Physics of lower dimensional systems: examples of research made possible through collaborations

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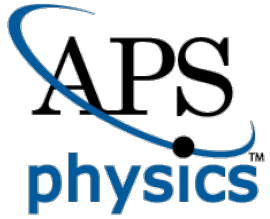

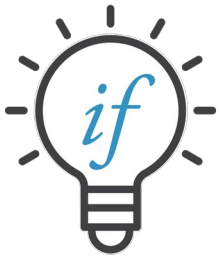

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Talk Outline:

- Low temperature phase of perovskite (ABO_3) materials: *structural, electronic and vibrational properties using extended Hubbard functional*

  Collaboration with Iurii Timrov and Nicola Marzari, EPFL, Switzerland

- Strontium ruthenate oxides of the Ruddlesden Popper (RP) series, $Sr_{n+1}Ru_nO_{3n+1}$ (n =1, 2, 3 , ∞): *Electronic and magnetic properties using PBEsol and Hubbard functionals*

  Collaboration with Richard Martin, Nicola Seriani, Prosper Ngabonziza, Omololu Akin-Ojo, Jonah Nagura

- Ideas for Collaborations

Low Temperature Phase ABO_3 Perovskites using Extended Hubbard functional

Outline

- Structural optimization
- Electronic properties
- Phonon calculations

Collaboration:

- **Prof. Nicola Marzari &**
- **Dr. Iurii Timrov**
- **http://theossrv1.epfl.ch/**

Low temperature phase Barium Titanite ($BaTiO_3$)

Barium Titanite is one of the ABO_3 perovskite materials.

The atomic positions can be represented:

Ba: $(0.0 + \Delta_{Ba} , 0.0 + \Delta_{Ba} , 0.0 + \Delta_{Ba})$

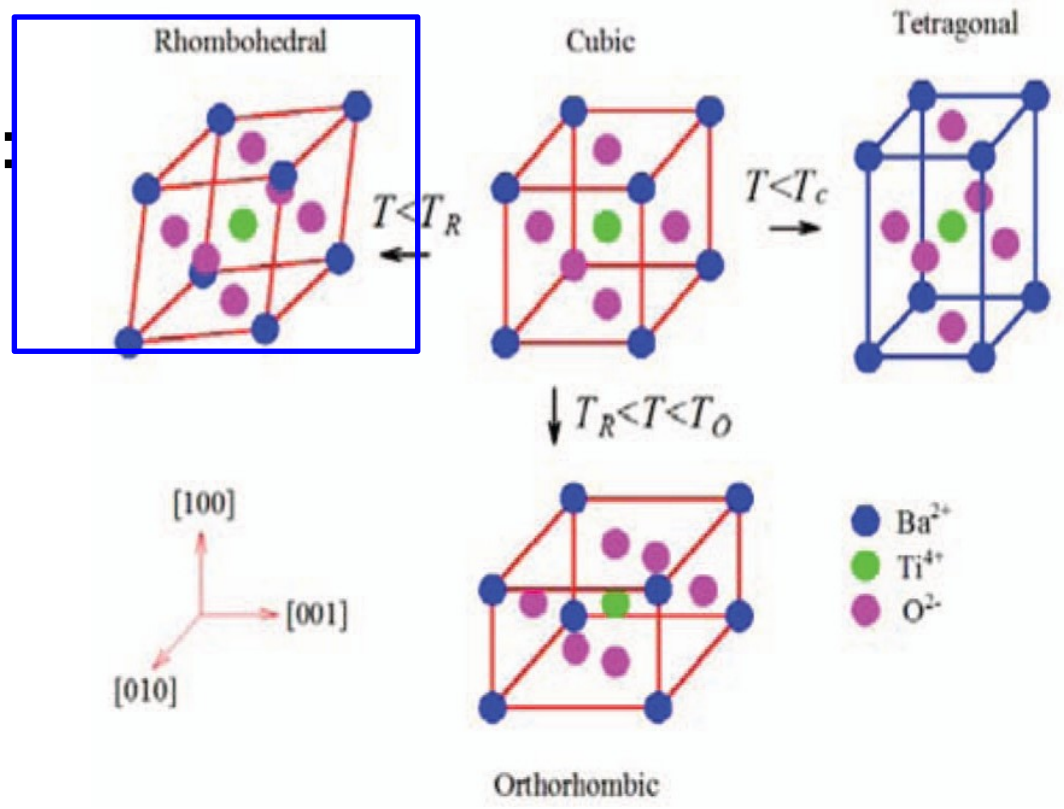
Ti: $(0.5 + \Delta_{Ti} , 0.5 + \Delta_{Ti} , 0.5 + \Delta_{Ti})$

O1: $(0.5 + \Delta_{O1}, 0.5 + \Delta_{O1} , 0.0 + \Delta_{O2})$

O2: $(0.5 + \Delta_{O1} , 0.0 + \Delta_{O2} , 0.5 + \Delta_{O1})$

O3: $(0.0 + \Delta_{O2} , 0.5 + \Delta_{O1} , 0.5 + \Delta_{O1})$

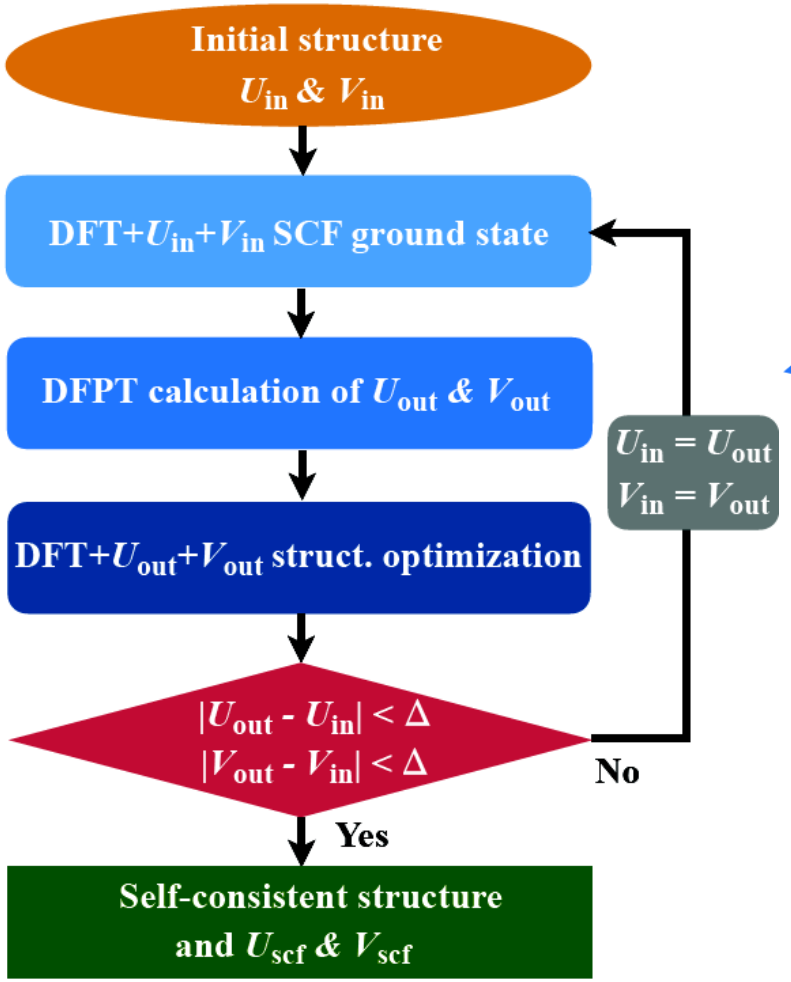
Where Δ_{Ba}, Δ_{Ti}, $\Delta_{O1,}$ and Δ_{O2} are atomic distortions.



We consider rhombohedral crystal structure with space group of *R3m* (160) (point group C_{3v}) and lattice Parameters a = b = c = 4.0036 Å and angle $\alpha = \beta = \gamma = 89.839^0$

Kwei G H *et. al* J.Phys. Chem. **97**, 2368, (1993).

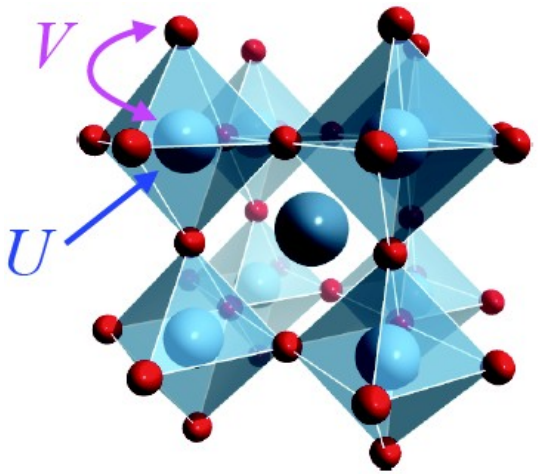
Self-consistent workflow of DFT+Hubbard



Calcultion of Hubbard parameters

$$U^I = \left(\frac{d^2 E_{\mathrm{DFT}}}{d\mathbf{n}^2} - \frac{d^2 E^{\circ}_{\mathrm{DFT}}}{d\mathbf{n}^2}\right)_{II}$$

$$V^{IJ} = \left(\frac{d^2 E_{\mathrm{DFT}}}{d\mathbf{n}^2} - \frac{d^2 E^{\circ}_{\mathrm{DFT}}}{d\mathbf{n}^2}\right)_{IJ}$$



atomic localisation & inter-atomic hybridisations

I. Timrov, N. Marzari, and M. Cococcioni, Phys. Rev. B **103**, 045141 (2021).

Structural Optimization

- DFT based structural optimization: PBEsol functional

	this work PBEsol				other theoretical works			Exp. [1]
Parameter	VC-R	VC-R-Ba-F	VC-R-vol-F	VC-R-vol-F-Ba-F	LDA (Ba-F)[2]	LDA-Vol-F (Ba-F)[3]	GGA-PBE (Ba-F)[2]	
a(A)	3.9949	3.9949	4.0036	4.0036	3.966	4.001	4.073	4.0036
α	89.854	89.854	89.834	89.835	89.958	89.87	89.710	89.839
Δ_{Ba}	-0.0006	0.0000	-0.001	0.0000	0.0000	0.000	0.0000	0.0000
Δ_{Ti}	-0.0123	-0.0117	-0.0132	-0.0122	-0.0080	-0.011	-0.0150	-0.0128
Δ_{O1}	0.0114	0.0119	0.0116	0.0126	0.0069	0.0133	0.0143	0.0109
Δ_{O2}	0.0184	0.0189	0.0193	0.0203	0.0109	0.0192	0.0249	0.0193

The deviations of optimized lattice constant and rhombohedral angle from the experimental values are 0.22 % and 0.017%, respectively.

1.Robert A. Evarestov and Andrei V. Bandura, J. Comp. Chem, **33**, 1123–1130, (2012).
2.Ph. Ghosez, X. Gonze, J. -P. Michenaud, Ferroelectrics **1**, 220, (1999).
3.Exp.- Kwei G H, Lawson A C, Billinge S J L and Cheong S W J.Phys. Chem. **97**, 2368, (1993).

Structural Optimization

- DFT+U structural optimization: PBEsol+U functional

	PBEsol+U (atomic)				PBEsol+U (ortho-atomic)				Exp. [1]
Parameter	VC-R	VC-R-Ba-F	VC-R-vol-F	VC-R-vol-F-Ba-F	VC-R	VC-R-Ba-F	VC-vol-F	VC-vol-Ba-F	
a(A)	3.9953	3.9953	4.0036	4.0036	3.999	3.990	4.0036	4.0036	4.0036
α	89.9994	90.000	89.9997	89.9994	89.9994	89.9999	89.9999	90.000	89.839
Δ_{Ba}	0.0057	0.0000	0.0057	0.0000	0.0057	0.000	0.0056	0.0000	0.0000
Δ_{Ti}	0.0056	0.0000	0.0058	0.0000	0.0057	0.0000	0.0056	0.0000	-0.0128
Δ_{O1}	0.0057	0.0000	0.0056	0.0000	0.0057	0.0000	0.0057	0.0000	0.0109
Δ_{O2}	0.0057	0.0000	0.0055	0.0000	0.0057	0.0000	0.0057	0.0000	0.0193

The structural optimization with the inclusion of **onsite Hubbard U parameter** leads to a phase transition from the **rhombohedral to cubic crystal structure**.

Structural Optimization

DFT+U+V structural optimization: PBEsol+U+V functional

	PBEsol+U+V (atomic)				PBEsol+U+V (ortho-atomic)				Other works	Exp. [1]
Parameter	VC-R	VC-R-Ba-F	VC-R-vol-F	VC-R-vol-F-Ba-F	VC-R	VC-R-Ba-F	VC-vol-F	VC-vol-F-Ba-F	PBE0[30]	
a(A)	4.03043	4.03040	4.0037	4.0037	4.02239	4.02235	4.0036	4.0036	4.029	4.0036
α	89.5489	89.5494	89.6459	89.6461	89.7035	89.704	89.7611	89.7609	89.727	89.839
Δ_{Ba}	-0.0032	0.0000	-0.0019	0.0000	-0.0022	0.000	-0.0012	0.0000	0.0000	0.0000
Δ_{Ti}	-0.0204	-0.0173	-0.0181	-0.0162	-0.0158	-0.0137	-0.0140	-0.0128	-0.0151	-0.0128
Δ_{O1}	0.0129	0.0161	0.0124	0.0000	0.0125	0.0146	0.0120	0.0132	0.0129	0.0109
Δ_{O2}	0.0259	0.0263	0.0235	0.0253	0.0212	0.0234	0.0195	0.0207	0.0242	0.0193
U	4.9407	4.9407	4.9512	4.9513	6.5237	6.5238	6.5369	6.5369	-	-
V	1.3367-1.8819	1.3368-1.8818	1.3808-1.8541	1.3809-1.8540	1.3194 - 1.7622	1.3196 - 1.7621	1.3510-1.7448	1.3509 - 1.7448	-	-

The deviations of optimized lattice constant and rhombohedral angle from the experimental values are 0.47% and 0.15%, respectively.

1.Robert A. Evarestov and Andrei V. Bandura, J. Comp. Chem, **33**, 1123–1130, (2012).
2.Exp.- Kwei G H, Lawson A C, Billinge S J L and Cheong S W, J.Phys. Chem. **97**, 2368, (1993).

Electronic properties

The calculated band gap values using PBEsol, PBEsol+U, PBEsol+U+V, and existing previous hybrid functional based calculations.

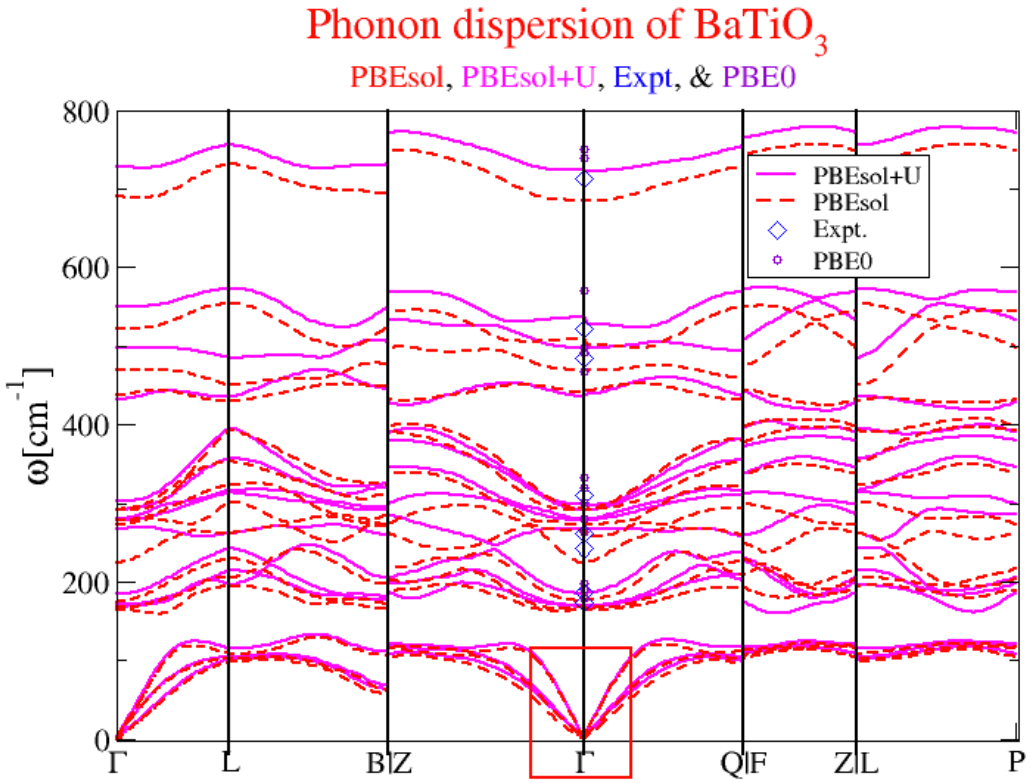
Band gap, E_g [eV]	**Ref.**
2.33	PBEsol (present work)
2.92	PBEsol+U (present work) U=5.3974 eV
3.81	PBEsol+U+V (present work) U=6.5369 eV & V= [1.35 – 1.74 eV]
3.72	HSE06 [1]
4.9	PBE0 [2]
3.4	Exp. (tetragonal) [3]

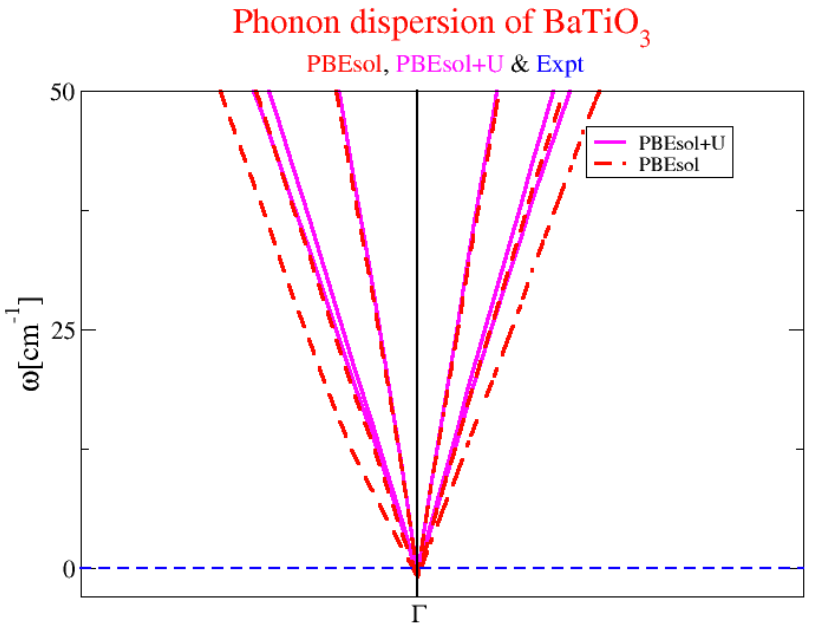
The inclusion of onsite and inter-site Hubbard U&V parameters predicts the band gap with higher accuracy than the hybrid functional calculations.

1. Wang, Grinberg, and Rappe Appl. Phys. Lett. **104**, 152903 (2014).
2. Robert A. Evarestov and Andrei V. Bandura, J. Comp. Chem, **33**, 1123–1130, (2012).
3. S. H. Wemple, Phys. Rev. B, **2**, 2679, (1970).

Phonon calculations

DFT+U based phonon calculations: PBEsol+U functional





Exp. data: D. A. TENNE *et al*. Phys. RevB. **69**, 174101, 2004

Miquel Royo, Konstanze R, Hahn, and Massimiliano Stengel, Phys Rev Lett **125,**217602 (2020).

Strontium ruthenate oxides RP-series, $Sr_{n+1}Ru_nO_{3n+1}$ (n =1, 2, 3 , ∞)

Outline

- Structural Properties
- Electronic properties
  - Band structures
  - Fermi Surfaces

Collaboration:

Prof. Richard Martin, University of Illinois, USA

Dr. Nicola Seriani, ICTP, Trieste, Italy

Dr. Omololu Akin-Ojo, EAIFR, Rwanda

Dr. Prosper Ngabonziza, Max Planck Institute for Solid State Research, Stuttgart, Germany

Jonah Nagura, ICTP, Trieste, Italy

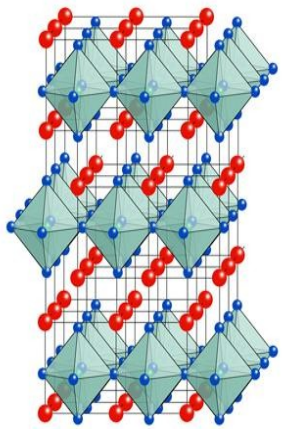
Prosper Ngabonziza, E Carleschi, V Zabolotnyy, A Taleb-Ibrahimi, F Bertran, R Fittipaldi, V Granata, M Cuoco, A Vecchione, BP Doyle, Scientific Reports, **10**, 21062, (2020).

Strontium ruthenate oxides RP-series, $Sr_{n+1}Ru_nO_{3n+1}$ (n =1, 2, 3 , ∞)

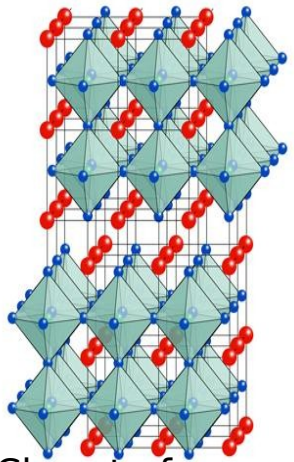
$Sr_{n+1}Ru_nO_{3n+1}$

$n=1$ $n=2$ $n=3$ $n=\infty$

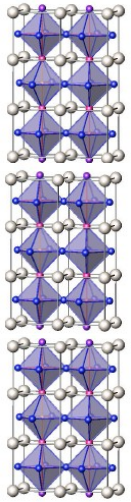
Sr_2RuO_4 $Sr_3Ru_2O_7$ $Sr_4Ru_3O_{10}$ $SrRuO_3$



Spin-triplet superconductor (T_c=1.5K)



Close to ferromagnetism, metamagnetic transition QCEP



Ferromagnetic
- T_c = 160 K.

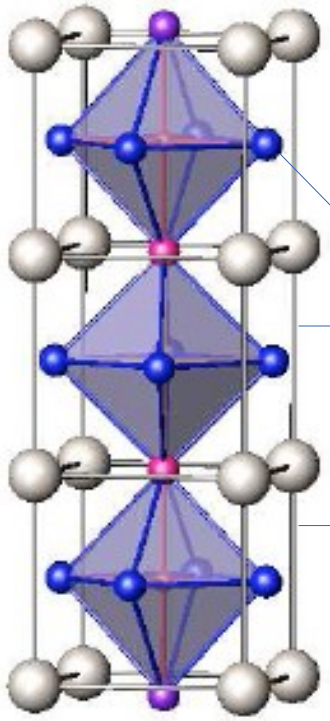
S. A Grigera *et al*., Science **294 ,** 329(2001).

K. Ishida *et al.*, Nature **396** (1998).

Y. J. Jo *et al.*, Phys. Rev. B **75**, 0944413(2007)

Structural Optimization

we consider orthorhombic crystal structure of $Sr_4Ru_3O_{10}$ with space group *Pbam* and lattice parameters a = 5.528 Å b=5.526 Å & c=28.651 Å



Optimized Structure

- **RuO_6** octahedra rotate 10.6° around the c-axis in the inner layer. → 11.339°
- **RuO_6** octahedra rotate 5.25° around the c-axis in the outer two layers. → 8.883°

a = 5.528 Å b=5.526 Å & c=28.651 Å → a = 5.474 Å b=5.473 Å & c= 29.038 Å

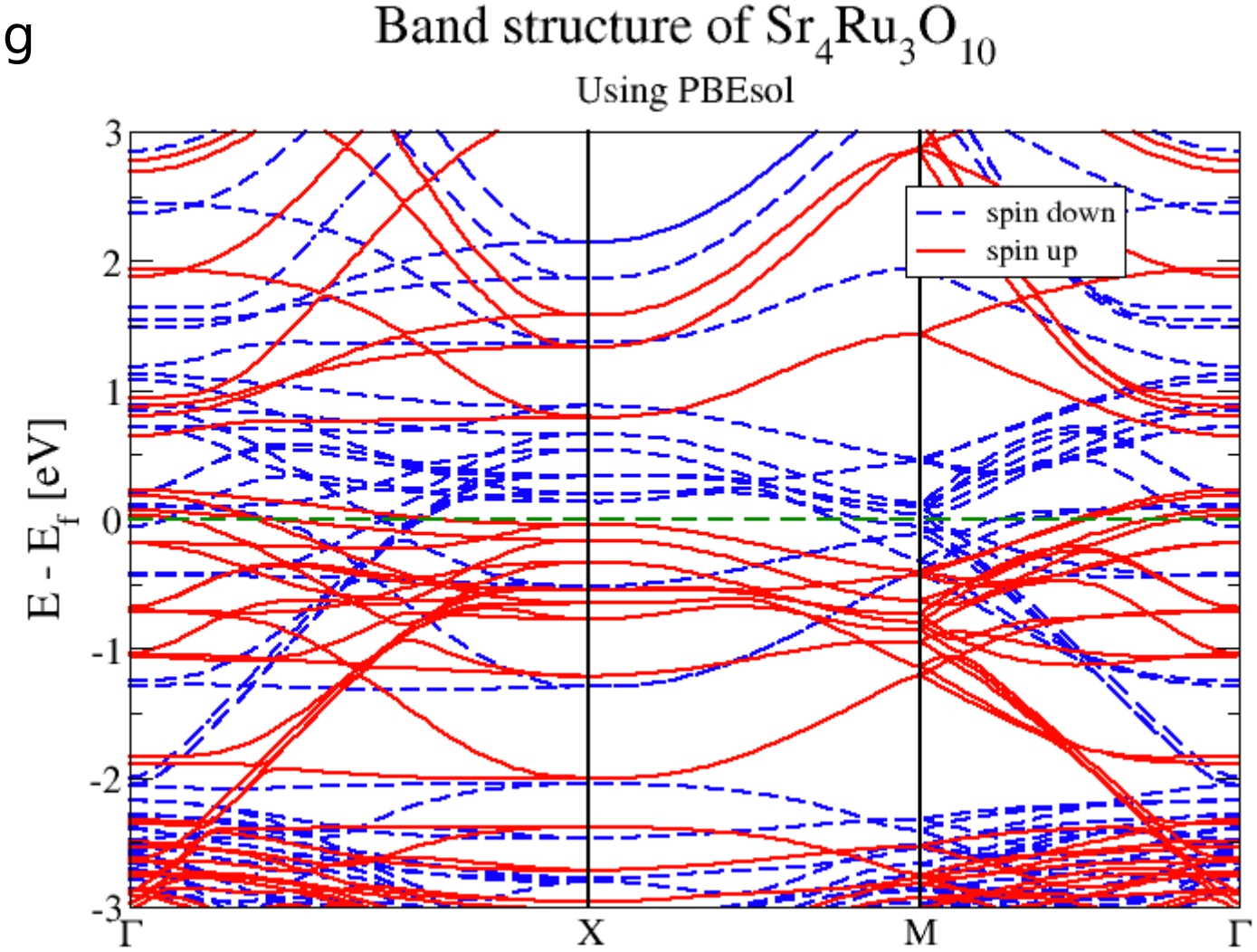
M. K. Crawford, R. L. Harlow, W. Marshall, Z. Li, G. Cao, R. L. Lindstrom, Q. Huang, and J. W. Lynn, Phys. RevB. **65,** 214412, (2002).

Electronic properties

The calculated band structure using PBEsol:

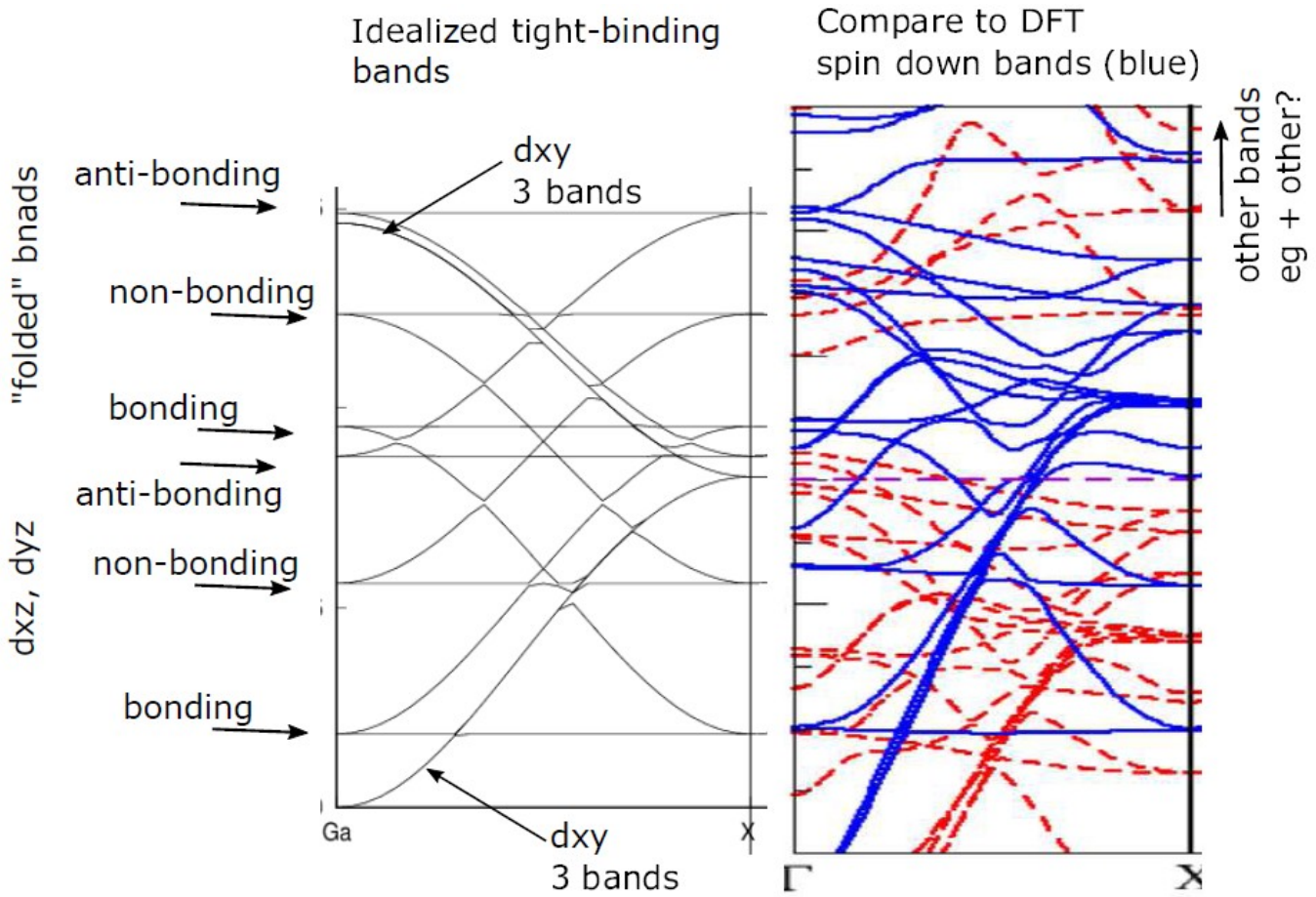
Effect of ferromagnetism:
- shift of the two spin bands.
- Both spins are partially occupied and each has bands that cross the Fermi energy.

The bands at the Fermi energy:
- mainly majority spin hole-like bands near completely minority spin and electron-like in the rest of the BZ.



Electronic properties

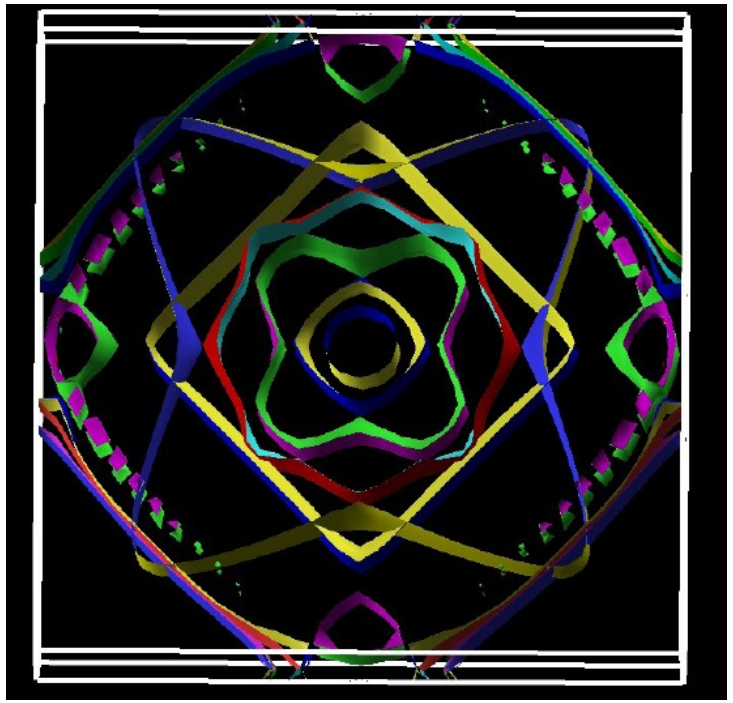
The nature of the bands arizing from the dxy , dxz and dyz t2g states



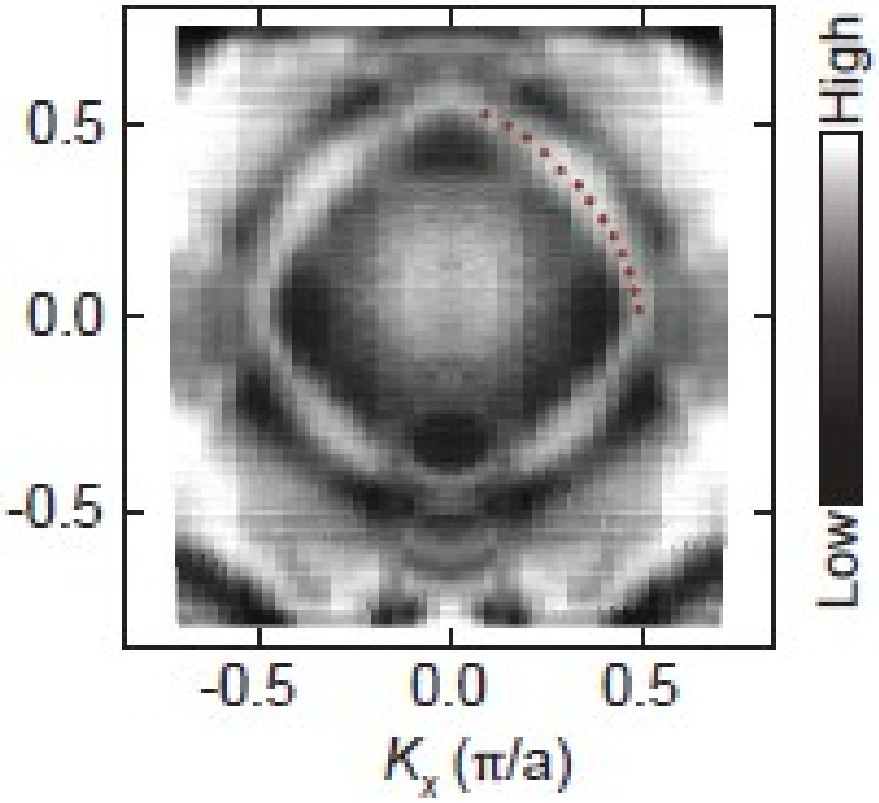
Prosper Ngabonziza, E Carleschi, V Zabolotnyy, A Taleb-Ibrahimi, F Bertran, R Fittipaldi, V Granata, M Cuoco, A Vecchione, BP Doyle, Scientific Reports, **10**, 21062, (2020).

Electronic properties

The calculated Fermi surface using PBEsol:



Fermi surface by ARPES from [1]



1. Prosper Ngabonziza, E Carleschi, V Zabolotnyy, A Taleb-Ibrahimi, F Bertran, R Fittipaldi, V Granata, M Cuoco, A Vecchione, BP Doyle, Scientific Reports, **10**, 21062, (2020).

Ideas For Collaborations:

1. Engaging with African Scientific Communities:

- Oraganise a workshop in Africa that involves large scientific community from US and Africa (create a platform for networking and showcases research outputs and/or ideas)
- Example of successful Scientific platforms: ASESMA, ICTP (some International Workshops).

2.Visiting Scientists programs

- Organise visit programs from the US scientific community to different African Universities.
- Organise a regional workshops that involves the Visiting scientist and local scientific communities.
- Develop and fund common research projects

3. PhD/Masters programs (co-advising)

Acknowledgment

Prof. Nicola Marzari

Dr. Iurii Timrov

MARVEL

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Dr. Nicola Seriani

Dr. Prosper Ngabonziza

Jonah Nagura