Mainz, Germany: 13 Nov. 2019
Novel Electronic and Magnetic Phases in Correlated Spin-Orbit Coupled Oxides

Importance of the quantification of Coulomb interaction in 5d oxides

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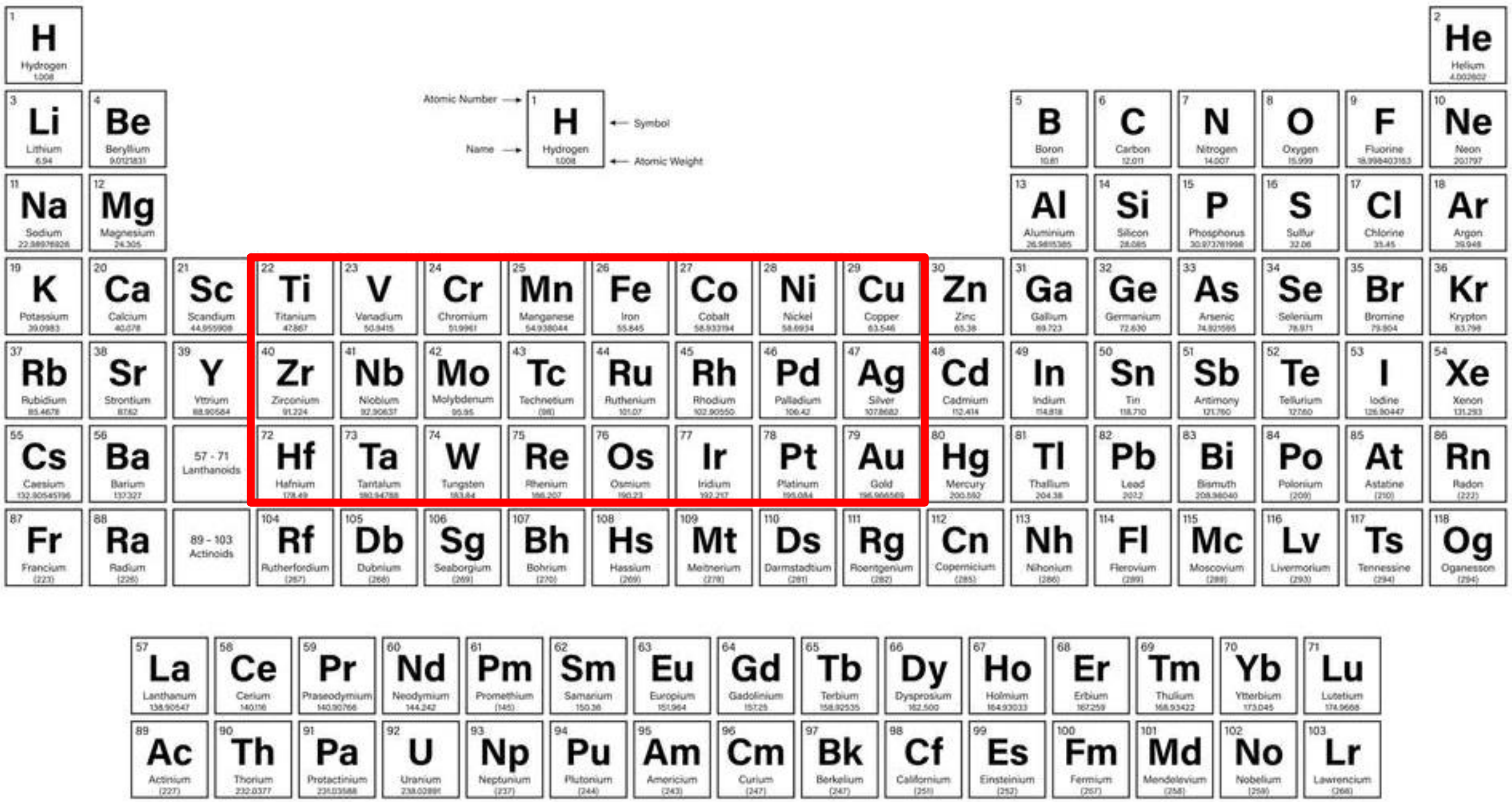
Abstract

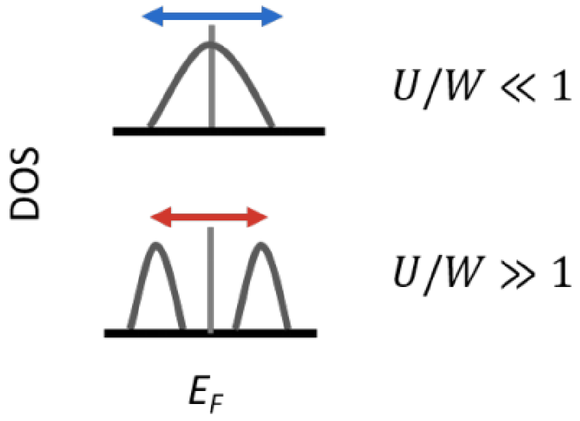
- **Introduction & Motivation**
- **iridates**
- **osmates**
- **Summary**

- **Transition metal oxides**

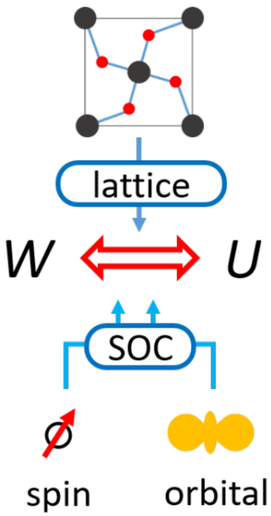


Intro.

- **Transition metal oxides**



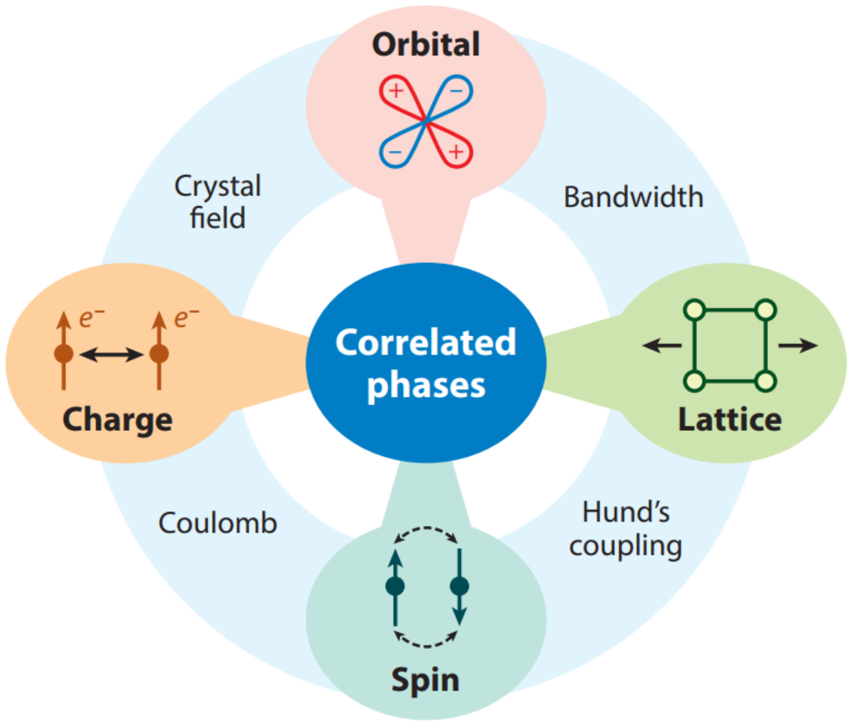
- Competition between *U* and *W*.



- Many degrees of freedoms.

Intro.

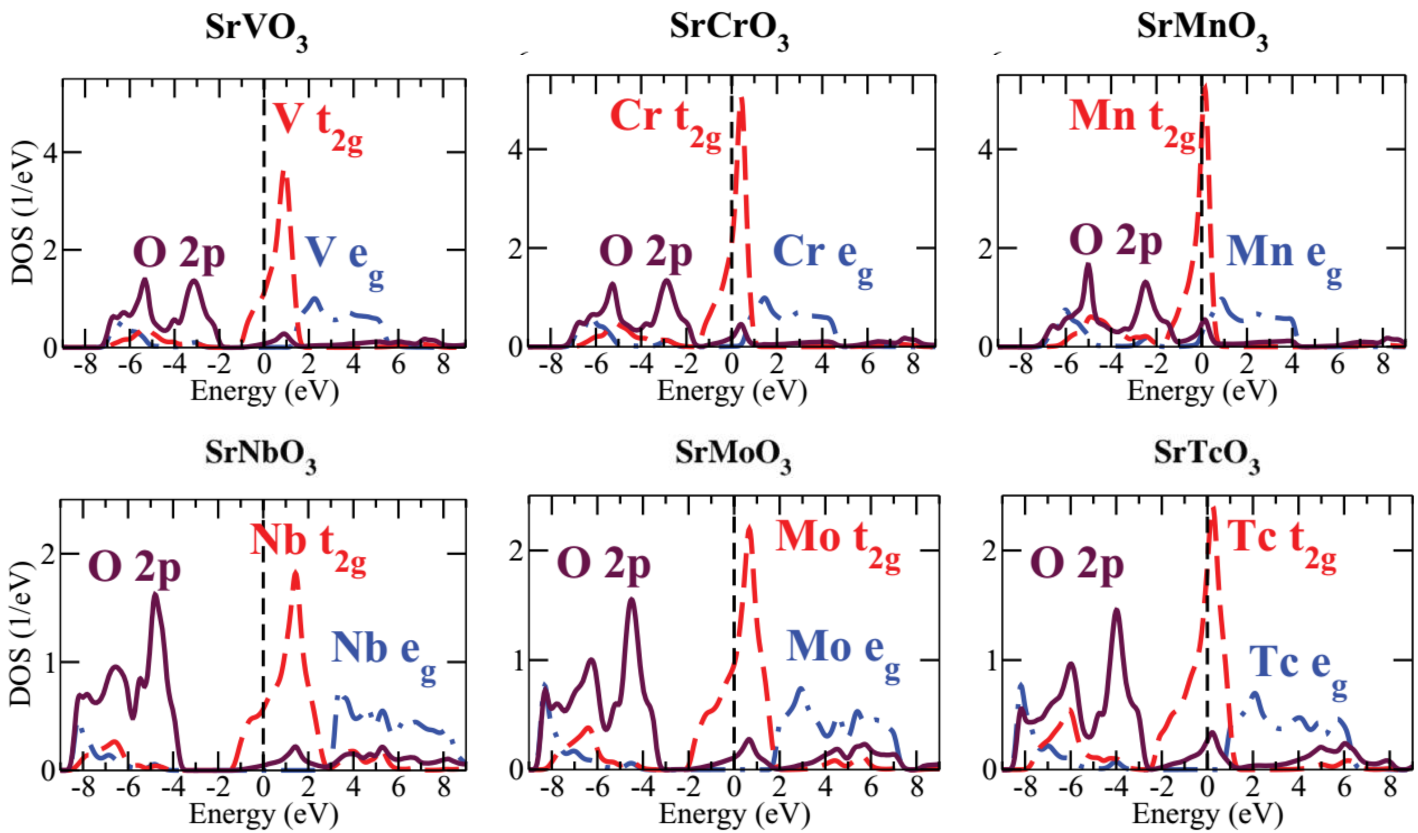
- **Transition metal oxides**



Ngai *et al.*, Annu. Rev. Mater. Res. **44** 1 (2014)

- High-T_C superconductors
- Colossal magnetoresistance (CMR)
- Unconventional magnetism
- (Multi)ferroicity
- Charge/spin/orbital order

Intro.



Vaugier *et al.*, PRB **86** 165105 (12)

- 3d oxides
  - Narrow bands
  - Strong correlation governs the physics
  - Role of SOC is minimal

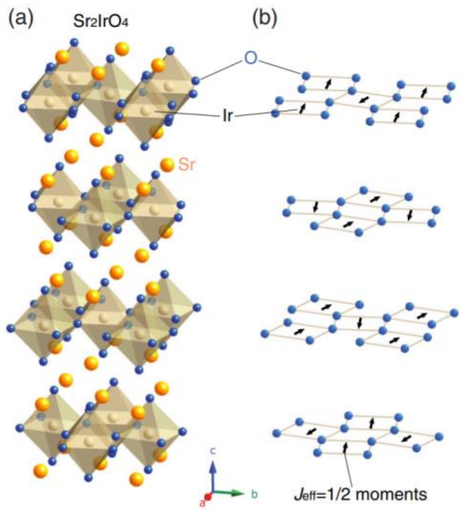
Intro.

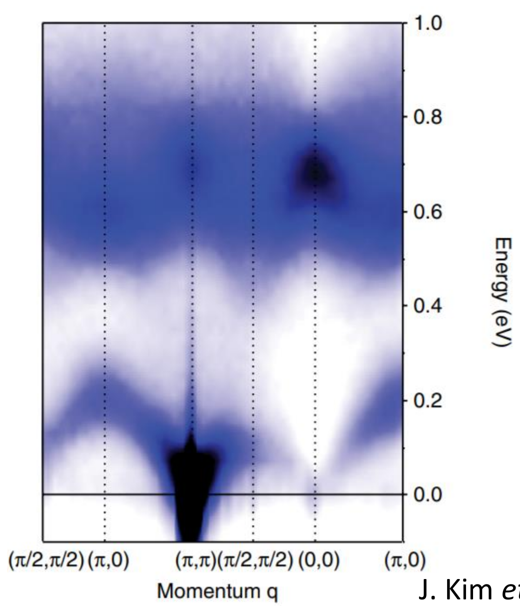
	3d oxides	5d oxides
	Cuprates, Manganites, ..	Iridates, Osmates, ...
U	5-10 eV	1-2 eV
W	a few eV	a few eV
SOC	minimal	~0.5 eV

- In 5d oxides..
  - ✓ energy scales of associated physical parameters are similar.
  - ✓ small tuning of specific parameter can change the essential physics.
  - ✓ recent experimental findings show physics in small energy scales.
  - ✓ correct quantification of parameters are important.

Intro.

- **Sr_2IrO_4: cuprate analogue**





J. Kim *et al.*, PRL **108** 177003 (12)

$$J_{ij} = \frac{t_{ij}t_{ji}}{U}$$

✓ Heisenberg interaction scale of 60-100 meV

Resolution of RIXS energy scale:
2000: 1500 meV
2010: 140 meV
2016: 8 meV

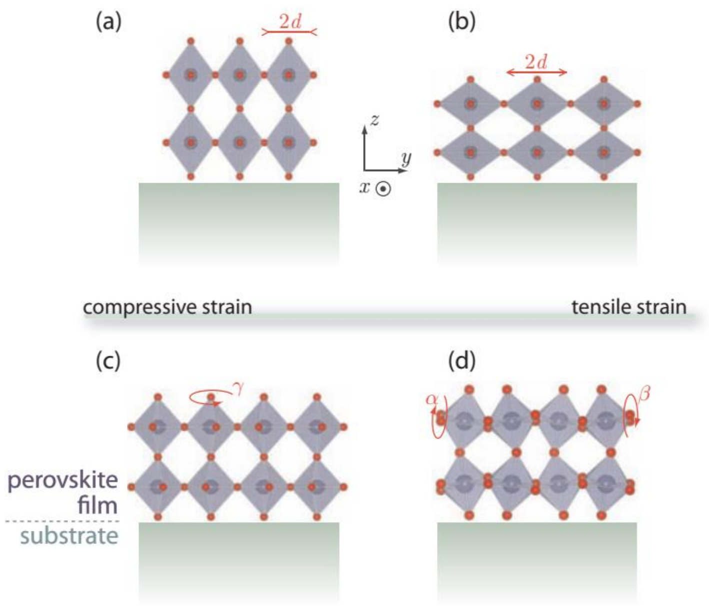
J=60 meV, *J'*= -20meV, *J''*=15meV, ..

✓ Small scale of physical parameters < eV are at play
: *U, t, J*, SOC, .. etc

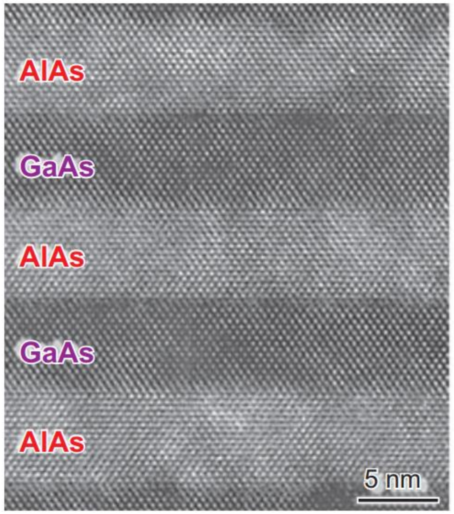
- Correct quantification of parameters are important.

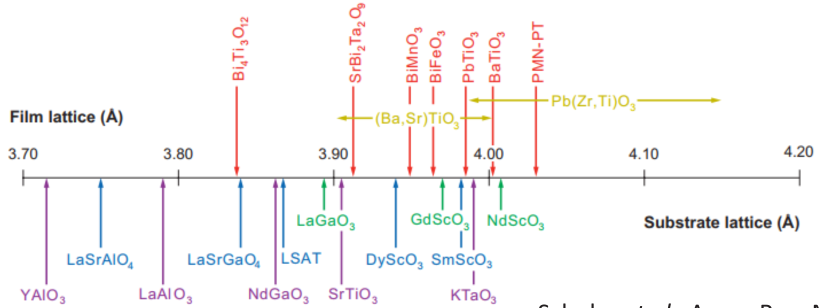
Intro.

- **Material tuning**
  - Epitaxial strain engineering
  - Heterostructuring



Rondinelli & Spaldin, Adv. Mater. **23** 3363 (2011)



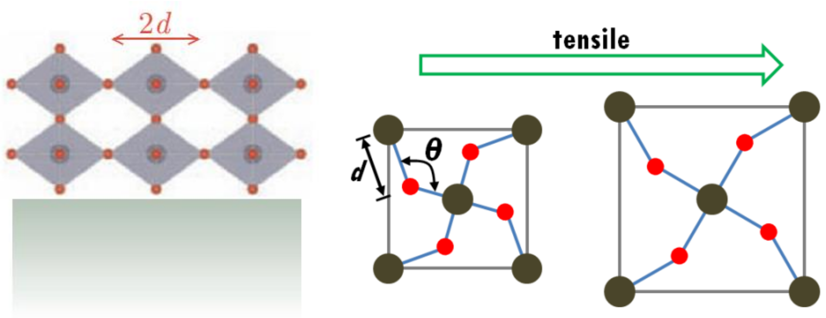


Scholm *et al.*, Annu. Rev. Mater. Res. **37** 589 (2007)

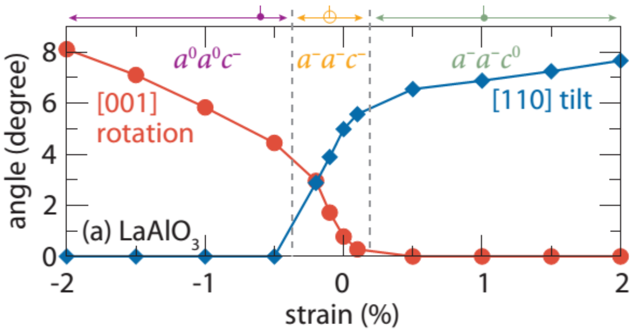
Intro.

- **Material tuning**

Strain

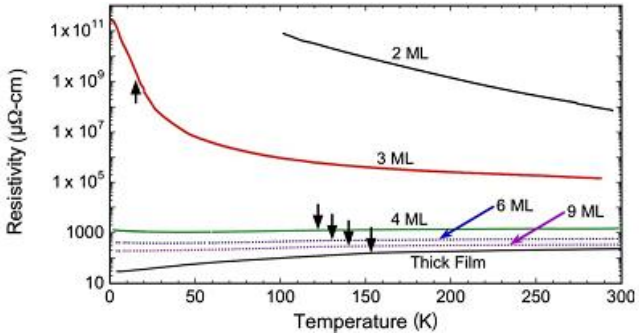


Symmetry



Rondinelli *et al.*, Adv. Mater. (2011)

Dimensionality



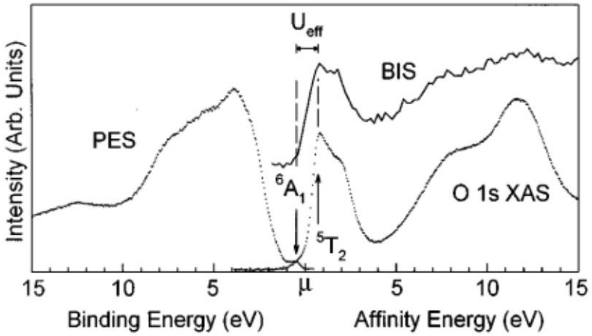
Xia *et al.*, PRB(R) **79** 040407 (2009)

- Tuning of bandwidth, hopping, crystal field splitting, symmetry, *etc*

Motivation

"what about ***U*** ?"

- In DFT-based studies, strong correlation is treated with adding Hubbard-type parameter *U*
  - DFT+*U*
  - DFT+DMFT
- The size of *U* is decided to fit experimental results
  - gap size
  - moment size
  - PES/IPES



- In practice for DFT studies, constant *U* has been employed upon structural modification.
  - strain, dimensional changes, etc.

"Is ***U*** parameter varies upon structural modification?"

Methods

- **cRPA (constrained random phase approximation)**

  - Exclude all the screening channels within the target correlated subspace (*d* orbitals for TMOs) from the total polarizability:

$$P^r = P - P^c$$

  - Partially screened Coulomb interaction can be obtained by solving following Bethe-Salpeter equation:

$$\mathbf{U}^{-1} = [\mathbf{U}^{bare}]^{-1} - P^r$$

, where the correlated subspace is constructed by means of MLWF.

  - Interaction parameters are calculated from

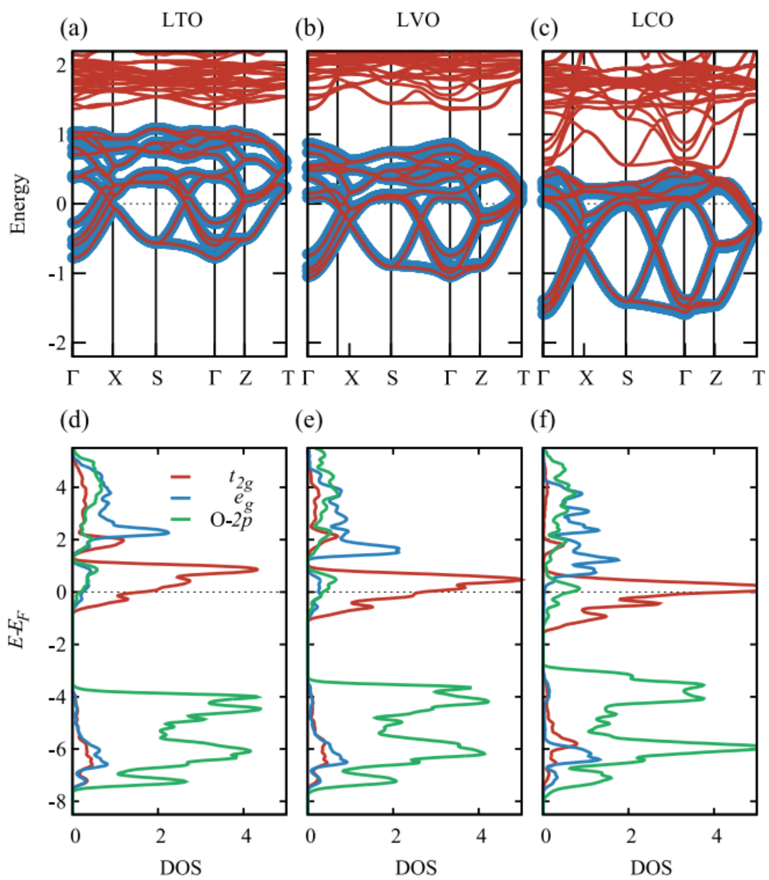
$$\mathbf{U}_{ijkl} = \lim_{\omega \to 0} \iint d^3r d^3r' w_i^*(\mathbf{r}) w_k^*(\mathbf{r}') \mathbf{U}(\mathbf{r}, \mathbf{r}', \omega) w_j(\mathbf{r}) w_l(\mathbf{r}'),$$

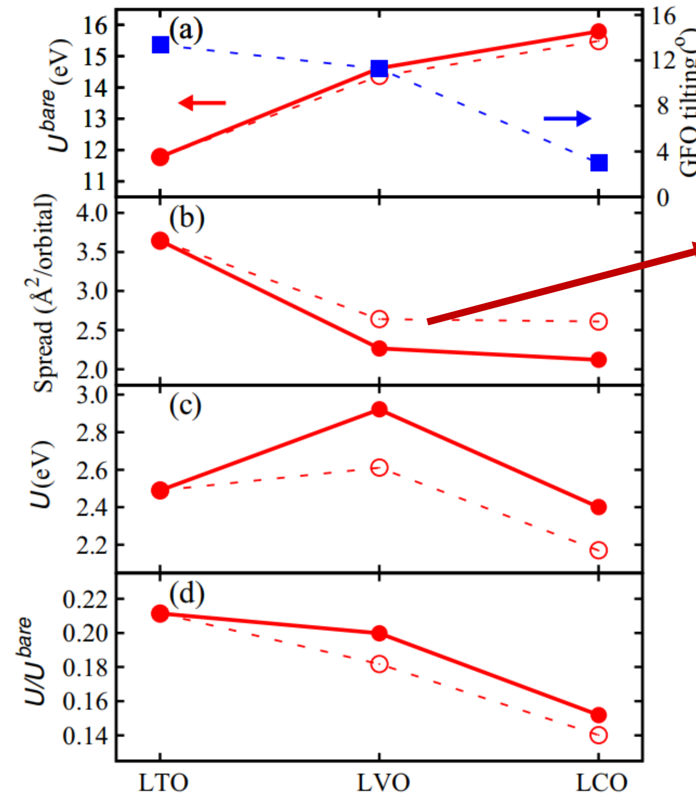
  - *U* and *J* are obtained by averaging $\mathbf{U}_{ijij}$ and $\mathbf{U}_{ijji} (i \neq j)$ matrix elements.
  - We employed both *t2g/t2g* and *t2g/t2g-p* models.

U-strain

3d oxides

BK *et al.*, PRB **98** 075130 (2018)



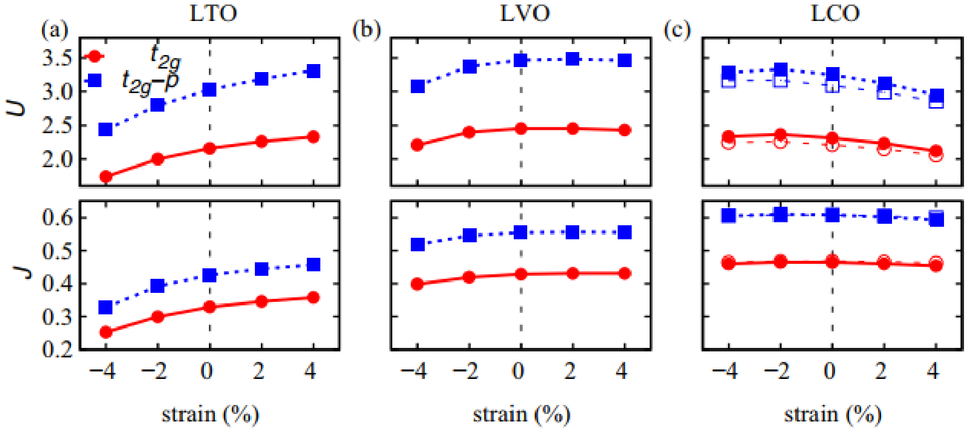


LVO/LCO in LTO structure

- Typical transition metal oxide perovskites chosen with varying occupancies:
  - $LaTiO_3$ (LTO, d^1), $LaVO_3$ (LVO, d^2) and $LaCrO_3$ (LCO, d^3)
  - t_{2g}-e_g levels are well-separated
  - *d-p* hybridization changes upon occupation
- Increase of U^{bare}
  - not from tilting but from occupation
- U does not follow trend of U^{bare}
- Strongly enhanced screening upon occupancy
- Competition between localization and screening

U-strain

BK *et al.*, PRB **98** 075130 (2018)



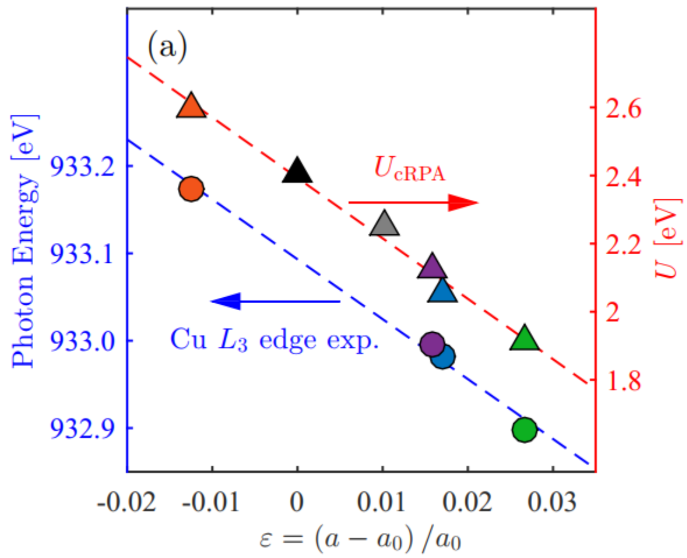
- Different strain dependence upon system.
  - LTO: *U* increase upon tensile strain
  - LCO: *U* decrease upon tensile strain

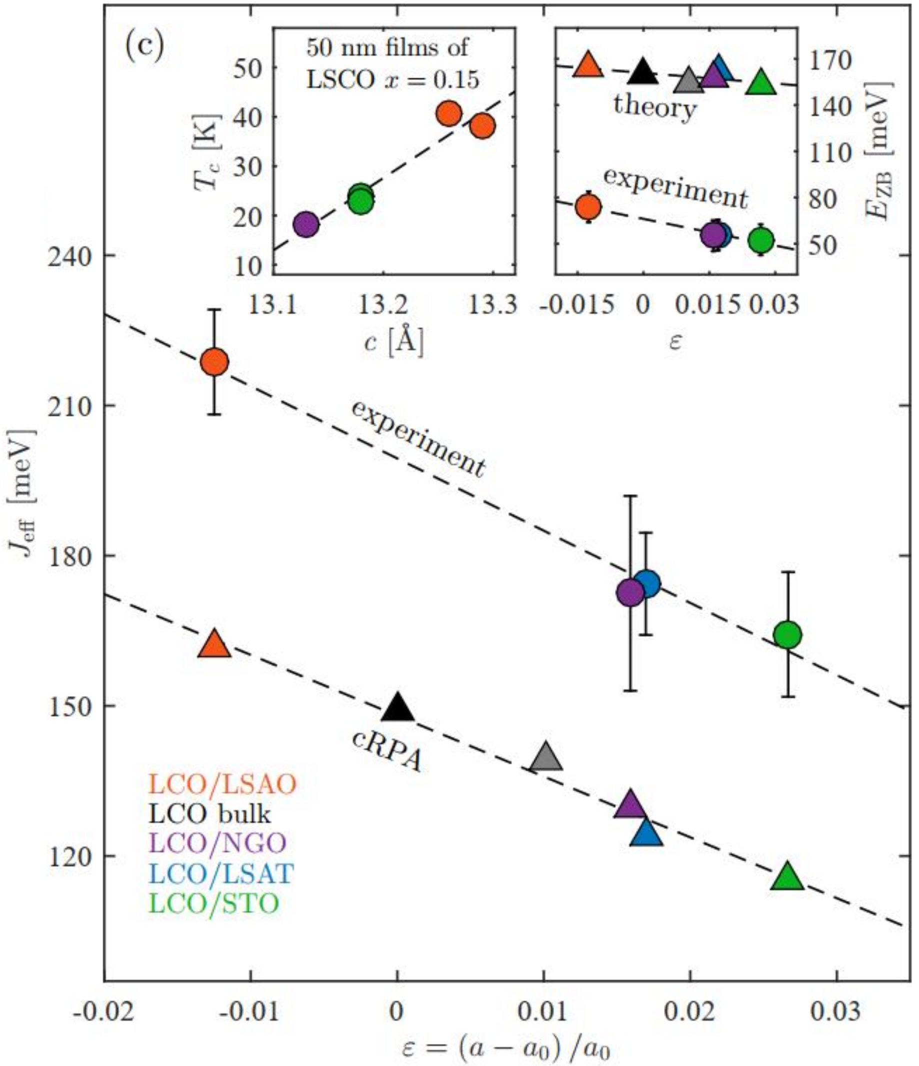
- Physics from localization, hybridization, and screening.
  - Wannier localization: effective in LTO
  - screening: more effective for LCO
  - Enhanced *d*-*p* overlap for LCO

U-strain

La2CuO4

Ivashko *et al.*, Nature Comm. 10 786 (2019)

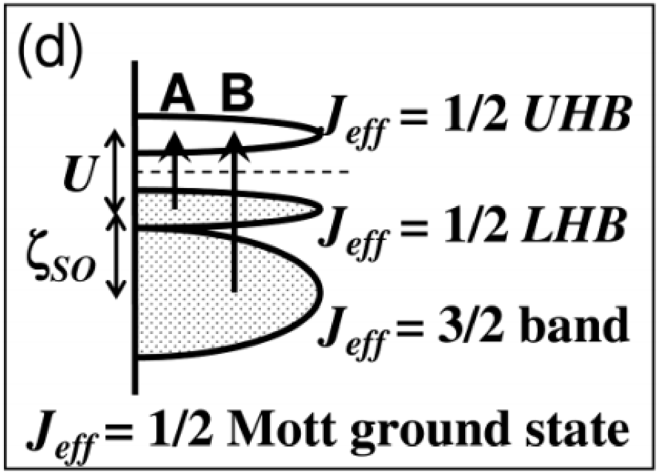




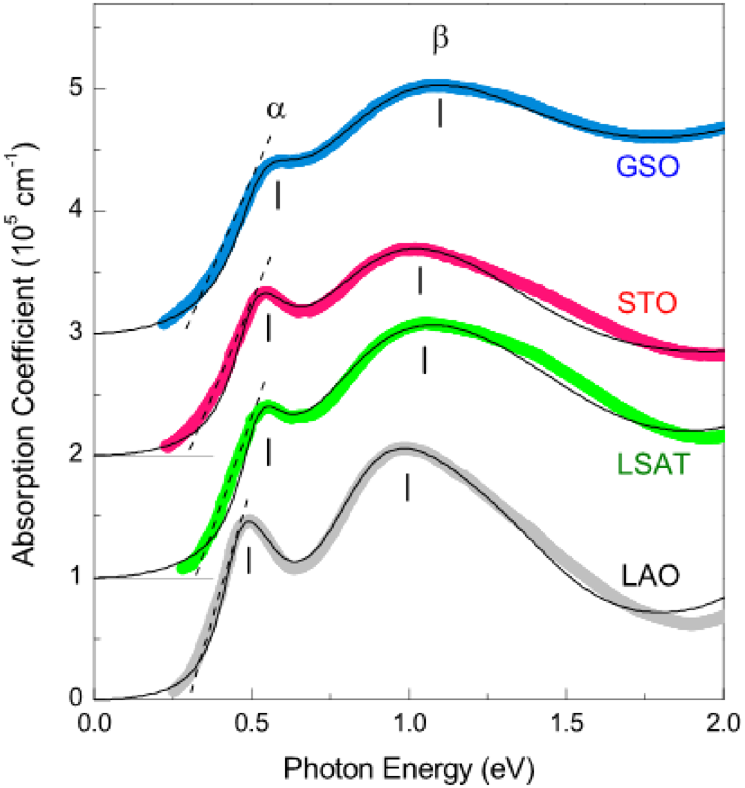
- Clear indication of change of *U* upon epitaxial strain.
- Clue to find the optimal condition for highest T_c.
  - enhancing Coulomb interaction & magnetic exchange interactions

iridates

Sr2IrO4



B.J.Kim *et al.*, PRL **101** 076402 (2008)

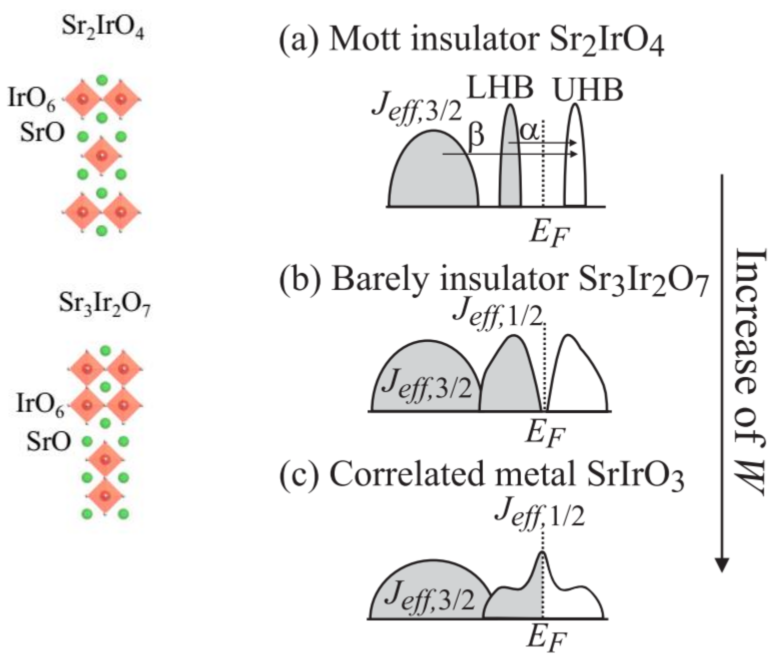


Nichols *et al.*, APL **102** 141908 (2013)

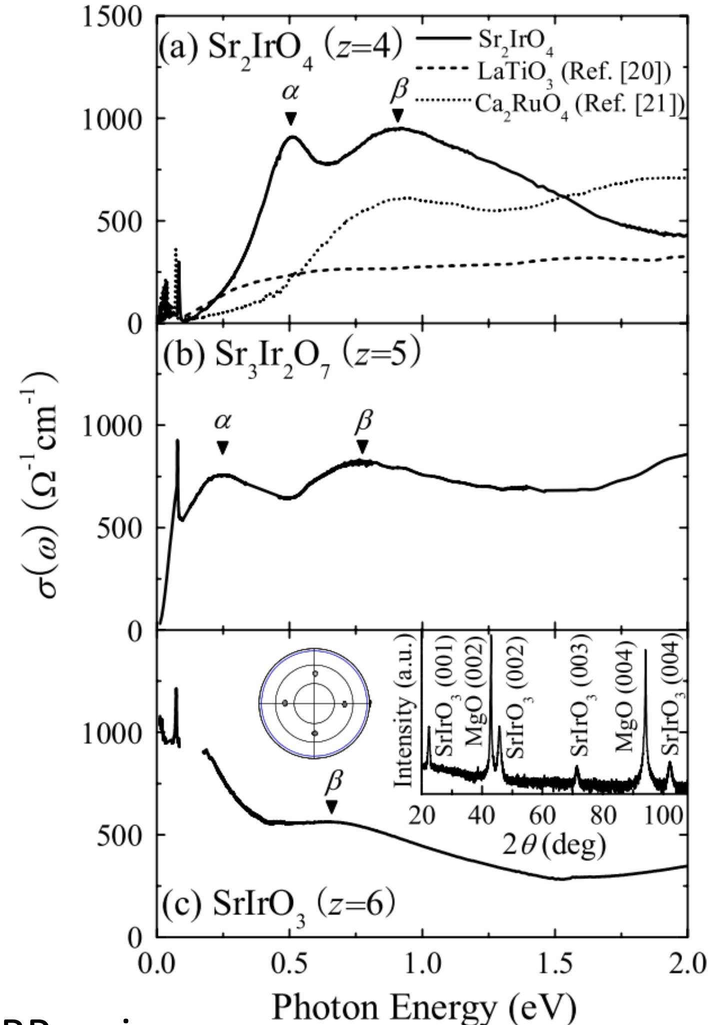
- Shift of α-peak: suggestive of *U* variations.

iridates

Ruddlesden-Popper series



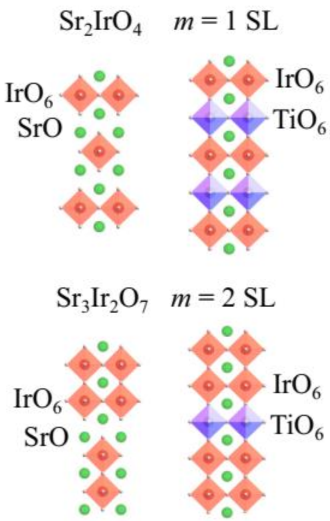
B.J.Kim *et al.*, PRL **101** 076402 (2008)

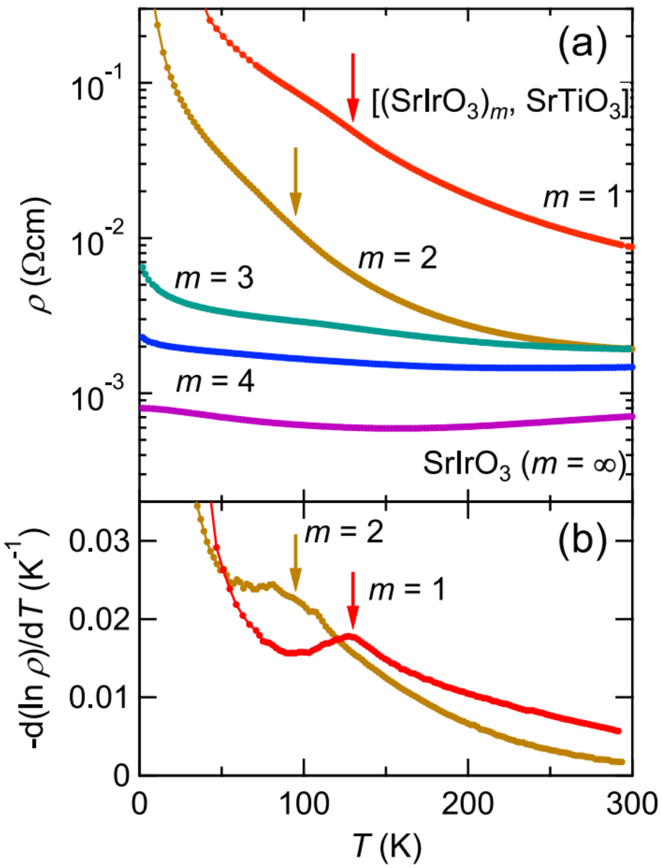


- Dimensional insulator-to-metal transition among RP series.
- Previously interpreted as increase of *W* upon dimensionality.
- Effective increase of *U* can be identified from optics.

iridates

(SrIrO3)m/(SrTiO3)



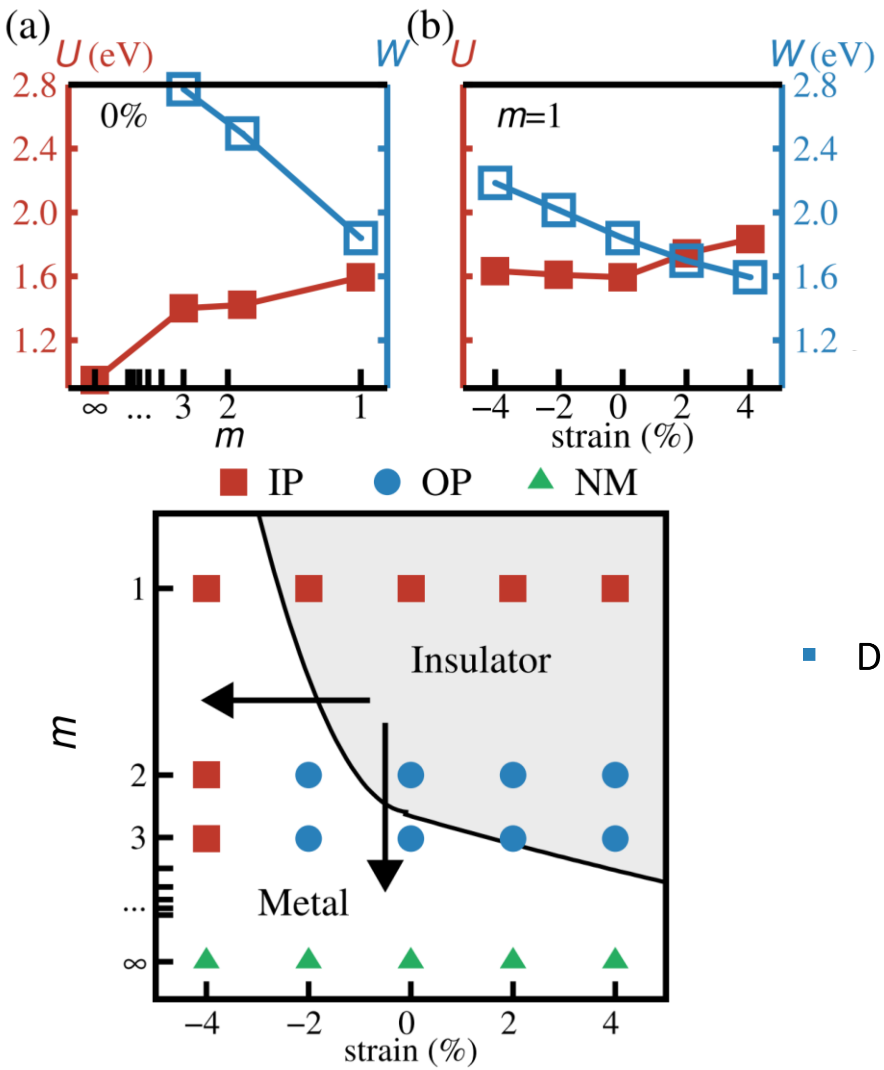


Matsuno *et al.*, PRL **114** 247209 (2015)

- Systematic study of dimensional effect using superlattice.
- Insulator-metal transition as well as magnetic transition observed.

$(SrIrO_3)_m/(SrTiO_3)$

BK *et al.*, PRB **95** 115111 (2017)

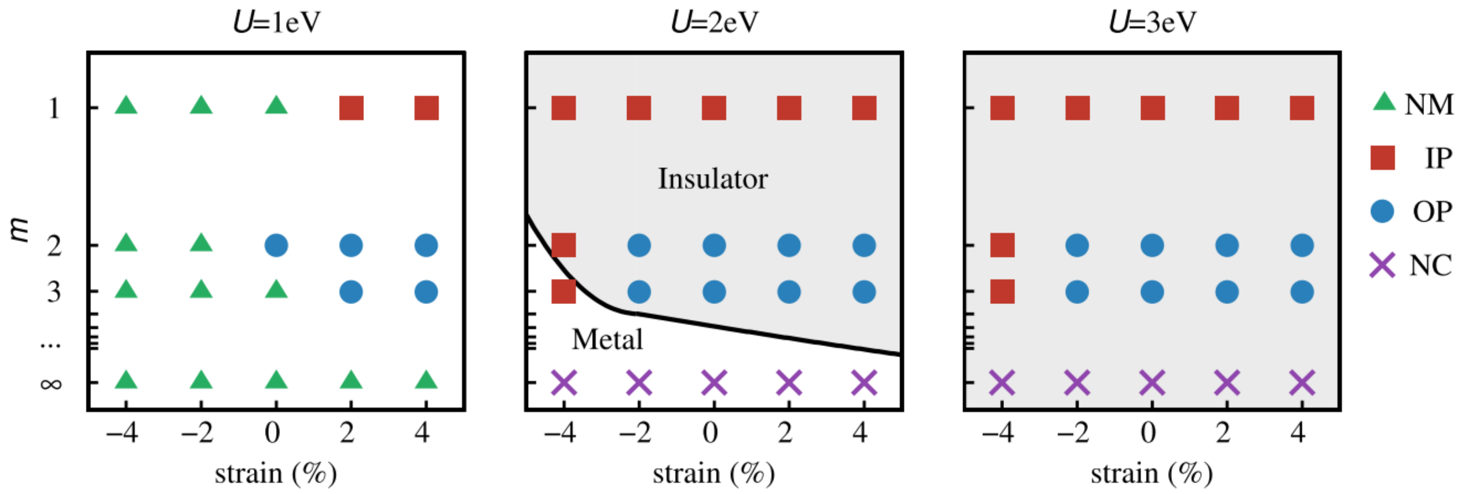


- Dimensional change of *U* & *W* can be quantified.
- Strain-dependence is not pronounced.

- Dimensionality-strain phase diagram
  - DFT+SOC+U with calculated *U* value.
  - Correct electronic and magnetic phase obtained.
  - Reproduces experimental findings.

(SrIrO3)m/(SrTiO3)

BK *et al.*, PRB **95** 115111 (2017)



- Employing the same *U* value for different structures results in wrong phase diagram.
- One need to take account of the varying *U*!

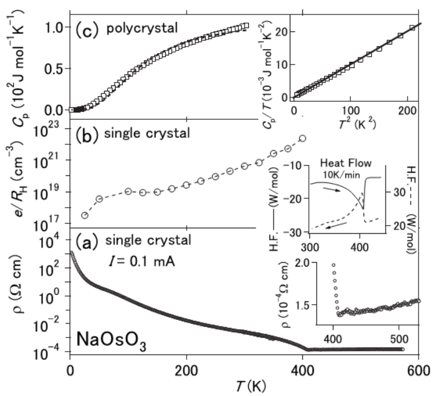
NaOsO3 and LiOsO3

RAPID COMMUNICATIONS

PHYSICAL REVIEW B **80**, 161104(R) (2009)

Continuous metal-insulator transition of the antiferromagnetic perovskite $NaOsO_3$

Y. G. Shi,[1,2] Y. F. Guo,[1,2] S. Yu,[3] M. Arai,[4] A. A. Belik,[1,2] A. Sato,[5] K. Yamaura,[2,3,*] E. Takayama-Muromachi,[1,2,3] H. F. Tian,[6] H. X. Yang,[6] J. Q. Li,[6] T. Varga,[7] J. F. Mitchell,[7] and S. Okamoto[8]



- Continuous MIT
  - Cooling and warming curve follows the same trace.
  - Continuous change in carrier density

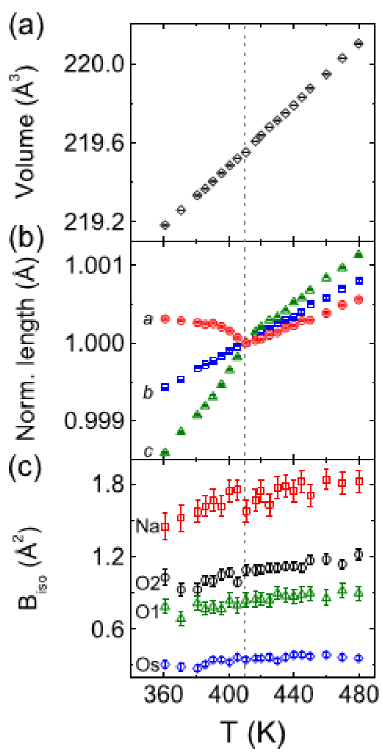
- **Ground state: G-type antiferromagnetic insulator**

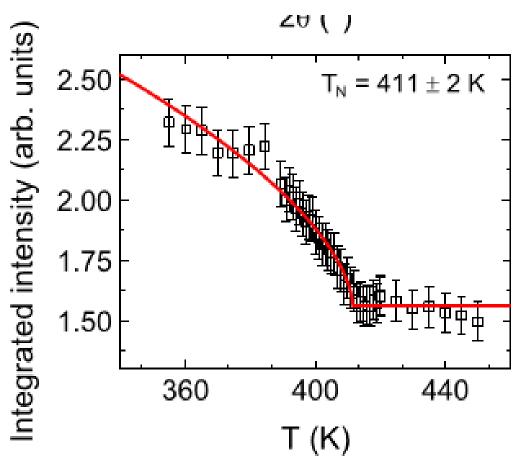
NaOsO3 and LiOsO3

PRL **108,** 257209 (2012) PHYSICAL REVIEW LETTERS week ending 22 JUNE 2012

Magnetically Driven Metal-Insulator Transition in $NaOsO_3$

S. Calder,[1,*] V. O. Garlea,[1] D. F. McMorrow,[2] M. D. Lumsden,[1] M. B. Stone,[1] J. C. Lang,[3] J.-W. Kim,[3] J. A. Schlueter,[4] Y. G. Shi,[5,6] K. Yamaura,[6] Y. S. Sun,[7] Y. Tsujimoto,[7] and A. D. Christianson[1]

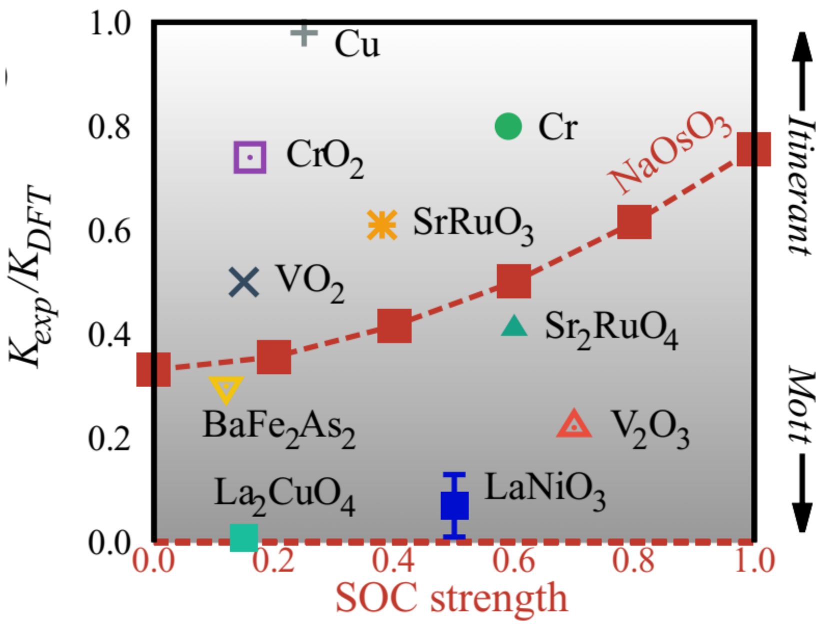


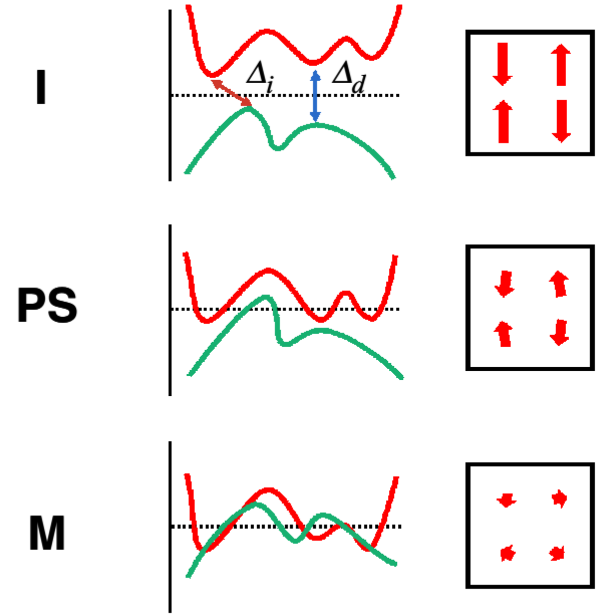


- limited structural effect.
- $T_{MIT} = T_N$.
- magnetism-driven.

NaOsO3 and LiOsO3

BK *et al.*, PRB **94** 241113R (2016)





- SOC-induced renormalization of *U*.
- T-dep. spin-fluctuation drives the MIT of the system.

NaOsO3 and **LiOsO3**

LETTERS

PUBLISHED ONLINE: 22 SEPTEMBER 2013 | DOI: 10.1038/NMAT3754

nature materials

A ferroelectric-like structural transition in a metal

Youguo Shi[1,2†], Yanfeng Guo[1,3†], Xia Wang[1], Andrew J. Princep[3], Dmitry Khalyavin[4], Pascal Manuel[4], Yuichi Michiue[5], Akira Sato[6], Kenji Tsuda[7], Shan Yu[1], Masao Arai[8], Yuichi Shirako[9], Masaki Akaogi[9], Nanlin Wang[2], Kazunari Yamaura[1*] and Andrew T. Boothroyd[3*]

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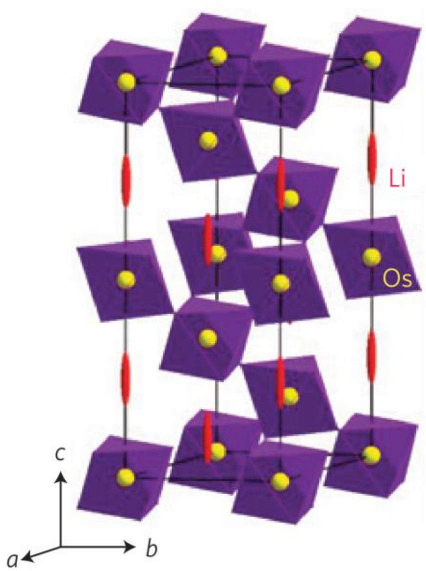


- First example of polar metal
- *R-3c* to *R3c* transition

- **Ground state: paramagnetic metal**

osmates

NaOsO3 and LiOsO3

- Low-*T*

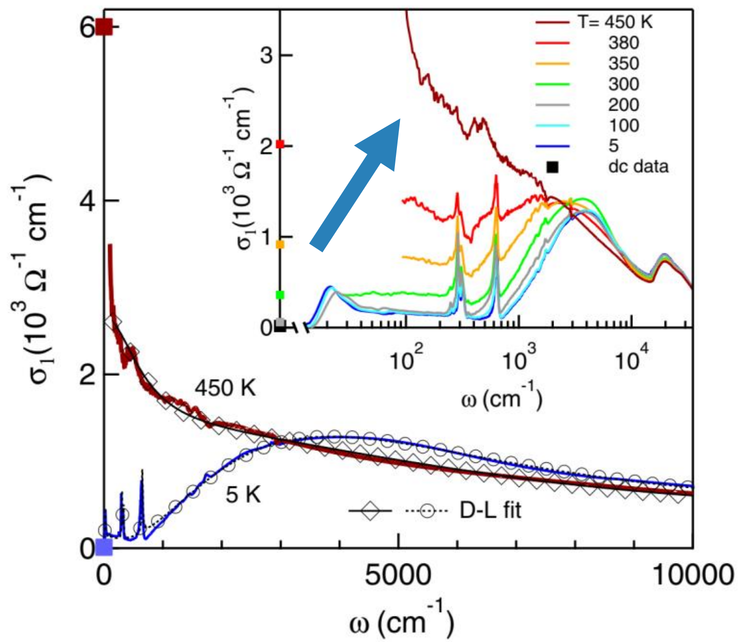
	NaOsO3	**LiOsO3**
Symmetry	*Pnma*	*R3c*
magnetism	G-AFM	paramagnetic
electronic	insulator	metal

- Similar electronic structure expected but very different ground state.

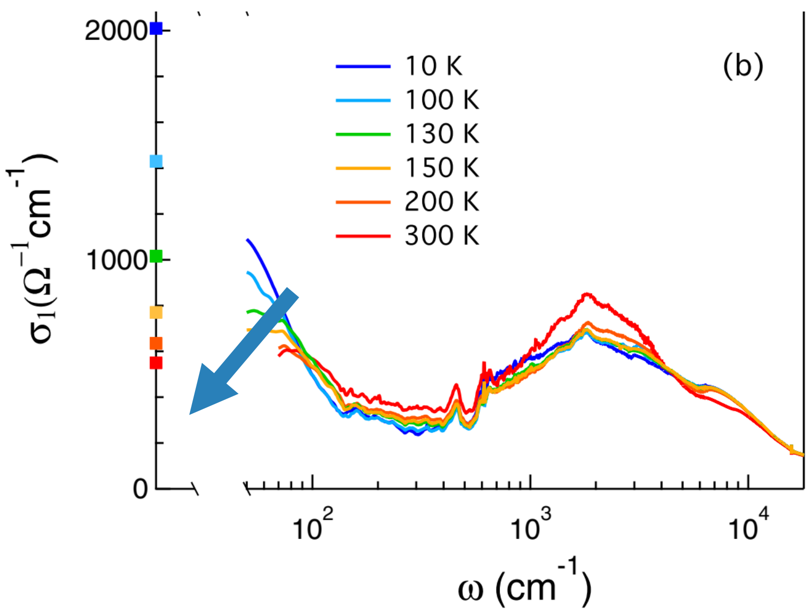
NaOsO3 and LiOsO3

- High-T: optical conductivity

Lo Vecchio *et al.*, Sci. Rep **3** 2990 (2013)



Lo Vecchio *et al.*, PRB **93** 161113R (2016)



NaOsO3

- Increase of Drude part
- Metallic behavior as T increases

LiOsO3

- Decrease of Drude part
- Insulating behavior as T decreases

osmates

NaOsO3 and LiOsO3

DFT+DMFT

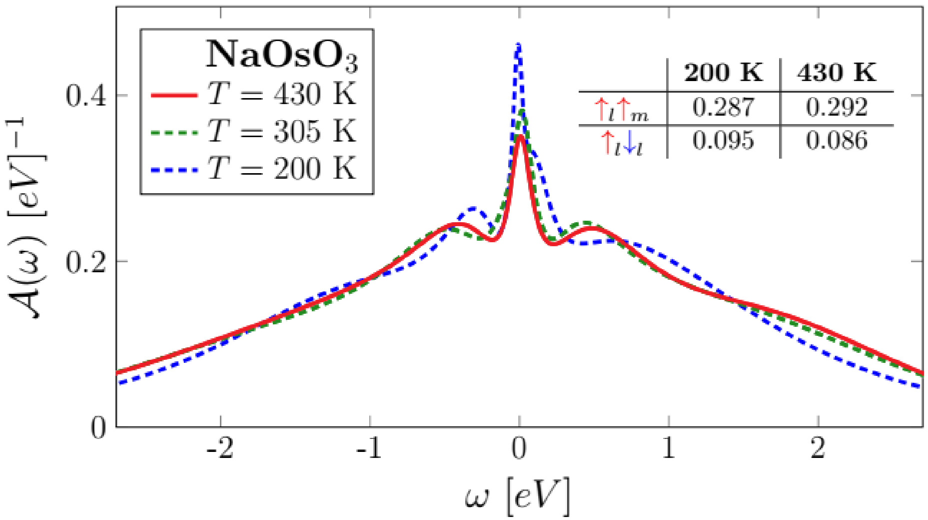
- Wannier-projected t2g orbitals.
- *w2dynamics* implementation.
- CTQMC

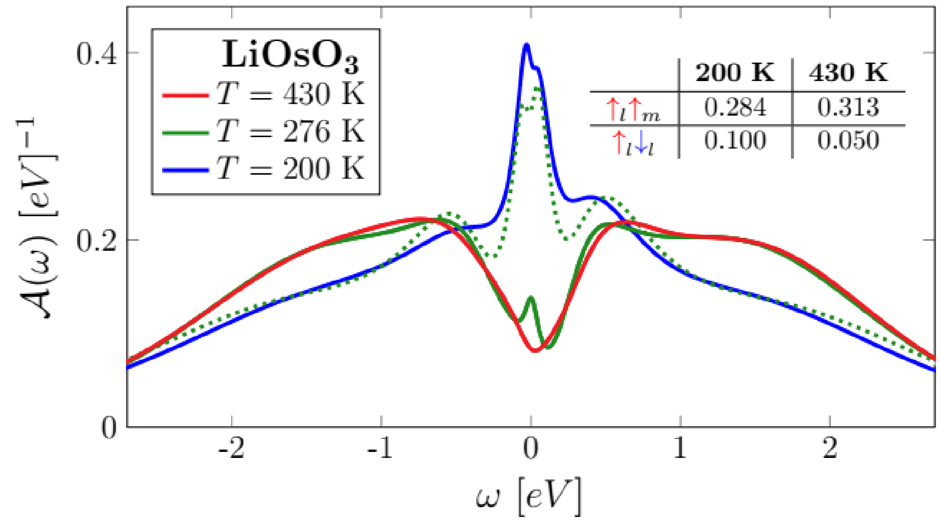
Physical parameters

	NaOsO3	LiOsO3
U	2.25 eV	2.35 eV
J	0.24 eV	0.25 eV
W	3.9 eV	3.5 eV
intra t2g splitting	150 meV	250 meV
SOC	0.3 eV	0.3 eV

On-site spectral function

NaOsO3 **LiOsO3**



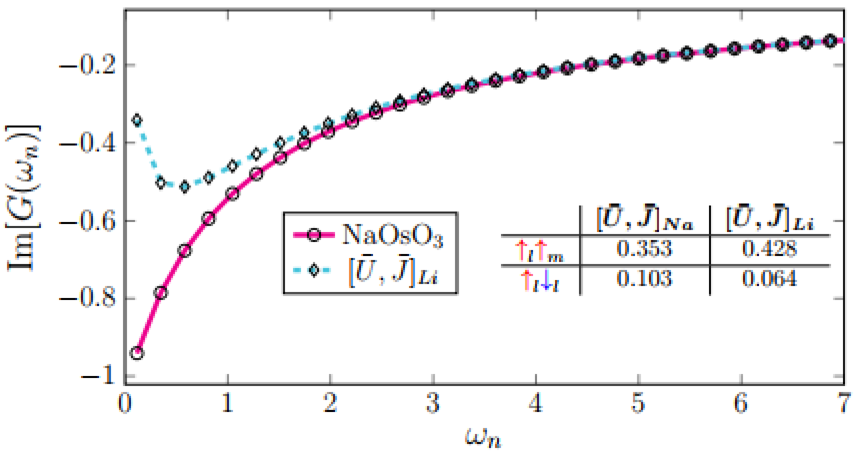


Springer, **BK,** *et al.*, arXiv:1910.05151

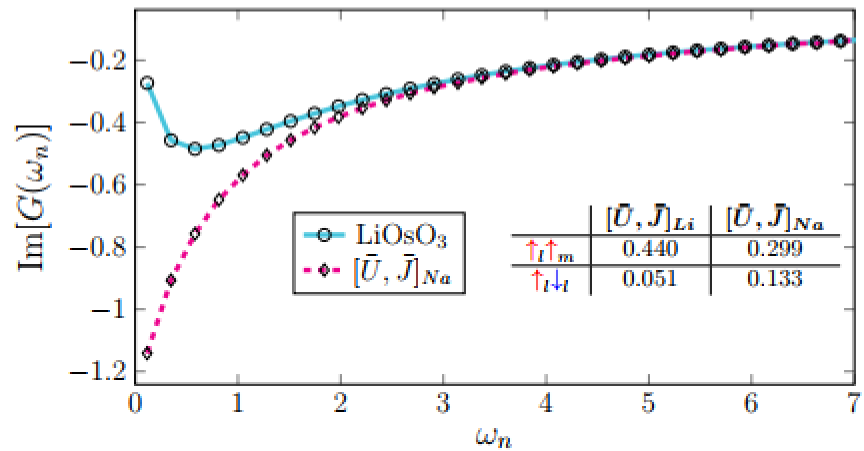
- Constructed from analytic continuation of on-site Green's function.
- Overall T-dep. behavior well-captured.
- Disappearance of metallic coherence for LiOsO3 as T increase.
- Increase of interorbital spin alignment & decrease of intraorbital double occupancy.

Local Green's function

NaOsO3

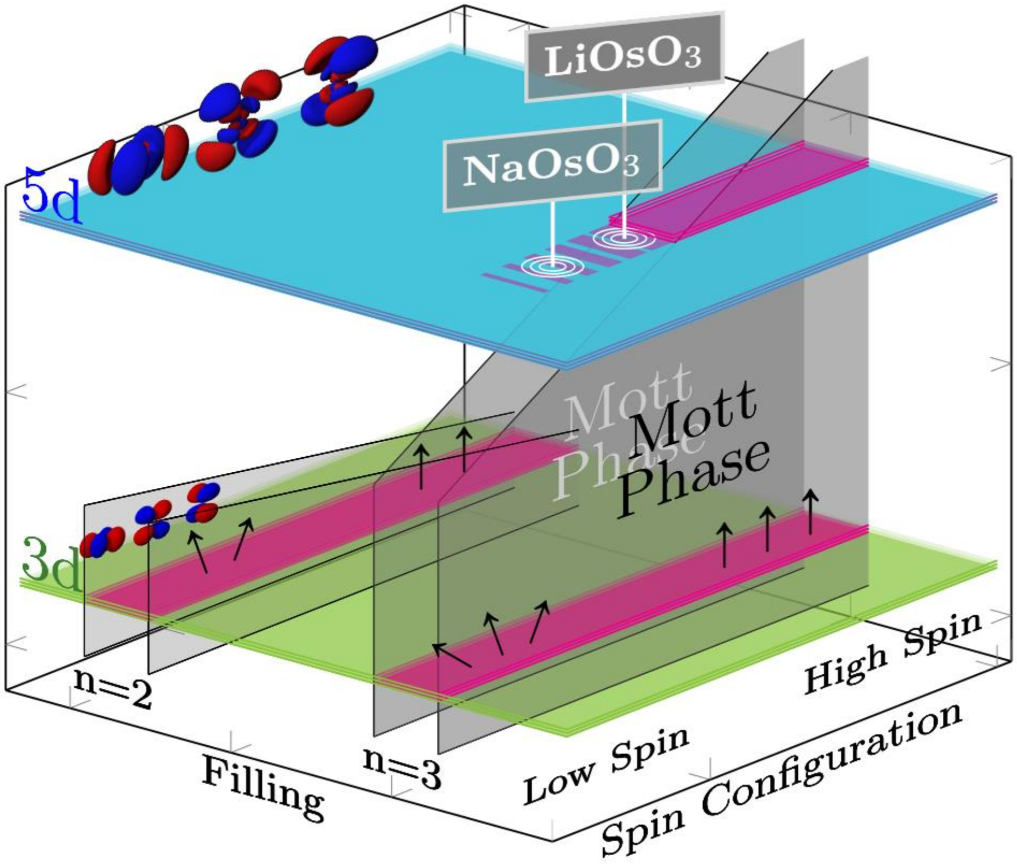


LiOsO3



- At high-*T*, metallic and insulating properties of NaOsO3 and LiOsO3 are well-described.
- Hund-Mott nature of insulating phase in LiOsO3.
  - Enhanced interorbital spin alignment: large local moment.
  - Suppression of intraorbital occupation.
- Interchange of Coulomb parameters, U & J, produces perfect mirroring of Green's function.
- Small details in the interaction parameters are responsible for the stark difference in two systems.

osmates



- Two systems are located at the proximity to the *n*=3 Hund-Mott transition.
- In 5d systems, better description of related physical parameters are important.

Summary

- *U* is not a fixed parameter – strongly dependent on the electronic structure and structural details.

- When employing DFT+manybody approaches (+U, +DMFT, etc), better quantification of Coulomb parameters are important – especially for 5d oxides.

- Implications for the TMO-based materials design.

BK, et. al, PRB **94** 241113 (2016)
BK, et. al ,PRB **95** 115111 (2017)
BK, et. al, PRB **98** 075130 (2018)
Springer, **BK**, et. al, arXiv: 1910.05151
Liu, He, **BK**, et. al, to be submitted

Thank you