Mott transition in 5d compounds

Xi Dai
Institute of Physics, CAS
collaborators:
Li Huang, Liang Du, H.M. Weng, Z. Fang

The materials with both strong interaction and spin-orbital coupling: 5d compounds

- Sr_2IrO_4: Mott insulators with AF order
- $A_2Ir_2O_7$: Pyrochlore lattice with possible Weyl fermions
- $Na_4Ir_3O_8$: frustrated spin system
- Na_2IrO_3: Topological insulator?
- ....

challenge to the electronic structure calculations

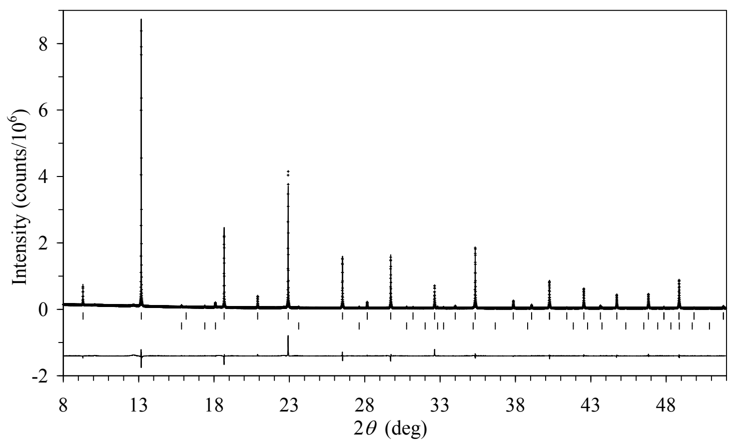
- rotational invariant form of the local Coulomb interaction
- LDA+DMFT: sign problem mostly induced by the spin orbital coupling
- LDA+Gutzwiller: generalized Gutzwiller projector

outline of this talk

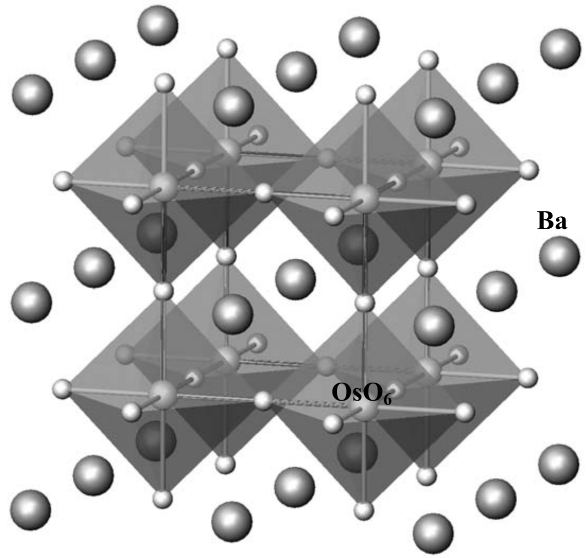
- Two possible Mott insulators in 5d systems: $BaOsO_3$, $NaIrO_3$
- Three band (t_{2g}) Hubbard model with spin orbital coupling
- two key questions: 1)How the nature of Mott transition will be changed by SOC; 2) How SOC will be changed by interaction
- calculations for the realistic materials

New material: $BaOsO_3$

Experimental geometrical structure

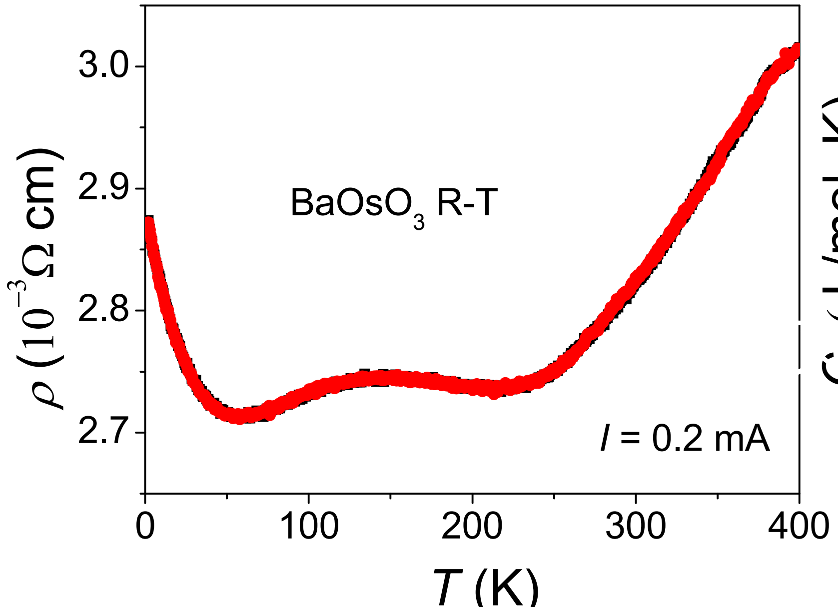


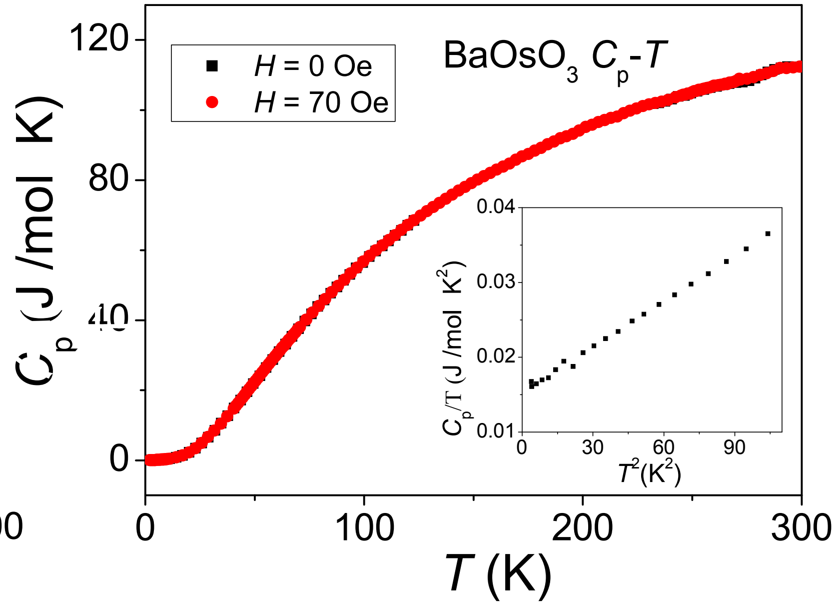
Observed (crosses), calculated (solid line), and difference synchrotron X-ray powder diffraction patterns of $BaOsO_3$ at 300 K. Bragg reflections are indicated by tick marks. The lower tick marks are given for reflections from the Os impurity (2.9 weight %).



Space group Pm-3m (no. 221)
a=b=c=4.02573(1) Å

Physical properties: exper. measurements

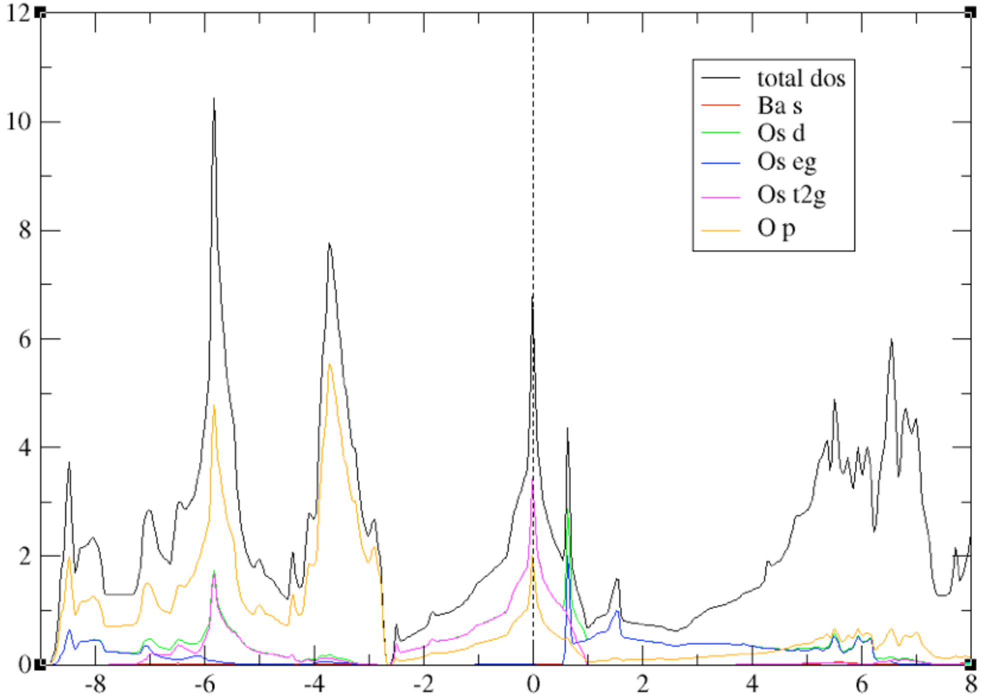


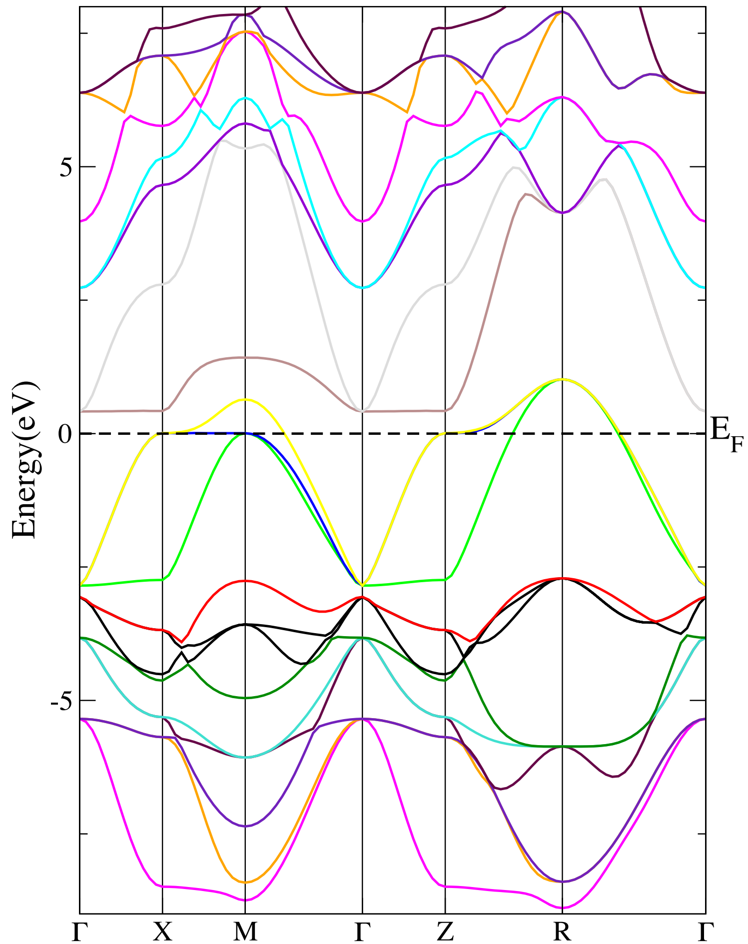


Y.G. Shi IOP & K. Yamaura NIMS

First-principles calculations: within GGA

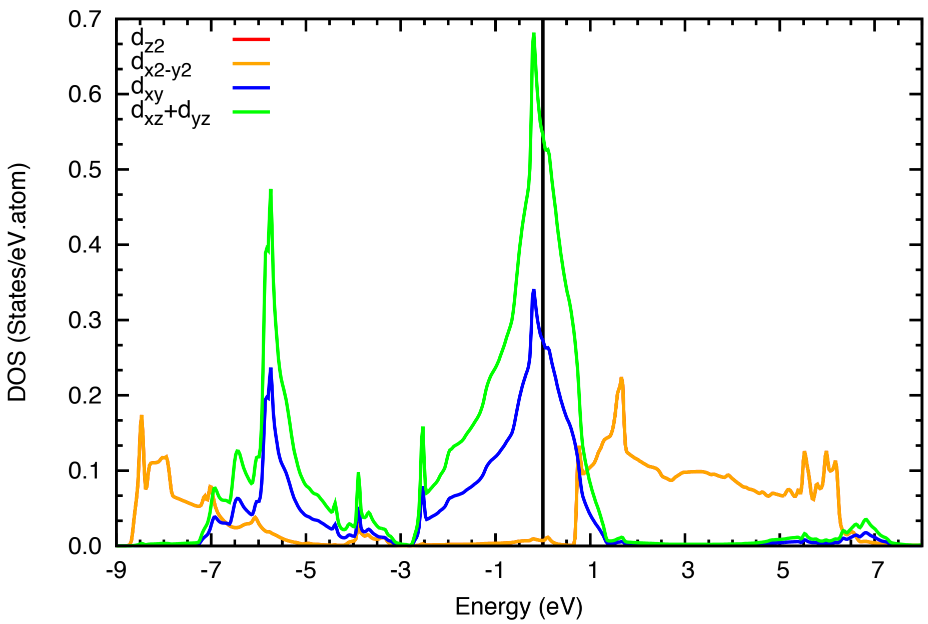
Non-magnetic case

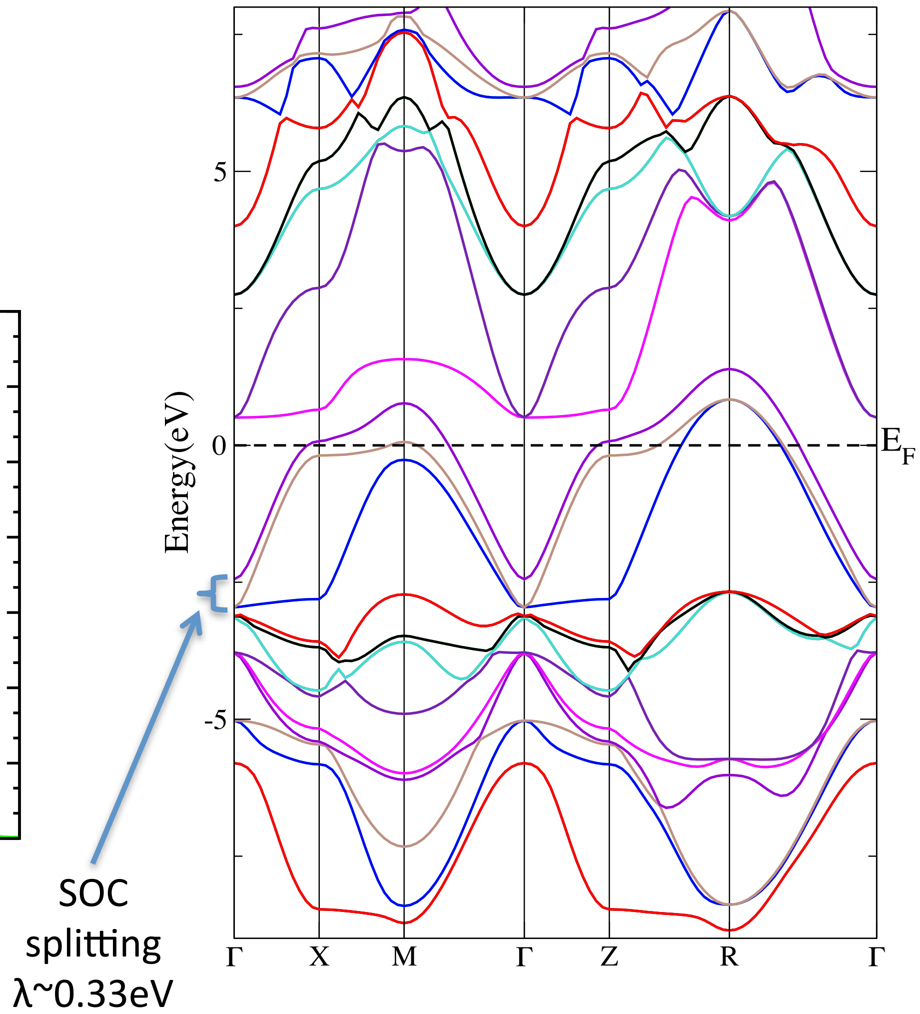




First-principles calculations: within GGA+SOC

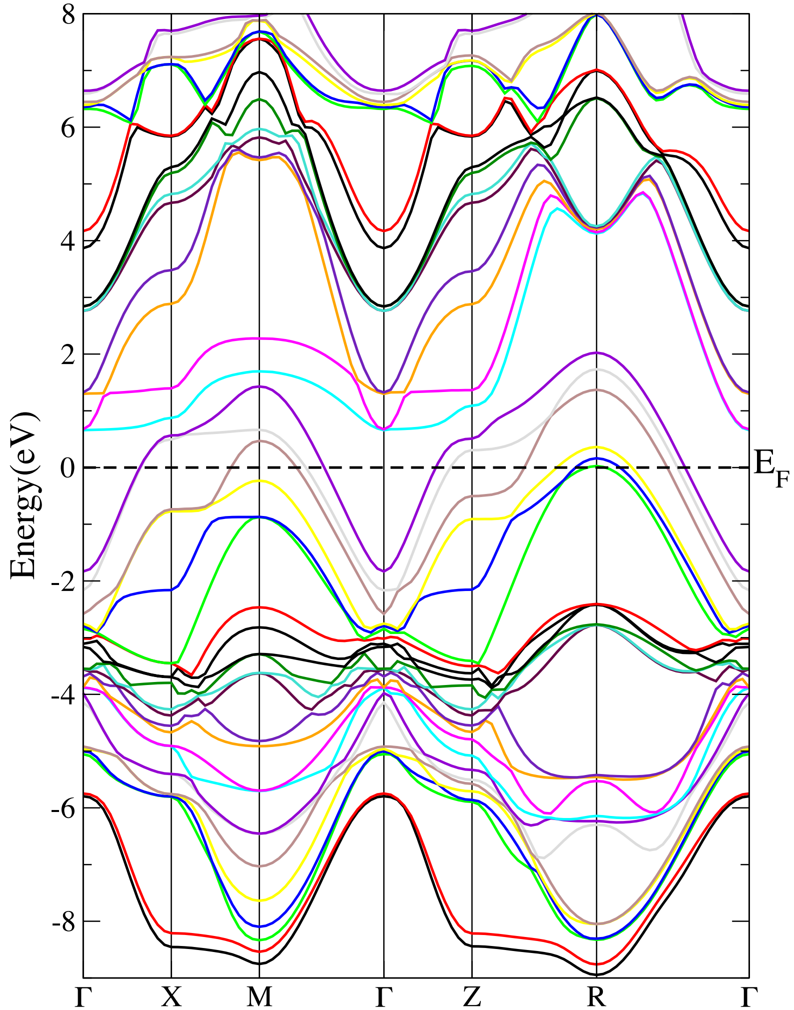
Starting from both Non-magnetic and Ferro-magnetic configureation, GGA +SOC calculations converge to the same Non-magnetic state.





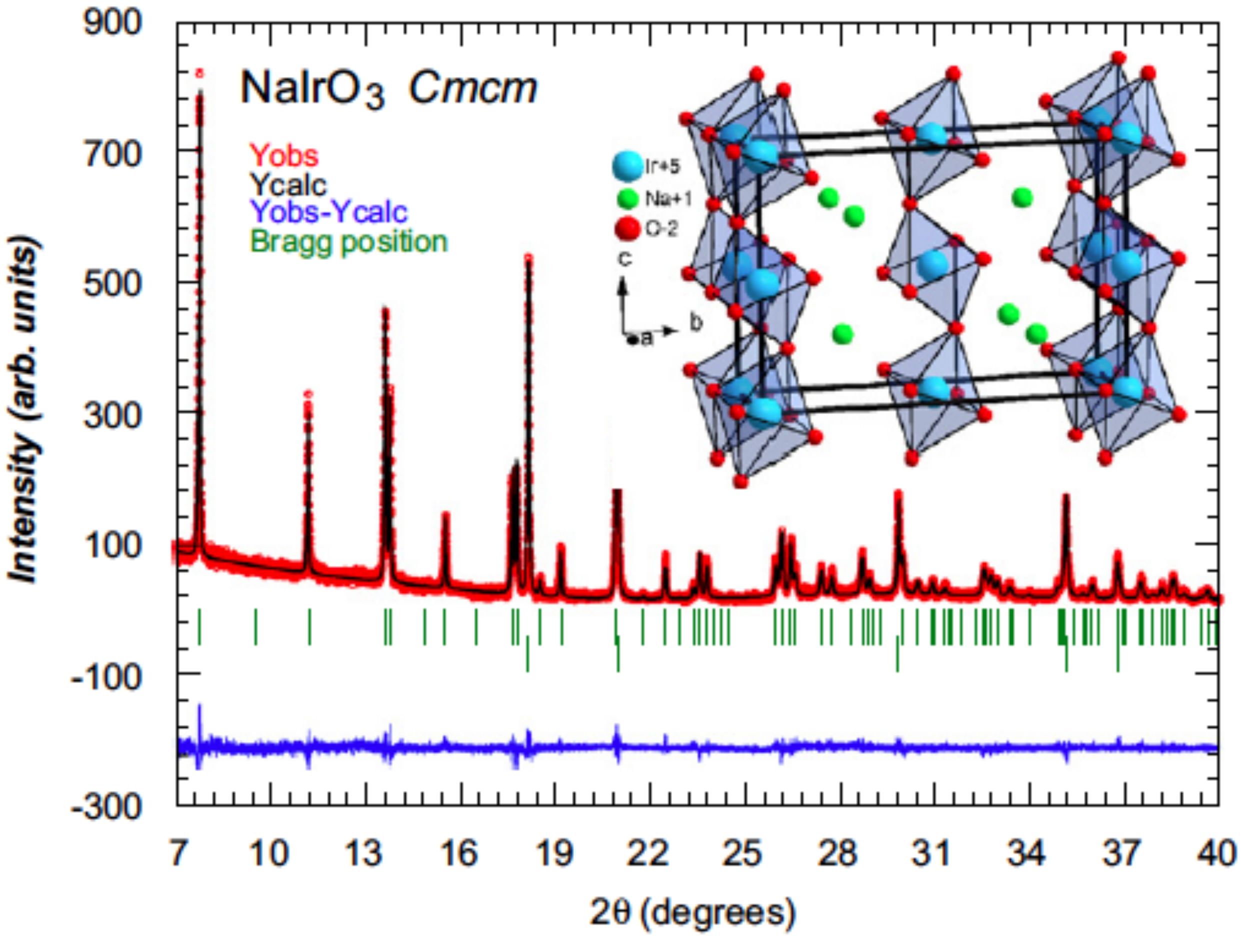
First-principles calculations: within GGA+SOC+U (U=3.0eV)

Ferromagnetic case



Post-perovskite $NaIrO_3$

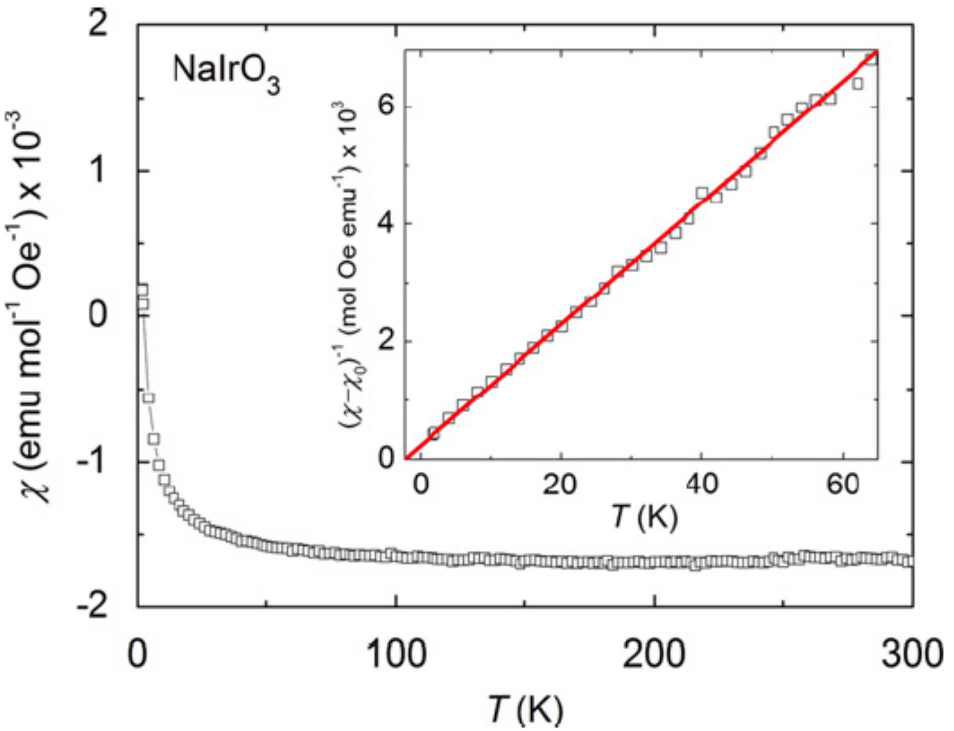
Experimental geometrical structure

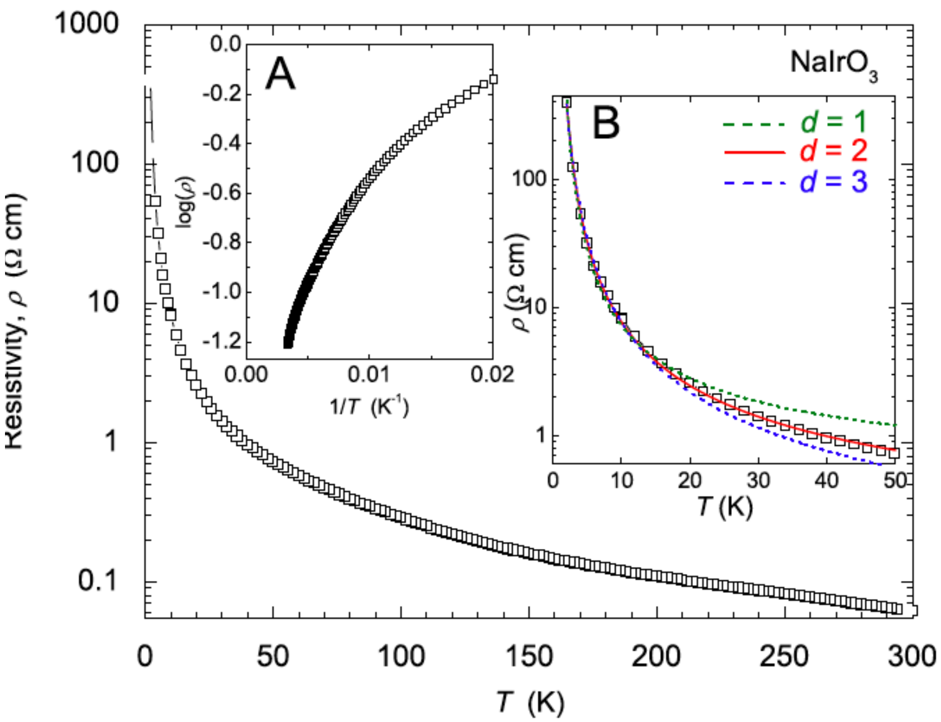


Space group
Cmcm (no. 63)
a=3.39683 Å
b=10.357612 Å
c=7.17663 Å

Cava's group in Princeton

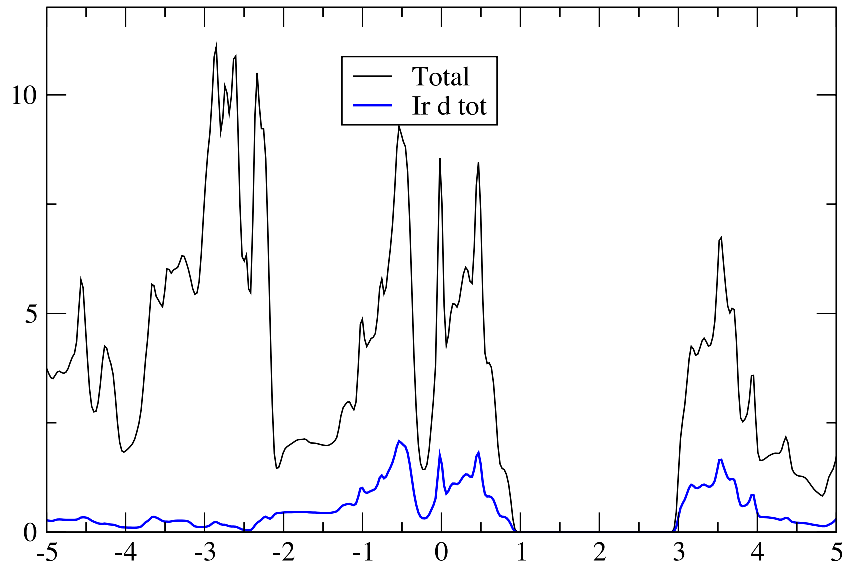
Physical properties: exper. measurements

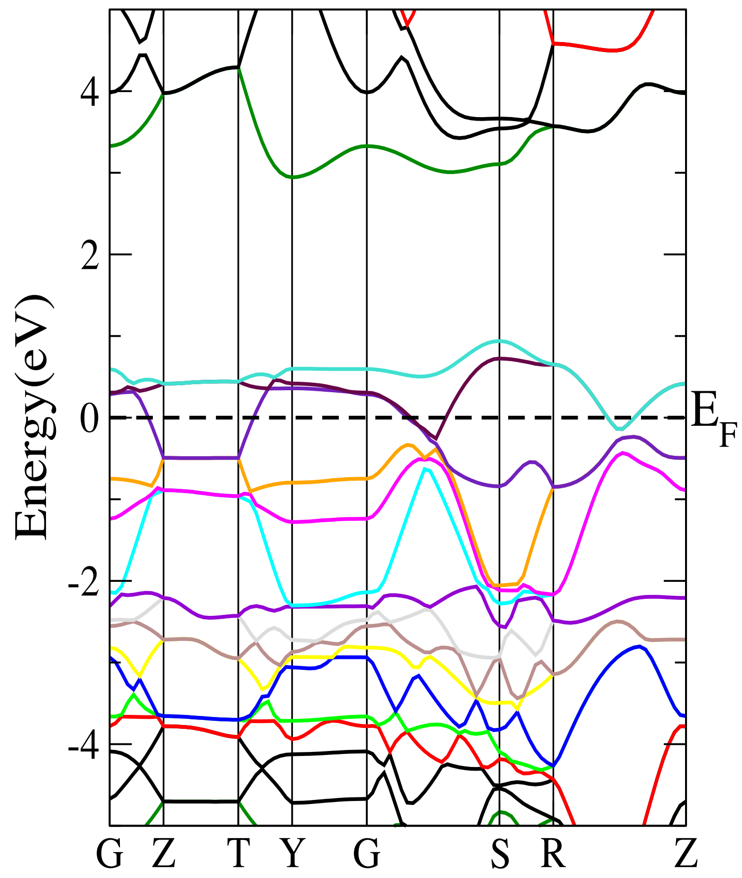




First-principles calculations: within GGA

Non-magnetic case



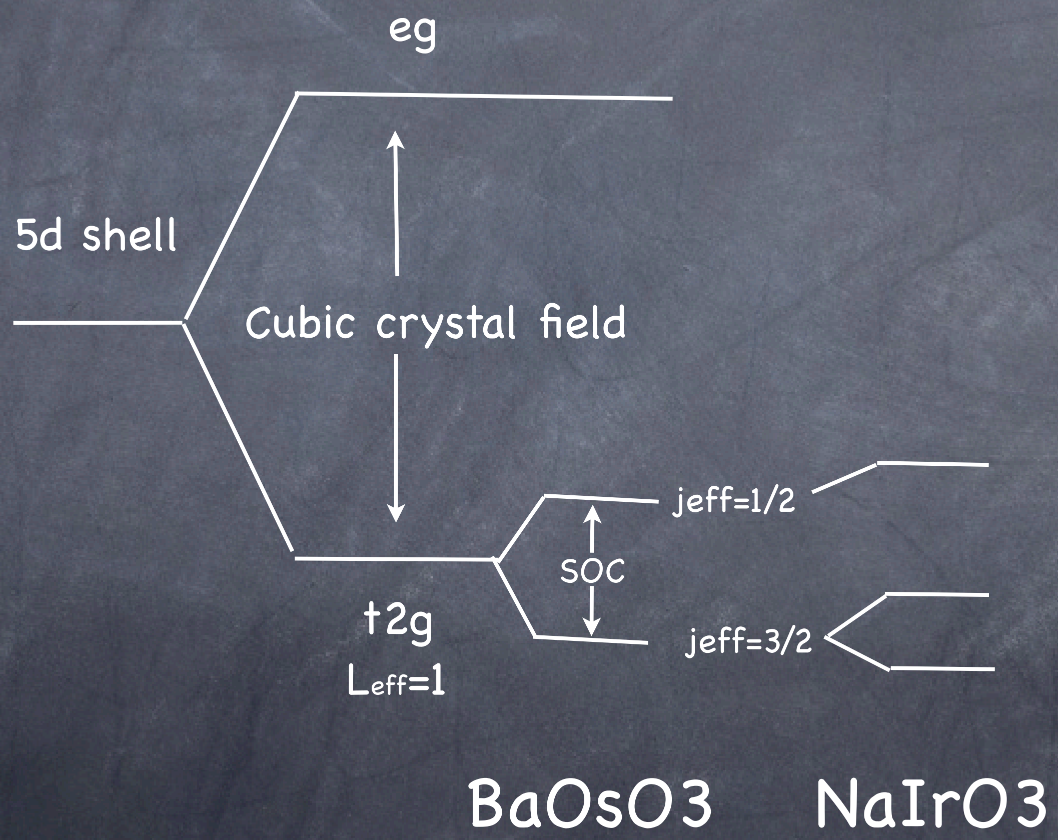


GGA+SOC+U predicts AF insulator only when U>7.0ev!

Conclusion from GGA+SOC+U calculation for the both materials

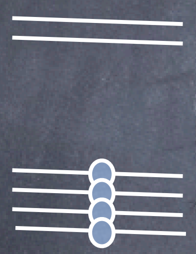
- The insulating states in both materials can only be obtained with GGA+SOC+U only when U>7.0eV, which is highly unrealistic
- Hartree-Fock mean field theory can not describe this featureless insulating state

The Crystal field splitting



Two different schemes to couple the spin and orbital degree of freedom for many electrons

JJ coupling



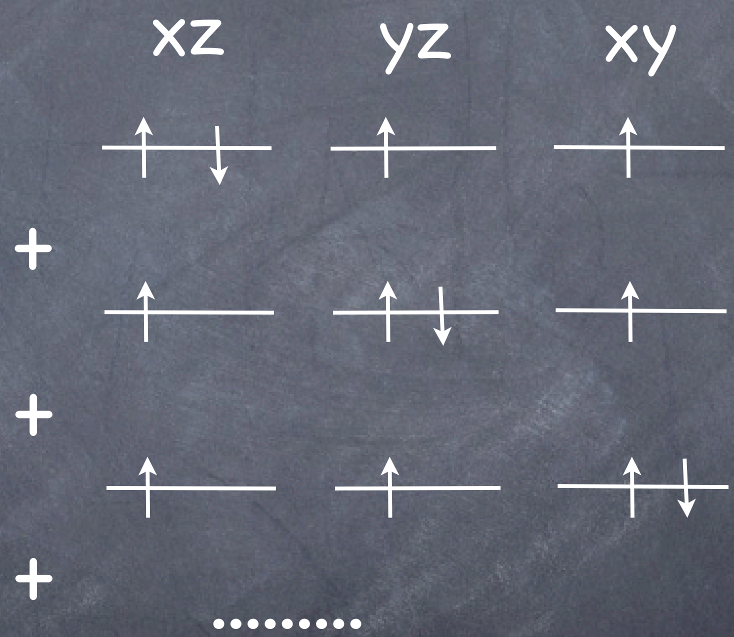
$j=1/2$

$j=3/2$

$J^2=0$

$U, J_H \ll \lambda$

LS coupling



$L^2=1(1+1);S^2=1(1+1);J^2=0$

$U, J_H \gg \lambda$

General tight-binding Hamiltonian

Three-band Hubbard Model With SOC:

$$H = H_0 + H_{SO} + H_U$$

Kinetic Energy Terms:

$$H_0 = \sum_{i \neq j, a\sigma} t_{ij}^{a\sigma} d_{i,a\sigma}^{\dagger} d_{j,a\sigma}$$

Spin-Orbit Coupling Terms:

$$H_{SO} = \sum_{a\sigma} \sum_{b\sigma'} \zeta \langle a\sigma | l_x s_x + l_y s_y + l_z s_z | b\sigma' \rangle d_{a\sigma}^{\dagger} d_{b\sigma'}$$

Coulomb Interaction Terms:

$$H_U = U \sum_a n_{a,\uparrow} n_{a,\downarrow} + U' \sum_{a<b,\sigma\sigma'} n_{a,\sigma} n_{b,\sigma'} - J_z \sum_{a<b,\sigma} n_{a,\sigma} n_{b,\sigma}$$

$$- J_{xy} \sum_{a<b} \left(d_{a,\uparrow}^{\dagger} d_{a,\downarrow} d_{b,\downarrow}^{\dagger} d_{b,\uparrow} + d_{a,\uparrow}^{\dagger} d_{a,\downarrow}^{\dagger} d_{b,\uparrow} d_{b,\downarrow} + h.c. \right)$$

Many body techniques used here

- Dynamical mean field theory (DMFT) with continuous time quantum Monte Carlo method as impurity solver
- Generalized Gutzwiller approximation

Rotational Invariant Gutzwiller Approximation

Gutzwiller variational wavefunction:

$$|\Psi_G\rangle = \mathcal{P}|\Psi_0\rangle = \prod_{\mathbf{R}} \mathcal{P}_{\mathbf{R}}|\Psi_0\rangle$$

$$\mathcal{P}_{\mathbf{R}} = \sum_{\Gamma\Gamma'} \lambda(\mathbf{R})_{\Gamma\Gamma'}|\Gamma, \mathbf{R}\rangle\langle\Gamma', \mathbf{R}|$$

$|\Gamma\rangle$: **eigenstates of atomic hamiltonian** H_U
Ψ_0: **uncorrelated wave function (Wick's Theorem holds)**
$\mathcal{P}_{\mathbf{R}}$: **projector operator modify weight of local configuration**
Gutzwiller Constraints:

$$\langle\Psi_0|\mathcal{P}^\dagger\mathcal{P}|\Psi_0\rangle = 1$$

$$\langle\Psi_0|\mathcal{P}^\dagger\mathcal{P}n_{i\alpha}|\Psi_0\rangle = \langle\Psi_0|n_{i\alpha}|\Psi_0\rangle$$

Total Energy In Gutzwiller Wavefunction:

$$E^G = E^G_{kin} + E^G_{loc} = \langle \Psi_G | H_0 | \Psi_G \rangle + \langle \Psi_G | (H_U + H_{SO}) | \Psi_G \rangle$$

Gutzwiller variational Procedure(Fixed n^0 Algorithm):

$$\frac{\partial E^G}{\partial \langle \Psi_0 |} = \sum_{i \neq j} \sum_{\gamma\delta} \sum_{\alpha\beta} t^{\alpha\beta}_{ij} \mathcal{R}^\dagger_{\alpha\gamma} \mathcal{R}_{\delta\beta} c^\dagger_{i\gamma} c_{j\delta} | \Psi_0 \rangle + \sum_{i\alpha} \eta_\alpha c^\dagger_{i\alpha} c_{i\alpha} | \Psi_0 \rangle = 0$$

$$\frac{\partial E^G}{\partial \lambda_{\Gamma\Gamma'}} = \sum_{\delta\beta} \left(\frac{\partial E_{kin}}{\partial \mathcal{R}_{\delta\beta}} \frac{\partial \mathcal{R}_{\delta\beta}}{\partial \lambda_{\Gamma\Gamma'}} + \frac{\partial E_{kin}}{\partial \mathcal{R}^\dagger_{\beta\delta}} \frac{\partial \mathcal{R}^\dagger_{\beta\delta}}{\partial \lambda_{\Gamma\Gamma'}} \right) + \frac{\partial E_{loc}}{\partial \lambda_{\Gamma\Gamma'}} + \sum_\alpha \eta_\alpha \frac{\partial n^G_\alpha}{\partial \lambda_{\Gamma\Gamma'}} = 0$$

The Lagrange parameters η_α come from Guztwiller Constraint.

$$\mathcal{R}^\dagger_{\alpha\gamma} = \frac{\mathrm{Tr}\left(\phi^\dagger c^\dagger_\alpha \phi c_\gamma \right)}{\sqrt{n^0_\gamma (1 - n^0_\gamma)}}$$

$$\phi_{II'} = \langle I | \hat{P} | I' \rangle \sqrt{\langle \Psi_0 | I' \rangle \langle I' | \Psi_0 \rangle}$$

Figure: Phase Diagram at plane of U and SOC ζ for $J/U = 0.25$ (a) RIGA (b) DMFT(CTQMC)

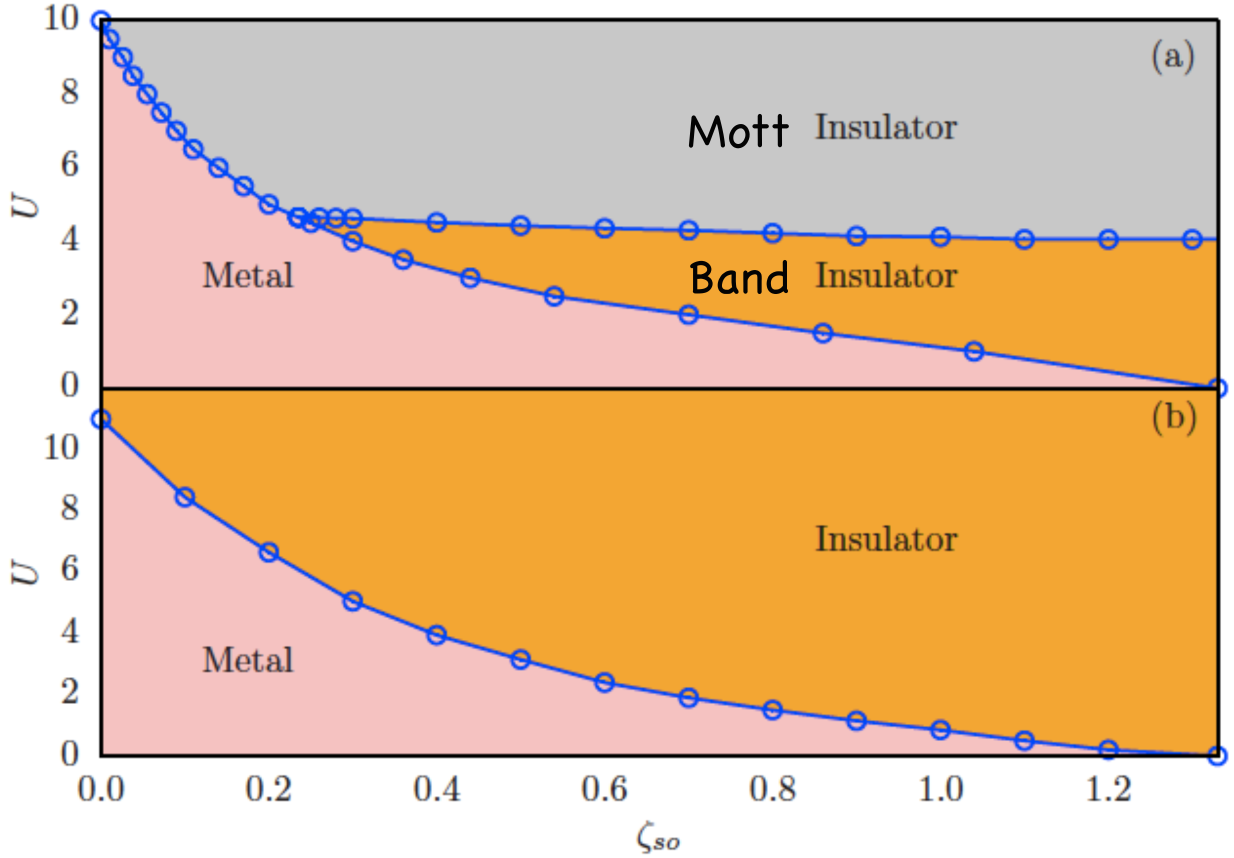


Figure: Energy and Quasiparticle weight as function of $0 \leq \delta n^0 \leq 1$ at fixed SOC $\zeta_{so} = 0.7$ and different $U = 1, 3, 6$, Derived by RIGA

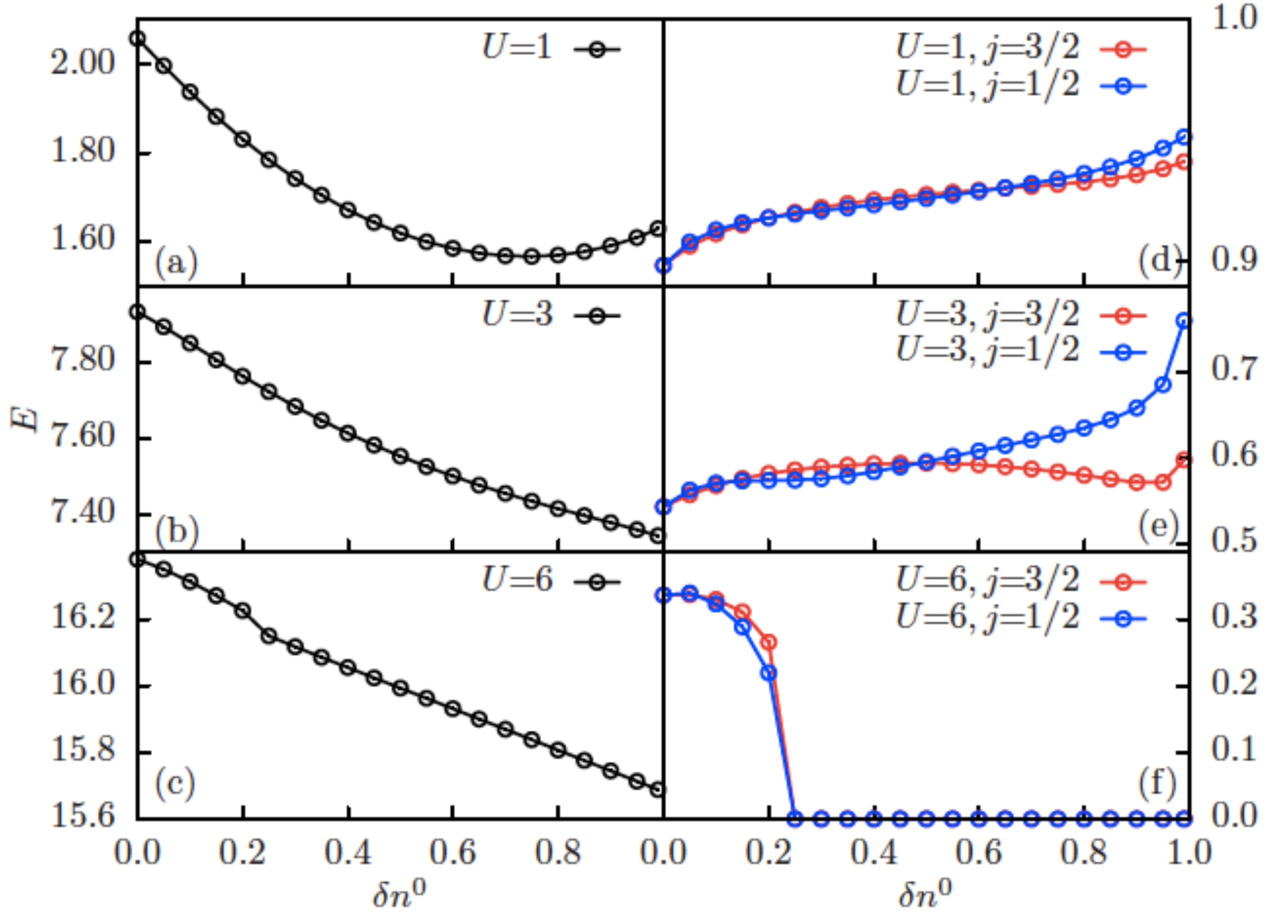
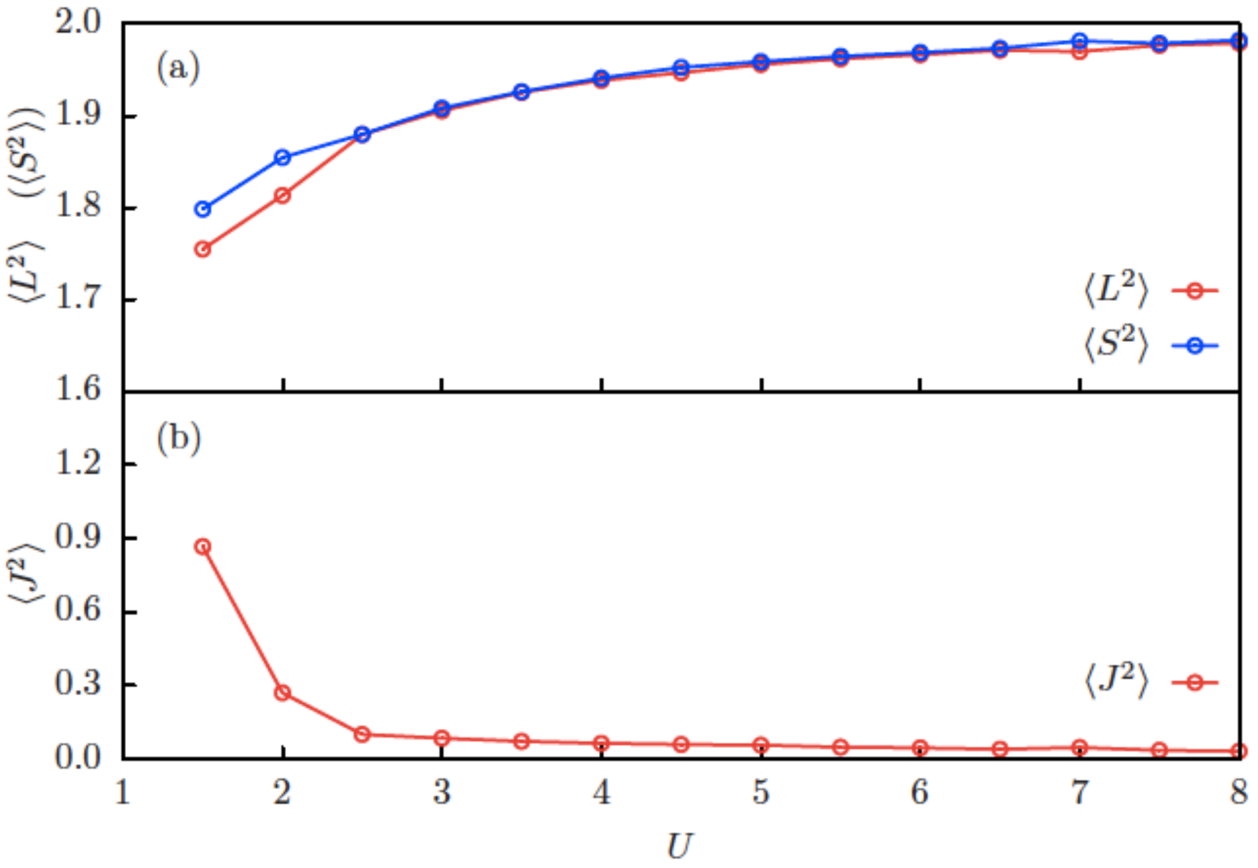
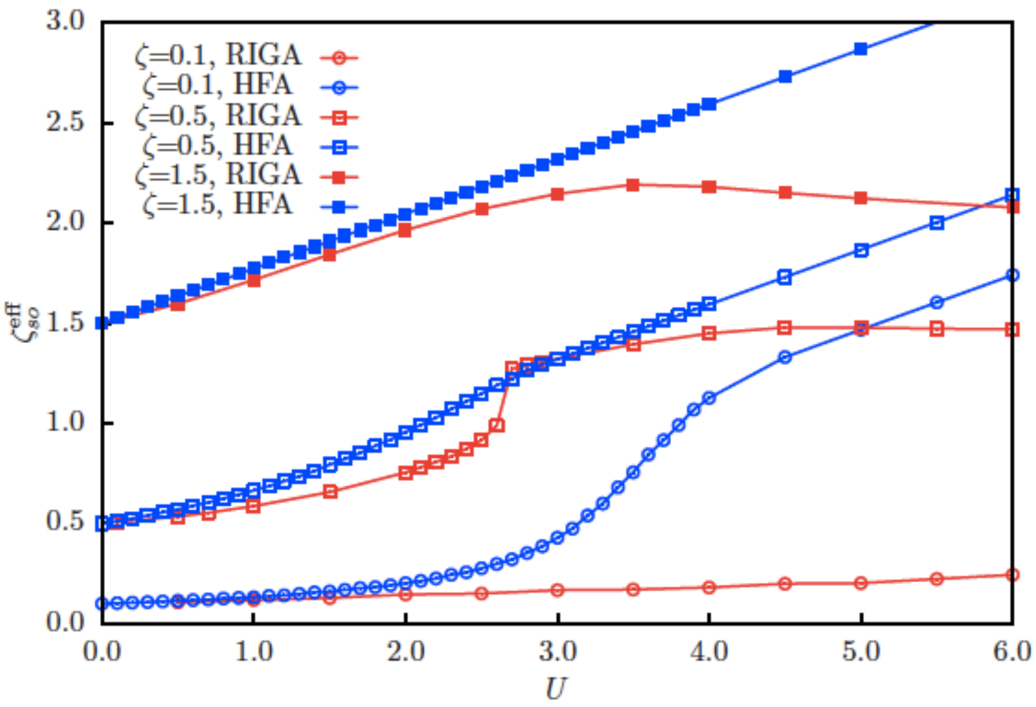


Figure: Expectational value of L^2, S^2, J^2 as function of U with fix SOC $\zeta = 0.7$, Derived by DMFT+CTQMC



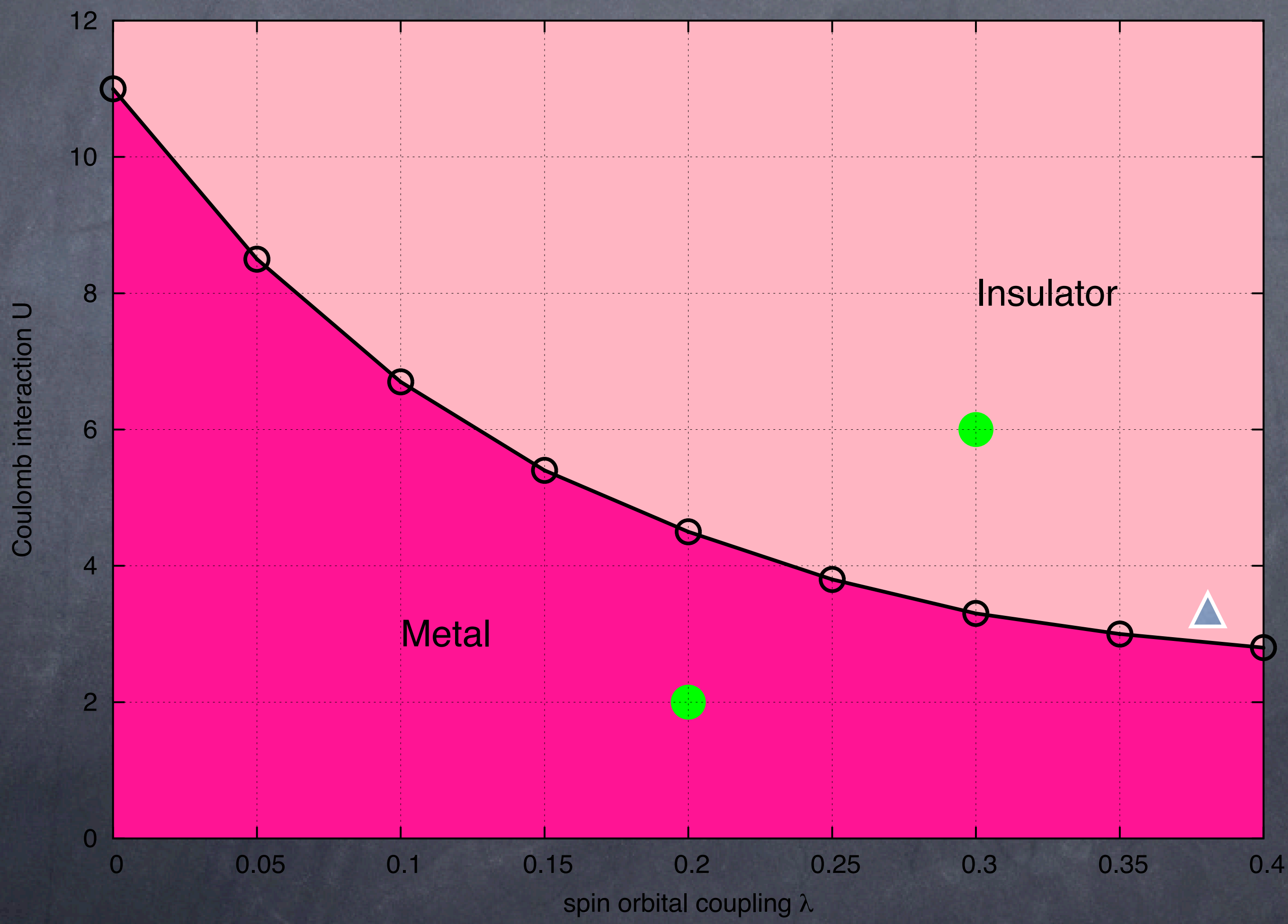
The effective spin orbital coupling modified by

Figure: Effective spin-orbit coupling ζ_{so}^{eff} as function U at fixed $\zeta = 0.1, 0.5, 1.5$, derived by HFA and RIGA

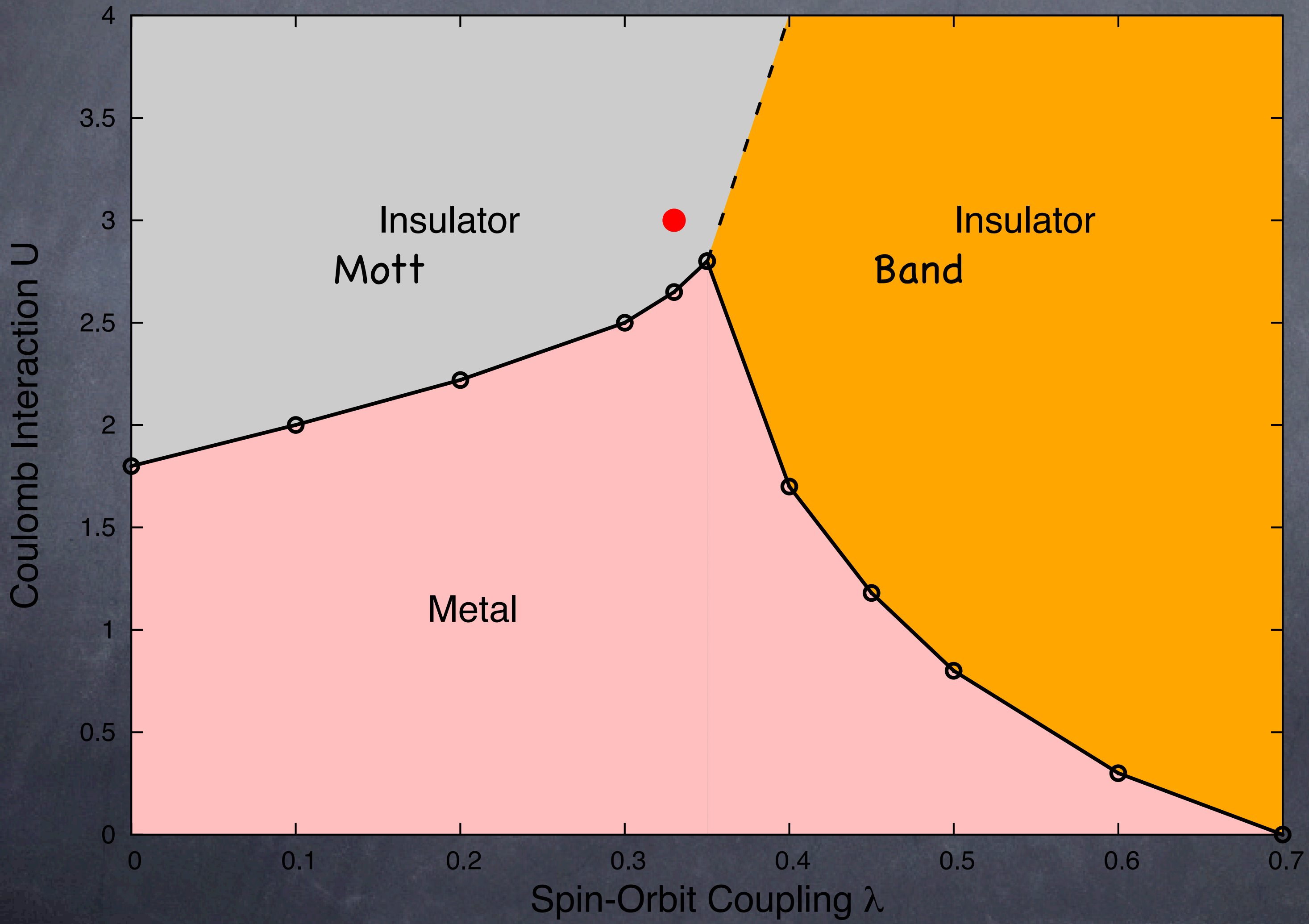


$$\lambda_{eff} = \frac{\partial E_{int}}{\partial n^0_{1/2}} - \frac{\partial E_{int}}{\partial n^0_{3/2}}$$

LDA+DMFT calculation for BaOsO3



The phase diagram of NaIrO3 obtained by LDA+Gutzwiller



Conclusions and outlook

- Both BaOsO3 and NaIrO3 are non-magnetic Mott insulators with the formation of local spin-orbital singlets
- The exact cancelation of spin and orbital moments
- doping? Possible exotic superconducting state
- magnetic solution?