Novel magnetism in d4 spin-orbit “Mott” insulators

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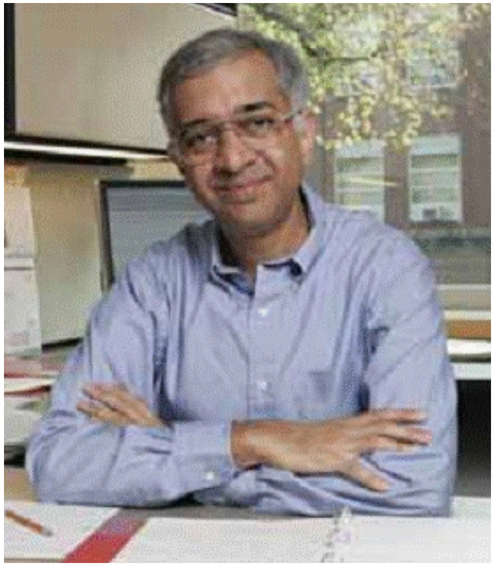
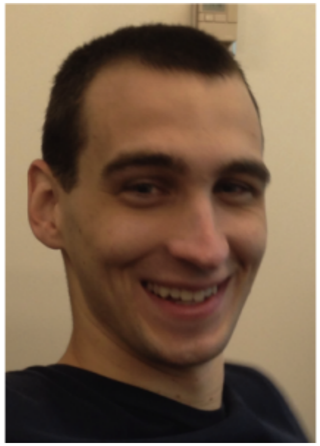


Mohit Randeria

O. Nganba Meetei, W. S. Cole, M. Randeria, N.T
PRB 91, 054412 (2015); *submitted Nov 2013*

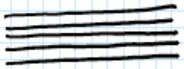


Chris Svoboda

4d & 5d oxides:

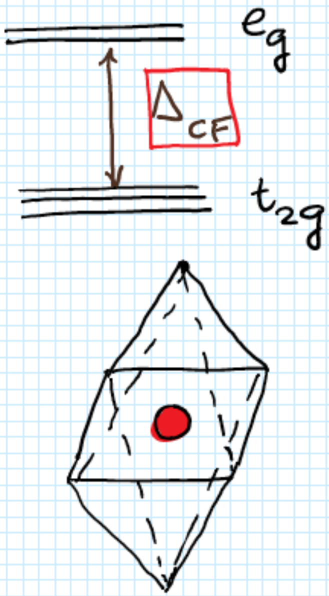
PARAMETERS

Multi orbitals:



d^5

(spherical environment)



(octahedral environment)

Atom: Coulomb

n (occupancy)

$\boxed{U}$ (intra orbital)

U' (inter orbital)

$U' \simeq U - 2\boxed{J_H}$

↑ Hund's

$\boxed{\lambda_{SO}}$ (spin-orbit coupling)

Xtal Structure

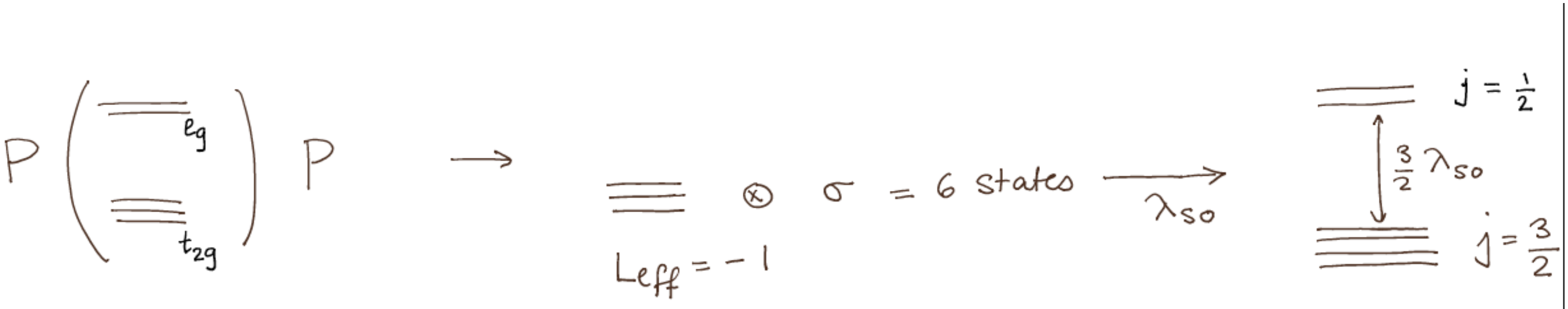
$\boxed{\overleftrightarrow{t}} \Rightarrow$ BW

Distortions of octahedra

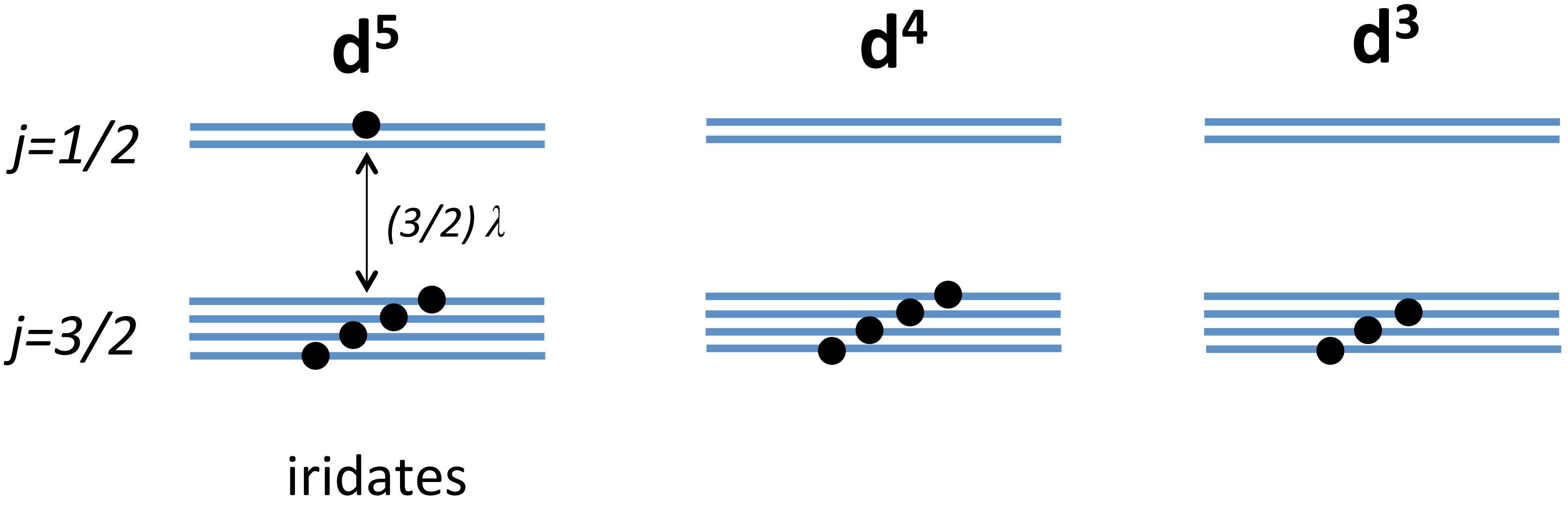
elongation, rotation, tilting

Energy Scales

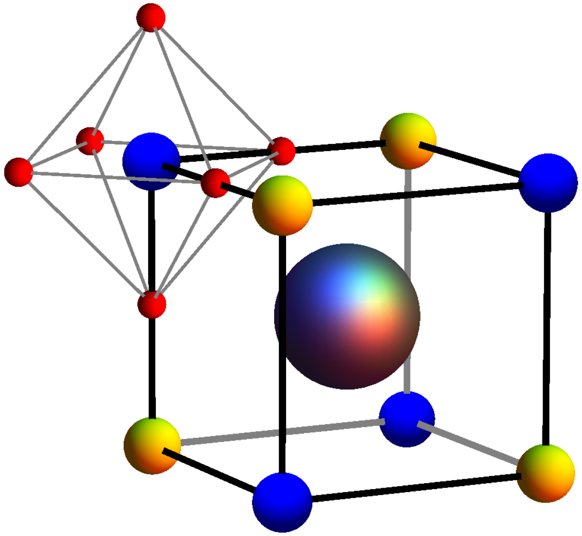
	U	J_H	Δ_{CF}	λ_{SO}	
3d	3-5 eV	0.8-0.9eV	$\Delta \lesssim J_H < U$ High Spin	0.01-0.1 eV	$U > \Delta_{CF} > \lambda_{SO}$
4d	2-3 eV	0.6-0.7 eV	$\Delta \simeq U > J_H$	0.1 -0.4 eV	$U \simeq \Delta_{CF} > \lambda_{SO}$
5d	1-2 eV	0.4 -0.5eV $\boxed{0.2\,U}$	$\Delta \simeq U > J_H$ low spin	0.4 -1 eV	$U \simeq \Delta_{CF} \simeq \lambda_{SO}$



different fillings in t2g



d1: Chen, Pereira, Balents, PRB **82**, 174440 2010
d2: Chen, Balents PRB **84**, 094420 (2011)
d3: *Theory of High Tc Ferrimagnetism in a Multiorbital Mott Insulator* Sr2CoOsO6
Meetei, Erten, Randeria, NT, Woodward PRL 110, 087203 (2013)
High antiferromagnetic transition temperature of a honeycomb compound SrRu2O6
W. Tian, C. Svoboda, M. Ochi, M. Matsuda, H. B. Cao, J.-G. Cheng, B. C. Sales, D. G. Mandrus, R. Arita, NT, and J.-Q. Yan, arXiv 1504.03642
d5: iridates



d3-d3

Sr_2CrOsO_6

Insulator
High **Tc~720K**
Net moment
Non-monotonic M(T)

New Mott criterion:

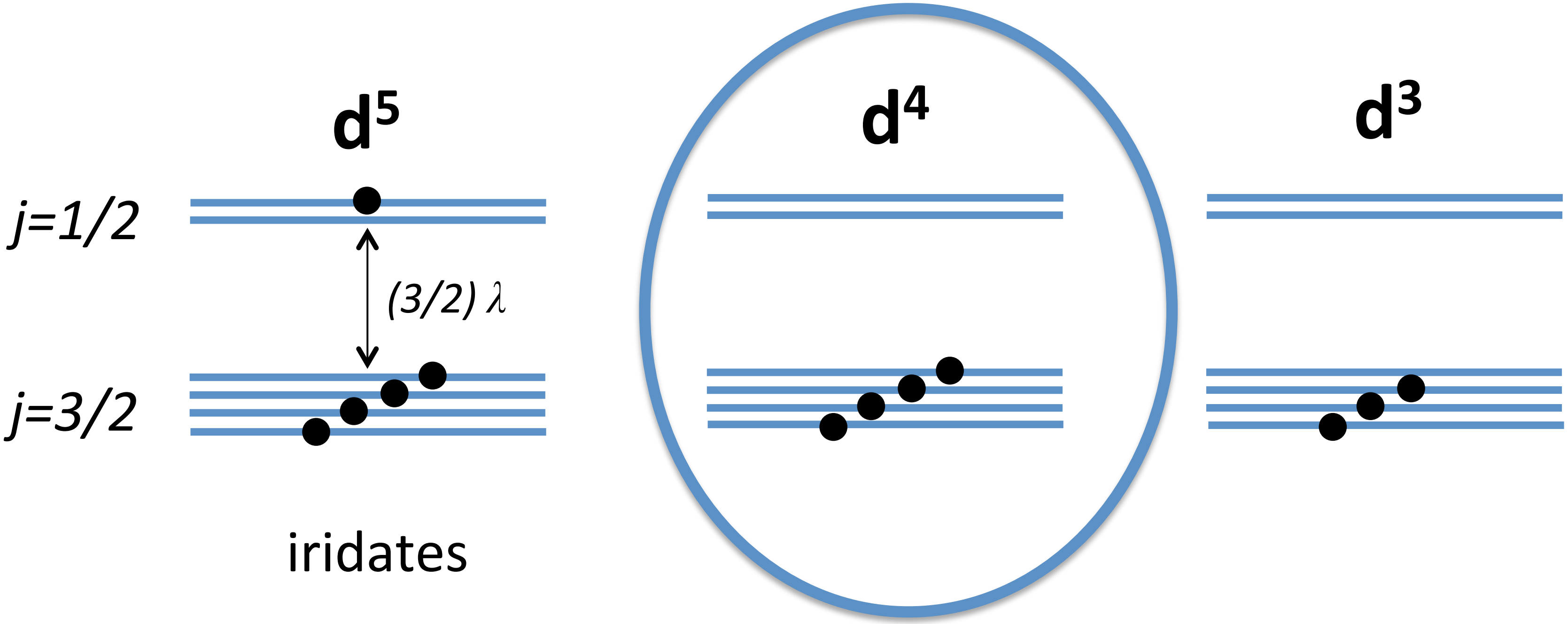
$$\sqrt{U_{Cr} \cdot U_{Os}} > 2.5\,W$$

Cr	Mn	Fe	Co
Mo	Tc	Ru	Rh
W	Re	Os	Ir

↑ U

Meetei, Erten, Randeria, NT, Woodward PRL 110, 087203 (2013)

What about other fillings?



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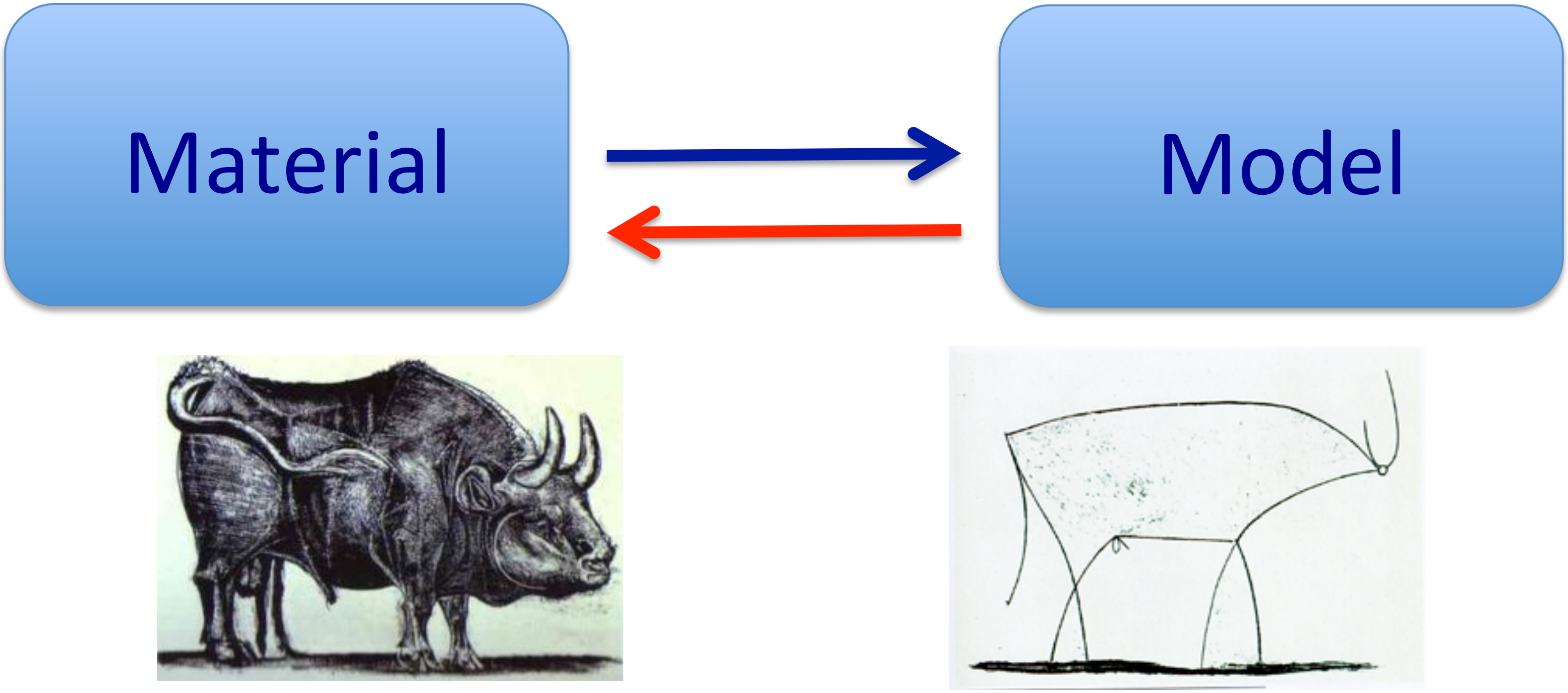


Material

Model

Outline:

(1) Puzzle of d4

(2) Insights from exact 2 site calculation

(3) Effective Hamiltonian: mean field theory

(4) Predictions for RXS

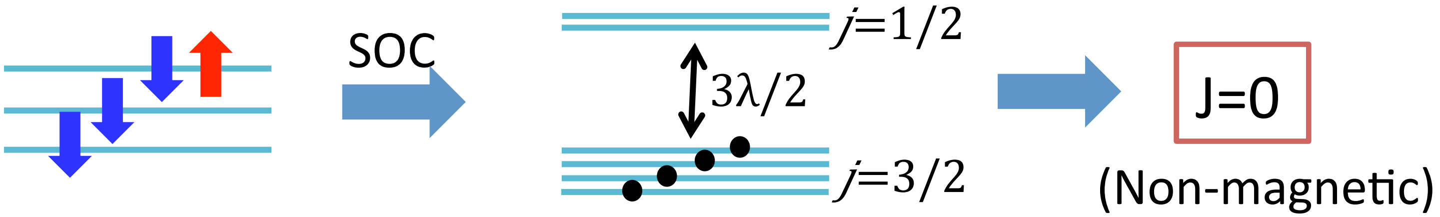
(5) Materials and Experiments

d4

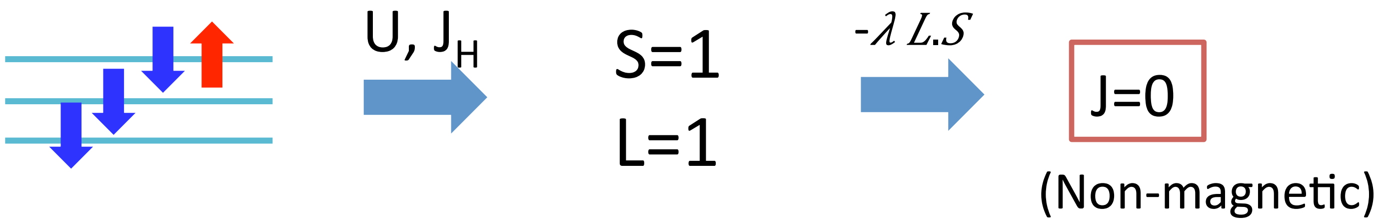
Cr	Mn	Fe	Co
Mo	Tc	Ru(4+)	Rh(5+)
W	Re(3+)	Os(4+)	Ir(5+)

d^4 systems are non-magnetic in atomic limit

$\lambda \gg U \gg t$



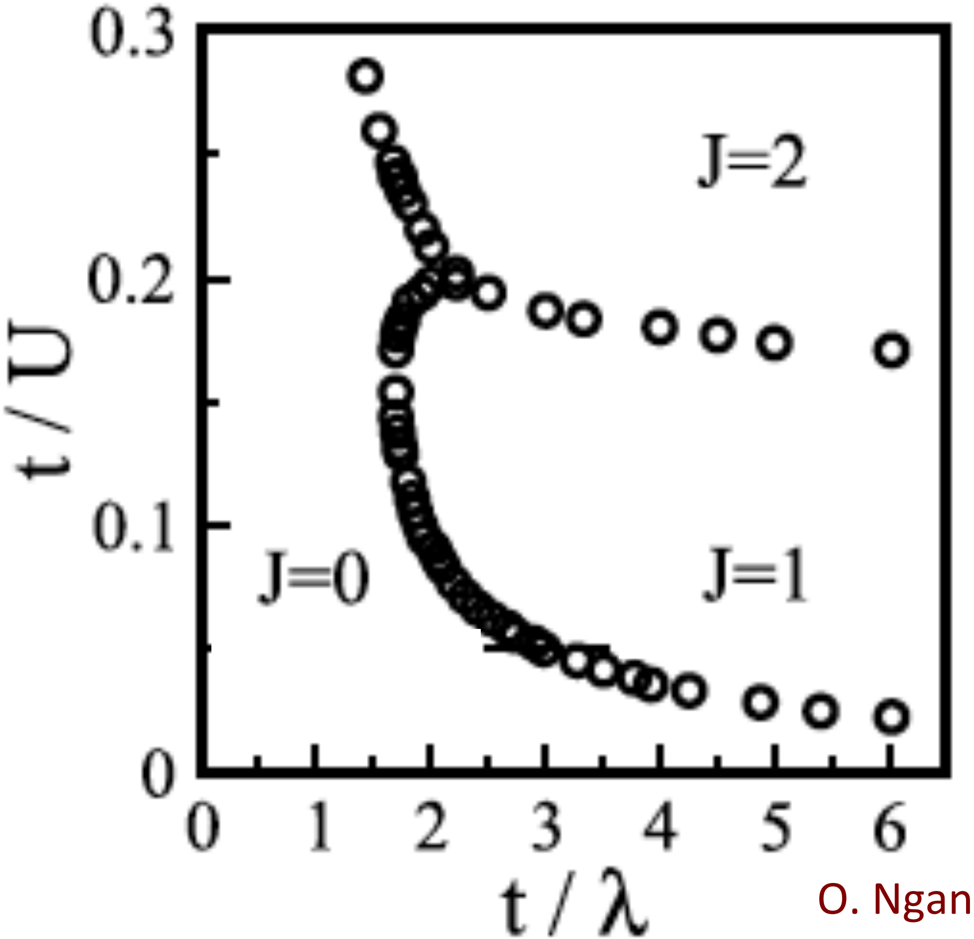
$U \gg \lambda \gg t$



Can there be any non-trivial magnetism in d^4 systems?

Hopping induced Ferromagnetism in d4 system

$$H = H_{hop} + \sum_i (H_{i,U} + H_{i,SOC})$$



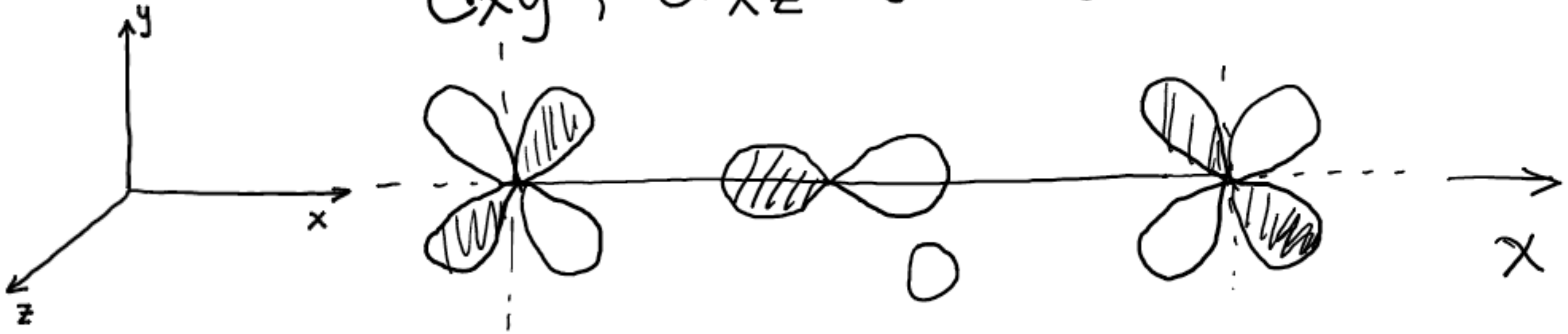
d^4: 2 sites

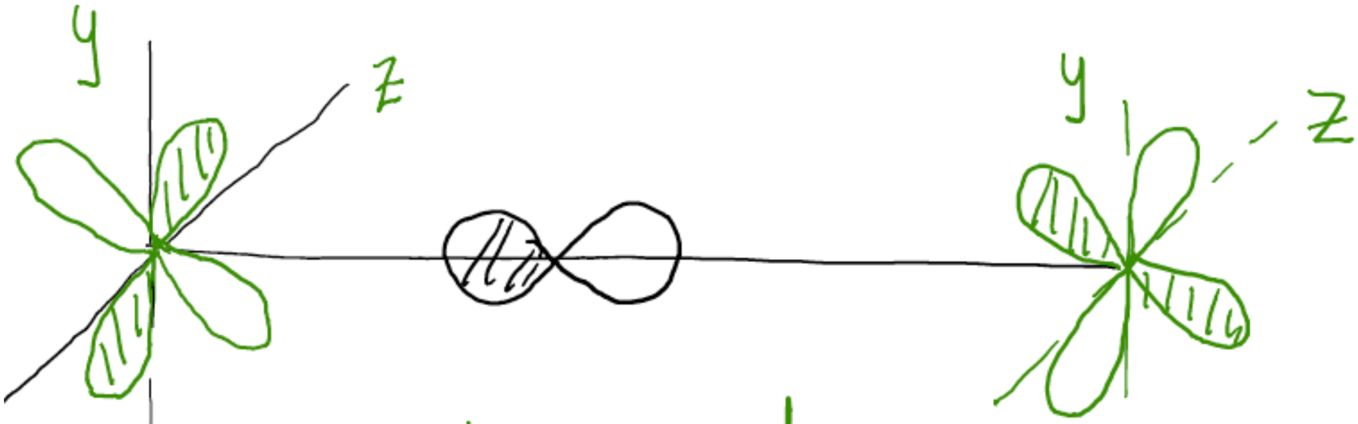
Exact diagonalization

$${}^{(6\times 2)}C_8 \approx 500$$

states

O. Nganba Meetei, W. Cole, M. Randeria, NT, PRB 91, 054412 (2015) dxy ; dxz active



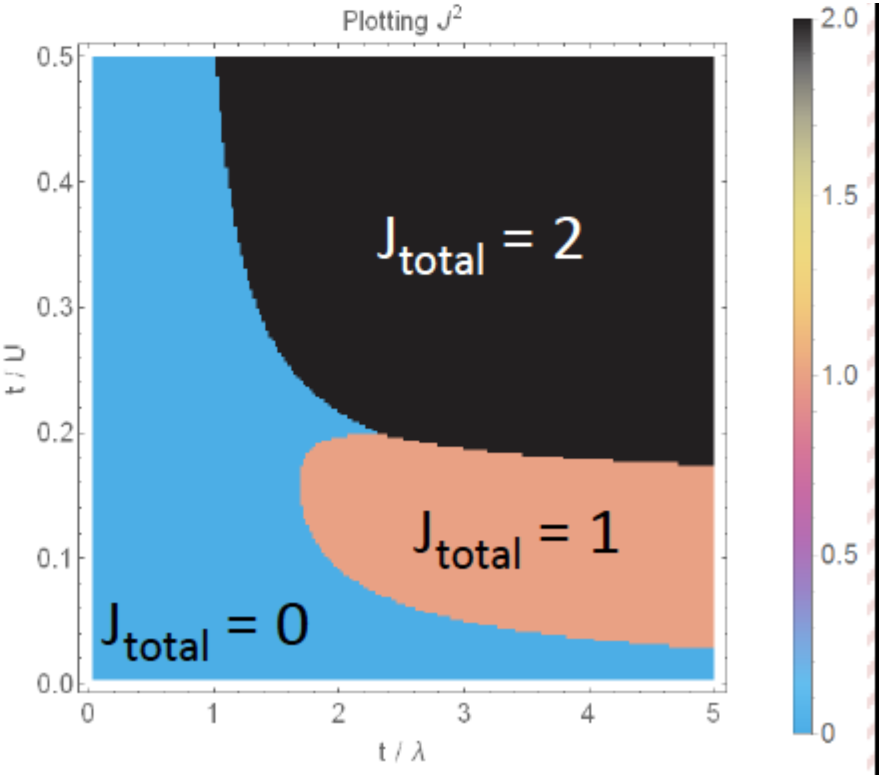


blocked

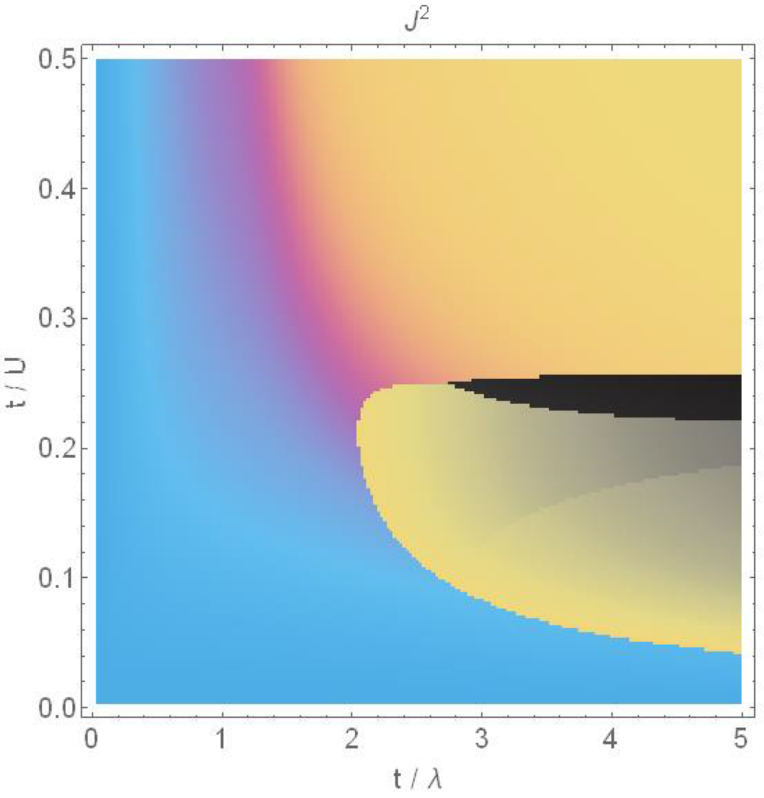
dyz

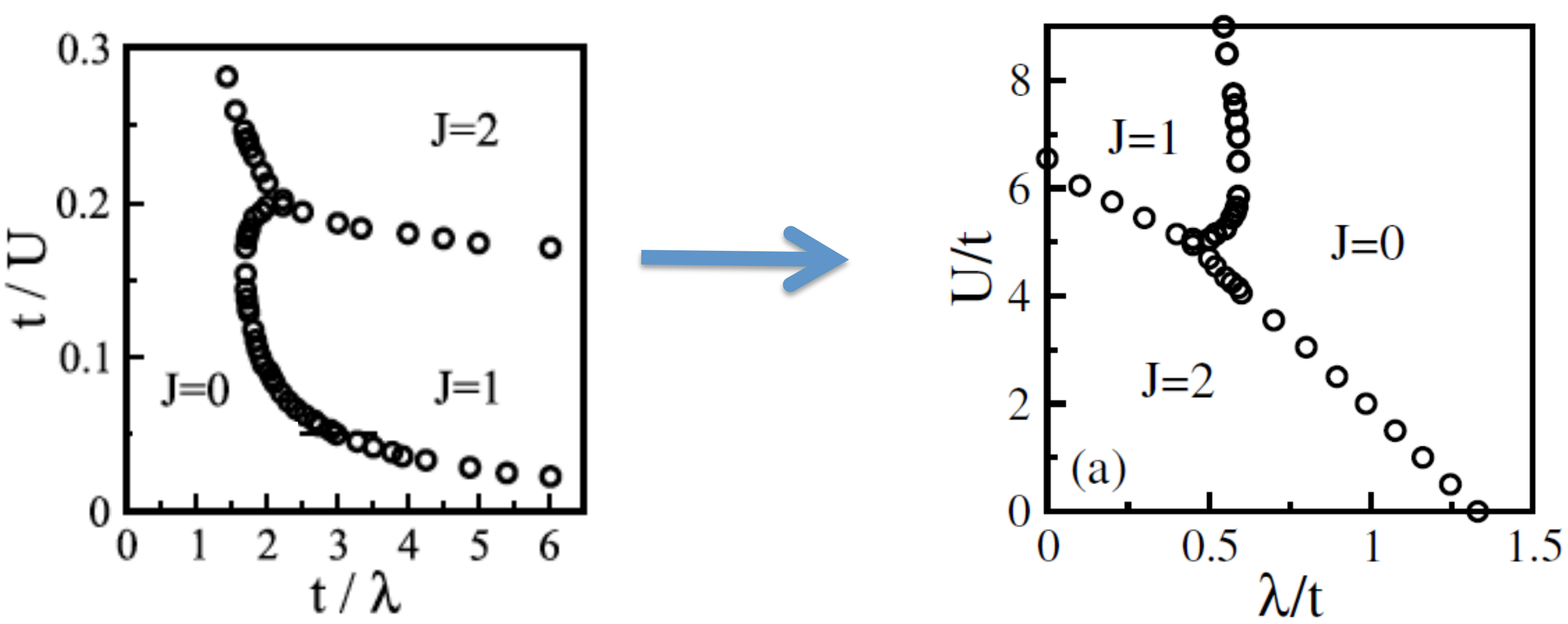
Magnetic ground states persist even with one blocked channel

Symmetric hopping
J good quantum number



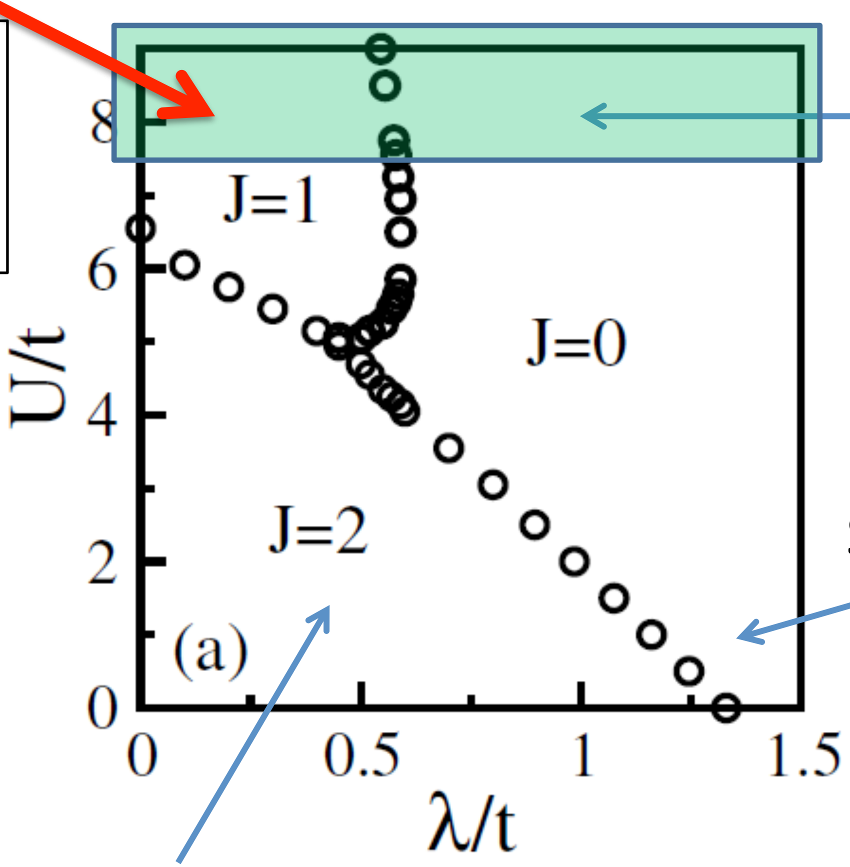
Asymmetric hopping
(one blocked channel)




0.3
0.2
0.1
0
t / U
J=2
J=0
J=1
0
1
2
3
4
5
6
t / λ
8
6
4
2
0
U/t
J=1
J=0
J=2
(a)
0
0.5
1
1.5
λ/t

Hopping induced magnetism

NOVEL
FM



Atomic picture
expected to work

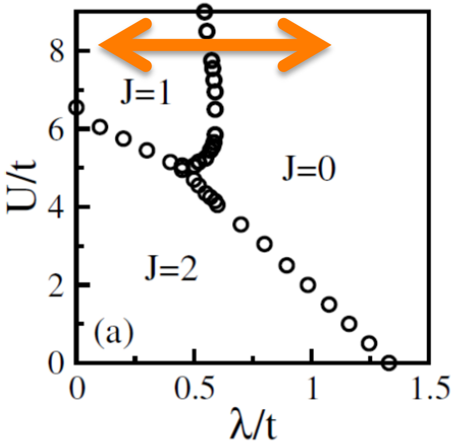
SOC Driven Band Insulator
($CaOsO_3$)
Band theory

Stoner Ferromagnet ($SrRuO_3$)
Mean field theory

Novel FM

(1) Hopping generates a local mom

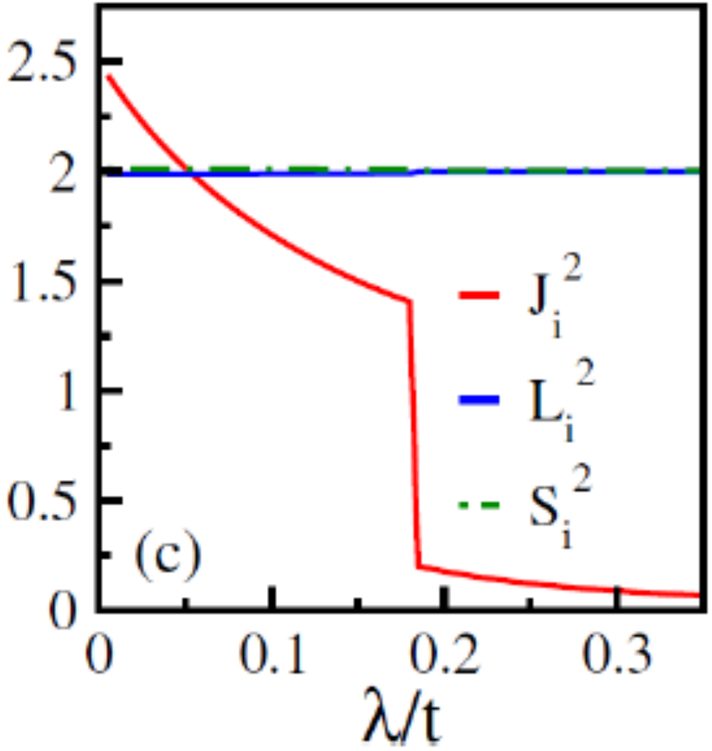
(2) Local moment is not robust



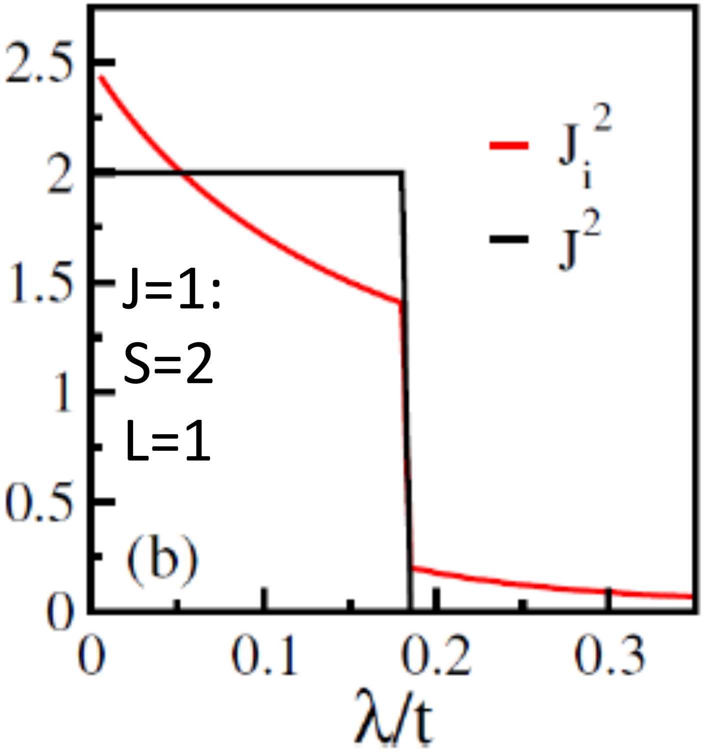
U/t=8

$$|L_i| = 1$$

$$|S_i| = 1$$



(3) Local moments are coupled ferromagnetically

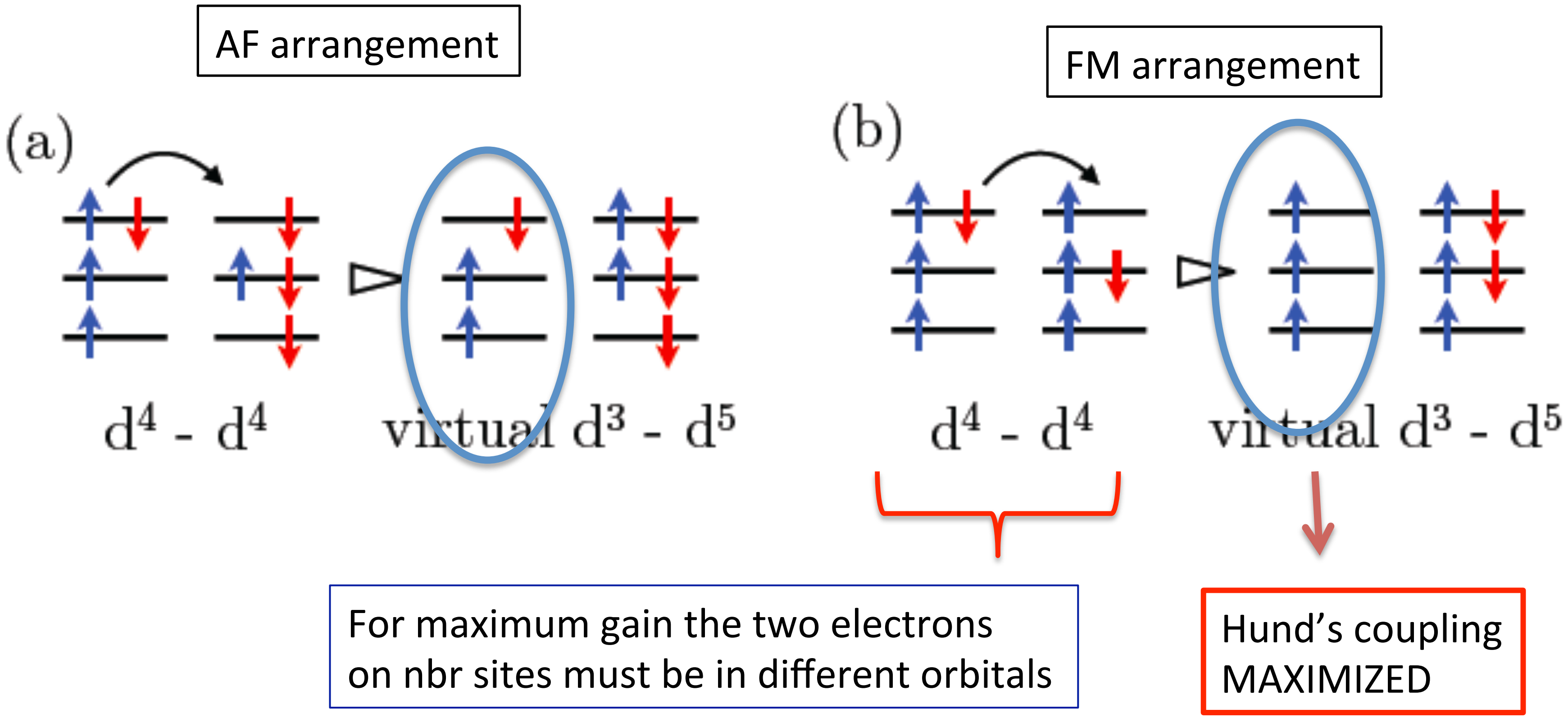


J=1:

S=2

L=1

Why *ferromagnetic* superexchange?



AF arrangement

FM arrangement

For maximum gain the two electrons on nbr sites must be in different orbitals

Hund's coupling MAXIMIZED

Perturbation Theory

$$\mathcal{H}_o = \sum \mathcal{H}_i^{at}$$

$$\mathcal{H}_i^{at} = \frac{U - 3J_H}{2} \hat{N}_i (\hat{N}_i - 1) + \frac{5}{2} J_H \hat{N}_i - 2 J_H \hat{S}_i^2 - \frac{1}{2} J_H \hat{L}_i^2$$

Ground state : $\hat{N}_i = 4$ $\hat{L}_i = 1$ $\hat{S}_i = 1$

$$E_0 = 12U - 26 J_H$$

$$\mathcal{H}_{hop} = -t \sum_{\alpha,\sigma} c^{\dagger}_{1\alpha\sigma} c_{2\alpha\sigma} + h.c.$$

FM pathway

d^4 d^4

$S_1 = 1$ $S_2 = 1$

$L_1 = 1$ $L_2 = 1$

$S = 2, 1, 0$

$L = 2, 1, 0$

d^3 d^5

$S_1 = 3/2$ $S_2 = \frac{1}{2}$

$L_1 = 0$ $L_2 = 1$

$S = \boxed{2}, 1$

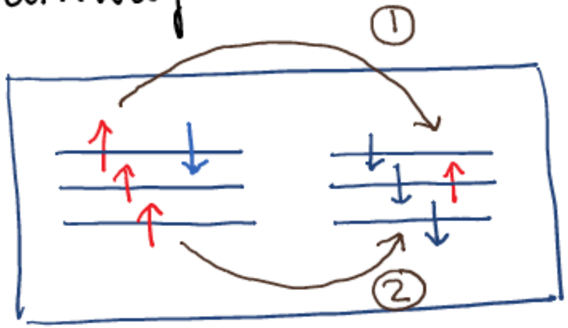
$L = \boxed{1}$

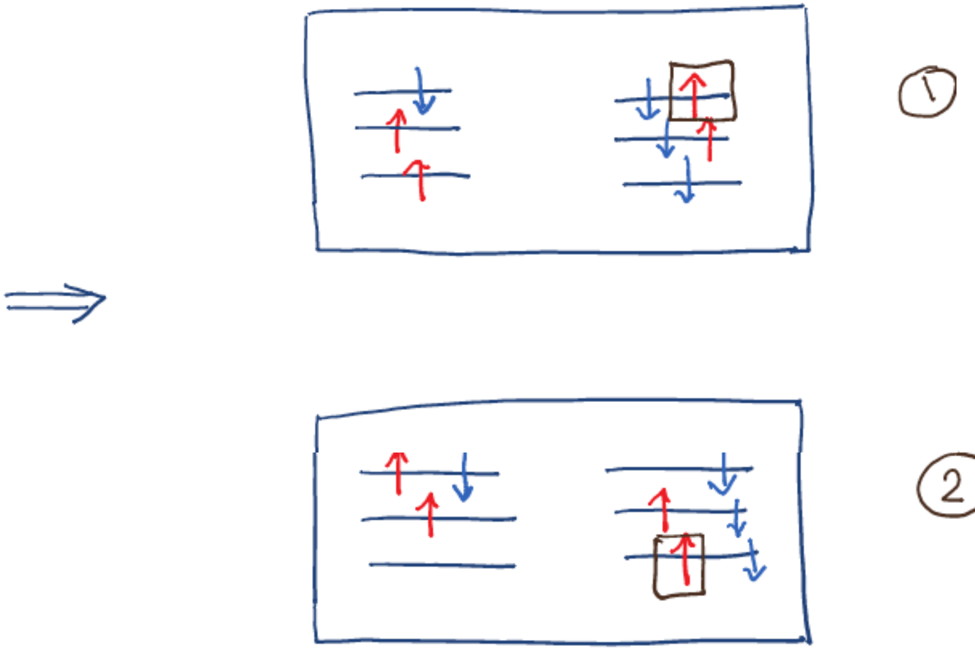
$$E_1 = 13U - 29 J_H$$

$$\Rightarrow \Delta E = E_1 - E_0 = U - 3J_H$$

$$\boxed{J_{FM} = -\frac{t^2}{U - 3J_H}}$$

AF Pathways :



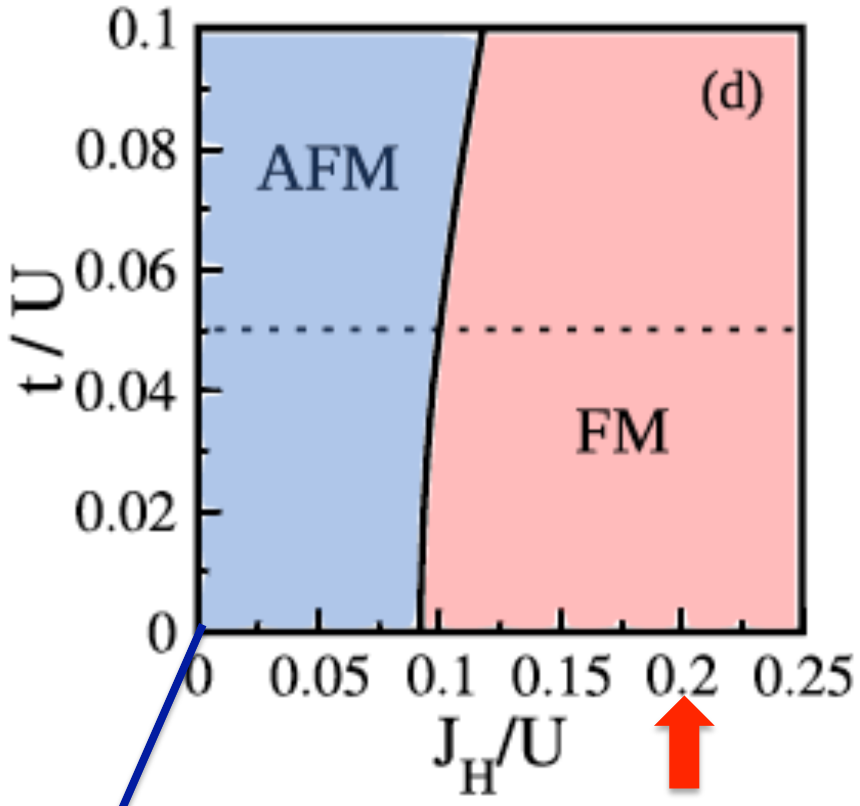


$$\boxed{J_{AF} = \frac{t^2}{3}\,\frac{1}{U-3J_H} + \frac{7}{6}\,\frac{t^2}{U} + \frac{t^2}{2}\,\frac{1}{U+2J_H}}$$

$J_F = J_{AF}$ when $\boxed{\frac{J_H}{U} \sim 0.15}$ estimate

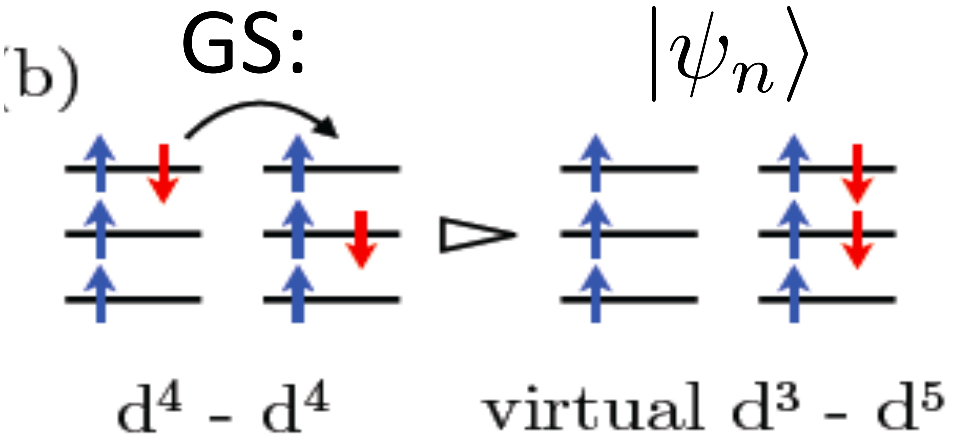
Role of Hund's coupling in determining magnetic ground state

- Hopping with one blocked channel
- Exact diagonalization results



Typical value for 4d/5d oxides
Khomskii book

AF superexchange for J_H=0
G. Khaliullin PRL 111, 197201 (2013)



GS: $|\psi_n\rangle$

$L_i = 1 \quad L_i = 1 \qquad L_i = 0 \qquad L_i = 1$

$S_i = 1 \quad S_i = 1 \qquad S_i = 3/2 \qquad S_i = 1/2$

$$\tilde{H}' = H_{hop} \left[\sum_n \frac{|\psi_n\rangle\langle\psi_n|}{E_G - E_n} \right] H_{hop}$$

$$\boxed{\tilde{H} \approx -J_{FM} \mathbf{S_1} \cdot \mathbf{S_2} \mathcal{P}(\mathbf{L_1} + \mathbf{L_2} = 1)}$$

Orbitally entangled Ferromagnet

S=2 antialigned with L=1

Effective Hamiltonian: Superexchange + SOC

$$H_{eff} = -J_{FM} \sum_{\langle ij \rangle} S_i . S_j \mathcal{P}(L_i + L_j = 1) + \lambda \sum_i L_i . S_i$$

FM superexchange

$S_1 + S_2 = 2$

and

$L_1 + L_2 = 1$

SOC

$L_1 + S_1 = 0$

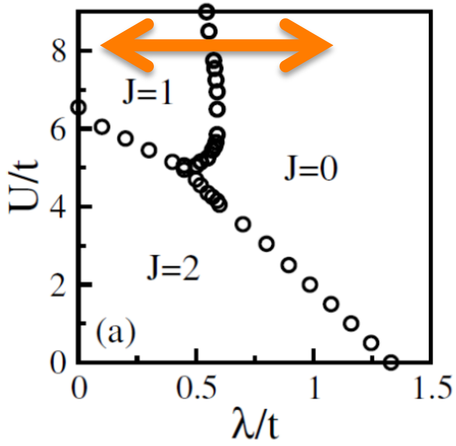
and

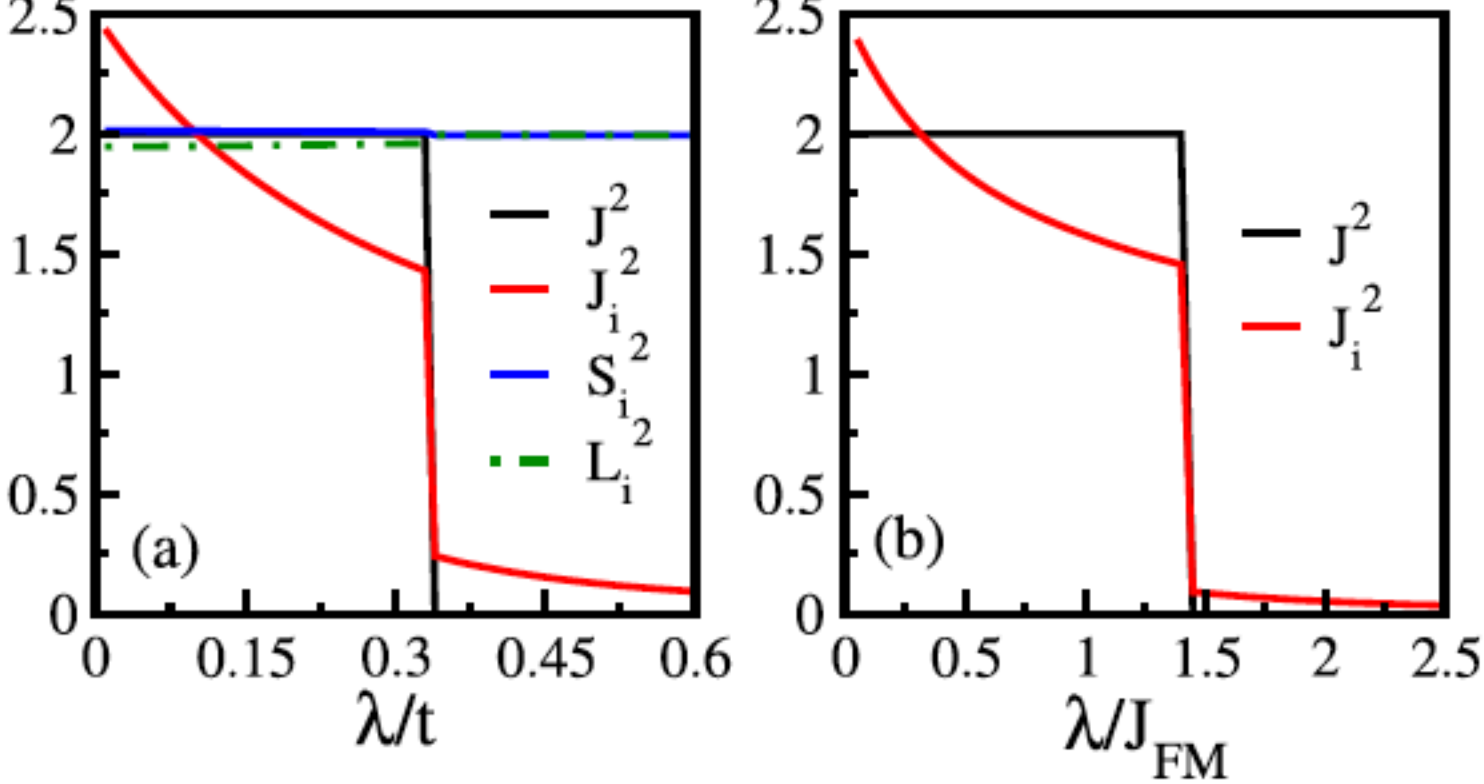
$L_2 + S_2 = 0$

Competition drives a phase transition

$$\sqrt{\lambda U_{eff}} \sim t$$

Comparison of effective model with full H





full H

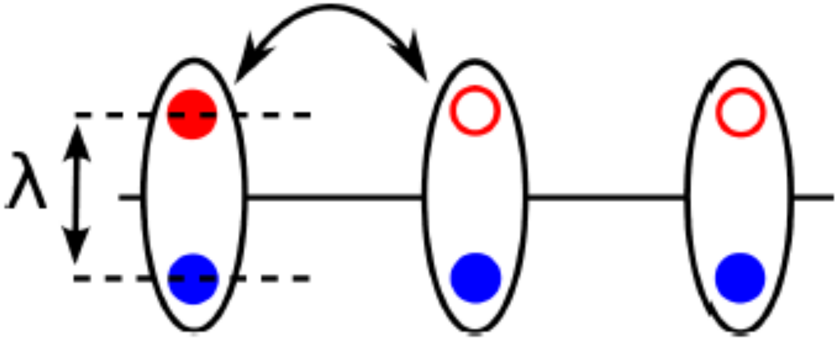
H_{eff}

Mean-field theory for effective Hamiltonian

Locally L=1 + S=1 → J=0, 1 and 2 (Ignore high energy J=2)

$$
\begin{aligned}
S_i^\alpha &= -\sqrt{\frac{2}{3}}\left(T^\dagger_{i\alpha}s_i + s_i^\dagger T_{i\alpha}\right) - \frac{i}{2}\epsilon_{\alpha\beta\gamma}T^\dagger_{i\beta}T_{i\gamma} \\
L_i^\alpha &= \sqrt{\frac{2}{3}}\left(T^\dagger_{i\alpha}s_i + s_i^\dagger T_{i\alpha}\right) - \frac{i}{2}\epsilon_{\alpha\beta\gamma}T^\dagger_{i\beta}T_{i\gamma}
\end{aligned}
$$

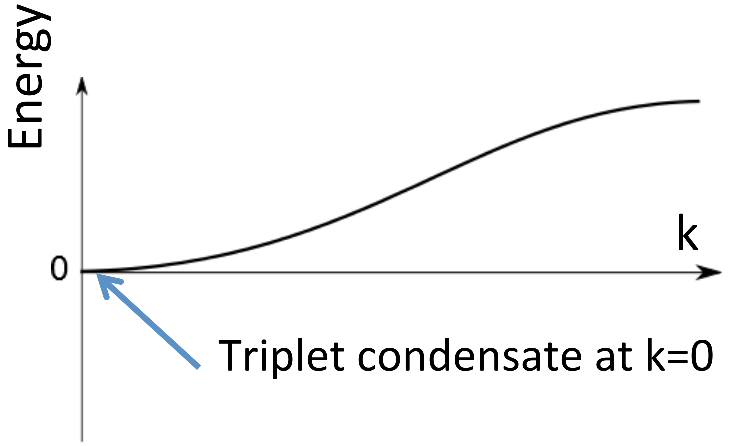
α = x,y,z



Sachdev, Bhatt PRB 41, 9323 (1989)

Mean-field theory for effective Hamiltonian

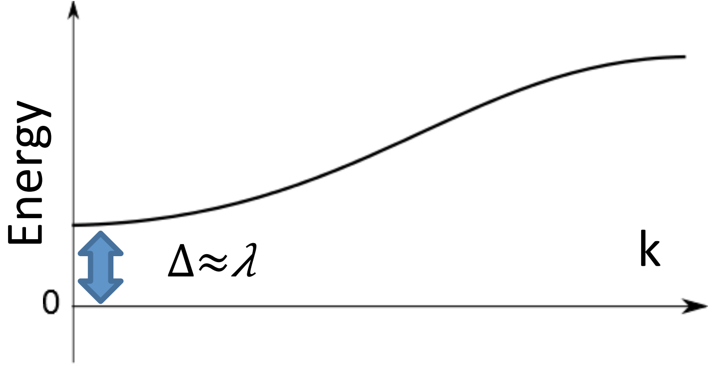
$J_{FM}/\lambda = (J_{FM}/\lambda)_c$



Triplet condensate at k=0

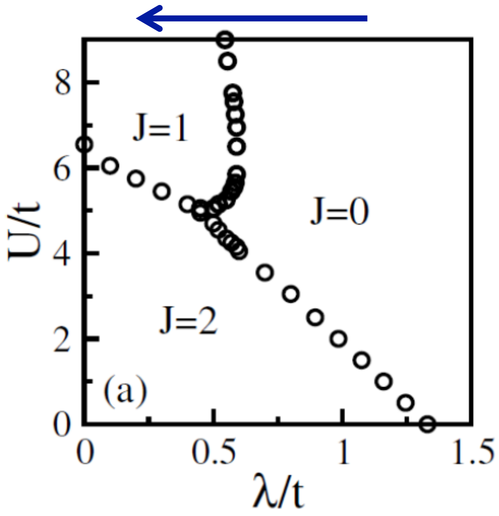
- Triplet gap closes
- Triplet condensate forms

$J_{FM}/\lambda < (J_{FM}/\lambda)_c$



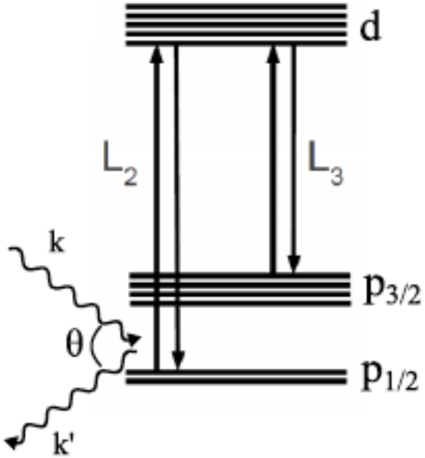
- Singlet condensate
- Gapped triplet band

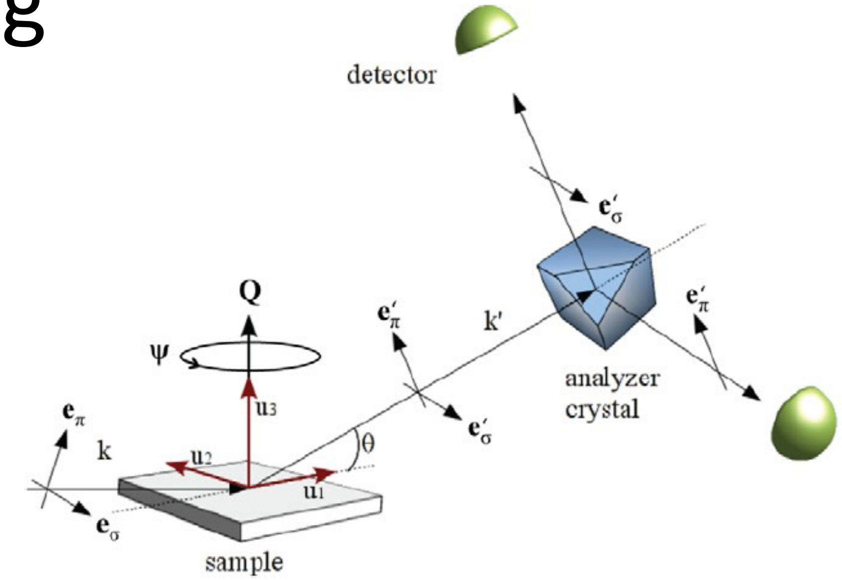
J_{FM}/λ



See also G. Khaliullin
PRL 111, 197201 (2013)

Resonant X-ray Scattering





e (e') polarization of incoming (outgoing) photon

J. Fink, E. Schierle, E. Weschke, and J. Geck, Rep. Prog. Phys. 76, 056502 (2013)

Free ion approximation (usually good for Mott insulators)

$$\Delta f(\omega) \propto \sum_n \frac{\langle \Psi_G|(\mathbf{e}'.\mathbf{D})^\dagger|\psi_n\rangle\langle\psi_n|\mathbf{e}.\mathbf{D}|\Psi_G\rangle}{E_n - E_G - \hbar\omega - i\Gamma}$$

When effect of neighboring sites are strong

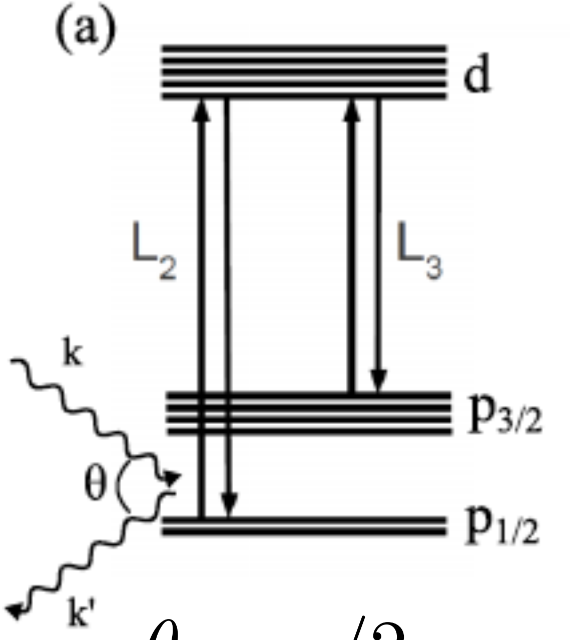
$$\Delta f(\omega) \propto Tr\left[\rho \sum_n \frac{(\mathbf{e}'.\mathbf{D})^\dagger|\psi_n\rangle\langle\psi_n|\mathbf{e}.\mathbf{D}}{E_n - E_G - \hbar\omega - i\Gamma}\right]$$

$$\vec{e}\cdot\vec{D} \approx \vec{e}\cdot\hat{\vec{r}} = \sum_{\alpha\beta\sigma} \vec{e}\cdot\langle d_\alpha|\hat{\vec{r}}|p_\beta\rangle d^\dagger_{\alpha\sigma} p_{\beta\sigma} + \mathrm{H.c.}$$

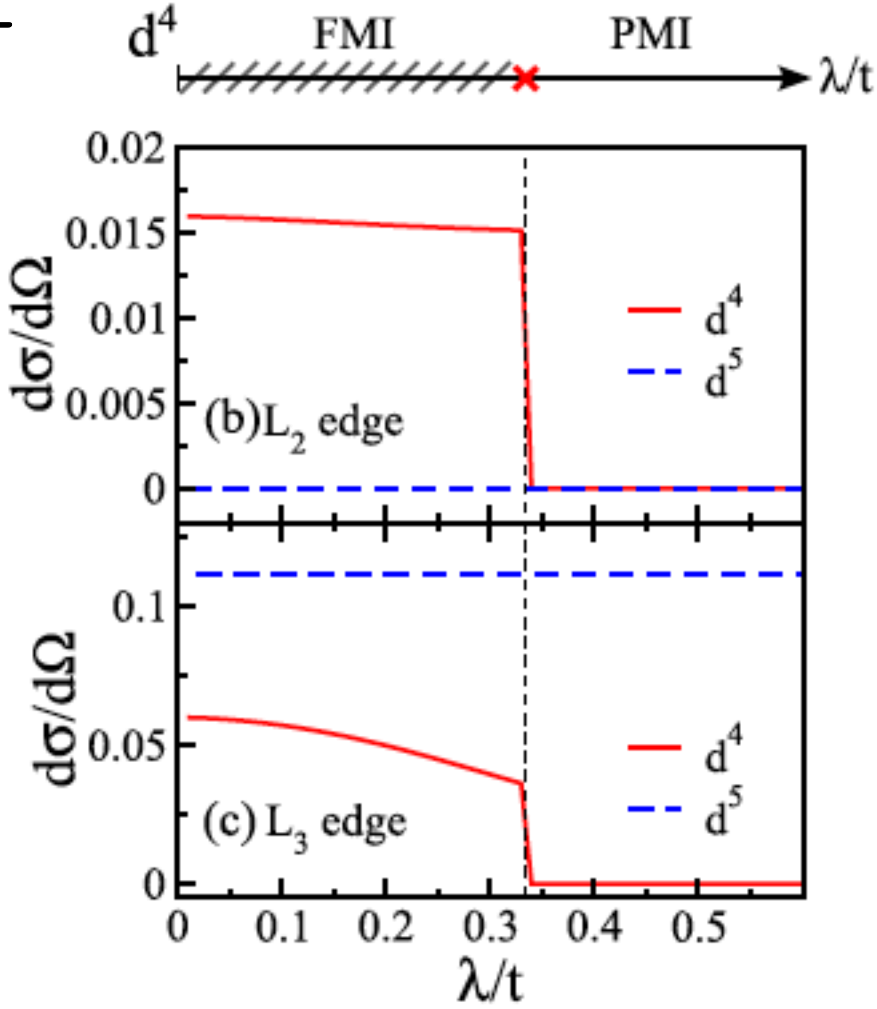
Resonant Xray scattering in d4

Magnetic
scattering
cross-section

$\sigma - \pi$



$\theta = \pi/2$



Conclusions: 4d/5d oxides

materials with 4 electrons in d-shell can be magnetic

→ Going beyond Ir(4+)

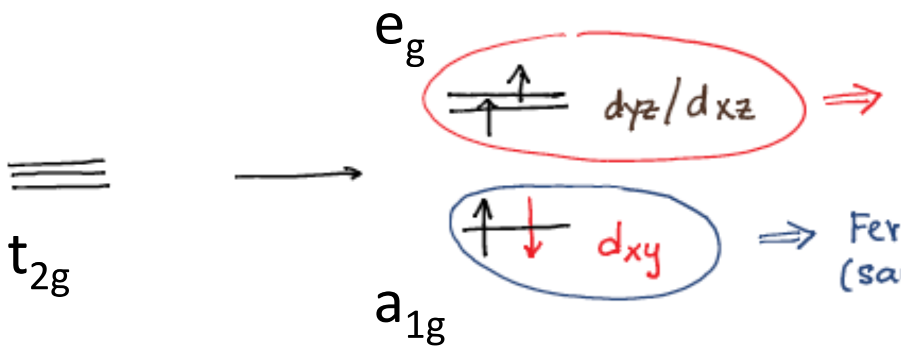
- Single atom with d4 is non-magnetic
- Hopping of electrons between atoms
  - -- generates the local moment
  - -- dictates the nature of ordering
- For typical values of $J_H/U \sim 0.2$
  (a) Ferromagnetic Ordering
  S_{total} maximized; L_{total} projected to intermediate value

  (b) Distortions F → AF

  (c) If $J_H/U < 0.1$ can get AF even without distortion

New paradigm
for magnetism

Distortion



half filled band leads to AF Mott Insulator

Ferro-orbital ordering (same orbital on all atoms)

Materials:

TM ions (d4): Ru^{4+}, Os^{4+}, Ir^{5+}

xtal structures:

- pyrochlore: $Y_2Os_2O_7$
- double perovskite: Sr_2MIrO_6, La_2MRuO_6
- layered perovskite: Ca_2RuO_4
- honeycomb: A_2RuO_3

strongly insulating and magnetic

Ca_2RuO_4 Optics + LDA/U J. H. Jung, Z. Fang, J. P. He, Y. Kaneko, Y. Okimoto, Y. Tokura, PRL 91, 056403 **(2003)**

K-edge RIXS + XAS gives Ru λso~200 meV cf Ir λso~400 meV

C. G. Fatuzzo, M. Dantz, S. Fatale, P. Olalde-Velasco, N. E. Shaik, B. Dalla Piazza, S. Toth, J. Pelliciari, R. Fittipaldi, A. Vecchione, N. Kikugawa, J. S. Brooks, H. M. Rønnow, M. Grioni, Ch. R¨uegg, T. Schmitt, and J. Chang, PRB 91, 155104 **(2015)**

Sr_2MIrO_6

[M=Mg, Ca, Sc, Ti, Ni, Fe, Zn, In, Y]

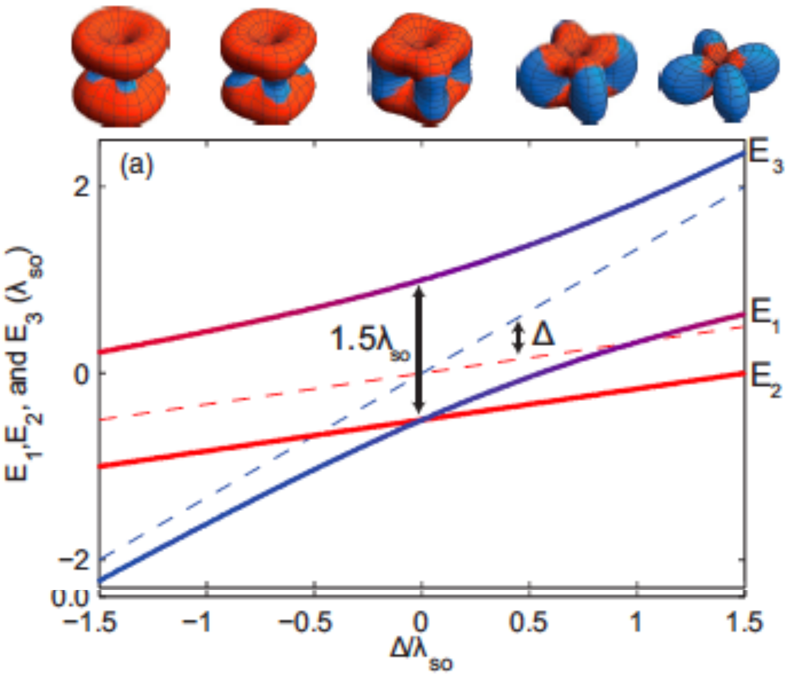
XAS+XMCD M. A. Laguna-Marco, P. Kayser, J. A. Alonso, M. J. Mart´ınez-Lope, M. van Veenendaal, Y. Choi, and D. Haskel, PRB 91, 214433 (2015)

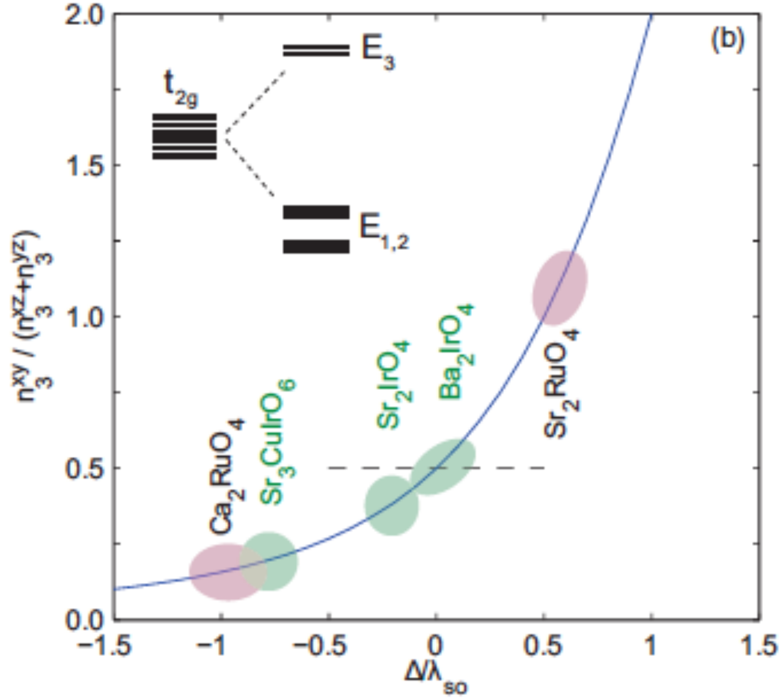
G. Cao, T. F. Qi, L. Li, J. Terzic, S. J. Yuan, L. E. DeLong, G. Murthy, and R. K. Kaul, PRL 112, 056402 (2014);

La_2MRuO_6 Zhiying Wang, J.-Q. Yan and collaborators

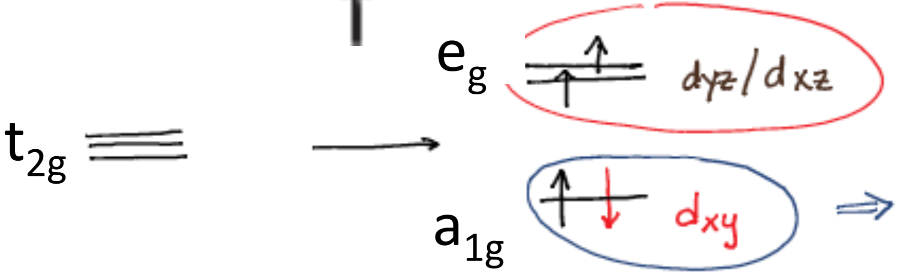
$Y_2Os_2O_7$ J.-Q. Yan and collaborators

A_2RuO_3 J. C. Wang, J. Terzic, T. F. Qi, Feng Ye, S. J. Yuan, S. Aswartham, S. V. Streltsov, D. I. Khomskii, R. K. Kaul, and G. Cao, PRB 90, 161110(R) (2014)



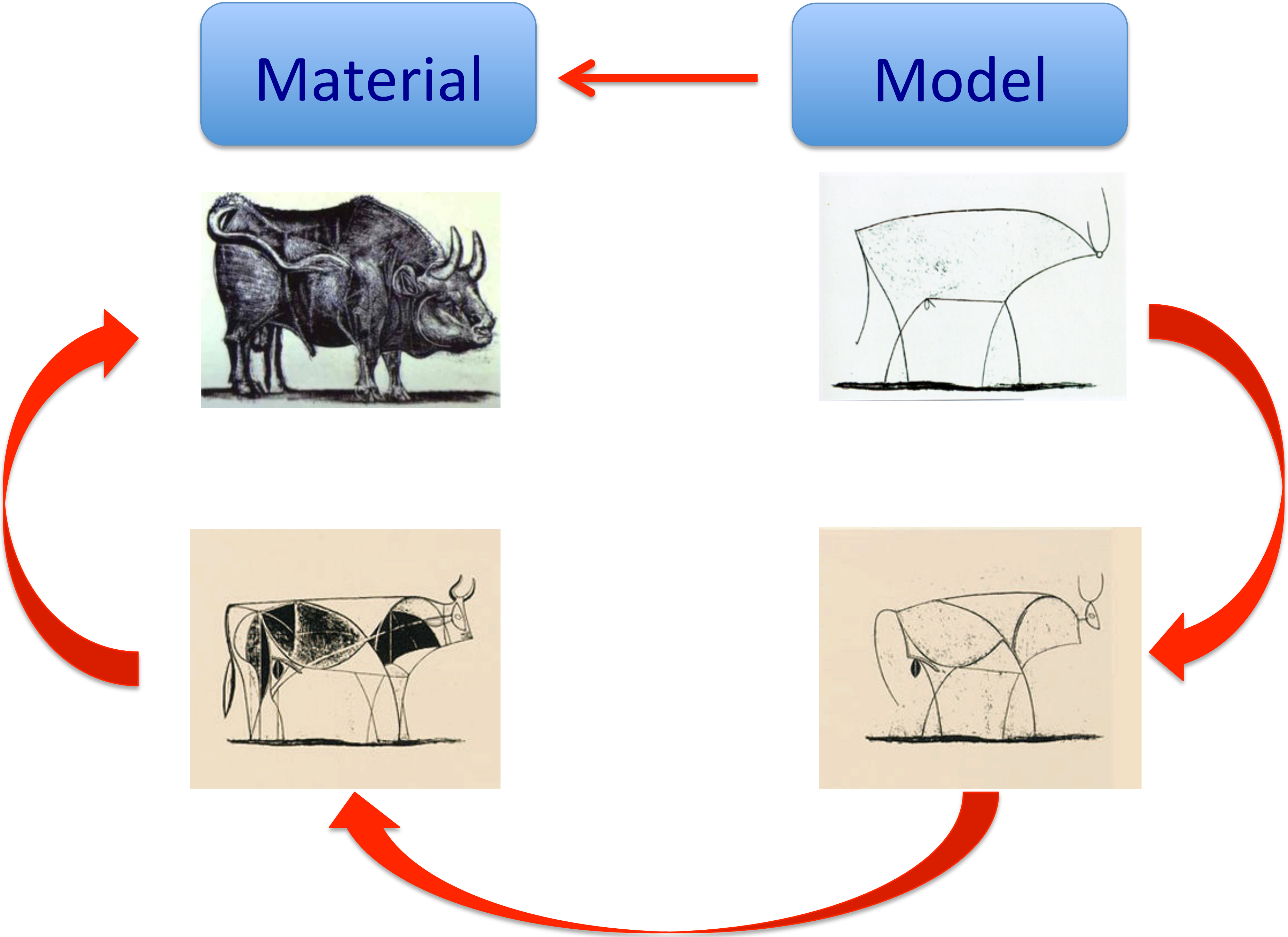


Fantozzo et al PRB 91, 155104 **(2015)**



⇒ half filled band leads to AF Mott Insulator

⇒ Ferro-orbital ordering (same orbital on all atoms)



Material

Model

Prediction:
Novel orbitally entangled ferromagnetism in d4 materials

Next steps….Develop theory for

1. Different d-O-d bond angles from different xtal structures
2. Distortion; pressure tuning
3. XMCD and XAS