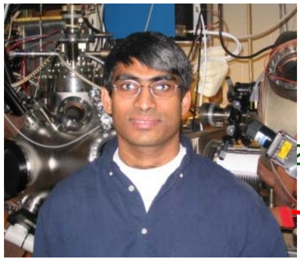
New collective and single particle phenomena at oxide interfaces and digital superlattices



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New phenomena at oxide interfaces (3 topics)

What are the new results?

- global symmetry can be controlled by architecture — Warusawithana
- design SL to obtain new optical transitions, σ(ω) — Zhai
- superconductivity with e-h antisymmetry — Davidson

Global symmetry:

Make a digital superlattice out of 3 dielectric phases and use SL architecture that breaks inversion symmetry

→ dielectric has permanent polarization and finite $\chi^{(2)}$.

New electronic transitions

Digital superlattice to mix bands at interface

→ new optical transitions between composite states.

Modified superconductivity at interface (unexpected)

a-axis interface between YBCO and CaTiO3 (NIS tunnel jct)

→ crystalline interface causes DoS measured by tunneling to have broken e-h symmetry at interface (not like BCS)

topic 1

Nanostructured asymmetric dielectrics

summary

Lattice strain distorts bonds at heterointerfaces

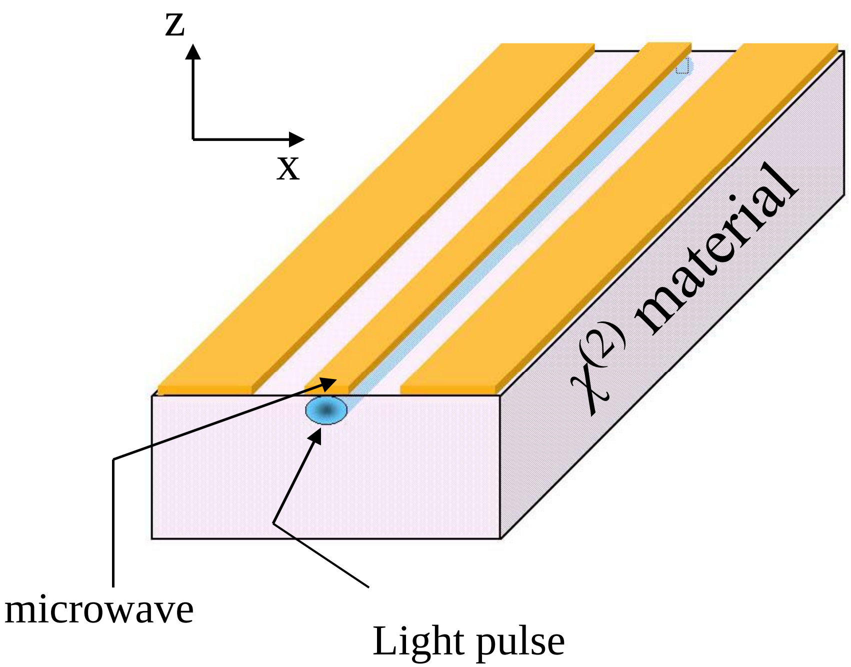
- asymmetric supercell → asymmetric strain "field"

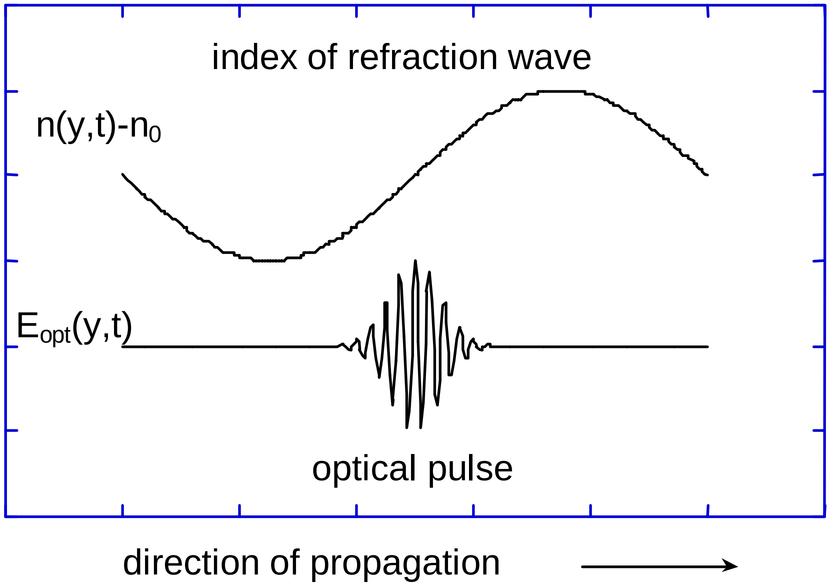
Supercells with broken inversion symmetry lead to self-poled materials (similar to ferroelectricity, but…)

- asymm. strain field acts like effective bias field, E_{eff}
- permanent polarization, with no two state switching

Strain "proximity effect" extends about 2 uc

Electro-Optic Frequency Shifter





$$E_\mu = E_0 \sin(ky - wt)$$

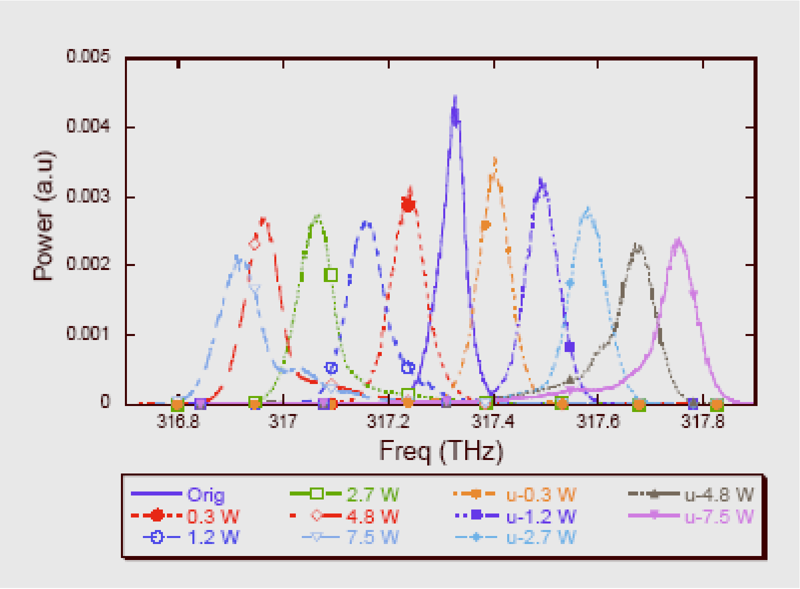
Index of refraction experiences a dynamic change given by

$$\delta n = -\frac{1}{2} n_0{}^3 r_c E_\mu$$

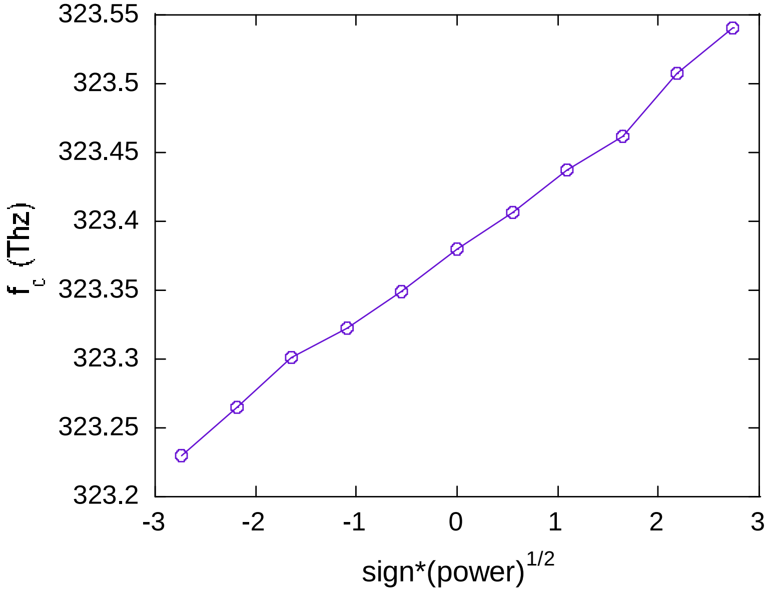
D. A. Farías and J. N. Eckstein. “Coupled-Mode Analysis of an Electro-optic Frequency Shifter,” IEEE J. of Quant. Elect. Vol 39. No 2. , pp. 358-363, Feb. 2003.

Experimental Results EOFS

power vs frequency



Frequency Shift vs. Microwave Power

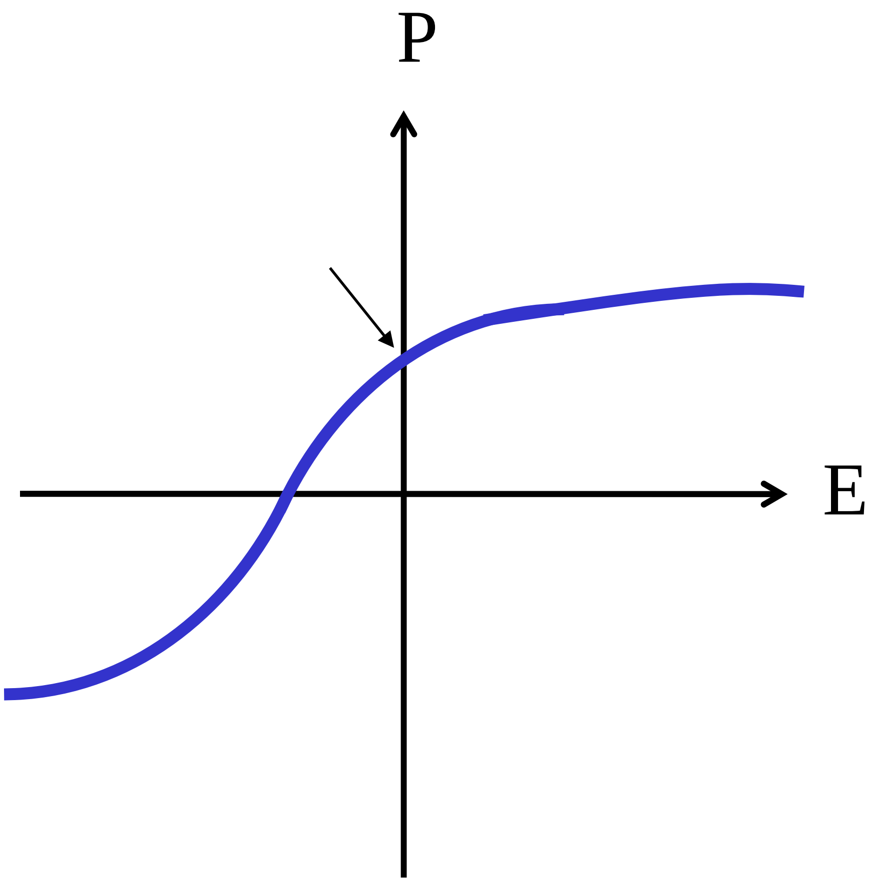


Frequency shifting proportional to $P^{1/2}$

Experiments done with 927 nm., 20 ps. pulses at a 75.7 MHz repetition rate. Microwave was chosen at the 80th harmonic (6.056 GHz), and powers of 0, 0.3, 1.2, 2.7, 4.8, and 7.5 Watts were applied to the modulator. Freq shifting range = 1 THz **limited by r_c**

D. A. Farías and J. N. Eckstein. IEEE J. of Quant. Elect. Vol 41. No 1. , pp. 94-99, Jan 2005.

What you would like for non-linear modulators, etc…



For non-linear optical and other field tuning applications would prefer a characteristic like this.

Stable, single solution
can't de-pole

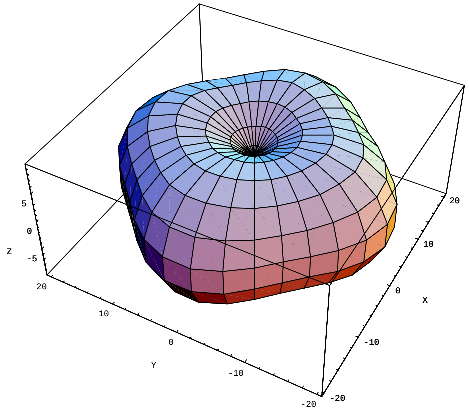
Permanently polarized dielectric with big $\chi^{(2)}$

Use molecular nanostructuring to make such a material (MBE)
(once you figure out what matters!)

Artificial structures

using ALL-MBE to synthesize materials and heterostructures not found in nature

anisotropy energy surface
($La_{0.7}Ca_{0.3}MnO_3$ on $SrTiO_3$ substrate)

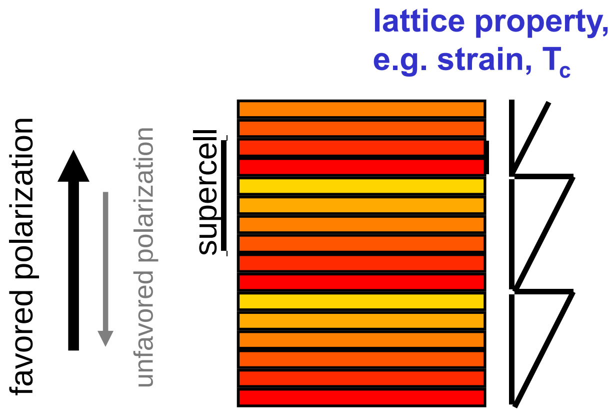


Controlling material properties via epitaxial strain

Tensile strain-induced magnetic anisotropy in magnetic oxide

Producing new “materials” by modulated heterostructure growth

Grow crystal using ferroelectric and related phases: stack with *structurally broken c-axis inversion symmetry*

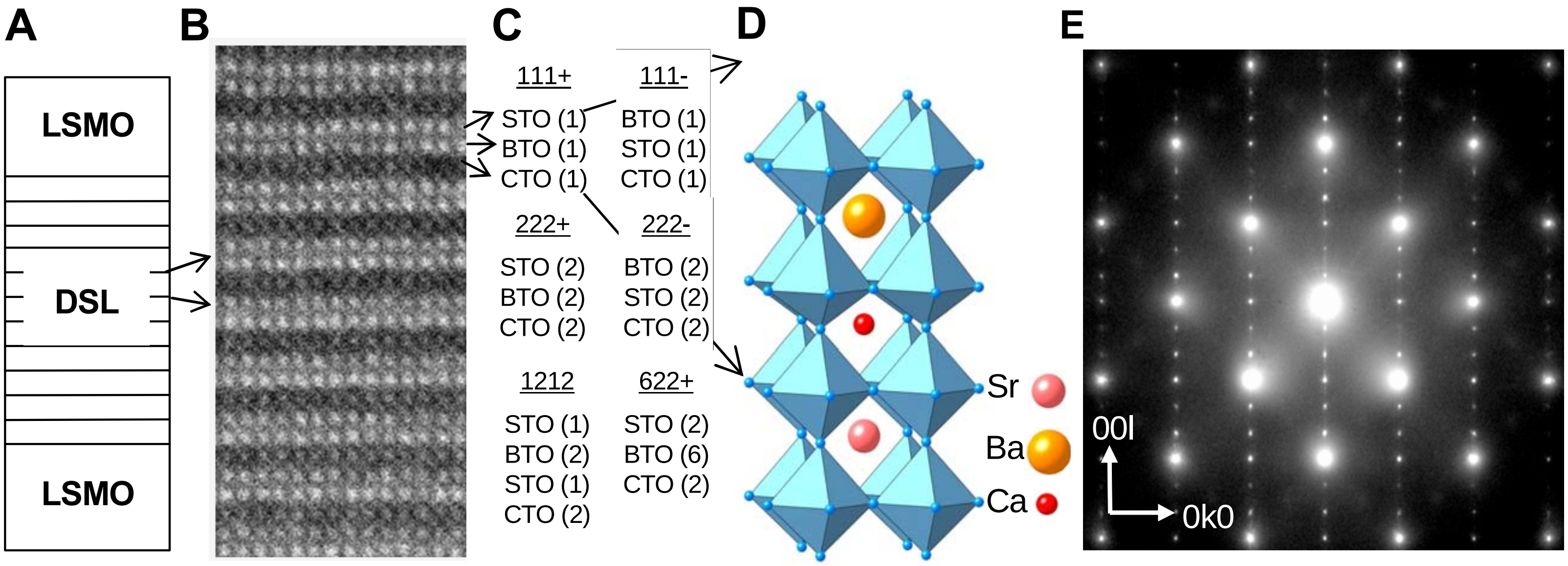


- **broken symmetry *throughout* film favors one polarization**
- **permits stable operation nearer to Curie temperature**
- **obtain larger response at zero bias**

Electrodynamic properties of atomically layered “meta-materials”

Asymmetric dielectric superlattices

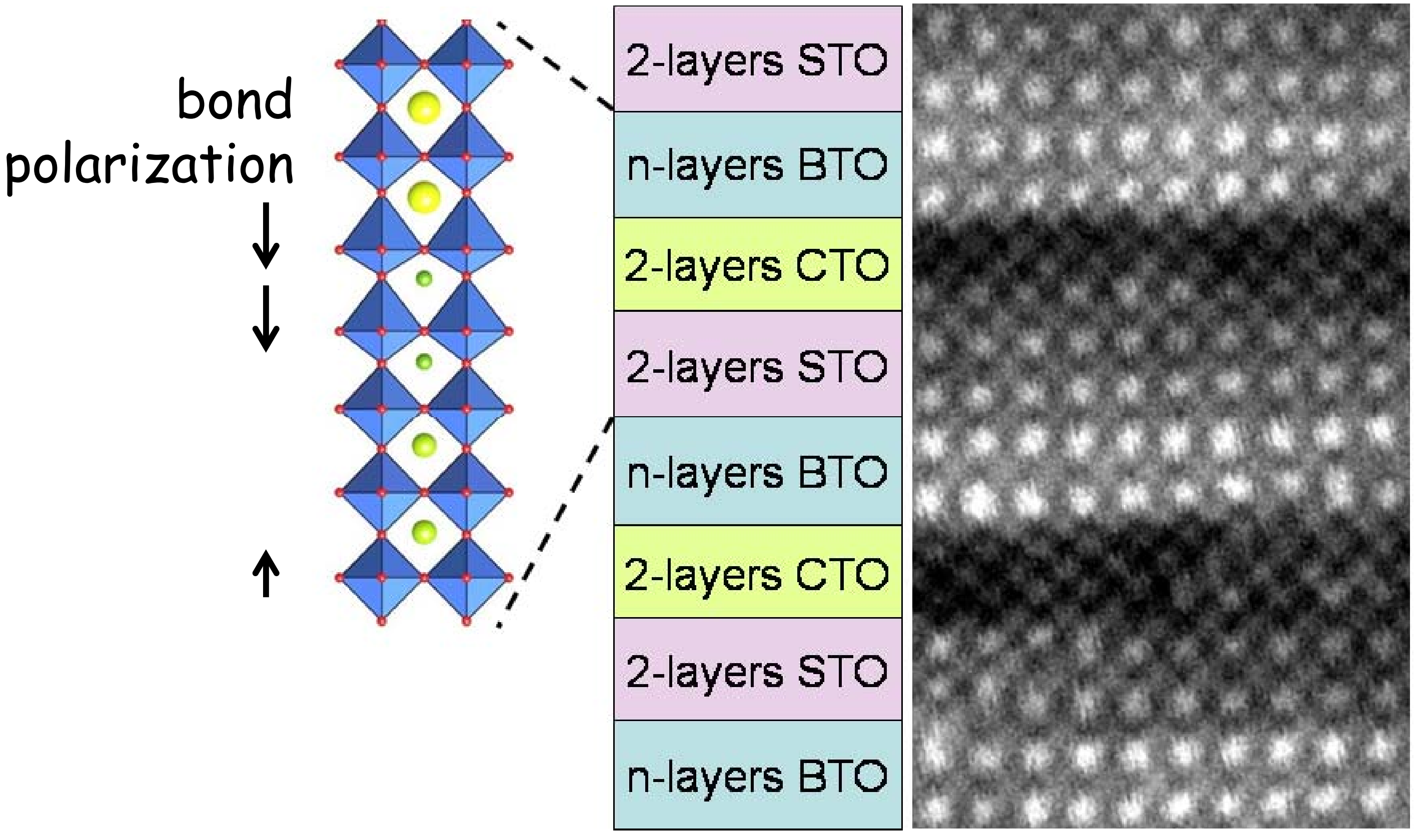
broken inversion symmetry



Calculation by Mike O'Keefe ASU
Microscopy by J. Zuo and H. Chen UIUC

bond valence sum calculation

Mike O'Keefe (ASU)



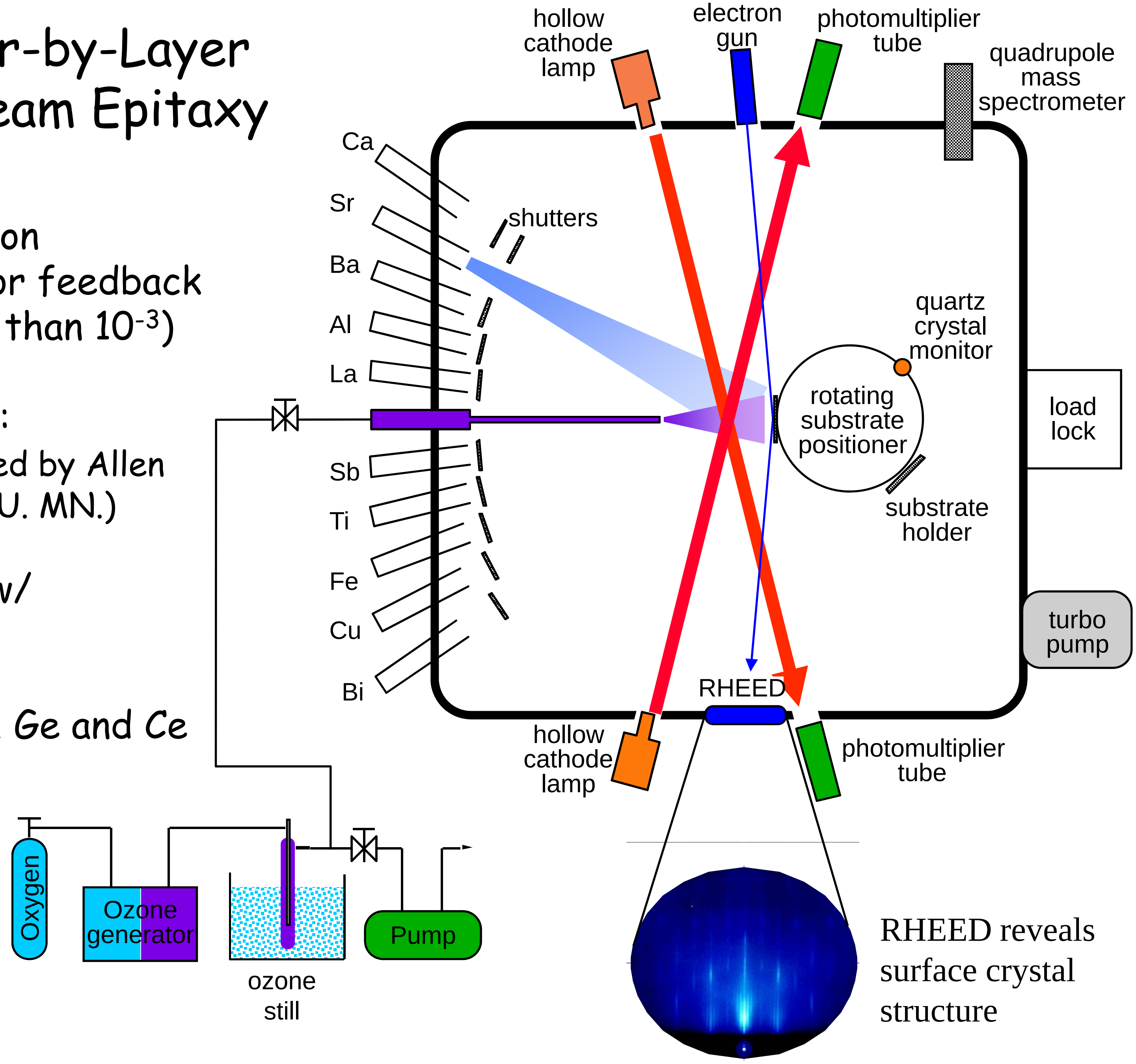
2-layers STO
n-layers BTO
2-layers CTO
2-layers STO
n-layers BTO
2-layers CTO
2-layers STO
n-layers BTO

Z-contrast STEM image

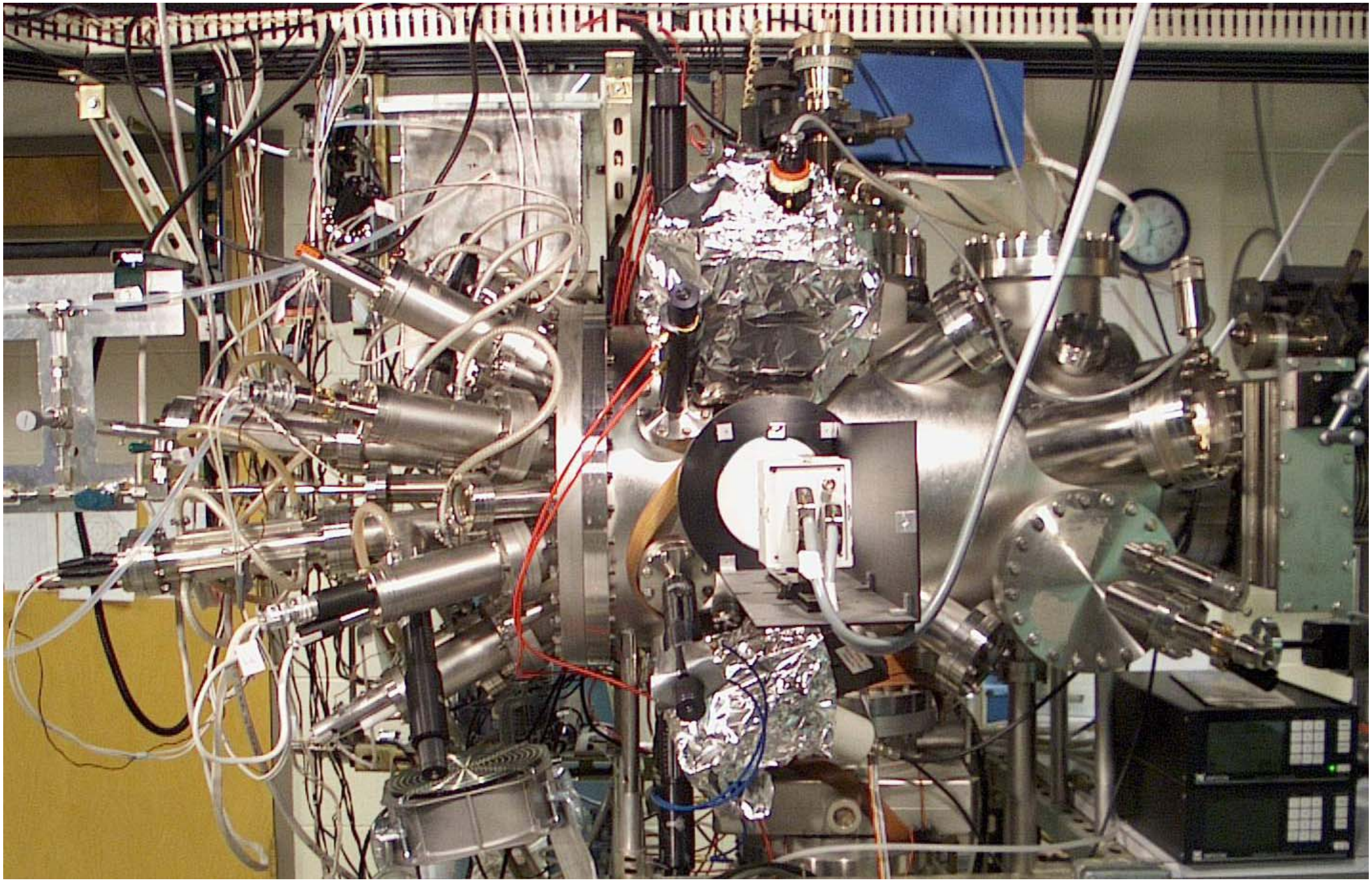
Hao Chen UIUC
Jim Zuo UIUC

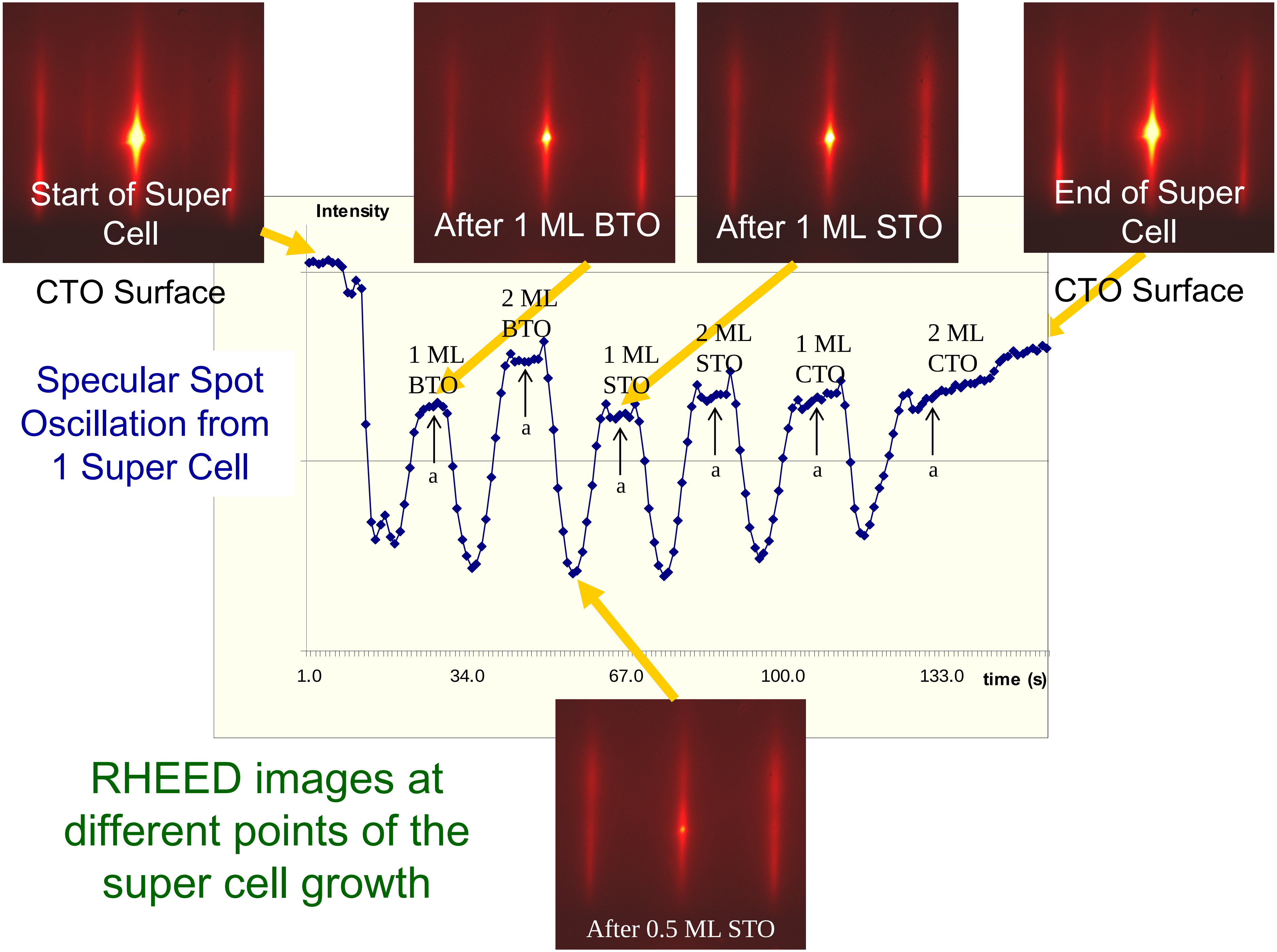
Atomic Layer-by-Layer Molecular Beam Epitaxy

- atomic absorption spectroscopy for feedback control (better than 10^{-3})
- ozone oxidation:
  (introduced by Allen Goldman, U. MN.)
- in-situ RHEED w/ **digital video**
- 12 sources, incl. Ge and Ce



RHEED reveals surface crystal structure

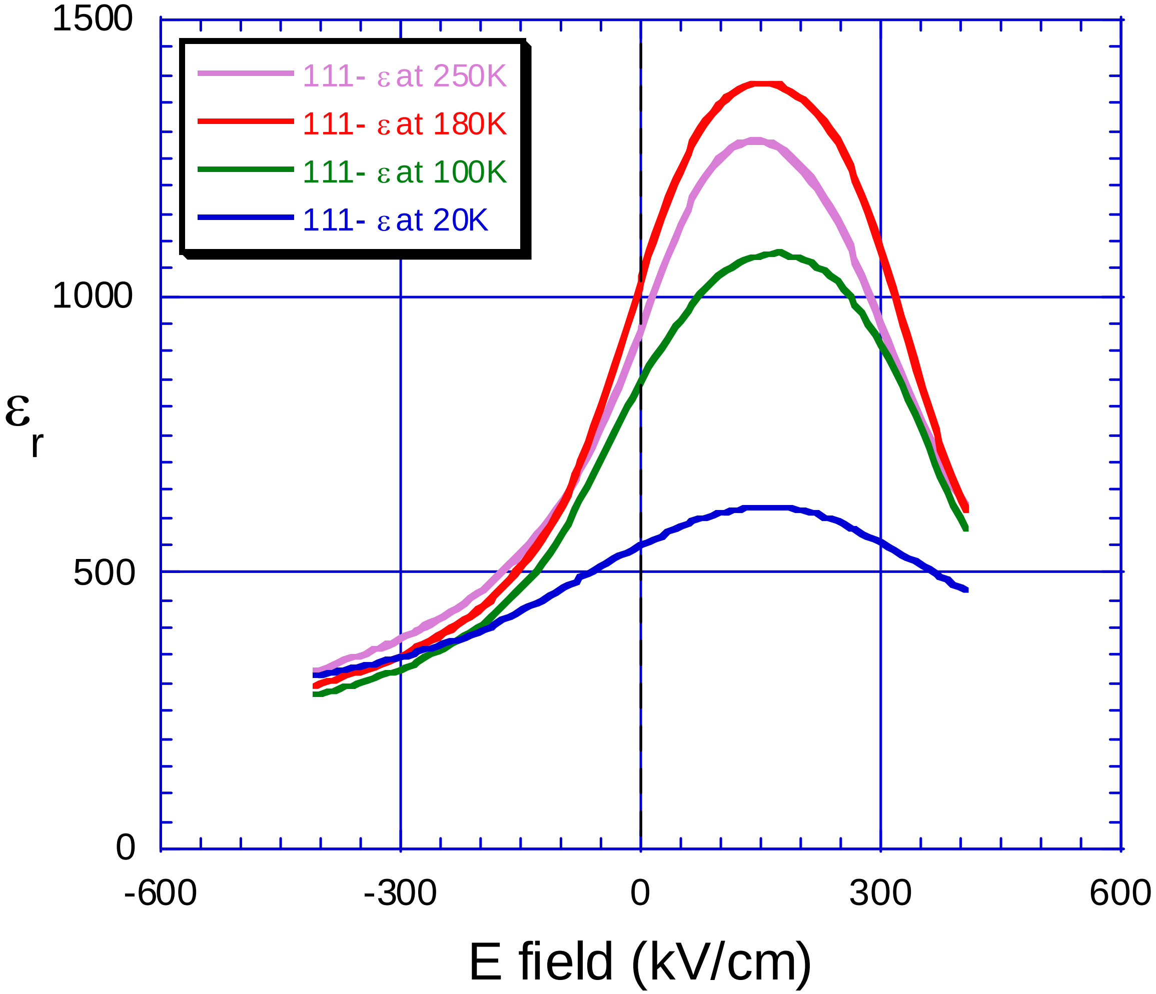




Specular Spot Oscillation from 1 Super Cell

RHEED images at different points of the super cell growth

digital superlattice
BTO-STO-CTO
111-

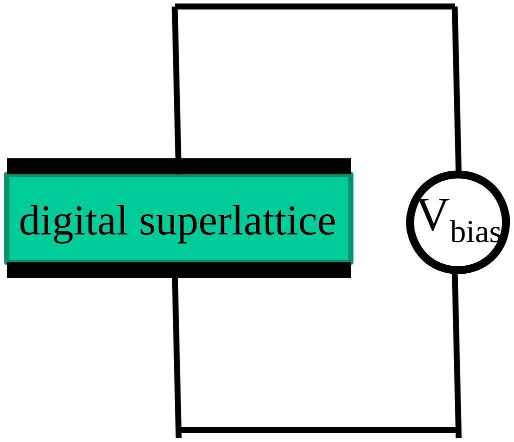


Behaves as though subject to effective bias field ~150 kV/cm

P(E=0) = 0.17 C/m^2

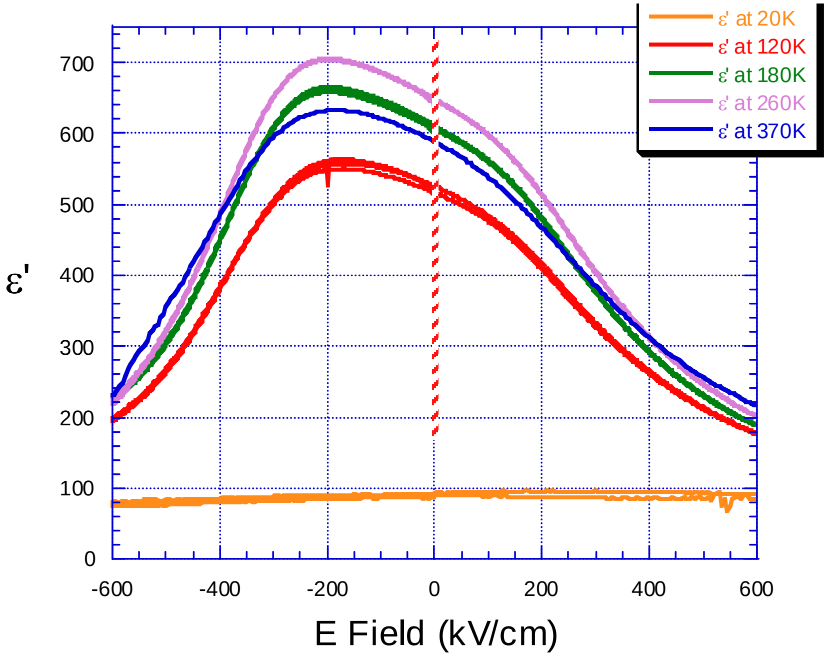
Samples without broken inv. sym. have sym. Curves

Sign of P depends on + or – superlattice architecture

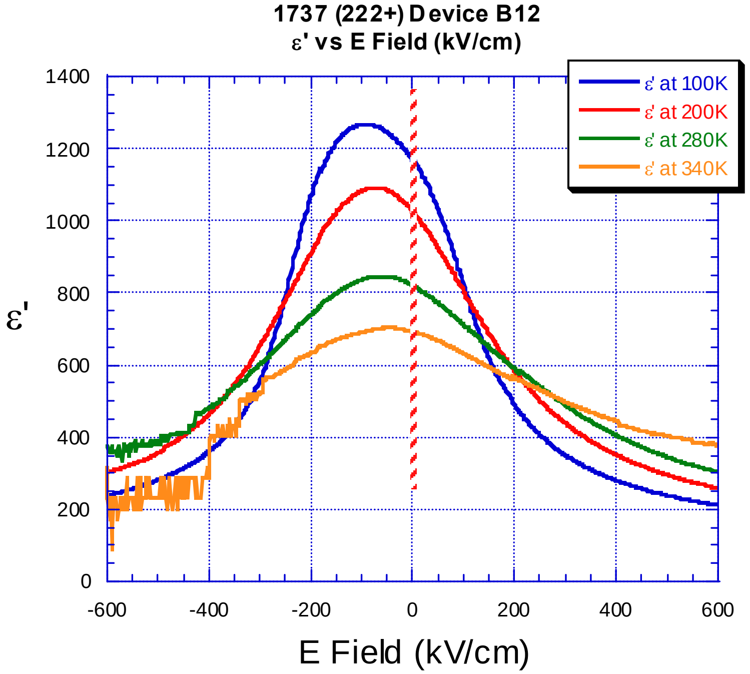


proximity effect

422+

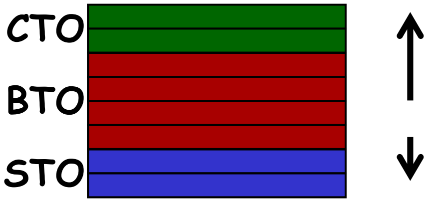


222+



422+ appears to be made up of a superposition of a larger (+) curve plus a smaller (-) curve

(+) response comes from stronger CTO/BTO heterointerface, and….



heterointerfacial strain → bond distortion → polarization
This says that the polarization extends ~ 2uc away from int

topic 1

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Lattice strain distorts bonds at heterointerfaces

asymmetric supercell → asymmetric strain "field"

Supercells with broken inversion symmetry lead to self-poled materials (paraelectric)

asymm. strain field acts like effective bias field, E_{eff}

permanent polarization, with no two state switching

Strain "proximity effect" extends about 2 uc

topic 2

New optical absorption bands in digital superlattices

summary

- Short period digital superlattices of perovskite phases having different TMs can be accurately grown.

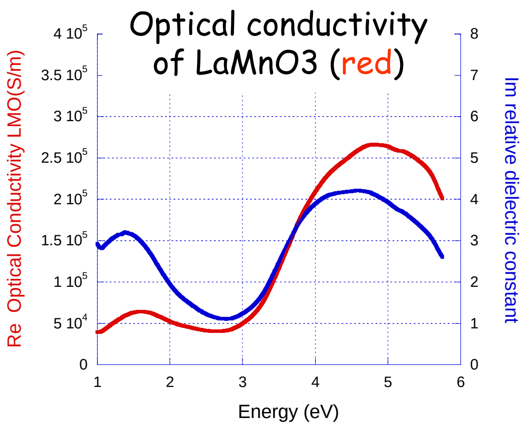
- New optical transitions can be created by placing different molecular layers that electronically hybridize next to each other

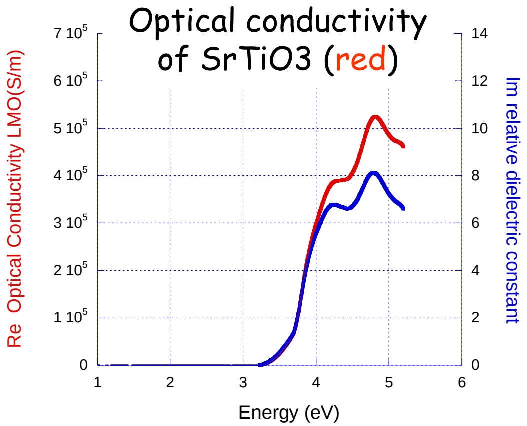
- Electron transitions from source state to destination state
    - Source state is mainly from one molecular layer
    - Destination state is mainly from other molecular layer

- Measure heterojunction band line-up

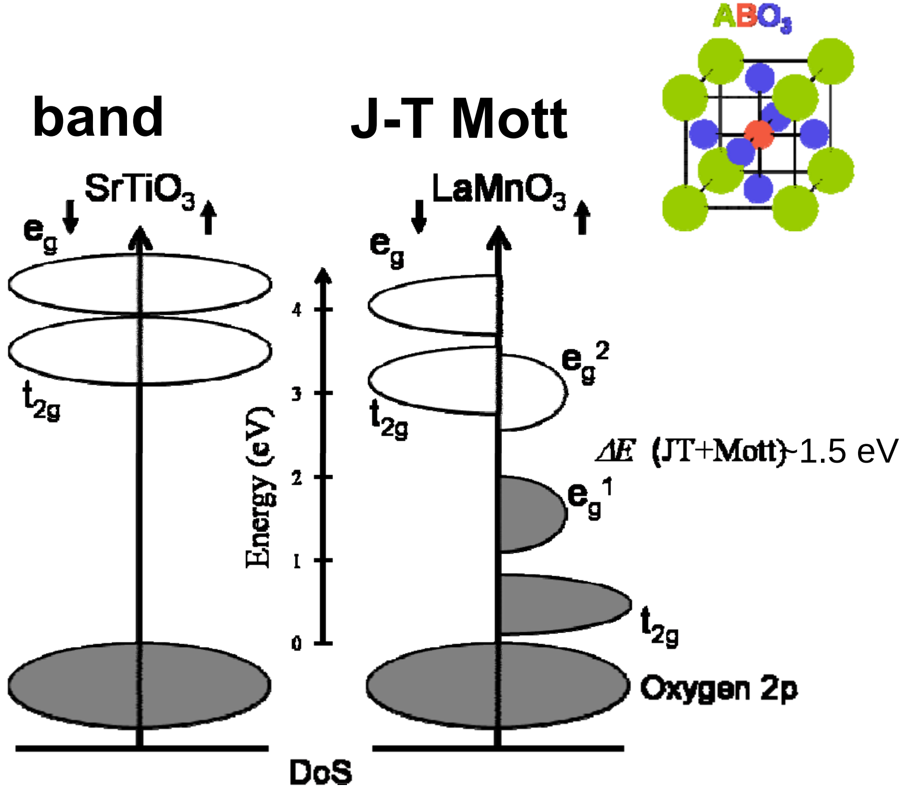
New optical absorption bands in atomically layered digital superlattices

Combine Jahn Teller - Mott Hubbard insulator with band insulator


Optical conductivity of LaMnO3 (red)


Optical conductivity of SrTiO3 (red)

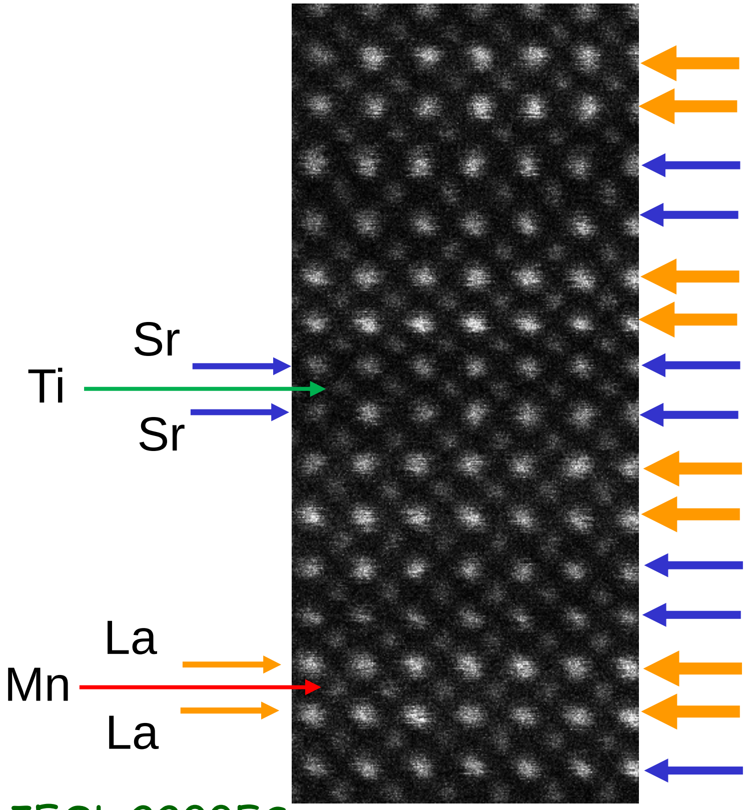
band line-up



LMO is an A-type antiferromagnet below 140 K, FM planes stacked AFM-ly

New optical absorption bands in atomically layered digital superlattices

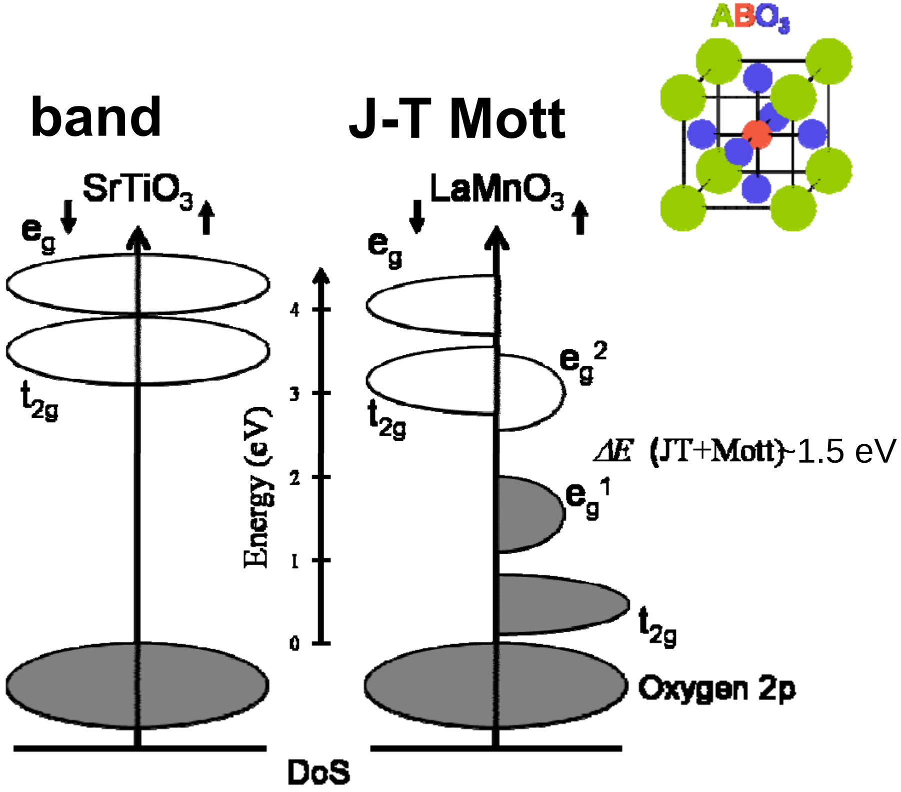
Combine Jahn Teller - Mott Hubbard insulator with band insulator



JEOL 2200FS
with probe forming
Cs Corrector

J-M Zuo group
Amish Shah

band line-up



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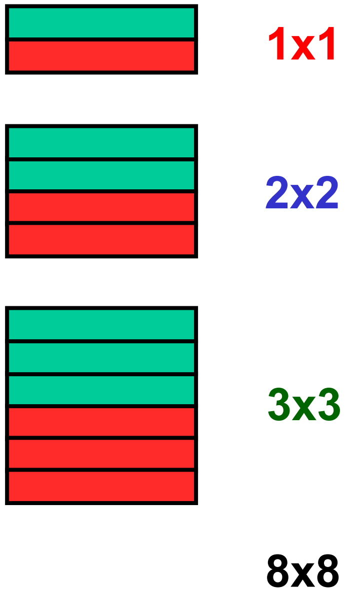
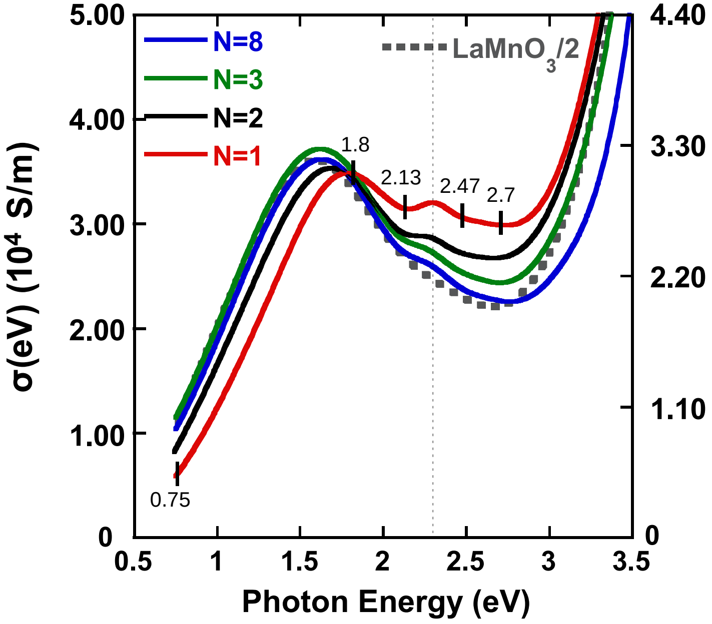
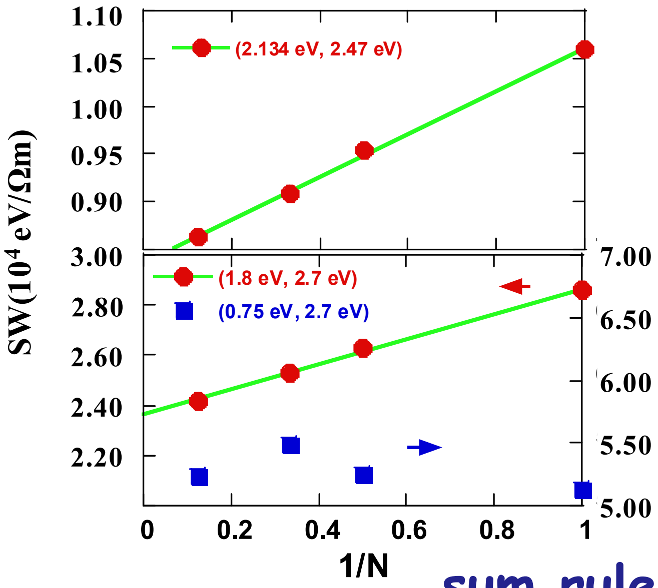
New bandstructure in short period superlattices

Key: 🟩 $SrTiO_3$
🟥 $LaMnO_3$







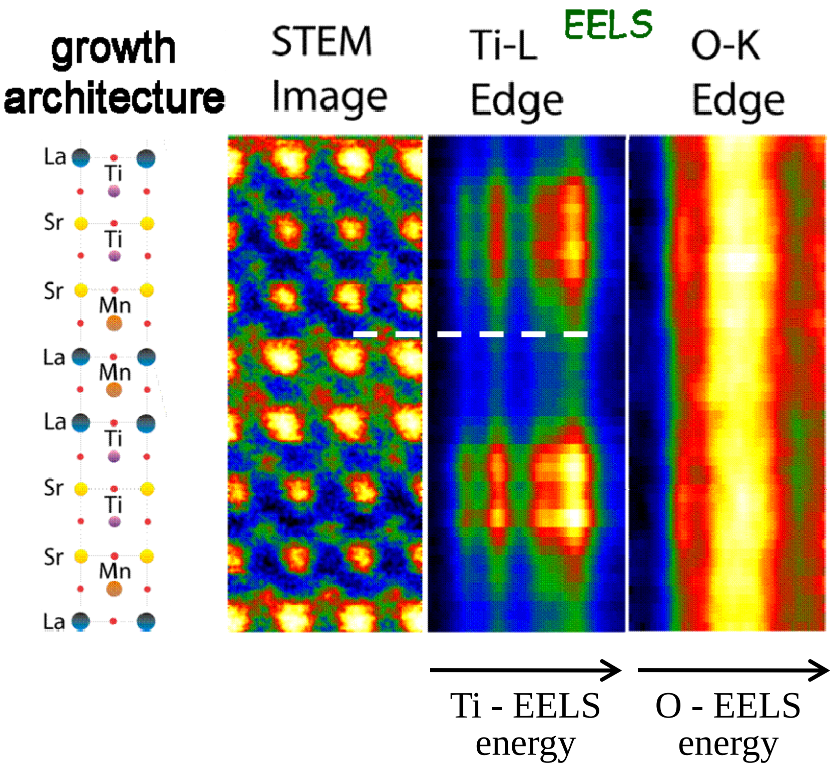
sum rule

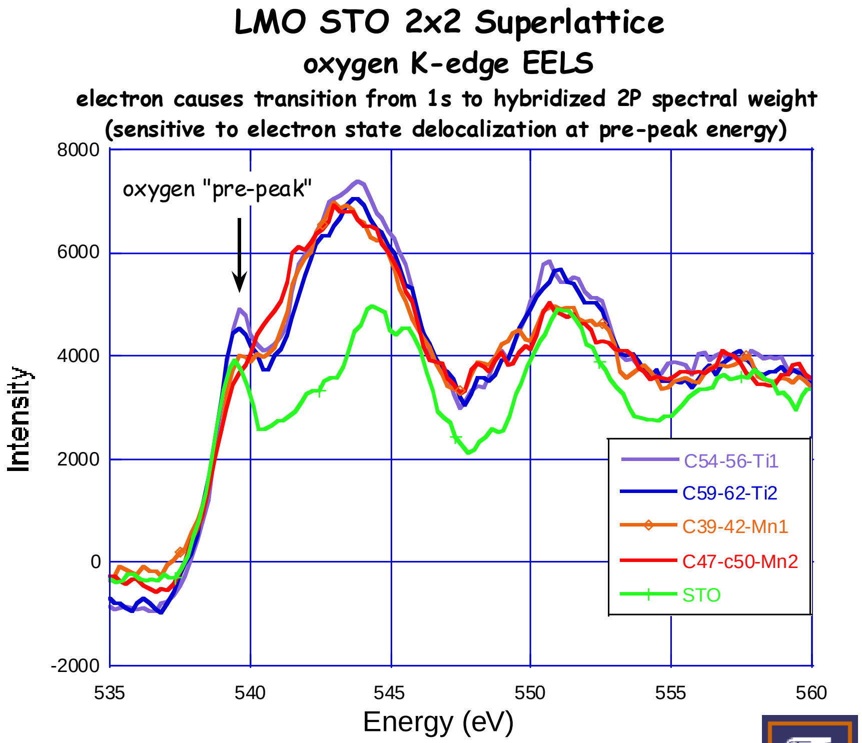
- Like GaAs/AlAs minibands, but here the quantum mechanically blended phases are very different: STO is band insulator while LMO is insulator due to e-ph and e-e interactons
  - Jahn Teller and Mott Hubbard
- *1.5 eV peak (MH-gap) blue shifts in shorter period SL*
- *New spectral weight emerges between 1 and 3 eV, and SW lost below 1.7 eV.*

Oxygen EELS shows similar spectra in Ti and Mn layers

Further evidence that the metal wavefunctions appreciably hybridize with oxygen

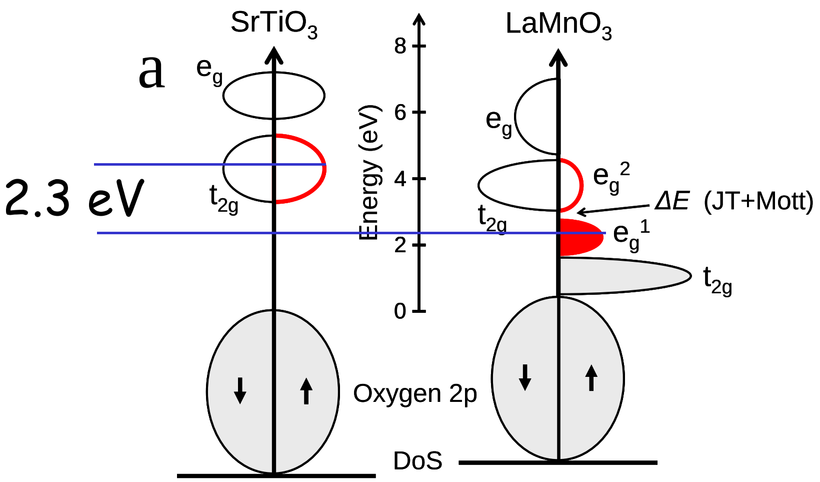
Spatially indirect or hybridized interface band transition at 2.4 eV (Mn→Ti)

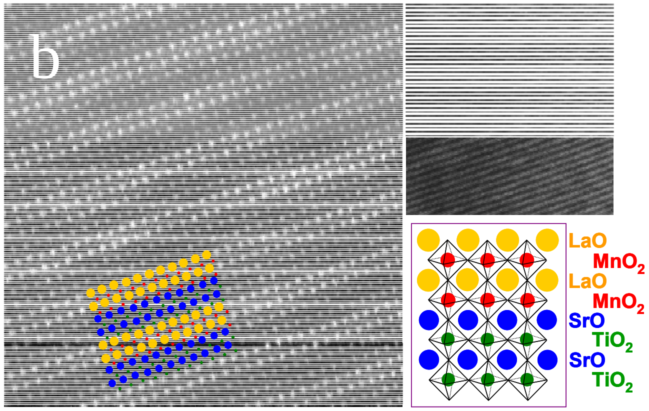



LMO STO 2x2 Superlattice
oxygen K-edge EELS
electron causes transition from 1s to hybridized 2P spectral weight (sensitive to electron state delocalization at pre-peak energy)

microscopy by Zuo and Shah

Band line-up and electronic structure

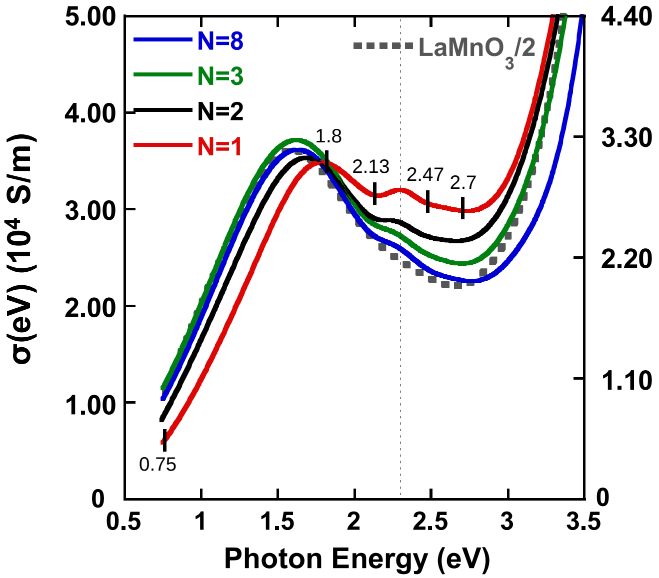




2.3 eV peak is Mn e_g^1 to Ti t_{2g}

for example, Mn d($3x^2$-r^2) to Ti d(xz)

oxygen bands nearly align



New optical absorption bands in digital superlattices

summary

- Short period digital superlattices of perovskite phases having different TMs can be accurately grown.

- New optical transitions can be created by placing molecular layers that electronically hybridize next to each other

- Electron transitions from source state to destination state
    - Source state is mainly from one molecular layer
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- Measure heterojunction band line-up

topic 3

Unusual quasiparticle DoS at cuprate/insulator interfaces (NIS tunnel junctions)

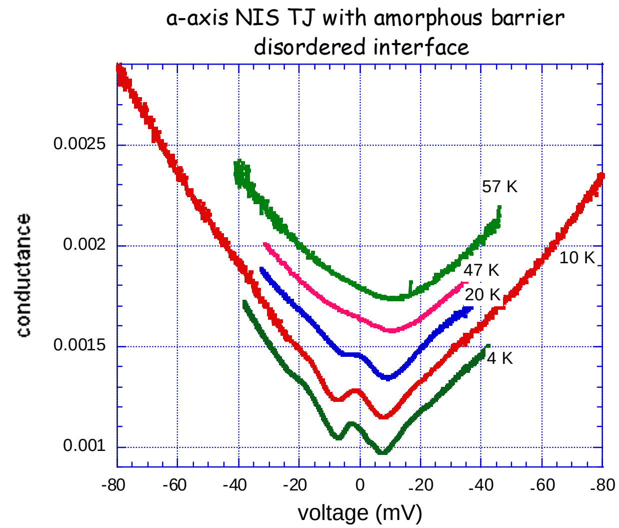
(not c-axis cuprate interfaces, rather they are ***a*-axis**)

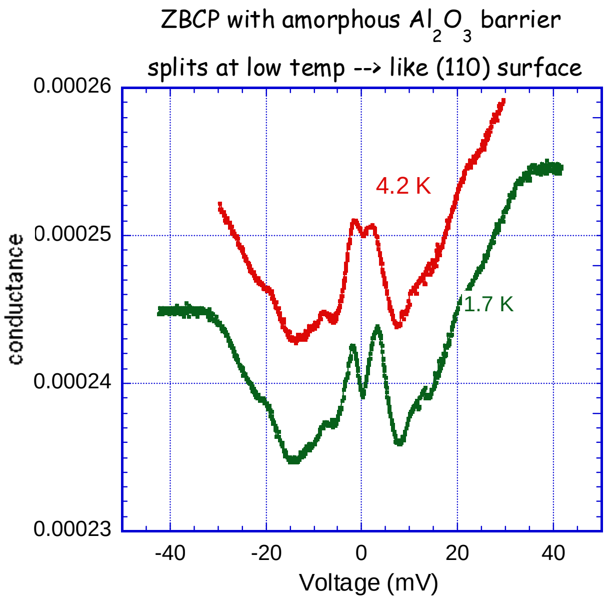
- If the interface causes a k-state to scatter to opposite signs of d_{x2-y2} then SC is quenched
  - At the surface a normal 2-D band is formed confined by the gap → the Andreev bound state
  - These electrons eventually pair in some other state and this is unique (expt. L.H. Greene group, theory J Sauls group)

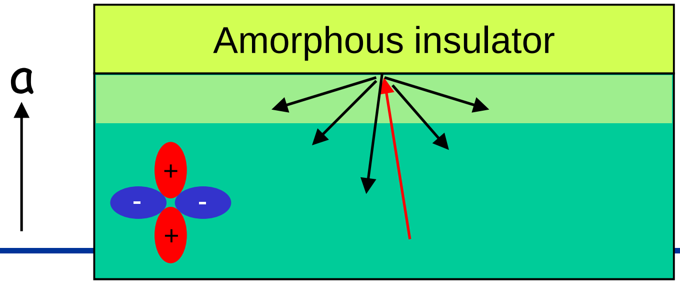
interface scattering at amorphous barriers!

Single crystal vs amorphous barriers

- non-specular reflection at **amorphous barriers**
- amorphous barrier devices show ZBCP DOS at surface similar to a+b direction devics: ZBCP which splits at lower temperatures **(LH Greene)**
- $G(V,T)/G(V,T_c)$ is particle/hole symmetric here →
- everything that follows now depends on **single crystal** heterostructure


a-axis NIS TJ with amorphous barrier
disordered interface


ZBCP with amorphous Al_2O_3 barrier
splits at low temp --> like (110) surface



Physica C **335**, 184 (2000)

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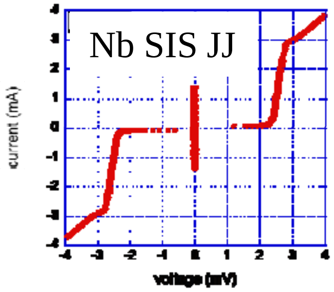
  - At the surface a normal 2-D band is formed confined by the gap → the Andreev bound state
  - These electrons eventually pair in some other state and this is unique (expt. L.H. Greene group, theory J Sauls group)
- If the interface is specular, and reflections don't mix + with -, then we find that the superconductivity at the surface doesn't have particle-hole symmetry (violates the core of BCS)

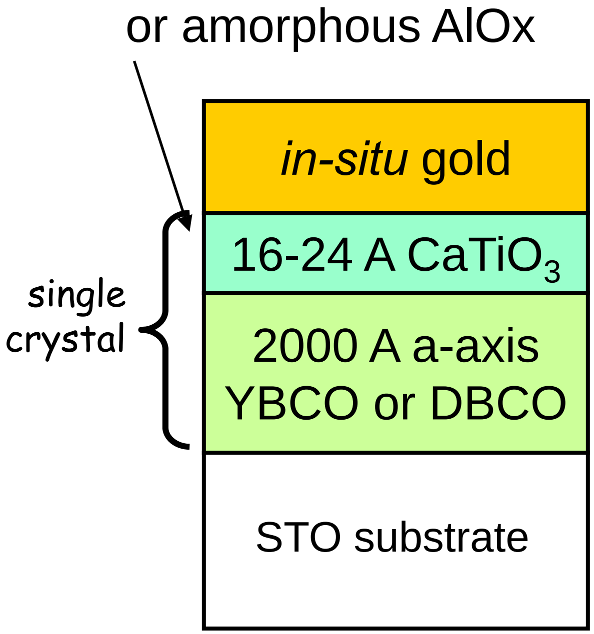
new electronic structure emerges because of the specular interface

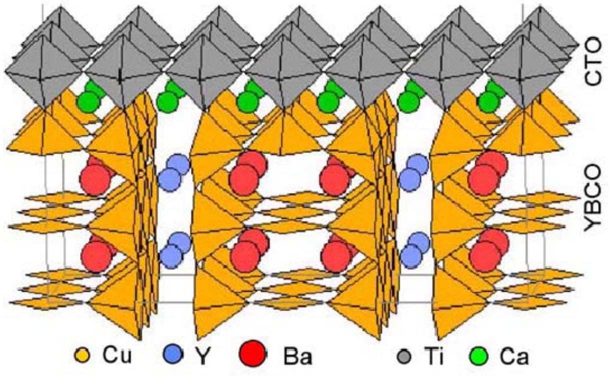


- In bulk, approximate e-h symmetry (BCS)
  - c.f. Renner, Davis, Yazdani, others (STM of 2212)
- At specular (100) surface we find this is really broken!
- Is it a superconducting state at interface?
  - It only appears below Tc
- How deep does it extend?
- Is it useful for engineering new electronic ground states that may have interesting properties?
- No theory yet.

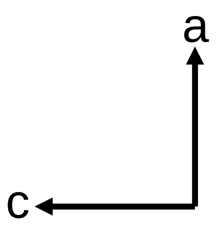
N-I-S tunnel junction fabrication

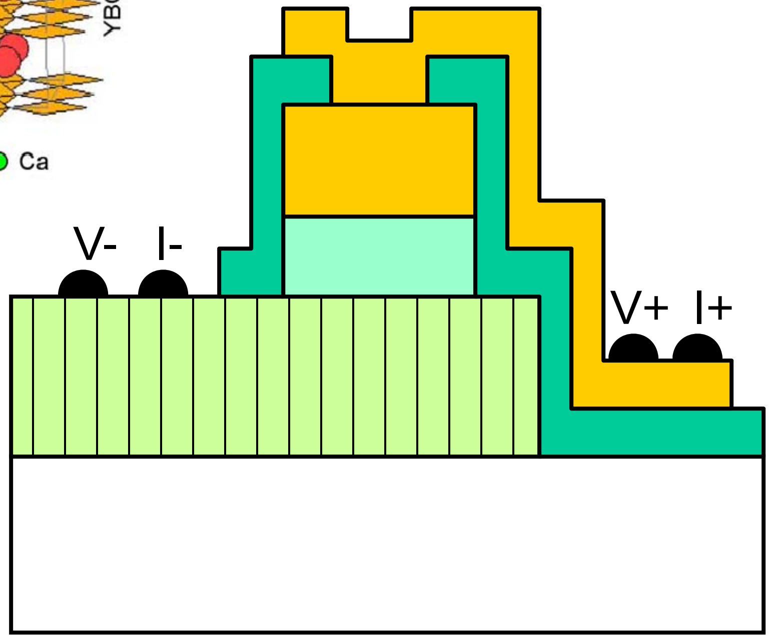
Trilayer film growth

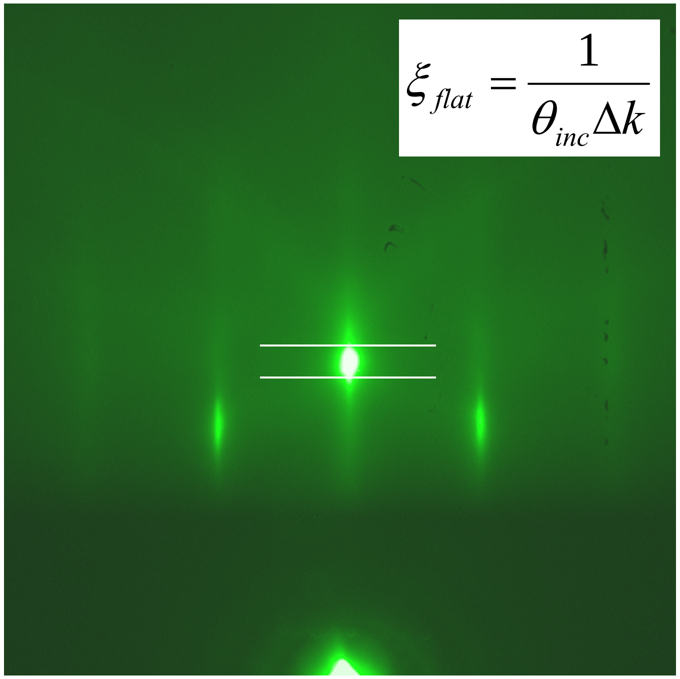




Junction process

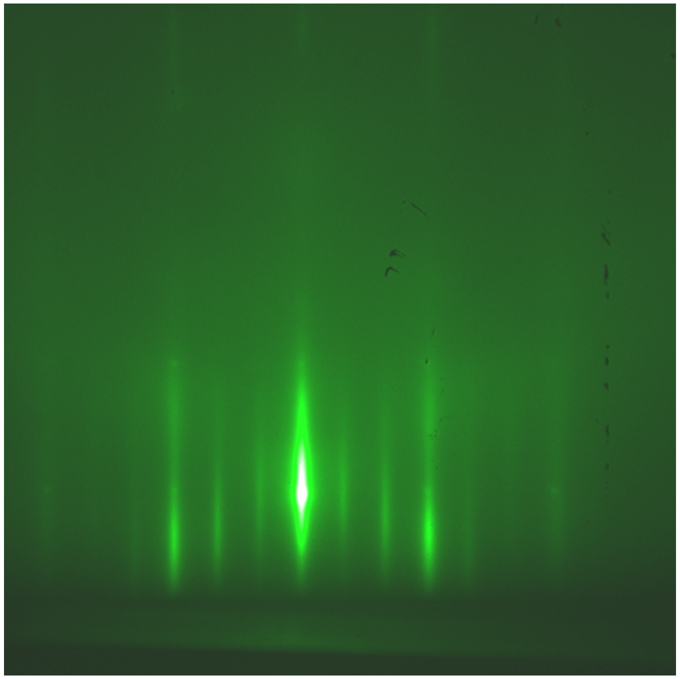






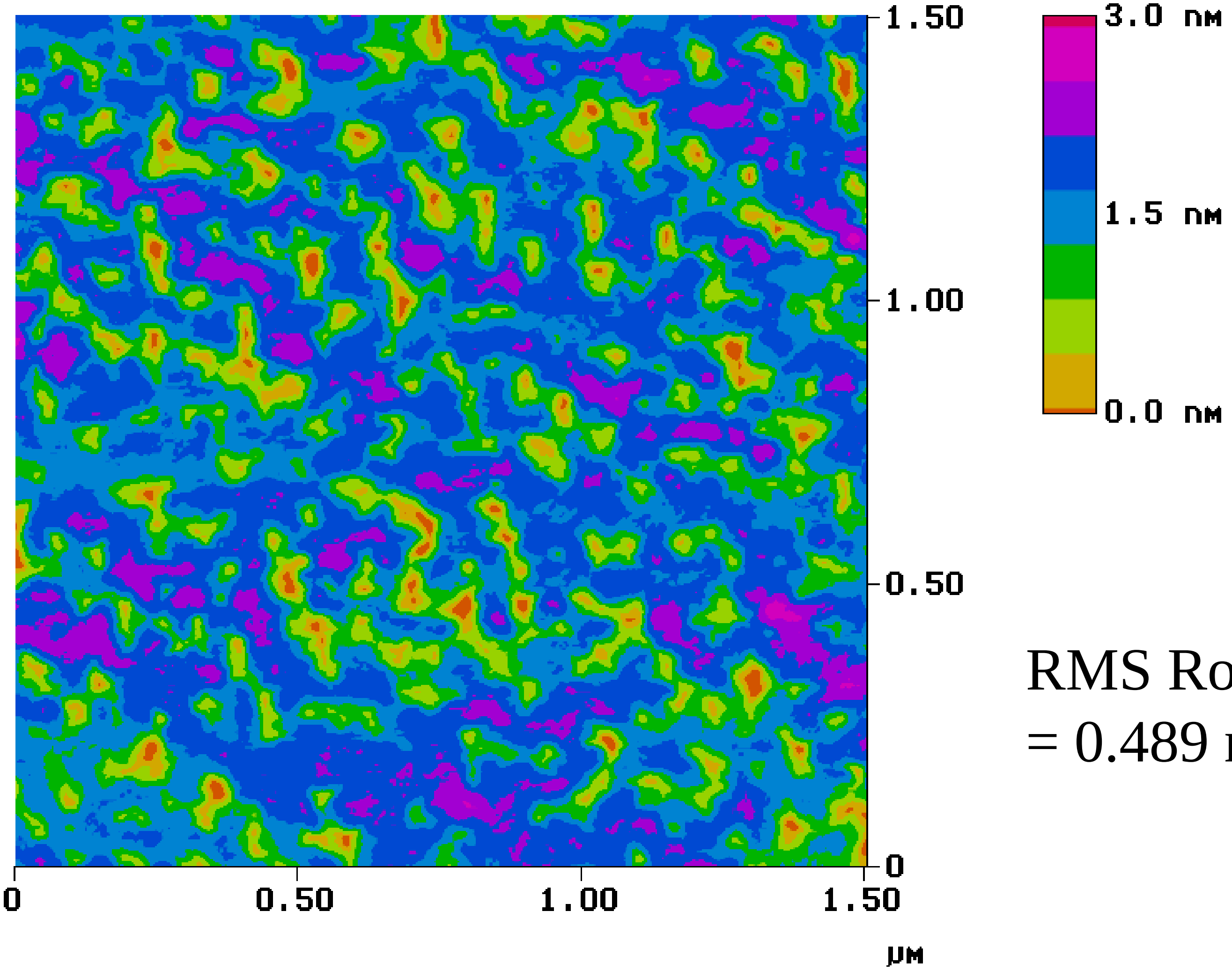
$$\xi_{flat} = \frac{1}{\theta_{inc}\Delta k}$$

Rheed image of substrate before growth



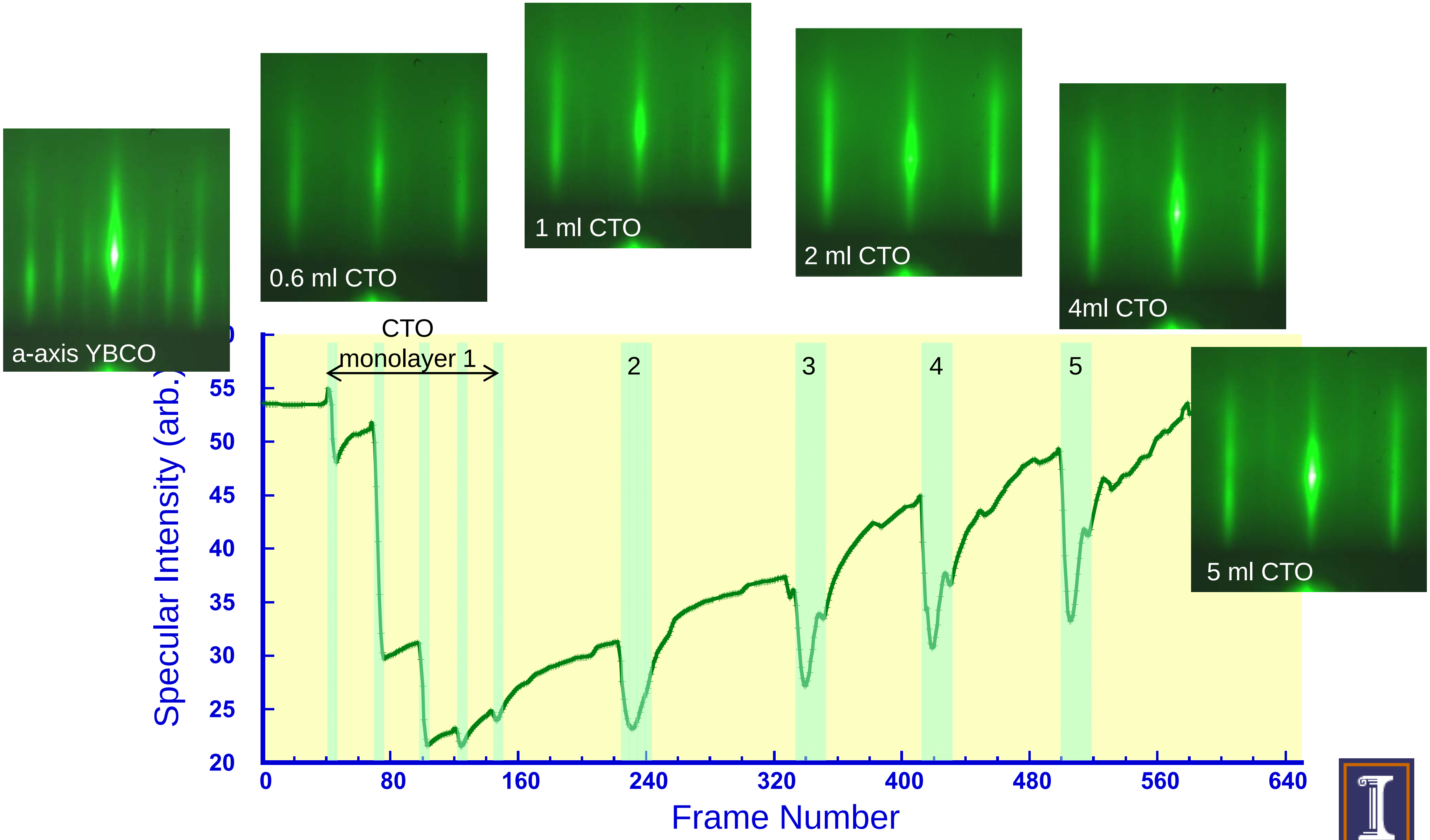
Rheed image of YBCO after growth of 2000 Angstroms

AFM Image of *a*-axis-YBCO



RMS Roughness
= 0.489 nm

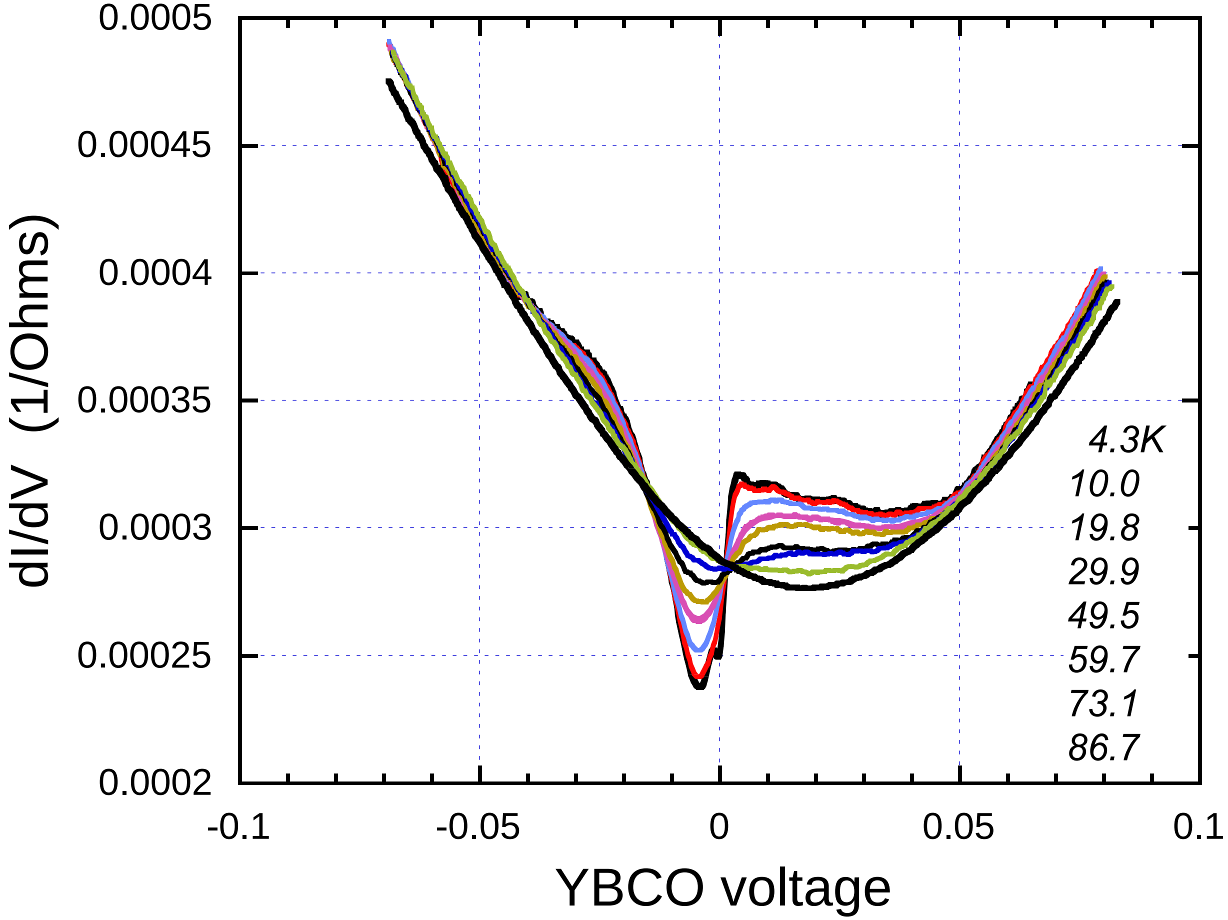
NIS tunnel junction
RHEED oscillations during crystalline barrier heteroepitaxy



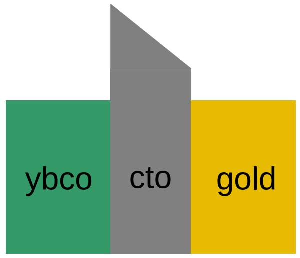
spectroscopy of electronic structure

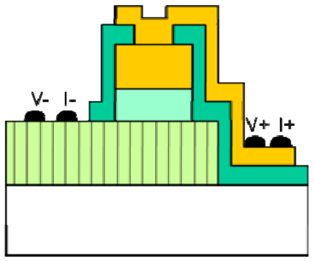
broken particle-hole symmetry

87K a-axis YBCO NIS tunnel junction



Offset parabola due to different band line up.

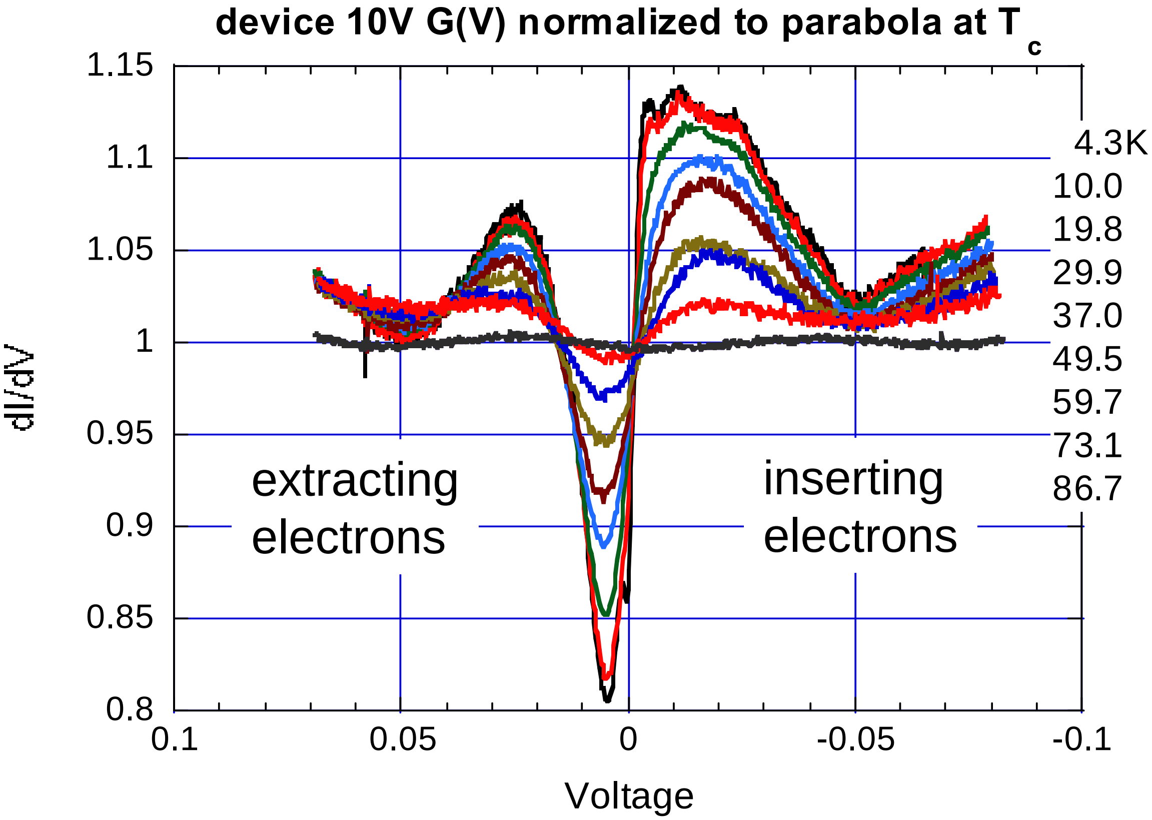




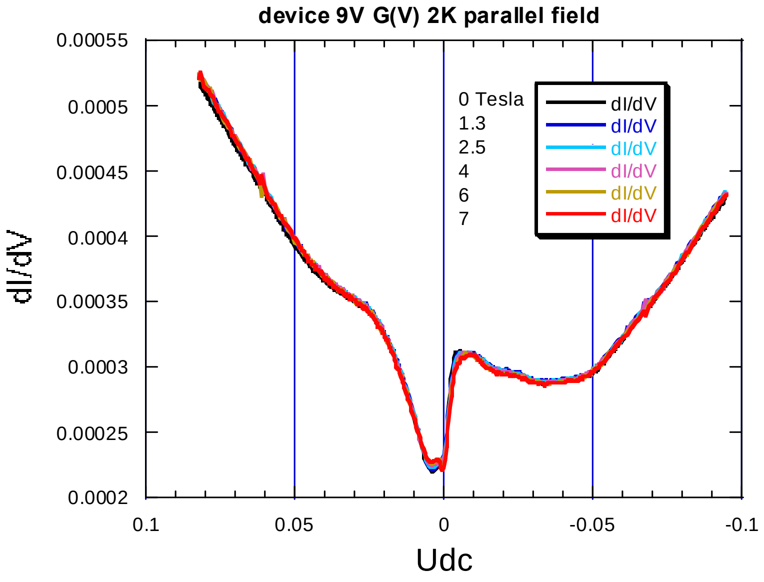
Phys Rev Lett **93**, 107004 (2004)

Normalize data to parabola above T_c

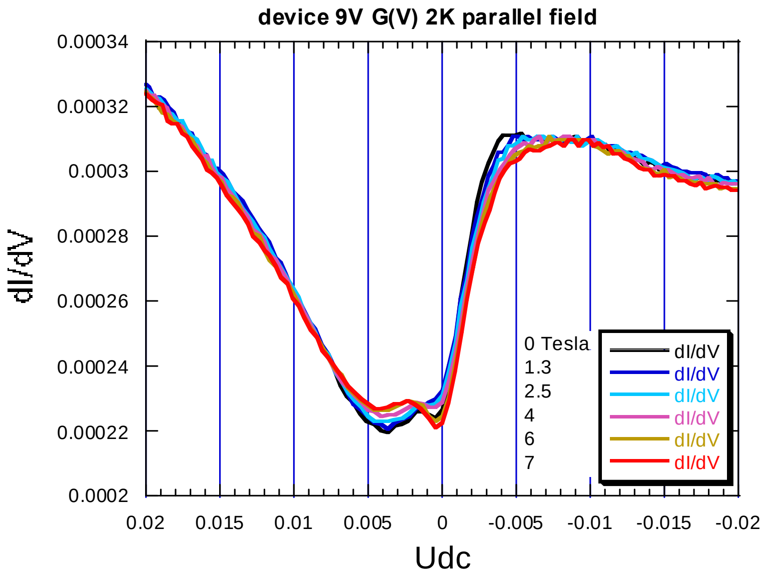
T_c=87K



Magnetic field parallel to junction

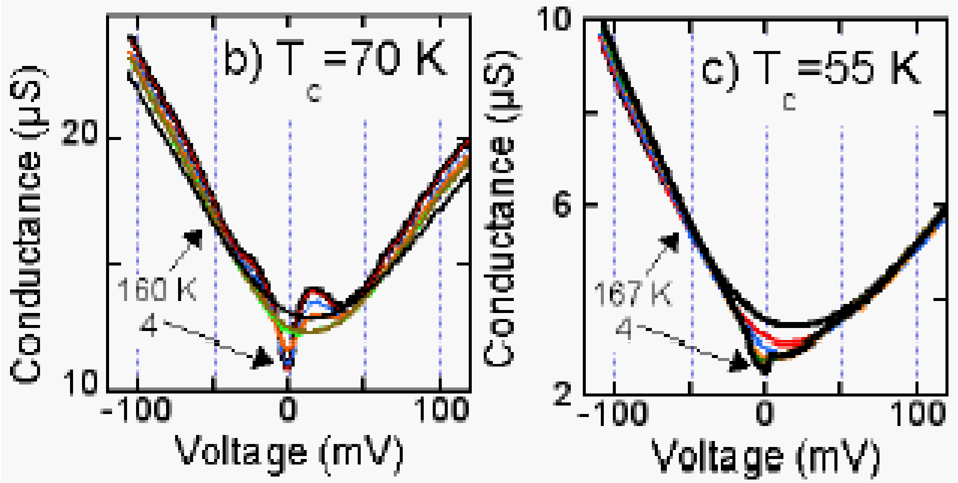


device 9V G(V) 2K parallel field



device 9V G(V) 2K parallel field

Similar results in underdoped cuprate



new electronic structure emerges because of the specular interface

- In bulk, approximate e-h symmetry
- At specular 100 surface this is really broken
  - For all carrier dopings
- Anti-symmetry tied to the Fermi energy
  - Not due to a single particle cause (Schottky barrier)
  - Due to interacting electrons
- How deep does it extend?
- Is it useful for engineering new electronic ground states that may have better properties?
  - SC is modified, can this be used?
- No theory yet (not that I know of).

Summary

Oxide interfaces and superlattices (3 topics)

What are the new results?

• global symmetry can be changed	Warusawithana
• design SL to obtain new optical transitions, $\sigma(\omega)$	Zhai
• emergence of modified collective order	Davidson

Global symmetry:

Make a digital superlattice out of 3 dielectric phases and use SL architecture that breaks inversion symmetry

→ dielectric has permanent polarization and finite $\chi^{(2)}$.

New electronic transitions

Digital superlattice to mix bands at interface

→ new optical transitions between composite states.

Modified superconductivity at interface (unexpected)

a-axis interface between YBCO and CaTiO3 (NIS tunnel jct)

→ crystalline interface causes DoS measured by tunneling to have broken e-h symmetry at interface (not like BCS)