Emergent phenomena of $SrRuO_3$ and their control in the 2-dimensional limit

Tae Won Noh

Center for Correlated Electron Systems, Institute for Basic Science (IBS), Seoul 08826, Korea

Department of Physics and Astronomy, Seoul National University, Seoul 08826, Korea

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Content

1. Introduction
    - ☐ Emergent phenomena in transition metal oxides and their heterostructures
    - ☐ Our experimental setup : cluster system for PLD, *in-situ* ARPES, RHEED & LEED
    - ☐ Basic Properties of $SrRuO_3$

2. MIT in $SrRuO_3$ ultrathin films
    - ☐ Fundamental thickness limit of SRO films
    - ☐ Control of MIT in 2D SRO films (i.e. 1 *u.c*) by controlling the titling angle
    - ☐ Strain Engineering of electronic properties of 2D SRO films

3. Dual ferromagnetism in $SrRuO_3$ thick films

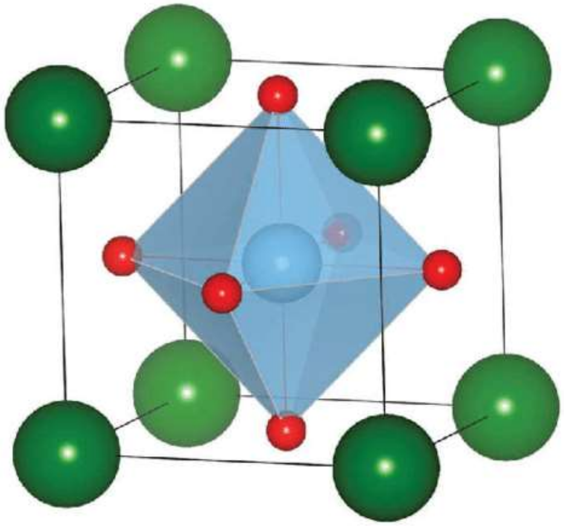
4. Topological band structures of $SrRuO_3$: Nodal features

5. Summary and Outlooks

Main Question in oxide electronics

How to discover & control novel emergent phenomena in TMO and their heterostructures?

Perovskite Oxides (ABO_3)



● *A* : Sr, Ca, Pb, Bi, ….

● *B* Transition metal ions

● *O* Oxygen

Emergent phenomena

- Ferroelectricity
- Ferroelasticity
- High-*k* dielectric
- Multiferroicity
- Flexoelectricity
- Magnetism
- Metal-insulator transition
- Correlated topological phases
- (Novel) Superconductors
- Resistance change, TER
- CMR, TMR, and GMR 2DEG
- Thermoelectric ….

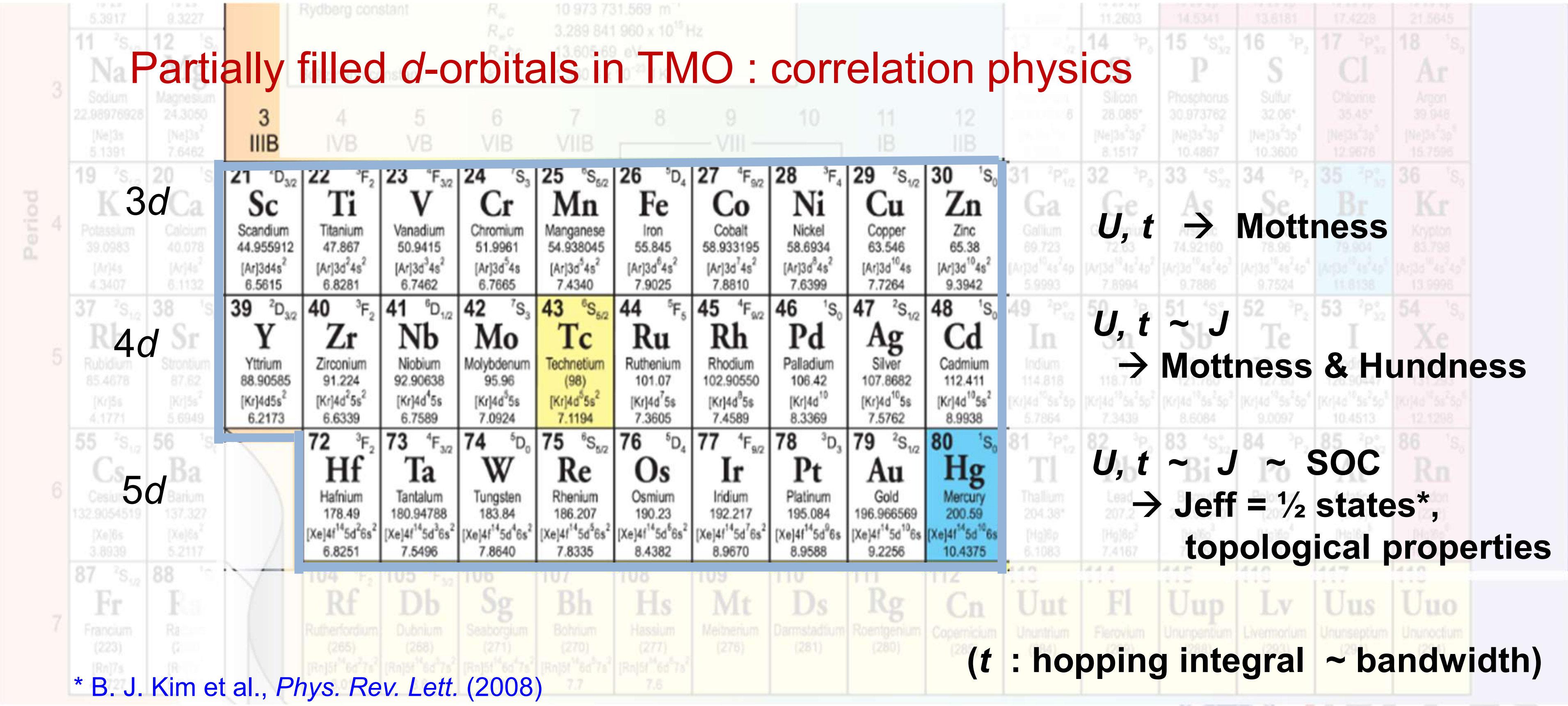
⋮

- BO_6 octahedra form a connecting network in 3D
- Filling of *d* electrons in *B* ions → emergent phenomena

Important interactions in transition metal oxides

Electronic correlations : On-site Coulomb interaction (U)
& Hund's rule coupling (J)

Topological effects : Spin-orbit coupling interaction (SOC)



Partially filled *d*-orbitals in TMO : correlation physics

3*d*

4*d*

5*d*

U*, *t → **Mottness**

U*, *t* ~ *J
→ **Mottness & Hundness**

***U*, *t* ~ *J* ~ SOC**
→ **Jeff = ½ states*,**
topological properties

(*t* : hopping integral ~ bandwidth)

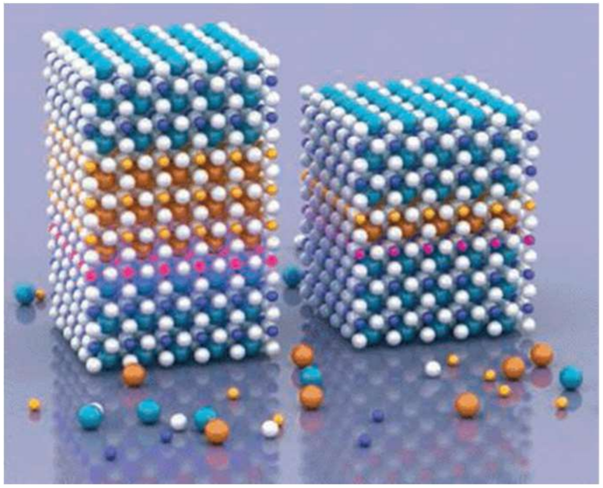
* B. J. Kim et al., *Phys. Rev. Lett.* (2008)

Center for Correlated Electron Systems

TMO heterostructures : a platform for searching novel EP

Design of artificial Transition Metal Oxides (TMOs) heterostructure

Novel emergent phenomena (EP) from TMO heterostructures



Controlling degrees of freedom

Charge

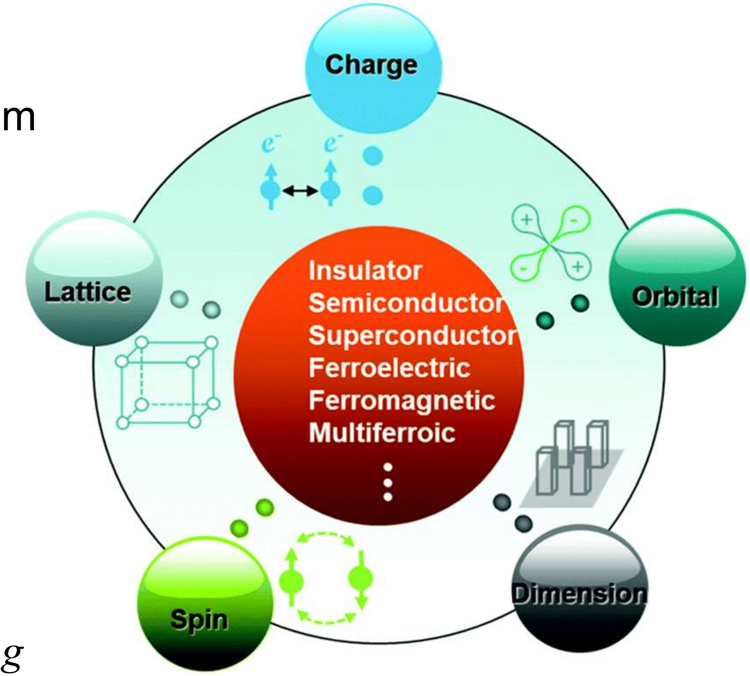
Spin

Lattice

Orbital

Dimension

Spin-Orbit coupling



J. M. Rondinelli *et al*, *MRS Bulletin*. (2012).
E. Dagotto *et al.*, *Science* (2007)
H. Y. Hwang *et al.*, *Nat. Mater.* (2012)

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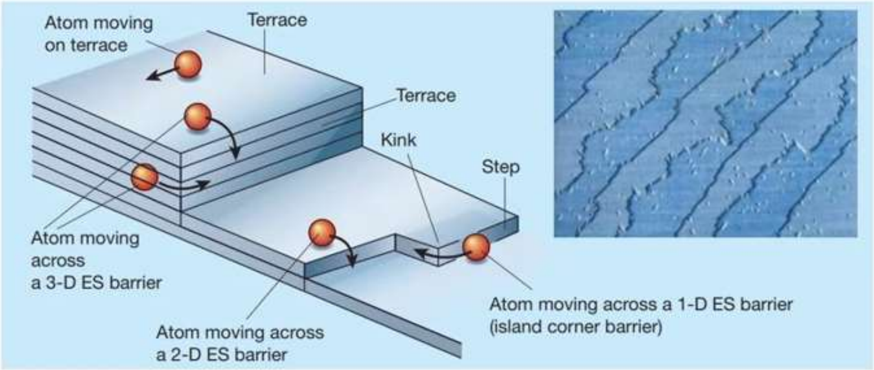
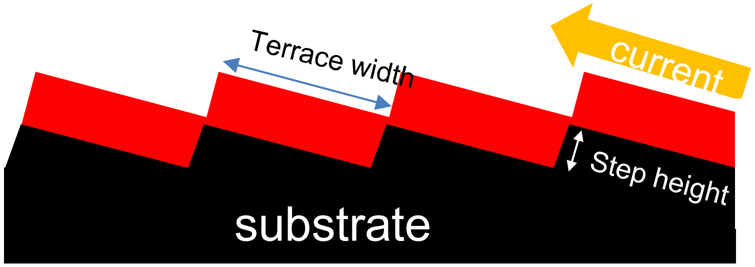
Difficulties on transport measurements of ultrathin TMO films

- ☐ Conducting channel broken near the step-edge of substrates
- ☐ Metal to insulator transition (MIT) in approaching the single atomic layer limit
  : correlation-driven Mott insulating phases or localization effects





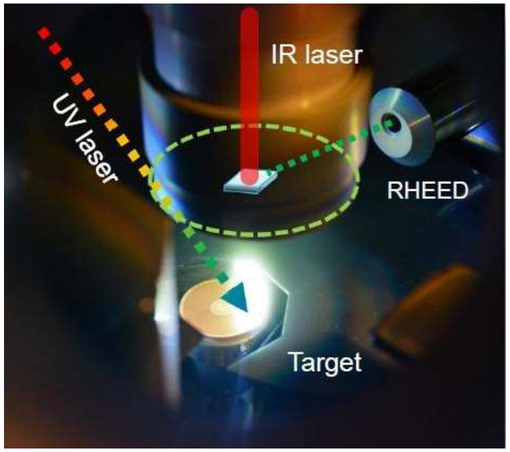
Inevitable broken channels near step edge!

➔ difficult to measure transport properties

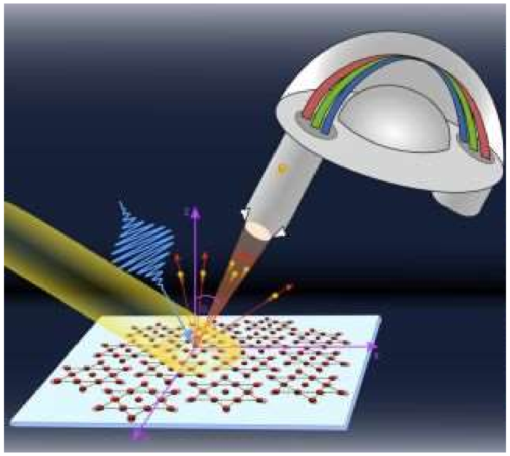
M. G. Lagally and Z. Zhang, *Nature* (2022)

Cluster system for studying electronic structure of TMO films

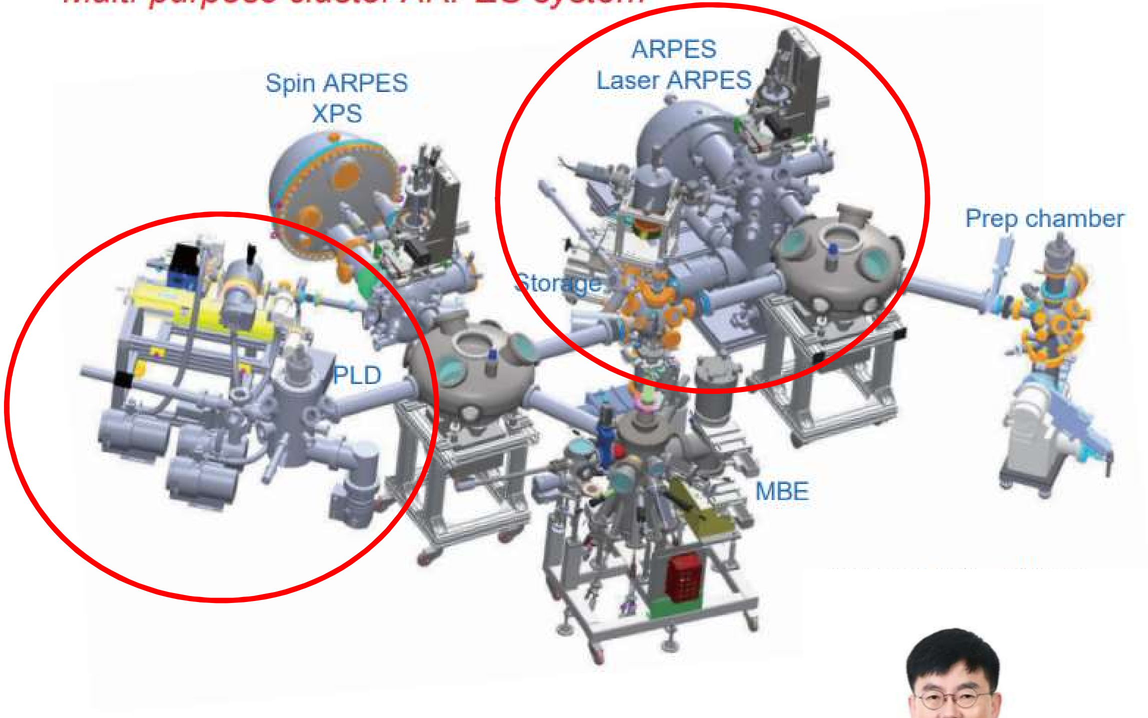
Pulsed Laser Deposition



in-situ Angle-resolved
Photoemission Spectroscopy

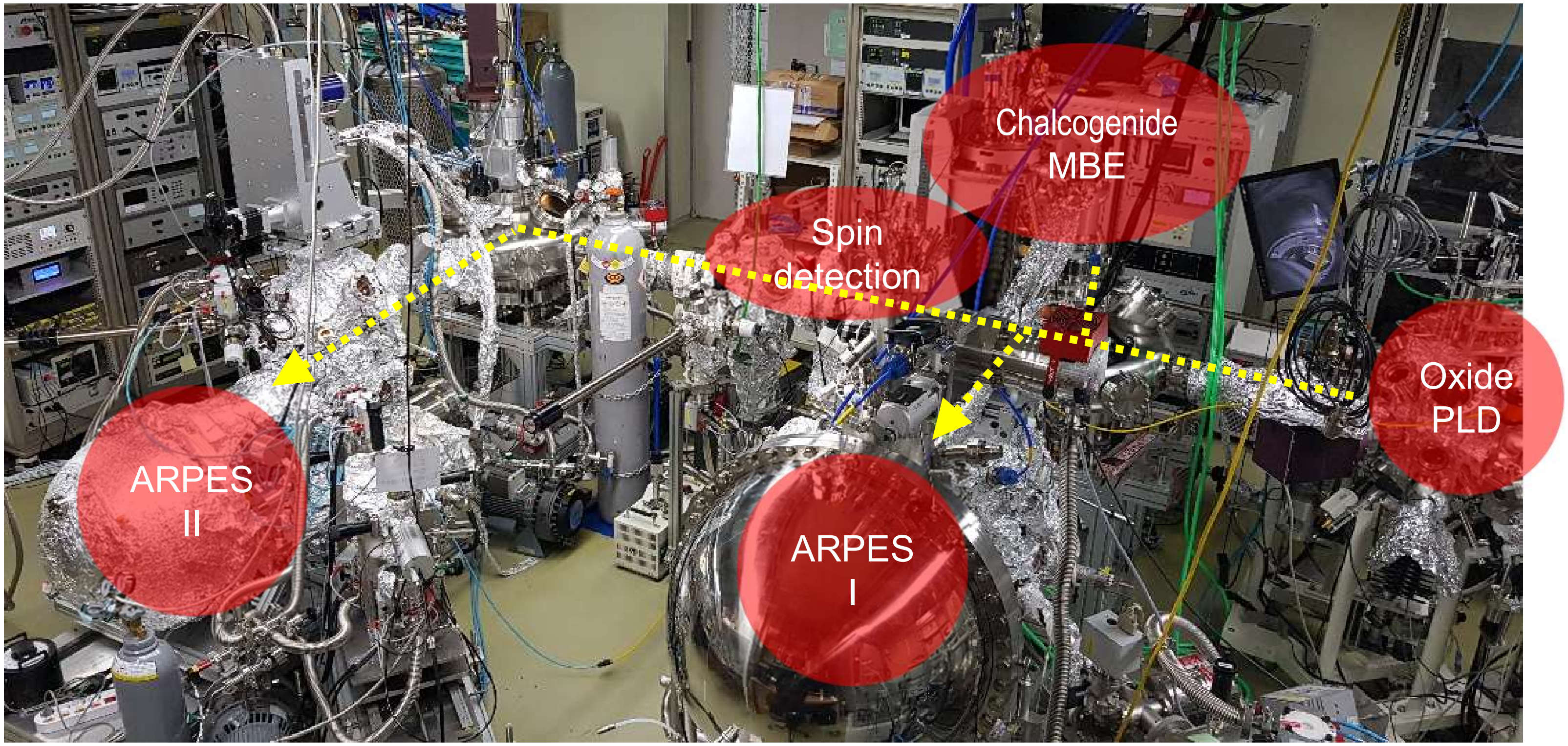


Multi-purpose cluster ARPES system



Collaboration with
Prof. Changyoung Kim

Cluster system for *in-situ* ARPES



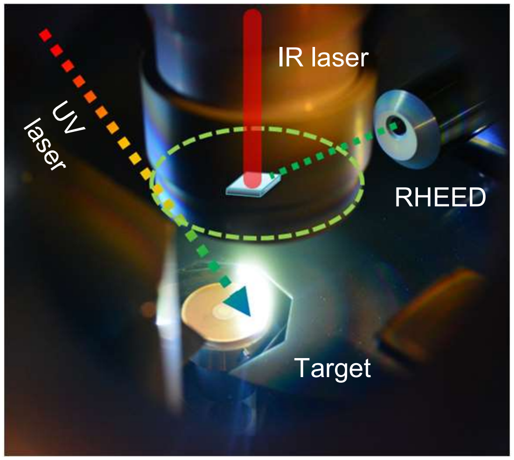
In-situ AREPS, ***In-situ*** *s*pin-resolved ARPES : measure average of local area responses

→ can characterize the electronic structures of the ultrathin films

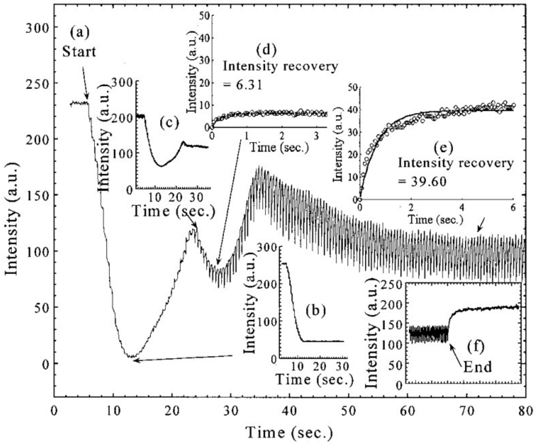
Energy Resolution: 6 meV

Growth of high-quality $SrRuO_3$ film growth

PLD growth

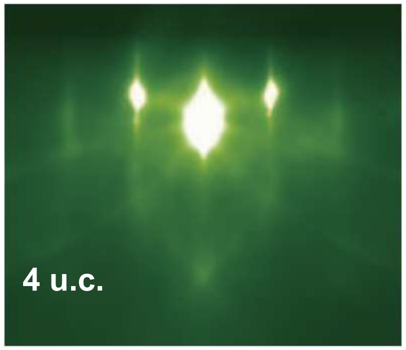


Step flow mode

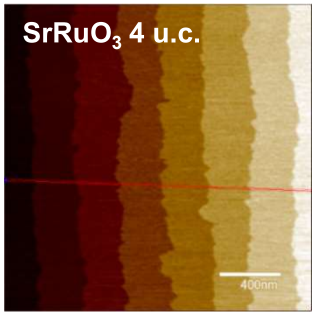


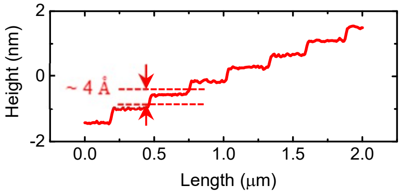
J. Choi et al., *Appl. Phys. Lett.* (2001)

RHEED



AFM



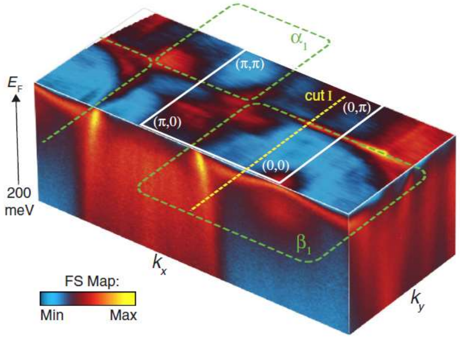


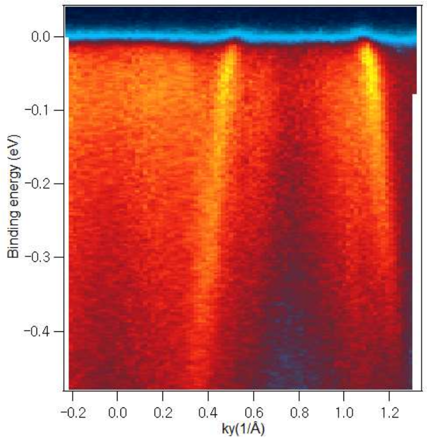
Via the step-flow growth mode of SRO films, we can grow very high-quality SRO films with atomic smoothness.

L. Wang et al., *Nano Lett.* (2020)

In-situ ARPES results on SRO thin-films

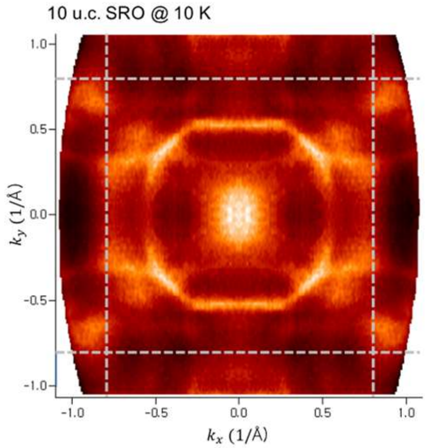
MBE @Cornell

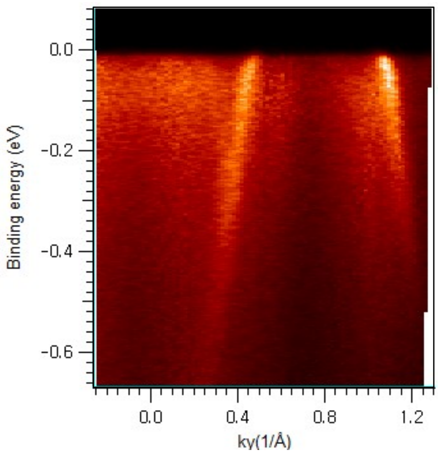




D. E. Shai et al., PRL **110**, 087004 (2013).

PLD @SNU





B. Sohn, C. Kim *et. al.,* Nature Materials (2021)

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ibS CCES
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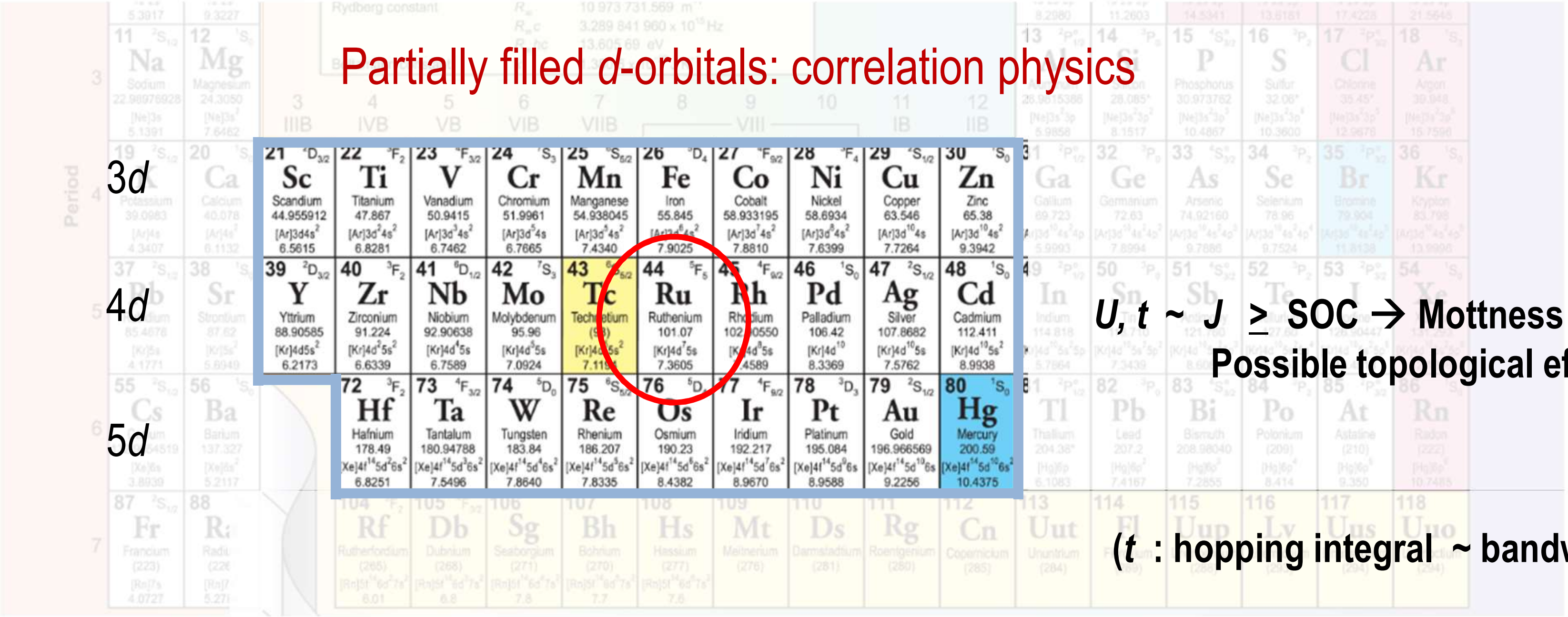
Material platforms of searching for CTPs : Ruthenates

Electronic correlations : On-site Coulomb interaction (*U*)

& Hund's rule coupling (*J*)

Topology : Spin-orbit coupling interaction (SOC)



Partially filled *d*-orbitals: correlation physics

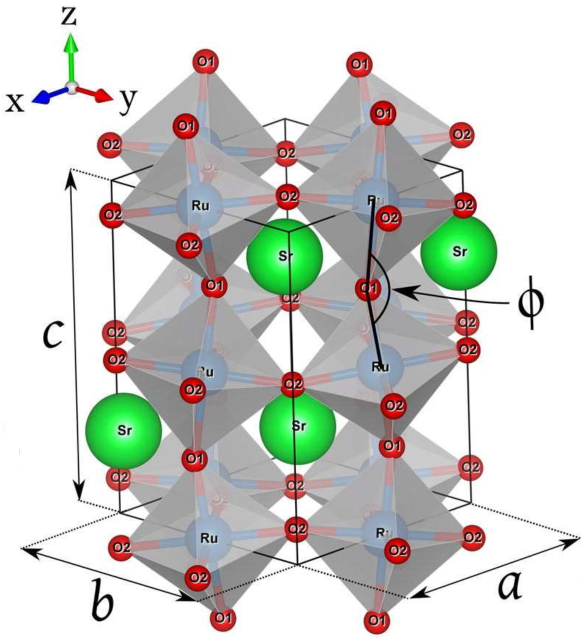
	3 IIIB	4 IVB	5 VB	6 VIB	7 VIIB	8	9	10	11 IB	12 IIB
3*d*	21 $^2D_{3/2}$ **Sc** Scandium 44.955912 $[Ar]3d4s^2$ 6.5615	22 $^3F_2$ **Ti** Titanium 47.867 $[Ar]3d^24s^2$ 6.8281	23 $^4F_{3/2}$ **V** Vanadium 50.9415 $[Ar]3d^34s^2$ 6.7462	24 $^7S_3$ **Cr** Chromium 51.9961 $[Ar]3d^54s$ 6.7665	25 $^6S_{5/2}$ **Mn** Manganese 54.938045 $[Ar]3d^54s^2$ 7.4340	26 $^5D_4$ **Fe** Iron 55.845 $[Ar]3d^64s^2$ 7.9025	27 $^4F_{9/2}$ **Co** Cobalt 58.933195 $[Ar]3d^74s^2$ 7.8810	28 $^3F_4$ **Ni** Nickel 58.6934 $[Ar]3d^84s^2$ 7.6399	29 $^2S_{1/2}$ **Cu** Copper 63.546 $[Ar]3d^{10}4s$ 7.7264	30 $^1S_0$ **Zn** Zinc 65.38 $[Ar]3d^{10}4s^2$ 9.3942
4*d*	39 $^2D_{3/2}$ **Y** Yttrium 88.90585 $[Kr]4d5s^2$ 6.2173	40 $^3F_2$ **Zr** Zirconium 91.224 $[Kr]4d^25s^2$ 6.6339	41 $^6D_{1/2}$ **Nb** Niobium 92.90638 $[Kr]4d^45s$ 6.7589	42 $^7S_3$ **Mo** Molybdenum 95.96 $[Kr]4d^55s$ 7.0924	43 $^6S_{5/2}$ **Tc** Technetium (98) $[Kr]4d^55s^2$ 7.119	44 $^5F_5$ **Ru** Ruthenium 101.07 $[Kr]4d^75s$ 7.3605	45 $^4F_{9/2}$ **Rh** Rhodium 102.90550 $[Kr]4d^85s$ 7.4589	46 $^1S_0$ **Pd** Palladium 106.42 $[Kr]4d^{10}$ 8.3369	47 $^2S_{1/2}$ **Ag** Silver 107.8682 $[Kr]4d^{10}5s$ 7.5762	48 $^1S_0$ **Cd** Cadmium 112.411 $[Kr]4d^{10}5s^2$ 8.9938
5*d*		72 $^3F_2$ **Hf** Hafnium 178.49 $[Xe]4f^{14}5d^26s^2$ 6.8251	73 $^4F_{3/2}$ **Ta** Tantalum 180.94788 $[Xe]4f^{14}5d^36s^2$ 7.5496	74 $^5D_0$ **W** Tungsten 183.84 $[Xe]4f^{14}5d^46s^2$ 7.8640	75 $^6S_{5/2}$ **Re** Rhenium 186.207 $[Xe]4f^{14}5d^56s^2$ 7.8335	76 $^5D_4$ **Os** Osmium 190.23 $[Xe]4f^{14}5d^66s^2$ 8.4382	77 $^4F_{9/2}$ **Ir** Iridium 192.217 $[Xe]4f^{14}5d^76s^2$ 8.9670	78 $^3D_3$ **Pt** Platinum 195.084 $[Xe]4f^{14}5d^96s$ 8.9588	79 $^2S_{1/2}$ **Au** Gold 196.966569 $[Xe]4f^{14}5d^{10}6s$ 9.2256	80 $^1S_0$ **Hg** Mercury 200.59 $[Xe]4f^{14}5d^{10}6s^2$ 10.4375

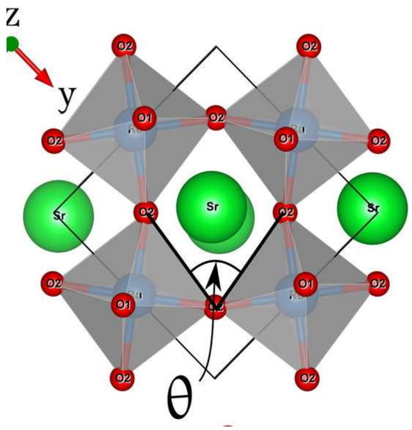
***U*, *t* ~ *J* ≥ SOC → Mottness & Hundness**

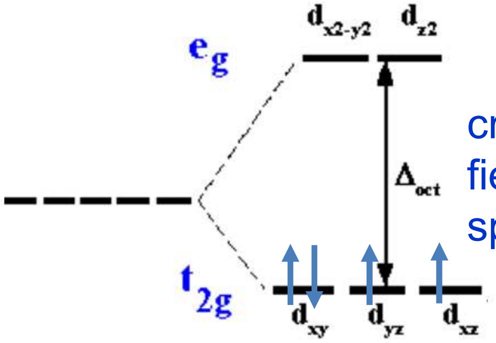
Possible topological effects

(*t* : hopping integral ~ bandwidth)

$SrRuO_3$: structural properties







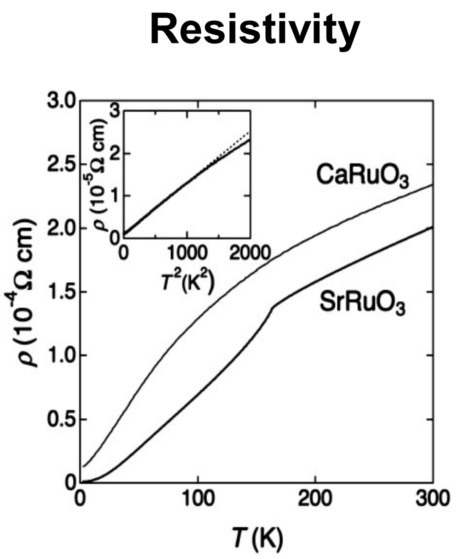
crystal
field
splitting

- Orthorhombic (*Pnma*) structure with oxygen octahedral rotation (OOR):

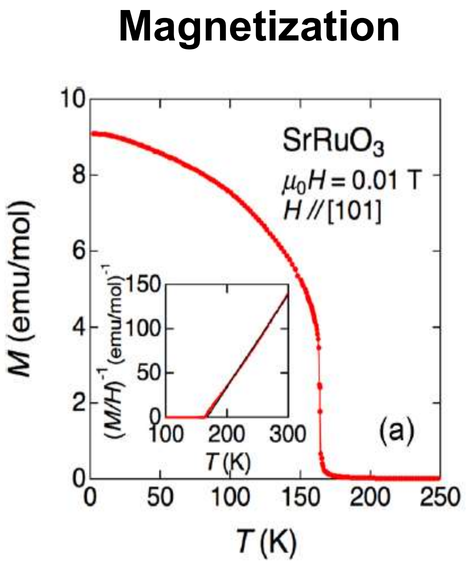
  tilting (ϕ) and rotation (θ) :
- In the Glazer notation, **$a^-a^-c^+$** rotation of RuO_6 octahedra (at RT)
- Ru^{4+} ion has 4 electrons in the t_{2g} bands (*xy, yz,* & *zx* orbitals).

J. Okamoto, M. Takano *et al.*, *Phys. Rev. B* (1999)

Basic Properties of $SrRuO_3$: Metallic Ferromagnet


Resistivity

Fermi liquid metal at low T


Magnetization

Ferromagnetism (T_c~160K)

☐ Highly metallic → widely used as electrode materials in oxide electronics

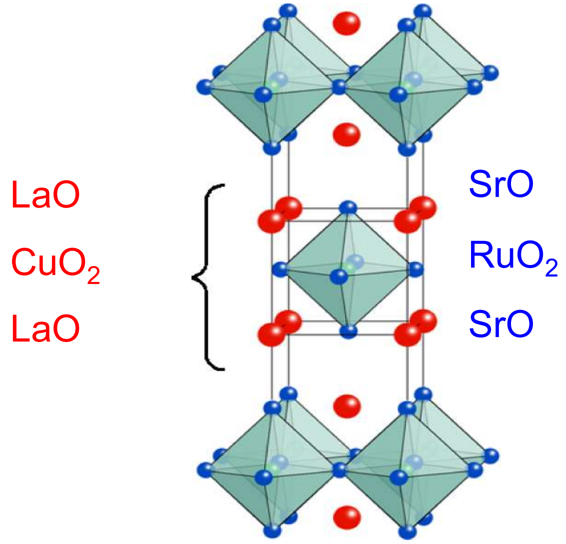
☐ Metallic & FM → Stoner mechanism?

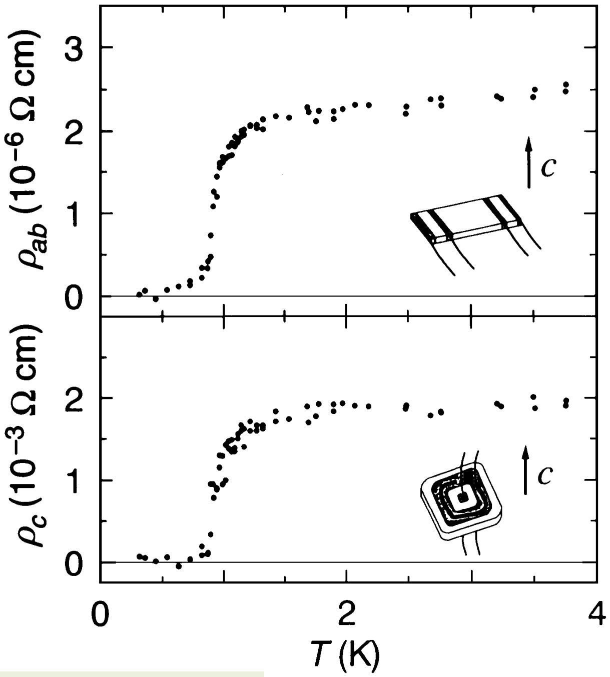
☐ Correlations due to *J* ~ 0.5 eV : orbital degeneracy effects → Hund's metal ? ….

☐ Anomalous Hall effects (AHE), due to the Berry curvature

☐ $SrRuO_3$ single crystals : hardly available → high quality thin films useful !!

Superconductivity in Sr_2RuO_4

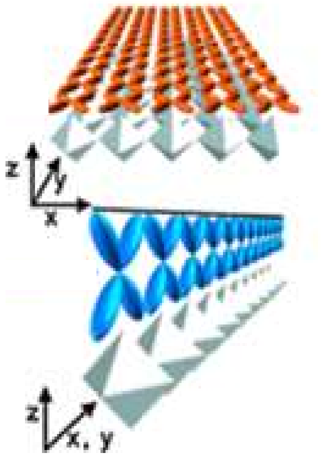


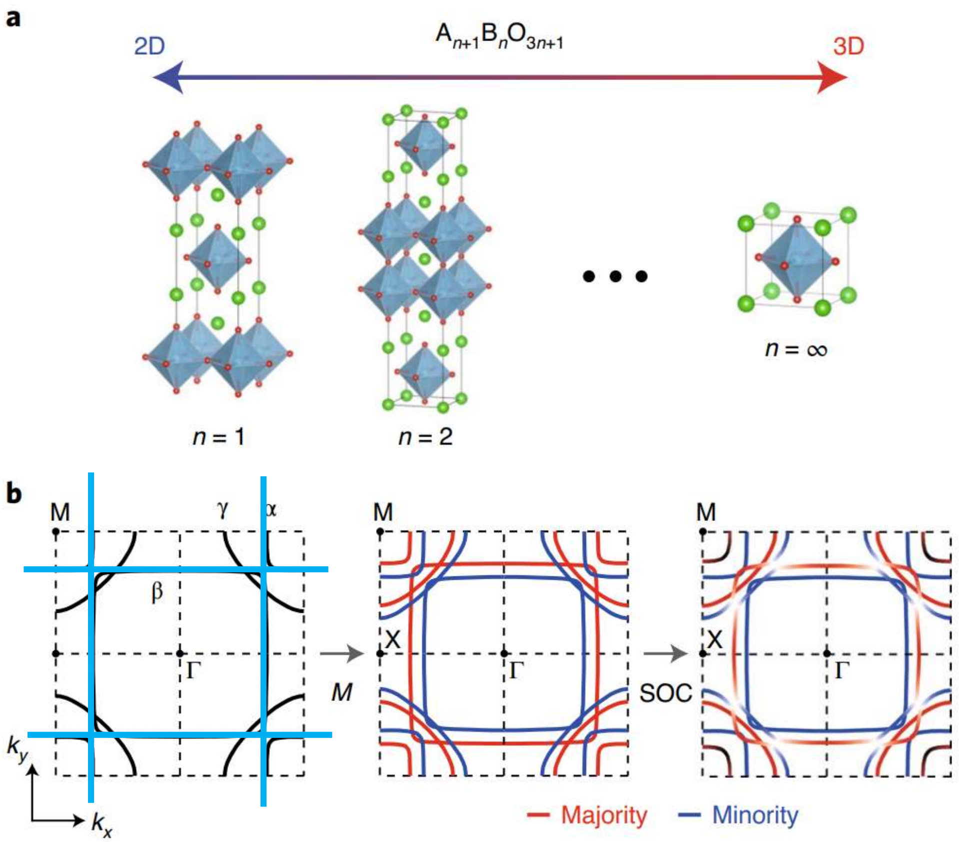


- 2D layer-perovskite structure
- Superconductivity at ~ 1.5 K
- A candidate of *p*-wave superconductor ?
- Similarily to 1 *uc* $SrRuO_3$ film (i.e. SRO in 2D limit) ?

Y. Maeno *et al*., Nature **372**, 532 (1994)

Electronic Structure of a $SrRuO_3$ ultrathin film : Fermi surface



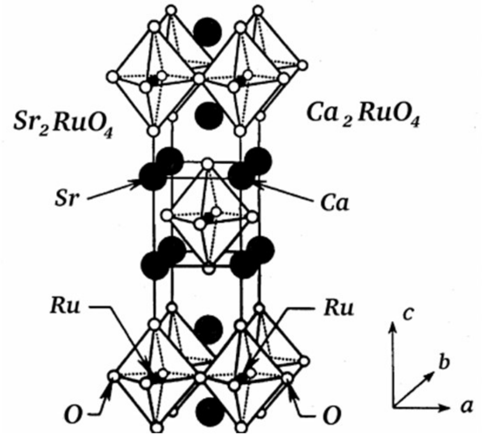


γ : d_{xy} orbital (2D-character)

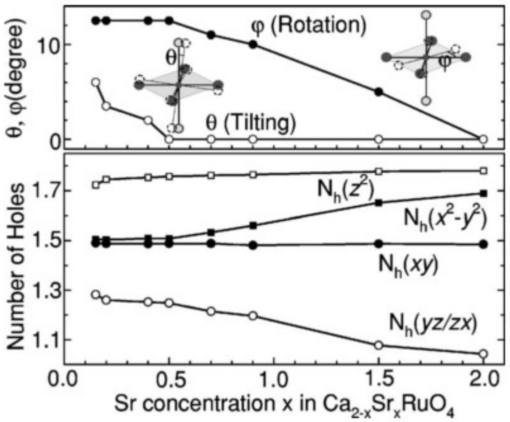
α , β : $d_{xz,yz}$ orbitals (1D-character)

B. Sohn, TWN, C. Kim *et. al.*, Nature Materials (2021)

Substitution effects in $Ca_{2-x}Sr_xRuO_4$

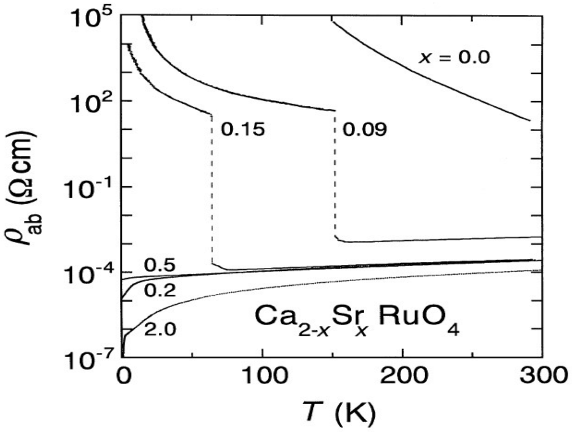


Octahedron rotation & tilt



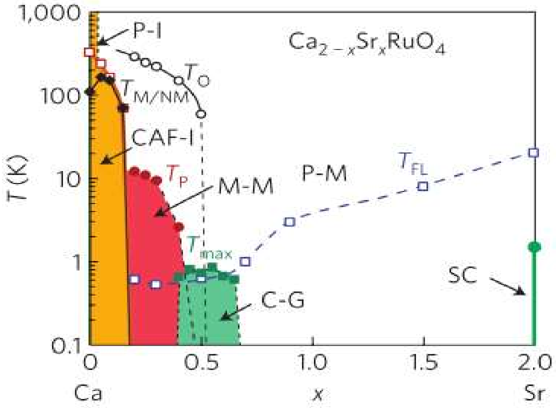
H.-J. Noh et al, PRB (2005)

Metal insulator transition



S. Nakatsuji, PRL 84, 2666(2000).

SCRO phase diagram



J. P. Carlo, Nature Materials (2012)

With Ca-substitution,

- RuO_6 tilting & rotation
- filling change of *d*-orbitals
- metal-insulator transition
- a very rich phase diagram

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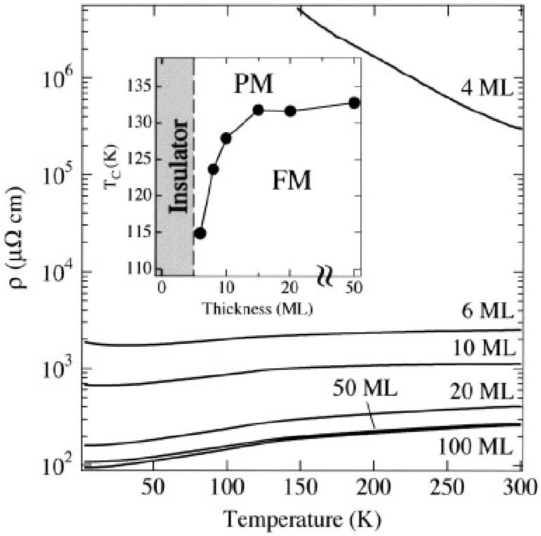
Thickness-dependent MIT of SRO films : previous studies

Question: At which thickness, does MIT occur in SRO films?



T_C **~ 4 u.c.**

Disorder in rough $SrRuO_3$ films

M. Izumi et al., JPSJ (1998)

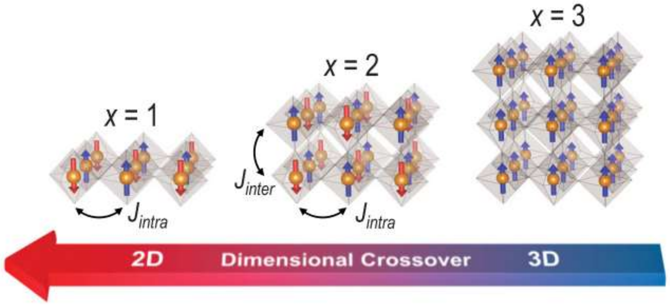
D. Toyota et al., APL (2005)

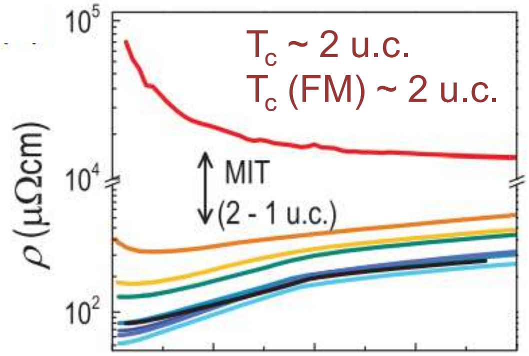
J. Xia et al., PRB (2009)

S. Kang et al., PRB (2019)

H. Jeong APL (2019)

Superlattices ($SrRuO_3$-$SrTiO_3$)

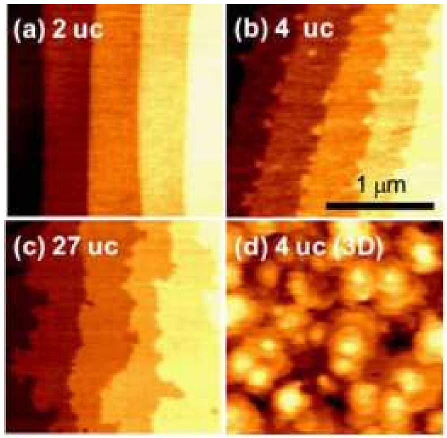


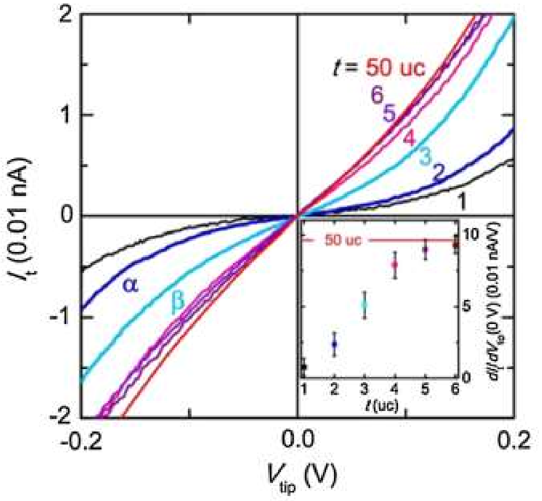


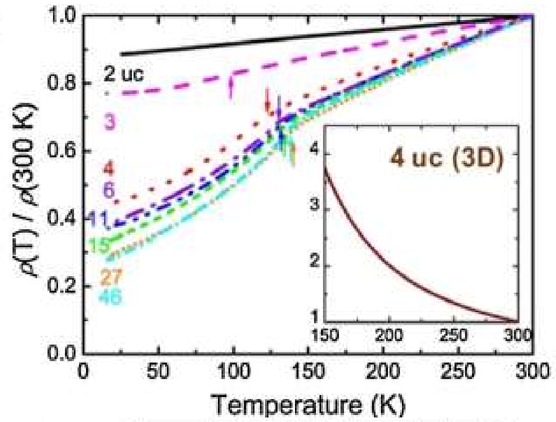
S. G. Jeong, and W. S. Choi et al., *PRL* (2020)

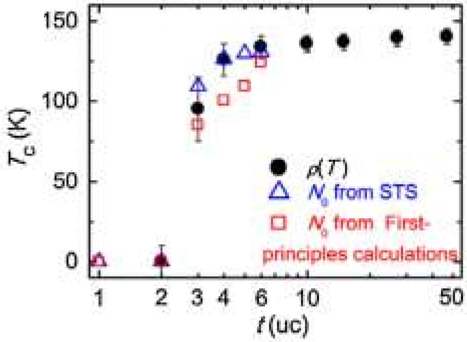
Fundamental thickness of $SrRuO_3$ ultrathin films

- Our previous study of SRO films









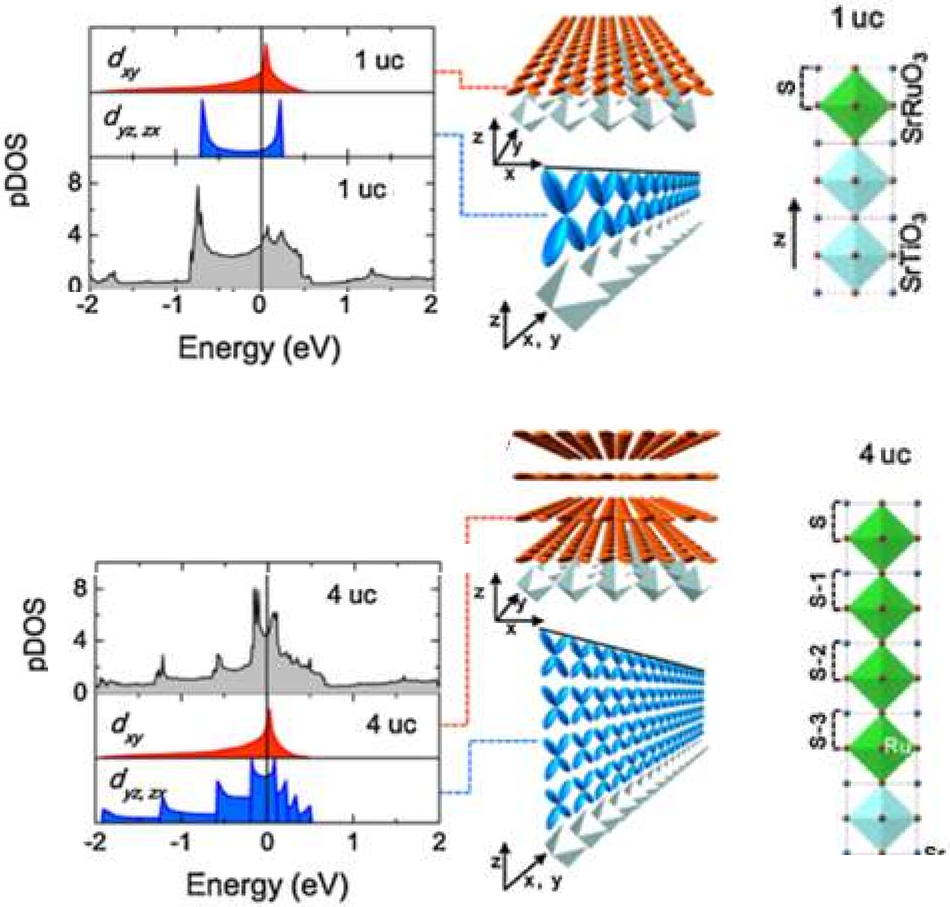
In 2009, we observed film thickness limits for metallicity and ferromagnetism.

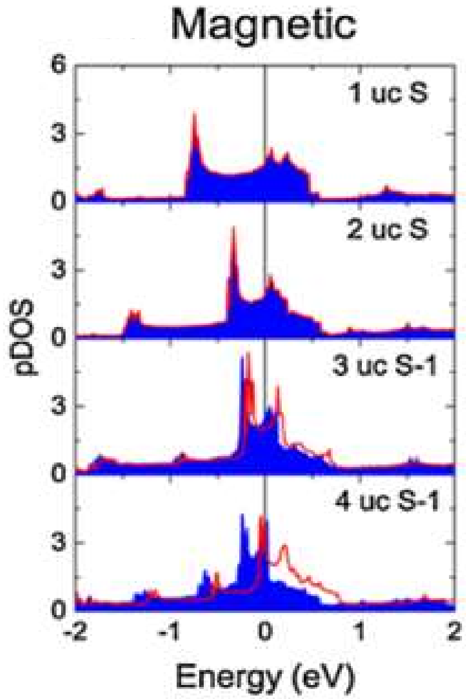
Metallicity : 1 u.c.
FM : 3 u.c.

Y. J. Chang, *et. al.*, *PRL* (2009)

Fundamental thickness of $SrRuO_3$ ultrathin films

- Orbital-selective quantum confinement effects (DFT)





DFT calculation-Critical thickness
for metallicity : 1 u.c.
for FM : 3 u.c.

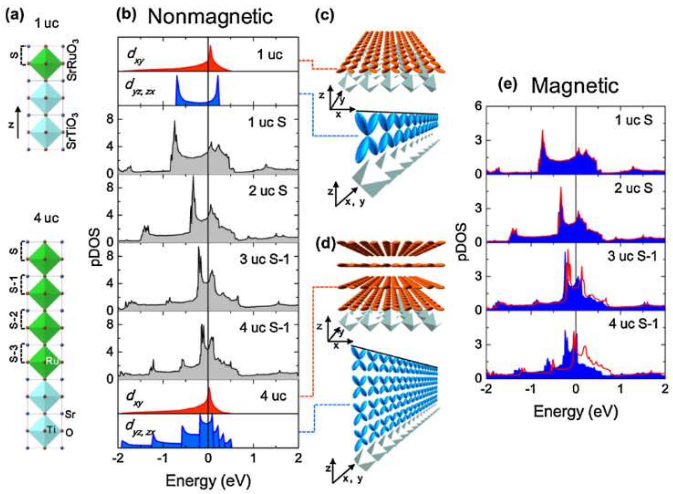
Y. J. Chang, *et. al., Phys. Rev. Lett.* (2009)

Fundamental thickness of SrRuO$_3$ ultrathin films

- Controversy on metallicity

Monolayer SRO is a metal

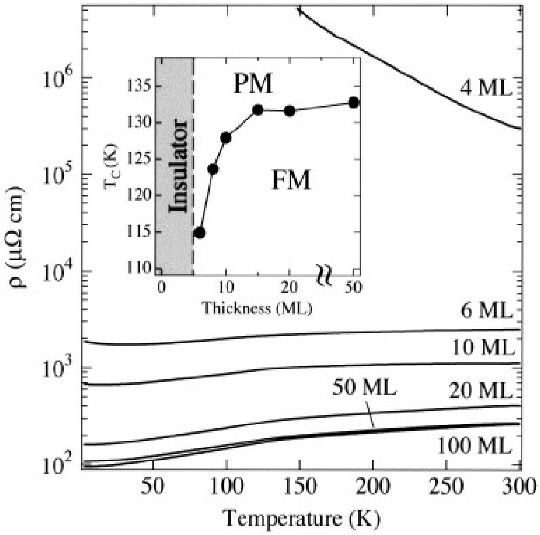
Para- or ferro-magnetic **metallic** ground state



Y. J. Chang, et al., Phys. Rev. Lett. 103, 057201 (2009)

VS

Monolayer SRO is an insulator

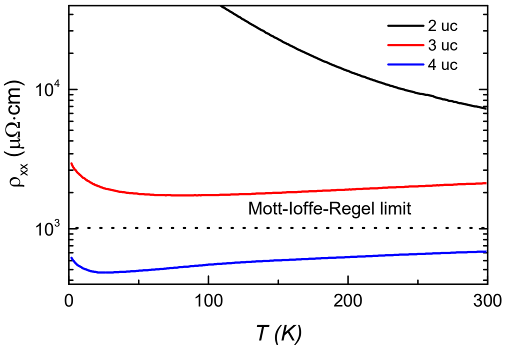


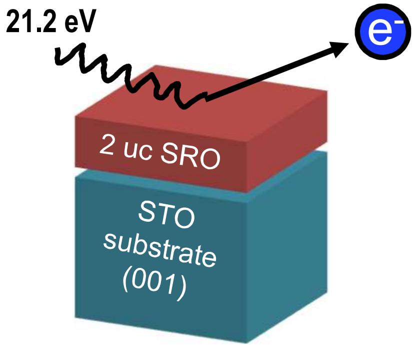
D. Toyota *et al.*, *APL* (2005)
M. Izumi *et al.*, *JPSJ* (1998)
...

Transport and magnetic measurements of ultra-thin films are quite difficult...
→ in-situ ARPES can be useful

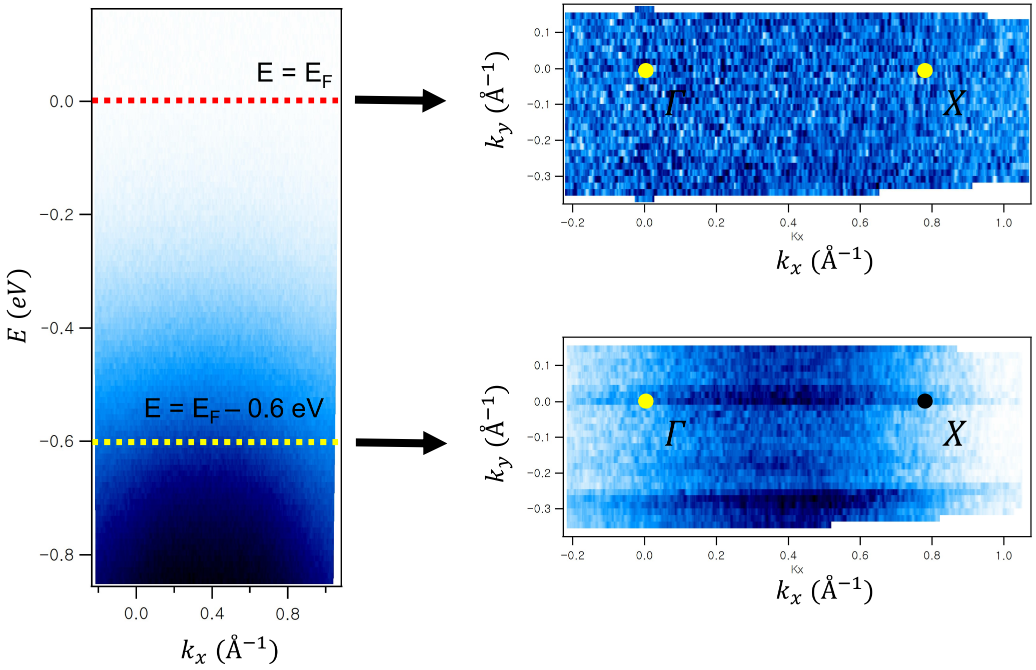
Our first ARPES on 2 u. c. SRO films

Resistivity of SRO/STO (001) films





In-situ ARPES on 2 uc SRO/STO (001) thin film

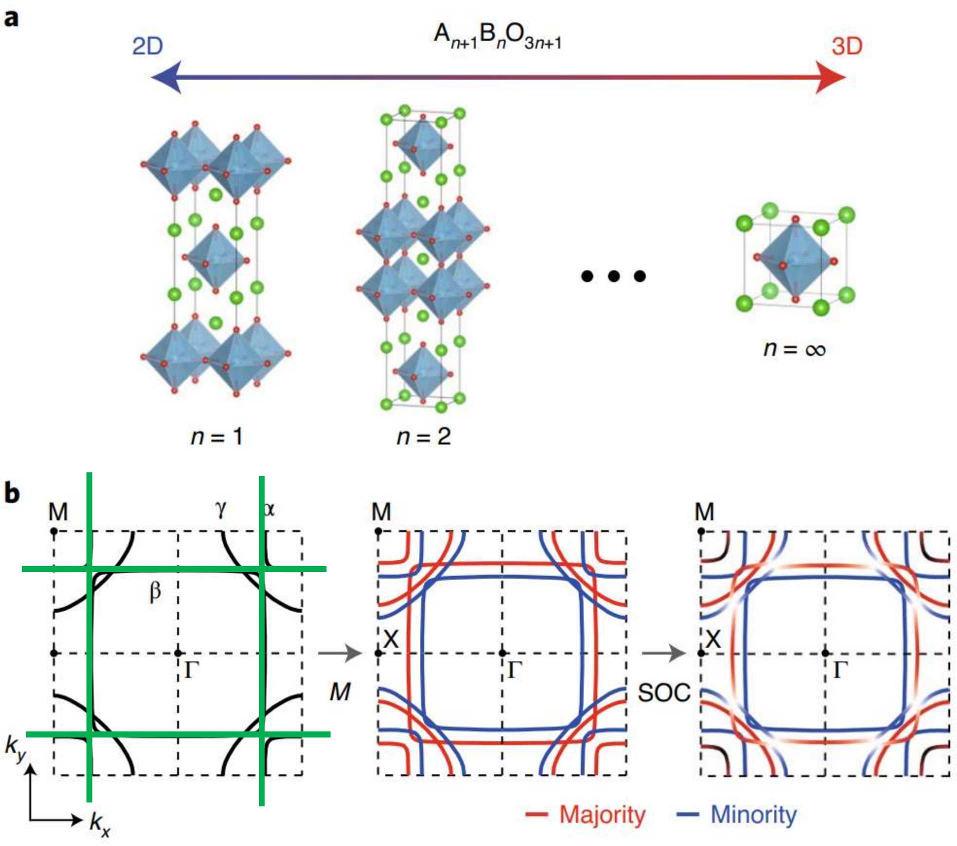


2 uc SRO film insulating in transport measurements

No band dispersion observed for 2 u.c. films

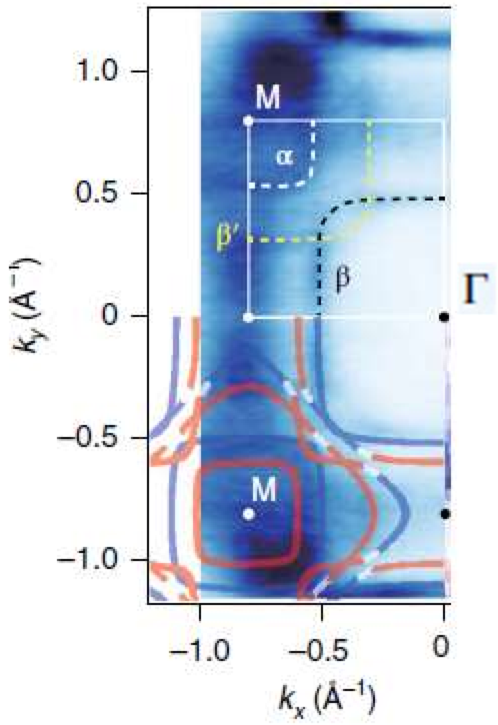
Are atomically-thin SRO films
intrinsic insulators? Or Charging effects ?

Electronic Structure of a $SrRuO_3$ film : Fermi surface



4uc SRO film

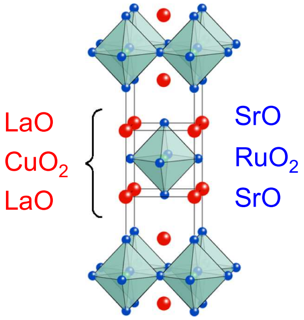
Synchrotron ARPES (80 eV)



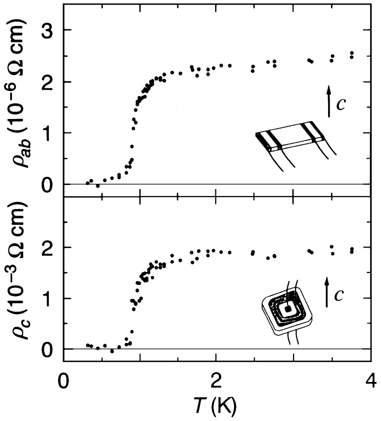
α , β : $d_{xz,yz}$ orbitals (1D-character)
Γ : d_{xy} orbital (2D-character)

B. Sohn, TWN, C. Kim *et. al.*, Nature Materials (2021)

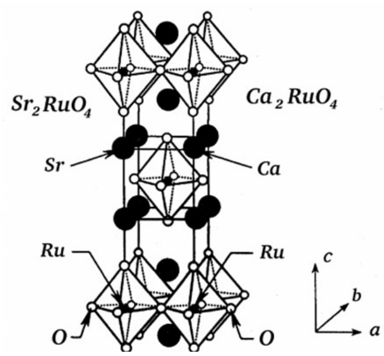
Octahedron distortions in bulk $Ca_{2-x}Sr_xRuO_4$



Sr_2RuO_4

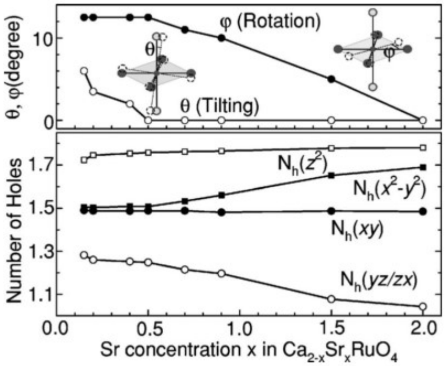


Y. Maeno *et al*., Nature **372**, 532 (1994)



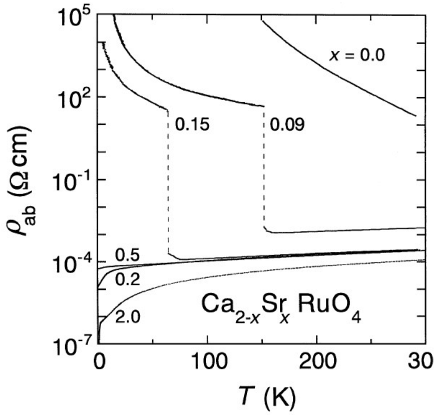
$Ca_{2-x}Sr_xRuO_4$

Octahedron rotation & tilt



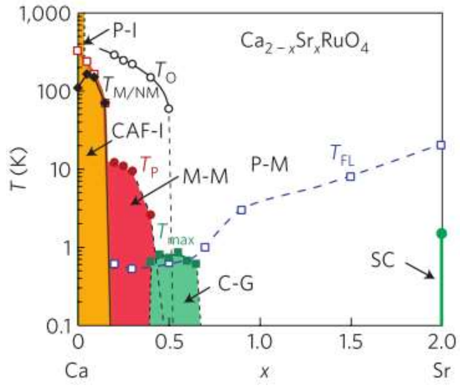
H.-J. Noh et al, PRB (2005)

Metal insulator transition



S. Nakatsuji, PRL 84, 2666(2000).

CSRO phase diagram



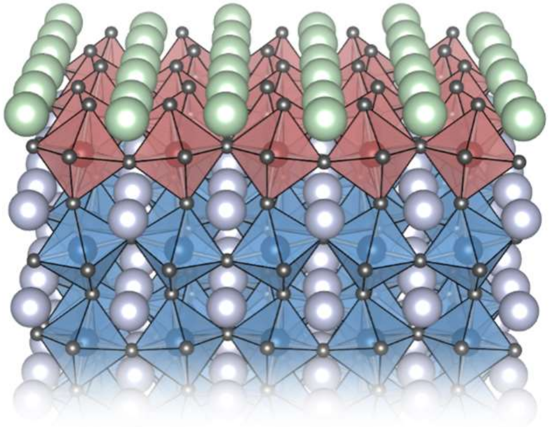
J. P. Carlo, Nature Materials (2012)

Challenging Task: Can we control octahedron distortion in 1 u. c. $SrRuO_3$ films?

Structure control of MIT in SRO ultrathin films

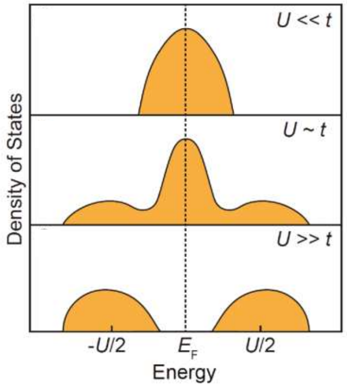
- Control of electronic states by octahedron distortion

Monolayer $SrRuO_3/CaTiO_3$
(SRO/CTO)



- **Electronic ground state?**
- $CaTiO_3$ buffer layer with oxygen octahedral rotation (OOR)
- CTO: $a^-b^+a^-$

Mott transition with increasing U/t



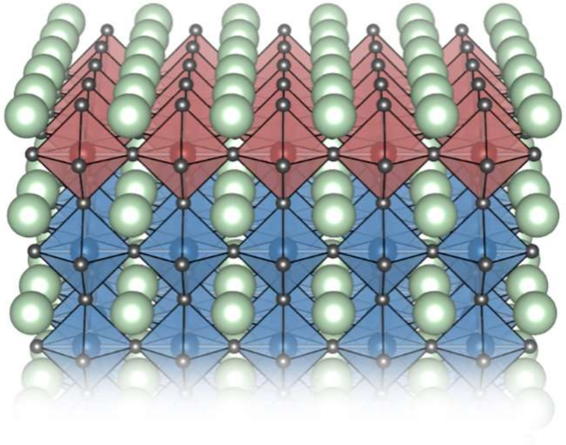
Increasing OR

←

Decreasing bandwidth, *t*

● Sr ● Ca ● Ru ● Ti · O

Monolayer $SrRuO_3/SrTiO_3$
(SRO/STO)

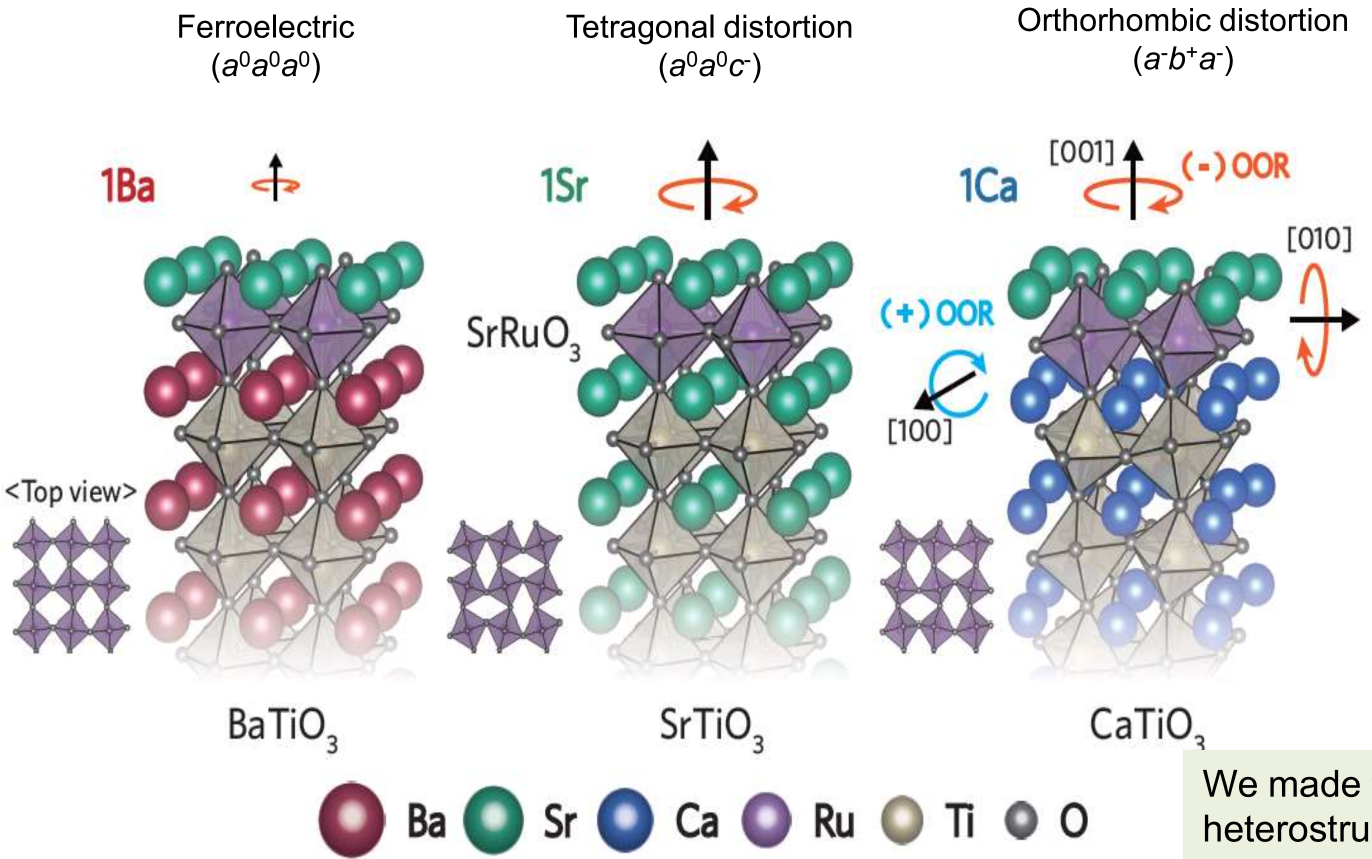


- **Metallic edge**
- Cubic $SrTiO_3$ buffer layer
- STO: $a^0b^0a^0$

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

Structure control of MIT in SRO ultrathin films

- Structural proximity effects using n-(SRO)$_n$/10-ATiO$_3$/4-SRO/STO



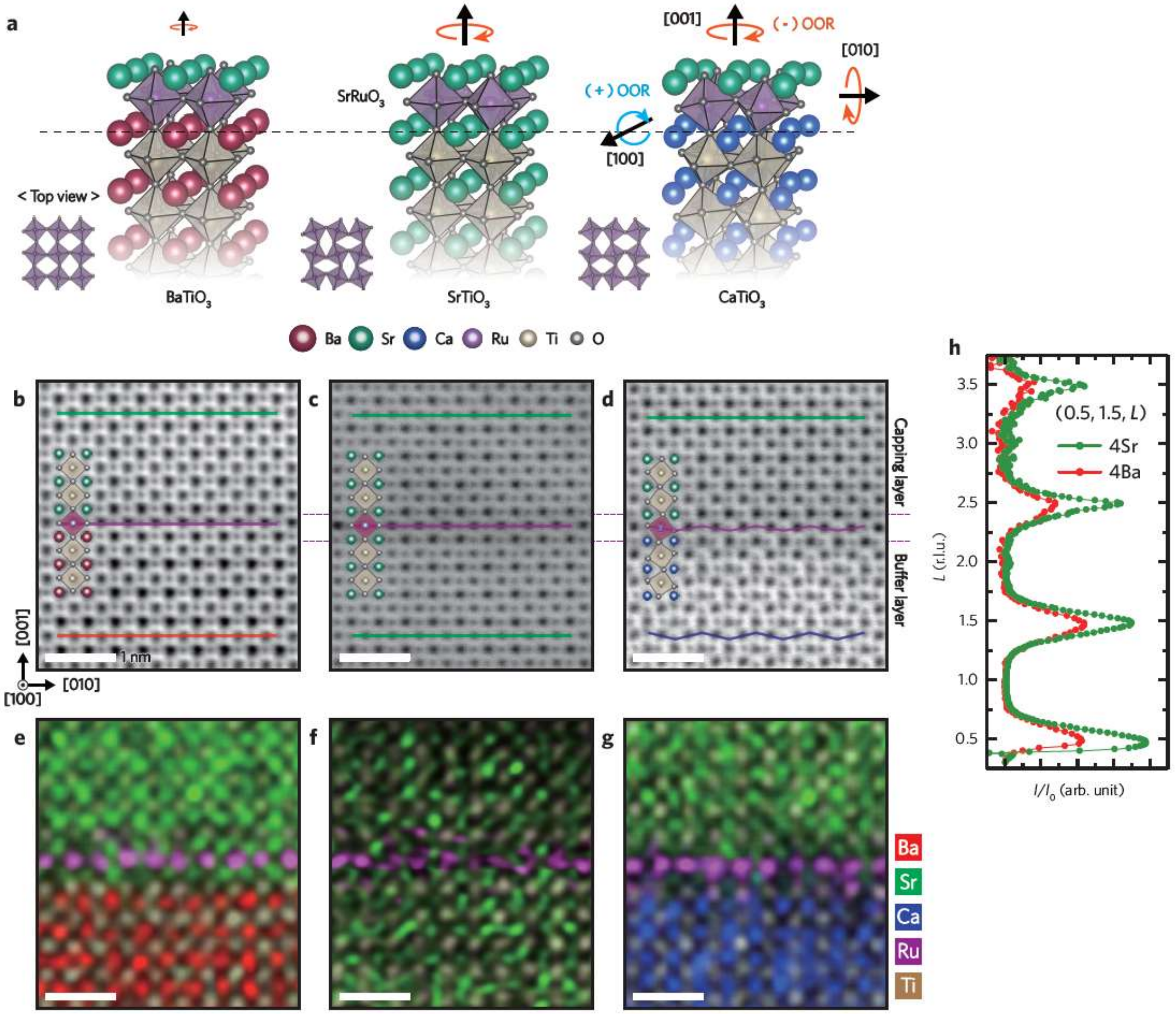
We made three different heterostructures by inserting layers of BTO, STO, and CTO, which were intended to create different distortions.

J. R. Kim, TWN, C. Kim *et. al., Advanced. Maters* (2023)

B. Sohn, J. R. Kim, TWN, C. Kim *et. al., Nat. Commun.* (2021)

Structure control of MIT in SRO ultrathin films

- STEM-EELS studies : chemical composition

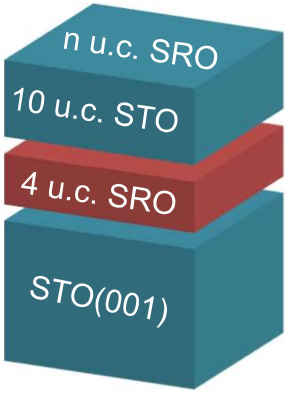


J. R. Kim, TWN, C. Kim *et. al., Advanced. Maters* (2023)

B. Sohn, J. R. Kim, TWN, C. Kim *et. al., Nat. Commun.* (2021)

Structure control of MIT in SRO ultrathin films

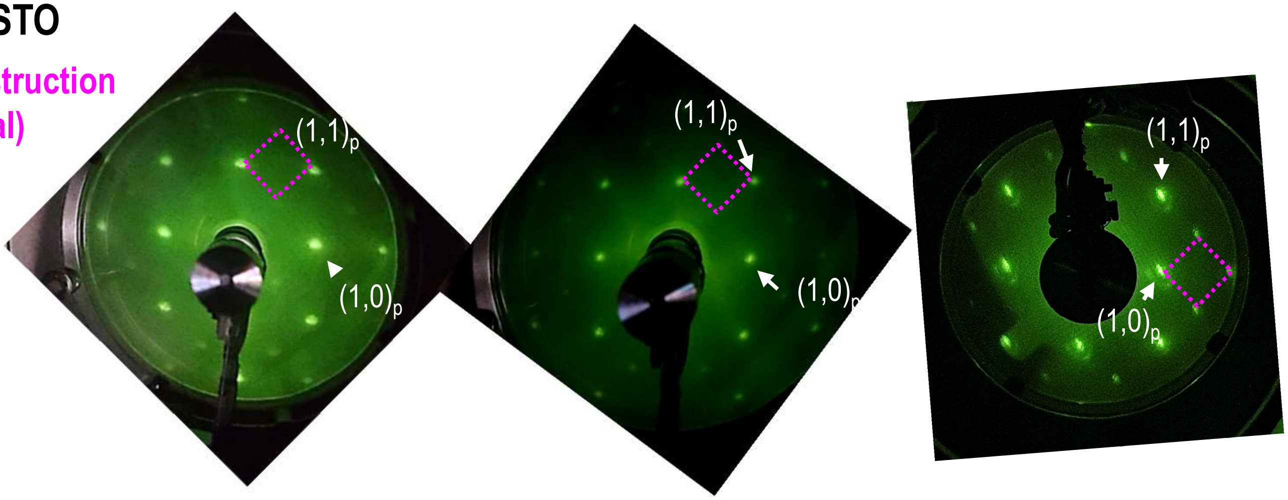
- LEED : surface reconstruction

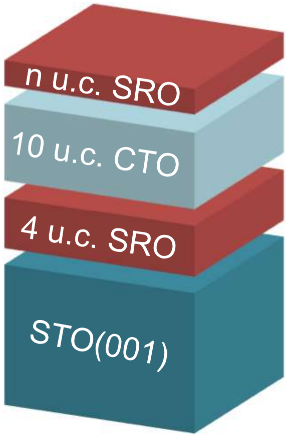


SRO/STO

$\sqrt{2} \times \sqrt{2}$ reconstruction (tetragonal)

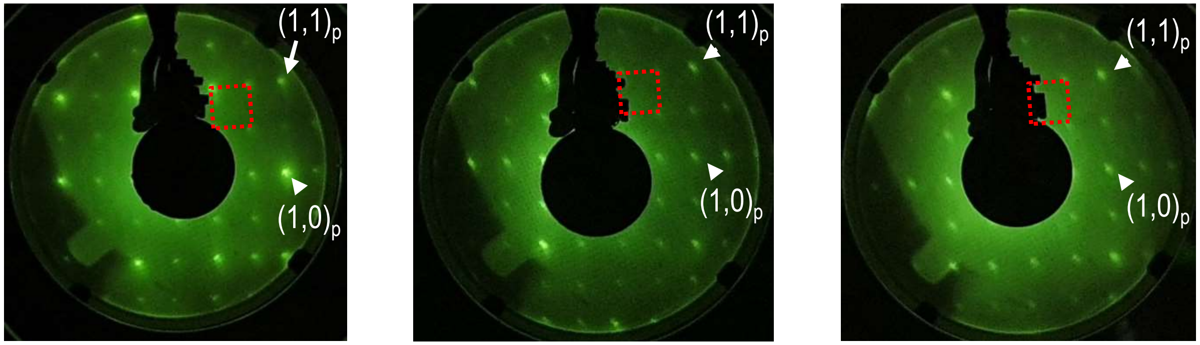
4 u.c. 2 u.c. 1 u.c.





SRO/CTO

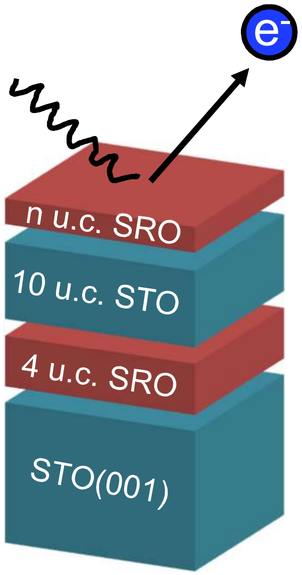
2 x 2 reconstruction (orthorhombic)



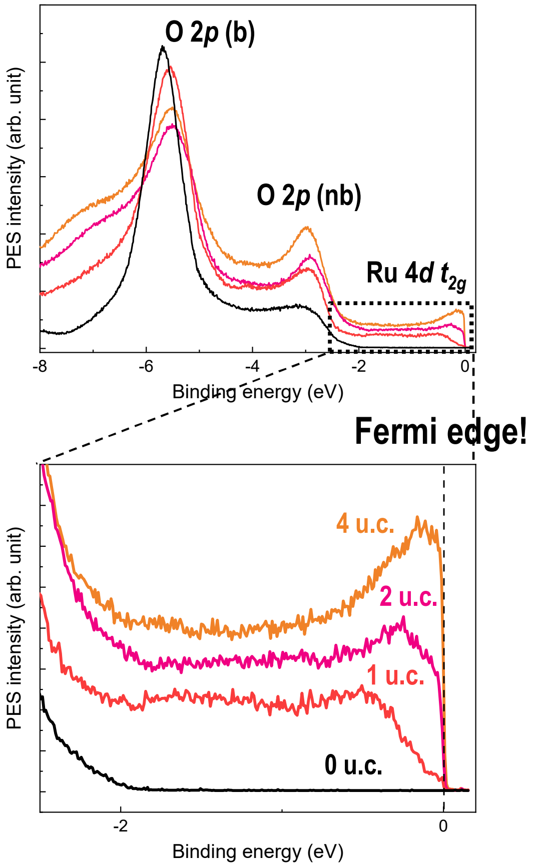
SRO crystal structure changes with buffer layers.

Structural control of MIT in SRO ultrathin films

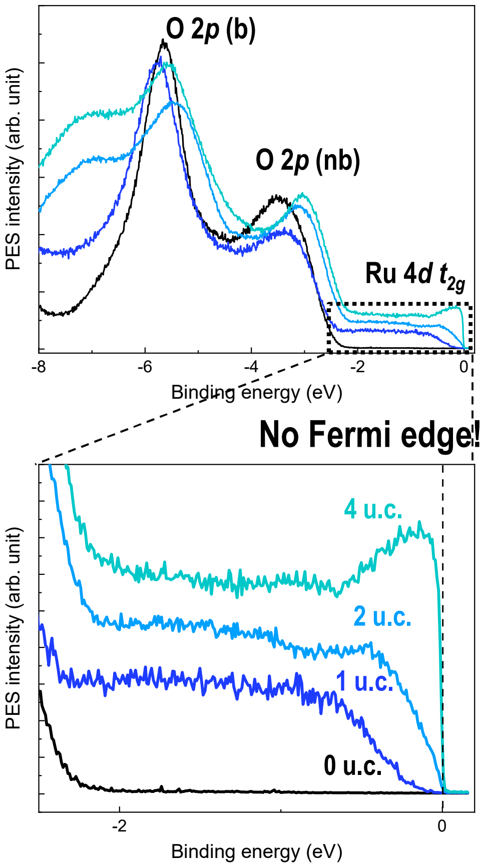
- Angle-averaged PES

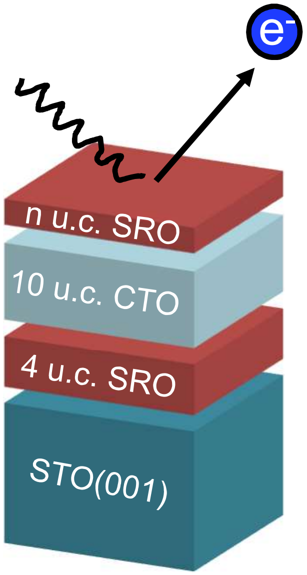


PES spectra of ultrathin SRO/STO



PES spectra of ultrathin SRO/CTO



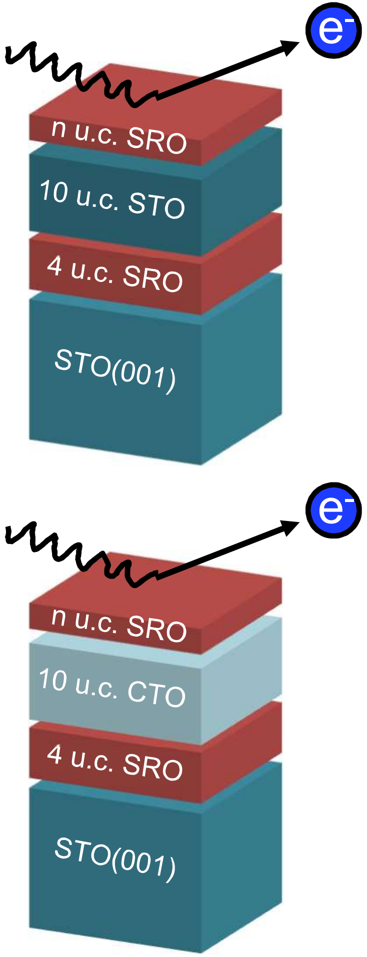


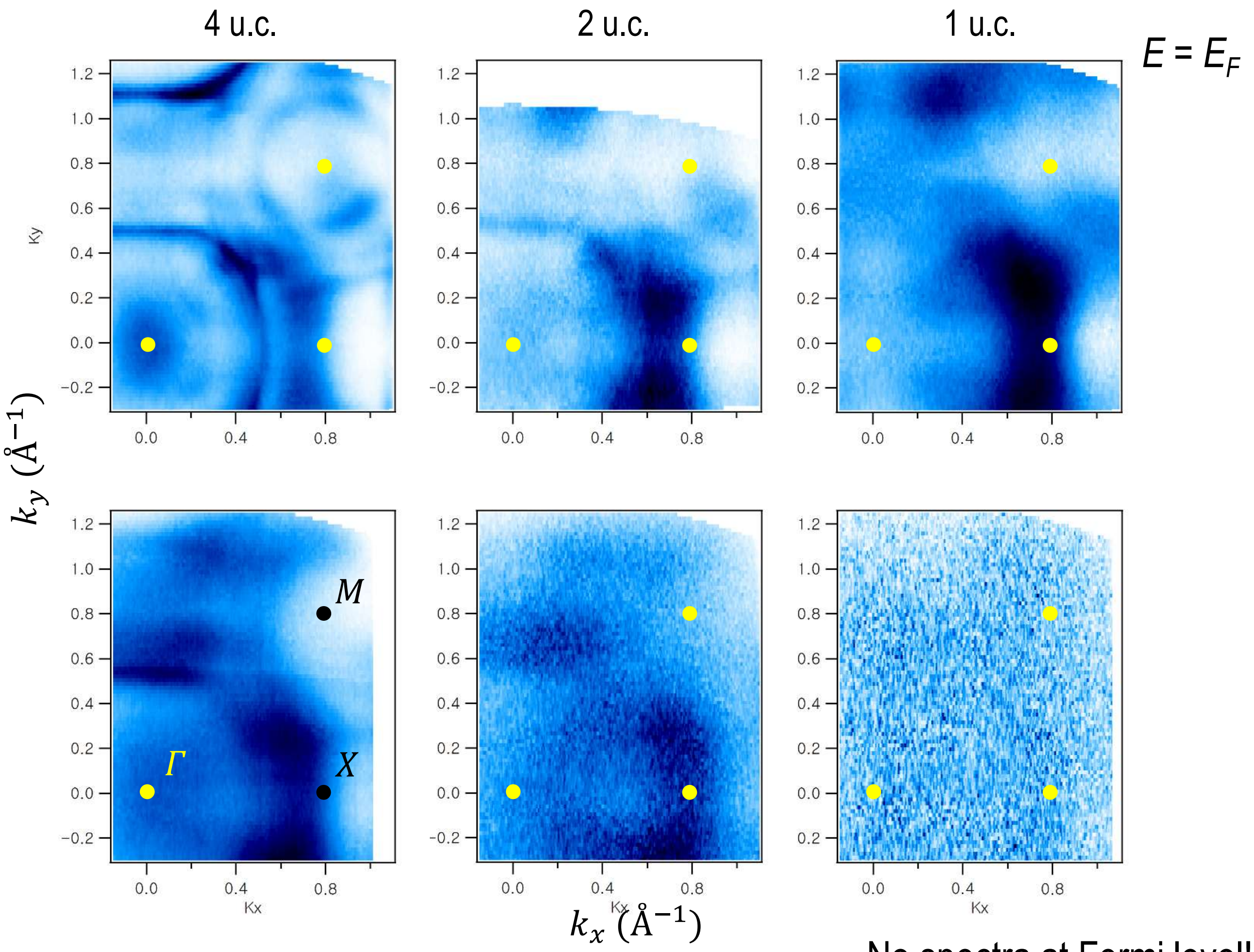
With STO, monolayer SRO film ($a^0a^0c^-$) is metallic

With CTO, monolayer SRO film (a^-b^+a) is insulating

Structural control of MIT in SRO ultrathin films

- ARPES spectra



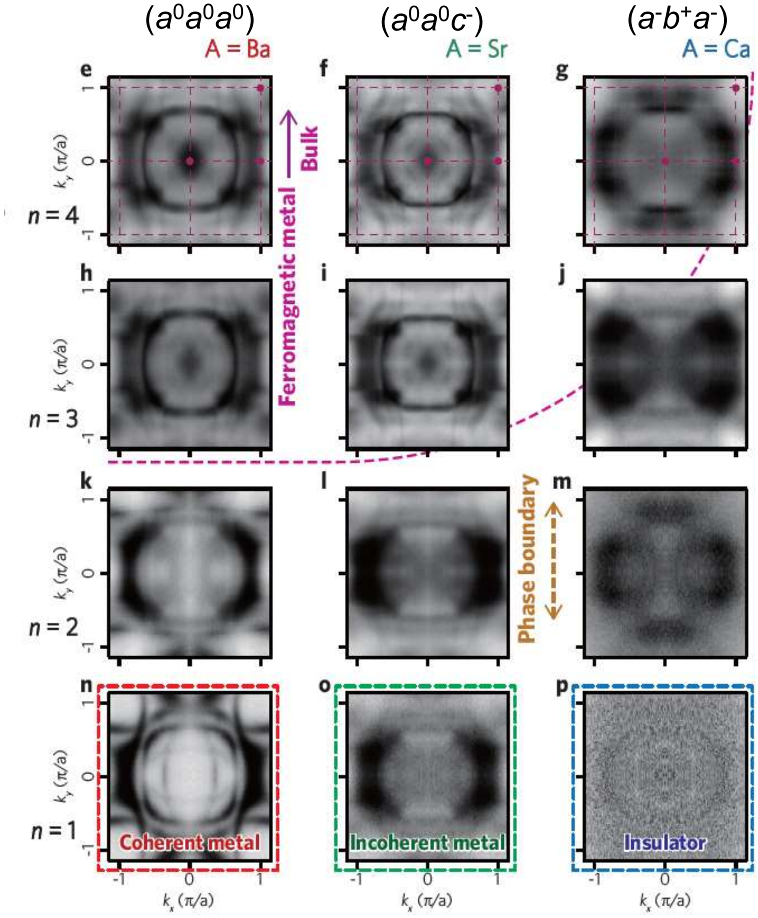


No spectra at Fermi level!

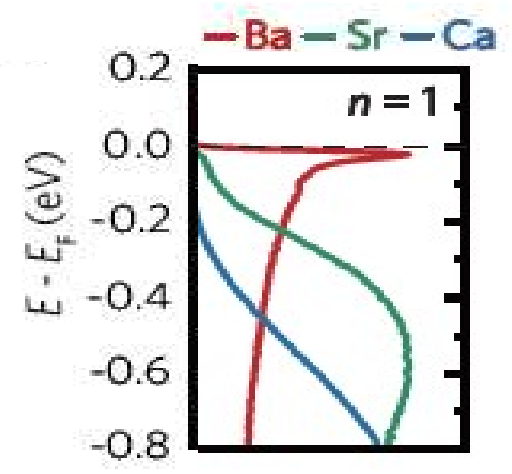
MIT can occur in 1 u.c. SRO film, depending on symmetry of SRO !

Structural control of electronic phases in SRO ultrathin films

- Emergence of 3 electronic phases



Coherent metal Incoherent metal Insulator



What is the nature of the inherent metallic state with small quasiparticle weight Z?

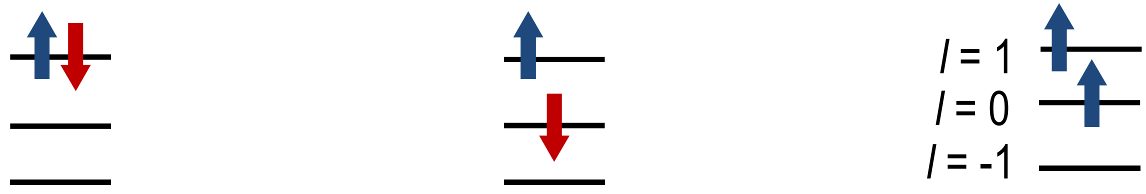
- Hund's metal?

J. R. Kim, TWN, C. Kim *et. al.*, *Advanced. Maters* (2023)

The Hund’s Rules for multiorbital systems

1. Total spin *S* should be maximized
2. Total angular momentum *L* should be maximized

For example, in 3-orbital (degenerate) t_{2g} system



Follows no rule
S = 0
L = 0

U

Follows rule 2
S = 0
L = 1

U-2*J*

Follows rule 1, 2
S = 1
L = 1

U-3*J*

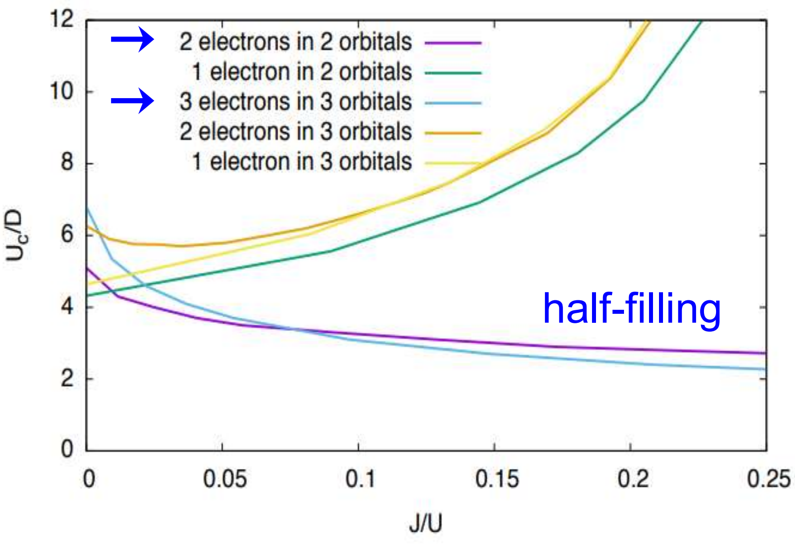
Interorbital exchange interaction *J* can plays important roles in multiorbital systems.

A. Georges et al., Annu. Rev. Condens. Matter Phys. (2013).

MIT in multiorbital systems

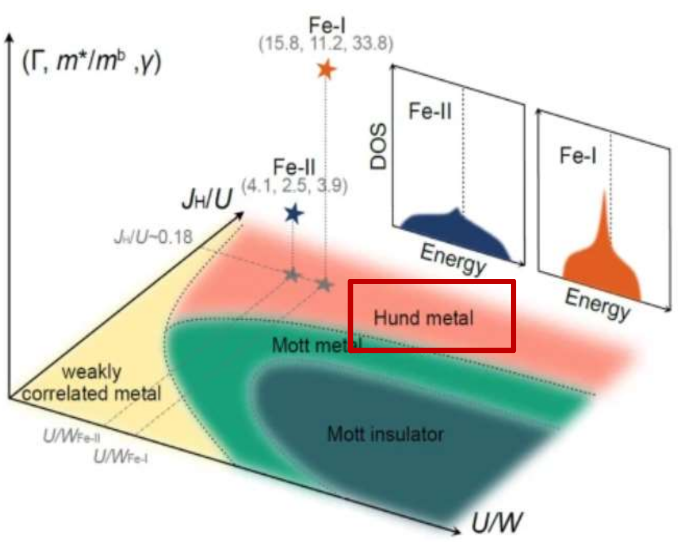
- Hund's coupling (*J*) plays the main role in determining electronic properties.

  The role of *J* varies depending on the number of orbitals and filling number.



half-filling

i. e. Fe_3GeTe_2

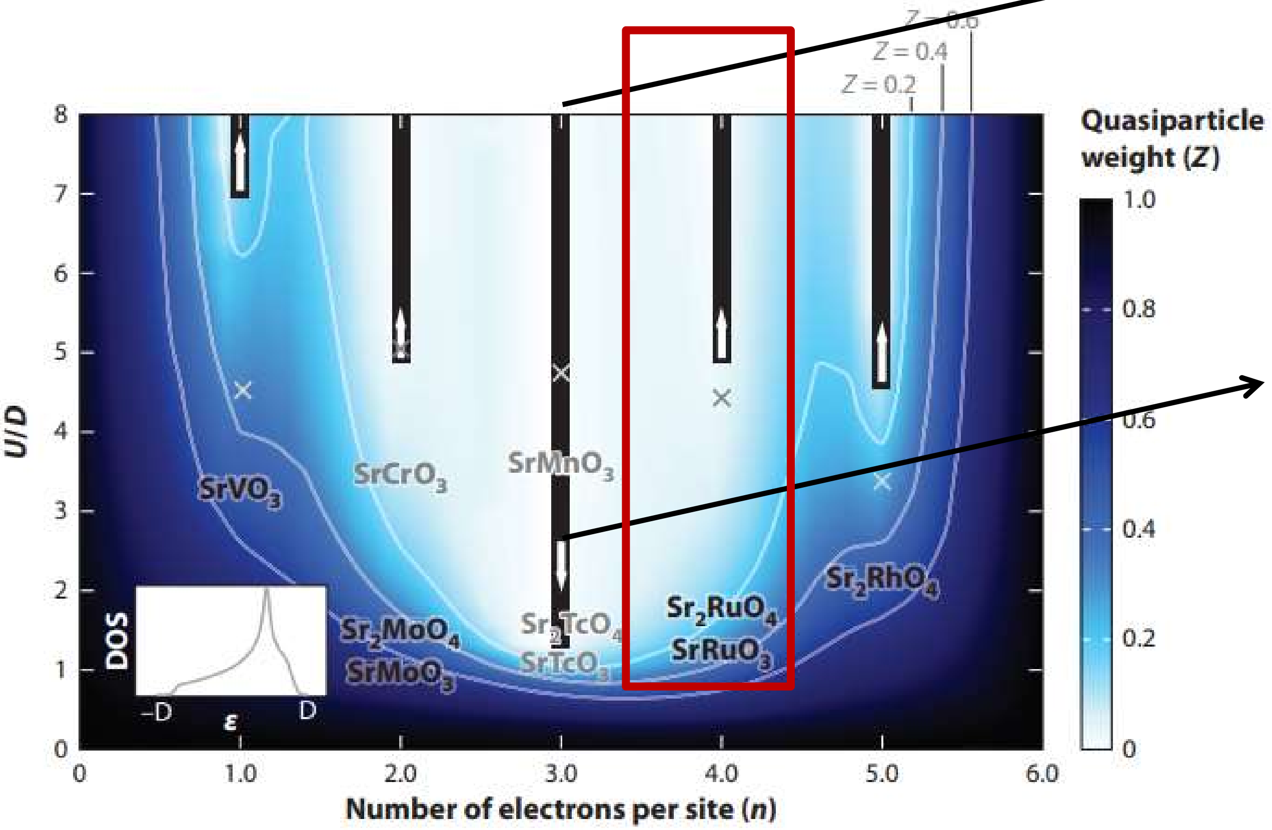


Hund's coupling polarizes the spin locally, which makes a small overlap between the noninteracting and spin-polarized atomic states.
Thus Z becomes suppressed, in concomitant with an enhancement of the spin fluctuations. → the Hund's metal. - R. H. Mckenzie

A. Georges et al., Annu. Rev. Condens. Matter Phys. (2013).

MIT in numerous multi-orbital systems

In 3-orbtial system (calculation)



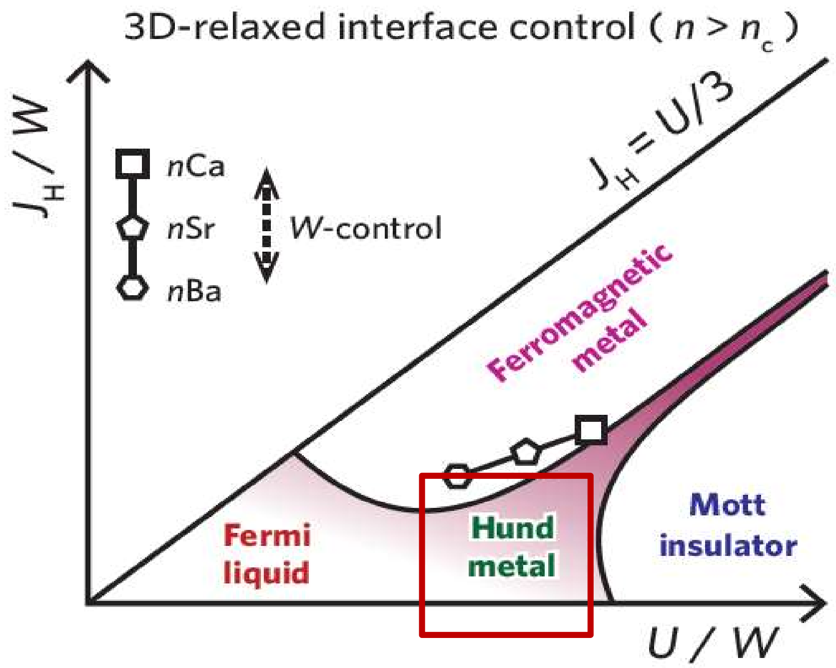
Black bar:
Mott insulating state

White arrow: increasing J

- MIT strongly depends on electron filling (Ambivalence of *J*).
- Near MIT, the Hund's metallic state can occur.

A. Georges et al., Annu. Rev. Condens. Matter Phys. (2013).

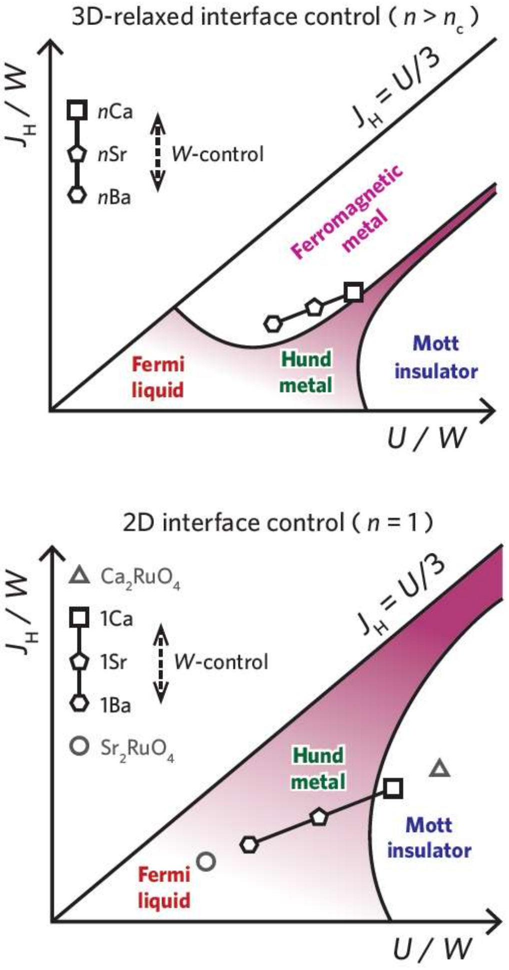
Difficulty to reach the Hund's metal in 3D $SrRuO_3$

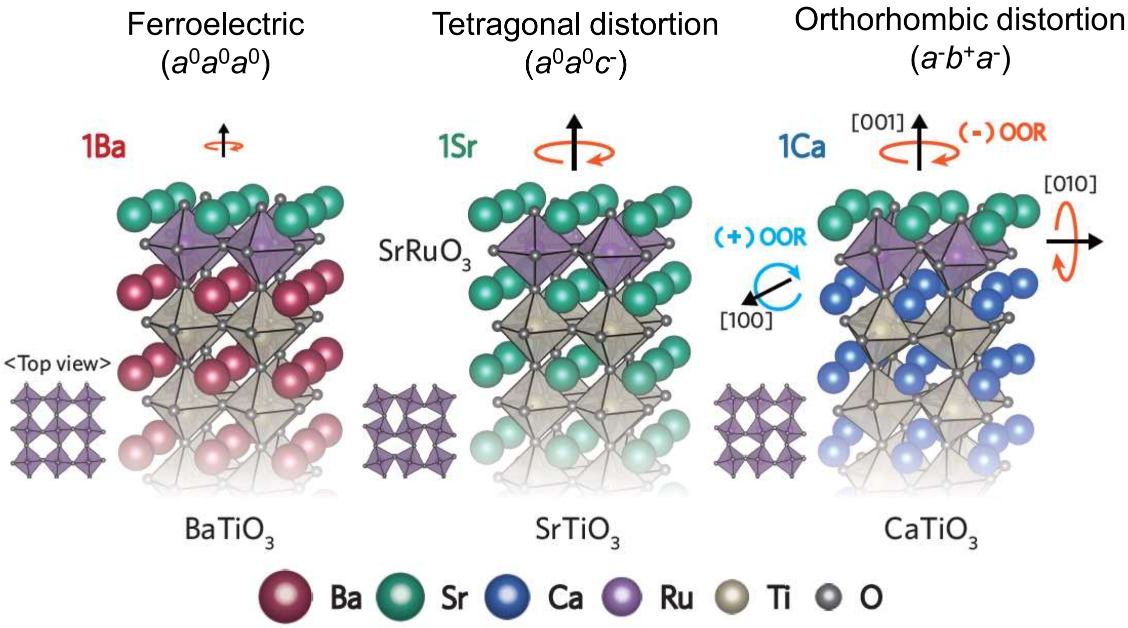


- By controlling the *U* and *J*, the multiple phases can be emerged in bulk SRO.
- However, **the Hund's metal near the MIT is difficult to reach.**

B. Sohn, J. R. Kim, TWN, C. Kim *et. al., Nat. Commun.* (2021)
J. R. Kim, TWN, C. Kim *et. al., Advanced. Maters* (2023)

MIT in the 2D SRO film through the Hund's metal state



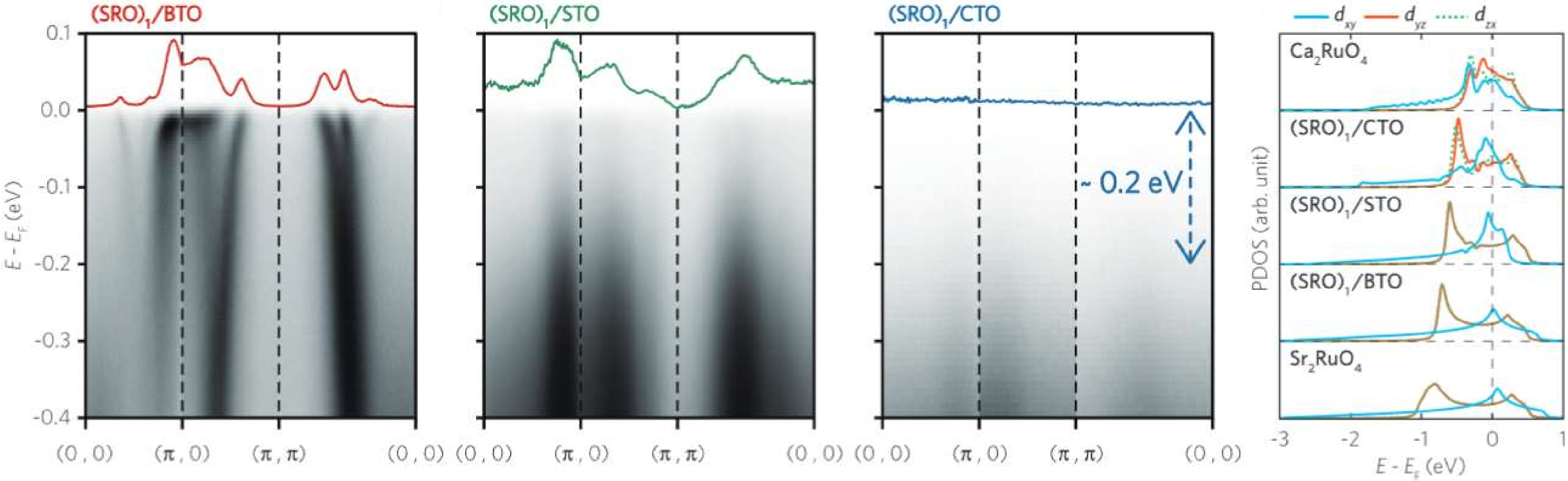


J. R. Kim, TWN, C. Kim *et. al., Advanced. Maters* (2023)

B. Sohn, J. R. Kim, TWN, C. Kim *et. al., Nat. Commun.* (2021)

MIT in 2D SRO film

- DFT calculations

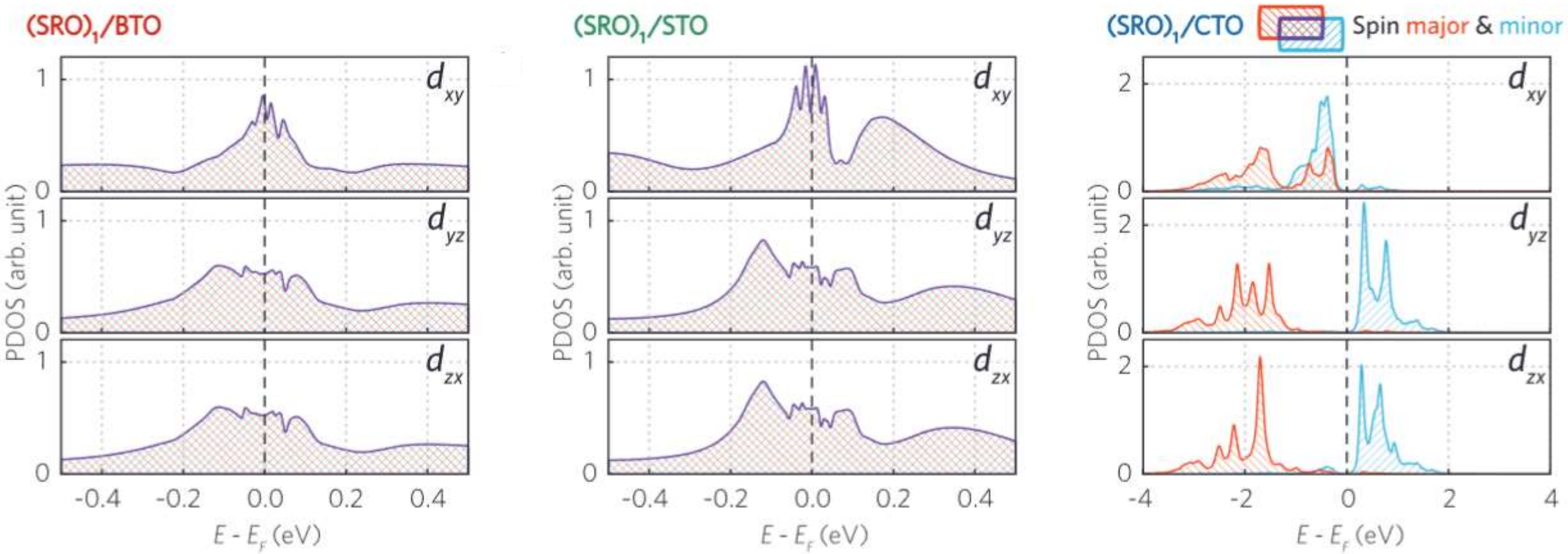


Based on our DFT calculation,

- 2D SRO/BTO is a coherent metal.
- a strong incoherency between DFT and experiments for 2D SRO/STO and SRO/CTO → indicating presence of a strong electronic correlation.

MIT in 2D SRO film

- DMFT calculations



Coherent Metal

Incoherent Metal

Insulator

$a^0a^0a^0$

$a^0a^0c^-$

$a^-b^+a^-$

Fermi liquid

Hund's metal

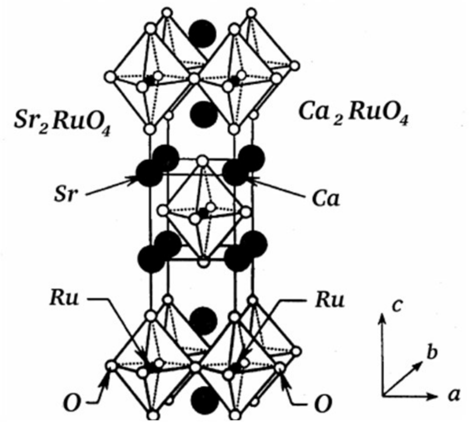
Mott insulator

Content

1. Introduction
   - ☐ Emergent phenomena in transition metal oxides and their heterostructures
   - ☐ Our experimental setup : cluster system for PLD, *in-situ* ARPES, RHEED & LEED
   - ☐ Basic Properties of $SrRuO_3$
2. MIT in $SrRuO_3$ ultrathin films
   - ☐ Fundamental thickness limit of SRO films
   - ☐ Control of MIT in 2D SRO films (i.e. 1 *u.c*) by controlling the titling angle
   - ☐ Strain Engineering of electronic properties of 2D SRO films
3. Dual ferromagnetism in $SrRuO_3$ thick films
4. Topological band structures of $SrRuO_3$: Nodal features
5. Summary and Outlooks

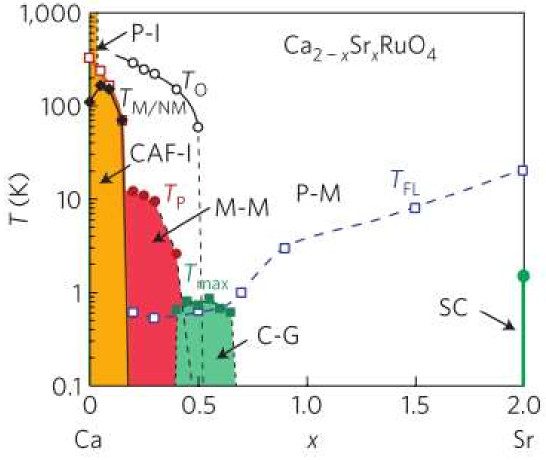
ibS CCES
Center for Correlated Electron Systems

Revisit of octahedron distortions in $Ca_{2-x}Sr_xRuO_4$



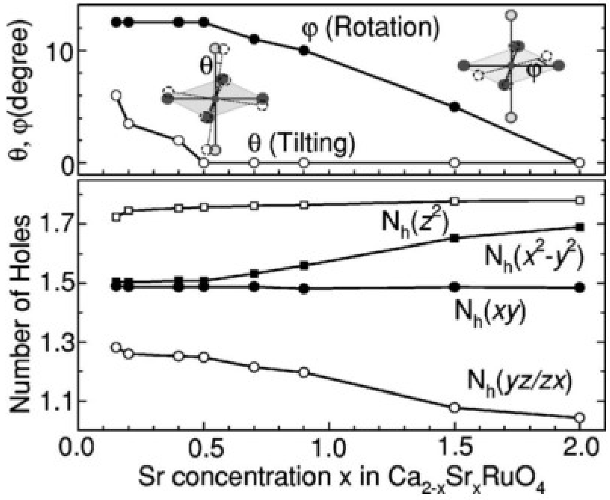
$Ca_{2-x}Sr_xRuO_4$

CSRO phase diagram



J. P. Carlo, Nature Materials (2012)

Octahedron rotation & tilt

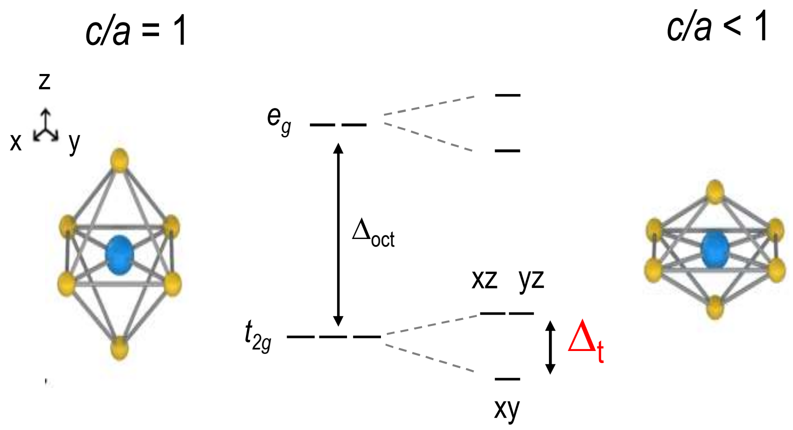


H.-J. Noh et al, PRB (2005)

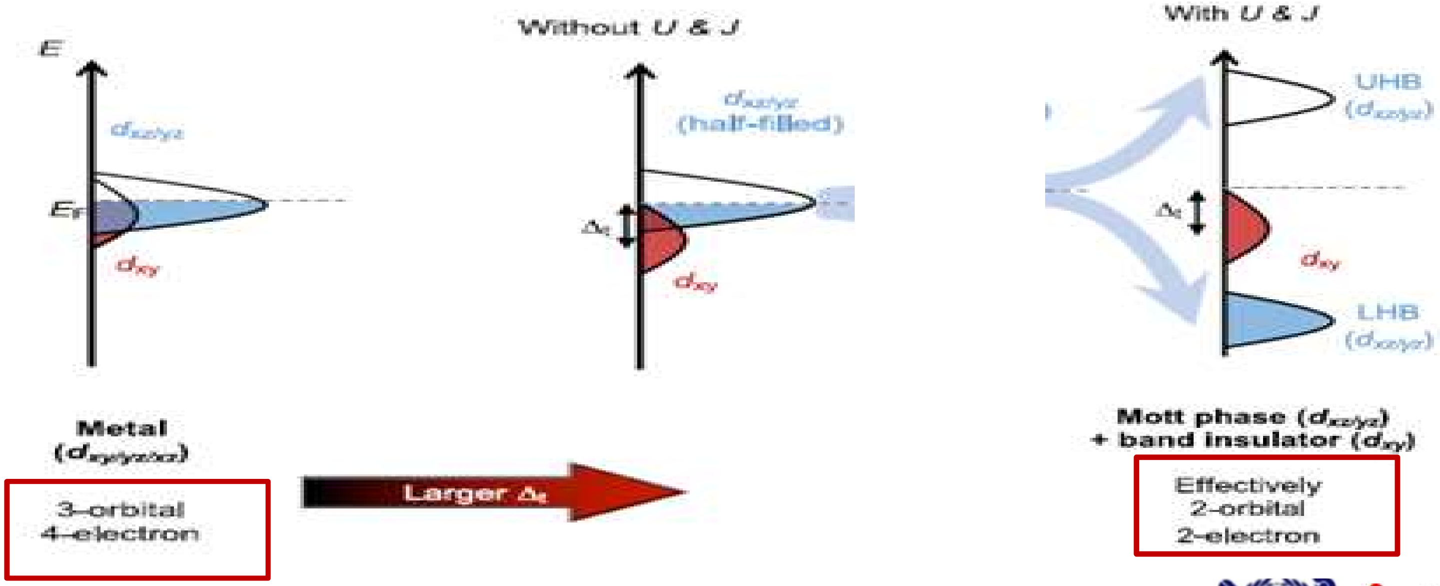
By changing the θ and ϕ, the crystal field splitting can be changed.

→ changes in the occupation number

Tuning of the crystal field splitting in $SrRuO_3$



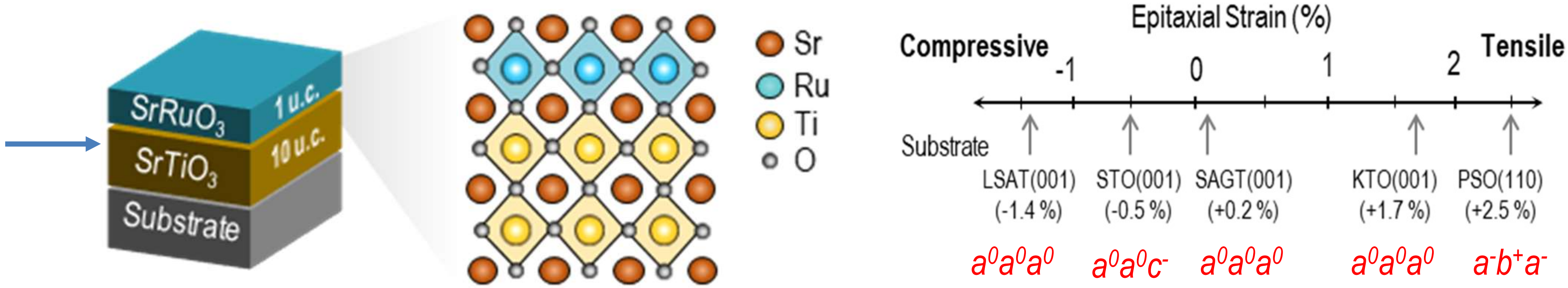
By tuning the crystal field splitting, we can change the energy levels of the Ru t_{2g} orbitals. It effectively varies the filling of orbitals in the $SrRuO_3$ films.



Experimental approaches using strain engineering.

- Difficulties in conventional strain engineering

$a^0a^0c^-$



Due to the proximity effects from substrate, the crystal structure of films will vary depending on the OOR of substrate. → crystalline symmetry breaking !

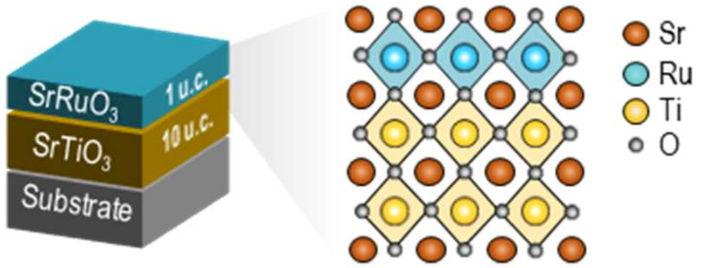
→ How to change lattice constants of the ultrathin films on different substrates with various OOR ?

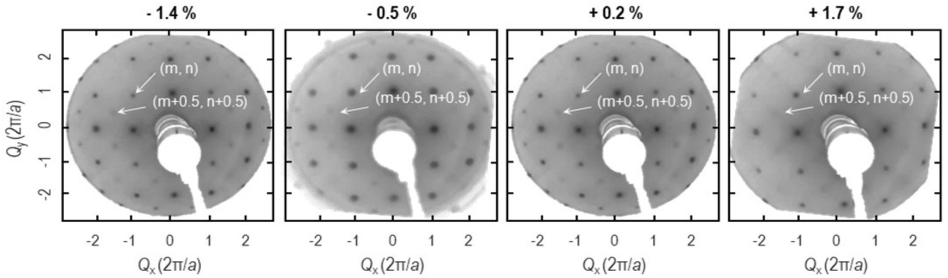
E. K. Ko, C. Kim, TWN *et. al.*, *Nature Comm.* (2023)

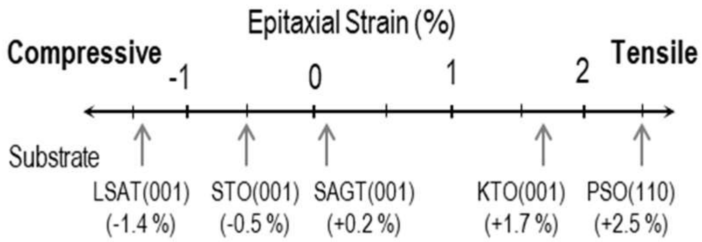
Strain engineering in SRO monolayer

- Symmetry preserving strain engineering

$a^0a^0c^-$
(tetrahedral)





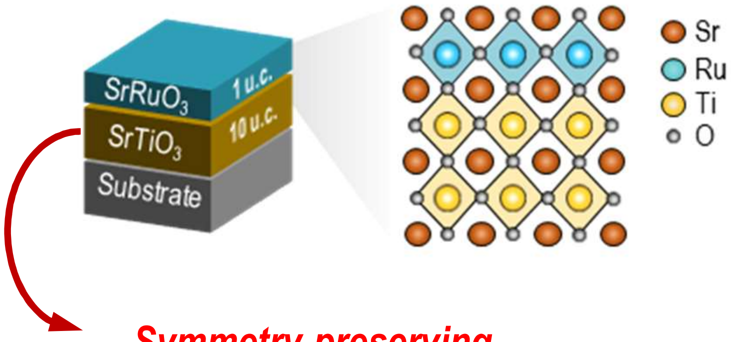


$a^0a^0c^-$

The STO layer guarantees that all SRO layers on 5 different substrates will have the same crystal symmetry of the $a^0a^0c^-$ OOR pattern.

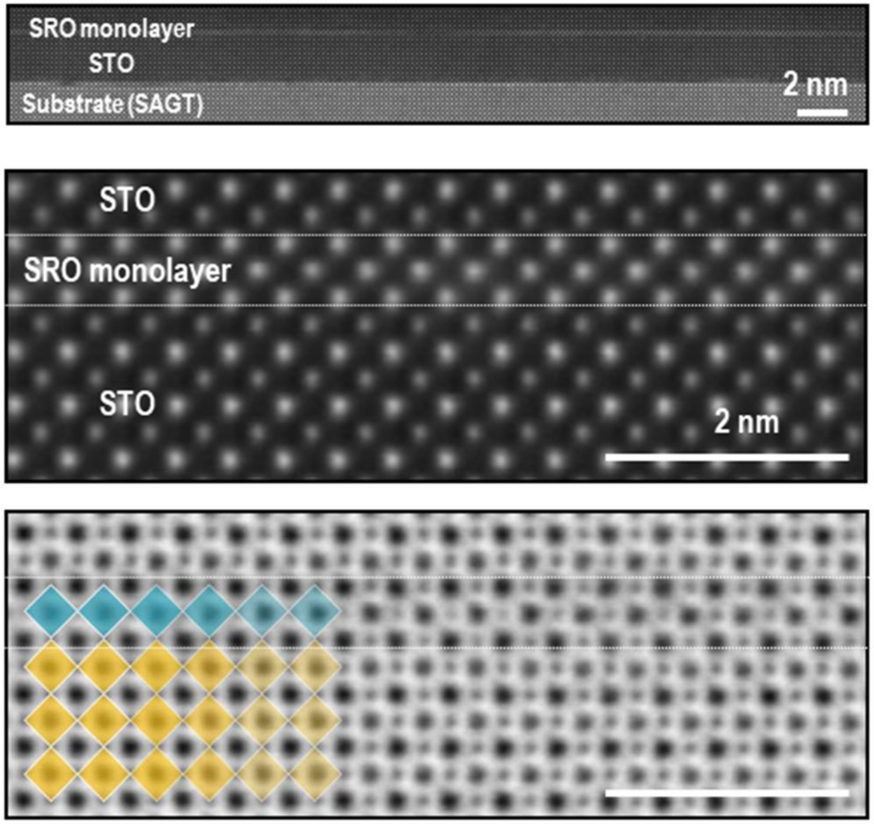
E. K. Ko, C. Kim, TWN *et. al.*, *Nature Comm.* (2023)

Structural characterization: STEM-HAADF



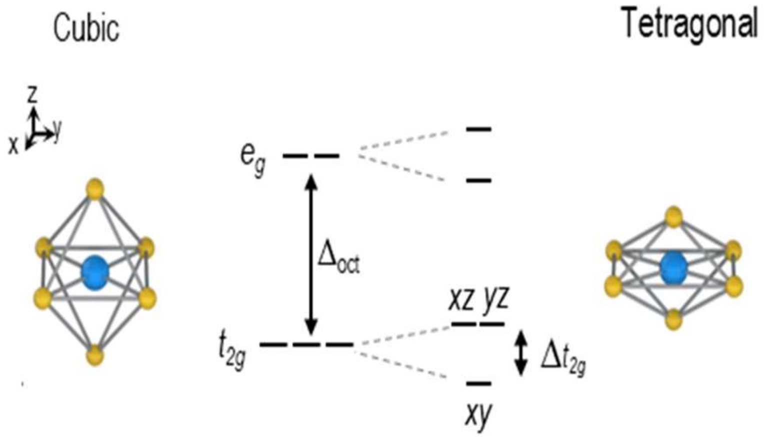
Symmetry-preserving buffer layer

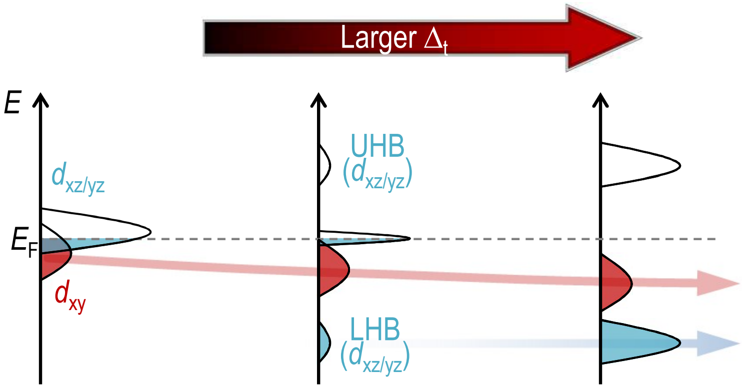
High-quality of SRO monolayers.



Strain engineering in SRO monolayer

- Symmetry-preserving epitaxial strain engineering





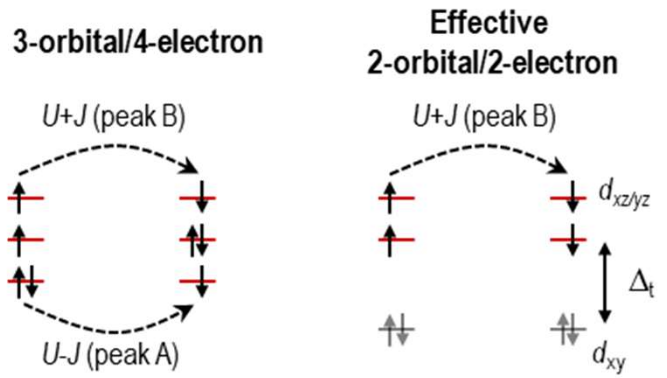
Insulating Mott system
effectively 2 electrons/2 orbitals

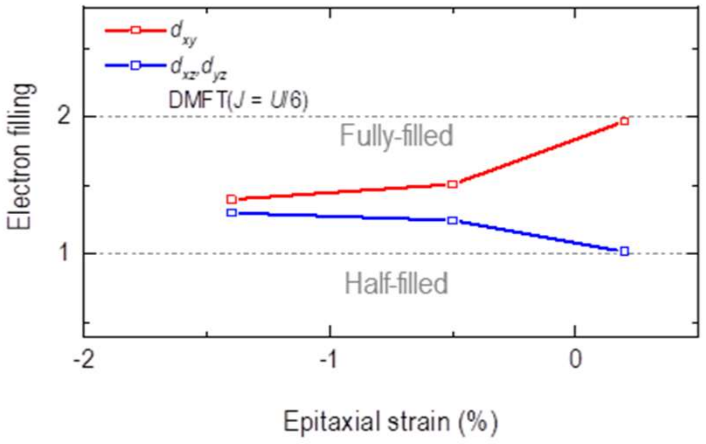
- By controlling the crystal field splitting by applying different in-plane strain, we can tune the associated band structure.
- In particular, the 2D SRO monolayer can exhibit MIT depending on different strain strength.→ possibility of appearance of new phases

E. K. Ko, C. Kim, TWN *et. al., Nature Comm.* (2023)

Strain-dependent orbital occupancy changes

(3-orbital/4-electron) → (2-orbital/2-electron)

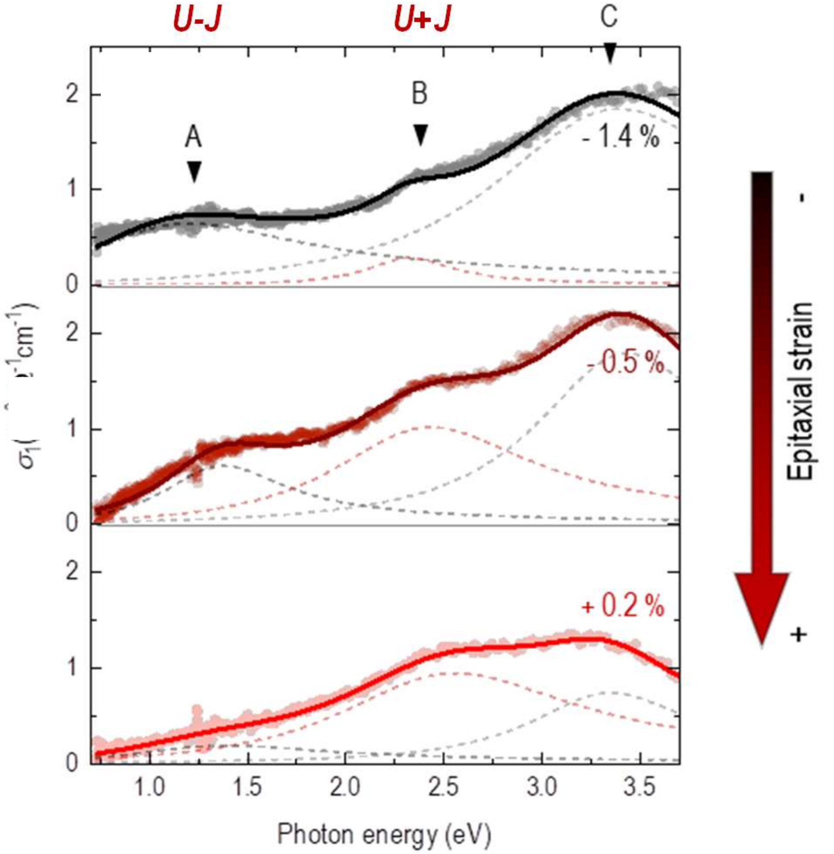




_d-d_ transition
_U-J_

_d-d_ transition
_U+J_

Charge transfer peak
O 2p → Ru t_{2g}



J. S. Lee, TWN, Y. Maeno *et al.*, Phys. Rev. Lett. (2002)
J. S. Lee, TWN *et al.* Phys. Rev. B (2001)

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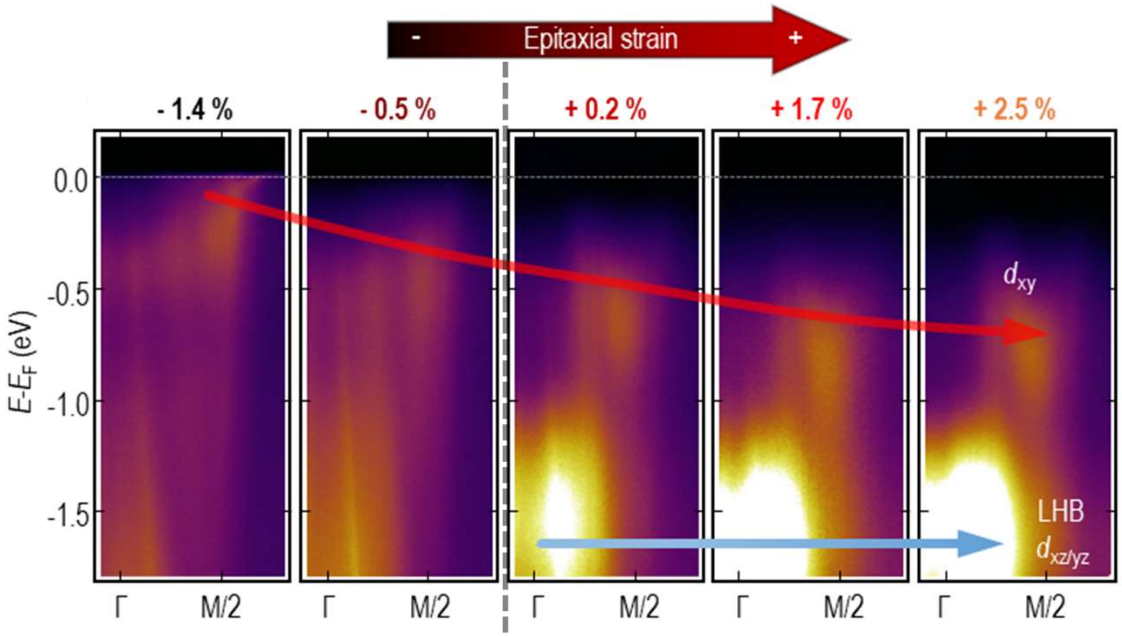
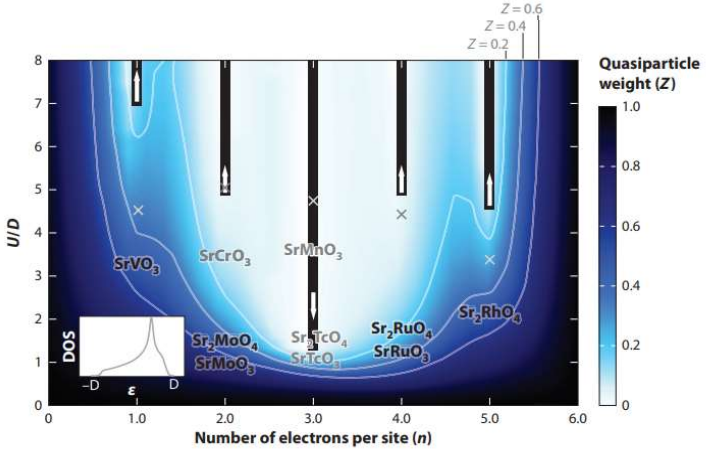
Concurrent opening of two types of gap

Energy-momentum dispersions at 6K



Band Insulating (d_{xy})

Mott gap ($d_{xz/yz}$)



MIT

- There are two types of gap opening:
  band insulator and Mott gap at MIT limit.
- Matt gap opening becomes feasible due to the half-filling of 2 orbitals (i.e. $d_{xy/yz}$) system

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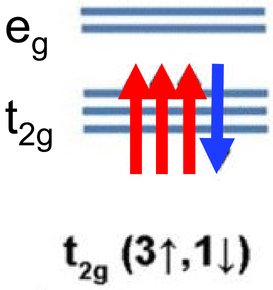
5. Summary and Outlooks

Itinerant vs local ferromagnetism in SRO

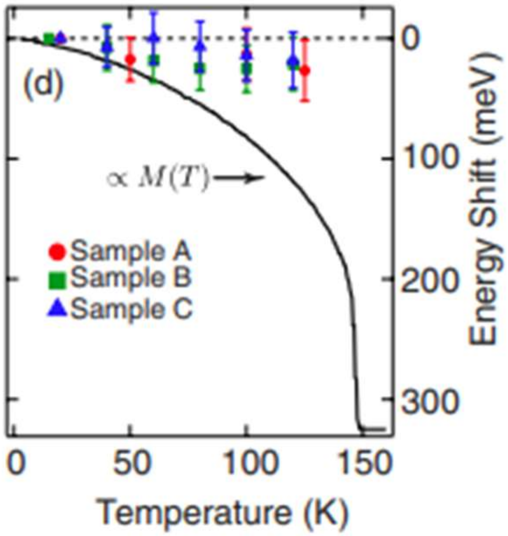
Non-integer magnetic moment



S = 2 expected in the local picture

Measured magnetic moment of ~1.6 μ_B

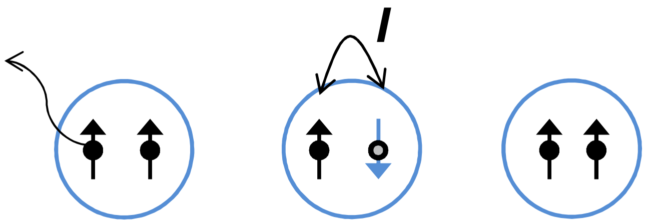
Temperature independent band splitting



Phys. Rev. Lett. 110, 087004 (2013)

Spectroscopic signature of the magnetism?

Stoner theory for ferromagnetic metals



Stoner criterion: $I N_0 > 1$

T_C estimation: $$T_C^2 = R\left(1 - \frac{1}{IN_0}\right)$$

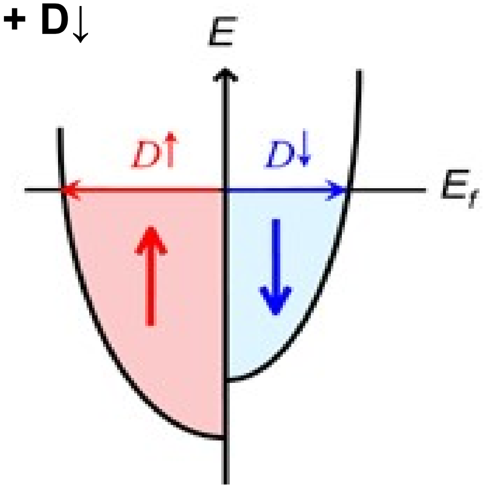
I : Stoner parameter (spin-spin interaction)

N_0 : DOS at E_F

N_0' & N_0" : 1st & 2nd derivative of DOS at E_F

$$R = \left(\frac{6}{\pi^2}\right)\left[\left(\frac{N_0'}{N_0}\right)^2 - \left(\frac{N_0''}{N_0}\right)\right]$$

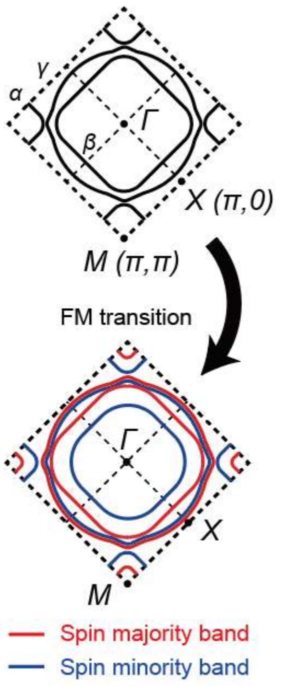
$N_0 = D\uparrow + D\downarrow$



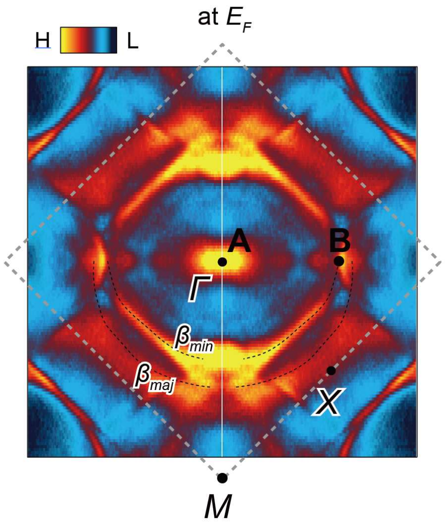
Origin of ferromagnetism in SRO: Stoner Ferromagnetism ?

ARPES : FM feature near E_F

Schematic FS

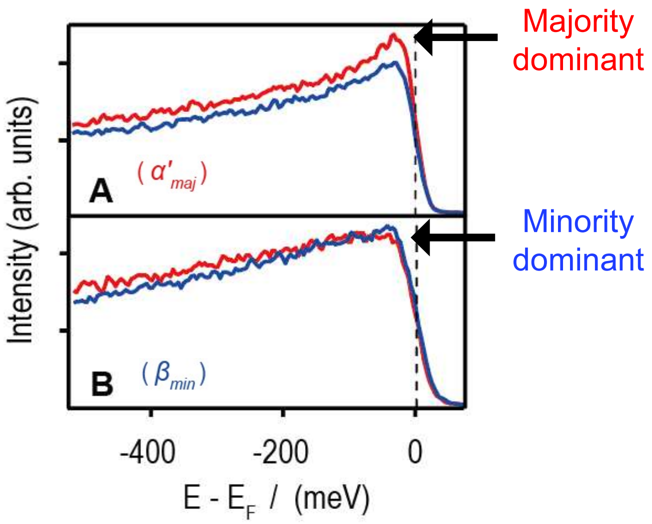


Measured Fermi surface



15 u.c. $SrRuO_3$ film

Spin ARPES



Spin polarization in dispersive bands

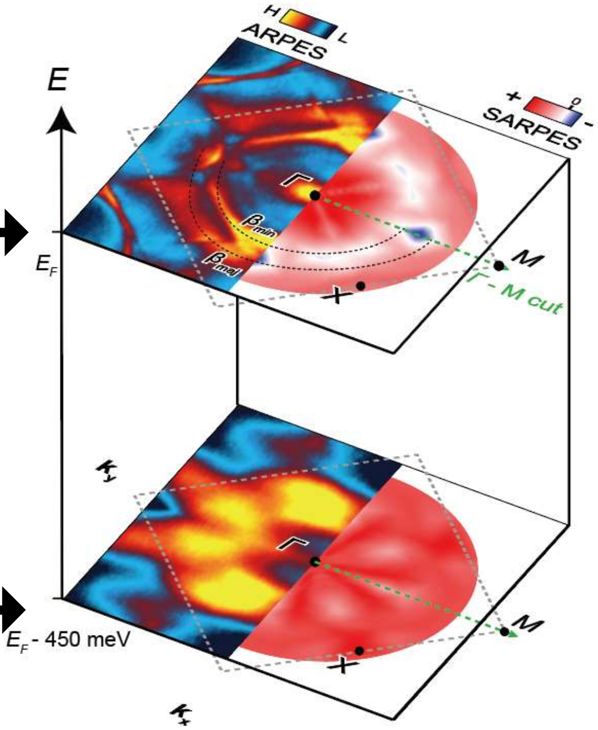
Observation of spin-polarized bands in ferromagnetic $SrRuO_3$ films

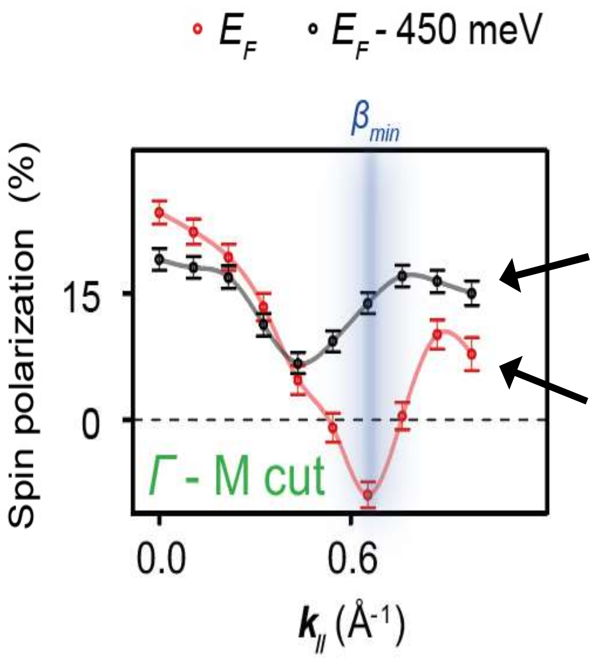
Localized FM feature at high energy

Energy dependent spin-polarization

Low E itinerant electrons

High E electrons





Local spin (small k dep.)

Itinerant spin (large k dep.)

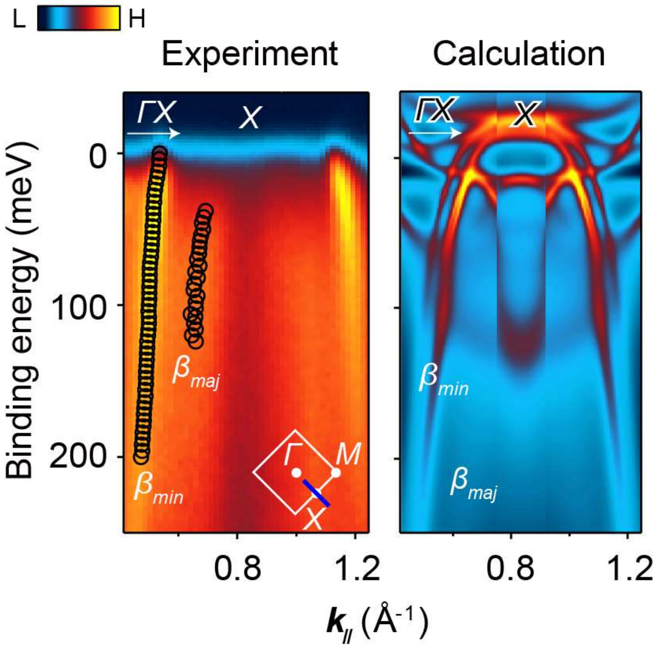
Dual character !!!

High energy electrons has a local spin character!

High energy electrons are strongly spin-polarized.

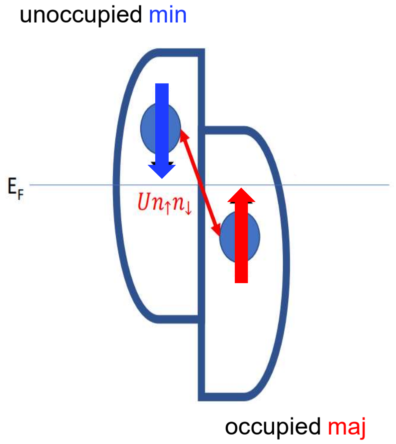
Spin-dependent correlation

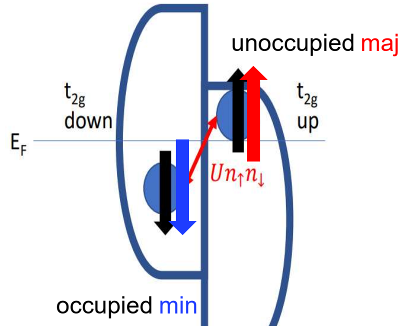
Difference in coherence



Broad majority and sharp minority bands

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





Wide interaction channel
: strong correlation for majority

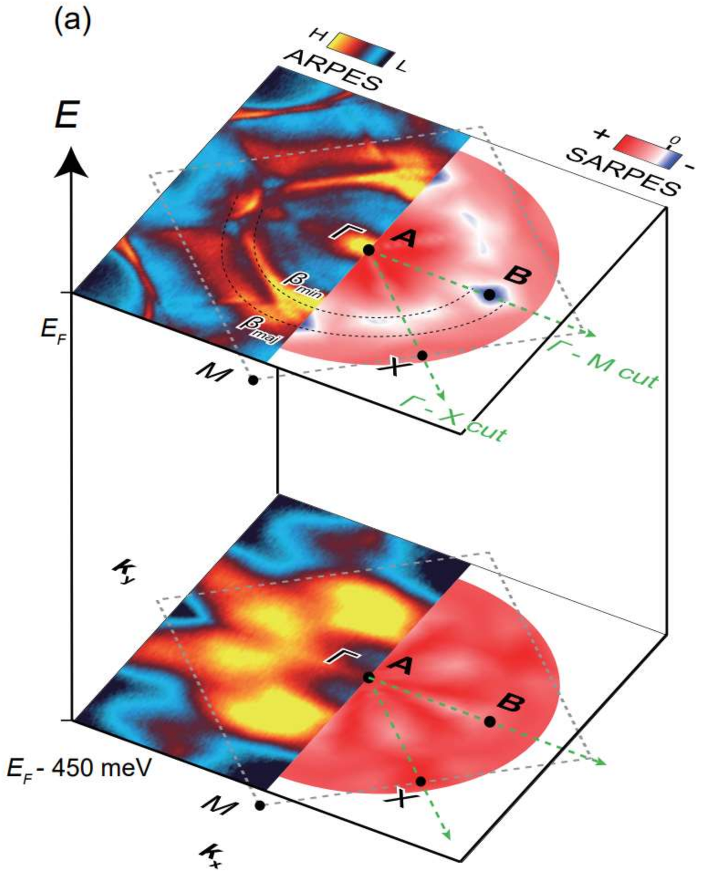
Narrow interaction channel
: weak correlation for minority

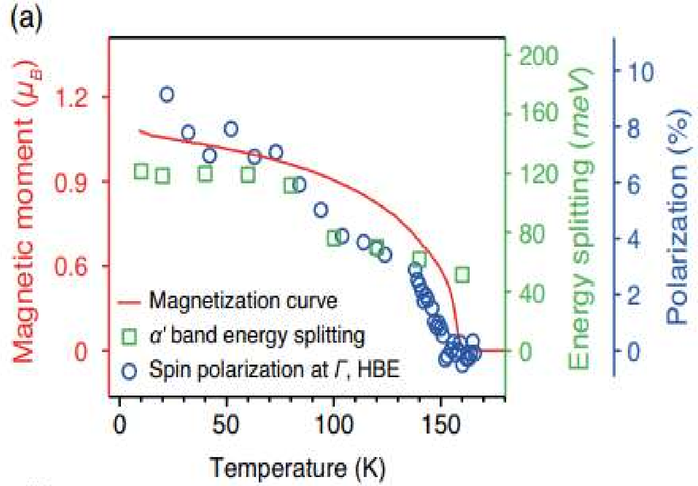
Localized spin majority electrons & itinerant spin minority electrons

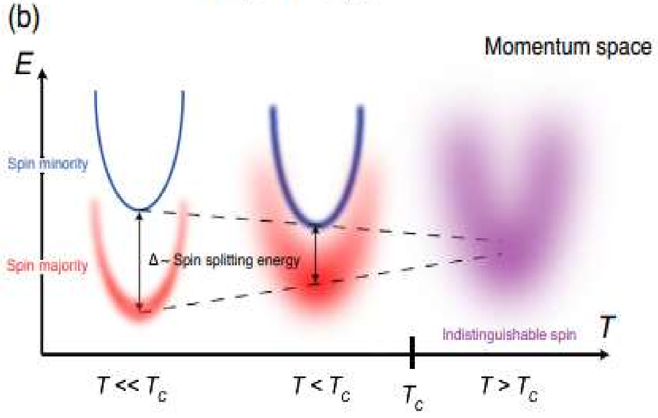
M.J. Kim et al, PRB 91, 205116 (2015)

Ferromagnetism in SrRuO$_3$ film : ARPES studies

15 u.c.-SrRuO$_3$ thin films



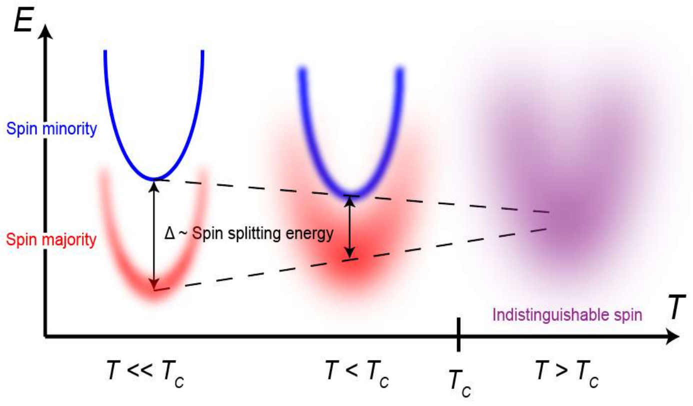




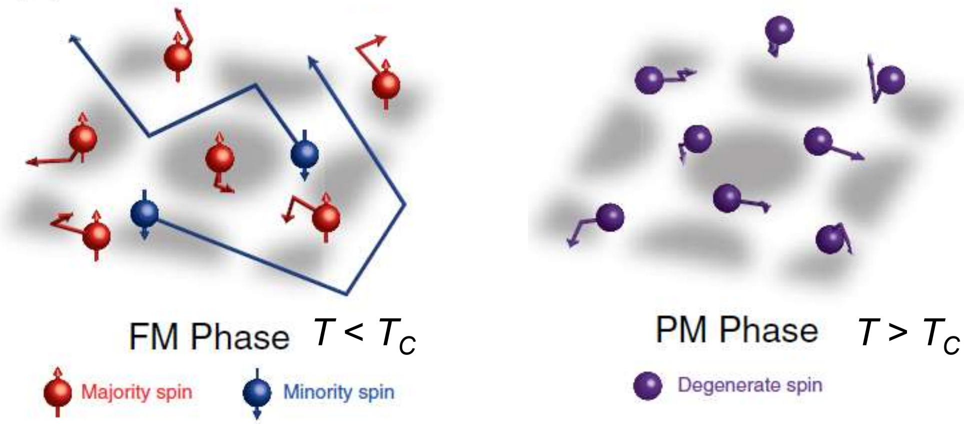
S. Hahn, TWN, C. Kim *et. al., Phys. Rev. Lett.* (2021)

Pictorial illustrations

Momentum space



Real space



- We observed spin-dependent electronic structure and dual ferromagnetism in thick SRO film.
- Below T_c, the majority and minority spin bands can have a large spin-split gap.
→ Ferromagnetic phase.
- Above T_c, the two spin bands become more incoherent and cannot be distinguished.
→ Paramagnetic phase.

S. Hahn, TWN, C. Kim *et. al., Phys. Rev. Lett.* (2021)

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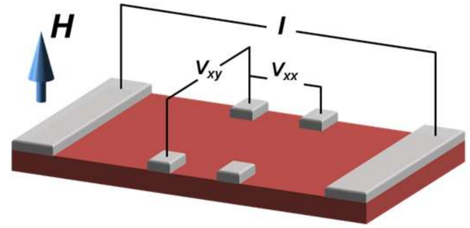
Anomalous Hall effects of SrRuO$_3$

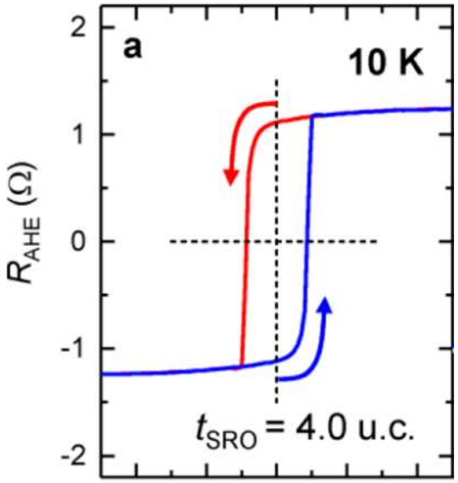
Hall measurement

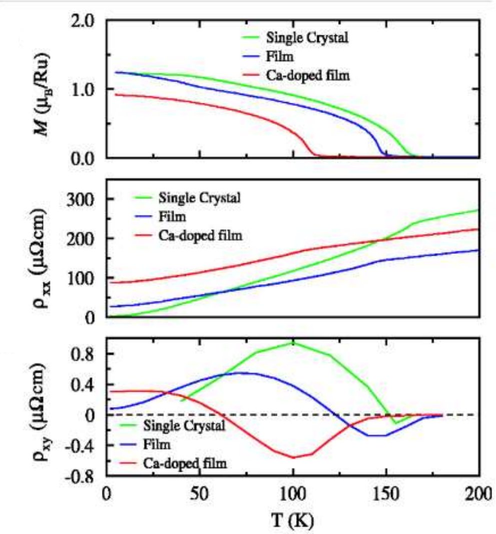


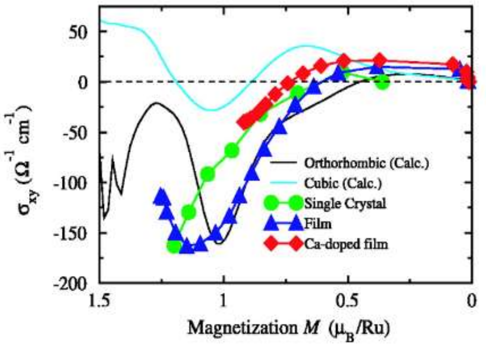
$$\rho_{xy} = R_0 B + 4\pi R_s M$$

Ordinary Hall signal

Anomalous Hall Effects (AHE)





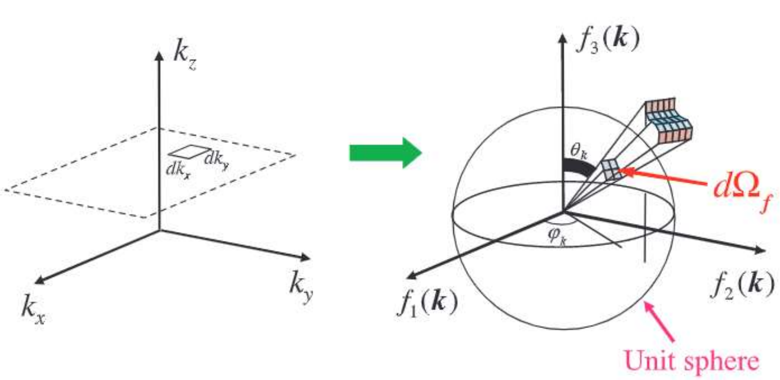


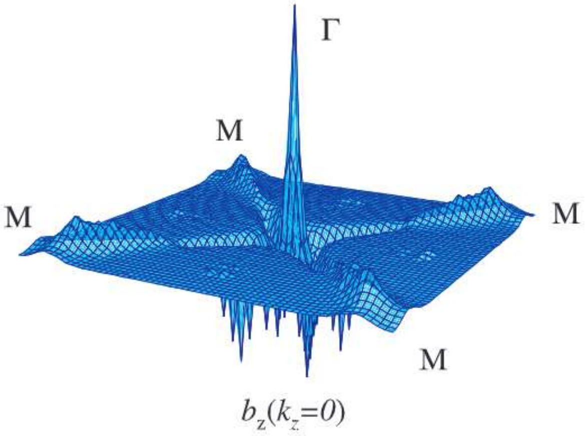
- Anomalous Hall effects (AHE) in SrRuO$_3$, due to the Berry curvature.
- However, the related electronic structure has not be reported experimentally.

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

Magnetic monopoles in *k*-space of $SrRuO_3$

Anomalous Hall effect (AHE) & magnetic monopoles in **k**-space





$$\rho_{xy} = R_0 B + 4\pi R_s M$$

$$\sigma_{xy} = \sum_{n,\mathbf{k}} n_F[\varepsilon_n(\mathbf{k})] b_z(\mathbf{k})$$

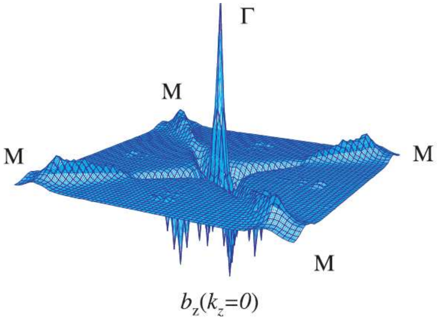
Ferromagnetic spin structure in r-space
→ magnetic monopoles in k-space
: experimentally observed in 3D magnetic metals with Weyl points
& Fe_3GeTe_2 (with NLs of 3D spin-polar. band)

However, not experimentally confirmed in $SrRuO_3$ yet.

Z. Fang, N. Nagaosa, *et. al.,* Science (2003)

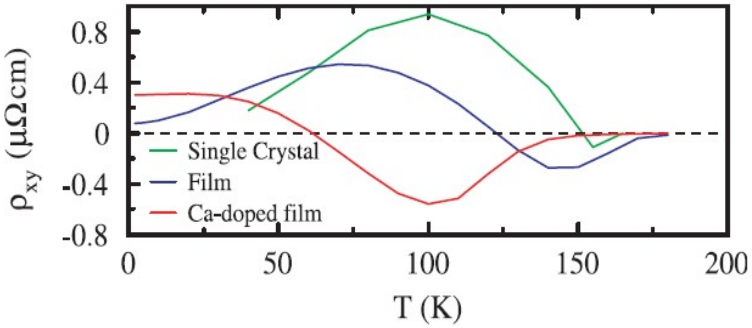
Magnetic monopoles in SRO?

Magnetic monopoles
in the momentum space



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

Sign changing anomalous Hall
effect in $SrRuO_3$



topological transport in itinerant FM ← Magnetism & SOC

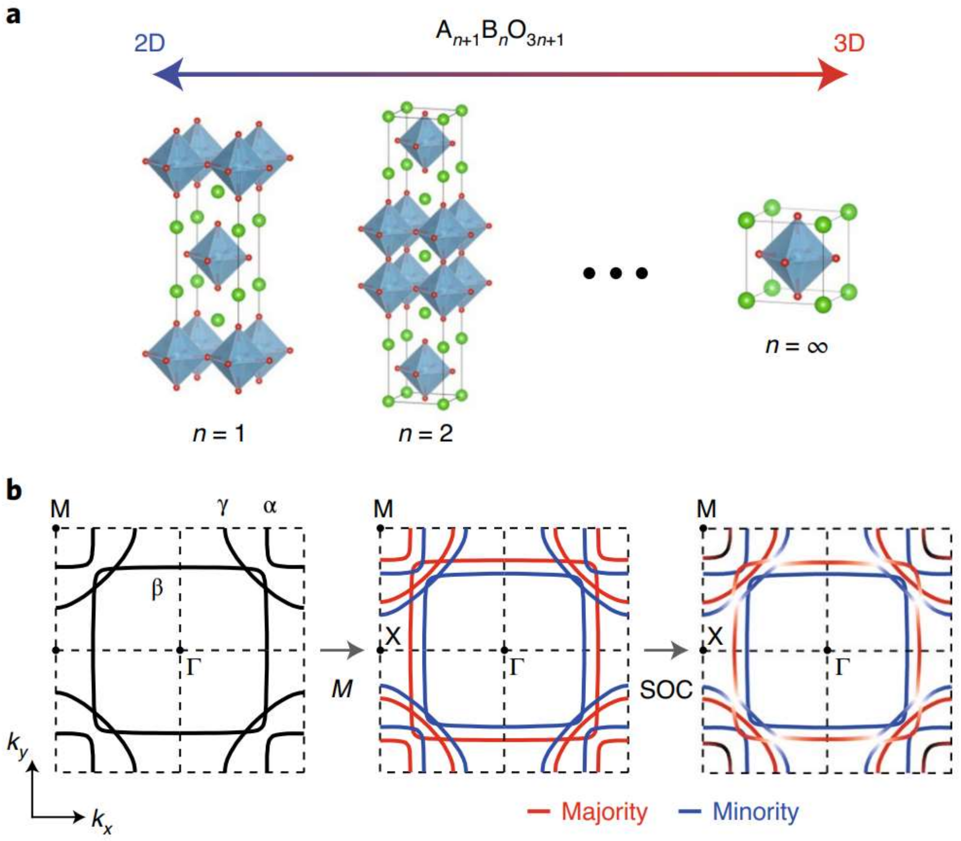
(1) spin-polarized bands with nodal points/lines
& (2) band degeneracy can be lifted by SOC

→ A source of Berry curvature, leading to a large AHE

It was theoretically predicted that 2D spin polarized bands of perovskite oxides generally can support symmetry protected nodal lines and points → AHE

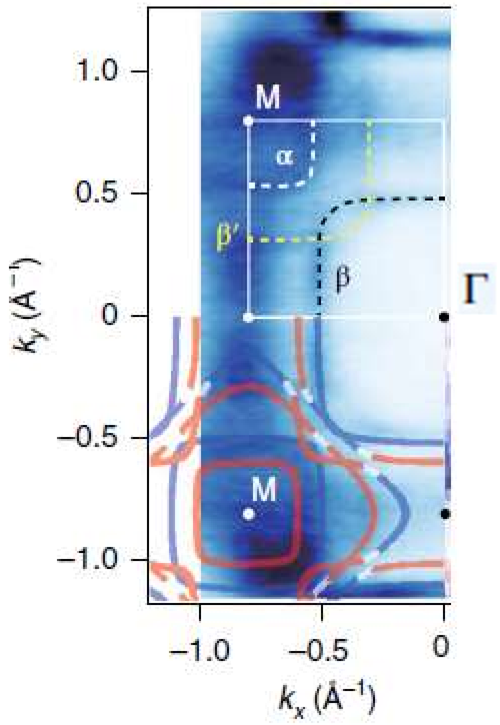
B. Sohn, TWN, C. Kim *et. al.*, *Nat. Mater.* (2021)

Electronic Structure of a $SrRuO_3$ ultrathin film



4uc SRO film

Synchrotron ARPES (80 eV)



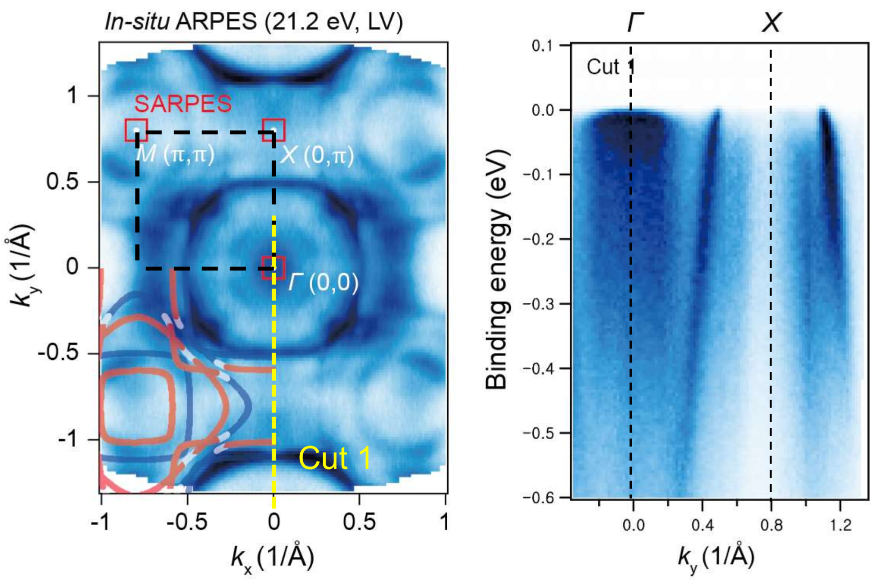
α , β : $d_{xz,yz}$ orbitals (1D-character)
Γ : d_{xy} orbital (2D-character)

(Question) Do it has nodal structures in the FM state?

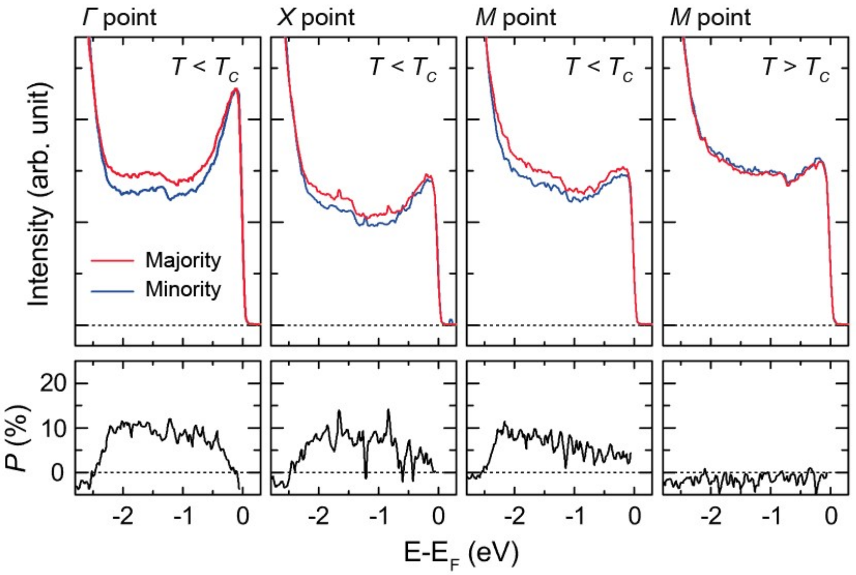
B. Sohn, TWN, C. Kim *et. al.*, Nature Materials (2021)

Spin polarization in 4 u.c. SRO film

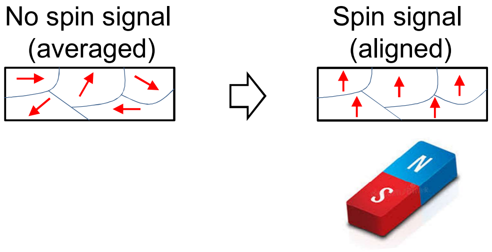
Fermi surface map and high symmetry cut



Spin-resolved ARPES (SARPES) from selected points



In situ magnetization



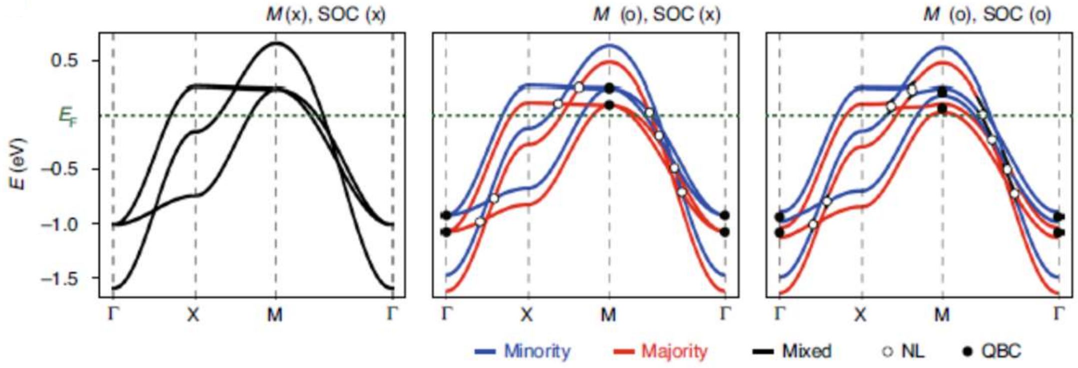
- Clear polarization below T_c was observed in spin-resolved ARPES

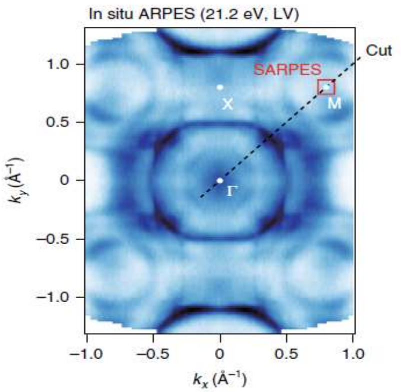
  → consistent with itinerant FM.
- We also observed band dispersion in SARPES → Origin?

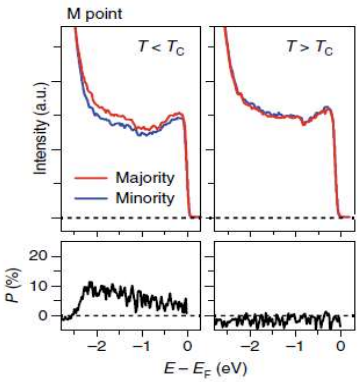
B. Sohn, TWN, C. Kim *et. al.*, Nature Materials (2021)

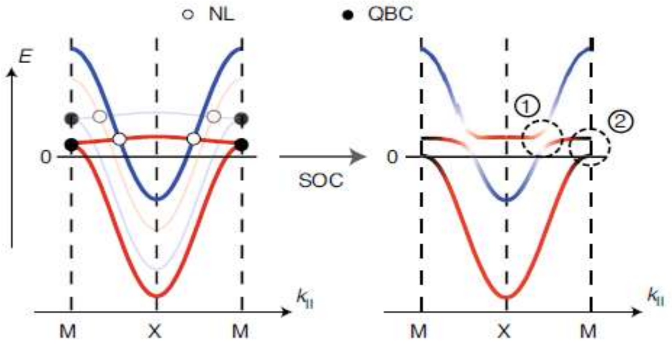
2D symmetry-protected nodal structures in ferromagnetic films

Tight-binding model (with M=0.33 μ_B per Ru)









2D FM perovskite of t_{2g} system can possess stable nodal structures :

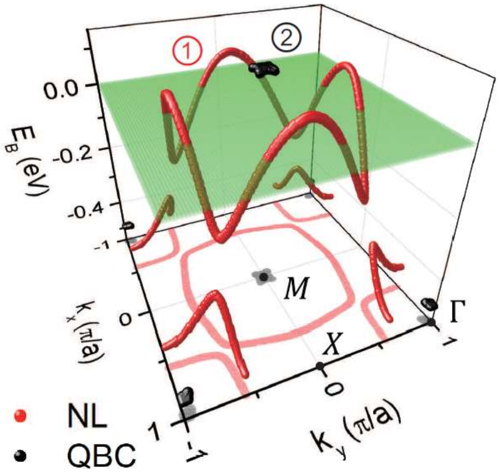
(i.e. nodal lines (NL) & quadratic band crossing (QBC)

with crystal and rotational symmetry, protecting topological features.

B. Sohn, TWN, C. Kim *et. al., Nat. Mater.* (2021)

Sign changes in AHE: Berry curvature due to Nodal structure

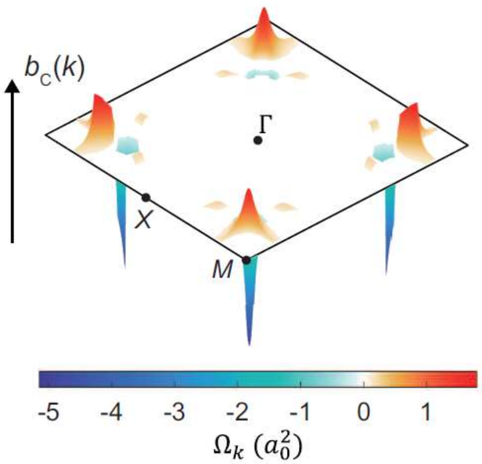
Berry curvature sources

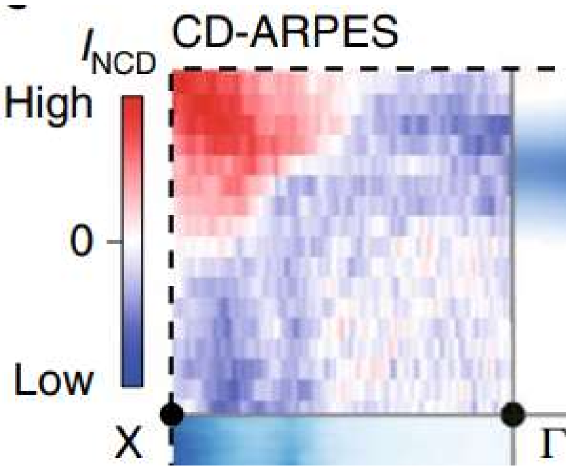


① Nodal lines (β - γ band crossing)

② Quadratic band crossing at M

Calculated Berry curvature
of 2D SRO





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

Quadratic band crossing (QBC) near the M point mainly responsible for the AHE.

The sign of AHE can change as the magnetization (or Fermi energy) is varied due to Berry curvature near the Fermi level induced by the nodal structures.

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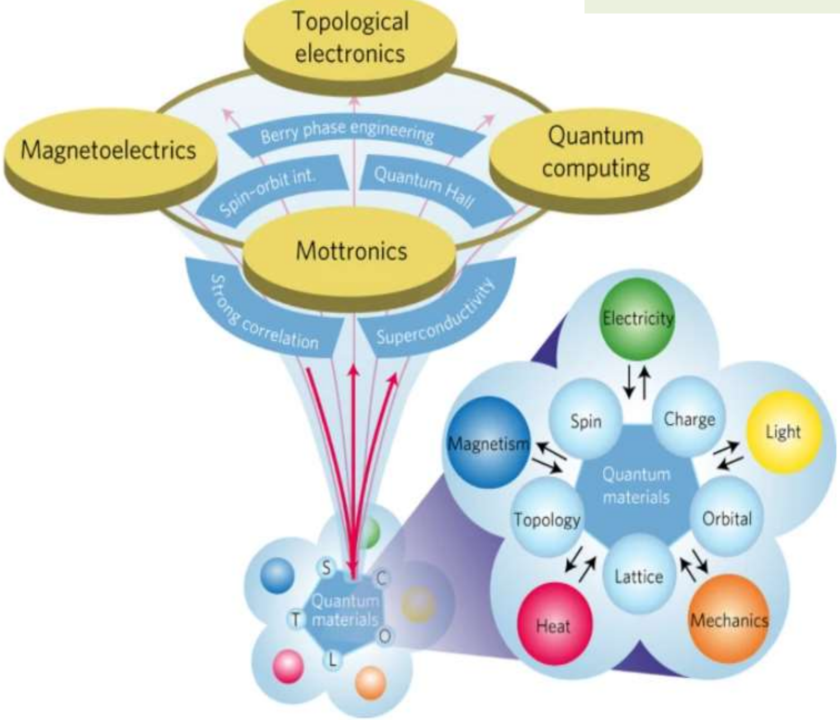
Summary on $SrRuO_3$ thin film studies

- We recently built a cluster system with atomic-scale epitaxy and angle-resolved photoemission spectroscopy (ARPES) to unravel the physics of conducting TMO films. [B. Sohn et al, Nature Comm. **12** 6171 (2021)]
- The fundamental thickness limit of SrRuO3 films on SrTiO3 substrates are 1 uc for metallicity and 3 uc for ferromagnetism, consistent with our earlier predictions. [Y. Chang et al., PRL 103(5), 057201 (2009)]
- In this 2D limit, we can manipulate a metal-insulator transition by controlling the rotation of the RuO_6 octahedron. We obtained some experimental indications of Hund's metallicity. [J.R. Kim et al., Adv. Mat. **35** 2208833 (2023)]
- We can split d_{xy} and d_{yz}/d_{zx} bands of 2D $SrRuO_3$ by changing crystal field splitting with the symmetry-preserved strain engineering technique. This tuning effectively changes the numbers of electrons and involved orbitals. We observed two types of gap opening: Band insulator and Mott gap near the MIT. [E. K. Ko et al., Nature Comm. **13** 3572 (2023)]
- We also addressed the nature of ferromagnetism by using spin-resolved ARPES. [S. Hahn et al., Phys. Rev. Lett. **127** 256401 (2021)] Lastly, we investigated symmetry-protected nodal structures in ferromagnetic $SrRuO_3$ films [B. Sohn et al, Nature Mater. **20** 1643 (2021)].

Outlooks on transition metal oxides

- Transition metal oxides is a fertile ground for investigating novel emergent phenomena arising from the interplay between degrees of freedom.
- Numerous strong interactions, such as on-site Coulomb interaction (*U*), Hund's rule coupling (*J*), and or spin-orbit coupling (SOC).



Y. Tokura *et al.*, *Nat. Phys.* (2017)

❑ TMO for Emergent phenomena

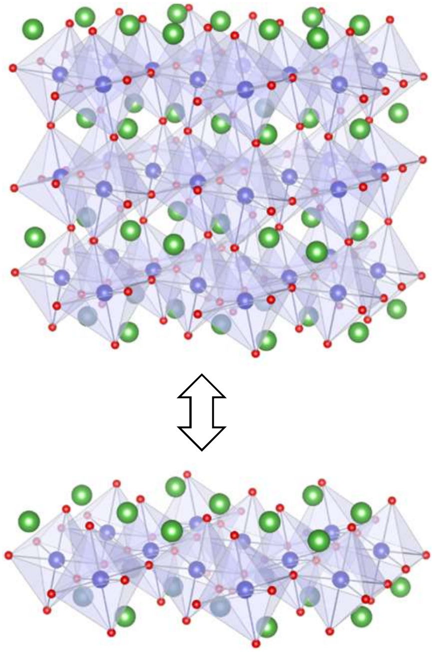
- High-T_c superconductor in cuprate (1980s)
- Colossal magnetoresistance in magnates (1990s)
- Sr_2RuO_4 (*Science*. (1998), Prof. Maeno)
- $SrRuO_3$ (*Science*. (2003), Prof. Nagaosa)
- Sr_2IrO_4 (*Phys. Rev. Lett*. (2008), Prof. B. J. Kim)
- $BaBiO_3$ (*Nat. Phys.* (2013), Prof. C. Felser)
- $[SrIrO_3/SrRuO_3]_n$ (*Sci. Adv*. (2016). Prof. J. Matsuno)
- $Nd_2Ir_2O_7$ (*Sci. Adv.* (2021), Prof. T. W. Noh)
- ……..

Ultrathin TMO film platforms

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

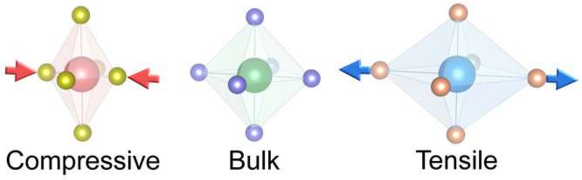
Dimensionality

Bulk crystal
3-D



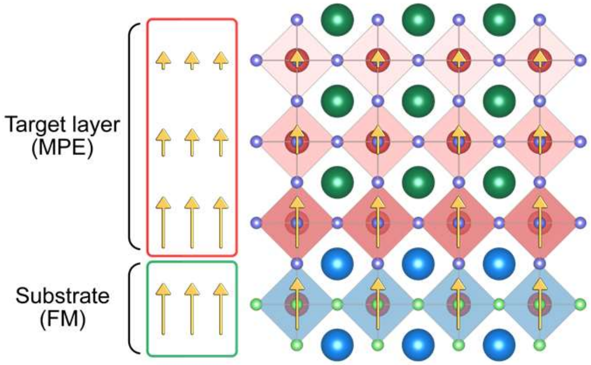
Ultrathin film (monolayer)
2-D limit

Epitaxial strain



Substrate-induced strain.

Proximity effect at the interface



Symmetry-breaking ferromagnetic, ferro electric layers as substrates.

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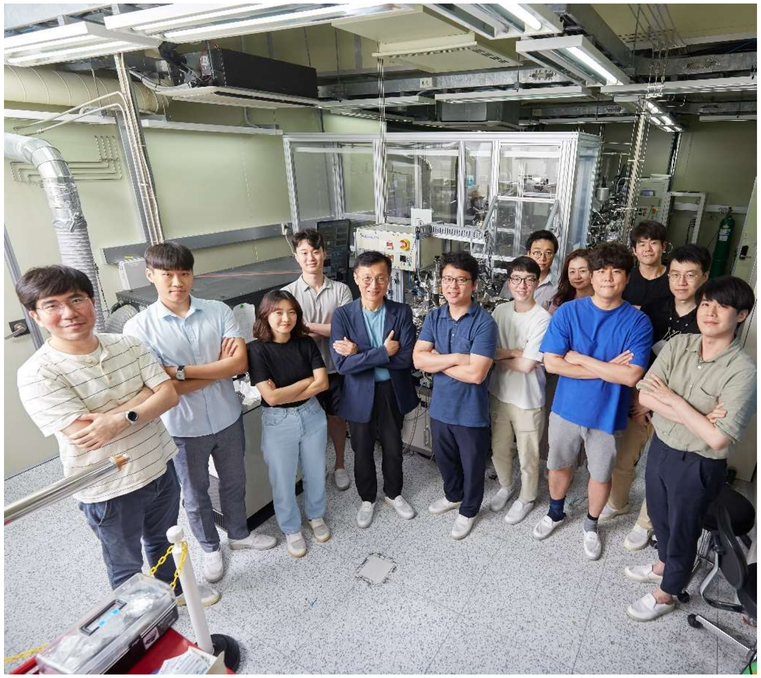

Atomic Scale Epitaxy group (early 2020)

Thank you very much! (~ 33 years in SNU)

ibs CCES
Center for Correlated Electron Systems