Phase IV in $\text{Ce}_{0.7}\text{La}_{0.3}\text{B}_6$: X-ray Resonant Scattering Results

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Magnetic Properties

LoadStone Magnetic Compass

Fe$_3$O$_4$ Magnetite

Magnetic Data Storage

Multi-billion dollar computer industry
Magnetic Order

Temperature, Pressure, Magnetic fields

\[ S \pm \frac{1}{2} \]

\[ J = L \pm S \]
Multipolar Order

Quadrupole charge order
Antiferroquadrupole order

Magnetic Octupole Order
Very Rare and exotic form of Magnetic Order
CeLaB$_6$, NpO$_2$. 
Multipolar Order

Magnetic Octupole Order

Very Rare and exotic form of Magnetic Order

\[ \text{CeLaB}_6, \text{NpO}_2. \]
X-Ray Single Crystal Diffraction

Bragg Law: \( n\lambda = 2d \sin(\theta) \)

\[ x = d \sin(\theta) \]
X-Ray Single Crystal Diffraction

\[ Q(HKL) = n\lambda = 2d \sin(\theta) \]

Crystal Lattice

\[ k_i, k_f \]
X-Ray Single Crystal Diffraction

Crystal Lattice

\[ \theta \]

\[ d = \frac{n\lambda}{2\sin(\theta)} \]

Detector

(111)

(222)

(333)

Detector

Q(HKL)
X-Ray Single Crystal Diffraction

Thomson Charge Scattering

\[ I_T \approx \left( \frac{Ze^2}{mc^2} \right)^2 \sum_j e^{iQ.r_j} \]

X-ray Magnetic Scattering

\[ I_m \approx -i \left( \frac{\hbar \omega}{mc^2} \right)^2 \left( \frac{N_m}{Z} \right)^2 \left( \frac{e^2}{mc^2} \right)^2 \sum_j e^{iQ.r_j} (L, \theta + S, B)^2 \]

\[ \left( \frac{\hbar \omega}{mc^2} \right)^2 \left( \frac{N_m}{Z} \right)^2 = \left( \frac{1 \times 10^4 \text{eV}}{0.511 \times 10^6 \text{eV}} \right)^2 \left( \frac{7}{26} \right) \]
Synchrotron Radiation ESRF

6 GeV Storage Ring
844M Circumference
40 Beamlines

Huge flux $10^{12} - 10^{13}$ photons/sec
High linear polarisation $\sim 100\%$
E1E1 X-Ray Resonant Scattering at Ce L$_2$ edge

Energy = $\frac{hc}{\lambda} \approx 6.164$ keV

$\lambda$  

$5d \quad \text{E1E1 } \Delta L = \pm 1$

$4f$

$2p^{3/2}$  

$2p^{1/2}$  

Ce L$_3$ edge 5.723 keV  

Ce L$_2$ edge 6.164 keV
E2E2 X-Ray Resonant Scattering at Ce L$_2$ edge

Energy = $\frac{hc}{\lambda}$

$\sim 6.164$ keV

$\lambda$
Detector

\( k_i \)

\( k_f \)

Energy vs. Intensity

Intensity

Energy

5d

4f
E1E1 XRS from multipole order

Detector

Antiferro-quadrupole order

Anisotropic Tensor Susceptibility ATS

$k_i$ $k_i$

5d E1E1 $\Delta L=\pm1$

4f

$2p^{3/2}$ $2p^{1/2}$
E2E2 XRS from multipole order

Antiferro-quadrupole order
Magnetic Octupole Order

5d E1E1 $\Delta L=\pm 1$
4f E2E2 $\Delta L=\pm 2$

$2p^{3/2}$ $2p^{1/2}$
X-ray Polarisation Dependence

\[ \sigma_i = \sigma_f \]

Diagram showing the path of X-rays through a sample, analyser, and detector. The incident beam is denoted by \( e_i = \sigma \) and the final beam by \( e_f = \sigma \). The crystal is at an angle of 45° and the sample is at angle \( 2\theta \).
Azimuthal Dependence

e_i = \sigma

X-rays \rightarrow k_i

2\theta

\theta

Sample

k_f

Detected

\epsilon_f = \pi

Analyser

Crystal 45°
Azimuthal Dependence

\[ F_{\text{XRMS}} = f_{\text{XRS}} [(e_i x e_f) \cdot M] \]

\[ \sigma \xrightarrow{x} \sigma = 0 \]

\[ \sigma \xrightarrow{x} \pi = -k_f \cdot M \quad \sin(\theta) \quad \cos(\theta) \]

\[ e_f = \pi \]

X-rays →

\[ e_i = \sigma \]

Sample

Detector

Analyser

Crystal 45°
Phase IV: $T<T_{IV}=1.5\text{K}$

- I Paramagnetic
- II Antiferroquadrupole order $q=(\frac{1}{2} \frac{1}{2} \frac{1}{2})$
- III Antiferromagnetic order $q=(\frac{1}{4} \frac{1}{4} \frac{1}{2})$
- IV – Proposed new phase

Phase IV ground state has remained elusive.
Enigmatic Phase IV

Specific Heat:
Large anomaly in specific heat is indicative of long range order.
Cusp in magnetic susceptibility: Antiferromagnetic Order?

No Magnetic Structure has been reported by neutron scattering.
Antiferroquadrupole Order?

Large softening of $c_{44}$ elastic constant.

This does not happen in pure CeB$_6$ in the AFQ phase?

? Magnetic Octupole Order in Phase IV?

RXS study of Phase IV

\[ \text{Ce}_{0.7}\text{La}_{0.3}\text{B}_6 \quad \text{Ce L}_2 \quad \text{RXS} \]

![Graph showing intensity vs. energy with peaks at 4f and 5d transitions.](image-url)
E1 and E2 Thermal and Spatial Independence!

\[ I = |T - T_{IV}|^{2\beta} \]

\[ \beta(5d) = 0.33 \]

\[ \beta(4f) = 0.99 \]

T-dep consistent with:

- E1 Dipole
- E2 Octupole
Azimuth dependence at 1.0 Kelvin

- E1 $\sigma\pi$ Only $\rightarrow$ Dipole
- NO AFQ order $\rightarrow$ Phase IV is new phase!
- E2 $\sigma\sigma$ 6-fold azimuth
- E2 $\sigma\pi$ 3-fold azimuth
Octupole Order of $T_{1u}$ ($\Gamma_{4u}$) symmetry elements


\[
f_{nE^2}(\Phi) = \sum_{i=1}^{3} \left[ -iF_{E^2}^{3} \right] \left[ (k_f \cdot Z_n^i(\Phi)) (k_i \cdot Z_n^i(\Phi)) (\epsilon_f \times \epsilon_i) \cdot Z_n^i(\Phi) + (\epsilon_f \cdot Z_n^i(\Phi)) (\epsilon_i \cdot Z_n^i(\Phi)) (k_f \times k_i) \cdot Z_n^i(\Phi) \right] + (\epsilon_f \cdot Z_n^i(\Phi)) (k_i \cdot Z_n^i(\Phi)) (k_f \times \epsilon_i) \cdot Z_n^i(\Phi) + (k_f \cdot Z_n^i(\Phi)) (\epsilon_i \cdot Z_n^i(\Phi)) (\epsilon_f \times k_i) \cdot Z_n^i(\Phi) \]
$I(\text{No PA}) = I(\sigma\sigma) + I(\sigma\pi)$

$\sigma\sigma$ intensity larger than $\sigma\pi$

$\Gamma_5$ Octupole model with equal domain population

Kusunose and Kuramoto
Quadrupole vs Octupole Order

Nagao and Igarashi
Conclusions

1. The first microscopic study of phase IV using RXS → Compact XMaS 1K cryostat – azimuth scans.

4. Thermal and spatial independence of E1 & E2 RXS → evidence two order parameters

7. 5d short range AFM order → below $T_{IV}=1.5K$ and above (~3K) at $q=(\frac{1}{2} \frac{1}{2} \frac{1}{2})$

4. Simple model for E2 T-dep, azimuth & Bragg dependence:
   → 4f octupole order with $T_{1u}$ symmetry elements
   → at $q=(\frac{1}{2} \frac{1}{2} \frac{1}{2})$ below $T_{IV}$ only.

14. No evidence for AFQ order in phase IV (No E1E1 $\sigma\sigma$) → direct evidence for new phase


6. Theory: Kusunose and Kuramoto, Nagao and Igarashi, Lovesey and Katsumata. Future experiments at (0.5 0.5 0.5) and Ce L$_3$ edge.