強相関Co酸化物の共鳴軟X線散乱

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Collaborators

- RSXS and XAS measurements

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- Samples

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- Cluster model calculations

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- Beamline and Machine

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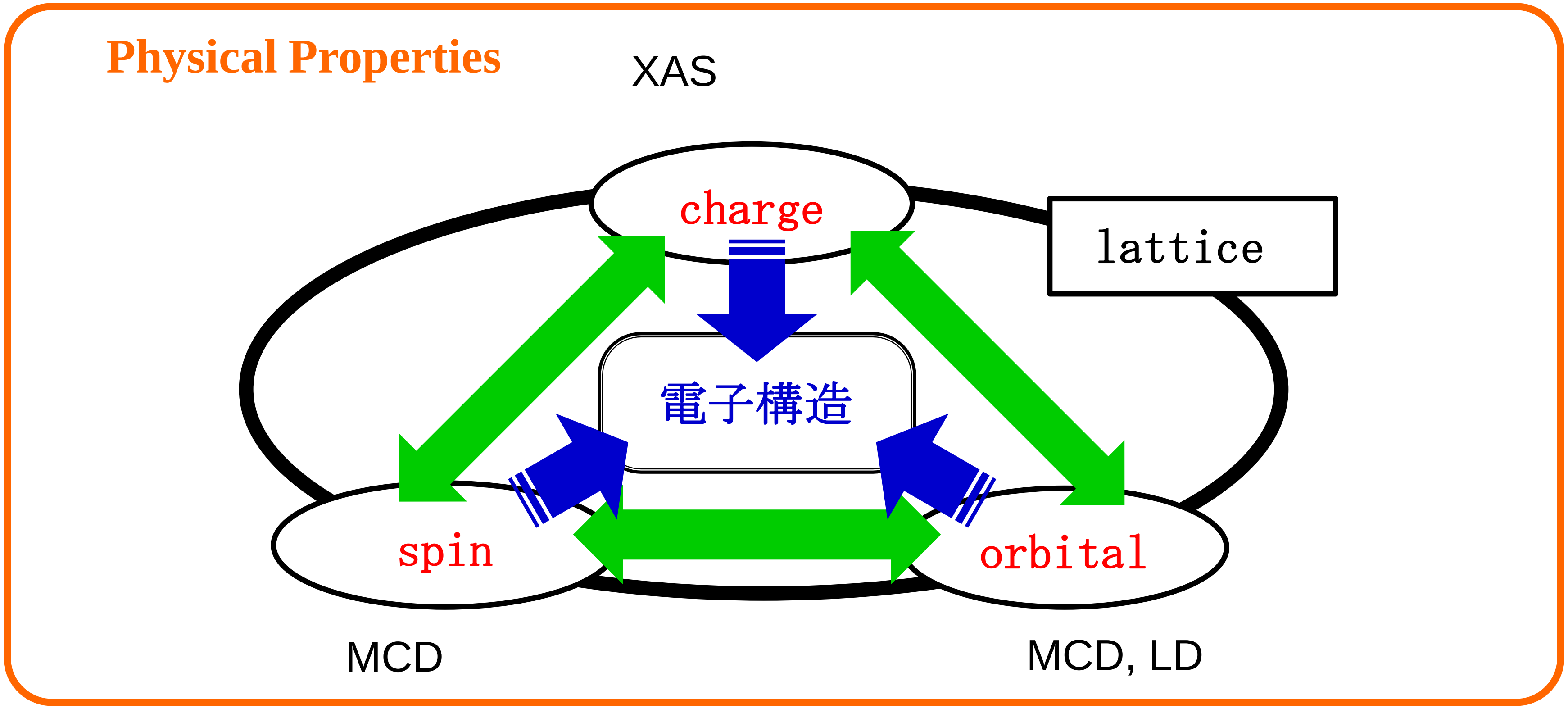
M. Kubota (JAEA)

Strongly correlated electron systems ~ spectroscopy & scattering ~



全体の電子構造
周期的に揃った電子構造

Resonant soft X-ray Scattering

Enhancement of Structure Factor

$f^{res} \propto$ ***Element selective***

$$\underbrace{(\varepsilon_s^*\cdot\varepsilon_i)[F_{1,1}+F_{1,-1}]}_{\text{電荷散乱}} - \underbrace{i(\varepsilon_s^*\times\varepsilon_i)\cdot\hat{\mathbf{z}}[F_{1,1}-F_{1,-1}]}_{\text{磁化散乱}} + \underbrace{(\varepsilon_s^*\cdot\hat{\mathbf{z}})(\varepsilon_i\cdot\hat{\mathbf{z}})[2F_{1,0}-F_{1,1}-F_{1,-1}]}_{\text{軌道秩序}}$$

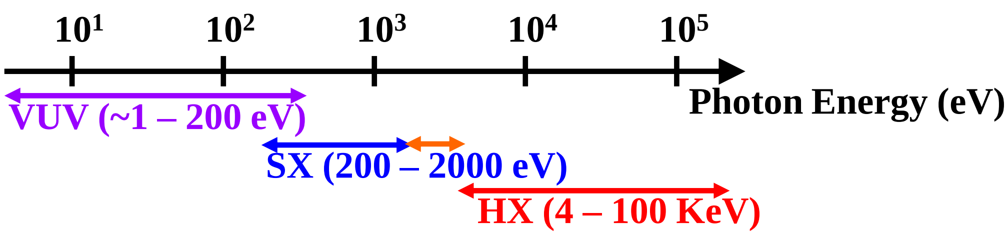
$\varepsilon_i, \varepsilon_s^*$: 入射光、散乱光の電場ベクトル

$F_{1,q}$: $\Delta m_l = q$の散乱振幅

$\hat{\mathbf{z}}$: 局在モーメント

J. P. Hannon *et al*., Phys. Rev. Lett. **61**, 1245 (1988).

偏光依存性を利用して、さまざまな電子自由度秩序構造を検出



*3d, 4d, 5d*遷移金属元素 *p* ➔ *d*遷移
希土類系 *d* ➔ *f* 遷移
酸素、硫黄 *s* ➔ *p* 遷移

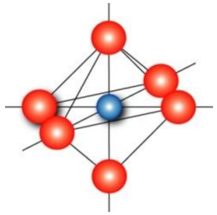
} **軟X線領域(200-2000 or 4000 eV)**

Fermi準位上電子状態の直接観測

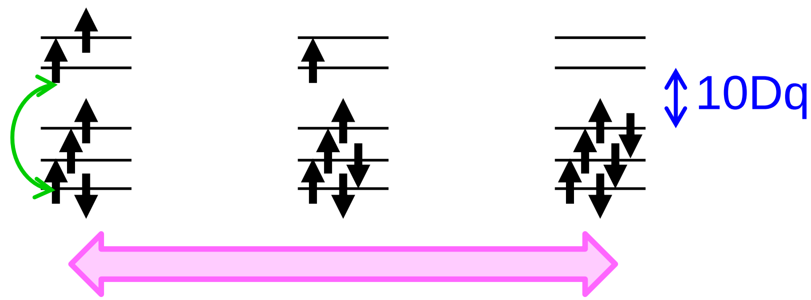
Co Oxides

Crystal Field

Co^{3+} (d^6) spin states



Hund, exchange



Spin moment

Orbital moment ?

Hybridization **between O *2p* and Co *3d* orbitals**

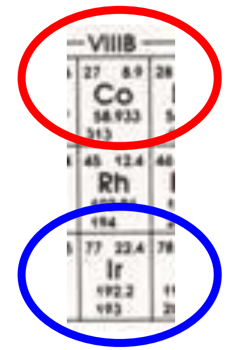
Sr_2IrO_4

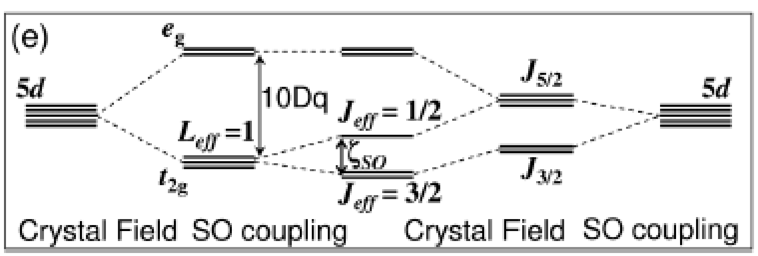
Mott insulator by J_{eff}

spin-orbit interaction ⇔ crystal field

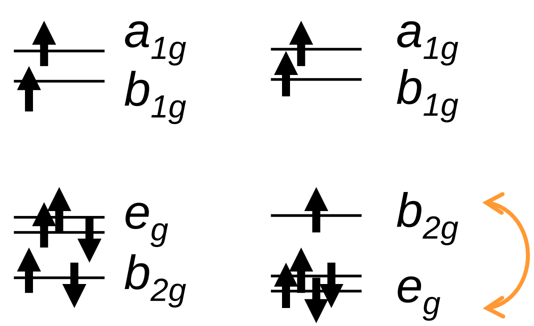
ζ_{SO}(Ir) ~ 0.4 eV

ζ_{SO}(Co) ~ 0.07 eV





(Tetragonal) Distortion



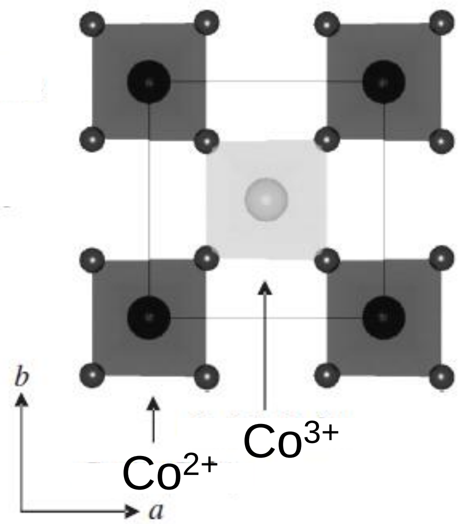
t_{2g} hole

SO interaction

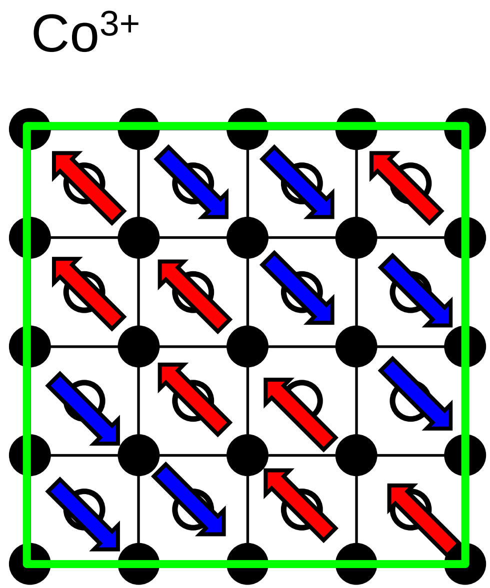
B. J. Kim *et al*., PRL **101**, 076402 (2009).

Co^{2+}とCo^{3+}を分けて電子構造を調べる

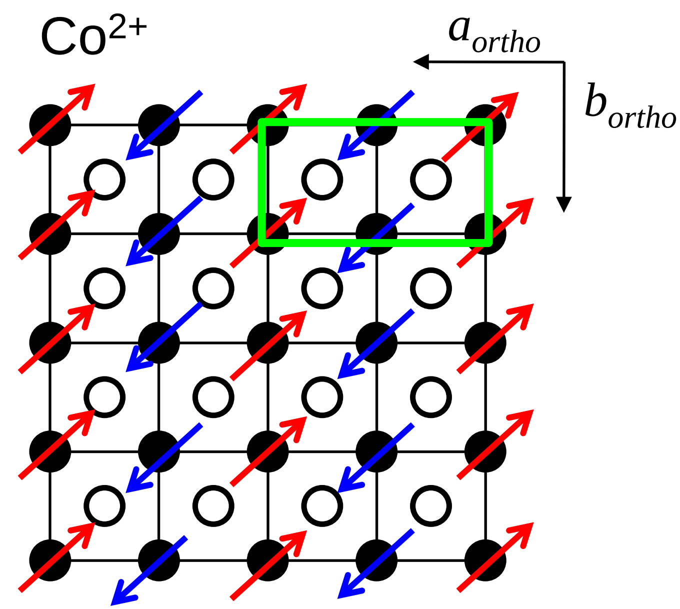
Charge ordering



Spin model



中性子散乱



K. Horigane *et al.*, J. Phys. Soc. Jpn. **76**, 114715 (2007).

磁気構造

***q* = (1/2, 0, 1/2)**

***q* = (1/2, 0, 1)**

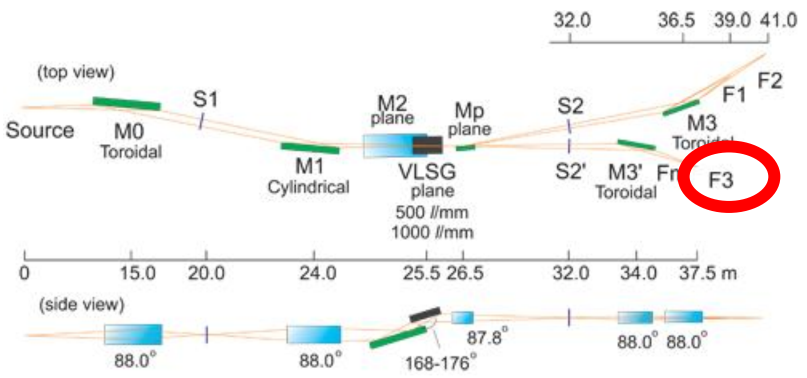
Co $L_{2,3}$-edge RSXS measurement

Co^{2+}-site electronic structures

Experimental

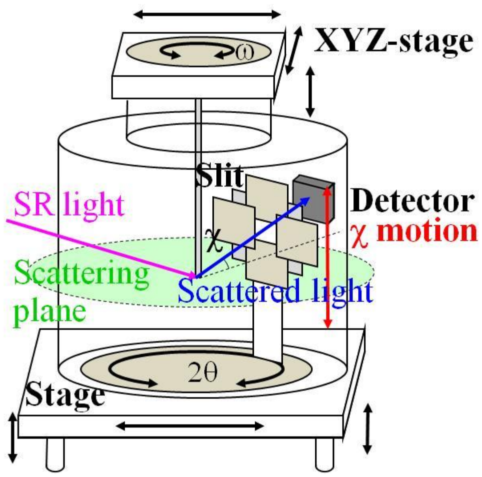
SX undulator beamline: BL16A

- Variable polarization: circular & linear
  APPLE II-type undulator
- $h\nu$: 250 – 1500 eV
- $E/\Delta E$ = 5000-10000 (500 – 1500 eV)
- Beam spot size: 50-100 μm(Vertical)
  100-300 μm (Horizontal)

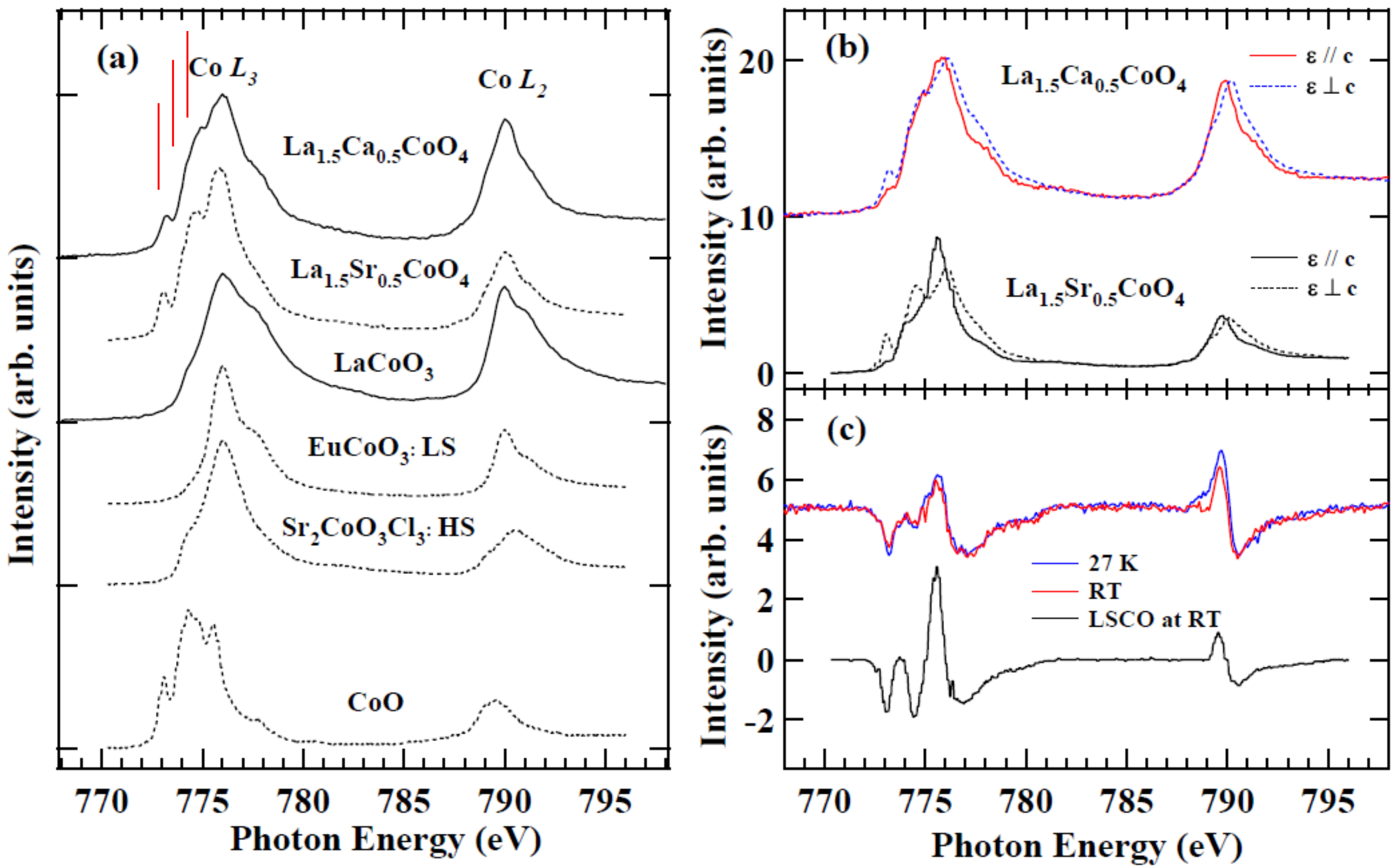


Resonant soft X-ray scattering measuring system

UHV, two-circle diffractrometer
- Base pressure : 1 × 10^{-5} Pa
- Lowest Temperature : 25 K
- Detector: Photodiode and MCP
- 2θ limit : 160°



Co $L_{2,3}$ - edge XAS

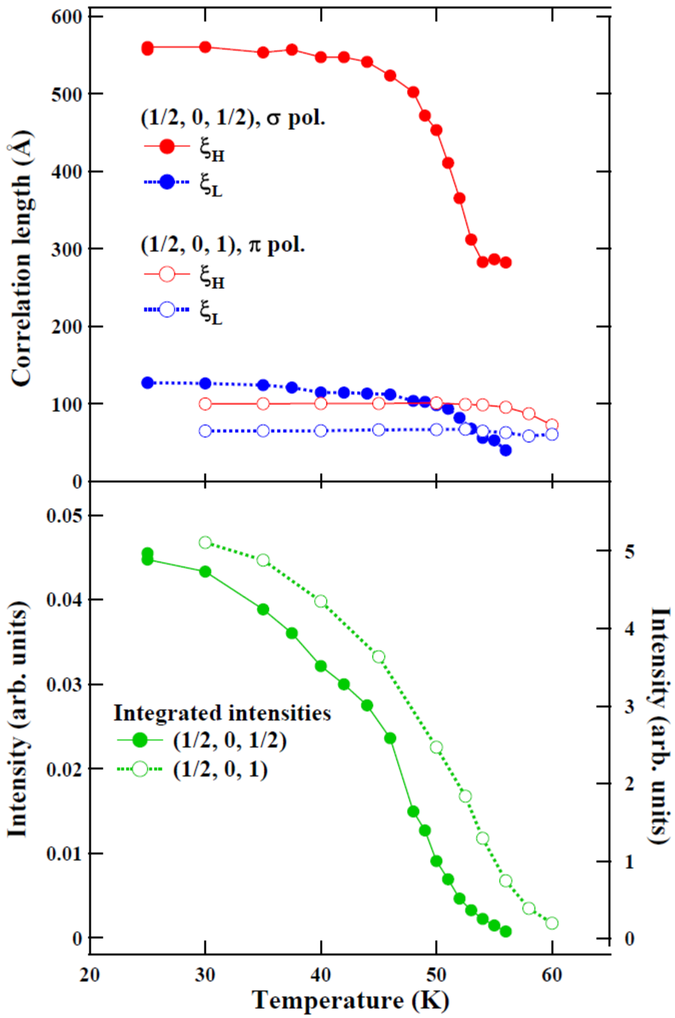


C. F. Chang *et al*., PRL **102**, 116401 (2009).

Co^{2+} and Co^{3+} exist.

LD XAS-like $La_{1.5}Sr_{0.5}CoO_4$ ⇔ Co^{2+} HS & Co^{3+} LS

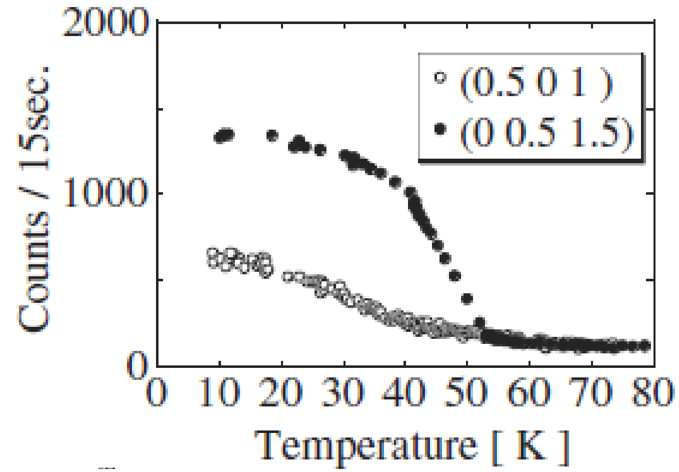
Magnetic ordering structures



	ξ_H	ξ_L	T_t
(1/2, 0, 1/2)	~ 550 Å	~ 120 Å	~ 56 K
(1/2, 0, 1)	~ 100 Å	~ 60 Å	~ 60 K

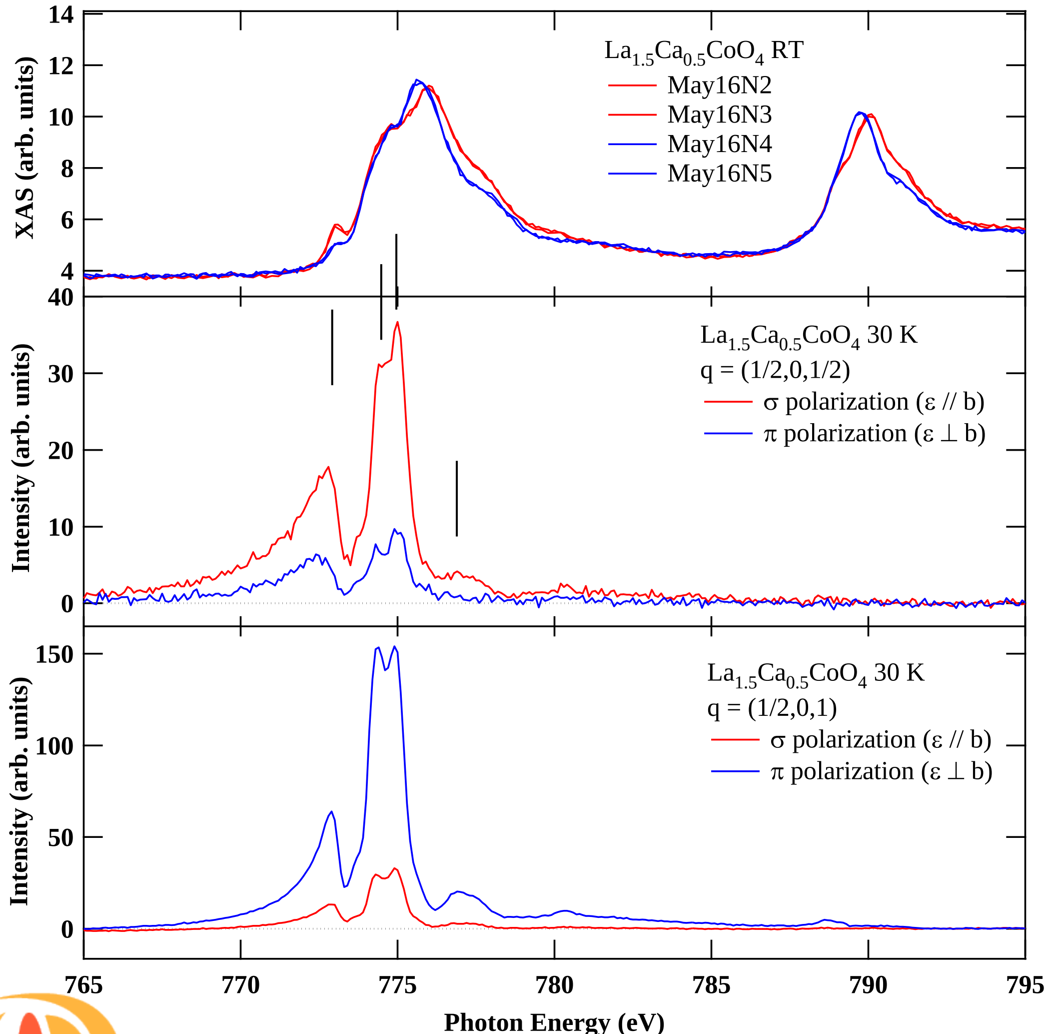
Neutron scattering

L = half-integer		L = integer	
ξ^{ab}_{so} (Å)	ξ^{c}_{so} (Å)	ξ^{ab}_{so} (Å)	ξ^{c}_{so} (Å)
195(4)	22(1)	98(8)	22(1)



K. Horigane *et al*., J. Phys. Soc. Jpn. **76**, 114715 (2007).

XAS & fix q scans

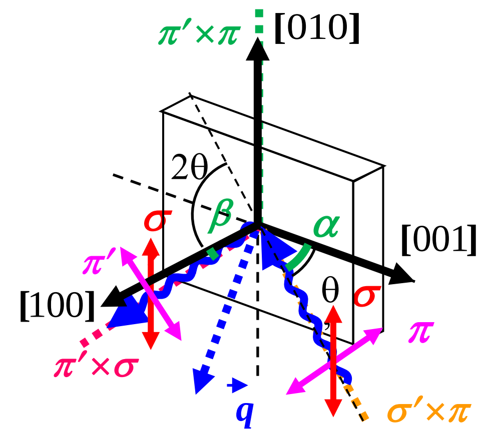


Sample:

$La_{1.5}Ca_{0.5}CoO_4$ single crystal

Orthorhombic

$a = b = 5.418$ Å, $c = 12.469$ Å



Co L₃-edge

Peaks : **773**, 775, 776, 778 eV

⬄ Mainly Co^{2+}

Co L₂-edge

No significant

Rough relation between XMCD & RSXS

XMCD

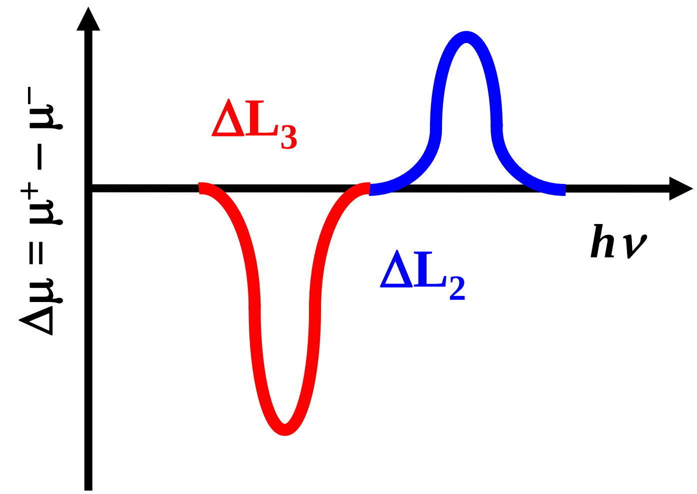
$$\Delta\mu = \mu_+ - \mu_+ = \boldsymbol{F}_{1,1} - \boldsymbol{F}_{1,-1}$$

XMCD Sum Rule

$$\boldsymbol{M}_{orb} \propto \Delta L_3 + \Delta L_2$$

$$\boldsymbol{M}_{spin} \propto \Delta L_3 - 2\Delta L_2$$

$$\frac{\boldsymbol{M}_{orb}}{\boldsymbol{M}_{spin}} \approx -\frac{2}{3}\frac{\Delta L_3 + \Delta L_2}{\Delta L_3 - 2\Delta L_2}$$



RSXS

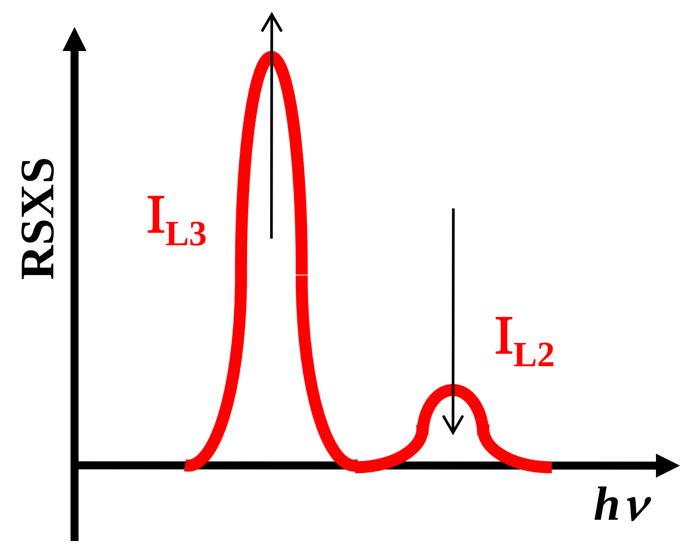
$$f^{res}_{mag} \propto (\varepsilon'^{*} \times \varepsilon) \cdot \hat{Z}[\boldsymbol{F}_{1,1} - \boldsymbol{F}_{1,-1}]$$

$$\sqrt{I_{L3}} \approx |\Delta L_3|$$

$$\sqrt{I_{L2}} \approx |\Delta L_2|$$

$$\boldsymbol{R}_{L3/L2} = \frac{I_{L3}}{I_{L2}}$$

$$\frac{\boldsymbol{M}_{orb}}{\boldsymbol{M}_{spin}} \approx \frac{2}{3}\frac{(-\boldsymbol{R}_{L3/L2} + 1)}{(-\boldsymbol{R}_{L3/L2} - 2)} \quad ?$$



Summary

- Co^{2+} HS states observed in magnetic orderings of (1/2, 0, 1/2) and (1/2, 0, 1).

- Co^{2+} has finite orbital moment of ~ $1\mu_B$.
  ➔ Co *3d* spin-orbit interaction > splitting of t_{2g} orbitals

Further Analysis

- Influence of tetragonal distortion on the Co^{2+} orbital moments