Title: Spin-orbit physics in the Mott regime

Date: Apr 22, 2010 10:00 AM

URL: http://pirsa.org/10040079

Abstract: Recent theory and experiment have revealed that strong spin-orbit coupling (SOC) can have dramatic qualitative effects on weakly interacting electrons. For instance, it leads to a distinct phase of matter, the topological band insulator. I will discuss the combined effects of SOC and strong electron correlation. For a "strong" Mott insulator, in which the electrons are well localized, SOC can compete with exchange interactions, leading to quenching of orbital degeneracy and even an instance of quantum criticality. For intermediate correlations, SOC has both quantitative and qualitative effects upon the Mott transition. An illustrative example of Ir-based pyrochlores will be presented, suggesting a rich interplay of correlations and SOC, and the possibility of distinct new electronic phases such a "topological Mott insulator".

Spin-orbit physics in the Mott regime

Leon Balents, KITP

4 corners conference, April 2010

The David and Lucile Packard Foundation

Collaborators

- $FeSc_2S_4$:

Gang Chen

Andreas Schnyder
KITP -> Stuttgart

- Mott transition (pyrochlore iridates)

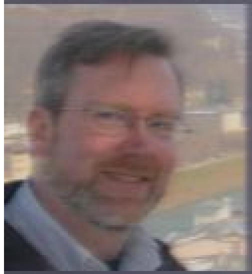
Dymtro Pesin
UT Austin

Spin-orbit physics

- Ashcroft+Mermin: an afterthought
- Recently, brought to the forefront:
    - Quantum spin Hall effect in HgTe quantum wells
    - Topological band insulators: $Bi_{1-x}Sb_x$, Bi_2Se_3
- This is an extremely hot topic, and deservedly so

Outline

1. Brief introduction to recent discoveries in systems with strong SOIs
2. SOIs deep in the Mott regime – understanding an experimental "spin liquid"
3. SOIs near the Mott transition, and a model for Ir pyrochlores



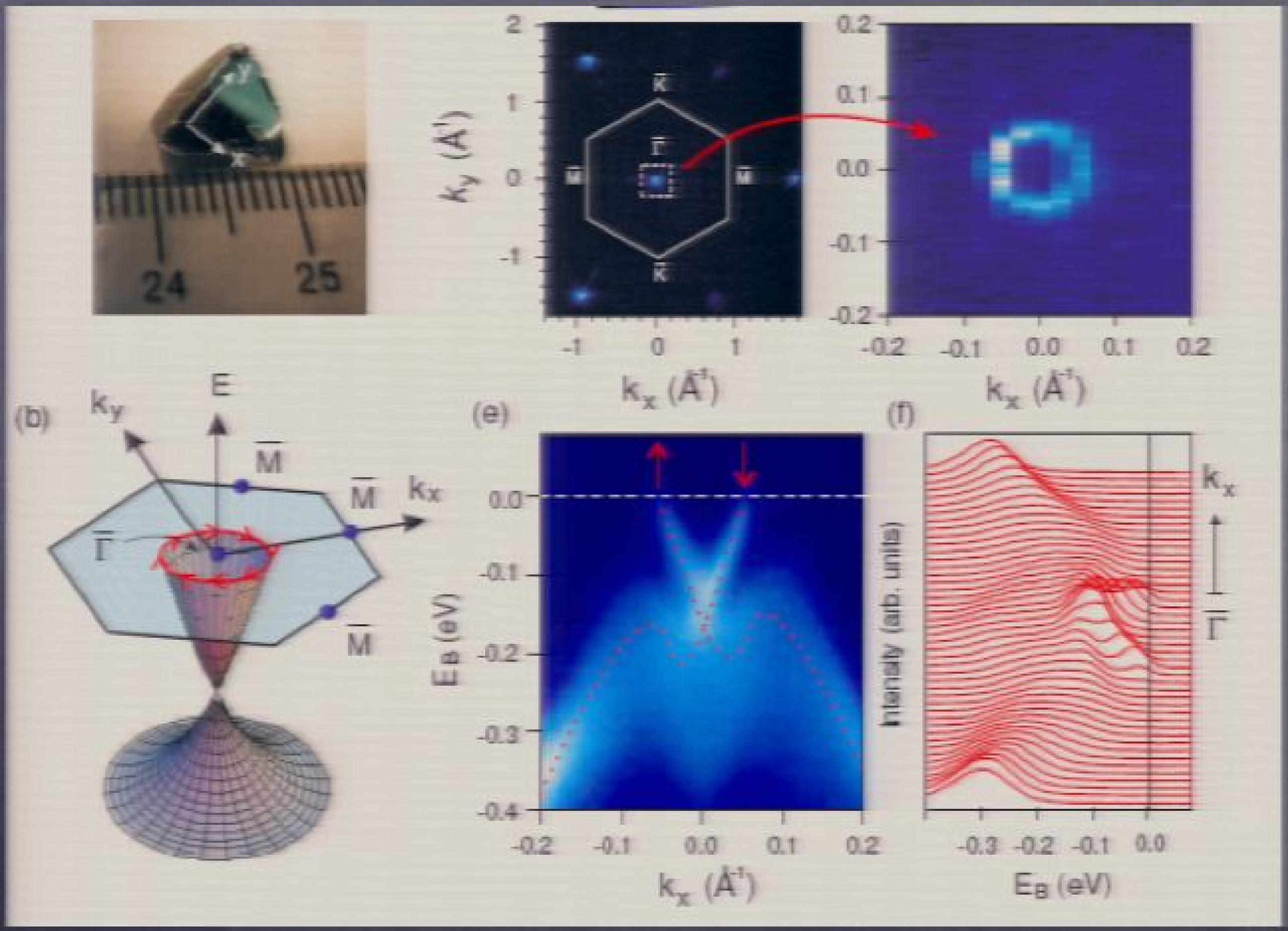
Topological Insulators

2d: Kane, Mele (2005); Bernevig, Hughes, Zhang (2006)
3d: L. Fu, C. Kane, E. Mele (2007); J. Moore, LB (2007)

- 3d band insulators w/ significant SOI can have hidden topological structure, somewhat similar to the IQHE
  - Exhibit "helical" surface states - 2d chiral Dirac fermions (evades Fermion doubling problem!)
    - Cannot be localized by disorder
  - Surface Hall effect <-> magnetoelectric response
- Sevral experimental examples
  - $Bi_{1-x}Sb_x$, Bi_2Se_3, Bi_2Te_3

Example: Bi_2Te_3

- M.Z. Hasan group – ARPES studies



Recent developments

- Experiments:
  - Superconducting and ferromagnetic versions of the materials have been made
  - STM measurements have confirmed suppressed backscattering
  - Transport measurements show surface conduction
- Theory
  - Novel magnetoelectric effects predicted
  - Superconducting-TI structures and materials predicted to host Majorana fermions

What about interactions?

Recent developments

- Experiments:
  - Superconducting and ferromagnetic versions of the materials have been made
  - STM measurements have confirmed suppressed backscattering
  - Transport measurements show surface conduction
- Theory
  - Novel magnetoelectric effects predicted
  - Superconducting-TI structures and materials predicted to host Majorana fermions

What about interactions?

Some theoretical suggestions

- Spontaneous TIs in models with microscopic SU(2) symmetry

S. Raghu et al, 2008
T. Grover + Senthil, 2008
Y. Zhang et al, 2009

- Antiferromagnetism from a TI – Na_2IrO_3

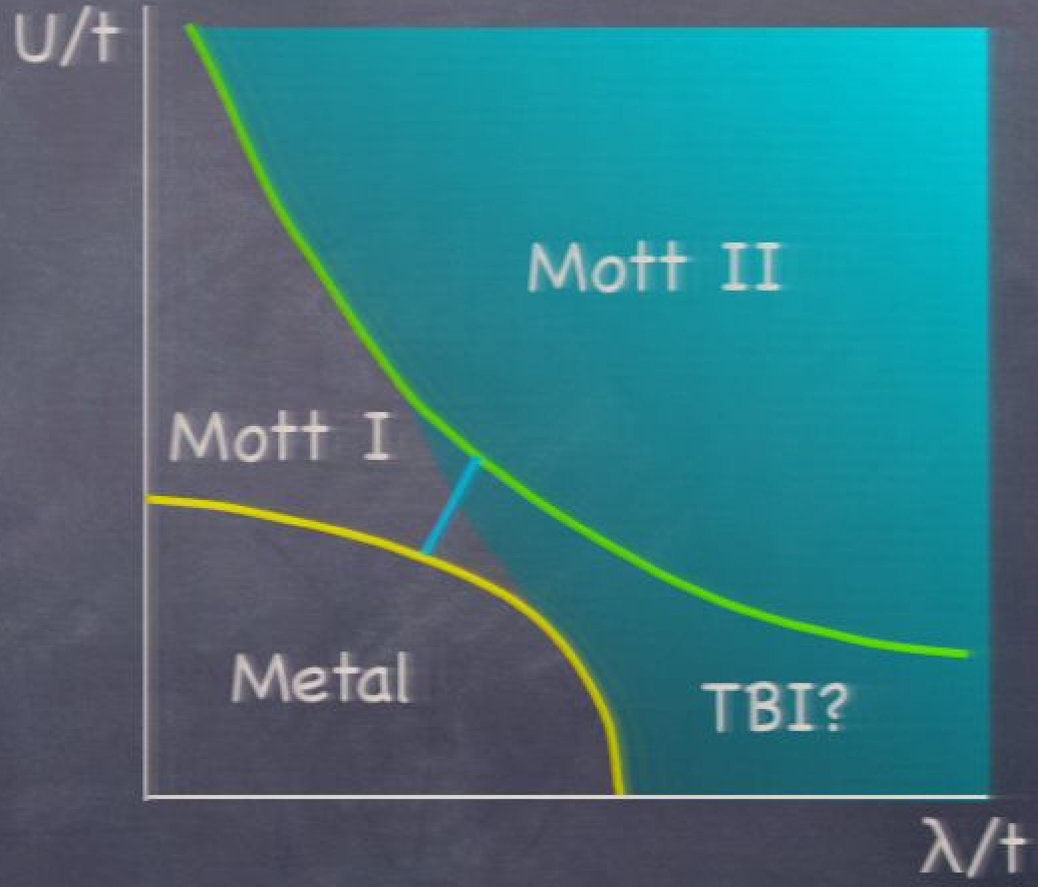
A. Shitade et al, 2009
H. Jin et al, arXiv:0907.0743

- 2d Fractionalized QSHE – spin-charge separated TI

M. W. Young et al, 2008

Materials perspective

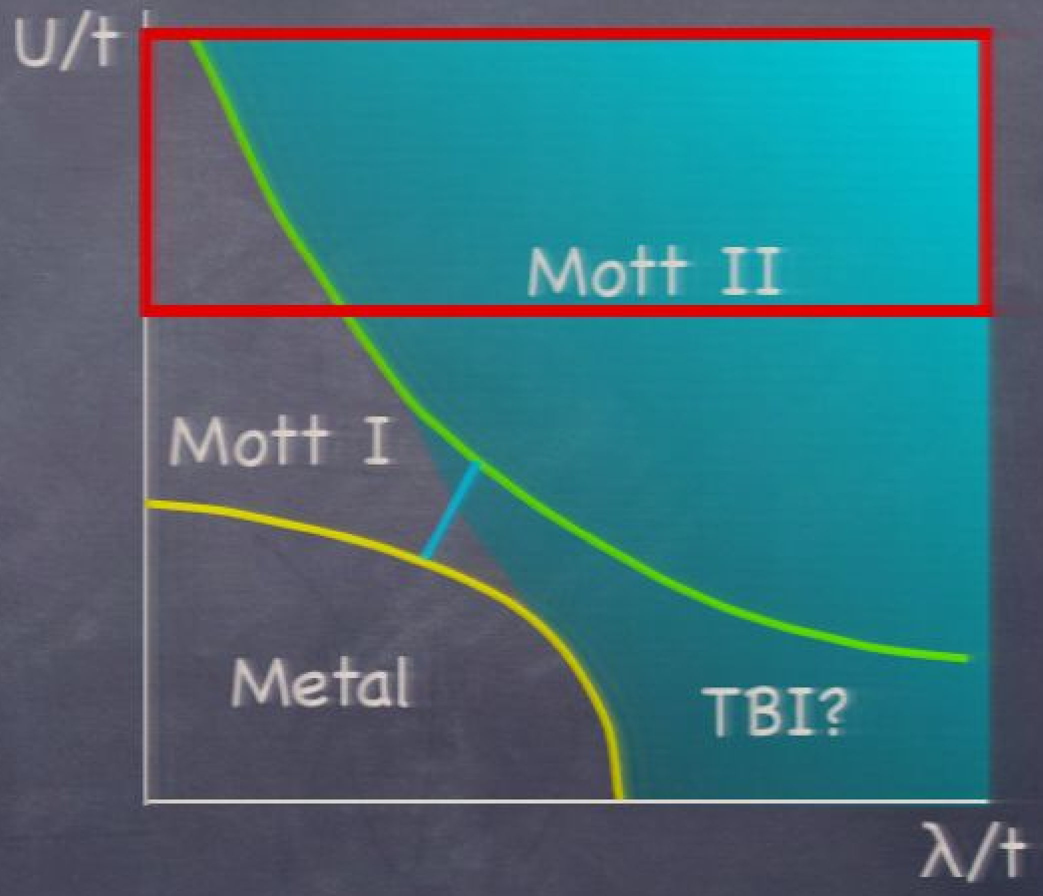
- Coulomb correlations reduce bandwidth
  - Spin-orbit enhanced relative to bandwidth
- In Mott insulator, compare SO to J not t.



schematic phase diagram

Materials perspective

- Coulomb correlations reduce bandwidth
  - Spin-orbit enhanced relative to bandwidth
- In Mott insulator, compare SO to J not t.



schematic phase diagram

Strong Mott Insulators with strong SOIs

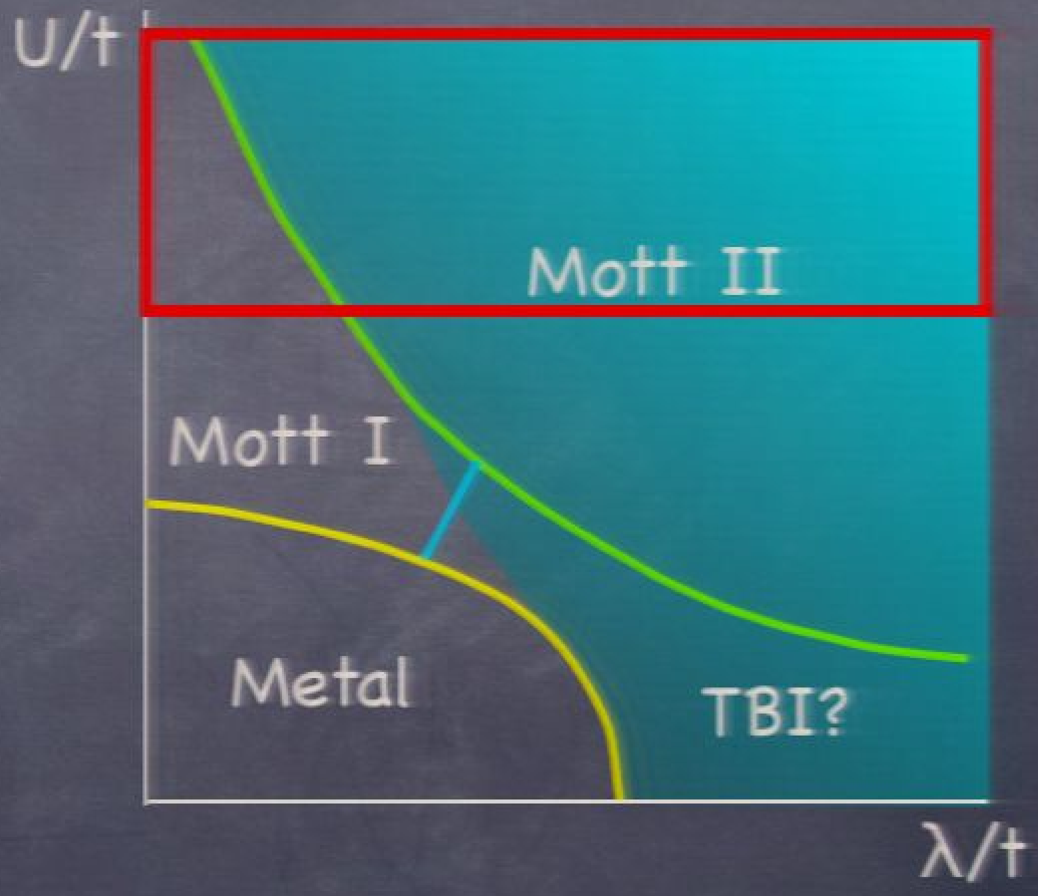
- some Fe and Co compounds, e.g. $FeSc_2S_4$ - orbitally degenerate spinel
- 4d and 5d double perovskites – Ba_2NaOsO_6, Ba_2LiOsO_6 etc.
- However, even in strong MIs with "weak" SOIs (e.g. Dzyaloshinskii–Moriya coupling at few % level), the SOIs can control the ground state when the exchange interactions are frustrated
  - e.g. triangular Cs_2CuCl_4, and probably most kagome materials

Strong Mott Insulators with strong SOIs

- some Fe and Co compounds, e.g. $FeSc_2S_4$ - orbitally degenerate spinel
- 4d and 5d double perovskites - Ba_2NaOsO_6, Ba_2LiOsO_6 etc.
- However, even in strong MIs with "weak" SOIs (e.g. Dzyaloshinskii-Moriya coupling at few % level), the SOIs can control the ground state when the exchange interactions are frustrated
  - e.g. triangular Cs_2CuCl_4, and probably most kagome materials

Materials perspective

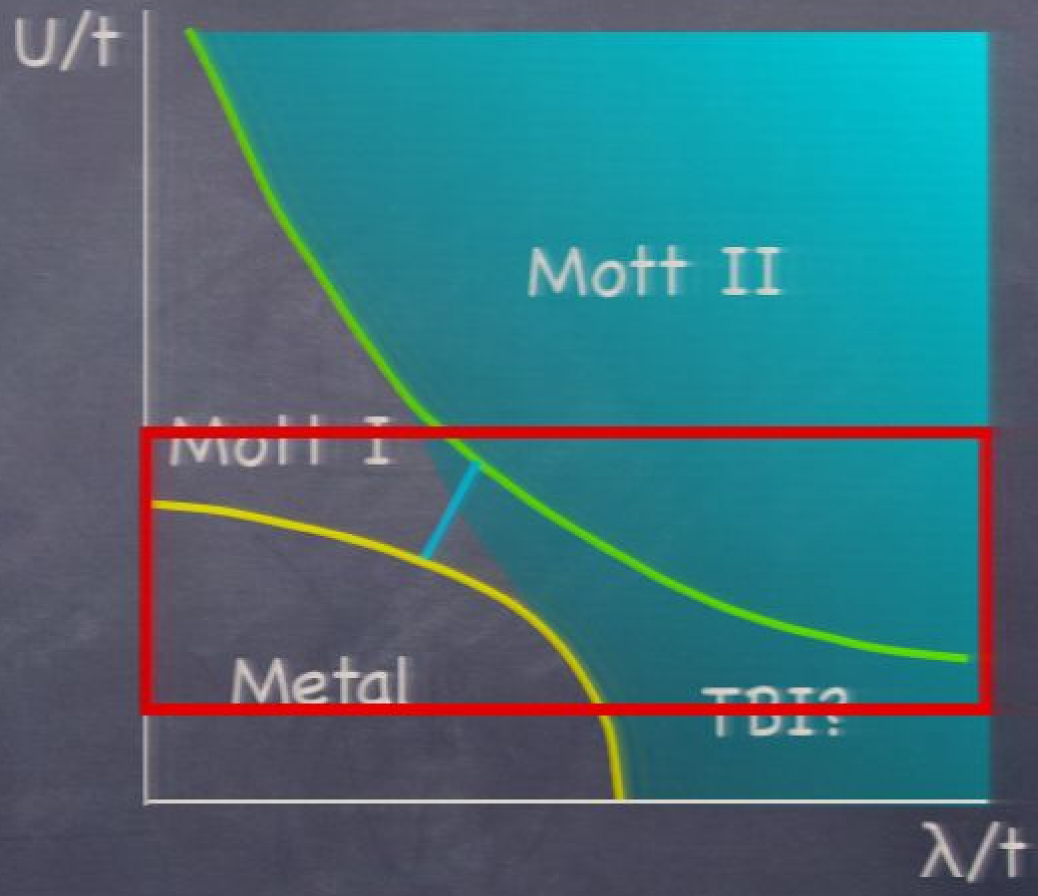
- intermediate regime



schematic phase diagram

Materials perspective

- intermediate regime



schematic phase diagram

Weak Mott Insulators with strong SOIs

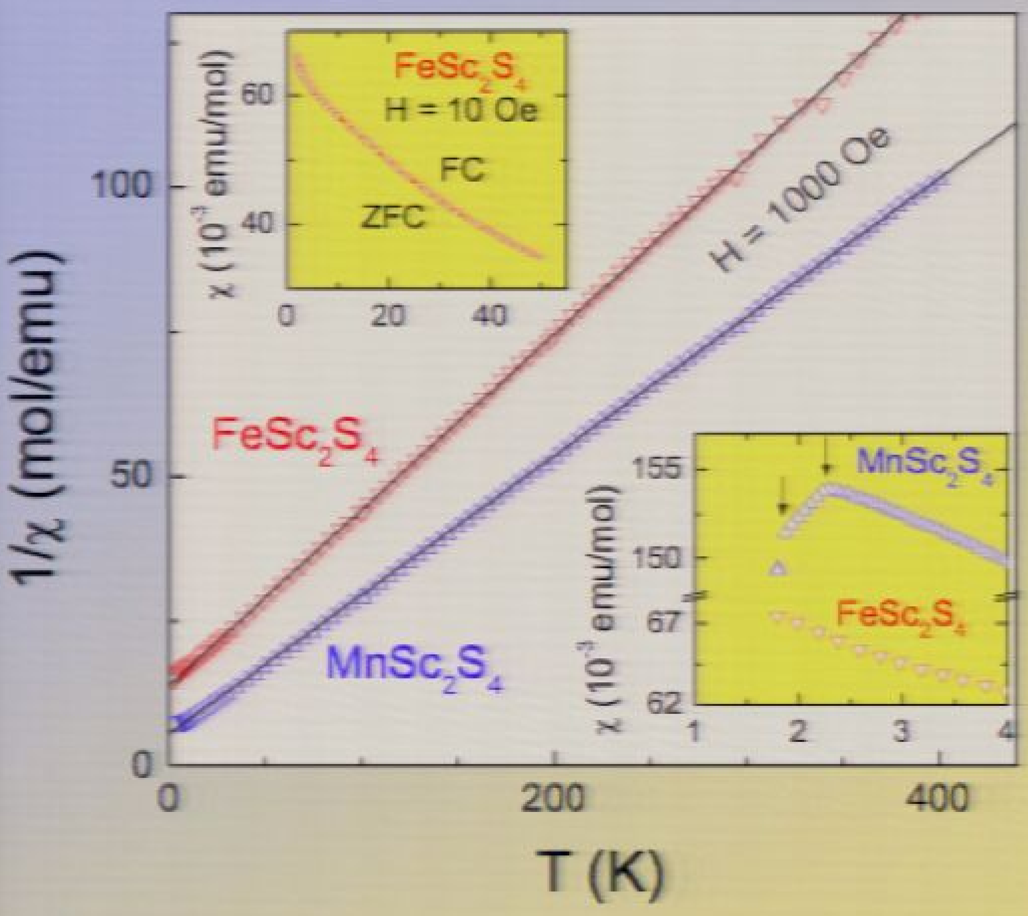
- Most 5d TM ions have smallish U≈1eV, and hence tend to be either metallic or weak Mott insulators
    - together, SOI and U can conspire to produce an insulating state
- e.g. 5d iridates - Sr_2IrO_4, Na_2IrO_3, $Na_4Ir_3O_8$ (hyperkagome), $Ln_2Ir_2O_7$ (pyrochlores)

$FeSc_2S_4$: spin-orbital quantum criticality

QSL candidates

- $CsCu_2Cl_4$ – spin-1/2 anisotropic triangular lattice
- $NiGa_2S_4$ – spin-1 triangular lattice
- κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$, $EtMe_3Sb[Pd(dmit)_2]_2$ – triangular lattice organics
- $FeSc_2S_4$ – orbitally degenerate spinel
- $Na_4Ir_3O_8$ – hyperkagome
- $ZnCu_3(OH)_6Cl_2$, $Cu_3V_2O_7(OH)_2 \cdot 2H_2O$, $BaCu_3V_2O_8(OH)_2$– kagome

Frustration Signature



$FeSc_2S_4$: $\theta_{CW} = 50$ K

T > 30 mK:

no long-range magnetic order

no spin-glass

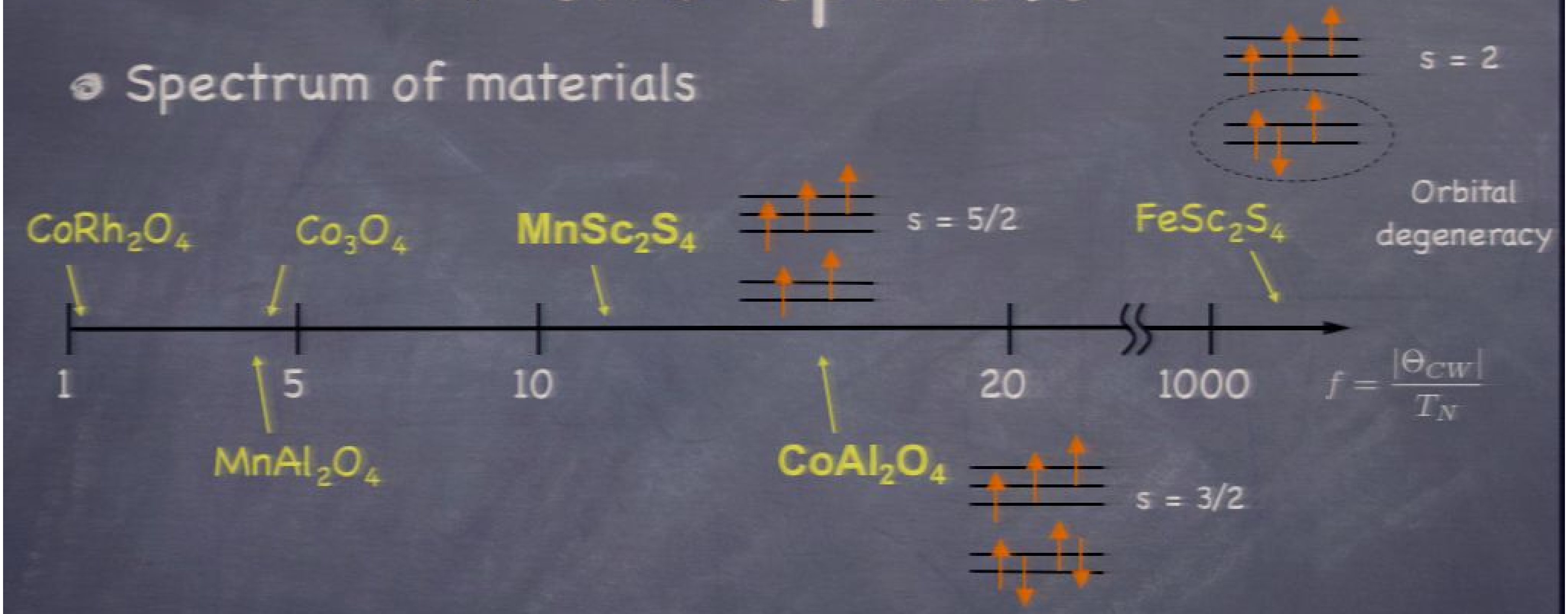
$MnSc_2S_4$: $\theta_{CW} = 25$ K

AFM transition @ 2 K

Fritsch *et al.*, PRL **92**, 116401, 2004

A-site spinels

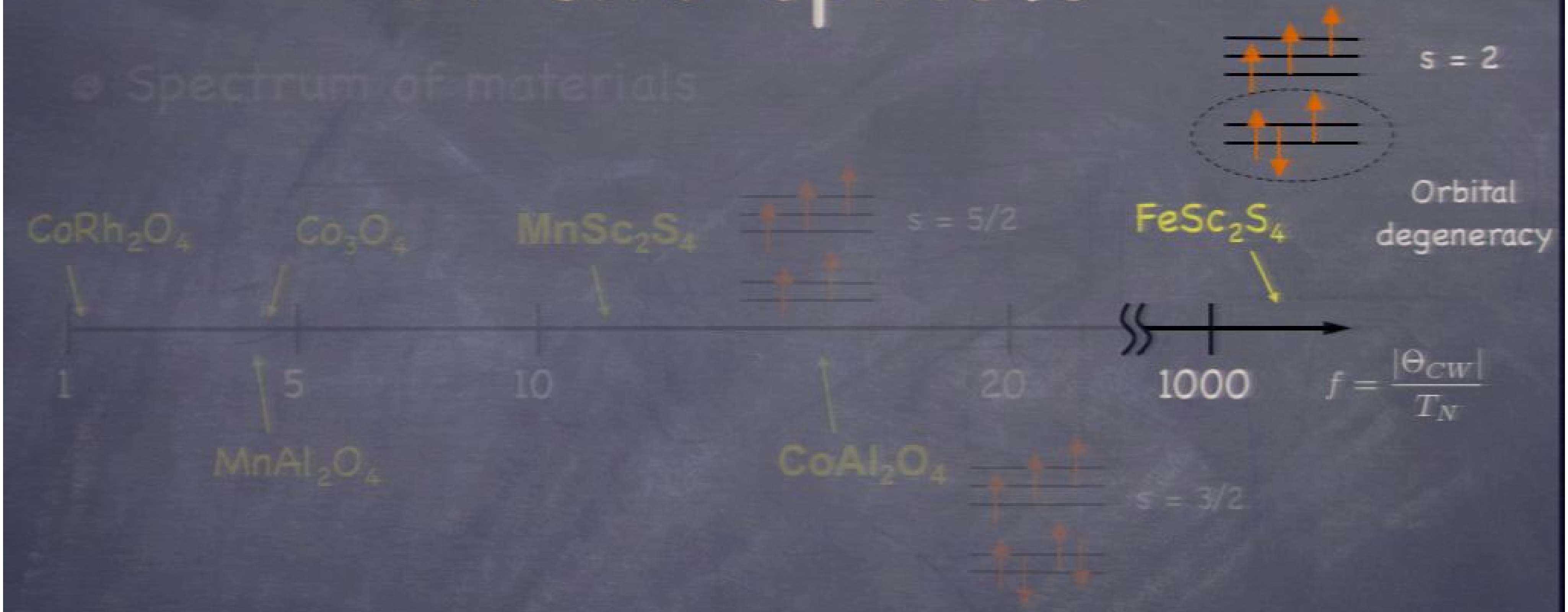
- Spectrum of materials



Fritsch et al. PRL **92**, 116401 (2004); N. Tristan et al. PRB **72**, 174404 (2005); T. Suzuki *et al*. (2006)

A-site spinels

- Spectrum of materials



s = 2

Orbital degeneracy

$CoRh_2O_4$

Co_3O_4

$MnSc_2S_4$

s = 5/2

$FeSc_2S_4$

1

5

10

20

1000

$f = \frac{|\Theta_{CW}|}{T_N}$

$MnAl_2O_4$

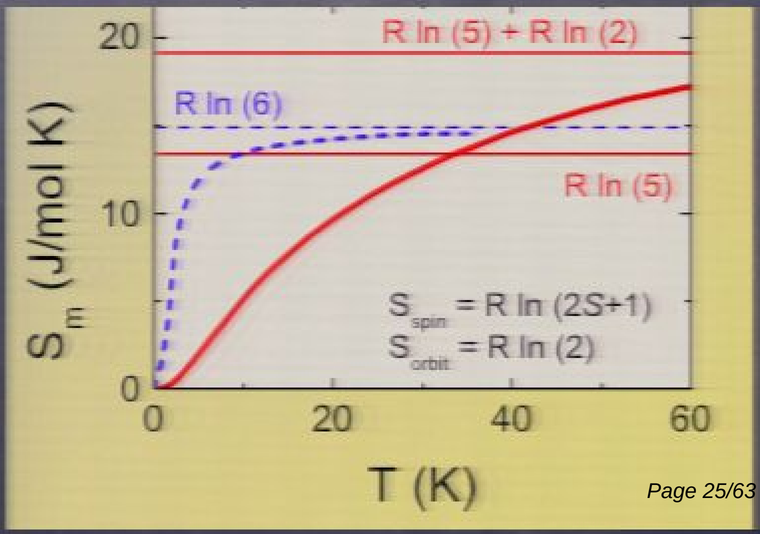
$CoAl_2O_4$

s = 3/2

Fritsch et al. PRL **92**, 116401 (2004); N. Tristan et al. PRB **72**, 174404 (2005); T. Suzuki *et al*. (2006)

Orbital degeneracy in $FeSc_2S_4$

- Chemistry:
  - Fe^{2+}: $3d^6$
  - 1 hole in e_g level
- Spin S=2
- Orbital pseudospin 1/2
- Static Jahn-Teller does not appear



Atomic Spin Orbit

- Separate orbital and spin degeneracy can be split!

$$H_{SO} = -\lambda\left(\frac{1}{\sqrt{3}}\tau^x\left[(S^x)^2-(S^y)^2\right]+\tau^z\left[(S^z)^2-\frac{S(S+1)}{3}\right]\right)$$

- Energy spectrum: singlet GS with gap = λ

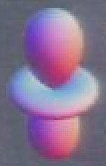
- Microscopically,

$$\lambda = \frac{6\lambda_0^2}{\Delta}$$

- Naive estimate λ ≈ 25K

Spin orbital singlet

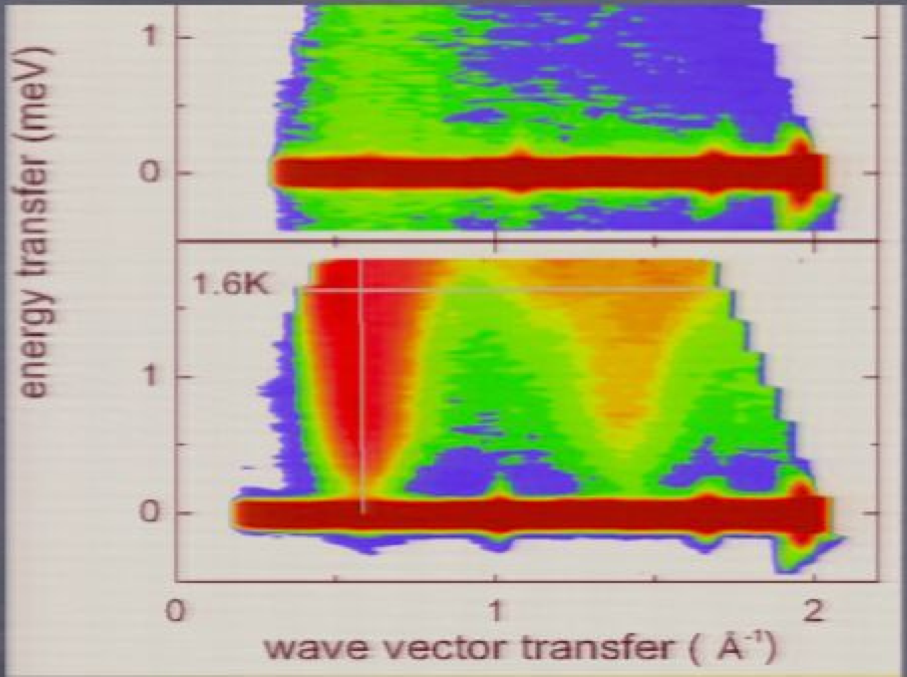
- Ground state of λ>0 term:

$|$  $\rangle |S^z\!=\!0\rangle - \frac{1}{\sqrt{2}}\,|$  $\rangle \big(|S^z\!=\!2\rangle + |S^z\!=\!-2\rangle \big)$

- Due to gap, there is a stable SOS phase for λ >> J.

Exchange

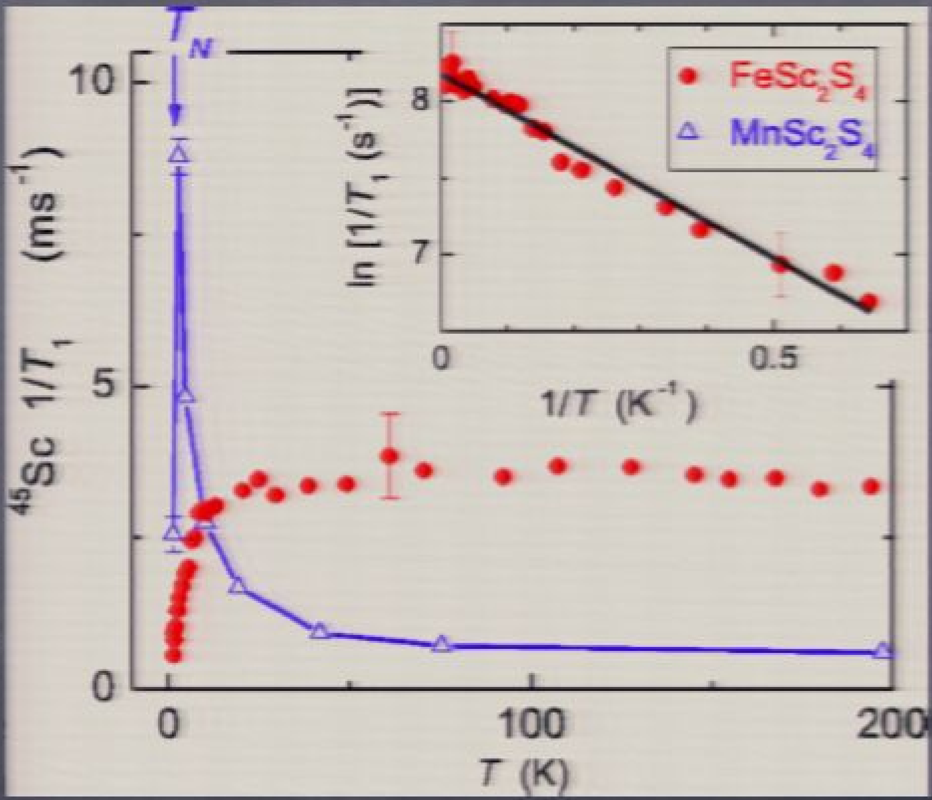
- Inelastic neutrons show significant dispersion indicating exchange
- Bandwidth ≈ 20K similar order as Θ_{CW} and estimated λ
- Gap (?) 1–2K
  - Small gap is classic indicator of incipient order



A. Krimmel *et al.*, Phys. Rev. Lett. **94**, 237402, 2005

Exchange

- Inelastic neutrons show significant dispersion indicating exchange
- Bandwidth ≈ 20K similar order as Θ_{CW} and estimated λ
- Gap (?) 1–2K
    - Small gap is classic indicator of incipient order



N. Büttgen et al, PRB **73**, 132409 (2006)

Exchange

- Most general symmetry-allowed form of exchange coupling (neglecting SOI)

$$H_{ex} = \frac{1}{2}\sum_{ij}\Big\{J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j + K_{ij}\boldsymbol{\tau}_i\cdot\boldsymbol{\tau}_j + \tilde{K}_{ij}\tau_i^y\tau_j^y \\ + \Big[L_{ij}\boldsymbol{\tau}_i\cdot\boldsymbol{\tau}_j + \tilde{L}_{ij}\tau_i^y\tau_j^y\Big]\mathbf{S}_i\cdot\mathbf{S}_j\Big\}$$

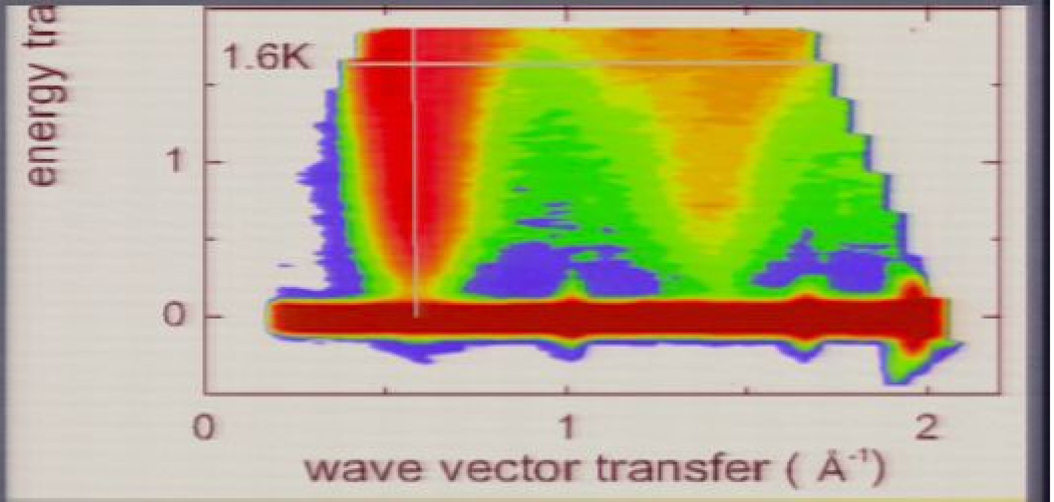
Exchange

- Largest interaction is just Heisenberg exchange

$$H_{ex} \approx \frac{1}{2}\sum_{ij} J_{ij}\mathbf{S}_i \cdot \mathbf{S}_j$$

  - More exchange processes contribute

Minimal Model

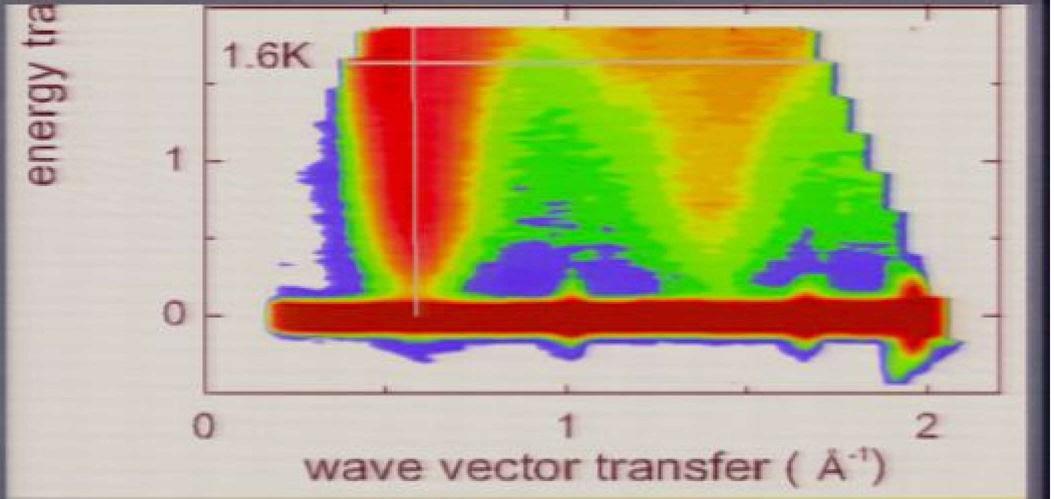


A. Krimmel *et al.*, Phys. Rev. Lett. **94**, 237402, 2005

- Neutron scattering suggests peak close to 2π(100)
- Indicates $J_2 \gg J_1$

$$H_{min} = J_2 \sum_{\langle\langle ij\rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j + H_{SO}$$

Minimal Model



A. Krimmel *et al.*, Phys. Rev. Lett. **94**, 237402, 2005

- Neutron scattering suggests peak close to $2\pi(100)$

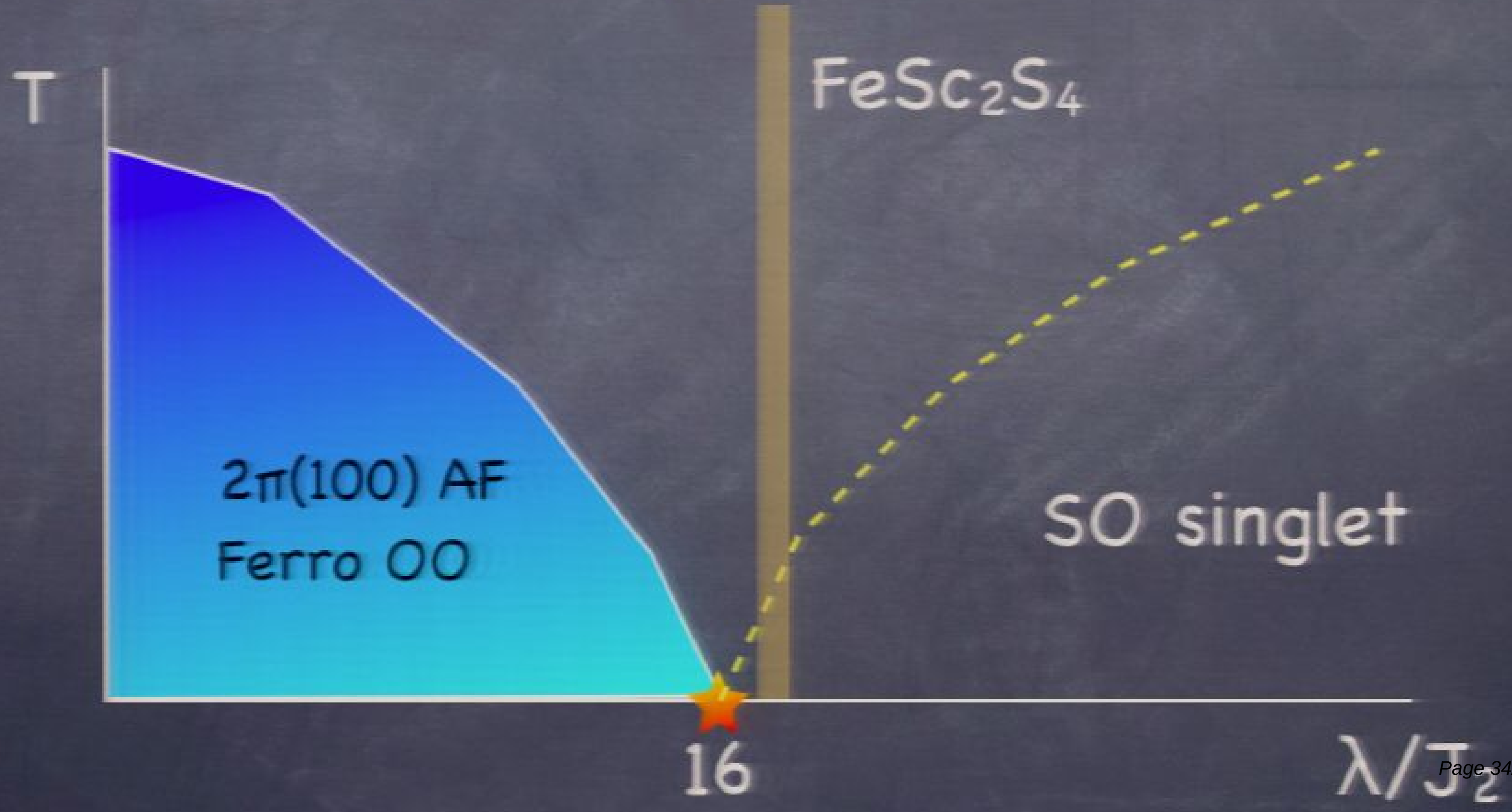
- Indicates $J_2 \gg J_1$

$$H_{min} = J_2 \sum_{\langle\langle ij \rangle\rangle} \mathbf{S}_i \cdot \langle \mathbf{S}_j \rangle + H_{SO}$$

Expect MFT good in 3+1 dimensions

Quantum Critical Point

- Mean field phase diagram

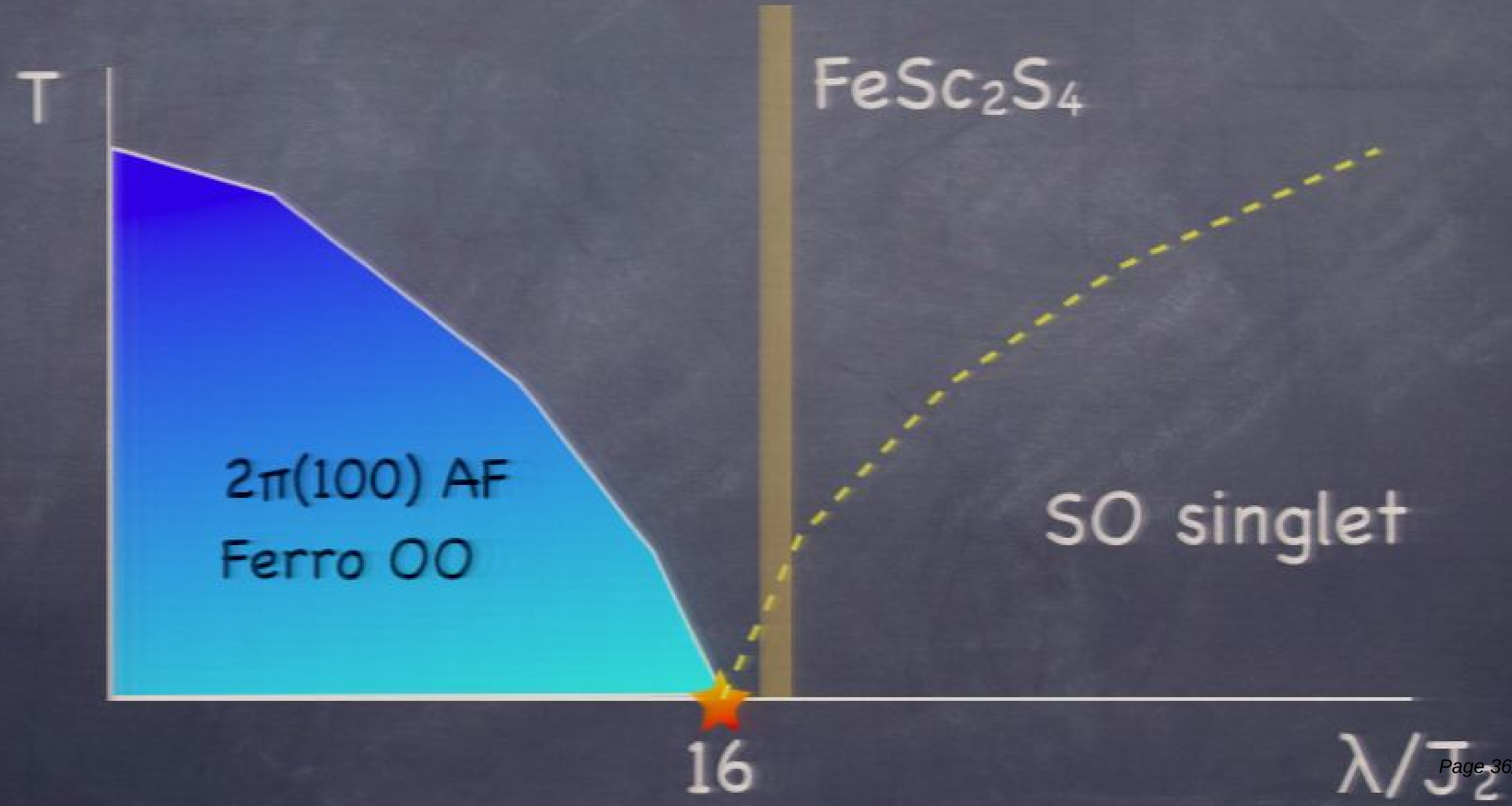


Predictions

- Large T=0 susceptibility (estimated) ✓
- Scaling form for $(T_1T)^{-1} \sim f(\Delta/T)$ ✓
- Specific heat $C_v \sim T^3 f(\Delta/T)$ ✓
- Possibility of pressure-induced ordering
- Magnetic field **suppresses** order
  - opposite to simple "dimer" antiferromagnet

Quantum Critical Point

- Mean field phase diagram



Predictions

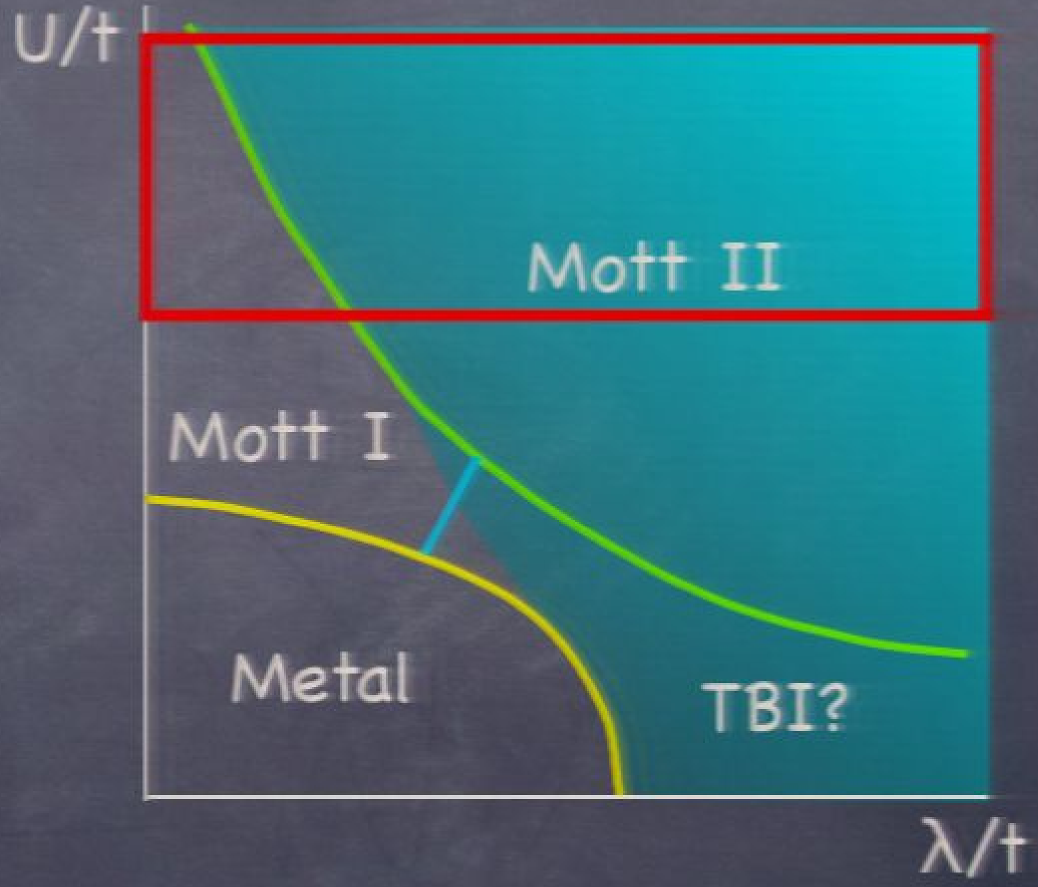
- Large $T=0$ susceptibility (estimated) ✓
- Scaling form for $(T_1T)^{-1} \sim f(\Delta/T)$ ✓
- Specific heat $C_v \sim T^3 f(\Delta/T)$ ✓
- Possibility of pressure-induced ordering
- Magnetic field **suppresses** order
  - opposite to simple “dimer” antiferromagnet

Conclusions on $FeSc_2S_4$

- Orbital degeneracy and spin orbit provides an exciting route to quantum paramagnetism and quantum criticality
  - entangled spin-orbital singlet ground state in an S=2 magnet!
  - Look in our papers for more details

Mott transition with SOIs

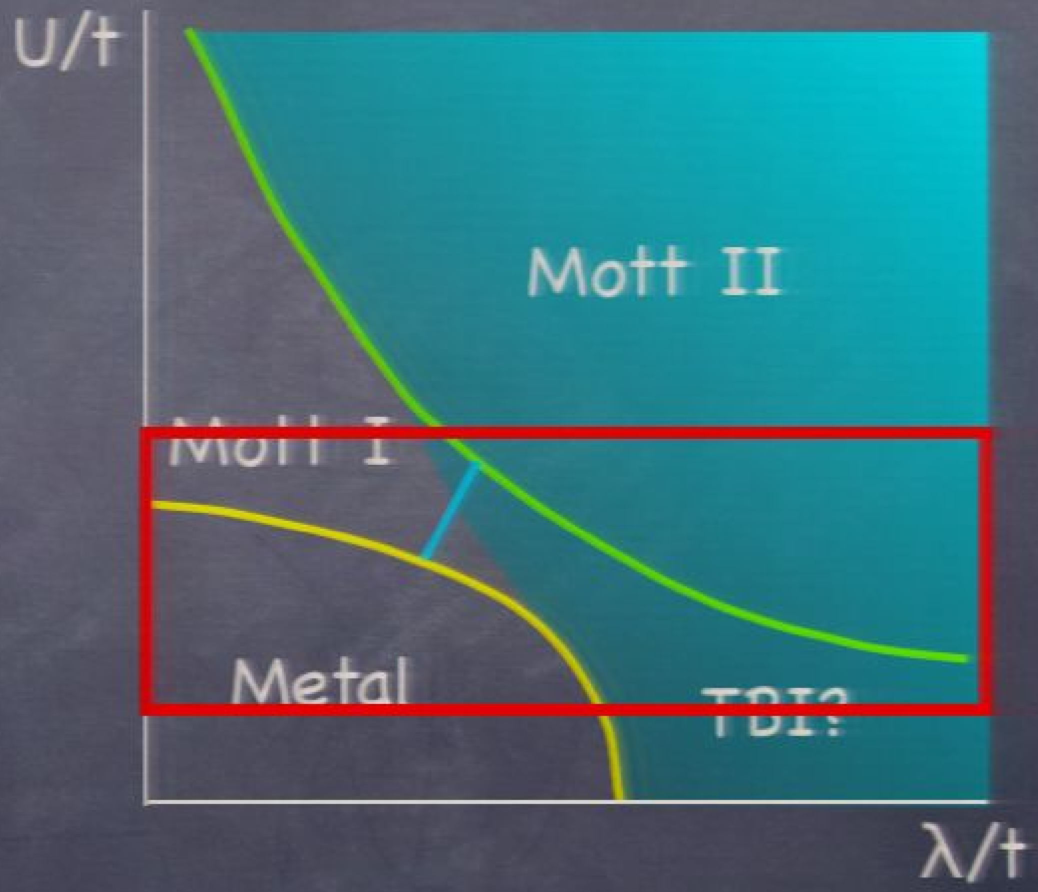
- Study this phase diagram in a concrete case



schematic phase diagram

Mott transition with SOIs

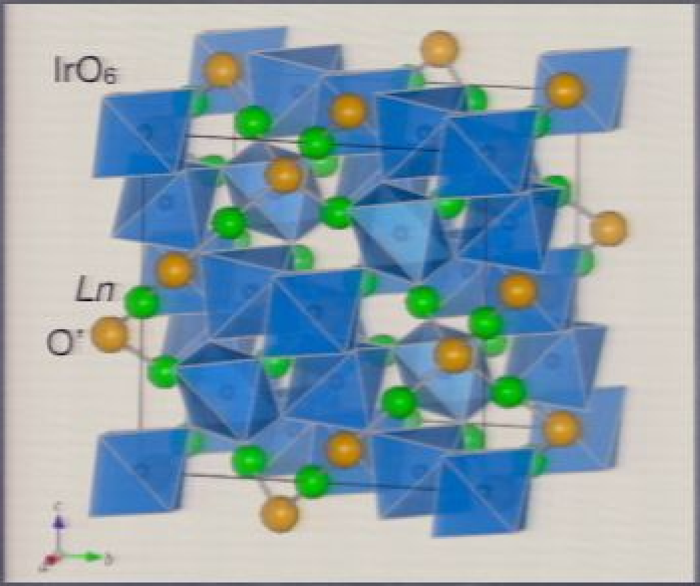
- Study this phase diagram in a concrete case

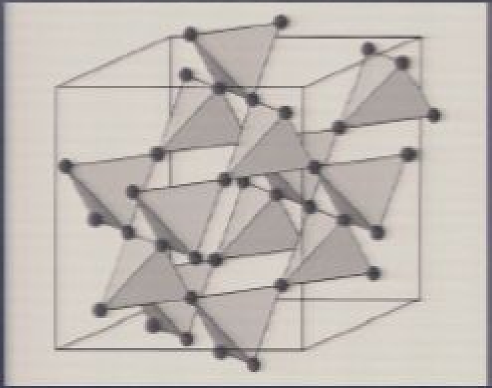


schematic phase diagram

Pyrochlore iridates

- Formula: $Ln_2Ir_2O_7$
    - both Ln and Ir atoms occupy pyrochlore lattices
    - Cubic, FCC Bravais lattice
- Ln carry localized moments only important at low T

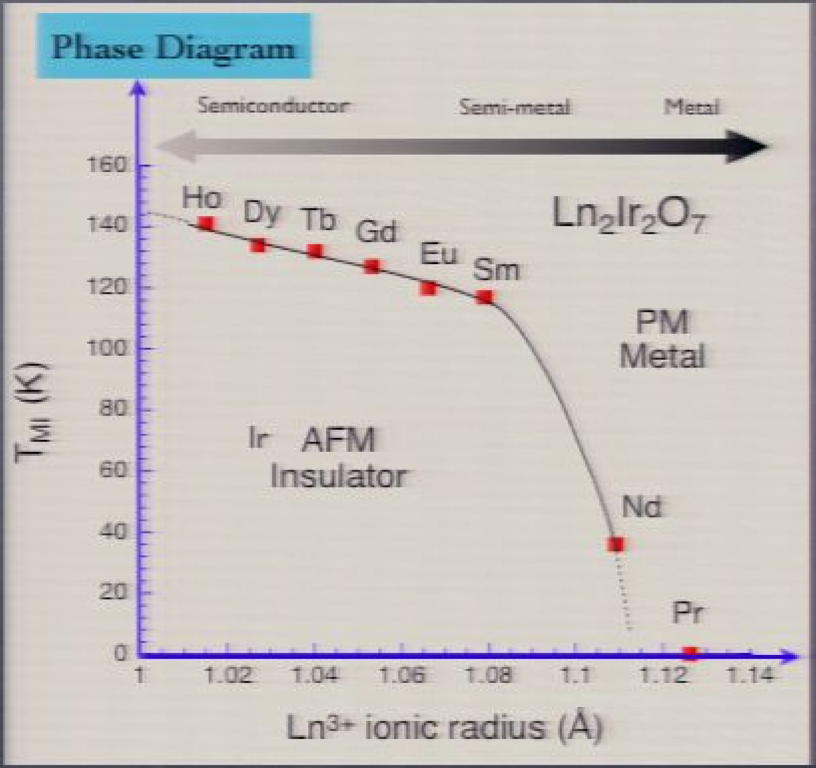




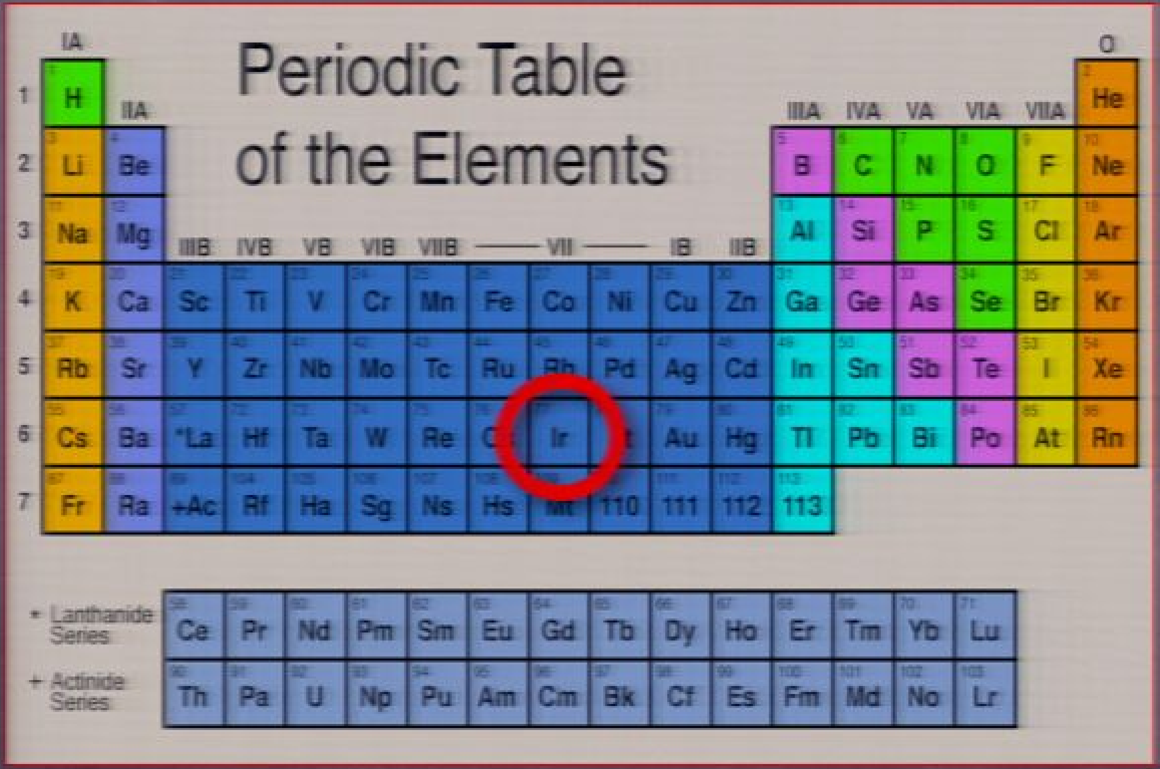
Metal-Insulator Transition

K. Matsuhira et al, 2007

- Decreasing Ir-O-Ir bond angle makes more insulating



Spin orbit coupling



Periodic Table of the Elements

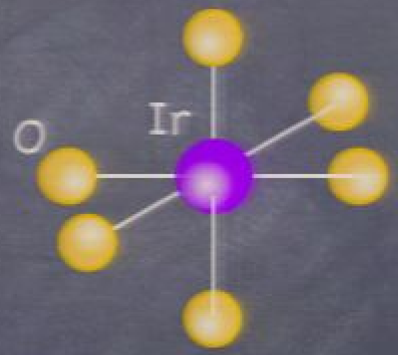
	IA	IIA	IIIB	IVB	VB	VIB	VIIB	— VII	—	—	IB	IIB	IIIA	IVA	VA	VIA	VIIA	O
1	H																	He
2	Li	Be											B	C	N	O	F	Ne
3	Na	Mg											Al	Si	P	S	Cl	Ar
4	K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
5	Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
6	Cs	Ba	*La	Hf	Ta	W	Re	[illegible]	Ir	t	Au	Hg	Tl	Pb	Bi	Po	At	Rn
7	Fr	Ra	+Ac	Rf	Ha	Sg	Ns	Hs	Mt	110	111	112	113					

* Lanthanide Series	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
+ Actinide Series	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr

- Estimate (?) $\lambda \approx 0.5$ eV

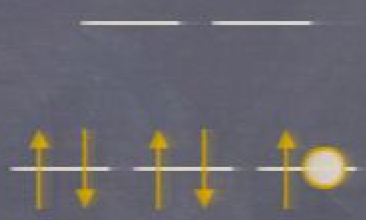
Model

- octahedral Ir^{4+}: $(t_{2g})^5$
  - effective l=1 orbital degeneracy
- Ir–O–Ir hopping
  - dominant $V_{pd\pi}$ channel
- Spin-orbit coupling
  - $H_{SOI} = -\lambda \vec{L} \cdot \vec{S}$
- Hubbard U

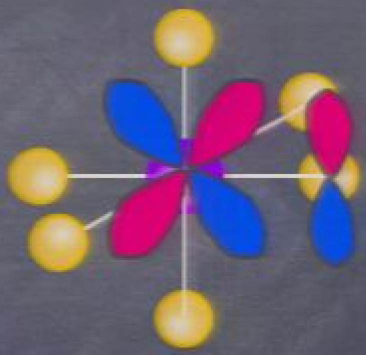


Model

- octahedral Ir^{4+}: $(t_{2g})^5$
  - effective l=1 orbital degeneracy



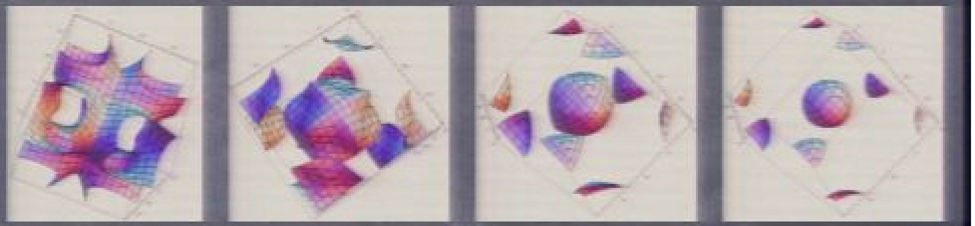
- Ir–O–Ir hopping
  - dominant $V_{pd\pi}$ channel

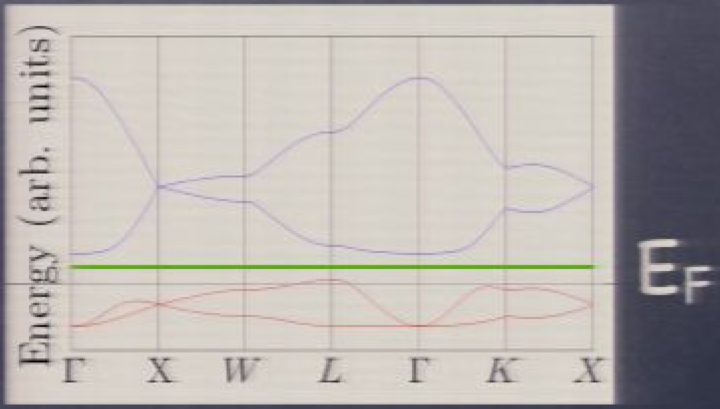


- Spin-orbit coupling
  - $H_{SOI} = -\lambda \vec{L} \cdot \vec{S}$
- Hubbard U

U=0 Band Structure

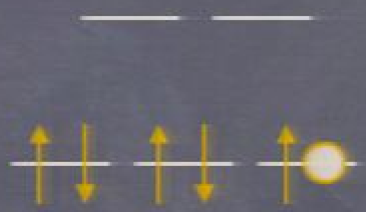
- 3 x 4 = 12 doubly degenerate bands
- λ<2.8t: overlap at Fermi energy: metal
- λ>2.8t: bands separate
    - only j=1/2 states near Fermi energy



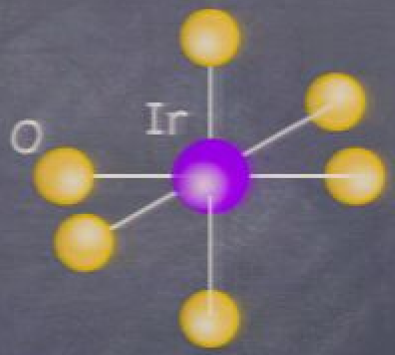


Model

- octahedral Ir^{4+}: $(t_{2g})^5$
  - effective l=1 orbital degeneracy



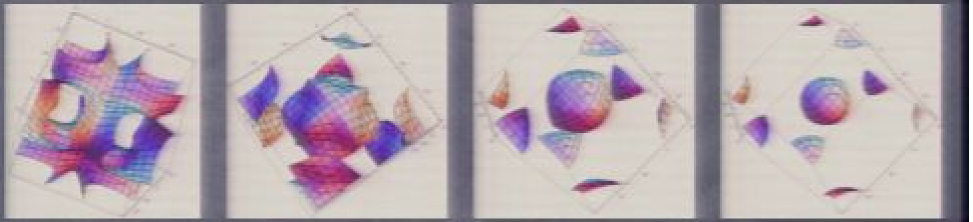
- Ir–O–Ir hopping
  - dominant $V_{pd\pi}$ channel

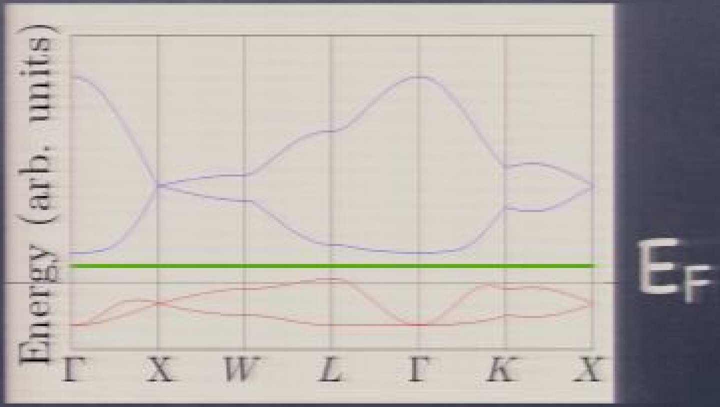


- Spin-orbit coupling
  - $H_{SOI} = -\lambda \vec{L} \cdot \vec{S}$
- Hubbard U

U=0 Band Structure

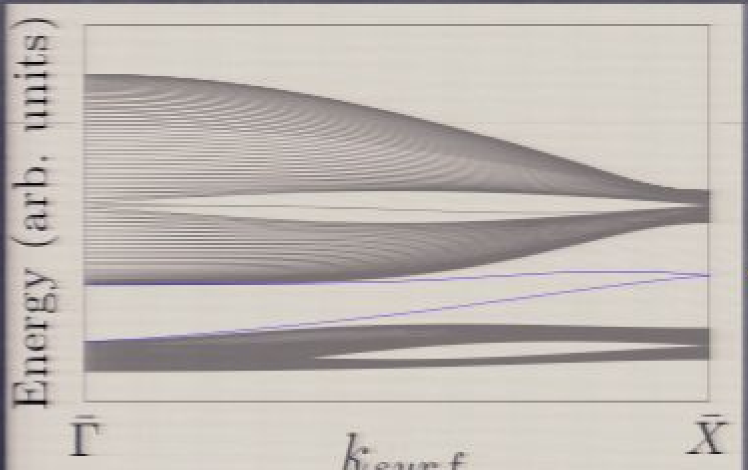
- 3 x 4 = 12 doubly degenerate bands
- λ<2.8t: overlap at Fermi energy: metal
- λ>2.8t: bands separate
    - only j=1/2 states near Fermi energy





Topological Band Insulator

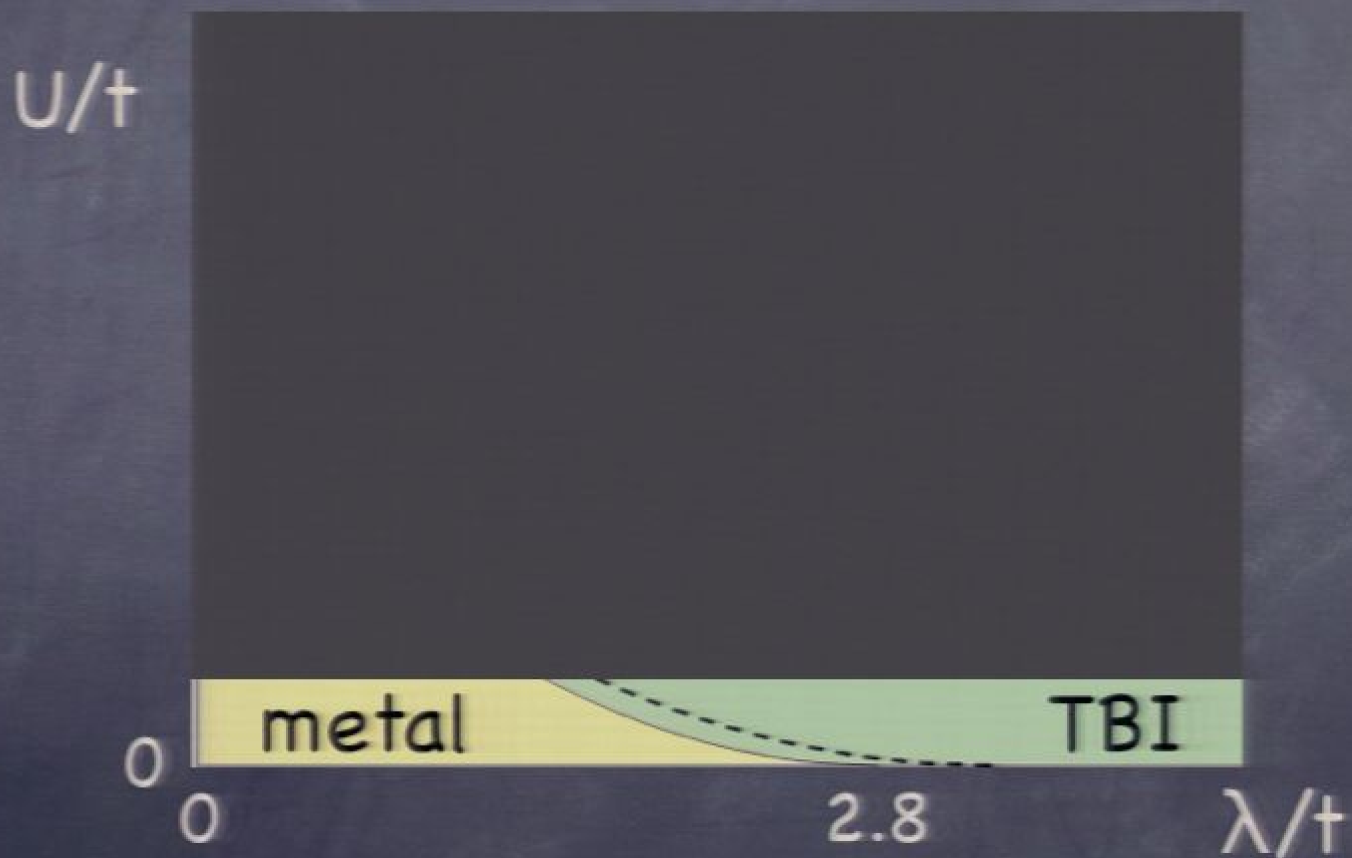
- Inversion Symmetry:
  - Fu-Kane give simple criterion for parity eigenvalues
  - Strong TBI (weak invariants all zero by cubic symmetry)
- Surface states



(100) surface

surface Dirac point

Phase Diagram

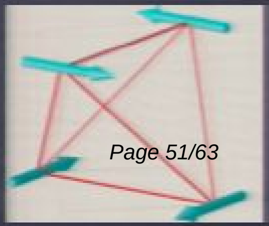


Very large U/t

- For λ >> J ~ t^2/U, reduces to Heisenberg "spin" model for j=1/2 eigenstates

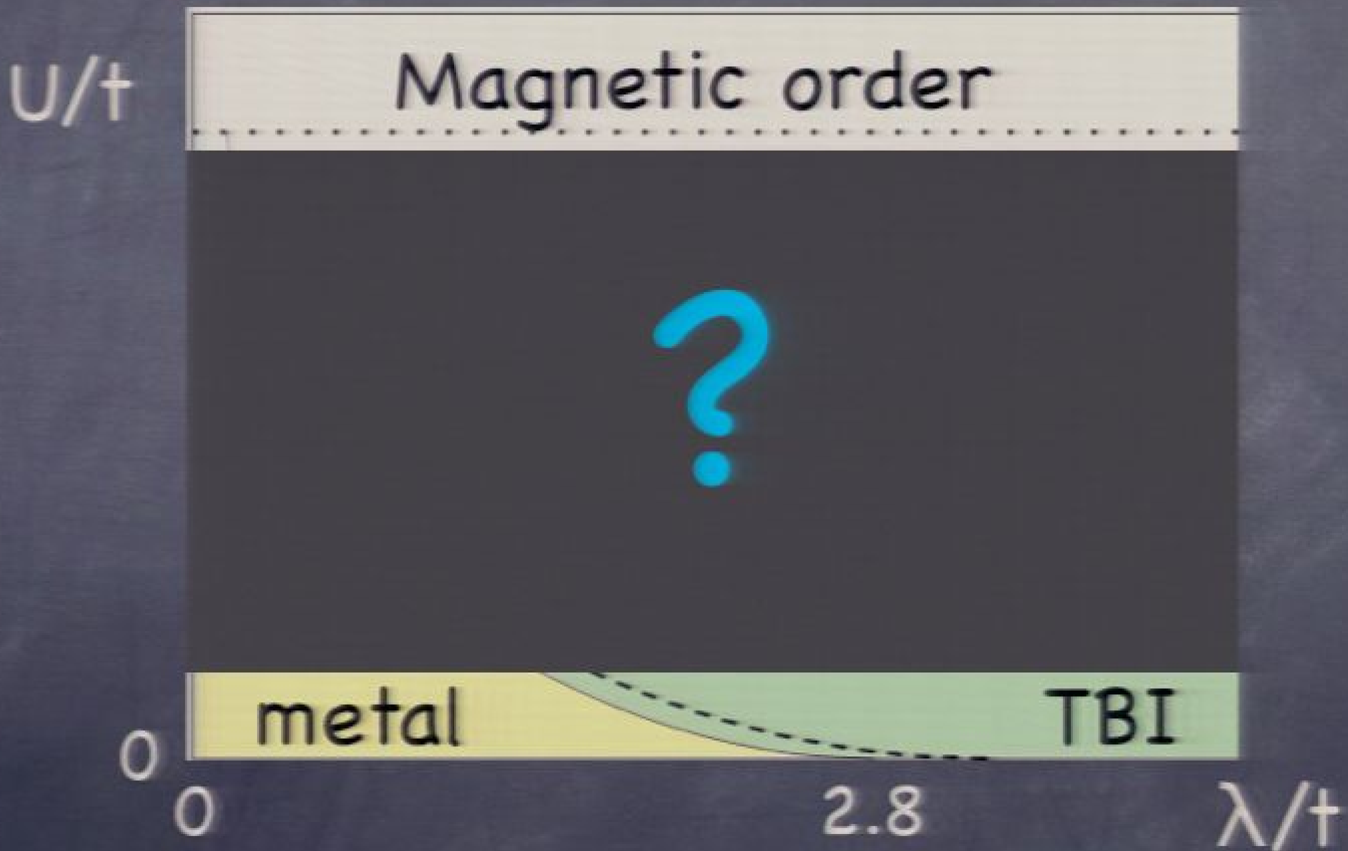
$$H_{spin} = \frac{4t^2}{U} \sum_{ii'} \left[J\vec{S}_i \cdot \vec{S}_{i'} + \vec{D}_{ii'} \cdot \vec{S}_i \times \vec{S}_{i'} + \vec{S}_i \cdot \overleftrightarrow{\Gamma}_{ii'} \cdot \vec{S}_{i'} \right]$$

- This model has been extensively studied Elhajal et al, 2005
  - Axis of D-vector fixed by symmetry
  - very large DM: $|D|/J = \frac{5460}{12283}\sqrt{2} \approx 0.63$
- Ground state for |D|/J > 0.3 is definitely magnetically ordered



  - Q=0 magnetic state

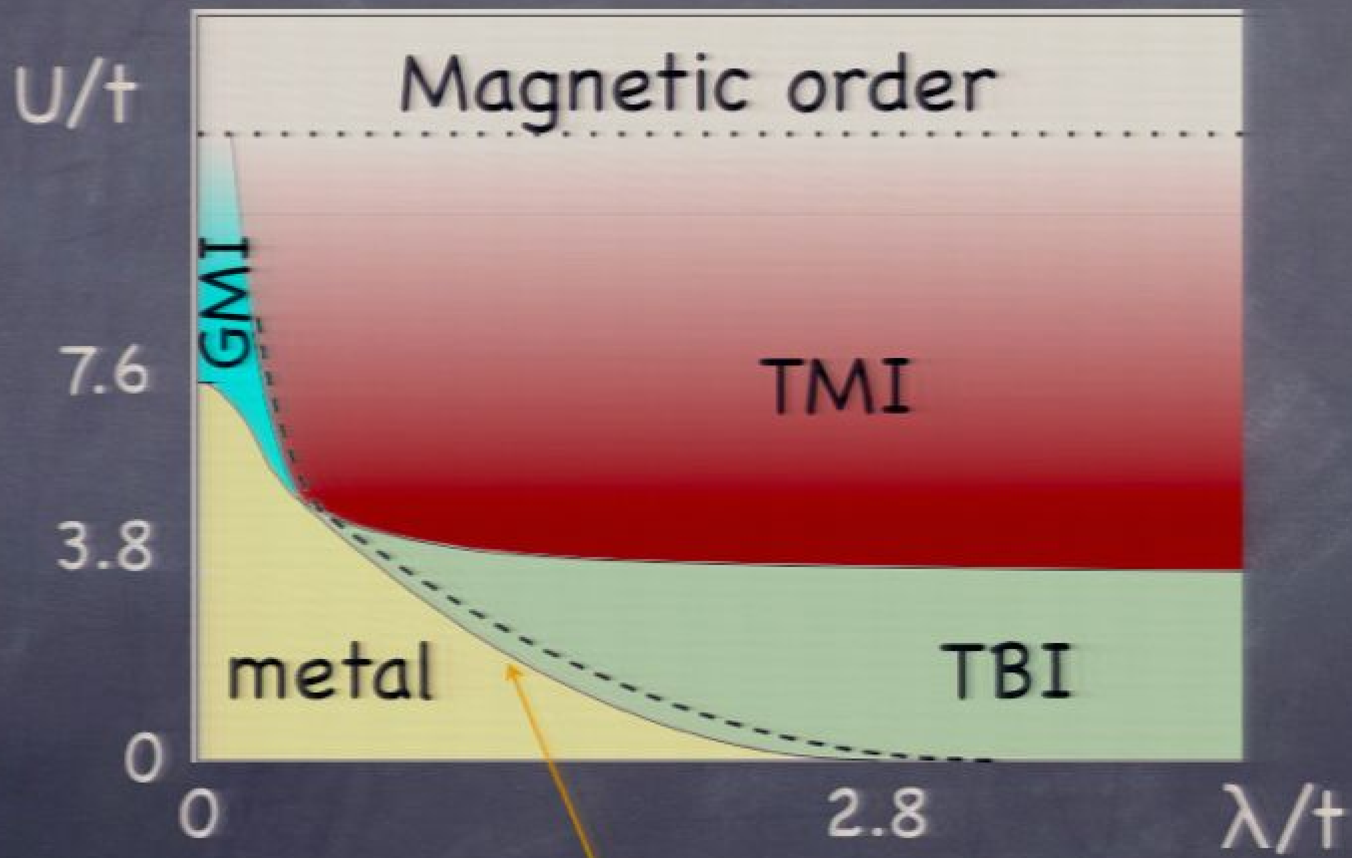
Phase Diagram



Intermediate U

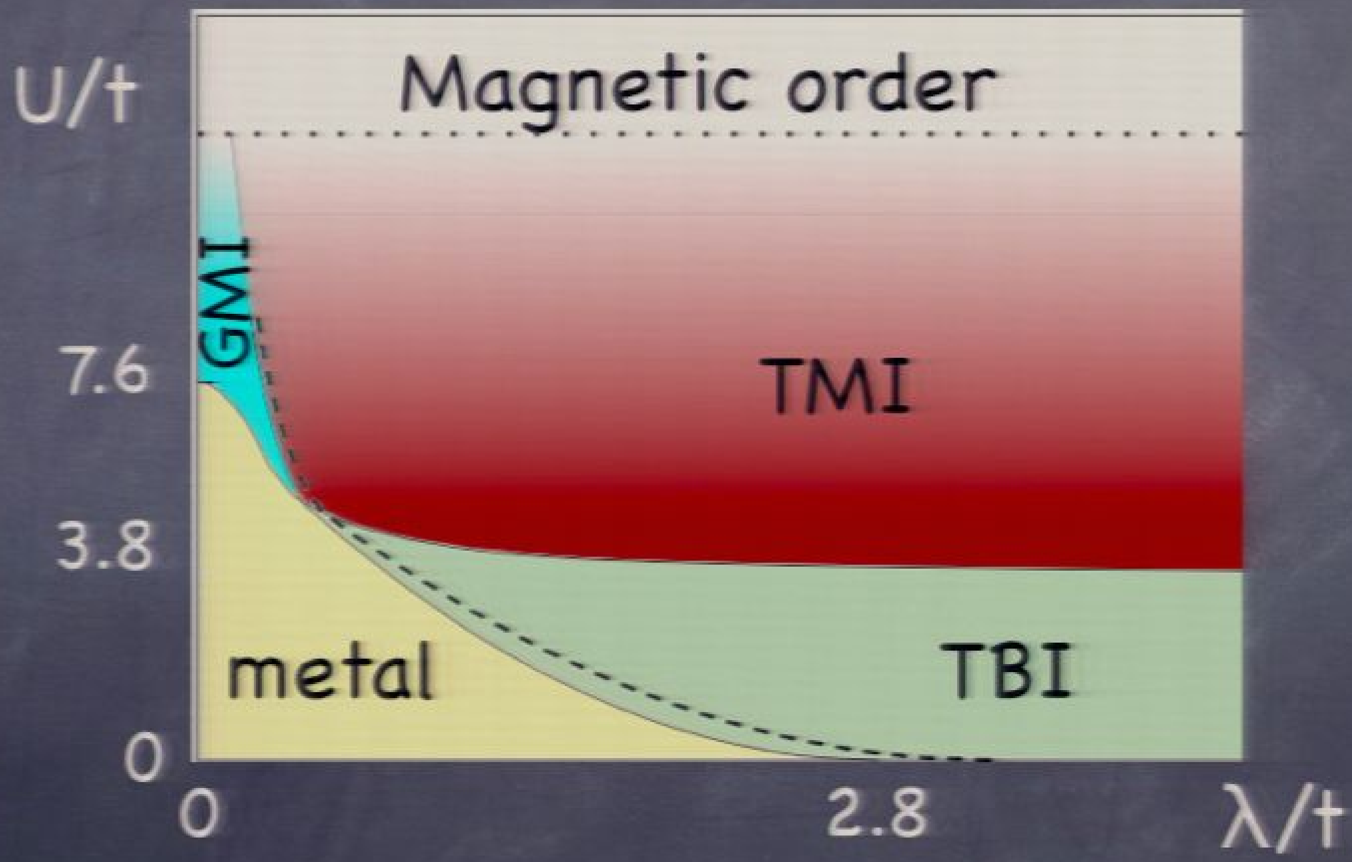
- Slave-rotor approximation Florens, Georges (2004)
  - Seems to give qualitatively reasonable results for frustrated Hubbard models (triangular, checkerboard, hyperkagome) in agreement with several numerical approaches
  - Does **not** describe nesting/SDW physics
- Simple to implement $c_a^\dagger = e^{i\theta} f_a^\dagger$
  - Decouple to produce independent MF dynamics for rotors (charge) and spinons
  - Should be solved self-consistently

Phase Diagram



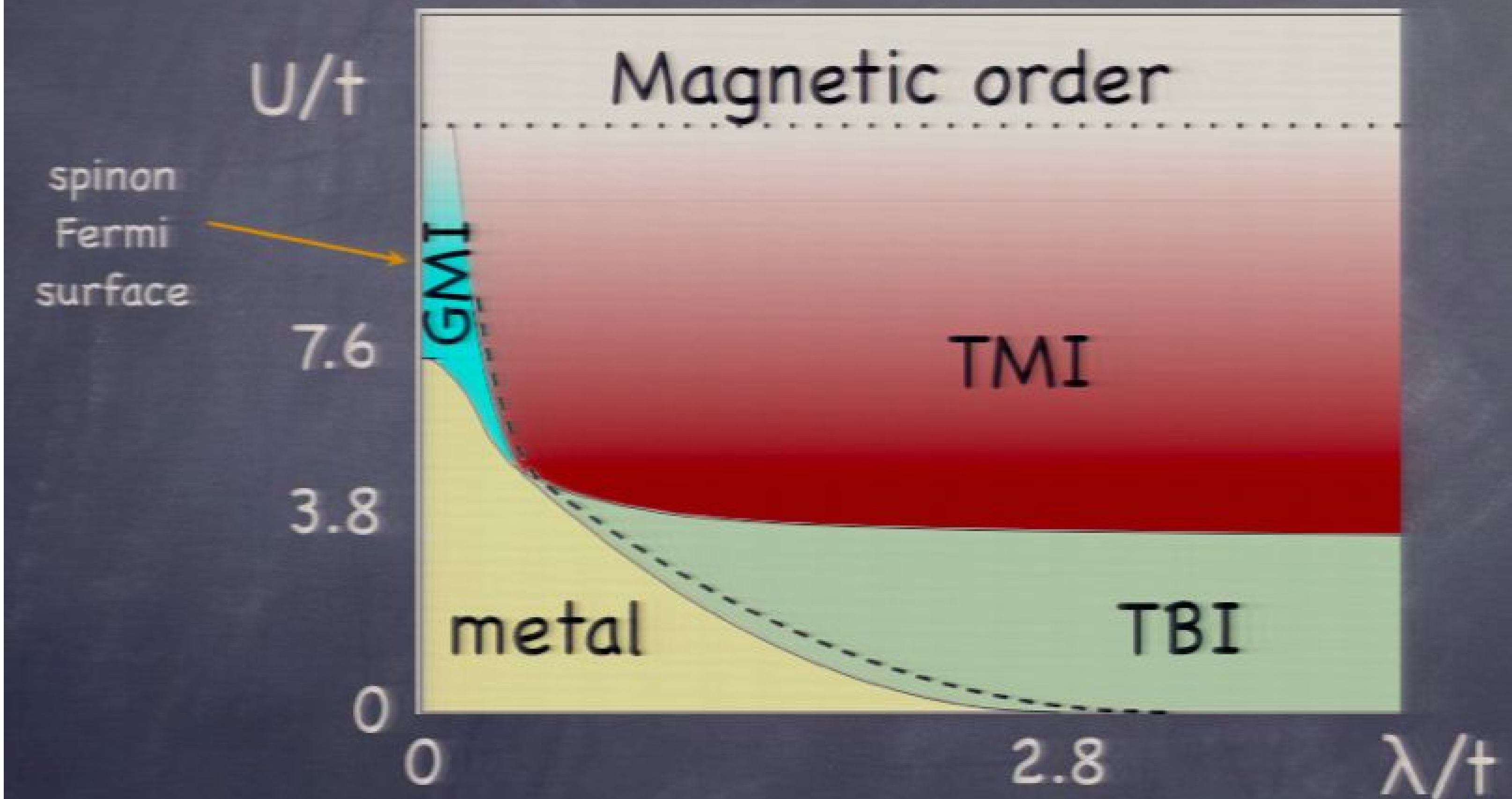
Bandwidth suppression

Phase Diagram

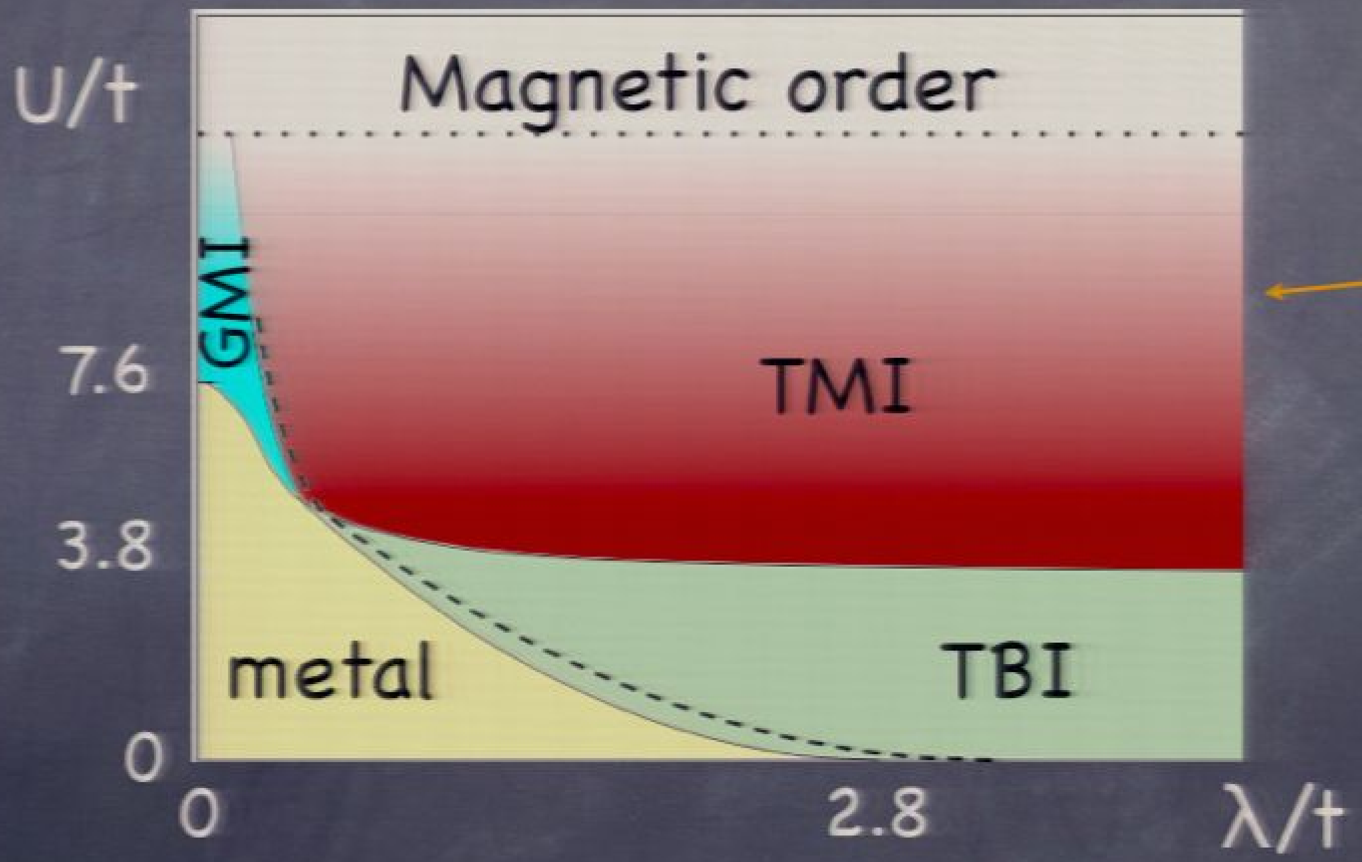


rotors
gapped
in MIs

Phase Diagram



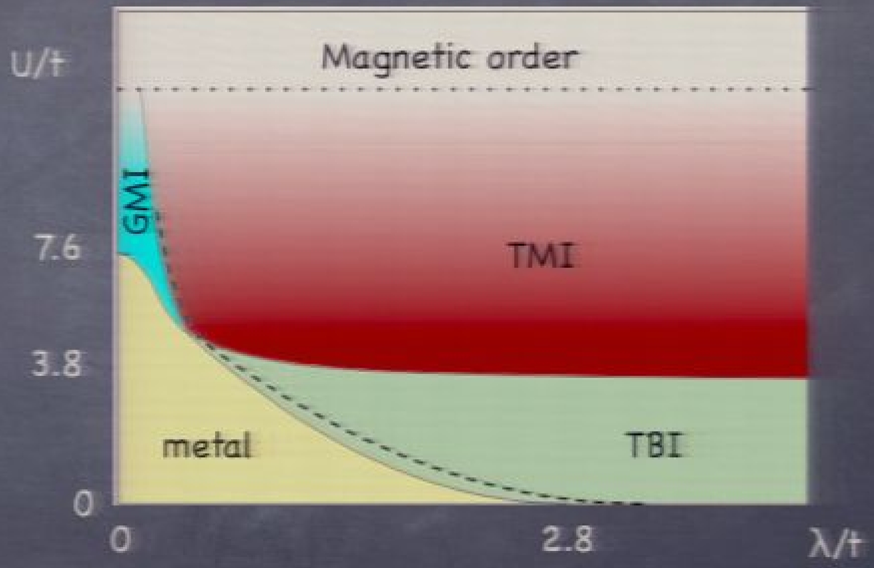
Phase Diagram



spinons in Z_2 TBI state

Topological Mott Insulator

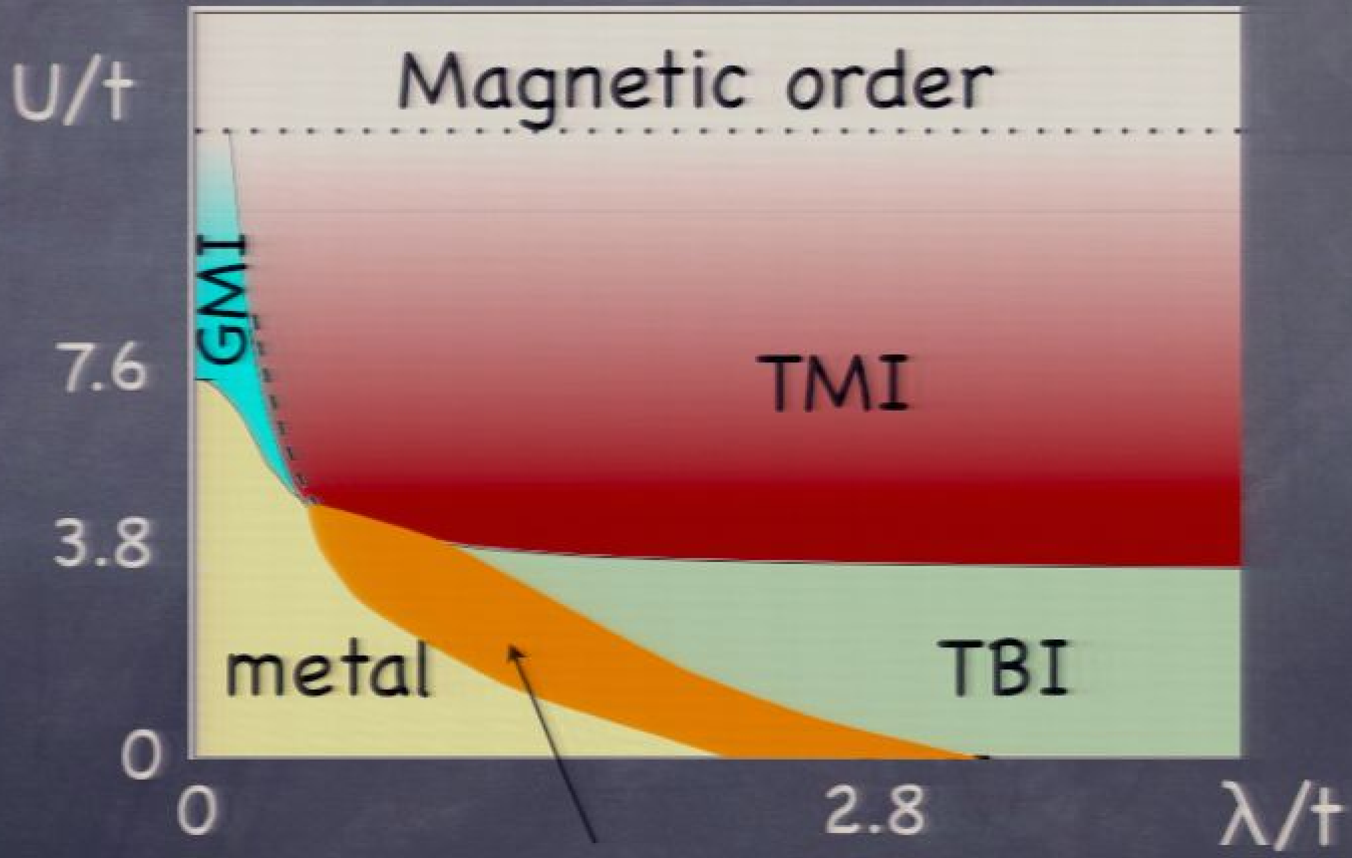
- A U(1) spin liquid
  - Gapless photon
  - Stable only in 3d
- Gapless "topological spin metal" at surface
- Magnetic monopole excitations carry spin or charge?



metal-TBI transition

- Long-range Coulomb: excitons

c.f. Halperin, Rice (1968)

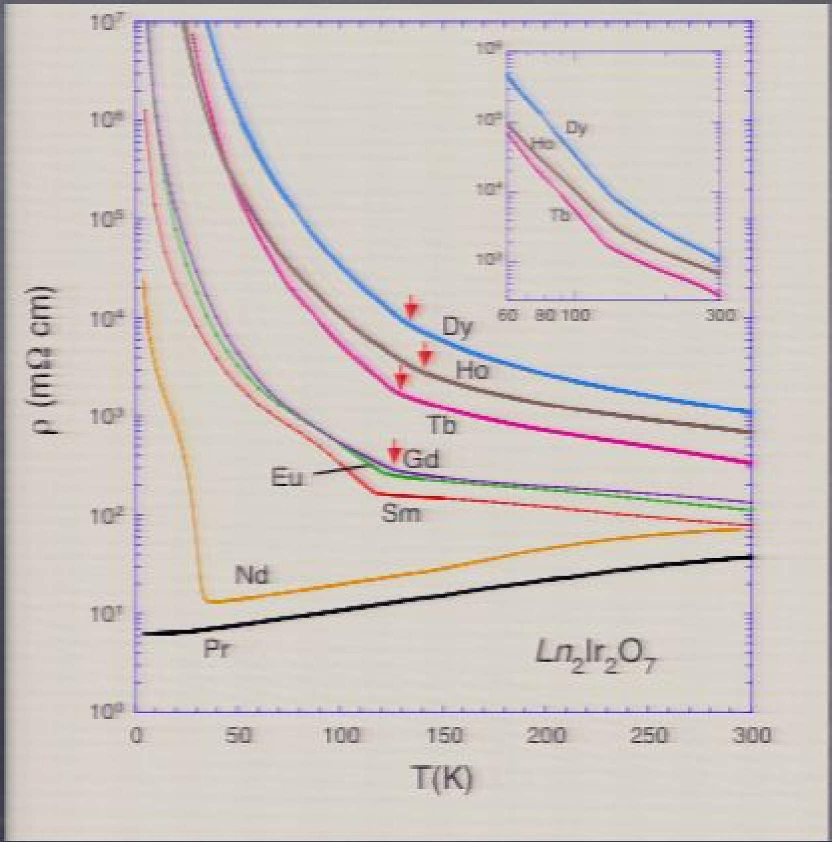


probably weakly magnetic

Back to iridates

K. Matsuhira et al, 2007

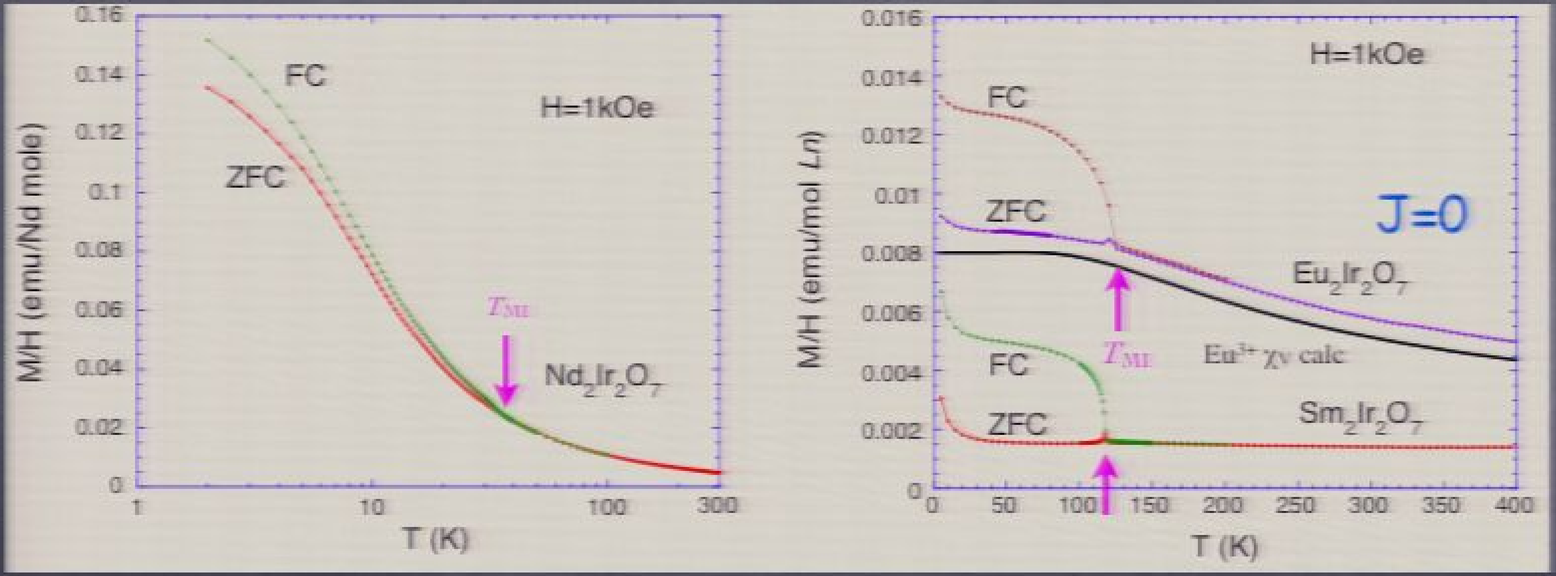
- Experiments show continuous T>0 MITs



Back to iridates

K. Matsuhira et al, 2007

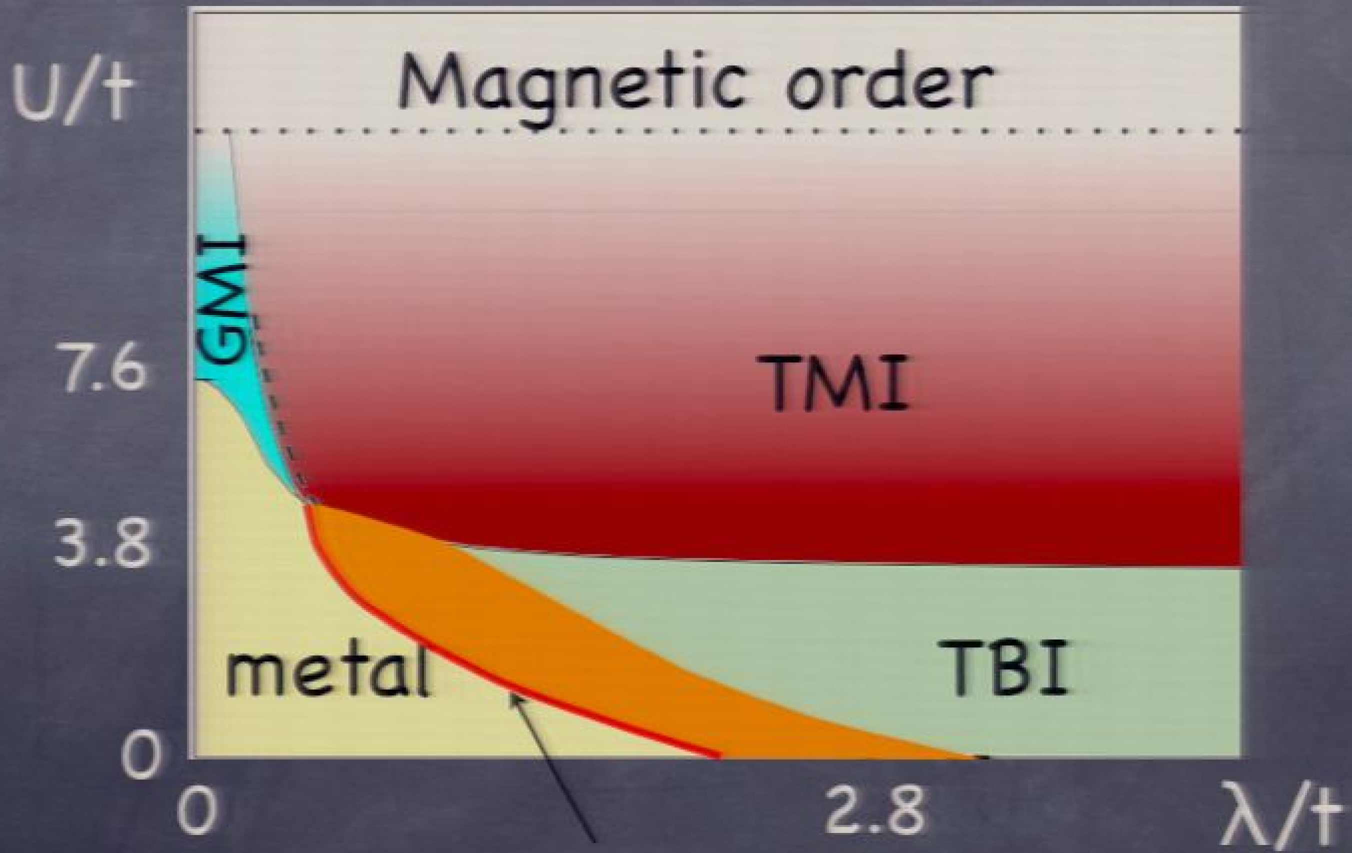
- Experiments show continuous T>0 MITs



closest to QCP

metal–TBI transition

- Perhaps consistent with an excitonic state?



this transition? probably too optimistic!

Conclusions

- Spin-orbit interactions become **increasingly** important with increased correlations due to reduction in effective bandwidth
  - especially true in situations with orbital degeneracy
- Interesting new phases and transitions possible in 5d TMOs
  - How long until interacting versions of TIs are discovered?

Reference - $FeSc_2S_4$: PRL 102, 096406 (2009), arXiv:0907.1692

Mott+SO: arXiv:0908.2962