Multipole Physics in Orbitally Degenerate Systems

Hiroaki Kusunose, (Tohoku U., Sendai)

in collaboration with Y. Kuramoto, (Tohoku U.)

Outline

Material science ... search for extraordinary phase.
Possibility of unexpected phenomena due to exotic phase.

- Anisotropic superconductivity (previous session).
- Exotic order parameter of electrons ... multipole (this session).

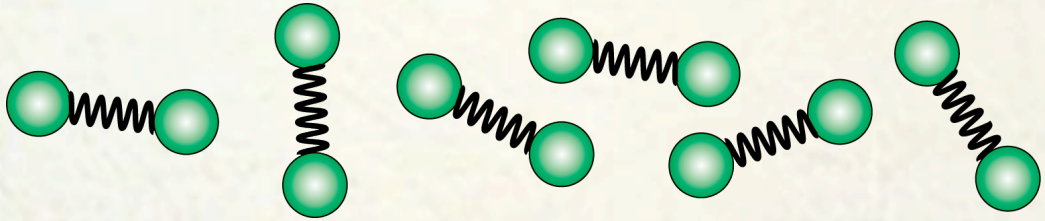
Previous talk by Dr. Mannix
first discovery of octupole order.

- What is multipole in solids ?
- Typical example: unusual behaviors in terms of octupole.
- Theoretical ground for first observation of octupole order.

Various orders in solids

related properties of electrons

Superconductivity



number

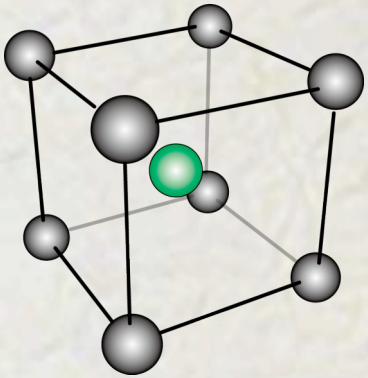
Charge density wave

charge
electric monopole

Magnetism

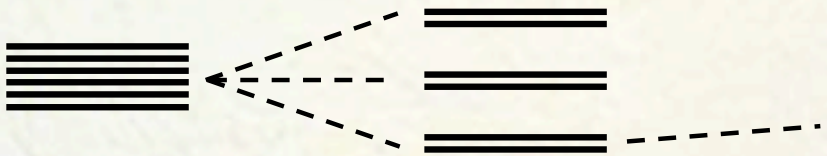
spin
magnetic dipole

Extra properties of electrons in solids



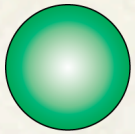
Crystalline electric field (CEF) lifts orbital degeneracy

spherical ⇒ CEF



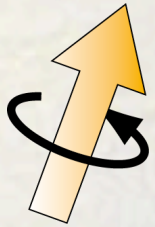
Kramers pair
(time reversal)

E-monopole
(charge)



scalar

M-dipole
(spin)

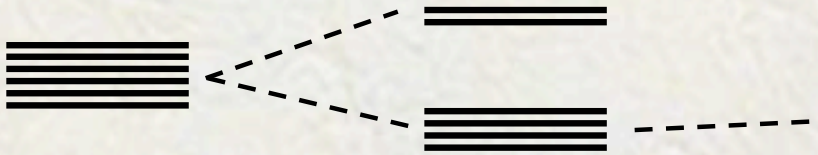


vector

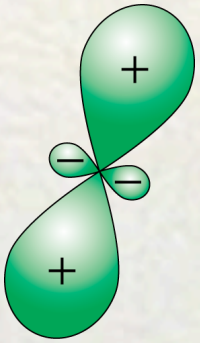
basic

high-symmetry CEF preserves orbital degeneracy

spherical ⇒ CEF

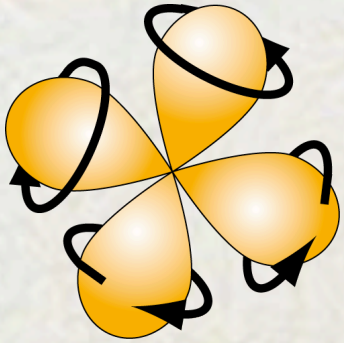


E-quadrupole



2nd-rank
tensor

M-octupole

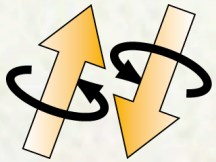


3rd-rank
tensor

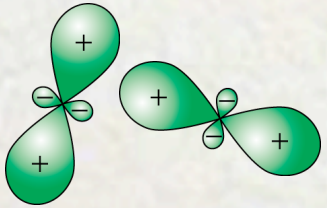
extra

Spontaneous order of multipoles

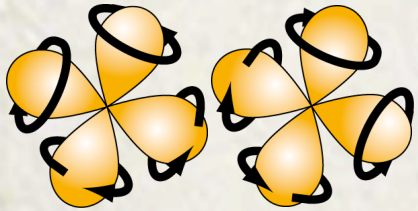
Normal (para) phase



equal population



equal population

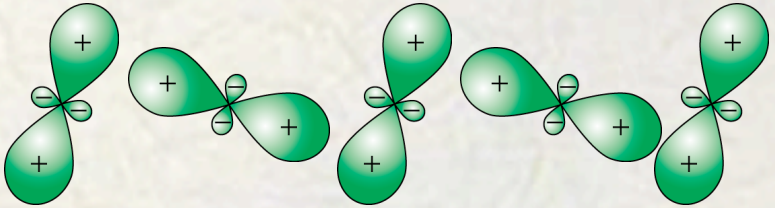


equal population

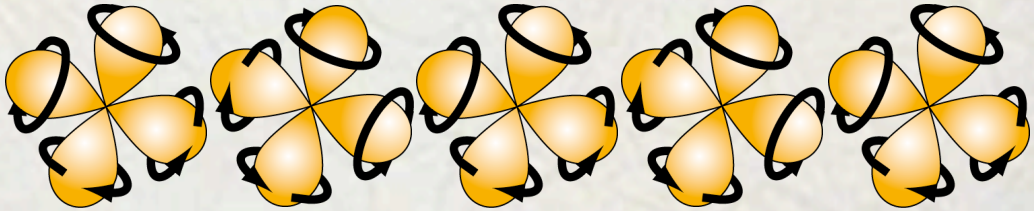
Ordered phase

magnetic phase

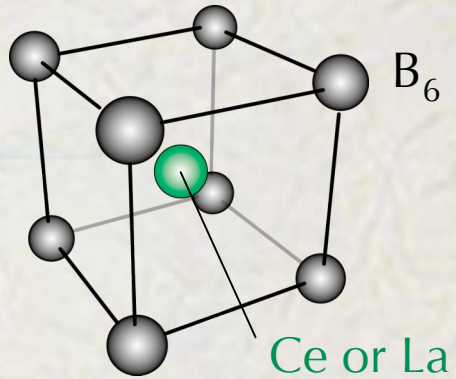
quadrupole phase



octupole phase



$Ce_xLa_{1-x}B_6$ ... cubic symmetry

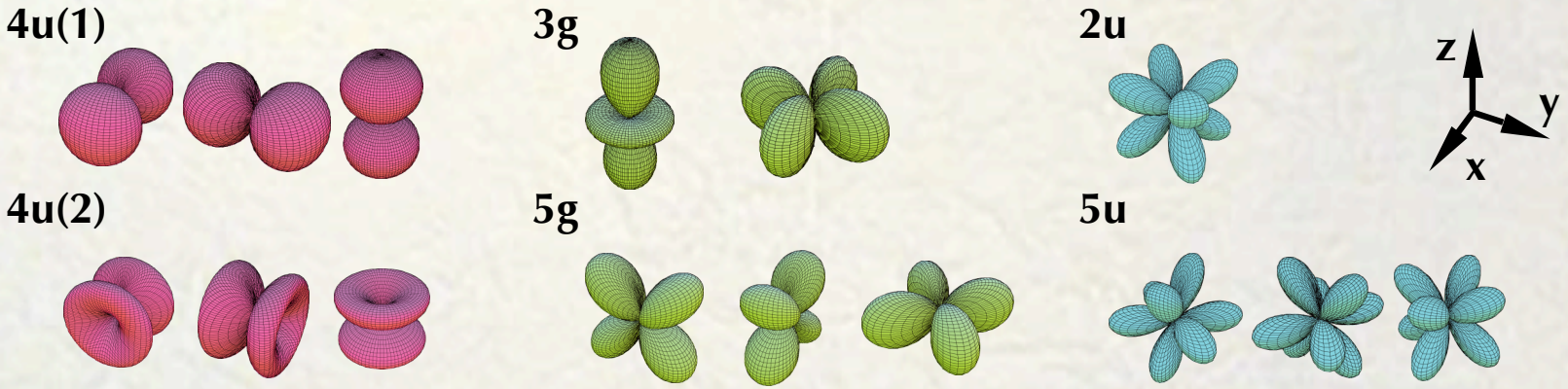


Ce ion has 4-fold CEF generacy

4x4 matrix = 1 + 15 independent multipoles

Irreducible rep. under cubic symmetry

symmetry



probe

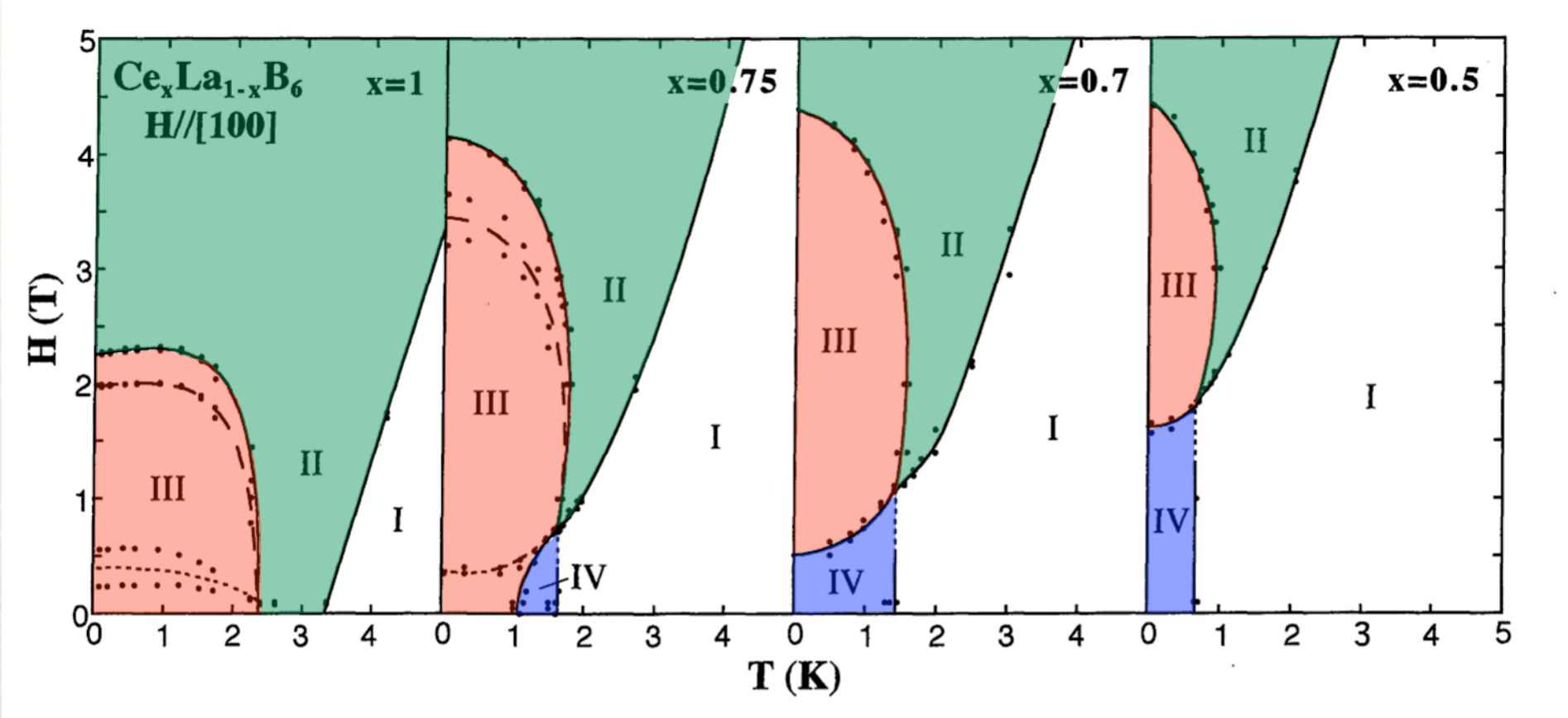
M-dipole (J)
- magnetic field
- neutron
- NMR
- μSR
- ...

E-quadrupole (O)
- uniaxial stress
- ultrasonic (elastic const.)
- polarized neutron
- resonant X-ray

M-octupole (T)
- local magnetic field (μSR, NMR)
- resonant X-ray

Multiple phases in H-T diagram

Tayama et al. JPSJ **66** 2268 '97



I. paramagnetic

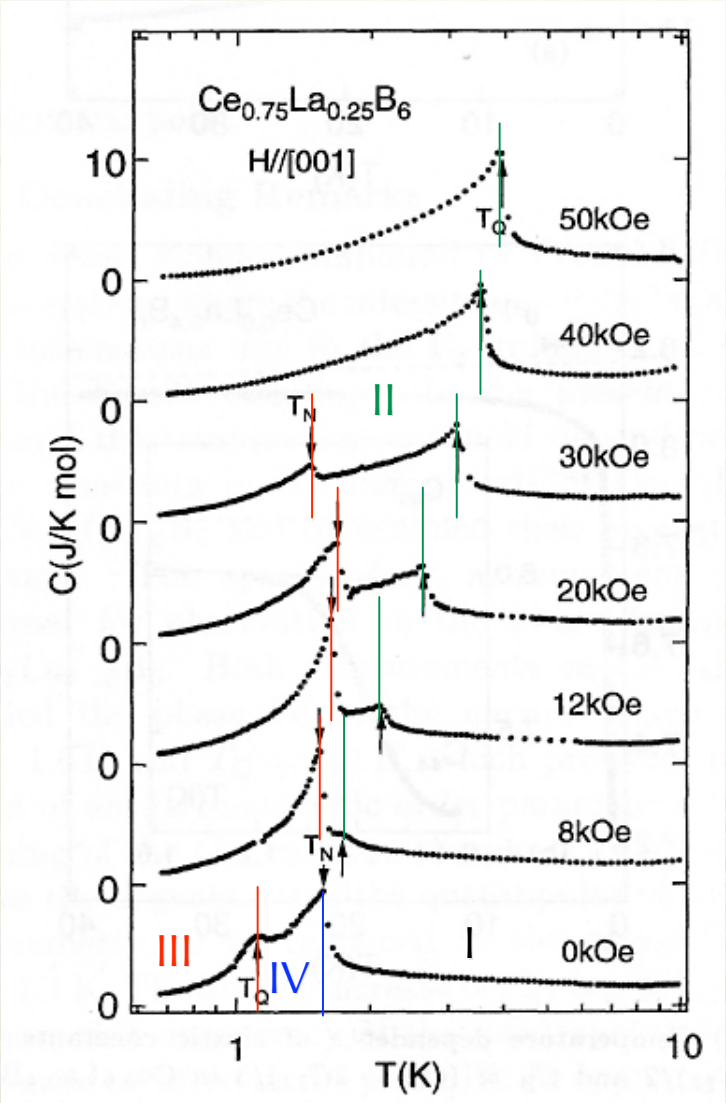
II. antiferro quadrupole (AFQ) RXS by Nakao et al. JPSJ **70** 1857 '01

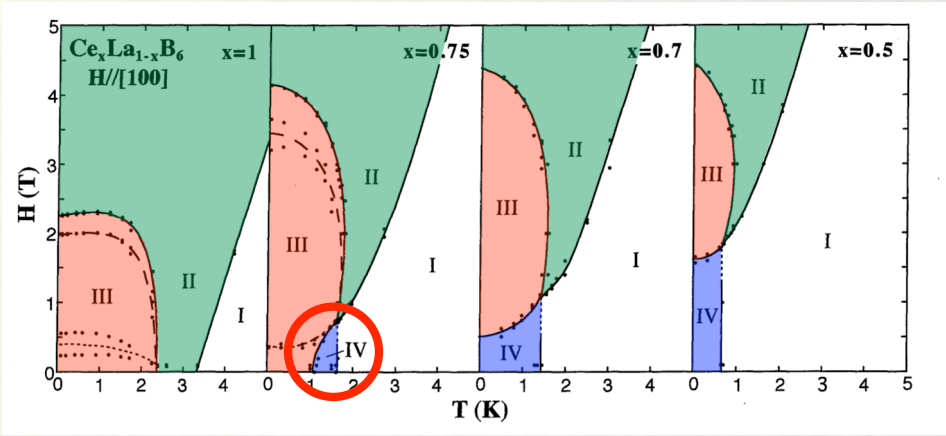
III. AFQ + dipole Neutron by Effantin et al. JMMM **47&48** 145 '85

IV. octupole order ?

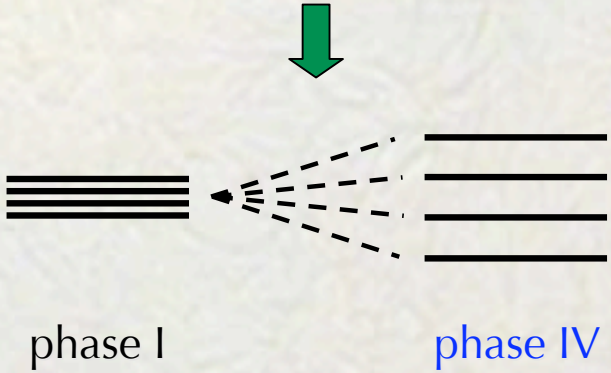
Hint for phase IV ... second order phase transition

Suzuki et al. JPSJ **67** 4243 '98





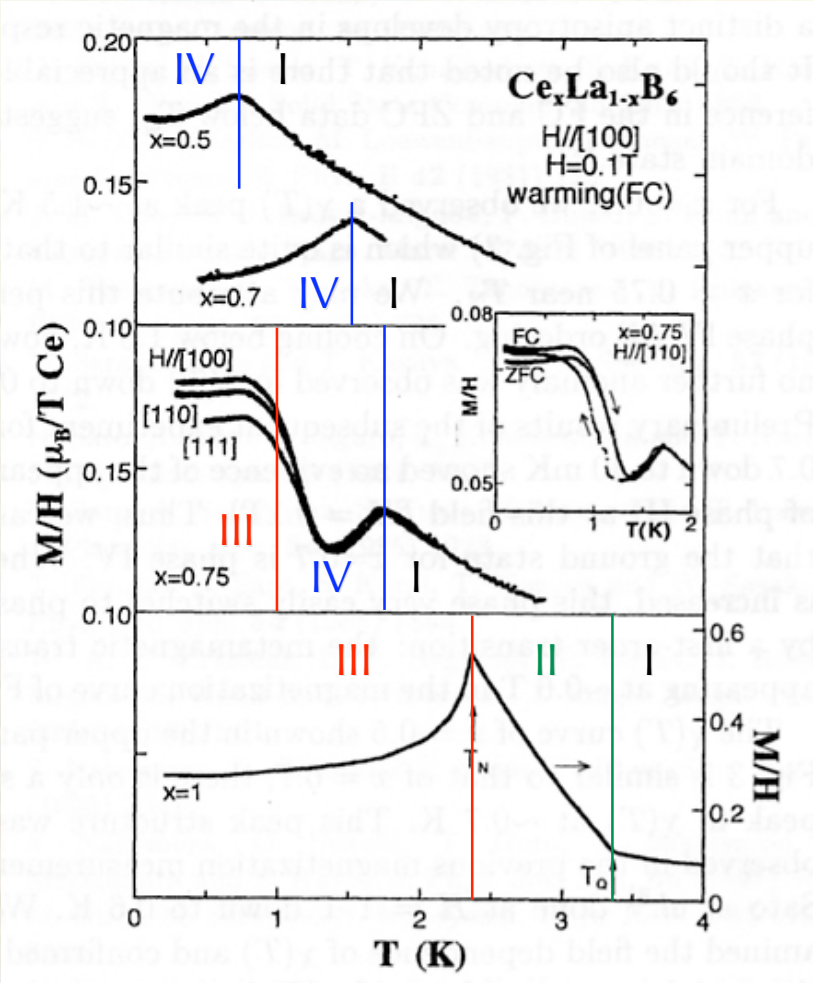
specific-heat jump at T_{IV}
entropy release ~ almost log(4)



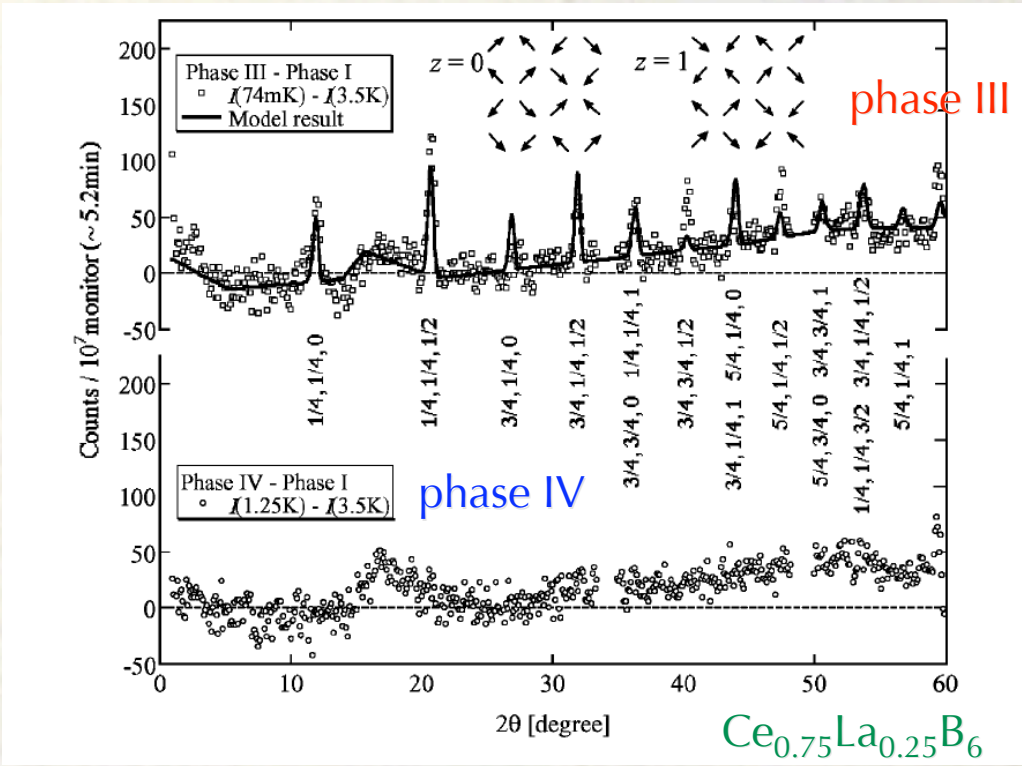
phase I phase IV

Hint for phase IV ... broken time-reversal symmetry

Tayama et al. JPSJ **66** 2268 '97



Iwasa et al. Physica B **329&333** 582 '03

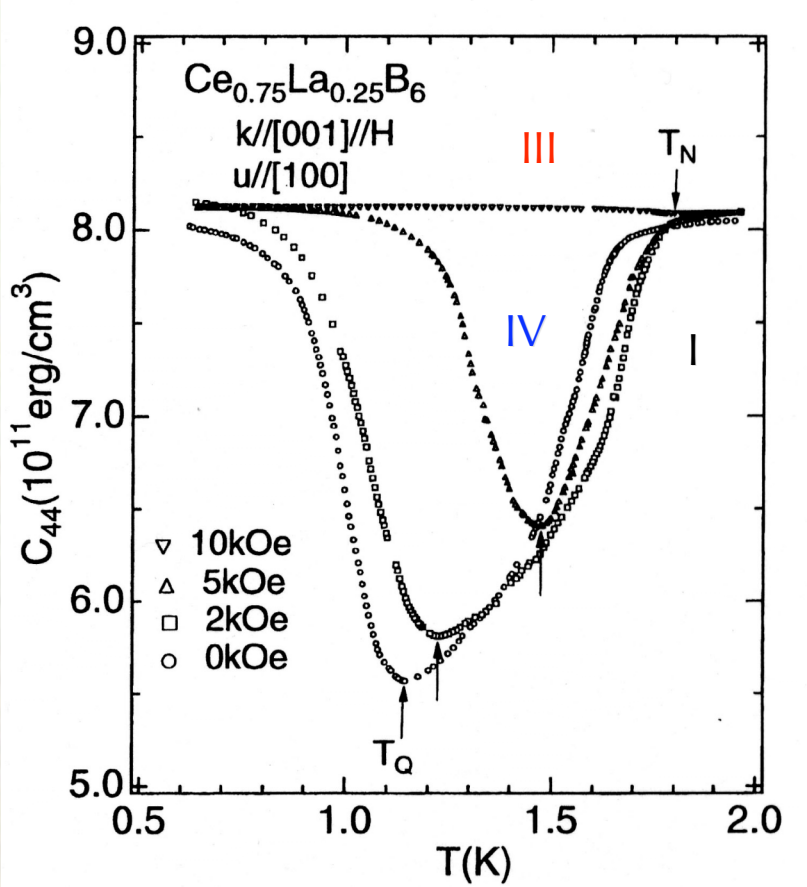


magnetic susceptibility shows cusp at T_{IV}
but, no Bragg peak in phase IV

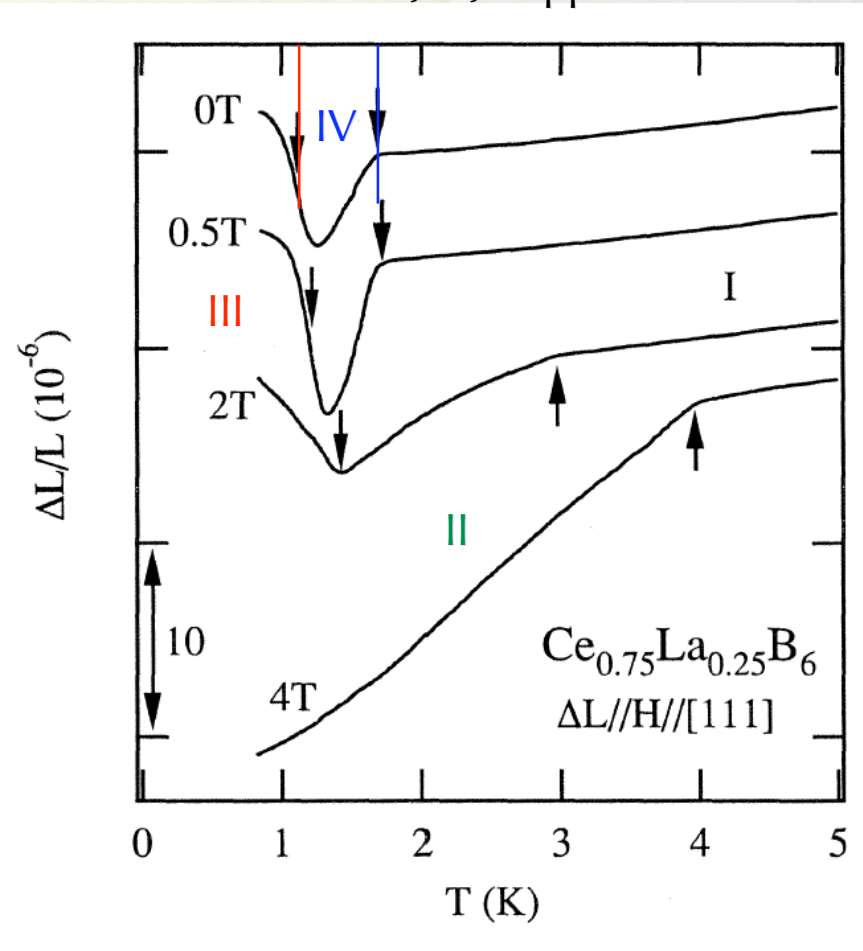
cf.
μSR, NMR also suggest broken time-reversal symmetry
Takagiwa et al. JPSJ **71** 31 '02,
Magishi et al. Z. Naturforsch **57**a 441 '02

Hint for phase IV ... huge softening & lattice distortion

Suzuki et al. JPSJ **67** 4243 '98



Akatsu et al. JPSJ Suppl. **71** 115 '02



elastic const. shows huge softening blow T_{IV}
lattice distortion along [111]

➡ coupling between E-quadrupole & hidden order

Octupole model for phase IV

HK and Kuramoto, JPSJ **70** 1751 '01
Kubo and Kuramoto, JPSJ **73** 216 '04

Order parameters

primary : AF octupole (5u type)

$$T(\boldsymbol{Q}) = (T^x_{5u} + T^y_{5u} + T^z_{5u})/\sqrt{3}$$

secondary : Ferro quadrupole (5g type)

$$O(\boldsymbol{0}) = (O^x_{5g} + O^y_{5g} + O^z_{5g})/\sqrt{3}$$

Free energy

3rd-order term possible (time-reversal even)

$$F = \frac{\alpha}{2}(T - T_{\rm IV})T^2(\boldsymbol{Q}) + \frac{b}{4}T^4(\boldsymbol{Q}) + \frac{a}{2}O^2(\boldsymbol{0}) \boxed{-gT^2(\boldsymbol{Q})O(\boldsymbol{0})} + \ldots$$

order parameters

$$T^2(\boldsymbol{Q}) = A(T_{\rm IV} - T)$$

$$O(\boldsymbol{0}) = \frac{g}{a}A(T_{\rm IV} - T)$$

$$A = a\alpha/(ab - 2g^2)$$

susceptibilities

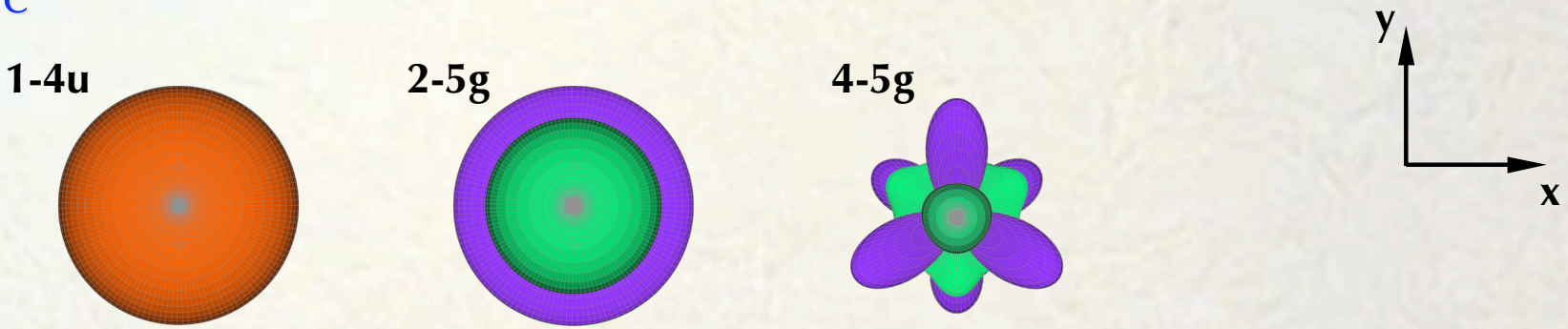
$$\chi_T(\boldsymbol{Q}) = \begin{cases} 1/2\alpha(T_{\rm IV} - T) & (T < T_{\rm IV}) \\ 1/\alpha(T - T_{\rm IV}) & (T > T_{\rm IV}) \end{cases}$$

$$\chi_O(\boldsymbol{0}) = \begin{cases} bA/a\alpha & (T < T_{\rm IV}) \\ 1/a & (T > T_{\rm IV}) \end{cases}$$

- Broken time-reversal symmetry
- Lattice distortion along [111]
- Huge softening (finite jump) in elastic const. [$\chi_O(\boldsymbol{0})$]
- Cusp structure in uniform magnetic susceptibility (due to singlet ground state)

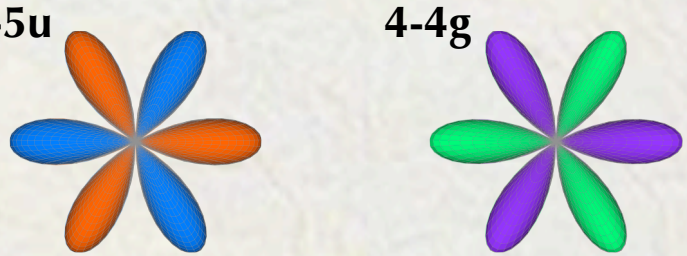
Shape (amplitude) of multipoles (view from z=[111])

isotropic

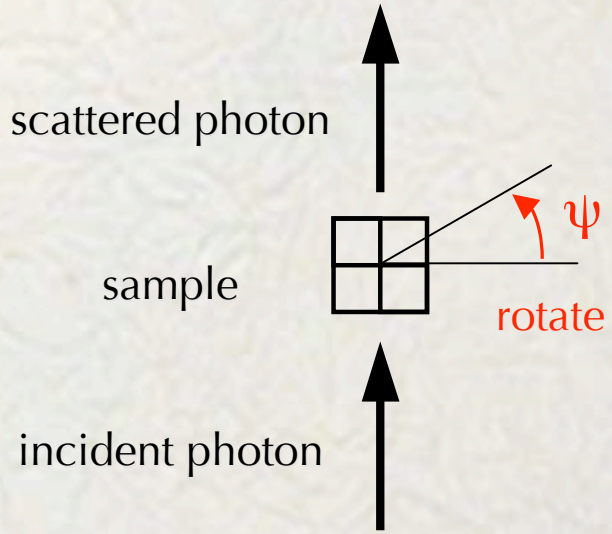


6-fold (max. in x direction)

6-fold (min. in x direction)



Resonant X-ray Scattering (RXS)



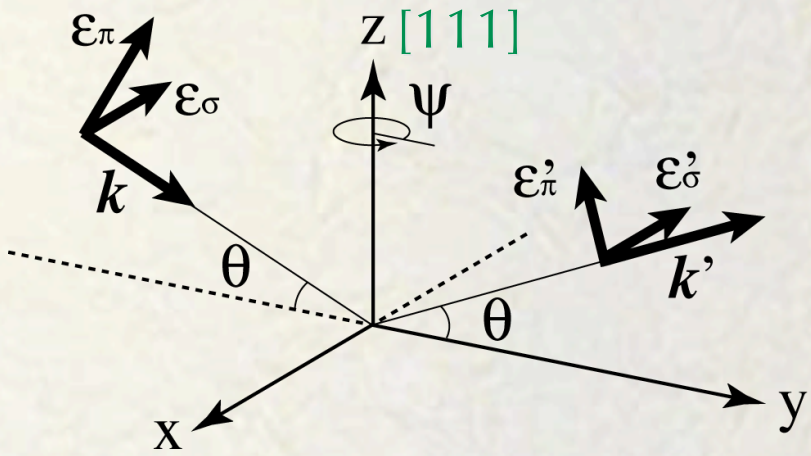
Azimuthal angle dependence for possible multipoles

E2σσ'

E-quadropole, non-rotated channel

E2σπ'

E-quadropole, rotated channel

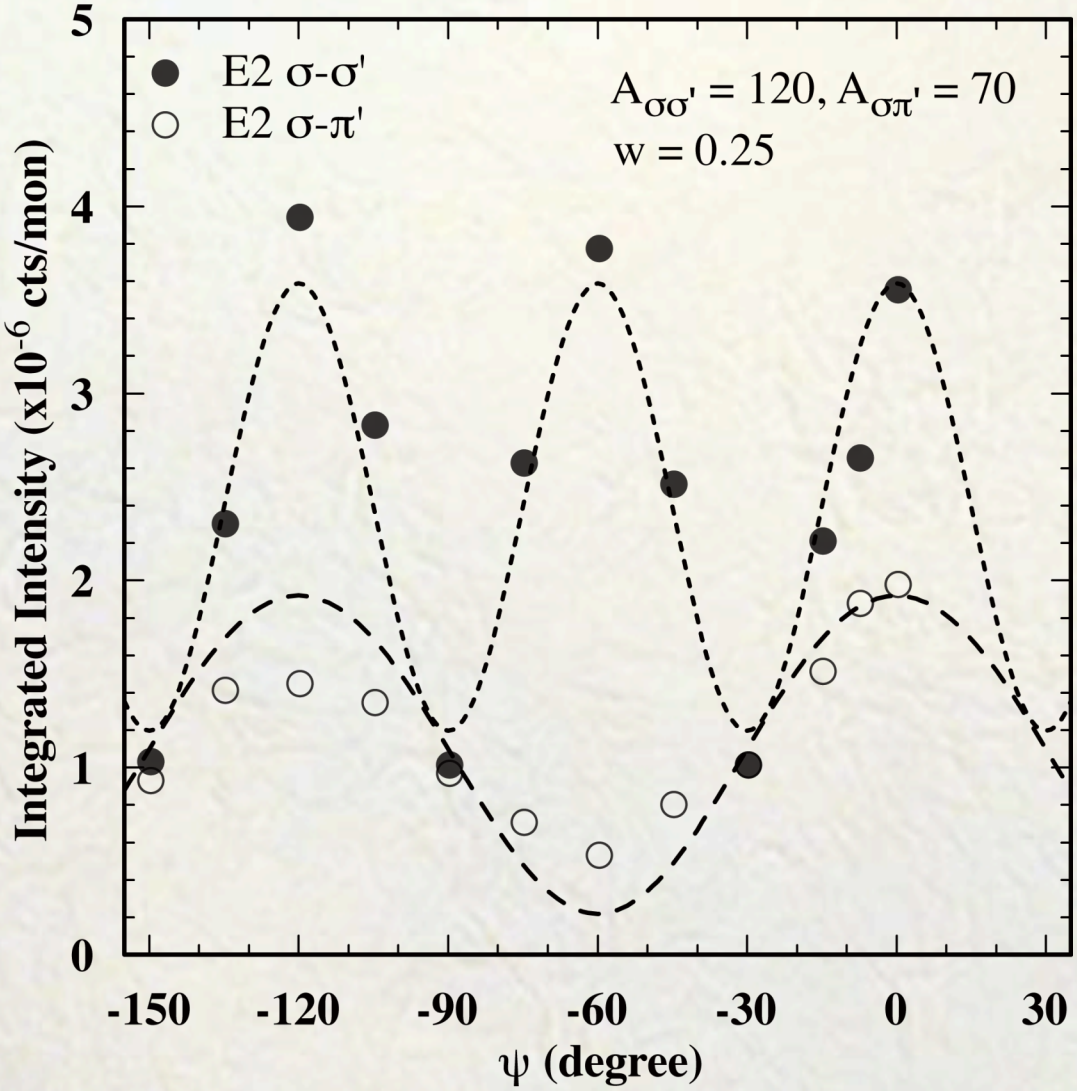


Scattering amplitude

p-Γ	E2 $\sigma\sigma'$	E2 $\sigma\pi'$
1-4u	0	$\frac{1}{40}\sin^2(3\theta)$
2-5g	$\frac{1}{224}(3+\cos(2\theta))^2$	0
3-2u	$\frac{1}{36}\sin^2(2\theta)\sin^2(3\psi)$	$\frac{1}{144}(3\cos(2\theta)-1)^2\cos^2\theta\left[\frac{1}{\sqrt{2}}\tan\theta-\cos(3\psi)\right]^2$
3-4u	$\frac{1}{36}\sin^2(2\theta)\sin^2(3\psi)$	$\frac{1}{144}(3\cos(2\theta)-1)^2\cos^2\theta\left[\frac{1}{\sqrt{2}}\tan\theta+\cos(3\psi)\right]^2$
3-5u	$\frac{1}{16}\sin^2(2\theta)\cos^2(3\psi)$	$\frac{1}{256}(\cos\theta+3\cos(3\theta))^2\sin^2(3\psi)$
4-4g	0	$\frac{1}{16}\cos^6\theta\cos^2(3\psi)$
4-5g	$\frac{1}{1512}(5-3\cos(2\theta))^2$	$\frac{1}{1296}\cos^6\theta\sin^2(3\psi)$

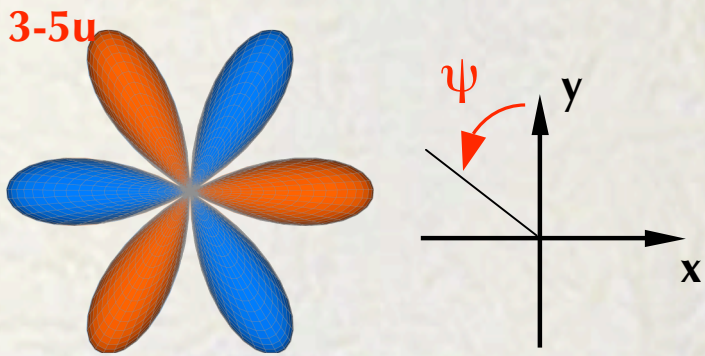
First observation of magnetic octupole order

Fit of experimental data by 5u octupole model



Symbols:
Mannix et al., PRL **95** 117206 '05

Theoretical lines:
HK and Kuramoto, JPSJ **74** 3139 '05



Note:
Assume same volume fraction
of 4 equivalent domains

Summary

Multipoles

- can be spontaneous order parameter.
- play important roles behind anomalous phenomena.
- are detectable directly by RXS.
- Phase IV in $Ce_{0.7}La_{0.3}B_6$ is the first example of spontaneous octupole order.