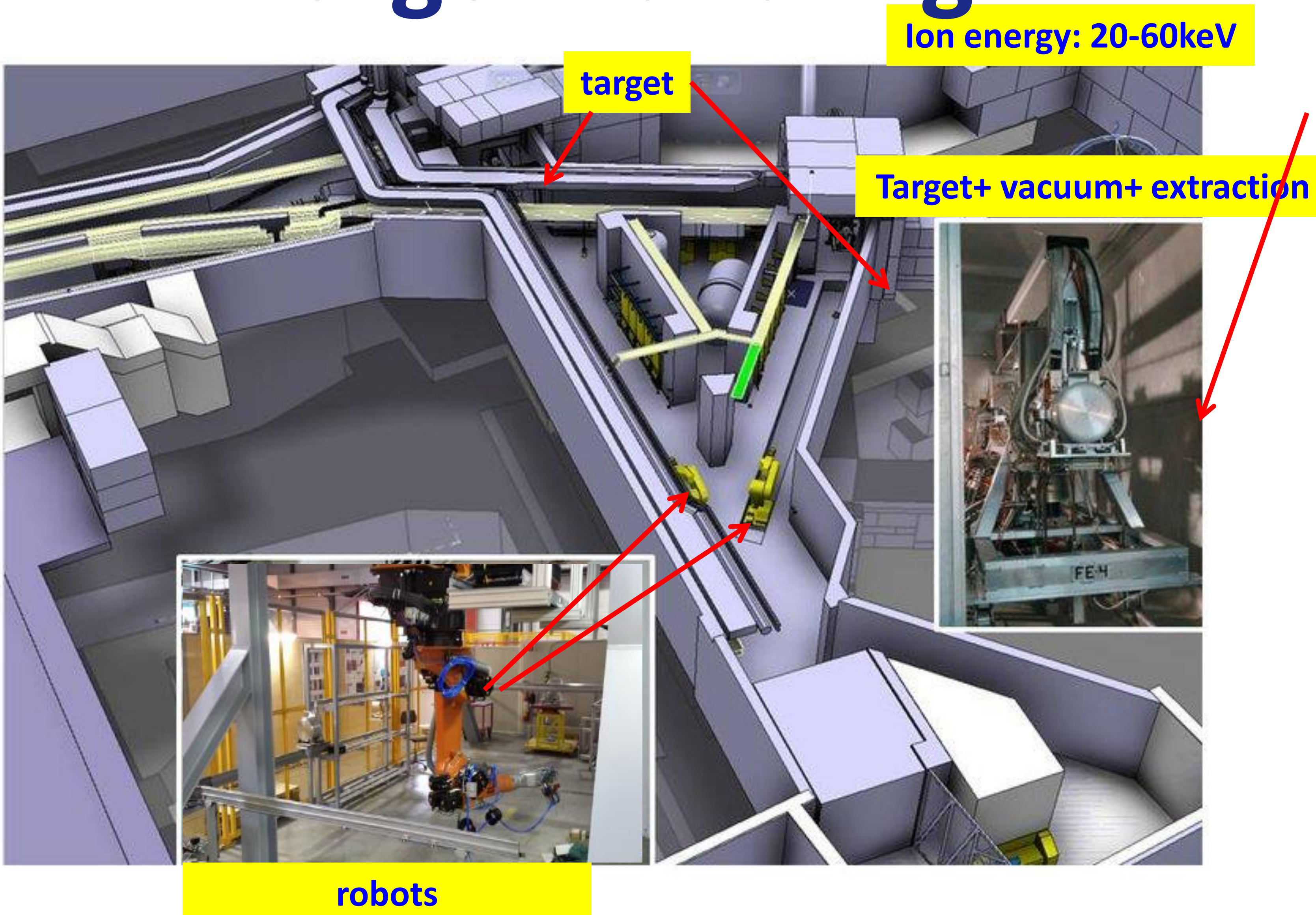


Post acceleration or Experiment

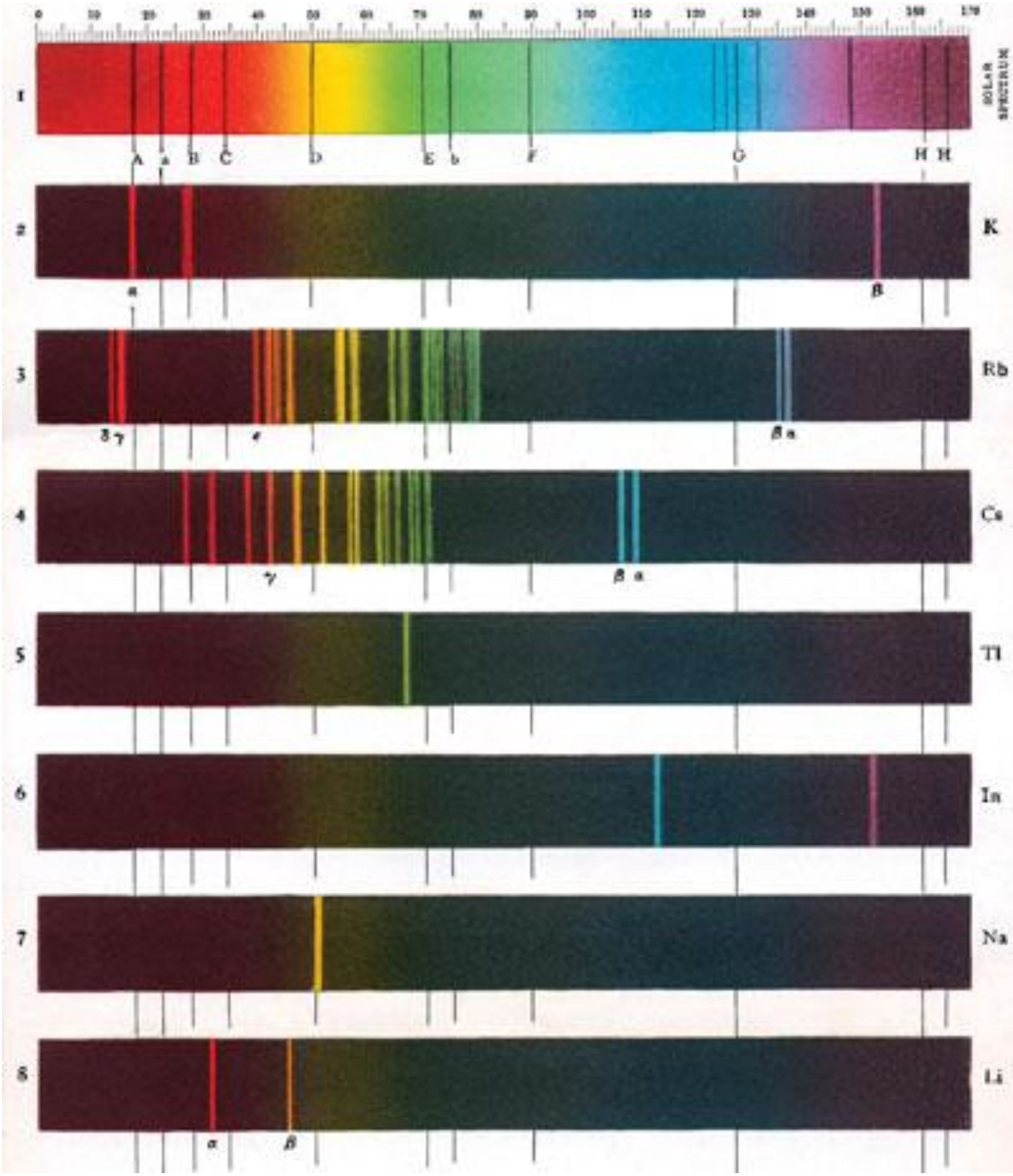


Fast, Efficient, Universal and Selective!

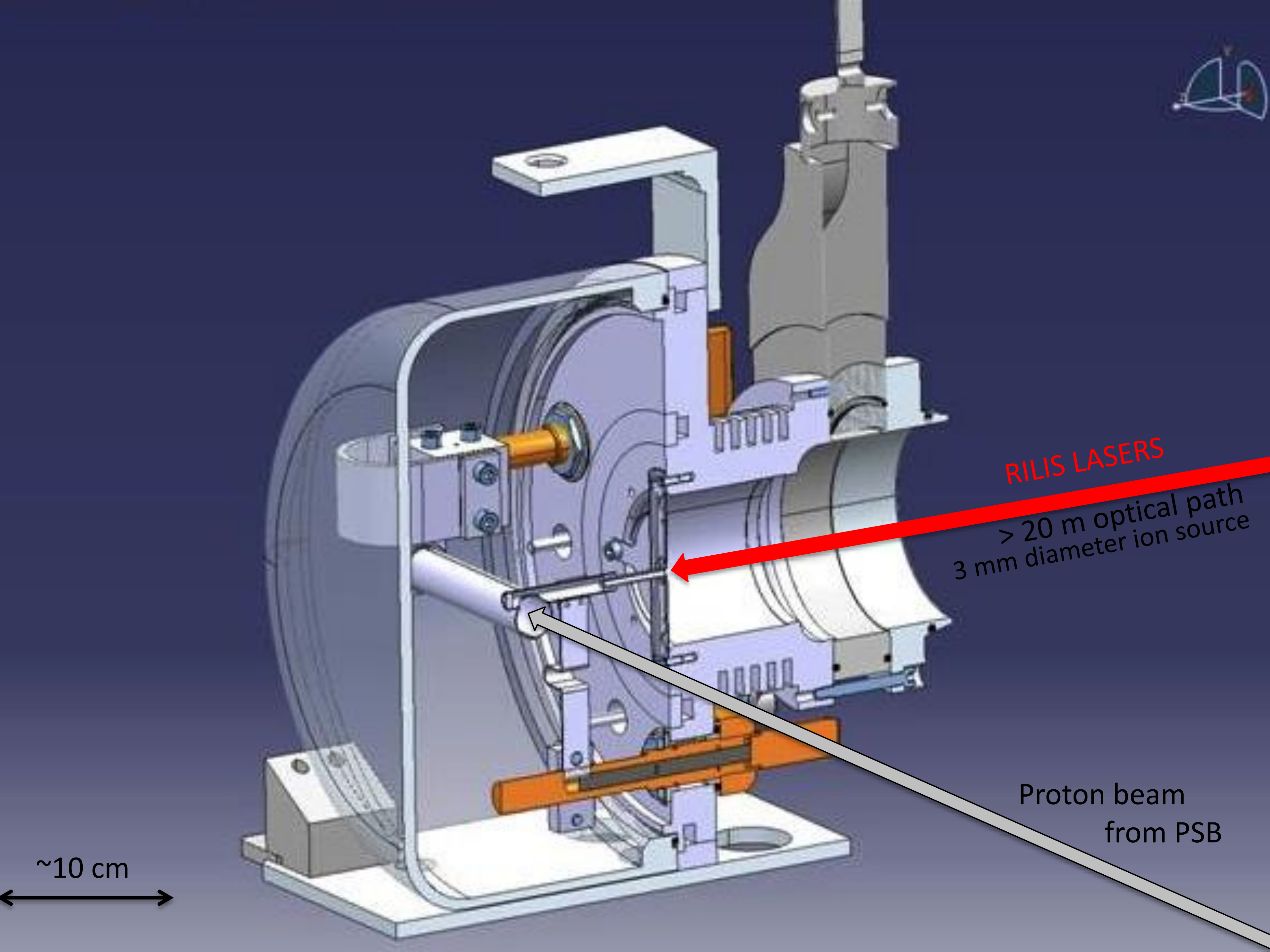
Target handling



RILIS (Resonant Ionisation Laser Ion Source)

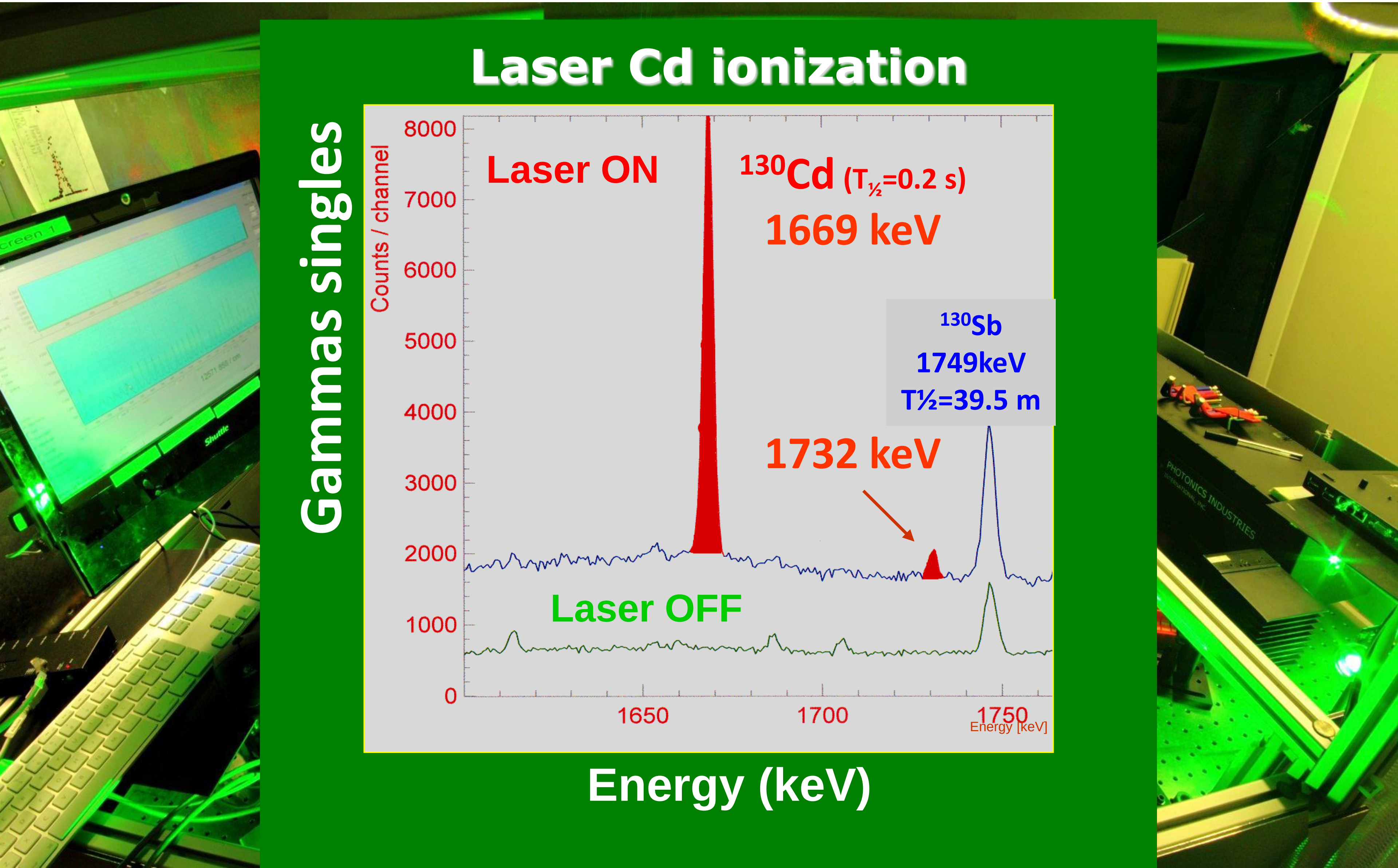


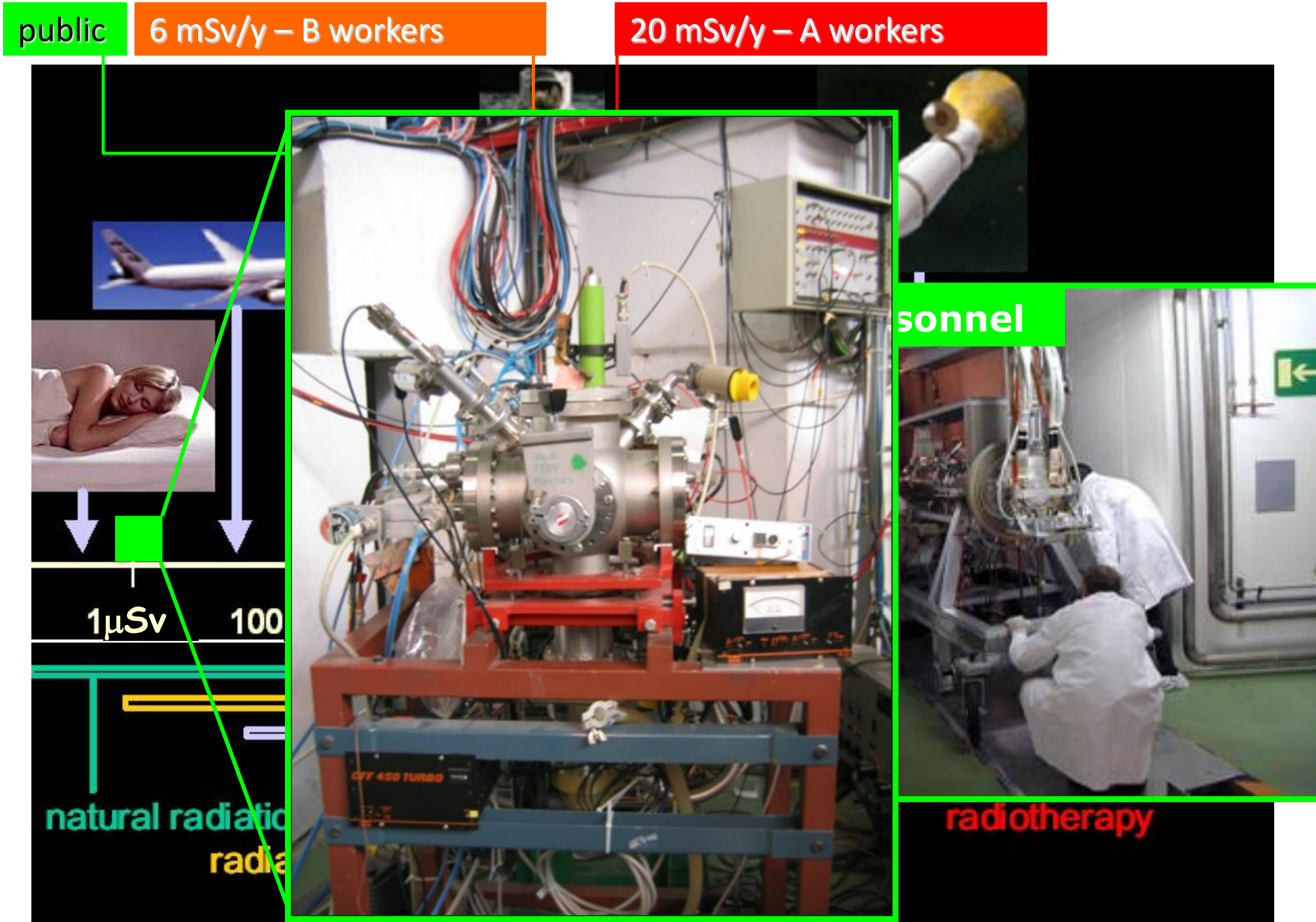
The atomic line spectra are the element's fingerprint



RILIS LASERS
> 20 m optical path
3 mm diameter ion source
Proton beam
from PSB
~10 cm

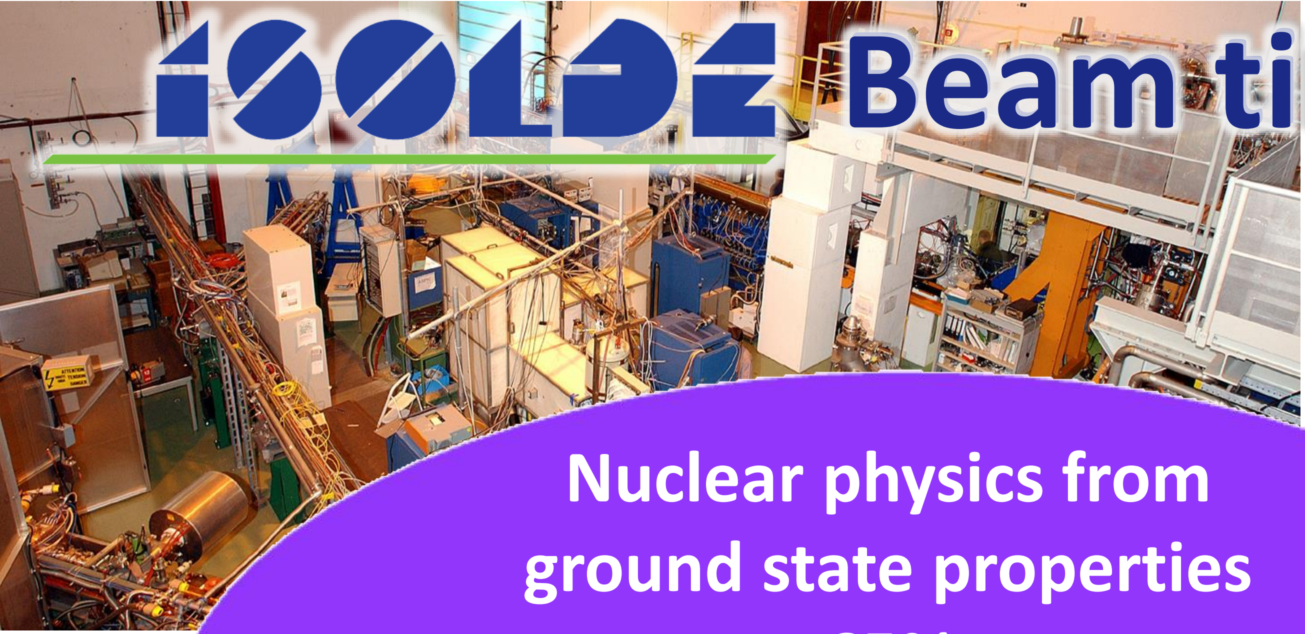
RILIS in action





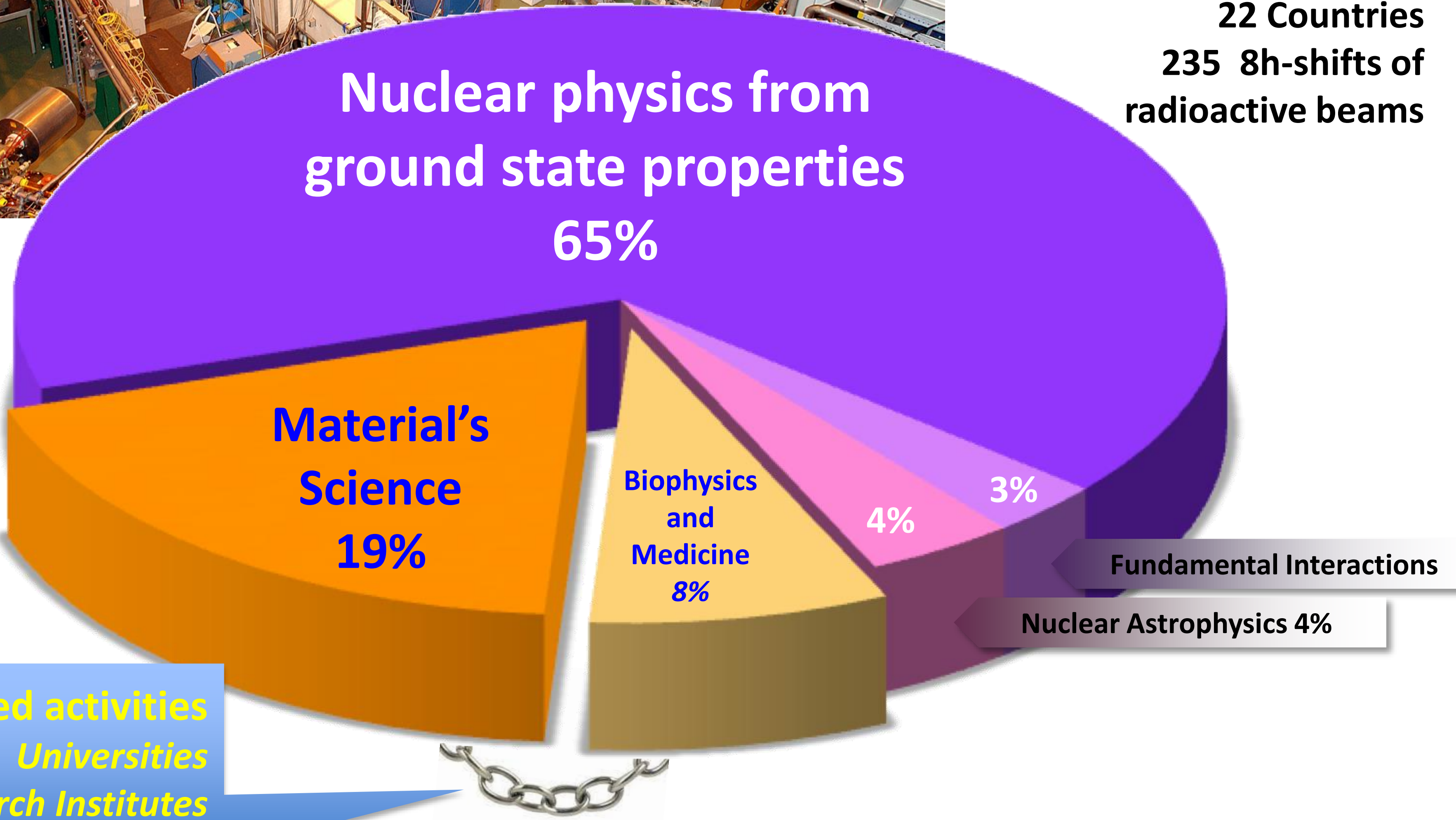
1 Gy = 1 J/Kg leads to a temperature increase of only 2.4 .10^{-4} °C

ISOLDE Beam time pie



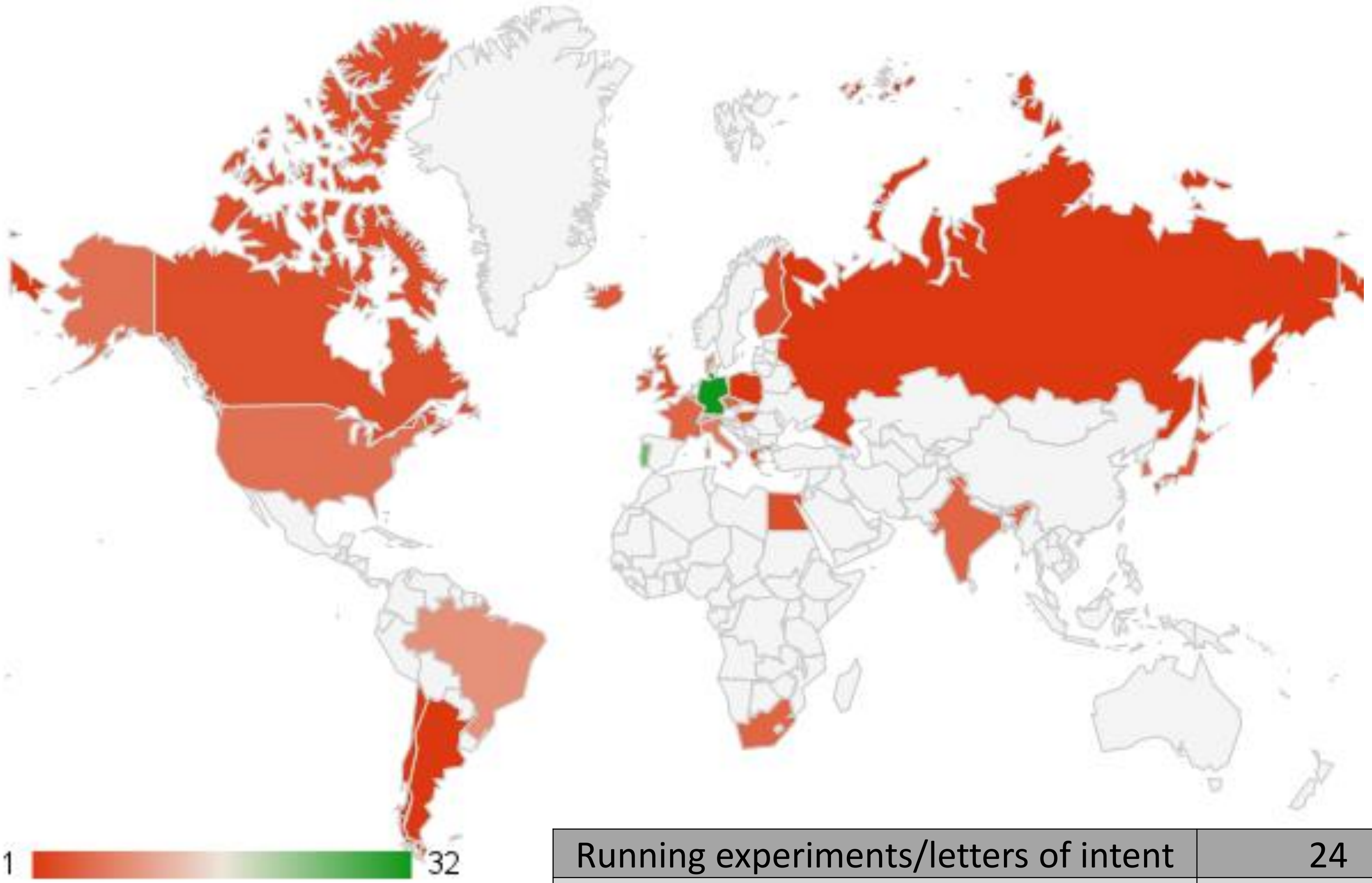
2015

37 Experiments
300 Users
96 Institutes
22 Countries
235 8h-shifts of radioactive beams



Linked activities
Universities
Research Institutes
Hospitals

SSP @ ISOLDE: Diverse community



Running experiments/letters of intent	24
Participating countries	26
Scientists	160

Current and PROSPECTIVE Work using EXOTIC Radioactive isotopes

➔ THINKING Materials and Molecular Properties ⇐

dealing with mass, electromagnetism, many body systems and scaling

Atomic-like information is the aim !

➔ Semiconductor Physics (Si, Oxides, organic compounds)

➔ Multi- ferroic- magnetic

➔ Nanomaterials (geometry, downsizing and integration)

➔ Surfaces and interfaces (bulk properties are modified)

➔ Soft matter : graphen,

➔ Bio / Molecular chemistry and physics

➔ THINKING Life Sciences ⇐

optimizing delivering and range of deposition of highly concentration of energy upon radioactive decay into the living body or cell of interest.

Enlarging the choices of radioisotopes is the aim !

➔ NEW isotopes and decay modes for diagnosis and treatments.

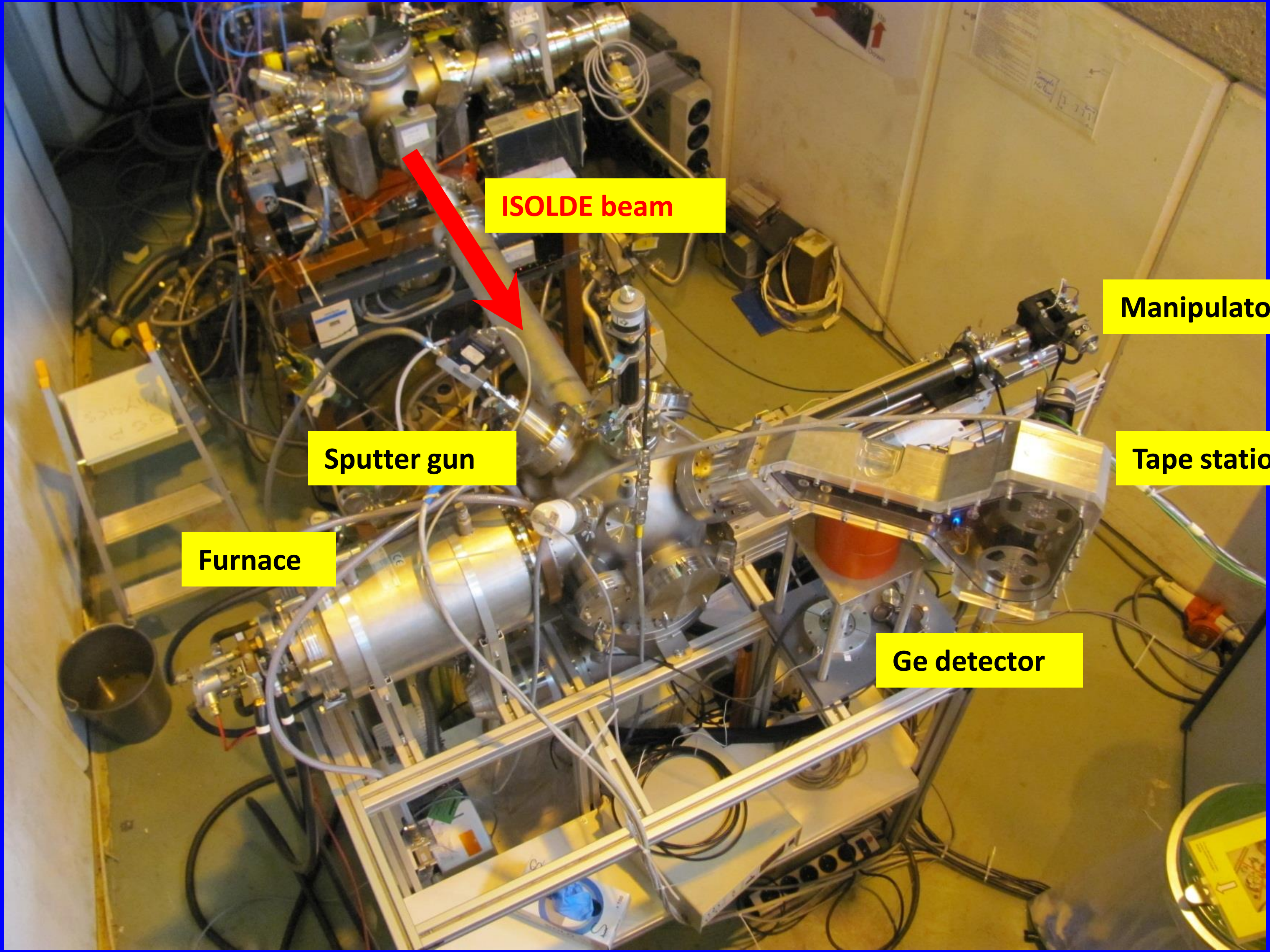
RECENT DEVELOPMENTS

Material's Science Applications

a commitment for the future based on facts !

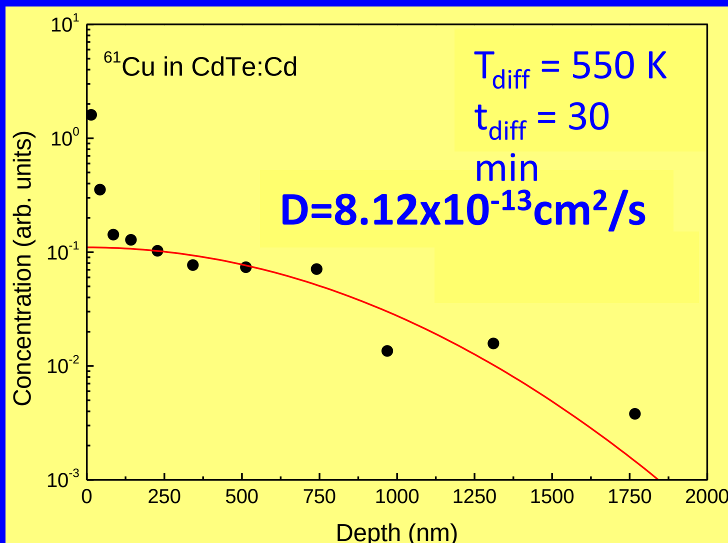
On-line diffusion chamber at ISOLDE

2011



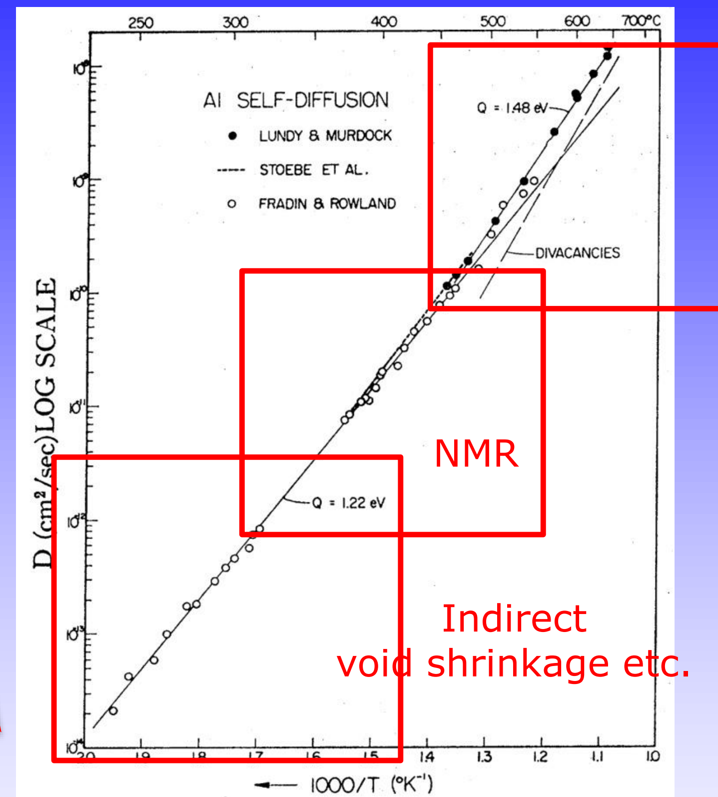
Understanding Diffusion and self diffusion ... On-line !

^{61}Cu (3.3h) diffusion in Cd saturated CdTe



Isotope	$t_{1/2}$	Detection
^{38}Cl	37.3 min	$\beta^-_{4800keV}$; $\gamma_{2168keV}$
^{11}C	20.38 min	β^+ ; γ_{511keV}
^{13}N	9.96 min	β^+ ; γ_{511keV}
(^{15}O)	122 s	β^+ ; γ_{511keV}
^{29}Al	6.6 min	$\gamma_{1273keV}$

Unknown Aluminum self diffusion



(T.G.Stoebe *et al.*, Phys. Rev. 166 (1968) 621)

- Single stable Al isotope: no SIMS measurements
  Unknown activation energy of self diffusion in Al
- Unknown role of vacancies, di-vacancies at different temperatures.
- **Unknown Al diffusion in Al-based compounds**

From the Avogadro Project : define Kg in terms of number of Si atoms...

Photo Luminescence – PL

Deep Level Transient Spectroscopy (DLTS)

$$N_A = \frac{V_{mol}}{V_o}.$$

$$N_A = \frac{V_{mol}}{(a^3/n)},$$

$$N_A = \frac{M_{Si}}{m}\frac{V}{(a^3/8)},$$

Scientific American
295, 102 – 109 (2006)

^{28}Si

ISOLDE

LASER

HeCd (3,8 eV)
Nd:YAG (2,3 nm)
Diode (1,9 nm)

Cryostat

He-Bathcryostat (1,5 – 300 K) Closed cycle

Monochromator

Focus: 0,75 m
Gratings: 150 – 1800 l/mm

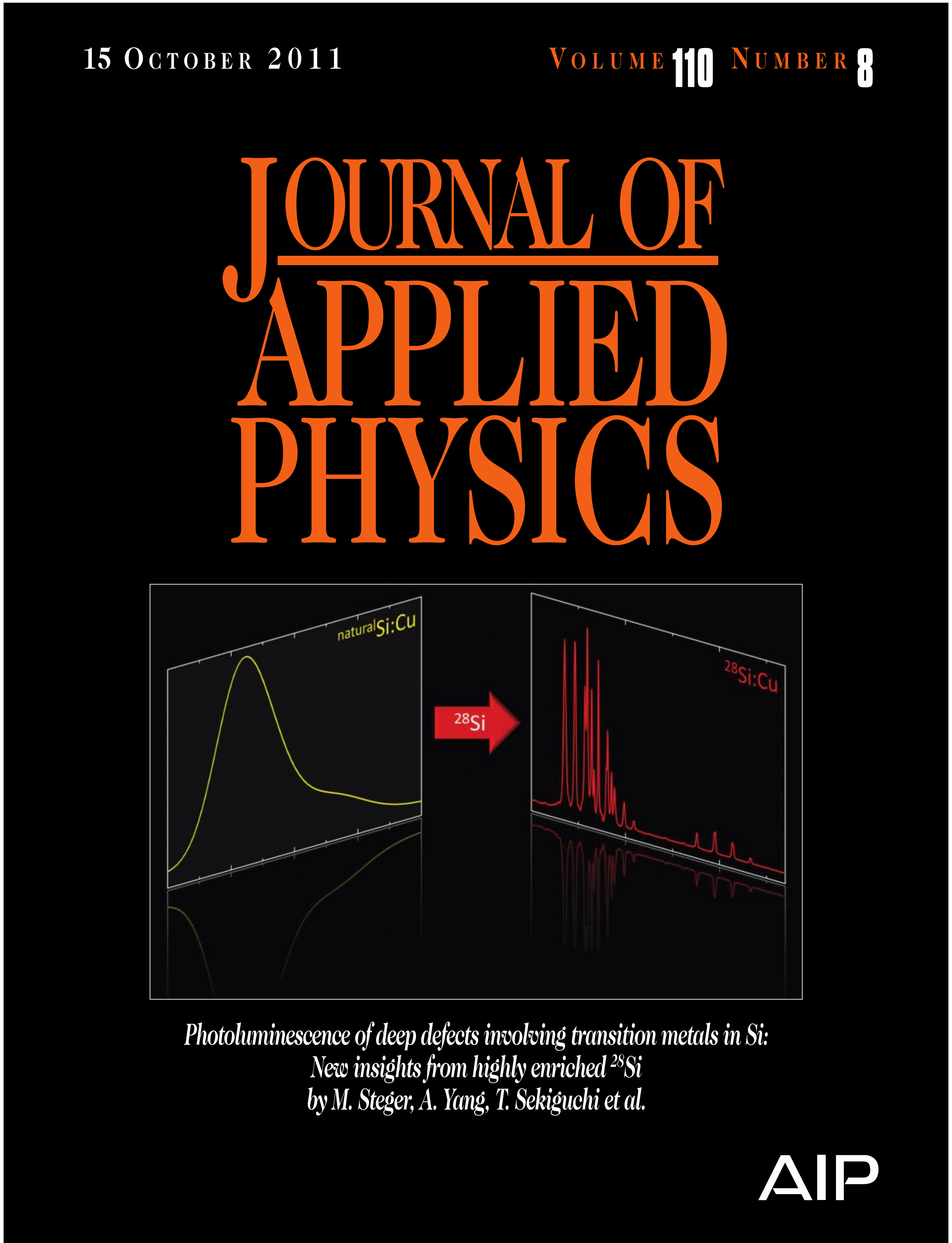
Detectors

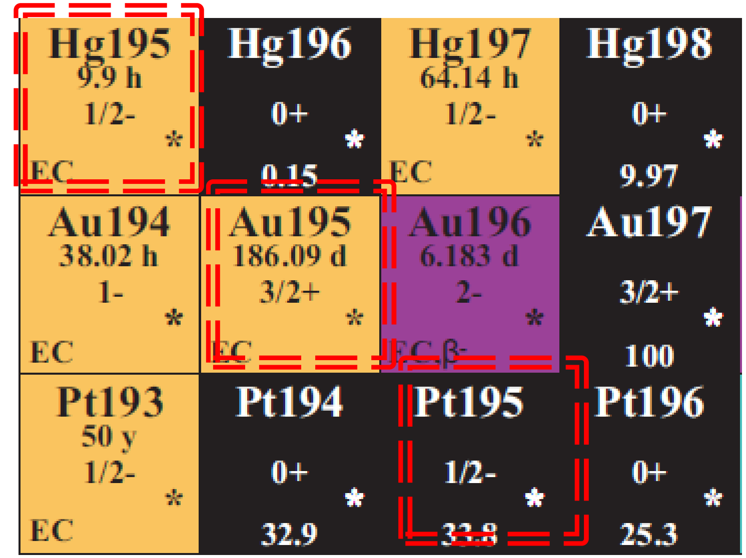
CCD-camera (1,1 - 6,2 eV)
Ge-Diode (0,7 - 1,5 eV)

experimental tool for studying electrically active defects and concentration in semiconductors.

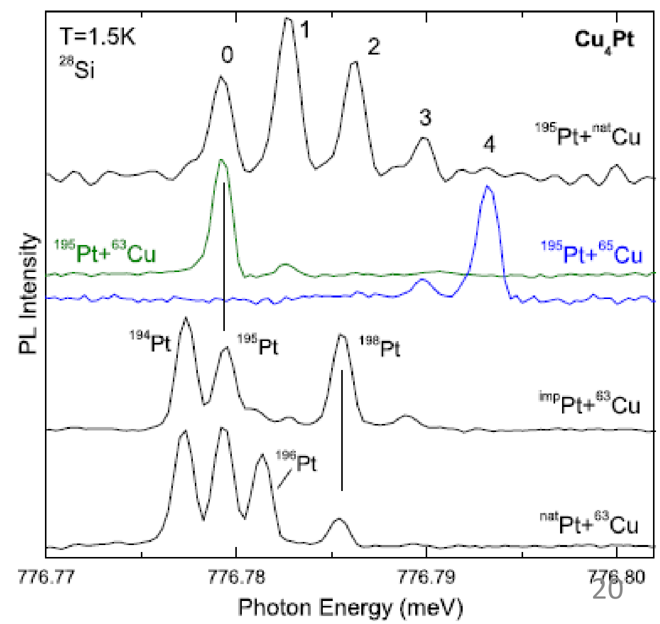
... to measure optical properties of mono-isotopic Si!

IS472

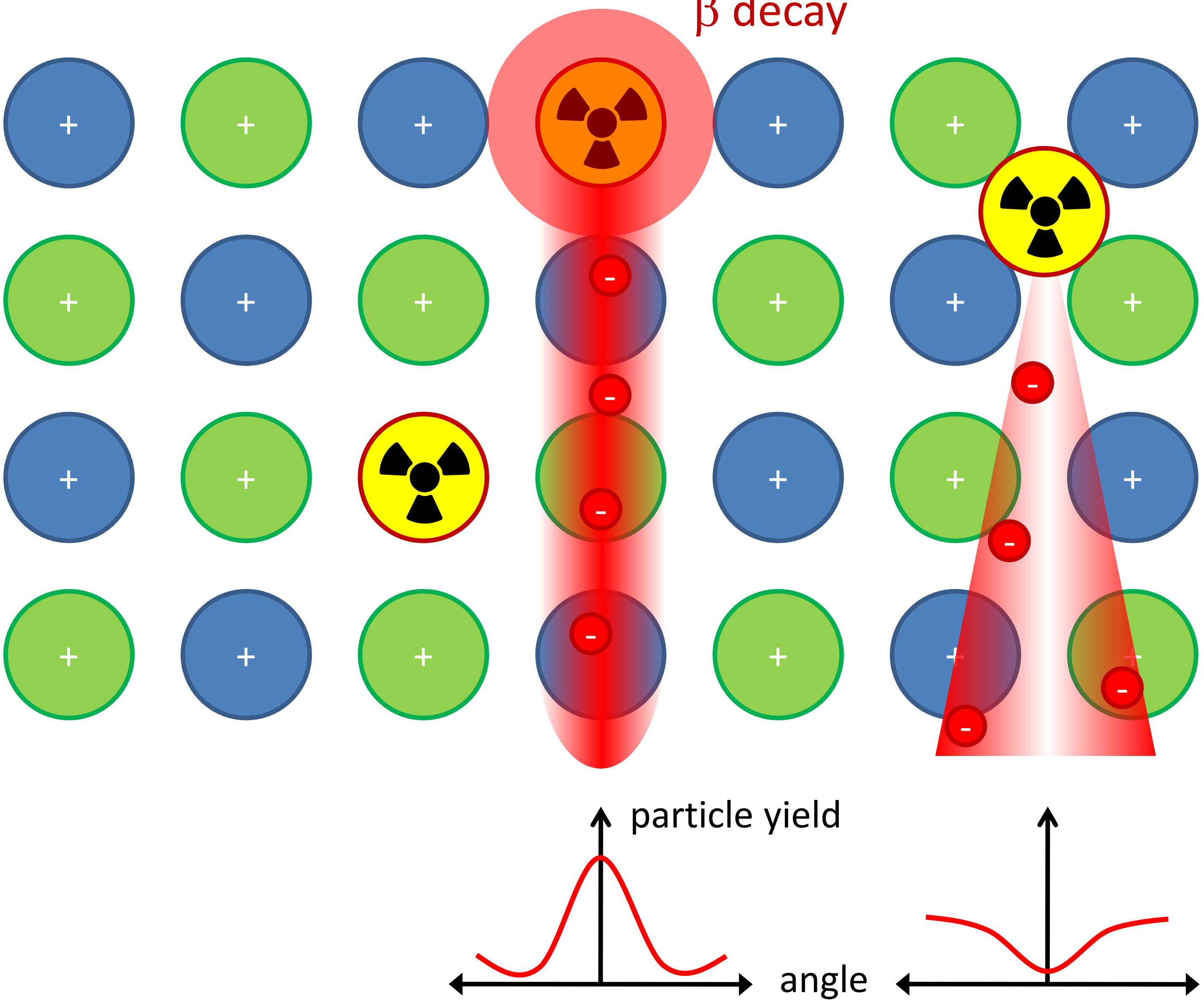




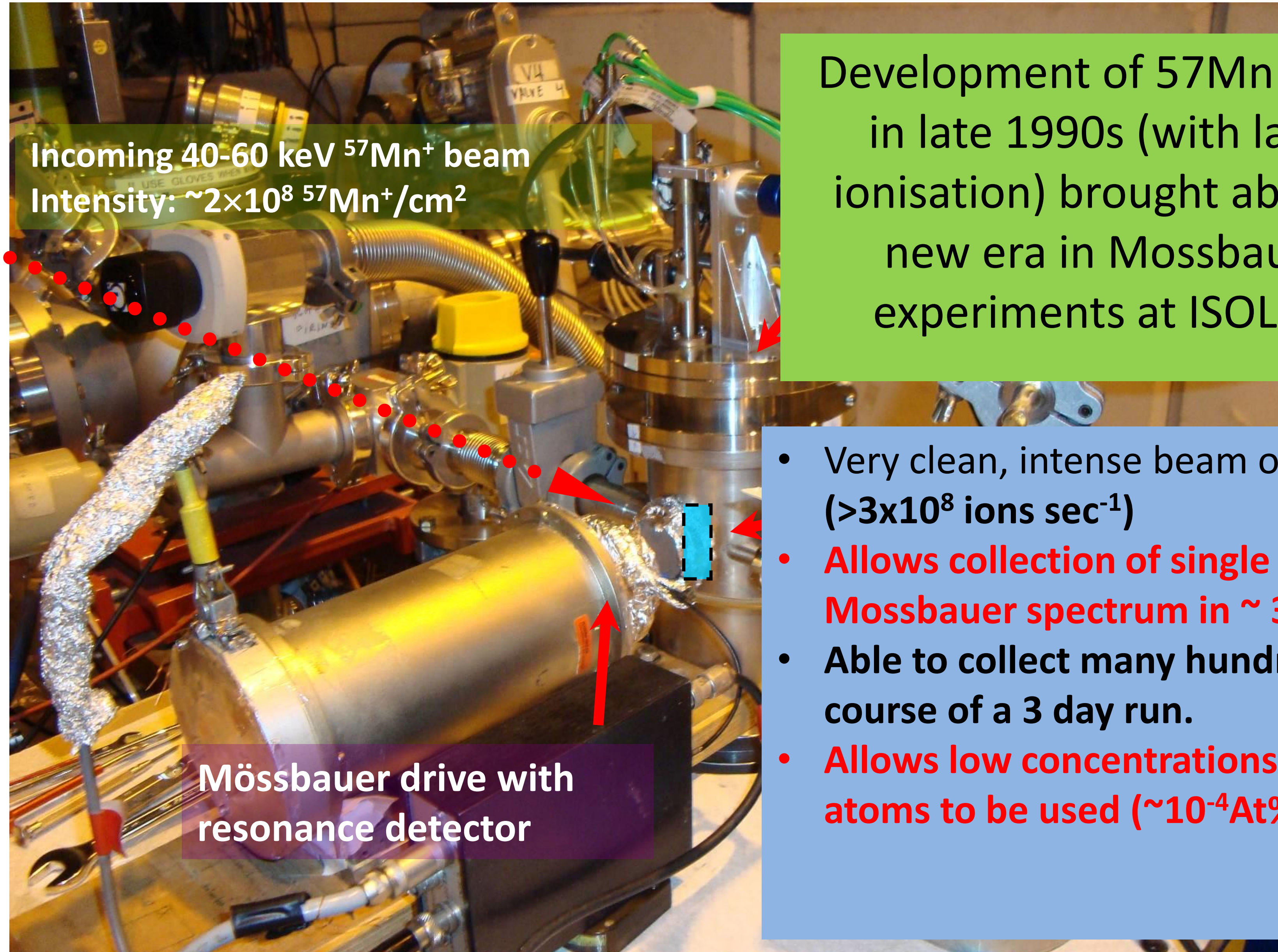
777meV feature, now shown to include Pt and 4 Cu atoms!



Emission Channeling of decay particles, on single crystals (β^-, β^+, c.e., α)



Hyperfine Interactions with Mossbauer spectroscopy



Development of 57Mn beam in late 1990s (with laser ionisation) brought about a new era in Mossbauer experiments at ISOLDE.

- Very clean, intense beam of ^{57}Mn **(>3x10^{8} ions sec^{-1})**
- **Allows collection of single Mossbauer spectrum in ~ 3 mins.**
- **Able to collect many hundreds over course of a 3 day run.**
- **Allows low concentrations of probe atoms to be used (~10^{-4}At%)**

Fe: ZnO a ferromagnetic semiconductor? (nope!)

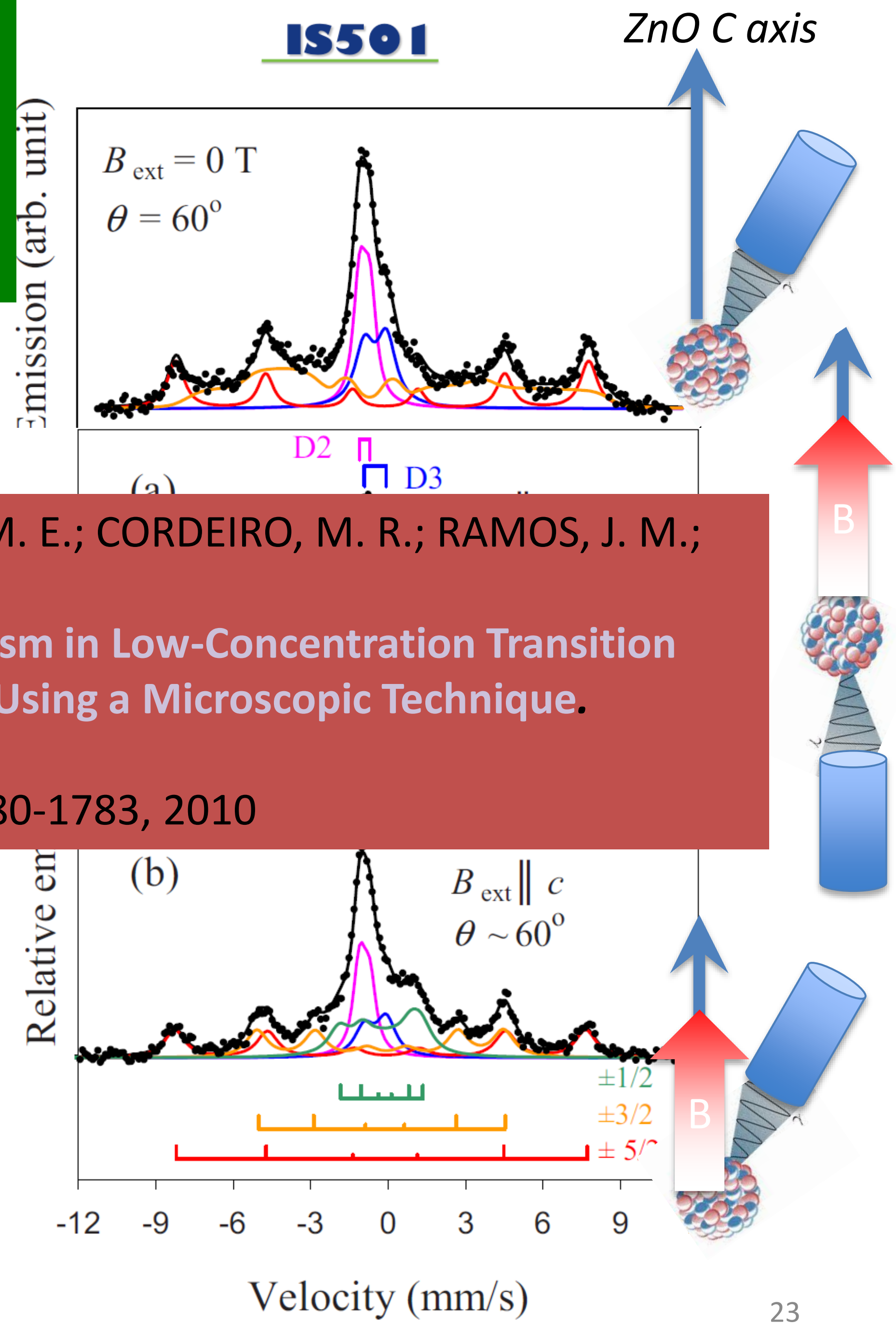
6 fold spectrum: characteristic of magnetic structure (at room temperature!!!).

DOGRA, R.; CARBONARI, A. W.; MERCURIO, M. E.; CORDEIRO, M. R.; RAMOS, J. M.; SAXENA, R. N.
Search for Room Temperature Ferromagnetism in Low-Concentration Transition Metal Doped ZnO Nanocrystalline Powders Using a Microscopic Technique.

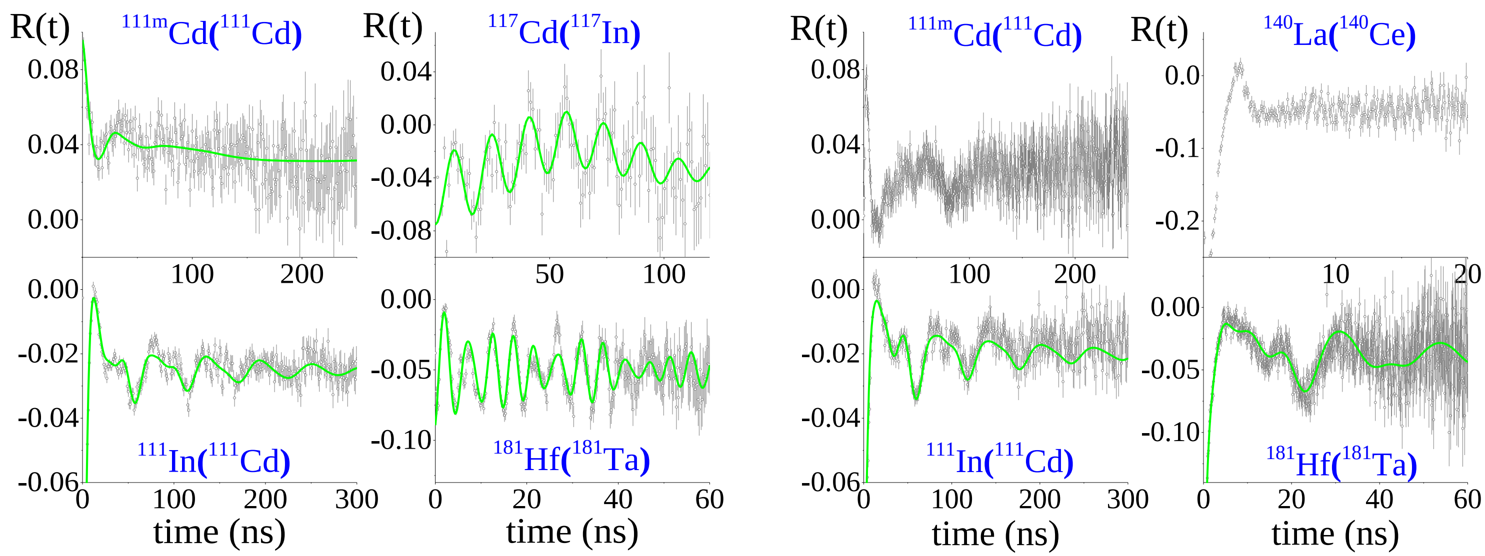
IEEE Transactions on Magnetics, v. 46, p. 1780-1783, 2010

After high-dose implantations, precipitates of Fe-III are formed. These form clusters yielding misleading information about the nature of magnetism in ZnO (as reported by many groups over the last number of years).

Gunnlaugsson *et al* APL **100 042109** 2012



PAC results: no DMS at RT!!!!



SnO_2 PAC Spectra or TiO_2 PAC Spectra

No magnetic interactions observed under the measurements conditions for intrinsic and doped SnO_2 or TiO_2

PAC reveals the presence of two different EFGs for each case: ^{111m}Cd(^{111}Cd), ^{181}Hf(^{181}Ta), ^{117}Cd(^{117}In) and ^{111}In(^{111}Cd), which are assigned to different local environments.

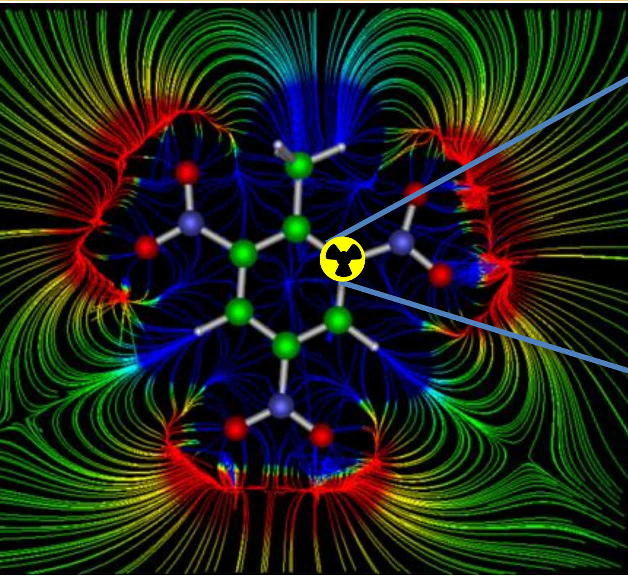
We stress the particular cases of ^{181}Hf(^{181}Ta) and ^{111}In(^{111}Cd), where a major fraction, f1 = 55-80 %, of probe atoms was assigned to substitutional Sn/Ti sites in the Sn/TiO_6 octahedral [Schell, J., CERN-THESIS-2015-229].

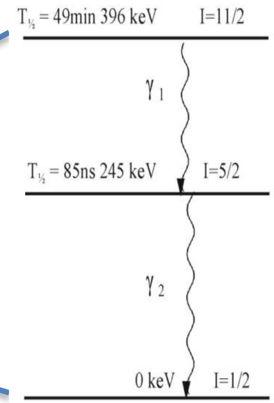
RECENT DEVELOPMENTS

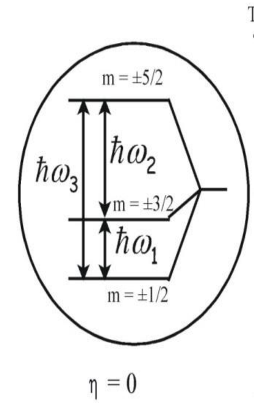
Bio-physics applications

a commitment for the future based on facts !

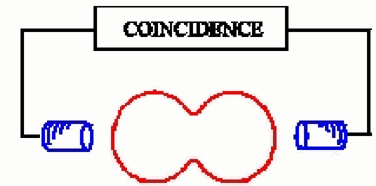
Perturbed angular correlation (PAC) Spectroscopy applied to BIOPHYSICS

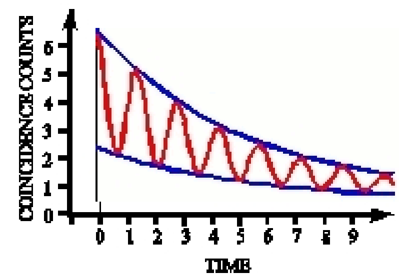




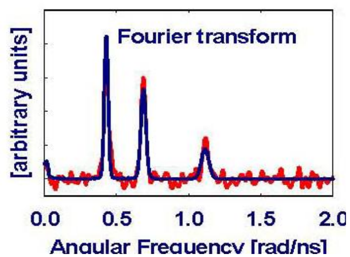


η = 0

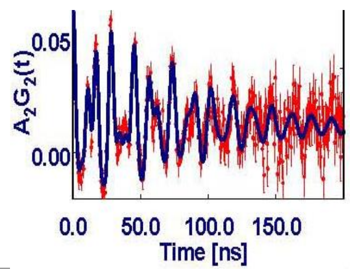




Fourier transform



Least χ^2 analysis



$$A_{22}G_{22}(t) = 2\frac{W(180°,t) - W(90°,t)}{W(180°,t) + 2W(90°,t)}$$

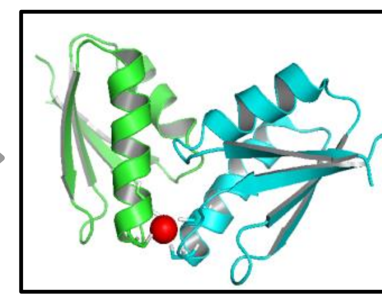
6·180° spectra and 24·90° spectra

ω_Q and η

+

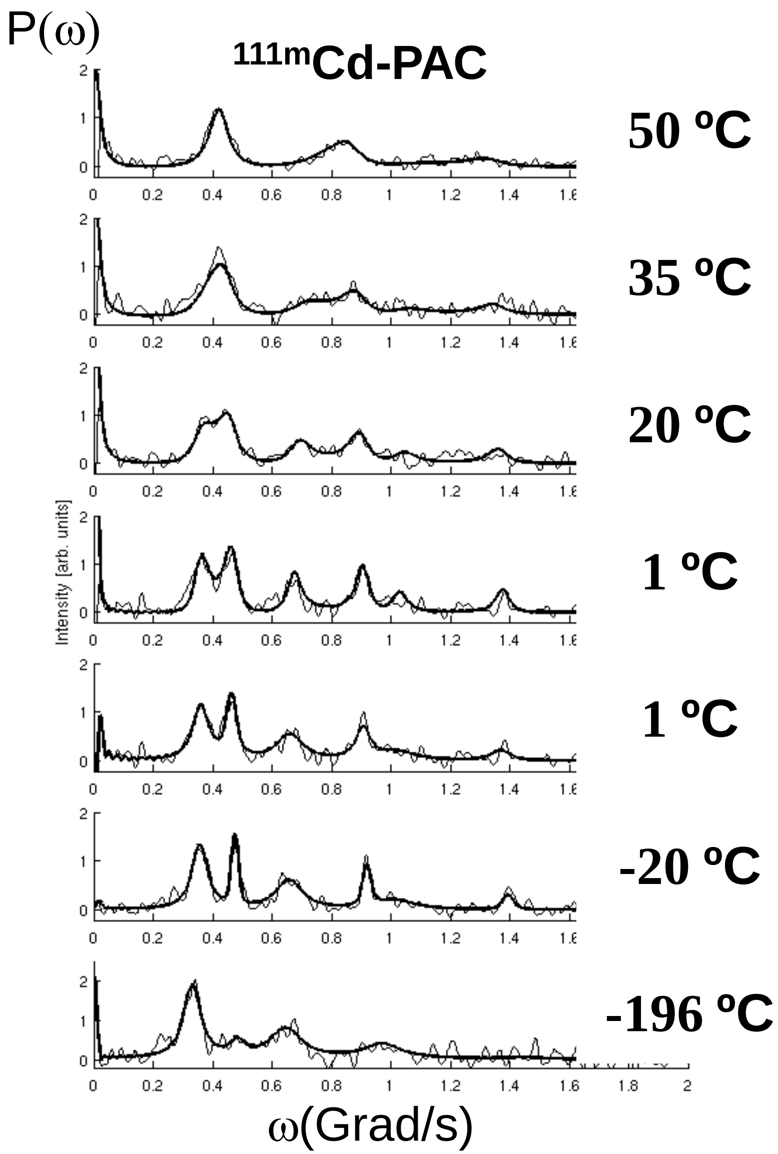
BASIL model

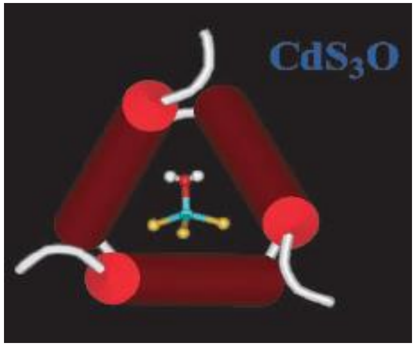
QM calculations

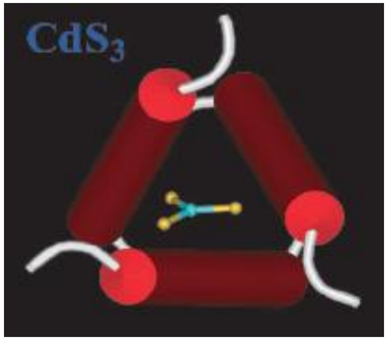


^{111m}Cd PAC (48M) - De novo designed heavy metal Ion binding proteins: ns dynamics

IS488







Temp [ºC]	τ_1 [ns]	τ_{-1} [ns]
1	52	48
20	42	36
35	28	20
50	19	12

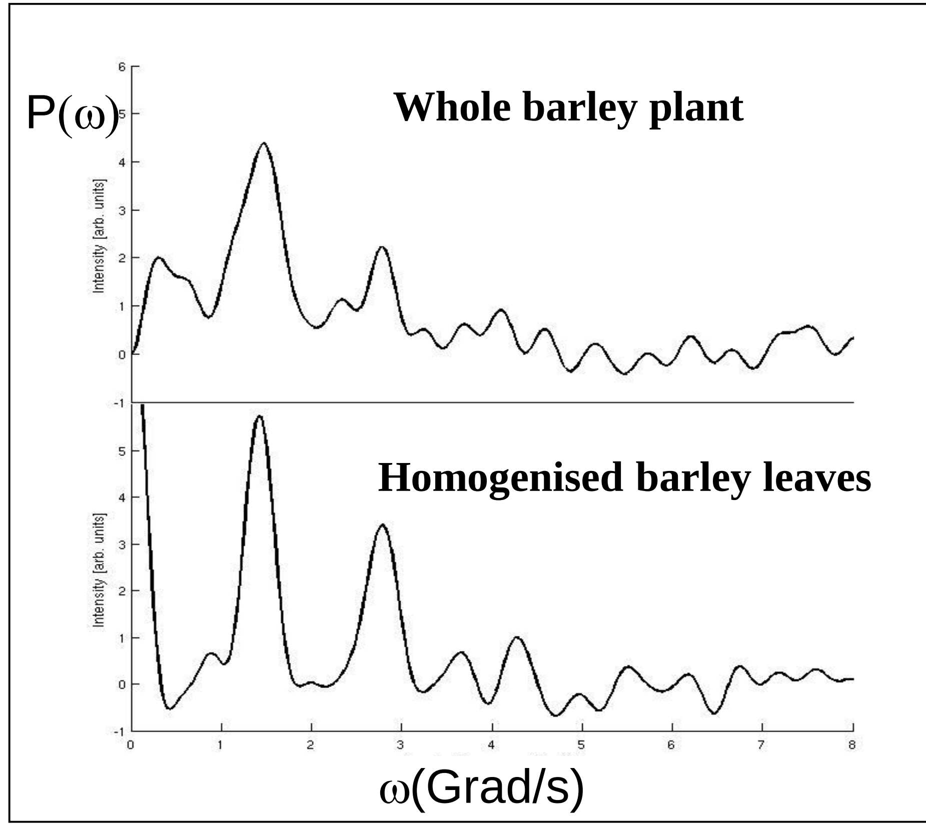
Stachura et al. Manuscript Science in preparation

In vivo experiments Hg(II) binding to barley

^{199m}Hg PAC (42M)

IS488

Adolph et al. *Chem. Eur. J.*, **2009**, 15, 7350 – 7358



- 5-7 days-old plants
- Plant inserted into test tube.
- Fast uptake of Hg(II) (<1h)
- Bound to large molecules, similarities to HgS_2 compounds

RECENT DEVELOPMENTS

Life Science applications

a commitment for the future based on facts !

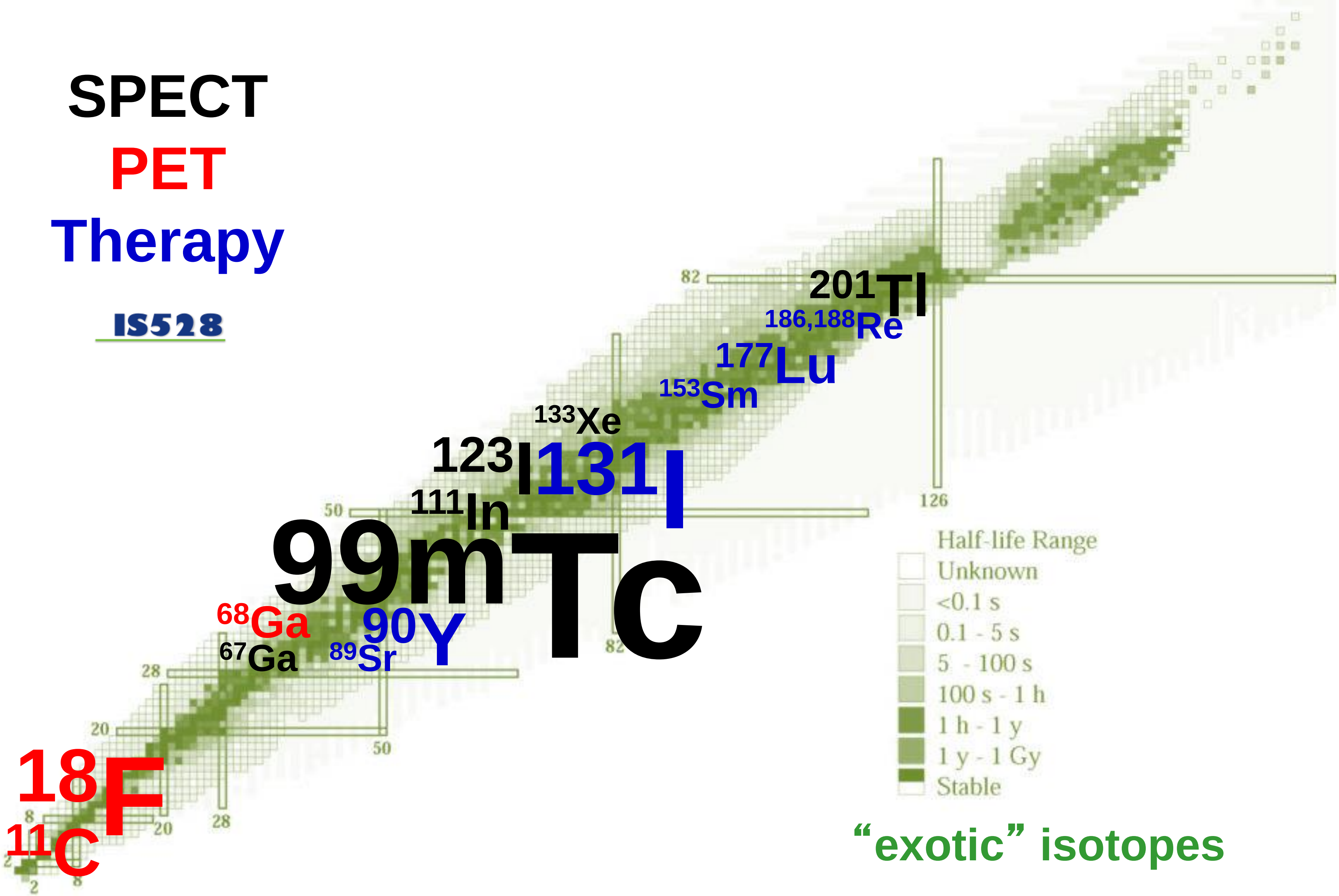
Radionuclides for diagnosis and therapy

SPECT

PET

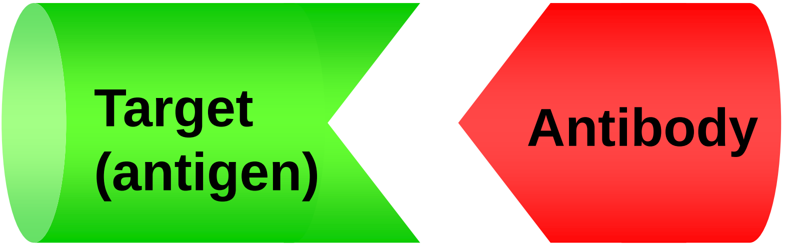
Therapy

IS528

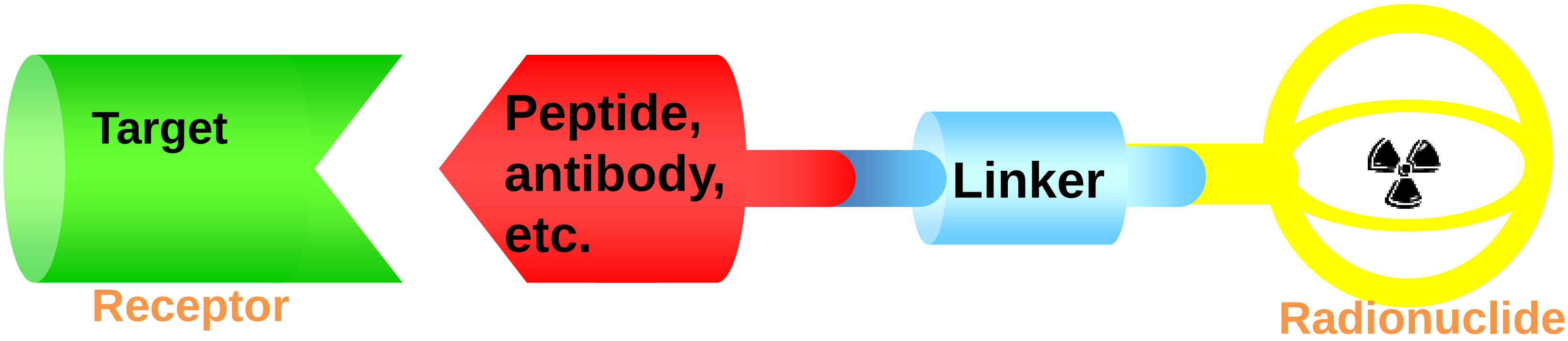


“exotic” isotopes

Immunology approach



Multidisciplinary collaboration to fight cancer



Immunology
Structural biology

Coordination chemistry

Nuclear physics and radiochemistry

Radionuclides for therapy

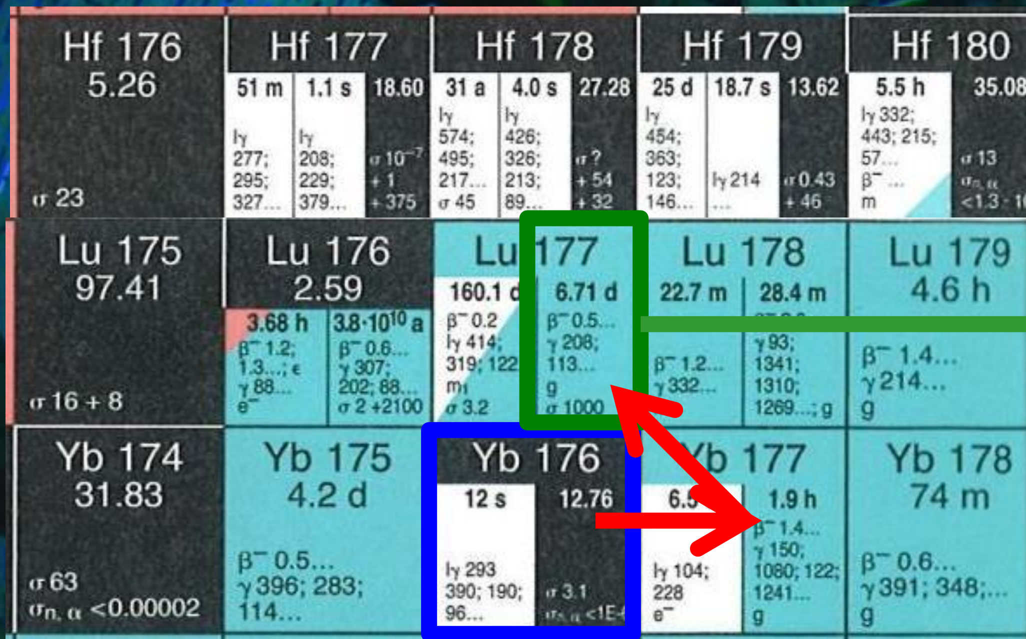
Radio-nuclide	Half-life	E mean (keV)	Eγ (B.R.) (keV)	Range
Y-90	64 h	934 β	-	**12 mm**
I-131	8 days	182 β	364 (82%)	**3 mm**
Lu-177	7 days	134 β	208 (10%) 113 (6%)	**2 mm**

Established isotopes

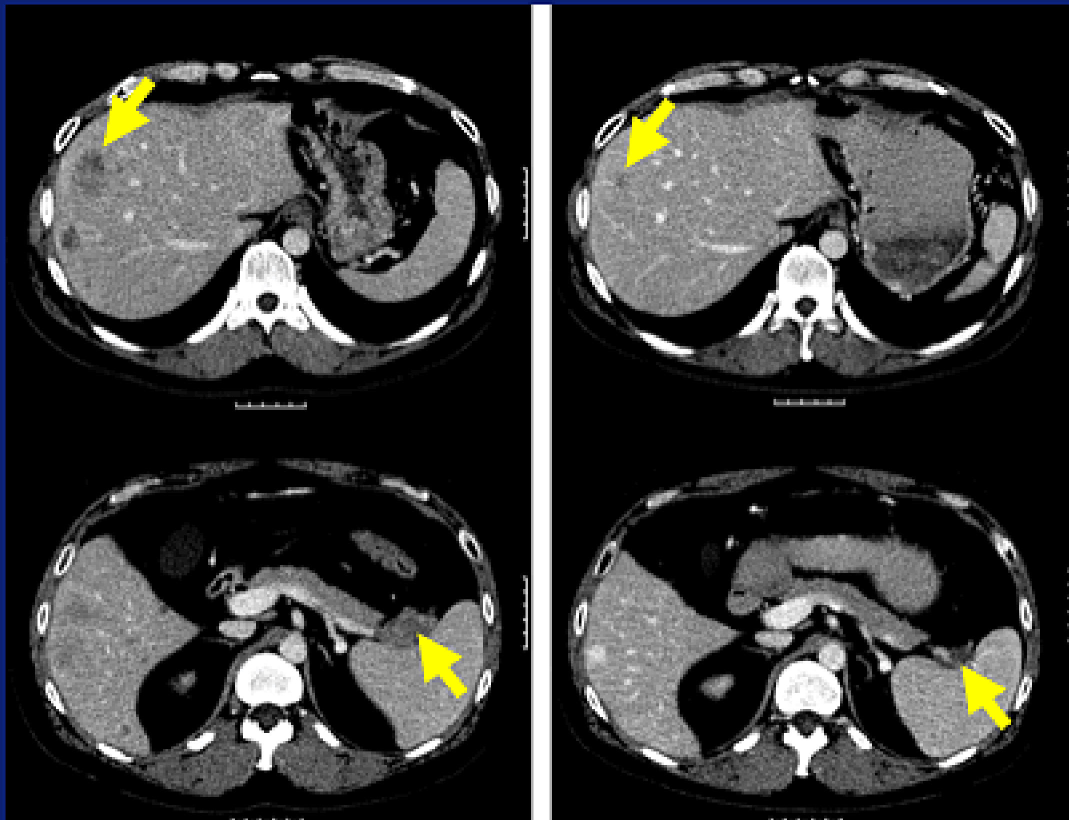
Emerging isotopes

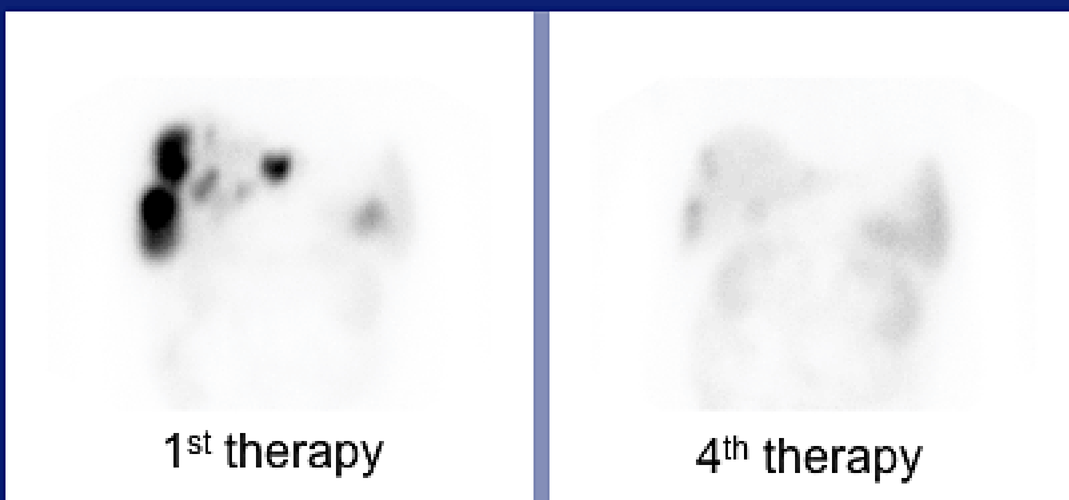
localized radiation

Production of non-carrier-added ^{177}Lu



Irradiation in high flux reactor (e.g. ILL Grenoble), then chemical separation of ^{177}Lu from stable Yb.





Male

36 years of age

Small cell pancreatic neuroendocrine tumour

Liver metastases

Ki-67 index 10-15% (liver biopsy)

4 cycles with ^{177}Lu-octreotate and capecitabine

Partial remission

Roelf Valkema, EANM-2008.

Radionuclides for therapy

Radio-nuclide	Half-life	E mean (keV)	Eγ (B.R.) (keV)	Range
Y-90	64 h	934 β	-	12 mm
I-131	8 days	182 β	364 (82%)	3 mm
Lu-177	7 days	134 β	208 (10%) 113 (6%)	2 mm
Tb-161	7 days	154 β 5, 17, 40 e^-	75 (10%)	2 mm 1-30 µm
Tb-149	4.1 h	3967 α	165,..	25 µm
Ge-71	11 days	8 e^-	-	1.7 µm
Er-165	10.3 h	5.3 e^-	-	0.6 µm

cross-fire

Estab-lished isotopes

Emerging isotopes

R&D isotopes: supply-limited!

localized

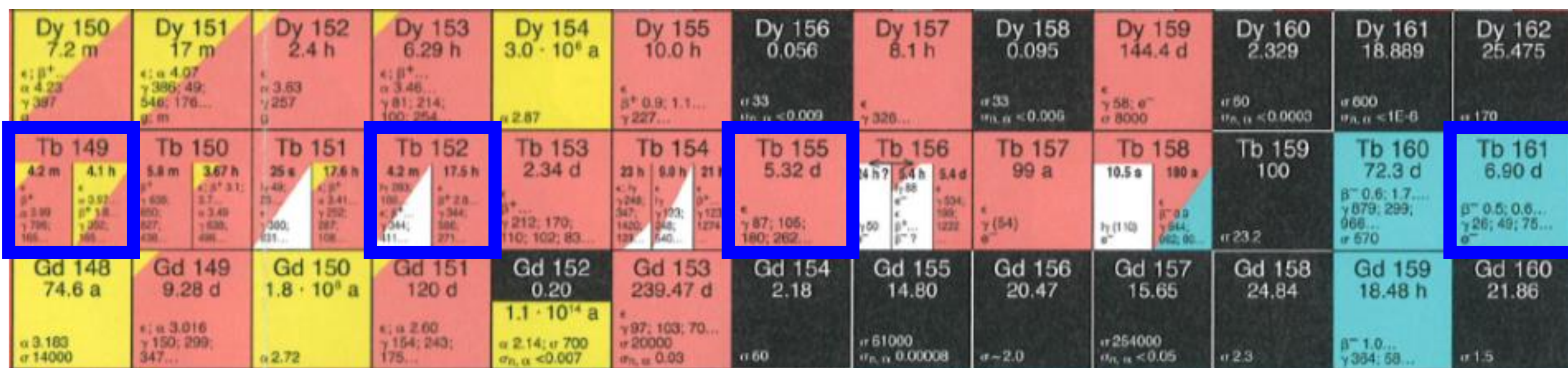
Modern, better targeted vectors require shorter-range radiation

⇒ need for adequate (R&D) radioisotope supply.

Radioisotopes for targeted alpha therapy

Radio-nuclide	Half-life (h)	Advantage	Problem
Tb-149	4.12	Known (bio)chemistry	Availability
Pb-212	10.6	Availability (recycled from U-fuel ^{228}Th)	Release of daughter?
Bi-212	1.01	Known (bio)chemistry Availability	Short half-life
Bi-213	0.76	Known (bio)chemistry	Short half-life Availability
At-211	7.22	Half-life	Challenging (bio)chemistry
Ra-223	274	Half-life Availability	Only for bone metastases (similar chemistry to Ca)
Ac-225	240	Half-life Known (bio)chemistry	Release of daughters? Availability

Terbium: a unique element for nuclear medicine



CERN-MEDICIS:
Medical isotopes collected from ISOLDE

R. Catherall, M. Dias, T. Giles, Z. Lawson, S. Marzari, **T. Stora** (**CERN**)

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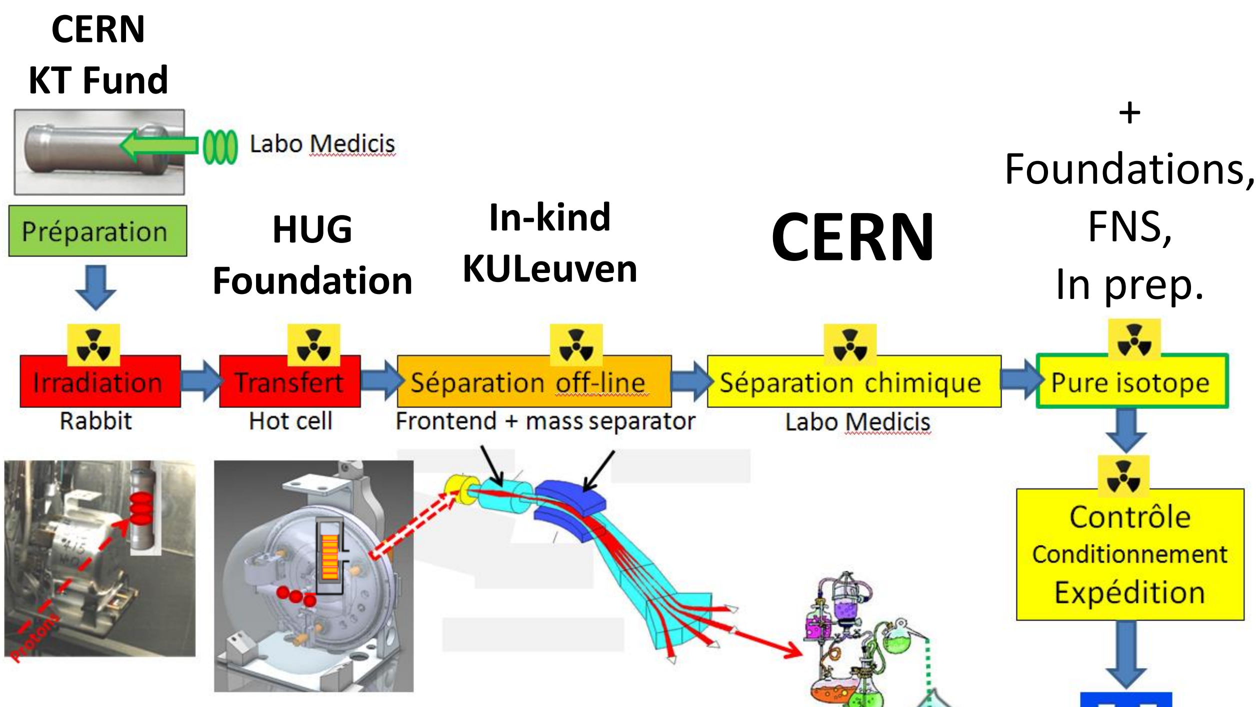
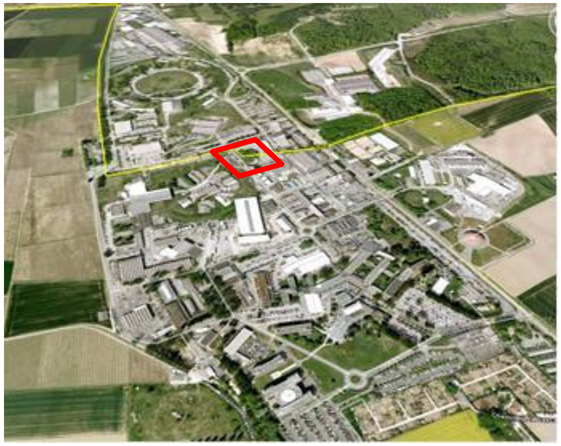
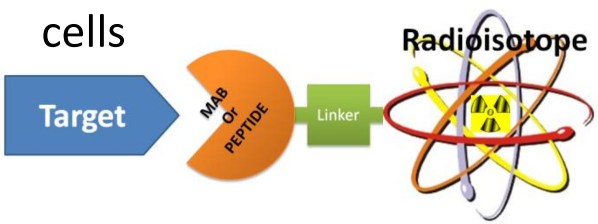


Expected to start end 2015

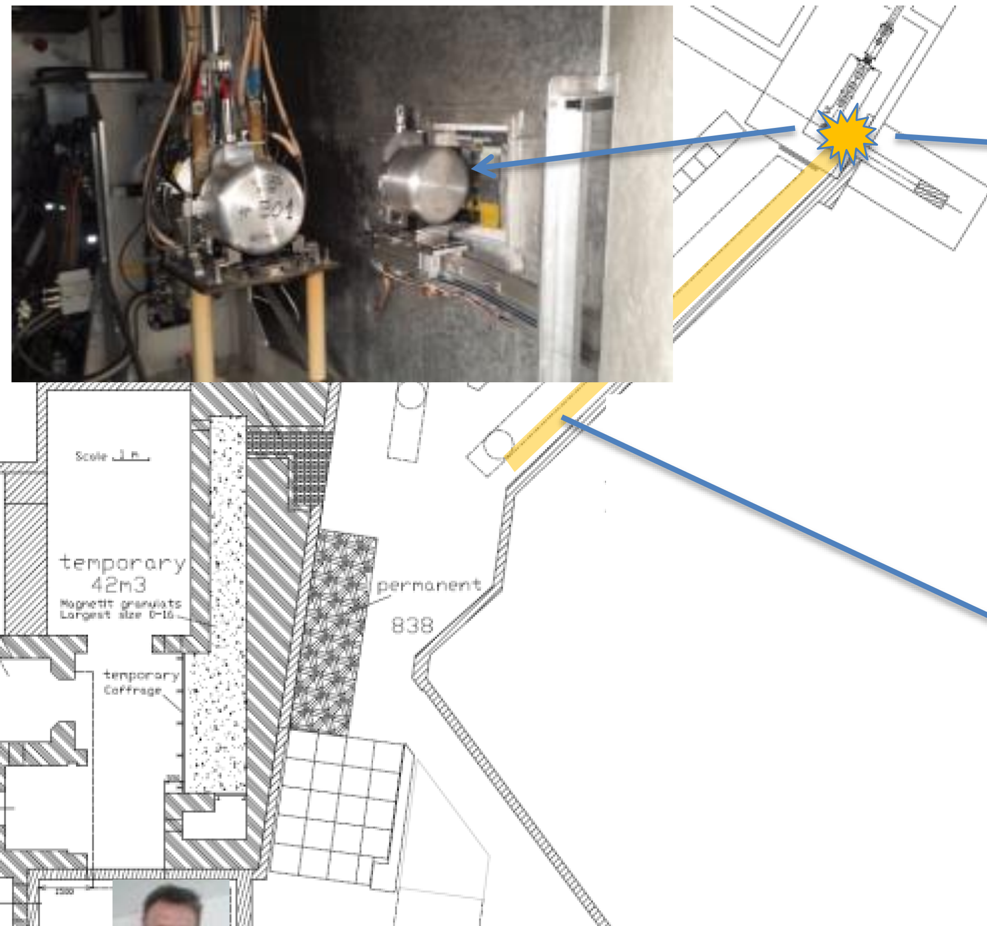


Prostate Cancer cells

152Tb From ISOLDE



Irradiation station commissioned with beam



Tests with protons:
Done successfully !

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The (potential) role of ISOL in nuclear medicine

1. Samples of R&D isotopes which are not commercially available or easily producible by other means.
2. Isotopes with ultimate specific activity for R&D, e.g. studies of efficacy versus specific activity.
3. Isotopes that are best produced by spallation (^{149}Tb,…).

Existing ISOL beams are sufficiently intense for preclinical studies, in certain cases even for clinical studies.

How to organize R&D with RIBs in nuclear medicine?
Physicists are used to “travel to the isotopes”,
but isotopes must “travel to physicians and patients”.

CONCLUSIONS

"SSP" of EXOCTIC radioactive isotopes

- Specific areas are identified:
  Life Sciences Materials Nuclear Chemistry...

- The methods follow the needs with progressing quality...

- Viability and Visibility of "Applications"
  depend at the long term from diversifying and optimizing RIB infrastructures with:
  Dedicated BEAM TIME and BEAM LINES
  Dedicated LABORATORY SPACE

...the future of "Applications" depend very much on the concept of the next generation of RIB facilities ....

Acknowledgments

To the WEMS 2016 organizers !

ISOLDE Solid State Physics

SPONSORED BY THE

Federal Ministry of Education and Research

KATHOLIEKE UNIVERSITEIT LEUVEN

FCT Fundação para a Ciência e a Tecnologia

MINISTÉRIO DA CIÊNCIA E DA TECNOLOGIA

... to colleagues and collaborators!

- R. Vianden
- D. C. Lupascu
- M. Deicher
- A. Fenta
- H. Gunnlaugsson
- T. E. Molholt
- K. Johnston
- L. B. S. Hemmingsen
- U. Köster
- M. Kowaslka
- A. L. Lopes
- J.G.M. Correia
- M. Stachura
- T. Stora
- U. Wahl
- *and The ISOLDE collaboration*