Center for Inverse Design

Achieving the grand challenge of materials and nanostructures by design

Understanding the Conductivity and Magnetism at Interfaces of Insulating Nonmagnetic Oxides: The case of $SrTiO_3/LaAlO_3$

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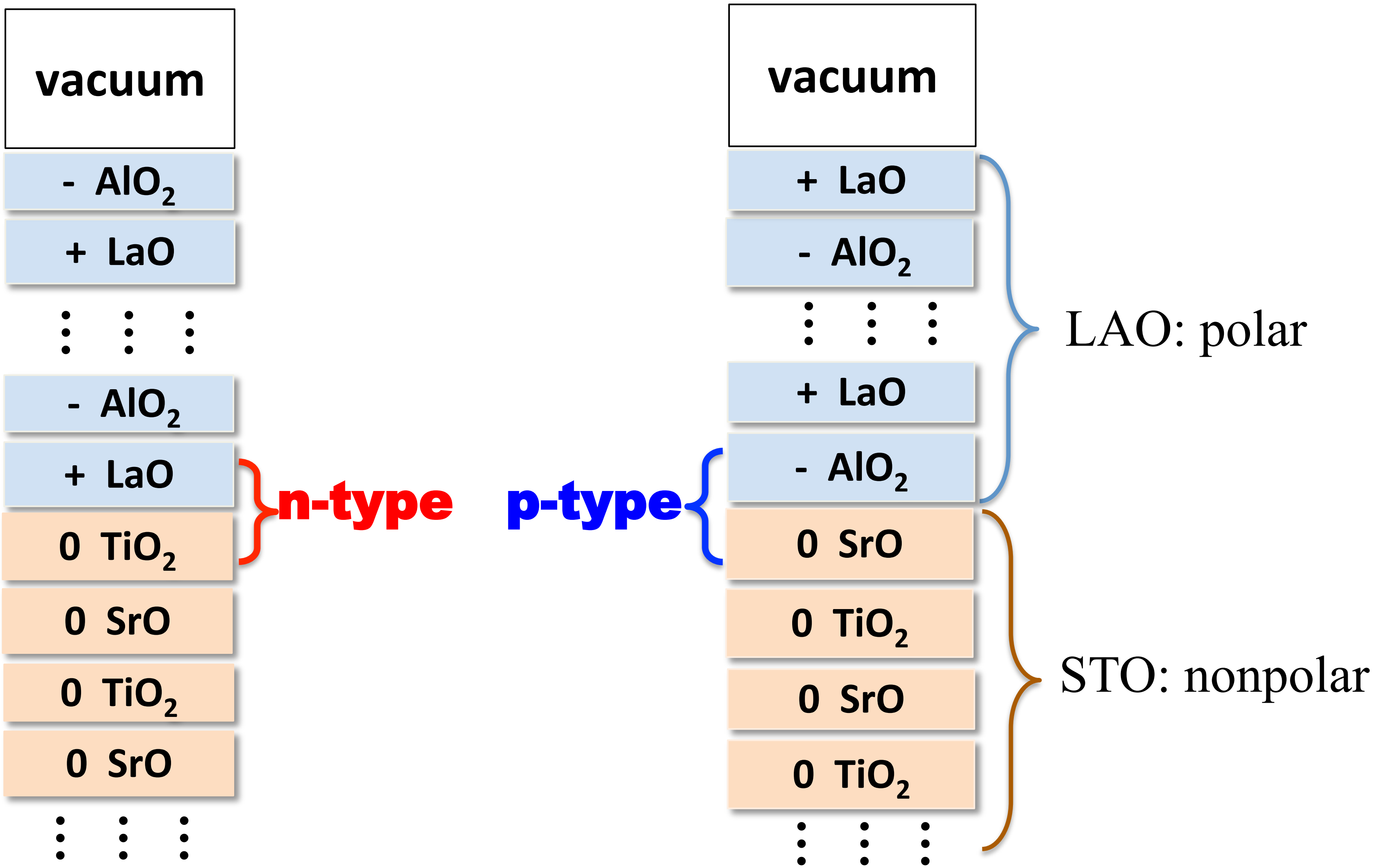
Two types of (001) interfaces



Six Leading Puzzles

① Where does 2DEG come from?

② What controls critical thickness?

③ What compensate electric field?

④ What controls 2DEG density?

⑤ What causes interface magnetism?

⑥ Why are p-interfaces insulating and magnetic?

A Unifying Mechanism

Polar Discontinuity across the interface

→ electric field in polar LAO film

→ spontaneous formation of defects
at LAO surface and STO/LAO interface

→ conductivity & magnetism @ interface

Puzzle 1

① Where does 2DEG come from?

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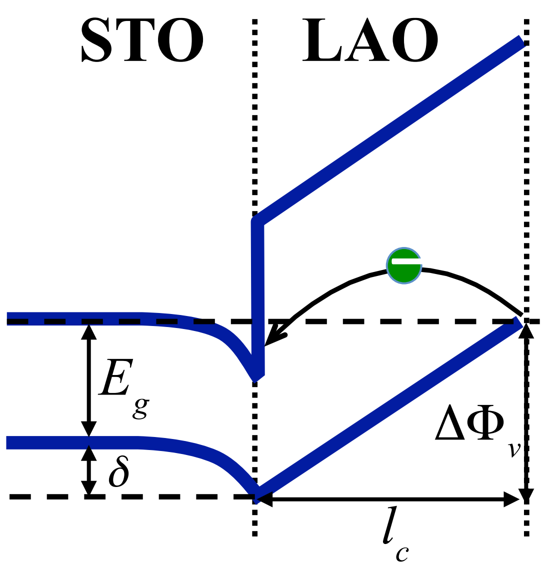
③ What compensate electric field?

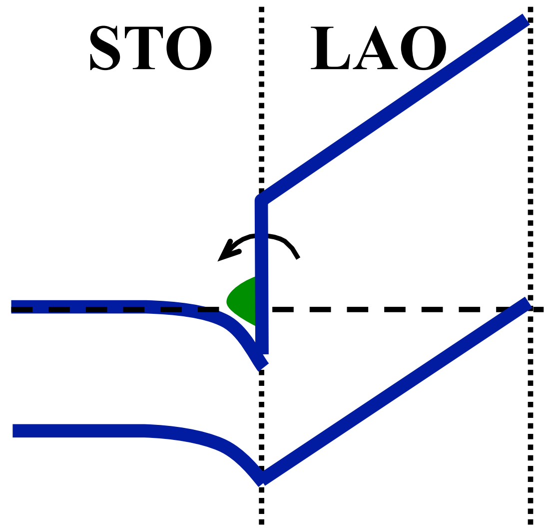
④ What controls 2DEG density?

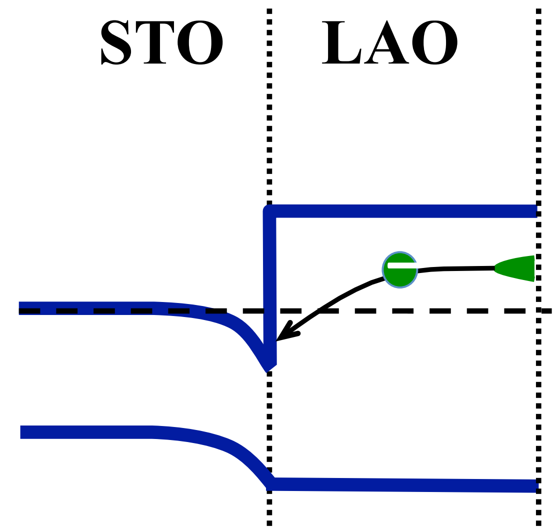
⑤ What causes interface magnetism?

⑥ Why are p-interfaces insulating and magnetic?

Four Proposed Mechanisms







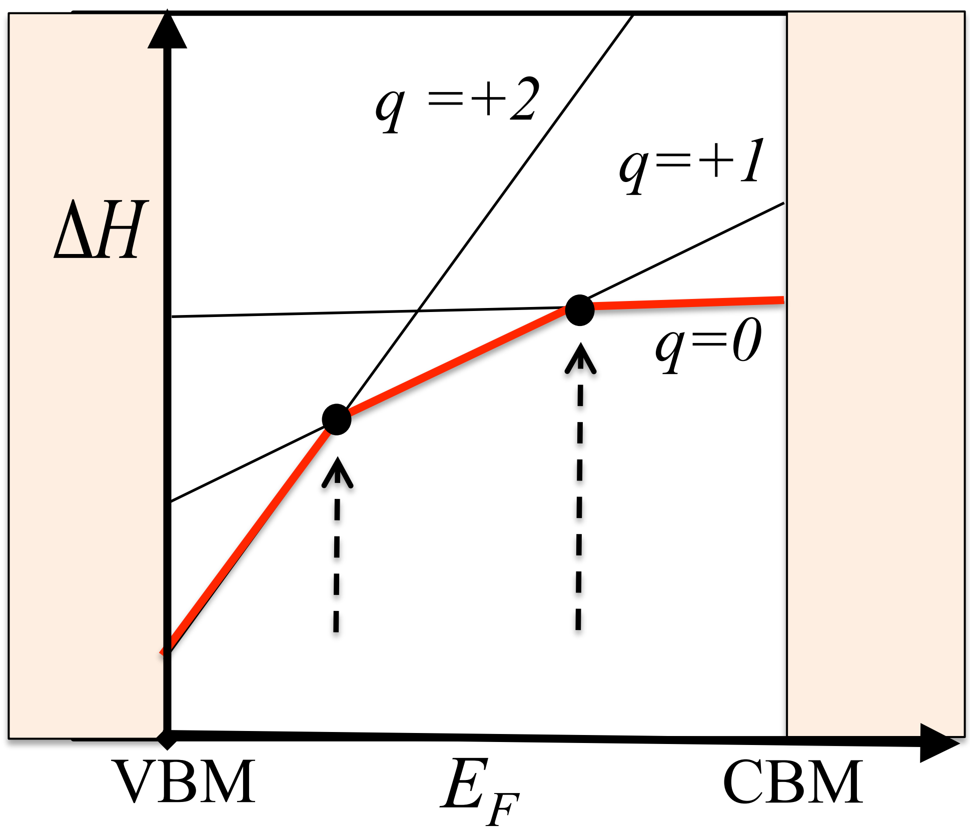
~~(1)~~ Polar catastrophe

(2) Interface V_O
(3) Interface La_{Sr}

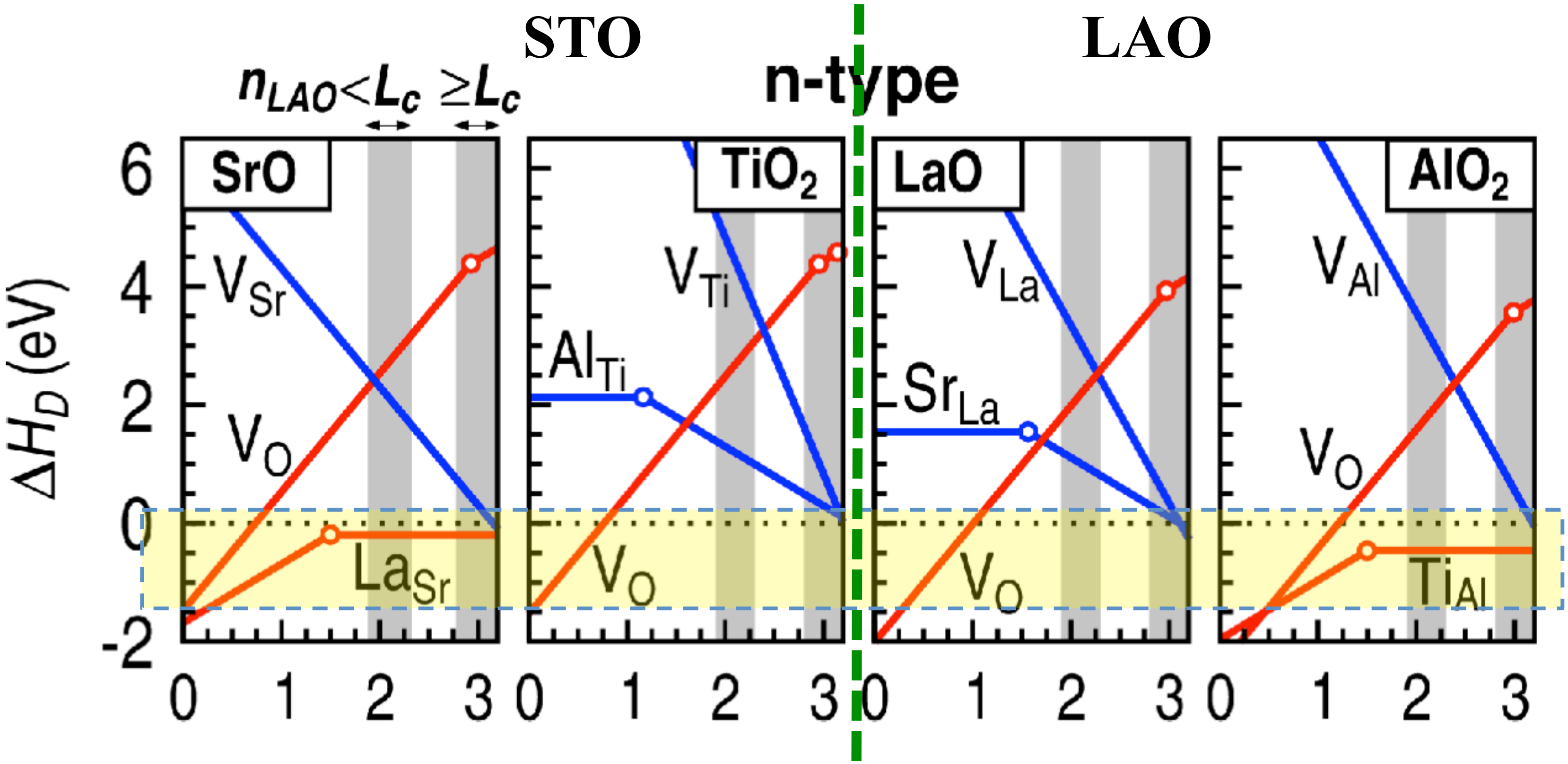
(4) Surface V_O

Method: first principles defect theory

$$\Delta H_D^q = E_D^q - E_H + \sum_\alpha n_\alpha \mu_\alpha + qE_F$$

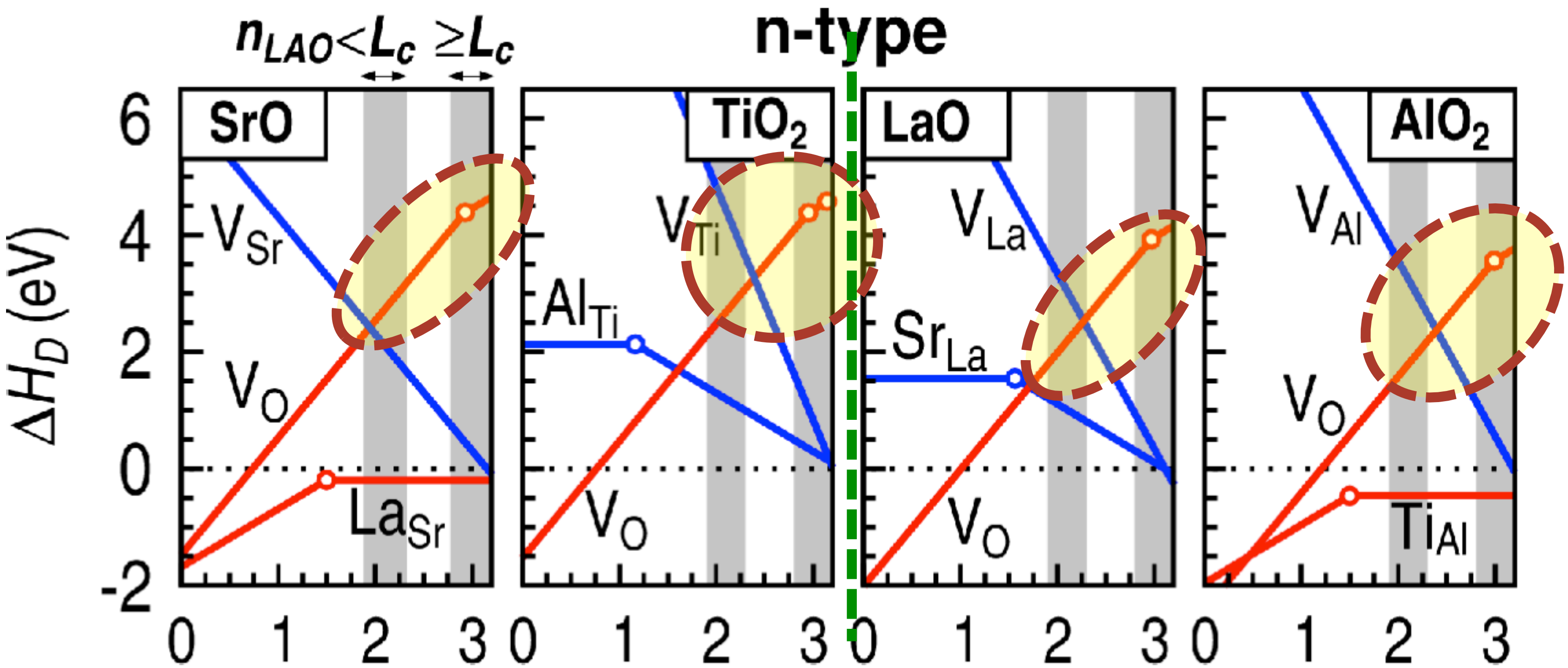


Interface La_{Sr} and Ti_{Al} donor defects are too deep to cause free carriers



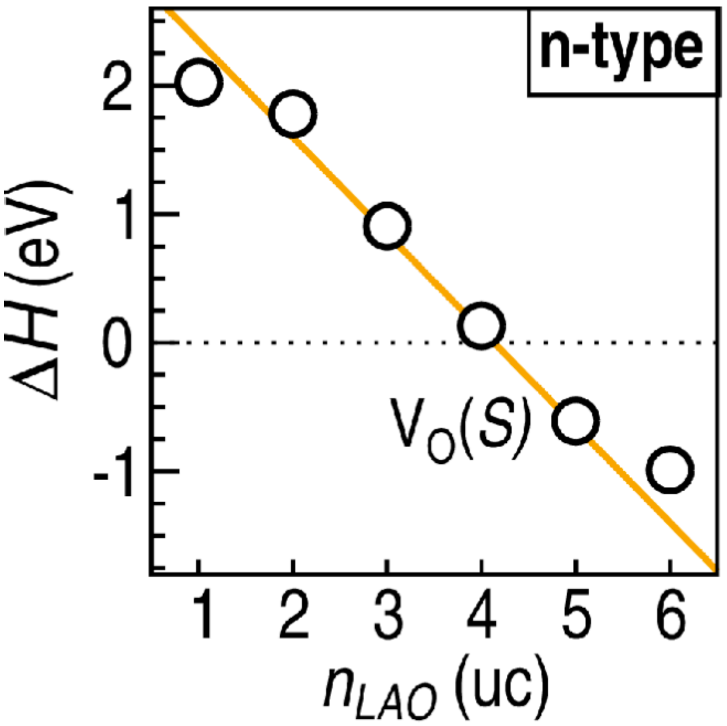
- Negative formation energies, thermodynamically unstable, cause cation mixing

Interface V_O defects do not contribute free carriers either



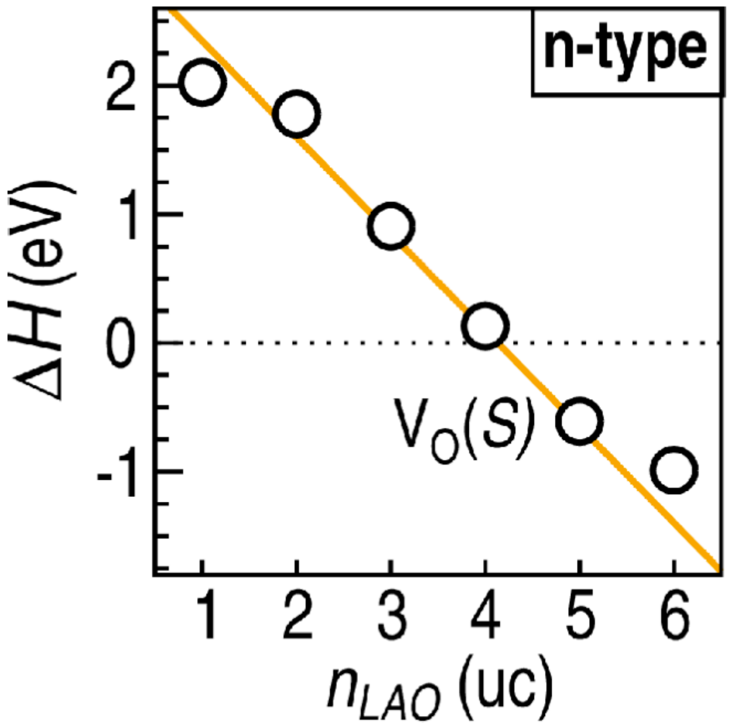
- Too high formation energy (> 3 eV)
- Shallow and independent of its layer location

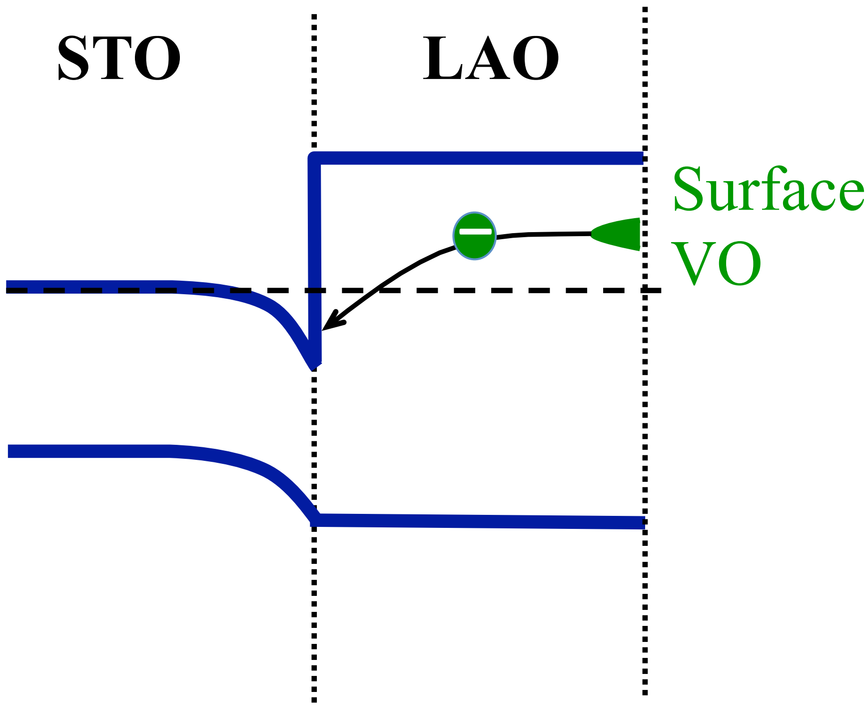
Surface V_O can form spontaneously when $n_{LAO} \geq \sim 4$uc



- Formation energy decreases linearly

Linear decreasing: a sign of charge transfer

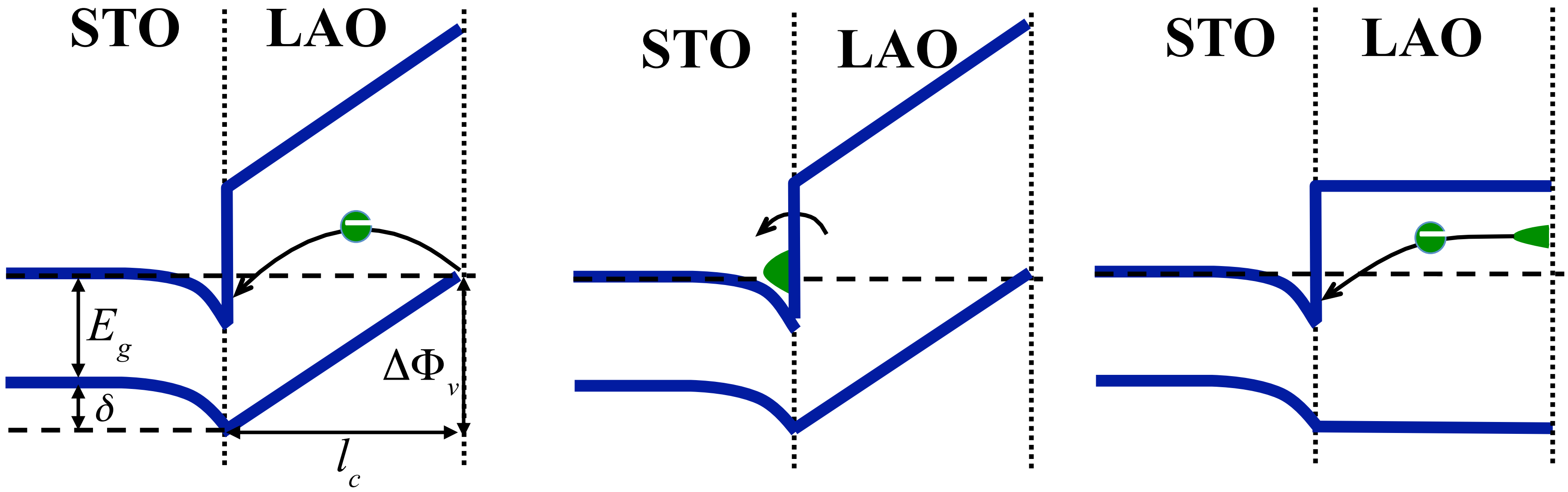




- Formation energy decreases linearly

- V_O donor level is higher than CB edge at interface
- Existence of built-in electric field

Summary: previous models on the 2DEG origin



(1) Polar catastrophe

(2) Interface V_O
(3) Interface La_{Sr}

(4) Surface V_O

Puzzle 2

① Where does 2DEG come from?

② **What controls critical thickness?**

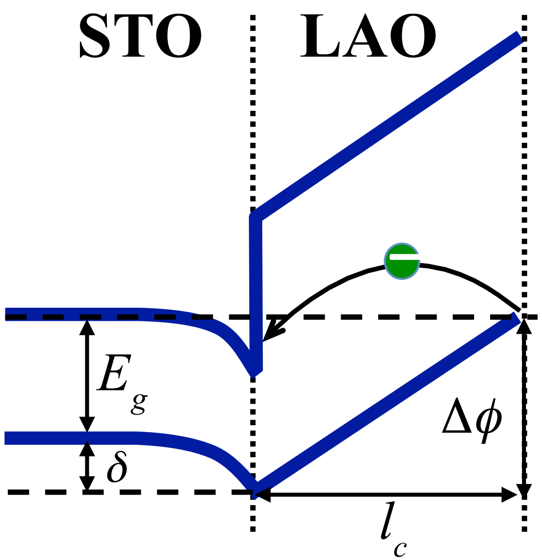
③ What compensate electric field?

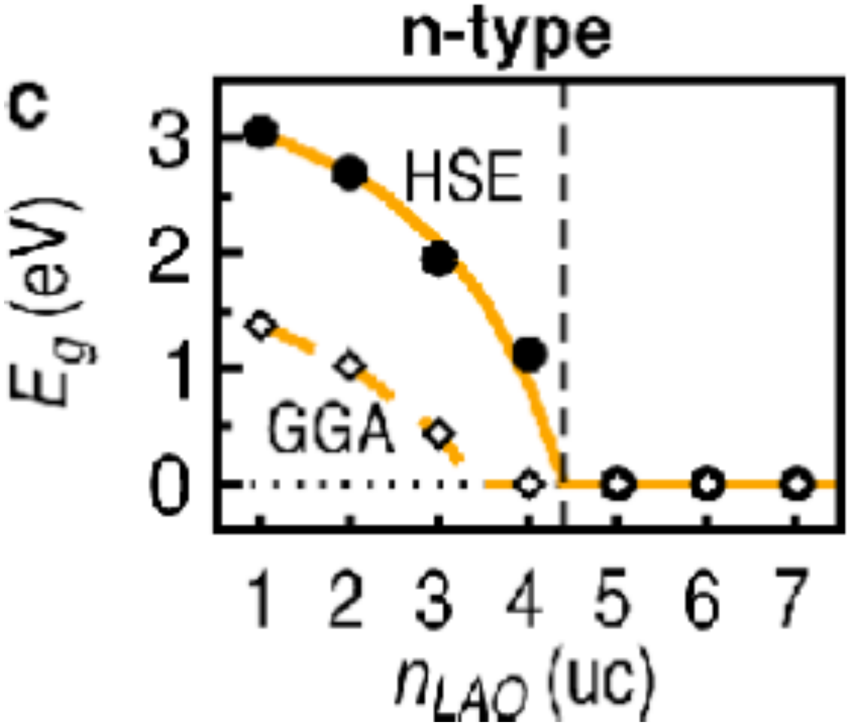
④ What controls 2DEG density?

⑤ What causes interface magnetism?

⑥ Why are p-interfaces insulating and magnetic?

Critical thickness from polar catastrophe model

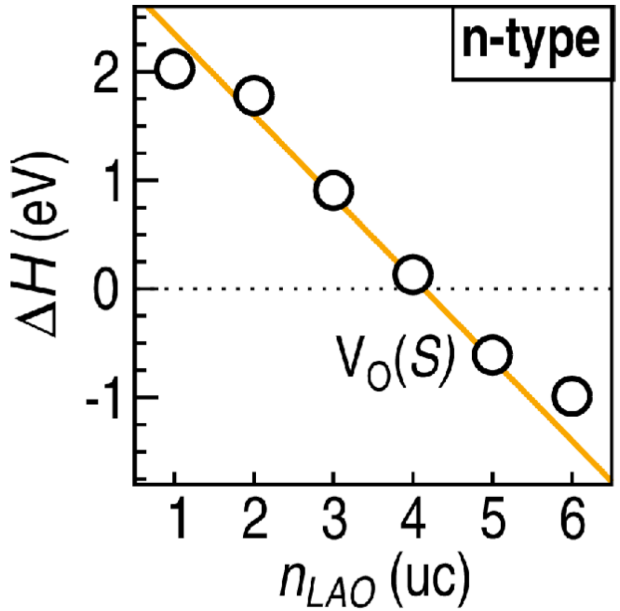


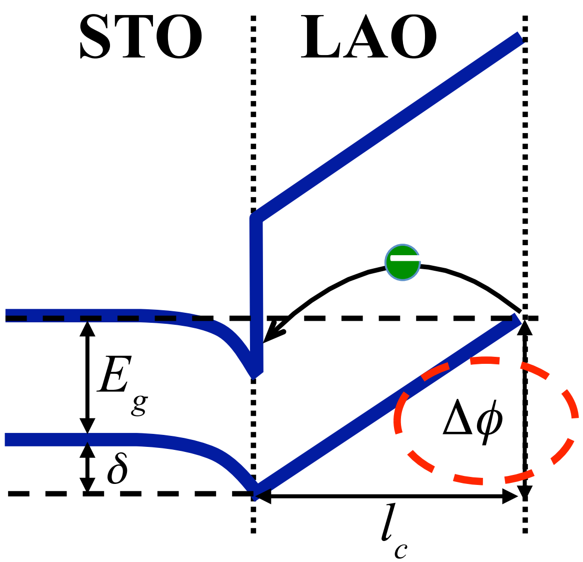


Electrostatics

$$l_c = \frac{\varepsilon_{LAO}\Delta\phi}{4\pi P^0_{LAO}}$$

Critical thickness from surface V_O model





Surface VO model:

$$l_c = \frac{\varepsilon_{LAO} \Delta H_{VO}}{4\pi P^0_{LAO}}$$

PC model:

$$l_c = \frac{\varepsilon_{LAO} \Delta \phi}{4\pi P^0_{LAO}}$$

Puzzle 3

① Where does 2DEG come from?

② What controls critical thickness?

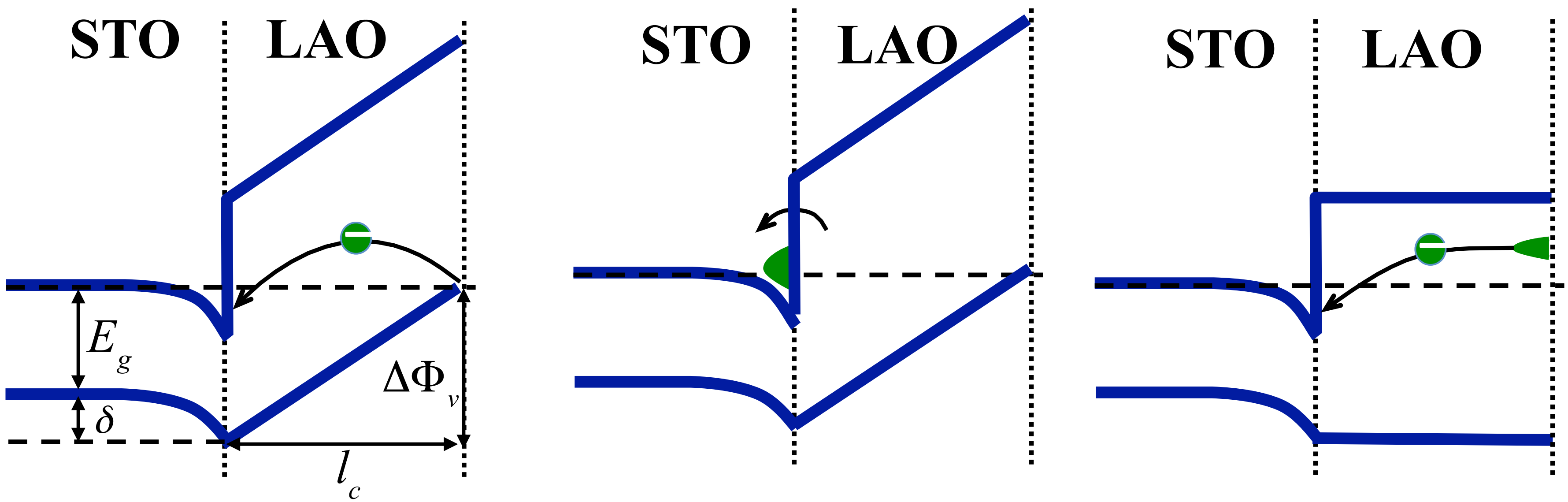
③ What compensate electric field?

④ What controls 2DEG density?

⑤ What causes interface magnetism?

⑥ Why are p-interfaces insulating and magnetic?

Four Existing Mechanisms

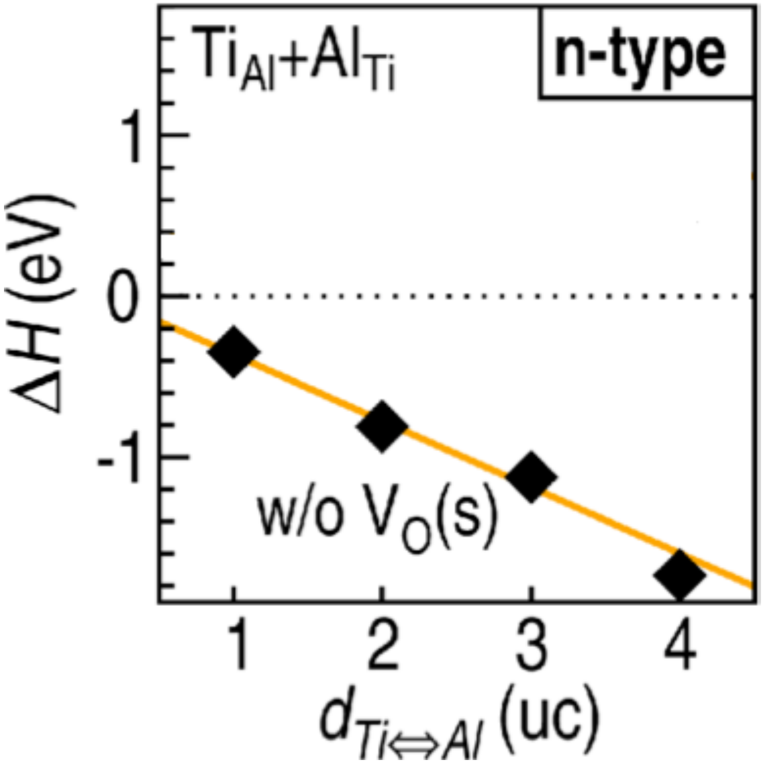


(1) Polar catastrophe

(2) Interface V_O
(3) Interface La_{Sr}

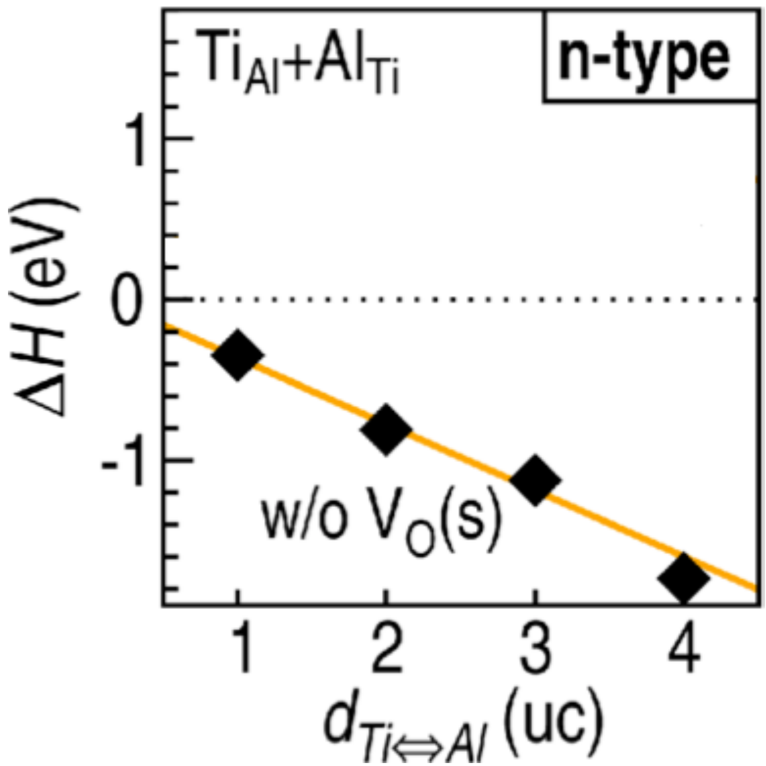
(4) Surface V_O

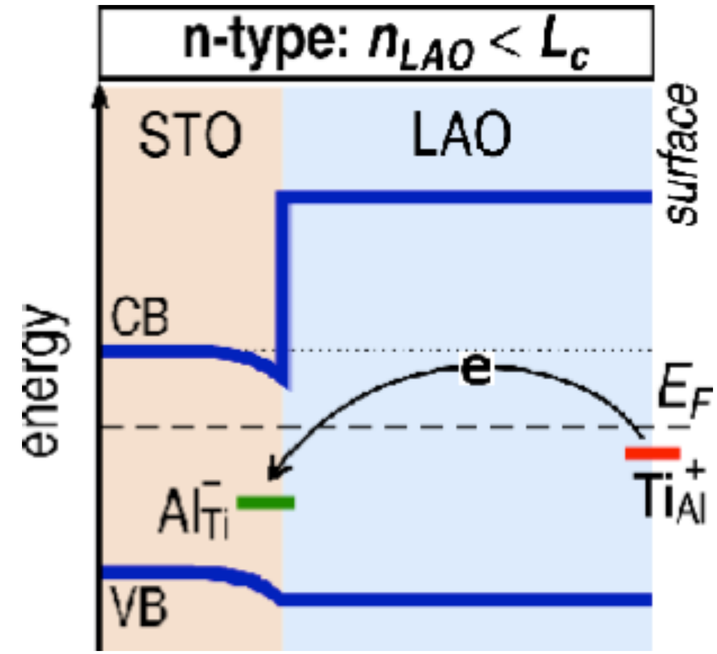
Why weak electric field below the Lc?



- Formation energy < 0
- Ti_{Al}(S) + Al_{Ti}(I) is most stable

Why weak electric field below the L_c?

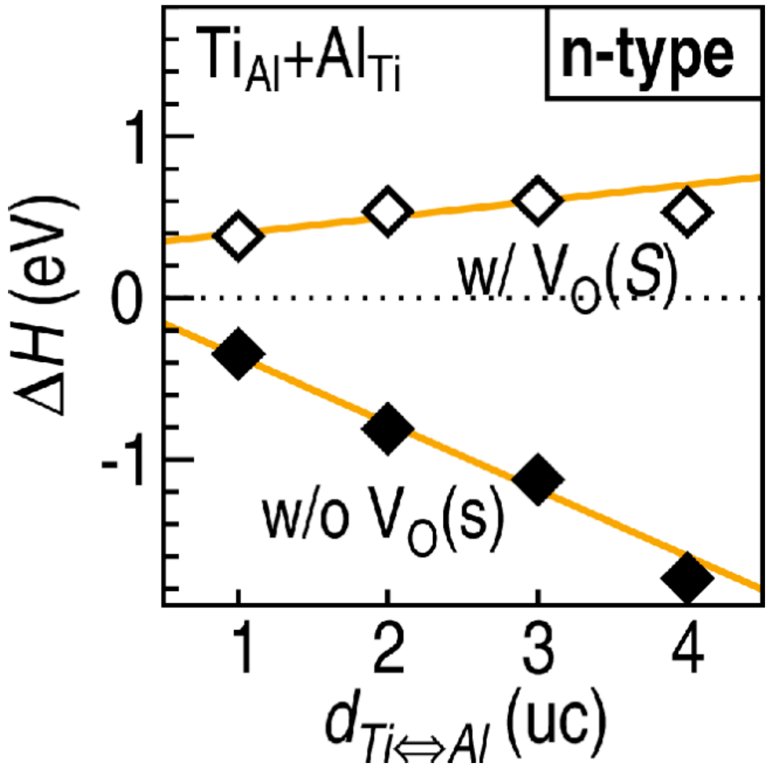


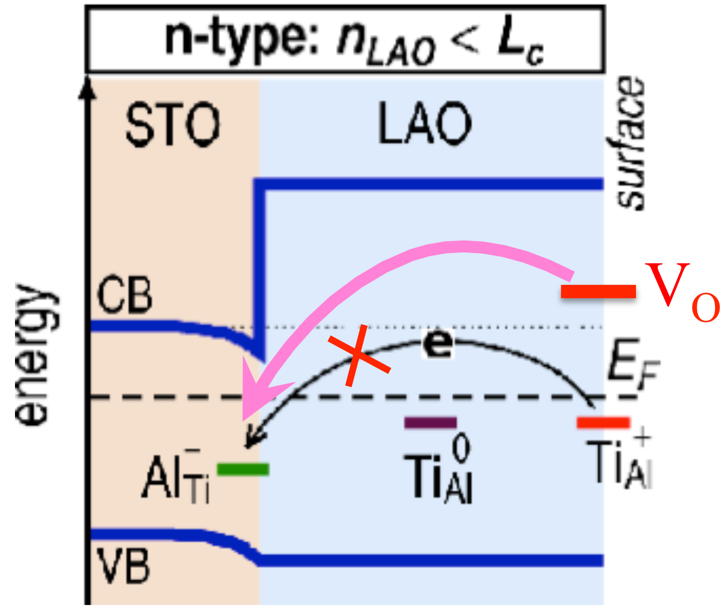


- Formation energy < 0
- Ti_{Al}(S) + Al_{Ti}(I) is most stable

- Donor level is higher than acceptor level
- Charge transfer cancels the built-in field, but induces no free carriers

What about Ti <-> Al exchange **above** the L_c?





- Positive formation energy

- "V_O → Al_{Ti}" is more energetically stable than "Ti_{Al} → Al_{Ti}"
- Interface Al_{TI} defects have lower concentration when $_VO$ present at surface

Puzzle 4

① Where does 2DEG come from?

② What controls critical thickness?

③ What compensate electric field?

④ What controls 2DEG density?

⑤ What causes interface magnetism?

⑥ Why are p-interfaces insulating and magnetic?

E~0 means ~0.5 e/uc interface charge

Gauss's law in electrostatics:

Maximum Q_e (at STO/LAO interface)

= D(LAO)-D(STO)

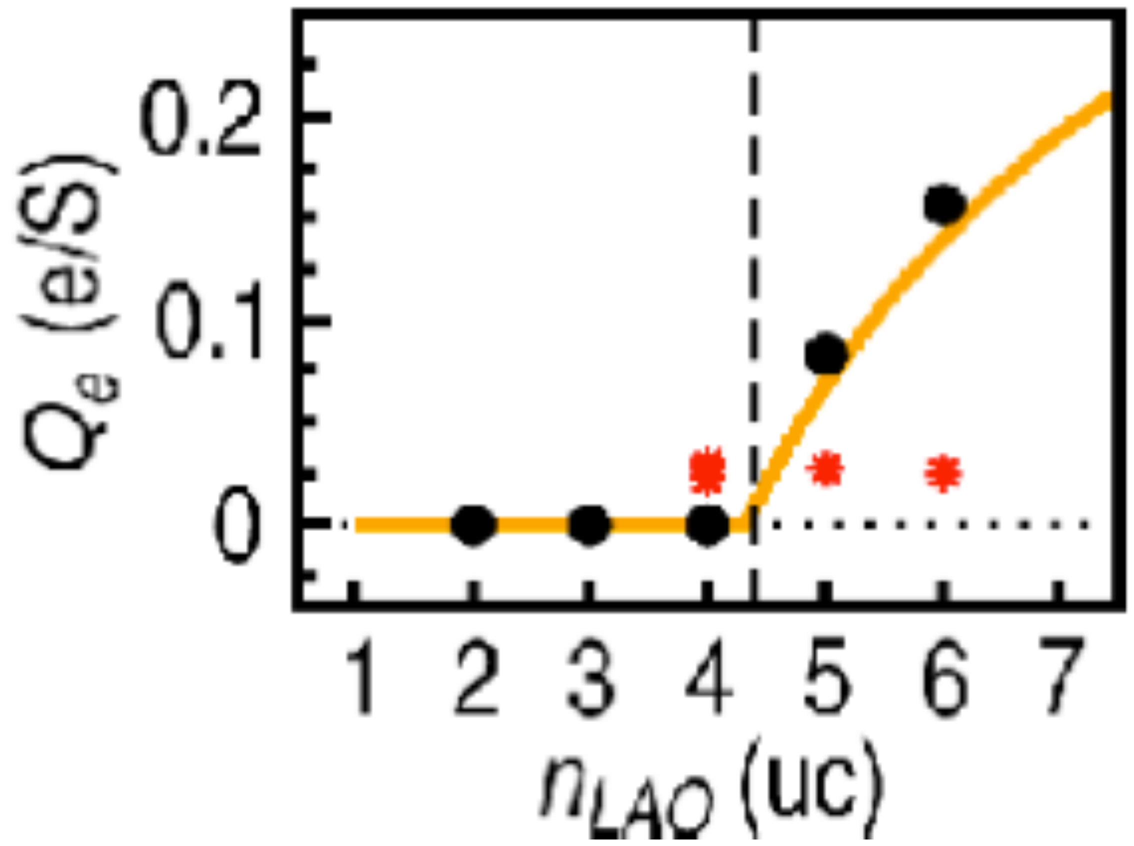
= D(LAO) - 0

= $E+4\pi P = 4\pi P_0$ (if E=0 in LAO)

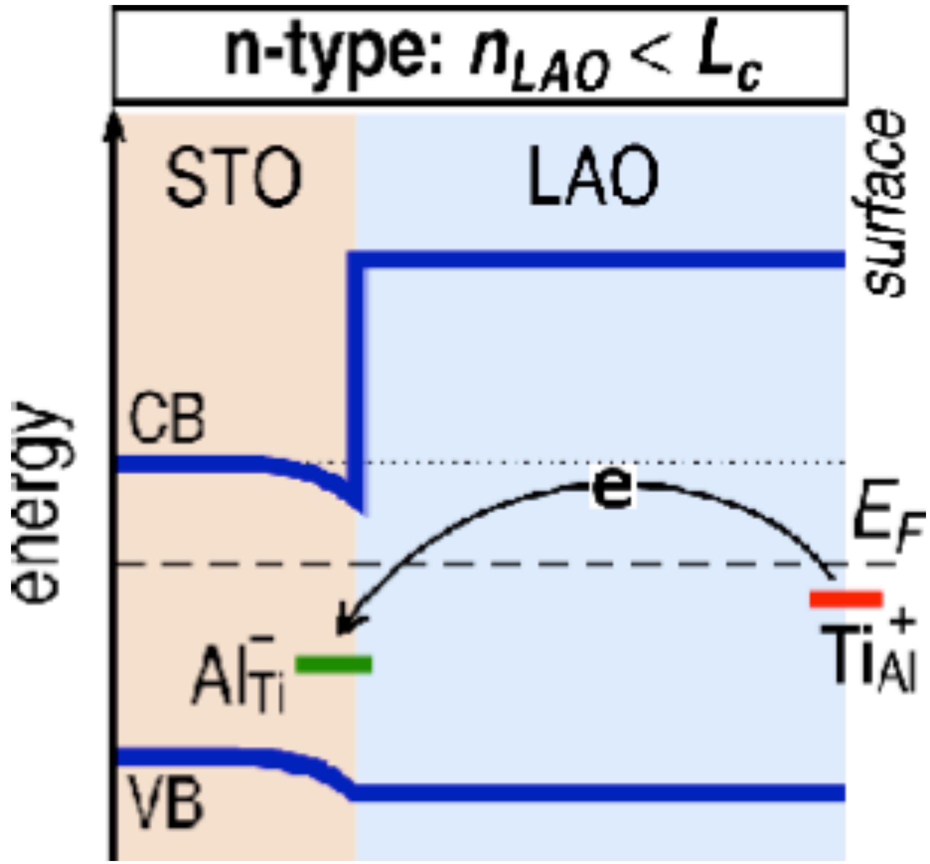
= 0.5 e/uc

The true questions: What traps electrons?

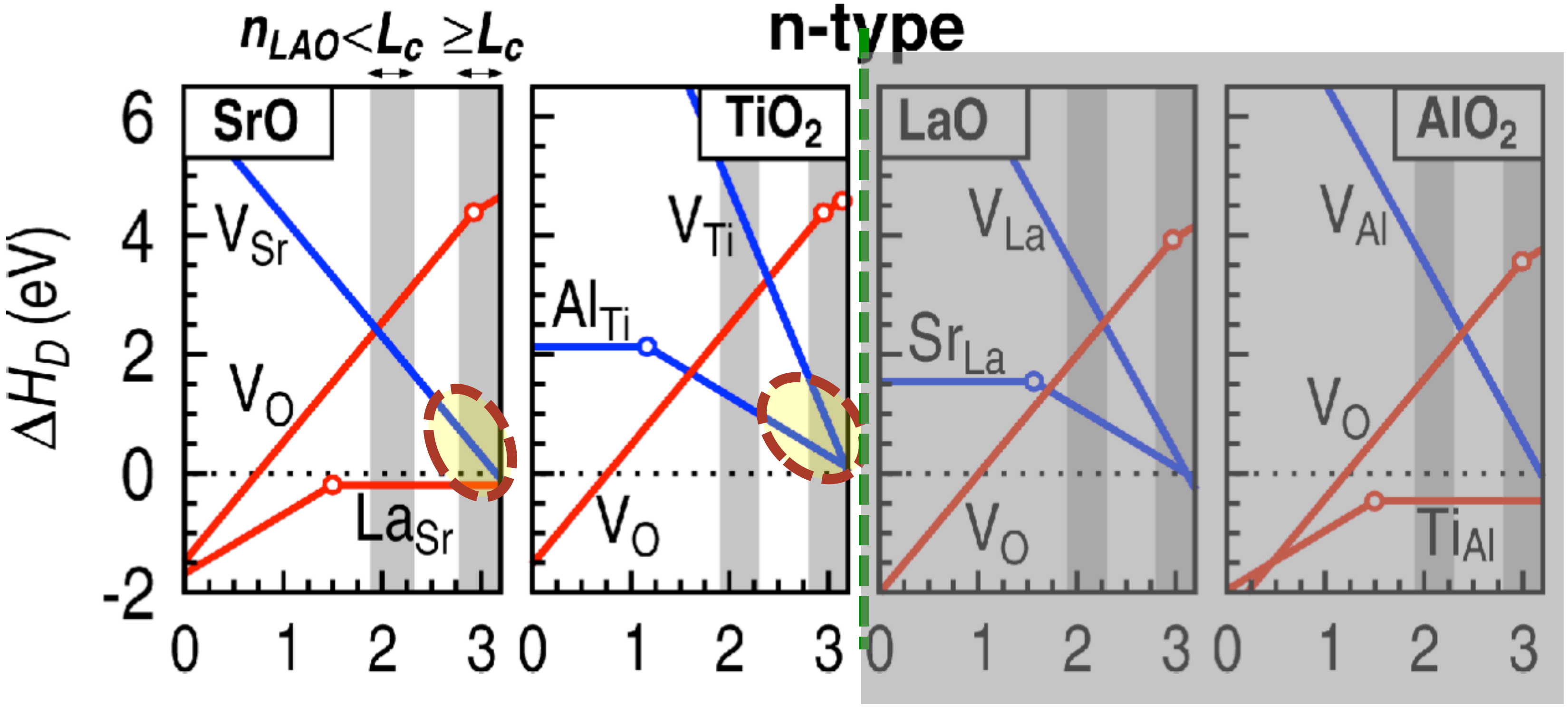
Polar catastrophe model predicts **ZERO** interface charge below L_c and **< 0.5 e/uc** at finite $n_{LAO} > L_c$



Below the L_c, Al-on-Ti antisite defects trap all 0.5e/uc interface electrons



Above the L_c, **part** of 2DEG is trapped by Al-on-Ti defects



Looking only at STO side where 2DEG locates:

- Al_{Ti} defects dominates over V_{Ti} defects
- La_{Sr} dominate over V_{Sr} defect in LaO layer

Puzzle 5

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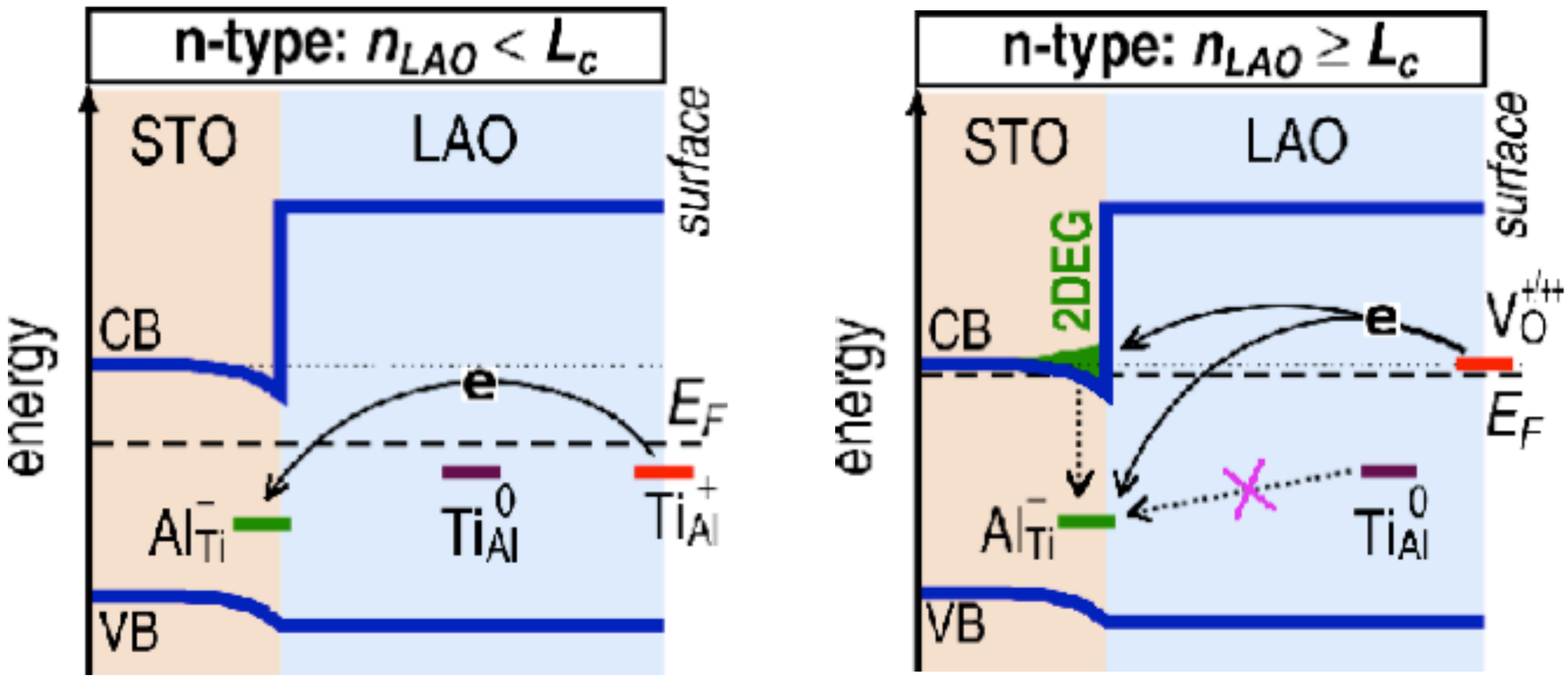
Un-ionized Ti_{Al}^{0} defects cause Ti^{3+} signals and local magnetic moment



Ti_{Al}^{+} = Ti^{4+}-on-Al^{3+}
zero local moment

Ti_{Al}^{0} = Ti^{3+}-on-Al^{3+}
Finite Local Moment

- **Magnetism has a similar L_c and resides in LAO side near interface**

Puzzle 6

① Where does 2DEG come from?

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③ What compensate electric field?

④ What controls 2DEG density?

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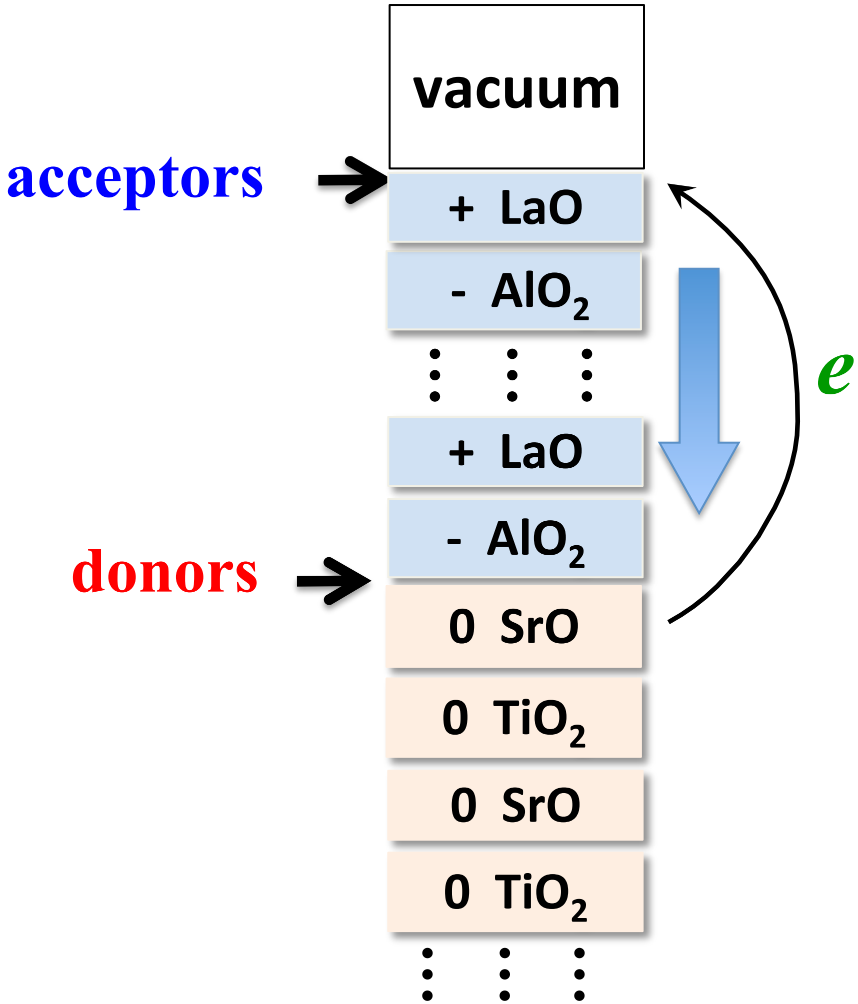
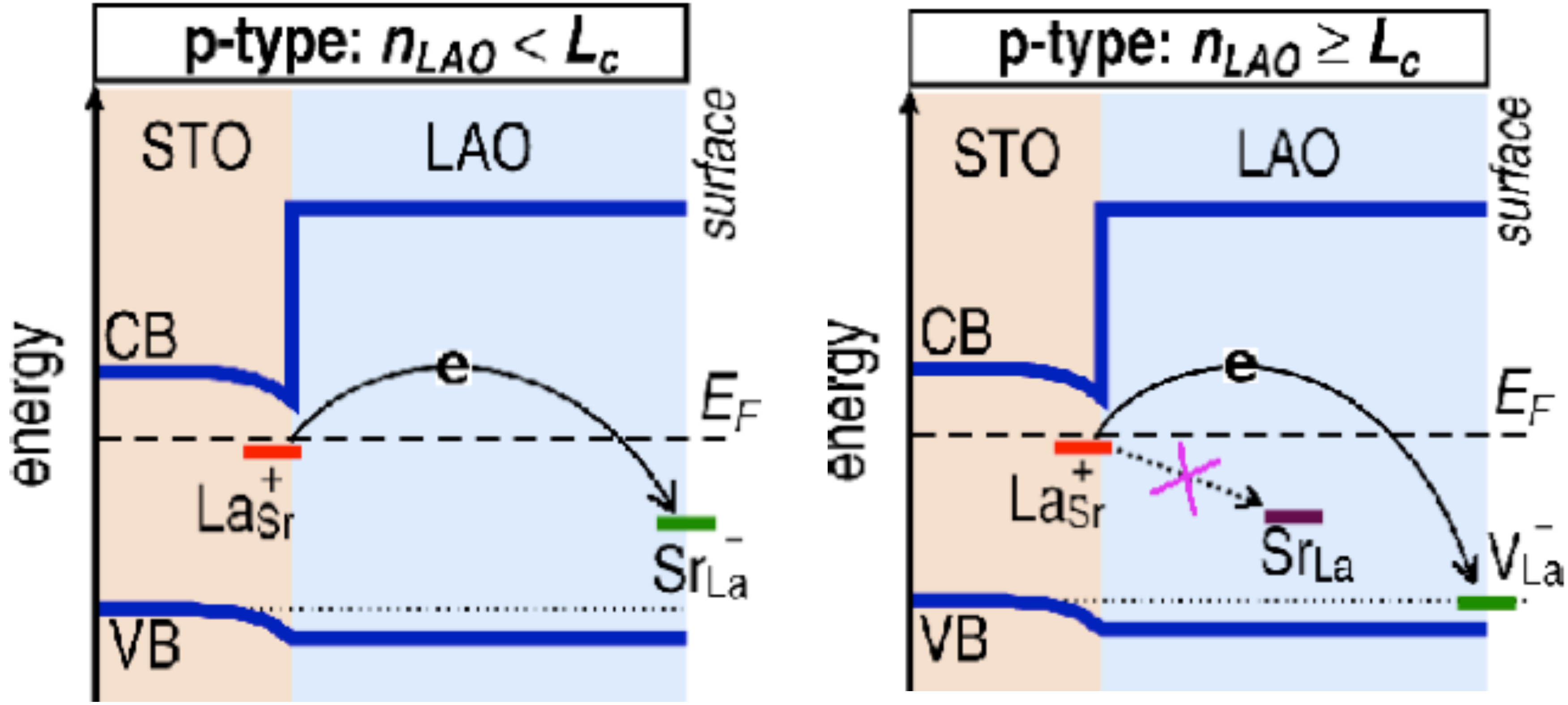
⑥ Why are p-interfaces insulating and magnetic?

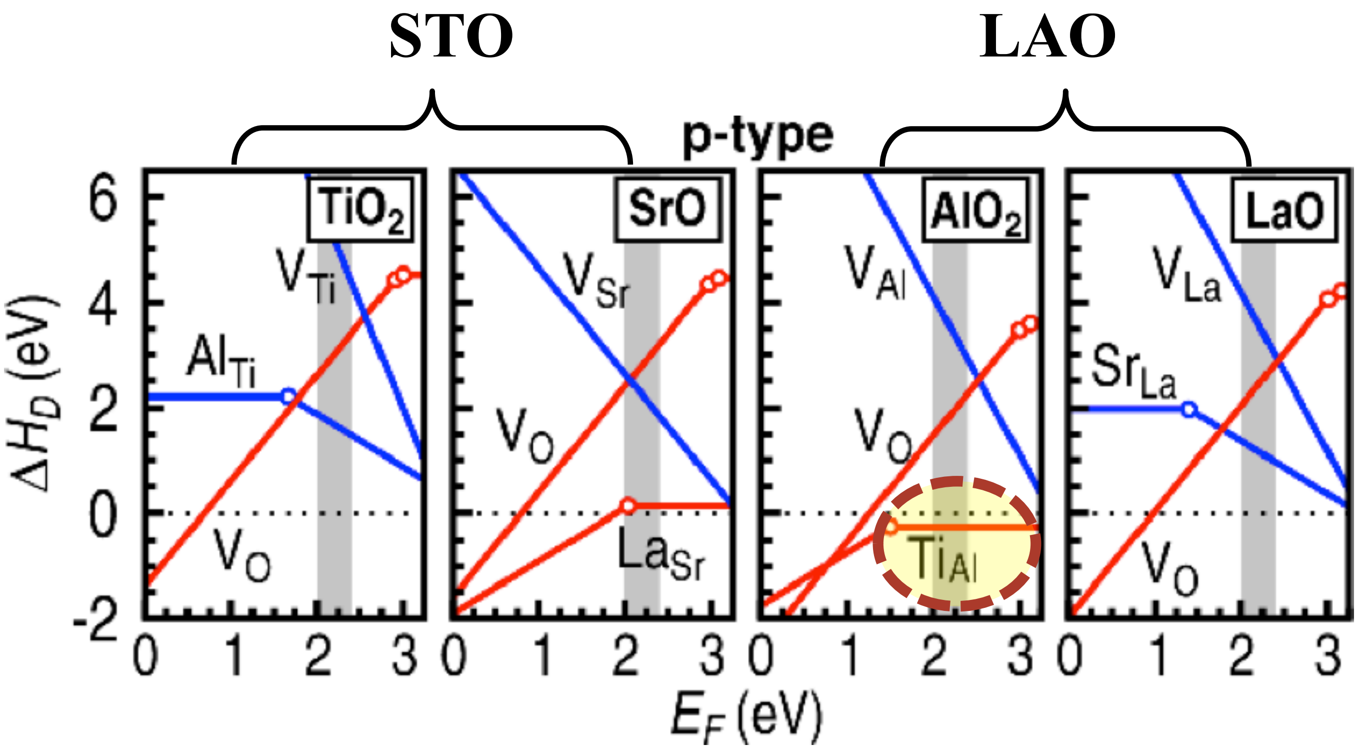
What defects should be considered?



Defects again!



Ti_{Al} defects in LAO cause interface magnetism



- Donor antisites: Low formation energy, but too deep
- Acceptor defects: too high formation energy
- Donor levers are higher than acceptor levels

The leading puzzles **SOLVED** !

- ✓ ① Where does 2DEG come from?
- ✓ ② What controls critical thickness?
- ✓ ③ What compensate electric field?
- ✓ ④ What controls 2DEG density?
- ✓ ⑤ What causes interface magnetism?
- ✓ ⑥ Why are p-interfaces insulating and magnetic?

Predictions for validation by experiments

n-type interface:

- V_O and Ti_{Al} at LAO surface below and above the L_c
- Distinction of Ti^{3+}, Ti^{4+} signals in STO side and LAO side (as defect)

P-type interfaces:

- V_{La} and Sr_{La} at LAO surface below and above the L_c

arXiv:1402.0895