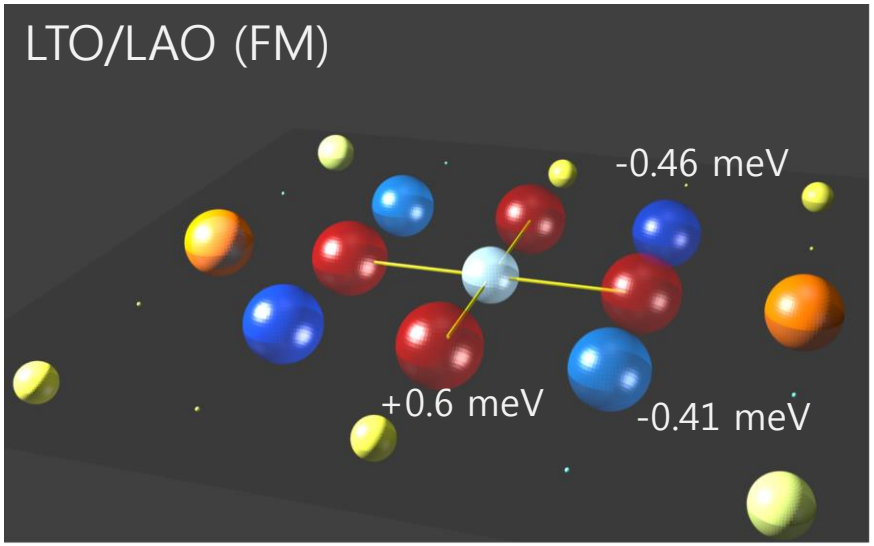
**OpenMX Developer's Meeting**
**(Nobemver 24th 2016, KAIST, Daejeon)**

# Calculating Branching Ratio and Heisenberg Exchange Constant with OpenMX

Myung Joon Han (KAIST, Physics)



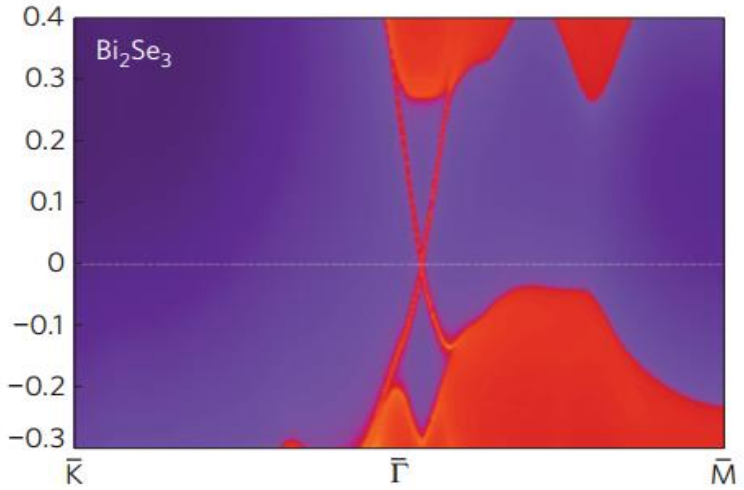

# SOC and Branching Ratio

$$H_{SO} = \lambda \langle L \cdot S \rangle$$

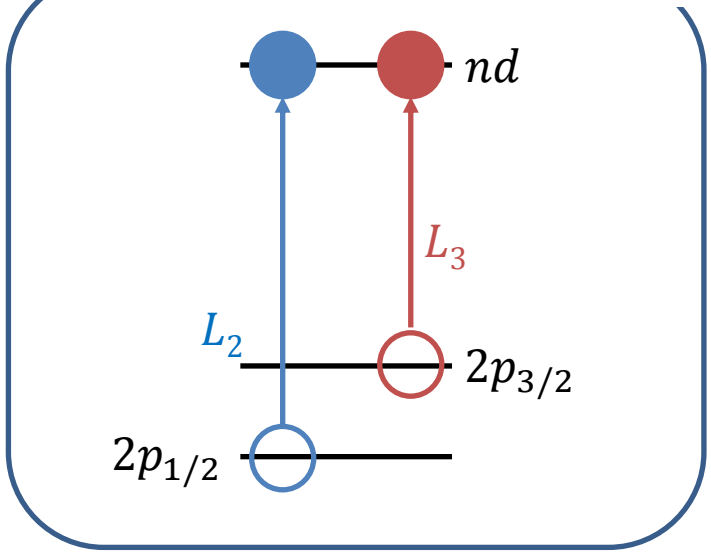
$$\bar{H}_{SO} = -\frac{e}{m^2c^2}\left(\frac{1}{r}\frac{d\phi}{dr}\right)(\mathbf{l}\cdot\mathbf{s})$$

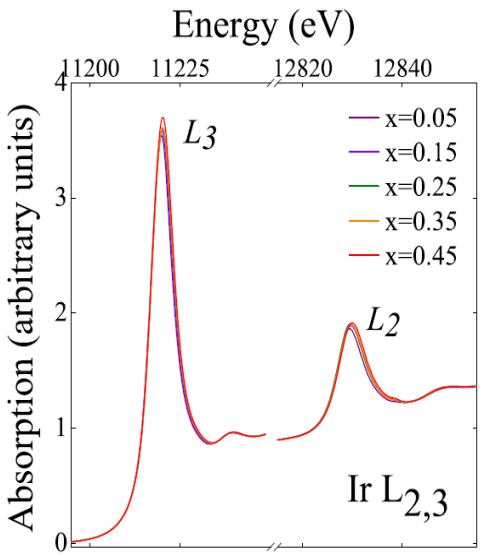
$$H_{SO} = \frac{-e}{2m^2c^2}\left[(\nabla\phi \times \mathbf{p})\cdot\mathbf{s}\right]$$

Zhang *et al.*, Nat. Phys. **5**, 438 (2009)




**X-ray absorption spectra (XAS)**




Chikara, *MJH et al.*
PRB-RC (2015)

$$I_{L_3}/I_{L_2}$$

# A Simple Way from First-Principles

$$\frac{L_j}{L_3+L_2} = \frac{2j+1}{2(2l_c+1)} \pm A(l_c, l_v, n_h)\,\langle \mathbf{L}\cdot\mathbf{S}\rangle \qquad \text{where } A = -\frac{1}{3n_h}\,.$$

$$I_{L_3}/I_{L_2} = \frac{2n_h - \langle \mathbf{L}\cdot\mathbf{S}\rangle}{n_h + \langle \mathbf{L}\cdot\mathbf{S}\rangle} = \frac{2-r}{1+r}$$

Thole and van der Laan,
PRL (1988), PRA (1988)

J.-H. Sim, H. Yoon, S. H. Park, *MJH*,
Phys. Rev. B (2016)

**For the case of iridates as an example:**

**( $l_c = p = 1$, $l_v = d = 2$ )**

$$|\psi_{n\mathbf{k}}\rangle = \sum_{m_J=-5/2}^{5/2} a_{m_J}^{n\mathbf{k}}\,|J=5/2, m_J\rangle_{\mathrm{Ir}} + \sum_{m_J=-3/2}^{3/2} b_{m_J}^{n\mathbf{k}}\,|J=3/2, m_J\rangle_{\mathrm{Ir}} + \sum_{(i,\alpha)\neq(\mathrm{Ir},d)} c_{\alpha,i}^{n\mathbf{k}}\,|\phi_{\alpha,i}\rangle$$

$$\langle \mathbf{L}\cdot\mathbf{S}\rangle = \sum_{n\mathbf{k}}^{\mathrm{occ}} \langle\psi_{n\mathbf{k}}|\,\mathbf{L}\cdot\mathbf{S}\,|\psi_{n\mathbf{k}}\rangle$$

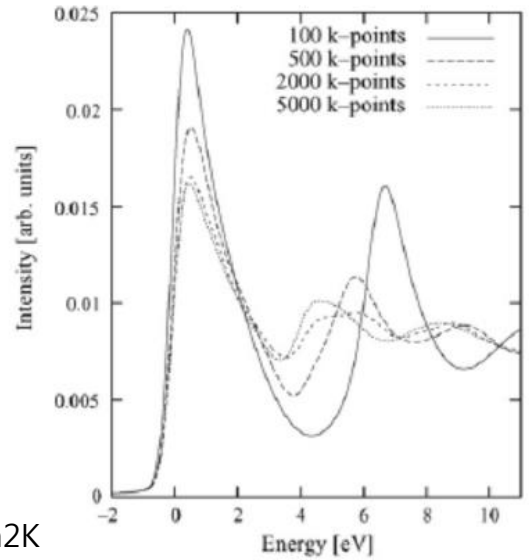
$$= \sum_{\epsilon_{n\mathbf{k}}<\epsilon_F} \sum_{m_J} (1.0\times|a_{m_J}^{n\mathbf{k}}|^2 - 1.5\times|b_{m_J}^{n\mathbf{k}}|^2)$$

# Advantages

## Direct simulation of XAS spectra

**(e.g., Wien2K and QuantumEspresso):**

- ✓ **How to simulate the core hole?**
- ✓ **Preparing special pseudopotentials**
- ✓ **Supercell, basis set, and other numerics issues**

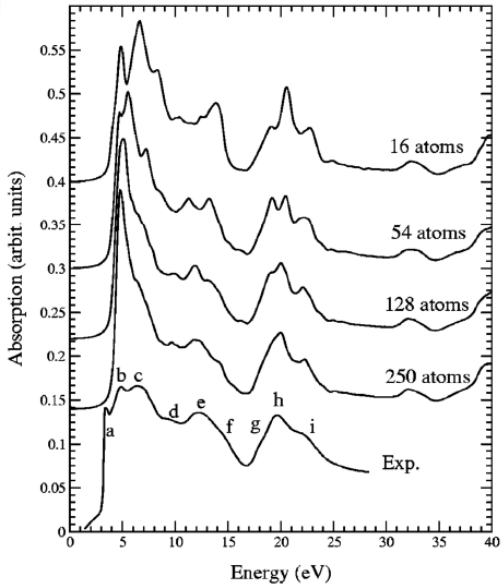



Jorissen's Wien2K Lecture Note

## Our approach

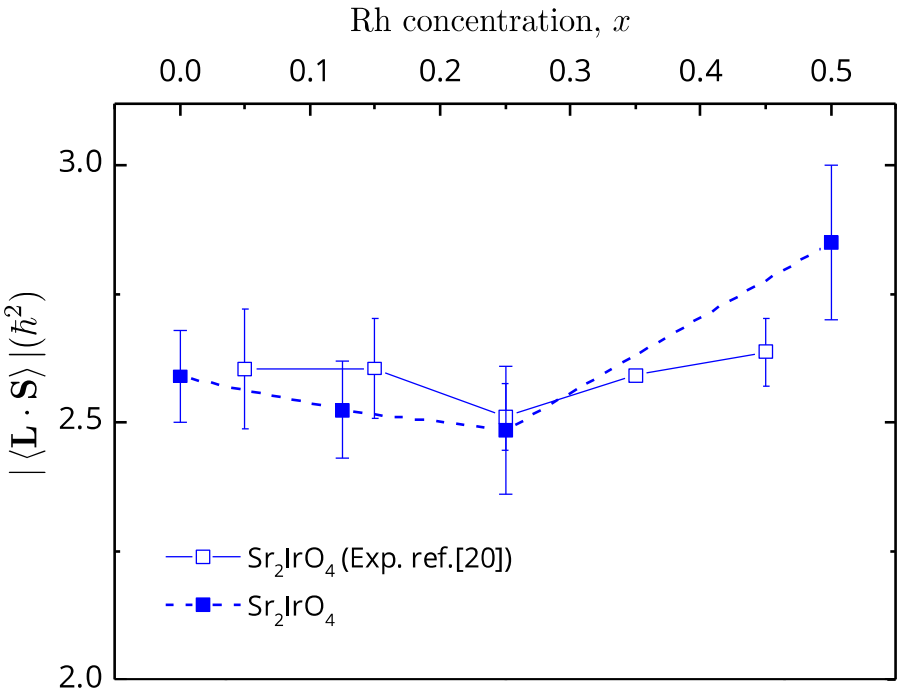
**(J.-H.Sim et al, PRB 94, 115149, 2016):**

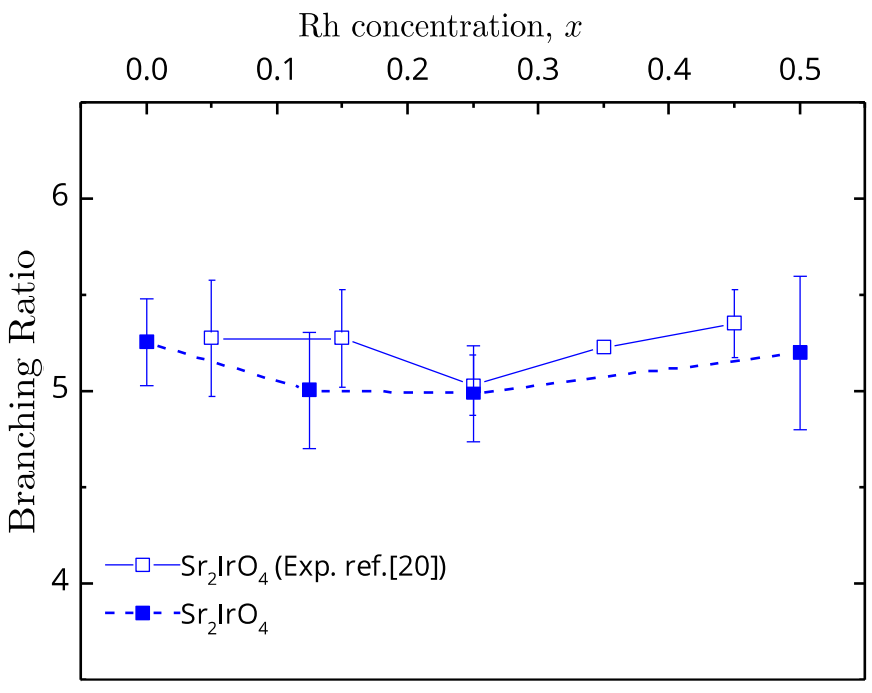
- ✓ **Almost zero additional cost**
- ✓ **Very simple implementation**
- ✓ **Still can make a bridge to the experiments**




Taillefumier, PRB (2002)

# Application (1): $Sr_2Ir_{1-x}Rh_xO_4$

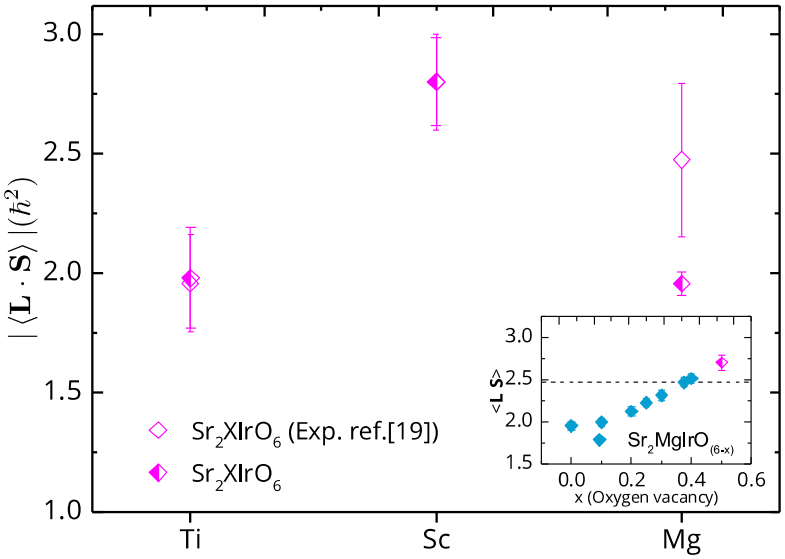


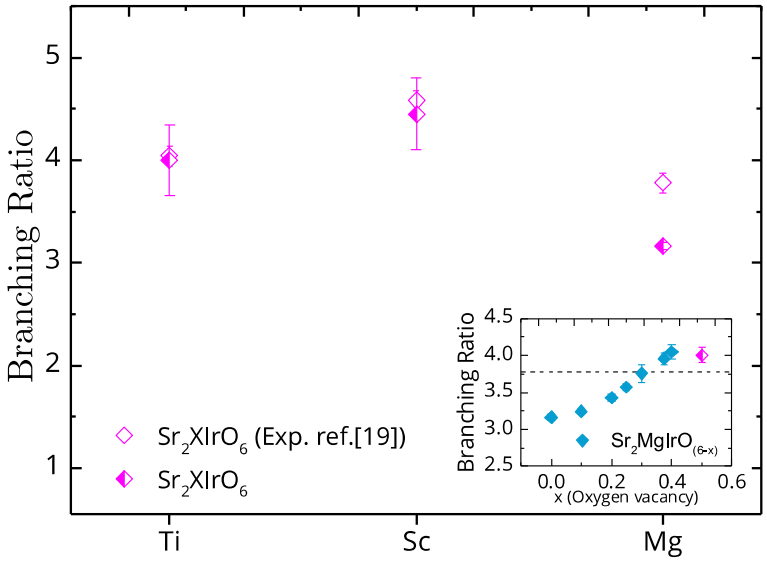




J.-H. Sim, et al., Phys. Rev. B (2016)

Exp: Chikara, *MJH* et al. PRB(R) (2015)

# Application (2): Double Perovskites

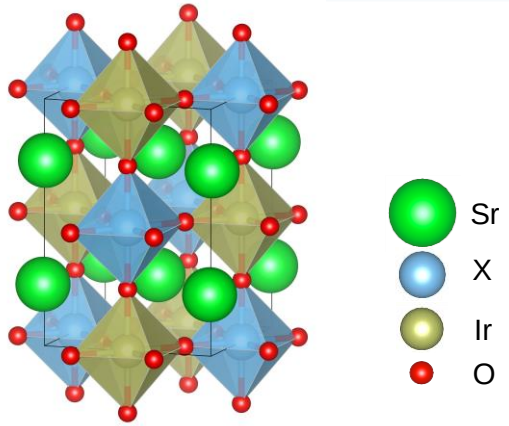


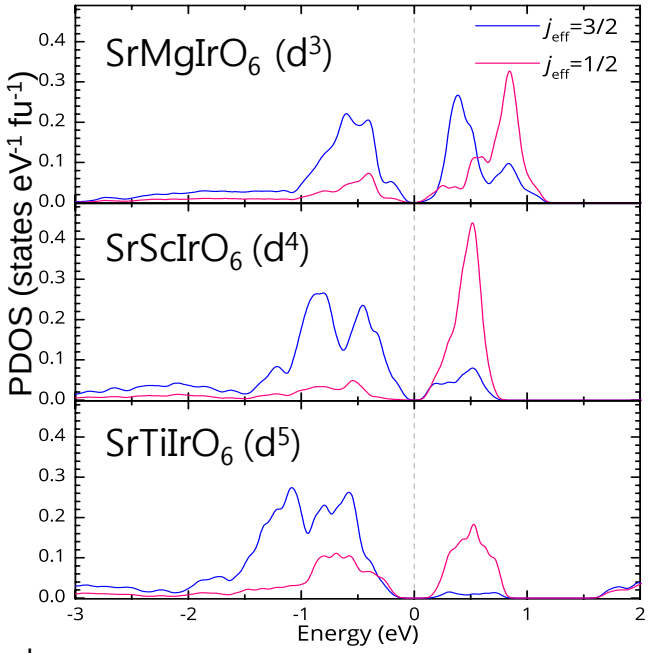




Exp:
Laguna-Marco et al.
PRB (2015)

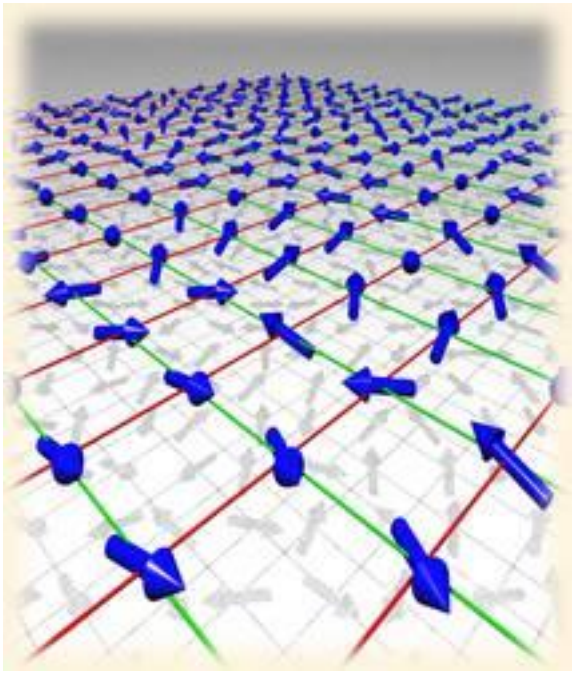
**$Sr_2XIrO_6$**
**(X=Mg, Sc, Ti)**

J.-H. Sim, et al.,
PRB (2016)



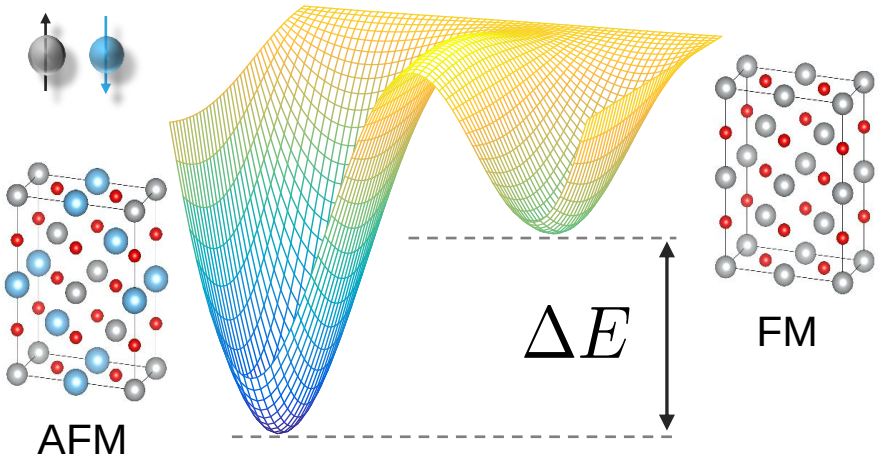
# Heisenberg Exchange Parameter



http://www.complexity-coventry.org/research/

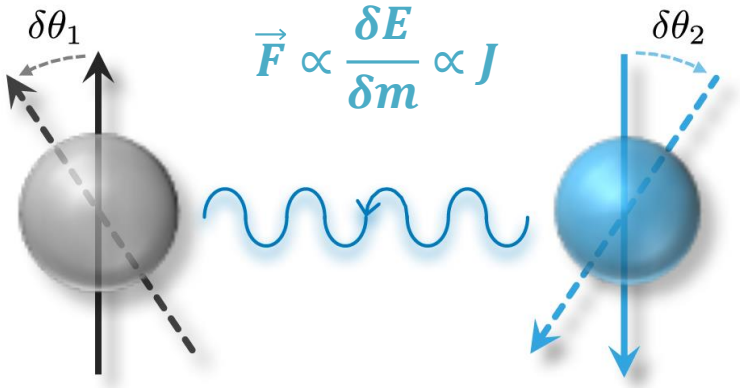
## Heisenberg Spin Hamiltonian

$$H = -\frac{1}{2}\sum_{ij} \boldsymbol{J_{ij}}\, \vec{S}_i \cdot \vec{S}_j$$




In the first-principles framework, it is often estimated by multiple supercell calculations of total energies.

# Magnetic Force Theory



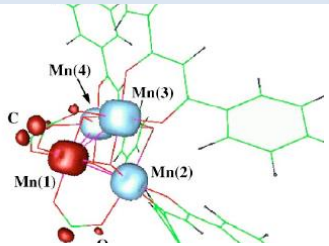

Liechtenstein et al. JMMM (1987)
Wan et al. PRL (2006)
Szilva et al. PRL (2013)
Pi et al. PRL (2014)
Kvashnin et al. PRL (2016)

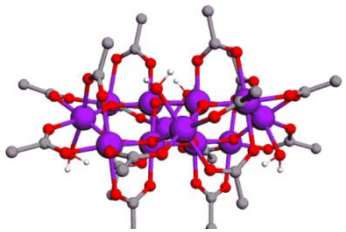
## Magnetic Force Theory in OpenMX : *MJH*, T. Ozaki, and J. Yu, PRB (2004)

**APPENDIX: A FORMALISM FOR THE CALCULATION OF EFFECTIVE EXCHANGE-COUPLING PARAMETERS**

We present a method for the calculations of effective exchange-coupling parameters by employing the nonorthogonal LCAO basis which was used for the results presented in Sec. V. Applying the rigid spin approximation (RSA) in the noncollinear magnetic perturbations[45–48] for the calculated DFT ground state, we can obtain the following expression for the exchange interaction $J_{ij}$ between two different sites $i$ and $j$:

$$J_{ij} = \frac{1}{2\pi} \int^{\varepsilon F} d\varepsilon \, \mathrm{Tr}[\hat{G}^{\uparrow}_{ij} \hat{V}_j \hat{G}^{\downarrow}_{ji} \hat{V}_i], \qquad \text{(A1)}$$



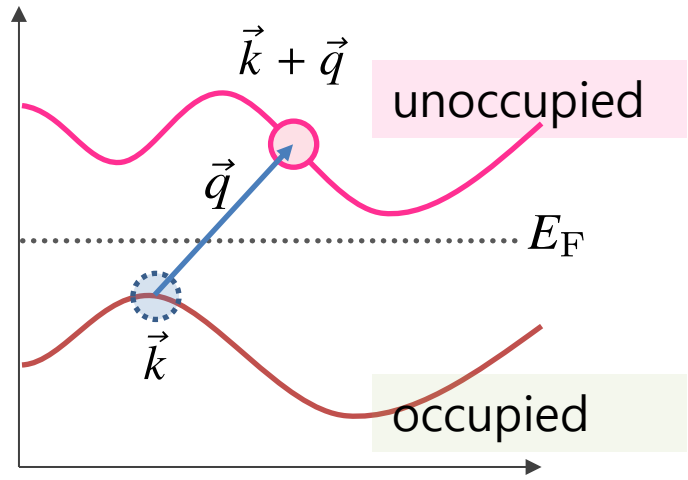
| | | $J_{12}$ | $J_{13}$ | $J_{23}$ |
|---|---|---|---|---|
| $Mn_{12}(N=84)$ | LSDA | −58.5 | −95.2 | −207.8 |
| $Mn_{12}(N=100)$ | LSDA | −84.8 | −83.1 | −71.0 |
| $Mn_{12}(N=148)$ | LSDA | −168.9 | −91.7 | −58.9 |
| $Mn_{12}(N=148)$ | LSDA($U$)[a] | −70.9 | −27.4 | −22.0 |
| Zeng *et al.*[b] | LSDA | −94.3 | −50.1 | −70.8 |
| Boukhvalov *et al.*[c] | LDA+$U$ | −36.8 | −32.7 | −3.4 |

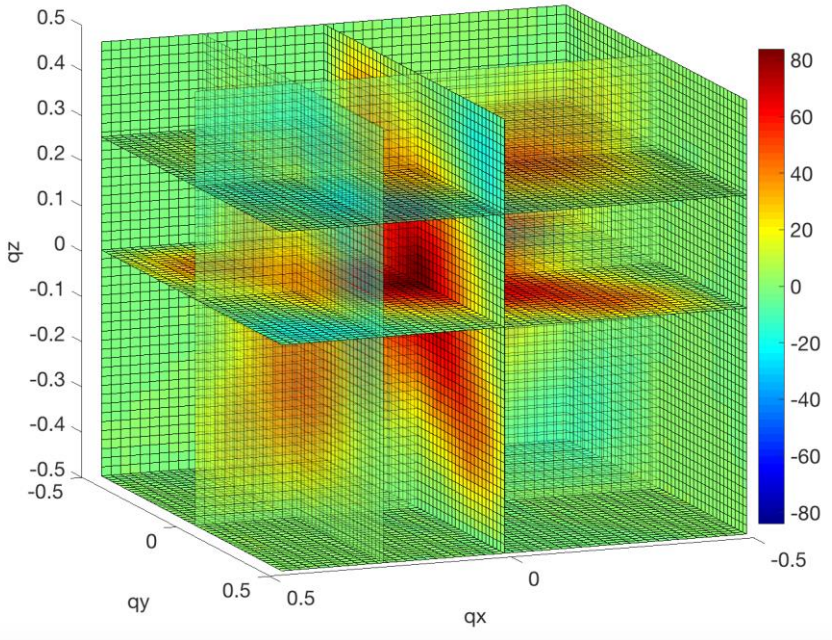
# Extension to Periodic Systems

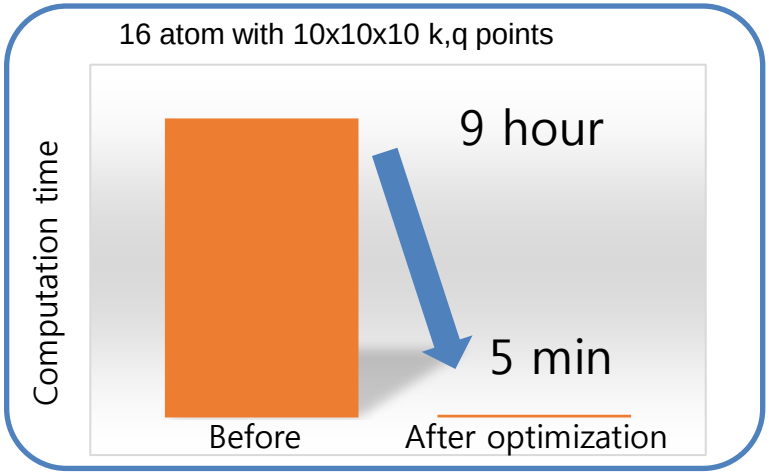
$$J_{ij}(\vec{R}) = \sum_{q} J_{ij}(\vec{q}) e^{-i\vec{R}\vec{q}}$$

$$J_{ij}(\vec{q}) = \sum_{n,n'} \sum_{k} \frac{f_{n,k\uparrow} - f_{n',k+q\downarrow}}{\epsilon_{n,k\downarrow} - \epsilon_{n',k+q\downarrow} - i\eta} M(n,n')$$

$$M(n,n') = Tr\left[\tilde{A}^{n\uparrow} V_i \tilde{A}^{n'\downarrow} V_j\right]$$

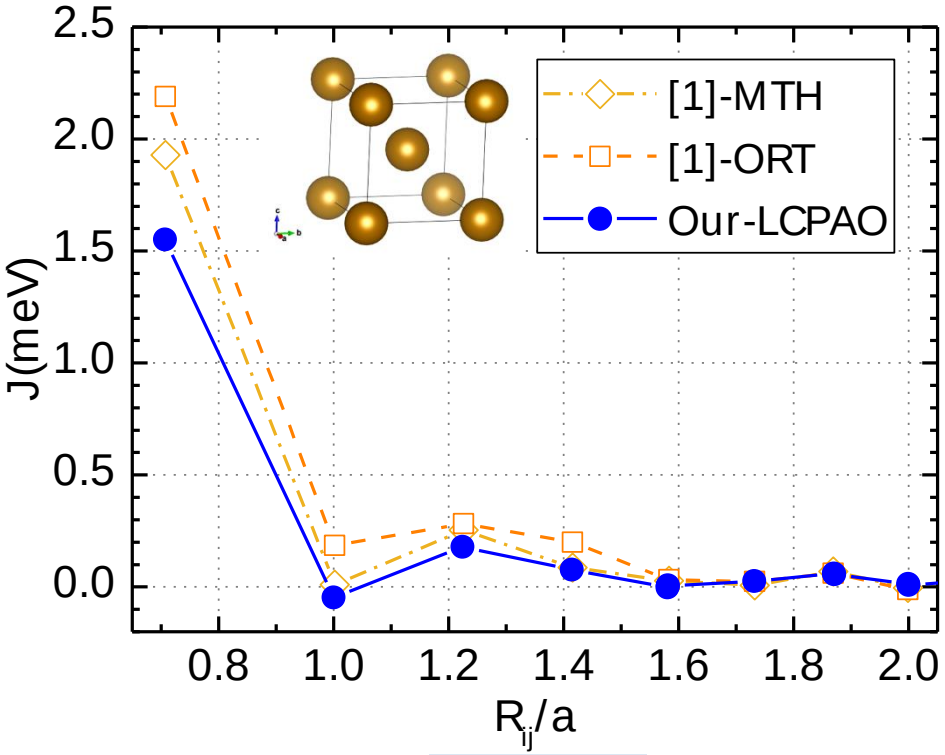


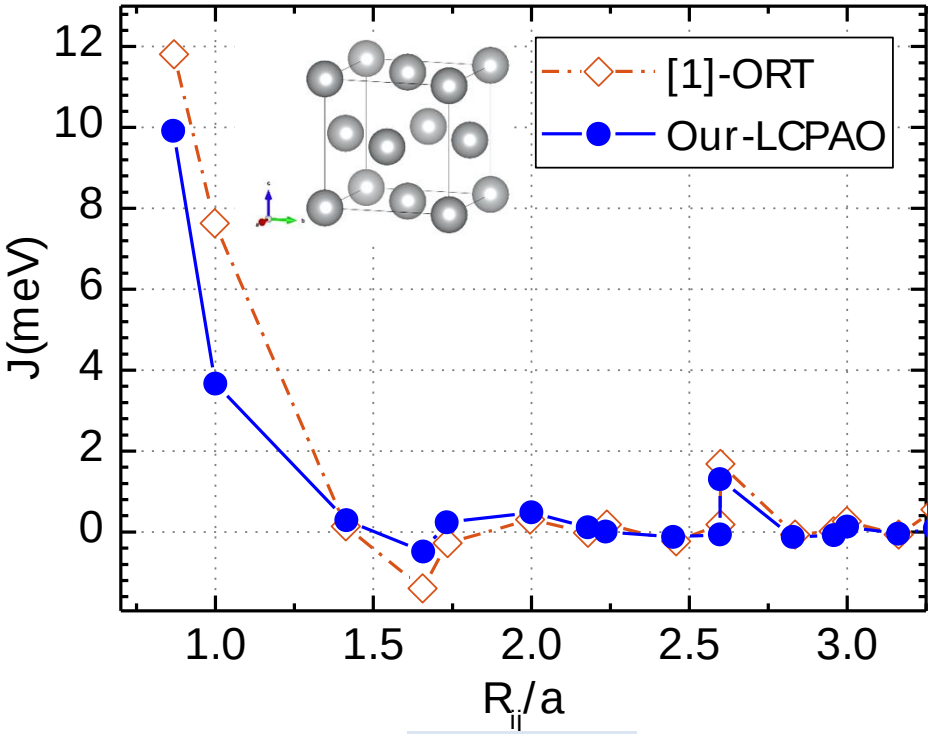



# Application to Transition Metals

**Hongki Yoon et al. (in preparation)**



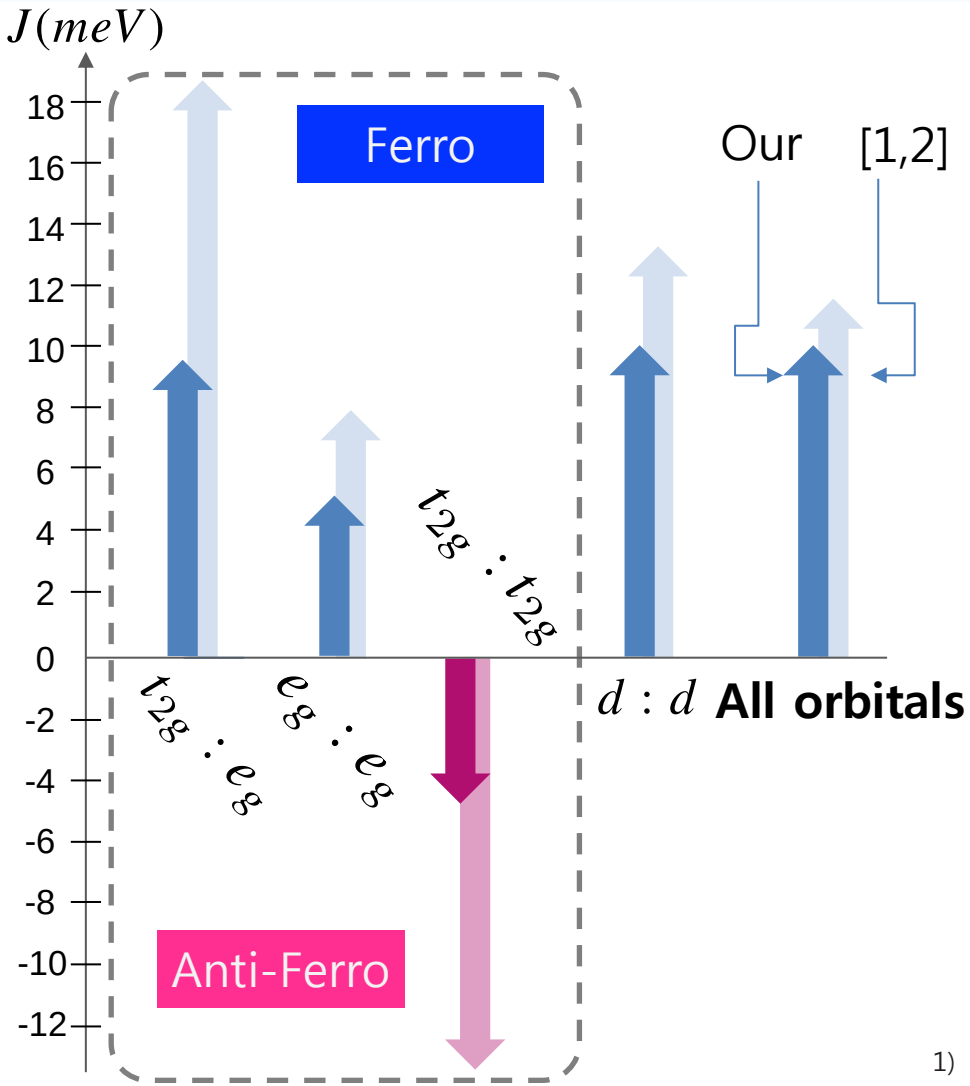

Fcc Ni




Bcc Fe

Ref 1: Kvashnin et al. Phys. Rev. B **91**, 125133 (2015)

# Orbital Decomposition (bcc Fe)




**Hongki Yoon et al. (in preparation)**

$J(meV)$

| Orbitals | Our | Other |
|---|---|---|
| eg-t2g | 9.8 | 19.1 |
| eg-eg | 5.3 | 8.1 |
| t2g-t2g | -4.8 | -13.6 |
| **d-d** | **10.3** | **13.6** |
| **All** | **10.3** | **11.9** |

Our result : $J_{d-d} = J_{tot}$

Ref 1,2 : $J_{d-d} \approx J_{tot}$


1) Kvashnin, Grånäs, Di Marco, Katsnelson, Lichtenstein, Eriksson, PRB (2015)

2) Kvashnin, Cardias, Szilva, Di Marco, Katsnelson, Lichtenstein, Nordström, Klautau, Eriksson PRL (2016)

# Summary

- ✓ We suggest a simple technique to directly calculate branching ratio through spin-orbit coupling. The calculation results of a series of iridate compounds are in good agreement with experiments.

- ✓ We extended our magnetic force formalism to the periodic bulk case and to have the orbital resolution. The result of Fe and NiO shows the good agreement with the previous calculations by FP-LMTO method. Combined with PP-LCPAO, this method can be a promising tool to study the large-scale magnetic systems.