SPICE-Workshop: Novel Electronic and Magnetic Phases

Numerical approach for general superexchange interaction: Application for the spin-orbital excitation of pentavalent iridates

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Outline

- Superexchange interaction
  - Superexchange interaction for spin $\frac{1}{2}$, general J, and LS cases
  - Numerical way to calculate the general superexchange interaction
- Magnetism and magnetic excitation in Ba_2YIrO_6
  - Effective spin-orbital interaction in Ba_2YIrO_6
  - Spin-orbital excitation and its potential condensation
  - Electric band structure (charge excitation) of Ba_2YIrO_6

Collaborators

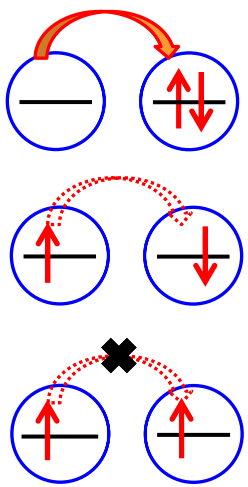
Dr. Dmitry V. Efremov and Prof. Jeroen van den Brink, IFW Dresden, Germany

Magnetic interaction in Hubbard model

Hubbard model

$$H = t \sum_{\langle i,j \rangle \sigma} \left(c_{j\sigma}^{\dagger} c_{i\sigma} + c_{i\sigma}^{\dagger} c_{j\sigma} \right) + U \sum_{i} n_{i\uparrow} n_{i\downarrow}$$

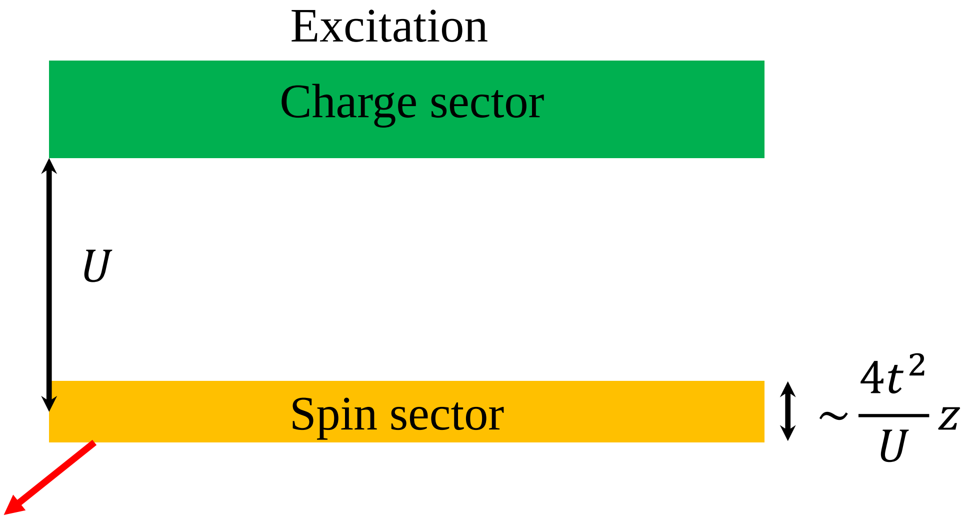
Strong coupling limit in half filled case $\quad U \gg t$



$\Delta E \sim U$

$\Delta E \sim \dfrac{t^2}{U}$

$\Delta E \sim 0$

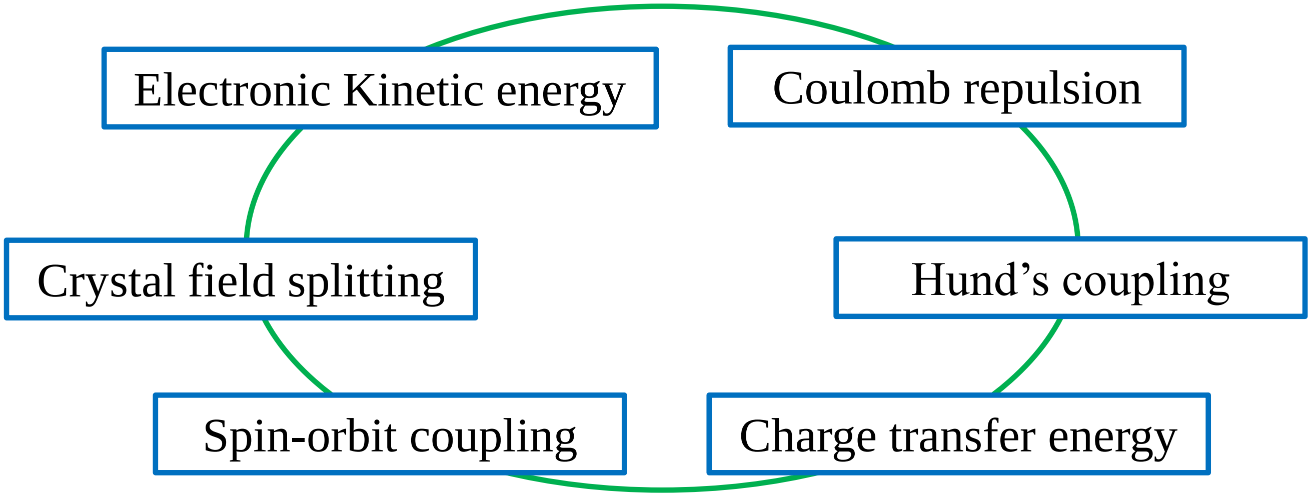


$$H_{SE} = J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j \quad \text{where} \quad J = \frac{4t^2}{U}$$

Superexchange interaction

Magnetism in *d*- and *f*-electron systems

Competing interactions

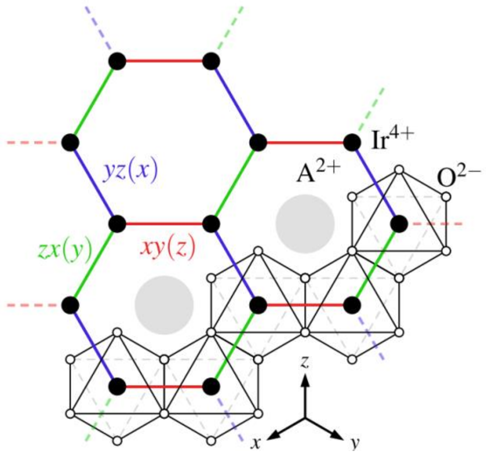


Magnetism is determined by spin and orbital, or spin-orbital entangled states

Beyond Heisenberg-type magnetic interaction

Beyond Heisenberg-type interaction

Na_2IrO_3, α-$RuCl_3$ $Ir4^+$, Ru^{3+} (t_{2g}^5)



J. G. Rau *et al.*, Phys. Rev. Lett. **112**, 077204 (2014)

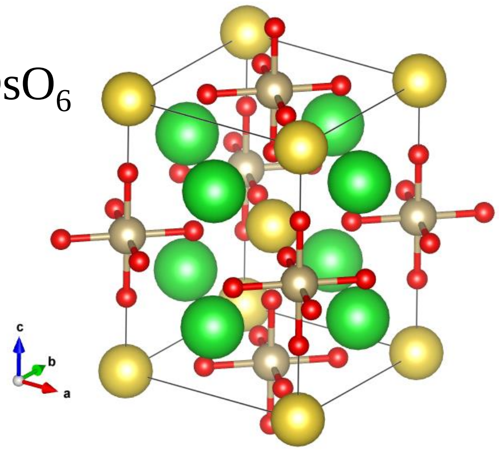
$$H = \sum_{\gamma \langle i,j \rangle_\gamma} K S_{i\gamma} S_{j\gamma} + \sum_{\gamma \langle i,j \rangle_\gamma} J \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$

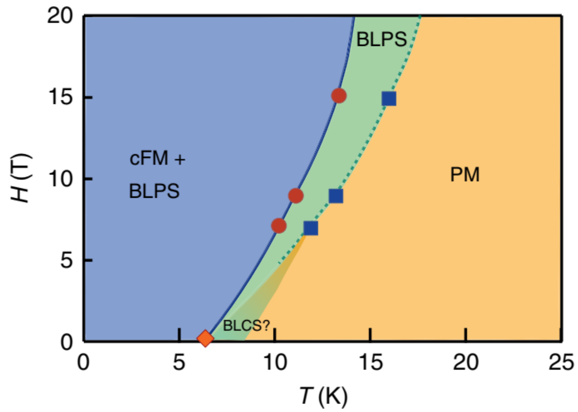
$$+ \sum_{\gamma \langle i,j \rangle_\gamma} \Gamma \left(S_{i\alpha} S_{j\beta} + S_{i\beta} S_{j\alpha} \right)$$

$$+ \sum_{\gamma \langle i,j \rangle_\gamma} \Gamma' \left(S_{i\alpha} S_{j\gamma} + S_{i\gamma} S_{j\alpha} + S_{i\beta} S_{j\gamma} + S_{i\gamma} S_{j\beta} \right)$$

Multipolar interaction in d^1 double perovskite

Ba_2NaOsO_6





L. Lu *et al.*, Nat. Commun. **8**, 14407 (2017)

Calculation of superexchang interaction

Hubbard model

$$H_0^{ij} = U\left(n_{i\uparrow}n_{i\downarrow} + n_{j\uparrow}n_{j\downarrow}\right) \qquad V_{ij} = t\left(c_{j\sigma}^{\dagger}c_{i\sigma} + c_{i\sigma}^{\dagger}c_{j\sigma}\right)$$

Effective Hamiltonian of spin sector with 2nd order perturbation

$$H_{\text{eff}}^{ij} = \sum_{\sigma_i\sigma_j\sigma_i'\sigma_j'}\sum_{n\notin G}\left[\frac{\langle\sigma_i\sigma_j|V_{ij}|\Psi_n^0\rangle\langle\Psi_n^0|V_{ij}|\sigma_i'\sigma_j'\rangle}{E_g^0 - E_n^0}\right]|\sigma_i\sigma_j\rangle\langle\sigma_i'\sigma_j'| \qquad \sigma,\sigma' \in \{\uparrow,\downarrow\}$$

$$= -\frac{t^2}{U}\left(|\uparrow\downarrow\rangle\langle\uparrow\downarrow| - |\uparrow\downarrow\rangle\langle\downarrow\uparrow| - |\downarrow\uparrow\rangle\langle\uparrow\downarrow| + |\downarrow\uparrow\rangle\langle\downarrow\uparrow|\right)$$

Relation between spin operators and $|\sigma\rangle\langle\sigma'|$ projection operators

$$|\uparrow\rangle\langle\uparrow| = \frac{1}{2}\mathbf{I} + S_z \qquad |\downarrow\rangle\langle\downarrow| = \frac{1}{2}\mathbf{I} + S_z \qquad |\uparrow\rangle\langle\downarrow| = S_x + iS_y \qquad |\downarrow\rangle\langle\uparrow| = S_x - iS_y$$

$$\Rightarrow \quad H_{\text{eff}}^{ij} = \frac{4t^2}{U}\left(-\frac{1}{4}\mathbf{I} + \mathbf{S}_i \cdot \mathbf{S}_j\right)$$

General superexchange interaction

J representation

$$|\sigma_i \sigma_j\rangle \longrightarrow |J m_i J m_j\rangle$$

$$m_i, m_j \in \{-J, \dots, J\}$$

LS representation

$$|\sigma_i \sigma_j\rangle \longrightarrow |m_i s_i m_j s_j\rangle$$

$$m_i, m_j \in \{-L, \dots, L\} \qquad s_i, s_j \in \{-S, \dots, S\}$$

Effective Hamiltonian of spin-orbital sector

$$H_{\text{eff}}^{ij} = \sum h_{m_i' m_j'}^{m_i m_j} |J m_i J m_j\rangle\langle J m_i' J m_j'|$$

$$H_{\text{eff}}^{ij} = \sum h_{m_i' s_i' m_j' s_j'}^{m_i s_i m_j s_j} |m_i s_i, m_j s_j\rangle\langle m_i' s_i', m_j' s_j'|$$

Relation between irreducible tensor operators and projection operalator

$$\hat{T}_q^k(\hat{J}) = \sum_{mm'} T_0^k(J) \frac{C_{Jm'kq}^{Jm}}{C_{JJk0}^{JJ}} |Jm\rangle\langle Jm|$$

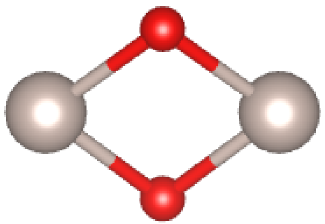
$$|Jm\rangle\langle Jm| = \sum_{kq} \frac{2k+1}{2J+1} \frac{C_{JJk0}^{JJ} C_{Jm'kq}^{Jm}}{T_0^k(J)} \hat{T}_q^k(\hat{J})$$

$$H_{\text{eff}}^{ij} = \sum \Lambda_{q_i q_j}^{k_i k_j} \hat{T}_{q_i}^{k_i}(\hat{J}_i) \hat{T}_{q_j}^{k_j}(\hat{J}_j)$$

$$H_{\text{eff}}^{ij} = \sum \Lambda_{q_i q_j s_i s_j}^{k_i k_j r_i r_j} \hat{T}_{q_i}^{k_i}(\hat{L}_i) \hat{T}_{q_j}^{k_j}(\hat{L}_j) \hat{T}_{s_i}^{r_i}(\hat{S}_i) \hat{T}_{s_j}^{r_j}(S_j)$$

Difficulty in calculation

Effect of accurate ligand states



Competitive physical interactions

Jahn-Teller effect

Spin-orbit coupling ⟷ Hund's coupling

Perturbation method is not easy to calculation magnetic interactions

Directly extracted from exact eigenvalues and states in two-site cluster

$$H_{\text{eff}} = \sum E_n \frac{P_{LS}|\Psi_n\rangle\langle\Psi_n|P_{LS}}{\langle\Psi_n|P_{LS}|\Psi_n\rangle} \quad \text{where} \quad P_{LS} = \sum \left|m_i s_i m_j s_j\right\rangle\left\langle m_i' s_i' m_j' s_j'\right|$$

Numerical method for general superexchange interaction

Constructing the multiband Hubbard model of two-site cluster

Finding local spin (S) and orbital (L) states : $|m_i s_i\rangle$

Finding lowest $n_{LS} = (2L+1)^2(2S+1)^2$ eigenvalues and states (E_n, Ψ_n)

Constructing projection operators $P_{LS} = \sum \left|m_i s_i m_j s_j\right\rangle\left\langle m_i' s_i' m_j' s_j'\right|$

Checking orthogonality of projected spin-orbital states $S_{nn'} = \langle\Psi_n|P_{LS}|\Psi_{n'}\rangle$

Extracting effective spin-orbital Hamiltonian in terms of projection operators

$$H_{\text{eff}}^{ij} = \sum h_{m_i' s_i' m_j' s_j'}^{m_i s_i m_j s_j} \left|m_i s_i, m_j s_j\right\rangle\left\langle m_i' s_i', m_j' s_j'\right|$$

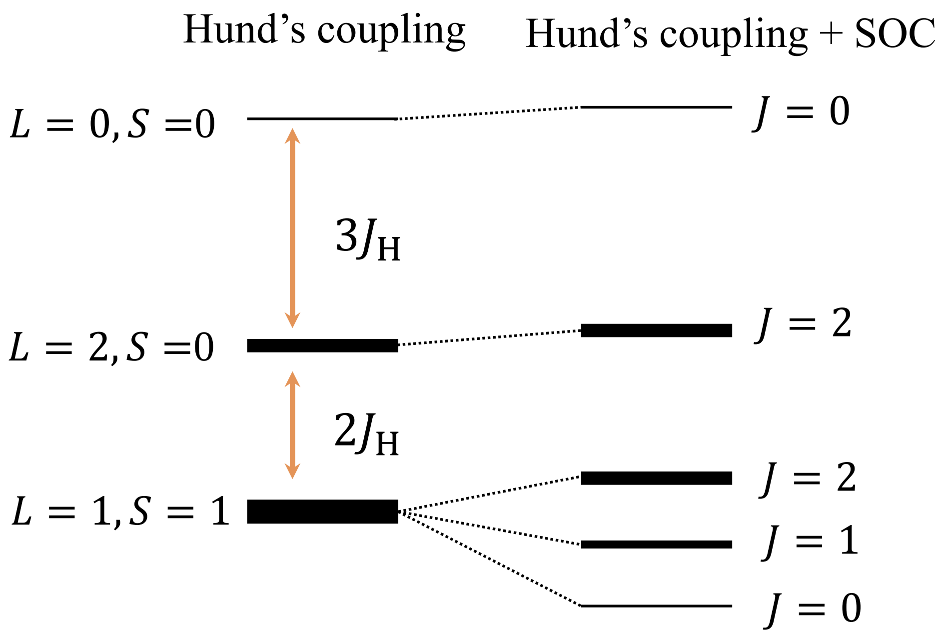
$$h_{m_i' s_i' m_j' s_j'}^{m_i s_i m_j s_j} = \sum_{nn'k} E_k \left[S^{-\frac{1}{2}}\right]_{nk} \left[S^{-\frac{1}{2}}\right]_{kn'} \left\langle m_i s_i, m_j s_j|\Psi_n\right\rangle\left\langle\Psi_{n'}|m_i' s_i', m_j' s_j'\right\rangle$$

Interaction with spin-orbital operators

$$H_{\text{eff}}^{ij} = C\mathbf{I} + H_i + H_j + H_{SE}^{ij} = \sum \Lambda_{q_i q_j s_i s_j}^{k_i k_j r_i r_j} \hat{T}_{q_i}^{k_i}(\hat{L}_i)\hat{T}_{q_j}^{k_j}(\hat{L}_j)\hat{T}_{s_i}^{r_i}(\hat{S}_i)\hat{T}_{s_j}^{r_j}(\hat{S}_j)$$

Van Vleck Paramagnetism of $4d^4/5d^4$

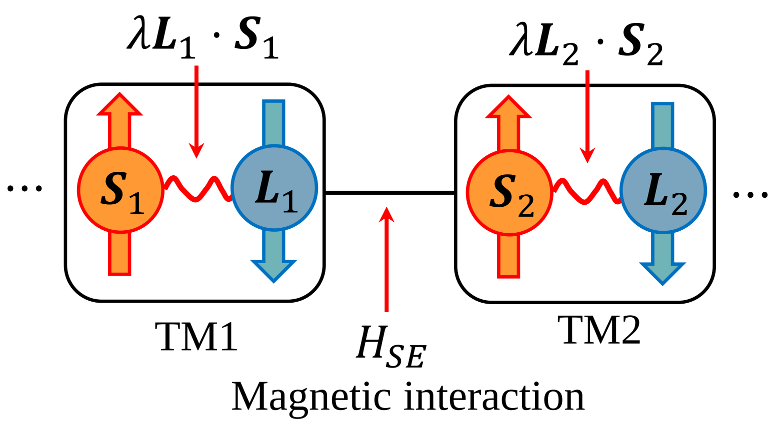
Ru^{4+} , Os^{4+}, Rh^{5+} and Ir^{5+} : d^4 ,two holes ($l_{eff} = 1, s = \frac{1}{2}$) of t_{2g} orbitals

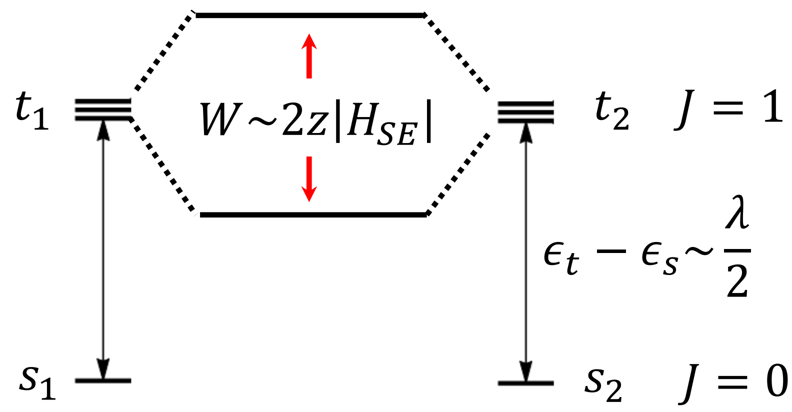


In strong SOC limit, singlet (Van Vleck-type nonmagnetic) ground state is stabilized

Excitonic magnetism in d^4

Magnetically interacting t_{2g}^4 systems





Similarity to the "quantum dimer model"

Triplon excitation (transition from $J = 0$ to $J = 1$) can have dispersion

$$T_i^\dagger = t_i^\dagger s_i$$

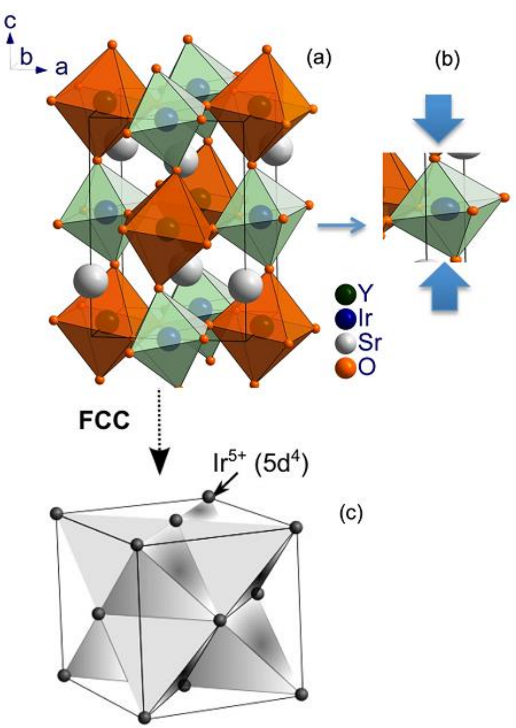
When $W > \epsilon_t - \epsilon_s$, Bose-Einstein condensation of triplons can occur

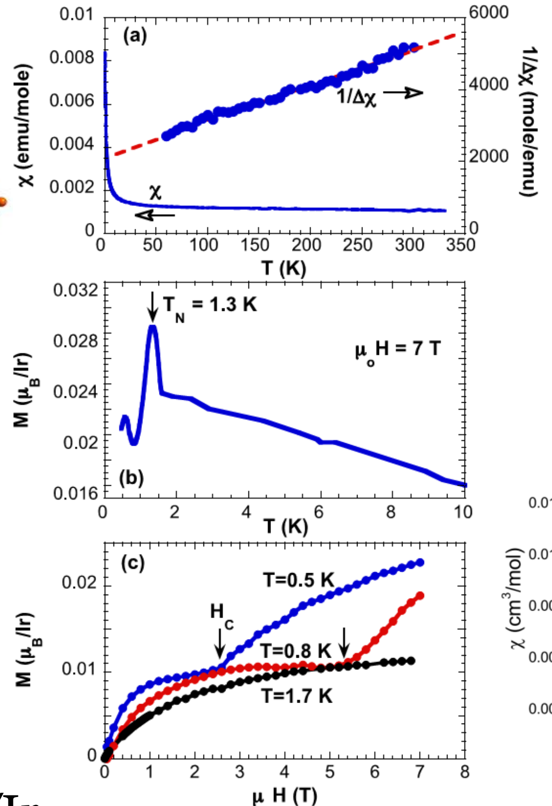
Triplon condensation leads to the magnetism (Exitonic magnetism)

G. Khaliullin Phys. Rev. Lett. 111, 197201 (2013)

Magnetism of pentavalent iridates

Sr_2YIrO_6 : distorted perovskite

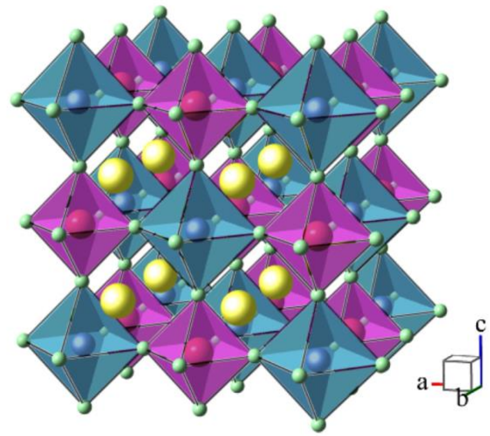


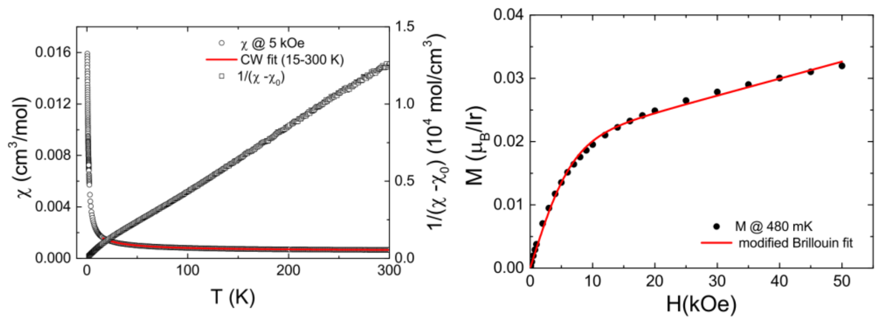


~0.91 μ_B/Ir

G. Cao *et. al*., Phys. Rev. Lett. **112**, 056402 (2014)

Ba_2YIrO_6 : cubic perovskite

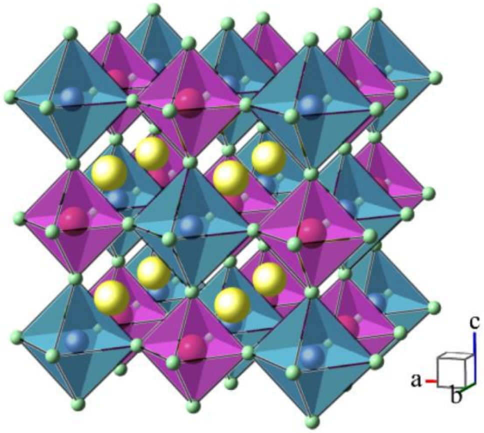




~0.44 μ_B/Ir

T. Dey *et. al*., Phys. Rev. B **93**, 014434 (2016)

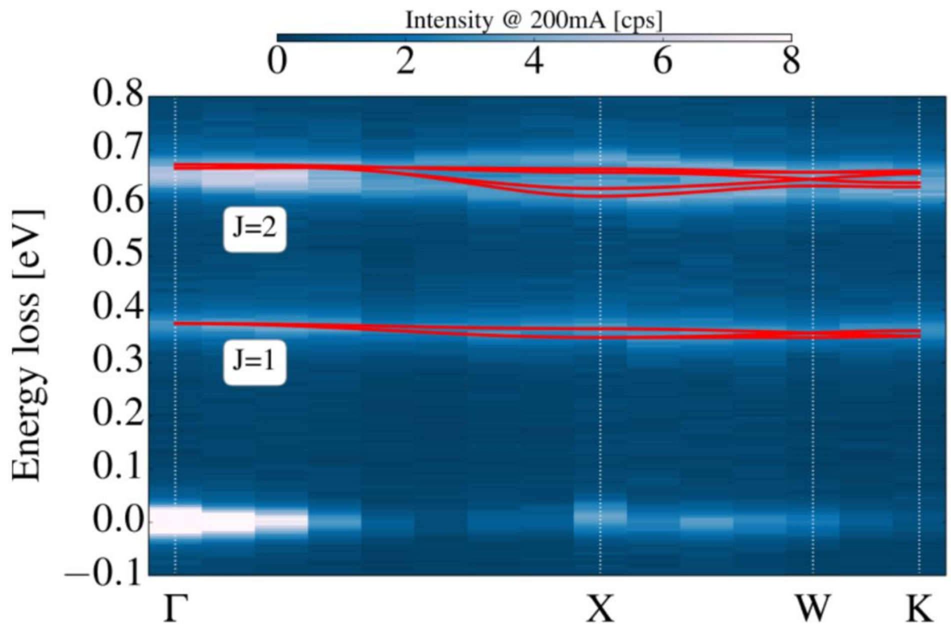
Spin-orbital excitations in Ba_2YIrO_6



Excitation energies

t_{2g}^4 terms	MRCI + SOC	Model	RIXS
$^3T_{1g}$	0.00	0.00 ($J = 0$)	0.00
	0.37	0.36 ($J = 1$)	0.371 ± 0.003
	0.70	0.66 ($J = 2$)	0.651 ± 0.003
$^1T_{2g}, {}^1E_g$	1.71, 1.79	1.50 ($J = 2$)	≃1.2
$^1A_{1g}$	3.09	2.72 ($J = 0$)	

Resonant inelastic x-ray scattering (RIXS) spectra



Dispersion is less than 50 meV

Triplon excitation hardly condenses

Kusch *et al.*, Phys. Rev. B 97, 064421 (2018)

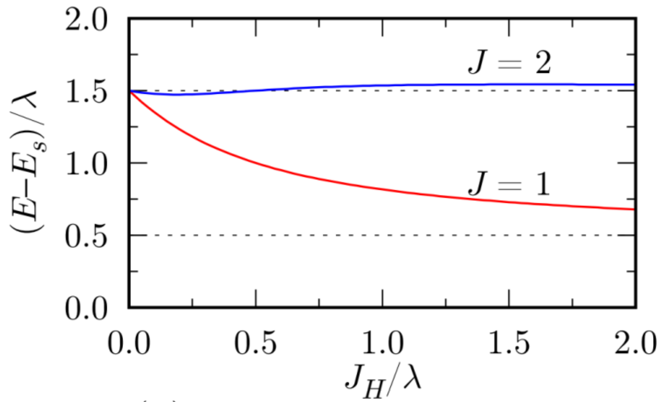
Model Hamiltonian for two-site cluster

Local multiplet energies of t_{2g}^4

Coulomb interaction, Hund's coupling, and spin-orbit coupling

$$H_{\text{ion}} = \lambda \sum_{\mu\nu\sigma\sigma'} (\boldsymbol{s}\cdot\boldsymbol{l})_{\mu\sigma,\nu\sigma'} c^{\dagger}_{\mu\sigma} c_{\nu\sigma'} + \frac{1}{2}\sum_{\mu\nu\sigma\sigma'} U_{\mu\nu} c^{\dagger}_{\mu\sigma} c^{\dagger}_{\nu\sigma'} c_{\nu\sigma'} c_{\mu\sigma}$$
$$+ \frac{1}{2}\sum_{\mu\neq\nu,\sigma\sigma'} J_{\mu\nu} c^{\dagger}_{\mu\sigma} c^{\dagger}_{\nu\sigma'} c_{\mu\sigma'} c_{\nu\sigma} + \frac{1}{2}\sum_{\mu\neq\nu,\sigma} J'_{\mu\nu} c^{\dagger}_{\mu\sigma} c^{\dagger}_{\mu,-\sigma} c_{\nu,-\sigma} c_{\nu\sigma}$$

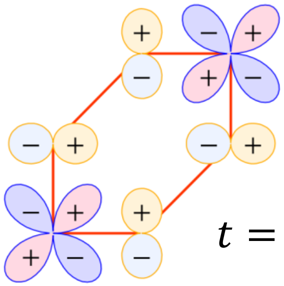
$$U_{\mu\mu} = U, U_{\mu\neq\nu} = U - 2J_H, J_{\mu\nu} = J'_{\mu\nu} = J_H$$



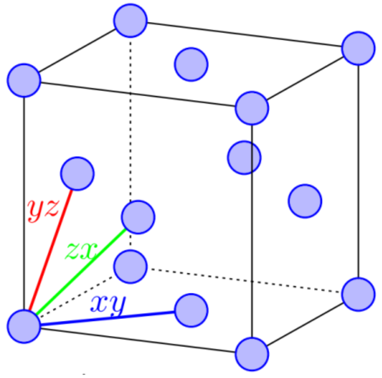
$J_H = 0.4\,\lambda = 0.5$ eV

Hopping

Dominant hopping process



$t = h_{xy,xy}$ in *xy*-plane



Effective spin-orbital Hamiltonian

Projection operator

$$P_J = \sum \left|J_i m_i J_j m_j\right\rangle\left\langle J_i' m_i' J_j' m_j'\right| \qquad J_i, J_i', J_j, J_j' \in \{0,1,2\}$$

Effective Hamiltonian

$$H_{\mathrm{eff}}^{ij} = \sum h_{J_i' m_i' J_j' m_j'}^{J_i m_i J_j m_j} \left|J_i m_i J_j m_j\right\rangle\left\langle J_i' m_i' J_j' m_j'\right|$$

Hard-core boson representation

Singlet boson ($J = 0$)

$$s^\dagger|vac\rangle = |J = 0,0\rangle$$

Triplet bosons ($J = 1$)

$$t_x^\dagger|vac\rangle = -\frac{1}{\sqrt{2}}(|J = 1,-1\rangle - |J = 1,1\rangle)$$

$$t_y^\dagger|vac\rangle = -\frac{i}{\sqrt{2}}(|J = 1,-1\rangle + |J = 1,1\rangle)$$

$$t_z^\dagger|vac\rangle = |J = 1,0\rangle$$

Quintet bosons ($J = 2$)

$$q_{xy}^\dagger|vac\rangle = \frac{i}{\sqrt{2}}(|J = 2,-2\rangle - |J = 2,2\rangle)$$

$$q_{yz}^\dagger|vac\rangle = \frac{i}{\sqrt{2}}(|J = 2,-2\rangle + |J = 2,1\rangle)$$

$$q_{z^2}^\dagger|vac\rangle = |J = 2,0\rangle$$

$$q_{zx}^\dagger|vac\rangle = \frac{1}{\sqrt{2}}(|J = 2,-2\rangle - |J = 2,1\rangle)$$

$$q_{x^2-y^2}^\dagger|vac\rangle = \frac{1}{\sqrt{2}}(|J = 2,-2\rangle + |J = 2,2\rangle)$$

Effective Hamiltonian of singlet, triplet, and quintet bosons

$$H_{\rm eff} = \sum_i \epsilon_s s_i^\dagger s_i + \sum_{i\alpha} \epsilon_t t_{\alpha i}^\dagger t_{\alpha i} + \sum_{i\mu} \epsilon_q q_{\mu i}^\dagger q_{\mu i}$$
$$+ \frac{1}{2}\sum_{i\delta\alpha\beta}\left([h_\delta^{11}]_{\alpha\beta} s_i^\dagger t_{\alpha i_\delta}^\dagger t_{\beta i} s_{i_\delta} + [d_\delta^{11}]_{\alpha\beta} t_{\beta i}^\dagger t_{\alpha i_\delta}^\dagger s_i s_{i_\delta} + h.c.\right)$$
$$+ \frac{1}{2}\sum_{i\delta\mu\nu}\left([h_\delta^{22}]_{\mu\nu} s_i^\dagger q_{\mu i_\delta}^\dagger q_{\nu i} s_{i_\delta} + [d_\delta^{22}]_{\mu\nu} q_{\nu i}^\dagger q_{\mu i_\delta}^\dagger s_i s_{i_\delta} + h.c.\right)$$
$$+ \frac{1}{2}\sum_{i\delta\alpha\mu}\left([h_\delta^{12}]_{\alpha\mu} s_i^\dagger t_{\alpha i_\delta}^\dagger q_{\mu i} s_{i_\delta} + [d_\delta^{12}]_{\alpha\mu} q_{\mu i}^\dagger t_{\alpha i_\delta}^\dagger s_i s_{i_\delta} + h.c.\right)$$
$$+ \frac{1}{2}\sum_{i\delta\mu\alpha}\left([h_\delta^{21}]_{\mu\alpha} s_i^\dagger q_{\mu i_\delta}^\dagger t_{\alpha i} s_{i_\delta} + [d_\delta^{21}]_{\mu\alpha} t_{\alpha i}^\dagger q_{\mu i_\delta}^\dagger s_i s_{i_\delta} + h.c.\right)$$
$$+ \cdots ,$$

When singlet bosons almost condense, $s_i = s_i^\dagger = s$, and $s^2 = 1$ are assumed

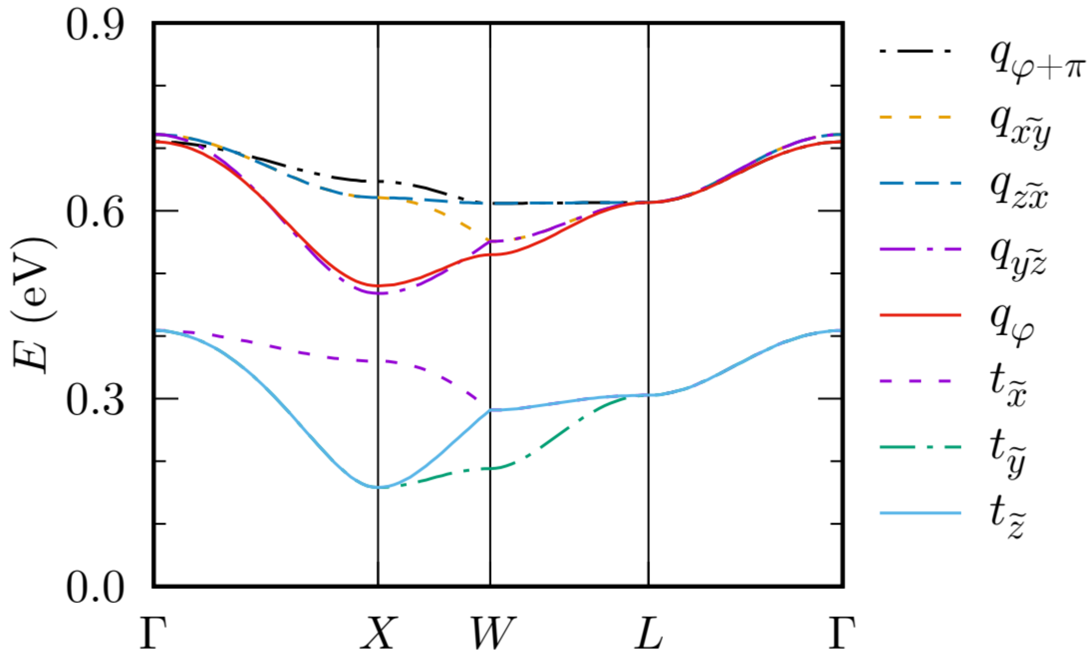
$$\boldsymbol{\psi}_i^\dagger = \left(t_{x,i}^\dagger \;\; t_{y,i}^\dagger \;\; t_{z,i}^\dagger \;\; q_{xy,i}^\dagger \;\; q_{yz,i}^\dagger \;\; q_{zx,i}^\dagger \;\; q_{z^2,i}^\dagger \;\; q_{x^2-y^2,i}^\dagger\right).$$

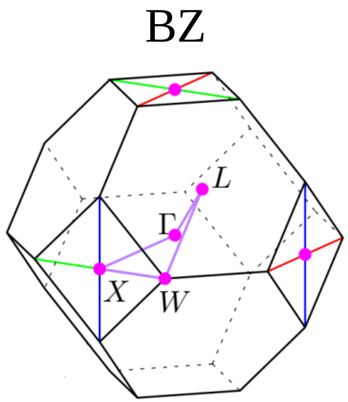
$$H_{\rm MF} \approx N\epsilon_s + \sum \left[\boldsymbol{\psi}_{\mathbf{k}}^\dagger \mathbf{h}(\mathbf{k}) \boldsymbol{\psi}_{\mathbf{k}} + \frac{1}{2}(\boldsymbol{\psi}_{\mathbf{k}}^\dagger \mathbf{d}(\mathbf{k}) \boldsymbol{\psi}_{-\mathbf{k}}^* + \text{H.c.})\right]$$

$$\mathbf{h}(\mathbf{k}) = \begin{pmatrix} \epsilon_t - \epsilon_s & 0 \\ 0 & \epsilon_q - \epsilon_s \end{pmatrix} + \sum_\delta \begin{pmatrix} \mathbf{h}_\delta^{11} & \mathbf{h}_\delta^{12} \\ \mathbf{h}_\delta^{21} & \mathbf{h}_\delta^{22} \end{pmatrix} e^{i\mathbf{k}\cdot\mathbf{r}_\delta} \qquad \mathbf{d}(\mathbf{k}) = \sum_\delta \begin{pmatrix} \mathbf{d}_\delta^{11} & \mathbf{d}_\delta^{12} \\ \mathbf{d}_\delta^{21} & \mathbf{d}_\delta^{22} \end{pmatrix} e^{i\mathbf{k}\cdot\mathbf{r}_\delta}$$

Spin-orbital excitation

$U = 4, J_H = 0.5, \lambda = 0.4, t = -0.4$ eV

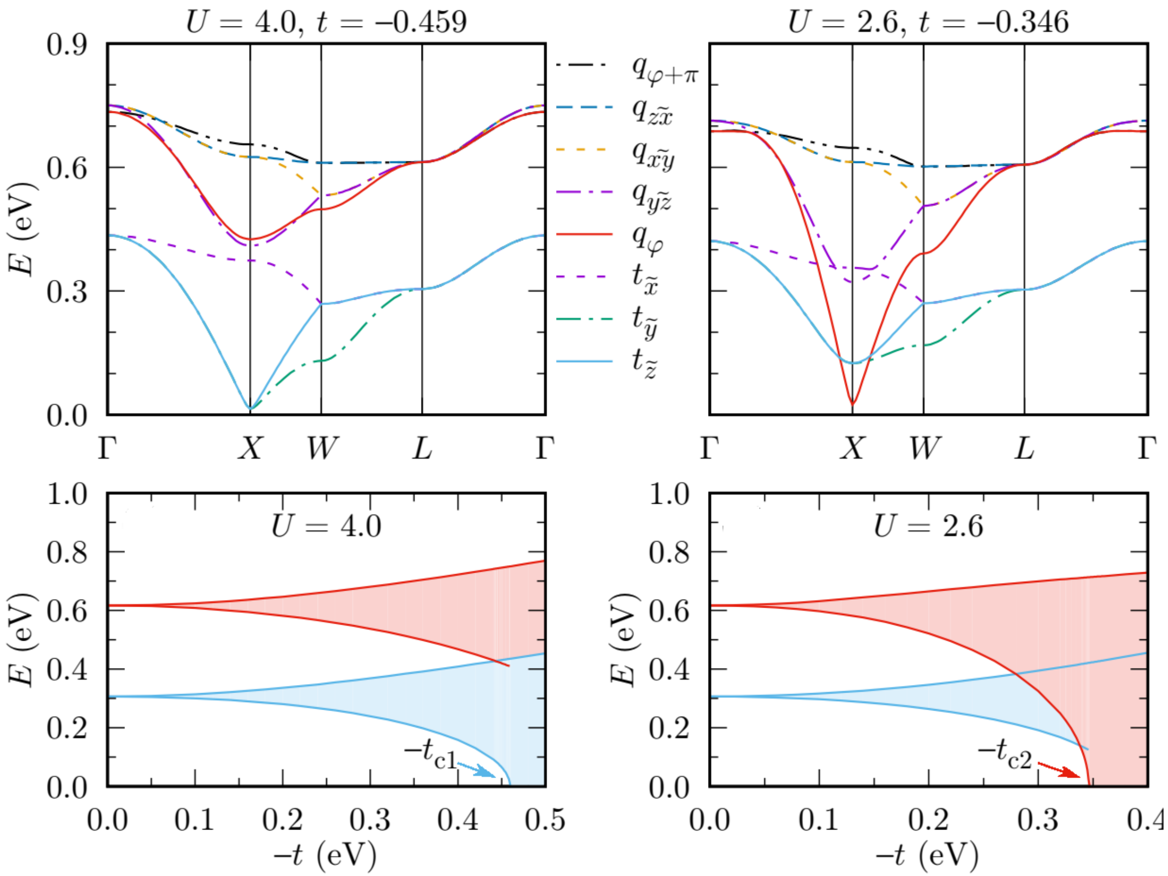




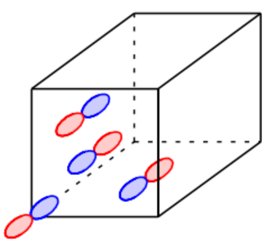
Minimum excitation at X (Y, and Z) point

Condensation of triplet and quintet excitons

$J_H = 0.5, \lambda = 0.4$



Dipole order

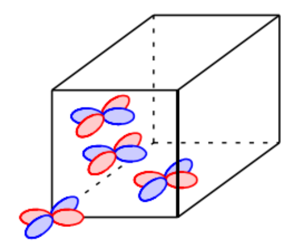


X point

$t_{\tilde{y}}$ and $t_{\tilde{z}}$ condense at X point

$q_{y^2-z^2}$ condense at X point

Quadrupole order

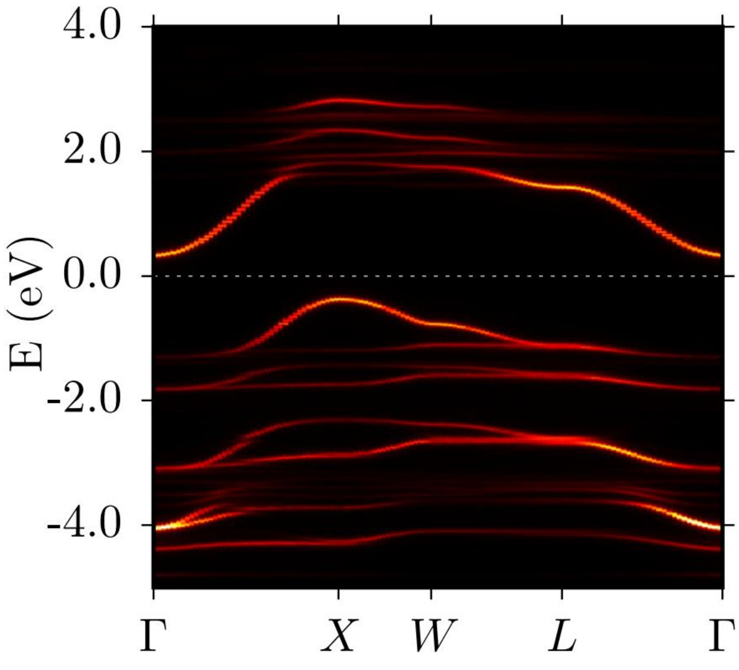


Z point

Electronic structure (Charge excitation)

Variational cluster perturbation theory with four-site cluster

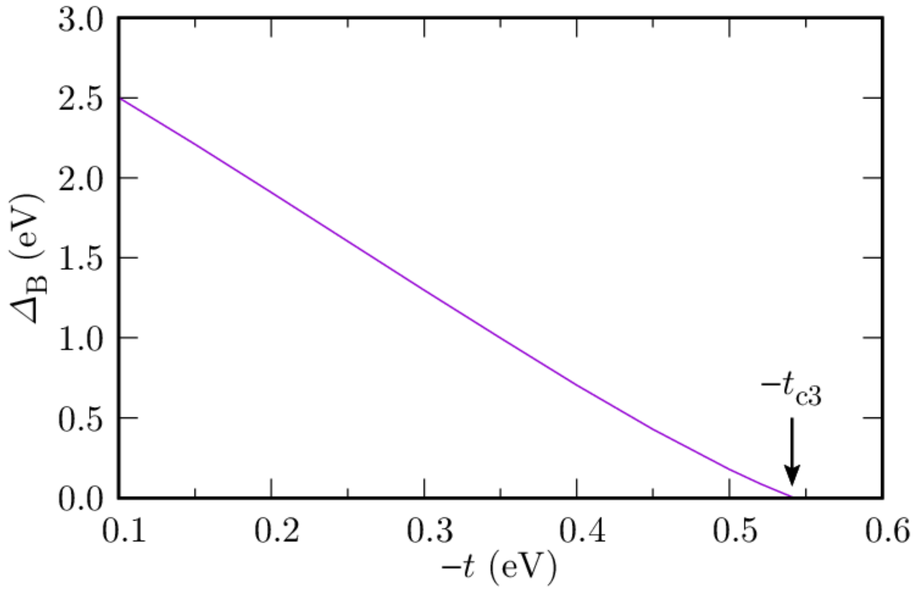
Spectral functions



Indirect gap (Γ & X)

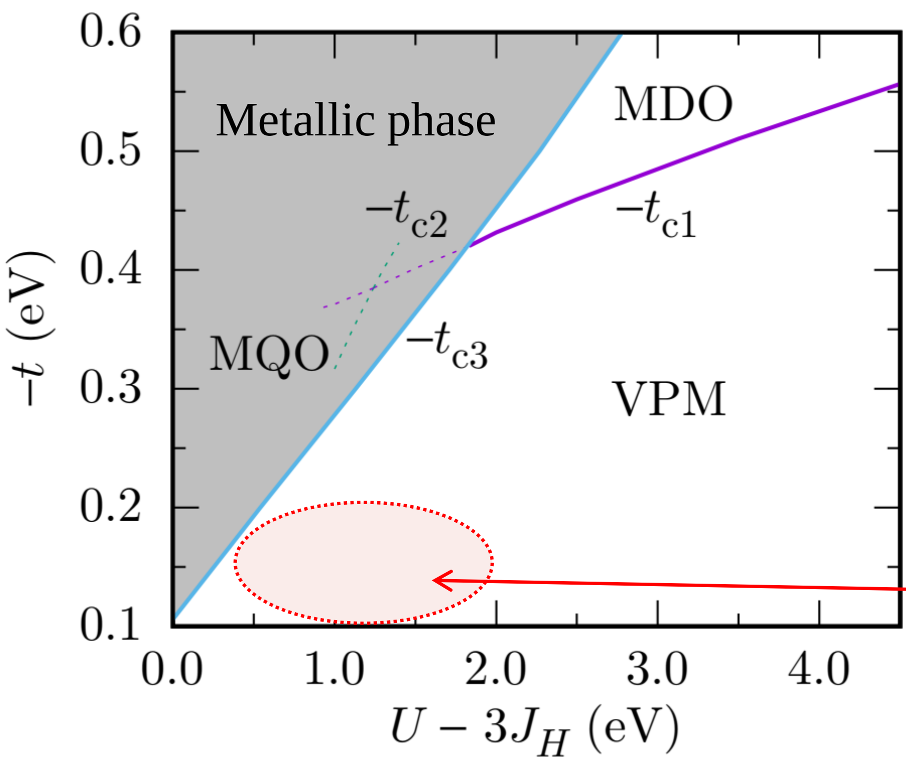
$U = 4, J_H = 0.5, \lambda = 0.4, t = -0.4$ eV

Electronic gap



Insulator-metal transition occurs at t_{c3}

Phase diagram



VPM : Van Vleck Paramagnetism

MDO : Magnetic dipole order

MQO : Magnetic quadrupole order

Double perovskite iridates

$|t| \approx 0.1 \sim 0.2$ eV

$U-3J_H \approx 0.4 - 2.0$ eV

Condensation of triplon or quinton could hardly occur in double perovskite iridates

Summary

- We introduce numerical way to calculate general sperexchange interactions (multipolar interactions) based on two-site cluster calculation

- We derive the effective spin-orbital Hamiltonian of double-perovskite pentavalent iridates (Ba_2YIrO_6)

- According to the mean-field approximation of spin-orbital Hamiltonian, triplet bosons (magnetic dipole) and quintet bosons (magnetic quadrupole) can condense in strong and moderate *U* limit

- We also check the electronic band structure for charge excitation gap

- In relevant *U* value for iridates, neither triplet nor quintet bosons condenses even if some physical parameters are tuned

Thank you for your attention