Spherical neutron polarimetry (SNP) in multiferroics under external stimuli

V. Hutanu

Institut für Kristallographie RWTH Aachen University,
JCNS outstation at MLZ, Garching, Germany

РНСИ-КС-2014

Sankt-Petersburg, Russian Federation, October 31, 2014

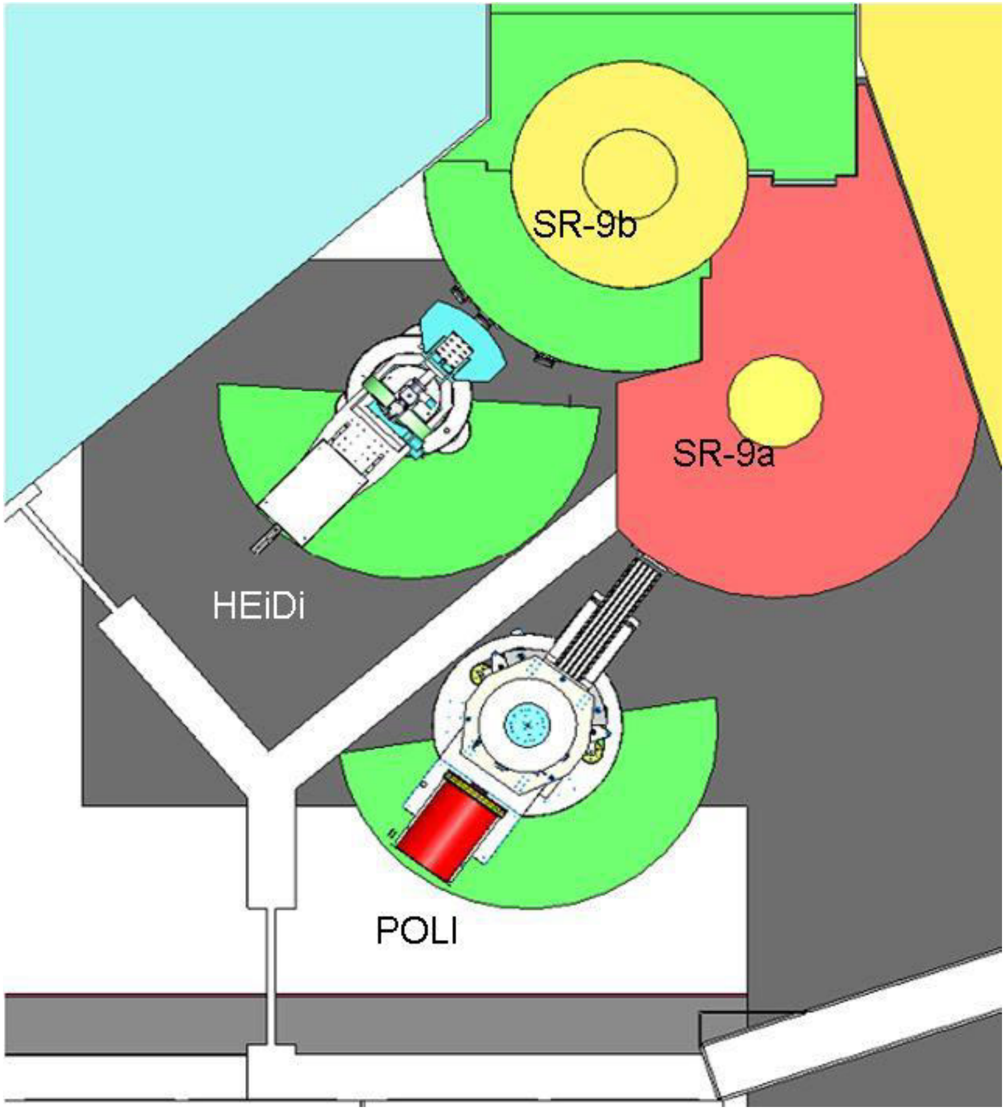
Outline

- Short introduction to POLI
- SNP on POLI
- Electric field in Cryopad

  - Cycloidal magnetic structures $TbMnO_3$, $DyMnO_3$
- Magnetic field influence (ZFC versus FC) on domain structure

  - 2D antiferromagnetic compound $Ba_2CoGe_2O_7$
- Electric + magnetic field in classical magnetoelectric Cr_2O_3
- Summary

POLI bekommt eigenen Strahl

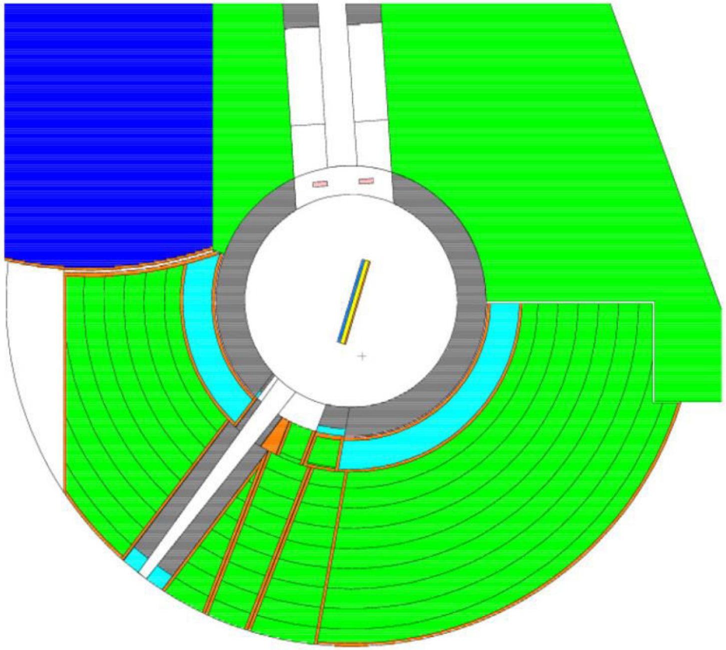


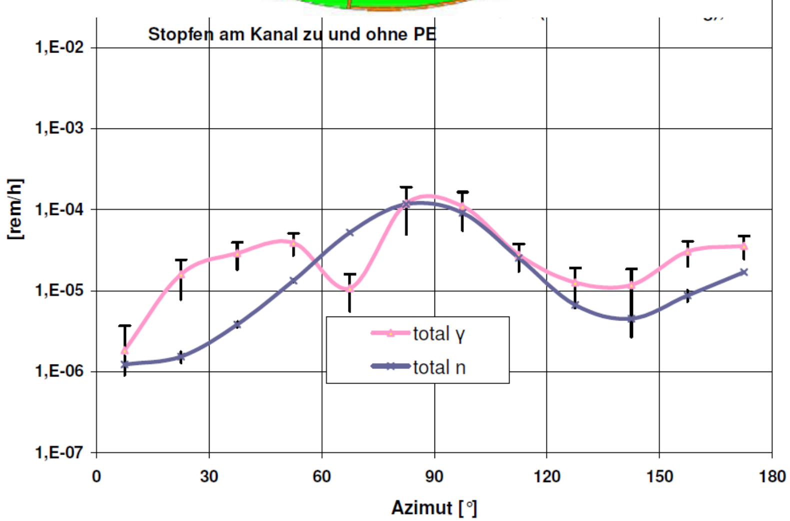
2010 – 2013 1.4 Mio. €
BMBF Project at RWTH Aachen for independent operation of the two diffractometers at the hot sourse FRM II (SR 9a / SR 9b)

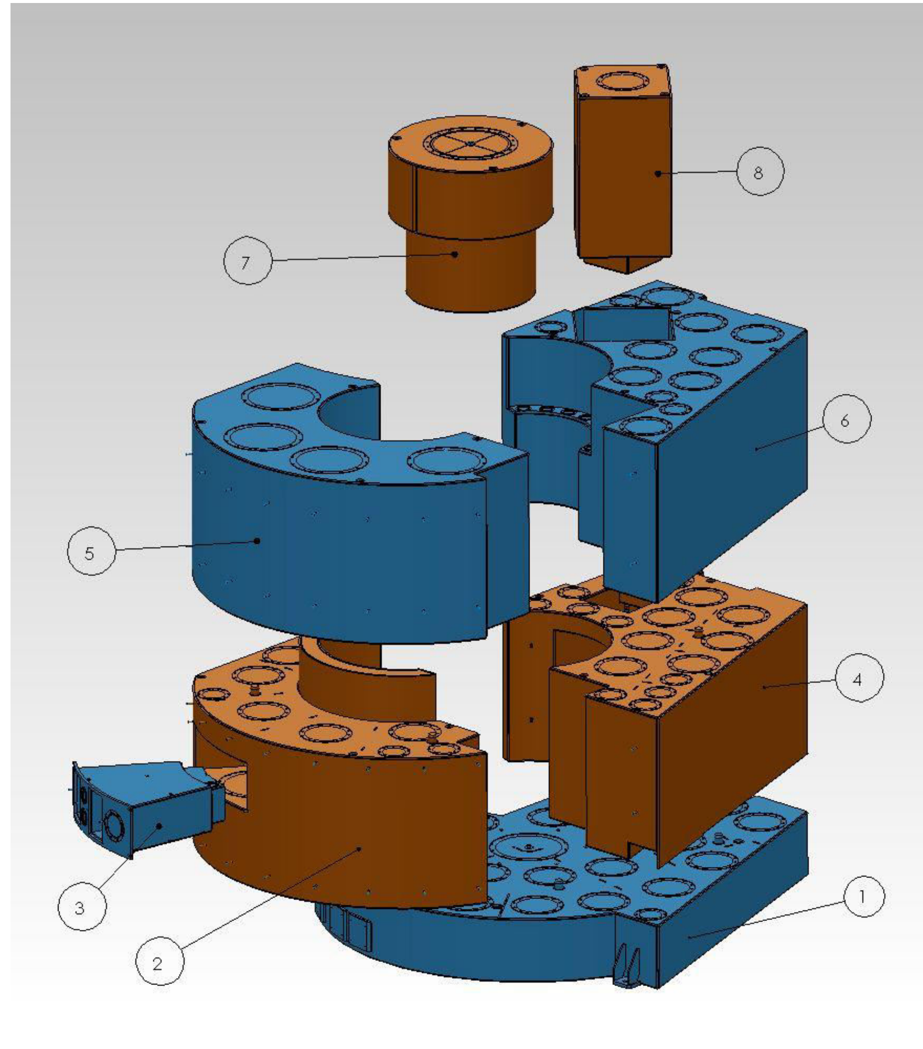
- Strahltrennung
- Neue Burg
- Kanal Aufbereitung
- Monochromatoren
- Infrastruktur für eigenen Messplatz
- Methodische Entwicklungen

POLI Burg

POLI Burg Model für MCNP Simulationen







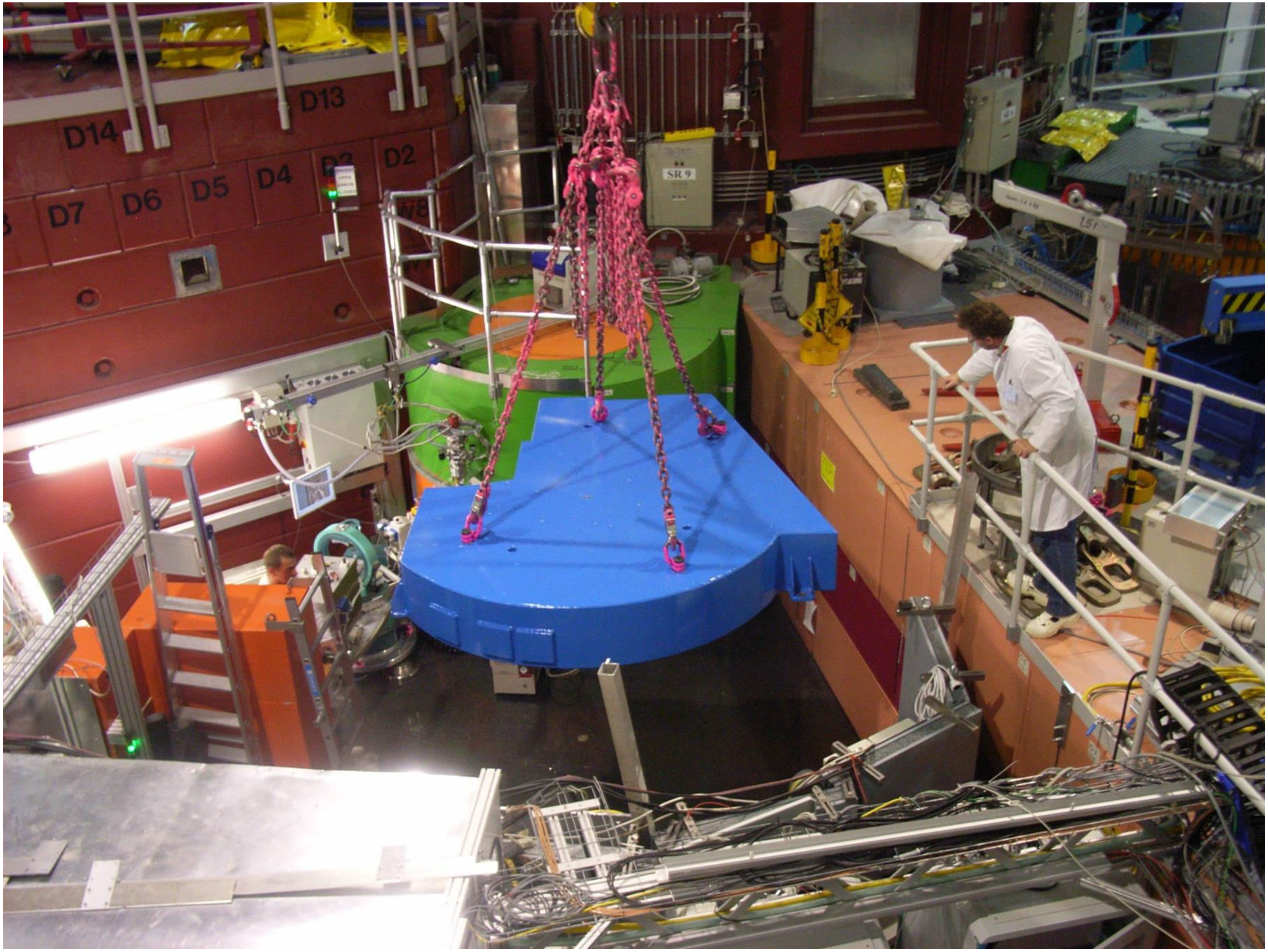
Novel recyclable TUM patented shielding material combined with Pb and BPE layers
Installation: October – November 2012

POLI Burg

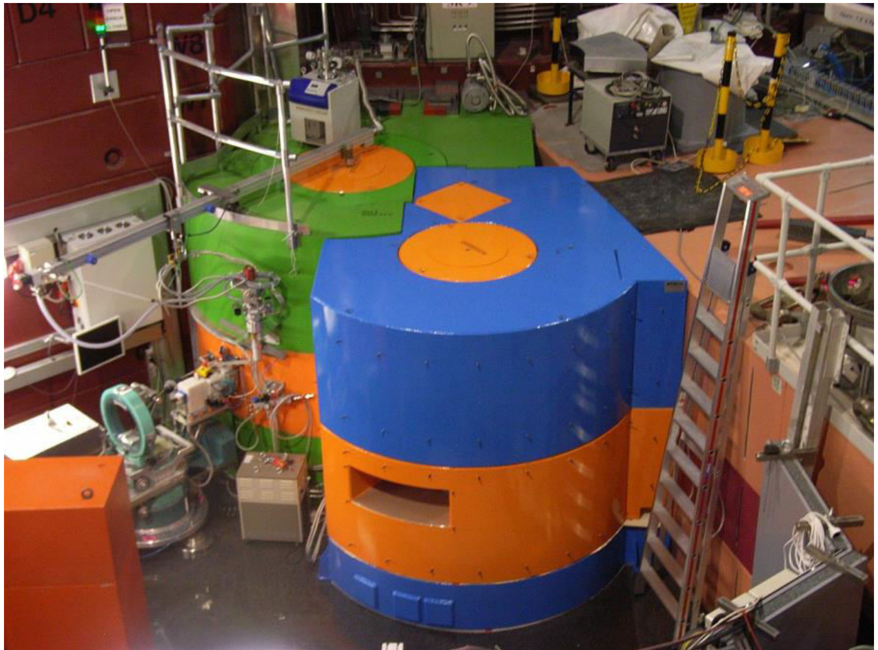
Intensive Begleitung bei der Fetigung bei Fa. …

- Überprüfung Stahlbau (Aufbesserungsprotokoll)
- Befühlung
- Dichtigkeittest (O-ring Material Änderung)
- Abname im Werk (Aufbesserungsprotokoll)
- Vorortabnahme (Aufbesserungsprotokoll)

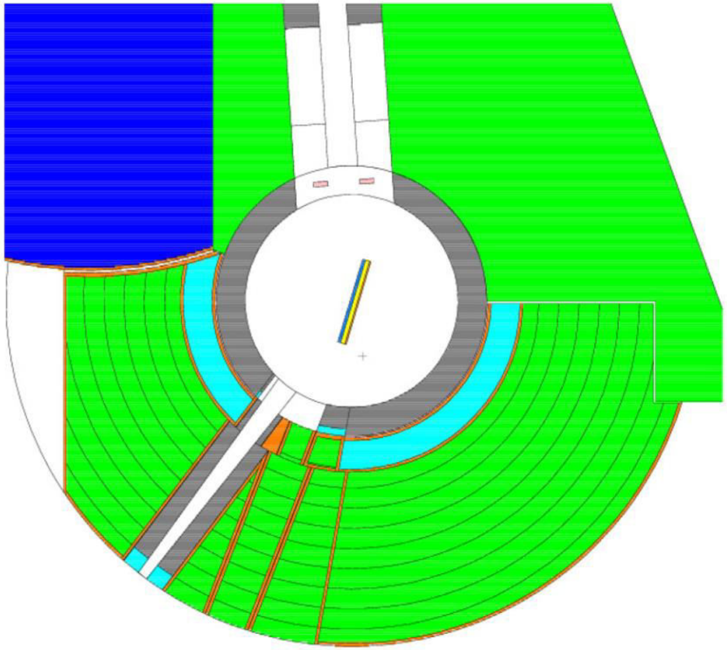
POLI Burg

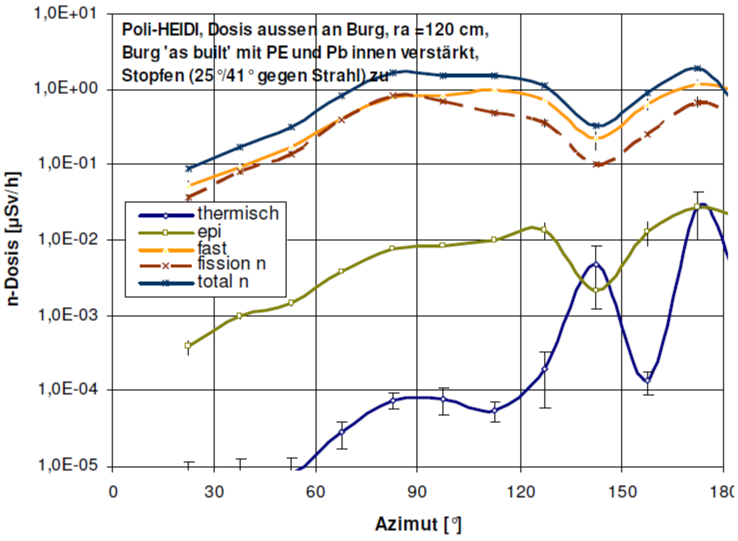


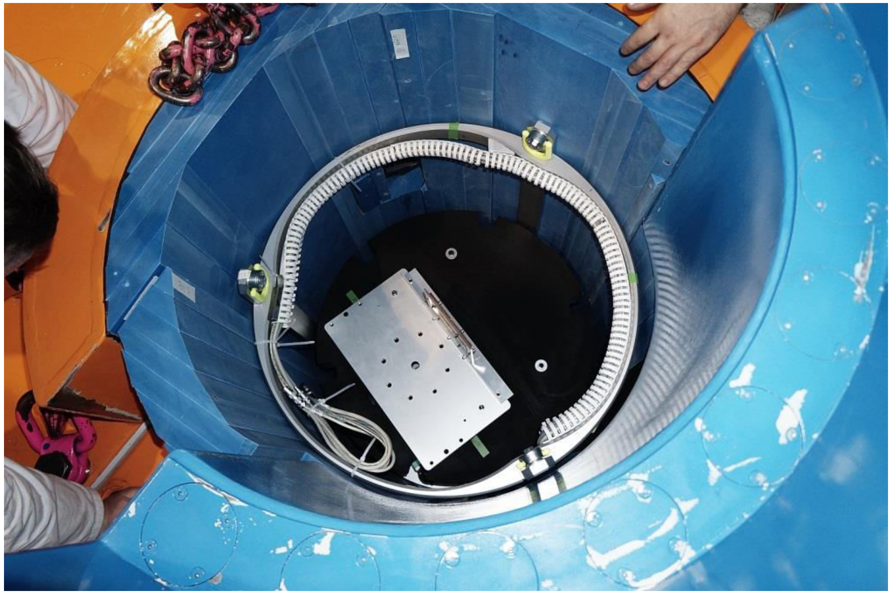
POLI Burg



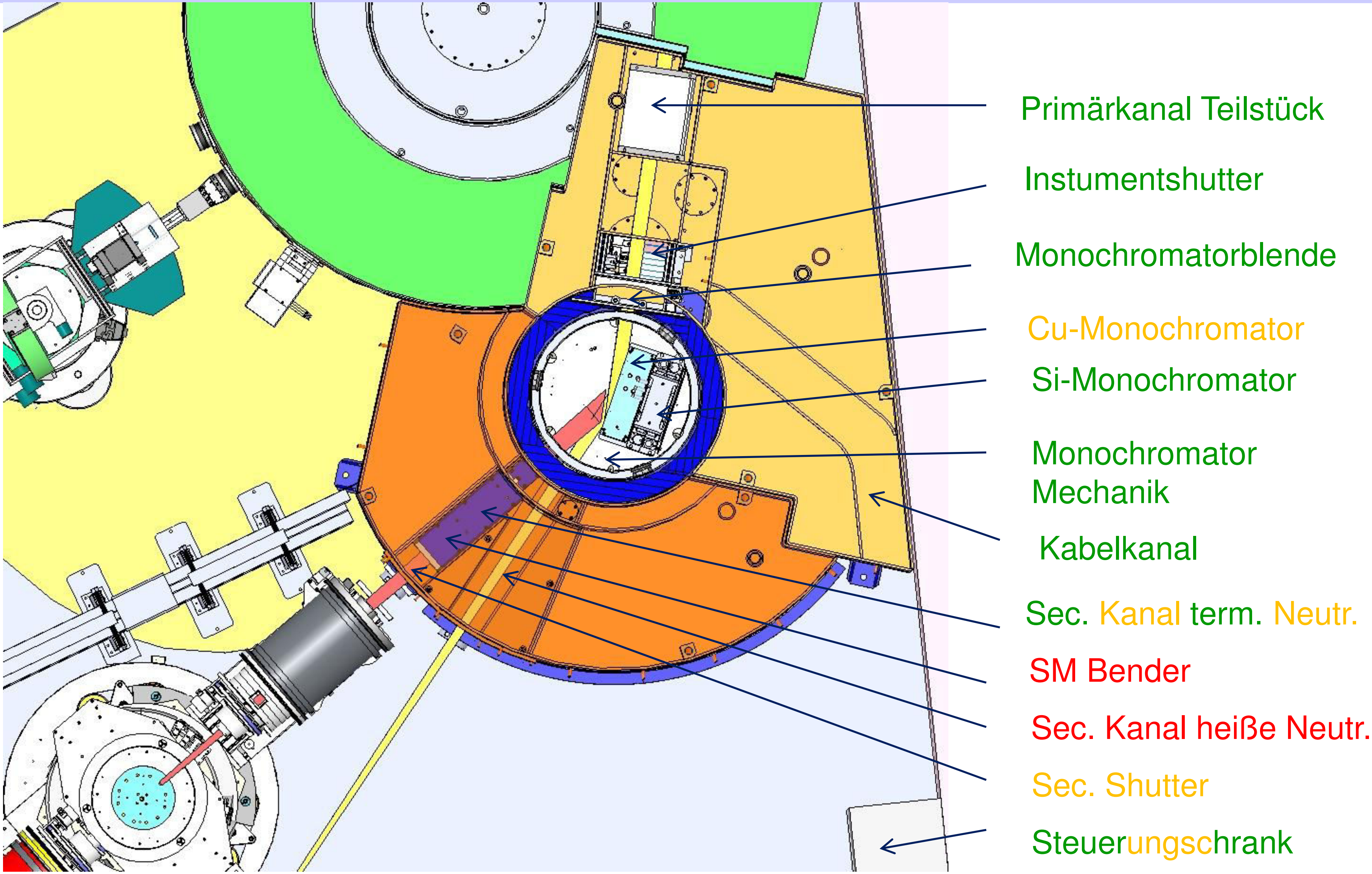
Burg Innenauskleidung





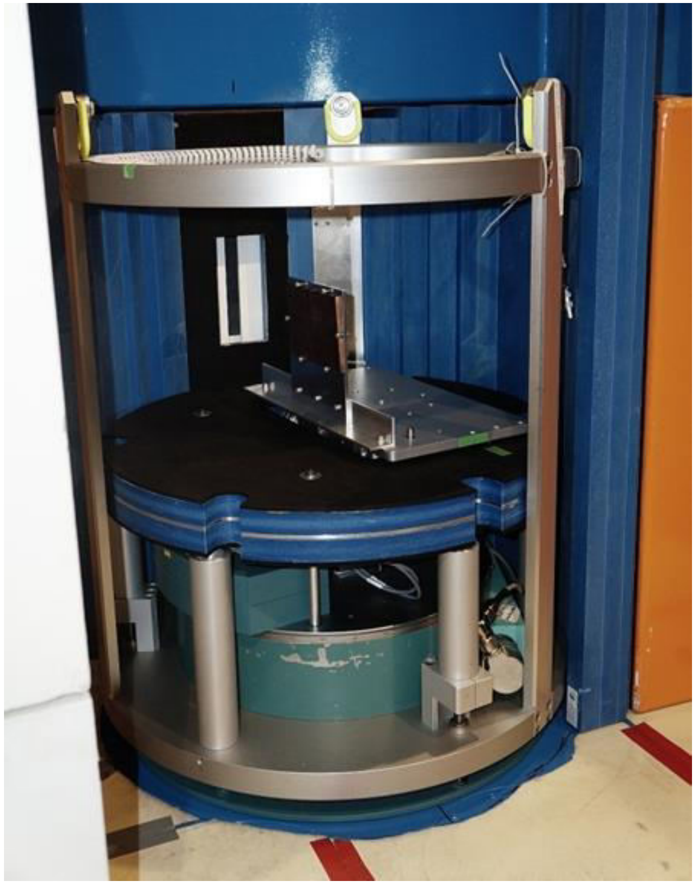


Innere "Werte"



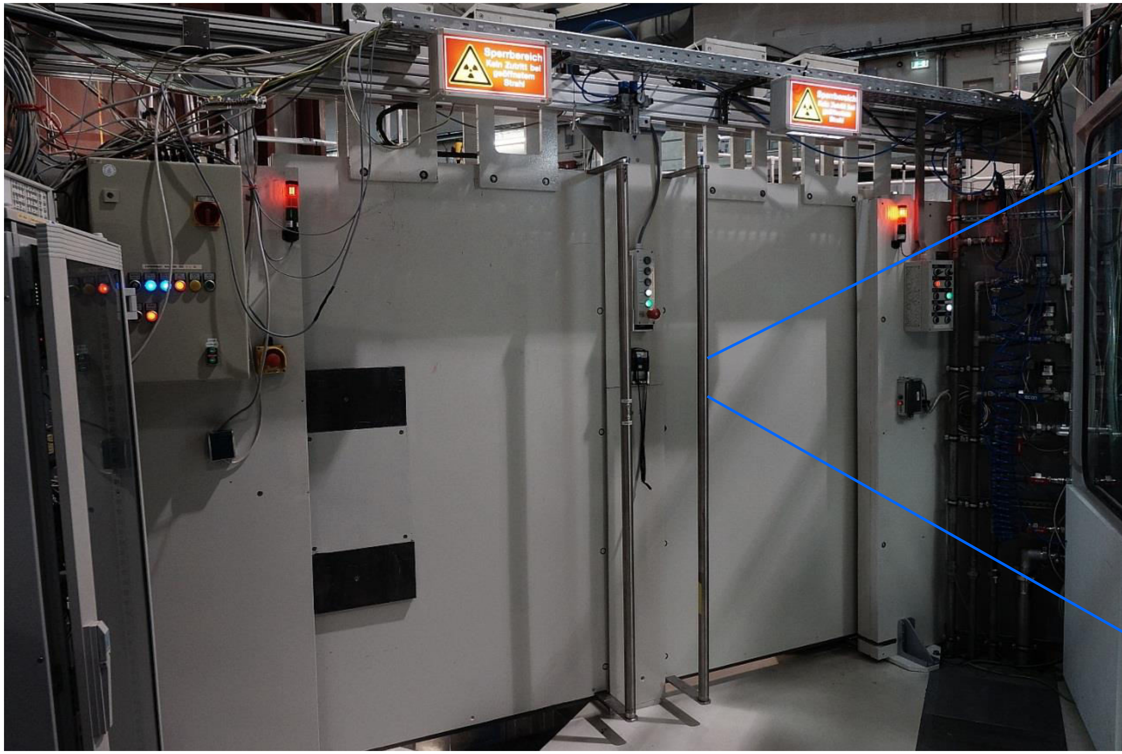
Monochromator Positioniermechanik

Konstruktion/Fertigung Fa. Huber,

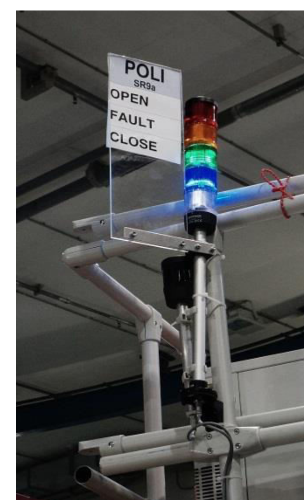


FZ Jülich ,(IFF Werkstatt) Anpassung für den Strahlumgebung, (GELI) Elektronik-montage, Verkabelung;
(ZEA 2) Steuerungsimplementierung

Sicherheitskonzept



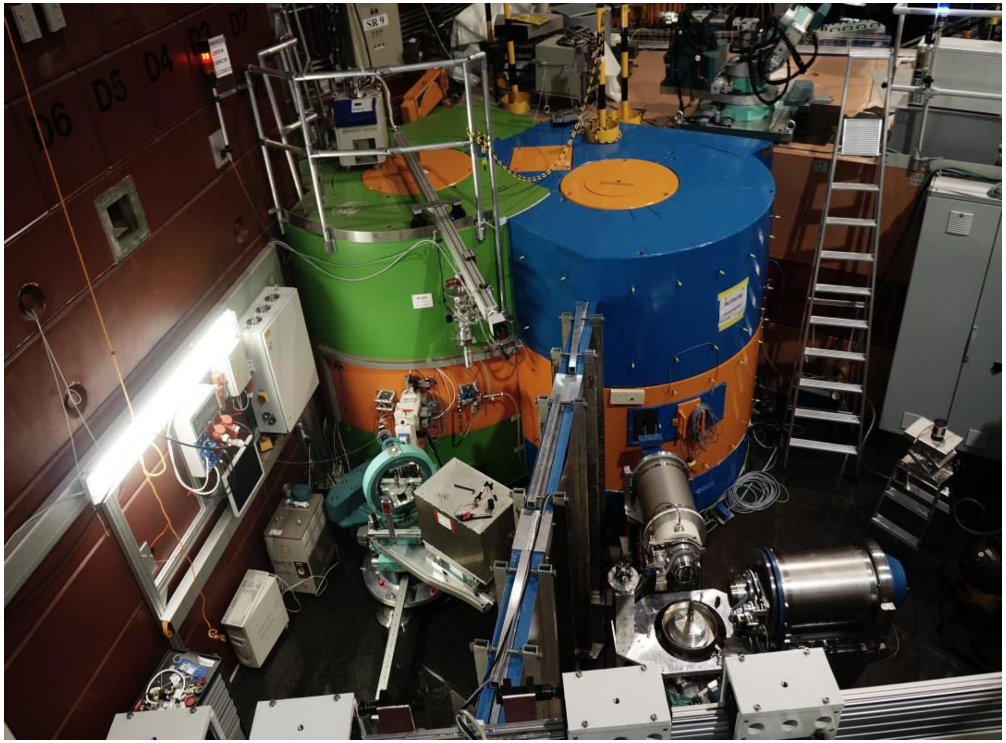
Pneumatische Unterstützung



Zugangsüberwachung mit Warnsignalen
3 x Not-auss (Tür, Exp. Feld, Messkabine)
SPS programmiert (Möller, Kämmerling)

R. Lorenz: „Testbetrieb zugelassen“

Testexperiment - Erfogsmeldung



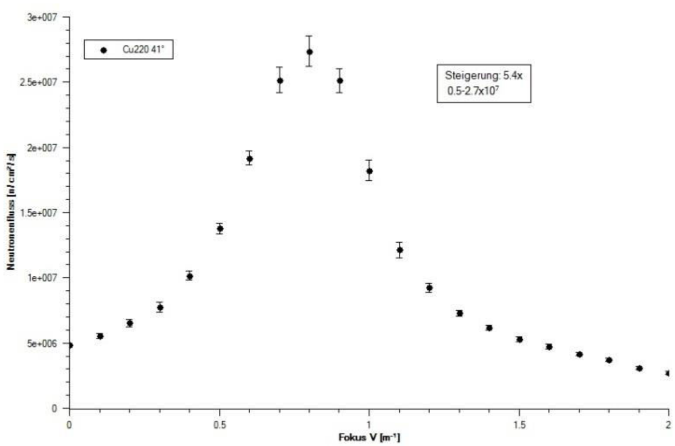
Cu (220) Testmonochromator

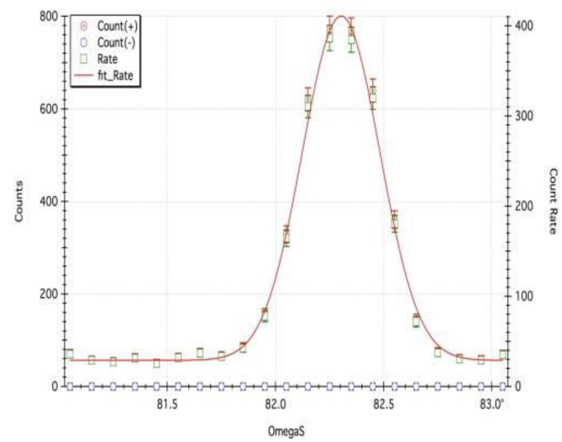
Hommogene Ausleuchtung von Probenort mit 0.9 Å monochromatischen Neutronenstrahl

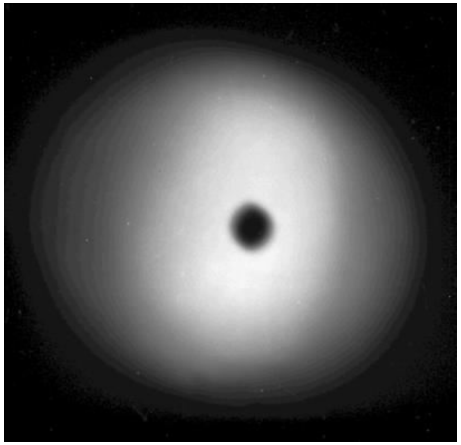
λ /2 Kontamination etwa 4%

Fluss: 5.8×10^6 n/s/cm^2 – Probe

2.8×10^9 n/s/cm^2 - Monochromator

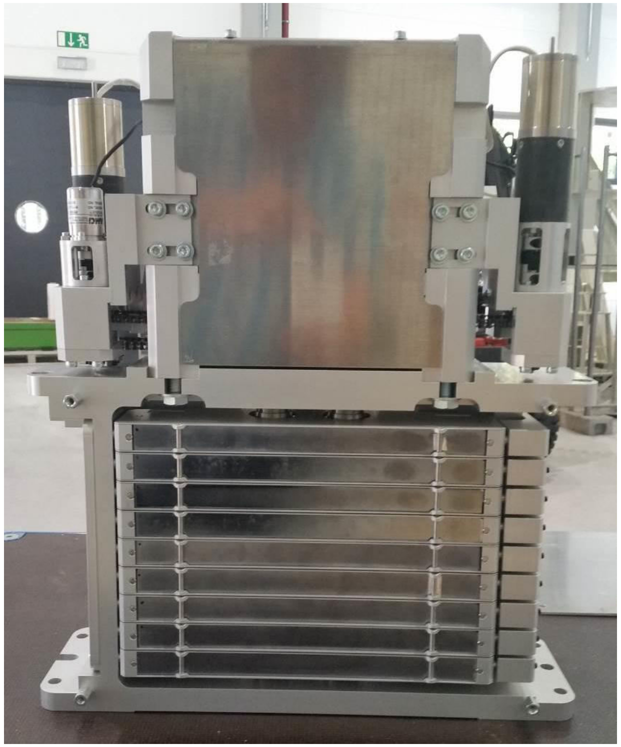






POLI Monochromatoren Si

Vertically focused horizontally bent Si (311) Monochromator (ILL; Fa. Bisson)

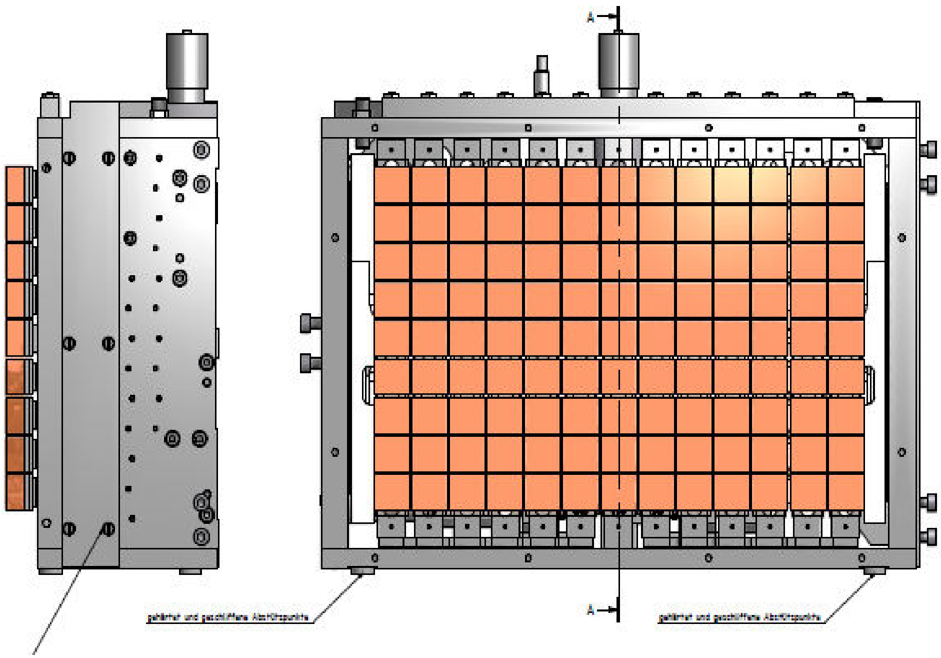


FERTIG !!!

Lieferung 06.2014

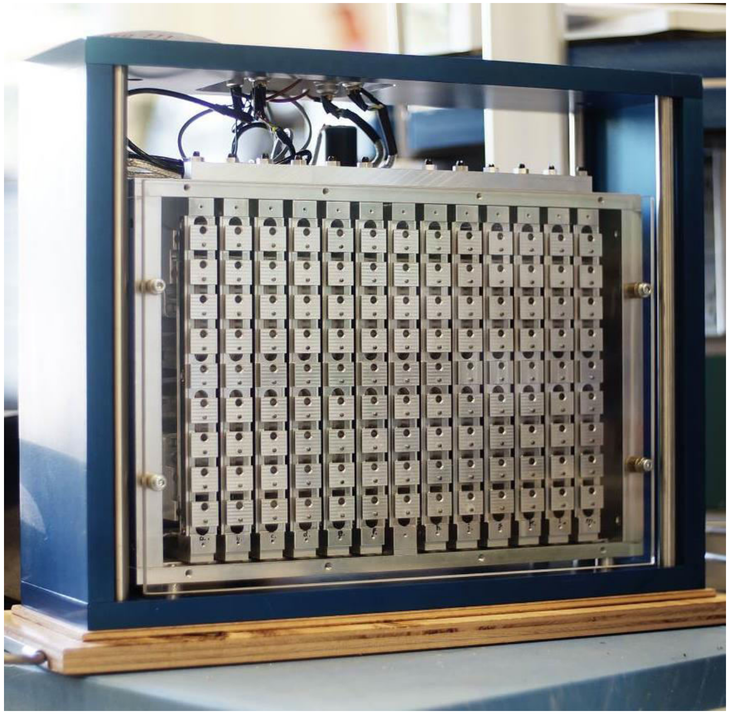
- Vertically focused
- 204 mm x 270 mm
- Horizontally bent
- 0.7 Å and 1.14 Å
- 9 x 20 mm Si wafers stacks

POLI Monochromatoren Cu



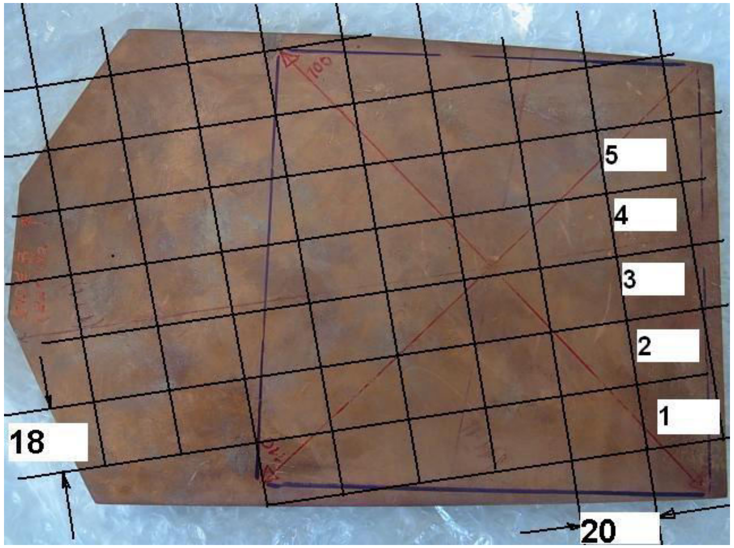
2013: Mechanik Fertig
in Jülich mit Huber Mechanik angepasst.
2014: In Göttingen zur Vermessung, Kristalljustierung, Befestigung

Douppellfokusierte Mechanik (type PUMA)
mit 117 (13 x 9) Cu (220) Einzelnkristallen
20 x 18 x 9 mm insgesamt 272 x172 mm^2

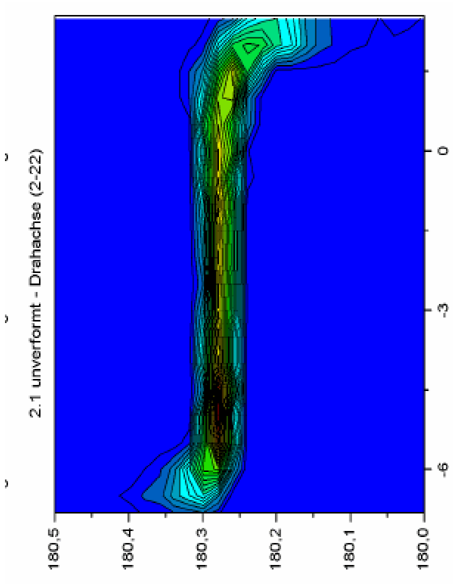


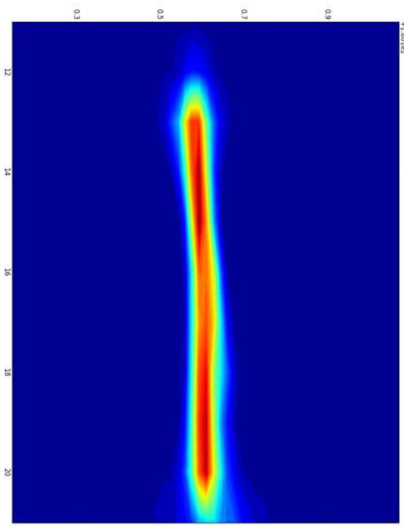
Kooperation IPC Uni Göttingen

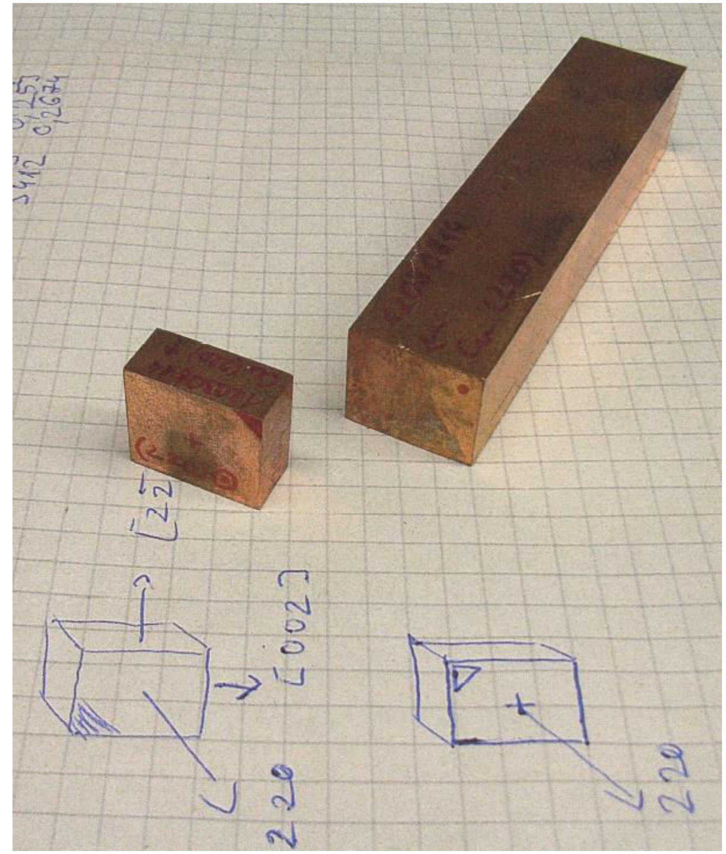
POLI Monochromatoren Cu



Cu (220) HEiDi Platte (46 Stück unverformt)

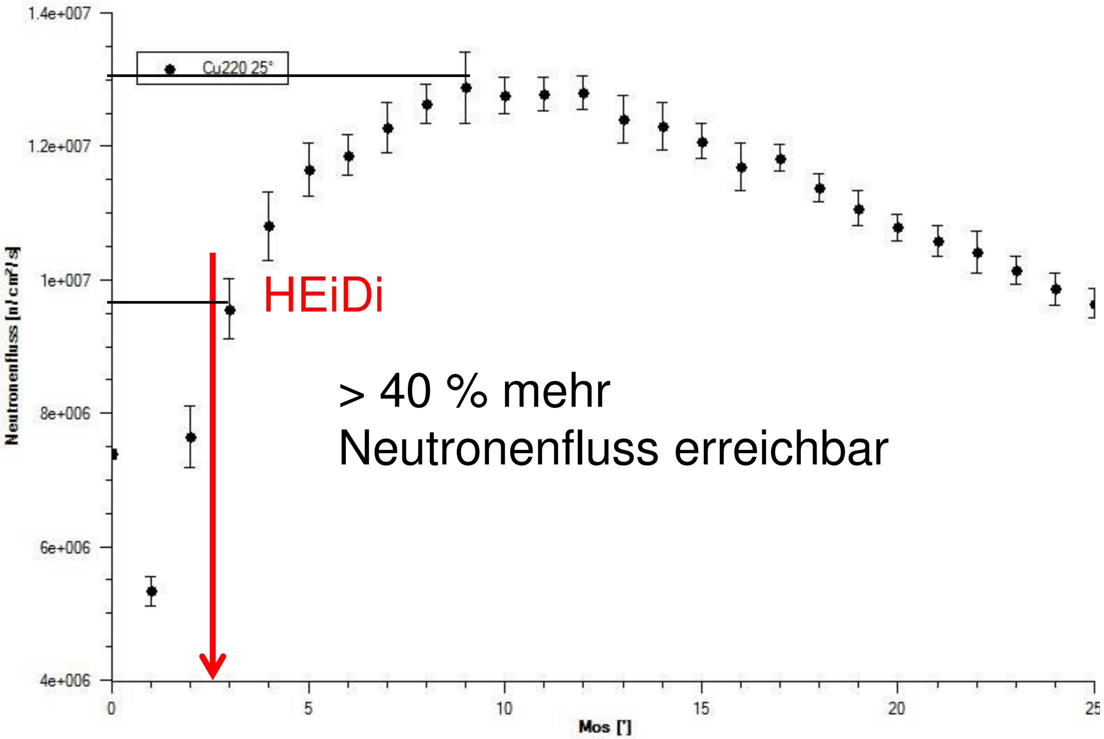




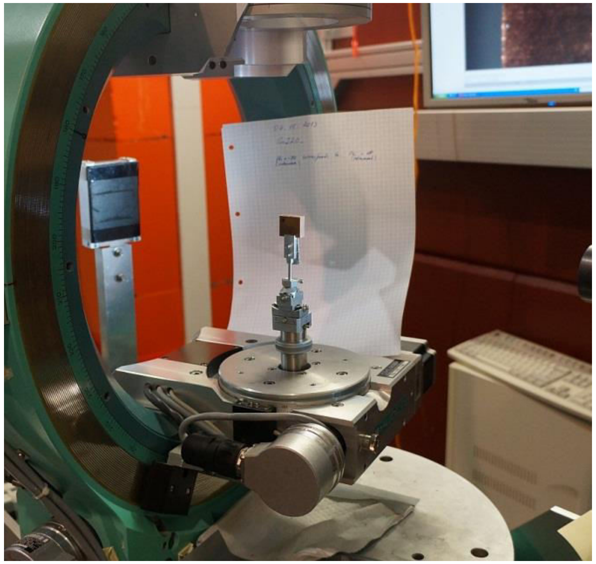


Fa. Mateck 100 Stück (7 Rods):
Jun. 2014: alle Gezüchtet,
4 Rods (60 Stück) geschnitten
/3 kaputt)

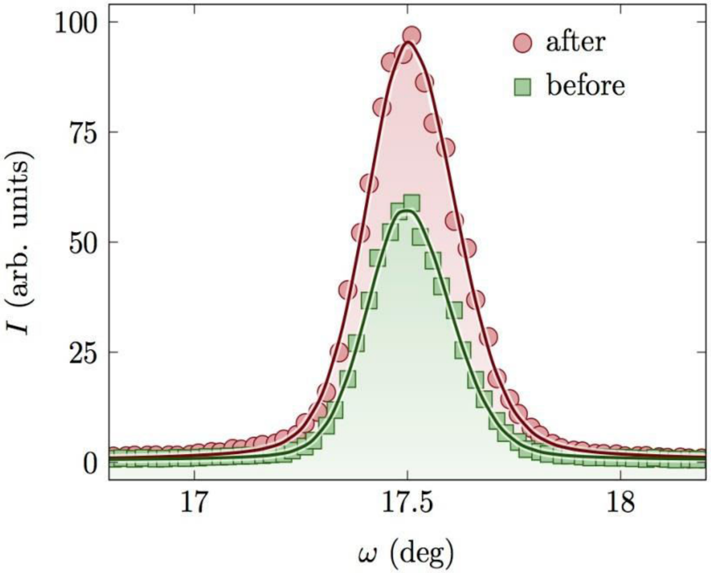
POLI Monochromatoren Cu



> 40 % mehr
Neutronenfluss erreichbar



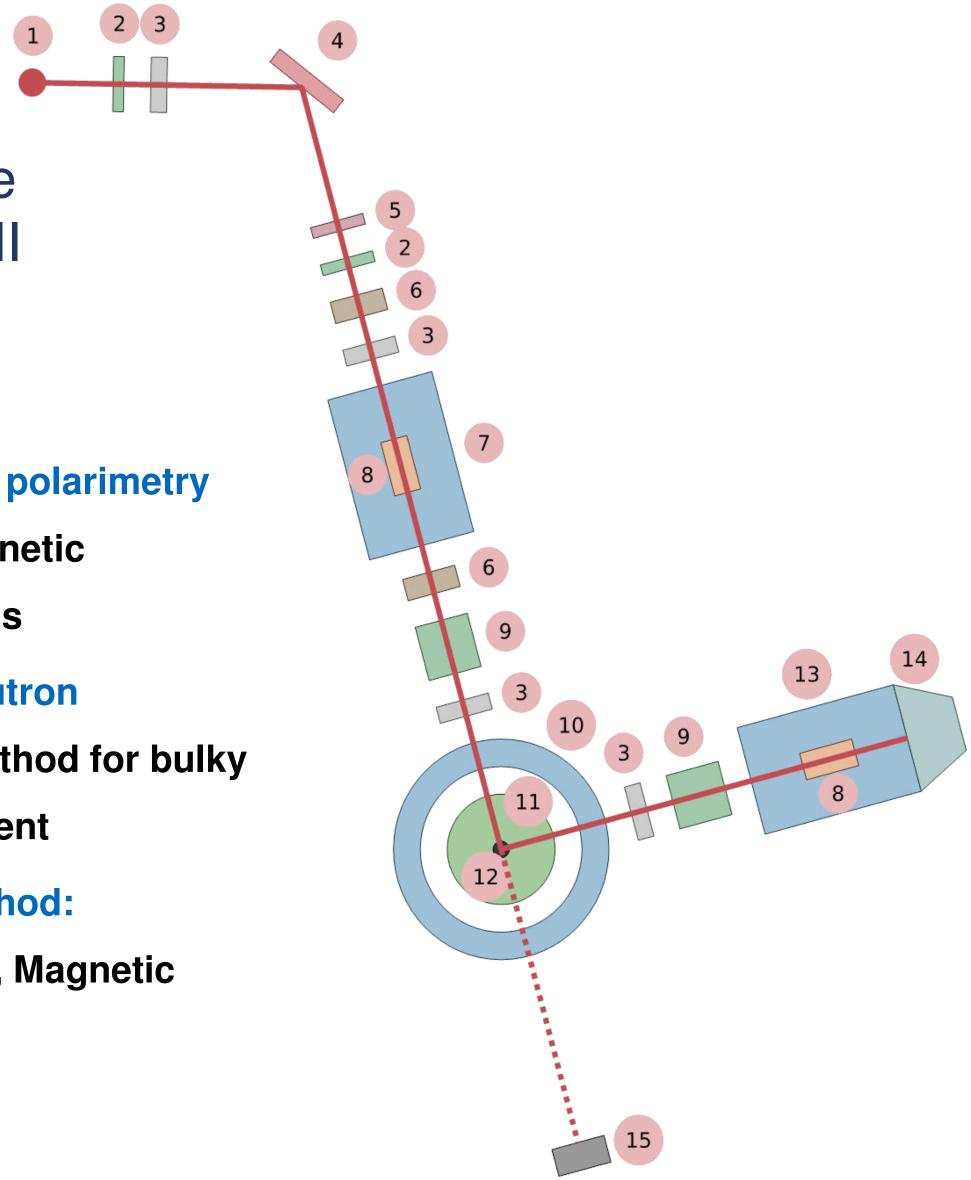
H4 vor und nach der Verformung



Verformung in Göttingen:
alle H Kristalle + 45 Mateck (etwa 90)
Zwieschenergebniss: 40 Fertig, 6 Abfall,
Rest – nachverformen

Termin: insgesamt noch etwa 2-3 Monate

New single-crystal diffractometer at the hot source of FRM II



1. Hot source
2. Shutter
3. Slits
4. Monochromator
5. λ/2 filter
6. Monitor
7. Polariser
8. ^{3}He cell
9. Nutator
10. Cryopad
11. Cryostat
12. Sample
13. Analiser
14. Detector
15. Beam stop

- **Option 1. Spherical neutron polarimetry with Cryopad:** **Complex magnetic structures, Magnetic domains**
- **Option 2. Non-polarised neutron diffraction:** **Conventional method for bulky and heavy sample environment**
- **Option 3. Flipping-ratio method:** **Magnetisation density maps, Magnetic structure form factors**

Option 1. Spherical neutron polarimetry (SNP)

SNP on POLI@ HEiDi with Cryopad is available for users since 2011, until now 11 experiments (7 ext.+ 4 int.)

2 Bachelor thesis,

1 Diploma thesis,

1 PhD thesis,

4 Instrumental papers,

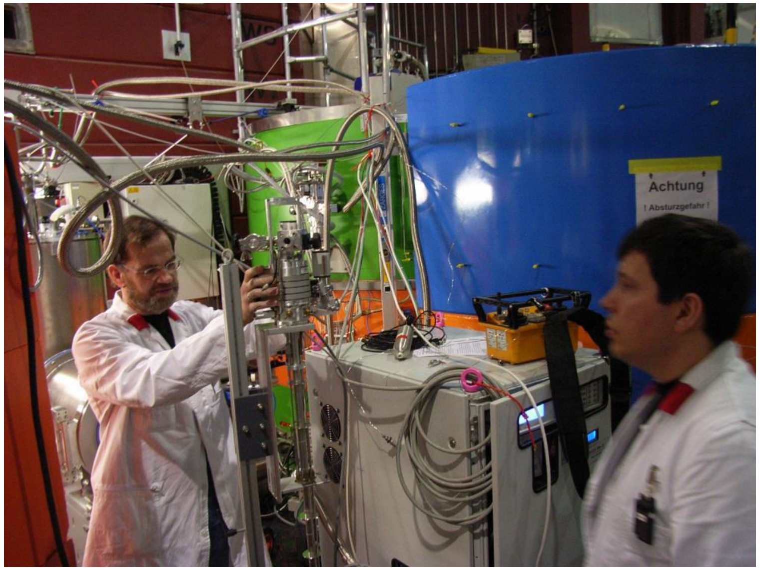
1 Scientific paper published , another 2 in preparation.

V. Hutanu et al., J. Phys.: Conf. Ser. 294 012012 (2011)
V. Hutanu et al., Phys. Rev. B 89, 064403 (2014)

Option 2. Non-polarised neutron diffraction

Complementary to the HEiDi diffractometer for the structure refinement under extreme conditions (T< 2.3 K, Magn. field, Pressure, etc.) using comparable wave length, resolution etc.

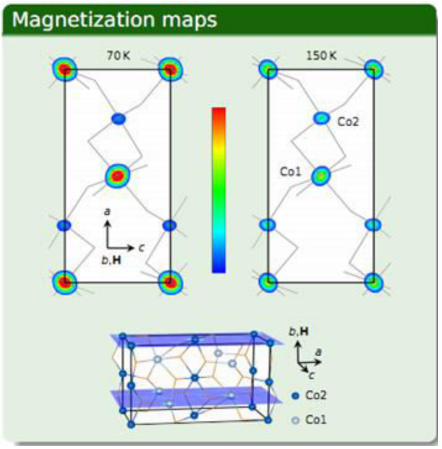
- Lifting counter setup
- 0.4–300 K using CCR cryostat
- New cryostat (4-800 K)
- 7.5 T magnet
- 10 kV el. field



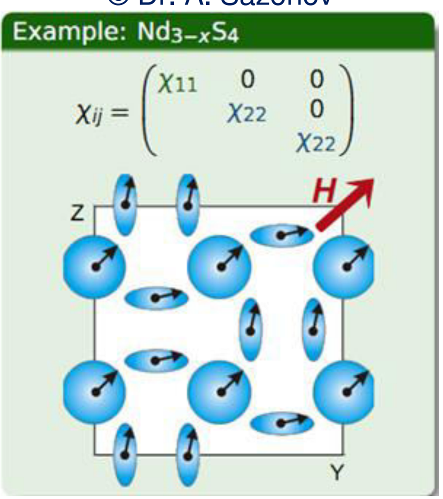
Option 3. Flipping-ratio measurements (PND)

Magnetisation density maps, Local susceptibility aproach, Magnetic form factors

Materials: ferromagnets, ferrimagnets and paramagnets in witch the atomic spins are highly orientated by an applied magnetic field. (Strong saturated field magnet need!)

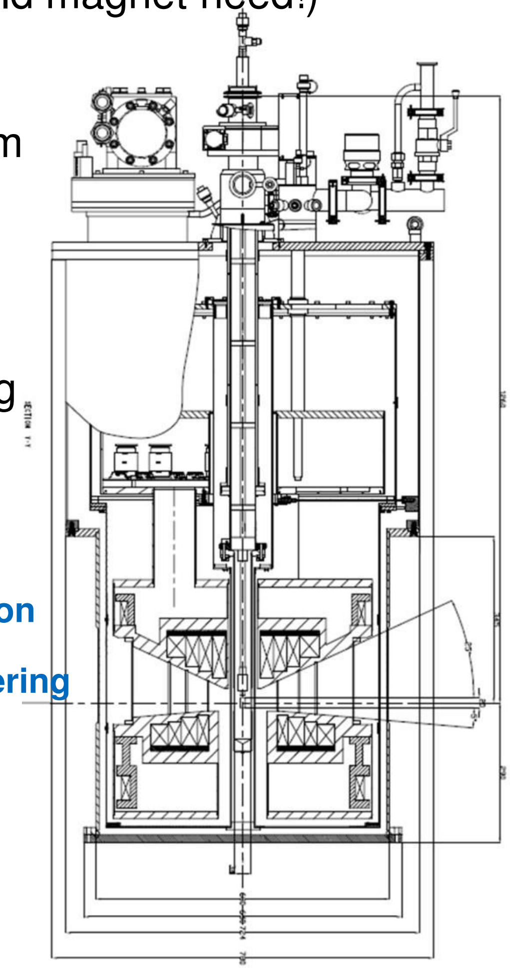


© Dr. A. Sazonov



- Max field: 8 - 9 T / Assymetric
- Actively shielded / stray field: < 50 G@ 1m
- Vertical access: -5° / +25°
- Horizontal access: 300°
- Compact design:

  Outer diameter: < 650 mm/ Weight: < 650 kg
- Low consume LHe cryostat



- **2013 – feasibility study and MLZ directorate decision**
- **2014 – elaboration of detailed specifications, tendering**
- **End 2015 – delivery**
- **2016 first half year – implementing on POLI**

Introduction to SNP

• SNP uses vector characteristic of the neutron polarisation;

• SNP is performed in zero field so the polarisation does not precess, rotation as well as change of the polarisation due to interaction with the sample are analysed;

•SNP distinguishes polarisation rotation from depolarisation;

•Determination of the direction of the magnetic interaction vector;

• Applications:

- Unique solution of complex magnetic structures (collinear or non-collinear AFM, incommensurate structures, direct evidence of chirality)

- Studies of magnetic domains;

- Determination of anti-ferromagnetic form factors;

It is based on the fundamental Blume-Maleev ecuations:

S.W. Maleev, V.G. Bar'yktar & R.A. Surkis, Sov. Phys.-Solid State 4, 2533 (1963)

M. Blume Phys. Rev. 130, 1970 (1963)

P. J. Brown, Spherical Neutron Polarimetry, Ch. 5 "Neutron Scattering From Magnetic Materials, ed. T. Chatterji, Elsevier, 2005

The polarisation tensor

The *Polarisation axes* are defined with:

- x parallel to the scattering vector $\mathbf{k}$.
- z perpendicular to the scattering plane (vertical)
- y completing the right handed cartesian set

With this choice of axes there are no components of the magnetic interaction vector $\mathbf{M}_\perp(\mathbf{k})$ parallel to x.

The Blume Maleev equations can be written in tensor form

$$\mathbf{P}' = \mathsf{P}\mathbf{P} + \mathbf{P}'' \quad \text{or in components} \quad P'_i = \mathsf{P}_{ij} P_j + P''_i$$

$\mathbf{P}''$ is the polarisation created in the scattering process.

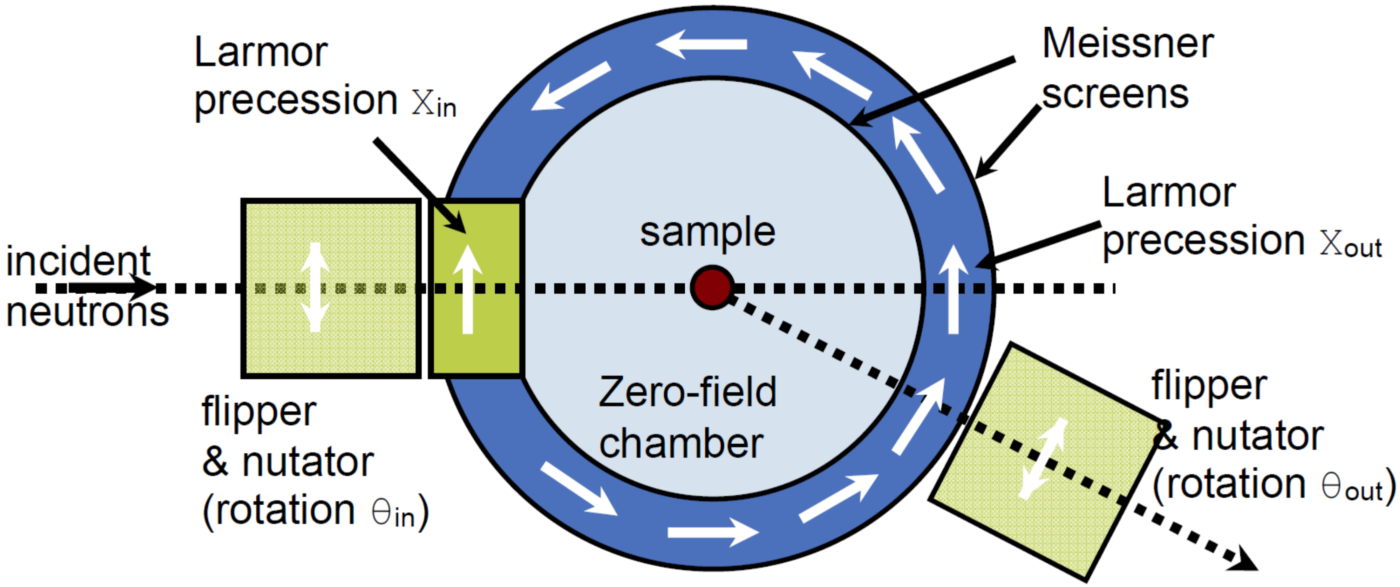
The experimental strategy: Polarisation Matrix

The usual experimental strategy is to measure the scattered polarisation $\mathbf{P}'$ with the incident polarisation $\mathbf{P}$ parallel to polarisation x, y, z in turn.
This determines the polarisation matrix.

The *polarisation matrix* P_{ij} is the experimentally measurable quantity related to the polarisation tensor.
The matrix element P_{ij} gives the ith component of scattered polarisation when the incident polarisation is in the jth direction.

$$P_{ij} = \left\langle \frac{\mathsf{P}_{ij} P_j + P_i''}{P_j} \right\rangle_{\text{domains}}$$

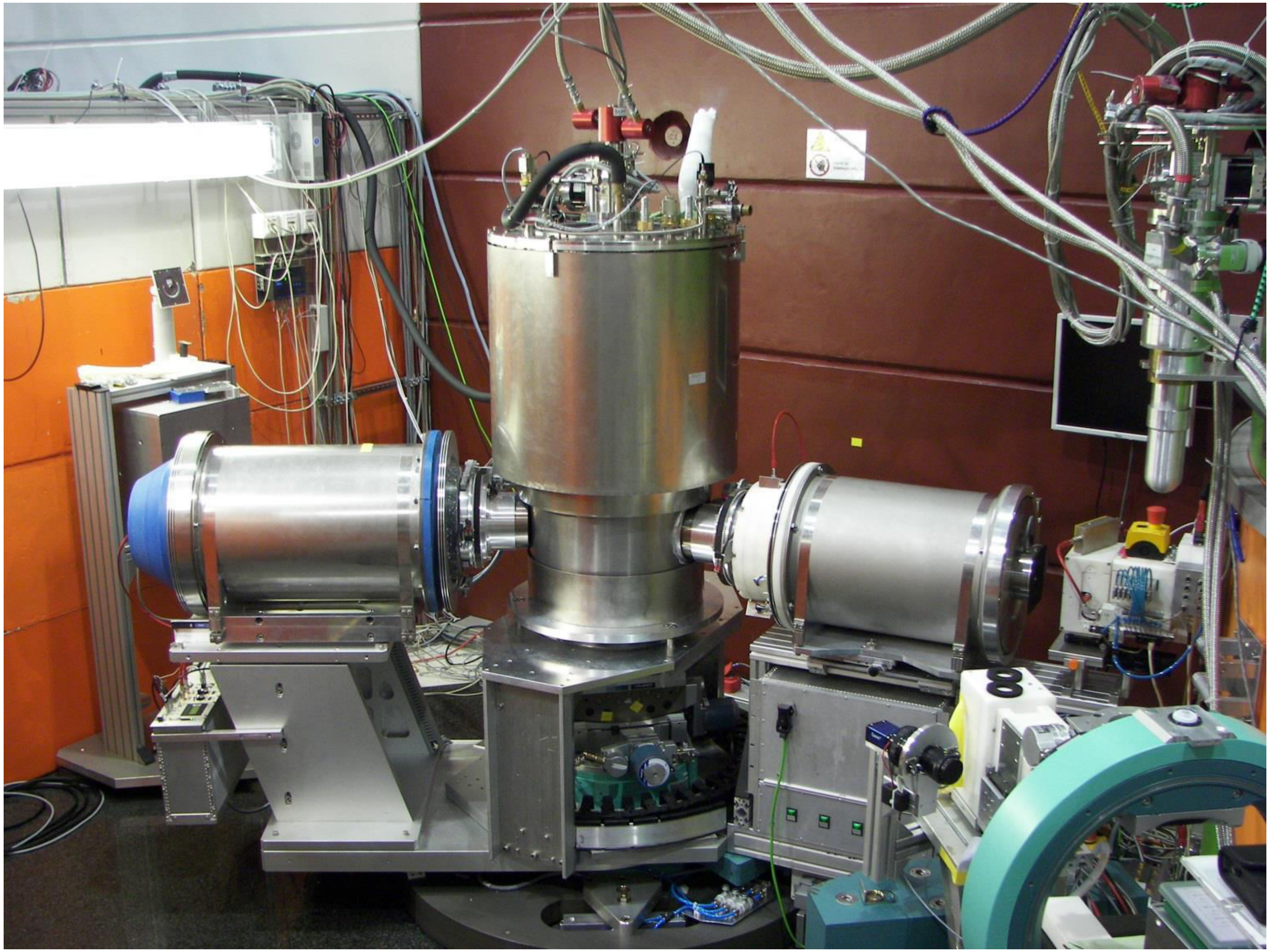
The experimental strategy: Cryopad



F. Tasset et al., Physica B 267}268 (1999) 69

$$\frac{n^+ - n^-}{n^+ + n^-} = P' \cdot A$$

SNP on POLI



V. Hutanu et al., J. Phys.: Conf. Ser. 294 012012 (2011)

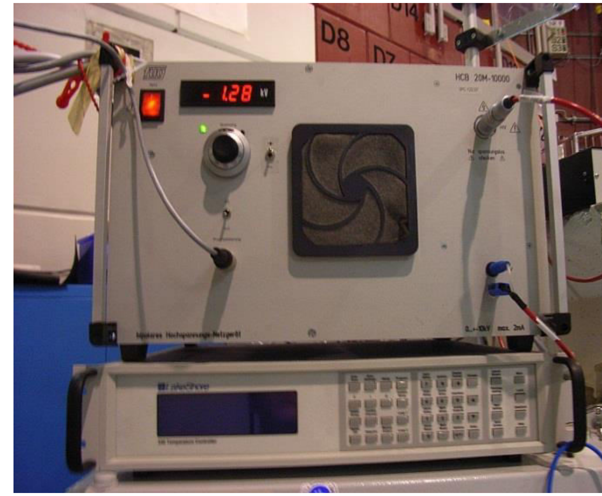
Electric field in Cryopad

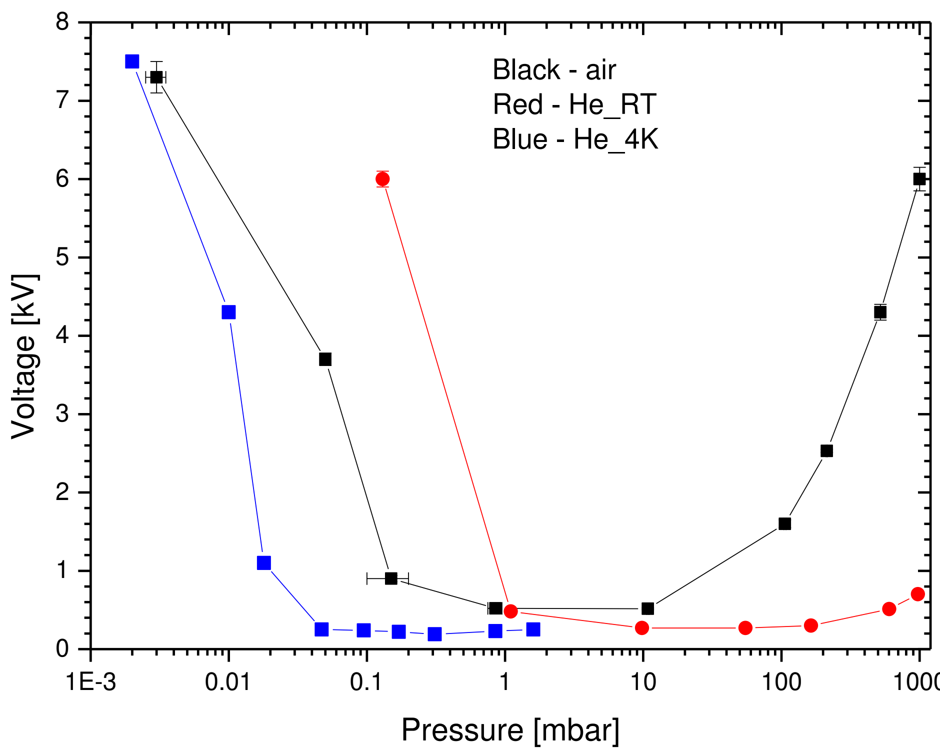
No problem with high el. field in vacuum

Low temp. cryostat with exachange He gas ?

Glow discharge at mBar pressure

Optimising pressure-voltage-tempearture condition for reliable temp & voltage control

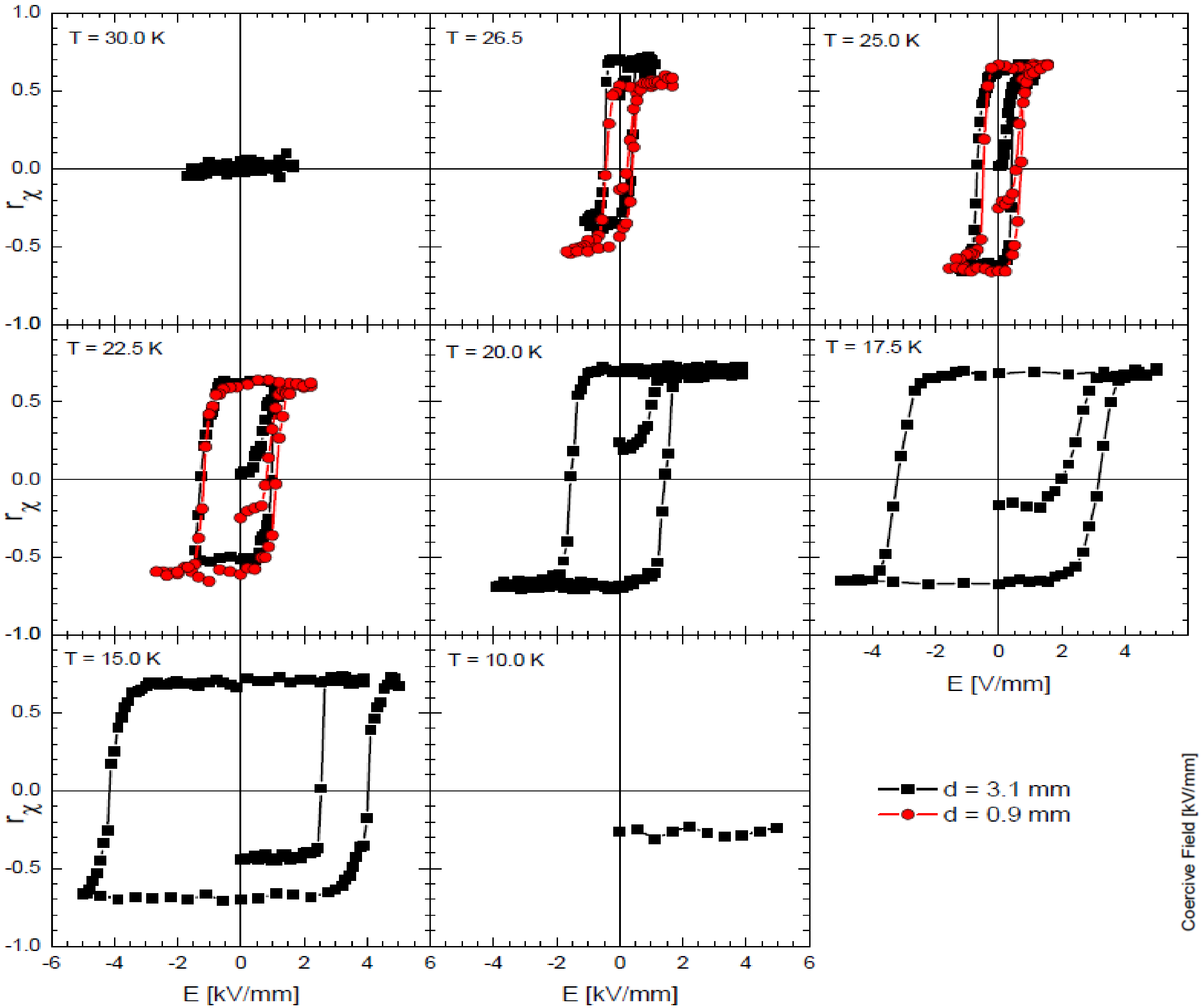




Valve control

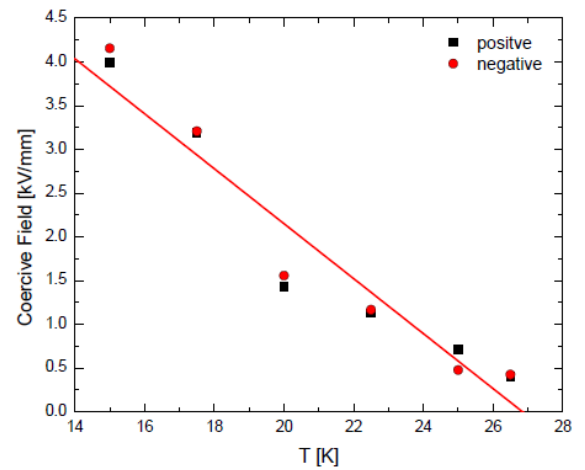
Switching magn. chirality in $TbMnO_3$

Electric field control of the magnetic structure chirality

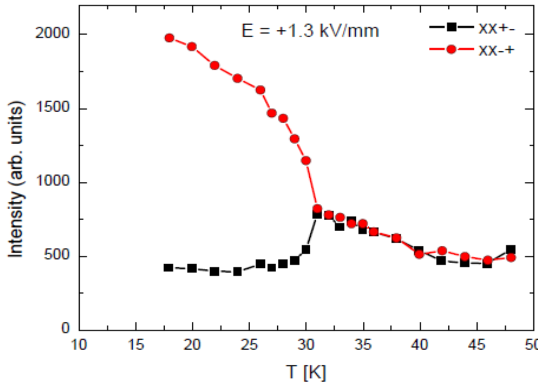


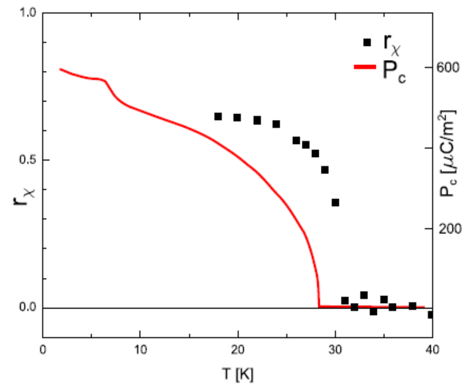
$$r_{chir} = \frac{\sigma_{xx}^{\uparrow\downarrow} - \sigma_{xx}^{\downarrow\uparrow}}{\sigma_{xx}^{\uparrow\downarrow} + \sigma_{xx}^{\downarrow\uparrow}}$$

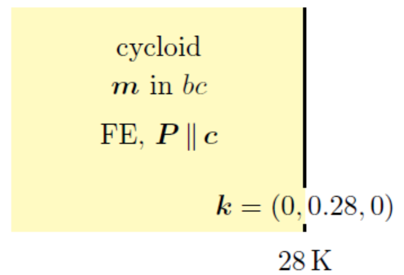
M. Baum PhD Thesis, University Cologne 2013

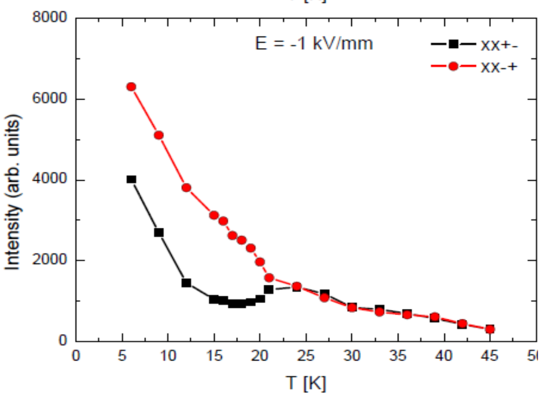


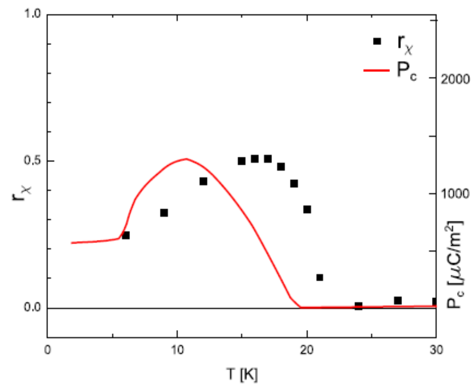
$TbMnO_3$ & $DyMnO_3$

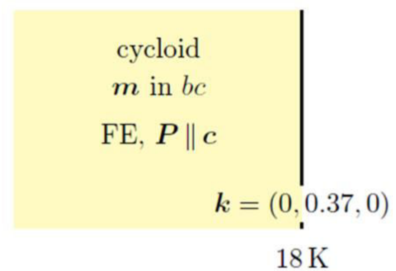




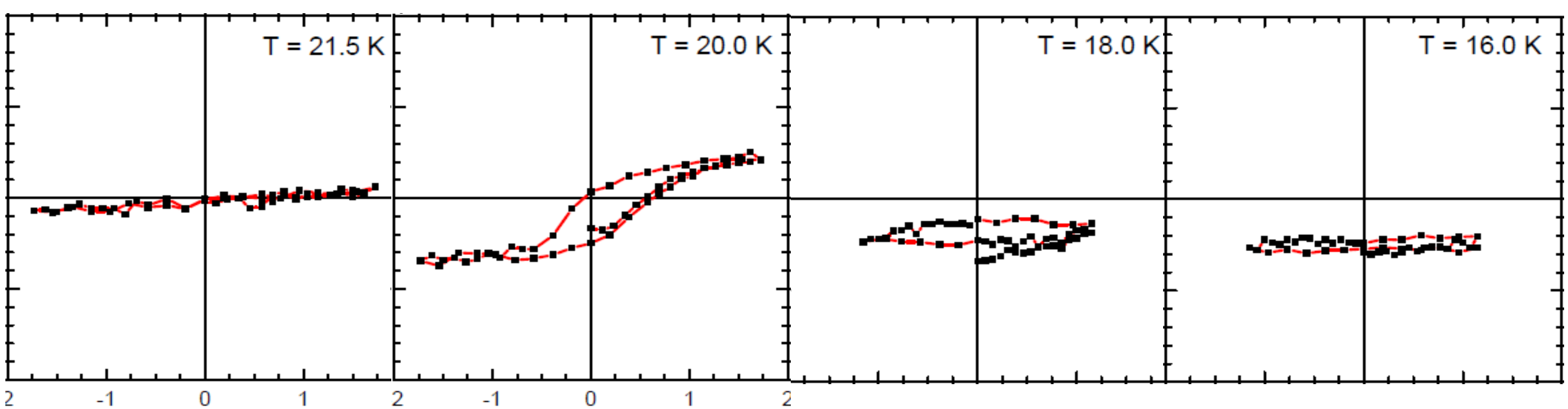




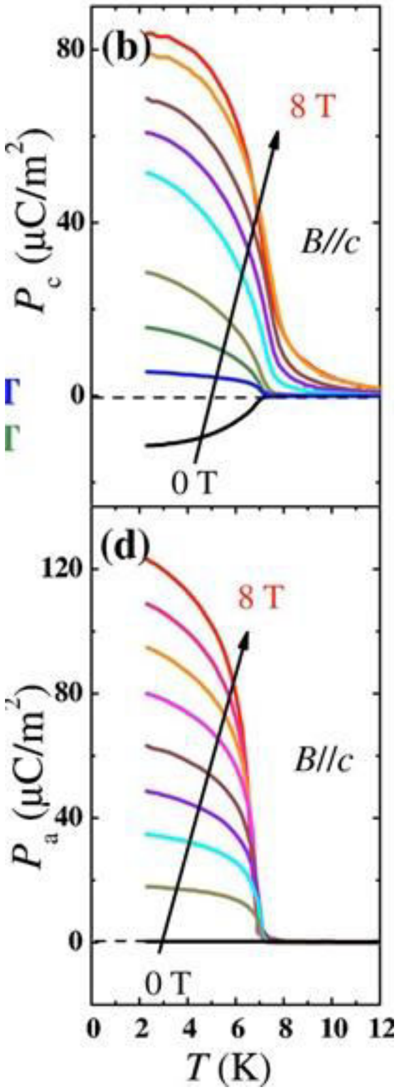




Reversing of chiral domains in $DyMnO_3$ is only possible close to the phase transition.



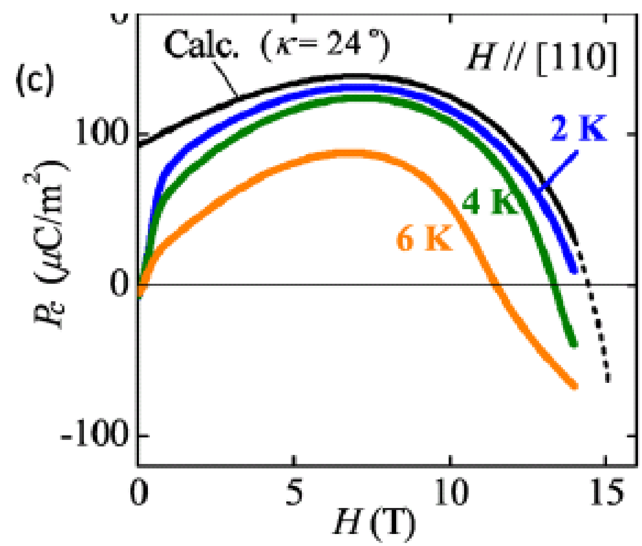
SNP application example: multiferroic $Ba_2CoGe_2O_7$



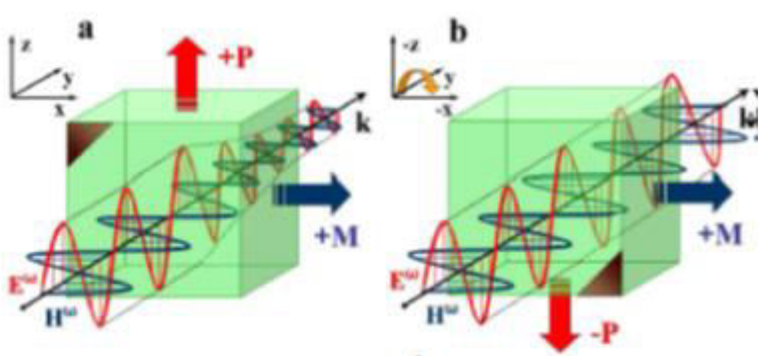
H.T. Yi et al., Appl. Phys. Lett. (2008)

A smooth rotation of the EP with the magnetic field rather than a sudden flip, is quite unique and cannot be explained by well-accepted spin-current model or by an exchange striction mechanism conventional for other multiferroics.

H. Murakawa et al., Phys. Rev. Lett. (2010)



A novel spin-dependent hybridization mechanism with a metal-ligand hybridization modified by local spin configurations through spin-orbit coupling.

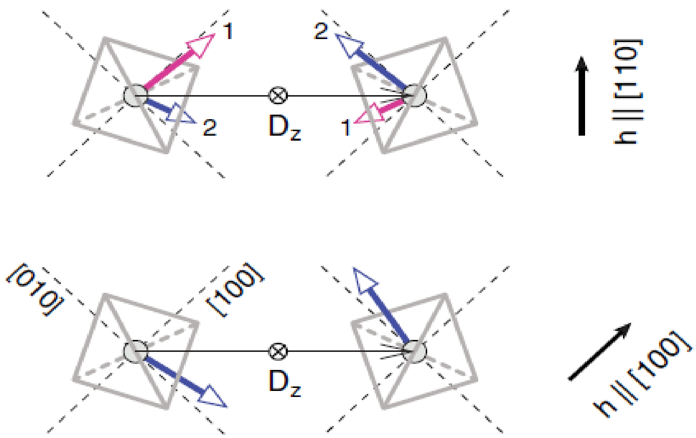


I. Kézsmárki et al. PRL 2011

Giant directional dichroism of terahertz light in resonance with magnetic excitations (electromagnons) .

Two different DM interactions along [1-10] and [001] ?

SNP application example: multiferroic $Ba_2CoGe_2O_7$



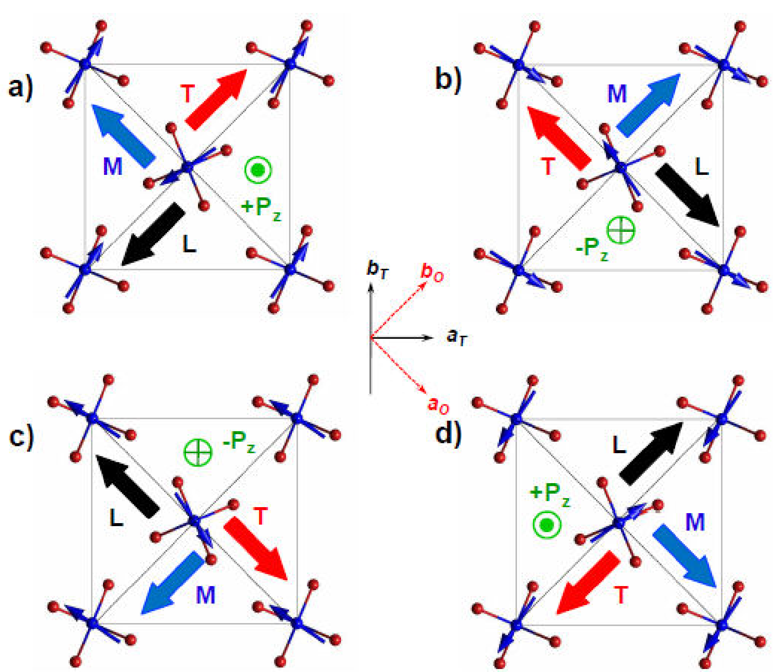
J. Romhanyi et al., Phys. Rev. B. 84, 224419 (2011)

Similar spin-dependent hybridization model, but adding in the Hamiltonian an exchange anysotropy and antiferroelectric polarization-polarization term. Mean field calculations.

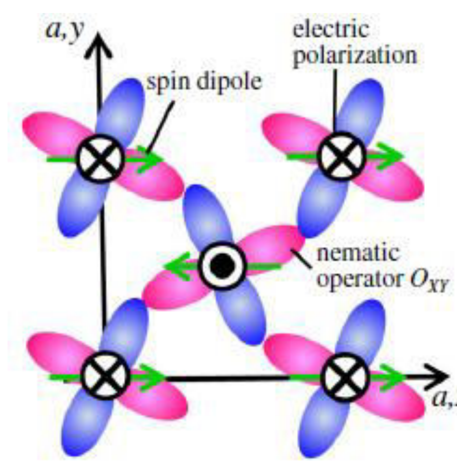
P. Toledano, at al. Phys. Rev. B **84**,094421 (2011)

Spontaneous toroidal moment, collinear to antiferromagnetic vector L = s_1-s_1 along a-plane.

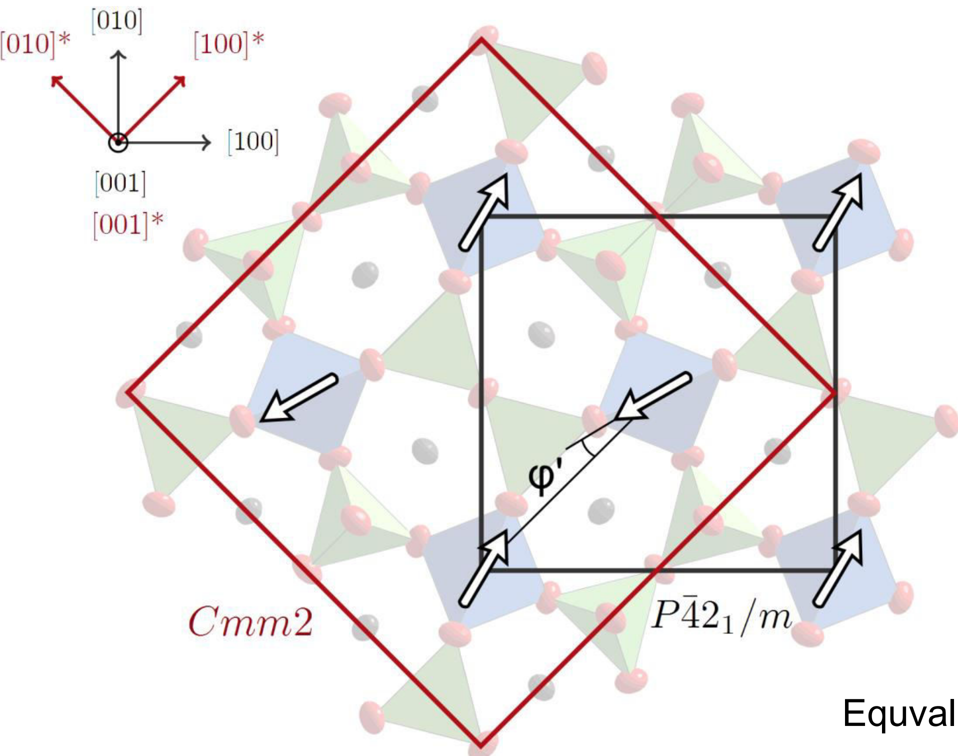
$$\vec{T} = \hat{\nu}(\vec{M} \times \vec{P})$$



Spin-nematic interaction
M. Soda et al. PRL 2014



Magnetic structure of Ba2CoGe2O7



Structure at RT & 90 K @ BM1 SNBL ESRF
Nonpolarised neutron diffraction on SCD HEiDi @ MLZ

–Week orthorombicity in the crystal structure

–No structural phase transition at T_N

–Precise magnitude of the AFM ordered moment :

M = 2.9 ± 0.1 μB , FM along c

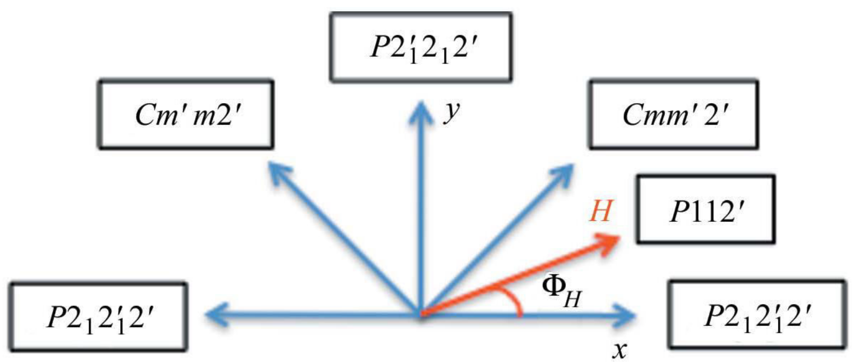
–No precise value vor canting φ' ≈ 8° ± 7° ???

V. Hutanu et. al Phys. Rev. B 86, 104401 (2012)

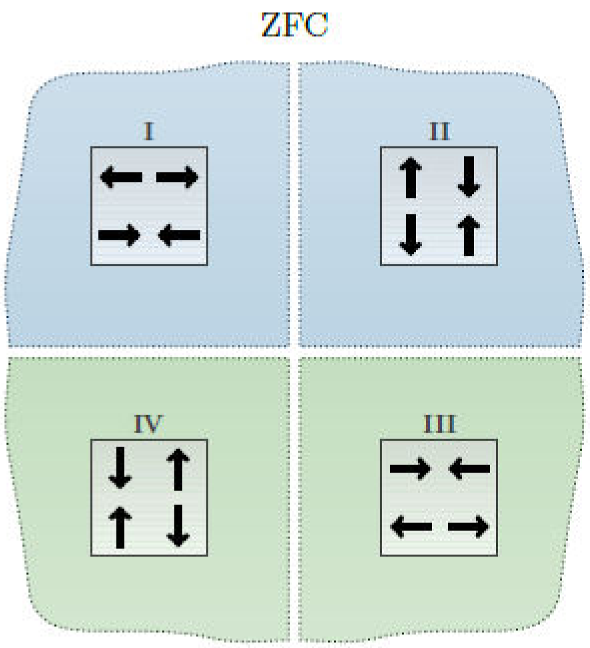
Equvalent AFM domains do not permit the unique solution ???

J.M. Perez-Mato, et al. Acta Cryst. A67, 264 (2011)

Different magnetic domains dependent on magnetic. moment direction



Example $Ba_2CoGe_2O_7$



$$\begin{bmatrix} xx & 0 & 0 \\ 0 & yy & 0 \\ 0 & 0 & zz \end{bmatrix}$$

Same ratio all domains

$$\begin{bmatrix} xx & 0 & a \\ 0 & yy & 0 \\ -a & 0 & zz \end{bmatrix}$$

Only domains type I ,II

$$\begin{bmatrix} xx & 0 & 0 \\ 0 & yy & b \\ 0 & b & zz \end{bmatrix}$$

Domains type I ,IV

$$\begin{bmatrix} xx & c & 0 \\ -c & yy & 0 \\ 0 & 0 & zz \end{bmatrix}$$

Domains type I ,III

$$\begin{bmatrix} xx & -c & 0 \\ c & yy & 0 \\ 0 & 0 & zz \end{bmatrix}$$

Domains type II ,IV

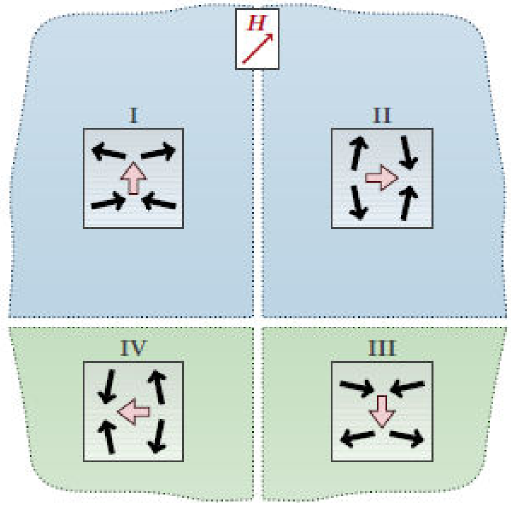
Example $Ba_2CoGe_2O_7$

	$\mathcal{P}_{ij}$	ZFC x'	y'	z'
Observed	x'	0.73(1)	0.01(2)	−0.06(4)
	y'	0.07(6)	0.76(2)	0.04(4)
	z'	0.04(2)	0.04(1)	0.76(4)
Calc100	x'	0.78	−0.01	−0.05
	y'	0.01	0.88	0.00
	z'	0.05	0.00	0.89
Calc110	x'	0.89	0.00	−0.03
	y'	0.00	0.94	0.00
	z'	0.03	0.00	0.95

Not cleare separation between the models in ZFC case

SNP application example: multiferroic $Ba_2CoGe_2O_7$

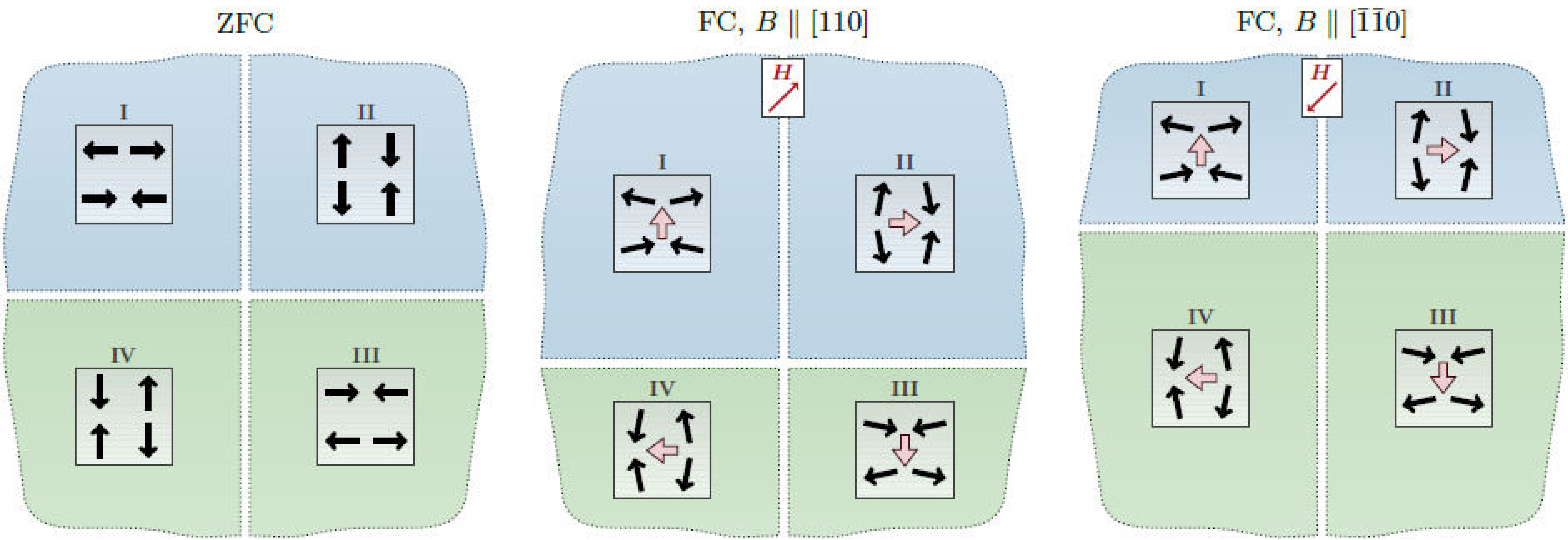
FC, $B \parallel [110]$



Field: 20 mT

	FC, $B \parallel [110]$		
	x'	y'	z'
Obs.	0.74(1)	0.03(2)	0.30(2)
	0.00(3)	0.82(2)	−0.02(6)
	−0.29(1)	0.00(3)	0.79(2)
(100)	0.78	0.01	0.22
	−0.01	0.88	0.00
	−0.22	0.00	0.89
(110)	0.83	0.00	0.17
	0.00	0.94	0.00
	−0.17	0.00	0.95

SNP application example: multiferroic $Ba_2CoGe_2O_7$



I	II	III	IV
28(3)%	24(3)%	21(3)%	27(3)%

I	II	III	IV
37(4)%	38(4)%	12(3)%	13(3)%

I	II	III	IV
11(3)%	13(3)%	36(4)%	40(4)%

Statistic distribution for the ZFC sample

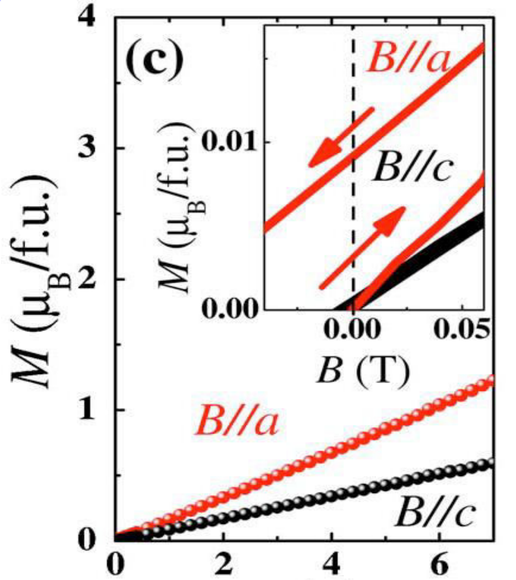
Symmetric picture (no preferential domain), same energies

Reversible ratio by field reverce,

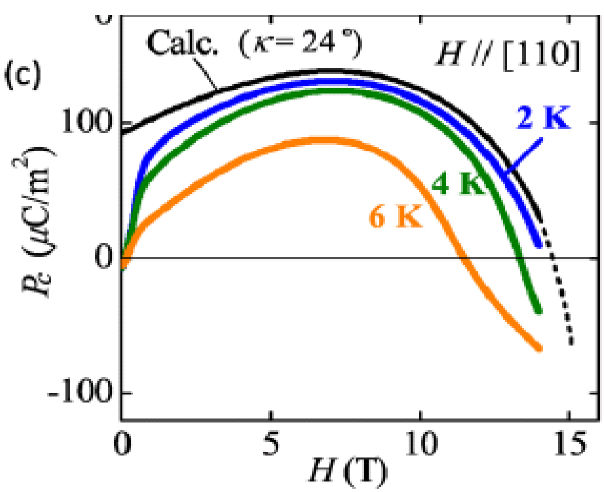
No memory effect after heating at 15 K

V. Hutanu, A. Sazonov et. al Phys. Rev. B 89, 64403 (2014)

SNP application example: multiferroic $Ba_2CoGe_2O_7$



Yi et al. Appl. Phys. Lett 2008



- Linear dependence domain ratio – field.

Field necessary to create double domain structure could be estimated: 40 mT.

-This is in disagreement with the hybridisation model of Murakawa (PRL 2010), where deviation at 1 T has been atributed to the domain formations.

- No dependence on the electric field direction along c

toroidal moment (Toledano, PRB 2011)

-Our results do not support the nematic interaction model of Soda (PRL 2014), neither on spin orientation, no on domain flop

-Best agreement to the Romhanyi (PRB 2011) model of hybridisation and antiferroelectric coupling

Examples: (Cr_2O_3)

Magnetic Compounds and Alloys Alan H. Morrish, Chairperson

Determination of the absolute magnetic moment direction in Cr_2O_3 using generalized polarization analysis

F. Tasset and P. J. Brown
Institut Laue Langevin, BP 156X, 38042 Grenoble, France
J. B. Forsyth
Neutron Division, Rutherford Appleton Laboratory, Chilton, Oxford, England

Cryopad I - 1988

A study of magnetoelectric domain formation in Cr_2O_3

P J Brown†, J B Forsyth‡ and F Tasset†
† Institut Laue–Langevin, BP 156 38042, Grenoble Cédex, France
‡ Rutherford Appleton Laboratory, Chilton, Oxon OX11 0QX, UK

Cryopad II - 1998

Determination of the magnetization distribution in Cr_2O_3 using spherical neutron polarimetry

P J Brown[1,2], J B Forsyth[3], E Lelièvre-Berna[1] and F Tasset[1]

Cryopad II - 2002

Magneto-electric coupling, colinear antiferromagnetic structure with zero propagation vector, polarisation independent cross section; 180° AFM domains with opposide ME effect.

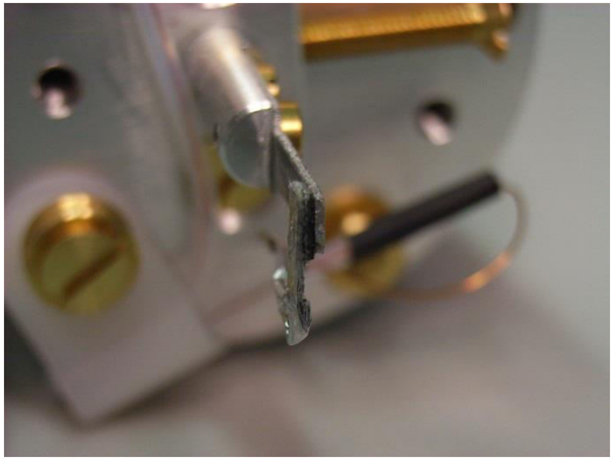
Solution: Magnetic and electric fields applied above T_N to disbalance the 180° domains

Examples: (Cr_2O_3)

If the moments are parallel to polarisation z

$$\boldsymbol{P} = \begin{pmatrix} \beta & \eta\xi & 0 \\ -\eta\xi & \beta & 0 \\ 0 & 0 & 1 \end{pmatrix} \quad \text{with} \quad \begin{aligned} \beta &= (1-\gamma^2)/(1+\gamma^2) \\ \xi &= 2q_z\gamma/(1+\gamma^2) \\ \gamma &= \mathbf{M}_\perp(\mathbf{k})/N(\mathbf{k}) \end{aligned}$$

q_z is $+1$ if $\mathbf{M}(\mathbf{k})$ is parallel to $\mathbf{z}$ and -1 if it is antiparallel.



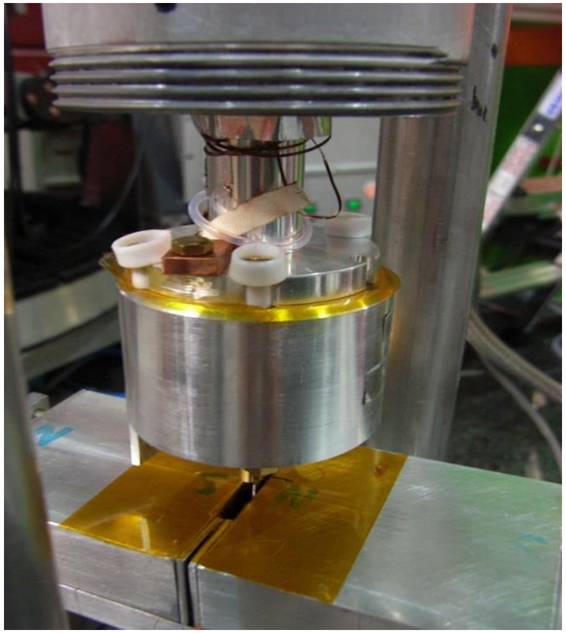
Simultaneously aplying of electric (1 kV/mm) and magnetic (0.9 T) fields at 320 K

Control of the 180° magnetic domains

Initial domain ratio: 57(4)/43(4)

Domain ratio after coling below T_N in paralell fields: 96(5)/4(5)

Antiparralel fields: 3(4)/ 97(4)



El. filed up to 10 kV and magnetic field up to 7.5 T are available on POLI

Conclusions

- SNP using third generation polarimeter Cryopad and new ^{3}He spin filter polariser and analyser has been recently implemented on instrument POLI at MLZ Garching Germany.

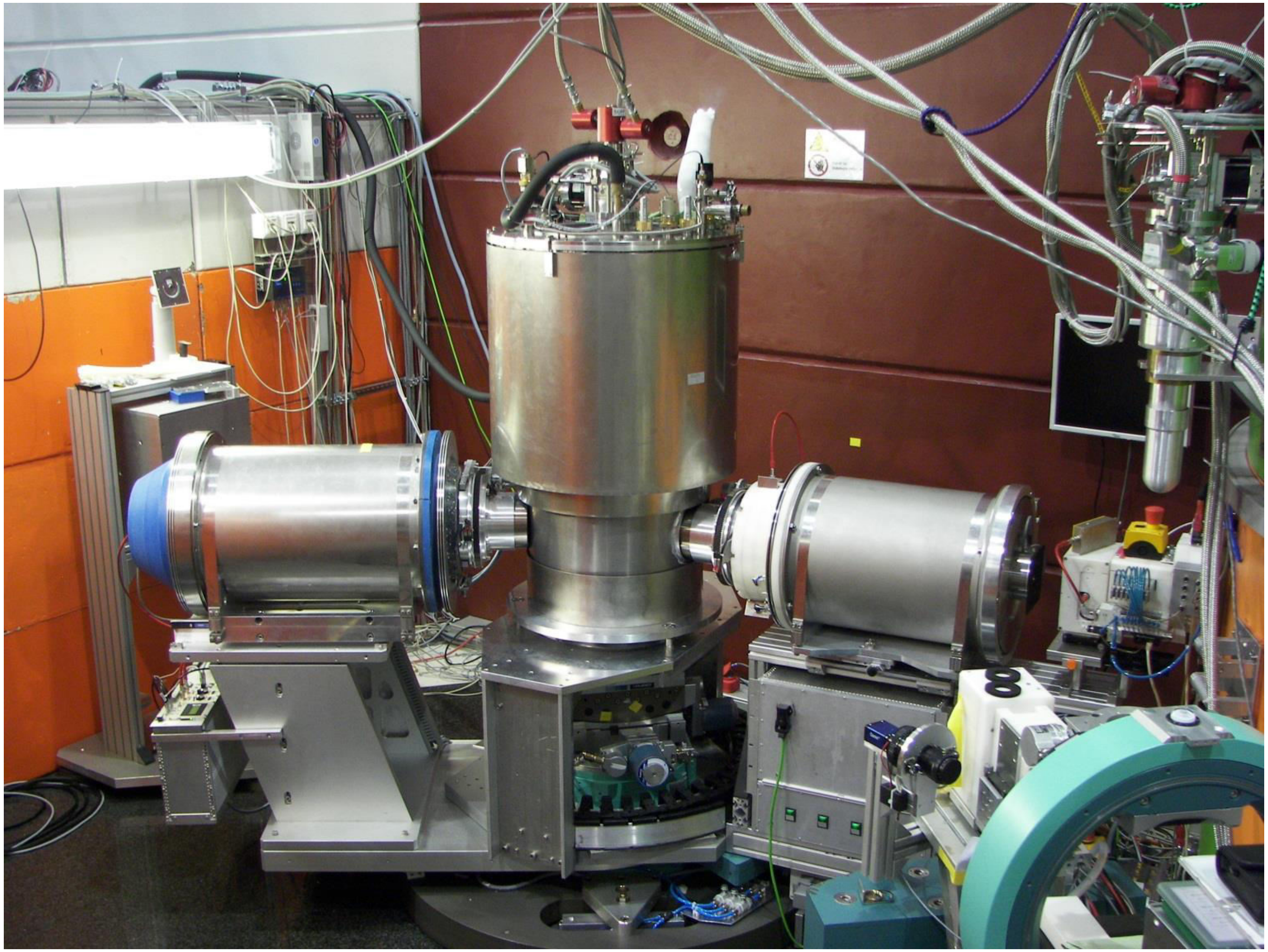
- This technique allowds a precise determining of the magnetic order in the ground state of the complex AFM structures. Also other types of structure like spiral (helical, cycloidal), spin dencity wave etc. can be determined.

- Different types of the magnetic domains in the sample (configuration, orientation, chiral, etc.) can be distingueshed, domain ratio determined and domain dynamics in dependance on external stimuli (e.g. electric and magnetic field) studied.

- Using SNP a collinear AFM order with the main moment pointing along (100) direction in orthorombic cell has been found in multiferroic $Ba_2CoGe_2O_7$. This result is in agreement to the Romhanyi (PRB 2011) model of hybridisation and antiferroilectric coupling between 2D layers.

- Last, but most important …….

You are welcome to use it



POLI with Cryopad (SNP) are available for external users

over the MLZ proposal system

Acknowledgements

Special thanks to:

A. Sazonov, M. Meven, W. Luberstetter, G. Roth - RWTH Aachen University

M. Baum, J. Stein, M. Braden - University of Cologne

H. Murakawa, Y. Tokura, - University of Tokyo

S. Bordács, I. Kézsmárki – University of Budapest

I.-H. Oh – KAERI Dajeong

D. Chernyshov – ESRF Grenoble

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D. Chernyshov – ESRF Grenoble

You for your attention !

If you know somebody interested in more than one of the following topics:

- Single crystal neutron diffraction,
- Polarised neutrons,
- Magnetic structures,
- Soffisticated Instrumentation,
- Bavarian Bier,

Please contact: vladimir.hutanu@frm2.tum.de /www.mlz-garching.de

3–year contract, expected to be started early in 2015