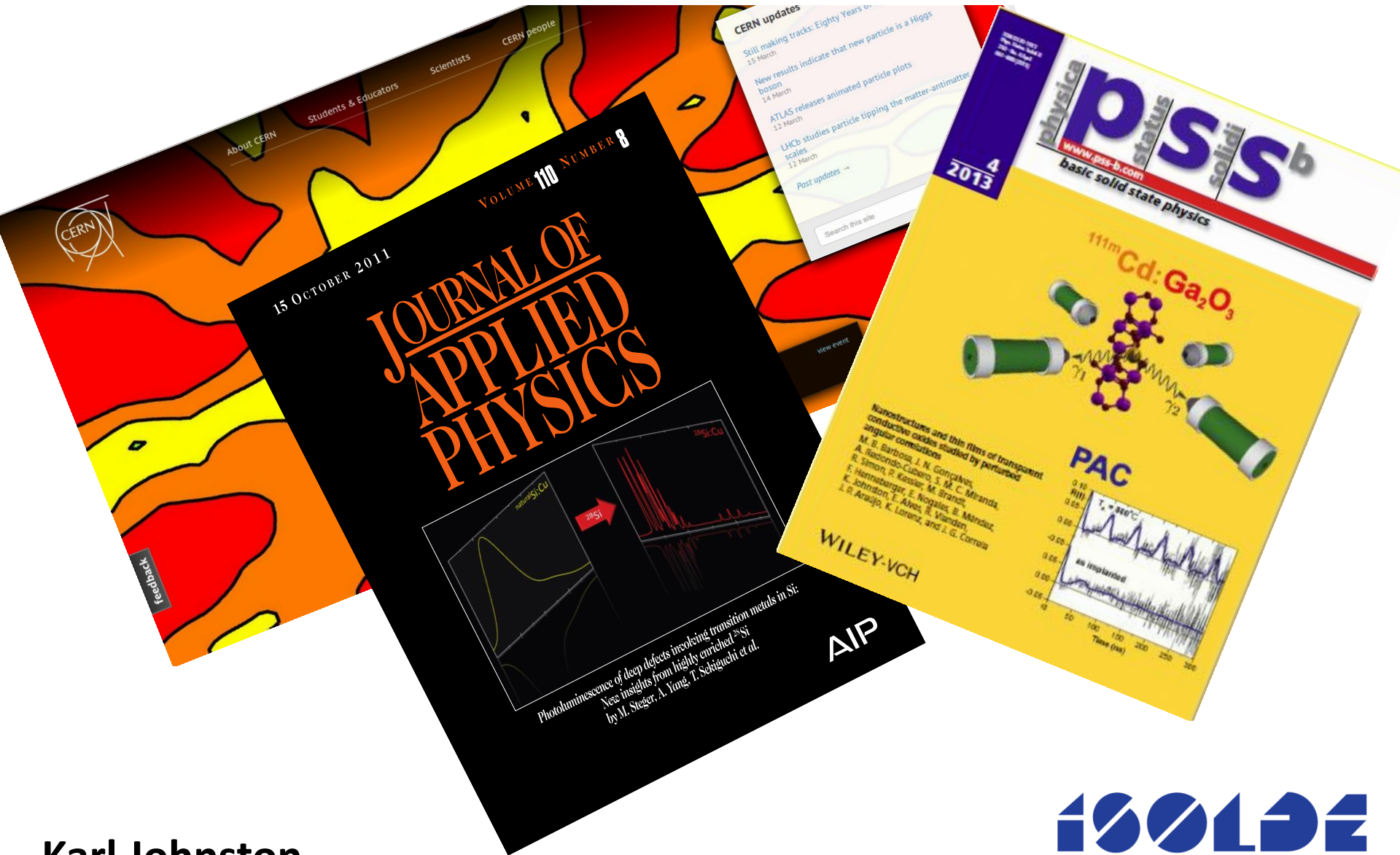


Status report of Solid State Physics, Biophysics and Collections at ISOLDE



Outline

- Summary of the materials/systems under study
- Overview of how the various techniques are sub-divided between experiments.
- Main results from experiments: semiconductors; Materials; biophysics; medicine; “miscellaneous”...
- Request for retaining shifts
- Brief addendum to IS486
- End

Variety!

Semiconductors

1. IS432/450
2. IS453
3. IS481
4. IS486
5. IS489
6. IS492
7. IS501
8. IS544

- Si
- Ge
- CdTe
- ZnO
- AlN
- InN
- Ga₂O₃

Biophysics/chemistry

12. IS488

- Model systems
- Proteins: designed and natural

Medicine

13. IS528

- Innovative isotopes for treatment/imaging: **Tb**

Multiferroic/nanomaterials/misc

9. IS487
10. IS514
11. IS515

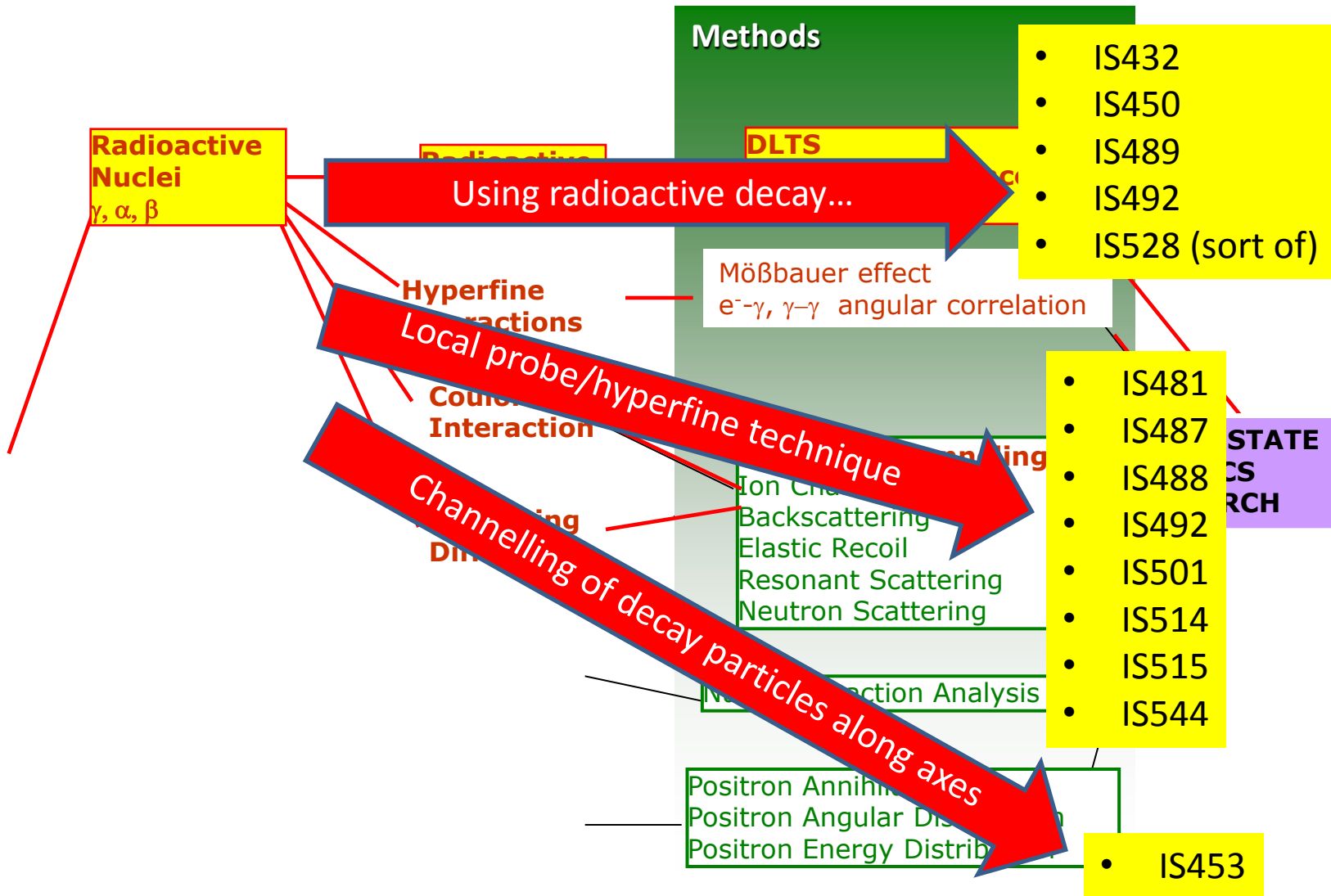
- RMNO₃
- Spinel CdCr₂S₄
- Intermetallic compounds
- Magnetic Nanoparticles

Collections (e.g. neutrino physics)

14. IS500
15. IS533

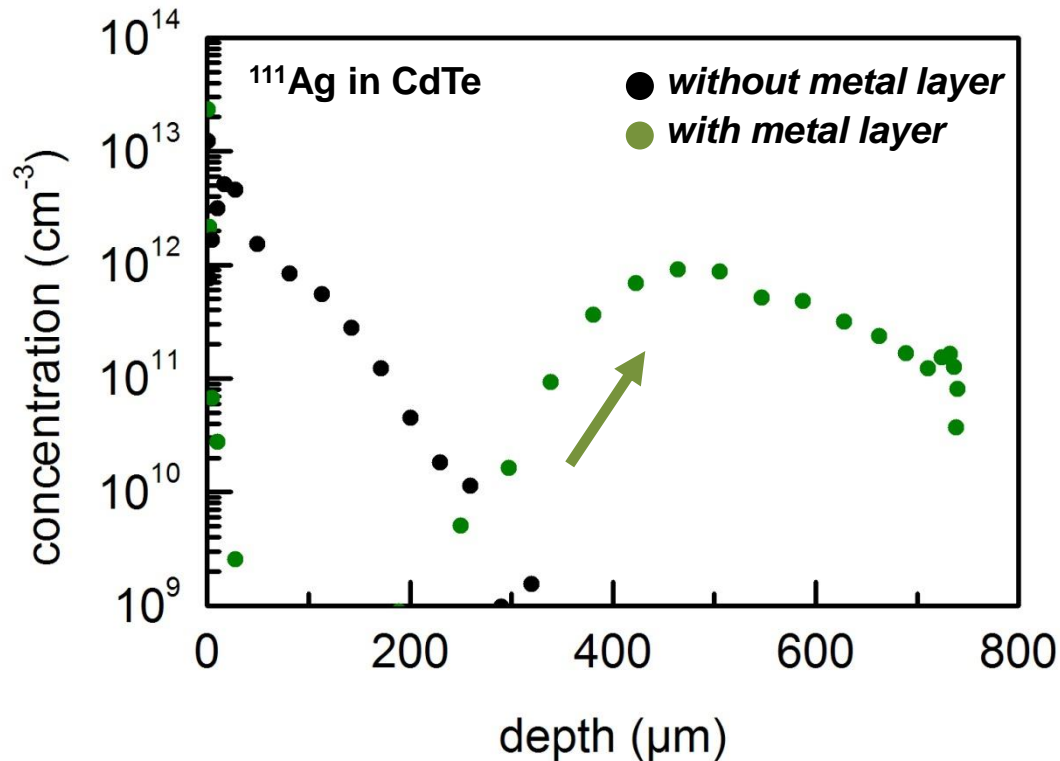
- Long-lived sources for calibration of experiment, ⁸³Rb

Applying radioactivity to solid state physics

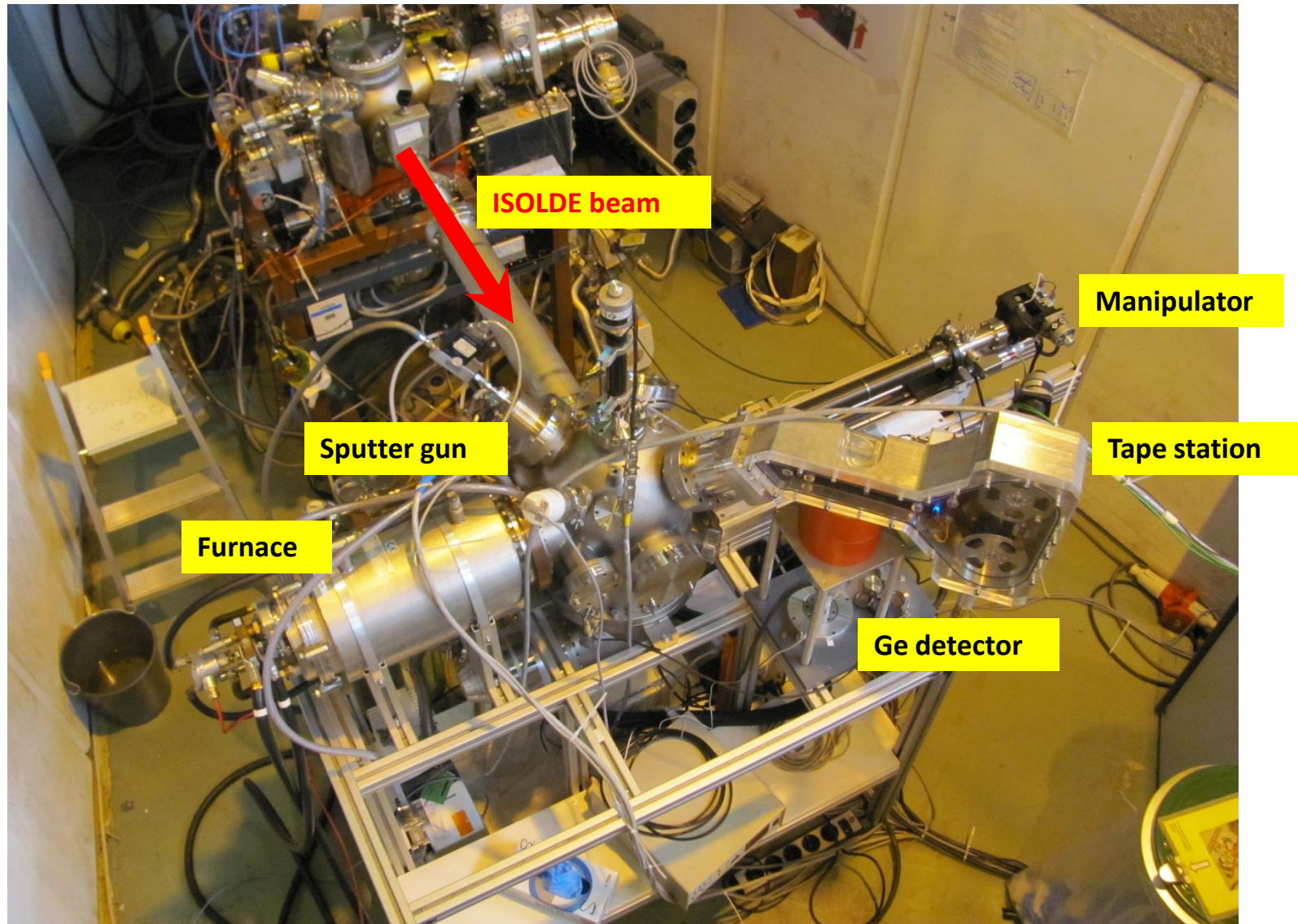


Diffusion without layer at 500 K

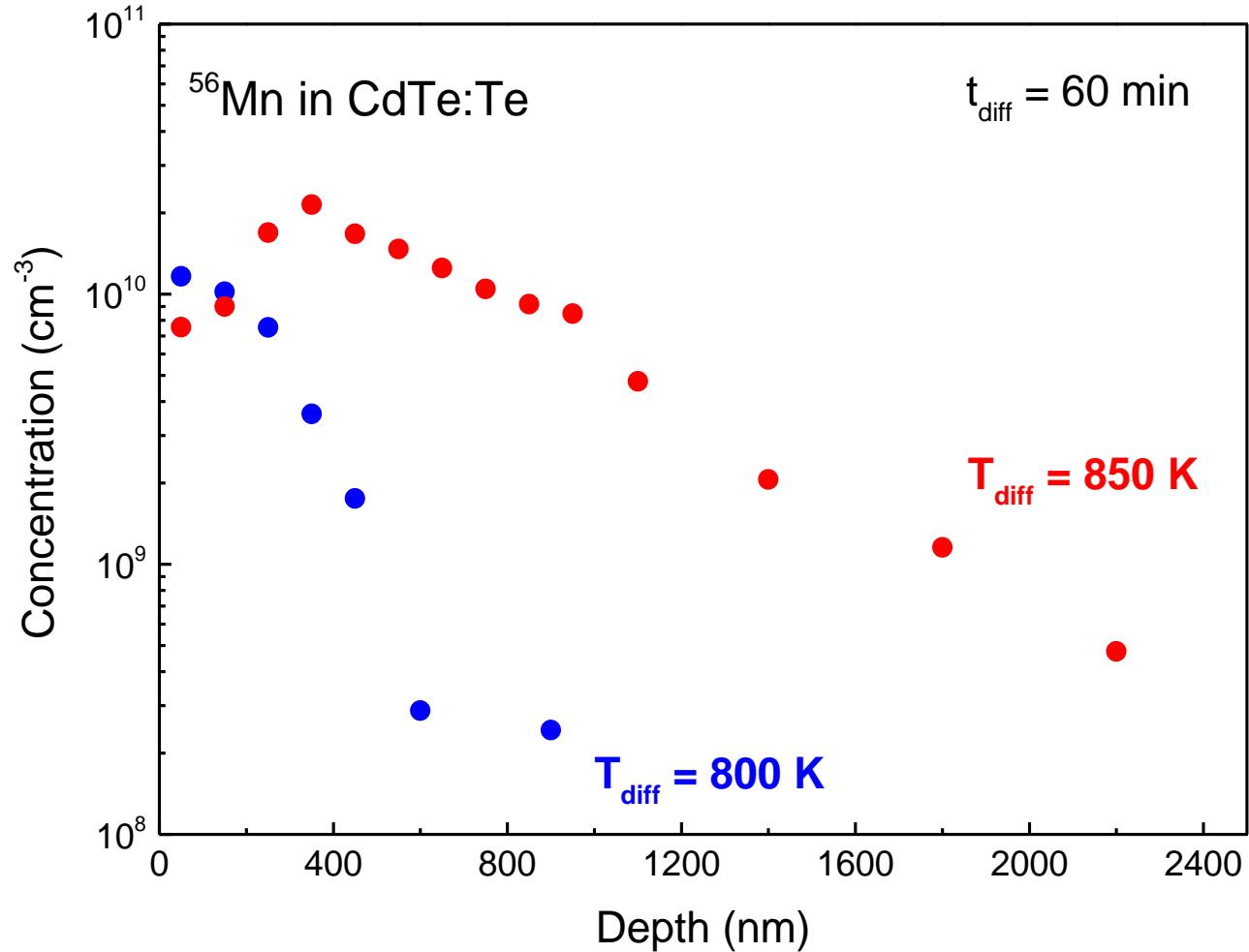
Diffusion with Cu layer at 500 K

**Metal layers can initiate uphill diffusion**

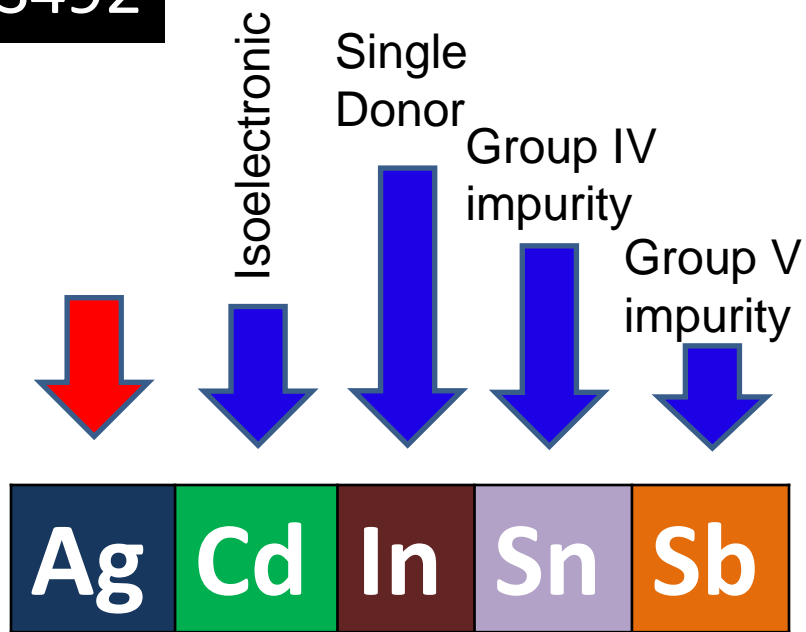
2012



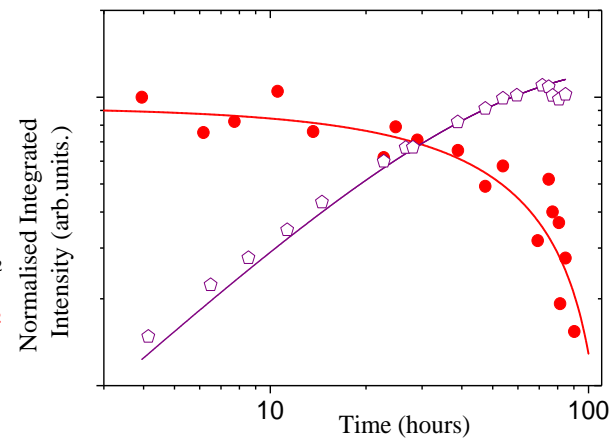
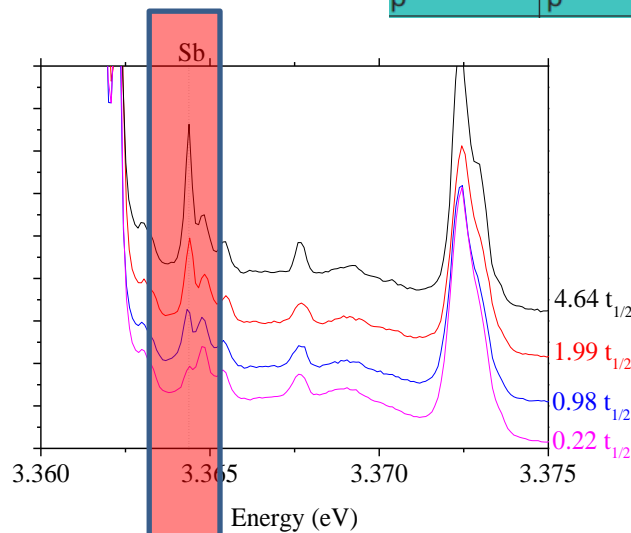
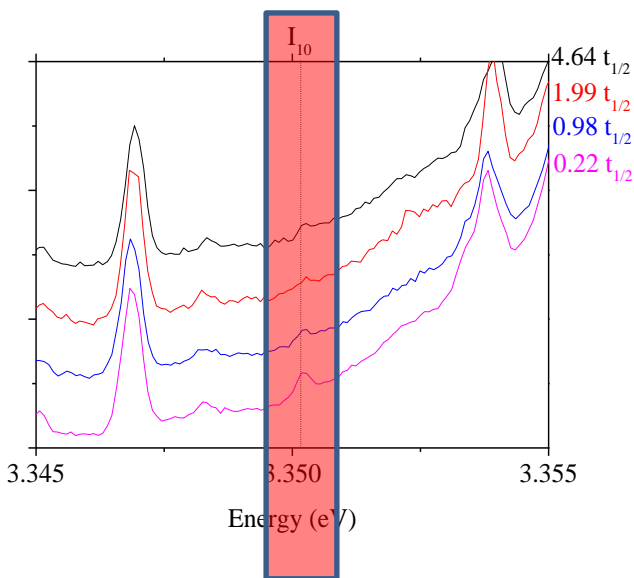
Commissioning of online diffusion chamber...Diffusion of metals in CdTe



IS492

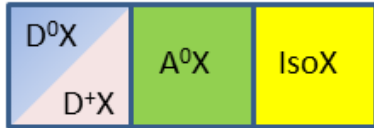


Sb121 5/2+ 57.36 0+ 32.59 β^-	Sb122 2.70 d 2- EC, β^- *	Sb123 7/2+ 42.64 0+ 4.63 β^-	Sb124 60.20 d 3- β^- *	Sb125 2.7582 y 7/2+ β^- *
Sn120 0+ 32.59 *	Sn121 27.06 h 3/2+ β^- *	Sn122 0+ 4.63 *	Sn123 129.2 d 11/2- β^- *	Sn124 0+ 5.79 *
In119 2.4 m 9/2+ β^- *	In120 3.08 s 1+ β^- *	In121 23.1 s 9/2+ β^-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



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

IS492



Tentative D^0X/A^0X



Defect complex?

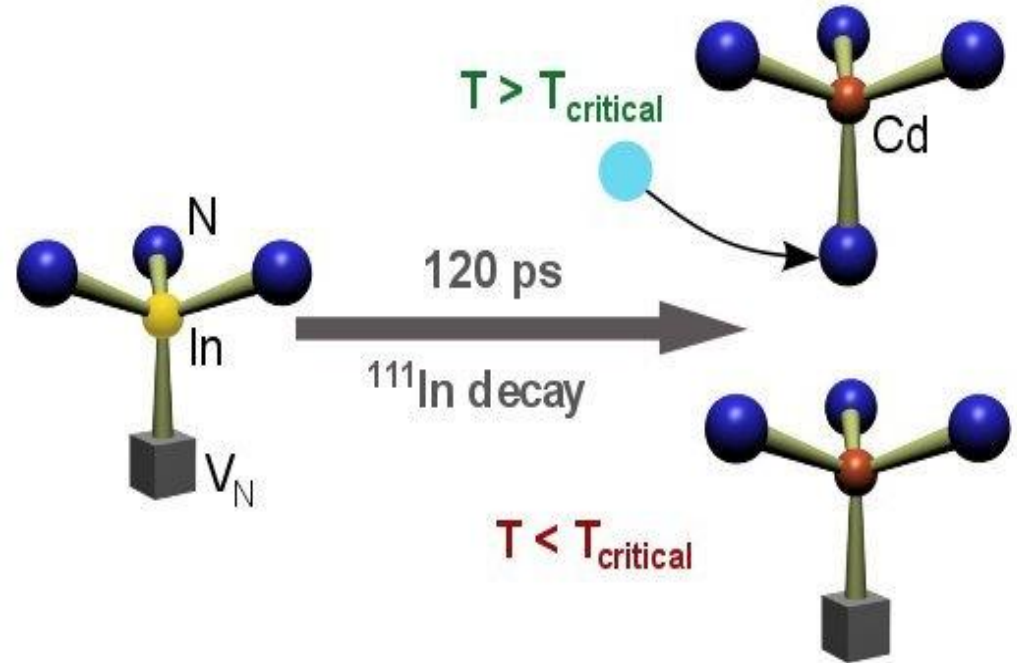
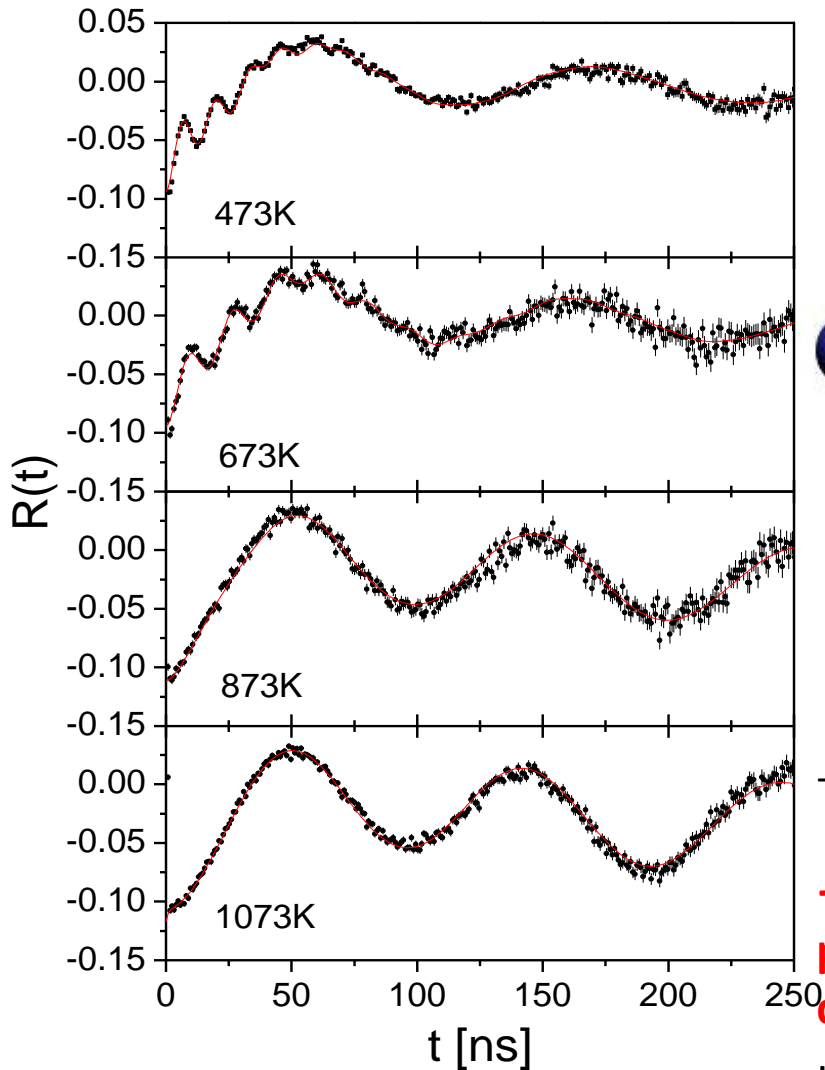
		B	C	N
		I_{6a} Al $I_0?$	Si	P
Cu	Zn	I_8 Ga I_1	Ge	As \times
Ag	\times Cd	I_9 In $I_2?$	I_{10} Sn	Sb \checkmark
Au	\checkmark Hg	Tl	Pb	Bi

- 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)

Defects and electronic effects in wide bandgap semiconductors

IS481

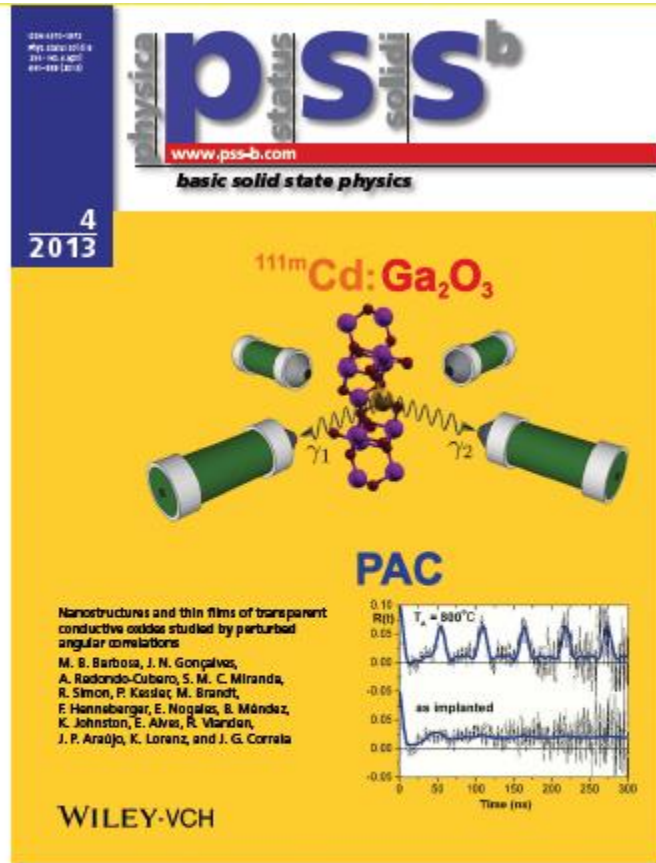


- The role of In in GaN and AlN has been studied by PAC:
→ A defect, probably V_N , is stabilized at the ^{111}In probe and is released at high temperatures after the decay to Cd.

Kessler et al. HFI 197 (2010) 187; pss c 9 (2012) 1032

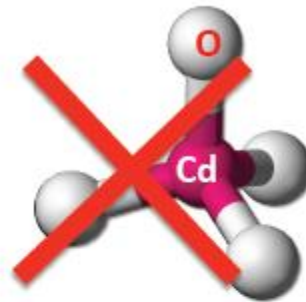
Electronic effects in Ga₂O₃

IS481

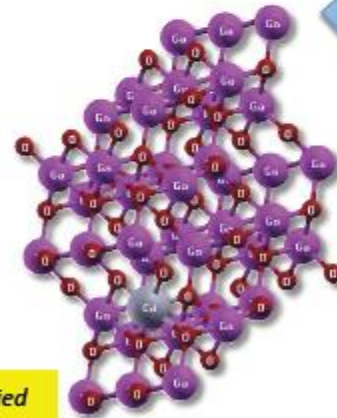
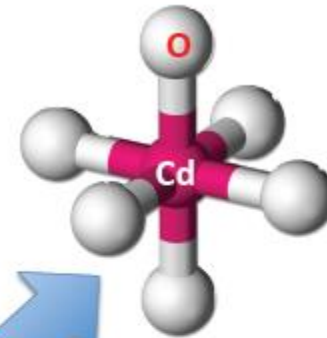


Nanostructures and thin films of transparent conductive oxides studied by perturbed angular correlations, M. B. Barbosa et al.
Phys. Status Sol. B 250, No. 4, 801–808 (2013)
DOI 10.1002/pssb.201200923

Tetrahedral Ga site



Octahedral Ga Site



PAC data + DFT simulations show that the (potential p-type) dopant Cd can only be surrounded by 6 oxygen atoms at the octahedral site

4

→ Future work:

Studying electronic effects in Ga₂O₃ nanostructures and bulk crystals using γ - γ and e- γ PAC

Motivation

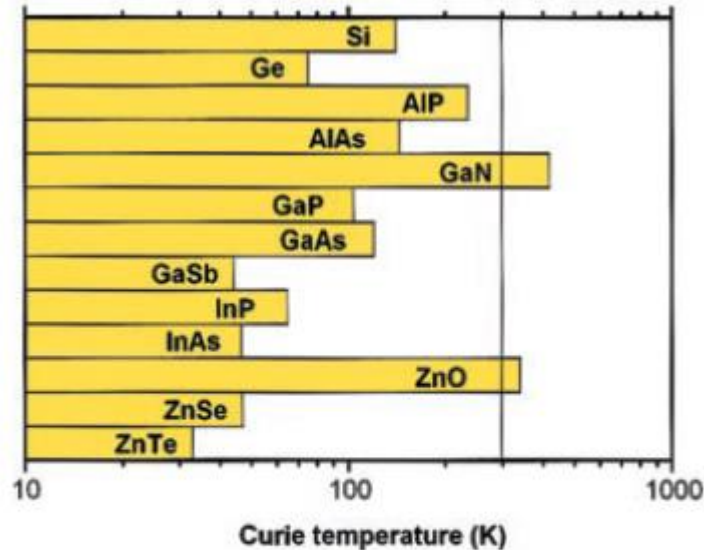


Fig. 3. Computed values of the Curie temperature T_C for various p-type semiconductors containing 5% of Mn and 3.5×10^{20} holes per cm^3 .

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.

Also motivation for IS432, IS450 and IS453

Fe: ZnO a ferromagnetic semiconductor?

IS501

(no!)

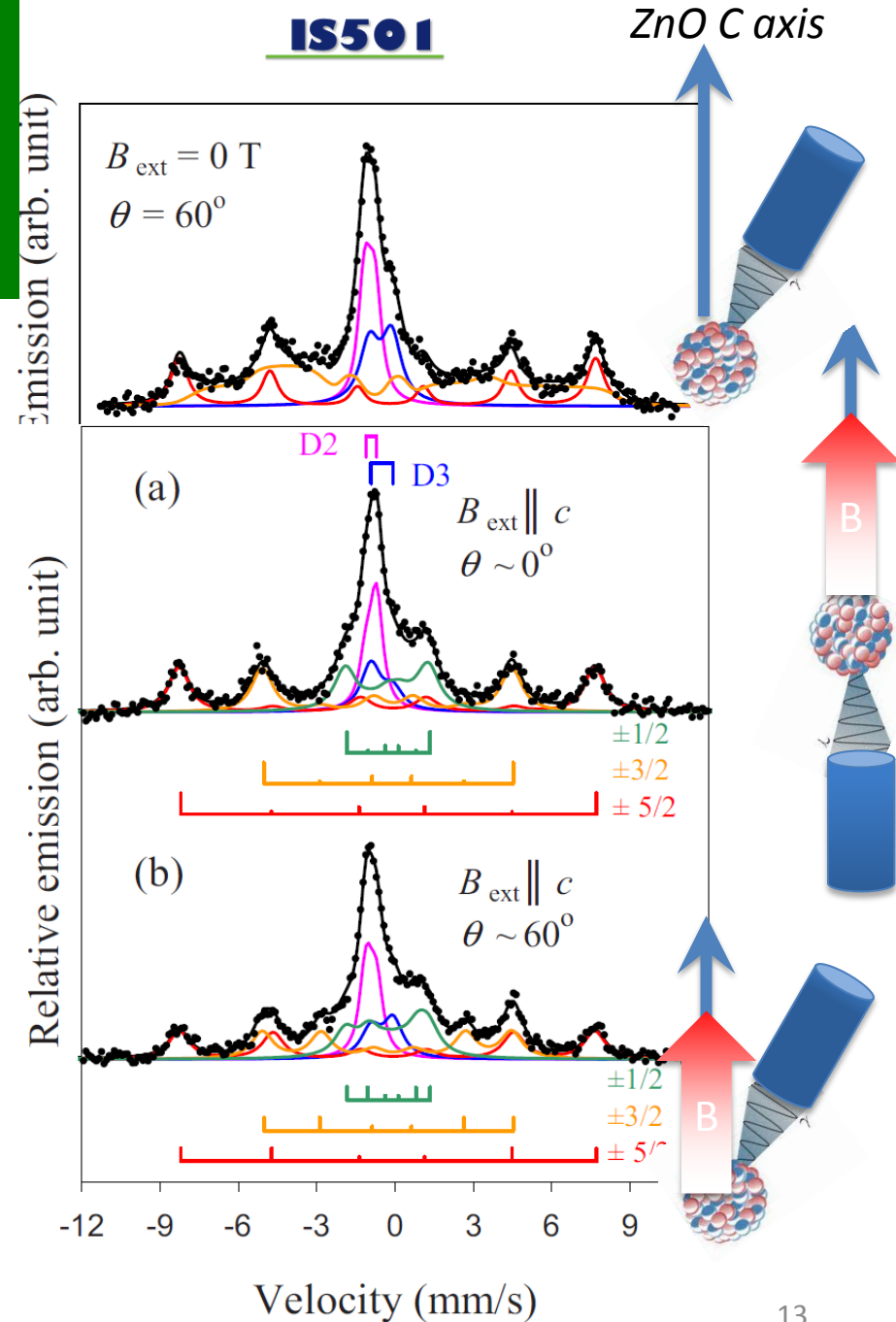
6 fold spectrum: characteristic of magnetic structure (at room temperature!!!).

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

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

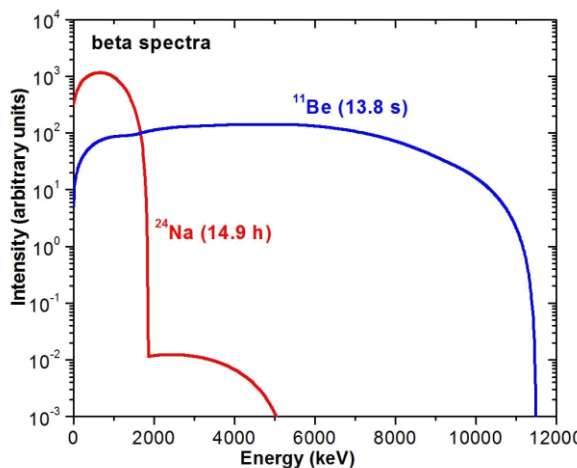
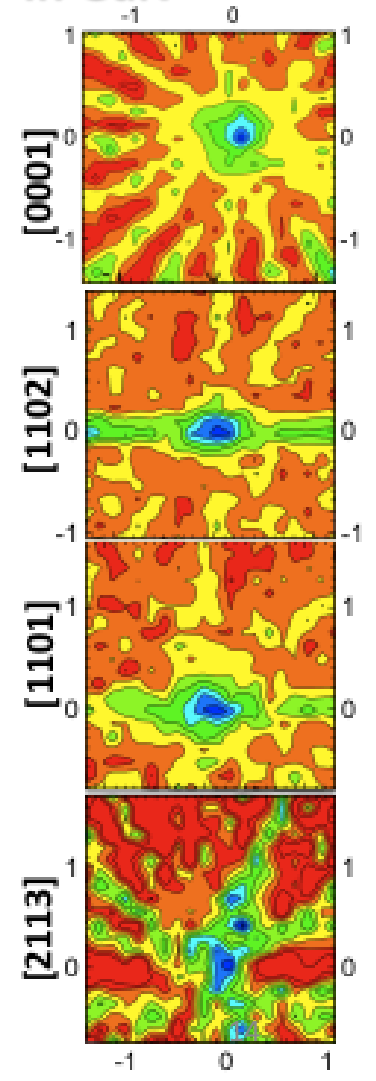
After high-dose implantations, precipitates of Fe-III are formed. These form **clusters** yielding misleading information about the nature of magnetism in ZnO (as reported by many groups over the last number of years).

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

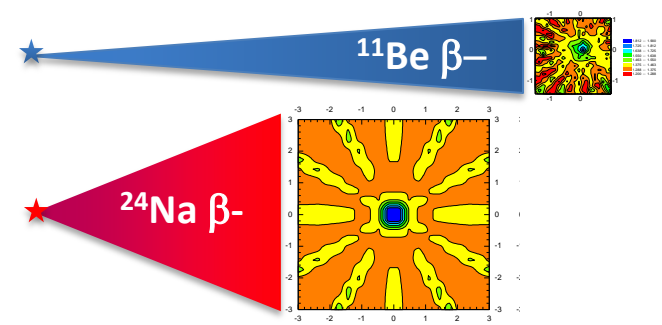


H	2012 probe isotopes																He						
Li	Be																	B	C	N	O	F	Ne
Na	Mg	2009																Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr						
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe						
Cs	Ba	La	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn						
Fr	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								

first ever done β^- EC patterns using ^{11}Be (14 s) in GaN



Source <-> detector : 60 cm



Source <-> detector : 30 cm



Emission channeling studies on a challenging case of impurity lattice location: cation versus anion substitution in transition-metal doped GaN and ZnO

L.M.C. Pereira, U. Wahl, J.G. Correia, L.M. Amorim, D.J. Silva, S. Decoster, M.R. da Silva, K. Temst, and A. Vantomme



Award for Best Student Oral Presentation at the 17th International Conference on Radiation Effects in Insulators (REI-17), in Helsinki, Finland, 30.6.-5.7.2013

Lattice sites of implanted Mg in the group-III nitrides

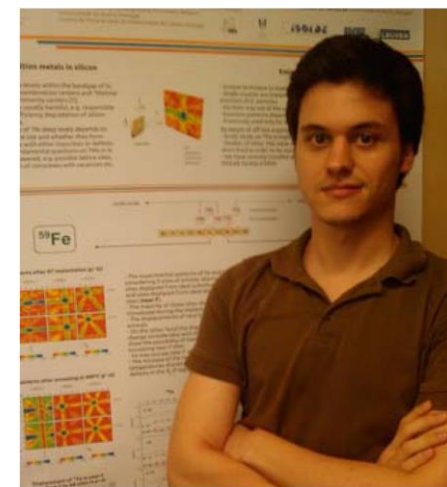
L.M. Amorim, U. Wahl, S. Decoster, L.M.C. Pereira, J.G. Correia, D.J. Silva, K. Temst and A. Vantomme



J.W. Corbett prize at the 27th International Conference on Defects in Semiconductors (ICDS-27), in Bologna, Italy, 21.-26.7.2013

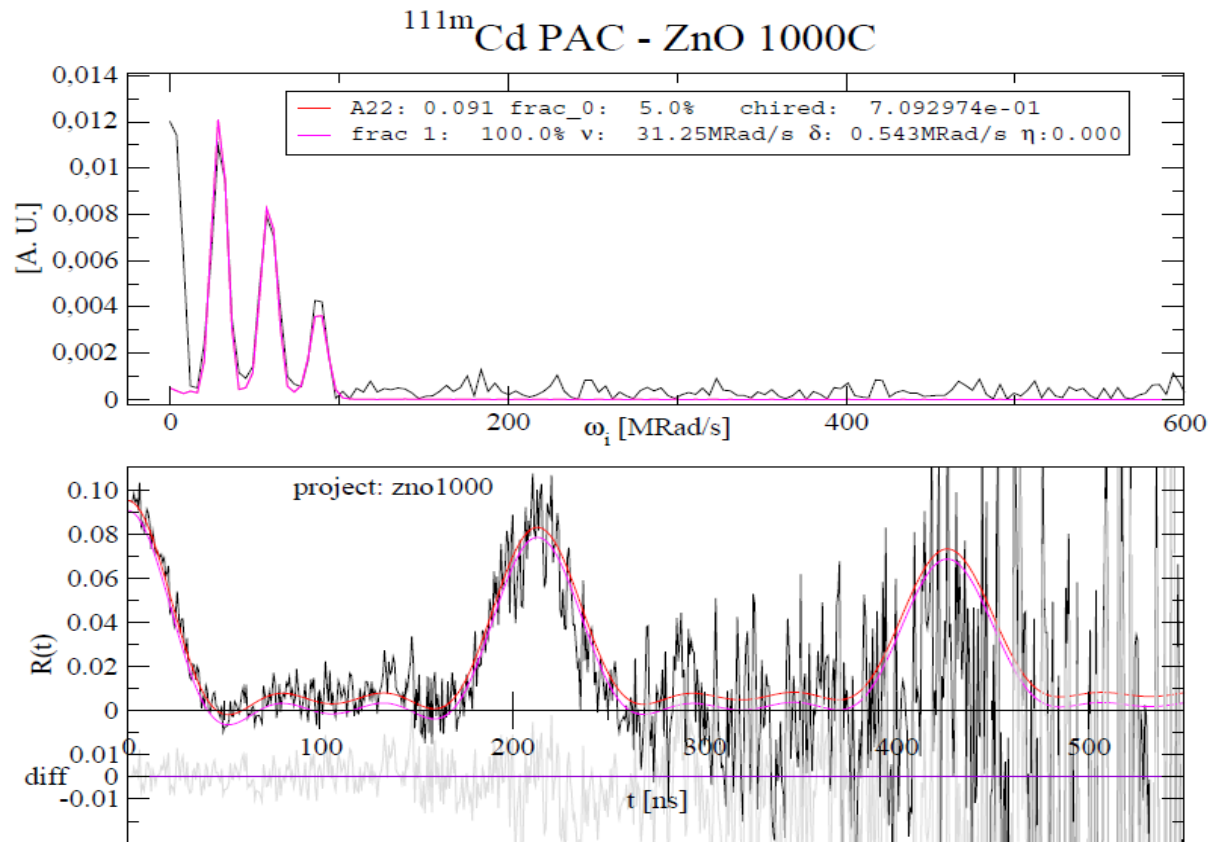
Influence of the doping on the lattice sites of Fe in Si

D.J. Silva, U. Wahl, J.G. Correia, and J.P. Araújo



Gauging Structural Aspects of ZnO nano-Crystal Growth Through X-ray Diffraction Studies and PAC

Use of ^{111m}Cd on ZnO nano size crystals , studying structural aspects

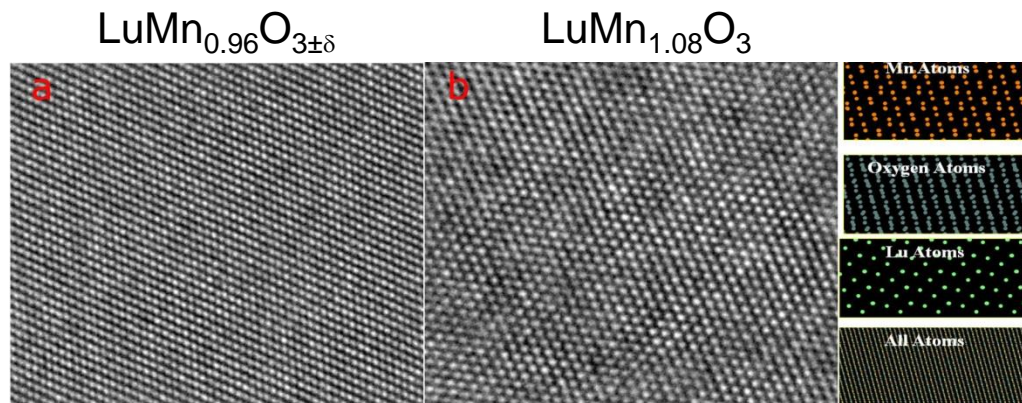
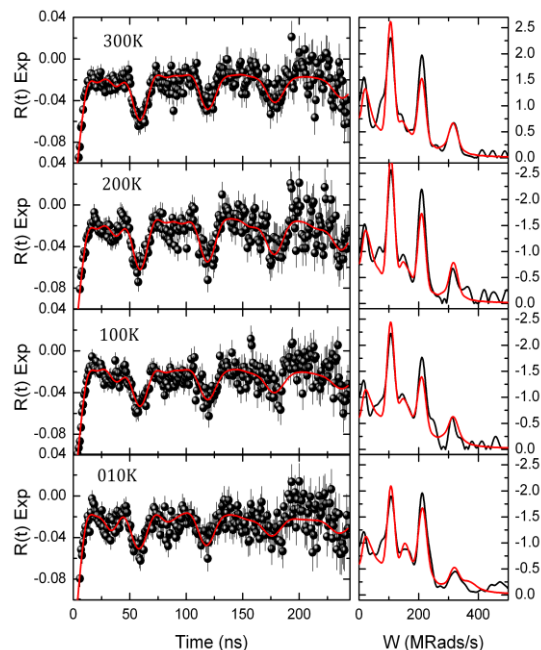


IS 544

Future projection : Growing ZnO nano-grains on folic acid template : biological applications , targeted drug delivery , use of $^{111m}\text{Cd}^{+2}$ again for the PAC studies. Growing GaO(OH) nano grains OR GaO_2O_3 grains and doping with Tl^{+3} or Ru^{+3} Or In^{+3} ions for PAC analysis.

Local PROBING of EFG and HMF in strongly correlated electron systems: spin, charge, orbital and lattice degrees of freedom

EFG and the electric polarization correlations . Multiferroic
EFG/HMF relations
Confined and strained conditions (films&nanop).



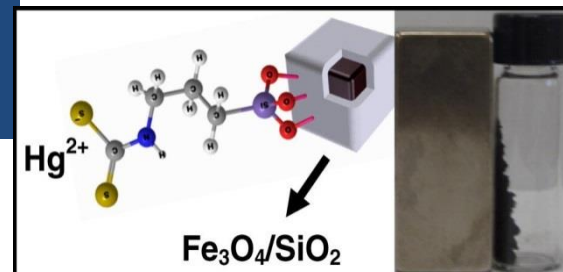
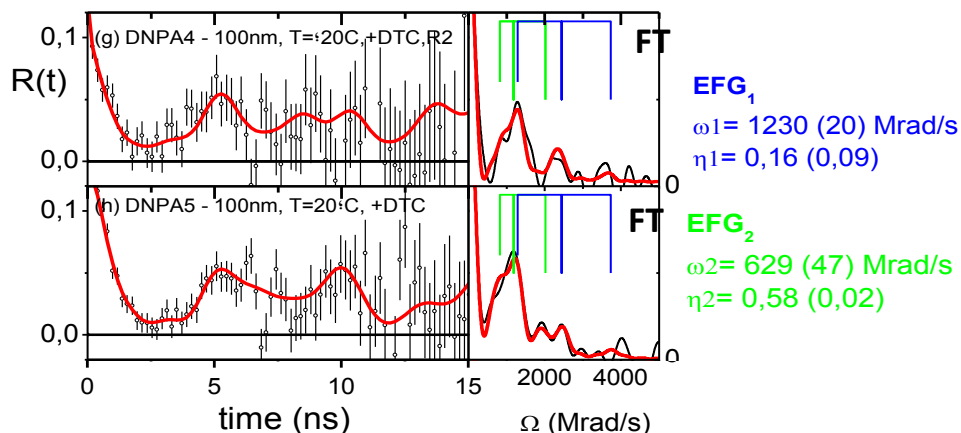
magnetic frustration reduction by local deformations in spin-ice systems DCr_2O_4 , (D=Mg, Cd, Zn, Hg)

hexagonal manganites:
single crystal (YMnO_3), magnetically frustrated LuMn_xO_3 .
Inequivalent positions for ions, in contrast to perovskite structure and clustering effects

11,5 Shifts isotopes: ^{111}mCd , ^{111}Ag , ^{117}Ag , ^{77}Br , $^{73}\text{Br}+^{73}\text{Se}$, $^{199\text{m}}\text{Hg}$

Local PROBING of EFG of local environment of ionic species (Hg^{2+} , Cd^{2+})

Two main electric field gradients in functionalized NP



Results in NP different from the pure Hg-DTC compound, where Hg is coordinated to 4 sulphurs instead of the expected 2 on the nanoparticle surface.

Limited number of experiments not yet significant statistics and systematic of results for control variables needed

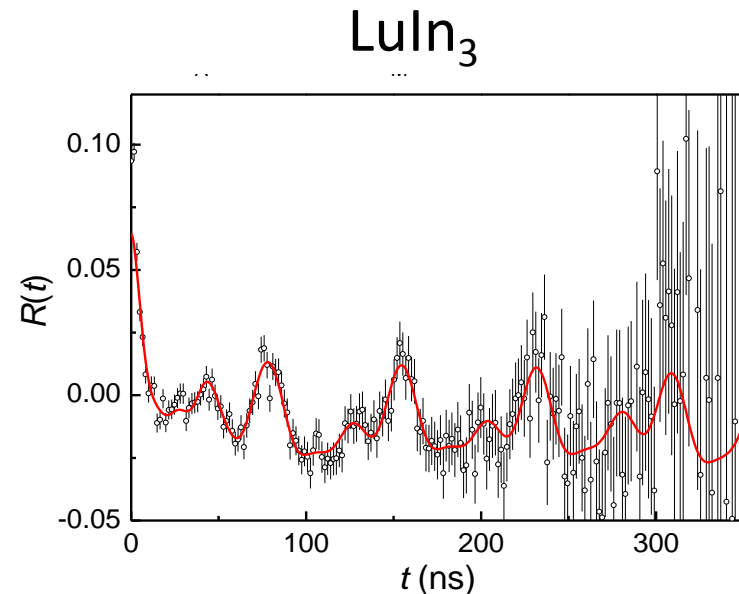
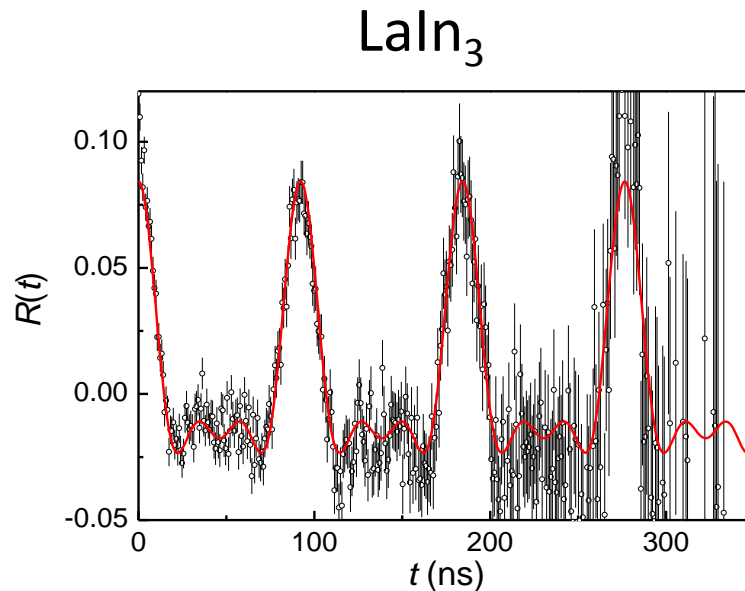
To complete the planned measurements program

- improvements on ion uptaking brought by different morphology/sizes, coverage and pH of the solutions, which determines the chemical speciation in the sample solution.
- study temperature dependence (0-30°C), for signal quality/molecular motion in adsorbed water remaining
- Modelling studies of the obtained EFG

^{111}mCd in $R\text{In}_3$ (diffusion aspects using PAC)

IS514

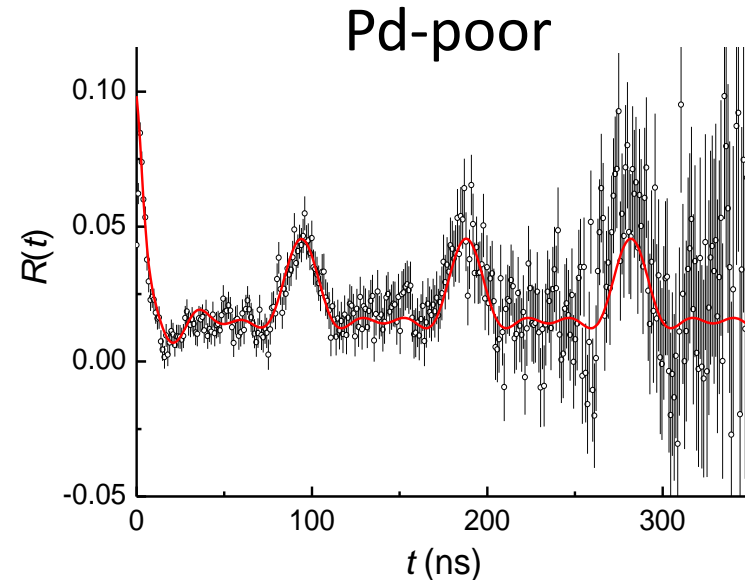
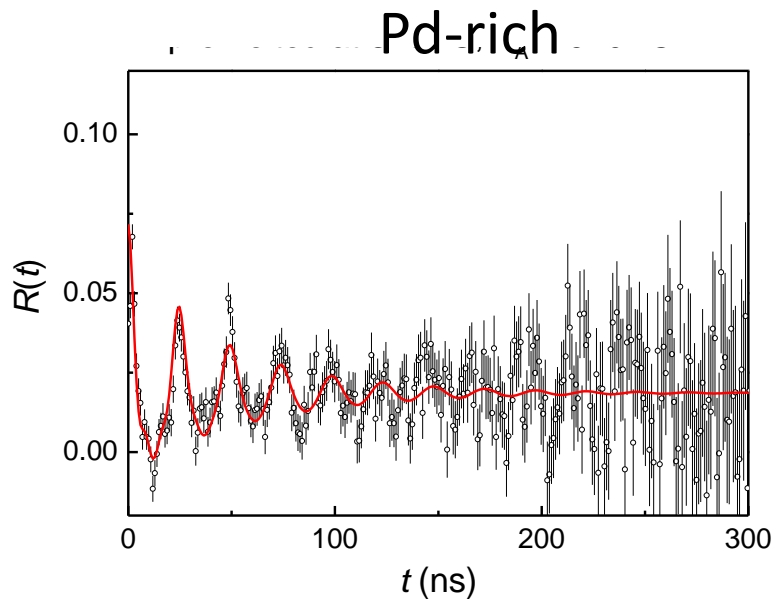
- Hope: observe defects bound to Cd impurity at room temperature
- Result:



Same signals as observed using ^{111}In PAC \rightarrow no bound defects observed, \rightarrow binding energy too low(?)

^{111}mCd PAC in Pd_3Ga_7

- Goal: measure quadrupole relaxation at elevated temperature to complement earlier ^{111}In measurements
- Result of room temperature test measurements:

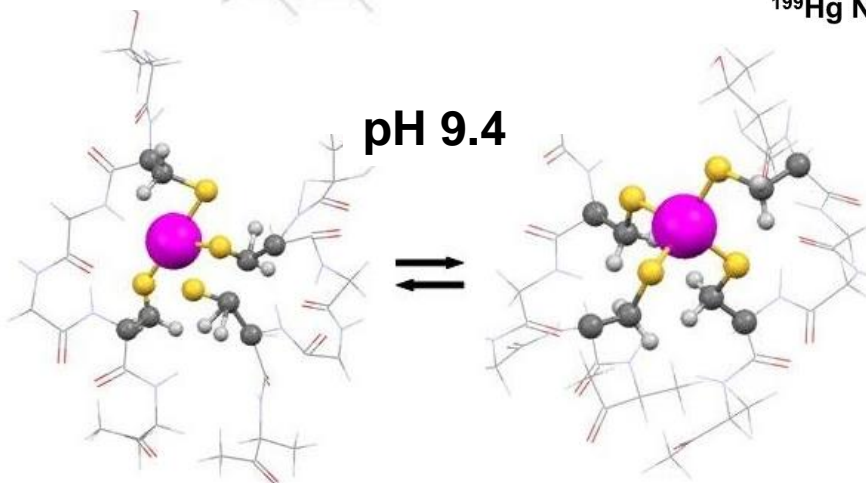
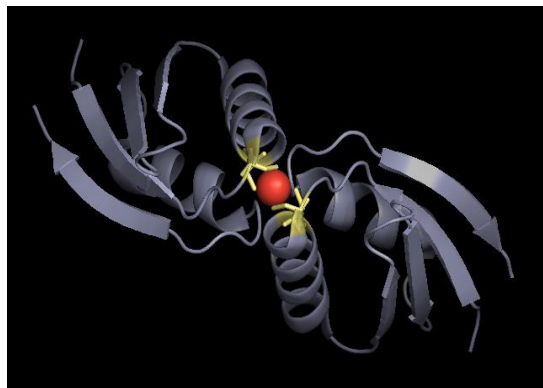
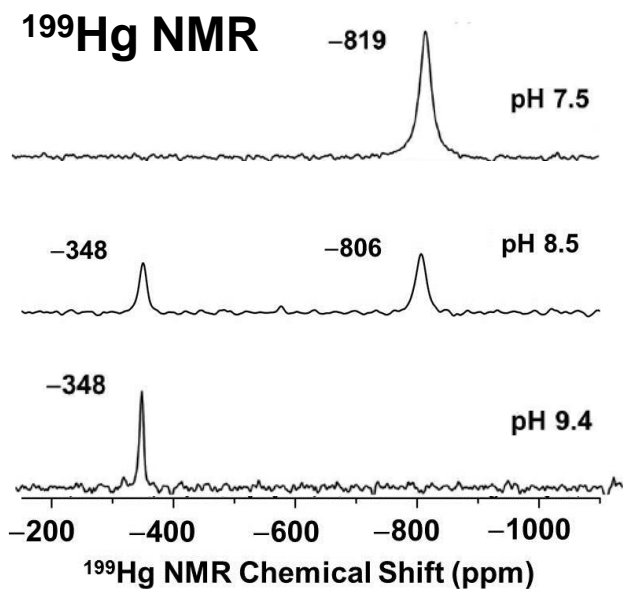
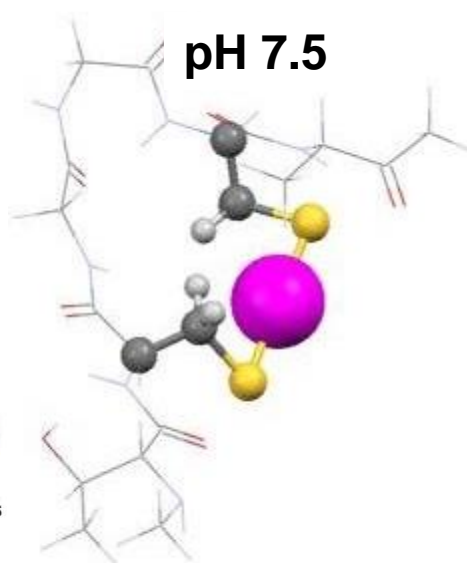
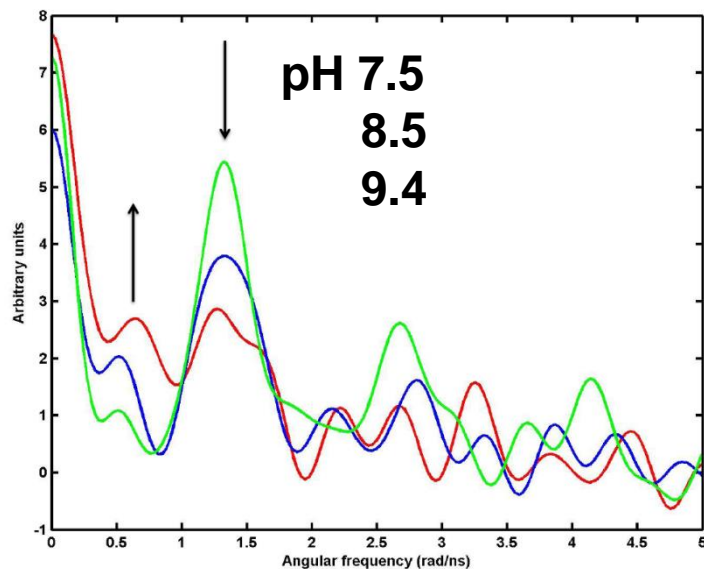


Signals differ from ^{111}In PAC results \rightarrow improper sample preparation prior to implantation at ISOLDE \rightarrow try again!

Metal ion transfer between proteins:

The Cu(I) binding protein HAH1

^{199}mHg PAC



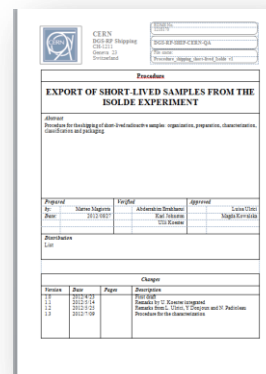
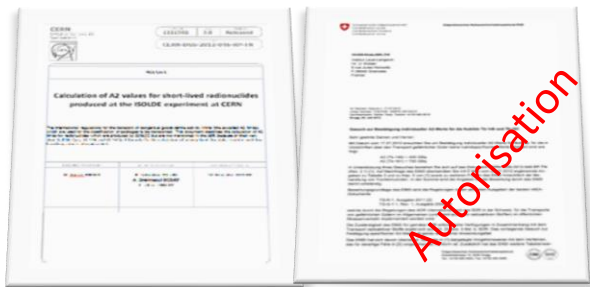
Luczkowski *et al.*,
Chem. Eur. J., 2013, 19, 9042

Spécificités : transports vie courte ISOLDE

Objectif 1 : Faire les transports Type A le plus rapidement possible

Valeurs A2 avec
U. Koester et K. Johnston

Procédure spéciale
En collaboration avec M. Magistris

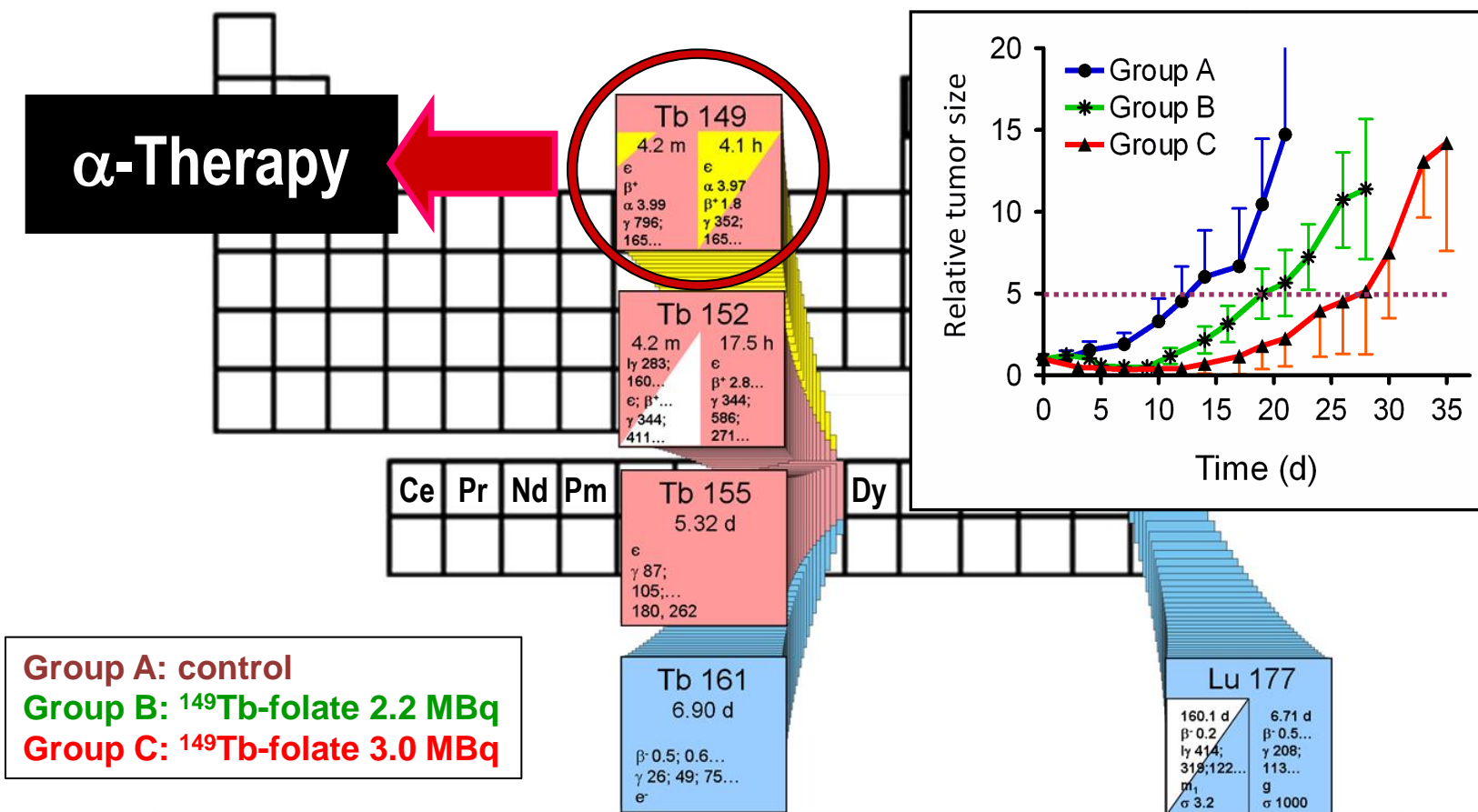


Tb 149 4.2 m c p ⁺ γ 796; 165...	4.1 h c α 3.97 p ⁺ 1.6 γ 352; 165...	Tb 152 4.2 m c p ⁺ γ 796; 165...	17.5 h c p ⁺ 2.8... α 3.97 γ 344; 271...	Tb 155 4.2 m c p ⁺ 2.8... γ 87; 105; 180, 262	5.32 d c p ⁺ 2.8... γ 87; 105; 180, 262	Tb 161 6.90 d p ⁺ 0.5, 0.6... γ 26, 49, 75... e
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Slide thanks to Abderrahim Errahhaoui

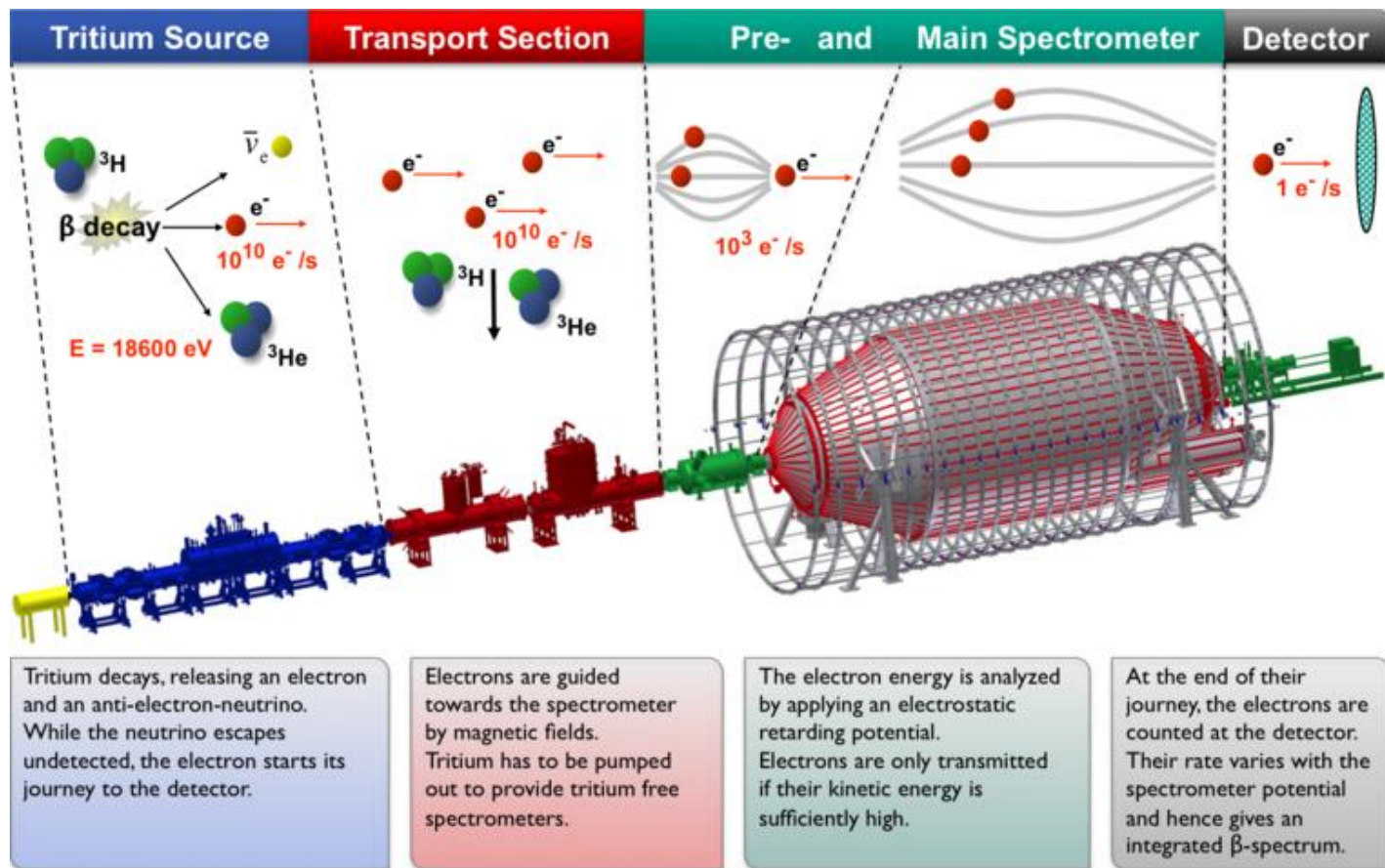
Terbium-149: alpha-Emitter

Folate Receptor Targeted α -Radionuclide Therapy



Müller et al. 2012, J Nucl Med 53:1951.

Müller et al. 2013, Pharmaceuticals, submitted



- For monitoring of KATRIN conversion electrons needed from decay of ^{83}mKr ($T_{1/2} = 1.8 \text{ h}$)
- ^{83}mKr is generated by decay of ^{83}Rb ($T_{1/2} = 86 \text{ d}$), which is implanted into a solid platinum substrate ($\varnothing 12 \text{ mm}$)
- ISOLDE produced 11 sources of ^{83}Rb
- Bonn can now produce more reliable sources.

(IS533 similar principle)

Isotope	yield (/uC)	target – ion source	Shifts (8h)
IS432/IS450			
⁵² Mn	5.10 ⁷	Nb foil RILIS	2
⁵⁶ Co	10 ⁶	YO/ZrO (VADIS)	3
IS481			
^{111m} Cd	10 ⁸	Molten Sn / plasma	4
¹¹⁵ Ag/ ¹¹⁵ Cd / ¹¹⁵ In	10 ⁸	UC2, Ag RILIS with Nb or Ta ion source cavity to decrease In contamination	0.5
¹¹⁷ Ag/ ¹¹⁷ Cd / ¹¹⁷ In	10 ⁸	UC2, Ag RILIS with Nb or Ta ion source cavity to decrease In contamination	3.5
IS487			
^{111m} Cd	10 ⁸	Molten Sn / plasma	7
^{199m} Hg	10 ⁸	Pb / Plasma, run it at LOW temp, avoiding TI	1
⁷⁷ Br, ⁷³ Br+ ⁷³ Se	10 ⁸	ZrO / plasma	1
IS488			
^{204m} Pb	1e8	UCx – RILIS	2
^{199m} Hg	1e8	Pb – VADIS	2
IS489			
²⁹ Al	10 ⁸	UC ₂ – W surface	1
³⁸ Cl	10 ⁸	UC ₂ – W surface	1
¹¹¹ Ag ¹¹⁷ Ag/ ¹¹⁷ Cd	10 ⁸	UC ₂ - LIS	1
IS492			
⁷⁷ Br	10 ⁸	ZrO ₂ –HP	1
¹¹¹ Ag ¹¹⁷ Ag/ ¹¹⁷ Cd	10 ⁸	UC ₂ - LIS	2
^{204m} Pb	1e8	UCx – RILIS	2

IS432/450

No recent results due to various technical problems : still relevant physics; RILIS Co scheme available...

IS481

Further work on Ga₂O₃. γ-γ, e-γ and β-γ studies ... allows for unique information.

IS487

Continue work on magnetic frustration, links between EFG and polarisation , investigate multiferroic ACrO₃ compounds...

IS488

Pb preferred,; **challenging**. Test with synthetic protein.

IS489

Further commissioning of online chamber. Role of metallic contacts.

IS492

Further studies on Br in ZnO. Examine in more detail recently discovered Sn feature.

IS500

⁸³ Rb-83	500pA/ 1.6μA	Uc-Ta	1
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IS500

Contingency shift in case problems at Bonn.

IS501

⁵⁷ Mn	(2- 3)×10 ⁸	UCx, Mn RILIS	2
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¹¹⁹ In	(2- 3)×10 ⁸	UCx, In RILIS	1
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IS501

Co available with RILIS...
Complete programme on oxides, location of In in oxides.

⁵⁷ Co	9×10 ⁷	ZrO ₂ /YtO ₂ , V/R(?)	1
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IS514

^{111m} Cd	2×10 ⁸	Sn – HP (VADIS)	8
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IS514

Previous results failed due to problems with sample preparation. Now resolved, experiments to be repeated.

IS515

^{111m} Cd	10 ⁸	Molten Sn / plasma	2
--------------------	-----------------	-----------------------	---

^{199m} Hg	10 ⁸	Pb / Plasma,	7
--------------------	-----------------	--------------	---

IS515

Understanding Hg-coordination, temperature dependence.

IS533

⁸³ Rb		UC-W / UC-Ta	6
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IS533

No previous shifts taken due to problems at ISOLDE. Still relevant

IS544

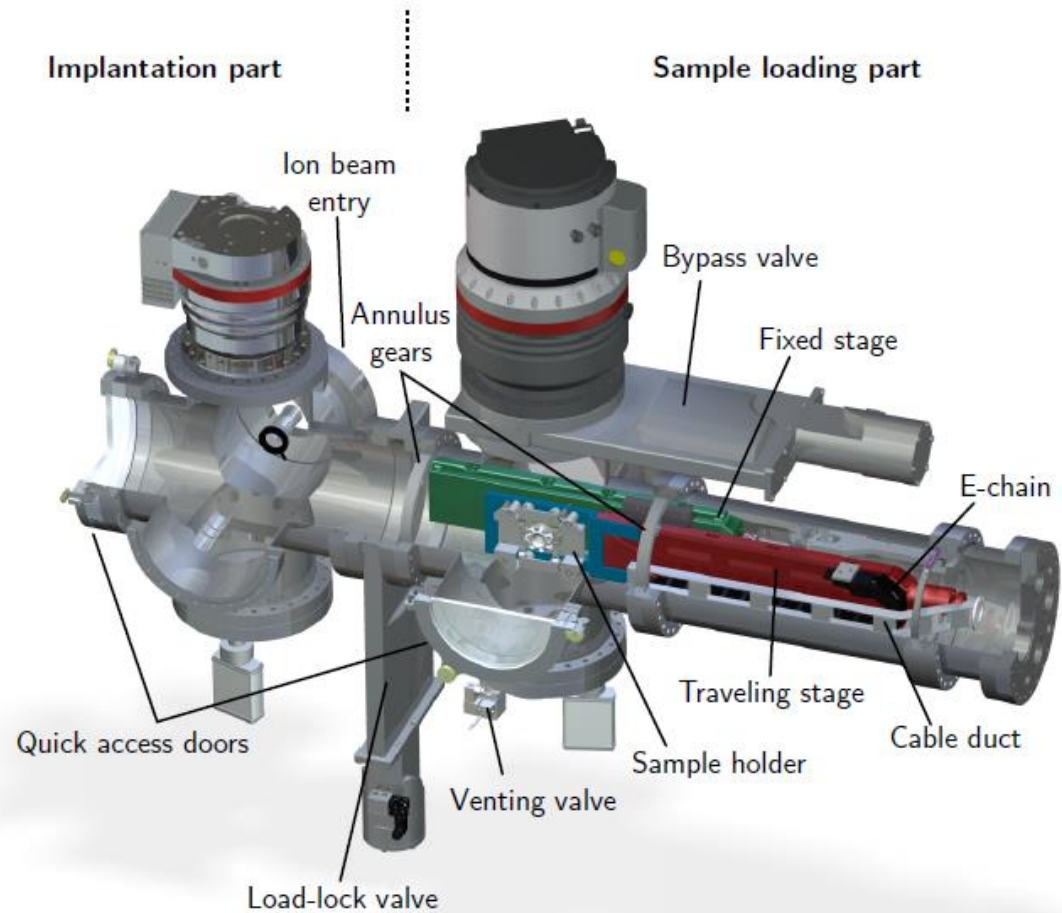
^{111m} Cd	5e8 μC	Molten Sn / plasma	2
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IS544

Further investigations on folic acid and cyclo

IS486: Addendum

- Request for extra shifts for commissioning the new general implantation chamber (due Summer 2014)
- Also: 2 shifts for development of online PAC using ^{19}O



isotope	yield (/uC)	target – ion source	Shifts (8h)
172-Lu	5e7	Ta or other (RILIS if possible)	1
149-Gd	5e7	Ta or other (RILIS if possible)	1
19-O	1e6	UCx VADIS	2

Summary

- 65 shifts to be kept for 13 experiments (of which 1 is for contingency only)
- 4 Shifts requested for IS486 to help complete their programme and commission the new GLM 2.0 chamber.

Publications & Theses

- 89 Publications published or accepted.
- 32 Theses (PhD, Masters either completed or about to be...)

Many thanks to collaborators for the slides

