RUHR UNIVERSITÄT BOCHUM

DFT+DMFT for Oxide Heterostructures

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Autumn School on Correlated Electrons:

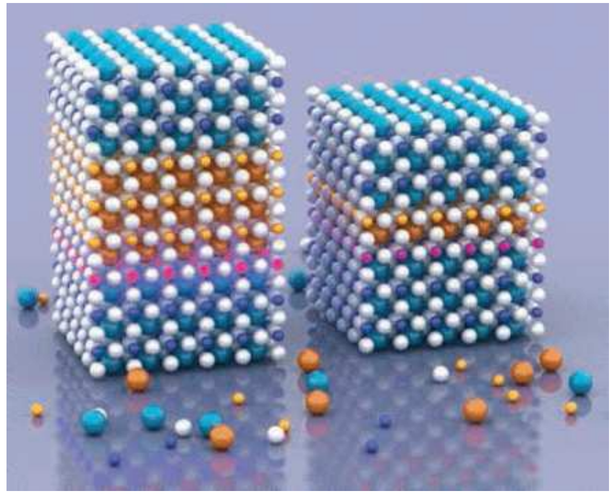
Dynamical Mean-Field Theory of Correlated Electrons

4-7 October 2022, Forschungszentrum Jülich

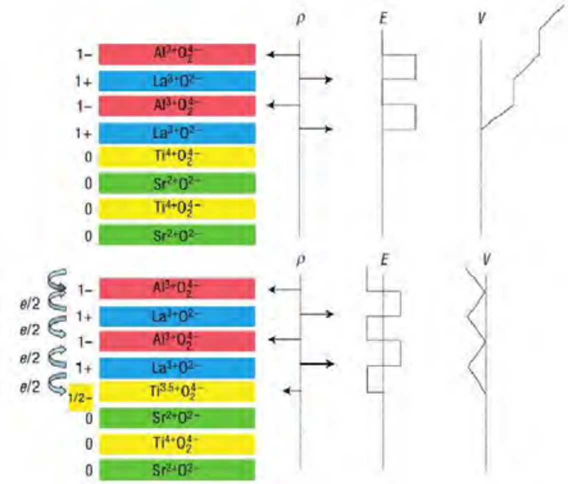
JÜLICH FORSCHUNGSZENTRUM

Oxide Heterostructures : (selected) Phenomenology

- superlattice/thin film build from
  - $SrTiO_3$ (band insulator)
  - $LaAlO_3$ (band insulator)
  - $LaTiO_3$ (Mott insulator)
  - ⋮
- possible quasi-twodimensional electron system (2DES)
- instabilities : ferromagnetism, superconductivity, . . .



e.g. [Hwang et al., Nature Mat. 11, 103 (2012)]



[Nakagawa et al., Nature Mat. 5, 204 (2006)]

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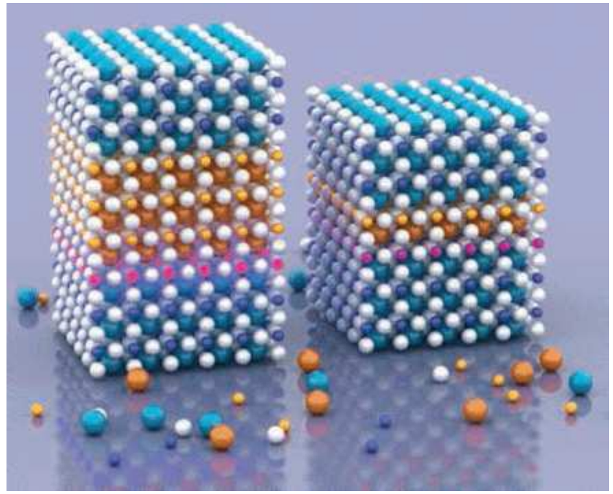

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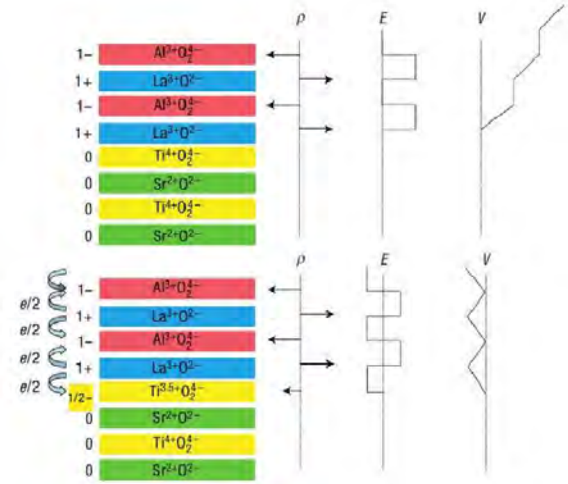
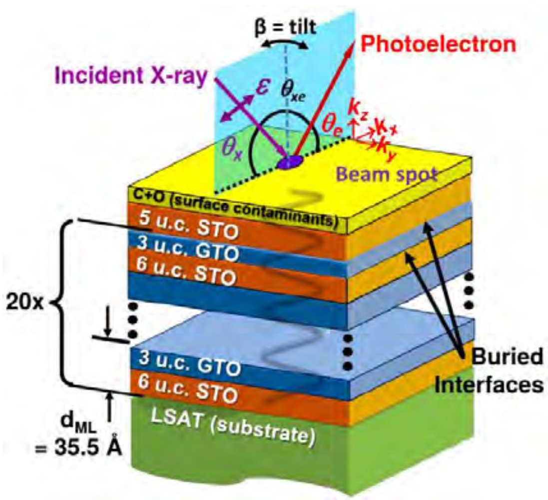
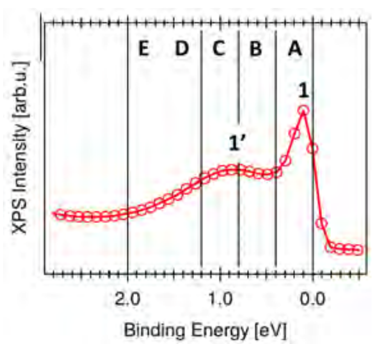


[Nakagawa et al., Nature Mat. 5, 204 (2006)]

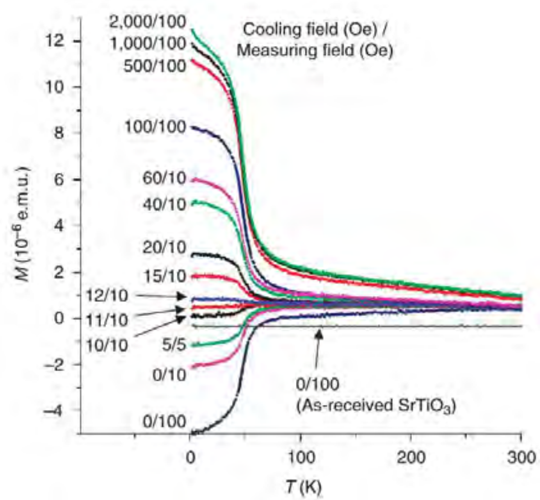
band-Mott insulators e.g. $SrO/GdTiO_3$





[Nemšák et al., PRB 93, 245103 (2016)]

band-band insulators e.g. $LaAlO_3/SrTiO_3$



[Ariando et al., Nat. Commun. 2, 188 (2015)]

Oxide Heterostructures : Theoretical Work

- model-Hamiltonian dynamical mean-field theory (DMFT) for Mott/band-insulator interfaces

[Okamoto and Millis, Nature 428, 630 (2004)]

- density functional theory (DFT) for band-insulating heterostructures

[Popovic and Satpathy, PRL 94, 176805 (2005)]

- DFT plus static Hubbard correlations (DFT+U) for band- and Mott heterostructures

[Pentcheva and Pickett, PRL 99, 016802 (2007)] [Pavlenko et al., PRB 86, 064431 (2012)]

- model-Hamiltonian field-theoretical approach to $LaAlO_3/SrTiO_3$

[Joshua et al., Nat. Commun. 3, 1129 (2012)]

- (realistic) Anderson-Hamiltonian exact-diagonalization/Lanczos method for isolated-defect $SrTiO_3$

[Lin and Demkov, PRL 111, 217601 (2014)]

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<u>this talk</u>

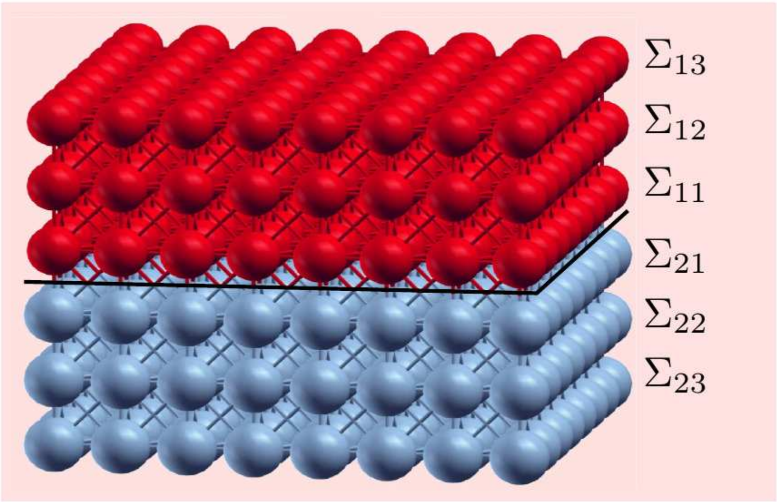
⋆ realistic many-body approach via DFT+DMFT

⋆ correlation physics by design

⋆ other recent DFT+DMFT approaches to oxide heterostructures

[K. Held, G. Sangiovanni, P. Hansmann, J. Tomczak] [C. Ederer] [C. Marianetti, A. J. Millis], [E. Dagotto], ...

Oxide Heterostructures : Why at all?



issues and features

- charge transfers and electrostatic boundary conditions
- spin and orbital reconstructions
- compressive and tensile strain
- source/sink for additional (point) defects
- two-dimensional characteristics

prospects

- unique, well-defined doping scenario
- tuning (proximity) options building up on the bulk
- (control of) layer-selective physics
- possibly novel (emergent) electronic phases
- oxide membranes and merging oxides with other materials

Outline

- theory: from DFT to DFT+DMFT
- interfacing band insulator with band insulator
- interfacing band insulator with Mott insulator
- natural-heterostructure systems: delafossites

Density Functional Theory (DFT) : Effective Single-Particle Approach

electronic density functional theory (DFT) with approximate exchange-correlation

- DFT is ground-state many-body theory with inhomogeneous charge density $n(\mathbf{r})$ ($\Leftrightarrow \; \Psi(\{\mathbf{r}_i\sigma_i\})$) as central quantity, formally exact

$$E[n] = T[n] \; + \int d\mathbf{r}\, n(\mathbf{r}) v_{\mathrm{ext}}(\mathbf{r}) \; + \; E_{\mathrm{H}}[n] \; + \; \mathbf{E_{xc}[n]}$$

- approximate exchange-correlation functional $\mathbf{E_{xc}}$ in effective single-particle Kohn-Sham mapping to render the approach practicable

local density approximation (LDA)	:	homogeneous electron gas is reference
generalized-gradient approximation (GGA)	:	∇n corrections to LDA

- **works very well for many realistic systems !**
  $\rightarrow$ many molecules, simple metals, semiconductors (not band gap), . . .

W. Kohn, Nobel Prize 1998

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- **PROBLEM: insufficient for materials with strong correlations among the electrons !**

  - some systems with odd electron number in unit cell are isolating
  - satellite structures in one-particle spectral function
  - strong changes of spectral function with doping
  - metal-to-insulator transition without symmetry change
  - coherence temperatures and generally subtle behavior with temperature

⋮

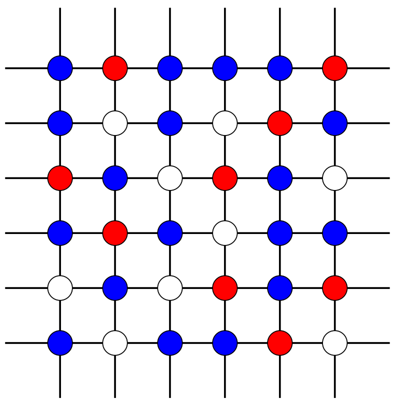
Correlated Electrons : Quasiparticles and Coherence, intuitively

.

Correlated Electrons : Mott-Hubbard picture

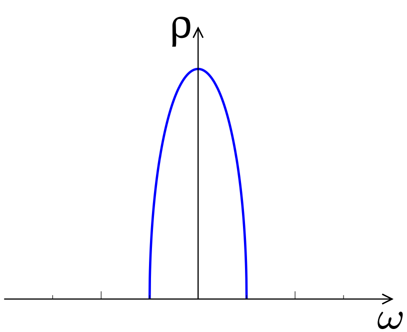
Hubbard model: $H = -\mathbf{t} \sum_{\langle ij \rangle \sigma} c^\dagger_{i\sigma} c_{j\sigma} + \mathbf{U} \sum_i n_{i\uparrow} n_{i\downarrow}$, for half filling $n = 1$

nonint. limit $(U = 0)$

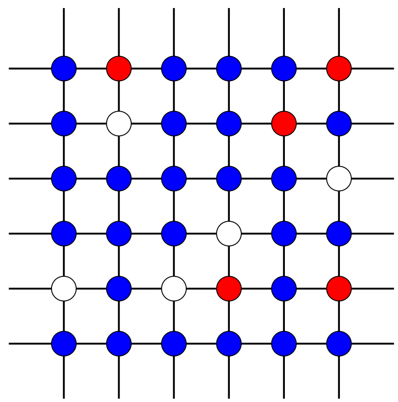


– $H = \sum_{\mathbf{k}\sigma} \varepsilon_{\mathbf{k}} \, c^\dagger_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma}$

– ideal metal, Fermi gas

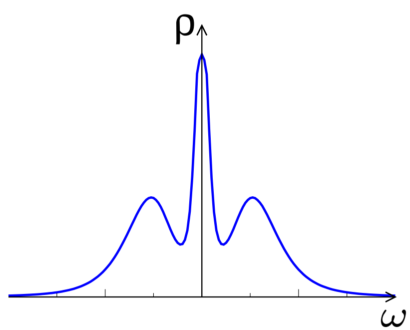


intermediate $(U \sim t)$



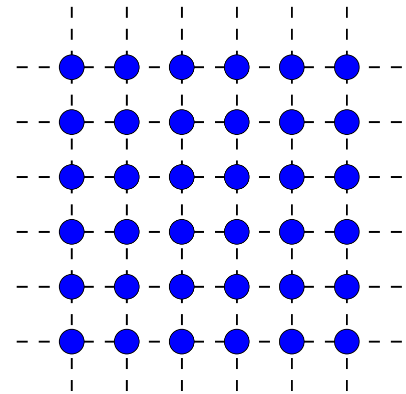
– correlated metal

– dominance of single occupations



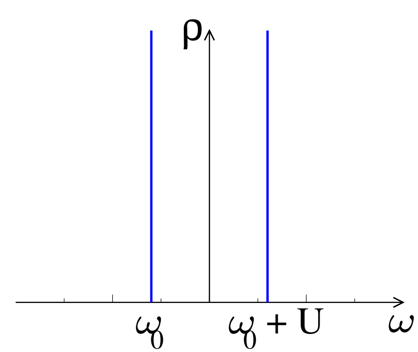
"Three-Peak Structure"

atomic limit $(t = 0)$



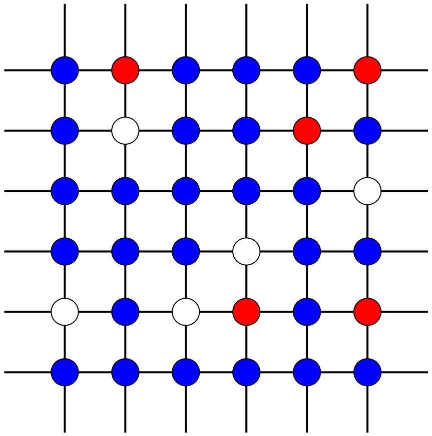
– decoupled lattice sites

– only single occupations in ground state

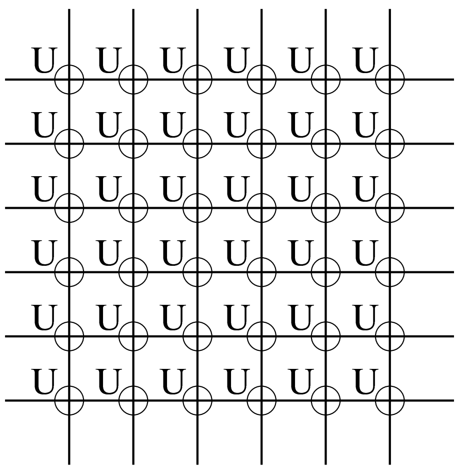


Dynamical Mean-Field Theory (DMFT) : Many-Body Approach

[Metzner, Vollhardt, PRL (1989)] [Georges, Kotliar, PRB (1992)]

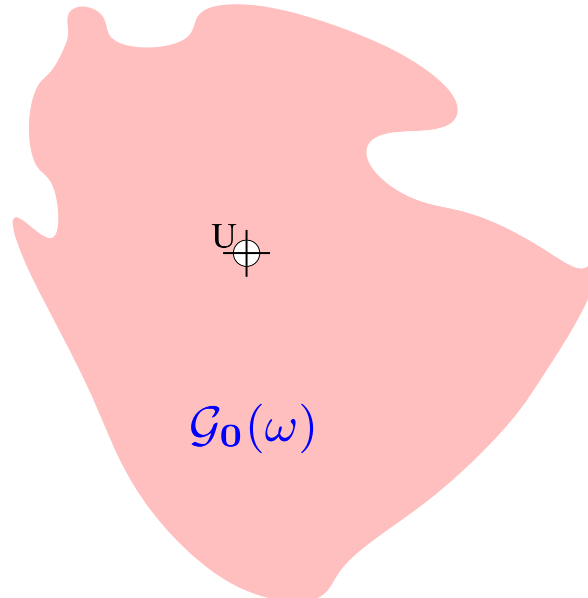


$$H = -t \sum_{\langle ij \rangle \sigma} c^{\dagger}_{i\sigma} c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}$$



$$G_{\mathrm{loc}}(\omega) = \sum_{\mathbf{k}} [\omega + \mu - \varepsilon_{\mathbf{k}} - \Sigma(\mathbf{k}, \omega)]^{-1}$$

$\Rightarrow$



$$G_{\mathrm{imp}}(\omega) = \left[\mathcal{G}_0^{-1}(\omega) - \Sigma_{\mathrm{imp}}(\omega) \right]^{-1}$$

DMFT approximation: $G_{\mathrm{loc}}(\omega) \stackrel{!}{=} G_{\mathrm{imp}}(\omega)$

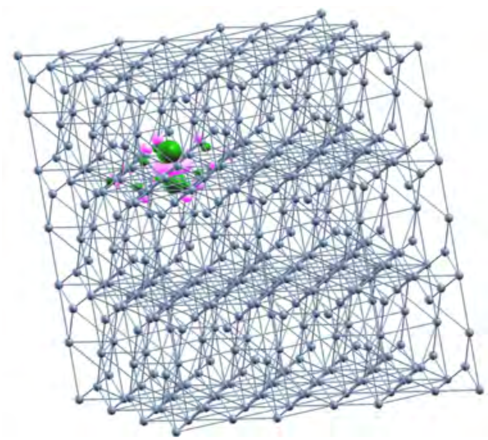
$$G^{\mathrm{DMFT}}_{\mathrm{loc}}(\omega) = \sum_{\mathbf{k}} [\omega + \mu - \varepsilon_{\mathbf{k}} - \Sigma_{\mathrm{imp}}(\omega)]^{-1}$$

$\rightarrow$ local correlations are fully taken into account non-pertubatively, explicit non-local correlations are neglected

Correlated Materials : different levels of correlation

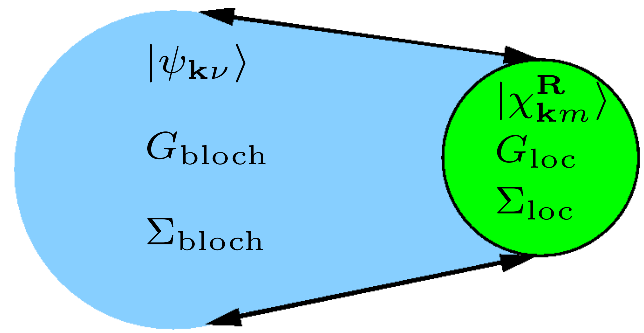
density functional theory (DFT)

- Hohenberg-Kohn theorems, Kohn-Sham construction, exchange-correlation functional, . . .
- mapping interacting electrons onto non-interacting electrons in an complicated effective potential
- very good description of the realistic, single-particle based chemical bonding ($\rightarrow$ band structure for solids)

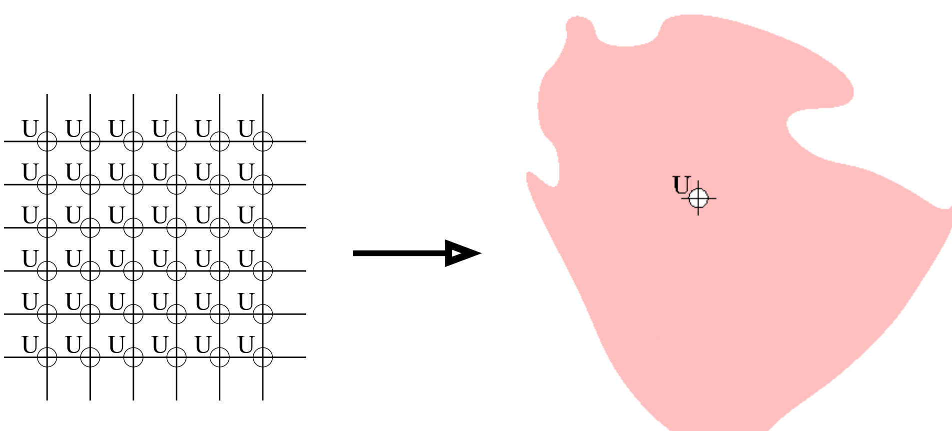


interface

- define correlated subspace by Wannier(-like) orbitals from correlated sites



dynamical mean-field theory (DMFT)

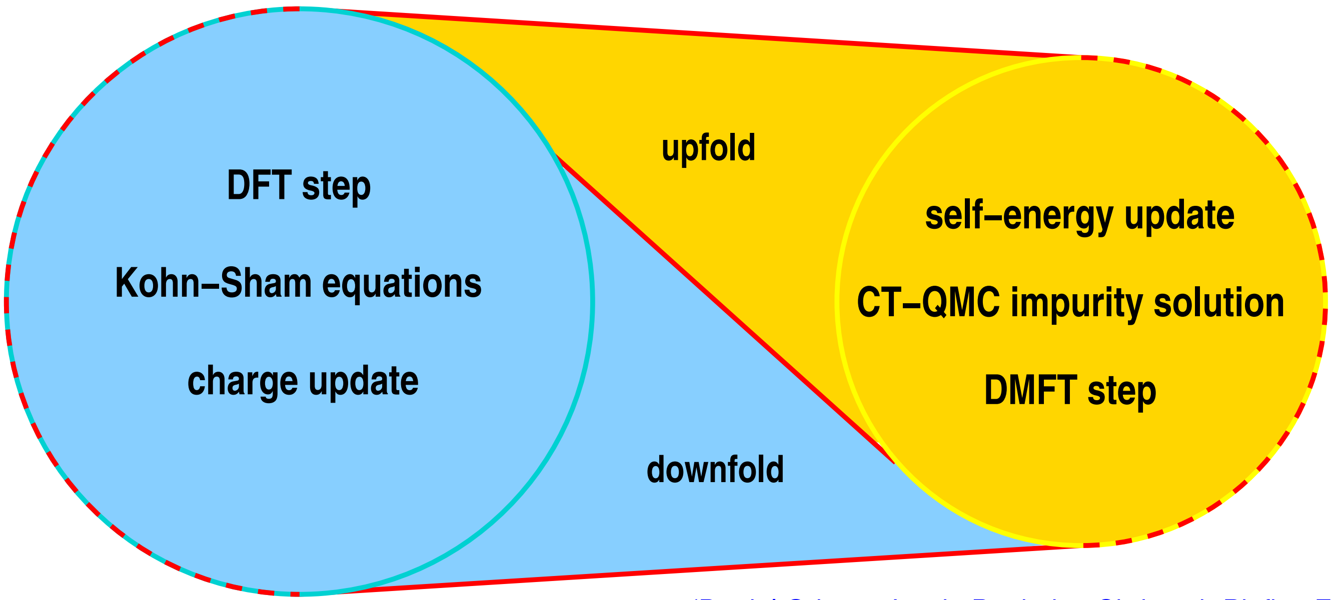


- many-body Green's functions, Hubbard model, quasiparticles vs. Hubbard bands, finite temperature, . . .
- mapping interacting electrons onto an interacting impurity in a self-consistent bath
- very good description of modelized interacting electrons on a lattice

Realistic Many-Body

[Anisimov, Poteryaev, Korotin et al., JPCM 9, 7359 (1997)]
[Lichtenstein and Katsnelson, PRB 57, 6884 (1998)]
[Held, Keller, Eyert et al., PRL 86, 5345 (2001)]
[Pavarini, Biermann, Poteryaev et al., PRL 92, 176403 (2004)]
[Minár, Chioncel, Perlov, at al., PRB 72, 045125 (2005)]
[Kotliar, S. Y. Savrasov, K. Haule et al, RMP 78, 865 (2006)]
[FL, Georges, Poteryaev et al., PRB 74, 125120 (2006)]
[Grieger, Piefke, Peil and FL, PRB 86, 155121 (2012)] . . . , . . . , . . .

Density Functional Theory (DFT) + Dynamical Mean–Field Theory (DMFT)



(Daniel Grieger, Lewin Boehnke, Christoph Piefke, FL)

- DFT part : mixed-basis pseudopotential

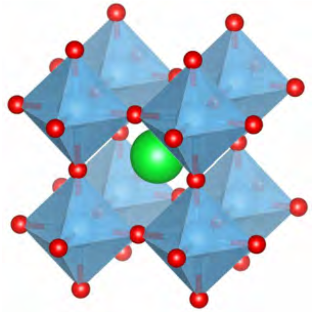
[Meyer, Elsässer, Lechermann and Fähnle, MPI for Metals Research]

- DMFT impurity solver : hyb-CT-QMC (TRIQS code)

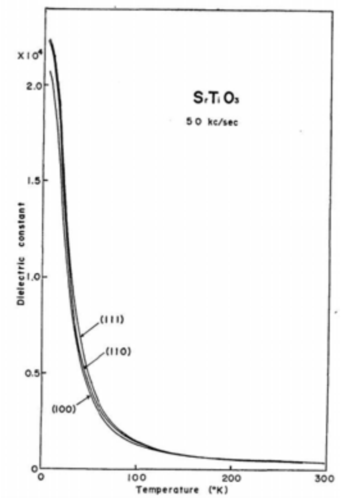
[Parcollet et al., Comput. Phys. Commun. 196, 398 (2015)]
[Seth et al, Comput. Phys. Commun. 200, 274 (2016)]

- charge self-consistency
- 1-5 local correlated orbitals
- up to 200-atom unit cells
- OX-HE: manifest multi-site DMFT

Bulk $SrTiO_3$: A Band Insulator prone to a Plethora of Physics

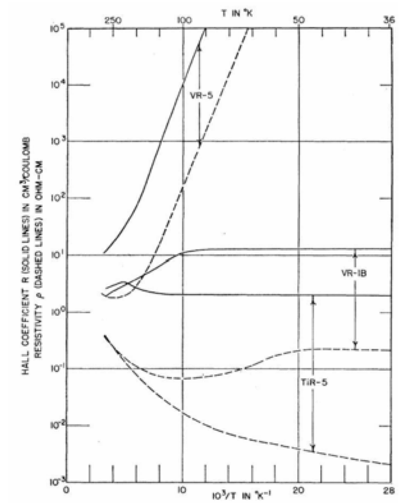


- cubic perovskite, transition to tetragonal phase at low temperature
- band insulator ($\Delta_g \sim 3.2$ eV)



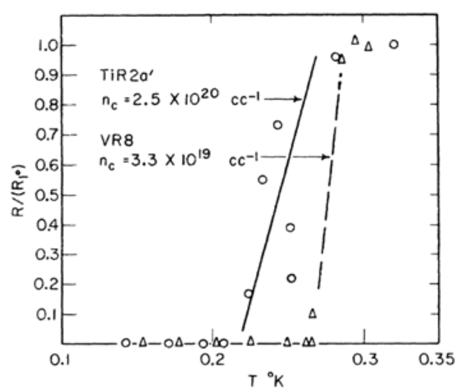
paraelectric

[Sawaguchi et al., JPSN 17, 1666 (1962)]



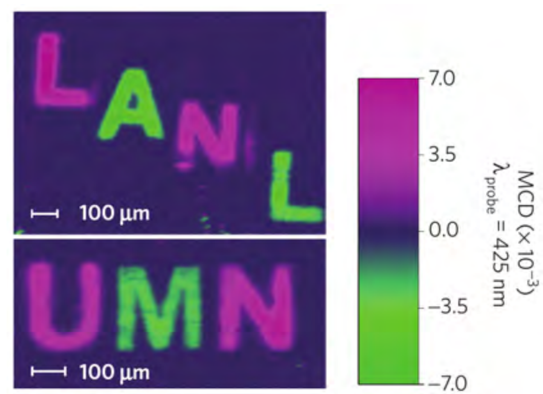
semiconductor

[Frederikse et al., PR 134, A442 (1964)]



superconductor

[Schooley et al., PRL 12, 474 (1964)]



magnet

[Rice et al. Nat. Mater. 13, 481 (2014)]

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LETTERS

PUBLISHED ONLINE: 4 SEPTEMBER 2011 | DOI: 10.1038/NPHYS2080

nature physics

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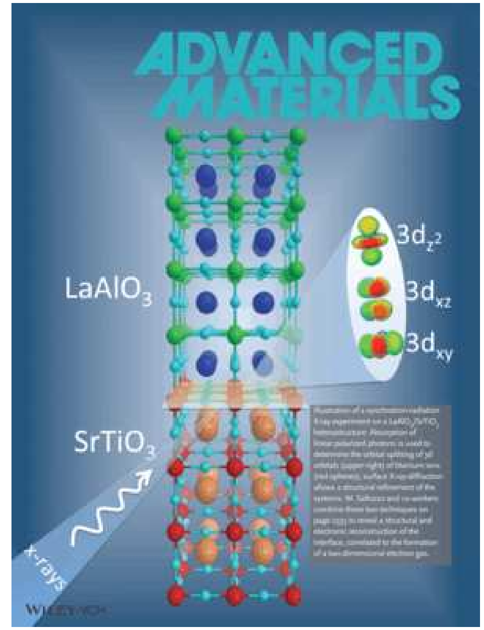

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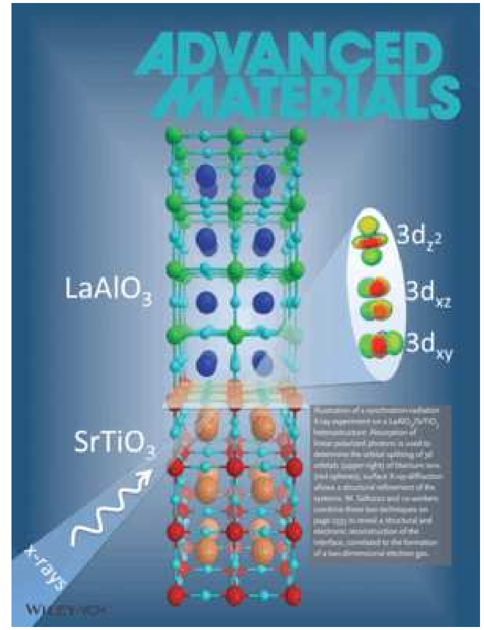
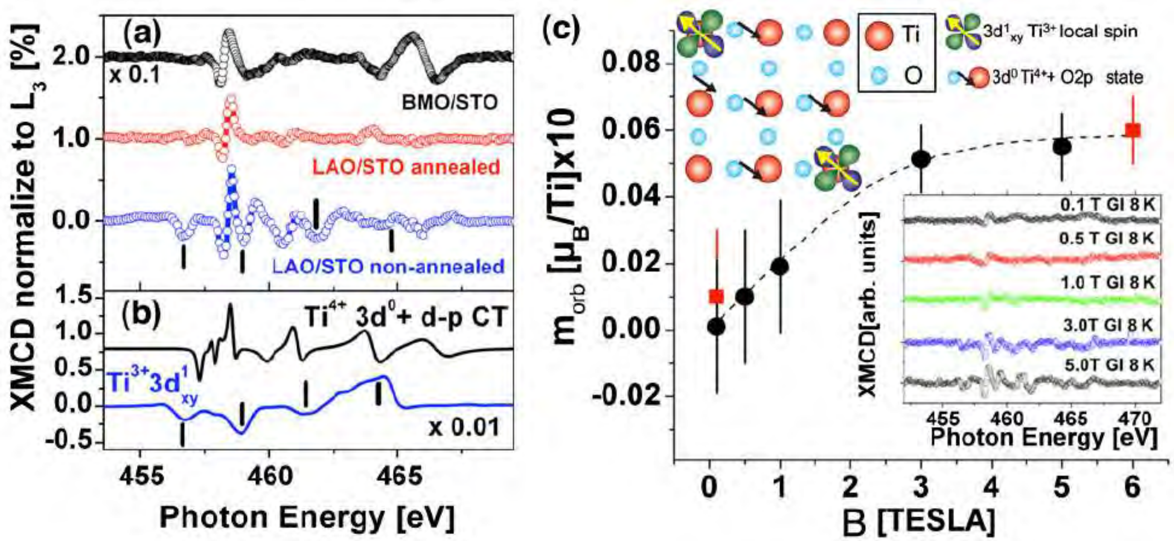


RIXS and XMCD measurements

relevance of oxygen vacancies at the interface

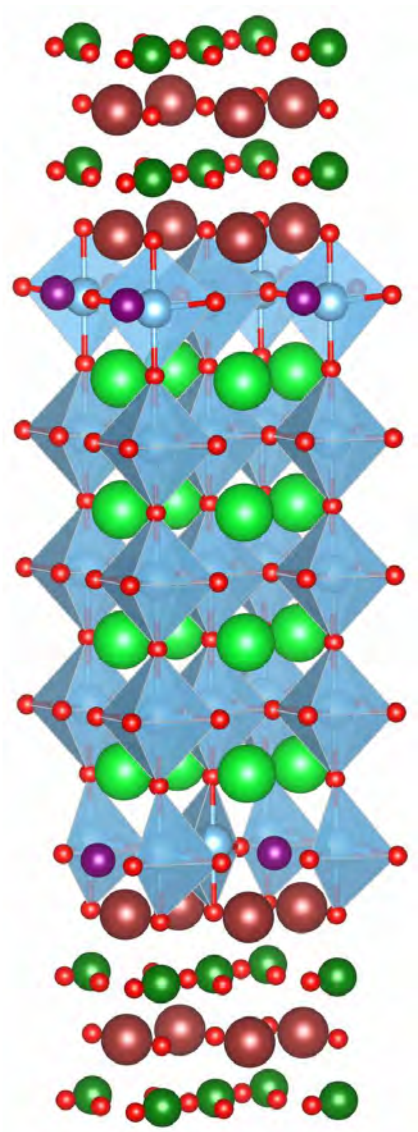
[Park et al., PRL 110, 017401 (2013)]

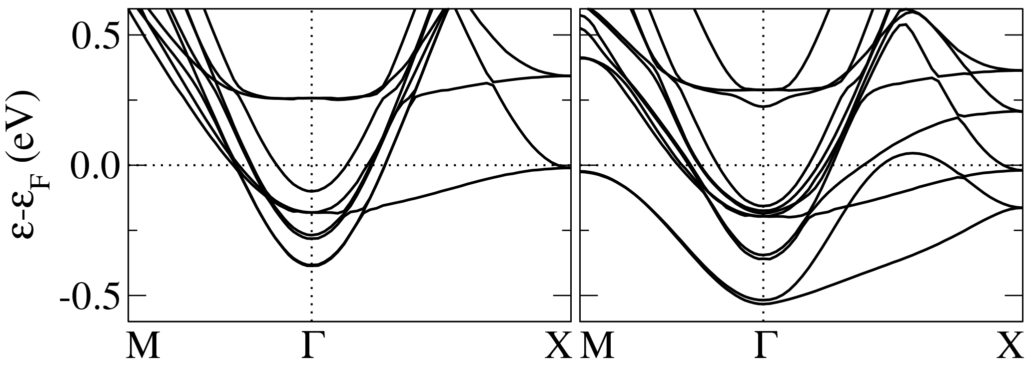
[Salluzzo et al., PRL 111, 087204 (2013)]

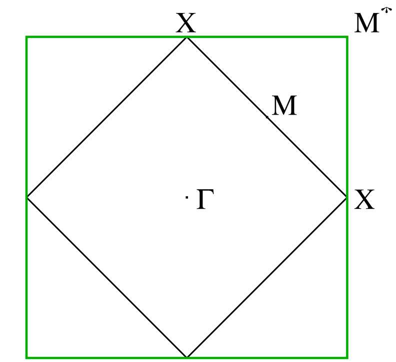


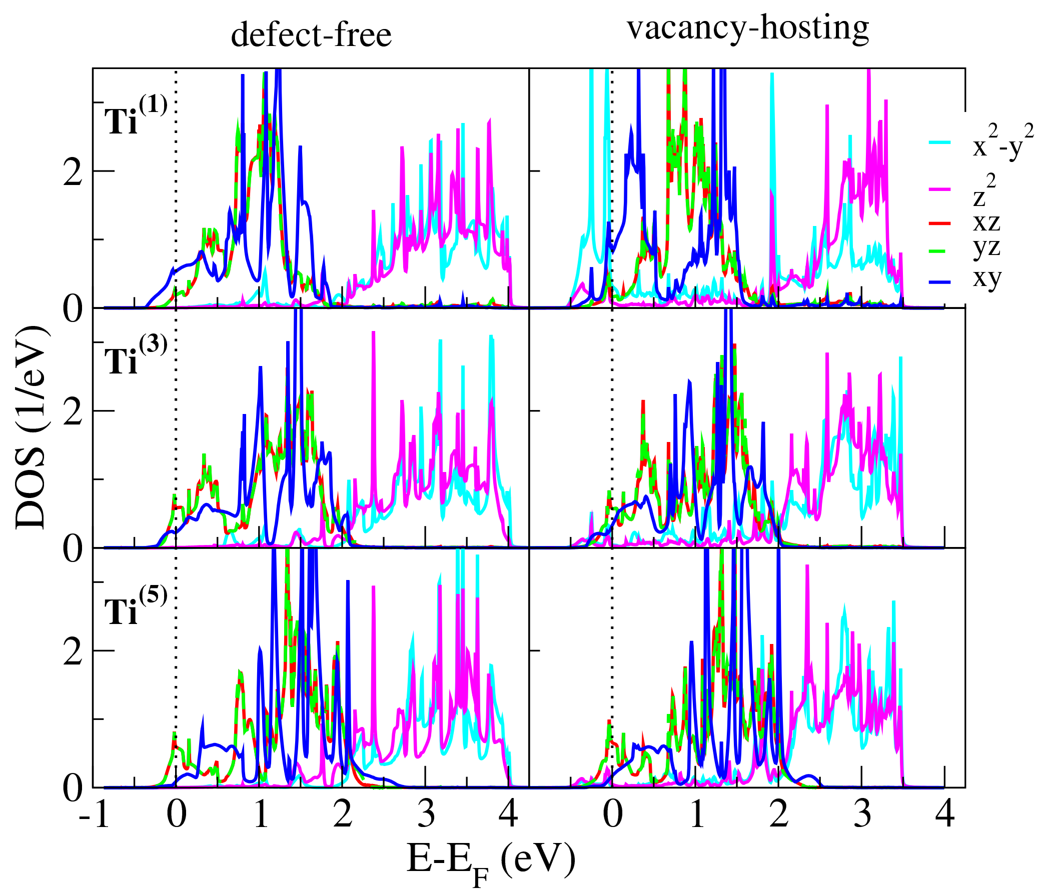
- oxygen vacancies introduce electrons that localize at the nearby Ti sites ($m \sim 0.1 - 0.2\mu_B$)

$LaAlO_3/SrTiO_3$ interface: DFT results



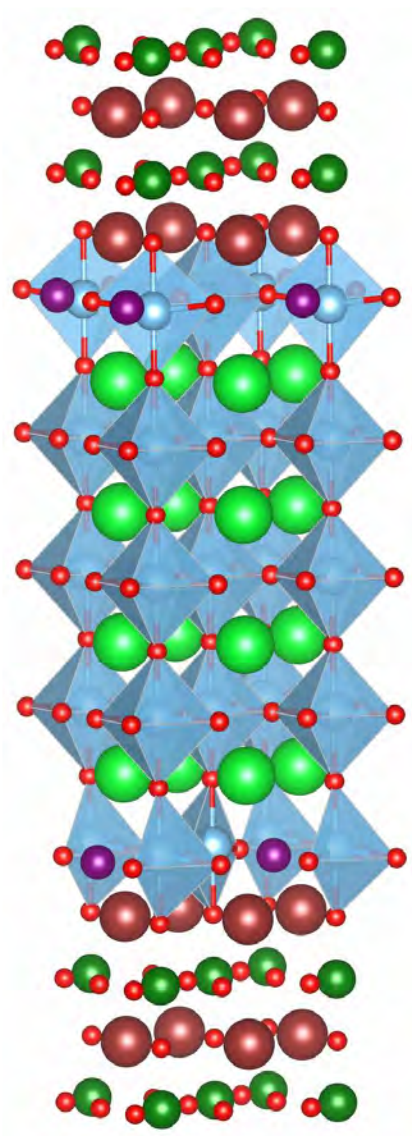




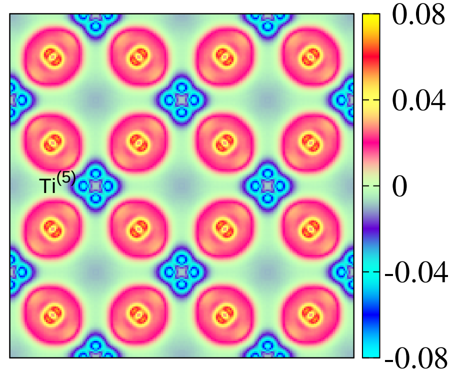


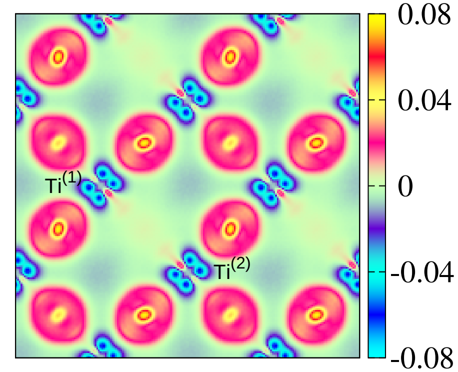
see also [Pavlenko et al., PRB 85, 020407(R) (2012); PRB 88, 201104(R) (2013)]

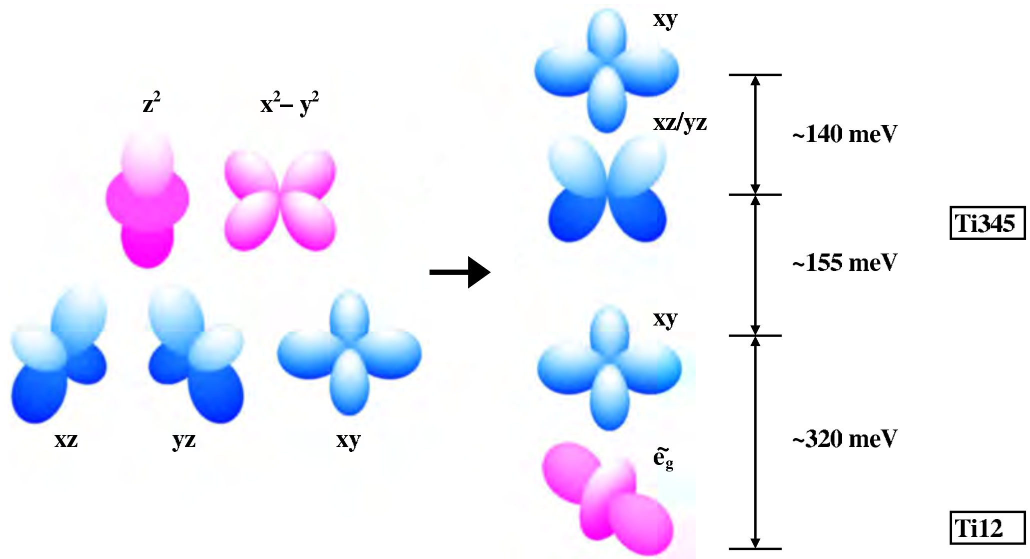
LAO/STO dense-defect interface : Relevant States



in-plane bond charge density
$\rho - \rho_{\rm at}$





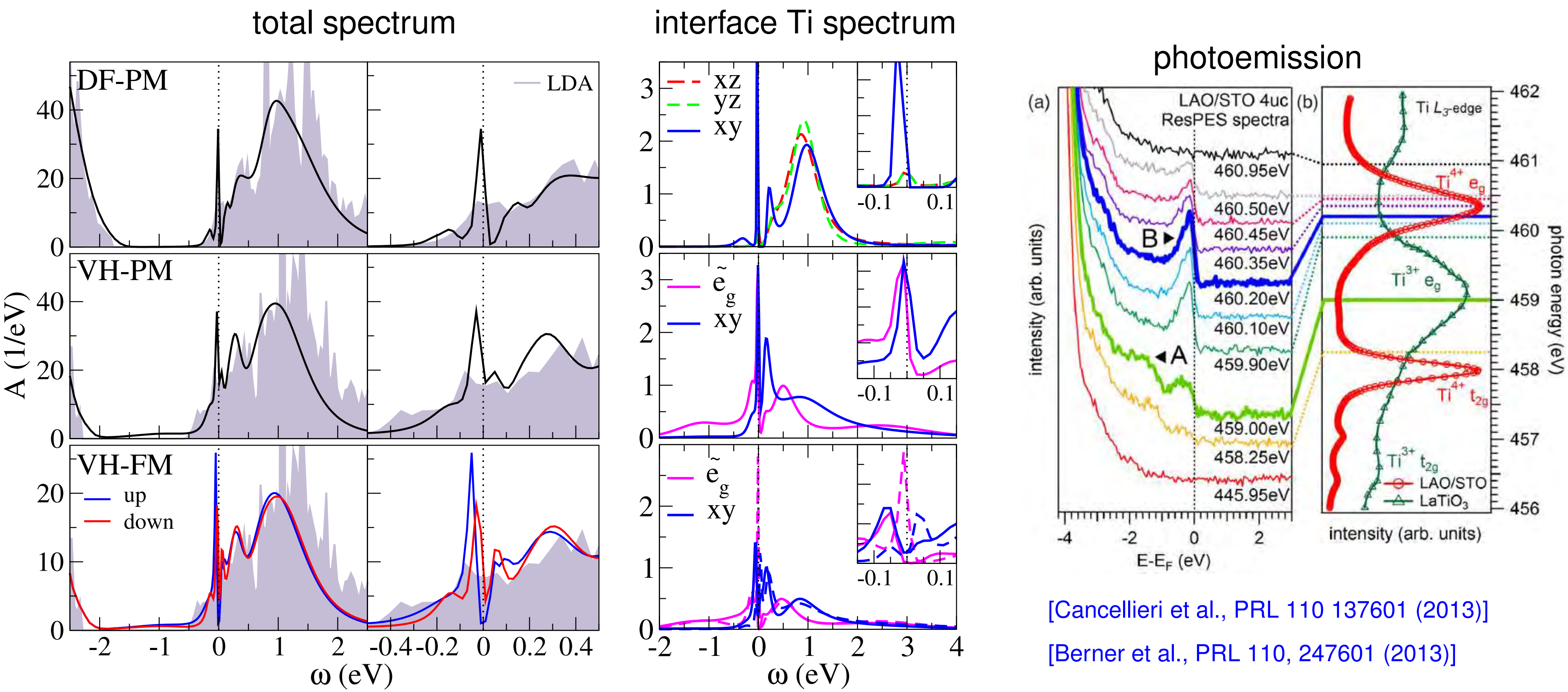


- two-orbital correlated subspace for each Ti ion from orbital projection
- interface Ti: $\tilde{e_g} - t_{2g}(xy)$ description, with $|\tilde{e_g}\rangle \sim 0.55|z^2\rangle \pm 0.84|x^2-y^2\rangle$

$\Rightarrow$ **coexisting localized and itinerant electrons expected !**

$\rightarrow$ in line with RIXS measurements [Zhou et al., PRB 83, 201402(R) (2011)]

LAO/STO dense-defect interface : DFT+DMFT

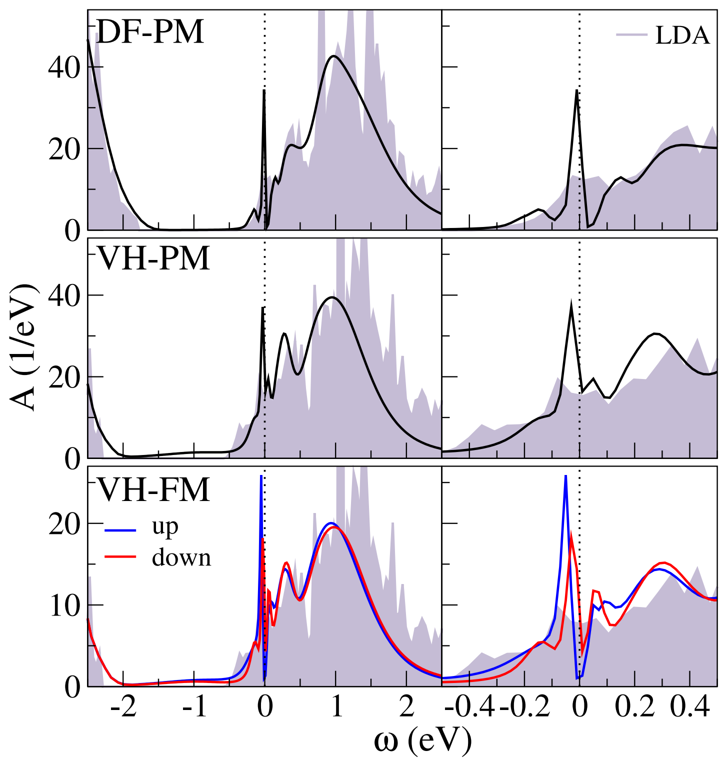


[Cancellieri et al., PRL 110 137601 (2013)]

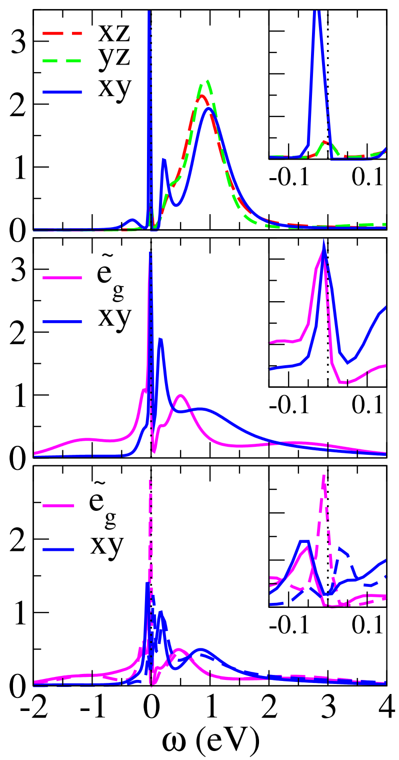
[Berner et al., PRL 110, 247601 (2013)]

LAO/STO dense-defect interface : DFT+DMFT

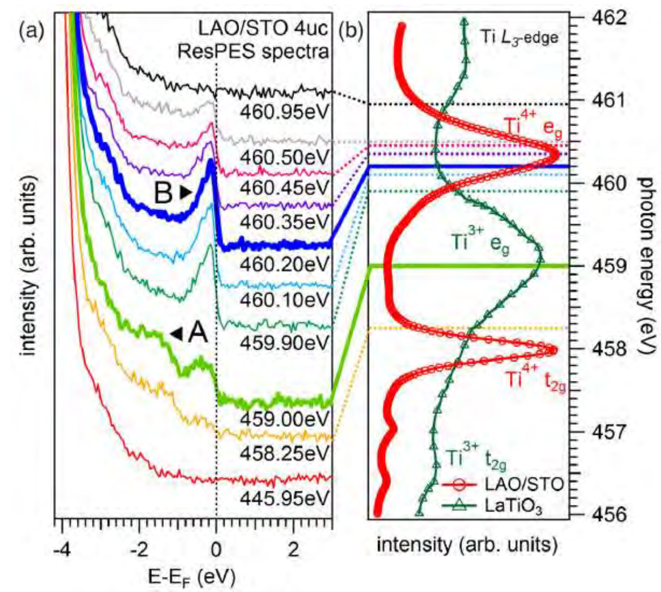
total spectrum



interface Ti spectrum



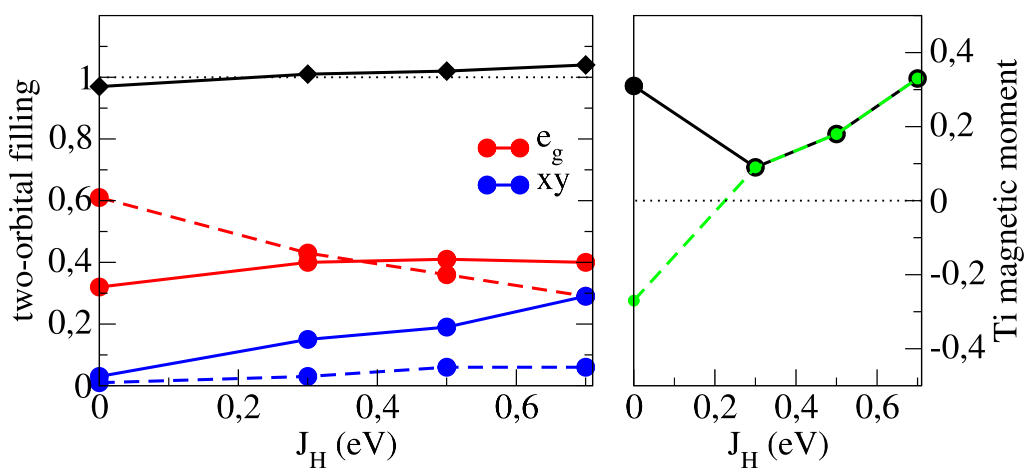
photoemission



[Cancellieri et al., PRL 110 137601 (2013)]

[Berner et al., PRL 110, 247601 (2013)]

local two-orbital fillings and polarizations with vacancies (U=2.5 eV)



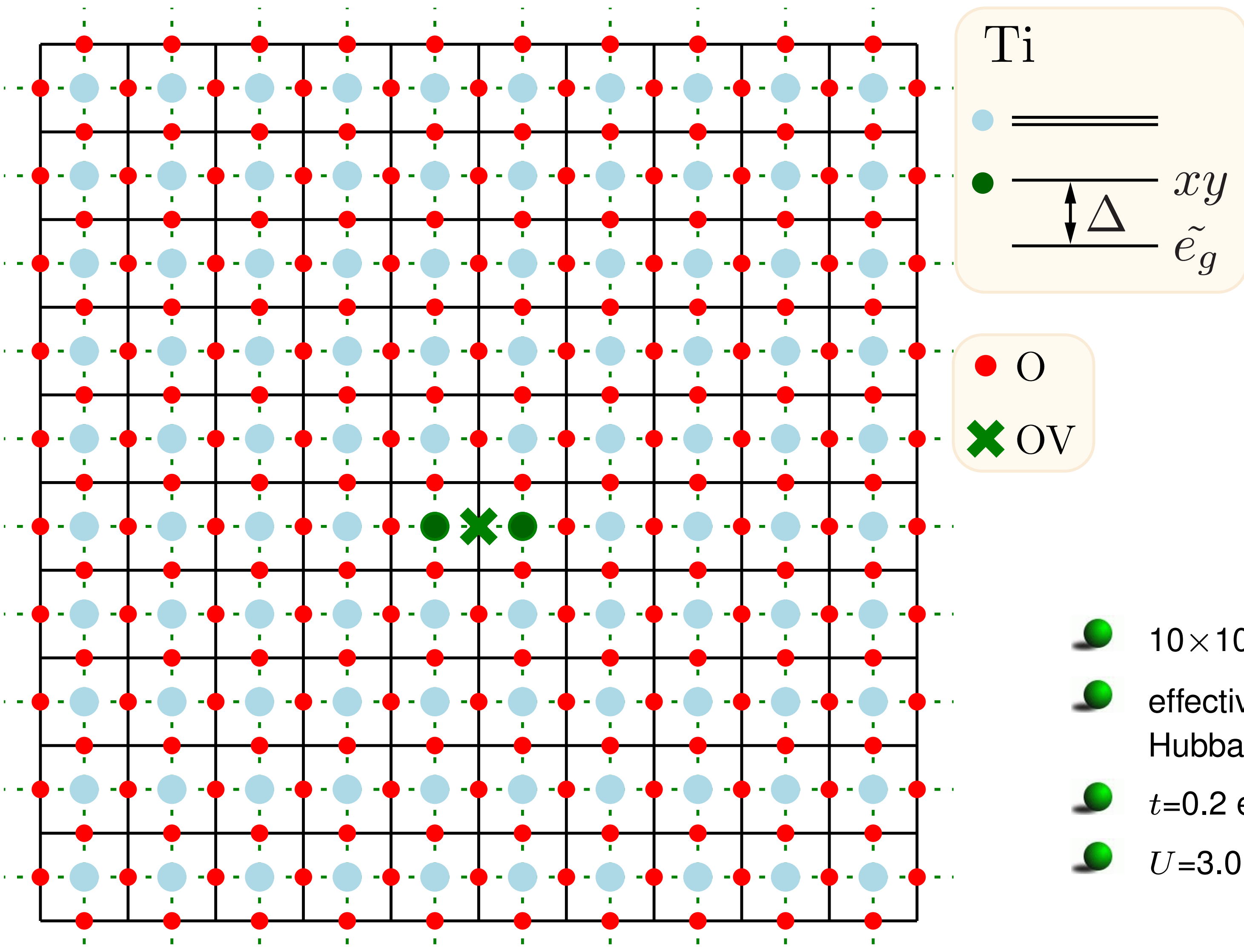
with oxygen vacancies (VH)

- one electron at interface Ti: quarter filling
- FM moment $m_{\mathrm{Ti12}} \sim 0.2\mu_B$
- double-exchange mechanism
- **very sensitive to J_H !**

[FL, Boehnke, Grieger, Piefke, PRB 085125 (2014)]

Arbitrary OV Concentration in LAO/STO : Real-Space Modelling

[Behrmann and FL, PRB 92, 125148 (2015)]



- 10×10 TiO_2 square lattice
- effective two-orbital Hubbard model
- t=0.2 eV, Δ=0.3 eV
- U=3.0 eV, $J_{\rm H}$=0.55 eV

- Gutzwiller/Slave-Boson approach : nearest-neighbor hopping and crystal field from former DFT calculation

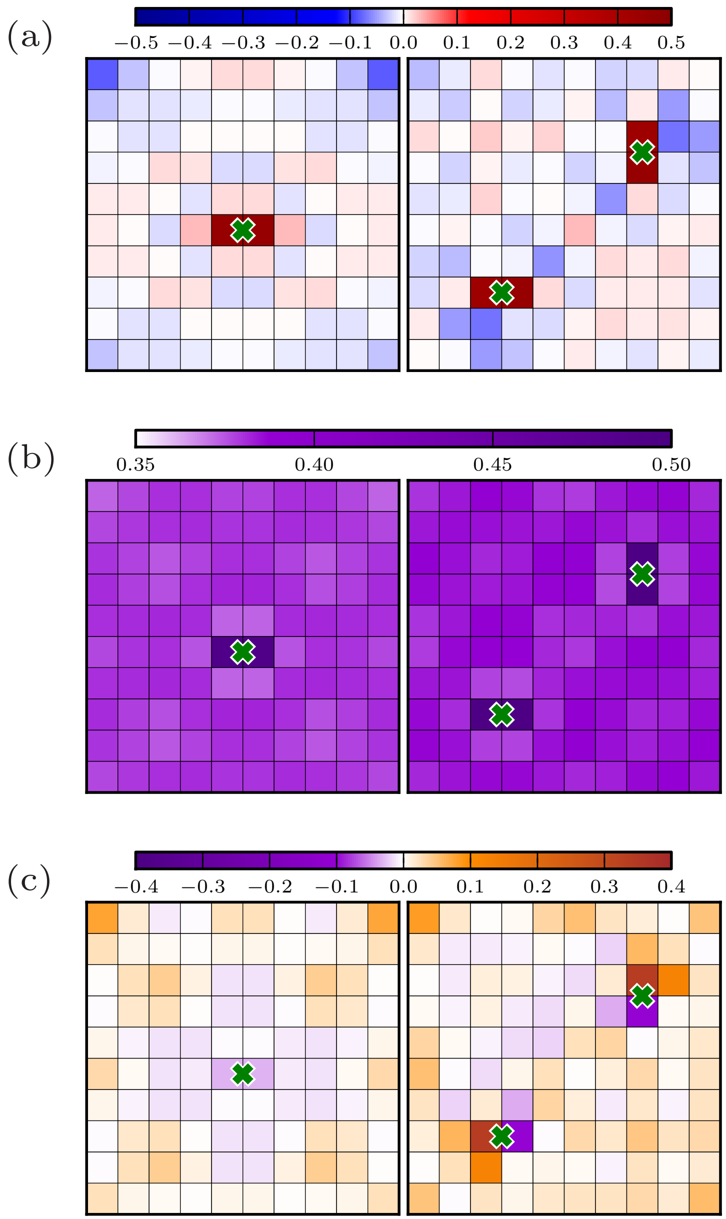
Arbitrary OV Concentration in LAO/STO : Dilute-Defect Limit

[Behrmann and FL, PRB 92, 125148 (2015)]

orbital polarization

local moment

ordered moment



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Arbitrary OV Concentration in LAO/STO : Dilute-Defect Limit

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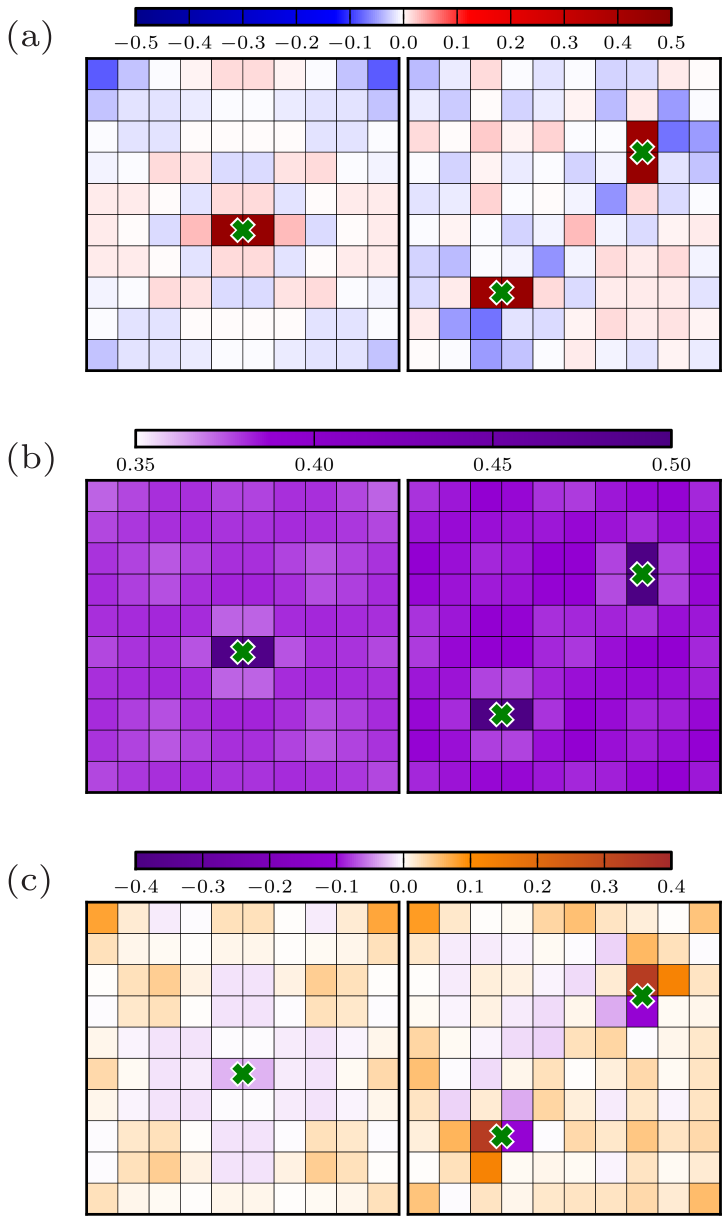
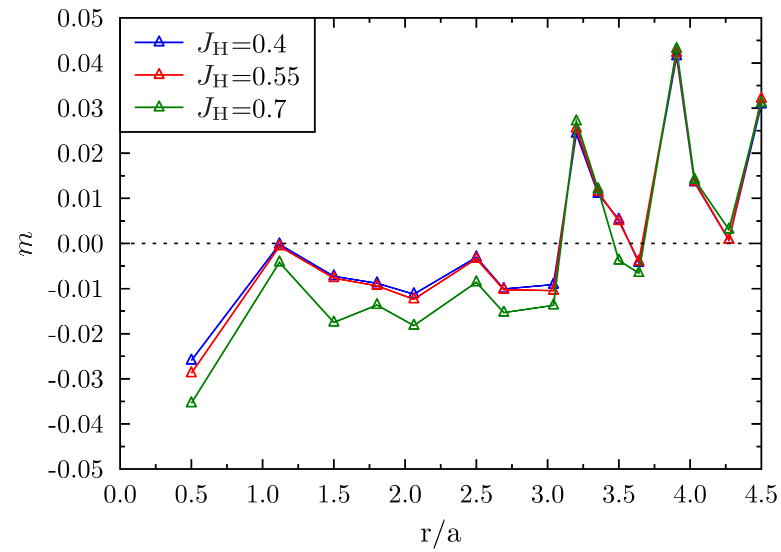




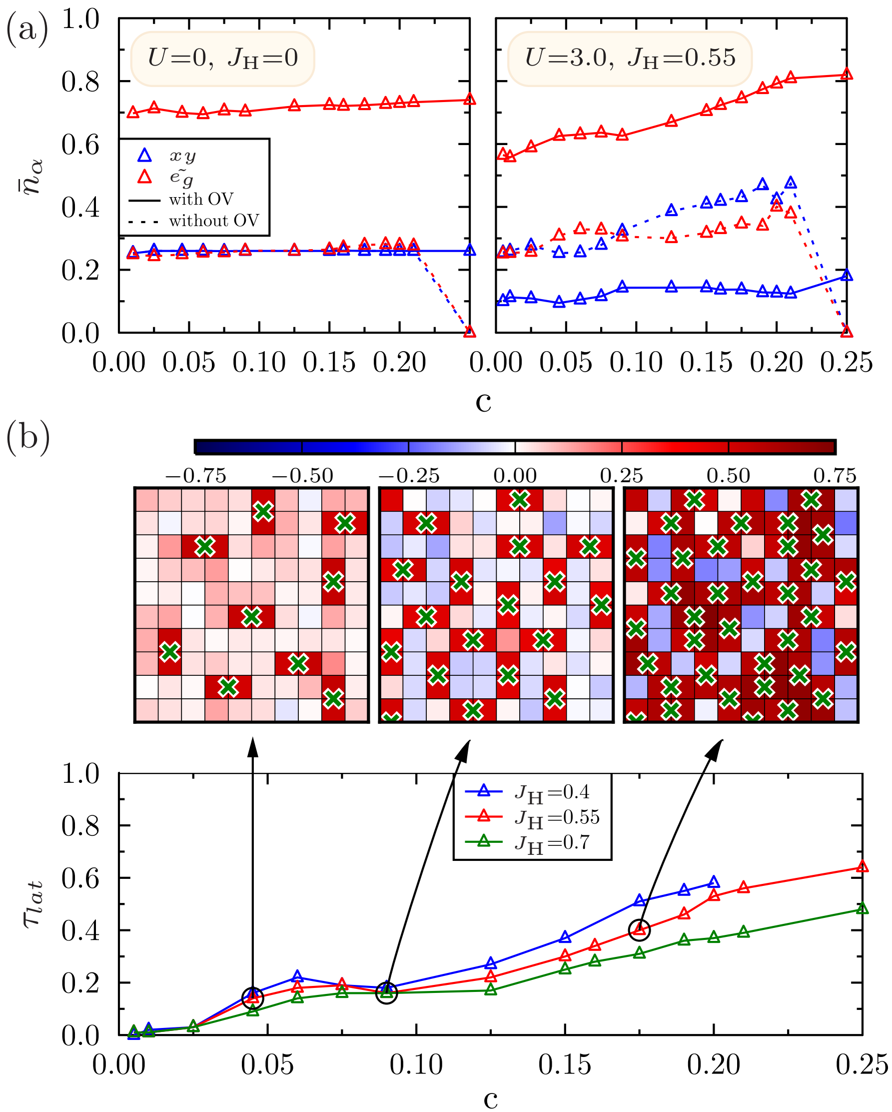
RKKY(-like) coupling

$$J(d)\ \cos\left(2\,k_F\,d\right) \quad , \quad d \sim 2.5 \ \Rightarrow\ k_F \sim \frac{\pi}{10}$$

$\rightarrow$ about 1/8 filling ✓

Arbitrary OV Concentration in LAO/STO : orbital and spin polarization

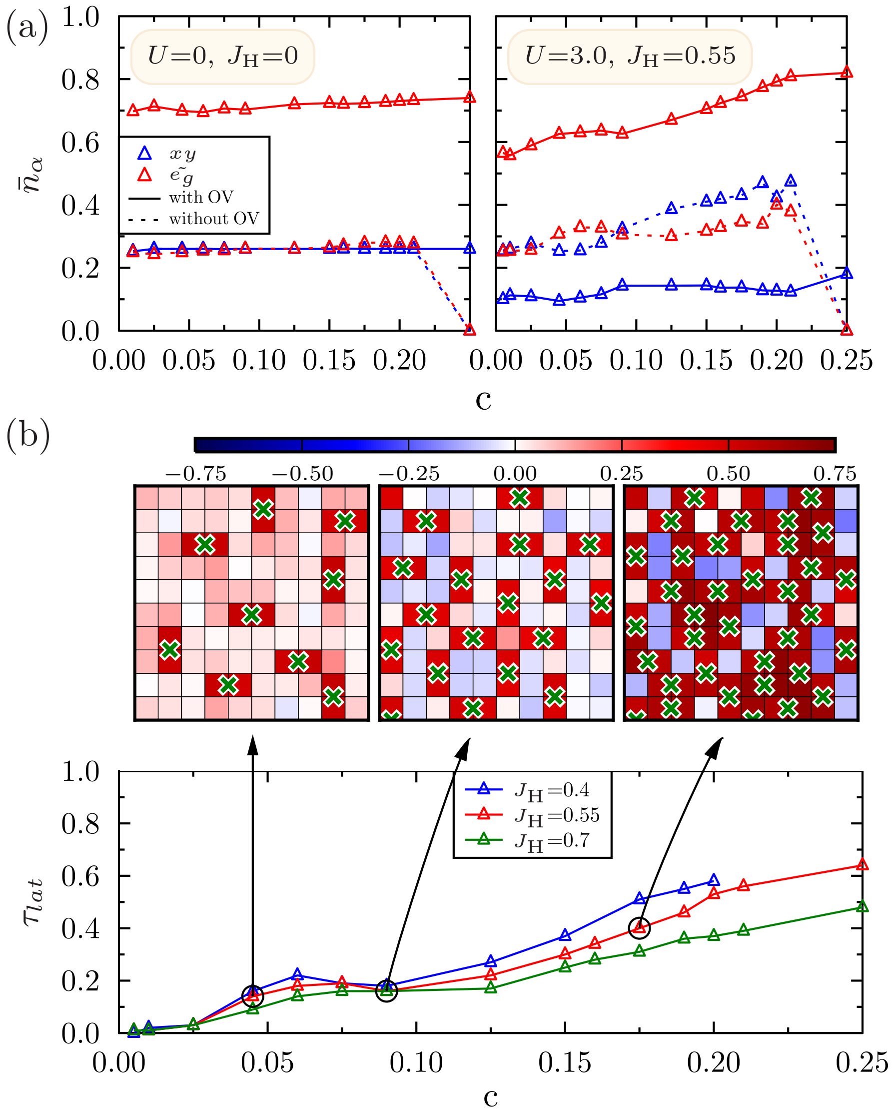
orbital occupation and orbital moment



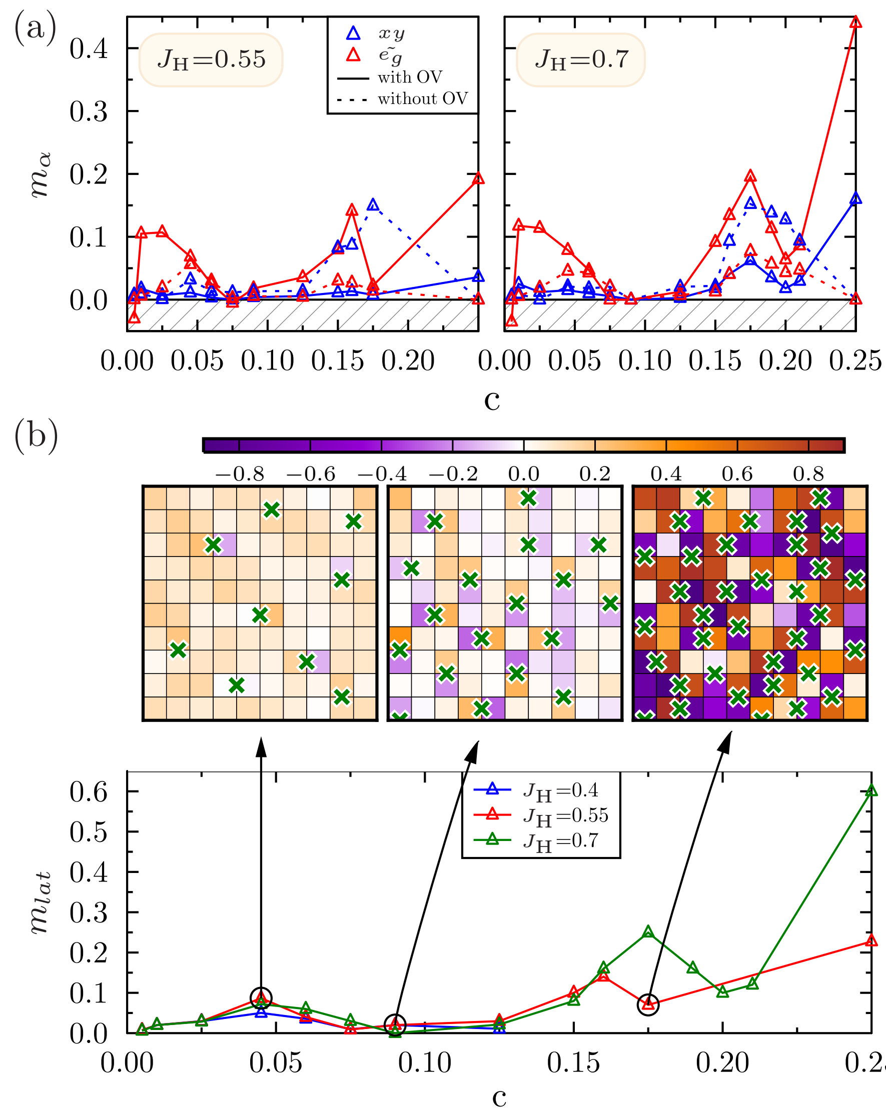
- intricate dichotomy between Ti sites with and without nearby oxygen vacancies

Arbitrary OV Concentration in LAO/STO : orbital and spin polarization

orbital occupation and orbital moment



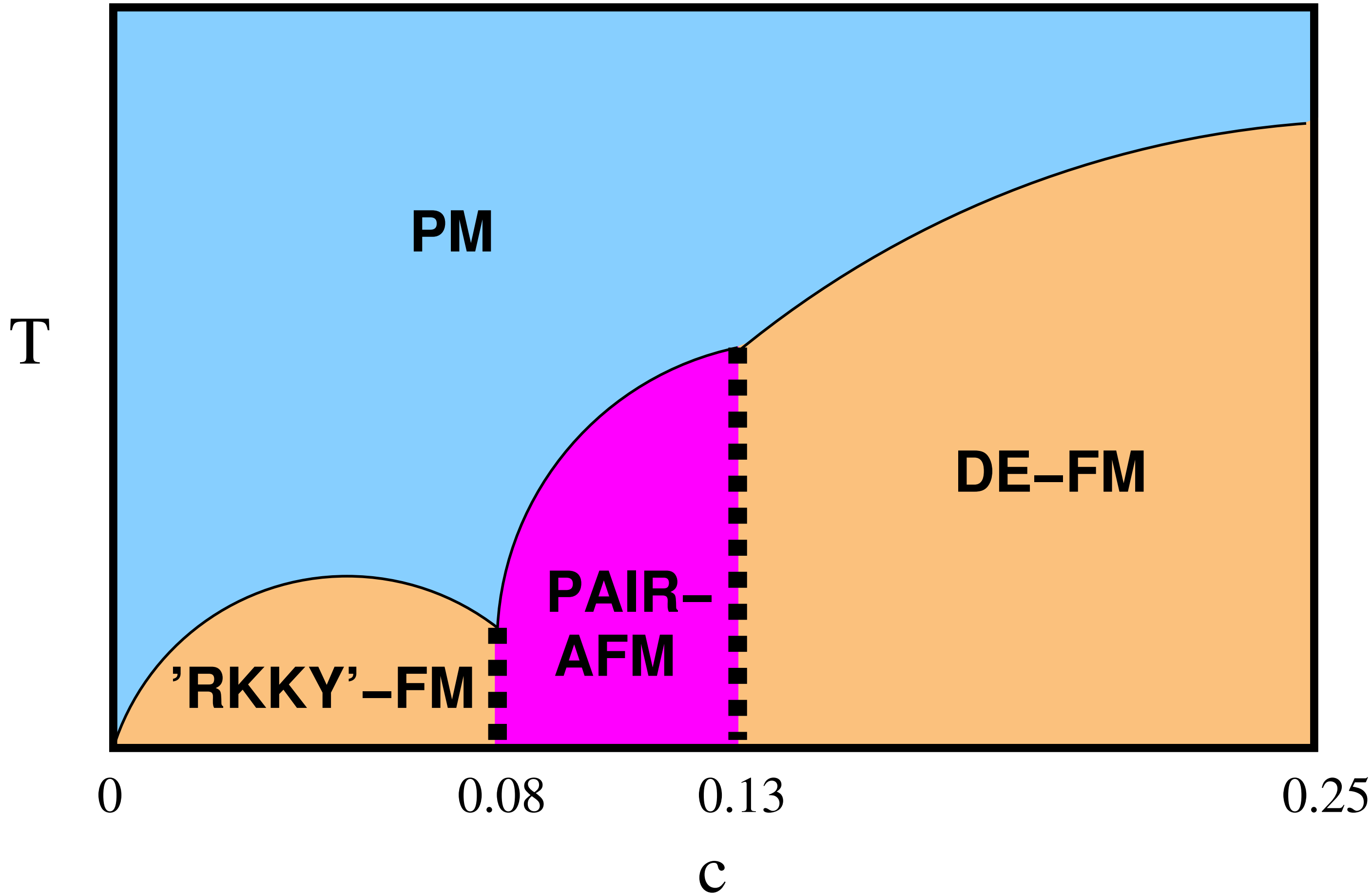
magnetic moment



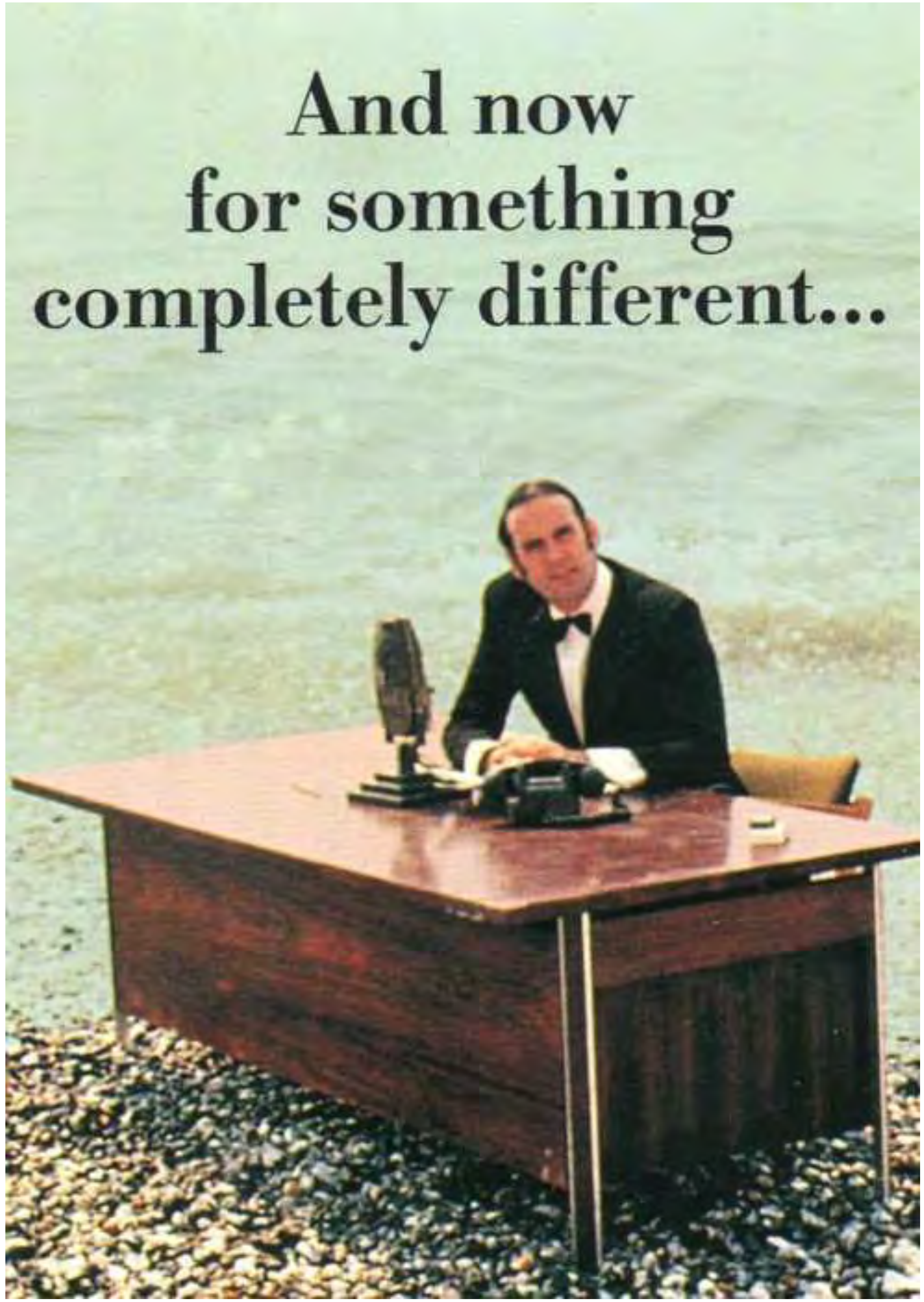
- intricate dichotomy between Ti sites with and without nearby oxygen vacancies

Arbitrary OV Concentration in LAO/STO : Magnetic Phase Diagram

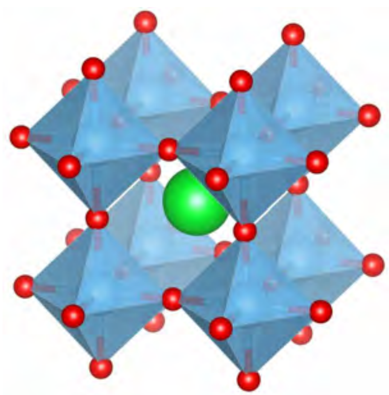
[Behrmann and FL, PRB 92, 125148 (2015)]

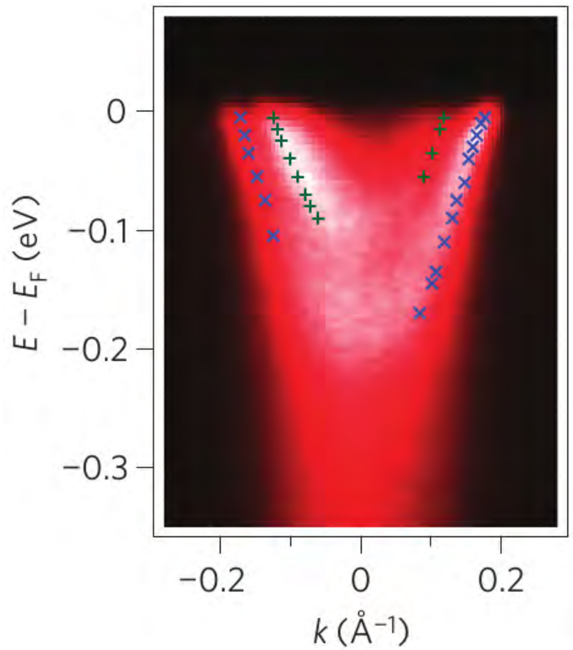


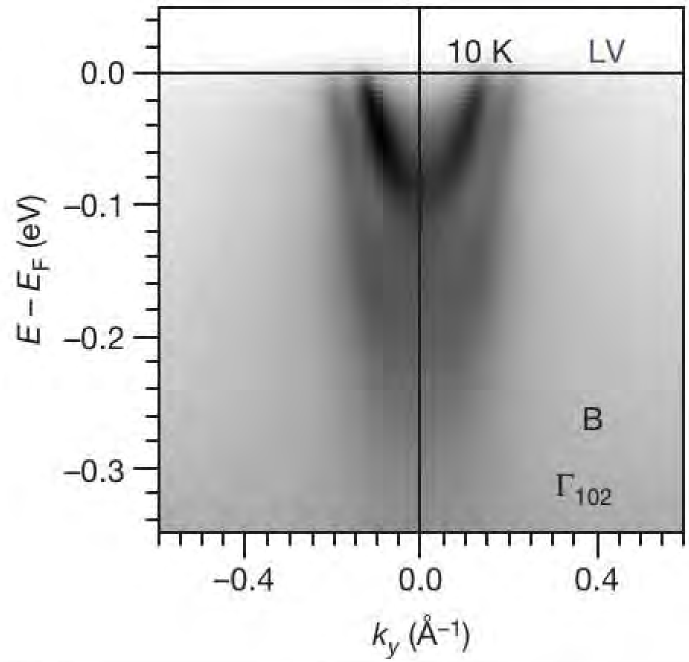
- nontrivial ferrmomagnetism triggered by even few oxygen vacancies
- double-exchange-like ferromagnetism for higher oxygen vacancy concentration


And now
for something
completely different...

Oxygen-Deficient $SrTiO_3$ surface : Experiment







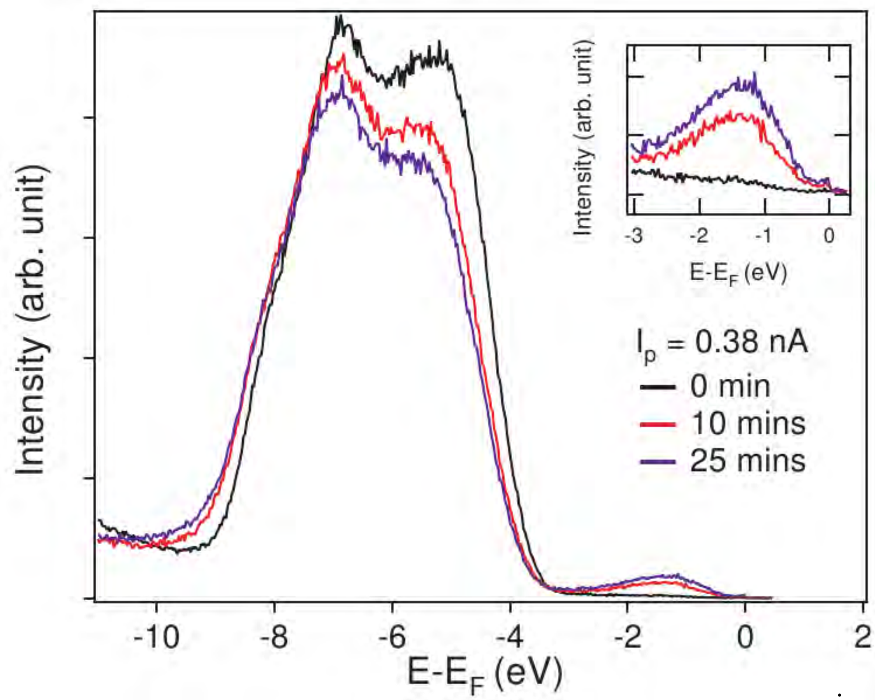
two-dimensional electron system on surface via

- exposure to UV light

[Meevasana et al., Nature Mat. 10, 114 (2011)]

- in-situ crystal fracture

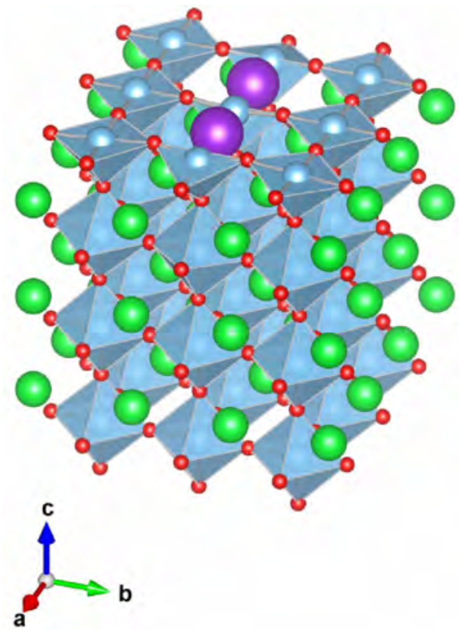
[Santander-Syro et al., Nature 469, 189 (2011)]

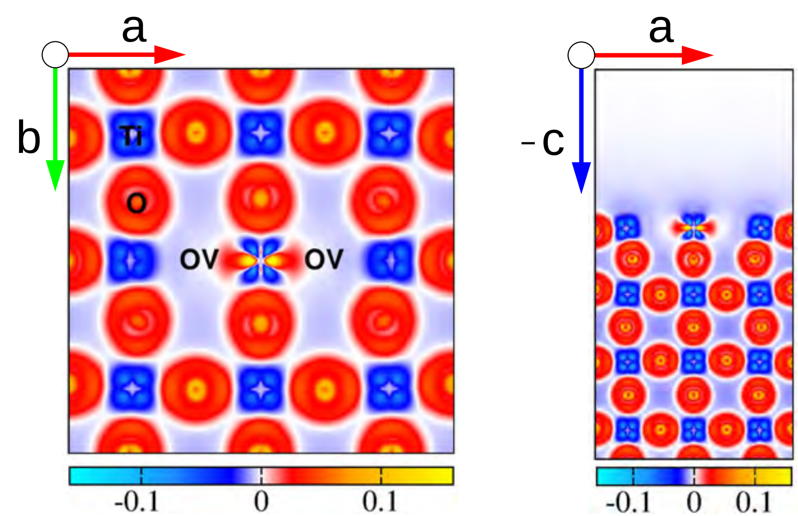


Oxygen-Deficient $SrTiO_3$ surface : DFT+DMFT

[FL, Jeschke, Kim, Backes and Valentí, PRB 93, 121103(R) (2016)]

- ⋆ 180-atom slab
- ⋆ two oxygen vacancies in TiO_2 surface layer
- ⋆ projections allow for e_g and t_{2g} states
- ⋆ U=3.5eV, $J_{\rm H}$=0.5eV





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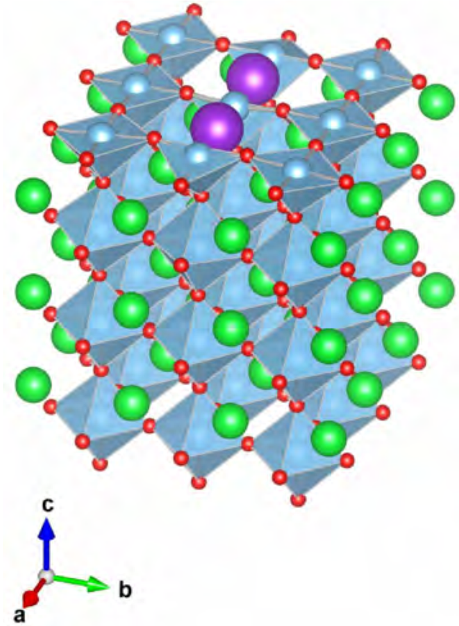
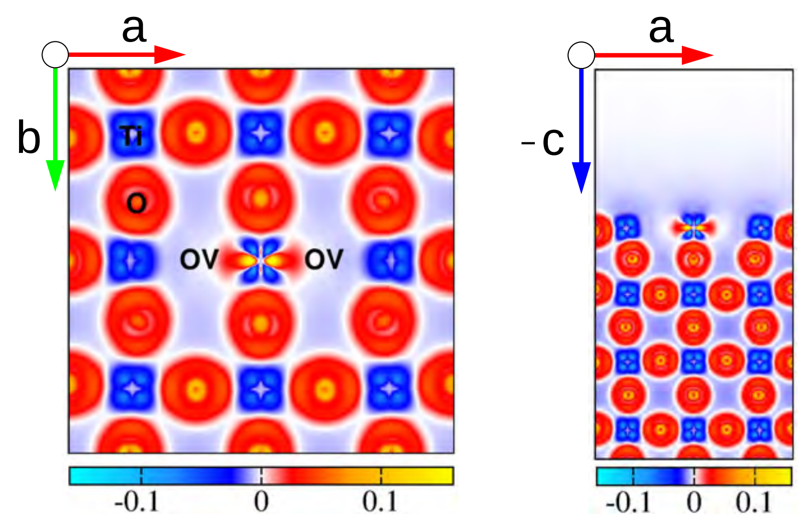
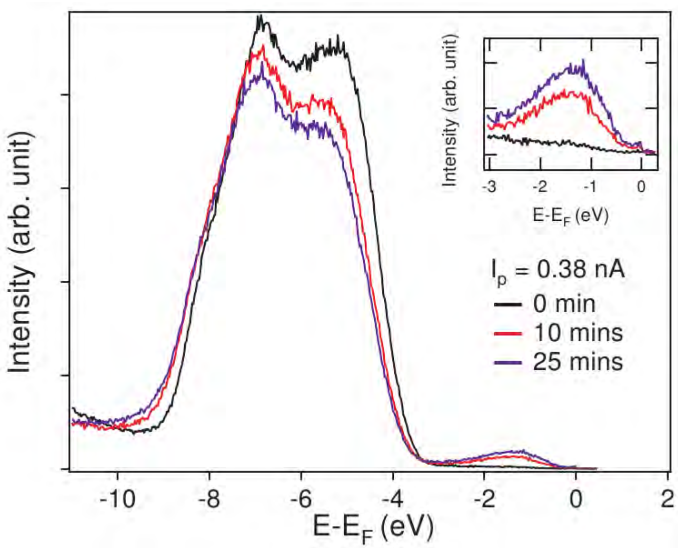
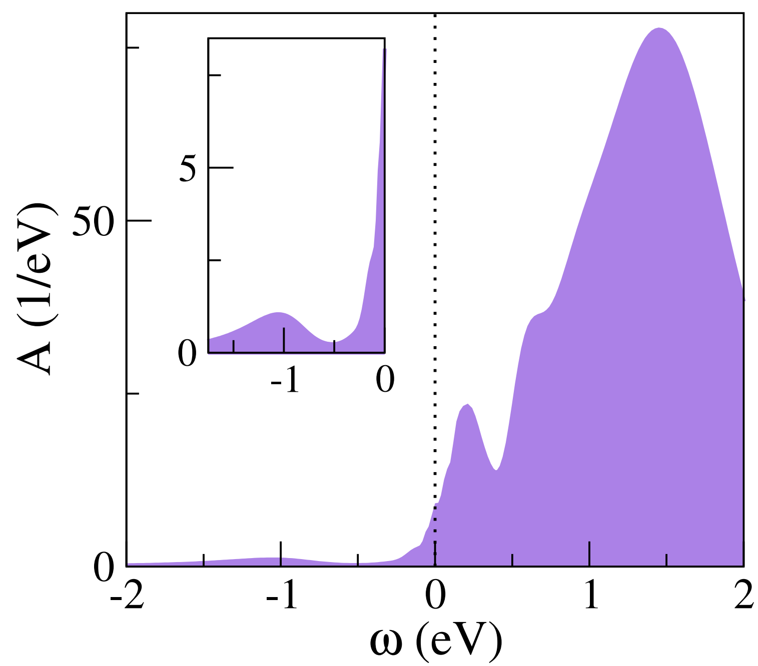






[Meevasana et al., Nature Mat. 10, 114 (2011)]



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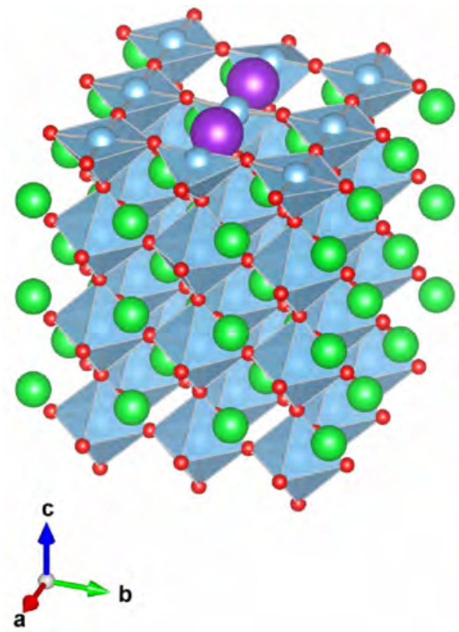
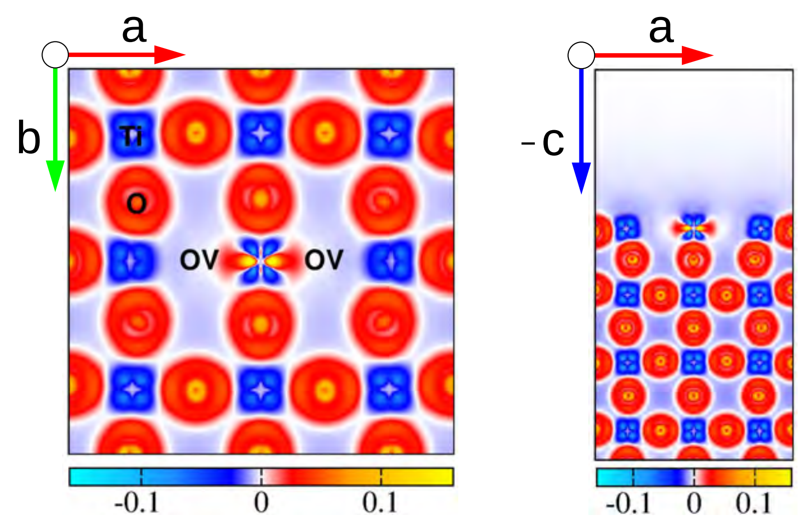
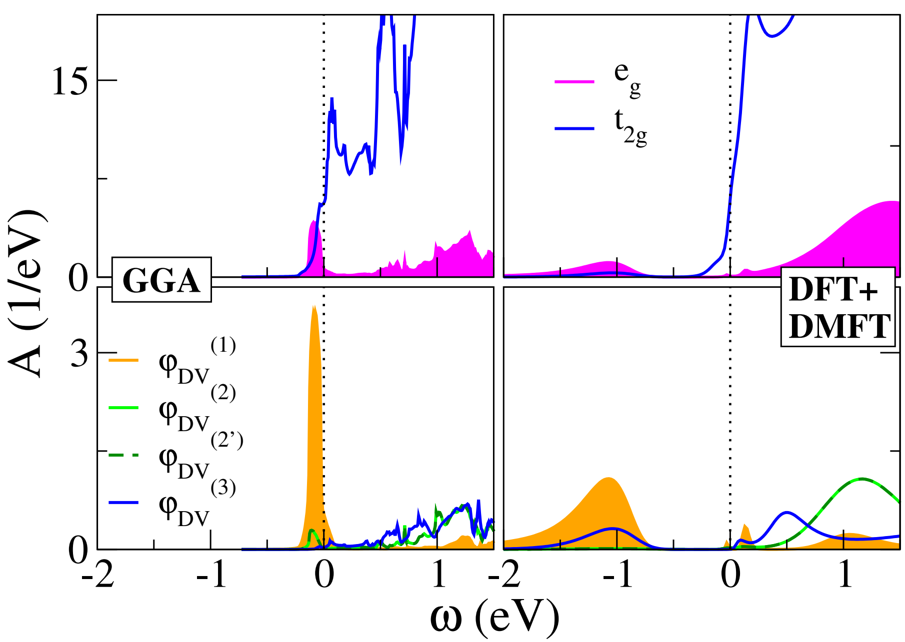
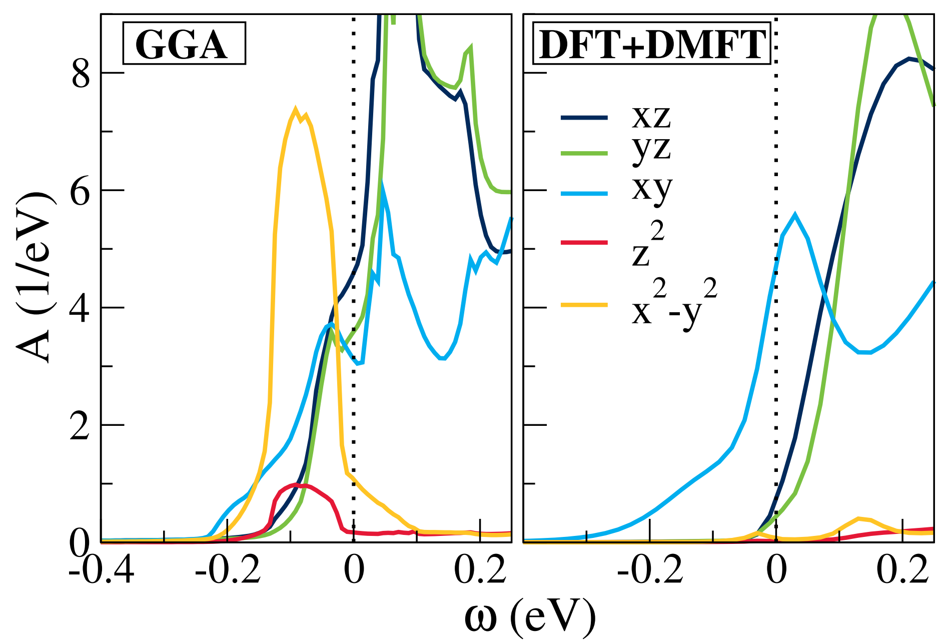




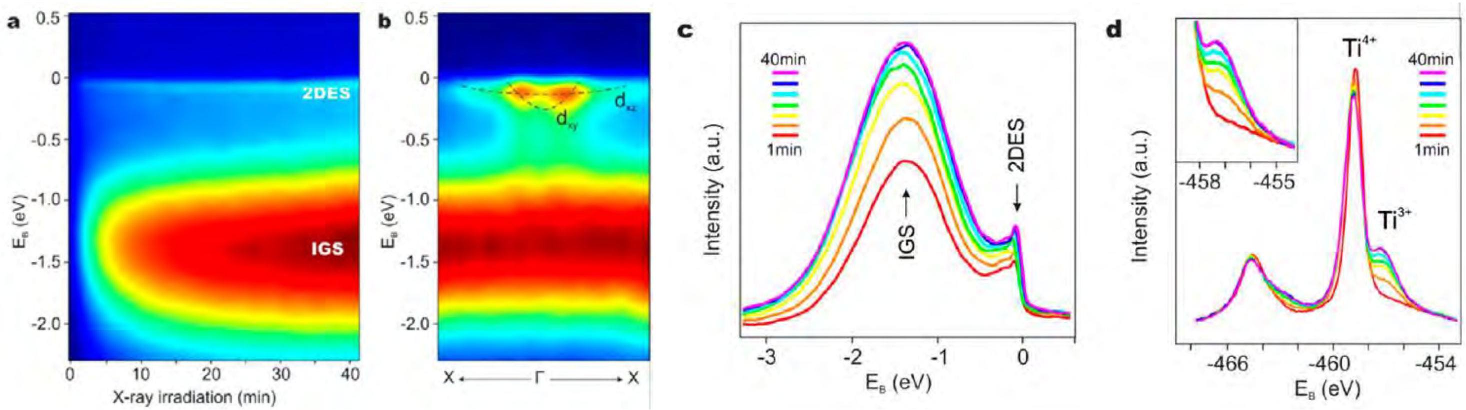




$LaAlO_3/SrTiO_3$ revisited : experiment and DFT+DMFT

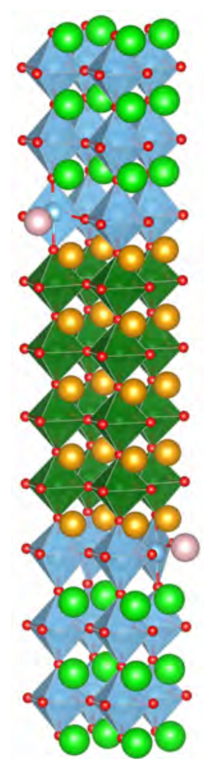
[FL, Boehnke, Grieger and Piefke, PRB 90, 085125 (2014)] [Chikina, FL, Husanu et al., ACS Nano 12, 7927 (2018)]

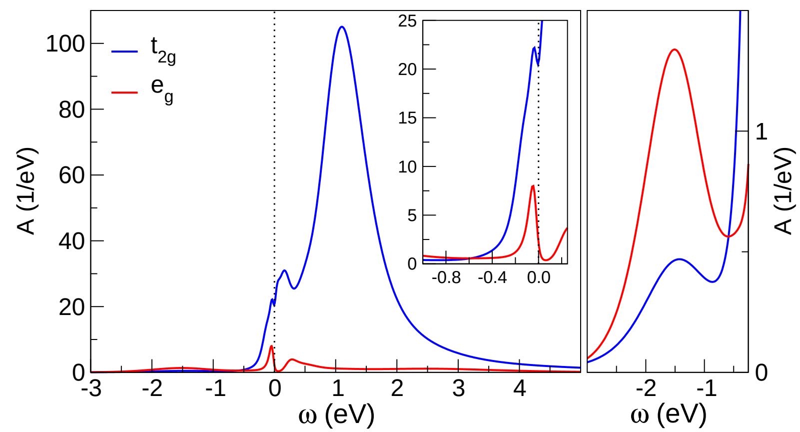
experimental spectrum

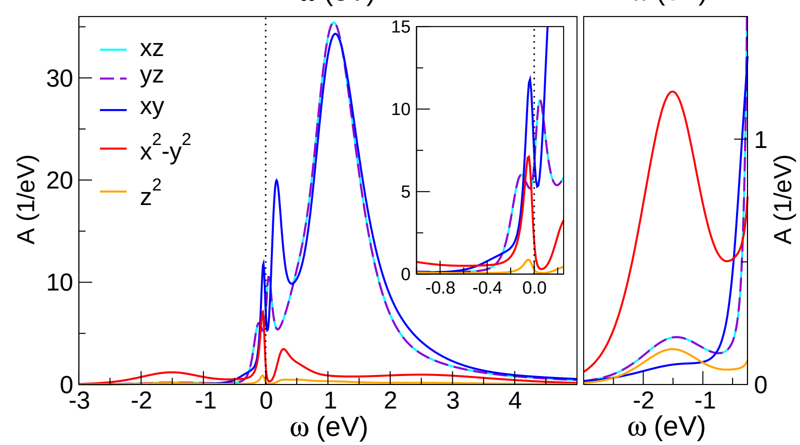


theoretical description

- 200-atom supercell
- Hubbard $U = 3\,\text{eV}$,
  Hund's exchange $J_{\text{H}} = 0.5\,\text{eV}$



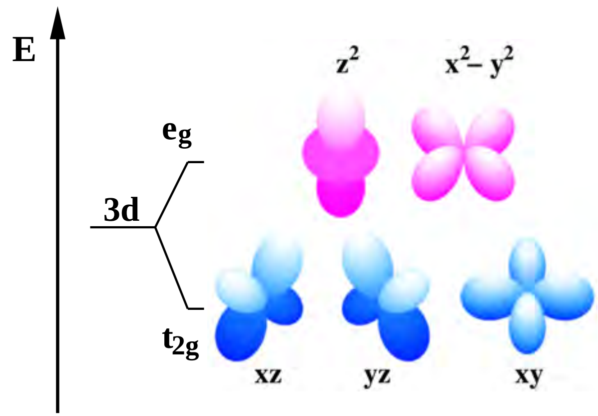


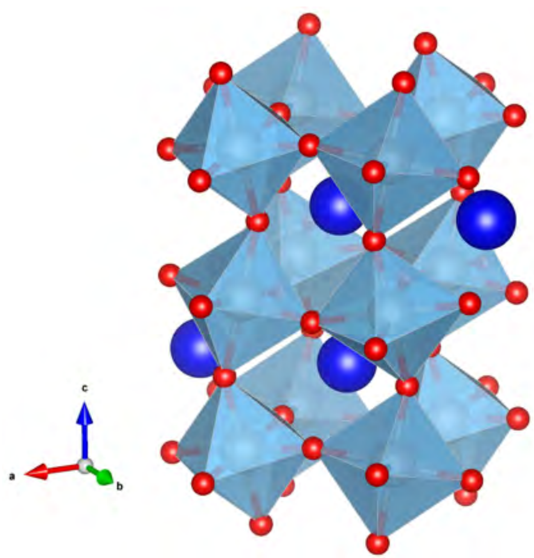


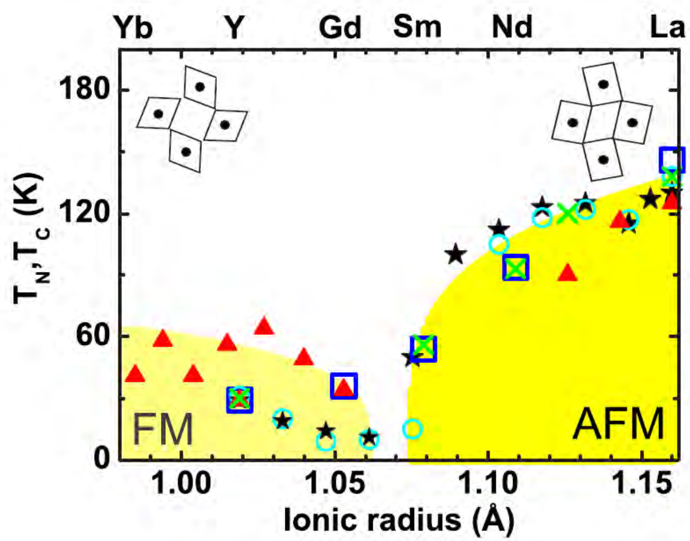
Doping the Mott-Insulating State : Phenomenology of Bulk Titanates

Mott-insulating distorted perovskites $R\mathrm{TiO}_3$

$\rightarrow$ Ti^{3+} with $3d^1(t_{2g})$







[Komarek et al., PRB 75, 224402 (2007)]

LaTiO_3	AFM T_{N}=146K, apical tilt Θ=13°, charge gap Δ=0.20 eV
GdTiO_3	FM T_{C}=36K, apical tilt Θ=18°, charge gap Δ=0.75 eV
SmTiO_3	AFM T_{N}=48K, apical tilt Θ=17°, charge gap Δ=0.50 eV

[Crandles at al., Physica C, 407 (1992)] [Okimoto et al., PRB 95, 9581 (1995)]

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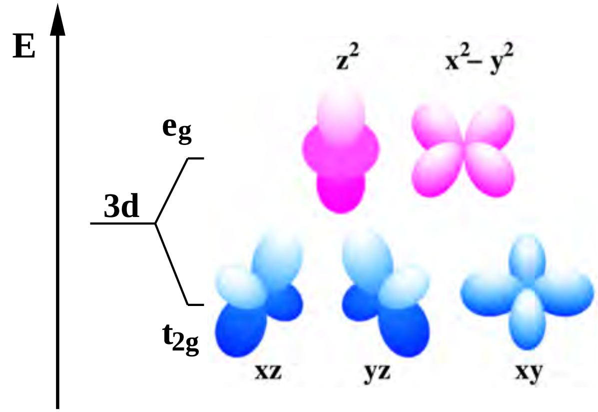

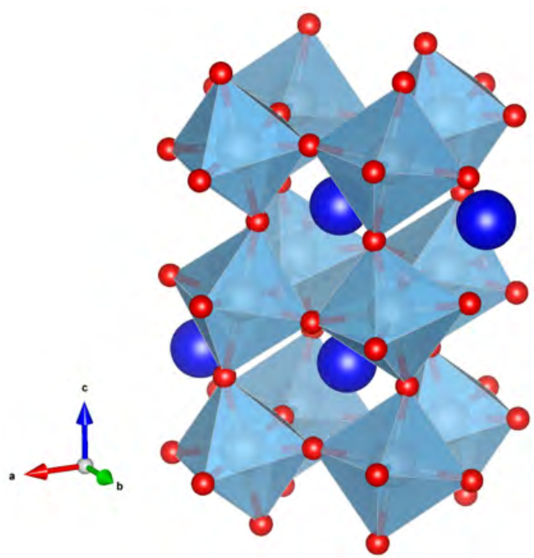

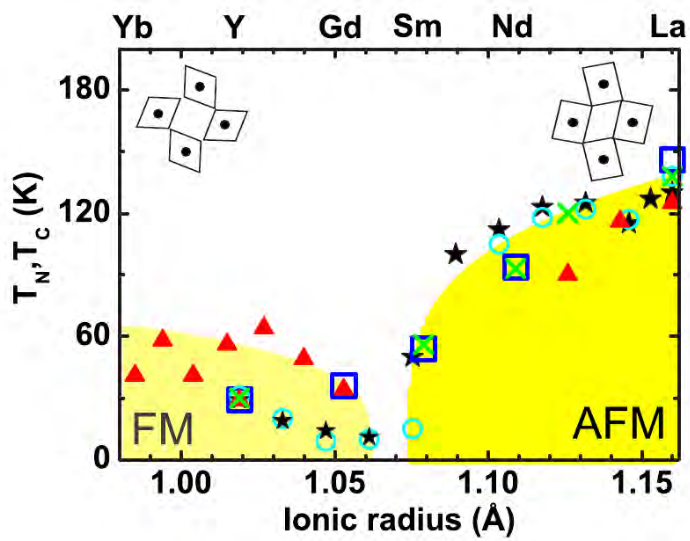

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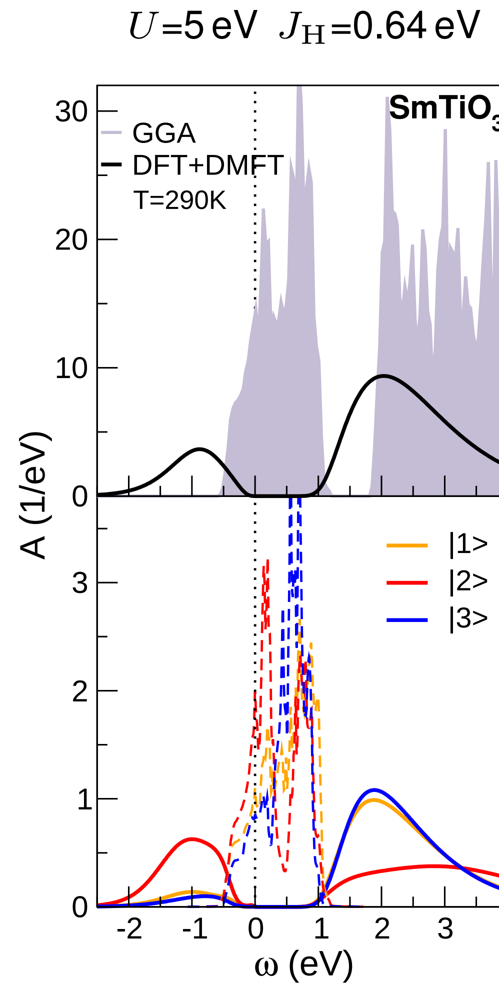

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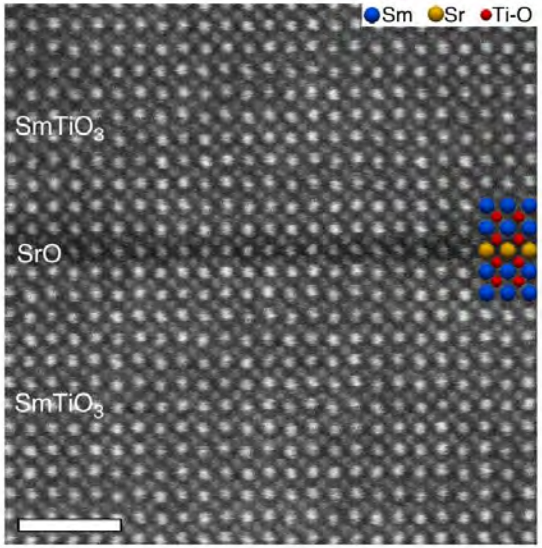
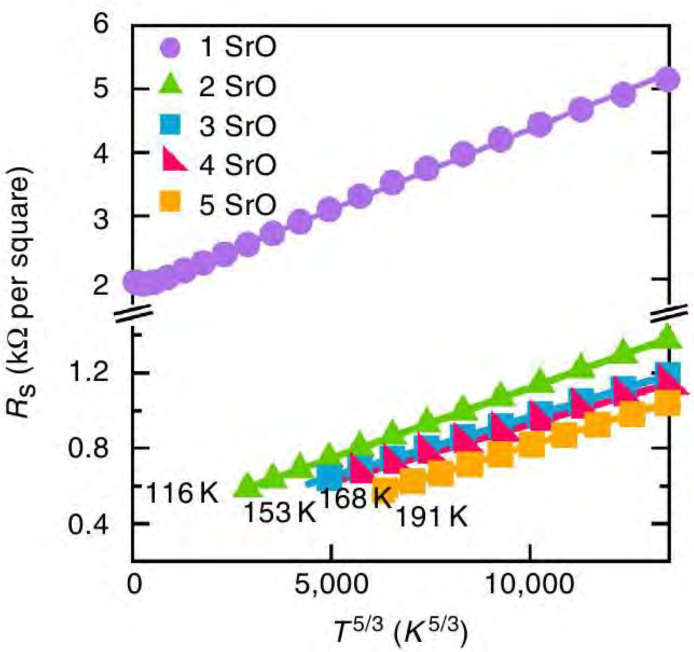
SmTiO_3 is close to an AFM-to-FM quantum phase transition

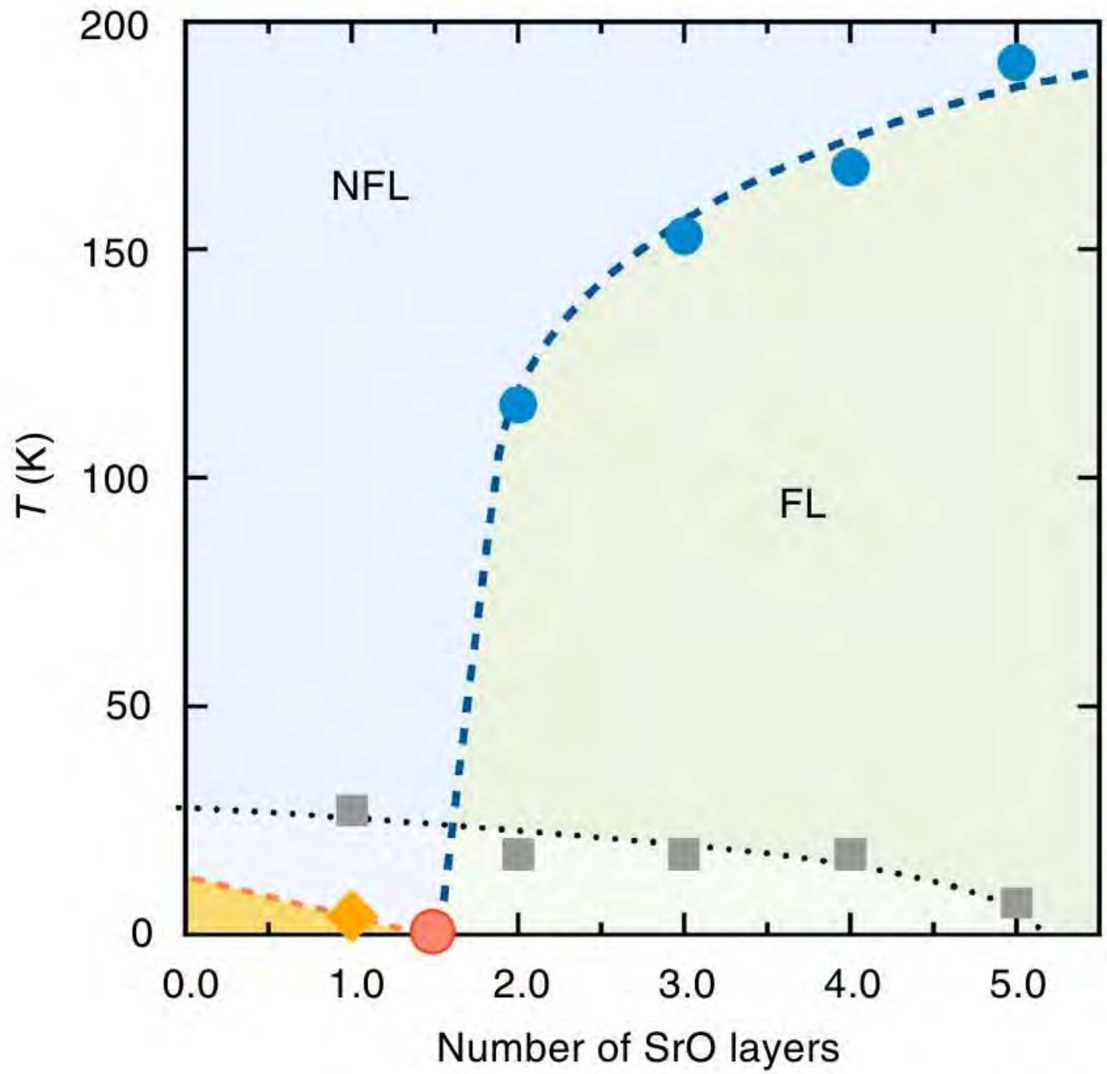
δ-doped $SmTiO_3$: Experimental Non-Fermi-Liquid Behavior

[Jackson et al., Nat. Commun. 5, 4258 (2014)]

well-defined doping with layers of SrO through molecular-beam epitaxy



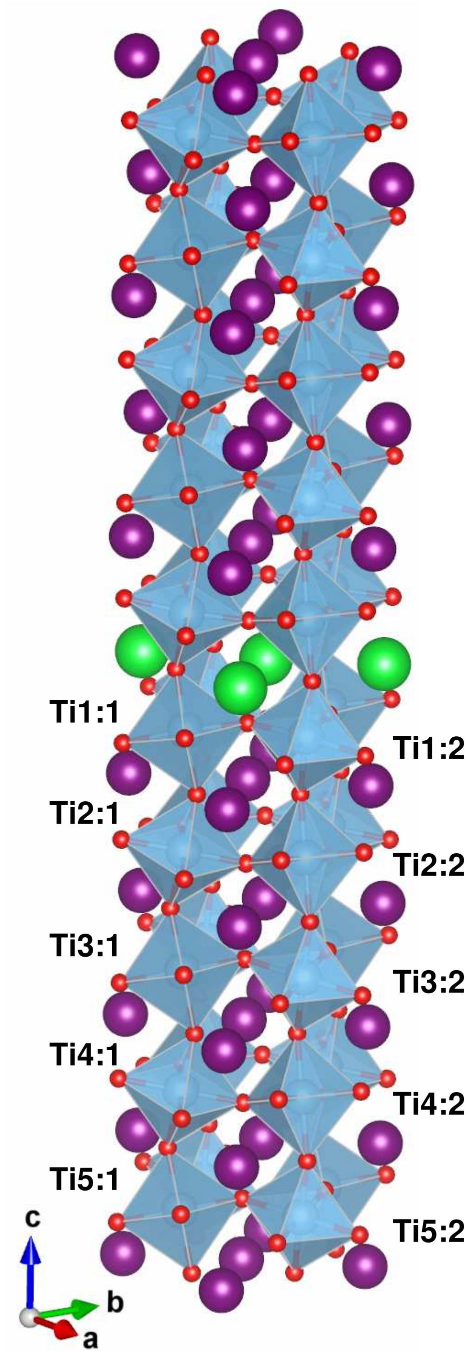


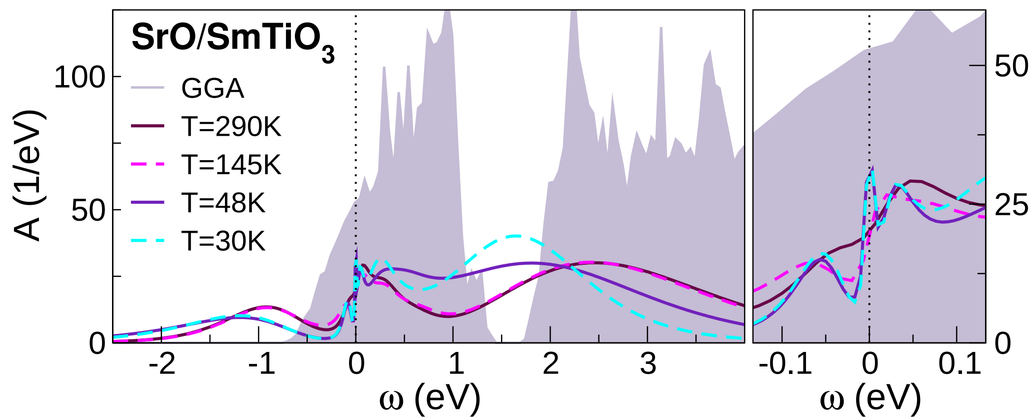


$T^{5/3}$-law for the resistivity

δ-doped $SmTiO_3$: Spectral Function from DFT+DMFT

[FL, Sci. Rep. 7, 1565 (2017)]





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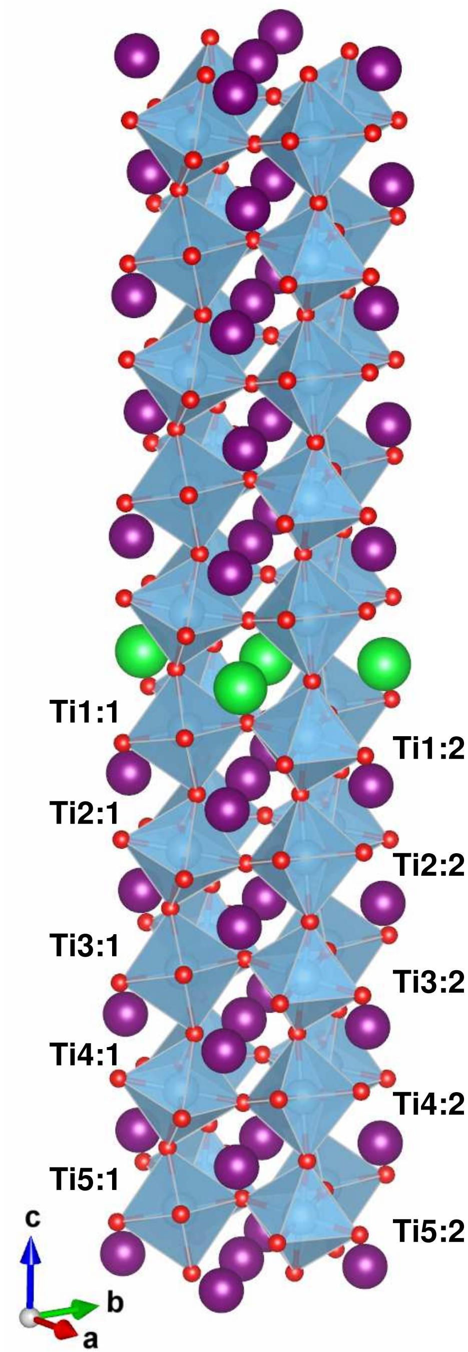
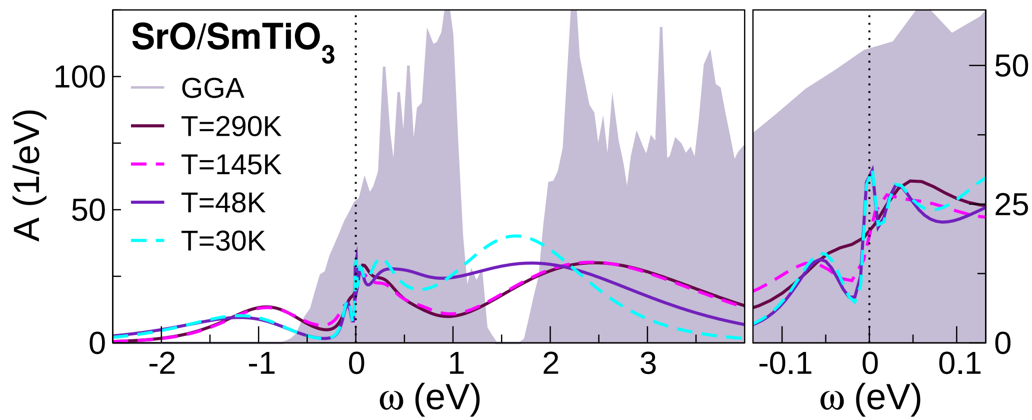
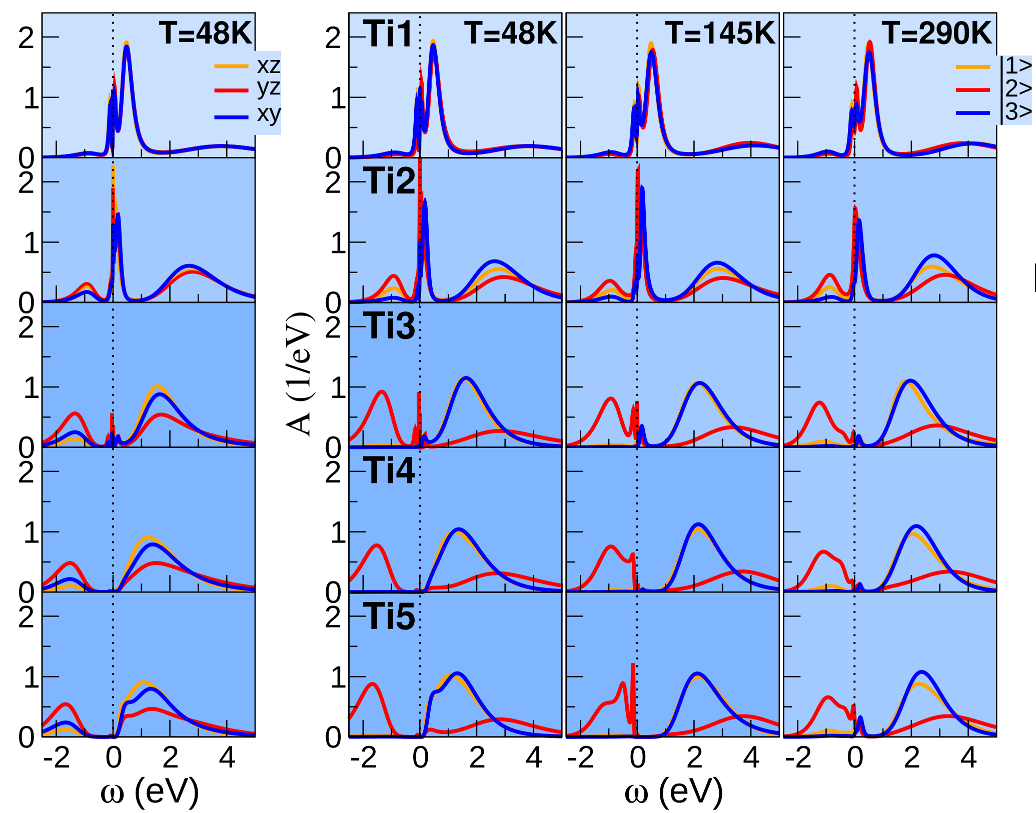


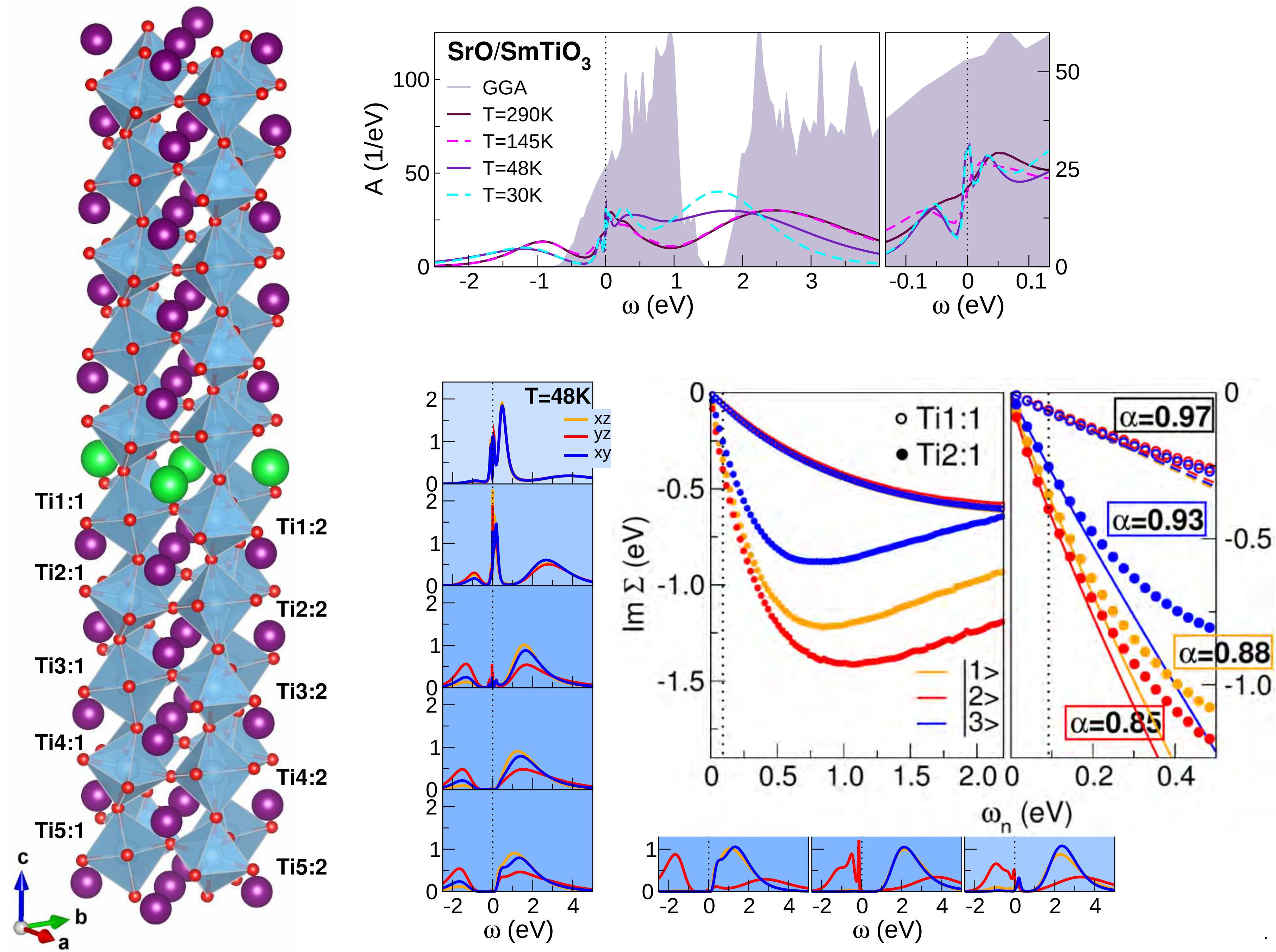




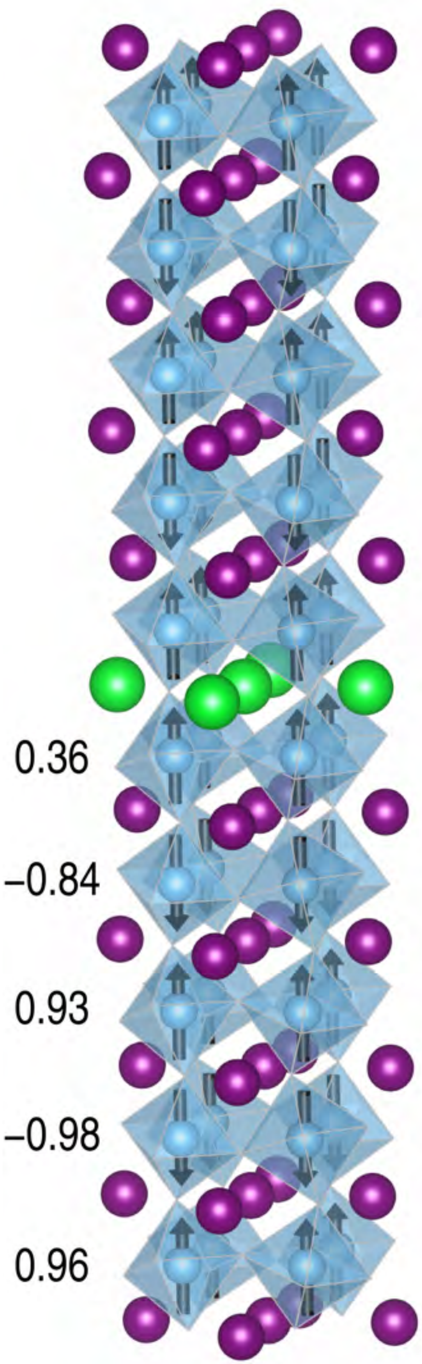
layer-dependent
multi-orbital
Mott transition

δ-doped $SmTiO_3$: Spectral Function from DFT+DMFT

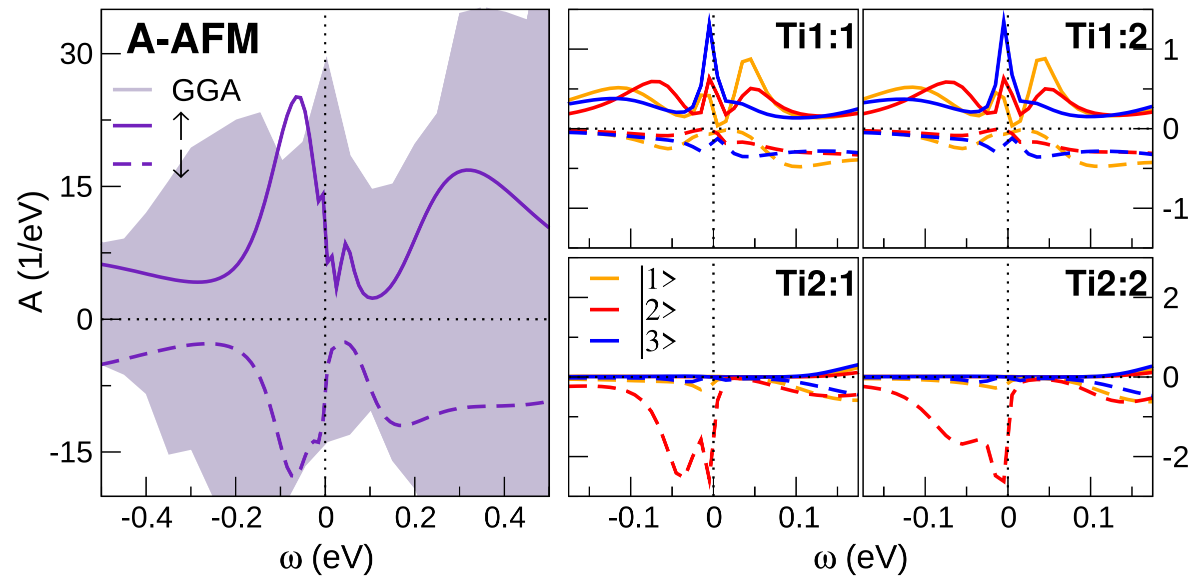
[FL, Sci. Rep. 7, 1565 (2017)]



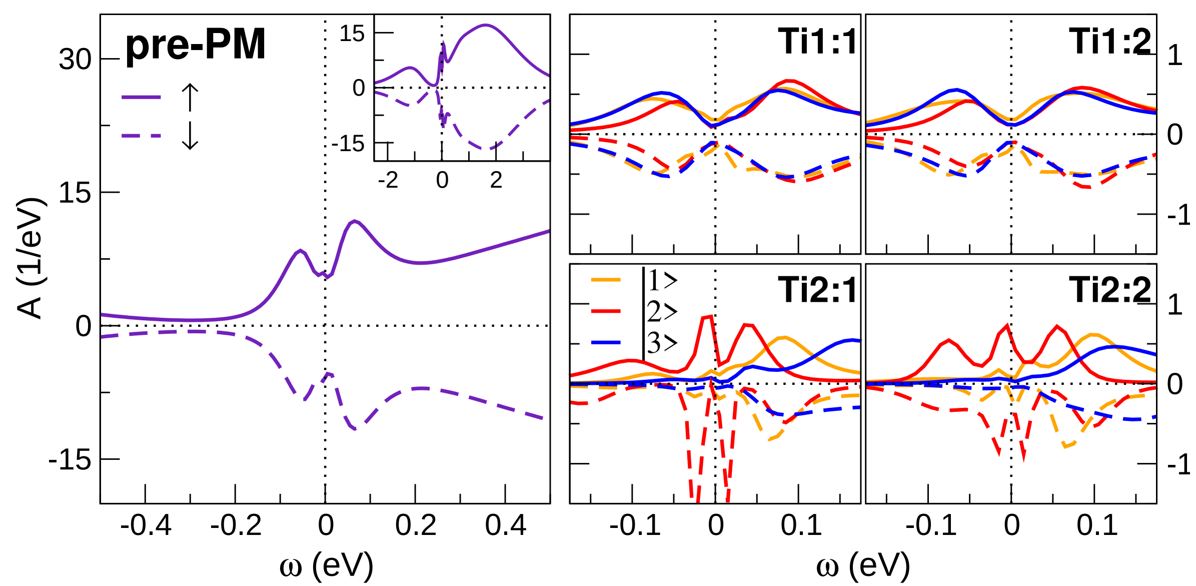
δ-doped $SmTiO_3$: Magnetic Order and Pseudogap Fingerprint



A-type AFM phase



pre-converged PM phase with pseudogap signature

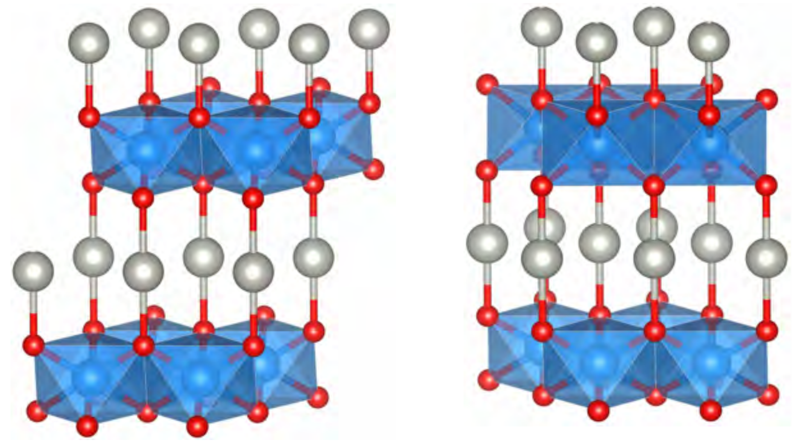


Delafossites: Introduction

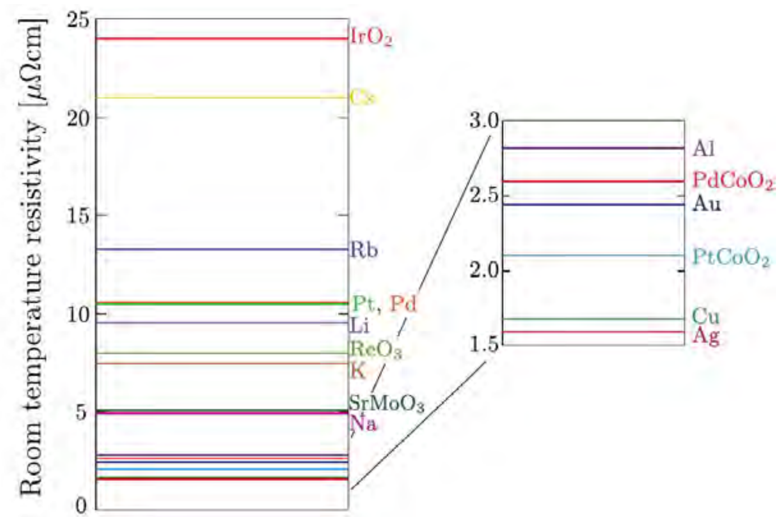
[Mackenzie, Rep. Prog. Phys. 80, 032501 (2017)]
[FL, npj Comput Mater 7, 120 (2021)]

- natural oxide heterostructure ABO_2, with metallic A^+ ions and metallic B^{3+} ions
- alternating A layers and BO_2 layers
- A layer forms triangular lattice
- unique dumbbell O-A-O structure of connecting layers
- two stackings
  $R\bar{3}m$: e.g. $PdCoO_2$, $CuCrO_2$, etc. (most)
  $2H$: e.g. $AgNiO_2$
- wide variety of compounds, insulators and metals,
  with metals among highest conducting materials!

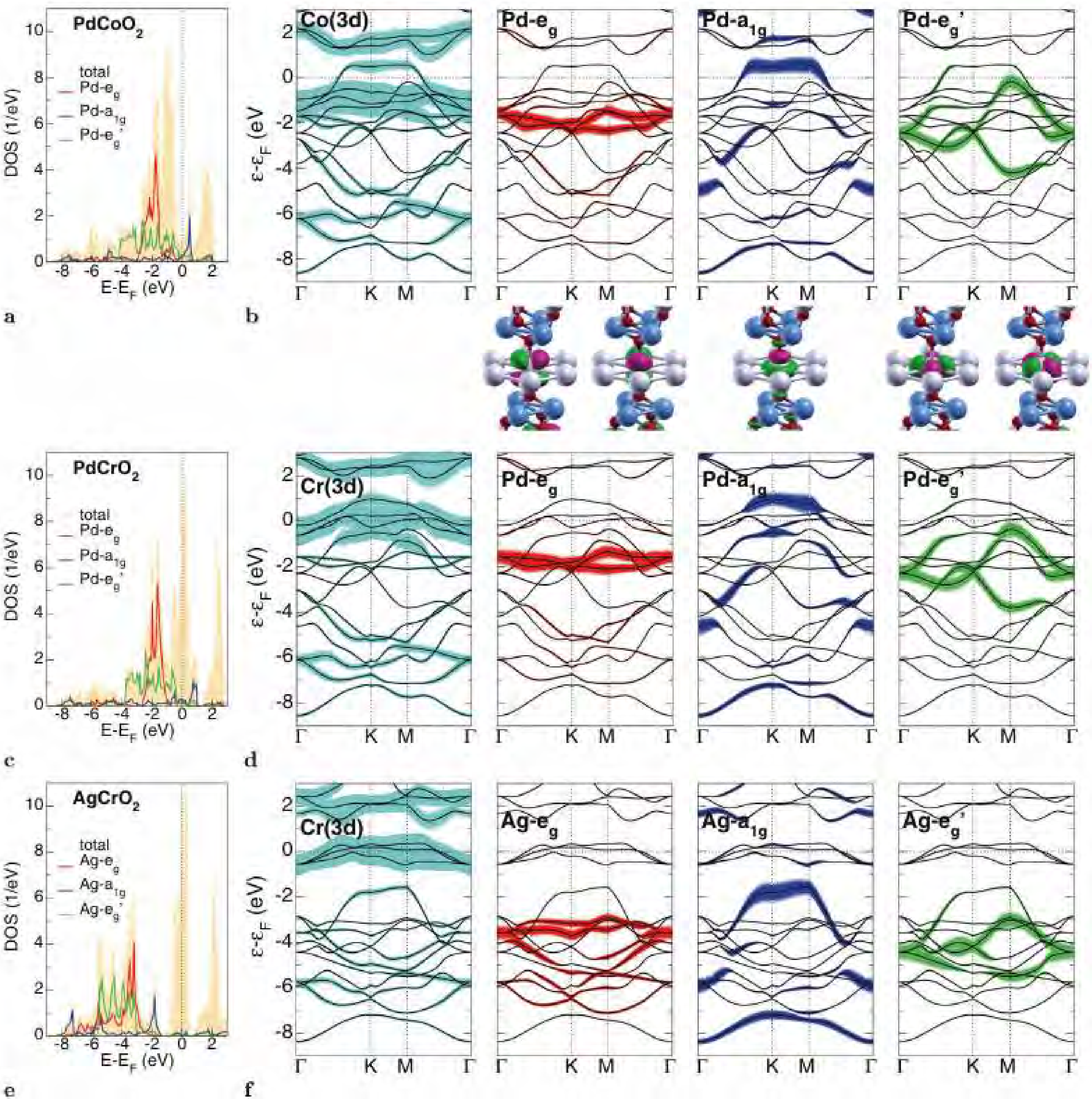
→ playground to design correlation physics



$R\bar{3}m$ $2H$

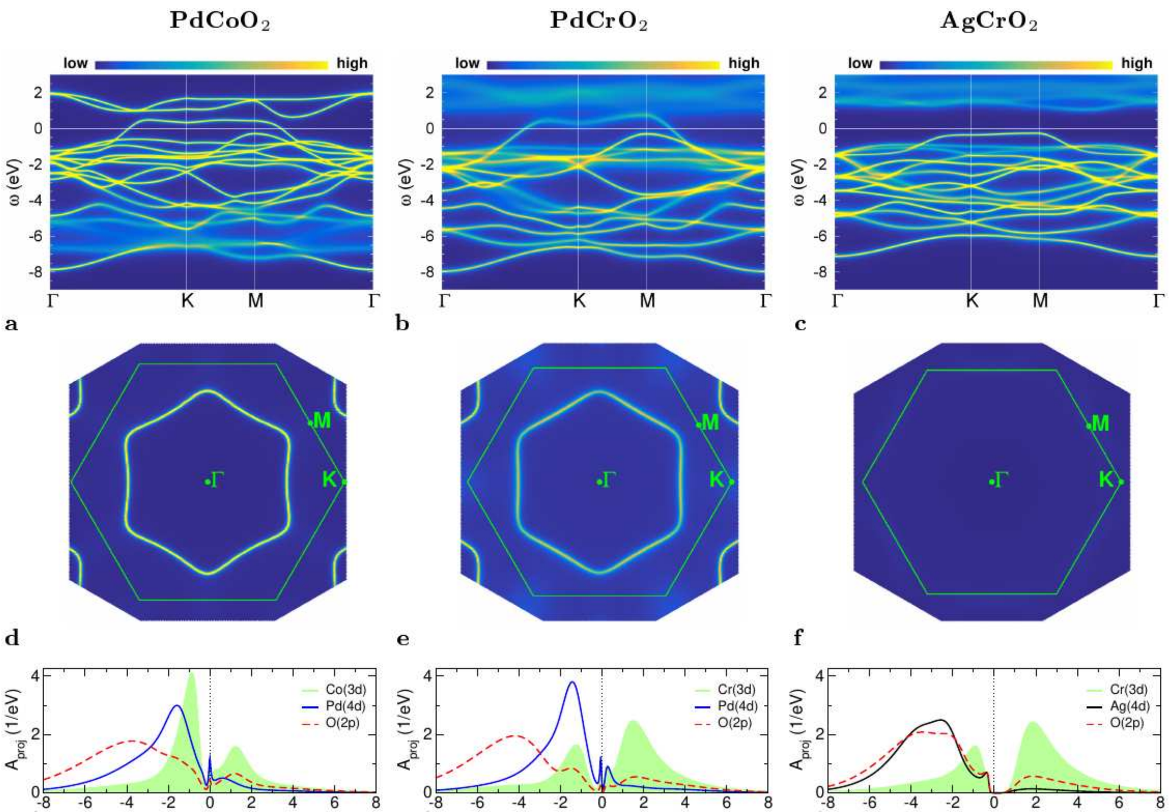


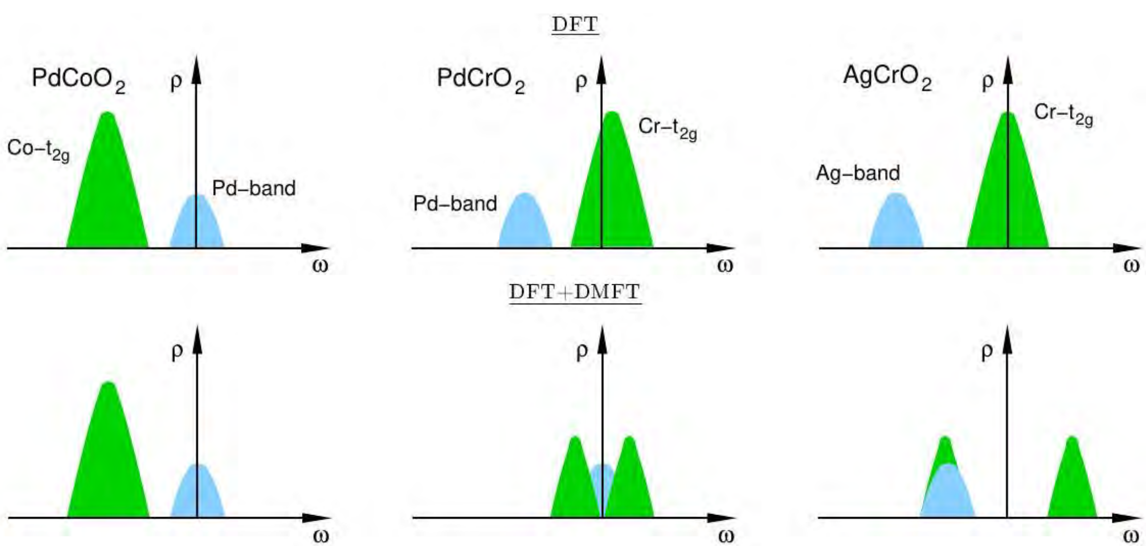
Delafossites: DFT picture



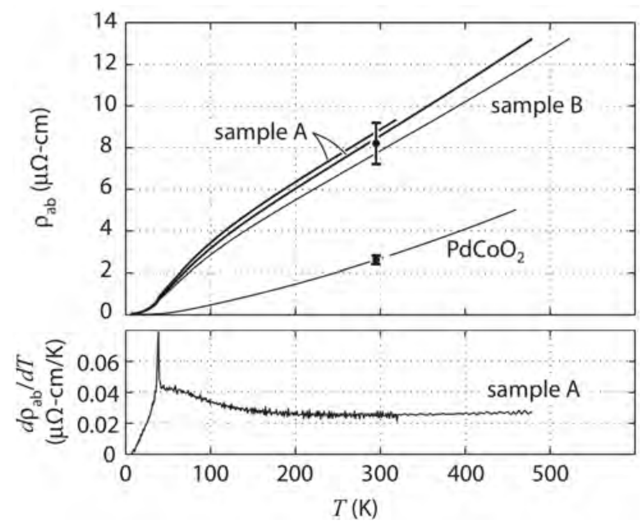
Delafossites: Correlation Scenarios

[FL, npj Comput Mater 7, 120 (2021)]





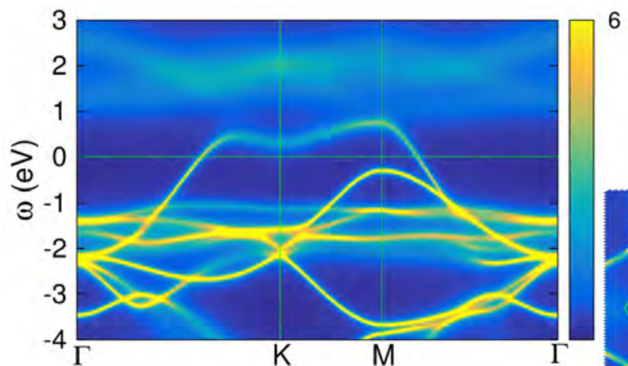
U(Co,Cr)$= 3 - 4$ eV,
$J_{\rm H} = 0.7$ eV

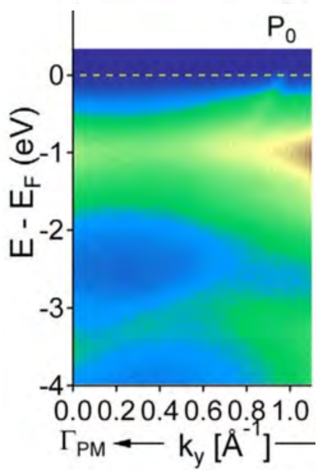


[Hicks et al, PRB 92, 014425 (2015)]

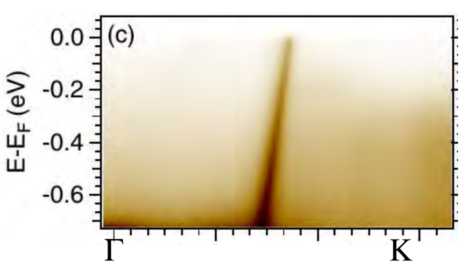
Delafossites: $PdCrO_2$ Correlated Electronic Structure

ARPES comparison in PM phase

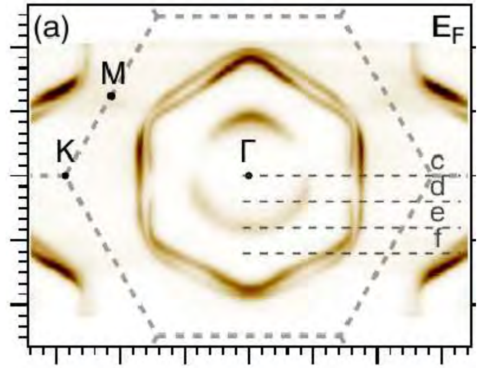


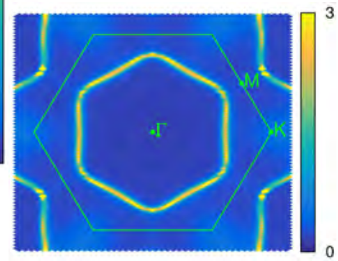


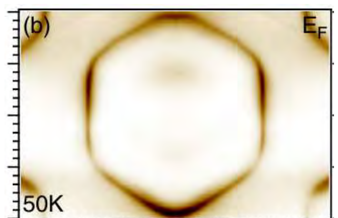
100 K [Noh et al., Sci. Rep. 4, 3680 (2014)]

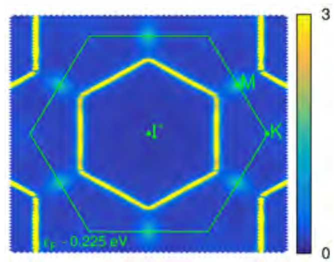


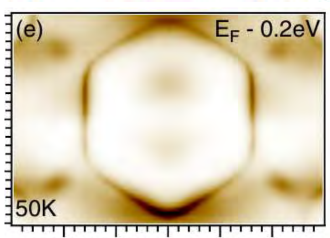
10 K [Sobota et al., PRB 88, 125109 (2013)]



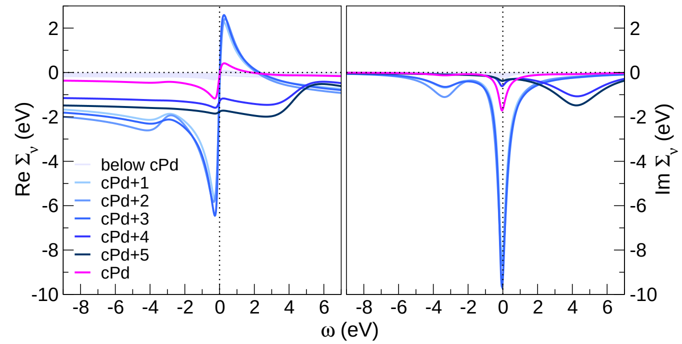








Coupling between itinerant and localized electrons



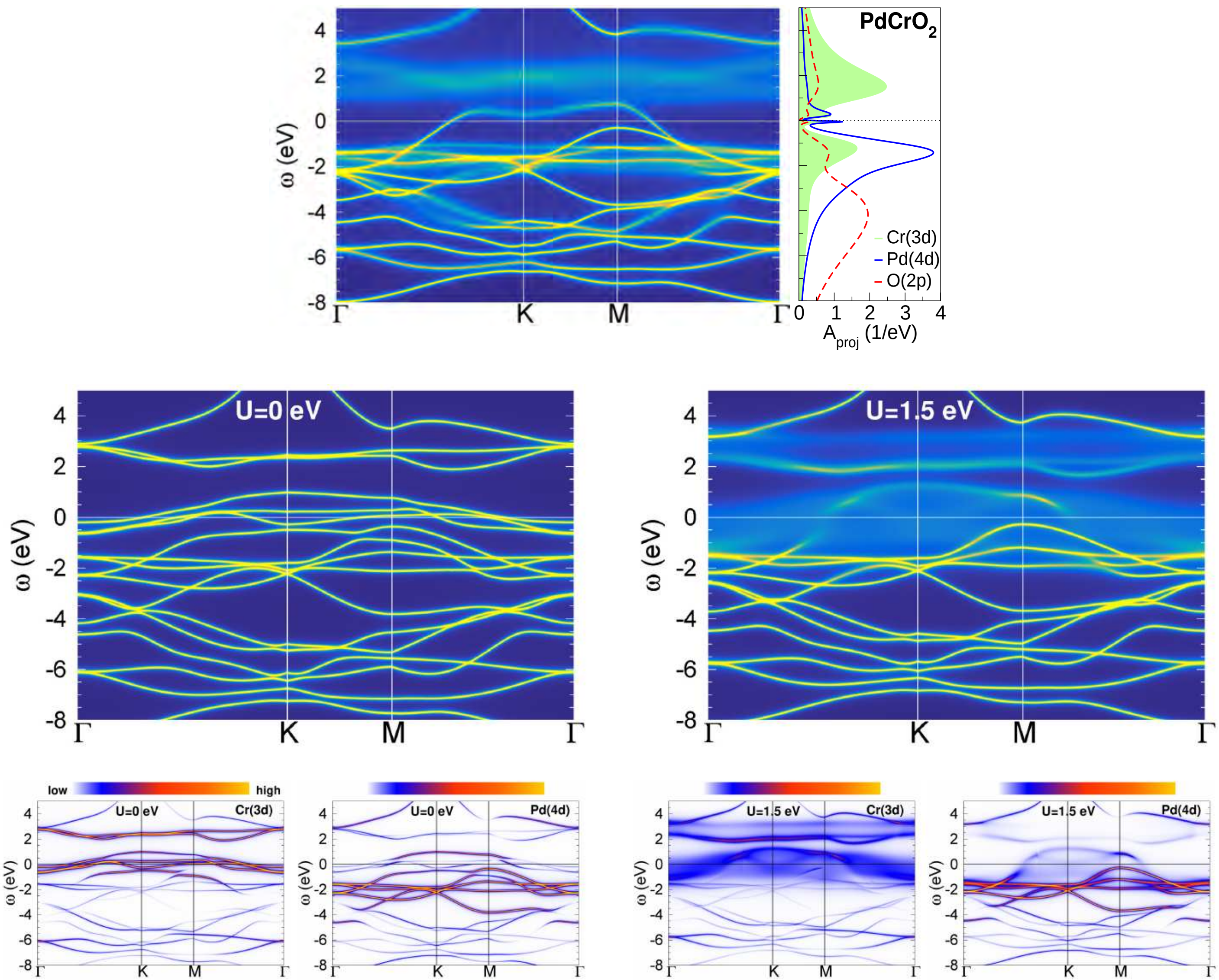
- from upfolded Cr self energy

$$\Sigma_{\nu\nu'}(\mathbf{k},\omega) = \sum_{\mathbf{R},mm'} \bar{P}^{\mathbf{R}*}_{\nu m}(\mathbf{k})\tilde{\Sigma}^{\mathbf{R}}_{mm'}(\omega)\bar{P}^{\mathbf{R}}_{m'\nu'}(\mathbf{k})$$

- significant self-energy contribution to conducting Pd band (no Hubbard interactions on Pd)
- relevant ee-scattering between Pd and CrO_2 layers
- Periodic Anderson model? Kondo model?

Delafossites: $PdCrO_2$ from weak to strong coupling

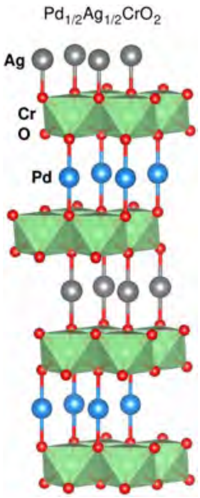
[FL, npj Comput Mater 7, 120 (2021)]

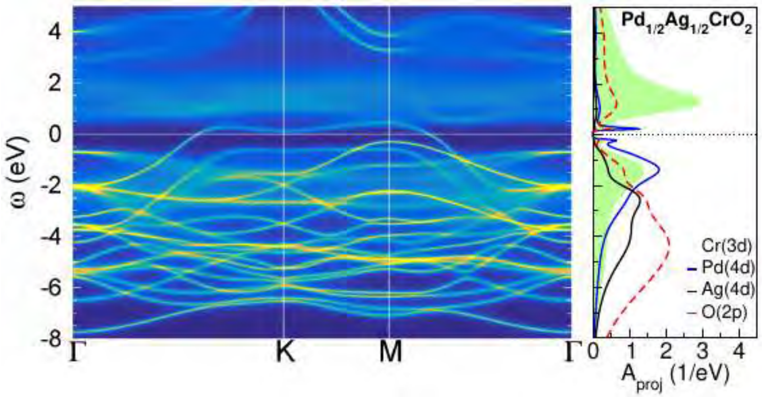


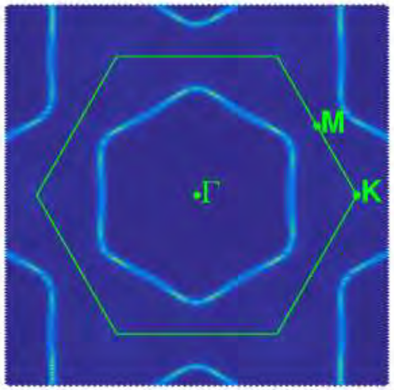
- metal-to-metal transition with growing interaction strength
- doping with isovalent Mo$(4d)$ for Cr$(3d)$?

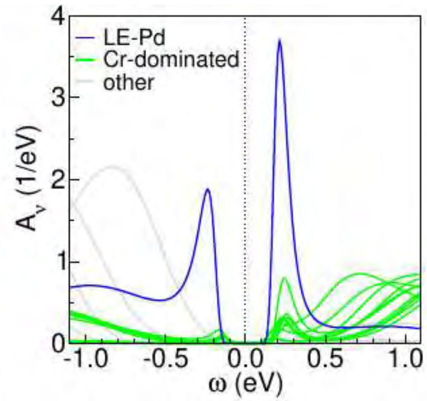
Delafossites: $PdCrO_2$ Mott Design I

[FL, npj Comput Mater 7, 120 (2021)]

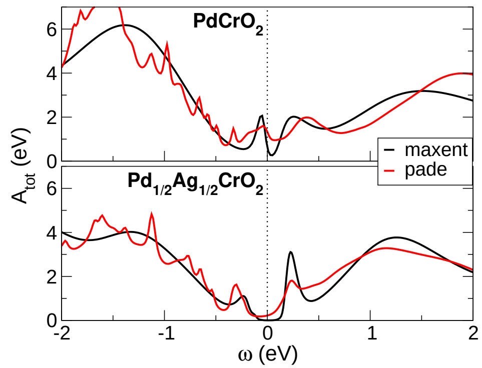








- prediction
  very weak dispersion crossing Fermi level
  → correlation-induced semimetal (CIS)



(no maxent artifact!)

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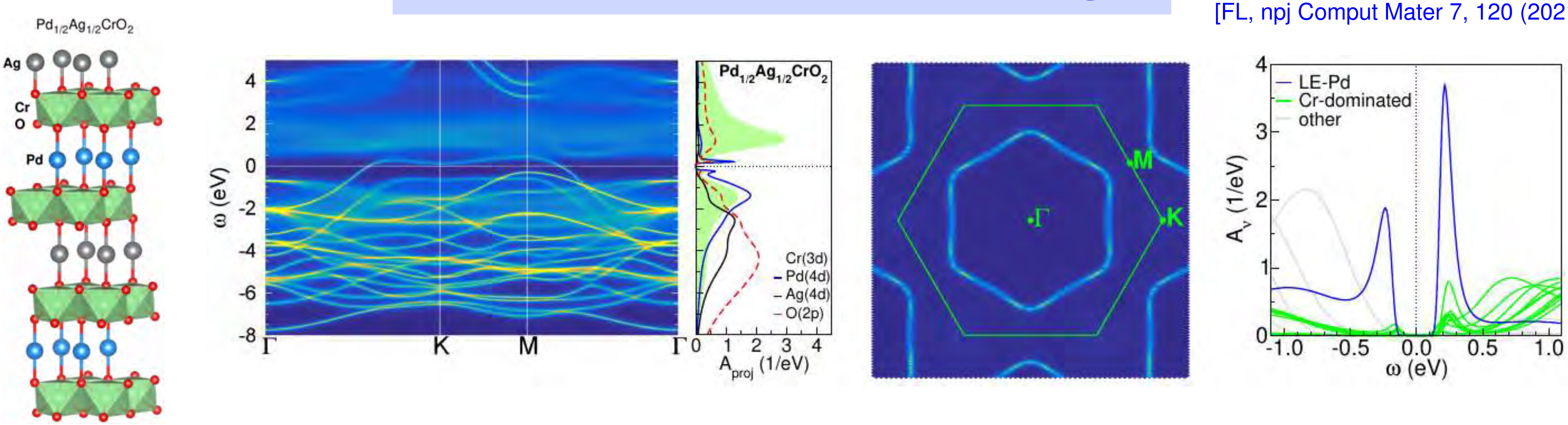
Delafossites: $PdCrO_2$ Mott Design I

[FL, npj Comput Mater 7, 120 (2021)]



- prediction
  very weak dispersion crossing Fermi level
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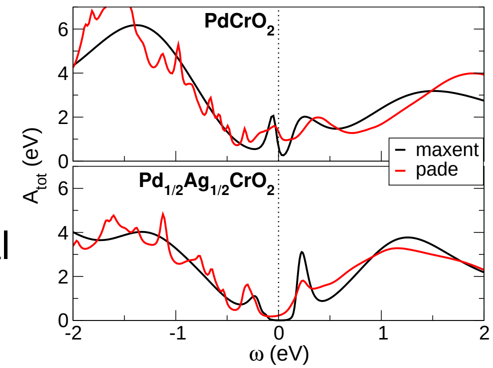
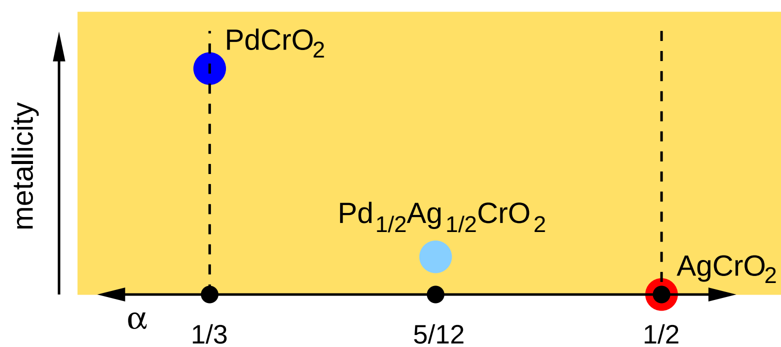


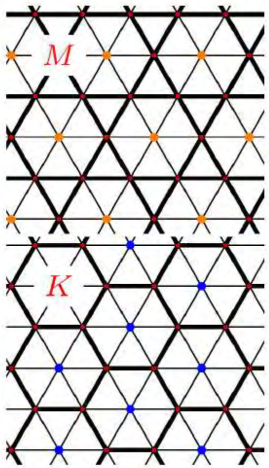
(no maxent artifact!)

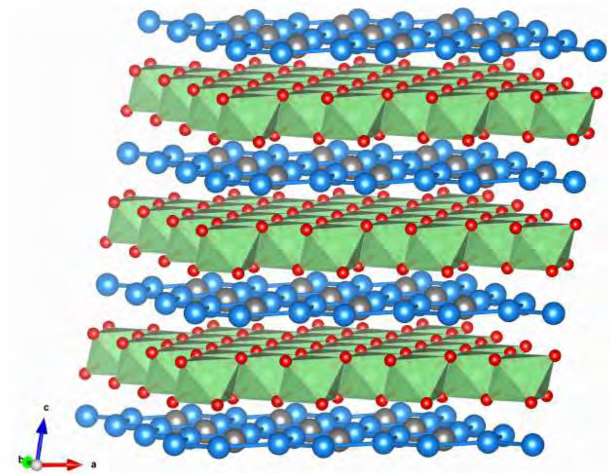
strange metal inbetween hidden-Mott and Mott insulator

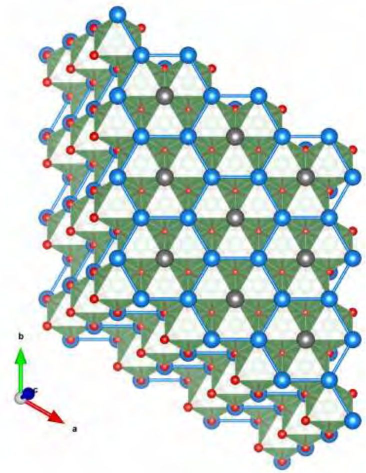


Delafossites: $PdCrO_2$ Mott Design II

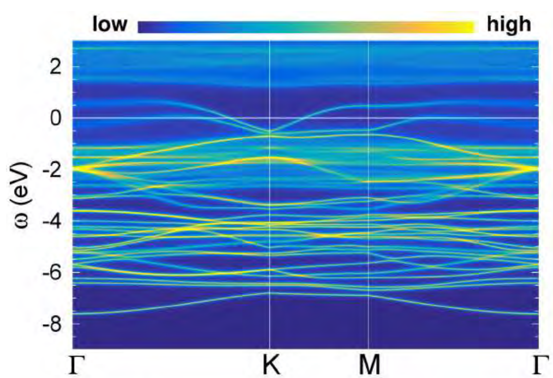
[FL, npj Comput Mater 7, 120 (2021)]

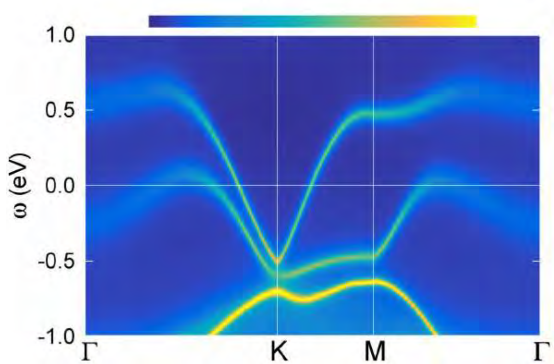




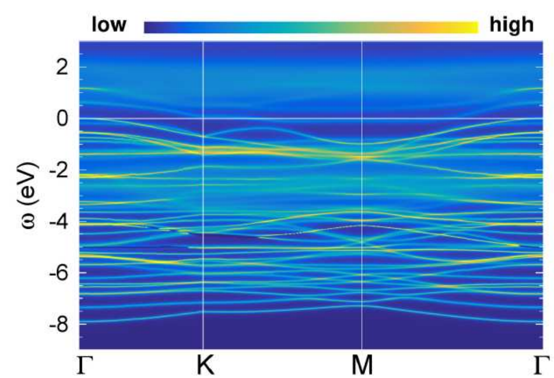


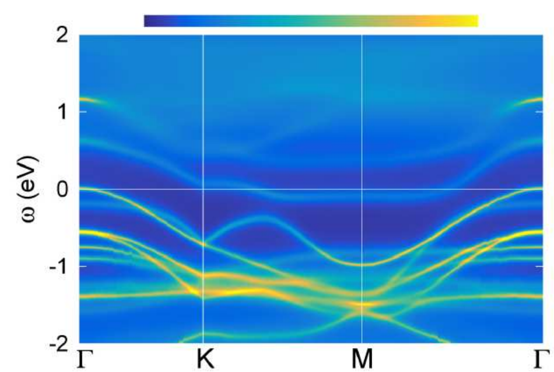
honeycomb-type (K)





kagomé-type (M)





Summary

- DFT+Many-Body methodology is capable of addressing oxide-heterostructure challenges on a realistic level
- $LaAlO_3/SrTiO_3$ interface
  - **electronic correlations *and* defects are relevant to allow for intricate physics**
  - **2DES vs. in-gap weight dichotomy**
- δ-doping of titanate Mott insulators
  - **layer-selective Mott transitions**
  - **triggering the AFM-to-FM criticality**
- delafossites
  - **natural heterostructures (“model system”)**
  - **various design options, to be further explored**