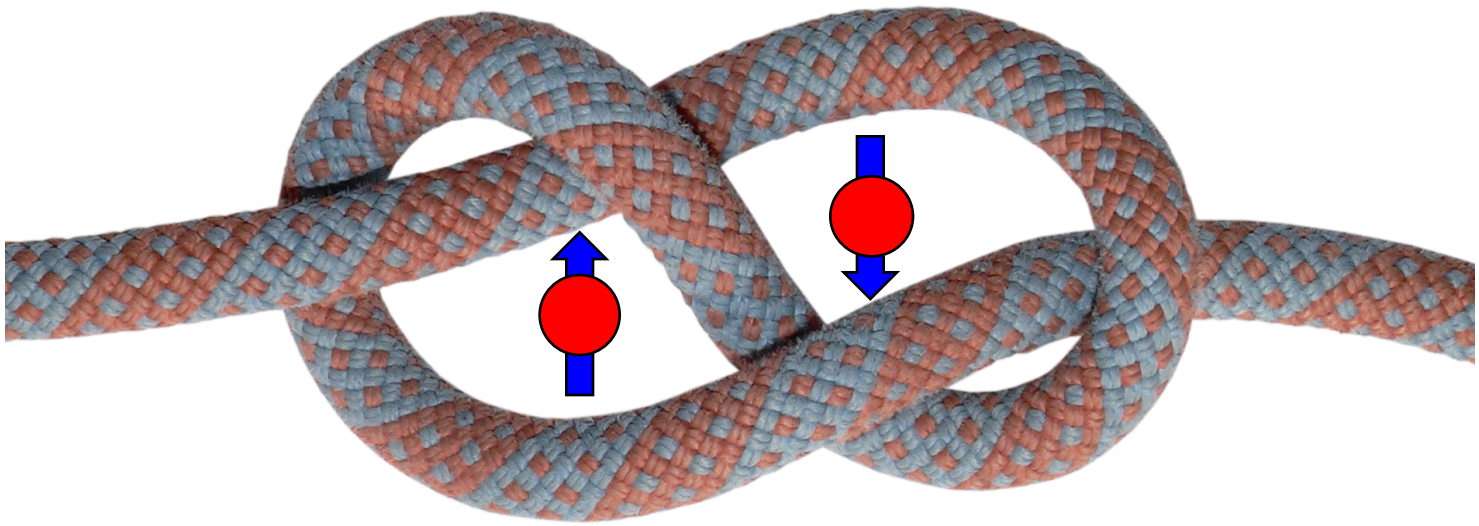
Oxide Heterostructures

A route to engineering correlated topological phases of matter

Arun Paramekanti
(University of Toronto)

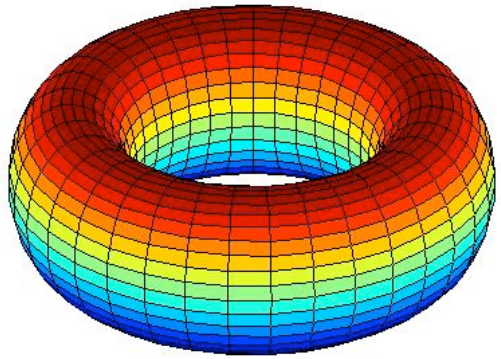


Multicomponent Superconductors (Nordita, 22 July, 2016)

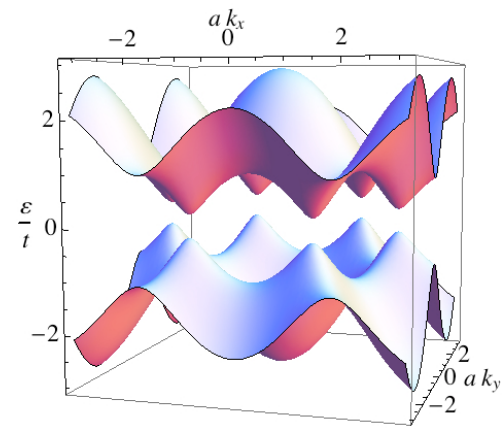
Funding: UNIVERSITY OF TORONTO, NSERC CRSNG, CIFAR CANADIAN INSTITUTE for ADVANCED RESEARCH

Bands in Crystals – Momentum Space Topology

Crystal momentum



2D Brillouin zone: Torus



k-space energy bands: $E_n(\mathbf{k})$

Bloch wavefunctions: $|\psi_n(\mathbf{k})\rangle$

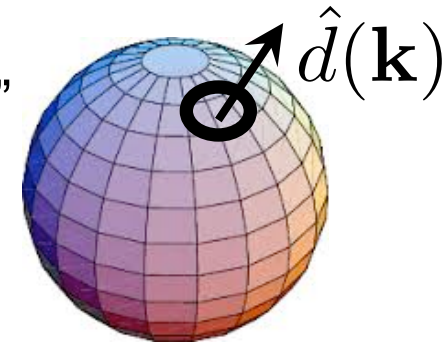
Two-Band System

$$H(\mathbf{k}) = \vec{d}(\mathbf{k}) \cdot \vec{\sigma}$$

pseudospin

$$E_\pm(\mathbf{k}) = \pm|\vec{d}(\mathbf{k})|$$

"Bloch sphere"



$\hat{d}(\mathbf{k})$: Information about wavefunction

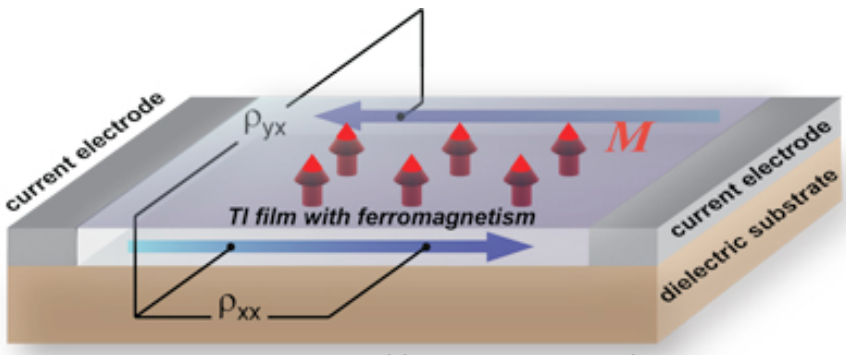
$$\boxed{\int \frac{dk_x dk_y}{4\pi} \hat{d}(\mathbf{k}) \cdot \partial_x \hat{d}(\mathbf{k}) \times \partial_y \hat{d}(\mathbf{k})}$$

→ Topological invariant (Chern number)

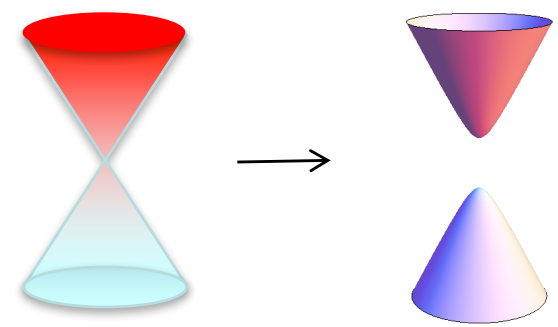
Sample boundary: Change of topology leads to gapless edge states

Quantum anomalous Hall effect
Magnetically doped topological insulators

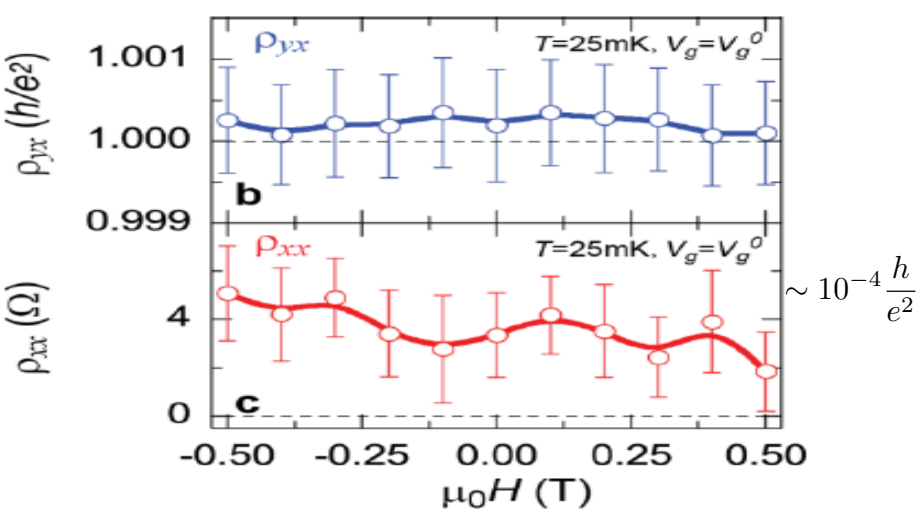
- Consider TIs near a band inversion transition (e.g., thin films)
- Breaking time-reversal strongly with dopant magnetization: QAH effect
- Spin orbit coupling is crucial



R. Yu, et al (Science 2010)



Surface Dirac fermions get gapped



$(Bi,Sb)_2Te_3$ film doped with Cr or V atoms
Ferromagnetic Tc ~ 10-15K

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

- **How can we get a "high temperature" QAH effect?**

Outline

- Strong correlations lead to a high temperature scale for magnetism
- But strong correlations also leads to Mott localization!

- How can we bring together strong correlations and band topology?
  - most natural in Kondo type systems (two-fluid picture)

- Can Kondo-type materials display “high temperature” QAH effect?
- Does the correlations+topology interplay lead to exotic phases?

Collaborators

Ashley Cook
(PhD:Toronto -> U. Zurich)

Ciaran Hickey
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Lukasz Cincio
(LANL)

Zlatko Papic
(Leeds)

- **Toronto: K. Plumb, J.P. Clancy, Young-June Kim**
- **SNU: B.-C. Jeon, T.-W. Noh**
- **ORNL: A.A. Aczel, G. Cao, T. J. Williams, S. Calder, A. Christianson, D. Mandrus. A. Kolesnikov**
- **India: Santu Baidya, Tanusri-Saha Dasgupta, Umesh Waghmare**

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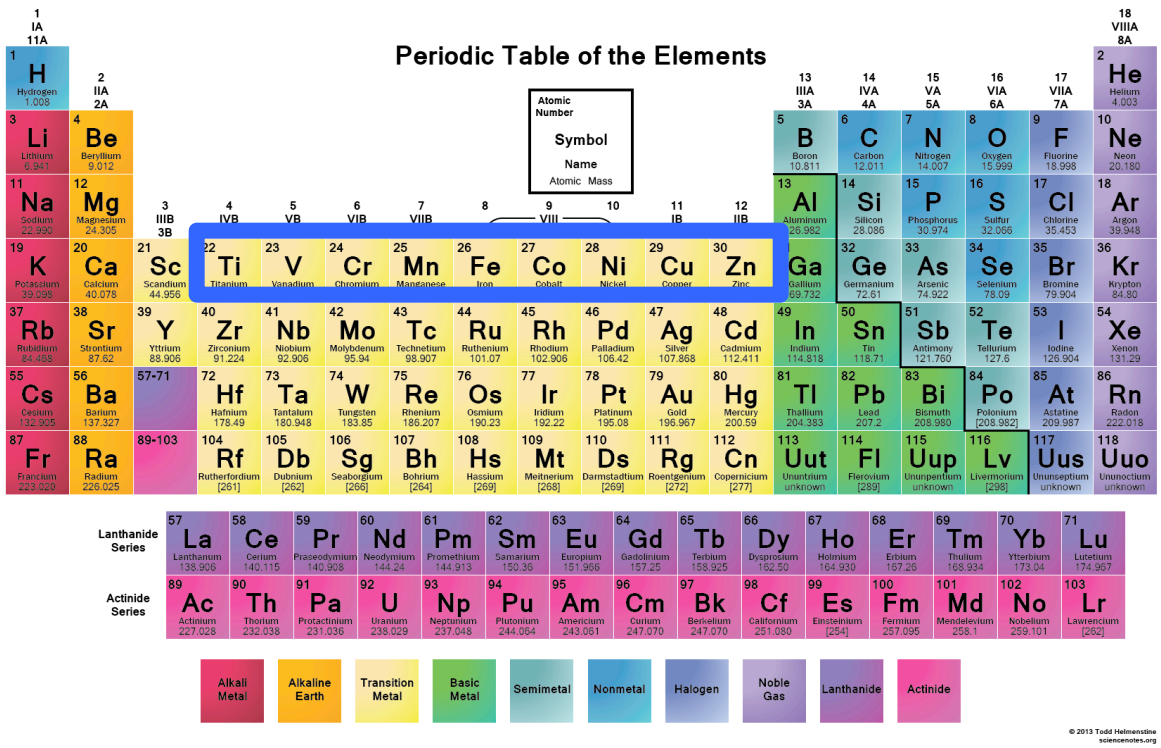
(1) Strong correlations

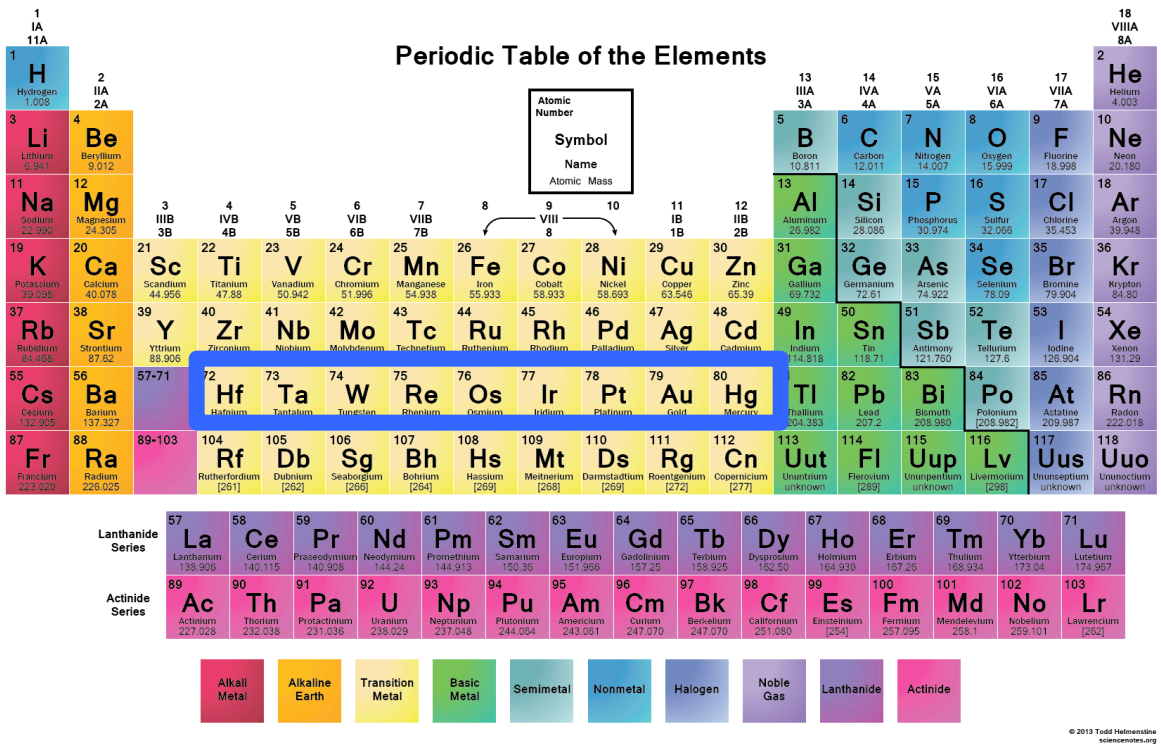
- Brink of Mott localization or deep Mott insulator regime
- Common in 3d oxides: Kinetic energy ~ Interactions
- “High” energy/temperature scales for correlations/magnetism



(2) **Band topology**

- Nontrivial band topology: SOC + conducting electron fluid
- Strong SOC needs heavy elements
- Expected in 4d/5d oxides (eg: Rhenium, Osmium, Iridium)

Increasing SOC

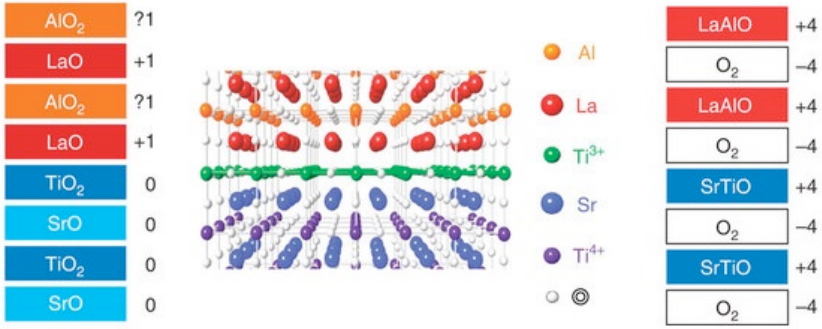


Engineering topology + correlations in solids

Oxide heterostructures and superlattices

LaAlO3/SrTiO3 interface [100]

H. Y. Hwang, J.M. Triscone, J. Mannhart,
R. Ashoori, K. A. Moler, ...
Basic physics: Magnetism + Superconductivity
Applications: Write/Erase circuits using electric field



3d-3d Superlattices along [111]

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]

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

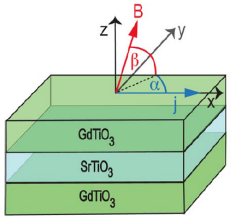
Citation: Applied Physics Letters **97**, 013105 (2010); doi: 10.1063/1.3455323

3d/5d Superlattices along [111]

H. Takagi group (APL 2015)

Quantum Wells, Modulation Doping

S. Stemmer group (UCSB)
Confined 2DEGs, high mobilities, magnetism



Prediction of topological insulators in simple TMO bilayers

ARTICLE

Received 20 Jun 2011 | Accepted 18 Nov 2011 | Published 20 Dec 2011 DOI: 10.1038/ncomms1602

Interface engineering of quantum Hall effects in digital transition metal oxide heterostructures

Di Xiao[1], Wenguang Zhu[1,2], Ying Ran[3], Naoto Nagaosa[4,5] & Satoshi Okamoto[1]

Rapid Communication

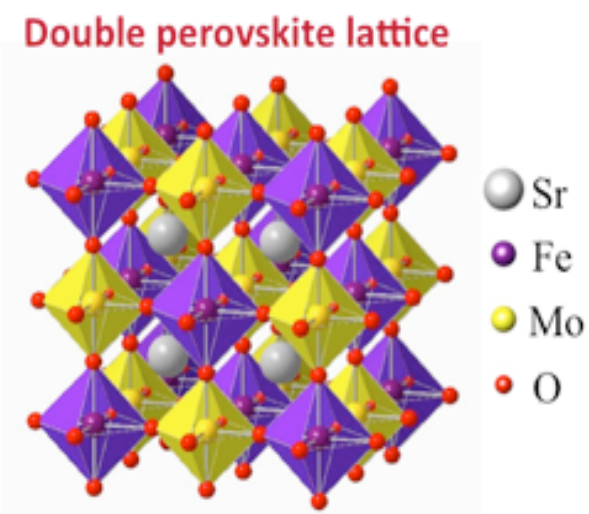
Topological insulators from complex orbital order in transition-metal oxides heterostructures

Andreas Rüegg and Gregory A. Fiete

Phys. Rev. B **84**, 201103(R) – Published 14 November 2011

Double Perovskites: Mixing correlations and SOC

General formula: $A_2BB'O_6$ (B,B' = 3d, 4d, 5d)



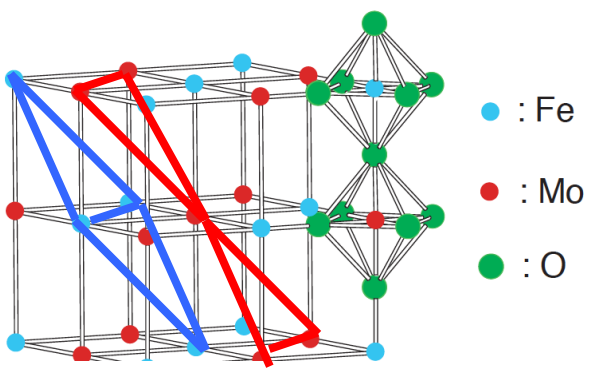
Metallic systems

B: Magnetism and B':Conduction electrons

. Half metallic ferrimagnets (eg:Sr_2FeMoO_6, T_C= 420K)

. Large polarization: good for spin injection

. Interplay of **Magnetism**, **SOC**, **Metallicity**



Layered along [111]

Mott insulators

B=magnetism, B'=inert or magnetism

. **Well isolated** TM-oxygen octahedra

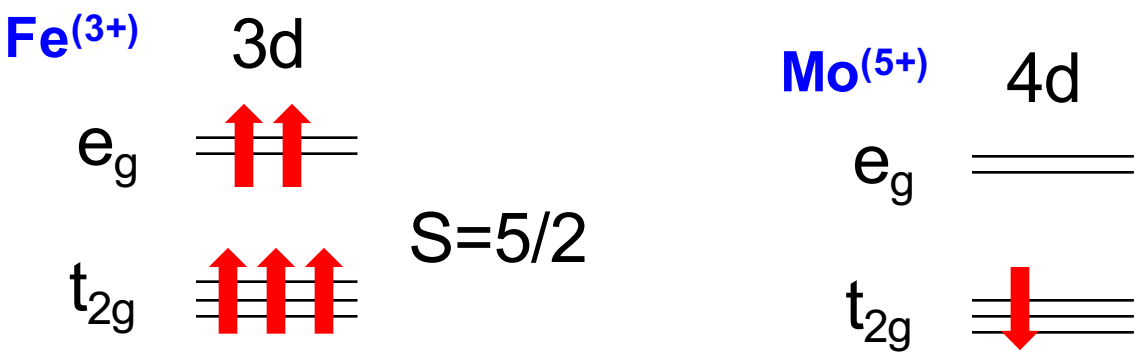
. **Frustrated** fcc lattice (eg: Ba_2YReO_6)

. Unusual spin-orbit coupled liquids?

. Insulating ferrimagnets (eg: Sr_2CrOsO_6, T_C=725K)

Single atom physics

Nominal valence: $Ba_2^{(2+)}Fe^{(3+)}Re^{(5+)}O_6^{(2-)}$
$Sr_2^{(2+)}Fe^{(3+)}Mo^{(5+)}O_6^{(2-)}$



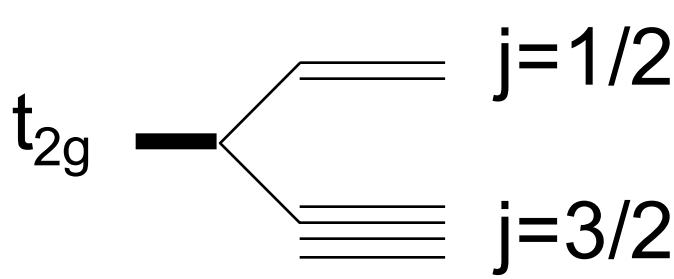
Hund's coupling > crystal field

SOC: λ ~100meV

Re: Spin orbit coupling in t_{2g} (L=1)

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

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



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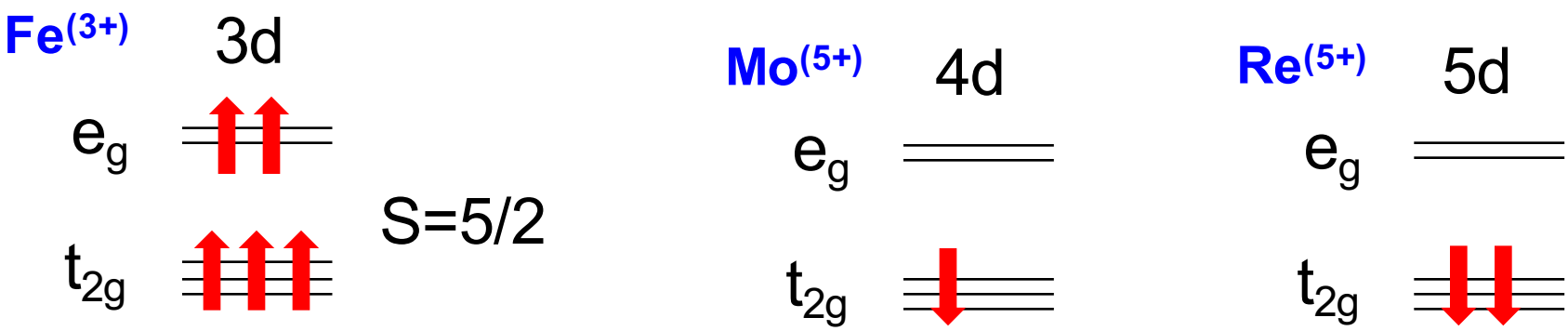


Hund's coupling > crystal field

SOC: λ ~100meV

SOC: λ ~ 500 meV
Hubbard U
important

Re: Spin orbit coupling in t_{2g} (L=1)

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

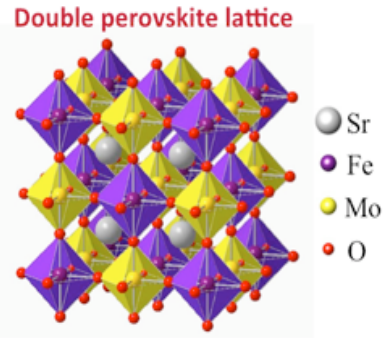
$$H_{\text{s.o.}} = -\lambda\vec{\ell}\cdot\vec{s}$$

Re: Interactions in t_{2g}

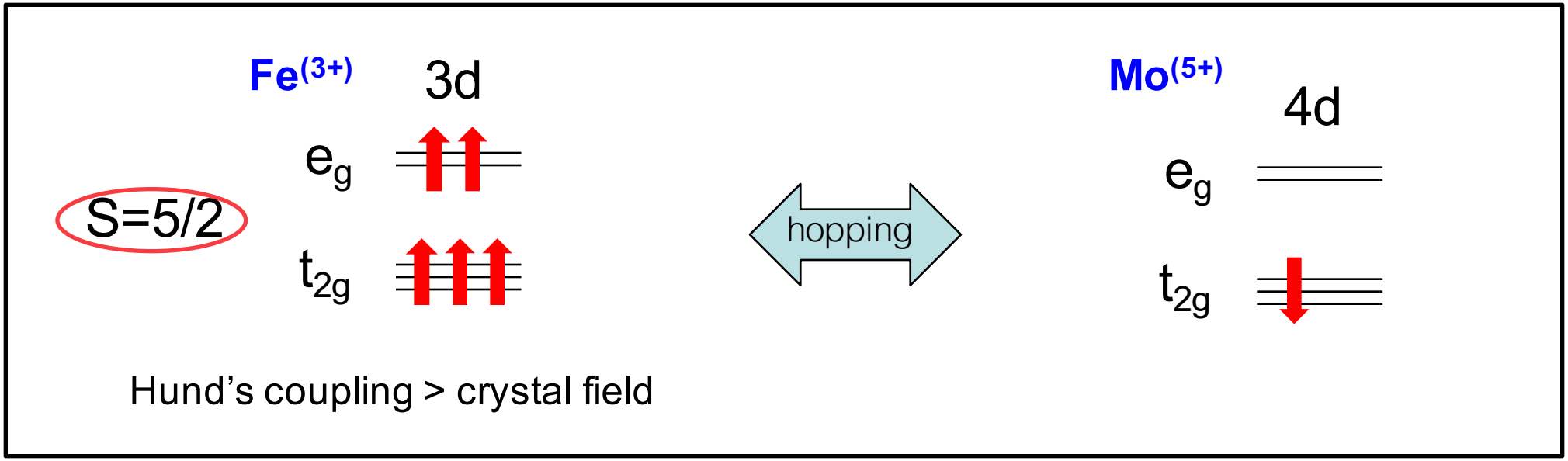
$$H_{\text{int}} = U\sum_{\alpha} n_{\alpha\uparrow}n_{\alpha\downarrow} + (U - 5\frac{J_H}{2})\sum_{\alpha<\beta} n_\alpha n_\beta$$
$$-2J_H\sum_{\alpha<\beta}\vec{S}_\alpha\cdot\vec{S}_\beta + J_H\sum_{\alpha\neq\beta} d^\dagger_{\alpha\uparrow}d^\dagger_{\alpha\downarrow}d_{\beta\downarrow}d_{\beta,\uparrow}$$

(Kanamori)

Double perovskites: Itinerant perspective

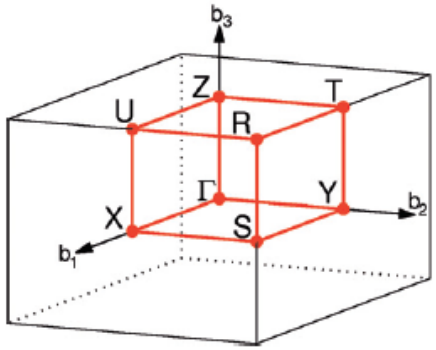


Origin of half-metallicity



D.D. Sarma, et al (PRL 2000); S. Di Matteo, G.Jackeli, N.Perkins (2003); G. Jackeli (PRB 2003); P. Sanyal, P. Majumdar (PRB 2009); O. Erten, O. Nganba-Meetei, M. Randeria, N. Trivedi, P. Woodward, (2011, 2013)

Electronic model for Ba_2FeReO_6: Hartree theory



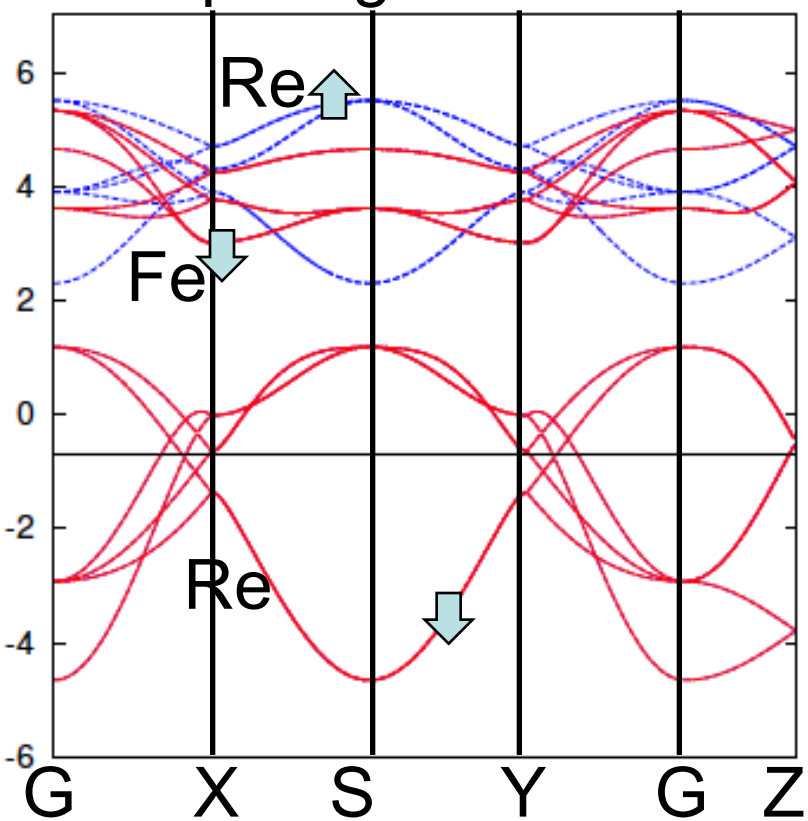
Correlation on Re: Stabilizes half-metallic state

Keep only intra-orbital U

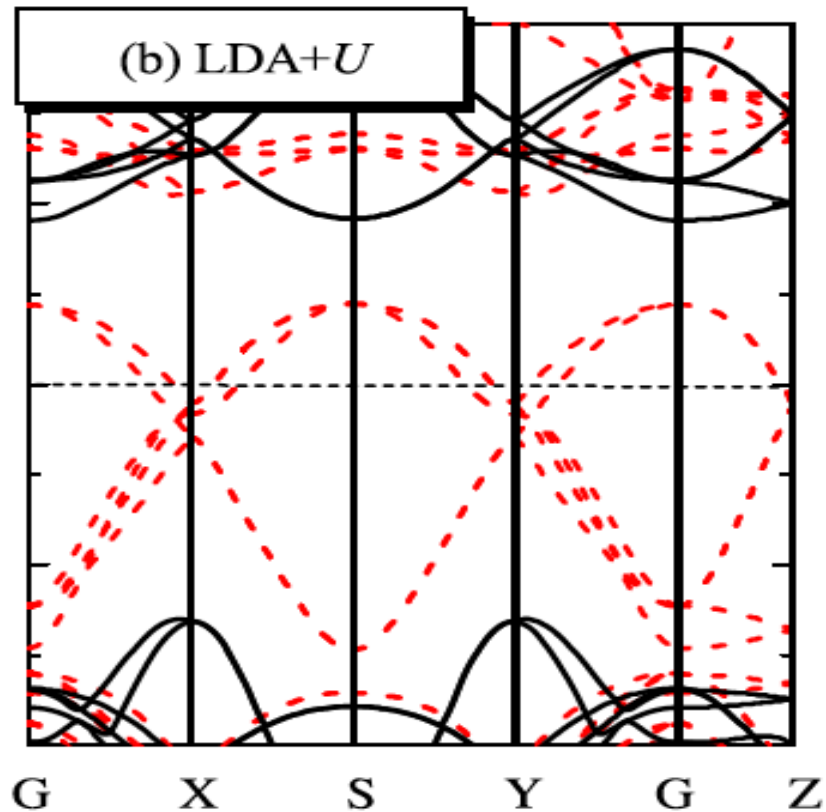
$t_{Fe\text{-}Re}$ ~ 330 meV, U ~ 2.5eV, Δ_{CT} ~ 1 eV

$t_{Re\text{-}Re}$ ~ 100 meV, Other hoppings small < 50 meV

Comparing Hartree-corrected dispersion with LDA+U



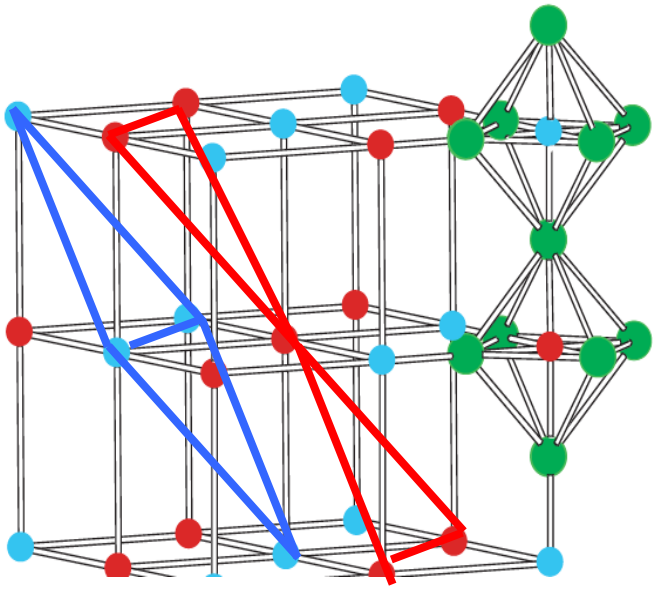
A. Cook, AP (PRB 2013)



B.C. Jeon, T.W. Noh, et al, (JPCM, 2010)

Ultrathin double perovskite films

- What if we dimensionally confine the half-metal to 2D?



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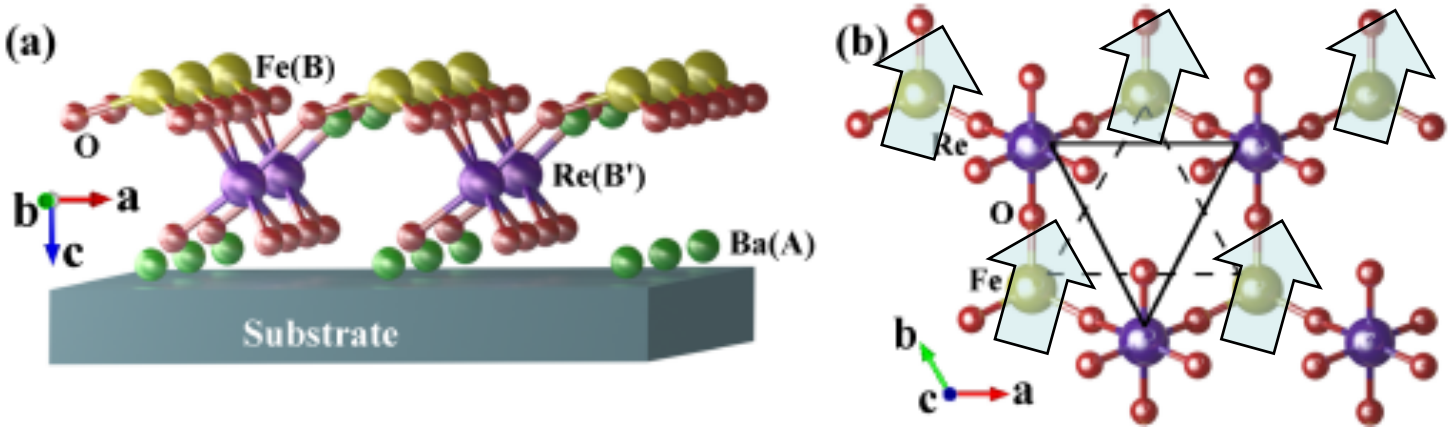
Citation: Applied Physics Letters **97**, 013105 (2010); doi: 10.1063/1.3455323

- Bilayer: Buckled honeycomb lattice

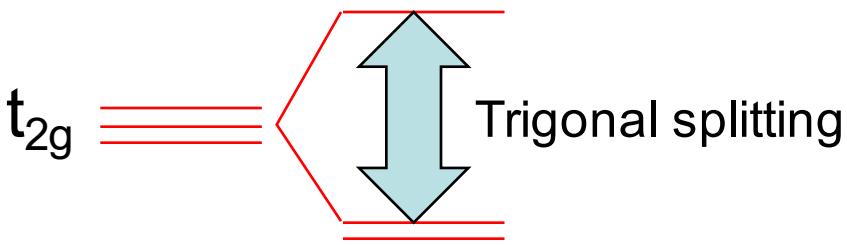


Re: Triangular lattice

Ultrathin double perovskite films

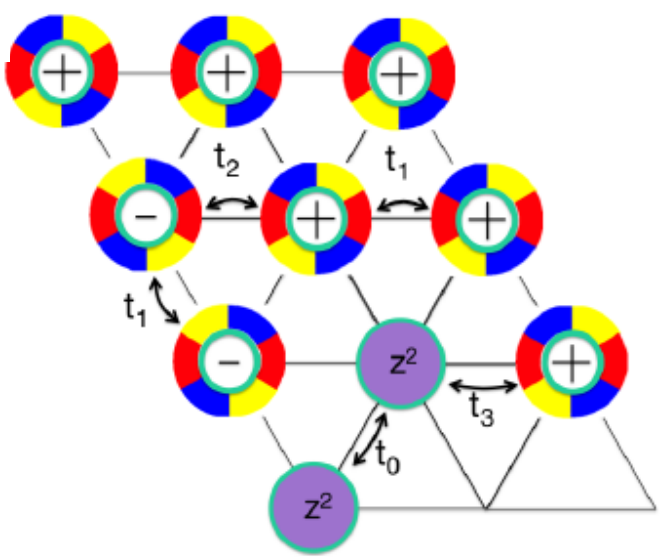
- Nominally Fe^{4+} ($3d^6$) and Re^{4+} ($5d^1$)
- Triangular half-metal

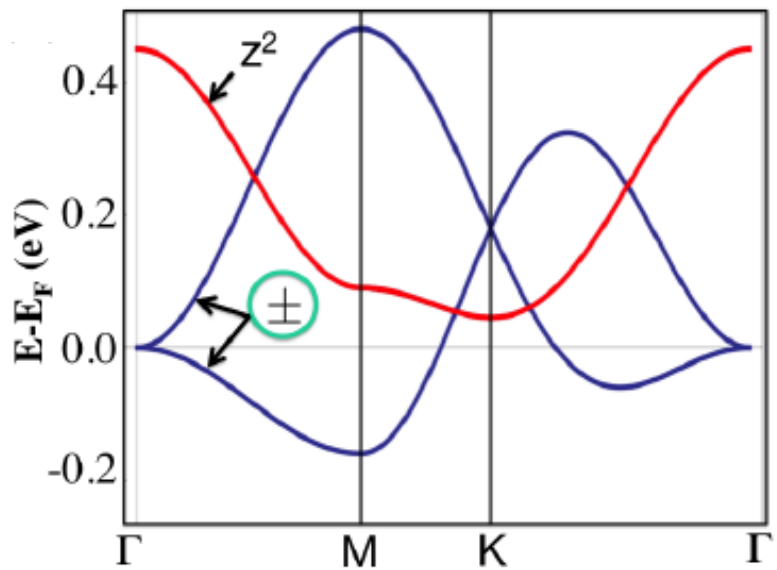
$$L_z = 0: \; d_{3z^2-r^2}$$



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

Non-Kramers doublet

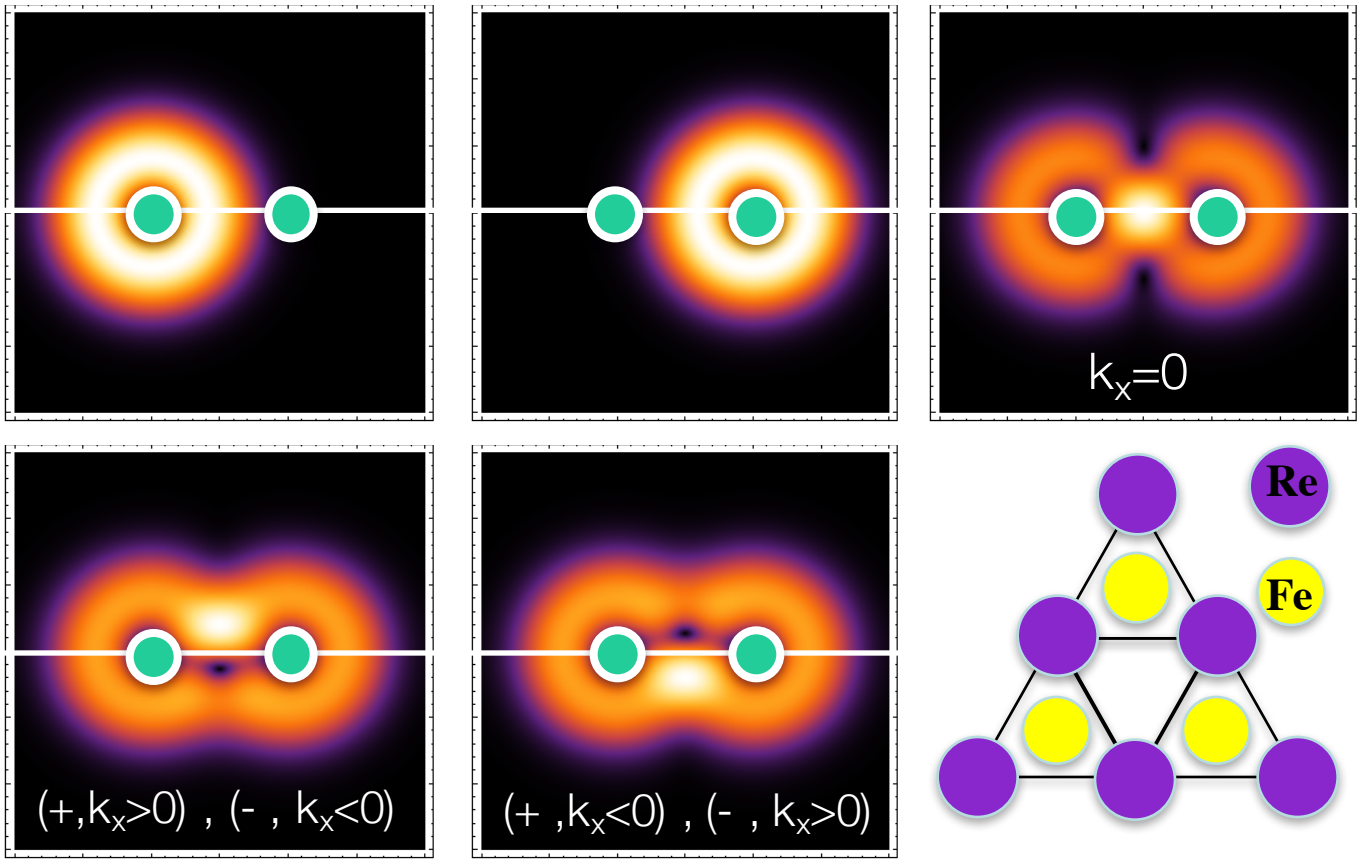




Dirac points at K: Inversion + "Time-reversal"
Quadratic band touching at Γ: C3 + "Time reversal"

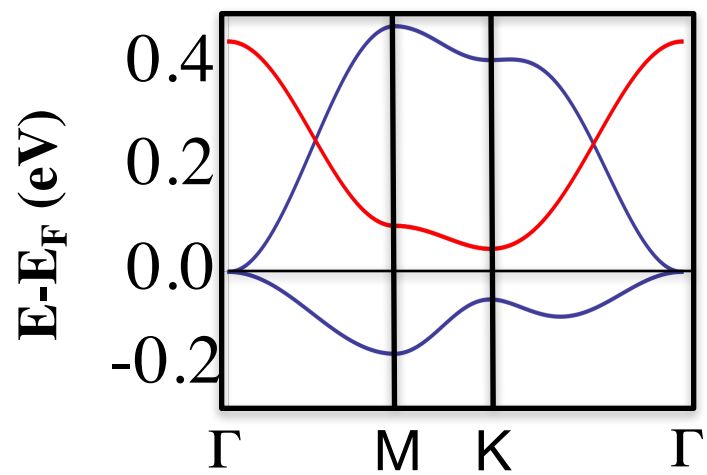
S. Baidya, U. Waghmare, AP, T. Saha-Dasgupta (to appear)

“Orbital Dipoles” and the “Orbital Rashba effect”

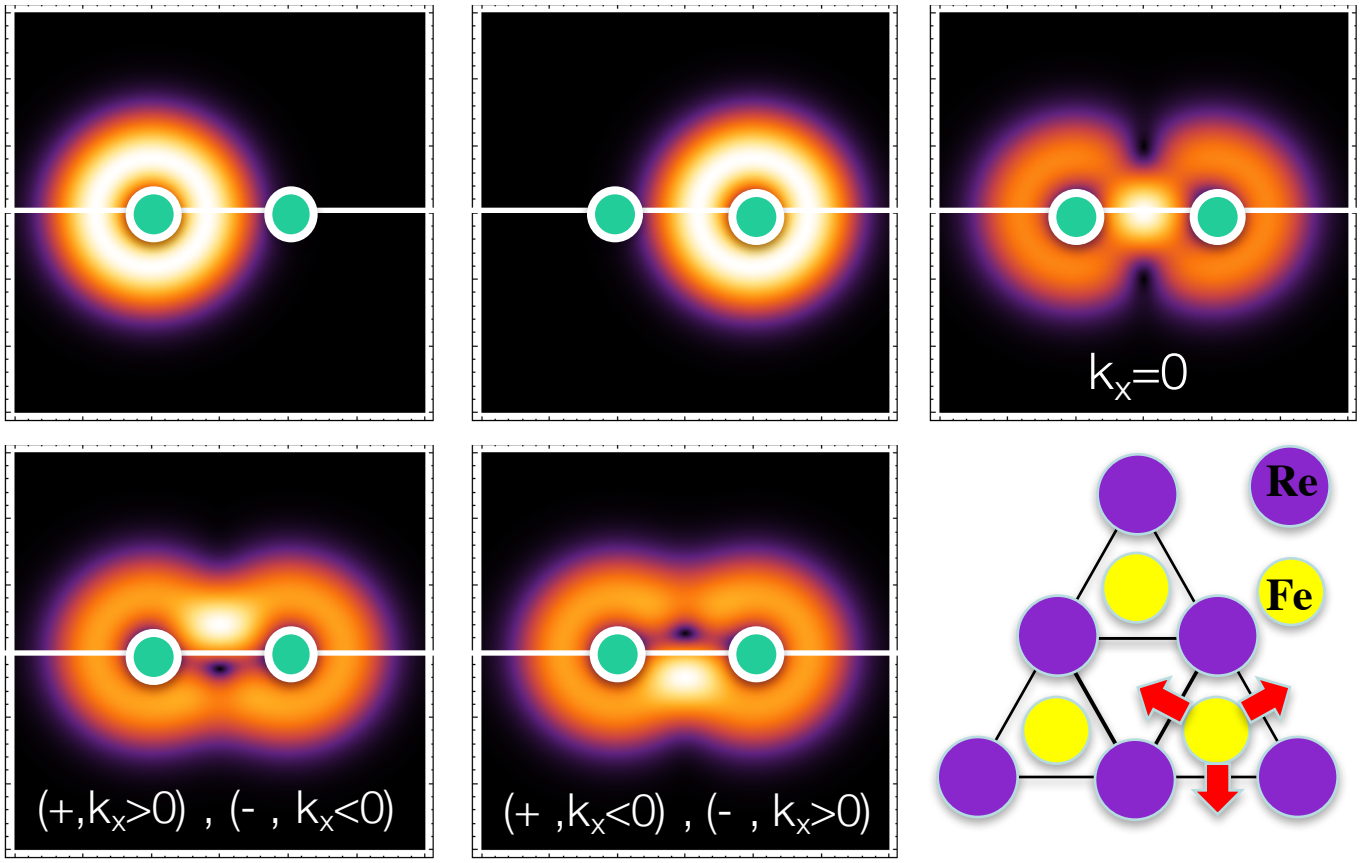


Inversion-symmetry breaking

- “Orbital Rashba” effect gaps out K Dirac point
- Half-semimetal



“Orbital Dipoles” and the “Orbital Rashba effect”



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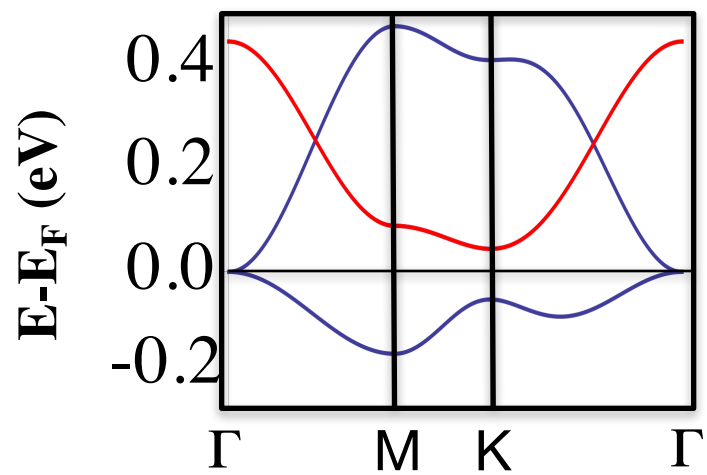
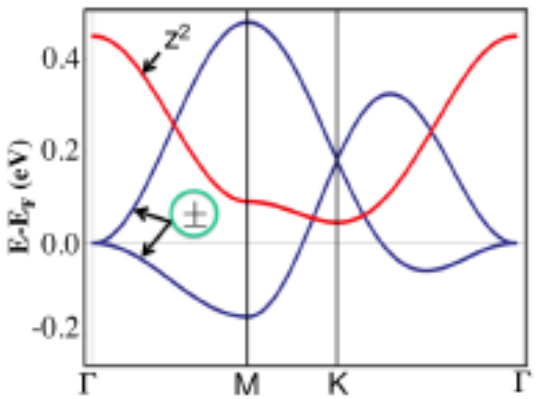
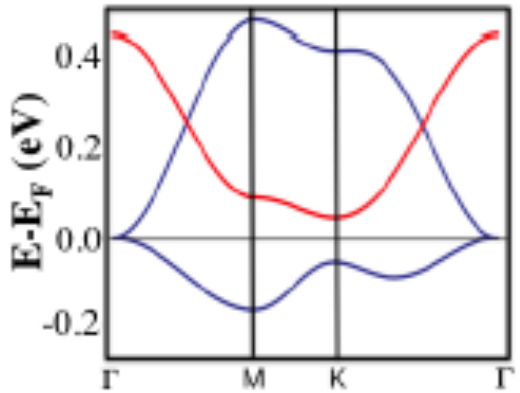


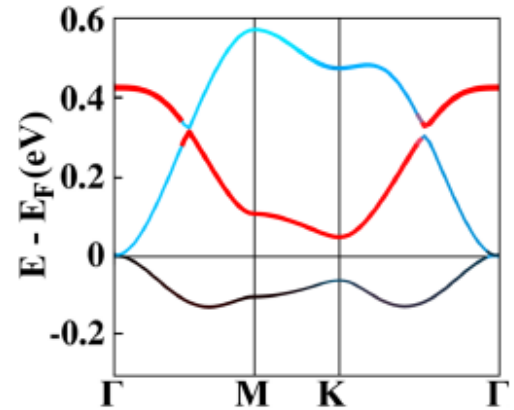
Large gap quantum anomalous Hall insulator



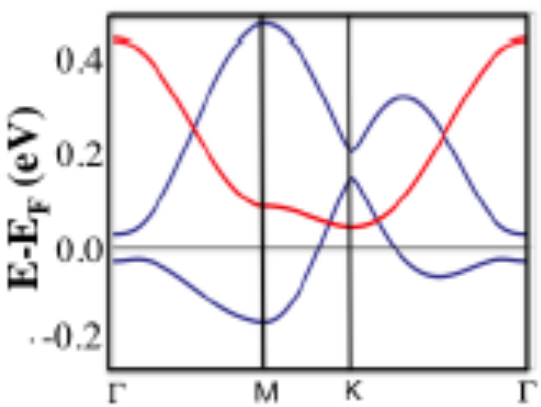
"Orbital Rashba" →



DFT calculations



↓ SOC



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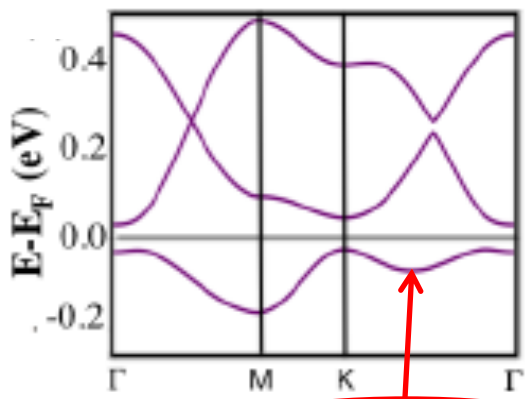
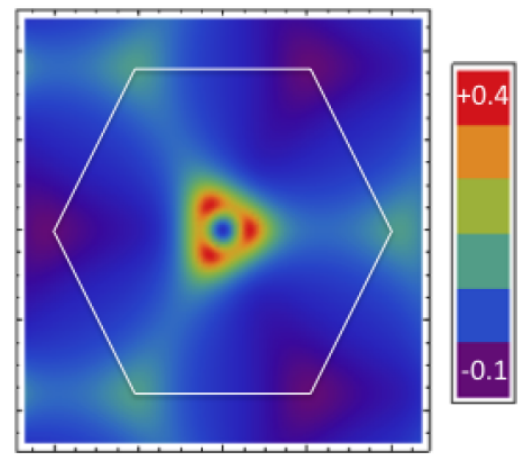


Chern band C=1



Berry curvature

QAH gap ~ 100 meV

Ferromagnetic Tc ~ 300 K

S. Baidya, U. Waghmare, AP, T. Saha-Dasgupta (to appear)

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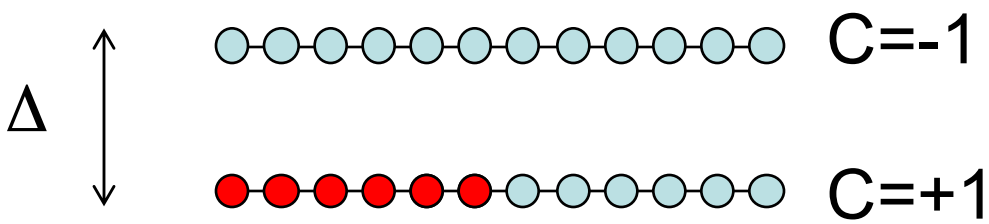
Exotic phases from topology + interactions

Correlations in Flat Chern Bands

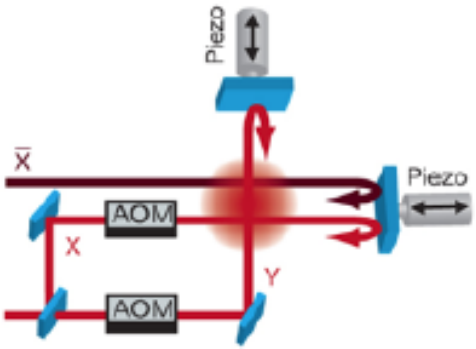


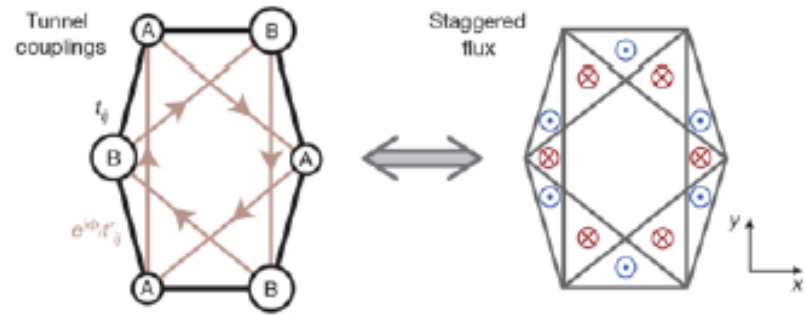
Bosons in nearly flat Chern bands ~ Landau levels

- Bandwidth W: W << Δ
- Interaction U: W << U << Δ
- ν=1/2 fractional QHE of bosons

D.N. Sheng, et al, Nat Comm 2011, PRL 2011, PRL 2012
T. Neupert, et al, PRL 2011

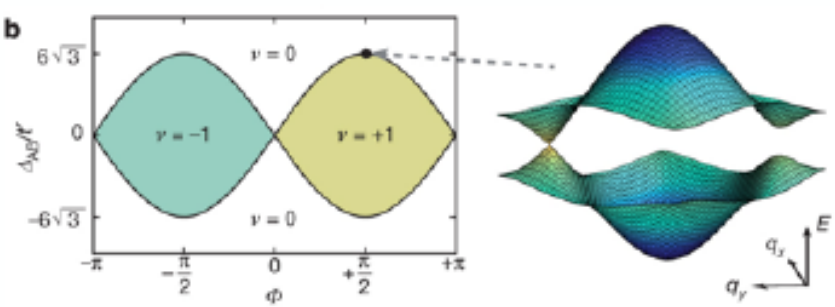
Chern bands for ultracold atoms: Atomic Haldane model



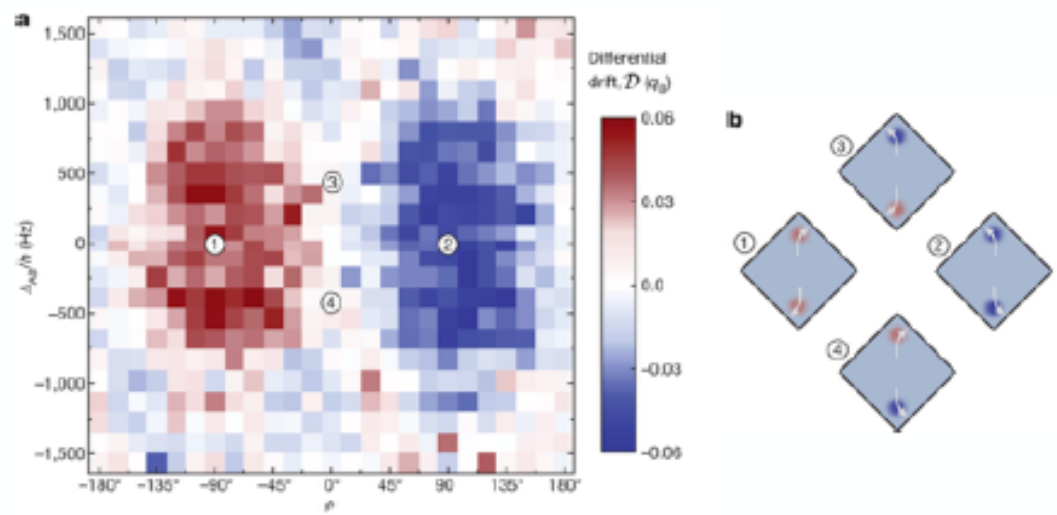


Periodic Lattice Shaking ⟷ Effective Staggered Flux Pattern

Second neighbor hopping = $t_2 e^{i\phi}$



Location of gap closing points measured using Bloch oscillations

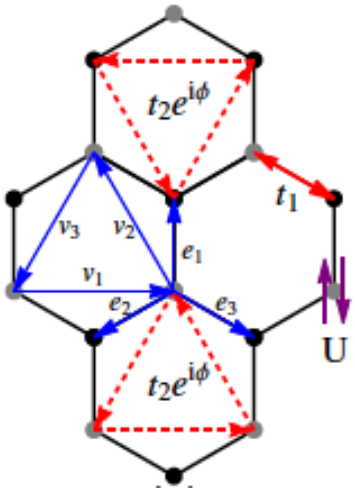


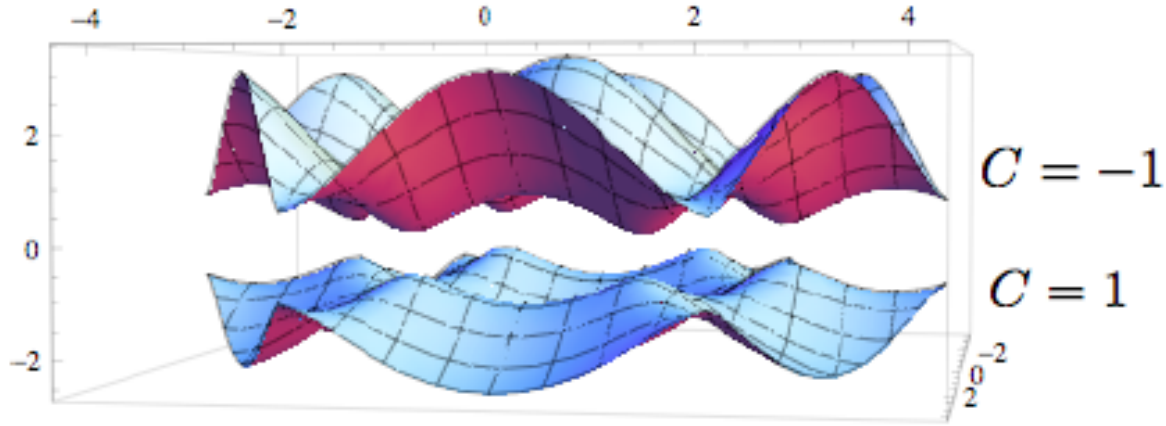
Berry curvature at Dirac points measured using differential drift

What happens with interactions?

Esslinger group: G. Jotzu, et al, Nature 2014

Equilibrium Haldane-Hubbard model of spin-1/2 fermions



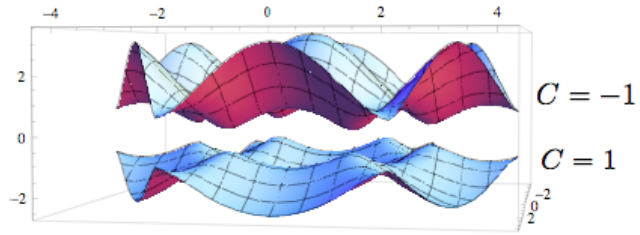


- Consider 2 flavors: $\uparrow, \downarrow$
- Fill up lower Chern band: $\sigma_{xy} = 2e^2/h$ ("2" due to spin)
- Local interactions between $\uparrow$ and $\downarrow$: "Haldane-Hubbard" model

$$H_{\rm HH} = -t_1 \sum_{\langle ij \rangle \sigma} (c^\dagger_{i\sigma} c_{j\sigma} + h.c.) - t_2 \sum_{\langle\langle ij \rangle\rangle \sigma} (e^{i\nu_{ij}\phi} c^\dagger_{i\sigma} c_{j\sigma} + h.c.) + U \sum_i n_{i\uparrow} n_{i\downarrow}$$

- What happens as we crank up interactions?
- What is the nature of magnetism in the Mott insulator?

Is the Mott Insulator an exotic spin liquid?



Mott insulator: View Spin $\uparrow$ = Boson, Spin $\downarrow$ = Empty

Chiral Spin Liquid = Bosonic Laughlin QH state

$$\Psi \sim \prod (z_i - z_j)^2 \qquad z \equiv \text{location of} \uparrow$$

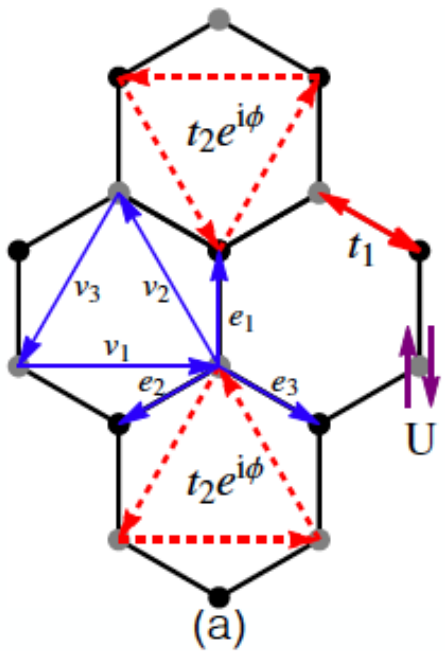
Theoretical proposals –

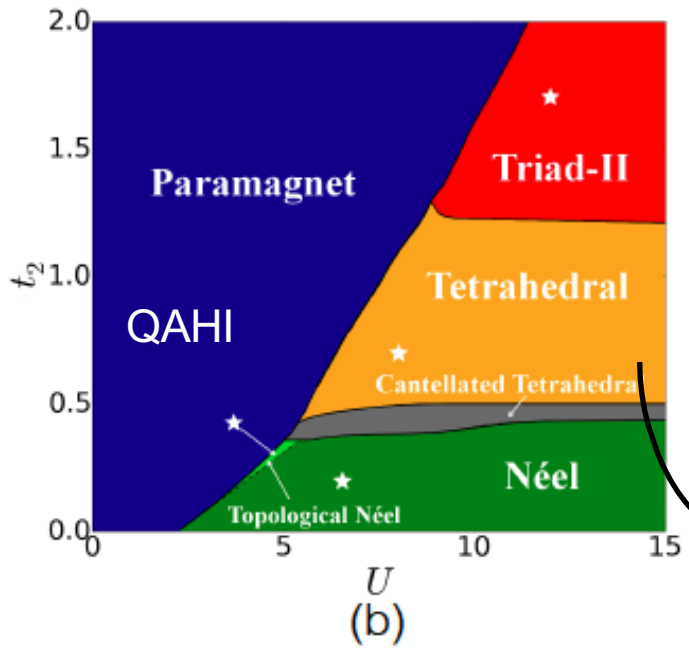
Kalmeyer, Laughlin (PRL 1987)
X. G. Wen, F. Wilczek, A. Zee (1989)
D. F. Schroeter, E. Kapit, R. Thomale, M. Greiter (PRL 2007, 2009)
Y. Zhang, T. Grover, A. Vishwanath PRB 2011
J. He, et al, PRB 2011, 2012 - Slave rotor mean field theory: Chiral spin liquid (ν=1/2 bosonic Laughlin state)

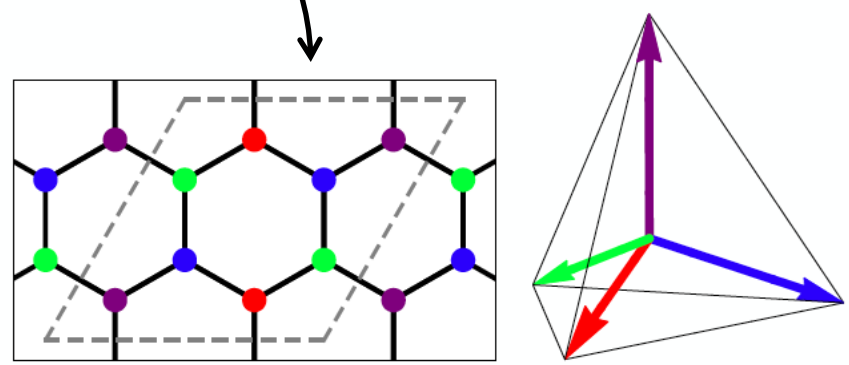
J. Maciejko, A. Ruegg, PRB 2013 - Exotic Z_2 CI* phase at intermediate U

D. Prychynenko, S. Huber arXiv:1410.2001 – Topological SDW order (with sublattice imbalance)

Hartree Mean Field Theory

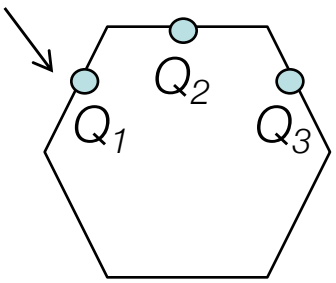






V.S.Arun, R. Sohal, C. Hickey, AP (PRB 2016)

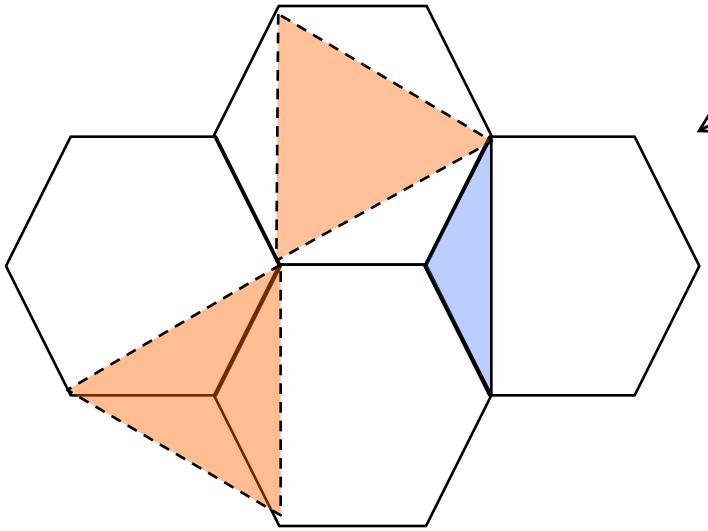
- Triple-Q noncoplanar state
- Large scalar spin chirality: “Skyrmion crystal”
- “Regular” Magnetic Order



L. Messio, C. Lhuillier, G. Misguich (PRB 2011)

Strong coupling limit: Chiral spin interactions

$$H_{\rm spin} = \frac{4t_1^2}{U}\sum_{\langle ij\rangle}\mathbf{S}_i\cdot\mathbf{S}_j + \frac{4t_2^2}{U}\sum_{\langle\langle ij\rangle\rangle}\mathbf{S}_i\cdot\mathbf{S}_j \quad \boxed{-\frac{24t_1^2t_2}{U^2}\sum_{\rm small-\triangle}\hat{\chi}_\triangle\sin\Phi_\triangle - \frac{24t_2^3}{U^2}\sum_{\rm big-\triangle}\hat{\chi}_\triangle\sin\Phi_\triangle}$$

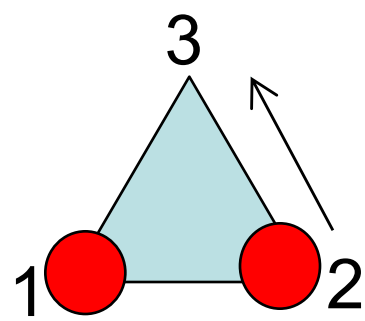


$$\chi_\triangle = \vec{S}_1\cdot\vec{S}_2\times\vec{S}_3$$

Scalar spin chirality

Boson language: Correlated hopping

$$in_1(b_2^\dagger b_3 - b_3^\dagger b_2)$$



Strong coupling limit: Chiral spin interactions

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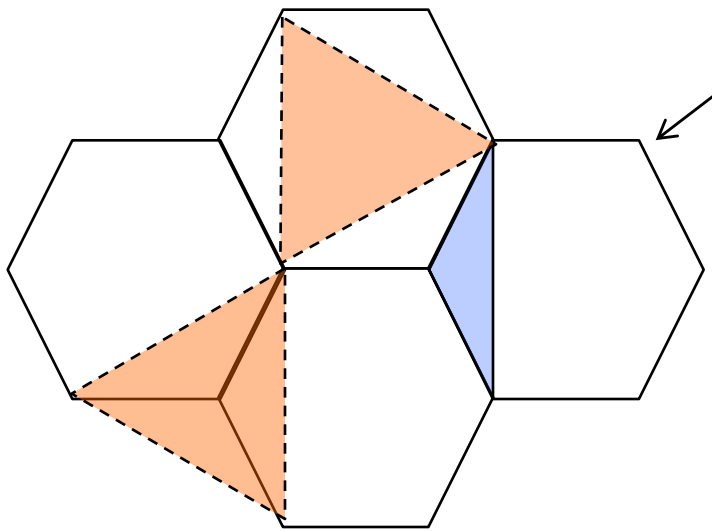
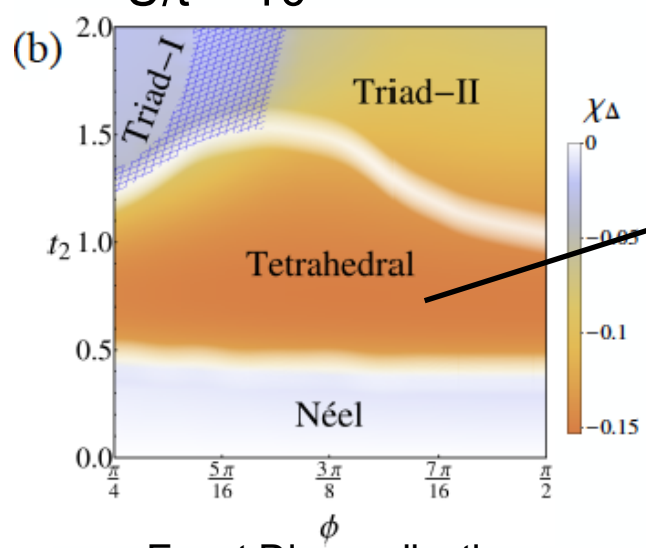
$$\chi_\triangle = \vec{S}_1\cdot\vec{S}_2\times\vec{S}_3$$

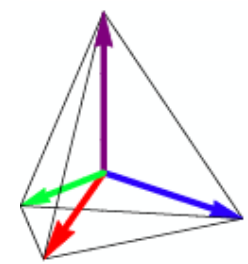
Scalar spin chirality



U/t = 10



Exact Diagonalization



Tetrahedral state
= Boson supersolid

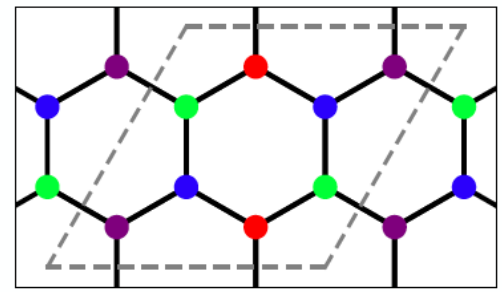
- Classical ground states
- Exact diagonalization
- Ground state: magnetically ordered!

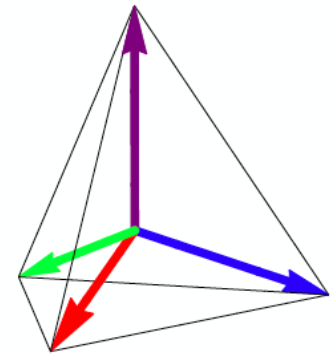
C. Hickey, L. Cincio, Z. Papic, AP (PRL 2016)

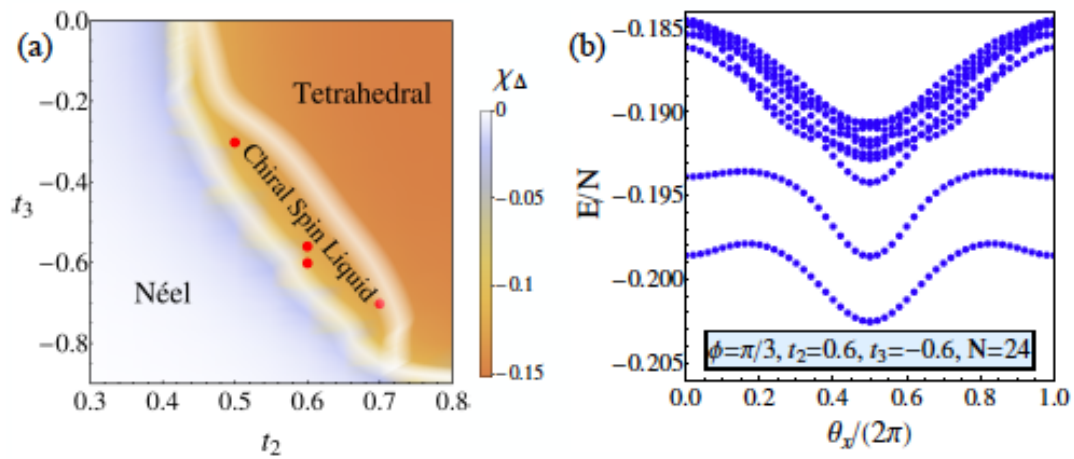
Cf: Nielsen, Sierra, Cirac, Nat. Comm. 2013 – Square lattice Chern insulator yields a CSL Mott insulator
Cf: B. Bauer, et al, Nature Communications 5, 5137 (2014) – Kagome chiral terms give CSL Mott insulator

Melting the Tetrahedral Order

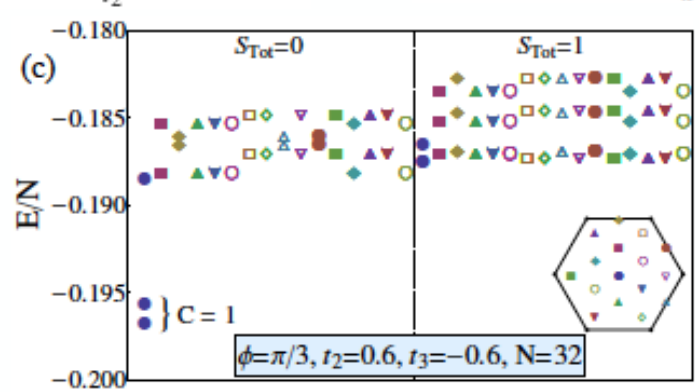
Add 3rd neighbor AFM interaction
Frustrates Tetrahedral order
Creates a CSL retaining large chirality!







Exact diagonalization results: N=18,24,32

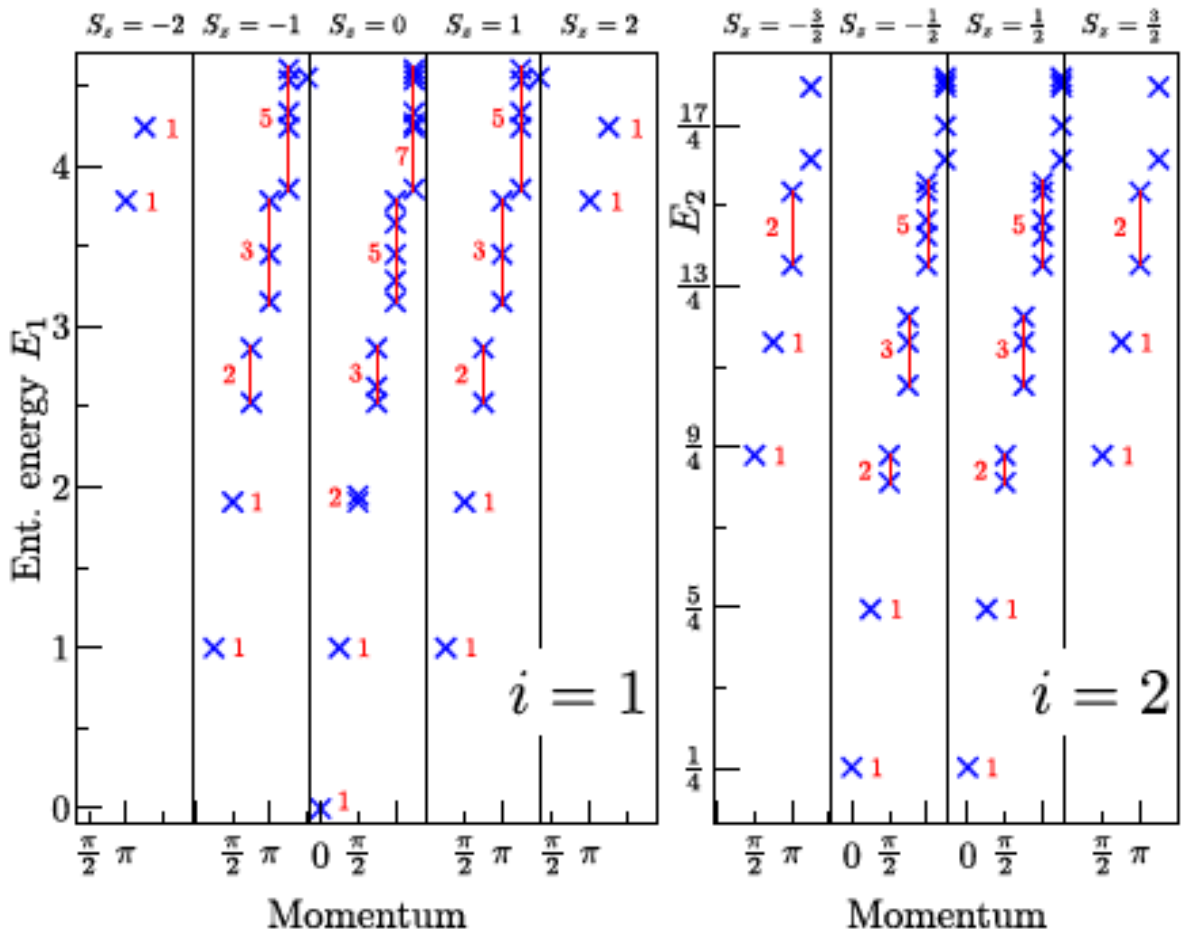


Bose supersolid -> FQH liquid

C. Hickey, L. Cincio, Z. Papic, AP (PRL 2016)

Infinite cylinder DMRG results

Y. Zhang, et al PRB 2013
L. Cincio and G. Vidal, PRL 2013

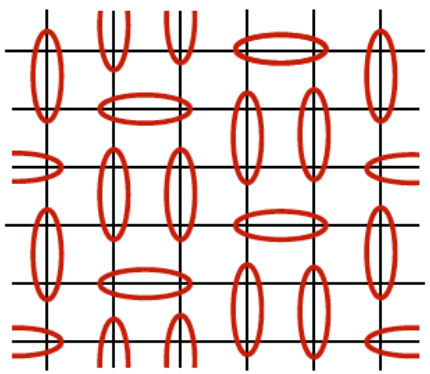


Agrees with chiral semion

C. Hickey, L. Cincio, Z. Papic, AP (PRL 2016)

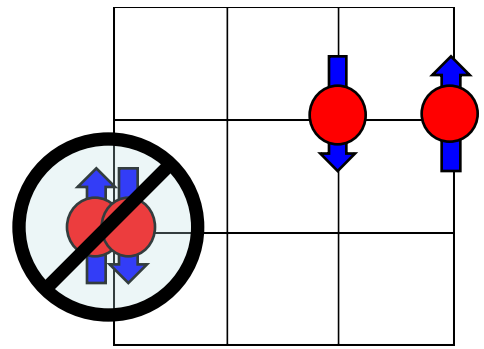
Routes to Quantum Spin Liquids

Resonating valence bonds
- Quantum dimer models
- Melting valence bond crystals

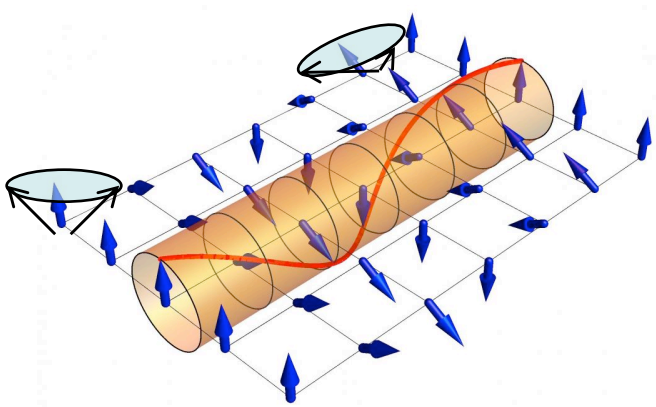


Gutzwiller projected band electrons

1 electron per site -> Variational spin wavefunction



Variational Monte Carlo studies or
slave particle gauge theory approaches



Disordering magnetically ordered states

Could there be a continuous Tetrahedral – Chiral Spin Liquid quantum phase transition?

Spin crystallization transition out of the CSL

- **Low energy theory of CSL: Pure topological Chern Simons theory**

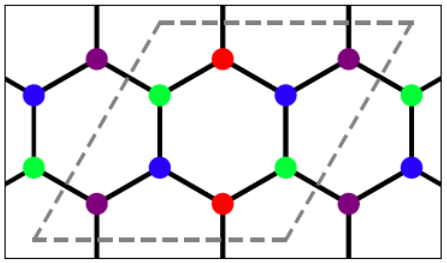
$$\mathcal{L}_{\rm CS} = \frac{1}{2\pi}\epsilon^{\alpha\beta\lambda} a_\alpha \partial_\beta a_\lambda$$

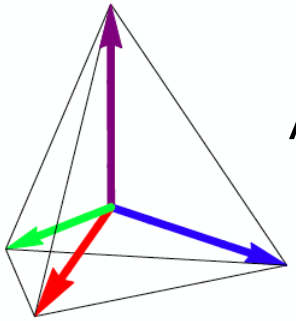
(Torus degeneracy=2)

- Need gapped excitations in CSL: Semions
- Like to examine continuous transition to a magnetically ordered state

Minimally couple bosonic spinons to CS gauge field

- In the CSL: Bosonic spinons bind π-flux to give semions
- Out of CSL: Condensing spinons can yield magnetic order



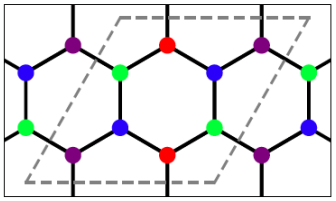


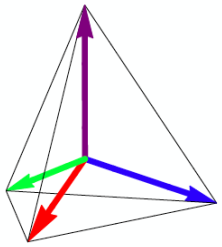
Adiabatic spinon transport: Berry Fluxes

Triangular loops: $\pi/2$

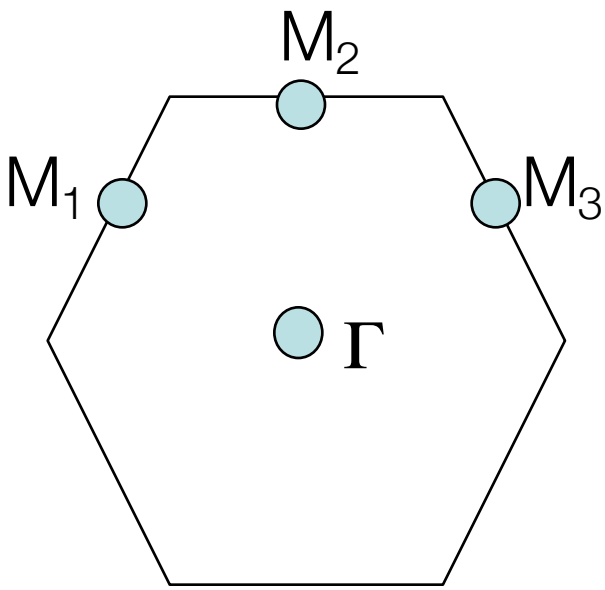
Hexagonal loops: π

Spin crystallization transition out of the CSL





Hofstadter problem
Triangular loops: $\pi/2$
Hexagonal loops: π



Dispersion minima

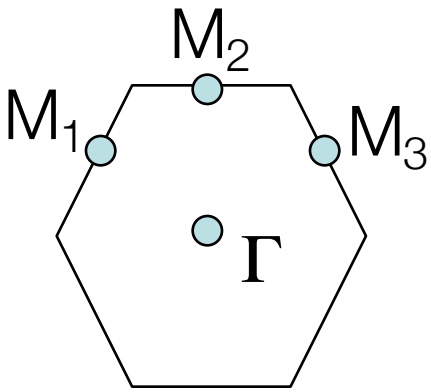
Suffices to focus on one Δ sublattice
Define spinon fields: $\phi_{i,\sigma}$

i = Mode label (1-4)
σ = Spin label

$$\mathcal{L}_{\mathrm{CS},\phi} = \frac{1}{2\pi}\epsilon^{\mu\nu\lambda} a_\mu \partial_\nu a_\lambda + \phi^*_{i\alpha}(\partial_\tau - i a_0)\phi_{i\alpha} + r|\phi_{i\alpha}|^2 + |(\vec{\nabla} - i\vec{a})\phi_{i\alpha}|^2$$

- Spinon condensates are “frustrated”: Lots of ways to condense into 4 minima
- Degeneracy will be broken by interaction effects

Spin crystallization transition out of the CSL



$$\mathcal{L}_{\mathrm{CS},\phi} = \frac{1}{2\pi}\epsilon^{\mu\nu\lambda} a_\mu \partial_\nu a_\lambda + \phi^*_{i\alpha}(\partial_\tau - i a_0)\phi_{i\alpha} + r|\phi_{i\alpha}|^2 + |(\vec{\nabla} - i\vec{a})\phi_{i\alpha}|^2$$

$$\rho_i = \phi^*_{i\alpha}\phi_{i\alpha}$$
$$\vec{\mathcal{S}}_i = \phi^*_{i\alpha}\vec{\sigma}_{\alpha\beta}\phi_{i\beta}$$

$u_2<0$ favors multimode

Fourth order interactions:

$$\mathcal{L}^{(1)}_{\mathrm{int}} = u_1\Big(\sum_i \rho_i\Big)^2 + u_2 \sum_{i\neq j} \rho_i \rho_j + u_3 \sum_{i\neq j} \vec{\mathcal{S}}_i \cdot \vec{\mathcal{S}}_j + u_4 \sum_{[ijk\ell]} \phi^*_{i\alpha}\phi^*_{j\beta}\phi_{k\alpha}\phi_{\ell\beta} + u_5 \sum_{i\neq j} \phi^*_{i\alpha}\phi^*_{i\beta}\phi_{j\alpha}\phi_{j\beta}$$

Sixth order interactions:

Favors tetrahedral order

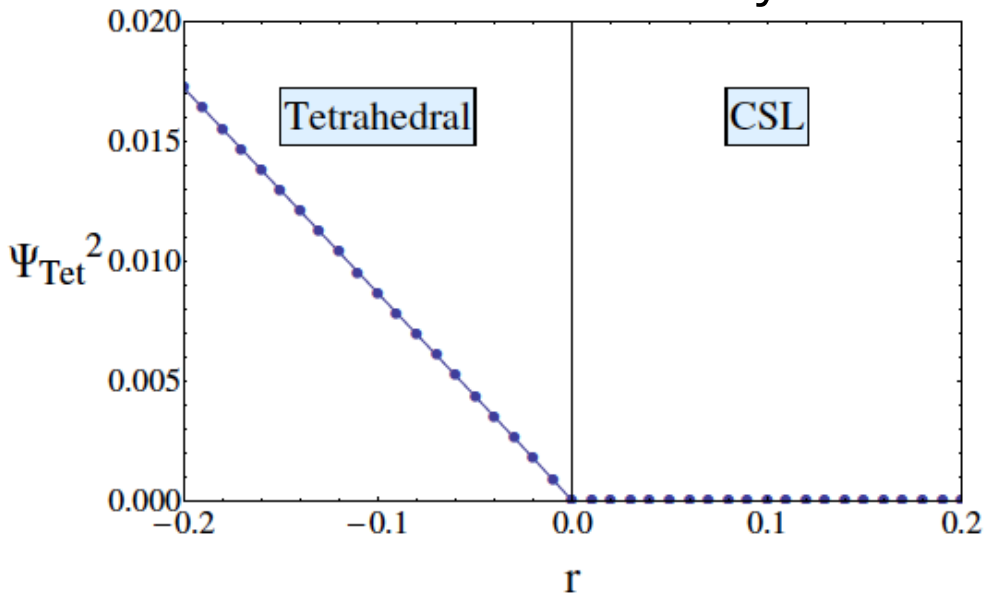
$$\mathcal{L}^{(2)}_{\mathrm{int}} = w_1\Big(\sum_i \rho_i\Big)^3 + w_2 \sum_{i,j,k} \epsilon^{ijk} \vec{\mathcal{S}}_i \cdot (\vec{\mathcal{S}}_j \times \vec{\mathcal{S}}_k) + \ldots$$

"Momentum space skyrmion crystal"

Spin crystallization transition out of the CSL

Chern-Simons-Higgs theory

Mean field theory



- The CSL arises from frustration induced melting of tetrahedral order
- We formulate a Chern-Simons-Higgs theory of the phase transition

Summary

- 2D double perovskites: Sr_2FeMoO_6, Ba_2FeReO_6
  - Topological phases including emergent Chern bands
  - C=1 quantum anomalous Hall insulators

- Chern bands and interactions
  - Novel magnetic orders
  - Melting noncoplanar order: route to chiral spin liquid