RIXS for 5d oxides in “atomic limit” Mott insulators

From Atomic Lego Blocks to Frustrated Magnets and Chern Insulators

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IXS 2019, Stonybrook, 24 June 2019

UNIVERSITY OF TORONTO

NSERC CRSNG

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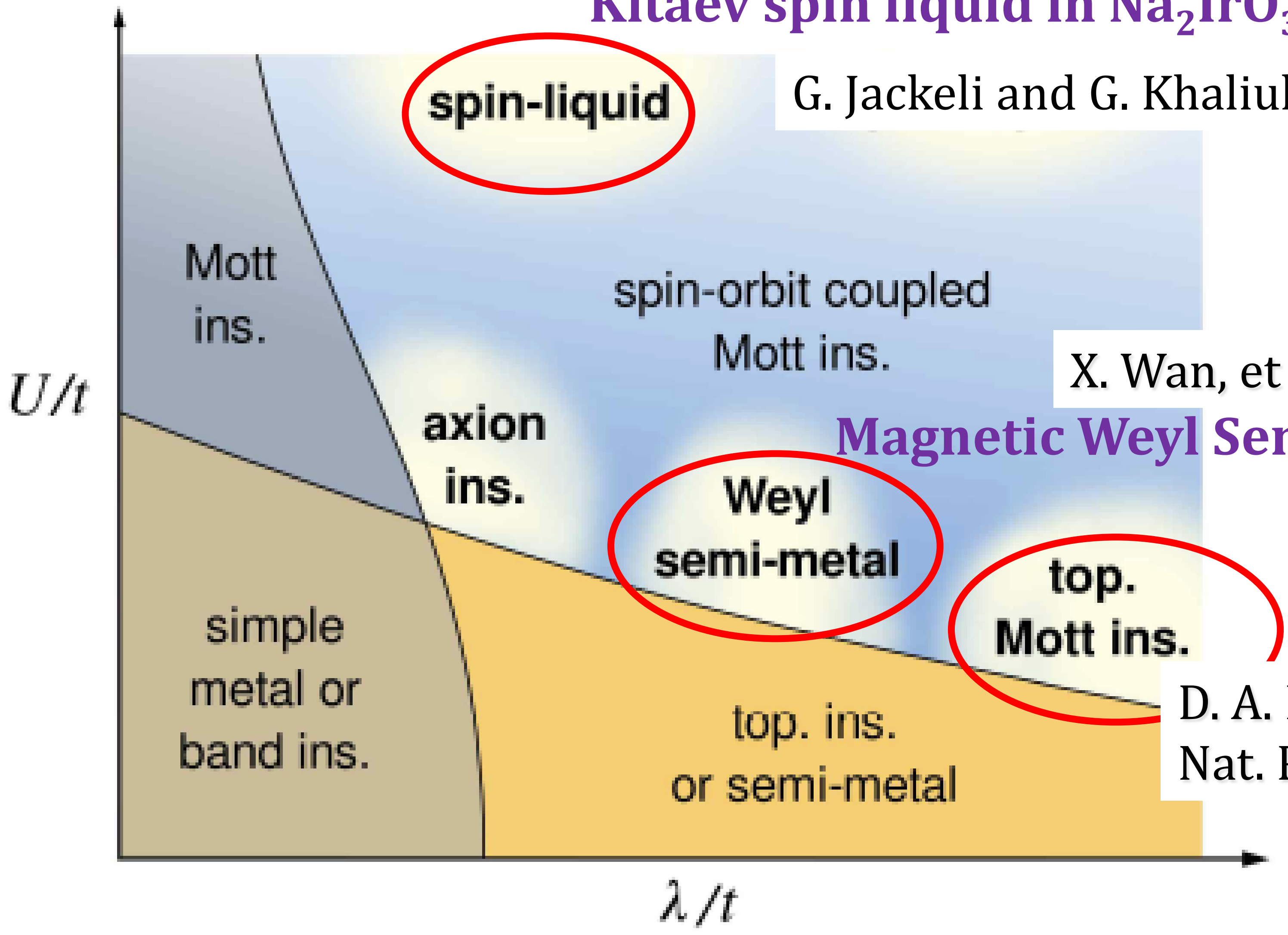
Interactions vs Spin-Orbit coupling

Kitaev spin liquid in Na_2IrO_3, Li_2IrO_3?

G. Jackeli and G. Khaliullin, PRL 2009



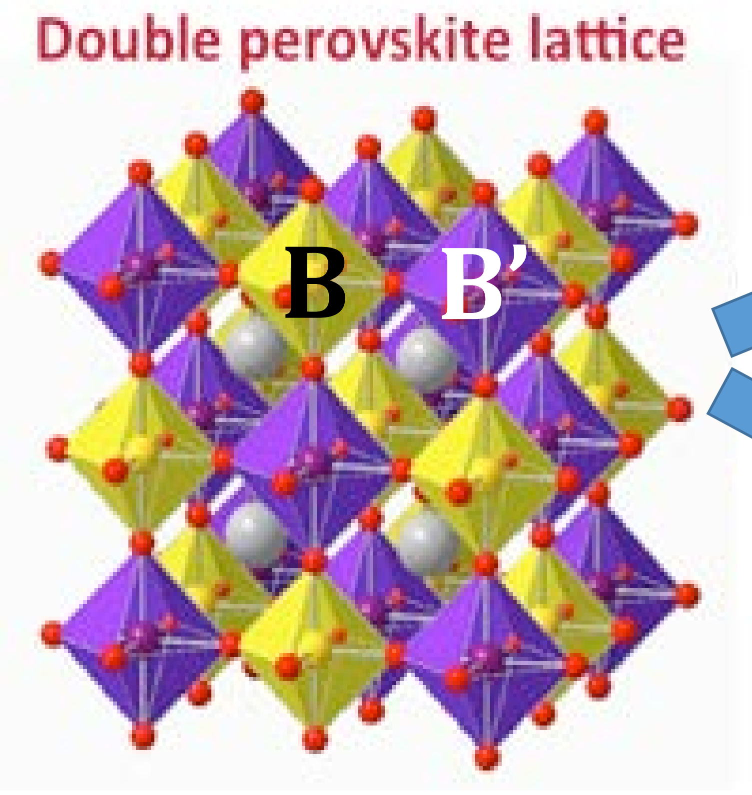
X. Wan, et al, PRB (2011)

Magnetic Weyl Semimetal in $Y_2Ir_2O_7$?

D. A. Pesin, L. Balents, Nat. Phys. (2010)

W. Witczak-Krempa, G. Chen, Y.B. Kim, L. Balents, Ann. Rev. Cond. Mat. (2014)

Ordered Double Perovskites



$A_2BB'O_6$

Mott insulators: Frustrated FCC Magnets

Ba_2Y**Mo**$O_{6,}$ Sr_2Y**Re**$O_{6,}$ Ba_2Y**Ir**$O_{6,}$ Ba_2Y**Ru**O_{6},...

B sublattice: Inert closed shell atom

B' sublattice: Magnetic ion with SOC

T. Aharen et al, Phys. Rev. B 81, 224409 (2010)
J. P. Carlo, et al, Phys. Rev. B 88, 024418 (2013)
G. Cao, et al, Phys. Rev. B 87, 155136 (2013)
G. Cao, et al, Phys. Rev. Lett. 112, 056402 (2014)

High Tc Ferromagnetic Metals

Sr_2**FeMo**$O_{6,}$ Ba_2**FeRe**$O_{6,}$ Pb_2**CrRe**$O_{6,}$ ...

B sublattice: 3d ion with strong magnetism

B' sublattice: 4d/5d ion with strong SOC

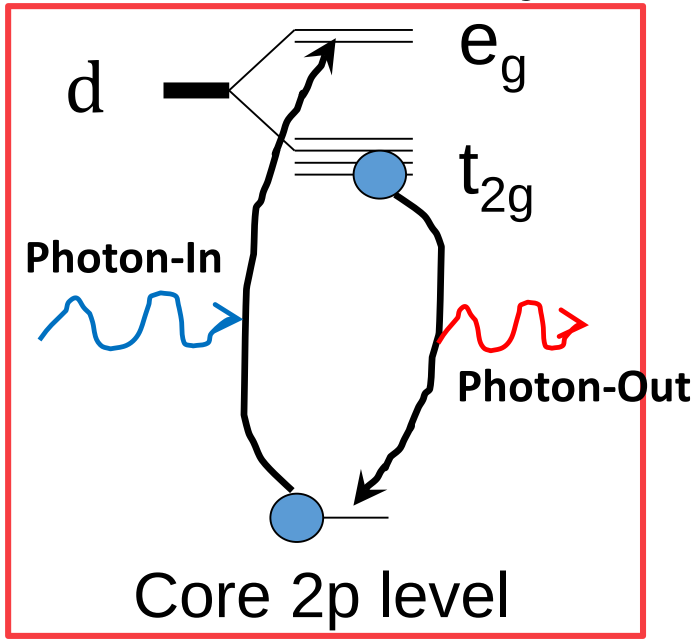
K.-I. Kobayashi, et al, Nature (1998) – Tokura group
W. Prellier, et al, J. Phys. Cond Matter (2000) – R. Greene group
N. Trivedi, M. Randeria, P. Woodward groups - OSU

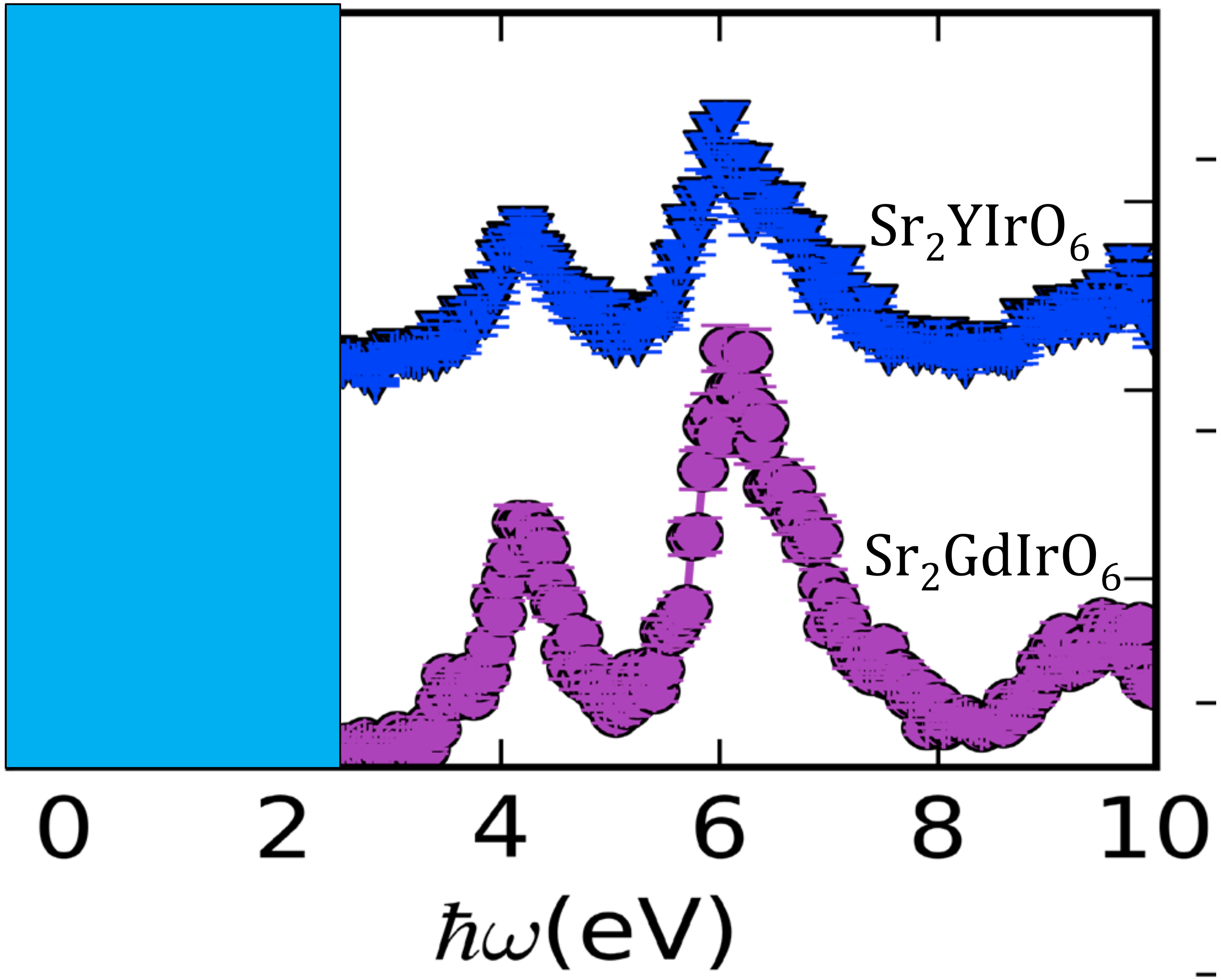
Outline

- Lego Block: $B'O_6$ Octahedra
- Deconstructing the Lego Block: RIXS
- Lego Block Assembly I: Frustrated Mott Insulators
- Lego Block Assembly II: High Tc Chern Insulators

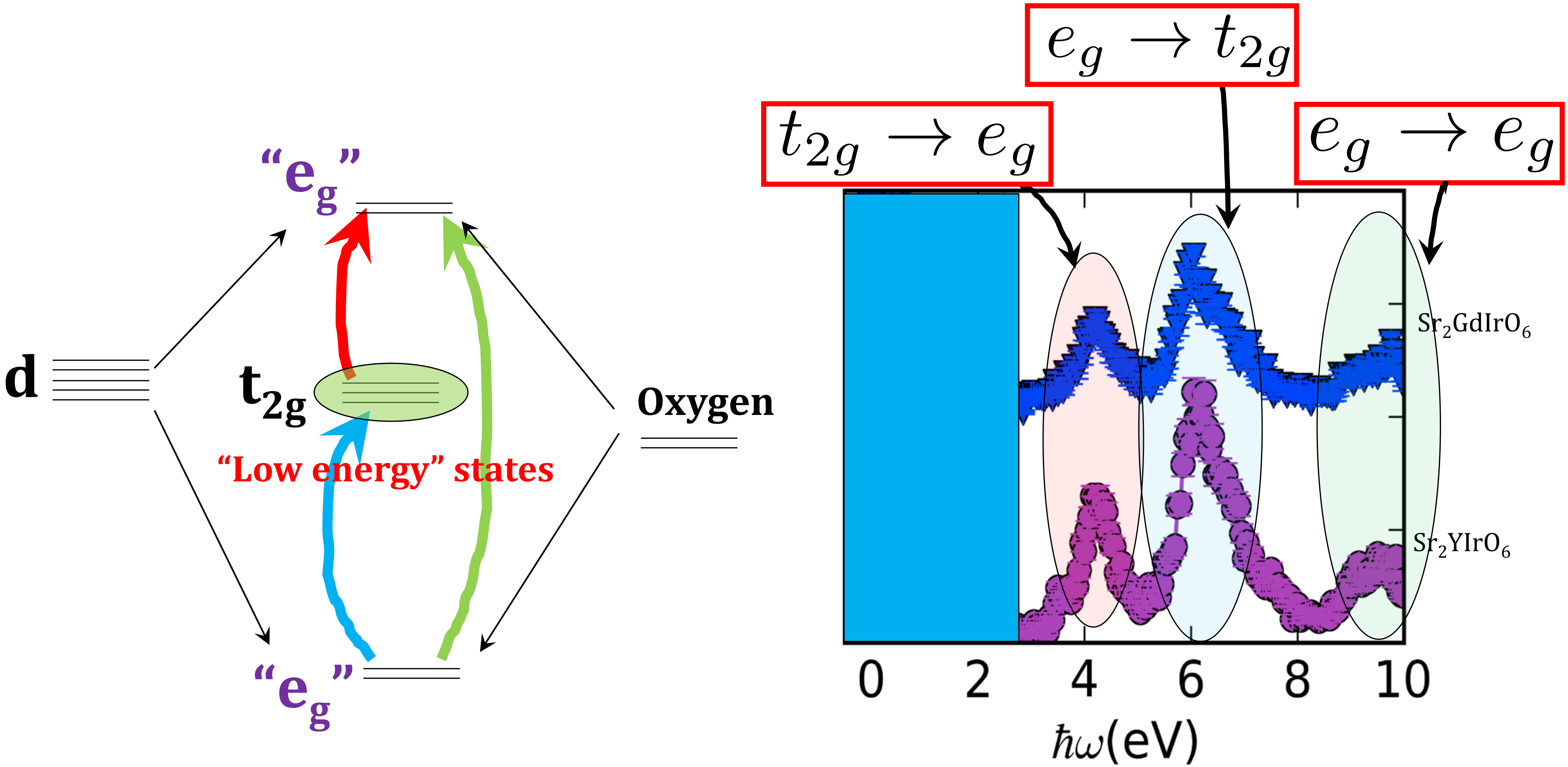
"Lego Block": Crystal Field Levels

Resonant Inelastic X-ray Scattering





B. Yuan, et al,PRB 2017



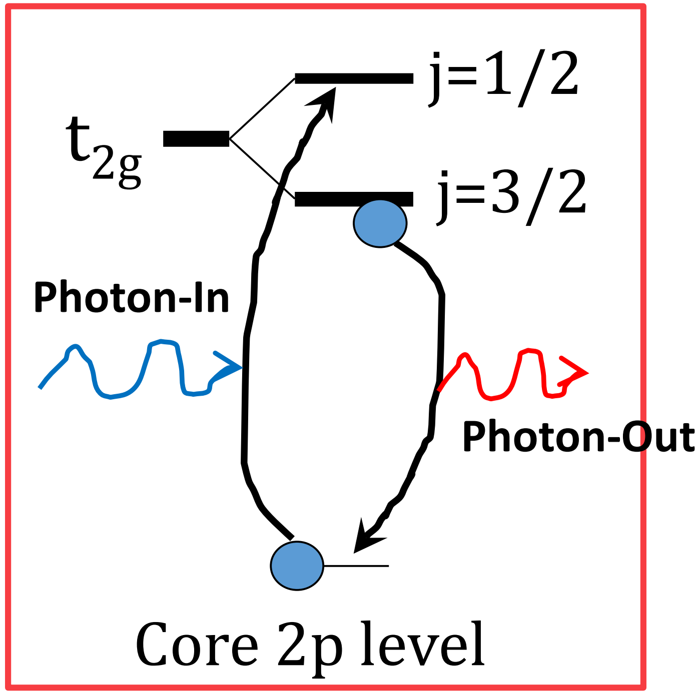
"e_g"
d
t_{2g}
"Low energy" states
Oxygen
"e_g"
$e_g \rightarrow t_{2g}$
$t_{2g} \rightarrow e_g$
$e_g \rightarrow e_g$
Sr_2GdIrO_6
Sr_2YIrO_6
0
2
4
6
8
10
$\hbar\omega$(eV)

"Lego Block": Spin-Orbit Coupling

Spin orbit coupling and interactions

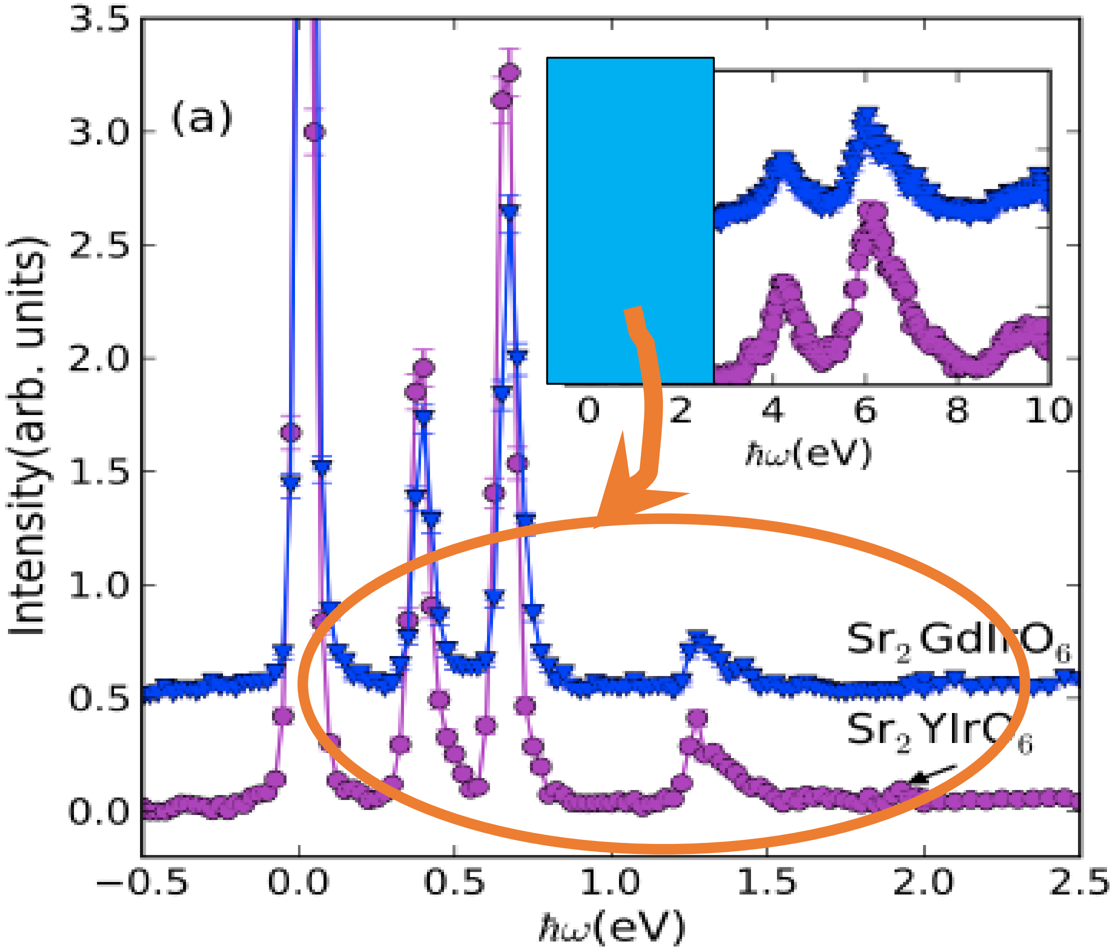
$$P_{t_{2g}} \vec{L} P_{t_{2g}} = -\vec{\ell} \quad (\ell = 1)$$

$$H_{\text{s.o.}} = -\lambda \vec{\ell} \cdot \vec{s}$$



Naive: Single peak at E = $3\lambda/2$
Works perfectly for d^5 iridates

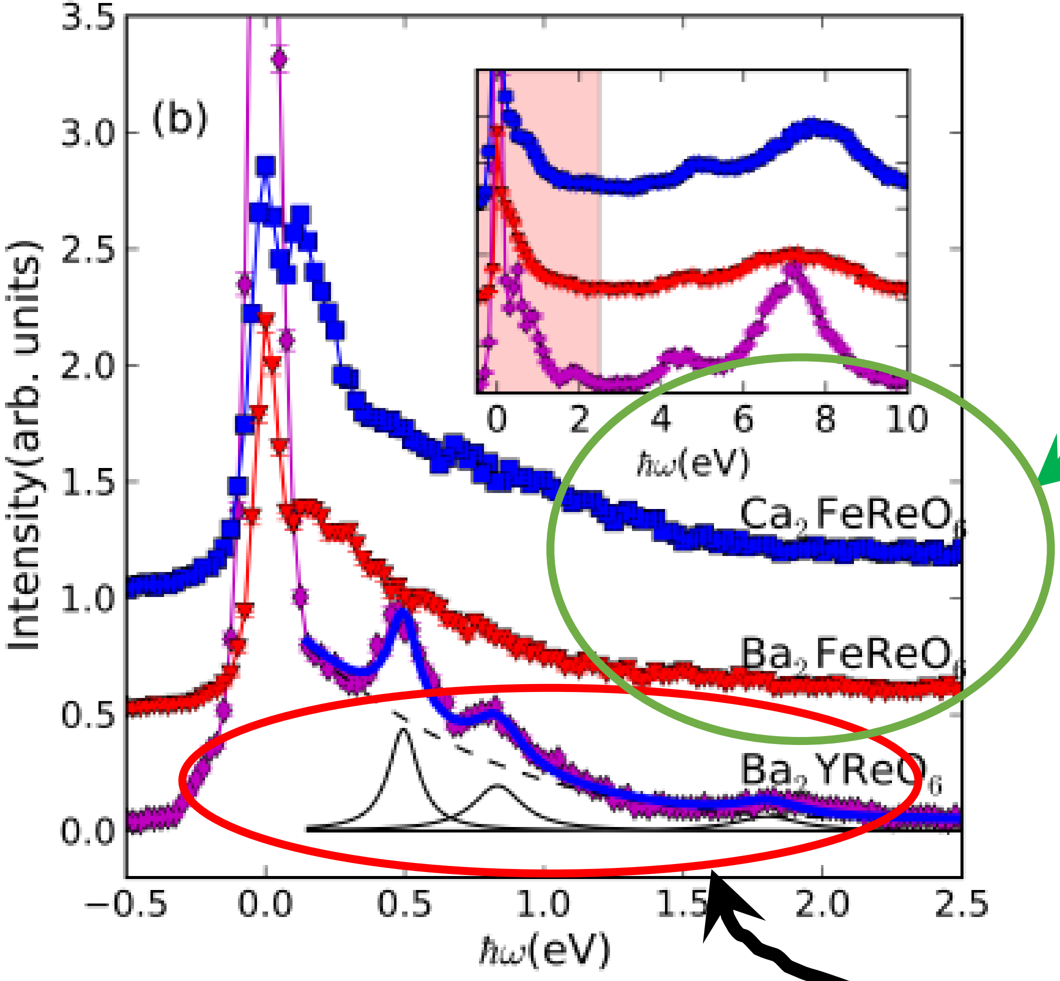
d^4 Iridates: Much richer structure



B. Yuan, et al, **AP**, Y. J. Kim, PRB (2017)

d^2 Rhenates

Metal/Semimetal
Nothing much visible

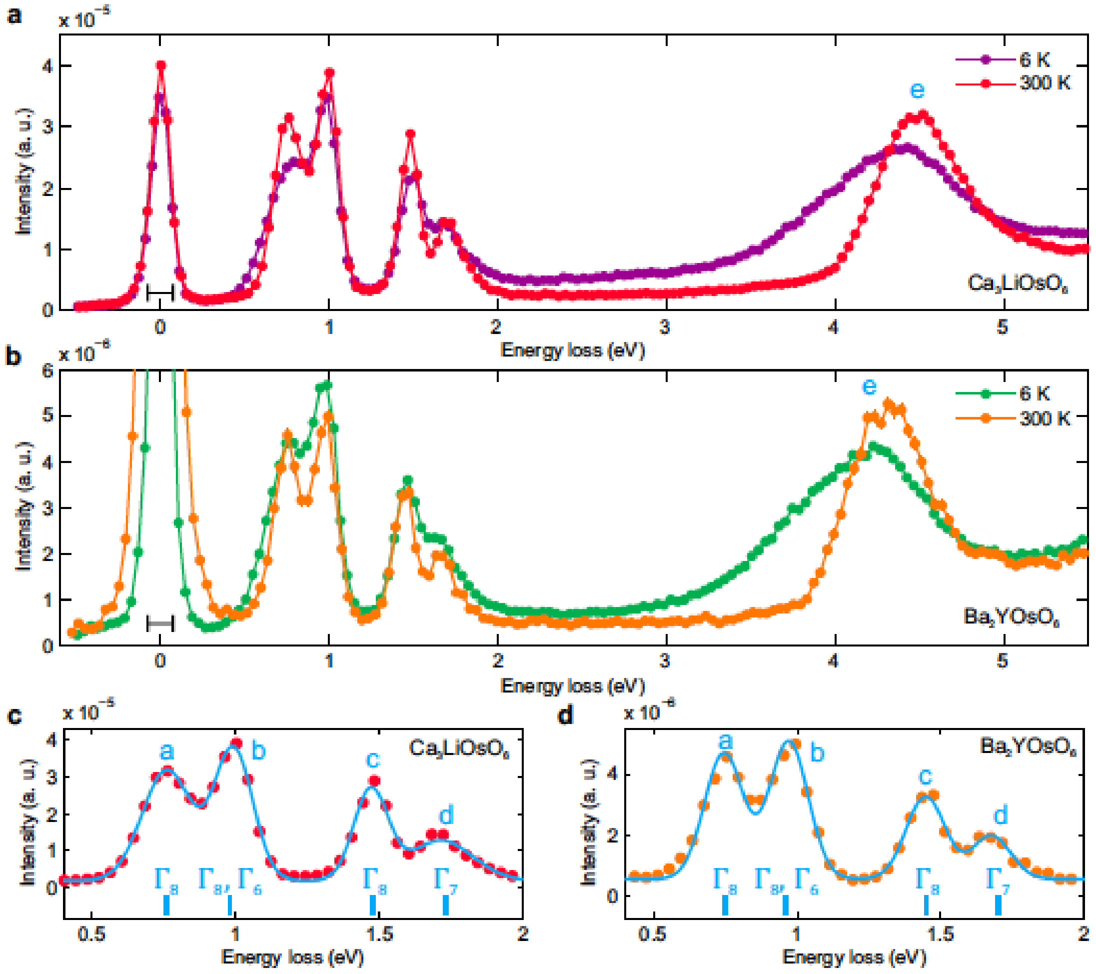


Insulators: Peaks

Bo Yuan, et al, PRB 2017

d^3 Osmates

Poorer resolution: Peaks still visible

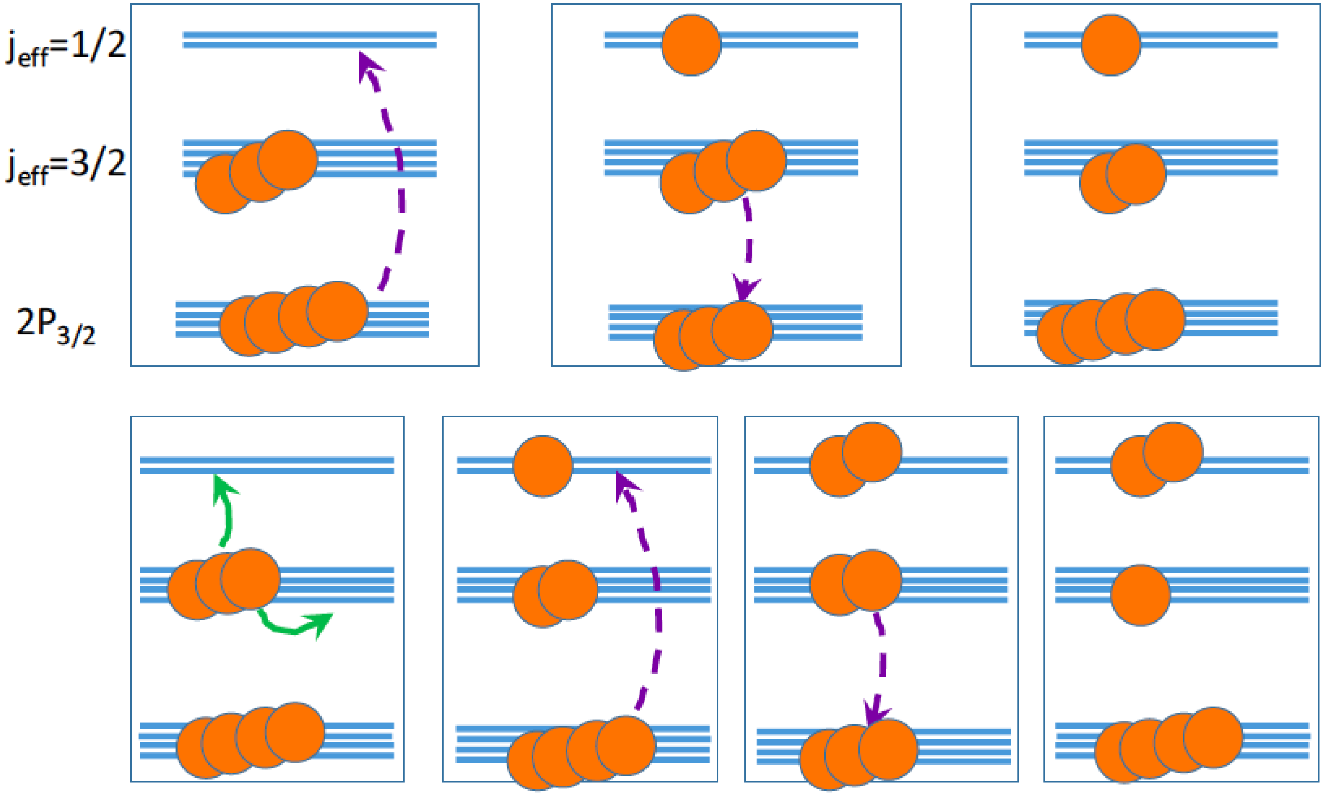


ORNL: A Taylor, et al, PRL 2017)

RIXS: Simple Picture

$$H_{\text{int}} = \frac{U}{2} : (\sum_{\ell} n_{\ell})^2 : -5\frac{J_H}{2} \sum_{\ell<\ell'} n_{\ell} n_{\ell'}$$

$$-2J_H \sum_{\ell<\ell'} \vec{S}_{\ell} \cdot \vec{S}_{\ell'} + J_H \sum_{\ell\neq\ell'} d^{\dagger}_{\ell\uparrow} d^{\dagger}_{\ell\downarrow} d_{\ell'\downarrow} d_{\ell'\uparrow}$$

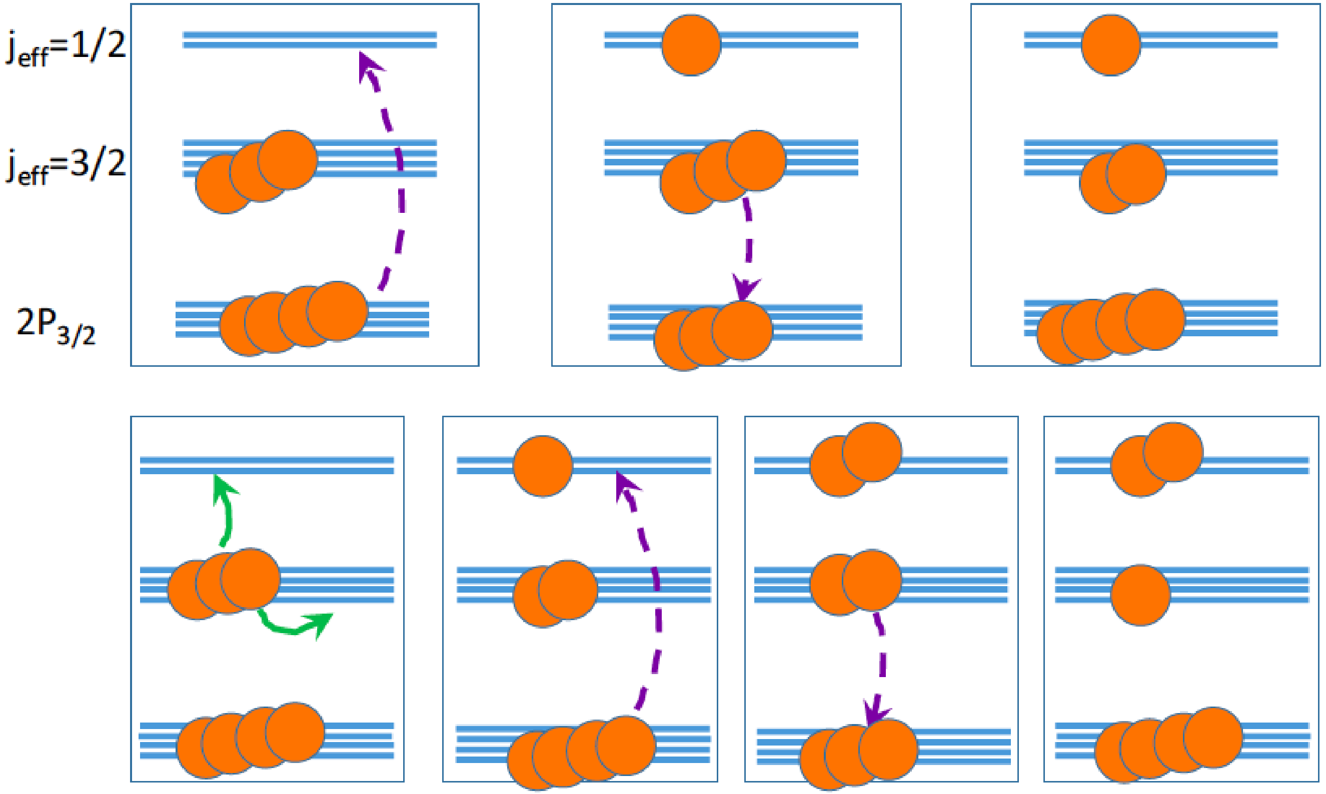


A. Paramekanti, et al, PRB 2018

RIXS: Simple Picture

$$H_{\rm int} = \frac{U}{2} : (\sum_\ell n_\ell)^2 : -5\frac{J_H}{2} \sum_{\ell<\ell'} n_\ell n_{\ell'}$$

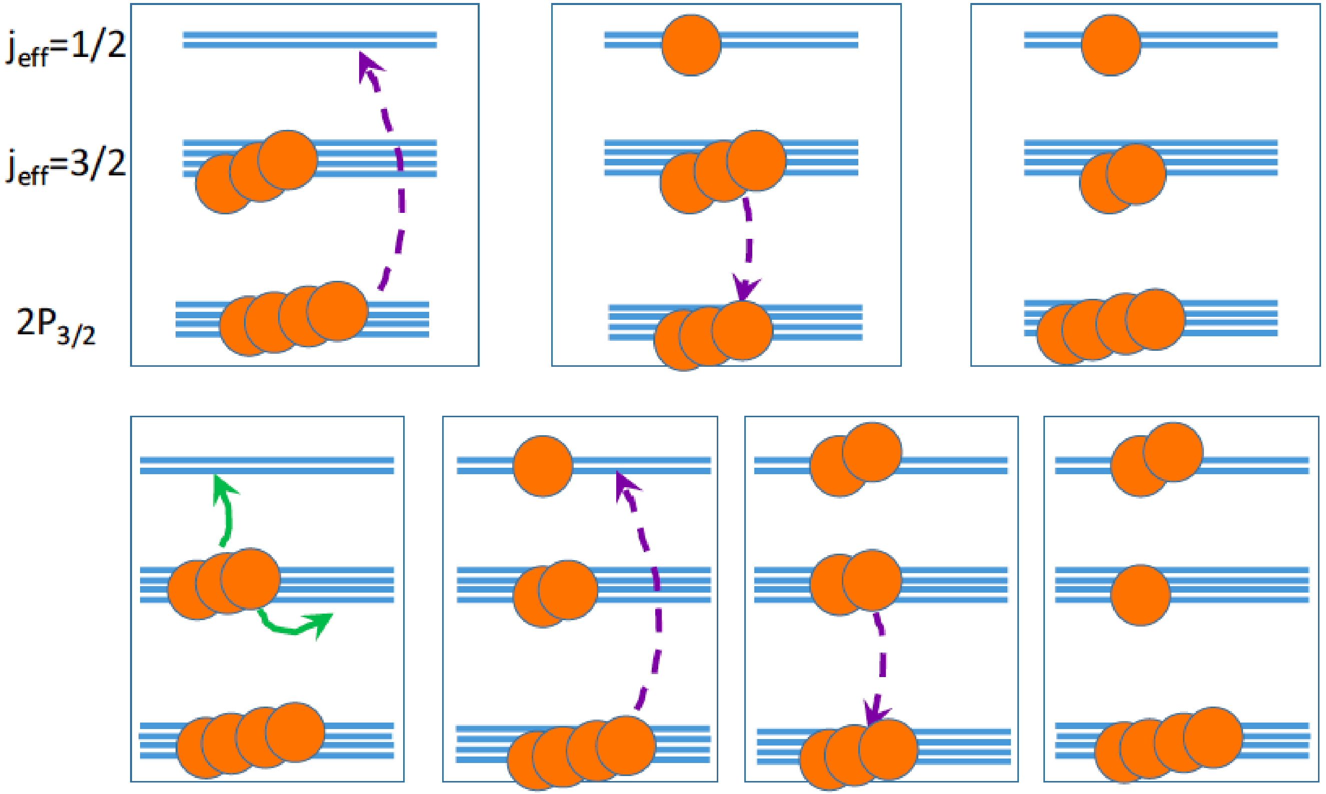
$$-2J_H \sum_{\ell<\ell'} \vec{S}_\ell \cdot \vec{S}_{\ell'} + J_H \sum_{\ell\neq\ell'} d^\dagger_{\ell\uparrow} d^\dagger_{\ell\downarrow} d_{\ell'\downarrow} d_{\ell'\uparrow}$$

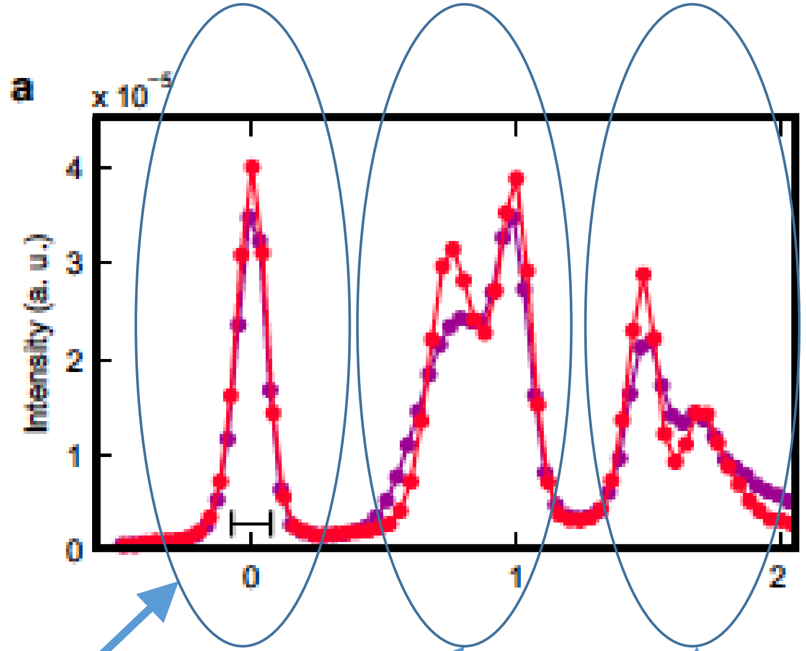


A. Paramekanti, et al, PRB 2018

RIXS: Simple Picture

$$H_{\rm int} = \frac{U}{2} : (\sum_\ell n_\ell)^2 : -5\frac{J_H}{2}\sum_{\ell<\ell'} n_\ell n_{\ell'} - 2J_H \sum_{\ell<\ell'} \vec{S}_\ell \cdot \vec{S}_{\ell'} + J_H \sum_{\ell\neq\ell'} d^\dagger_{\ell\uparrow} d^\dagger_{\ell\downarrow} d_{\ell'\downarrow} d_{\ell'\uparrow}$$





Elastic line

1-electron excitation

2-electron excitation

A. Paramekanti, et al, PRB 2018

RIXS Theory: Simplifying Assumptions

$$\frac{d^2\sigma}{d\Omega dE_i} \approx \left|\frac{1}{E_g - \bar{E}_n + E_i + i\frac{\bar{\Gamma}_n}{2}}\right|^2 \sum_f \left|\sum_n \langle f|T^\dagger|n\rangle\langle n|T|g\rangle\right|^2 \delta(E_g - E_f + E_i - E_o)$$

Average out denominator;
short core-hole lifetime

Focus on matrix elements, energies
Ignore core hole interactions

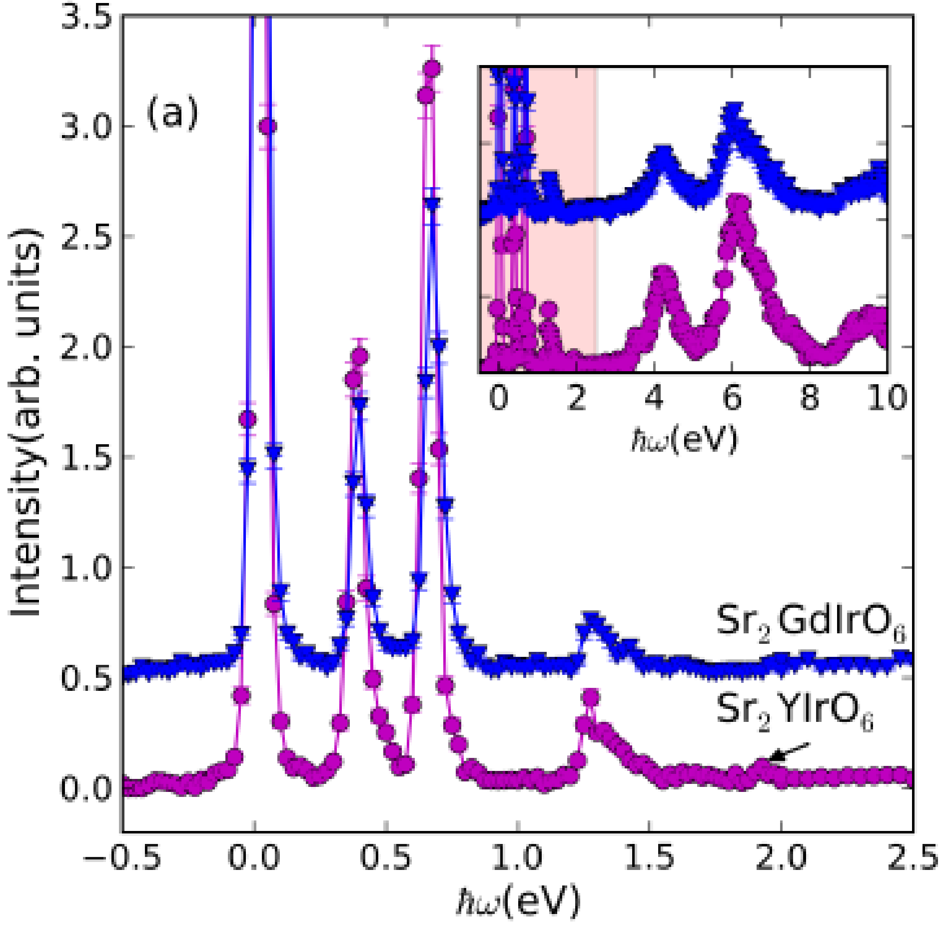
Dipole Approximation, Ignore Core-Hole interaction

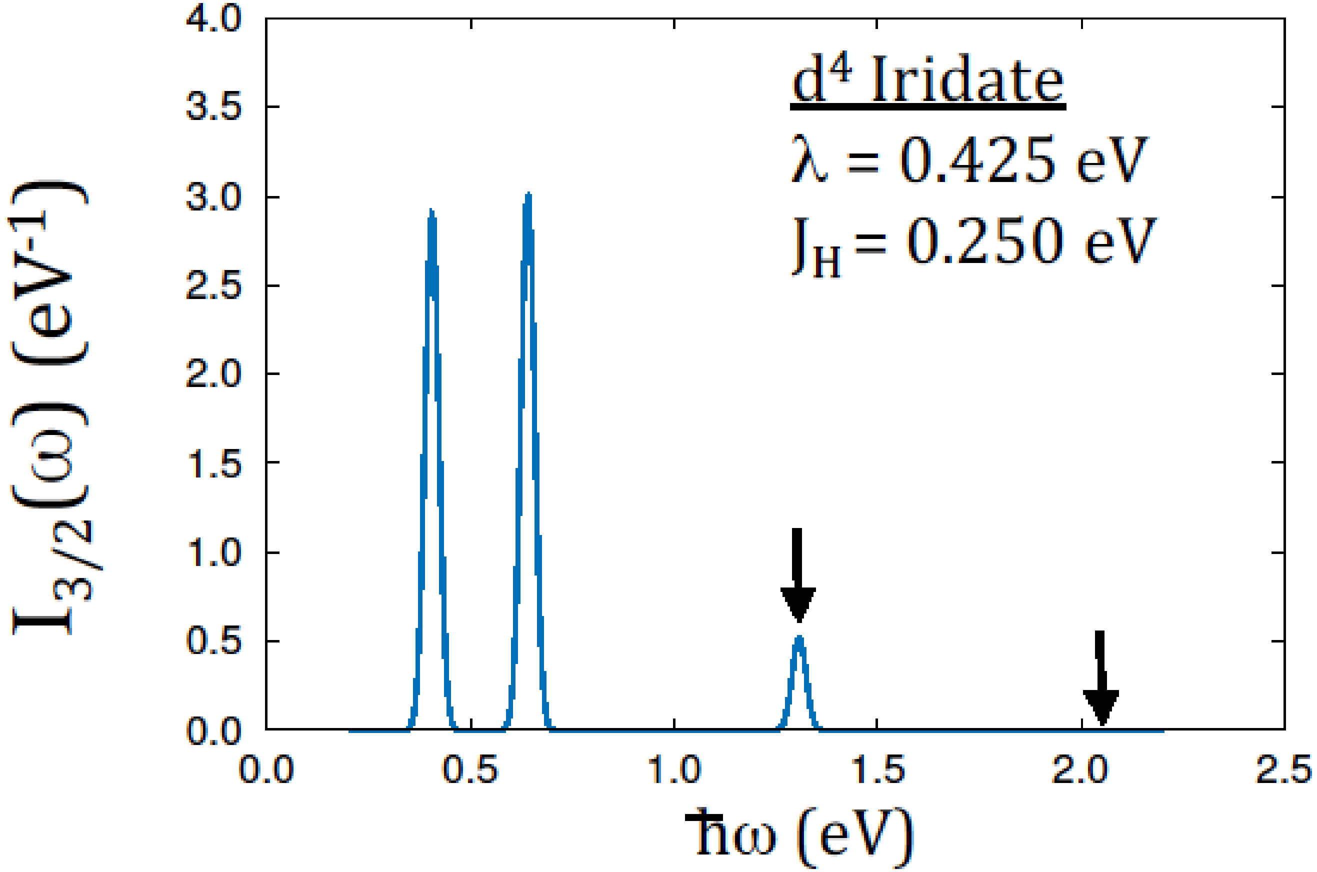
$$\langle n|T|g\rangle = \hat{\epsilon}^{\alpha}_{\text{in}}\langle n|p^\dagger_{\beta\sigma}d^\dagger_{\alpha\beta\sigma}|g\rangle$$
$$\langle f|T^\dagger|n\rangle = \hat{\epsilon}^{\mu}_{\text{out}}\langle f|d_{\mu\nu\sigma'}p_{\nu\sigma'}|n\rangle$$

$$H_{\text{int}} = \frac{U}{2} : (\sum_\ell n_\ell)^2 : -5\frac{J_H}{2}\sum_{\ell<\ell'} n_\ell n_{\ell'} - 2J_H \sum_{\ell<\ell'} \vec{S}_\ell \cdot \vec{S}_{\ell'} + J_H \sum_{\ell\neq\ell'} d^\dagger_{\ell\uparrow}d^\dagger_{\ell\downarrow}d_{\ell'\downarrow}d_{\ell'\uparrow}$$

Exact diagonalization results

d^4 Iridates

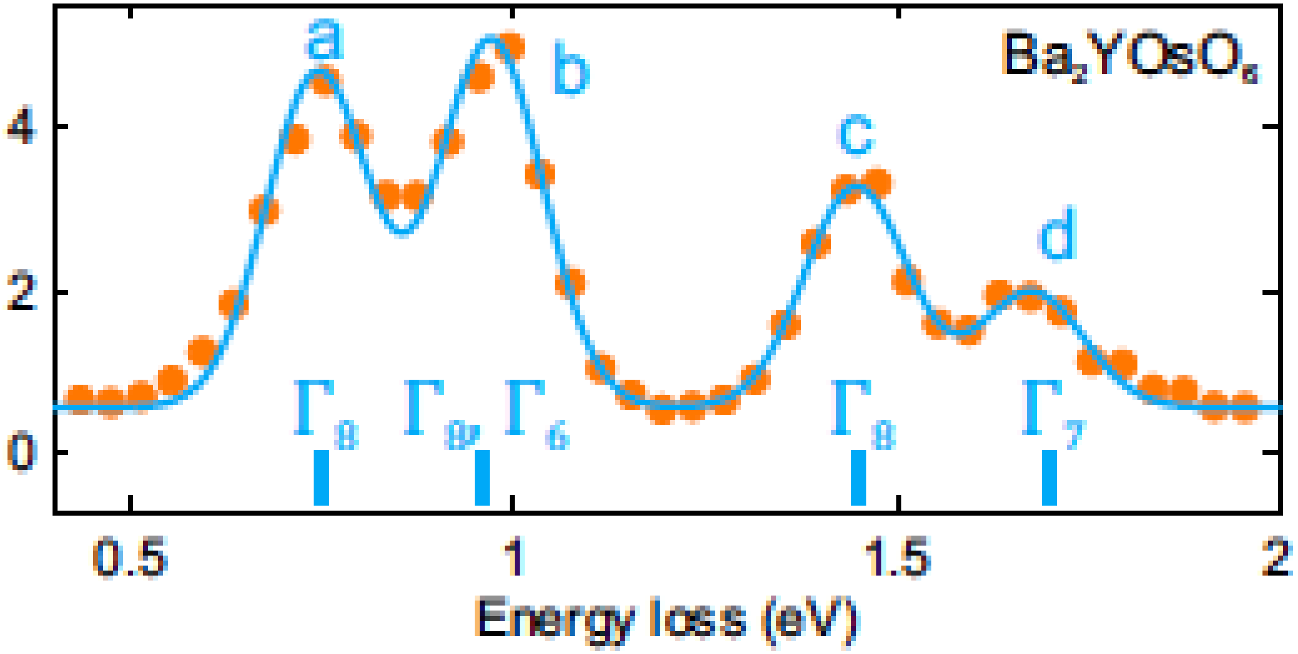


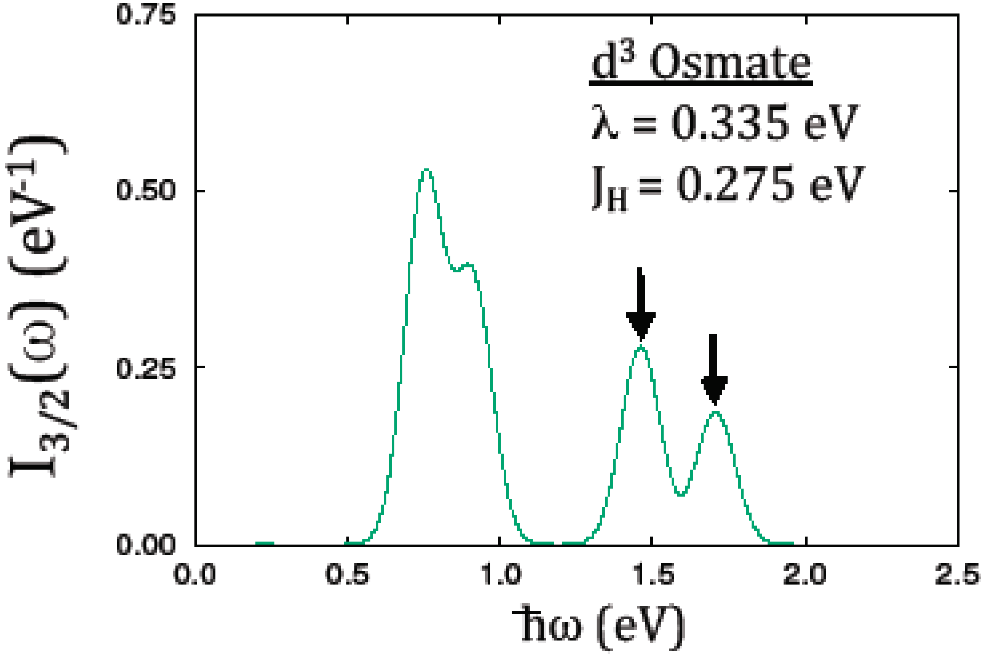


A. Paramekanti, et al, PRB 2018

Exact diagonalization results

d^3 Osmates

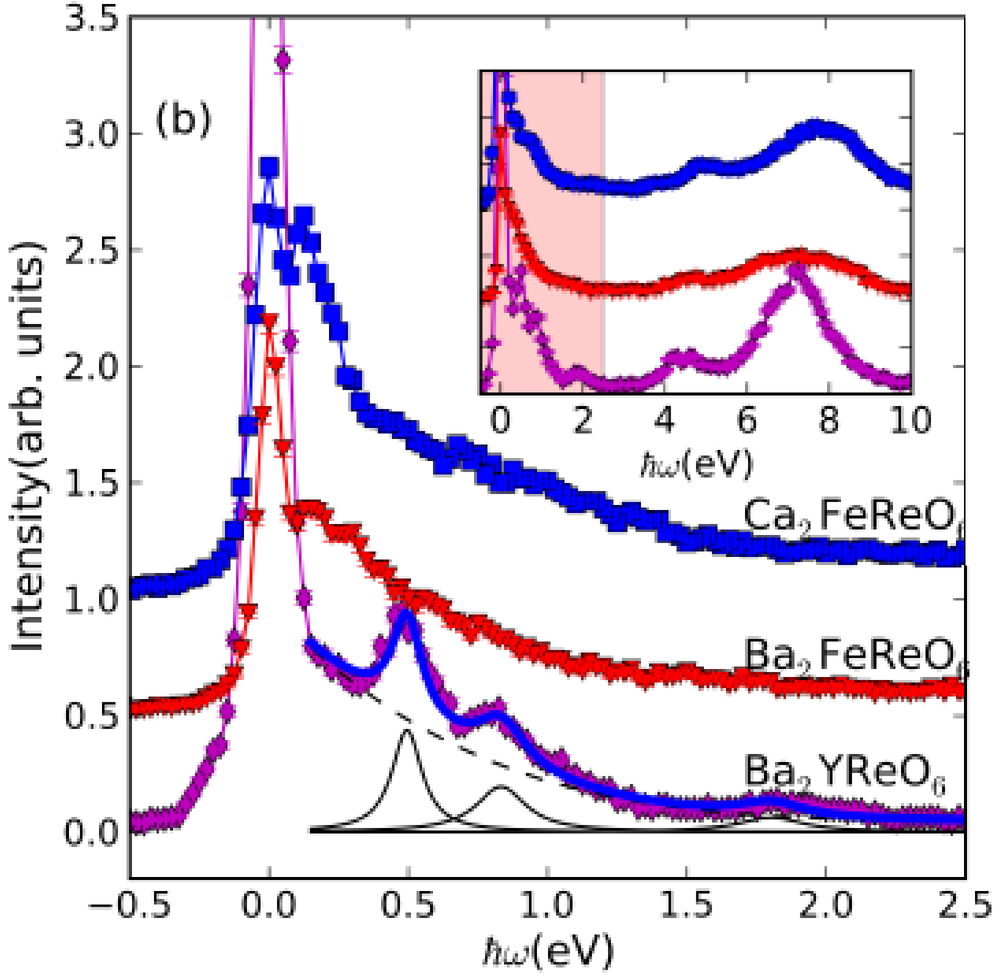


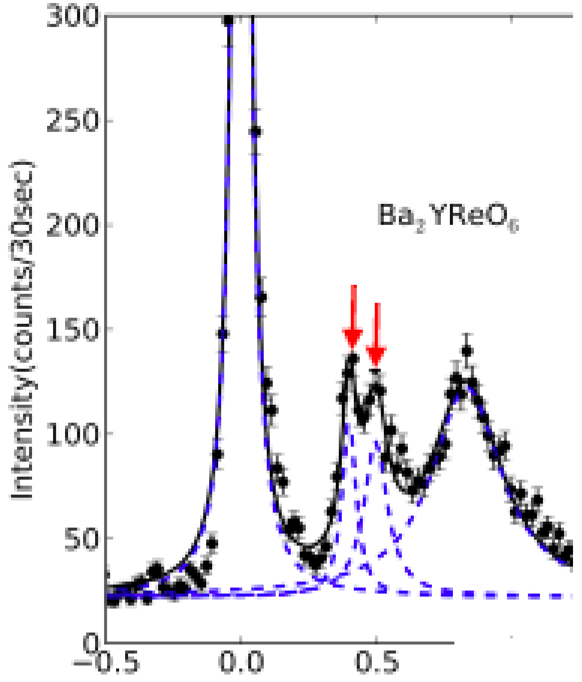


A. Paramekanti, et al, PRB 2018

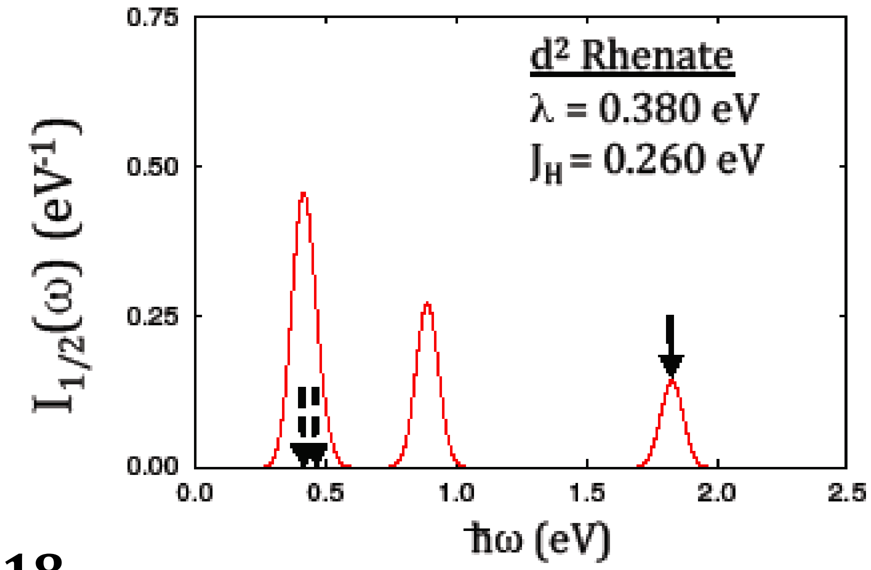
Exact diagonalization results

d^2 Rhenates





d^2 configuration has **only one** 2-electron excited state



A. Paramekanti, et al, PRB 2018

"Lego Block" Hamiltonian

$$H = H_{\mathrm{SOC}}[\lambda] + H_{\mathrm{int}}$$

Material		λ	J_H	Peak 1	Peak 2	Peak 3	Peak 4
Ba_2YReO_6	Ex			*[0.40]*	*[0.50]*	*0.83*	*1.85*
(ref.55,this)	Th	0.380	0.260	0.41	0.47	0.89	1.83
Ba_2YOsO_6	Ex			*0.745*	*0.971*	*1.447*	*1.68*
(ref.54)	Th	0.335	0.275	0.75	0.91	1.46	1.71
Sr_2YIrO_6	Ex			*0.39*	*0.66*	*1.30*	*∼2*
(ref.55)	Th	0.425	0.250	0.41	0.64	1.31	2.06
Ba_2YIrO_6	Ex			*0.35*	*0.60*	*1.18*	-
(ref.56)	Th	0.385	0.230	0.37	0.58	1.19	1.88

d^3 (Osmates)

SOC still important!

d^4 (Ba_2YIrO_6)

Magnetism?

Impurities?

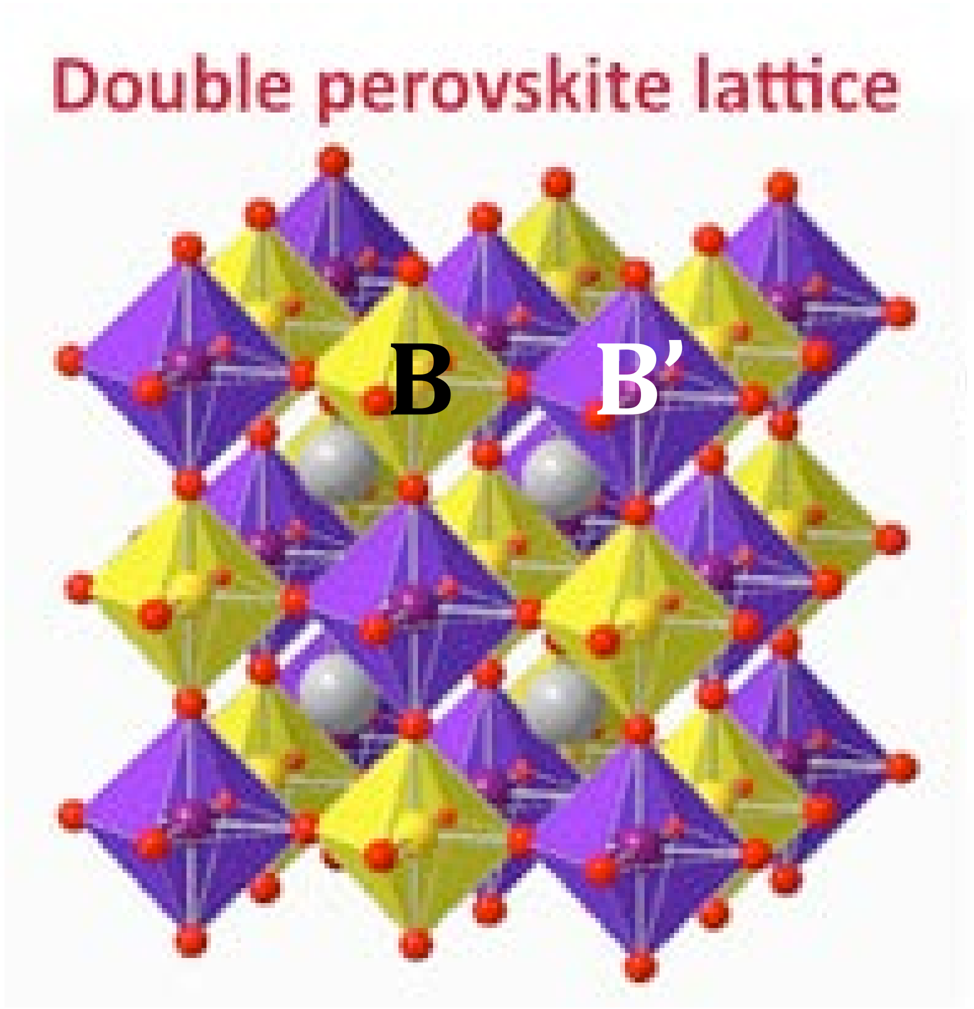
Cluster Excitons?

A. Paramekanti, et al, PRB 2018

Material Variations

- Different Atomic Numbers
- Different Degrees of Oxygen Hybridization

Coupling the “Lego Blocks”
Mott Insulators on the Frustrated FCC Lattice



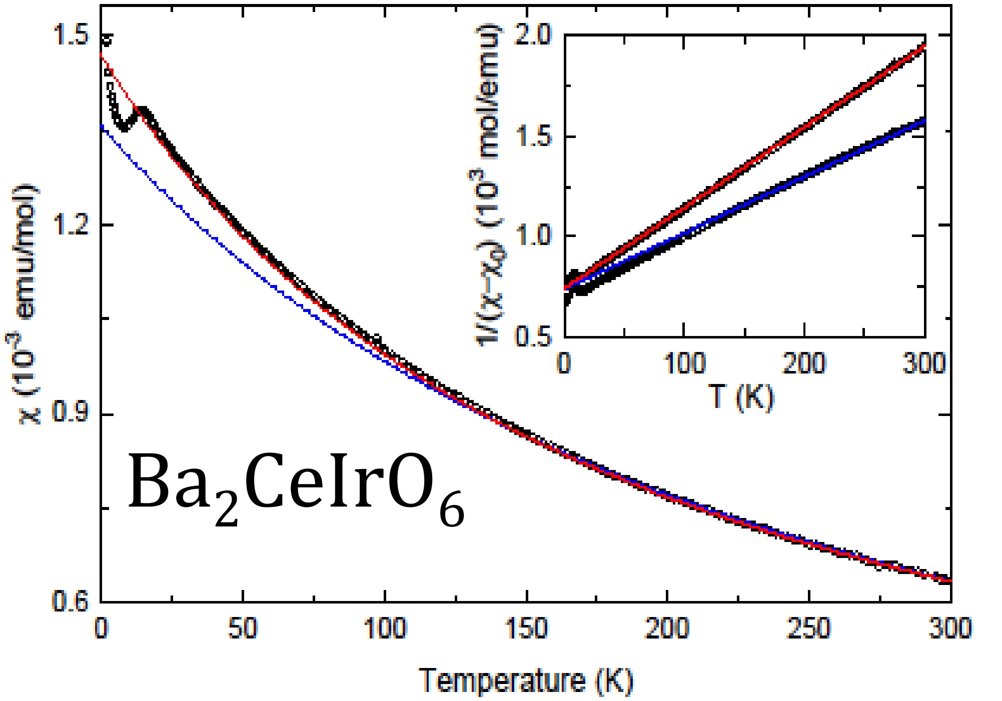
d^5 iridates

La_2MgIrO_6
La_2ZnIrO_6
Monoclinic $P2_1/n$

Ba_2CeIrO_6
Cubic $Fm\bar{3}m$

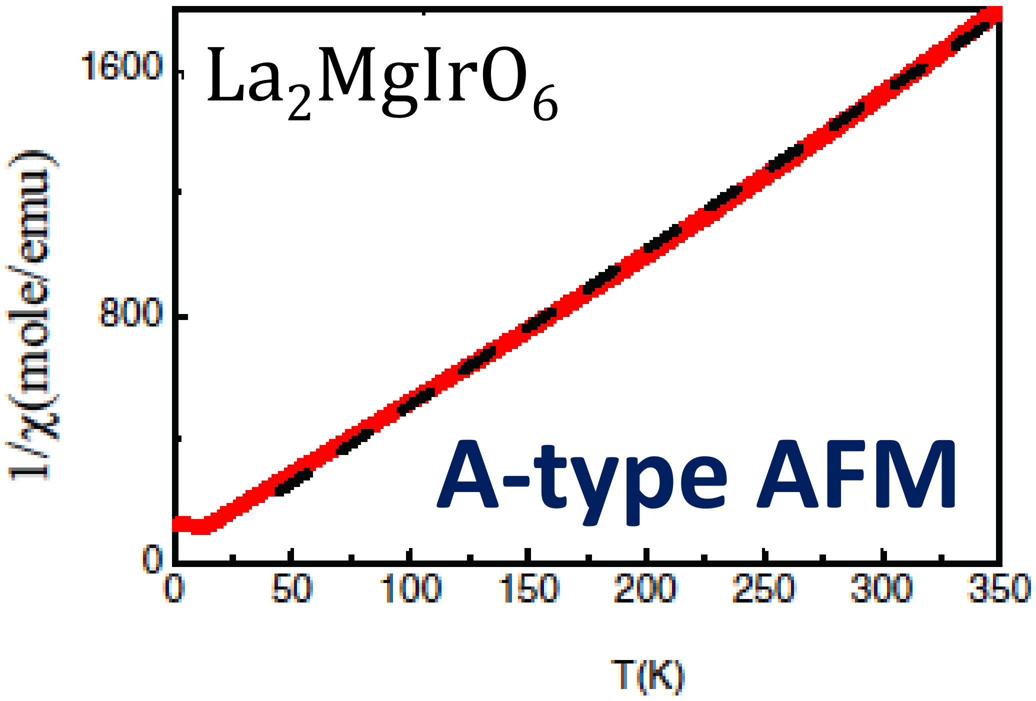
Other fillings:
Chen, Pereira, Balents (2010); Chen, Balents (2011);
Svoboda, Randeria, Trivedi (2017, 2018); AP et al (2018)

j=1/2 Mott Insulators on the FCC Lattice



$$\Theta_{CW} \approx -200K \quad T_N \approx 14K$$

A.Revelli, et al, **arXiv:1901.06215 (Cologne)**
A.Aczel, et al, **arXiv:1901.08146 (ORNL/UTK); A-type AFM**

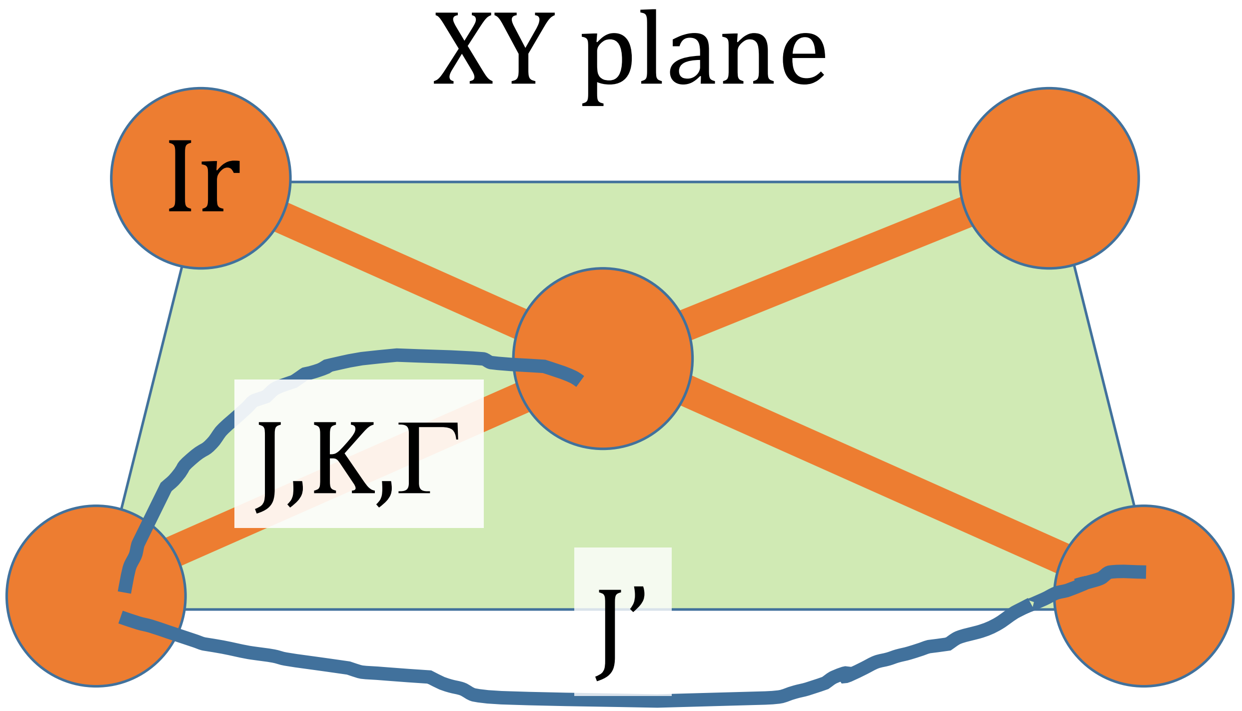


$$\Theta_{CW} \approx -24K \quad T_N \approx 12K$$

Guixin Cao, et al,PRB (2013) (ORNL/UTK)
A.Aczel, et al, PRB (2017); A.Cook, et al, PRB (2017)

- **Strong Frustration for ideal FCC magnet**
- **Highly Tuned by Weak Distortion: Magnetoelastic**

j=1/2 Mott Insulators on the Ideal FCC Lattice
Theory: Exchange Couplings



Perturbative + DFT

$K/J \approx 0.2$ AFM

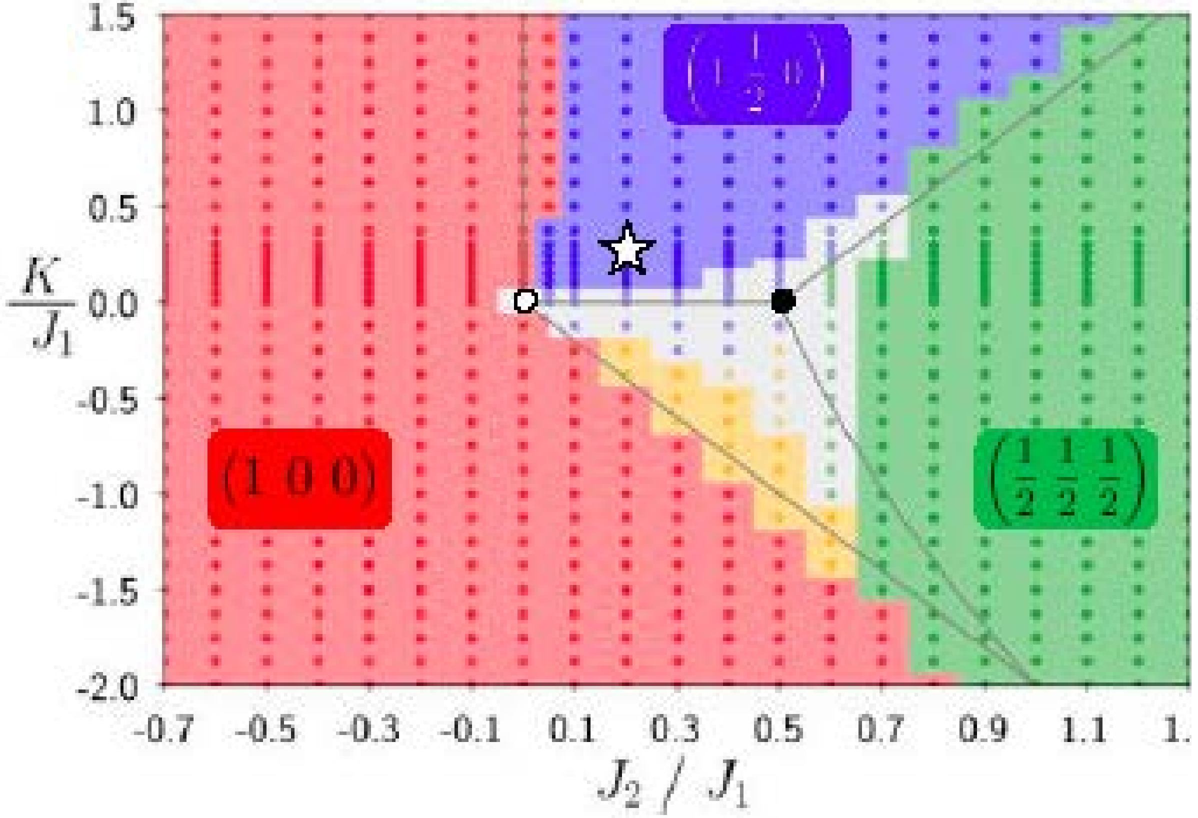
$J'/J \approx 0.2$ nnbr

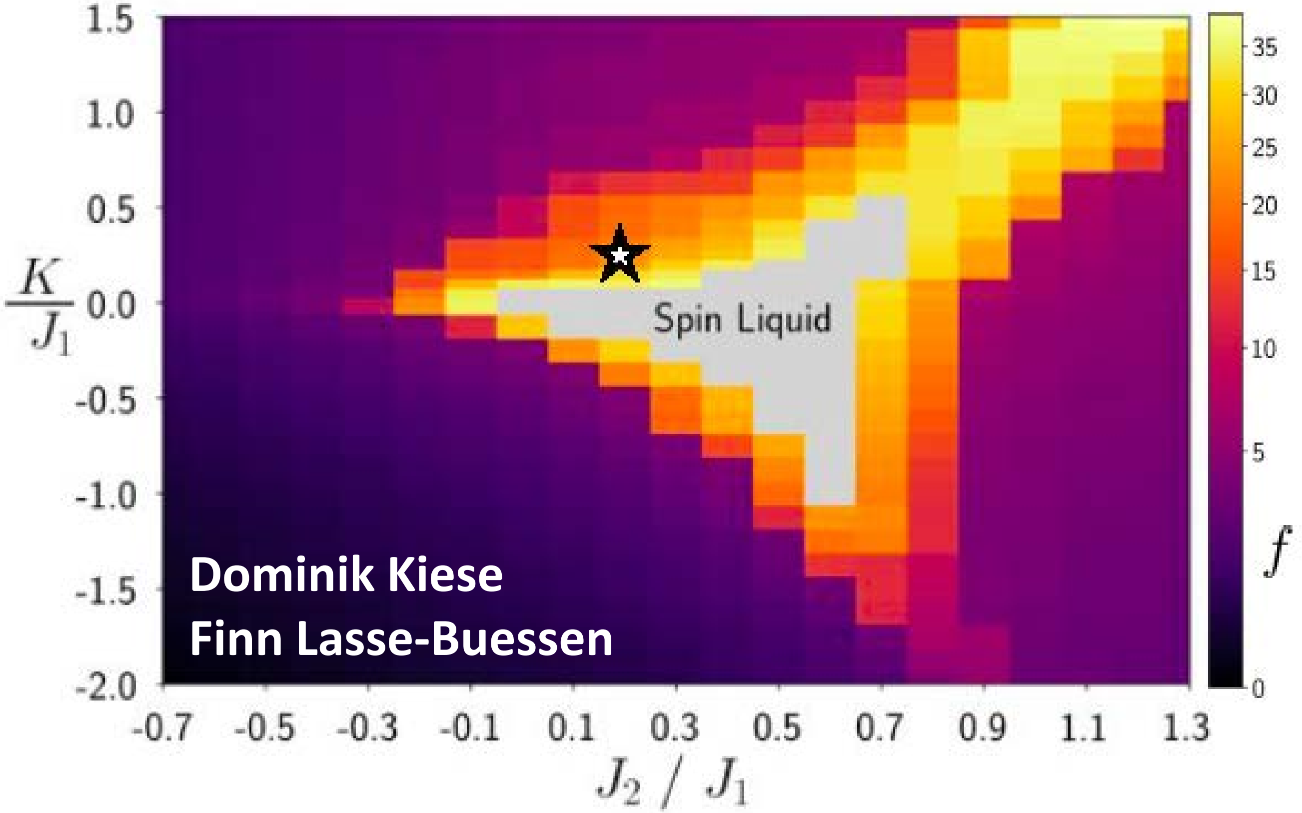
$\Gamma/J \approx 0$

$$H_{ij} = J\vec{S}_i \cdot \vec{S}_j + K S_i^z S_j^z + \Gamma(S_i^x S_j^y + S_i^y S_j^x)$$

A.Revelli, et al, **arXiv:1901.06215 (Cologne)**

j=1/2 Mott Insulators on the Ideal FCC Lattice
Theory: Spin Functional-RG

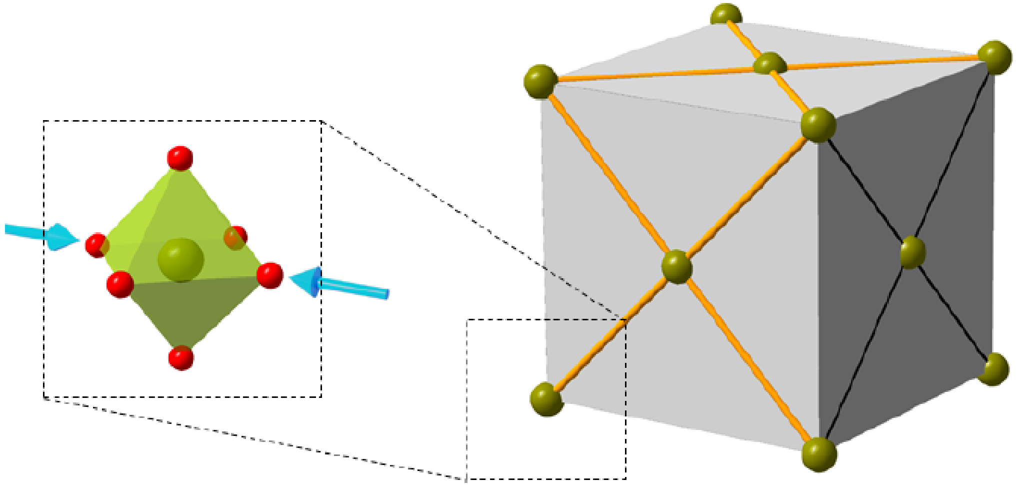




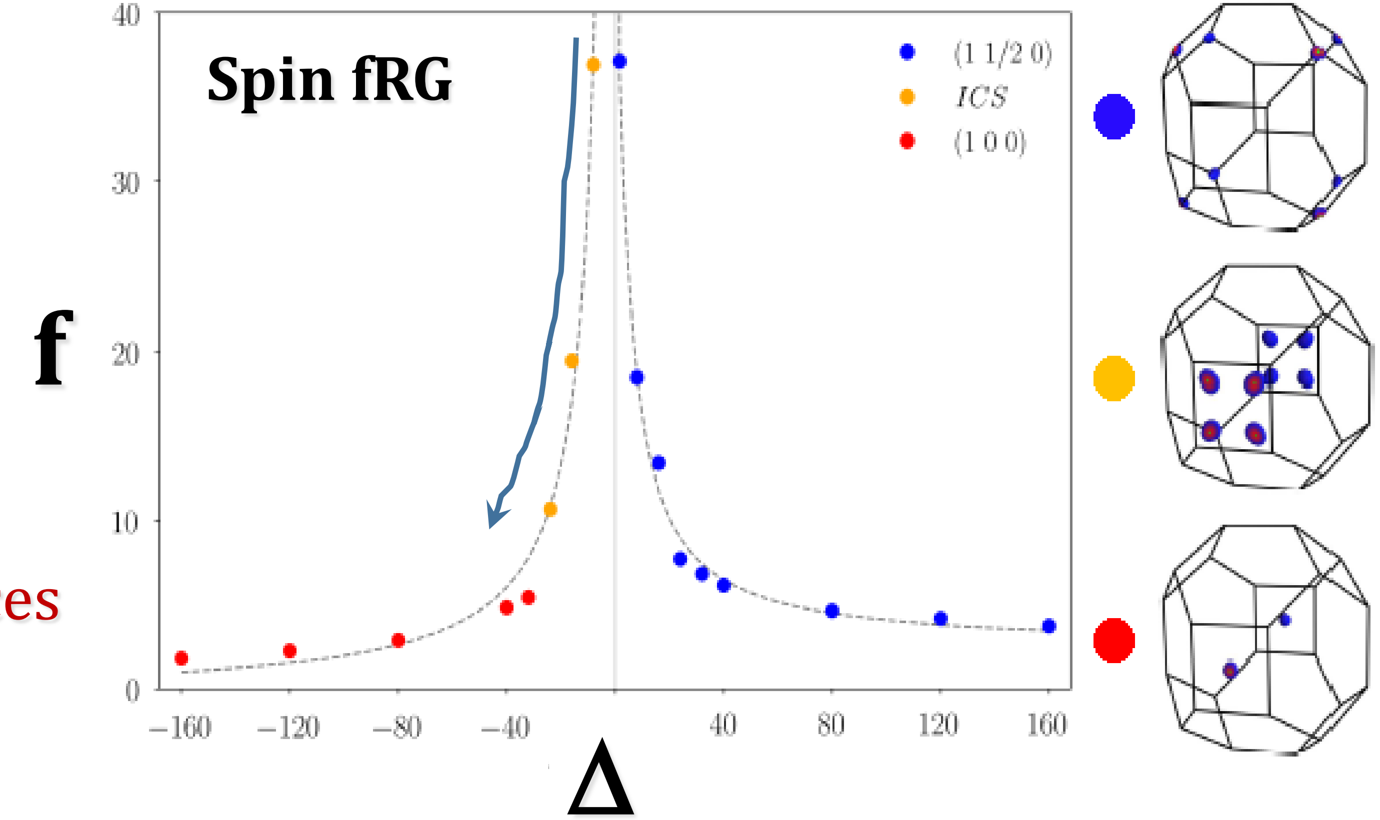
- Proximate to Quantum Spin Liquid (type of QSL unclear)
- Very large frustration parameter, f ~ 30 **(>> classical)**
- Kitaev interaction **suppresses** frustration

A.Revelli, et al, **arXiv:1901.06215**

Mott Insulators on the FCC Lattice
Theory: Impact of Weak Distortion



- Z-compression ($\Delta > 0$) strongly enhances XY-plane Heisenberg exchange
- Z-stretch ($\Delta < 0$) strongly suppresses XY-plane Heisenberg exchange

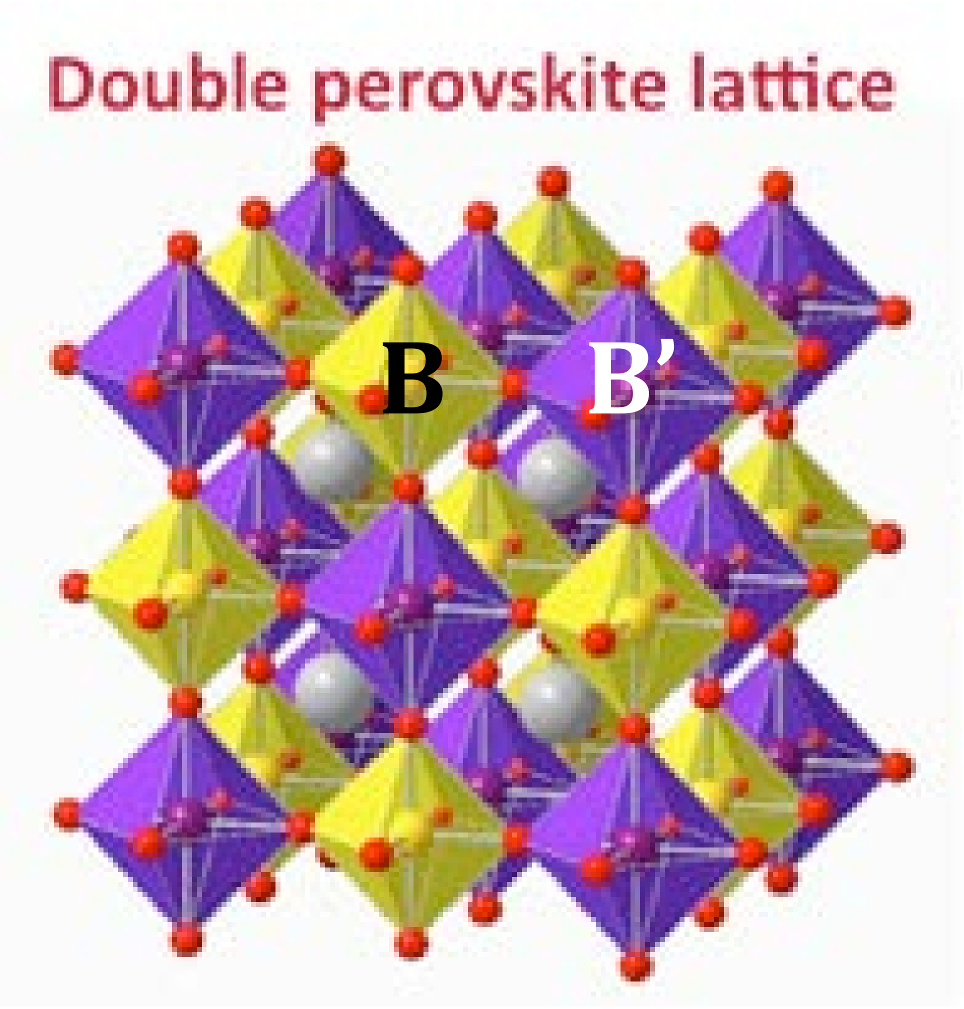


La_2MgIrO_6: **Global** distortion, f ~ 2
Ba_2CeIrO_6: **Local** distortions, f ~ 14

Strong magneto-elastic effect

A.Revelli, et al, **arXiv:1901.06215**

Coupling the "Lego Blocks"
High Tc Chern Insulators in Ultrathin Films

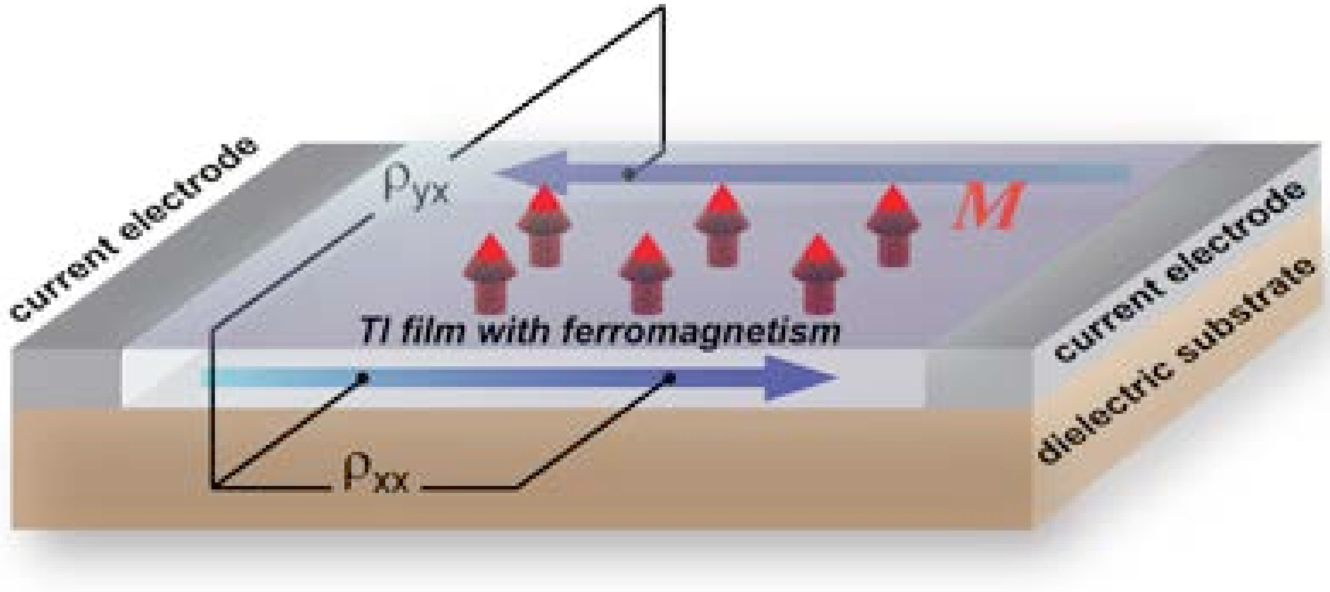


Ba_2FeReO_6
Sr_2FeMoO_6

- **Half-Metallic FMs**
- **Spin-Polarized**
- **High Bulk FM Tc**

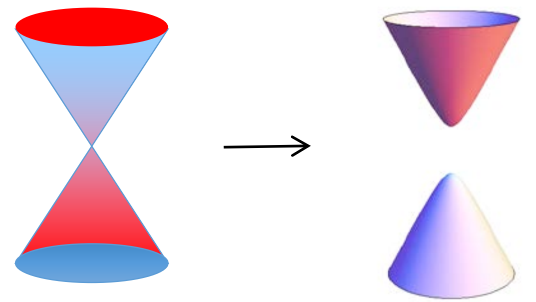
Quantum Anomalous Hall Effect

- Topological insulators: Surface Dirac Fermions
- Breaking time-reversal with dopant magnetization, no B-field!
- Spin orbit coupling is crucial: Gives a **"Lorentz force"**



R. Yu, et al (Science 2010)

$(Bi,Sb)_2Te_3$ film doped with Cr or V

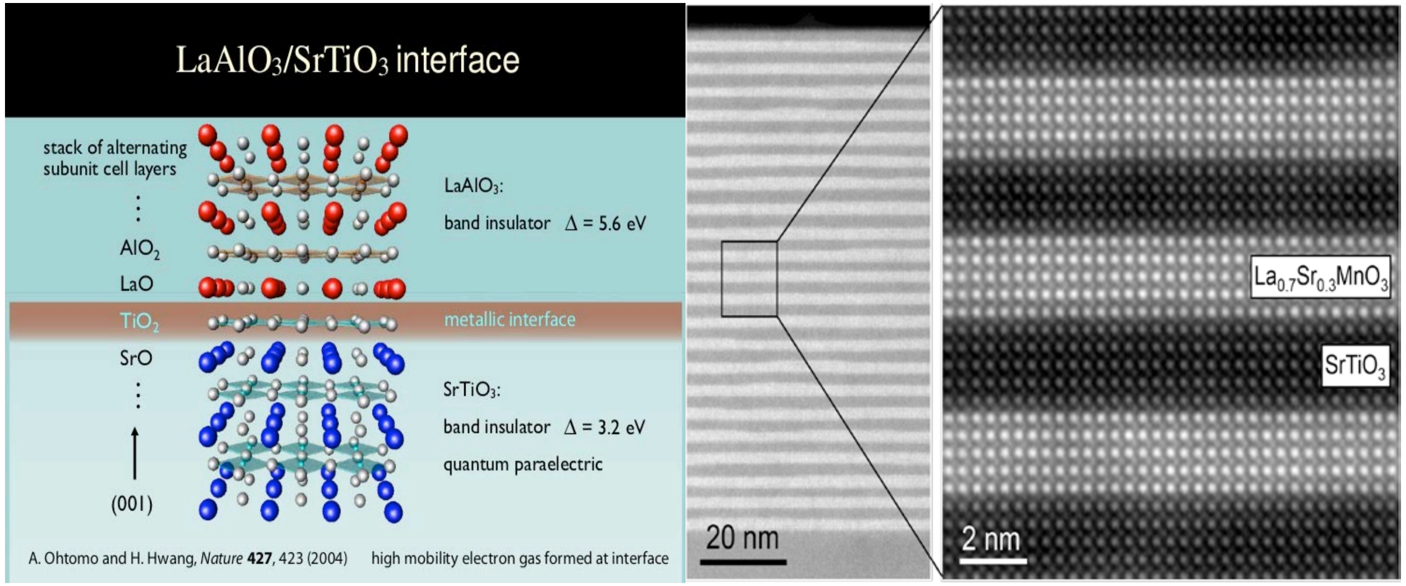


Surface Dirac fermions get gapped

Ferromagnetic Tc ~ 10K
Hall quantization: T ~ 25 mK

C.Z. Chang et al, Science 2013 (Xue group, Tsinghua)
C.Z. Chang et al, arXiv (M. Chan + J. Moodera groups)
A. J. Bestwick et al, arXiv (Goldhaber-Gordon group)
A. Kandala, et al, arXiv (N. Samarth group)

Oxide heterostructures



A. Ohmoto, H. Hwang, Nature (2004)

Boost temperature scale for Quantum Anomalous Hall effect?

Multicomponent Superlattices

nature materials | LETTERS
PUBLISHED ONLINE: 22 JANUARY 2012 | DOI: 10.1038/NMAT3224

Exchange bias in $LaNiO_3$-$LaMnO_3$ superlattices

Marta Gibert[1]*, Pavlo Zubko[1], Raoul Scherwitzl[1], Jorge Íñiguez[2] and Jean-Marc Triscone[1]

Triscone Group (Geneva)
M. Gibert (Zurich)

AIP | Applied Physics Letters

Local electronic and magnetic studies of an artificial La 2 FeCrO 6 double perovskite

Benjamin Gray, Ho Nyung Lee, Jian Liu, J. Chakhalian, and J. W. Freeland

Citation: Applied Physics Letters 97, 013105 (2010); doi: 10.1063/1.3455323

Jak Chakhalian group (Rutgers)

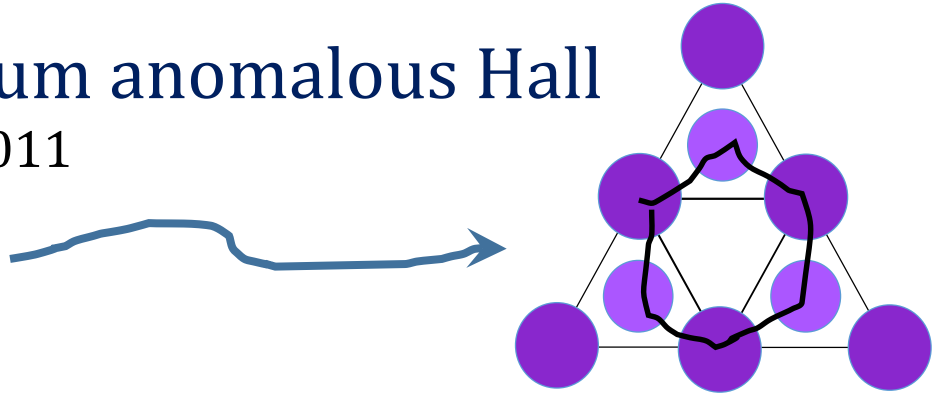
H. Takagi group (APL 2015)
SrIrO3/SrTiO3

ORNL/UTK/ANL (H.N. Lee)
SrIrO3/SrMnO3 – large AHE
Nat Comm. 2016

Topological Bands in (111) Heterostructures

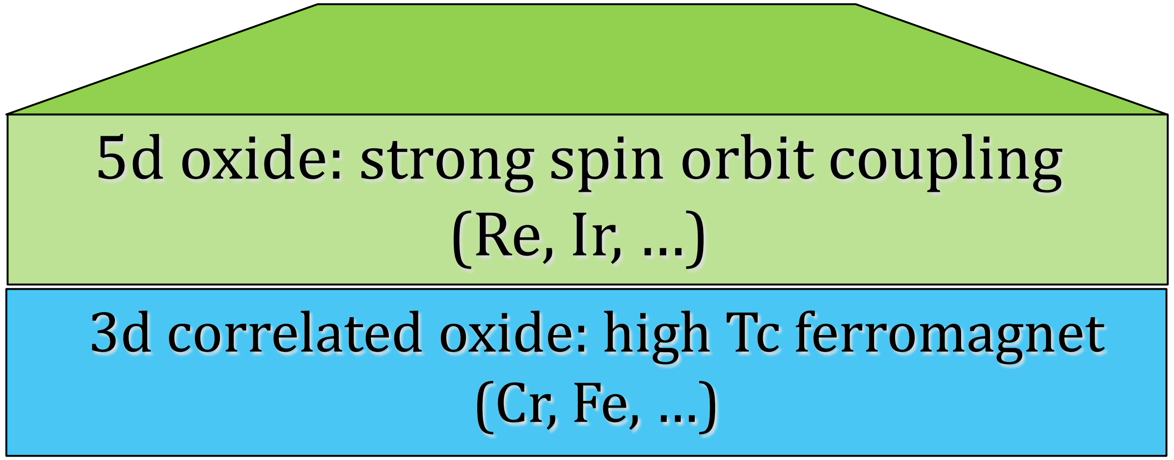
Transition metal oxide bilayers: Quantum anomalous Hall

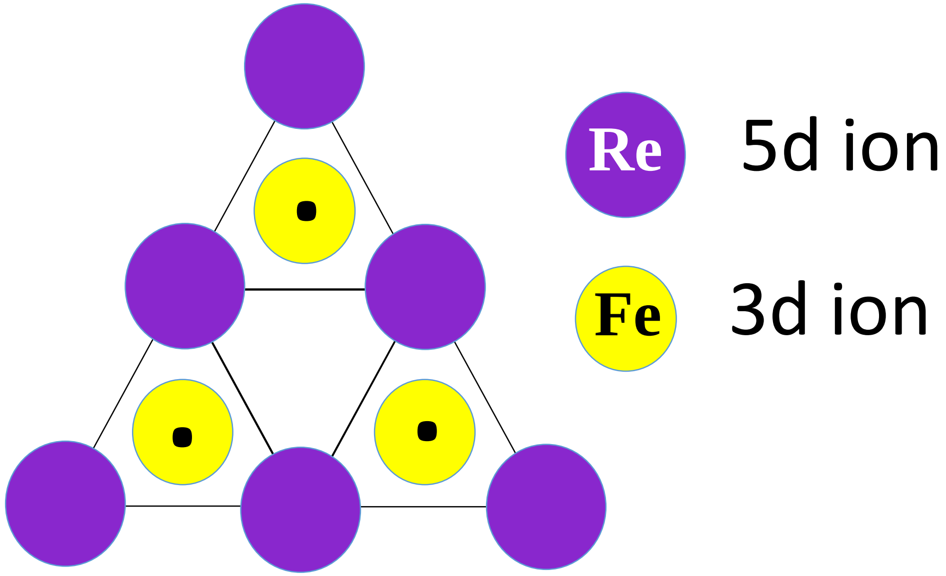
- D. Xiao, Zhu, S. Okamoto, Y. Ran, Nagaosa, Nat Comm 2011
- G. Fiete group, PRB 2011, PRL 2013
- K. Held group, PRL 2017 ($SrRuO_3$ bilayers)



Multicomponent bilayers

Predict high Tc QAH effect

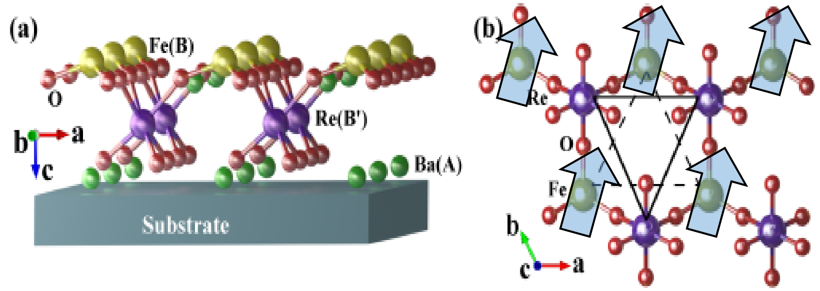




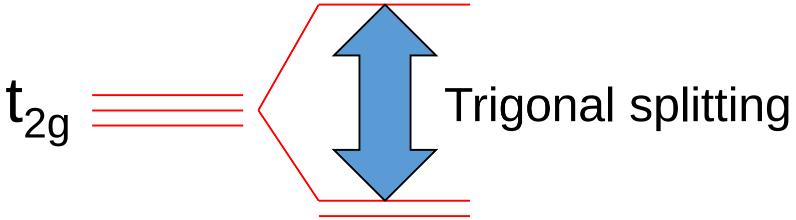
A.M. Cook, **AP** (PRL 2014); K. Garrity, D. Vanderbilt, PRB (2014);
S. Baidya, U.Waghmare, **AP**,T.Saha-Dasgupta (PRB 2015+16)

“Multicomponent” (111) bilayer

- Triangular half-metal

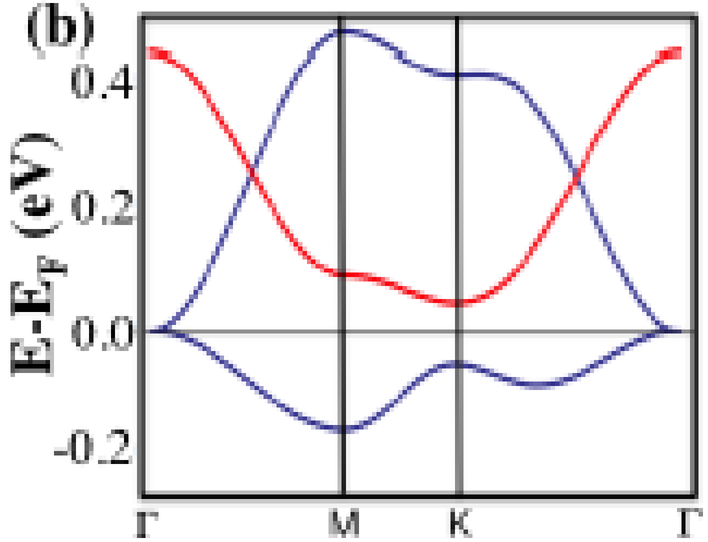


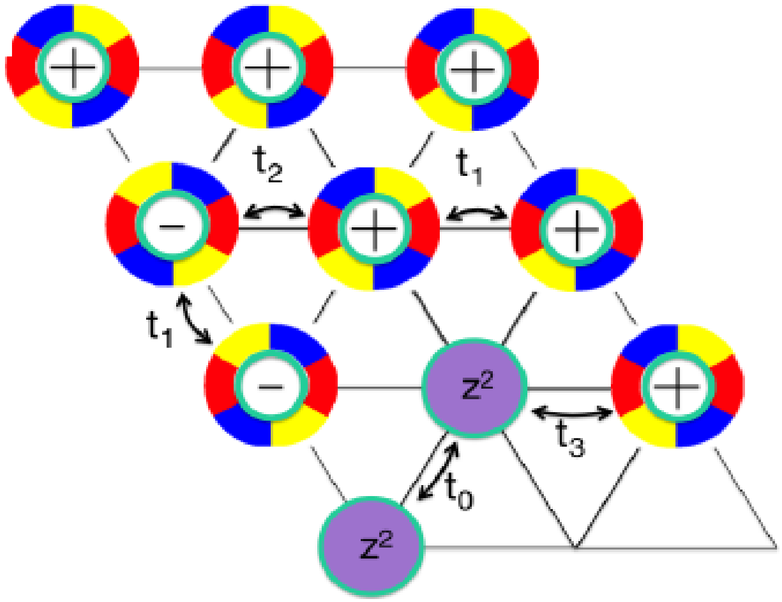
$L_z = 0 : \; d_{3z^2-r^2}$



$L_z = \pm 2 : d_{x^2-y^2} \pm i d_{xy}$

Non-Kramers doublet





Quadratic band touching at Γ: C3 + “Time reversal”

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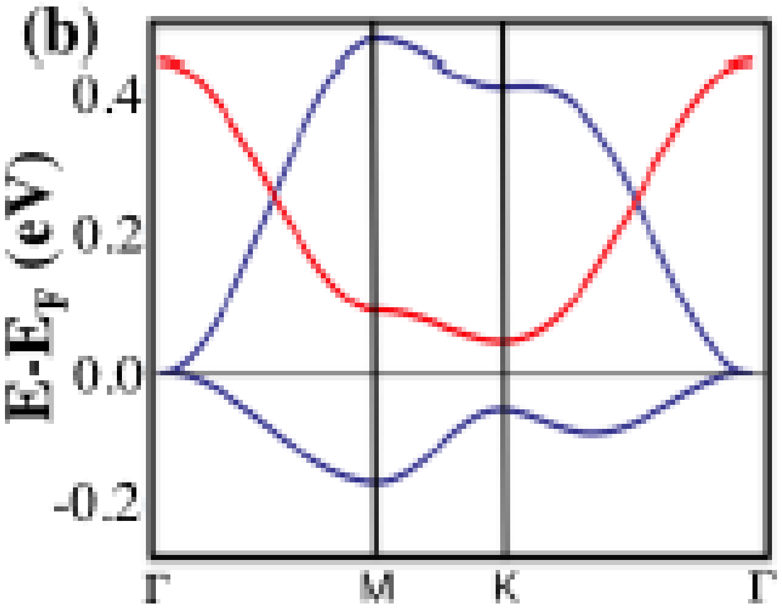
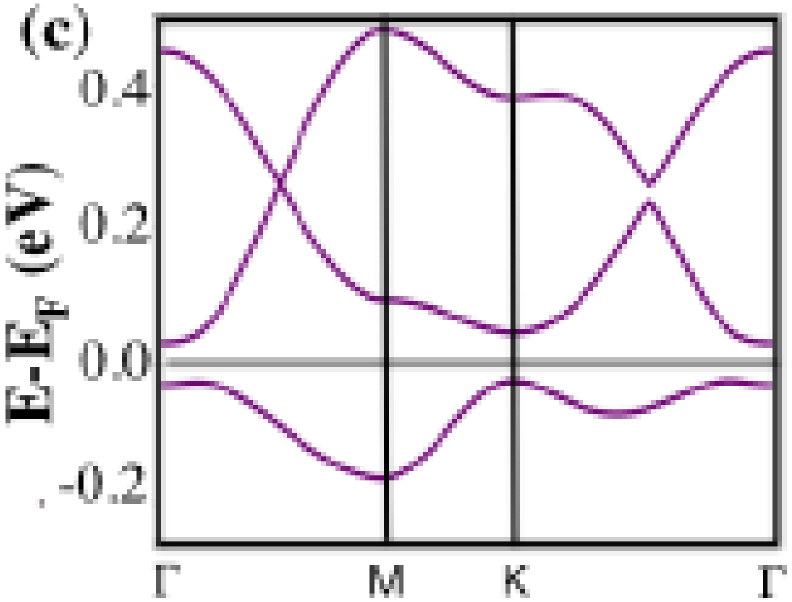
Large gap Chern insulator



SOC

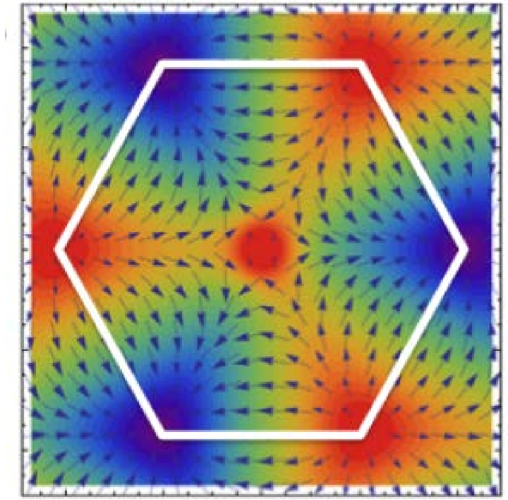
$$\xrightarrow{}$$

$$\lambda L_z \langle S_z \rangle$$



DFT: QAH gap ~ 100 meV

Monte Carlo: Ferromagnetic Tc ~ 250 K



k-space skyrmion

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Summary

- **Double Perovskites: Lego-Blocks with Strong SOC**

- **RIXS: Can be used to extract SOC and Hund’s Coupling**
- **Iridates, Rhenates, Osmates: Hund’s coupling $<$ SOC**

- **Lego Assembly: FCC Mott Insulators [Strong Magnetoelastic Effect]**
- **Lego Assembly: Chern Insulators [High Tc QAH Effect]**