Spin-orbit physics in the Mott regime

Leon Balents, KITP

February 2010

The David and Lucile Packard Foundation

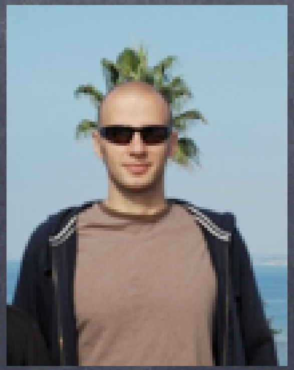
Collaborators

- $FeSc_2S_4$:

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

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

Warning: no DMFT!
(Maybe there should be some?)

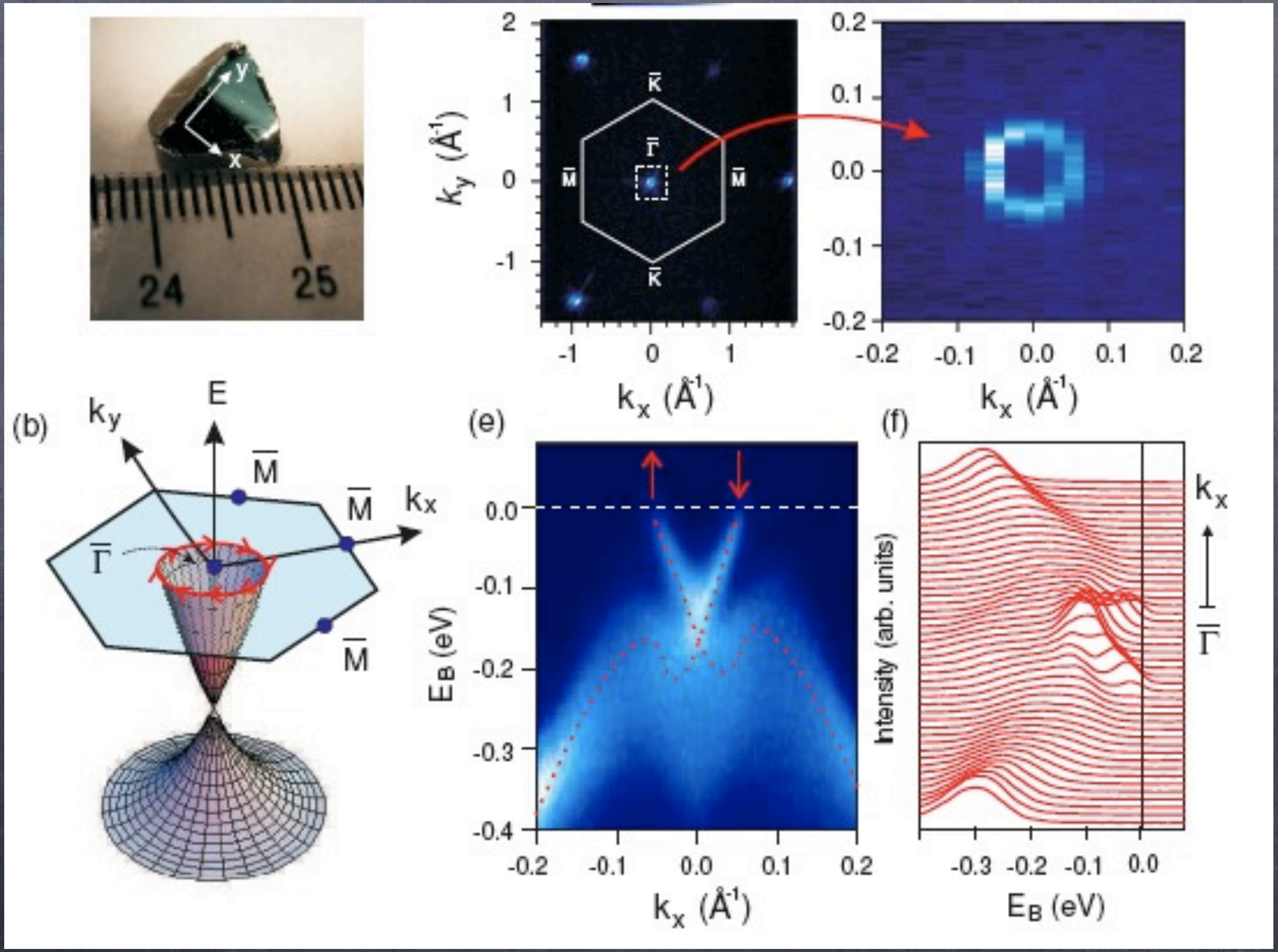
Topological Insulators

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?

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 - $\mathrm{Na_2IrO_3}$

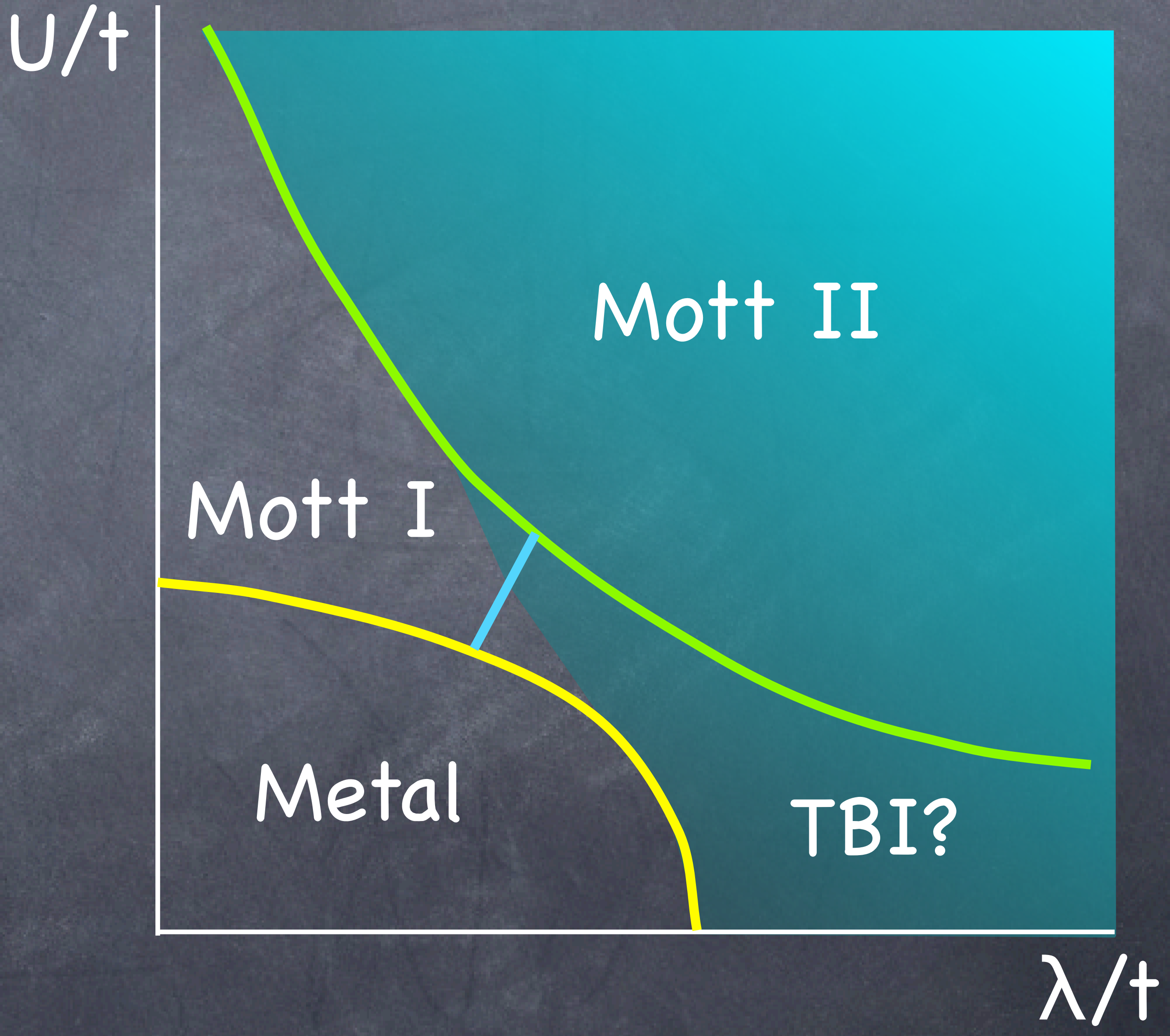
A. Shitade et al, 2009
H. Jin et al, arXiv:0907.0743

- 2d Fractionalized QSHE - spin-charge separated TI

M. W. Young et al, 2008

Materials perspective

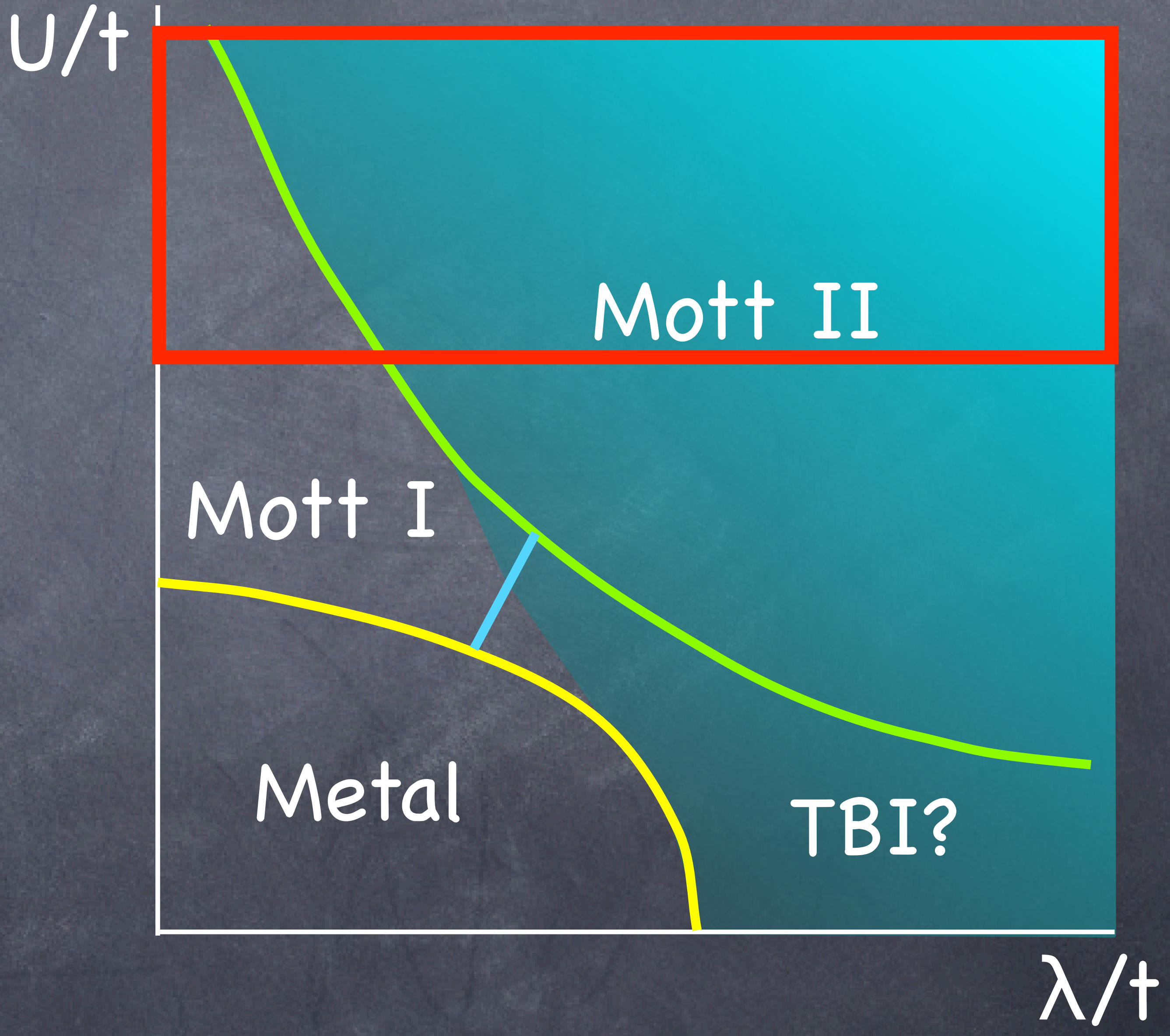
- Coulomb correlations reduce bandwidth
  - Spin-orbit enhanced relative to bandwidth
- In Mott insulator, compare SO to J not t.



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

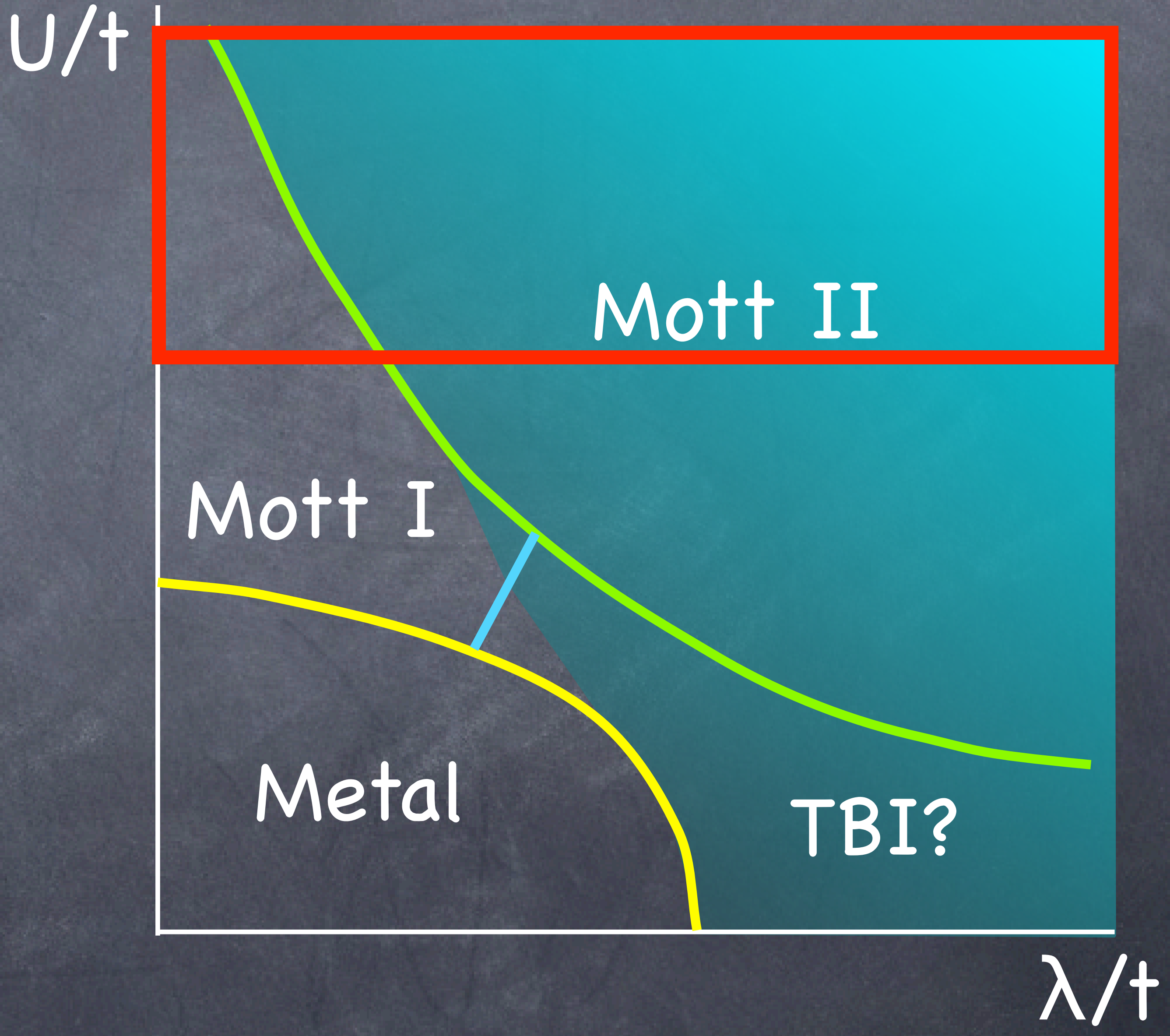
- some Fe and Co compounds, e.g. $FeSc_2S_4$ - orbitally degenerate spinel
- 4d and 5d double perovskites - Ba_2NaOsO_6, Ba_2LiOsO_6 etc.
- However, even in strong MIs with “weak” SOIs (e.g. Dzyaloshinskii-Moriya coupling at few % level), the SOIs can control the ground state when the exchange interactions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

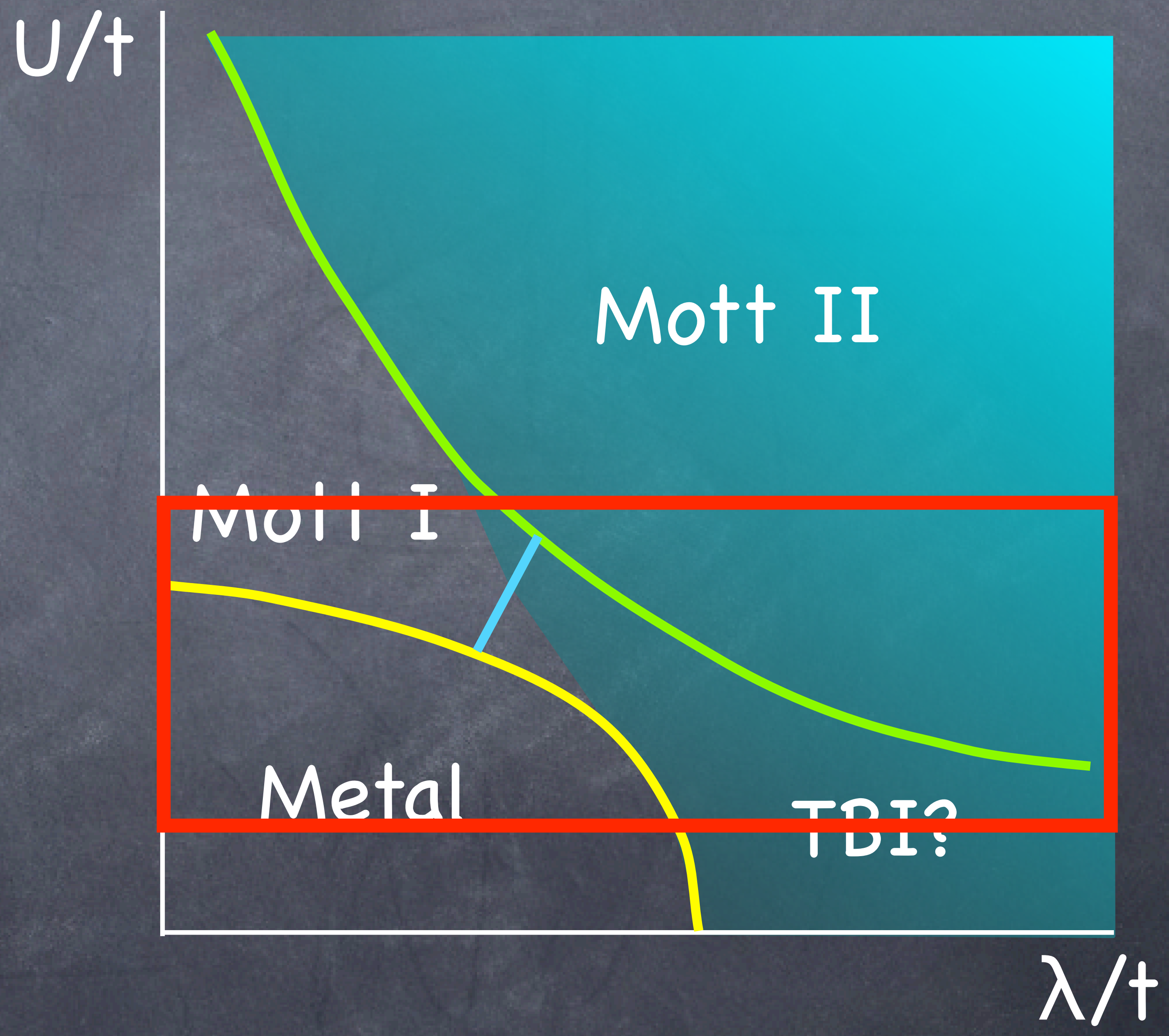
- intermediate regime



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)

Weak Mott Insulators with strong SOIs

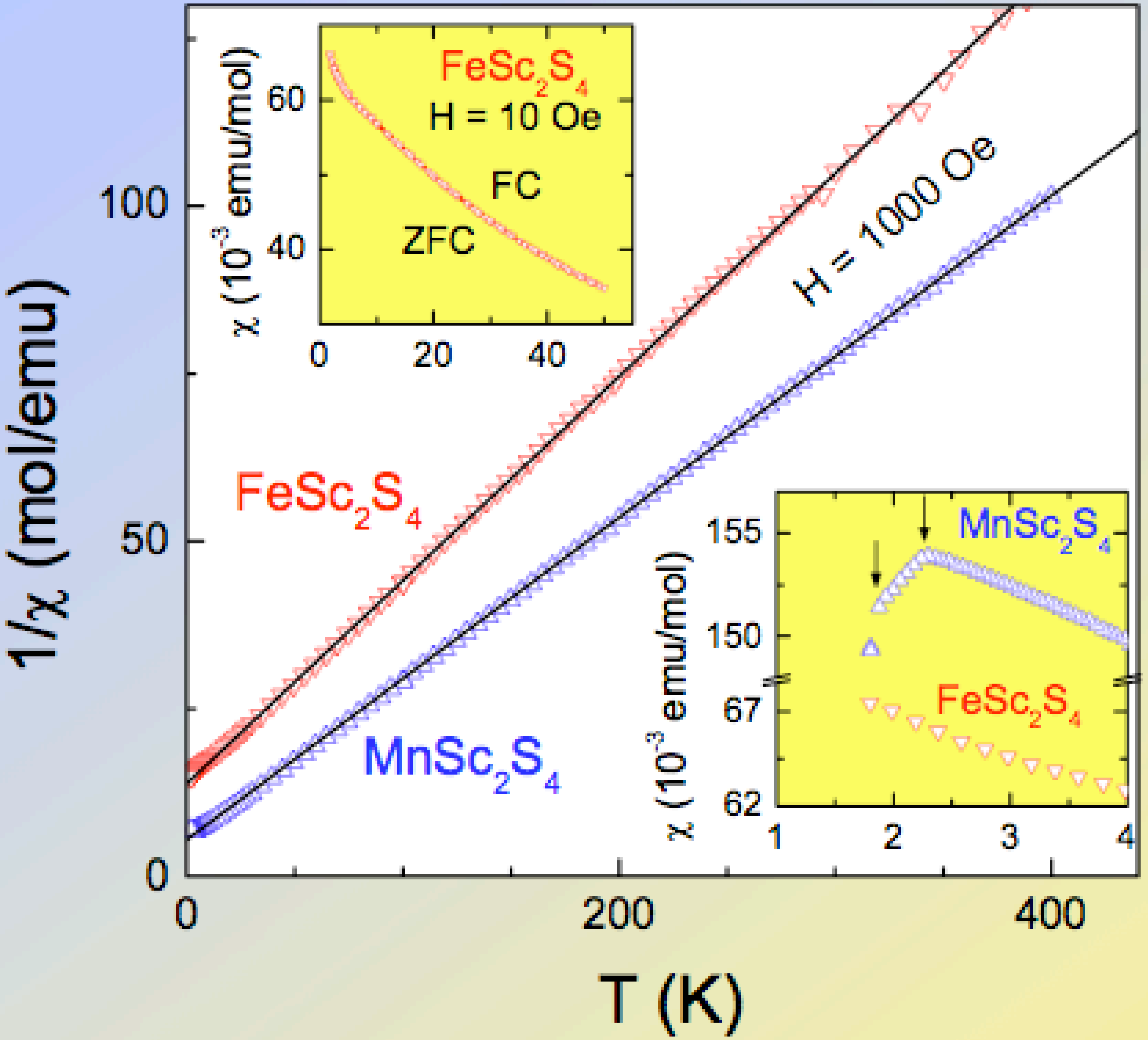
- Most 5d TM ions have smallish U≈1eV, and hence tend to be either metallic or weak Mott insulators
    - together, SOI and U can conspire to produce an insulating state
- e.g. 5d iridates - Sr_2IrO_4, Na_2IrO_3, $Na_4Ir_3O_8$ (hyperkagome), $Ln_2Ir_2O_7$ (pyrochlores)

$FeSc_2S_4$: 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

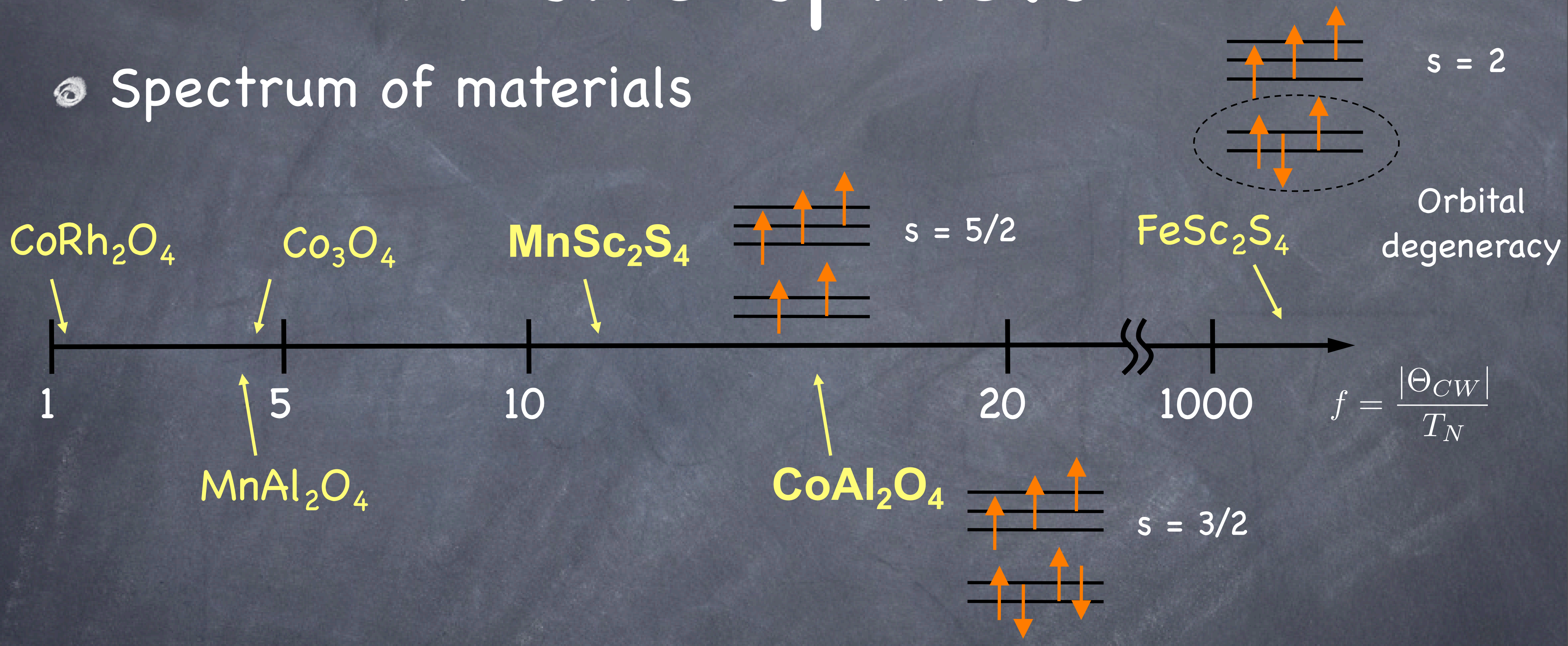
$MnSc_2S_4$: $\theta_{CW} = 25$ K

AFM transition @ 2 K

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

A-site spinels

- Spectrum of materials



$CoRh_2O_4$

Co_3O_4

$MnSc_2S_4$

s = 5/2

$FeSc_2S_4$

s = 2

Orbital degeneracy

1

5

10

20

1000

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

$MnAl_2O_4$

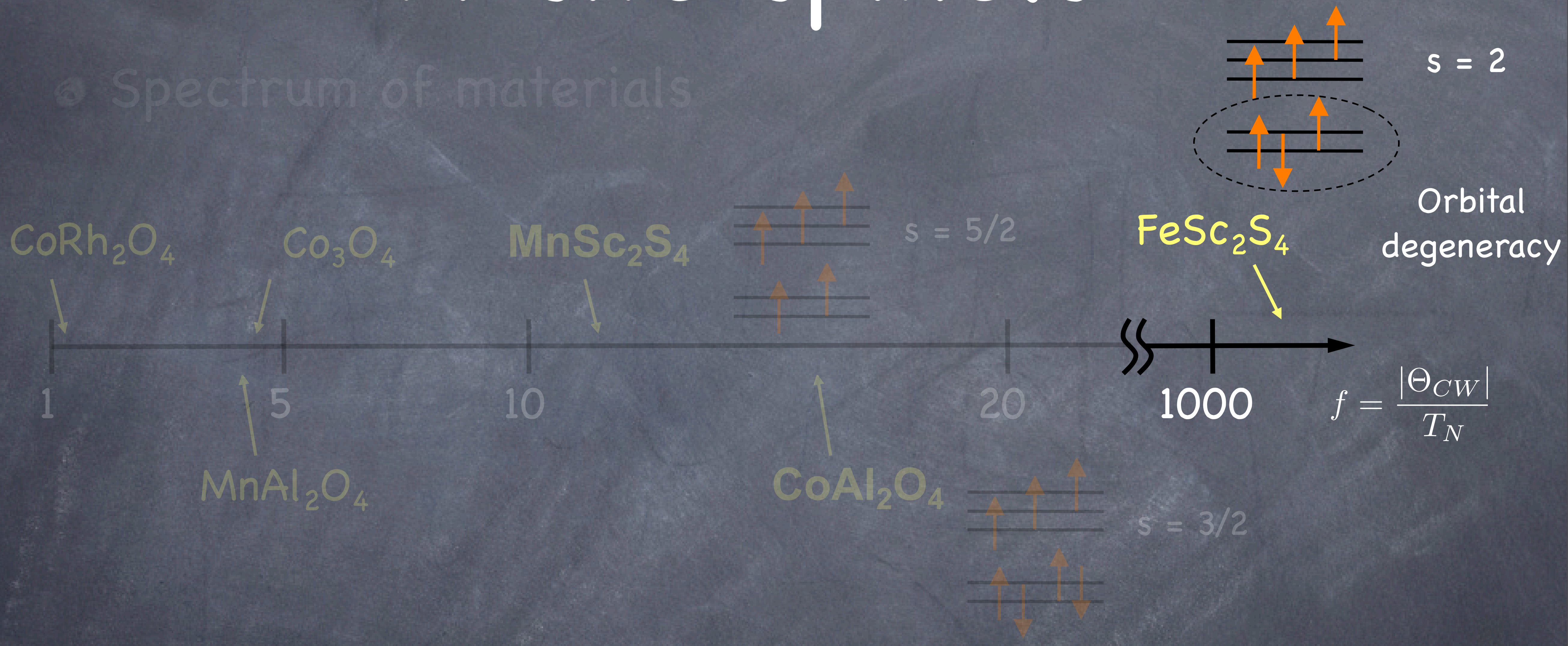
$CoAl_2O_4$

s = 3/2

V. 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

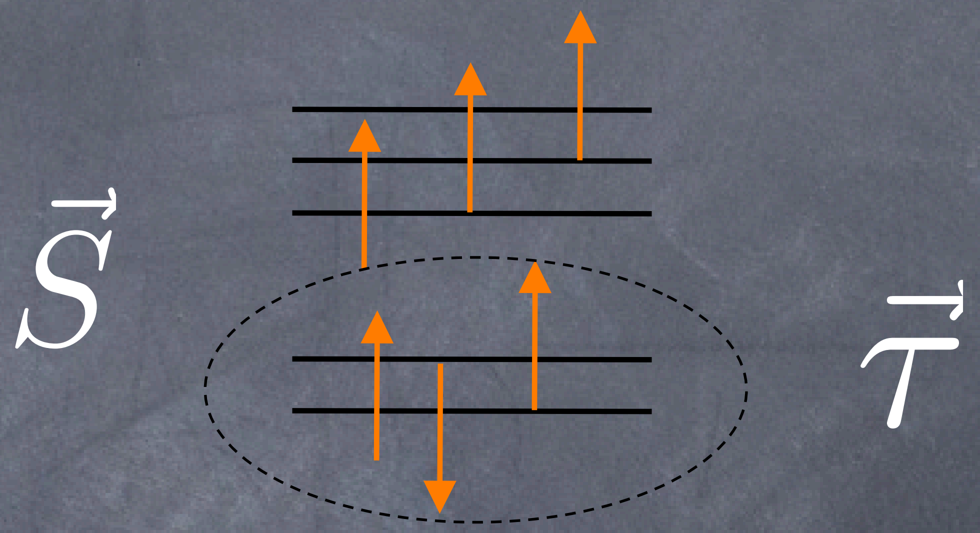


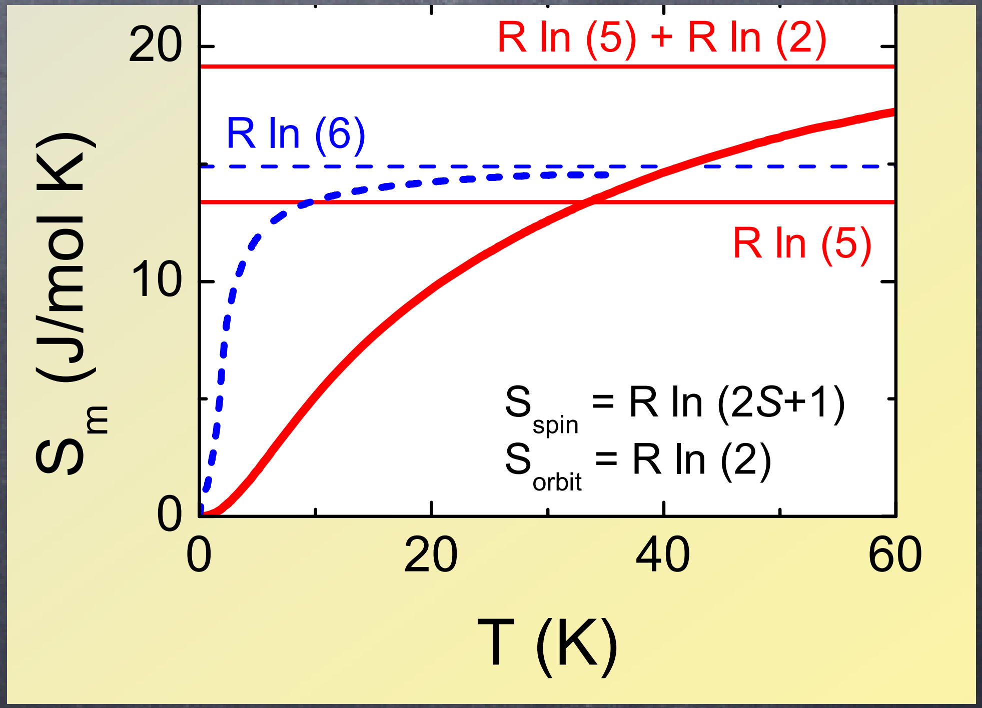
$$f = \frac{|\Theta_{CW}|}{T_N}$$

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

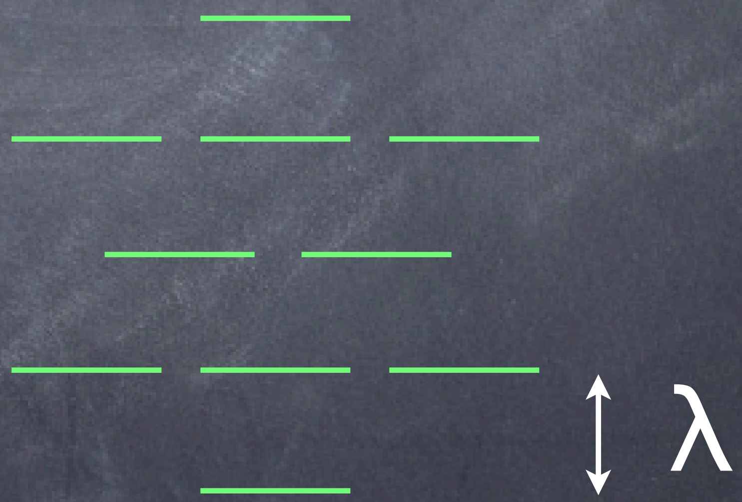
- Separate orbital and spin degeneracy can be split!

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

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

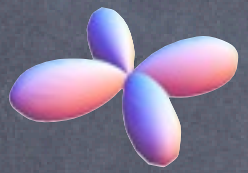
$$\lambda = \frac{6\lambda_0^2}{\Delta}$$

- Naive estimate λ ≈ 25K



Spin orbital singlet

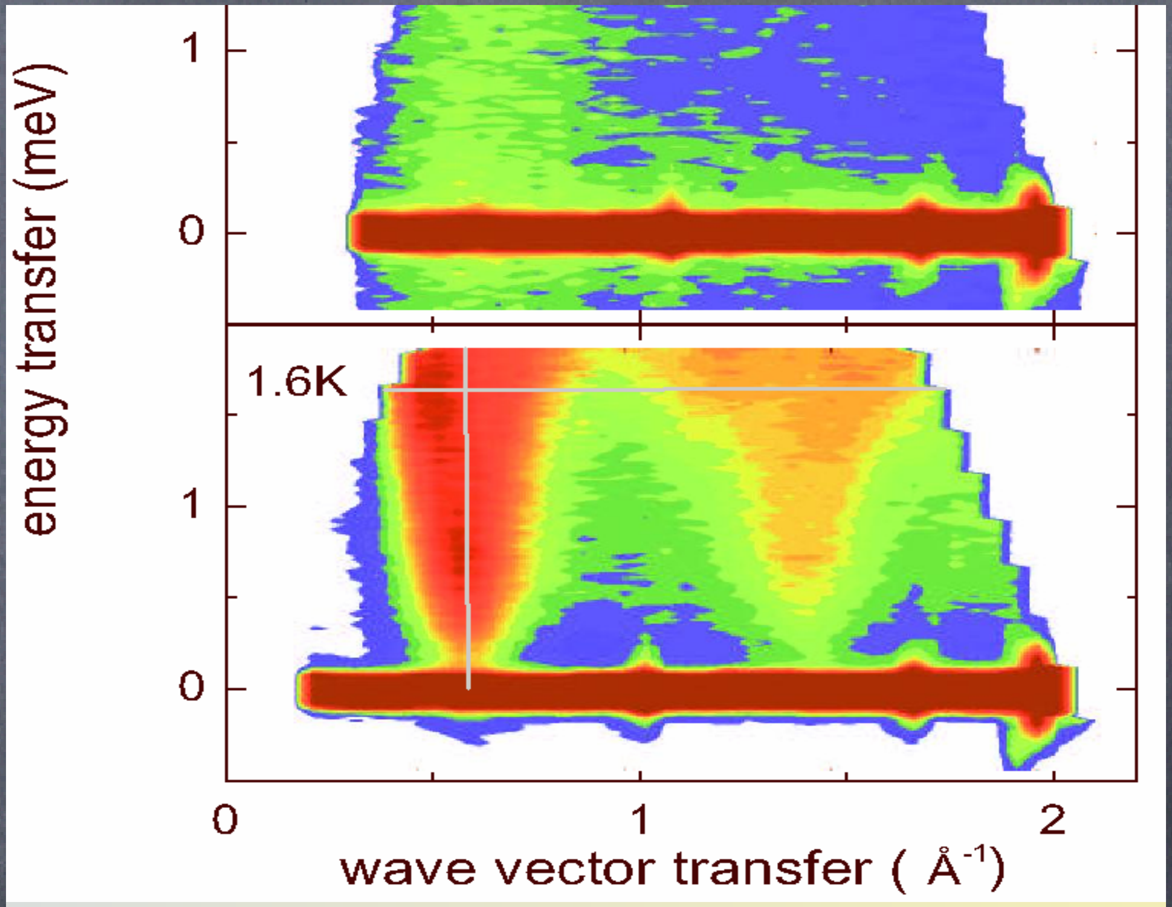
- Ground state of λ>0 term:

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

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

Exchange

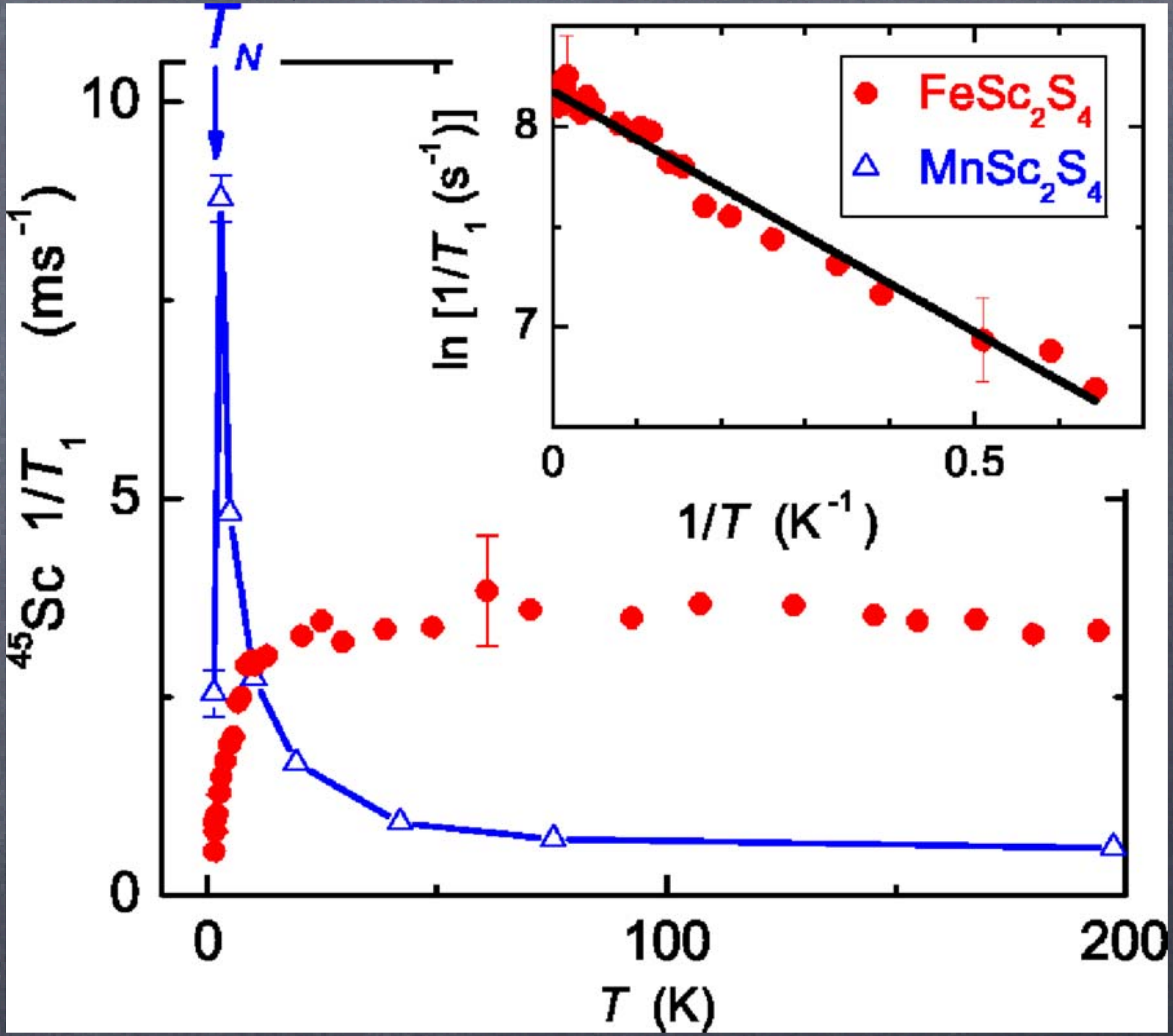
- Inelastic neutrons show significant dispersion indicating exchange
- Bandwidth ≈ 20K similar order as Θ_{CW} and estimated λ
- Gap (?) 1-2K
  - Small gap is classic indicator of incipient order



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)

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

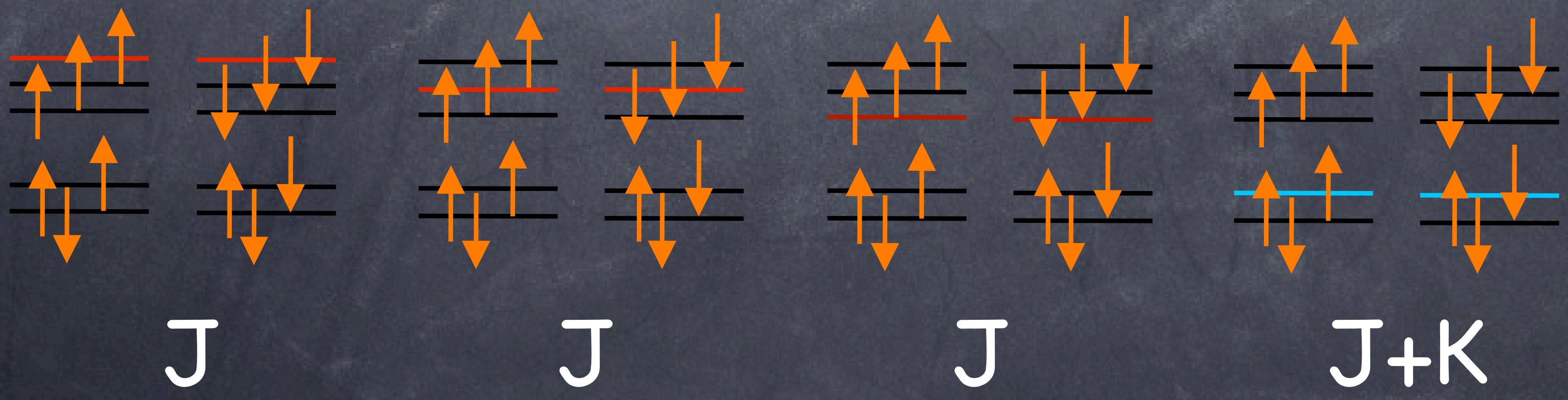
$$+ \Big[L_{ij}\boldsymbol{\tau}_i \cdot \boldsymbol{\tau}_j + \tilde{L}_{ij}\tau_i^y \tau_j^y\Big]\mathbf{S}_i \cdot \mathbf{S}_j\Big\}$$

Exchange

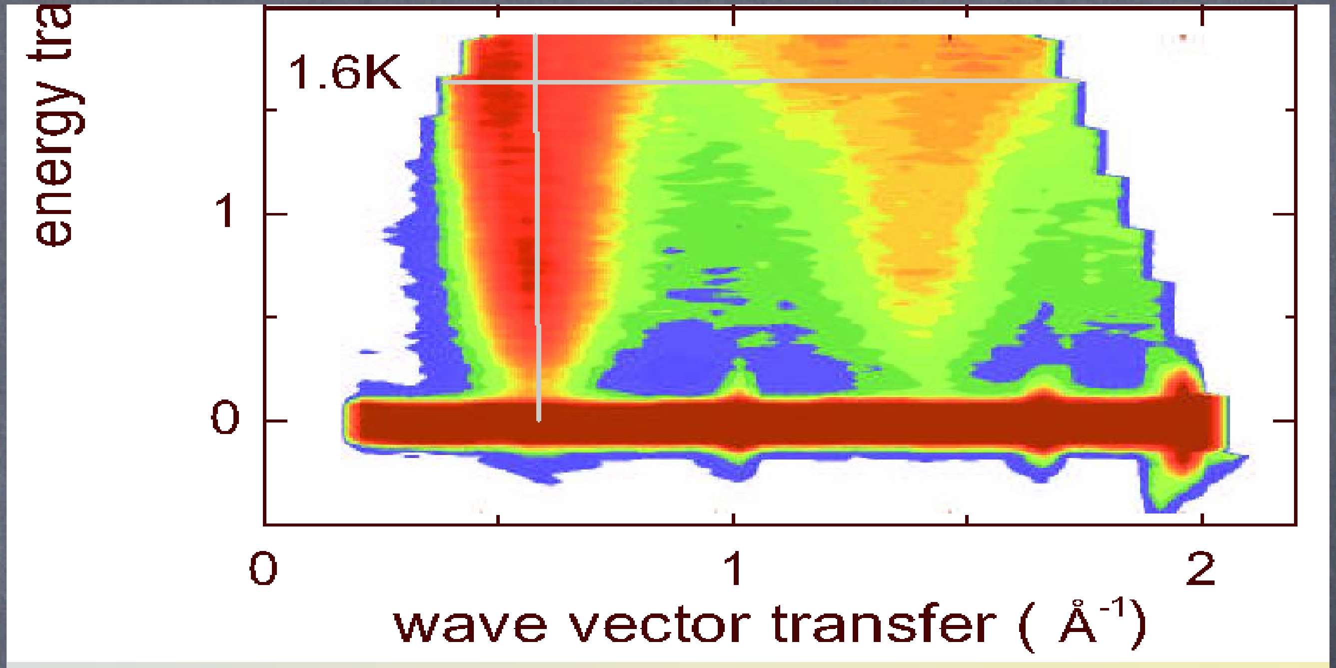
- Largest interaction is just Heisenberg exchange

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

  - More exchange processes contribute



Minimal Model

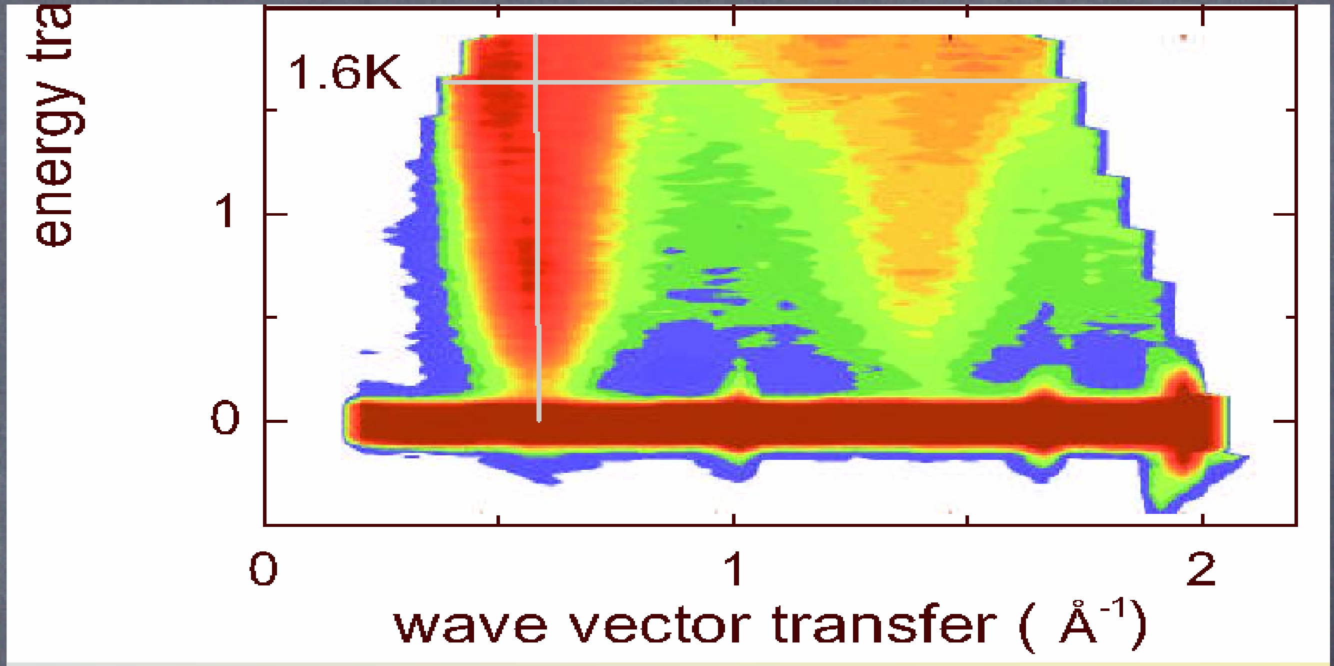


A. Krimmel *et al*., Phys. Rev. Lett. **94**, 237402, 2005

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

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

Minimal Model



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

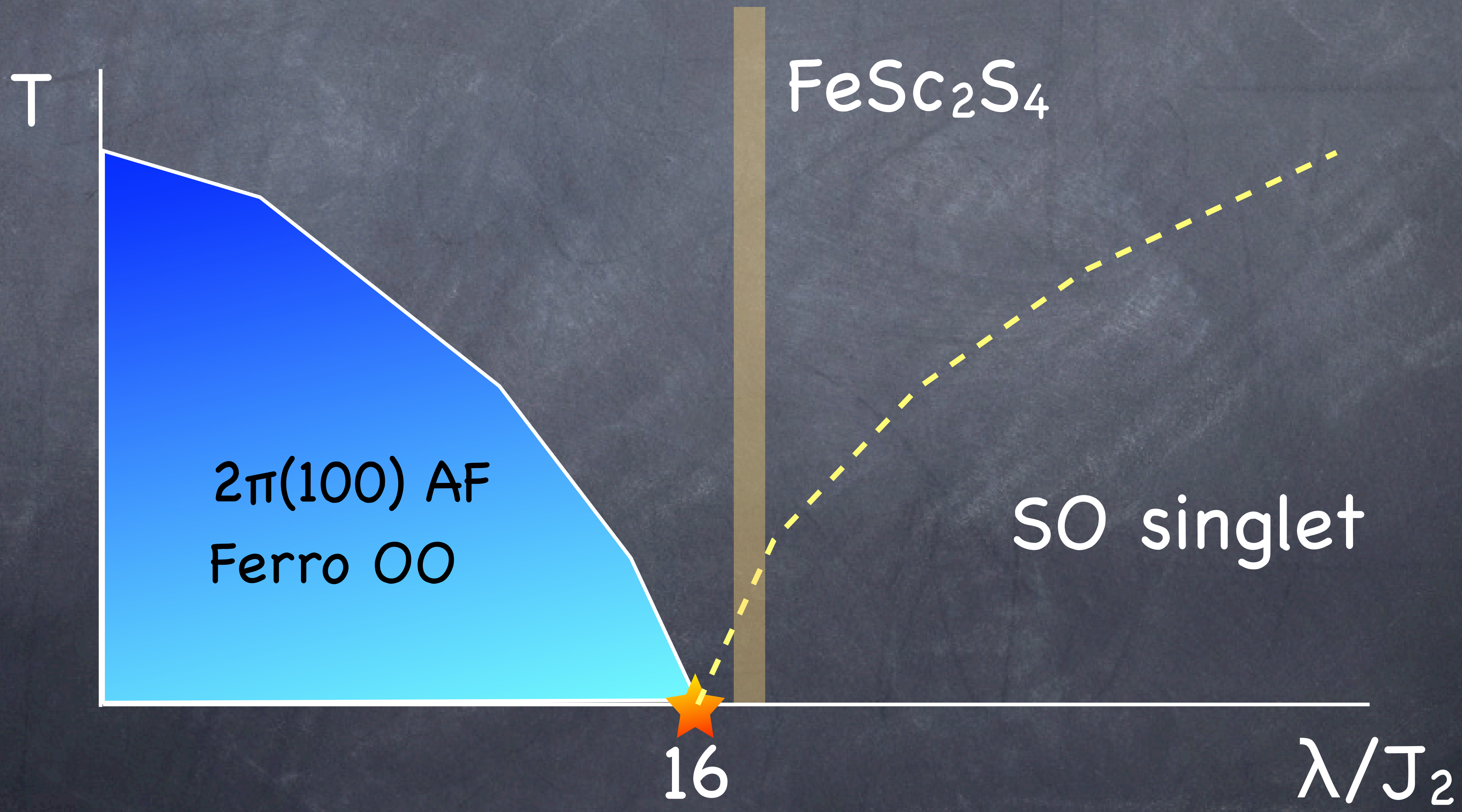
- Neutron scattering suggests peak close to $2\pi(100)$
- Indicates $J_2 \gg J_1$

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

Expect MFT good in 3+1 dimensions

Quantum Critical Point

- Mean field phase diagram



Predictions

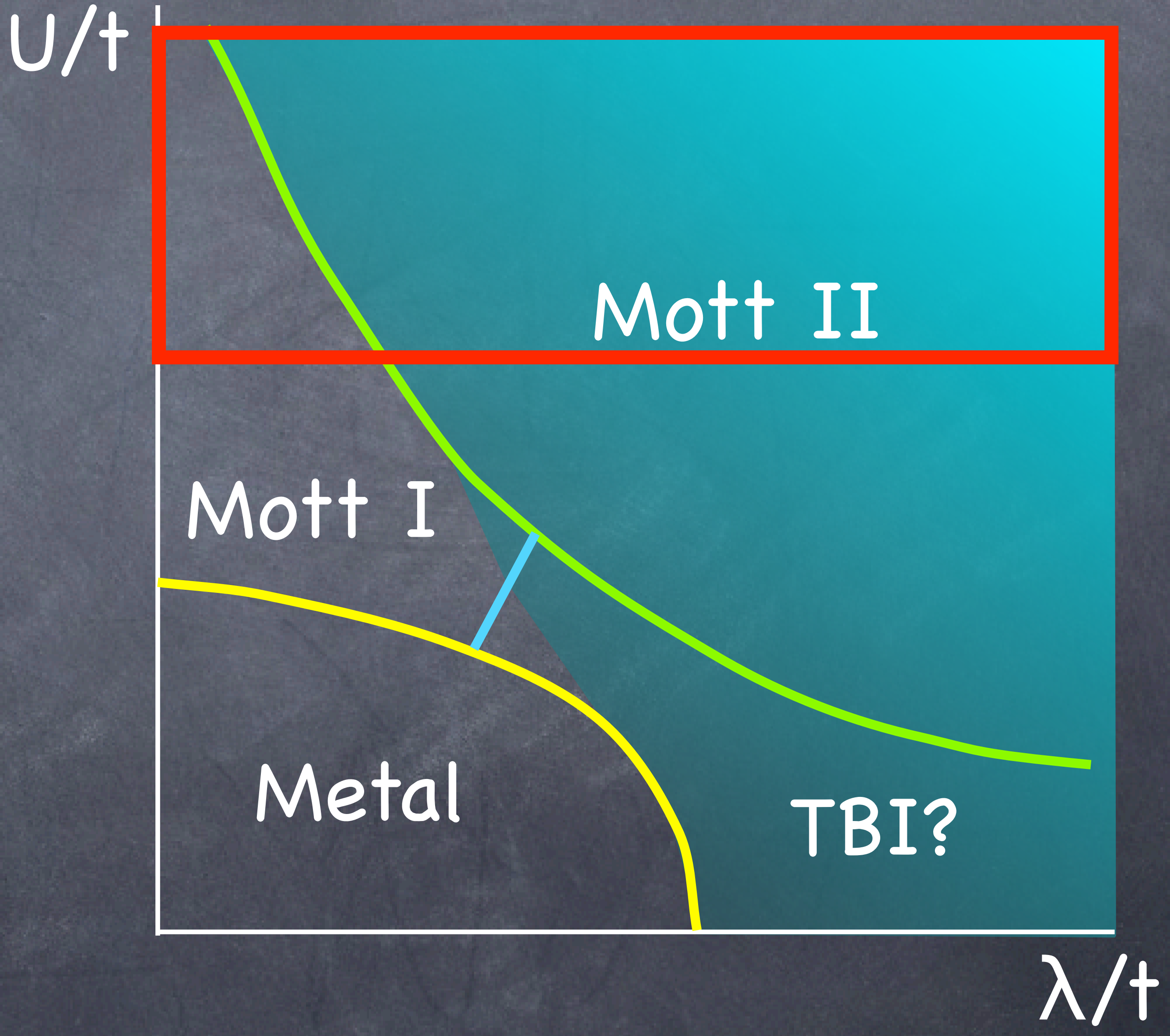
- Large T=0 susceptibility (estimated) ✓
- Scaling form for $(T_1T)^{-1} \sim f(\Delta/T)$ ✓
- Specific heat $C_v \sim T^3 f(\Delta/T)$ ✓
- Possibility of pressure-induced ordering
- Magnetic field **suppresses** order
    - opposite to simple "dimer" antiferromagnet

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 for our papers for more details

Mott transition with SOIs

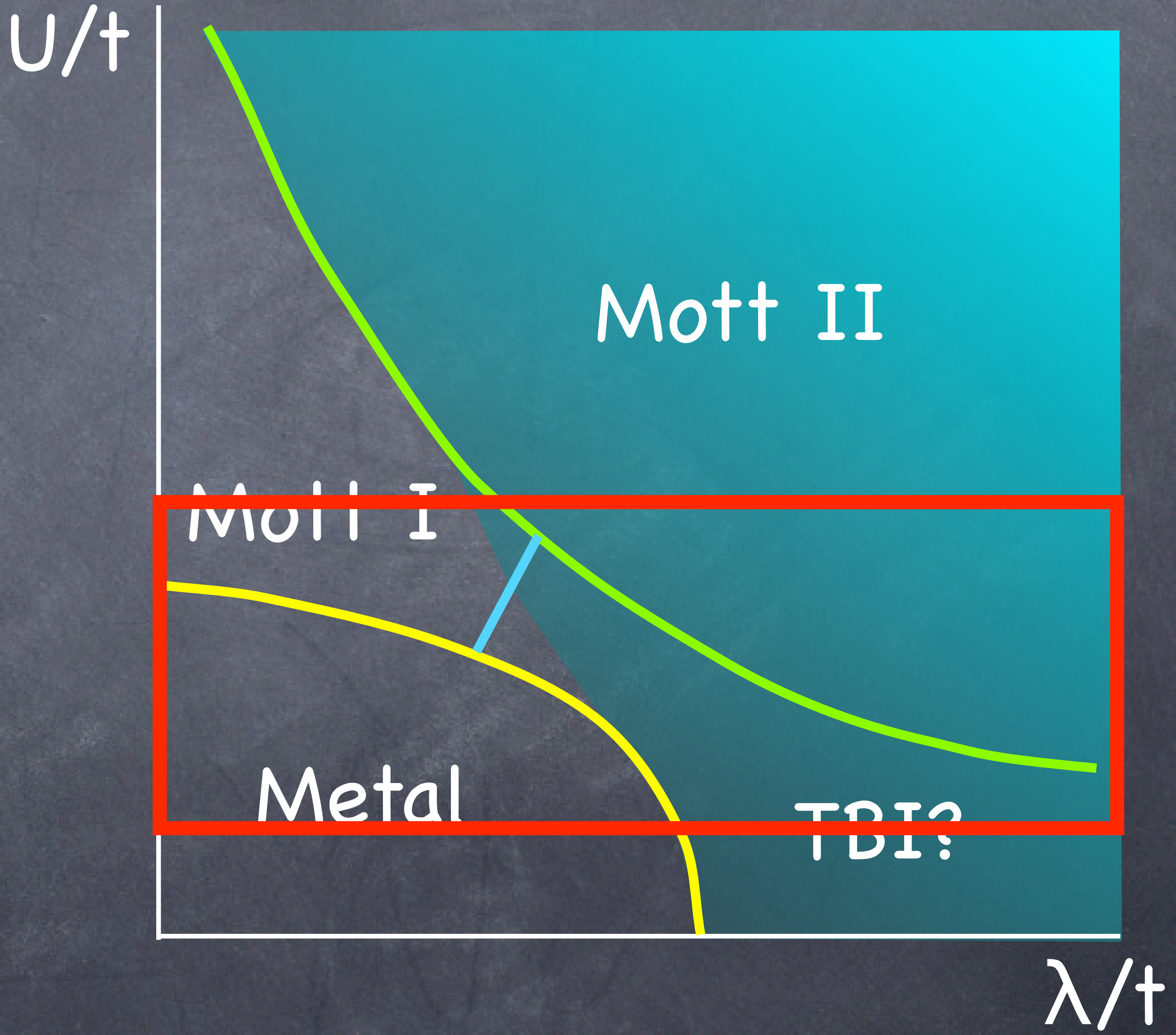
- Study this phase diagram in a concrete case



schematic phase diagram

Mott transition with SOIs

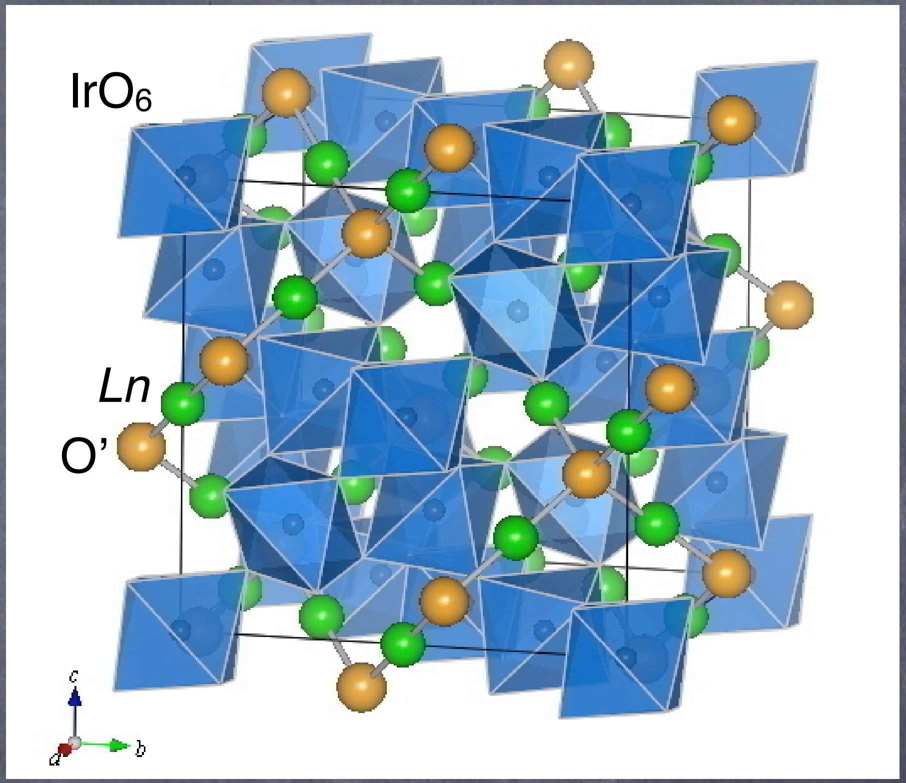
- Study this phase diagram in a concrete case

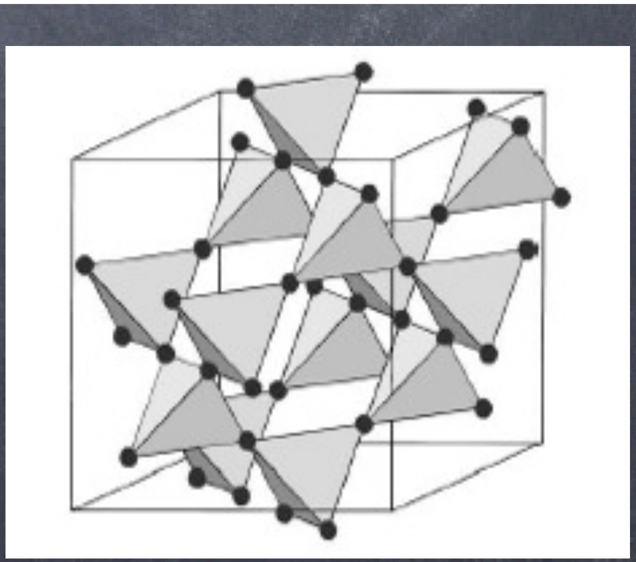


schematic phase diagram

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

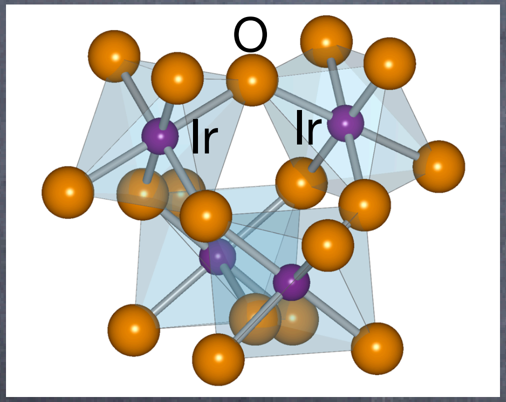


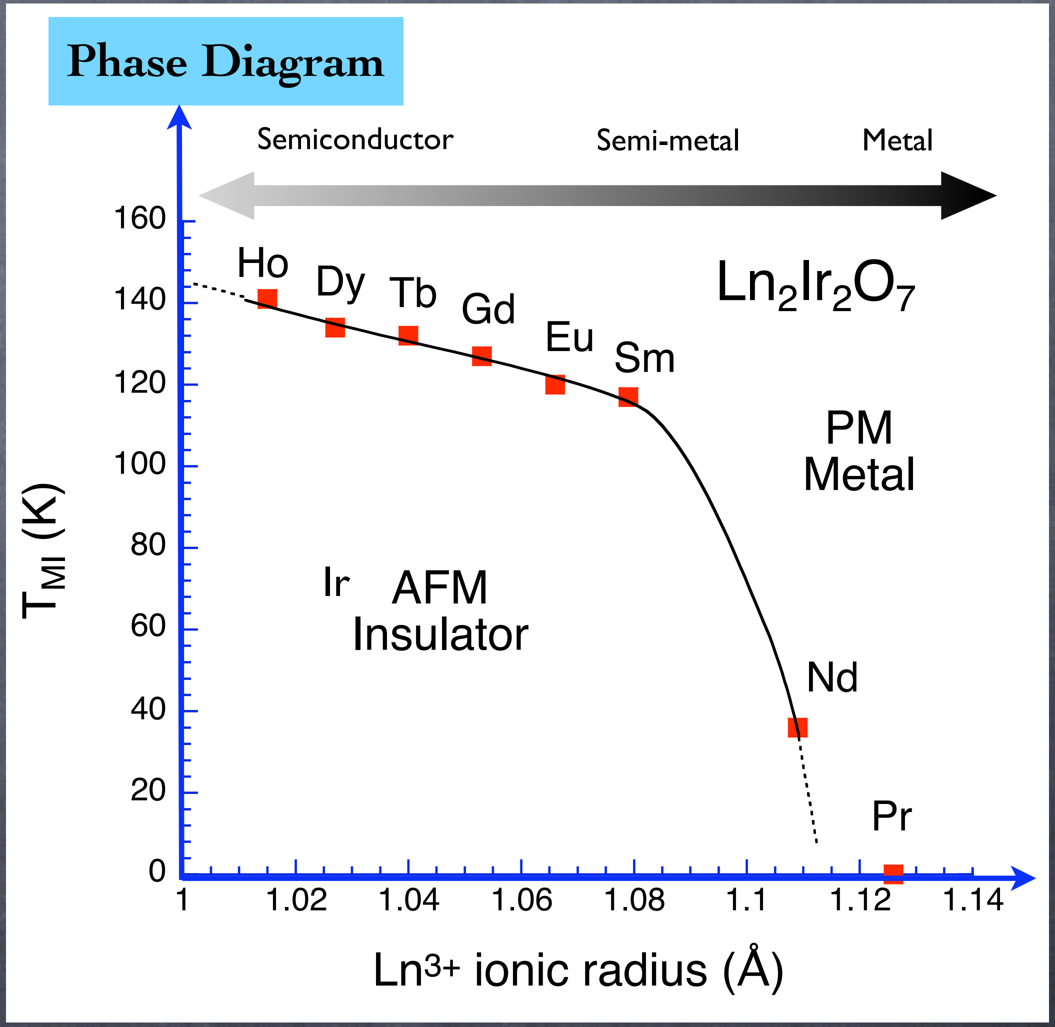


Metal-Insulator Transition

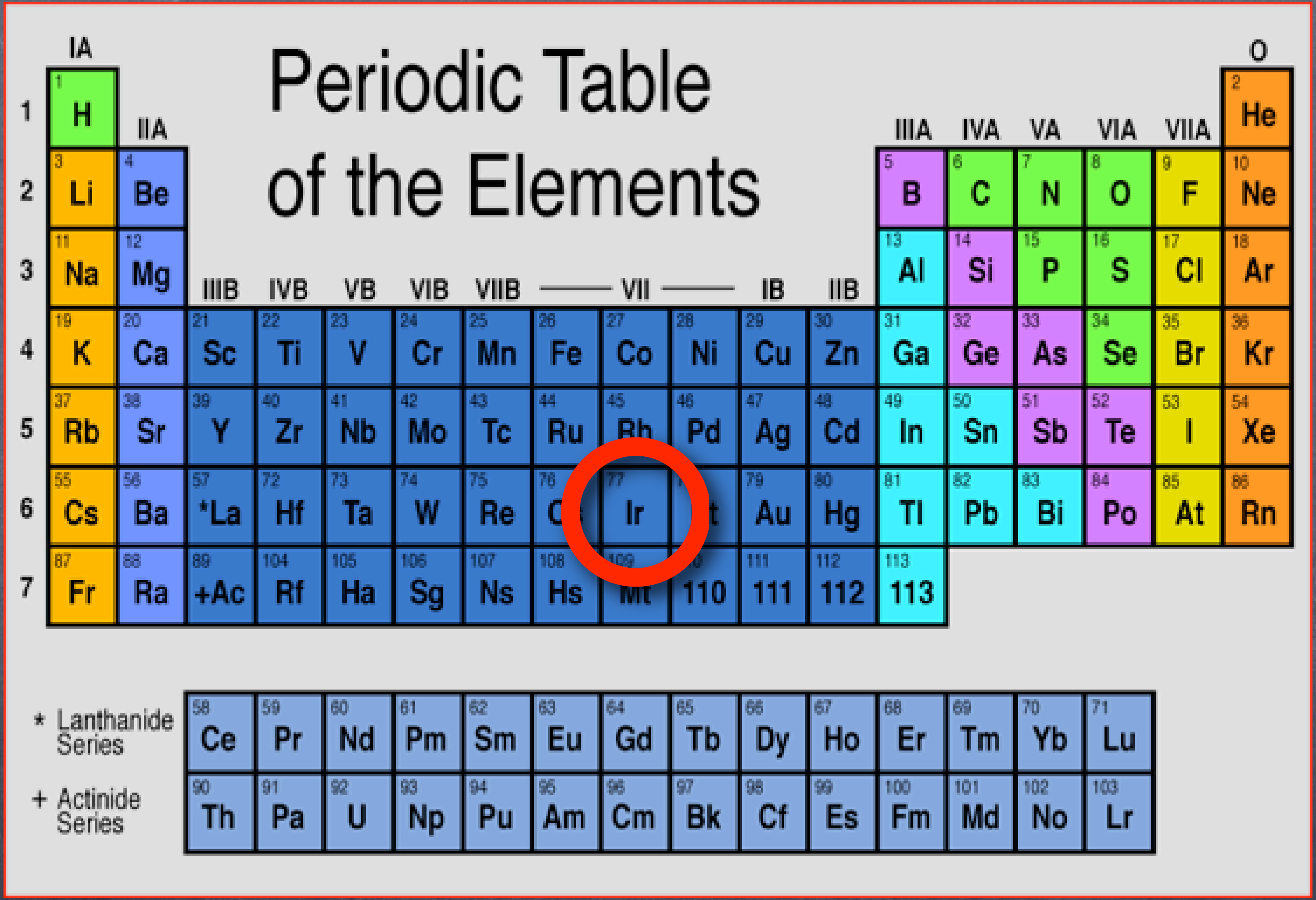
K. Matsuhira et al, 2007

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





Spin orbit coupling



Periodic Table of the Elements

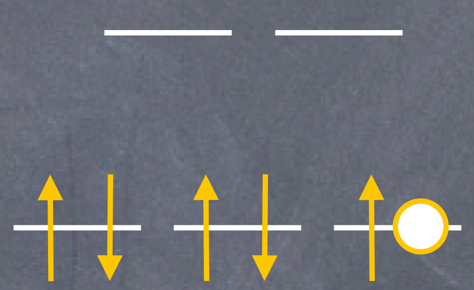
	IA	IIA	IIIB	IVB	VB	VIB	VIIB	VII			IB	IIB	IIIA	IVA	VA	VIA	VIIA	O
1	1 H																	2 He
2	3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
3	11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
4	19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 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 O	77 Ir	t	79 Au	80 Hg	81 Tl	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	111 111	112 112	113 113					

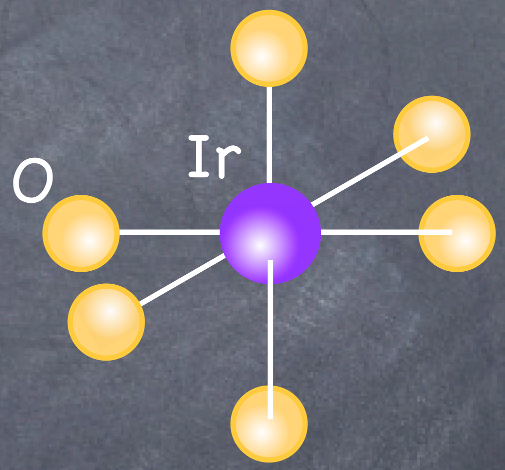
* Lanthanide Series	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
+ Actinide Series	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr

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

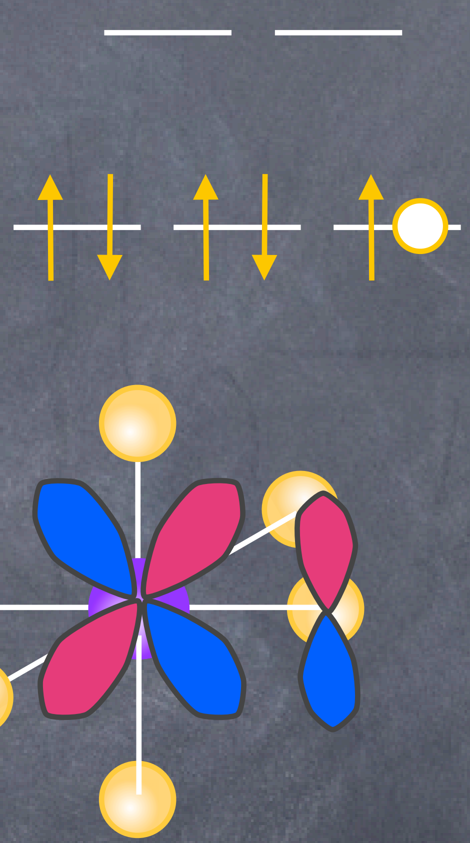
Model

- 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





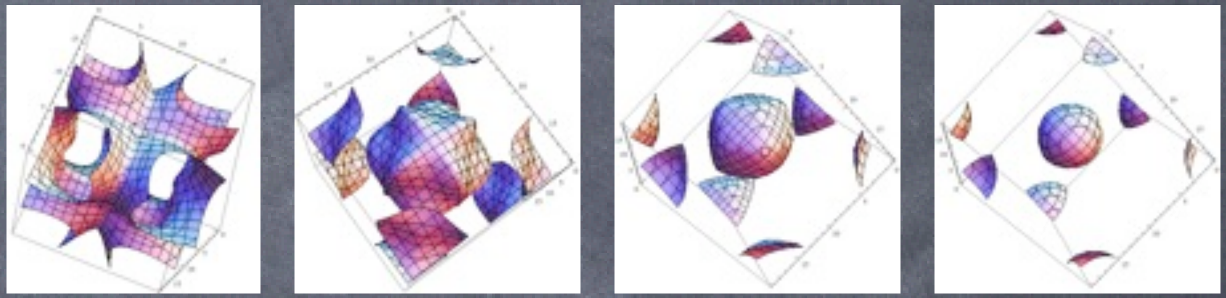
Model

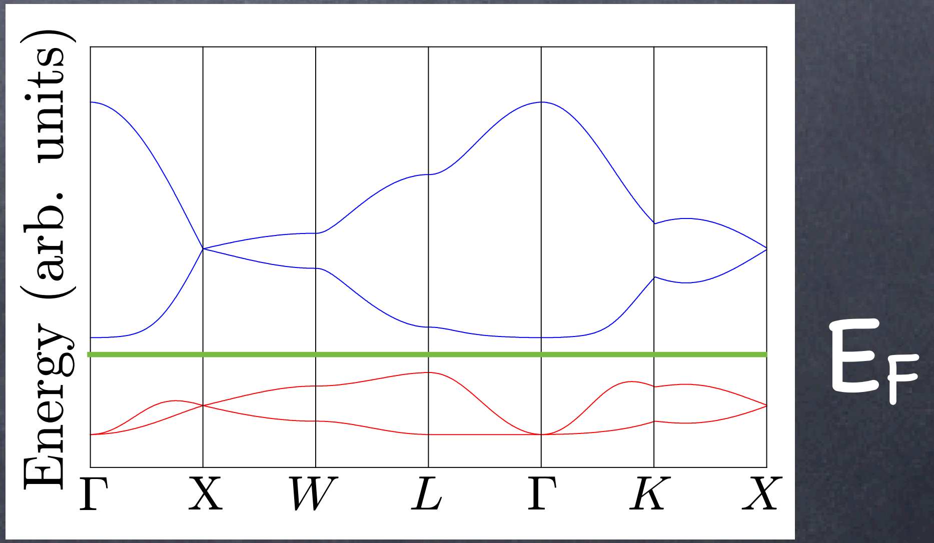


- octahedral Ir^{4+}: $(t_{2g})^5$
  - effective l=1 orbital degeneracy
- Ir-O-Ir hopping
  - dominant $V_{pd\pi}$ channel
- Spin-orbit coupling
  - $H_{SOI} = -\lambda \vec{L} \cdot \vec{S}$
- Hubbard U

U=0 Band Structure

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

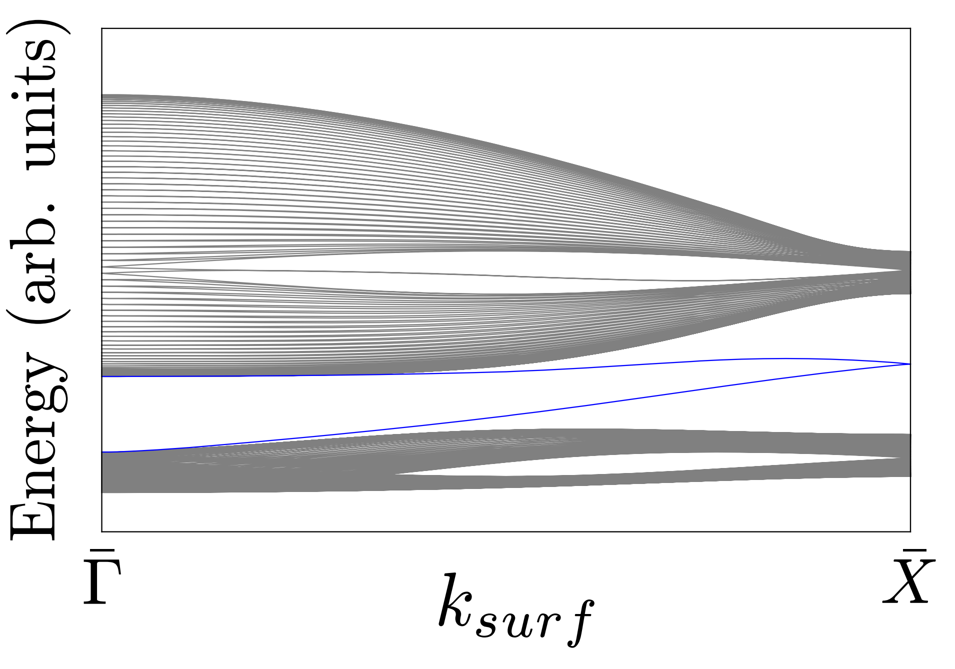




Topological Band Insulator

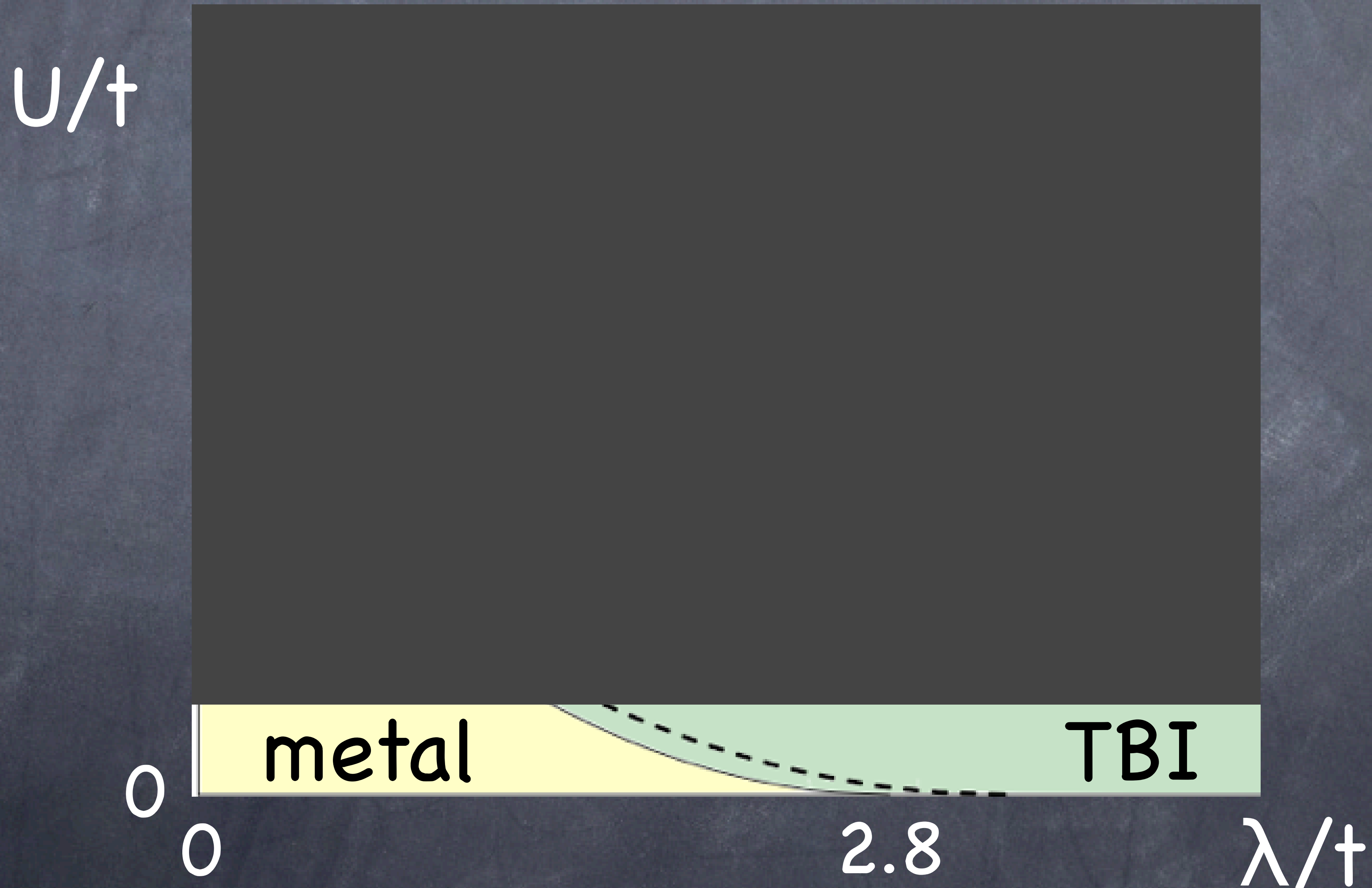
- Inversion Symmetry:
  - Fu-Kane give simple criterion for parity eigenvalues
  - Strong TBI (weak invariants all zero by cubic symmetry)
- Surface states

(100) surface



surface Dirac point

Phase Diagram



Very large U/t

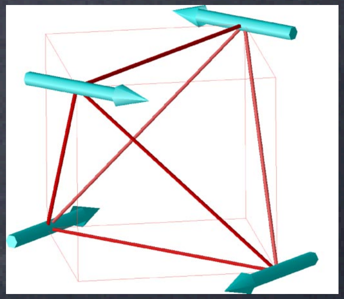
- For λ >> J ~ t^2/U, reduces to Heisenberg "spin" model for j=1/2 eigenstates

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

- 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



Very large U/t

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

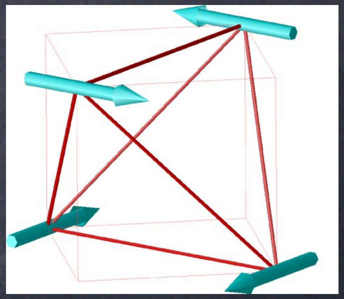
$$H_{spin} = \frac{4t^2}{U}\sum_{ii'}\left[J\vec{S}_i\cdot\vec{S}_{i'} + \vec{D}_{ii'}\cdot\vec{S}_i\times\vec{S}_{i'} + \cancel{\vec{S}_i\cdot\overleftrightarrow{\Gamma}_{ii'}\cdot\vec{S}_{i'}}\right]$$

- This model has been extensively studied

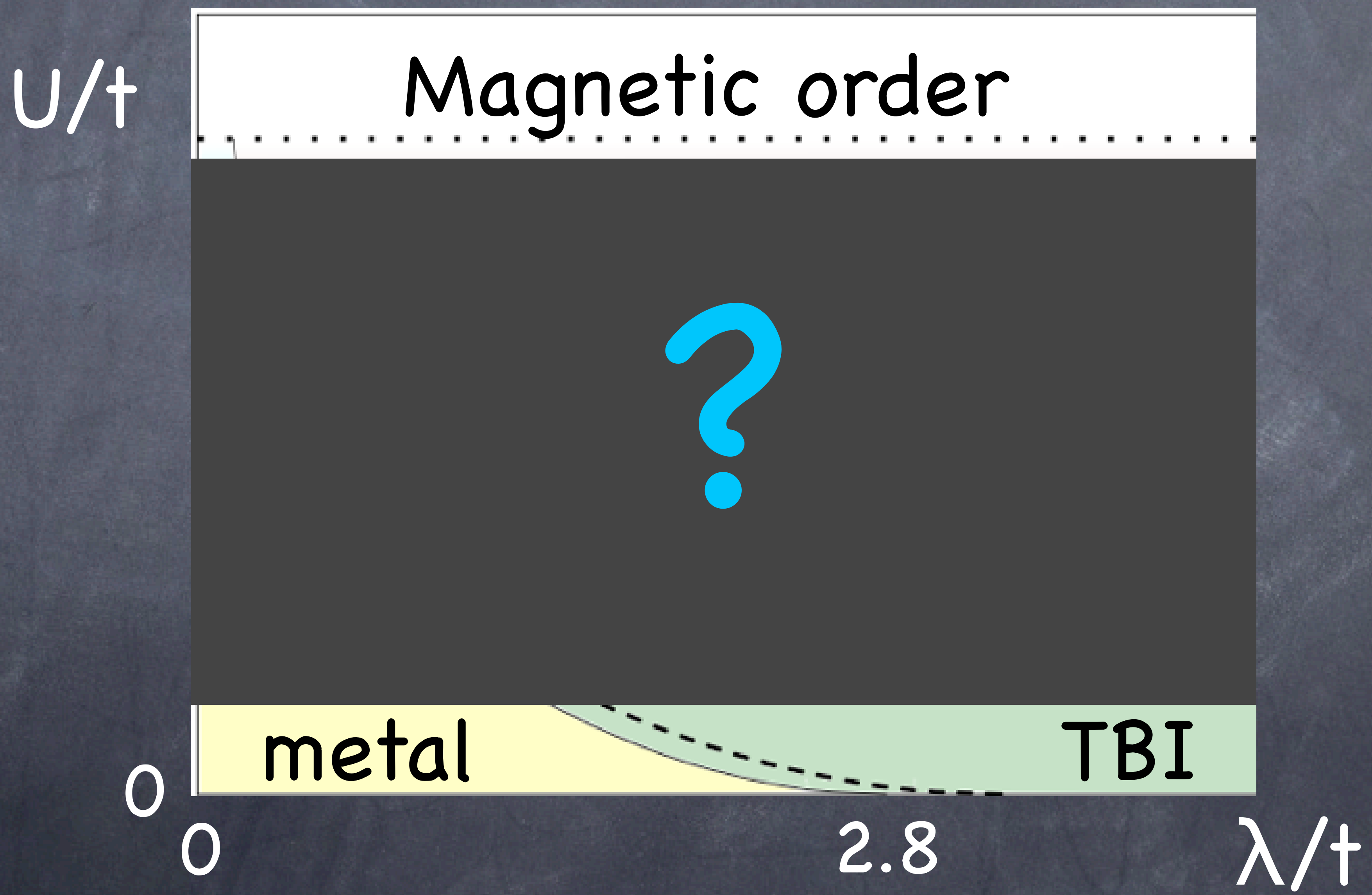
Elhajal et al, 2005

  - 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



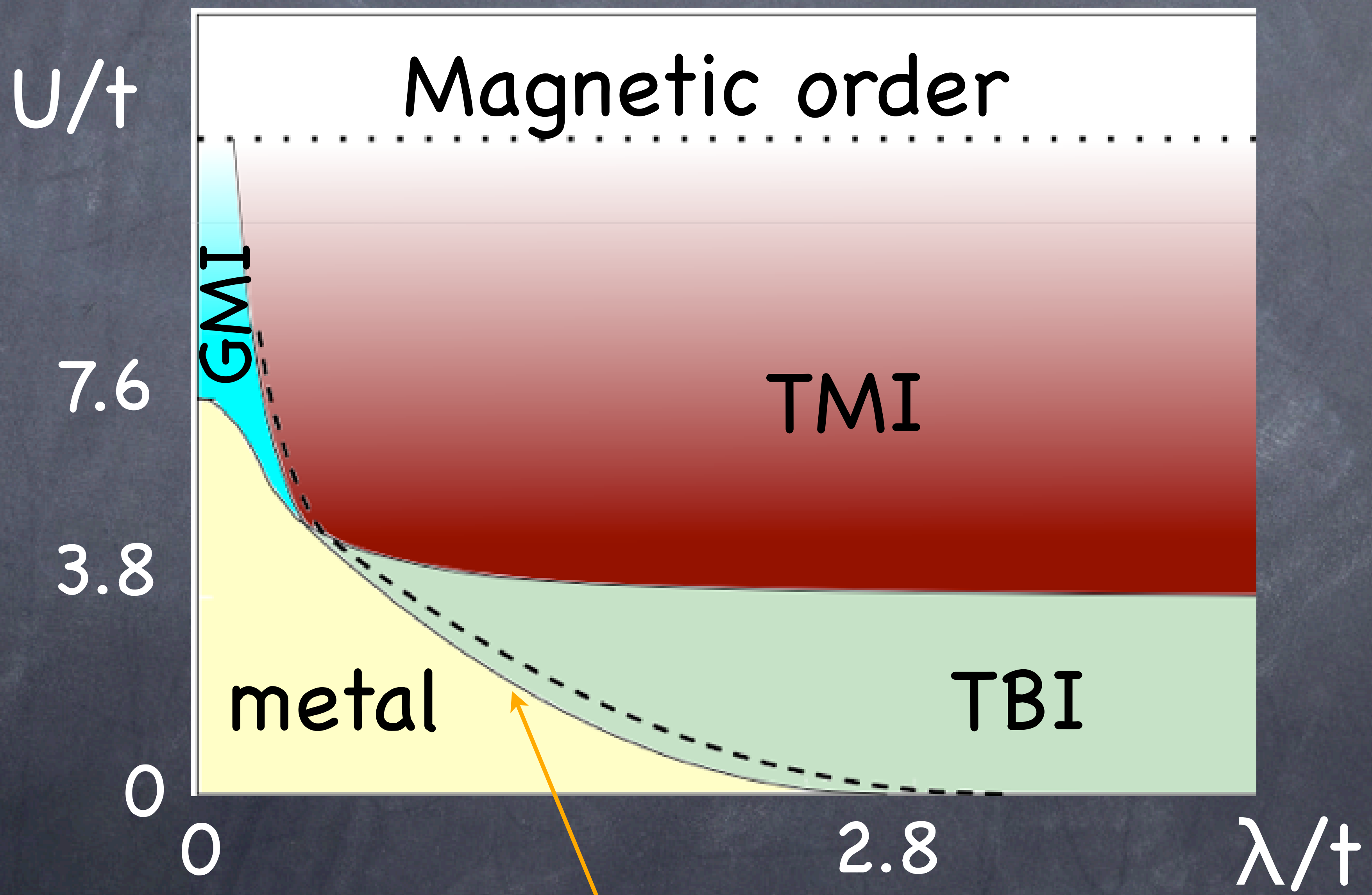
Phase Diagram



Intermediate U

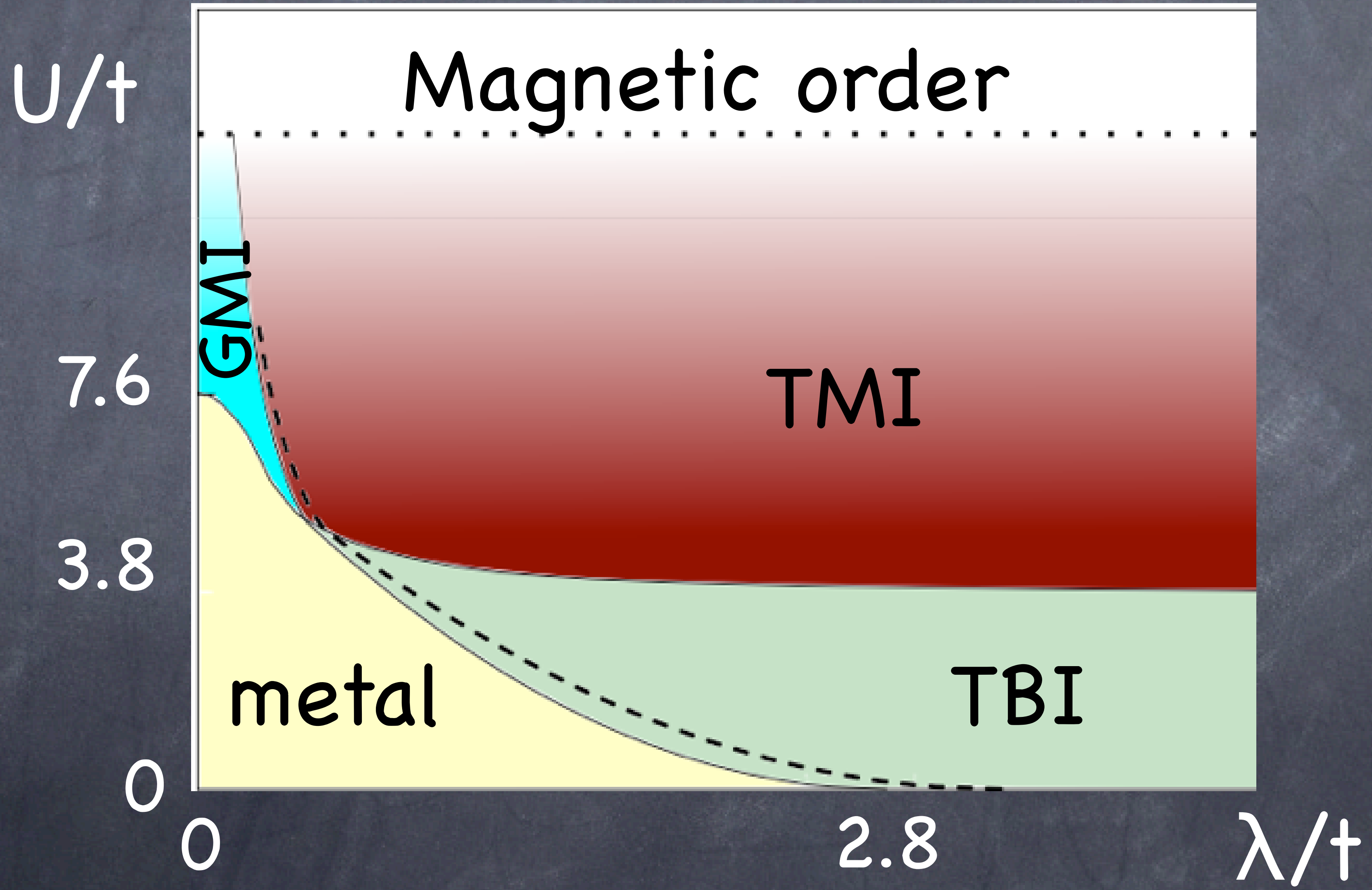
- Slave-rotor approximation Florens, Georges (2004)
  - Seems to give qualitatively reasonable results for frustrated Hubbard models (triangular, checkerboard, hyperkagome) in agreement with several numerical approaches
  - Does **not** describe nesting/SDW physics
- Simple to implement $c_a^\dagger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

Phase Diagram



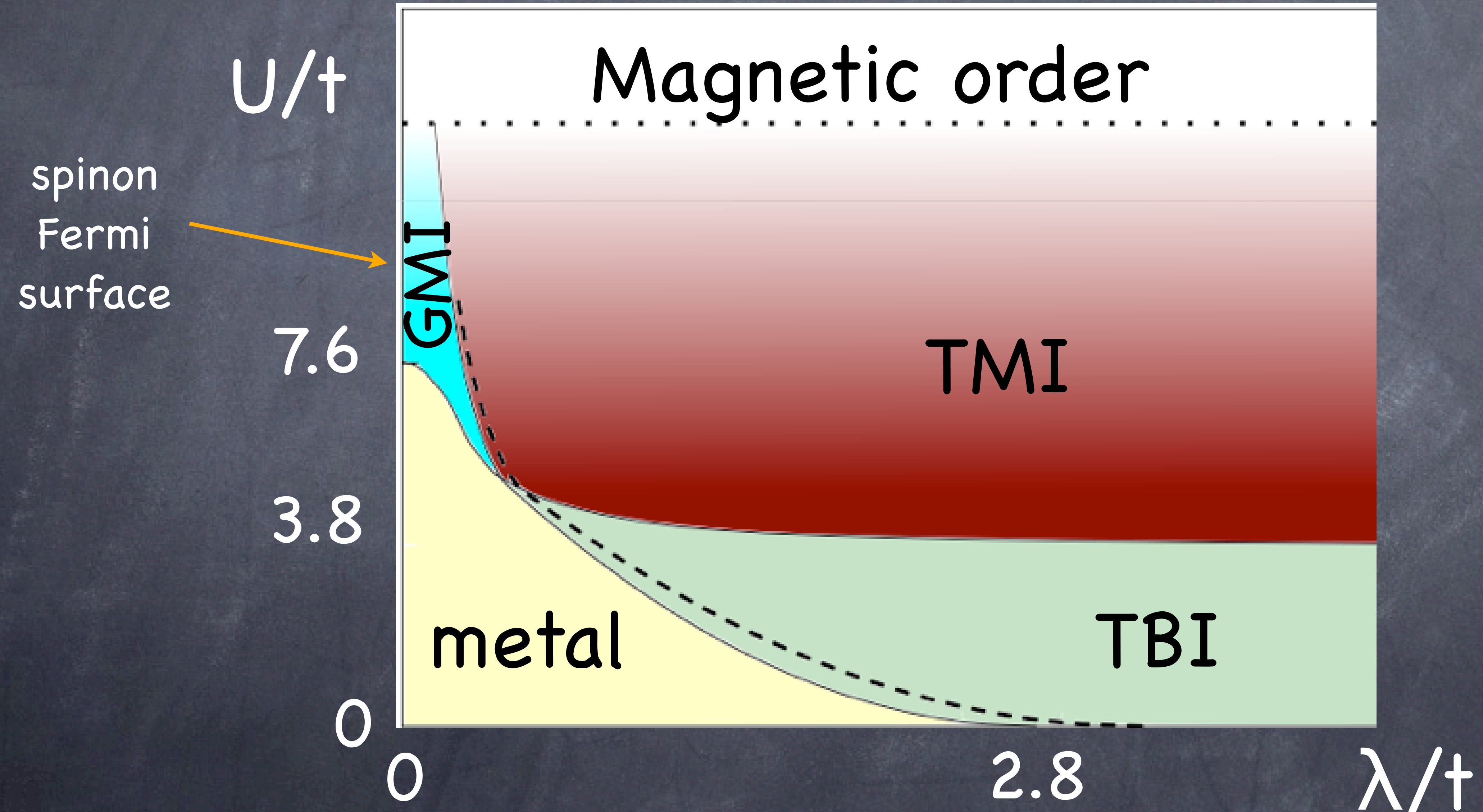
Bandwidth suppression

Phase Diagram

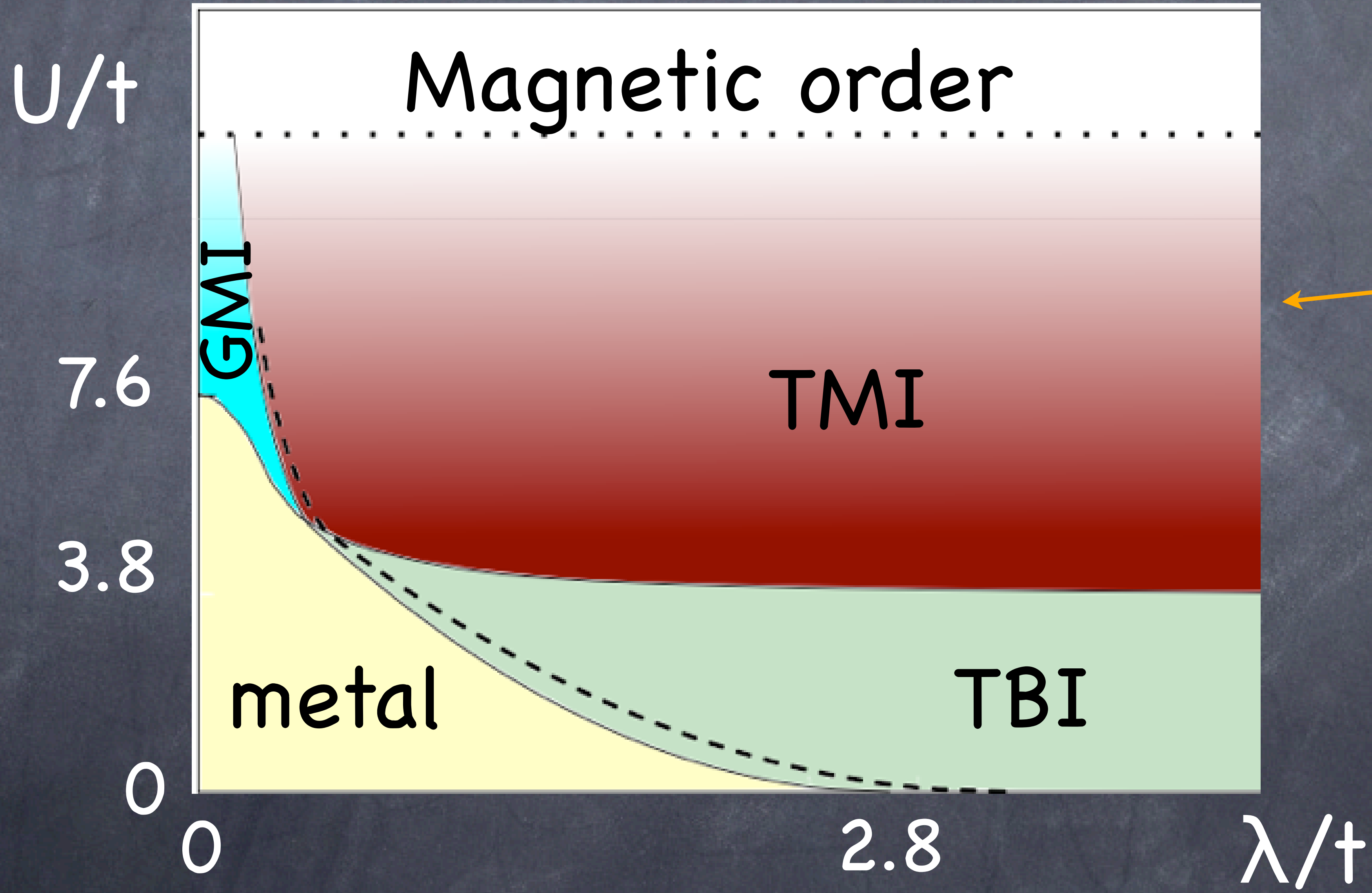


rotors gapped in MIs

Phase Diagram



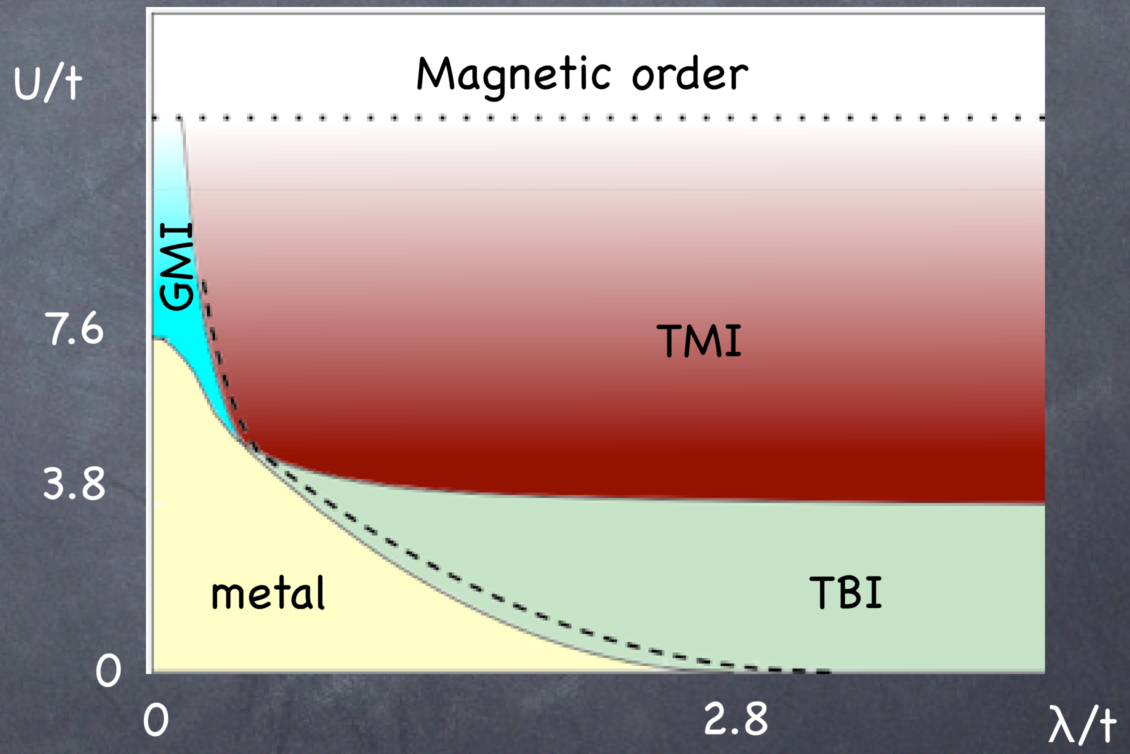
Phase Diagram



spinons
in Z_2 TBI
state

Topological Mott Insulator

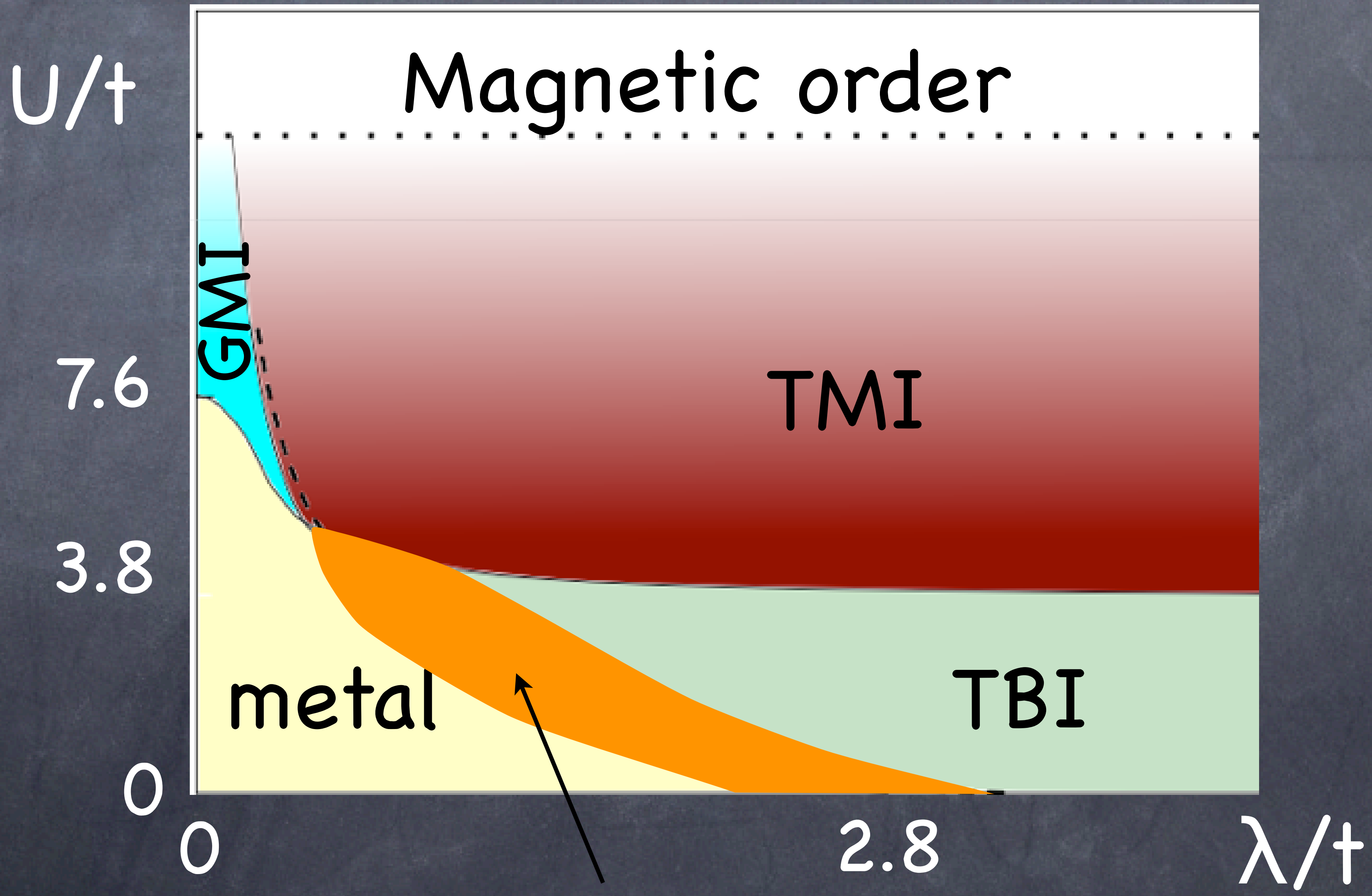
- A U(1) spin liquid
  - Gapless photon
  - Stable only in 3d
- Gapless "topological spin metal" at surface
- Magnetic monopole excitations carry spin or charge?



metal-TBI transition

- Long-range Coulomb: excitons

c.f. Halperin, Rice (1968)

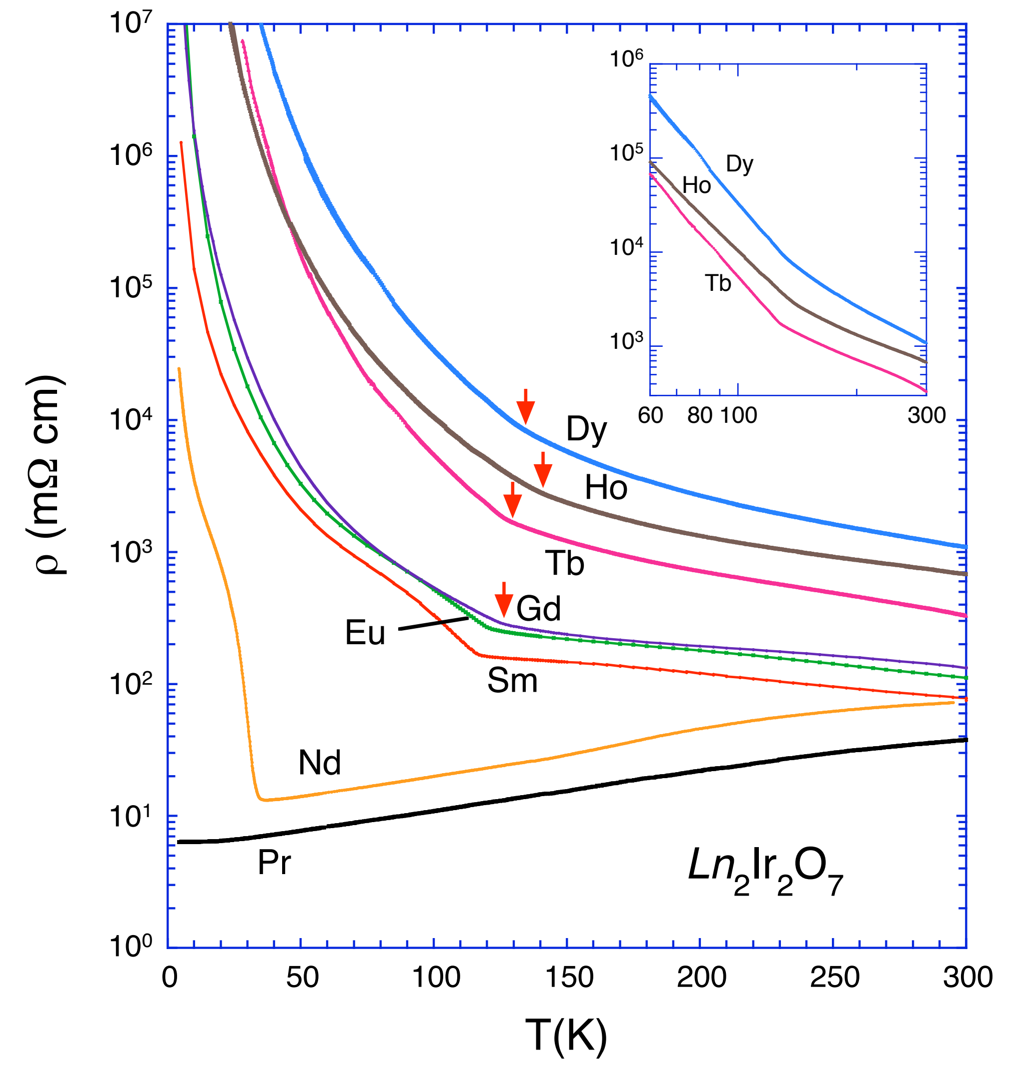


probably weakly magnetic

Back to iridates

K. Matsuhira et al, 2007

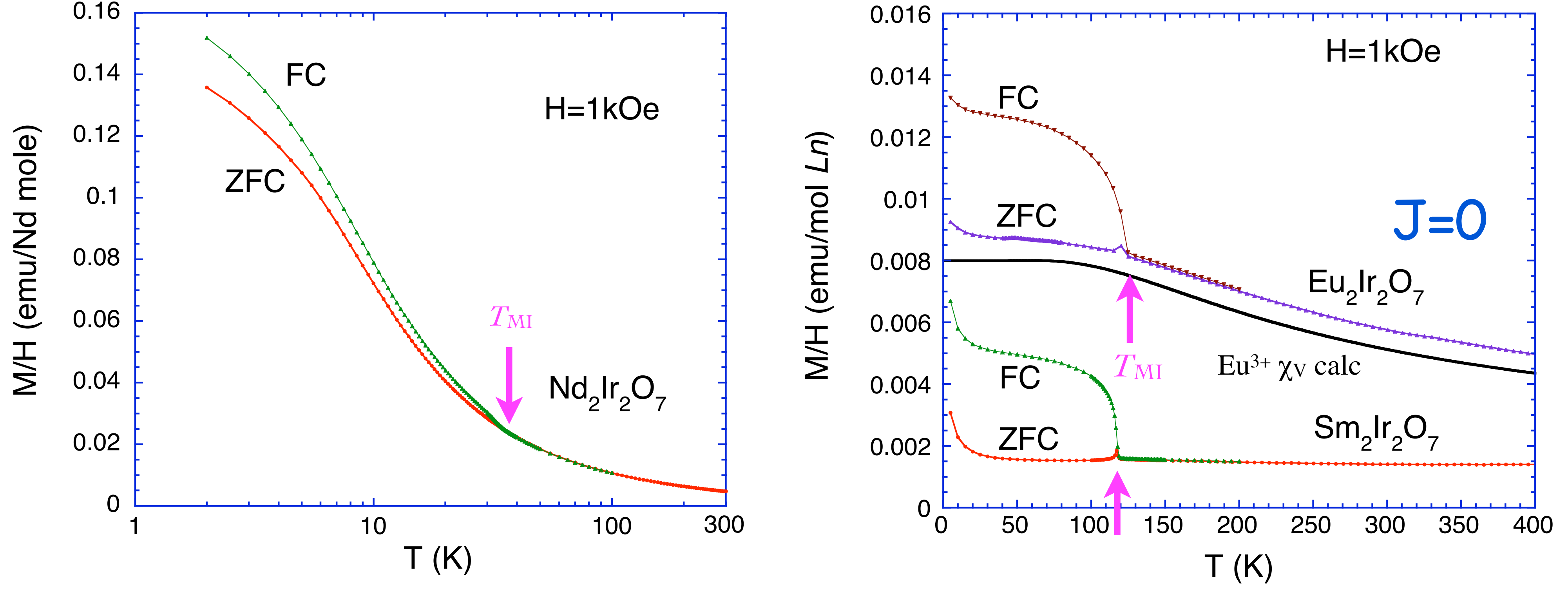
- Experiments show continuous T>0 MITs



Back to iridates

K. Matsuhira et al, 2007

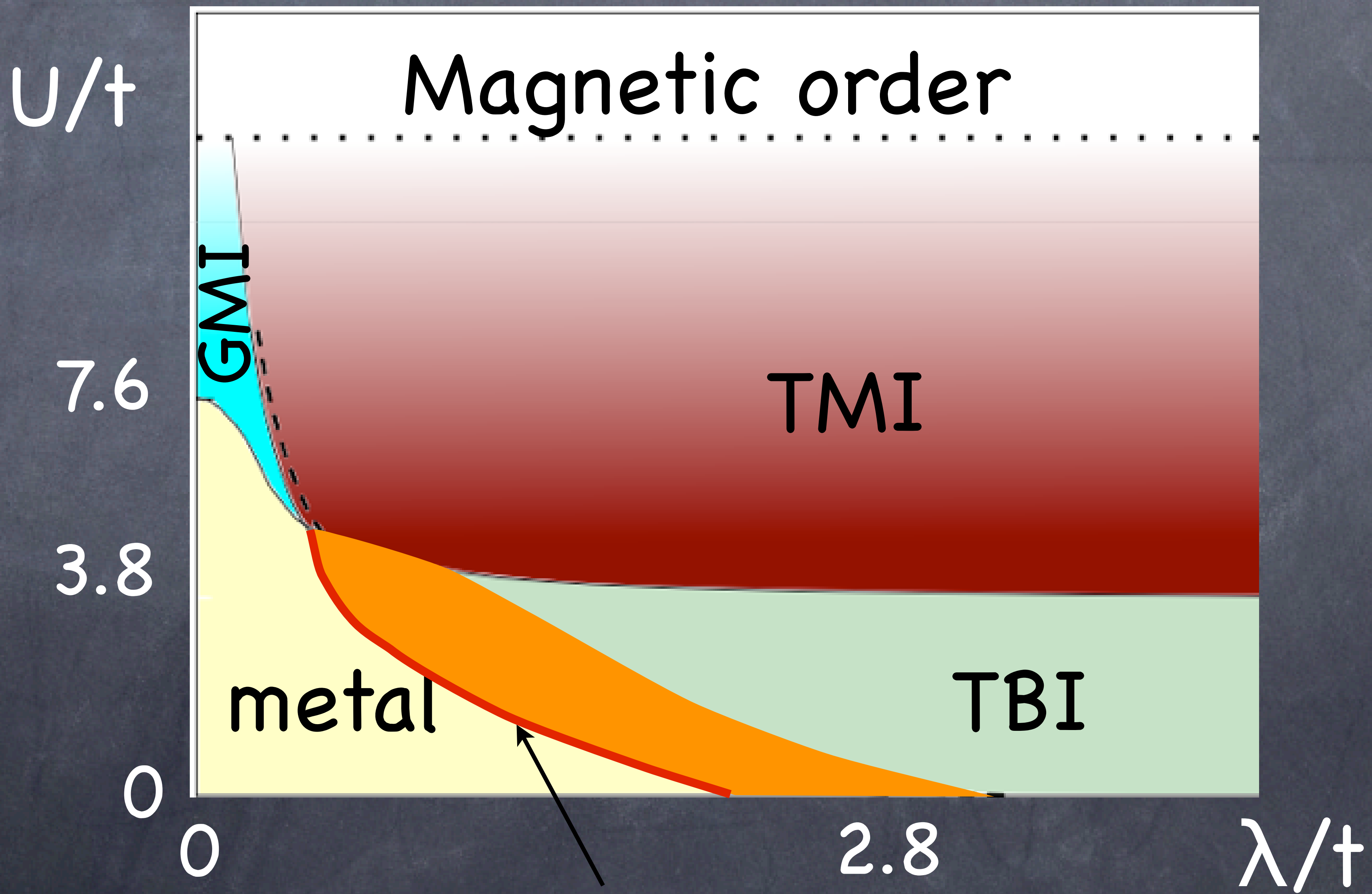
- Experiments show continuous T>0 MITs



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
- Advertisement for other group activities:
  - Double perovskites (Ba_2NaOsO_6...)
  - Various frustrated quantum magnets
  - Mott transition in TMO heterostructures

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

Mott+SO: arXiv:0908.2962