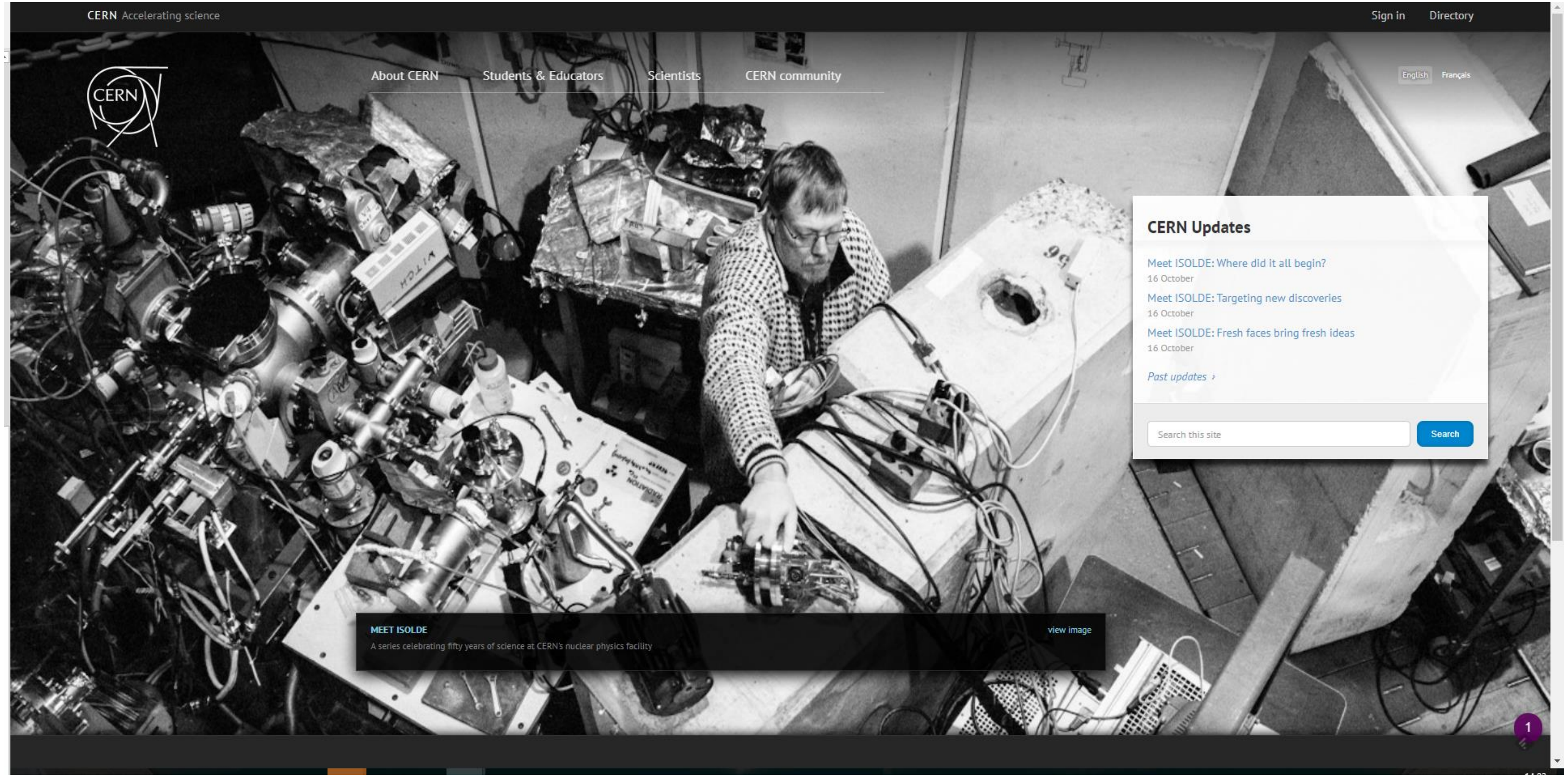
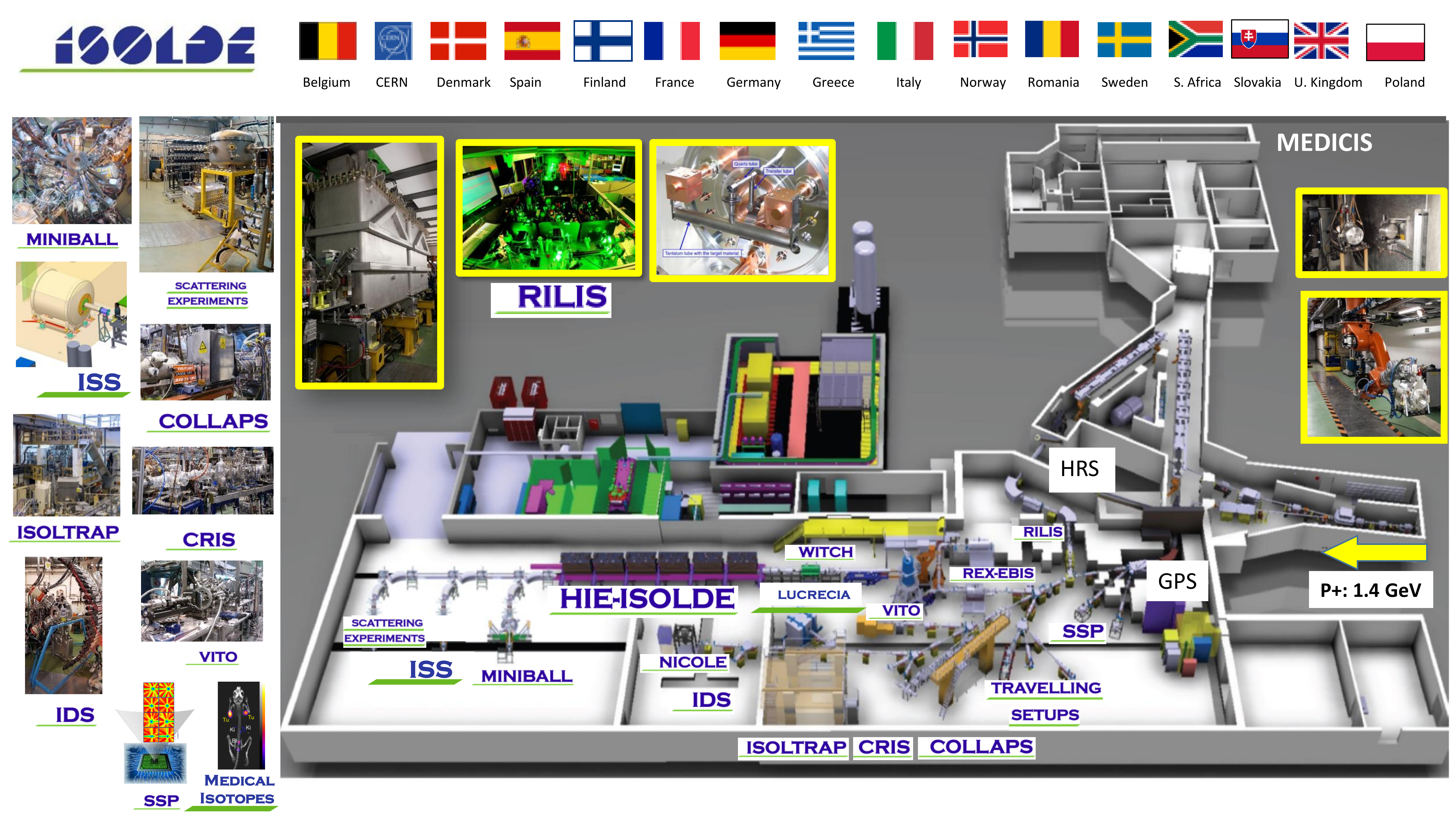
The Solid state programme at ISOLDE



Karl Johnston


ISOLDE
Belgium
CERN
Denmark
Spain
Finland
France
Germany
Greece
Italy
Norway
Romania
Sweden
S. Africa
Slovakia
U. Kingdom
Poland
MINIBALL
SCATTERING EXPERIMENTS
ISS
COLLAPS
ISOLTRAP
CRIS
IDS
VITO
SSP
MEDICAL ISOTOPES
RILIS
MEDICIS
HRS
GPS
P+: 1.4 GeV
RILIS
WITCH
REX-EBIS
HIE-ISOLDE
LUCRECIA
VITO
SSP
SCATTERING EXPERIMENTS
ISS
MINIBALL
NICOLE
IDS
TRAVELLING SETUPS
ISOLTRAP
CRIS
COLLAPS

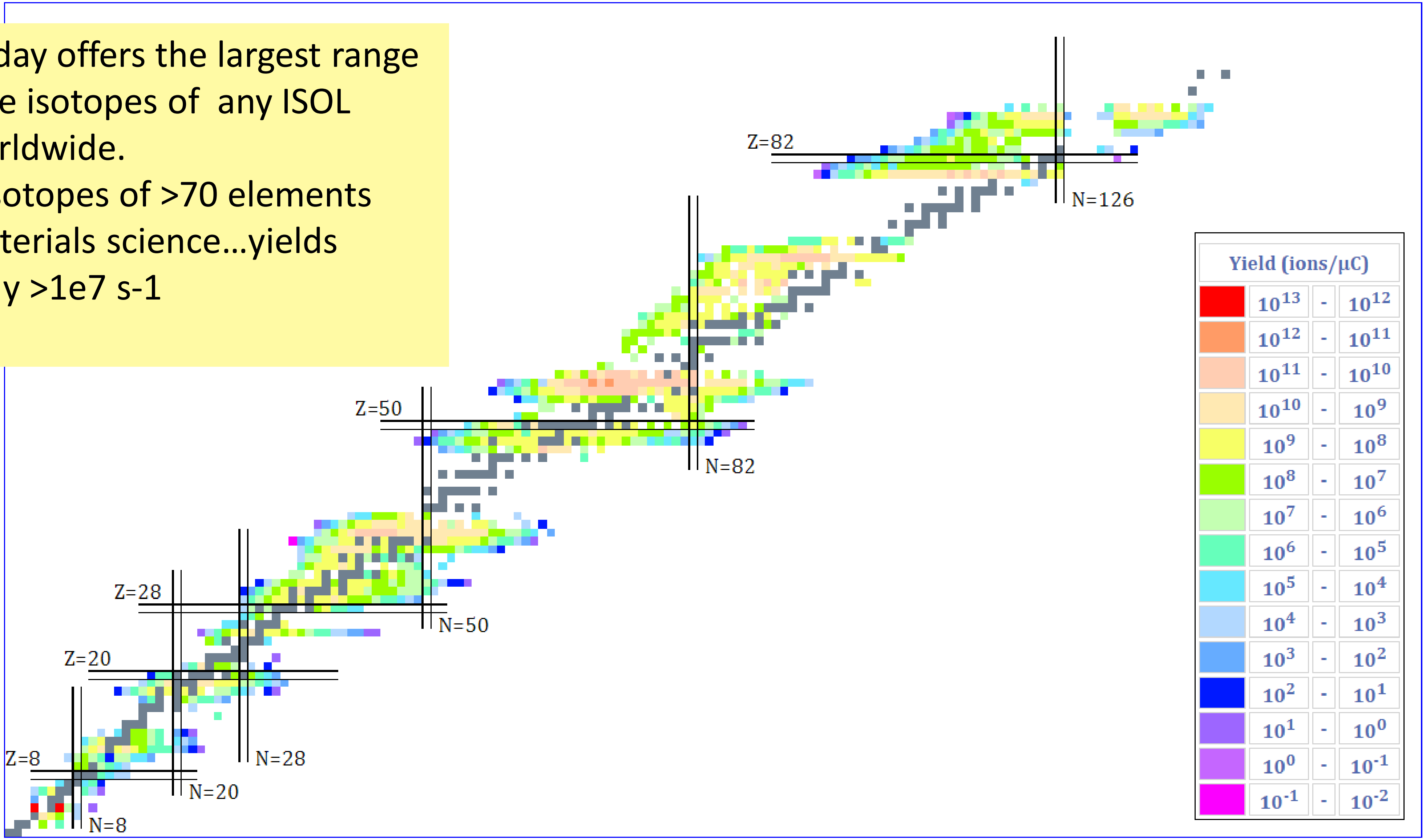
Oct 16th 2017: 50 years of beams at ISOLDE

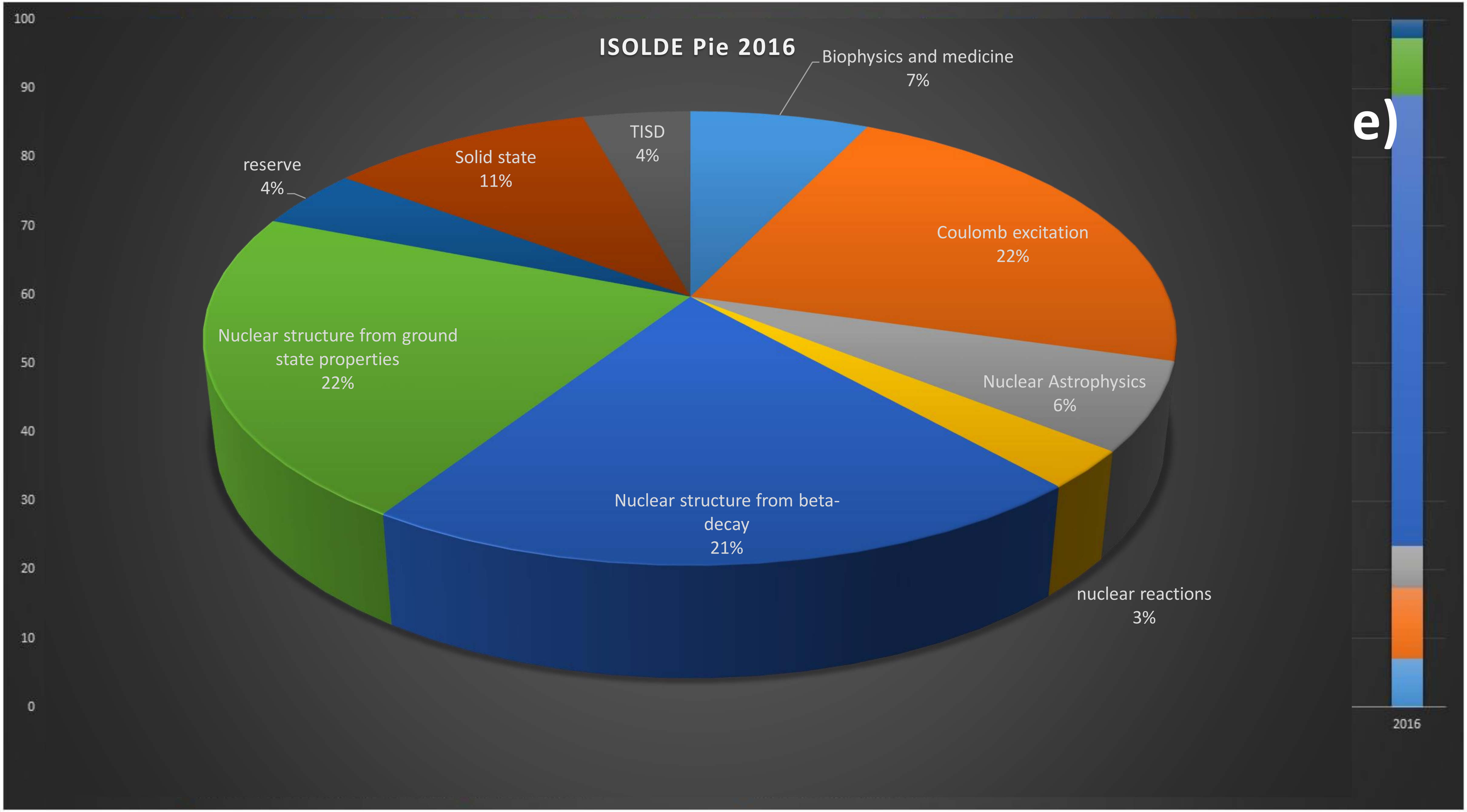
1975

Nuclear chart for ISOLDE

ISOLDE today offers the largest range of available isotopes of any ISOL facility worldwide.

- 1000 isotopes of >70 elements
- For materials science…yields typically >1e7 s-1




ISOLDE Pie 2016
Biophysics and medicine 7%
TISD 4%
Solid state 11%
reserve 4%
Coulomb excitation 22%
Nuclear structure from ground state properties 22%
Nuclear Astrophysics 6%
Nuclear structure from beta-decay 21%
nuclear reactions 3%
e)
100
90
80
70
60
50
40
30
20
10
0
2016

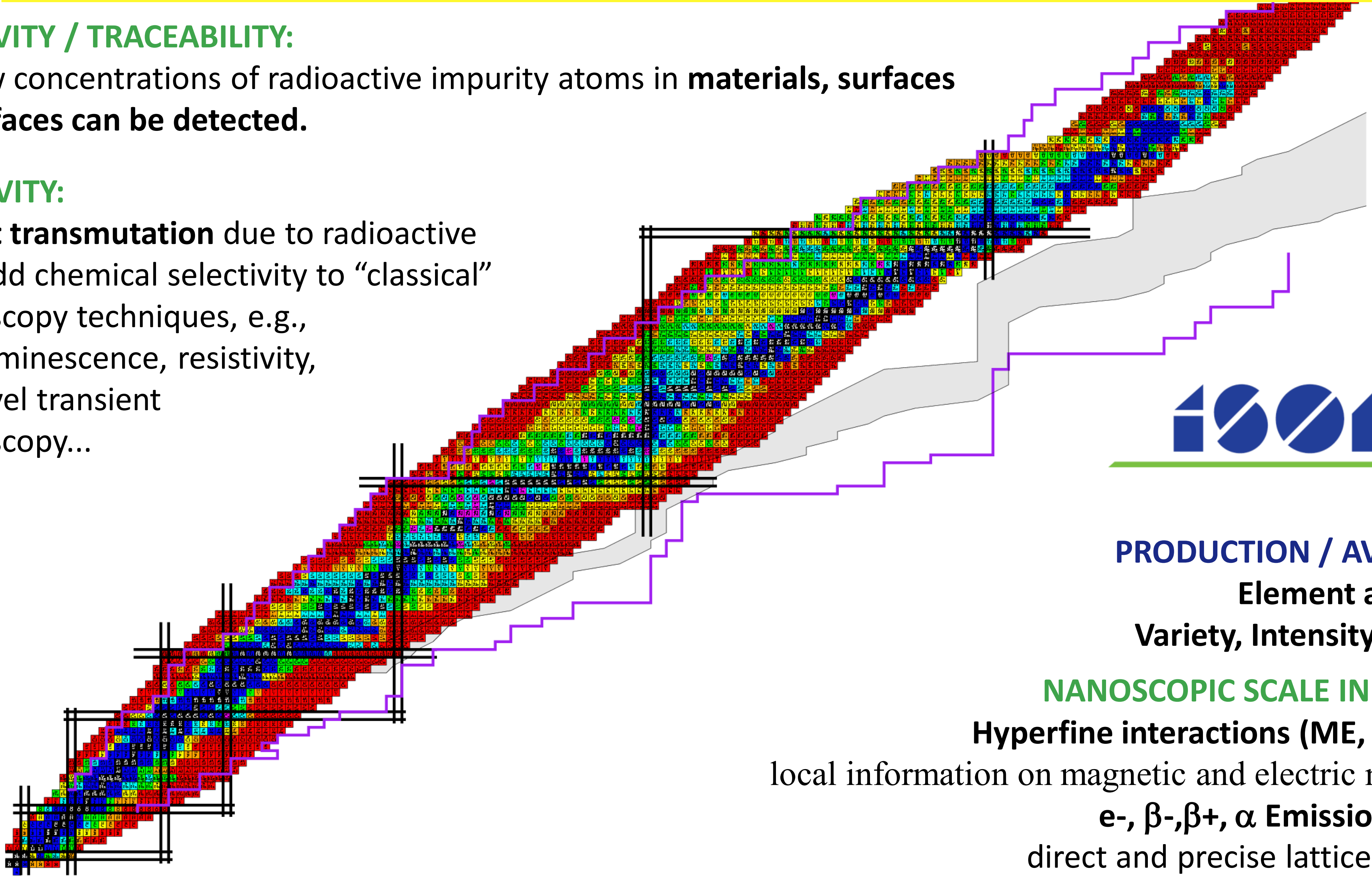
Key features of RIB for materials research

SENSITIVITY / TRACEABILITY:

Very low concentrations of radioactive impurity atoms in **materials, surfaces or interfaces can be detected.**

SELECTIVITY:

Element transmutation due to radioactive decay add chemical selectivity to "classical" spectroscopy techniques, e.g., photoluminescence, resistivity, deep level transient spectroscopy...



ISOLDE

PRODUCTION / AVAILABILITY

Element and isotope Variety, Intensity and Purity

NANOSCOPIC SCALE INFORMATION:

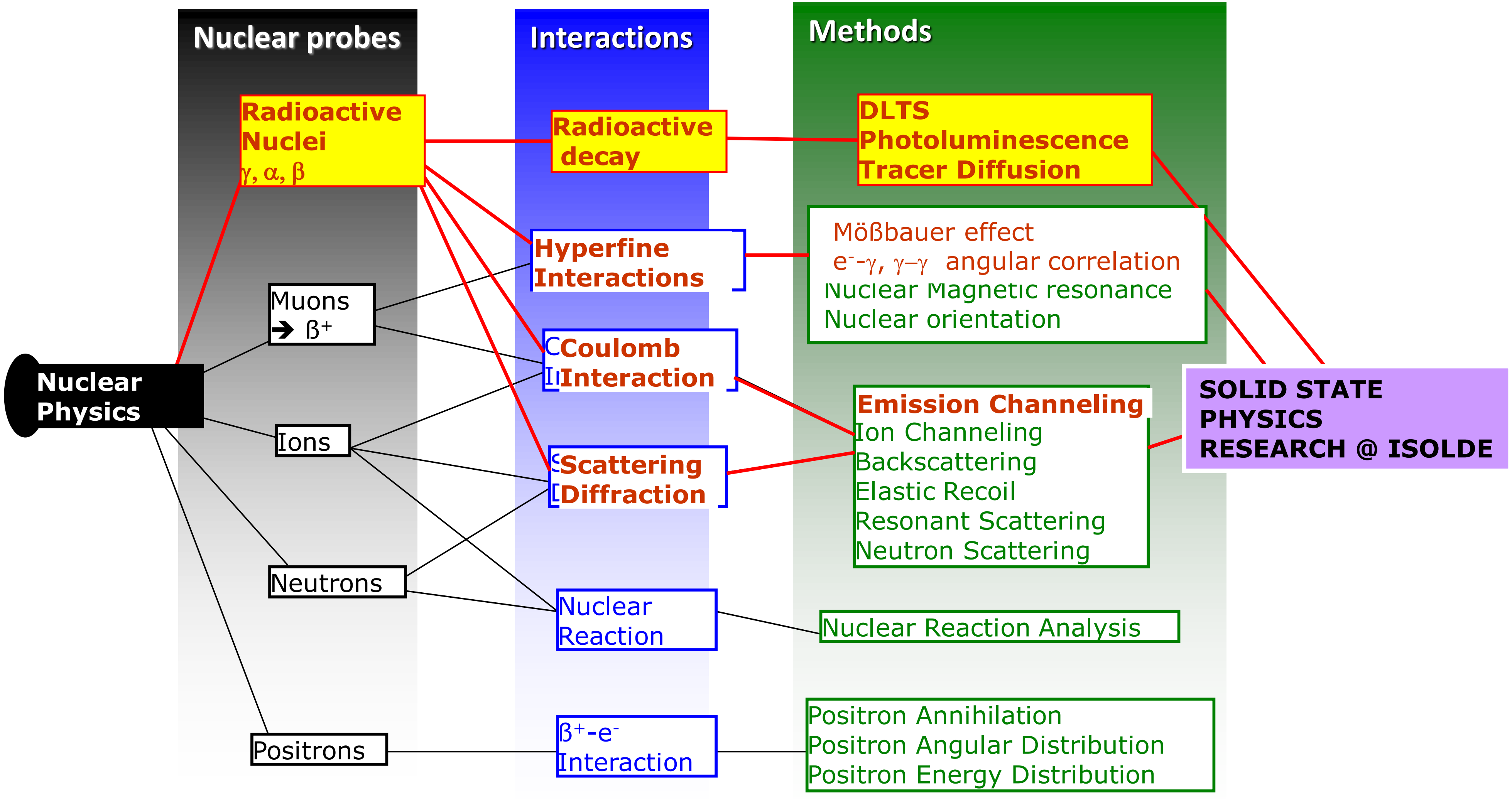
Hyperfine interactions (ME, PAC, β-NMR)

local information on magnetic and electric neighborhood

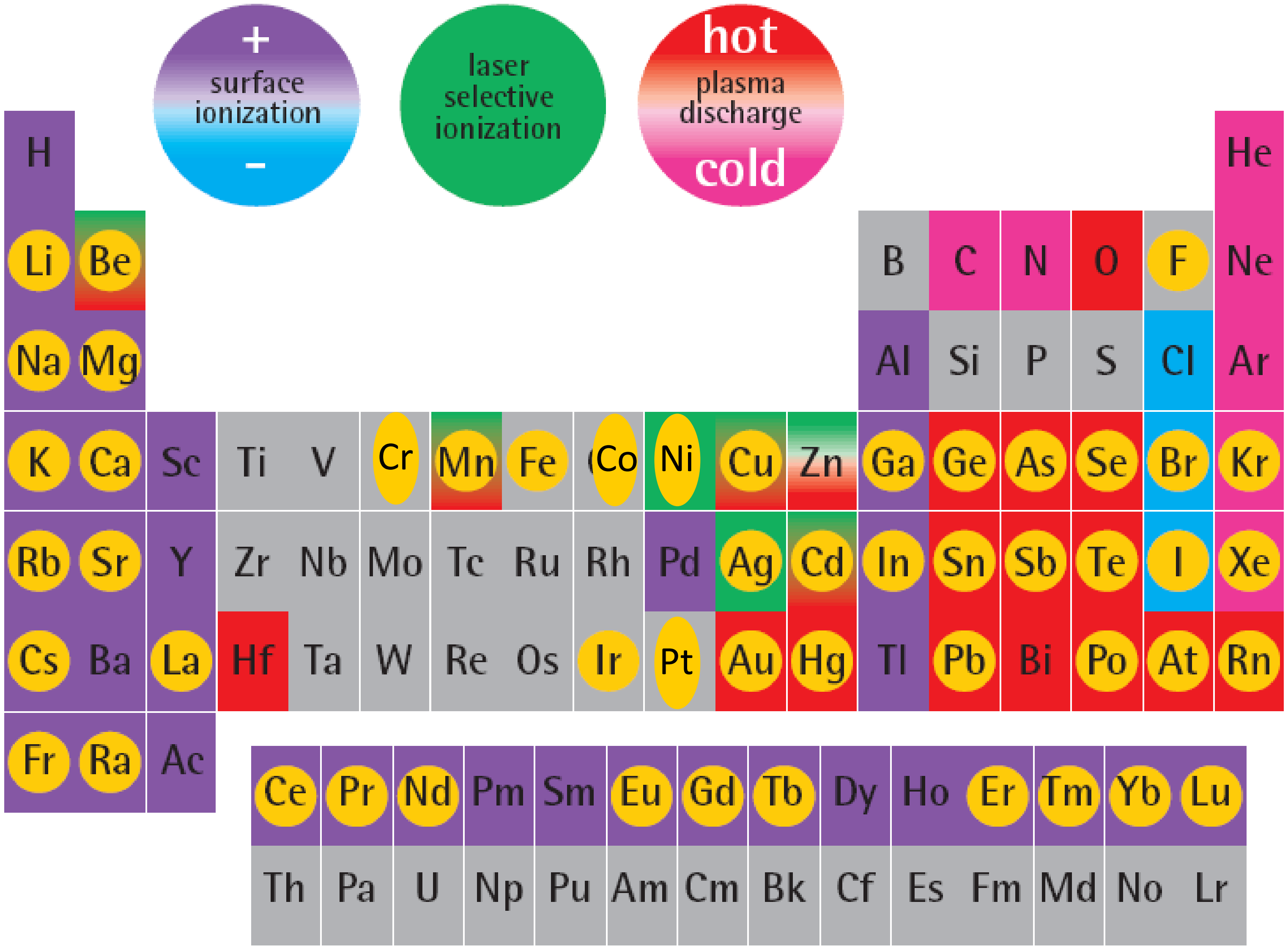
e-, β-,β+, α Emission Channeling

direct and precise lattice site location.

Applying radioactivity to solid state physics



ISOLDE table of elements



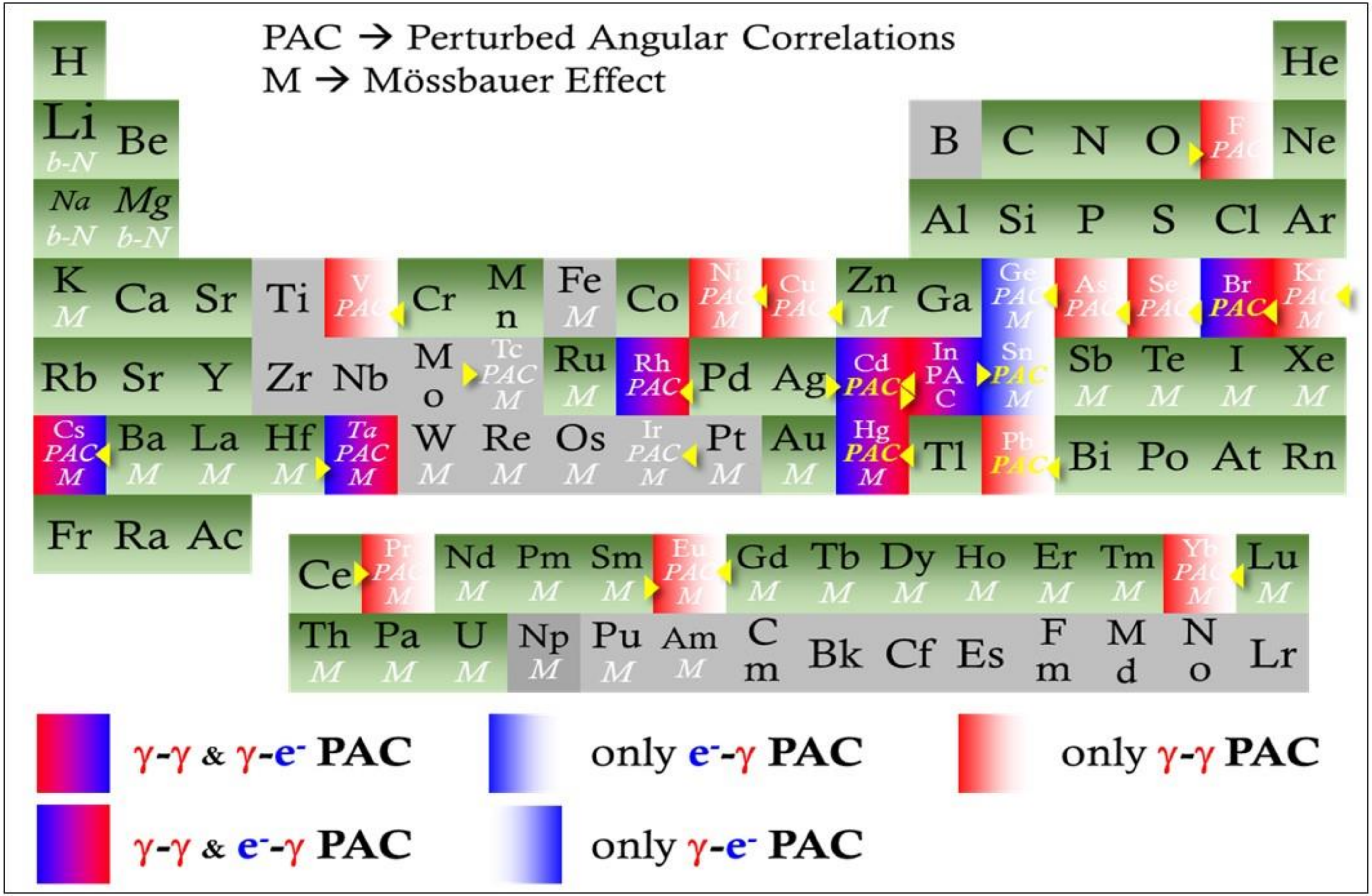
Workhorse probes:

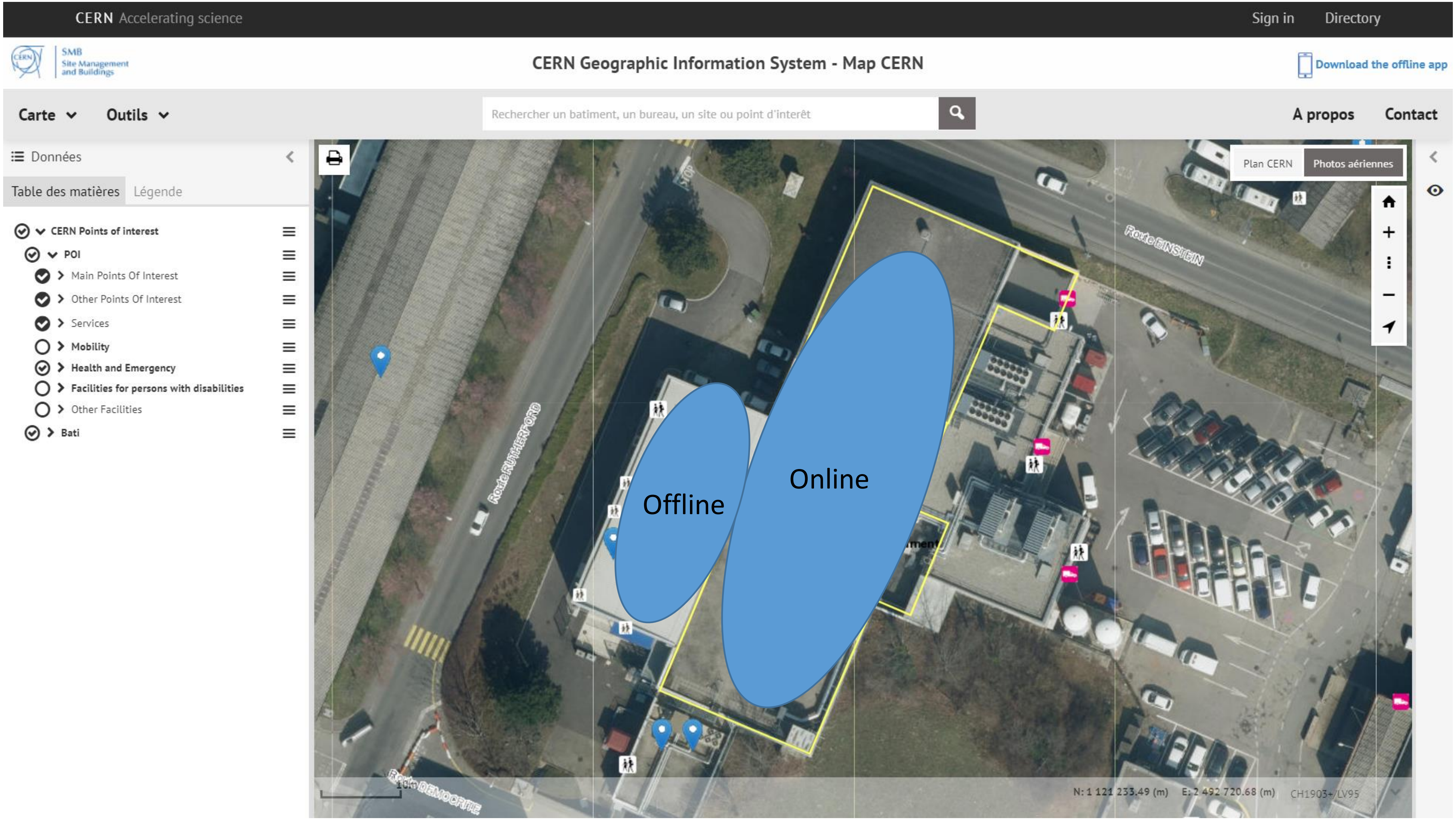
^{111}Cd, ^{199}Hg, ^{117}Cd, ^{57}Mn, ^{73}As

New promising probes:

^{68}Cu, ^{149}Gd, ^{172}Lu, ^{151}Gd, ^{197}Hg

Isotopes of this element used for solid state physics or life science


PAC → Perturbed Angular Correlations
M → Mössbauer Effect
H
He
Li b-N
Be
B C N O
F PAC
Ne
Na b-N
Mg b-N
Al Si P S Cl Ar
K M
Ca Sr Ti
V PAC
Cr Mn
Fe M
Co
Ni PAC M
Cu PAC
Zn M
Ga
Ge PAC M
As PAC
Se PAC
Br PAC
Kr PAC M
Rb Sr Y Zr Nb Mo
Tc PAC M
Ru M
Rh PAC
Pd Ag
Cd PAC
In PAC
Sn PAC M
Sb M
Te M
I M
Xe M
Cs PAC M
Ba M
La M
Hf M
Ta PAC M
W M
Re M
Os M
Ir PAC M
Pt M
Au M
Hg PAC M
Tl
Pb PAC
Bi Po At Rn
Fr Ra Ac
Ce
Pr PAC M
Nd M
Pm M
Sm M
Eu PAC M
Gd M
Tb M
Dy M
Ho M
Er M
Tm M
Yb PAC M
Lu M
Th M
Pa M
U M
Np M
Pu M
Am M
Cm Bk Cf Es Fm Md No Lr
γ-γ & γ-e⁻ PAC
only e⁻-γ PAC
only γ-γ PAC
γ-γ & e⁻-γ PAC
only γ-e⁻ PAC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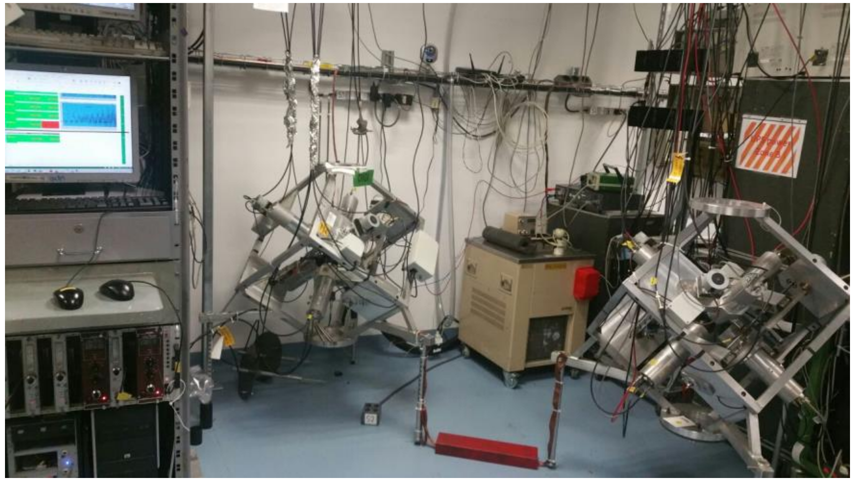
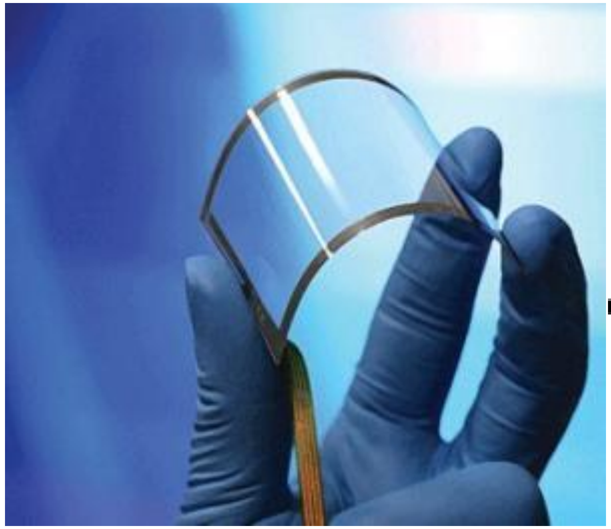
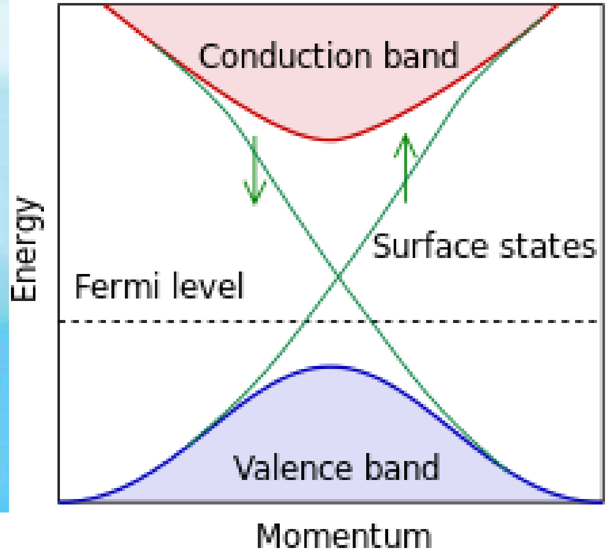
Services
Mobility
Health and Emergency
Facilities for persons with disabilities
Other Facilities
Bati
Plan CERN
Photos aériennes
Route EINSTEIN
Route RUTHERFORD
Offline
Online
N: 1 121 233.49 (m)
E: 2 492 720.68 (m)
CH1903+/LV95

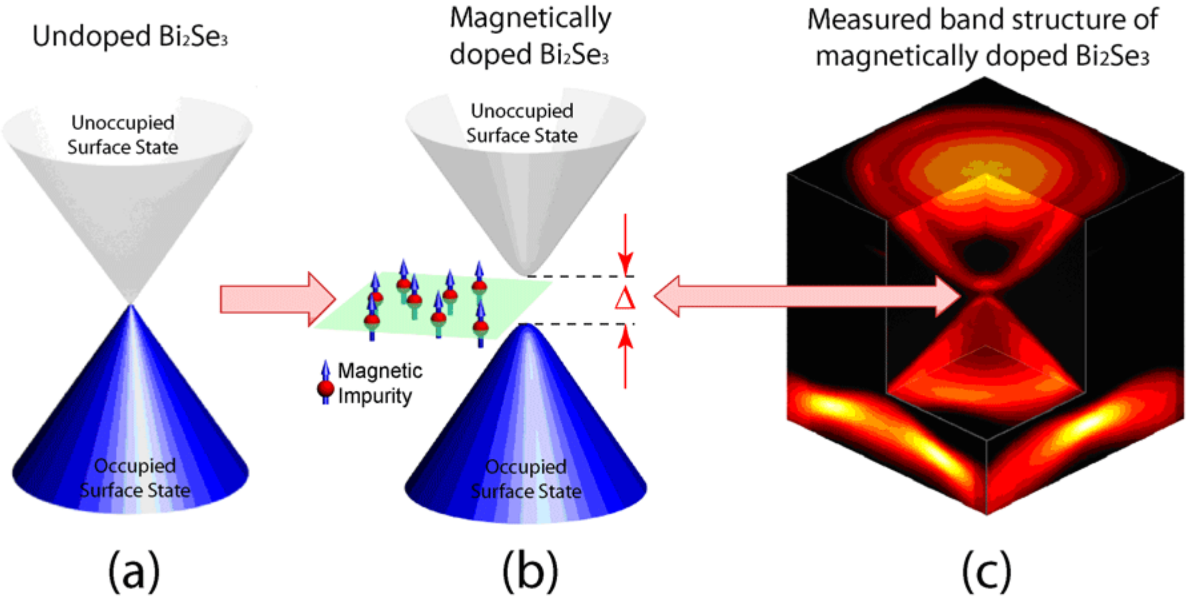
Offline labs at ISOLDE: B. 508



Staying relevant: Materials studied at ISOLDE

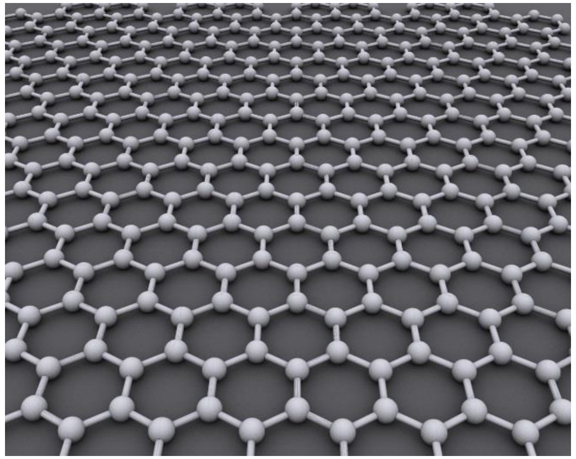






graphene

Topological Insulators



IOP Publishing

Journal of Phy

J. Phys. D: Appl. Phys. **50** (2017) 393002 (26pp)

https://doi.org/10.

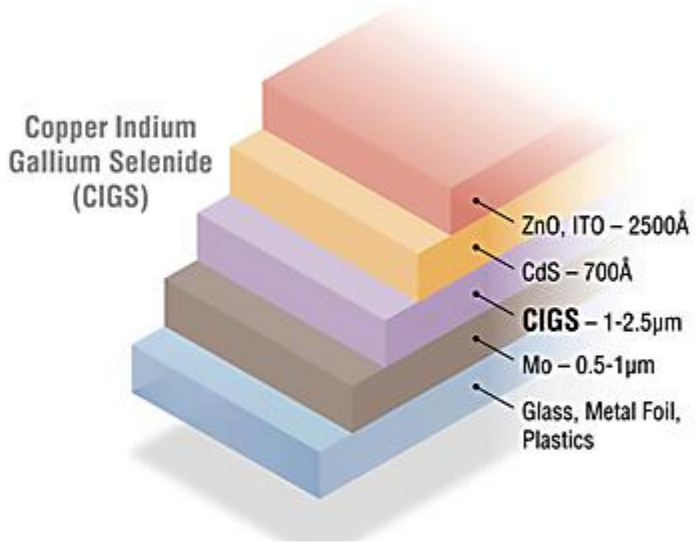
Topical Review

Experimentally evaluating the origin of dilute magnetism in nanomaterials

L M C Pereira

KU Leuven, Instituut voor Kern- en Stralingsfysica, 3001 Leuven, Belgium

Hyperfine techniques are particularly successful in unravelling subtle magnetic behaviour in materials


Copper Indium
Gallium Selenide
(CIGS)
ZnO, ITO – 2500Å
CdS – 700Å
CIGS – 1-2.5µm
Mo – 0.5-1µm
Glass, Metal Foil,
Plastics

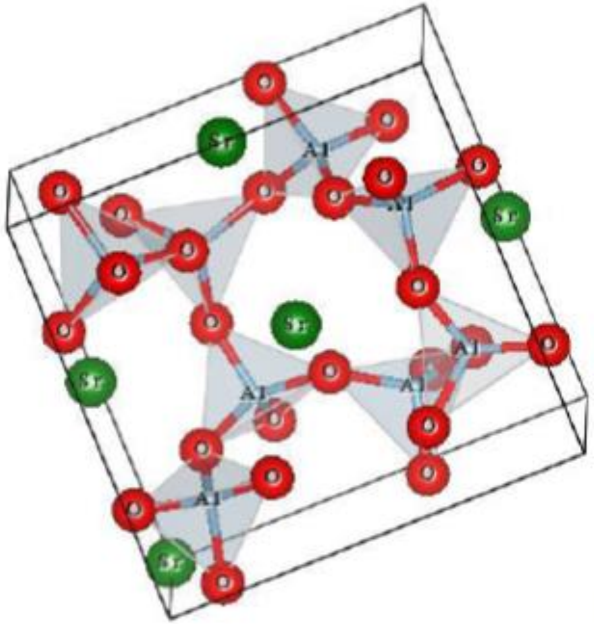
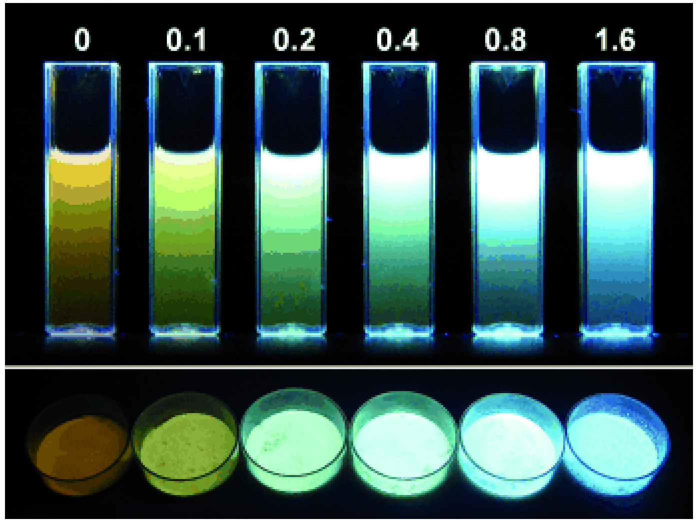


Fig. 1



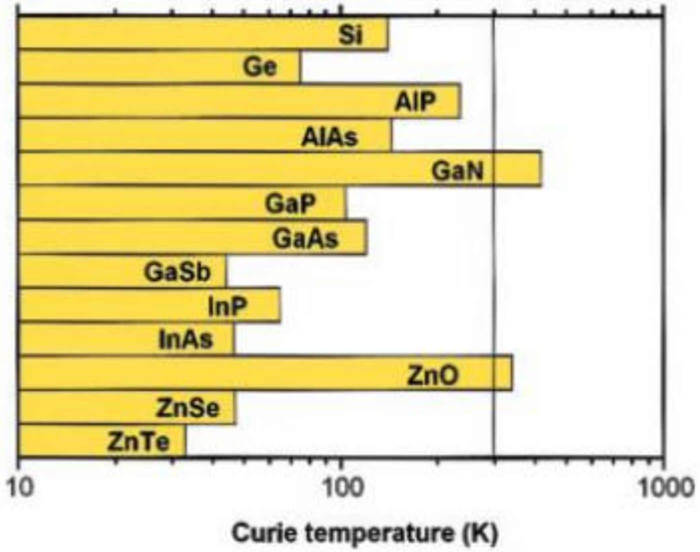
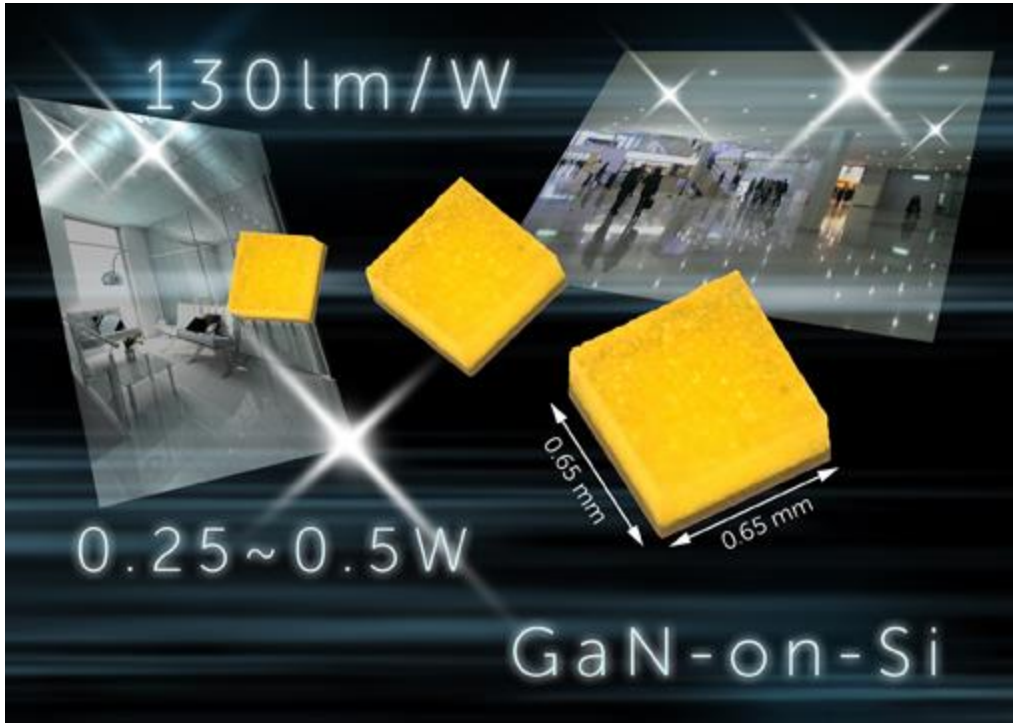


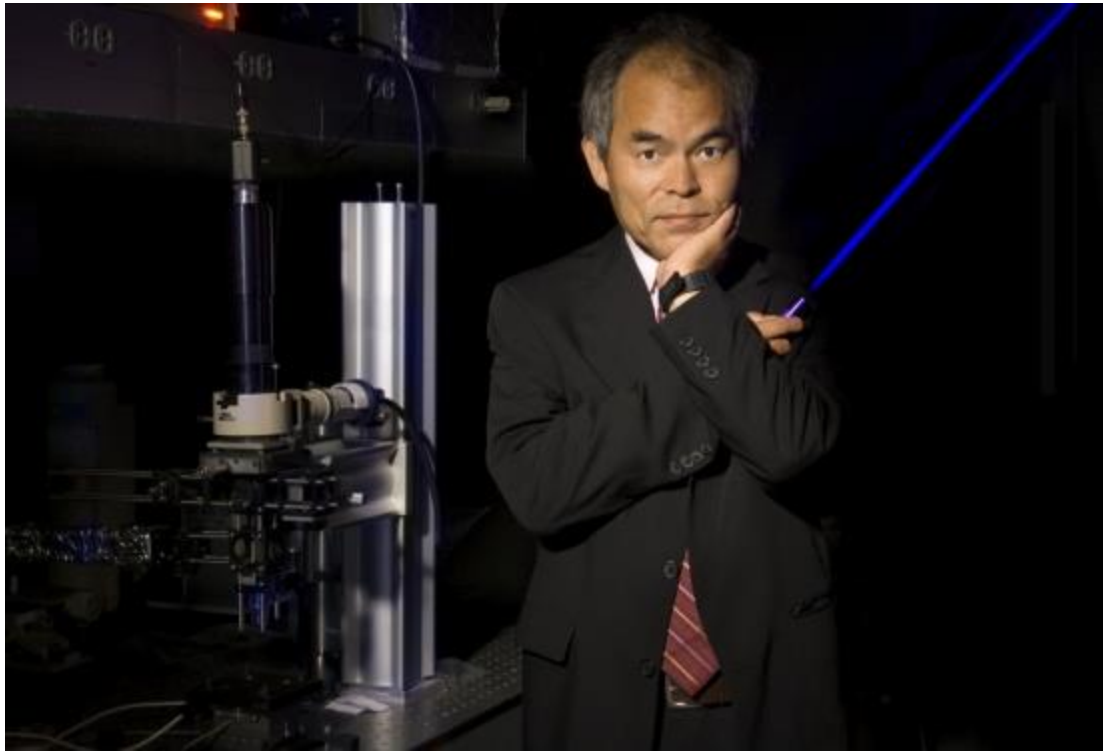
Fig. 3. Computed values of the Curie temperature T_C for various p-type semiconductors containing 5% of Mn and 3.5×10^{20} holes cm^3.

Dietl *et al*, *Science* 287 (2000) 1

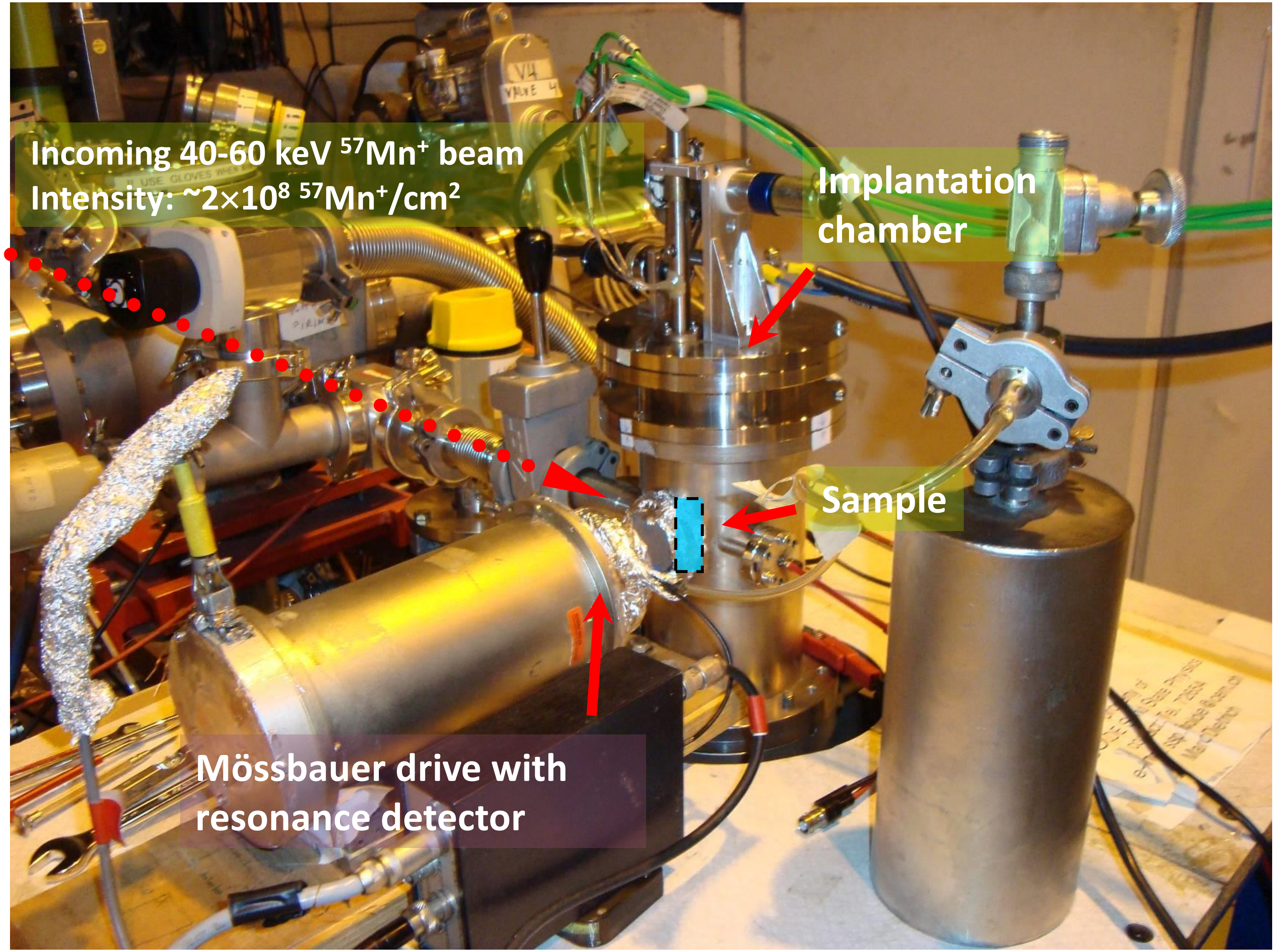
Is it possible to create magnetic semiconductors that work at room temperature?
Such devices have been demonstrated at low temperatures but not yet in a range warm enough for spintronics applications.

Next generation semiconductors : doping





Hyperfine Interactions with Mossbauer spectroscopy

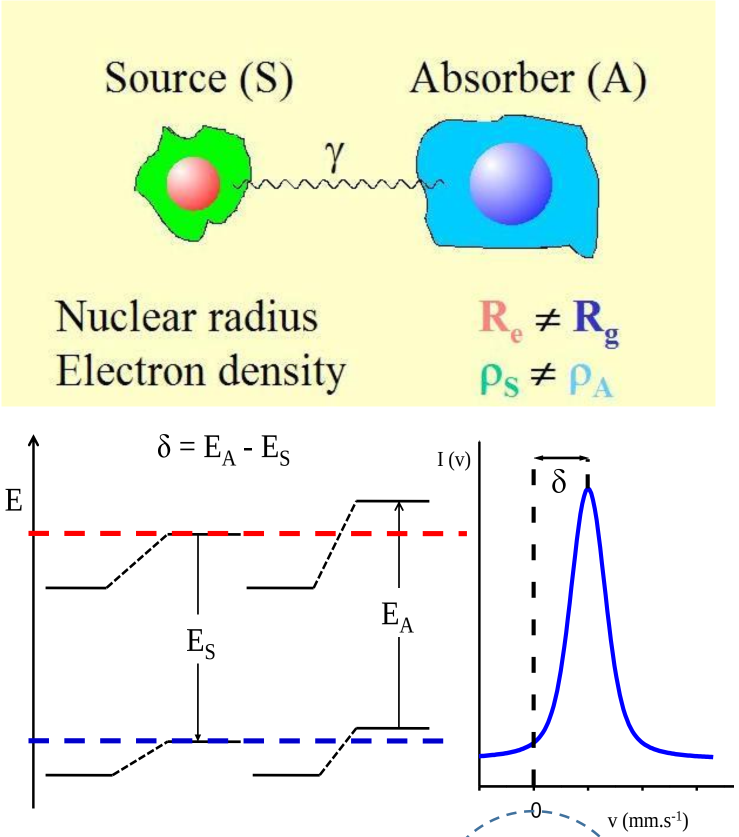


Laser Ionised **^{57}Mn** beam : a new era for Mossbauer experiments at ISOLDE. 20th anniversary in 2016

- 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….

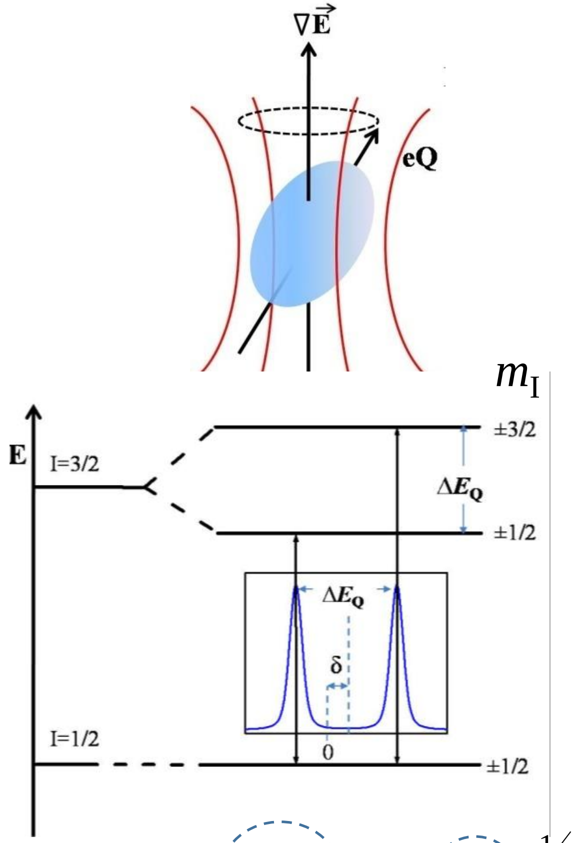
Isomer Shift



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

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

Fe: ZnO a ferromagnetic semiconductor? (no!)

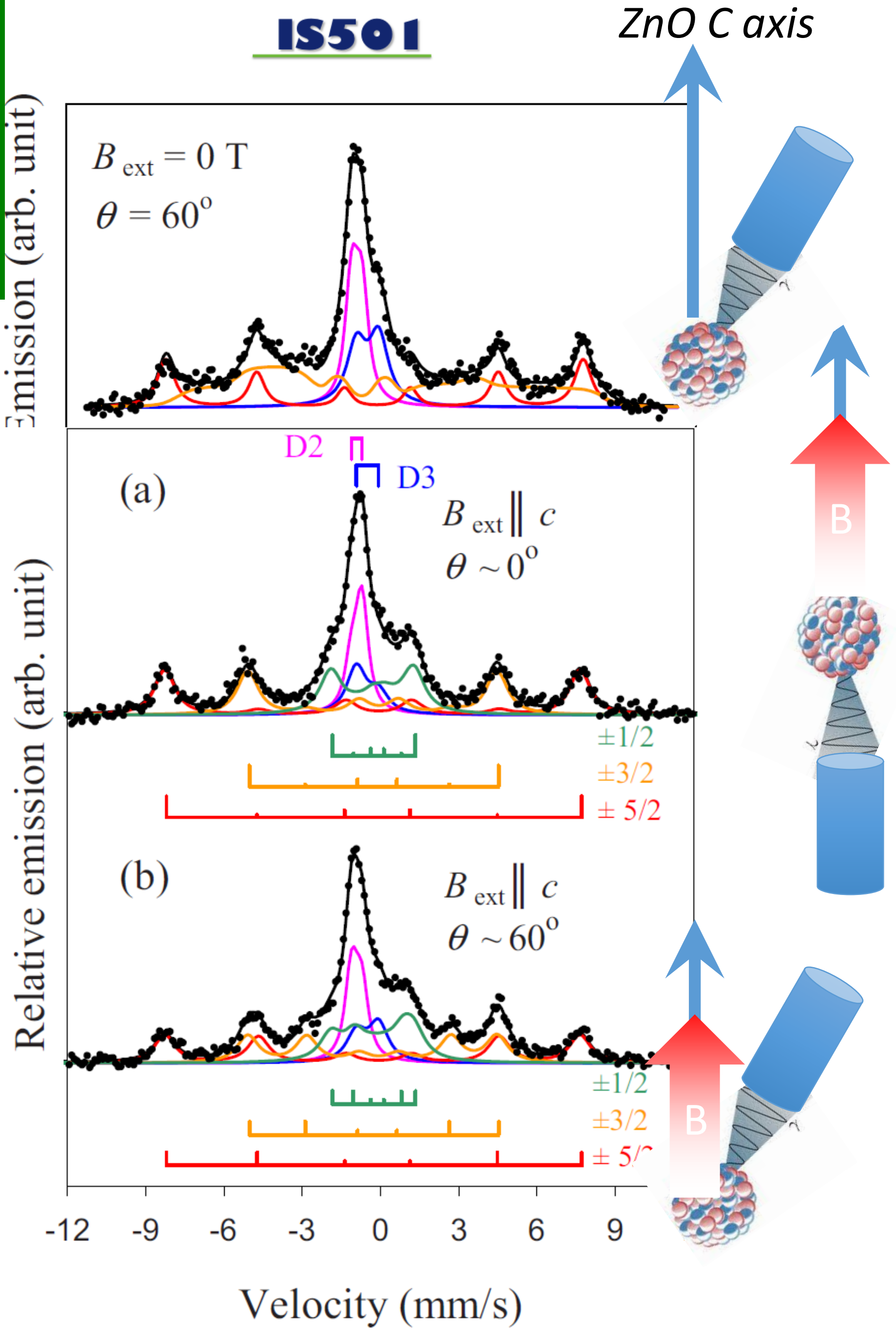
6 fold spectrum: characteristic of magnetic structure (at room temperature!!!).

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

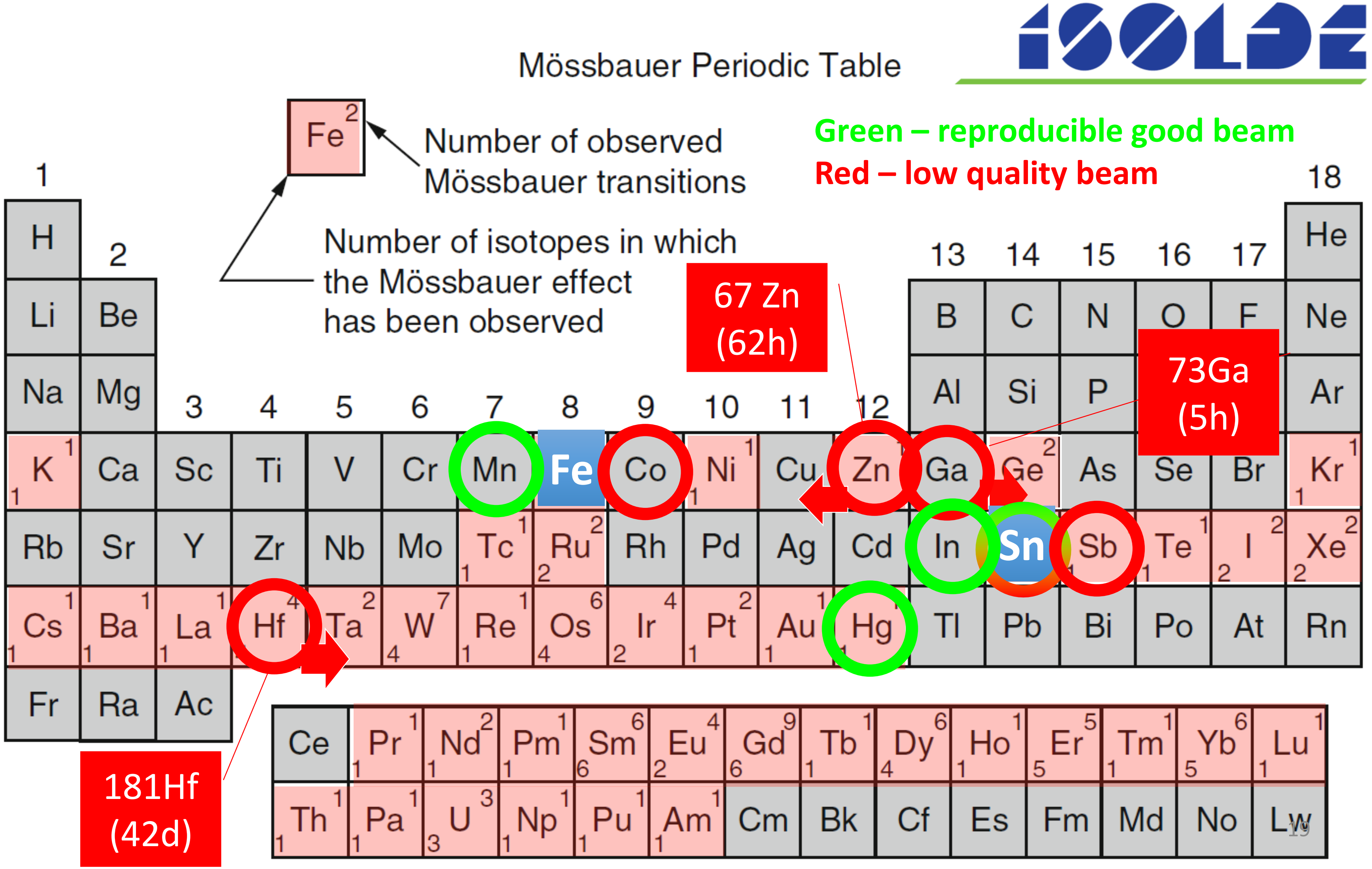
Gunnlaugsson *et al* (APL **97** 142501 2010)

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).*

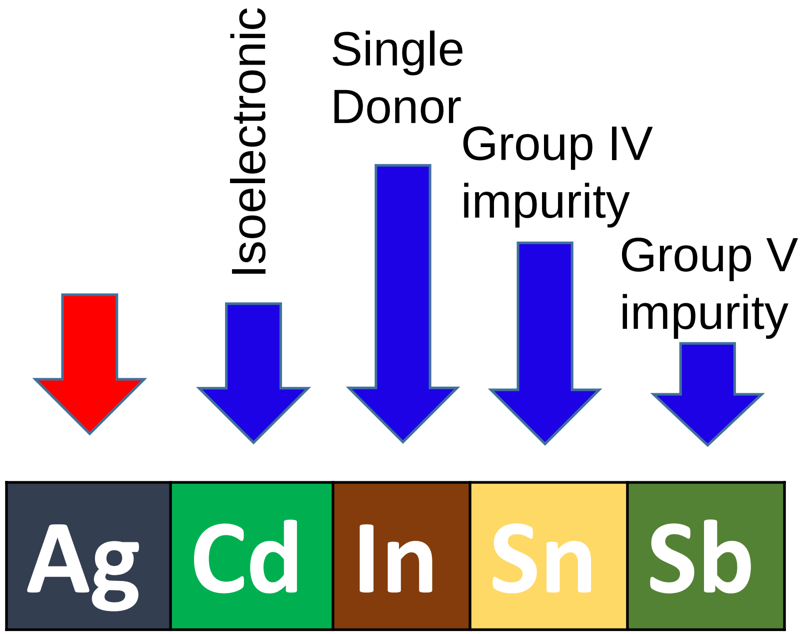
Gunnlaugsson *et al* APL **100** 042109 **(**2012)



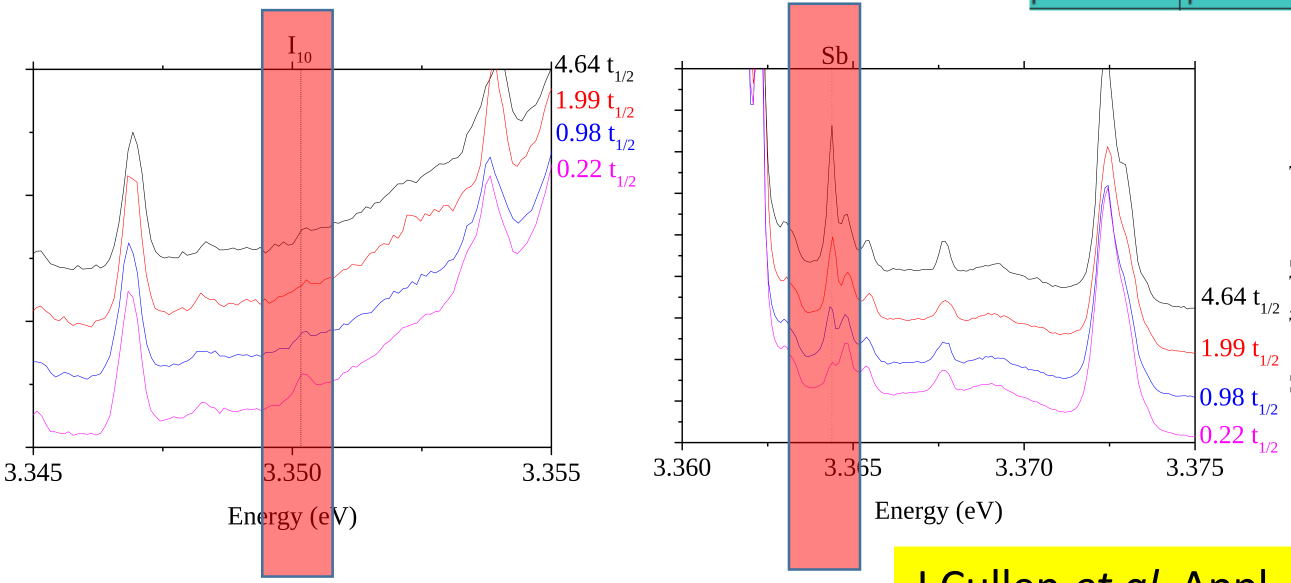
Mössbauer periodic table

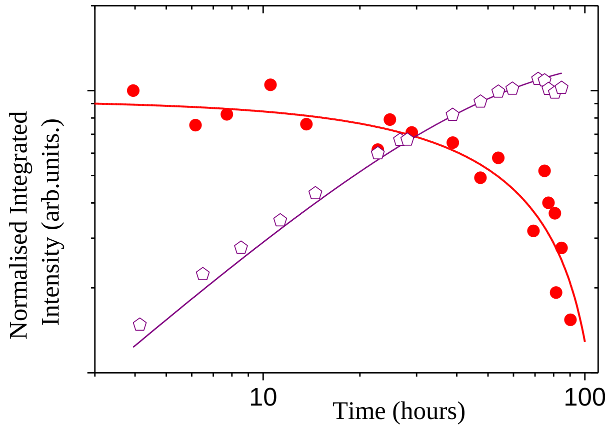


ZnO: wide band gap semiconductor



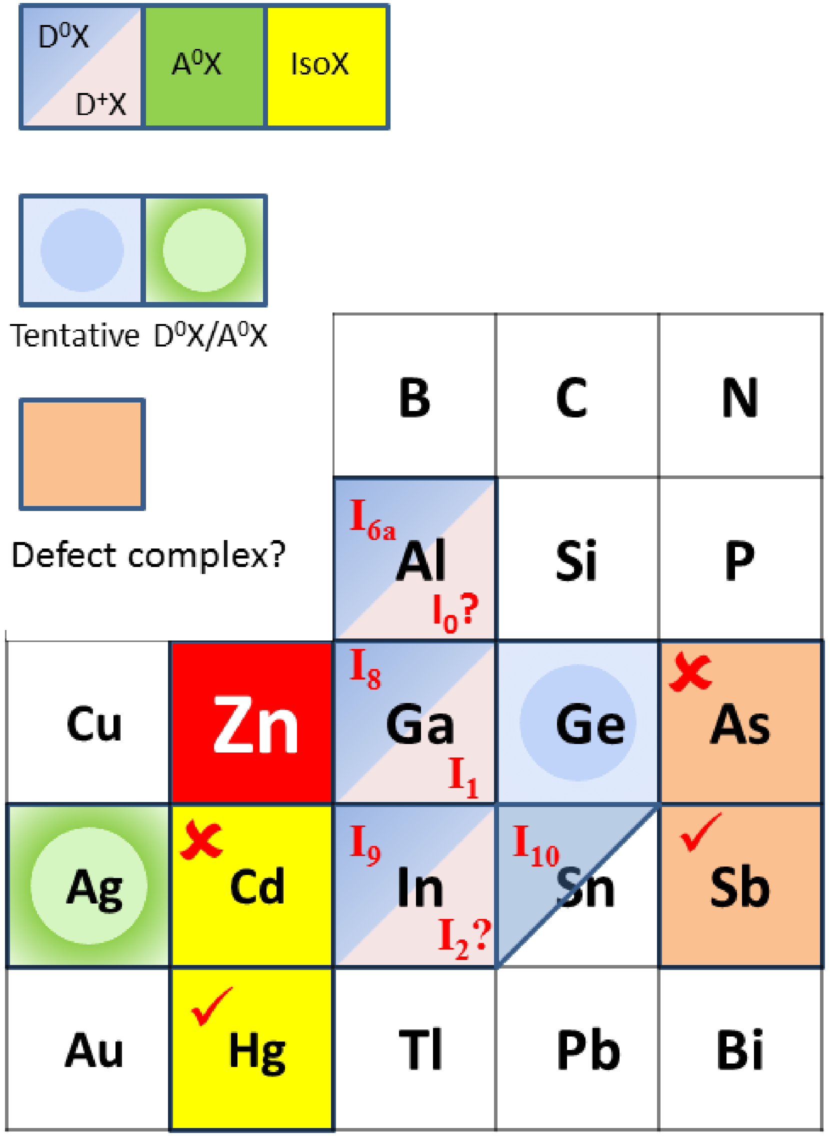
Sb121 5/2+ * 57.36	Sb122 2.70 d 2- * EC,β-	Sb123 7/2+ * 42.64	Sb124 60.20 d 3- * β-	Sb125 2.7582 y 7/2+ * β-
Sn120 0+ * 32.59	Sn121 27.06 h 3/2+ * β-	Sn122 0+ * 4.63	Sn123 129.2 d 11/2- * β-	Sn124 0+ * 5.79
In119 2.4 m 9/2+ * β-	In120 3.08 s 1+ * β-	In121 23.1 s 9/2+ * β-	In122 1.5 s 1+ * β-	In123 5.98 s 9/2+ * β-
Cd118 50.3 m 0+ β-	Cd119 2.69 m 3/2+ * β-	Cd120 50.80 s 0+ β-	Cd121 13.5 s (3/2+) * β-	Cd122 5.24 s 0+ β-
Ag117 72.8 s (1/2-) * β-	Ag118 3.76 s (1)- * β-	Ag119 2.1 s (7/2+) * β-	Ag120 1.23 s * β-	Ag121 0.78 s (7/2+) β-n





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

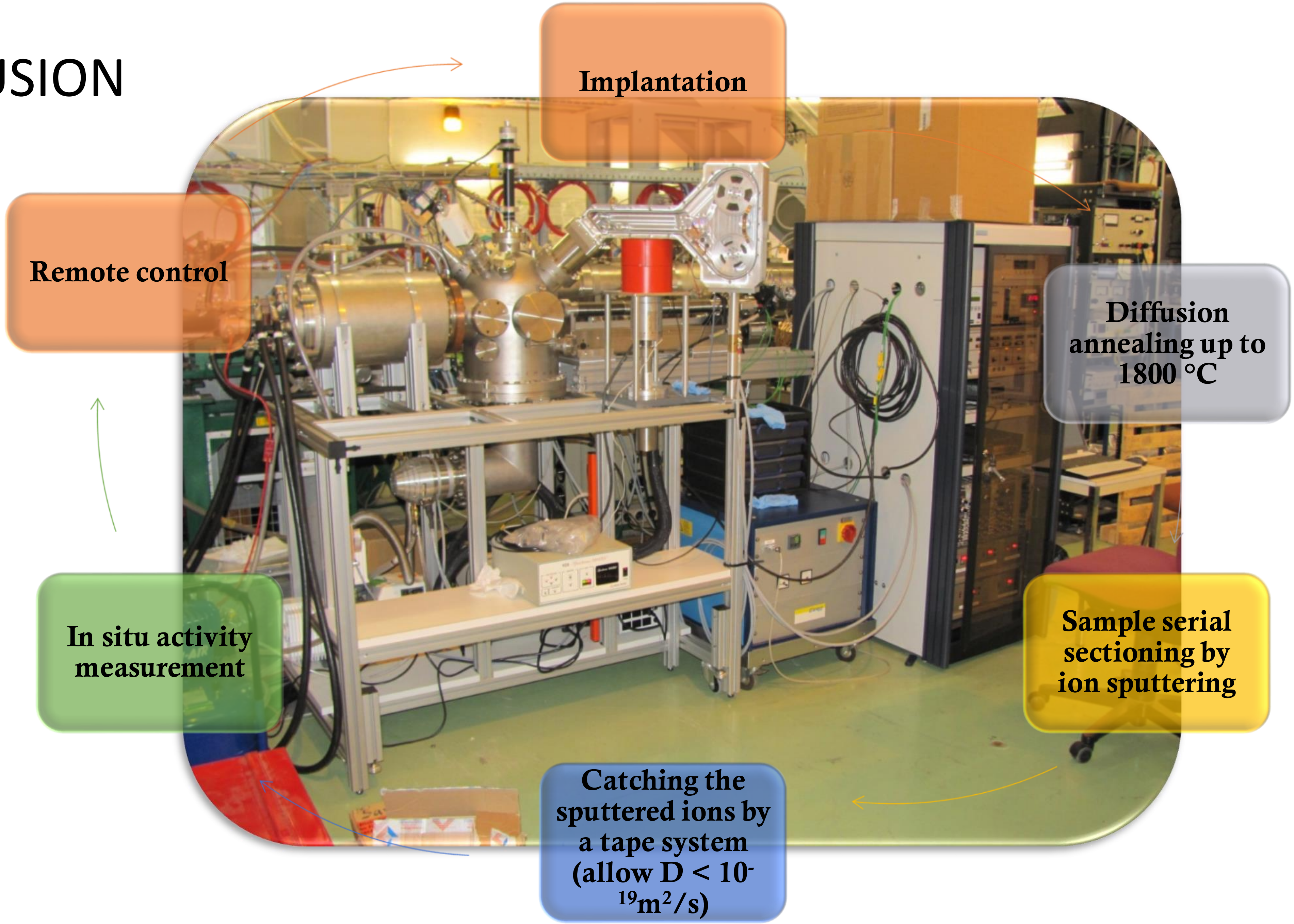
Radiotracer PL has allowed for the full classification of the dominant impurities in ZnO



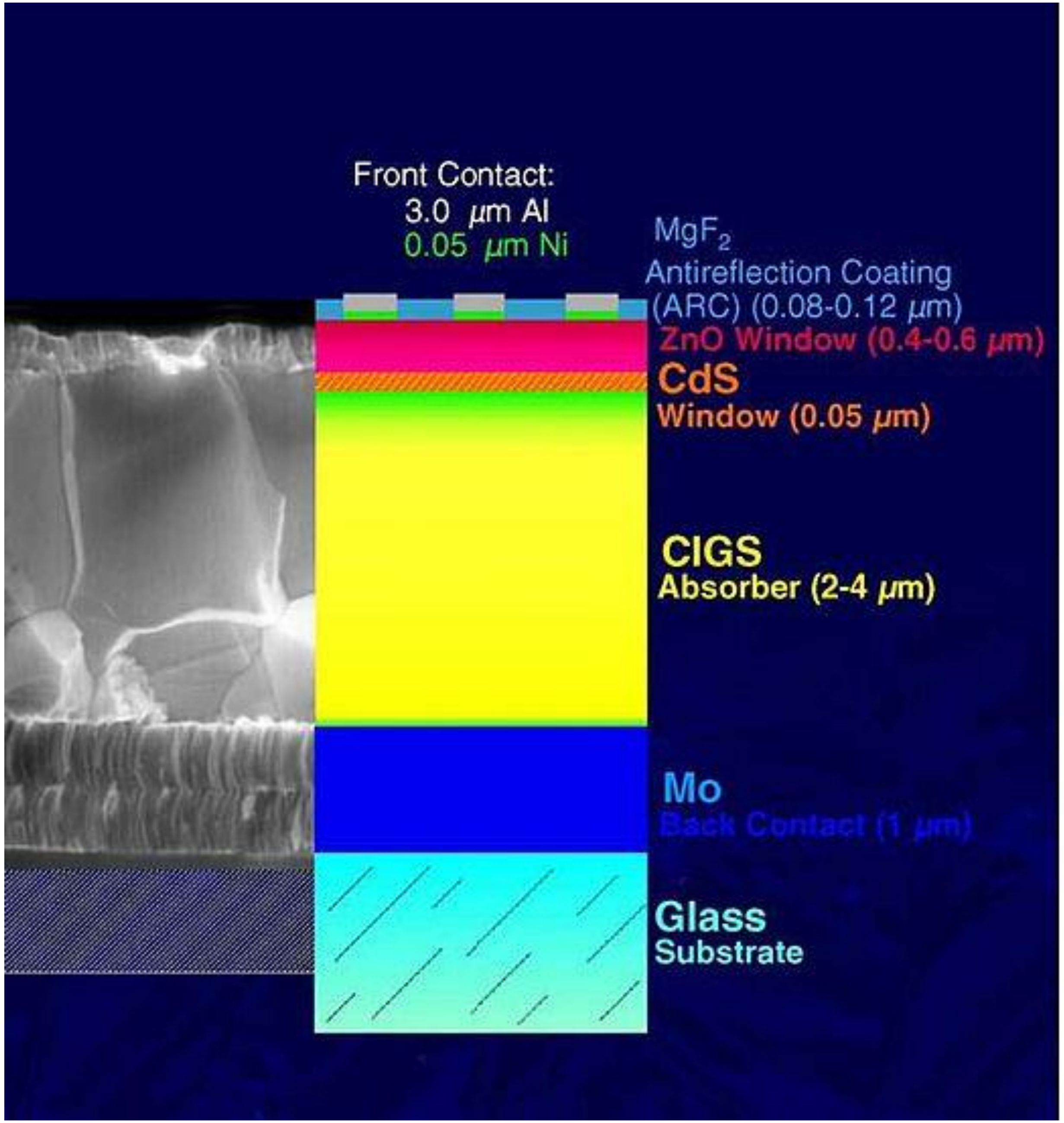
- Radio PL allows for the subtle chemical identification of luminescence through different decay chains.
- Has allowed for the identification of neutral and ionised donors [1, 2], complexed impurities [3], "double donor" centres [2, 4], and isoelectronic centres [5].

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)

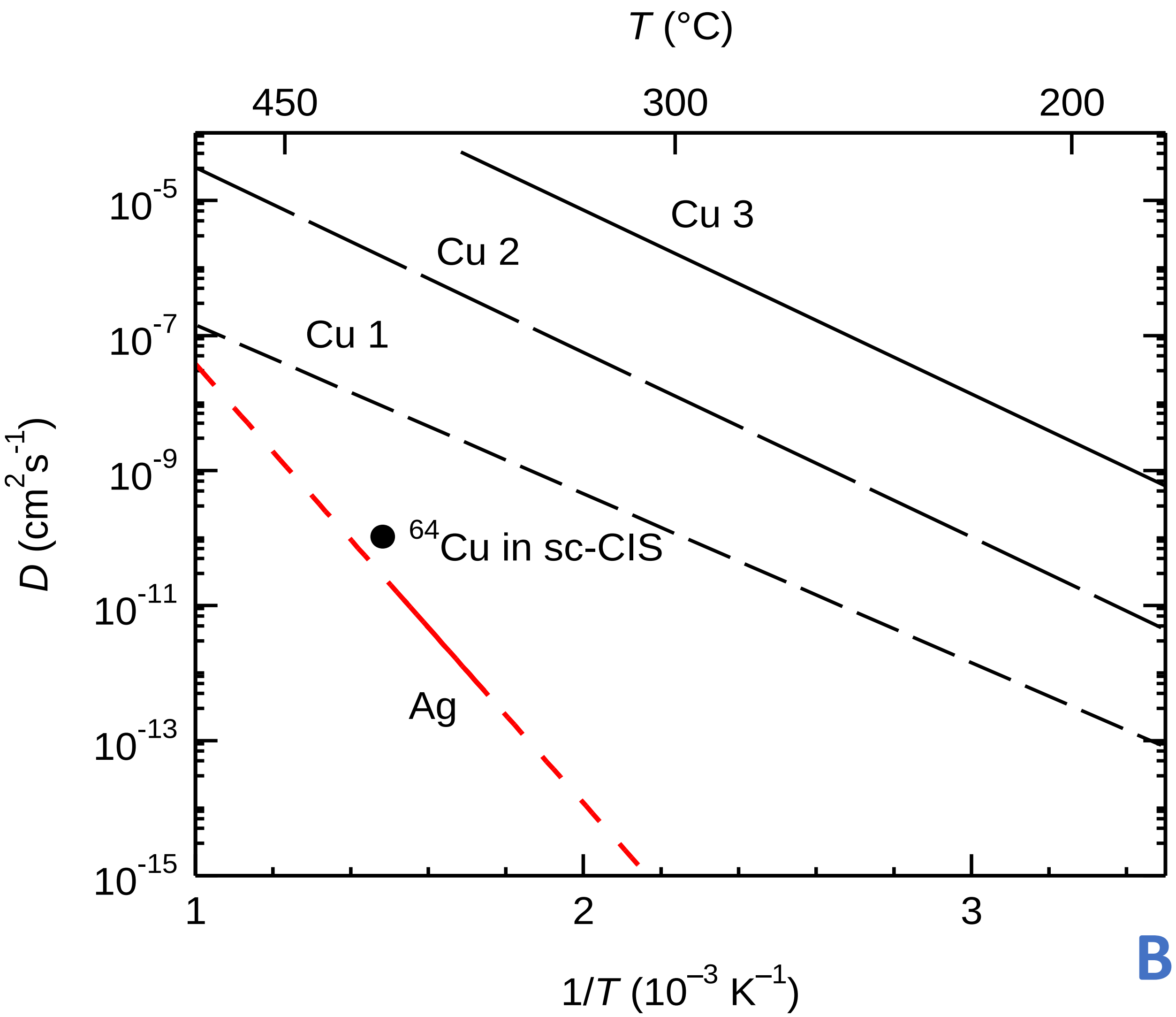
DIFFUSION


Implantation
Remote control
Diffusion annealing up to 1800 °C
In situ activity measurement
Sample serial sectioning by ion sputtering
Catching the sputtered ions by a tape system (allow D < 10-19m2/s)

New collaboration with Uni Munster: diffusion processes in CIGS (Copper Indium Gallium Selenide) solar cells: in particular Cu



Why? Cu diffusion in CIGS: the entire literature...



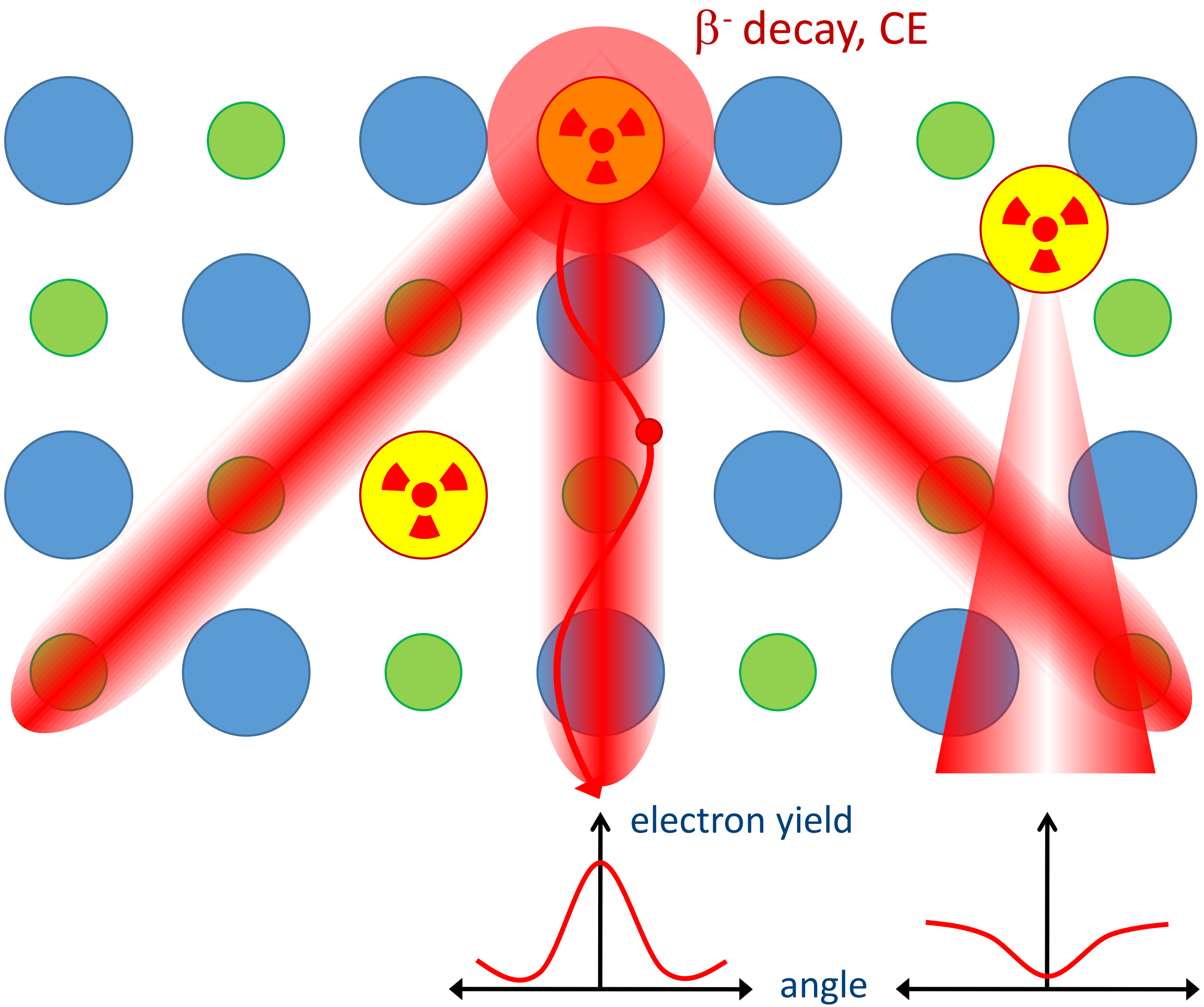
Lubomirsky et al., JAP 83 (1998) 4678

Cu 1, Cu 2, Cu 3:
- various authors,
- various (electrical) methods

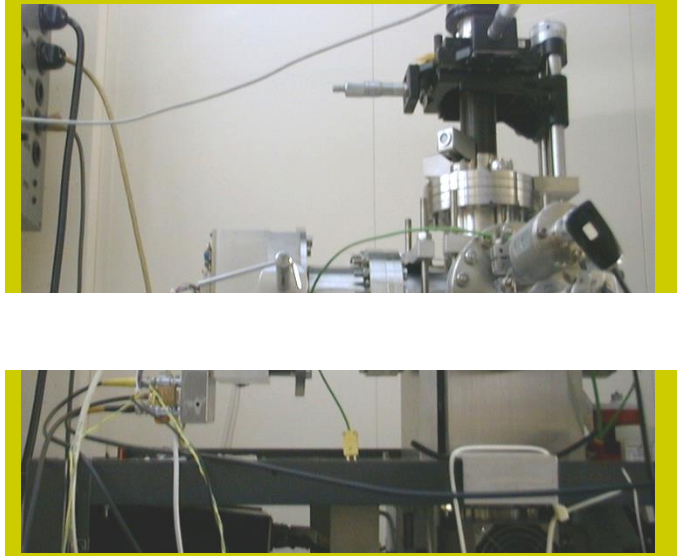
^{64}Cu:
- tracer experiment
- bulk single-crystal CIS

Beamtime scheduled for Nov 2017

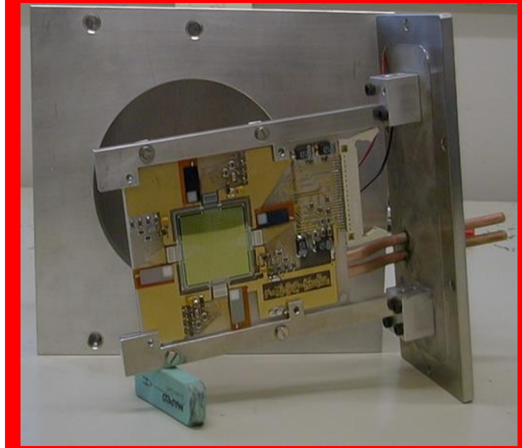
Emission channeling: (β^-, β^+, c.e., α)



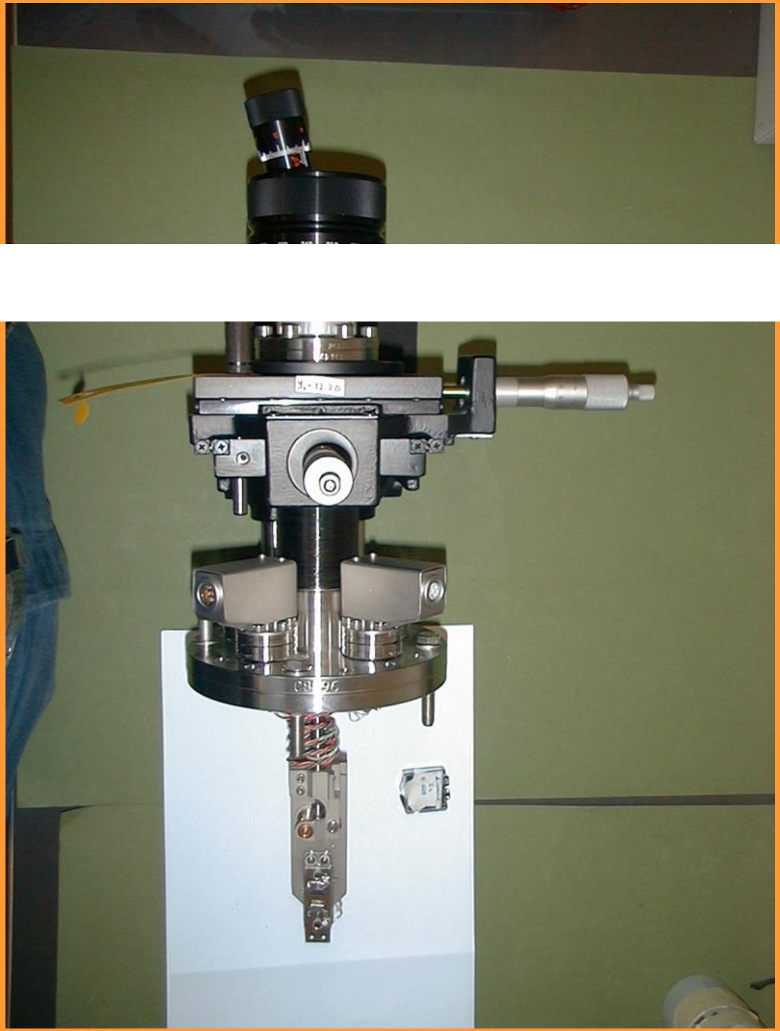
What do you need to do Emission Channeling



Vacuum chamber

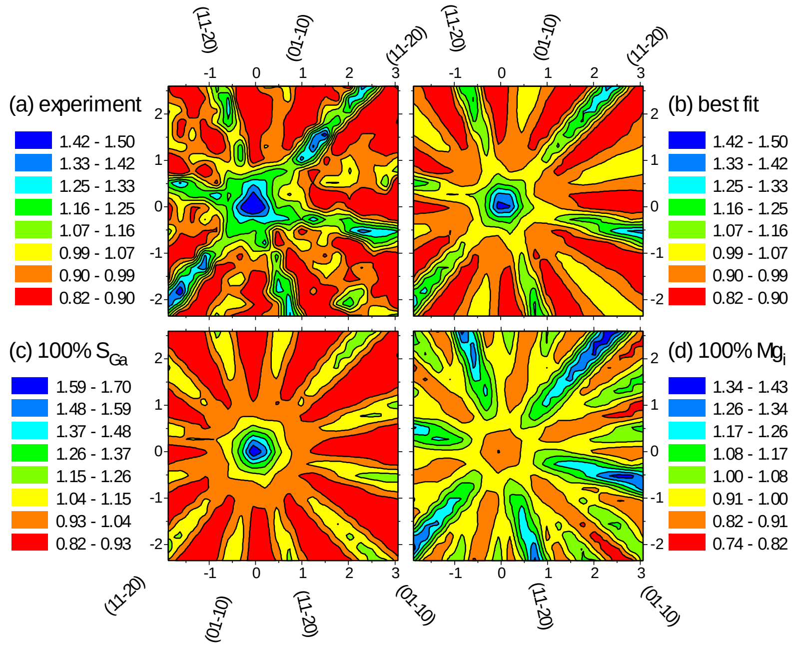


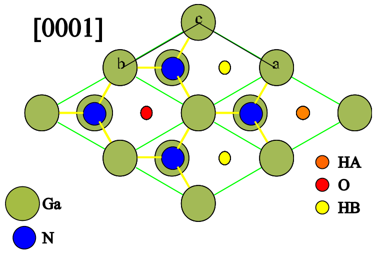
Position-sensitive detector

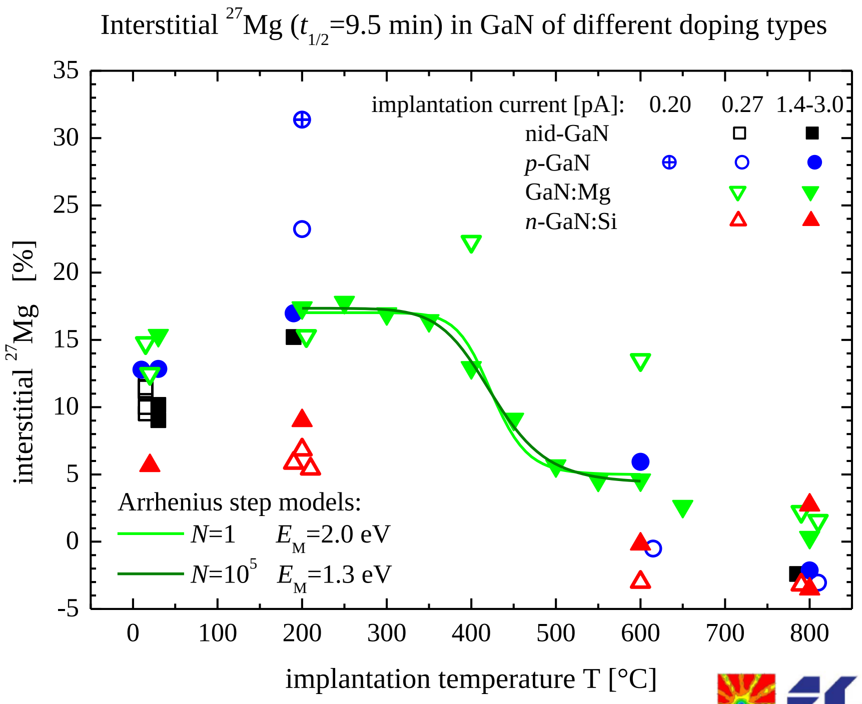


Goniometer

Lattice sites of ^{27}Mg in different pre-doped GaN







- Electron emission channeling patterns show mix of substitutional + interstitial ^{27}Mg

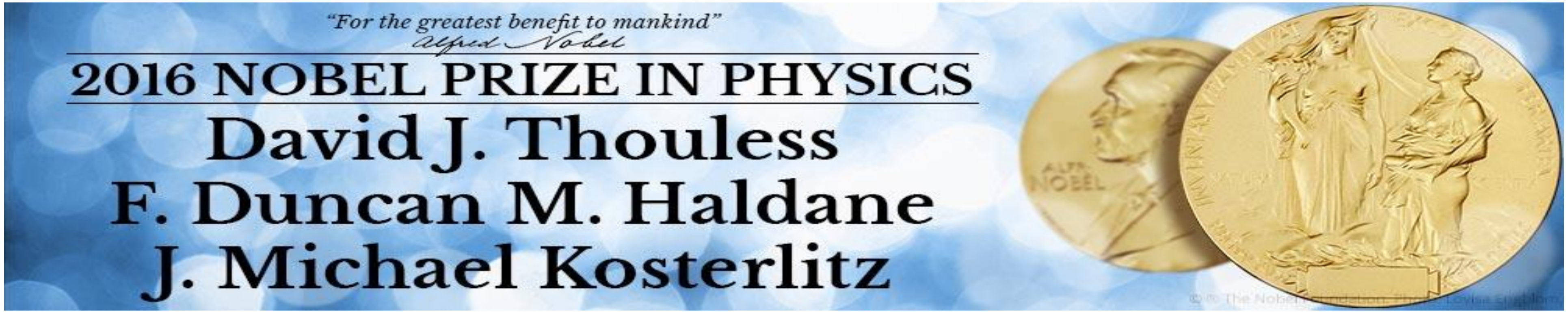
- Interstitial Mg fraction highest in *p*-GaN:Mg
- Lowest in *n*-GaN:Si

⇒ Direct evidence for amphoteric character of Mg that is coupled to the doping type

- Site change interstitial - substitutional Mg_{Ga}

⇒ Activation energy for migration of interstitial Mg: E_M » 1.3 - 2.0 eV

Phys. Rev. Lett. **118**, 095501(2017)



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David J. Thouless

Prize share: 1/2

Photo: Princeton University, Comms. Office, D. Applewhite

F. Duncan M. Haldane

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Ill: N. Elmehed. © Nobel Media 2016

J. Michael Kosterlitz

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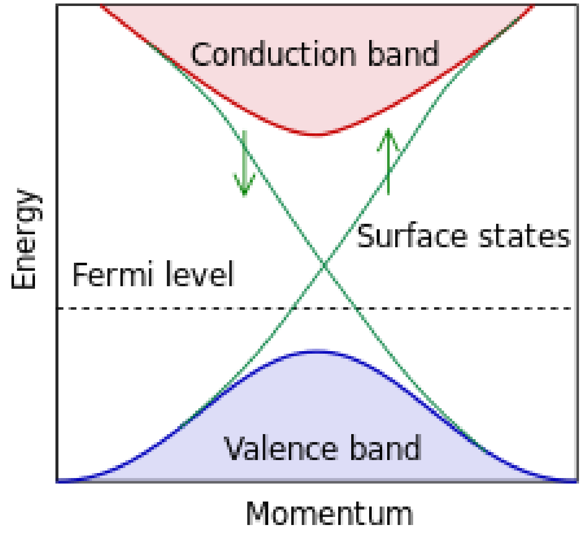
"for theoretical discoveries of topological phase transitions and topological phases of matter"

Source: "The Nobel Prize in Physics 2016". *Nobelprize.org.* Nobel Media AB 2014. Web. 4 Dec 2016.
<http://www.nobelprize.org/nobel_prizes/physics/laureates/2016/>

Topological insulators

Semi-metalic surface states originating from non-trivial topology of the electronic band structure in the bulk (insulator)



Dirac fermions at the surface (equivalent to graphene)
+ spin-locking → spin current in a *non-magnetic* material

(spintronics, quantum computation...)

Z_2 topological insulators: time reversal symmetry

Topological crystalline insulators: crystal mirror symmetry

Topological crystalline insulators

Rhombohedral distortion: breaking crystal mirror symmetry

PbTe

↓

(Pb,Sn)Te

↓

SnTe

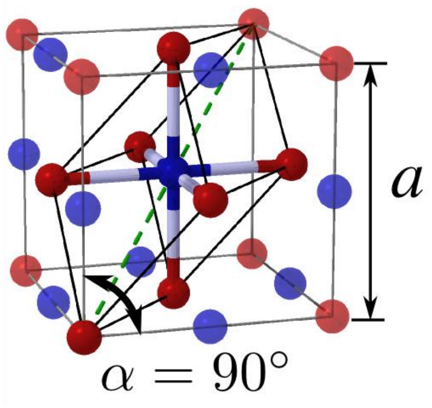
↓

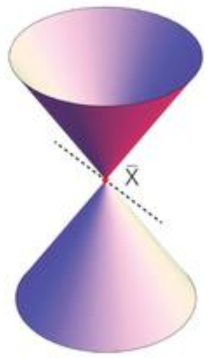
(Ge,Sn)Te

↓

GeTe

cubic



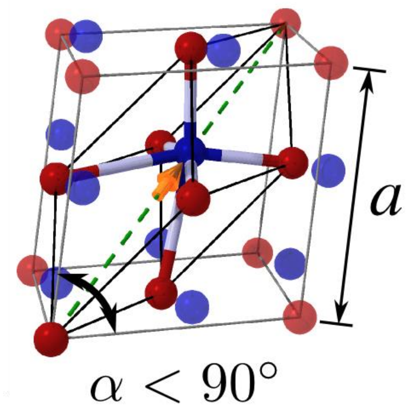


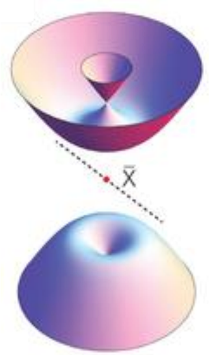
topological crystalline insulator (TCI)

Pb, Sn or Ge ●

Te ●

rhombohedral

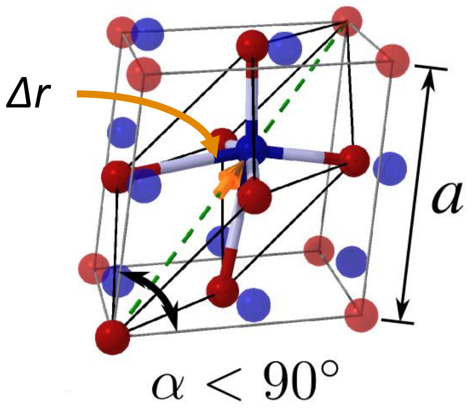


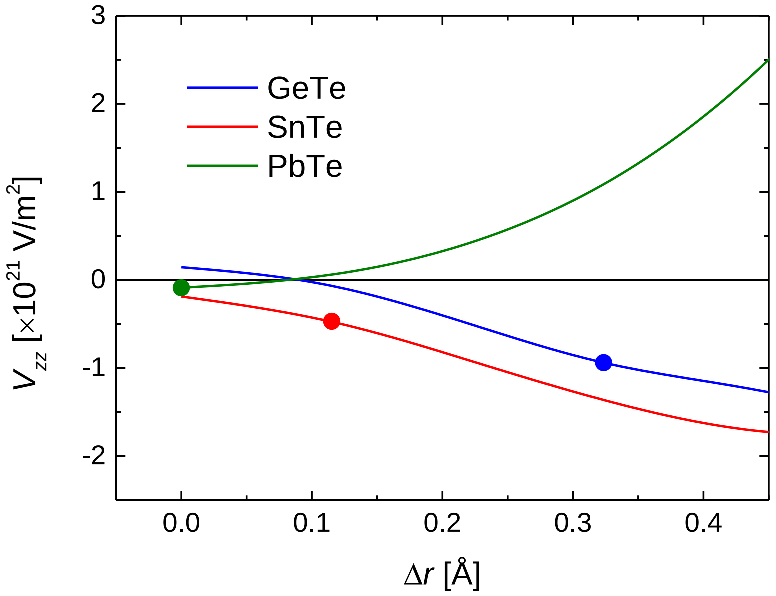


ferroelectric Rashba semiconductor (FERS)

Perturbed angular correlation

... with hyperfine interactions





$$\omega_Q \text{ or } \Delta E_Q \propto Q\, V_{zz}$$

	technique	parent	$t_{1/2}$	Q [b]
Pb	PAC	^{204m}Pb	67 min	0.44
Sn	eMS	^{119}In ^{119m}Sn	2.4 min 293 d	0.094
Ge	PAC	^{73}As	80 d	0.70

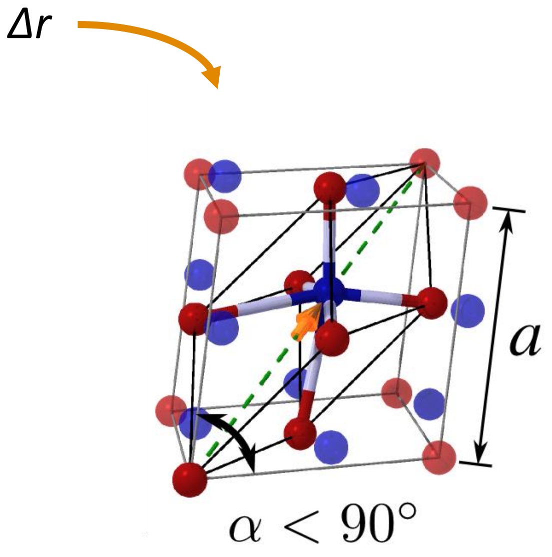
density functional theory calculations

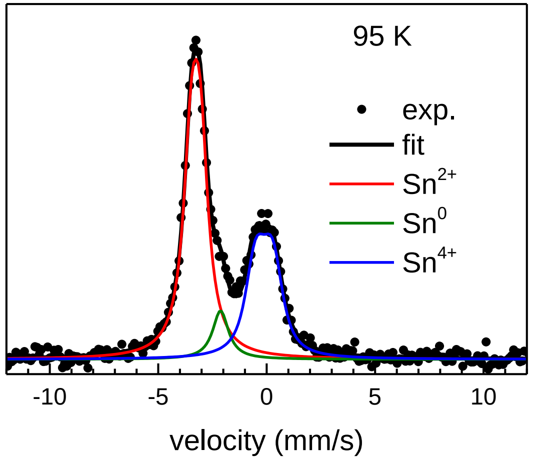
to establish relation between measure HFI parameters and structural parameters

Mossbauer spectroscopy

... with hyperfine interactions

$^{119}In \rightarrow {}^{119}Sn$ emission Mössbauer (PbTe)





Proof-of-principle with most challenging case:

> PbTe is cubic

> smallest *Δr* (< 0.10 Å)

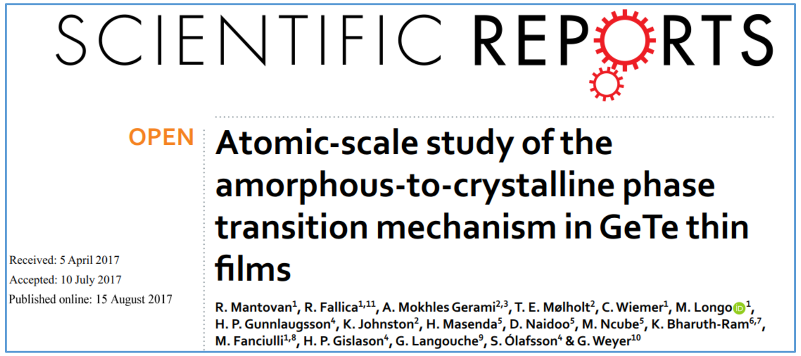
> ^{119}Sn – smallest *Q*

> <10 % precision in *Δr* (corresponding to < 0.05 Å)

> non-regular sites: OK

> damage: OK

See also:



SCIENTIFIC REPORTS

OPEN

Atomic-scale study of the amorphous-to-crystalline phase transition mechanism in GeTe thin films

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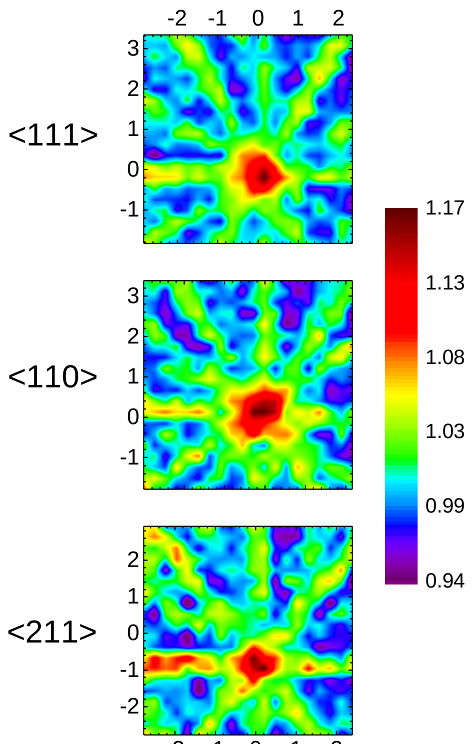
R. Mantovan[1], R. Fallica[1,11], A. Mokhles Gerami[2,3], T. E. Mølholt[2], C. Wiemer[1], M. Longo[1], H. P. Gunnlaugsson[4], K. Johnston[2], H. Masenda[5], D. Naidoo[5], M. Ncube[5], K. Bharuth-Ram[6,7], M. Fanciulli[1,8], H. P. Gislason[4], G. Langouche[9], S. Ólafsson[4] & G. Weyer[10]

…with emission channeling

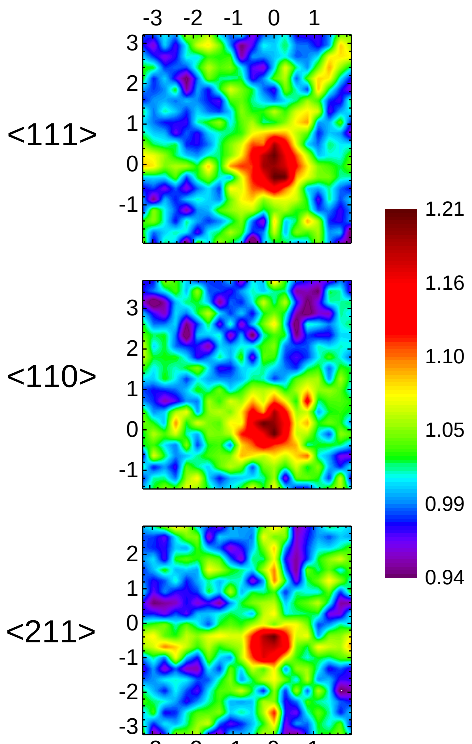
Successful emission channeling measurements on topological insulators



Mn-doped PbTe

^{56}Mn (2.6 h)

- Mn as magnetic dopant
- EC is used to determine the lattice location



Sn-doped PbTe

^{123}Sn (40 min)

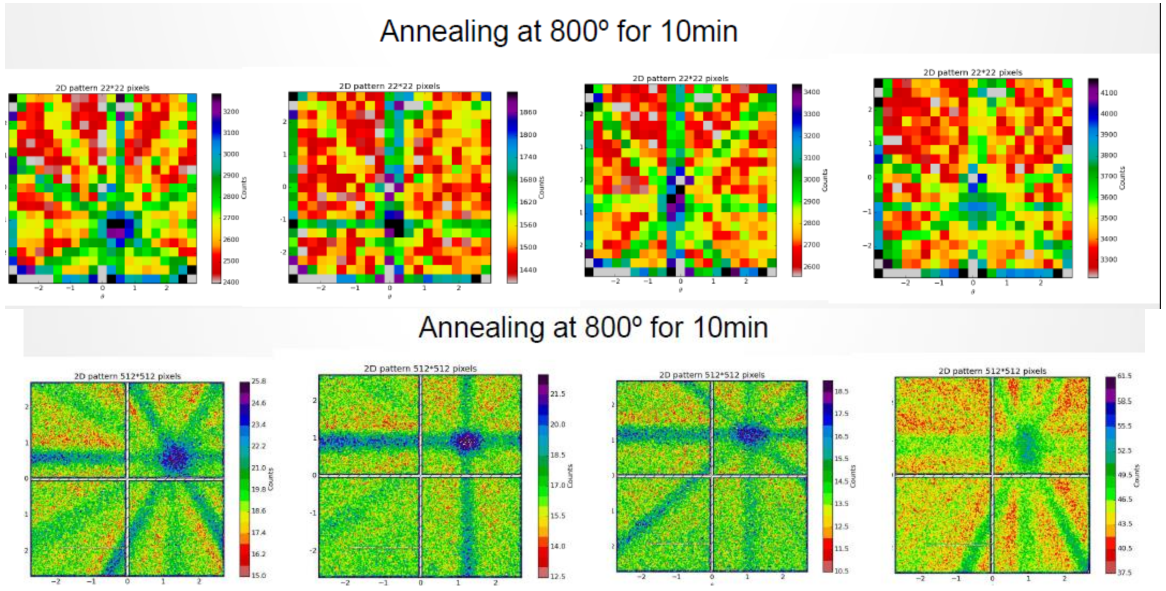
- parent: ^{123}In
- *isotope used for first time for EC*
- EC is used to characterize the distortion

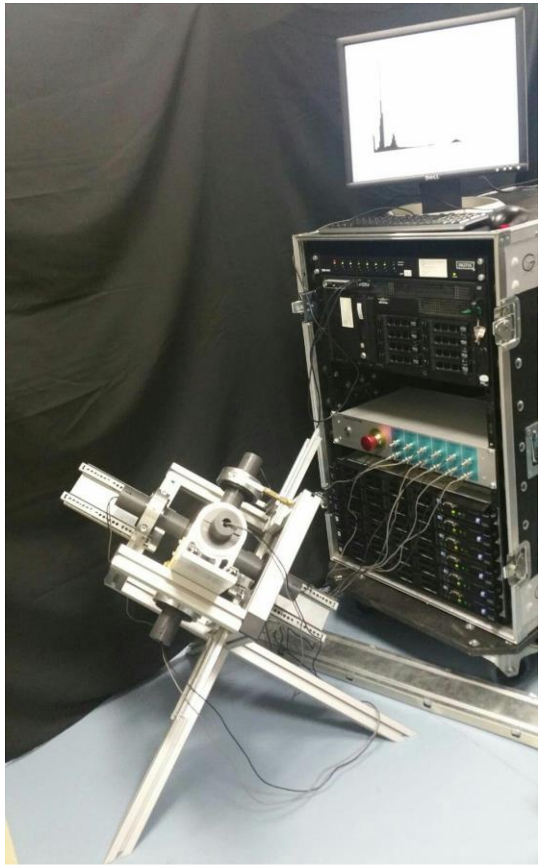
Combination with multitude of techniques, at ISOLDE and beyond, and strong link to theory

Challenges

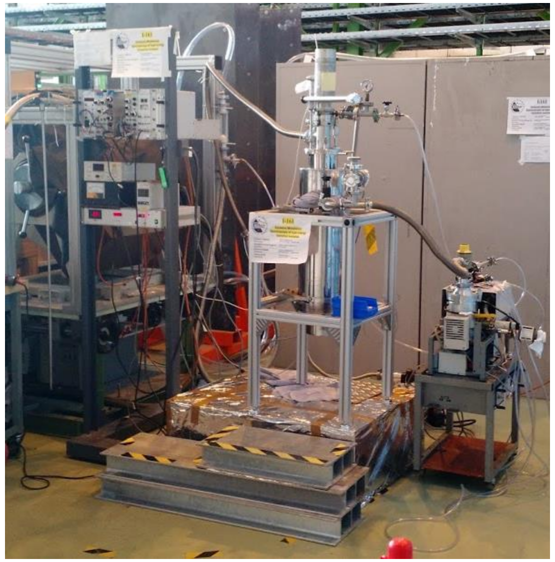
- Still a niche field, unfamiliar beyond the core community, or necessarily small: emission channelling needs to be attached to an ISOLDE …
- Limitations with beamtime → shift requests can be comparatively modest compared to nuclear physics. → only one run a year….(Mossbauer work previously mentioned took about 2.5 years to complete).
- Core community e.g. in Germany not being renewed, nuclear solid state physics not popular at home labs: licenses etc… Leads to problems with training.
- Pace of materials science is very fast… difficult to react to new developments/materials….experiments which don't get beam within one year can lose relevance….
- Safety administration becoming ever heavier

New developments

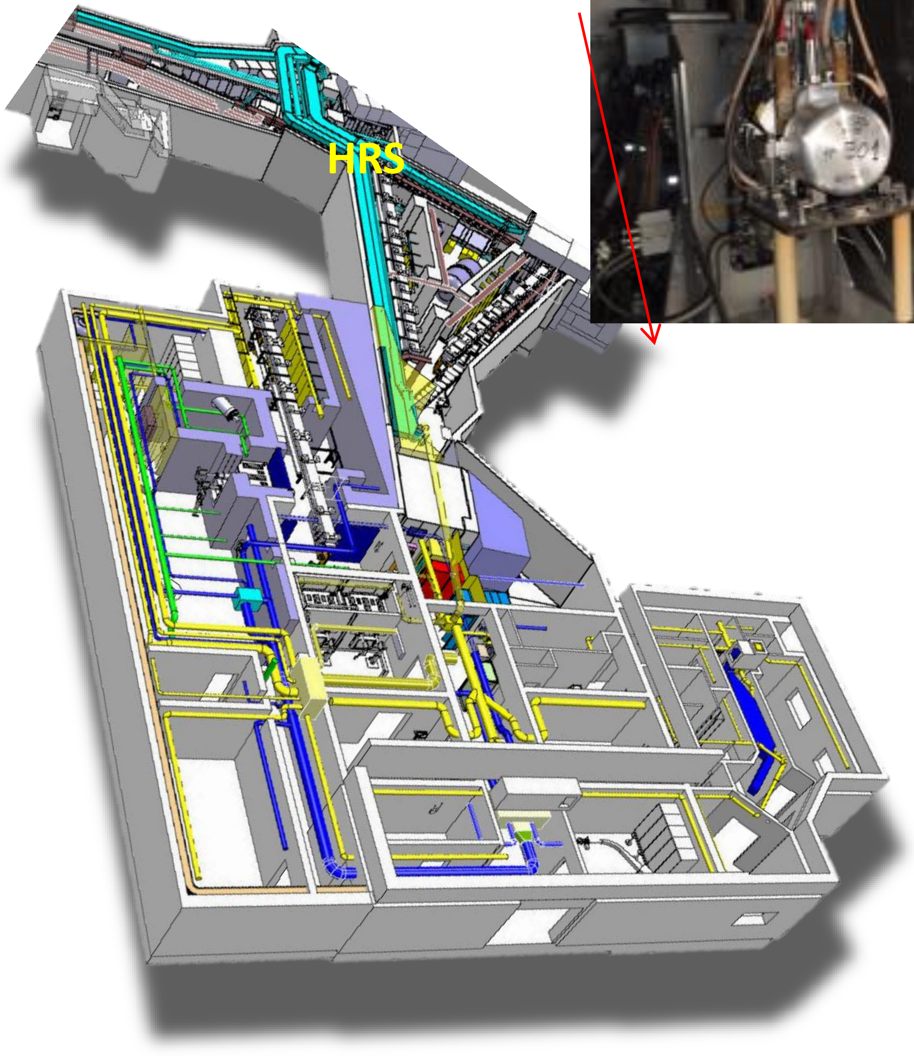


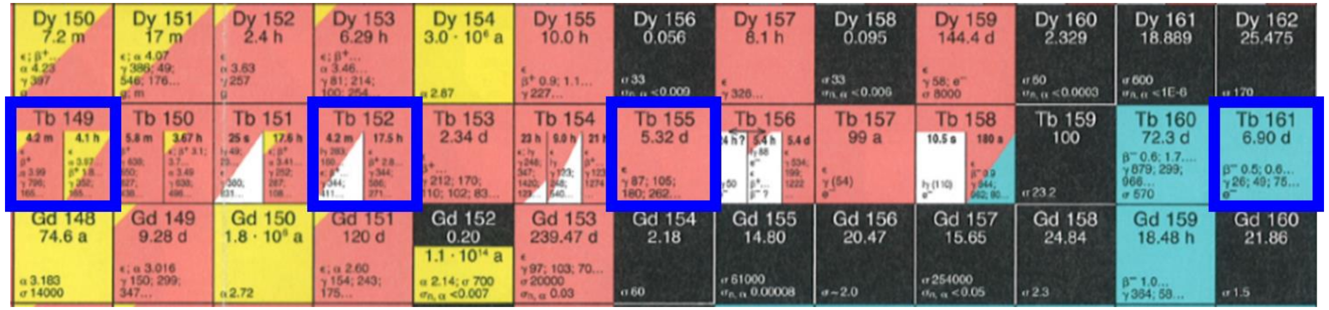


- Upgrading of detectors/spectrometers/ upgrade DAQ.
- LaBr and CeBr detectors, allow for wider range of probes to be used.
- Spectrometer for biophysics for Mossbauer spectroscopy.



CERN-MEDICIS: new Facility for medical isotopes (and solid state/materials?)





Summary

- Solid State programme at ISOLDE: well established and a unique combination of techniques on site.

- Varied experimental programme capable of resolving otherwise difficult problems. Remaining relevant in spite of the difficulties in obtaining beam time..

- New developments should help make spectrometers more user friendly

- Possible link with MEDICIS could allow for wider community to be served (beyond ISOLDE)

- OPEN ACCESS
  The solid state physics programme at ISOLDE: recent developments and perspectives
  Karl Johnston *et al* 2017 *J. Phys. G: Nucl. Part. Phys.* **44** 104001
  + View abstract   View article   PDF

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