Large anomalous Hall conductivity in CoS_2

Symmetry protected nodal structures in ultrathin $SrRuO_3$ film

“Tunable Anomalous Hall conductivity”

Changyoung Kim

Department of Physics & Astronomy, SNU
IBS Center for Correlated Electron Systems, SNU

Large anomalous Hall conductivity in CoS_2

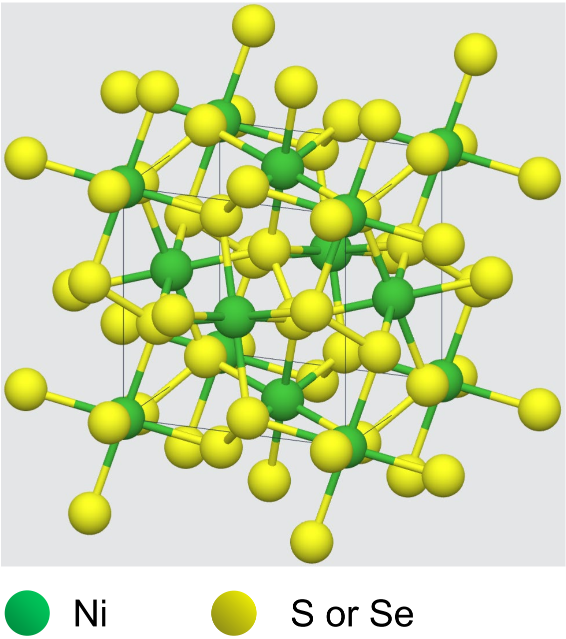
In collaboration with

- Joonyoung Choi, Younjung Jo (Kyungbook Nat U)
- Se-Young Park (Soongsil U)
- Jinghong Park, Joonwon Rhim (Ajou U)

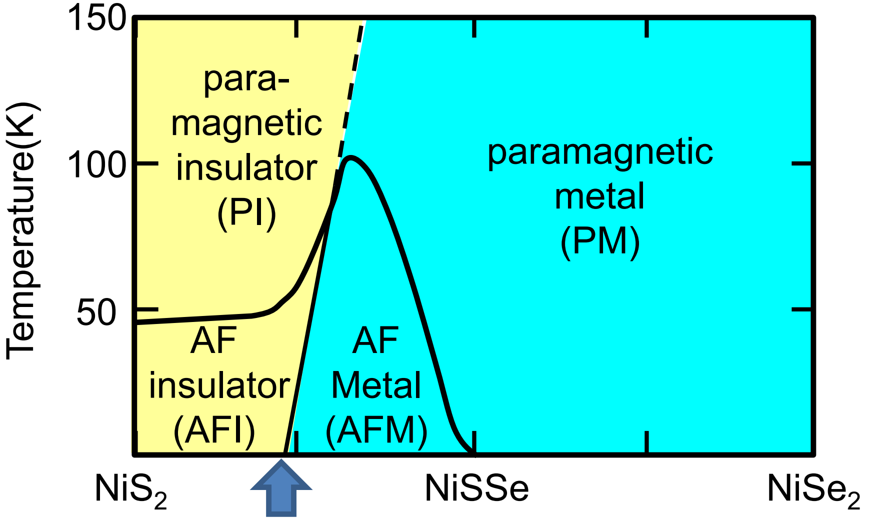
$NiS_{2-x}Se_x$ - Ideal MIT system

- Ni and S-S or Se-Se dimers form rock-salt structure
- NiS_2 and $NiSe_2$ are isovalent and isostructural
- Metal-insulator transition occurs around x=0.45 at T=20K
- Chemical pressure enhanced from Se to S

Pyrite structure ($Pa\bar{3}$ [205])



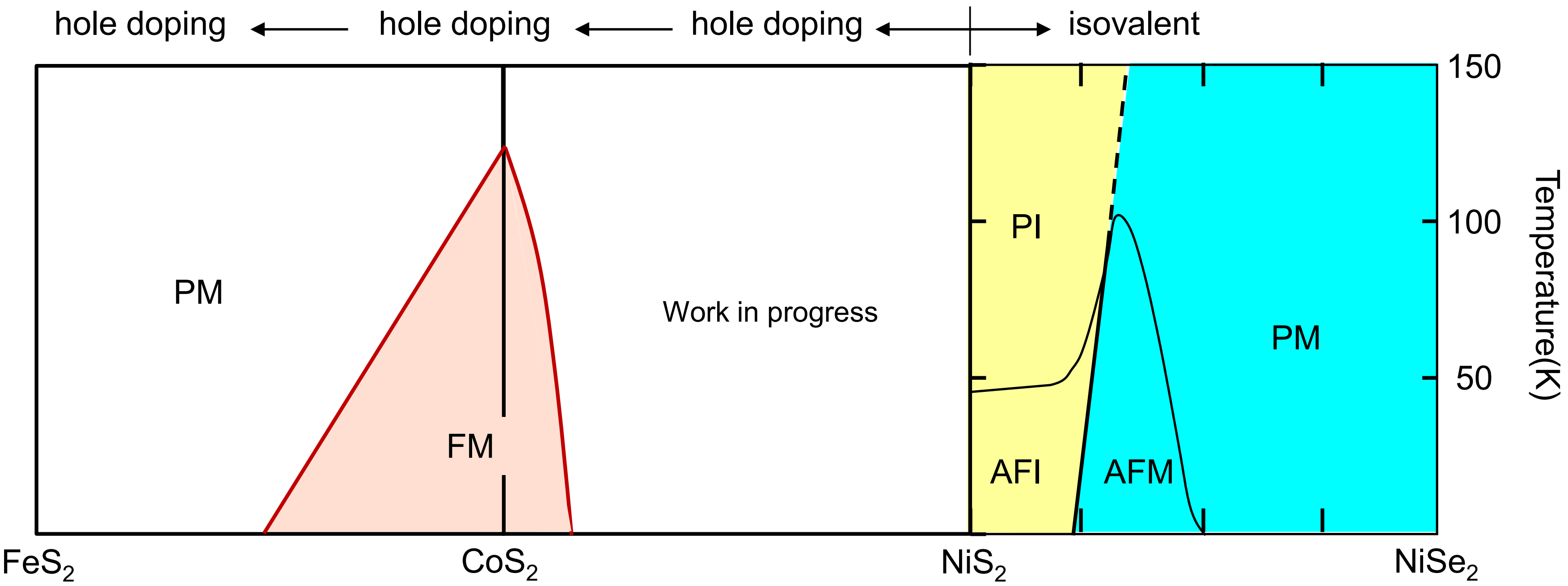
P. G. Niklowitz *et al*, PRB **77**, 115135(2008)

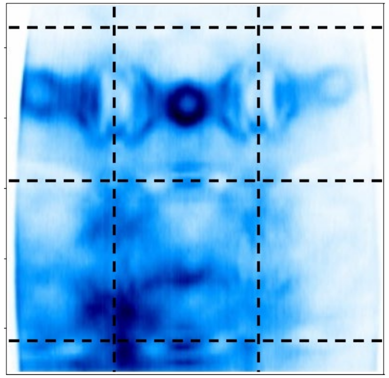


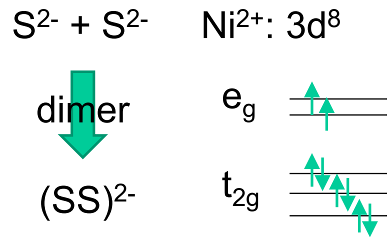
Insulator (Large U/t)

Metal (Small U/t)

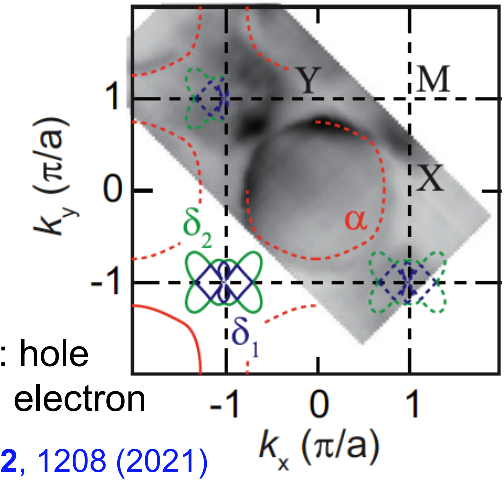
(Ni,Co,Fe)(S,Se)$_2$ phase diagram







Partially (half) filled e$_g$

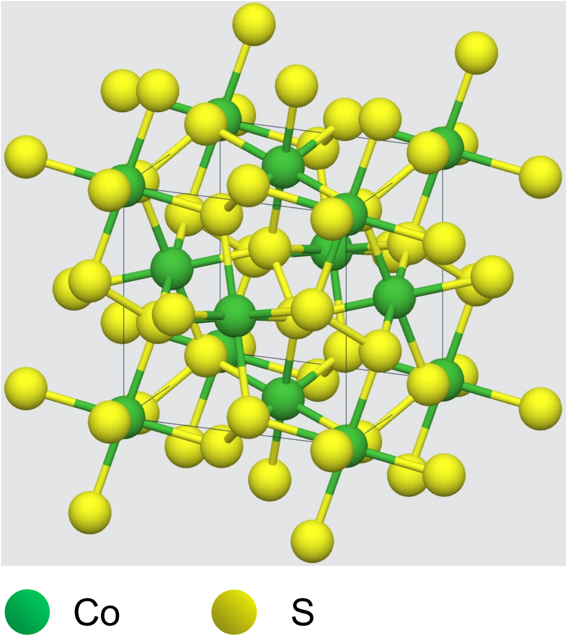


α: hole
δ: electron

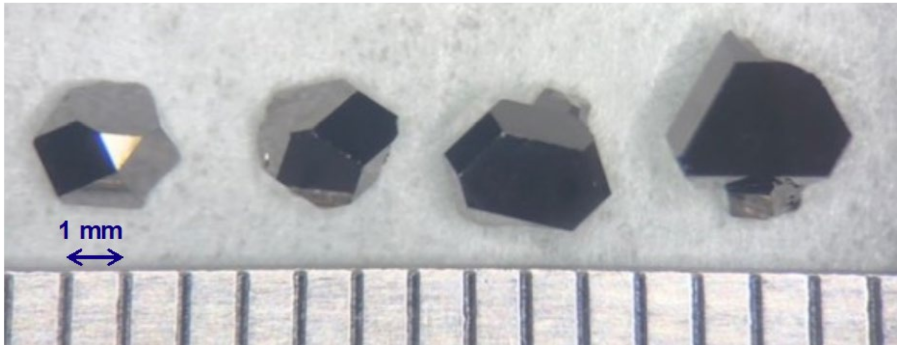
Nat Comm **12**, 1208 (2021)

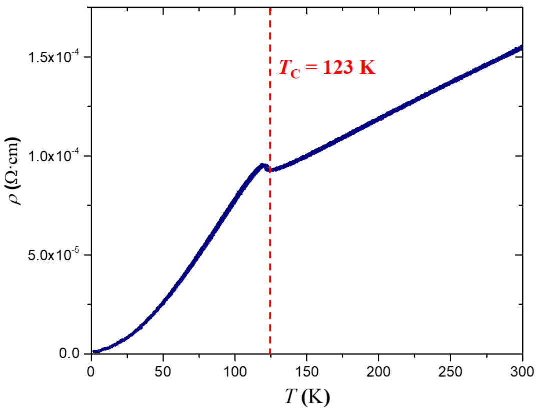
CoS_2

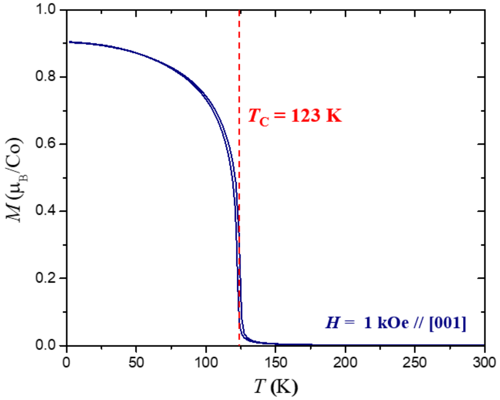
Pyrite structure ($Pa\bar{3}$ [205])



CoS_2 Single Crystals



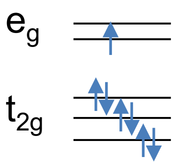


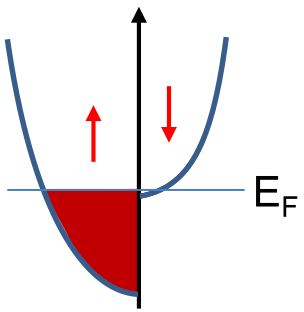


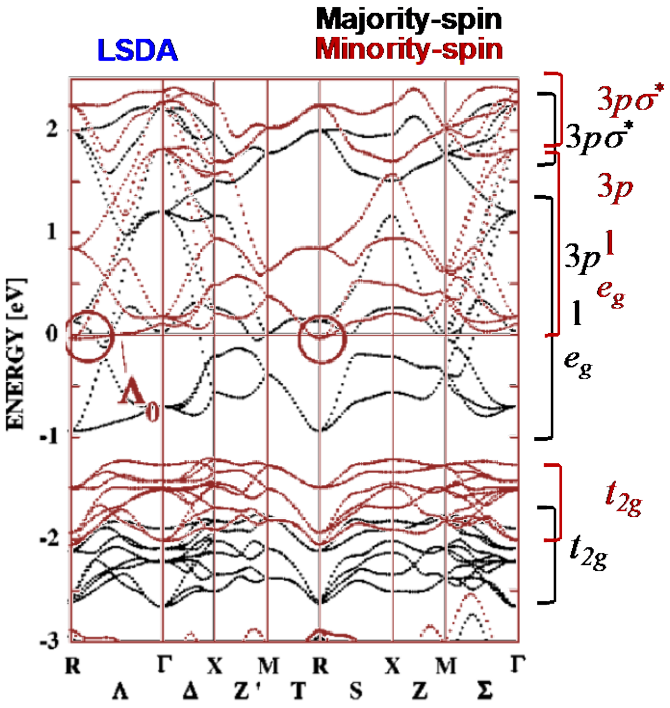
- Anomaly with thermal hysteresis at $T_{FM} = 123$ K
- $\rho_{2\,K} = 2.086 \times 10^{-6}\ \Omega\cdot cm$, $\rho_{300\,K} = 1.542 \times 10^{-4}\ \Omega\cdot cm$
- $RRR = \rho_{300\,K} / \rho_{2\,K} = 73.921$

CoS_2 – nearly half metal

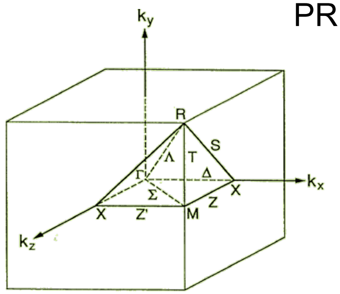
Co^{2+}: $3d^7$



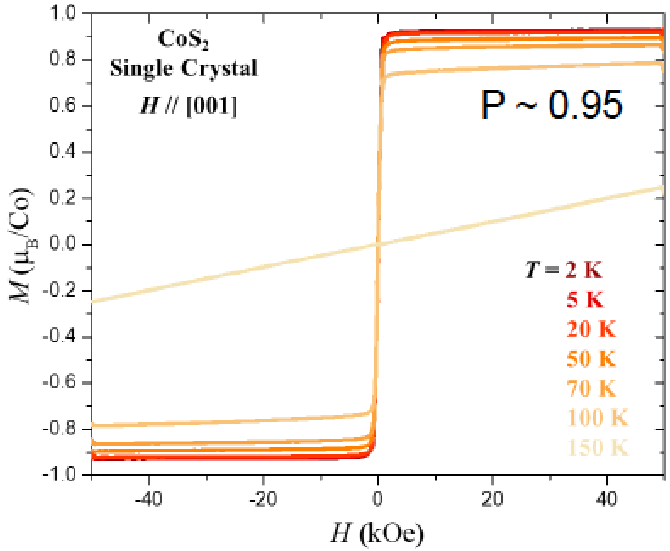




PRB **64**, 180401 (2001)

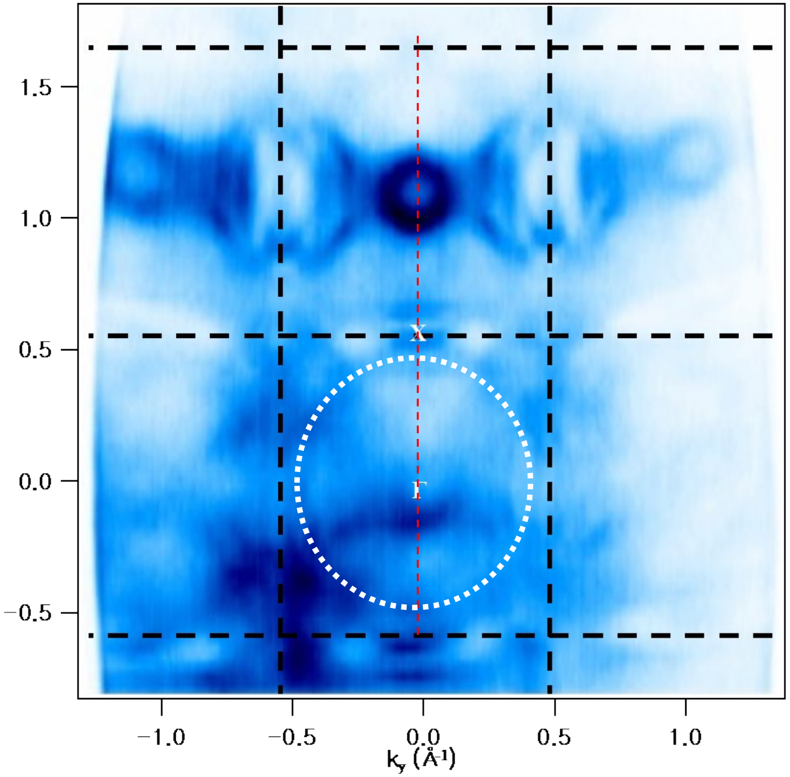


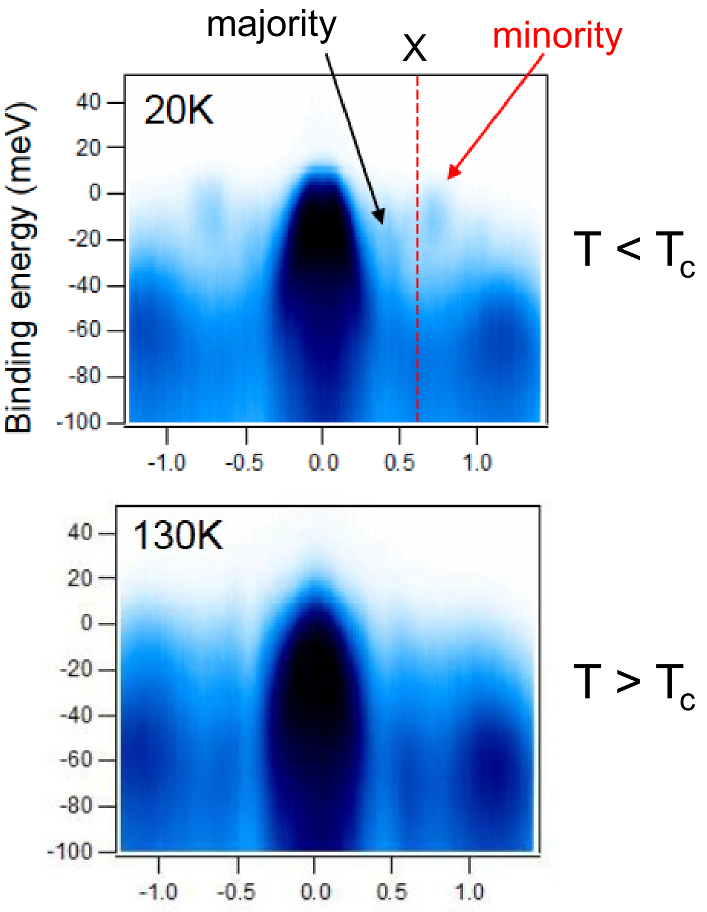
M-H



ARPES data

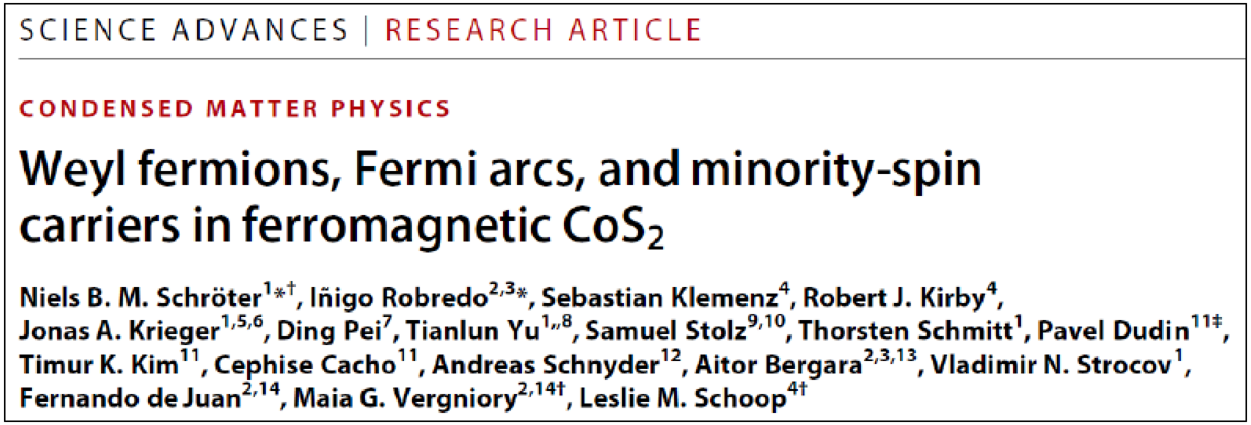
Fermi surface map





- Complex surface states ➔ to be addressed (B J Yang group, in progress)
- Split bands with mostly majority electrons ➔ Stoner type
- Can be hole doped to have only the majority band occupied ➔ perfect half metal

Weyl fermions in CoS_2?

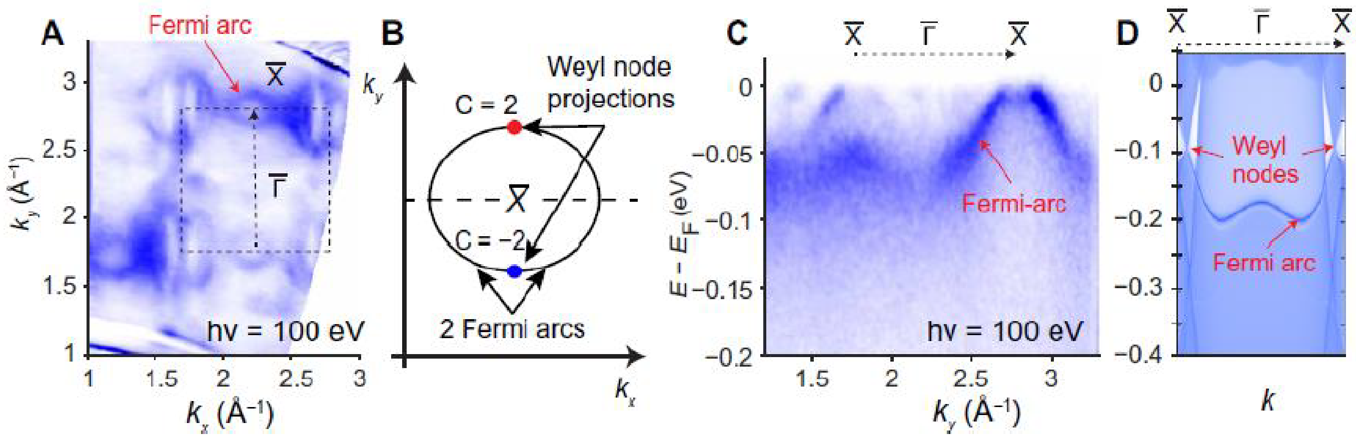


SCIENCE ADVANCES | RESEARCH ARTICLE

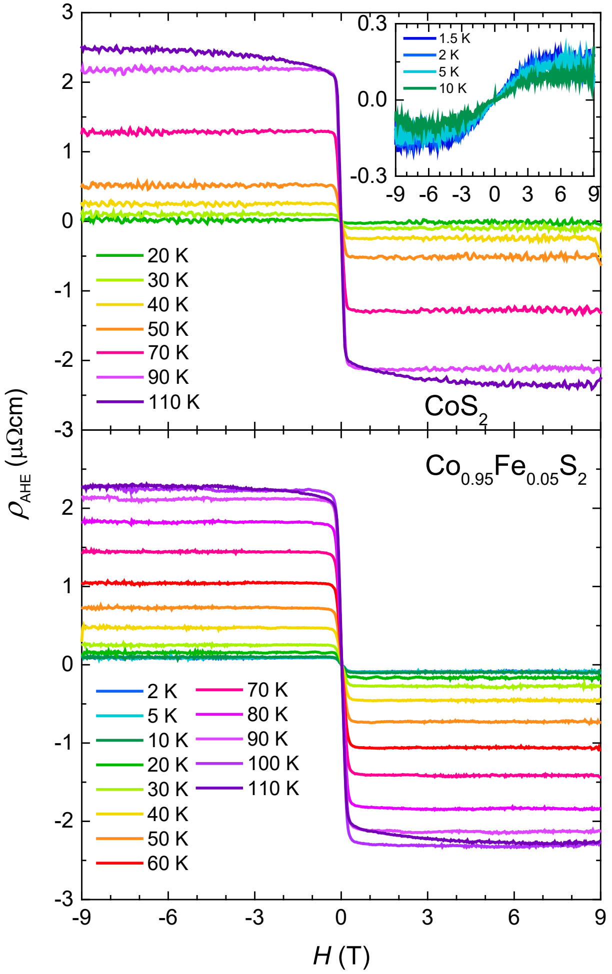
CONDENSED MATTER PHYSICS

Weyl fermions, Fermi arcs, and minority-spin carriers in ferromagnetic CoS_2

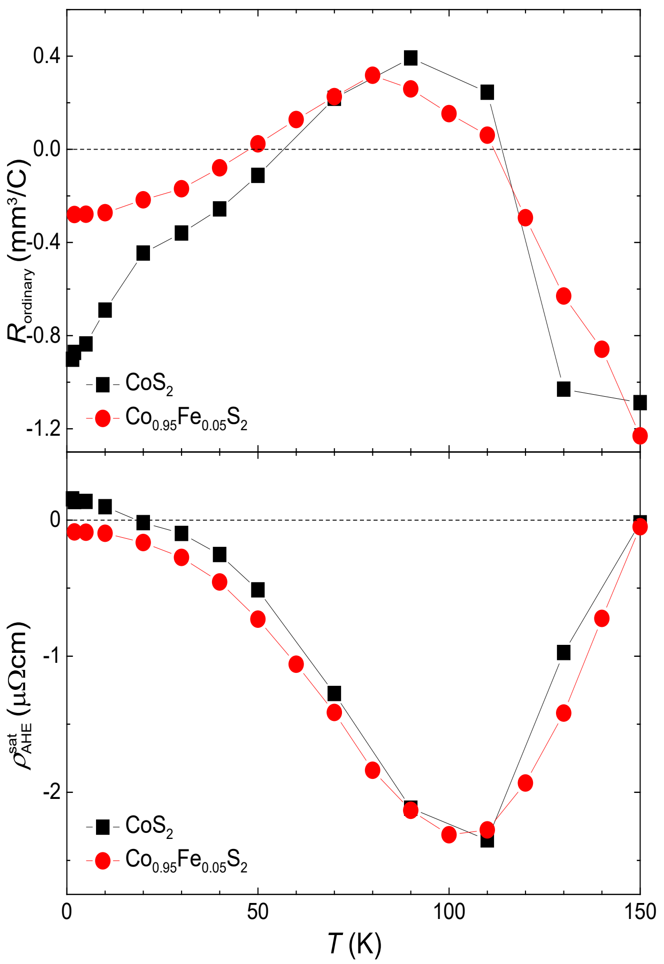
Niels B. M. Schröter[1*†], Iñigo Robredo[2,3*], Sebastian Klemenz[4], Robert J. Kirby[4], Jonas A. Krieger[1,5,6], Ding Pei[7], Tianlun Yu[1,8], Samuel Stolz[9,10], Thorsten Schmitt[1], Pavel Dudin[11‡], Timur K. Kim[11], Cephise Cacho[11], Andreas Schnyder[12], Aitor Bergara[2,3,13], Vladimir N. Strocov[1], Fernando de Juan[2,14], Maia G. Vergniory[2,14†], Leslie M. Schoop[4†]



Hall data



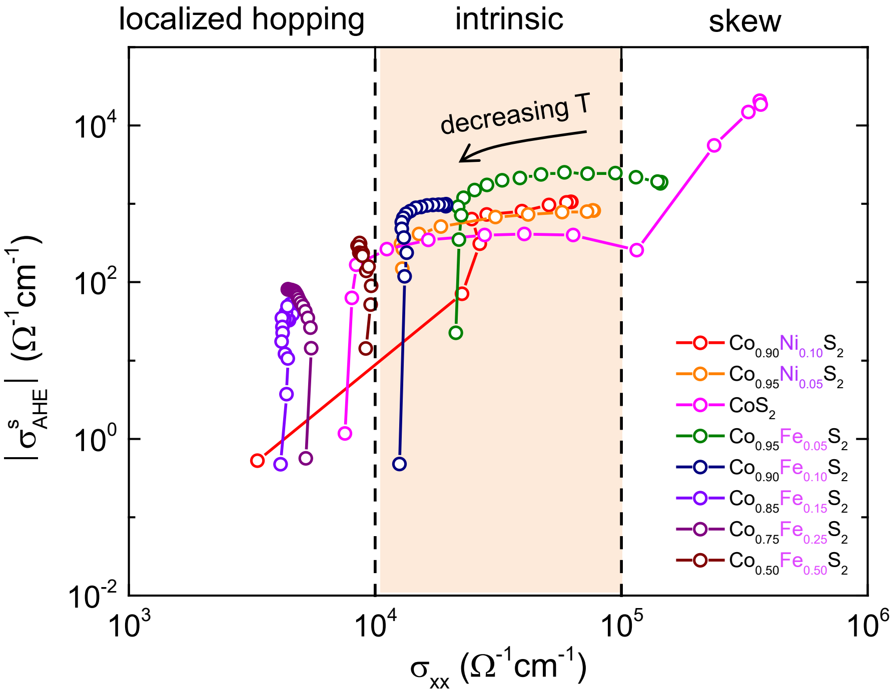
ρ_{AHE} (μΩcm)
3
2
1
0
-1
-2
-3
0.3
0.0
-0.3
-9 -6 -3 0 3 6 9
1.5 K
2 K
5 K
10 K
20 K
30 K
40 K
50 K
70 K
90 K
110 K
CoS_2
$Co_{0.95}Fe_{0.05}S_2$
2 K
5 K
10 K
20 K
30 K
40 K
50 K
60 K
70 K
80 K
90 K
100 K
110 K
-9
-6
-3
0
3
6
9
H (T)

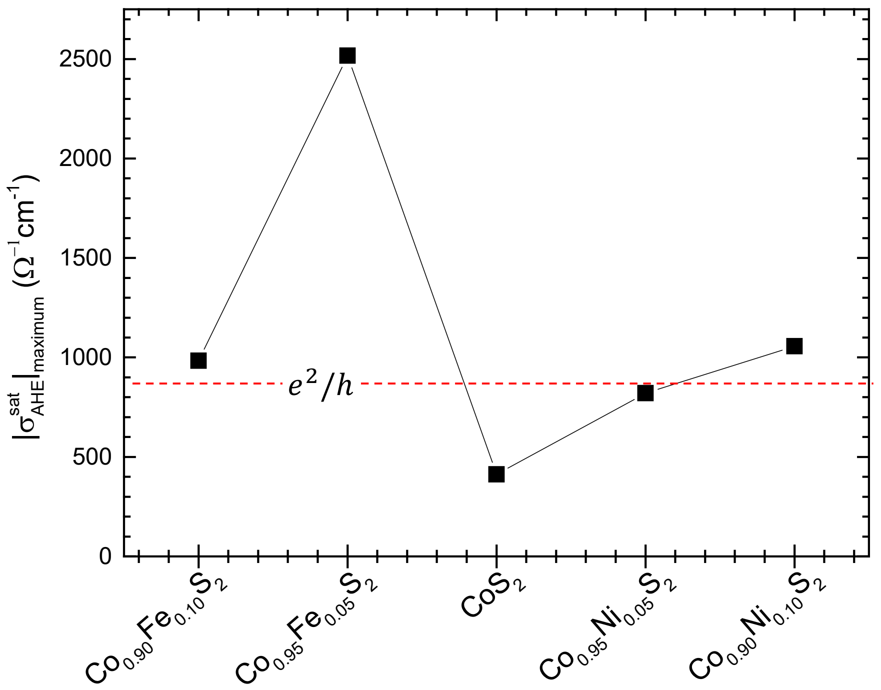


$R_{ordinary}$ (mm^3/C)
0.4
0.0
-0.4
-0.8
-1.2
CoS_2
$Co_{0.95}Fe_{0.05}S_2$
ρ_{AHE}^{sat} (μΩcm)
0
-1
-2
CoS_2
$Co_{0.95}Fe_{0.05}S_2$
0
50
100
150
T (K)

Anomalous Hall conductivity

$Co_{1-x}Fe_xS_2$



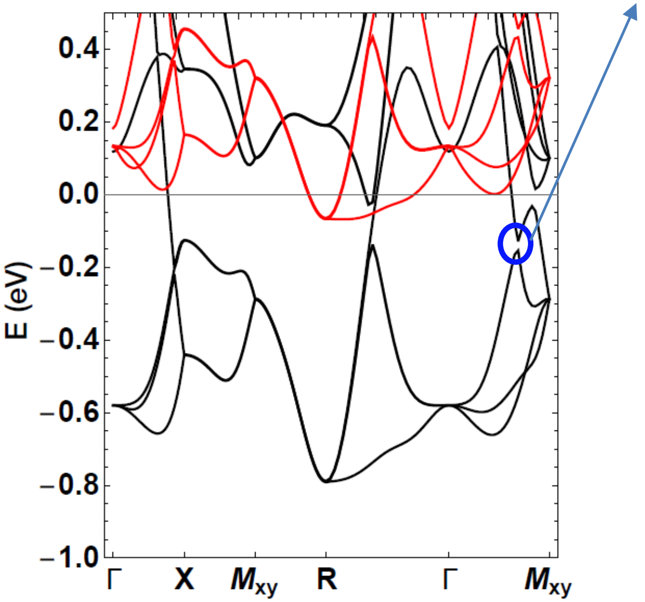


- Why is it large?
- Why does it peak at *x*=0.05?

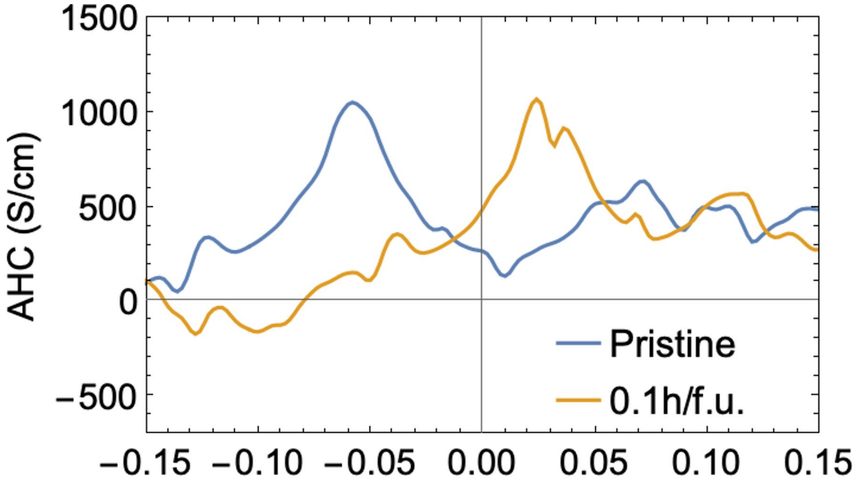
*σ_{AHE} for Co_2MnGa ~ 800 $\Omega^{-1}cm^{-1}$

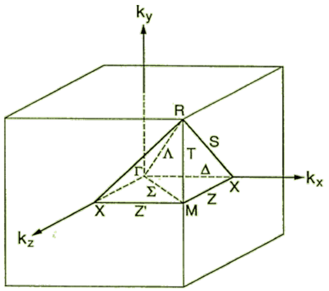
(npj QM **6**, 17(2021))

Calculation



Strong BC source





- Strong BC sources near E_F
- All of them have the same sign (add up)
- Hole doping moves the source to E_F

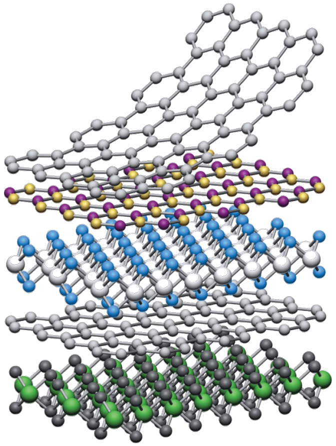
Symmetry protected nodal structures in ultrathin $SrRuO_3$ film

In collaboration with

- Bohm-Jung Yang group (CCES & SNU)
- Taewon Noh group (CCES & SNU)
- Se-Young Park (Soongsil U)

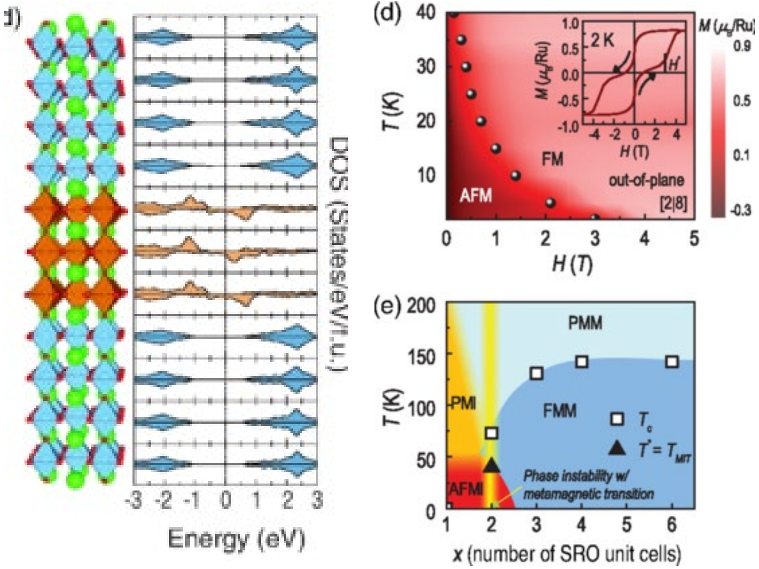
Artificial systems

Exfoliation & stacking

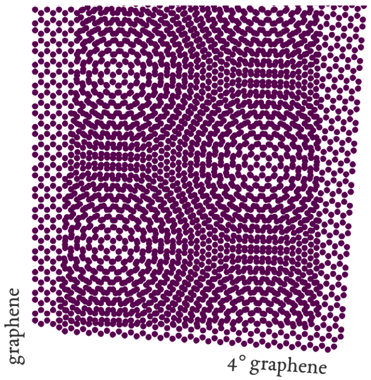


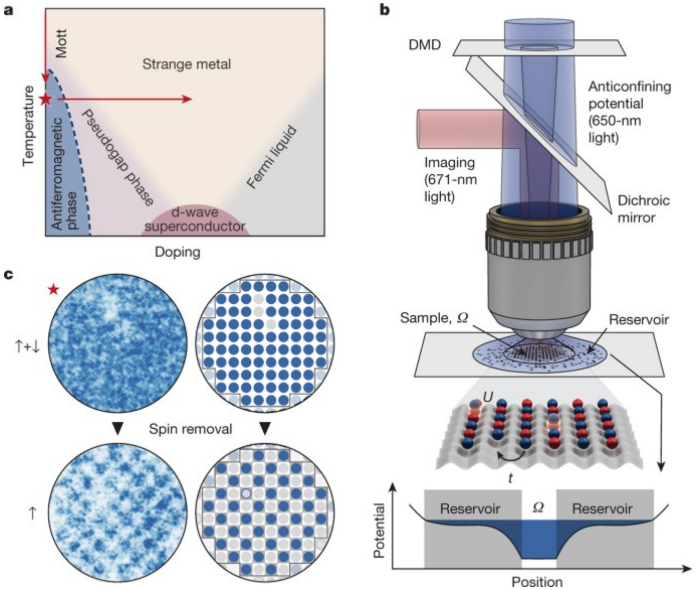
Nature **499**, 419-425 (2013).

Atomically designed superlattices
("Artificial oxide crystal")
PRL **124**, 026401 (2020)



Twisted bilayer





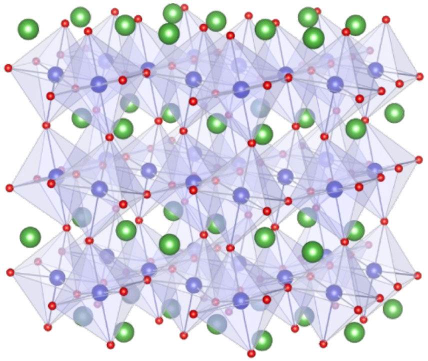
Cold atoms
Fermi-Hubbard AF
Nature **545**, 462 (2017)

Ultrathin film platforms

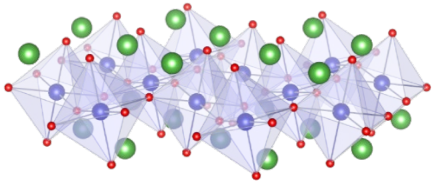
- Tailor the electronic structures via **symmetry breaking**:
  wide range of **tunability** available in **ultrathin films and heterostructures**.

Dimensionality

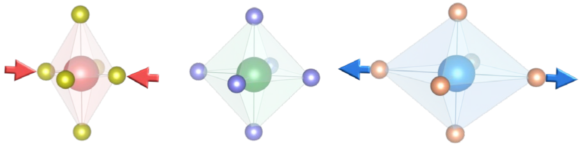
Bulk crystal
3-D



Ultrathin film (monolayer)
2-D limit

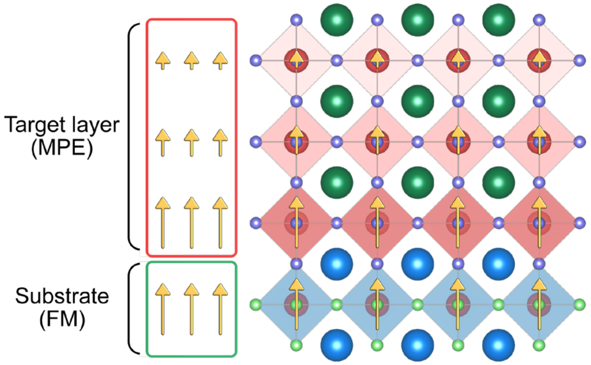


Epitaxial strain



Substrate-induced strain.

Proximity effect at the interface

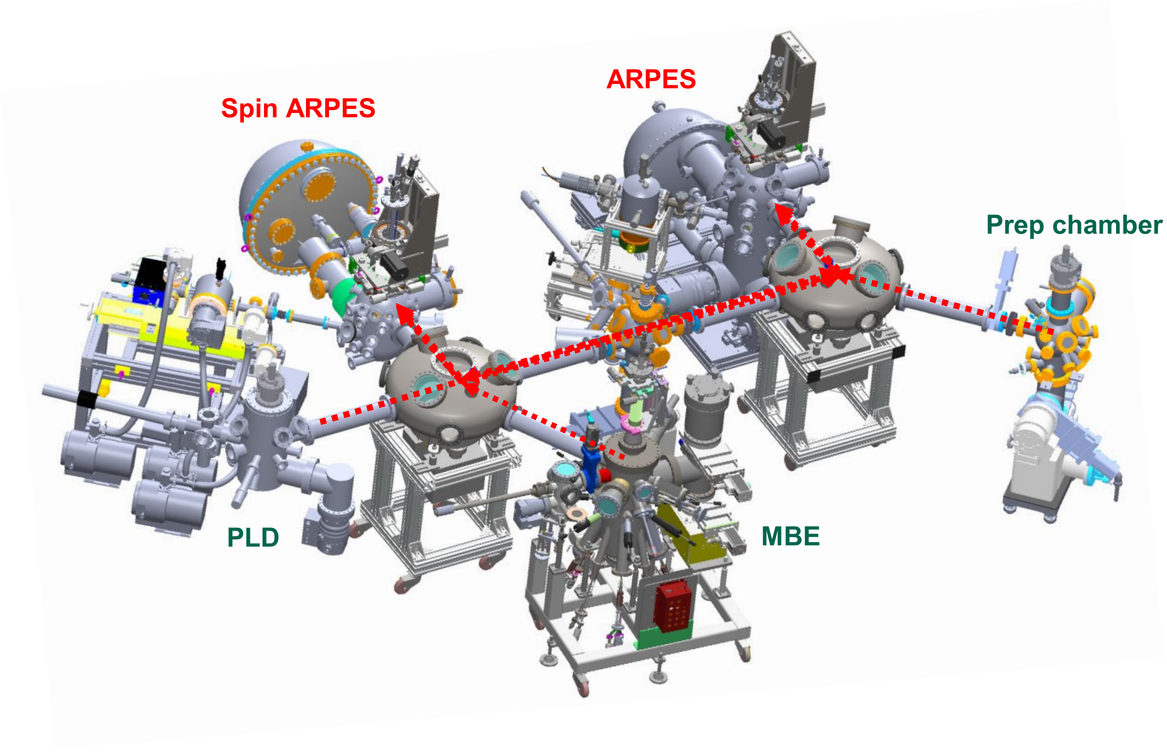


Symmetry-breaking ferromagnetic,
ferroelectric layers as substrates.

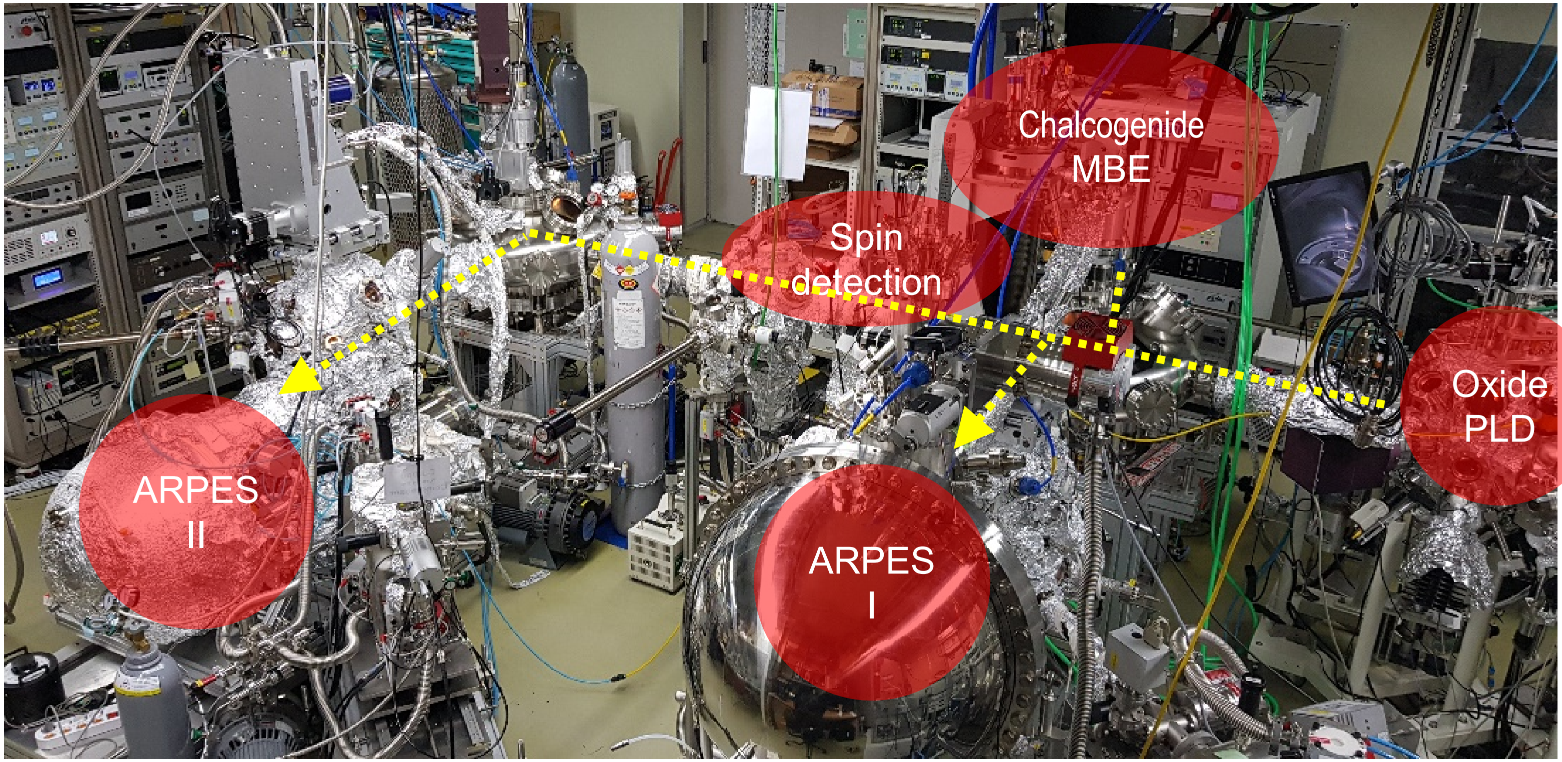
Cluster system for *in-situ* ARPES

Characterize the electronic structure of novel correlated phases via **multi-purpose cluster ARPES system**

- ***In-situ*** AREPS
- ***In-situ*** *s*pin-resolved ARPES

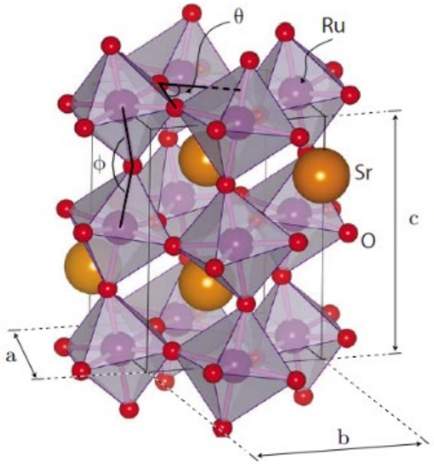


Cluster system for *in-situ* ARPES



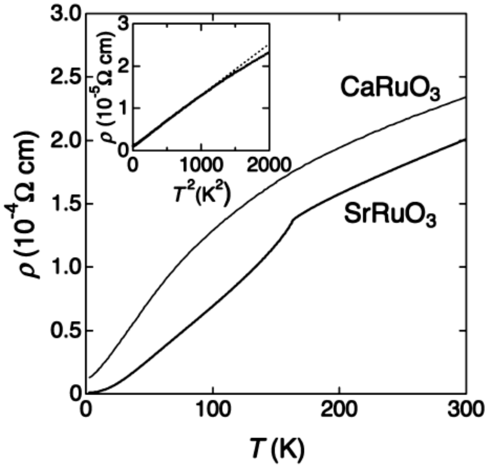
$SrRuO_3$ – oxide ferromagnetic metal

Structure

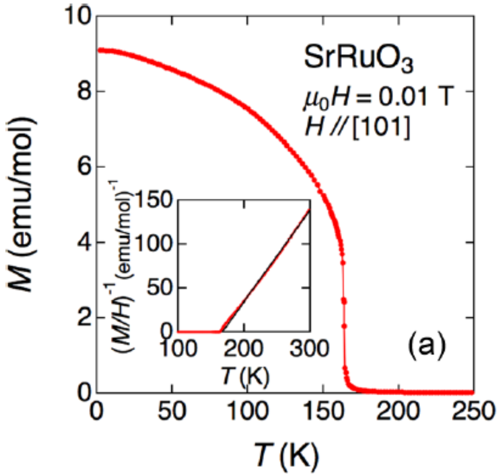


- Pnma , Orthorhombic

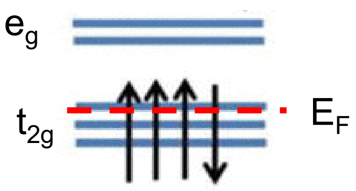
Resistivity



Magnetization



Ru^{4+} 4 electrons in t_{2g}



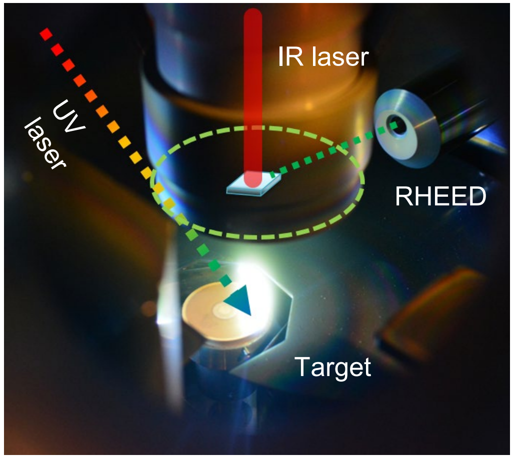
t_{2g} (3↑,1↓) U ~ 2 eV

- **Ferromagnetic** transition at 160 K
- Moderate *U*
- *4d* transition-metal oxide (**spin-orbit coupling**)

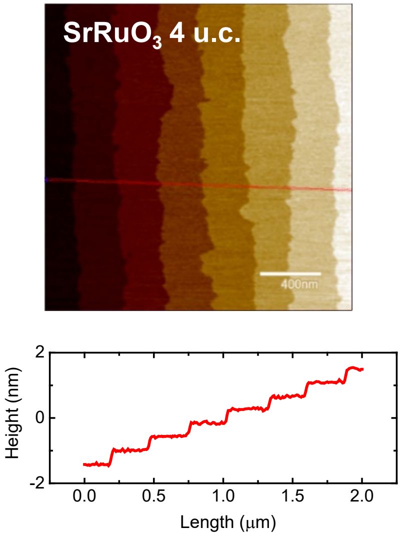
- Q. Gan et al. J. Appl. Phys. 85, 5297 (1999).
- N. Kikugawa et al. Cryst. Growth Des. 15, 5573 (2015).

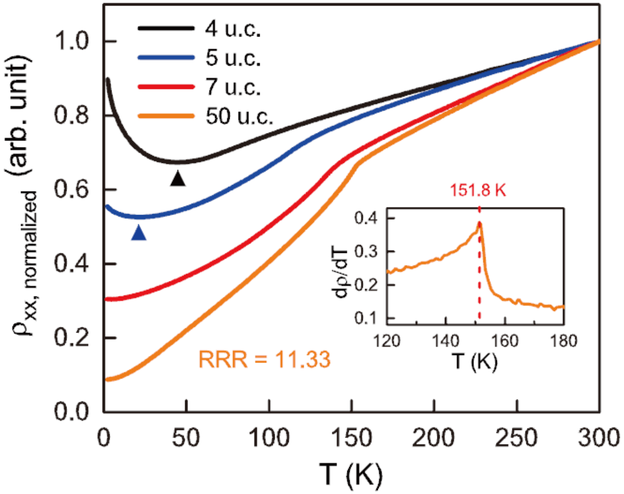
SrRuO$_3$ film growth

PLD growth

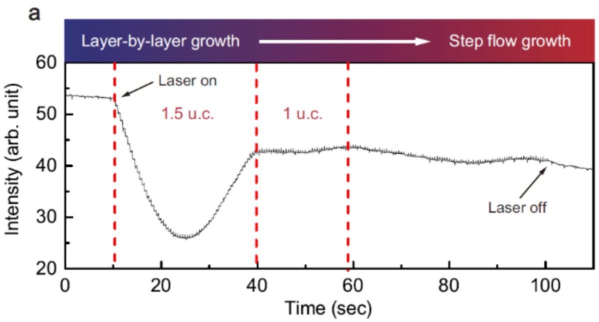


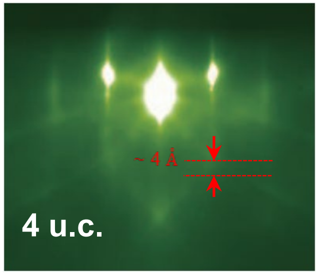
AFM

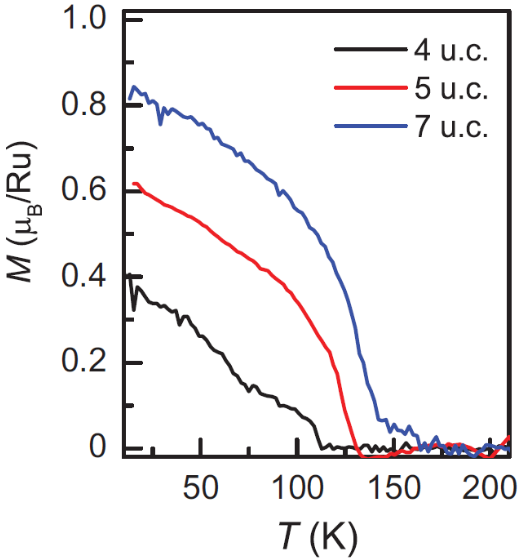




RHEED

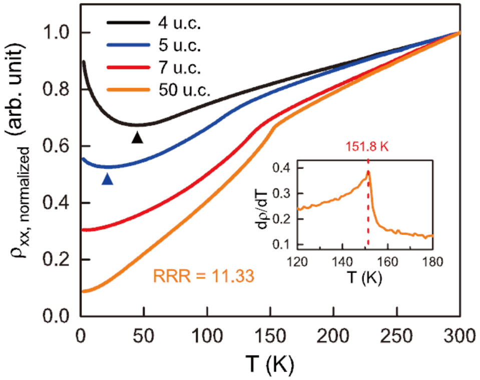


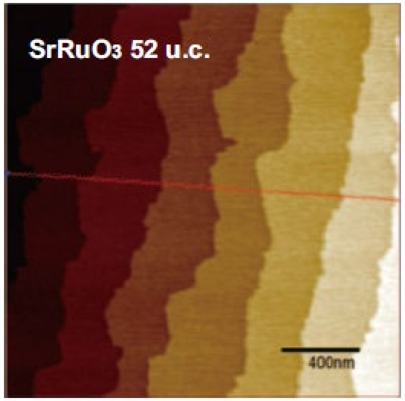


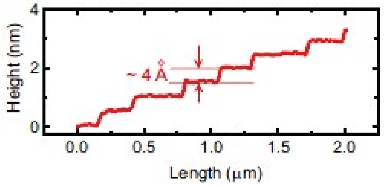


In-situ ARPES on SRO thin-film

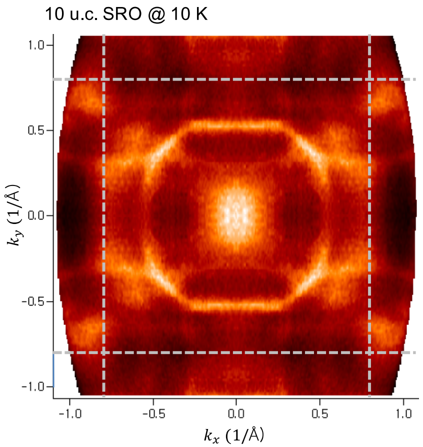
@SNU

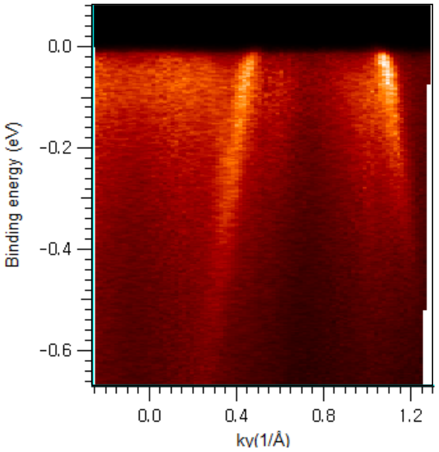






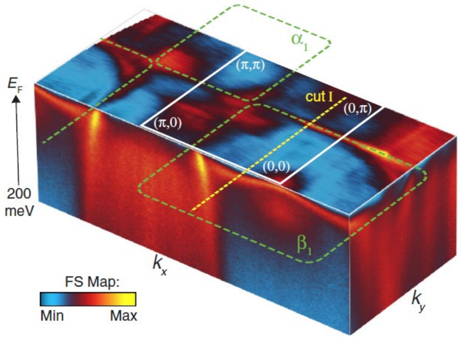
CCES/PLD

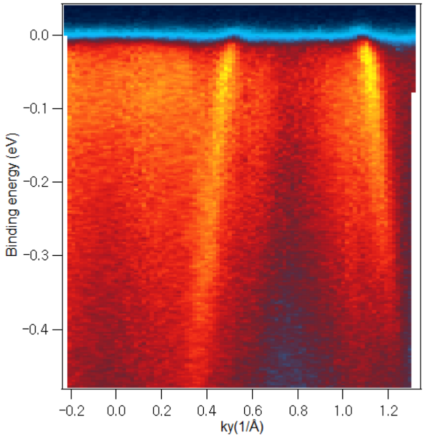




B. M. Sohn et al.

Cornell/MBE





D. E. Shai et al., PRL **110**, 087004 (2013).

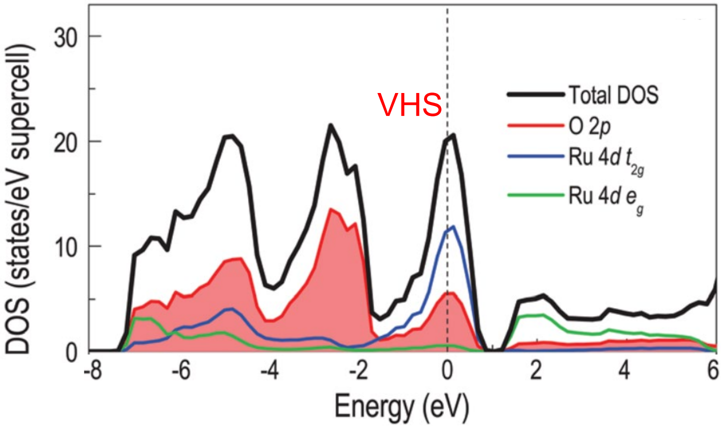
Dual ferromagnetism
(with 15 u.c. films)

References

1. S. Hahn et al., Phys. Rev. Lett. **127**, 256401 (2021)

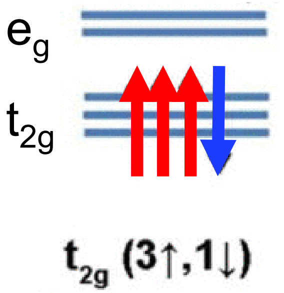
Signature of itinerant magnetism

DOS of $SrRuO_3$



High DOS at E_F favorable for itinerant magnetism

Non-integer magnetic moment



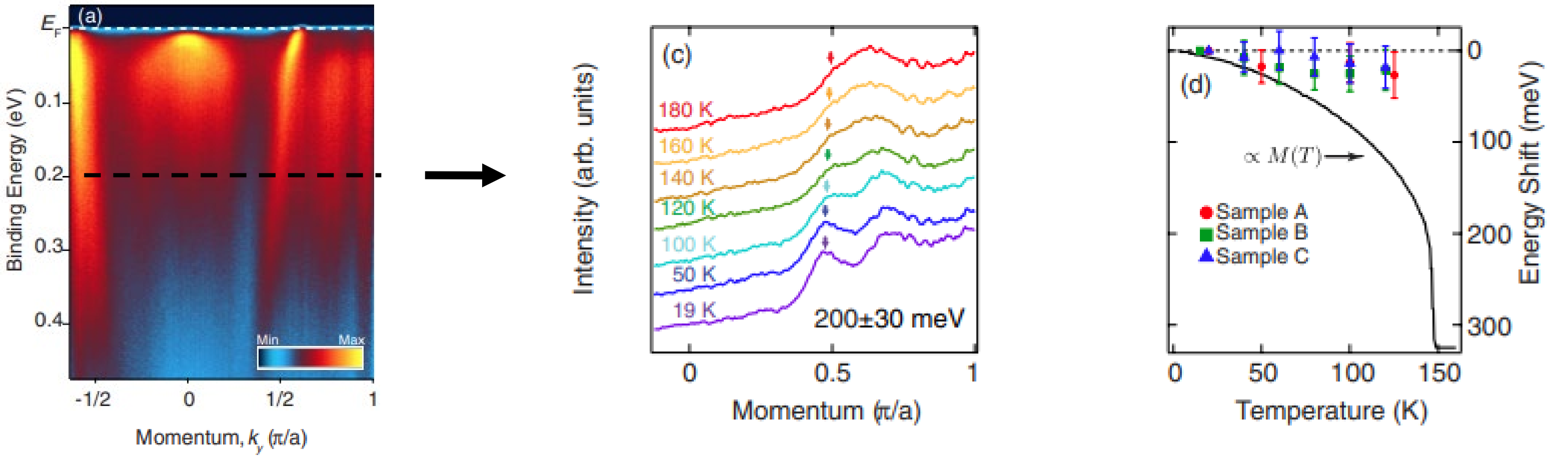
S = 2 expected in the local picture

Measured magnetic moment of ~ 1.6 μ_B

S. A. Lee et al., Energy Environ. Sci. 10, 924 (2017).

Signature of local magnetism

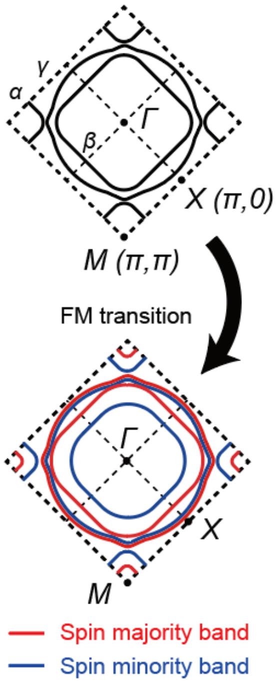
Temperature independent band splitting



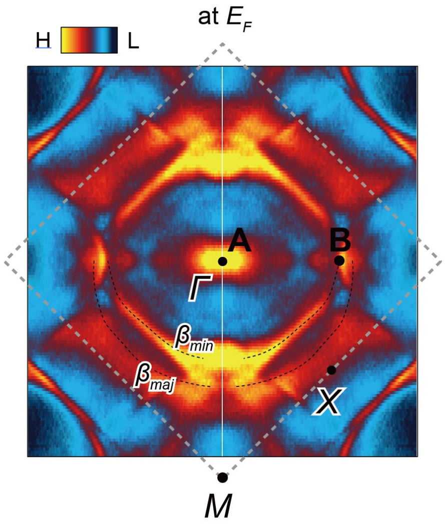
D. E. Shai et al., Phys. Rev. Lett. 110, 087004 (2013).

Itinerant FM feature near E_F

Schematic FS

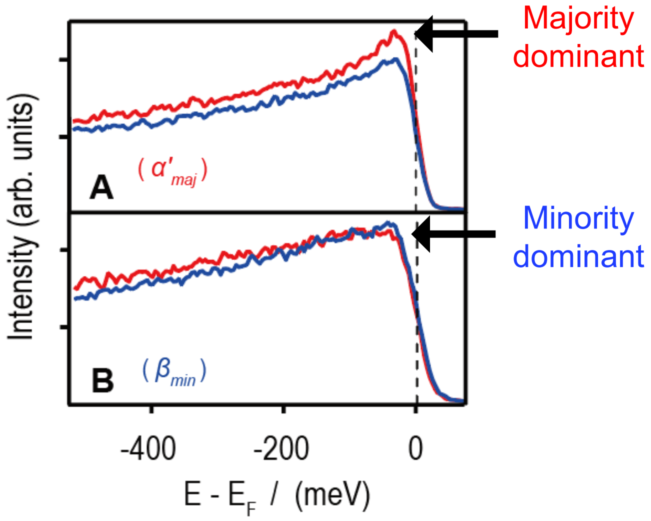


Measured Fermi surface



15 u.c. $SrRuO_3$ film

Spin ARPES

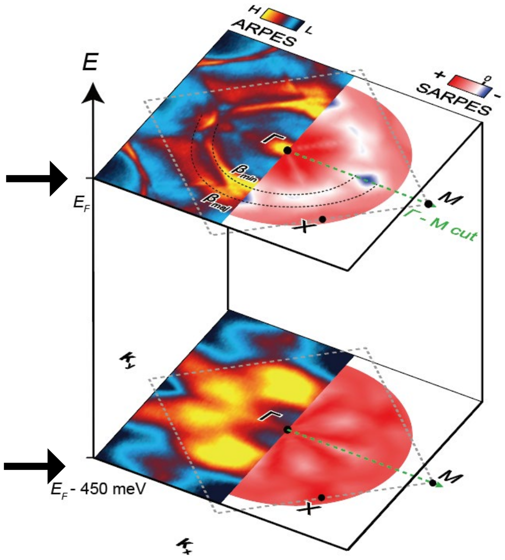


Spin polarization in dispersive bands

Itinerant electrons have momentum dependent spin-polarization.

Localized FM feature at high energy

Energy dependent spin-pol

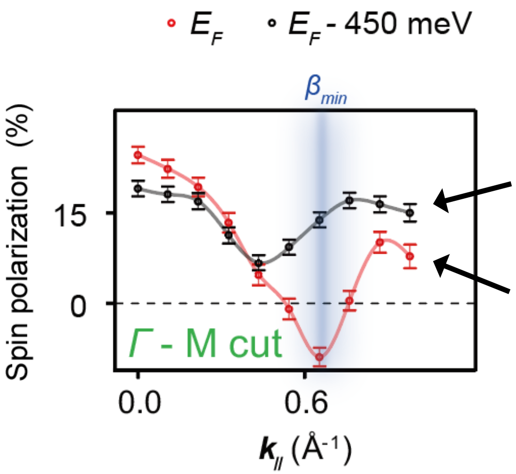


Low E
itinerant
electrons

High E
electrons

High energy electrons are spin-polarized.

Local spin character of high energy electrons



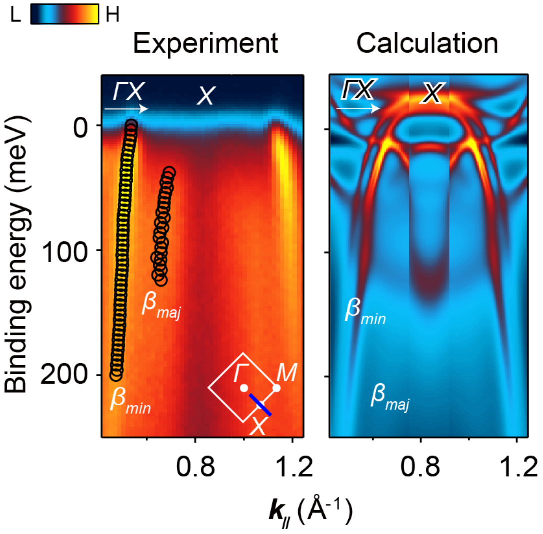
Local spin
(small k dep.)

Itinerant spin
(large k dep.)

Dual character observed!

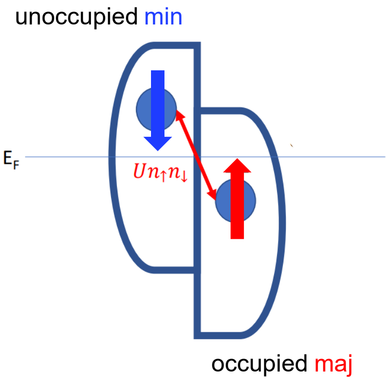
Spin-dependent correlation

Difference in coherence

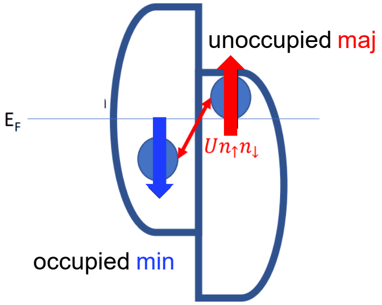


Broad majority and sharp minority bands

Spin-dep Coulomb interaction ($Un_\uparrow n_\downarrow$)



Wide interaction channel
: strong correlation for majority



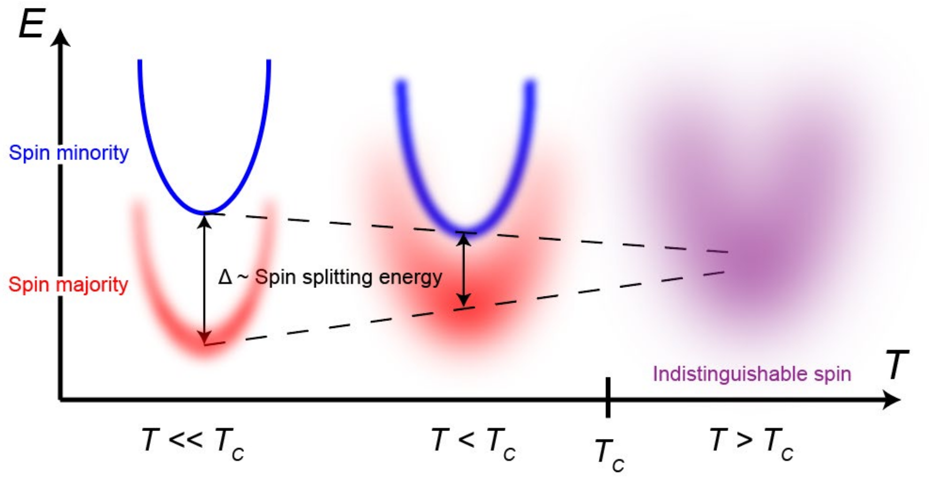
Narrow interaction channel
: weak correlation for minority

Localized spin majority electrons & itinerant spin minority electrons

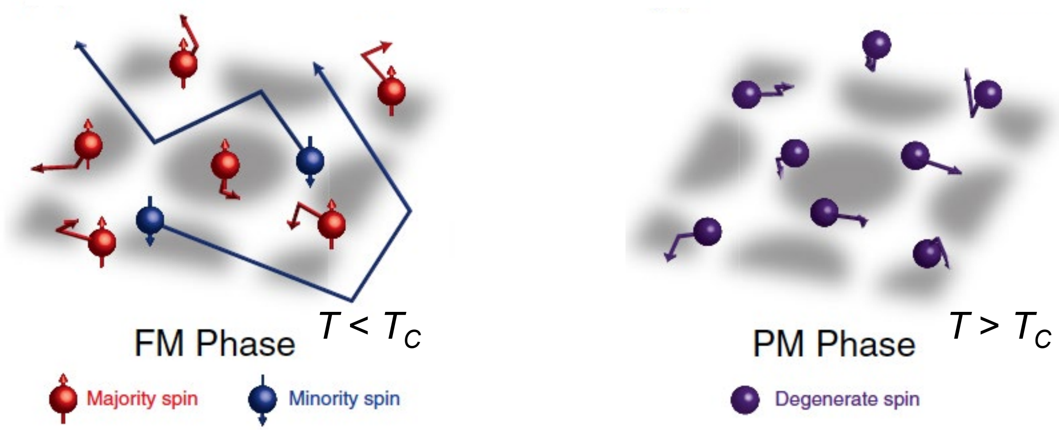
M.J. Kim et al, PRB 91, 205116 (2015)

Pictorial illustration

Momentum space



Real space



Hahn, PRL **127**, 256401 (2021)

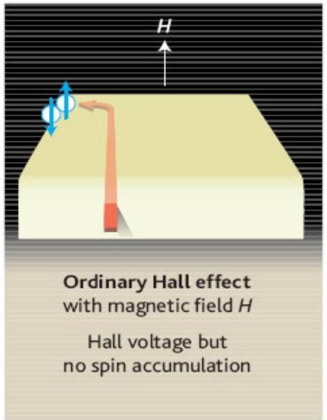
Nodal features
(with 4 u.c. films)

Reference

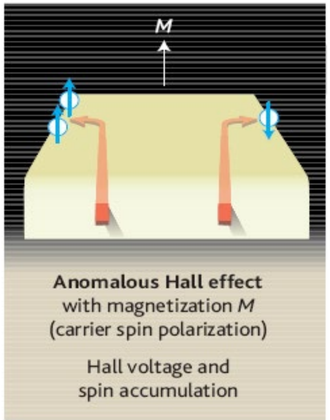
1. Sohn et al, Nature Materials **20**, 1643 (2021)

Magnetic monopoles in SrRuO$_3$

1879

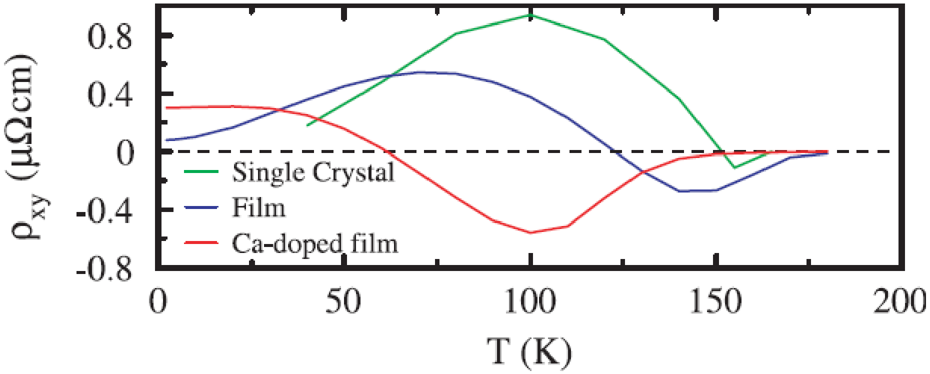


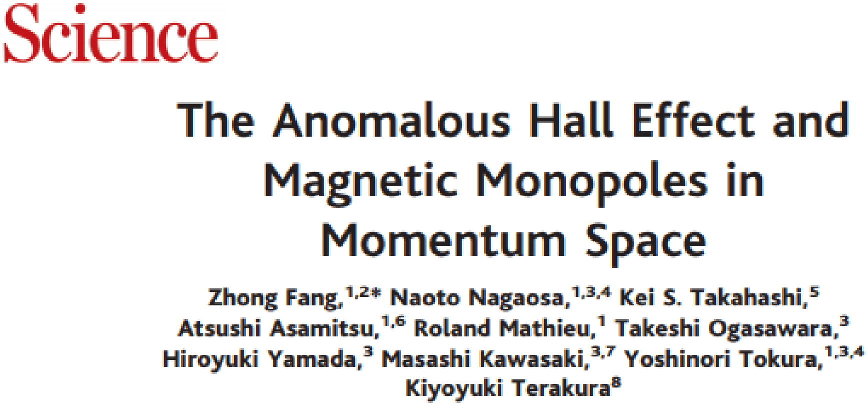
1881



$\text{AHE} \propto \text{M}$

Sign changing anomalous Hall effect in SrRuO$_3$





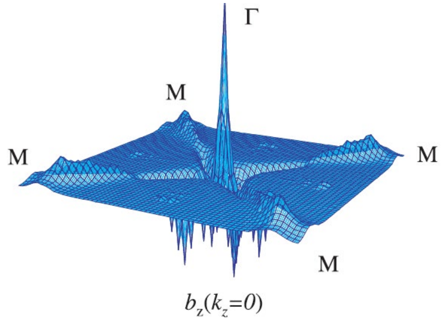
Science

The Anomalous Hall Effect and Magnetic Monopoles in Momentum Space

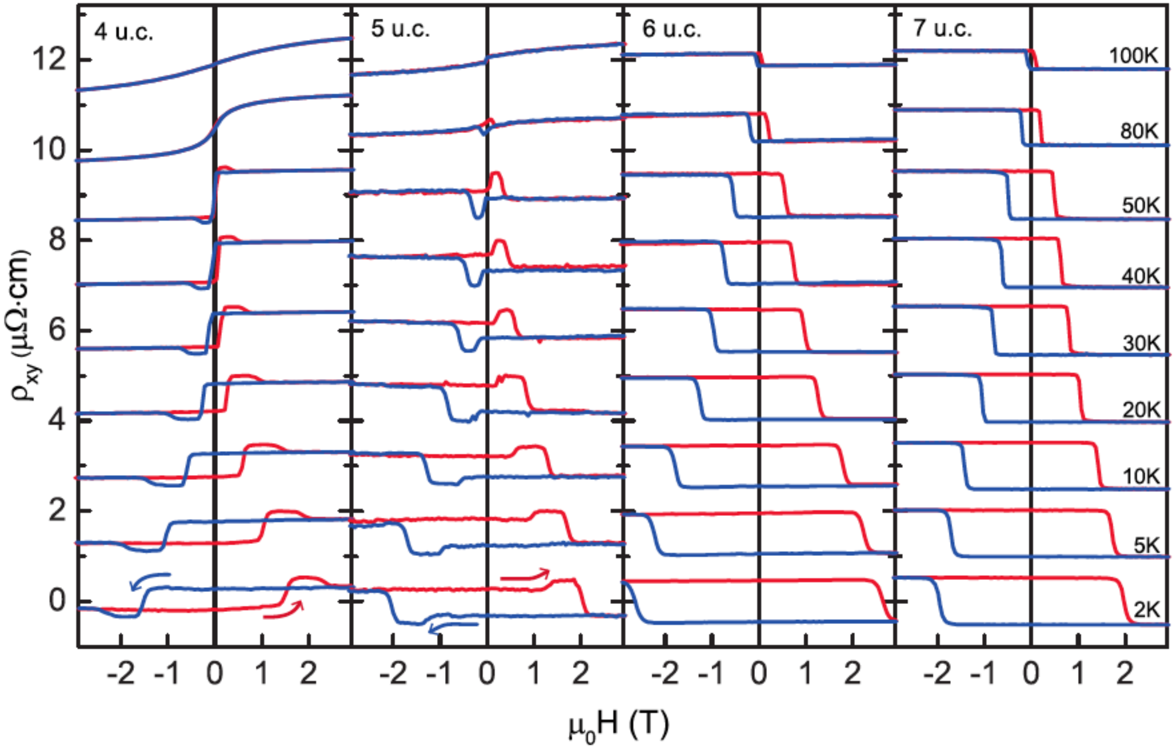
Zhong Fang,[1,2*] Naoto Nagaosa,[1,3,4] Kei S. Takahashi,[5]
Atsushi Asamitsu,[1,6] Roland Mathieu,[1] Takeshi Ogasawara,[3]
Hiroyuki Yamada,[3] Masashi Kawasaki,[3,7] Yoshinori Tokura,[1,3,4]
Kiyoyuki Terakura[8]

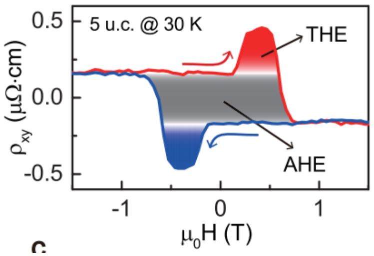
Z. Fang et al., Science **302**, 5642 (2003)

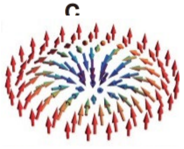
Magnetic monopoles in the momentum space



Anomalous and Topological Hall effects







Topological Hall effect?

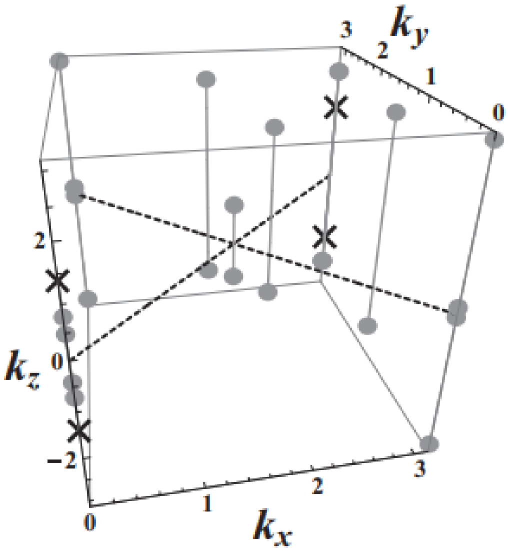
B. Sohn et al., PRR 3, 023232 (2021)

Sohn et al., Current Applied Physics 20, 186 (2019)

1. ‘Topological Hall’ effect in ultrathin films (controversial)
2. Sign changing AHE

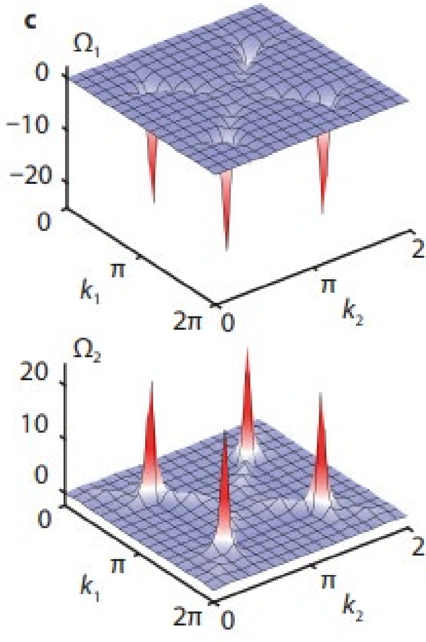
Issues on magnetic monopole in $SrRuO_3$

3D



PRB **88**, 125110 (2013)

2D

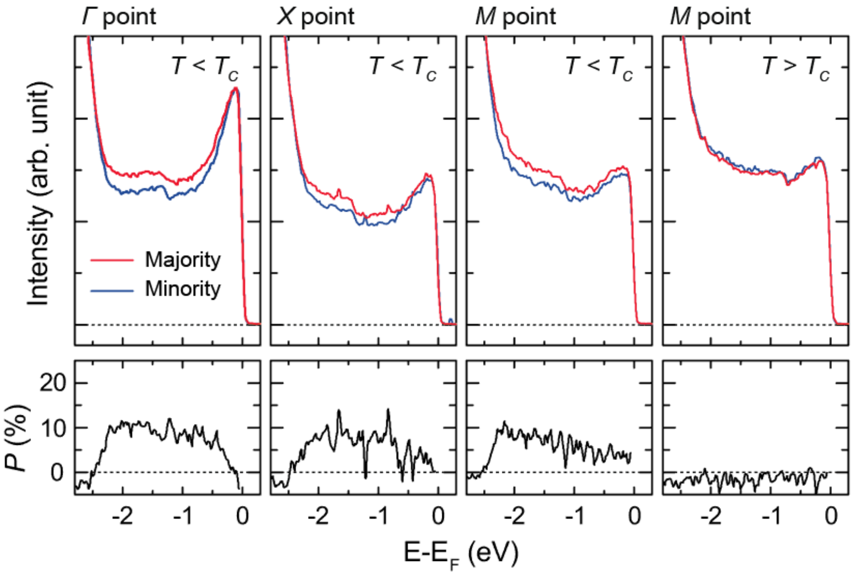


Phys. Rev. Res. 2, 023404 (2020)

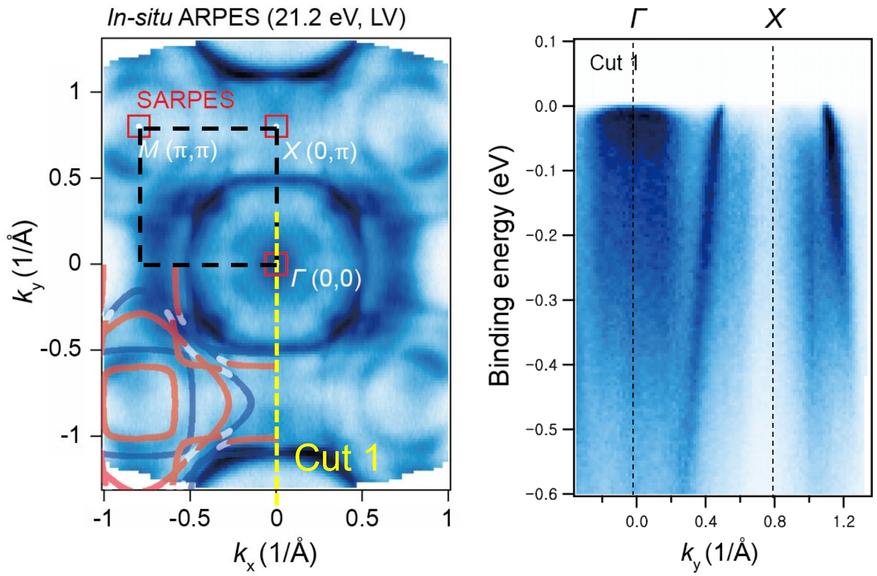
- DFT does not work well
- No experimental dispersions (ARPES)
    - no high quality single crystals for ARPES

Spin polarization & tight binding fit

Spin-resolved ARPES from selected points



Fermi surface map and high symmetry cut



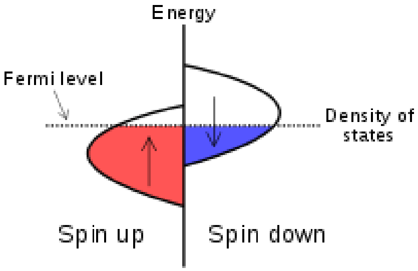
4 u.c. $SrRuO_3$ film

Clear polarization below T_C

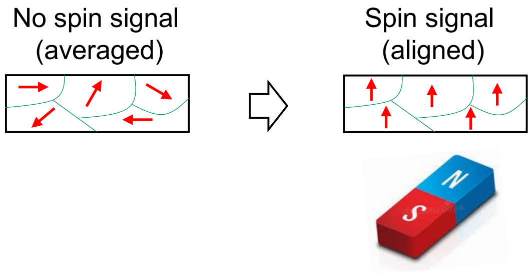
$$H = \sum_k [\underbrace{\epsilon^a_{\boldsymbol{k}\sigma}\,\delta_{ab}\delta_{\sigma\sigma'}}_{\text{Nearest}} + \underbrace{f^{ab}_{\boldsymbol{k}}\delta_{\sigma\sigma'}}_{\text{Next-nearest}} + \underbrace{i\lambda\epsilon^{abc}\tau^c_{\sigma\sigma'}}_{\text{SOC}}]\, d^\dagger_{\boldsymbol{k}a\sigma} d_{\boldsymbol{k}b\sigma}$$

(Considering only t_{2g} orbitals)

2D tight-binding fit

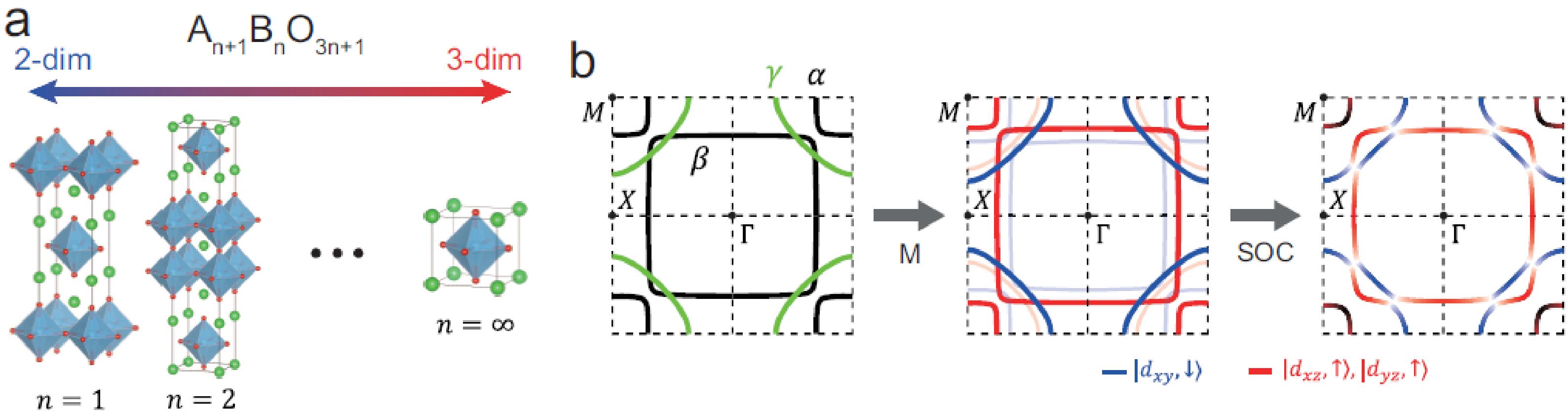


In situ magnetization



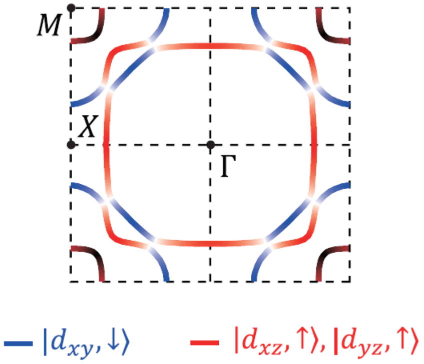
Electronic structure of single layer

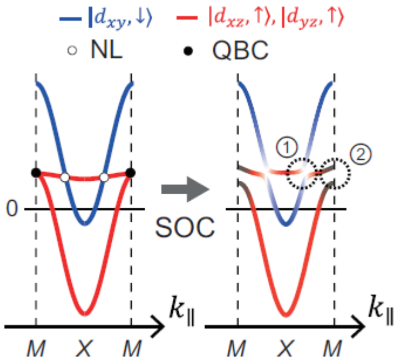
Band structure of 2D ferromagnetic Ruddlesden-Popper phases



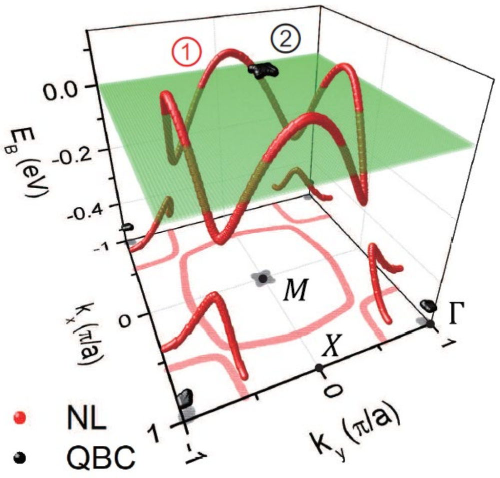
Berry curvature sources

TB fit and analysis





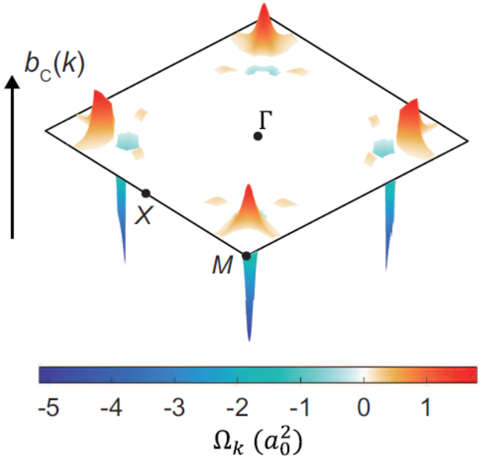
Berry curvature sources



① Nodal lines (β - γ band crossing)

② Quadratic band crossing
(xz – yz band crossing)

Calculated Berry curvature
at the Fermi level



- Different sign of BC near E_F
- BC mostly from QBC in 2D FM perovskite

(Also see PRR 2, 023404 (2020))

Quadratic band crossing (QBC) near the M point responsible for the sign-tunable AHE.

Berry curvature, OAM & Circular dichroism

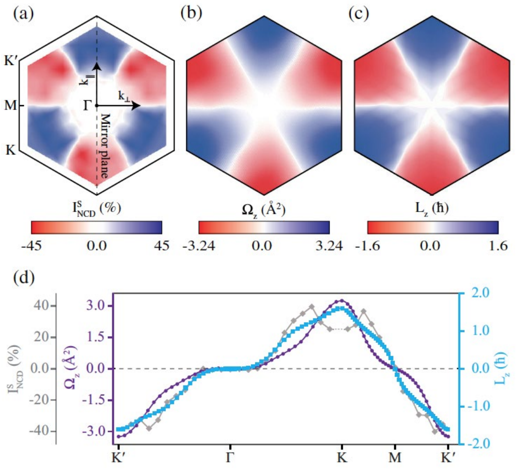
Berry curvature & OAM

$$\vec{A}(\vec{k}) = \lambda_p^s \vec{k} \times \vec{L}$$

$$\vec{B}(\vec{k}) \propto \vec{L}$$

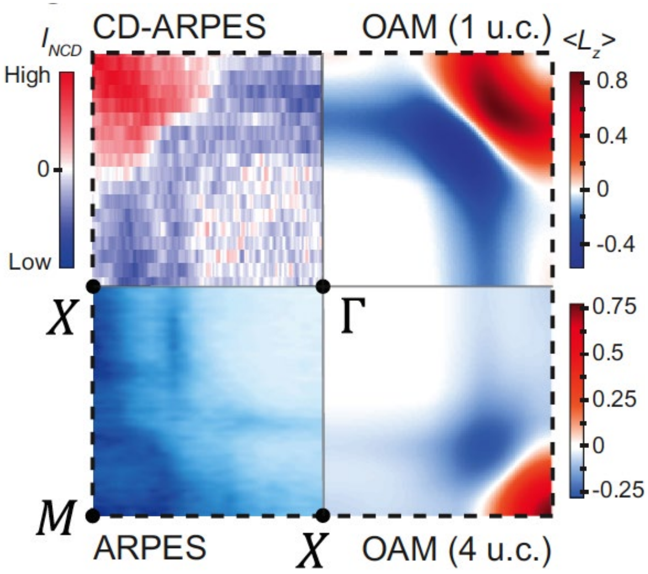
PRL **121**, 086602 (2018)

OAM & Circular dichroism



"Hidden Berry curvature in WSe$_2$"

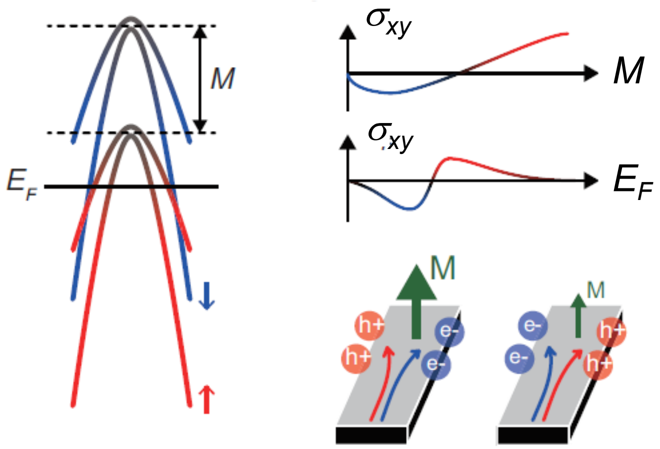
PRL **121**, 186401 (2018)



*More direct evidence for existence of Berry curvature

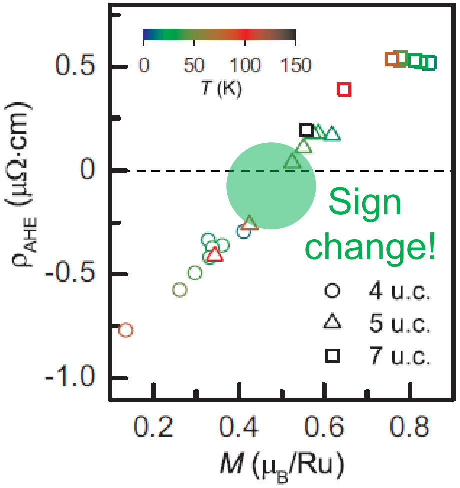
Origin of sign-tunable AHE

QBC induced sign-changing AHE



QBC responsible for sign changing AHE

Magnetization-dependent AHE



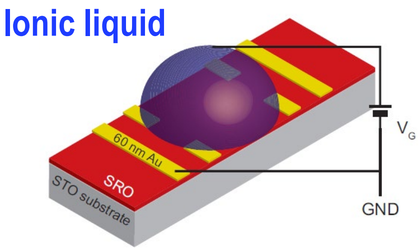
Sign change!

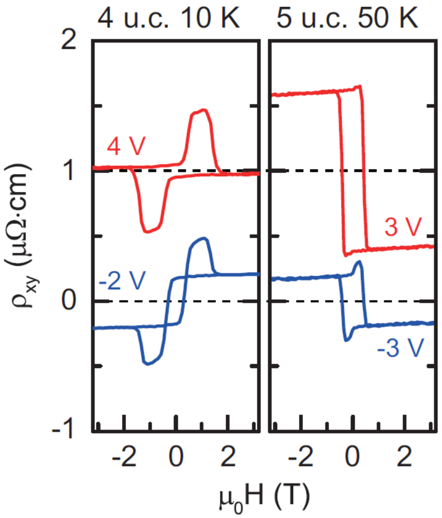
M determines AHE

Sohn et al., Nature Materials (2021)

Thickness & ionic gating dependent AHE sign

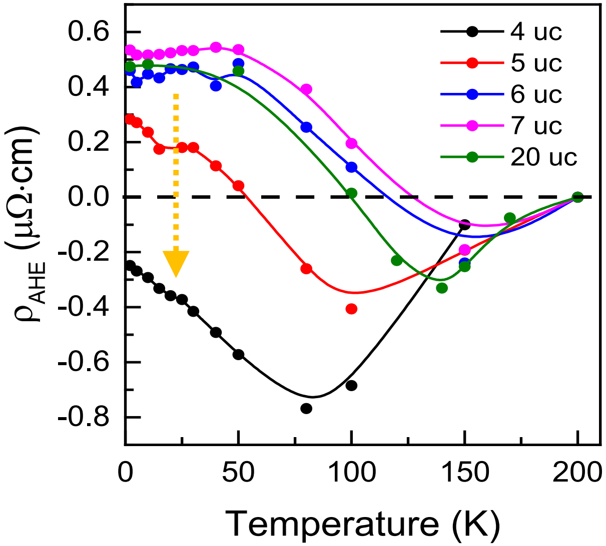
Chemical potential-dependent AHE by **ionic-liquid gating**





Kim et al., APL 118, 173102 (2021)

Thickness-dependent AHE



Sign changing AHE in ultra-thin limit

Future studies – proximity (strain)

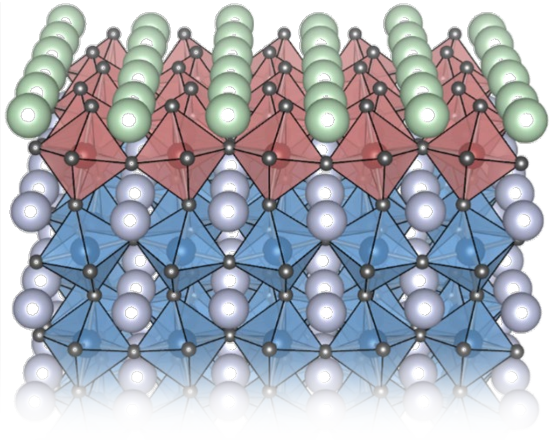
References

1. Nat Comm **12**, 6171 (2021) (Our 1st ARPES work on 1 uc film)
2. ArXiv : 2203.04244 (Kim et al, control of MIT through octahedron distortion)
3. In preparation (Ko et al, control of MIT through strain)

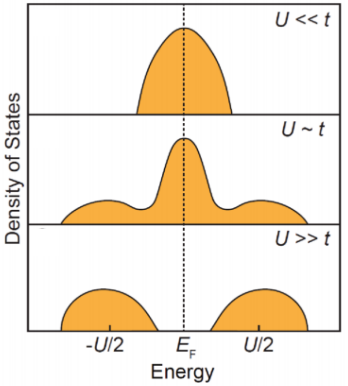
Structure control of 1 u.c. SRO

Metal-to-insulator transition in 1 u.c. SRO by octahedron distortion?

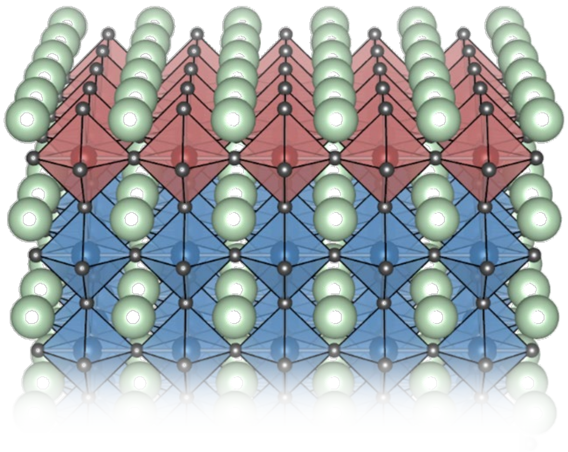
Monolayer $SrRuO_3/CaTiO_3$
(SRO/CTO)



Mott transition with increasing U/t



Monolayer $SrRuO_3/SrTiO_3$
(SRO/STO)



Increasing OR

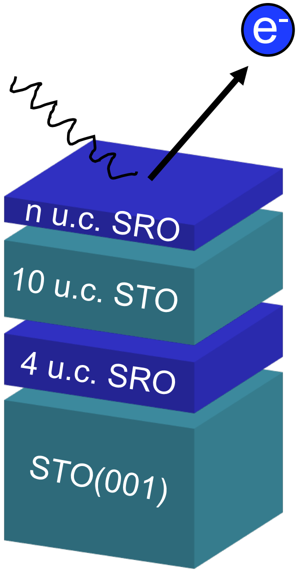
Decreasing bandwidth, *t*

- **Electronic ground state?**
- $CaTiO_3$ buffer layer with oxygen octahedral rotation (OOR)

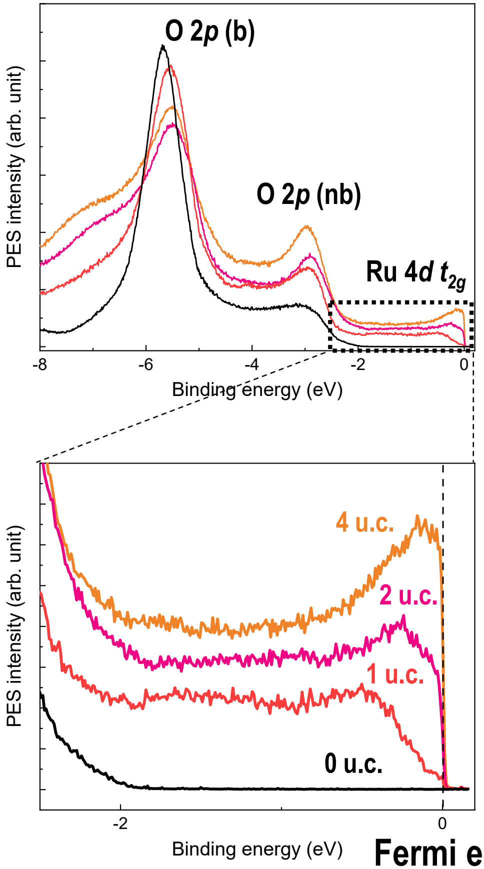
- **Metal**
- Cubic $SrTiO_3$ buffer layer

S. Y. Kim, *et al*., Adv. Mater. **30**, 1704777 (2018).

Interface-driven MIT in 1 u.c. films

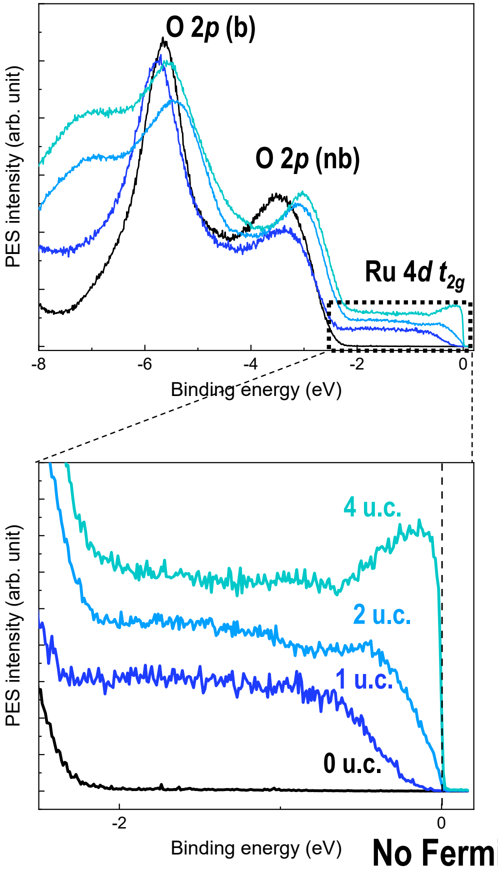


PES spectra of ultrathin SRO/STO

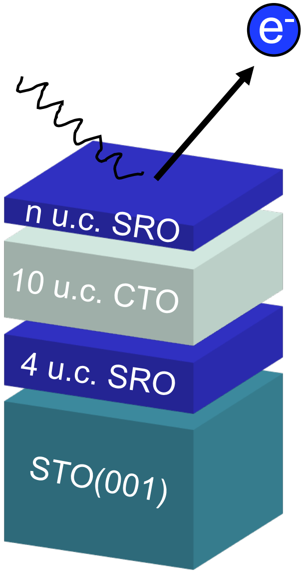


Fermi edge!

PES spectra of ultrathin SRO/CTO



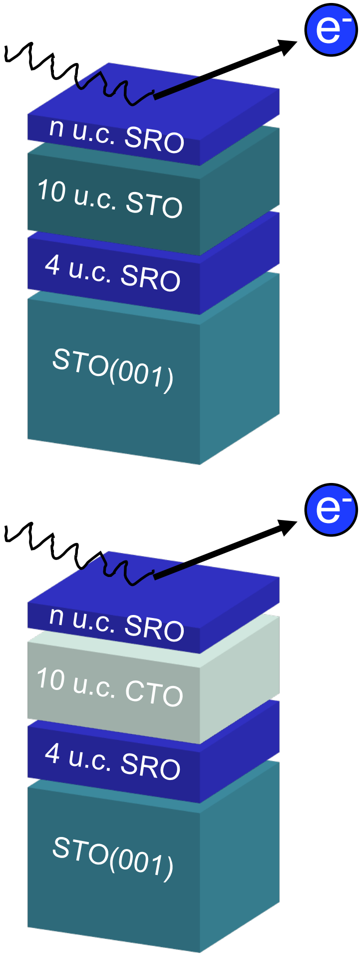
No Fermi edge!

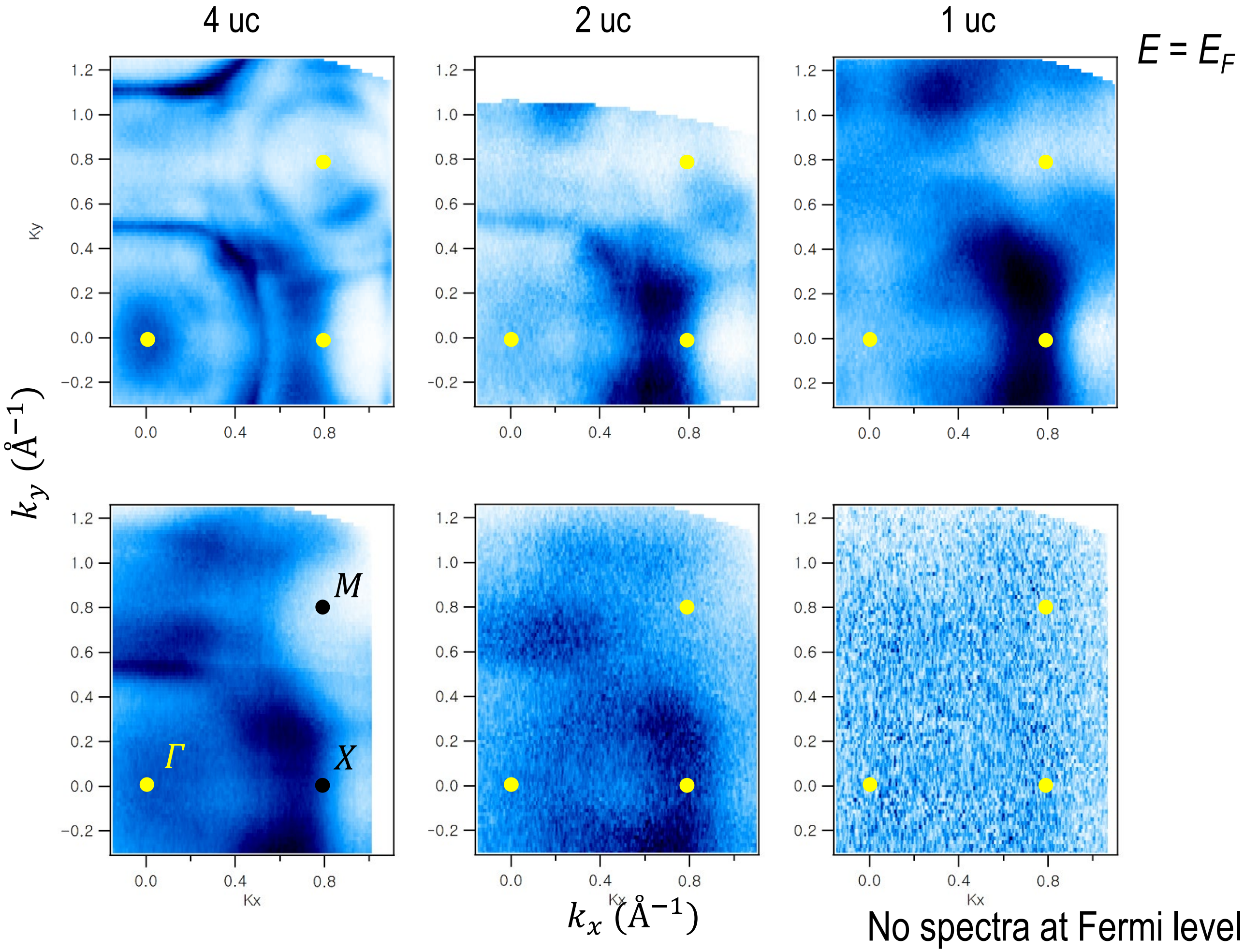


Metal-to-insulator transition in monolayer SRO!

Nat Comm **12**, 6171 (2021)

Fermi surfaces





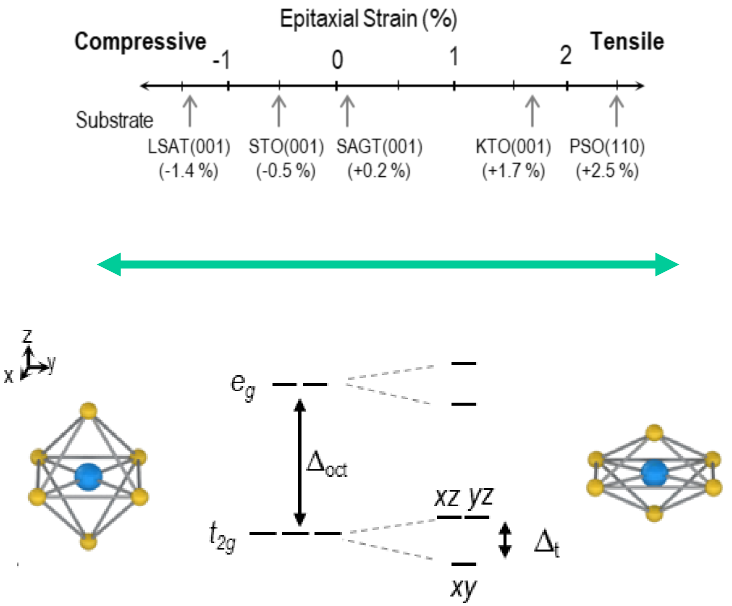
No spectra at Fermi level!

Control MIT in monolayer SRO!

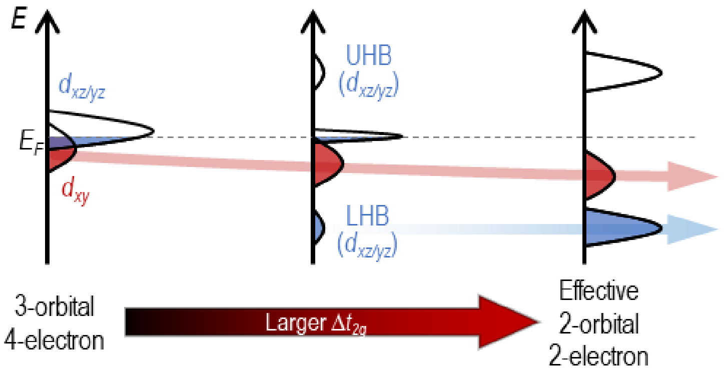
J R Kim et al., ArXiv : 2203.04244

Strain engineering of monolayer $SrRuO_3$

Orbital tuning with strain engineering



Strain-dependent band structure



4 electrons / 3 orbitals

2 electrons / 2 orbitals

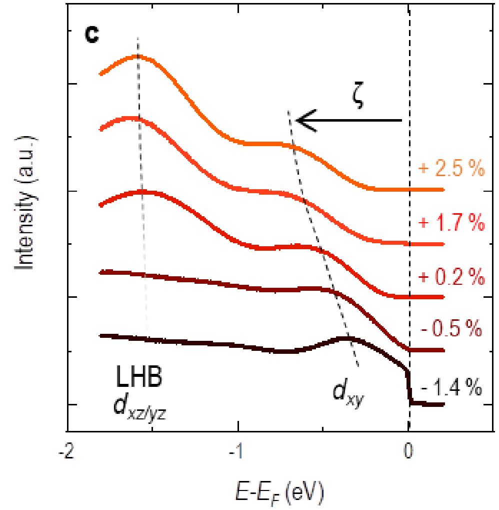
Larger Δ_t with strain

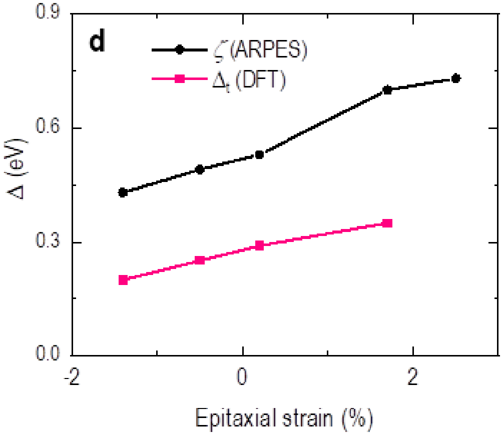
Fermi liquid
(weakly correlated)

Correlated metal
(Hund's system)

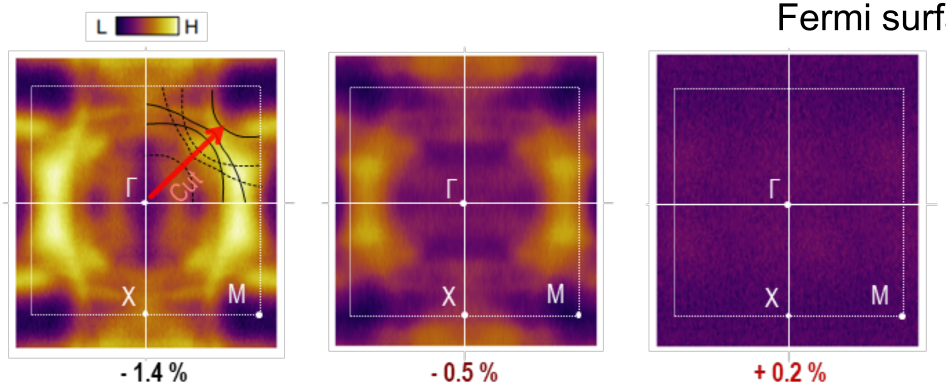
Mott Insulator
(Hund's assisted Mott state)

Strain dependent electronic structure

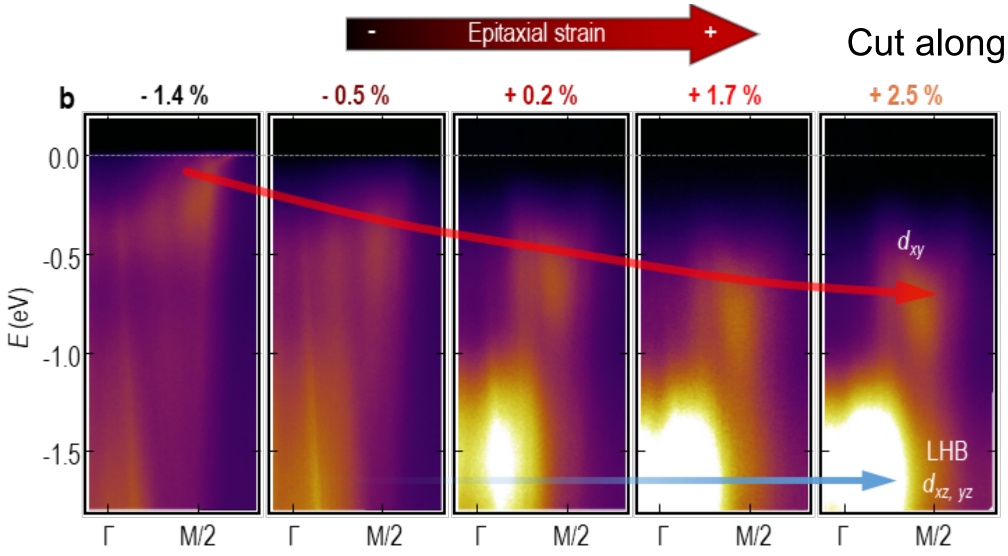




Fermi surfaces



Cut along ΓM



Large Mott-gap size unexpected from DFT

E K Ko, in progress

Summary

Large anomalous Hall conductivity in CoS_2

- Few, small gap, near E_F, same sign BC sources

Symmetry protected nodal structures in ultrathin $SrRuO_3$ film

- Nodal lines and QBCs generic to pervskite oxides

“Tunable Anomalous Hall conductivity”

Collaborators

ARPES

Byungmin Sohn, Sungsoo Hahn, Donghan Kim, Young-Do Kim

Wonshik Kyung, Yoonsik Kim, Hanyoung Ryu and Soonsang Huh

Jinwoong Hwang, J D Denlinger, Jiseop Oh, Eli Rotenberg

Crystal/Thin-film growth

Mikyung Kim

Byungmin Sohn, Sungsoo Hahn, Donghan Kim, Eun-Kyo Ko, Jeong Rae Kim, Tae Won Noh

Transport and magnetic measurement

Mikyung Kim

Byungmin Sohn, Bongju Kim, Tae Won Noh

Joonyoung Choi, Younjung Jo

Ionic-liquid gating

Minsoo Kim, Donghan Kim

Theory

Bohm-Jung Yang, Eunwoo Lee

Se Young Park, Choong Hyun Kim

Ji Hoon Shim, Minjae Kim

Joon Won Rhim, Jinhong Park