Double Perovskites

Spin-Orbit Coupling, Magnetism, Chern Bands

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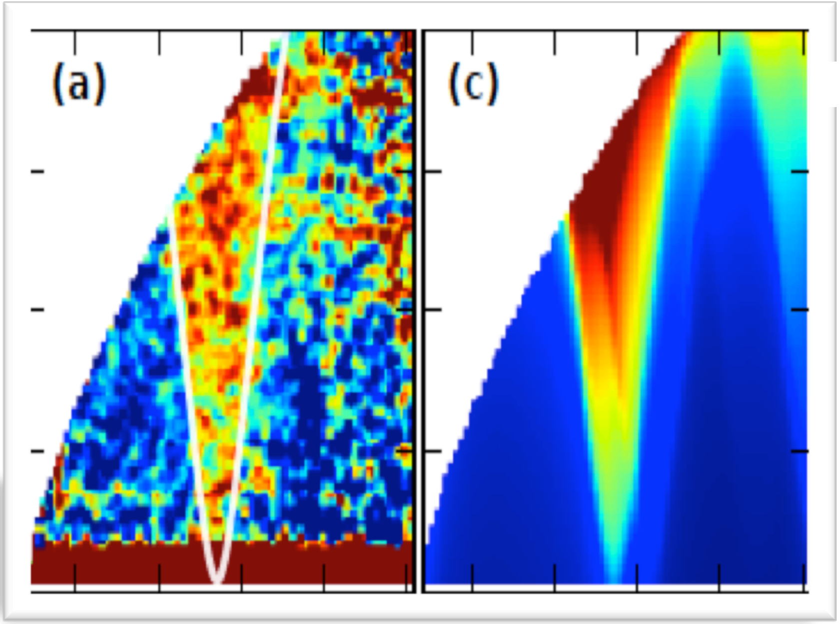
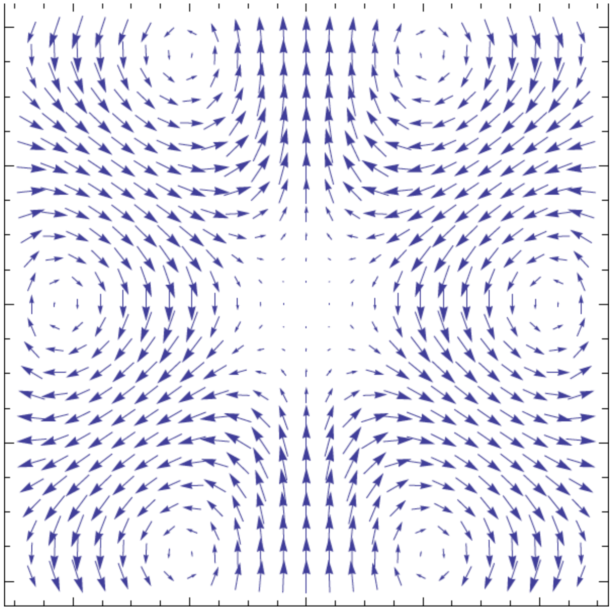
(University of Toronto)





Oxides Workshop, FTPI Minnesota, May 2014

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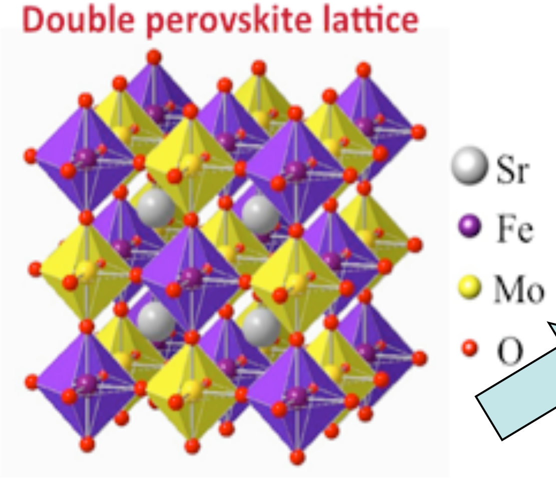
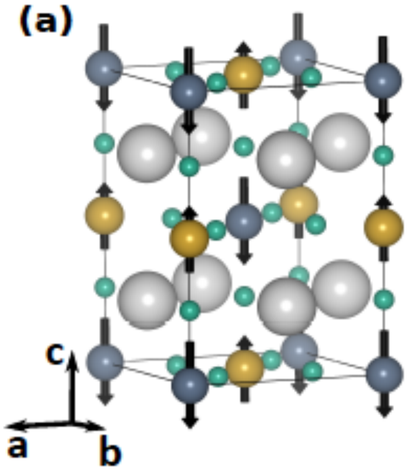
Double Perovskites

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





Frustrated magnetism

B=mag, B'=nonmag

. Get FCC lattice of spins (eg: Ba_2YMoO_6)

. Unusual spin-orbit coupled liquids?

High T_C ferromagnetism

B,B' both magnetic

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

  - Large polarization: good for spin injection

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

Interplay of SO coupling, magnetism, transport

Ba_2FeReO_6 [BFRO]

Sr_2FeMoO_6 [SFMO]

Collaborators

Theory


Ashley Cook
(Toronto)


Ciaran Hickey
(Toronto)

Neutron scattering

K.Plumb, P.Clancy, Y.J. Kim (Toronto), A.Kolesnikov (ORNL)

Samples

B.-C. Jeon, T.-W. Noh (SNU, Korea)

Experiments: Bulk magnetism and transport

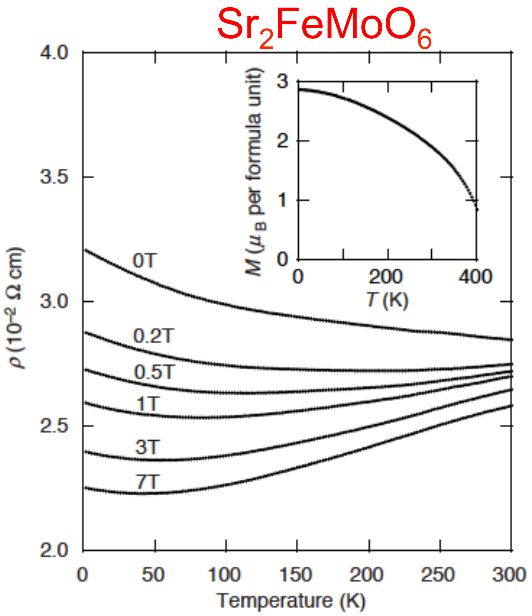
- **Metallic transport**, but evidence of half-metallicity

- **High Ferromagnetic T_C** (BFRO:315K, SFMO:420K)
- Antiparallel magnetization on Mo/Re and Fe

- **Tetragonal distortion below magnetic Tc**

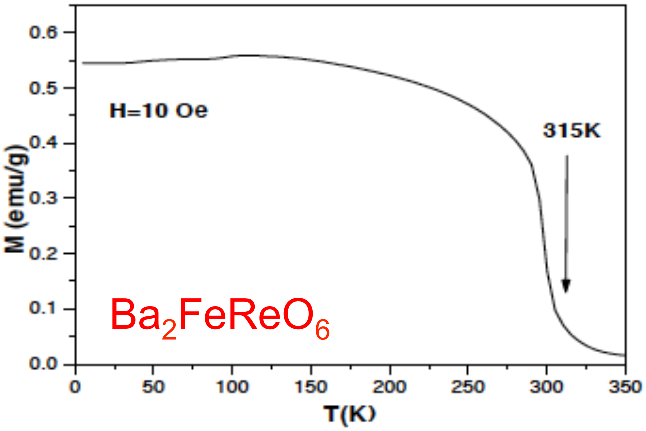
- **XMCD in BFRO shows orbital+spin magnetism**
  Re: Orbital and spin moments
  Fe: Spin moment

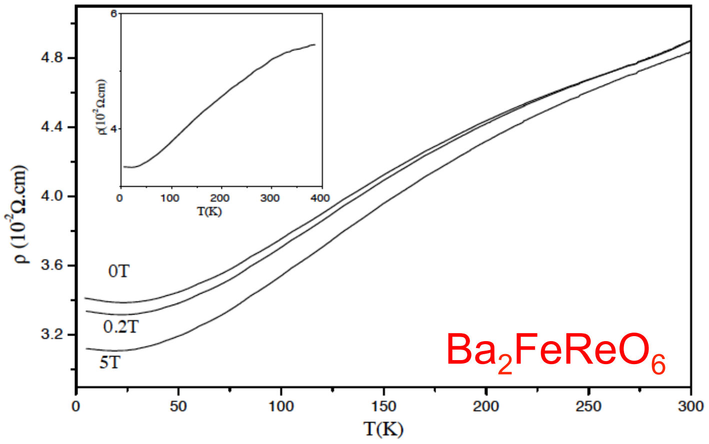
Azimonte, et al, PRL ***98****, 017204 (2007)*



Tokura group, Nature (1998)

Indirect evidence of half-metallicity



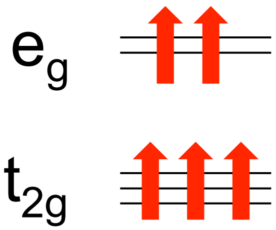


R. L. Greene group JPCM (2000)

Atomic states

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

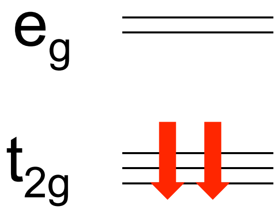
Fe$^{(3+)}$ 3d



S=5/2

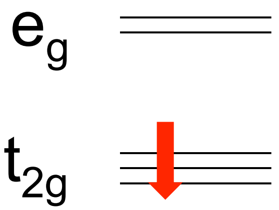
Hund's coupling dominates over crystal field

Re$^{(5+)}$ 5d



S.O. λ ~ 500 meV
Hubbard U important

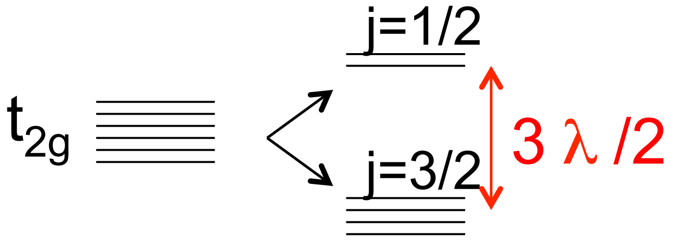
Mo$^{(5+)}$ 4d



S.O. λ ~100meV

Spin orbit coupling in t_{2g}

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



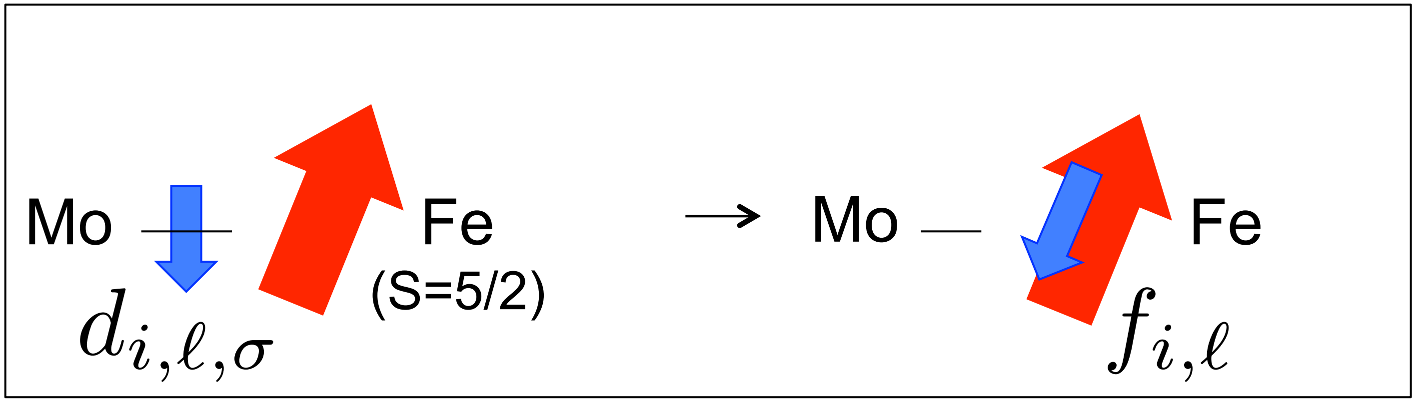
Interactions in t_{2g}

$$H_{\text{int}} = U \sum_{\alpha} n_{\alpha\uparrow} n_{\alpha\downarrow} + (U - 5\frac{J_H}{2}) \sum_{\alpha<\beta} n_\alpha n_\beta$$

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

(Kanamori)

Electronic model



Kinetically stabilized ferromagnetism

Manganites - Double exchange, Hund's coupling: Ferromagnetic

Double perovskite ferrimagnets - Pauli blocking: Ferrimagnetic

D.D. Sarma, et al (PRL 2000); S. Di Matteo, G. Jackeli, N. B. Perkins (PRB 2003)
Phillips, Chattopadhyay, A.J. Millis (PRB 2003);
L. Brey, M.J. Calderon, S. Das Sarma, F. Guinea (PRB 2006)
O. Erten, O. Nganba-Meetei, M. Randeria, N. Trivedi, P. Woodward (PRL 2011)
O. Nganba-Meetei, O. Erten, M. Randeria, N. Trivedi, P. Woodward (PRL 2013)

Open issues

- Correlation effects?
- Spin-orbit coupling?
- Spin-orbital dynamics?
- Berry curvature and anomalous Hall effects?

Hamiltonian

Charge transfer gap on Fe

Fe-Re nearest-neighbour hopping

$$H = -\sum_{\langle ij\rangle,\ell,\sigma}\left[t_\ell^{ij}\, g_\sigma(j)\, d^\dagger_{i\ell\sigma} f_{j\ell} + \mathrm{H.c.}\right] + \Delta\sum_{i\ell} f^\dagger_{i\ell} f_{i\ell}$$

$$-\sum_{\langle\langle ij\rangle\rangle,\ell,\sigma} \eta^{ij}_{\ell\ell'}\, d^\dagger_{i\ell\sigma} d_{j\ell'\sigma} + i\frac{\lambda}{2}\sum_{i} \varepsilon_{\ell mn}\,\tau^{n}_{\sigma\sigma'}\, d^\dagger_{i\ell\sigma} d_{im\sigma'}$$

spin-orbit coupling

Re-Re nearest-neighbour hopping

A. Cook, AP, PRB 88, 235102 (2013)

$$g_\uparrow(j) = \sin\frac{\theta_j}{2}\mathrm{e}^{-i\phi_j/2}$$

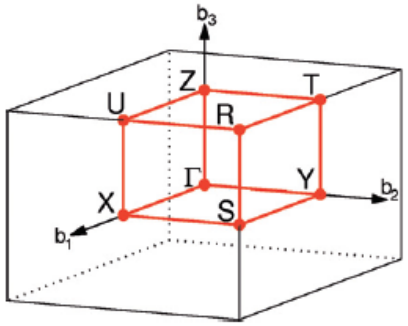
$$g_\downarrow(j) = \cos\frac{\theta_j}{2}\mathrm{e}^{i\phi_j/2}$$

$$H_{\text{int}} = U\sum_{\alpha} n_{\alpha\uparrow}n_{\alpha\downarrow} + (U - 5\frac{J_H}{2})\sum_{\alpha<\beta} n_\alpha n_\beta$$

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

Importance of SO + correlations
in “Cr-5d” DPs: H.Das, et al (PRB 2011)

Electronic model for BFRO: Hartree theory



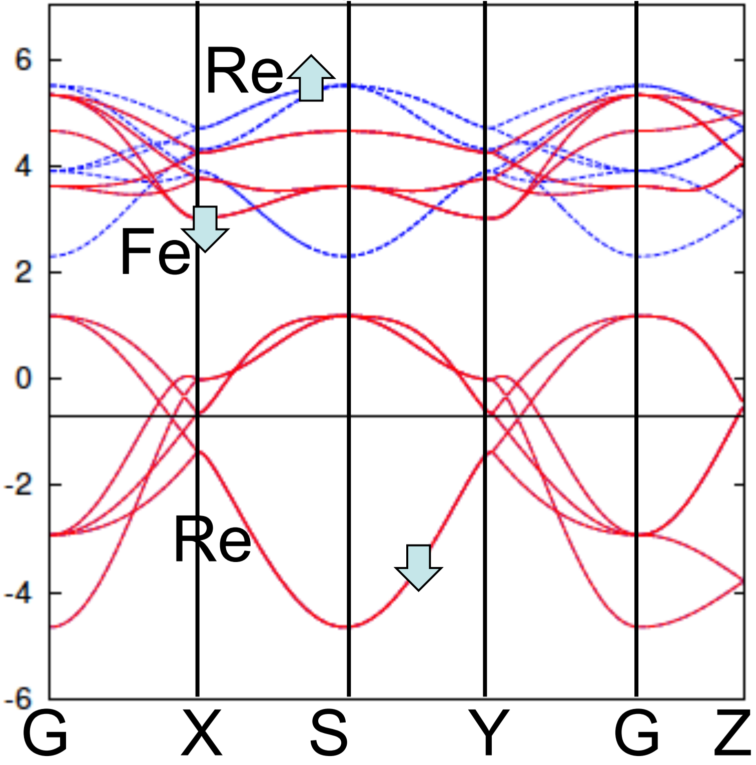
Correlation on Re: Stabilizes half-metallic state

Keep only intra-orbital U

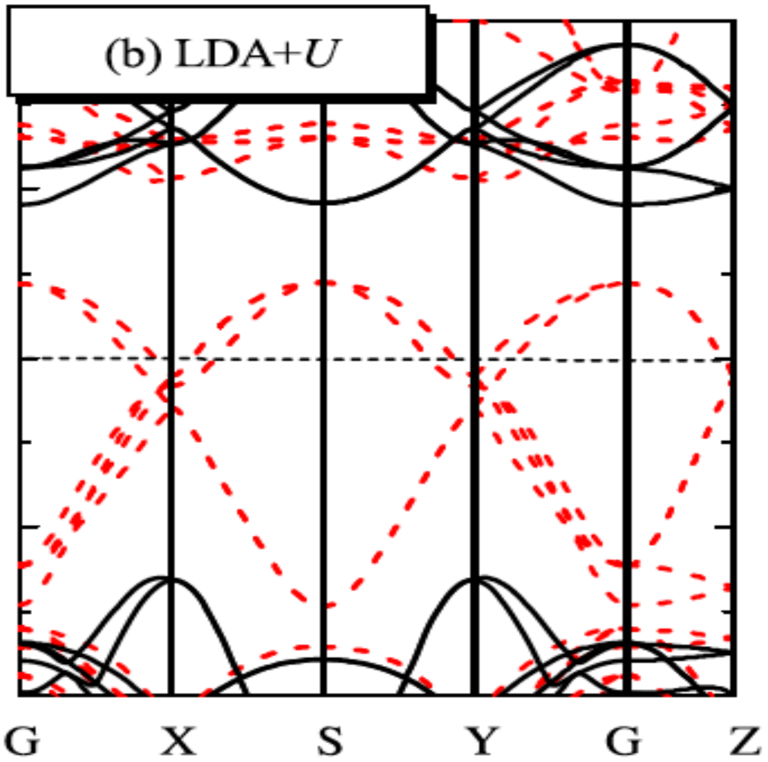
$t_{Fe\text{-}Re}$ ~ 330 meV, U ~ 2.5eV, Δ_{CT} ~ 1 eV

$t_{Re\text{-}Re}$ ~ 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)

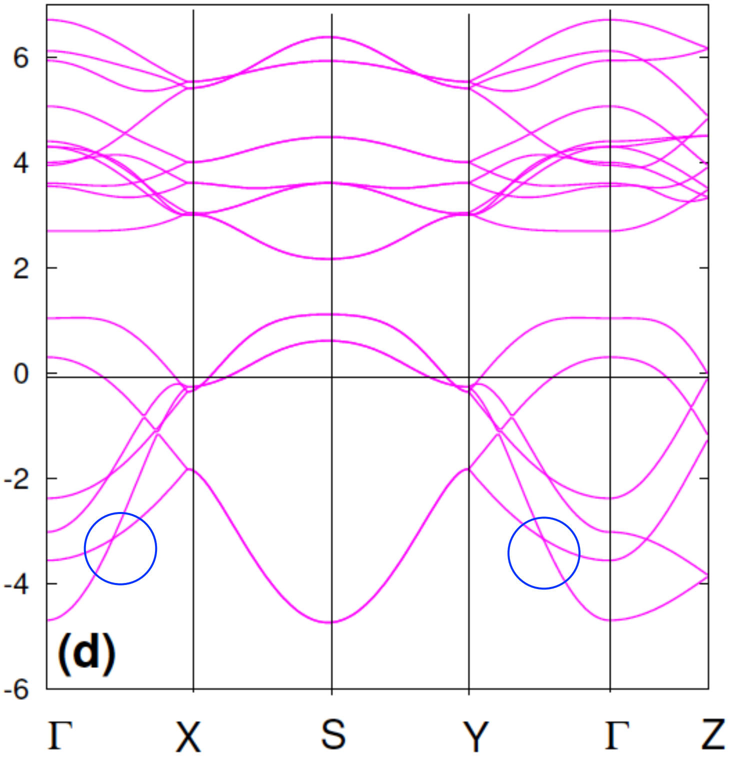
Electronic model for BFRO: Correlations and SOC

Spin-Orbit coupling lifts many degeneracies

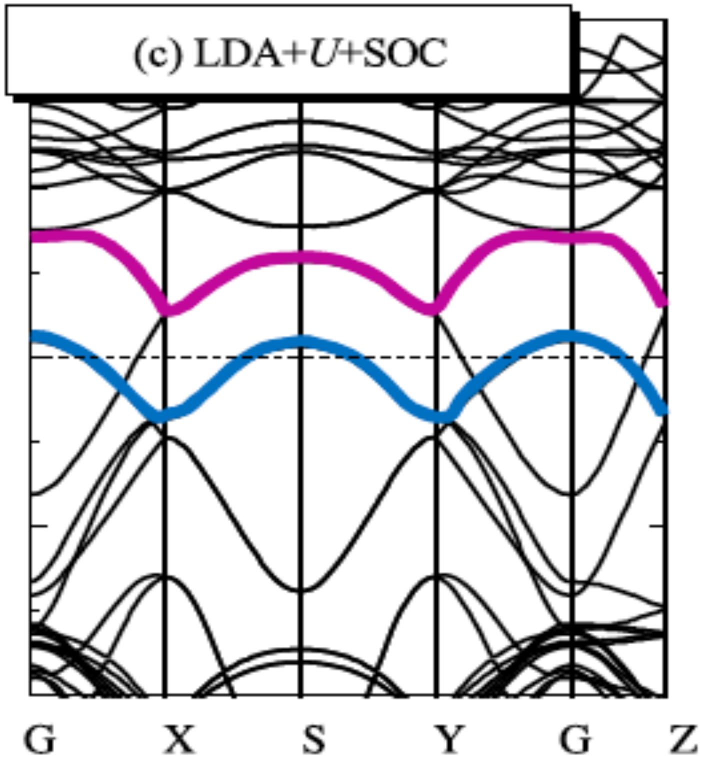
Stable Weyl nodes: Discrete band touching points with topological properties

[X.Wan, A. Turner, A. Vishwanath, S. Savrasov (PRB 2011); A. Burkov, L. Balents (PRL 2012)]

Comparing Hartree + SO dispersion with LDA+U+SOC



A. Cook, AP (PRB 2013)



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

Polarization at Fermi level ~ 90%

Electronic model for BFRO: Correlations and SOC

No correlations (U=0)

Sz (Fe) ~ +2.40 (i.e., 4.8 μ_B)
Sz (Re) ~ -0.15
Lz (Re) ~ -0.09

With correlations (U=2.5eV)

Sz (Fe) ~ +2.30 (i.e., 4.6 μ_B)
Sz (Re) ~ -0.78
Lz (Re) ~ -0.48

Important: $m_{sat} \sim 3.5\ \mu_B$

XMCD data on Re

C. Azimonte, et al, PRL **98**, 017204 (2007)

Theory: $(\mu_{orb}/\mu_{spin}) \sim -0.31$
Expt: $(\mu_{orb}/\mu_{spin}) \sim -0.29$

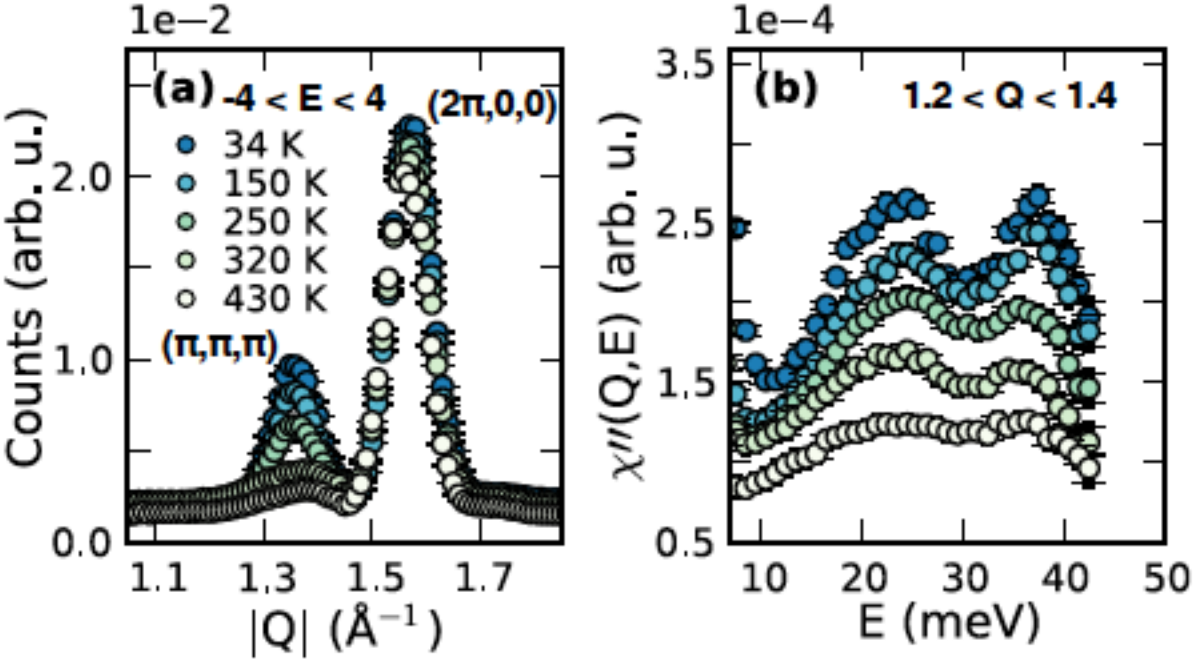
Tetragonal distortion

Weak orbital order in the Ferromagnetic state

$$\frac{\rho_{xy}}{\rho} - \frac{\rho_{xz}}{2\rho} - \frac{\rho_{yz}}{2\rho} \simeq 0.05$$

Magnetic fluctuations

Neutron scattering results



Y.J. Kim group

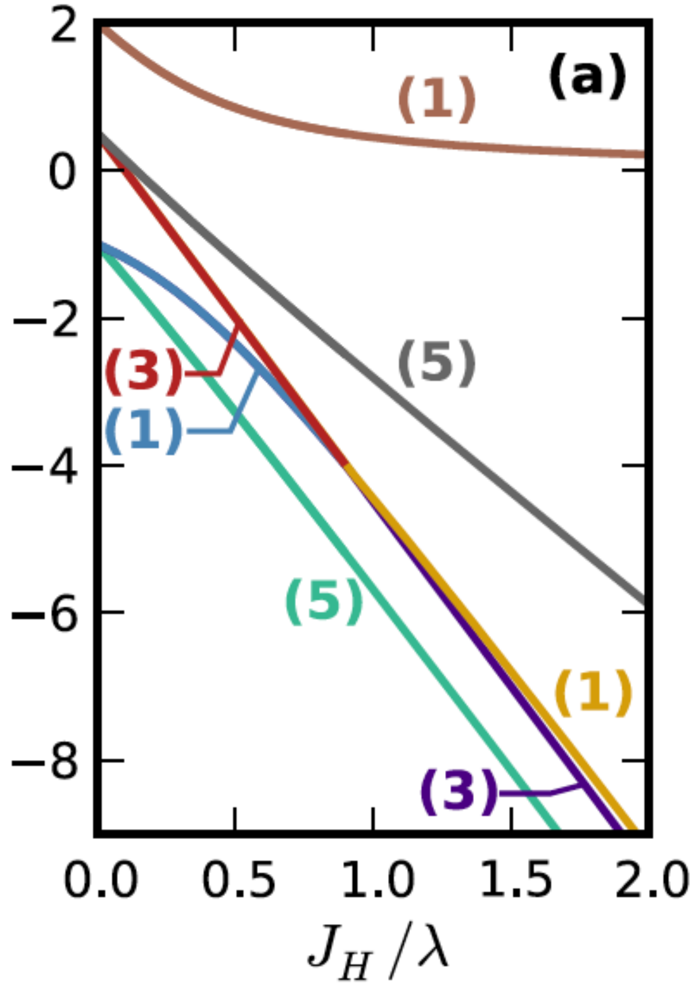
- Weak antisite disorder < 1% (from Xray studies)
- Re - friendly for neutron scattering (unlike Ir)
- Evidence for scattering at the magnetic Bragg peak
- Evidence for signal disappearing near magnetic Tc
- Confirm with Q-dependence of signal over wider range
  - weaker at large Q, unlike phonons

K. Plumb, A. Cook, et al, PRB 87,184412 (2013)

Local moment starting point: Atomic states

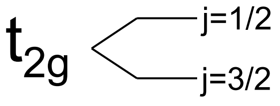
Simplified interaction for d^2

$$H_{\mathrm{at}}^{(2)} = -2J_H \vec{S}_{\mathrm{tot}}^2 - \frac{J_H}{2} \vec{L}_{\mathrm{tot}}^2 - \lambda_0 (\vec{L}_1 \cdot \vec{S}_1 + \vec{L}_2 \cdot \vec{S}_2)$$



J_H=0 states

Degeneracy: 6,8,1



t_{2g}

j=1/2

j=3/2

Large J_H states simple

L=1,S=1: J=2,1,0

L=2,S=0: No s.o.

L=0,S=0: No s.o.

Lowest 5 states smooth

For modest values $J_H > 2\lambda$

L=1 + S=1 giving J=2

K. Plumb, A. Cook, et al, PRB 87,184412 (2013)

Local moment Hamiltonian

On symmetry grounds, expect:

$$H = \mathcal{J} \sum_{\langle \mathbf{r}\mathbf{r}' \rangle} \vec{S}_{\mathbf{r}} \cdot \vec{\mathcal{F}}_{\mathbf{r}'} - \lambda \sum_{\mathbf{r} \in Re} \vec{L}_{\mathbf{r}} \cdot \vec{S}_{\mathbf{r}} \quad \xrightarrow[\lambda \gg \mathcal{J}]{\text{simplify}} \quad H_{\text{eff}} = \mathcal{J}_{\text{eff}} \sum_{\langle \mathbf{r}\mathbf{r}' \rangle} \vec{\mathcal{R}}_{\mathbf{r}} \cdot \vec{\mathcal{F}}_{\mathbf{r}'}$$

2-branch dispersion

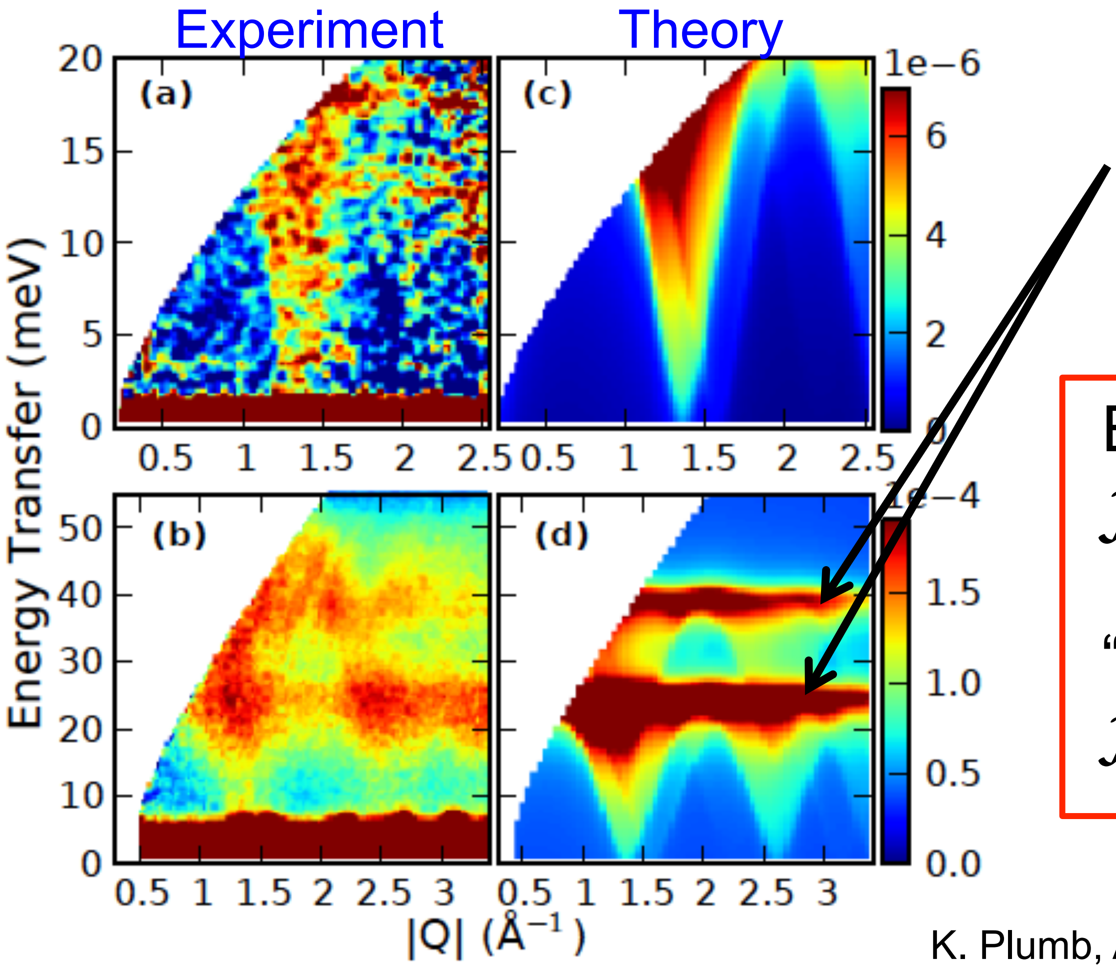
$$\Omega_{\pm}(\mathbf{Q}) = \sqrt{\mathcal{S}_{+}^{2} \gamma_{0}^{2} - \mathcal{F}\mathcal{R}\gamma_{\mathbf{Q}}^{2}} \pm \mathcal{S}_{-}\gamma_{0}$$

$$\mathcal{S}_{\pm} = (\mathcal{F} \pm \mathcal{R})/2$$

Dynamic structure factor

$$S_{\perp}(\mathbf{Q}, \omega) = 2\pi \sum_{\sigma = \pm} (G_{\mathbf{Q}} - \sigma \mathcal{S}_{-}) \delta(\omega - \Omega_{\sigma}(\mathbf{Q}))$$

Local moment Hamiltonian



Zone boundary energy ratio: $\mathcal{F}/\mathcal{R}$

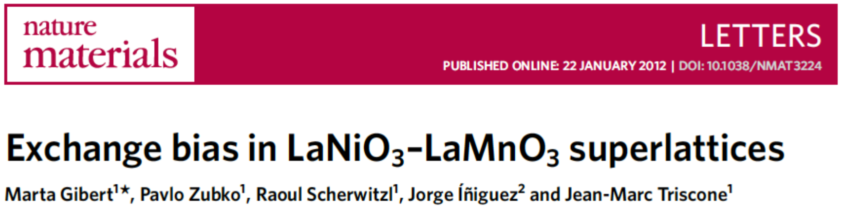
Experimental estimate $\mathcal{F}/\mathcal{R} \approx 1.6$

"Mean field" estimate $\mathcal{F}/\mathcal{R} \approx 1.9$

K. Plumb, A. Cook, et al, PRB 87,184412 (2013)

- Assuming F=2.1-2.3, find R ~ 1.3-1.4
- R > 1: Orbital moments participate in dynamics
- AFM Re-Fe coupling ~ 3 meV (rough Tc ~ 350K)
- Weak locking to lattice – consistent with weak orbital order

{111} grown oxide heterostructures



nature materials

LETTERS

PUBLISHED ONLINE: 22 JANUARY 2012 | DOI: 10.1038/NMAT3224

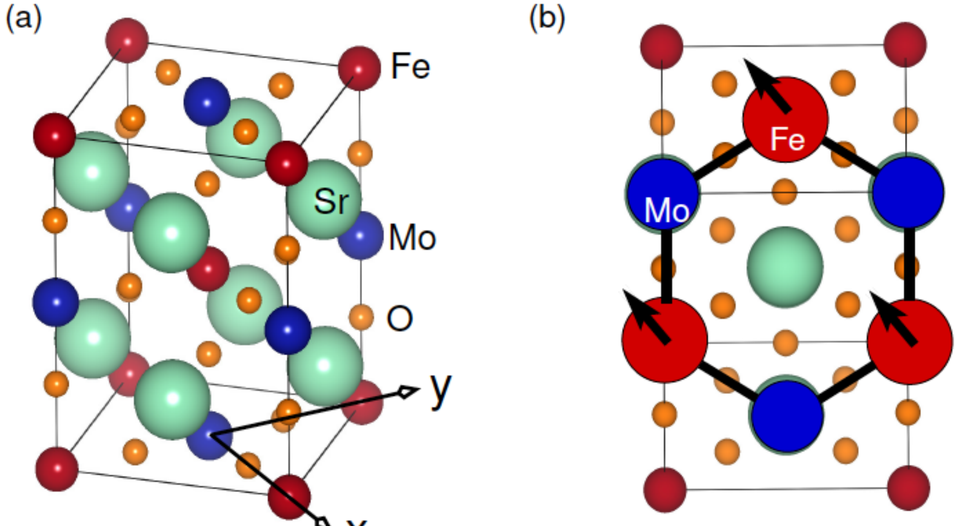
Exchange bias in $LaNiO_3$–$LaMnO_3$ superlattices

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

$(LaNiO3)_m$-$(LaMnO3)_n$ superlattices along {111}

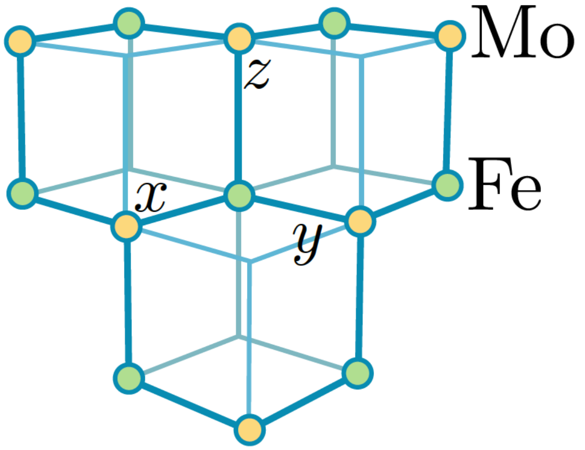
Infinite m=1,n=1 superlattice: La_2NiMnO_6 double perovskite

SFMO: {111} bilayer



Bulk crystal structure

{111} view

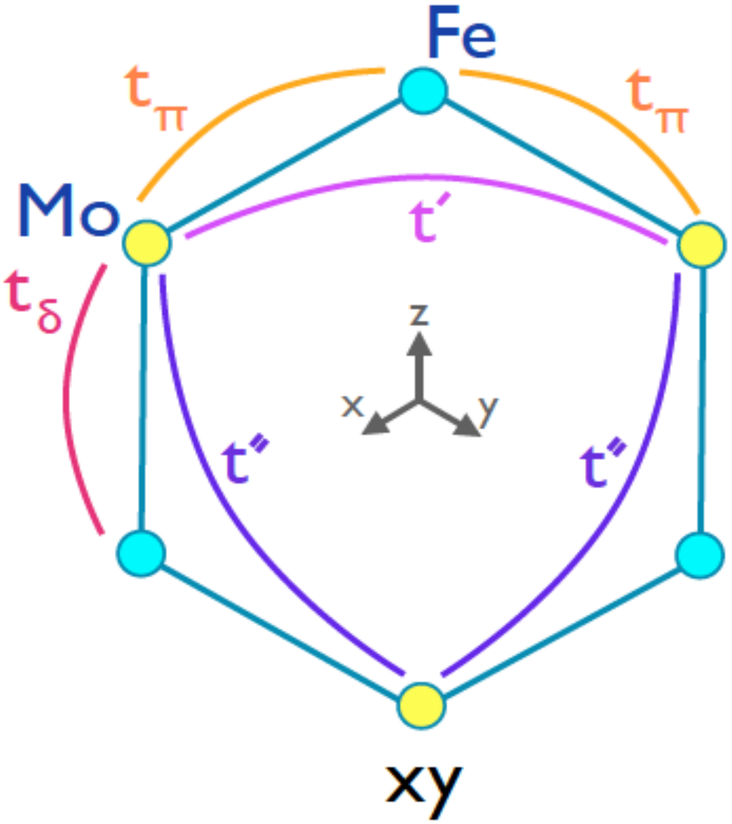


Fe and Mo/Re on honeycomb lattice

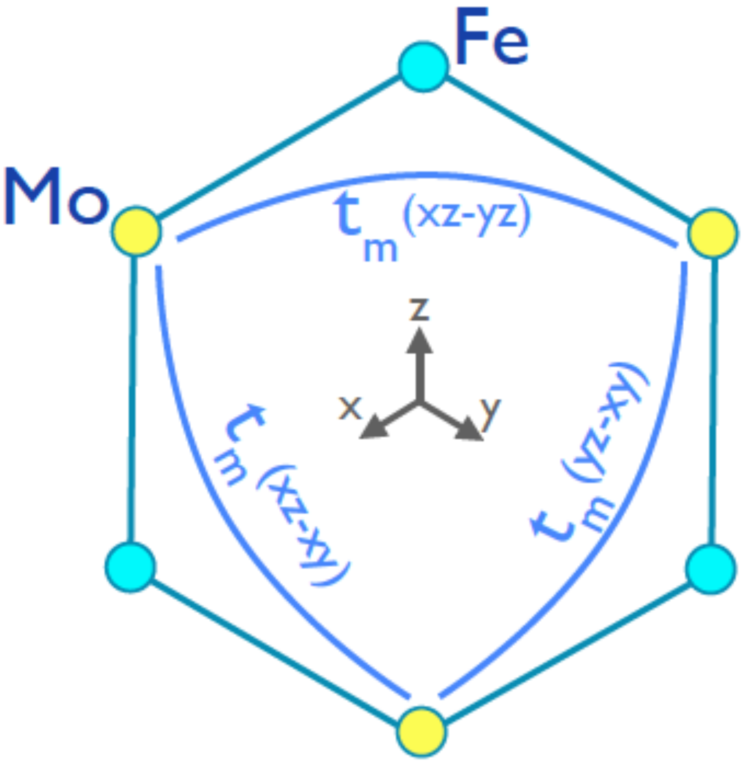
Fe: Local moments

Mo/Re: Itinerant t_2g electrons

Bilayer hoppings



Quasi-1D
intra-orbital hoppings



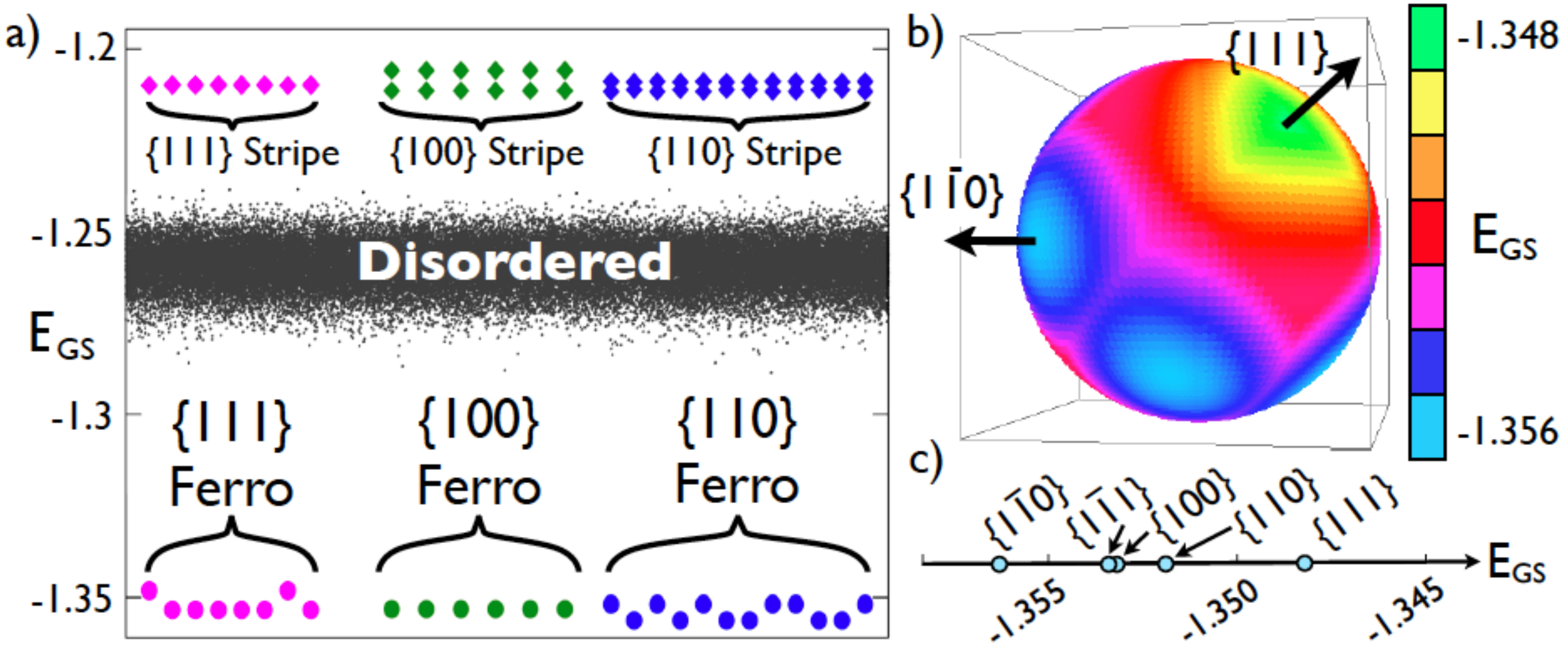
Orbital mixing
second-neighbor hoppings

$$t_\pi \sim 250\mathrm{meV} \qquad t_\delta, t', t'', t_m \sim 0.1 t_\pi$$

Magnetism of SFMO in {111} Thin Film Geometry

Adapt spins+fermions model to bilayer

Ferro orientation



Lowest energy: Ferromagnetic orientation of Fe moments
Exchange energy between Fe (S=5/2) moments ~ 1.5meV
Ising-like anisotropy due to SOC
Estimated Tc ~ 100 K

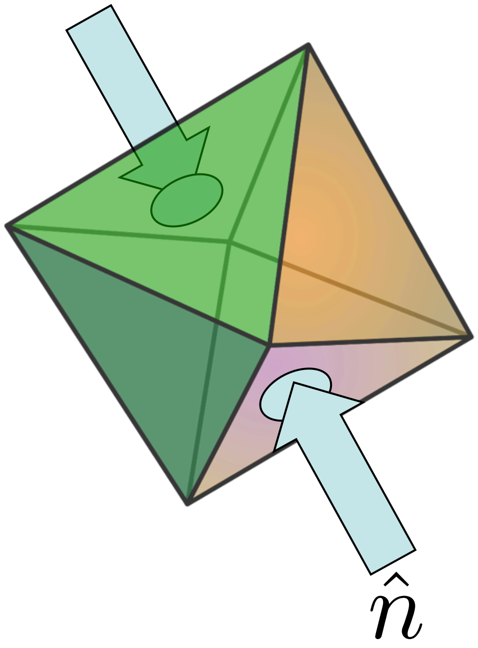
. A. Cook, AP, arXiv:1402.6347

Trigonal distortion

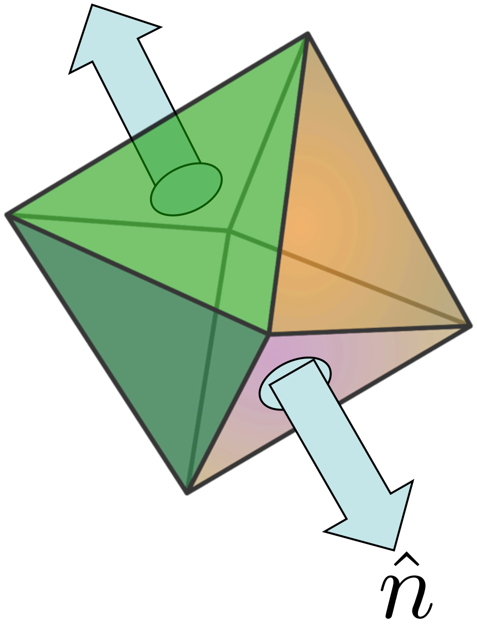
$$\boxed{H_{\mathrm{tri}} = \chi_{\mathrm{tri}}(\vec{L}.\hat{n})^2}$$

trigonal distortion of Mo oxygen cage

$\chi_{\mathrm{tri}} > 0$



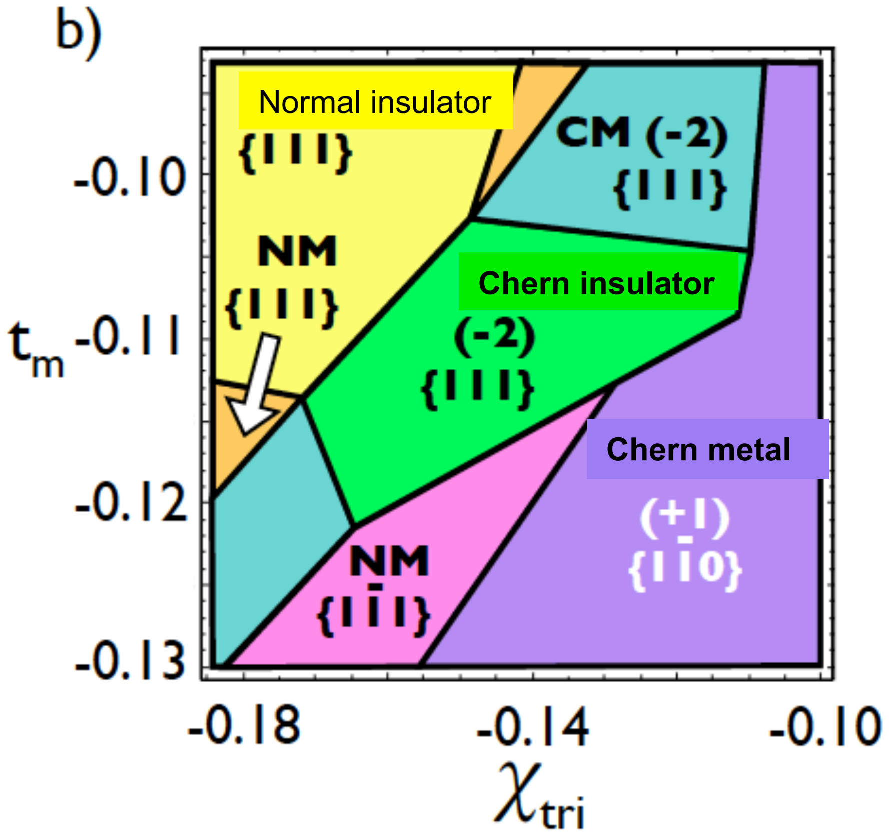
$\chi_{\mathrm{tri}} < 0$

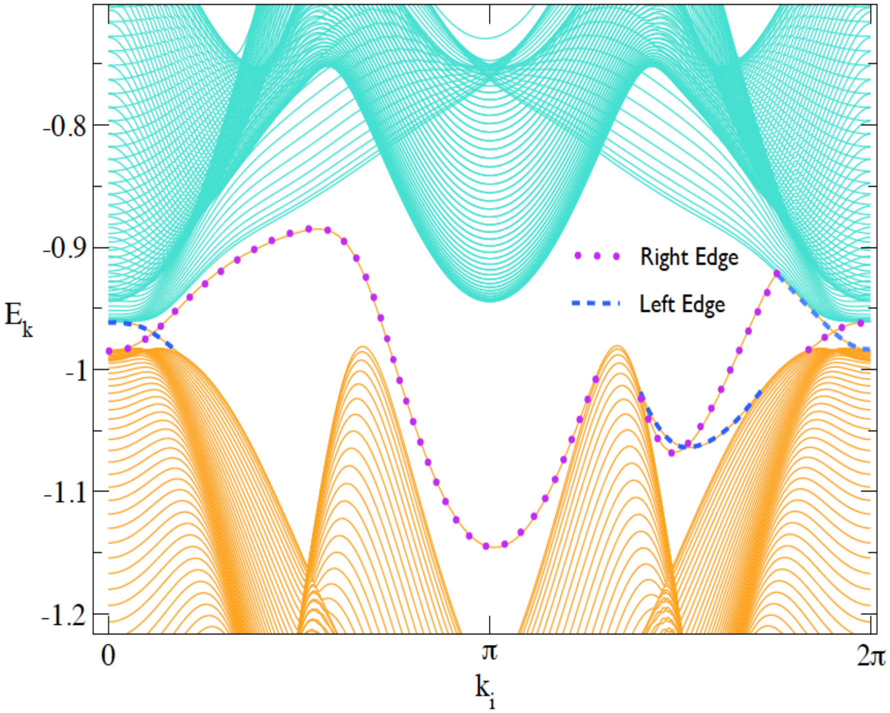


$\hat{n}$: Squeezing / Stretching axis

Symmetry: {111} perpendicular to bilayer

Phase diagram of SFMO in {111} bilayer





Edge state spectrum in C=2 Chern insulator

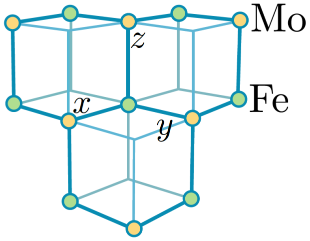
SFMO bilayer supports topologically nontrivial metals and C=2 Chern insulator

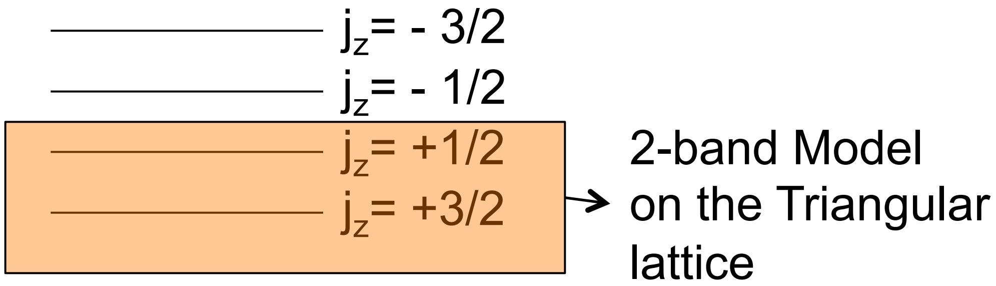
Quantum Hall gap ~ 75K

. A. Cook, AP, arXiv:1402.6347

Emergence of C=2 Chern bands in the {111} ferromagnet

Exchange split j=3/2 states on Mo due to Fe ferromagnetism





j_z= - 3/2

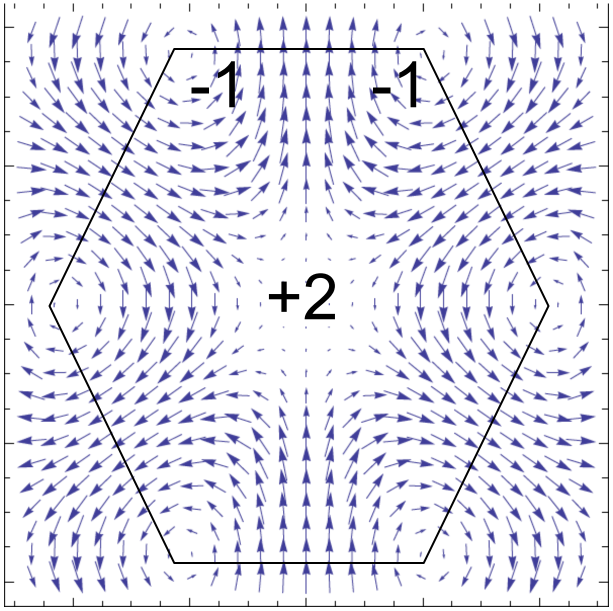
j_z= - 1/2

j_z= +1/2

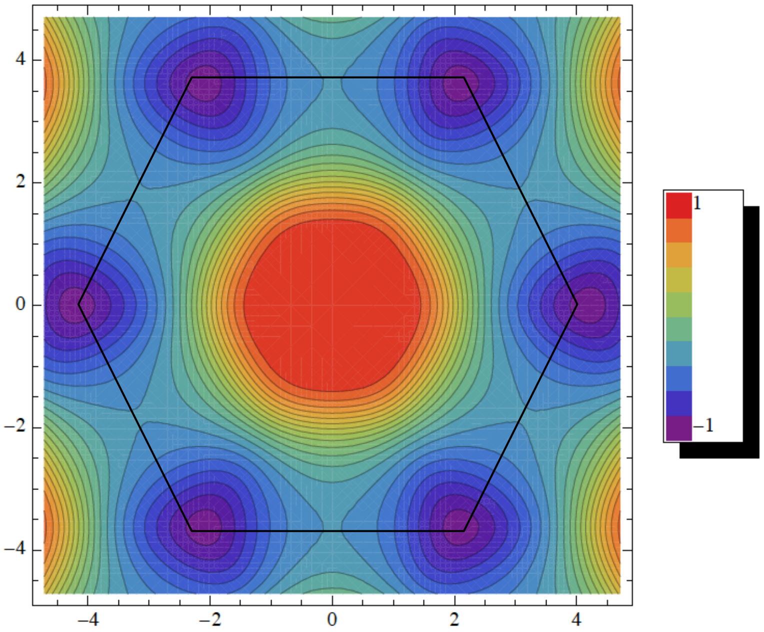
j_z= +3/2

2-band Model on the Triangular lattice

xy

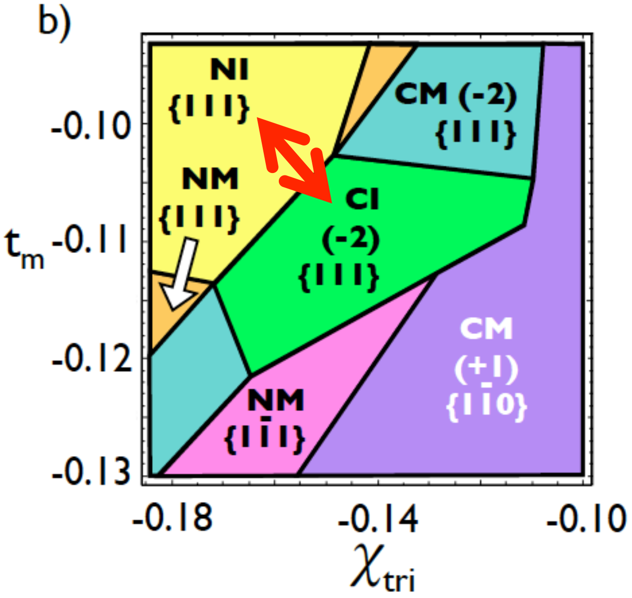


z (mass)



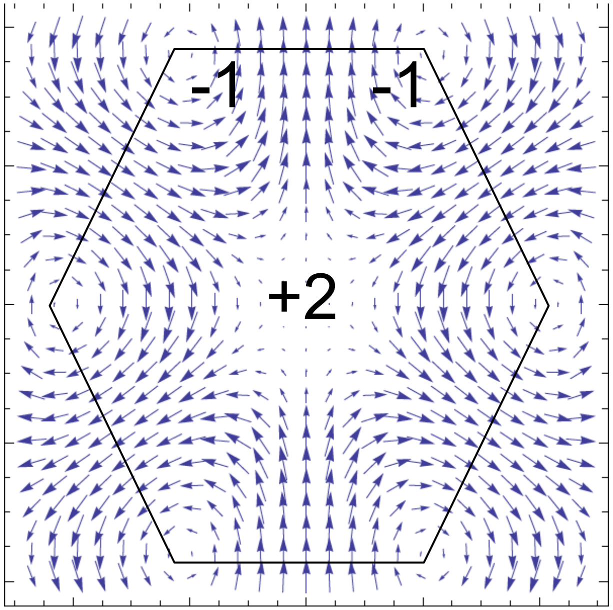
Skyrmion number = +/- 2

C=2 Chern insulator to Normal insulator transition



Haldane model: Chern number changes by “1”.

Chern number change by “2” is unusual: Symmetry protection needed (inversion or C_6 of Mo triangular lattice)



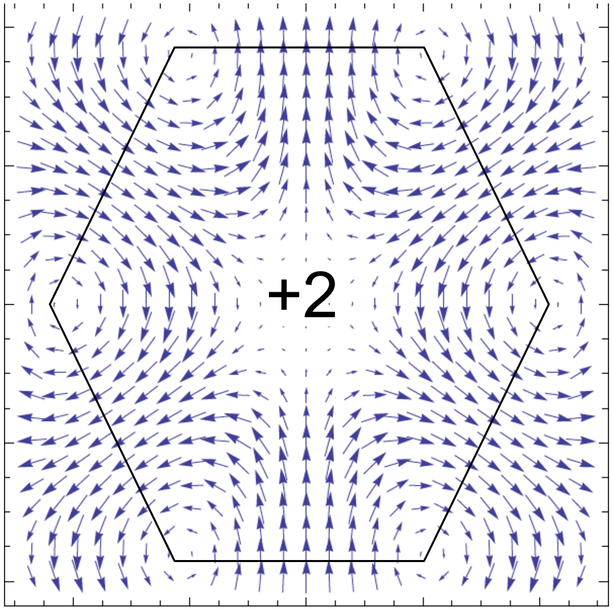
- Massless Dirac fermions at the BZ corners having “vorticity” 1 (inversion protected)
- Quadratic band touching at Γ-point with skyrmion texture having winding number=2 (protected by C_6 symmetry)

C.Hickey, A. Cook, AP (preprint)

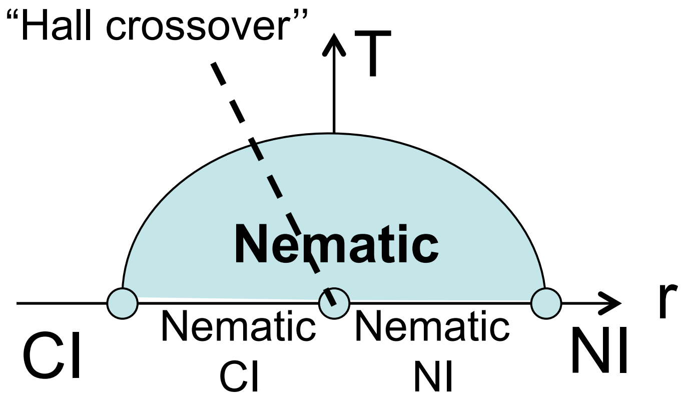
C=2 Chern insulator to Normal insulator transition

Near the Γ-point

$$H_0(\mathbf{k}) = \begin{pmatrix} (t_1+t_2)k^2 + r & t_3(k_x+ik_y)^2 \\ t_3(k_x-ik_y)^2 & -(t_1-t_2)k^2 - r \end{pmatrix}$$



$$\boxed{\begin{aligned} \frac{dr}{d\ell} &= 2r + \frac{g\Lambda^2}{4\pi}\frac{t_1}{\sqrt{t_1^2+t_3^2/4}} \\ \frac{dg}{d\ell} &= \frac{g^2}{6\pi}\frac{1}{\sqrt{t_1^2+t_3^2/4}}. \end{aligned}}$$



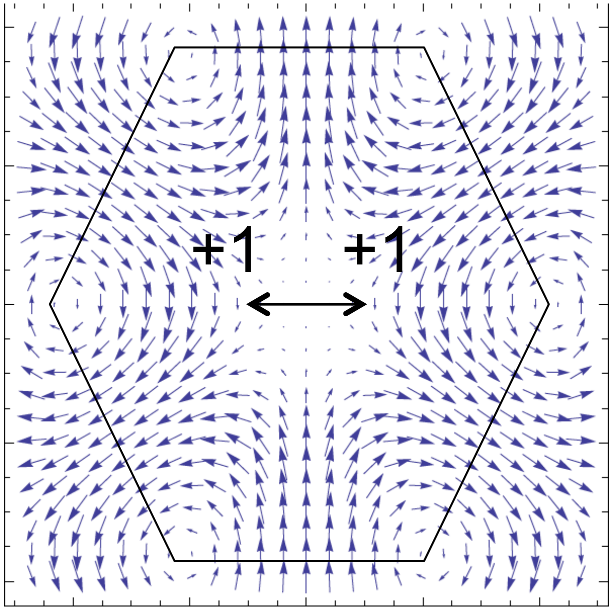
Marginally relevant interactions drive **nematic order** around the putative z=2 critical point

C. Hickey, A. Cook, AP (preprint)

C=2 Chern insulator to Normal insulator transition

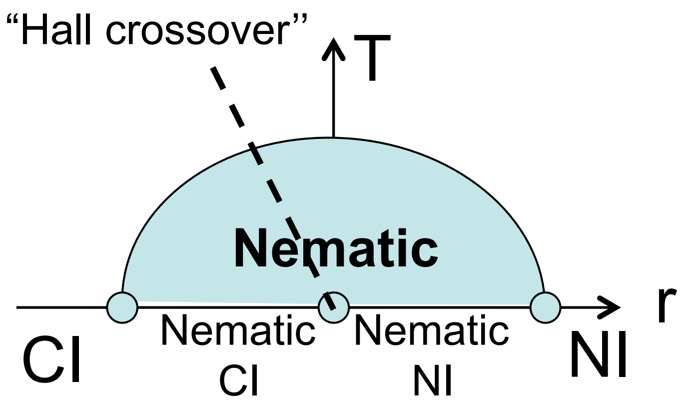
Near the Γ-point

$$H_0(\mathbf{k}) = \begin{pmatrix} (t_1+t_2)k^2 + r & t_3(k_x+ik_y)^2 \\ t_3(k_x-ik_y)^2 & -(t_1-t_2)k^2 - r \end{pmatrix}$$



$$\frac{dr}{d\ell} = 2r + \frac{g\Lambda^2}{4\pi}\frac{t_1}{\sqrt{t_1^2+t_3^2/4}}$$

$$\frac{dg}{d\ell} = \frac{g^2}{6\pi}\frac{1}{\sqrt{t_1^2+t_3^2/4}}.$$



Marginally relevant interactions drive **nematic order** around the putative z=2 critical point

C. Hickey, A. Cook, AP (preprint)

Summary

Spin orbit coupled double perovskites host rich physics

- 3D Bulk complex oxide: Ba_2FeReO_6
  - band structure with Weyl nodes
  - spin and orbital magnetization from strong correlations
  - spin dynamics in agreement with neutron data

- 2D Heterostructures {111}: Sr_2FeMoO_6
  Topological phases including emergent Chern bands and C=2 quantum anomalous Hall insulators

References

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4. C. Hickey, A. Cook, AP (in preparation, 2014)