Phase IV in $Ce_{0.7}La_{0.3}B_6$:
X-ray Resonant Scattering Results

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Magnetic Properties

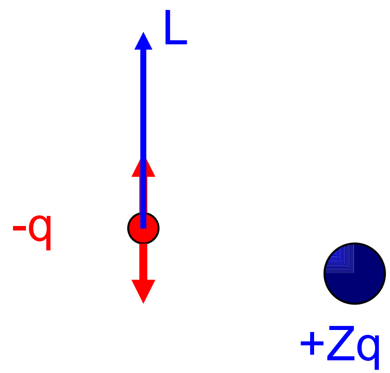
LoadStone Magnetic Compass

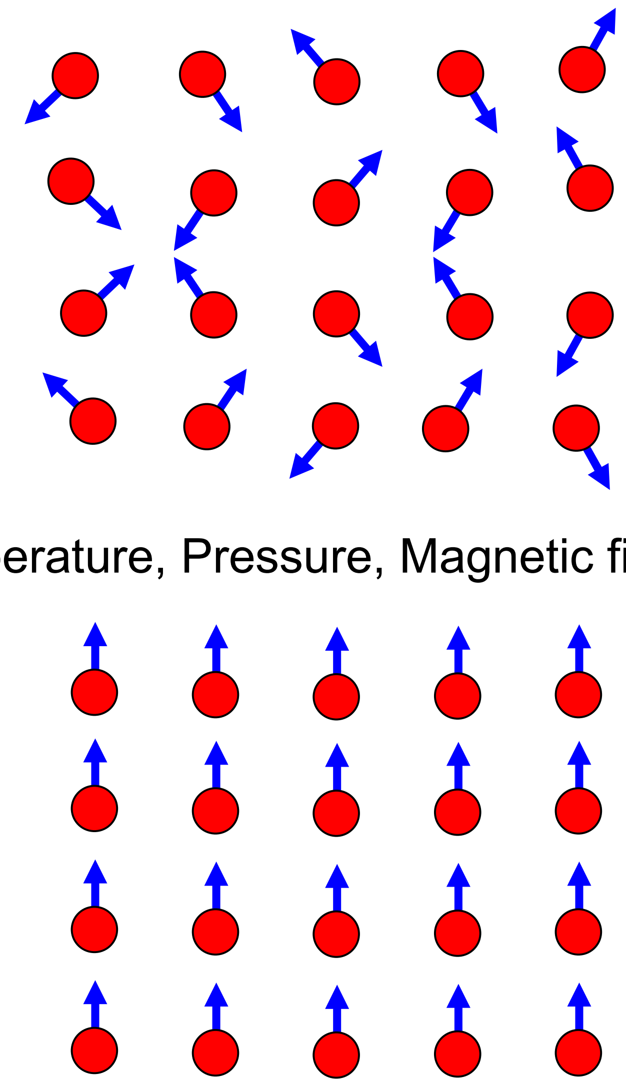
Magnetic Data Storage

Multi-billion dollar computer industry

Fe_3O_4 Magnetite

Magnetic Order

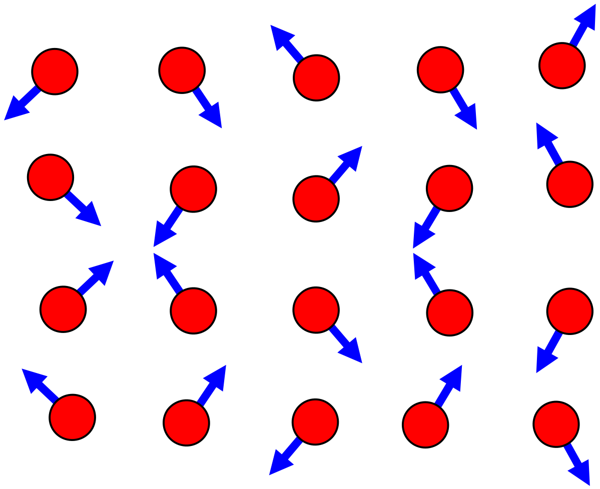


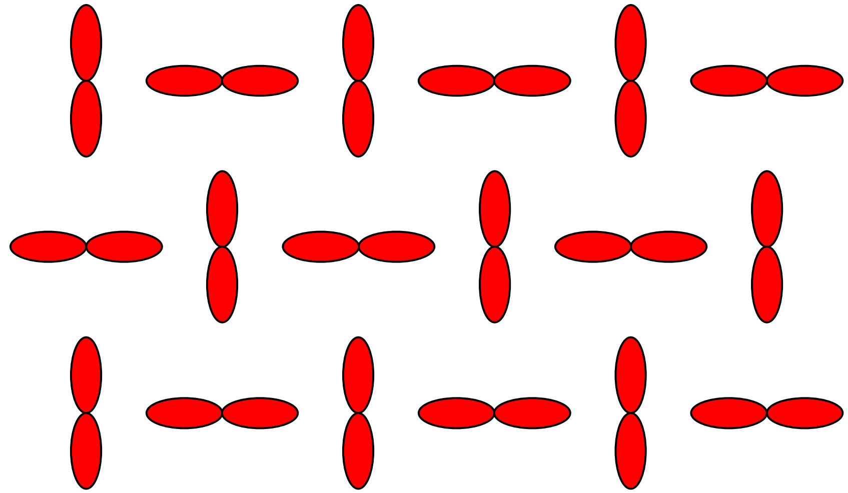


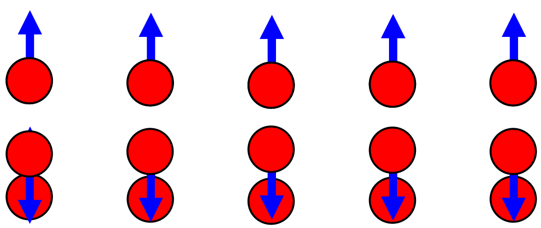
S±½

J=L±S

Multipolar Order

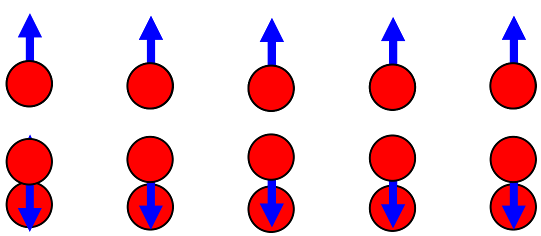






Ferromagnetic

Bulk response



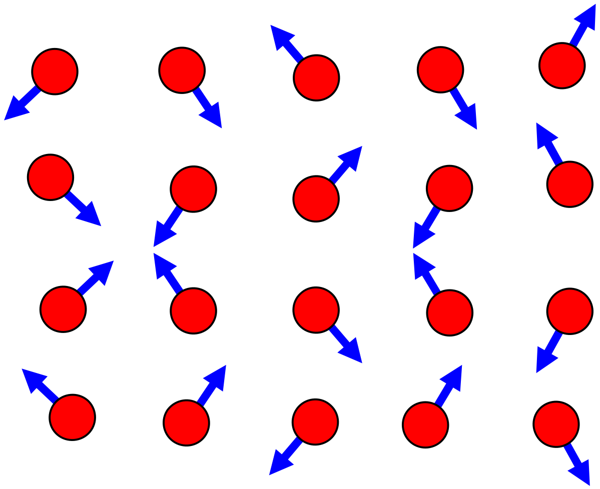
Antiferromagnetism

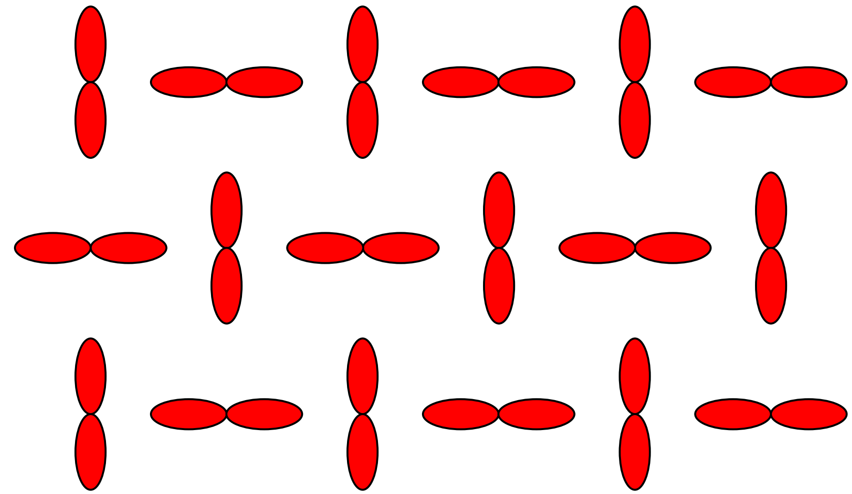
Neutron Scattering

Quadrupole charge order

Antiferroquadrupole order

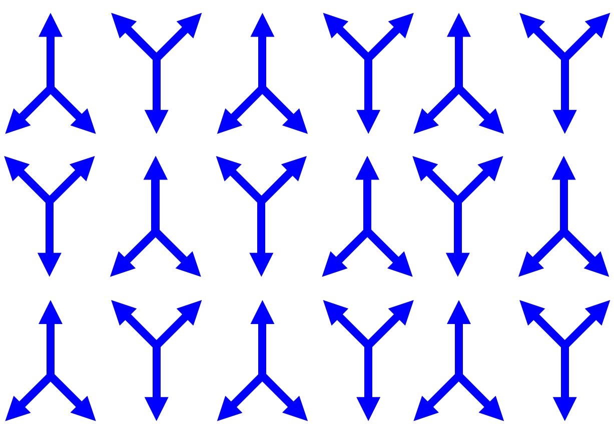
Multipolar Order





Quadrupole charge order

Antiferroquadrupole order

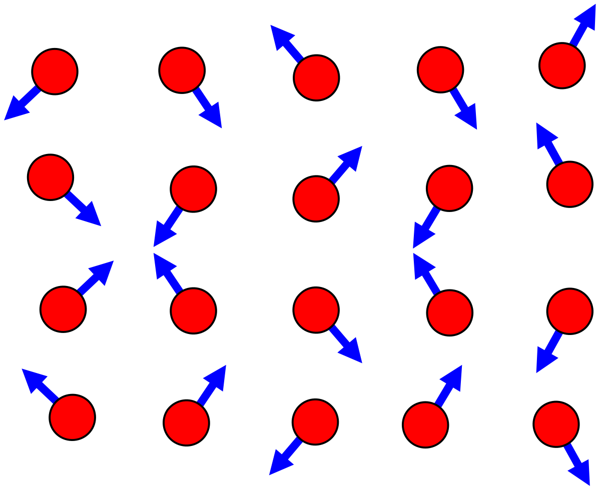


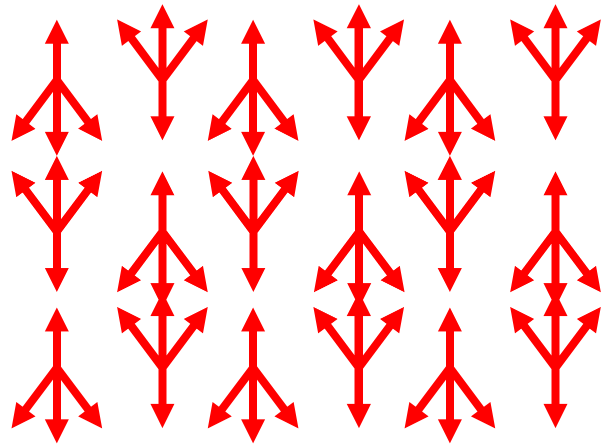
Magnetic Octupole Order

Very Rare and exotic form of Magnetic Order

$CeLaB_6$, NpO_2.

Multipolar Order



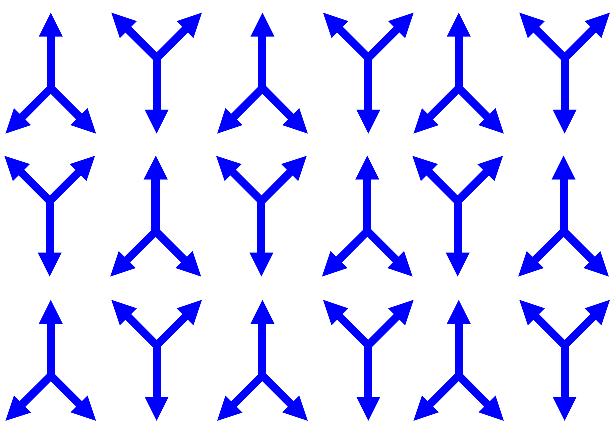


Hexadecapole Charge Order

Multipole Order: large L small S

e.g. Ce, Nd, Tm, Dy, U and Np compounds

RXS probe: Weak interaction for Neutrons



Magnetic Octupole Order

Very Rare and exotic form of Magnetic Order

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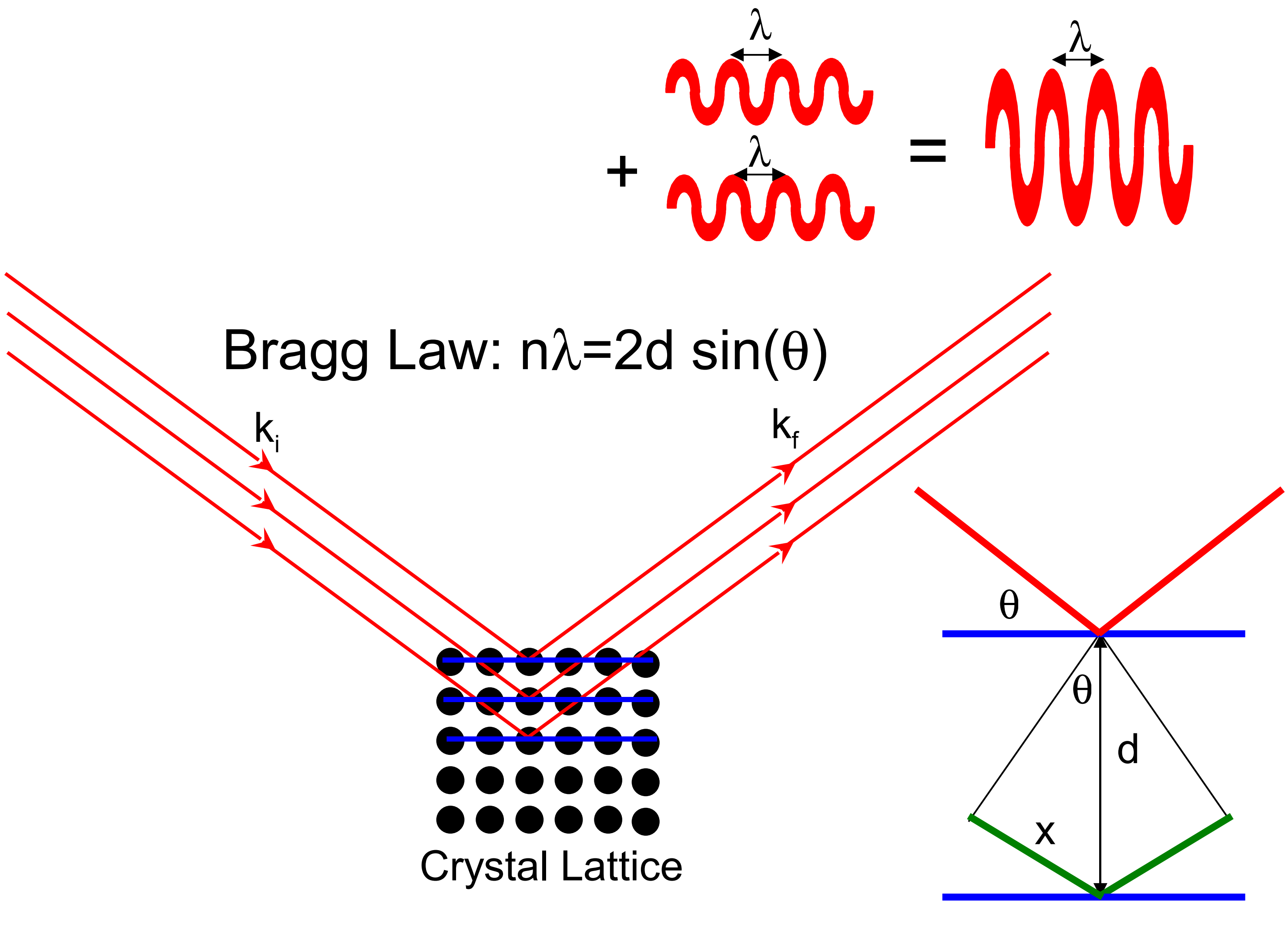
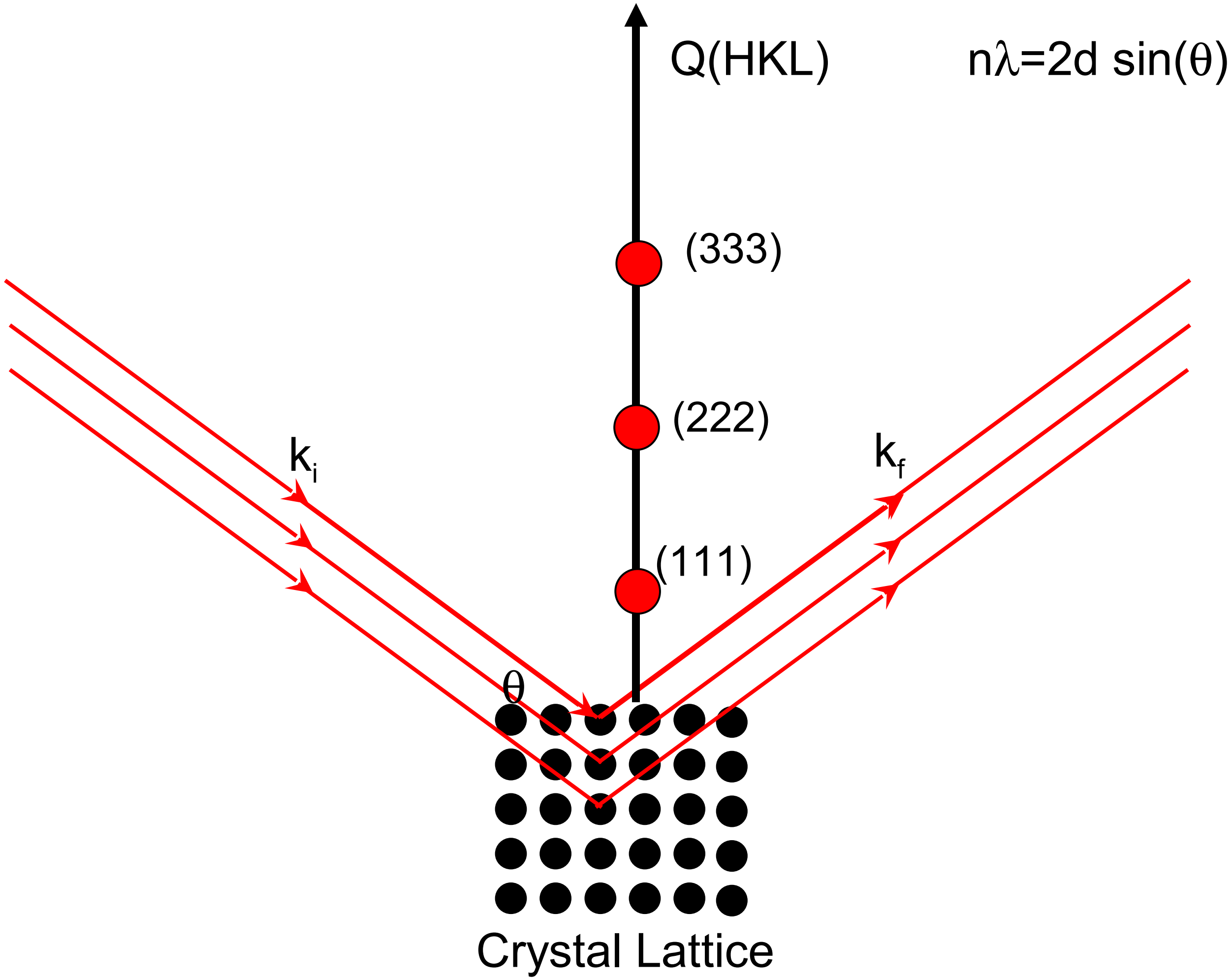
X-Ray Single Crystal Diffraction

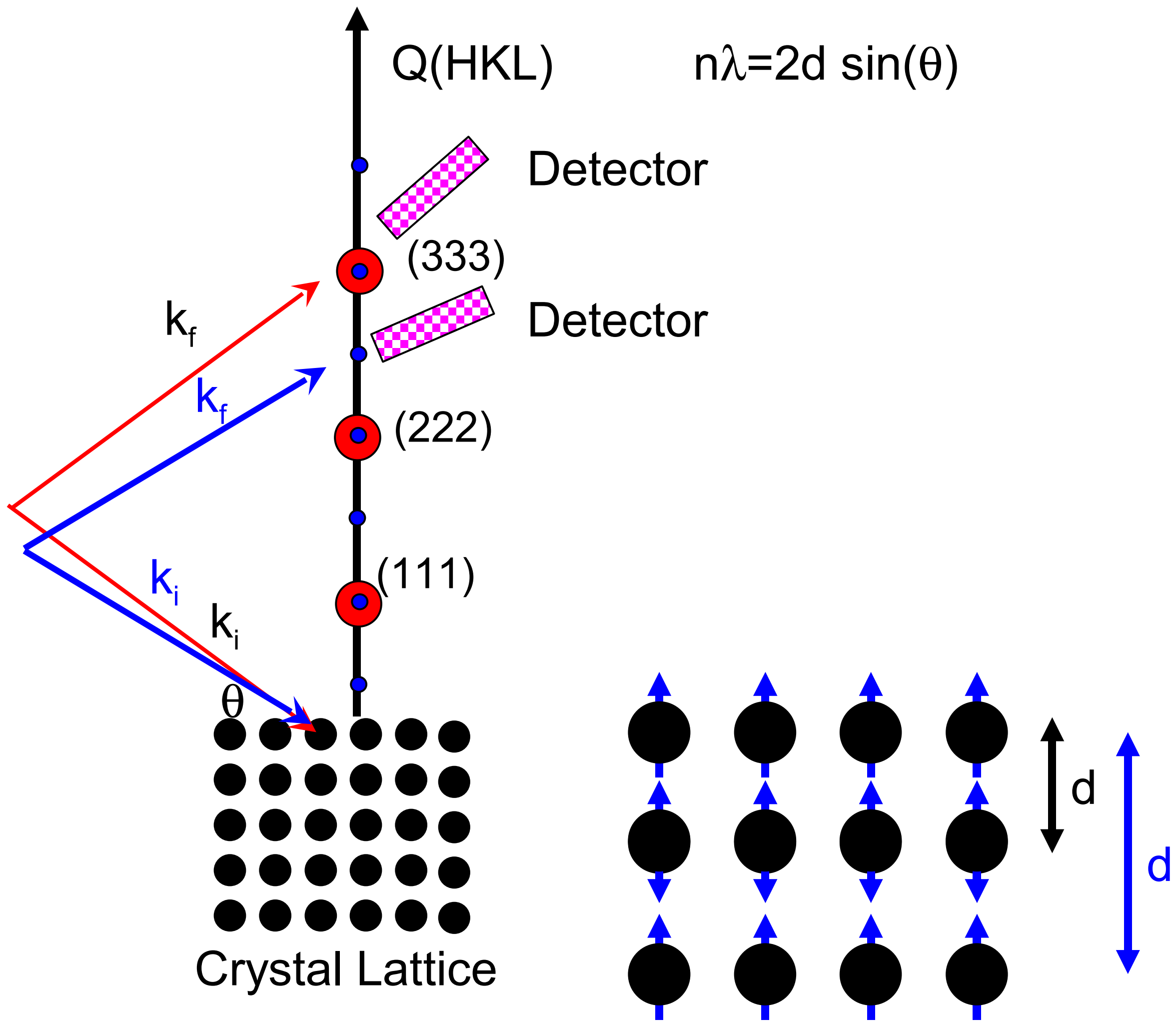


x=d sin(θ)

X-Ray Single Crystal Diffraction



X-Ray Single Crystal Diffraction



X-Ray Single Crystal Diffraction

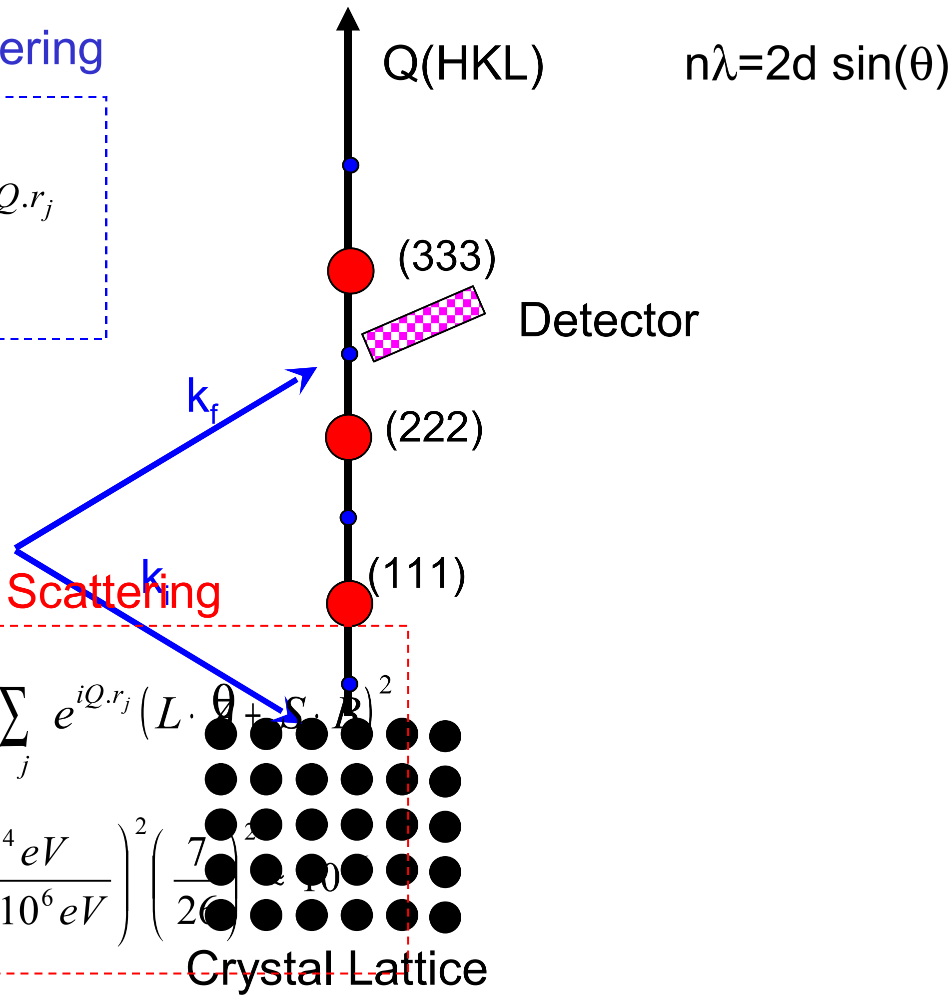
Thomson Charge Scattering

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

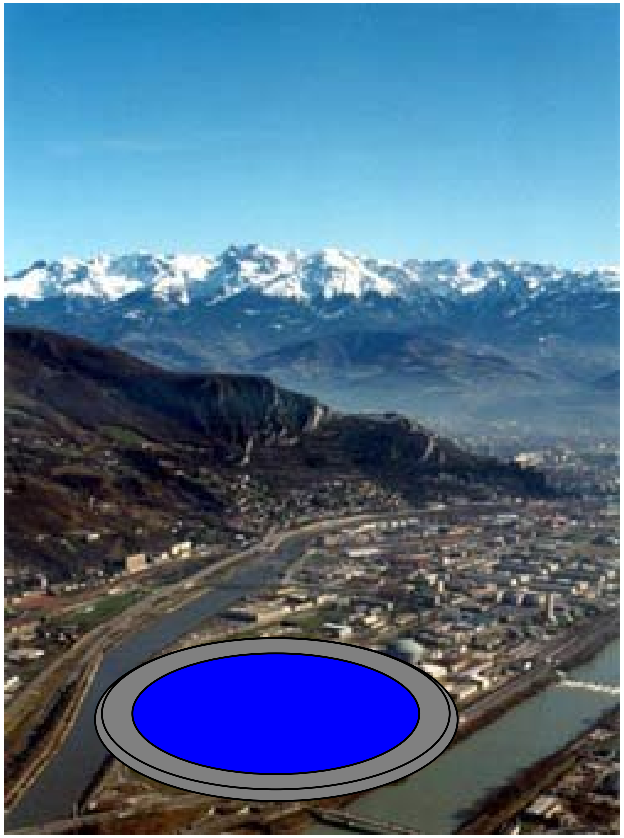
X-ray Magnetic Scattering

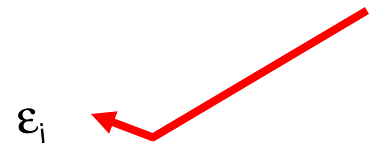
$$I_m \approx -i \left(\frac{\hbar\omega}{mc^2} \right)^2 \left(\frac{N_m}{Z} \right)^2 \left(\frac{e^2}{mc^2} \right)^2 \sum_j e^{iQ.r_j} \left(L \cdot Q + S \cdot B \right)^2$$

$$\left(\frac{\hbar\omega}{mc^2} \right)^2 \left(\frac{N_m}{Z} \right)^2 = \left(\frac{1 \times 10^4 eV}{0.511 \times 10^6 eV} \right)^2 \left(\frac{7}{26} \right)^2 \approx 10$$



Synchrotron Radiation ESRF





6 GeV Storage Ring

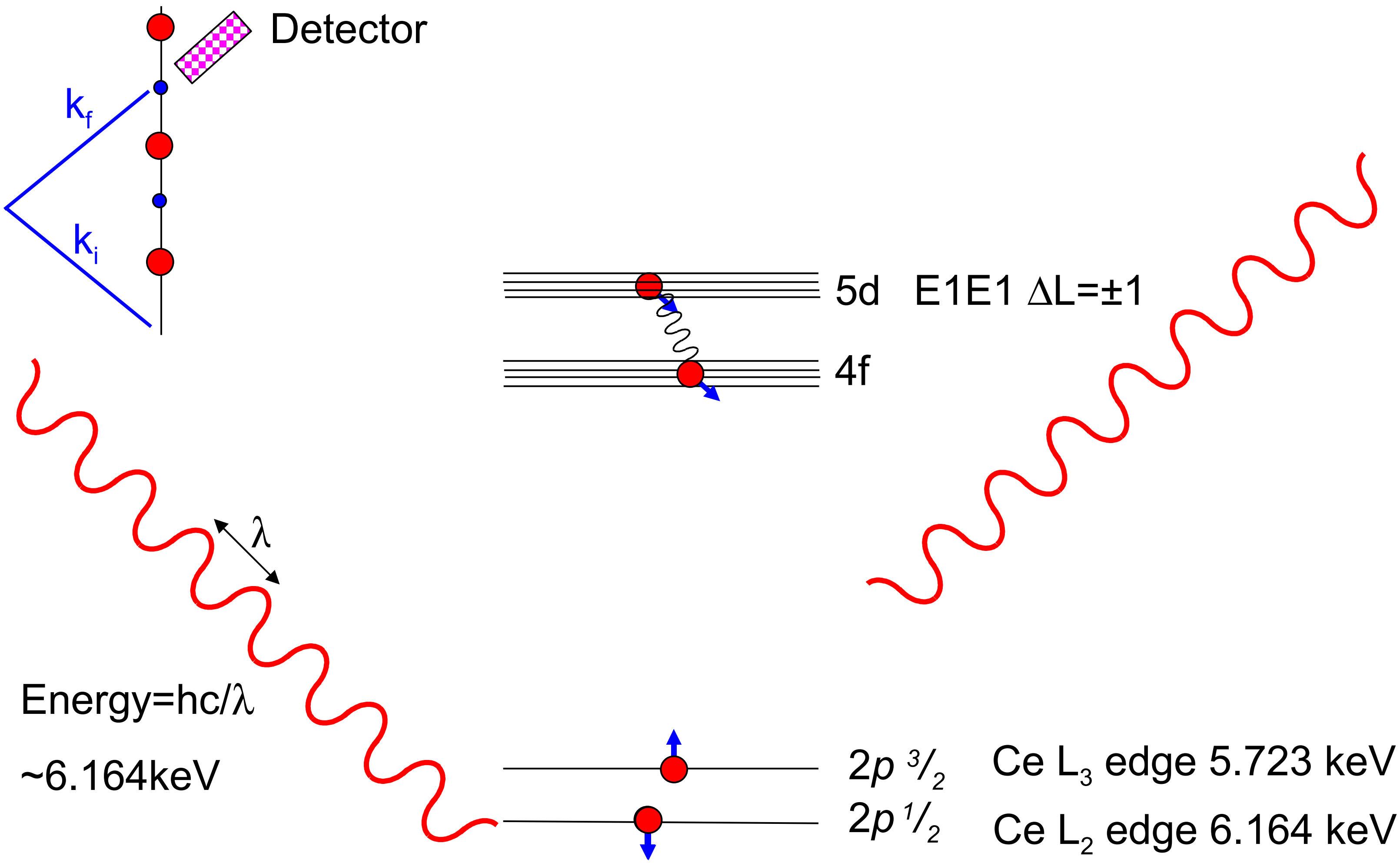
844M Circumference

40 Beamlines

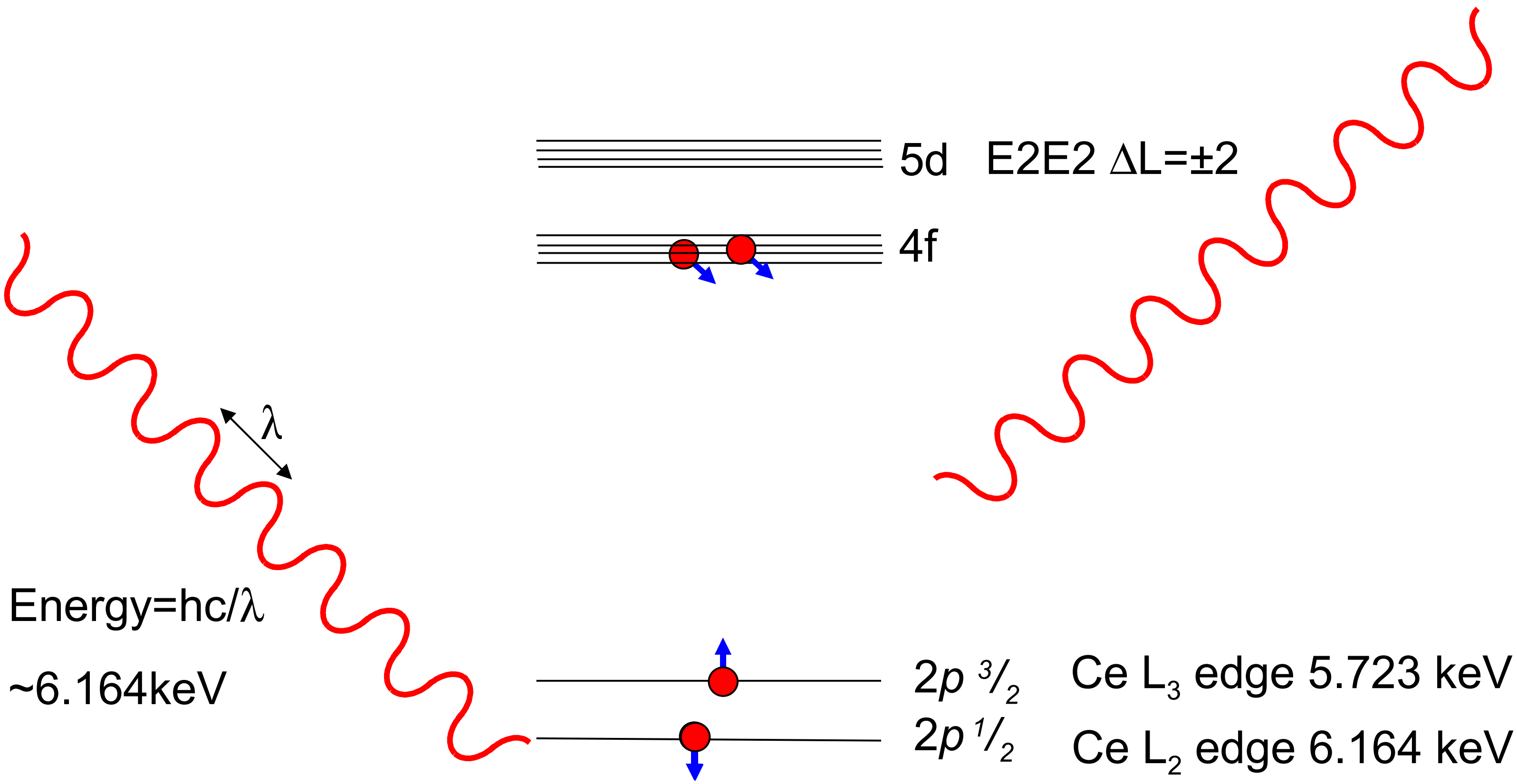
Huge flux $10^{12} - 10^{13}$ photons/sec

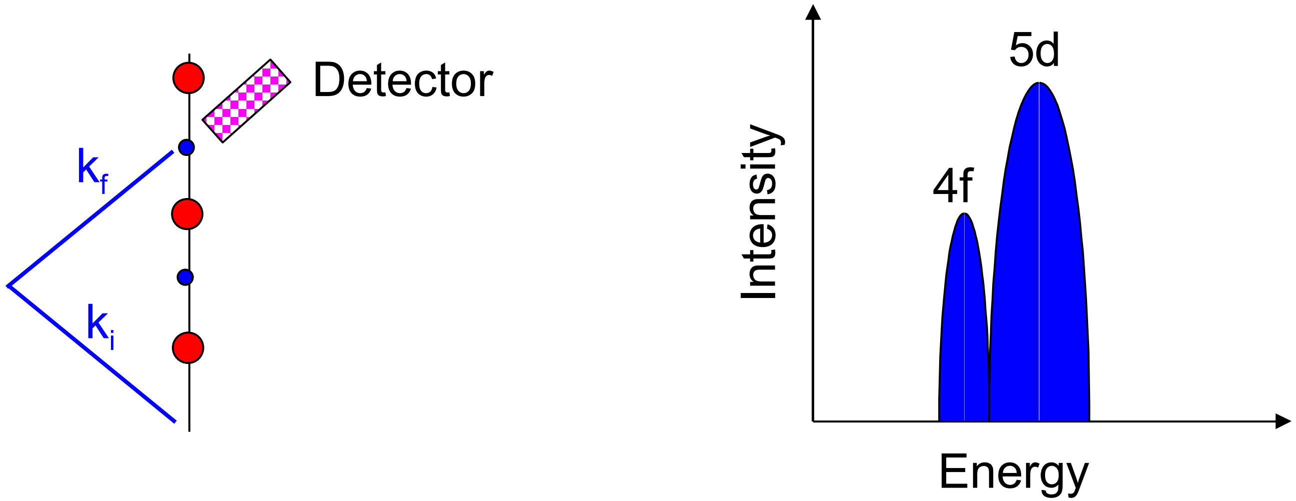
High linear polarisation ~100%

E1E1 X-Ray Resonant Scattering at Ce L_2 edge



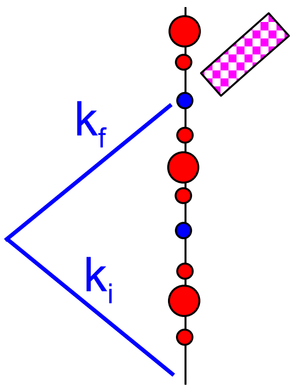
E2E2 X-Ray Resonant Scattering at Ce L_2 edge

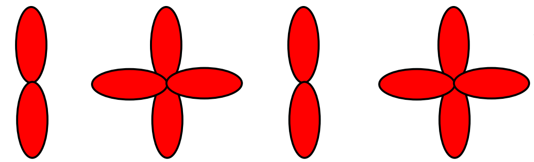


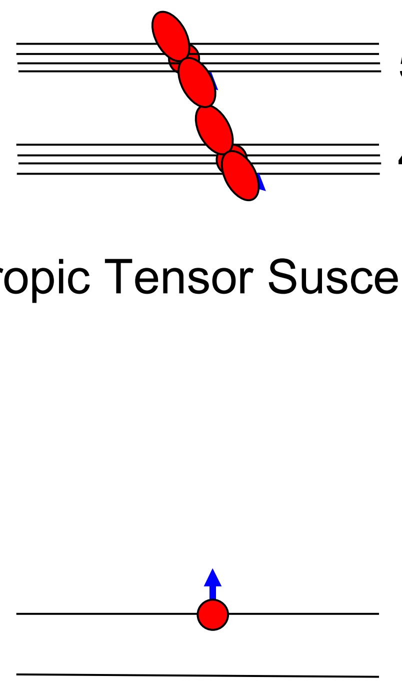


Detector
k_f
k_i
Intensity
Energy
4f
5d

E1E1 XRS from multipole order

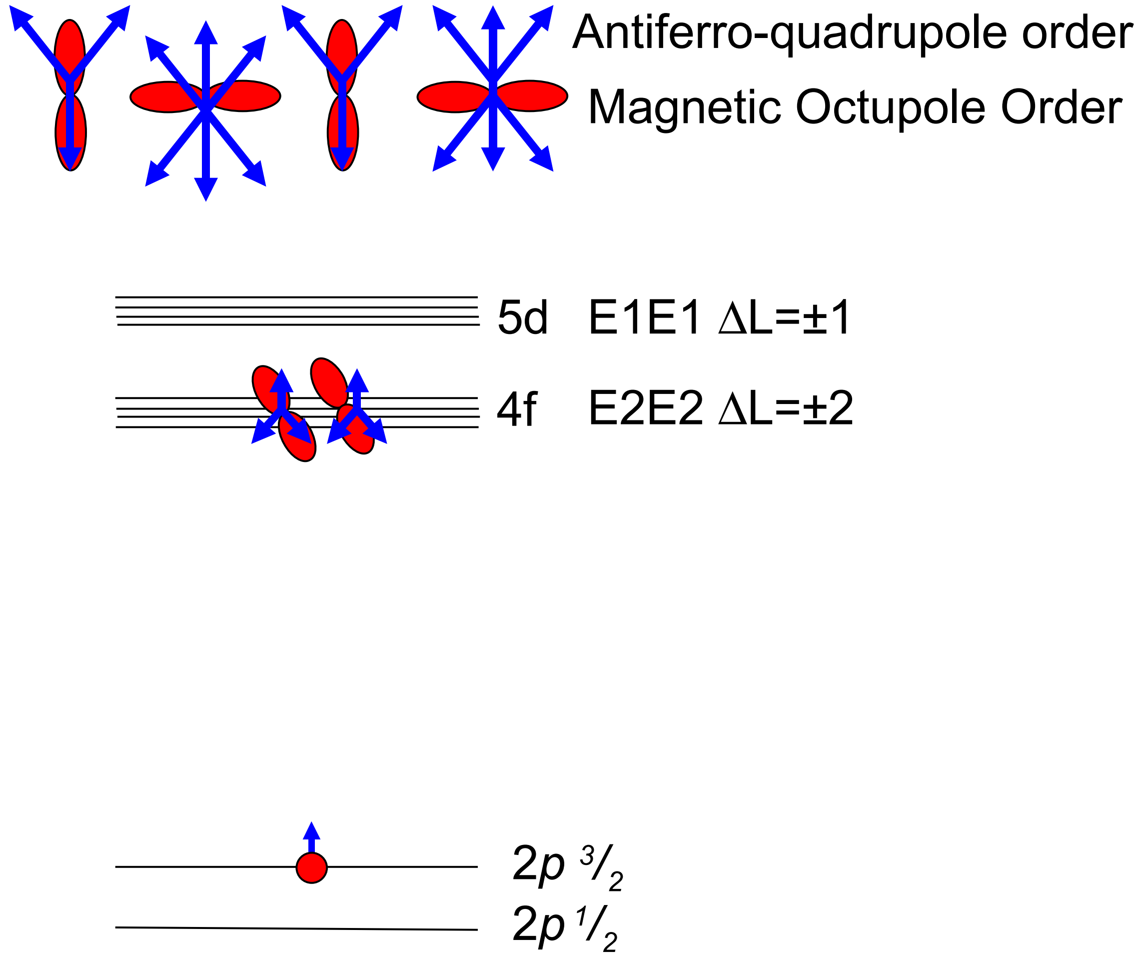




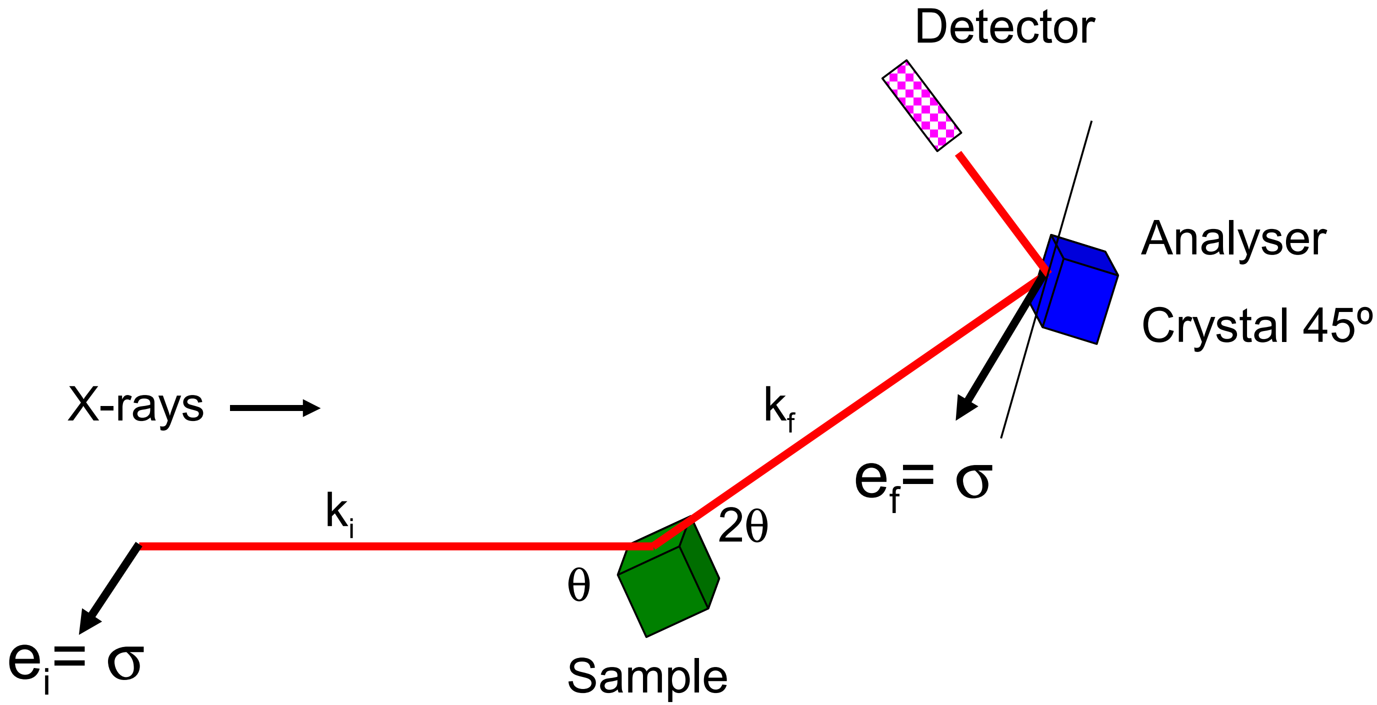


Anisotropic Tensor Susceptibility ATS

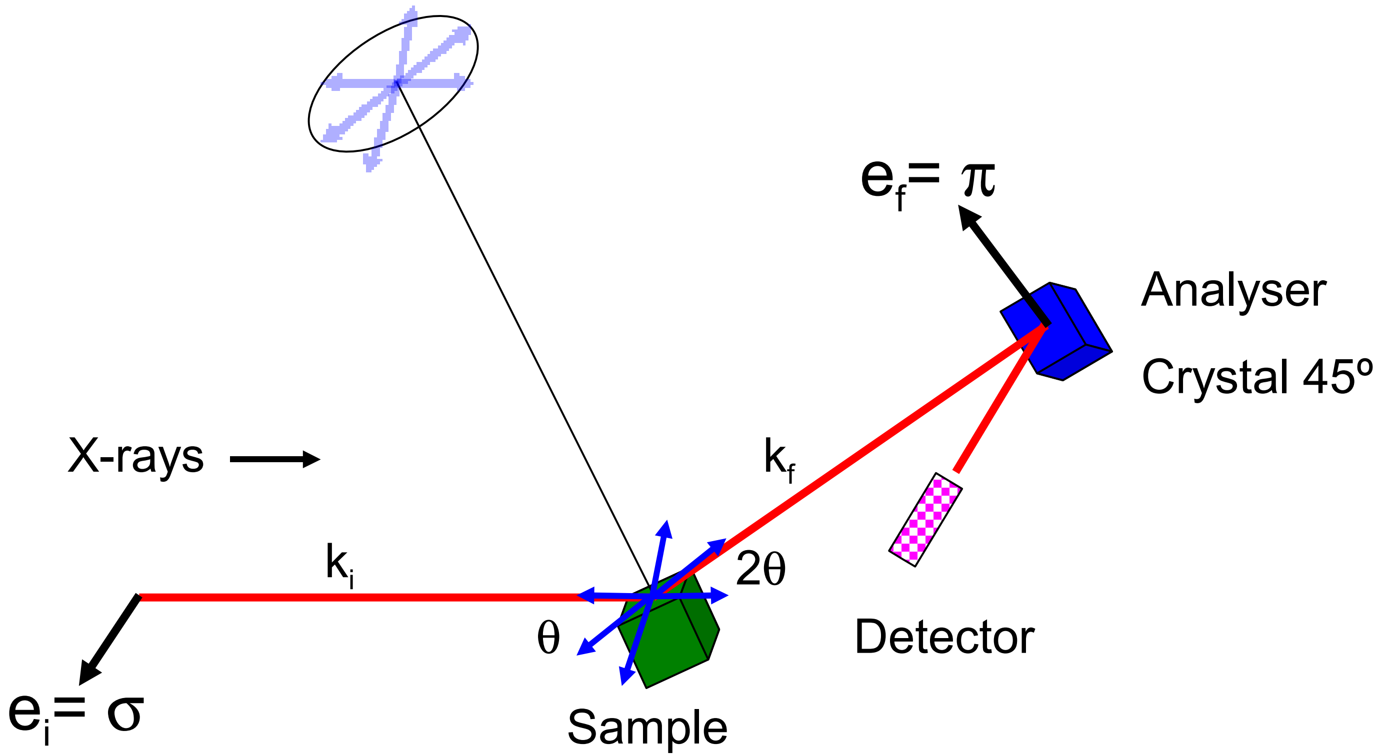
E2E2 XRS from multipole order



X-ray Polarisation Dependence



Azimuthal Dependence

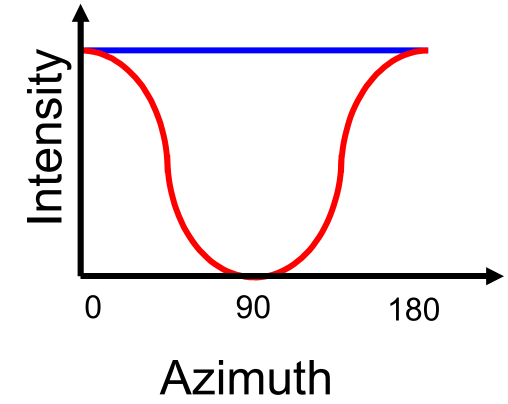


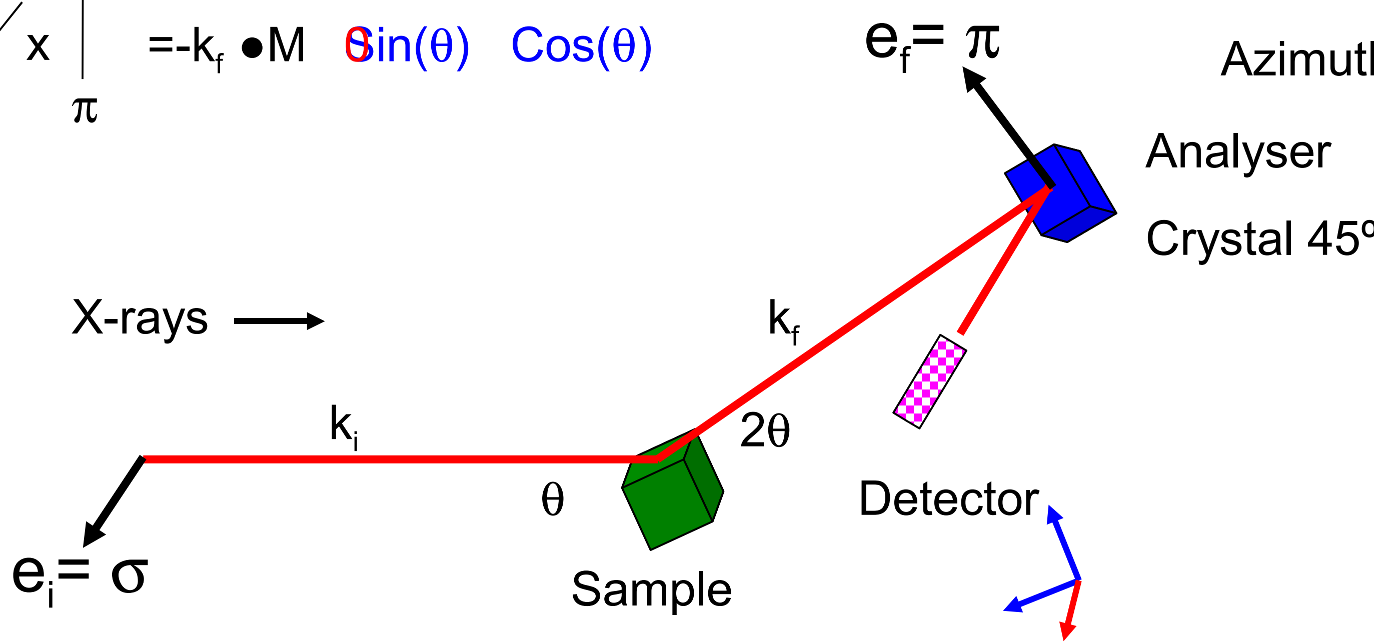
Azimuthal Dependence

$F_{XRMS} = f_{XRS}\,[(e_i x e_f) \bullet M]$

$\sigma \times \sigma = 0$

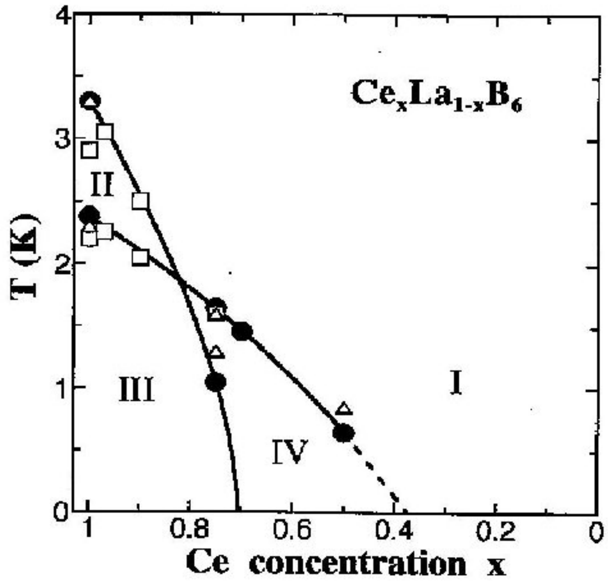
$\sigma \times \pi = -k_f \bullet M$ 0 Sin(θ)   Cos(θ)

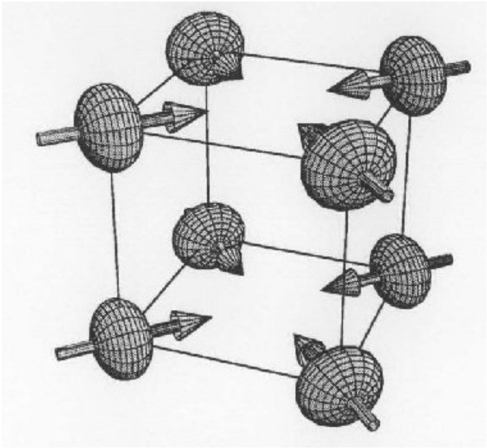




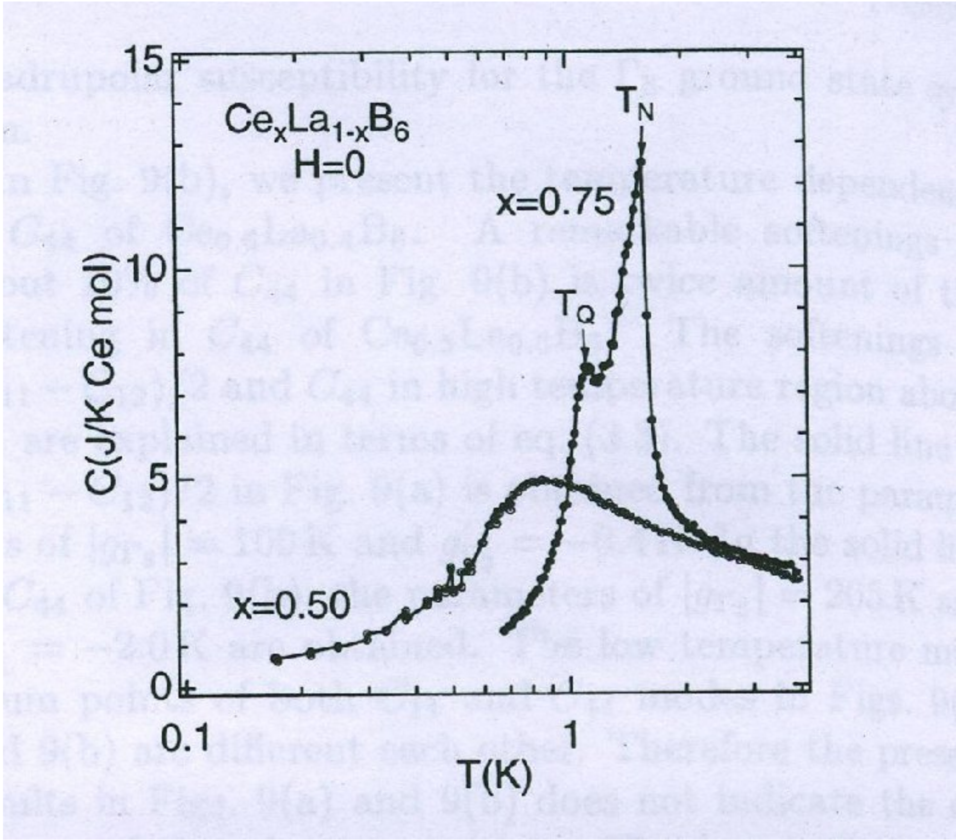
Phase IV: $T<T_{IV}=1.5K$

- I Paramagnetic
- II Antiferroquadrupole order q=(½ ½ ½)
- III Antiferromagnetic order q=(¼ ¼ ½)
- IV – Proposed new phase
- Phase IV ground state has remained elusive.



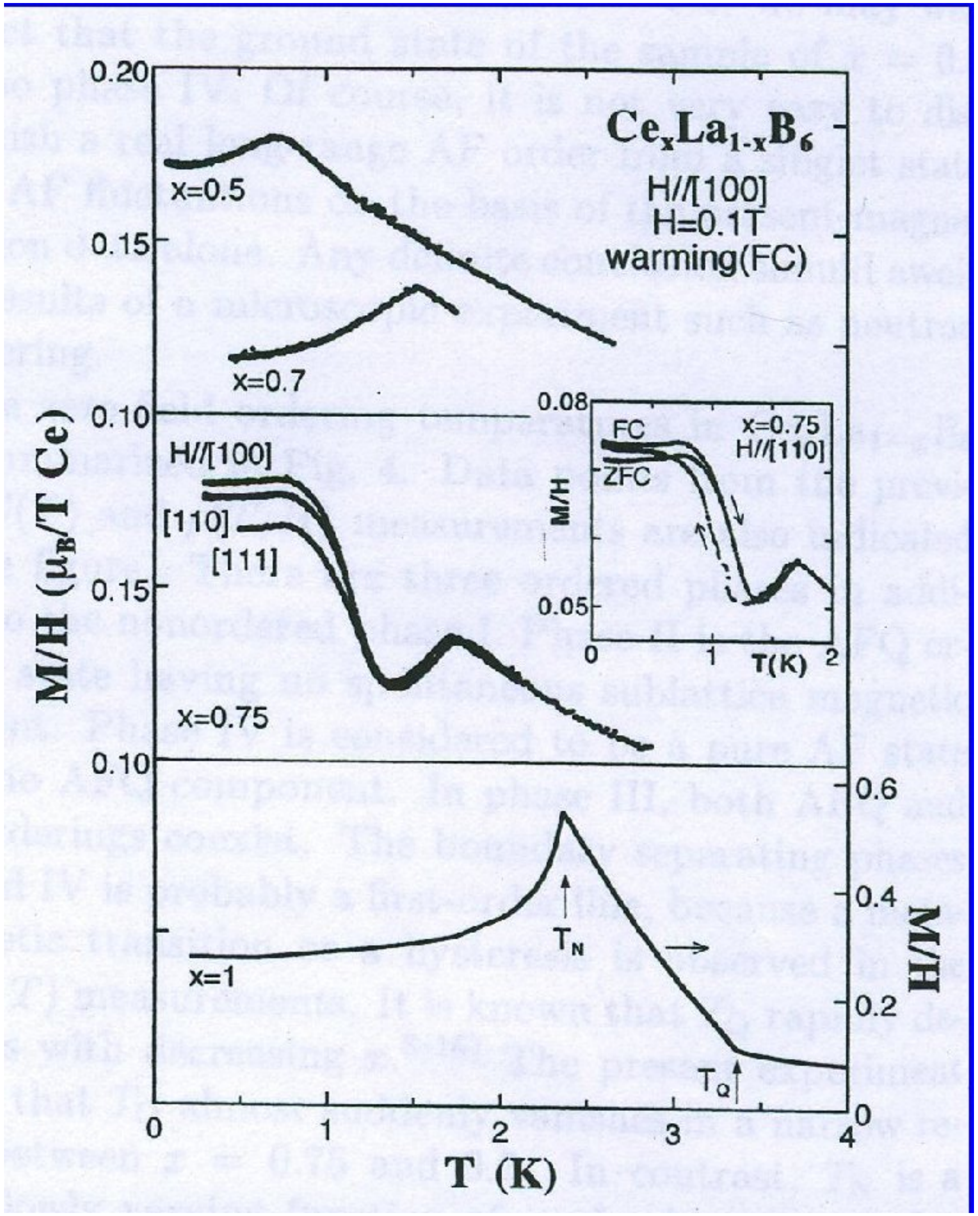


Enigmatic Phase IV



Specific Heat:

Large anomaly in Specidif heat is indicative of long rang order.

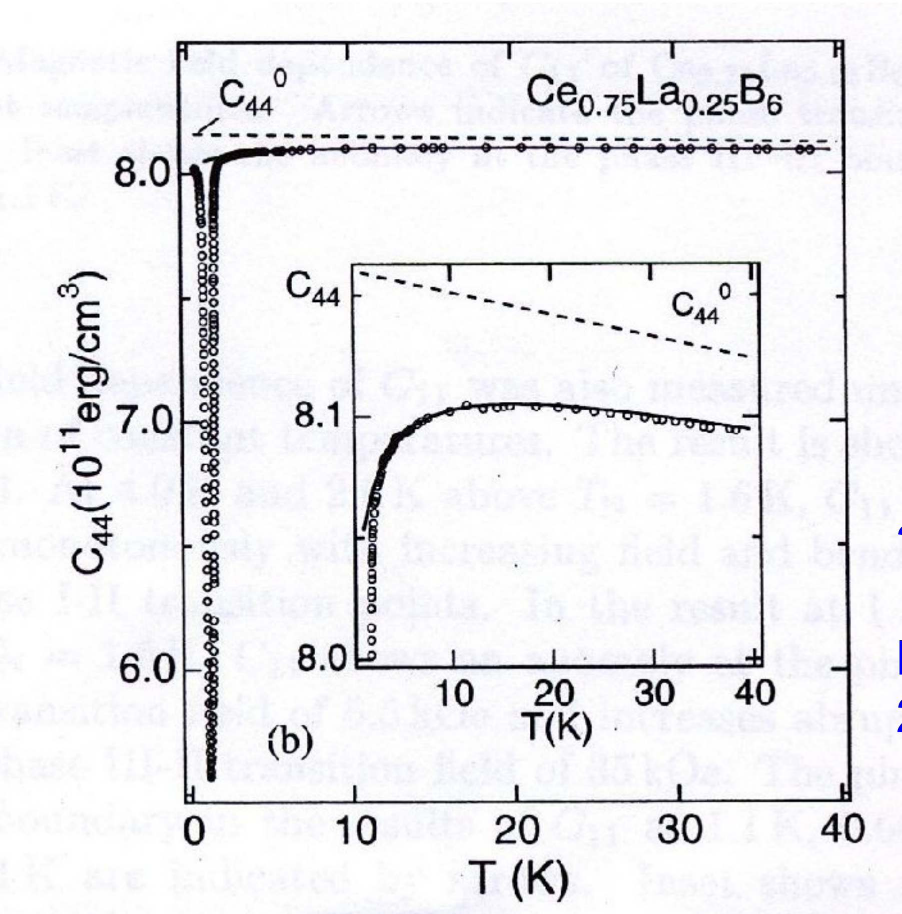


Cusp in magnetic susceptibility:

Antiferromagnetic Order?

No Magnetic Structure has been reported by neutron scattering.

Antiferroquadrupole Order ?



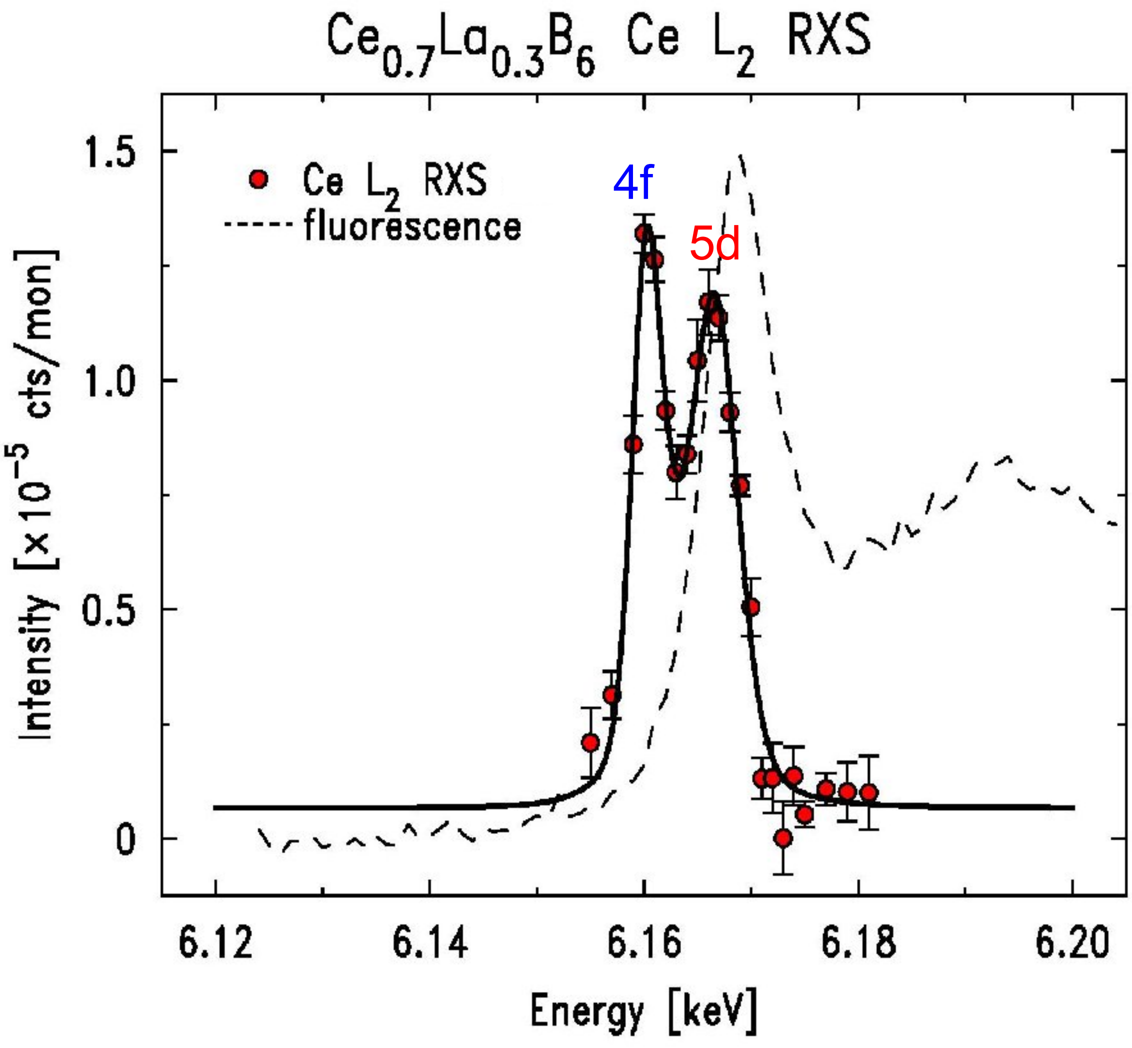
Large softening of c_{44} elactic constant.

This does not happen in pure CeB_6 in the AFQ phase?

? Magnetic Octupole Order in Phase IV?

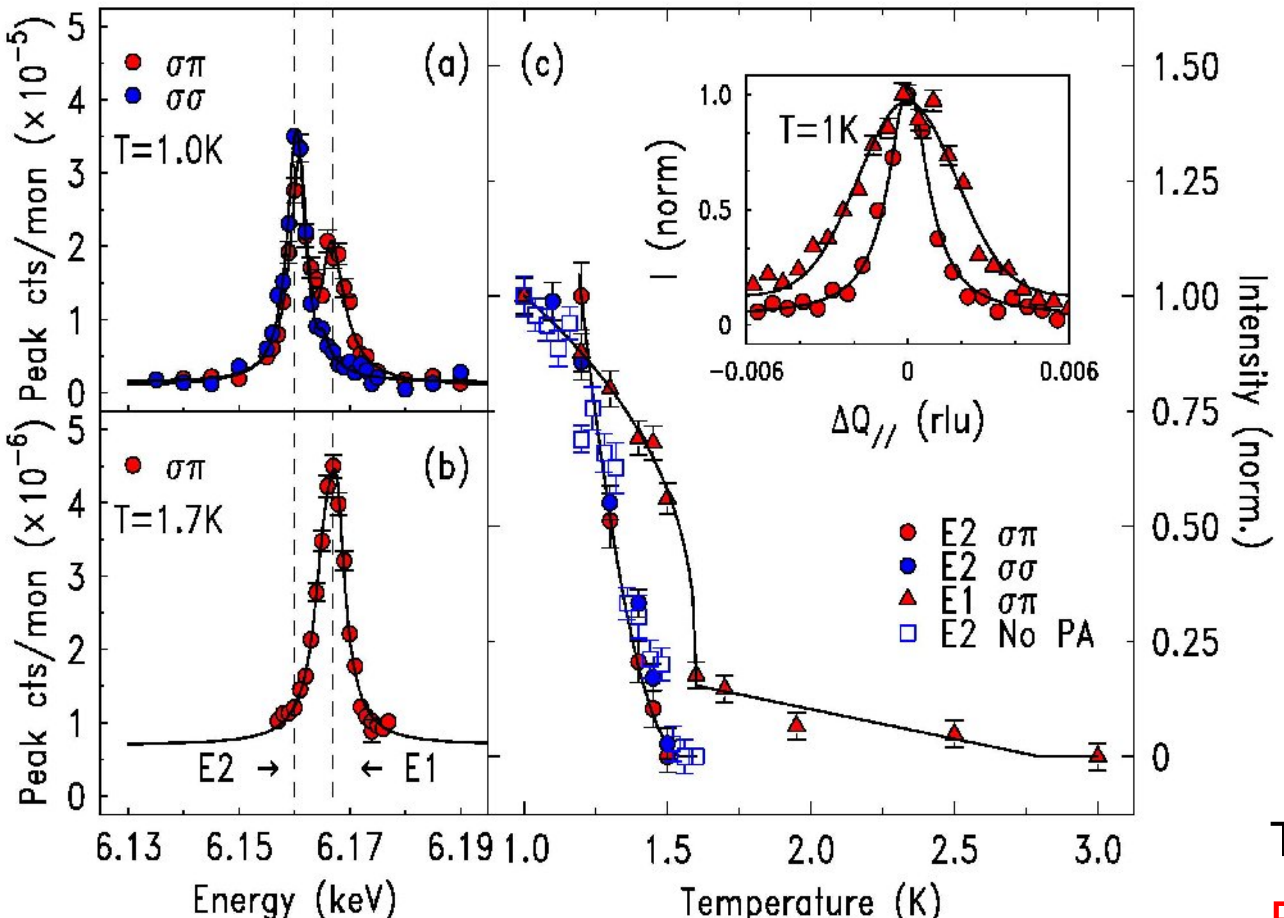
Kubo & Kuramoto J. Phys. Soc. Jpn. 73 216 (2004).

RXS study of Phase IV



E1 and E2 Thermal and Spatial Independence !

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



$\beta(5d) = 0.33$

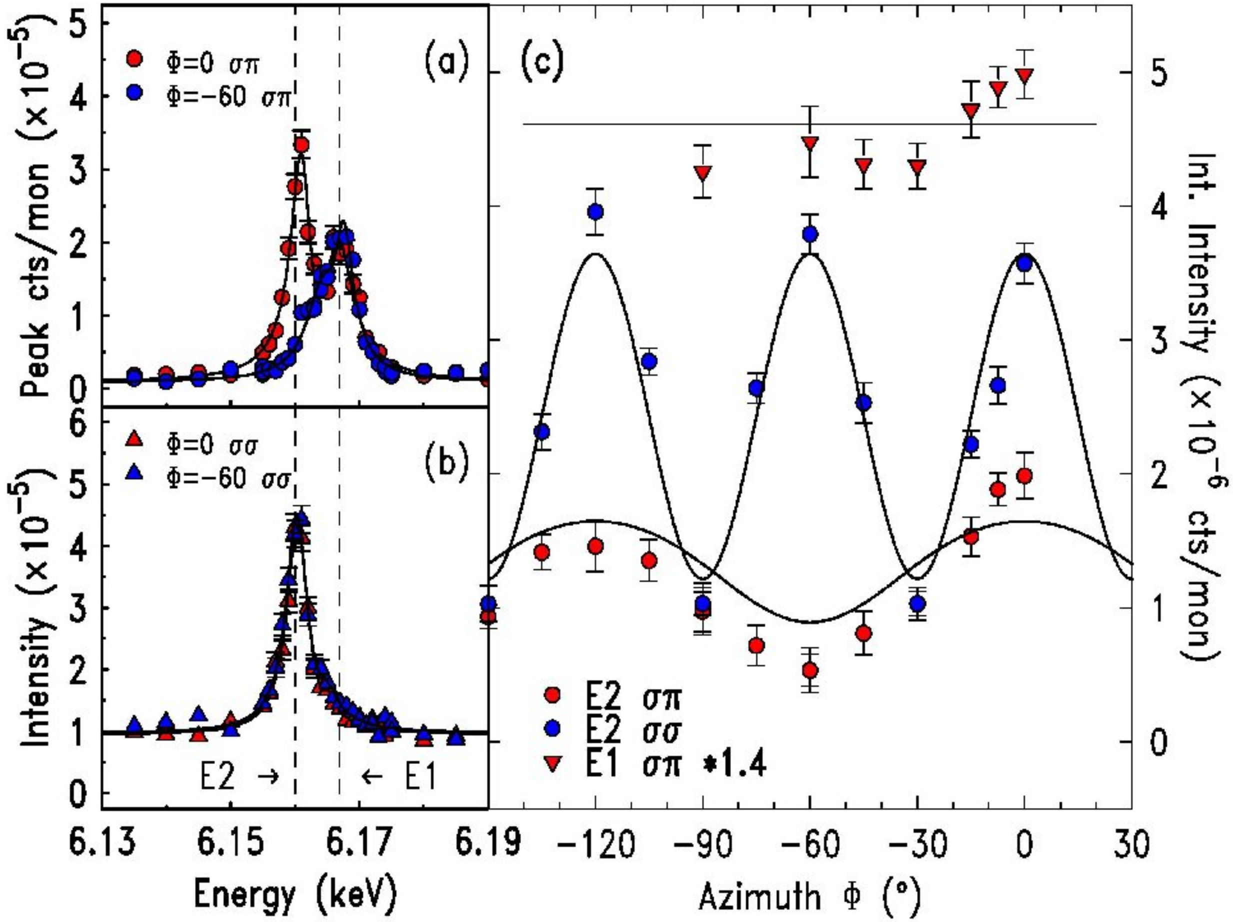
$\beta(4f) = 0.99$

T-dep consistent with:

E1 Dipole

E2 Octupole

Azimuth dependence at 1.0 Kelvin



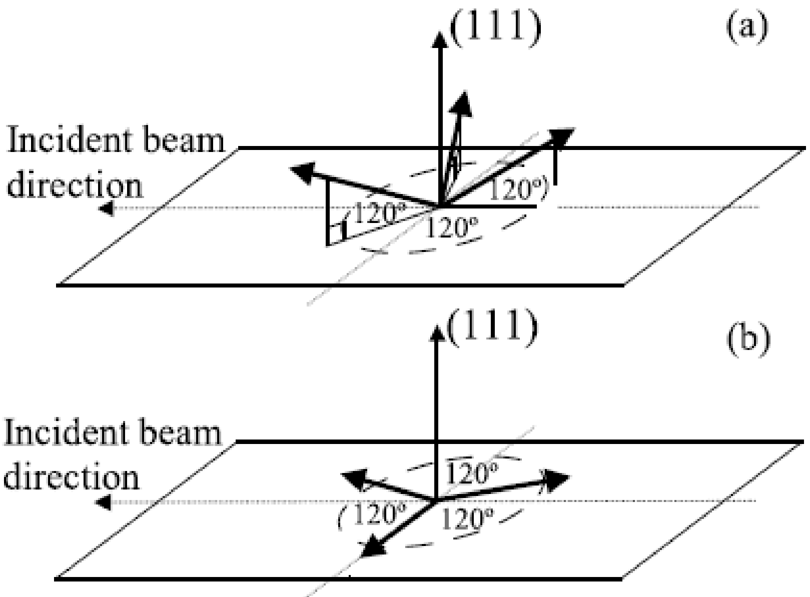
E1 σπ Only → Dipole

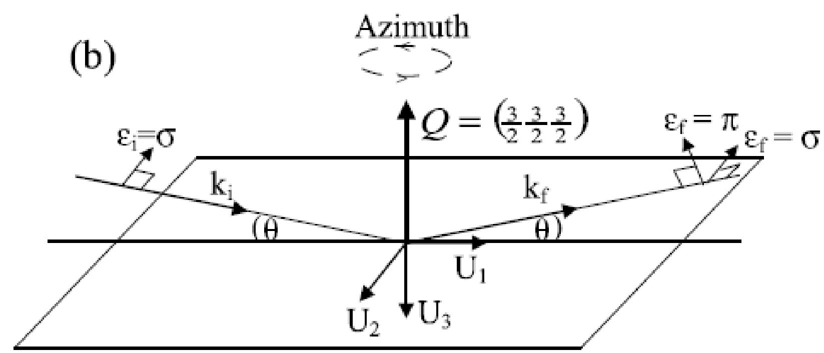
NO AFQ order → Phase IV is new phase!

E2 σσ 6-fold azimuth

E2 σπ 3-fold azimuth

Octupole Order of T_{1u} (Γ_{4u}) symmetry elements

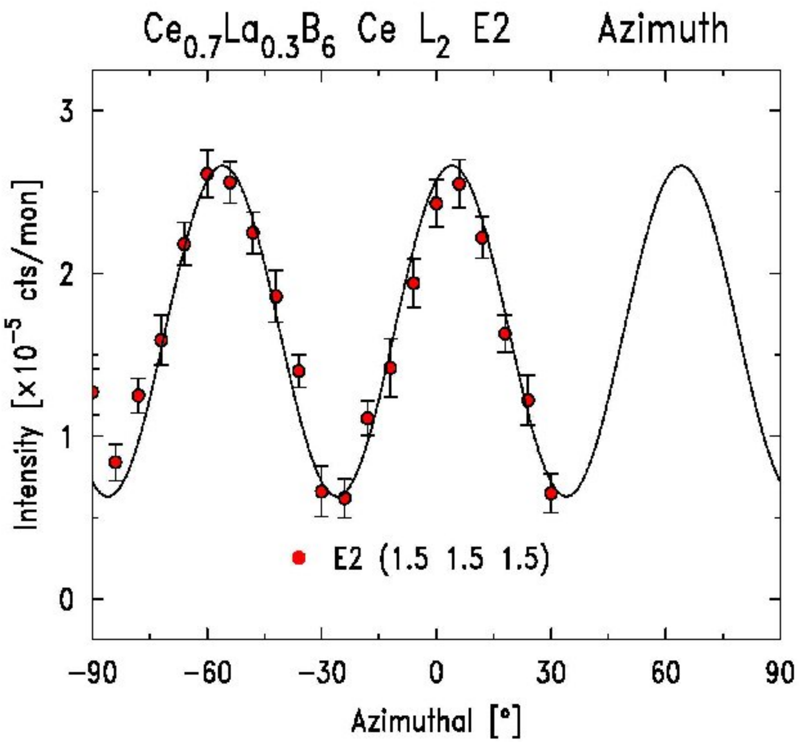


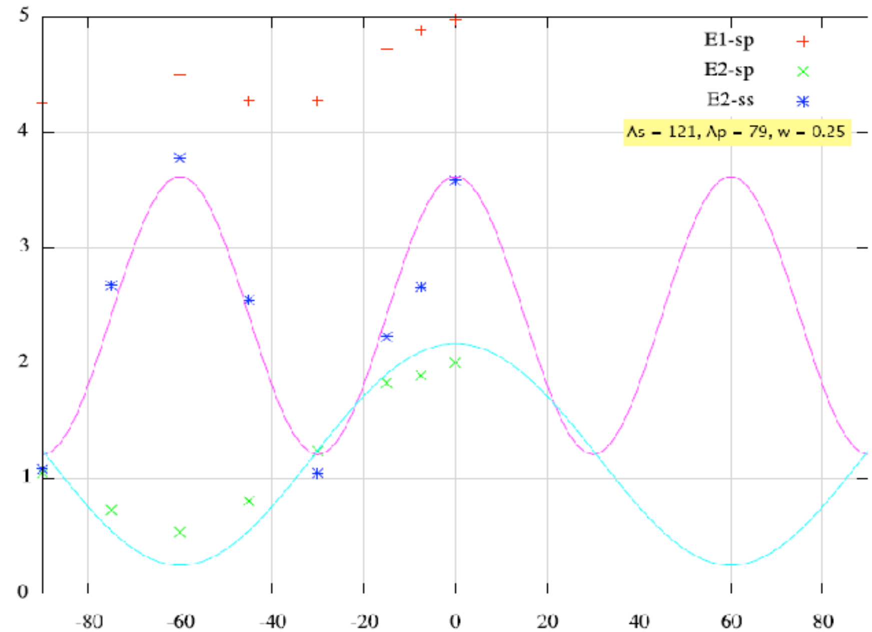


$$f_{nE2}(\Phi) = \sum_{i=1}^{3}[-i(F^3_{E2})][(k_f \cdot Z^i_n(\Phi))(k_i \cdot Z^i_n(\Phi))(\epsilon_f \times \epsilon_i) \cdot Z^i_n(\Phi) + (\epsilon_f \cdot Z^i_n(\Phi))(\epsilon_i \cdot Z^i_n(\Phi))(k_f \times k_i) \cdot Z^i_n(\Phi)$$
$$+(\epsilon_f \cdot Z^i_n(\Phi))(k_i \cdot Z^i_n(\Phi))(k_f \times \epsilon_i) \cdot Z^i_n(\Phi) + (k_f \cdot Z^i_n(\Phi))(\epsilon_i \cdot Z^i_n(\Phi))(\epsilon_f \times k_i) \cdot Z^i_n(\Phi)]$$

Following Hill & McMorrow Acta Cryst. A52 236 (1996).

RXS without Polarisation Analysis





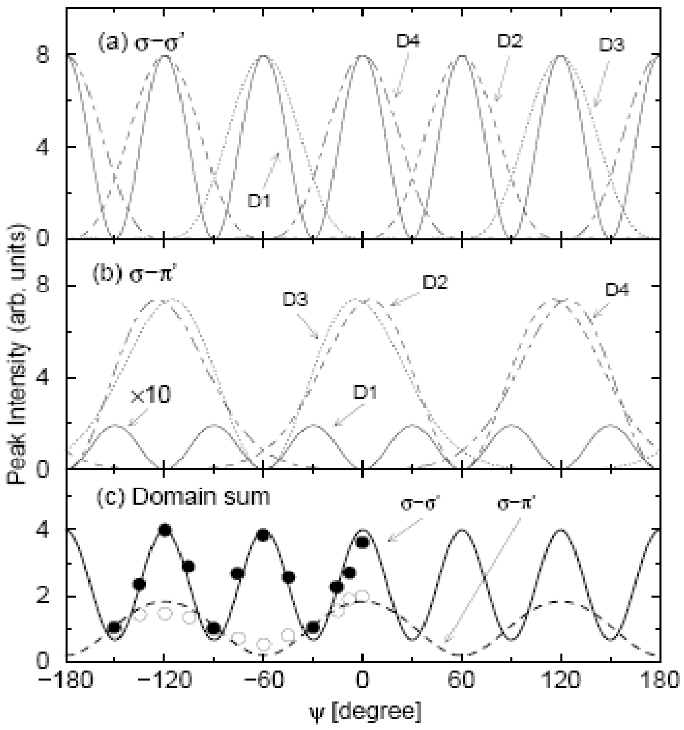
I(No PA) = I(σσ)+I(σπ)

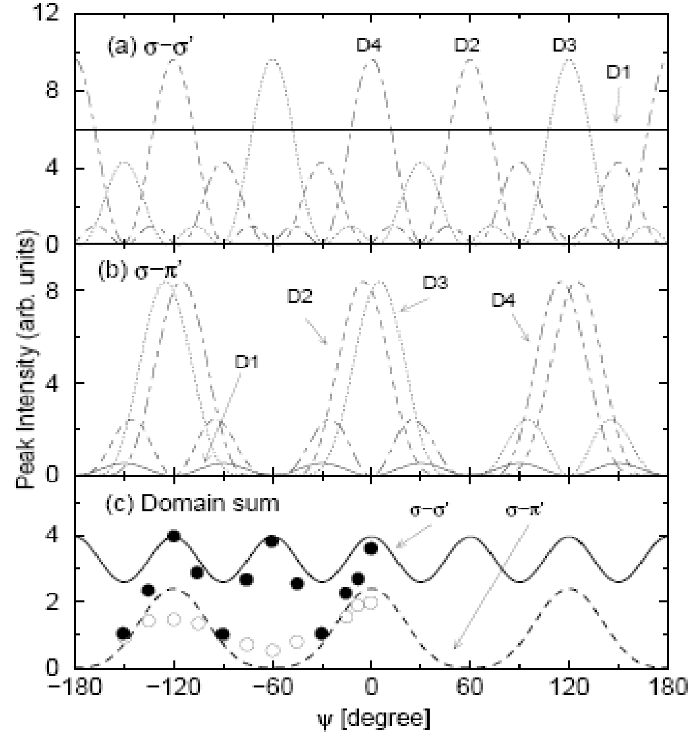
σσ Intensity larger than σπ

Γ_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=(½ ½ ½)

4. Simple model for E2 T-dep, azimuth & Bragg dependence:
   → 4f octupole order with T_{1u} symmetry elements
   → at q=(½ ½ ½) below T_{IV} only.

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

D. Mannix et al. Physical Review Letters 95 117206 (2005)

6. Theory: Kusunose and Kuramoto, Nagao and Igarashi, Lovesey and Katsumata.
   Furure experiments at (0.5 0.5 0.5) and Ce L_3 edge.