Spin-orbit physics and the Mott regime

Leon Balents, KITP

CDF Seminar, June 2010

The David and Lucile Packard Foundation

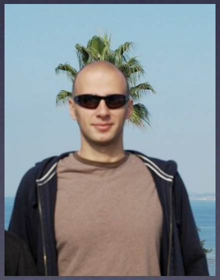
Collaborators

- $FeSc_2S_4$:

Gang Chen

Andreas Schnyder
MPI Stuttgart

- Mott transition (pyrochlore iridates)



Dymtro Pesin
UT Austin

Outline

1. Introduction to SOC in solids, and the recent discovery of topological insulators
2. SOC deep in the Mott regime
3. SOC near the Mott transition, and iridium oxides

Spin-orbit coupling

$$H_{SOC} = \lambda \mathbf{L} \cdot \mathbf{S}$$

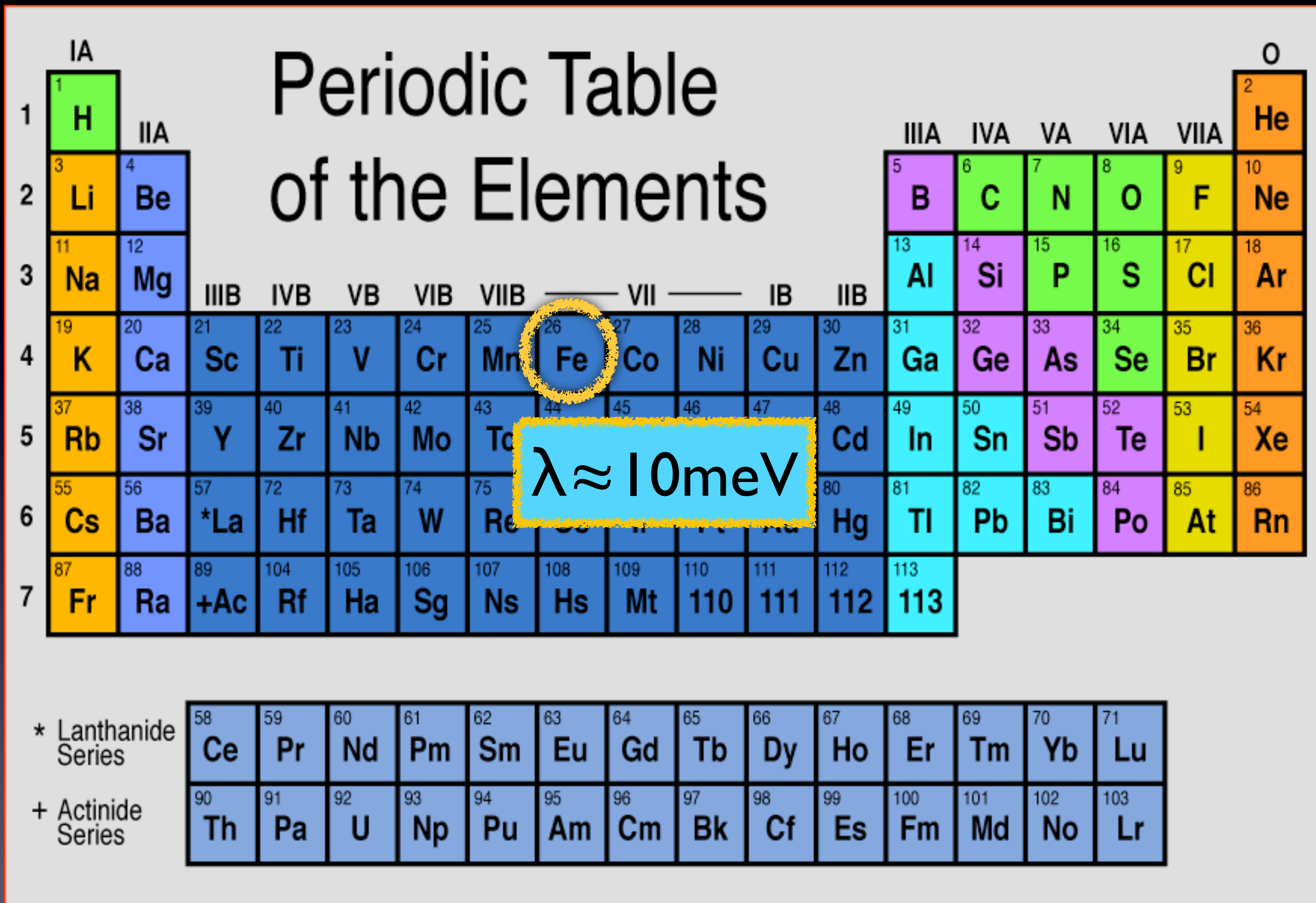
- This is a *relativistic* effect, usually considered weak
  - responsible for the "fine structure" of atomic spectra
- Typically not considered very significant in solids
- But there are exceptions, and the exceptions are interesting...

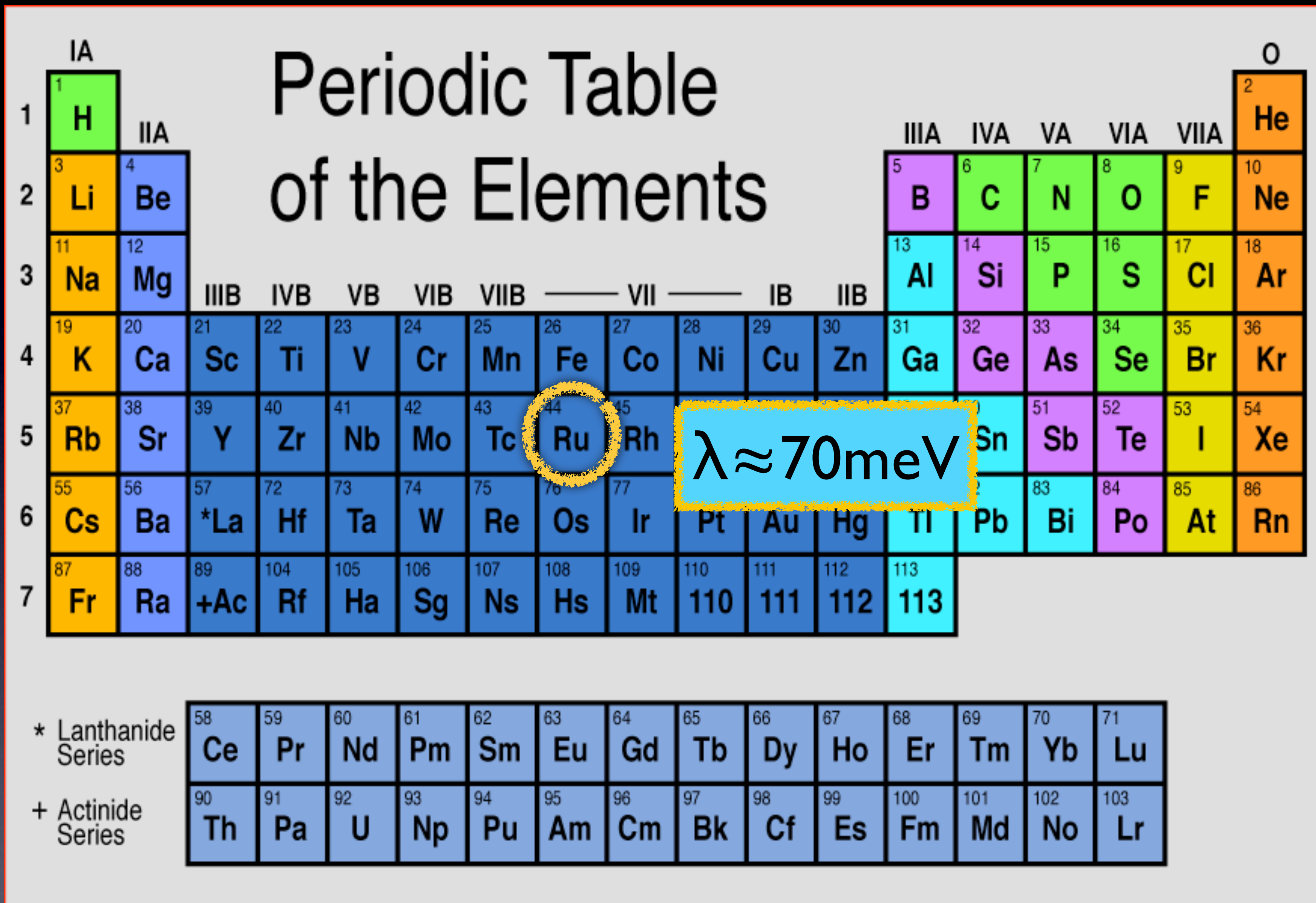
When is SOC important?

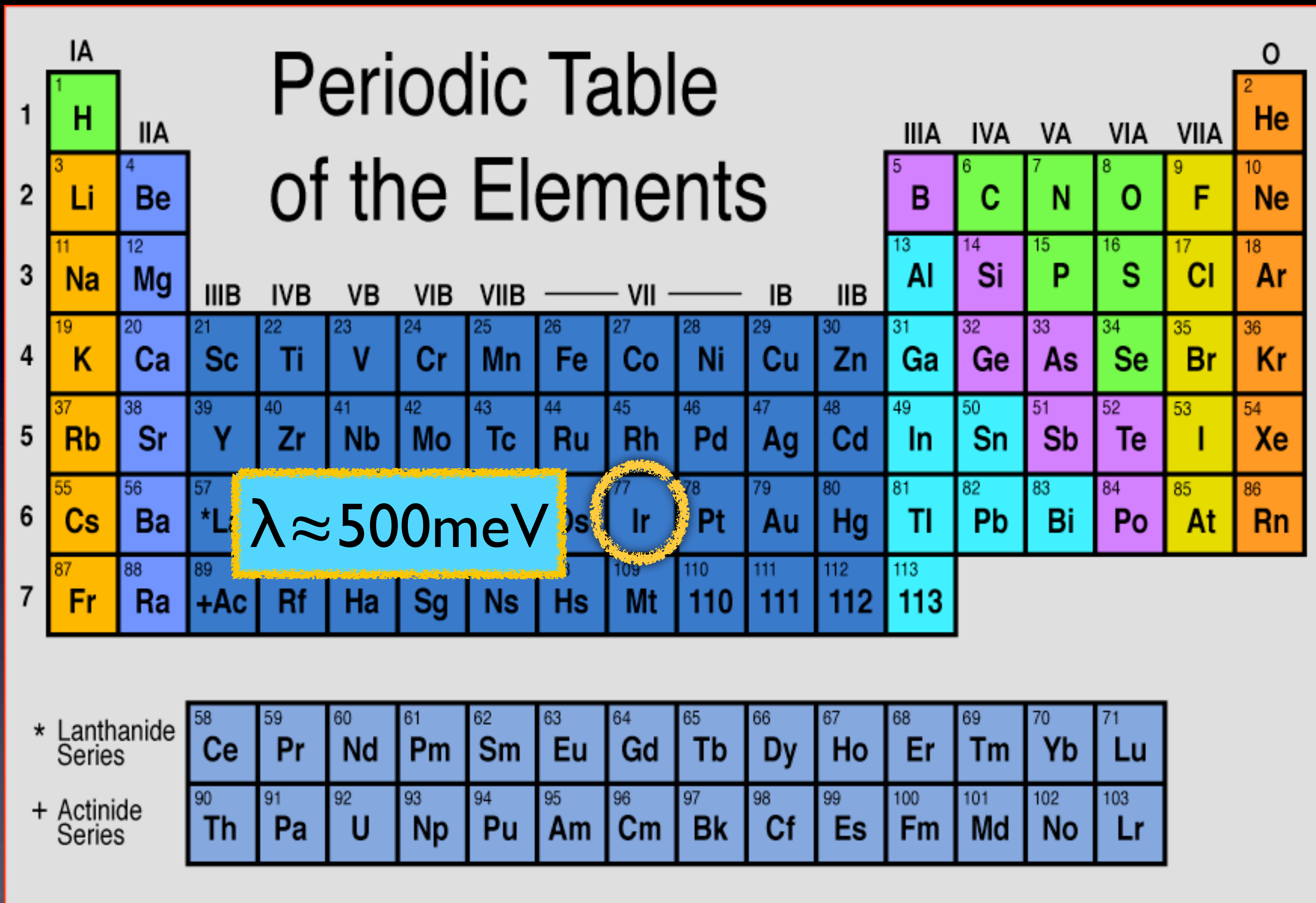
- Phenomena where spin-rotational symmetry is broken
  - Magnetic anisotropy
  - Spintronics - e.g. spin Hall effect in semiconductors
  - Spin relaxation
- In these situations, SOC can still be treated as weak, usually

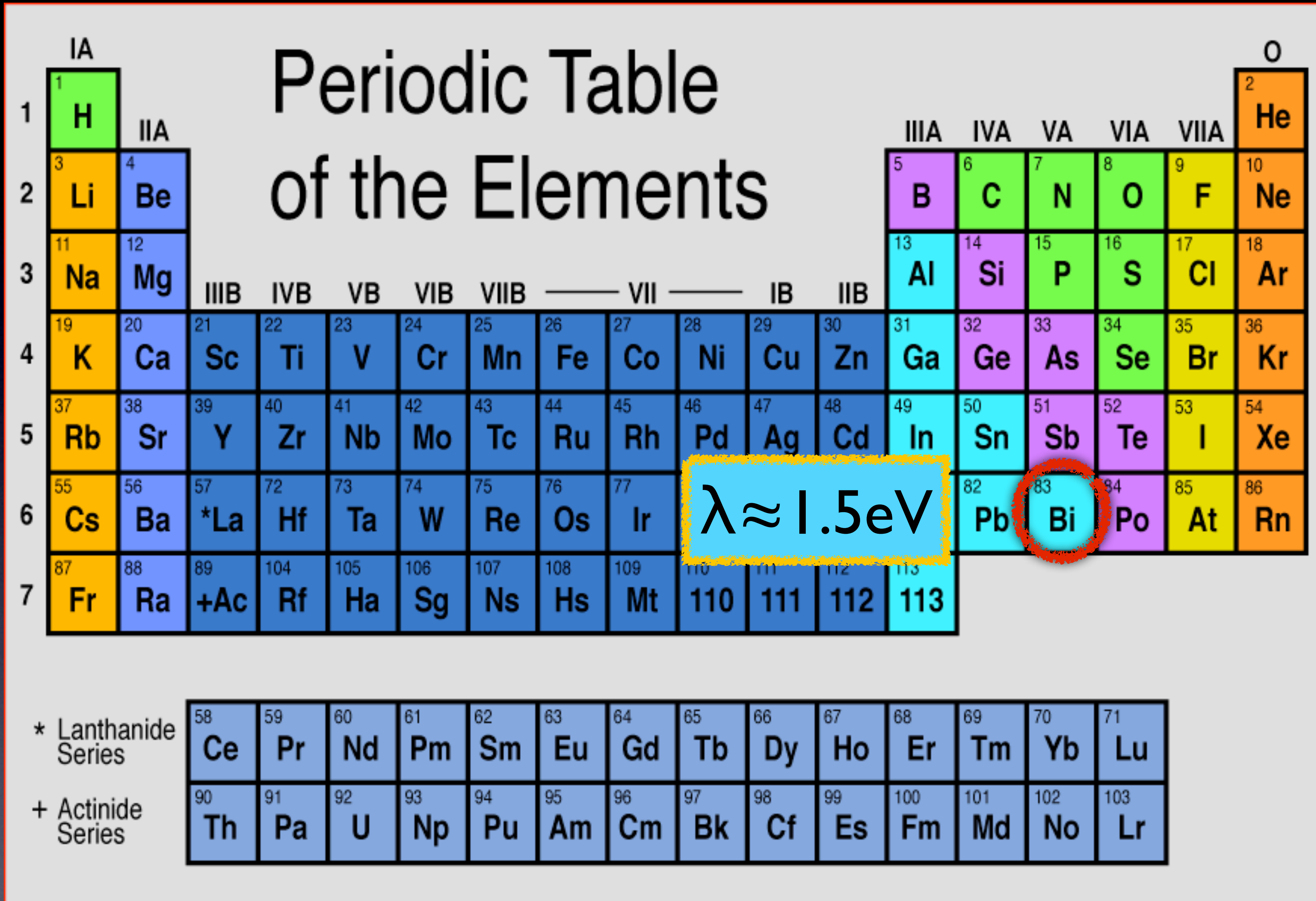
Strong SOC?

- Atomic SOC grows with atomic number
  - $\lambda \sim Z^4$, where Z is atomic number
- Typical values?


Periodic Table
of the Elements
λ≈10meV
IA
IIA
IIIB IVB VB VIB VIIB — VII — IB IIB
IIIA IVA VA VIA VIIA
O
1 H
2 He
3 Li 4 Be
5 B 6 C 7 N 8 O 9 F 10 Ne
11 Na 12 Mg
13 Al 14 Si 15 P 16 S 17 Cl 18 Ar
19 K 20 Ca 21 Sc 22 Ti 23 V 24 Cr 25 Mn 26 Fe 27 Co 28 Ni 29 Cu 30 Zn 31 Ga 32 Ge 33 As 34 Se 35 Br 36 Kr
37 Rb 38 Sr 39 Y 40 Zr 41 Nb 42 Mo 43 Tc 44 45 46 47 48 Cd 49 In 50 Sn 51 Sb 52 Te 53 I 54 Xe
55 Cs 56 Ba 57 *La 72 Hf 73 Ta 74 W 75 Re 80 Hg 81 Tl 82 Pb 83 Bi 84 Po 85 At 86 Rn
87 Fr 88 Ra 89 +Ac 104 Rf 105 Ha 106 Sg 107 Ns 108 Hs 109 Mt 110 110 111 111 112 112 113 113
* Lanthanide Series
58 Ce 59 Pr 60 Nd 61 Pm 62 Sm 63 Eu 64 Gd 65 Tb 66 Dy 67 Ho 68 Er 69 Tm 70 Yb 71 Lu
+ Actinide Series
90 Th 91 Pa 92 U 93 Np 94 Pu 95 Am 96 Cm 97 Bk 98 Cf 99 Es 100 Fm 101 Md 102 No 103 Lr


Periodic Table
of the Elements
λ≈70meV
* Lanthanide Series
+ Actinide Series

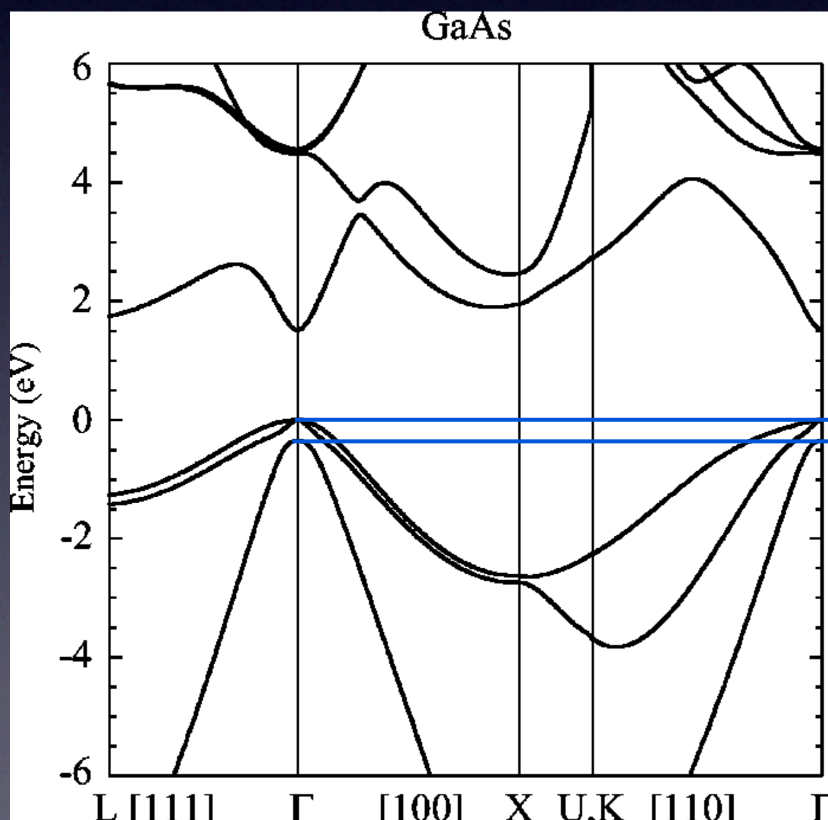

Periodic Table
of the Elements
λ≈500meV
IA
IIA
IIIB IVB VB VIB VIIB VII IB IIB
IIIA IVA VA VIA VIIA O
1 H
2 He
3 Li 4 Be 5 B 6 C 7 N 8 O 9 F 10 Ne
11 Na 12 Mg 13 Al 14 Si 15 P 16 S 17 Cl 18 Ar
19 K 20 Ca 21 Sc 22 Ti 23 V 24 Cr 25 Mn 26 Fe 27 Co 28 Ni 29 Cu 30 Zn 31 Ga 32 Ge 33 As 34 Se 35 Br 36 Kr
37 Rb 38 Sr 39 Y 40 Zr 41 Nb 42 Mo 43 Tc 44 Ru 45 Rh 46 Pd 47 Ag 48 Cd 49 In 50 Sn 51 Sb 52 Te 53 I 54 Xe
55 Cs 56 Ba 57 77 Ir 78 Pt 79 Au 80 Hg 81 Tl 82 Pb 83 Bi 84 Po 85 At 86 Rn
87 Fr 88 Ra 89 +Ac Rf Ha Sg Ns Hs 109 Mt 110 110 111 111 112 112 113 113
* Lanthanide Series
58 Ce 59 Pr 60 Nd 61 Pm 62 Sm 63 Eu 64 Gd 65 Tb 66 Dy 67 Ho 68 Er 69 Tm 70 Yb 71 Lu
+ Actinide Series
90 Th 91 Pa 92 U 93 Np 94 Pu 95 Am 96 Cm 97 Bk 98 Cf 99 Es 100 Fm 101 Md 102 No 103 Lr


Periodic Table of the Elements
IA IIA IIIB IVB VB VIB VIIB VII IB IIB IIIA IVA VA VIA VIIA O
1 1 H 2 He
2 3 Li 4 Be 5 B 6 C 7 N 8 O 9 F 10 Ne
3 11 Na 12 Mg 13 Al 14 Si 15 P 16 S 17 Cl 18 Ar
4 19 K 20 Ca 21 Sc 22 Ti 23 V 24 Cr 25 Mn 26 Fe 27 Co 28 Ni 29 Cu 30 Zn 31 Ga 32 Ge 33 As 34 Se 35 Br 36 Kr
5 37 Rb 38 Sr 39 Y 40 Zr 41 Nb 42 Mo 43 Tc 44 Ru 45 Rh 46 Pd 47 Ag 48 Cd 49 In 50 Sn 51 Sb 52 Te 53 I 54 Xe
6 55 Cs 56 Ba 57 *La 72 Hf 73 Ta 74 W 75 Re 76 Os 77 Ir 82 Pb 83 Bi 84 Po 85 At 86 Rn
7 87 Fr 88 Ra 89 +Ac 104 Rf 105 Ha 106 Sg 107 Ns 108 Hs 109 Mt 110 111 112 113
λ≈1.5eV
* Lanthanide Series 58 Ce 59 Pr 60 Nd 61 Pm 62 Sm 63 Eu 64 Gd 65 Tb 66 Dy 67 Ho 68 Er 69 Tm 70 Yb 71 Lu
+ Actinide Series 90 Th 91 Pa 92 U 93 Np 94 Pu 95 Am 96 Cm 97 Bk 98 Cf 99 Es 100 Fm 101 Md 102 No 103 Lr

Weak correlations

- SO splittings are very small compared to bandwidth (s and p electrons)

GaAs

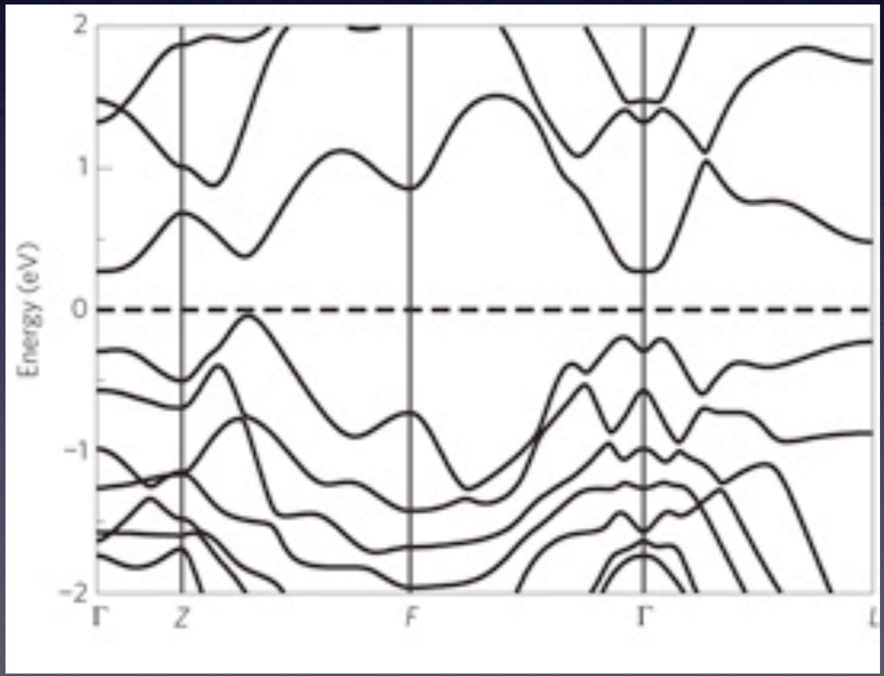


0.34eV

Weak correlations

- In some rare materials, SOC is comparable to bandwidth

Bi_2Se_3



λ

Topological Insulators

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

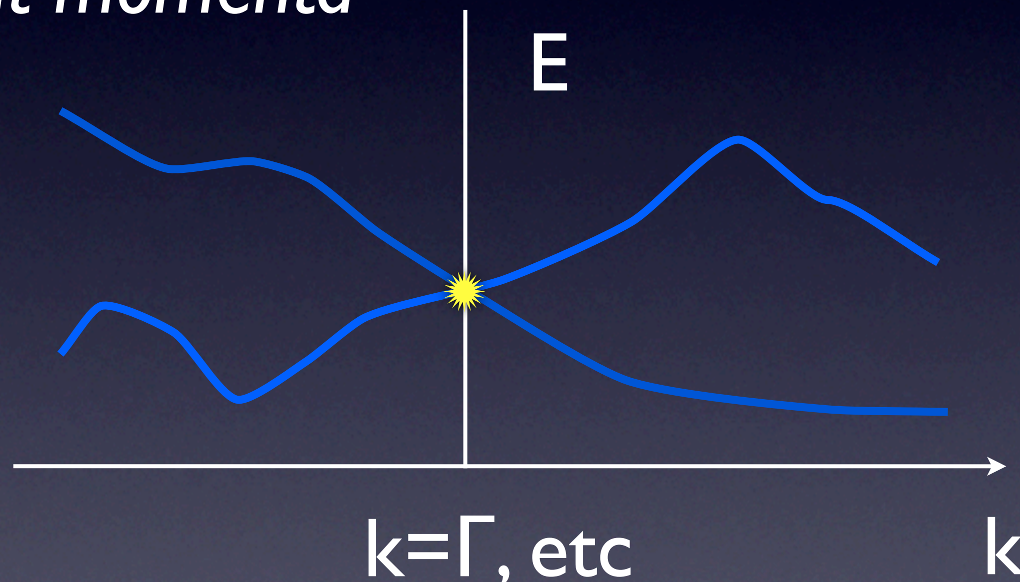
- Band insulators w/ significant SOI can have a topological structure, similar to the integer quantum Hall effect
- TBIs have *protected* surface states that are like *chiral Dirac fermions* and cannot be localized by disorder

Bands with SOC

- Consider for simplicity a solid with Inversion symmetry
  - I: $E_{s,-k} = E_{s,k}$
  - TR: $E_{-s,-k} = E_{s,k}$
- Together, this implies $E_{-s,k}=E_{s,k}$, i.e. all bands are 2-fold degenerate

Bands with SOC

- Pairs of levels can approach one another at *TR-invariant momenta*



- Such a point is a 3(+1)d Dirac point

3+1d Dirac Point

- Since there are 4 levels that approach one another, this is described by a 4×4 Bloch Hamiltonian
- Can be parametrized by the Dirac Matrices
  - Here Γ_1, Γ_2, Γ_3, Γ_4 are odd under TR + I
  - and Γ_5 is even under TR and I
  - other matrices $\Gamma_{a,b}$ are odd under TR*I

3+1d Dirac Point

- Can be parametrized by the Dirac Matrices
  - Here Γ_1, Γ_2, Γ_3, Γ_4 are odd under TR + I
  - and Γ_5 is even under TR and I
  - other matrices $\Gamma_{a,b}$ are odd under TR*I
- Then one can always choose coordinates so

$$H_{\text{Bloch}} = \sum_{a=1}^{3} v_a k_a \Gamma_a + m\Gamma_5$$

3+1d Dirac Point

- Then the Dirac point closes when m=0 only

$$E_{\text{Bloch}} = \pm\sqrt{\sum_{a=1}^{3}(v_a k_a)^2 + m^2}$$

- This describes a *quantum critical point* between two types of band insulators, e.g.
  - m>0 ordinary band insulator
  - m<0 topological band insulator

Surface states

- Solve Dirac equation for an interface

$$\left(k_1\Gamma_1 + k_2\Gamma_2 - i\Gamma_3\partial_z + m(z)\Gamma_5\right)\Psi = E\Psi$$

- This equation has a 2d *chiral bound state*

$$\Gamma_{3,5} = i\Gamma_3\Gamma_5 = \mathrm{sign}[m(\infty) - m(-\infty)]$$

- With a 2d chiral Dirac wavefunction

$$(k_x\Gamma_1 + k_y\Gamma_2)\psi = E\psi$$

$$E = \pm v\sqrt{k_x^2 + k_y^2}$$

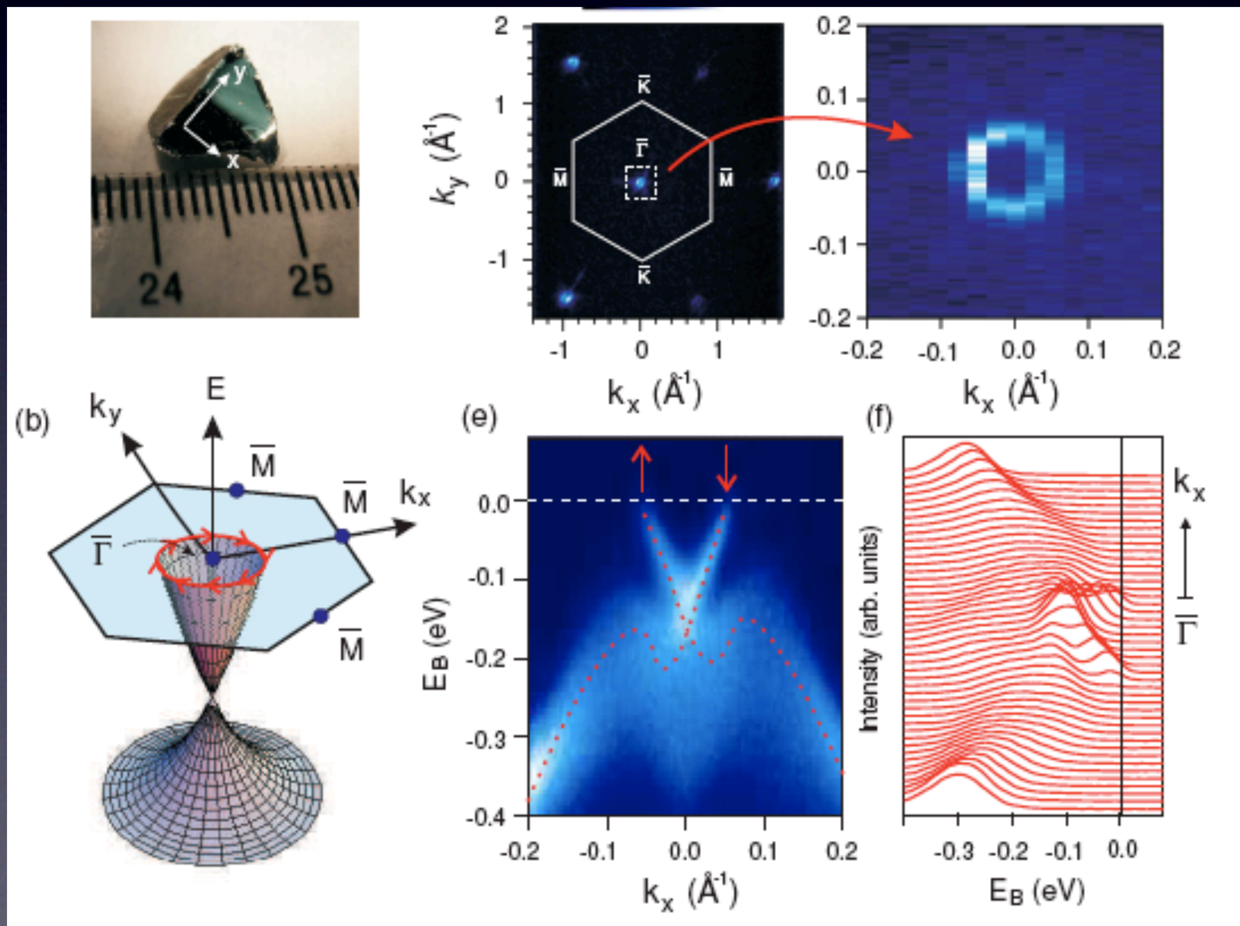
Surface states

- The chiral Dirac state = "1/4 graphene"
- It "violates" the Nielsen-Ninomiya theorem
  - prohibits an odd number of Dirac cones in a 2d lattice model
  - this is only possible because it is the *edge* of a bulk state

Example: Bi_2Te_3

- M.Z. Hasan group - ARPES studies

backscattering prohibited: no localization



More...

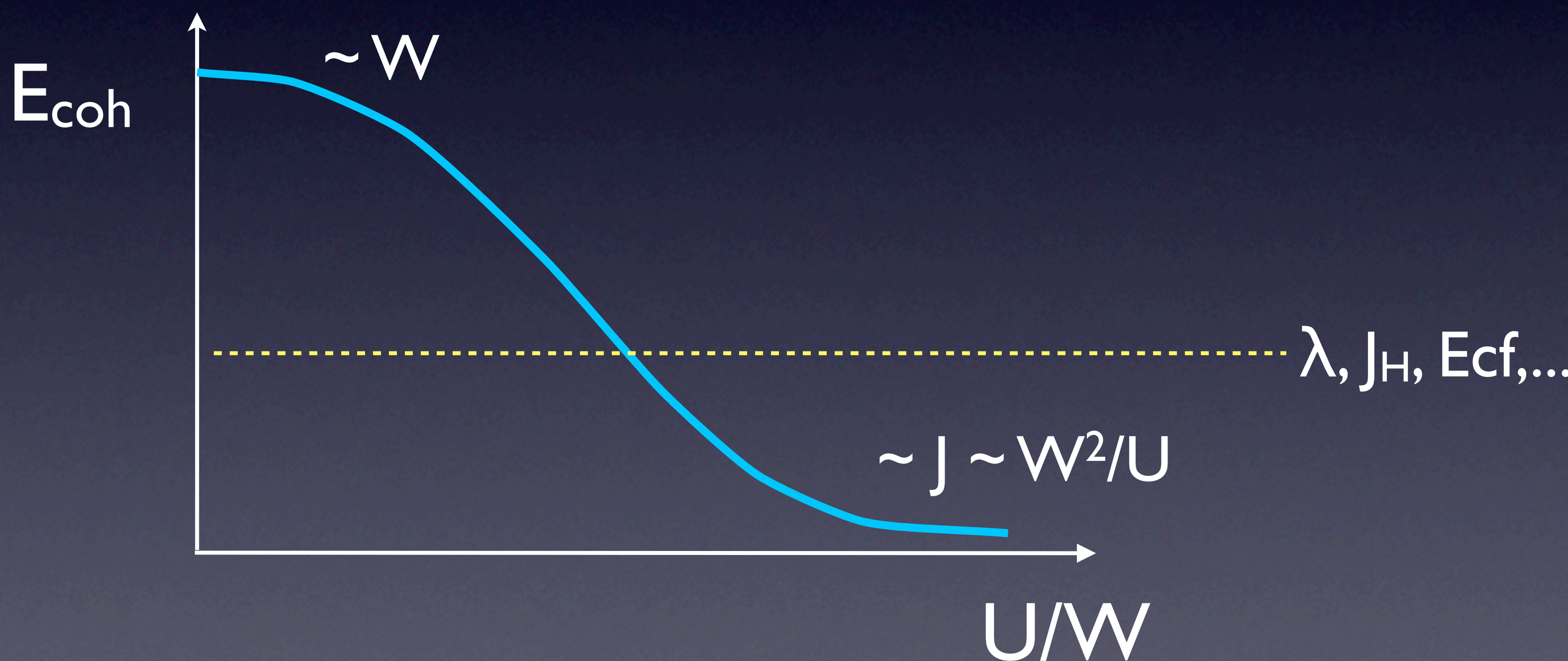
- Topological insulators are also predicted to
  - have strong "quantized" magnetoelectric response
  - show zero modes at certain crystal defects
  - be a platform for novel hybrid structures (e.g. with superconductors or ferromagnets)

Correlations and SOC

- Key observation:
  - Hopping/hybridization is suppressed by Mott physics
  - This allows SOC to compete more effectively

Coherence scale

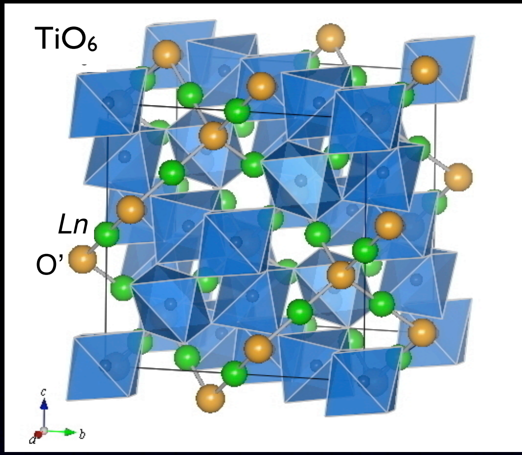
- Local physics is enhanced by Hubbard U



Strong Mott insulators

- U >> W
- In this case, need to compare SOC to J, E_{cf}
- Look for situations with
  - large λ or small J
  - exact or approximate orbital degeneracy

Rare earths



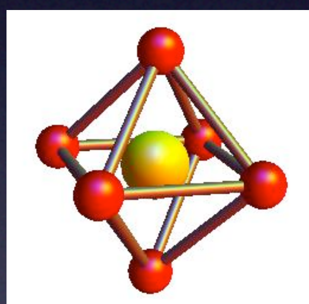
- Generally, 4f electrons are tightly bound and well-shielded from crystal fields
  - leads to large J = L + S local moments
  - usually (but not always) classical magnetism, with strong magnetic anisotropy
- Example: spin ice $Ho_2Ti_2O_7$, $Dy_2Ti_2O_7$
- Most heavy fermions involve such states

Transition Metals

- Typically, crystal fields and exchange are much larger
- In 3d and 4d TMs, strong SO situations are relatively rare

Cubic systems

- in ideal octahedron with cubic symmetry, the d levels split into e_g and t_{2g} multiplets

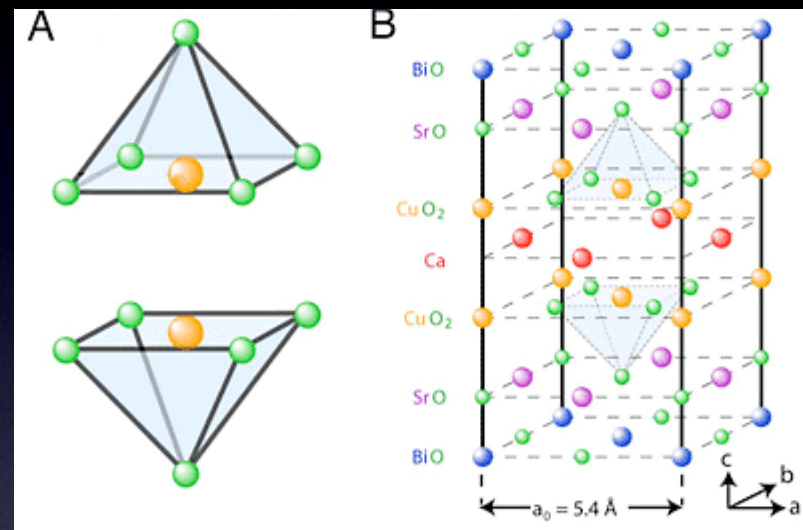


e_g	— —	x^2-y^2, $2z^2-x^2-y^2$
t_{2g}	— — —	yz, xz, xy

- Need to consider SOC within these multiplets, which are often further split by non-cubic distortions

Cuprates

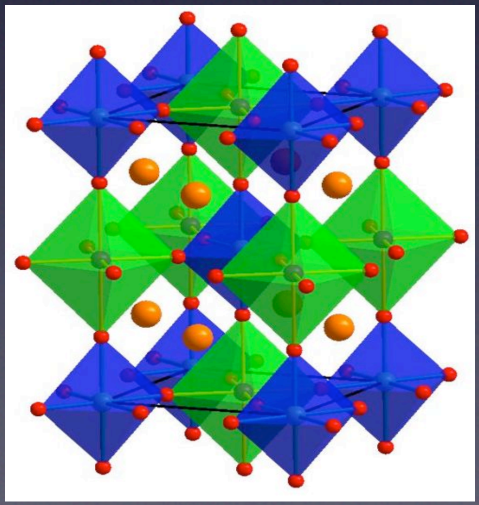
- Octahedra in cuprates have a large elongation along z axis
- completely removes orbital degeneracy



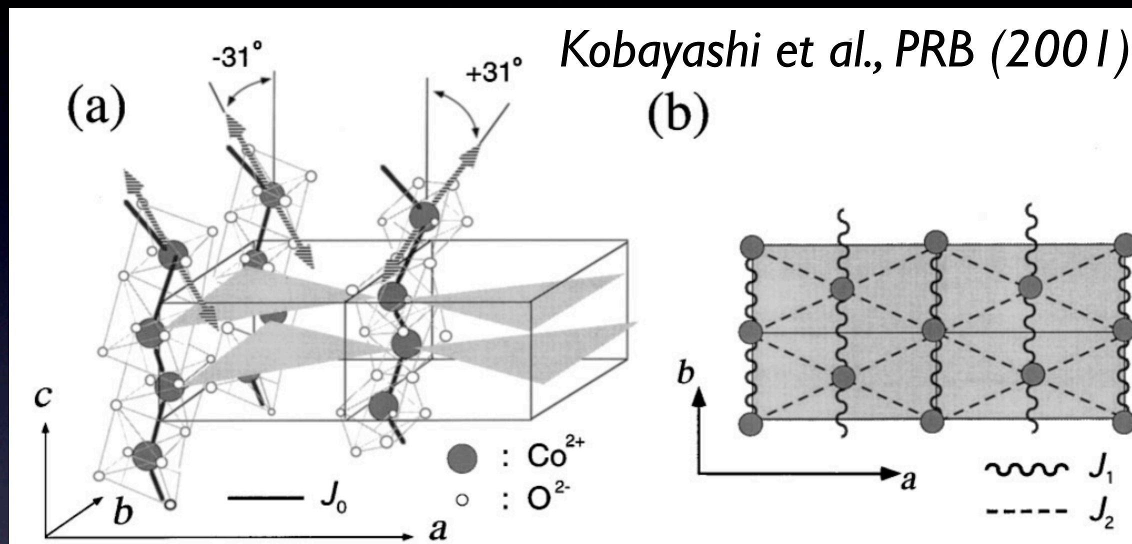
$Cu^{2+}=3d^9$

Double Perovskites

- $A_2BB'O_6$ often form with 4d and 5d TMs
- For instance, Ba_2NaOsO_6, Ba_2LiOsO_6
- Large separation of B' ions minimizes exchange
  - $J \sim 5meV$
  - $\lambda \sim 0.2eV$

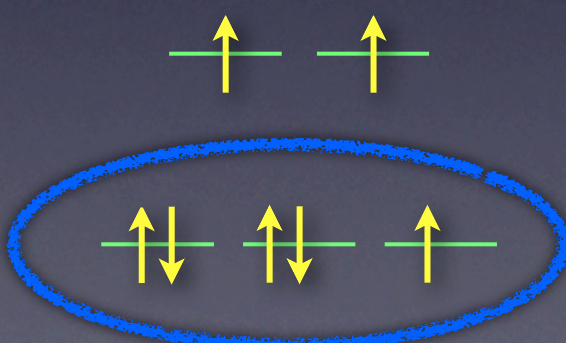


$CoNb_2O_6$



Kobayashi et al., PRB (2001)

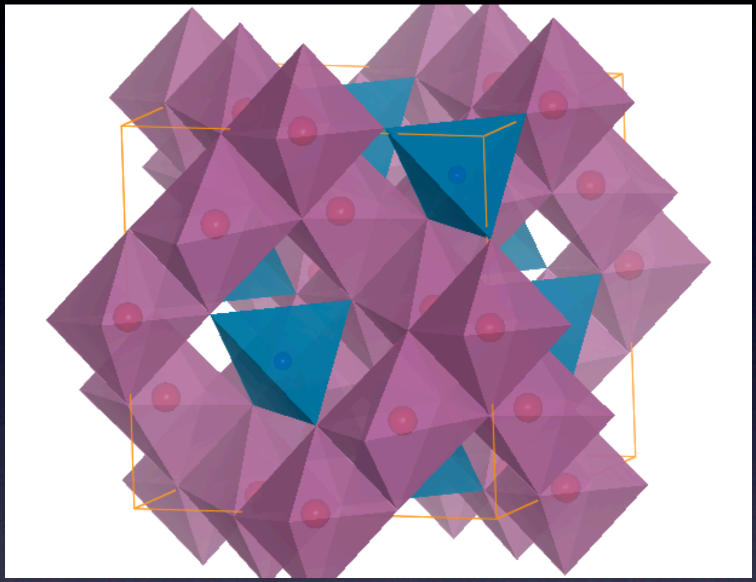
- Co^{2+} in *high spin* state (J_H)



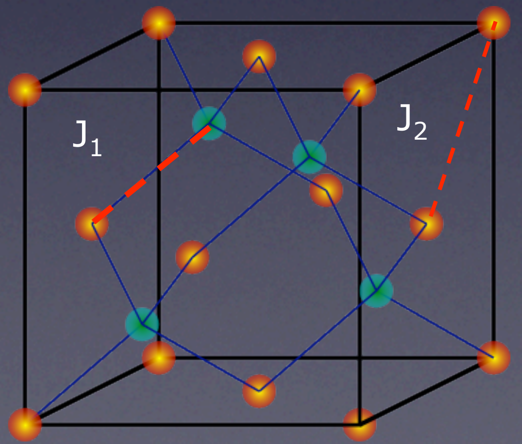
orbital degeneracy

strong Ising anisotropy

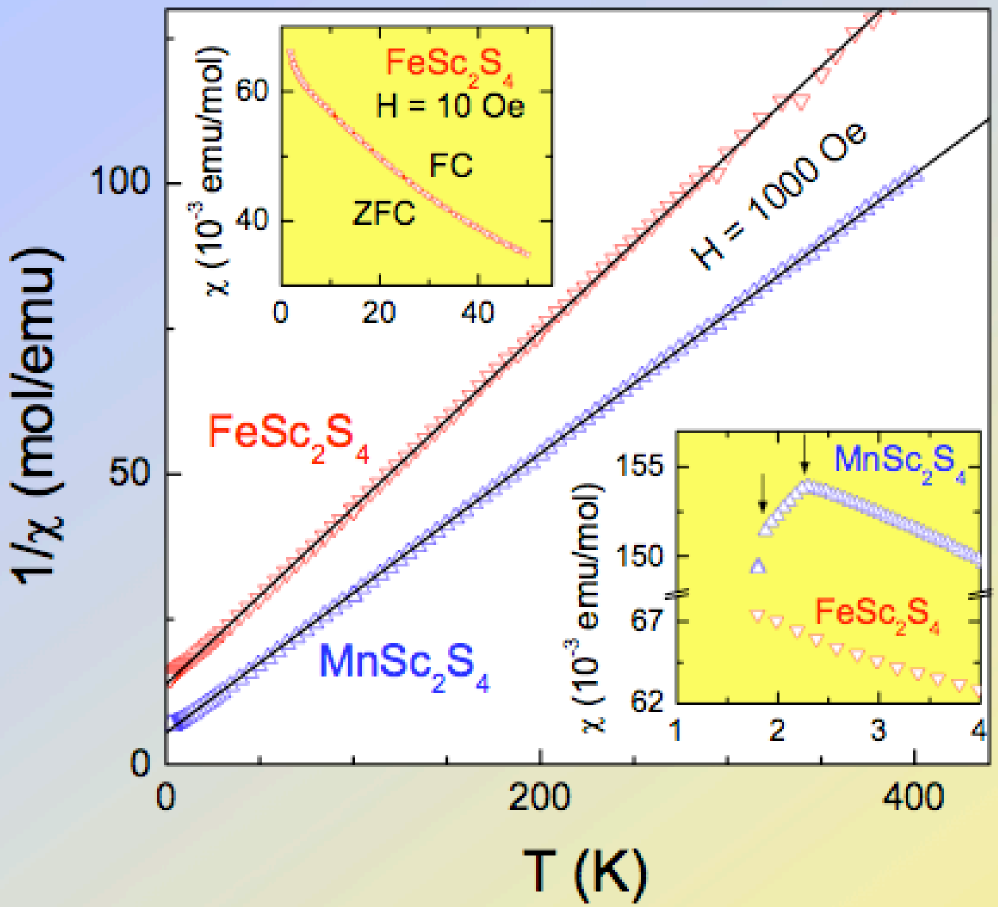
$FeSc_2S_4$



- Fe atoms occupy A sites of the spinel, which are *tetrahedrally* coordinated
- A-sites forming a *diamond lattice*
- Widely separated A sites leads to *weak exchange*
- Here exchange and *SOC* can *compete* in an interesting way



Frustration?



$FeSc_2S_4$: θ_{CW} = 50 K

T > 30 mK:

no long-range magnetic order

no spin-glass

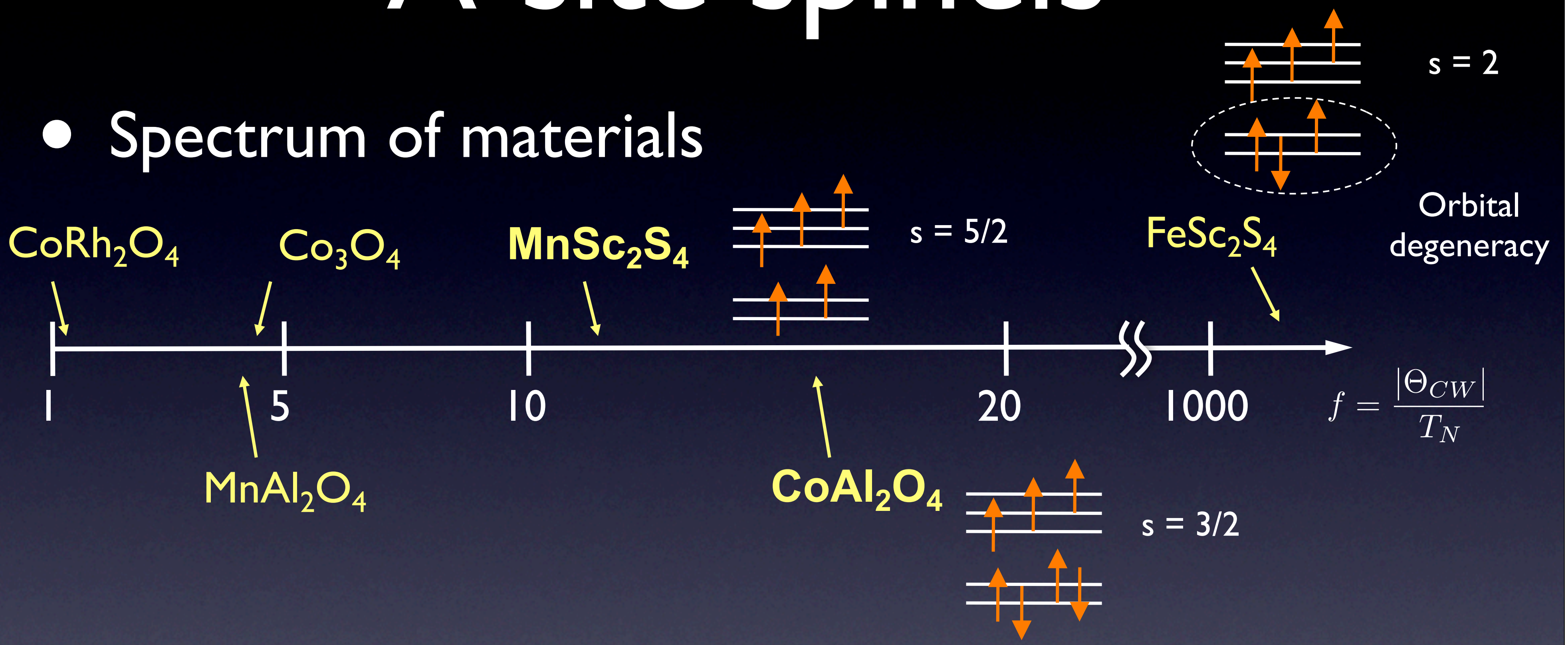
$MnSc_2S_4$: θ_{CW} = 25 K

AFM transition @ 2 K

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

A-site spinels

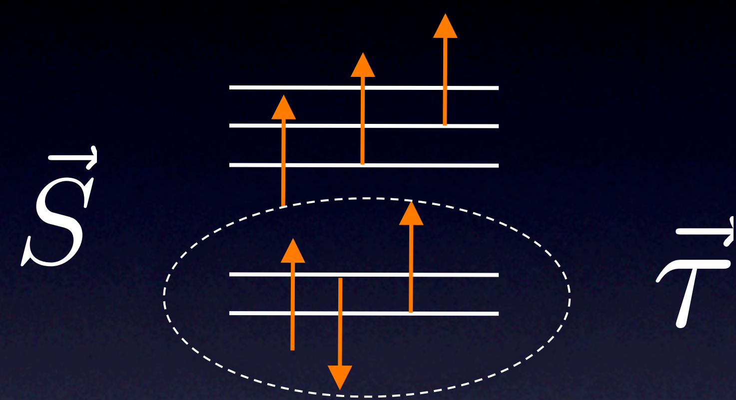
- Spectrum of materials

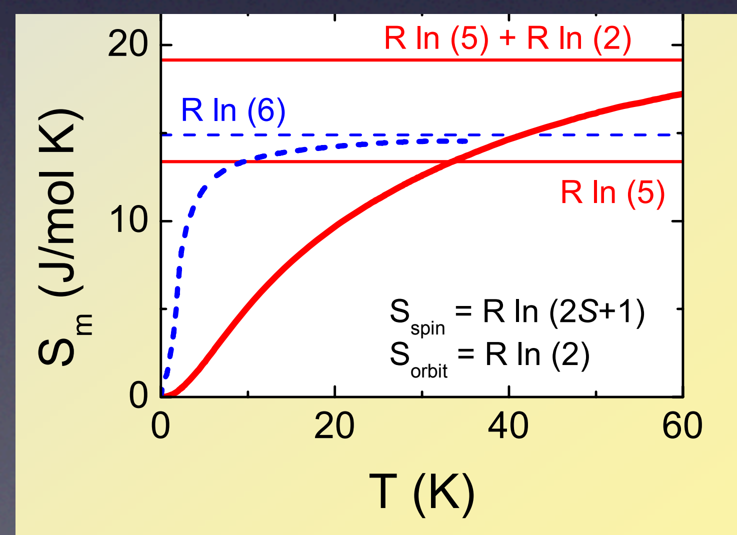


V. Fritsch et al. PRL **92**, 116401 (2004); N. Tristan et al. PRB **72,** 174404 (2005); T. Suzuki *et al.* (2006)

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

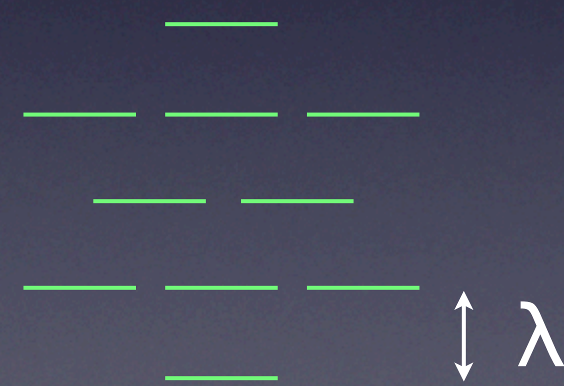
- Separate orbital and spin degeneracy can be split!

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

- Energy spectrum: singlet GS with gap = λ
- Microscopically,

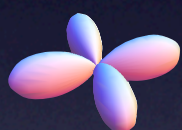
$$\lambda = \frac{6\lambda_0^2}{\Delta}$$

- Naive estimate $\lambda \approx 25\text{K}$



Spin orbital singlet

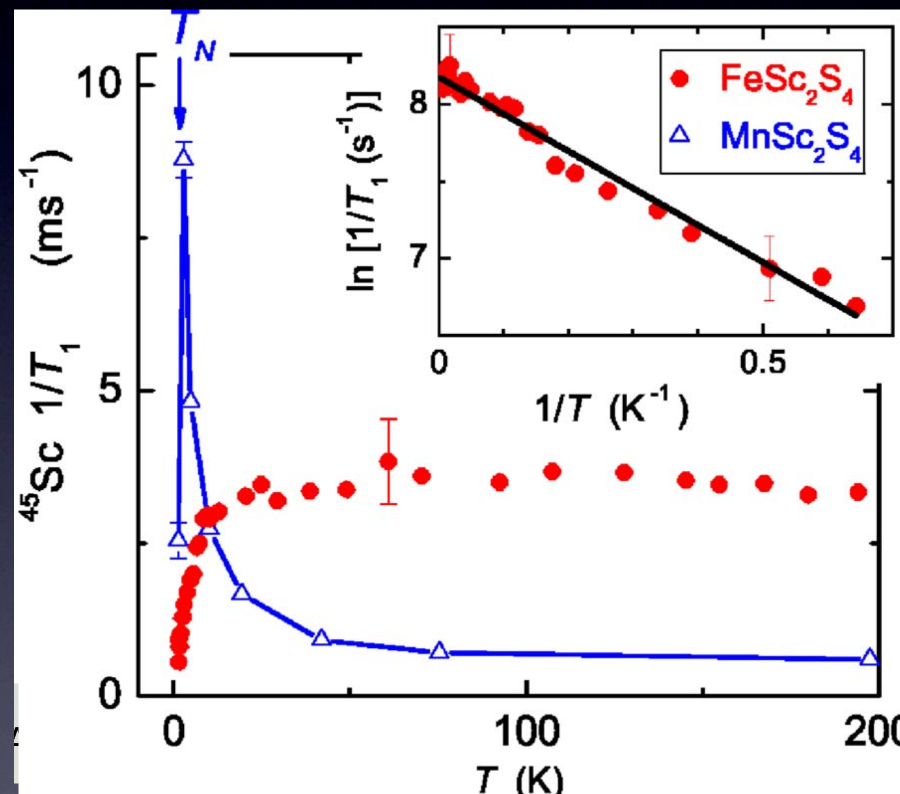
- Ground state of λ>0 term:

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

- 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



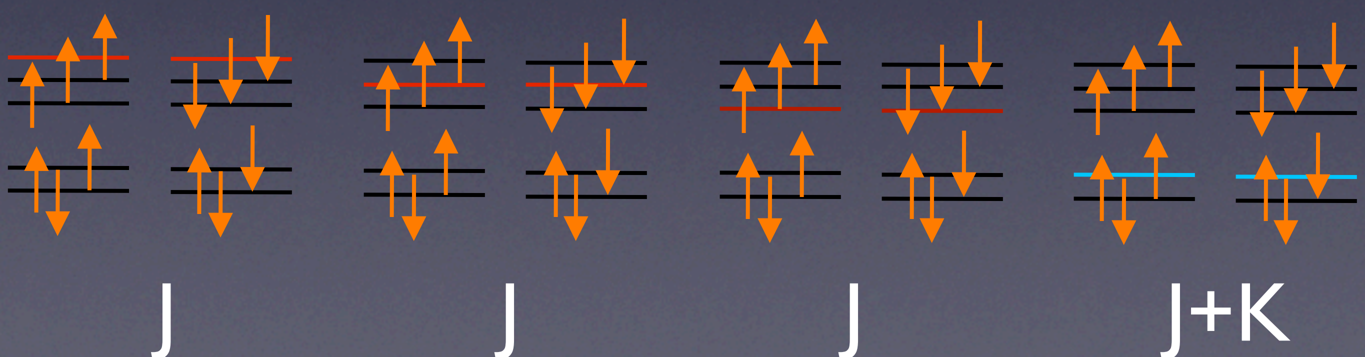
N. Büttgen *et al, PRB* ***73****, 132409 (2006)*

Exchange

- *Largest* interaction is just Heisenberg exchange

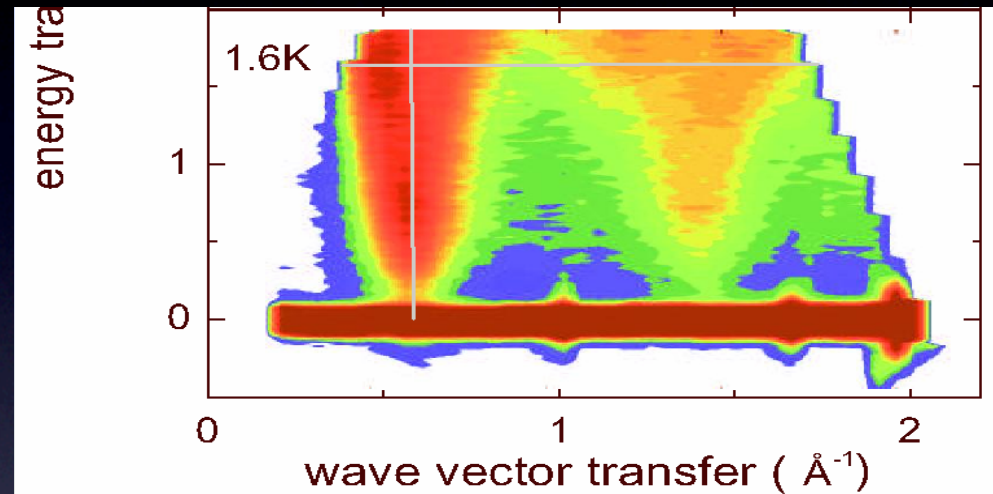
$$H_{ex} \approx \frac{1}{2}\sum_{ij} J_{ij}\mathbf{S}_i \cdot \mathbf{S}_j$$

- More exchange processes contribute

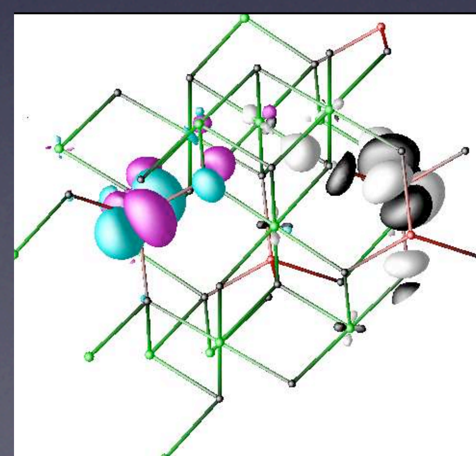


Minimal Model

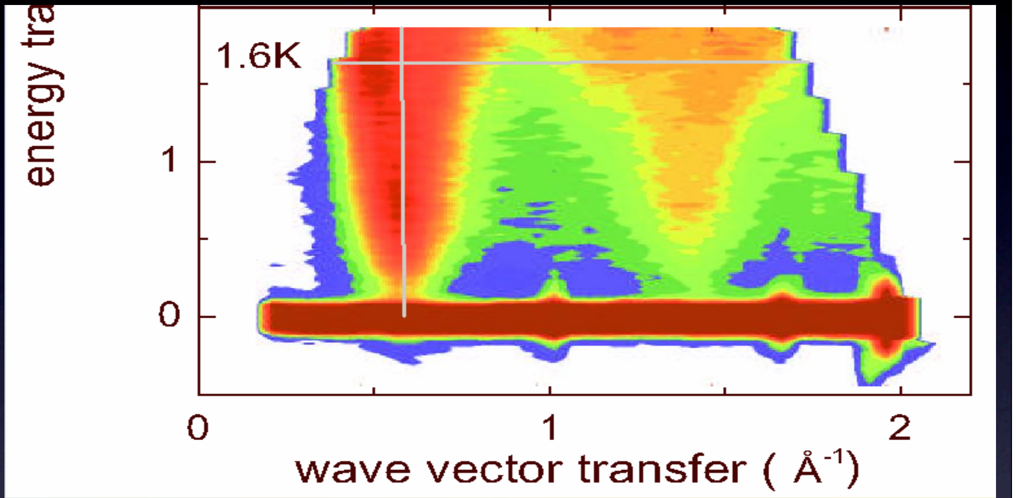
- Neutron scattering suggests peak close to $2\pi(100)$
- Indicates $J_2 >> J_1$
- Recent LDA calculations confirm this microscopically (S. Sarkar *et al*, 2010)



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



Minimal Model



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

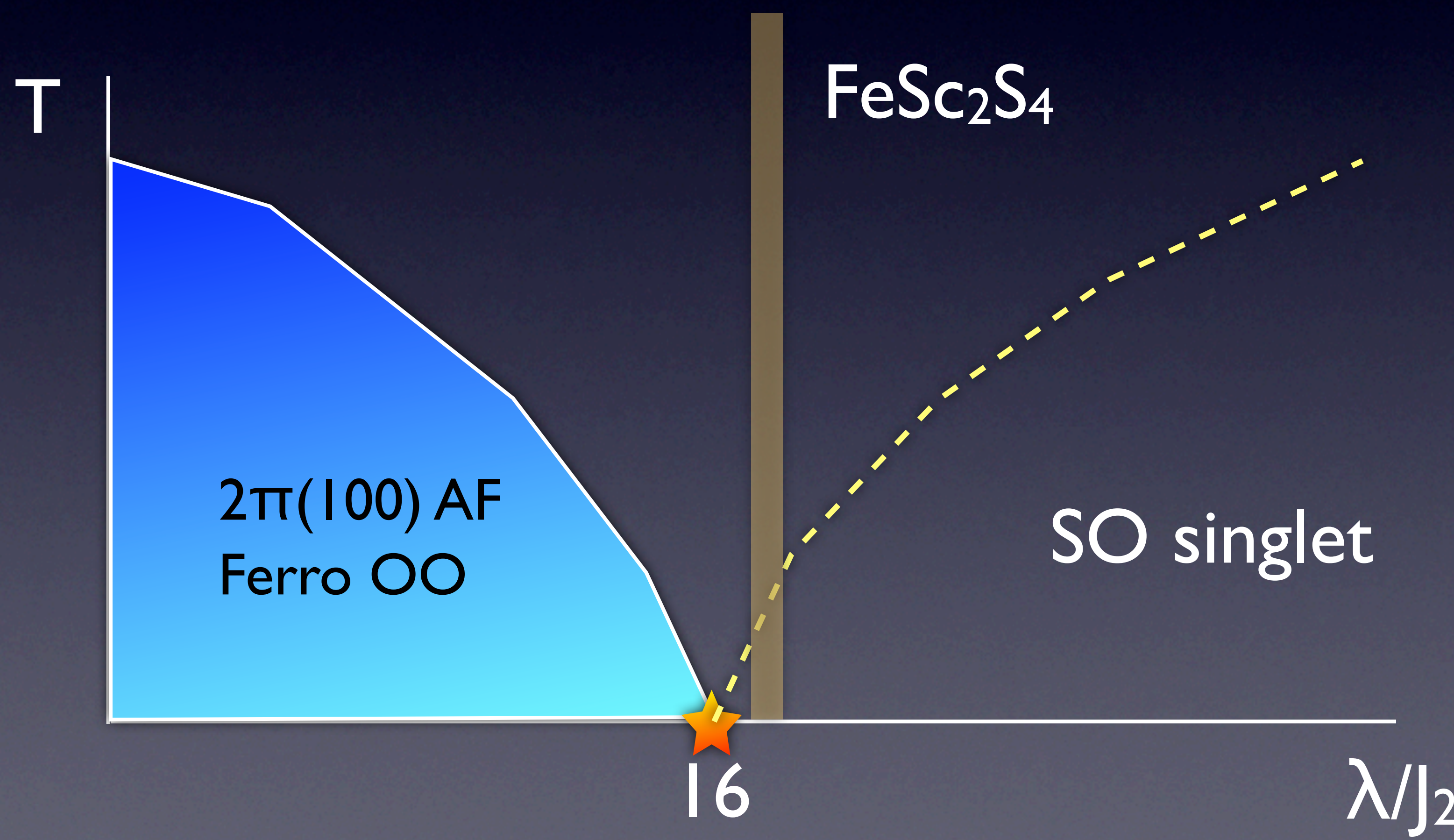
- Neutron scattering suggests peak close to 2π(100)
- Indicates $J_2 >> J_1$

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

Expect MFT good in 3+1 dimensions

Quantum Critical Point

- Mean field phase diagram



Predictions

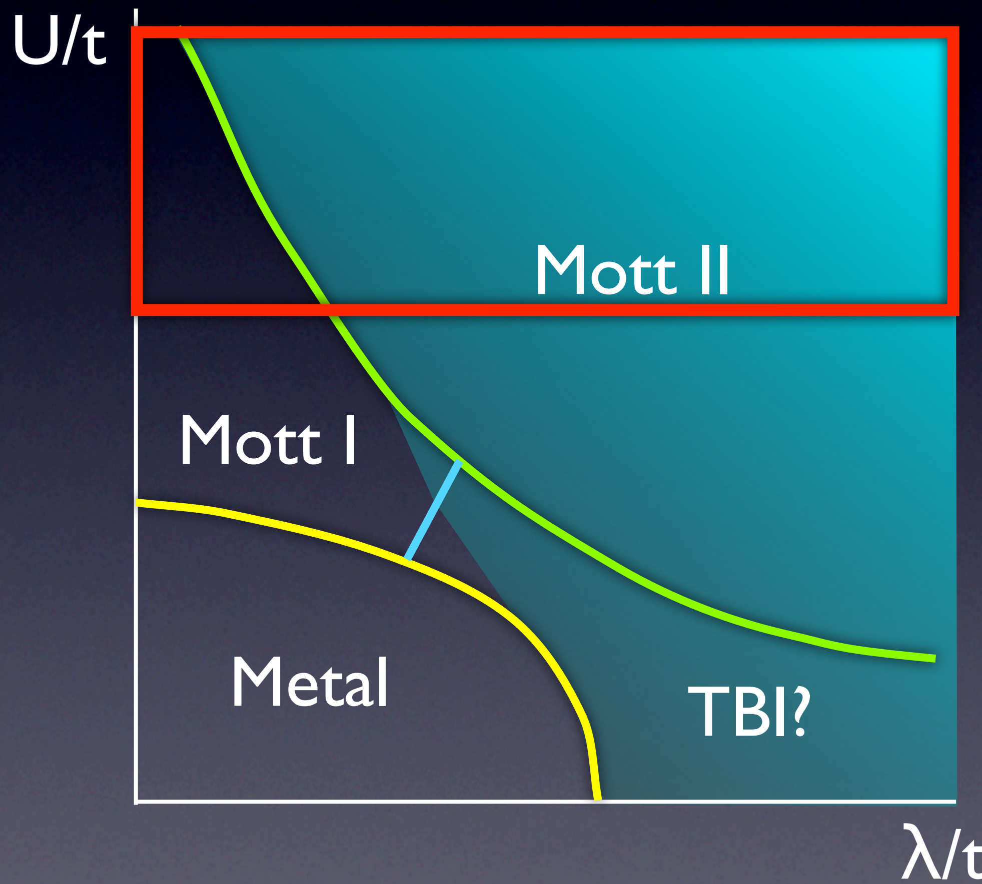
- Large T=0 susceptibility (estimated) ✓
- Scaling form for $(T_1 T)^{-1} \sim f(\Delta/T)$ ✓
- Specific heat $C_v \sim T^3 f(\Delta/T)$ ✓
- Possibility of pressure-induced ordering
- Magnetic field *suppresses* order
  - opposite to simple "dimer" antiferromagnet

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 regime

- As correlations increase, SOC becomes increasingly important
- It can easily be at least comparable to *effective* bandwidth near the transition from metal to insulator
- Here all 3 effects: SOC, U, and W are comparable!



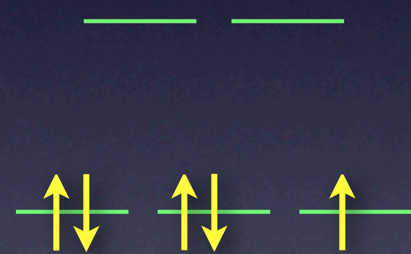
schematic phase diagram

5d Transition Metals

- 5d transition metal oxides are especially interesting
  - $\lambda \sim 0.5$ eV
  - U ~ 1-2eV (5d orbitals rather extended)
  - W ~ 1-4eV
- Together, SOC and U can conspire to produce Mott insulators

Iridates

- Ir is particularly interesting
- Most common valence Ir^{4+} is $S=1/2$ in octahedral coordination



$5d^5$

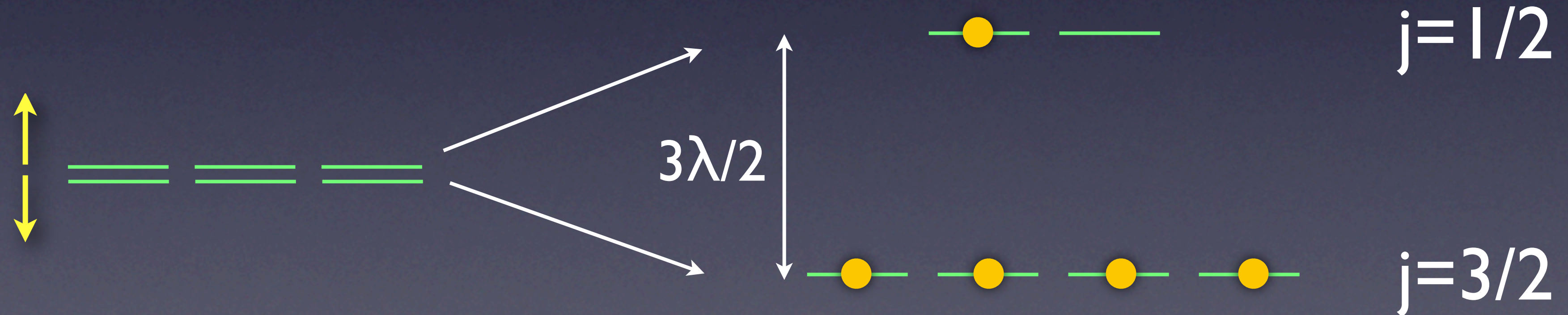
- Interesting interplay of orbital and spin degeneracy

SOC for Ir^{4+}

- Orbital angular momentum in the t_{2g} manifold behaves like for p states

$$P_{t_{2g}} \mathbf{L} P_{t_{2g}} = -\mathsf{L}_{\ell=1}$$

- As a consequence get effective j=1/2 state

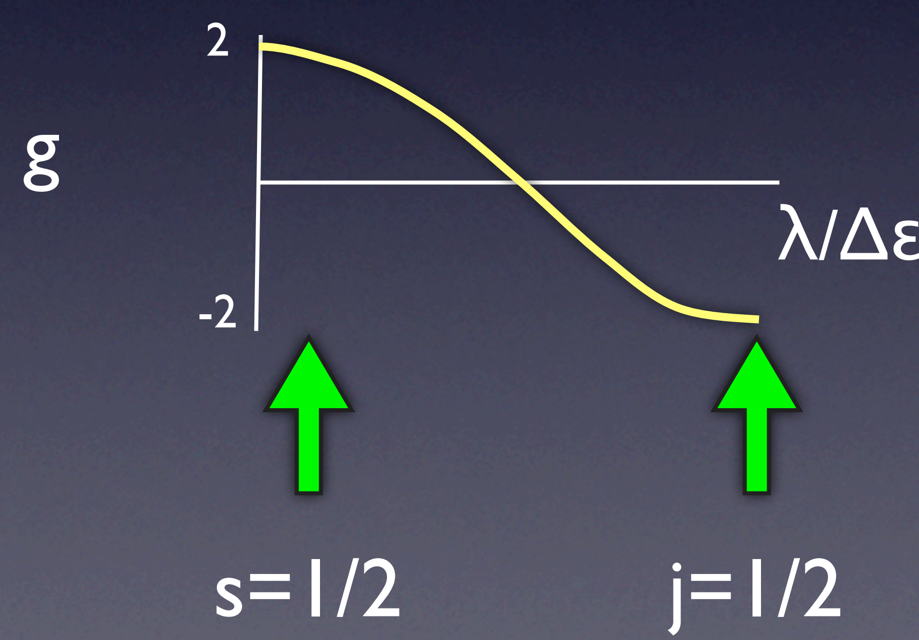


- complex: $|J_z = \frac{1}{2}\rangle = \frac{1}{\sqrt{3}} \left[|\mathrm{xy}\rangle|\uparrow\rangle + |\mathrm{xz}\rangle|\downarrow\rangle + i|\mathrm{yz}\rangle|\downarrow\rangle \right]$

SOC versus H_{cf}

$$H = H_{non-cubic} + H_{SOI}$$

- Spin orbit and non-cubic crystal fields compete to split orbital degeneracy



Some Iridates

material	structure	behavior
Sr_2IrO_4	single-layer perovskite	AF Mott insulator
Na_2IrO_3	honeycomb lattice	AF insulator
$Na_4Ir_3O_8$	hyperkagome lattice	spin-liquid Mott insulator
Ir_2O_4	spinel-based pyrochlore	small gap insulator
$Ln_2Ir_2O_7$	pyrochlore	MITs with magnetic Mott insulator

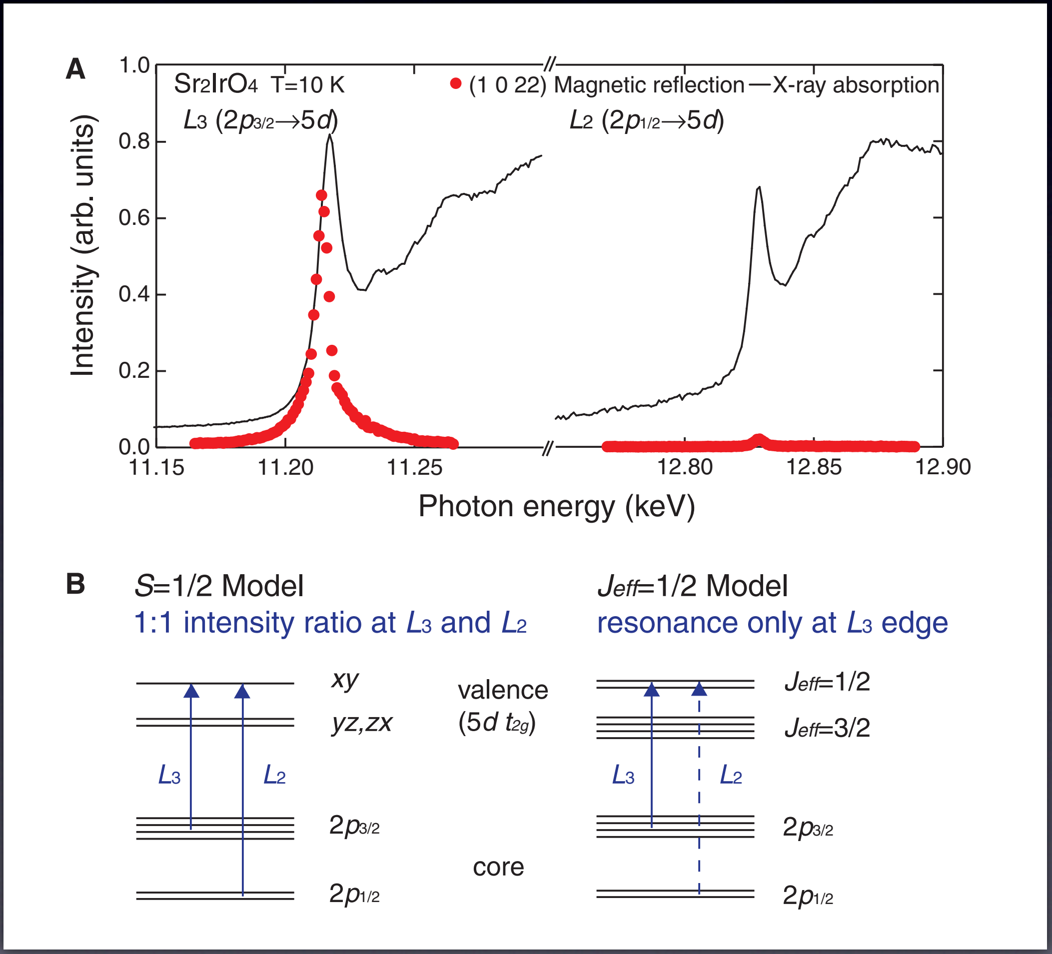
Issues

- J=1/2 or S=1/2 or in between?
- Mott or Slater insulators?
- Are there non-trivial band topologies?
  - can topological insulator physics pertain to Mott insulators?
- How is the Mott transition affected by SOC?

Sr_2IrO_4

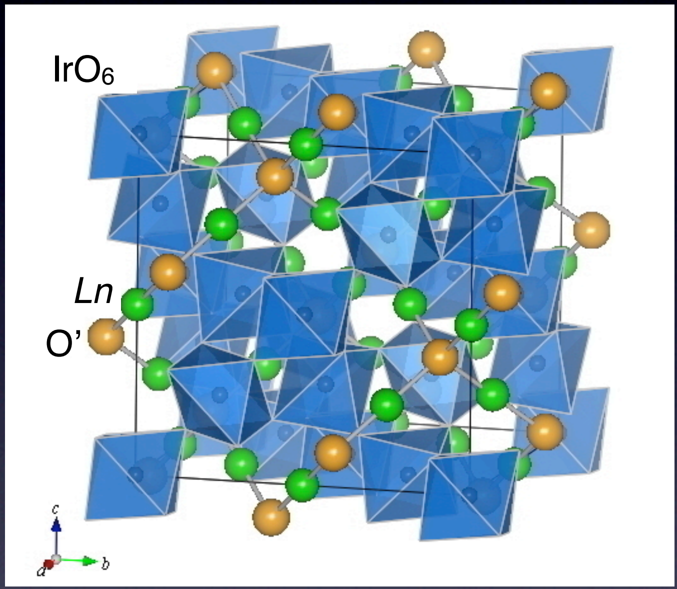
resonant X-ray scattering clearly show J=1/2 state

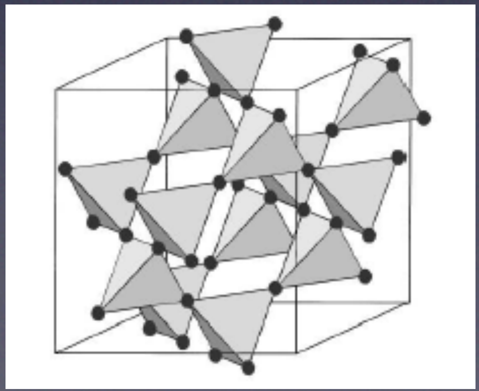
BJ Kim *et al*, Science (2009).



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

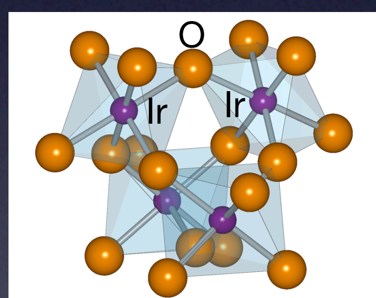


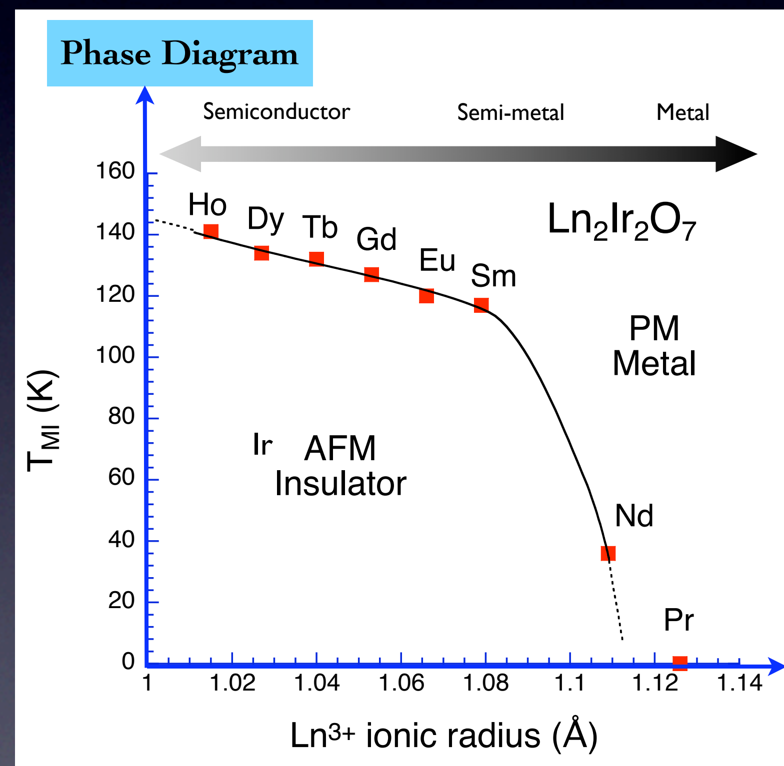


Metal-Insulator Transition

K. Matsuhira et al, 2007

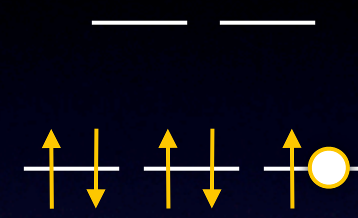
- Decreasing Ir-O-Ir bond angle makes more insulating



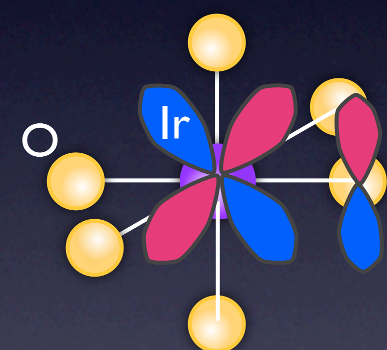


Model

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



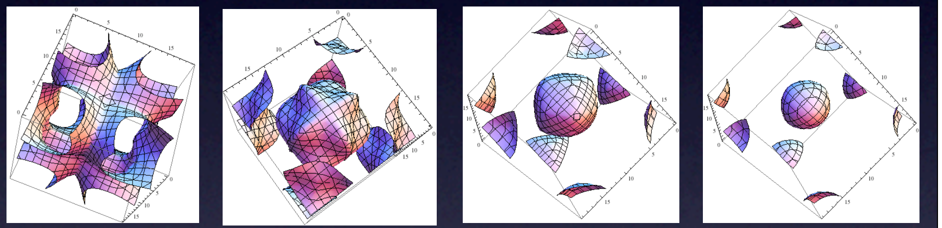
- Ir-O-Ir hopping
  - dominant $V_{pd\pi}$ channel

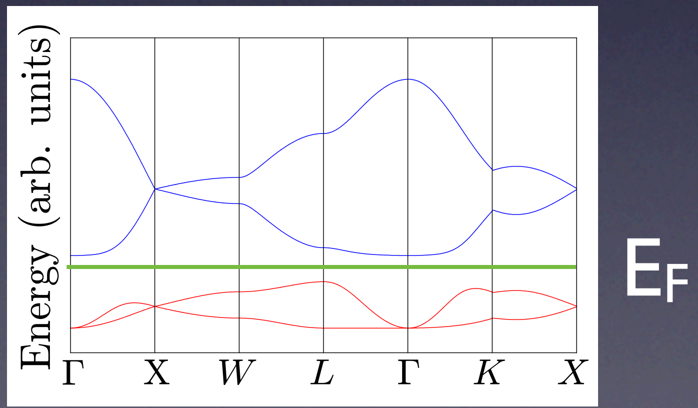


- Spin-orbit coupling
  - $H_{SOI} = -\lambda \vec{L} \cdot \vec{S}$
- Hubbard U

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

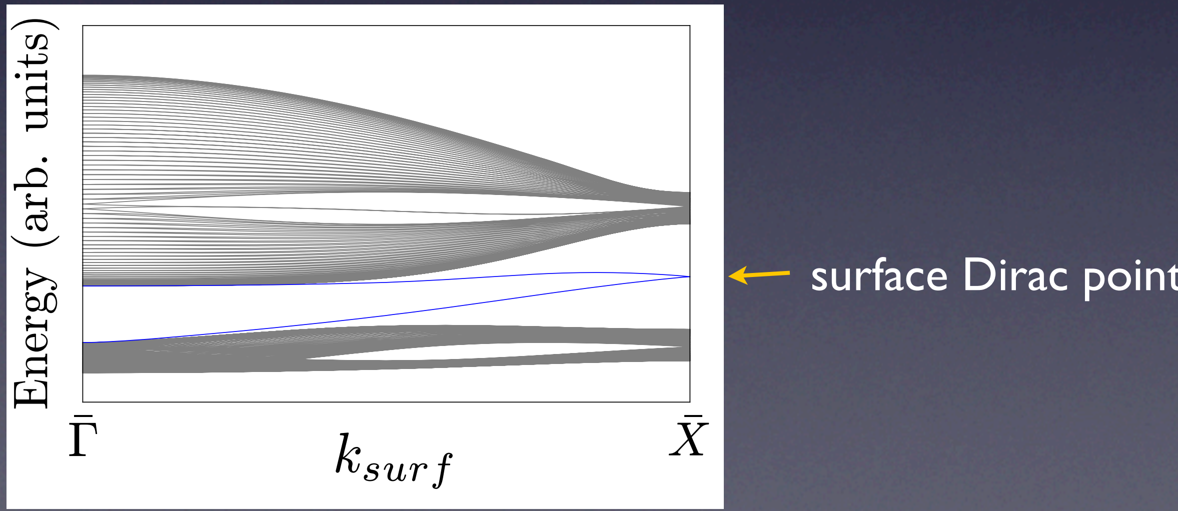




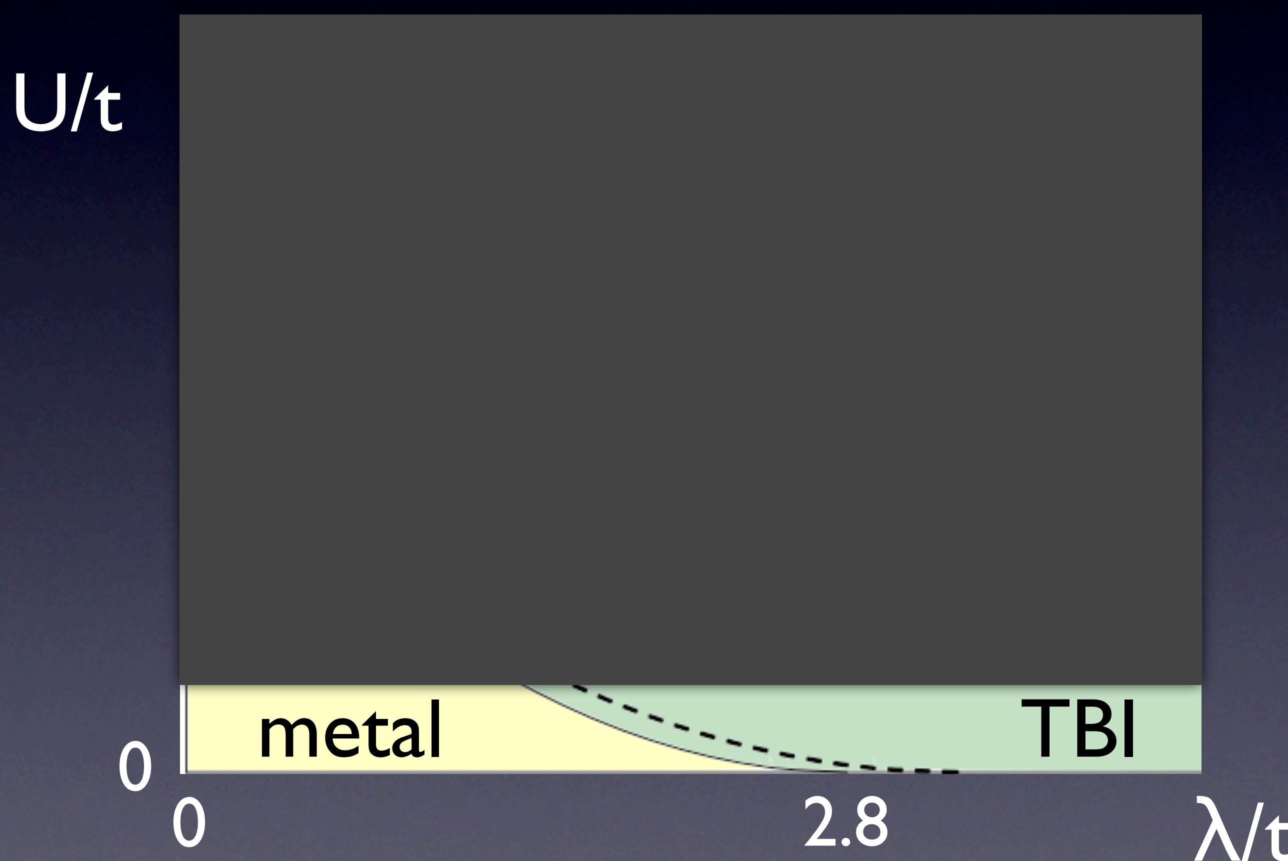
Topological Band Insulator

- We can show using criteria developed by Fu and Kane that this is a "strong" topological band insulator
- Surface states

(100) surface



Phase Diagram



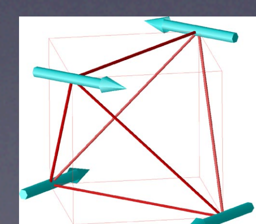
Very large U/t

- Heisenberg "spin" model for j=1/2 eigenstates

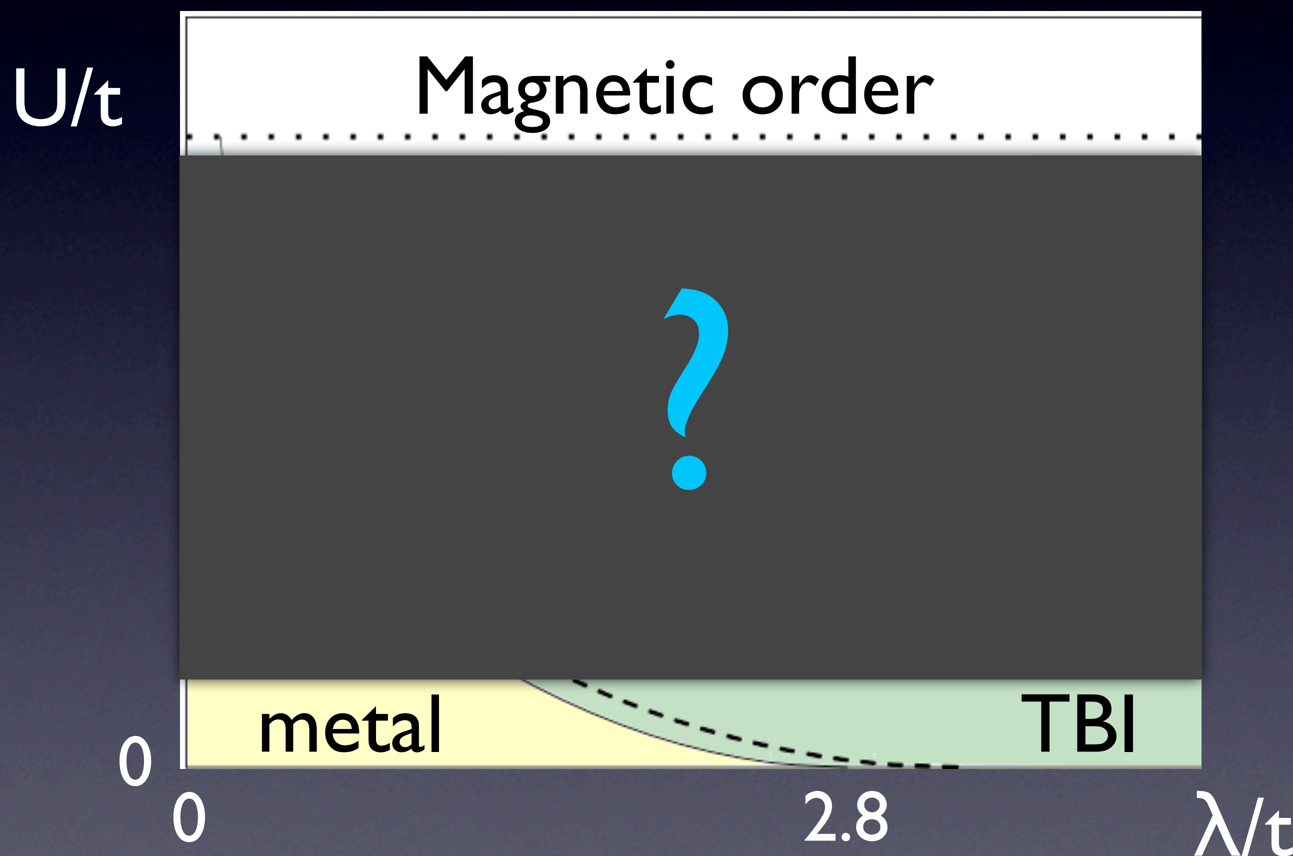
$$H_{spin} = \frac{4t^2}{U} \sum_{ii'} \left[J \vec{S}_i \cdot \vec{S}_{i'} + \vec{D}_{ii'} \cdot \vec{S}_i \times \vec{S}_{i'} + \cancel{\vec{S}_i \cdot \overleftrightarrow{\Gamma}_{ii'} \cdot \vec{S}_{i'}} \right]$$

Elhajal et al, 2005

- This model has been extensively studied
  - 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



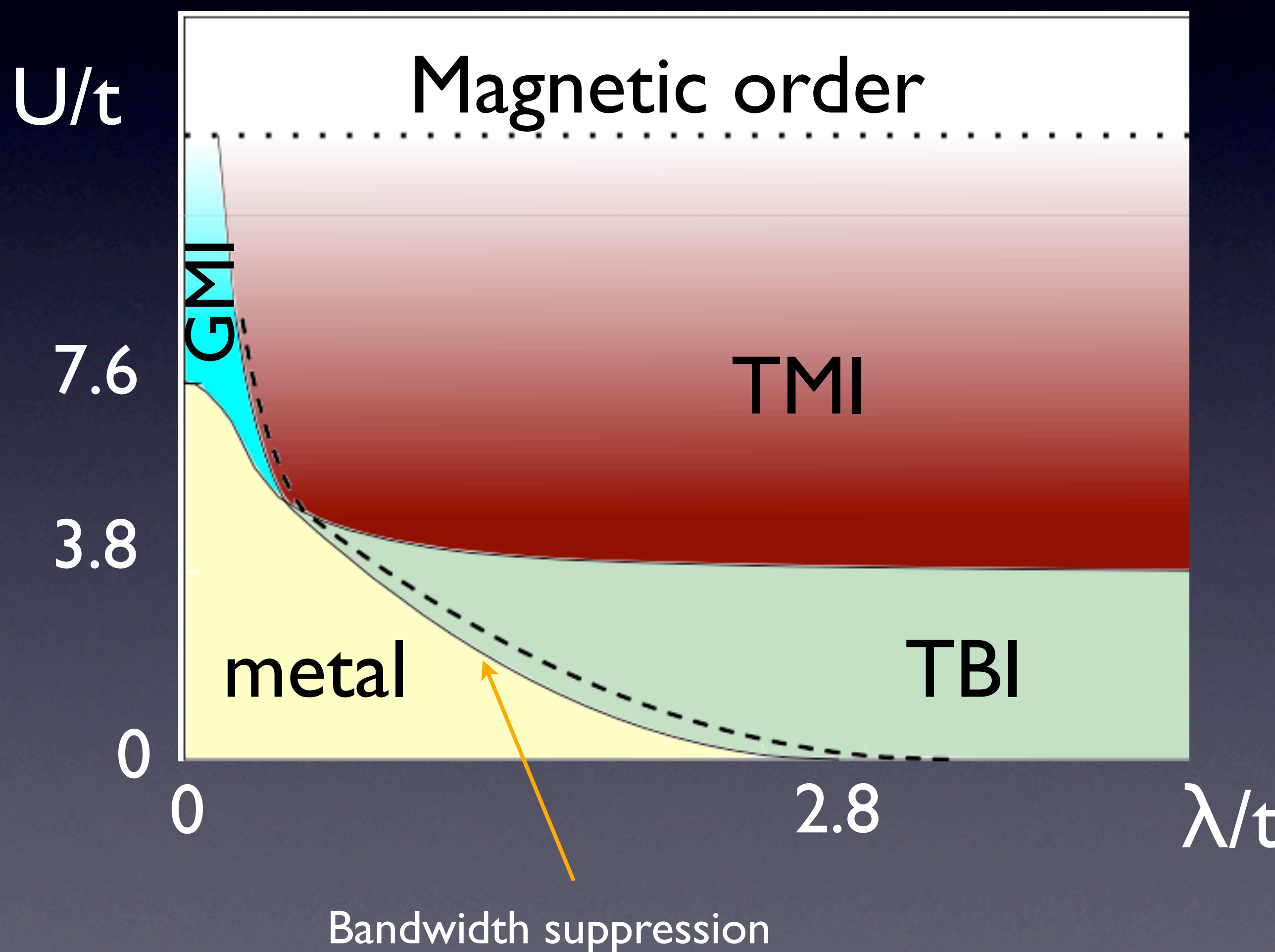
Phase Diagram



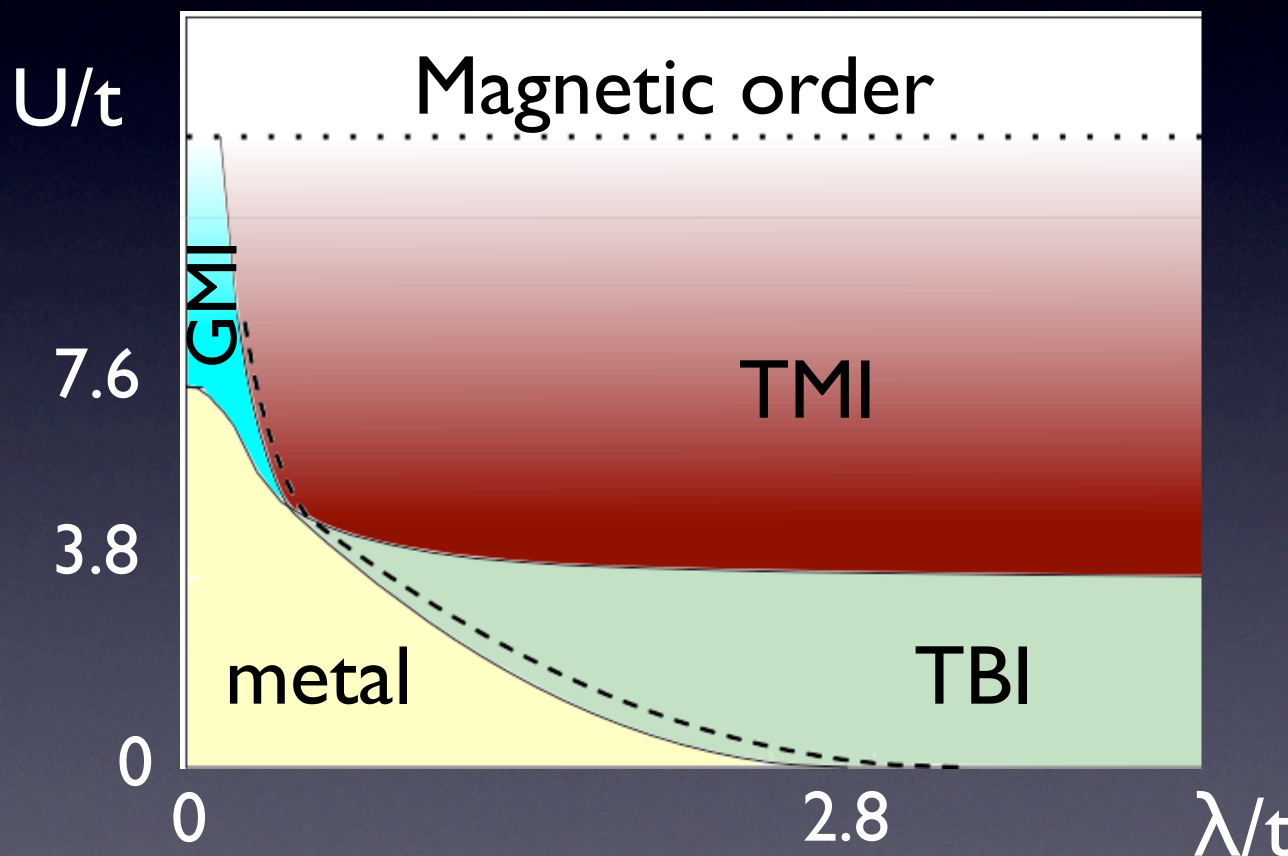
Intermediate U

- 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

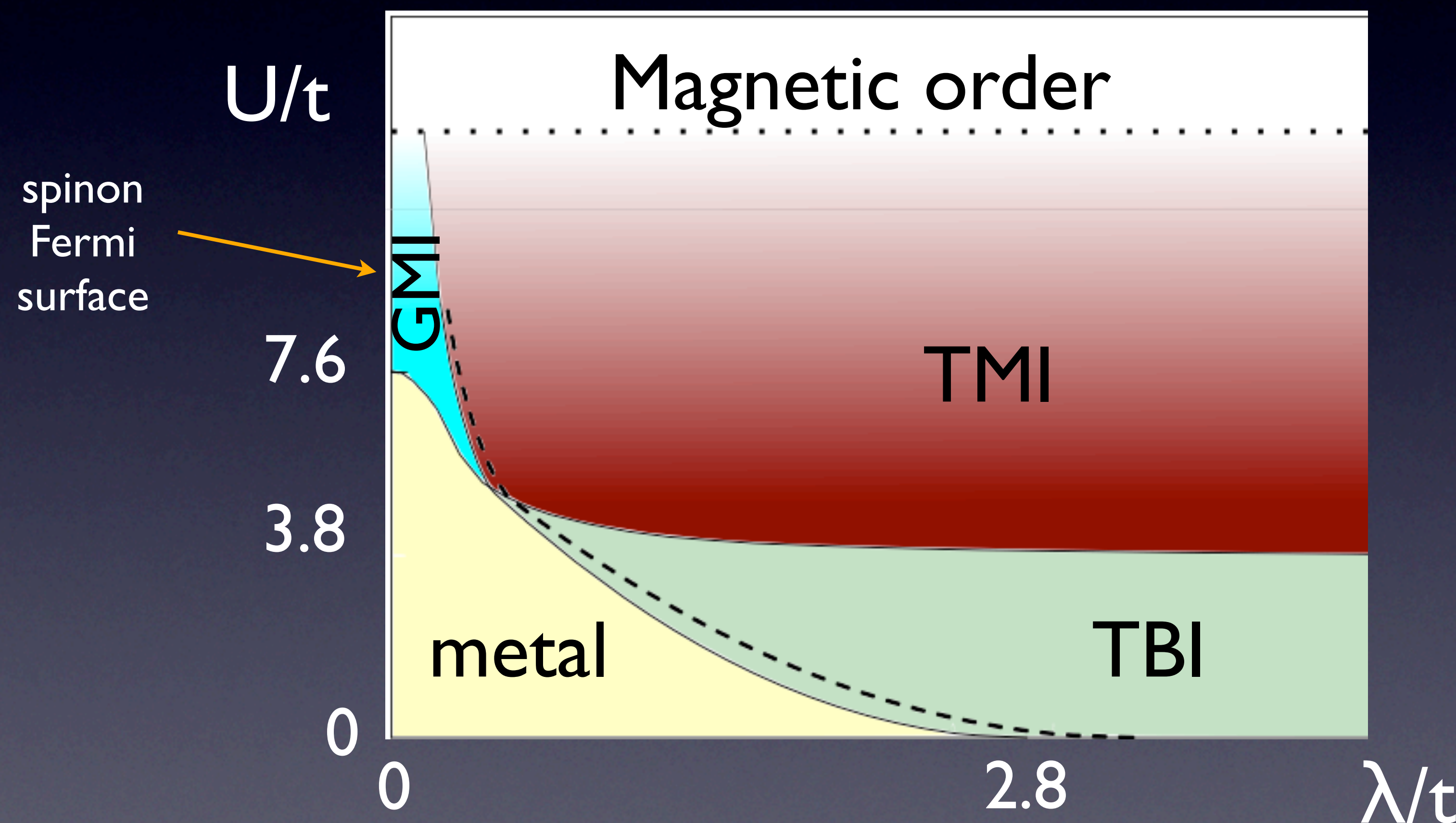


Phase Diagram

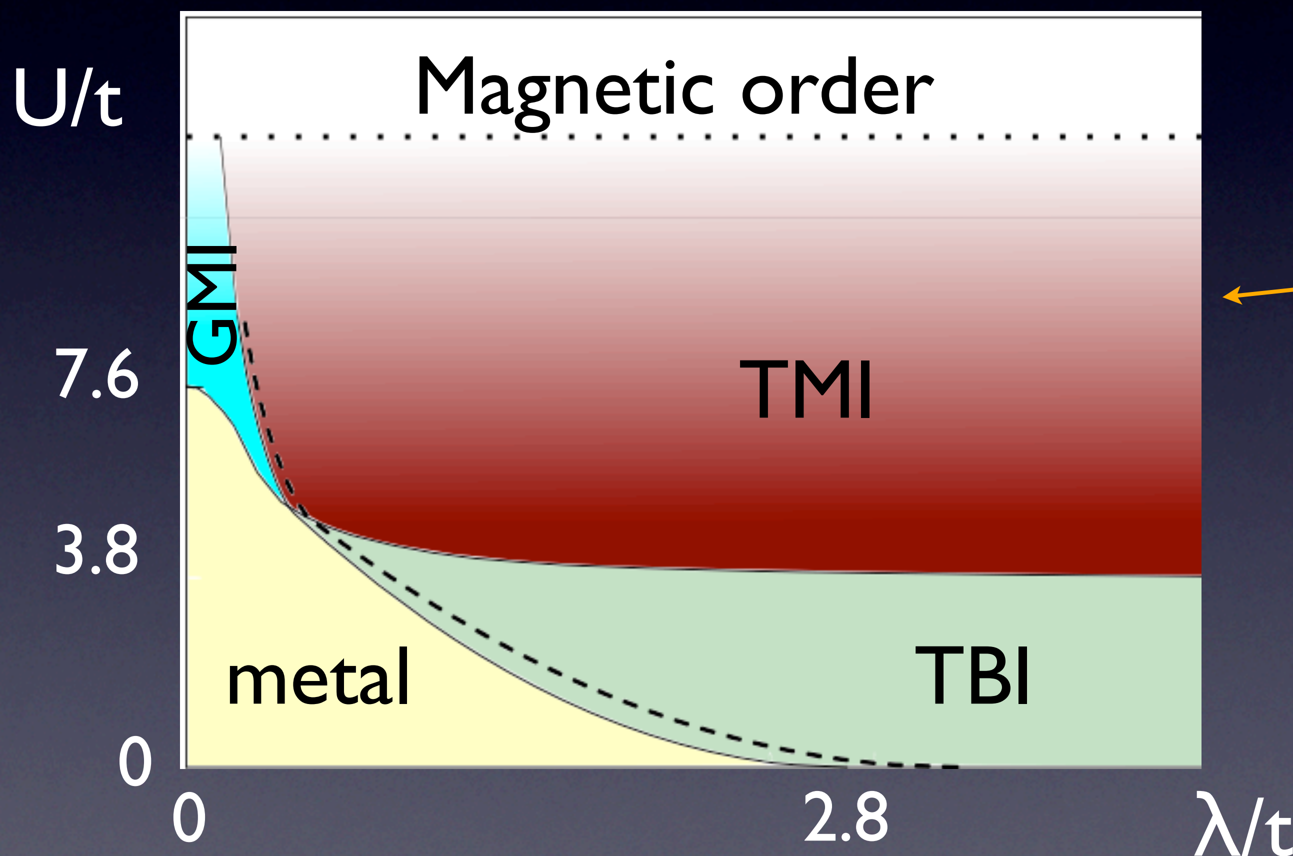


rotors gapped in MIs

Phase Diagram



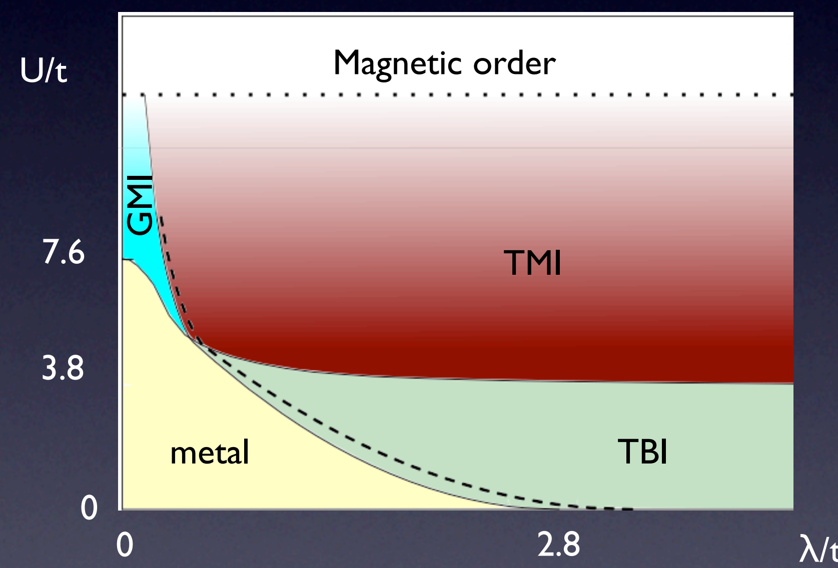
Phase Diagram



spinons in
Z_2 TBI
state

Topological Mott Insulator

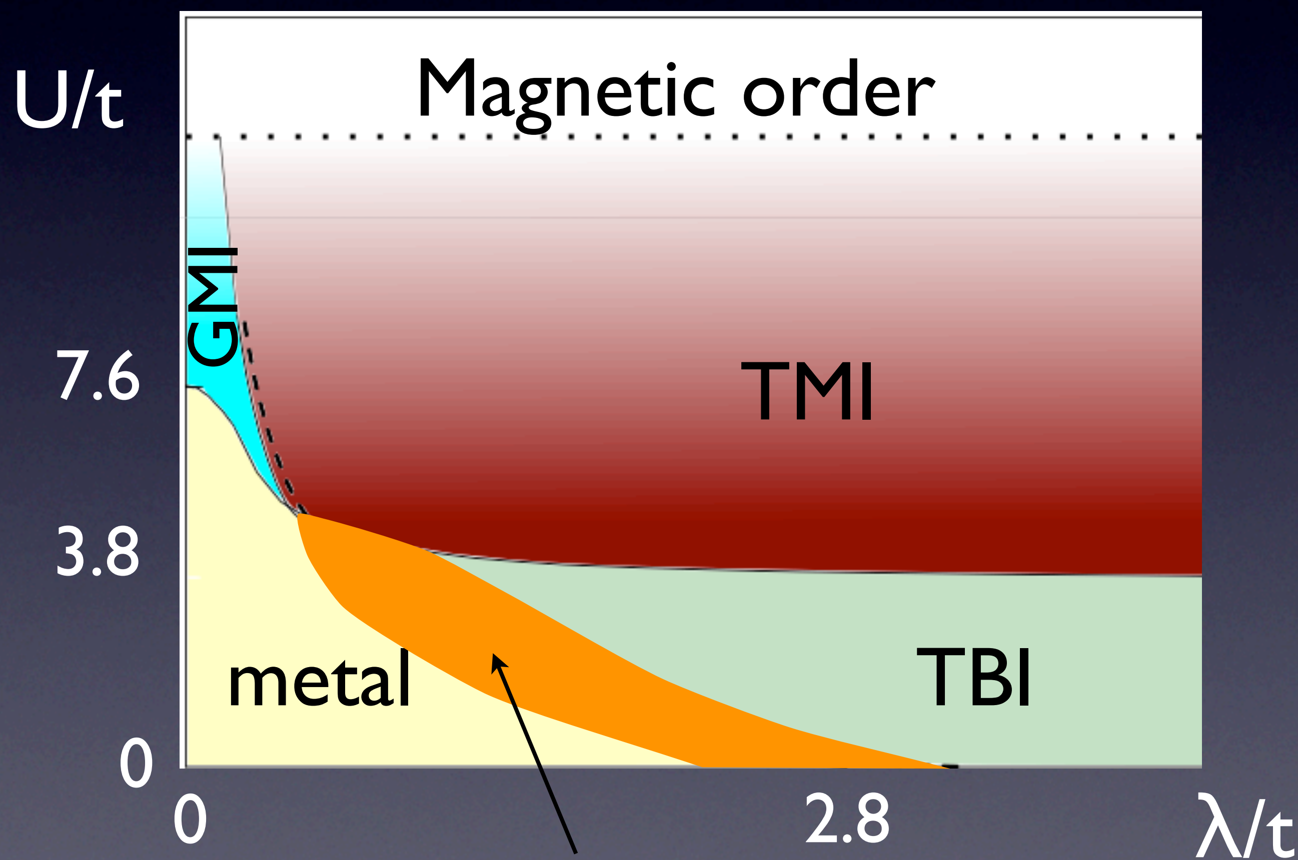
- A U(1) spin liquid
  - Gapless photon
  - Stable only in 3d
- Gapless "topological spin metal" at surface
- Magnetic monopole excitations carry spin or charge?



metal-TBI transition

- Long-range Coulomb: excitons

c.f. Halperin, Rice (1968)

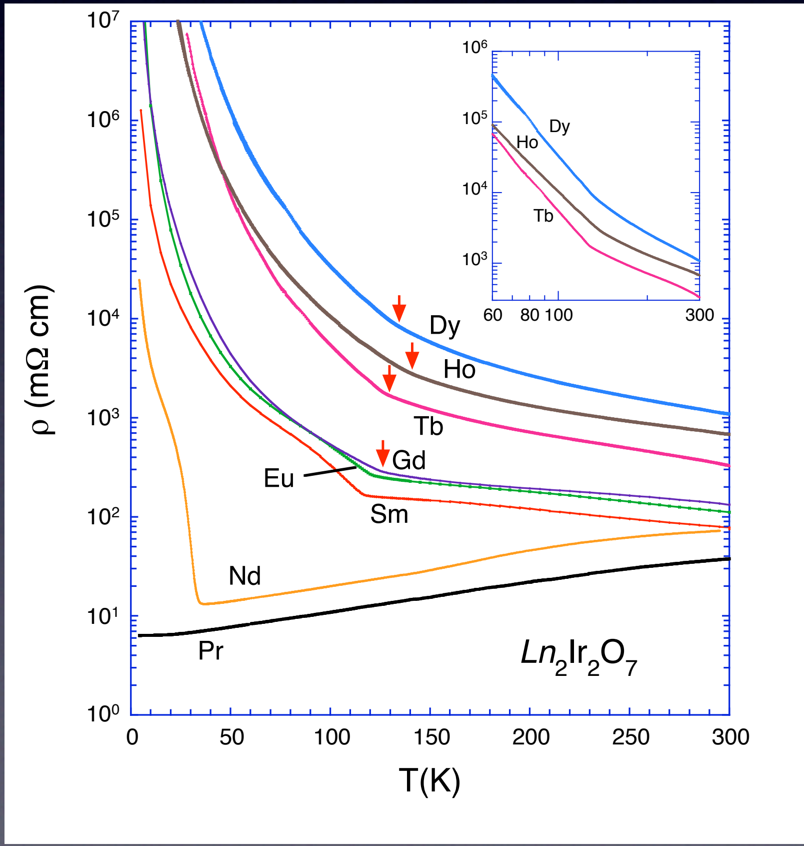


probably weakly magnetic

Back to iridates

K. Matsuhira et al, 2007

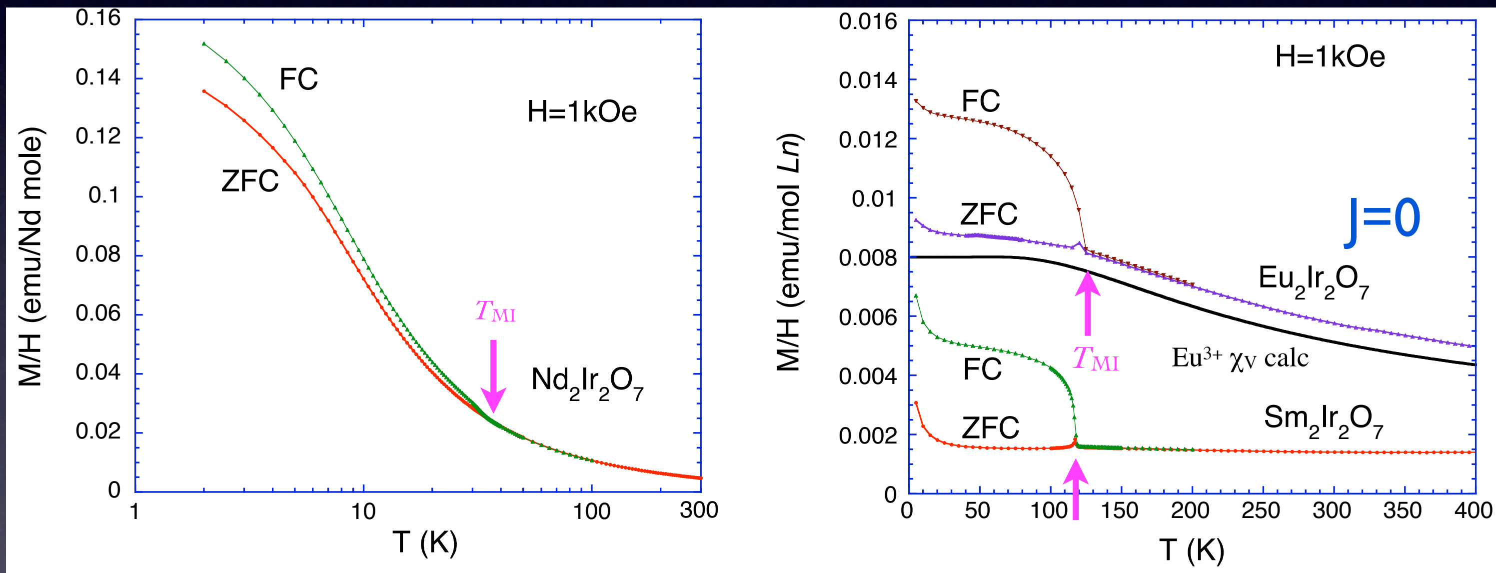
- Experiments show continuous T>0 MITs



Back to iridates

K. Matsuhira et al, 2007

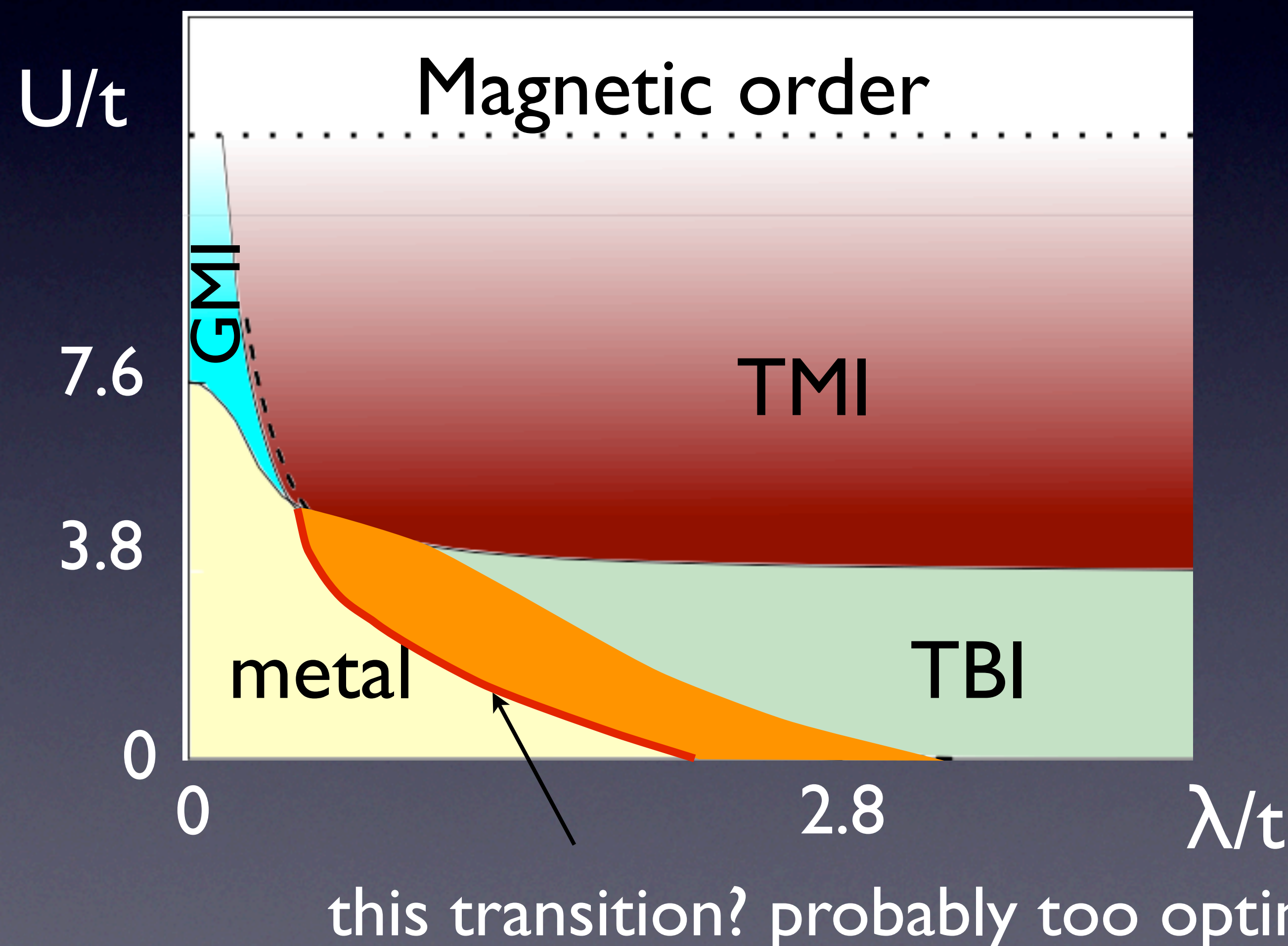
- Experiments show continuous T>0 MITs



closest to QCP

metal-TBI transition

- Perhaps consistent with an excitonic state?



Other possibilities?

- X. Wan *et al* predict an antiferromagnetic Mott state using LSDA+U+SO methods, and find a *non-topological* band structure
- A. Vishwanath *et al* find that at intermediate U/W a magnetic "semimetal" with 3d Dirac nodes obtains
- Clearly more experiments are needed here!

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?

References - $FeSc_2S_4$: PRL 102, 096406 (2009), PRB 80, 224409 (2009).
$Na_4Ir_3O_8$: PRB 78, 094403 (2008); Mott+SO: Nat. Phys. 6, 376 (2010).