Novel spin-orbit coupled electronic states in Ir oxides

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Outline

- Background: why 5*d* transition metal oxide?
- Sr_2IrO_4
  - ✓ Novel spin-orbit integrated J_{eff}=1/2 state
  - ✓ Anisotropic exchange interaction

Kim et al., PRL 101, 076402 (2008)
Jin et al., Phys. Rev. B 80, 075112(2009)
Moon et al., PRL 101, 226402 (2008)

- Na_2IrO_3
  - ✓ anti-ferromagnetic order with spin-orbit Zeeman field
  - ✓ Z_2 topological number?

Jin et al., arXiv:0907.0743
Kim et al., (to be published)

Why 4*d* and 5*d* Transition Metal Oxides?

- Close to metal-insulator transition instability
  - ✓ 4d and 5d orbitals are more extended than 3d's
  - ✓ reduced on-site Coulomb interaction strength
  - ✓ sensitive to lattice distortion, magnetic order, etc.
- Strong spin-orbit (SO) couplings
  - ✓ large atomic numbers: relativistic effect
    - V_{SO} (3d) $\leq$ 50 meV
    - V_{SO} (5d) $\approx$ 500 meV

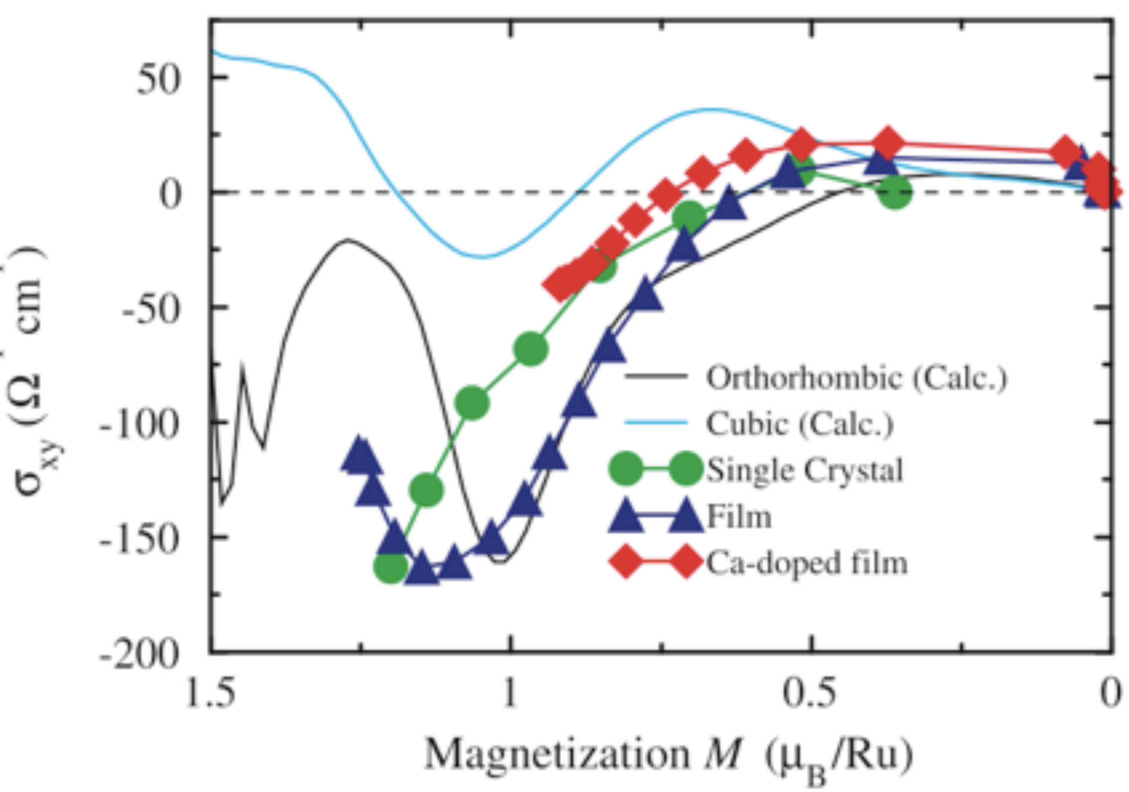
Physics driven by spin-orbit (SO) coupling

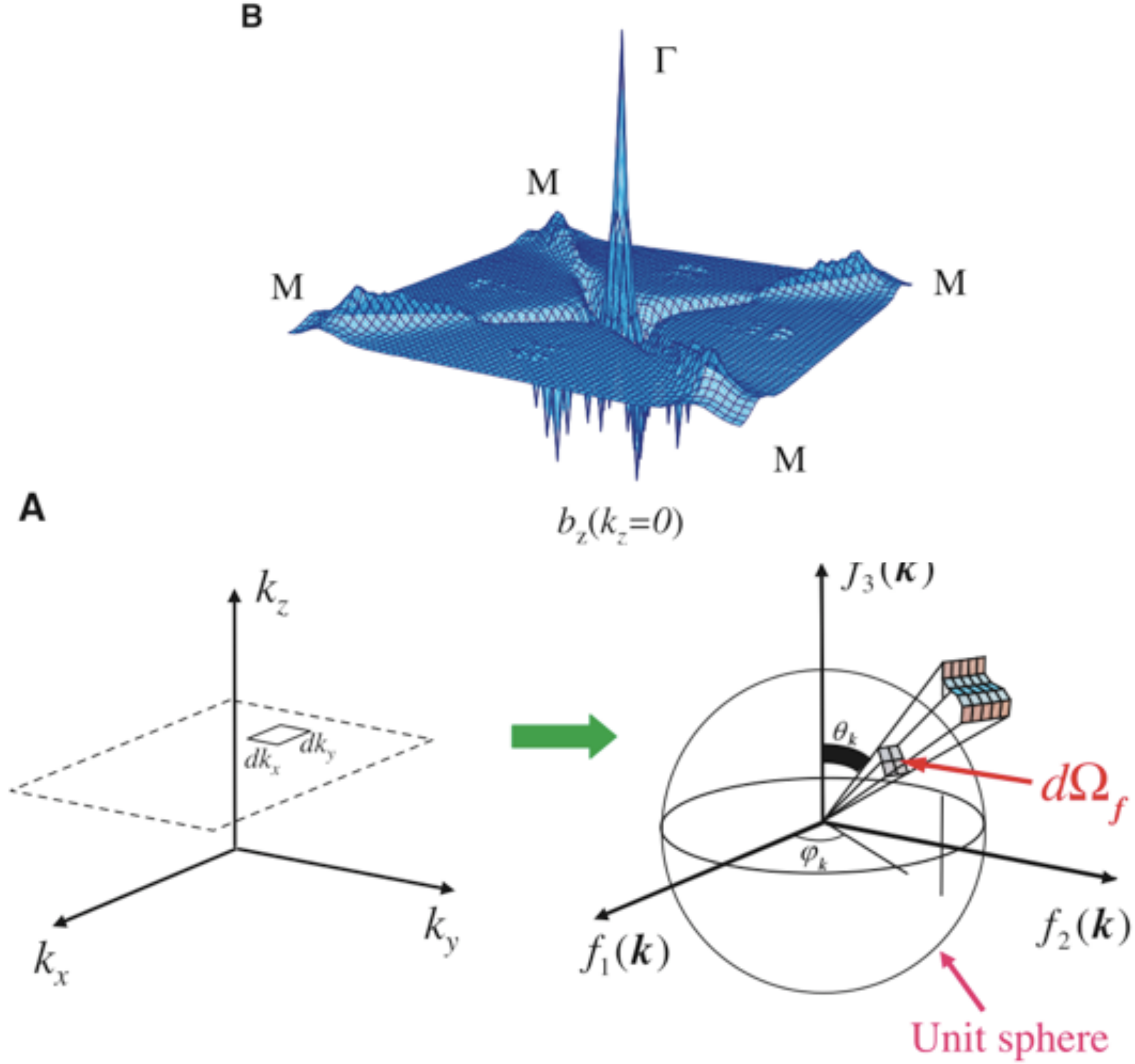
- Anisotropic magnetic exchange interactions:
  - ✓ Dzyaloshinskii-Moriya interaction
  - ✓ Multiferroic physics ...
- Anomalous Hall effect:
  - ✓ $SrRuO_3$
- Quantum spin Hall effect:
  - ✓ Spintronics
- Topological insulator
  - ✓ Magneto-electric effect, axion, ...

Anomalous Hall effect and magnetic monopoles in momentum space

$$a_{n\mu}(\boldsymbol{k}) = i\langle u_{n\mathbf{k}} | \frac{\delta}{\delta \mathbf{k}_\mu} | u_{n\mathbf{k}} \rangle \qquad \sigma_{xy} = \sum_{n,\mathbf{k}} n_F[\varepsilon_n(\mathbf{k})] b_z(\mathbf{k}) \qquad b_z(\mathbf{k}) = F_{xy}(\mathbf{k})$$

$$F_{\mu\nu} = \partial_{k_\mu} a_{n\nu} - \partial_{k_\nu} a_{n\mu}$$

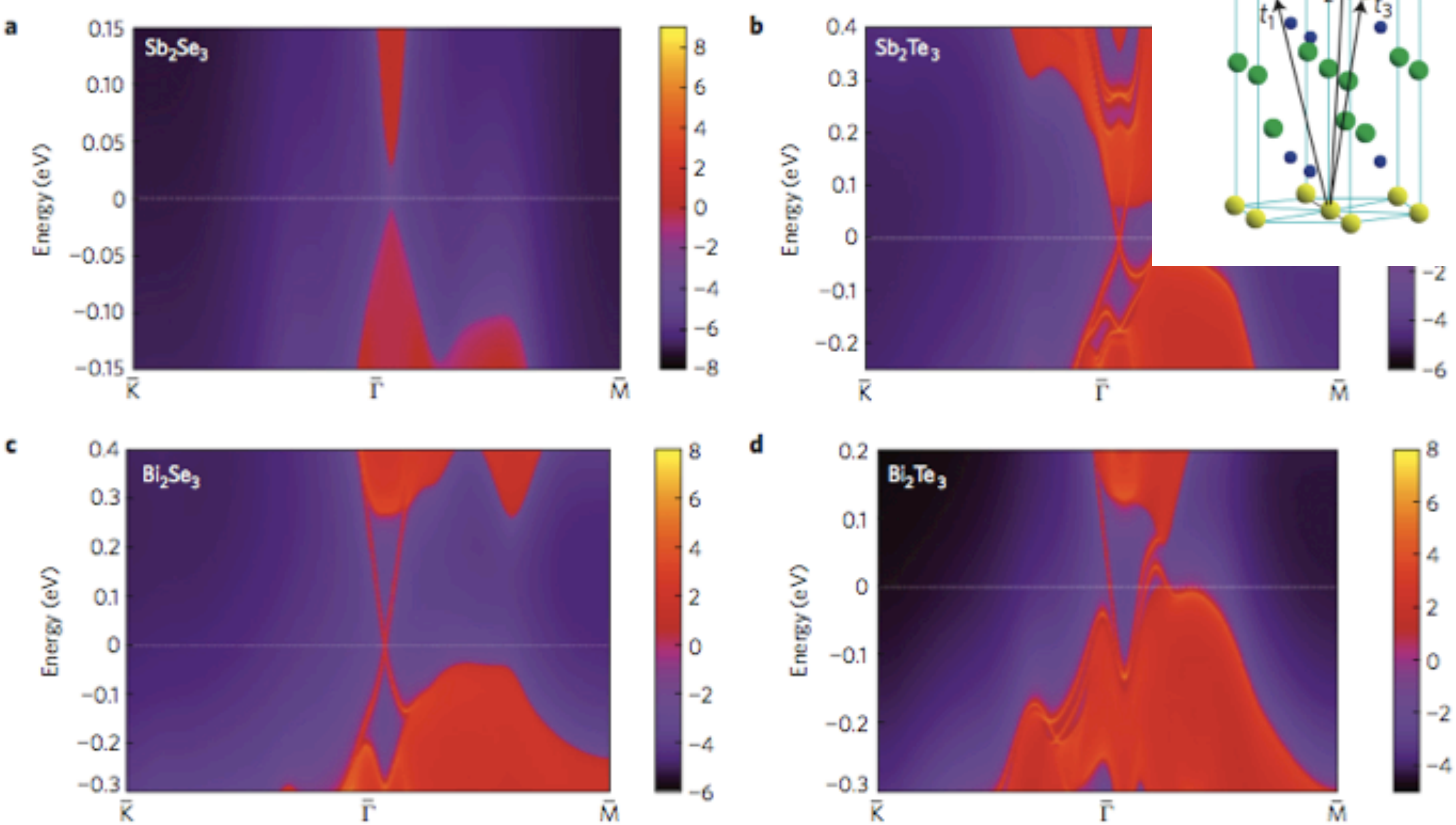


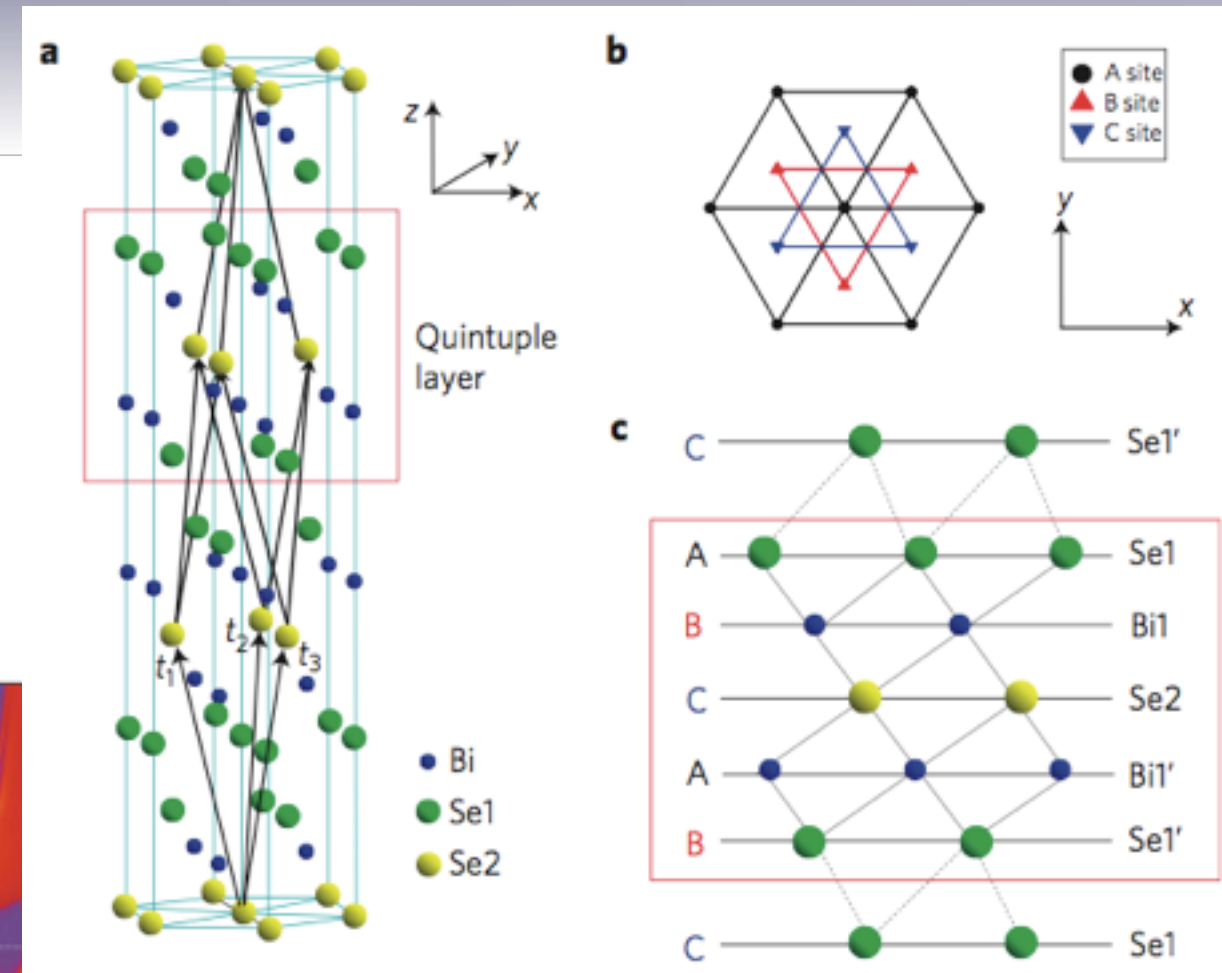


Z. Fang et al., Science 302, 92 (2003)

Quantum spin Hall effect and Topological insulators

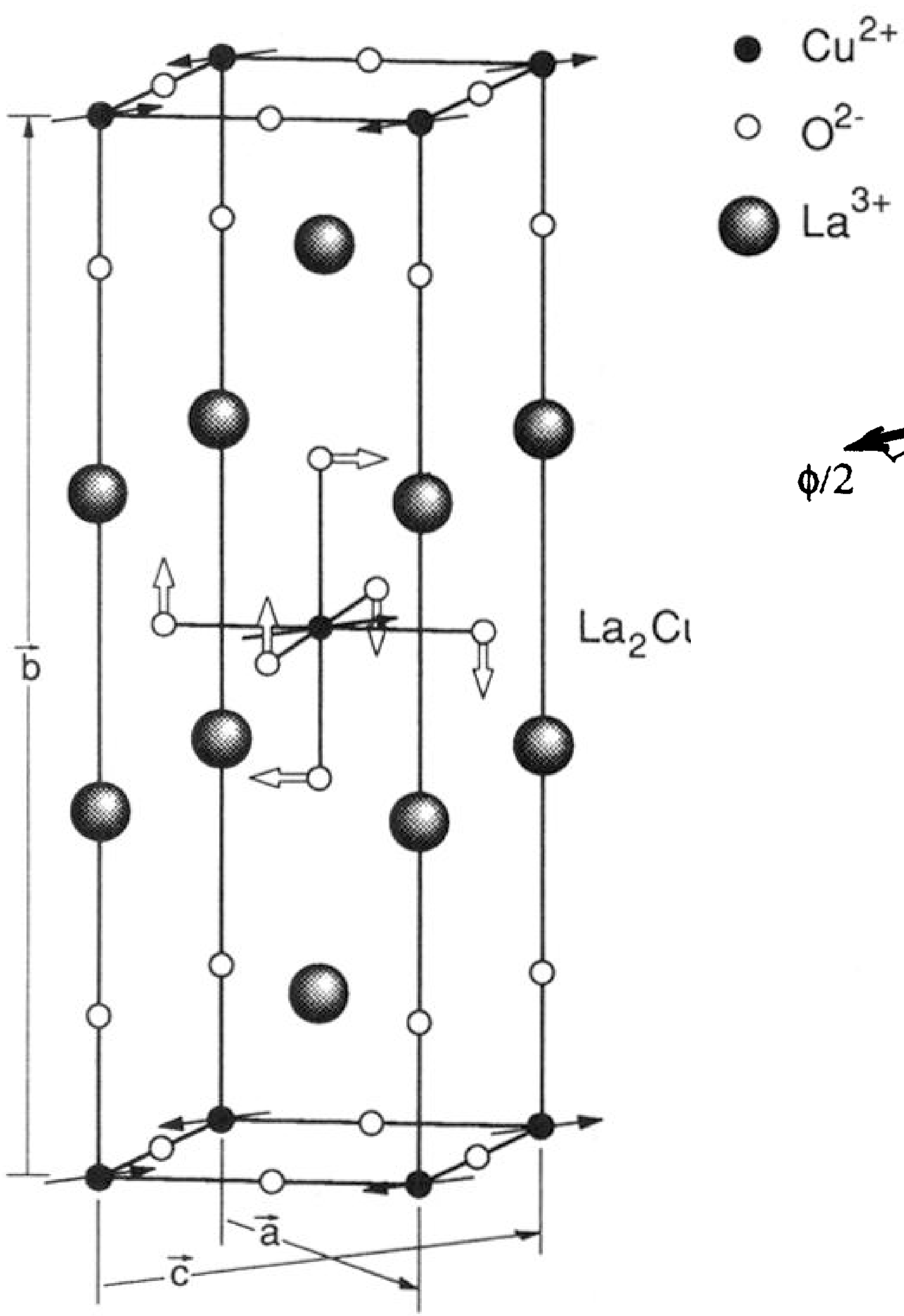
Topological insulators in Bi_2Se_3, Bi_2Te_3 and Sb_2Te_3 with a single Dirac cone on the surface

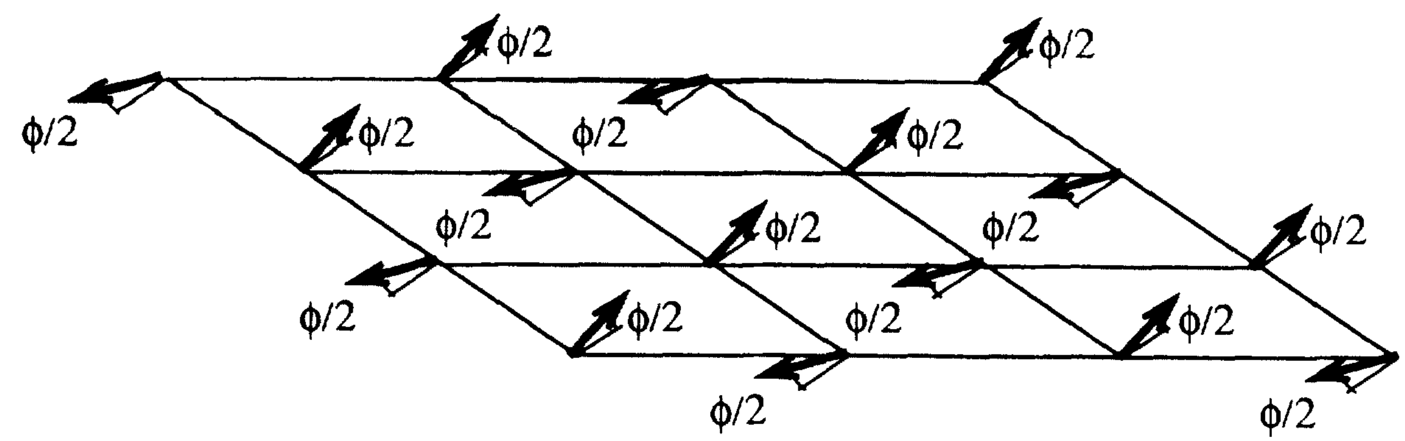




Zhang et al., Nat. Physics (10 May 2009)

Dzyaloshinskii-Moriya interactions in La_2CuO_4



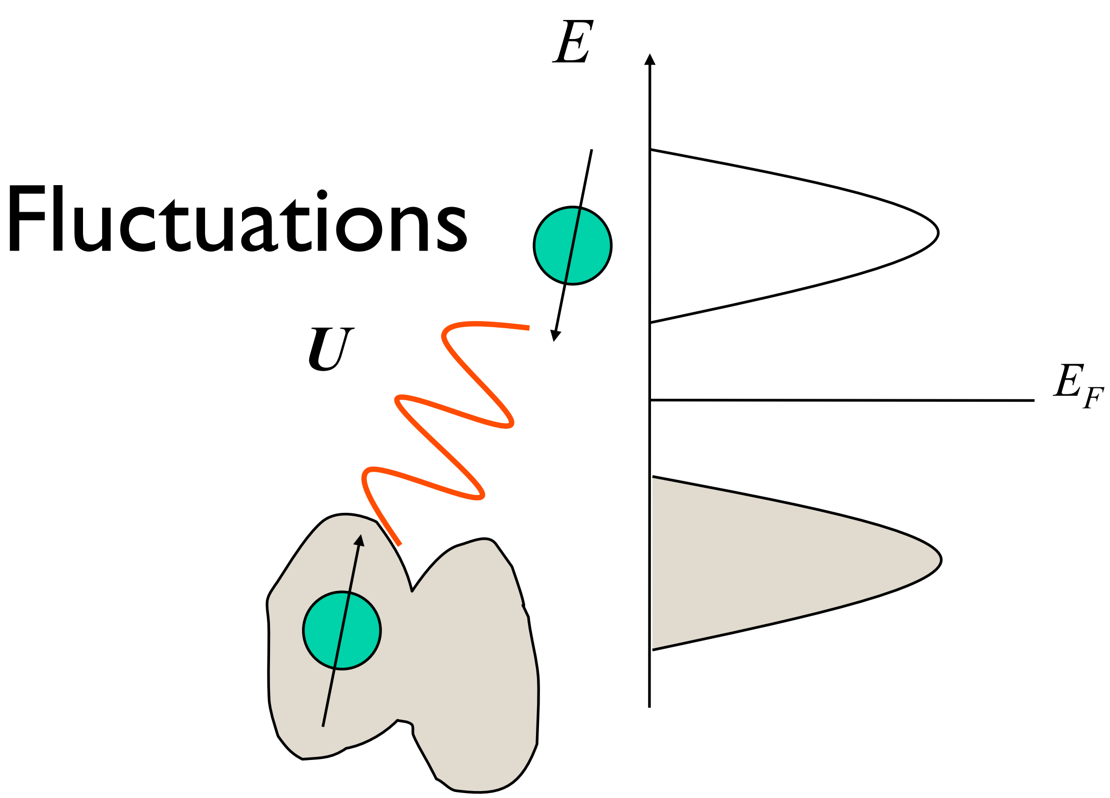


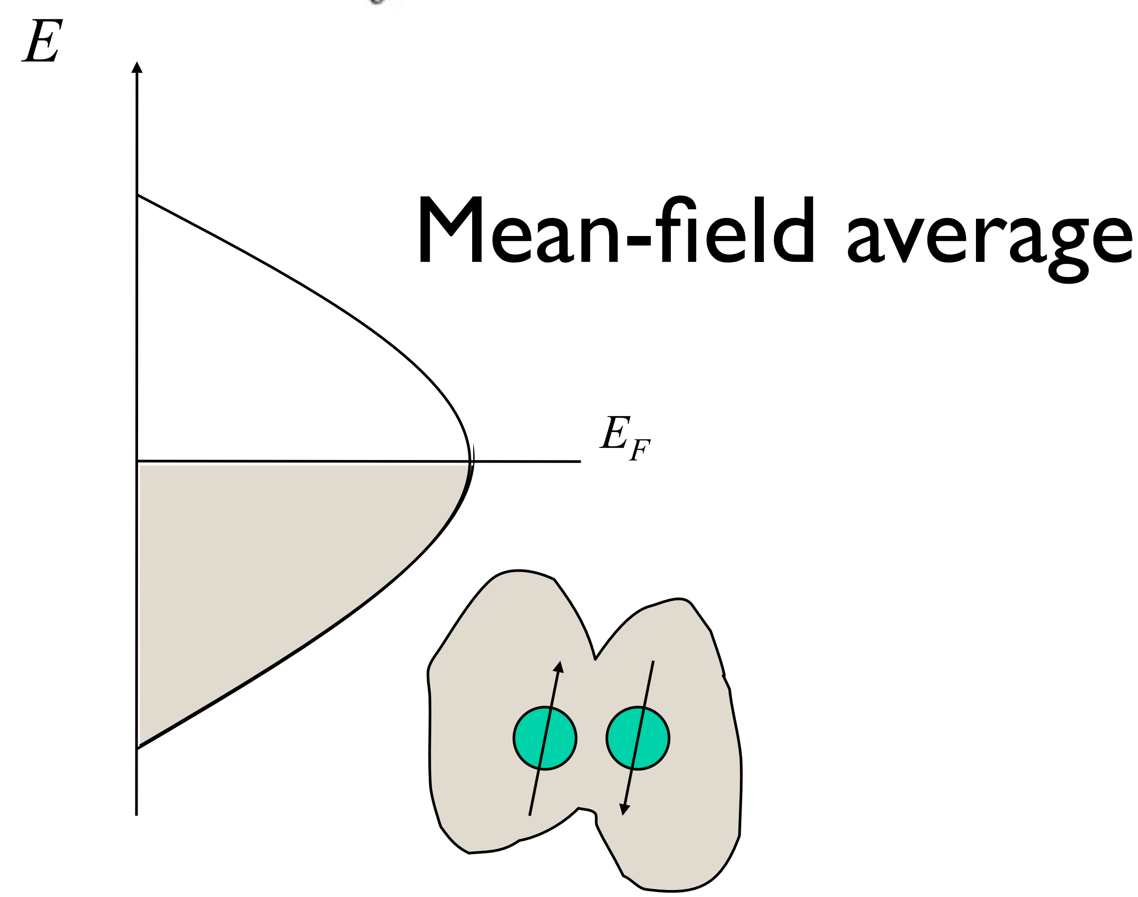
Cheong et al., PRB 39 (1989)

Metal-Insulator Transition

- Hubbard model:

$$H = \sum_{\langle ij \rangle \sigma} t_{ij} c^{+}_{i\sigma} c_{j\sigma} + U \sum_{i} n_{i\uparrow} n_{i\downarrow}$$

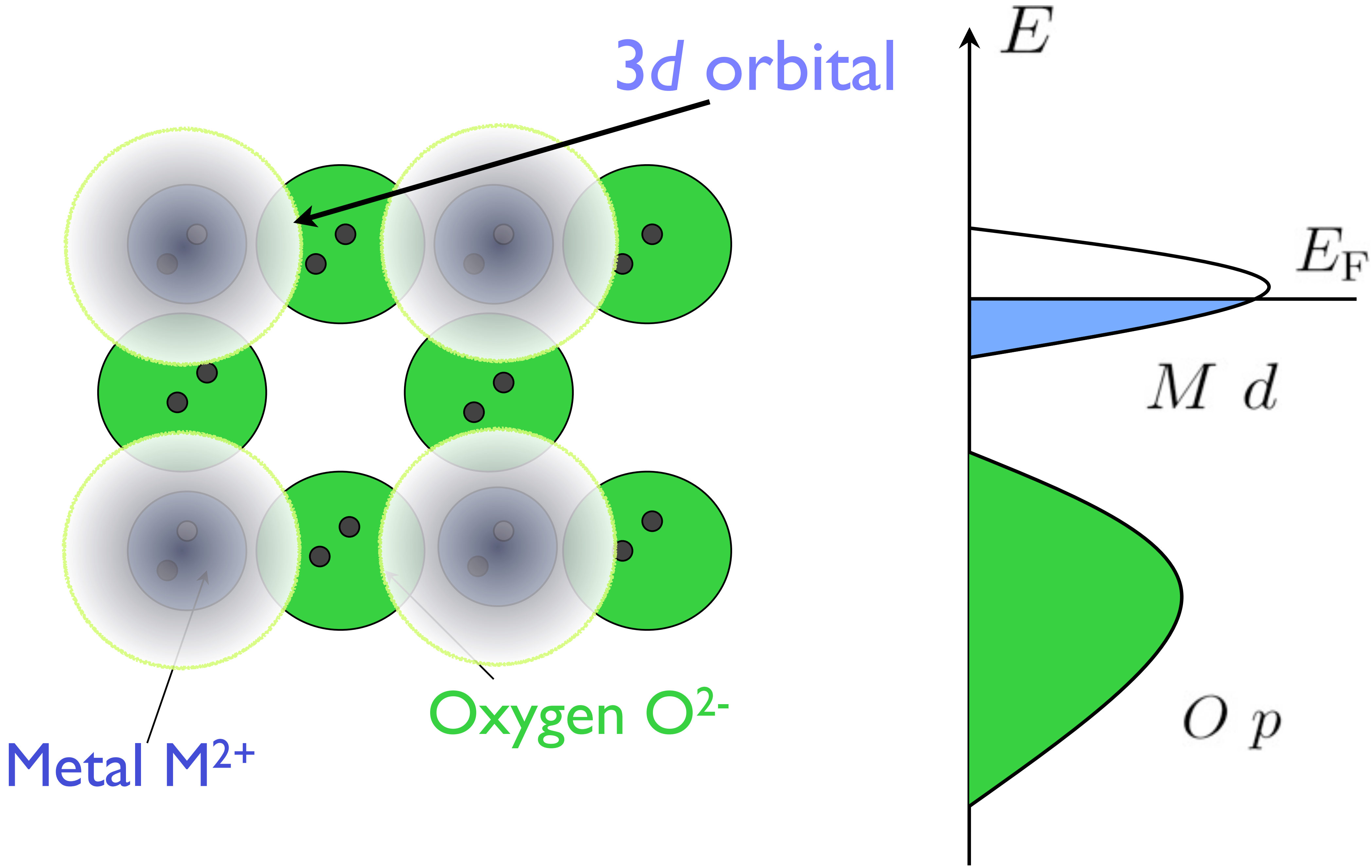




Mott insulator | Band metal

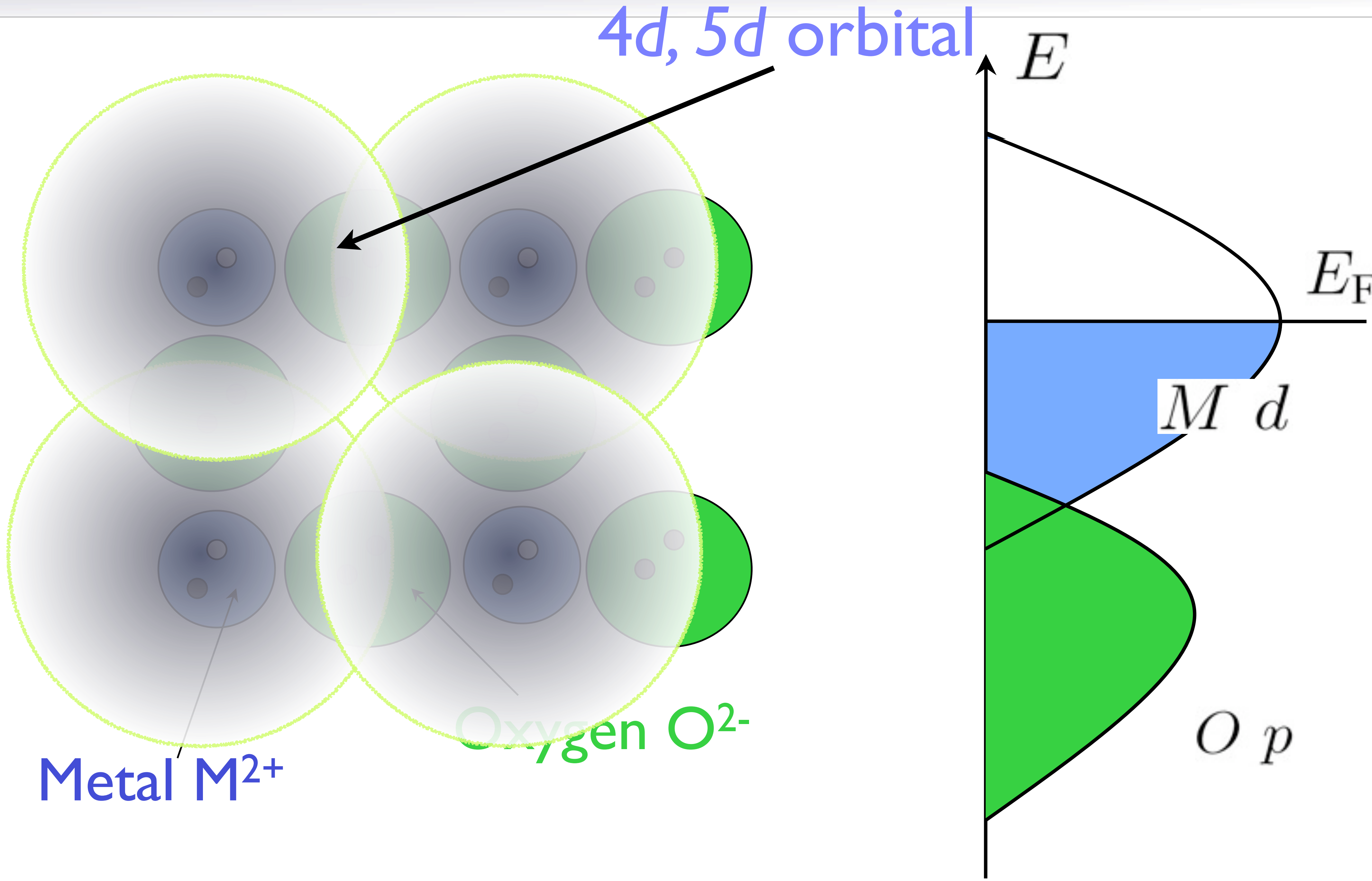
U >> W $\Leftarrow$ $\Rightarrow$ *W>>U*

3*d* transition metal oxides (TMO)



Localized 3*d* orbital → a narrow band!

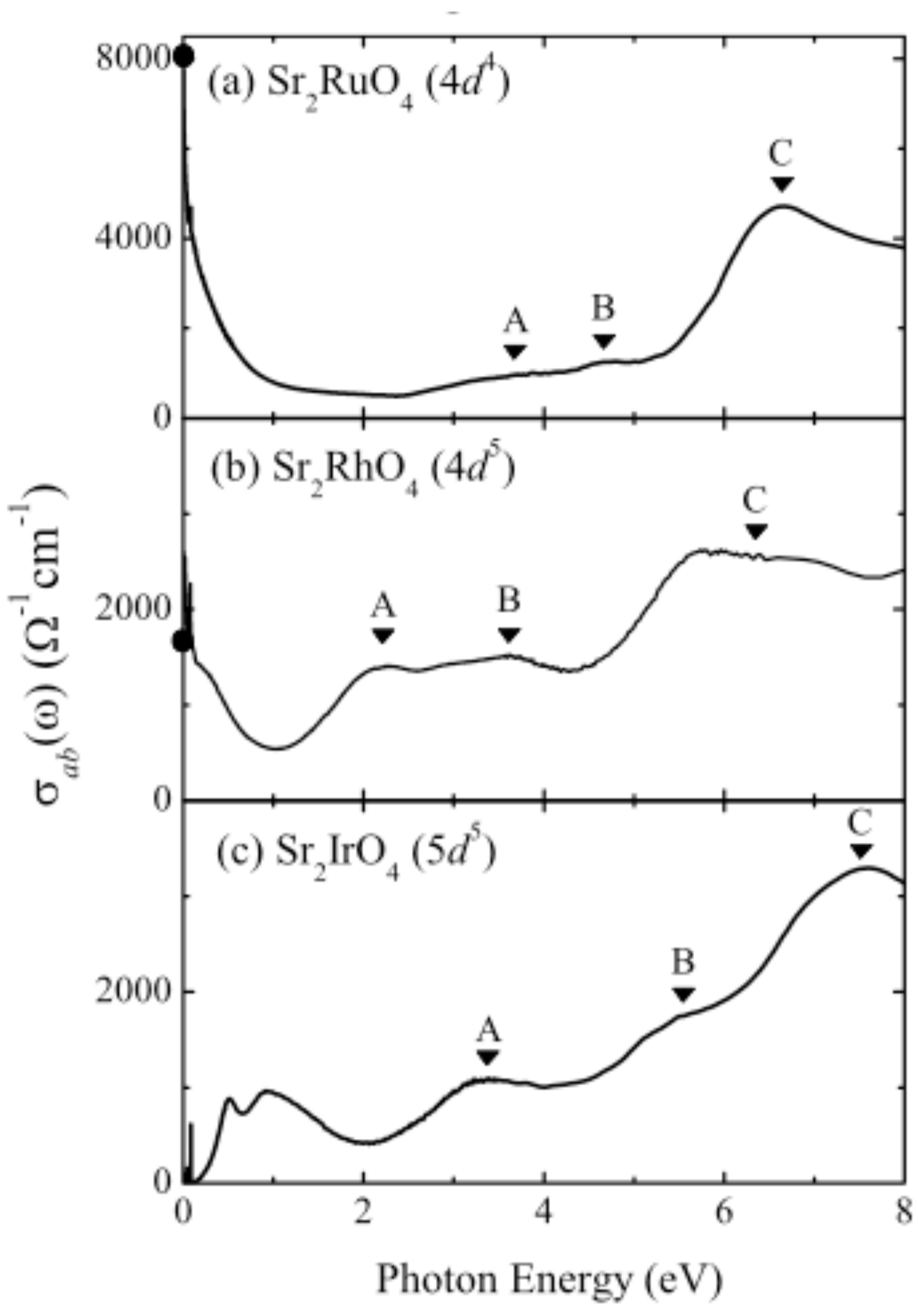
4*d* and 5*d* transition metal oxides

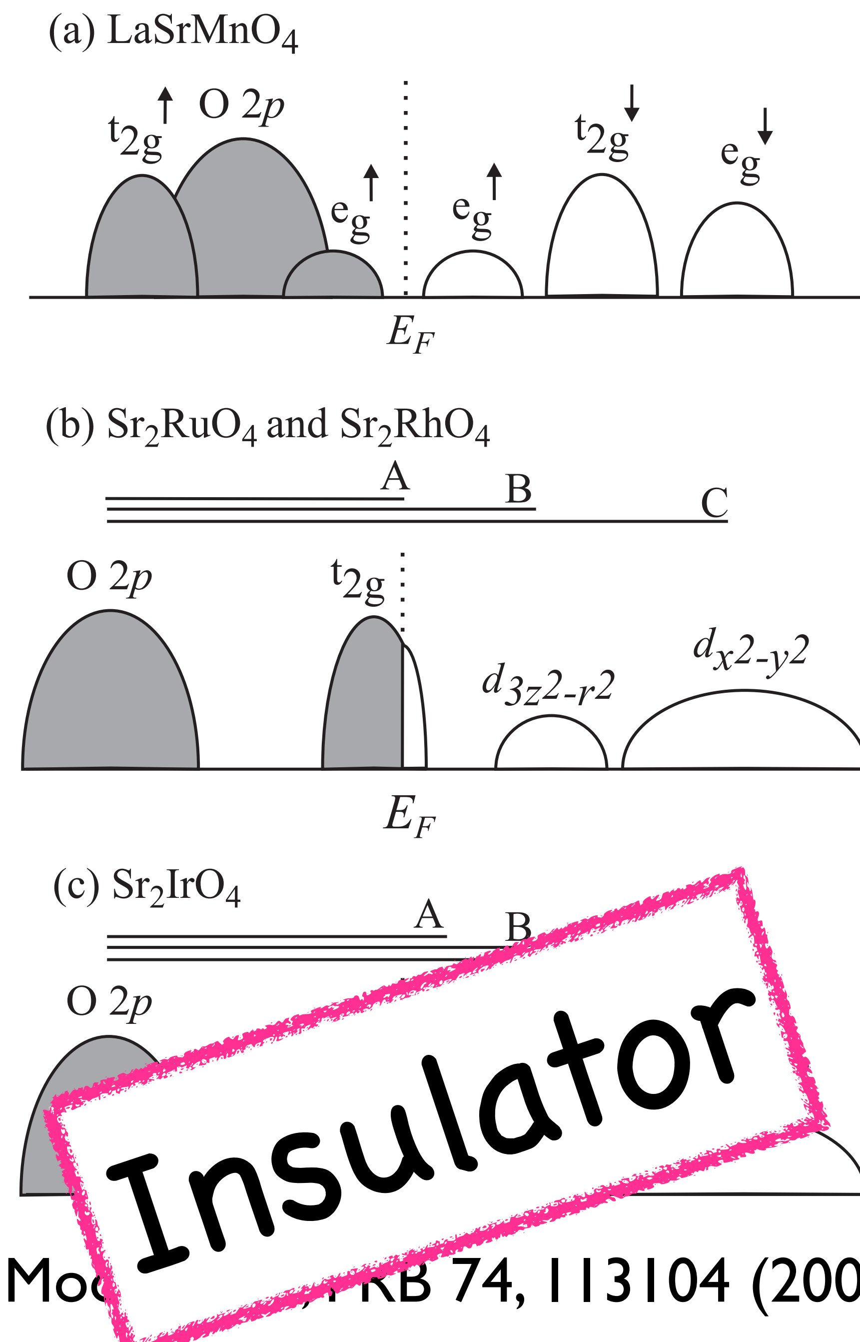


Extended 4*d*, 5*d* orbitals → **wider** band!

4d and 5d transition metal oxides?

For the same K_2NiF_4 structure: La_2CuO_4





3d

⇓

4d

⇓

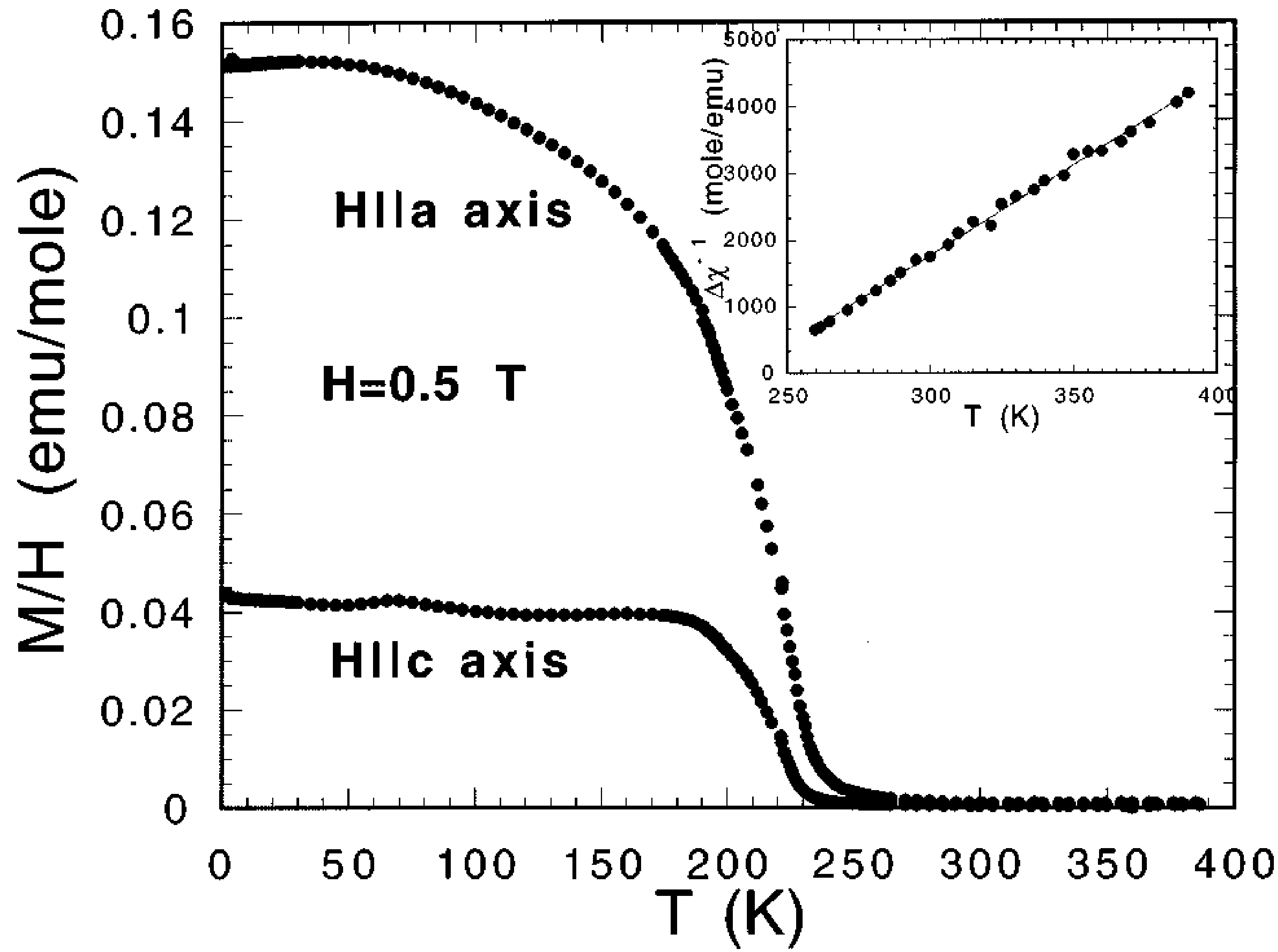
5d

Mo... RB 74, 113104 (2006)

4d, 5d orbitals: more extended → larger band width

→ expect a metallic band structure in Sr_2IrO_4!??

So why is Sr_2IrO_4 insulating?



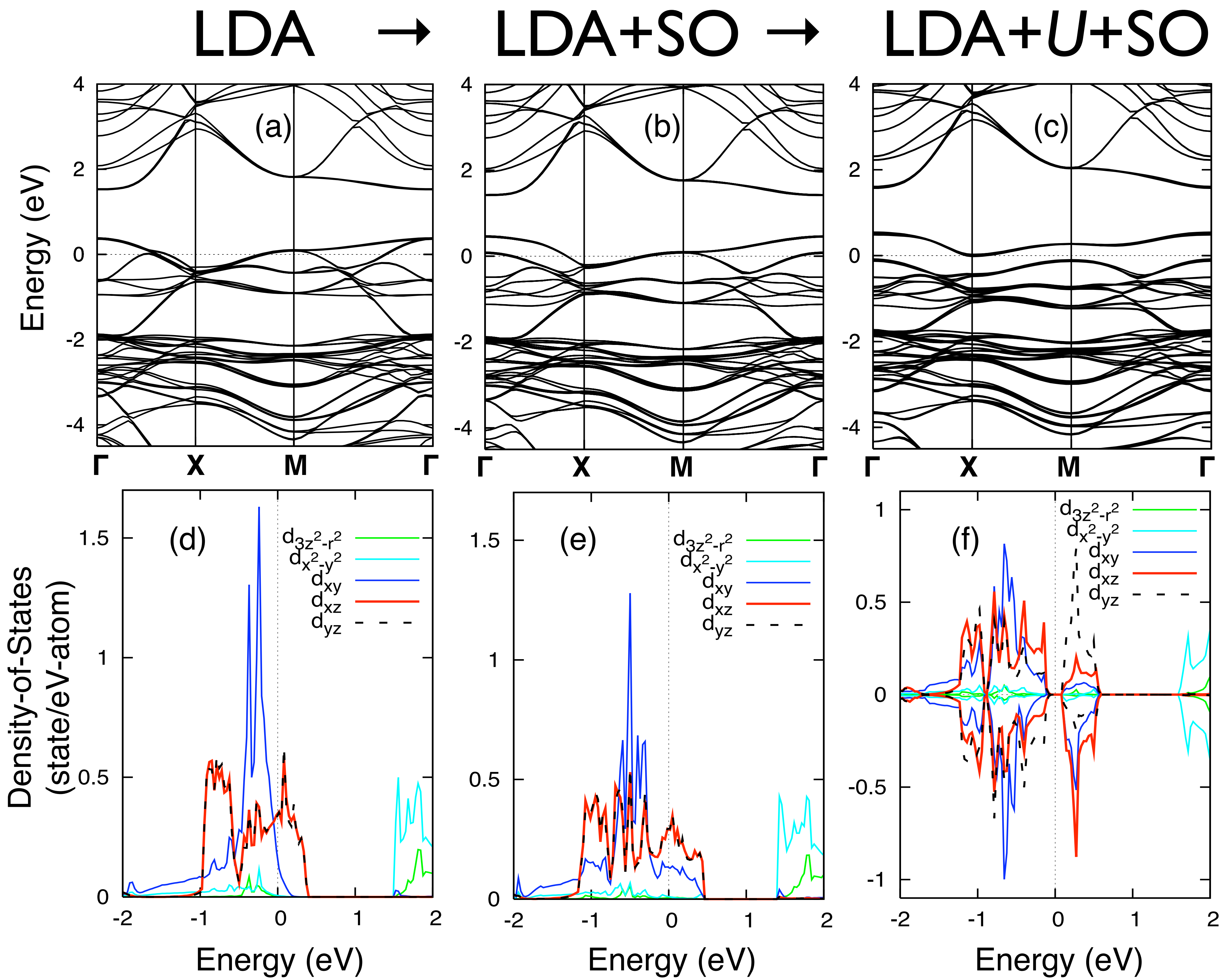
Even more puzzling is that Sr_2IrO_4 exhibits weak ferromagnetism!

Cao et al., PRB 57, R11039 (1998)

Calculation methods

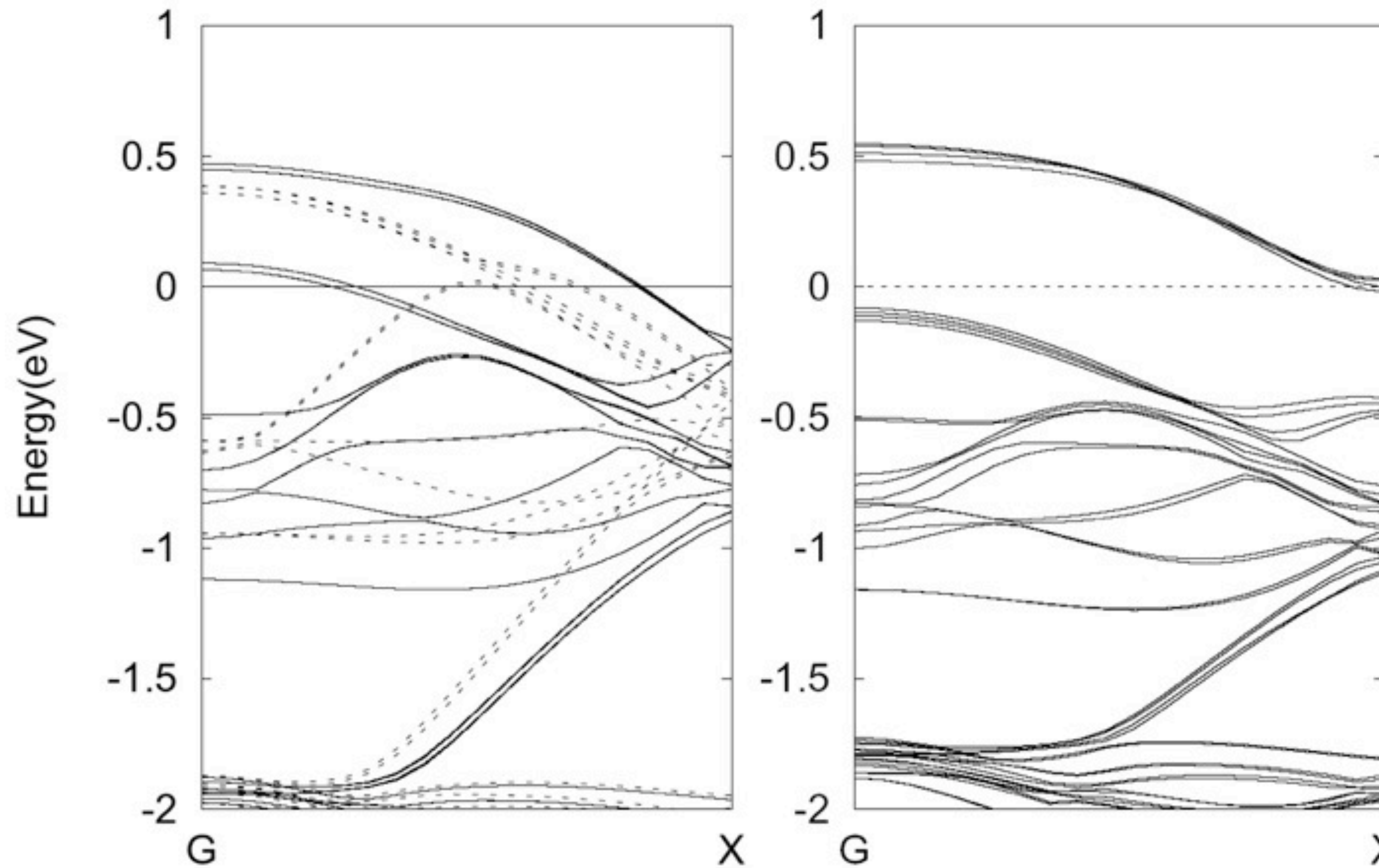
- OpenMX code (http://www.openmx-square.org)
- LDA+*U* methods
- Relativistic pseudo-potential including spin-orbit terms: LDA+*U*+SO calculations
- LDA exchange-correlation potential
- Non-collinear spin configurations

Sr_2IrO_4 band structure



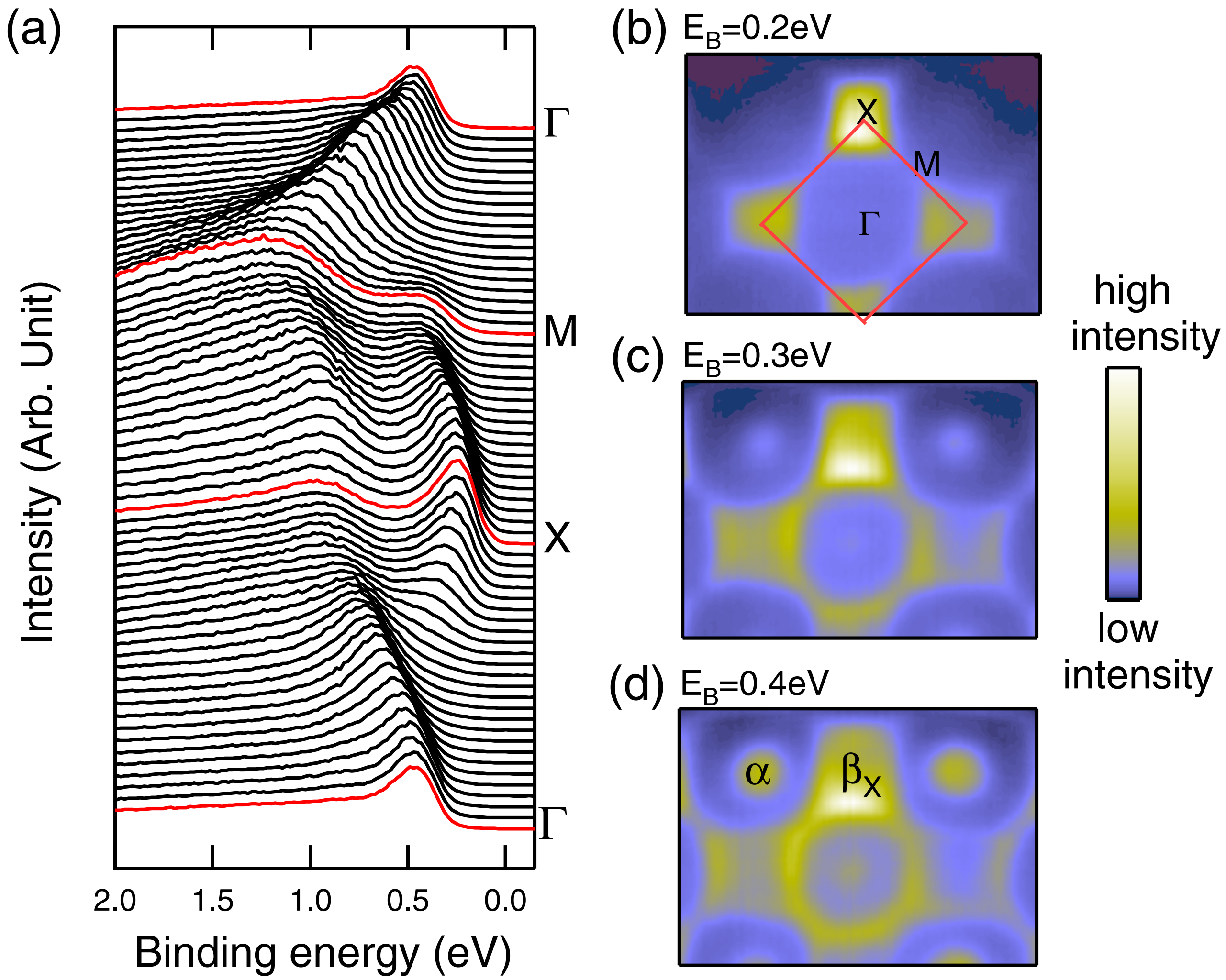
Tight-binding t_{2g} bands of Sr_2IrO_4

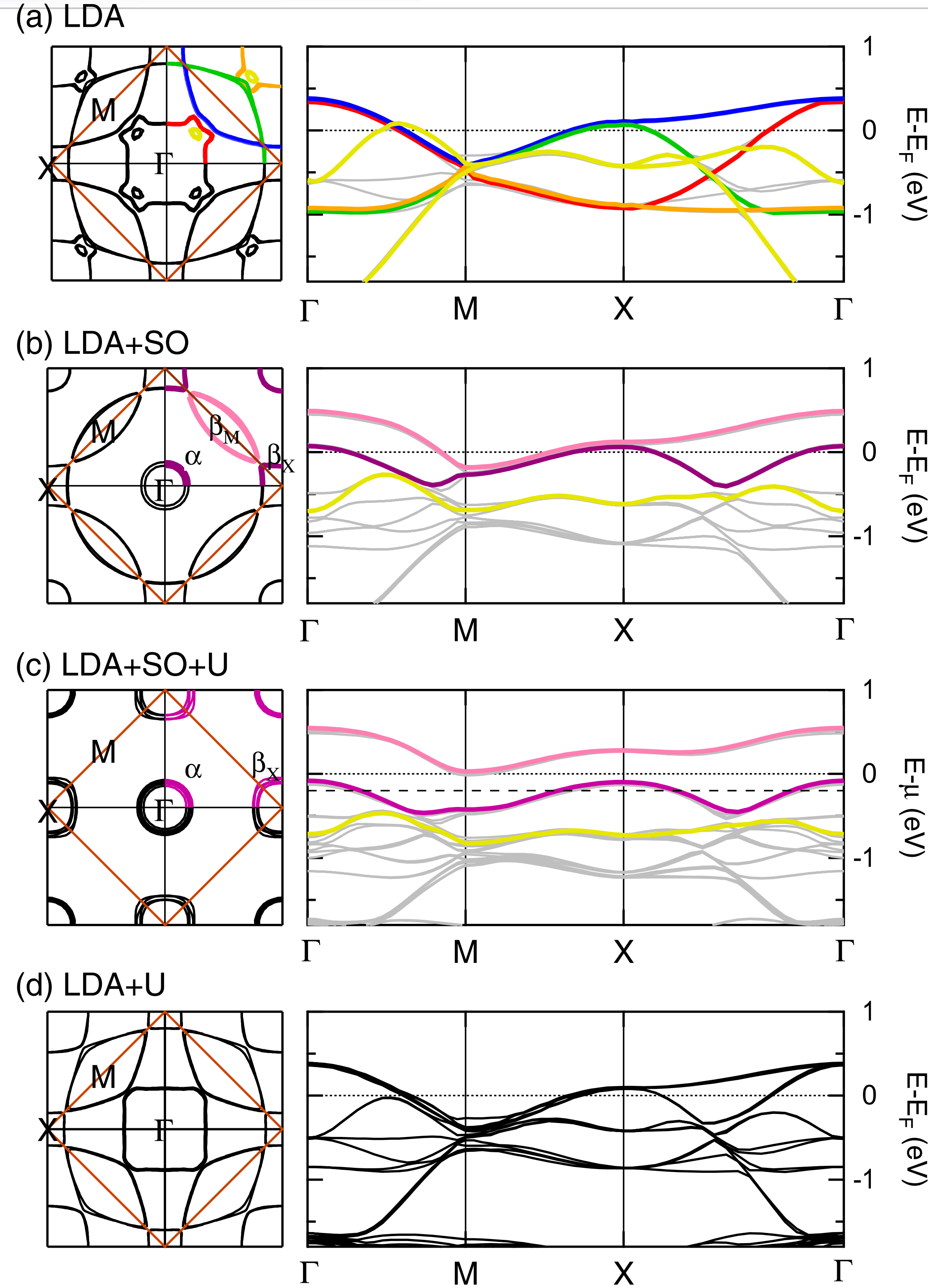
Evolution of t_{2g} bands by SO coupling and on-site U



LDA → LDA+SO → LDA+U+SO

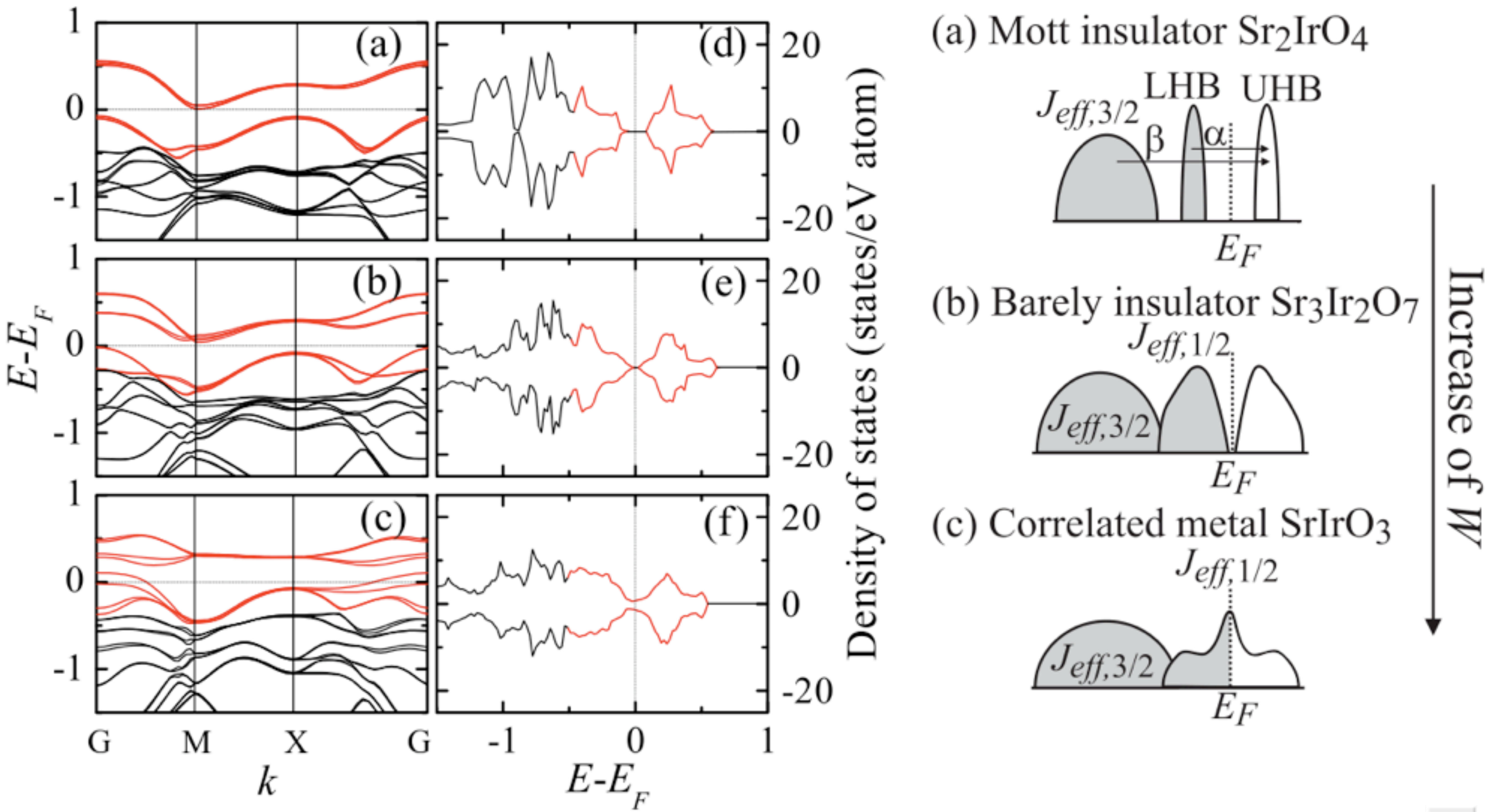
Comparison with ARPES Experiment





Kim et al., PRL 101, 076402 (2008)

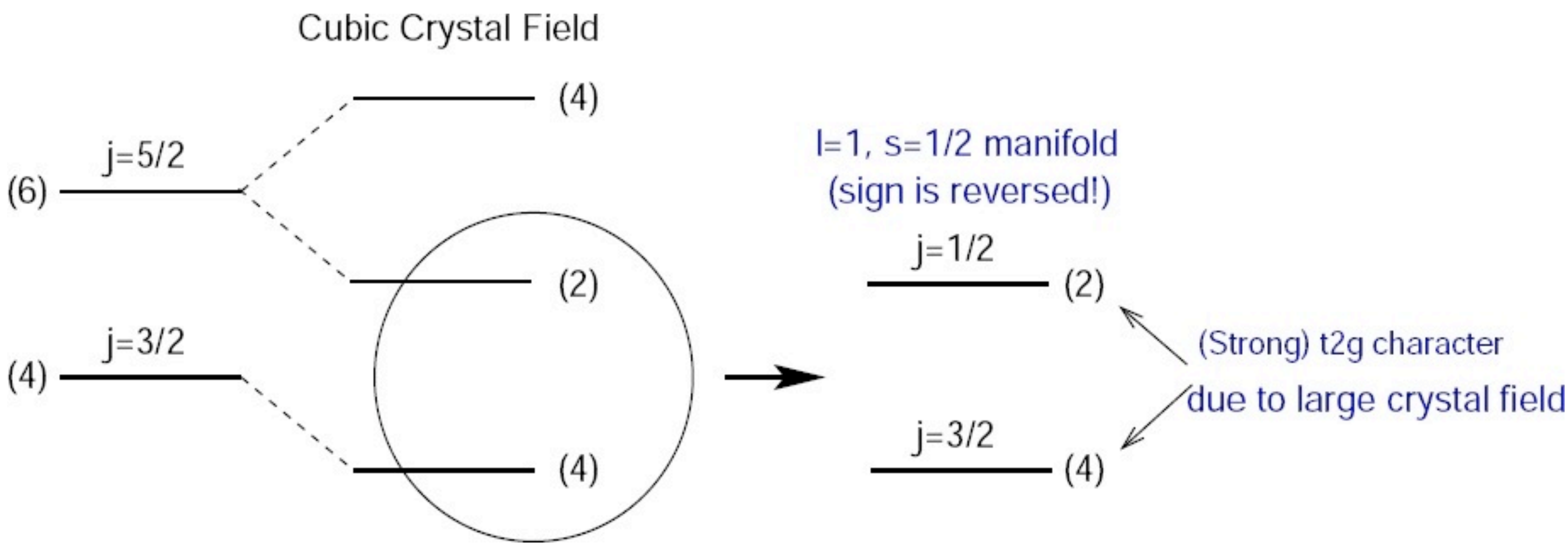
Insulator-to-metal transition in $Sr_{n+1}Ir_nO_{3n+1}$



Moon et al., PRL 101, 226402 (2008)

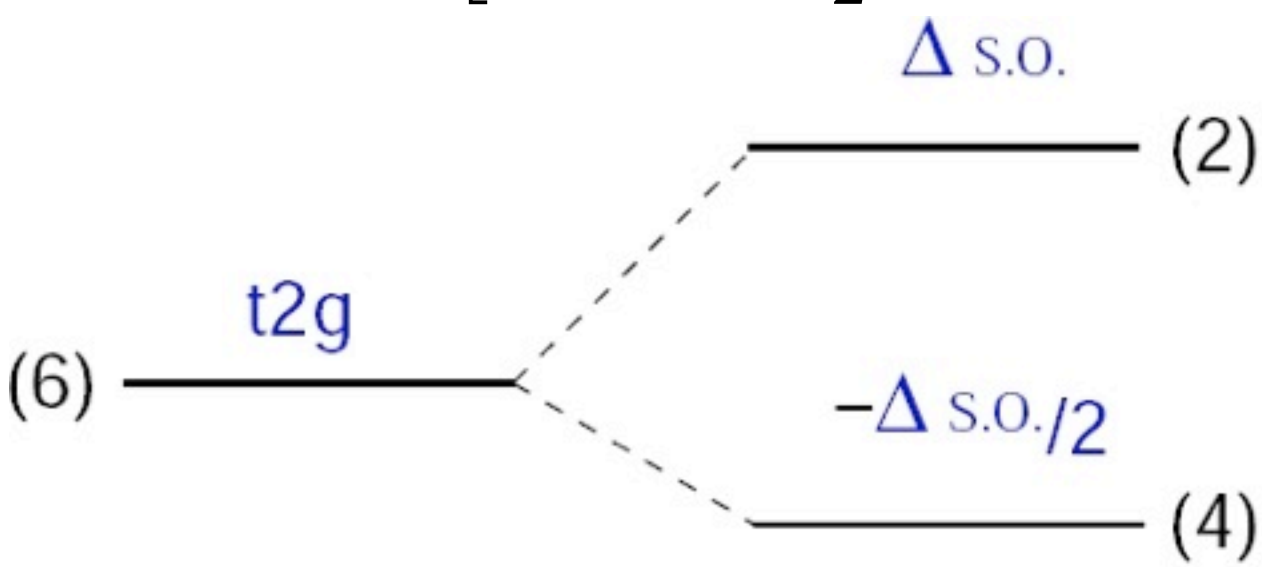
Large λSO coupling in the atomic limit

$$\mathcal{H}_{\mathrm{SO}} = \lambda \mathbf{L} \cdot \mathbf{S} = \lambda \left[L_z S_z + \frac{1}{2}(L_+ S_- + L_- S_+) \right]$$



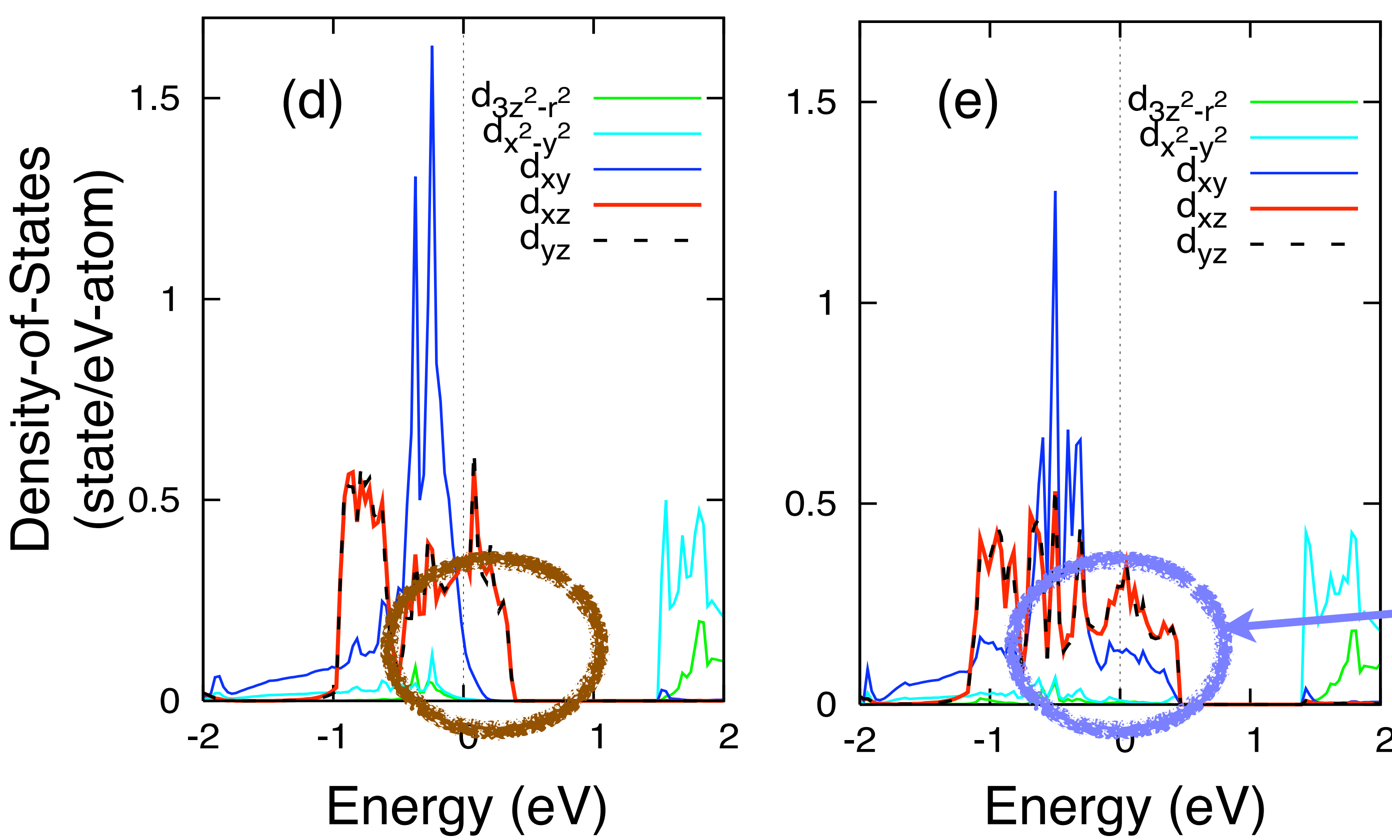
Small λ_{SO} in the band limit

$$\mathcal{H}_{\rm SO} = \lambda \mathbf{L}\cdot\mathbf{S} = \lambda\left[L_z S_z + \tfrac{1}{2}(L_+ S_- + L_- S_+)\right]$$



$|xy,\uparrow\rangle + |yz,\downarrow\rangle + i\,|zx,\downarrow\rangle$,
$|xy,\downarrow\rangle - |yz,\uparrow\rangle + i|zx,\uparrow\rangle$

$|yz,\downarrow\rangle + i|zx,\downarrow\rangle$
$|yz,\uparrow\rangle - i|zx,\uparrow\rangle$
$|xy,\uparrow\rangle - (|yz,\downarrow\rangle + i|zx,\downarrow\rangle)/2$
$|xy,\downarrow\rangle + (|yz,\uparrow\rangle - i|zx,\uparrow\rangle)/2$

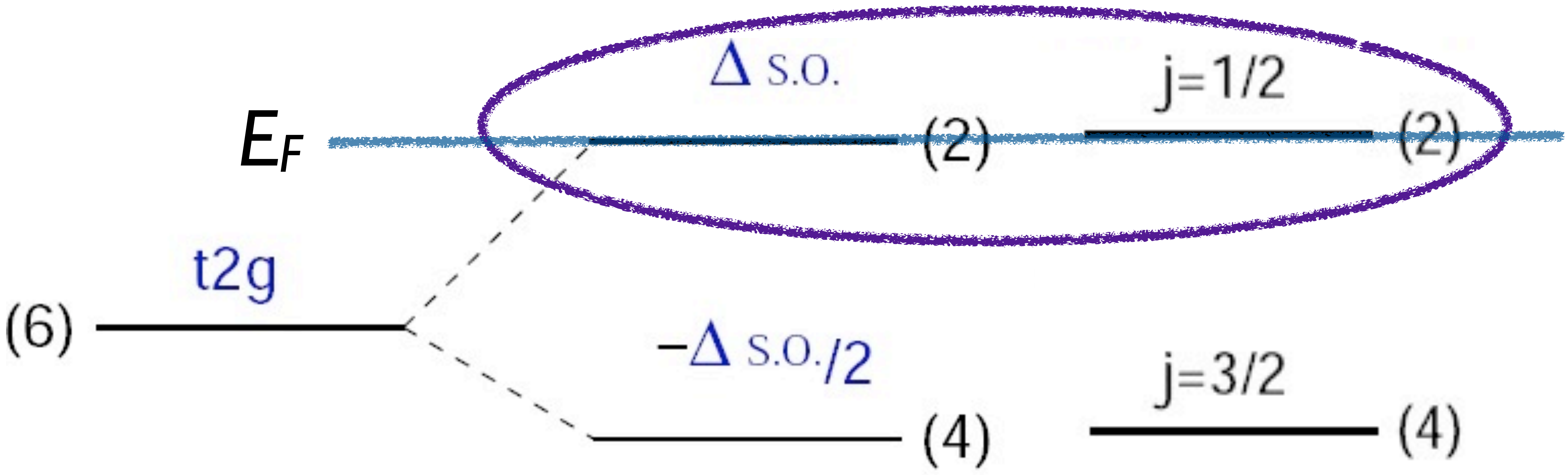


LDA

LDA+SO

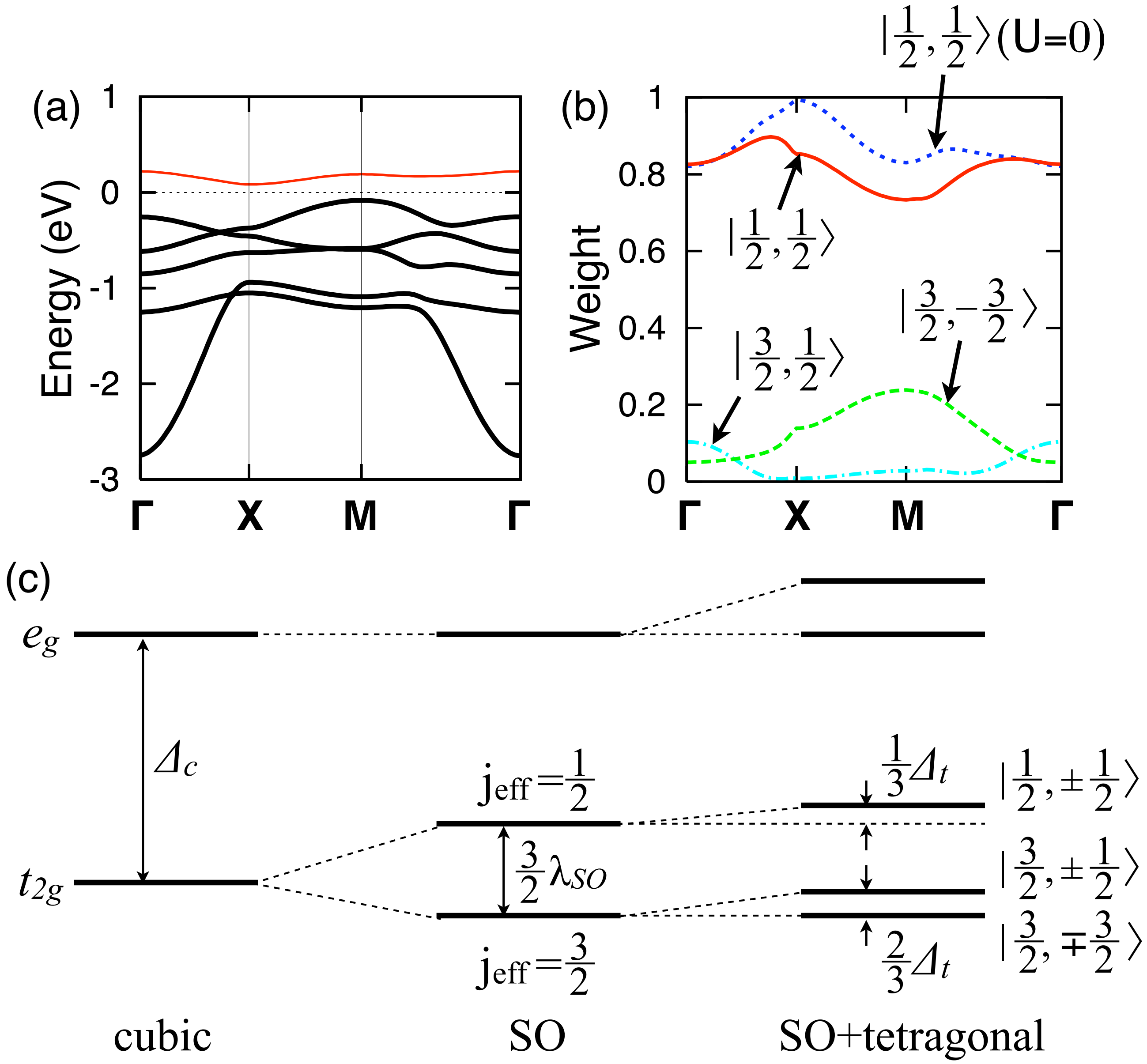
Increase of d_{xy}-component

Localized and Itinerant Pictures

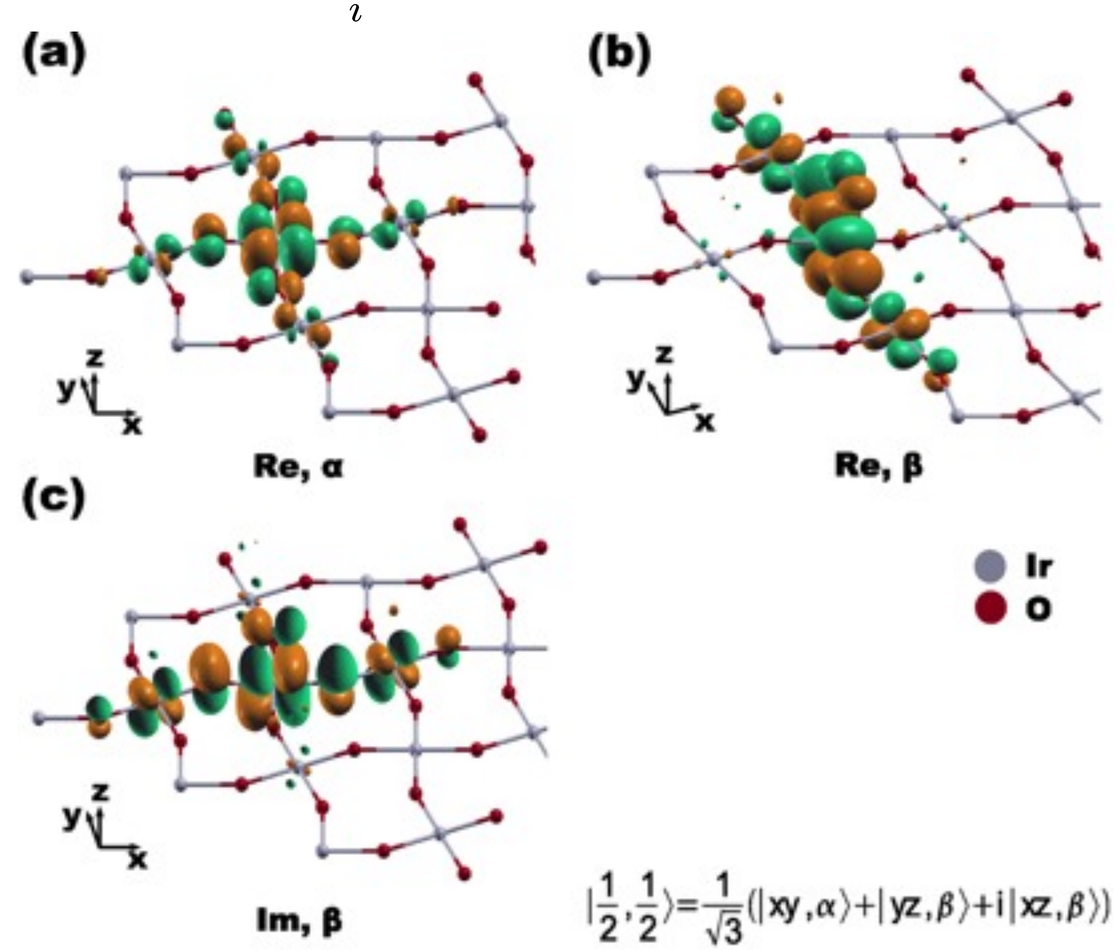


Ir^{4+} d^5

t2g tight-binding model and Wannier functions



$$\mathcal{H} = \sum_{\langle ij\rangle\alpha\beta\sigma} t_{ij}^{\alpha\beta} c_{i\alpha\sigma}^{\dagger} c_{j\beta\sigma} + \frac{1}{2}\sum_{i(\alpha\sigma)\neq(\beta\sigma')} U^{\alpha\beta} n_{i\alpha\sigma} n_{i\beta\sigma'} + \lambda_{\text{SO}}\sum_{i}\mathbf{L}_i\cdot\mathbf{S}_i$$

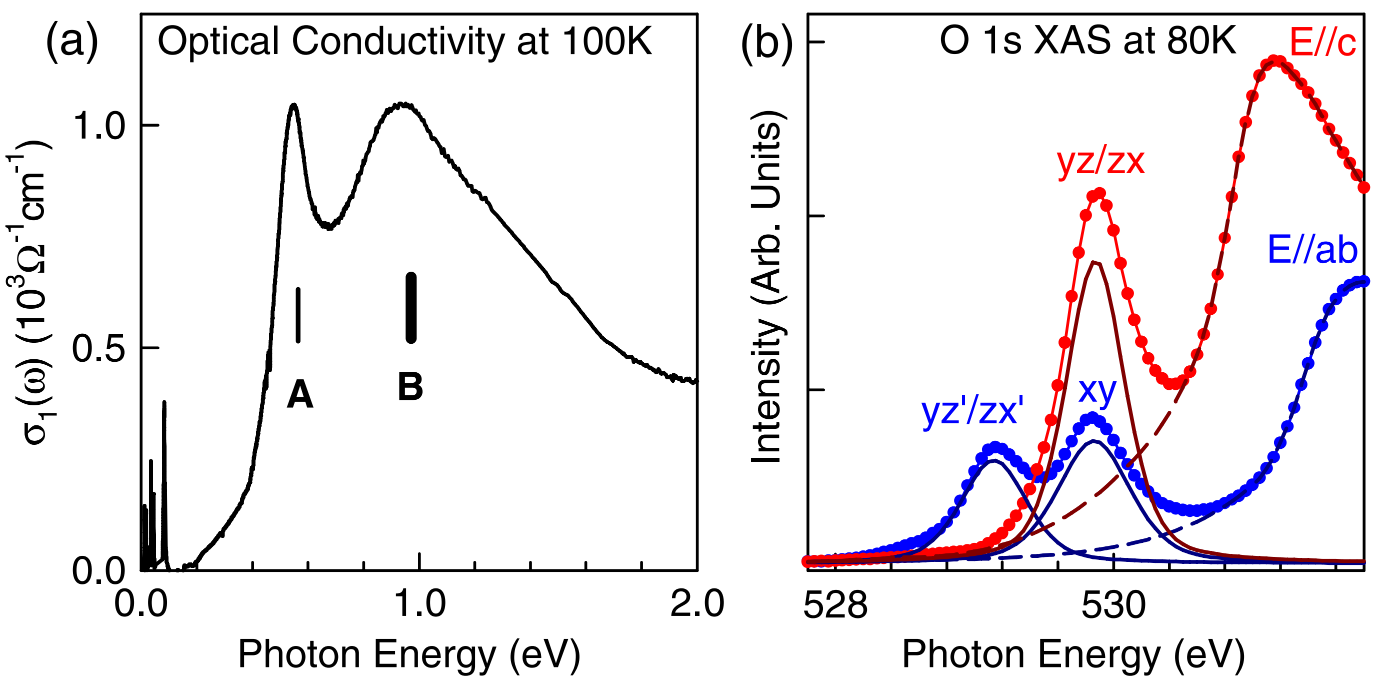


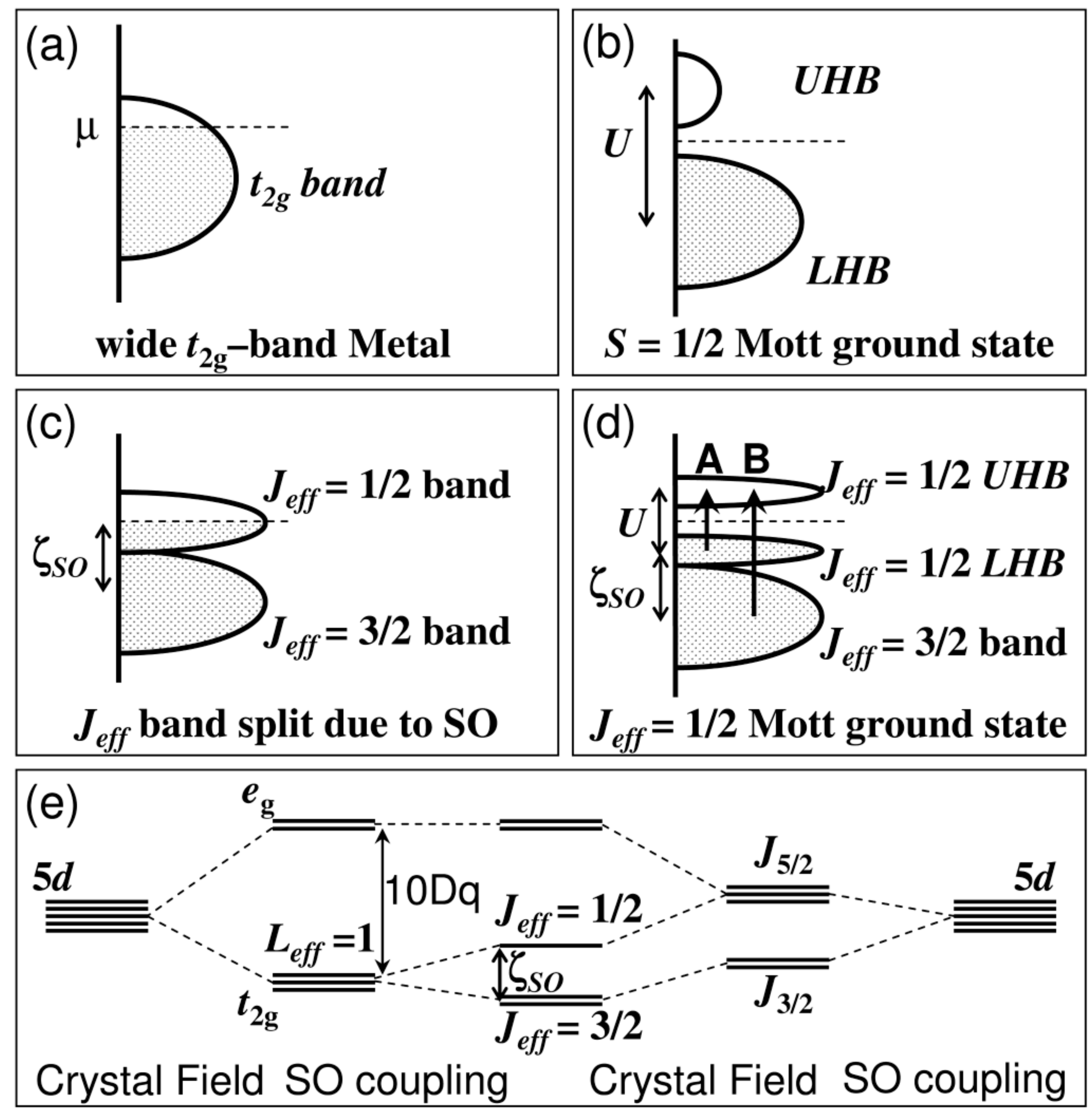
$$|j_{\text{eff}} = 1/2, \pm 1/2\rangle = \mp\frac{1}{\sqrt{3}}|xy\rangle|\pm\rangle - \frac{1}{\sqrt{3}}\left(|yz\rangle \pm i|zx\rangle\right)|\mp\rangle$$

Robust J_{eff}=1/2 spin-orbit integrated state

Sr_2IrO_4: J_{eff}=1/2 Mott insulator

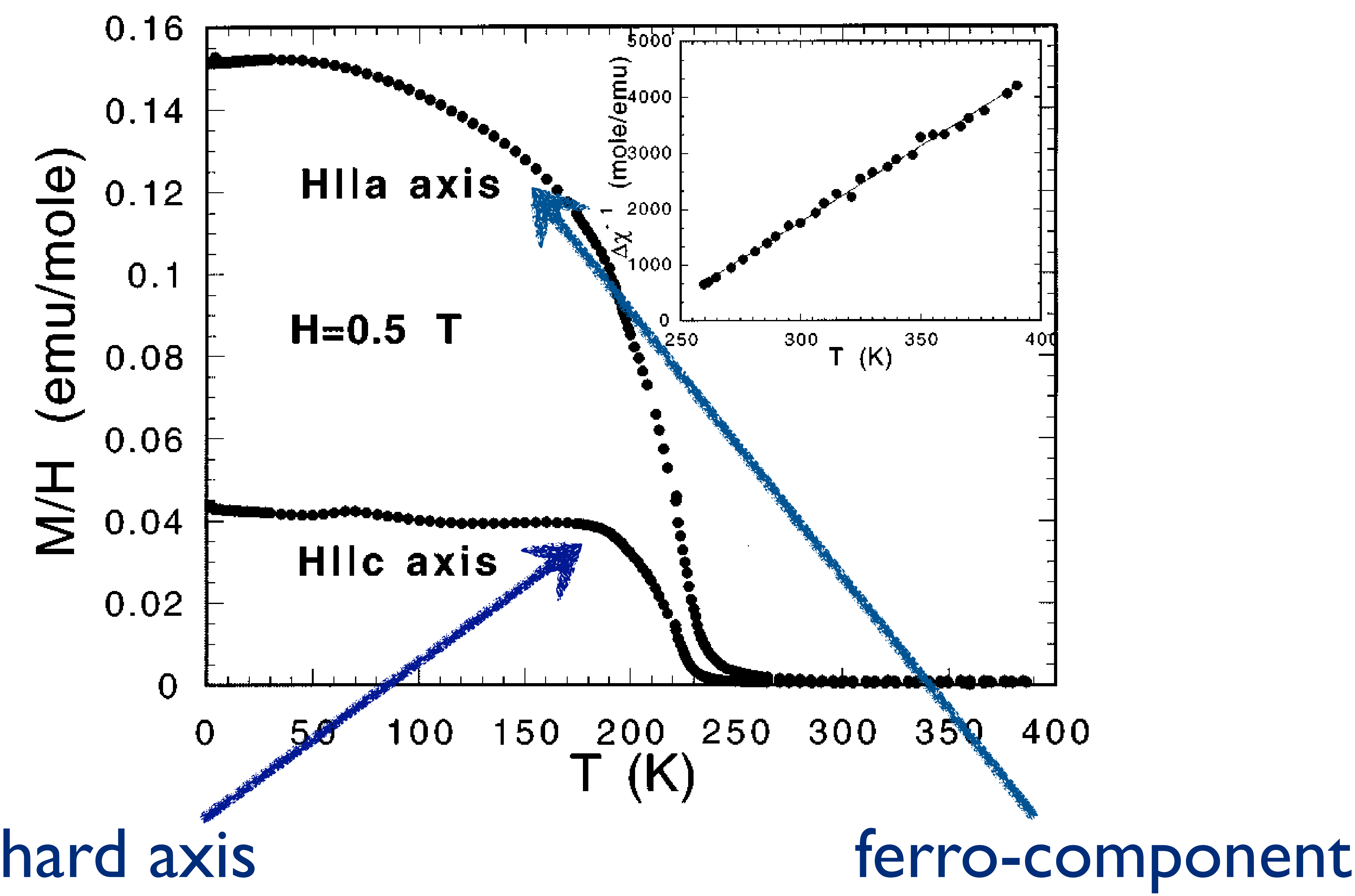
$$|j_{\text{eff}} = 1/2, \pm 1/2\rangle = \mp \frac{1}{\sqrt{3}} |xy\rangle|\pm\rangle - \frac{1}{\sqrt{3}} \left(|yz\rangle \pm i|zx\rangle\right) |\mp\rangle$$





Kim et al., PRL 101, 076402 (2008)

Now what about magnetism in Sr_2IrO_4?

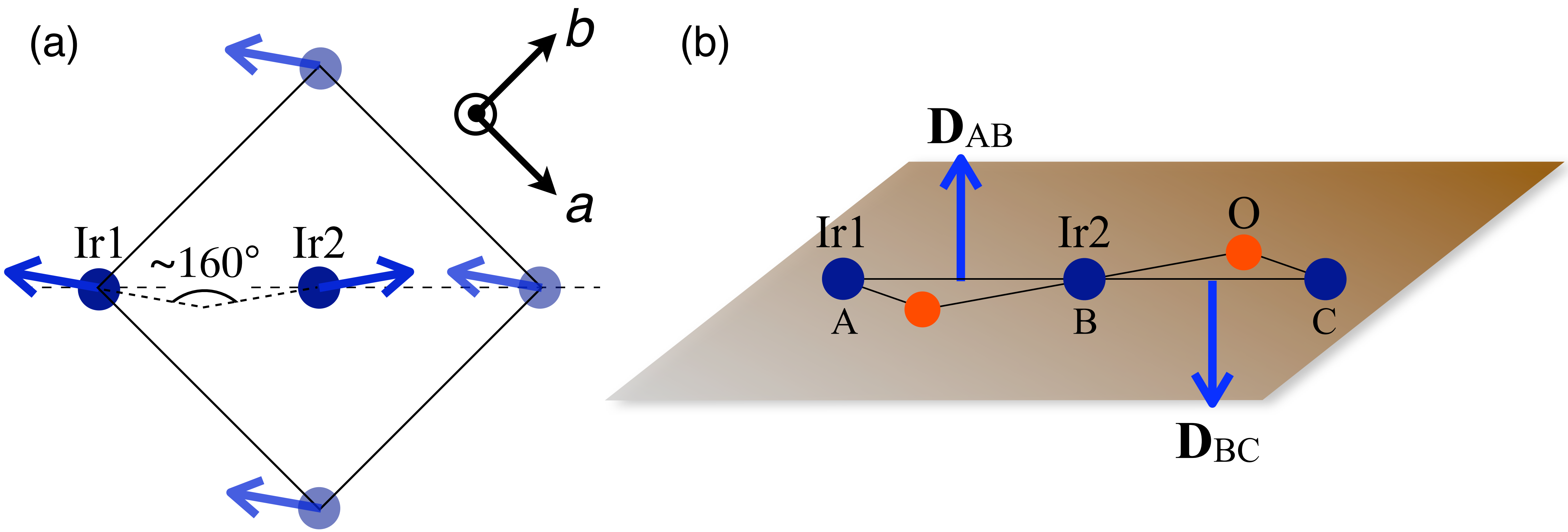


Cao et al., PRB 57, R11039 (1998)

Canted Antiferromagnetic State: Sr_2IrO_4

LDA+U+SO calculation predicts canted AF ordering

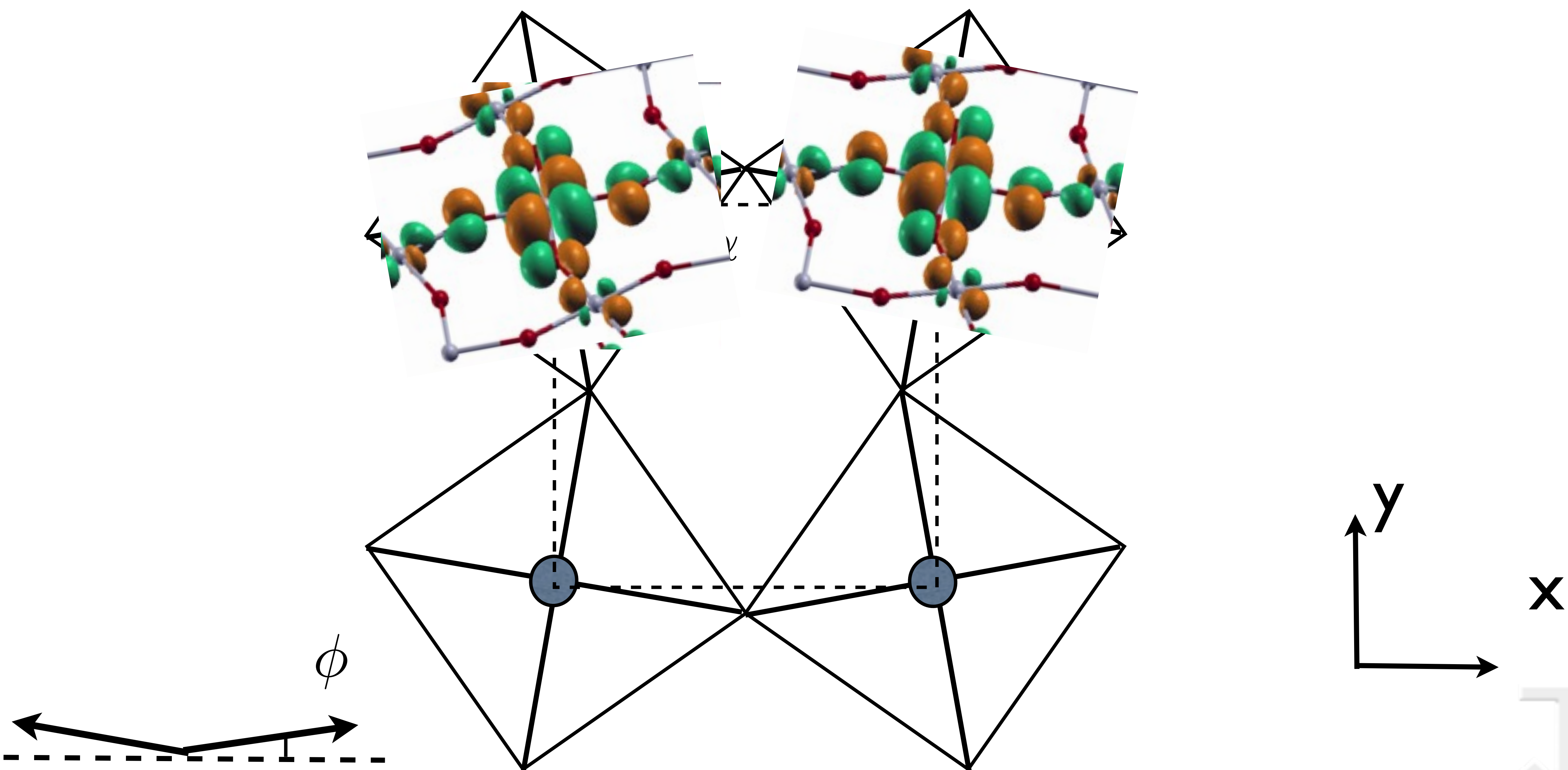
$$m_{\mathrm{AFM}} = 0.36\ \mu_{\mathrm{B}} \qquad m_C = 0.063\ \mu_{\mathrm{B}}$$



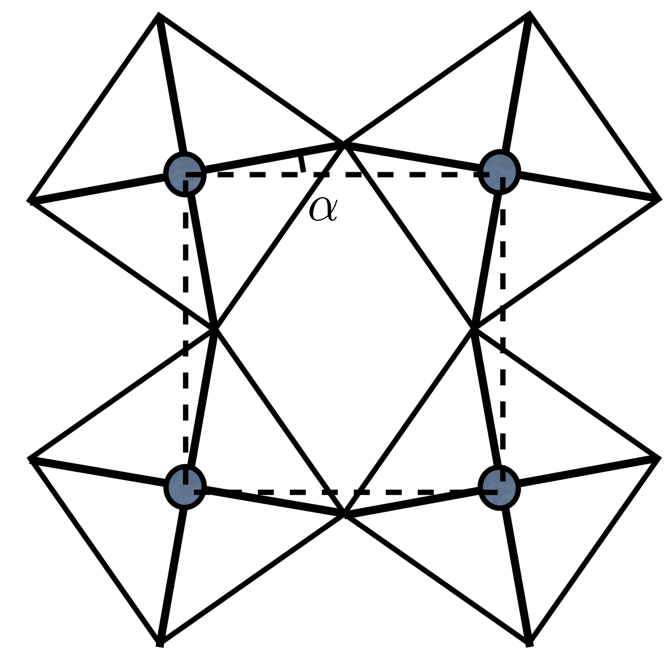
One-band J_{eff}=1/2 Hubbard model for Sr_2IrO_4

$$\mathcal{H} = \sum_{\langle ij \rangle m_j} t_{ij} d^{+}_{im_j} d_{jm_j} + U \sum_i n_{im_j=+1/2} n_{im_j=-1/2}$$

$$|j_{\text{eff}} = 1/2, \pm 1/2\rangle = \mp \frac{1}{\sqrt{3}} |xy\rangle|\pm\rangle - \frac{1}{\sqrt{3}} \left(|yz\rangle \pm i|zx\rangle\right)|\mp\rangle$$



Rotation of IrO_6 octahedron by α



$$\mathcal{H}_{ij} = I_0 \mathbf{J}_i \cdot \mathbf{J}_j + I_1 J_{zi} J_{zj} + \mathbf{D}_{ij} \cdot \mathbf{J}_i \times \mathbf{J}_j$$

$$I_0 = 4(\bar{t}_0^2 - \bar{t}_1^2)/\bar{U} \qquad \mathbf{D}_{ij} = D_z \hat{\mathbf{z}}$$

$$I_1 = 8\bar{t}_1^2/\bar{U} \qquad D_z = 8\bar{t}_0\bar{t}_1/\bar{U}$$

$$\bar{t}_0 \approx \frac{2t_0}{3} \qquad \bar{t}_1 \approx -\frac{2t_0}{3}\sin\alpha$$

Dzyaloshinskii-Moriya interaction: $\left|\frac{D_z}{I_0}\right| \approx \left|\frac{2t_1}{t_0}\right| \sim 2\alpha$

Jin et al., Phys. Rev. B (2009)

Effective Pseudo-Spin Hamiltonian

$$\begin{aligned}\mathcal{H}_{\text{eff}} &= \langle\gamma|\mathcal{H}_{\text{SO}} + \mathcal{H}_{\text{SO}}\frac{1}{\varepsilon - \mathcal{H}'}\mathcal{H}_{\text{SO}} + \ldots|\gamma\rangle \\ &= \sum_{i\mu,j\nu} d_{i\mu,j\nu} J_{i\mu} J_{j\nu} + \ldots\end{aligned}$$

Unquenched orbital degrees of freedom:
J_{eff}=1/2 state

$$\mathcal{H}_{\text{eff}} = \sum_{\langle ij\rangle} \vec{\mathbf{D}}_{ij} \cdot \vec{\mathbf{S}}_i \times \vec{\mathbf{S}}_j$$

Summary

- New form of Mott insulator Sr_2IrO_4:
  spin-orbit entangled j_{eff}=1/2 ground state

  ✓ Strong anisotropic magnetic interactions: Dzyaloshinskii-Moriya interactions driven by the J_{eff}=1/2 state

- Proximity to spin-orbit or topological insulator in Na_2IrO_3

  ✓ Not j_{eff}=1/2 but SO-entangled e_g' state

  ✓ AFM insulator with strong anisotropy

- Both on-site Coulomb and spin-orbit interactions contribute to the non-trivial spin and orbital orderings.