相関系の非弾性X線散乱

における偏光依存性

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PF研究会「軟X線分光・散乱測定を用いた物性研究の現状と展望」
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Acknowledgement

S. Ihara (Tohoku Univ.)

K. Ishii, T. Inami, J. Mizuki, Y. Murakami, Y. Endoh, K. Tsutsui

D. J. Huang, Y. Harada

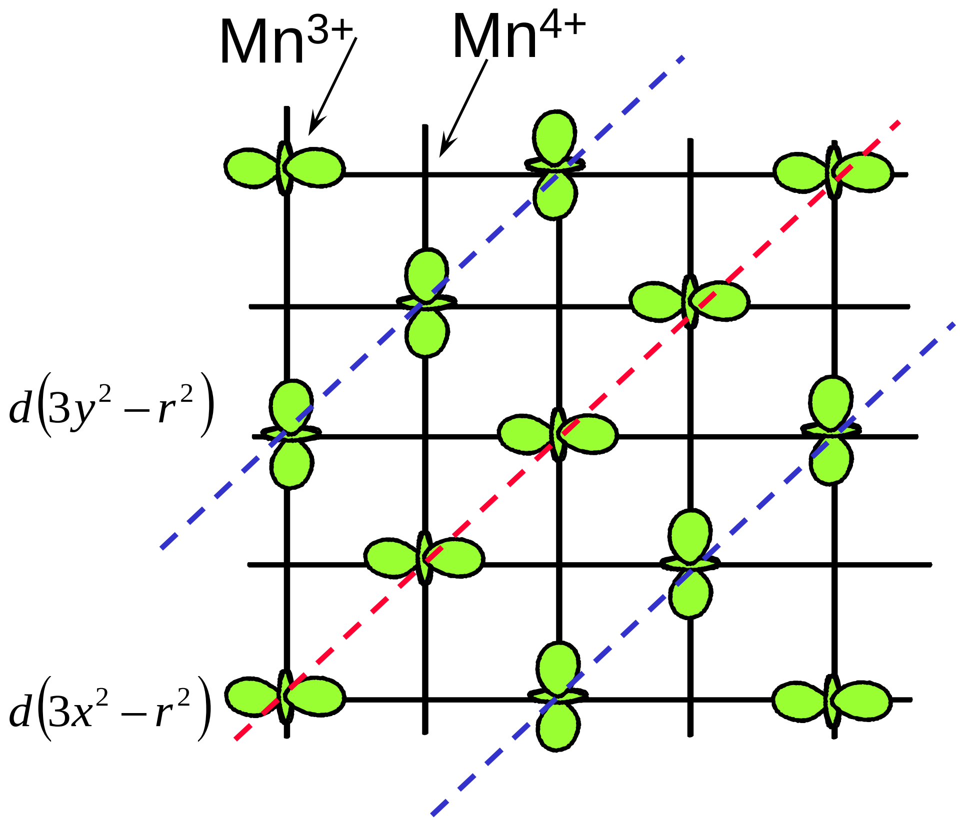
References

K. Ishii, S. Ishihara, Y. Murakami, K. Ikeuchi, K. Kuzushita, T. Inami, K. Ohwada, M. Yoshida, I. Jarrige, N. Tatami, S. Niioka, D. Bizen, Y. Ando, J. Mizuki, S. Maekawa, Y. Endoh
Phys. Rev. B 83, 241101(R) (2011) (editor's suggestion)

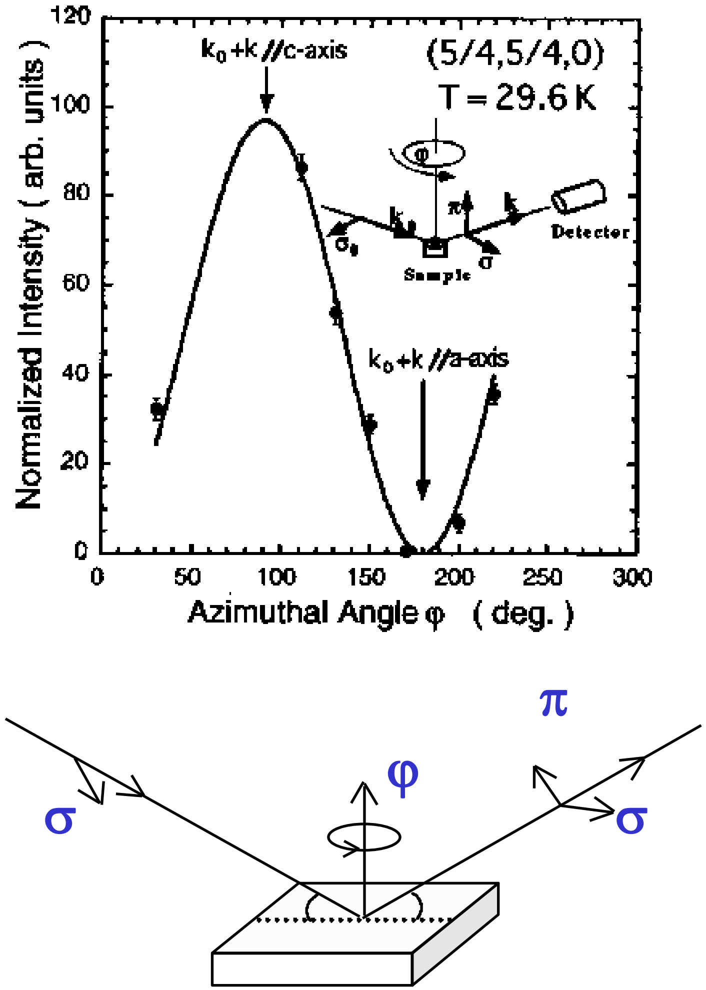
S. Ishihara and S. Ihara Jour. Phys. Chem. Sol. 69, 3184 (2008)

Resonant X-ray Scattering

$La_{0.5}Sr_{1.5}MnO_4$
(Murakami et al.)



Azimuthal angle dependence



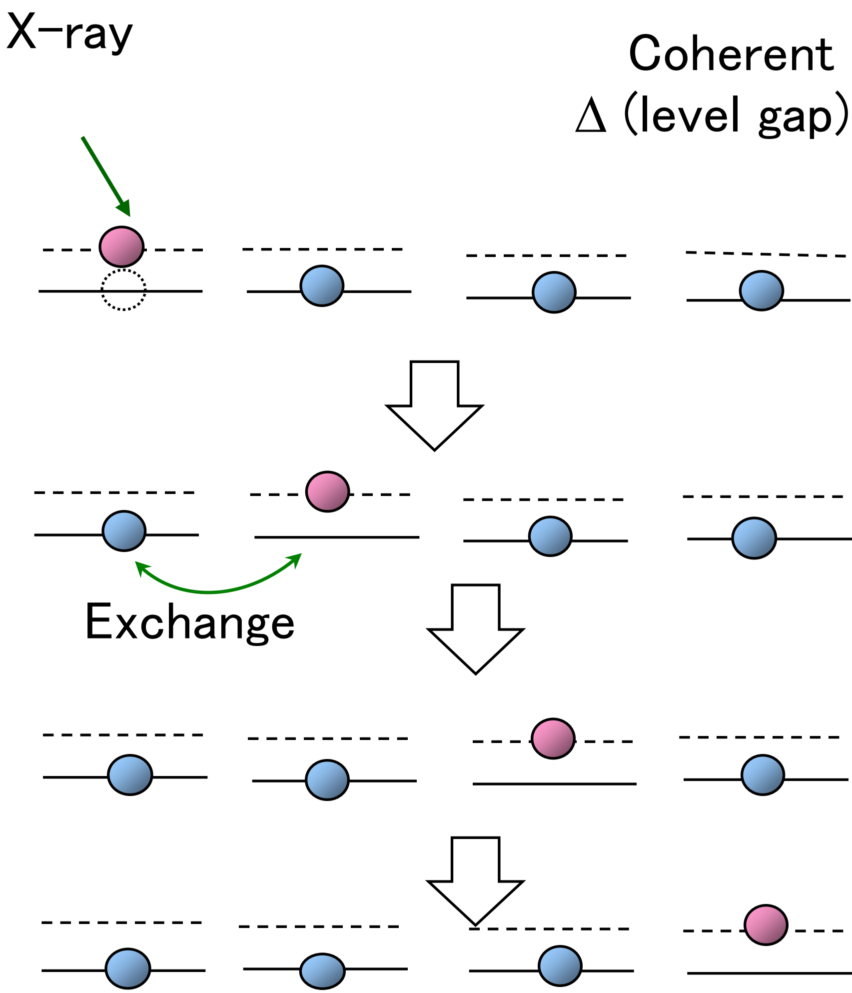
Orbital order & Polarization dependence

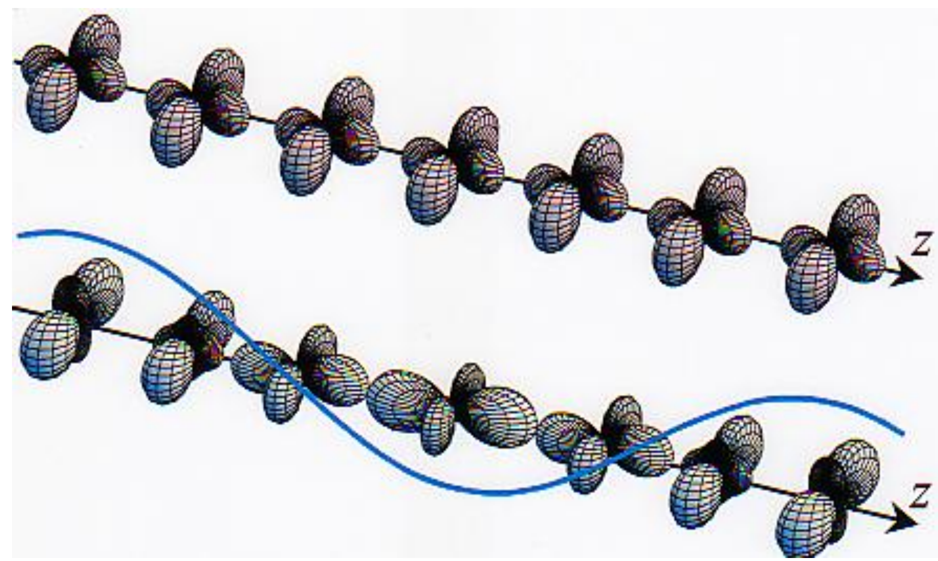
Orbiton

Orbital wave (orbiton) in orbital order

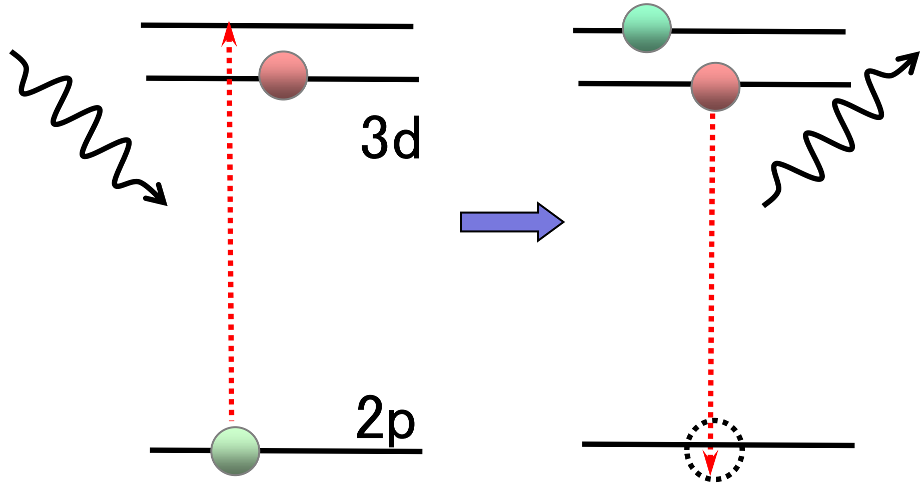
Coherent d–d excitation

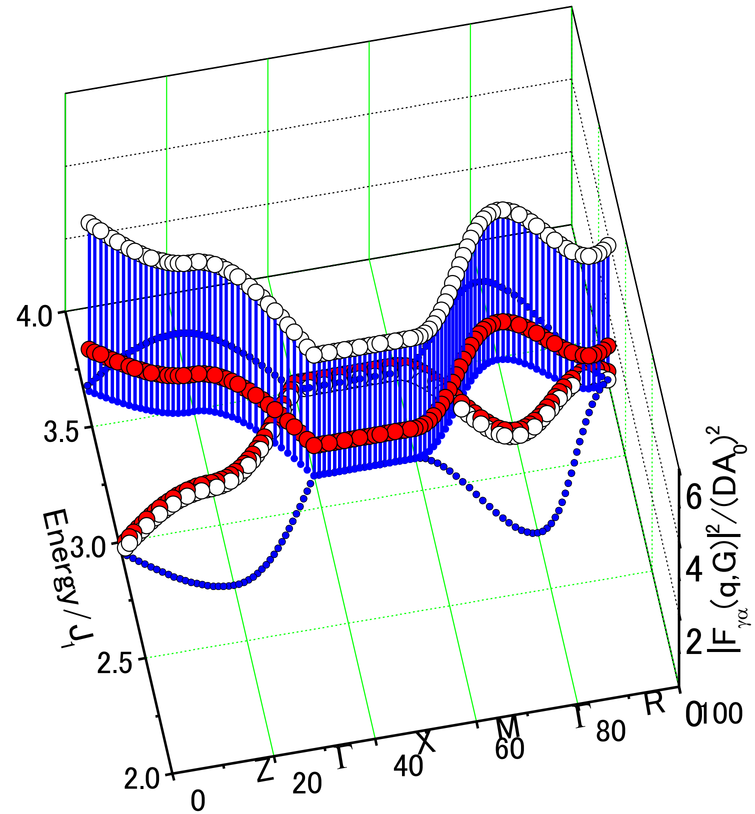
Δ (level gap) $\ll$ J (inter-site)





Orbiton detected by L-edge RIXS





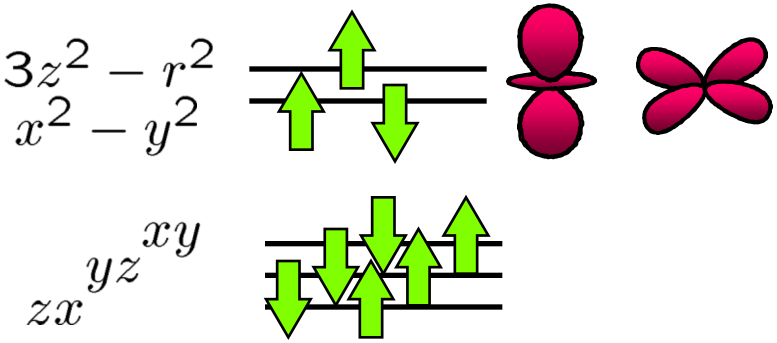
$$\frac{d\sigma^2}{d\Omega d\omega_f} = \left(\frac{e^2}{mc^2}\right)^2 \left(\frac{\omega_f}{\omega_i}\right)^2 (\vec{e}_i)^2_\alpha (\vec{e}_f)^2_\alpha S\left(\vec{k}_i - \vec{k}_f, \omega_i - \omega_f\right)$$

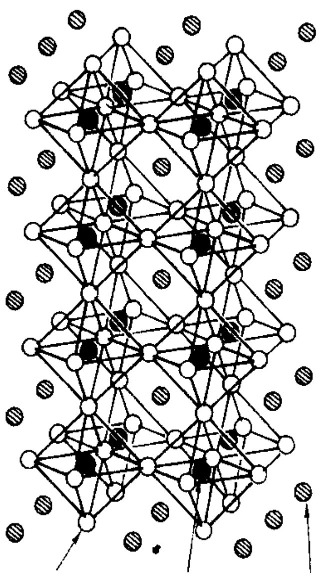
$$S\left(\vec{K}, \omega\right) = N\delta_{k_f - k_i + G + q}\,\delta\left(\omega_f - \omega_i - \omega_q^{(\mu)}\right)\left|\frac{F_{\mu\alpha}(q)}{\Delta E}\right|^2$$

SI, & Maekawa PRB 62, 2338 (‘00).

$KCuF_3$

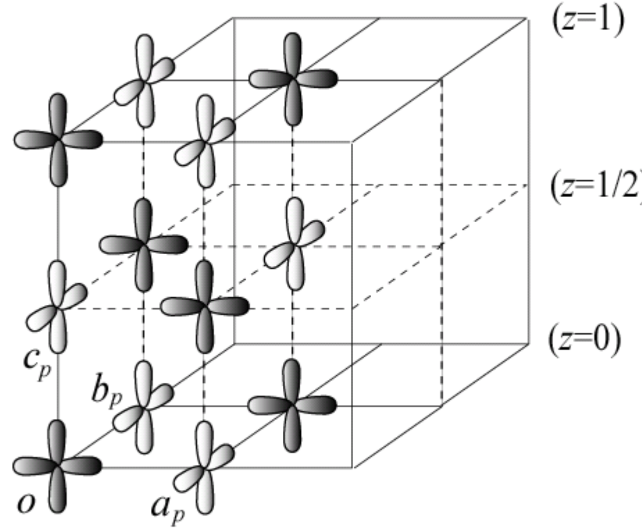
Cu^{2+} (d^9)





3-dim. Perovskite crystal

Orbital order with
Jahn-Teller distortion



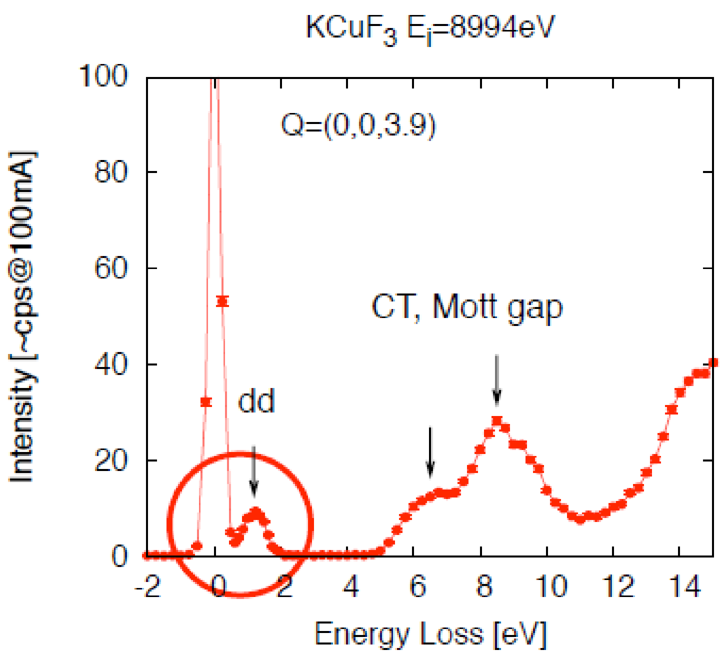
$d(y^2 - z^2)/d(z^2 - x^2)$ type

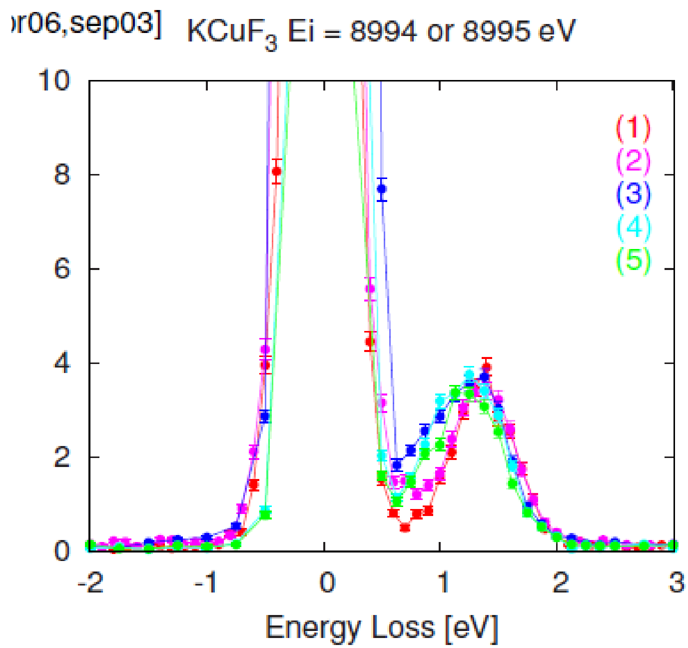
$$Q = (\pi, \pi, \pi)$$

Resonant x-ray scattering

Polarization dependence of RIXS

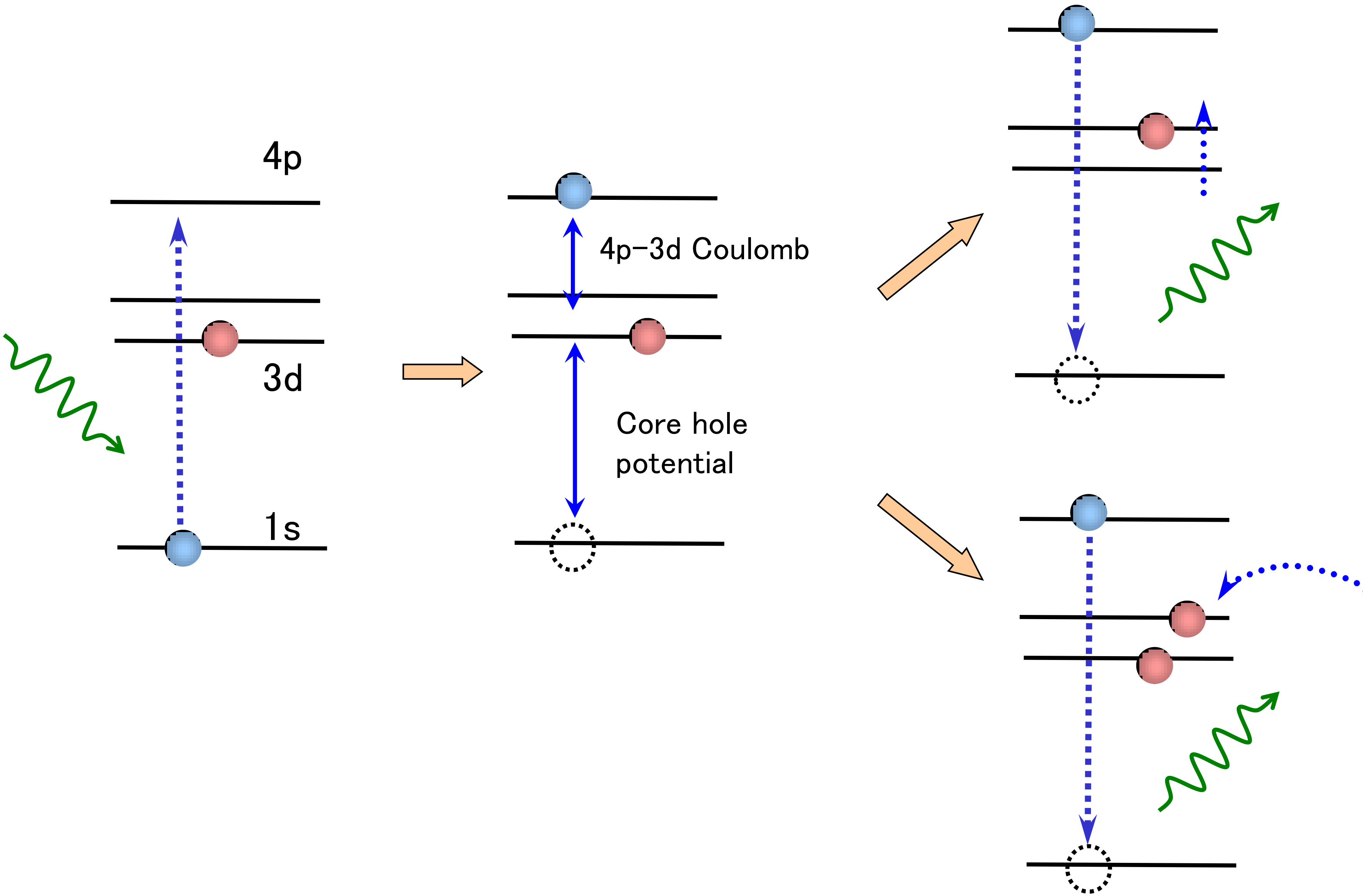
$KCuF_3$





Ishii–Kuzushita–Inami–Ohwada–
Niioka–Tatami–Mizuki–Endoh–Murakami

RIXS @ K-edge



Polarization dependence of RIXS

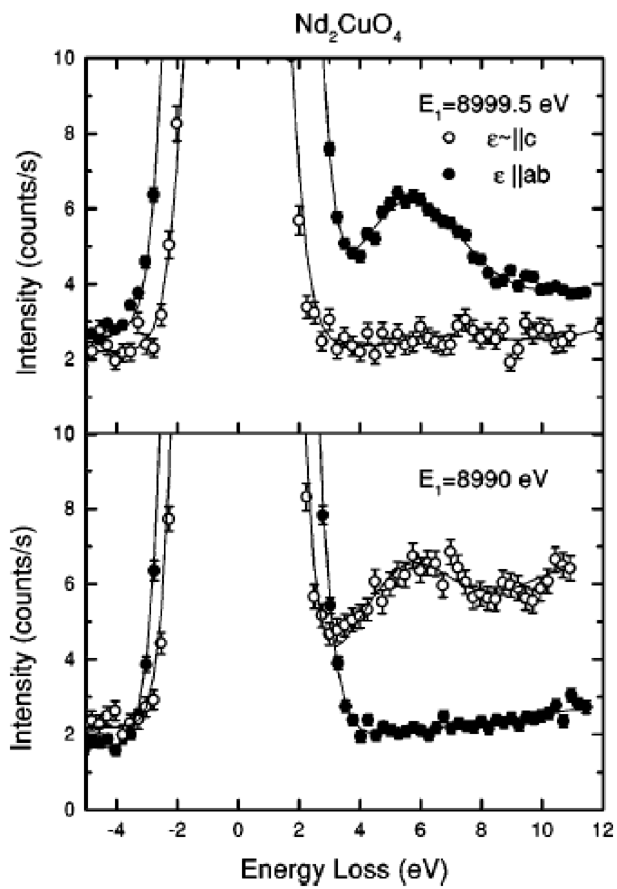


FIG. 2. Top panel: inelastic scattering for an incident energy of 8999.5 eV. Closed circles: incident polarization in the *ab* plane; open circles: incident polarization approximately along *c*. Bottom panel: same as top, with incident energy set to 8990 eV [$\epsilon \| c$ data (open circles) are multiplied by 5 to highlight the peak].

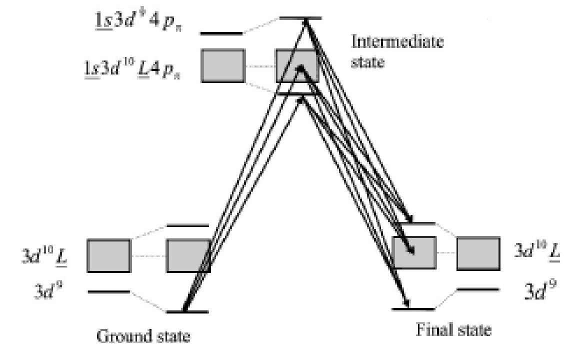


FIG. 4. Schematic energy level diagram for inelastic scattering from a copper site. Arrows indicate processes summed over in the calculation of the scattered intensity.

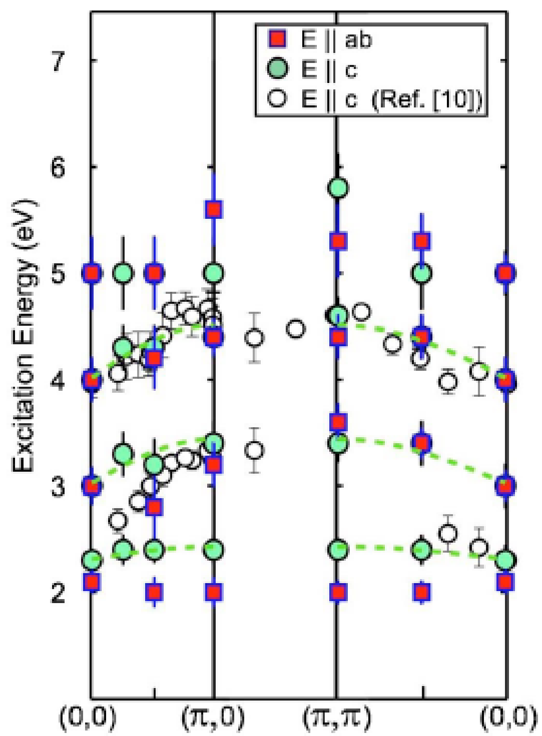
Hamainen-Hill-Kotani, PRB, 61 1836 (2000)

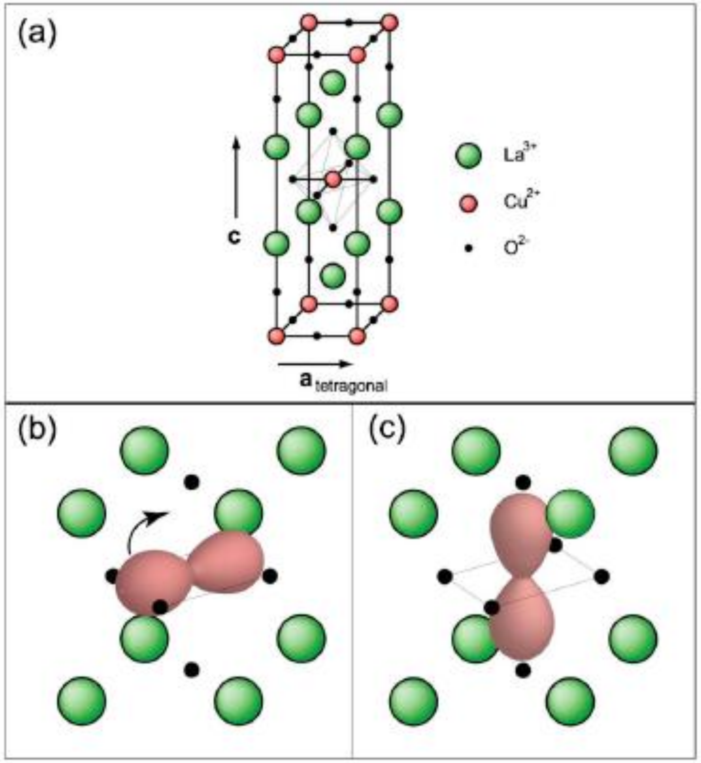
c. f. J. P. Hill, C. C. Kao, W. A. L. Caliebe, M. Matsubara, A. Kotani, J. L. Peng, and R. L. Greene, Phys. Rev. Lett. **80**, 4967 1998.

Polarization dependence of RIXS

Cuprate La_2CuO_4

Lu et al. PRB 74 224509 ('06)





Cu 4p –O 2p Coulomb interaction

O 2p –Cu 3d charge transfer

dd excitation in K-edge RIXS

M. van Veenendaal et al. PRB 83, 045101 ('11)

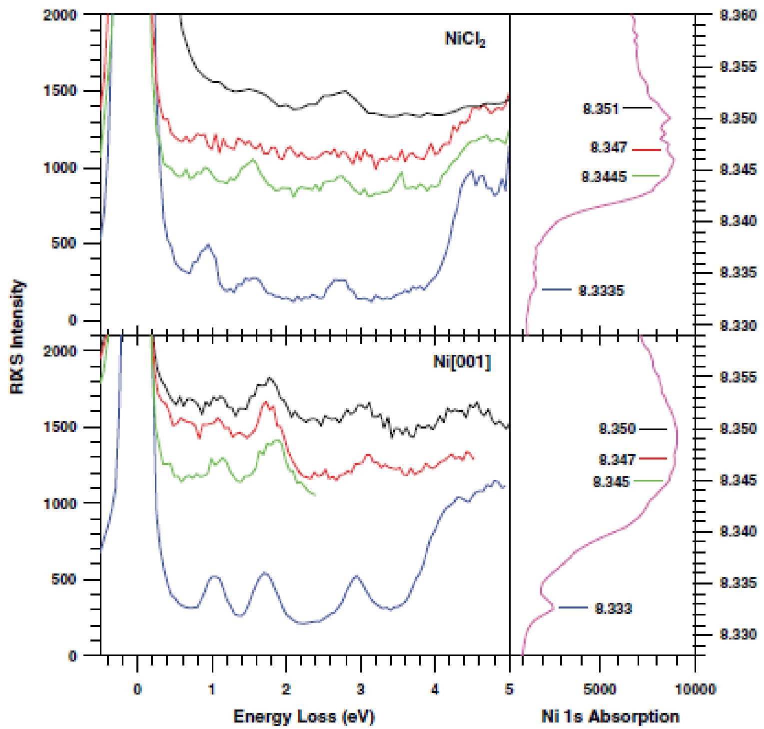


FIG. 1. (Color online) Ni K-edge RIXS of NiO[001] and NiCl$_2$. The x-ray absorption is shown in the right panel. RIXS spectra for different excitation energy are shown in the left panel.

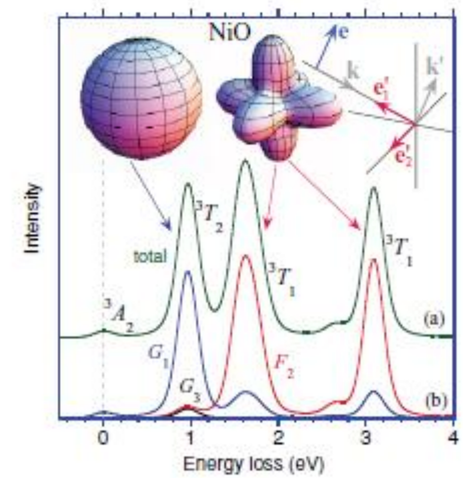


FIG. 2. (Color online) (a) The total RIXS intensity due to the p-d Coulomb interaction between the excited $4p$ electron and the $3d$ valence electrons in the intermediate state. A 90° horizontal scattering condition with the incoming polarization vectors 45° with respect to the z axis is used; see inset. (b) The RIXS intensity, but now separated into different terms in the p-d Coulomb interaction, where F^2 is related to the direct interaction and G^1 and G^3 are exchange interactions. The top shows the angular distributions as a function of the direction of the incoming polarization vector.

dd excitation & possibility of polarization flipping

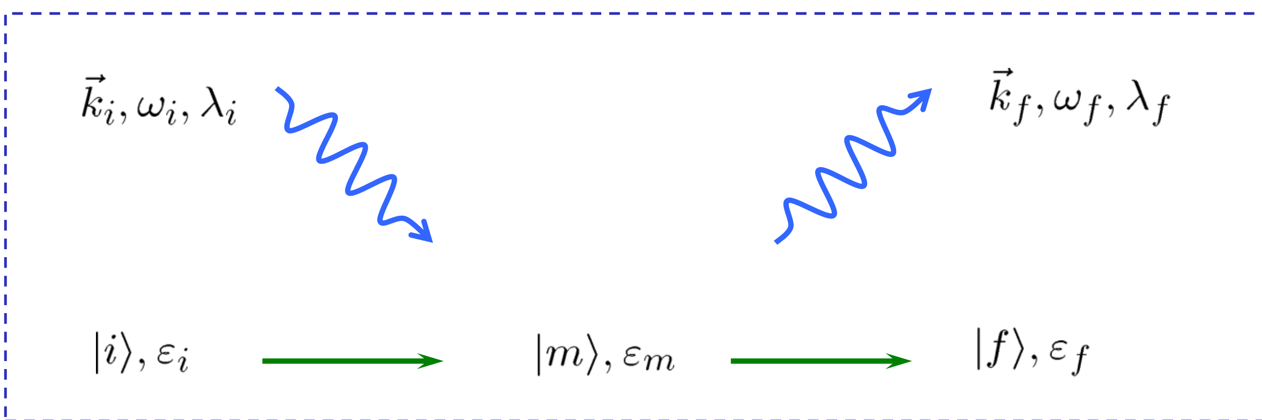
RIXS Cross Section

Kramers–Heisenberg formula

$$\frac{d^2\sigma}{d\Omega d\omega_f} = \left(\frac{e^2}{mc^2}\right)\left(\frac{\omega_f}{\omega_i}\right)\sum_{|f\rangle}\left|\sum_{\alpha\beta} e^{\alpha}_{k_f\lambda_f} S^{\alpha\beta} e^{\beta}_{k_i\lambda_i}\right|^2 \delta\left(\omega_f - \omega_i - E_f + E_i\right)$$

$$S^{\alpha\beta} = \sum_m \frac{\langle f|j^{\alpha}_{k_f}|m\rangle\langle m|j^{\beta}_{-k_i}|i\rangle}{E_i - E_m + \omega_i + i\Gamma/2}$$

$$j^{\alpha}_{k} = \sum_{k_0\sigma} A\left(p^{\dagger}_{k-k_0\alpha\sigma} s^{\dagger}_{k_0\sigma} + p_{k_0-k\alpha\sigma} s_{-k_0\sigma}\right)$$



Phenomenological theory

Ishihara-Maekawa, PRB 62, R9252, (00)
Ishihara-Kondoh-Maekawa, Physica B 345, 15 (04)

"For both L- & K-edge RIXS"

$$\frac{d^2\sigma}{d\Omega d\omega_f} = \left(\frac{e^2}{mc^2}\right) \sum_{\alpha\alpha'\beta\beta'} P_{\alpha\alpha'\beta\beta'} \Pi_{\beta'\alpha'\beta\alpha}(\vec{K},\omega)$$

$$\Pi_{\beta'\alpha'\beta\alpha}(\vec{K},\omega) = \frac{1}{2\pi}\int dt e^{i\omega t} \sum_{ij} e^{-i\vec{K}\cdot(\vec{r}_i-\vec{r}_j)} \langle \alpha_{j\alpha'\beta'}(t)^\dagger \alpha_{i\beta\alpha}(0)\rangle$$

Correlation function of polarizability

$$P_{\alpha\alpha'\beta\beta'} = \vec{e}_{i\alpha}\vec{e}_{f\beta}\vec{e}_{i\alpha'}\vec{e}_{f\beta'}$$ Polarization part

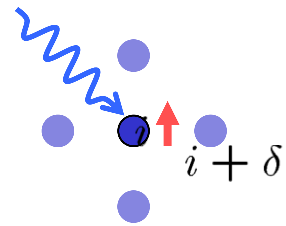
$$\alpha_{i\alpha\beta} = (-i)\int_{-\infty}^{\infty} \theta(-t) e^{i\omega t}[j_{i\beta}(t), j_{i\alpha}(0)]$$

Polarizability tensor at i-site

Phenomenological theory

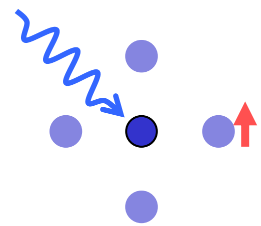
Expansion of polarizability based on symmetry

$$\alpha_{i\alpha\beta} = \sum_{\Gamma} I^{\Gamma}_{\beta\alpha} O^{\Gamma}_{i} \xrightarrow{\text{F.T.}} \sum_{\Gamma} I^{\Gamma}_{\beta\alpha} O^{\Gamma}_{K}$$



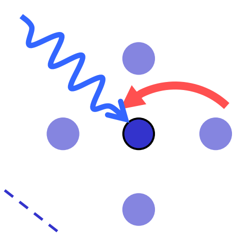
On-site excitation

$$+\sum_{\Gamma\gamma} I^{\Gamma}_{\beta\alpha} \sum_{\delta\Gamma_1} C^{\Gamma\Gamma_1}_{\delta} O^{\Gamma_1}_{i+\delta} \xrightarrow{\text{F.T.}} \sum_{\Gamma} \boxed{\tilde{I}^{\Gamma K}_{\beta\alpha}} O^{\Gamma}_{K}$$



Off-site excitation

$$+\sum_{\Gamma} I^{\Gamma}_{\beta\alpha} \sum_{\delta} \sum_{\Gamma_2\Gamma_1} C^{\Gamma\Gamma_1\Gamma_2}_{\delta} O^{\Gamma_1}_{i} O^{\Gamma_2}_{i+\delta} \xrightarrow{\text{F.T.}} \sum_{\Gamma_1\Gamma_2 p} \boxed{\tilde{I}^{\Gamma_1\Gamma_2 K+p}_{\beta\alpha}} O^{\Gamma_1}_{-p} O^{\Gamma_2}_{K+p}$$



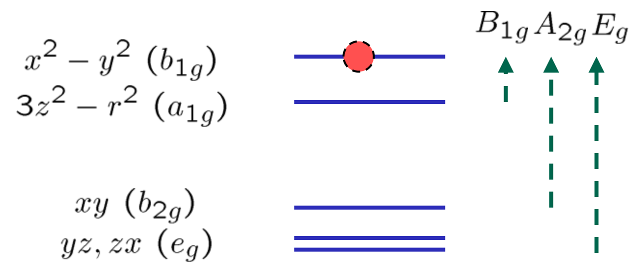
Inter-site excitation

Momentum-Polarization cross term

Γ	Irrducible representation and its basis
O^{Γ}_{i}	Excitation at i-site with symmetry Γ
$I^{\Gamma}_{\beta\alpha}$	(β,α) componet of polarization with symmetry Γ

Phenomenological theory

Example: d^9 D_{4h} in 2-dim. square lattice



On-site excitation

$$\alpha_{K\alpha\beta} \sim I_{\alpha\beta}^{B_{1g}} O_K^{B_{1g}} + I_{\alpha\beta}^{A_{2g}} O_K^{A_{2g}} + I_{\alpha\beta}^{E_g} O_K^{E_g}$$

B_{1g} polarization　　B_{1g} excitation

B_{1g}(A_{1g}) excitation is detected in B_{1g} (A_{1g}) polarization (no-momentum dependence)

Off-site excitation

$$\begin{aligned}\alpha_{K\alpha\beta} \sim\ & \left[I_{\alpha\beta}^{B_{1g}}(\cos K_x+\cos K_y)+I_{\alpha\beta}^{A_{1g}}(\cos K_x-\cos K_y)+I_{\alpha\beta}^{E_{gx}}\sin K_x+I_{\alpha\beta}^{E_{gy}}\sin K_y\right]O_K^{B_{1g}}\\ +\ & \left[I_{\alpha\beta}^{A_{2g}}(\cos K_x+\cos K_y)+I_{\alpha\beta}^{B_{2g}}(\cos K_x-\cos K_y)+I_{\alpha\beta}^{E_{gx}}\sin K_x+I_{\alpha\beta}^{E_{gy}}\sin K_y\right]O_K^{A_{2g}}\\ +\ & \cdots\end{aligned}$$

B_{1g} excitation is detected in B_{1g} as well as A_{1g} and E_g polarization

At K=0, momentum-polarization cross effect disappears

Selection Rule for RIXS

$$\langle \phi_{3d\gamma}\phi_{4p\alpha}|e^{iq(r_1-r_2)}|\phi_{3d\gamma'}\phi_{4p\alpha'}\rangle \sim A_{1g}$$

c.f. P. Abbamonte et al. cond-mat/9911215

$I(xx) \neq I(yy)$ along $\Gamma - X$

$(zz) : (x^2-y^2) \rightarrow (3z^2-r^2)$
along $\Gamma - X$

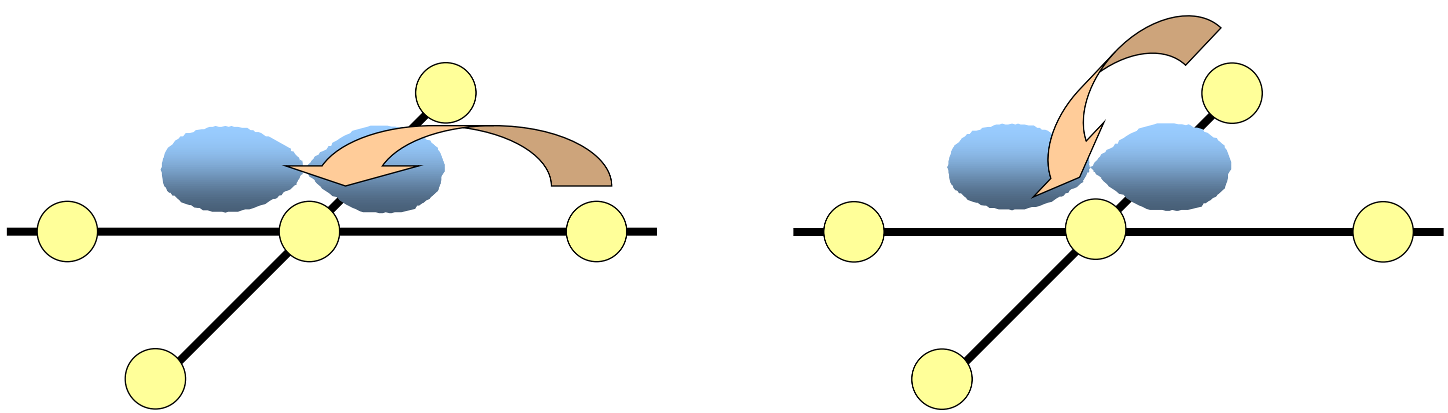
Local excitation

$$\langle \phi_{3d\gamma}(\vec{e}_i)_\alpha|1|\phi_{3d\gamma'}(\vec{e}_f)_{\alpha'}\rangle \sim A_{1g}$$

$(xx) : (3x^2-r^2) \rightarrow (x^2-y^2)$

$(xy) : (xy) \rightarrow (3z^2-r^2)$

Same with Raman



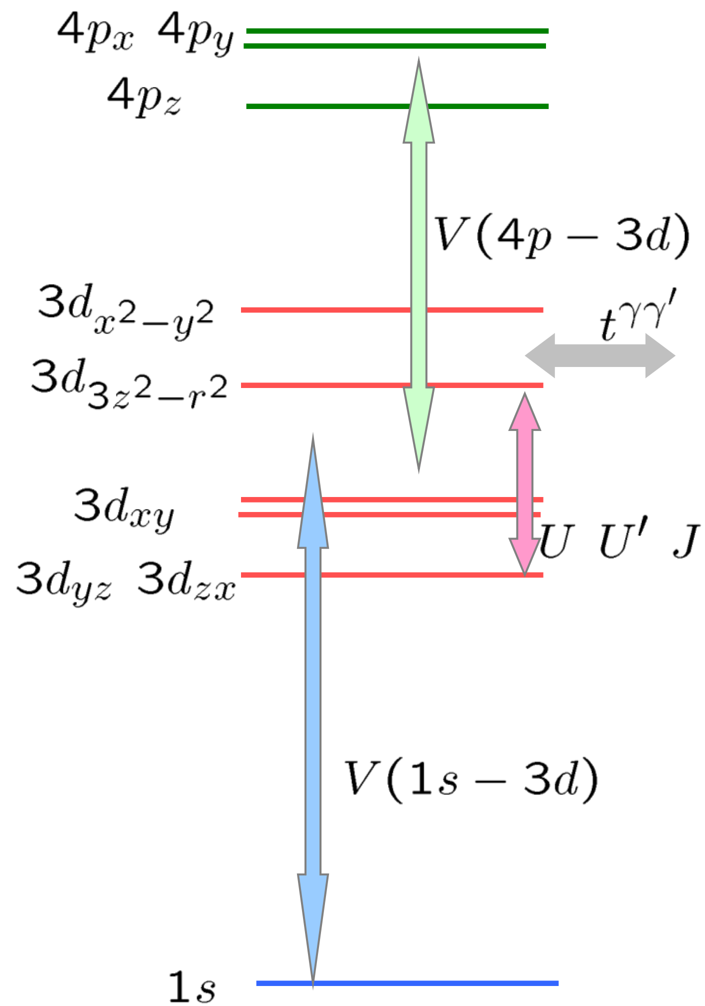
Polarization/momentum combining effect

Model

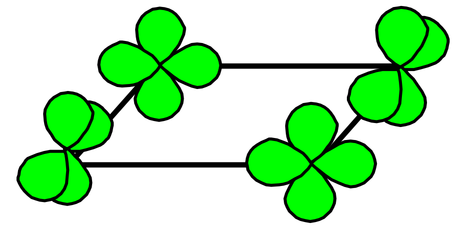
$$\mathcal{H} = \mathcal{H}_{3d} + \mathcal{H}_{1s} + \mathcal{H}_{4p} + \mathcal{H}_{3d-1s/4p}$$

$$\begin{aligned}
\mathcal{H}_{3d} &= \sum_{i\gamma\sigma} \varepsilon_\gamma d^\dagger_{i\gamma\sigma} d_{i\gamma\sigma} \\
&+ \sum_{\langle ij\rangle \gamma\gamma'\sigma} \left(t^{\gamma\gamma'}_{ij} d^\dagger_{i\gamma\sigma} d_{j\gamma'\sigma} + \text{H.c.} \right) \\
&+ \sum_{i\gamma} U n_{i\gamma\uparrow} n_{i\gamma\downarrow} \\
&+ \sum_{i\gamma>\gamma'} U'_{\gamma\gamma'} n_{i\gamma} n_{i\gamma'} \\
&+ \sum_{i\gamma>\gamma'} J_{\gamma\gamma'} d^\dagger_{i\gamma\sigma} d^\dagger_{i\gamma'\sigma'} d_{i\gamma\sigma} d_{i\gamma'\sigma'}
\end{aligned}$$

$$\mathcal{H}_{4p} + \mathcal{H}_{1s} = \sum_{i\alpha\sigma} \varepsilon^p_\alpha p^\dagger_{i\alpha\sigma} p_{i\alpha\sigma} + \sum_{i\sigma} \varepsilon^s s^\dagger_{i\sigma} s_{i\sigma}$$



Method



- 2×2 2-dimensional square lattice
  with periodic boundary condition

- 5×3d orbitals
  3×4p orbitals
  1s orbital at each sites

- Total hole # of d(3z), d(yz), d(zx),d(xy) <= 2

- 4p orbital : flat band

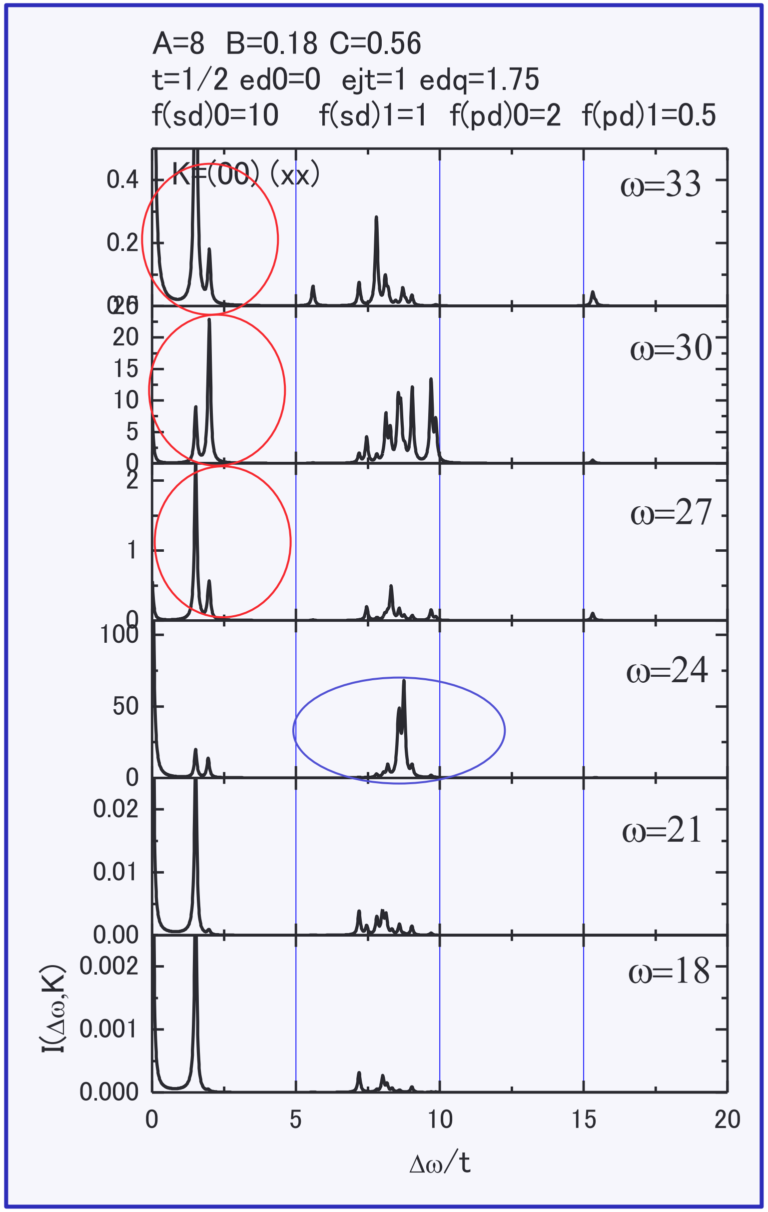
- Exact diagonalization by Lanczos algorithm

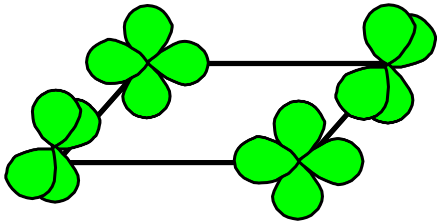
- RIXS spectra by the modified conjugate gradient method
  and the recursion method

$$S^{\alpha\beta} = \sum_m \frac{\langle f|j^{\alpha}_{k_f}|m\rangle\langle m|j^{\beta}_{-k_i}|i\rangle}{E_i - E_m + \omega_i + i\Gamma/2}$$

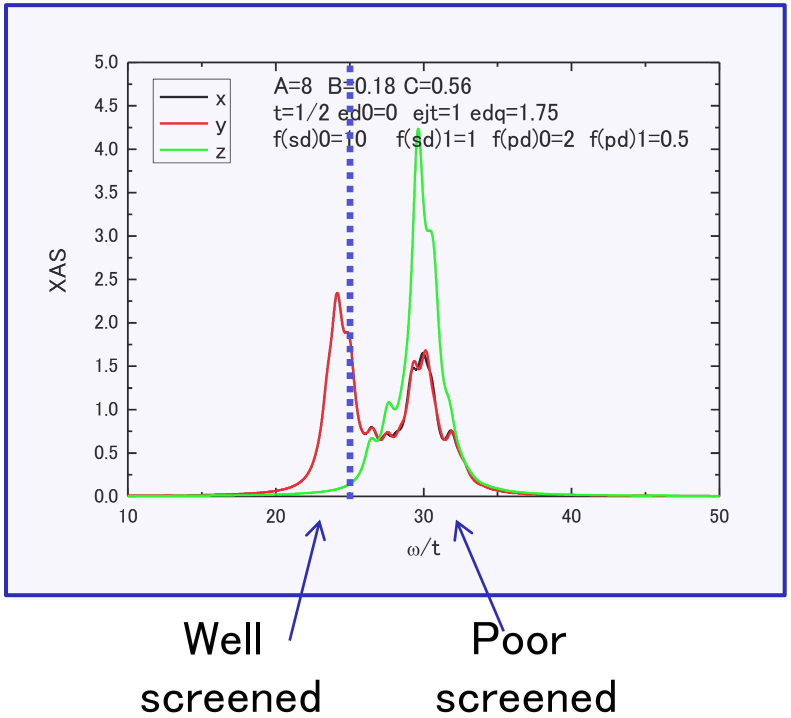
XAS & RIXS

RIXS

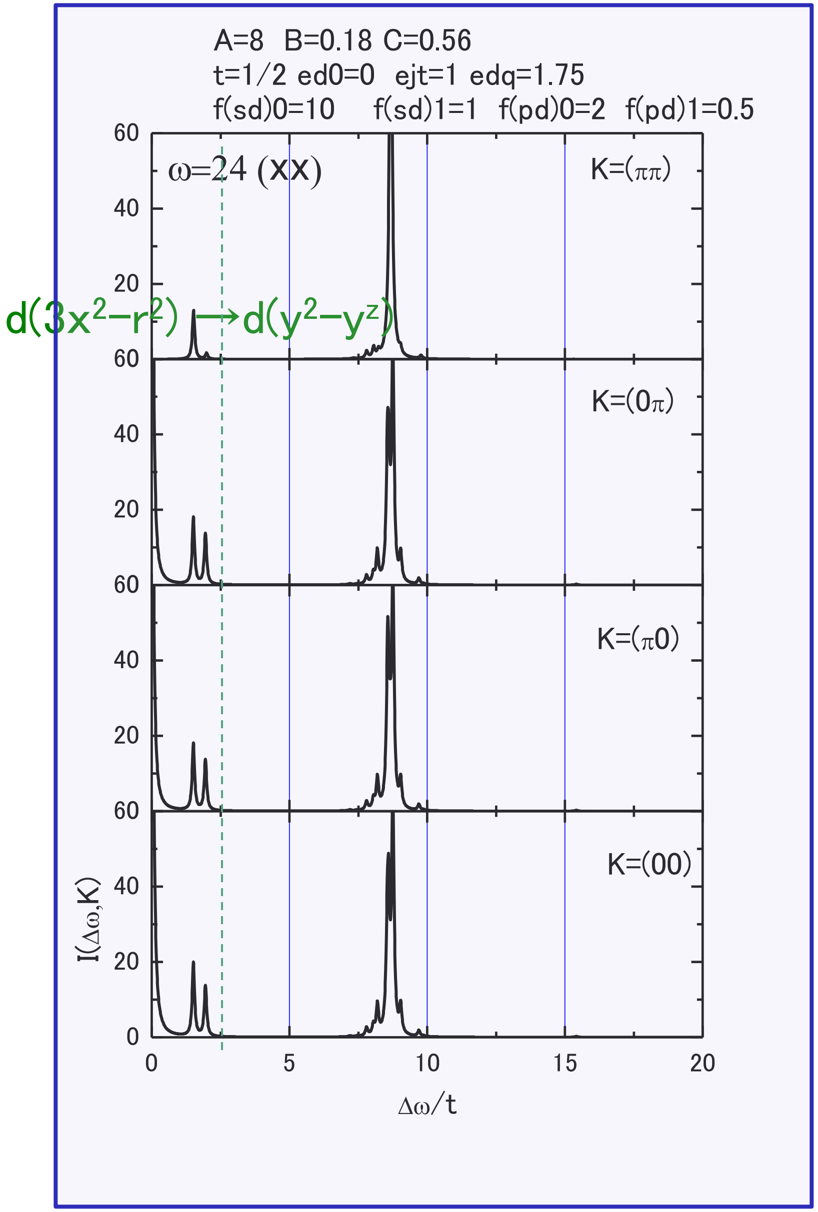




XAS



$KCuF_3$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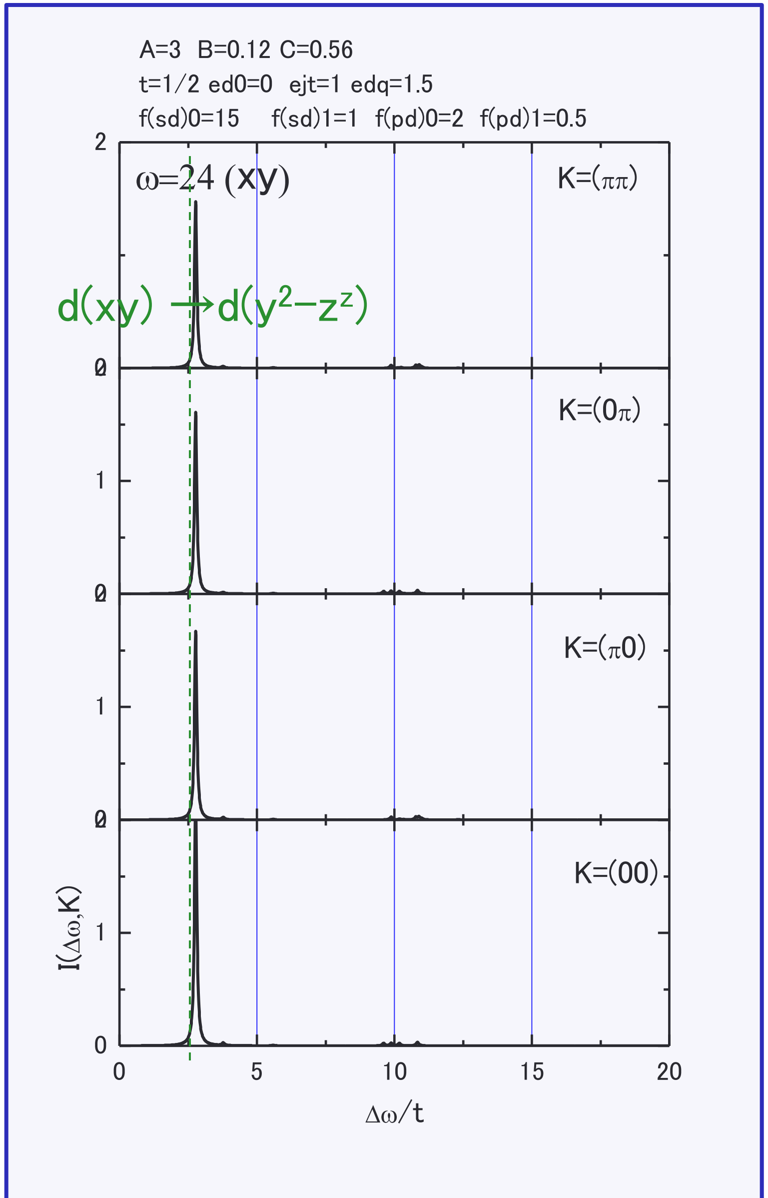
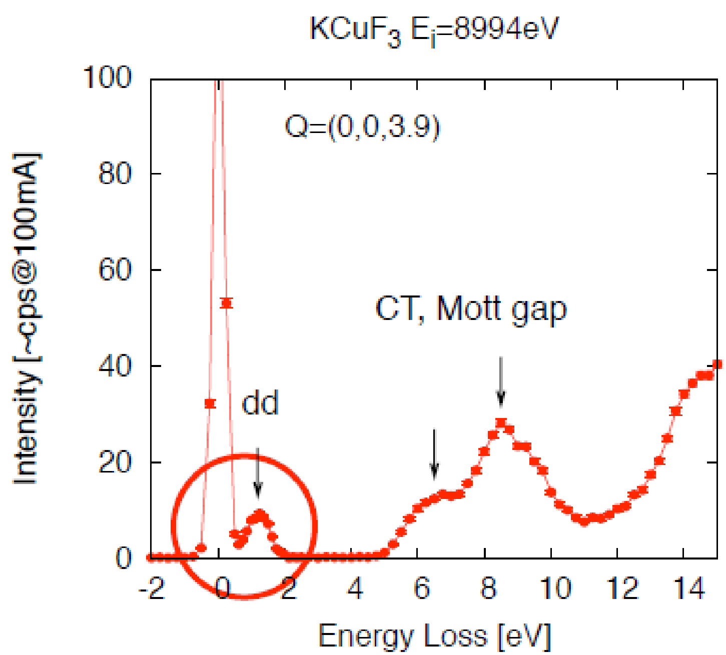
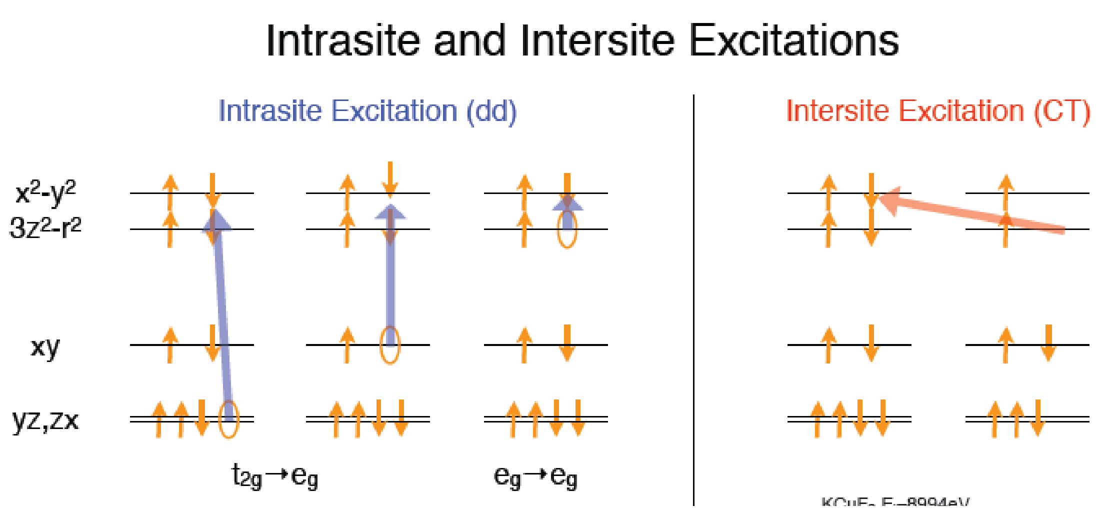
t=1/2 ed0=0 ejt=1 edq=1.75
f(sd)0=10 f(sd)1=1 f(pd)0=2 f(pd)1=0.5
ω=24 (xx)
K=(ππ)
K=(0π)
K=(π0)
K=(00)
d(3x²−r²) →d(y²−yᶻ)
I(Δω,K)
Δω/t
0
5
10
15
20
60
40
20
0


A=3 B=0.12 C=0.56
t=1/2 ed0=0 ejt=1 edq=1.5
f(sd)0=15 f(sd)1=1 f(pd)0=2 f(pd)1=0.5
ω=24 (xy)
K=(ππ)
K=(0π)
K=(π0)
K=(00)
d(xy) →d(y²−zᶻ)
I(Δω,K)
Δω/t
0
5
10
15
20
2
1
0

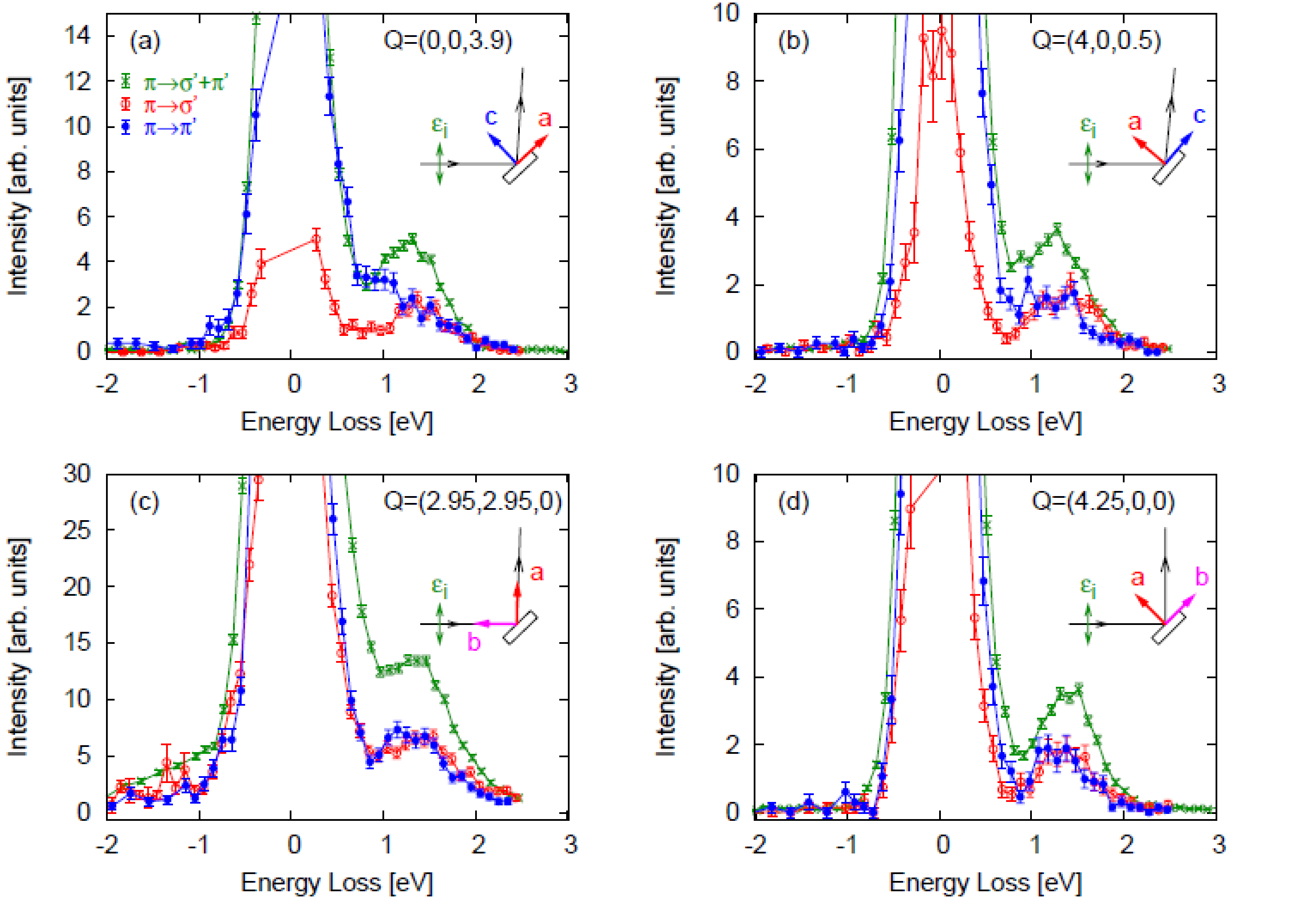
RIXS experiments in $KCuF_3$



Ishii–Kuzushita–Inami–Ohwada–Niioka–Tatami–Murakami



RIXS experiments in $KCuF_3$



RIXS experiments in $KCuF_3$

TABLE I: Summary of the polarization conditions of Fig. 2.

configuration	polarization	ϵ_i	ϵ_f	symmetry of $P_i \times P_f$
(a)	$\pi \to \sigma'$	$y+z$	x	$A_{2g}+B_{2g}+E_g$
	$(a+c) \to b$	$x+y$	z	E_g
	$\pi \to \pi'$	$y+z$	$y-z$	$A_{1g}+B_{1g}+E_g$
	$(a+c) \to (a-c)$	$x+y$	$x-y$	$B_{1g}+A_{2g}$
(b)	$\pi \to \sigma'$	$y+z$	x	$A_{2g}+B_{2g}+E_g$
	$(a+c) \to b$	$x+y$	z	E_g
	$\pi \to \pi'$	$y+z$	$y-z$	$A_{1g}+B_{1g}+E_g$
	$(a+c) \to (a-c)$	$x+y$	$x-y$	$B_{1g}+A_{2g}$
(c)	$\pi \to \sigma'$	z	y	E_g
	$a \to c$	y	x	$A_{2g}+B_{2g}$
	$\pi \to \pi'$	z	x	E_g
	$a \to b$	y	z	E_g
(d)	$\pi \to \sigma'$	$x+z$	y	$A_{2g}+B_{2g}+E_g$
	$(a+b) \to c$	$y+z$	x	$A_{2g}+B_{2g}+E_g$
	$\pi \to \pi'$	$x+z$	$x-z$	$A_{1g}+B_{1g}+E_g$
	$(a+b) \to (a-b)$	$y+z$	$y-z$	$A_{1g}+B_{1g}+E_g$

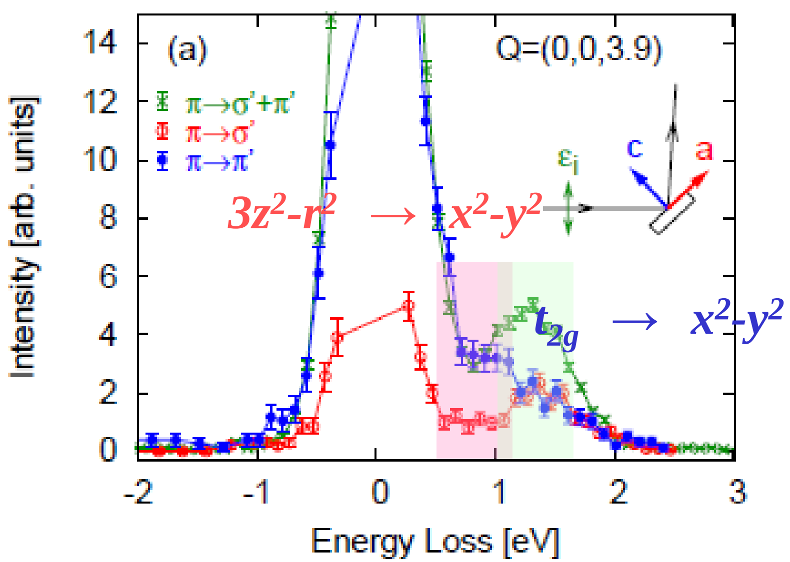


TABLE II: Symmetry of the orbital excitations of $KCuF_3$.

excitation	symmetry of $\Gamma_i \times \Gamma_f$
$(3z^2-r2) \to (x^2-y^2)A$	B_{1g}
$(xy) \to (x^2-y^2)$	A_{2g}
$(yz) \to (x^2-y^2)$	E_g
$(zx) \to (x^2-y^2)$	E_g

Ishii–Kuzushita–Inami–Ohwada–Niioka–Tatami–Murakami

Summary

Polarization dependence of RIXS
in correlated electron systems

Selection Rule for RIXS
Scattering vector reflects on q
Raman type rule for local excitations & its breaking at finite q

Orbital ordered $KCuF_3$
Polarization dependence for dd excitation

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Charge (collective) excitation in electronic ferroelectricity