Mott Quantum Wells

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Motivation for Complex Oxide Heteroepitaxy

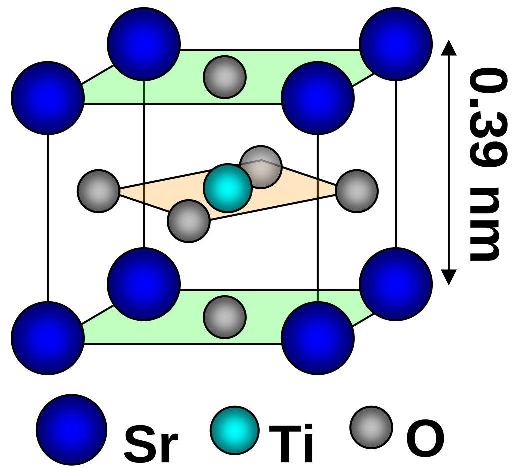
Unconventional superconductivity

Charge,orbital, spin ordering

Colossal magnetoresistance

Multiferroics

Perovskite structure



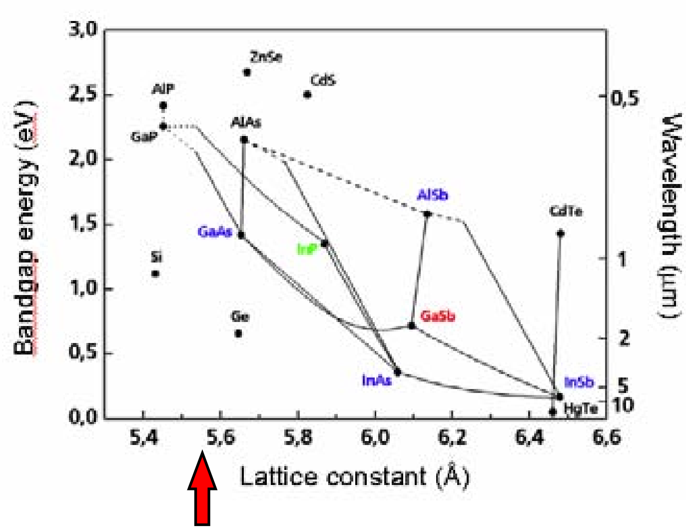
Integrated electro-optics, ferroelectrics, dielectrics

Josephson junctions

Magnetic tunnel junctions

Atomic scale synthesis

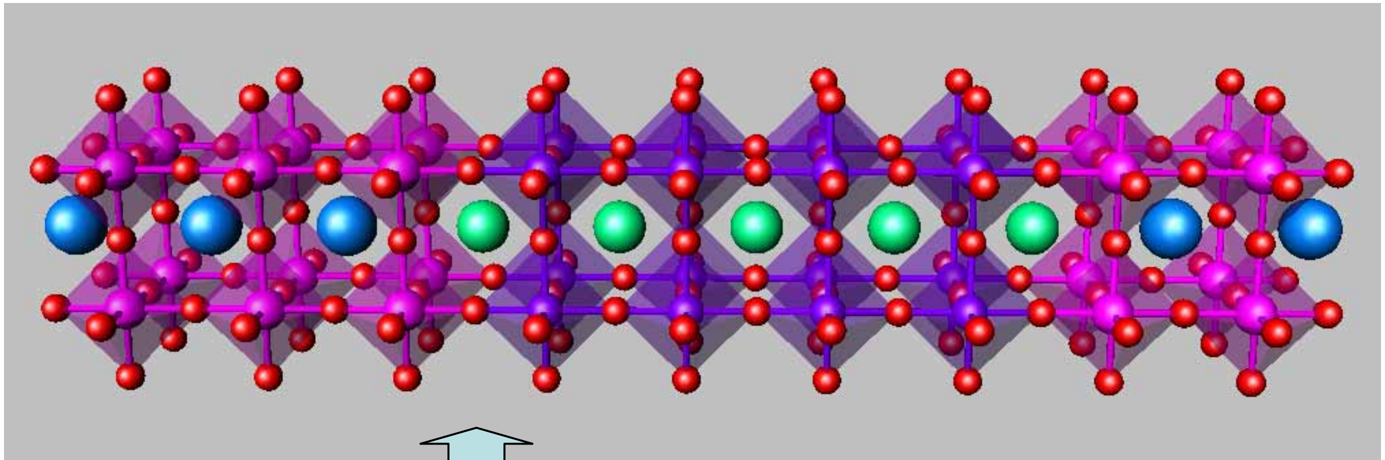
Resistive switching



$\sqrt{2}a_o$ for perovskite-based oxides

Almost every ground state is available for heteroepitaxy!

Fundamental drive to increase the degrees of freedom in semiconductor heterostructures (e.g. "spintronics").



What is happening here?

"The interface is the device" Herbert Kroemer

Focus here on strong correlations

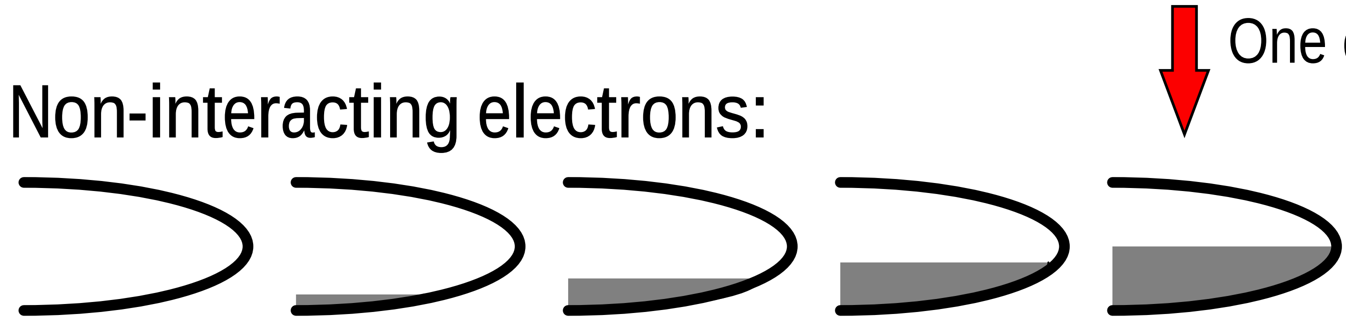
Outline:

1) $LaTiO_3/SrTiO_3$ superlattices – role of lattice screening
2) $LaTiO_3/LaAlO_3$ superlattices – quantum confinement
3) $LaAlO_3/SrTiO_3$ – aside on polar discontinuities and their couplings
4) Modulation doping of $LaVO_3$ quantum wells via a polar surface

Filling Electrons into a Band Insulator

One electron per site

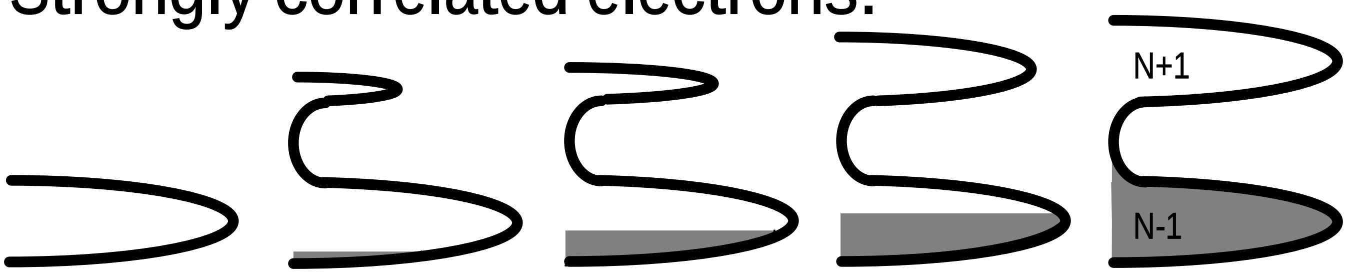
Non-interacting electrons:



Band metal

Strongly correlated electrons:

Upper Hubbard band



Mott insulator

Lower Hubbard band

$$H = -t \sum_{<i,j>} (c^{+}_{i,\sigma} c_{j,\sigma} + h.c.) + U \sum_{i} n_{i\uparrow} n_{i\downarrow}$$

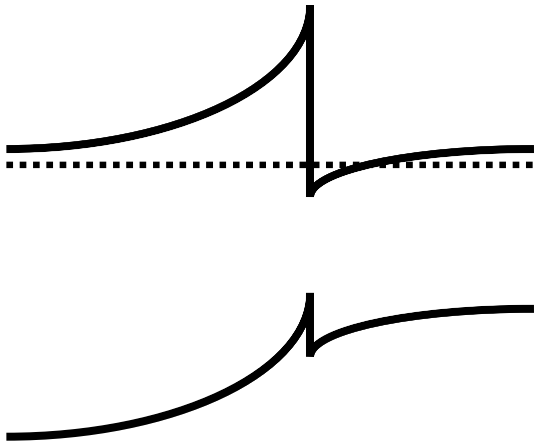
Antiferromagnetic ground state

$$H = J \sum_{<i,j>} (\vec{S}_i \bullet \vec{S}_j - \frac{1}{4}); \quad J = \frac{4t^2}{U}$$

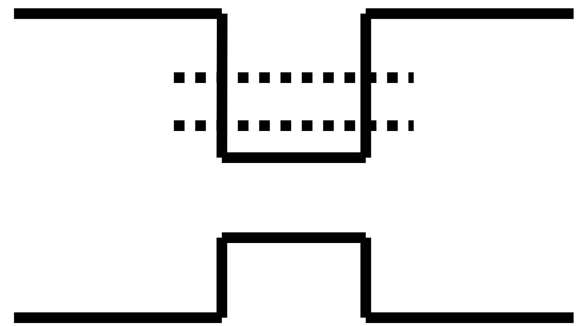
The electronic structure is a function of the number of electrons - not a rigid single particle band!

"Mott Semiconductor" at Interfaces and in Confined Geometries

Semiconductor heterojunction



Quantum well



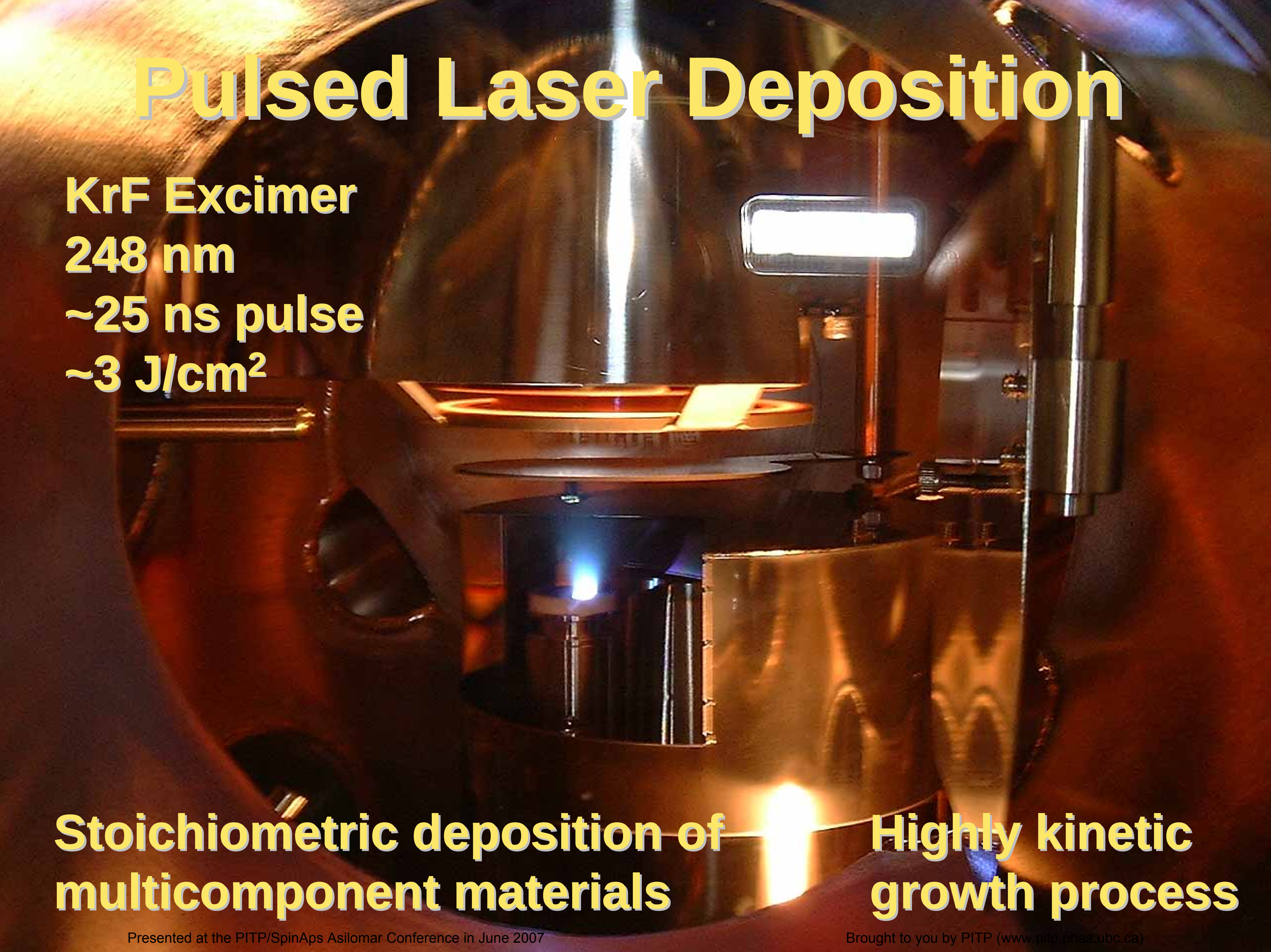
These "band diagrams", so central to semiconductor devices, assume rigid, single-particle bands.

Questions:

- *What is the correlated equivalent of band bending and quantum confinement?*
- *Can we create new charge states at these interfaces?*

Pulsed Laser Deposition

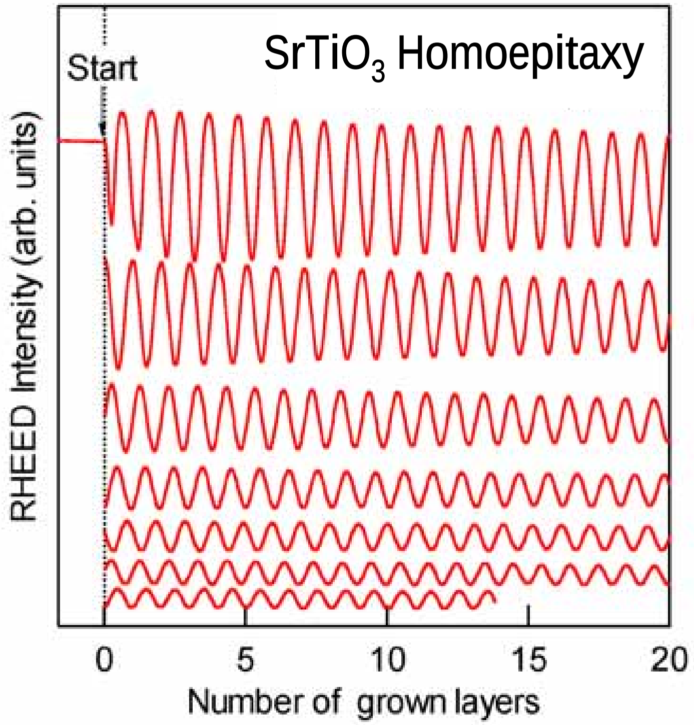
KrF Excimer
248 nm
~25 ns pulse
~3 J/cm^2



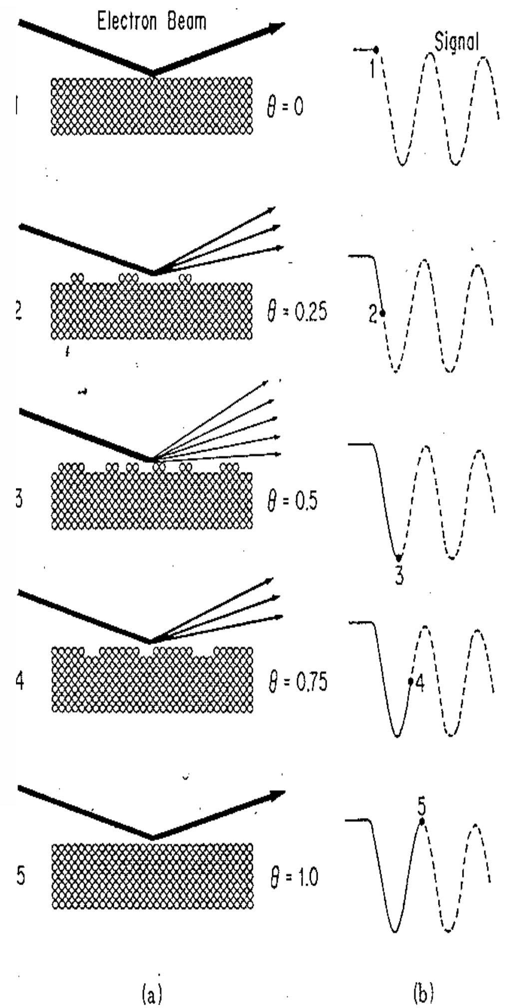
Stoichiometric deposition of multicomponent materials

Highly kinetic growth process

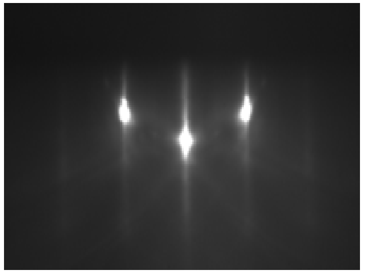
In-situ RHEED Monitoring



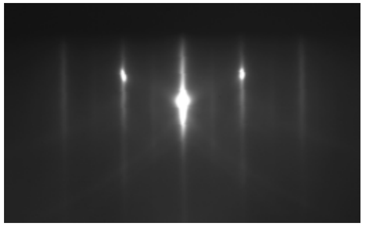
Unit cell RHEED oscillations
1 unit cell for ~ 25 laser pulses



substrate



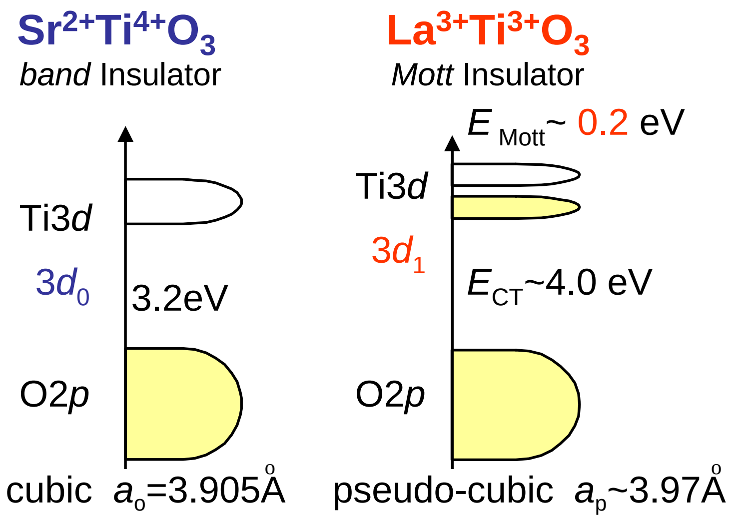
film

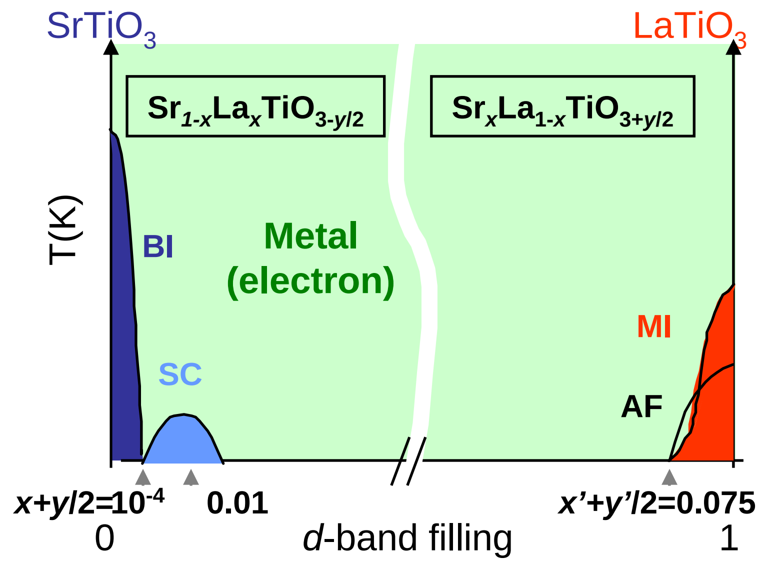


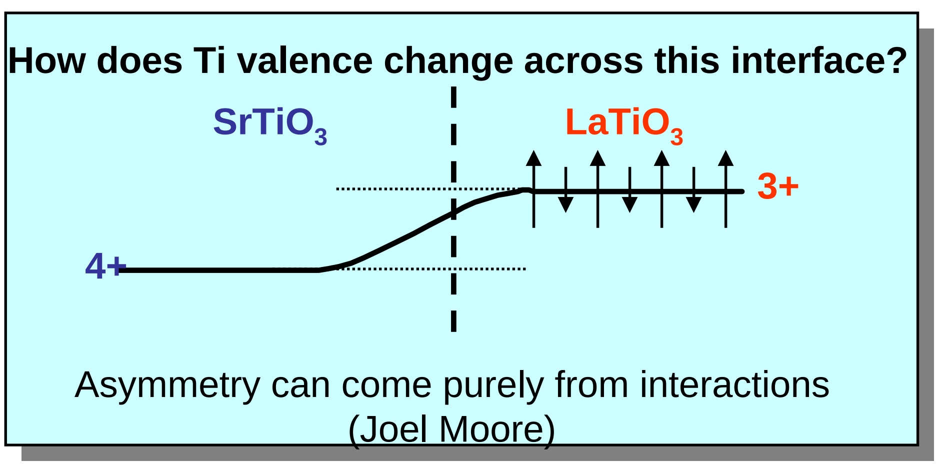
[100]

<u>R</u>eflection <u>H</u>igh <u>E</u>nergy <u>E</u>lectron <u>D</u>iffraction

1. $SrTiO_3/LaTiO_3$ Interfaces/Superlattices



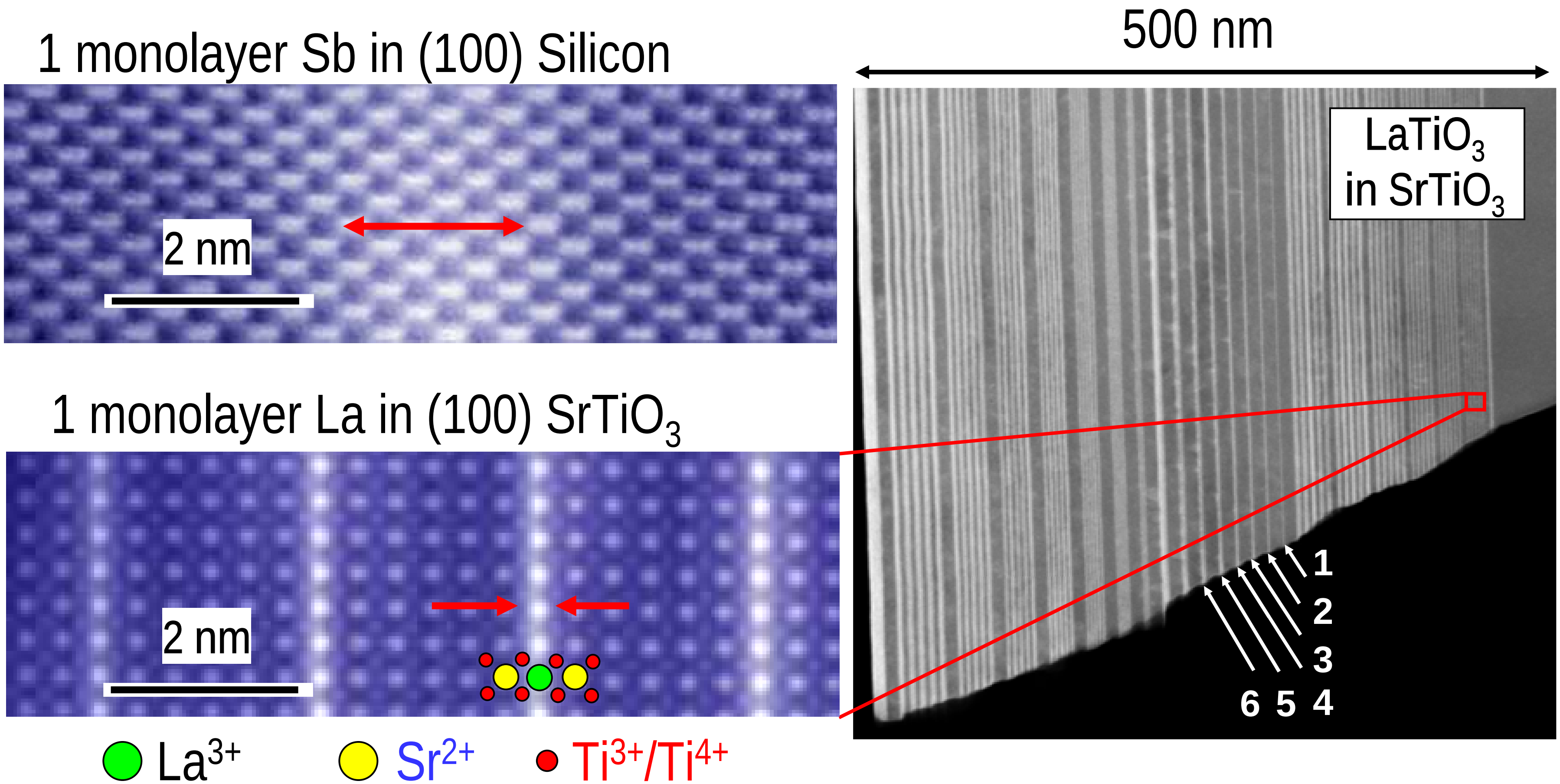




- The electronic compressibility and $\varepsilon_{eff}(x)$ sets a new length
- The mixed valence interface is a confined conductor
- TiO_6 octahedral network continuous, only changing A-site

Central issue is the control of the Ti oxidation state.

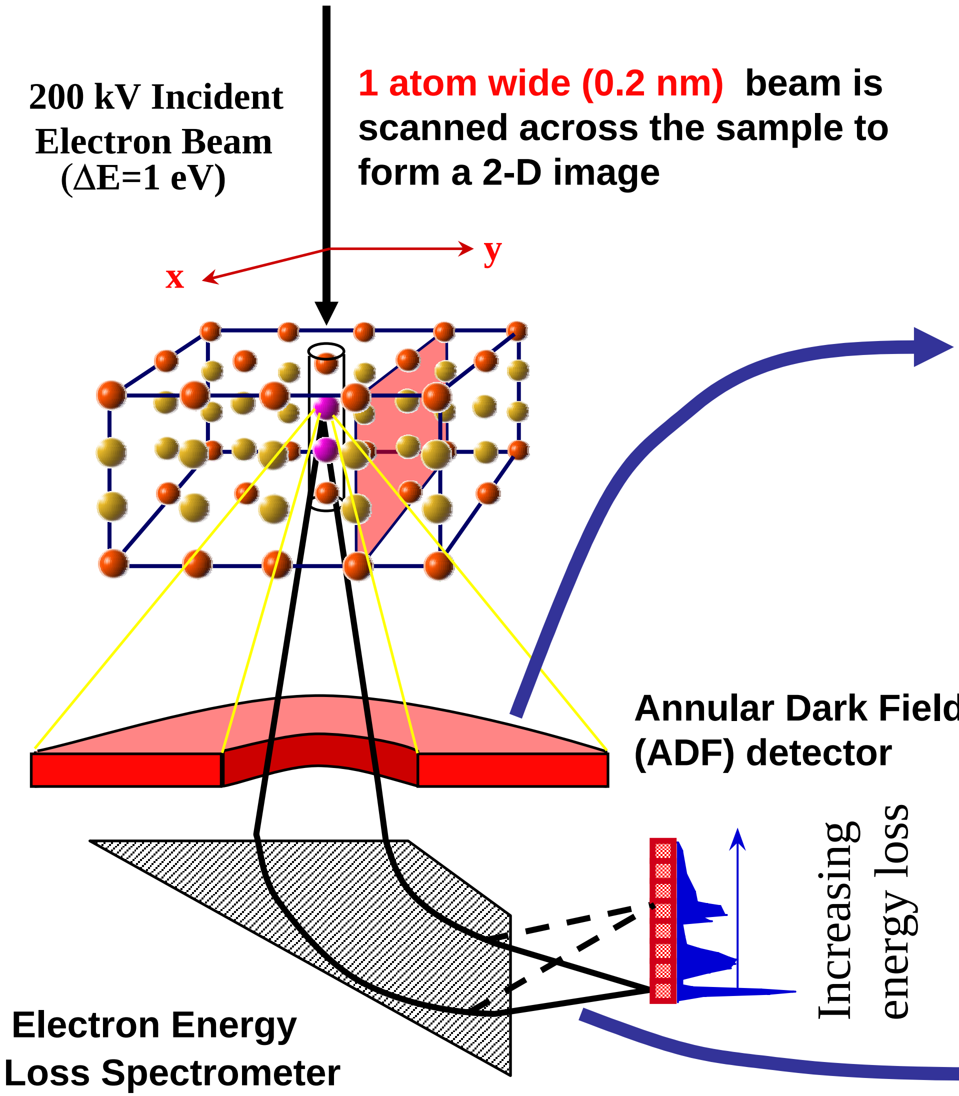
Atomic Scale Delta-Doping: $LaTiO_3/SrTiO_3$

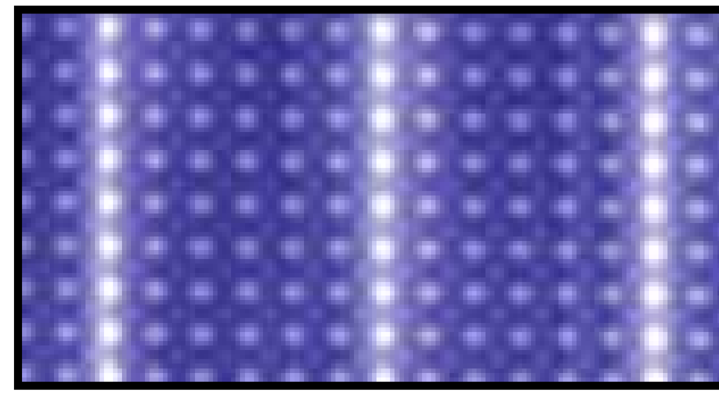


Superlattices by design

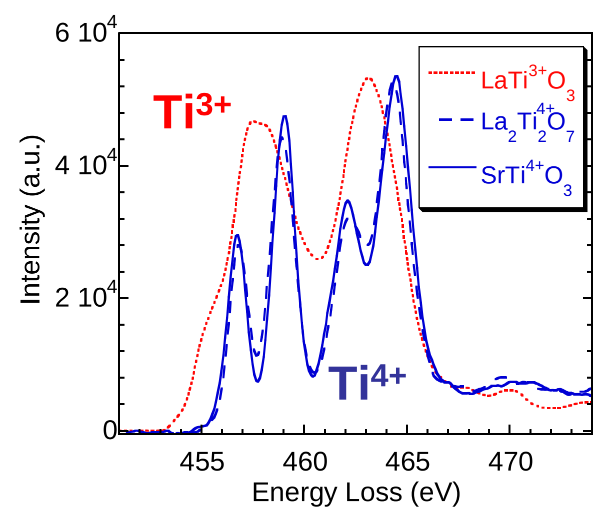
A. Ohtomo, D. A. Muller, J. A. Grazul, and H. Y. Hwang, *Nature* 419, 378 (2002).

Scanning Transmission Electron Microscopy





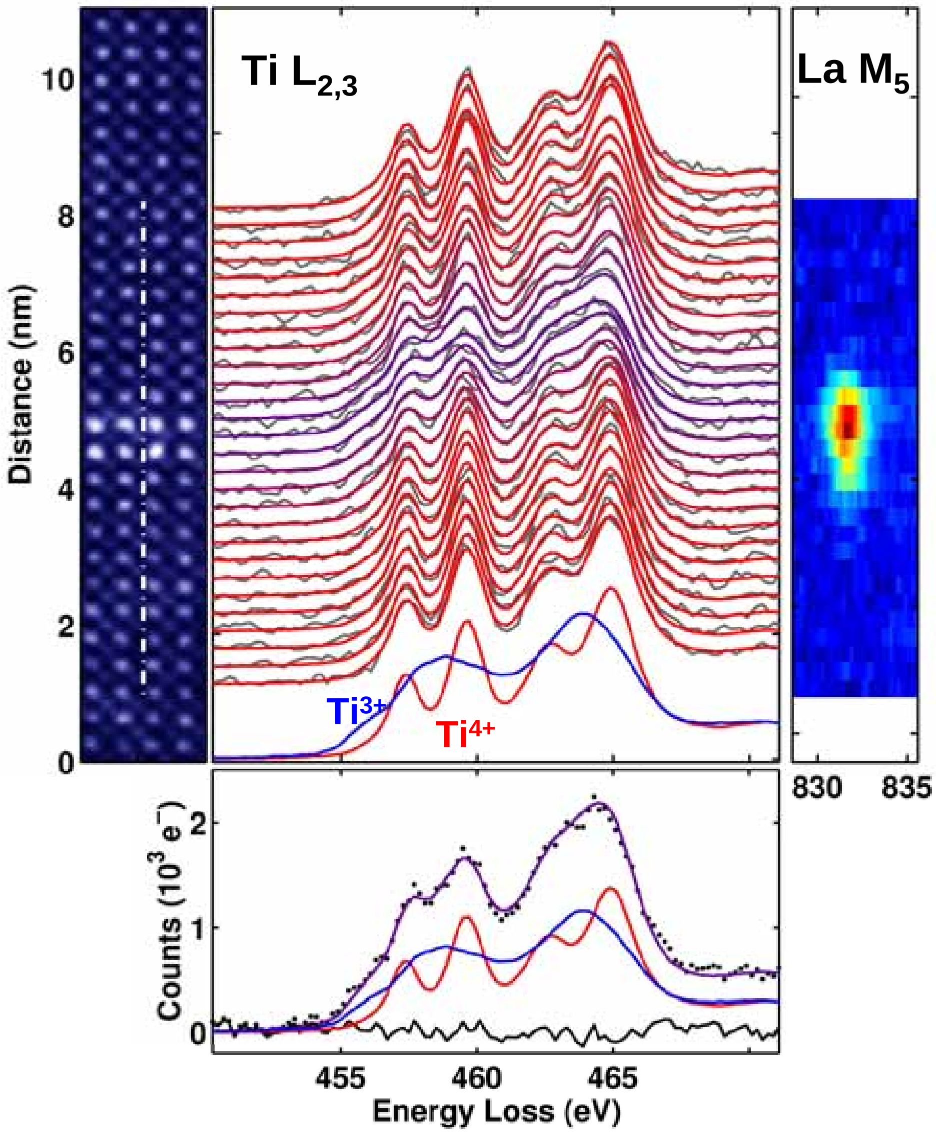
Elastic Scattering ~ "Z contrast"



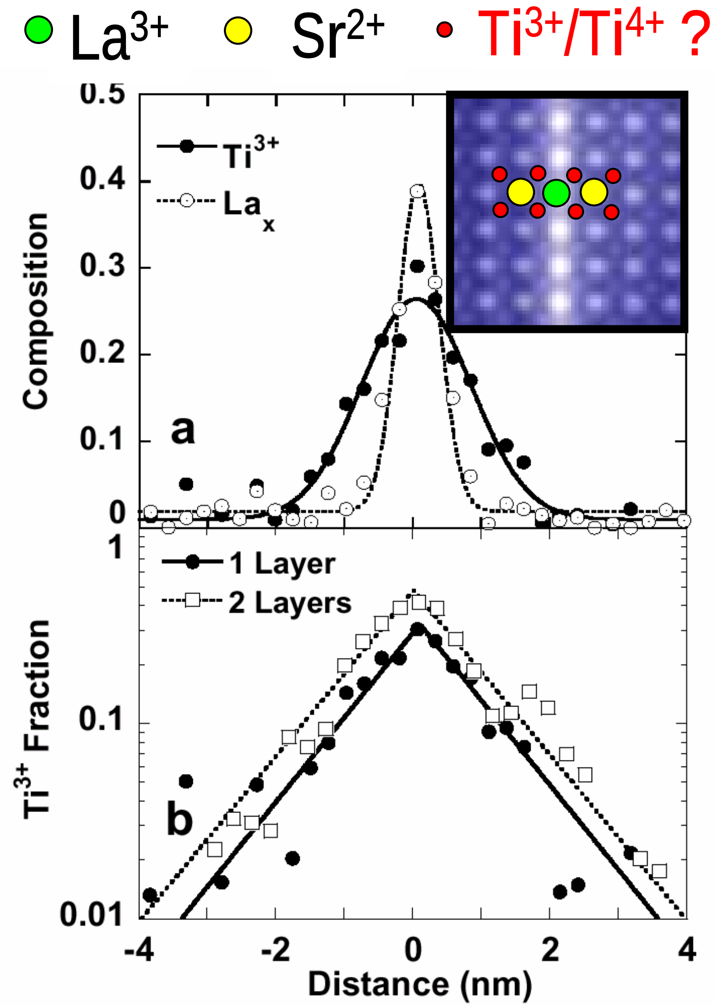
Ti $L_{2,3}$ Edge from $LaTiO_3$ and $SrTiO_3$

A. Ohtomo, D. A. Muller, J. A. Grazul, and H. Y. Hwang, *Appl. Phys. Lett.* 80, 3922 (2002).

Electron Distribution on an Atomic Length Scale



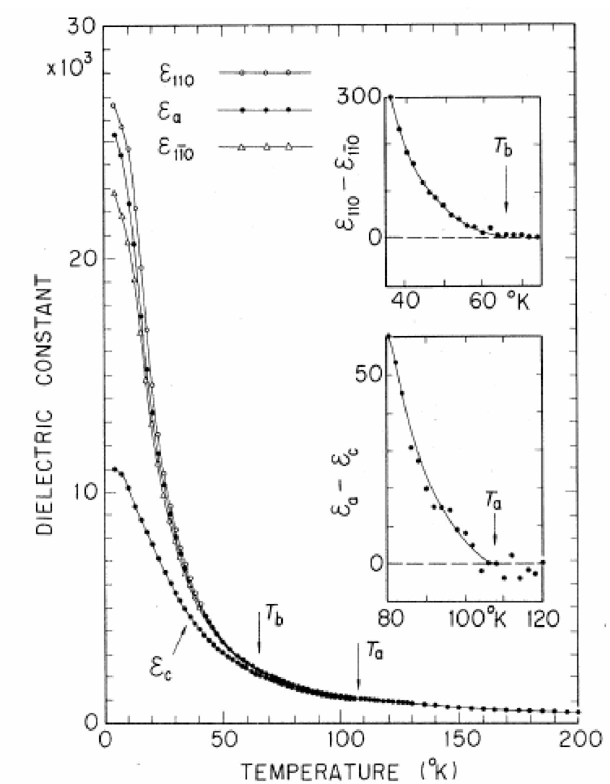
Spectral decomposition of the Ti L edge.



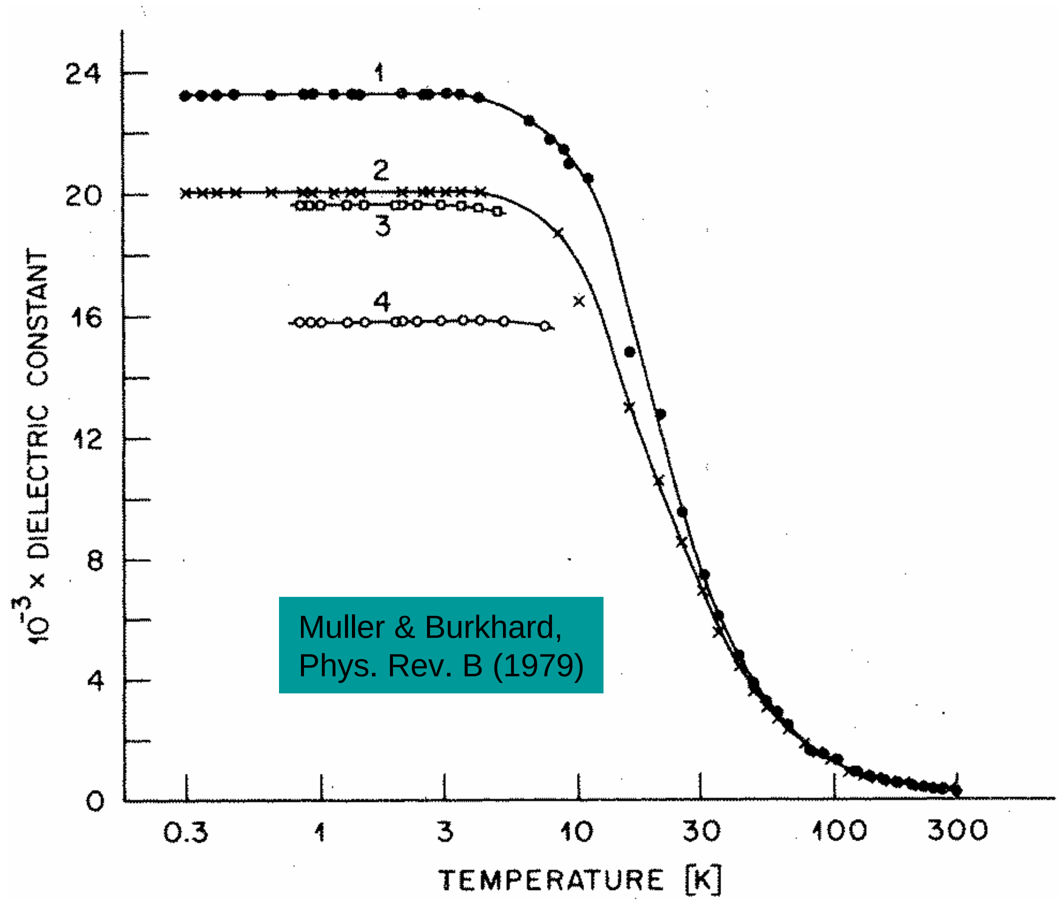
Screening length: $\lambda = 1.0 \pm 0.2$ nm

$\lambda_{TF\text{-}free} = 0.072$ nm

SrTiO$_3$ Dielectric Properties

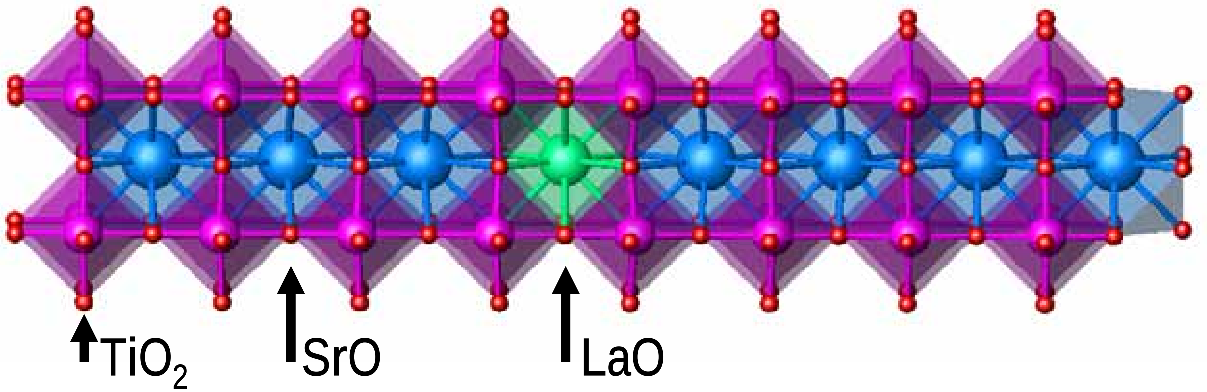


T. Sakudo and H. Unoki,
Phys. Rev. Lett. 26, 851 (1971).

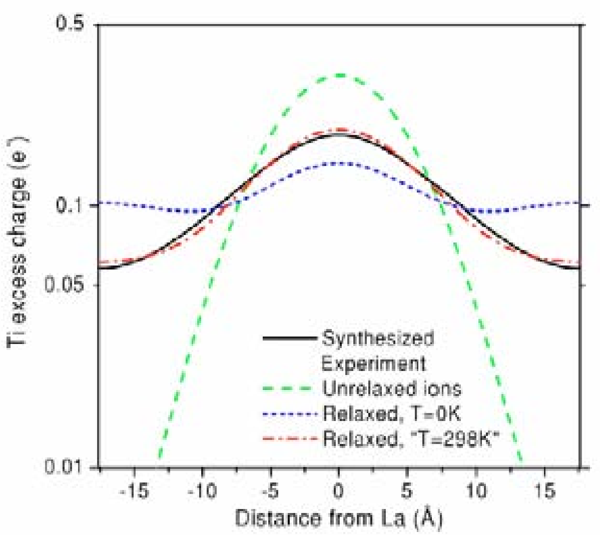


"Quantum Paraelectric"

Density Functional Studies of Lattice Polarization

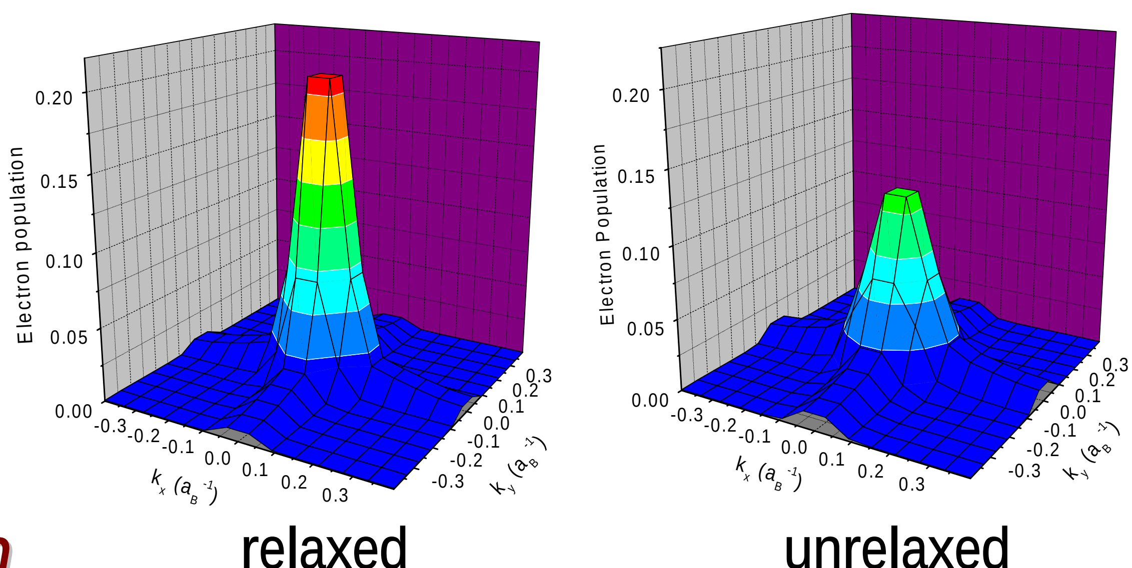


Electronic structure calculations with and without lattice relaxation.



real space distribution

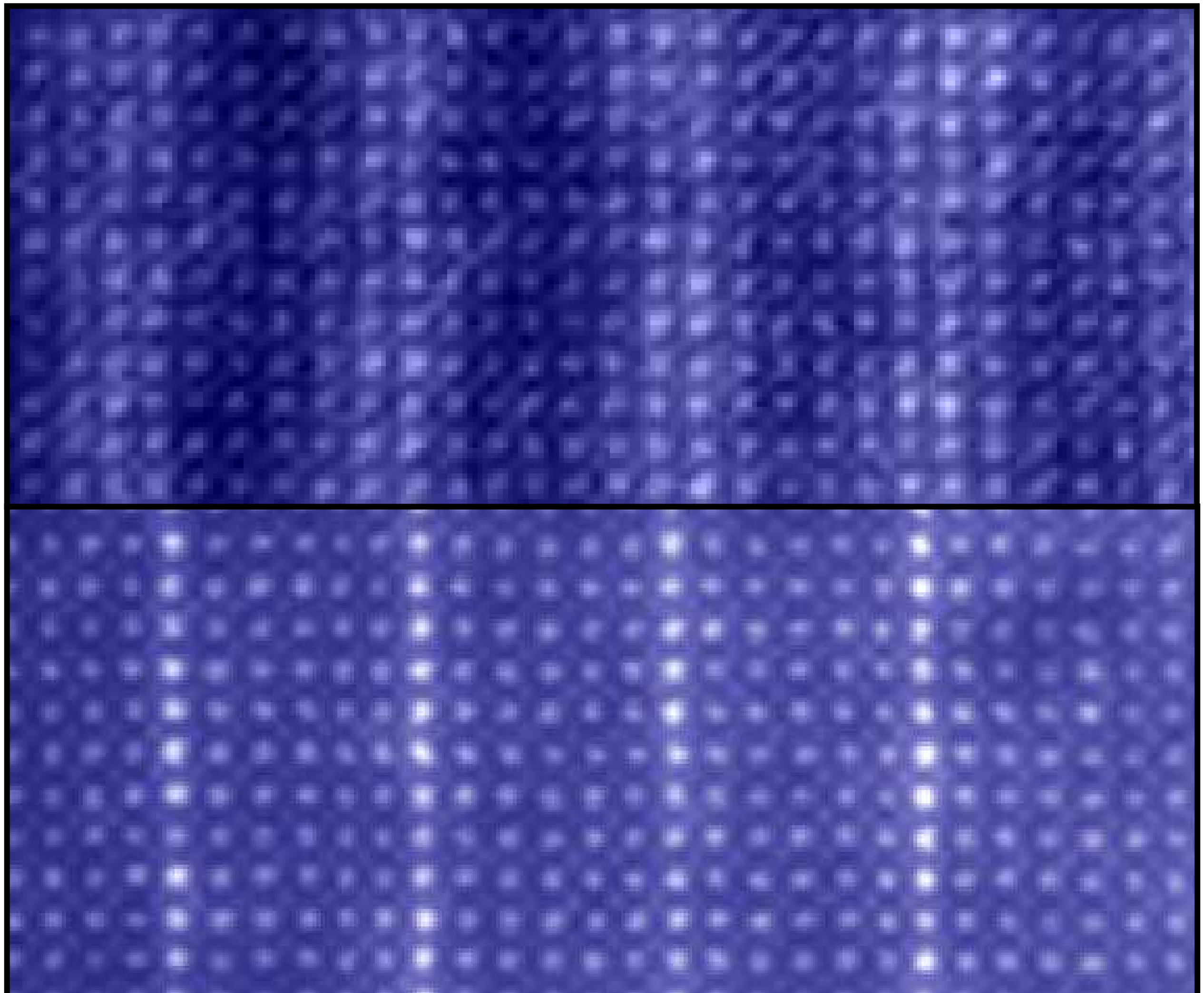
k-space distribution



D. R. Hamann, D. A. Muller, and H. Y. Hwang, *Phys. Rev. B* 73, 195403 (2006).
S. Okamoto, A. J. Millis, and N. A. Spaldin, *Phys. Rev. Lett.* 97, 056802 (2006).

Diffusion of Cations With Oxygen Defects

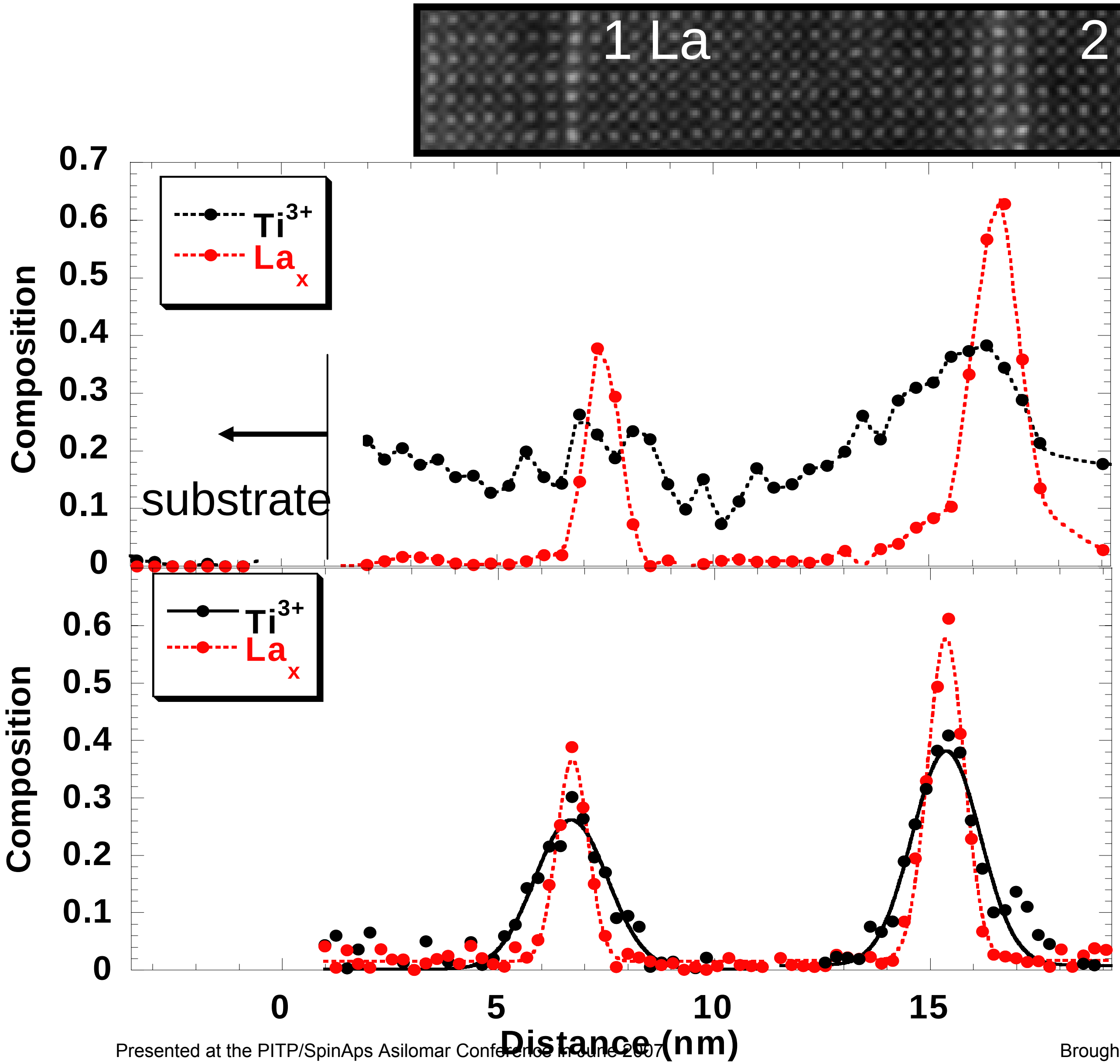
$(LaTiO_3)_1(SrTiO_3)_5$



Superlattices grown at 10^{-6} Torr of O_2. The high vacancy concentration has led to La diffusion. The "fuzzy" quality of the lattice image is indicative of La/Sr disorder.

Superlattices grown at 10^{-5} Torr of O_2. With reduced oxygen vacancies, the structure remains abrupt and well ordered.

Oxygen Vacancies/La Modulate the Ti Valence



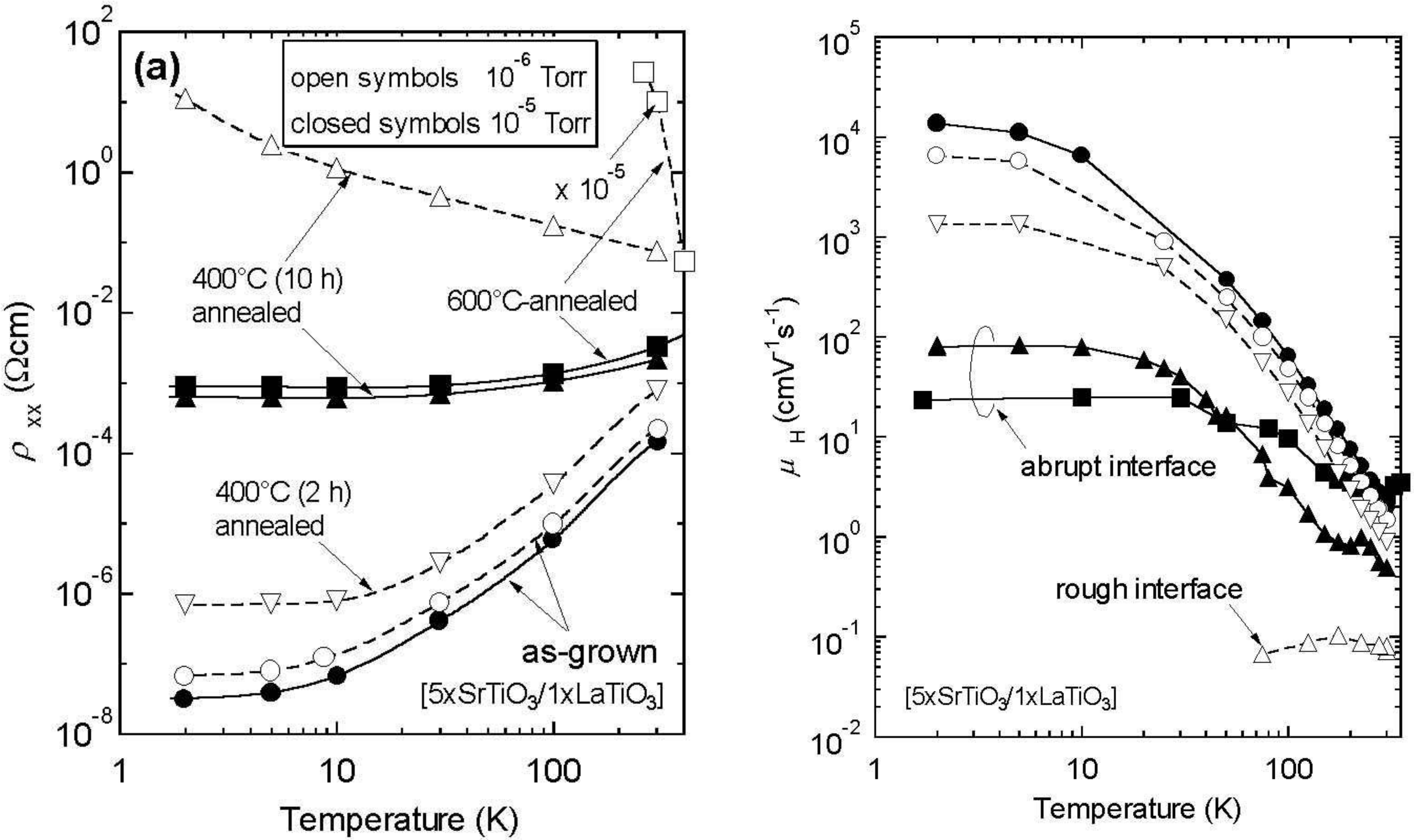
As-grown:

- **10% Oxygen deficient**
- **Weak charge modulation from La (hidden by O δ)**

Annealed in O_2:

- **removes O vacancies**
- **Strong charge modulation from La now visible**

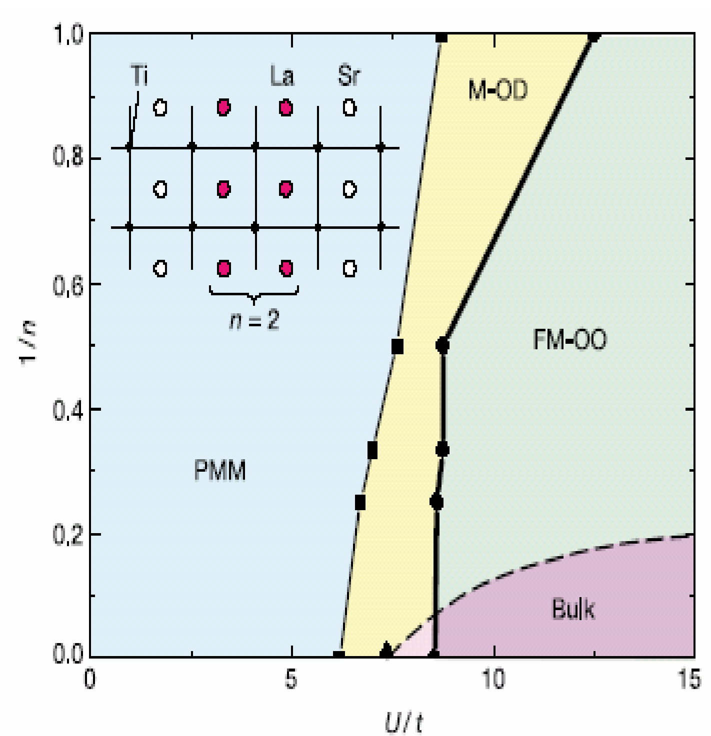
Fuzzy versus Sharp Delta-Layers



Mobility Edge

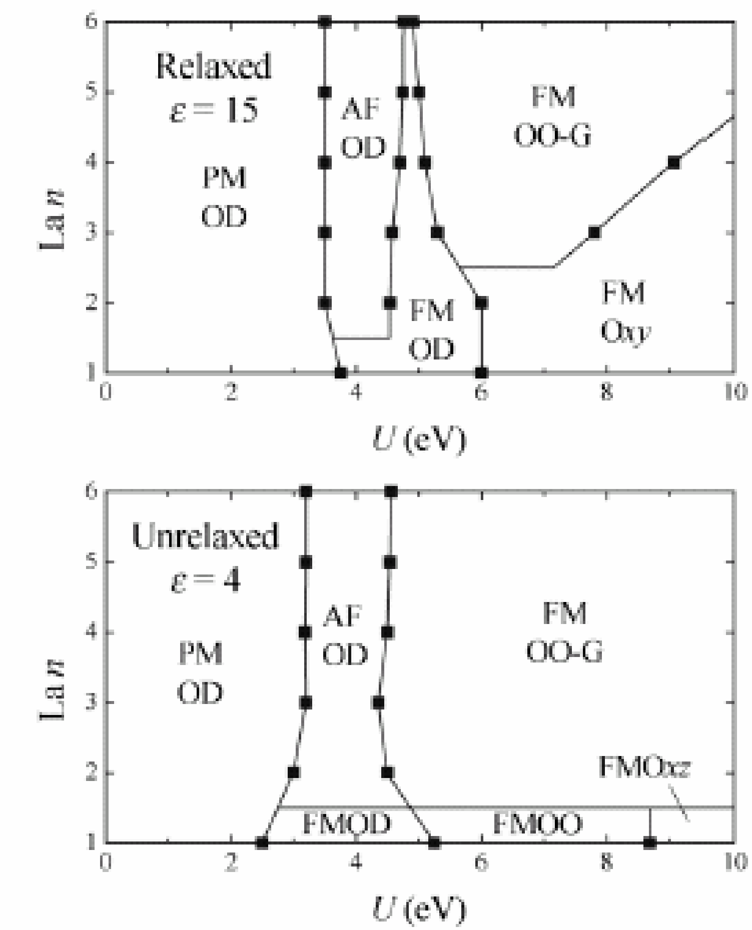
(Proximate) Magnetic Ground States?

Hartree-Fock



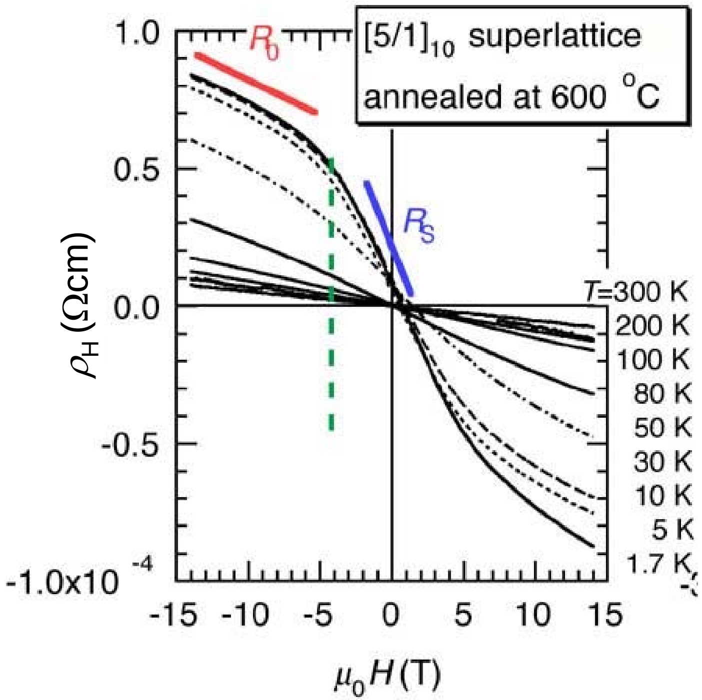
S. Okamoto and A. J. Millis, *Nature* 428, 620 (2004).

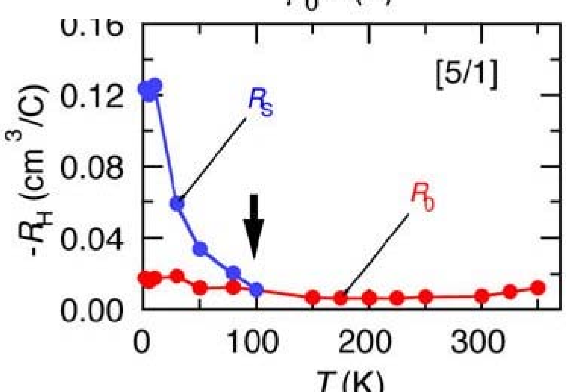
LDA-U + HF

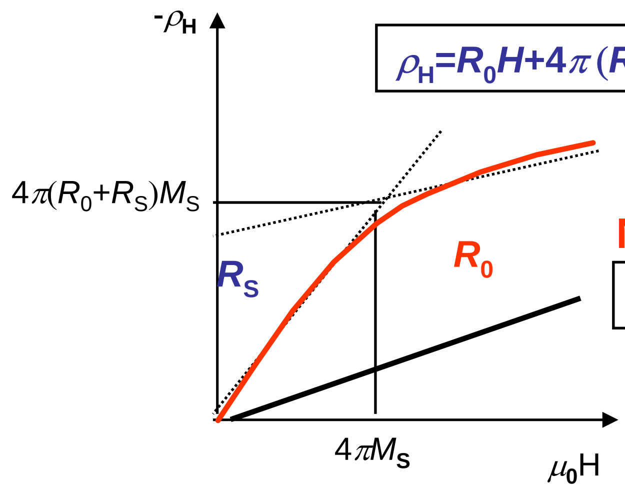


S. Okamoto, A. J. Millis, and N. A. Spaldin, *Phys. Rev. Lett.* 97, 056802 (2006).

Anomalous Hall Effect - Associated with Magnetic Correlations?







Anomalous Hall

$\rho_H=R_0H+4\pi(R_0+R_S)M_S$

Normal Hall

$\rho_H=R_0H$

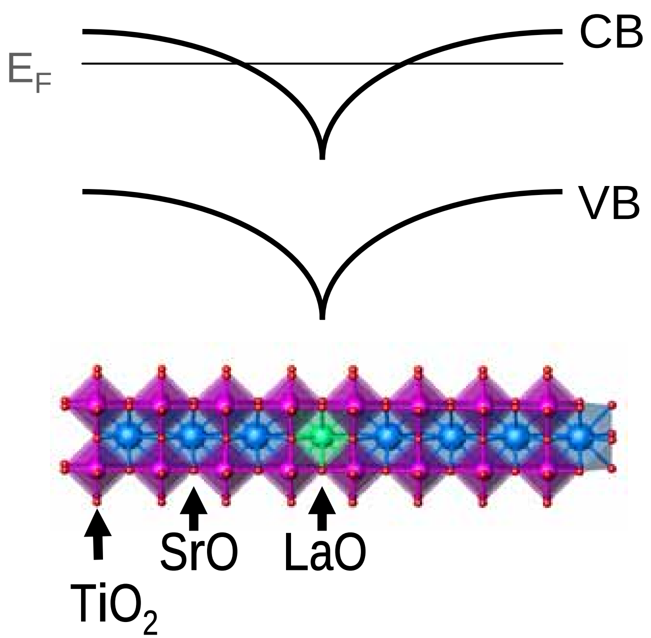
appearance of *magnetic phase* below 100 K?

cf.) Bulk $Sr_xLa_{1-x}TiO_{3+y/2}$
T_N=100 K (x+y/2=0.06)
T_N= 145 K (x+y/2=0)

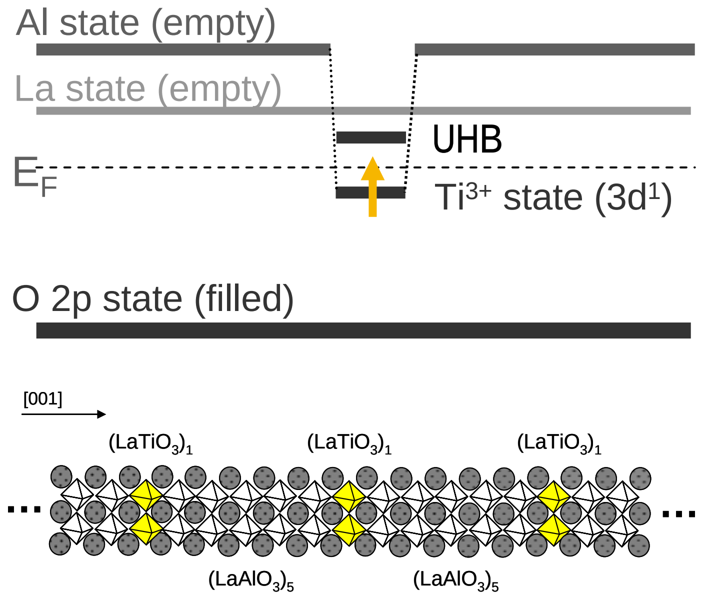
usually, $R_S \to 0$ as $T \to 0$ for FM

2. $LaTiO_3$ in $LaAlO_3$ – Confinement

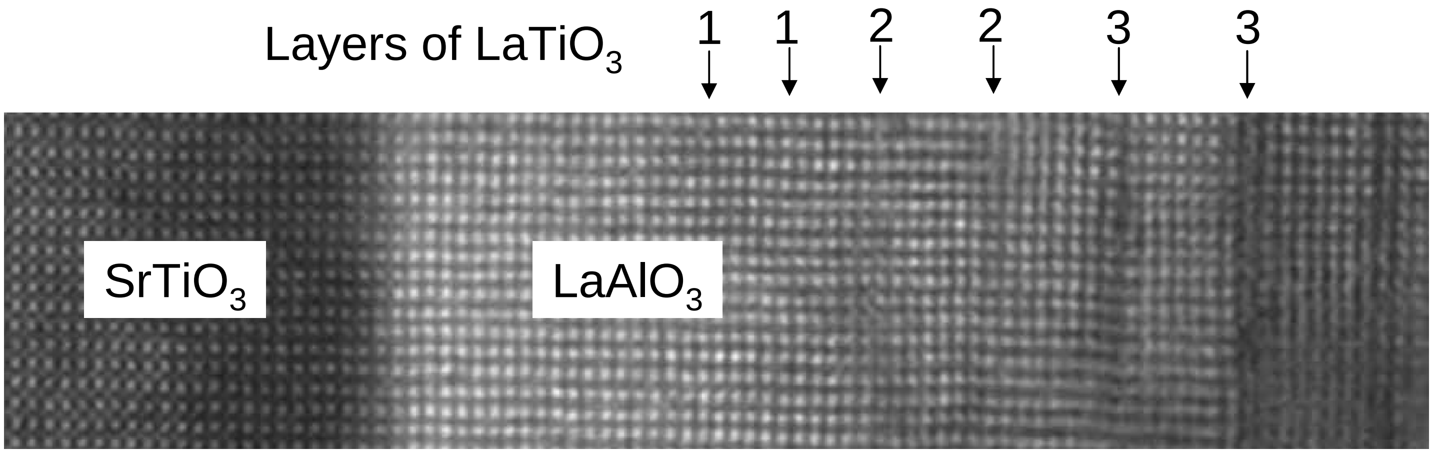
$LaTiO_3$ in $SrTiO_3$ can be considered δ-doping - interesting correlated metallic state.

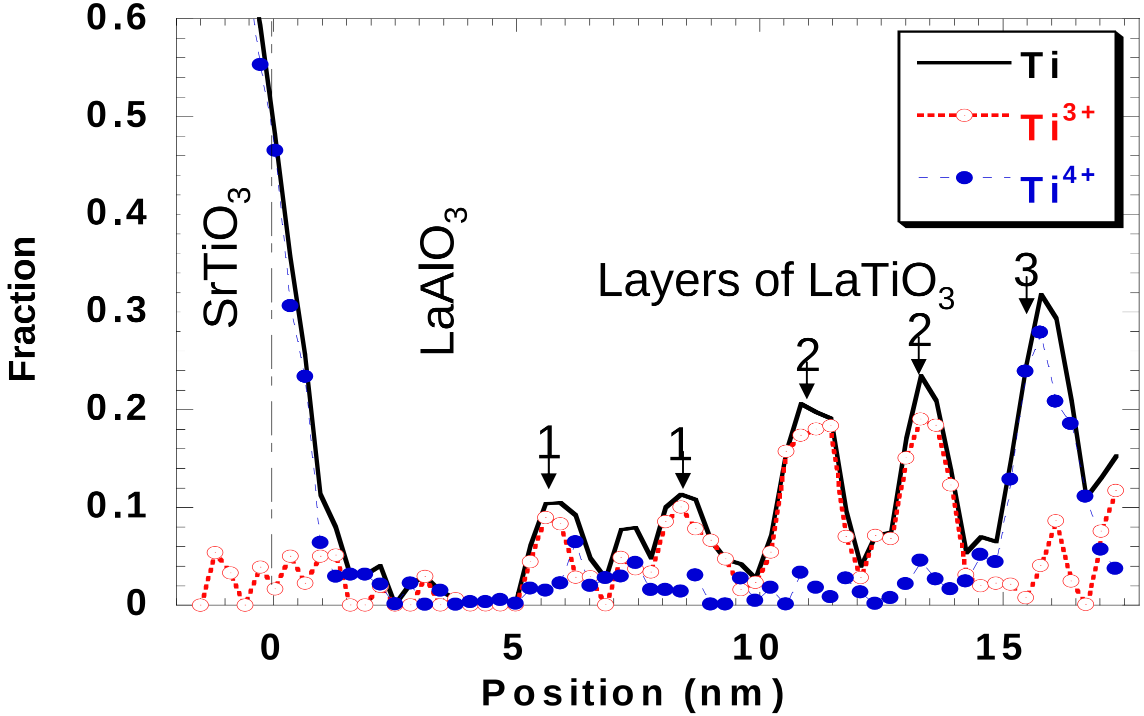


By embedding $LaTiO_3$ in wider bandgap $LaAlO_3$, can we quantum confine?



$LaTiO_3$ in $LaAlO_3$





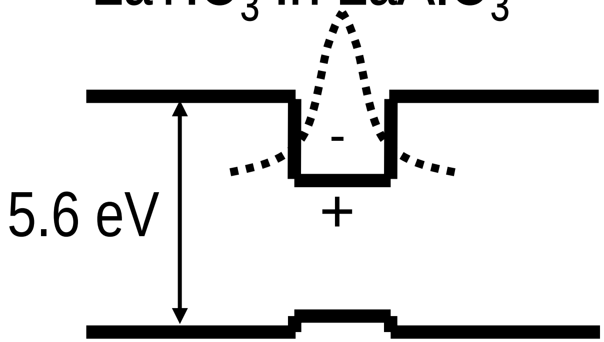
Ti L edge EELS (fitted to Ti^{3+}, Ti^{4+} spectra).

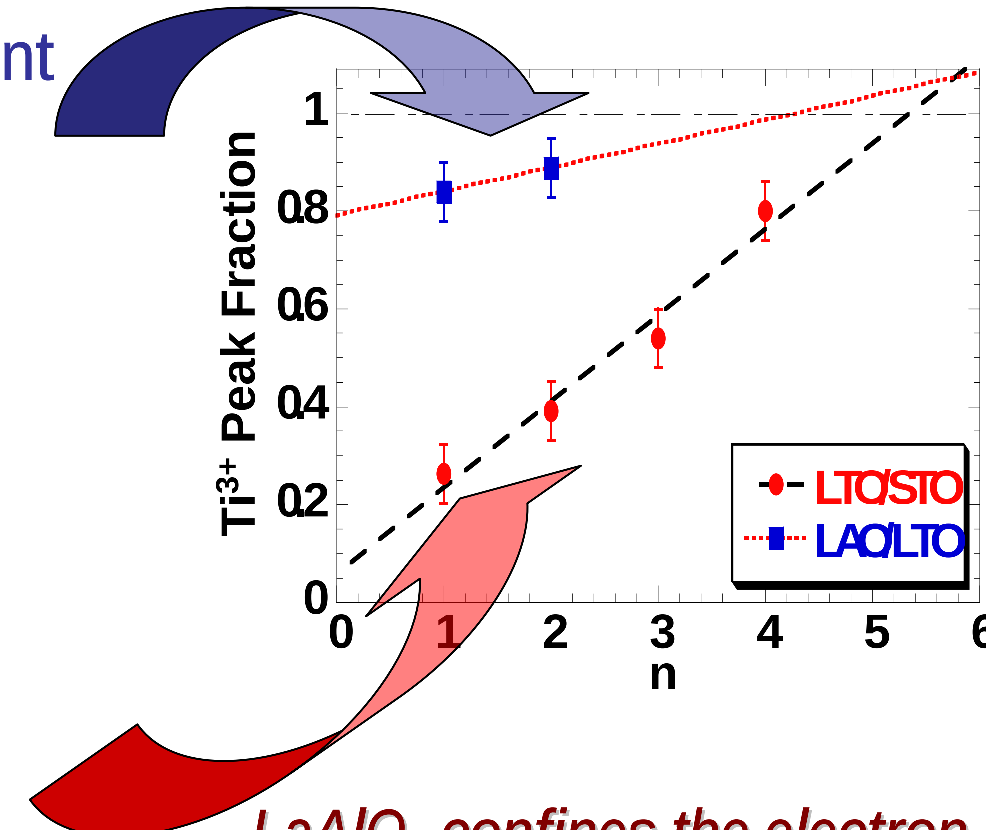
Lower resolution due to charging in $LaAlO_3$ and low density of Ti sites

Quantum Confinement of Mott Insulators

Quantum confinement

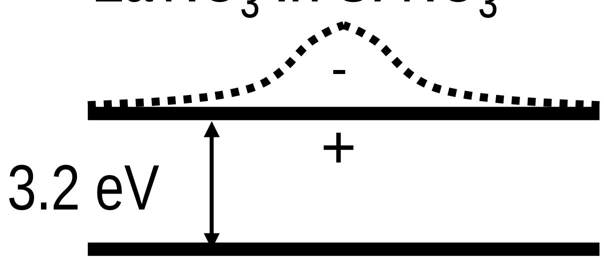
LaTiO$_3$ in LaAlO$_3$





δ-doping

LaTiO$_3$ in SrTiO$_3$

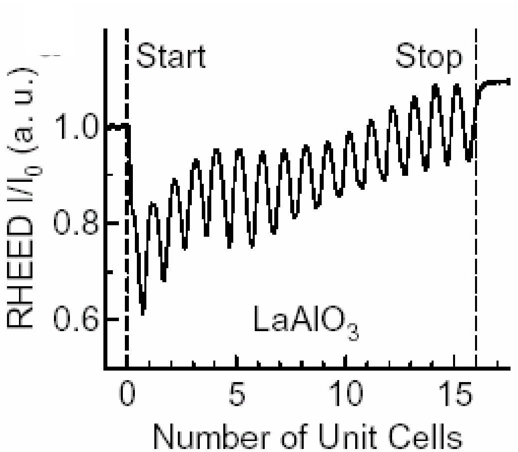


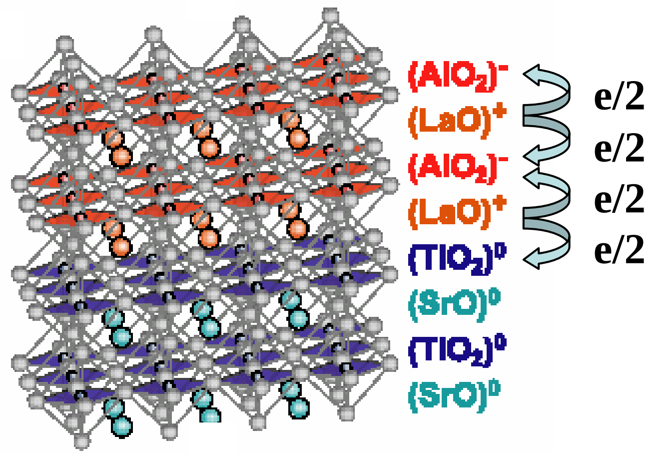
LaAlO$_3$ confines the electron to the thin LaTiO$_3$ layer (increasing the peak electron density).

Is it a Mott insulator? Insulating, gap appears robust.

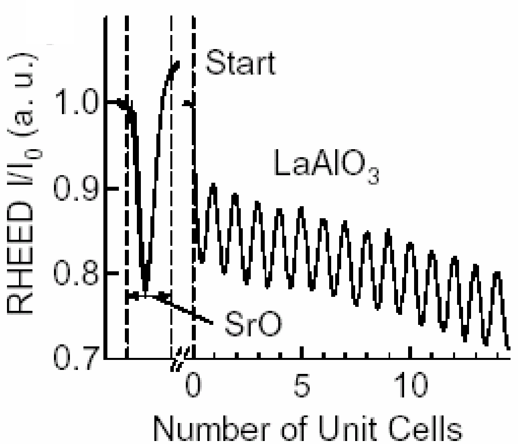
3. Charged Interfaces Between Polar and Nonpolar (Band) Insulators

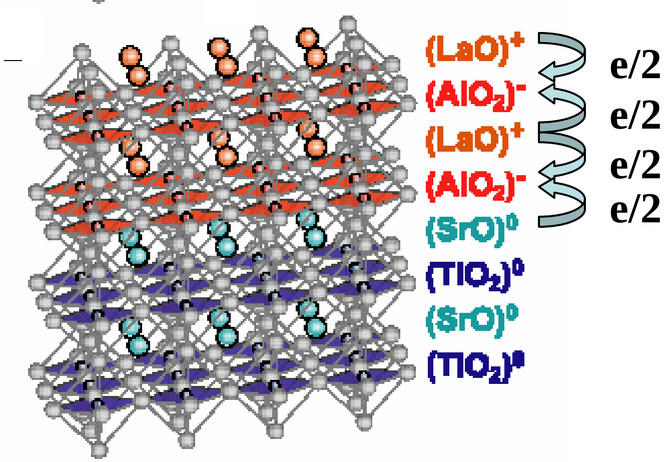
$LaAlO_3$ on $SrTiO_3$





n-type:
Metallic interface – electronic reconstruction

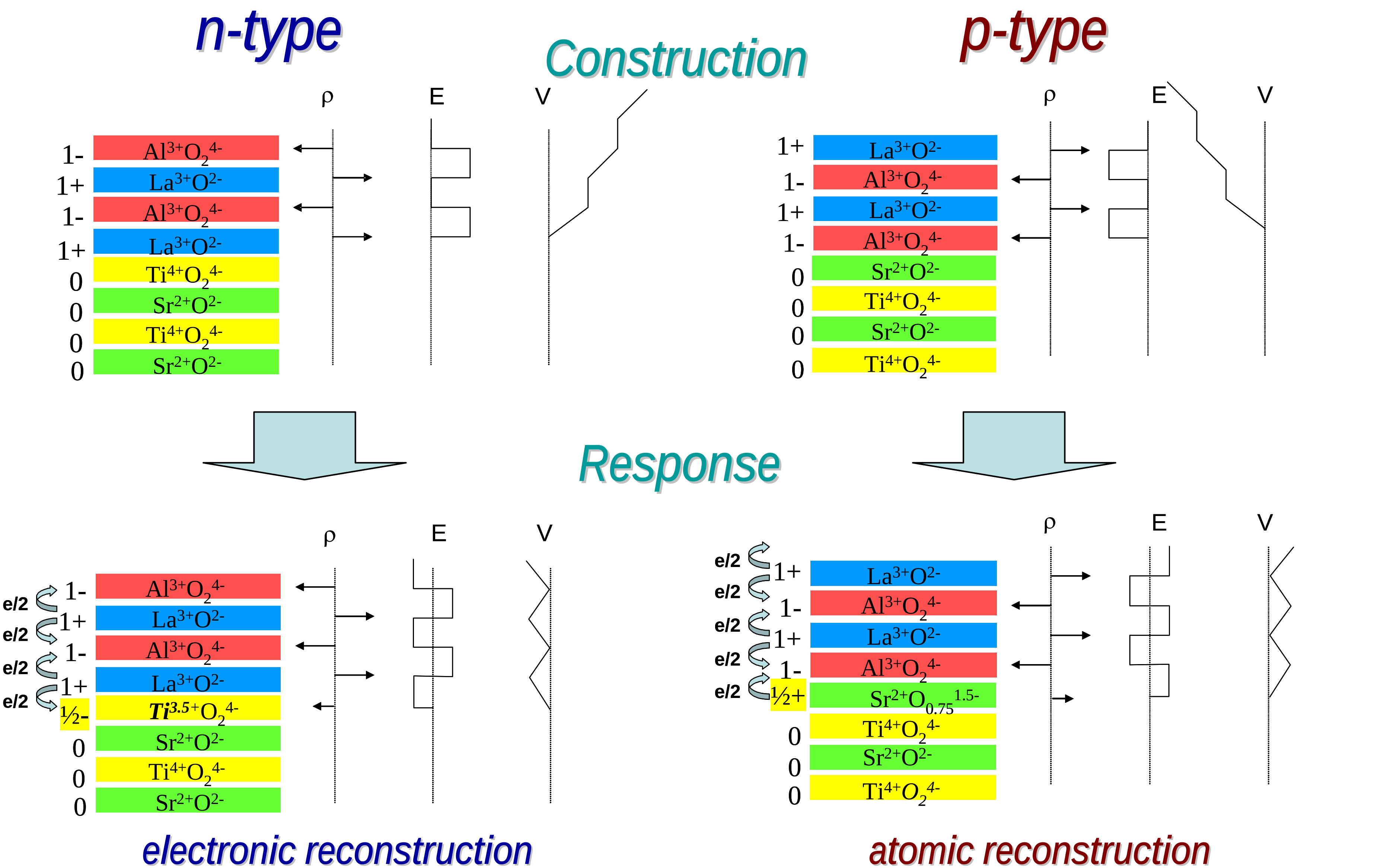




P-type
Insulating interface – atomic reconstruction

A. Ohtomo and H. Y. Hwang, *Nature* 427, 423 (2004).

Resolving the Potential Divergence at the $LaAlO_3/SrTiO_3$ Interface



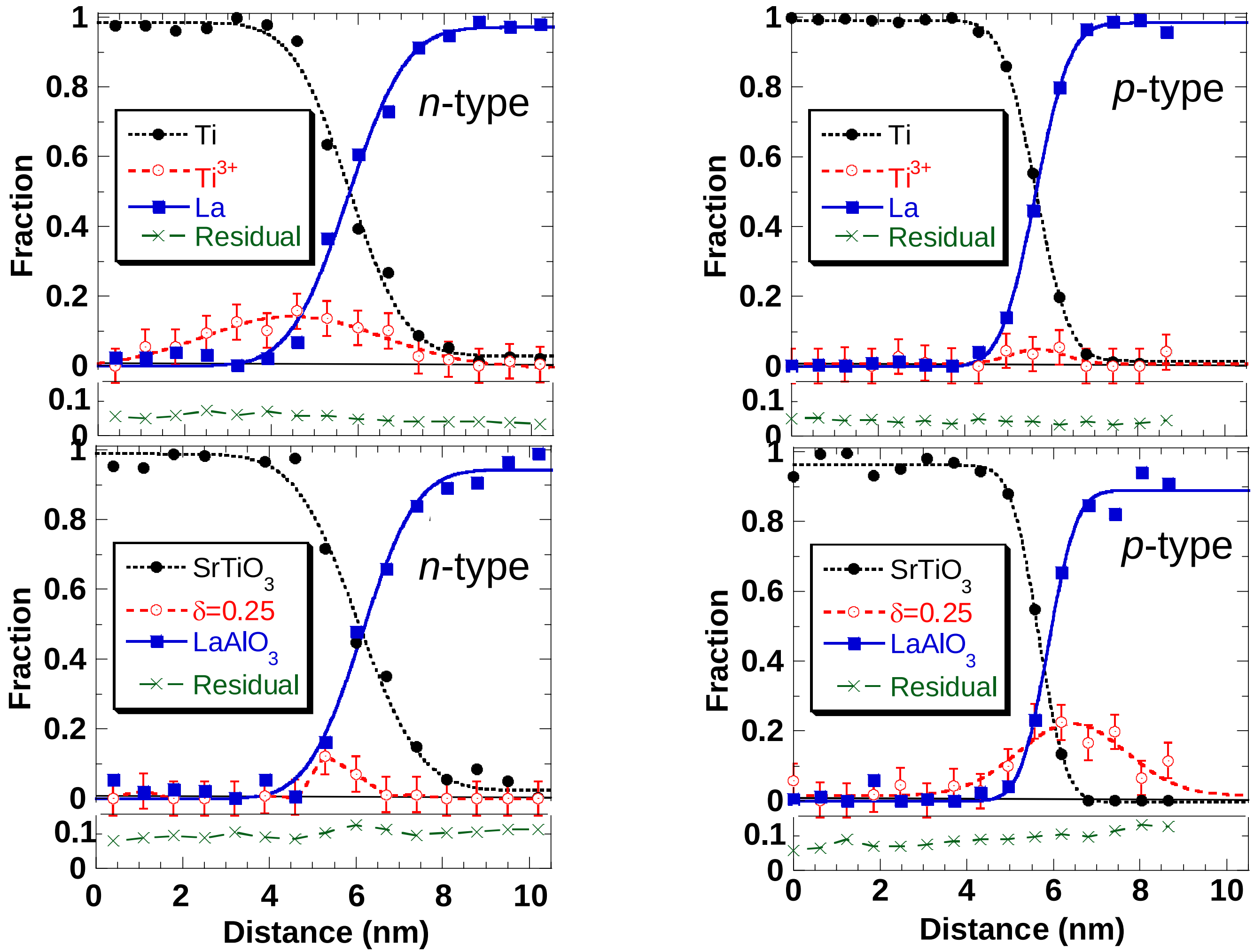
N. Nakagawa, H. Y. Hwang, and D. A. Muller, *Nature Materials* 5, 204 (2006).

Charges at the $SrTiO_3/LaAlO_3$ Interface

n-type ***p-type***

Electrons on Ti sites

Holes compensated by oxygen vacancies



0.7±0.1 e-, 0.2±0.08 h

0.1±0.1 e-, 0.6±0.1 h

Open Issues on the $LaAlO_3/SrTiO_3$ Interface

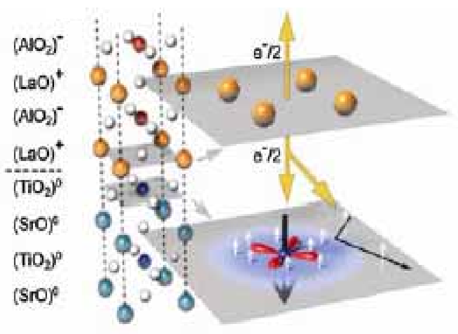
Growth induced oxygen vacancies?

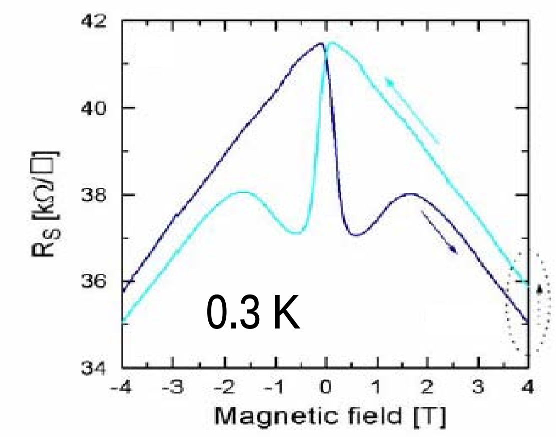
A. S. Kalabukhov *et al*., cond-mat/0603501
W. Siemons *et al*., cond-mat/0603598
G. Herranz *et al*., cond-mat/0606182

Ferromagnetism at the interface?

LDA-U R. Pentcheva & W. E. Pickett, *Phys. Rev. B* 74, 035112 (2006).

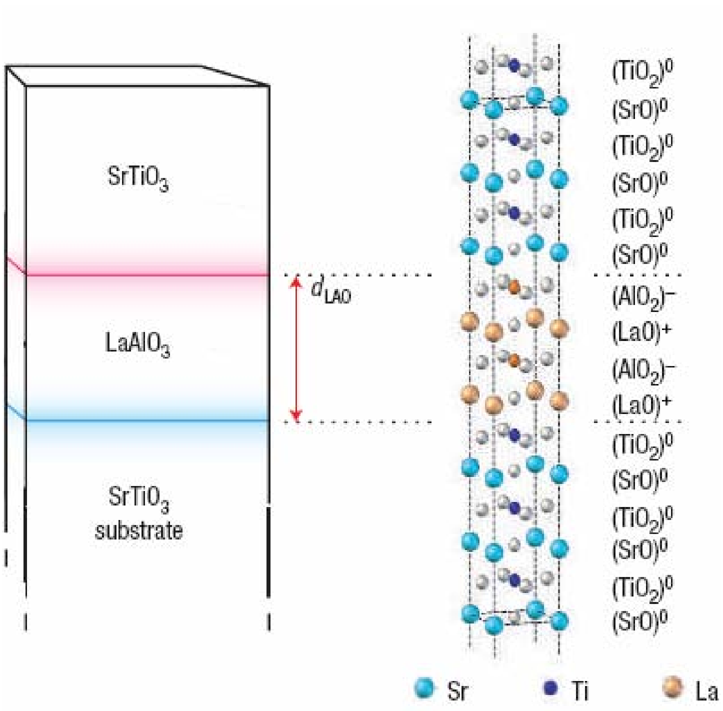
A. Brinkman *et al.*, Nat. Mater. in press.

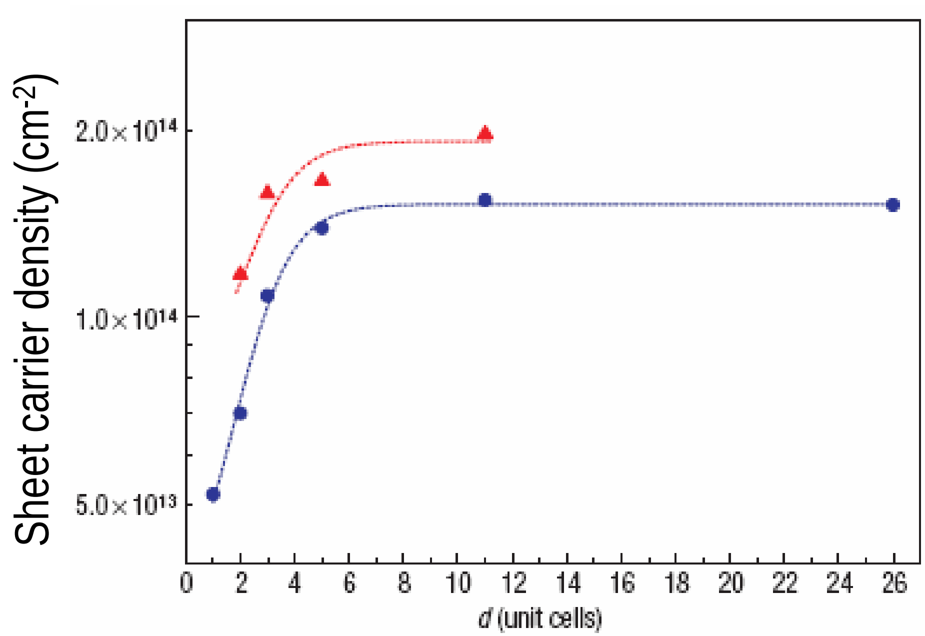




Superconductivity (~200 mK, 2D-KT) Mannhart, Triscone, *et al.*

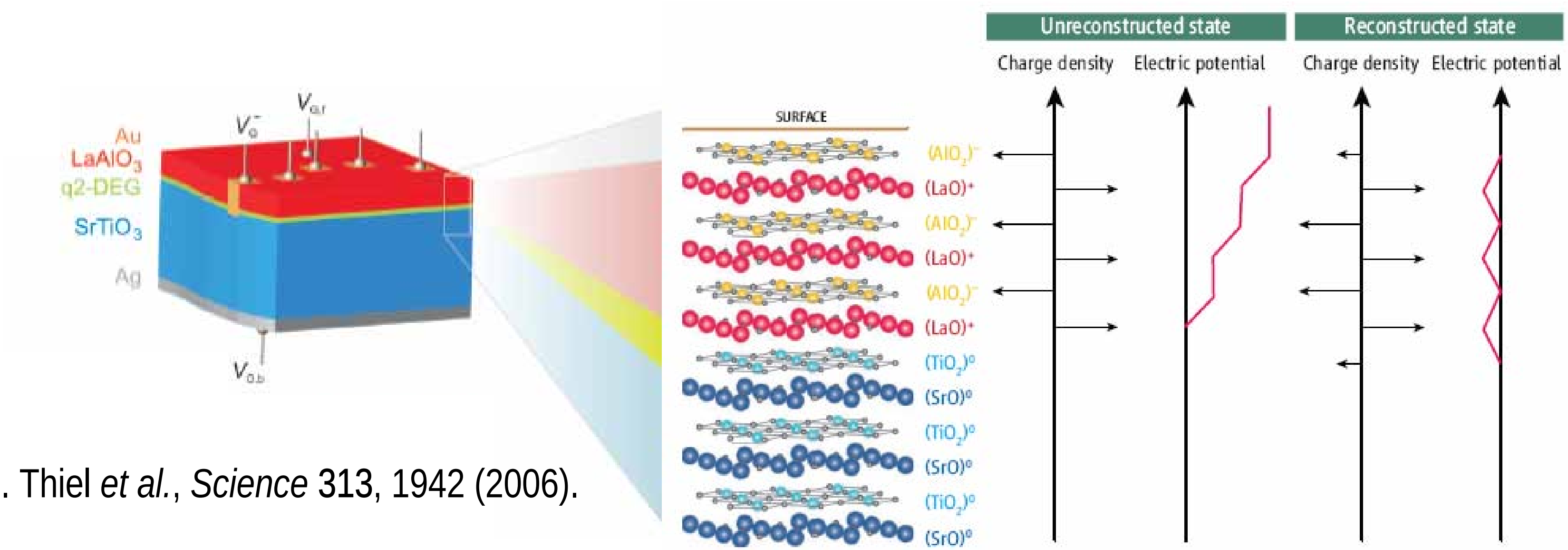
Static Tuning of Polar Interfaces by Proximity Coupling





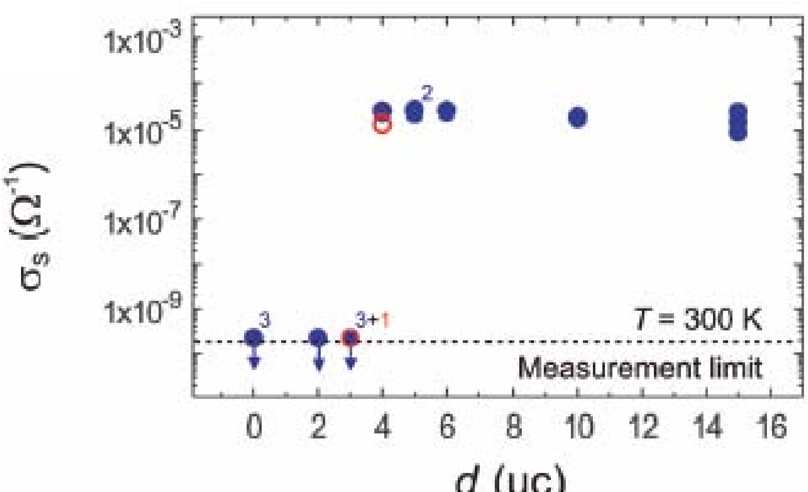
M. Huijben, G. Rijnders, D.H.A. Blank, S. Bals, S.V. Aert, J. Verbeeck,
G.V. Tenderloo, A. Brinkman, H. Hilgenkamp, *Nature Materials* 5, 556 (2006).

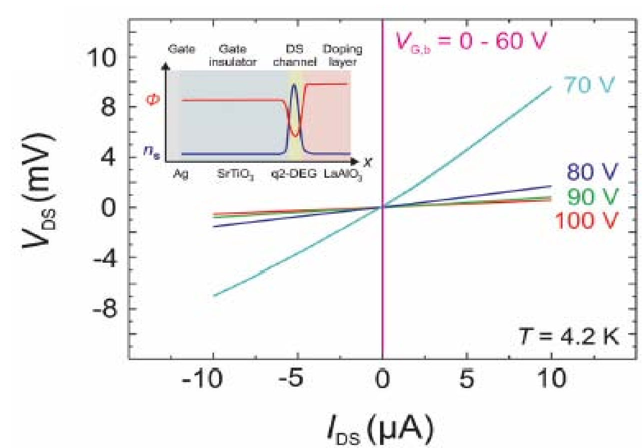
Dynamic Tuning by Gating (HEMT-type structure)



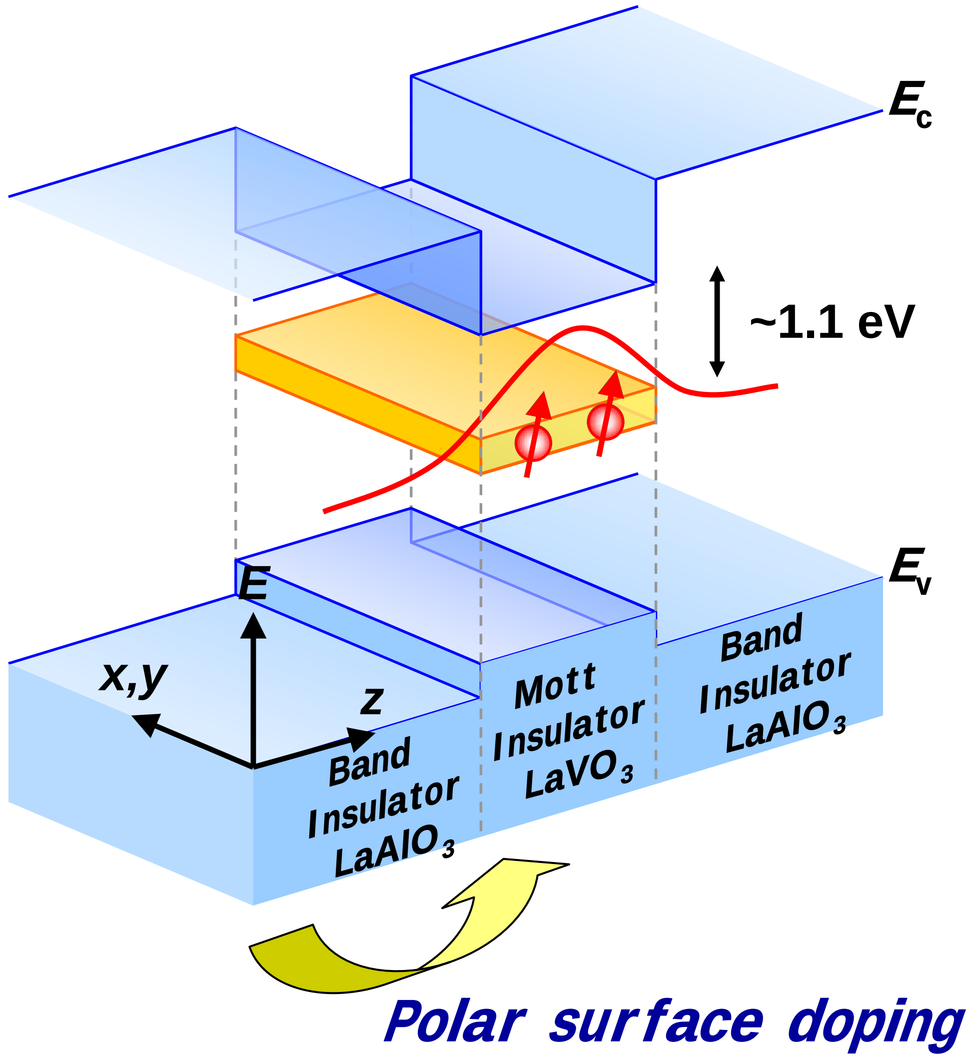
S. Thiel *et al.*, *Science* **313**, 1942 (2006).

H. Y. Hwang, *Science* **313**, 1895 (2006).

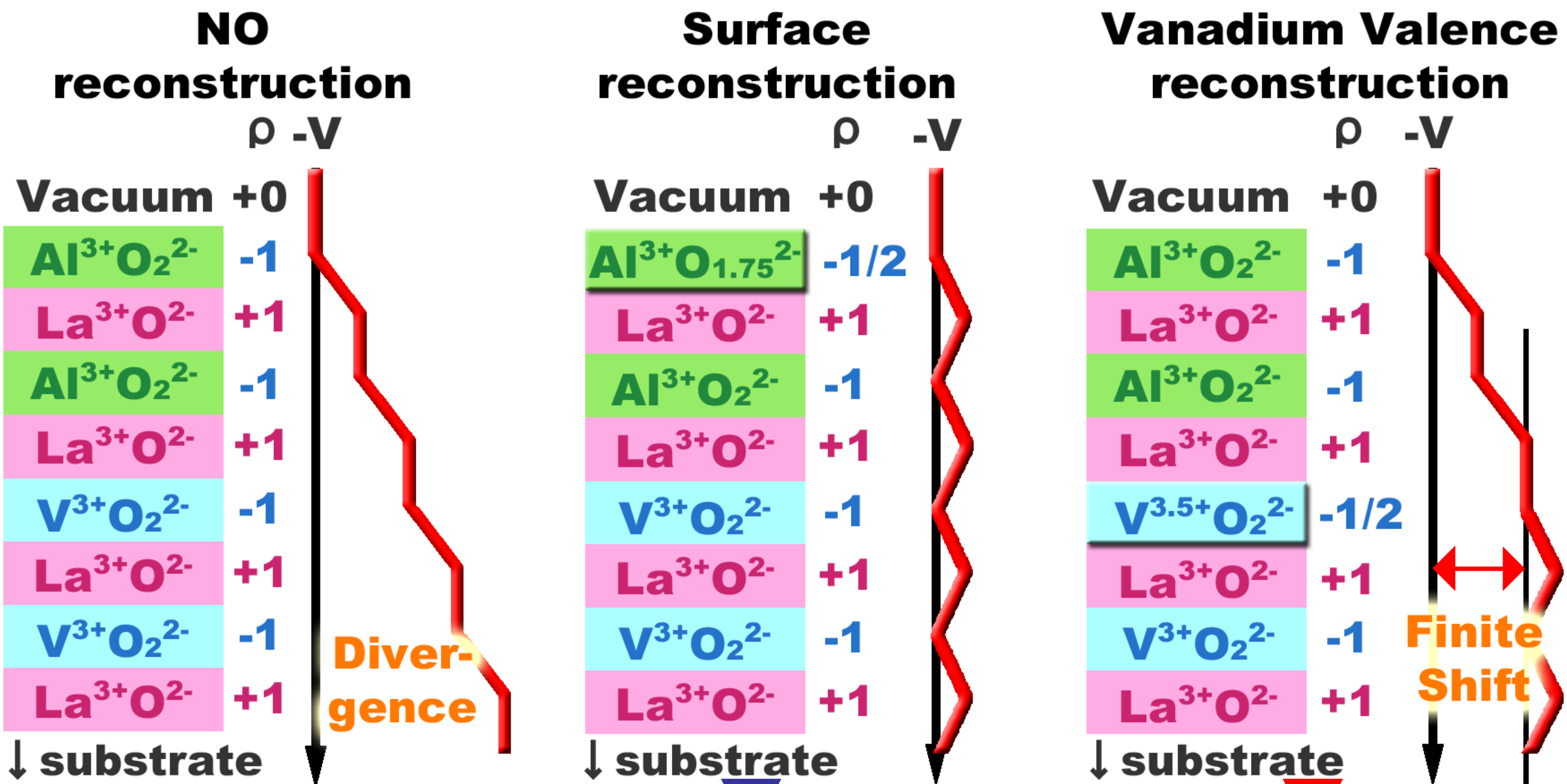




4. Modulation-doping $LaVO_3$ Mott Quantum Wells via Polar Surface



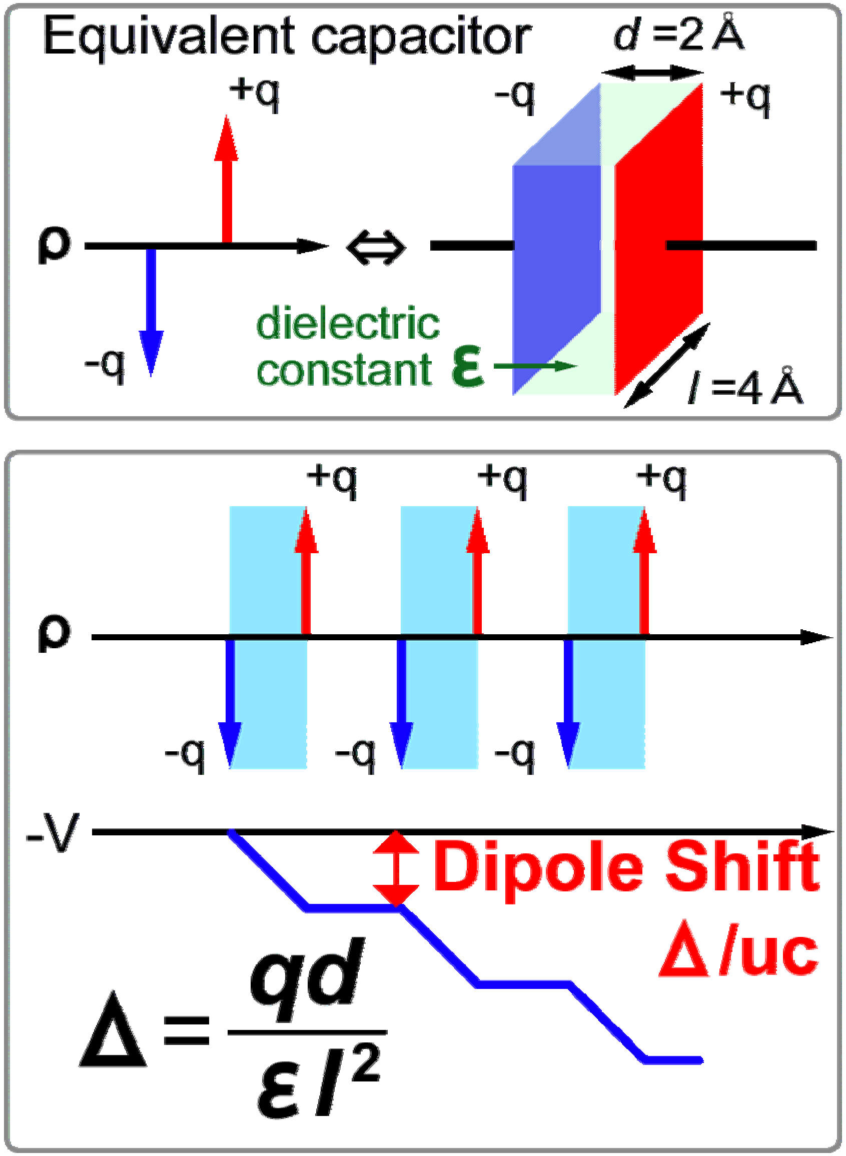
Competing Near-Surface Reconstructions

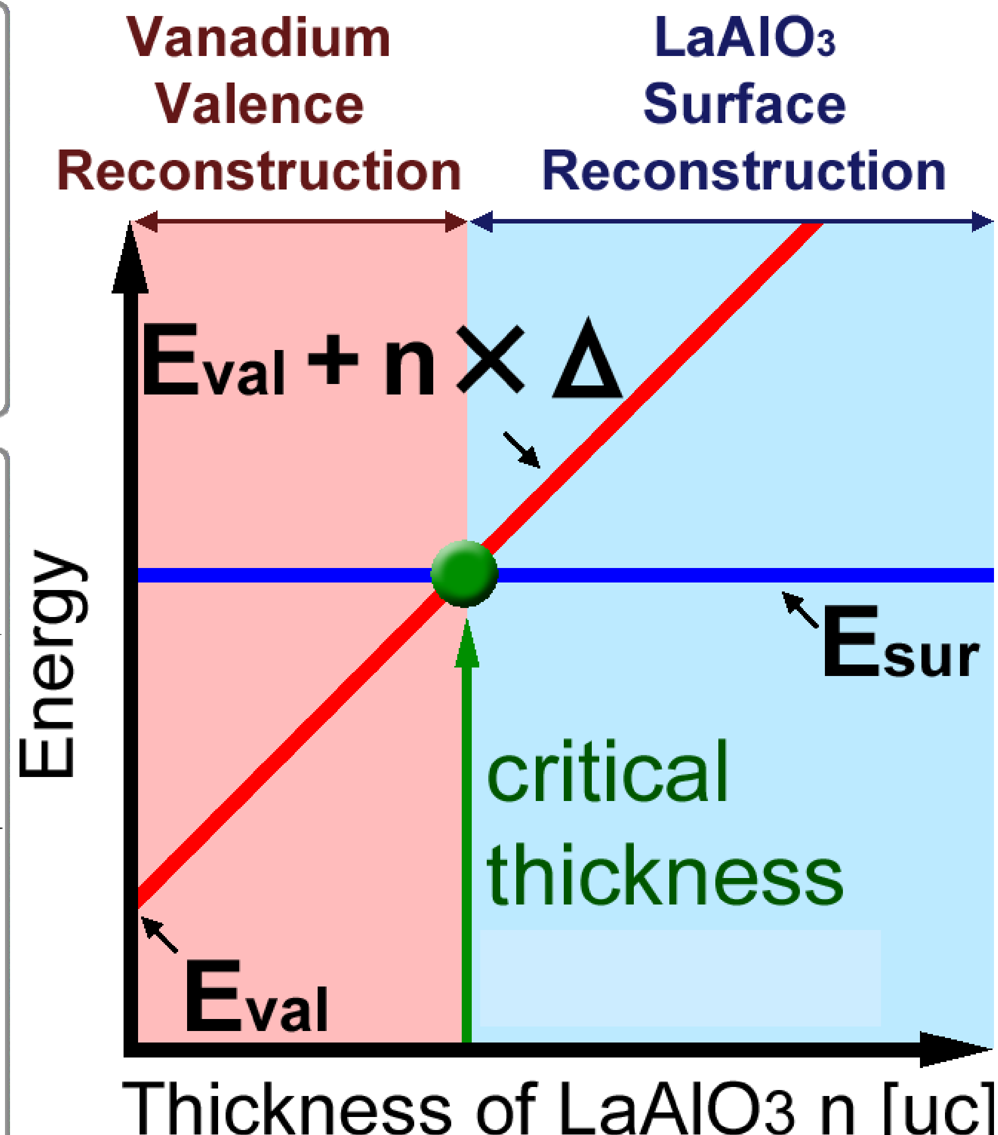


No change in V^{3+}: insulating

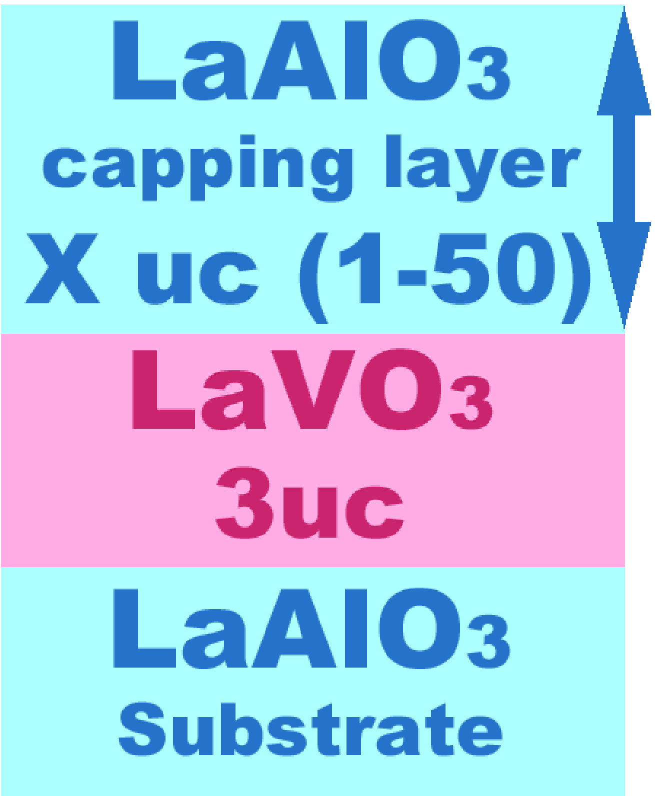
Hole injection: conducting?

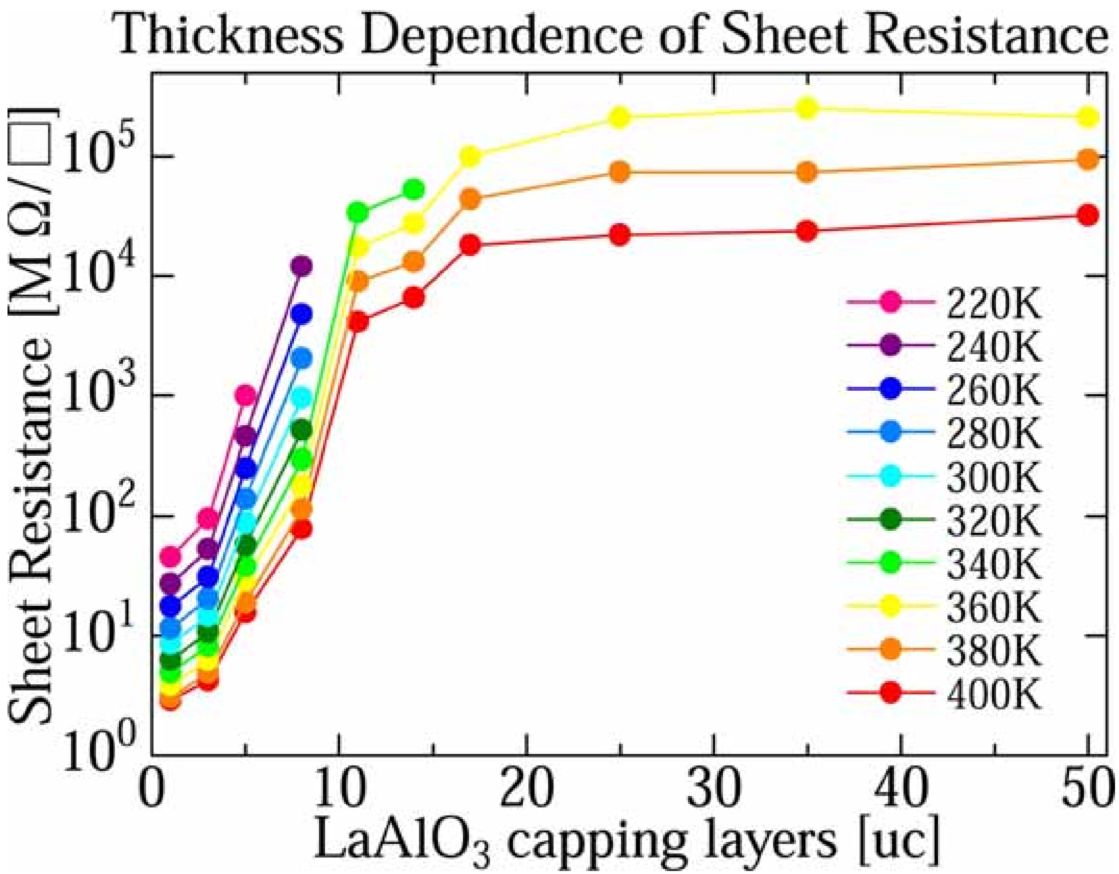
Competing Near-Surface Reconstructions





Hole Doping - Transport





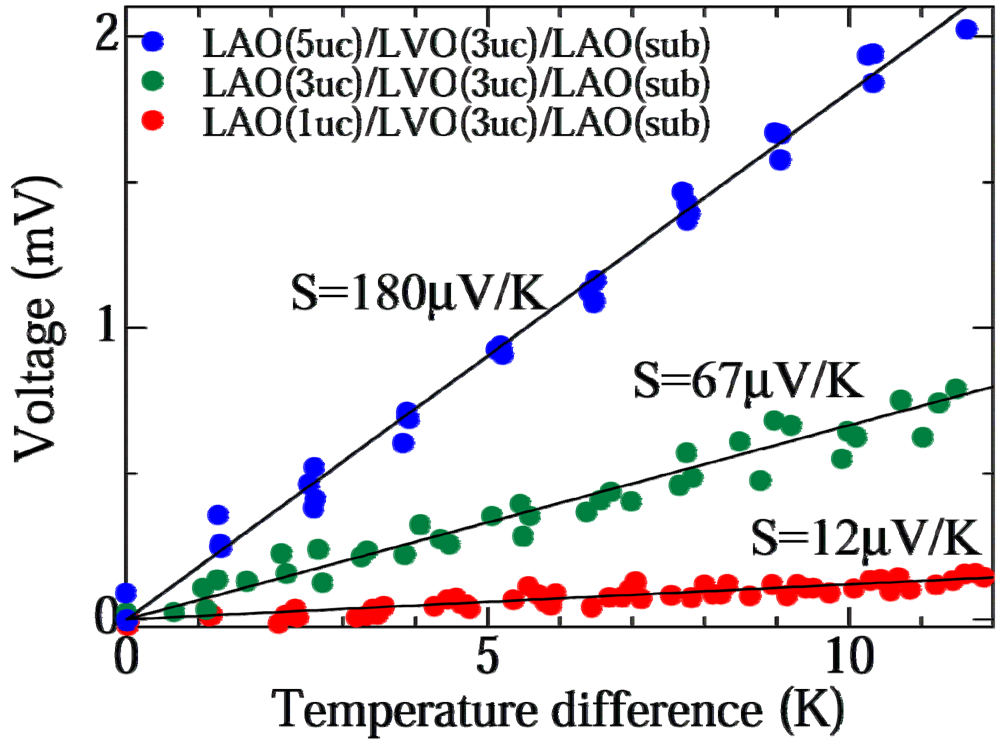
Critical thickness seen in transport

Seebeck Coefficient @ 300K

- Sign of carriers is positive.
  > Carriers are holes
- Seebeck coefficient increased as a function of $LaAlO_3$ capping layers.
  > Fewer holes in samples with thicker capping

Cf. for carriers in a parabolic band

$$S = \frac{k_B}{e}\left(\frac{5}{2} + r + \ln\left\{\frac{2(2\Pi mkT)^{3/2}}{nh^3}\right\}\right)$$



> Consistent with the reconstruction coupling scenario

Conclusions

(i) Mixed valence and lattice polarizability allow significant electronic reconstructions at perovskite heterointerfaces

(ii) By embedding Mott insulators in wide bandgap hosts, a Mott quantum well can be formed.

(iii) A proximate polar surface/discontinuity can be coupled to modulation dope the well.