Correlated quantum phenomena in the strong spin-orbit regime

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(24 September 2013)

Outline

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  - Interactions with rare earth moments
  - Issues and Outlook

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- Full degeneracy lifting and honeycomb iridates
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  - Connections to experiments

IV. Concluding Remarks and Outlook

I. Introduction

- Two central threads of quantum materials research
  - Correlated electron physics (e.g. mainly 3*d* transition metal oxides)
    - Local moment formation and magnetism
    - Quantum criticality
    - Unconventional superconductivity
  - Non-trivial physics from strong Spin-Orbit Coupling (SOC)
    - *f*-electron materials
    - Topological insulators and superconductors (*s*- and *p*- orbitals)
- What about systems with correlation + SOC?
  - Heavy Transition Metal Oxides (TMOs) mainly from 5*d* series
  - Both SOC and electronic repulsion strengths, λ and U respectively, become comparable
  - Several arguments suggest that λ and U tend to **cooperate** rather than compete
- A **mean-field** model: Hubbard model with SOC

$$H = \sum_{i,j;\alpha\beta} t_{ij,\alpha\beta}\, c^{\dagger}_{i\alpha} c_{j\beta} + \text{h.c.} + \lambda \sum_{i} \mathbf{L}_i \cdot \mathbf{S}_i + U \sum_{i,\alpha} n_{i\alpha}(n_{i\alpha} - 1)$$

No correlations and no SOC — **with SOC** — **with correlations**

I. Introduction

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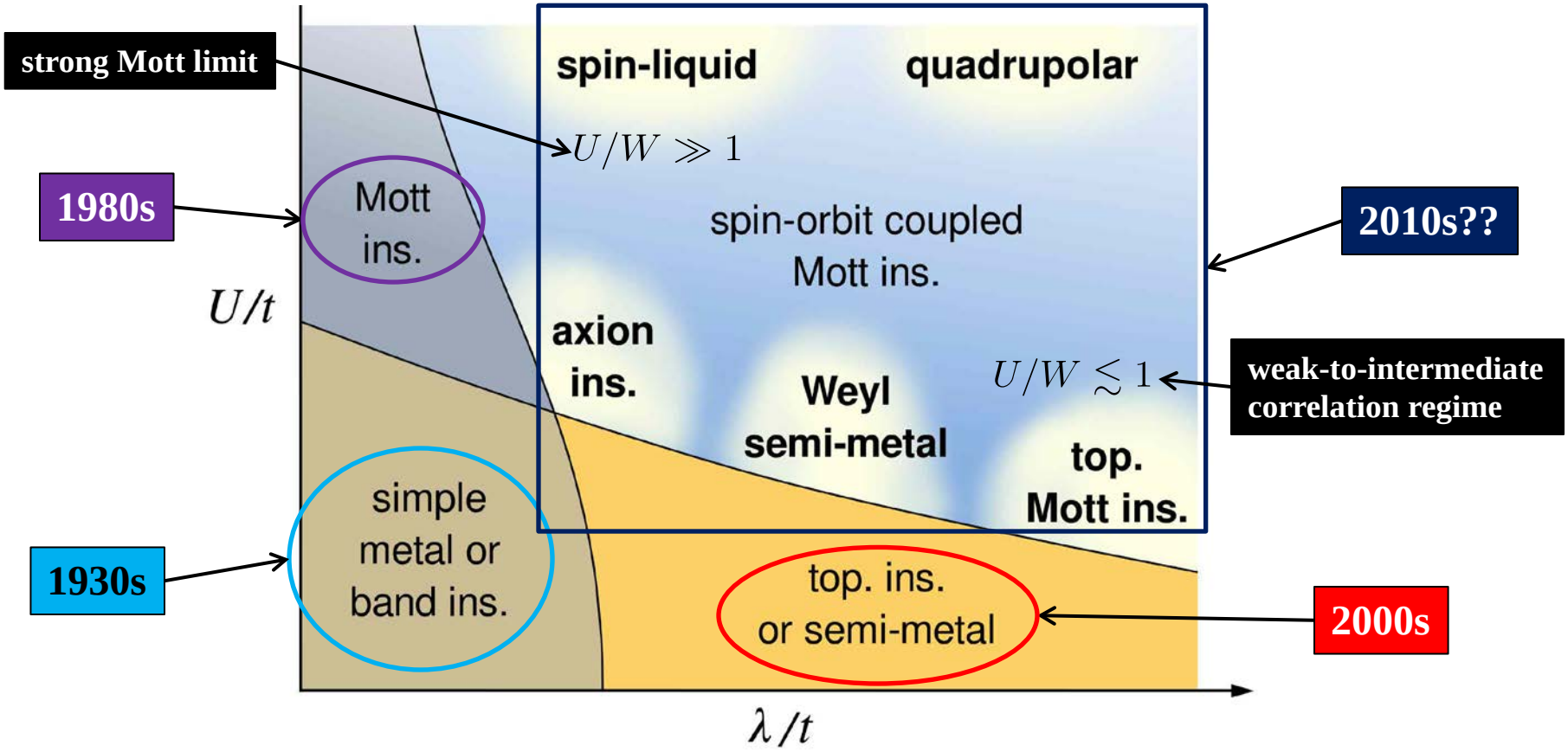

I. Introduction

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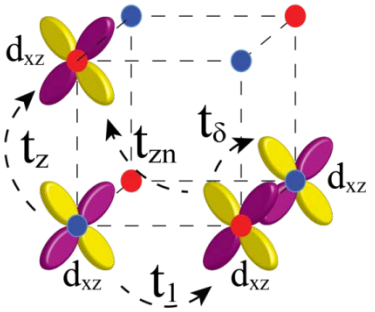
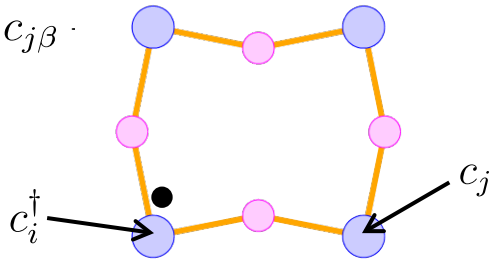
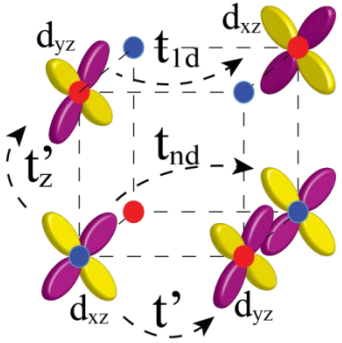


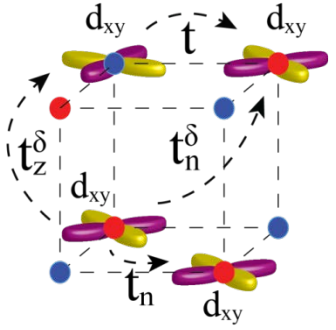
- Consider example: Sr_2IrO_4

$$H = \sum_{i,j;\alpha\beta} t_{ij,\alpha\beta}\, c^{\dagger}_{i\alpha} c_{j\beta}$$

Kinetic energy with which electron hops from site *j* to *i*







- Angular momentum ($\mathbf{L}_i$) and spin ($\mathbf{S}_i$) of electrons on site *i* couple
- Energy cost of repulsion between electrons on the **same** site = *U*
  - One electron **localized** per site
  - The operator $n_{i\alpha} = c^{\dagger}_{i\alpha} c_{i\alpha}$ counts the number of electrons on site *i* in orbital α
  - Last terms kicks in when $n_{i\alpha} \neq 0, 1$

I. Introduction

- Proposals for Iridates

Emergent quantum phases in correlated spin-orbit coupled materials. Abbreviations are as follows: TME = topological magnetoelectric effect, (F)QHE = (fractional) quantum Hall effect. Correlations are W-I = weak-intermediate, I = intermediate (requiring magnetic order, say, but mean field-like), and S = strong.

Phase	Correlation	Properties	Proposed Materials
Axion Insulator	W-I	Magnetic insulator, TME, no protected surface states	$R_2Ir_2O_7$
Weyl semi-metal	I	Dirac-like bulk states, surface Fermi arcs, anomalous Hall	$R_2Ir_2O_7$
Chern Insulator	W-I	Bulk gap, QHE	$SrIrO_3$
Fractional Chern Insulator	I-S	Bulk gap, FQHE	$SrIrO_3$
Fractional Topological Insulator, Topological Mott Insulator	I-S	Several possible phases. Charge gap, fractional excitations	$R_2Ir_2O_7$
Quantum spin liquid	S	Several possible phases. Charge gap, fractional excitations	Na_2IrO_3

II. Weak to Intermediate Correlations

- Topological insulators: non-trivial topology of the **bands** in a **gapped** system
- Gapless systems: Weyl semi-metals (WSMs)

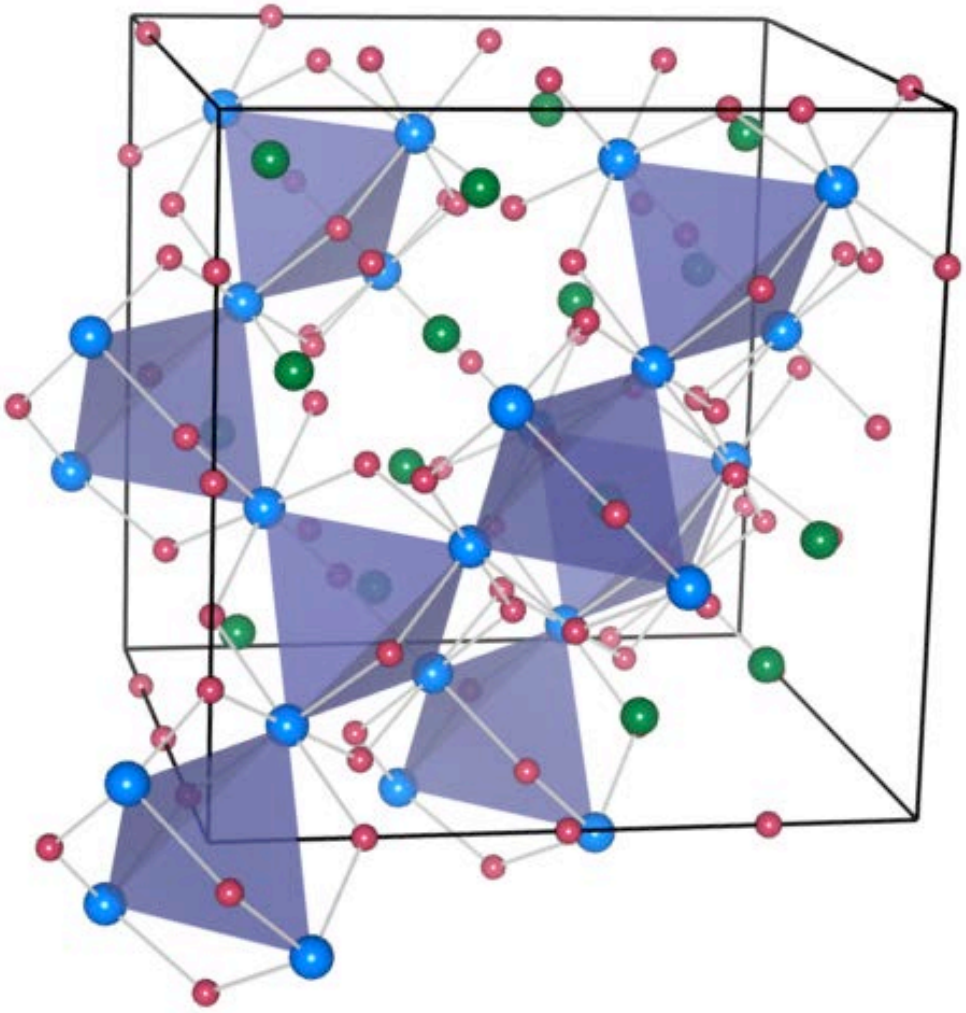
$$\mathcal{H}(\mathbf{k}) = \pm v(\delta k_x \sigma_x + \delta k_y \sigma_y + \delta k_z \sigma_z)\,, \quad \delta\mathbf{k} = \mathbf{k} - \mathbf{k}_W$$

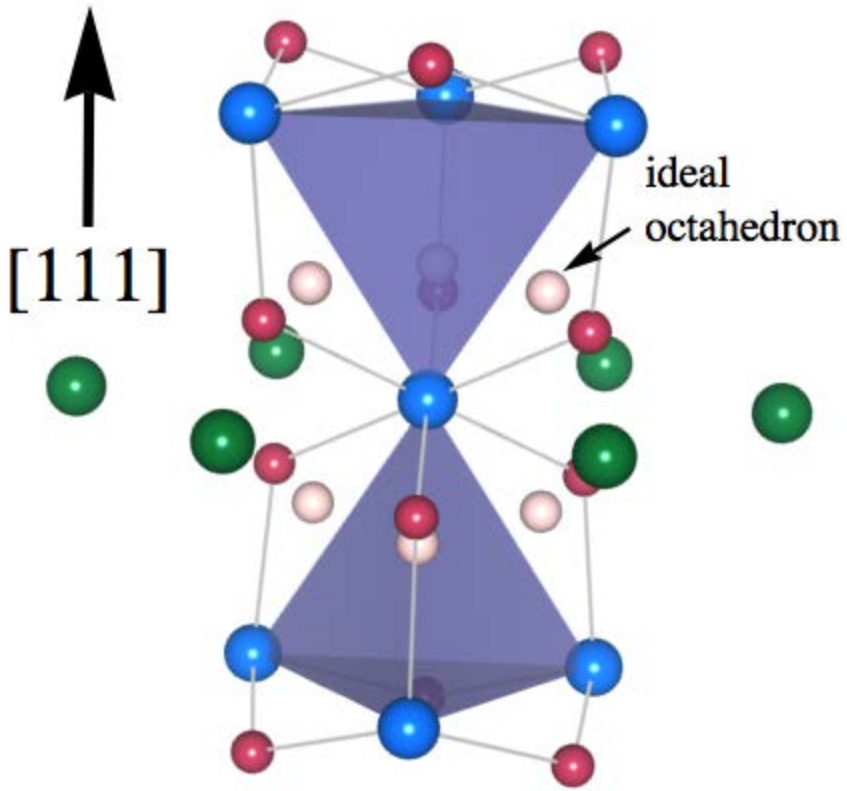
- Notion of band topology ➔ some degree of itinerancy
- Non-TI, but still topological phases, require: *intrinsic* symmetry breaking
- Any form of intrinsic magnetization ➔ correlations “weak” enough for mean-field
- Examples of non-TI topological phases:
  - Antiferromagnetic Topological Insulator (AFTI)
  - Axion Insulator
  - Weyl semi-metal
- Strong Mott regime ➔ electrons atomically localized; “band” topology doesn’t make sense
- Exotic phases due to orbital- and spin-ordering when both are entangled
- The spin + orbit *entanglement* lifts degeneracies of the ground states to give interesting lattice models

II. Weak to Intermediate Correlations

A. Pyrochlore iridates

- Formula: $R_2Ir_2O_7$ where R is a rare earth element



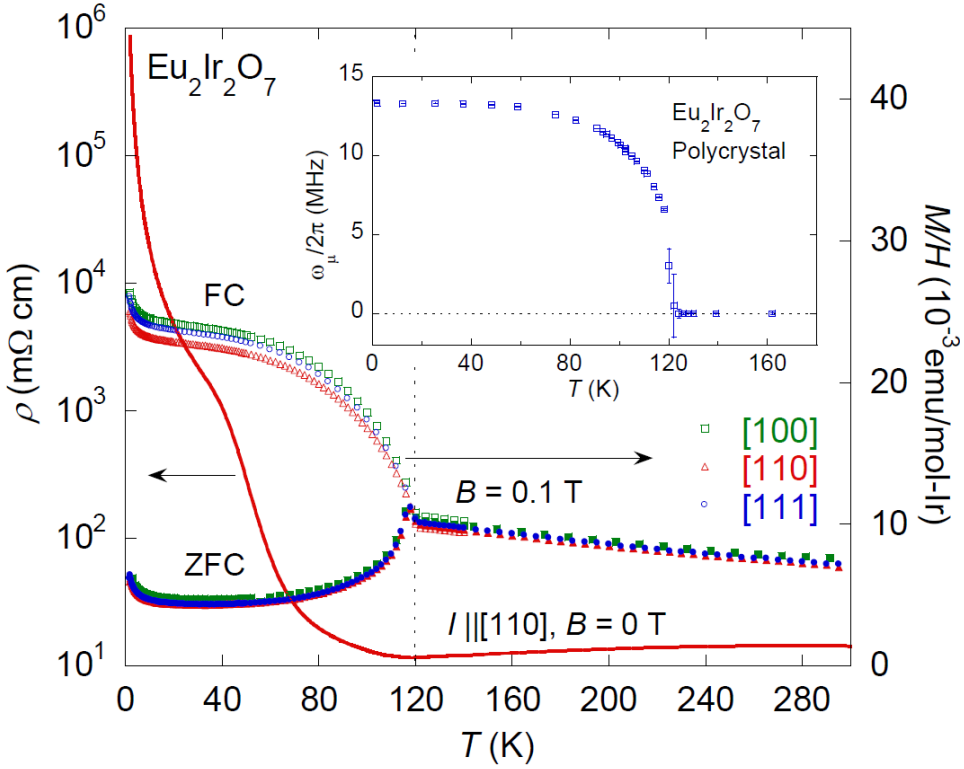


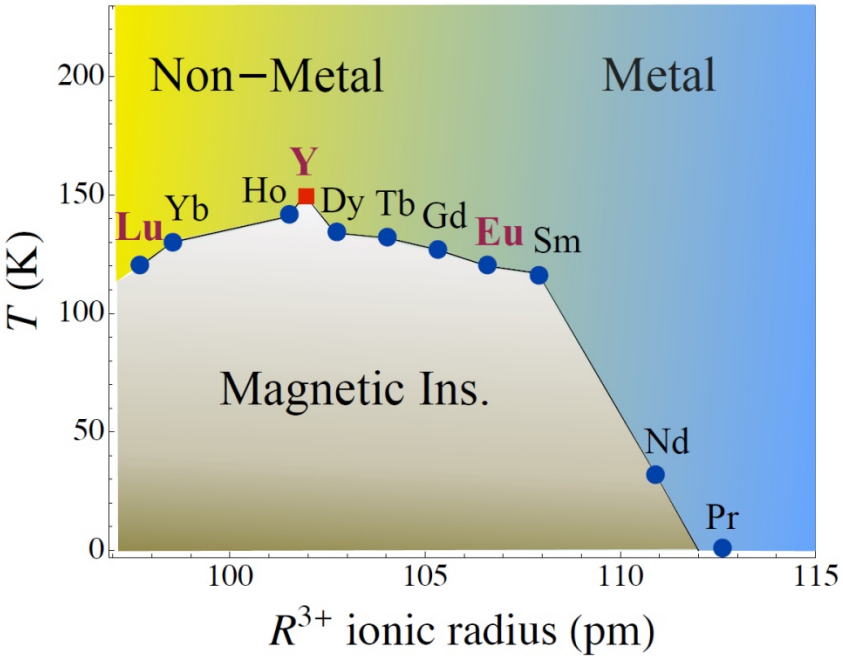
II. Weak to Intermediate Correlations

A. Pyrochlore iridates

1. Experimental resume

- Resistivity goes from being "metallic" ($d\rho/dT > 0$) at $T > T_c$ to "non-metallic" ($d\rho/dT < 0$) at $T < T_c$
- The rare earth ion affects crystal field splitting; T_c is changed
- Larger R^{3+} cation ➔ more metallicity; larger cation ➔ decreased trigonal compression ➔ increased the Ir-O orbital-overlap



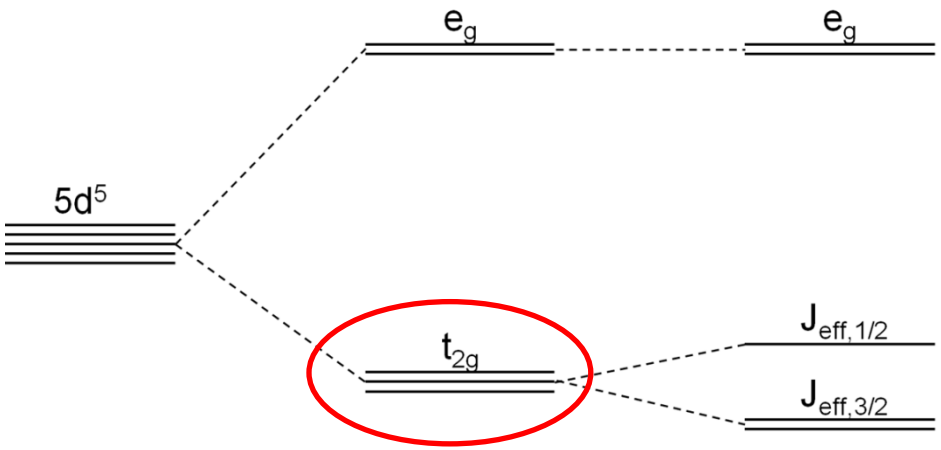


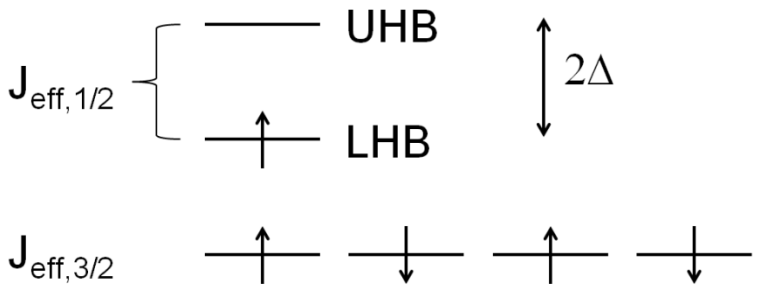
II. Weak to Intermediate Correlations

A. Pyrochlore iridates

2. Electronic structure

- Focus on Ir-electron physics; neglect the rare earth magnetism (relevant at very low temperatures)
- Outer-shell electrons of Ir^{4+} cation are in a $5d^5$ configuration





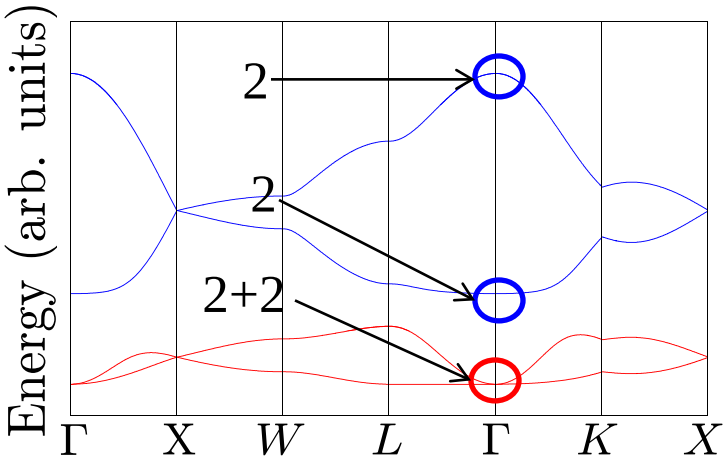
- Full angular momentum operator projected to the t_{2g} manifold: $\mathcal{P}_{t_{2g}}\mathbf{L}\mathcal{P}_{t_{2g}} = -\mathbf{L}^{\text{eff}}_{\ell=1}$
- SOC splits the t_{2g} spinful manifold into a higher energy $J_{\text{eff}} = 1/2$ doublet and a lower $J_{\text{eff}} = 3/2$ quadruplet
- Only (half-filled) $J_{\text{eff}} = 1/2$ doublet near the Fermi energy; 2 bands per Ir atom
- 4 Ir atoms in the tetrahedral unit cell ➔ total 8 Bloch bands near Fermi energy

II. Weak to Intermediate Correlations

A. Pyrochlore iridates

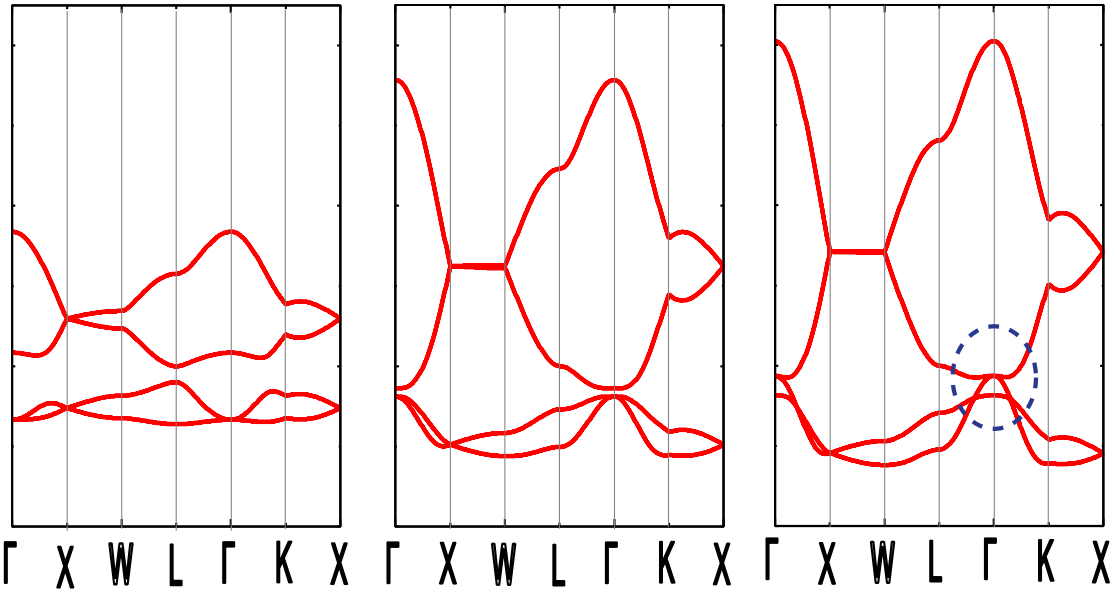
2. Electronic structure

- Consider band structure of the 8 Bloch bands near the Γ point
- Classification of 8 Bloch bands: two 2-D irreps and one 4-D irrep (cubic symmetry)
- Pesin and Balents obtained "4-2-2"
- The "2-2-4" and "4-2-2" can be TIs due to insulating ground state



- Yang *et al*. found "2-4-2" metallic state due to trigonal distortion
- Wan *et al*. also found "2-4-2" metallic state from LDA calculations
- TI state in (metallic) $Y_2Ir_2O_7$ is impossible

Increase distortion ➔



II. Weak to Intermediate Correlations

A. Pyrochlore iridates

2. Electronic structure

- Convenient tight-binding model for both metallic and insulating regimes

$$H_0 = \sum_{\langle ij \rangle} c_i^\dagger \left(t_1 + i t_2 \mathbf{d}_{ij} \cdot \boldsymbol{\sigma} \right) c_j$$

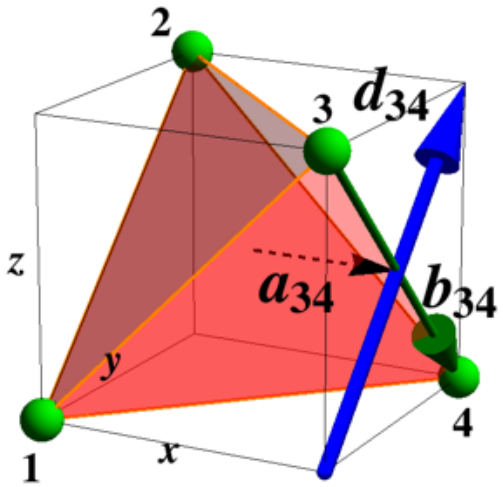
Gives non-trivial Berry phase

$$\mathbf{d}_{ij} = 2\mathbf{a}_{ij} \times \mathbf{b}_{ij}$$

$$\mathbf{a}_{ij} = \frac{1}{2}(\mathbf{b}_i + \mathbf{b}_j) - \mathbf{x}_c$$

$$\mathbf{b}_{ij} = \mathbf{b}_j - \mathbf{b}_i$$

$$\mathbf{b}_1 = (0,0,0),\ \mathbf{b}_2 = (0,1,1),\ \mathbf{b}_3 = (1,0,1),\ \mathbf{b}_4 = (1,1,0)$$



- Diagonalization gives "2-4-2" semi-metallic state for $-2 \leq t_2/t_1 \leq 0$ and a Topological Insulator otherwise
- Semi-metallic state is a zero-gap semiconductor
- This semi-metallic state forms stable *non-Fermi liquid* phase with a quadratic band touching at the Γ point: "Luttinger-Abrikosov-Beneslavskii" (LAB) phase
- About LAB:
  - Electron-hole pair excitations susceptible to "excitonic instability" due to *unscreened* Coulomb interactions
  - Excitonic instability circumvented in the presence of time-reversal and cubic symmetries
  - Enormous zero field anomalous Hall effect

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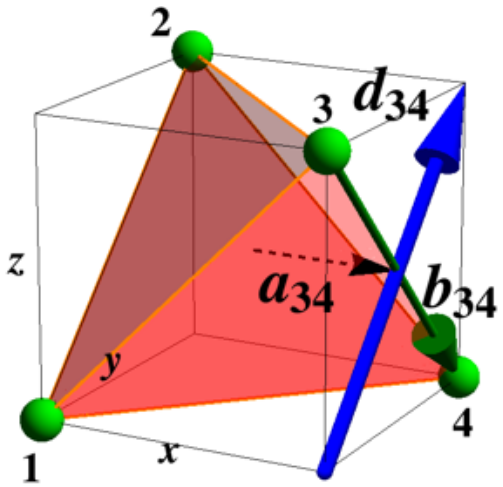
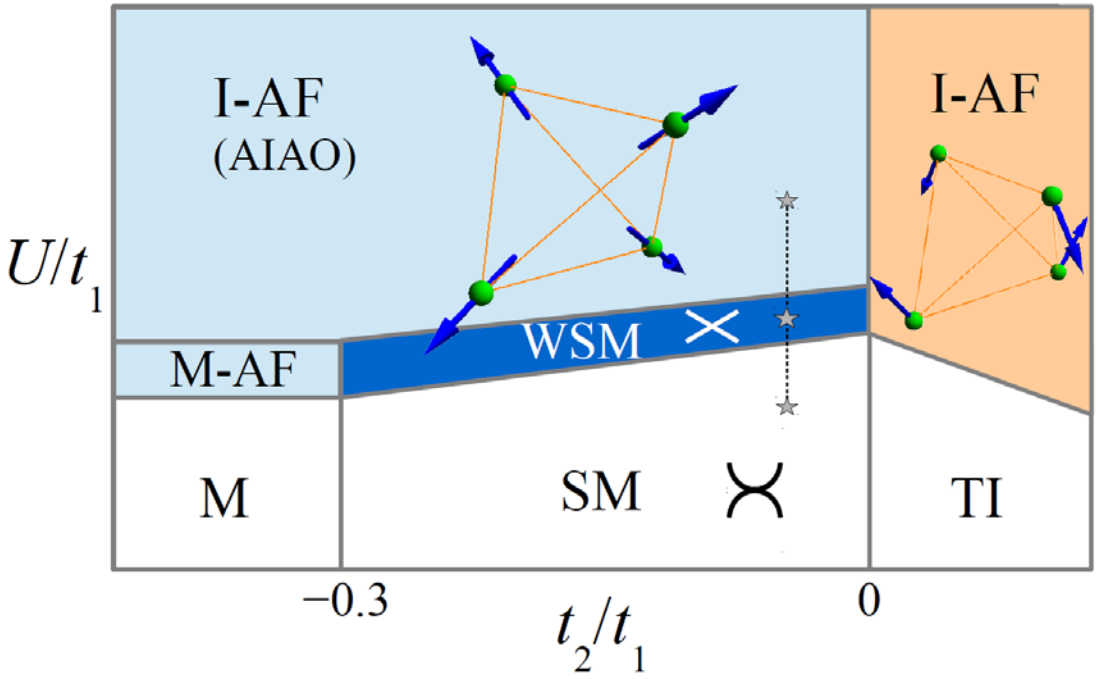
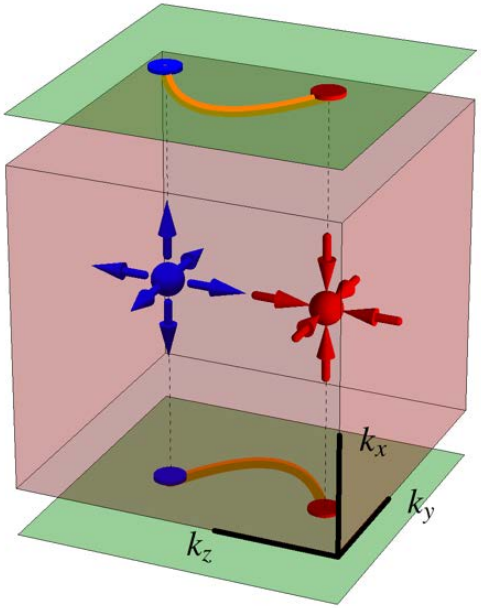
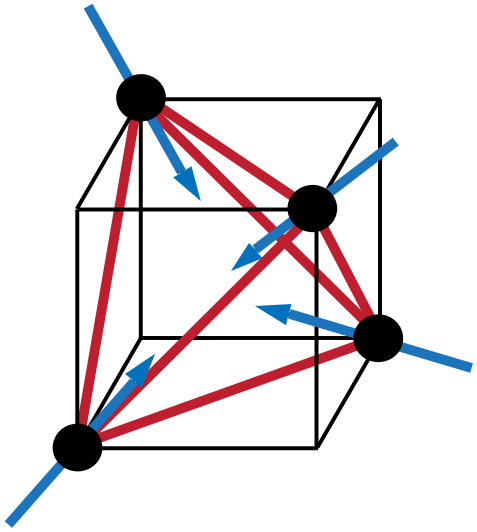






II. Weak to Intermediate Correlations

A. Pyrochlore iridates



3. Magnetism and Weyl Fermions

- Local C_3 axes for four Ir ions constituting a tetrahedron
- Experiments suggest “all-in/all-out” (AIAO) ground state
- Wan *et al*. found Weyl semi-metal with 24 Weyl nodes and *suggested* an axion insulator state

4. The role of many-body effects

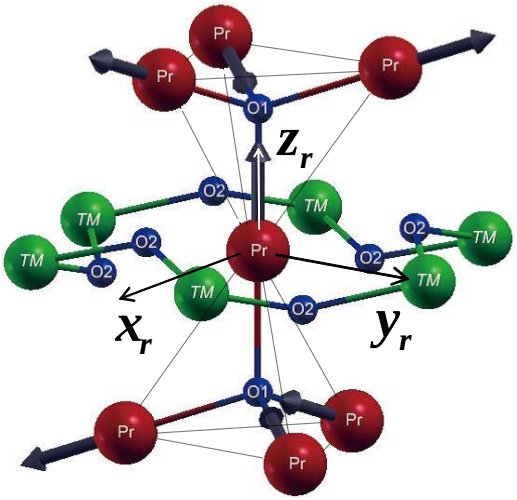
- TI, AIAO, WSM stable to (perturbative) interactions
- Axion insulator state appears in the CDMFT analysis but **not** at the Hartree-Fock level
- Wang *et al*. formulated Z_2 invariant in terms of zero-frequency Green’s function
- Both CDMFT and Hartree-Fock theory cannot capture topological Mott insulator

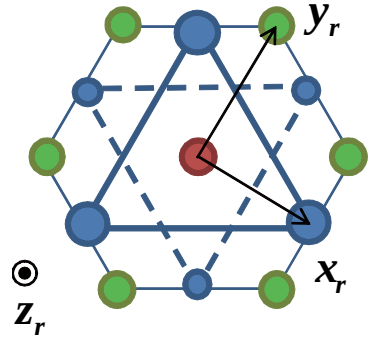
II. Weak to Intermediate Correlations

A. Pyrochlore iridates

5. Interactions with rare earth moments

- What about interactions between *R*-site *f*-electrons and the Ir *d*-electrons?
- Non-Kramers R^{3+} ions (*R* = Pr, Tb, Ho) have an even and Kramers ions (*R* = Nd, Sm, Gd, Dy, Yb) have an odd number of *f*-electrons
- Example: $Yb_2Ir_2O_7$; two ordering temperatures: T_M = 130 K (Ir sublattice) and T^* ≈ 20 K (Yb sublattice)
- Most studied *f*-electron physics in iridates: Pr_2IrO_7 (no MIT)
- Zero field anomalous Hall effect at 0.3 K < *T* < 1.5 K
- Pr moments exhibit spin-ice type physics; "2-in/2-out" configurations on each tetrahedron
- Pr ordering via RKKY interaction
- Chen *et al*. suggest coupling to Ir may help to stabilize the WSM and axion insulator phases



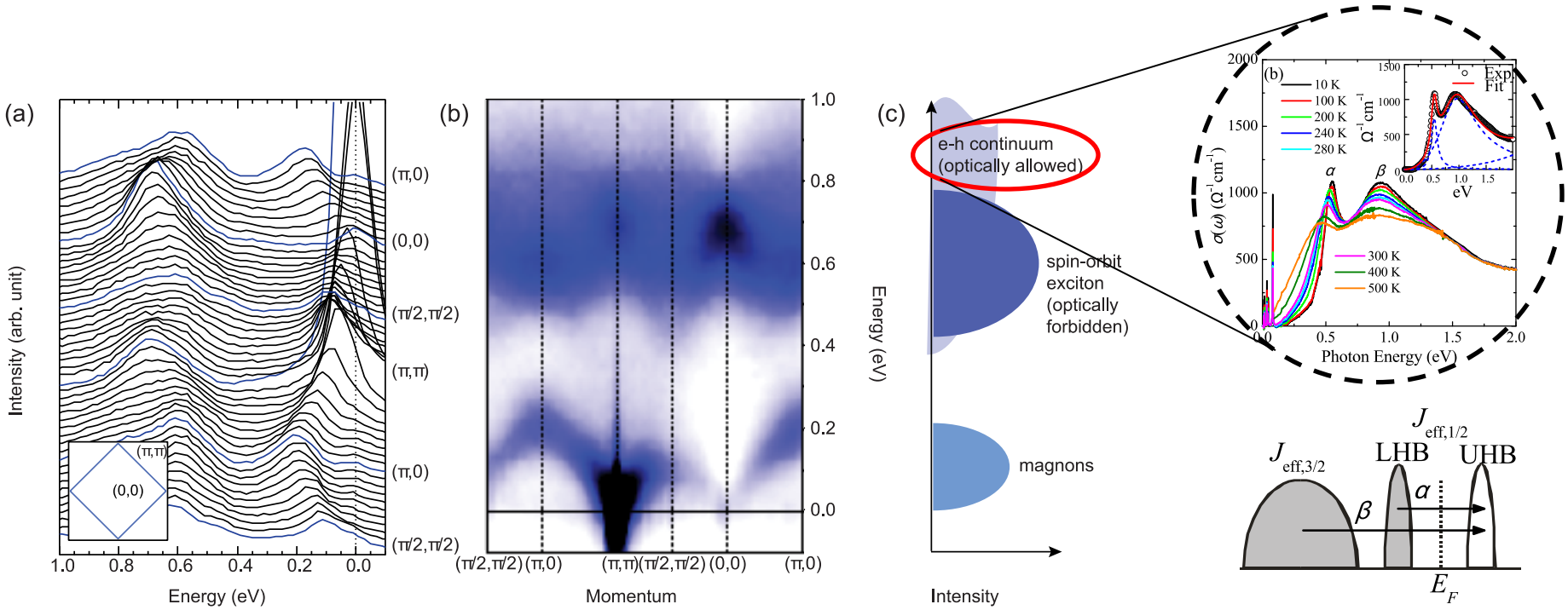


II. Weak to Intermediate Correlations

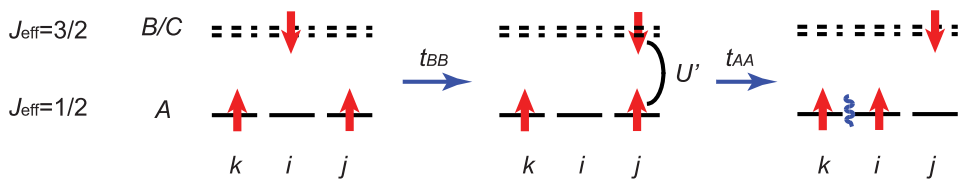
A. Pyrochlore iridates

6. Issues and Outlook

- Pyrochlore iridates undergo MIT with the onset of AIAO magnetic order
- $Nd_2Ir_2O_7$: AIAO at the Nd-sites may imply AIAO at the Ir-sites
- Resonant x-ray diffraction measurements suggest $Eu_2Ir_2O_7$ has AIAO order

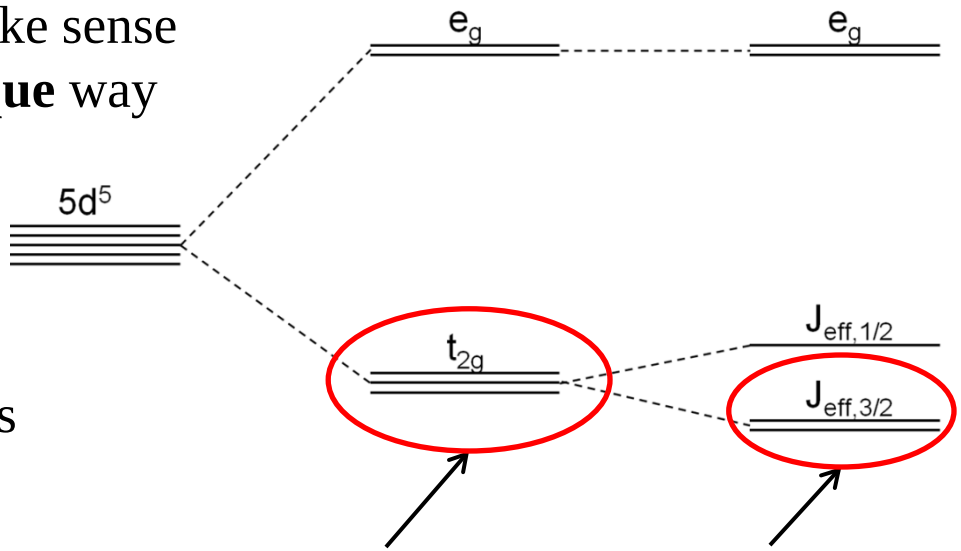


- Generation of the spin-orbit exciton



III. Strong Mott Regime

- Electrons effectively localized to single atoms
- Description in terms of local spin and orbital degrees of freedom (DOF) applies
- Charge gap ≫ energy of spin and orbital excitations
- Notion of *band* topology does **not** make sense
- Orbital degeneracy resolved in a **unique** way
- Orbital DOF behaves as additional "pseudo-spin" **quantum** variable
- Exchange of spin + pseudo-spin ➔ *Kugel-Khomskii* models
- Jahn-Teller effect ➔ lattice distortions split orbital degeneracy
- "Quantumness" washed away by phonon modes
- SOC trades Jahn-Teller effect for entanglement of spin and orbital DOF
- Exchange of spin + pseudo-spin ➔ possibilities of exotic new ground states
- *Quantum spin liquid* and *multipolar ordered* phases possible in honeycomb iridates and the double perovskites

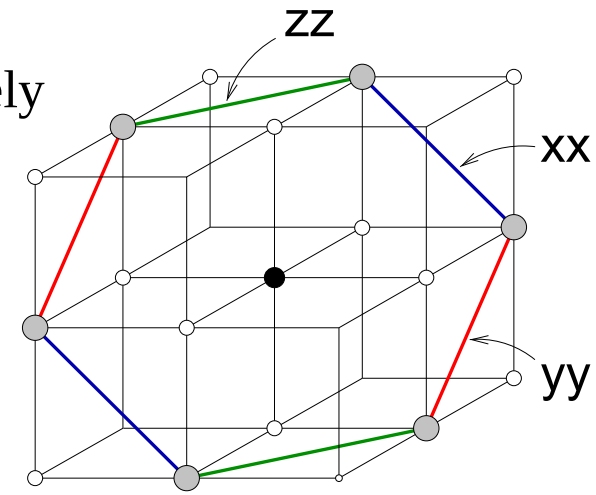


3-fold degenerate for 1, 2, 4, and 5 electrons

2-fold degenerate for 1 and 3 electrons

III. Strong Mott Regime

A. Full degeneracy lifting and honeycomb iridates



- Ir^{4+} with $5d^5$ ➔ orbital degeneracy removed completely
- Na_2IrO_3 and Li_2IrO_3 ➔ Ir^{4+} + strong Mott regime
- Anisotropic exchange model

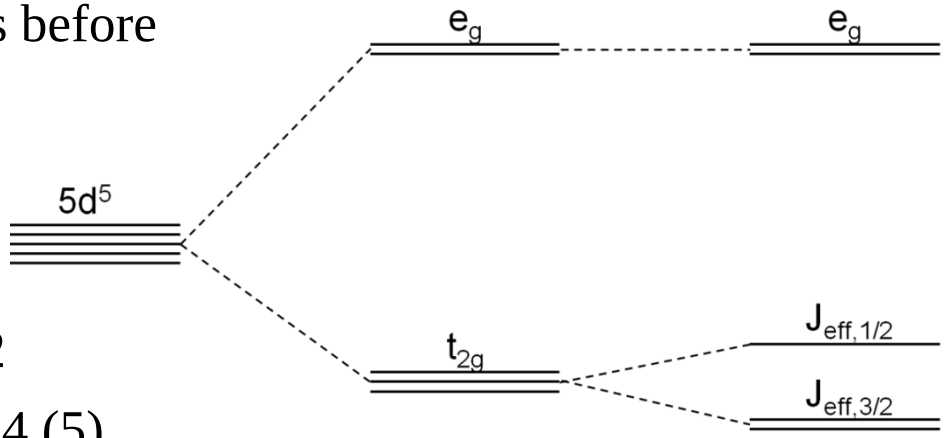
$$H_K = -K \sum_{\alpha=x,y,z} \sum_{\langle ij \rangle \in \alpha} S_i^\alpha S_j^\alpha$$

- The only example of an exactly soluble model for a quantum spin liquid state!
- No magnetic order + charge-neutral “spin”-carrying elementary excitations ➔ Majorana fermions!
- Unfortunately experiments on Na_2IrO_3 have **not** confirmed the Kitaev model yet

III. Strong Mott Regime

B. Partial degeneracy lifting and ordered double perovskites

- Need only 1 or 2 electrons in the 4*d* or 5*d* shells ➔ strongly spin-orbit coupled analogs of Ti^{3+} and V^{3+} or V^{4+}
- V^{3+} or V^{4+} constitute classic families undergoing Mott transitions
- With SOC, degeneracy lifting same as before
- d^1 case ➔ local $J_{eff} = 3/2$ spin
- d^2 case ➔ two parallel (spin-1/2) electrons with aligned spins due to Hund's rule ➔ total spin $S = 1$
- Since t_{2g} has $L_{eff} = 1$, $J_{eff} = L_{eff} + S = 2$
- Overall degeneracy for d^1 (d^2) case is 4 (5)
- *Multipolar* spin exchange common for large J_{eff}
- Multipolar interactions connect directly states with very different S^z quantum numbers ➔ wavefunction delocalization in spin space

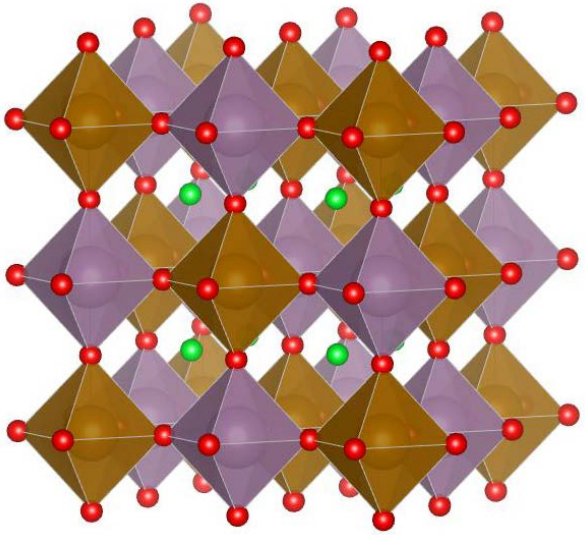


III. Strong Mott Regime

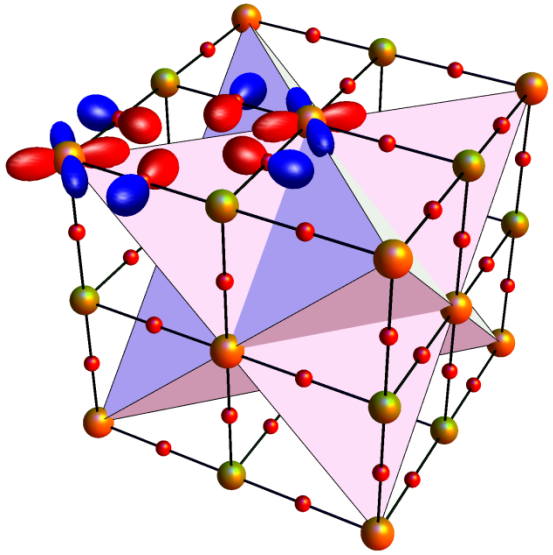
B. Partial degeneracy lifting and ordered double perovskites

1. Double perovskites

- $A_2BB'O_6$ ➔ regular ABO_3 perovskites with alternating B (non-magnetic) and B' (magnetic) atoms
- Consequence of SOC ➔ for $J_{\text{eff}} = 3/2$ the g-factor vanishes
- Magnetic entropy ($R\ln(4)$) estimated from experiments ➔ indication of strong SOC



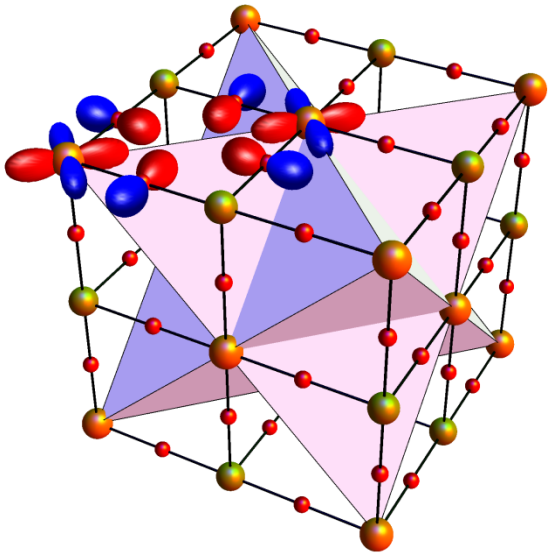
Compound	B'	electron config.	Θ_{CW} (K)	μ_{eff} (μ_B)
Ba_2YMoO_6	Mo^{5+}	$4d^1$	$-91 \sim -219$	$1.34 \sim 1.72$
Sr_2MgReO_6	Re^{6+}	$5d^1$	-426	1.72
Ba_2NaOsO_6	Os^{7+}	$5d^1$	~ -10	~ 0.6
Ba_2CaOsO_6	Os^{6+}	$5d^2$	-157	1.61
La_2LiReO_6	Re^{5+}	$5d^2$	-204	1.97



III. Strong Mott Regime

B. Partial degeneracy lifting and ordered double perovskites

2. Multipolar exchange



- Consider Kugel-Khomskii type exchange with all orbitals are included ➔ then project to the effective spins in the strong SOC limit
- For d^1 case consider exchange: $\mathcal{H}_{\rm ex} = \mathcal{H}^{xy}_{\rm ex} + \mathcal{H}^{yz}_{\rm ex} + \mathcal{H}^{xz}_{\rm ex}$

$$\mathcal{H}^{xy}_{\rm ex} = J \sum_{\langle ij\rangle \in xy \text{ plane}} \left(\mathbf{S}_{i,xy} \cdot \mathbf{S}_{j,xy} - \frac{1}{4} n_{i,xy} n_{j,xy} \right)$$

- Consider Kugel-Khomskii type exchange with all orbitals are included ➔ then project to the effective spins in the strong SOC limit
- In strong for t_{2g} we have $\mathbf{S}_{i,xy} = S_i \left[1 - (L^z_i)^2\right]$ $\qquad n_{i,xy} = 1 - (L^z_i)^2$

$$S_i = \mathbf{S}_{i,xy} + \mathbf{S}_{i,xz} + \mathbf{S}_{i,yz}$$

- Performing the projections we get

$$\tilde{S}^{\alpha}_{i,xy} = \frac{1 + 2\delta_{\alpha,z}}{4} S^{\alpha}_i - \frac{1}{3} S^z_i S^{\alpha}_i S^z_i \quad (\alpha = x, y, z), \qquad \tilde{n}_{i,xy} = \frac{3}{4} - \frac{1}{3}(S^z_i)^2$$

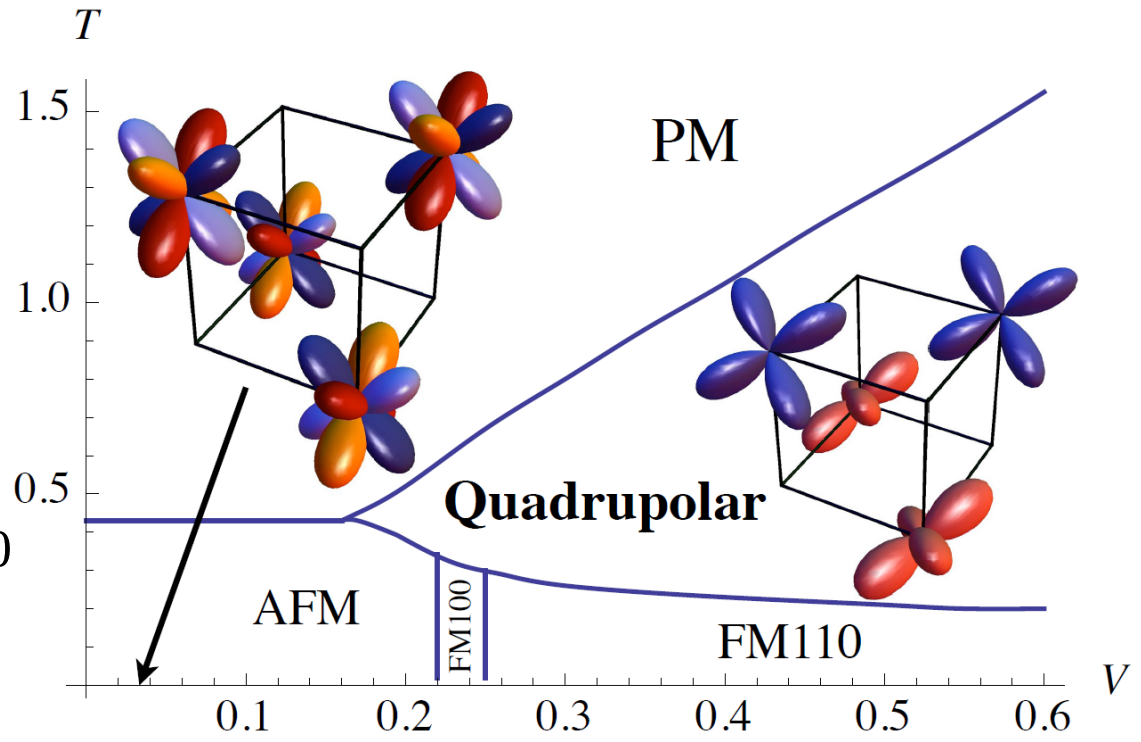
- For d^1 we have two exchange channels: ferromagnetic exchange between orthogonal orbitals (J') and electrostatic quadrupole interaction (V)

III. Strong Mott Regime

B. Partial degeneracy lifting and ordered double perovskites

3. Mean field theory

- Exotic phases even in mean field
- Anisotropic contributions come from quadrupolar and octupolar interactions
- Antiferromagnetic phase for small J'/J and V/J
- Ferromagnetic phases (FM110 and FM100) for large J'/J and V/J
- Quadrupolar states classified by eigenstates of $Q_i^{\mu\nu} = \langle S_i^\mu S_i^\nu \rangle - \frac{1}{3} S(S+1)\delta^{\mu\nu}$



- Only 1 independent eigenvalue (q, q, -2q) ➔ Uniaxial nematic phase
- 2 independent eigenvalues (q_1, q_2, $-q_1$, $-q_2$) ➔ Biaxial nematic phase
- Quadrupolar phase appears in d^2 perovskite even for $T = 0$; d1 must always break time reversal symmetry at $T = 0$ to avoid ground state degeneracy.

III. Strong Mott Regime

B. Partial degeneracy lifting and ordered double perovskites

4. Beyond mean-field theory

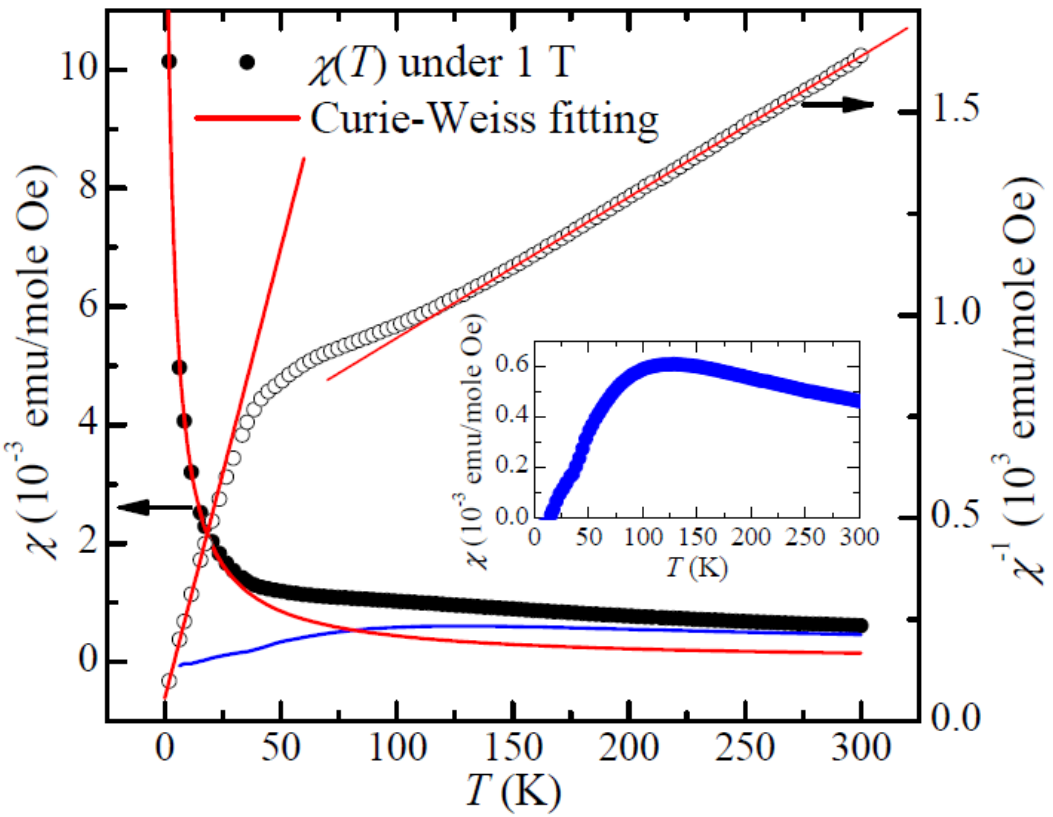
- Multipolar interactions destabilize conventional, magnetically ordered semiclassical ground states
- More “spin flip” terms analogous to the $S_i^+ S_j^-$ couplings
- Quantum disordered ground states can be established rigorously for AKLT models
- Multipolar Hamiltonians are intermediate between conventional spin models and these special cases
- Check for disordered states ➔ gauge the magnitude of quantum fluctuations within a spin-wave expansion
- Valence bond solids and quantum spin liquid states predicted in various parameter regimes
- Non-cubic crystal fields give highly frustrated systems ➔ quantum fluctuations support a spin liquid phase

III. Strong Mott Regime

B. Partial degeneracy lifting and ordered double perovskites

5. Connections to experiments

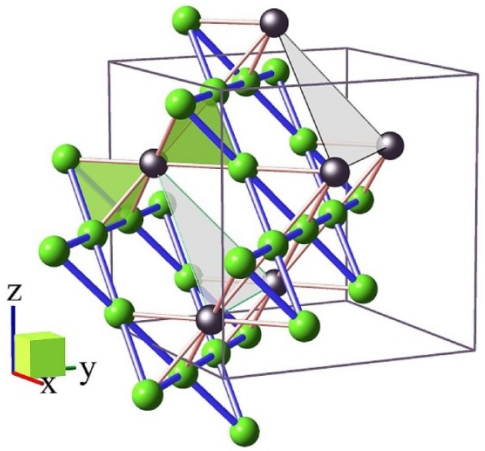
- Ba_2YMoO_6 cubic to low temperatures
- Like many double perovskites has a two Curie regime
- Phonon mode above 130 K; consistent with local structural change

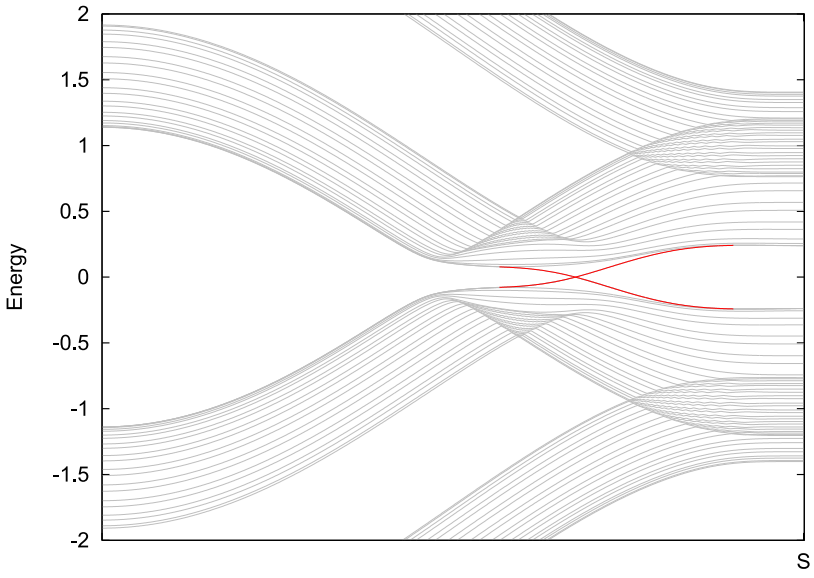


- Ba_2NaOsO_6 has a ferromagnetic ground state below 6.8 K with [110] easy axis
- Landau theory predicts [100] or [111] as the easy axis
- Quadrupolar ordering mechanism can account for it; associates with a structural change; not observed so far

IV. Concluding Remarks and Outlook

- Not discussed ➔ Ruddlesdon-Popper series of perovskite iridates ➔ formula for a *n*-layer quasi-2D system ➔ $Sr_{n+1}Ir_nO_{3n+1}$ for $n = 1, 2, \infty$
- The $n = 1$ case (Sr_2IrO_4) expected to be a high-Tc superconductor, upon doping, owing to its similarity cuprate parent compound to La_2CuO_4
- This review mainly discusses bandwidth controlled MITs; filling (or doping) controlled MITs might reveal interesting physics
- Exotic fractionalized phases possible: fractional Chern insulators from heterostructures of $SrIrO_3$-$SrTiO_3$
- Controversies ➔ Mott vs. Slater insulator in Sr_2IrO_4? ➔ contradictory results from different calculations ➔ experimental evidence needed
- Heterostructures of $SrIrO_3$ and $R_2Ir_2O_7$ along the [111] direction can give topological insulators and IQHE





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- William Witczak-Krempa, Gang Chen, Yong Baek Kim, and Leon Balents. “Correlated quantum phenomena in the strong spin-orbit regime.” arXiv preprint arXiv:1305.2193 (2013)