Field induced phase transition in $\text{Ca}_2\text{FeReO}_6$ double perovskite
an XMCD study in 30T pulsed magnetic field

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X-ray Magnetic Circular Dichroism

Difference in the spectral shape of absorption spectra acquired at opposite relative orientation of photon helicity and sample magnetization

Proportional to the difference in the spin-up and spin-down DOS above $E_F$

Element selective probe of localized magnetic moments

$$\Delta \mu = \mu^+ - \mu^-$$
XMCD magnetometry & imaging

Assuming no change in the spectral shape element specific $M(B,P,T)$ profiles can be measured

High resolution, element specific imaging of magnetic domains

Sum Rules

B.T.Thole et al., PRL 68 (1992) 1943
P.Carra et al., PRL 70 (1993) 694

\[
L_z = -\frac{4}{3} \cdot n \cdot \frac{\int (\Delta \mu) dE}{\int (\mu_0) dE} \quad \frac{L_3 + L_2}{L_3 + L_2}
\]

\[
S_z = -n \cdot \frac{\int (\Delta \mu) dE - 2 \int (\Delta \mu) dE}{\int (\mu_0) dE} \quad \frac{L_3}{L_3 + L_2}
\]

where \( n \) denotes the number of holes in the final states

Full spectra necessary at the energy step \( \sim \) lifetime broadening
Sum Rules for Re $L_{2,3}$-edges

$$\frac{m_L}{m_S} \approx \frac{L_z}{2S_z} \sim \frac{1}{3}$$

- Unoccupied d-like final states with...
- Pure spin polarisation
- Pure orbital polarisation

$T=10K, B=2T$
High magnetocrystalline anisotropy → high saturation magnetization and coercive field

Pulsed magnetic field generation

High, steady field magnets are huge and very expensive. Max. at SR facility: 17T at Spring-8.

Higher field may be generated at low cost using pulsed technique. Max. at SR facility: 40T at Spring-8, 30T at ESRF.
Portable pulsed field setup at ESRF

High duty cycle minicoil

- monolithic
- slit coil

Cooling surface

C = 1 mF  L = 20 \mu H
U = 2650 V  I = 13 000 A

Duty cycle: $1 \cdot 10^{-4}$
B = 30 (38) T
rep. rate: 6/min
at working T: 120K

Portable pulsed field setup at ESRF

A: base
B: window block
C: vacuum shroud
D: coil cup
E: chimney
F: sample cryostat
G: sample holder

Continous flow sample cryostat

He in

He out

Operating temperature 5–300K

Pulsed fields at ED beamline

<table>
<thead>
<tr>
<th>Beamline type</th>
<th>Monochromatic</th>
<th>Energy dispersive</th>
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<tbody>
<tr>
<td>Circular polarization</td>
<td>ID, QWP</td>
<td>QWP</td>
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<td>Spectral distortions</td>
<td>Sensitive for highly non-homogenous samples only</td>
<td>Sensitive to beam motions, very sensitive for non-homogenous samples</td>
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<tr>
<td>sample or beam related</td>
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<tr>
<td>Detection techniques</td>
<td>Transmission, fluorescence, TEY</td>
<td>Transmission only?</td>
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<tr>
<td>Systematic errors</td>
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<td>low</td>
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<tr>
<td>due to ring current decay</td>
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<tr>
<td>Number of pulses per spectrum</td>
<td>at least 50</td>
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</tbody>
</table>
Data treatment 1

10K, 30T
LCP

Re $L_3$

VARIATION WITHOUT GIVEN SCAN

NORMALIZED INTEGRAL OF GIVEN SCAN

MAXIMUM / MINIMUM IN THE ENTIRE SCAN RANGE
Data treatment 2

XAS

10K, 30T
LCP

Re $L_3$

XMCD

VARIATION WITHOUT GIVEN SCAN

NORMALIZED INTEGRAL OF GIVEN SCAN

MAXIMUM / MINIMUM IN THE ENTIRE SCAN RANGE
Left & right CP comparison
Systematic error
Double perovskites: $A_2BB’O_6$

- $B’: \text{Mo, Re, W, Os}$
- $B: \text{Fe, Cr, Mn}$

Half doped $B$ site: regularly stacked $BO_6$ and $B’O_6$ octahedra

Ferrimagnetic, metallic double-exchange-like interaction
Magnetoresistive double perovskites

Ferrimagnetic half metals
100% spin polarization

\( T_C \approx 400-750\text{K} \)


Ca\(_2\)FeReO\(_6\) reveals:

High coercivity at low \( T \)

Magnetoresistive double perovskites

Ferrimagnetic half metals
100% spin polarization

$T_C \sim 400$-$750K$

$\text{Sr}_2\text{FeReO}_6$

DOS (states/eV/f.u.)

Energy (eV)

Ca$_2$FeReO$_6$ reveals:

- **High coercivity at low $T$**
- **Large magnetoresistance**
- **Phase coexistence at $T<150K$**


Phase transition in Ca$_2$FeReO$_6$


below $T_S$

above $T_S$
Low and high field XMCD

$L_3$ shape altered by B & T

$L_2$ shape unchanged
Stronger $L_3 \rightarrow$ higher absolute $m_L/m_S$

Similar increase of $L_2$ (XMCD integral) $\rightarrow m_L$ follows bulk magnetization
$m_L/m_S$ evolution over $B$-$T$ space

Within statistical error margin

Unique $m_L/m_S$ expected for given electronic configuration

Relative increase of the absolute $m_L/m_S$ ratio in ‘metallic’ phase

$T<100K$
$\textbf{$m_L/m_S$ evolution}$
Induced by magn. field
$\rightarrow$ phase coexistence

$T>200K$
\textbf{constant $m_L/m_S$} $\rightarrow$ single phase
**Re & bulk magnetization evolution**

$M(B)$ profiles normalized at 30T
→ collinear magn. $T > 200$K
→ excess of Re magnetization at low fields for $T < 150$K

$M(T)$ normalized at high $T$
→ excess of Re magnetization at low temperatures & fields

May be explained by charge redistribution
→ increase in Re population at low $T$ & $B$
$m_L$ & bulk magnetization evolution

$M(B)$ profiles normalized at 30T
\rightarrow collinear magn. $T > 200K$
\rightarrow excess of Re magnetization at low fields for $T < 150K$

$M(T)$ normalized at high $T$
\rightarrow excess of Re magnetization at low temperatures & fields

... or by non-collinear alignment
\rightarrow decrease of projected $M_{Fe}$
Conclusions and perspectives

Re $L_{2,3}$ XMCD spectra acquired up to 30T over wide $T$ range: 10-250K

Field induced phase transition observed in $\text{Ca}_2\text{FeReO}_6$, confirmed phase coexistence

Phase transition associated with charge redistribution and ...

... non-collinear alignment in insulating (low $B$ & $T$) phase

XMCD spectroscopy successfully combined with pulsed generation of magnetic field

A number of 20-50 pulses per spectrum is sufficient

Reliable but complex setup (quick reparation time)

Automatic data selection (correction) techniques to be developed
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