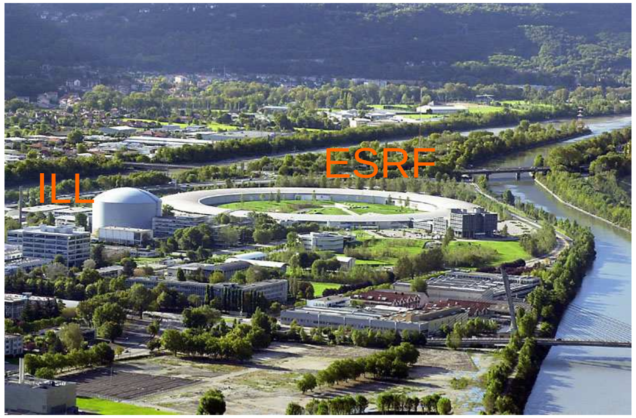
Réunion du GdR "Matériaux et Interactions en Compétition",
Roscoff – 7-10 Janvier 2013

Champs Magnétiques Pulsés
et
Diffusion des rayons X et des neutrons

Fabienne Duc

LNCMI-Toulouse



Grenoble

Collaborators

LNCMI, Toulouse, France

J. Béard, J. Billette, X. Fabrèges, P. Frings,
W. Knafo, M. Nardone, J. P. Nicolin, A. Zitouni
B. Vignolle, G. Rikken

X-ray experiments

- ESRF, Grenoble, France

  C. Detlefs, T. Roth, L. Paolasini, W. Crichton (ID06)
  P. Van der Linden (ESRF)
  C. Strohm, O. Mathon, S. Pascarelli (ID24)

Neutron experiments

- Institute for Material Research
  Tohoku University, Japan

  H. Nojiri, S. Yoshii, K. Ohoyama

- Ibaraki University, Japan

  K. Kuwahara

- Japan Atomic Energy Agency
  (JAEA, Tokai), Japan

  M. Matsuda

- INAC/SPSMS/MDN, CEA-Grenoble

  L. P. Regnault (CRG IN22@ ILL)
  D. Aoki, F. Bourdarot, J. Flouquet

- Institut Laue Langevin, Grenoble

  E. Lelièvre-Berna, B. Rollet, X. Tonon

- Institut Néel, Grenoble, France

  J.E. Lorenzo

Outline

- **Introduction - Motivations**
- **High pulsed magnetic field**
- **X-ray diffraction in pulsed fields**
  - Pulsed magnet with conical bore for powder diffraction
  - Split-pair magnet for single crystal diffraction
  - Perspectives
- **Neutron diffraction in pulsed fields**
  - Neutron experimental configuration
  - Application to the frustrated spinel $CdCr_2O_4$
  - Metamagnetic transition in 4%Rh-URu_2Si_2
  - Perspectives

Motivations: why ? Outline of scientific cases

Quantum magnetism
- magnetization plateaus
- quantum critical points

High T_c
- low temperature normal state studies
- competing phases in the cuprates

Magnetic oxides
- charge and orbital ordering
- structural transitions

Heavy fermions
- metamagnetic transitions (URu_2Si_2…)

Multiferroics
- magnetically driven charge displacements

and lots more!!!

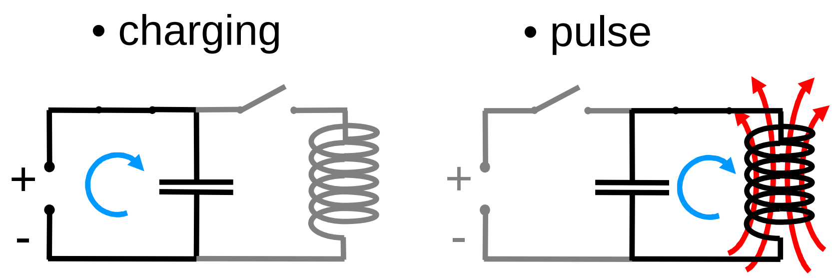
Synchrotron and neutron radiation are powerful and universal tools to determine magnetic and structural properties as well as dynamic modes of condensed matter

The combination of synchrotron and neutron radiation with high magnetic fields open many new research opportunities.

Pulsed magnetic fields

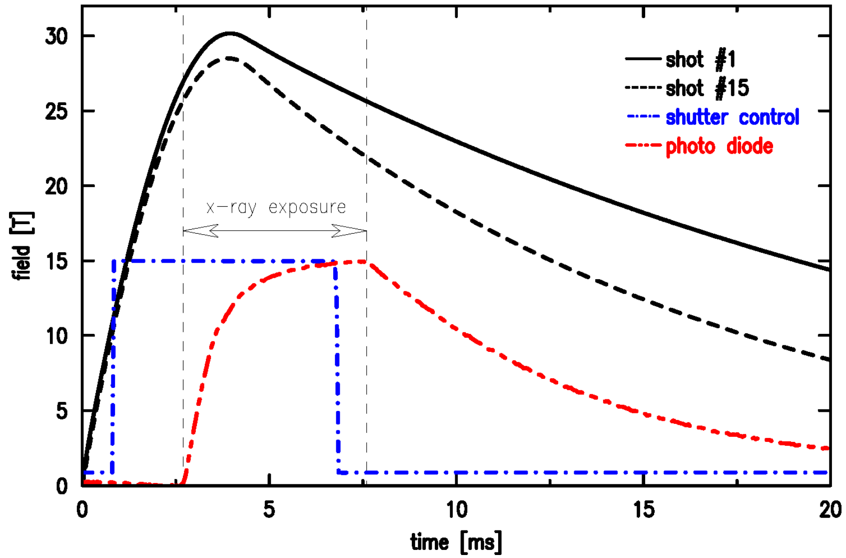
Advantages

- Scalable
  Trade-off between pulse length
  and energy/size of installation
- Well-known technology
- Quite easy (and cheap) to reach 30 T
  technology for 60 T is mature
- Trade-off between max. field
  and duty cycle/repeat frequency



Disadvantages

- Duty cycle:
  1 msec every 10 sec
  30 msec every 5 min…
- Eddy currents ⇒ heating
- Vibrations induced by magnetic forces



State of the art

Synchrotron pulsed field devices developed across the world during the last 8 years:

Japan: Spring-8

- Nojiri et al. (IMR, Tohoku University, Sendai)
  Miniature coil, 40 T (~ 7-8 ms), single crystal diffraction, X-ray absorption spectroscopy (XAS), X-ray magnetic circular dichroism (XMCD)

- Katsumata, Kindo and Narumi et al. (ISSP, Tokyo University, Japan)
  Split-pair coil, 40 T (~ 27 ms), single crystal diffraction

USA: APS

- Collaboration Z. Islam and Nojiri et al.
  Split-pair and solenoid minicoil, 30 T (~ 7-8 ms)
  Powder and single crystal diffraction

France: ESRF

- Collaboration between LNCMI, ESRF, Institut Néel and INPAC (Leuven)
  Classical solenoid coil, 30 T (~ 30-100 ms), powder diffraction, XAS, XMCD
  Coil with conical bore, 30 T (~ 30-100 ms), powder diffraction
  Split-pair coil, 30 T , (~ 60 ms), single crystal diffraction
  + ESRF dedicated beamline (ID06) C. Detlefs, T. Roth

- ESRF, P. Van der Linden, O. Mathon, C. Strohm
  Miniature coil, 30 T (~ 1 ms), XAS, XMCD, nuclear forward scattering

State of the art

Neutron pulsed field devices developed across the world during the last 8 years:

Japan: JAEA
- Nojiri et al.
  Miniature coil, 30 T (~ 1 to 10 ms), single crystal diffraction

J-PARC
- Nojiri et al.
  Miniature coil, 50 T (~ 1 to 10 ms), single crystal diffraction

England: ISIS
- Collaboration with Nojiri et al.

USA: SNS, Oak Ridge
- Collaboration with Nojiri et al.

France: ILL
- Collaboration between Nojiri el al. (IMR), LNCMI-T and CEA-SPSMS
  Miniature coil, 30 T (~ 7-8 ms), single crystal diffraction
- ANR project: Dec. 2010- Dec 2014
  Collaboration between LNCMI-T, CEA-SPSMS, ILL and Inst. Néel
  Coil with conical bore, 40 T (~100 ms), single crystal diffraction

Neutron DC fields projects under development

Germany: HZB, Berlin
- Collaboration with NHMFL (Florida, USA)
  Hybrid magnet, 25 T, elastic and inelastic scattering

Mobile power supplies (LNCMI-T)

- 2 storage units + 1 charger/control unit
- 2 polarities
- Charging and commutation in only few min (< 3 min)

2 versions

290 kJ
1 mF, 24 kV, 5 m^3, 2.8 t

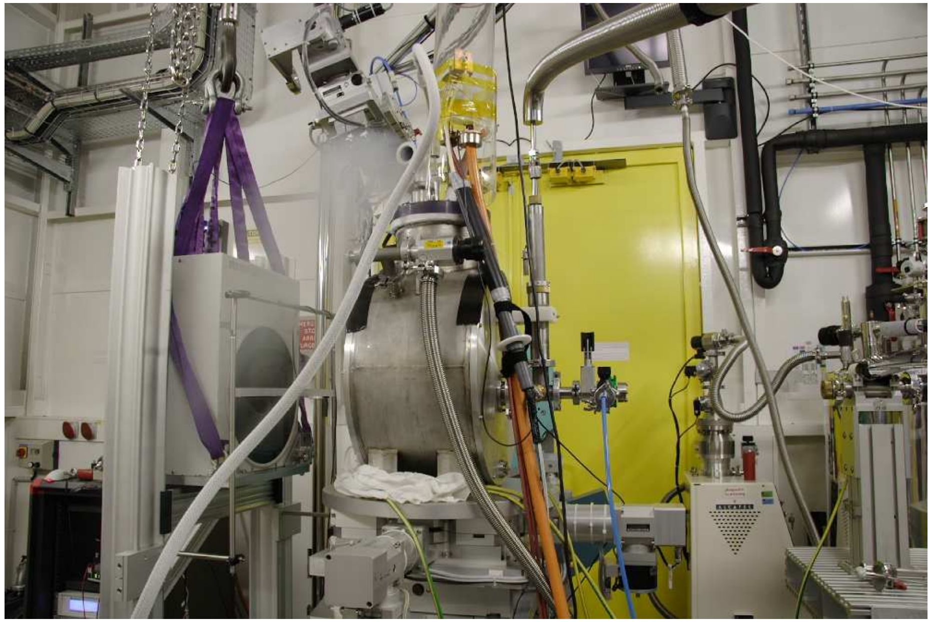
1.15 MJ
4 mF, 24 kV, 11 m^3, 5 t

and will be available in 2 x 3 MJ in 2013


Daniel Michon, Artechnique Grenoble

X-ray diffraction in pulsed fields

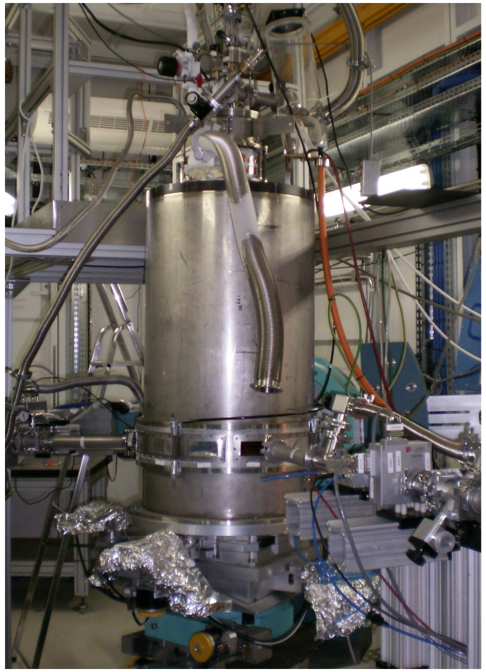
Collaboration: LNCMI-Toulouse, ESRF-Grenoble



30 T magnet with conical bore on ID20@ESRF

LNCMI capacitor bank
on ID06@ESRF

4 mF, 24 kV, 1.15 MJ,
11 m^3, 5 t

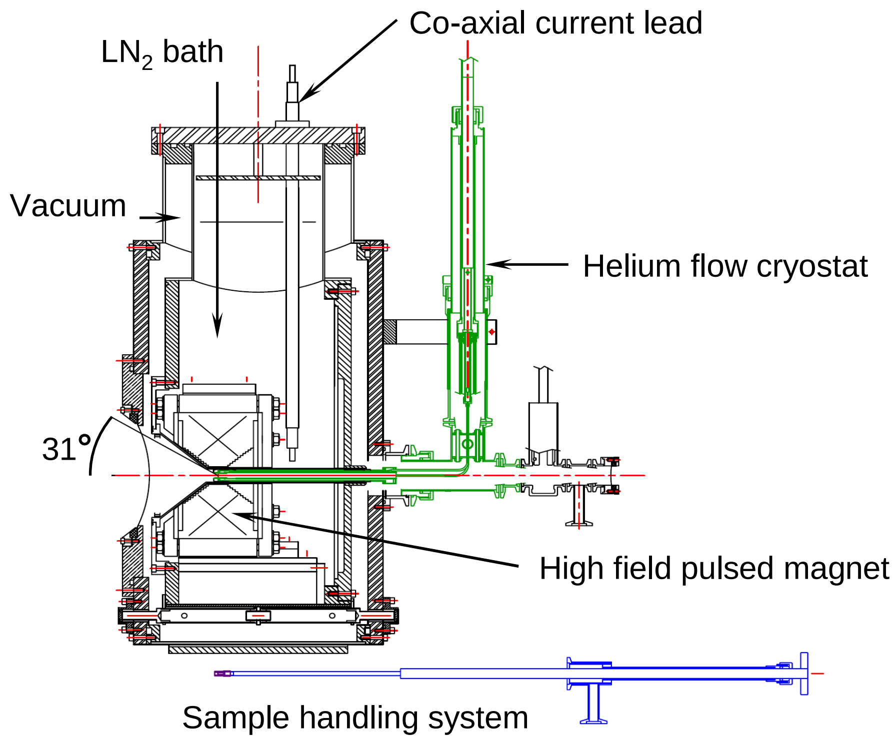


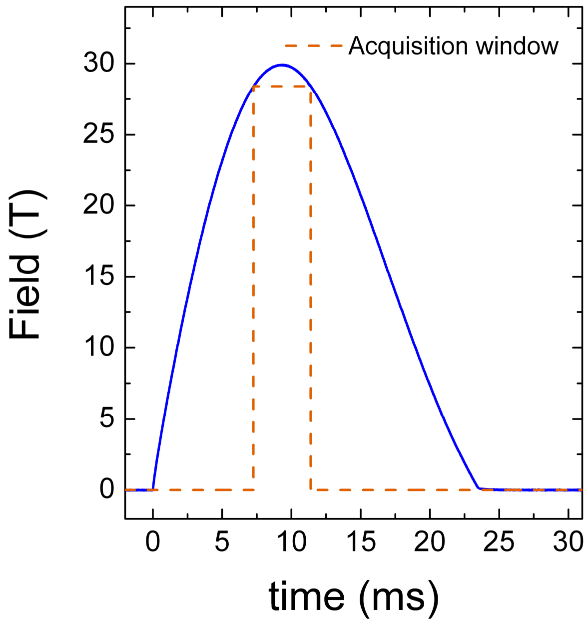
30 T split pair magnet
on ID06@ESRF

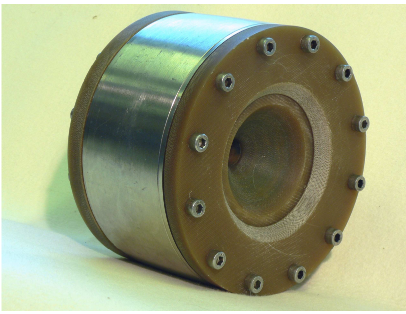
- Magnets and cryostats: LNCMI-Toulouse
- Pulsed field generator: LNCMI-Toulouse
- Synchrotron beamlines: BM26, ID20, ID06 @ ESRF

30 T pulsed magnet and cryogenics for X-ray powder diffraction

- Maximum field **30 T**
- Pulse duration (total) **23 ms**
- Repetition rate at B_{max} **6 pulses/hour**
- Geometry **conical ±31°**
  B // incident beam
- Sample diameter 4 mm
- Sample temperature **5-250 K**



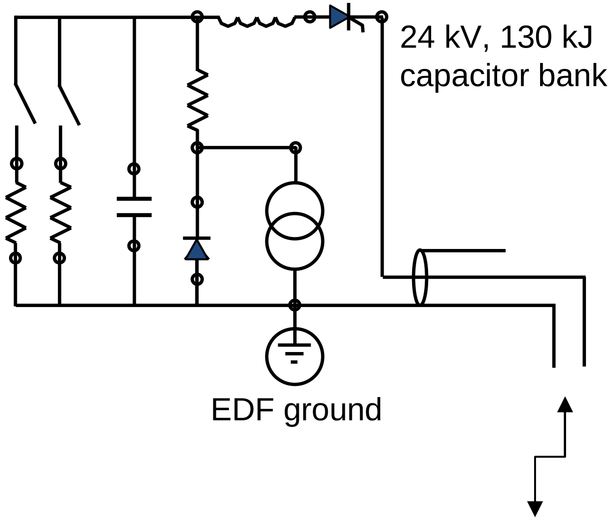


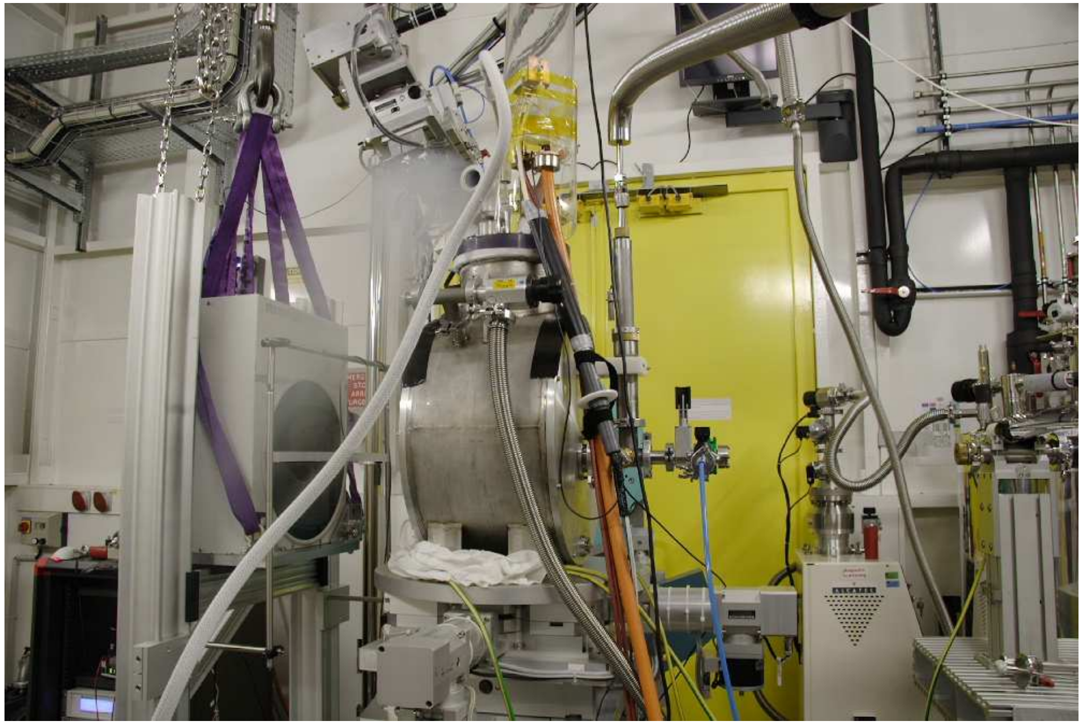


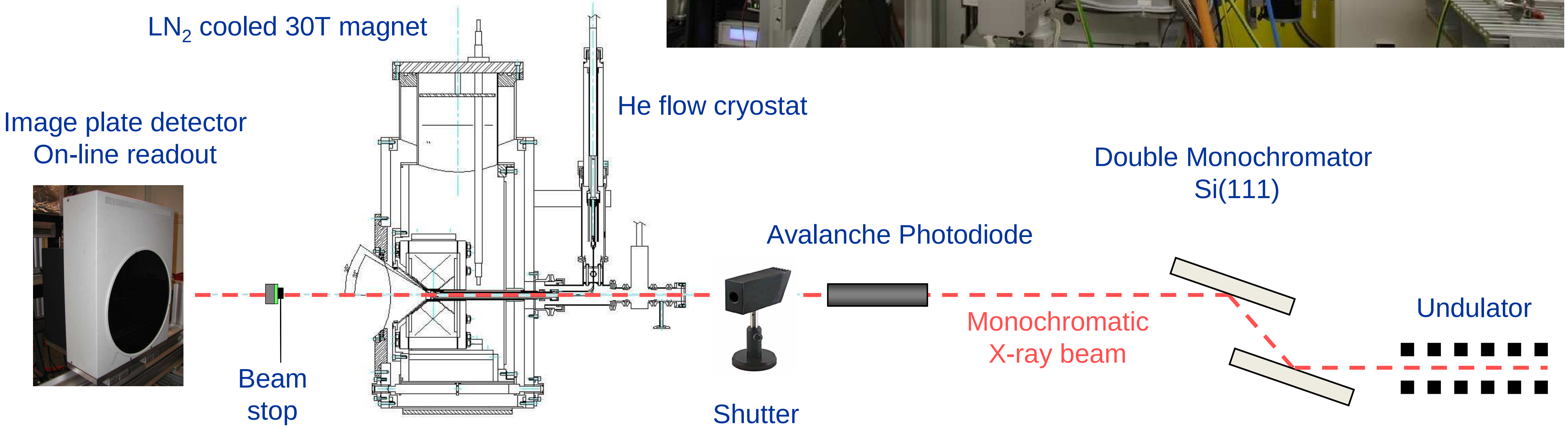
- Powder embedded in a polymer matrix
  to suppress grain movement
  to improve thermal contact

Billette *et al.*, Rev. Sci. Instrum. 83, 043904 (2012)

High field X-ray powder diffraction on ID20 (ESRF)



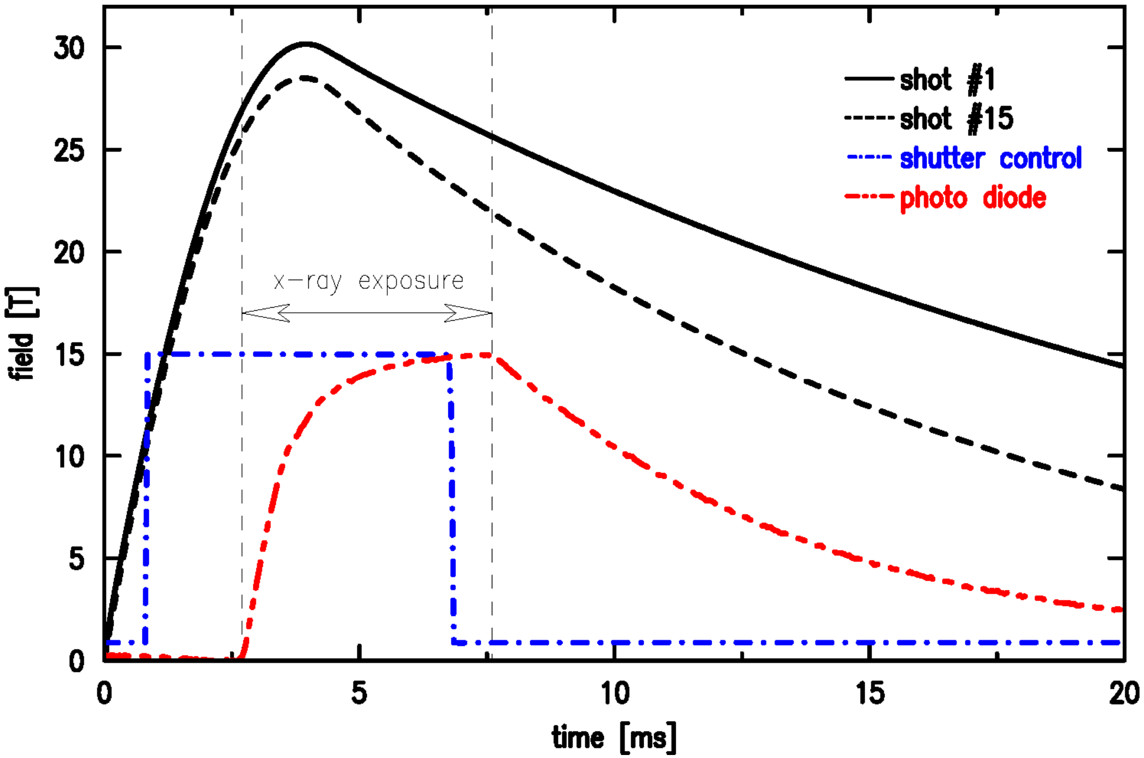




Data acquisition with image plate detector

Image plate detector (MAR 345)

- Exposed by opening a fast shutter near maximum magnetic field, integrating ca. 2-5 ms
- Simple, robust exp.
- Timing with fixed delays
- One (average) field/spectrum



Data acquisition with image plate detector

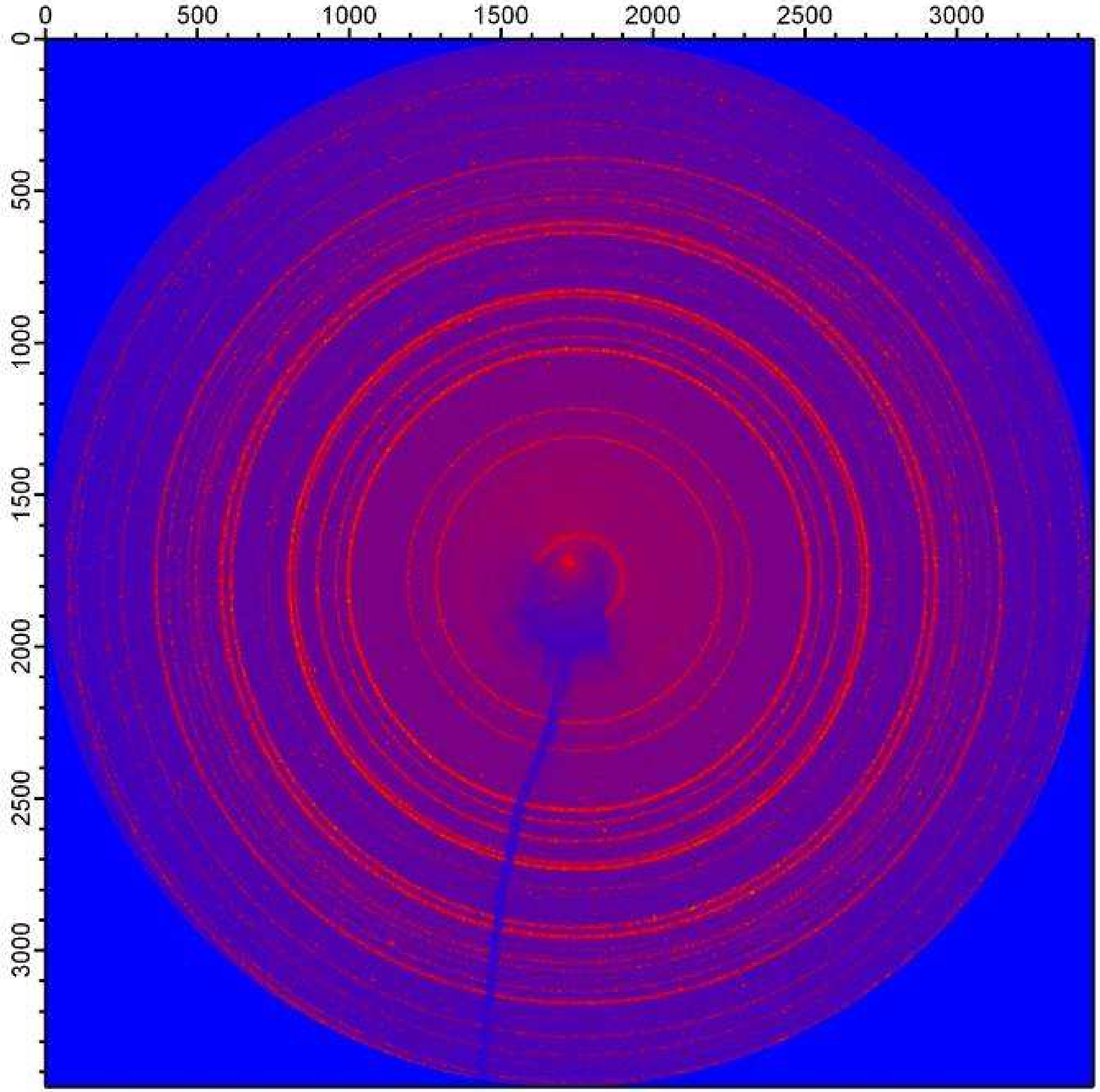
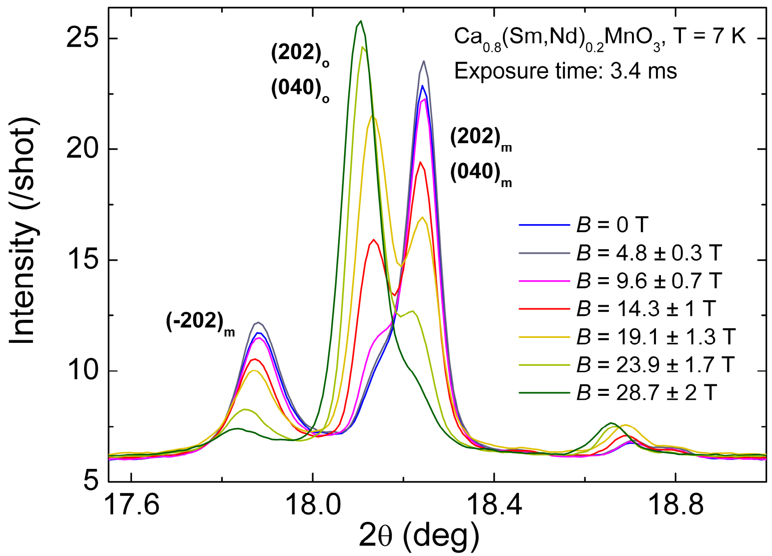


Image plate detector (MAR 345)

- Exposed by opening a fast shutter near maximum magnetic field, integrating ca. 2-5 ms
- Simple, robust exp.
- Timing with fixed delays
- One (average) field/spectrum

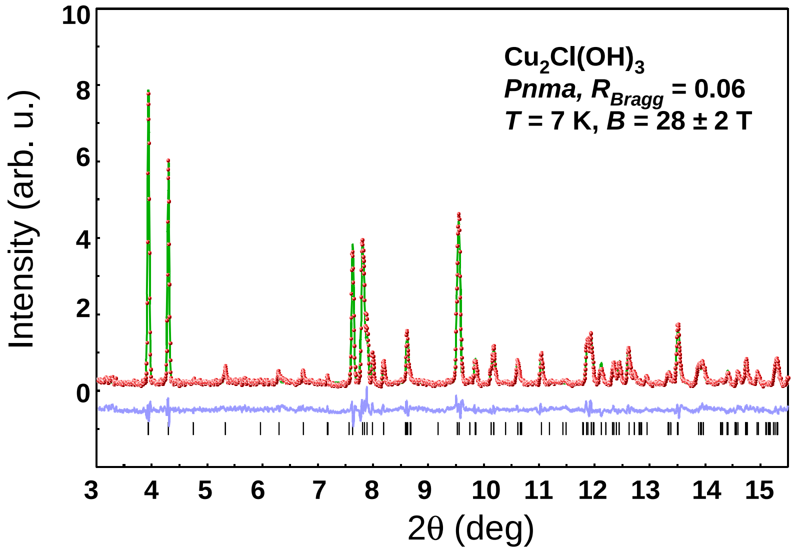
High magnetic field X-ray powder diffraction

Duc *et al.*, Phys. Rev. B 82, 054105 (2010)



- ⇒ **Field induced structural transition**
- ⇒ **Group-subgroup transition**
- ⇒ **Magnetostriction**

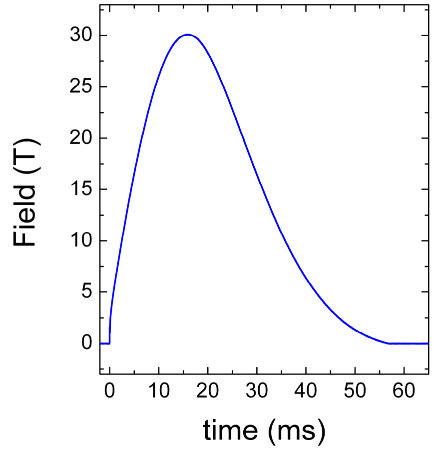
- ⇒ **Rietveld analysis possible**
- ⇒ **cell parameters + internal parameters**



Billette *et al.*, Rev. Sci. Instrum. 83, 043904 (2012)

30 T split-pair magnet for single-crystal X-ray diffraction

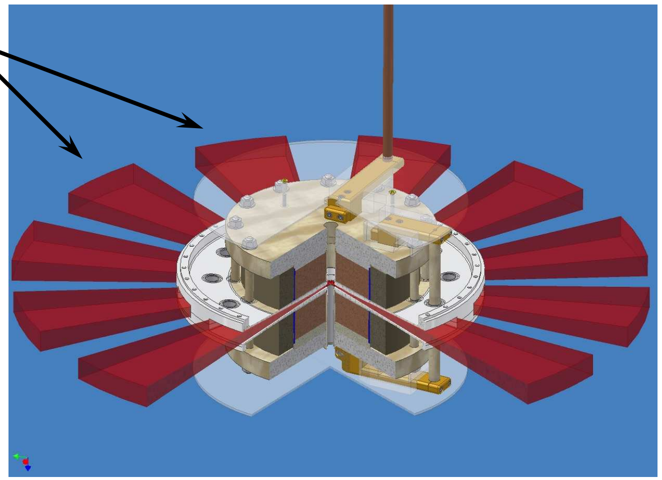
- Maximum field **30 T**
- Pulse duration (total) **57 ms**
- Repetition rate at B_{max} **2 pulses/hour**
- Sample temperature **1.5-250 K**

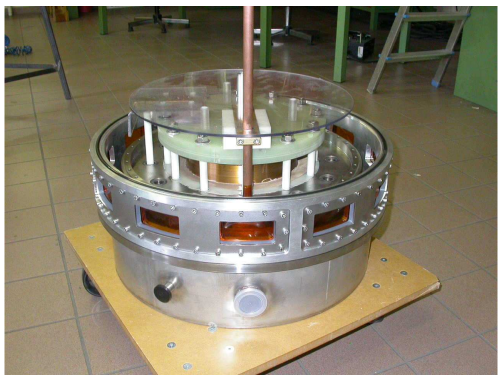


Wide angle space accessible

Split coil with ***B*** ⊥ **X-ray beam**

H

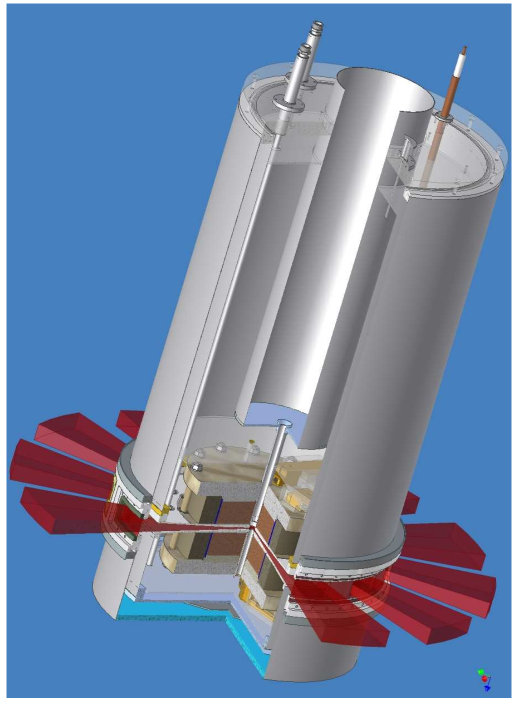




- Single crystal diffraction

Transmission or reflection geometry

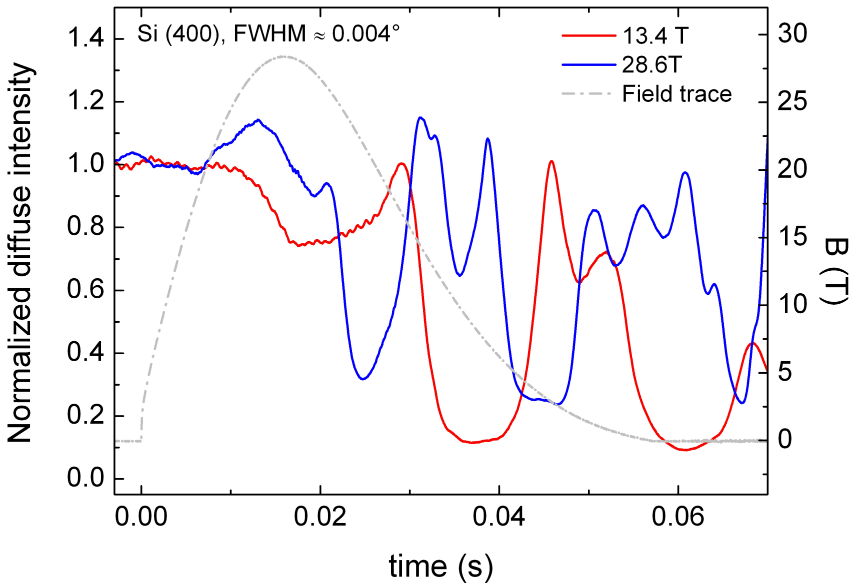
Grazing incidence

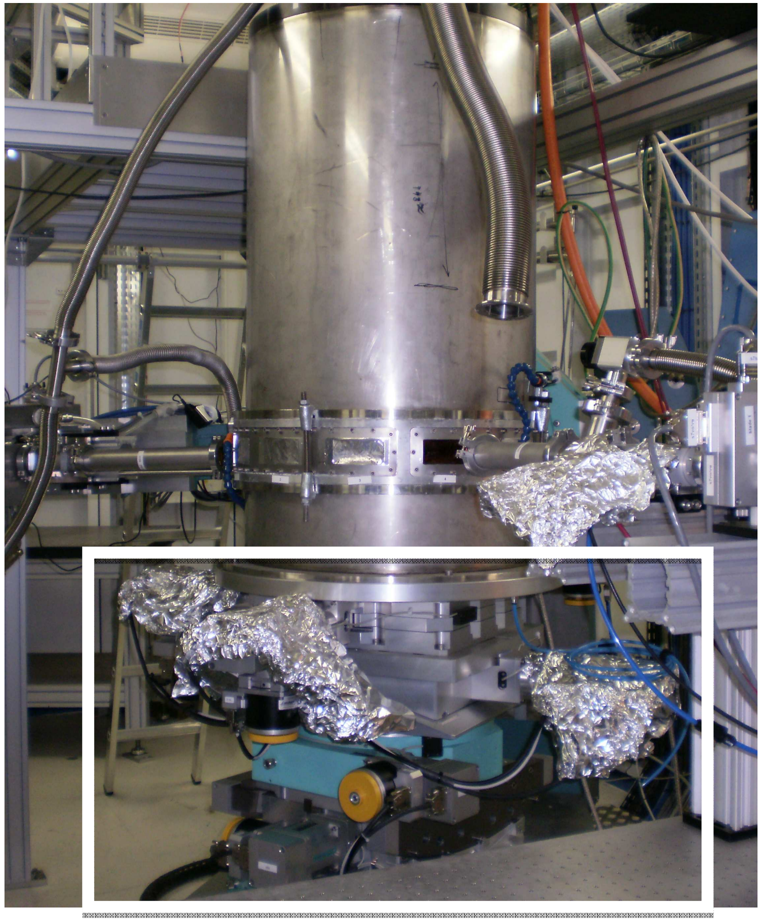


High field single-crystal diffraction on ID06@ESRF

- Single crystal diffraction in monochromatic beam
  - Highest resolution
  - Strongest absolute signal
  - Best signal/noise ratio

- ID06: angular resolution > 10^{-3} deg





⇒ **Sample rotation > 3.10^{-3} deg at 30 T**

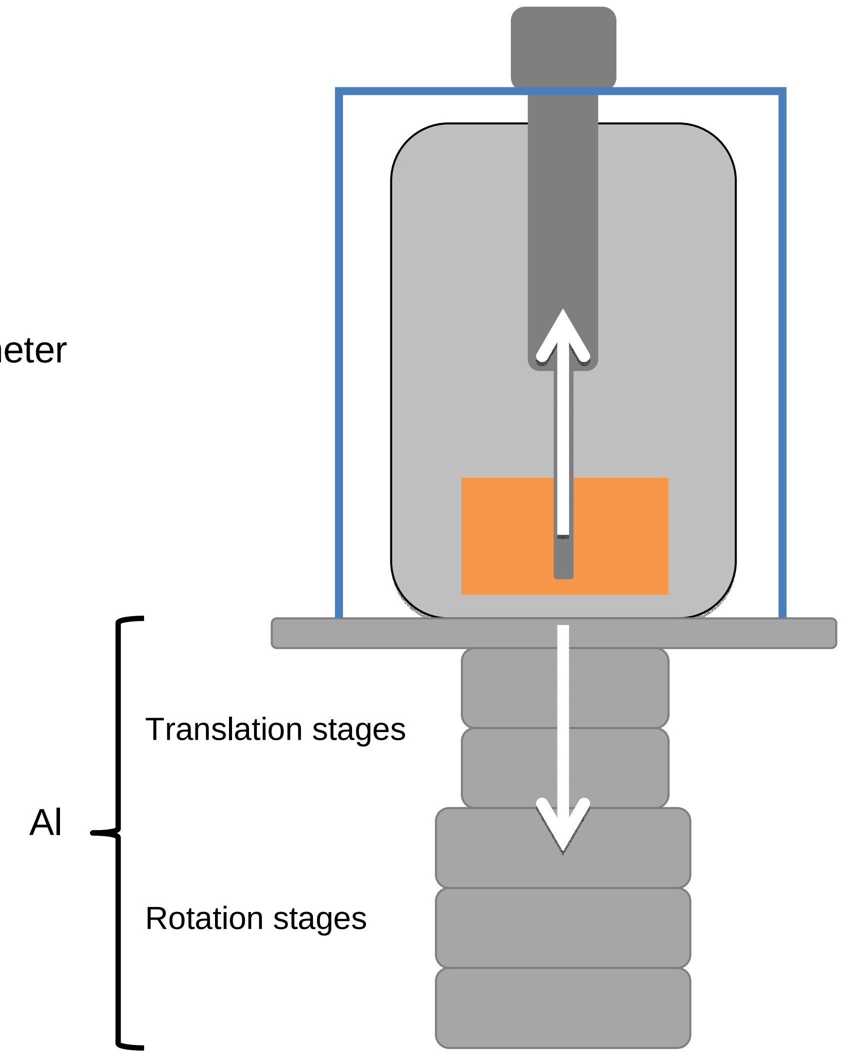
Vibration sensitivity

Eddy currents in Al parts of the diffractometer

Goniometer plate (diam. 1m)

F = 9.6 kN

Repulsive induced forces in coil and diffractometer



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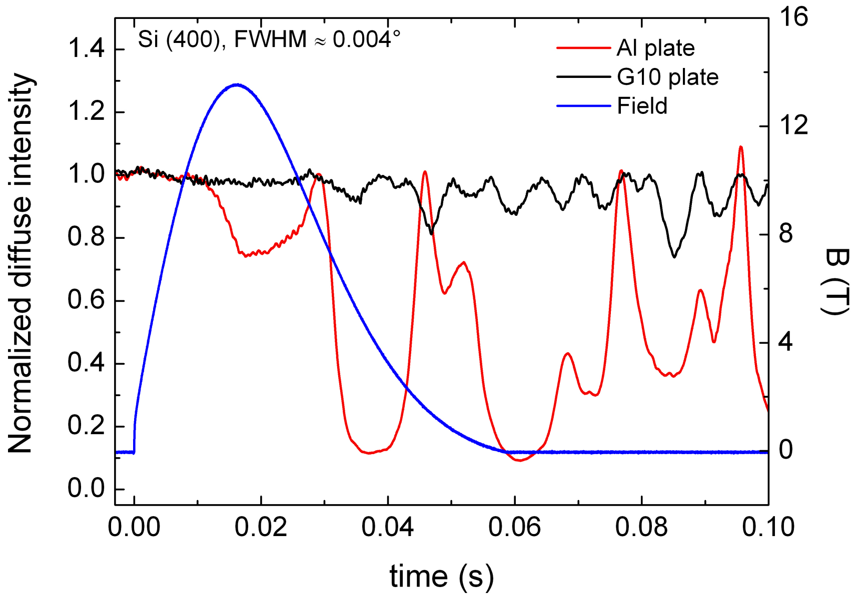

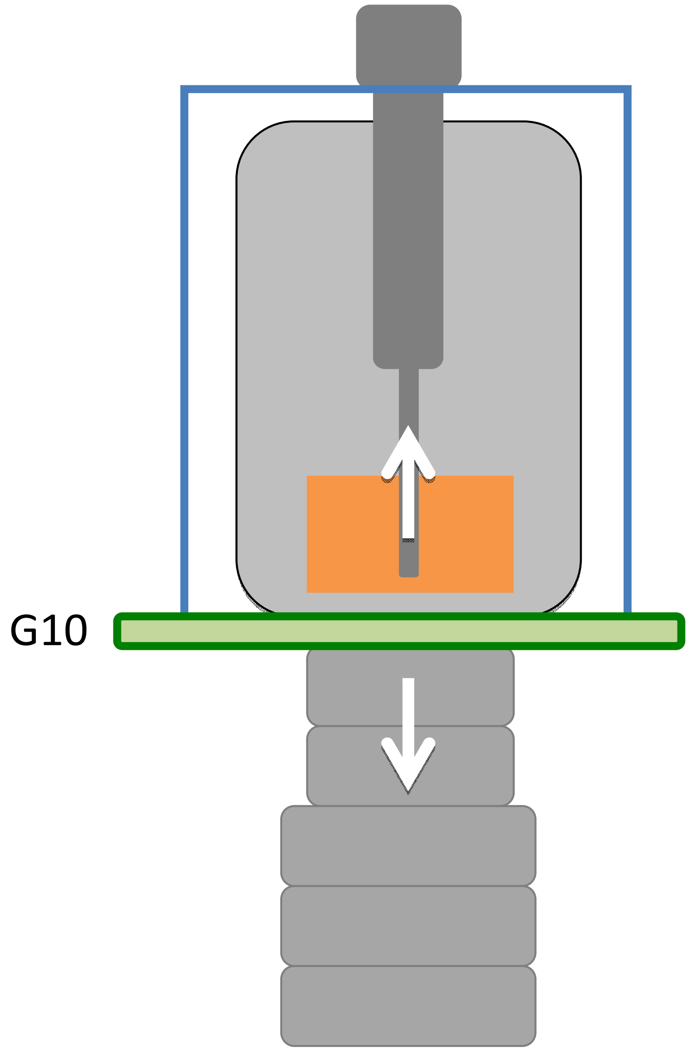

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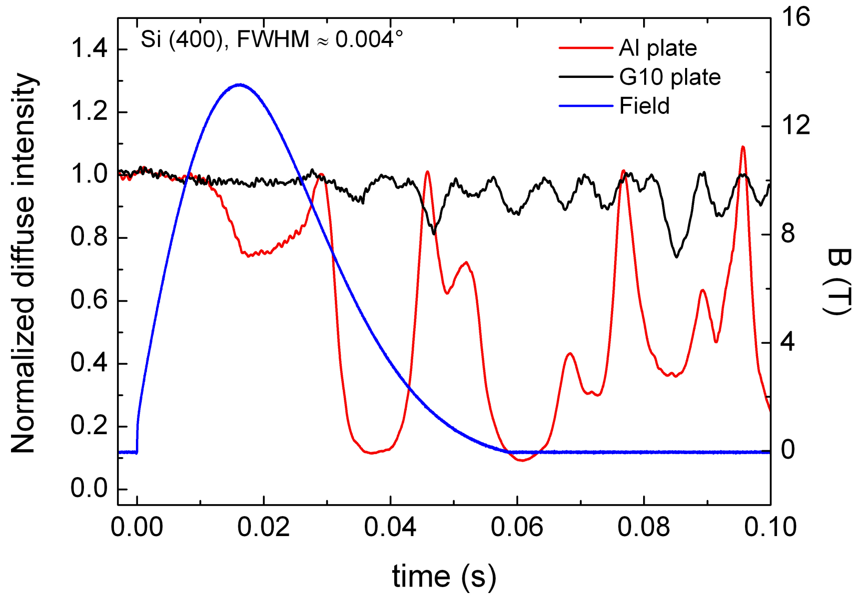
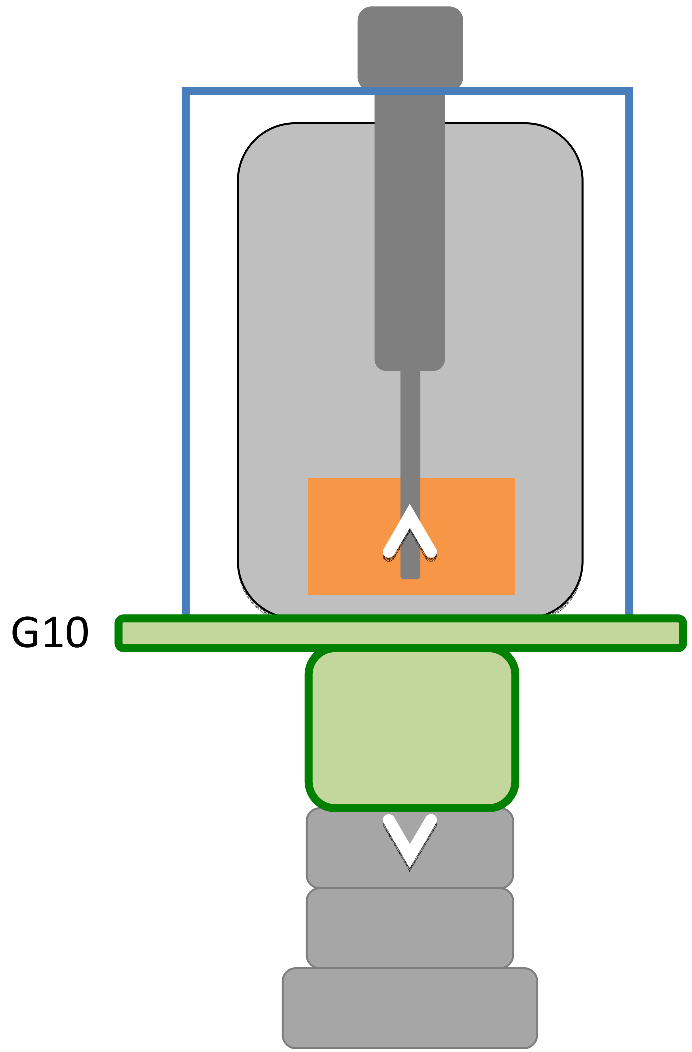
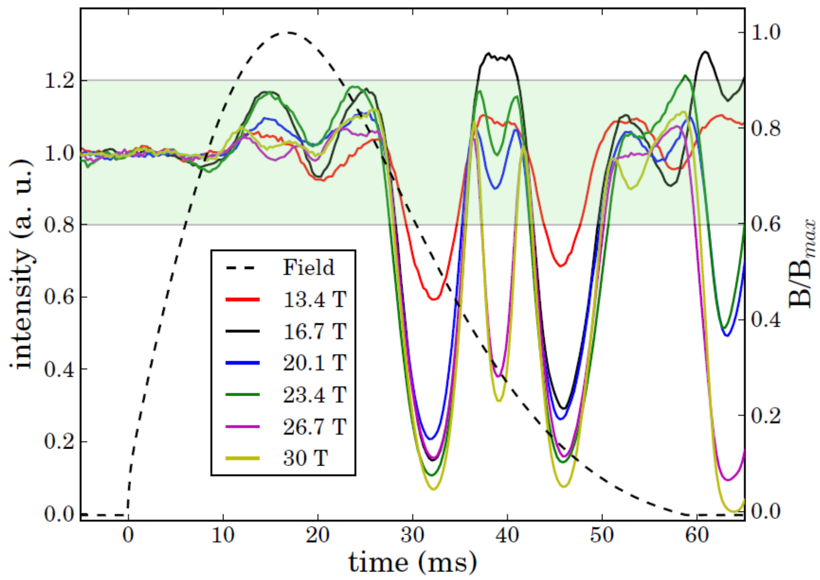
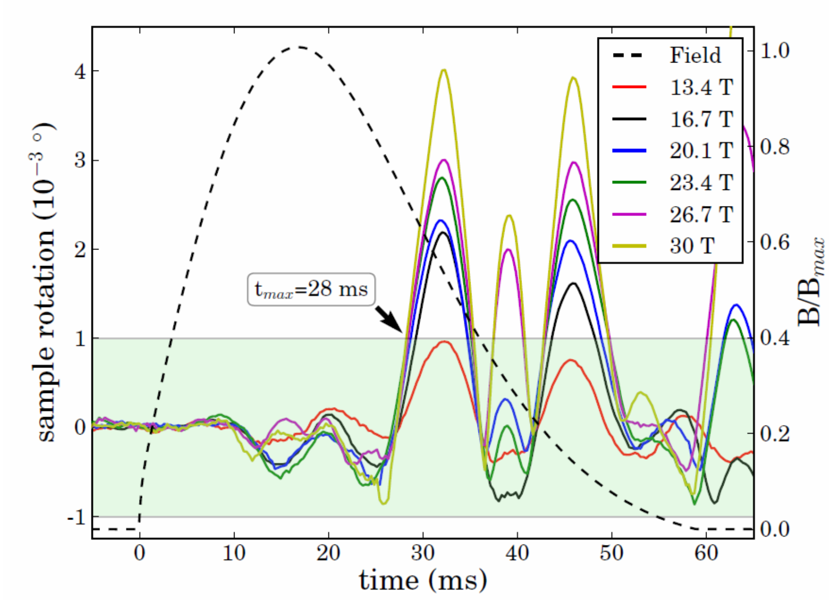




Vibration sensitivity

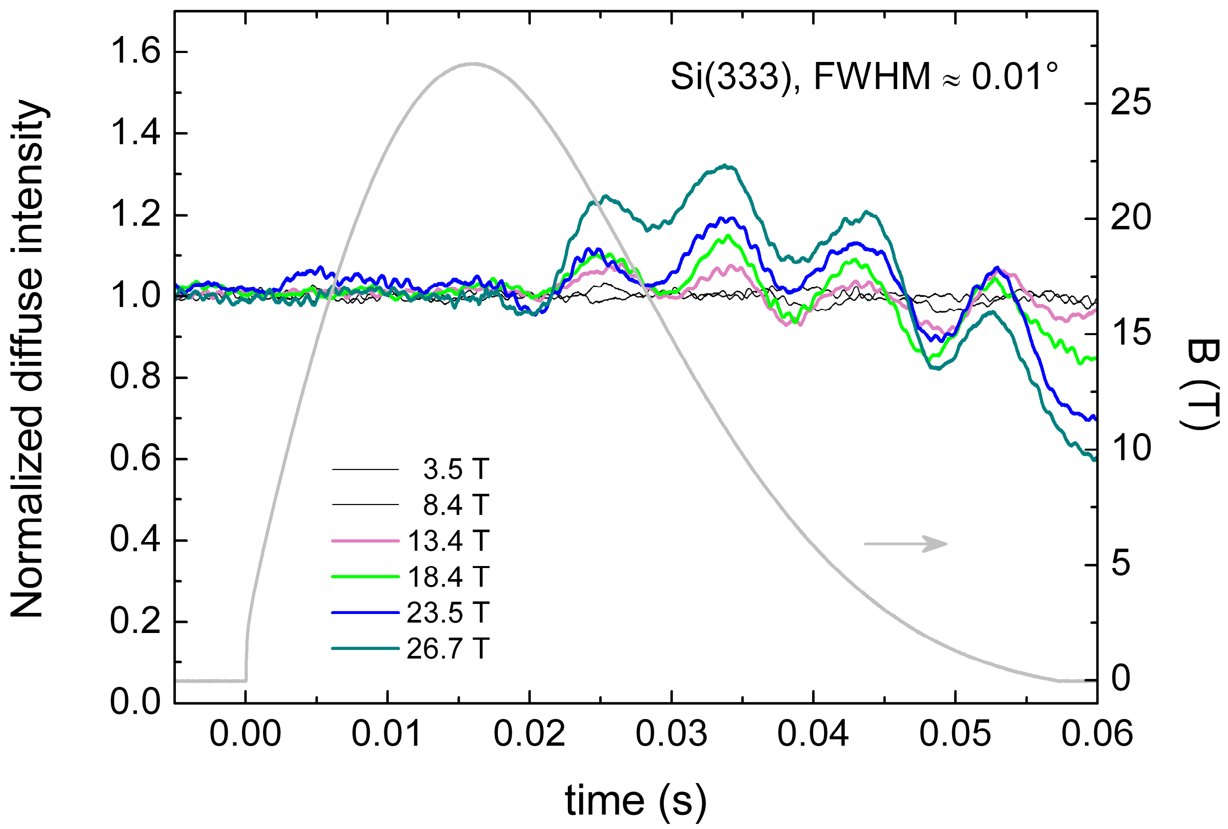
Si(400), FWHM(θ) ~ 0.004°





⇒ Up to ~ 30 ms, sample rotation < 6.10^{-4} deg at 30 T

Vibration sensitivity



⇒ High stability of sample is achieved up to 20 ms

Perspectives for high field X-ray scattering

- ***Time-resolved detector for X-ray single crystal diffraction on ID06***

  MAXIPIX high frame rate pixel detector

  (developed for time-resolved, noiseless and high spatial resolution X-ray detector)

  $\Rightarrow$ *Magneto-structural behavior of frustrated spinel systems*

- ***New developments***

  30 T magnet with conical bore *will be modified* ***to allow single crystal diffraction***

  $\Rightarrow$ ***Single crystal diffraction measurements in backscattering geometry***

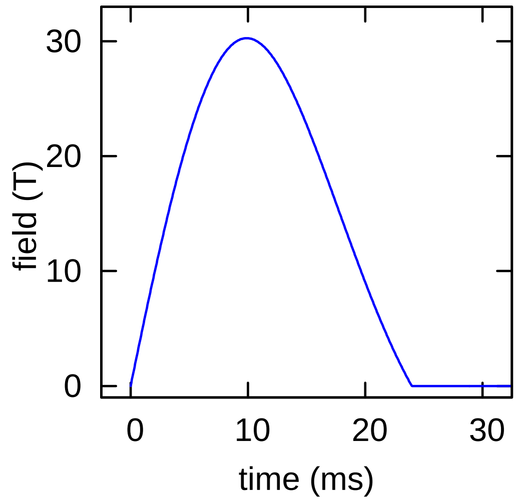
  $\Rightarrow$ *Charge fluctuations in high T_c cuprates*

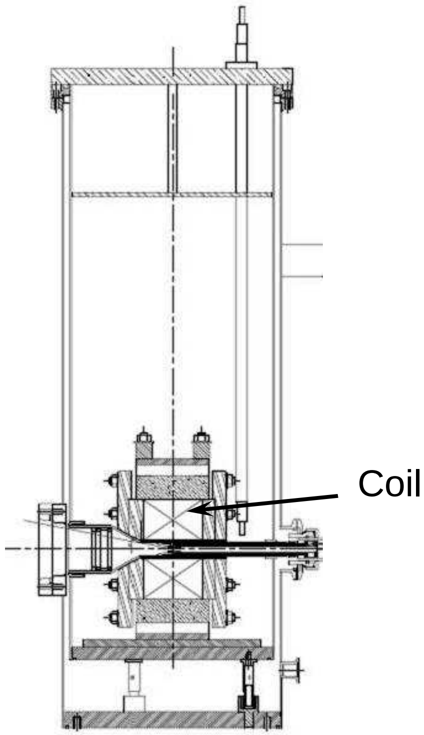
Perspectives: XAS and XMCD on a dispersive XAS beamline ID24 @ ESRF

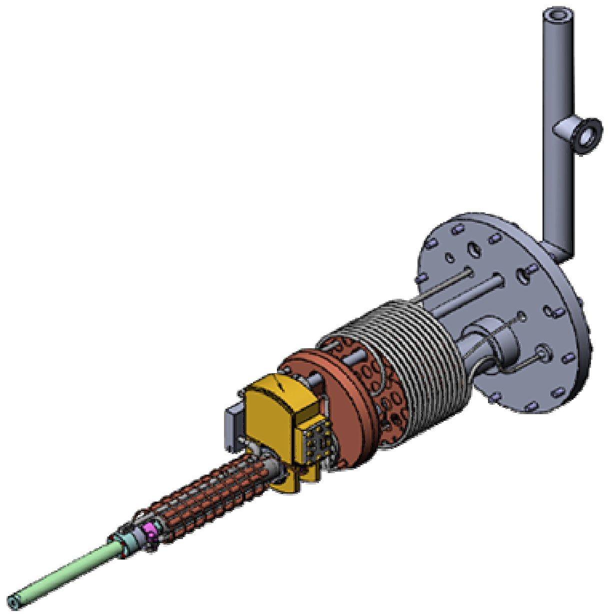
Collaboration: P. van der Linden, C. Strohm

- Solenoid coil: LNCMI Toulouse
  30 T in dia. 16 mm @ 77 K (✔)
- Pulse duration (total) **23 ms**
- Repetition rate at B_{max} **6 pulses/hour**
- Geometry ***B // incident beam***

- cryostat: P. van der Linden (ESRF)
  1.5 K in dia. 9 mm (✔)
- 'top – loading'
- 1.5 K to 300 K







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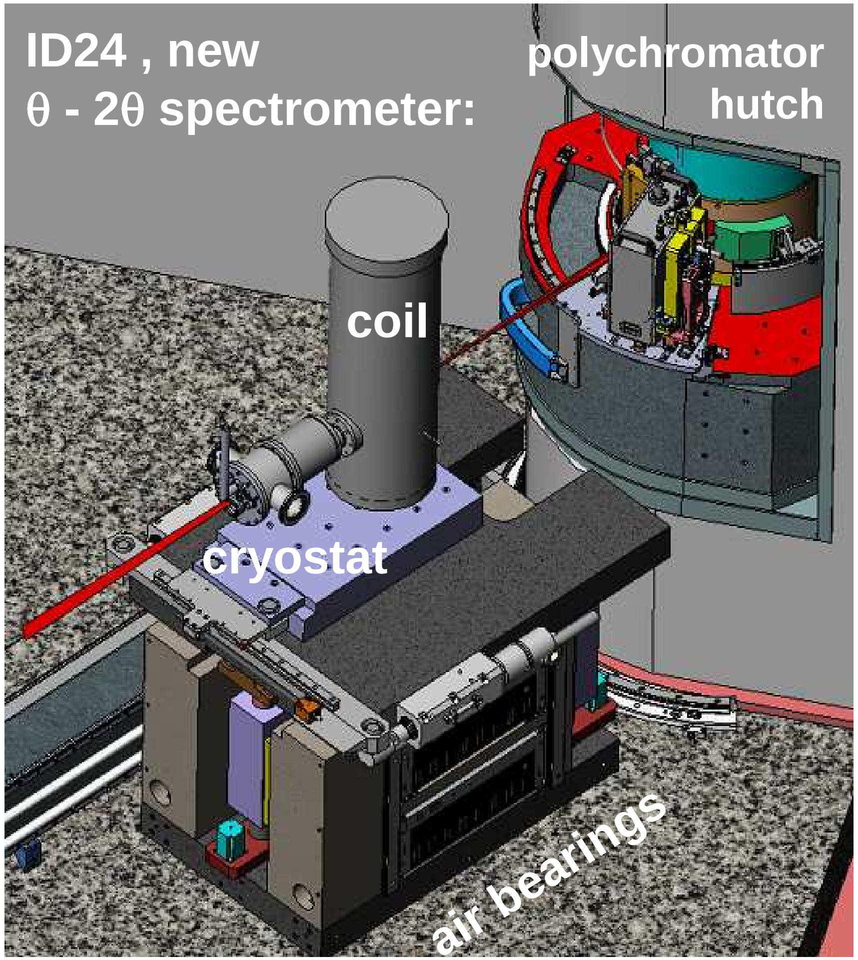
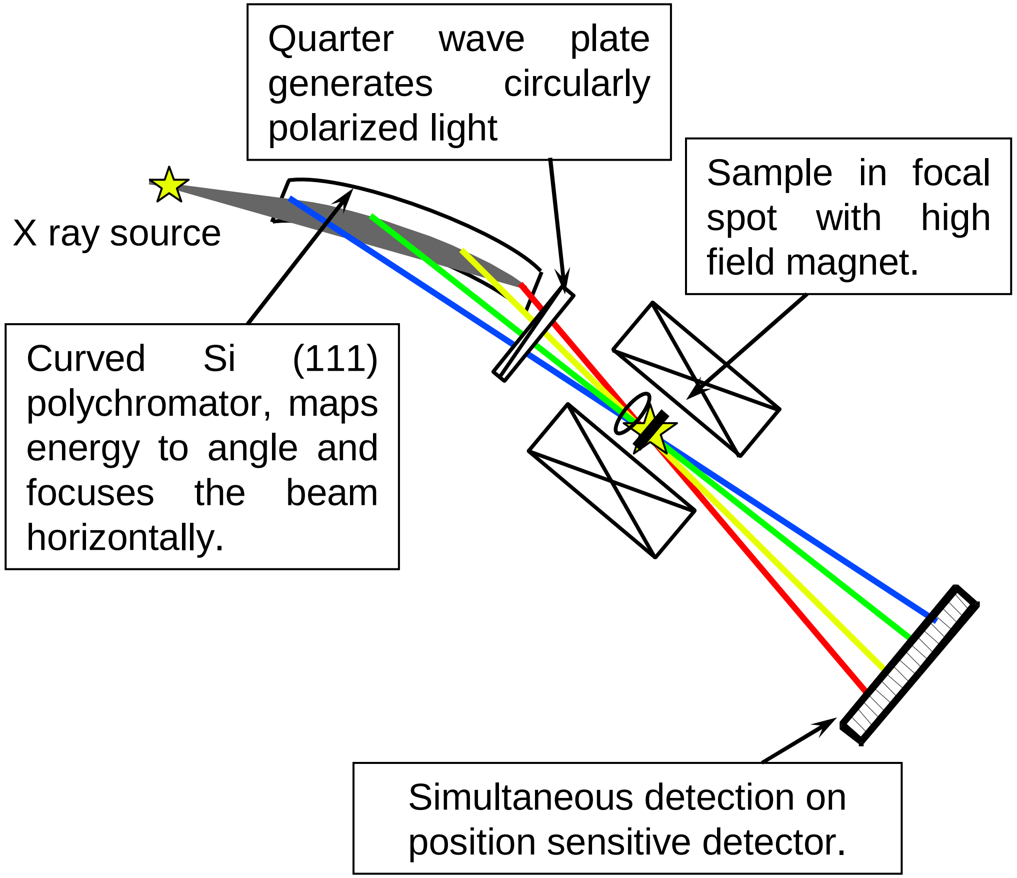




⇒ *Field-induced valence changes in Ce- and Yb-based heavy fermions*

Perspectives: Development of a new end-station for soft X-ray magnetic dichroism experiments SIM beamline, SLS

Collaboration: IPCMS-Strasbourg, SLS Villigen, SOLEIL, LNCMI-T, IMPMC-Paris

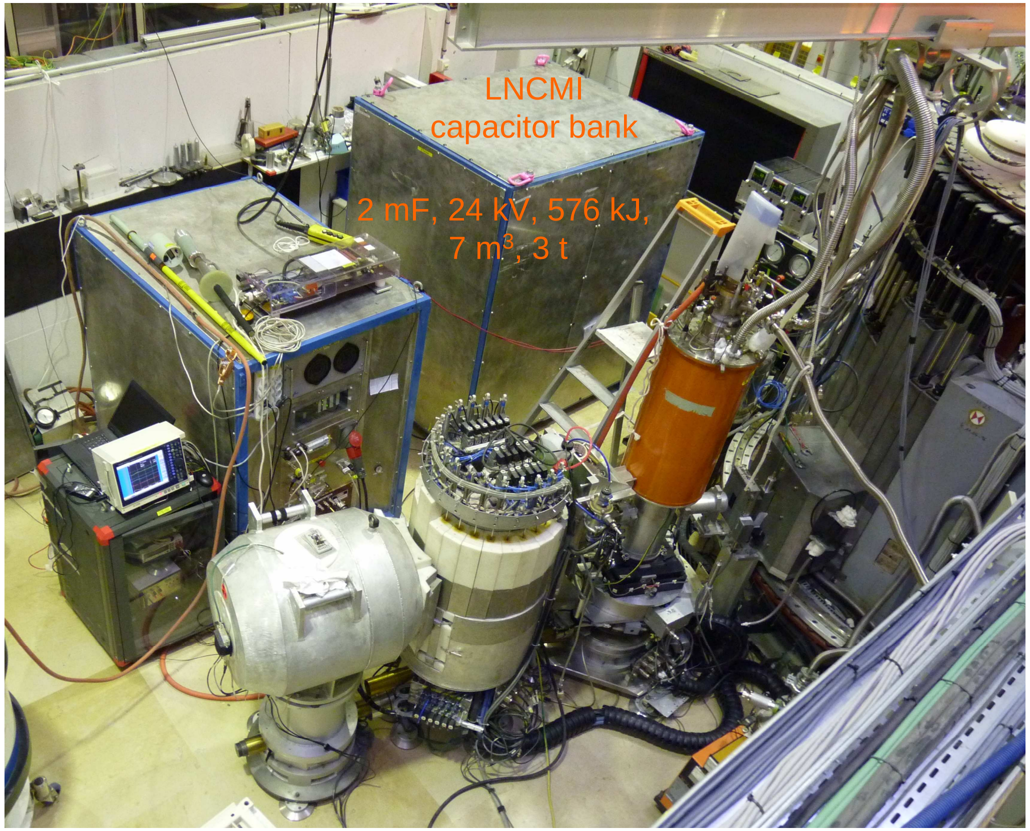
- Solenoid coil: LNCMI Toulouse
  30 T in dia. 16 mm @ 77 K (✔)
- Pulsed field generator: LNCMI Toulouse

- cryostat: IPCMS-Strasbourg
- 10 K

⇒ *Study of the first-order field-induced transition from paramagnetic to ferromagnetic state in the* $Co(S_{1-x}Se_x)_2$ *pseudobinary compounds, by recording and modeling the* $Co\text{-}L_{2;3}$ *edges as a function of magnetic field.*

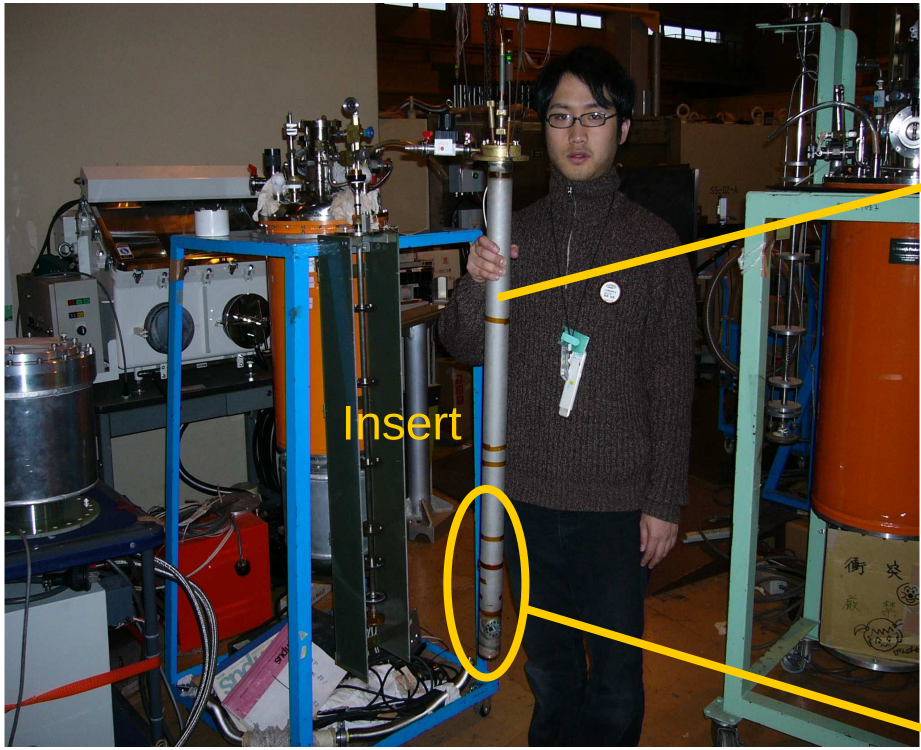
Neutron diffraction in pulsed fields
on IN22 spectrometer (CEA-CRG @ ILL, Grenoble)

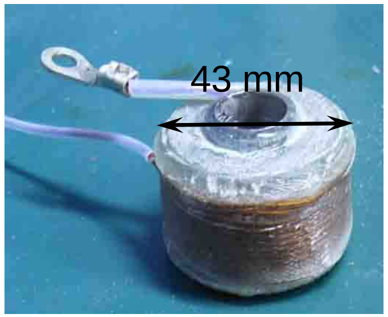
Collaboration: IMR/Sendai, INAC/CEA-Grenoble, LNCMI-Toulouse



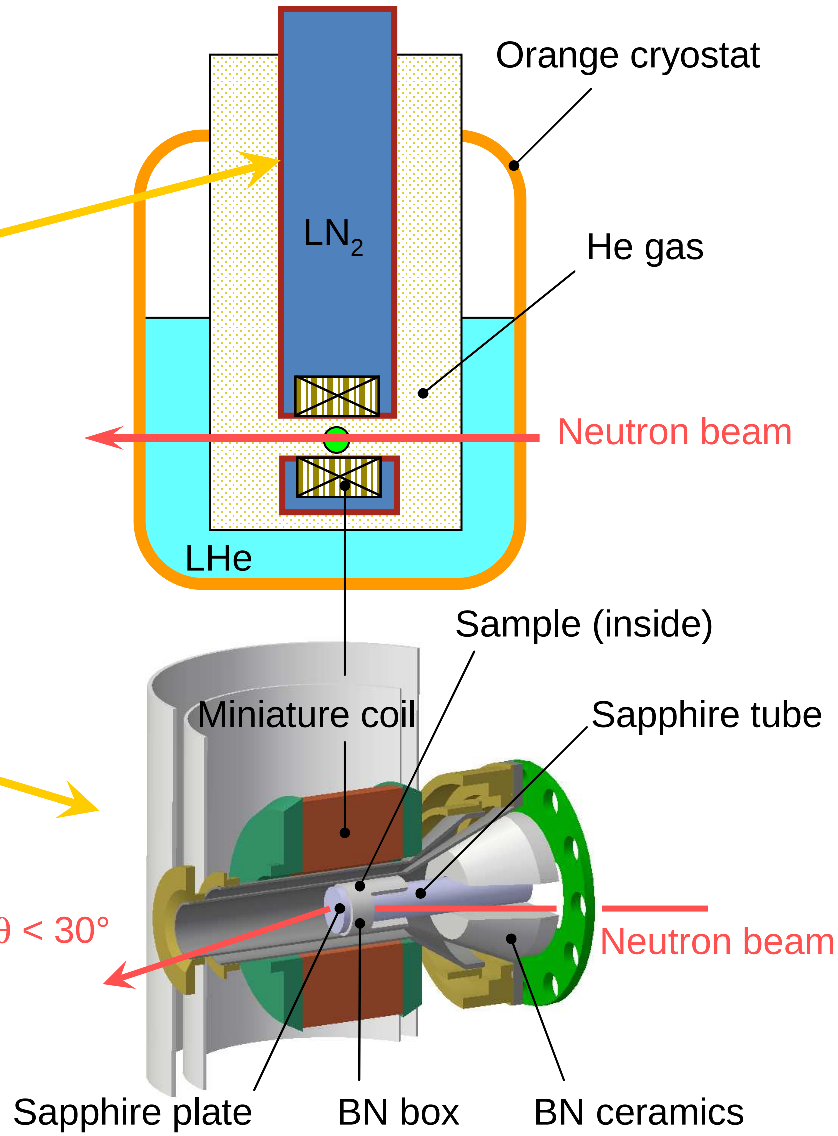
- Magnet and cryostat insert: IMR/Sendai
- Pulsed field generator: LNCMI-Toulouse
- Neutron spectrometer: CEA-CRG @ ILL

Magnet coil and cryostat insert (IMR, Japan)

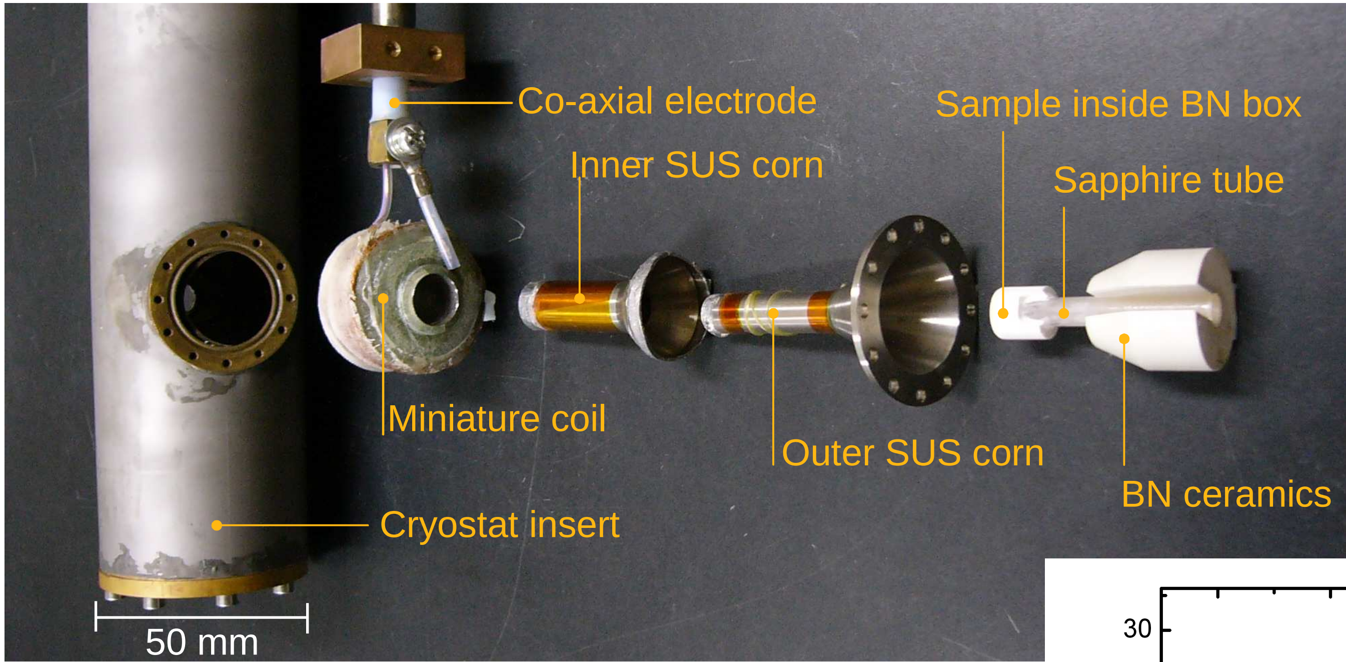




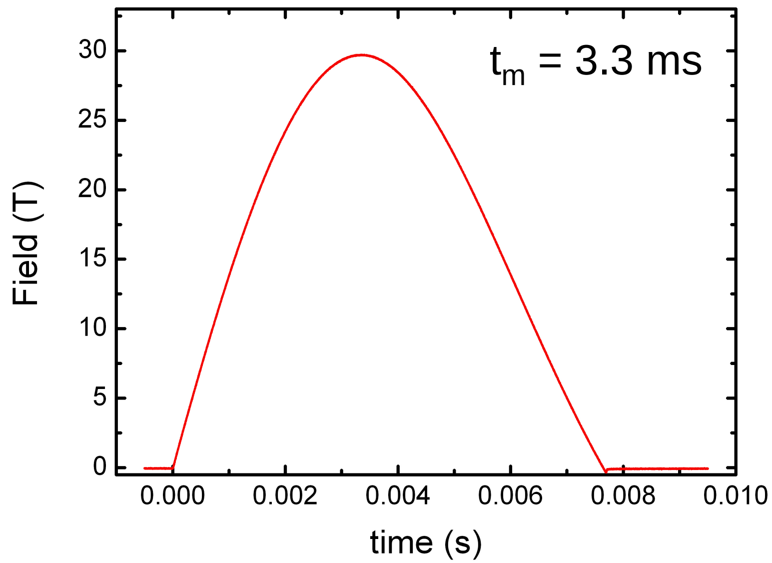
Sample space: ∅ = 9.4 mm, 2θ < 30°



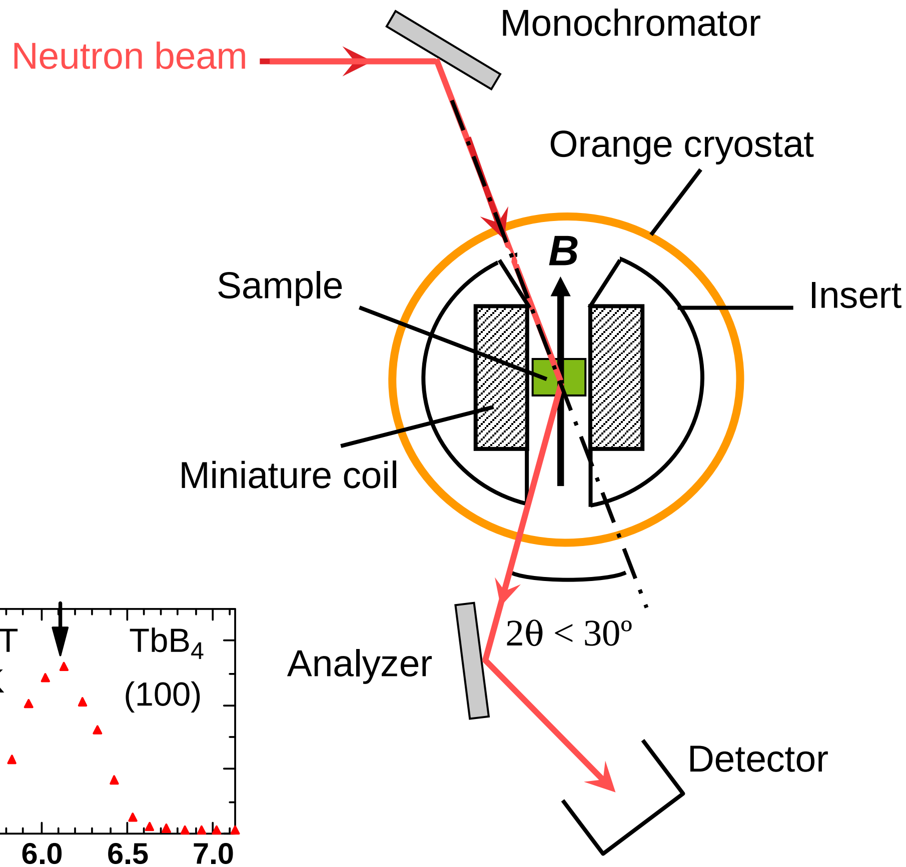
Magnet coil and cryostat insert (IMR, Japan)

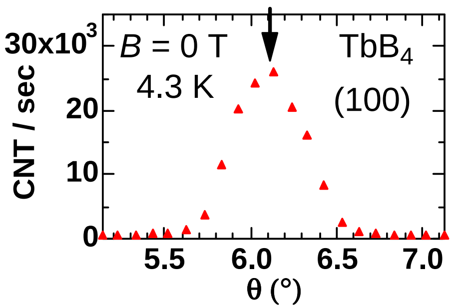


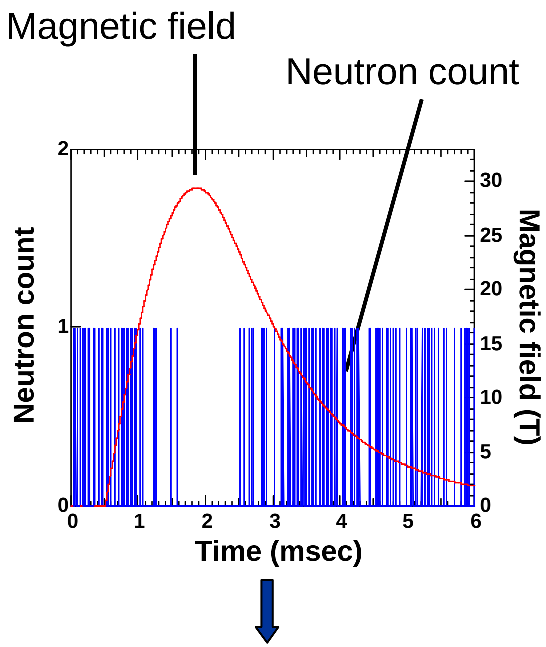
- Maximum field **30 T**
- Pulse duration (total) **7 ms**
- Cool down time **6-7 min**
- Duty cycle **1.7 .10^{-5}**
- Geometry ***B* // incident beam**

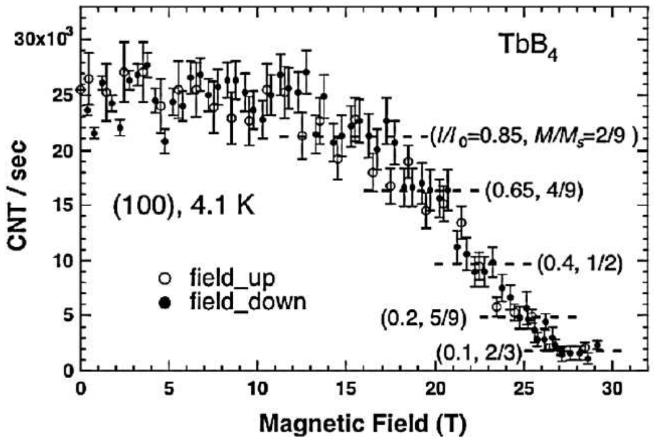


Data acquisition scheme









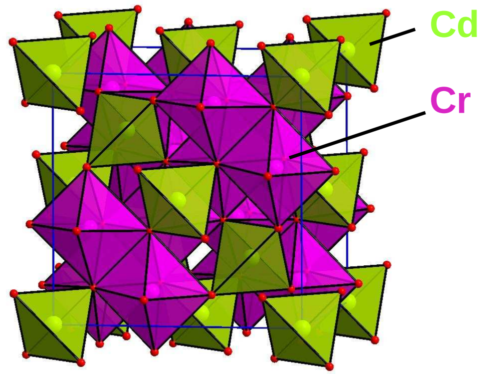
- Fixed Q
- Synchronous collection of field and neutron count
- Time-binning of neutron count
- Accumulation of 100-200 pulses/Bragg reflection (12-24H)

Yoshii *et al.*, Phys. Rev. Lett. 103, 077203 (2009)

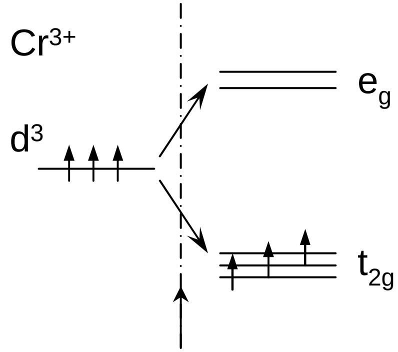
Application to the frustrated spinel $CdCr_2O_4$

⇒ geometrically frustrated system

- Magnetic Cr^{3+} (S = 3/2)
- AF interactions: Curie-Weiss θ_{CW}= -88 K



Spinel $CdCr_2O_4$



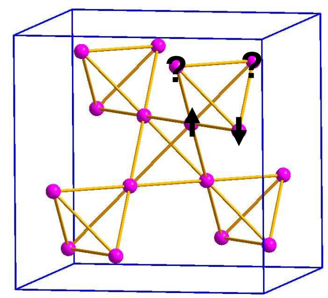
Crystal field

No orbital degree of freedom
No spin-orbit coupling

Application to the frustrated spinel $CdCr_2O_4$

⇒ geometrically frustrated system

- Magnetic Cr^{3+} (S = 3/2)
- AF interactions: Curie-Weiss θ_{CW}= -88 K

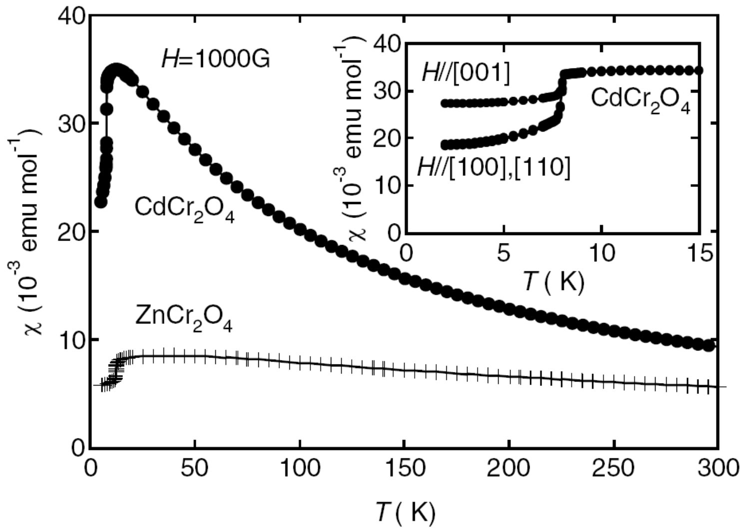


Cr^{3+} pyrochlore lattice

$T_N = T_s$ = 7.8 K

- Transition to a noncollinear AF state

Ueda *et al.*, Phys. Rev. Lett. **94**, 047202 (2005)



Ueda *et al.*, Phys. Rev. Lett. 94, 047202 (2005)

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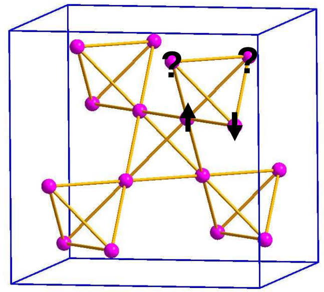


Cr^{3+} pyrochlore lattice

$T_N = T_s$ = 7.8 K

- Transition to a noncollinear AF state

Ueda *et al*., Phys. Rev. Lett. **94**, 047202 (2005)

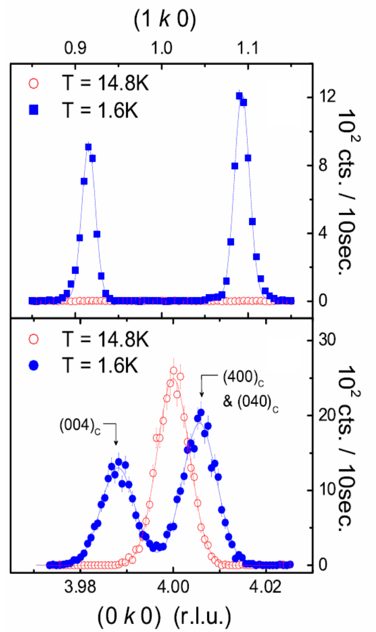
- Zero field magnetic structure = incommensurate helical spin structure

Chung *et al*., Phys. Rev. Lett. **95**, 247204 (2005)

- Structural transition cubic *Fd -3m* ⇒ tetragonal $I4_1/amd$

  with c > a = b

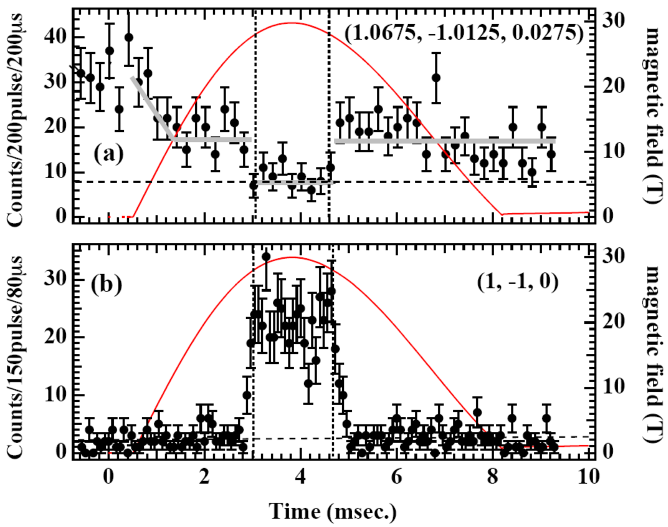
Chung *et al*., Phys. Rev. Lett. **95**, 247204 (2005)

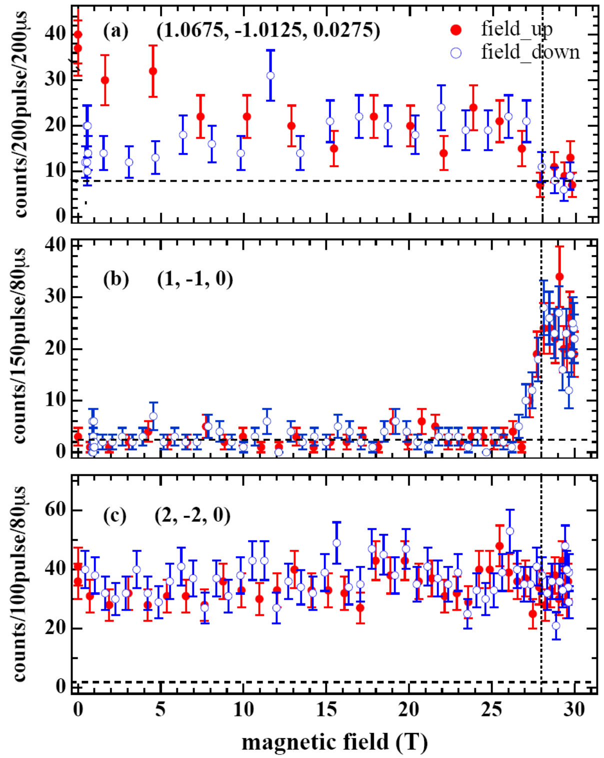


Chung *et al*., Phys. Rev. Lett. 95, 247204 (2005)

Magnetic structure of the half-magnetization plateau phase

T = 2.5 K, sample mass 40 mg

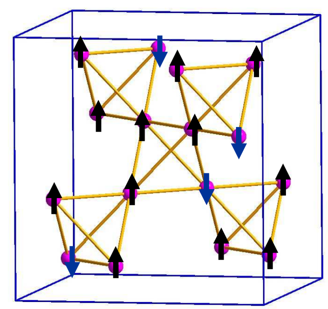




Incommensurate

Commensurate

***P*4$_3$32**



⇒ Incommensurate-commensurate transition observed

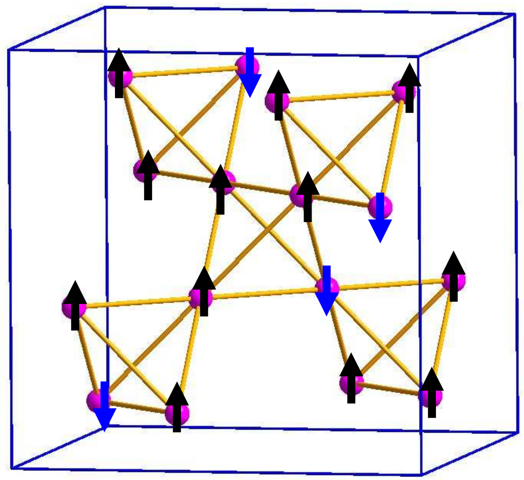
⇒ High field magnetic structure established

Matsuda *et al.*, Phys. Rev. Lett. 104, 047201 (2010)

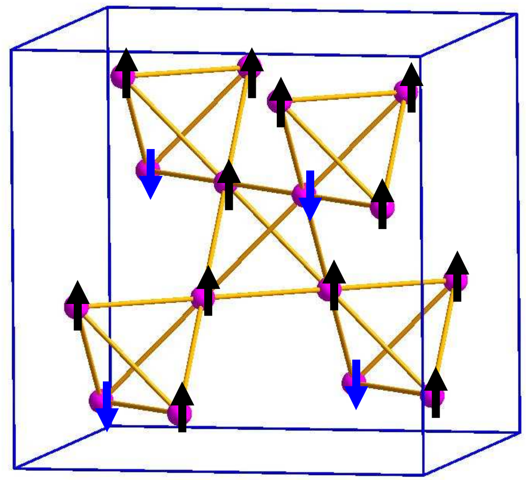
Magnetic structure of the half-magnetization plateau phase

2 possible spin orders

P4$_3$32



R-3m



↑⇩↑⇩ along [111]
(FM in the plane)

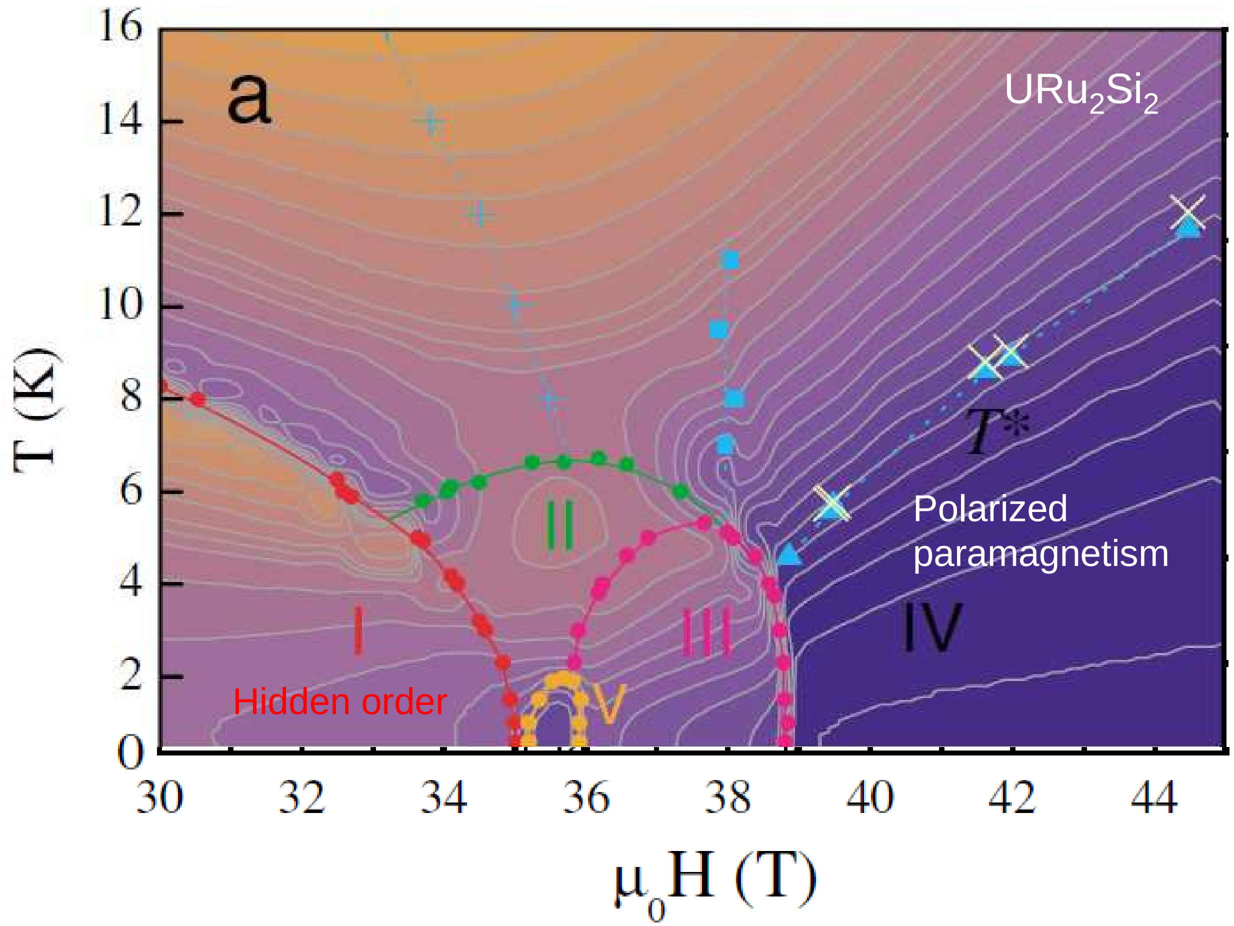
Extinctions rules are different

- (1,-1,0), (2,1,0), (111) only observed in ***P4$_3$32*** ⇒ **(1,-1,0)**, (2,1,0) purely magnetic
  small nuclear contribution to (111)

- Magnetic scattering on **(2,-2,0)** only observed in ***R-3m*** (nuclear contribution to (2,2,0))

Ueda et al., Phys. Rev. Lett. 94, 047202 (2005)

Metamagnetic transition in URu_2Si_2



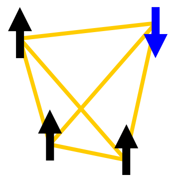
Kim *et al*., Phys. Rev. Lett. 91, 256401 (2003)

Magnetic structure of the half-magnetization phase in the frustrated spinel $CdCr_2O_4$

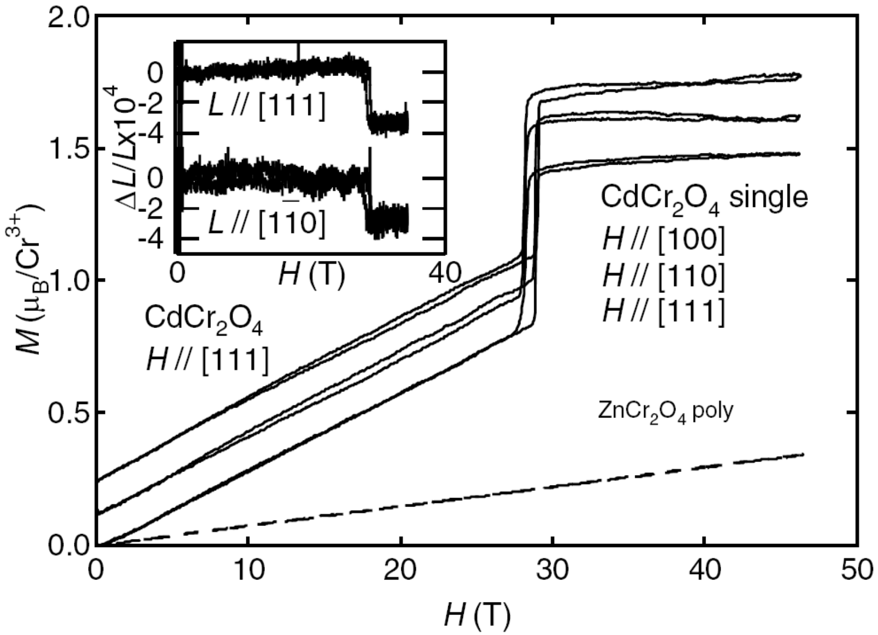
H_{c1} = 28 T

- 1^{rst} order isotropic transition
- Half-magnetization plateau phase

⇒ Ferrimagnetic phase uuud realized



- Large lattice distortions: $\Delta L/L = -4x10^{-4}$

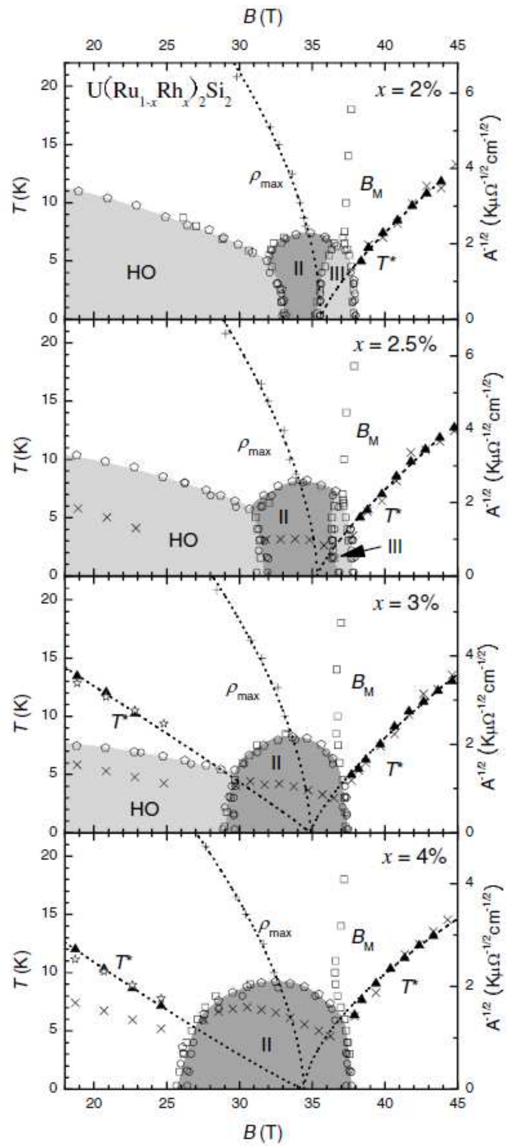


⇒ Strong spin-lattice couplings

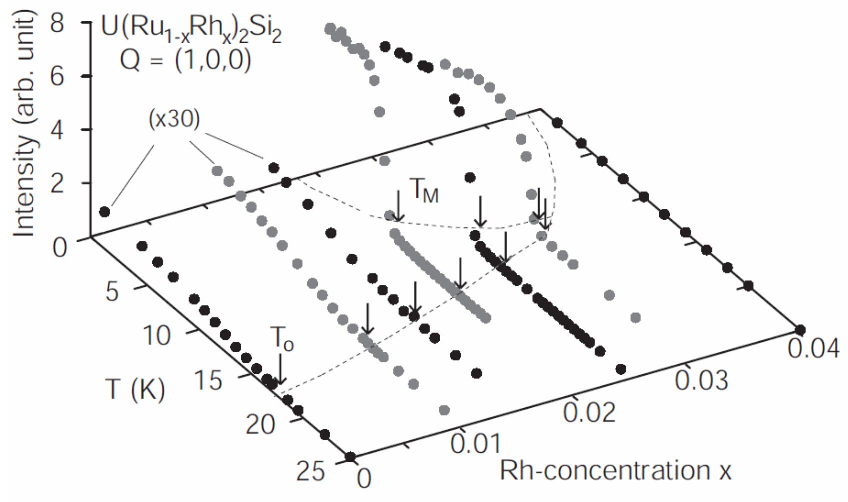
⇒ Half-magnetization plateau phase stabilized by lattice distortion

Ueda *et al*., Phys. Rev. Lett. 94, 047202 (2005)

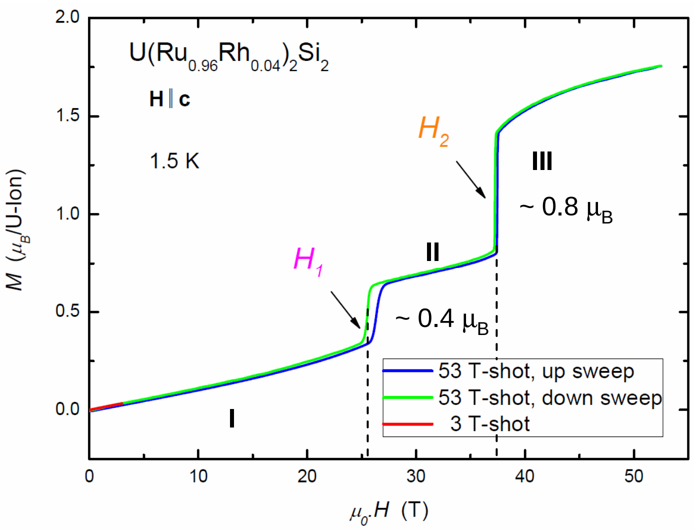
4%Rh-URu_2Si_2 sample (CEA-Grenoble)



Kim *et al.*, Phys. Rev. Lett. 93, 206402 (2004)



Yokoyama et al., J. Phys. Soc. Jpn. 76, 136 (2007)



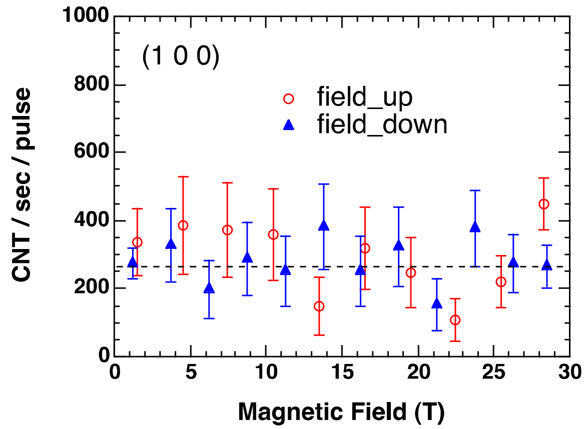
- Field induced AF order ?
- Phase II

1/3 magnetization ⇒ uud state

In-plane

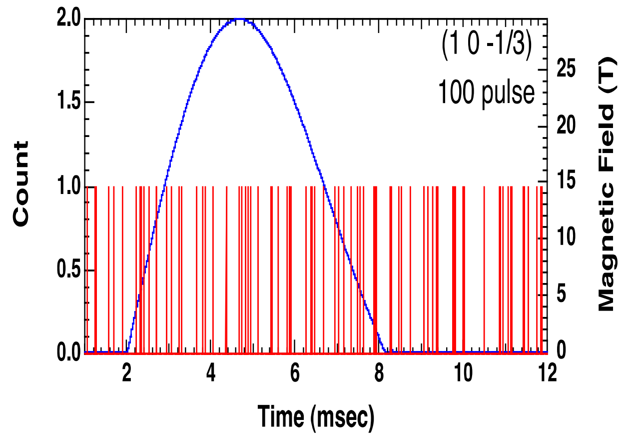
or inter-plane modulation ?

AF (100) and uud along *c*-axis

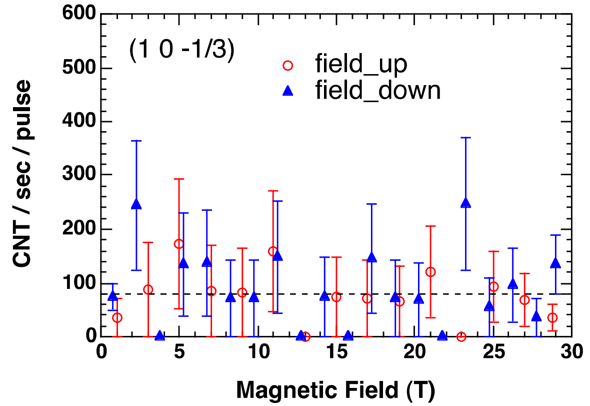


⇒ **No intensity beyond the BG**

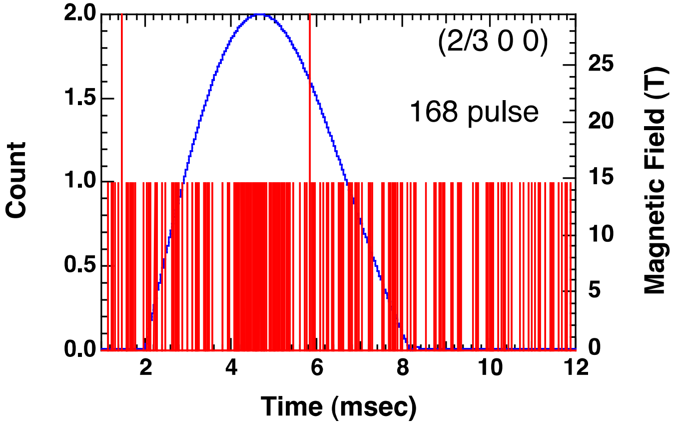
⇒ **AF (100) order not observed**

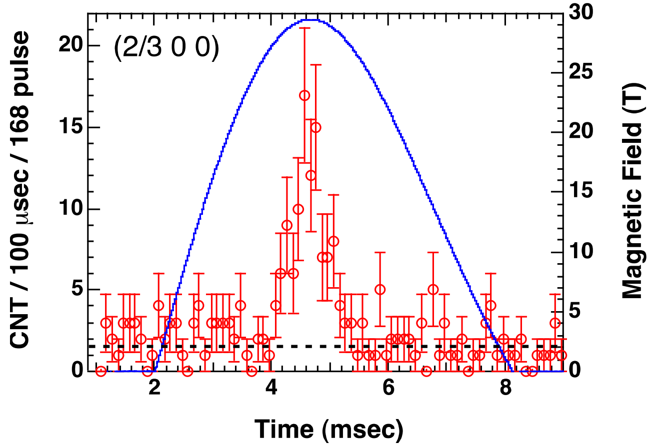


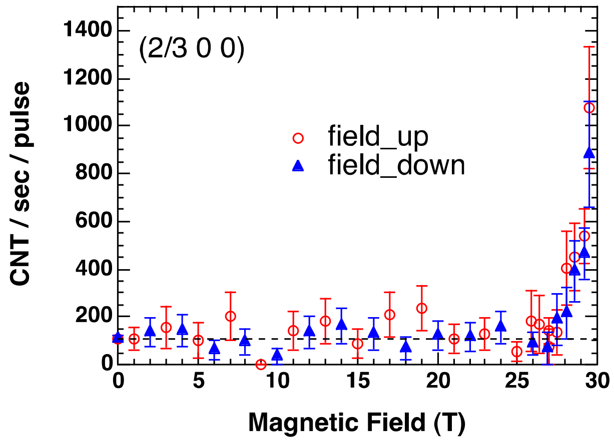
⇒ **(1, 0,-1/3) = no intensity beyond the BG**



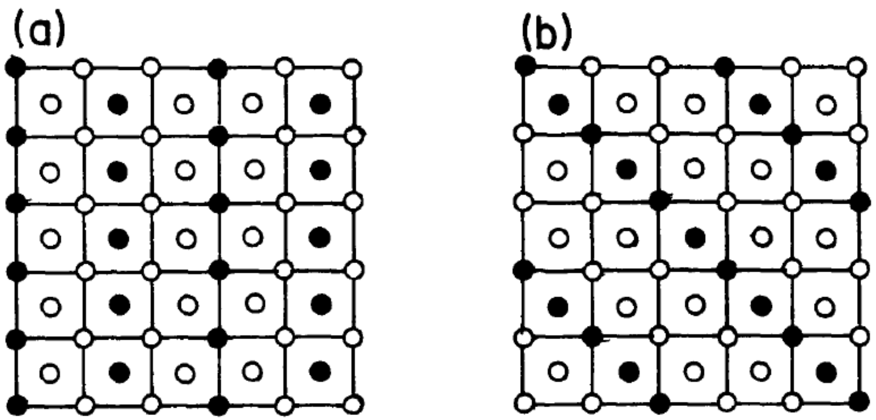
⇒ **uud spin modulation is not along *c*-axis**







In plane uud modulation



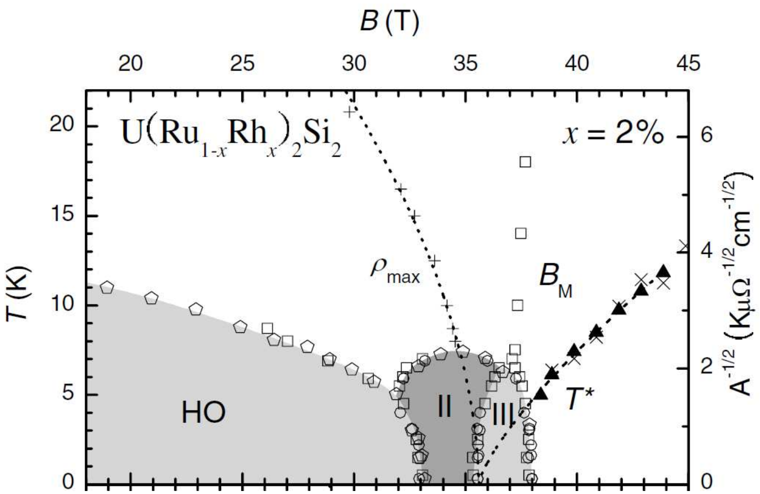
Sugiyama *et al*., J. Phys. Soc. Jpn. 59, 3331 (1990)

⇒ (2/3, 0, 0) = wave vector of phase II

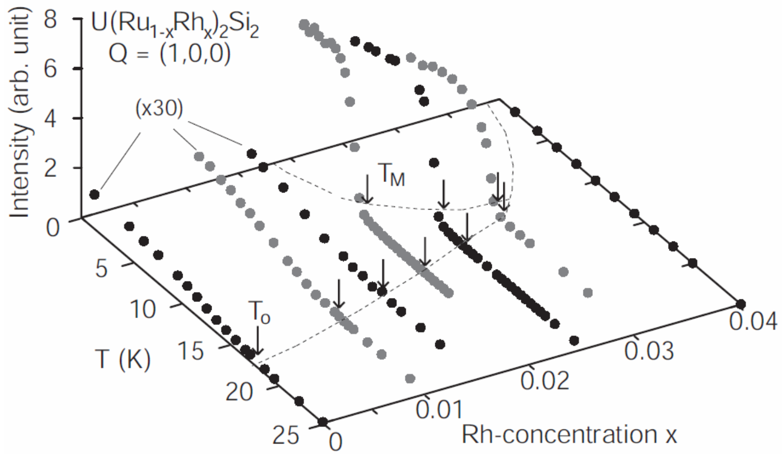
Kuwahara *et al*., to be published

Perspectives: 2%Rh-URu_2Si_2 and pure sample

Collaboration: IMR/Sendai, INAC/CEA-Grenoble, LNCMI-Toulouse



Kim *et al.*, Phys. Rev. Lett. 93, 206402 (2004)



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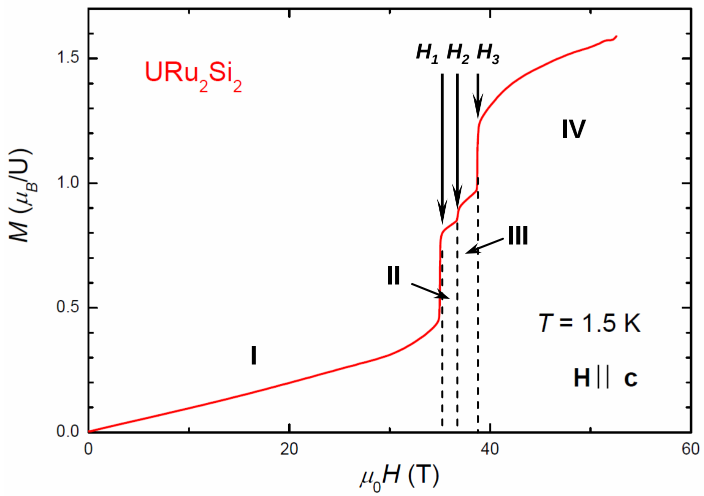
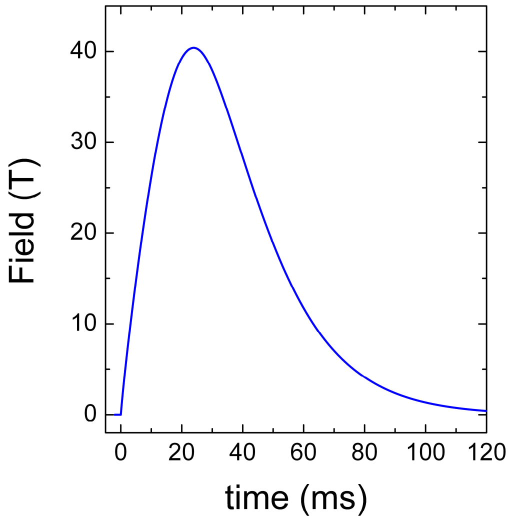


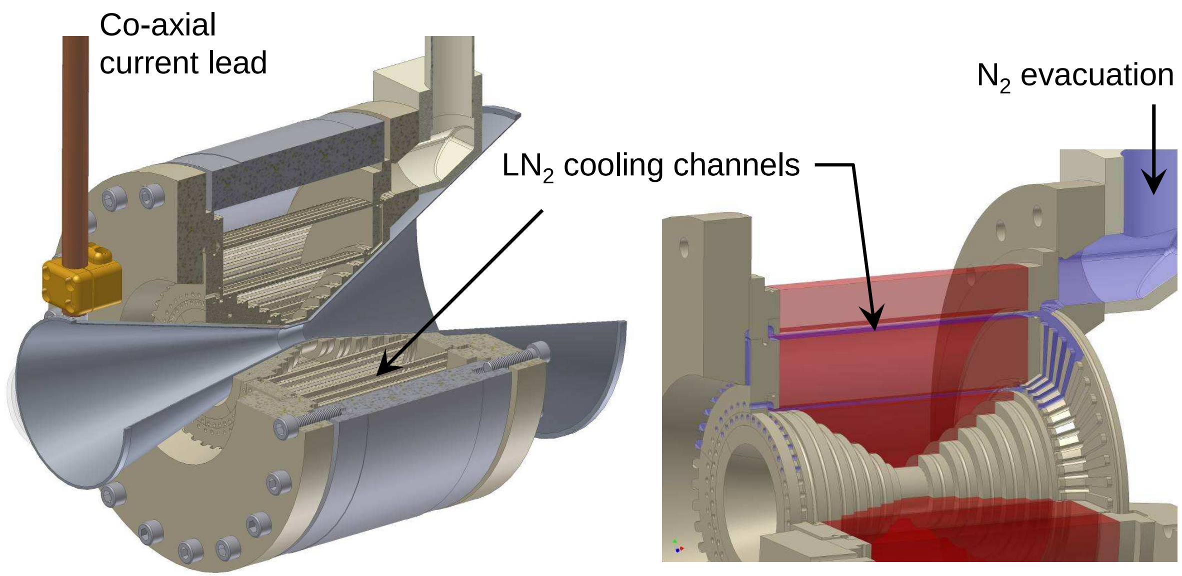
Perspectives for high field neutron diffraction

Collaboration: LNCMI, INAC/CEA, ILL, Institut Néel
ANR financial support: Dec. 2010- Dec.2014

40 T long pulse rapid cooling magnet

• Maximum field	**40 T (ΔB <2%)**	• Geometry	**conical**
• Pulse duration (total)	**100 ms**		***B* // incident beam**
• Cool down time	**7 min**	• Sample volume	**~ 0.15-0.2 cm³**
• Duty cycle	**2.4 .10⁻⁴**	• Opening angle	**± 15°(in), ± 30°(out)**





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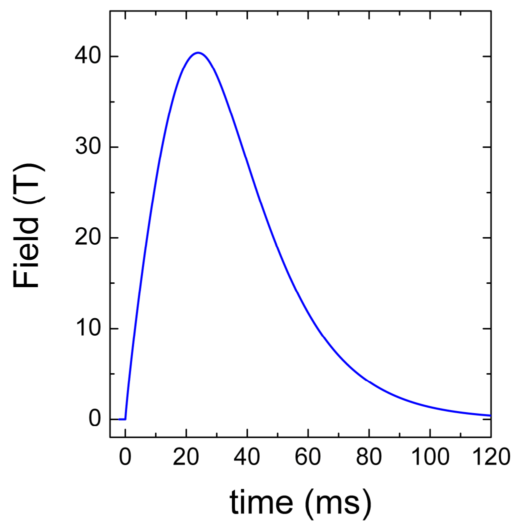
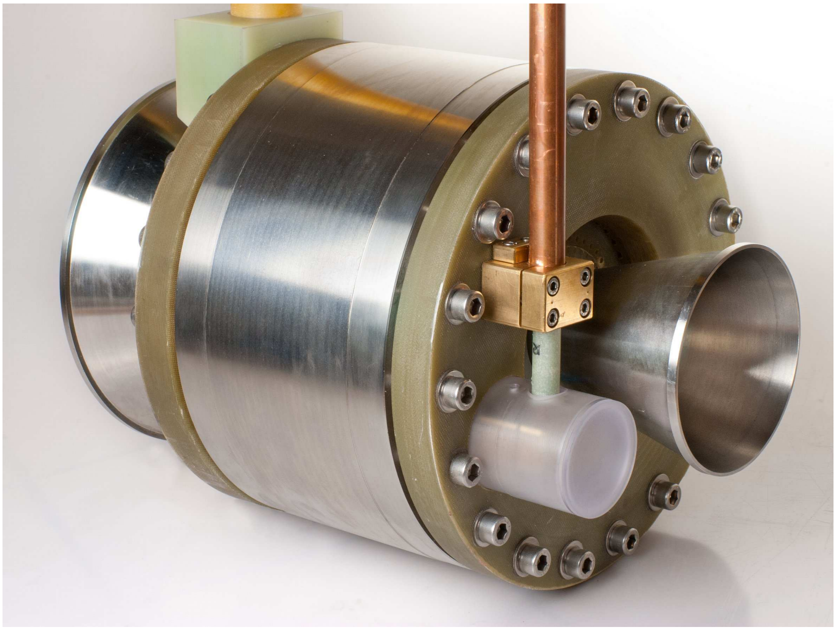




Perspectives for high field neutron diffraction

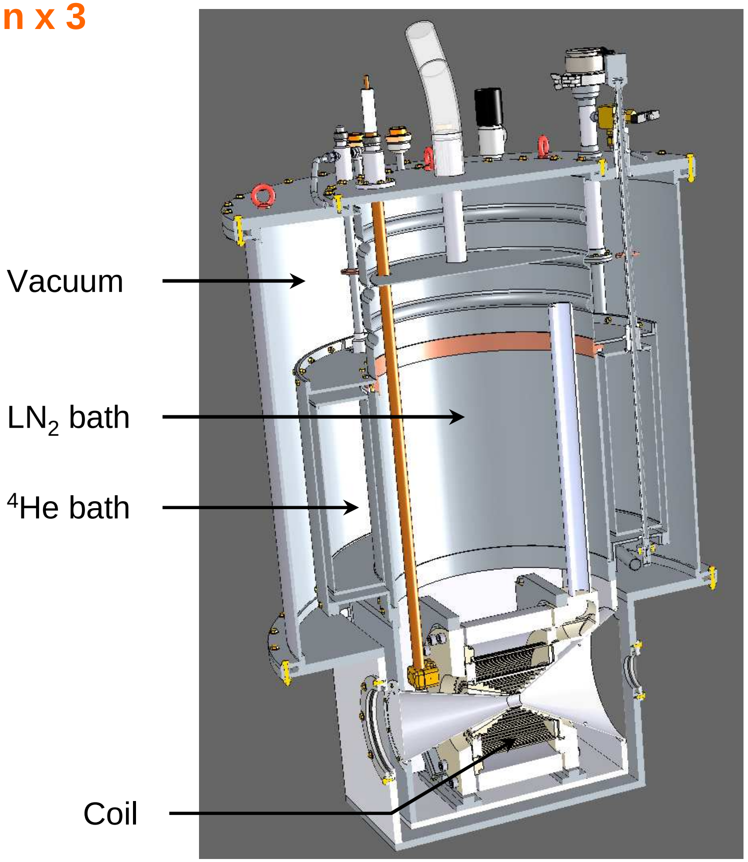
Collaboration: LNCMI, INAC/CEA, ILL, Institut Néel
ANR financial support: Dec. 2010- Dec.2014

- Duty-cycle improvement
  - Increase the pulse duration (~ 100 ms) → **Gain x 15**
  - Optimization of neutron equipment: → **Gain x 3**

    maximize the number of neutrons detected/pulse

    - Efficient focusing neutron optics
    - New fast and high counting rate detector

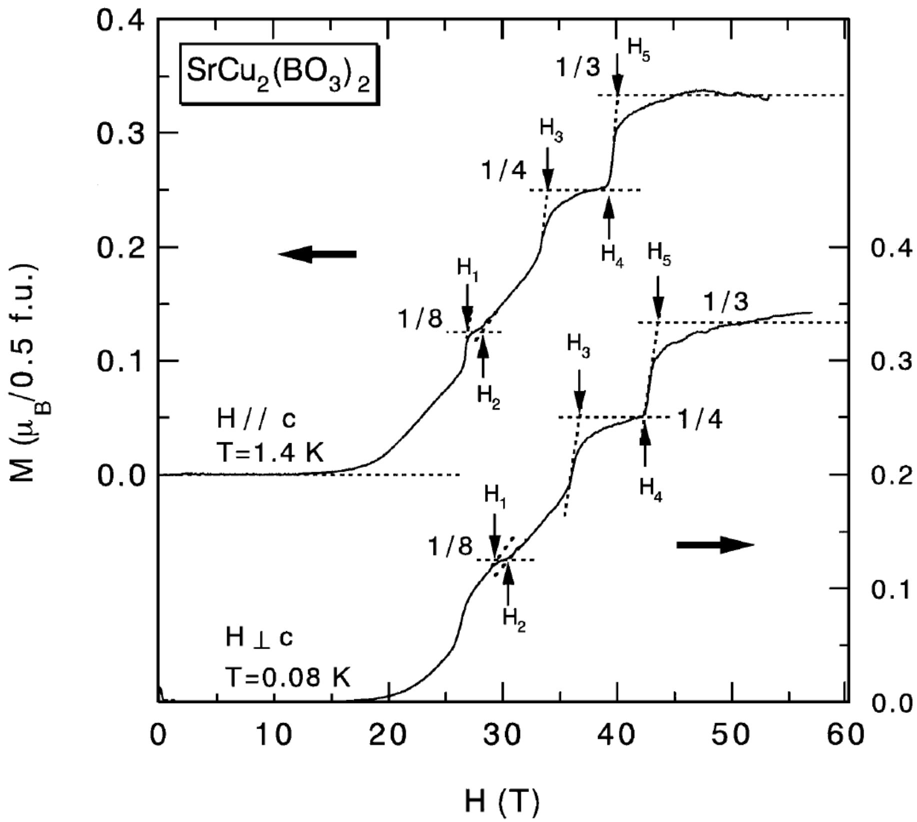
- Cryogenic environment
  - Coil in LN_2 bath
  - ^{4}He bath to cool the sample
  - Heat exchanger in vacuum, inside the cone
  - Sample temperature: 1.5-2 K



Perspectives for high field neutron diffraction

Powerful tool to investigate high magnetic field induced phases

- strongly correlated electron systems and quantum magnets:
  - competing phases in cuprates
  - heavy fermions
  - frustrated systems
  - Haldane chains…



Onizuka *et al.*, J. Phys. Soc. Jpn. 69, 1016 (2000)

Merci !