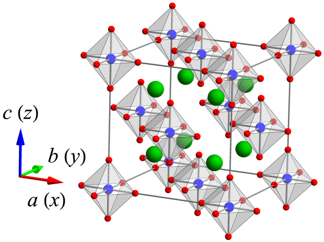
Vibronic excitations in the resonant inelastic x-ray scattering spectra of spin-orbit Mott insulators

Naoya Iwahara[1], Shouta Shikano[2], and Wataru Furukawa[2]

[1] Graduate School of Engineering, Chiba University, Japan.
[2] Department of Materials Science, Faculty of Engineering, Chiba University, Japan.

The 25th international conference on the Jahn-Teller conference, York, 11, 14-18 May 2023

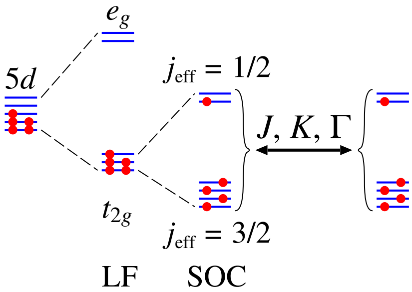
N. Iwahara and S. Shikano, Physical Review Research **5**, 023051, (2023).
N. Iwahara and W. Furukawa, arXiv:2305.05853



Antifluorite structure
Green: K
Blue: Ir ($5d^5$)
Red: Cl

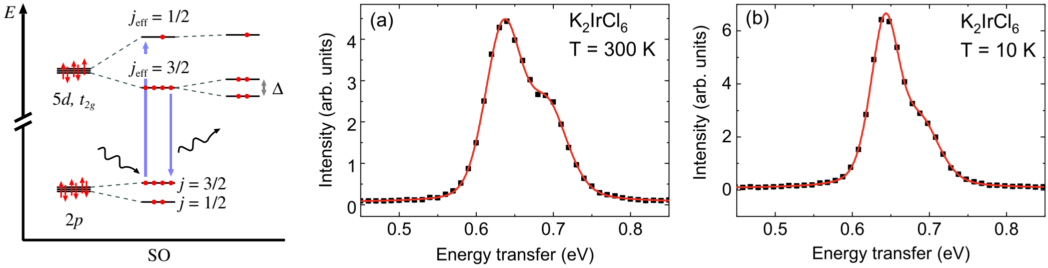
N. Khan *et al.*, Phys. Rev. B **99**, 144425 (2019).
D. Reig-i-Plessis *et al.*, Phys. Rev. Mater. **4**, 124407 (2020).

- Fcc down to 0.3 K (neutron diffraction data)
- Spin-orbit coupling on $5d^5$ site
  $\Rightarrow J_{\rm eff} = 1/2$ ground state
- Kitaev exchange interaction between $5d^5$ sites



Resonant inelastic x-ray scattering (RIXS) measurements

To confirm $J_{\mathrm{eff}} = 1/2$ ground state $\Rightarrow$ RIXS measurements



D. Reig-i-Plessis *et al.*, Phys. Rev. Mater. **4**, 124407 (2020).

What is the origin of the splitting of the peak in the RIXS spectra?

This work

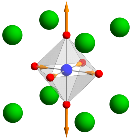
Demonstrate that the dynamic Jahn-Teller effect changes the shape of the RIXS spectra

Model Hamiltonian for single $5d^5$ site:

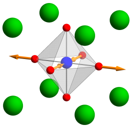
$$\hat{H} = \hat{H}_{\mathrm{SO}} + \hat{H}_{\mathrm{JT}}, \tag{1}$$

$$\hat{H}_{\mathrm{SO}} = \lambda \hat{\boldsymbol{l}} \cdot \hat{\boldsymbol{s}}, \tag{2}$$

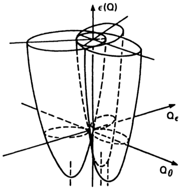
$$\hat{H}_{\mathrm{JT}} = \sum_{\alpha=u,v} \frac{\hbar\omega}{2}\left(\hat{p}_\alpha^2 + \hat{q}_\alpha^2\right) + \sum_{\sigma=\uparrow\downarrow} \hbar\omega g \left[\hat{P}_{yz,\sigma} \underbrace{\left(-\frac{1}{2}\hat{q}_u + \frac{\sqrt{3}}{2}\hat{q}_v\right)}_{x^2} + \hat{P}_{zx,\sigma} \underbrace{\left(-\frac{1}{2}\hat{q}_u - \frac{\sqrt{3}}{2}\hat{q}_v\right)}_{y^2} + \hat{P}_{xy,\sigma}\hat{q}_u\right]. \tag{3}$$



$u(z^2)$



$v(x^2 - y^2)$



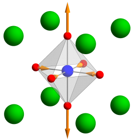
Potential surface

Model Hamiltonian (SO multiplets)

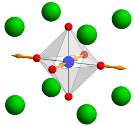
In the SO basis:

$$\hat{H} = \lambda \begin{pmatrix} -\boldsymbol{I}_2 & \boldsymbol{0} \\ \boldsymbol{0} & \frac{1}{2}\boldsymbol{I}_4 \end{pmatrix}$$

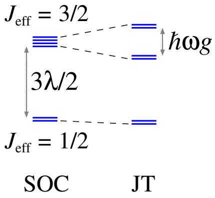
$$+ \sum_{\alpha=u,v} \frac{\hbar\omega}{2}\left(\hat{p}_\alpha^2 + \hat{q}_\alpha^2\right) + \frac{\hbar\omega g}{2} \begin{pmatrix} 0 & 0 & 0 & \sqrt{2}\hat{q}_v & 0 & -\sqrt{2}\hat{q}_u \\ 0 & 0 & \sqrt{2}\hat{q}_u & 0 & -\sqrt{2}\hat{q}_v & 0 \\ 0 & \sqrt{2}\hat{q}_u & -\hat{q}_u & 0 & -\hat{q}_v & 0 \\ \sqrt{2}\hat{q}_v & 0 & 0 & \hat{q}_u & 0 & -\hat{q}_v \\ 0 & -\sqrt{2}\hat{q}_v & -\hat{q}_v & 0 & \hat{q}_u & 0 \\ -\sqrt{2}\hat{q}_u & 0 & 0 & -\hat{q}_v & 0 & -\hat{q}_u \end{pmatrix}. \qquad (4)$$

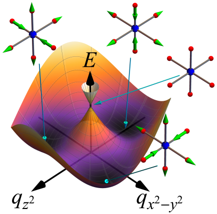


$u(z^2)$



$v(x^2-y^2)$

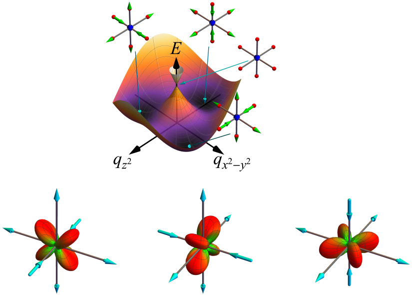




Potential surface

Static Jahn-Teller effect

q, $p = 0$: c-numbers

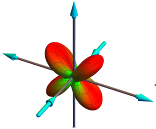
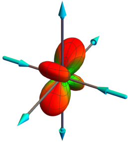


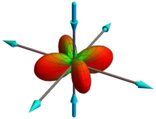
One orbital-lattice configuration

Dynamic Jahn-Teller effect

$\hat{q}$, $\hat{p}$: Quantum dynamic variables

$\Rightarrow$ Tunneling between the minima:

$|\Psi_\nu\rangle \approx c_{1\nu}$  $+ c_{2\nu}$ 

$+ c_{3\nu}$ 

Orbital-lattice entangled (vibronic) state

Hamiltonian:

$$\hat{H} = \underbrace{\hat{H}_{\text{SO}} + \sum_{\alpha=u,v} \frac{\hbar\omega}{2}\left(\hat{p}_\alpha^2 + \hat{q}_\alpha^2\right)}_{\text{Unperturbed Hamiltonian}, \hat{H}_0} + \frac{\hbar\omega g}{2} \begin{pmatrix} 0 & 0 & 0 & \sqrt{2}\hat{q}_v & 0 & -\sqrt{2}\hat{q}_u \\ 0 & 0 & \sqrt{2}\hat{q}_u & 0 & -\sqrt{2}\hat{q}_v & 0 \\ 0 & \sqrt{2}\hat{q}_u & -\hat{q}_u & 0 & -\hat{q}_v & 0 \\ \sqrt{2}\hat{q}_v & 0 & 0 & \hat{q}_u & 0 & -\hat{q}_v \\ 0 & -\sqrt{2}\hat{q}_v & -\hat{q}_v & 0 & \hat{q}_u & 0 \\ -\sqrt{2}\hat{q}_u & 0 & 0 & -\hat{q}_v & 0 & -\hat{q}_u \end{pmatrix}.$$

Vibronic basis (Eigenstates of $\hat{H}_0$):

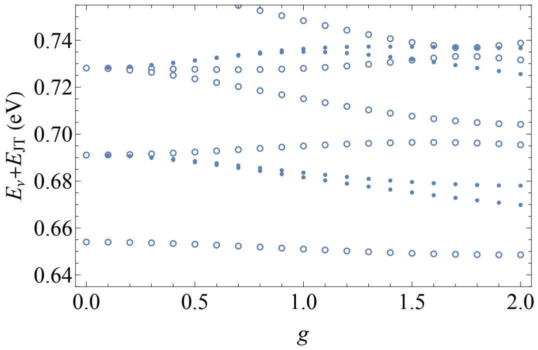
$$\{|J_{\text{eff}}, M_J\rangle \otimes |n_u, n_v\rangle | J_{\text{eff}} = 1/2, 3/2; 0 \le n_u + n_v \le 20\}. \quad (5)$$

Vibronic states:

$$|\Psi_\nu\rangle = \sum_{J_{\text{eff}} M_J} \sum_{n_u n_v} |J_{\text{eff}}, M_J\rangle \otimes |n_u, n_v\rangle c_{J_{\text{eff}} M_J, n_u n_v; \nu}. \quad (6)$$

Vibronic states

The distribution of the vibronic levels (E_ν) varies much with respect to g.



E_{JT}: Classical Jahn-Teller stabilization energy, $\hbar\omega/2 \cdot (g/2)^2$

Model Hamiltonian:

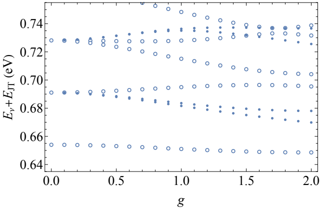
$$\hat{H} = \hat{H}_{\mathrm{JT}} + \hat{V}, \tag{7}$$

$$\hat{V} \propto \hat{\boldsymbol{p}}_{\mathrm{el}} \cdot \hat{\boldsymbol{A}}. \tag{8}$$

Cross-section (2nd order perturbation theory)

$$\frac{d^2\sigma}{d\Omega dk} \propto \left| \langle \nu_{\mathrm{f}}; \boldsymbol{k}_{\mathrm{f}} \lambda_{\mathrm{f}} | \hat{V} \hat{G} \hat{V} | \nu_{\mathrm{i}}; \boldsymbol{k}_{\mathrm{i}} \lambda_{\mathrm{i}} \rangle \right|^2 \delta\left(E_{\nu_{\mathrm{i}}} + \hbar\omega_{\mathrm{i}} - E_{\nu_{\mathrm{f}}} - \hbar\omega_{\mathrm{f}}\right). \tag{9}$$

$|\nu\rangle$, E_ν: Vibronic states



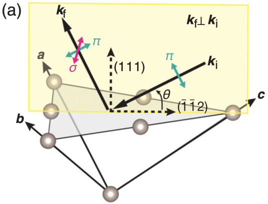
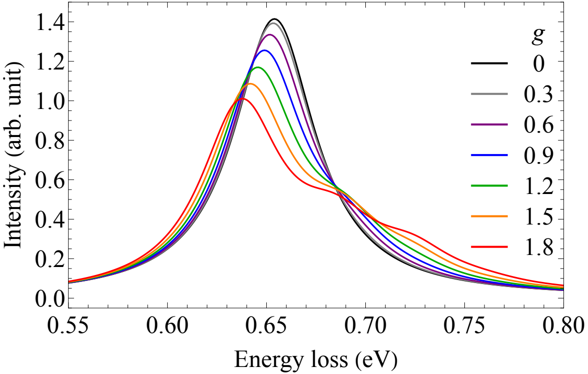


Fig. H. Takahashi *et al.*, Phys. Rev. Lett. **127**, 227201 (2021).



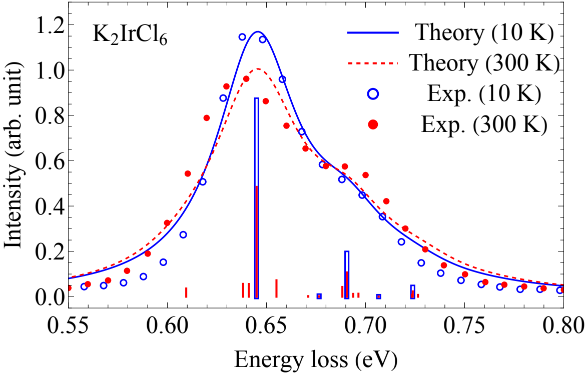
- $\lambda = 0.44$ eV (RIXS)
- $\omega = 33$ meV (Raman)
- $\Gamma = 50$ meV
- g: vary from 0.2 till 2 by 0.2

RIXS: D. Reig-i-Plessis *et al.*, Phys. Rev. Mater. **4**, 124407 (2020).

Raman: S. Lee *et al.*, Phys. Rev. B **105**, 184433 (2022).

As g increases, several peaks appear.

Comparison between the theoretical and experimental RIXS spectra



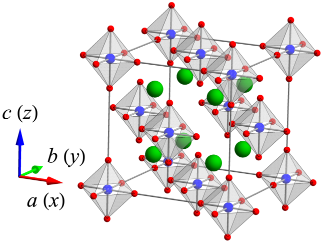
- $\lambda = 0.44$ eV (RIXS)
- $\omega = 33$ meV (Raman)
- $\Gamma = 50$ meV
- $g = 1.2$

RIXS: D. Reig-i-Plessis *et al.*, Phys. Rev. Mater. **4**, 124407 (2020).

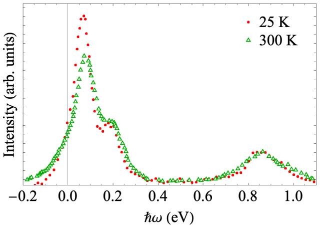
Raman: S. Lee *et al.*, Phys. Rev. B **105**, 184433 (2022).

Good agreement between the theoretical and the experimental RIXS spectra.

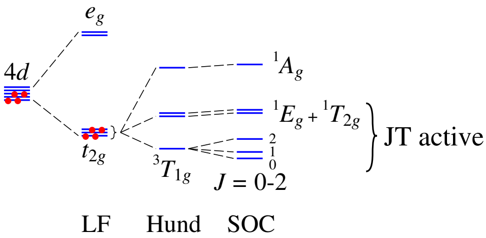
K_2RuCl_6: a candidate material of excitonic magnetism



Structure of K_2RuCl_6



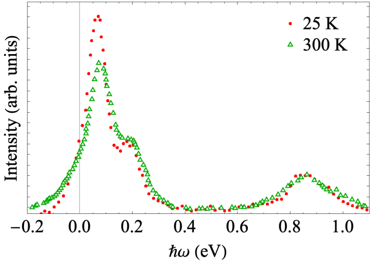
- Spin-orbit coupling on $4d^4$ site
  $\Rightarrow J = 0$ nonmagnetic ground state
- Exchange interaction between J multiplets could induce Excitonic magnetism.



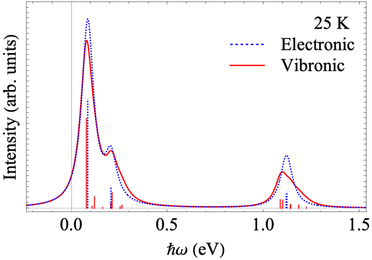
Theory: G. Khaliulline, Phys. Rev. Lett. **111**, 197201 (2013).

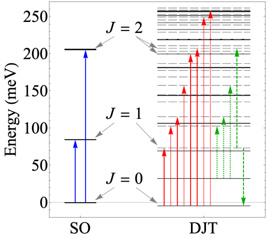
Exp.: H. Takahashi *et al.*, Phys. Rev. Lett. **127**, 227201 (2021).

Exp.



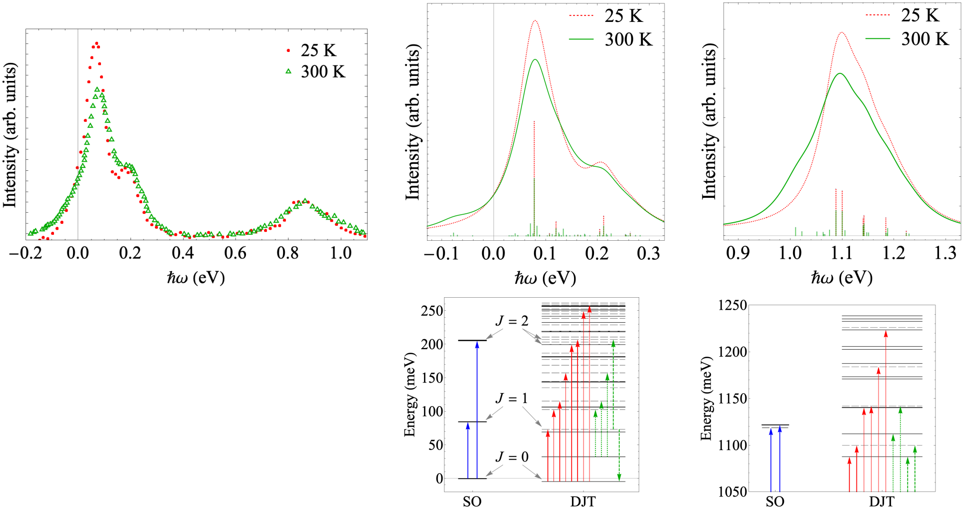
Theory





With the dynamic JT effect, the shape of the RIXS spectrum becomes closer to the exp. one.

Temperature evolution of the RIXS spectra (25 K and 300 K)



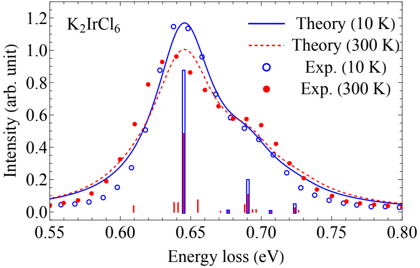
Our theoretical spectra capture the main features of the T-evolution of the exp. RIXS spectra

Conclusion

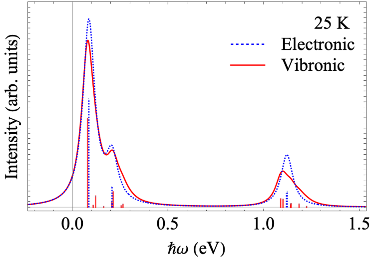
The fingerprints of the dynamic Jahn-Teller effect appear in RIXS spectra.

- Shape
- T-dependence

K_2IrCl_6



K_2RuCl_6



N. Iwahara and S. Shikano, Physical Review Research **5**, 023051, (2023).
N. Iwahara and W. Furukawa, arXiv:2305.05853