Closing the circle: d-wave order parameter symmetry in the electron-doped $Sr_{1-x}La_xCuO_2$ superconductors

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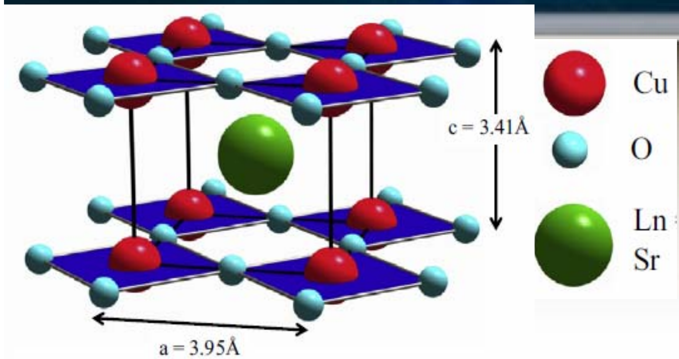
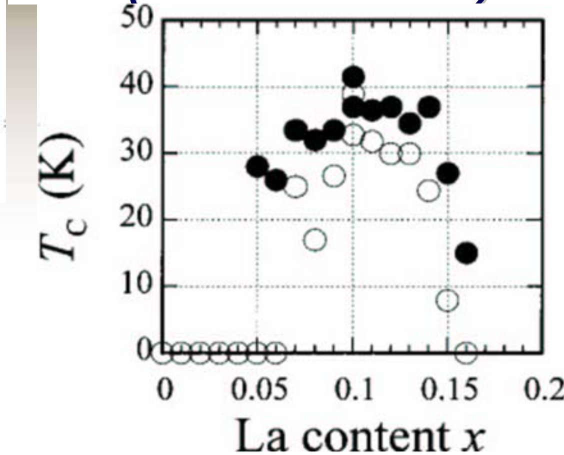
The $Sr_{1-x}La_xCuO_2$ (SLCO) compounds

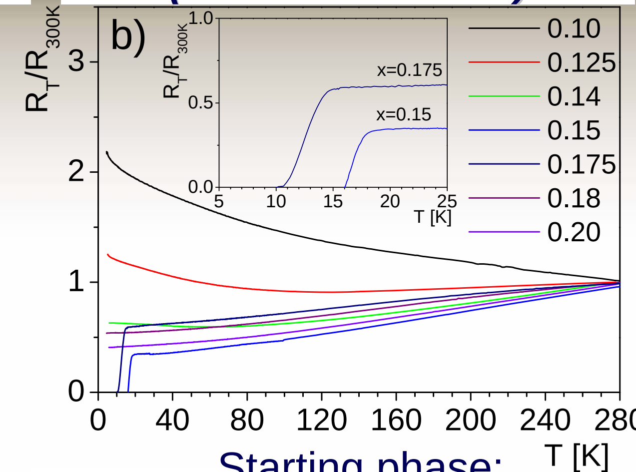


$Sr_{1-x}La_xCuO_2$
infinite-layer type structure
$T_{c,max}$ = 43 K

MBE growth on (001) $KTaO_3$ (reduction method)

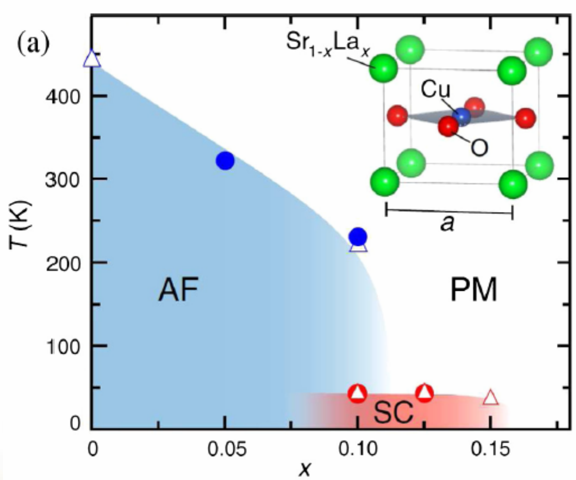


PLD growth on (001) $KTaO_3$ (oxidation method)

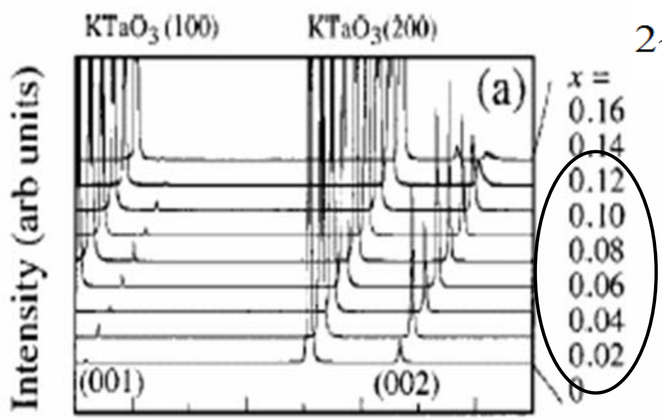


Starting phase:

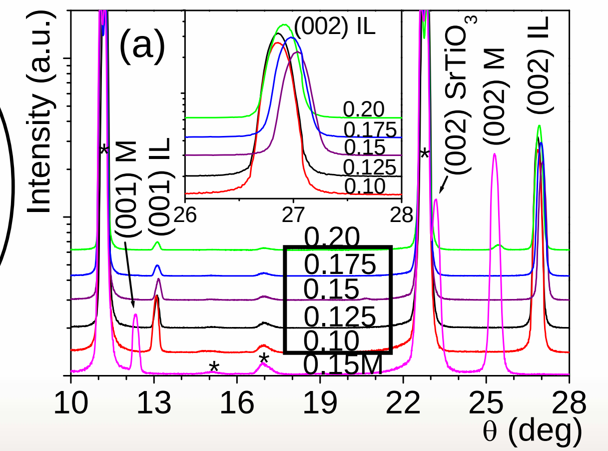
$$2\sqrt{2}a_p \times 2\sqrt{2}a_p \times c \text{ (super)structure}$$



T_c vs. dopant concentration phase diagram for bulk $Sr_{1-x}La_xCuO_2$

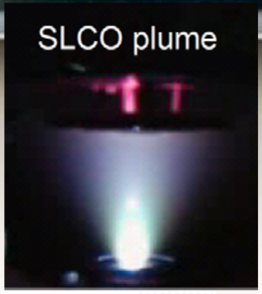


Karimoto et al. (APL 79, 2001)

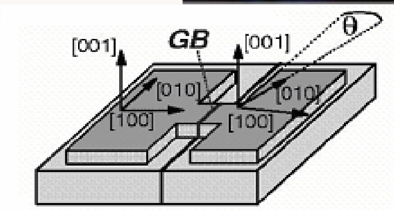


Leca et al. (APL 89, 2006)

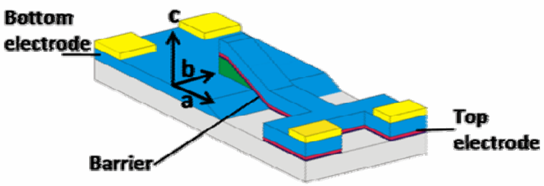
Outline



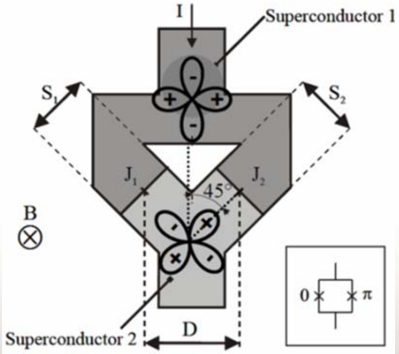
1. *Correlation between strain, microstructure, and electrical transport properties in $Sr_{1-x}La_xCuO_2$ (SLCO, x=0.10-0.175) **thin films** grown by PLD on different substrates ($KTaO_3$, $SrTiO_3$, and $DyScO_3$)*



2. *$Sr_{1-x}La_xCuO_2$ (x=0.125-0.15)* ***Josephson junctions***:
   - *grain-boundary junctions* based on c-axis oriented films
   - *ramp-type* SLCO/Au/Nb junctions



3. ***Order parameter symmetry*** – phase sensitive tunneling experiments based on 0/π-SQUIDs (on $SrTiO_3$ tetracrystals)

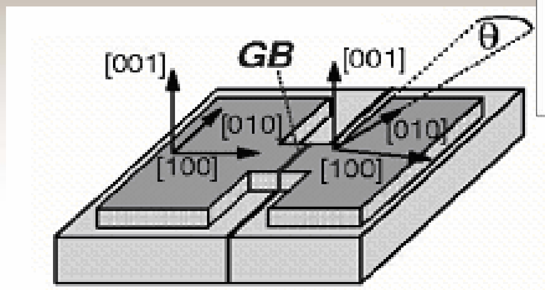


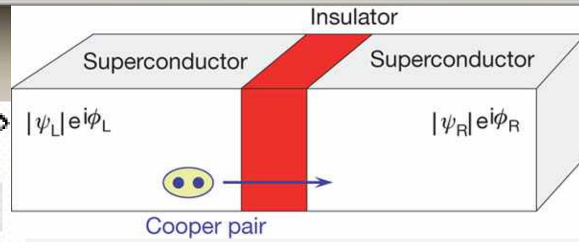
Is d-wave symmetry inherent to cuprate superconductivity?

Thin films: growth and properties

Requirements for $Sr_{1-x}La_xCuO_2$-based junctions

Grain-boundary

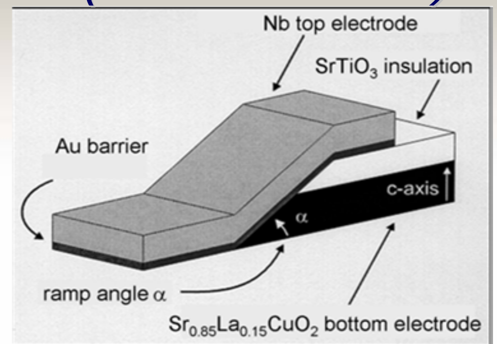


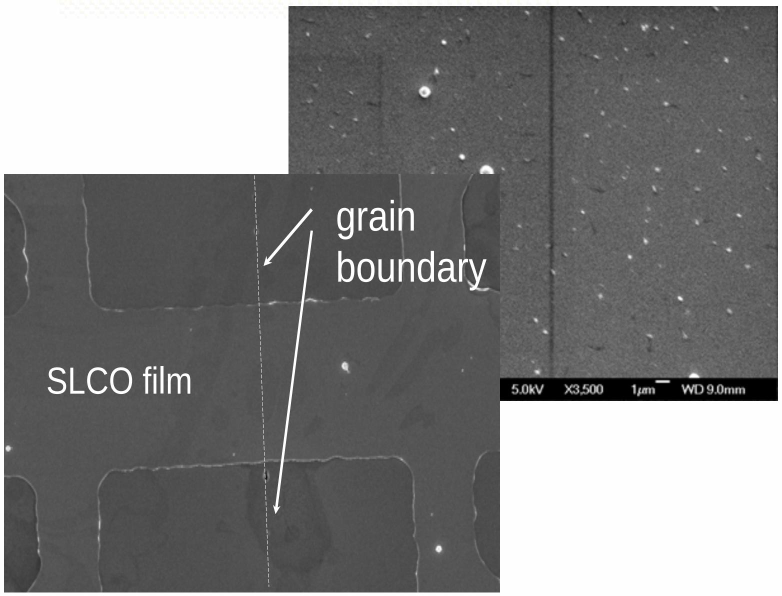


Josephson junction

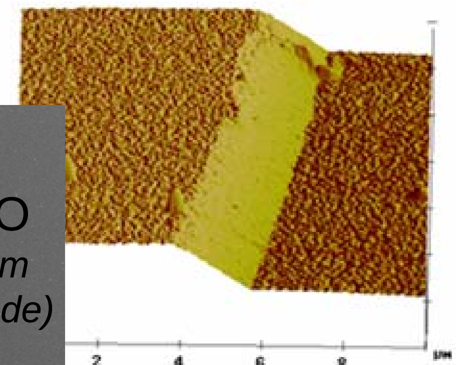
Ramp-type

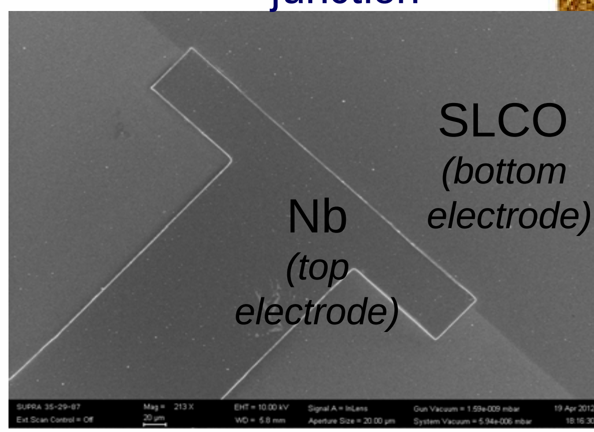
(SrLaCuO/Au/Nb)





AFM of a ramp-type junction





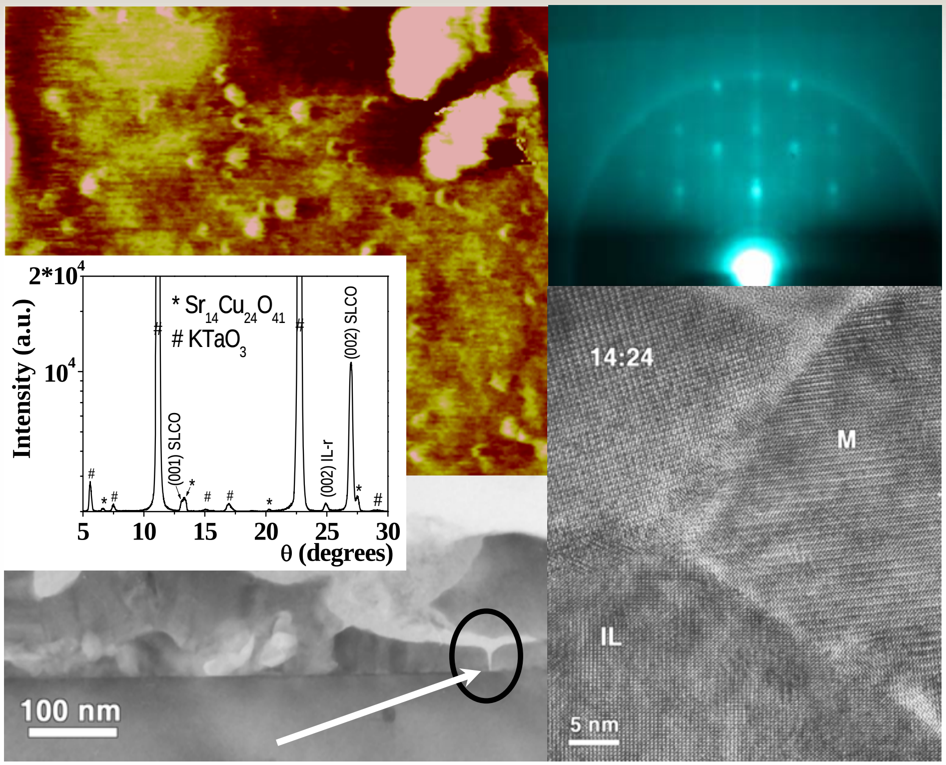
SEM of a 200 μm wide junction

Single phase, c-axis oriented films required

Single phase, c-axis oriented SLCO films, with smooth surface and sharp oxide-metal interface required

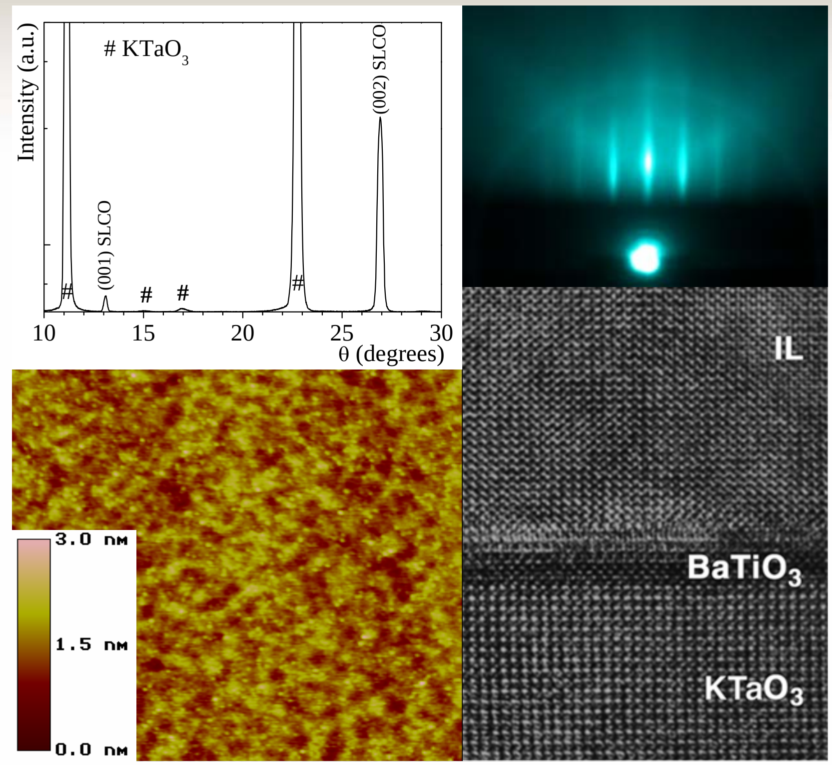
Role of the substrate-film interface (oxidation method)

$Sr_{1-x}La_xCuO_2$ grown on as-received (001) $KTaO_3$



XRD, HRTEM: Structural defects/secondary phases developing at the substrate-film interface
RHEED, AFM: 3D growth mode induced by the high interface roughness

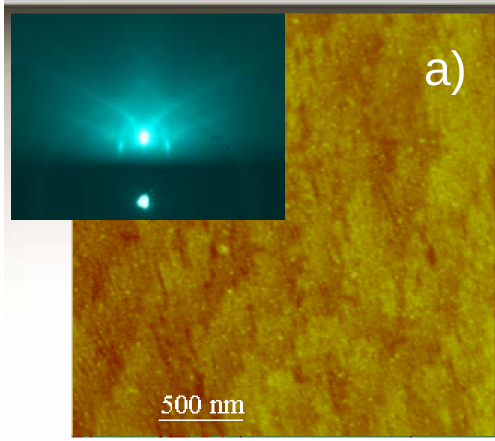
$Sr_{1-x}La_xCuO_2$ grown on $BaTiO_3$-buffered (001) $KTaO_3$

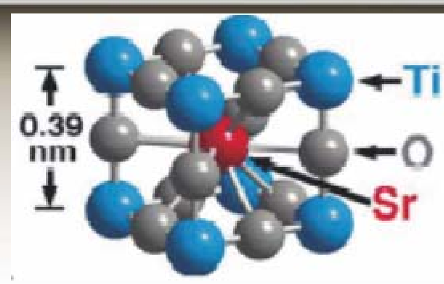


XRD, HRTEM, AFM: single-phase, reduced density of structural defects, improved morphology
RHEED, AFM: 2D growth mode

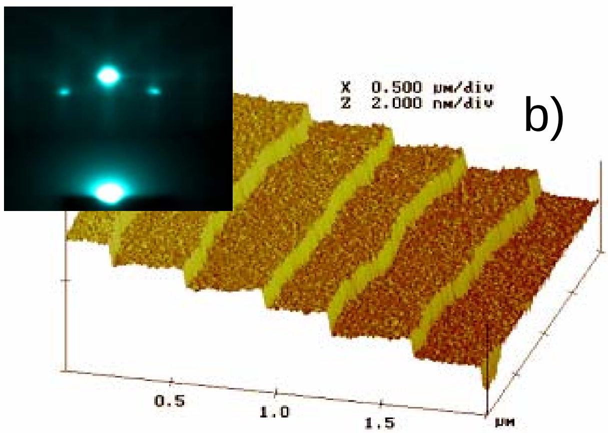
Leca, PhD thesis (2003), Leca et al. APL 89 (2006)

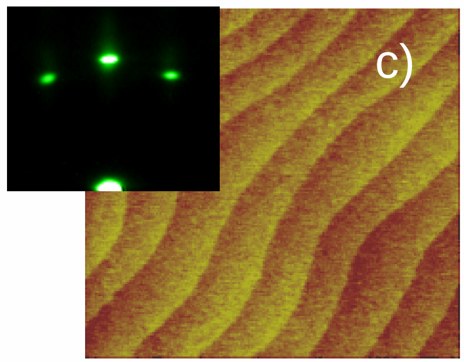
Improved substrate morphology – $SrTiO_3$ and $KTaO_3$





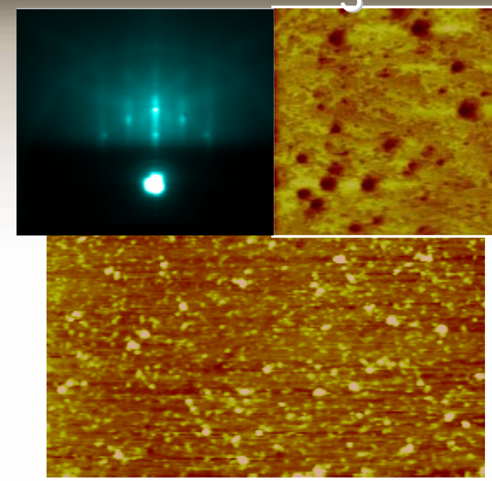
$SrTiO_3$ structure



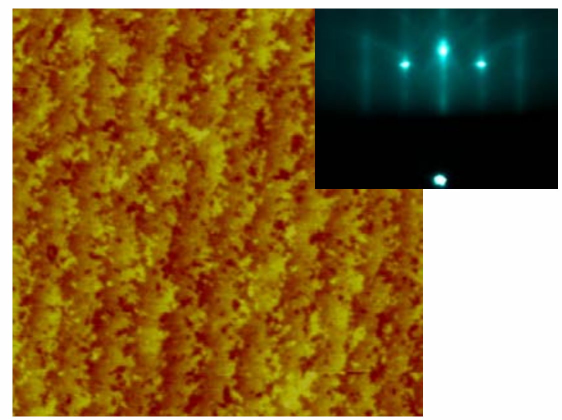


a) as-received substrate; b) after chemical treatment (BHF) and annealing (950ºC/1h/O_2) - TiO_2 termination
c) after annealing (950ºC/1h/O_2) - SrO termination

$KTaO_3$



as-received substrate



after annealing at 750ºC/O_2/1h

Kawasaki et al. Science 226 (1994); Koster et al. APL 73 (1998); Leca, PhD thesis (2003)

$Sr_{1-x}La_xCuO_2$ ($x \approx 0.15$) growth parameters

On (001) $KTaO_3$ and (110) $DyScO_3$

$Sr_{1-x}La_xCuO_2$ (SLCO):

T_d = 500-575 °C
P_d = 0.20-0.40 mbar O_2
Post-deposition vacuum annealing (reduction) at T_d and 10^{-7} mbar:
- 10-20 min ($KTaO_3$ case)
- 20-30 min ($DyScO_3$ case)

T_c: up to 24 K, on $KTaO_3$
up to 18 K, on $DyScO_3$

Film's composition (x_t=0.15 target), from RBS data:
- x_f = 0.145, film grown on $KTaO_3$
- x_f = 0.135, film grown on $DyScO_3$

a_{KTO}=3.988 Å, a_{SLCO}~3.978 Å
$a_{DSO(110)}$=3.944 Å, a_{SLCO}~3.950 Å

On $BaTiO_3$- buffered (001) $SrTiO_3$

$BaTiO_3$ (BTO):

T_d = 700-850 °C
P_d = 0.10 mbar O_2
Post-deposition annealing:
-15 min @ 950°C, under 0.10 mbar O_2, and 30 min @ 950°C, in vacuum (10^{-7} mbar)

$Sr_{1-x}La_xCuO_2$ (SLCO):

T_d = 550-600 °C
P_d = 0.20 mbar O_2
Post-deposition annealing (reduction):
45-60 min @ 550 °C, in vacuum (10^{-7} mbar)

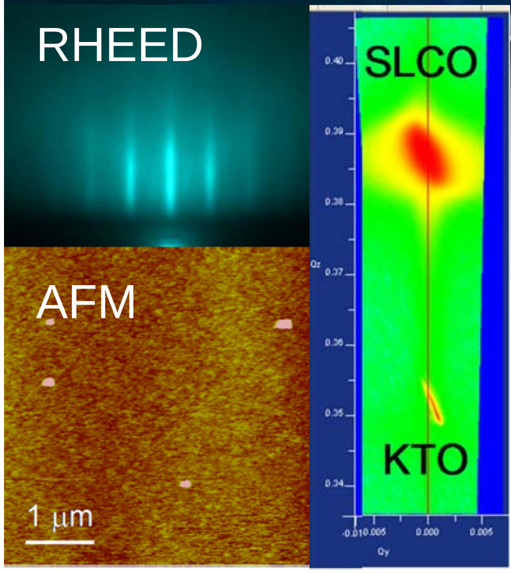
T_c: up to 21 K

Film's composition (x_t = 0.15 target), from RBS data: x_f = 0.145

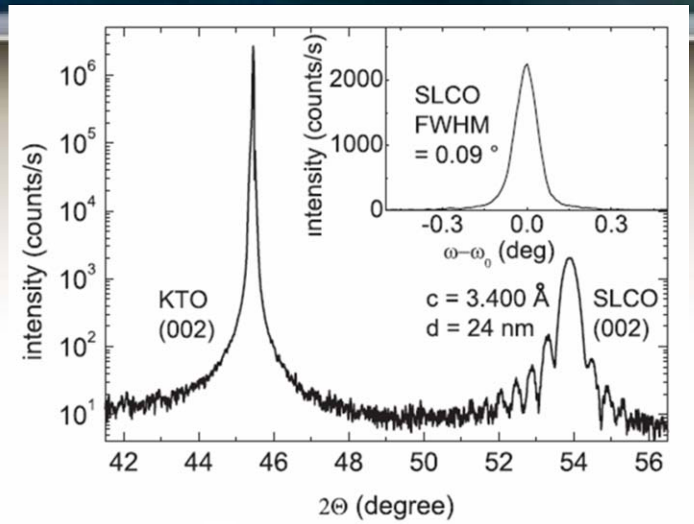
a_{STO}=3.905 Å, a_{BTO}~3.990 Å, a_{SLCO}~3.967 Å

Tomaschko, Leca, Selistrovski, Diebold, Jochum, Kleiner, Koelle, Phys. Rev. B 85 (2012)

$Sr_{1-x}La_xCuO_2$ (x=0.15) grown on $KTaO_3$ (001)

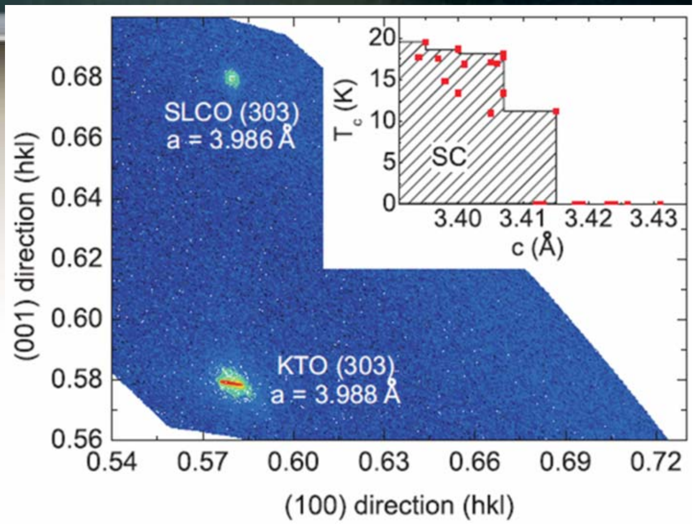


RHEED, AFM, and
XRD rsm of (101) plane



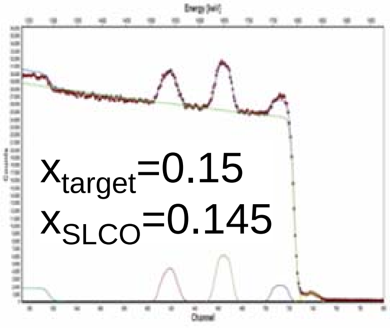
$2\theta/\omega$ XRD scan of (002) SLCO/KTO

a_{SLCO}~3.978-3.988 Å
c_{SLCO}~3.395-3.405 Å

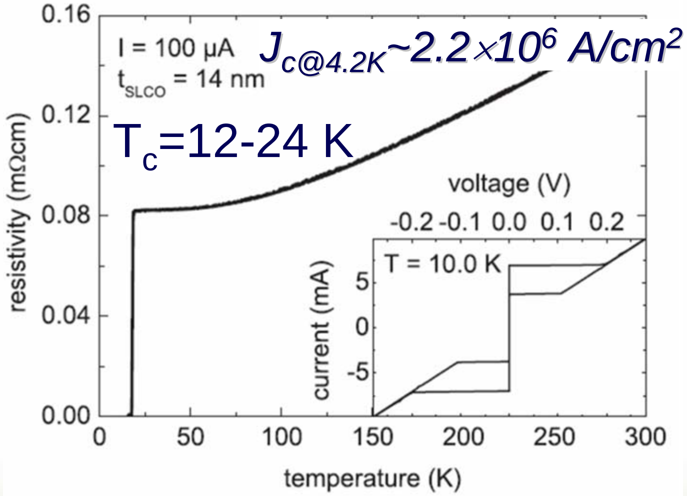


XRD reciprocal space map

Growth parameters:
$Sr_{0.85}La_{0.15}CuO_2$ (SLCO): T_d=575°C,
P_d=0.30 mbar O_2; E_d=2 J/cm^2;
20 min/550°C/10^{-7} mbar
films thickness: 15-50 nm.



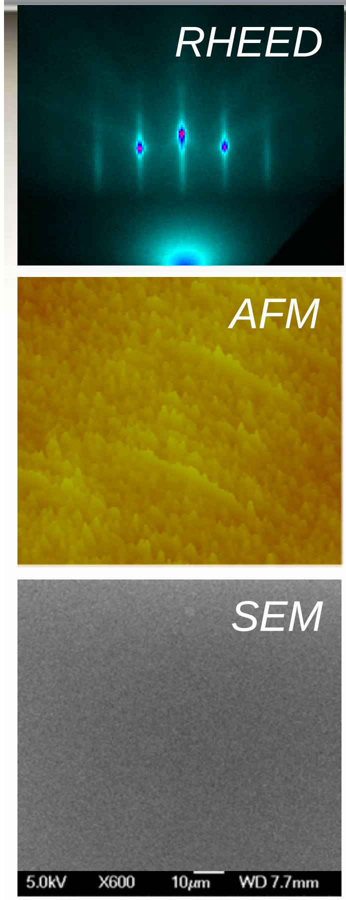
RBS



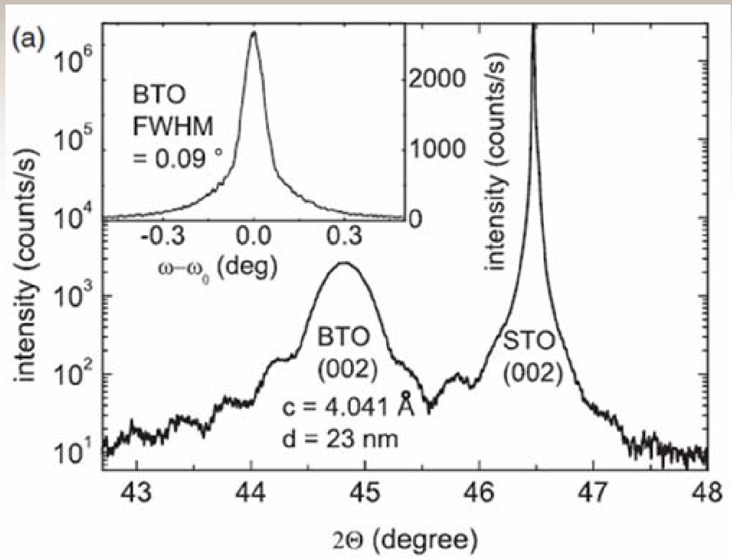
ρ vs. T

Tomaschko, Leca, Selistrovski, Diebold,
Jochum, Kleiner, Koelle, Physical Review B 85 (2012)

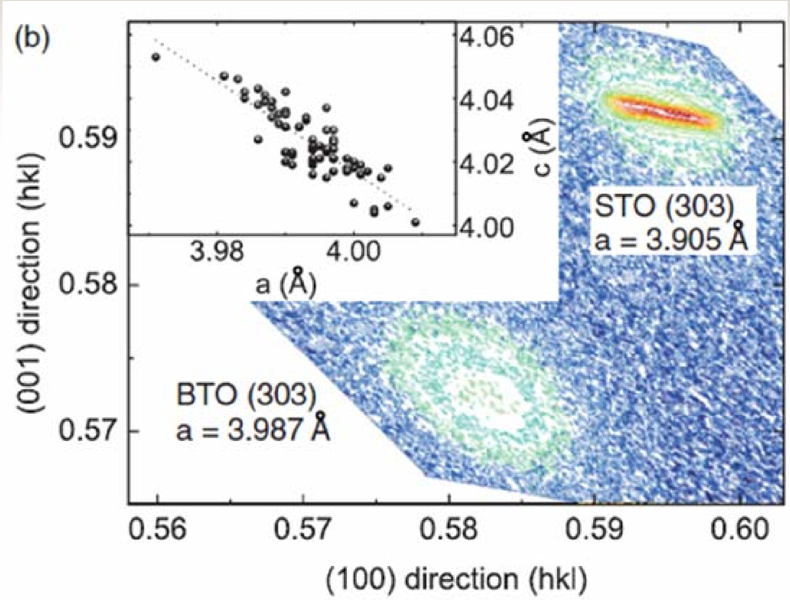
$Sr_{1-x}La_xCuO_2$ (x=0.15) grown on $BaTiO_3/SrTiO_3$ (001)



The $BaTiO_3$ buffer layer



2θ/ω XRD scan of (002) BTO/STO

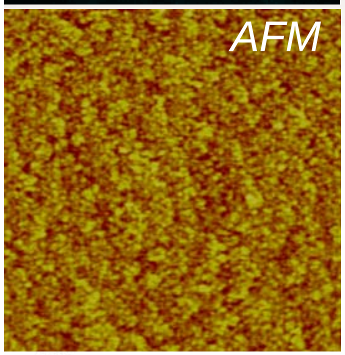


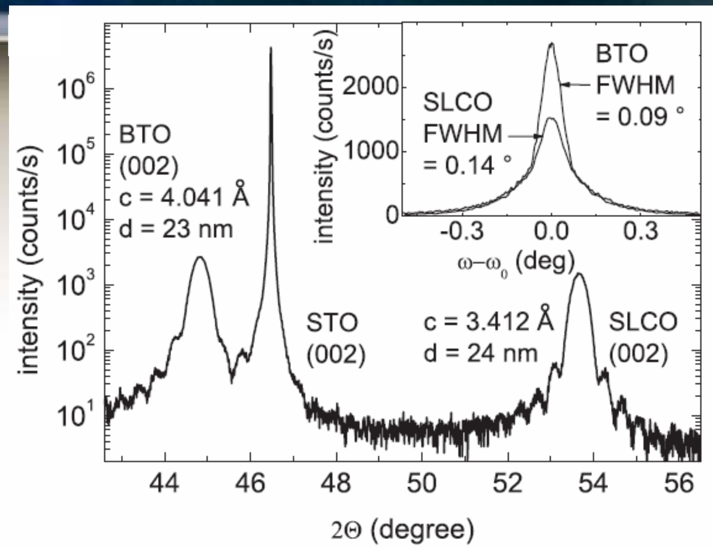
XRD rsm of (303) BTO/STO

Deposition conditions for $BaTiO_3$:
T_d=750°C, P_d=0.10 mbar O_2, E_d=1.75 J/cm²; 30 min/950°C/10^{-7} mbar

Tomaschko, Leca, Selistrovski, Diebold, Jochum, Kleiner, Koelle, Phys. Rev. B 85 (2012)

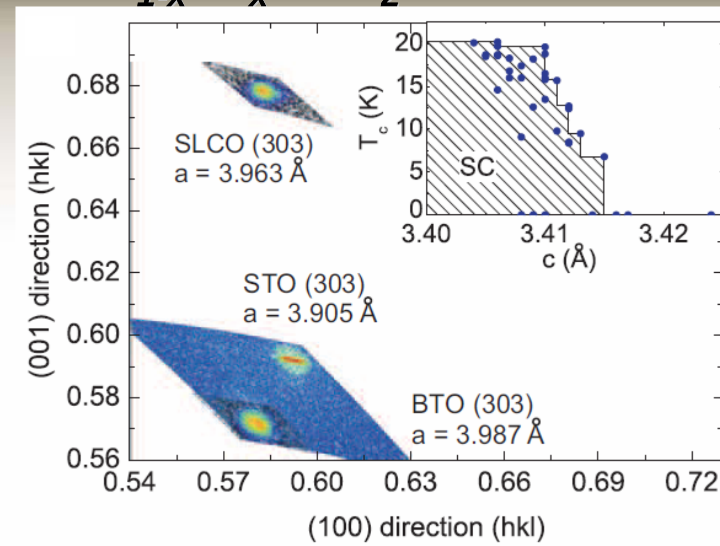
$Sr_{1-x}La_xCuO_2$ (x=0.15) grown on $BaTiO_3/SrTiO_3$ (001)



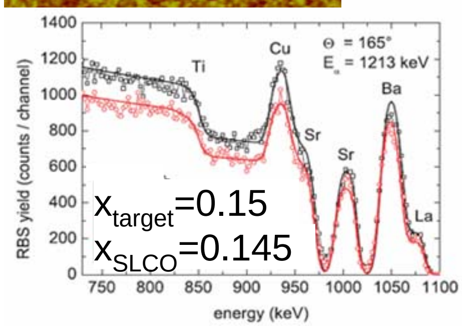


2θ/ω XRD scan of (002) planes

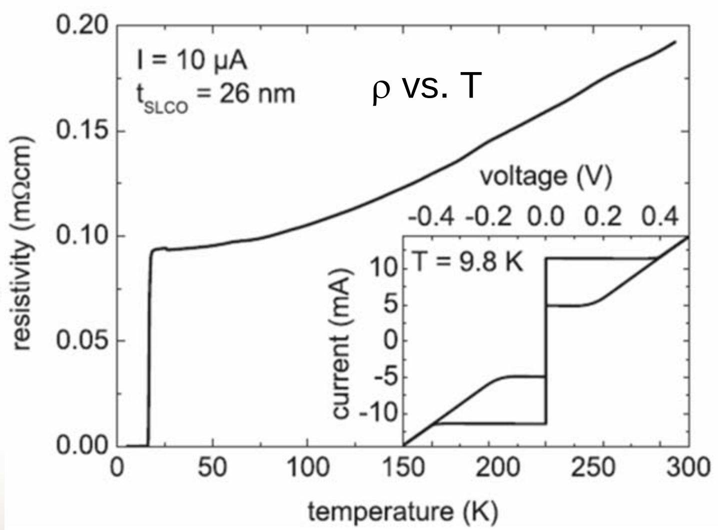
$Sr_{1-x}La_xCuO_2$ films



XRD rsm of (303) SLCO/BTO/STO



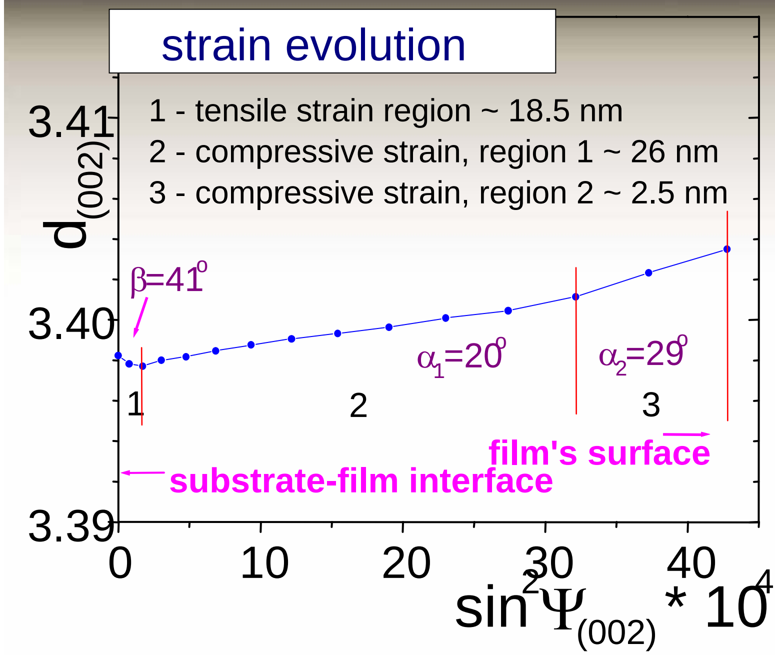
RBS

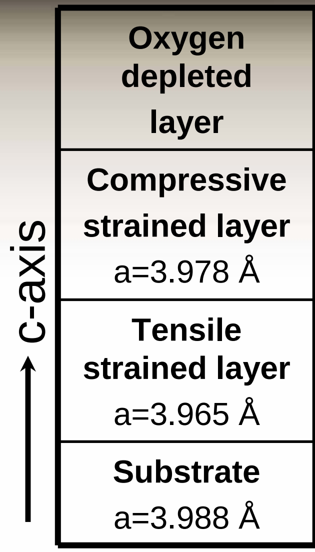


Deposition conditions:
T_d=550°C, 0.40 mbar O_2;
50 min/550°C/10^{-7} mbar
$T_{c,0}$=12-21 K
$J_{c@4.2K}$=2.1×10^6 A/cm^2

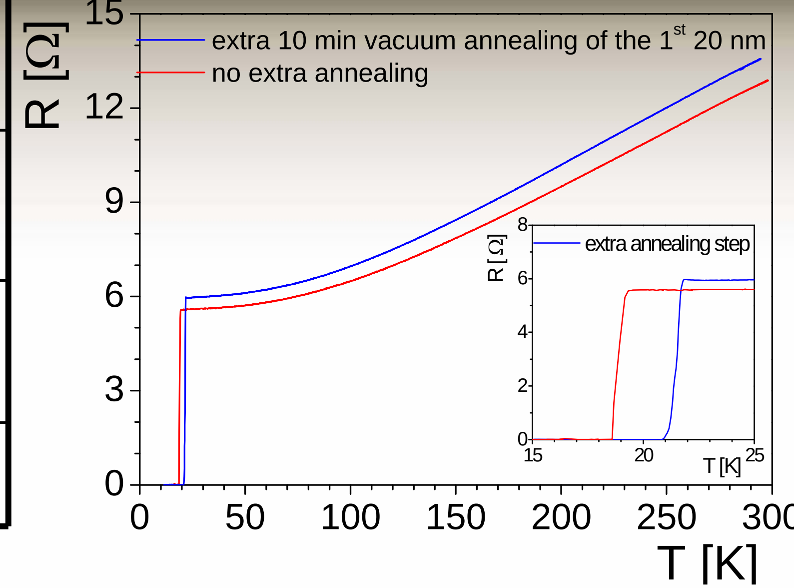
Tomaschko, Leca, Selistrovski, Diebold, Jochum, Kleiner, Koelle, Phys. Rev. B 85 (2012)

Role of the epitaxial strain





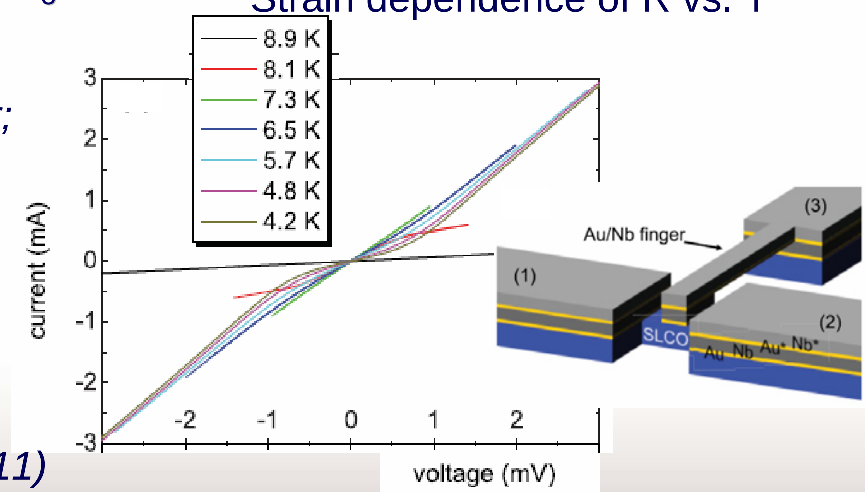
SLCO on $KTaO_3$



Strain dependence of R vs. T

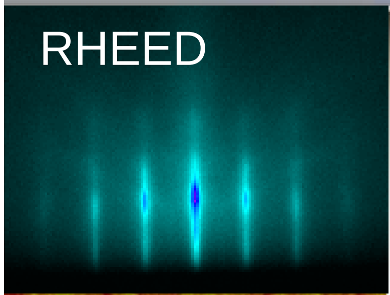
***XRD and XPS**: existence of an oxygen depleted top layer;*
- implications: no Copper pair tunneling in planar SLCO/Au/Nb junctions

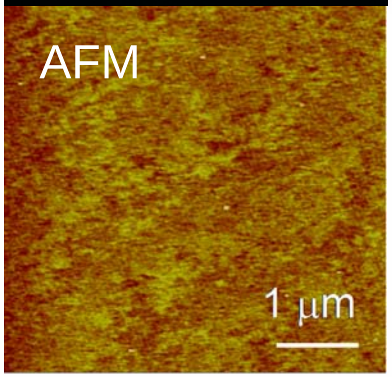
***XRD**: two types of strain: tensile (@ substrate-film interface) and compressive, on top*



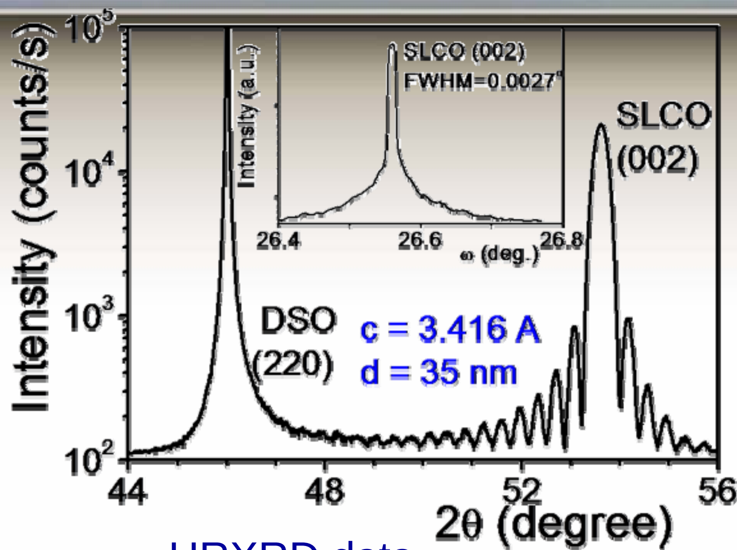
Tomaschko et al., PRB 84 (2011)

$Sr_{1-x}La_xCuO_2$ ($x \approx 0.15$) grown on $DyScO_3$ (110)

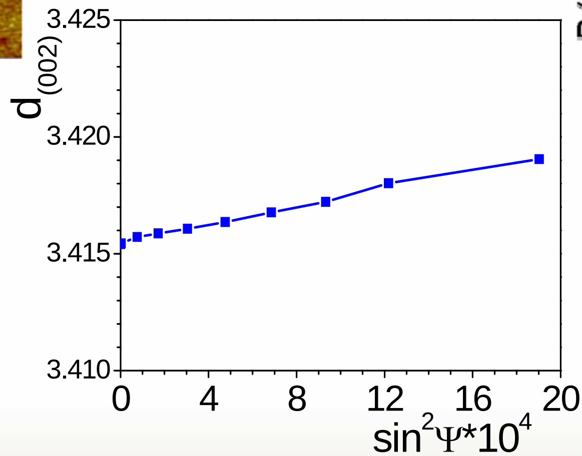




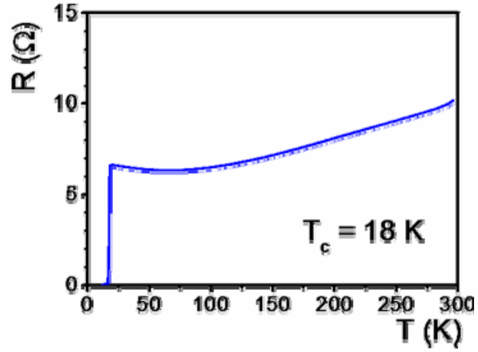
No interface tensile strain layer



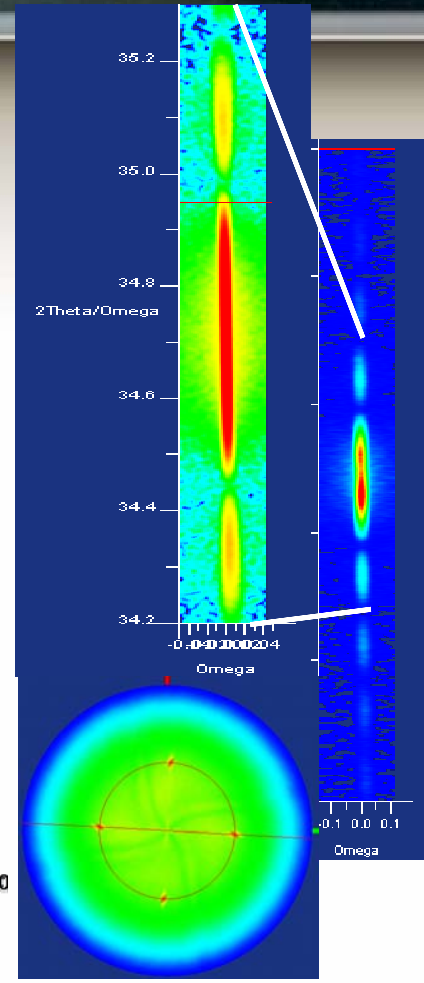
HRXRD data



Strain evolution:
in-plane compressive strain

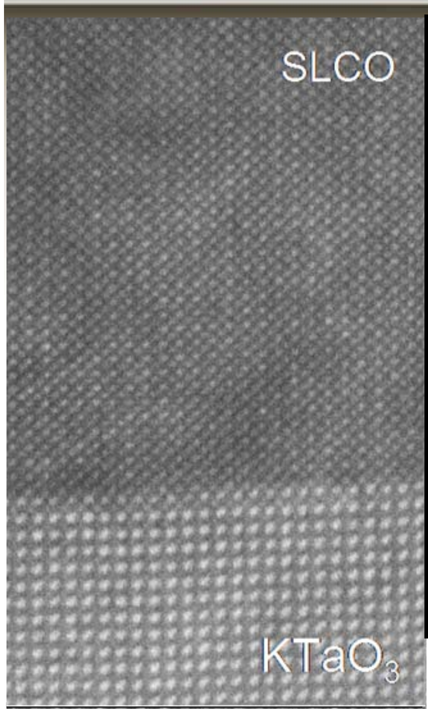


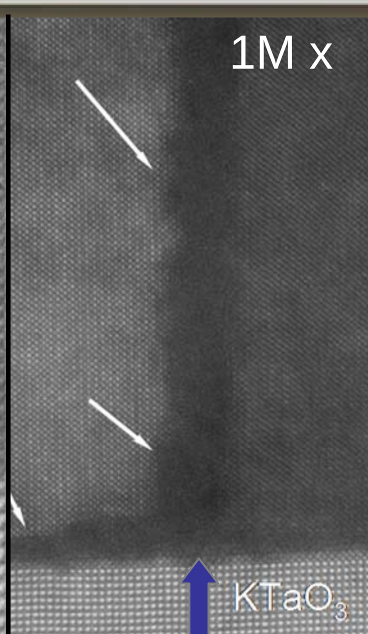
R vs. T evolution



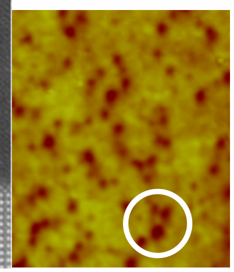
HRXRD rsm and pole figure
of the asymmetric (101) plane
Presence of Laue fringes
indicates high crystalinity

HRTEM results on $Sr_{1-x}La_xCuO_2$ ($x \approx 0.15$) film

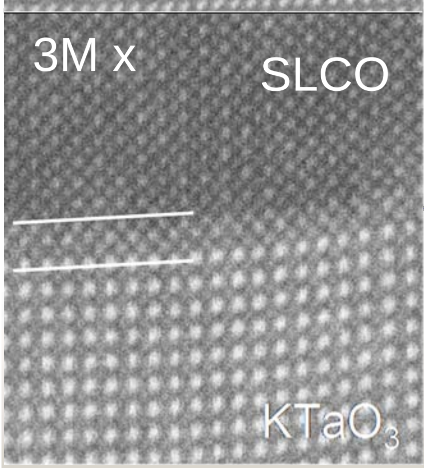




discontinuity of the SLCO film

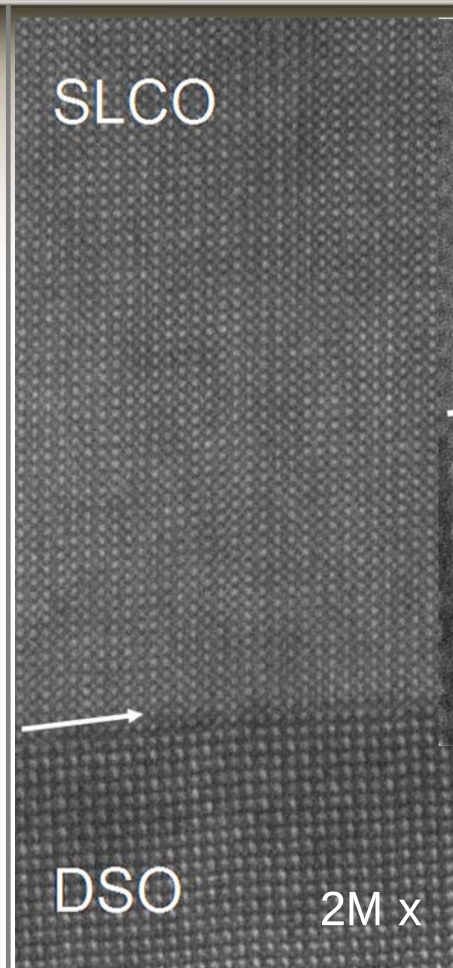


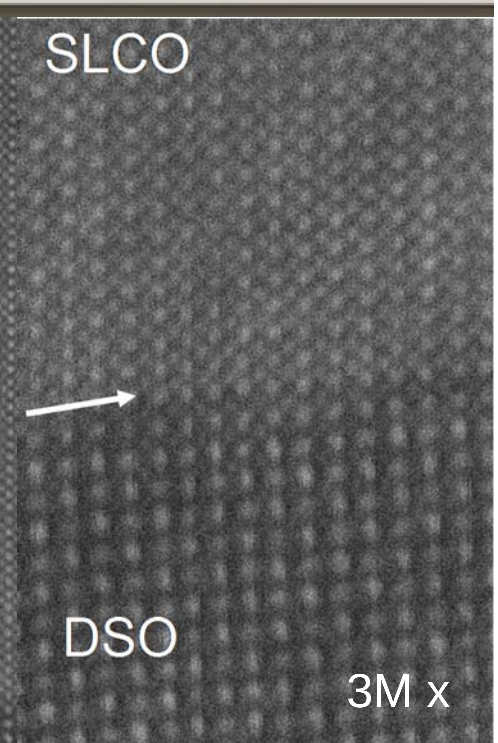
AFM of $KTaO_3$



Interface roughness

on (001) $KTaO_3$

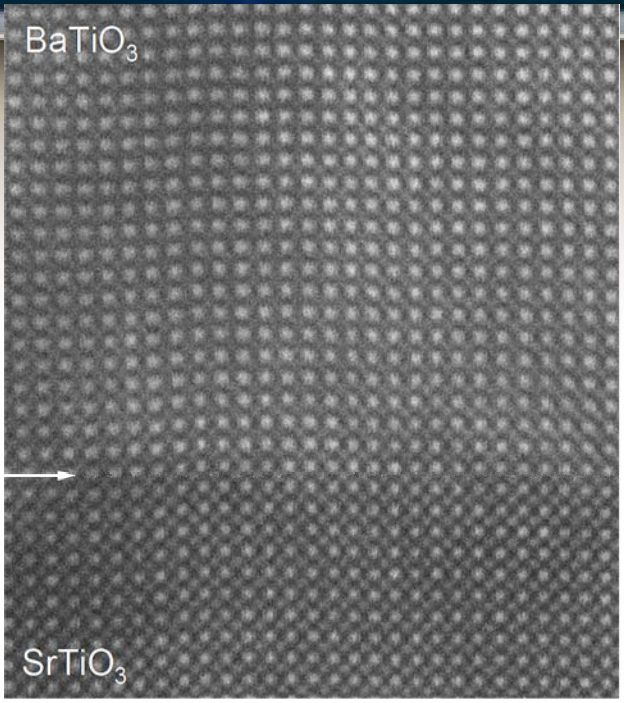




no defects;
sharp interface;
no interface strained layer

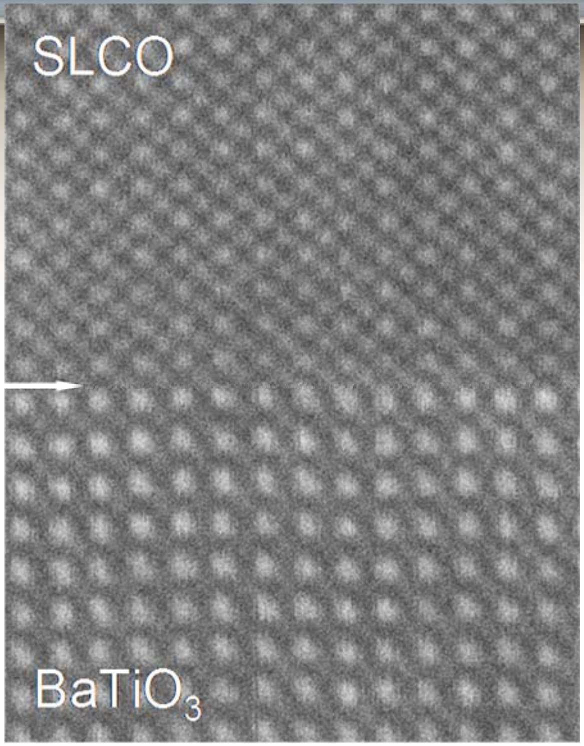
on (110) $DyScO_3$

HRTEM results on $Sr_{1-x}La_xCuO_2$ ($x \approx 0.15$) film

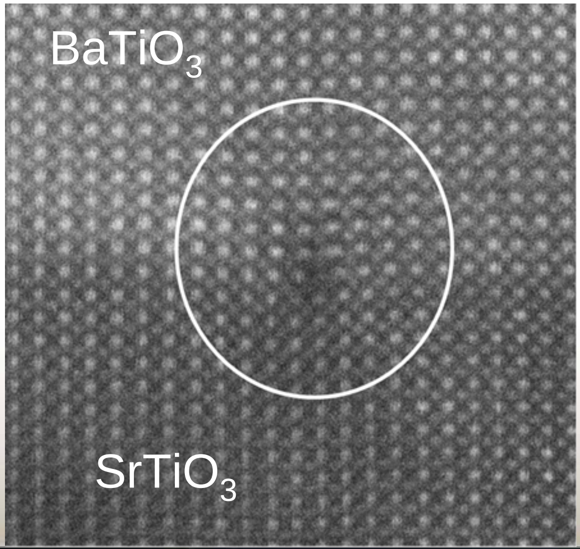


HRTEM

Sharp interfaces



Sharp BTO-SLCO interface



edge dislocation at the STO/BTO interface

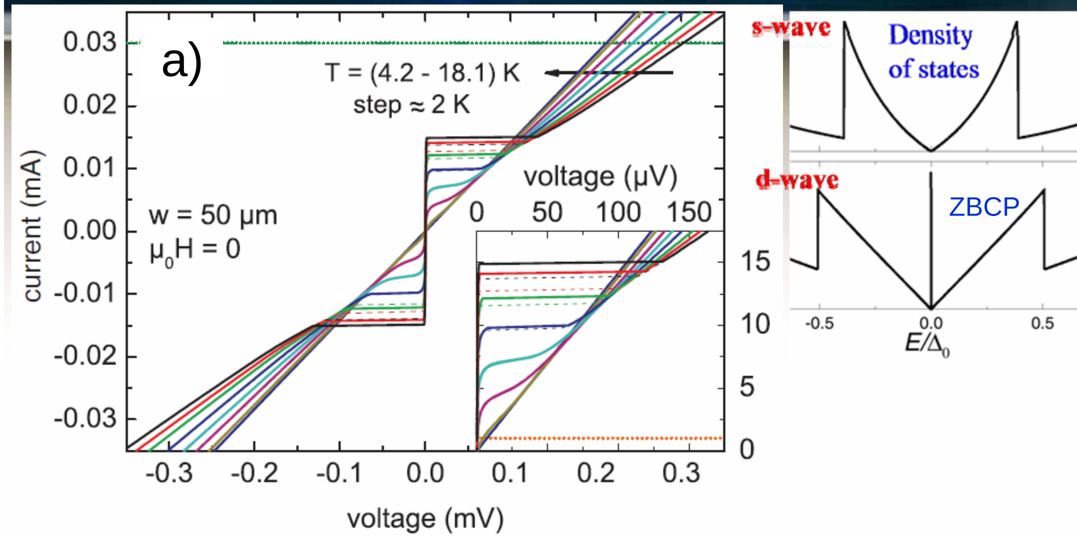
SLCO on $BaTiO_3$/(001) $SrTiO_3$

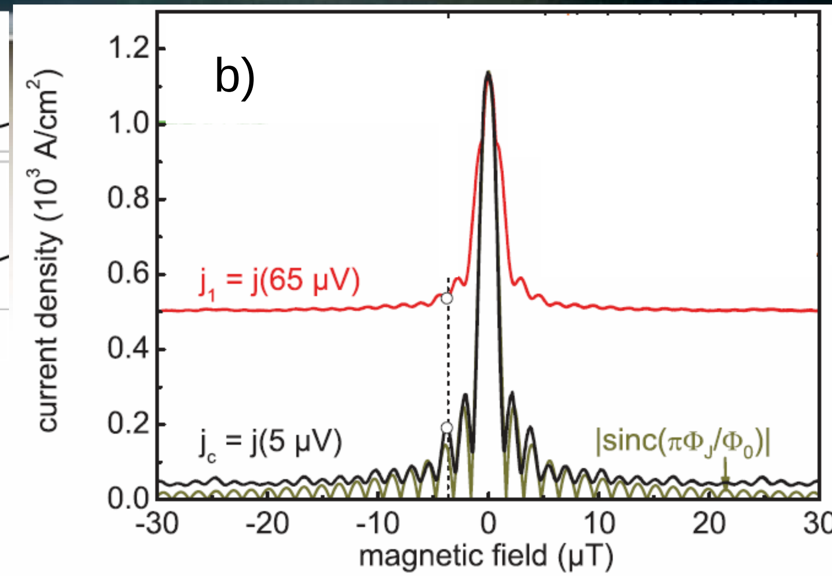
Migration of the structural defects at the BTO/STO interface due to high temperature annealing

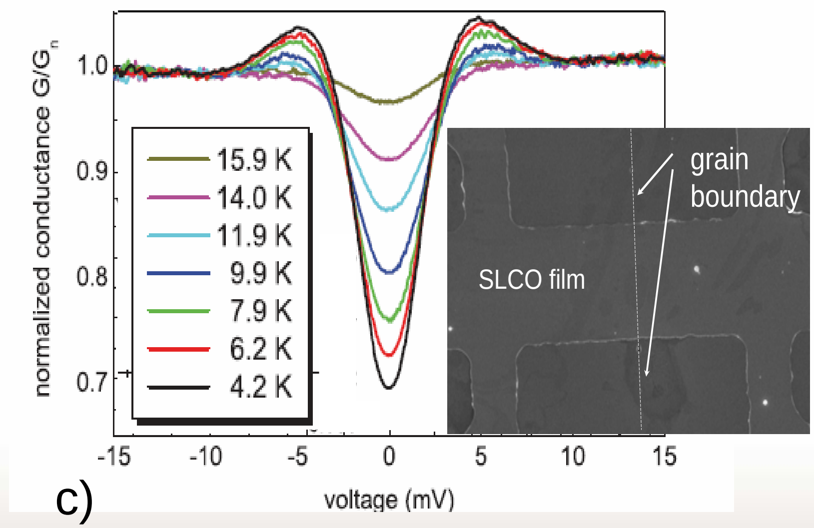
Terai et al., APL 80 (2002)

Josephson junctions:
fabrication and transport properties

$Sr_{1-x}La_xCuO_2$ 24º symmetric grain boundary junctions







a) ICVs: resistively and capacitively shunted junction (RCSJ)-like, with no significant excess current;
- J_c (@ 4.2 K) ~ 1.2 kA/cm^2 – 1-2 orders of magnitude above J_c of 24º GB based on NCCO and LCCO

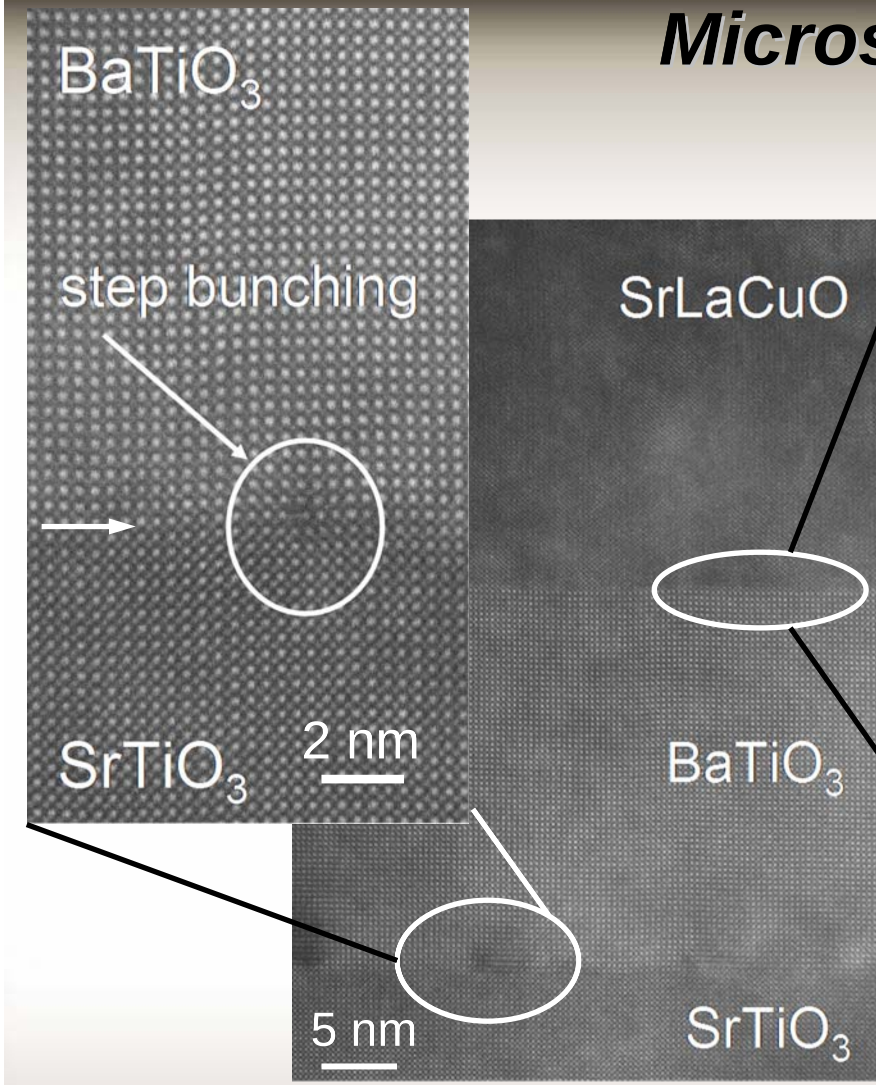
b) highly regular Fraunhofer-like patterns for different voltage criterion

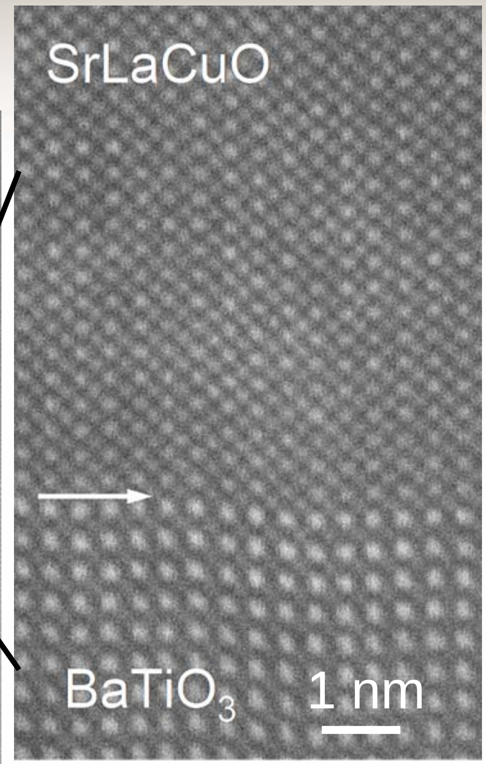
c) conductance spectra did not show a zero-bias conductance peak. s-wave symmetry?
but the V-shaped of the spectra in the subgap regime may indicate an order parameter with nodes

Tomaschko, Leca, Selistrovski, Kleiner, Koelle, Phys. Rev. B 84 (2011)

$Sr_{1-x}La_xCuO_2$ 24º symmetric grain boundary junctions

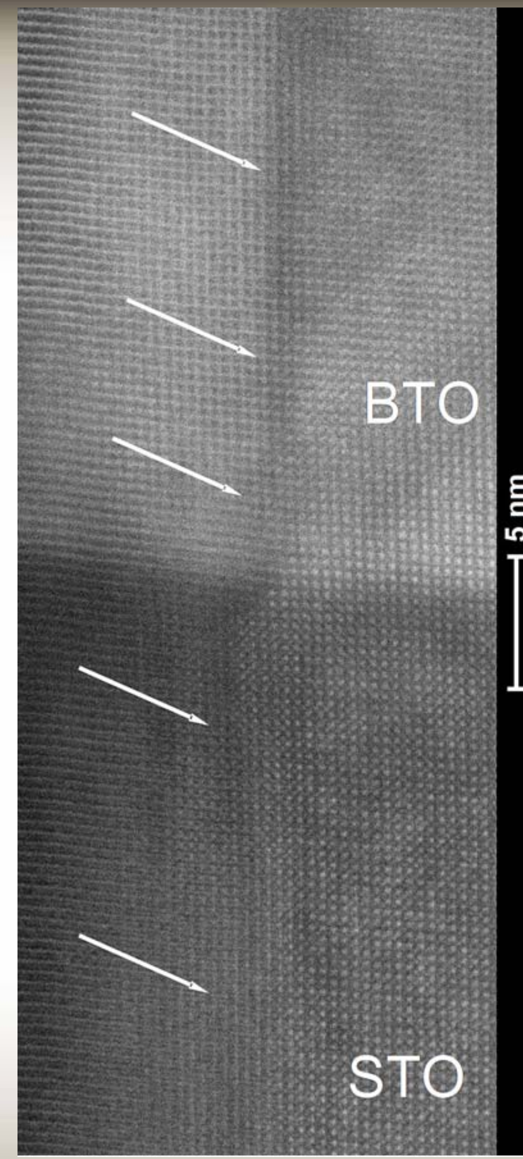
Microstructure



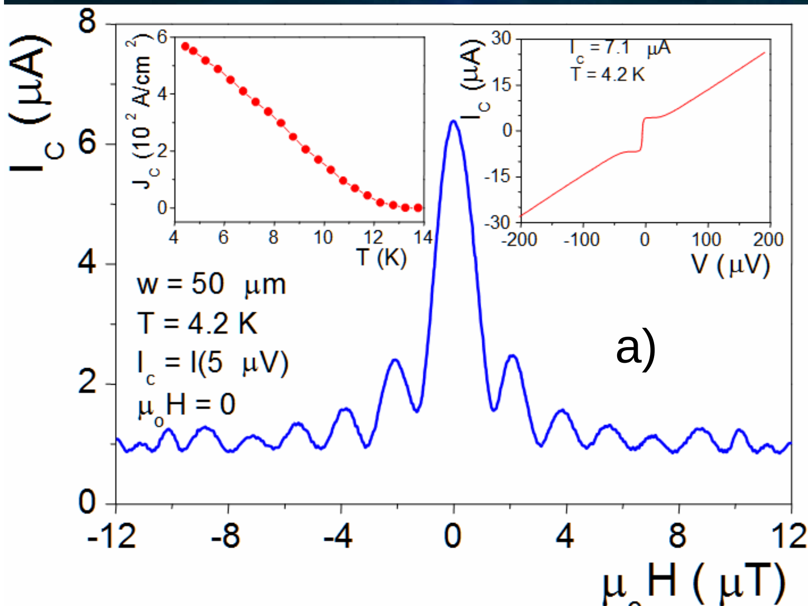


HRTEM data
$Sr_{0.85}La_{0.15}CuO_2$
on $BaTiO_3/SrTiO_3$

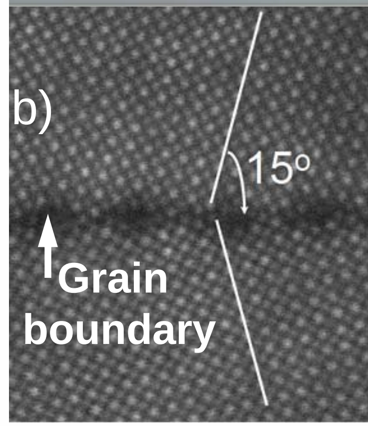
24º grain boundary



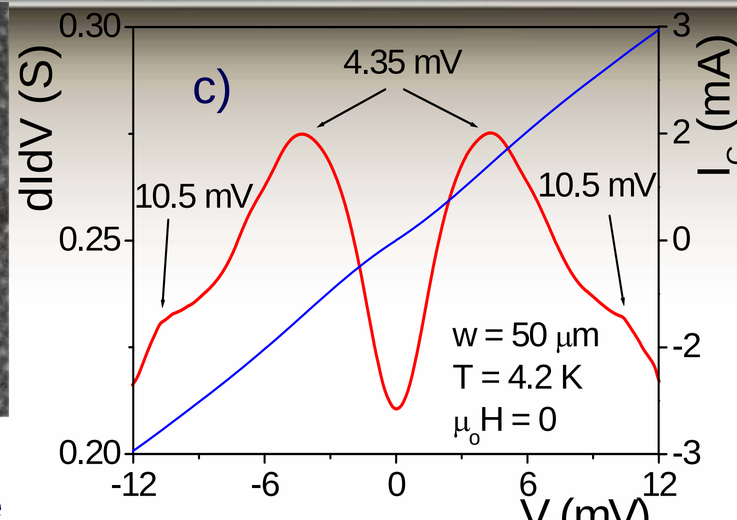
$Sr_{1-x}La_xCuO_2$ 30^o symmetric grain boundary junctions



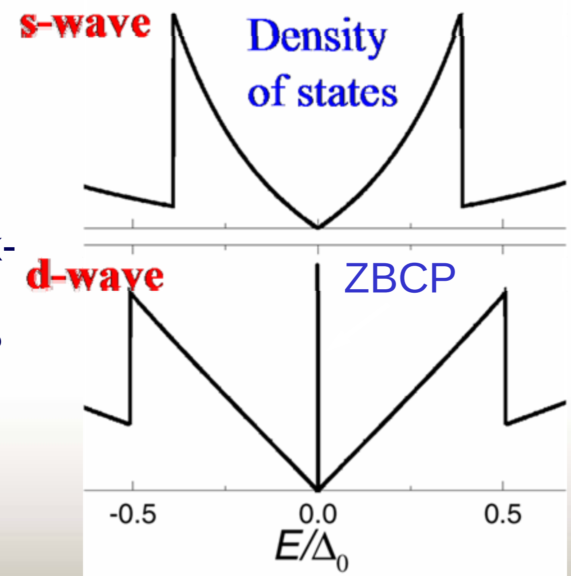
I_CH, J_cT, and I_cV characteristics



HRSTEM image ($SrTiO_3$ bicrystal)



Conductance spectra

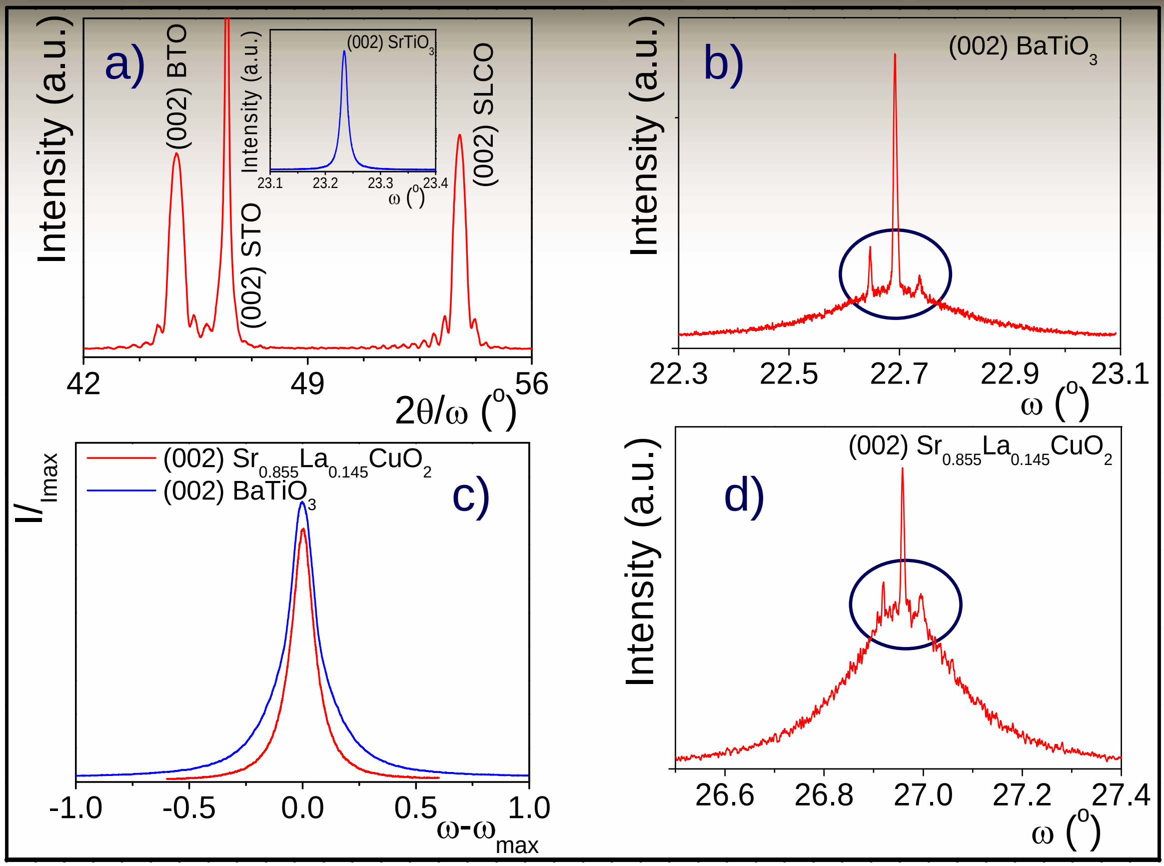


a) I_cVs: resistively and capacitively shunted junction (RCSJ)-like;
- highly regular Fraunhofer-like patterns;
- high J_c @ 4.2 K of ~ 0.55 kA/cm^2; I_cR_n of ~ 50 μV

b) conductance spectra did not show a zero-bias conductance peak-ZBCP, but a V-shaped subgap spectra ***and*** extra peaks above the coherence peaks; microscopic roughness due to structural defects?

c) plain view HRTEM of the substrate (30^o) grain boundary.

$Sr_{1-x}La_xCuO_2$ 30º symmetric grain boundary junctions



Microstructure

Misfit strain accommodation by lattice modulations
Two components in ω scans: strained and relaxed

lattice modulations visible *only* in the HRXRD scans

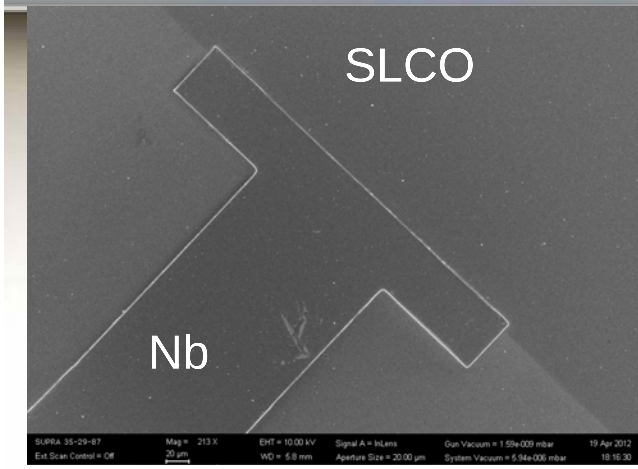
XRD (a,c) and HRXRD (b, d) data for BTO/SLCO films grown on 30º GB

XRD scans in double (one monochromator) and triple (two monochromators) axes configuration showing the presence of dislocations in the (buffer) $BaTiO_3$ and SLCO layers.

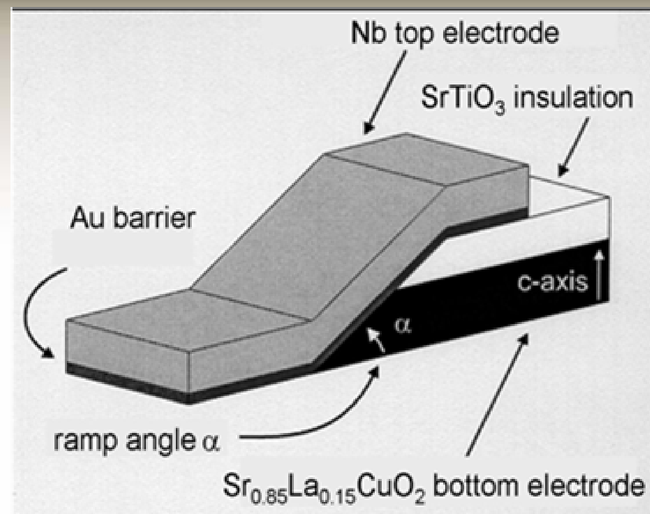
HRXRD: triple axes configuration
XRD: double axes configuration

Leca, Tomaschko, Danila, Kleiner, Koelle, unpublished

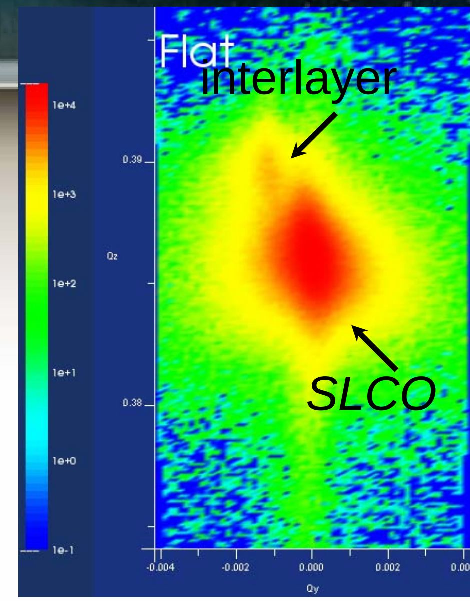
$Sr_{1-x}La_xCuO_2$ ramp-type junctions



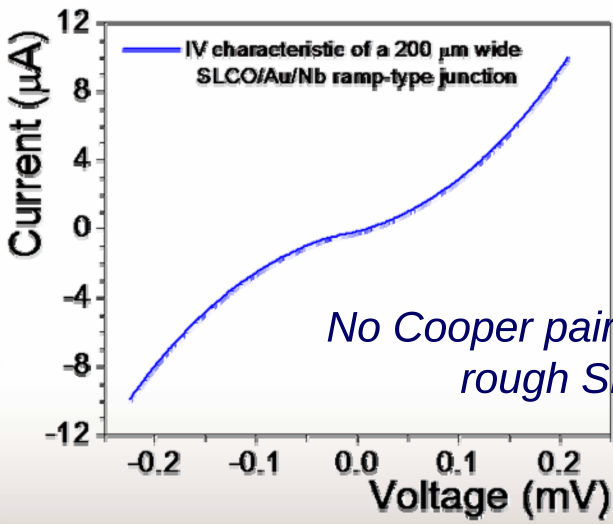
SEM image of a 200μm wide SLCO/Au/Nb ramp-type junction



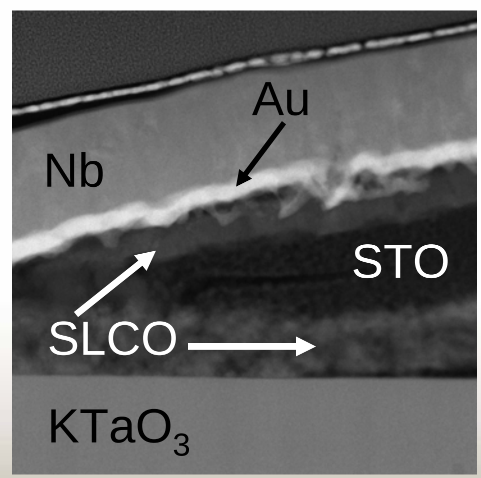
Schematic representation of the SLCO/Au/Nb ramp-type junction



XRD rsm scan of (101) showing the separation of the SLCO and interlayer peaks



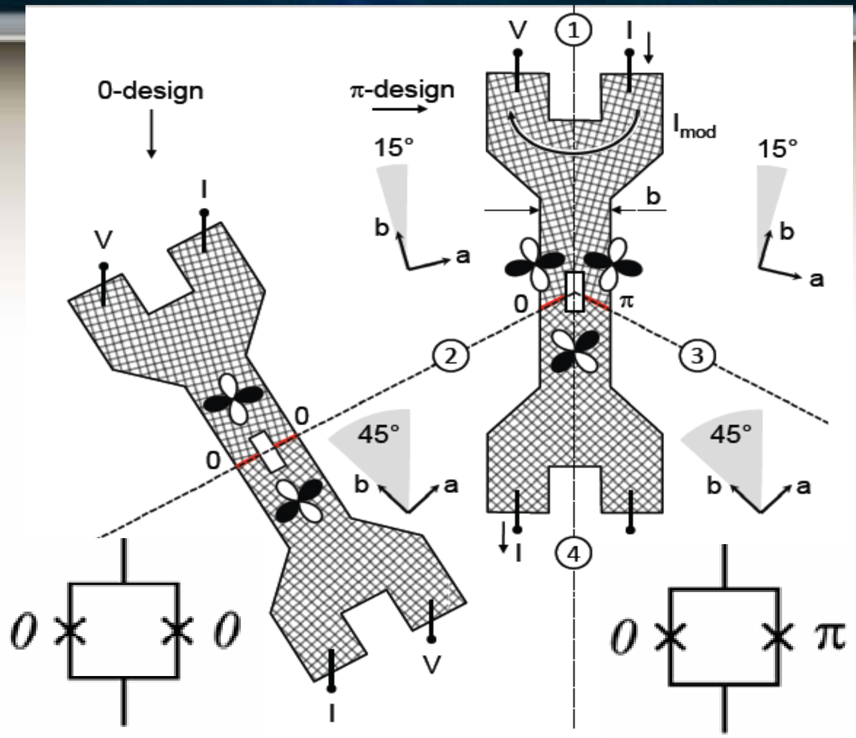
No Cooper pair tunneling observed; rough SLCO/Au interface



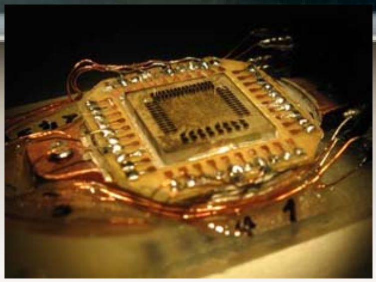
HRTEM

Phase sensitive experiments using π-SQUIDs

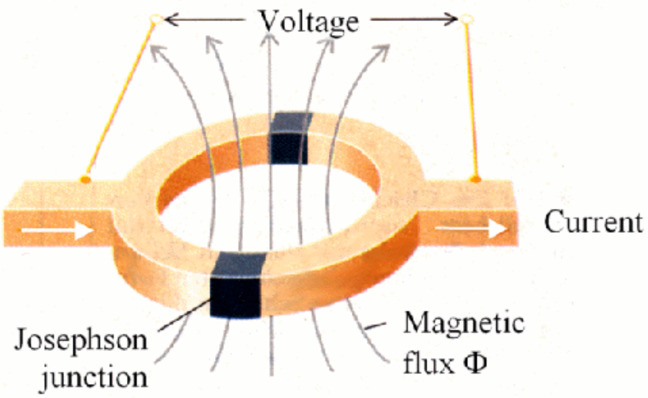
Pairing symmetry from phase sensitive experiments



Schematic layout of the
0 - and π-SQUIDs



The $SrTiO_3$ tetracrystal with structured SQUIDs



Design of a dc-SQUID

Geometry designed to be frustrated for d-wave pairing.
If d-wave: the dc-SQUID ring around the tetracrystal point contains one 0-junction and one π-junction (forming the π-SQUID, which exhibits an additional π shift in its phase).

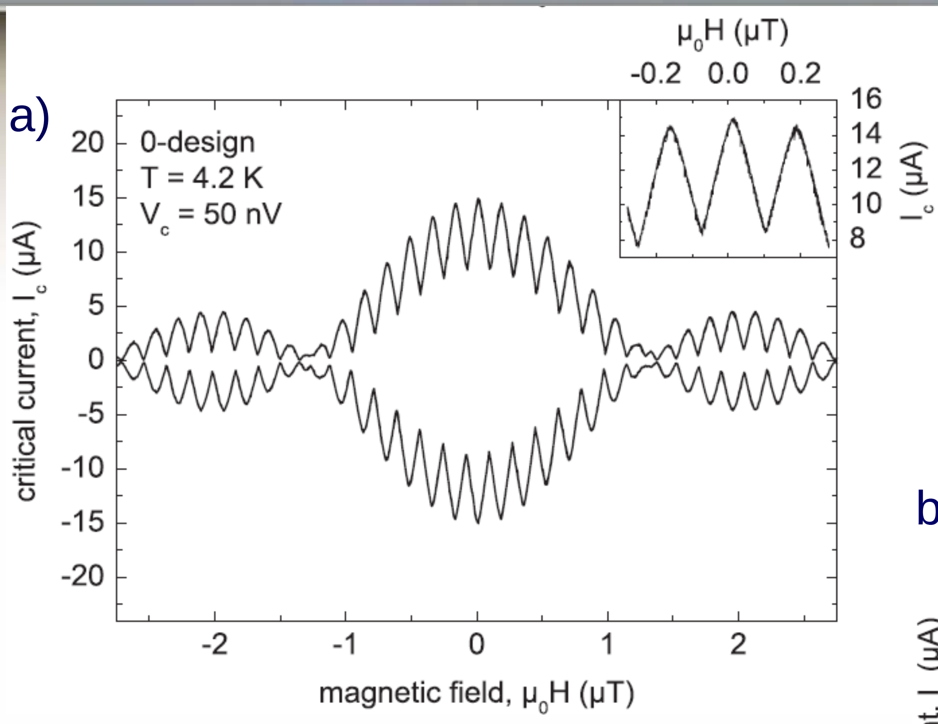
Phase-sensitive experiments rely on sign change in the Josephson I_c (a qualitative signature of unconventional Sc), and not on quantitative magnitude of the I_c.

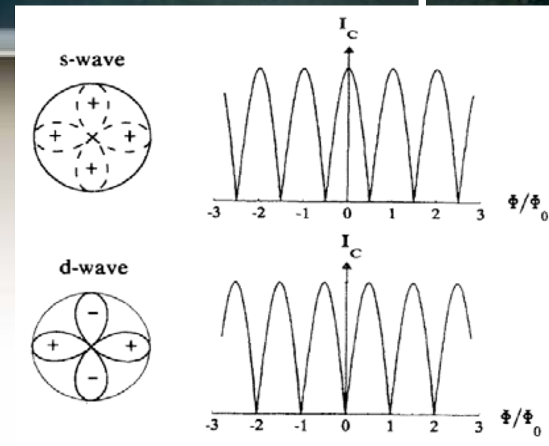
Tsuei et al. RMP 72 (2000)

[Schulz et al., APL 76 (2000); Chesca et al., PRL 90 (2003)]

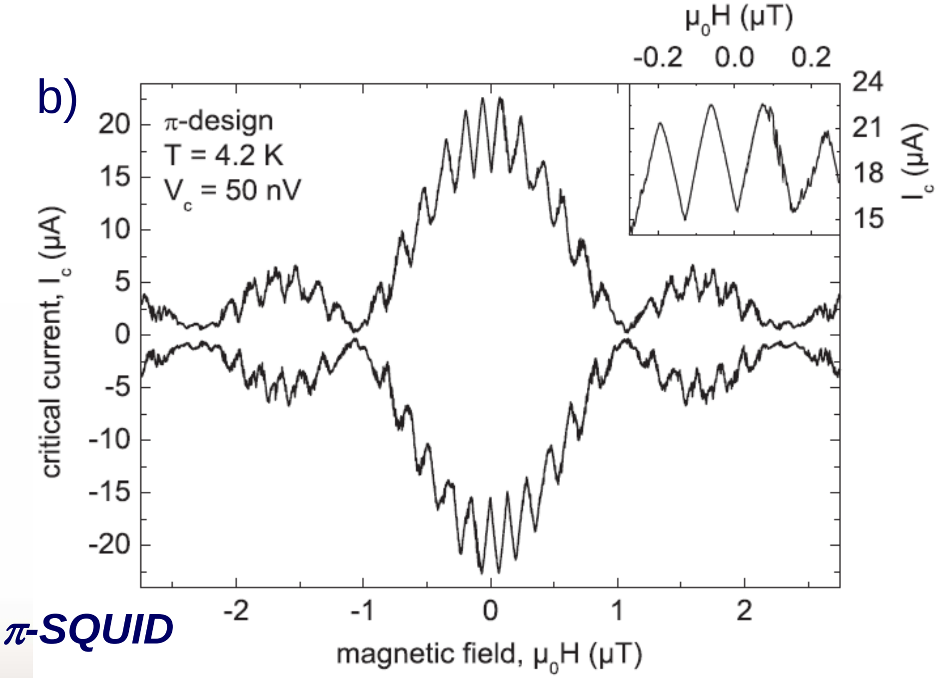
Pairing symmetry from phase sensitive experiments

a)





b)



I_c vs magnetic field for
a) 0 - and b) π-SQUID

The I_c vs. H shows a minimum at H=0 for the π-SQUID due to the phase shift across the π-junction.

Predominantly $d_{x^2-y^2}$ pairing symmetry

Tomaschko et al., Phys. Rev. B 86 (2012)

Conclusions

The superconducting properties of the PLD grown electron-doped $Sr_{1-x}La_xCuO_2$ thin films are still far of those of the bulk or of the MBE grown films; however, higher phase stability can be achieved by PLD (in the over doped region).

No zero bias conductance peak observed in grain-boundary junctions, most probably due to oxygen vacancies along the grain boundary.

Phase sensitive experiments shown a predominantly d-wave order parameter symmetry in the $Sr_{1-x}La_xCuO_2$ (x ~ 0.15) thin films.