X-ray powder diffraction
under pulsed magnetic fields up to 30T

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Overview

- Why high magnetic fields?
- How to generate magnetic fields?
- Pulsed magnetic fields
- Application to X-ray diffraction
- Example: Jahn-Teller transition of TbVO$_4$
- Outlook: Future developments
Why high magnetic fields?

- The magnetic field is a thermodynamic variable of fundamental importance, as temperature or pressure.

- All electrons carry a spin, and therefore a magnetic moment. Therefore, in principle, all condensed matter is concerned:
  
  - Magnetically ordered systems (changes of magnetic structure),
  - Polymers (orientation),
  - Semiconductors (quantum Hall effect),
  - Superconductors (flux line lattices, destruction of superconductivity)
  
  ... and many others

- The higher the available field, the larger the number of phase transitions and other effects that can be observed.
How do you generate a magnetic field?

- Up to 1 T: Permanent magnets.
- Up to 15 T: Superconducting magnets → ID20, 10 T.
- Up to 33 T: Resistive magnets, 20 MW.
- Up to 45 T: Hybrid superconducting and resistive magnets, (NHMFL, Tallahassee, 24 MW, ≈ 15 M$).
- Up to (existing) 80 T: Pulsed resistive magnets, ←
  (project) 100 T:
  - Up to ≈ 130 T: Destructive pulsed magnets (destroys magnet only).
  - Up to ≈ 600 T: Destructive pulsed magnets (destroys everything).
- Above that: Neutron stars, solar storms, . . .

Current maximum field for x-ray or neutron diffraction: 15 T (17.5 T with “booster”)

Motivation/Scientific case

• There are many laboratories in Europe and elsewhere in the world which are dedicated to high magnetic field research.

• These labs employ a large number of different techniques:
  → Magnetization and susceptibility.
  → Transport (resistivity, Hall effect, magneto-resistance).
  → Specific heat.
  → Dilatometry and sound velocity.
  → De-Haas-van-Alphen effect (Fermi surface mapping)
  → NMR (Nuclear magnetic resonance)
  → Optical spectroscopy (Raman scattering, reflectivity, ellipsometry, . . .)
Motivation/Scientific case

• All of the current techniques are macroscopic measurements.

... but there is no information about the microscopic structure of the sample at fields above 15 T!

• At the same time we know (from measurements at lower fields) that often field-induced phase transitions have a structural component.

• Sound velocity and dilatometry measurements at high fields also indicate structural effects.

There is an urgent need for diffraction for fields above 15 T!

→ Find the easiest and most cost-effective way to explore this region of the phase diagram...
How do you generate a magnetic field?

- Up to $1 \text{T}$: Permanent magnets.
- Up to $15 \text{T}$: Superconducting magnets $\rightarrow$ ID20, $10 \text{T}$.
- Up to $33 \text{T}$: Resistive magnets, $20 \text{MW}$.
- Up to $45 \text{T}$: Hybrid superconducting and resistive magnets, (NHMFL, Tallahassee, $24 \text{MW}$, $\approx 15 \text{M\$}$).

$\rightarrow$- Up to (existing) $80 \text{T}$: Pulsed resistive magnets, $\leftarrow$
  (project) $100 \text{T}$:
- Up to $\approx 130 \text{T}$: Destructive pulsed magnets (destroys magnet only).
- Up to $\approx 600 \text{T}$: Destructive pulsed magnets (destroys everything).
- Above that: Neutron stars, solar storms, . . .

Installations become progressively bigger, more expensive, and more difficult to manage, with exception of pulsed fields, which are scalable.
How to generate pulsed magnetic fields?

The principle is very simple:

1) Charge capacitor
2) Close switch
3) Current flows
4) \ldots repeat

\[
\tau = \sqrt{LC}
\]

\[
E = \frac{1}{2} CV^2 = \frac{1}{2} LI^2
\]

\[
E = \frac{1}{2\mu_0} \int dV B^2
\]
... but some details need to be considered:

- **High voltage/high current risks**: 24 kV, 6 kA
  → Grounding, protection of beamline electronics ...

- **Stored energy**: 110 kJ (upgrade to 1.5 MJ planned)
  → transformed into heat at the end of the pulse ...
  → ...need efficient cooling of the coil
  → What happens in case of a fault?

High field laboratories know very well how to handle this.
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Application to x-ray diffraction

- Magnet and capacitor bank supplied by LNCMP/Toulouse
  - Transportable capacitor bank, 130 kJ energy, 2.8 tons, \( \approx 4 \text{ m}^3 \).
  - Solenoid magnet, lq. N\textsubscript{2} cooled, maximum field 30 T, bore 22 mm, max. opening angle 22°.
  - rise time 5 msec, decay time \( \approx 20 \text{ msec} \), 10 shots per hour.
- X-ray powder diffraction at 21 keV
  - Online-image plate detector
  - Fast shutter to synchronize the x-ray exposure to the magnetic field pulse.

30 T limit convenient because of wire material \( \rightarrow \) duty cycle, fatigue, \ldots
\( \rightarrow \) upgrade to 60 T relatively straightforward
X-ray powder diffraction on BM26B DUBBLE

Transportable generator:

- 2 storage modules, 1 charger/control module
- \( C = 1 \text{ mF}, V_{\text{max}} = 16 \text{ kV}, E_{\text{max}} = 130 \text{ kJ} \)
- Total weight \( \approx 2.8 \text{ t} \)
- Total size \((h \times d \times w)\)
  \( 1.25 \times 1.30 \times 2.85 \text{ m}^3 \)
- Generator and load magnet installed in radiation hutch.
- Interlocked through radiation hutch PSS.
- Remote control over fiber optical cables.

Generator design: P Frings (LNCMP).
X-ray powder diffraction on BM26B DUBBLE

Coil design: J. Billette (LNCMP), cryostat design: M. Nardone, A. Zitouni (LNCMP).
X-ray powder diffraction on BM26B DUBBLE

- Shutter synchronized to magnetic field pulse
- Warming of coil after sequence of pulses.
- Signal integrated over $\approx 5\text{ ms}$ per pulse.

Not ultra-fast, but not stroboscopic: Small number of pulses.
Fatigue life: Design system such that 1 shot is enough.
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**Example: Jahn-Teller transition in** \( \text{TbVO}_4 \)

- \( \text{TbVO}_4 \) is a textbook example of a cooperative Jahn-Teller transition mediated by quadrupolar interactions. \( T_{JT} \approx 34 \text{ K} \).

- The system is known since the 1970’ies and has been studied intensively at zero field. G. A. Gehring and K. A. Gehring, Rep. Prog. Phys. 38, 1 (1975).

- Driven by \( \text{Tb} \, 4f \) quadrupole moment

- Orthorhombic distortion, 2\% along \((111)\)

- Space group \( I4_1/amd \rightarrow Fdd \)
Example: Jahn-Teller transition in \( \text{TbVO}_4 \)

- Recently, first studies in high magnetic fields.
- Strongly anisotropic response (CF).
- Theory (qualitative): 1970’ies
  Competition of magnetization and quadrupolar moment

- Theory (quantitative):
  Field of \( \approx 28 \text{T} \) along the c-axis suppresses the JT state.

- Indirectly observed in magnetization.
  Kazei et al, JETP Lett. 82, 609 (2005).

- But so far no direct observation
Jahn-Teller transition in $\text{TbVO}_4$: Raw data

- DUBBLE CRG at ESRF
- 21 keV
- MAR 345 image plate detector
- Exposure time 60 s
- $B = 0 \, \text{T}$, $T = 7.5 \, \text{K}$
- Sample: Ground single crystals embedded in a polymer matrix to suppress grain movement and improve thermal contact.
**Jahn-Teller transition in TbVO$_4$:**

Raw data

- DUBBLE CRG at ESRF
- 21 keV
- MAR 345 image plate detector
- Exposure time $15 \times 5$ ms
- $B = 30$ T, $T = 7.5$ K
- Sample:
  Ground single crystals embedded in a polymer matrix to suppress grain movement and improve thermal contact.
Jahn-Teller transition in $\text{TbVO}_4$: $2\theta$ scans

- High temperature: small splitting induced by magnetic field.
- Low temperature: Splitting reduced by magnetic field.

→ Complex average over phase diagram because of powder average
Interpretation (qualitative)

• The system is driven by the Tb $4f$ quadrupole moment

\[ \epsilon \propto Q_{xy} = \frac{1}{2} (J_x J_y + J_y J_x) \]

Very strong L-S coupling in rare earths
links magnetic moment and charge distribution.

• Coupling between quadrupole and magnetic dipole induced by magnetic field.

→ $B \parallel (001)$: Magnetization $\propto J_z$ in competition with $Q_{xy}$.

→ $B \parallel (110)$: Magnetization $\propto (J_x + J_y)$ increases $Q_{xy}$.

• In a powder sample: Average over all possible directions

→ Average over different phase diagrams

• Working on quantitative data analysis with Z. A. Kazeï.
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**Toulouse 30T magnet system: Second generation**

- New coil design for increased optical access (J. Billette, LNCMP)
  - Coil wound onto a double-cone
  - Opening angle up to $31^\circ$
  - More powder lines available for measurement

- Installation on undulator beamline ID20
  - $\approx \times 50$ gain in intensity
  - Generator installed outside the radiation hutch
    - First tests on the beamline 08–14/11/2006
      - Sufficient intensity with 2–4 msec exposure time
      - Need only one shot per spectrum
Toulouse 30T magnet system: Second generation
Miniature pulsed magnetic field coils
(Peter van der Linden, Olivier Mathon)

Successfully tested in X-ray magnetic circular dichroism (XMCD) experiments on ID24

- Very compact system, can be installed on any beamline.
- Rise time $250 \, \mu s$, one pulse every $10 \, \text{sec.}$
Nuclear resonant forward scattering using pulsed magnetic fields
(Cornelius Strohm, Peter van der Linden, Rudolf Rüffer)

- Nuclear Resonant Forward Scattering of $^{57}\text{Fe}$ foil
- Using mini-coil system
- Total data acquisition time $\approx 8\text{ h}$
Future developments

Short term:

→ The technical solution we are using now has a lot of potential.

→ Significant improvements are necessary before this can become a standard experiment with a user program.

→ For most experiments a split coil geometry with $\vec{B} \perp \vec{k}$ is desired.

→ Try other x-ray techniques: Spectroscopy (EXAFS, XMCD), Laue diffraction can be done by installing our equipment on different beamline.

Medium/long term:

→ Need to improve the detection efficiency. Fast 2D pixel detector?

→ Very low temperatures, down to $100 \text{ mK}$.

→ Higher field, up to $60 \text{ T}$. Improved duty cycle of the magnet system.

→ A permanent setup for capacitor banks, optimized detection system, etc.
**Summary/Conclusions**

- X-ray diffraction under high magnetic fields is virtually virgin ground. There is plenty to be done.
- Steady magnetic fields have the advantage that we can use proven measurement strategies, measure very small signals, etc.
- Pulsed magnetic fields require much more development of x-ray diffraction.
- But because of sample volume, time structure, etc, they can boldly go where no neutron has gone before (and very likely will ever go*).

→ There is a scientific case for both of them.

→ Steady fields solution is lower risk, but limited to 30–40 T.

→ Pulsed fields solution is much more speculative. But it also requires less capital investment, and the ms time resolved x-ray techniques may be of interest in other fields, such as on-line chemistry, shock waves, . . . .

* . . . with the possible exception of neutron stars!