物構研シンポジウム'09「放射光・中性子・ミュオンを用いた表面・界面科学の最前線」
エポカルつくば, 2009年11月17日～18日

強相関酸化物ヘテロ界面の相競合と新規物性

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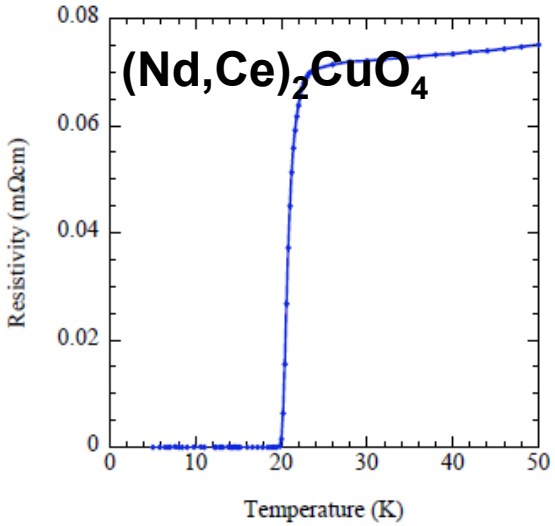
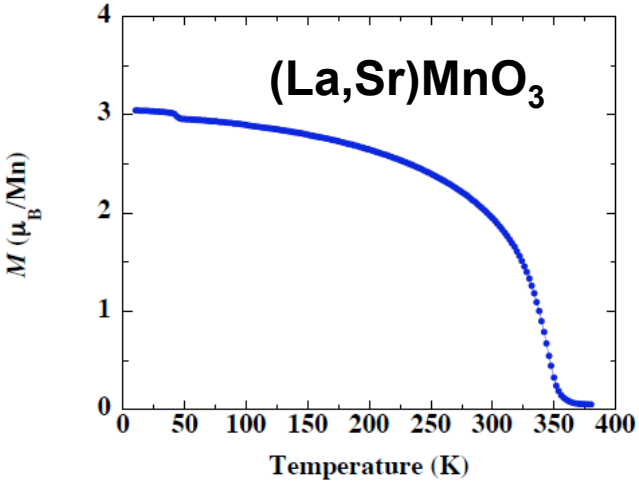
[1]産業技術総合研究所
[2]JST-CREST

強相関遷移金属酸化物

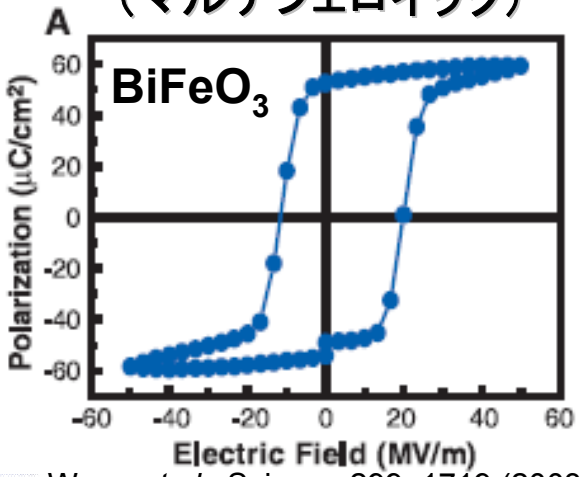
超伝導



強磁性

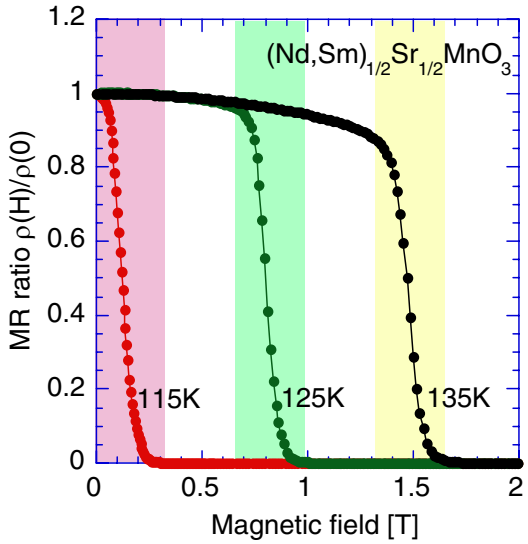


強誘電＋反強磁性（マルチフェロイック）



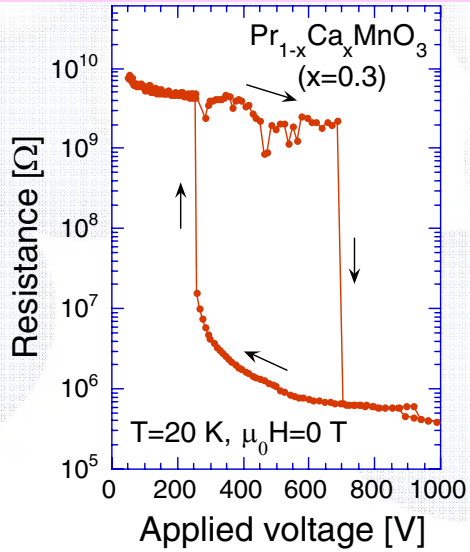
Wang *et al.*, Science 299, 1719 (2003)

磁場制御



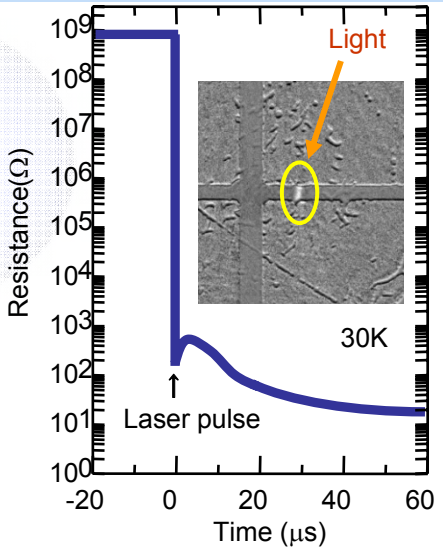
Tomioka *et al.*

電場制御



Asamitsu *et al.*, Nature 388, 50 (1997)

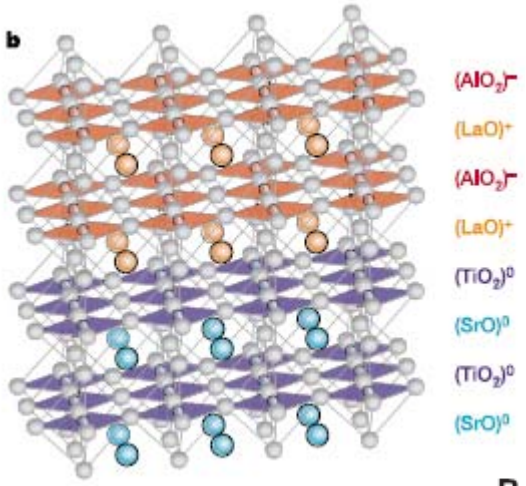
光制御

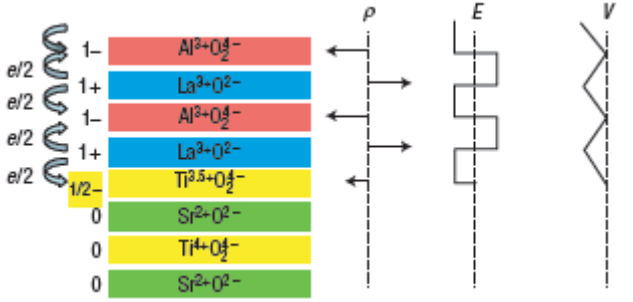


Fiebig *et al.*, Scinence 280, 1925 (1998)

遷移金属酸化物界面の特異な電子状態

$LaAlO_3$-$SrTiO_3$ Interface (band insulators)

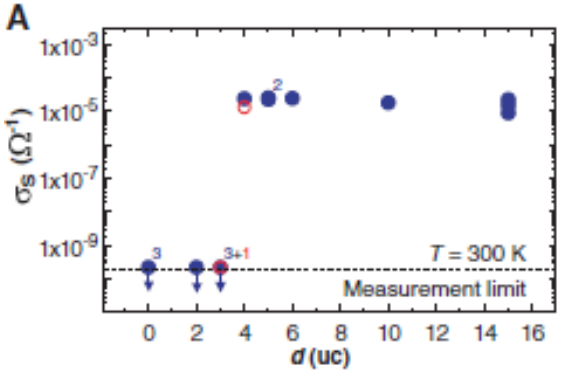




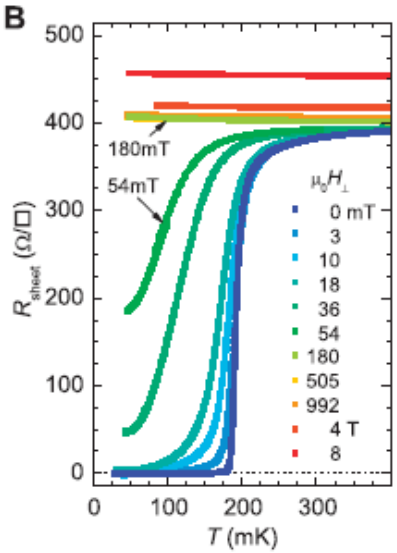
Polar catastrophe

Ohtomo and Hwang,
Nature 427,423 (2004)
Nakagawa *et al.*,
Nature Mater. 5, 204 (2006)

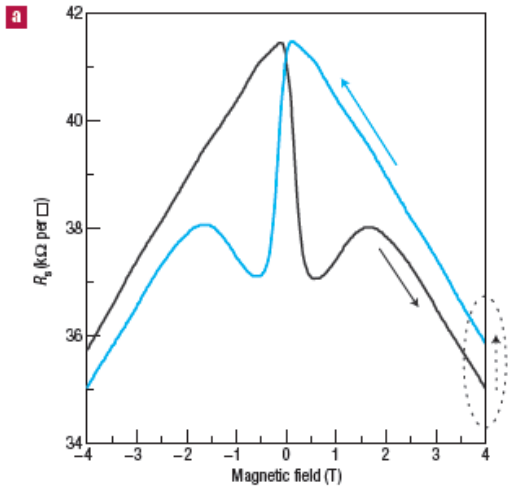
two-dimensional electron gas?



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



Reyren *et al.*,
Science 317,1196 (2007)

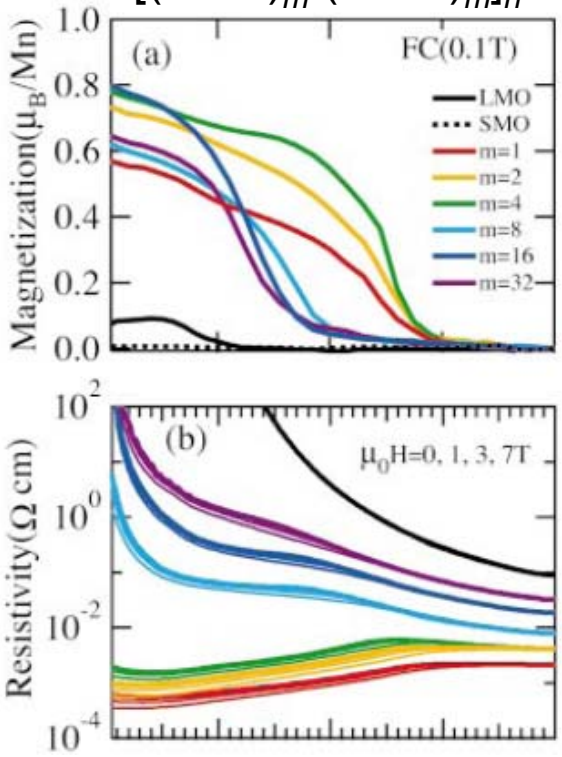


Brinkman *et al.*,
Nature Mater. 6, 493 (2007)

強相関酸化物界面の特異な電子状態

$LaMnO_3$-$SrMnO_3$ Interface (Mott insulators)

$[(LMO)_m/(SMO)_m]_n$

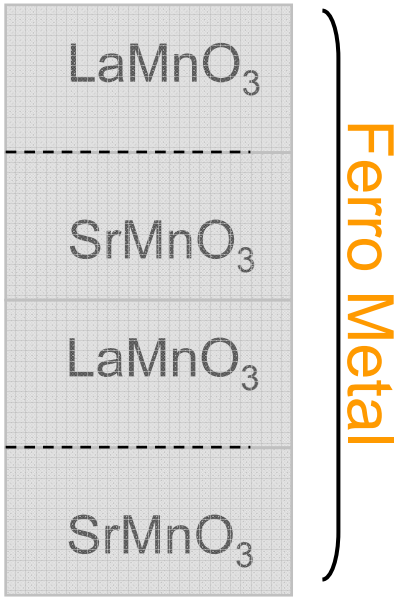


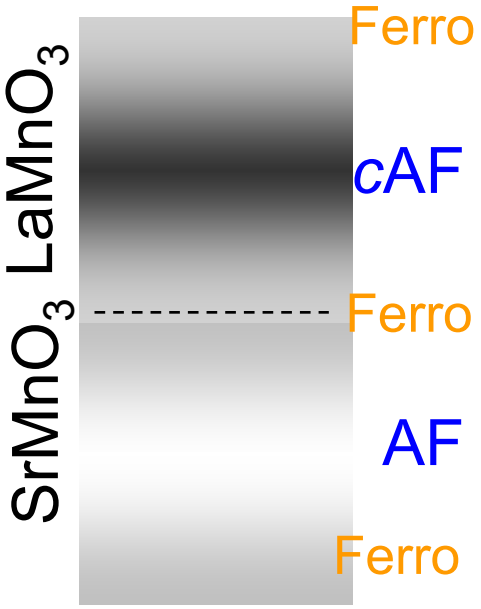
Koida *et al.*,
PRB 66, 144418 (2002)

Salvador *et al.*, APL 75, 2638 (1999)
Yamada *et al.*, APL 89, 052506 (2006)
Bhattacharya *et al.*, PRL 99, 196404 (2007)

Short-Period
Uniform Valence
Alloy-like metal

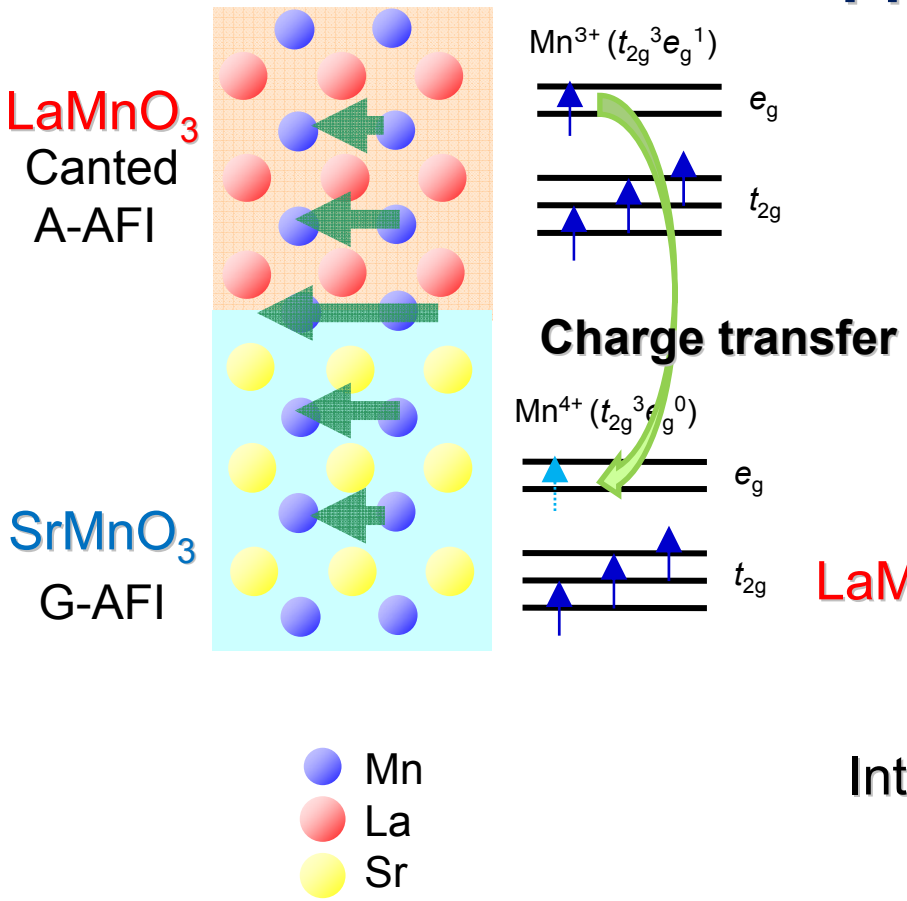
Long-Period
Nonuniform Valence
Interface Ferro.



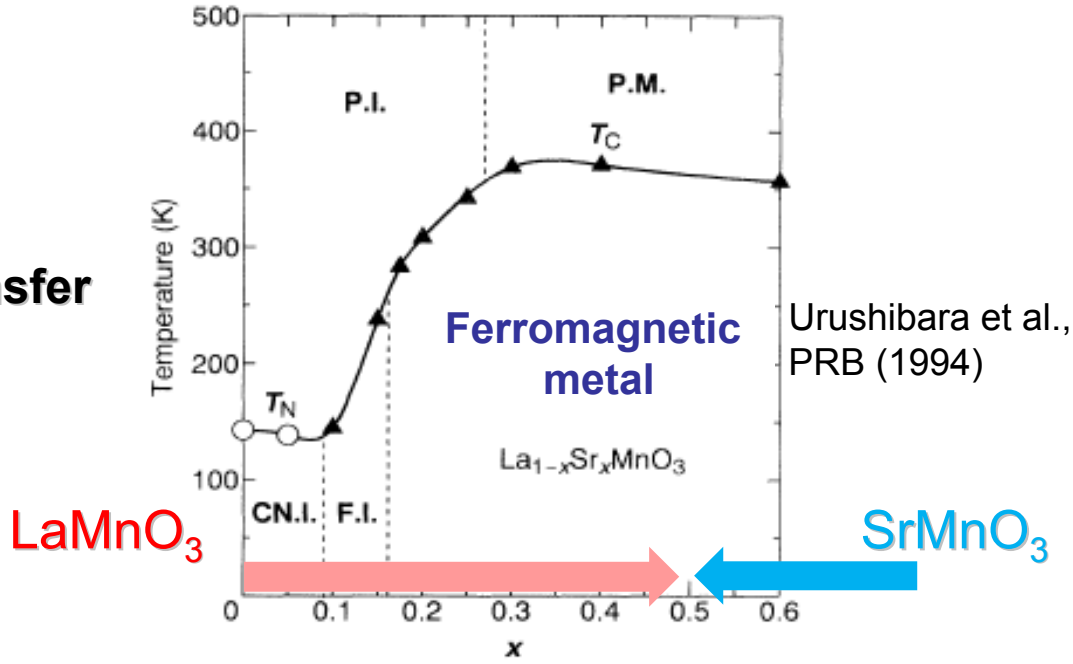


Charge transfer at interfaces

Electronic reconstruction at LMO-SMO interface



Phase diagram of $La_{1-x}Sr_xMnO_3$

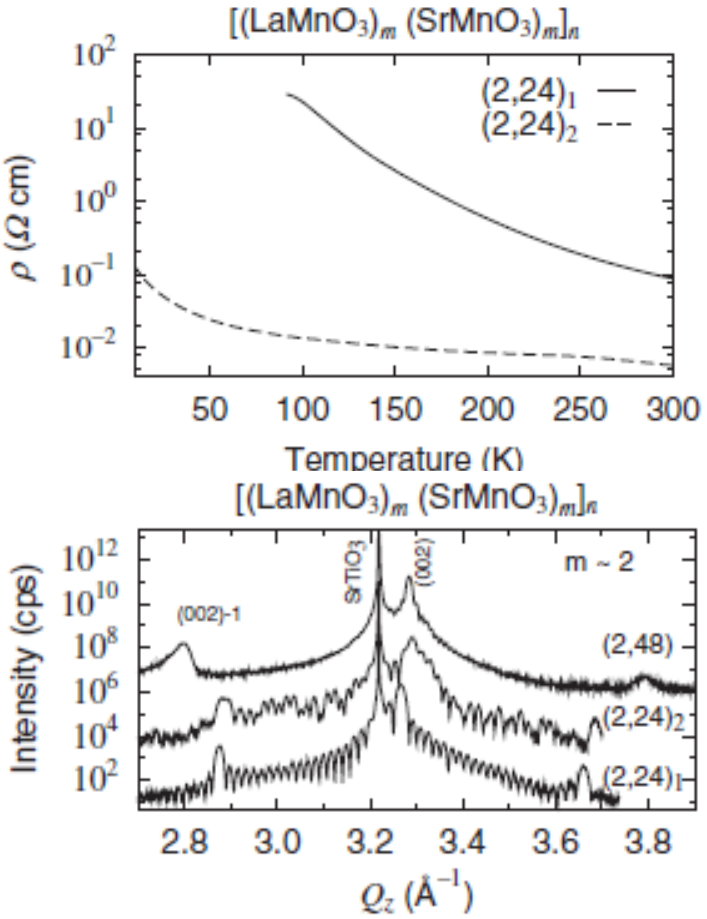


Urushibara et al., PRB (1994)

Interface has a doping level intermediate between LMO and SMO?
(~0.5)

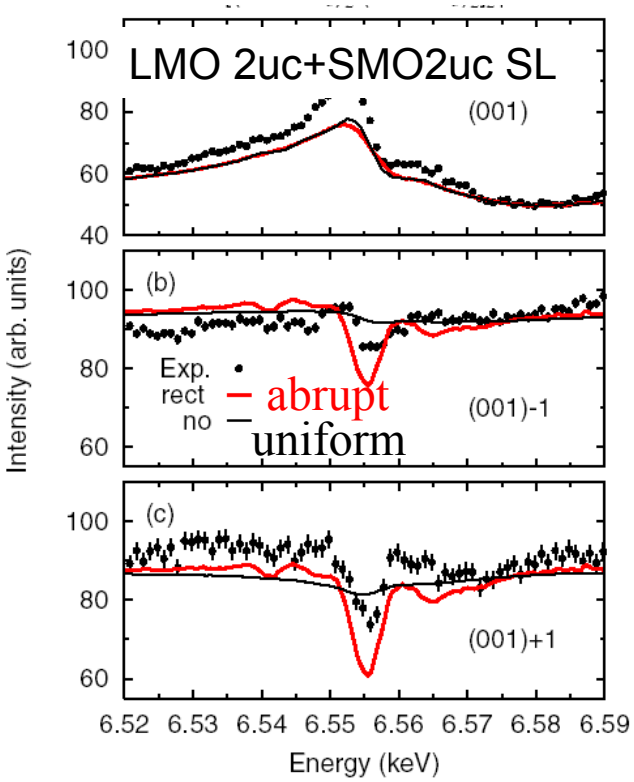
Recent study

H. Nakao (PF-KEK) *et al*., JPSJ 78, 024602 (2009)



Interface roughness changes interfacial electronic states

Resonant X-ray Scattering

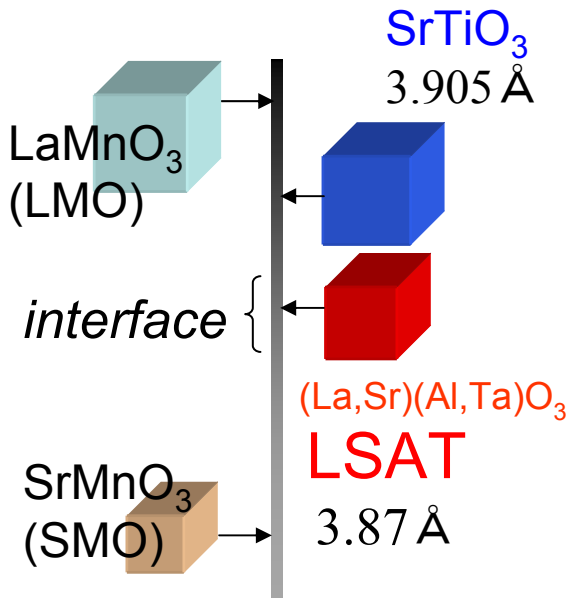


Valence modulation may be very small.

Intrinsic electronic states at LMO-SMO interface ??

Strategy for high-quality superlattice synthesis

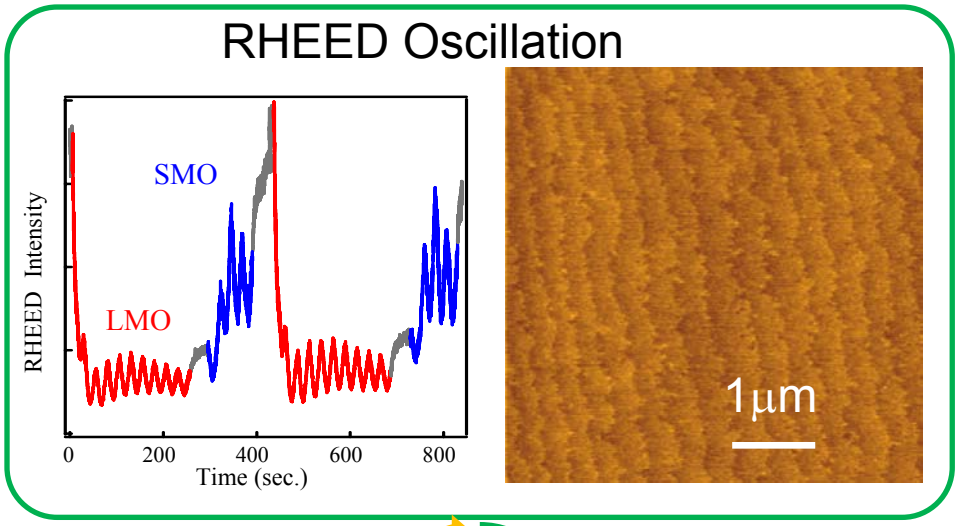
(1) Substrate-choice



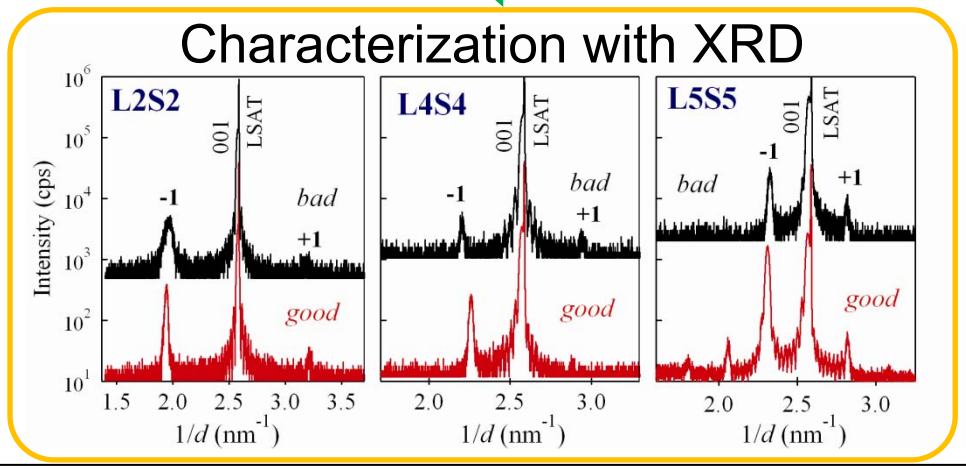
STO：tensile-strain

LSAT: lattice-matching

(2) Improvement of Samples

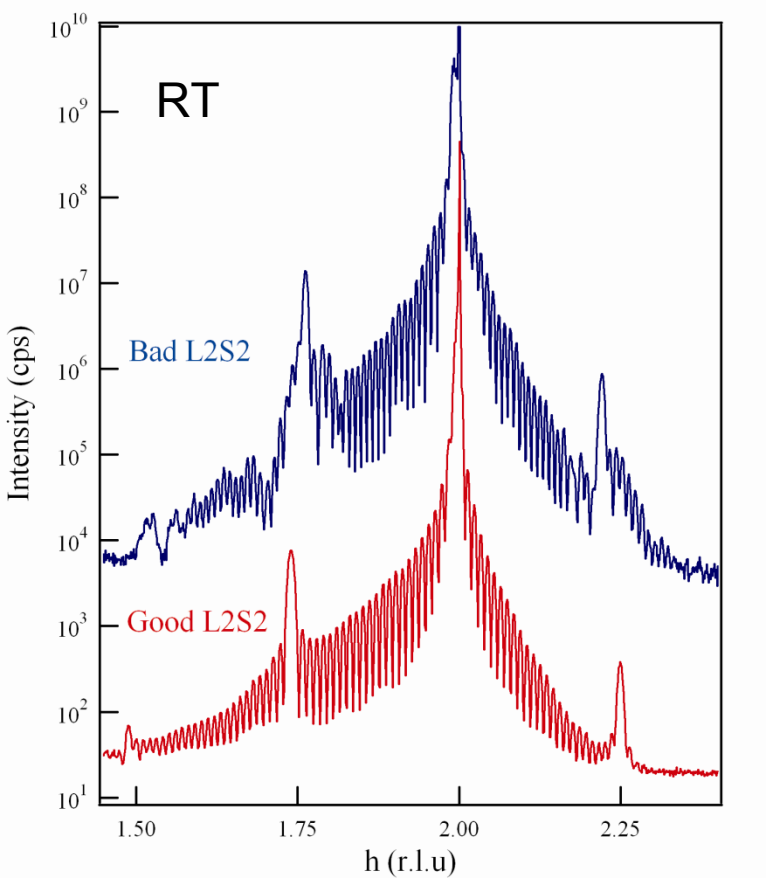


Feedback



Characterization with Synchrotron XRD

BL4c , Photon Factory, KEK, Tsukuba, 2009 (Prof. Nakao)



Bad superlattice

- Sharp superlattice reflection
- Laue Fringes until *h*=1.5

✓ Modulation in superstructure

⇩

Good as *films*, but structural imperfection

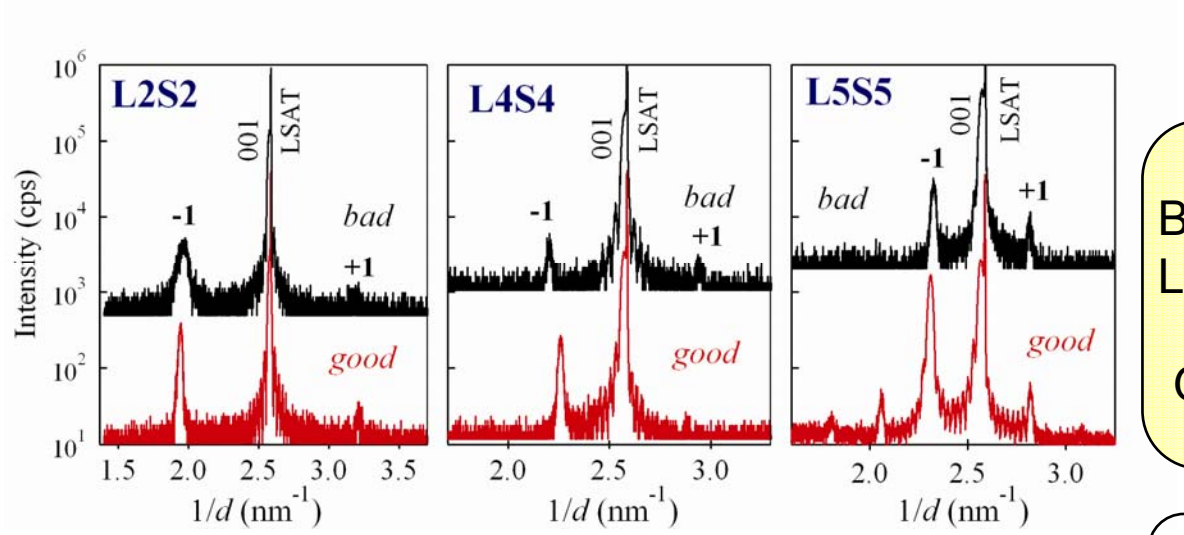
Good superlattice

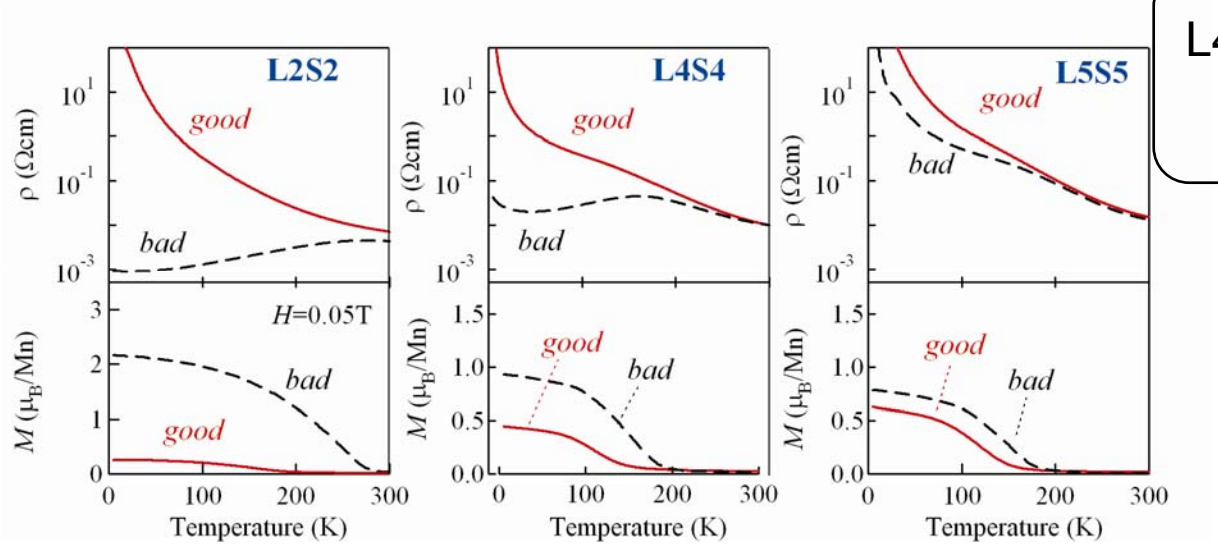
Clear Laue Fringes

⇩

perfect structure

Impact of Improvement on Properties





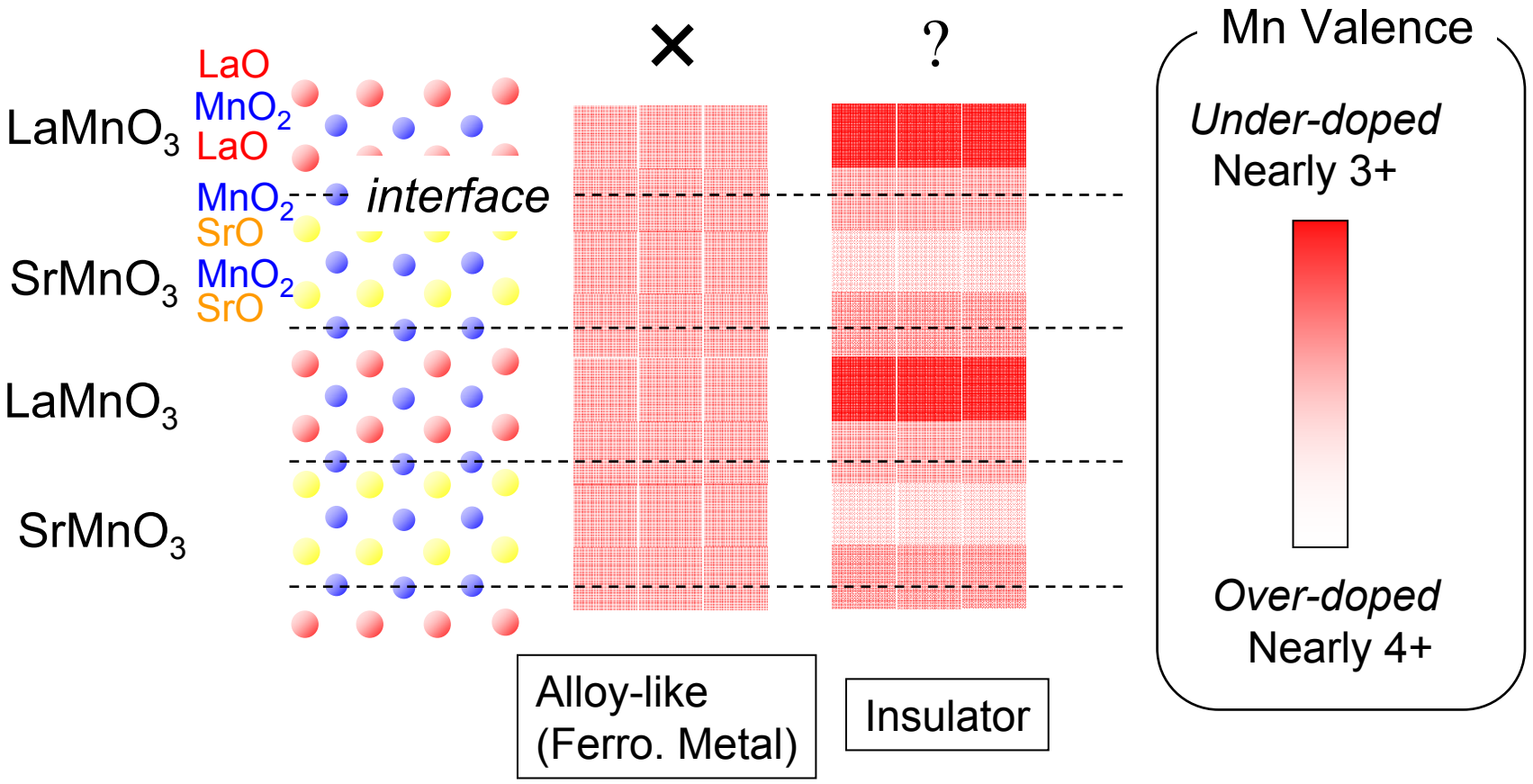
Bad L2S2 = Ferro Metal
$La_{0.5}Sr_{0.5}MnO_3$

Good L2S2 = Insulator

L4S4, L5S5= Ferro Insulator
M, 1/ρ, Tc
Bad > Good

Bad- Good
= Extrinsic FM
(interface rougness etc.)

Short-Period LMO-SMO SL



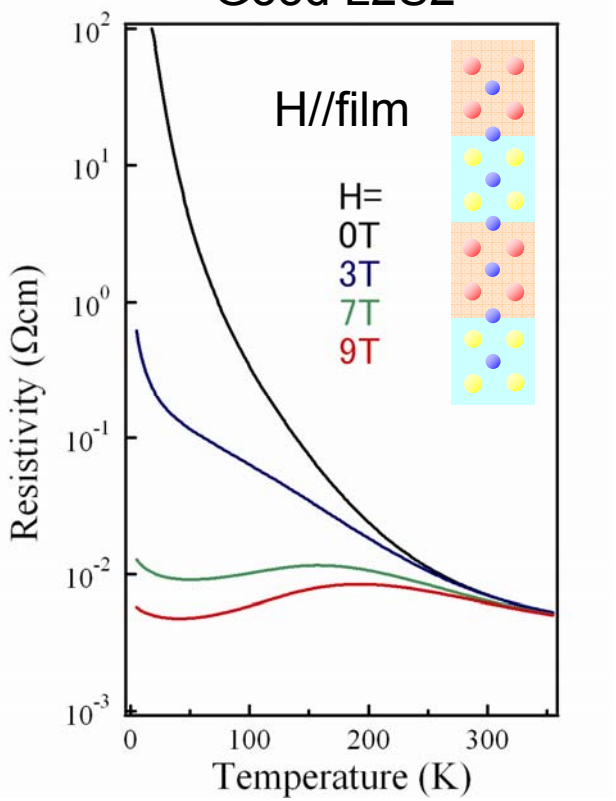
? Even in the short periodicity, electronic state is not uniform.

? Charge transfer alone cannot explain the properties.

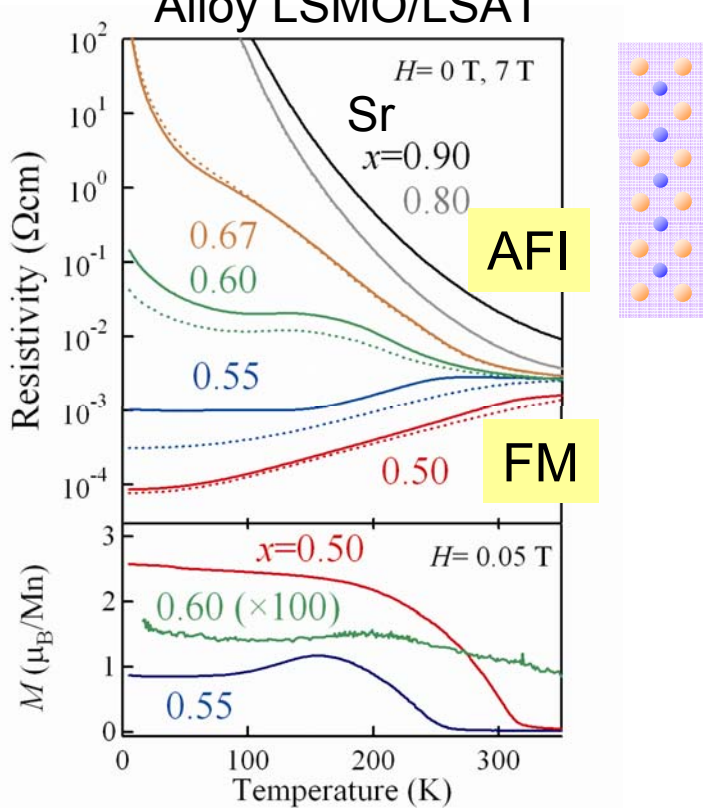
➡ Additional mechanism?

H-Induced Insulator-to-Metal Transition

Good L2S2



Alloy LSMO/LSAT



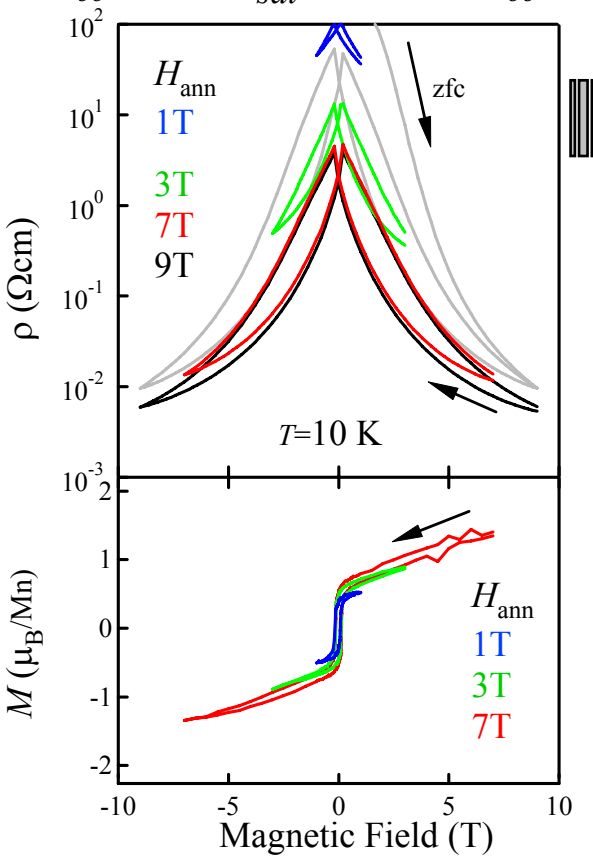
- Metallic for $H \geq 7$T : strong coupling with spin/orbital degrees of freedom

- No large MR effect:

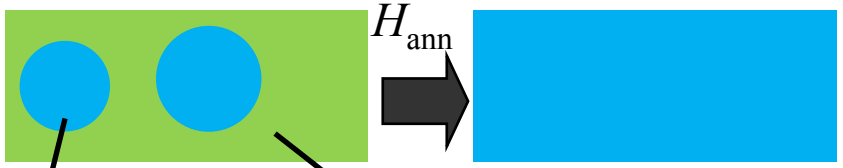
L2S2: Magnetic-Field Dependence

H-annealing effect

History of H-application affects M_{sat} and MR effect.

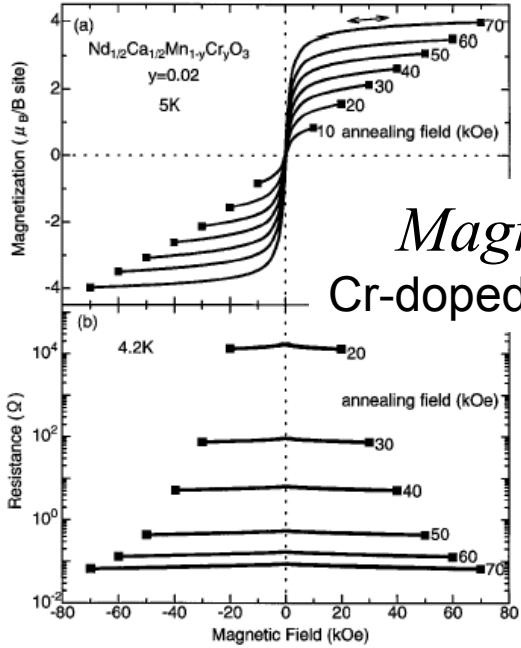


Phase Separation



FM domain

Short-range Orbital-Order (AFI)



T. Kimura et al., PRL 83 (1999)

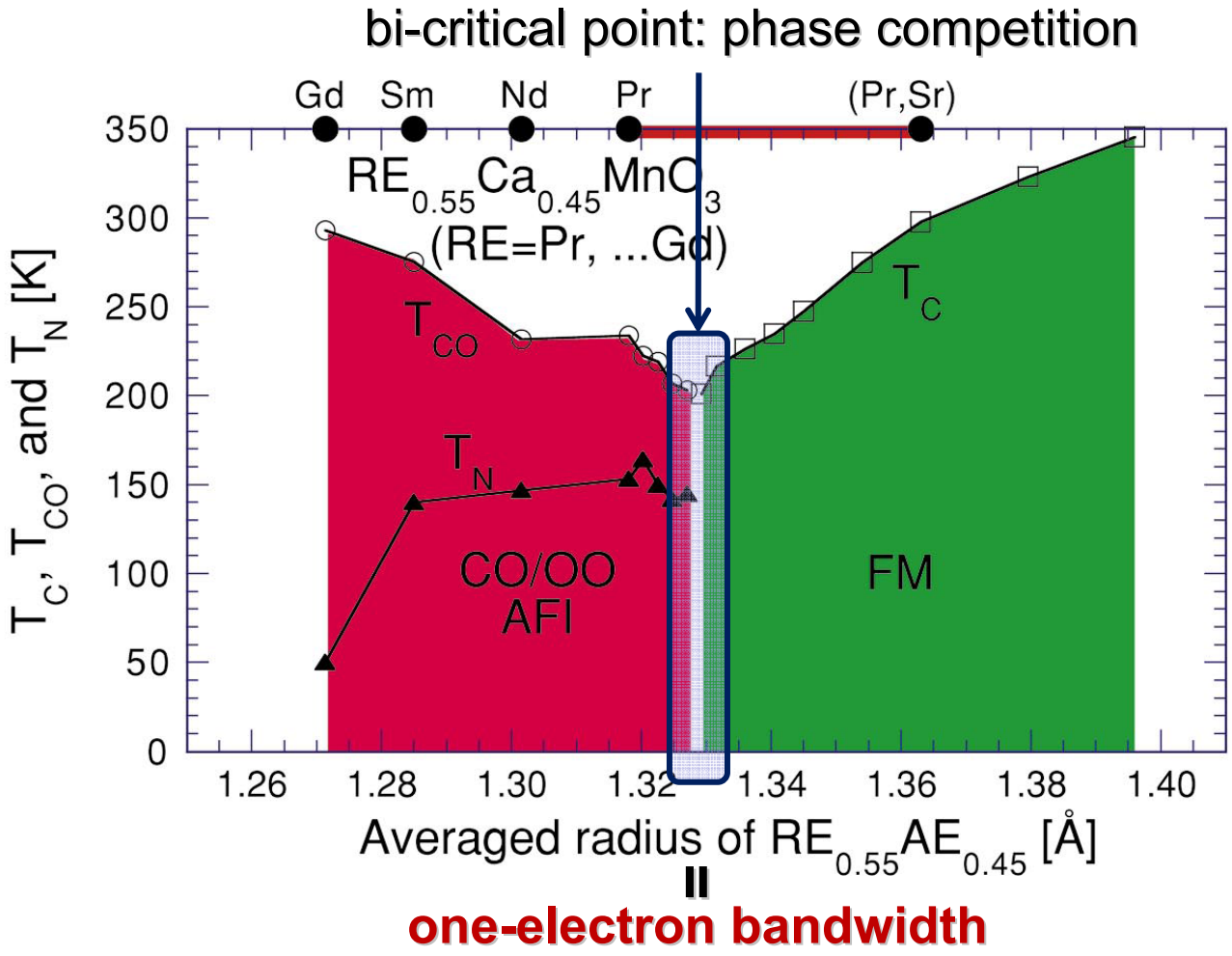
Magnetorelaxor

Cr-doped $Nd_{0.5}Ca_{0.5}MnO_3$

Phase competition at LMO-SMO interface

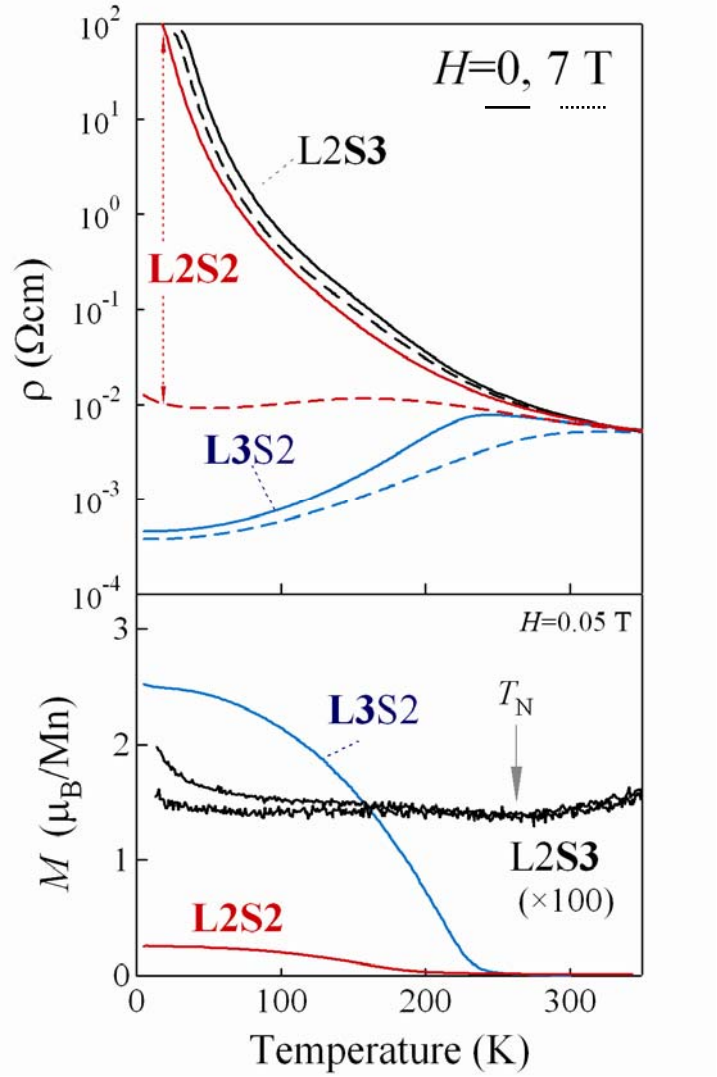
Phase diagram of perovskite Mn-oxides

Tomioka and Tokura, PRB 70, 014432 (2004)



Lattice distortion induces an electronic phase variation

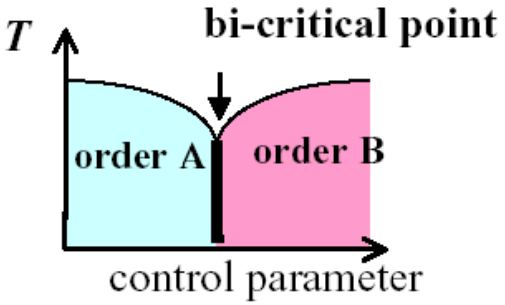
Layer-Thickness Dependence (asymmetric)



LMO 2uc < SMO 3uc

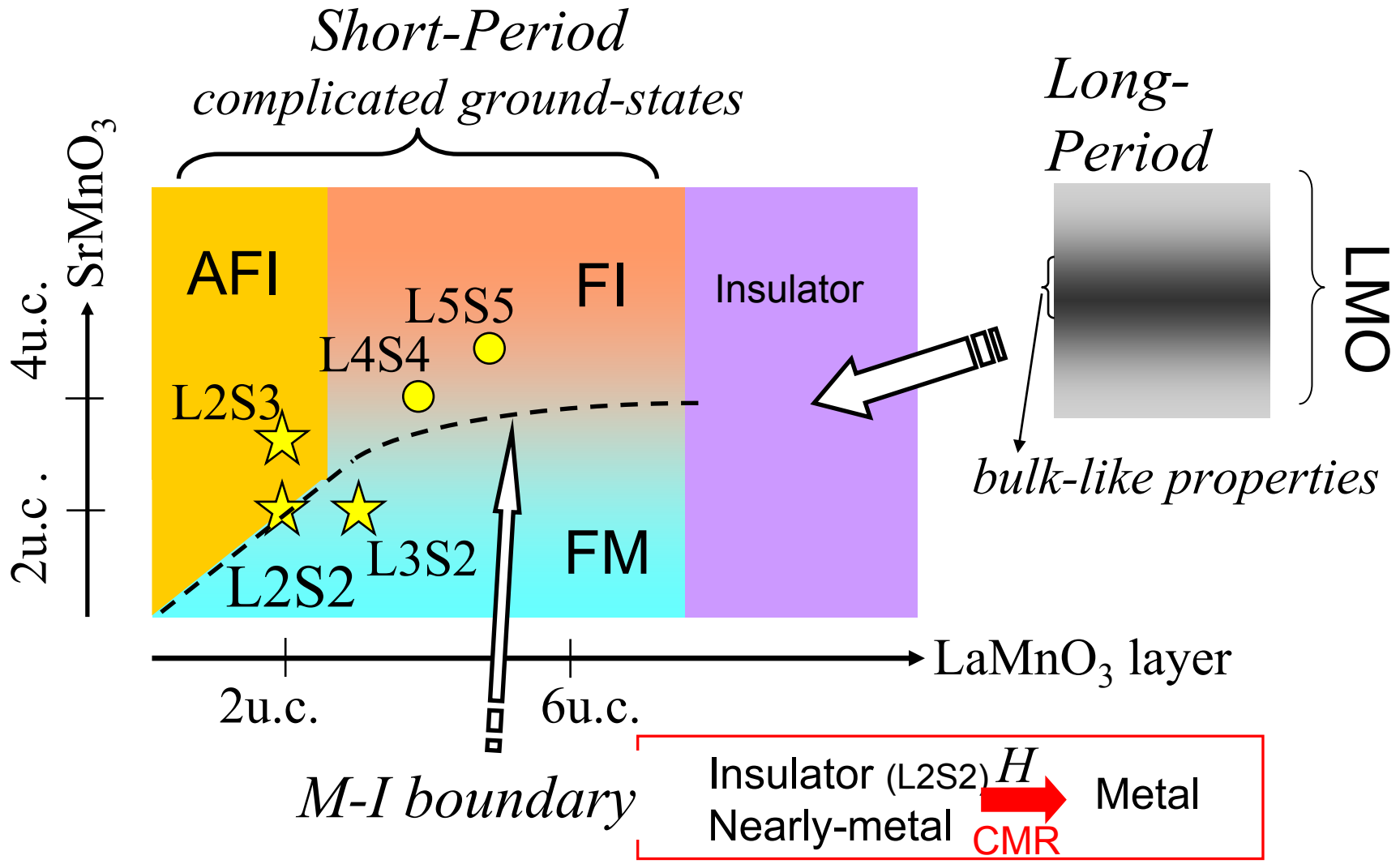
Antiferromagnetic Insulator
(T_N~250K)
Reduced MR (Insulator for $H \leq 9$T)

L2S2
(T_N~200K)



LMO 3uc > SMO 2uc
Ferromagnetic metal
(T_C=240K, M<3μ_B)

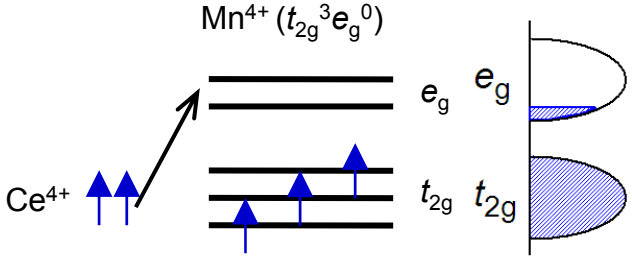
Ground-State Phase Diagram

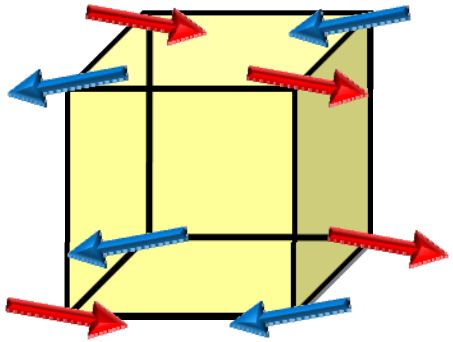


Electron-doped $CaMnO_3$

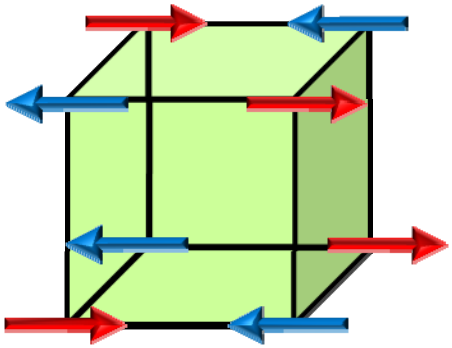
Metal-insulator transition with lightly electron doping

$Ca_{1-x}Ce_xMnO_3$

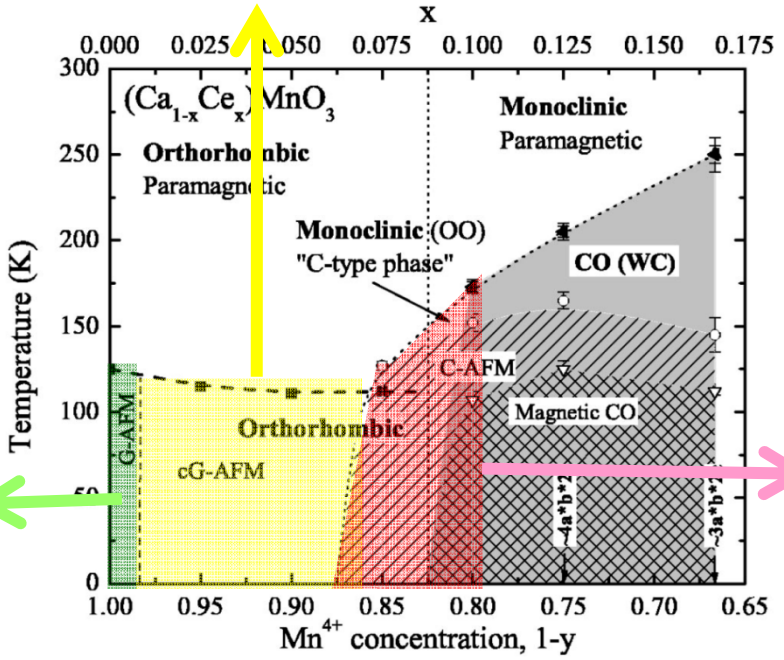


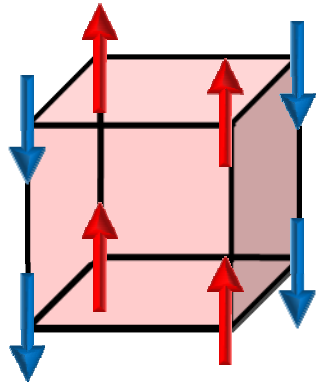


cG-type AF **Metal**



G-type AF **Insulator**





C-type AF **Insulator**

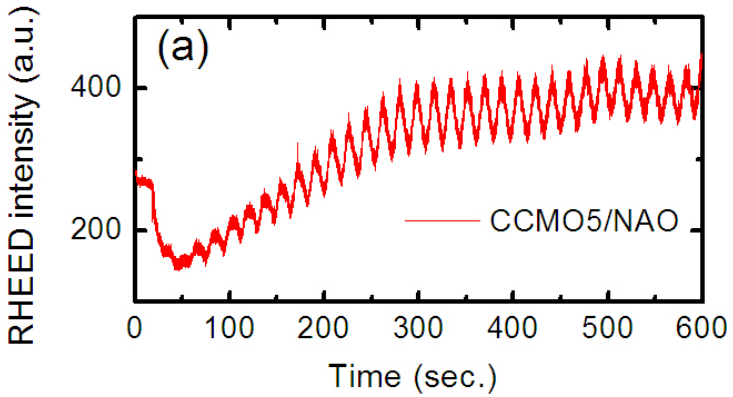
Caspi *et al.*, PRB 69, 104402 (2004)
Tomioka *et al.*, unpublished

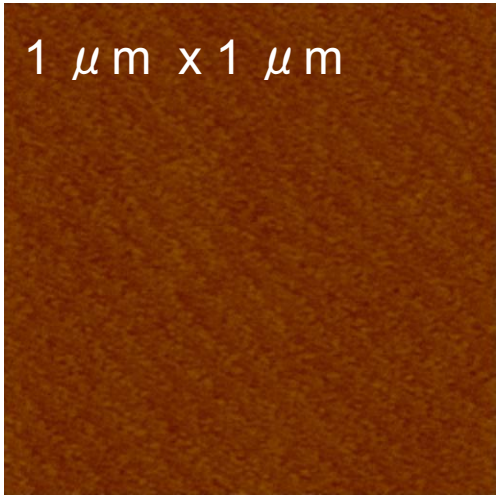
$Ca_{1-x}Ce_xMnO_3$ (CCMO): x = 0 – 0.08

a (CCMO x=0.05) ~ 0.374 nm

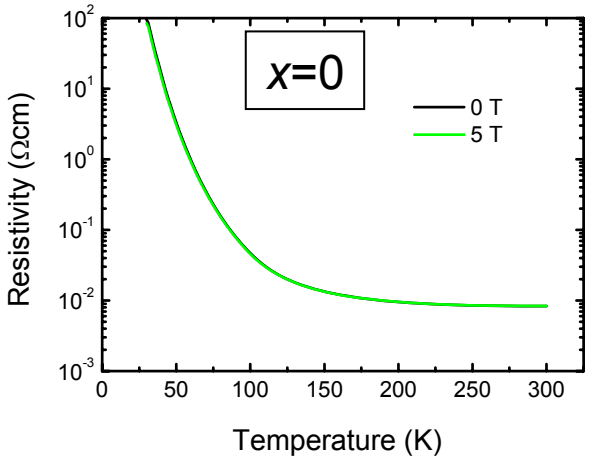
Substrate: (100)-oriented

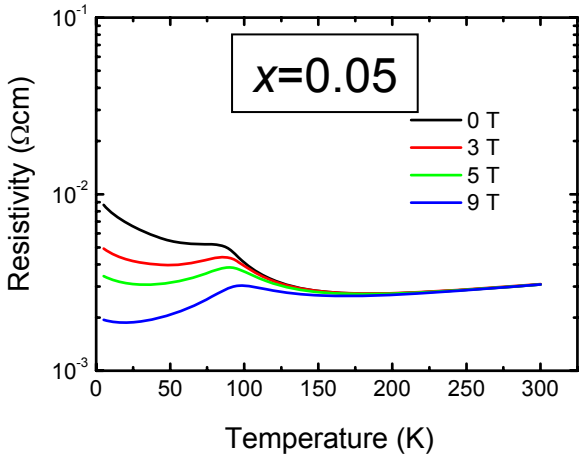
NAO: $NdAlO_3$ (*a* = 0.3751 nm, **+0.3%**)

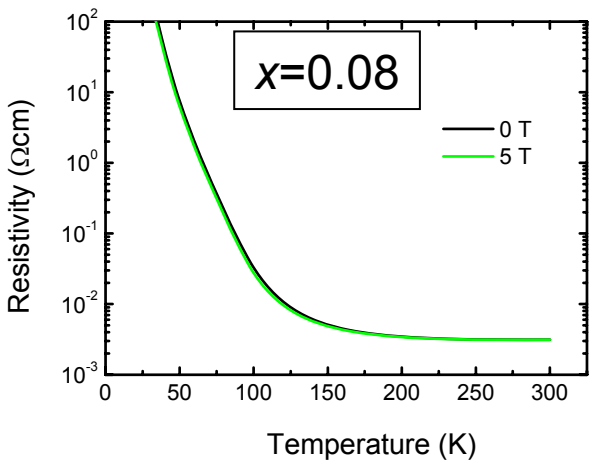


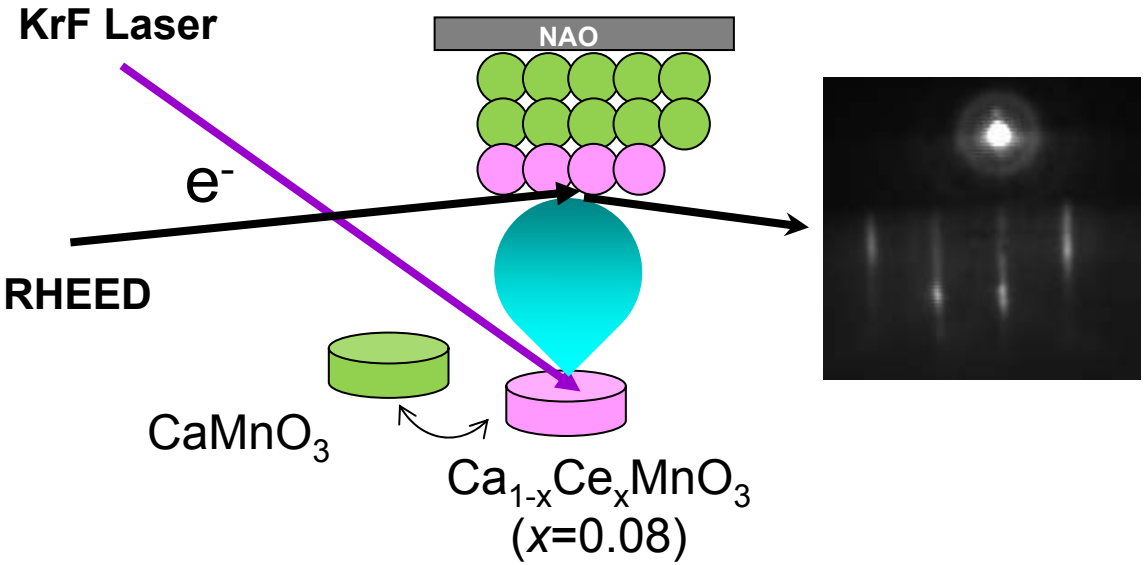


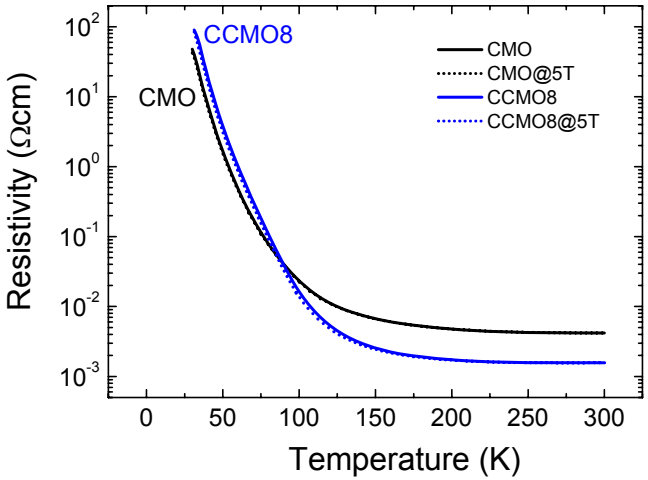
Xiang *et al*., APL 94, 062109 (2009)

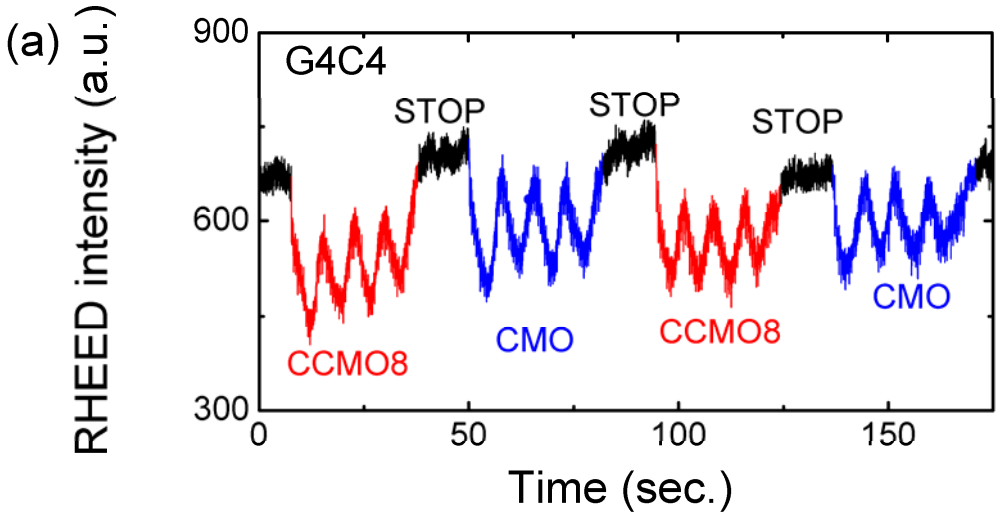


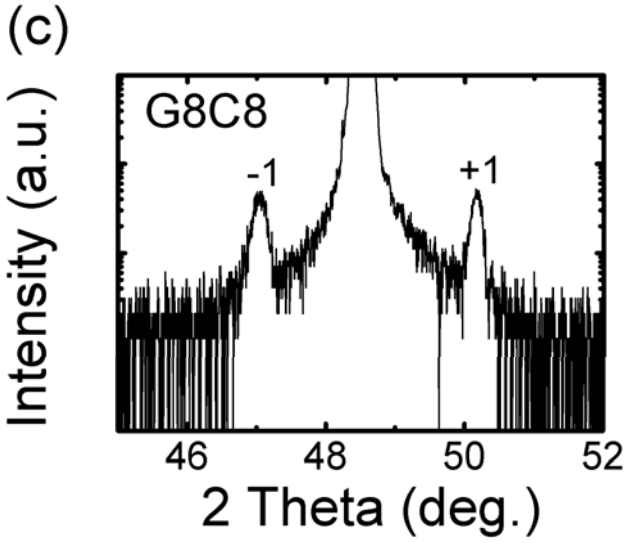





KrF Laser
NAO
e-
RHEED
CaMnO3
Ca1-xCexMnO3
(x=0.08)

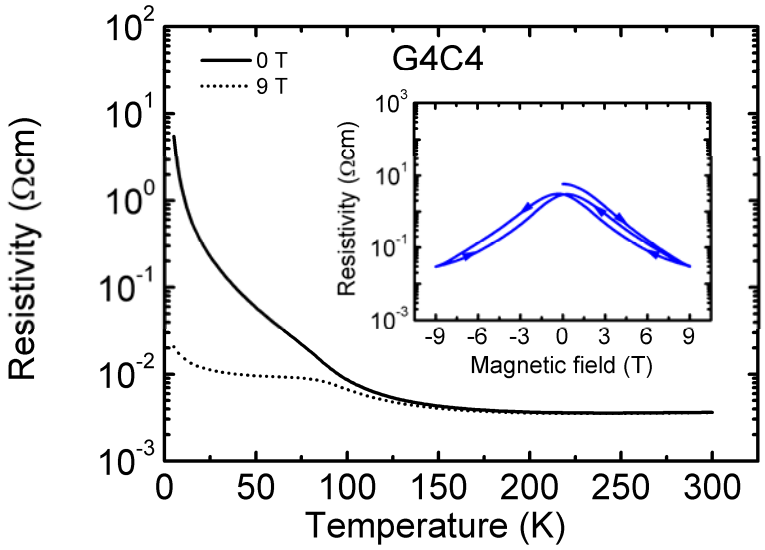

CCMO8
CMO
CMO
CMO@5T
CCMO8
CCMO8@5T
Resistivity (Ωcm)
Temperature (K)


(a)
G4C4
STOP
STOP
STOP
CCMO8
CMO
CCMO8
CMO
RHEED intensity (a.u.)
Time (sec.)

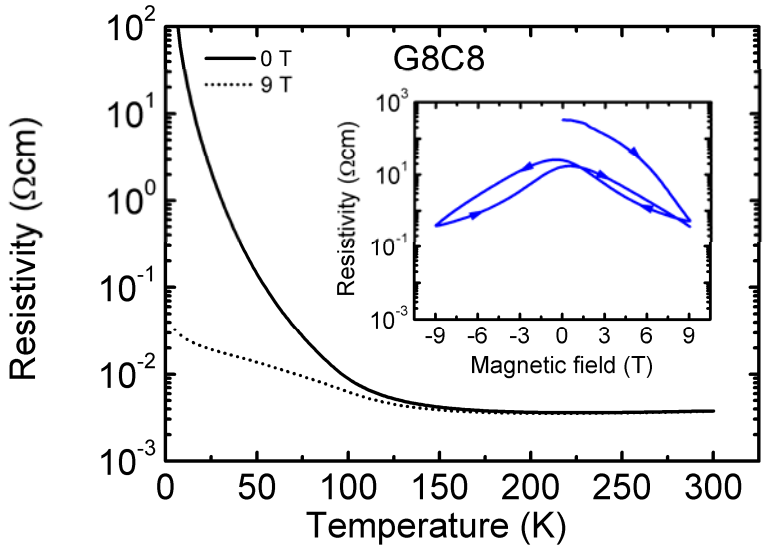

(c)
G8C8
-1
+1
Intensity (a.u.)
2 Theta (deg.)

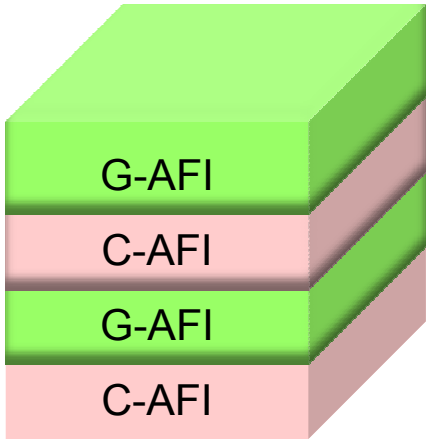
[CMO(x=0) × m/CMO(x=0)) × n]

G-AFI

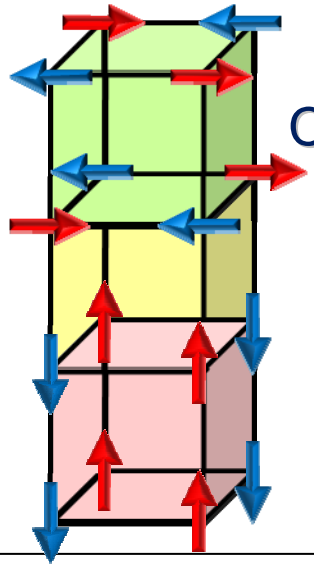


C-AFI





interface

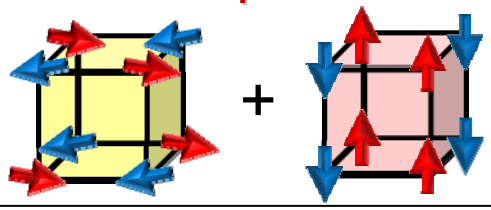


Charge transfer +

Competition of different spin structures at interfaces

⇩

Phase separation?



まとめ

強相関酸化物ヘテロ界面

・電荷移動　・・・・　限定的（？）
　　キャリア濃度は界面でシャープに変化
　　界面1ユニットセル程度の領域だけでキャリア濃度が変化
・電子相競合
　　強相関電子系の効果
　　・・・・　軌道（格子）、スピンの協調と競合

↓

界面電子相・新機能

今後

・界面の局所構造・電子状態（価数、スピン状態など）の詳細な評価
　　シンクロトロンX線実験，透過電子顕微鏡観察
・界面電子相競合状態の外場（電場、光など）による制御　・・・・　機能化