Emission Mössbauer Spectroscopy at ISOLDE/CERN

Torben Esmann Mølholt

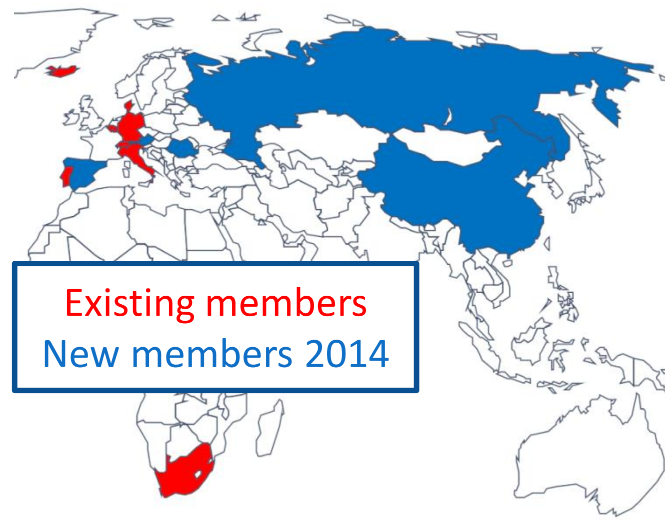
ISOLDE Seminar, 25. Nov. 2015

ISOLDE Solid State Physics

Outline

- Experimental setup at ISOLDE
- Brief on the Mössbauer spectroscopy technique
- Examples and Results
- Future/ongoing measurements

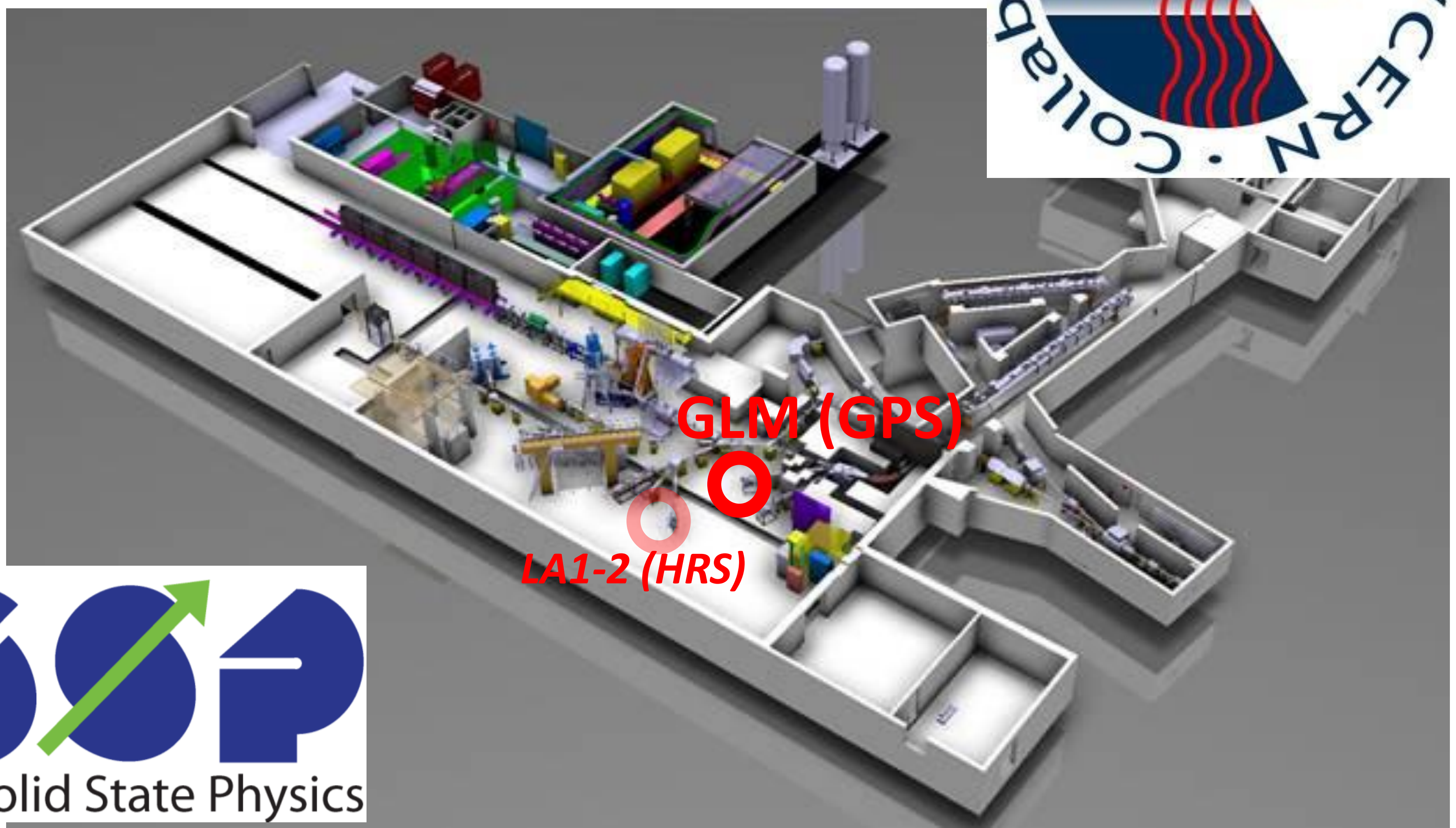
Acknowledgements



The Mössbauer collaboration at ISOLDE/CERN, **>30 active members** with new members (2014) from China, Russia, Bulgaria, Austria, Spain: Four experiments (IS-501, IS-576, IS-578, I-161)

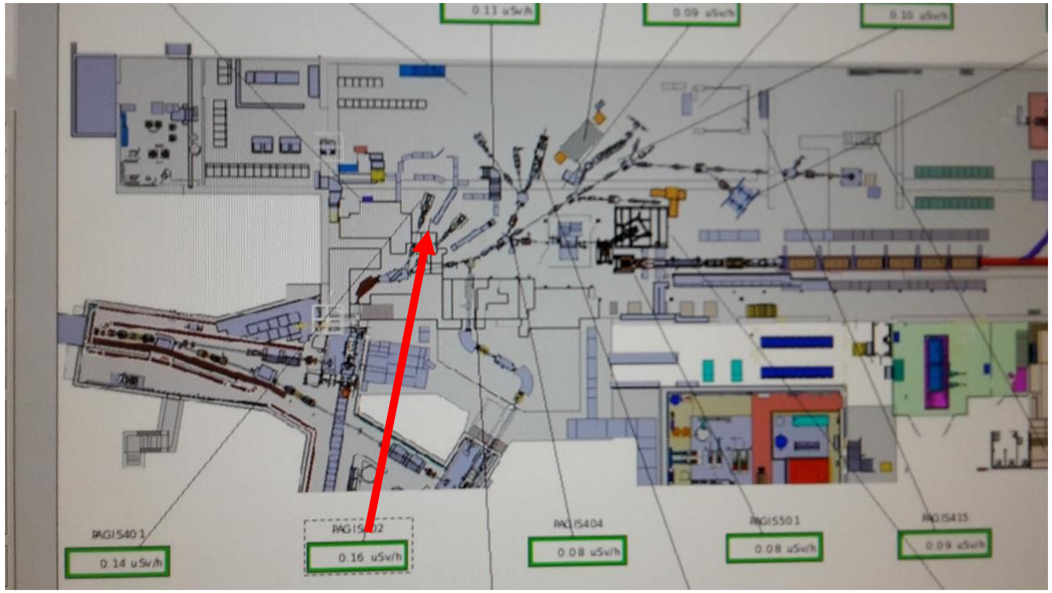
Emission Mössbauer Spectroscopy at ISOLDE/CERN

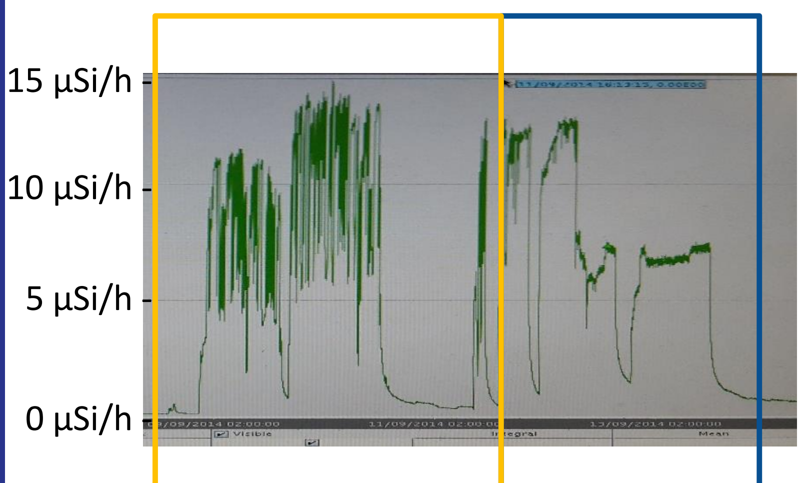
http://e-ms.web.cern.ch/

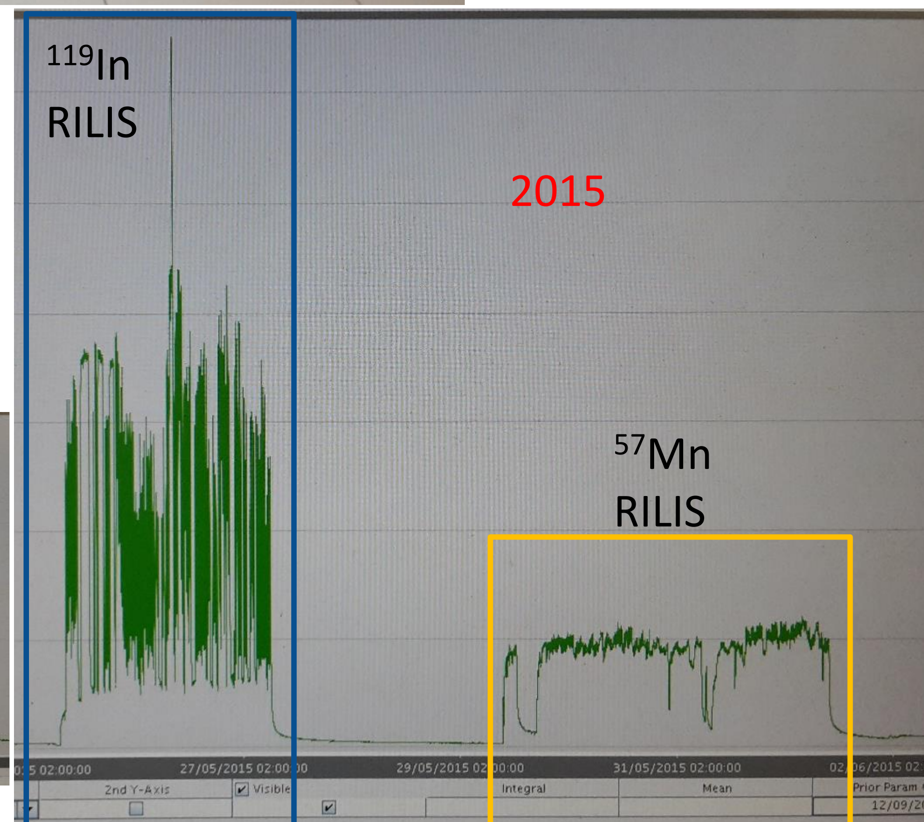


Mössbauer Collaboration at ISOLDE/CERN

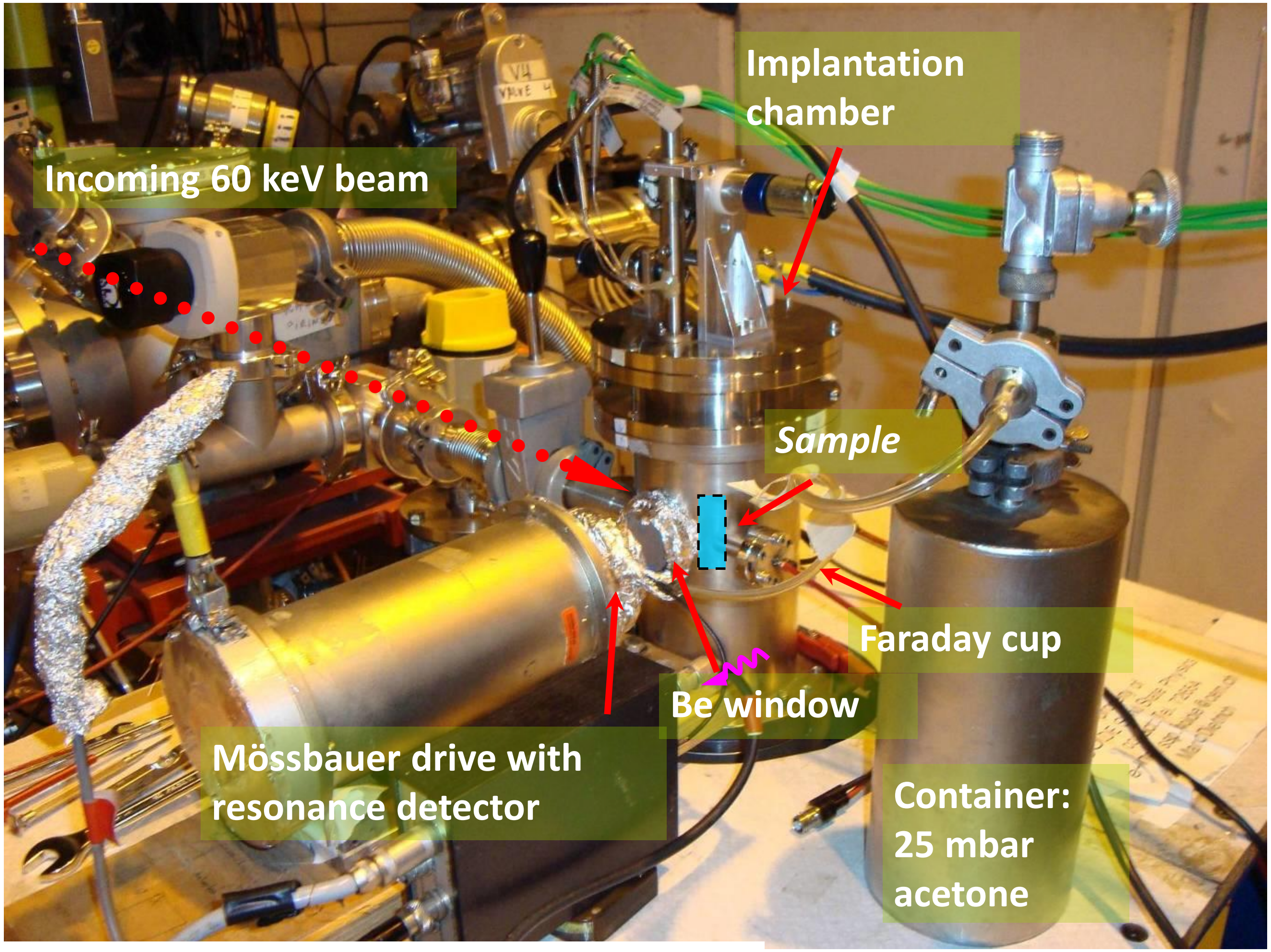
ISOLDE Solid State Physics

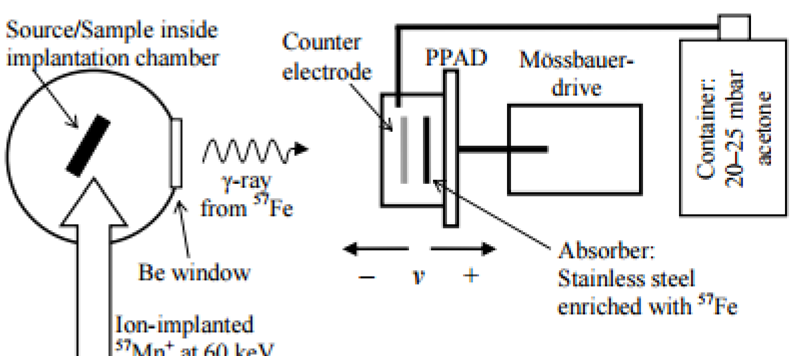

0.11 uSv/h
0.09 uSv/h
0.10 uSv/h
PAGIS401
0.14 uSv/h
0.16 uSv/h
PAGIS404
0.08 uSv/h
PAGIS501
0.08 uSv/h
PAGIS415
0.09 uSv/h


2014
^{57}Mn
RILIS
^{119}In
15 µSi/h
10 µSi/h
5 µSi/h
0 µSi/h


^{119}In
RILIS
2015
^{57}Mn
RILIS
27/05/2015 02:00:00
29/05/2015 02:00:00
31/05/2015 02:00:00
2nd Y-Axis
Visible
Integral
Mean

Mössbauer Experimental setup



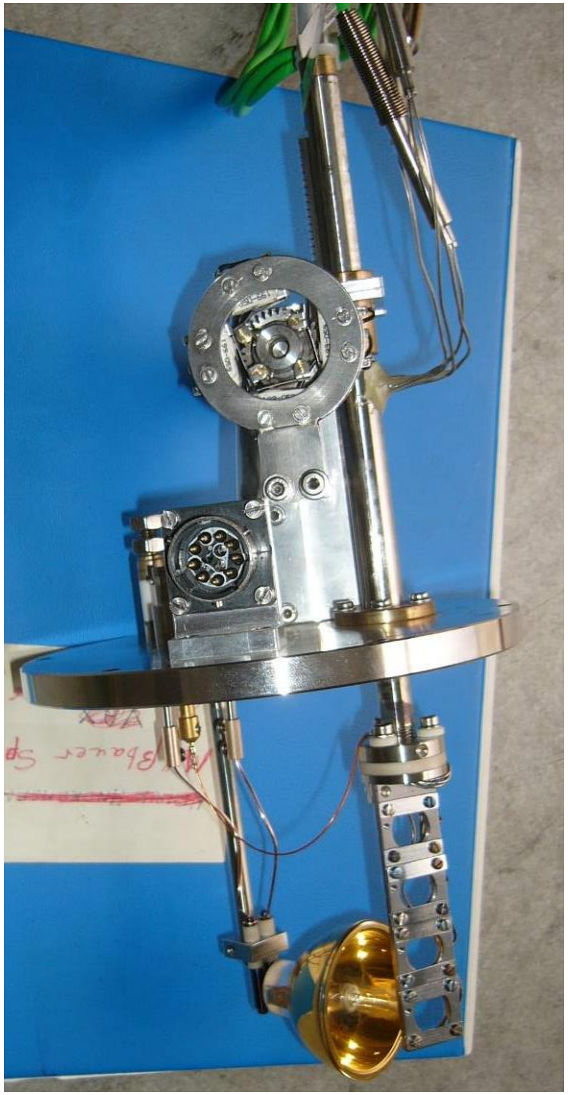


- Intensity (~1×10^{8} atoms/s)
- High statistics spectrum (5 – 10 min.)
  - On-line (short lived)
- Collections for Off-line (long lived)
  - Hours - days

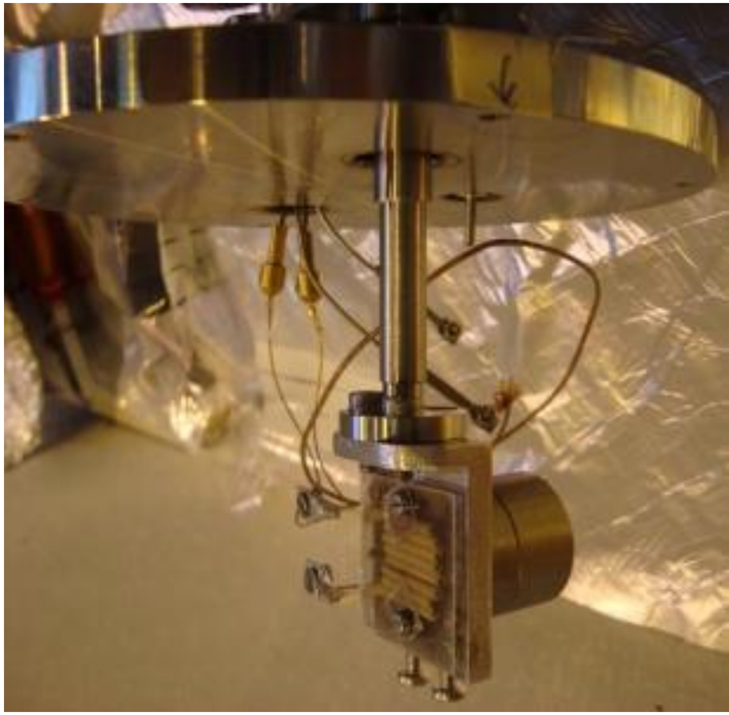
Mössbauer Experimental setup

Sample holder

- Temperature range 90 – 700 K



- Measurements at different emission angles
- Applied magnetic field ($B_{ext} \leq 0.6$ T)



Mössbauer Experimental setup

Sample holder

- Quenching:
  - ➢ Implant at high temperature
  - ➢ Measure at low temperature (off-line)

Mössbauer Experimental setup

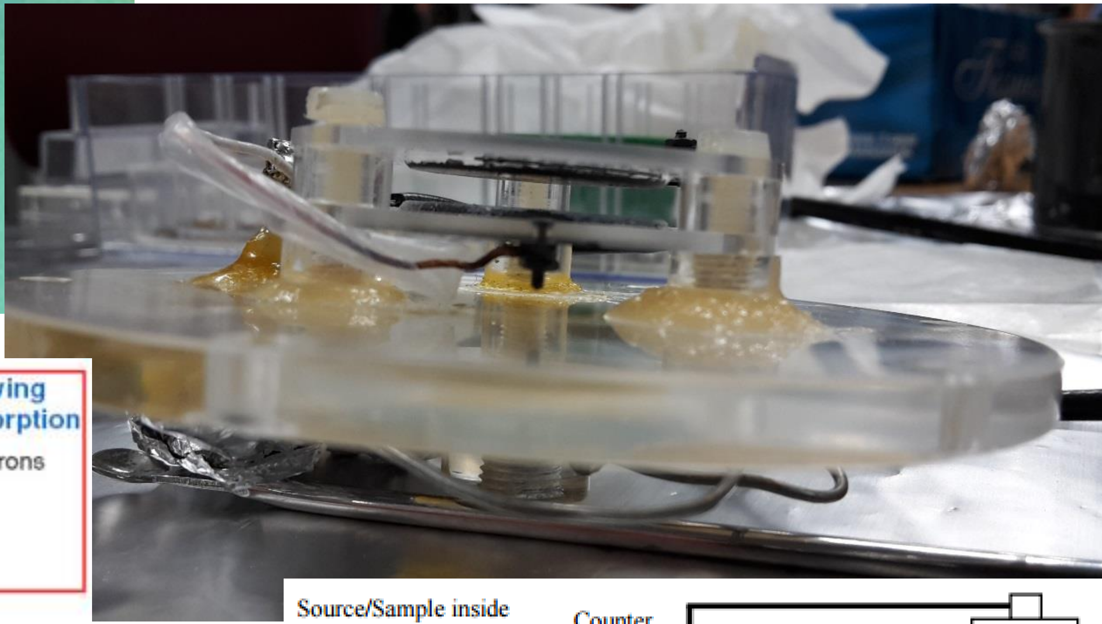
Resonance detector

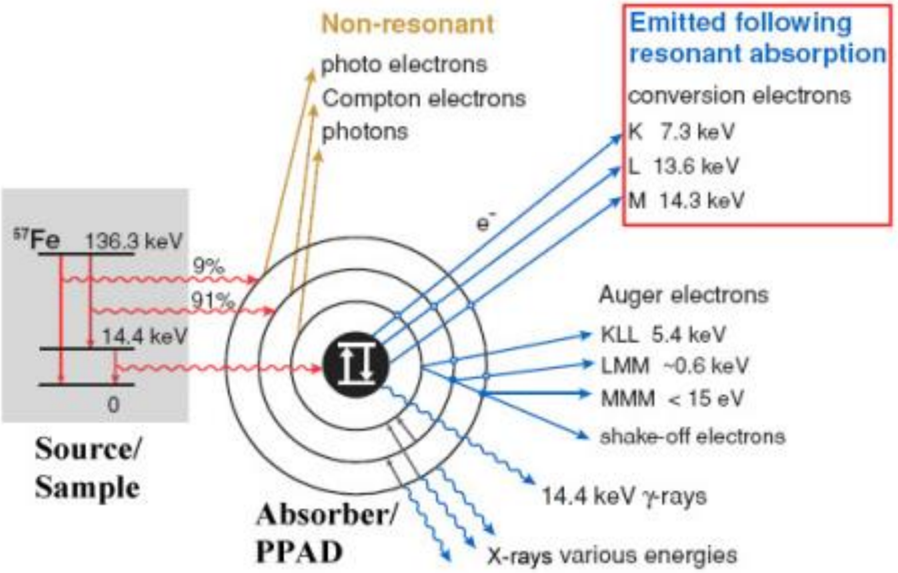
- G. Weyer, Mössbauer Eff. Meth., 10 (1976) 301

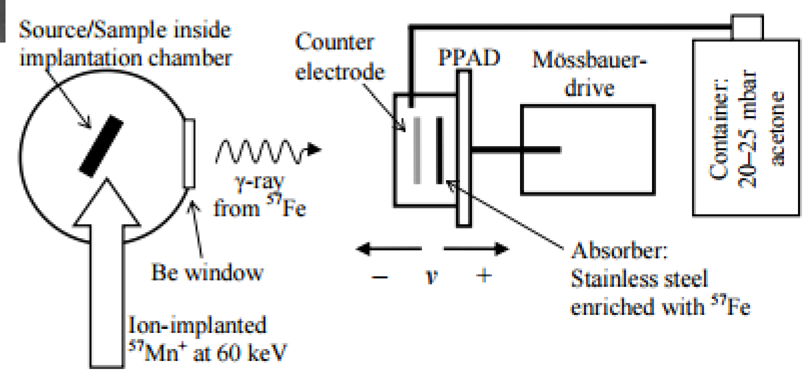
PPAD: Parallel Plate Avalanche Detector

- Single line resonance detector.

0.1 cps (~0.1 µCi) – 50k cps (~500 mCi)



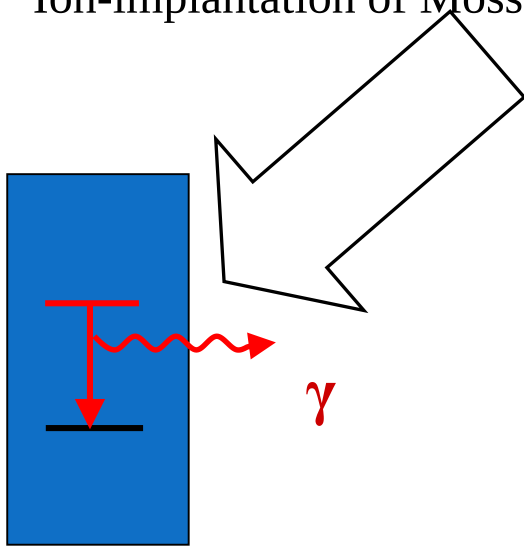




Mössbauer spectroscopy technique

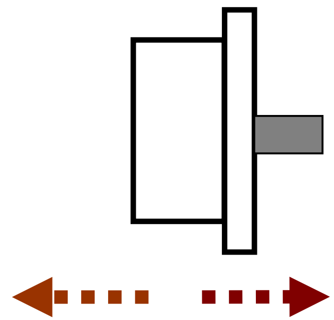
Emission Mössbauer spectroscopy

40-60 keV
Ion-implantation of Mössbauer Probe



Source/sample:
ion-implanted crystal

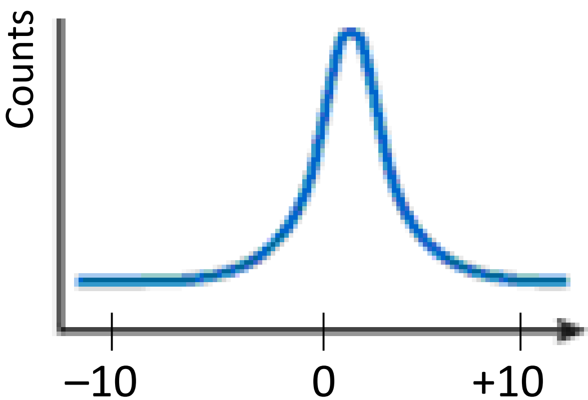
Measurement of spectrum



$$E(v) = E_{\gamma}\left(1 + \frac{v}{c}\right)$$

Absorber/detector:
Single line resonance detector

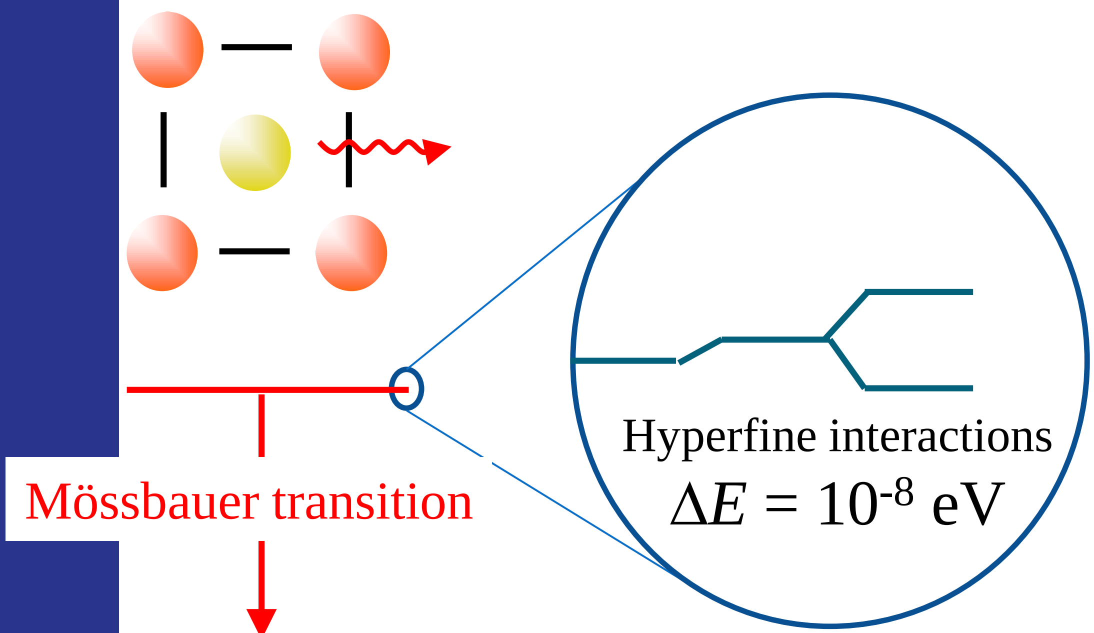
Mössbauer spectroscopy:
High spectral resolution
$v = \pm 10$ mm/s (Doppler)
$\Delta E = \pm 4.8\times 10^{-7}$ eV



Emission Mössbauer spectrum

Emission Mössbauer spectroscopy

Measure hyperfine interactions



Dilute Probe:

Below 10^{-3} at.%

1×10^{18} atoms/cm^3

<u>Important info on an atomic scale:</u>

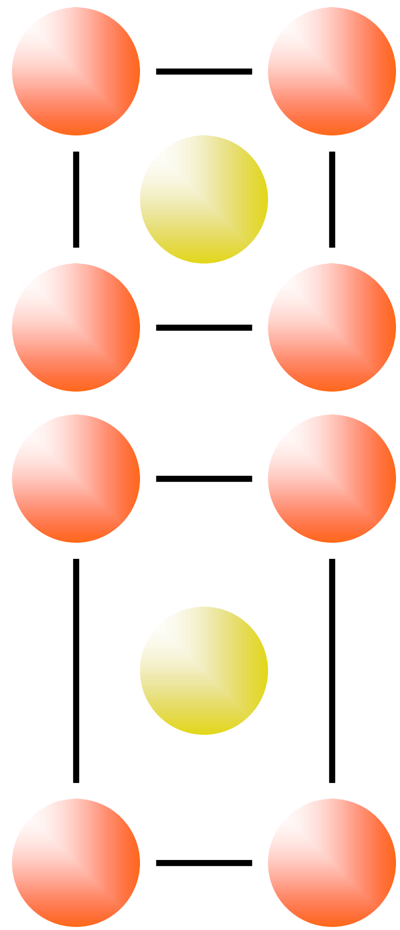
- Valence/Spin state (line position, δ)
- Site symmetry (doublet?)
- Magnetic interactions (Sextet)
- Binding properties
- Relaxation effects
- Diffusion

.....

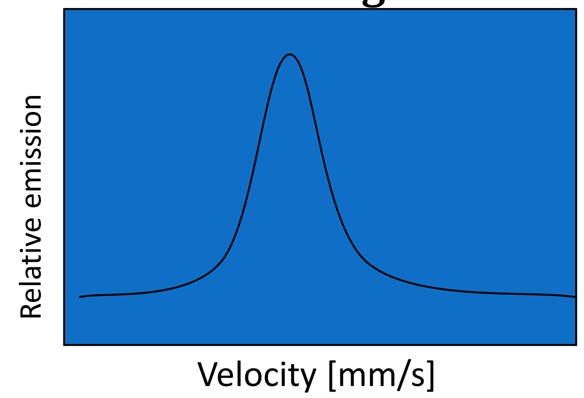
The resolution of Mössbauer spectroscopy can measure **hyperfine interactions**

- **Position of spectral line**
  → Valence state

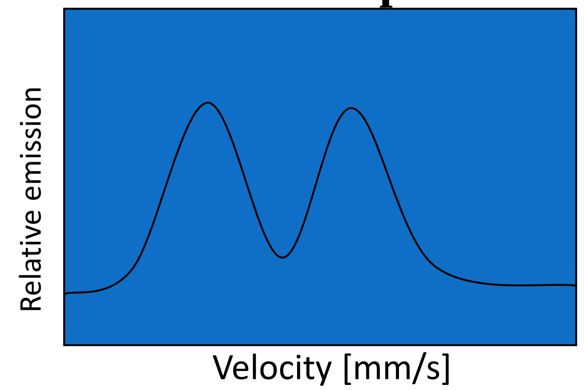
- **Quadropol splitting**
  → Cubic?



Cubic: Single line



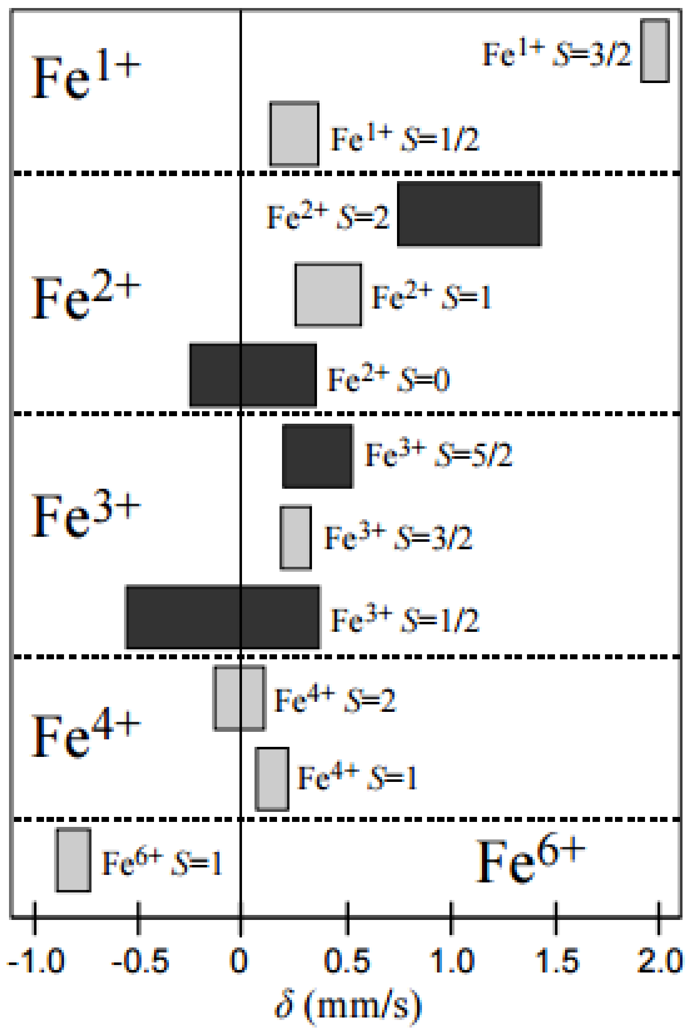
Non-cubic: Split line



Valence/Spin state

^{57}Fe emission Mössbauer spectroscopy

Spectral line position,
Isomer shift, δ

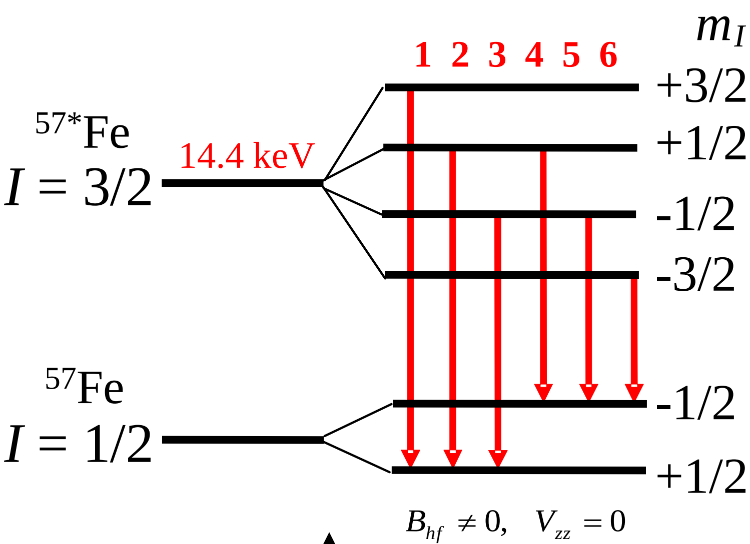


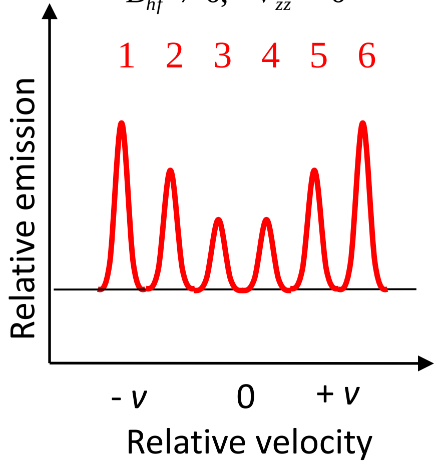
Shielding ↑
$\rho(0)$ ↓, δ ↑

1 mm/s =
48 neV

^{57}Fe emission Mössbauer spectroscopy

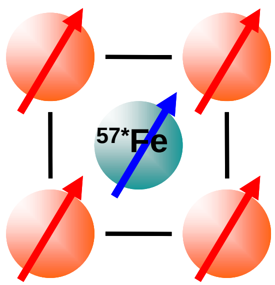
Magnetic hf. splitting of ^{57}Fe → Sextet



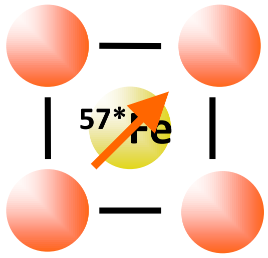


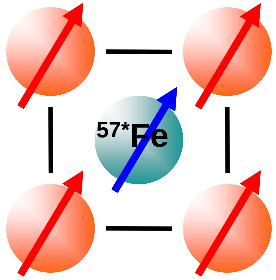
If the spin is stable for longer than **140 ns** – Sextet is observed

→Ferromagnetic material



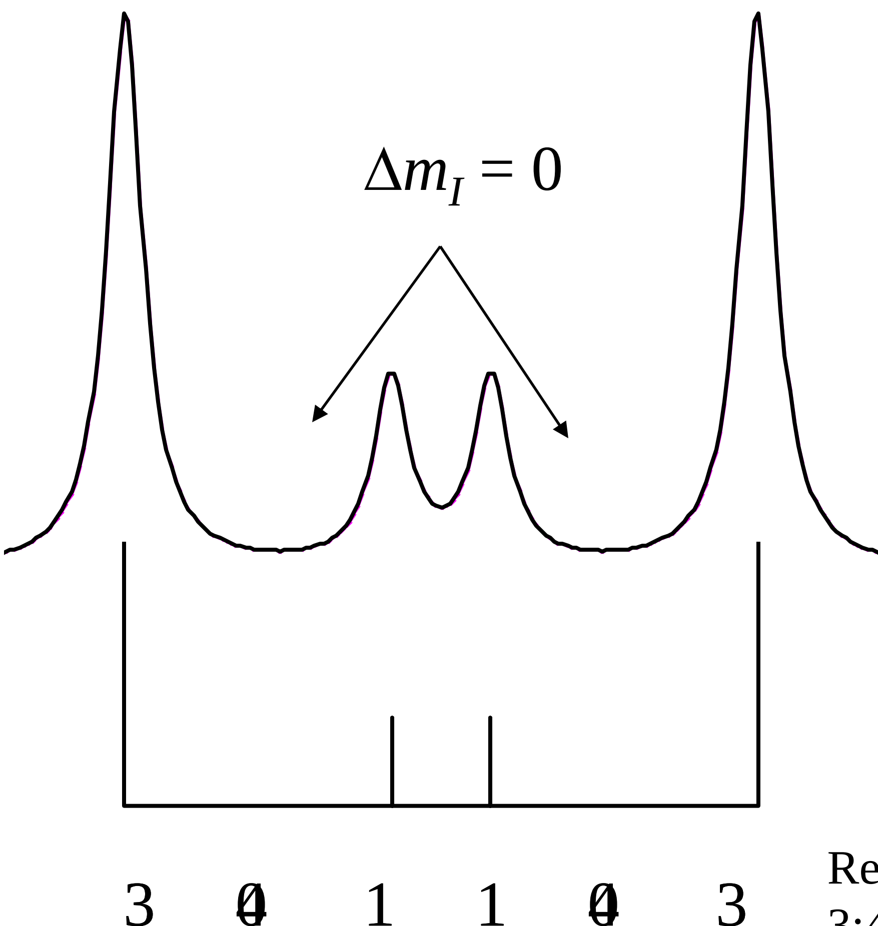
→ Slow relaxing paramagnetism (not only one sextet)

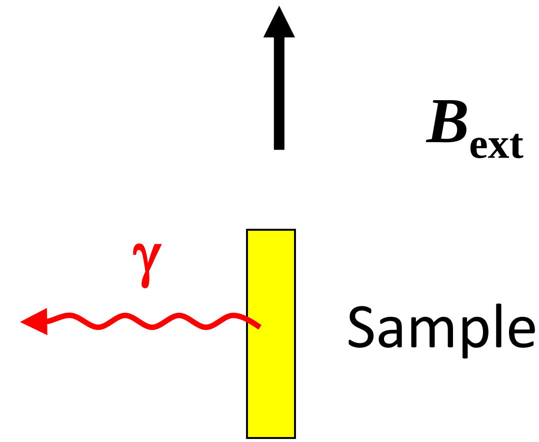




Magnetic order

Angular dependence in B_{ext}





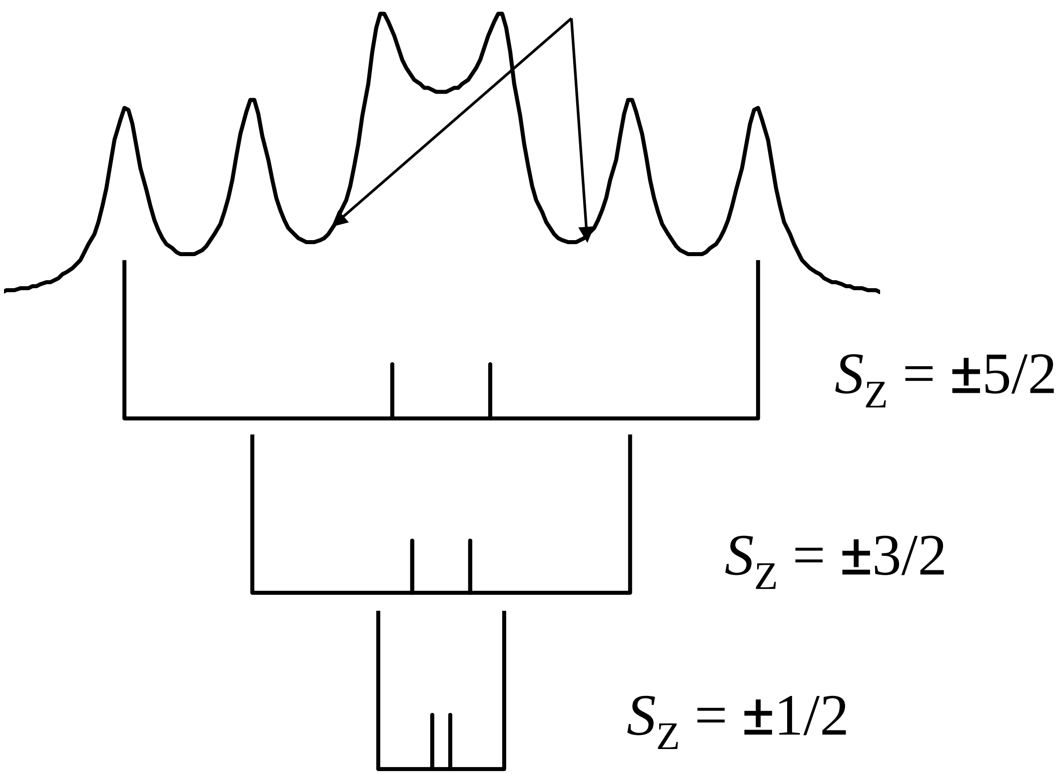
Individual line ratios depend on the angle between B_{ext} and the γ direction

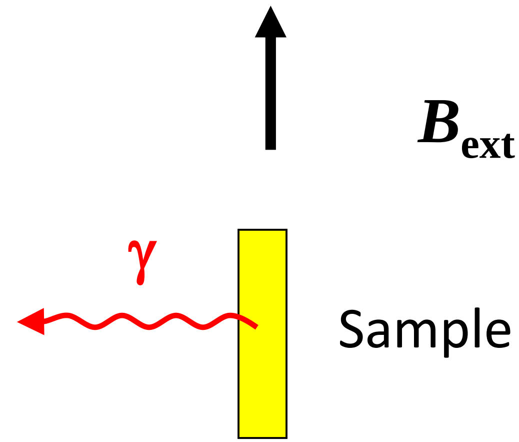
Relative line ratios:
3:4:1 (90°) → 3:0:1 (0°)

Angular dependence in B_{ext}
(same as ordered, but Kramer doublets)

Paramagnetism (slow relaxation)

$\Delta m_I = 0$ from $S_Z = \pm 3/2$

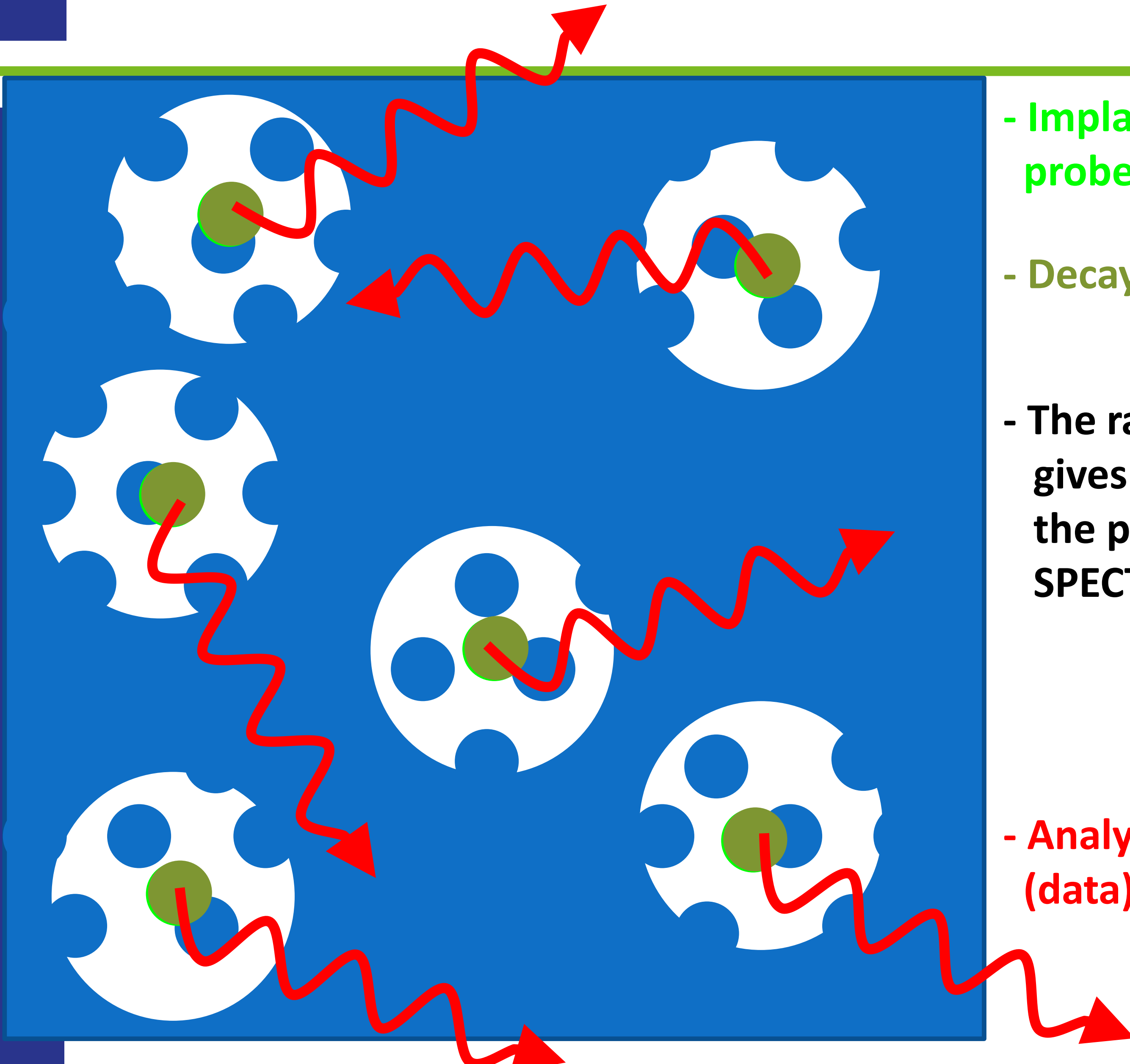




Individual line ratios depend on the angle between B_{ext} and the γ direction

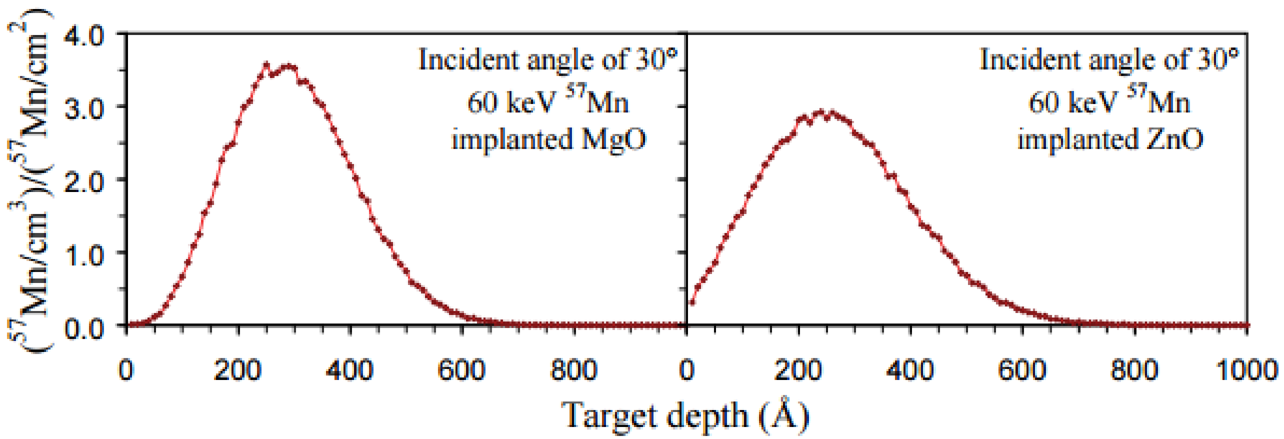
Relative line ratios:
3:4:1 (90°) → 3:0:1 (0°)

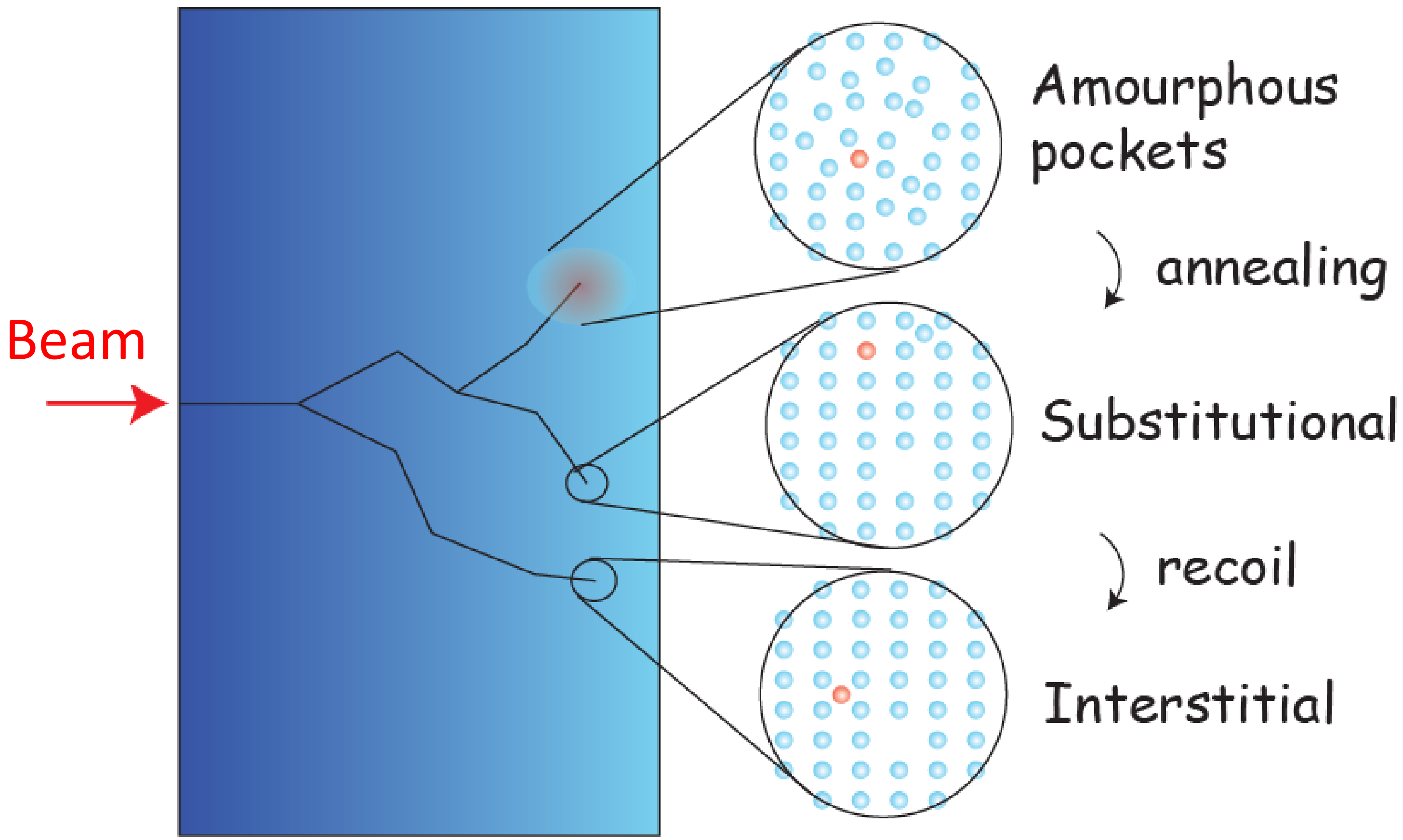
Sample of interest (Crystal, solid)



- Implant Radioactive probes / impurities
- Decay → Probe the crystal
- The radioactive decay gives information about the probe sites → SPECTRUM (data)
- Analysis of Spectra (data) → Crystal properties

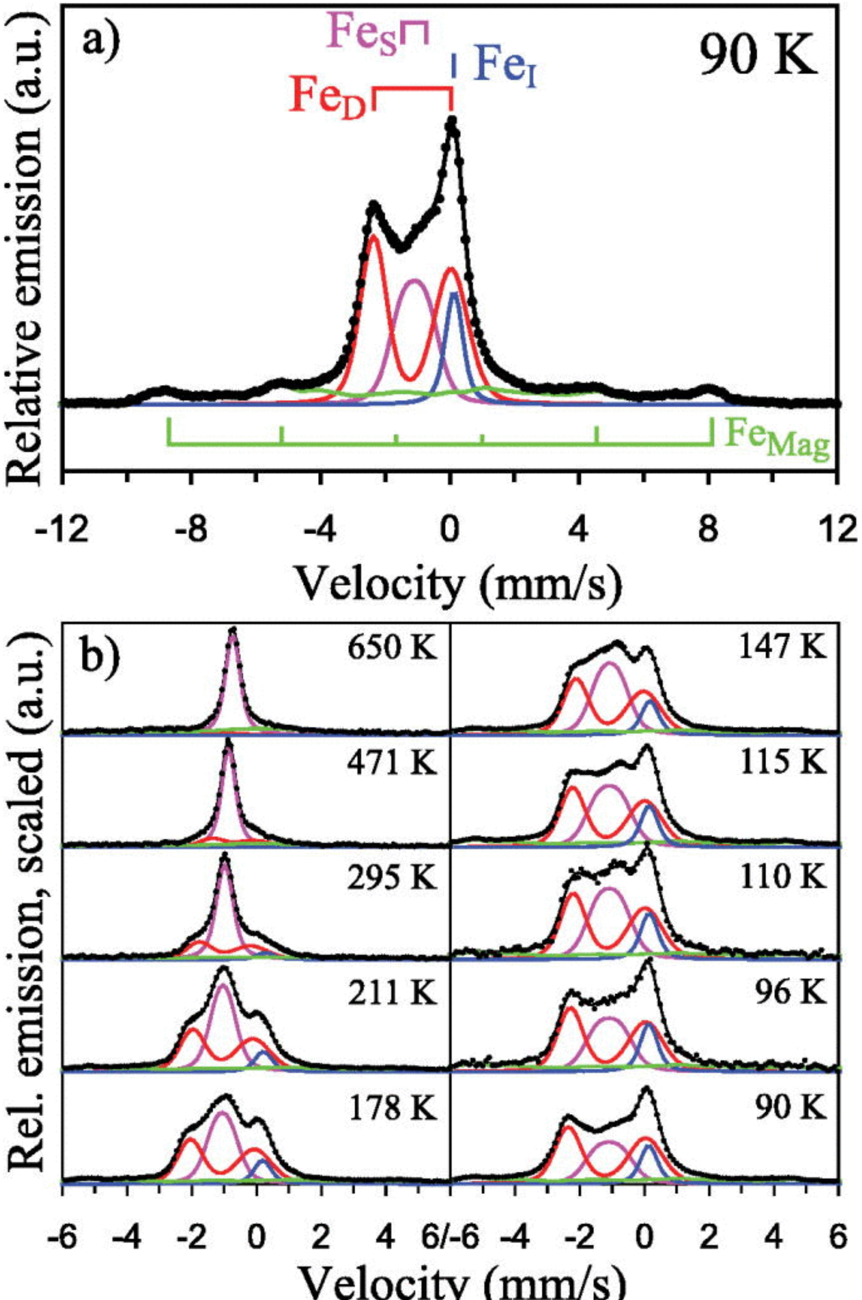
Ion-implantation

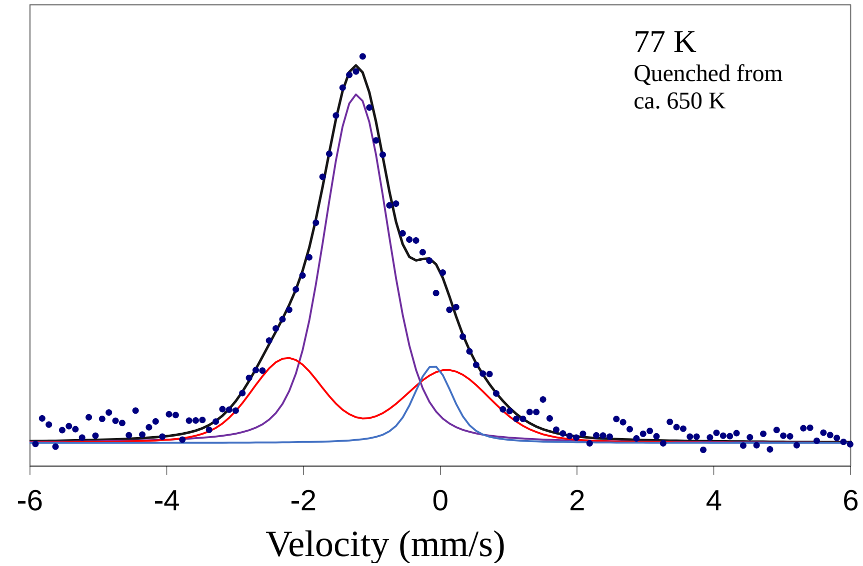




Examples and results

Interstitial in MgO





Quenching setup:

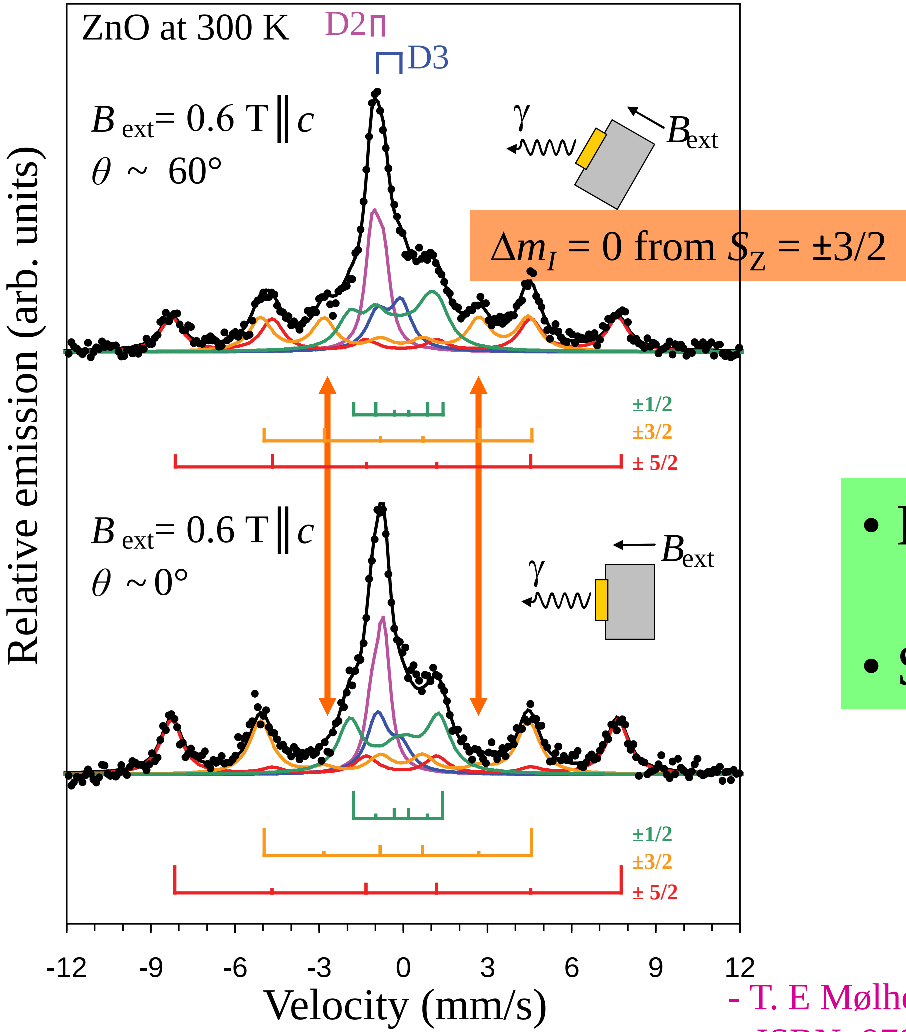
Reduction of Fe_D (damage)

- "More clear" Fe_I line

- *Low statistics spectrum (no Fe_{Mag})*

T. E. Mølholt *et al.* J. Appl. Phys. **115**, 023508 (2014)

Magnetic identification

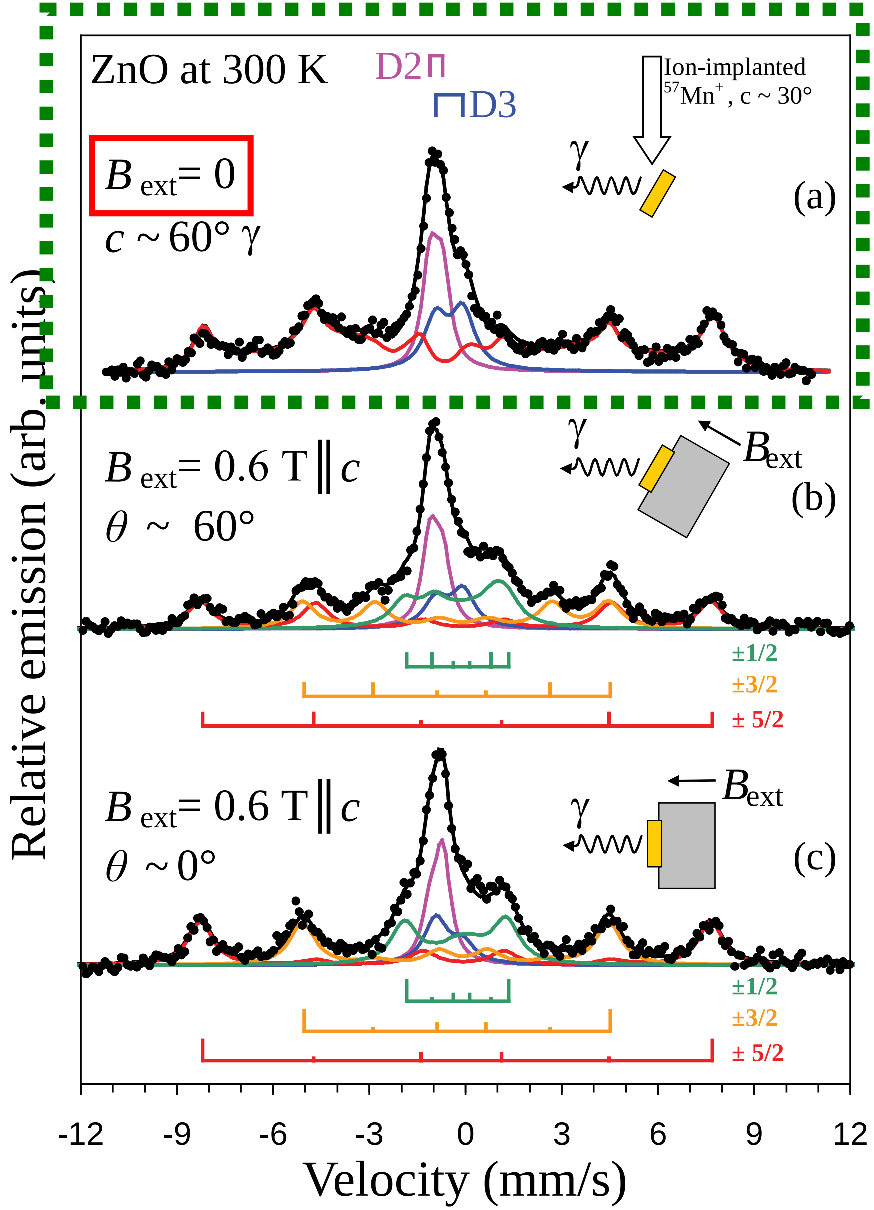


Magnetic structure originate from Kramers doublets is clearly observed.

- NO ordered magnetism
- Slow relaxing Paramagnetism

- T. E Mølholt, Paramagnetism in ion-implanted oxides (2012) ISBN: 978-9935-9069-5-3
- H. P. Gunnlaugsson *et al.* , Appl. Phys. Lett. 97 (2010) 142501

Paramagnetic relaxation of dilute ^{57}Fe?



Know it is of paramagnetic origin:

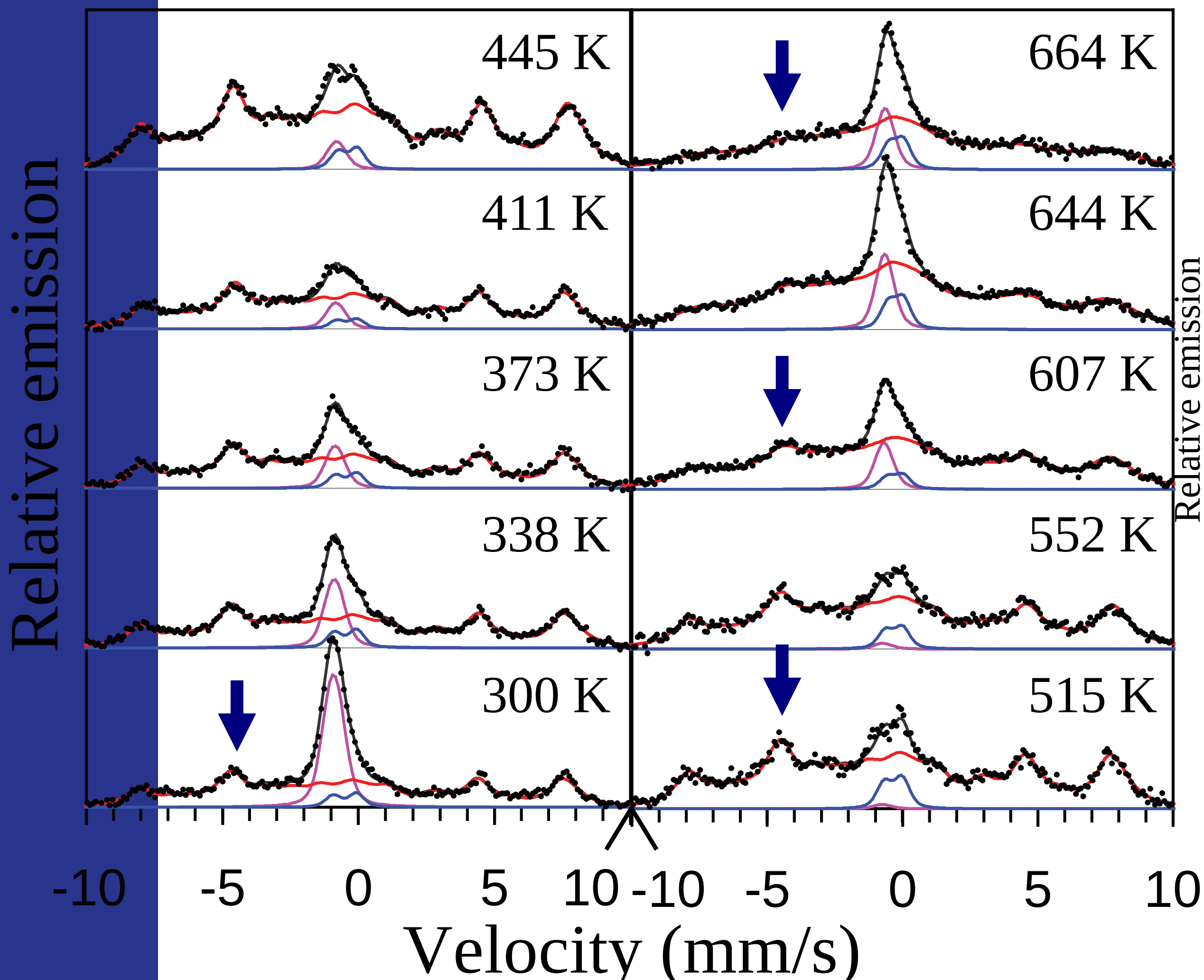
→ Examine temperature dependence of the paramagnetic structure

B_{ext} = 0 T:

→ More complex magnetic sextet structure

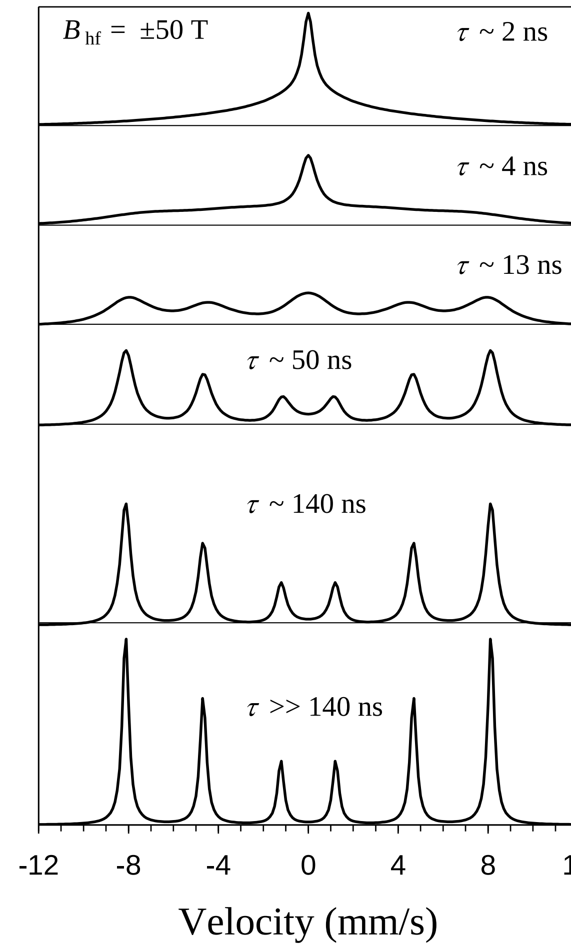
Temperature ↑ : Broadening ↑

ZnO: B_{ext} = 0 T



- T. E Mølholt *et al.* Physica Scripta, T148 (2012) 014006

simulation *Blume M. and Tjon J.A.: Phys. Rev.* ***165****, 446 (1968)*

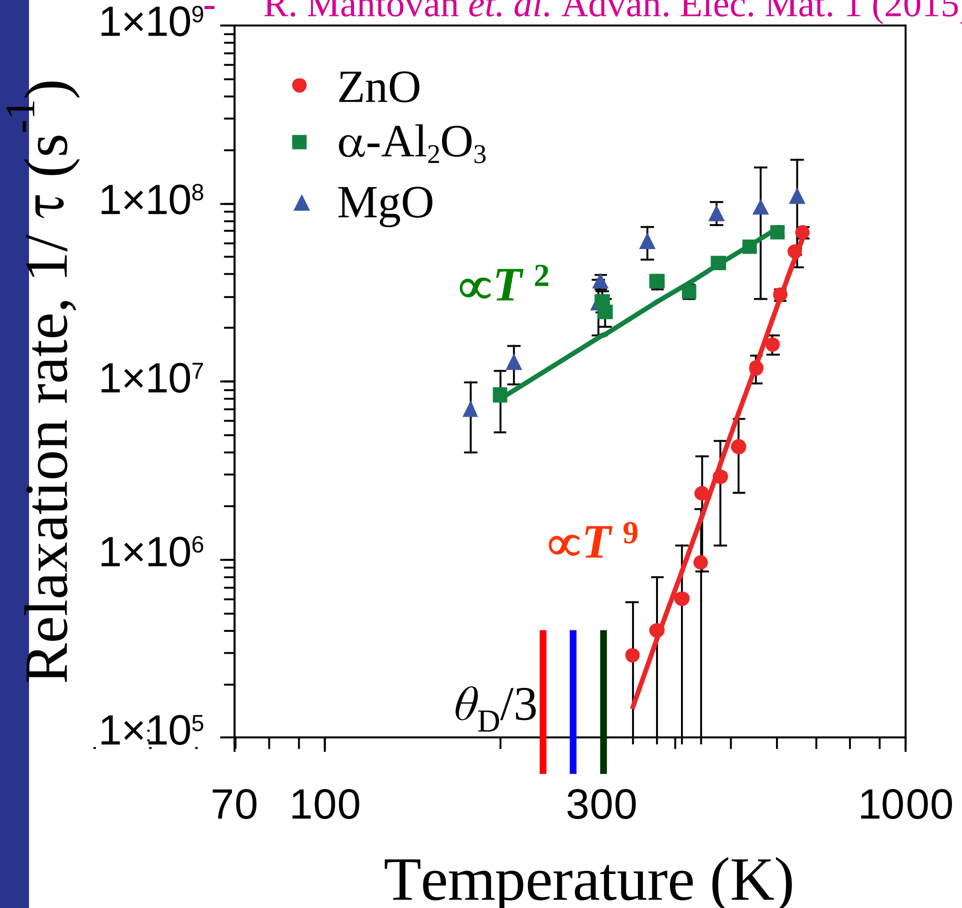


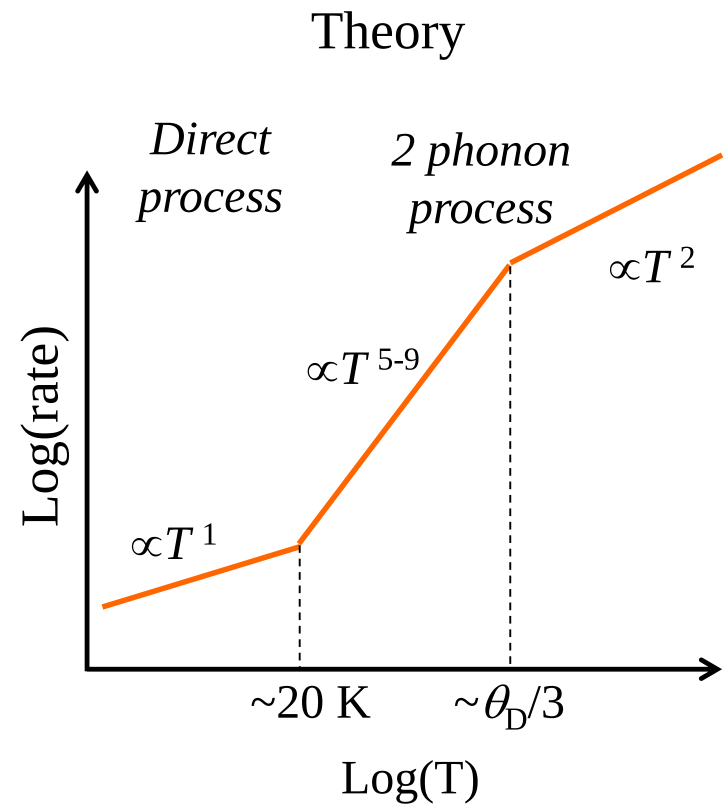
$$\Delta\Gamma = \frac{2\hbar c}{E_0} \cdot \tau^{-1}$$

- T. E Mølholt *et al.* Hyp. Int. 197(2010) 89-94

Spin-lattice relaxation rates in studied oxides

- T. E Mølholt *et al.* Physica Scripta, T148 (2012) 014006
- T. E Mølholt *et al.* Hyp. Int. 197 (2010) 89-94
- H.P. Gunnlaugsson *et al.* Hyp. Int. 198 (2010) 5-14
- R. Mantovan *et. al.* Advan. Elec. Mat. 1 (2015) 1400039





θ_D
- MgO: ~ 730 K
- α-Al_2O_3: ~ 1050 K
- ZnO: ~ 300 – 700 K

On-going and future Mössbauer studies at ISOLDE

Make use of more ISOLDE beams

- On-line
- Off-line (longer lived Mossbauer isotopes), b508

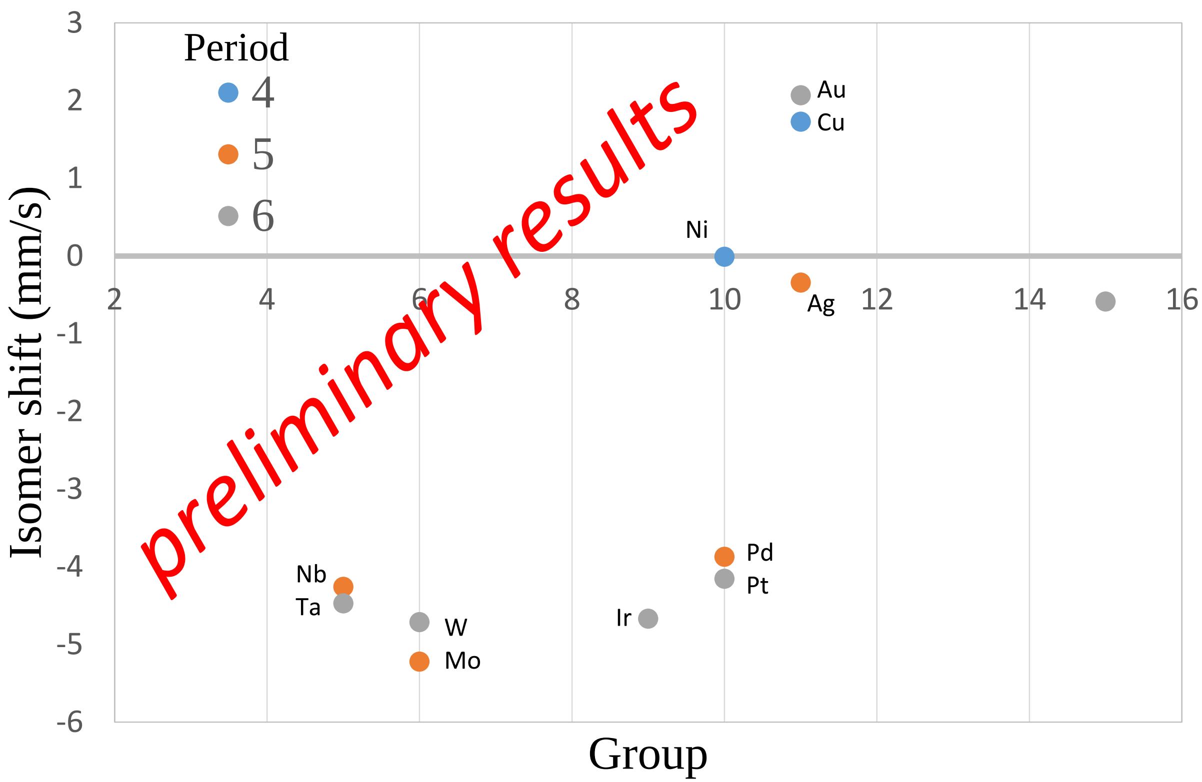
Please see Talk at the ISOLDE Workshop by Haraldur Páll Gunnlaugsson:

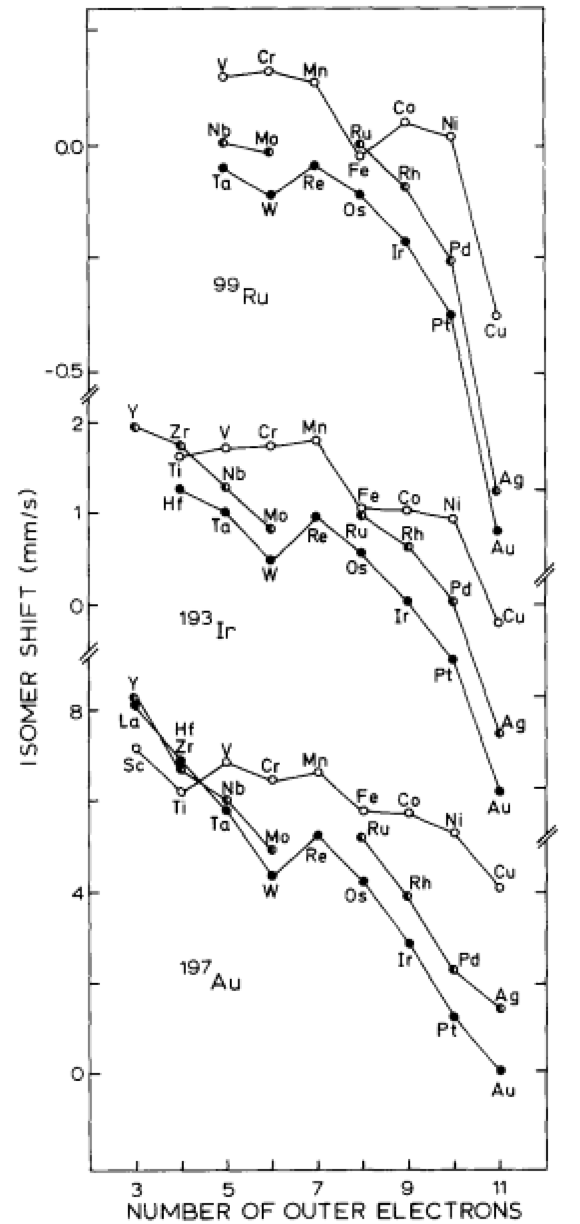
- Friday 4th Dec. 09:30

^{151}Eu Mössbauer

June/July 2015: ^{151}Dy-beam, $T_{½}$~124d (^{151}Gd)

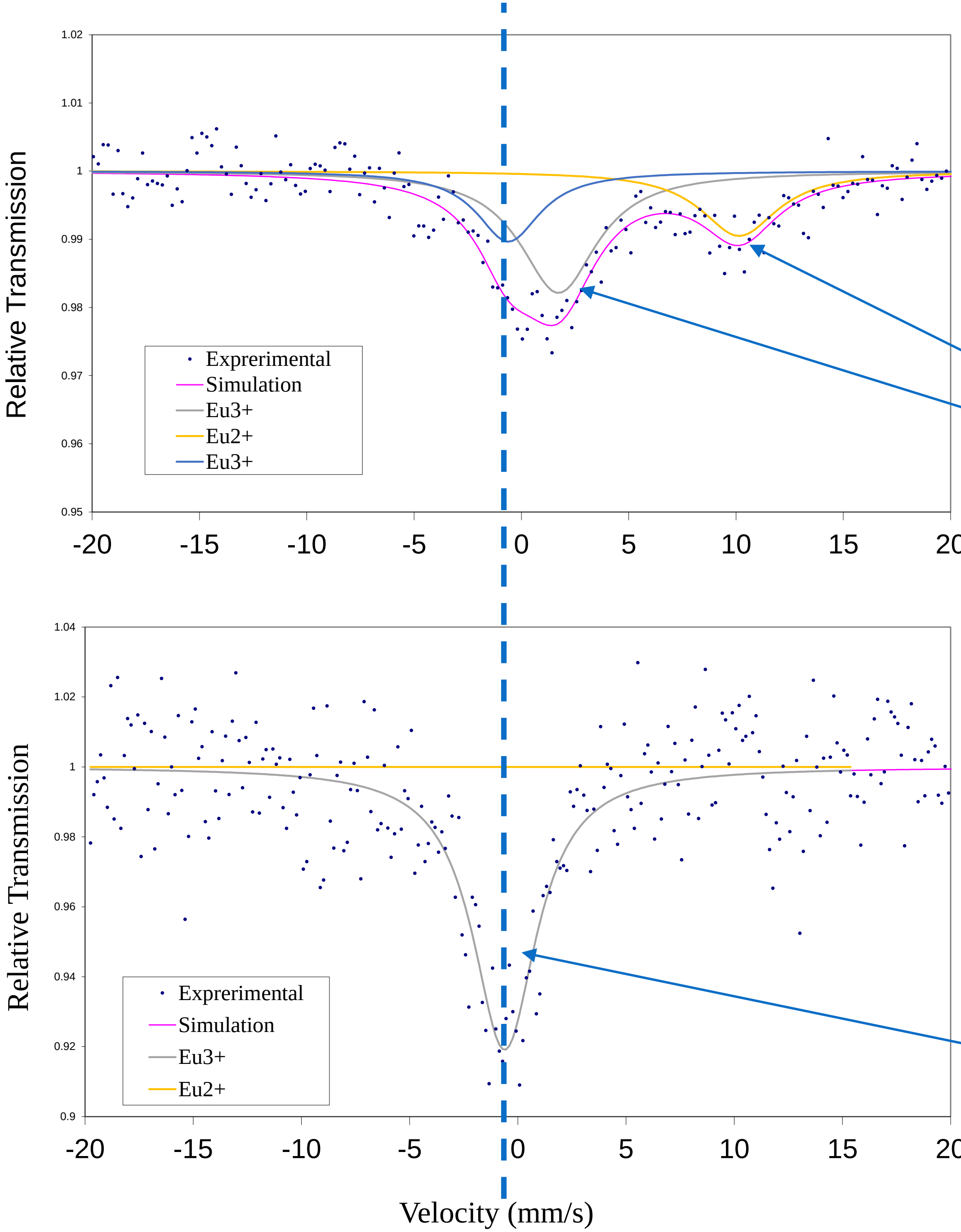
- RE doping: manipulate optical properties in semiconductors
- Samples made in minutes
- Measurements of ~20 samples ongoing





F.E. Wagner *et. al.*, Physics Letters A **42**, 7, (1973) 483

Cu sample. ^{151}Eu Measured at RT



As implanted

Implantation related sites

- *Damage (not perfect lattice)*
- *Vacancies*
- *Interstitial*

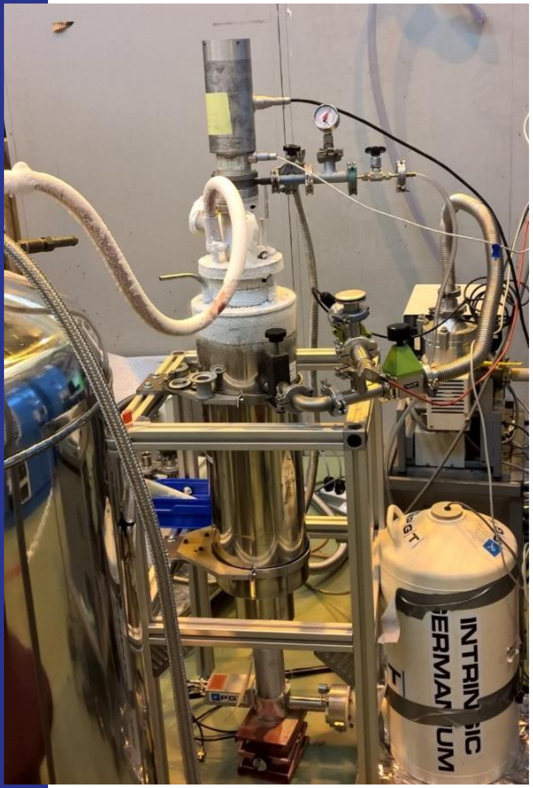
After annealing:
350°C for 30 min.

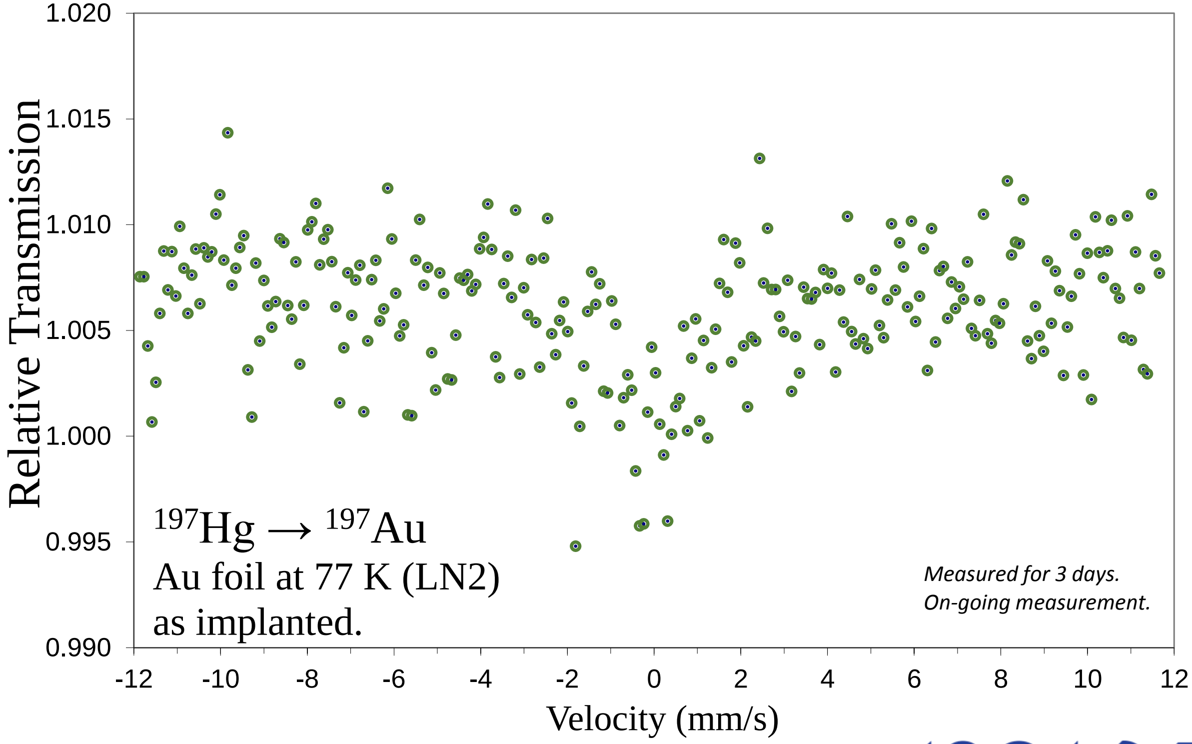
Substitutional site

^{197}Au Mössbauer

November 2015: ^{197}Hg beam, $T_{½}$~64h

- Test for Bio-physics (INTC-2015-008, I-161).
- Low Hg-yields to LA2 (sample made in *several* hours):
  - → No bio-physics. But proof of feasibility/calibration.





Emission Mössbauer at ISOLDE

- ! The ISOLDE isotope beams are our **tools** for Mössbauer studies !

Usage of **additional isotopes** for extended studies and possibilities

Conclusions

- Mössbauer is a unique atomic-scale measurements of electronic, magnetic, and structural properties within materials.
  - ISOLDE is the perfect tool to create and study doping and defects in materials.
- Showed some specific results.
  - Interstitial Fe in MgO.
  - Paramagnetism is oxides.
- Expanding the isotopes used for eMS at ISOLDE.
  - Further doping possibilities.
  - Bio-physics.

Thanks for your attention