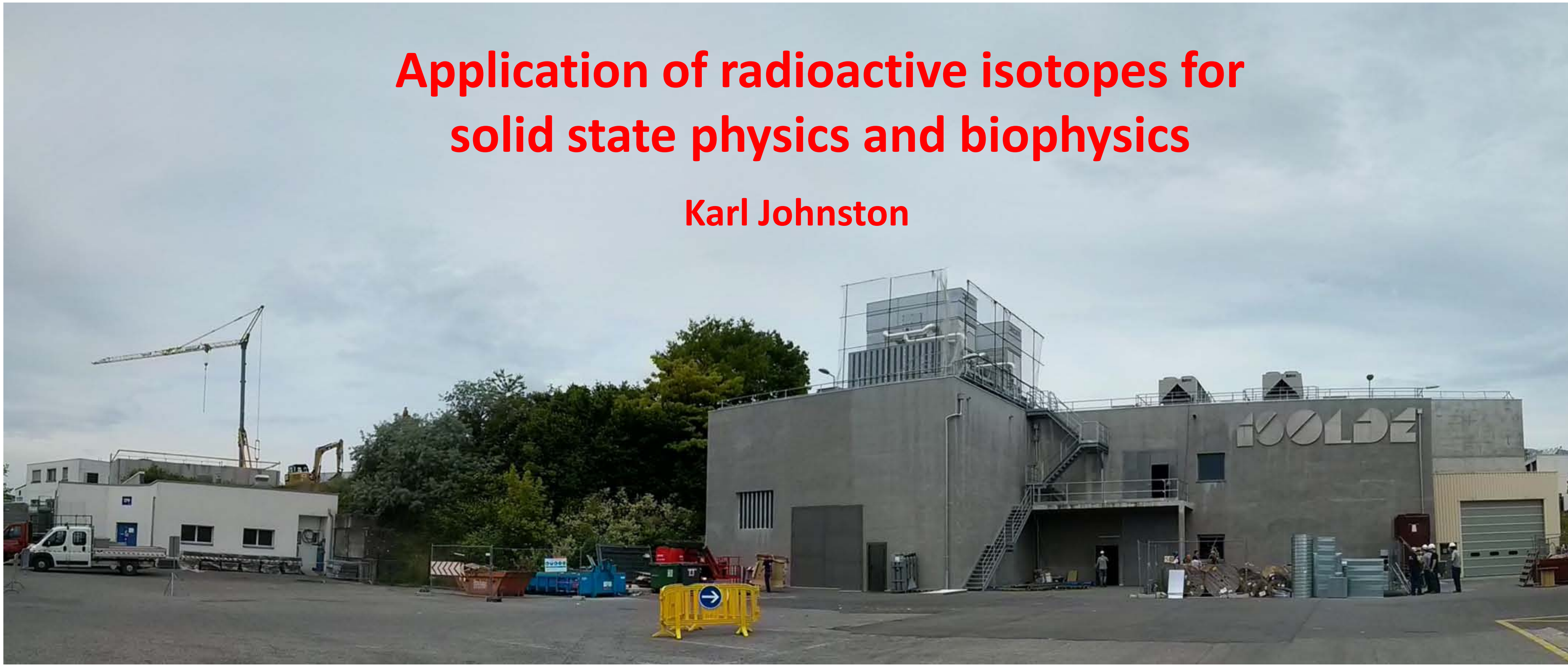
Application of radioactive isotopes for solid state physics and biophysics

Karl Johnston

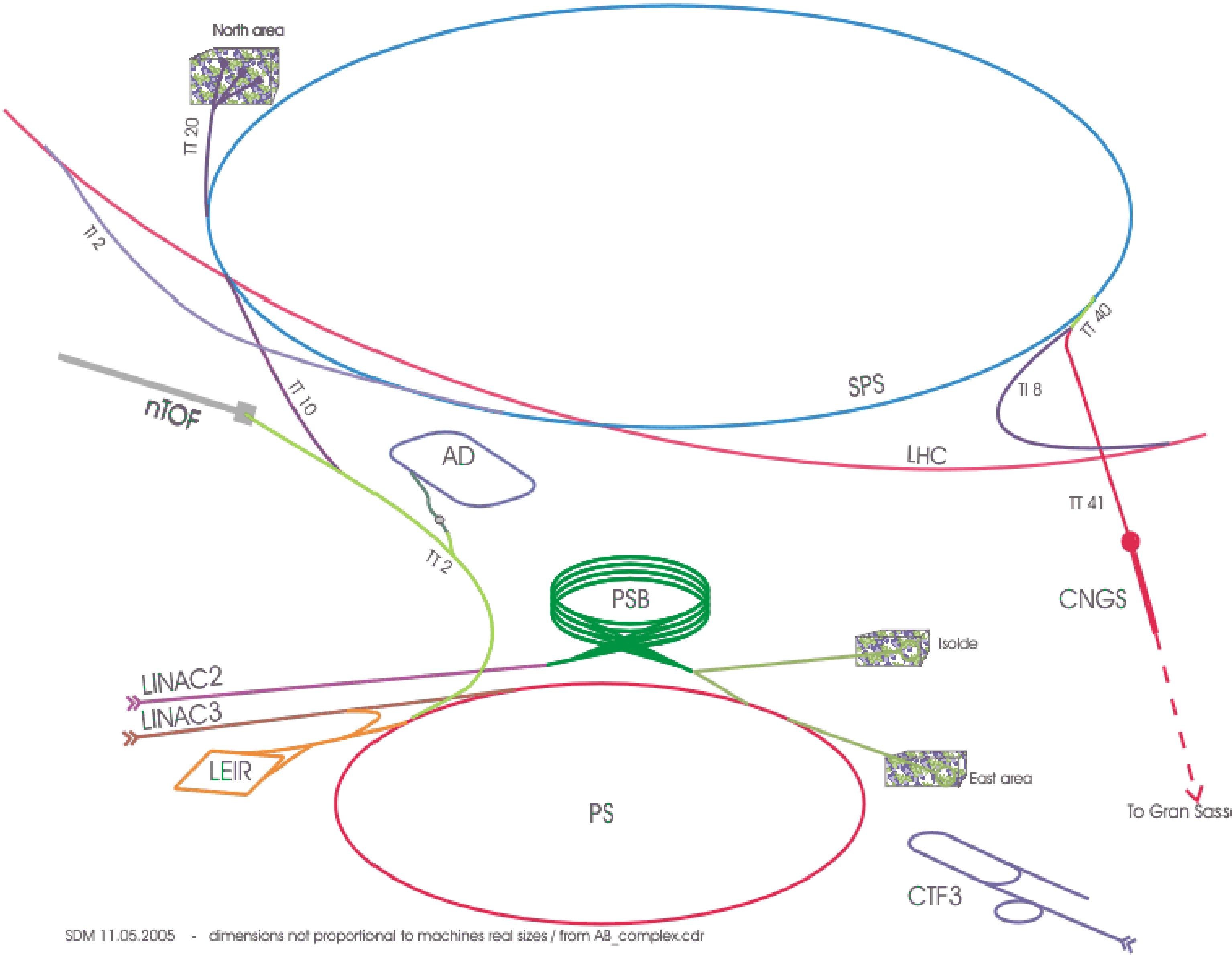


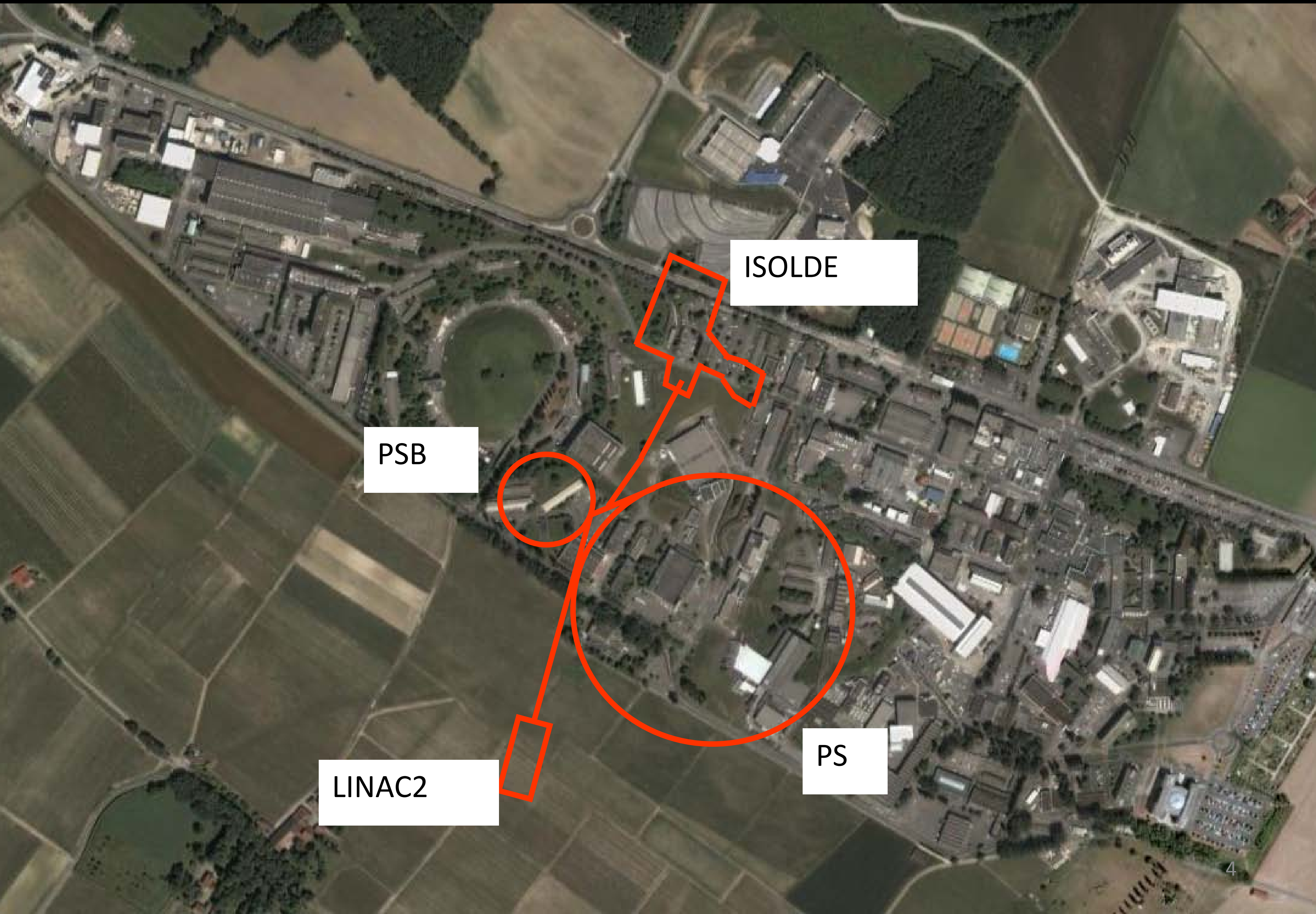
YEARS/ANS CERN

Overview

- (Very brief) Introduction to ISOLDE…
- (even shorter) “flash” of some recent nuclear physics highlights.
- “Applied” programme at ISOLDE: Solid state physics, biophysics.
- Techniques and materials:
    - Radiotracer Diffusion
    - Optical studies in semiconductors
    - Emission Channelling
    - Mossbauer spectroscopy using 57Mn/57Fe
    - Perturbed angular correlation and β-NMR for biophysics
    - MEDICIS

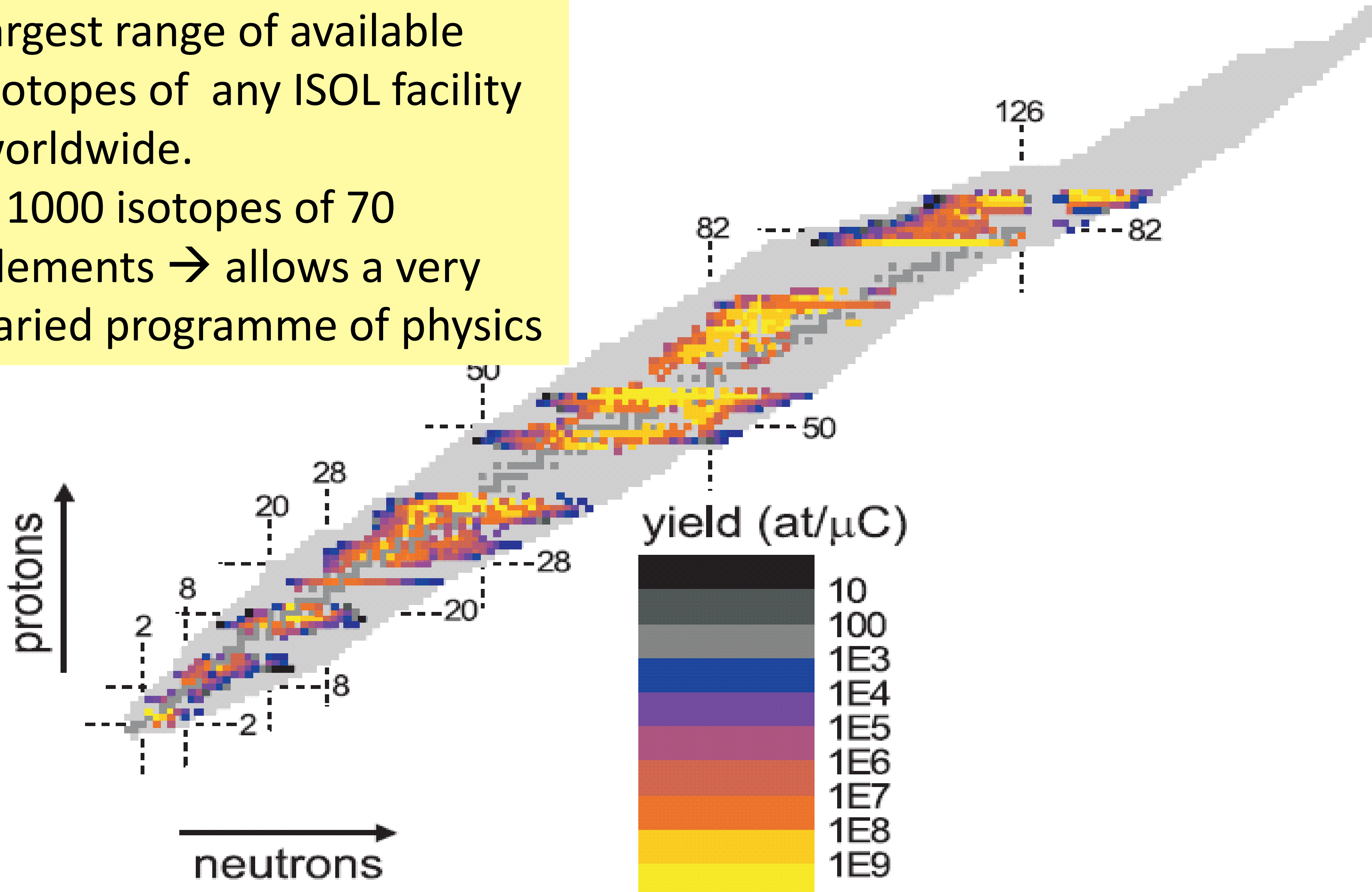
Accelerators at CERN



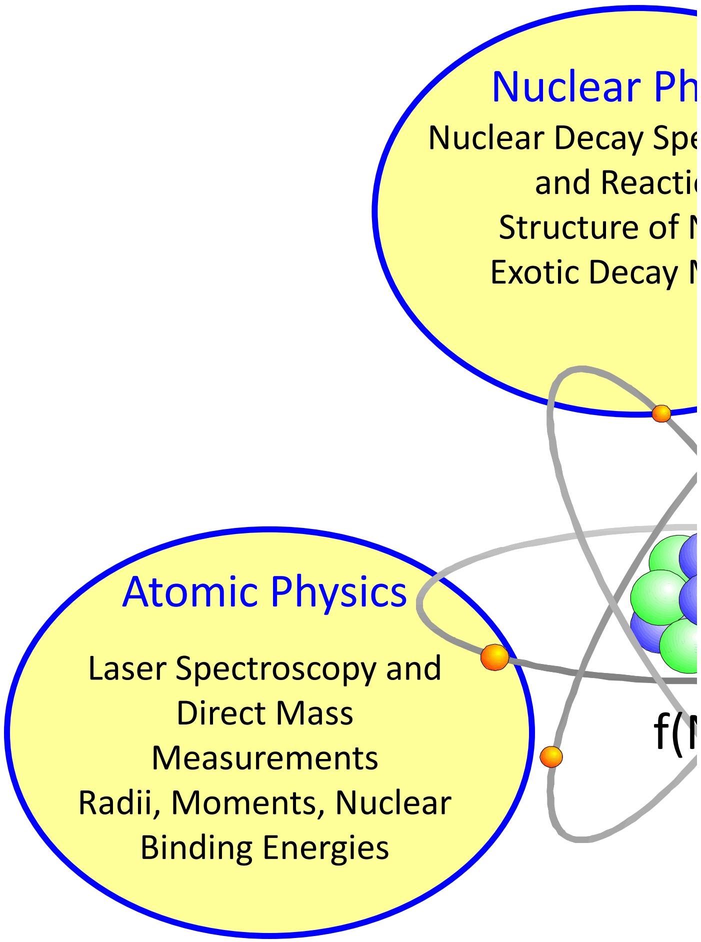

ISOLDE
PSB
PS
LINAC2

ISOLDE / 50 y Experience

ISOLDE today offers the largest range of available isotopes of any ISOL facility worldwide.
> 1000 isotopes of 70 elements → allows a very varied programme of physics



ISOLDE physics



ARTICLE

doi:10.1038/nature12073

Studies of pear-shaped nuclei using accelerated radioactive beams

L. P. Gaffney[1], P. A. Butler[1], M. Scheck[1,2], A. B. Hayes[3], F. Wenander[4], M. Albers[5], B. Bastin[6], C. Bauer[2], A. Blazhev[5], S. Bönig[2], N. Bree[7], J. Cederkäll[8], T. Chupp[9], D. Cline[3], T. E. Cocolios[4], T. Davinson[10], H. De Witte[7], J. Diriken[7,11], T. Grahn[12], A. Herzan[12], M. Huyse[7], D. G. Jenkins[13], D. T. Joss[1], N. Kesteloot[7,11], J. Konki[12], M. Kowalczyk[14], Th. Kröll[2], E. Kwan[15], R. Lutter[16], K. Moschner[5],

LETTER

doi:10.1038/nature12226

Masses of exotic calcium isotopes pin down nuclear forces

F. Wienholtz[1], D. Beck[2], K. Blaum[3], Ch. Borgmann[3], M. Breitenfeldt[4], R. B. Cakirli[3,5], S. George[1], F. Herfurth[2], J. D. Holt[6,7], M. Kowalska[8], S. Kreim[3,8], D. Lunney[9], V. Manea[9], J. Menéndez[6,7], D. Neidherr[2], M. Rosenbusch[1], L. Schweikhard[1], A. Schwenk[7,6], J. Simonis[6,7], J. Stanja[10], R. N. Wolf[1] & K. Zuber[10]

nature COMMUNICATIONS

ARTICLE

Received 21 Aug 2012 | Accepted 27 Mar 2013 | Published 14 May 2013
DOI: 10.1038/ncomms2819 OPEN

Measurement of the first ionization potential of astatine by laser ionization spectroscopy

Radioactive isotopes for solid state physics? Why & How??

Radiation: very easy to detect……..

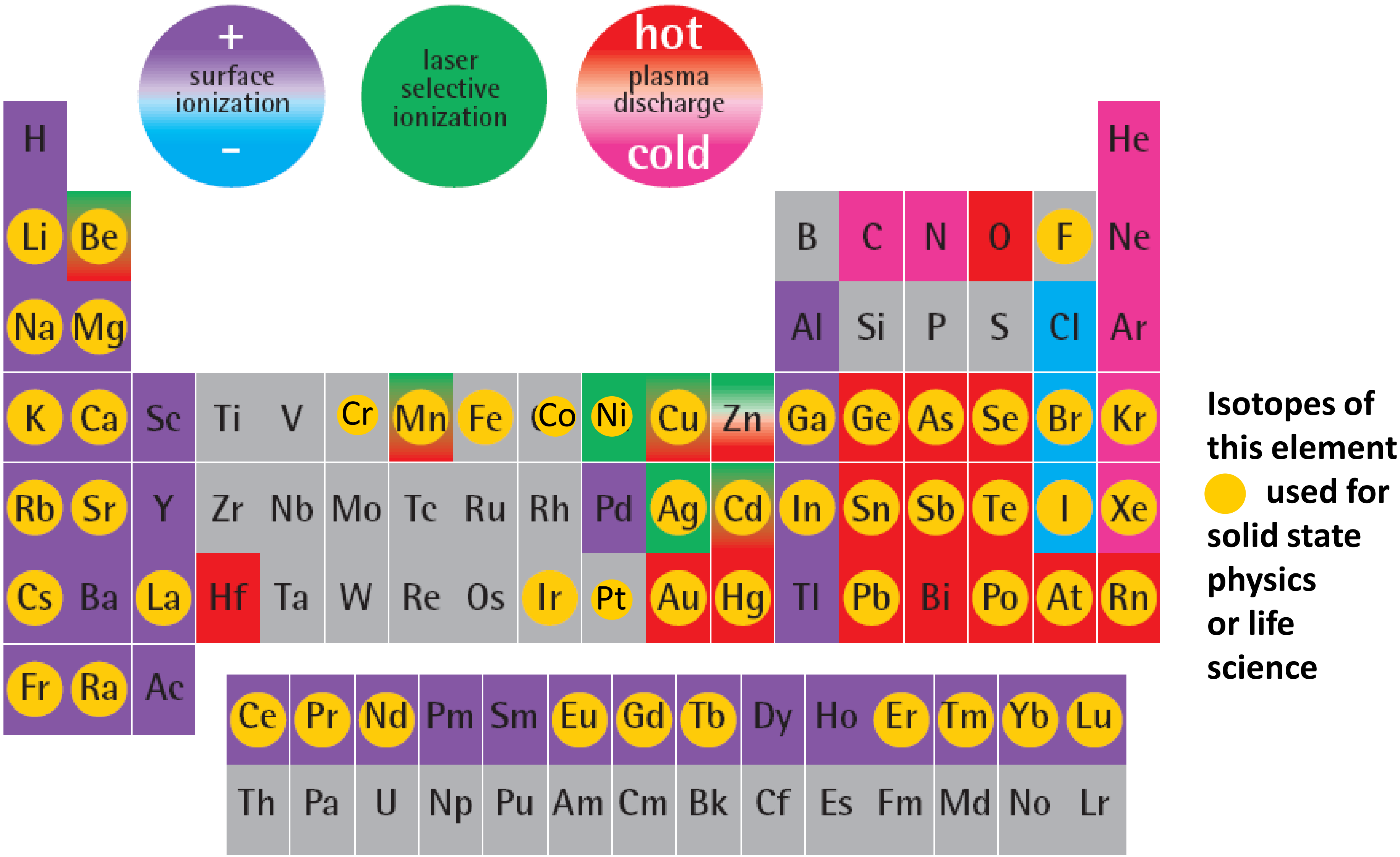
The radioactive isotopes ("probes") act as "spies" transmitting information with **atomic** resolution via their decay.

- What information do you get?????
  - Adds extra dimension to "normal spectroscopies" e.g. optical / electrical characterisation of semiconductors
  - Provides extremely **local probe** which can be applied to variety of systems

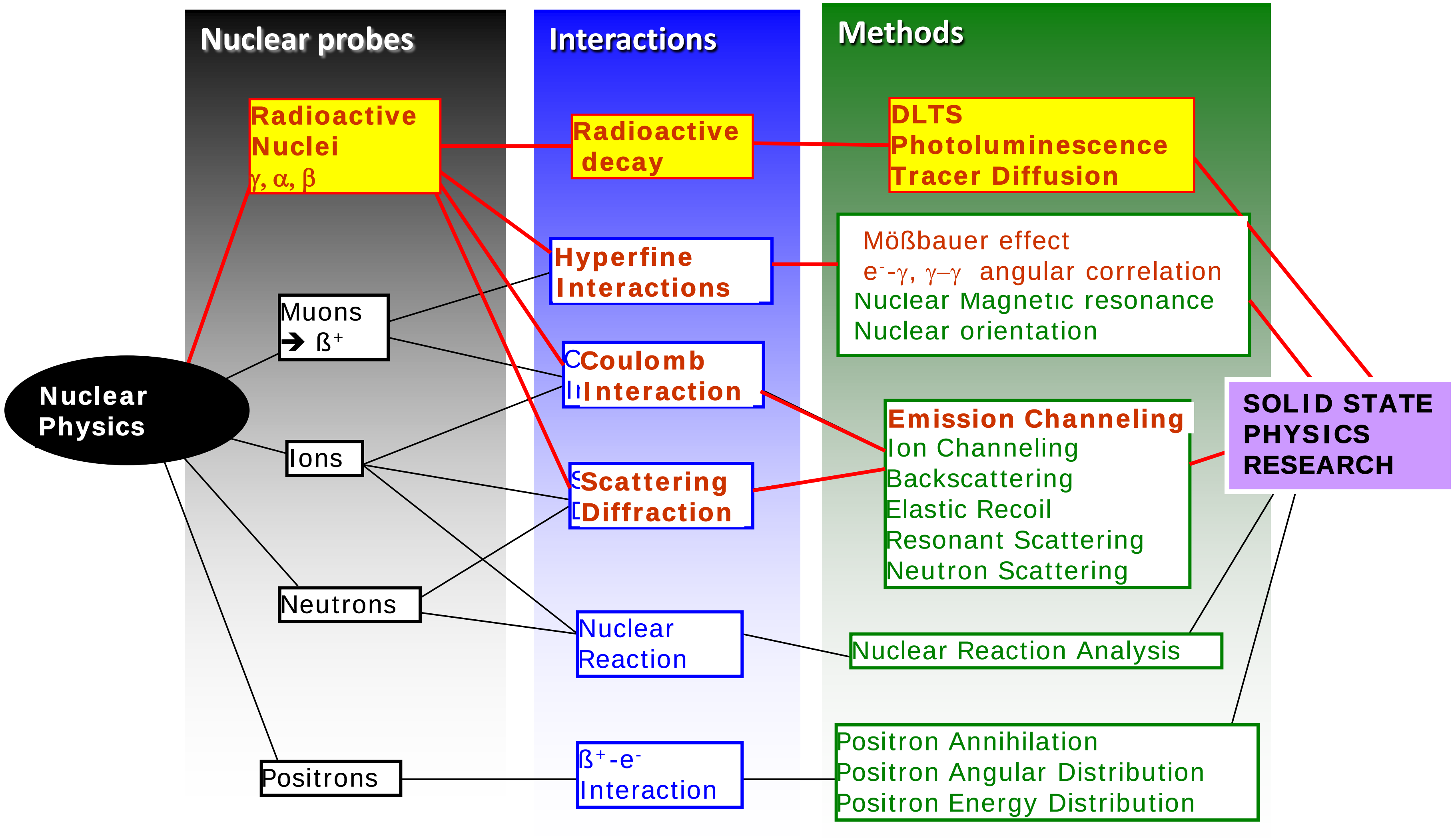
You need

A facility providing a large variety of radioactive isotope (if possible, isotopically clean)

ISOLDE table of elements

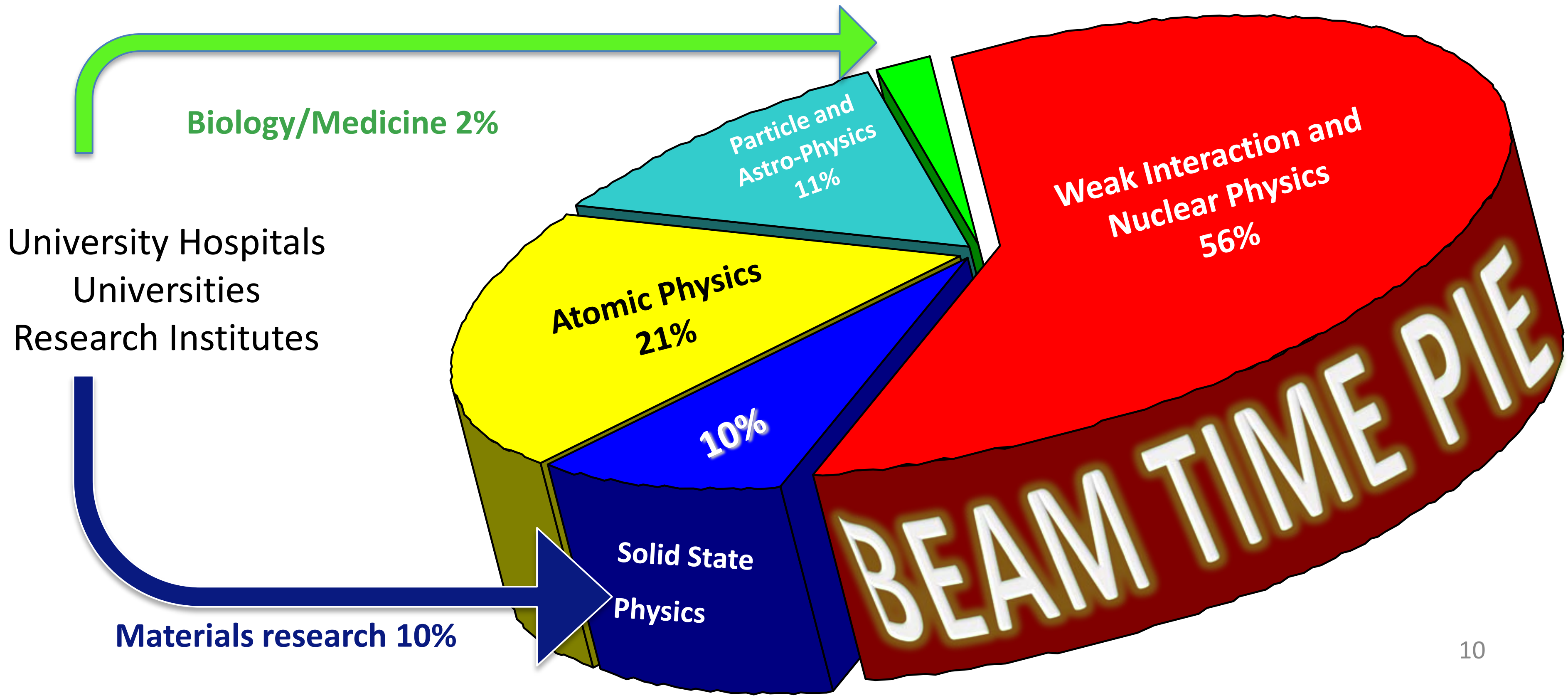


Applying radioactivity to solid state physics



“APPLICATIONS”

- Intense beams
- Clean beams
- Ability to share time with more exotic nuclear physics: running in parallel

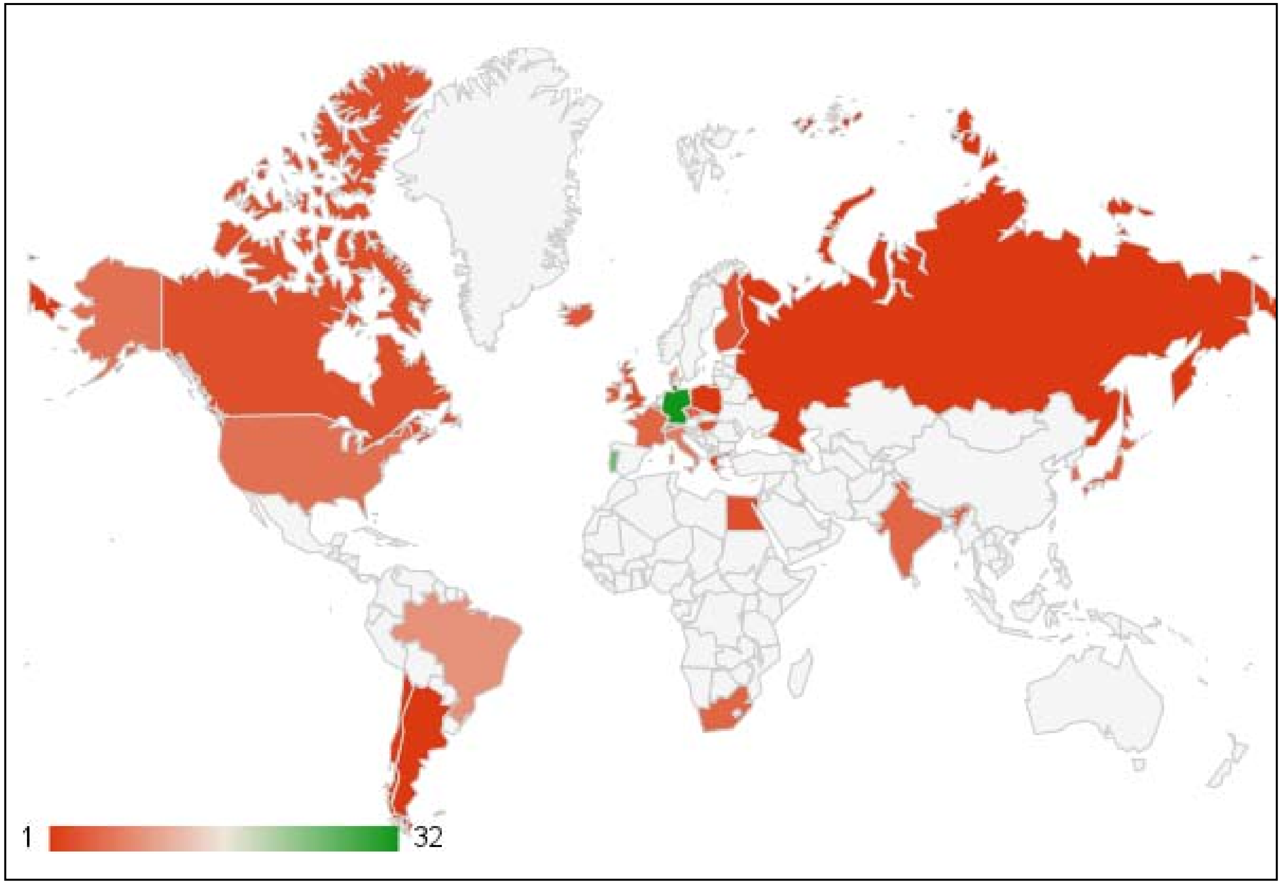


Solid state physics @ ISOLDE: Diverse community

Solid State physics:
Semiconductors,
Metals

Materials scientists:
High Tc
Superconductors,
Multiferroic materials

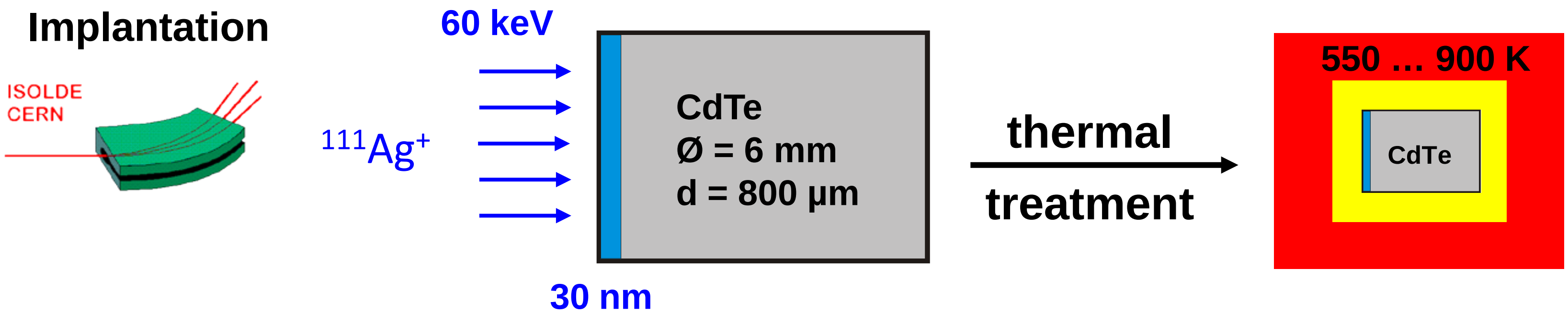
Biophysics:
structural properties &
role of heavy metals in
proteins, DNA.

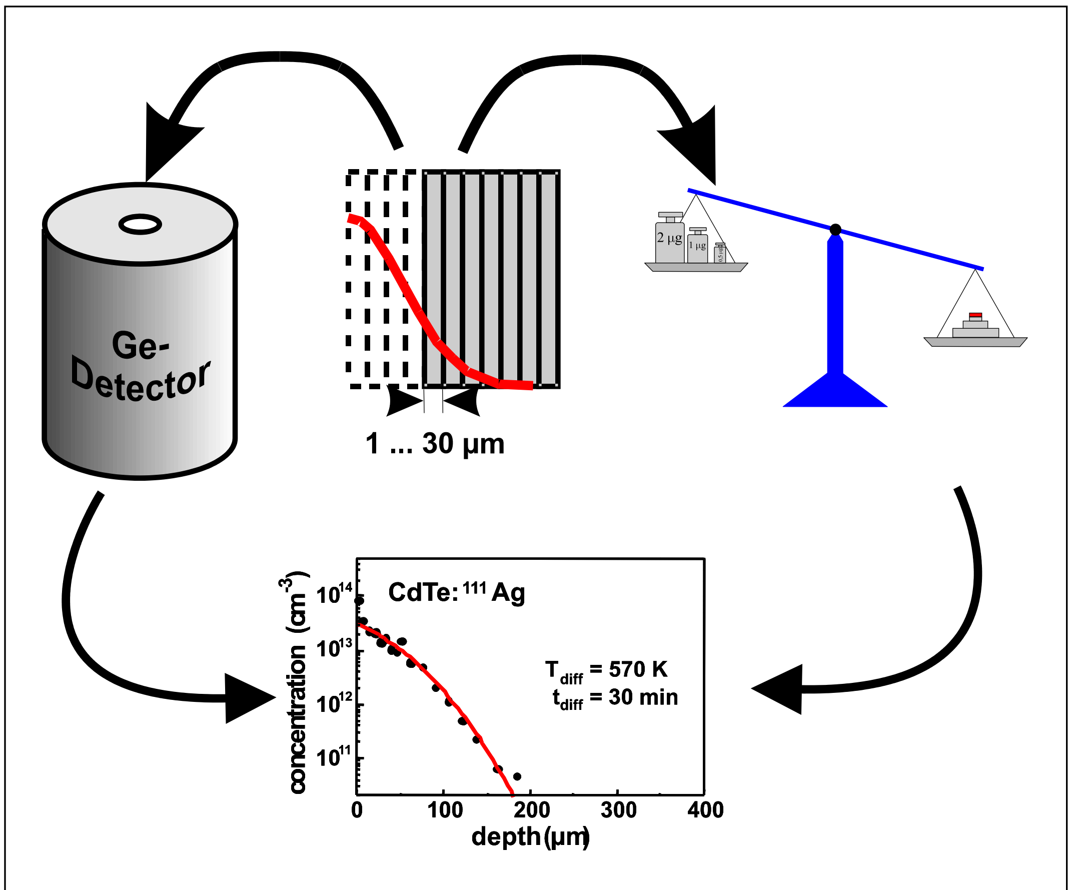


SSP collaborations

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

Radiotracer technique





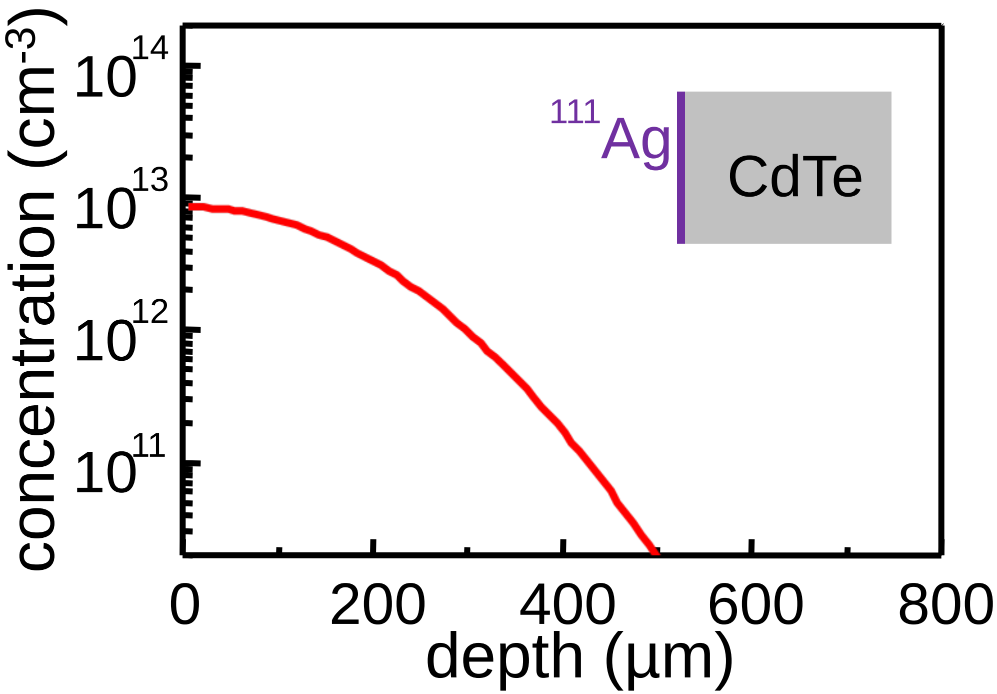
Mechanical sectioning

- Resolution > 1 µm
- $T_{1/2}$ > 1 h

12

Diffusion in solids

$$J(X) = -D_X \frac{\partial [X]}{\partial x} \qquad \frac{\partial [X]}{\partial t} = -\frac{\partial}{\partial x} J(X)$$

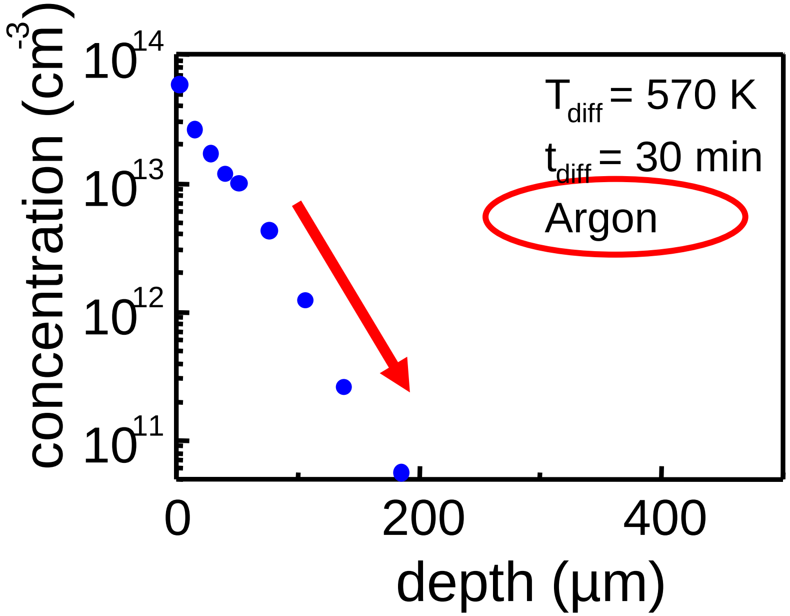


for $C(x,t=0) = N \cdot \delta(x)$

$$C(x,t) = \frac{N}{\sqrt{2\pi \cdot Dt}} \cdot e^{-\frac{x^2}{4Dt}}$$

Monotonously decreasing profiles

Diffusion of ^{111}Ag in CdTe



Monotonously decreasing profile

Uphill diffusion profile

(H. Wolf et al., Phys. Rev. Lett. 94 (2005) 125901)

Model

- Deviation from stoichiometry

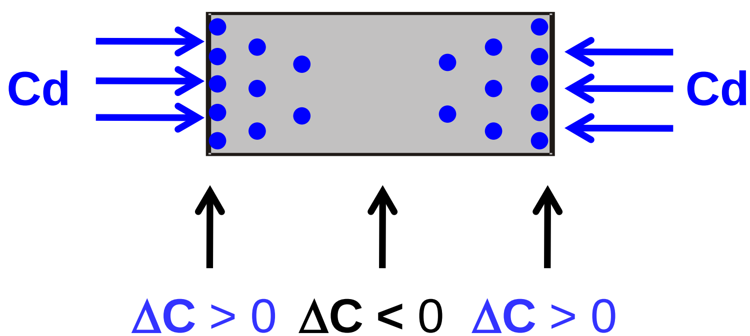
$$[\Delta C]=[Cd_i]-[V_{Cd}]$$

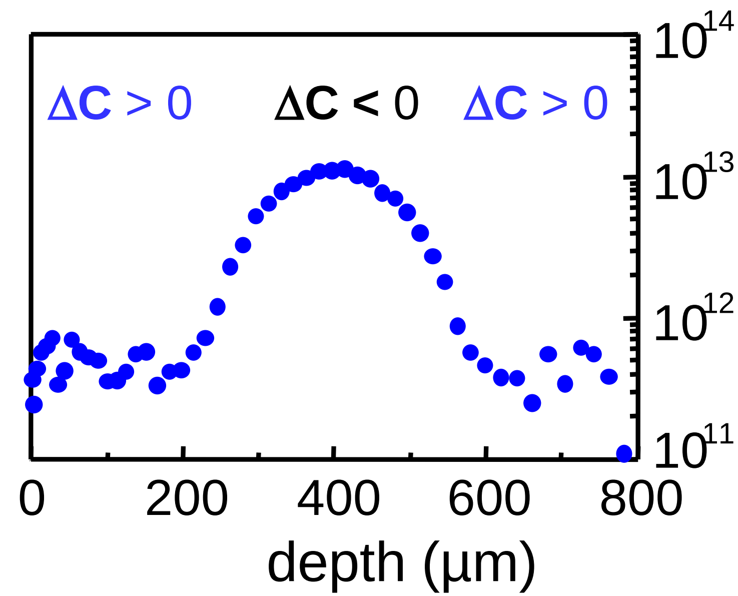
- Initial state of the crystal Te rich

ΔC < 0

- Highly mobile Cd interstitials
- Highly mobile Ag dopant (interstitial)

- Source of Cd interstitials during diffusion

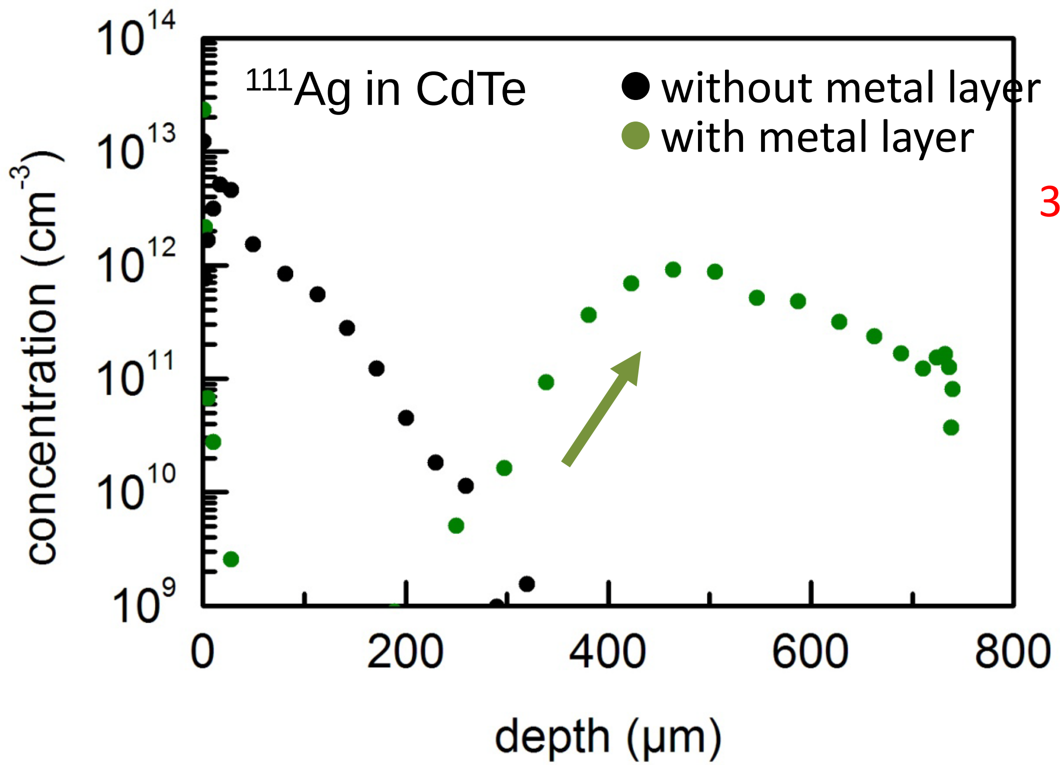


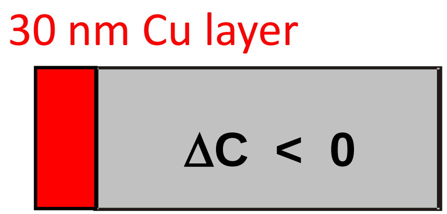


- Ag dopants images the profile of the intrinsic defects

Also seen from metal layers (Cu)

Diffusion without Cu layer at 550 K
Cu layer at 500 K





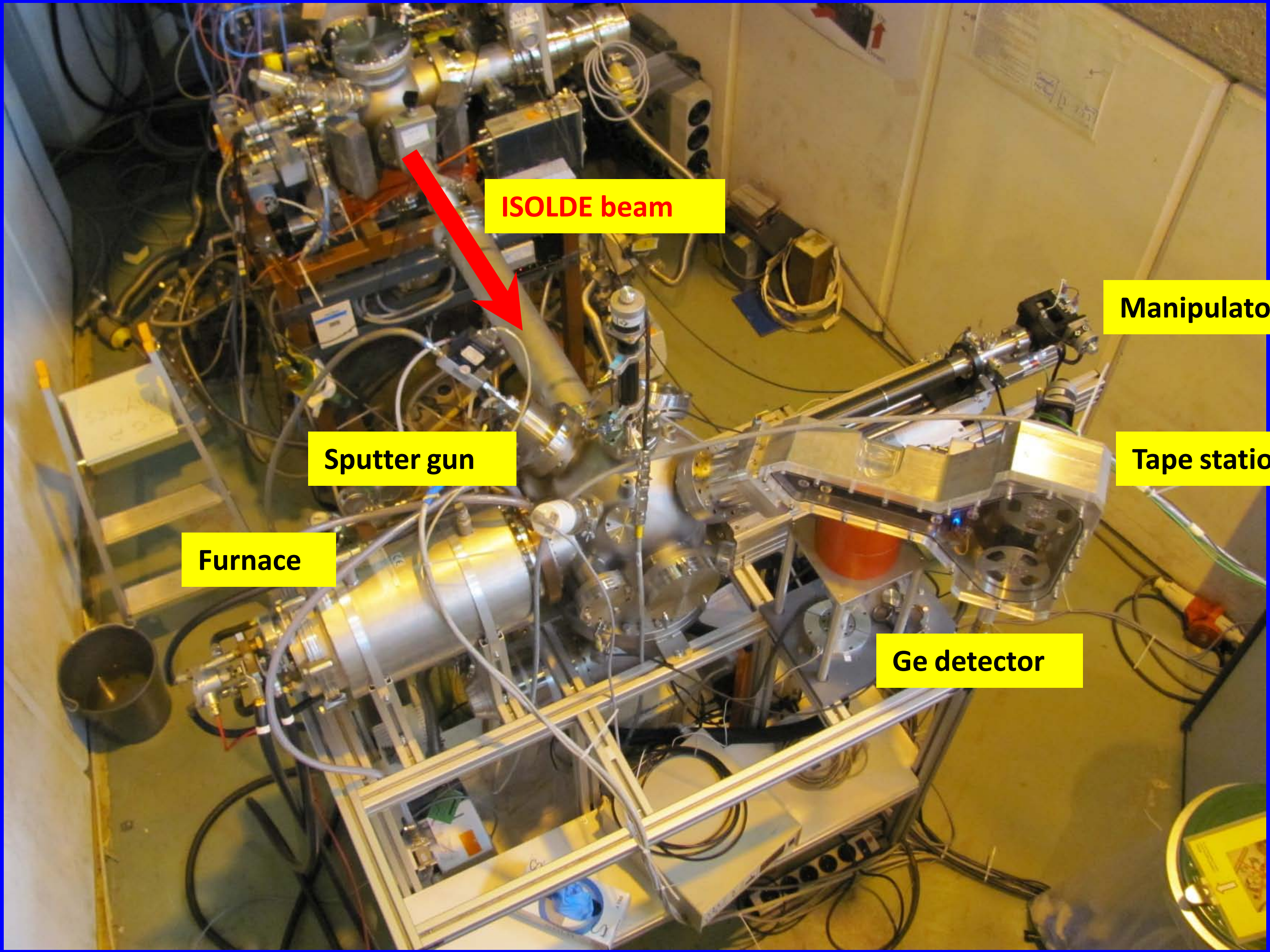
Metal layers can initialize uphill diffusion

(H. Wolf et al., Appl. Phys. Lett. 100 (2012) 171915)

Consequences for contacts to semiconductors?

On-line diffusion chamber at ISOLDE

2012

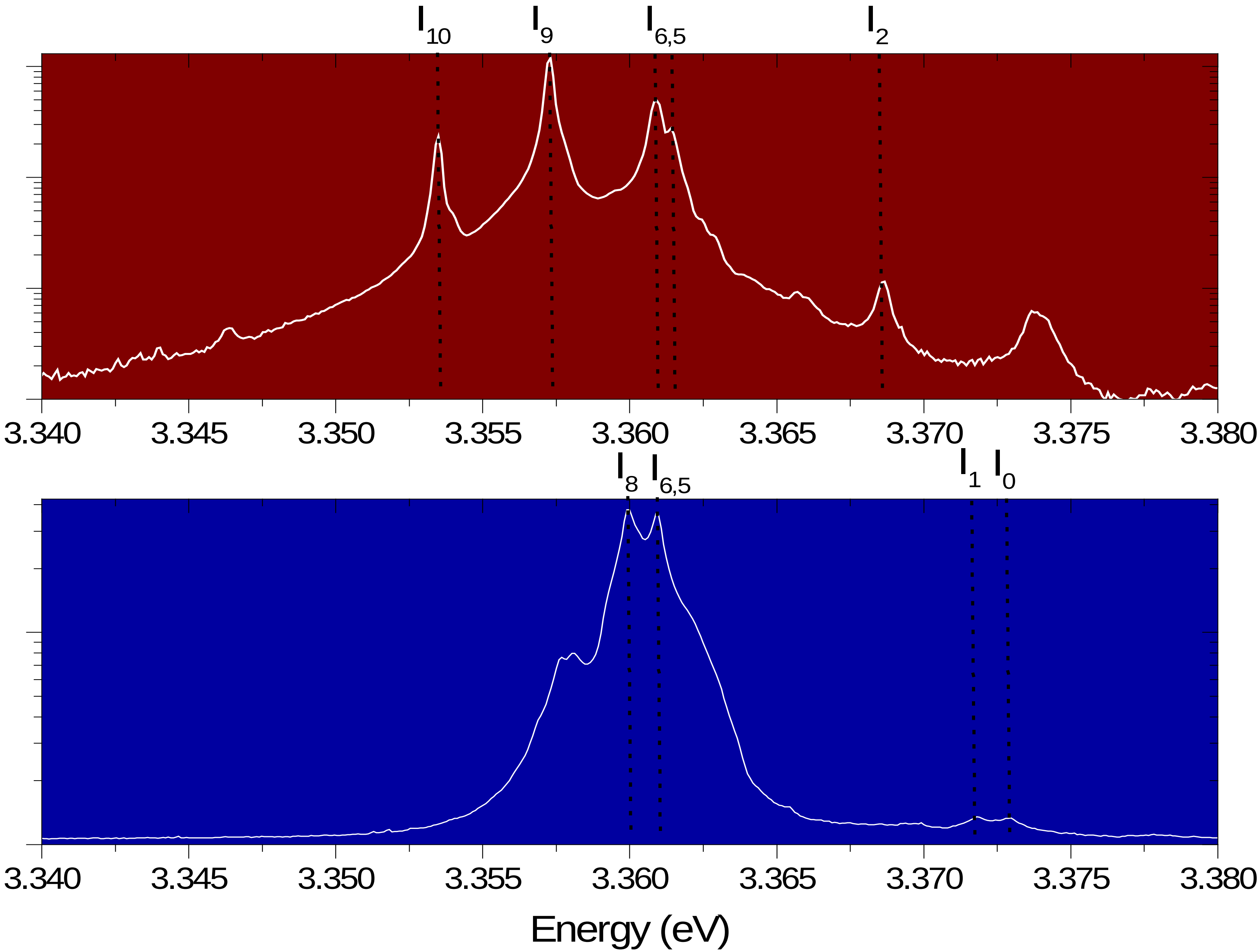


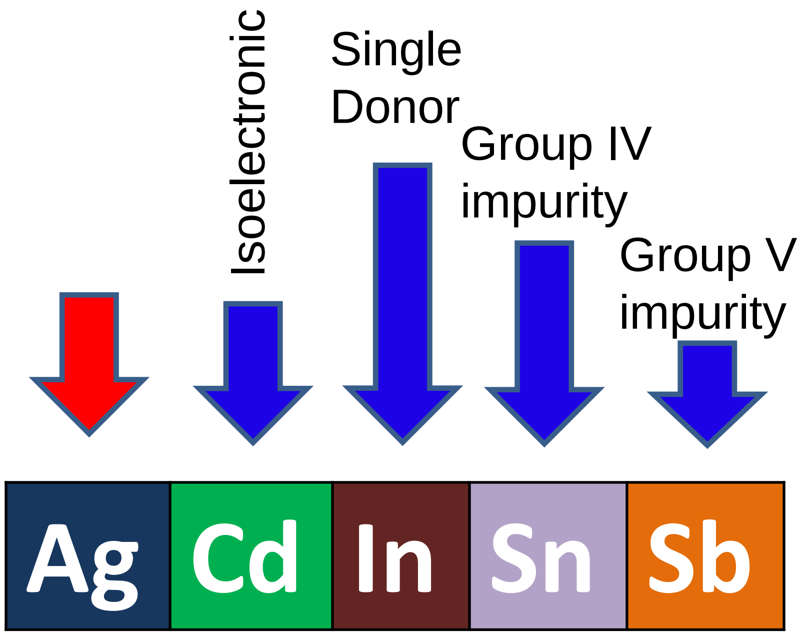
Photoluminescence Characterisation of Semiconductors

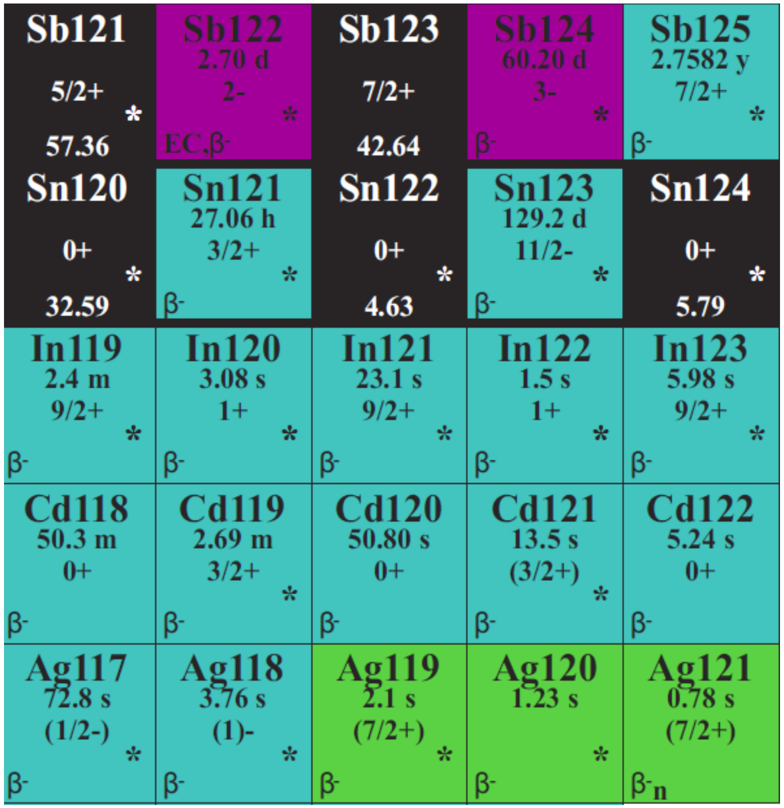
- ✓ One of the principal techniques in semiconductor research.
- ✓ High Spectral resolution
- ✓ Non-destructive
- ✓ No need for contacts
- ✓ Very sensitive (can probe ppb)
- ✓ Relatively flexible and straightforward
- ✓ Can be extended to include external perturbations.

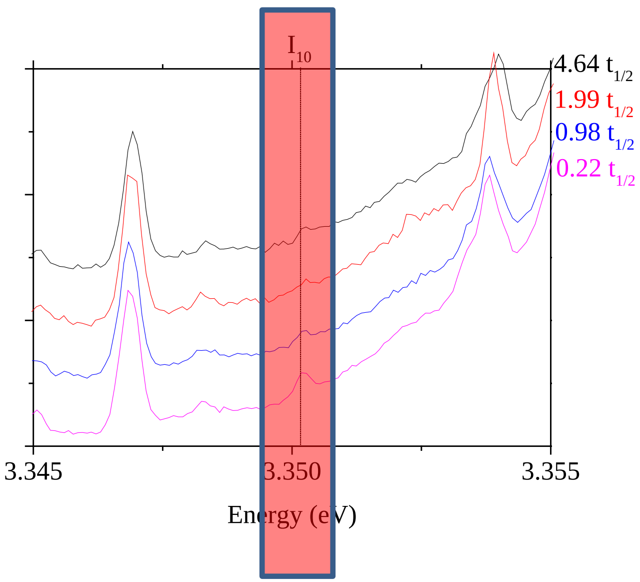
X Not quantitative
X Low concentrations may be more optically efficient
X **Lack of chemical information (except for some rare cases where isotope shifts are observed)**

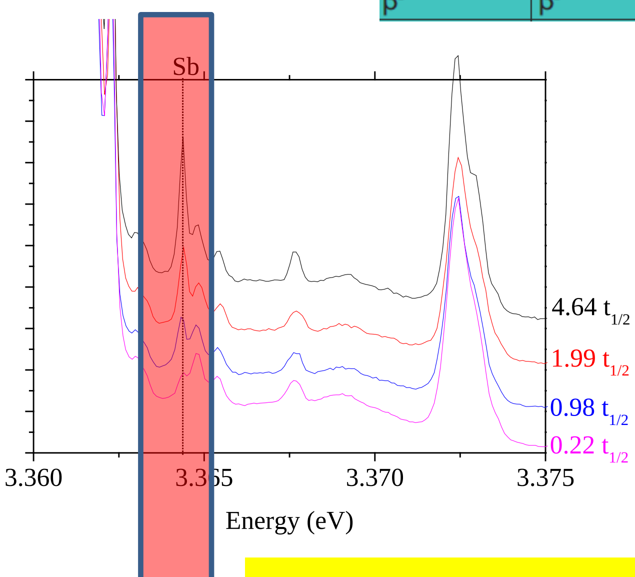
Optical Spectroscopy of Semiconductors: ZnO

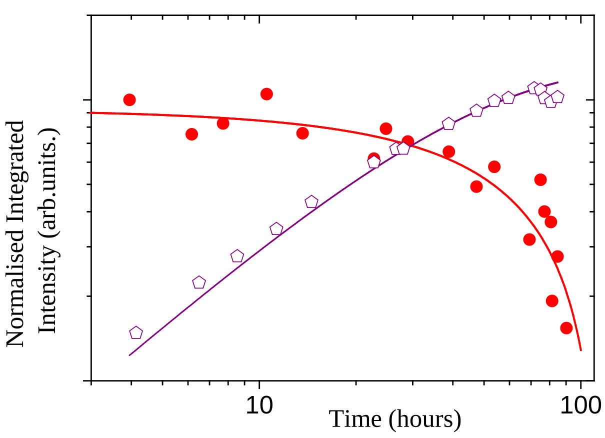






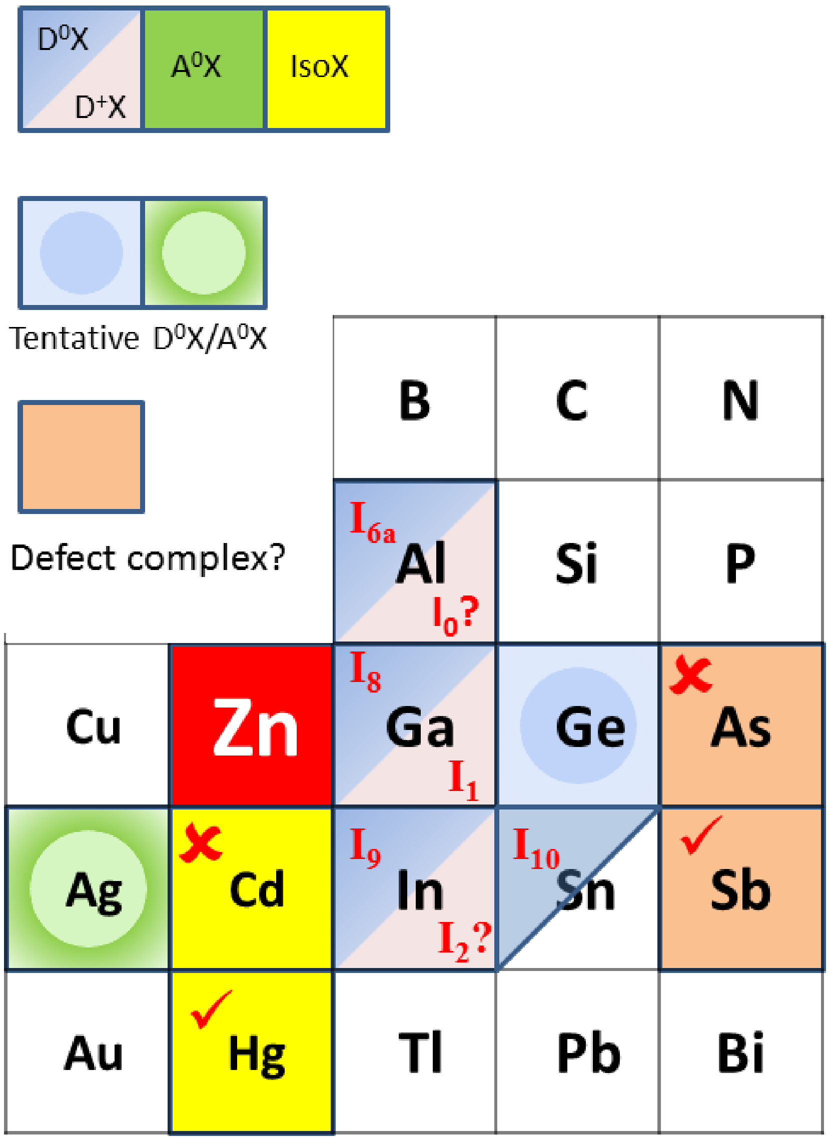






J Cullen *et al* Appl. Phys. Lett. **102** 192110 (2013)

Radiotracer PL has allowed for the full classification of the dominant impurities in ZnO

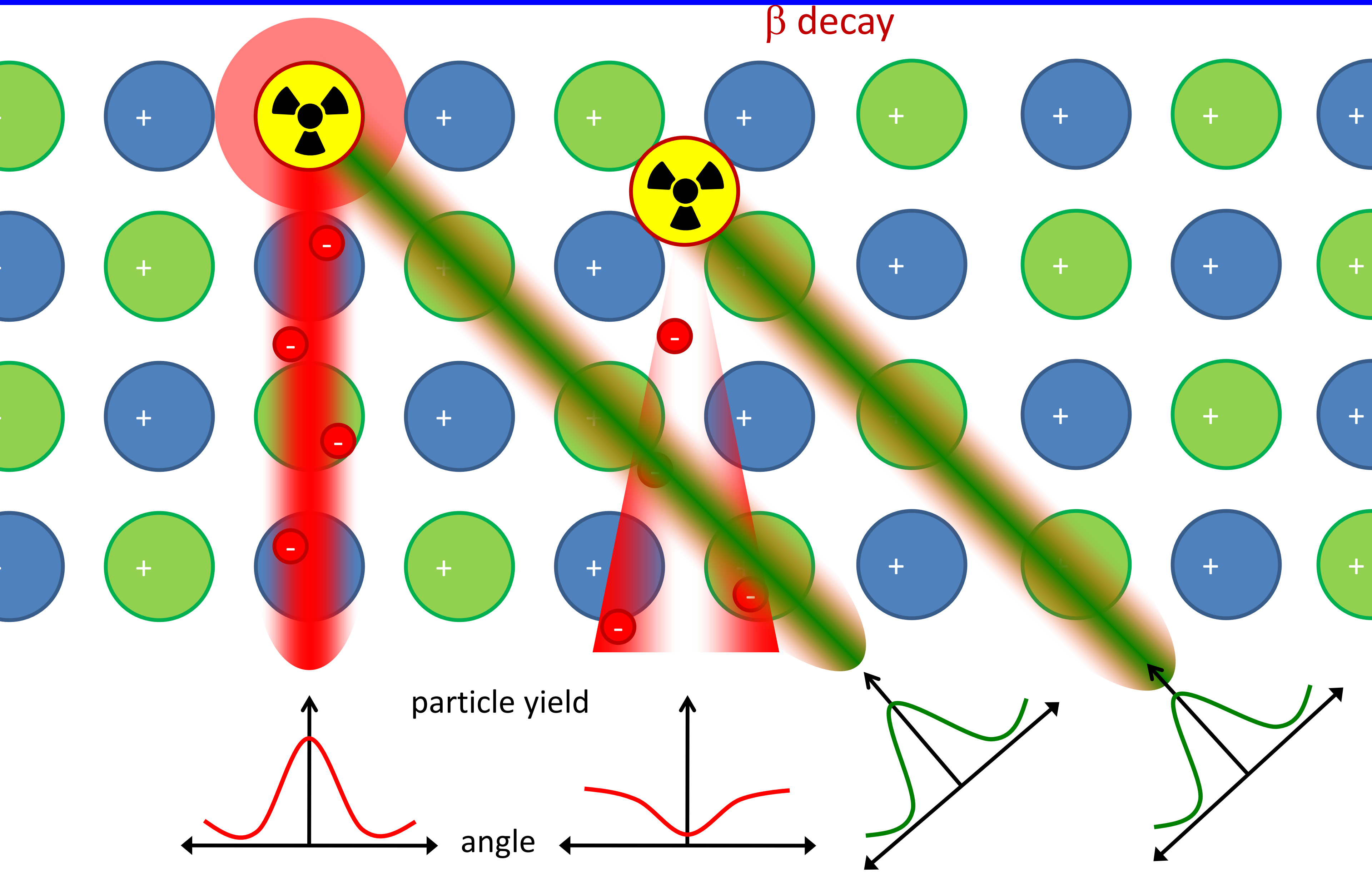


- Radio PL allows for the subtle chemical identification of luminescence through different decay chains.

- Has allowed for the identification of neutral and ionised donors [1, 2], complexed impurities [3], “double donor” centres [2, 4], and isoelectronic centres [5].

1. K. Johnston *et al* Phys Rev B **73** 165212 (2006).
2. K. Johnston *et al* Phys Rev B **83** 125205 (2011).
3. J Cullen *et al* Appl. Phys. Lett. **102** 192110 (2013)
4. J. Cullen et al Phys Rev B **87** 165202 (2013)
5. J. Cullen et al J. Appl Phys (2013)

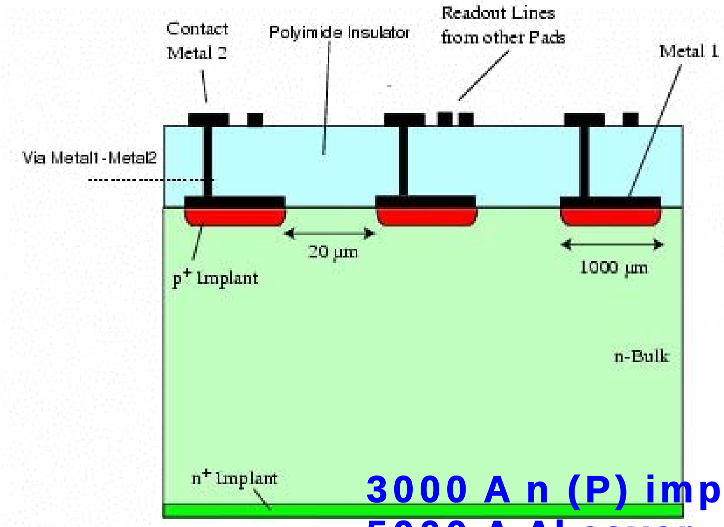
Emission Channeling of decay particles, on single crystals (β^-, β^+, c.e., α)...lattice location of impurities in crystals



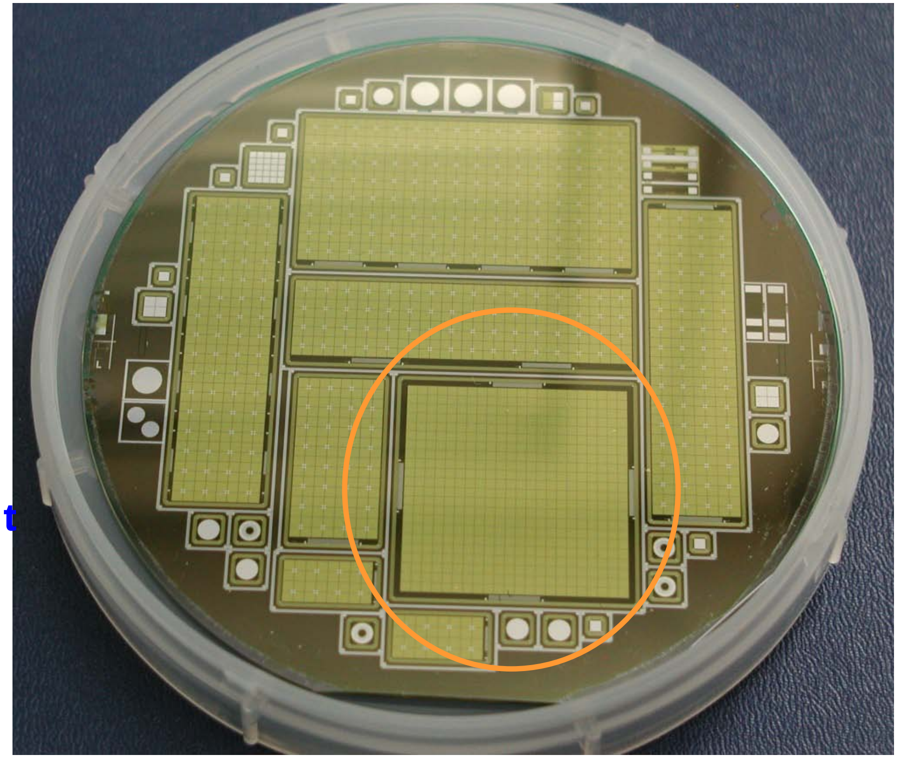
What do you need to do Emission Channeling

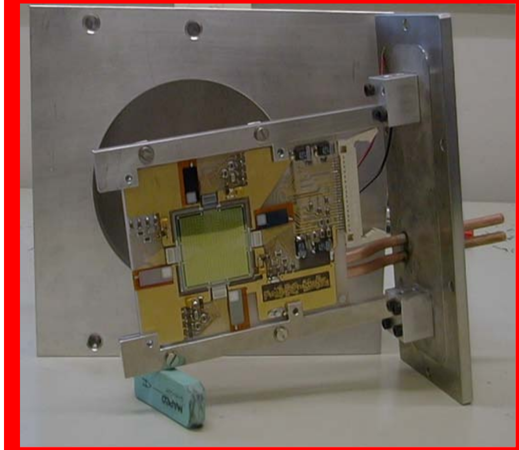
Si PAD electron detectors

5μm kapton
2μm SiO2
0.3mm p (B) implant



3000 A n (P) implant
5000 A Al cover

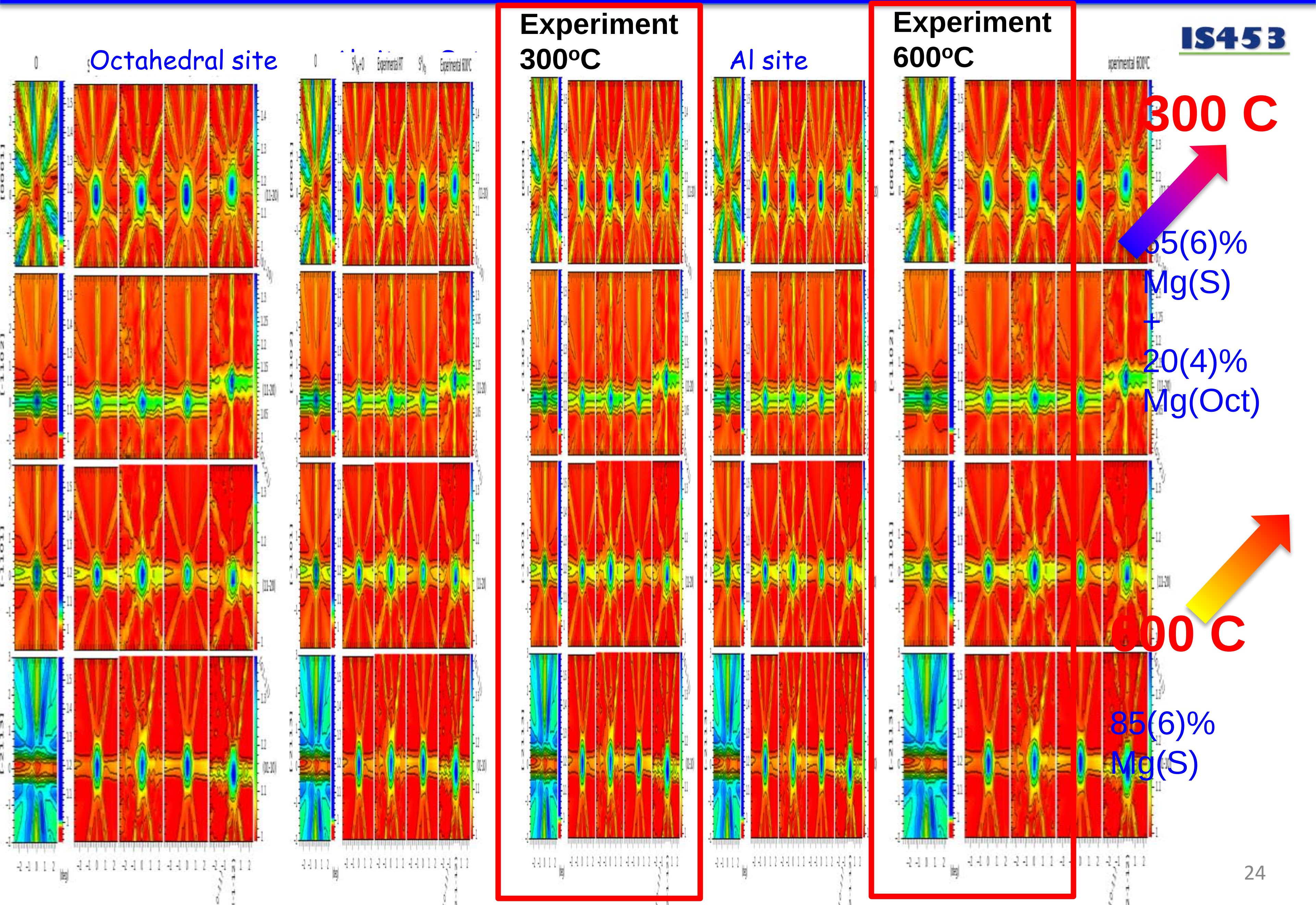


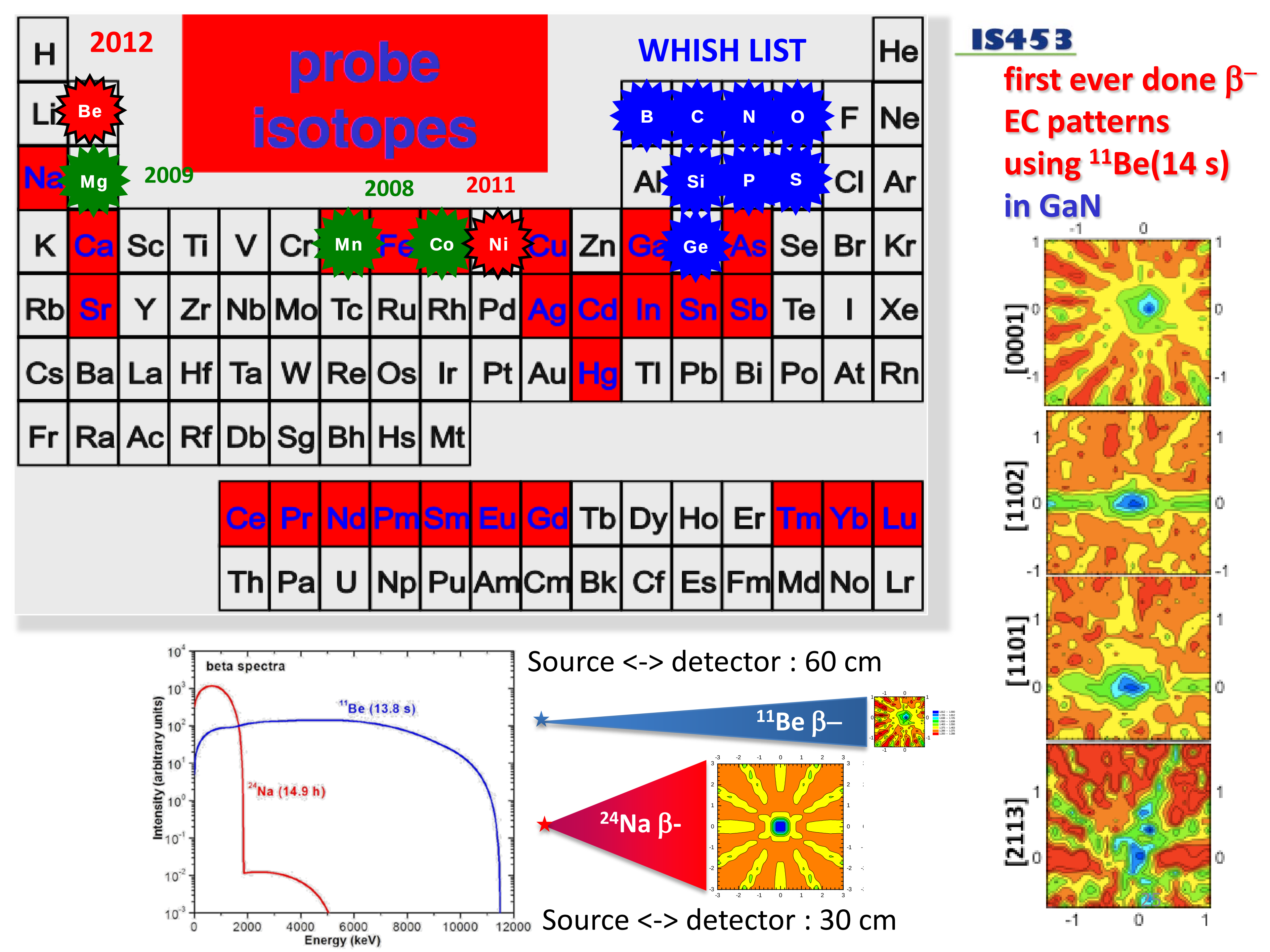


Position-sensitive detector

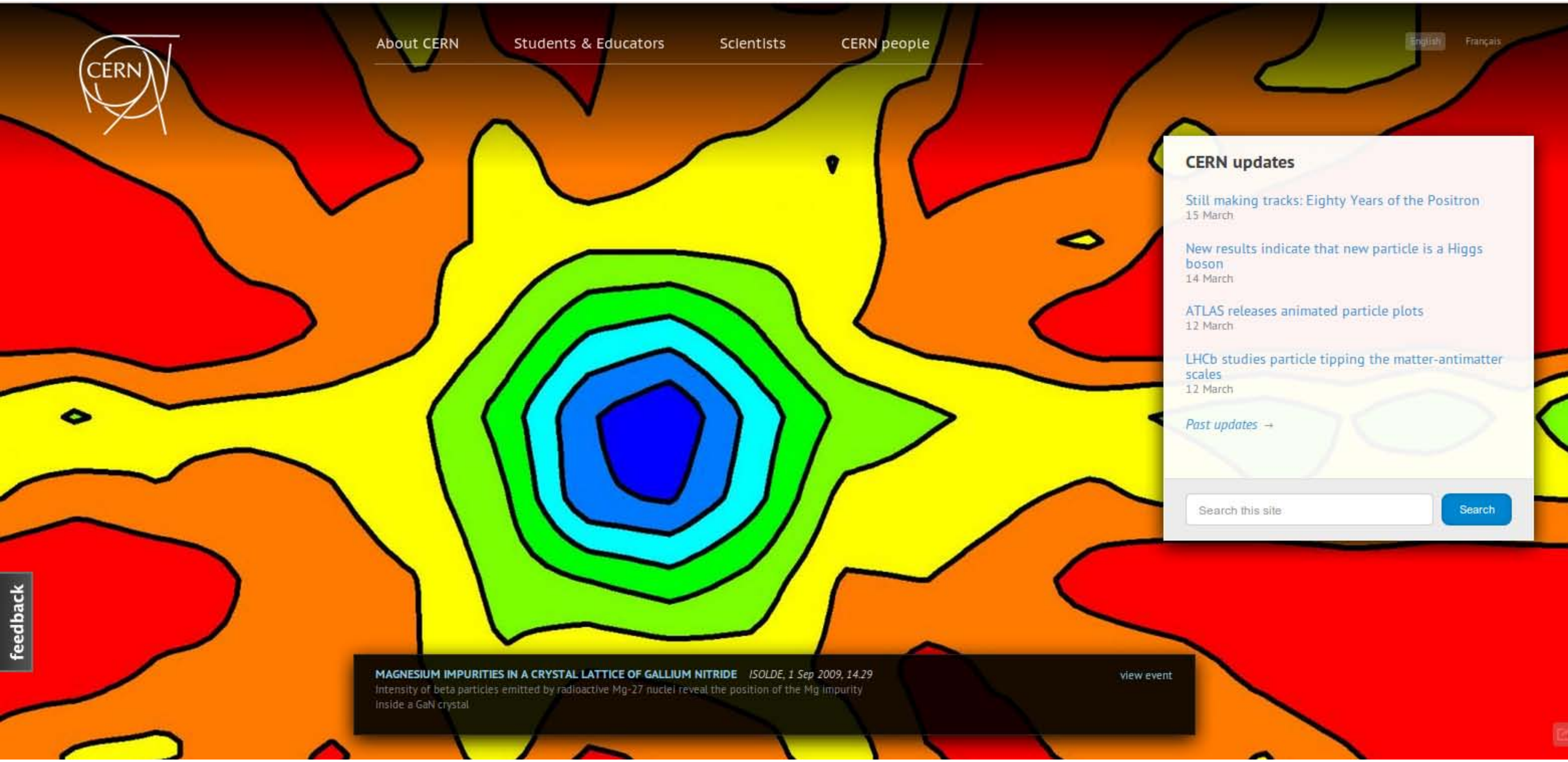
Good energy resolution ~3 keV
Large pad – 1.4x1.4 mm2
Dead / unbonded channels
Leakage current limiting depletion
15keV << E(e^-) << 300 keV
Readout → 200Hz … 5 kHz

Lattice location study of implanted ^{27}Mg (13min) : AlN

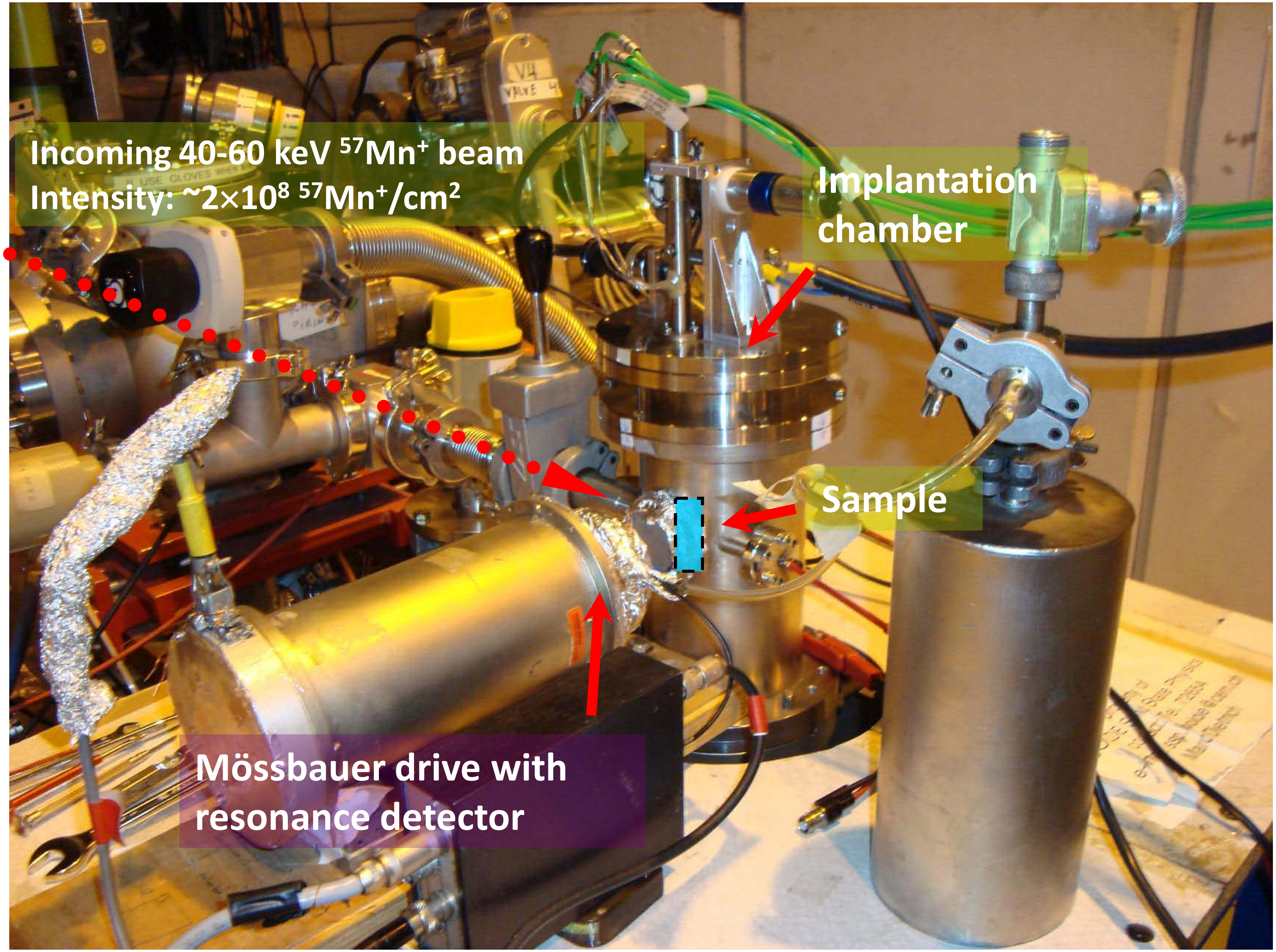


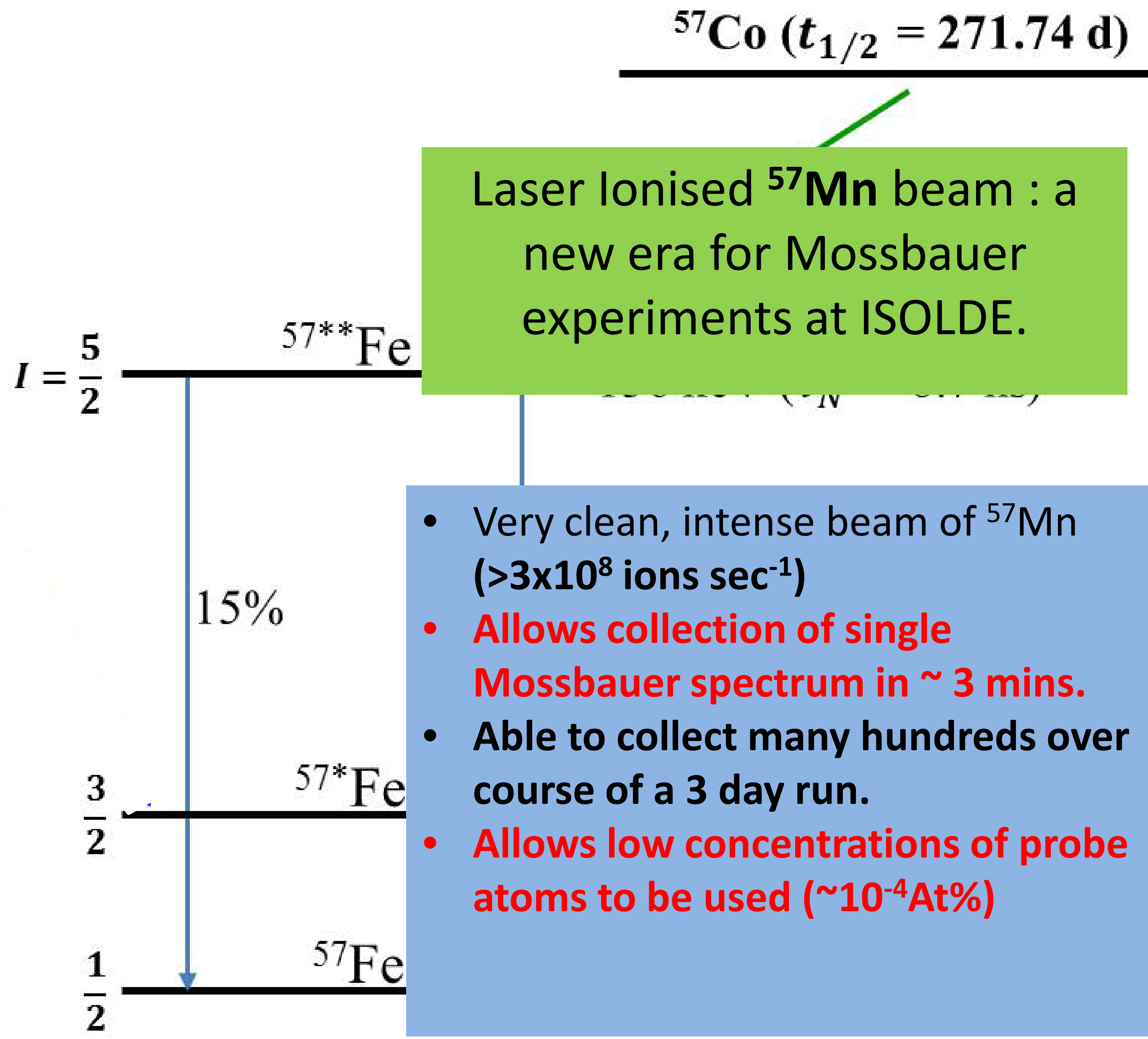

IS453
first ever done β⁻ EC patterns using ¹¹Be(14 s) in GaN
probe isotopes
WHISH LIST
2012
2009
2008
2011
H He
Li Be B C N O F Ne
Na Mg Al Si P S Cl Ar
K Ca Sc Ti V Cr Mn Fe Co Ni Cu Zn Ga Ge As Se Br Kr
Rb Sr Y Zr Nb Mo Tc Ru Rh Pd Ag Cd In Sn Sb Te I Xe
Cs Ba La Hf Ta W Re Os Ir Pt Au Hg Tl Pb Bi Po At Rn
Fr Ra Ac Rf Db Sg Bh Hs Mt
Ce Pr Nd Pm Sm Eu Gd Tb Dy Ho Er Tm Yb Lu
Th Pa U Np Pu Am Cm Bk Cf Es Fm Md No Lr
[0001]
[1102]
[1101]
[2113]
beta spectra
¹¹Be (13.8 s)
²⁴Na (14.9 h)
Intensity (arbitrary units)
Energy (keV)
Source <-> detector : 60 cm
¹¹Be β–
²⁴Na β-
Source <-> detector : 30 cm

....and good for publicity



Hyperfine Interactions with Mossbauer spectroscopy



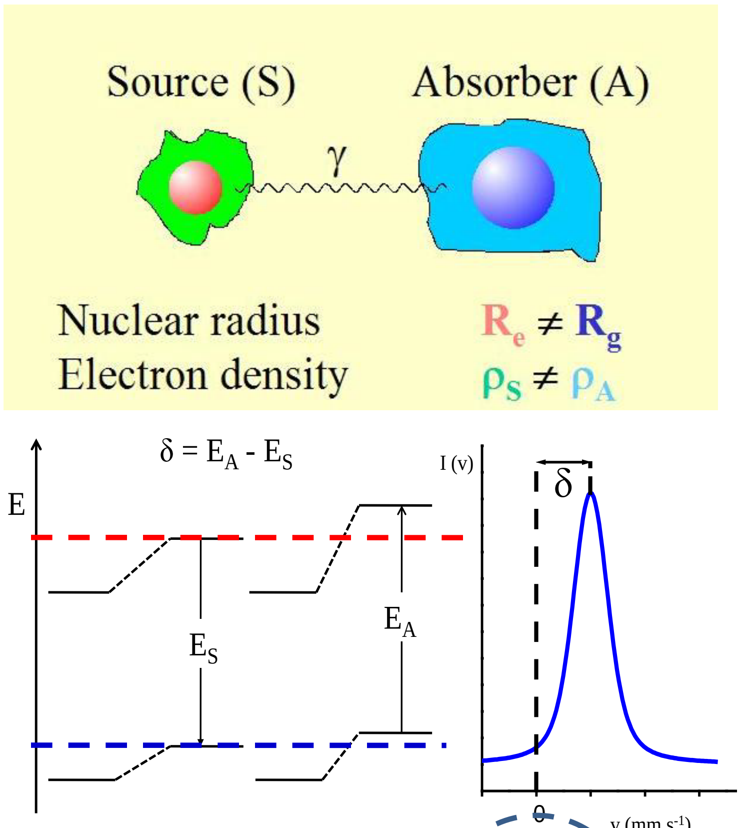


Laser Ionised ^{57}**Mn** beam : a new era for Mossbauer experiments at ISOLDE.

- 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%)**

Local information….

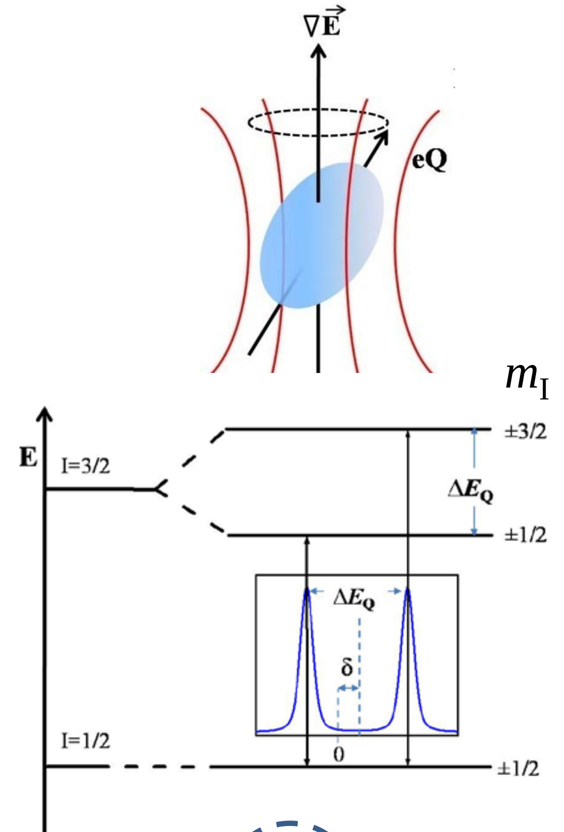
Isomer Shift



$$\delta_{IS} = \alpha\left[\rho_a(0) - \rho_s(0)\right]$$

Chemical bonding
Charge states

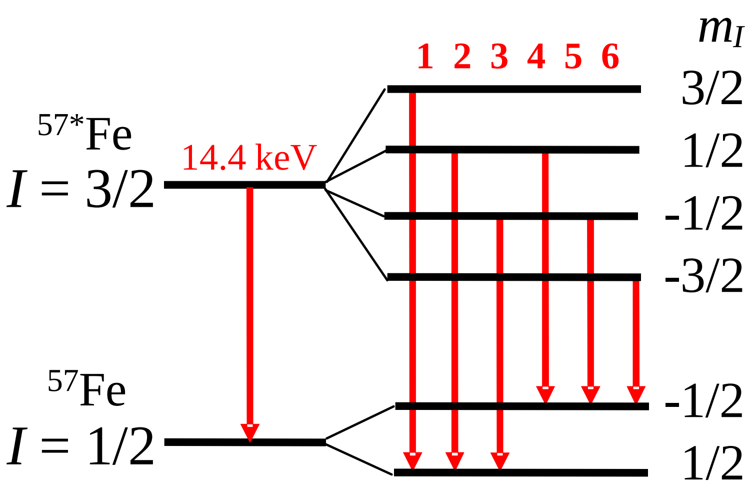
Quadrupole Splitting



$$\Delta E_Q = \pm \frac{eQV_{ZZ}}{2}\left(1 + \frac{\eta^2}{3}\right)^{1/2}$$

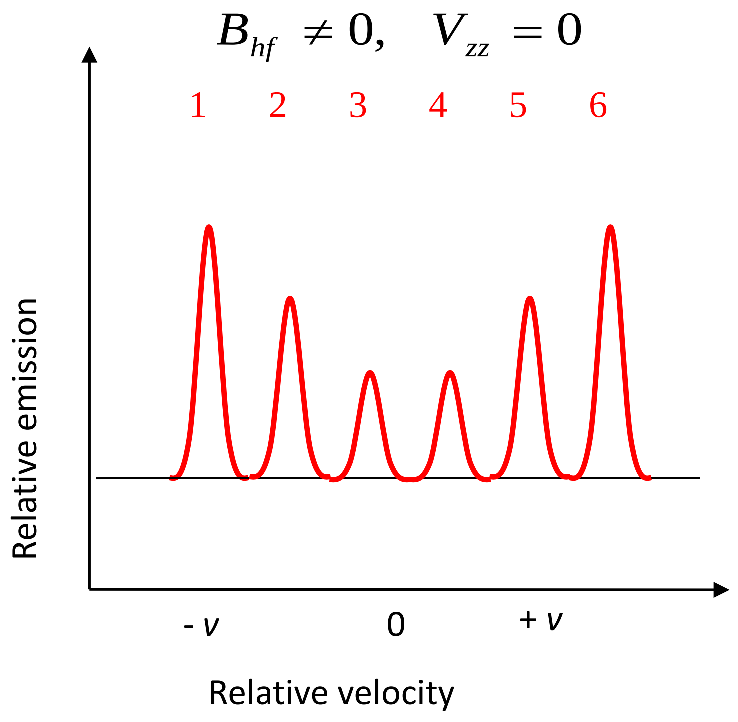
Lattice asymmetry
Clustering of atoms

Magnetic hf. splitting of ^{57}Fe: Sextet



Source/sample:
Mn/Fe implanted MgO

$$B_{\text{hf}} \propto \left|\psi_s^{\downarrow}\right|^2 - \left|\psi_s^{\uparrow}\right|^2$$



Motivation .....

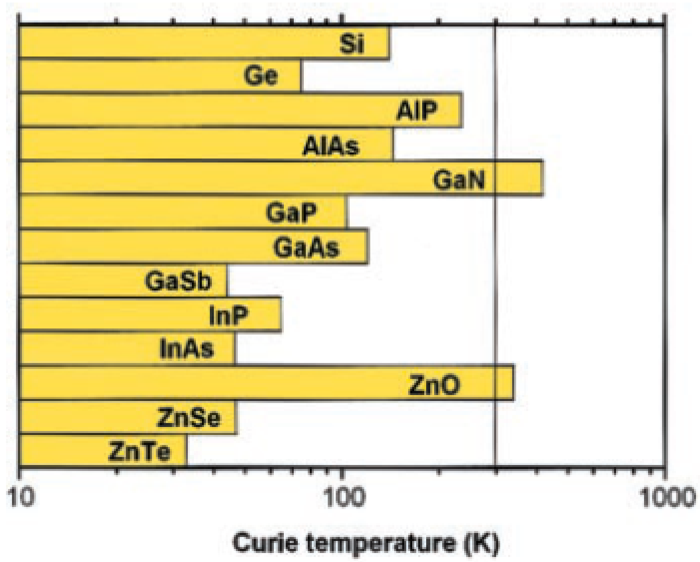


Fig. 3. Computed values of the Curie temperature T_C for various p-type semiconductors containing 5% of Mn and 3.5×10^{20} holes per cm^3.

Dietl *et al*, *Science* 287 (2000) 1019

Is it possible to create magnetic semiconductors that work at room temperature?
Such devices have been demonstrated at low temperatures but not yet in a range warm enough for spintronics applications.

Fe: ZnO a ferromagnetic semiconductor? (no!)

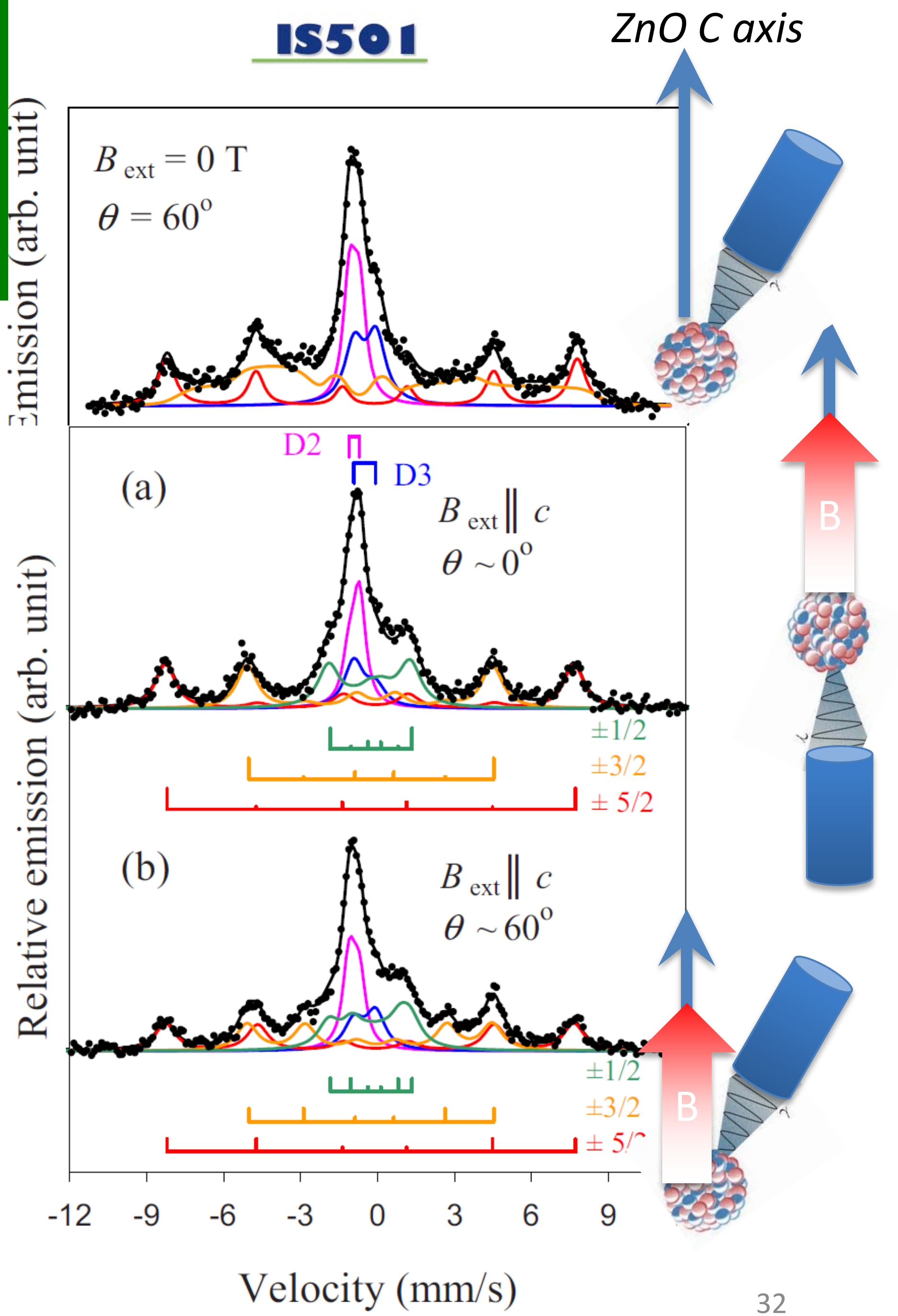
6 fold spectrum: characteristic of magnetic structure (at room temperature!!!).

Results in an external magnetic field show that the spectrum shown to be a **slowly relaxing paramagnetic system** .

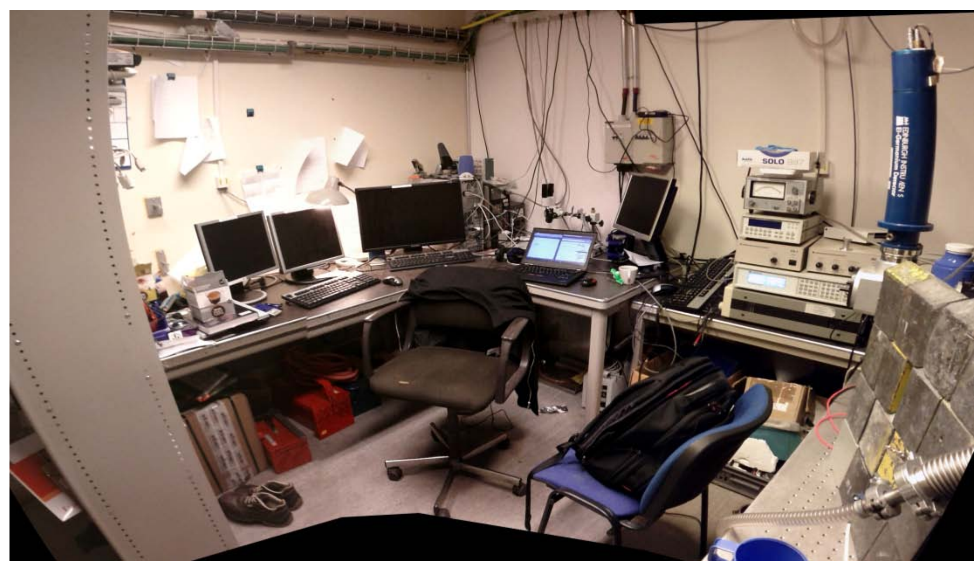
Gunnlaugsson *et al* (APL **97** 142501 2010)

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)



Also….Infrastructure



Time for a change…

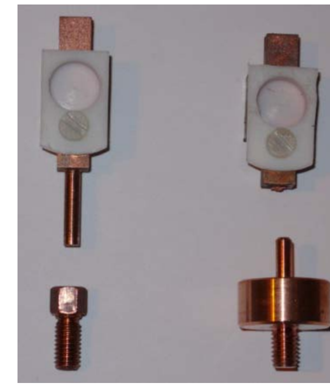
New 120m² lab…completion ~ sometime …2014

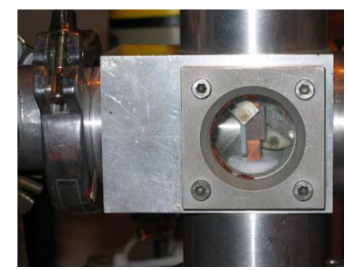
Biophysics at ISOLDE: Radioisotopes for Probing Biomolecular Functionality in Living Matter

PAC isotopes used for biophysics at ISOLDE:

parent	half-life	decay	isomer	half-life (ns)
^{62}Zn	9.186(13) h	EC/β^+	^{62}Cu	4.57(18)
^{99}Mo	65.94(1) h	β^-	^{99}Tc	3.61(7)
^{111m}Cd	48.54(5) min	IT	^{111}Cd	85.0(7)
^{111}In	2.8049(1) days	EC	^{111}Cd	85.0(7)
^{111}Ag	7.45(1) days	β^-	^{111}Cd	85.0(7)
^{133}Ba	10.52(13) years	EC	^{133}Cs	6.27(2)
^{160}Tb	72.3(2) days	β^-	^{160}Dy	2.02(1)
^{181}Hf	42.39(6) days	β^-	^{181}Ta	10.8(1)
^{199m}Hg	42.6(2) min	IT	^{199}Hg	2.45(2)
^{204m}Pb	67.2(3) min	IT	^{204}Pb	265(10)
		isotopes that have only been used for the sum-p		
^{147}Nd	10.98(1) days	β^-	^{147}Pm	2.50(5)
^{152}Eu	13.542(10) years	EC	^{152}Sm	1.428(7)

Hemmingsen et al. *Chem. Rev.*, **2004**, 104: 4027

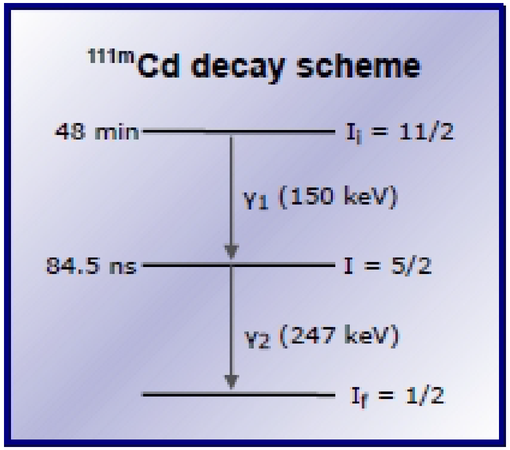




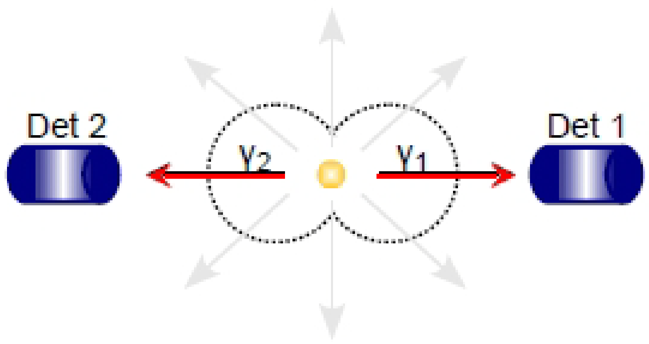
Suitable isotopes exist for probing toxicology in bio-systems: Pb, Cd and Hg

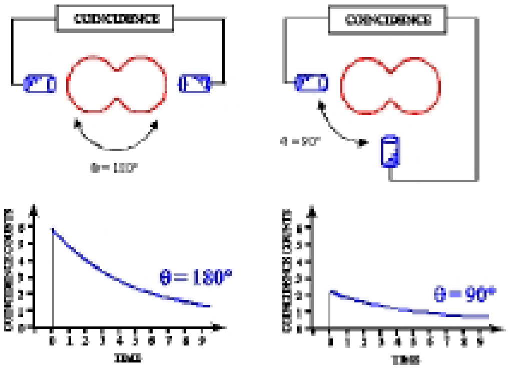
No implantations: collect activity in ice → chemistry

PAC Spectroscopy



Angular correlation of γ-rays

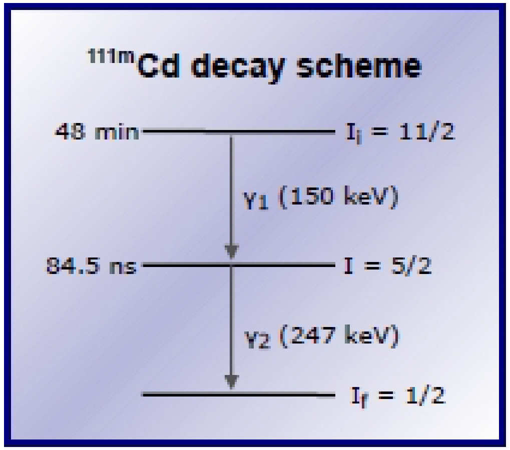




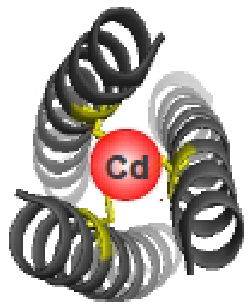
(www.uni-leipzig.de)

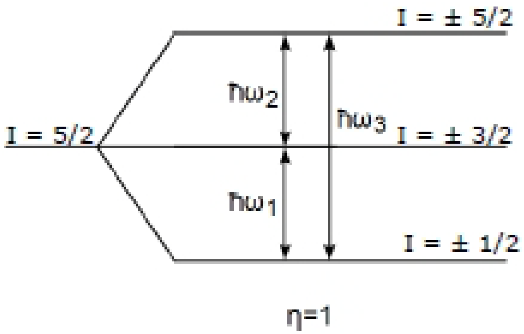
- Angular correlation of γ-rays is a property of the nuclear decay
- The distance from the center to the curve gives the probability of emission of γ_2 in that direction
- Coincidence count rate depends on the angle between detectors

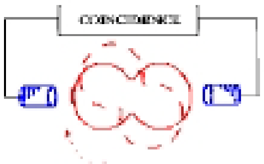
PAC Spectroscopy

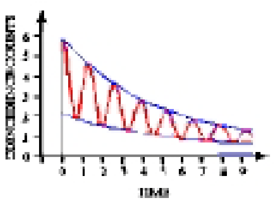


Perturbed angular correlation of γ-rays: the influence of extra-nuclear fields









(www.uni-leipzig.de)

HFI causes precession of the nuclear spin

↓

Rotation of the angular correlation

↓

Information about ω_1, ω_2 and ω_3
(the finger print of the local charge distribution)

Metal Ion Binding Site Structure: Fast inter-conversion between species

B

^{111m}Cd-PAC

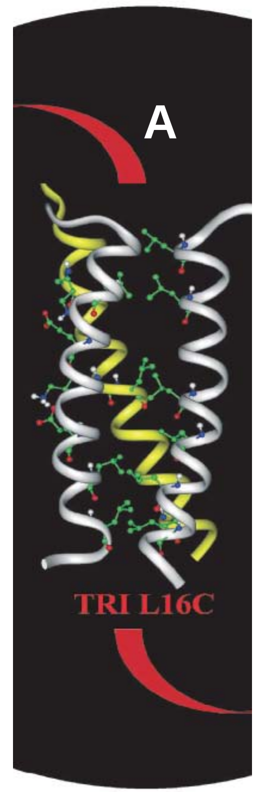
C

^{113}Cd-NMR

B

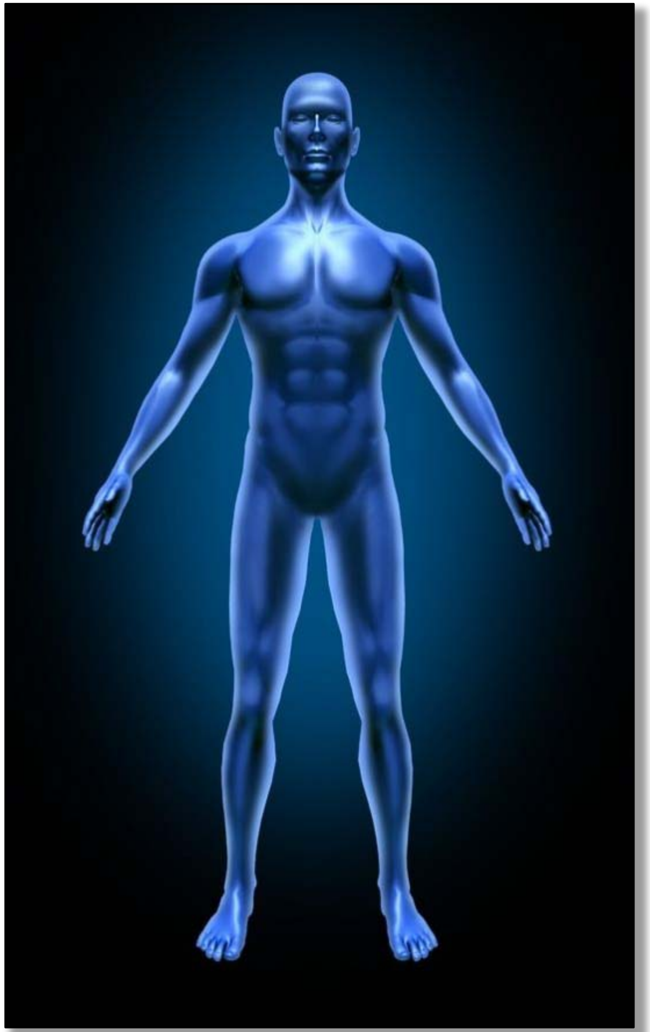
C

Chemical shift / ppm



Matzapetakis *et al*. J. Am. Chem. Soc. **2002**, 124: 8042; Lee *et al*. Angew. Chem., **2006**, 45: 2864; Peacock *et al.* Proc. Nat. Acad. Sci. **2008**, 105: 16566

Elements in Human Body



Human body:

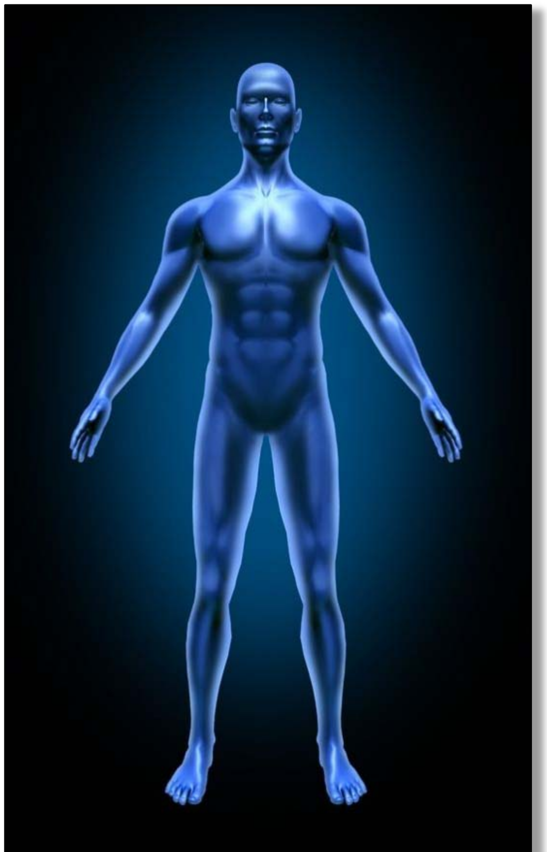
- $6.7 \bullet 10^{27}$ atoms
- 99% 4 major elements:

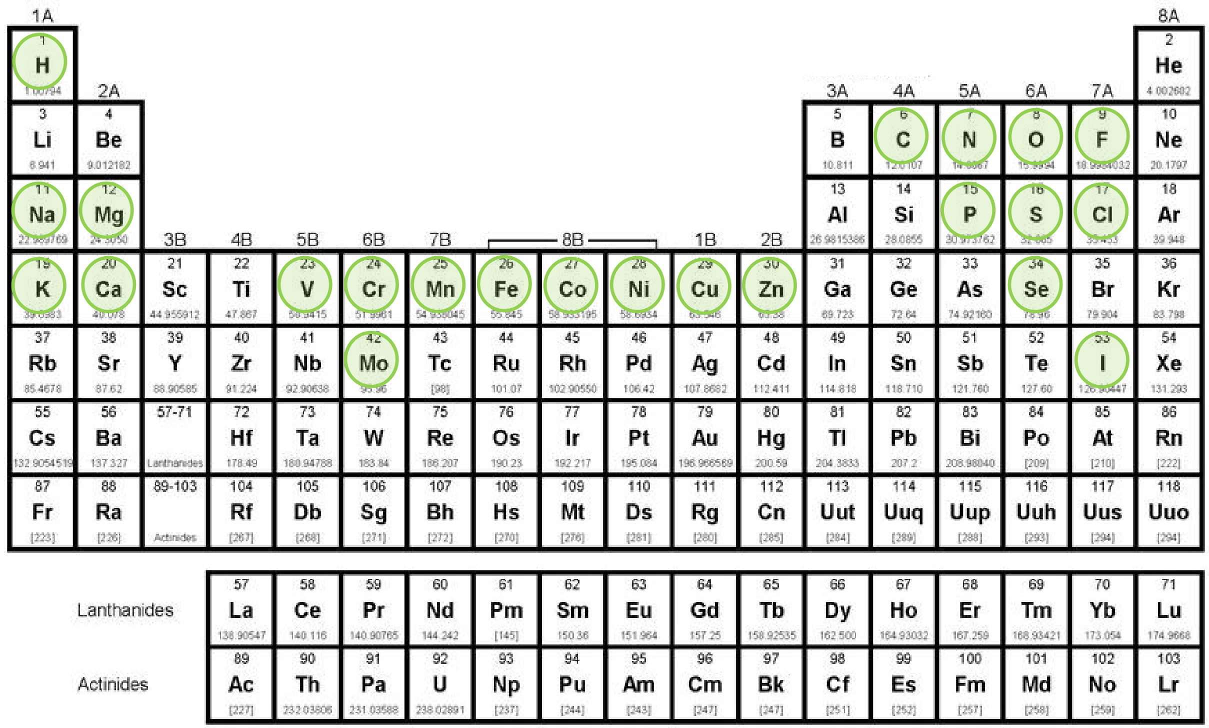
  O (43kg), C (16kg), H (7kg), N (1.8kg)
- 1% 21 other elements:

  (Ca:1kg, Mg:19g, Fe:3-5g, Zn:3g,

  Cu: 300mg)

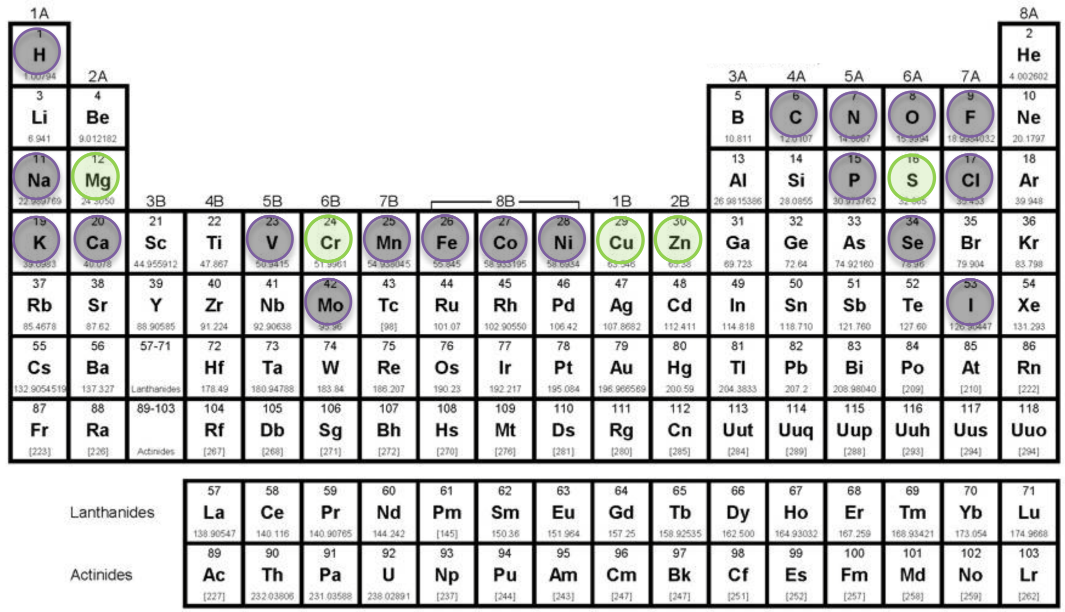
Elements in Human Body





Natural elements

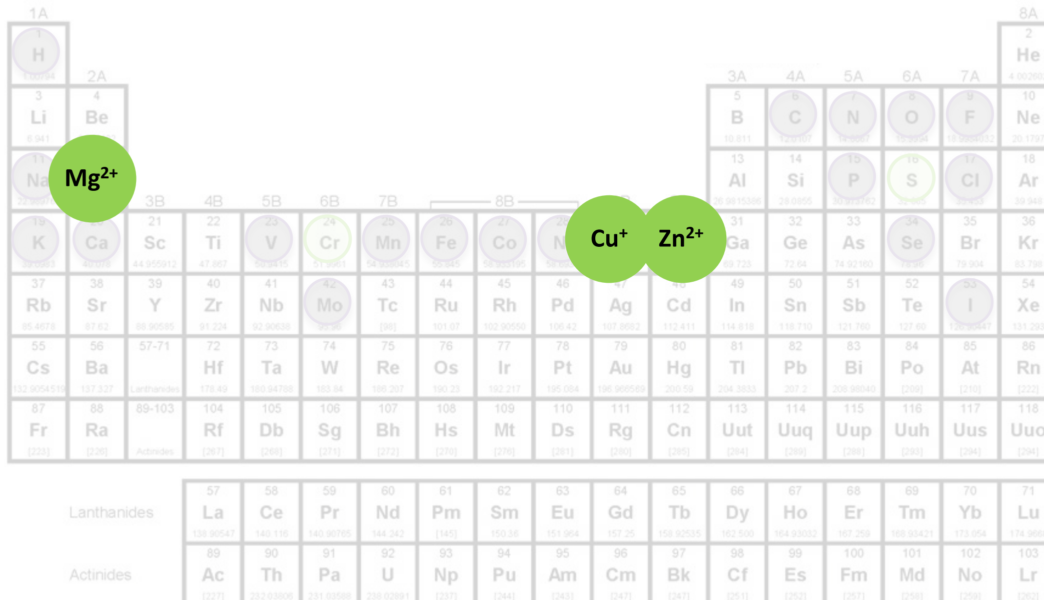
Elements in Human Body



Natural elements

Ions accessible in established techniques

Elements in Human Body



Mg(II), Cu(I), Zn(II)

Natural elements

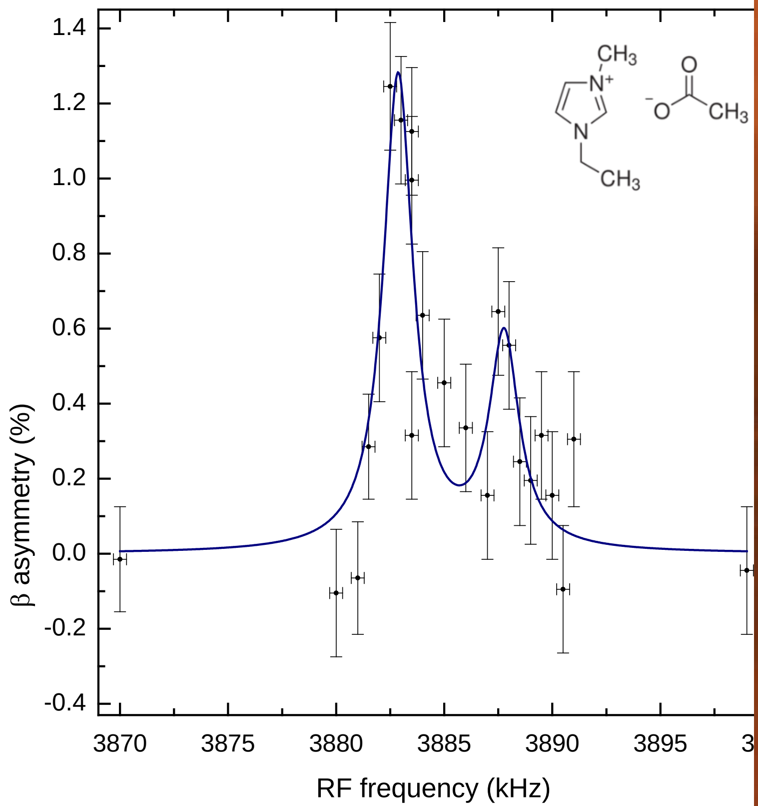
Ions accessible in established techniques

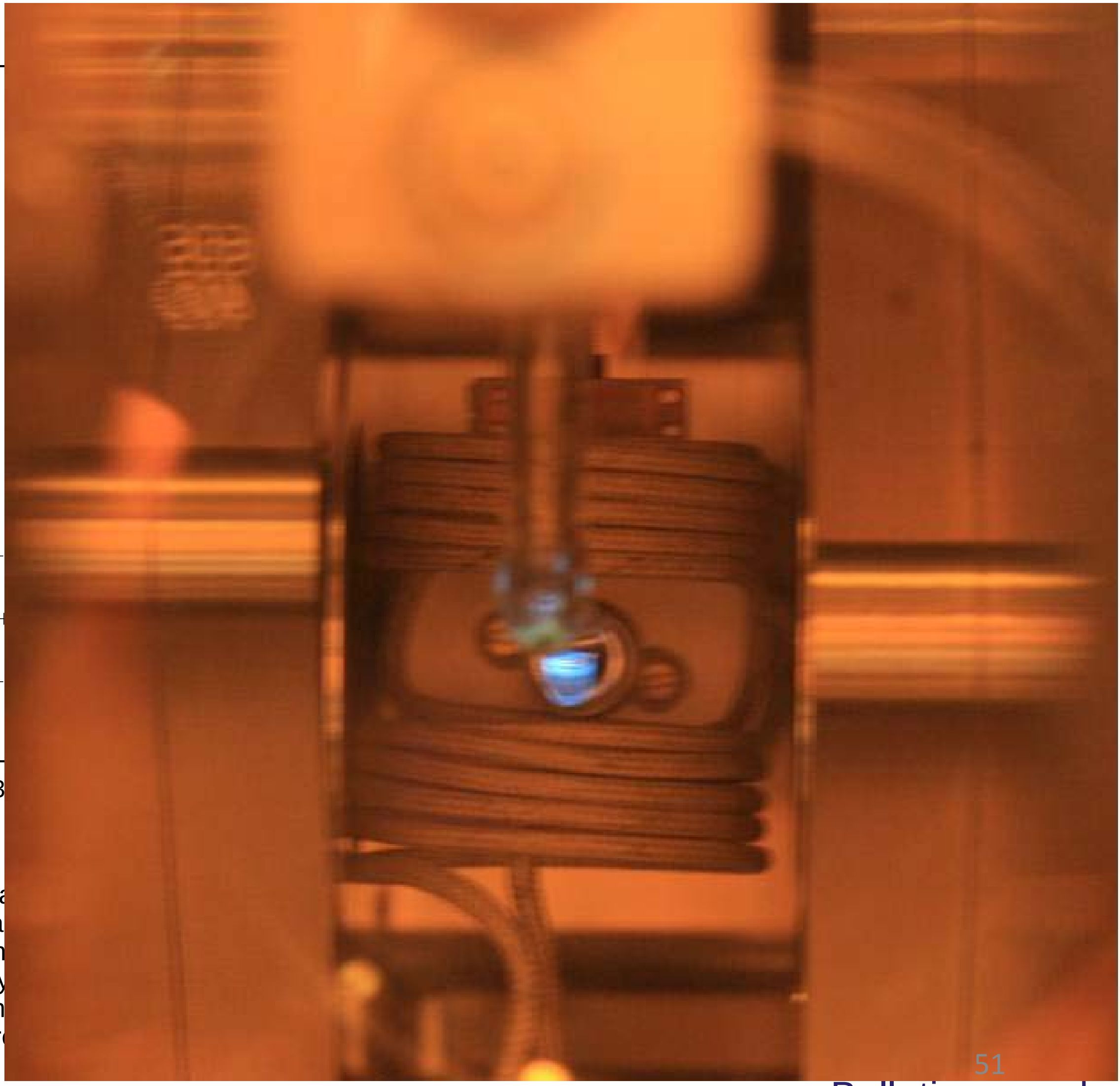
- Most abundant ions in human body
- Closed shell ions → silent in most spectroscopic techniques

2012 – first (and successful) $^{31}Mg^{+}$ β-NMR experiment applied to soft condensed matter

$^{31}Mg^{+}$ implanted into an ionic liquid (EMIM-Ac):

Differential pumping and drop
Mounted @ COLLAPS experiment

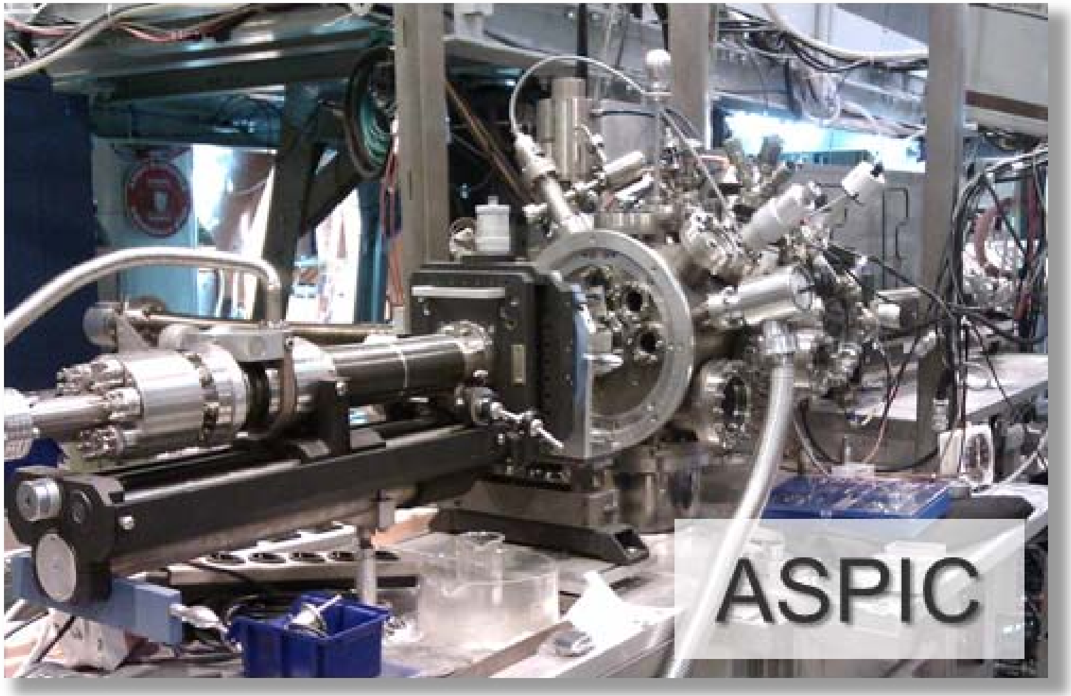




Monika Stachura, University of Copenhagen; Magdalena Kowa
CERN, Geneva; Alexander Gottberg, CSIC, Madrid; Klaus Bla
Planck Institute for Nuclear Physics, Heidelberg; Gerda Neyen
Leuven University, (Leuven); Rainer Neugart, Mainz University
(Mainz); Deyan Yordanov, Max Planck Institute for Nuclear Ph
Heidelberg; Mark Bissell, Leuven University, (Leuven); Kim Kr
Planck Institute for Nuclear Physics, Heidelberg

The VITO project

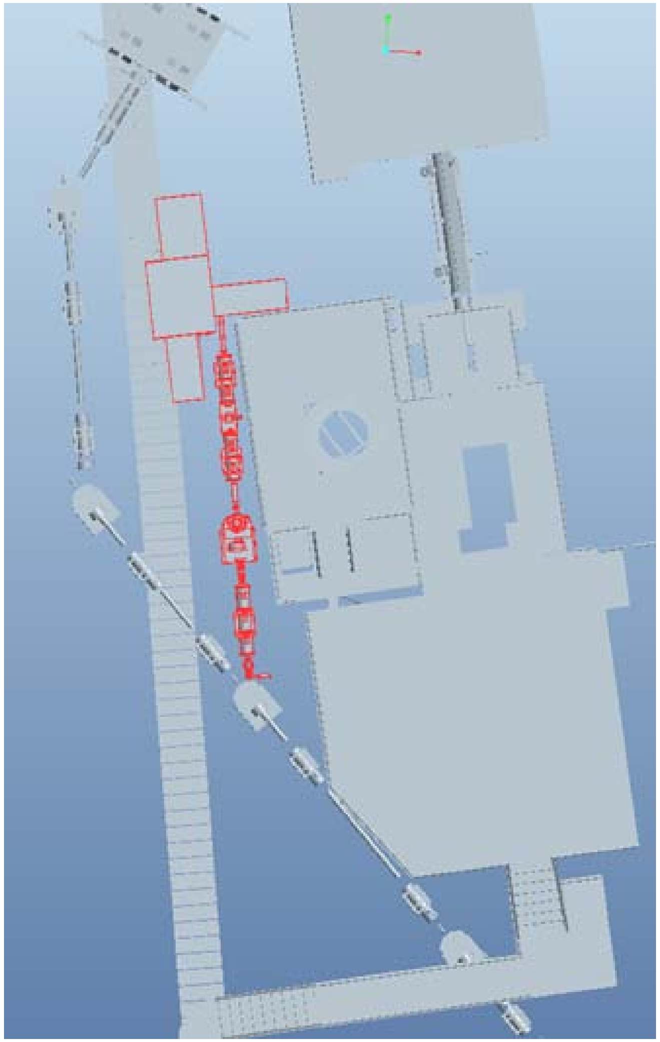
(Versatile Ion-polarized Techniques Online)



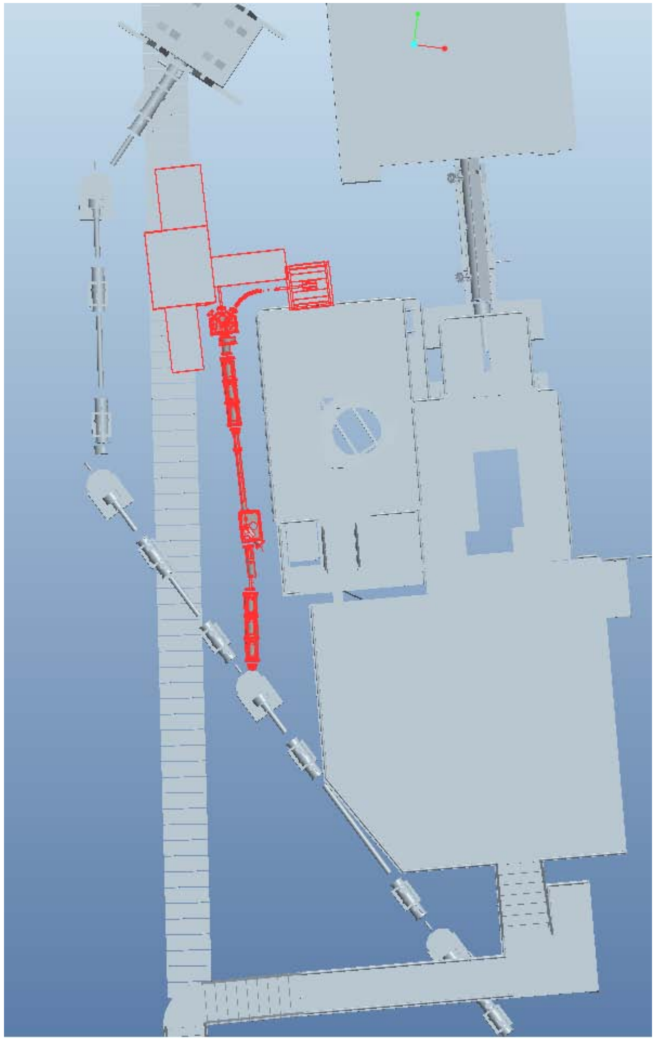
Cern.ch/vito

ASPIC → ViTO Upgrade

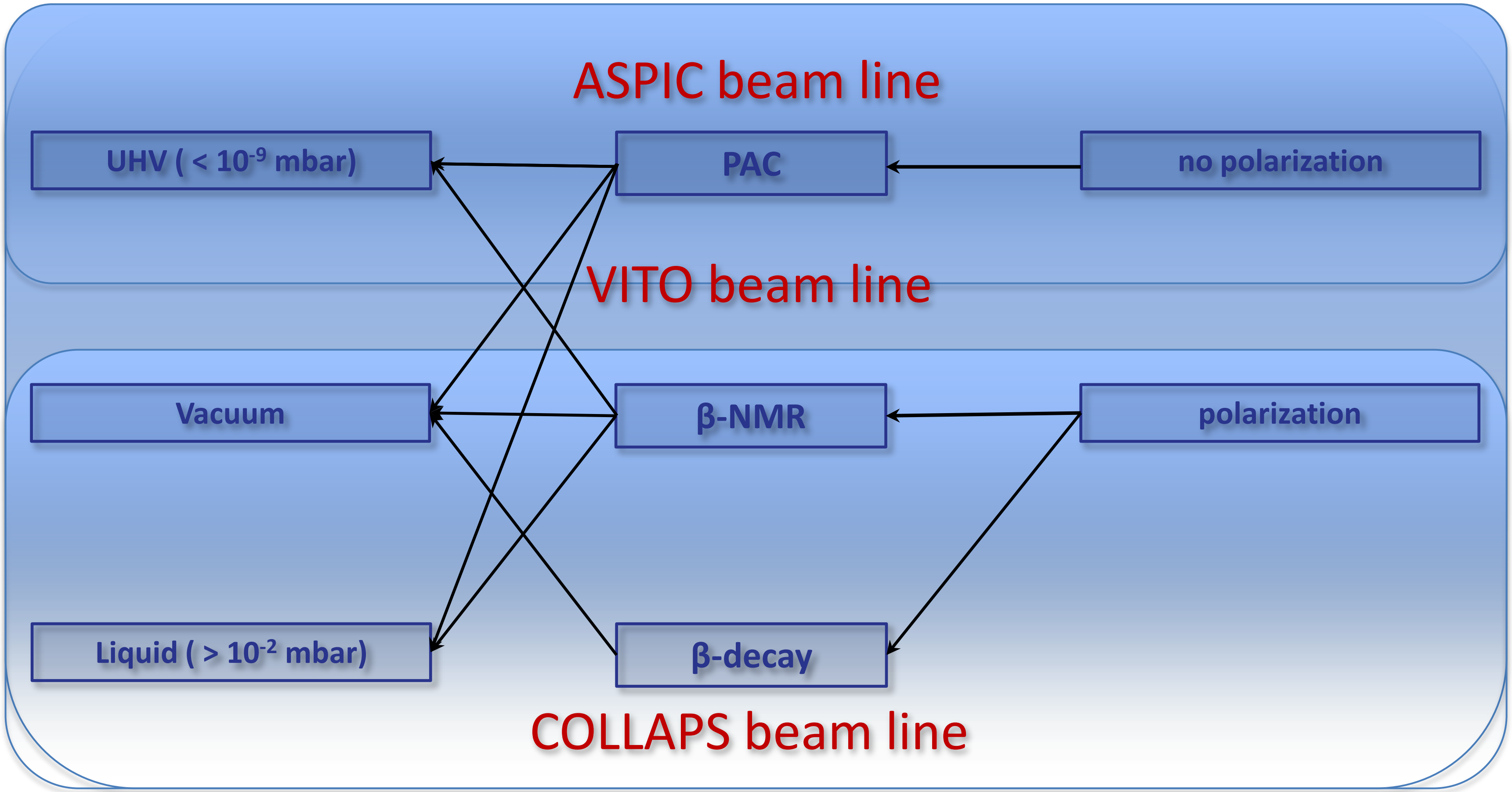
ASPIC



ViTO



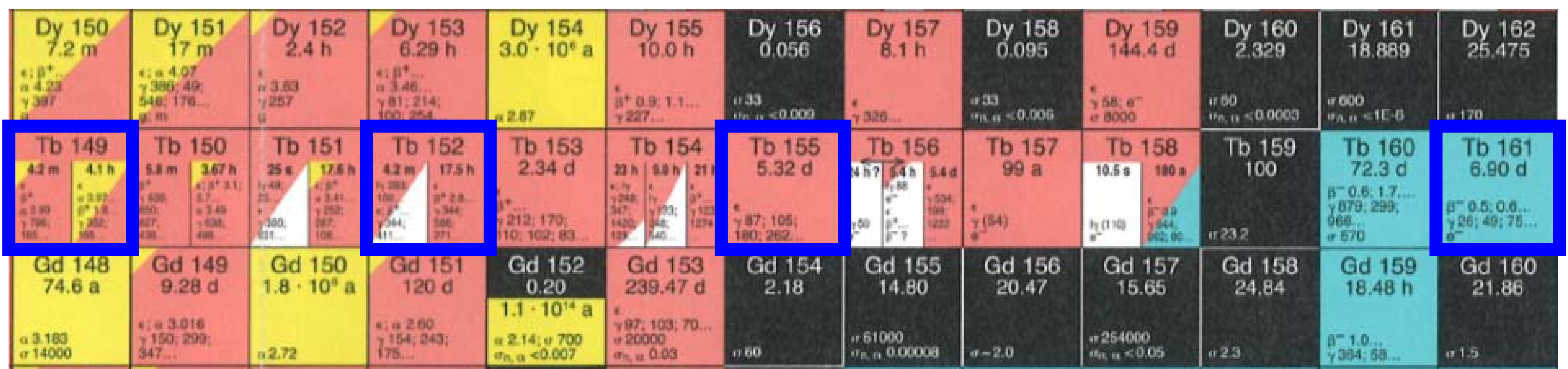
Physics at ViTO



Radioisotopes for targeted alpha therapy

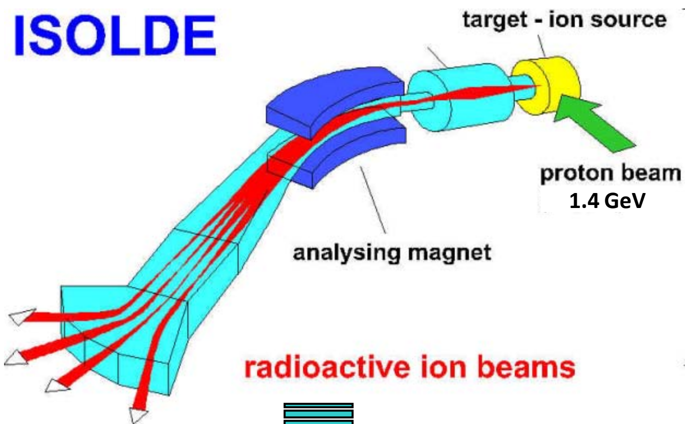
Radio-nuclide	Half-life (h)	Advantage	Problem
Tb-149	4.12	Known (bio)chemistry	Availability
Pb-212	10.6	Availability (recycled from U-fuel ^{228}Th)	Release of daughter?
Bi-212	1.01	Known (bio)chemistry Availability	Short half-life
Bi-213	0.76	Known (bio)chemistry	Short half-life Availability
At-211	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



In practice (at the moment)

i. Collection at ISOLDE

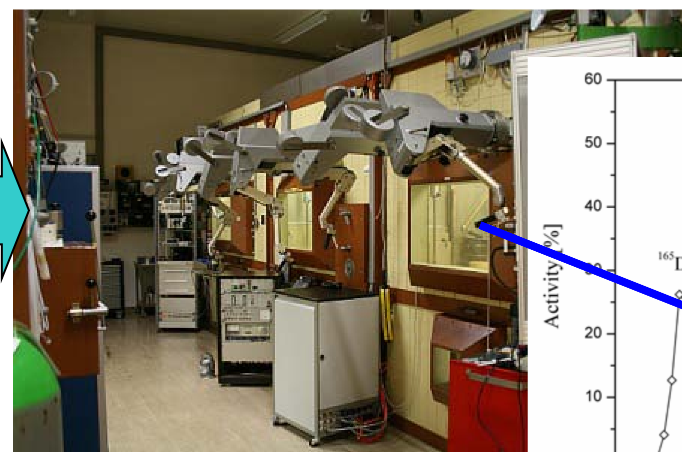


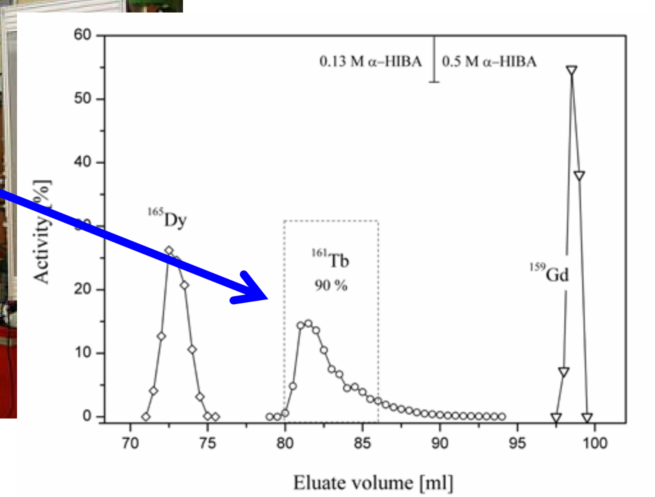
iv. Injection into mouse

v. PET/SPECT imaging and tumor treatment

ii. Shipping to PSI

iii. Radiochemical purification and labeling





Imaging Studies Using PET and SPECT

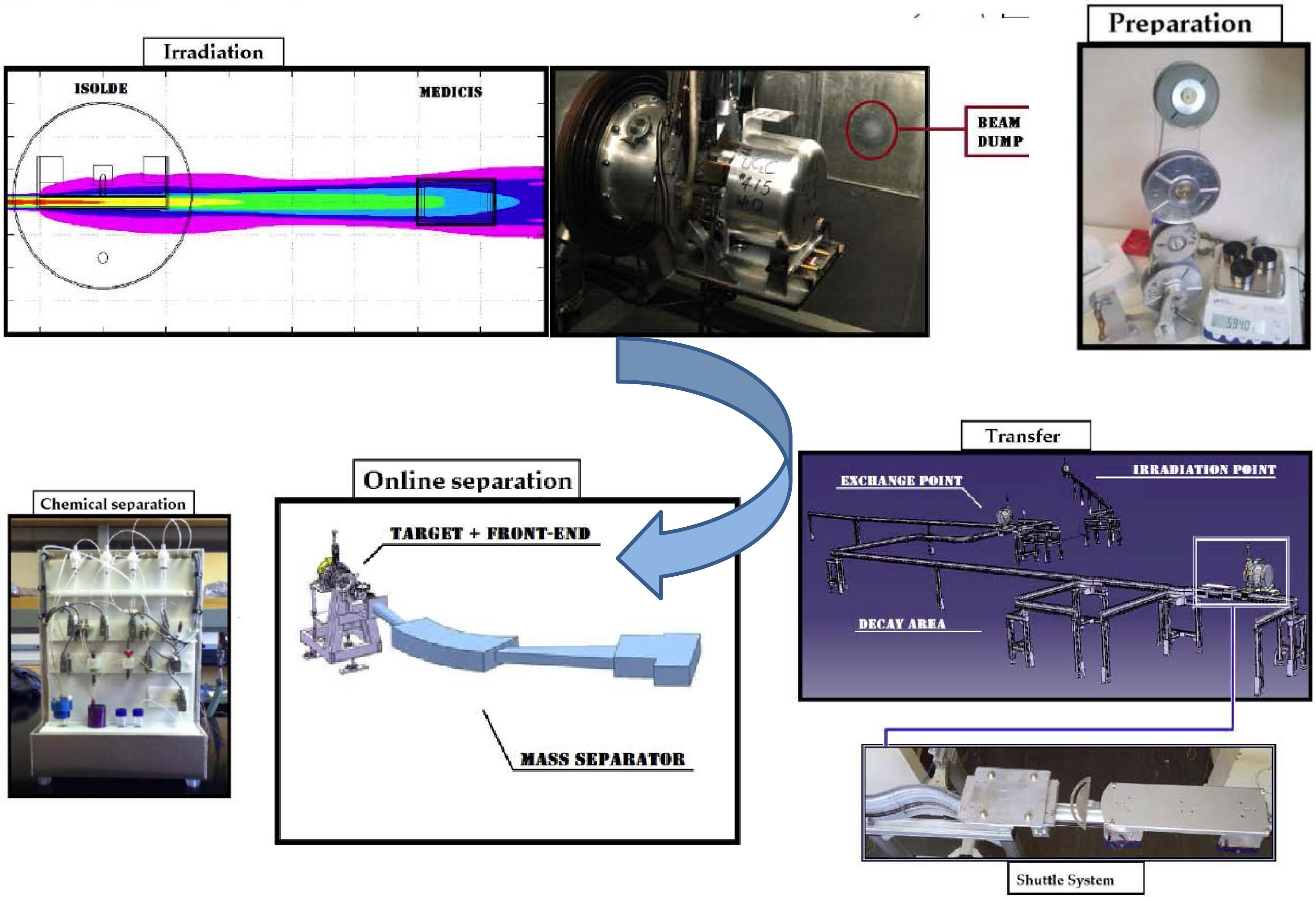
All very nice, but: how to have a more regular source of beam for these kinds of experiments? (once a year not enough)

C. Müller et al., J. Nucl. Med. 53 (2012) 1951

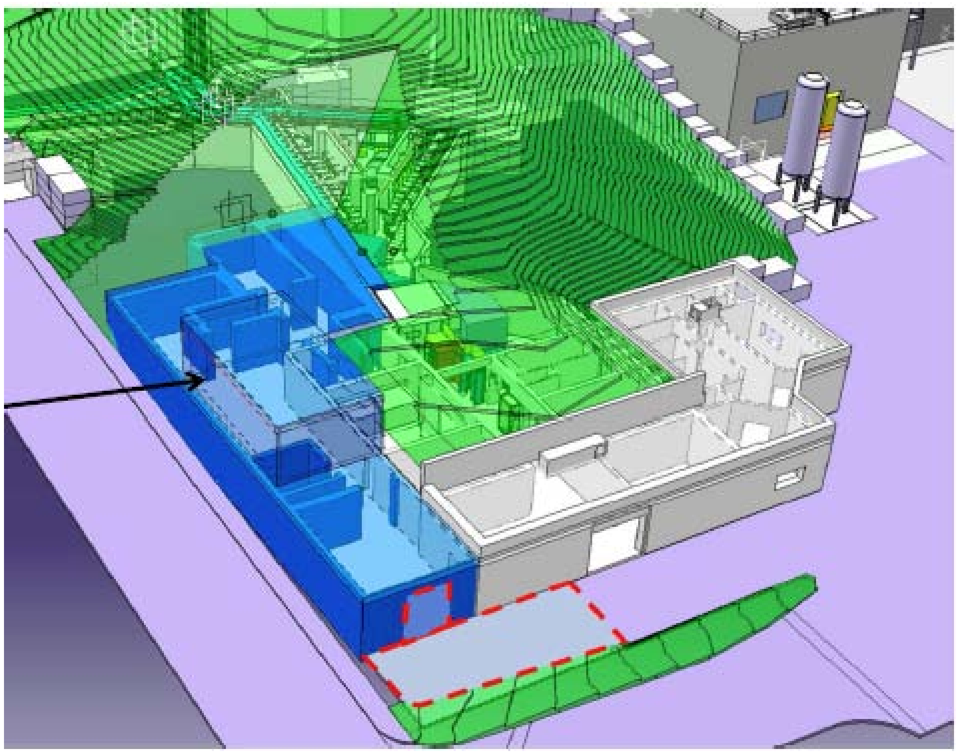
IS528 collaboration: supported by ENSAR, Swiss National Science Foundation and Swiss South African Joint Research Program.

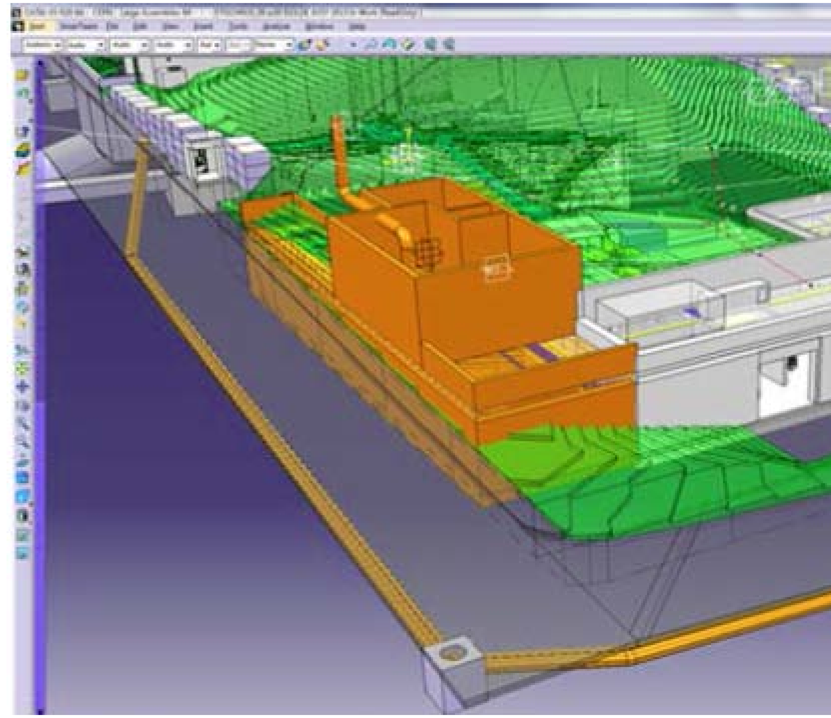
(*Medical Isotopes Collected at ISOLDE*)

Augusto et al *Appl. Sci.* **2014**, *4*, 265-281



Ground breaking Sept 4th 2013





INNOVATIVE ISOTOPES

Field of Application	Radiation	Chemical elements	Half lives
PET	β^+	Alkaline earth Halogen Lanthanide Transition metals …	10's min. Hours Days Months
SPECT	γ		
TAT	α		
Beta therapy	β^-		
Auger therapy	e^-		

A new office for biomedical applications at CERN

Interview with Steve Myers

Oct 20 2013

Medical applications play an important role at CERN and recently the DG has decided to structure them under a common umbrella. Steve Myers, CERN's Director of Accelerators and Technology has been appointed as the first Head of CERN's Office for Medical Applications. We have decided to dedicate October's issue of the PH Newsletter to this interesting field and asked Steve Myers for an interview. Following his long-standing career on accelerators and his experience from LEP and the LHC, Steve discusses about his future ambitions and the challenges of his new role.

What do you think about your new role and what is your greatest ambition?

This is the first time that CERN has put (into its medium-term) a budget line for medical applications. It is a small budget line but can be the seed for important projects. Over the last years, many medical activities have been going on at CERN; however these efforts were not focused. There are several activities related to CERN's technologies including the design of specialized accelerators for cancer therapy, the conversion of the Low Energy Ion Ring (LEIR) into a biomedical facility, radio-isotope production using ISOLDE, medical imaging and applications to improve dosimetry for patients and finally large scale computing applications. The DG thought that the time had come to put these projects under one umbrella and asked me to be the coordinator.

I want to set a common goal and try to coordinate these

Summary

- **<u>Unique</u>** information – be it chemical or local – which is only achievable using radioactive implantations/probes.
- Ability to profit from the large range of beams available at ISOLDE (especially now with online *and* offline setups)….imagination (and sometimes money) the only limitation.
- Can go beyond the "standard" isotopes used … e.g. ^{111}In, ^{57}Co.
- Many new developments underway e.g. β-NMR for biophysics and new dedicated offline labs.

Funding and Acknowledgements

- Manfred Deicher, Herbert Wolf, University of Saarlandes, Germany
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- Monika Stachura, CERN
- Peter Butler, University of Liverpool, UK
- Bruce Marsh, Sebastian Rothe, Valentin Fedesseov, CERN
- Palle Gunnlaugsson, University of Arhus, Denmark
- Thierry Stora, CERN
- Frank Wienholtz, CERN

Advantages and Limitations of PAC Spectroscopy for biophysics

Advantages:

- Characterisation of structure and dynamics at the PAC probe site (including rotational correlation times)
- High sensitivity to **structural changes**
- Small amount of PAC probe needed (in principle about **1 pmol**)
- **Different physical states** (crystals, surfaces, solutions, *in vivo*...)
- Mechanically stable, allowing for stirring, flow, ...

Limitations:

- Suitable PAC isotopes do not exist for all elements
- PAC isotope **must bind strongly** to the molecule of interest
- Spectral parameters do not uniquely determine structure
- After effects can cause problems (in particular for EC). **(^{111}In)**
- Production of PAC-isotopes

Hemmingsen et al. *Chem. Rev.*, **2004**, 104: 4027; Hemmingsen and Butz, in "Application of Physical Methods to Inorganic and Bioinorganic Chemistry" **2007**, Ed. R.A. Scott, Wiley