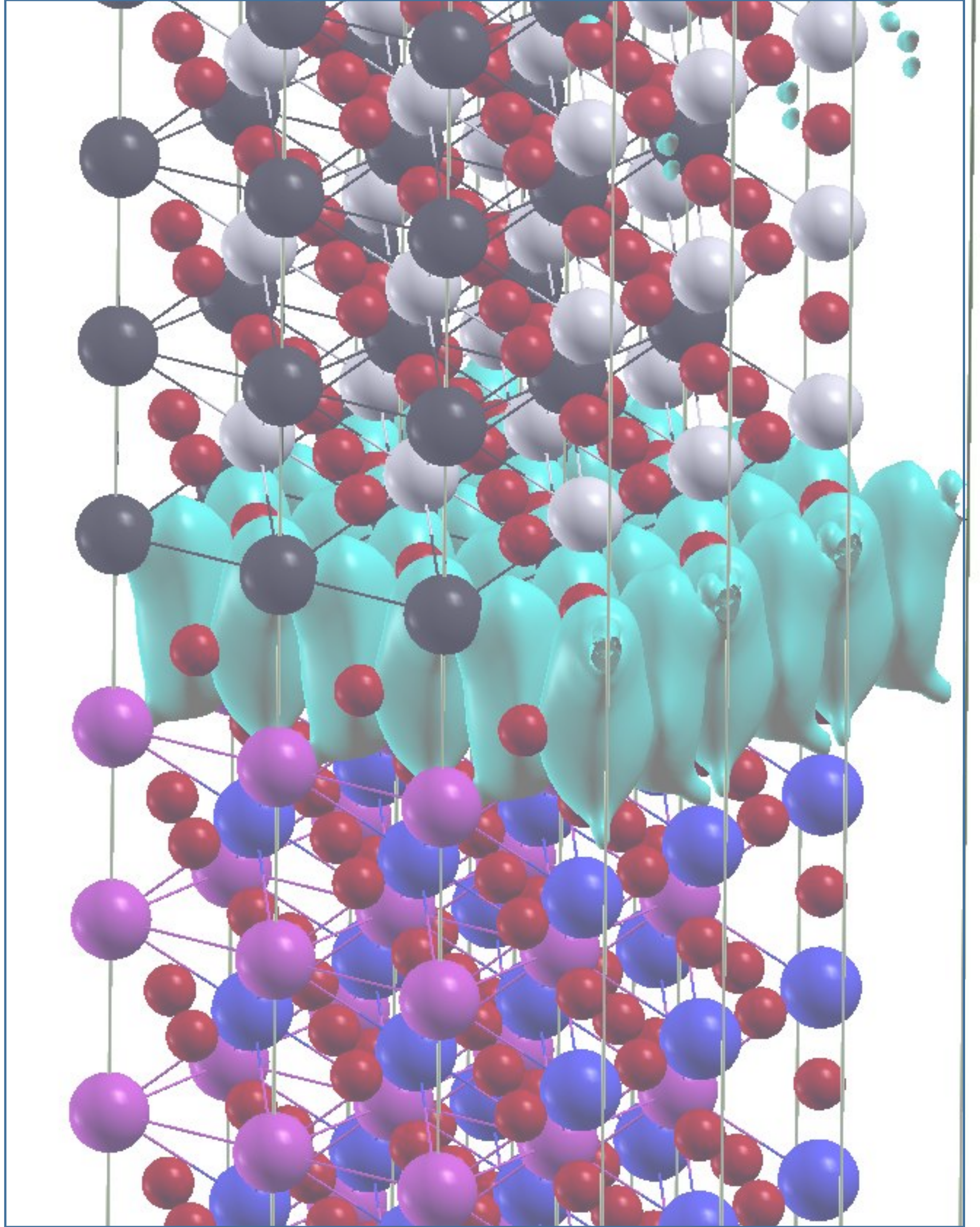

Two Dimensional Electron Gas at LVO/KTO Interface

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Jawaharlal Nehru Centre for Advanced Scientific Research

Bangalore

Engineered 2D Quantum Materials
ICTS-TIFR, Bangalore
July, 2024

Collaborators

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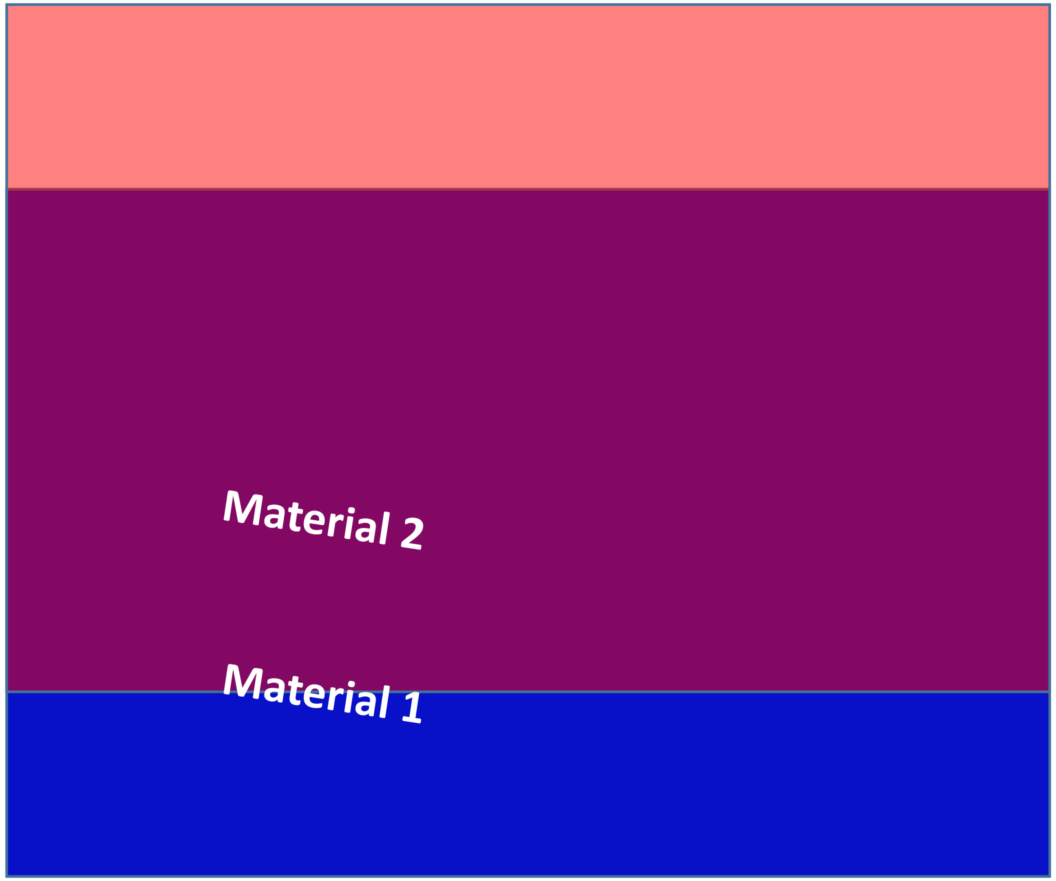
INST, Mohali, India

Prof. Shobhana Narasimhan,
JNCASR, Bangalore, India

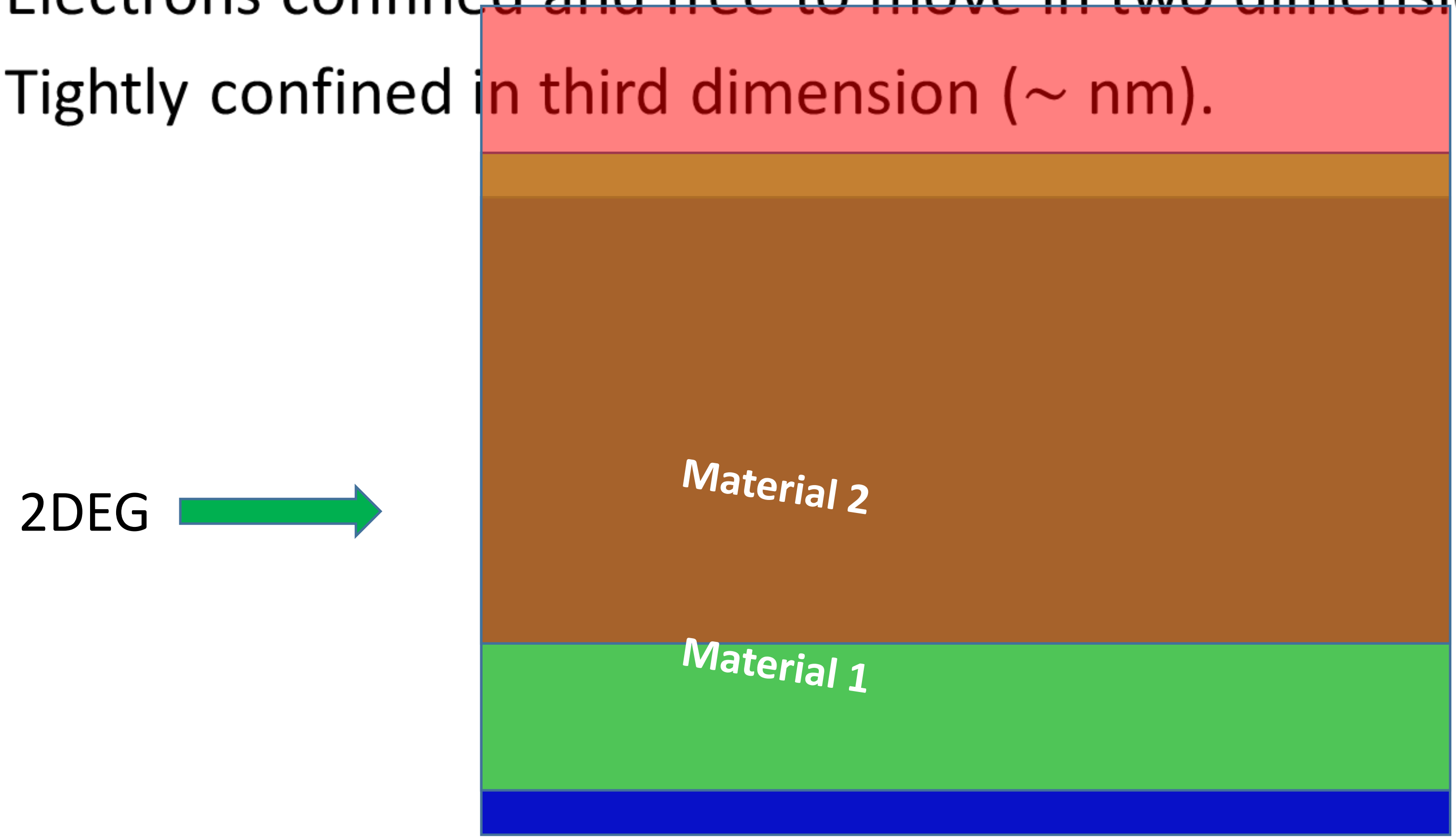
2DEG at Interface

2DEG at Interface



2DEG at Interface

- 2DEG can form at heterointerface.
- Electrons confined and free to move in two dimensions.
- Tightly confined in third dimension (~ nm).



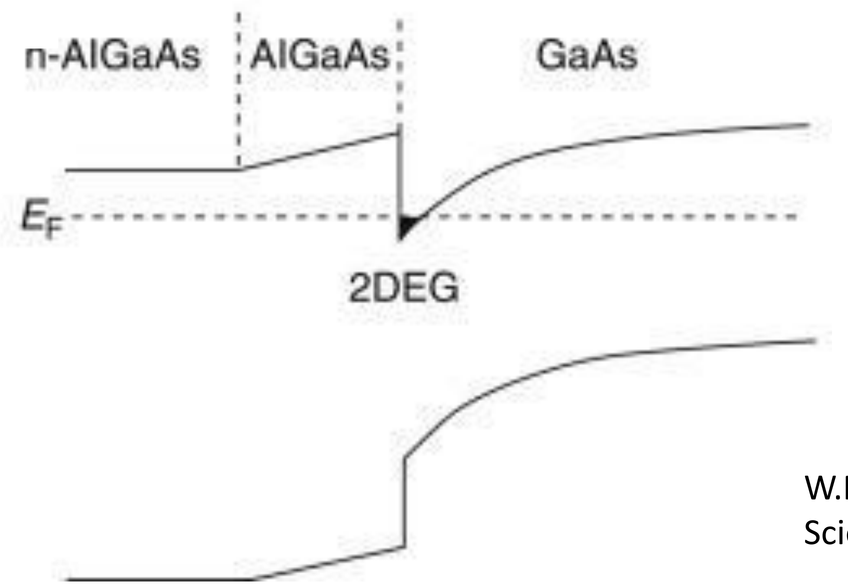
Why is 2DEG Interesting?

- Important for fundamental physics and exotic phenomena in 2D
  (2 Nobel prizes so far!)
  e.g.,
    - Quantum Hall effect.
    - Superconductivity.
    - Wigner Crystallization.

- Important for applications.
    - High carrier concentration and high carrier mobility attractive for devices.
    - e.g., MOSFETs.

Why 2DEG forms - 1

- At semiconductor-semiconductor interfaces:
  e.g., Si/Si-Ge or AlGaAs/GaAs



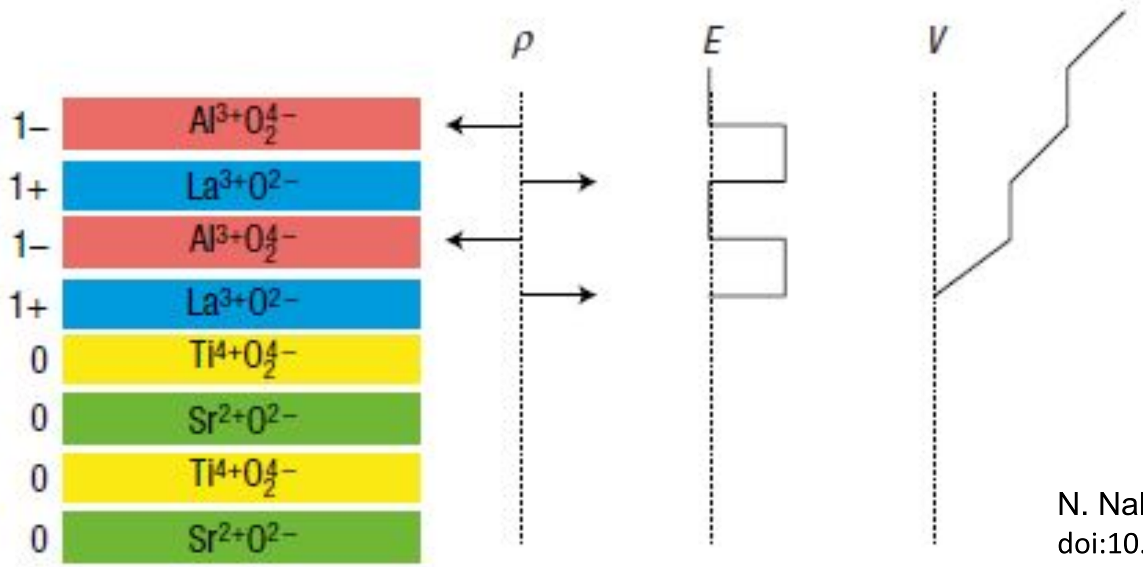
W.R. Clarke et al., Reference Module in Materials Science and Materials Engineering, 2016

- Band gap engineering: band bending + modulation doping + gating.
  (Recall talk by Garima Ahuja on 23rd July)

Why 2DEG forms - 2

- At polar/non-polar interfaces:

  e.g., $LaAlO_3/SrTiO_3$, $LaCrO_3/SrTiO_3$



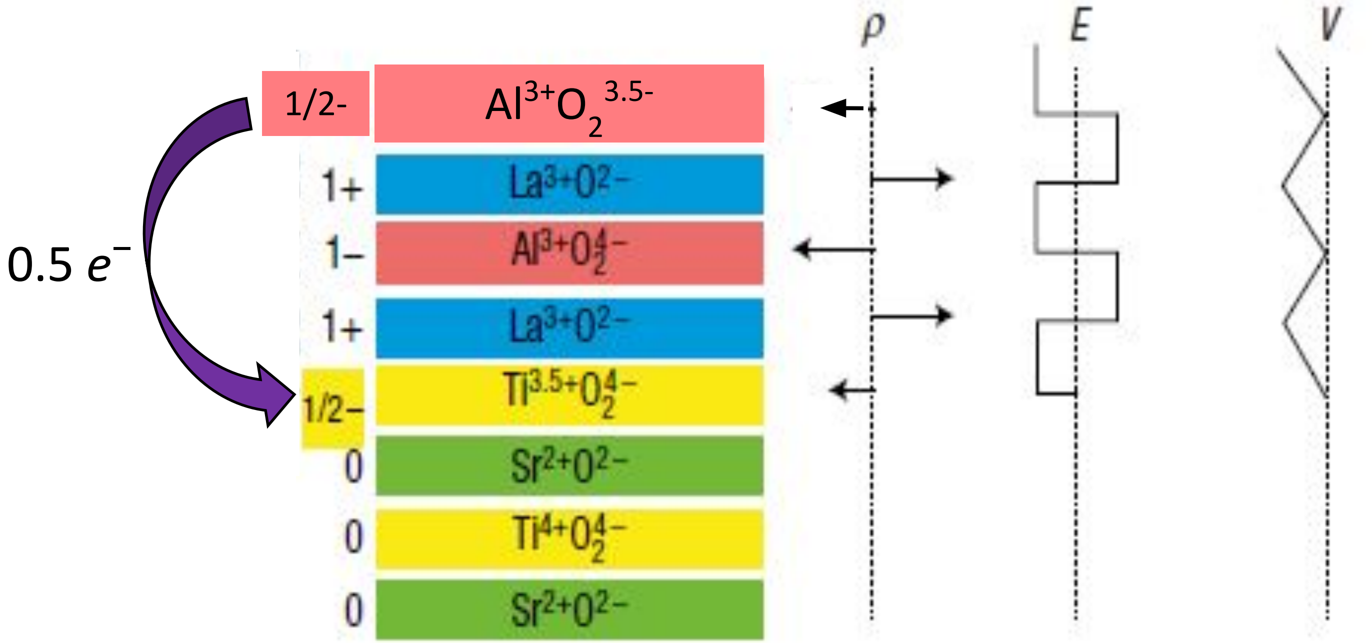
N. Nakagawa et al., Nature Materials, doi:10.1038/nmat1569

- Neutral planes of STO interfaced with alternating charged planes of LAO.
- Produces non-negative electric field.
- Electrostatic potential diverges with increasing film thickness.
- Known as "Polar catastrophe".

Why 2DEG forms - 2

- At polar/non-polar interfaces:

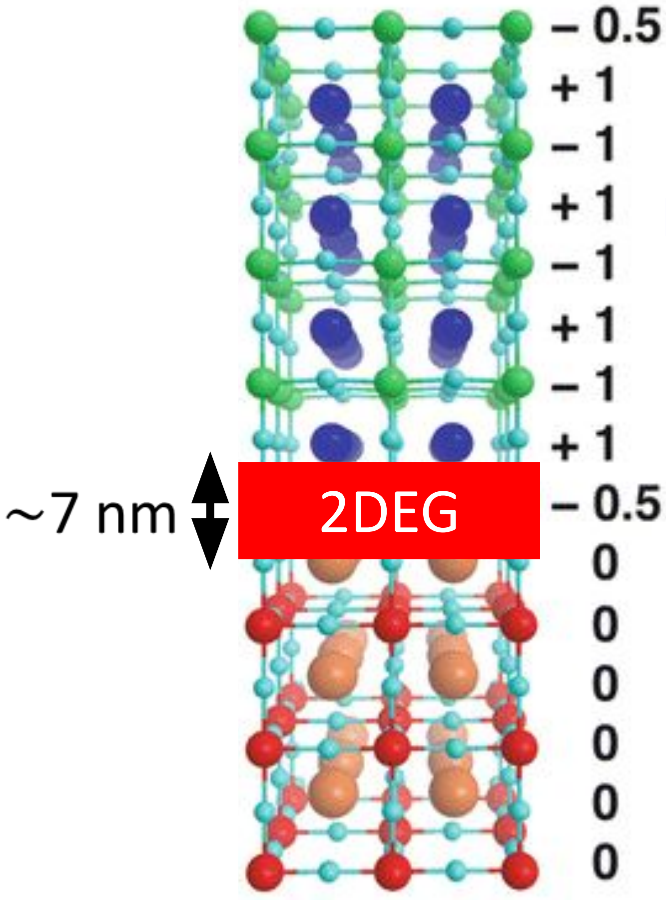
  e.g., $LaAlO_3/SrTiO_3$, $LaCrO_3/SrTiO_3$



N. Nakagawa et al., Nature Materials, doi:10.1038/nmat1569

- Polar catastrophe is avoided by $\frac{1}{2}$ electron transfer to the interfacial TiO_2 layer.
- Electric field oscillates about zero and potential remains finite.
- Known as "electronic reconstruction".

Early Work: Polar/Non-polar Oxide Interface

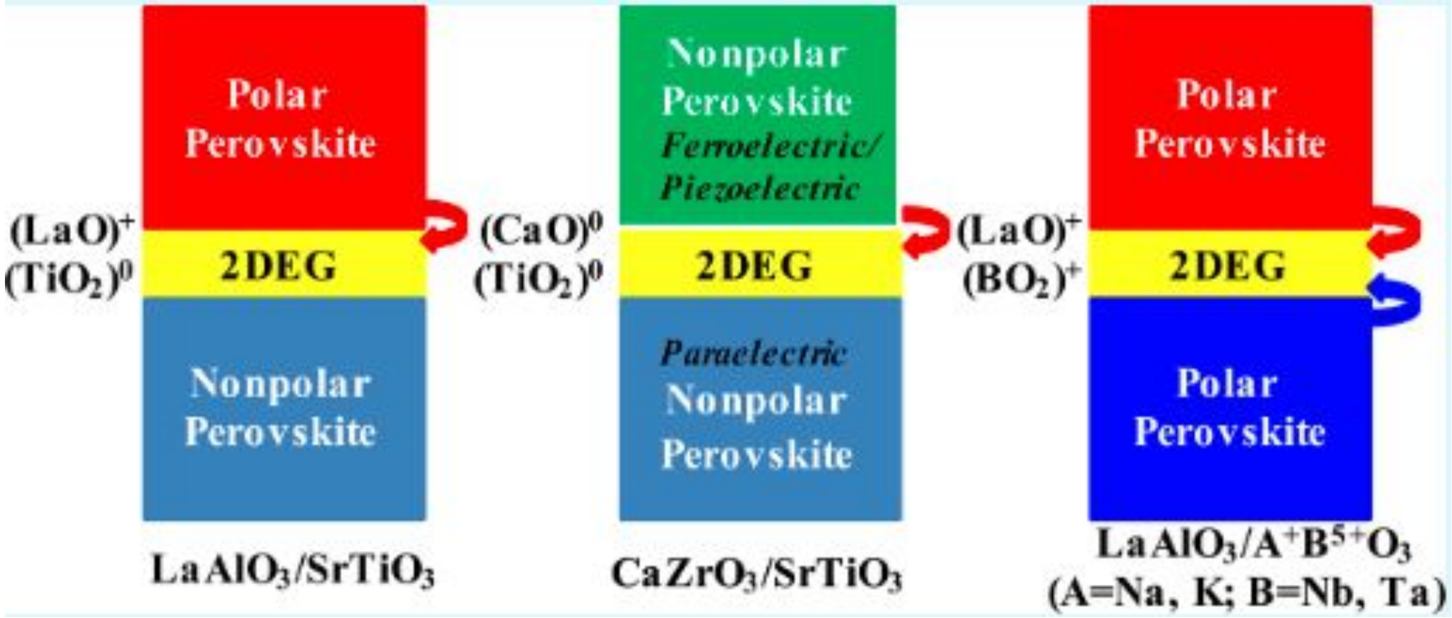


Polar $LaAlO_3$ — Band insulator (gap = 5.6 eV)

Non-polar $SrTiO_3$ — Band insulator (gap = 3.2 eV)

1. Ohtomo et al., NATURE | VOL 427 | 29 JANUARY 2004.
2. M. Basletic et al., *Nature Materials* **7**, pages 621–625 (2008).

Early Work: Other Oxide Interfaces

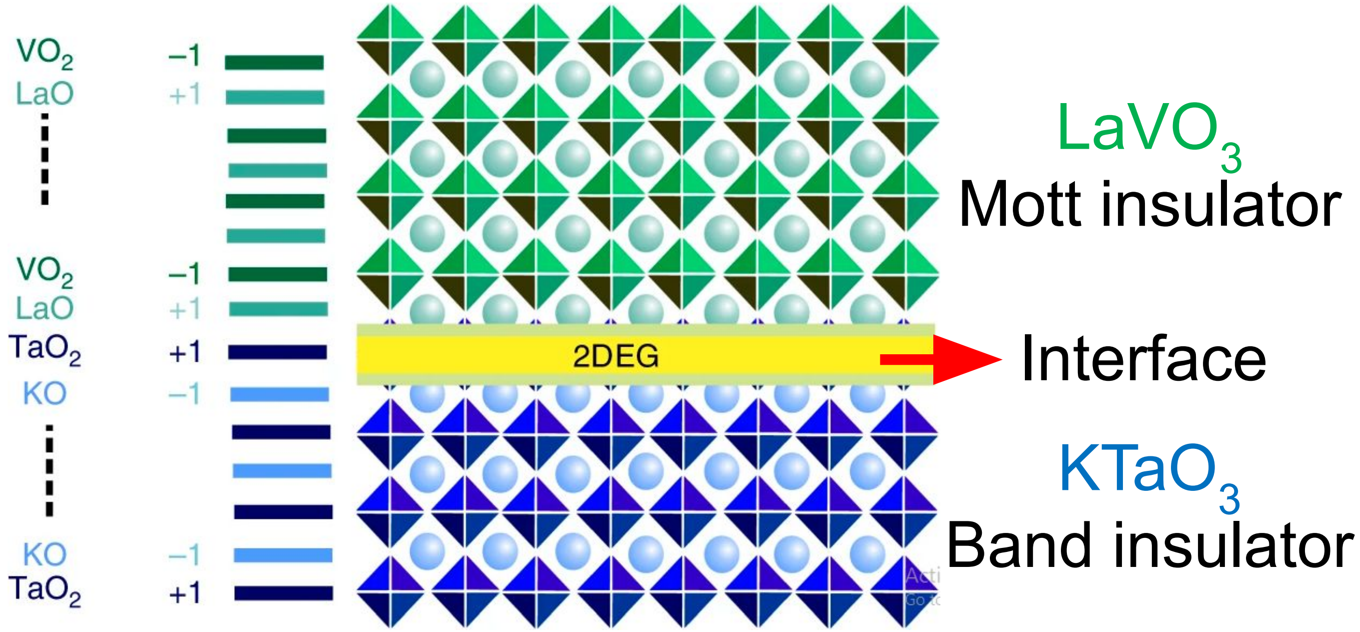


- Non-polar/non-polar oxide interfaces:
  e.g., $CaZrO_3/SrTiO_3$ etc.

- Polar/polar oxide interfaces:
  e.g., $LaTiO_3/KTaO_3$, $LaAlO_3/KNbO_3$, $LaAlO_3/NaTaO_3$ etc.

Fig. is taken from: Y. Wang et al., ACS Appl. Mater. Interfaces **8**, 13659−13668 (2016).

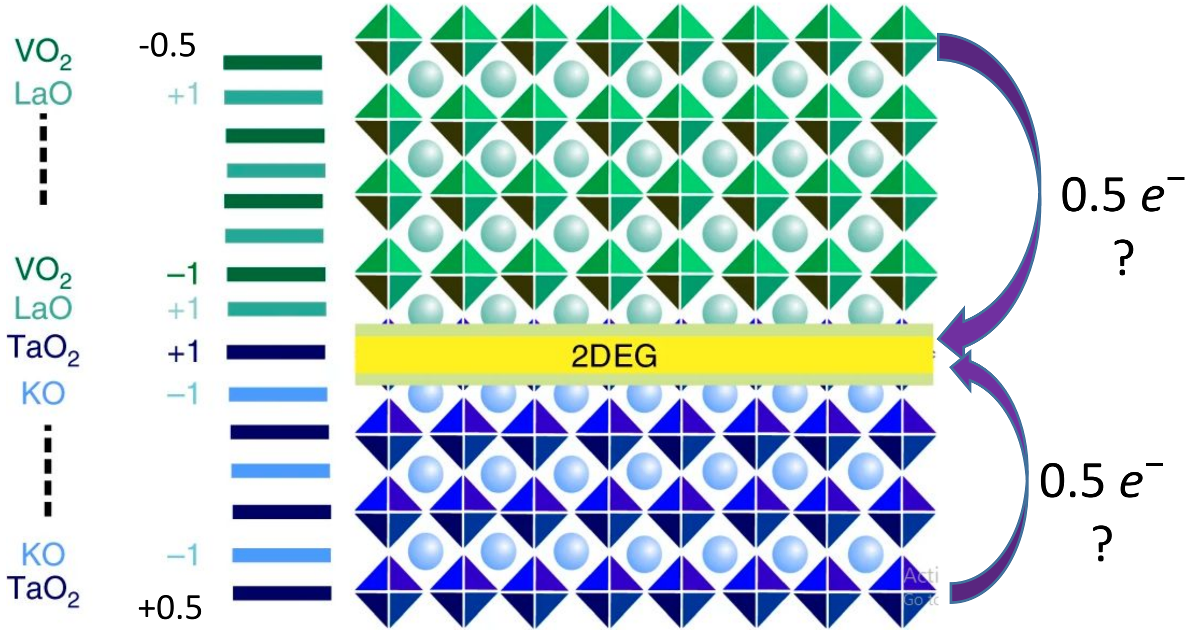
System Under Study



- Deposited thin film of $LaVO_3$ on TaO_2 terminated $KTaO_3$(001) surface.
- LVO is a Mott insulator. KTO is a band insulator.
- Both LVO and KTO are polar i.e., consist of alternating charged layers.

Ref.: N. Wadehra et al., NATURE COMMUNICATIONS | (2020) 11:874.

Why LVO/KTO?



- Polar/polar interface.
- What we hope will happen:

(i) Both the materials will donate electrons increase in carrier density.

(ii) Ta-5d orbitals are less localized compared to Ti-3d orbitals higher e^- mobility.

(iii) Large spin-orbit coupling Rashba physics.

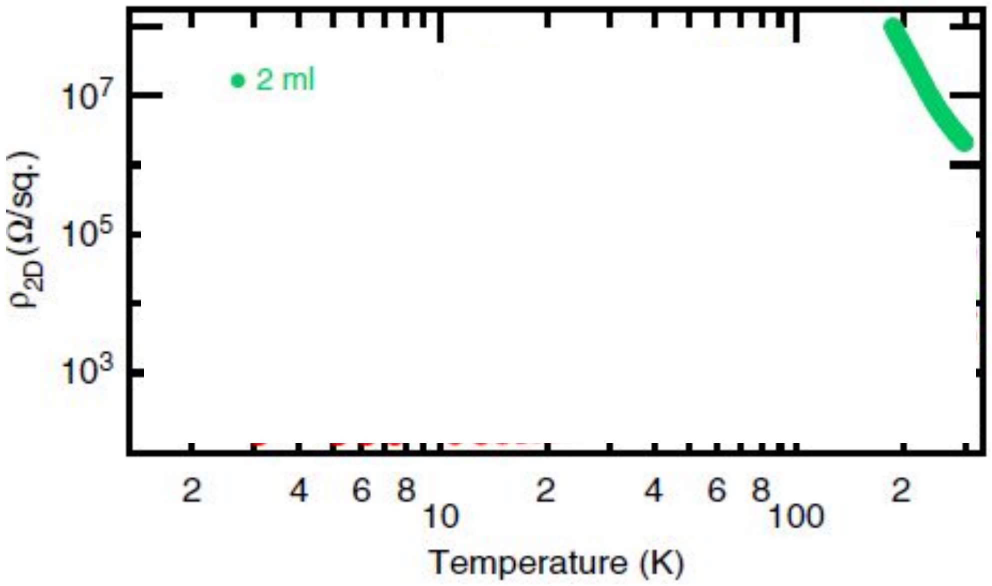
Ref.: N. Wadehra et al., NATURE COMMUNICATIONS | (2020) 11:874.

Aims of this Study

- Joint experimental and theoretical study.
- Experiments done in the group of Prof. Suvankar Chakraverty, INST, Mohali.
- Want to find other systems which exhibit 2DEG at oxide interface, in particular $LaVO_3/KTaO_3$ (LVO/KTO).
- LVO/KTO is a polar/polar interface – Can give higher interfacial electron density of 2DEG?
- Determine the origin of 2DEG at LVO/KTO interface.
- DFT calculations to confirm experimental observations and provide additional insights.

Experimental Observations: Signature of Formation of 2DEG at LVO/KTO Interface

Resistivity vs. Temp for diff thicknesses of LVO

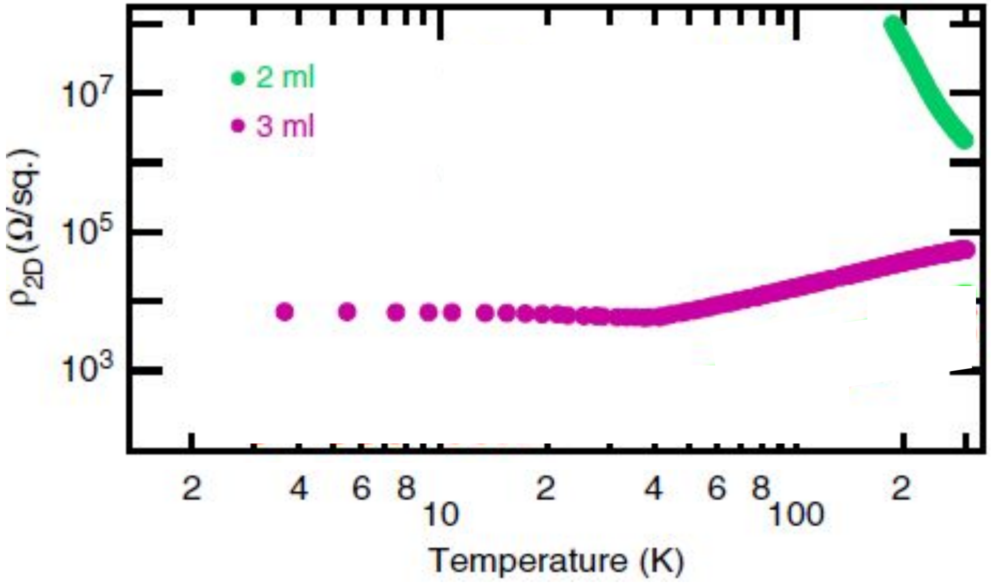


- At 2 ML LVO film thickness, resistivity is very high.
- Resistivity decreases with increasing temp.
- Interface is insulating.

Ref.: N. Wadehra et al., NATURE COMMUNICATIONS | (2020) 11:874.

Experimental Observations: Signature of Formation of 2DEG at LVO/KTO Interface

Resistivity vs. Temp for diff thicknesses of LVO

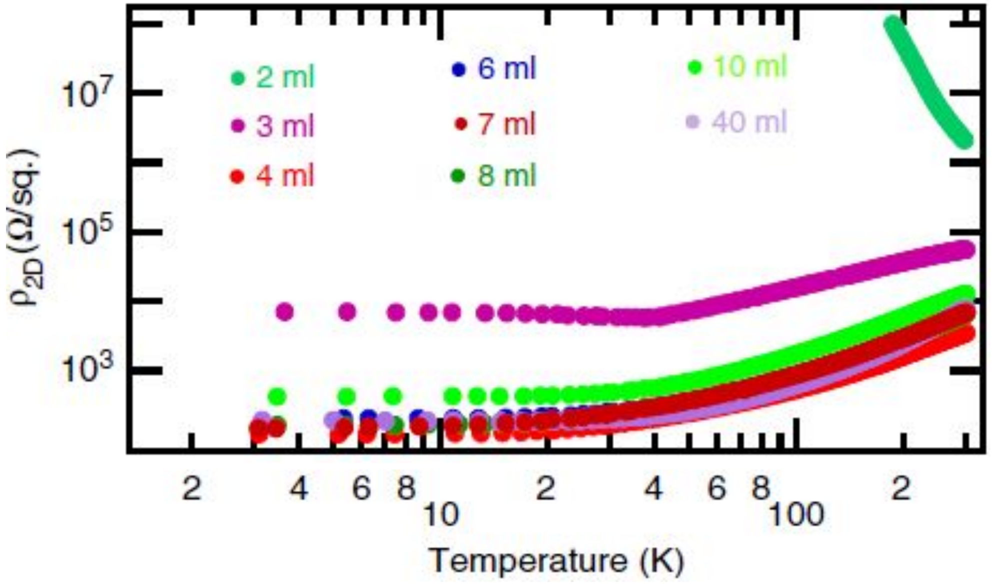


- At 3 ML LVO film thickness, resistivity became smaller, but still high.
- Resistivity increases linearly with increasing T.

Ref.: N. Wadehra et al., NATURE COMMUNICATIONS | (2020) 11:874.

Experimental Observations: Signature of Formation of 2DEG at LVO/KTO Interface

Resistivity vs. Temp for diff thicknesses of LVO

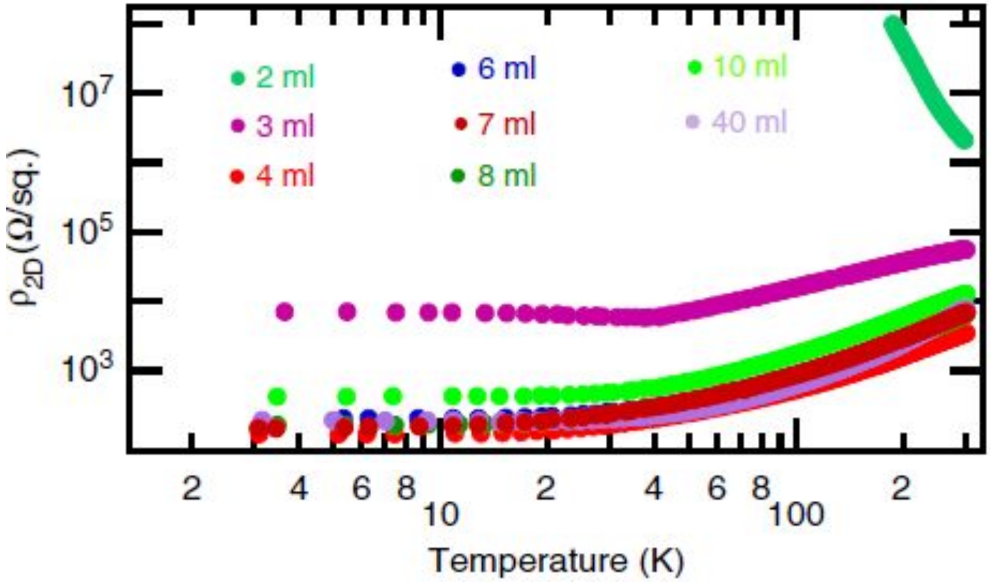


- Above 3 ML LVO film thickness, interfacial resistivity drops down few orders of magnitude.
- Resistivity increases linearly with increasing T.
- Interface becomes metallic above 3 ML.

Ref.: N. Wadehra et al., NATURE COMMUNICATIONS | (2020) 11:874.

Experimental Observations: Signature of Formation of 2DEG at LVO/KTO Interface

Resistivity vs. Temp for diff thicknesses of LVO



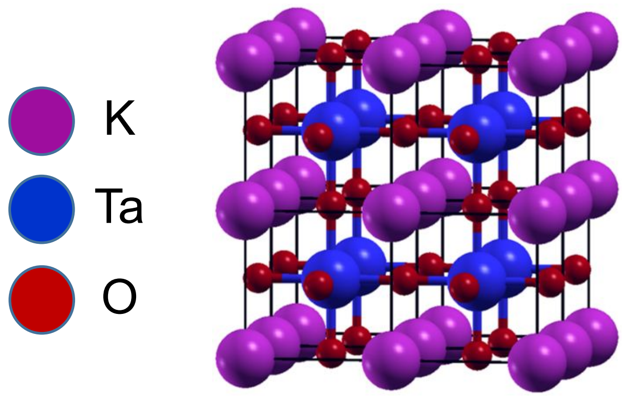
- Above 3 ML LVO film thickness, interfacial resistivity drops down few orders of magnitude.
- Resistivity increases linearly with increasing T.
- Interface becomes metallic above 3 ML.
- Metallicity is confined within ~ 10 nm around interface.
- High carrier mobility ~ 600 $cm^2\ V^{-1}\ s^{-1}$ at 1.8 K.
- High carrier density of 1×10^{14} electrons/cm^{-2}. (compare with 1×10^{13} electrons/cm^{-2} for LAO/STO system)

Ref.: N. Wadehra et al., NATURE COMMUNICATIONS | (2020) 11:874.

Computational Details

- Spin-polarized Density functional theory, Quantum ESPRESSO package.
- Plane wave basis (ecutwfc = 45 Ry), ultrasoft pseudopotentials.
- PBE-GGA exchange-correlation.
- Hubbard *U* and *J* for strongly correlated V-3*d* and Ta-5*d* orbitals.
- BZ sampling using 8×8×1 Monkhorst-Pack k-mesh.
- Marzari-Vanderbilt cold smearing of width of 0.005 Ry.
- Geometry optimization using Broyden-Fletcher-Goldfarb-Shanno (BFGS) algorithm.

Bulk KTO and LVO Structures

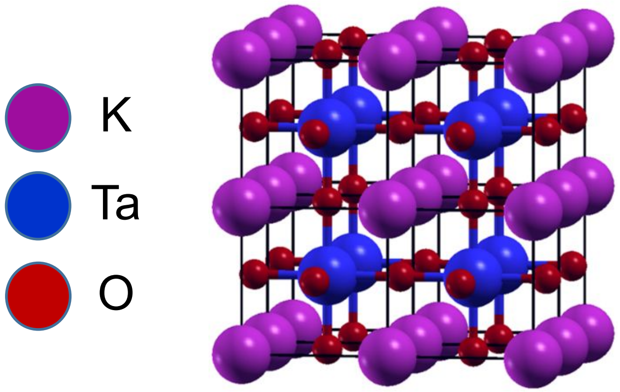


Cubic
NM

DFT optimized lattice constant:
a_0^{KTO} = 4.011 Å (3.989 Å)

Band insulator

Bulk KTO and LVO Structures



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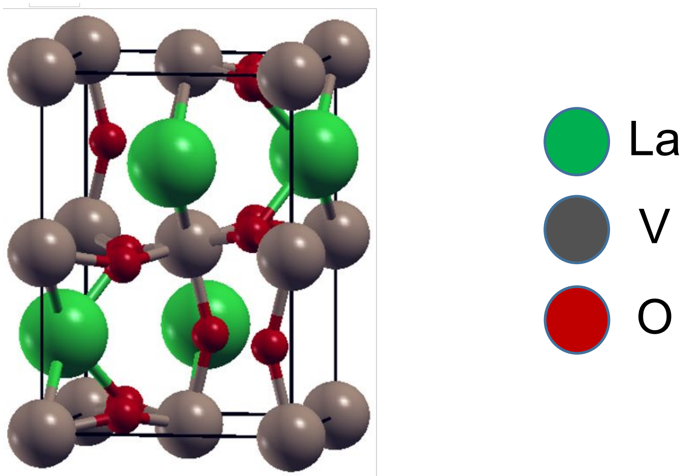


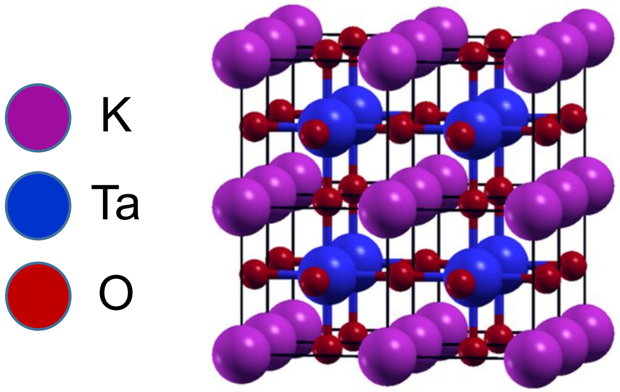
Orthorhombic
NM

RT structure

Mott insulator

DFT optimized lattice constants:
a = 5.547 Å (5.555 Å)
b = 5.571 Å (5.553 Å)
c = 7.952 Å (7.848 Å)

Bulk KTO and LVO Structures



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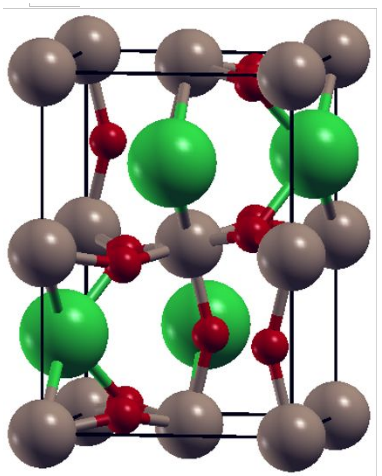
Band insulator



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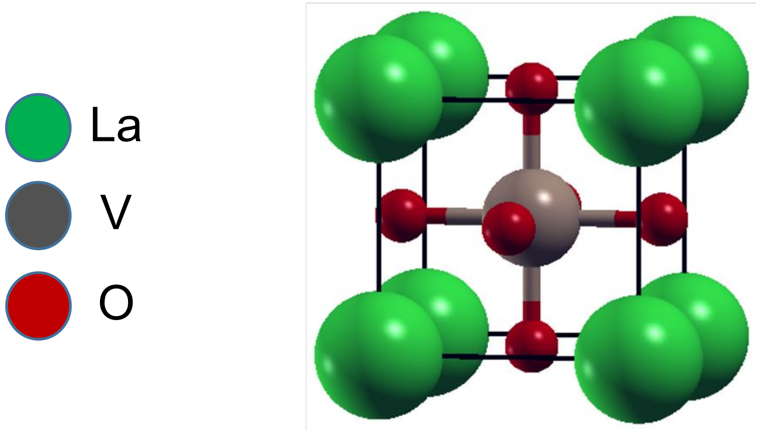


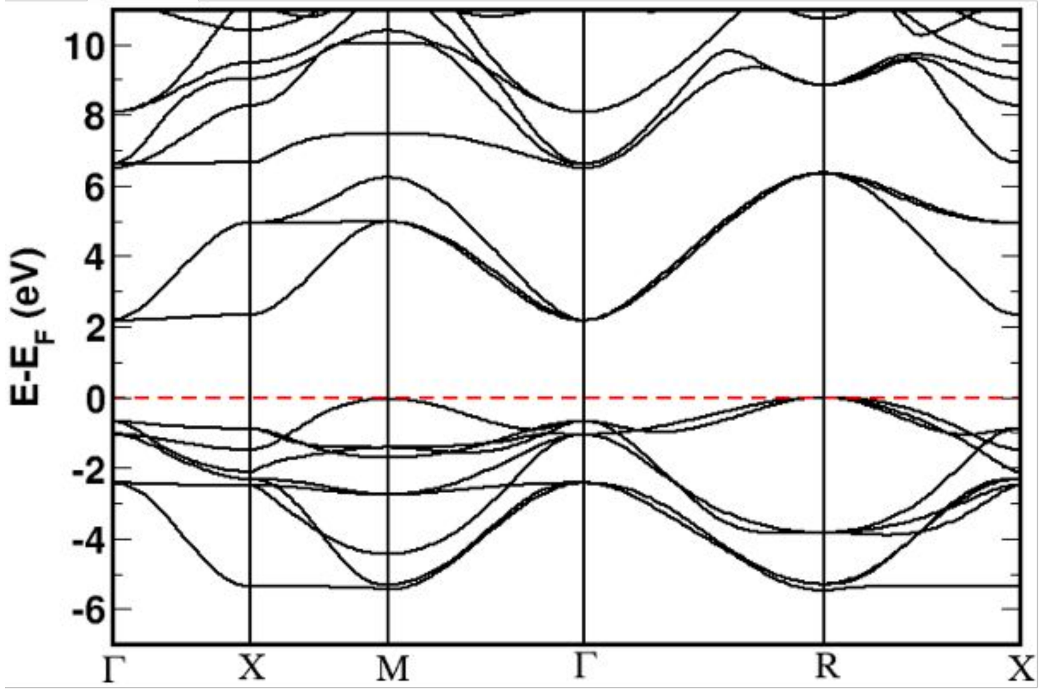
Tetragonal
A-AFM

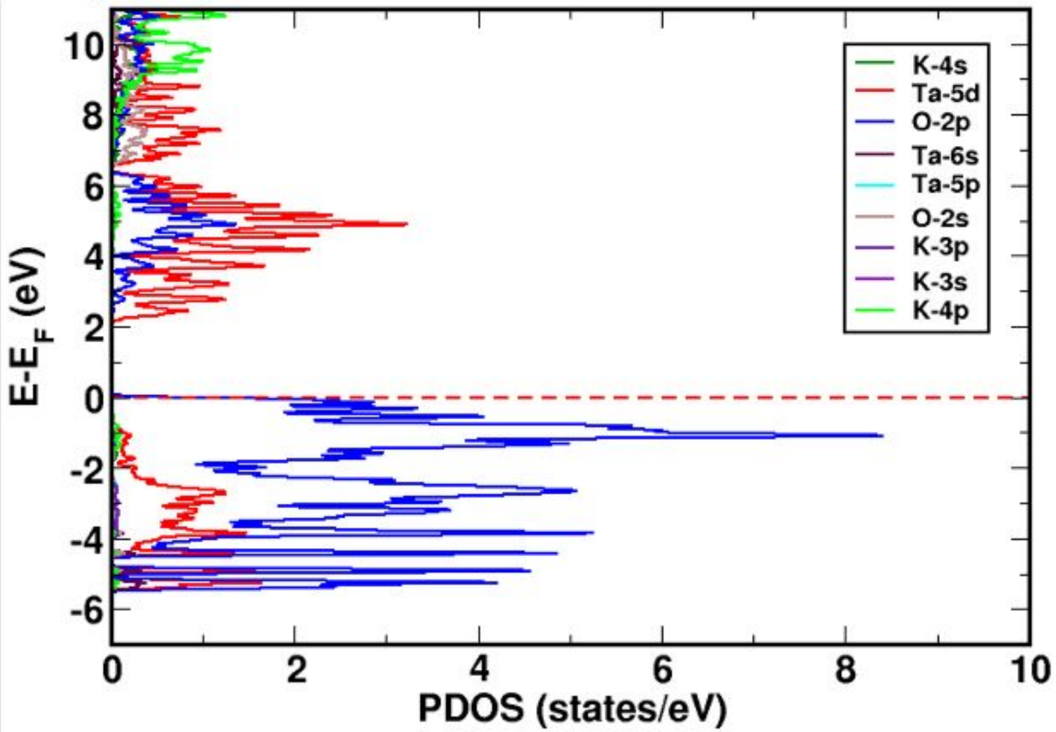
Hypothetical structure considered as LVO takes tetragonal structure when grown on KTO(001)

a = b = a_0^{KTO} = 4.011 Å
c = 3.952 Å (DFT optimized)

A-AFM ordering favored over NM, FM, C-AFM and G-AFM

Electronic Structure of Bulk KTO

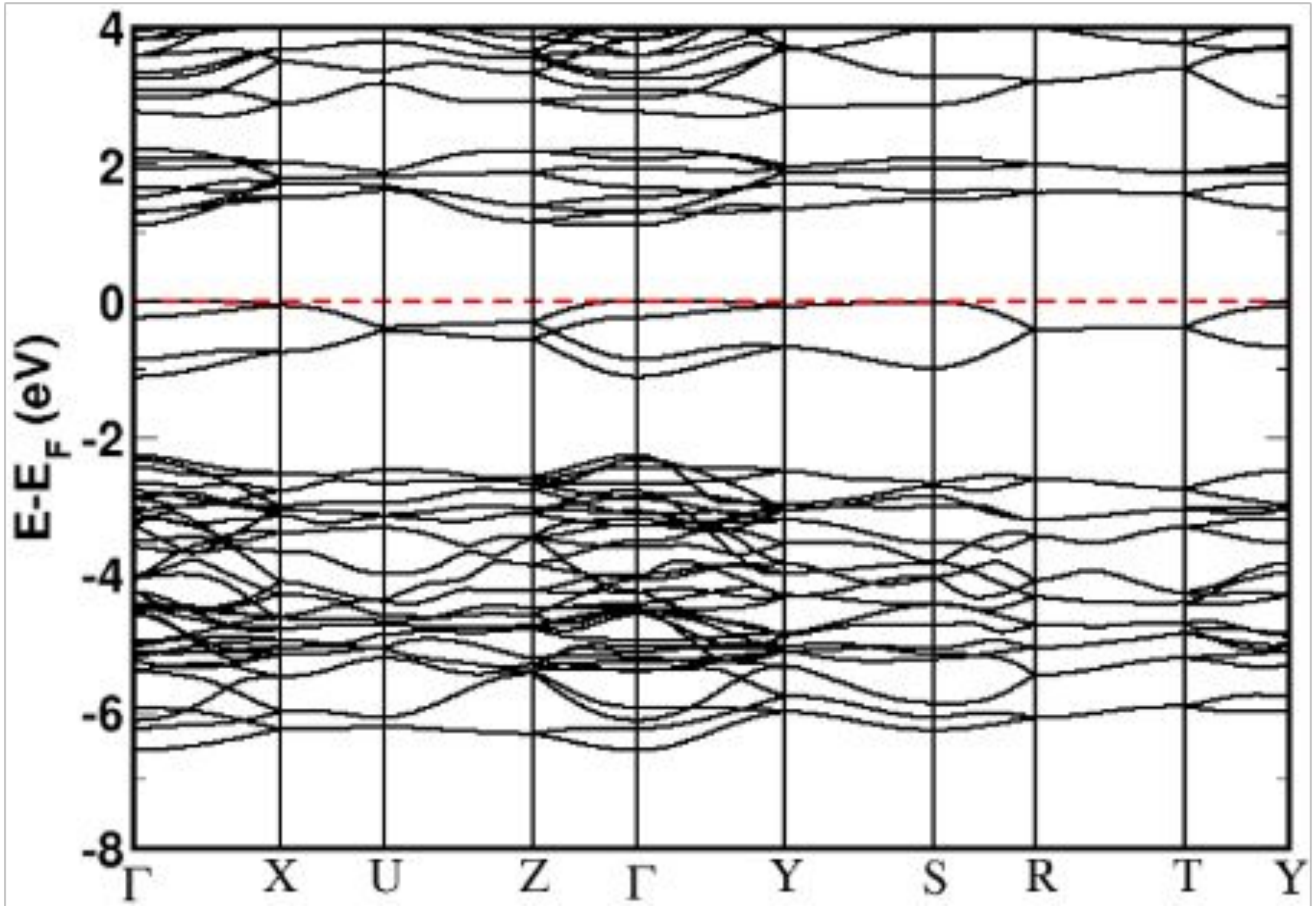




- Calculated band gap = 2.18 eV;
  Previous DFT: 2.1 eV
  Experimental value = 3.60 eV.
- VBM at R point, CBM at Γ point. Indirect gap.

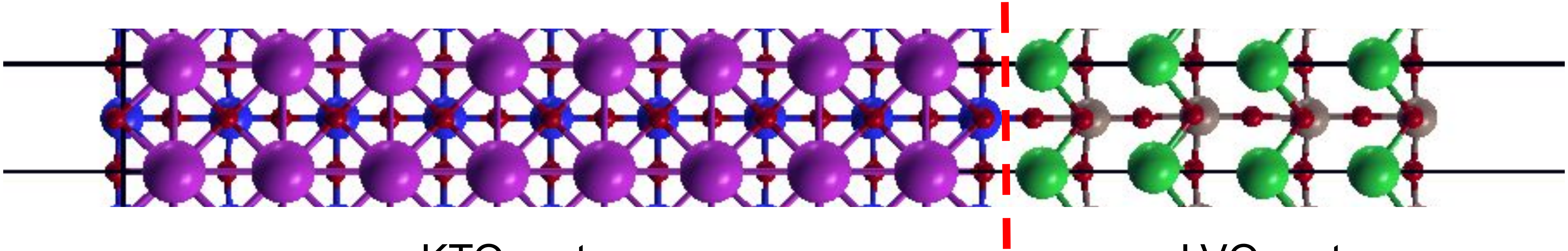
- Valence bands mainly come from O-2*p* orbitals.
- Conduction bands mainly come from Ta-5*d* orbitals.

Electronic Structure of Bulk LVO



- LVO is a Mott insulator. Therefore need *U* to get correct physics.
- For *U* = 5.65 eV and *J* = 0.65 eV, band gap matches with expt. value of 1.1 eV.

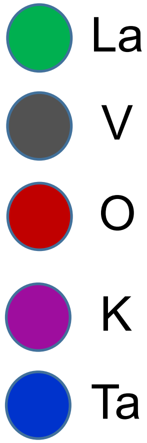
Atomic Structure of LVO/KTO Interface



KTO part

LVO part

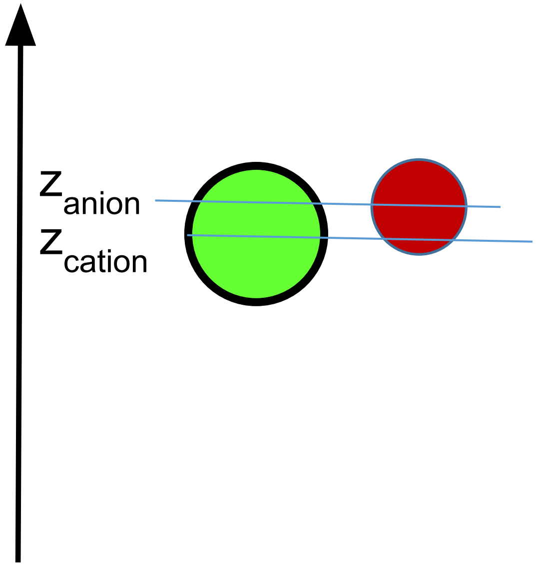
$(LaO)^{+}/(TaO_2)^{+}$
interface



Slab structure of $(LVO)_4/(KTO)_{8.5}$ interface
(with vacuum)

Polar Distortions at LVO/KTO Interface

Relative displacement = $z_{cation} - z_{anion}$



Polar Distortions at LVO/KTO Interface

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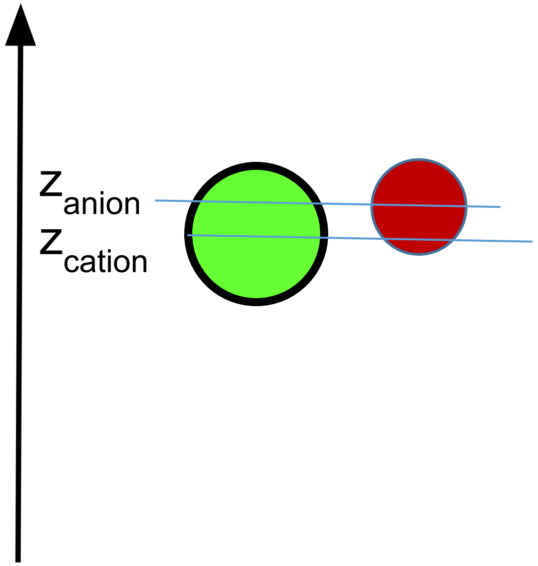
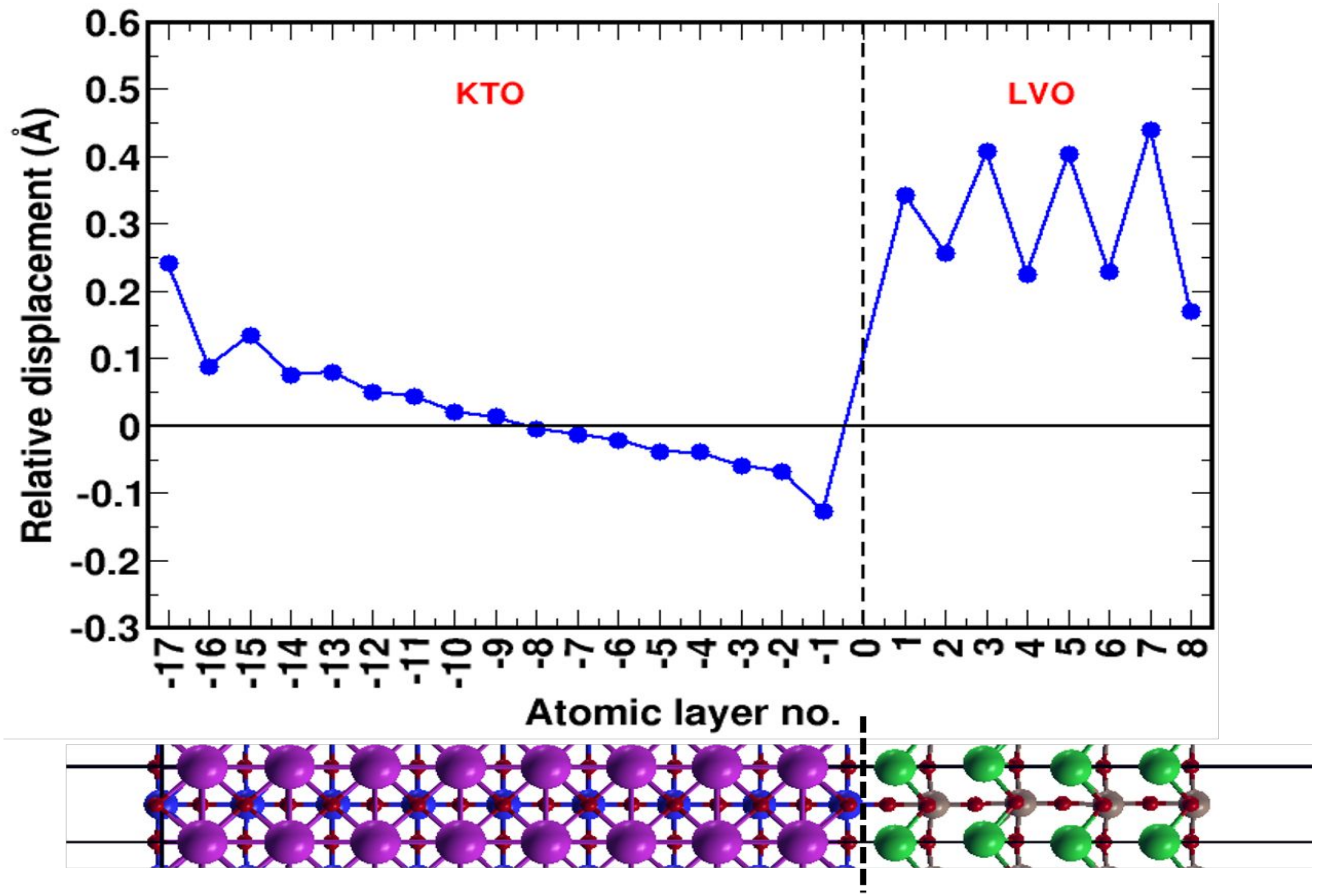
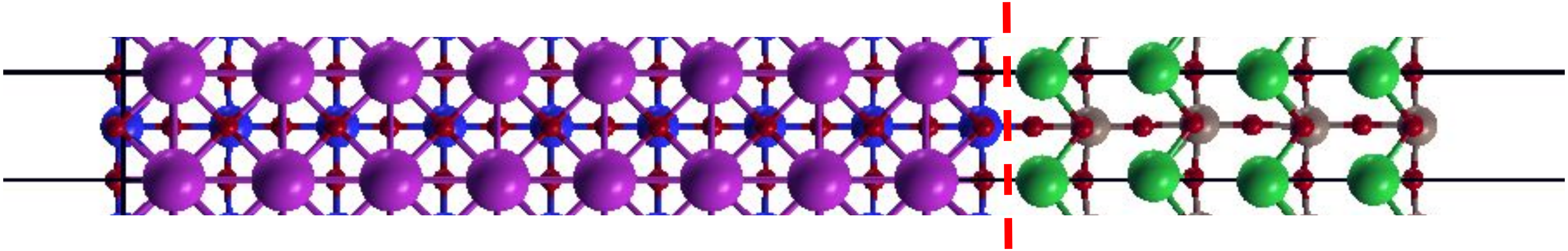




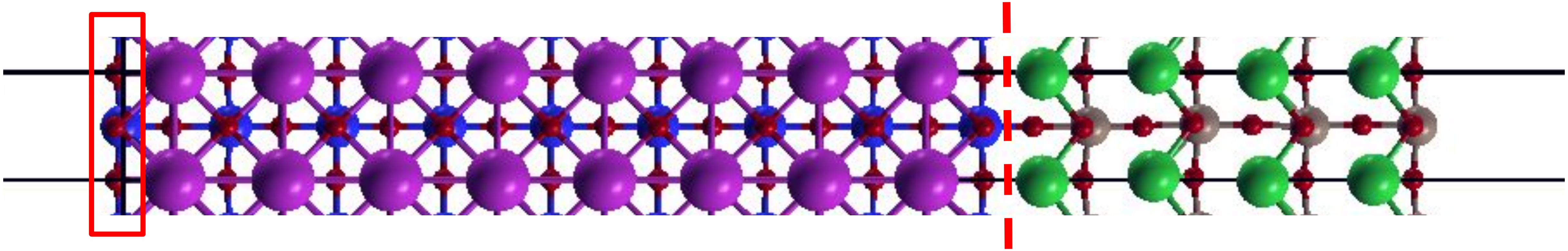
- O^{2-} anions want to come closer to the interface.
- Ferroelectric like polar distortions create local dipoles.
- Similar behavior observed at LAO/STO and LSO/BSO interfaces.[1,2]

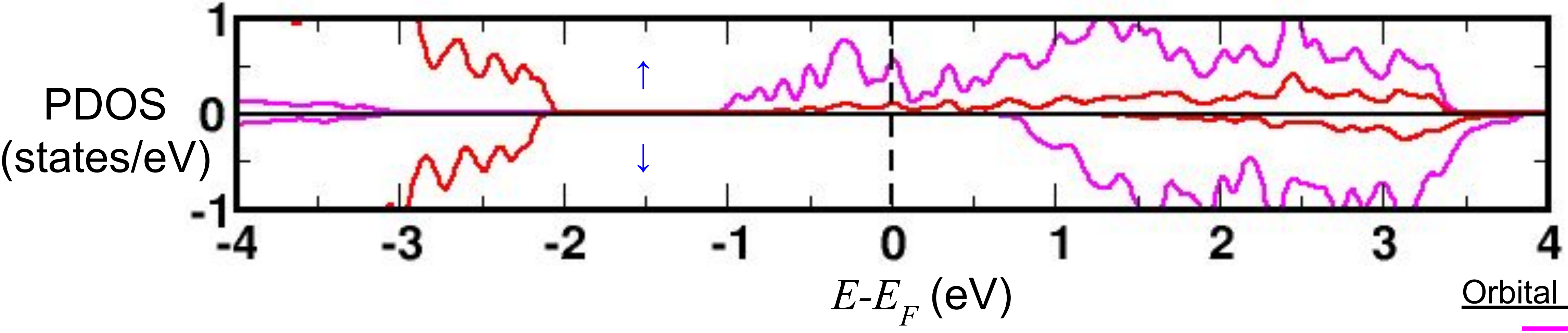
[1, 2]: Phys. Rev. B 96, 245423 (2017) and Appl. Phys. Lett. 111, 141604 (2017)].

Layer-resolved PDOS of LVO/KTO Interface



Layer-resolved PDOS of LVO/KTO Interface
(layer: -17)

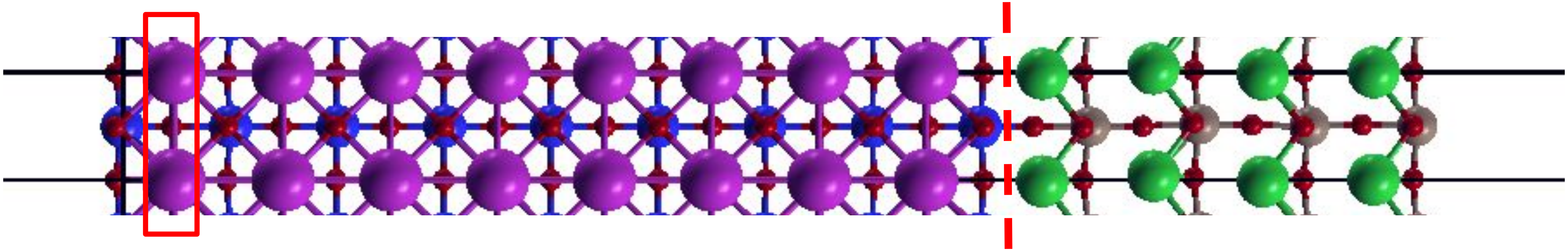


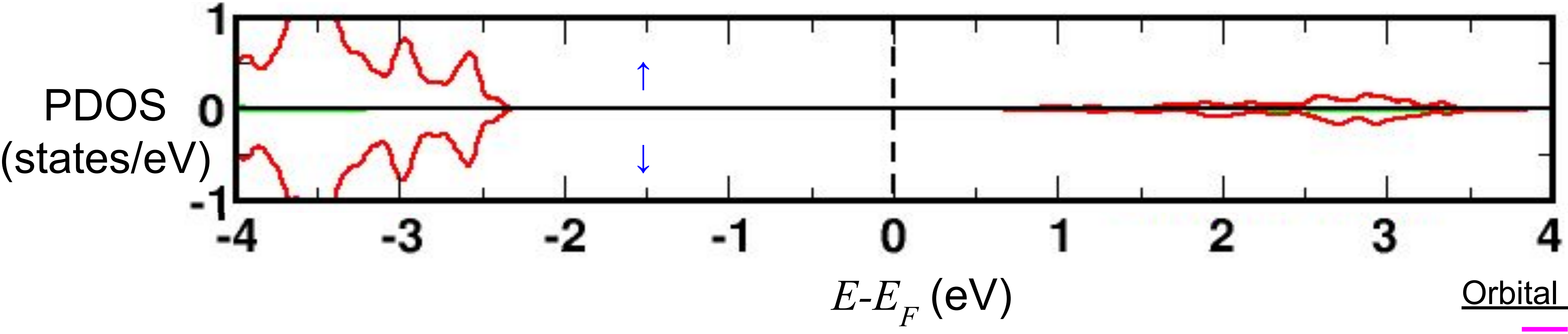


Orbital color code:
- Ta-5d
- V-3d
- O-2p
- La-5d

- Spin-polarized 2DEG at KTO surface.

Layer-resolved PDOS of LVO/KTO Interface
(layer: -16)

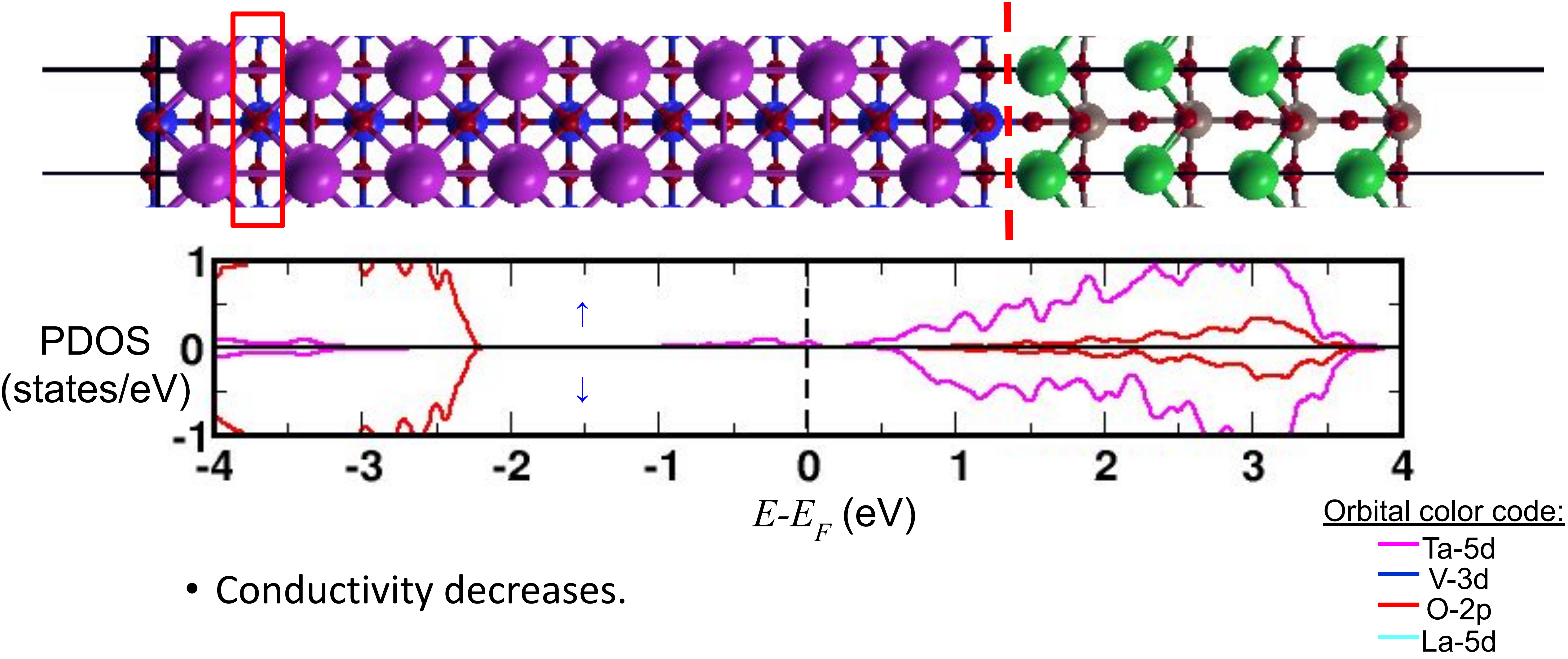




Orbital color code:
- Ta-5d
- V-3d
- O-2p
- La-5d

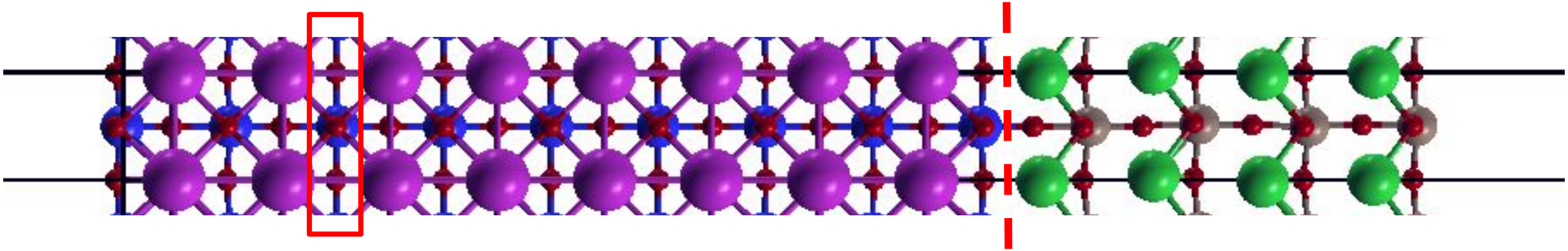
- Insulating.

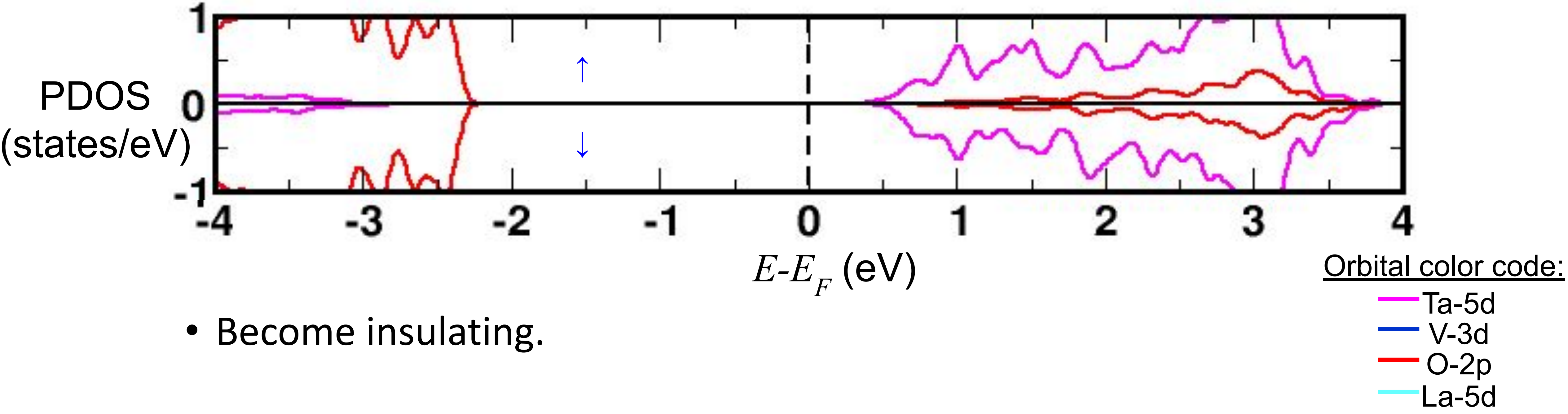
Layer-resolved PDOS of LVO/KTO Interface
(layer: -15)



- Conductivity decreases.

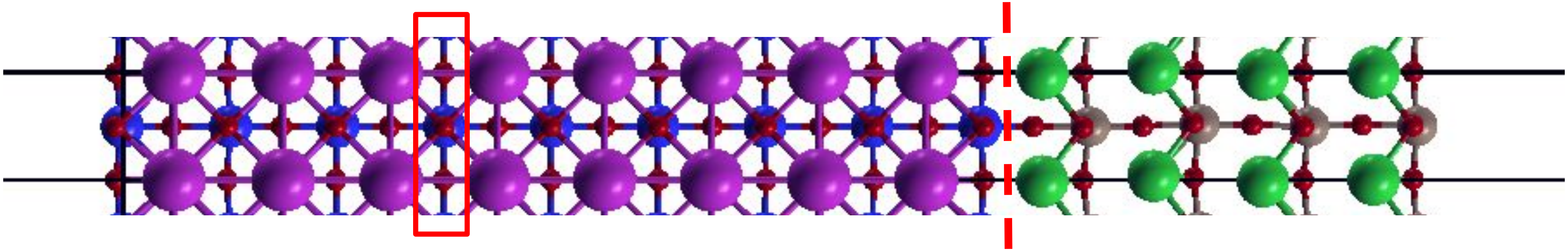
Layer-resolved PDOS of LVO/KTO Interface (layer: -13)

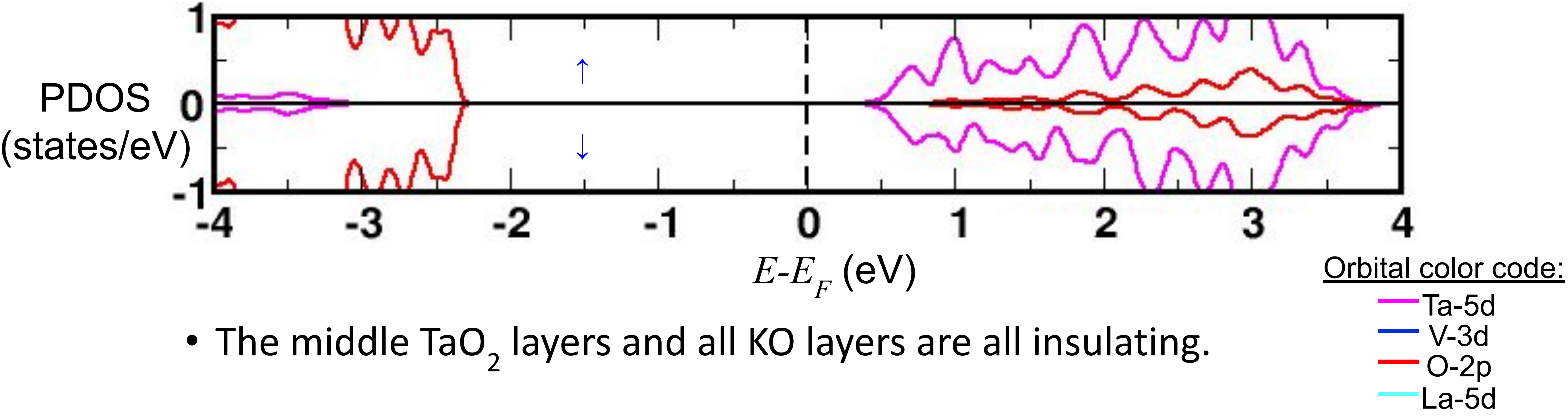




- Become insulating.

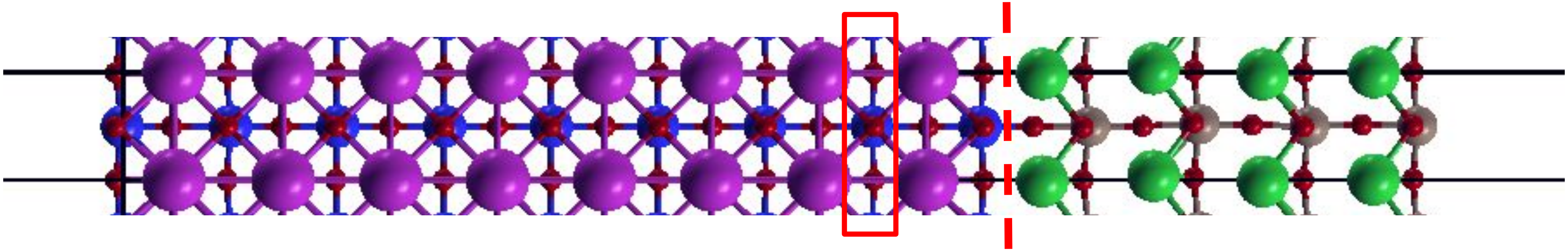
Layer-resolved PDOS of LVO/KTO Interface (layer: -11)

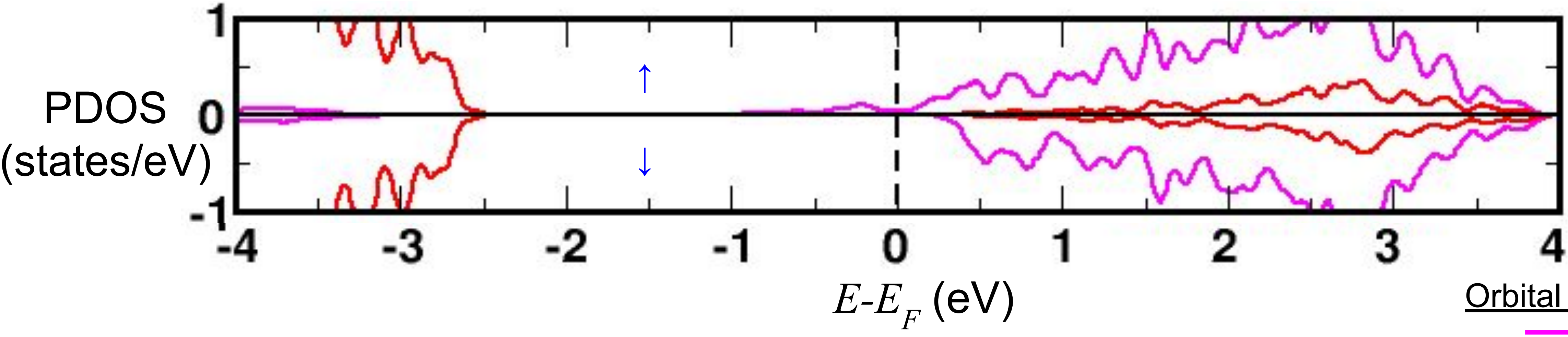




- The middle TaO_2 layers and all KO layers are all insulating.

Layer-resolved PDOS of LVO/KTO Interface
(layer: -3)

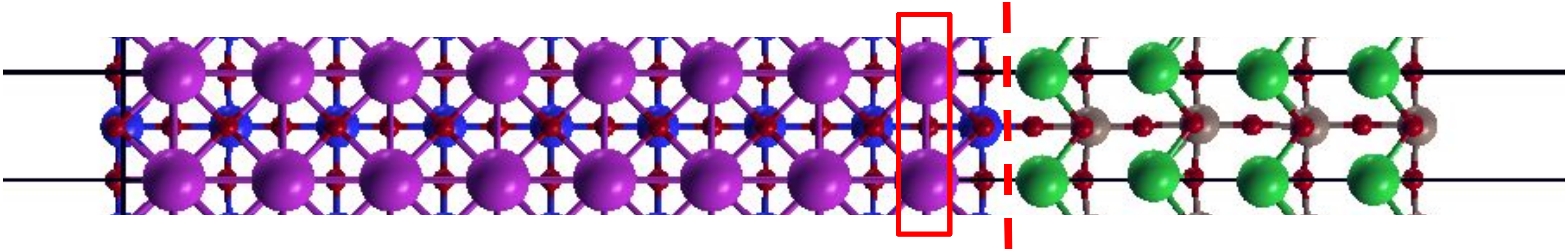


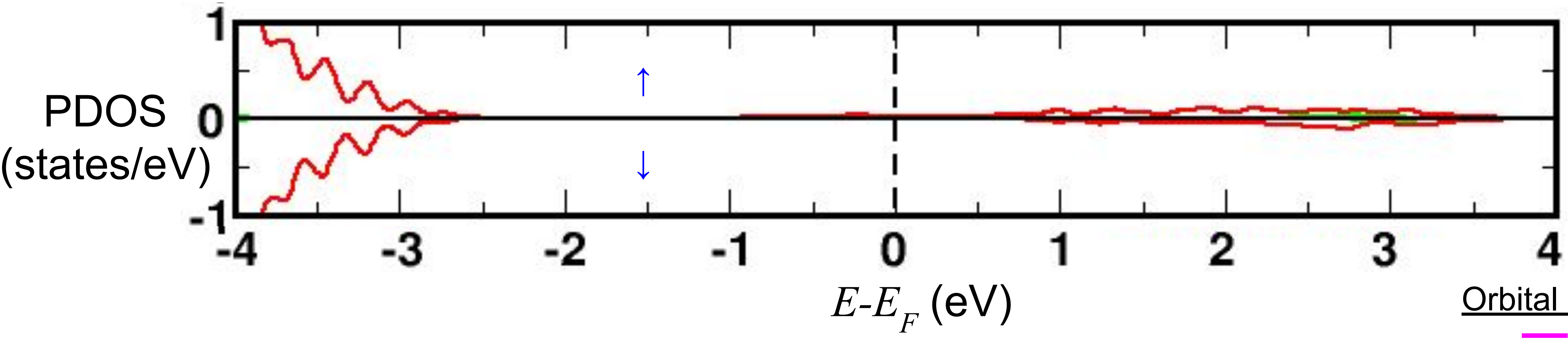


- Little contribution to conduction and hence to 2DEG.

Orbital color code:
Ta-5d
V-3d
O-2p
La-5d

Layer-resolved PDOS of LVO/KTO Interface
(layer: -2)

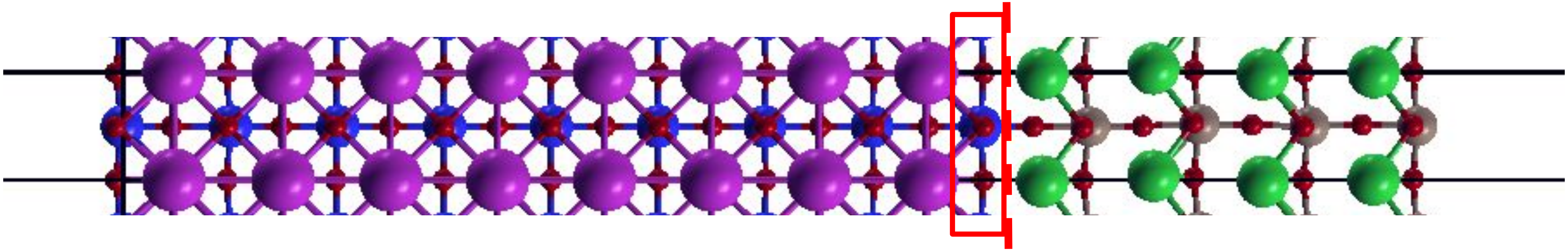


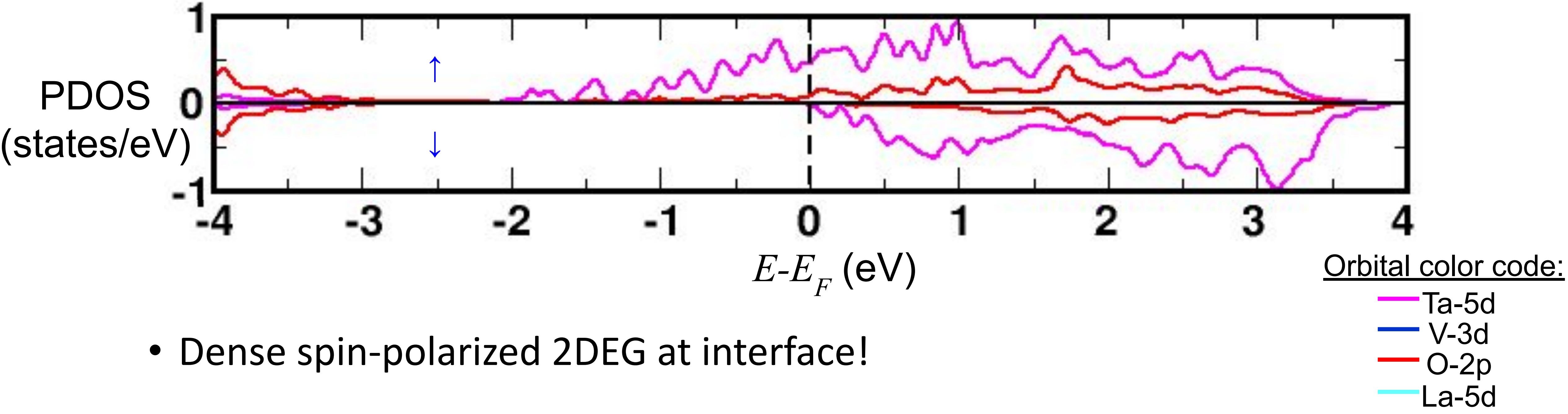


Orbital color code:
- Ta-5d
- V-3d
- O-2p
- La-5d

• Insulating.

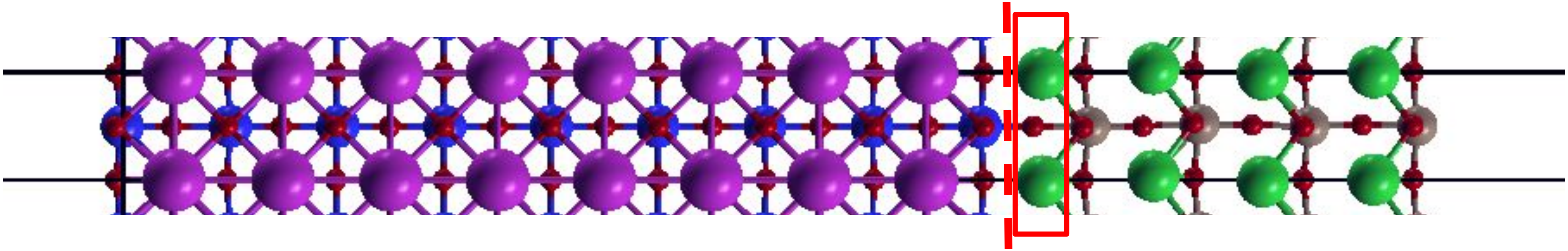
Layer-resolved PDOS of LVO/KTO Interface (layer: -1)

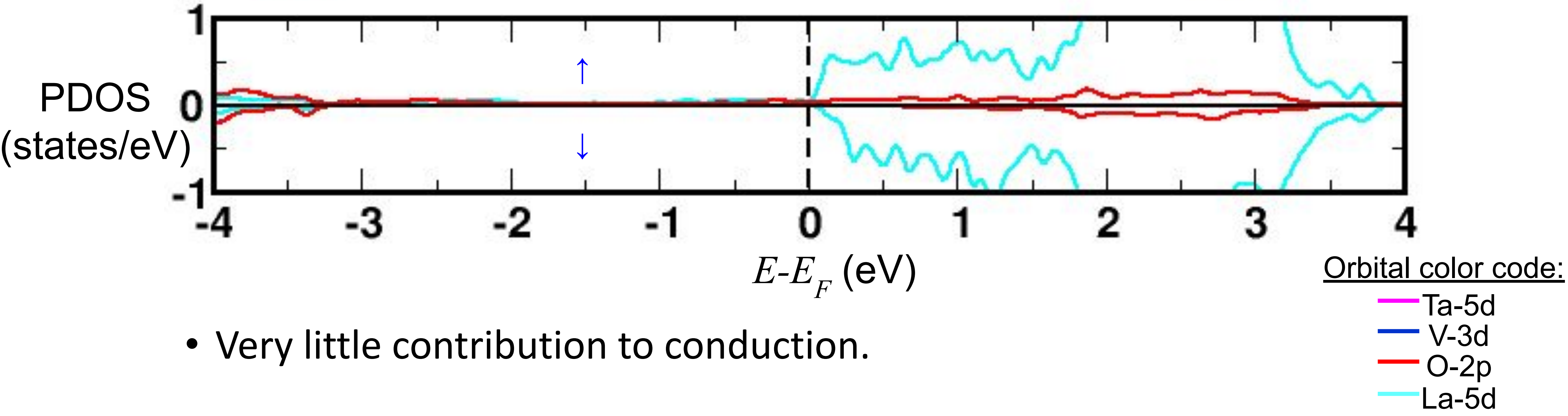




- Dense spin-polarized 2DEG at interface!

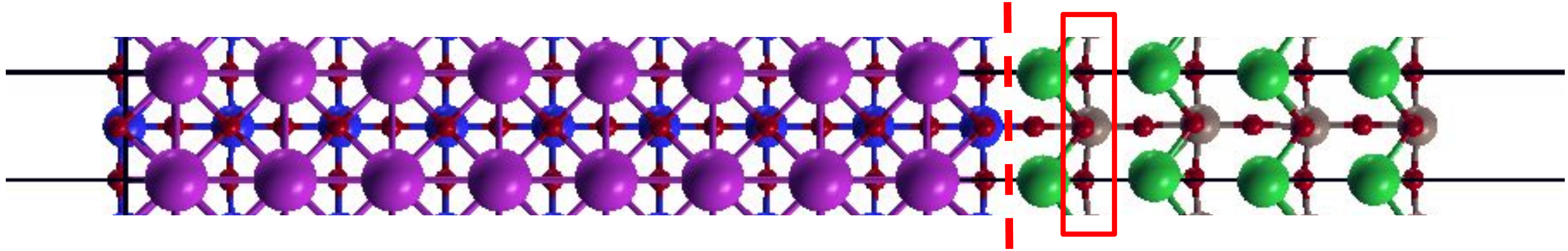
Layer-resolved PDOS of LVO/KTO Interface
(layer: 1)

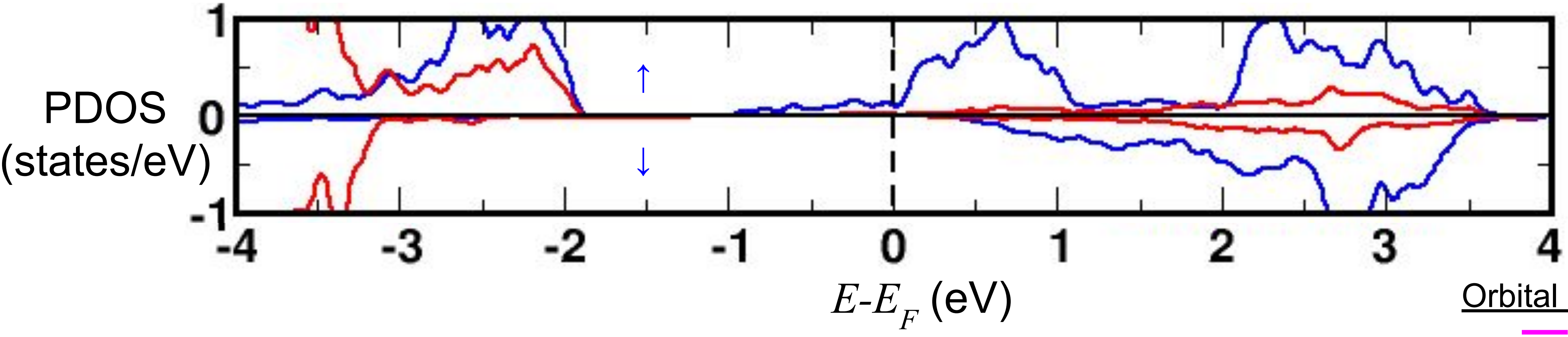




- Very little contribution to conduction.

Layer-resolved PDOS of LVO/KTO Interface
(layer: 2)

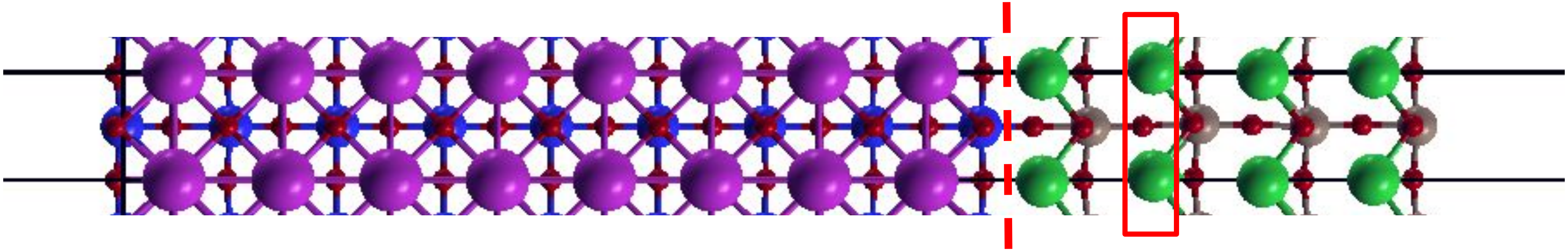


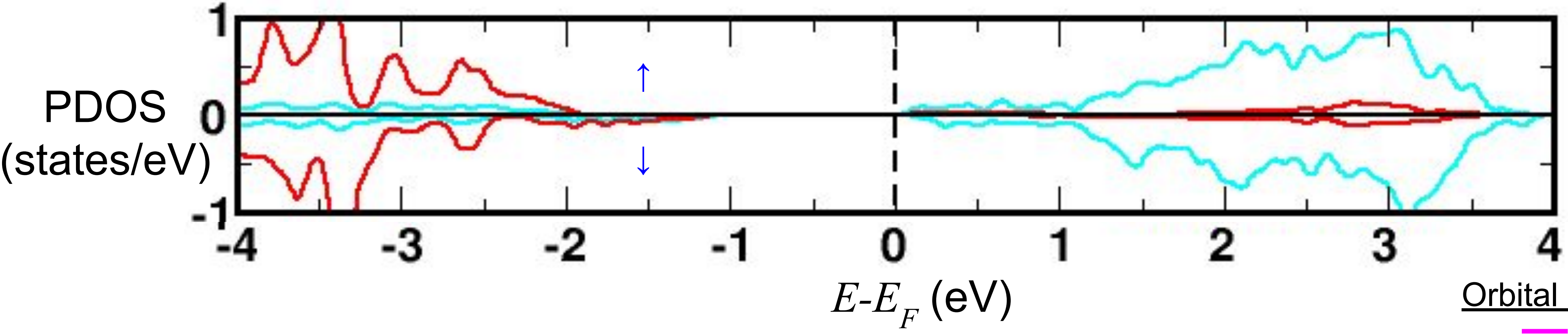


Orbital color code:
Ta-5d
V-3d
O-2p
La-5d

- Conducting → little contribution to 2DEG.
- Mainly V-3*d* orbitals hosts the 2DEG in this layer.

Layer-resolved PDOS of LVO/KTO Interface
(layer: 3)

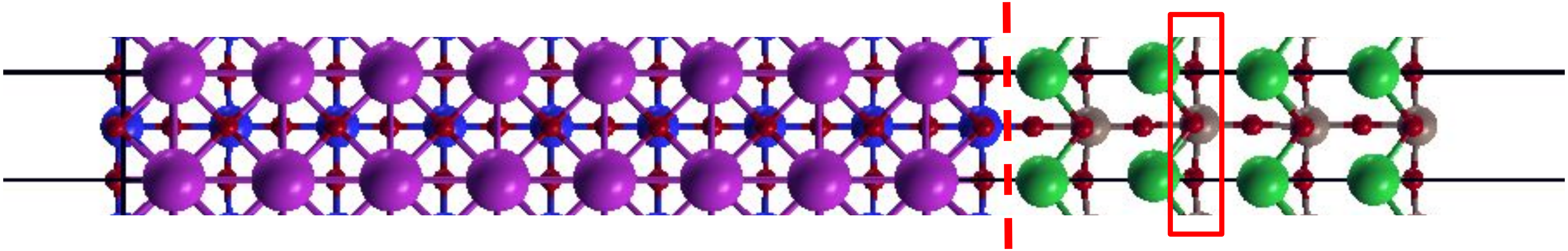


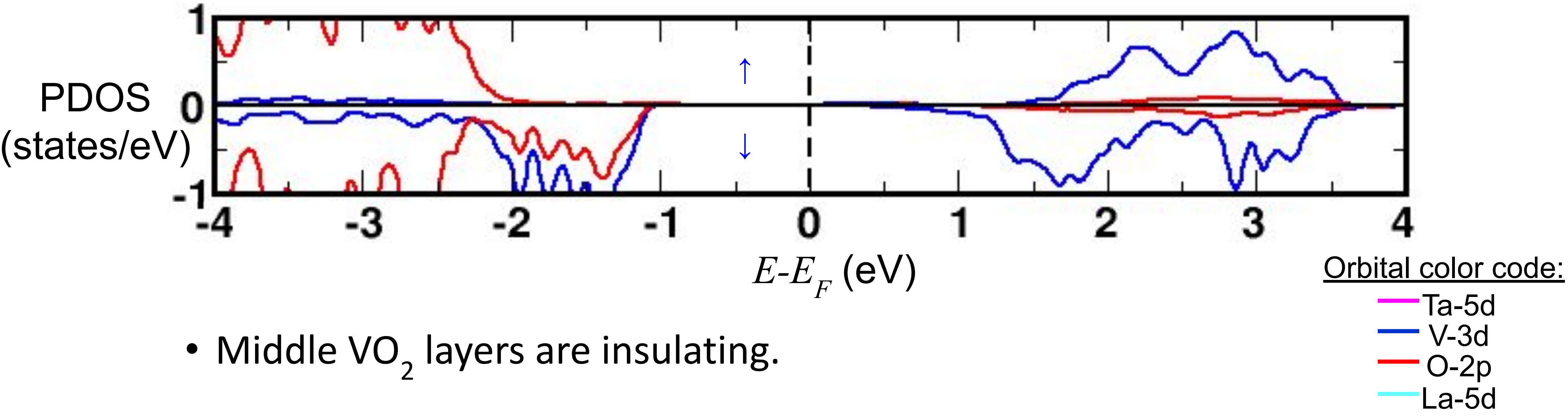


Orbital color code:
- Ta-5d
- V-3d
- O-2p
- La-5d

- Middle LaO layers are insulating.

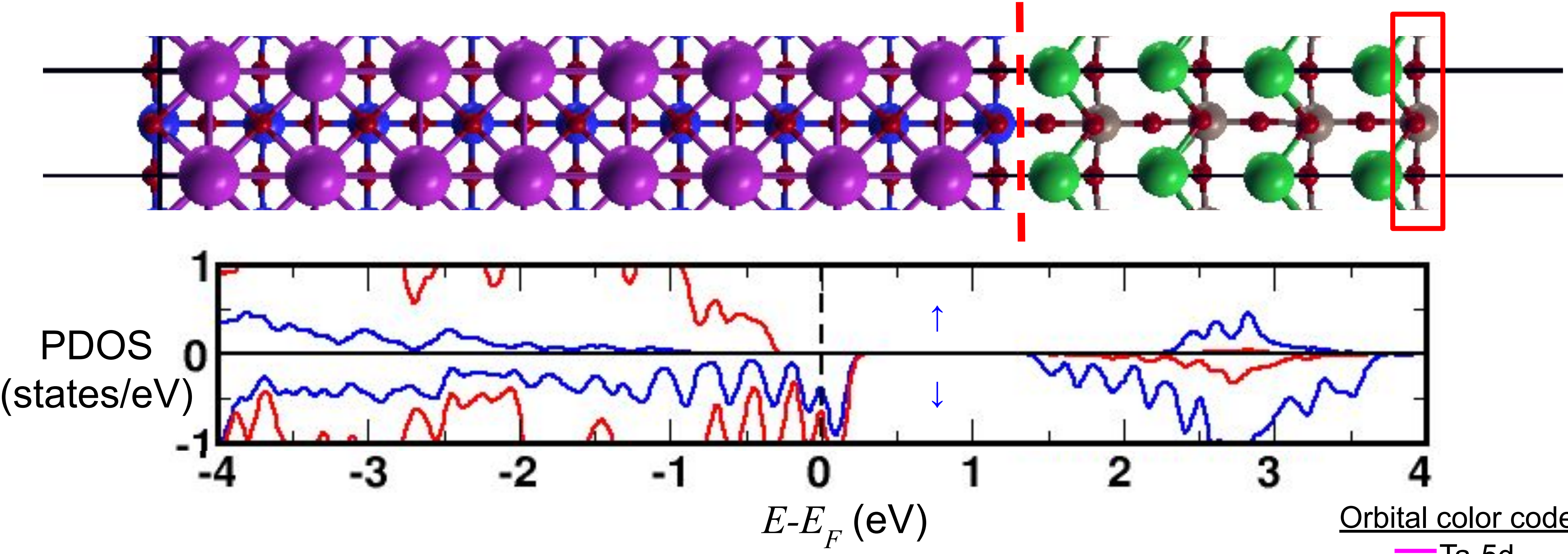
Layer-resolved PDOS of LVO/KTO Interface (layer: 4)





- Middle VO_2 layers are insulating.

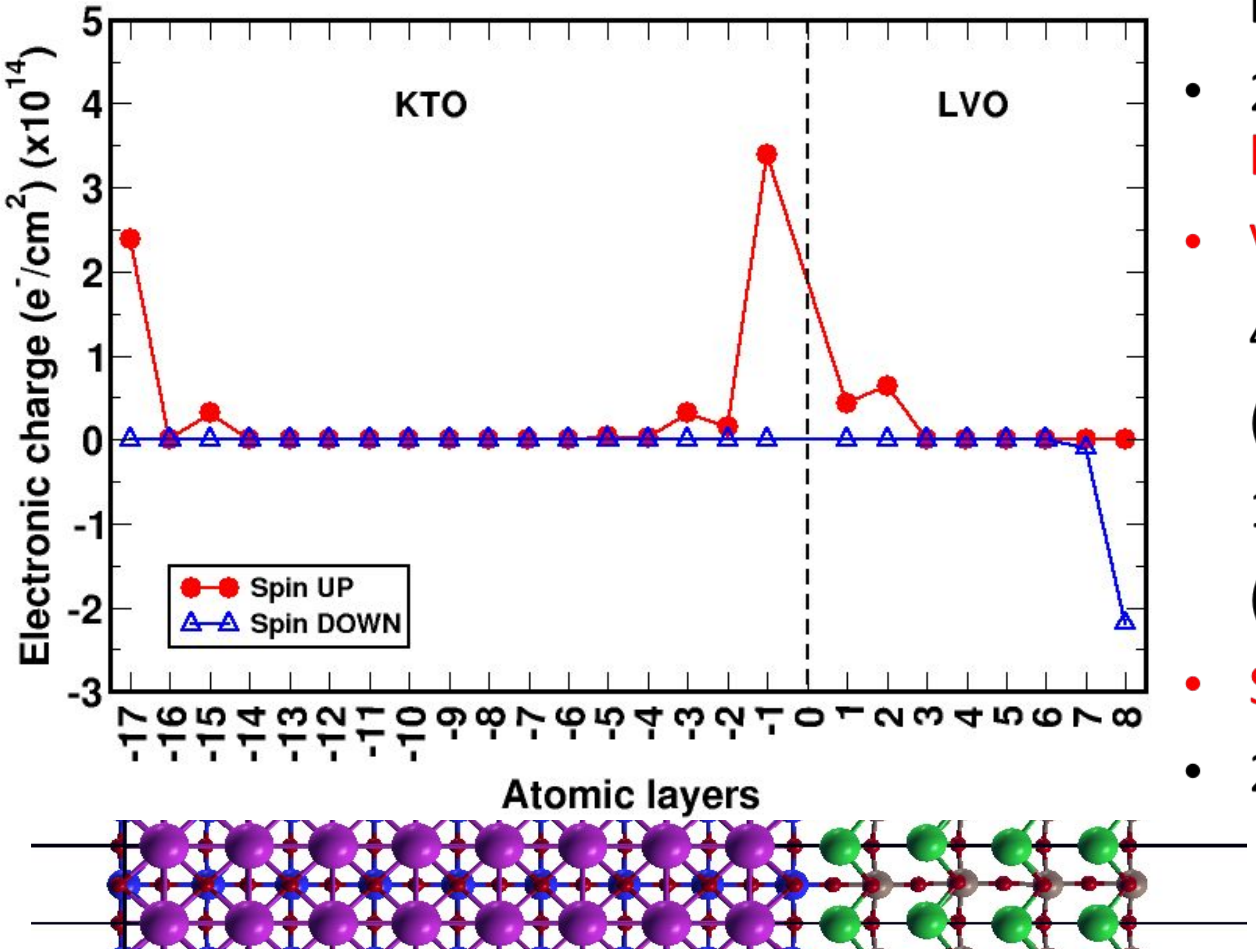
Layer-resolved PDOS of LVO/KTO Interface (layer: 8)



Orbital color code:
- Ta-5d
- V-3d
- O-2p
- La-5d

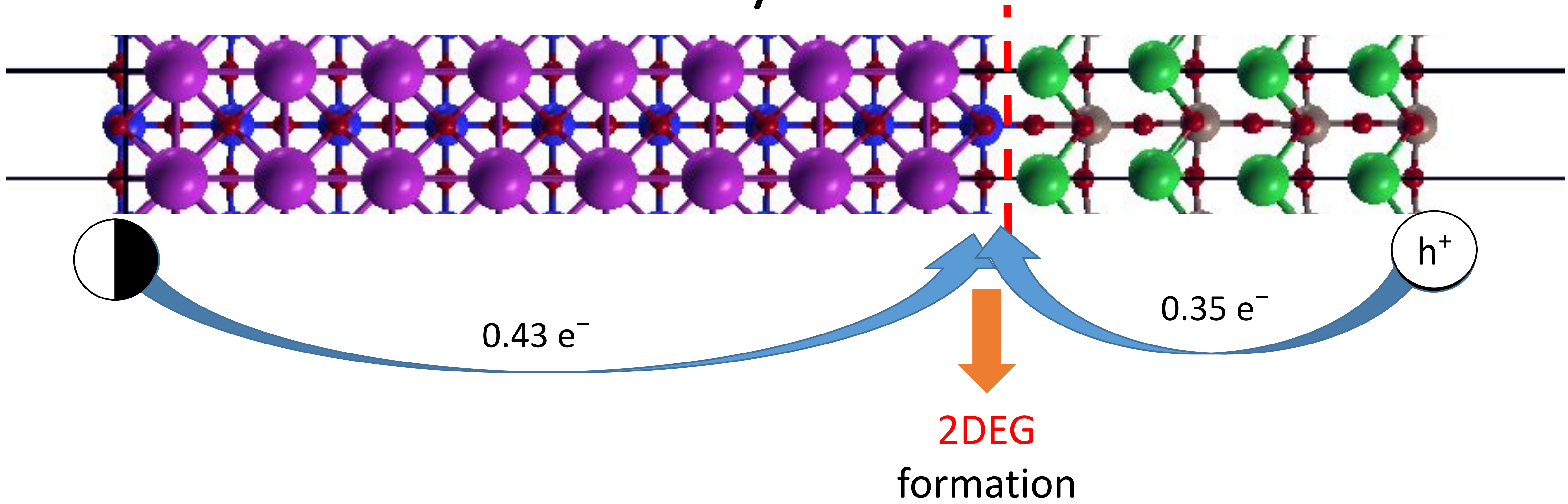
- Spin-polarized 2DHG at LVO surface.

Layer-resolved Electronic Charge



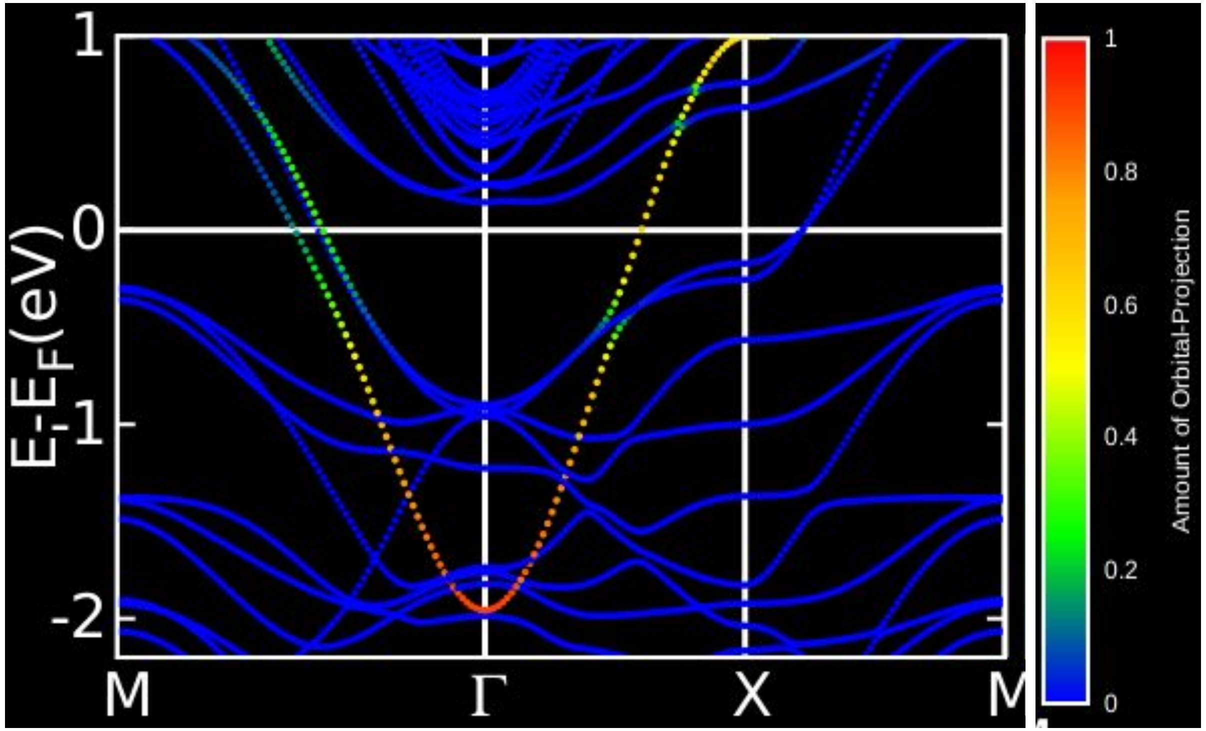
- Charge is calculated by integrating layer-resolved PDOS.
- 2DEG is primarily hosted by Interfacial TaO_2 layer on KTO side.
- Very high interfacial free e^- density = 4.89×10^{14} e^-/cm^2 (reasonable agreement w/ expt. value of 1×10^{14} e^-/cm^2) (at LAO/STO interface ~ 1×10^{13} e^-/cm^2)
- Spin-polarized 2DEG.
- 2DEG is extremely confined to the interface.

"Electronic Reconstruction" is the Origin of 2DEG at LVO/KTO Interface



- We indeed find charge contributed from both polar materials!

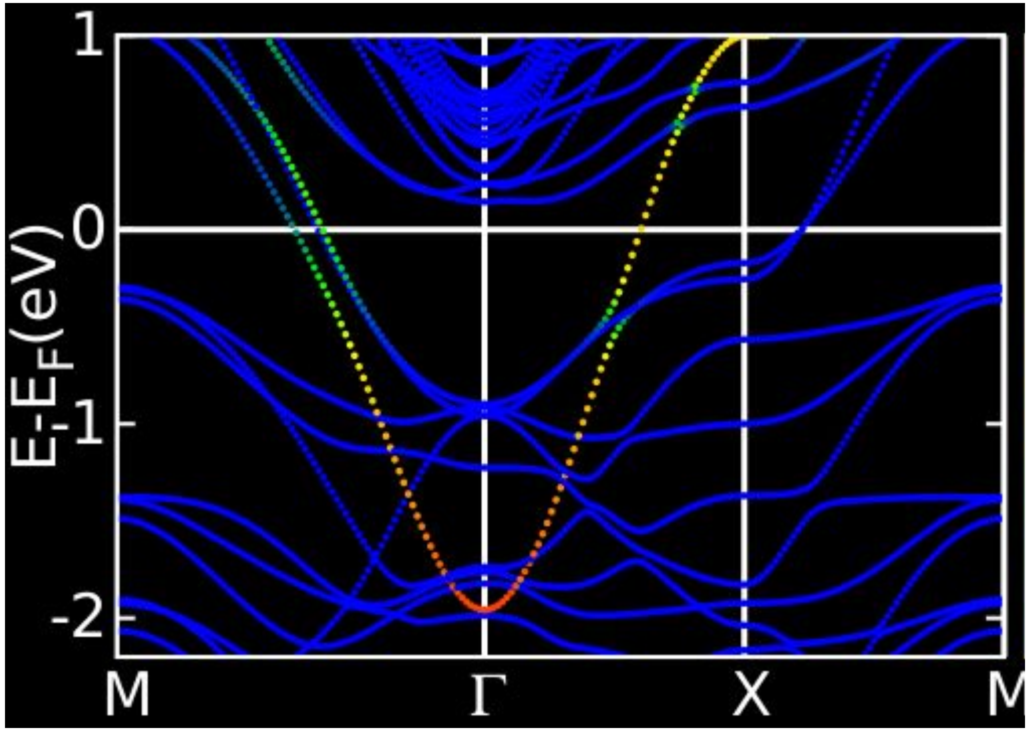
Projected Band Dispersion at Interface



Projected on interfacial Ta-5d_{xy} orbitals spin ↑

- Parabolic band confirms existence of free electrons forming 2DEG; primarily hosted by interfacial Ta-5d_{xy} orbitals.

Effective Mass & Mobility of 2DEG Electrons



Projected on interfacial Ta-5d_{xy} orbitals spin ↑

- 2DEG at the interface forms a parabolic band.

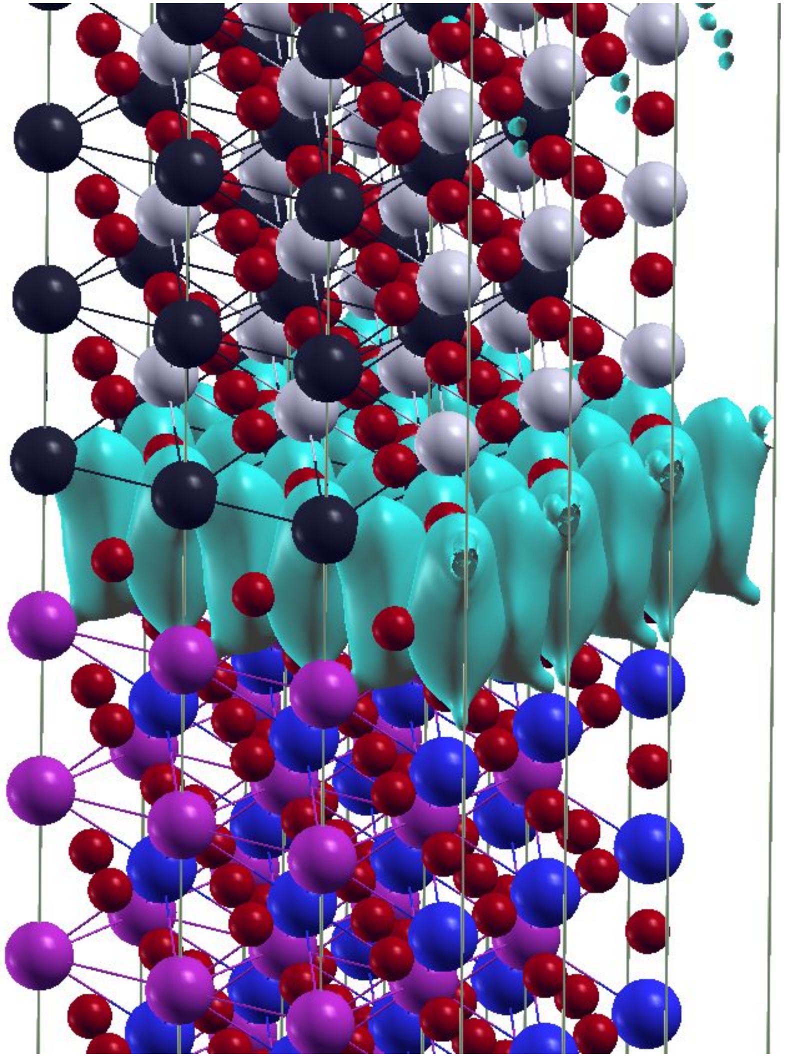
- Electron effective mass:

$$m^* = \frac{\hbar^2}{\frac{d^2E}{dk^2}} = 0.11m_0.$$

Much lower than $m^* = 0.4 - 0.6m_0$ for LAO/STO.

- Results in Higher mobility ($\mu = \frac{e\tau}{m^*}$).

Band-decomposed Charge Density: 2DEG at LVO/KTO Interface



- Band-decomposed charge density for parabolic band hosting 2DEG at Γ point.
- Cyan lobes correspond to iso-surface of charge density $|\psi_{n\boldsymbol{k}\uparrow}(\boldsymbol{r})|^2$ with iso-value = 0.0001 e^-/bohr^3.
- Cyan lobes showing 2DEG.
- Electrons are confined and free to move in plane parallel to interface; cannot move perpendicular to interface.

Conclusions

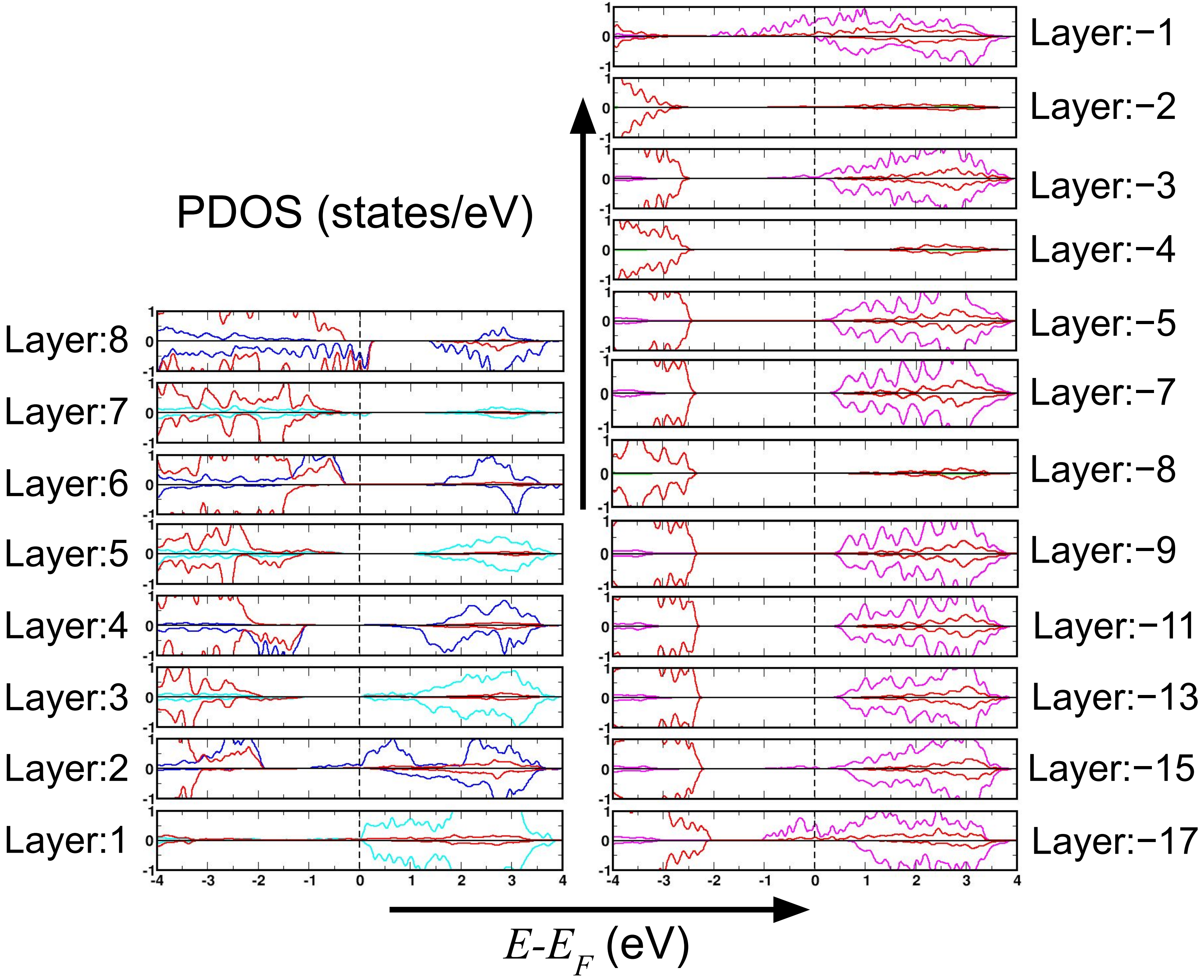
- Very thin layer of spin-polarized 2DEG formed at the interface between KTO (non-magnetic) and LVO (A-AFM).
- 2DEG has higher carrier density and lower electron effective mass expect higher mobility than previously studied systems.
- Carrier density is higher in 2DEG because of charge contributed from both (polar) LVO and (polar) KTO.

Thank You

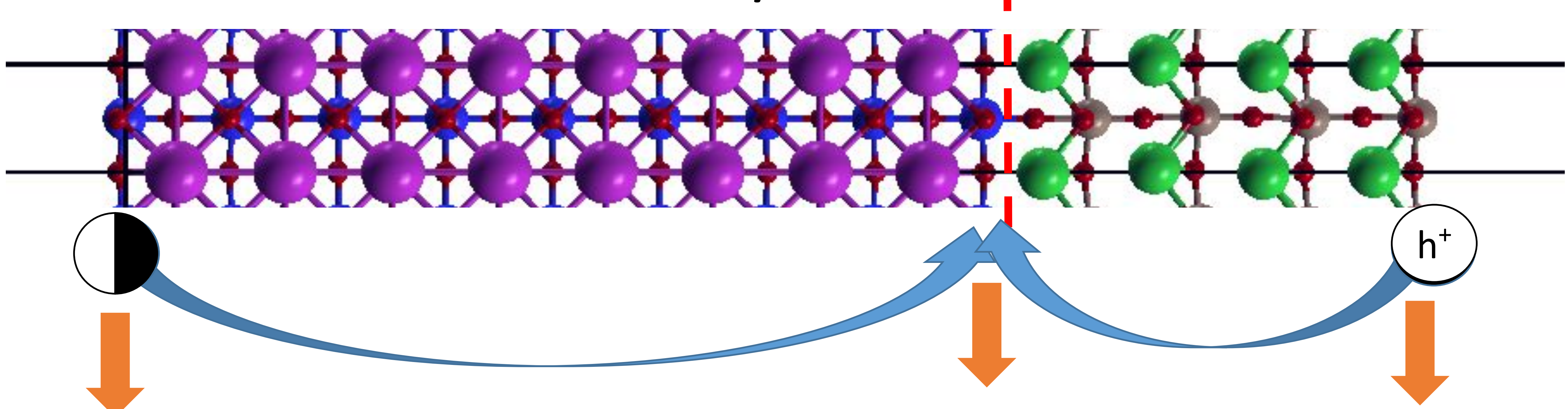
Backup Slides

Conclusions

- LVO/KTO interface shows metallicity though the constituents are insulators.
- Parabolic bands around the Γ point cross the Fermi level which shows the existence of conduction/free electrons.
- Electrons are transferred from surface of both LVO and KTO to interfacial TaO_2 layer 'electronic reconstruction' is the origin of 2DEG.
- Calculated interfacial electron density agrees well with expt.
- Higher electron mobility because of lower effective mass, due to less localized Ta-5d orbitals compared to Ti-3d orbitals.
- Conduction electrons forming the 2DEG primarily reside on interfacial TaO_2 layer.
- 2DEG is spin-polarized.
- 2DEG is extremely confined or localized to the interface.
- Higher interfacial carrier density and higher mobility of 2DEG compared to polar/non-polar oxide interfaces.


PDOS (states/eV)
Layer:8
Layer:7
Layer:6
Layer:5
Layer:4
Layer:3
Layer:2
Layer:1
Layer:−1
Layer:−2
Layer:−3
Layer:−4
Layer:−5
Layer:−7
Layer:−8
Layer:−9
Layer:−11
Layer:−13
Layer:−15
Layer:−17
E-E_F (eV)

“Electronic Reconstruction” is the Origin of 2DEG at LVO/KTO Interface



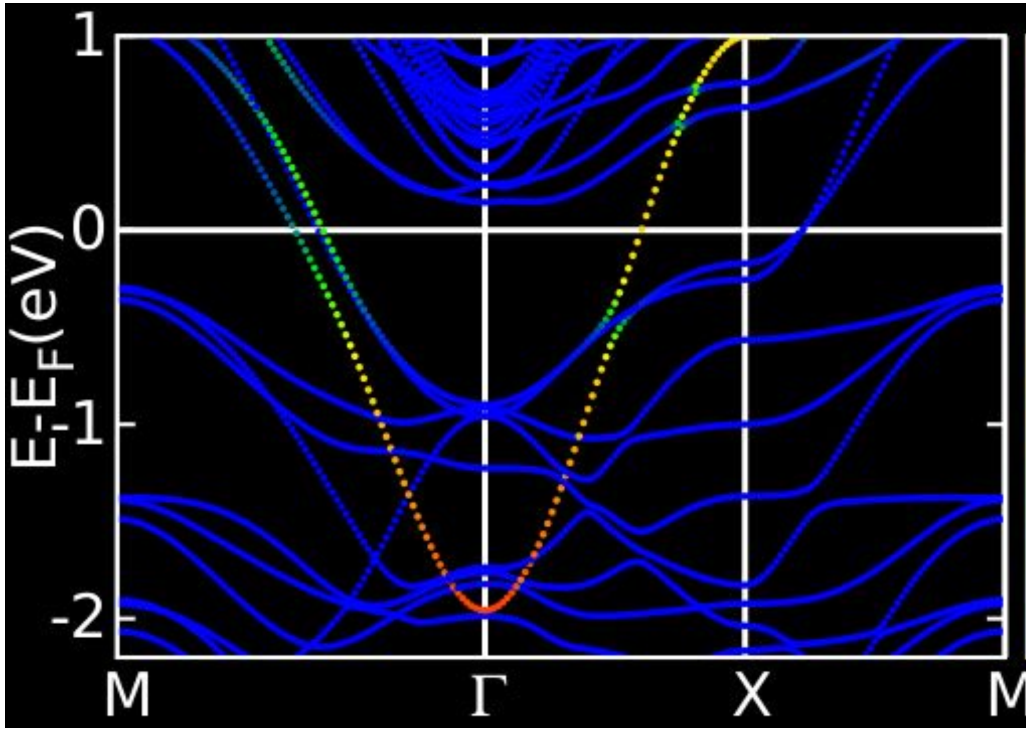
- CBM of surface TaO_2 layer move up in energy.
- Free e^- density at KTO surface decreases.

2DEG formation

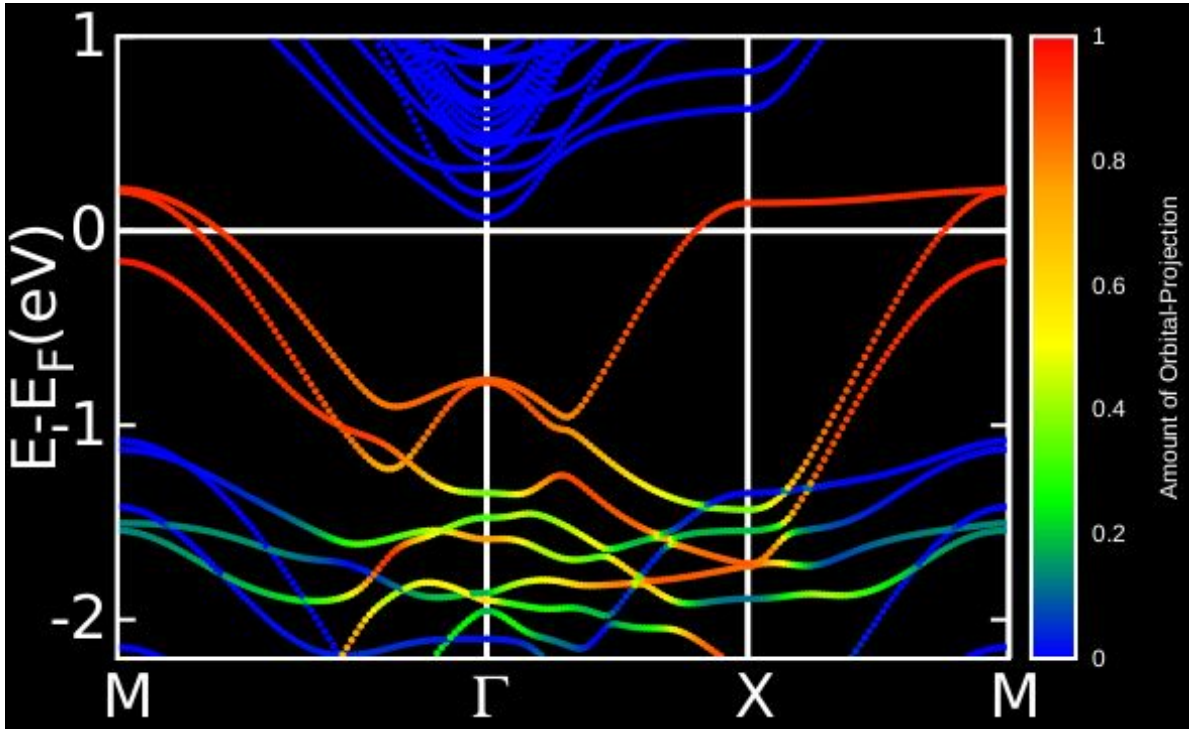
LVO/KTO being polar/polar, both LVO & KTO donate e^- to the interface □ higher e^- density compared to polar/non-polar LAO/STO interface.

- Valence bands of surface VO_2 layer move up in energy & cross Fermi level.
- Creation of holes.

Orbital Projected Band Dispersion



Projected on interfacial Ta-5d_{xy} orbitals spin ↑



Projected on surface VO_2 layer spin ↓

- Parabolic band confirms existence of free electrons forming 2DEG; primarily hosted by interfacial Ta-5d_{xy} orbitals.
- Surface valence V-3*d* and O-2*p* states cross the Fermi level: they donate electrons to the interface.