

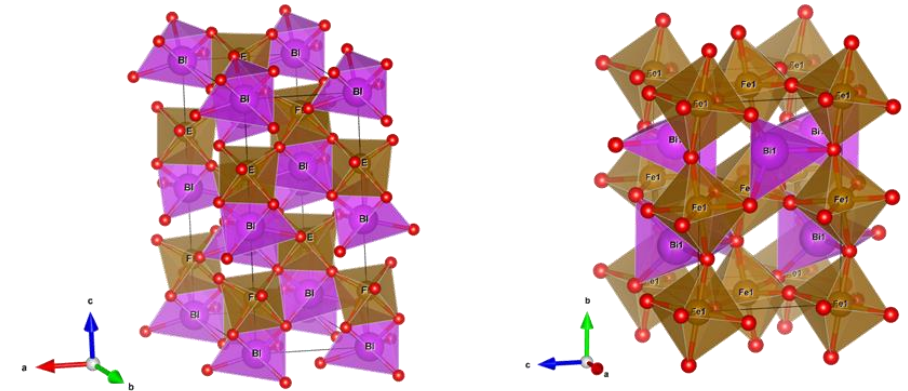
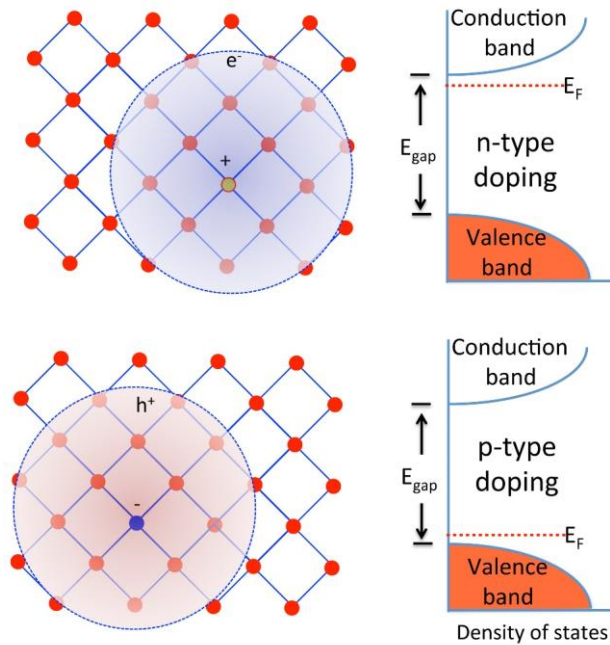
Spicing up solid state physics with radioactive isotopes: recent highlights from ISOLDE
Karl Johnston, CERN



- Introduction to some aspects of solid state physics
- Why use radioactive isotopes?
- Techniques
- Examples of recent results

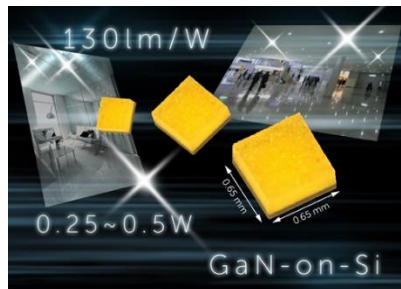
Thanks to Juliana Schell, Guilherme Correia, Georg Marschick
and all groups in SSP@ISOLDE

Some aspects of solid state physics



Multiferroics e.g. BiFeO_3

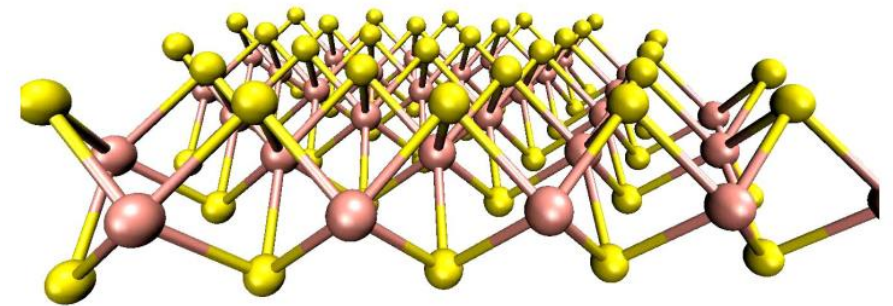
What form of material produces a particular effect?



Doping in semiconductors

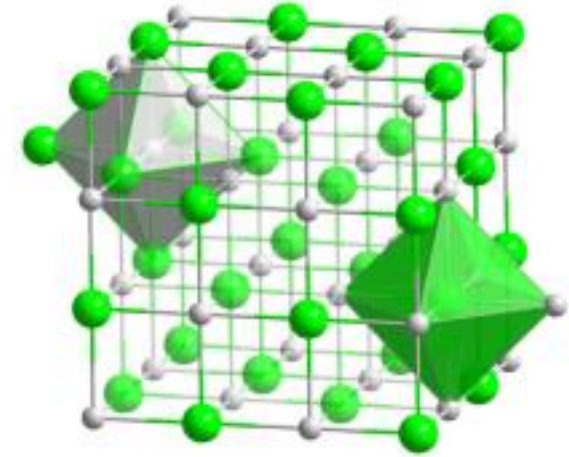
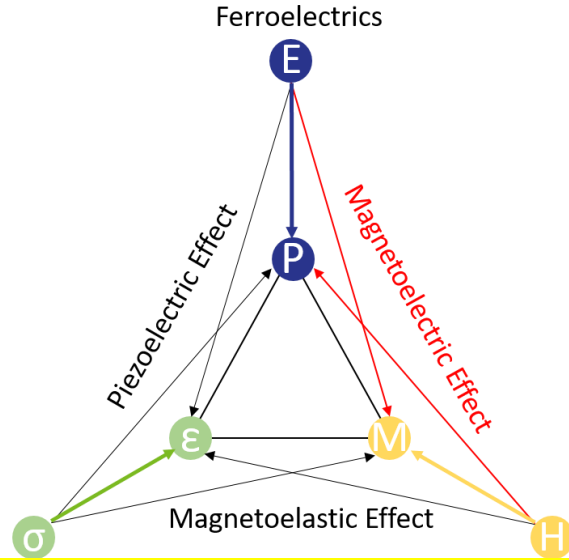
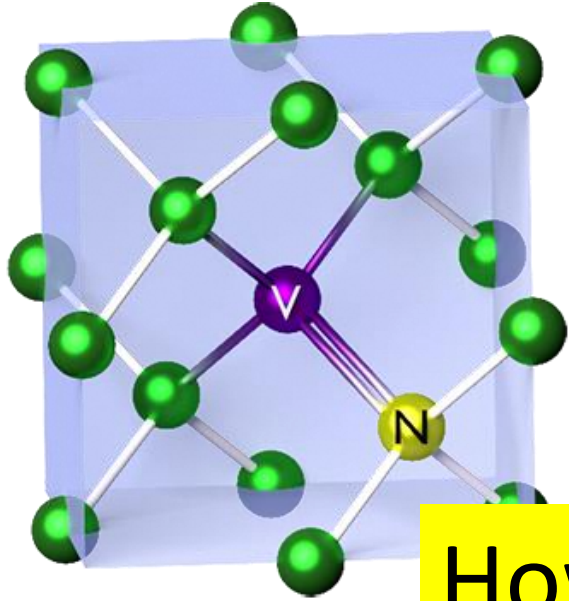
What causes doping (element, or defect)?

Where does it sit in the lattice?



Surface studies of low dimensional materials...

Current “hot” topics

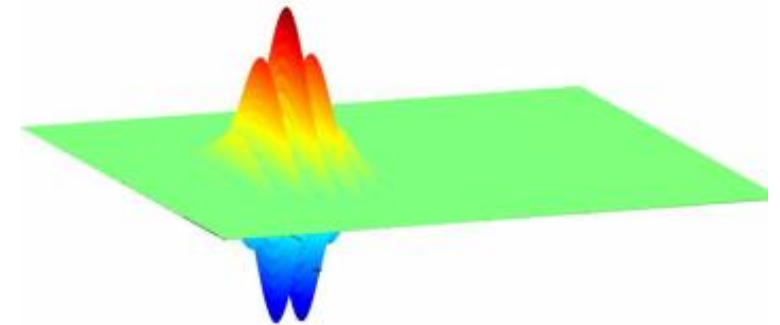
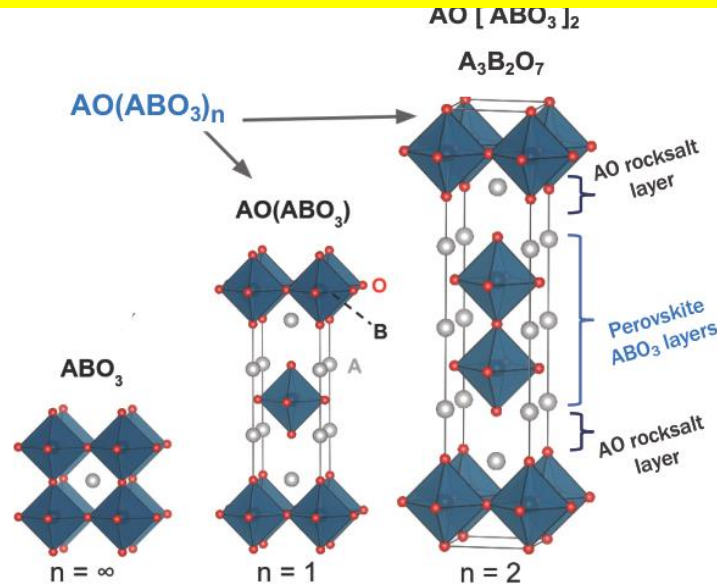


How can radioactive ion beams help?

K

Defects in diamond

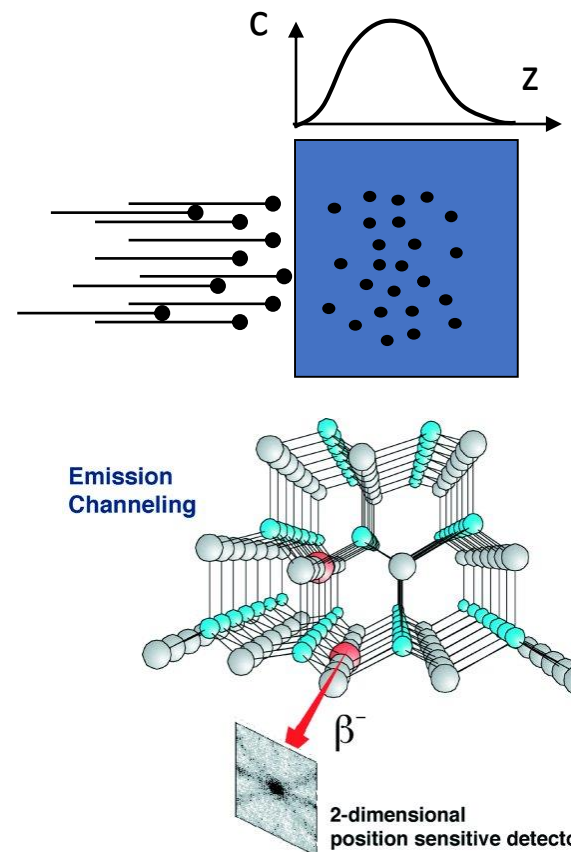
Quantum bits:
control and application



Spin or superconducting fluctuations

Unique features of radioactive probe atoms for SSP applications


- **Chemically selective and isotope specific**
- **Extremely good detection limit**
 - among the most sensitive methods, no reaction cross section limitation
 - $10^{15} - 10^{18}$ probes/cm³
 - $10^{11} - 10^{12}$ probe atoms
- **Depth distribution and concentration control**
 - Ion energy and ion fluence control
 - Circumventing solubility and diffusion limits
- **Highly local Information**
 - Nucleus-size sensors for **local** magnetic and electric fields
 - Electric Field Gradient $\sim r^{-3}$
 - Emission channeling: ~ 0.02 nm position resolution

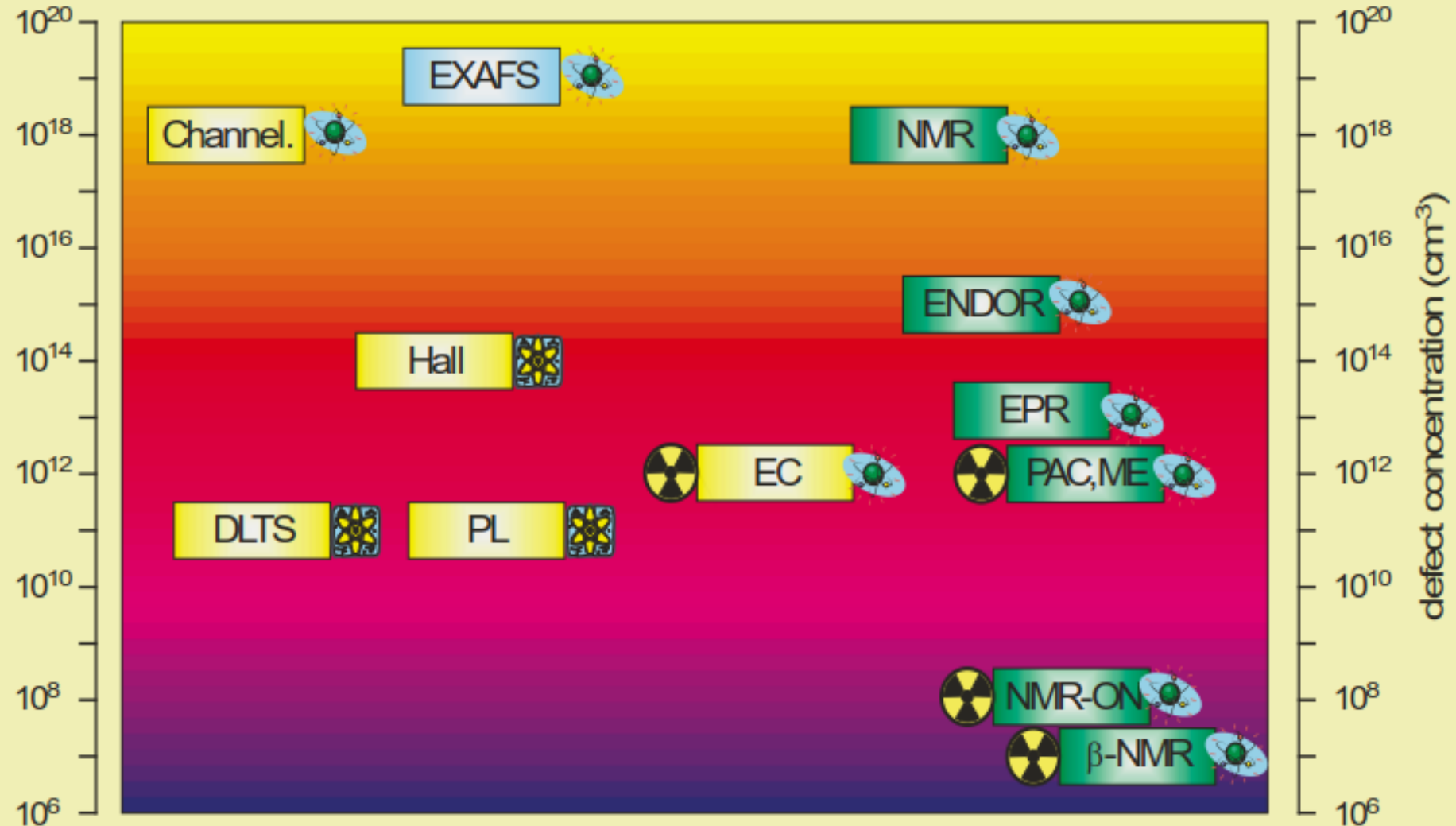


Why radioactive probes ? Sensitive – Selective - Controllable – Local
Often relatively easy isotopes for RIB facilities to produce (not always a good thing...)

Semiconductor Spectroscopy

sensitive to chemical nature  or electronic properties 

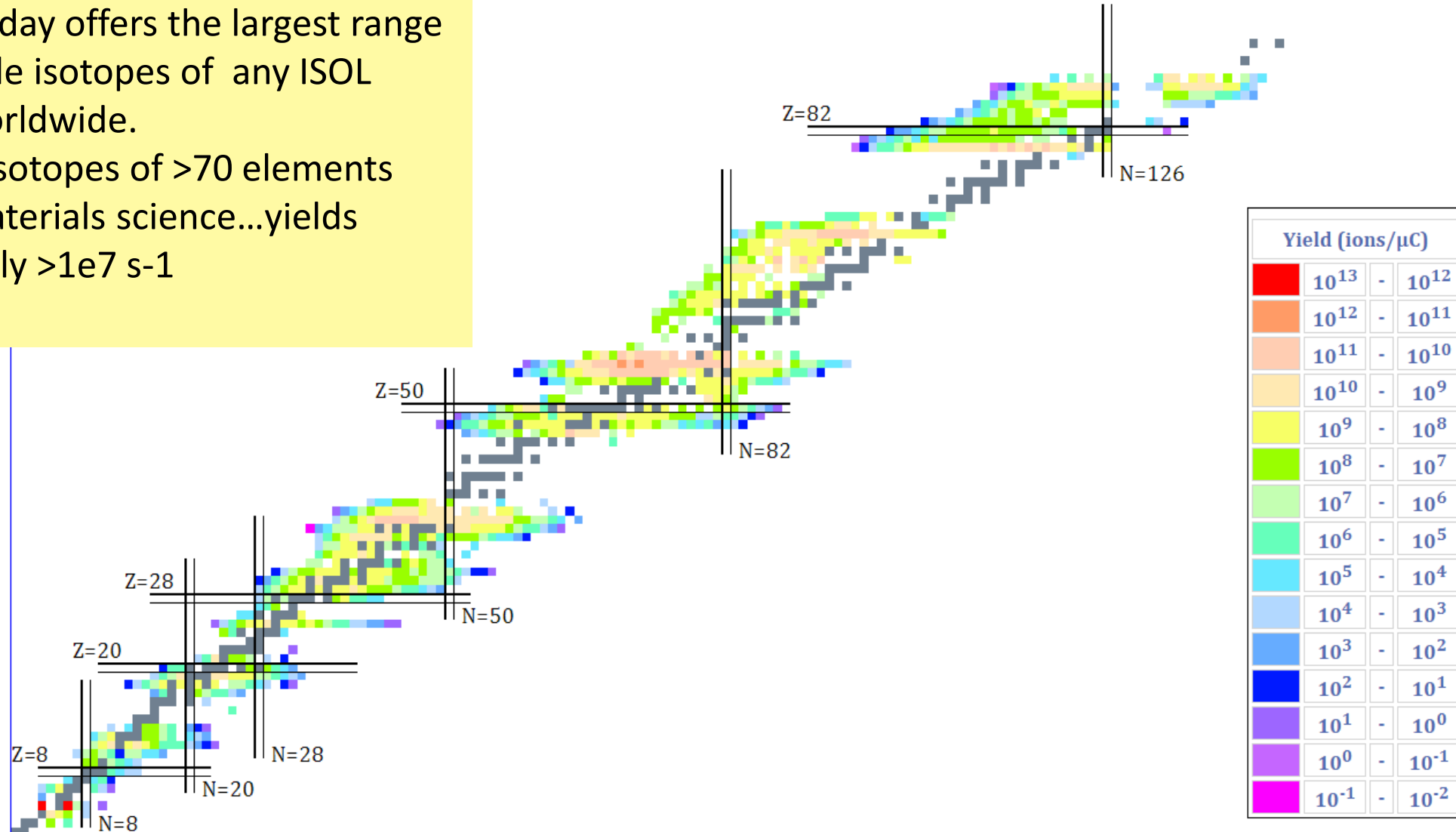
(some require radioactive isotopes )



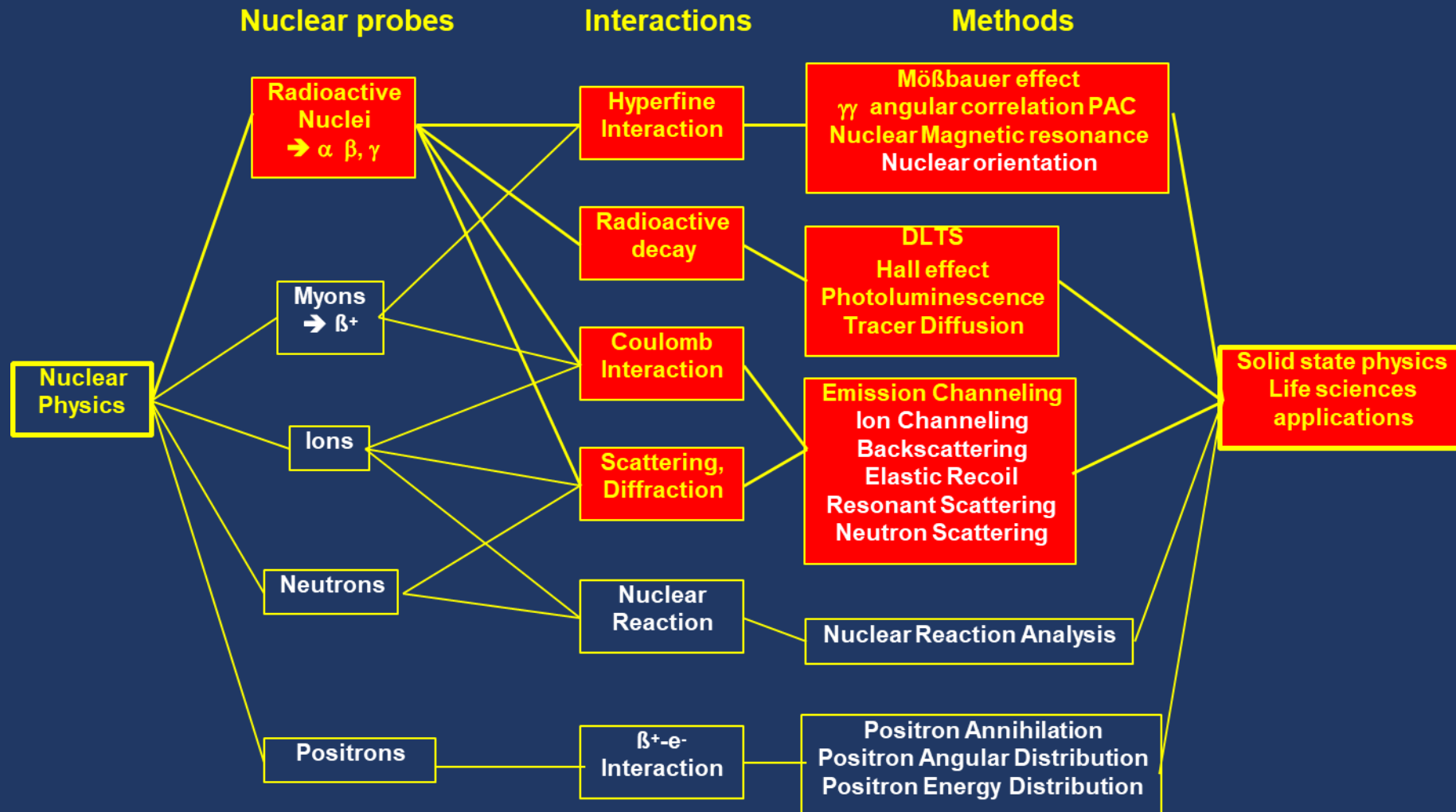
Nuclear chart for ISOLDE

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

- 1000 isotopes of >70 elements
- For materials science...yields typically $>10^7$ s⁻¹



Applying radioactivity to solid state physics/biophysics



ISOLDE table of elements

<div><div><div>+</div><div>surface ionization</div><div>-</div></div><div>laser selective ionization</div><div><div>hot</div><div>plasma discharge</div><div>cold</div></div></div>																			
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																	
			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			

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

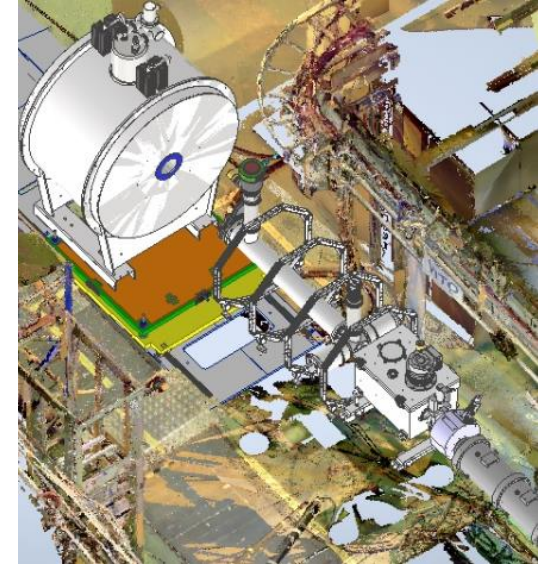
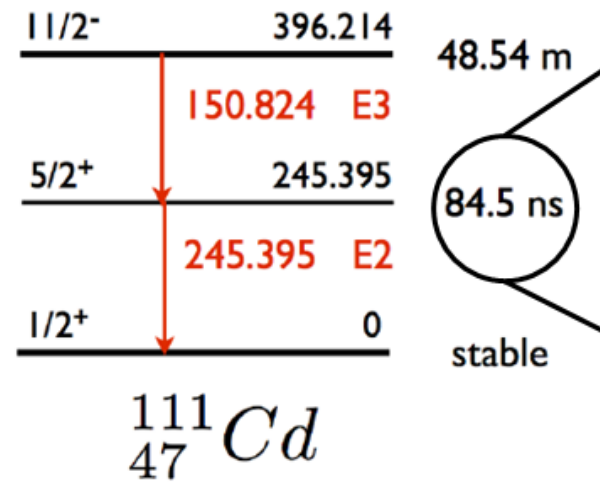
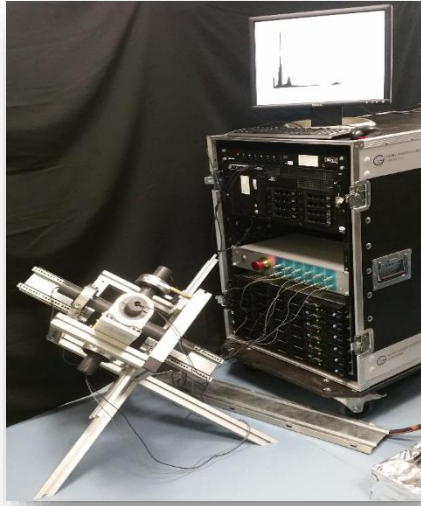
Solid state demands have been a driver in the development of new beams at ISOLDE

Overview of techniques for «applied» nuclear physics I: local probes/channelling

Perturbed angular correlation

Mossbauer effect

Beta-NMR

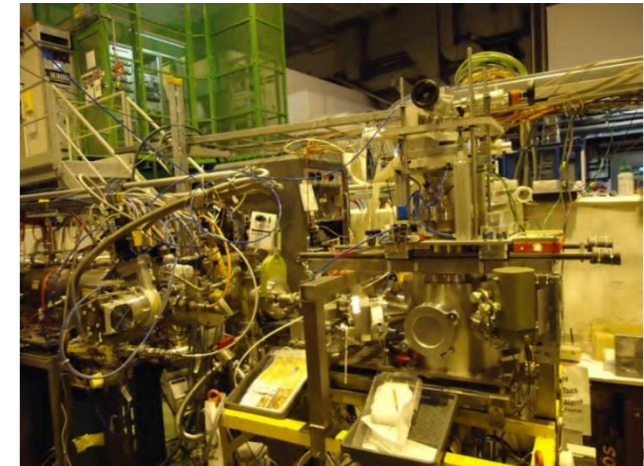
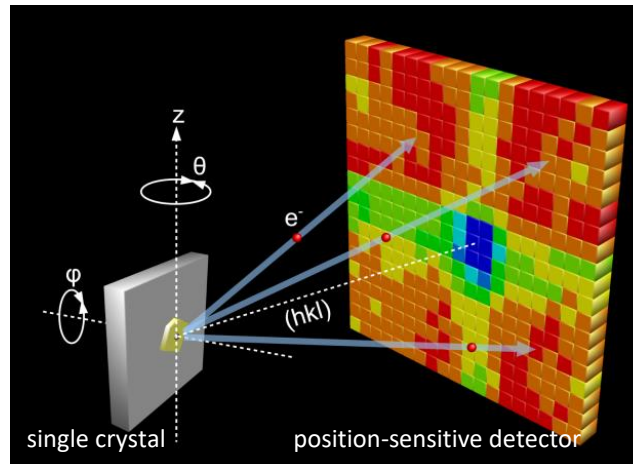


Very sensitive probes to the local environment of a material/protein etc, **magnetic interactions**, dynamical processes. Very intense (and often unique) beams available at ISOLDE

Emission channelling:

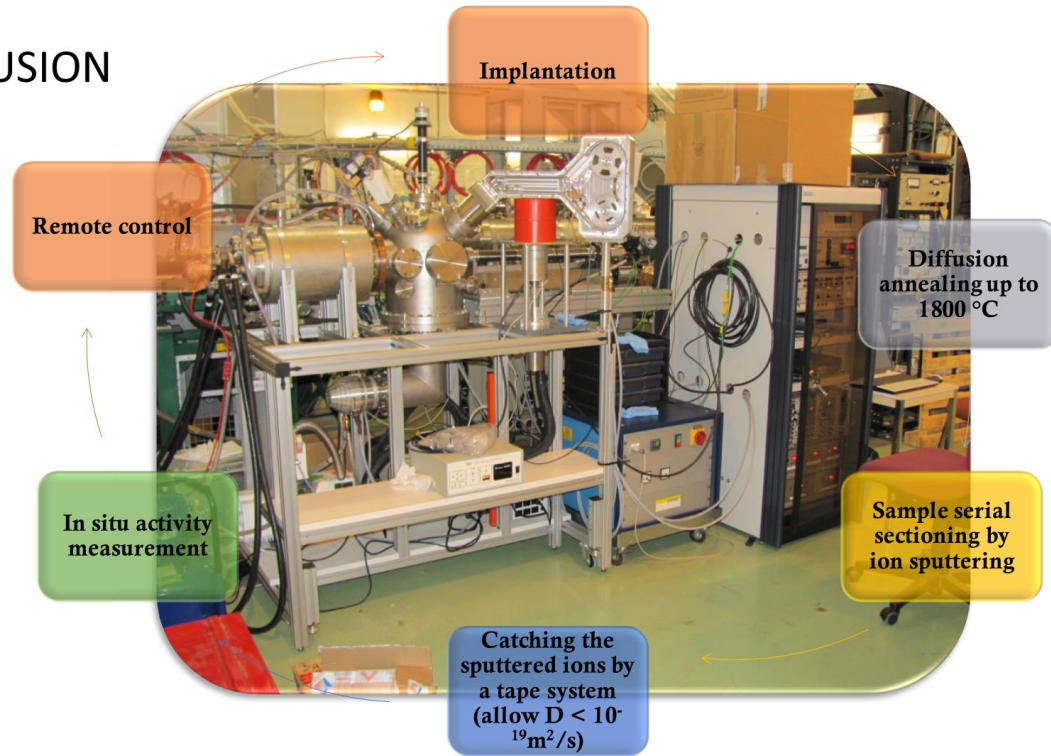
Almost unique to ISOLDE. 4 orders of magnitude more sensitive than RBS.

Ability to utilise low concentrations to determine position: increasingly important...



Overview of techniques for «applied» nuclear physics II: Tracer methods

DIFFUSION



Photoluminescence spectroscopy



Most general approach....tracking half-life of implanted isotopes or their daughters

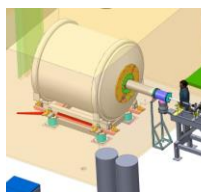
Can be applied to almost any «typical» laboratory technique: offers unique sensitivity



MINIBALL



SCATTERING
EXPERIMENTS



ISS



COLLAPS



ISOLTRAP



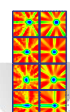
CRIS



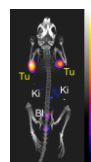
IDS



VITO



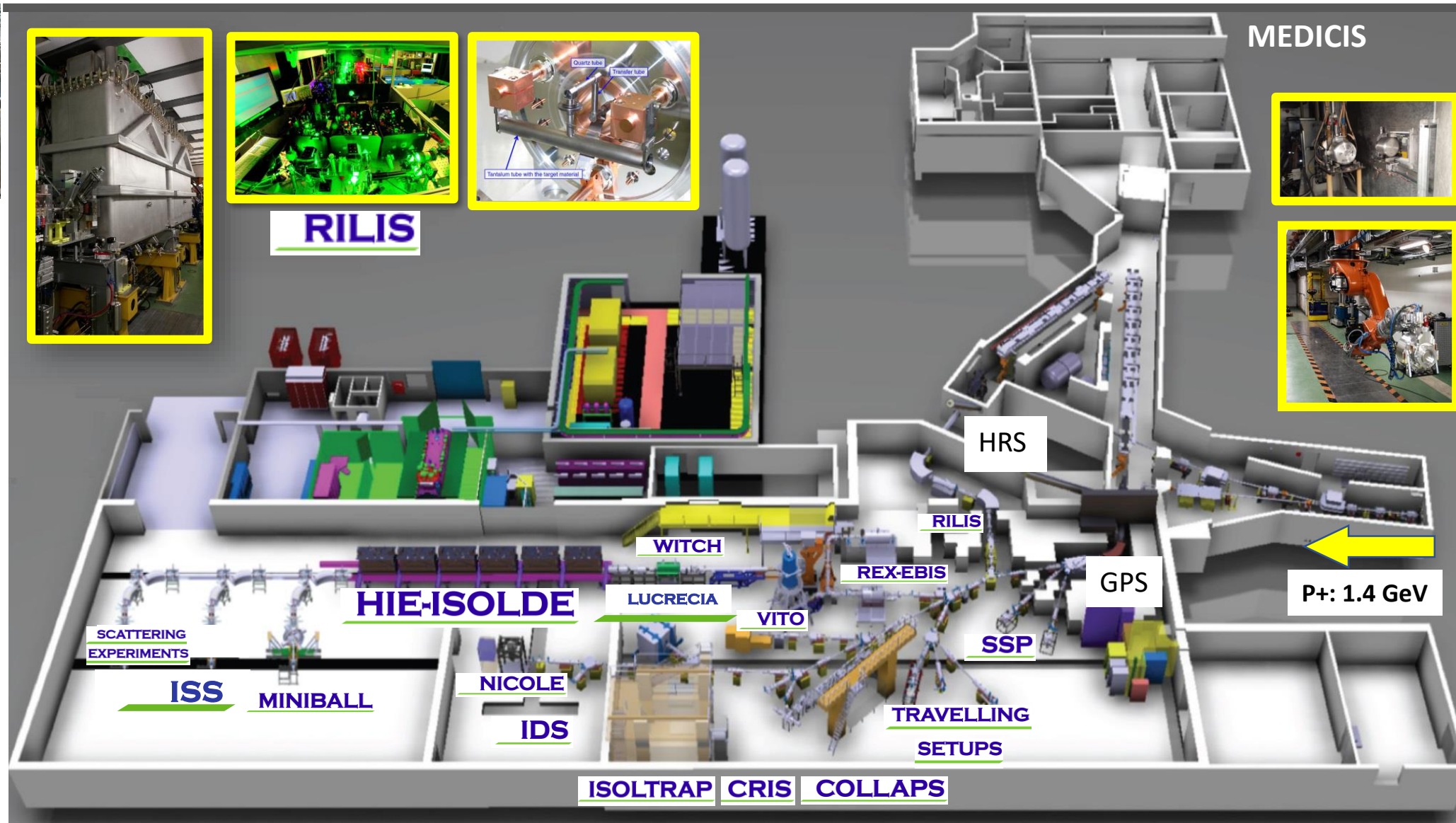
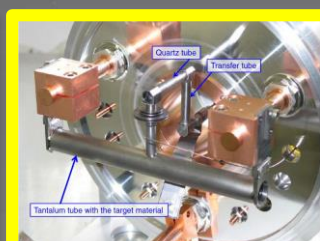
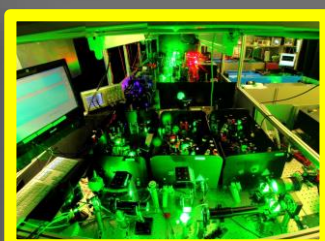
SSP



MEDICAL
ISOTOPES

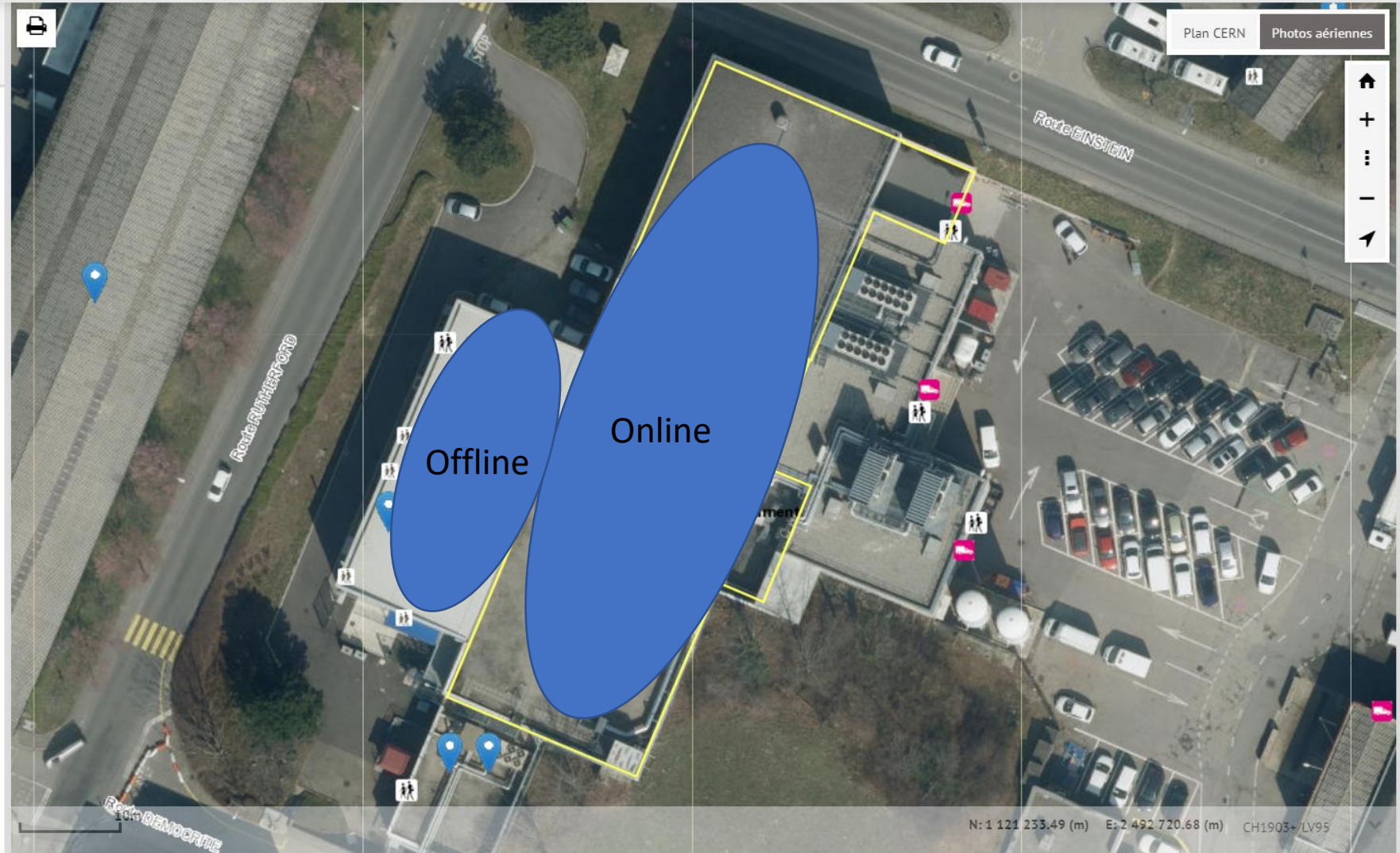


RILIS

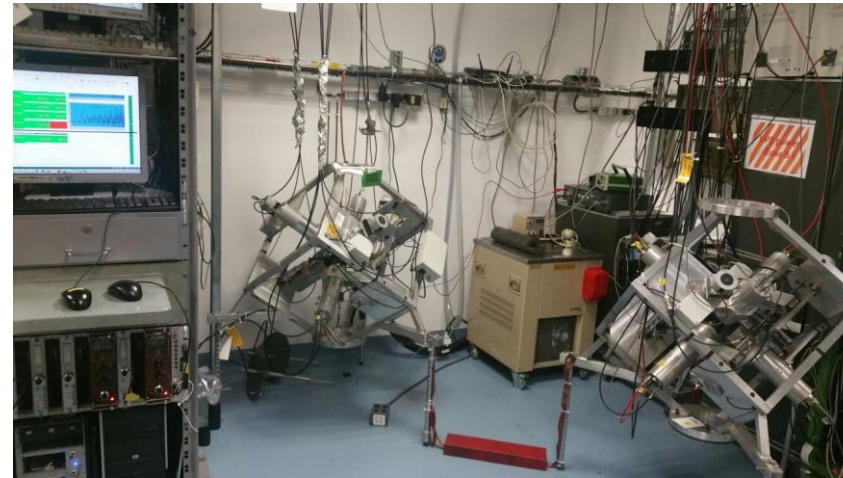


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Rechercher un bâtiment, un bureau, un site ou point d'intérêt

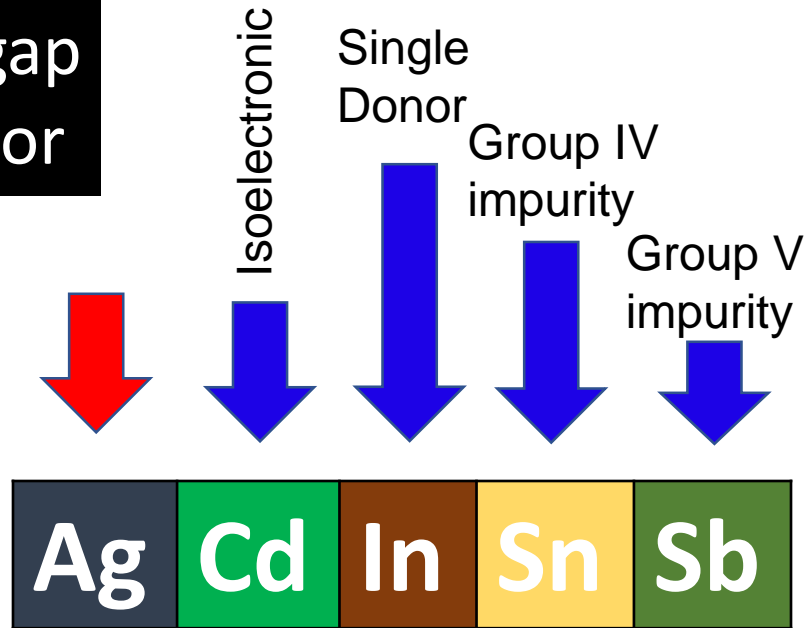
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Offline labs at ISOLDE: chemistry; spectroscopy; characterisation

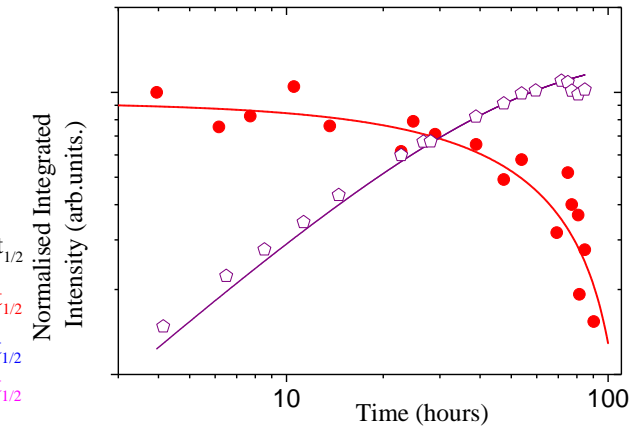
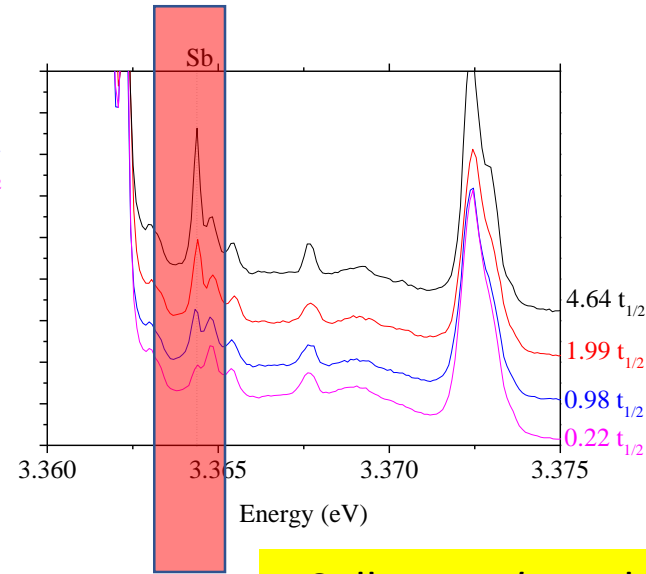
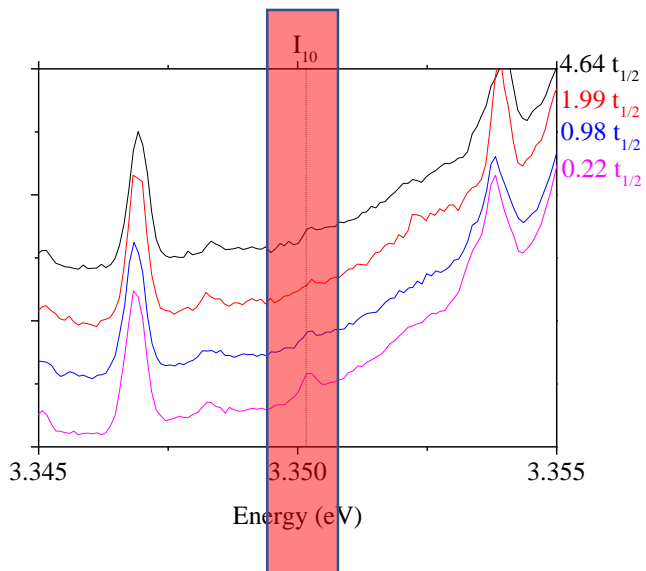


ZnO: wide
band gap
semiconductor

Radiotracer PL



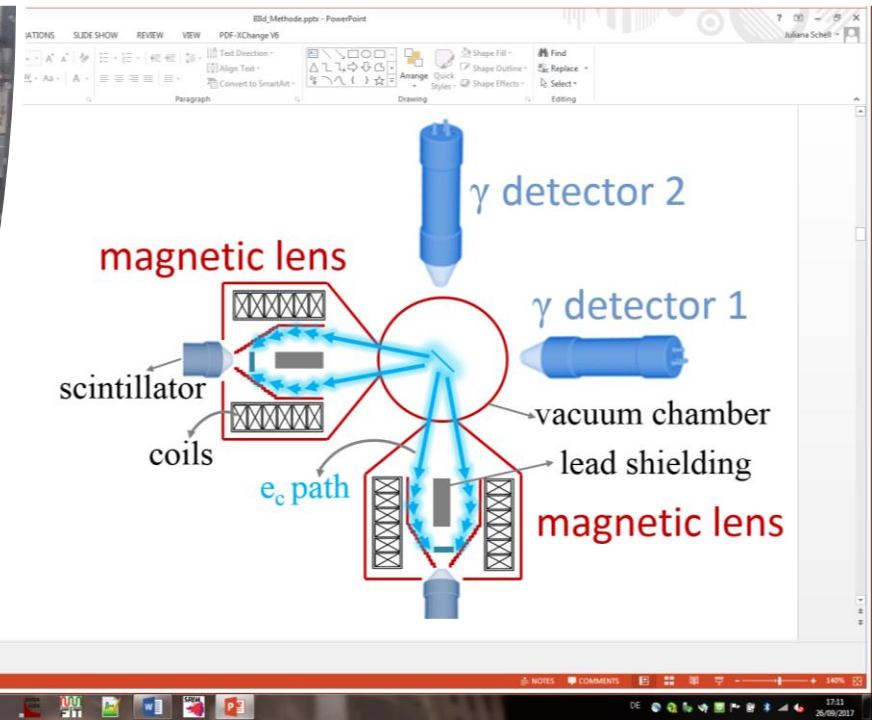
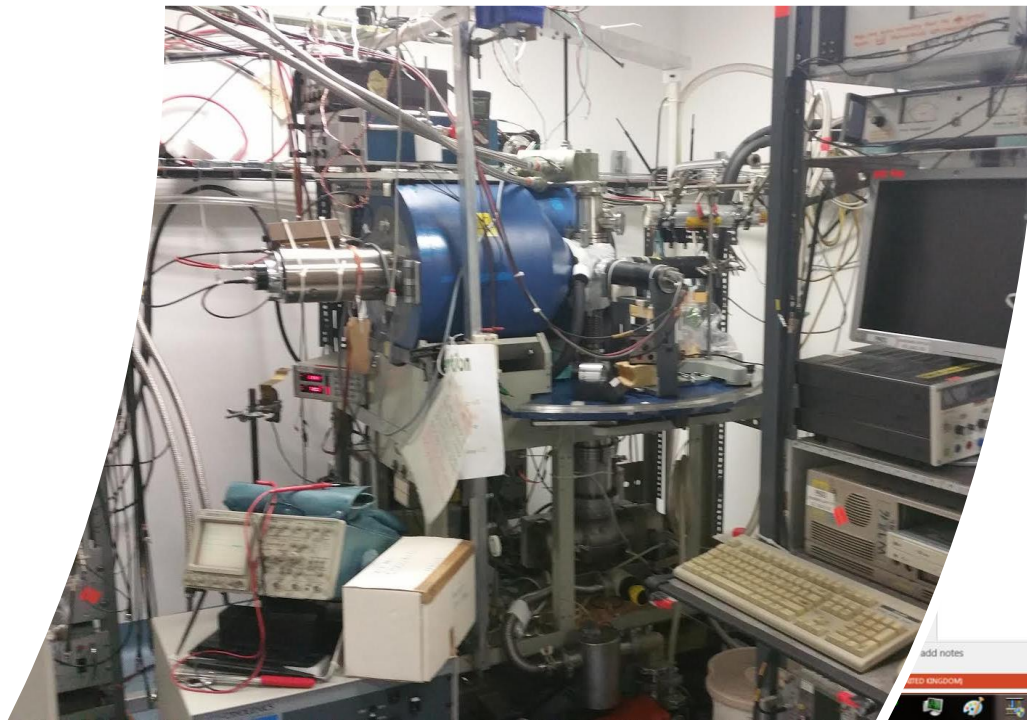
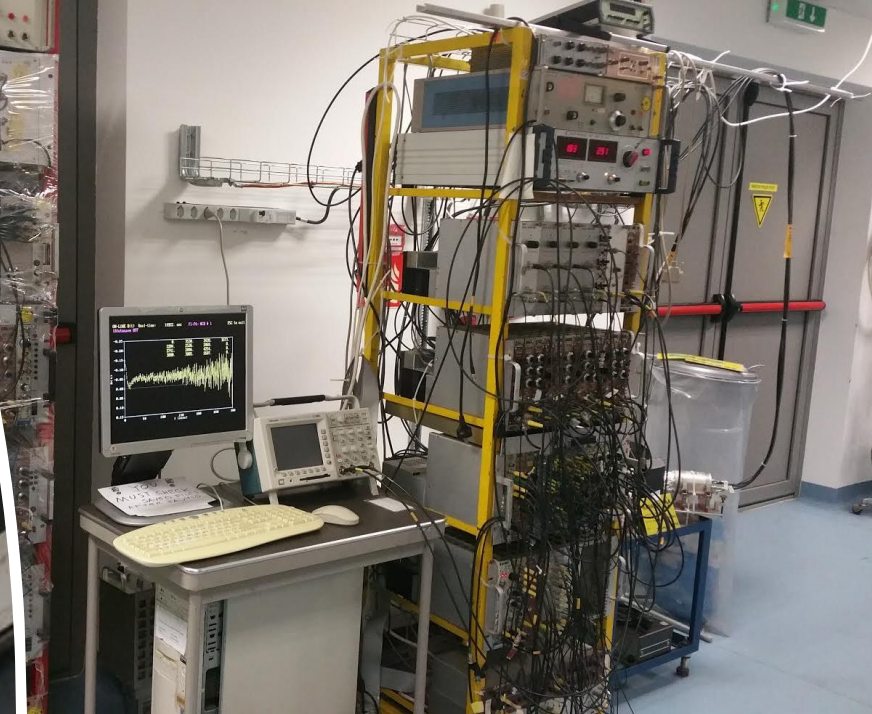
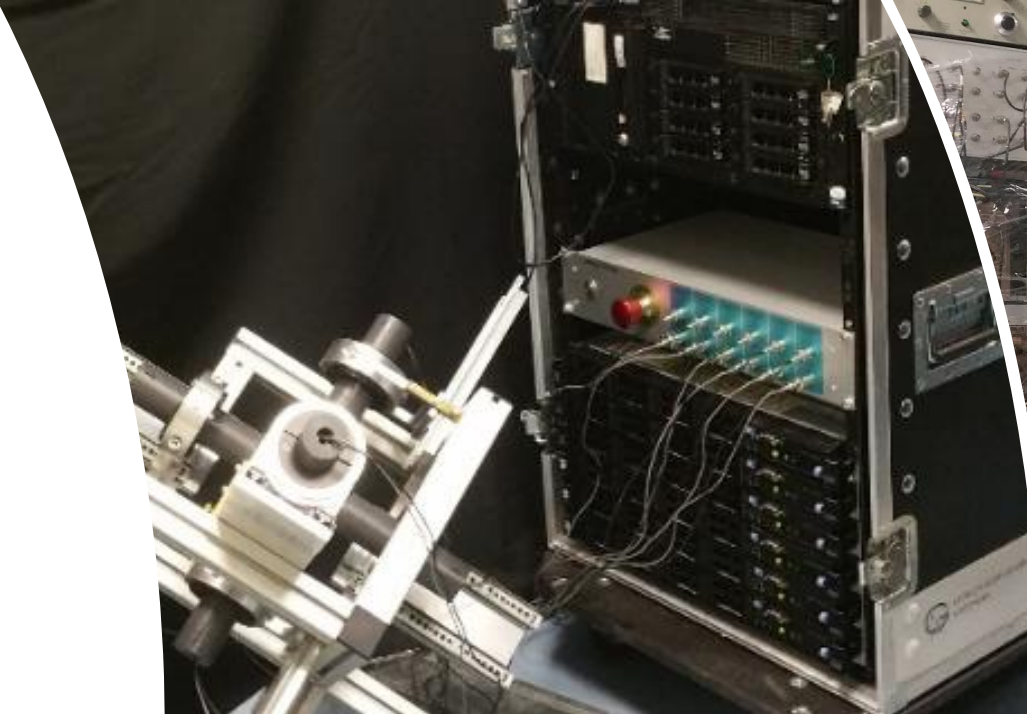
Sb121 5/2+ 57.36 0+ 32.59 β ⁻	Sb122 2.70 d 2- EC, β ⁻ β ⁻	Sb123 7/2+ 42.64 0+ 4.63 β ⁻	Sb124 60.20 d 3- β ⁻	Sb125 2.7582 y 7/2+ β ⁻
Sn120 0+ 32.59 β ⁻	Sn121 27.06 h 3/2+ β ⁻	Sn122 0+ 4.63 β ⁻	Sn123 129.2 d 11/2- β ⁻	Sn124 0+ 5.79 β ⁻
In119 2.4 m 9/2+ β ⁻	In120 3.08 s 1+ β ⁻	In121 23.1 s 9/2+ β ⁻	In122 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



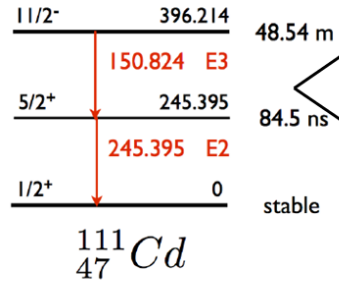
J Cullen *et al* Appl. Phys. Lett. **102** 192110 (2013)

Perturbed angular correlation @ ISOLDE

- Most established technique at ISOLDE: benefitting from upgrade of spectrometers in recent years and improved relation to theory.
- Electron gamma unique to ISOLDE.
- Range of novel isotopes also only useable at ISOLDE: allows for varied programme in materials physics, biophysics and beyond



Perturbed angular correlation @ ISOLDE



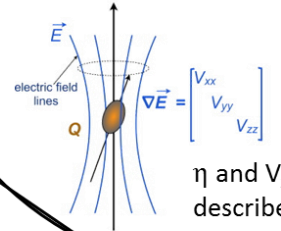
Electric quadrupole interaction

Electric Field Gradient (EFG)

$$\begin{pmatrix} V_{xx} & 0 & 0 \\ 0 & V_{yy} & 0 \\ 0 & 0 & V_{zz} \end{pmatrix}$$

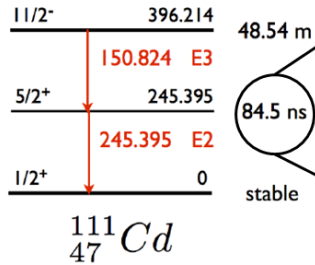
$$\omega \propto \Delta E \propto Q \otimes \text{EFG}$$

interacts with core quadrupole moment Q



$$\eta = \frac{V_{xx} - V_{yy}}{V_{zz}}$$

η and $V_{zz}(\omega_0)$ describe the EFG



Nuclear level splitting

Electric Field Gradient (EFG)

$$\begin{pmatrix} V_{xx} & 0 & 0 \\ 0 & V_{yy} & 0 \\ 0 & 0 & V_{zz} \end{pmatrix}$$

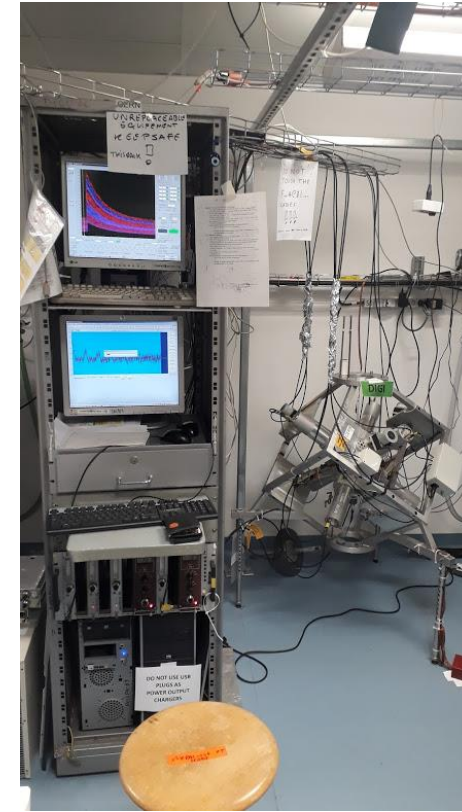
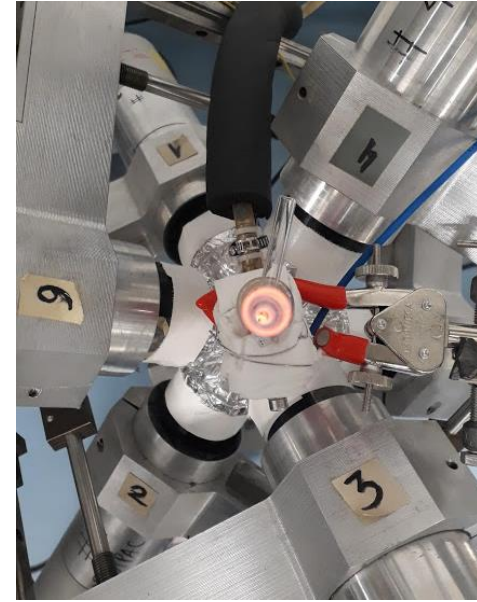
$$\omega \propto \Delta E \propto Q \otimes \text{EFG}$$

+ combined interactions

Magnetic Hyperfine field

$$(0,0,B_z)$$

$$\omega \propto \Delta E \propto -\mu \cdot B$$



PAC: not sensitive to chemical shift, but more flexible than Mossbauer for temperature and environment in which can be measured.

New setups to measure in magnetic field and for surface science on horizon

PAC → Perturbed Angular Correlations
M → Mössbauer Effect

PAC → Perturbed Angular Correlations
M → Mössbauer Effect

H																	He	
Li <i>b-N</i>	Be											B	C	N	O	F <i>PAC</i>	Ne	
Na <i>b-N</i>	Mg <i>b-N</i>											Al	Si	P	S	Cl	Ar	
K <i>M</i>	Ca	Sr	Ti	V <i>PAC</i>	Cr	Mn	Fe <i>M</i>	Co	Ni <i>PAC</i>	Cu <i>PAC</i>	Zn <i>M</i>	Ga	Ge <i>PAC</i>	As <i>PAC</i>	Se <i>PAC</i>	Br <i>PAC</i>	Kr <i>PAC</i>	
Rb	Sr	Y	Zr	Nb	Mo <i>PAC</i>	Tc <i>PAC</i>	Ru <i>M</i>	Rh <i>PAC</i>	Pd	Ag	Cd <i>PAC</i>	In <i>PAC</i>	Sn <i>PAC</i>	Sb <i>M</i>	Te <i>M</i>	I <i>M</i>	Xe <i>M</i>	
Cs <i>PAC</i>	Ba <i>M</i>	La <i>M</i>	Hf <i>M</i>	Ta <i>PAC</i>	W <i>M</i>	Re <i>M</i>	Os <i>M</i>	Ir <i>PAC</i>	Pt <i>M</i>	Au <i>M</i>	Hg <i>PAC</i>	Tl	Pb <i>PAC</i>	Bi	Po	At	Rn	
Fr	Ra	Ac																
			Ce	Pr <i>PAC</i>	Nd <i>M</i>	Pm <i>M</i>	Sm <i>M</i>	Eu <i>PAC</i>	Gd <i>M</i>	Tb <i>M</i>	Dy <i>M</i>	Ho <i>M</i>	Er <i>M</i>	Tm <i>M</i>	Yb <i>PAC</i>	Lu <i>M</i>		
			Th <i>M</i>	Pa <i>M</i>	U <i>M</i>	Np <i>M</i>	Pu <i>M</i>	Am <i>M</i>	Cm <i>M</i>	Bk	Cf	Es	Fm	Md	No	Lr		



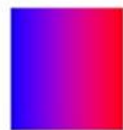
γ - γ & γ - e^- PAC



only e^- - γ PAC



only γ - γ PAC



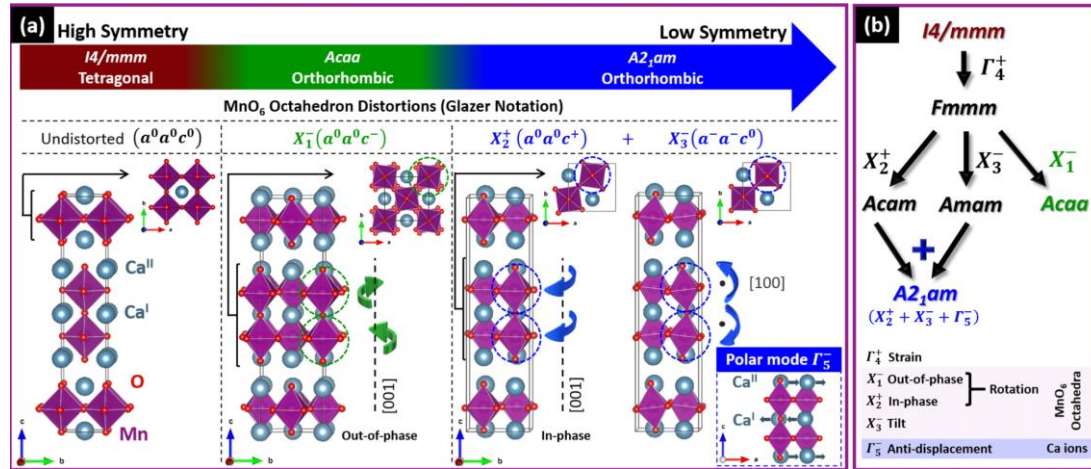
γ - γ & e^- - γ PAC



only γ - e^- PAC

PAC: hyperfine studies of multiferroics

$\text{Ca}_3\text{Mn}_2\text{O}_7$ structural path unraveled by atomic-scale properties: A combined experimental and *ab initio* study



Probing multiferroic materials with PAC on the atomic scale: revealing the atomic changes behind the transitions from polar to ferroelectric behaviour

Persistence of polar – phase clusters up to 500C

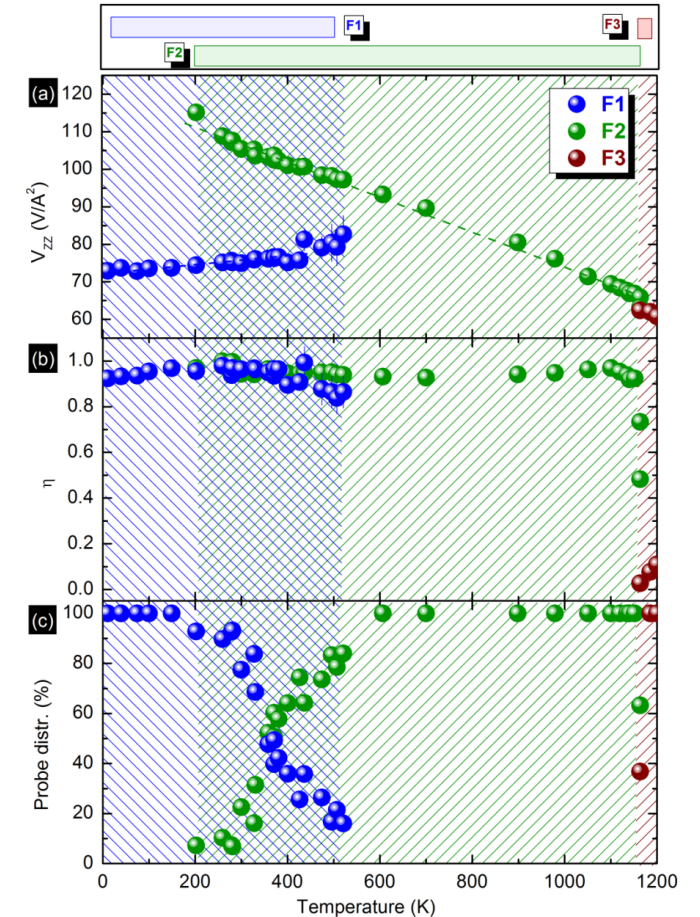
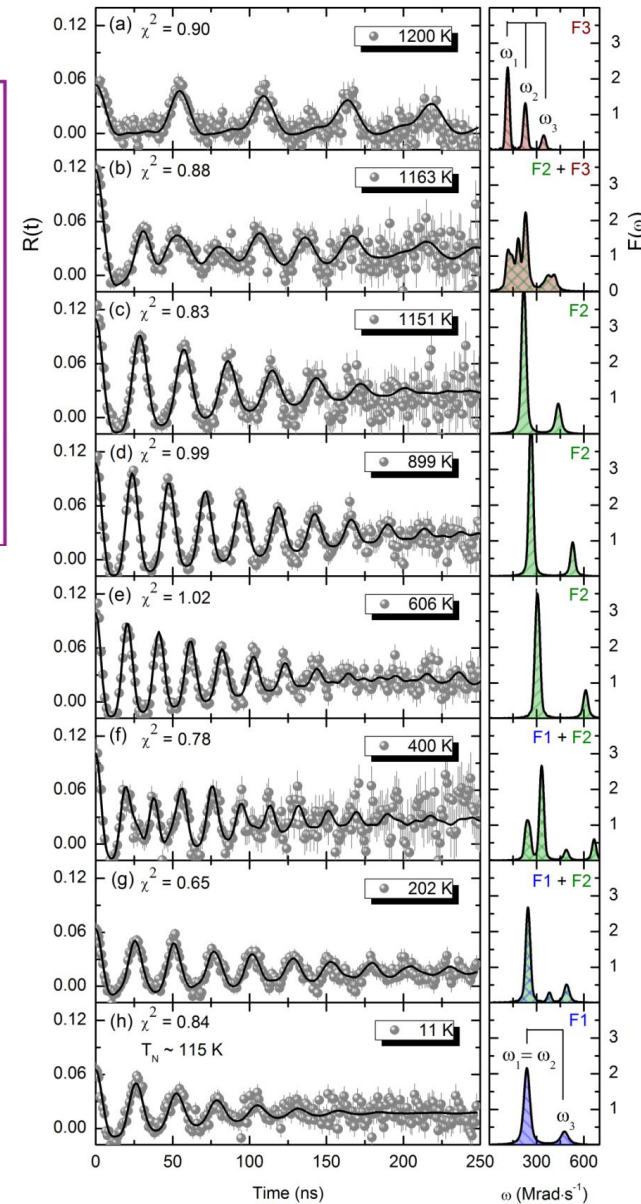
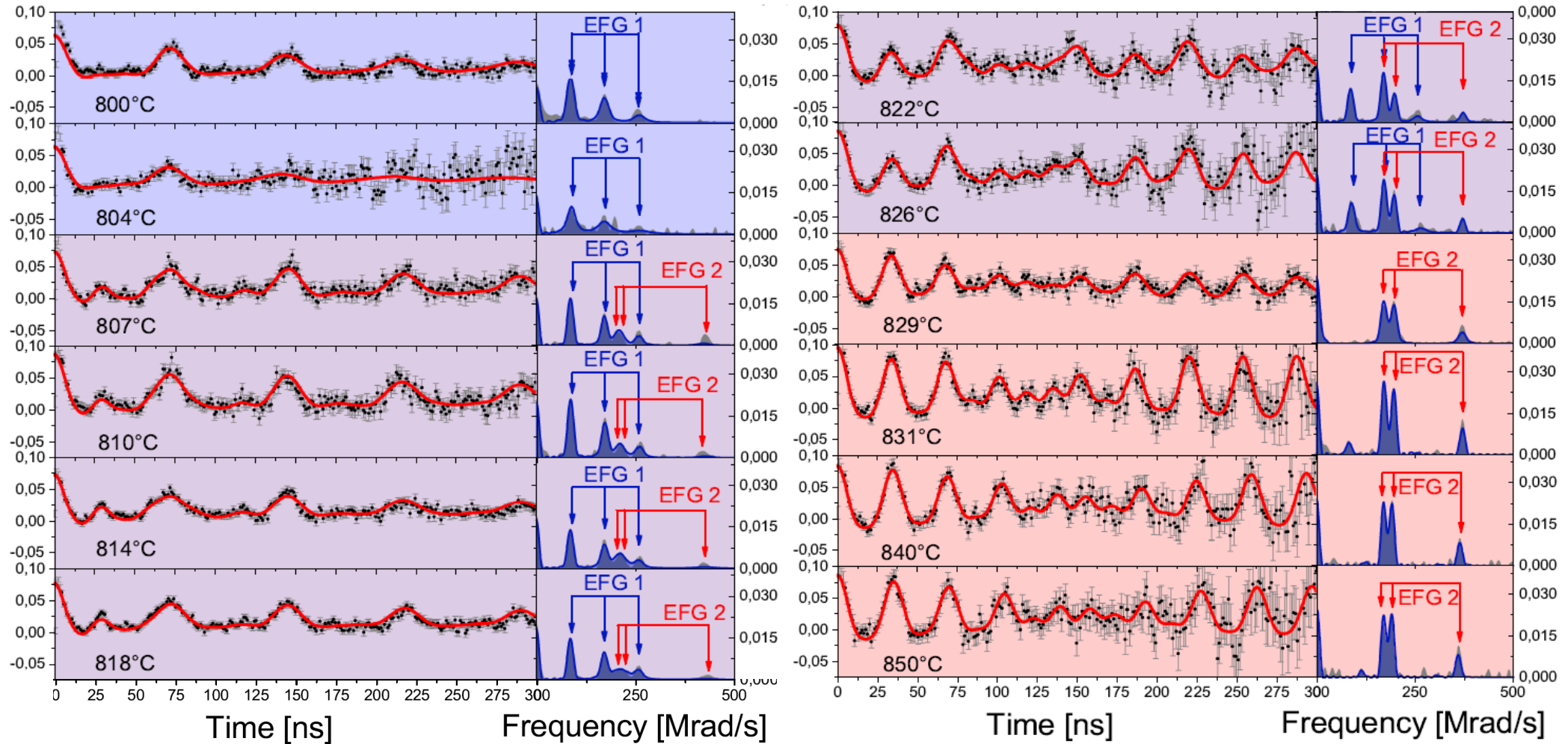
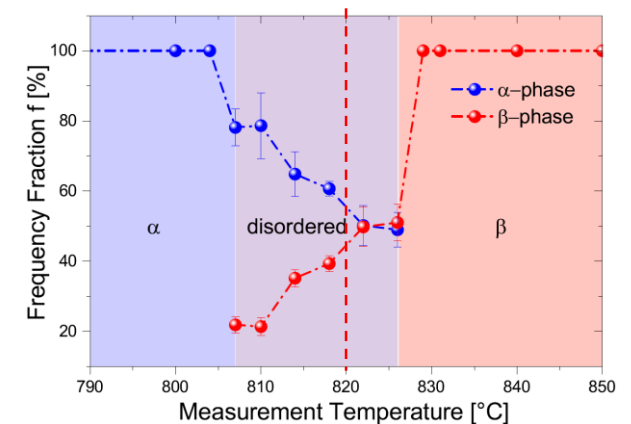
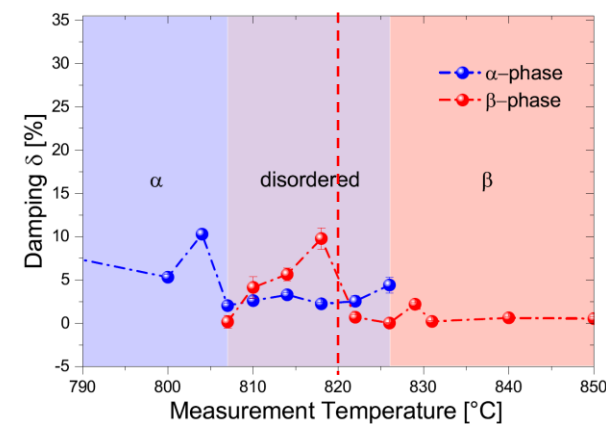
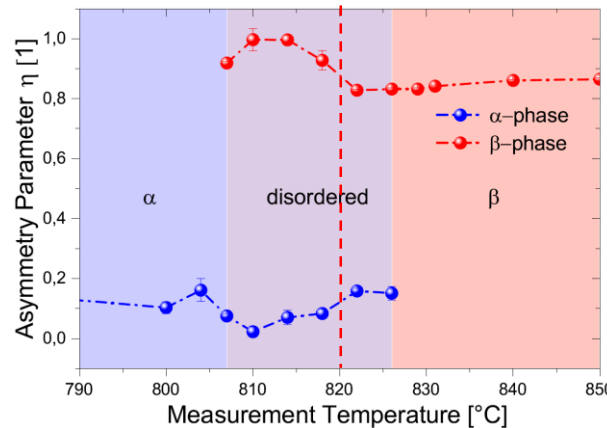
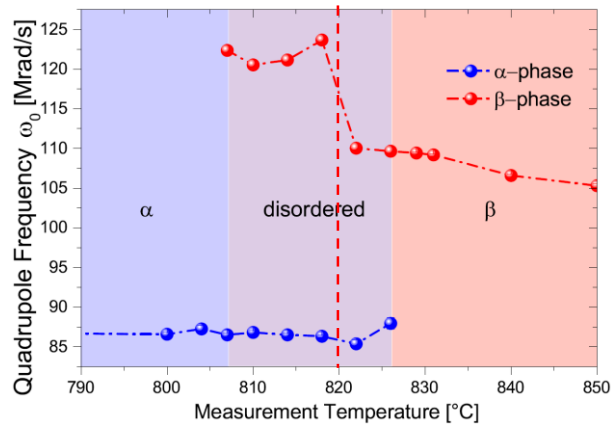
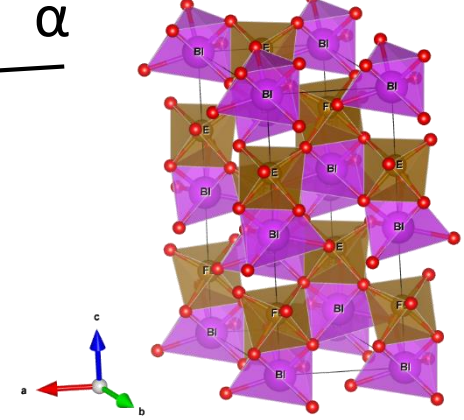
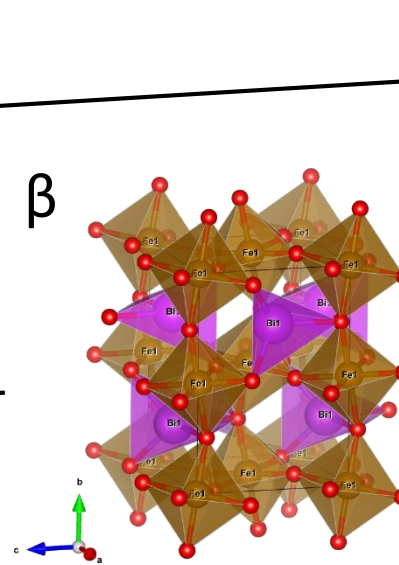
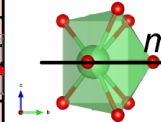
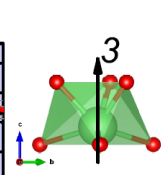
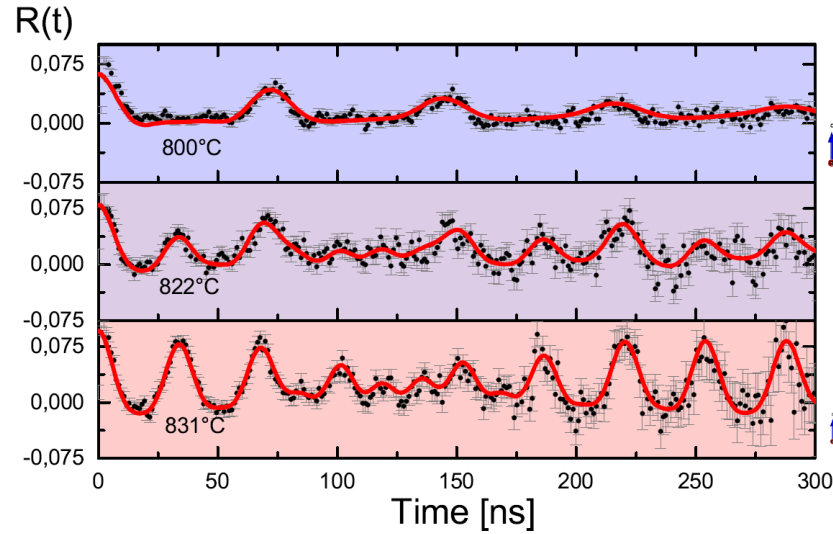


FIG. 3. Experimental EFG tensor at ^{111}Cd probe for the $\text{Ca}_3\text{Mn}_2\text{O}_7$ sample. (a) Principal component $|V_{zz}|$; (b) asymmetry parameter η ; (c) probe distribution. The dashed lines are a guide for the eyes.

Results in BiFeO₃

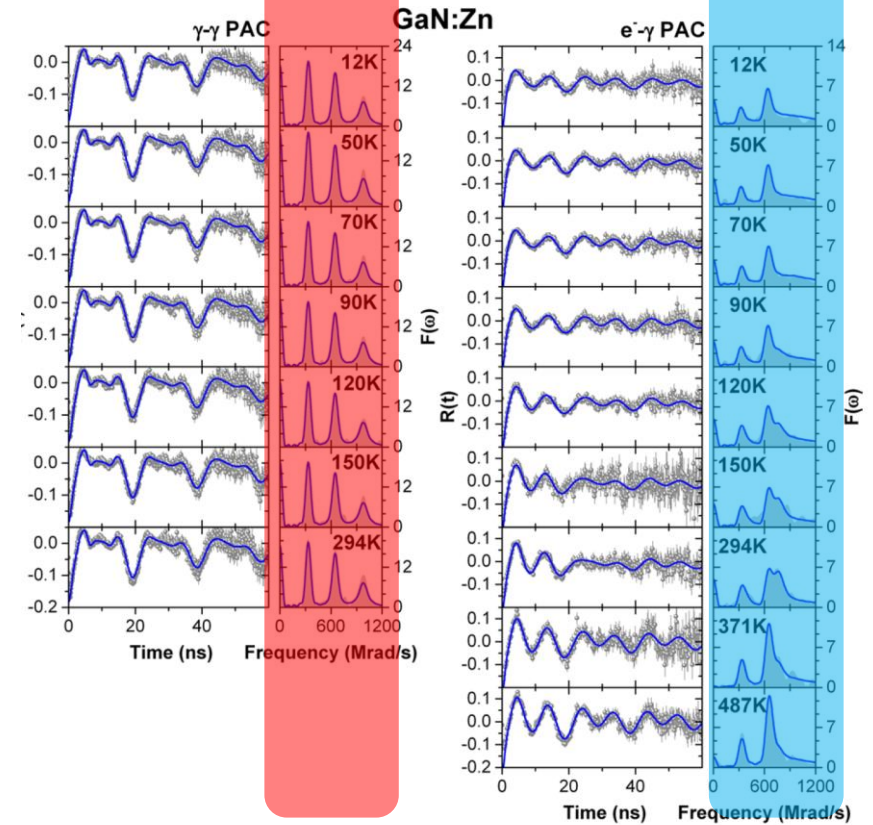
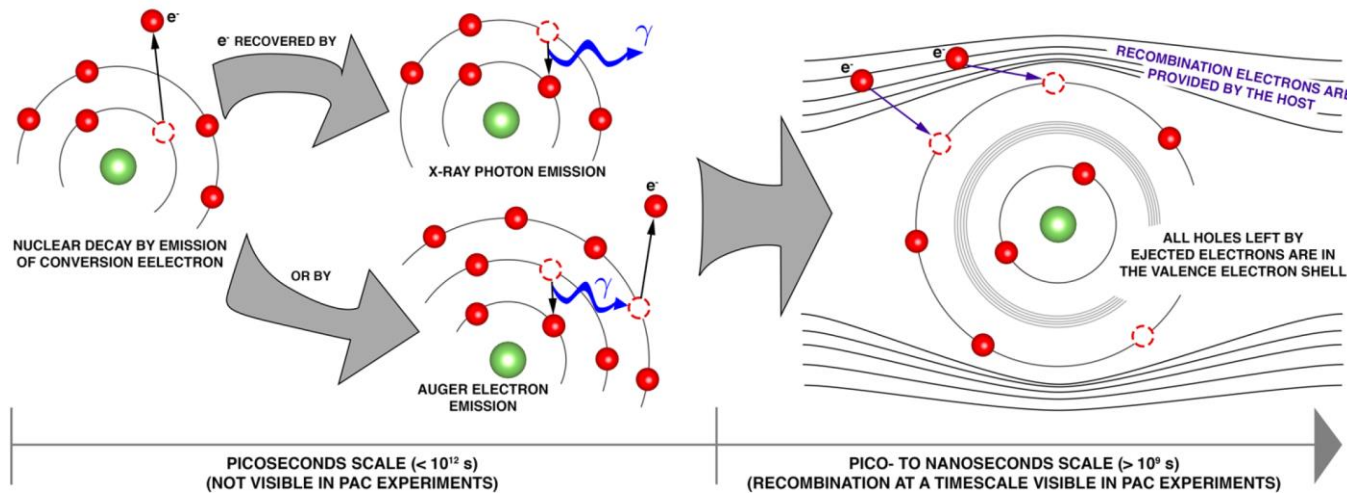


Results in BiFeO₃



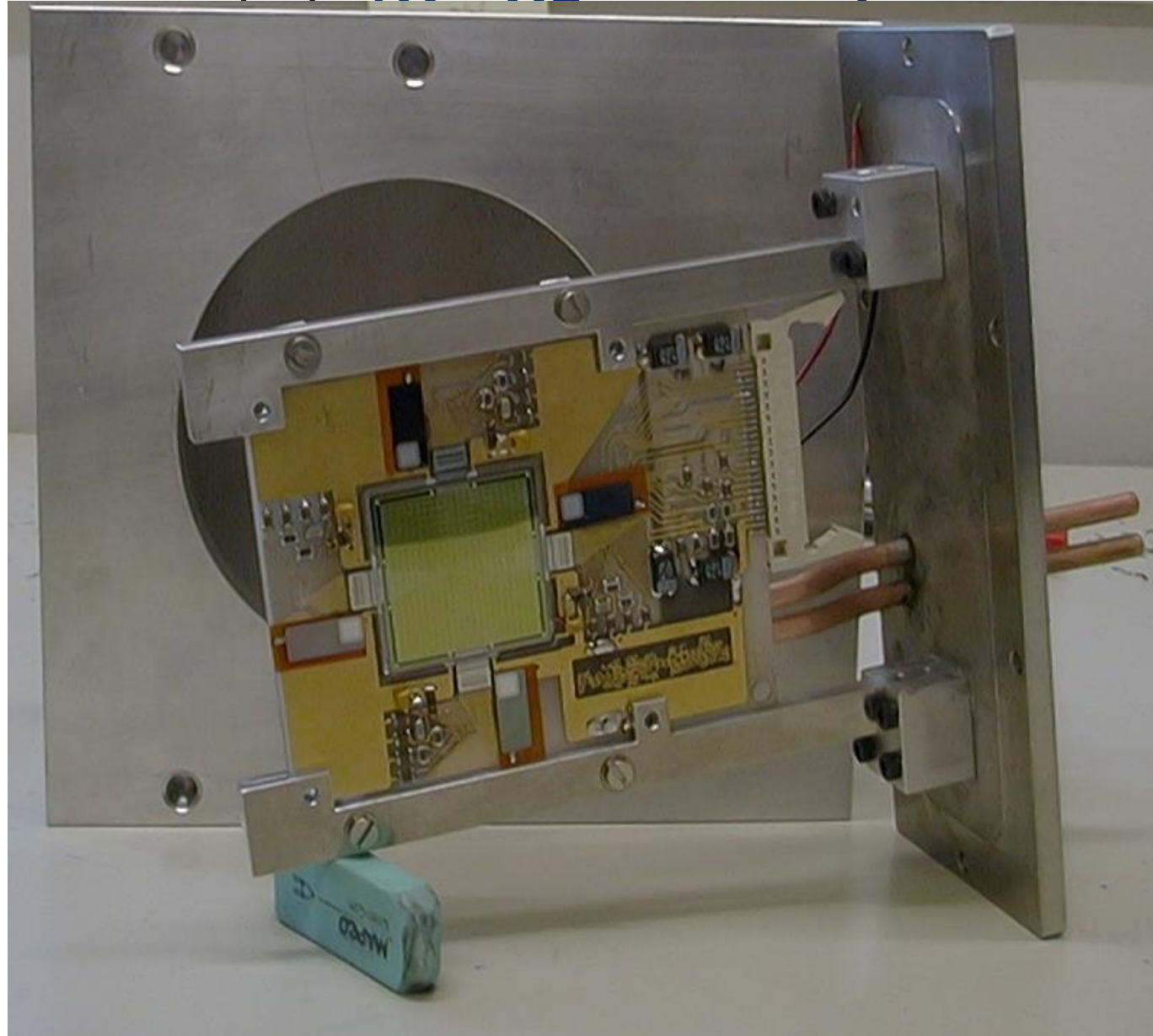
phase transition from rhombohedral α -BFO in $R3c$ setting to orthorhombic β -BFO with its $Pbnm$ space group at 820°C

Studying electronic properties in GaN without electrical contacts using γ - γ vs e^- - γ Perturbed Angular Correlations



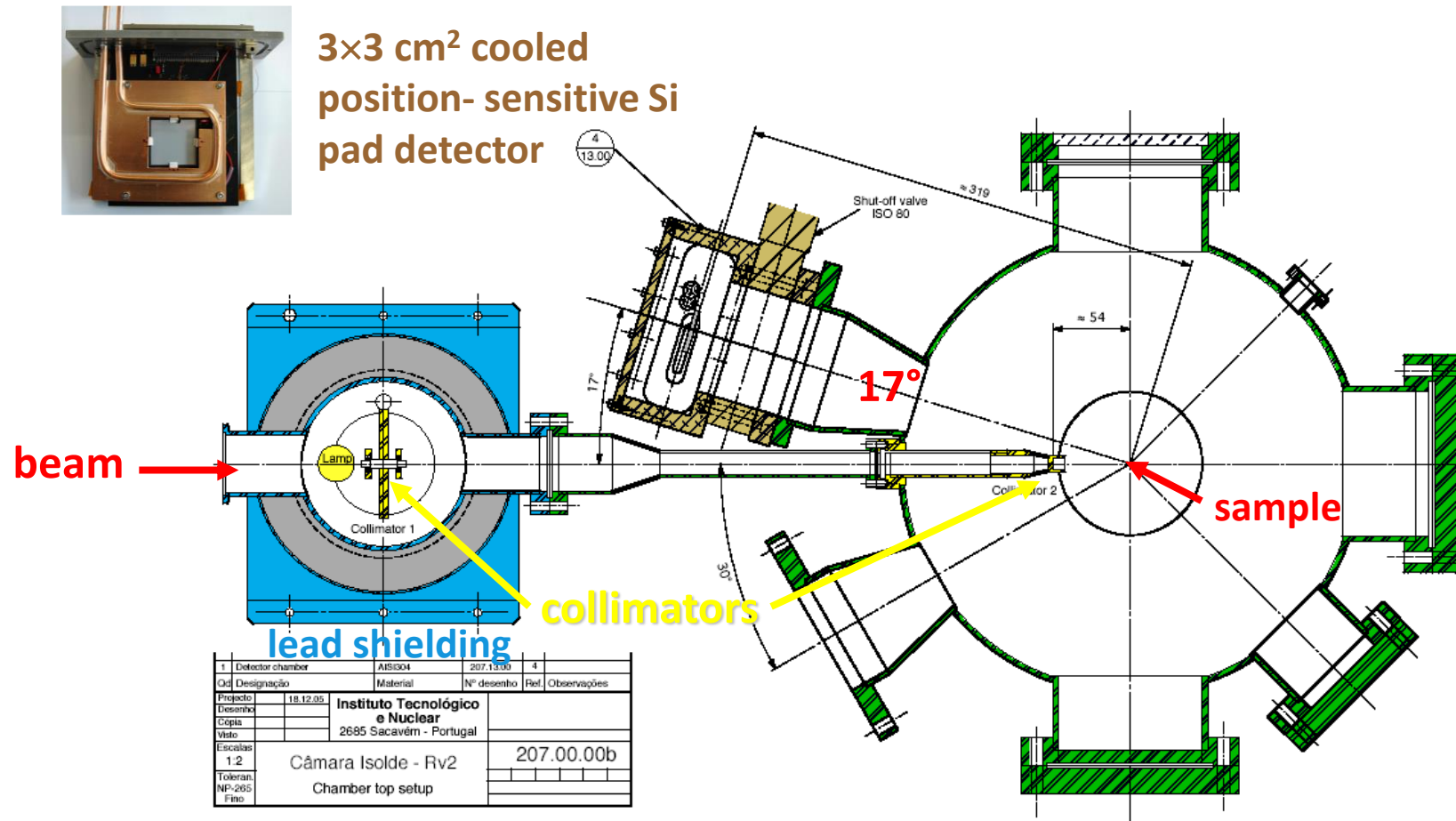
PAC revealing bulk properties of Si and Zn-doped Ga using γ - γ and e^- - γ PAC identification of double donor

Emission channeling lattice location inside the



angle

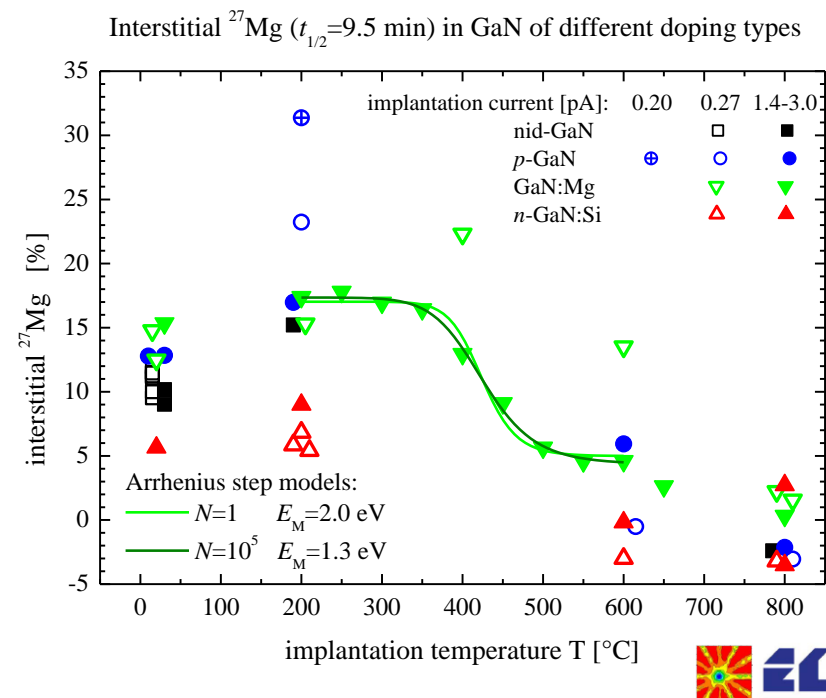
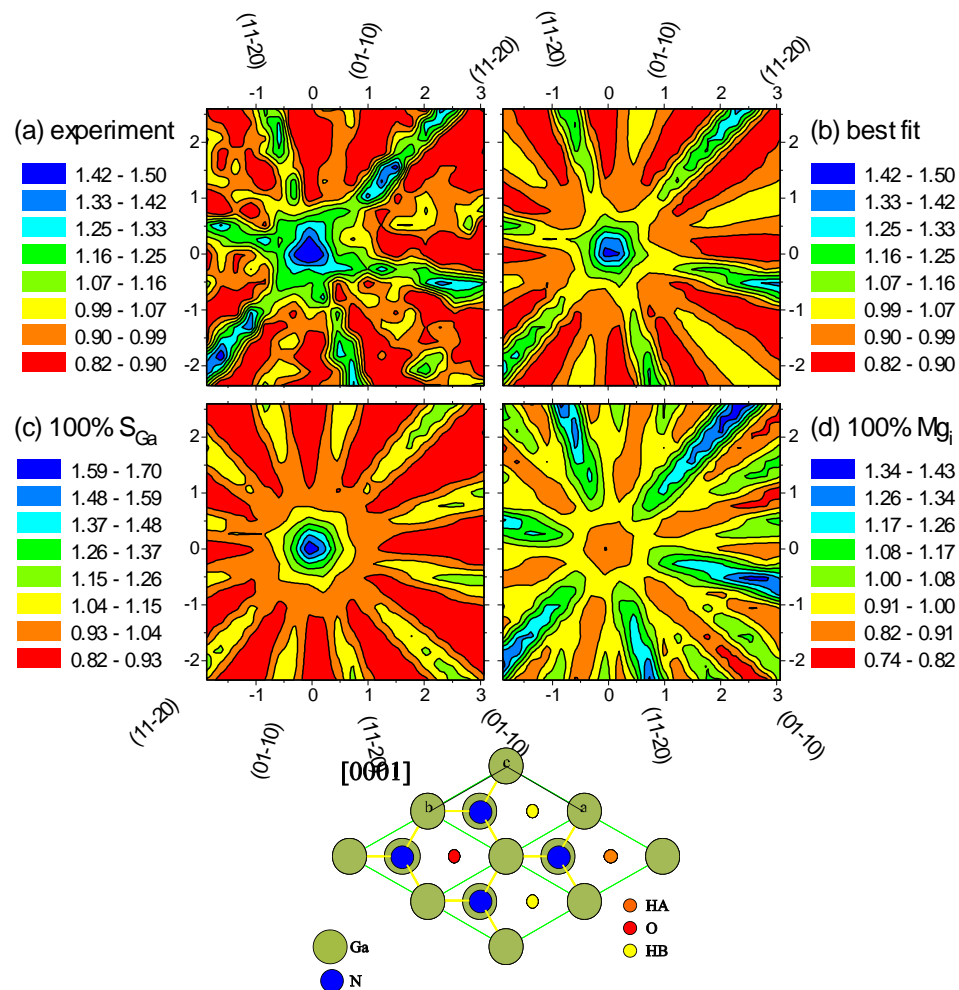
Emission channeling setup



- beam collimated on sample by two apertures (last one \varnothing 1mm)
- detector at 17° backward geometry for simultaneous implantation and measurement
- 22x22 pixels of 1.3 mm position-sensitive Si pad detector, water cooled

Emission channelling in semiconductors

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

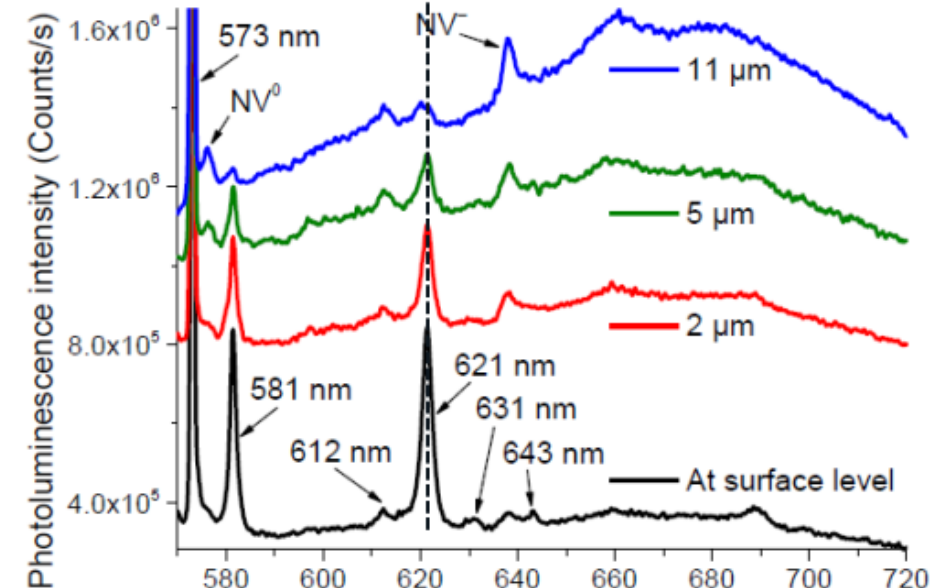
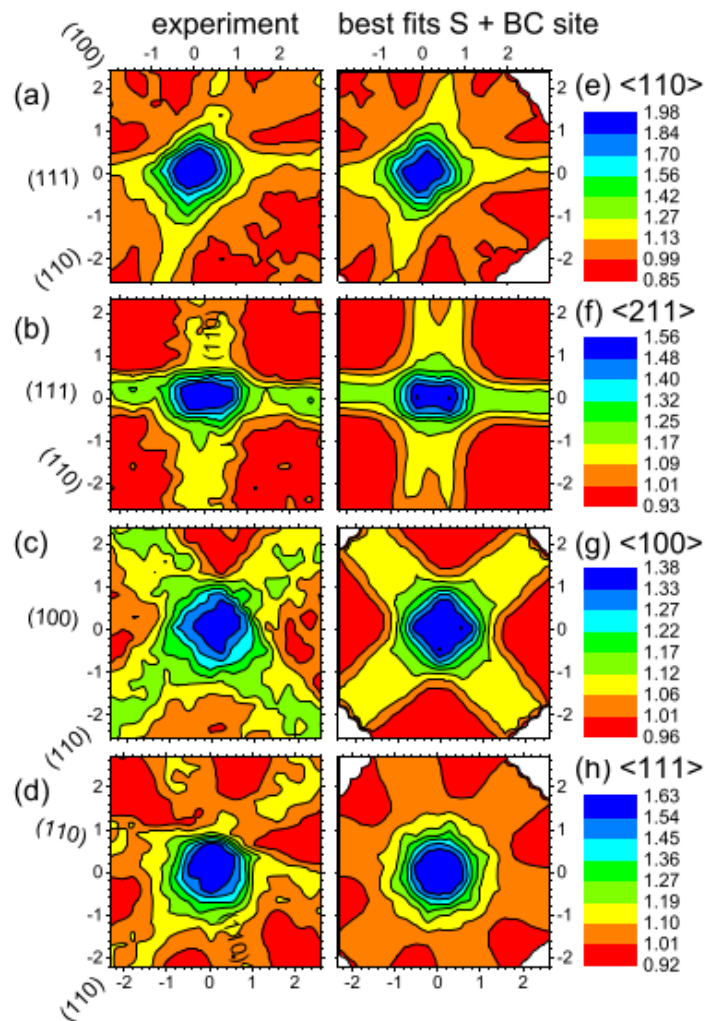
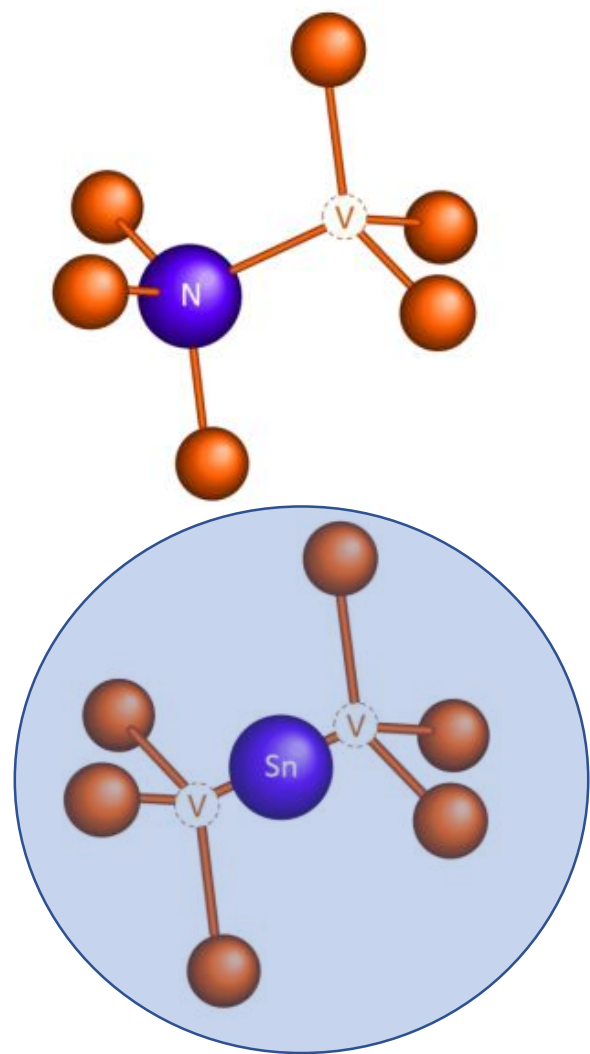


- Electron emission channeling patterns show mix of substitutional + interstitial ^{27}Mg

- Interstitial Mg fraction highest in $p\text{-GaN:Mg}$
- Lowest in $n\text{-GaN:Si}$
- \Rightarrow Direct evidence for amphoteric character of Mg that is coupled to the doping type
- Site change interstitial - substitutional Mg_{Ga}
- \Rightarrow Activation energy for migration of interstitial Mg: $E_M \gg 1.3 - 2.0$ eV

Colour Centres in diamond: future quantum bits?

Sn?

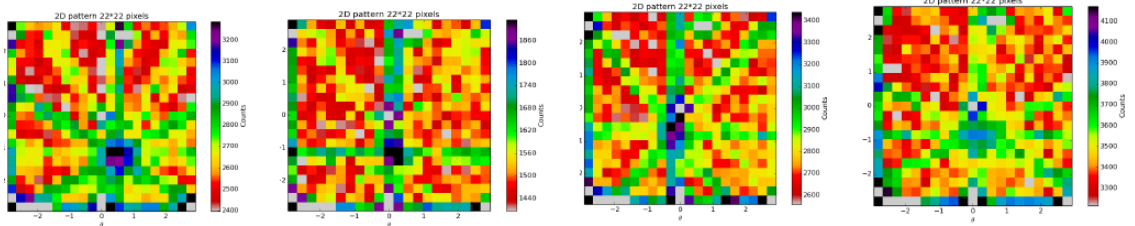


Direct Structural Identification and Quantification of the Split-Vacancy Configuration for Implanted Sn in Diamond

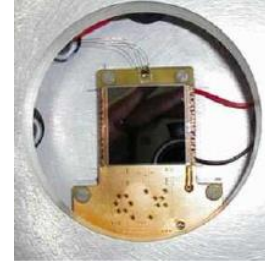
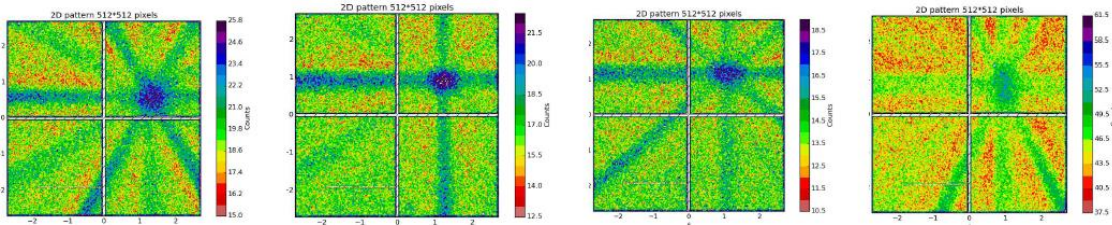
PHYSICAL REVIEW LETTERS 125, 045301 (2020)

New developments

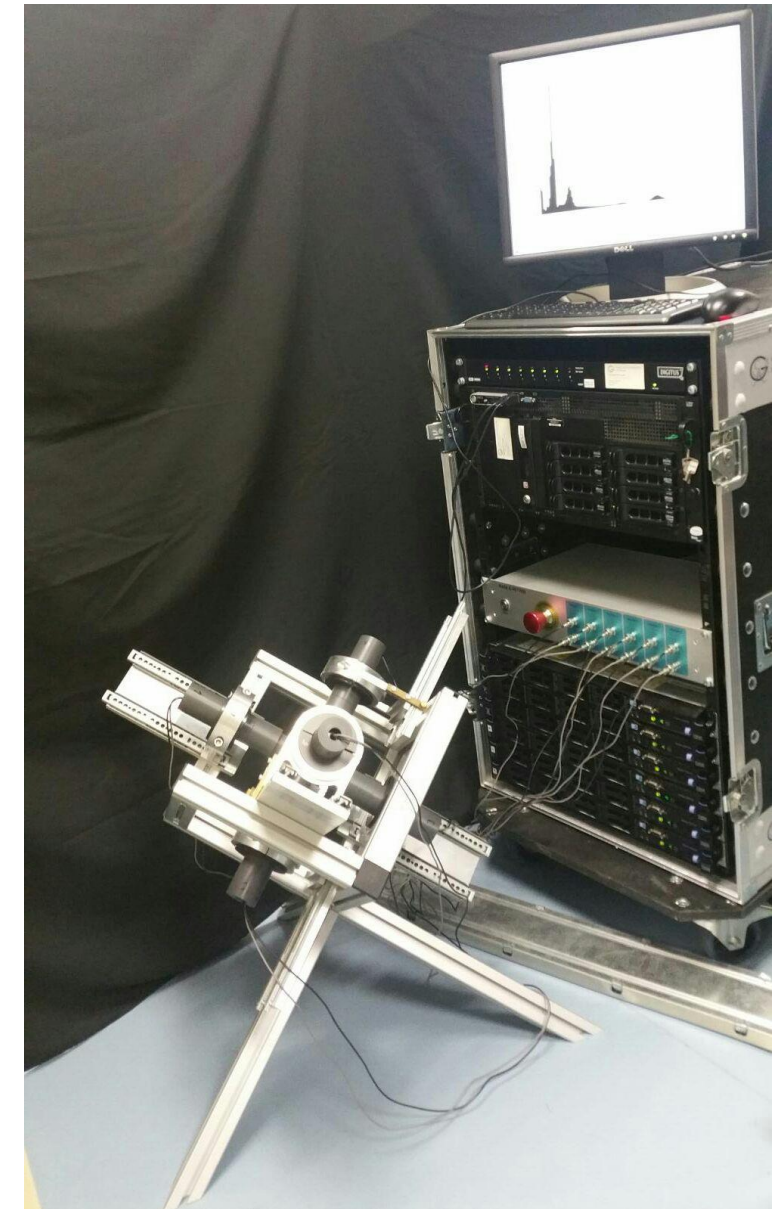
Annealing at 800° for 10min



Annealing at 800° for 10min



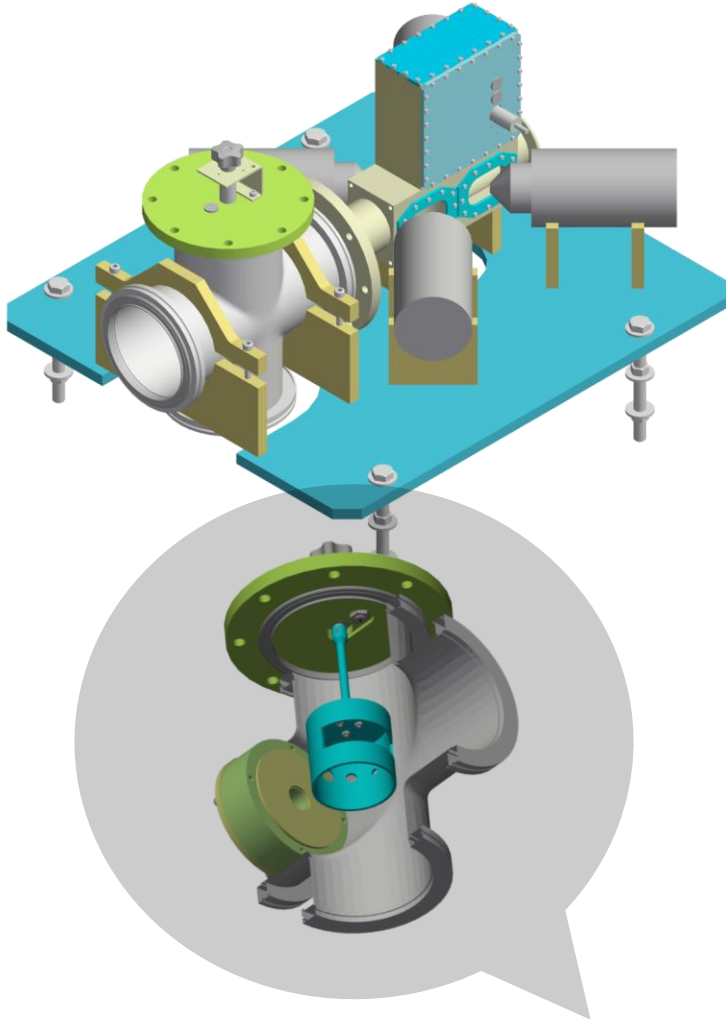
- Upgrading of detectors/spectrometers/upgrade DAQ.
- LaBr and CeBr detectors, allow for wider range of probes to be used.
- Spectrometer for biophysics for Mossbauer spectroscopy.



Controlling implantation

Implantations on ice:
biology and surface science

Online setup for ^{68}Cu PAC



Surface implantation



Summary

- Ability to probe magnetic properties in dilute and sensitive way is essentially unique to hyperfine techniques such as PAC and Mossbauer and most radioactive methods... extremely attractive and becoming more so: devices are not getting any bigger...
- The interface of materials can be impossible to study reliably...radioactive probes are the ideal tool
- Flexibility of PAC techniques allows for temperature, surface, magnetic and other perturbative studies in materials
- Chemical information which can be addressed with radiotracer methods could reveal hidden aspects of quantum materials
- Especially powerful when various techniques are combined.
- With the range of isotopes available at ISOLDE can always attack a problem in a fresh and unique way...

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

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