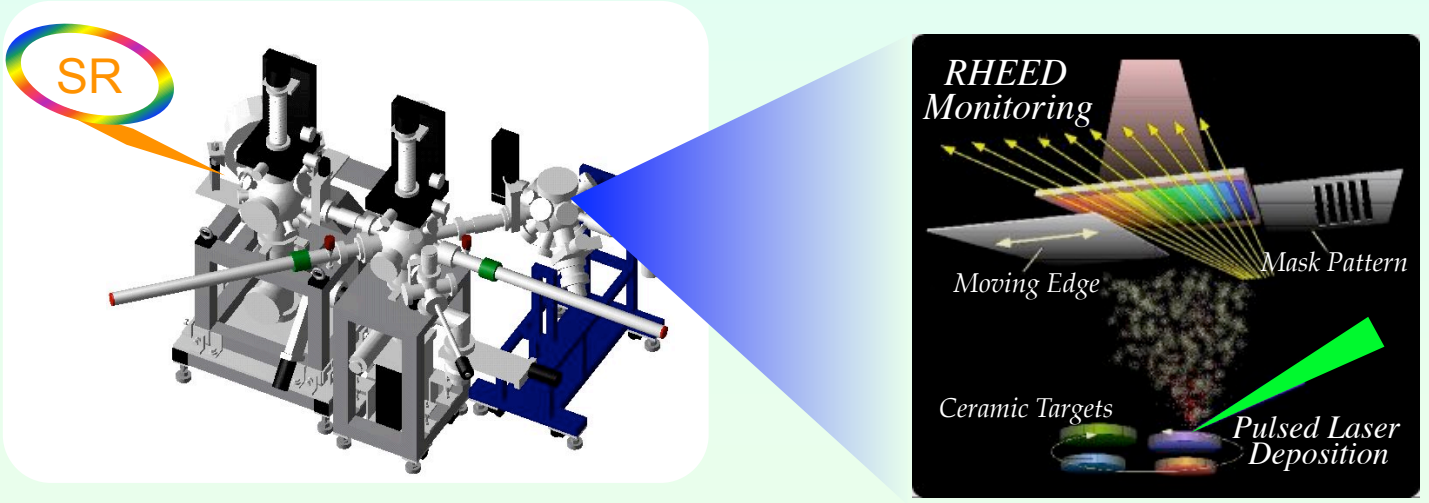
PF研究会「磁性薄膜・多層膜を究める：キャラクタリゼーションから新奇材料の創製へ」
2011年10月15（14-15）日@KEK-PF小林ホール

放射光光電子分光による$La_{0.6}Sr_{0.4}MnO_3/SrTiO_3$スピントンネル接合界面の電子状態解析

組頭　広志
KEK-PF（物構研＆構造物性研究センター）
JSTさきがけ



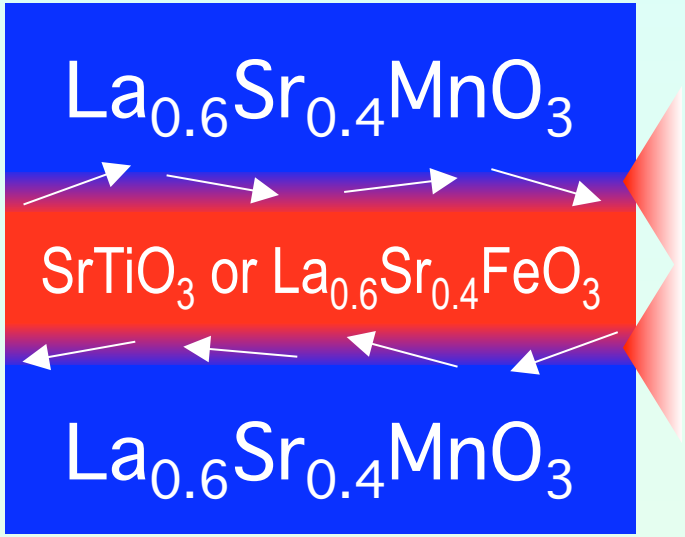
Motivation

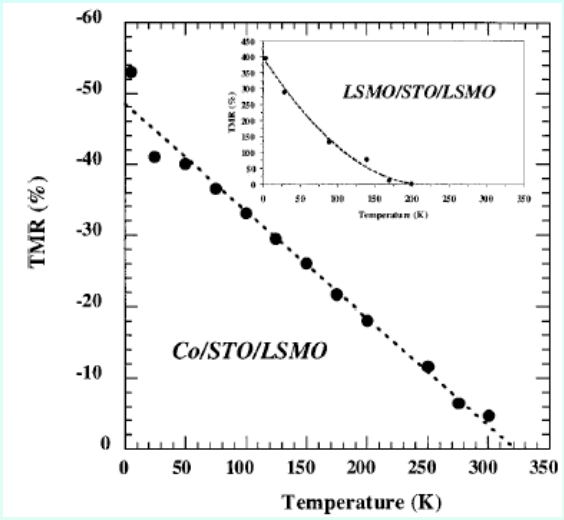
$La_{0.6}Sr_{0.4}MnO_3$

Half Metallic

Tc ~ 370K

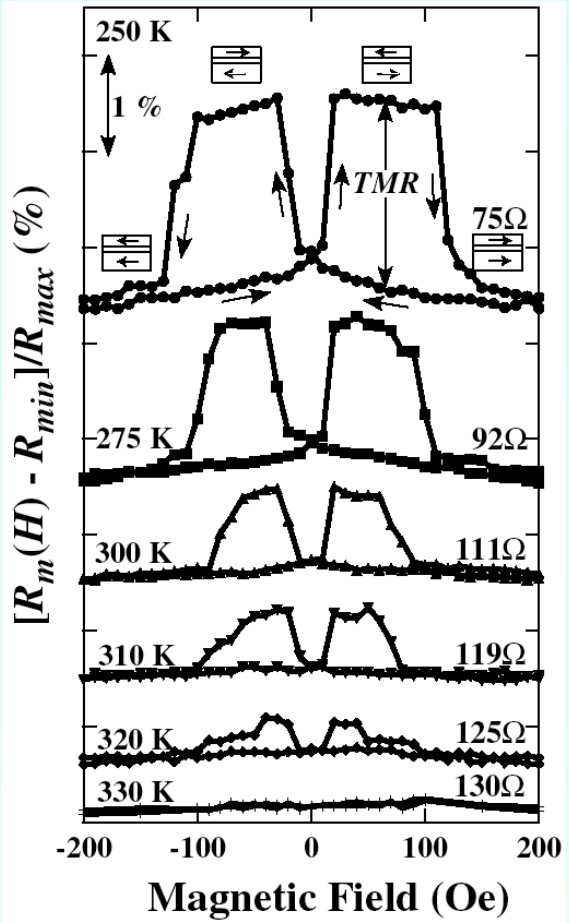
Spin Tunneling Junctions





F. Pailloux *et al.*, PRB **66**, 014417 (02).

Disappearance of TMR@R.T.



Y. Ogimoto *et al.*, JJAP **42**, L369 (03)

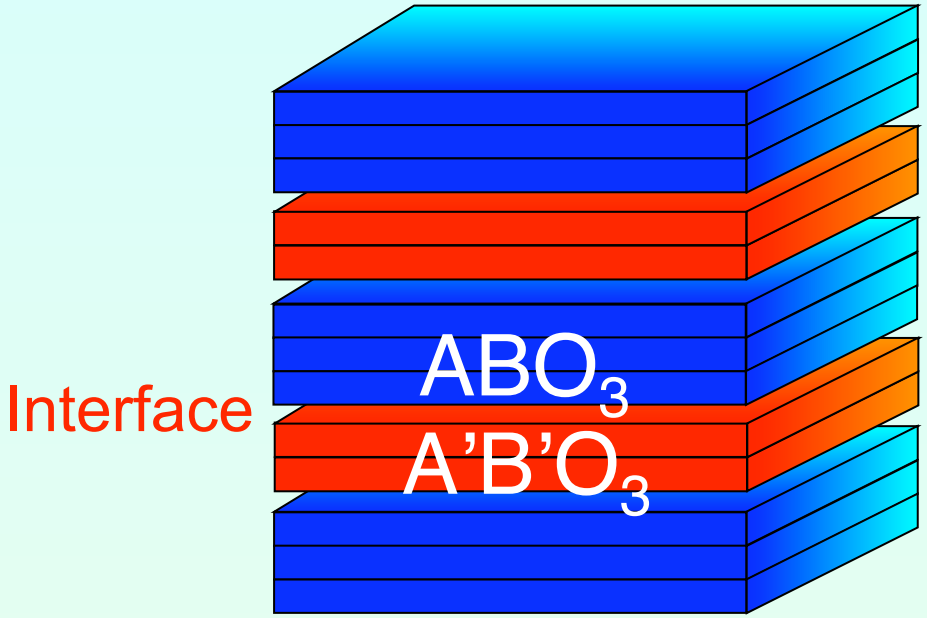
In actual TMR devices, the performance is far worse than what is expected from these physical properties.

Small TMR ratio

Magnetic "dead layer" formation at the STO/LSMO interface

Charge Transfer at the interface

Superlattice based on Transition Metal Oxides



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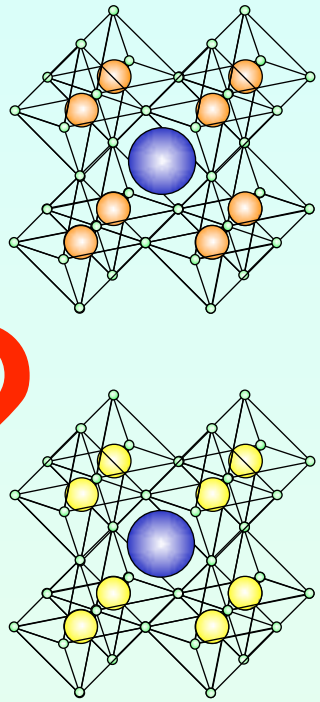
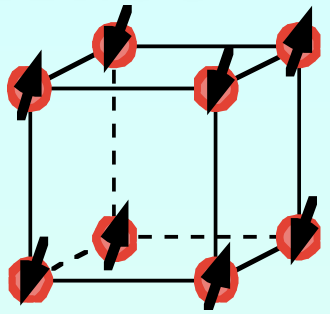
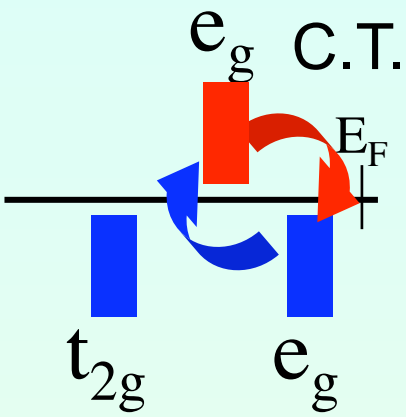
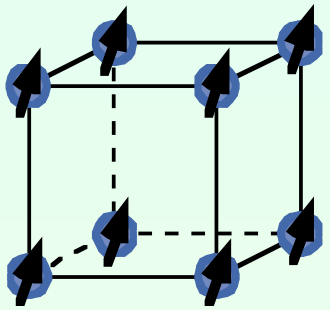








It is indispensable to reveal the electronic structure at the heterointerface

"in-situ Photoemission"

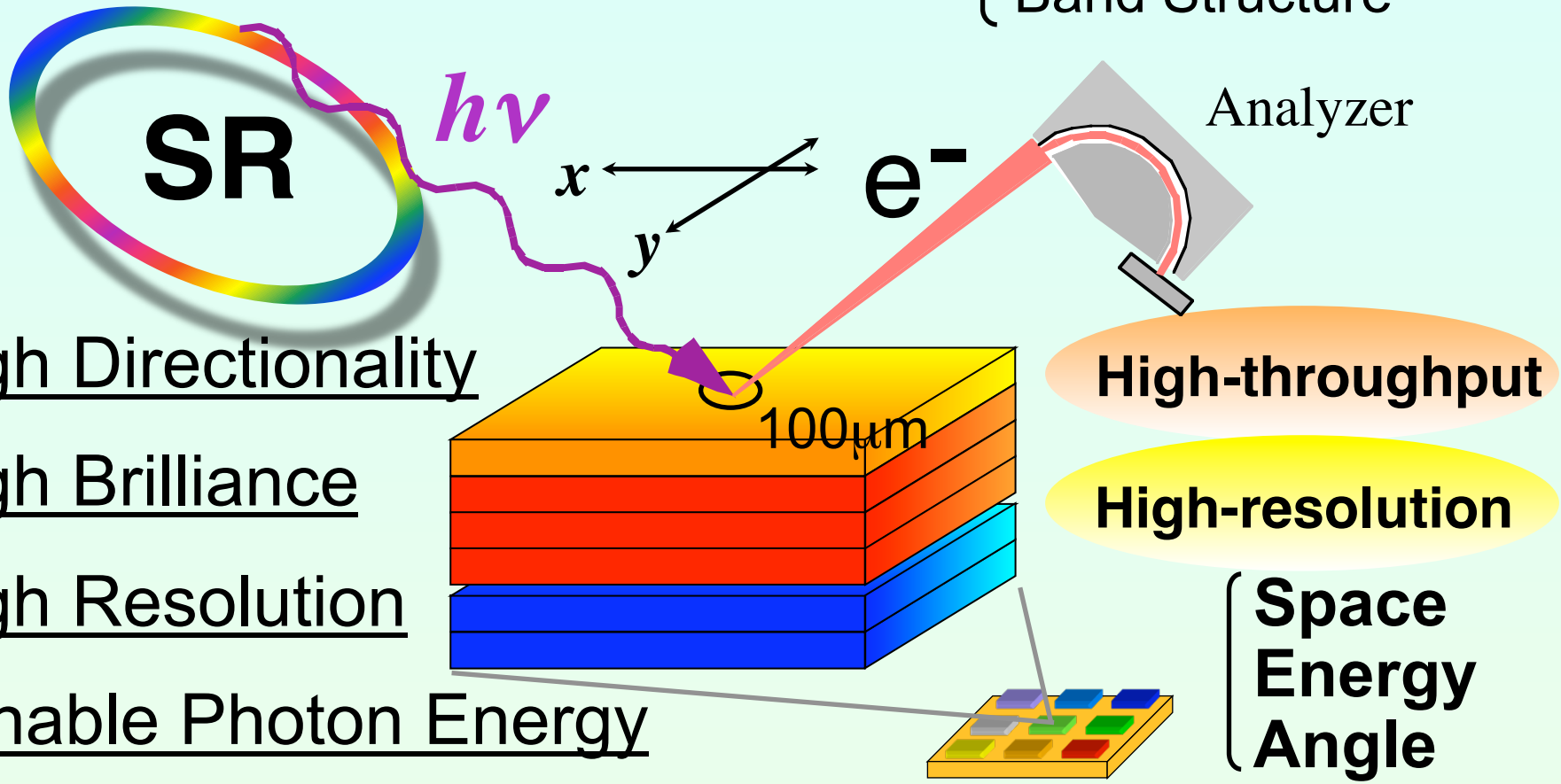
Advantage of SR-PES

☆Non-destructive

☆Surface (Interface) Sensitive （5～30Å）

☆Direct Determination of Electronic States
- Chemical Shift
- DOS
- Band Structure

Elemental Selectivity



High Directionality

High Brilliance

High Resolution

Tunable Photon Energy

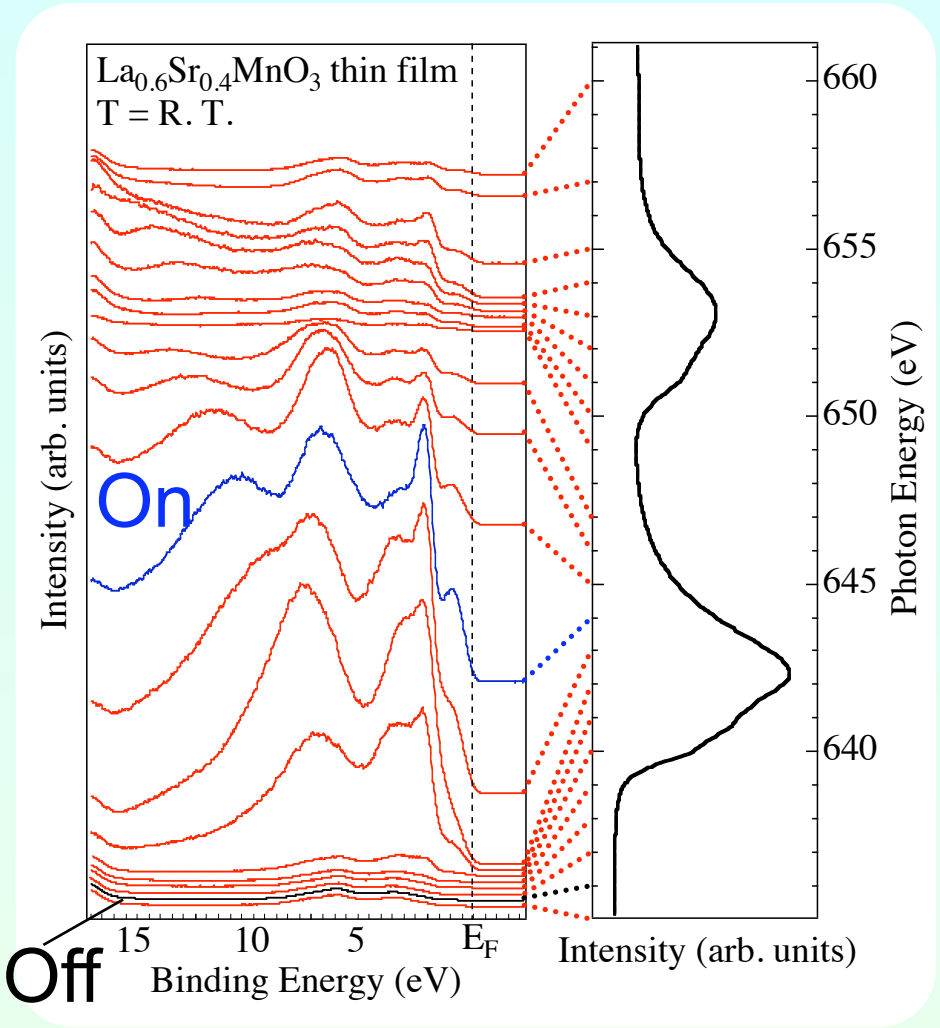
High-throughput

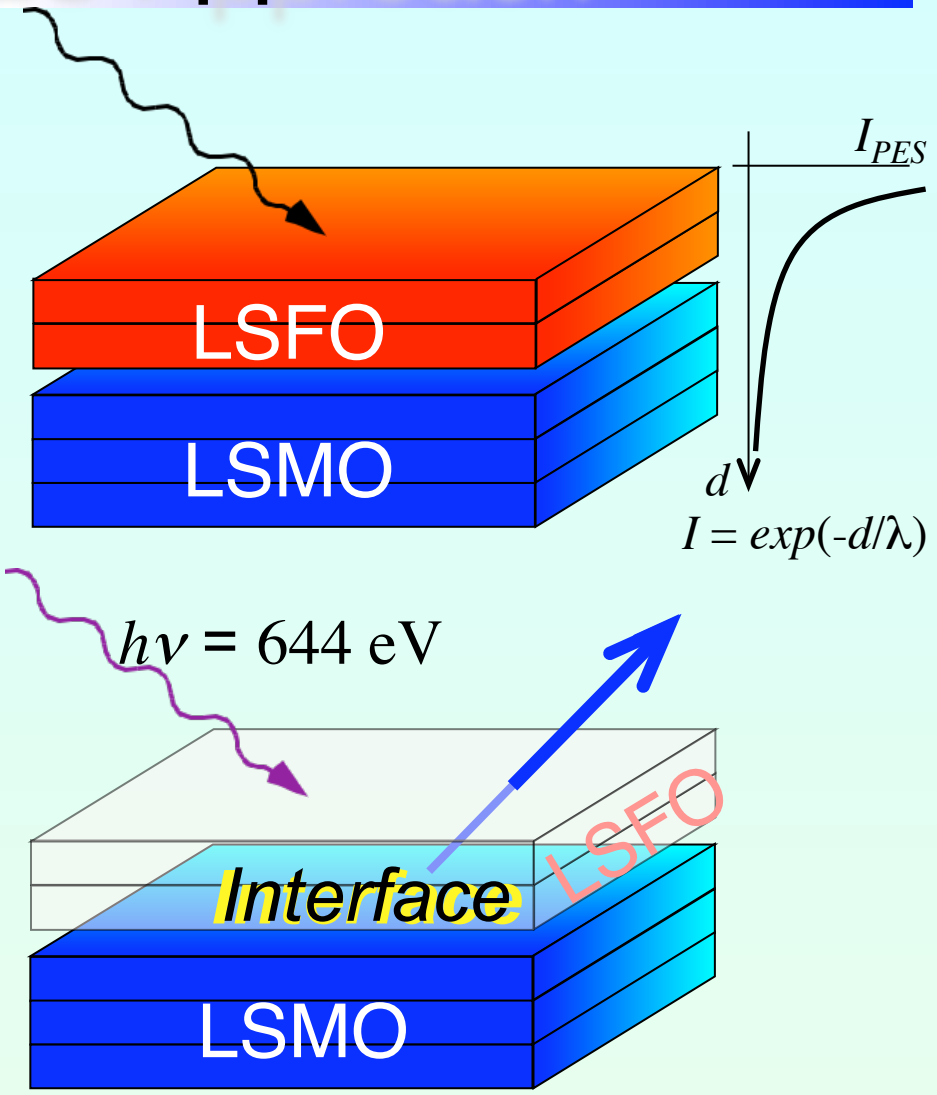
High-resolution

Space
Energy
Angle

Resonant PES Approach

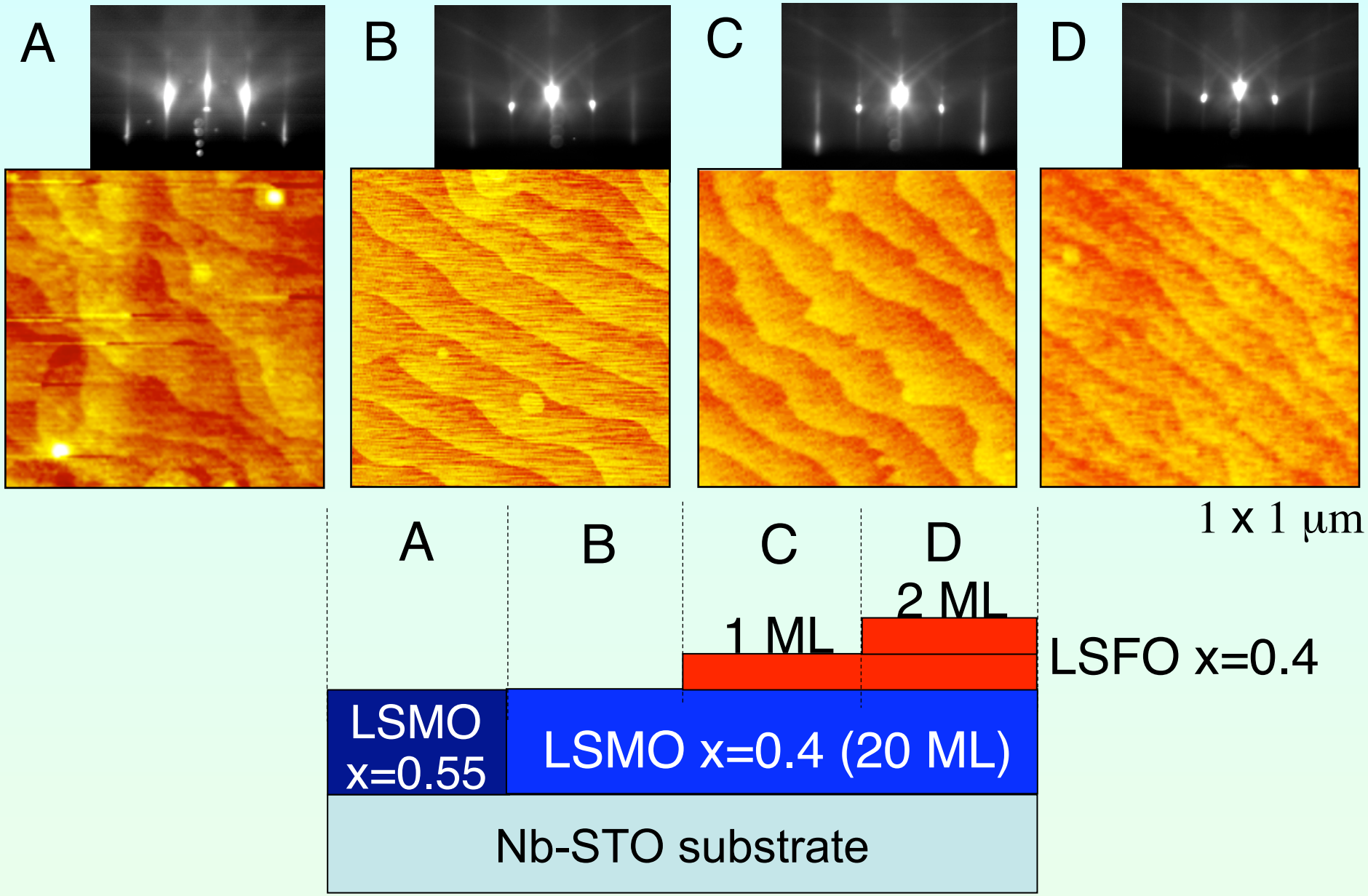
Resonant PES of LSMO (x=0.4)





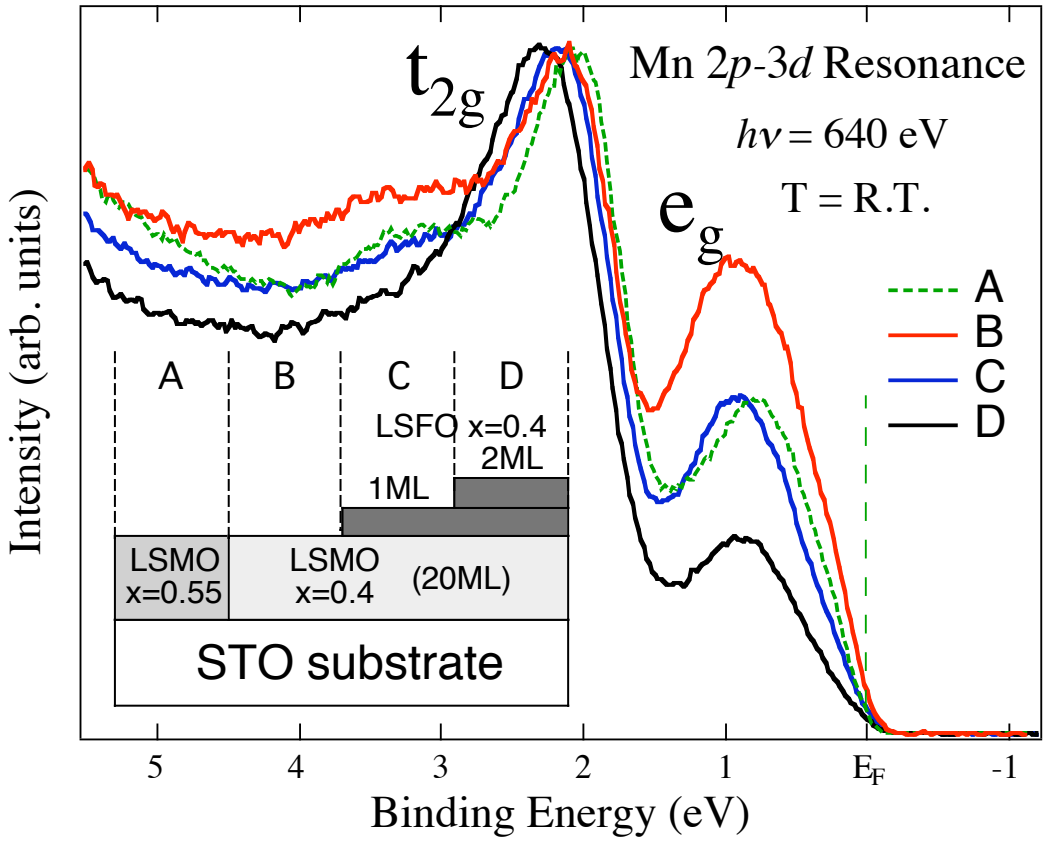
Probing Electronic Structure at the Interface (Mn 3*d* PDOS)

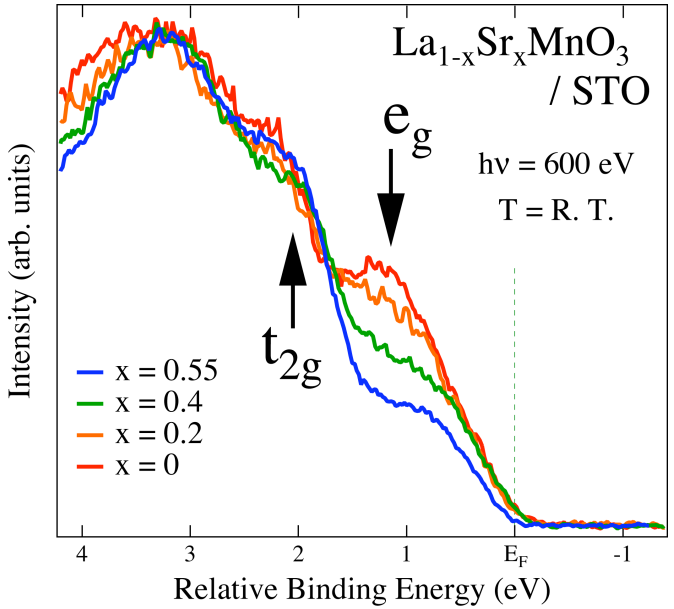
RHEED Pattern & AFM Images



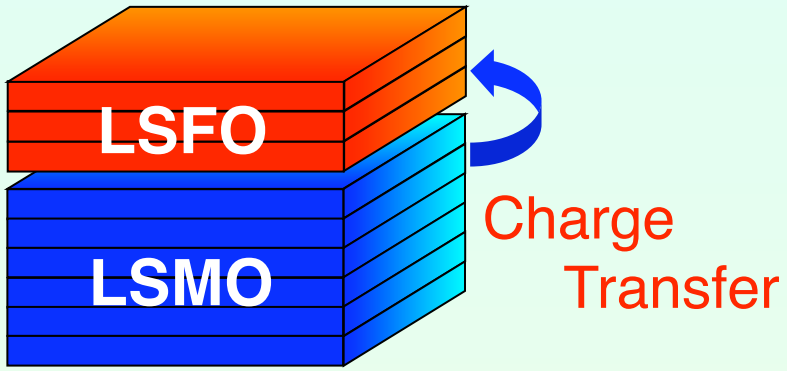
Resonant PES of LSMO at Interface

Mn 2*p*-3*d* Resonant PES





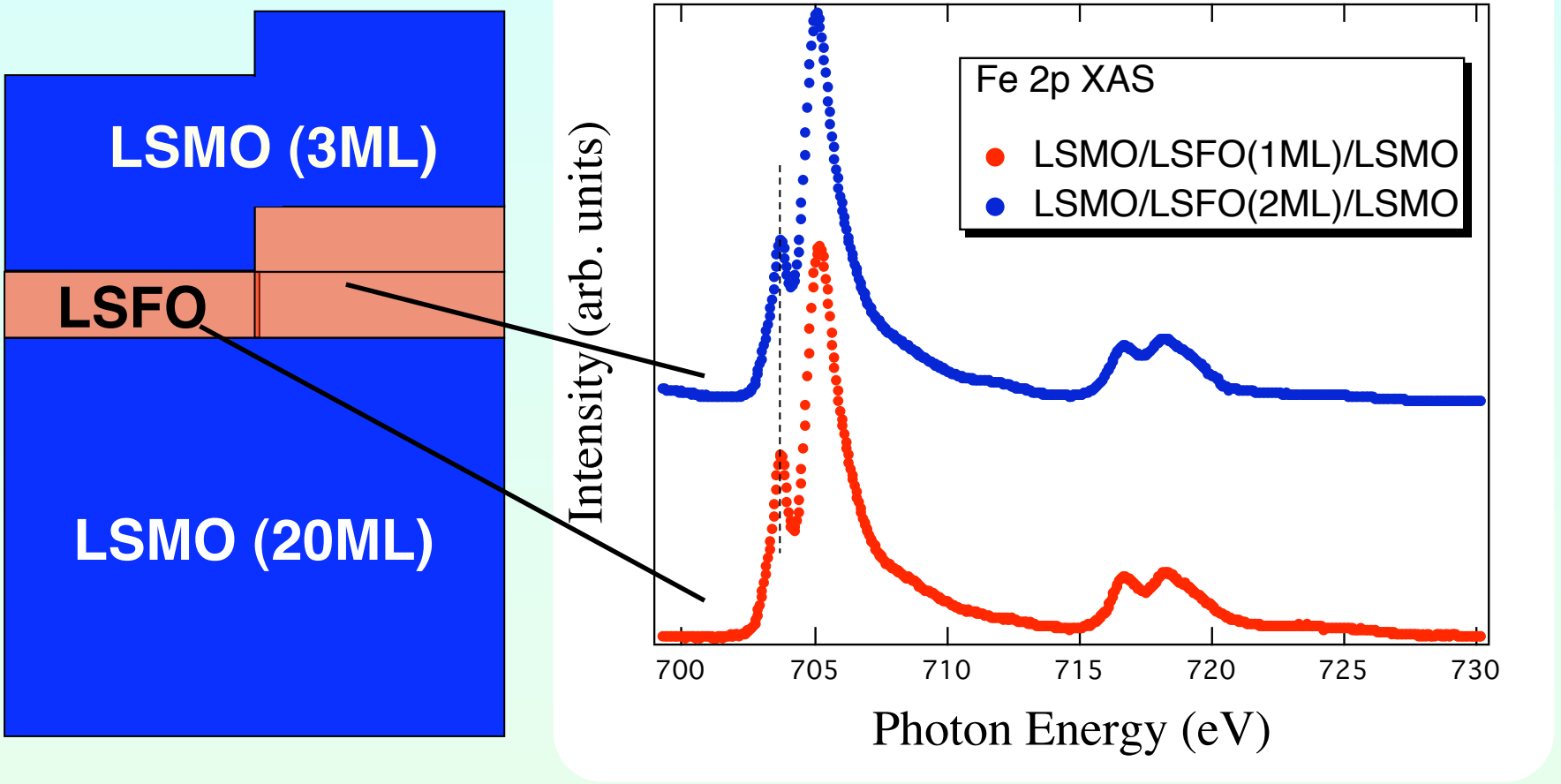
K. Horiba, H.K. *et al*., Phys. Rev. B **71**, 155420 ('05)



Spectral Evidence of Charge Transfer at LSMO/LSFO Interface

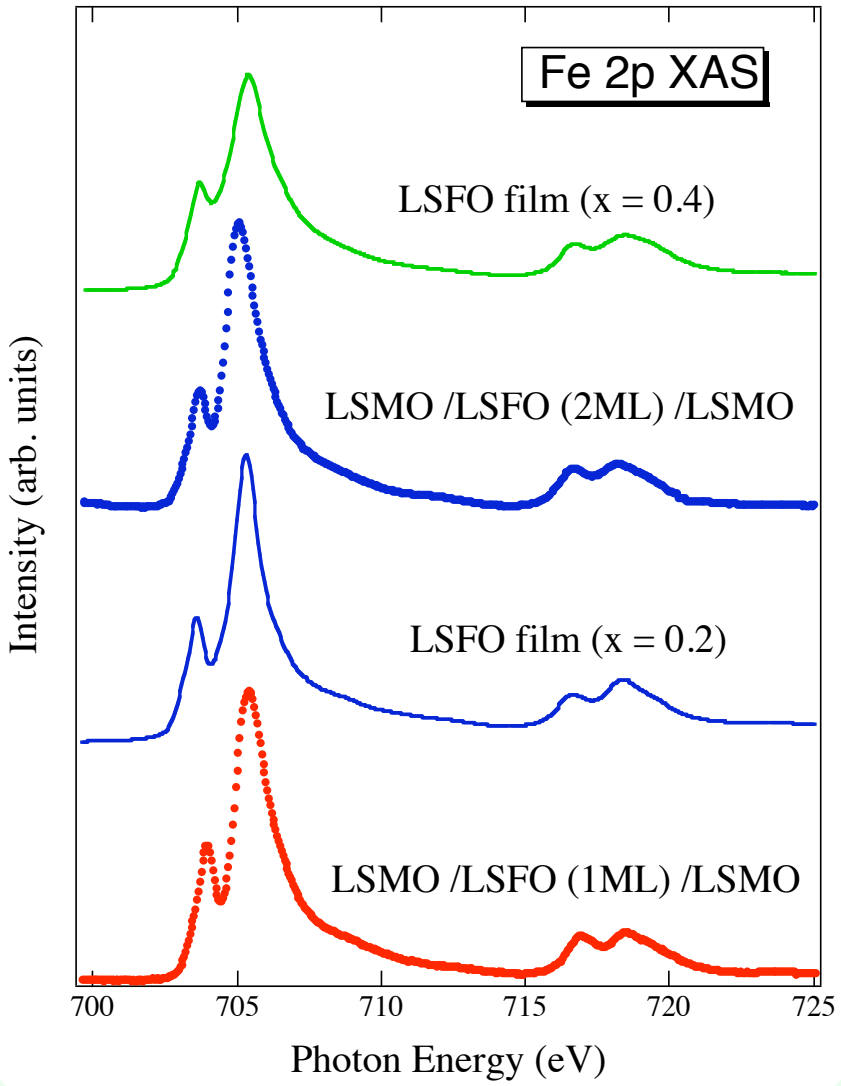
H. Kumigashira *et al*., *Appl. Phys. Lett.* **84**, 5353 (2004).

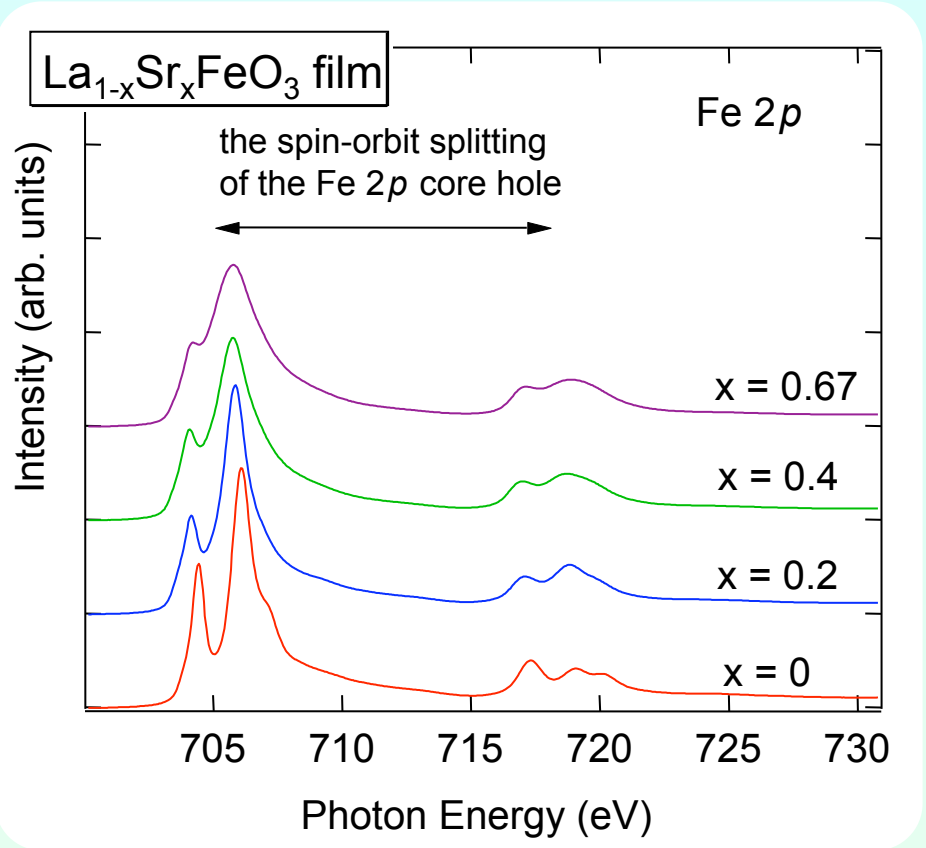
XAS Spectra of Interfacial LSFO Layer



Comparison of Fe-2*p* XAS spectra between Interfacial LSFO layer and LSFO films

Fe-2*p* XAS spectra



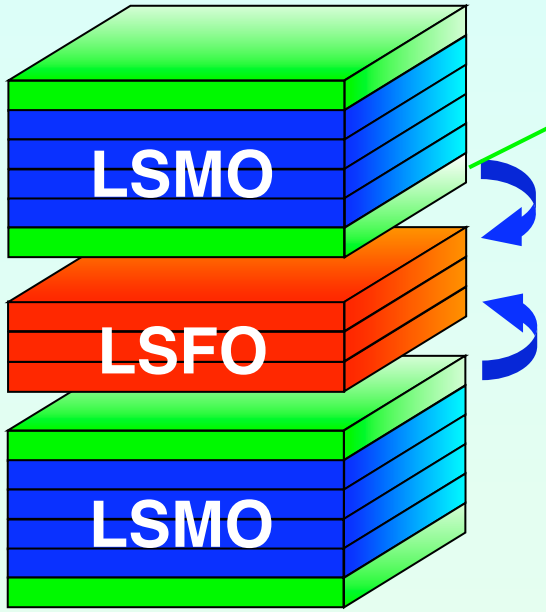


H. Wadati, H.K. *et al.*, Phys. Rev. B **71**, 035108 (2005).

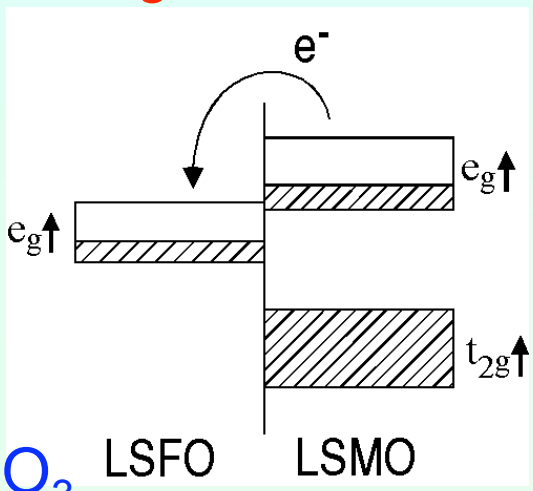
Evidence of Charge Transfer from LSMO to LSFO

Charge-Transfer at Interface

Interface in LSMO/LSFO superlattices

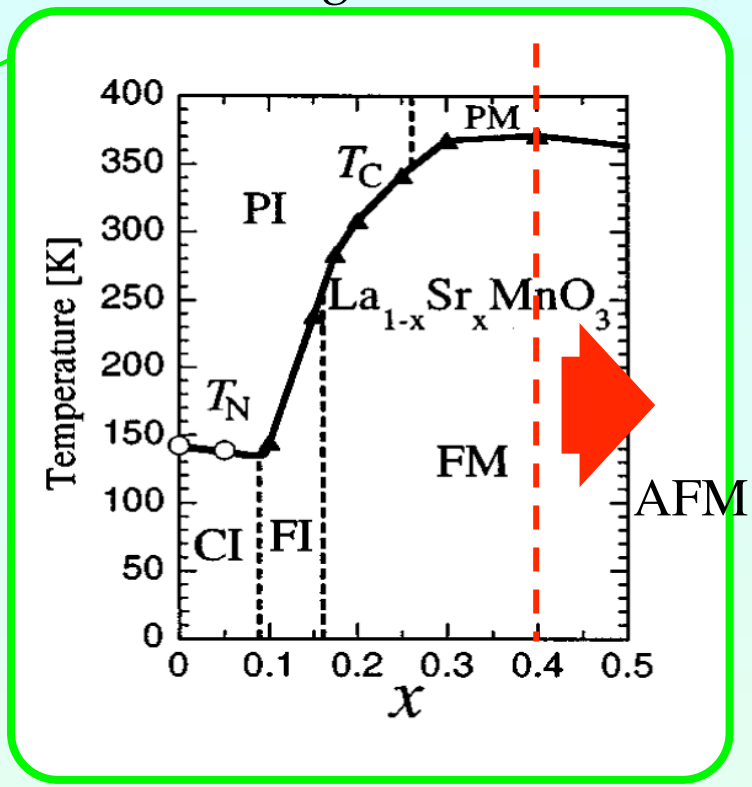


Charge Transfer



$(La_{0.6}Sr_{0.4})BO_3$

Phase diagram of LSMO

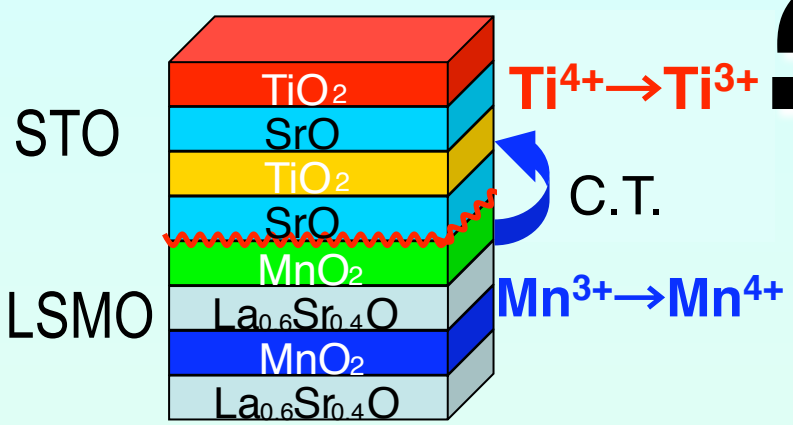


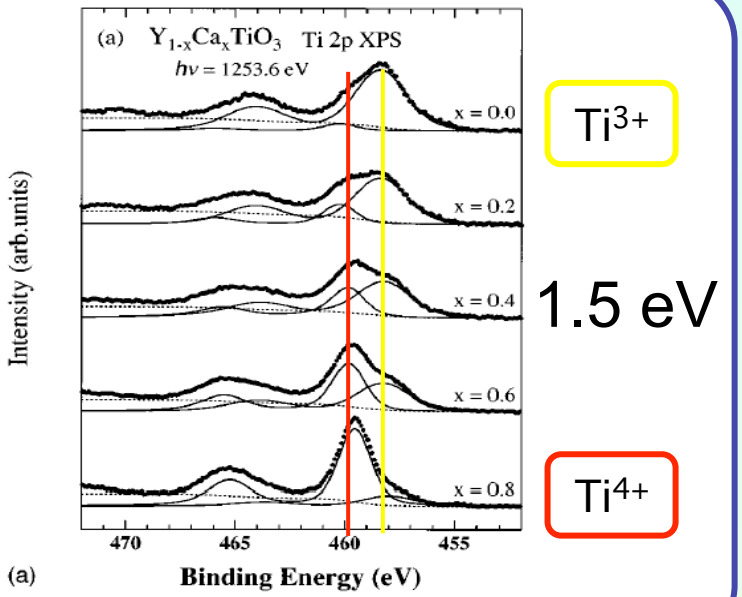
A. Urushibara *et al*., Phys. Rev. B **51**, 14103 (1995).

AFM or PM dead-layer formation at Interface

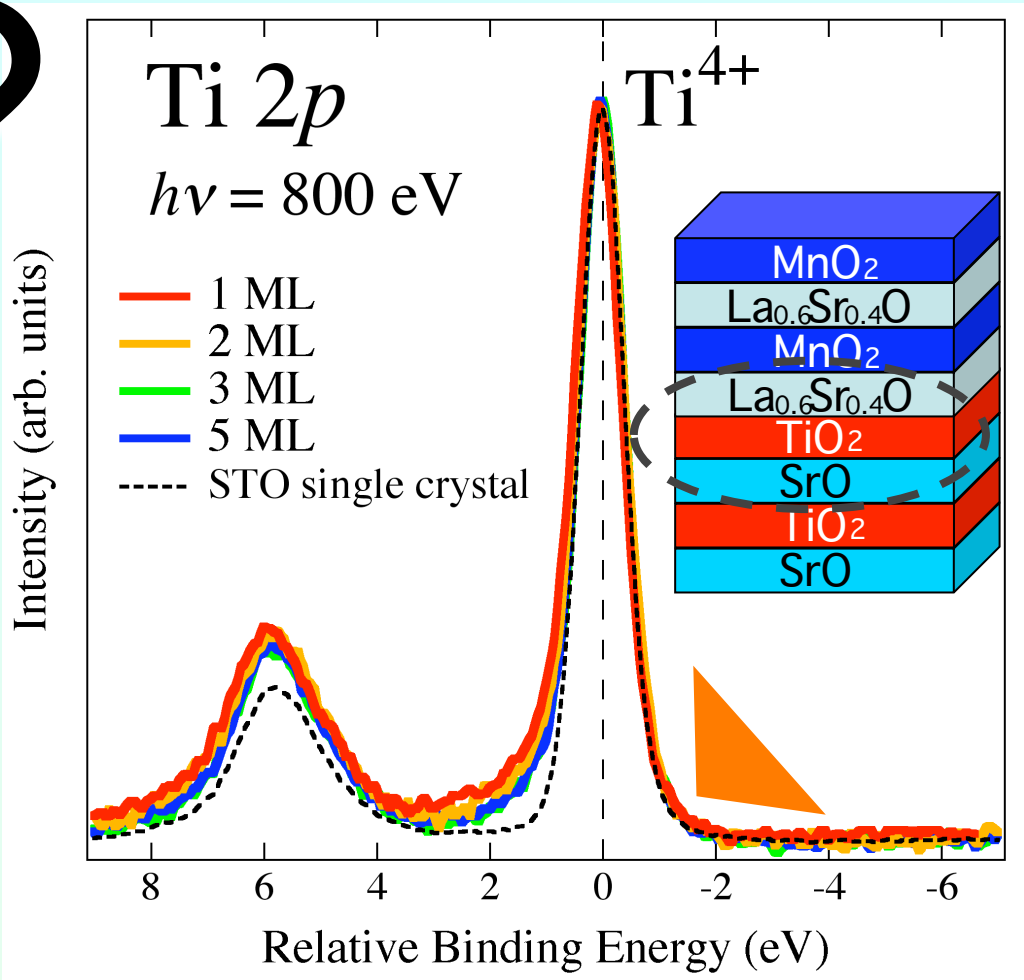
Ti 2*p* Core Level Spectra at Interfaces

Charge Transfer ?





K. Morikawa *et al.*, Phys. Rev. B **54**, 5446 (1996).

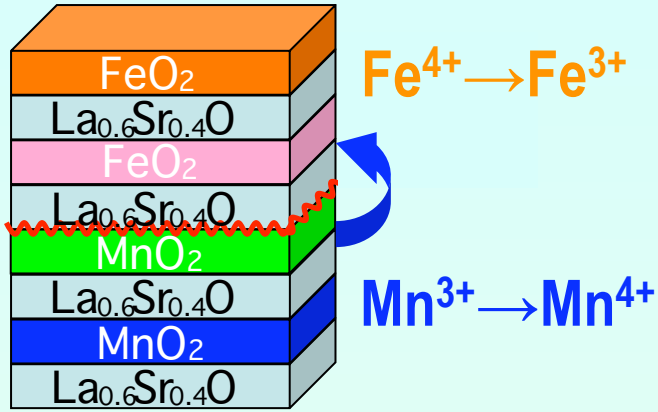


H. Kumigashira *et al.*, Appl. Phys. Lett. **88**, 192504 ('06).

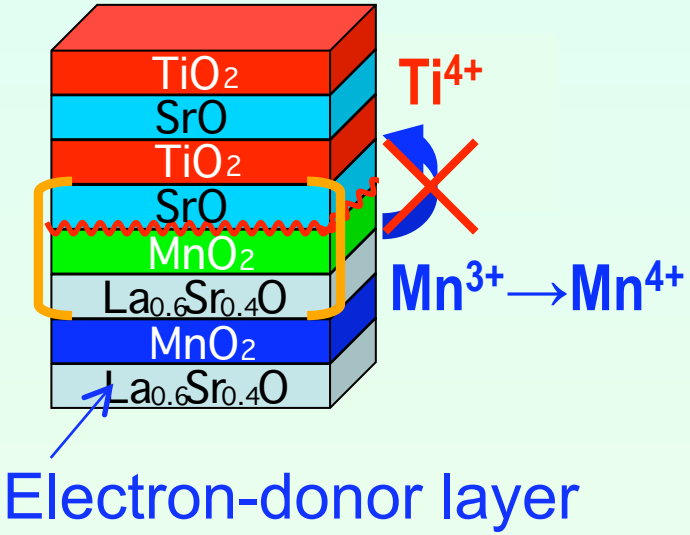
Robust Ti^{4+} states

Origin of Charge Transfer

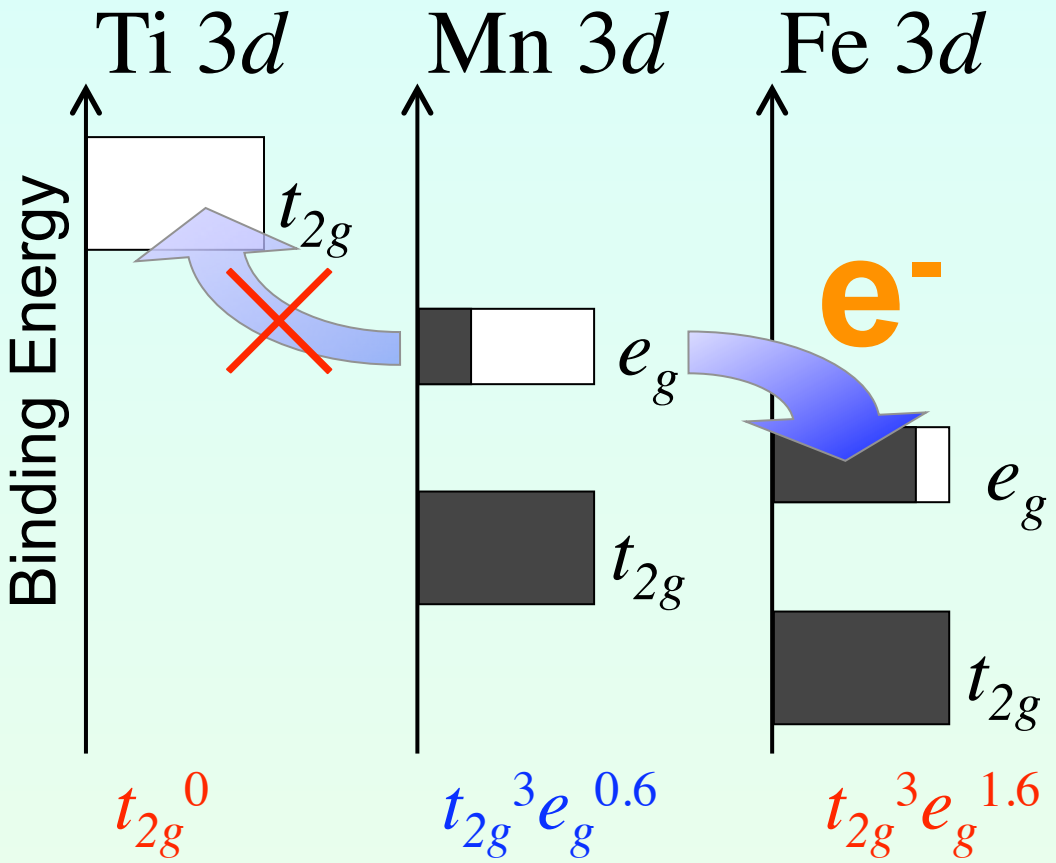
LSFO/LSMO



STO/LSMO



Difference of 3*d* levels among transition metals

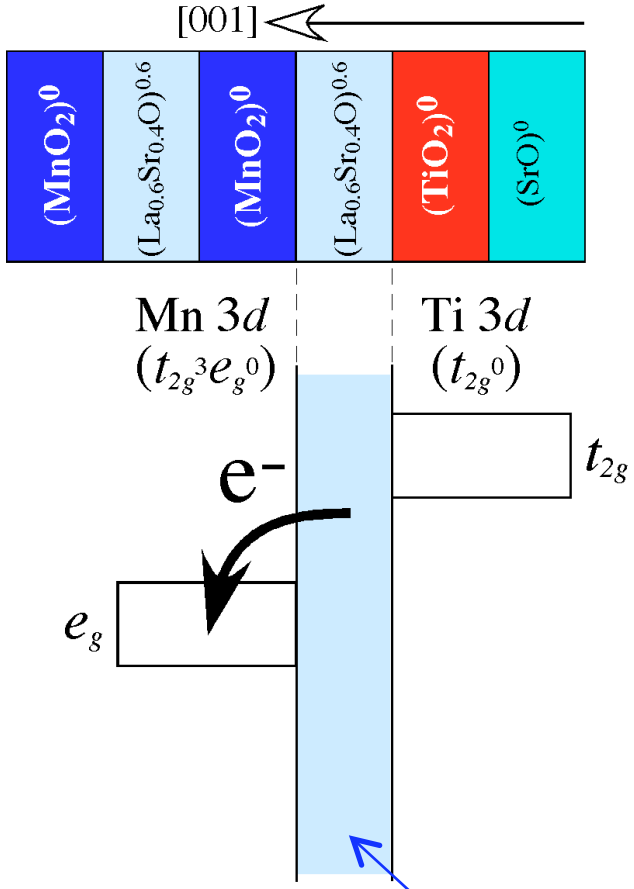


Charge redistribution at interface

C. T. from AO Layer to BO_2 Layer

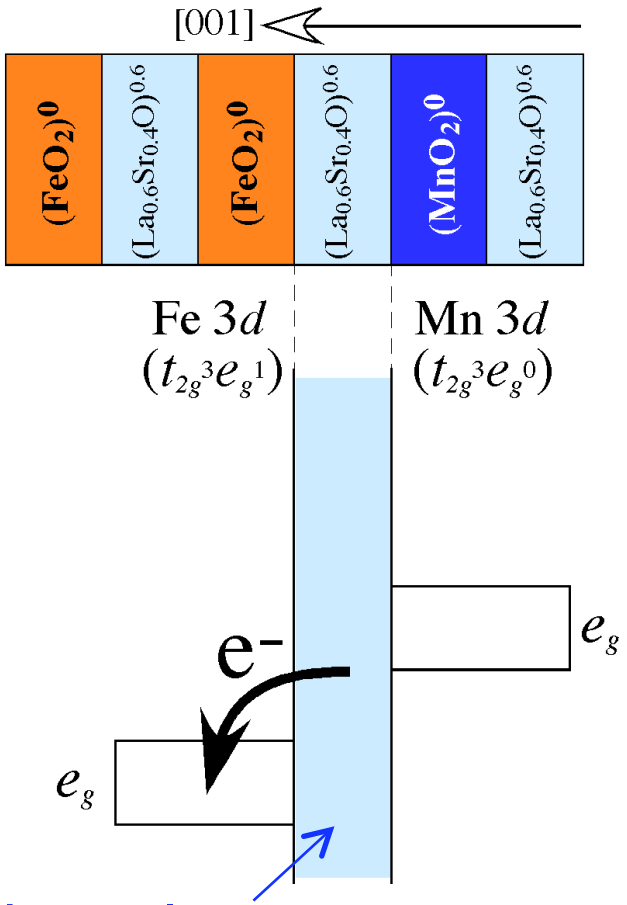
LSMO/STO interface

(Valence mismatch)



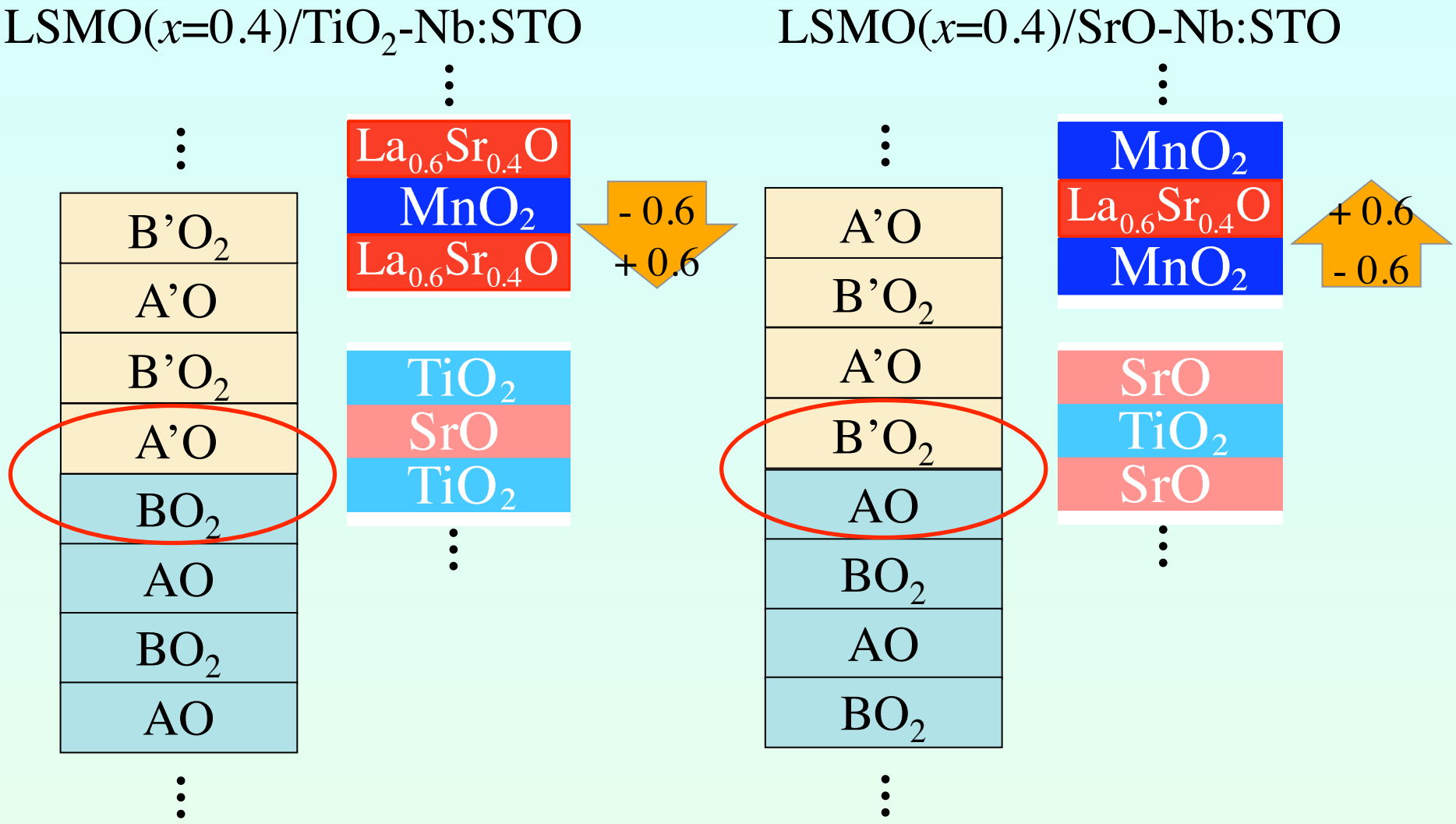
LSFO/LSMO interface

(Charge transfer between TM ions)



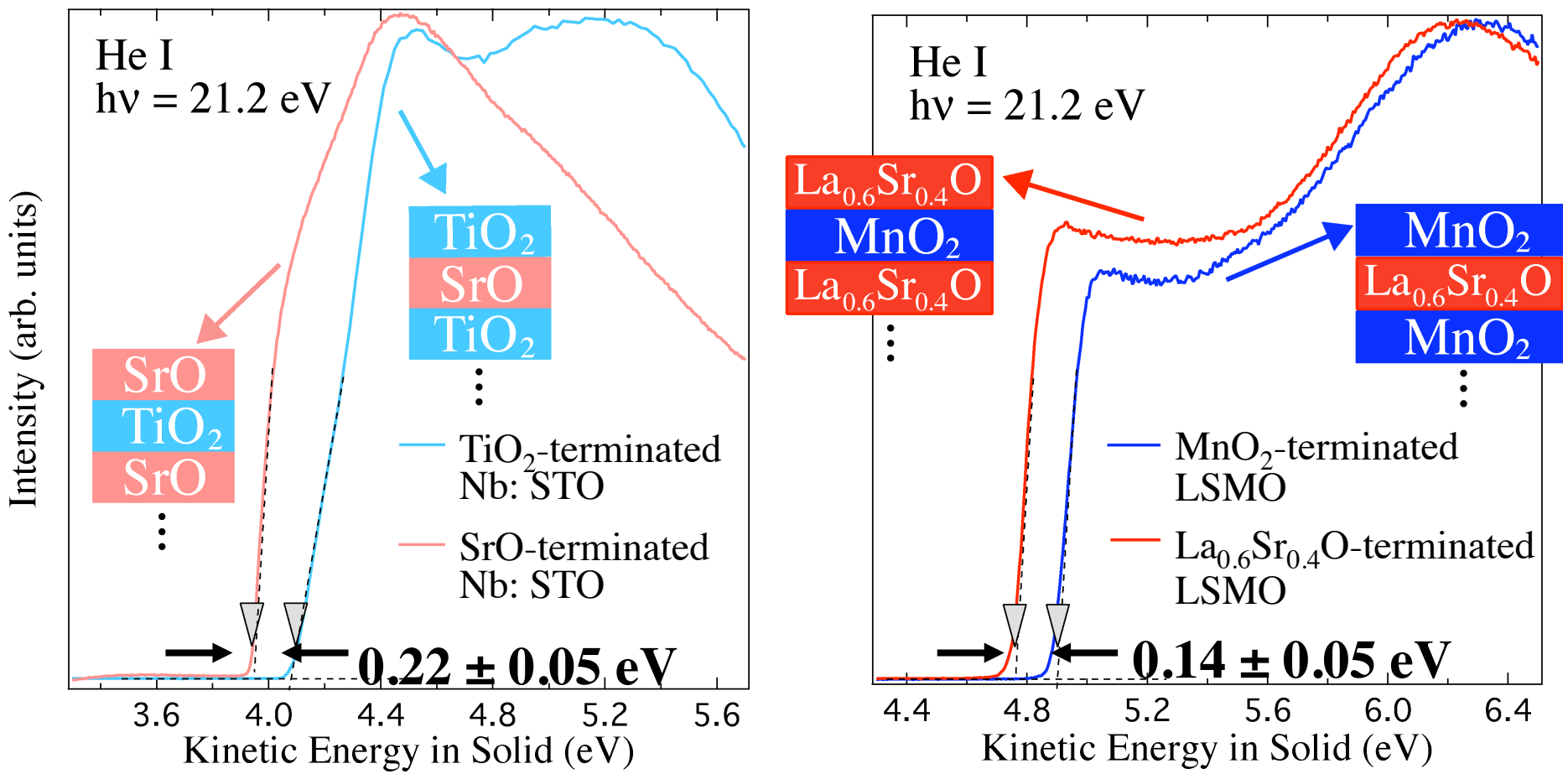
$La_{0.6}Sr_{0.4}O$ electron-donor layer

$A'B'O_3/ABO_3$ interface: $A'B'O_3/BO_2$-ABO_3 vs $A'B'O_3/AO$-ABO_3



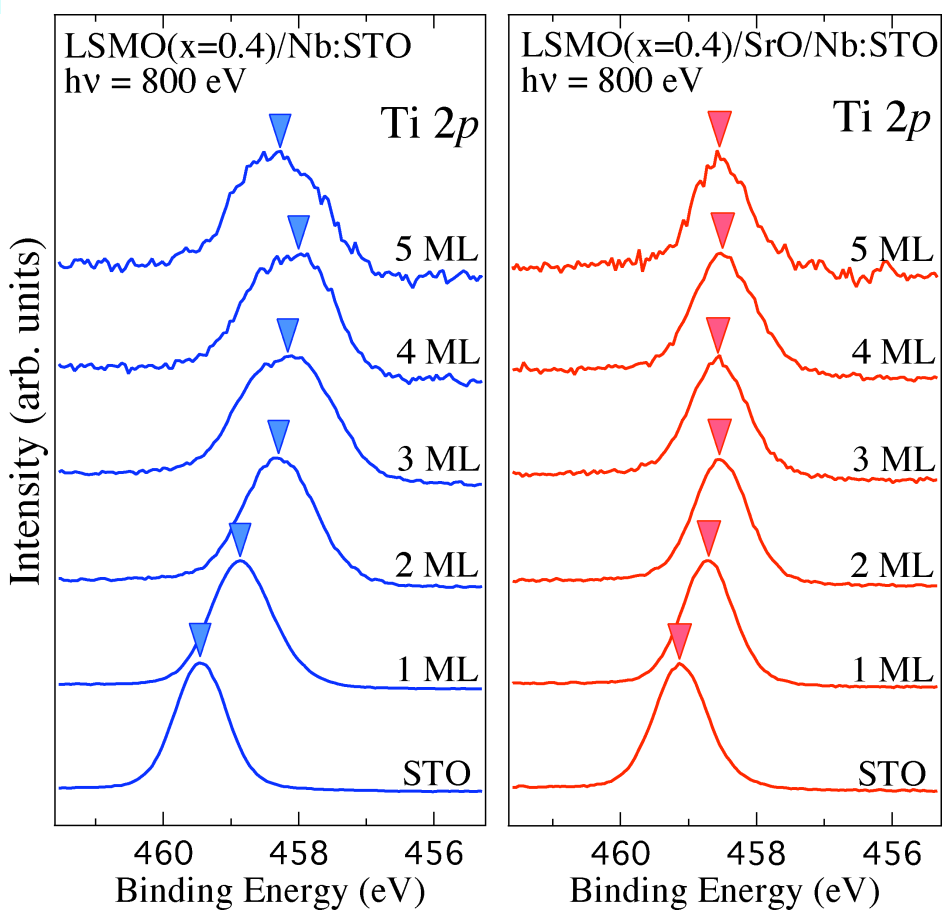
How does the band diagram modulates by changing the terminating layer at the interface?

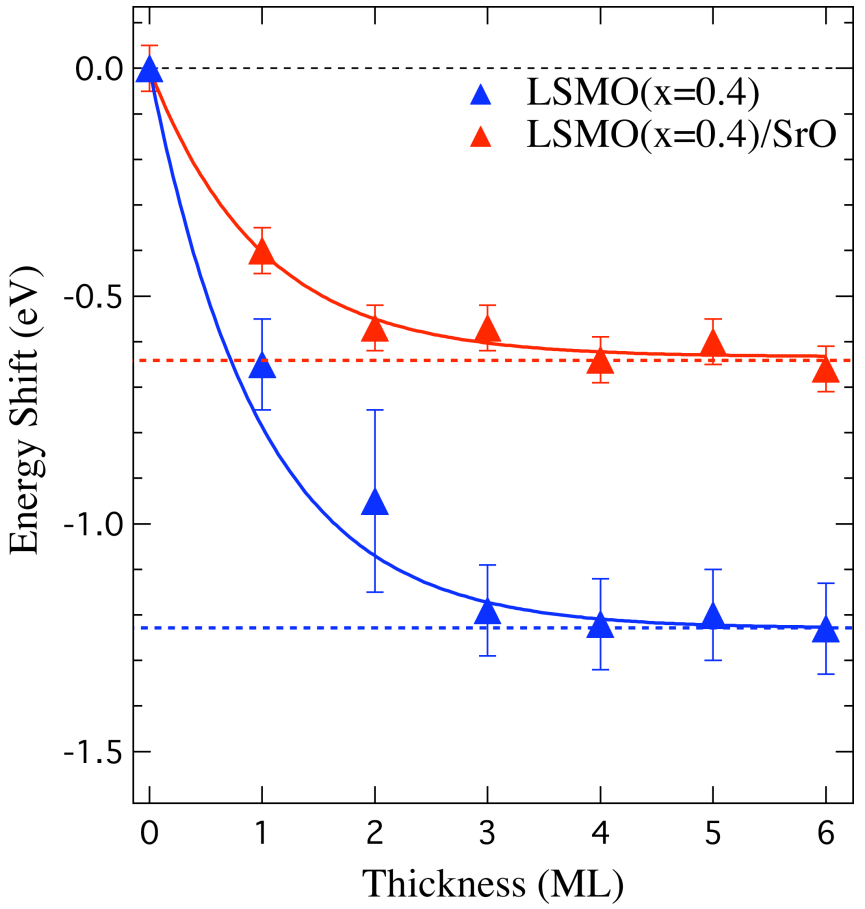
Work functions of termination controlled STO and LSMO



Termination	Nb:STO (χ_i)	LSMO(x=0.4) (ϕ_m)
BO_2	4.08 eV	4.87 eV
AO	3.86 eV	4.73 eV

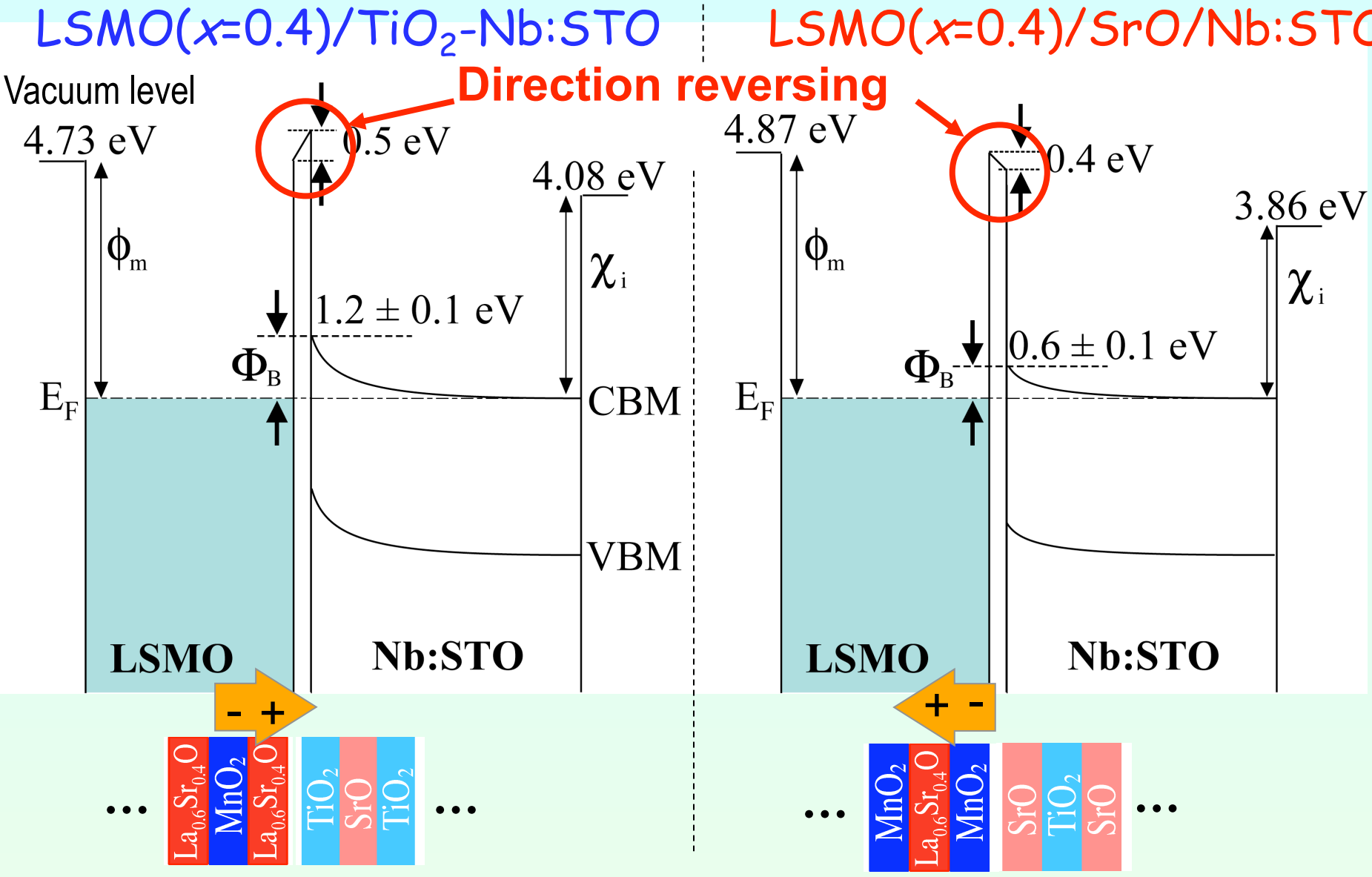
Built-in potential of termination controlled LSMO





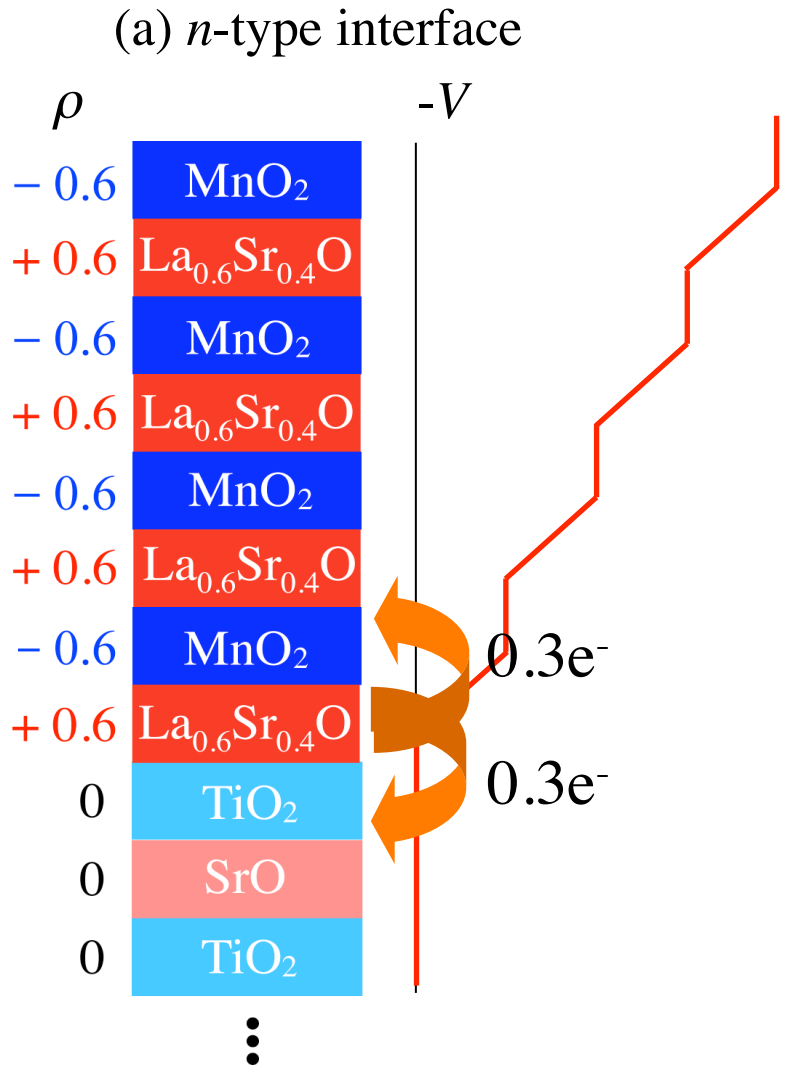
Heterojunctions	Φ_B [exp.]	$\phi_m - \chi_i$ [exp.]
LSMO/TiO_2-Nb:STO	1.2 ± 0.1 eV	0.65 eV
LSMO/SrO/Nb:STO	0.6 ± 0.1 eV	1.01 eV

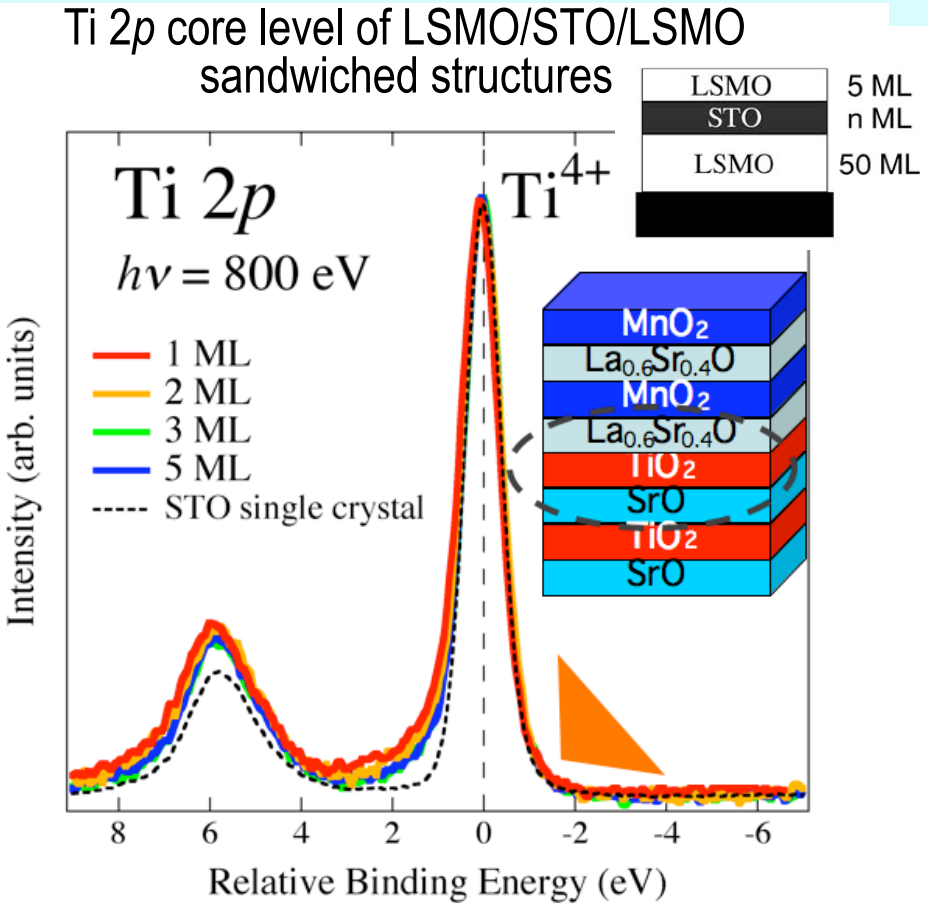
Band diagrams of termination controlled LSMO



The direction of interface dipole changes depending on the terminating layer.

Origin of the interface dipole

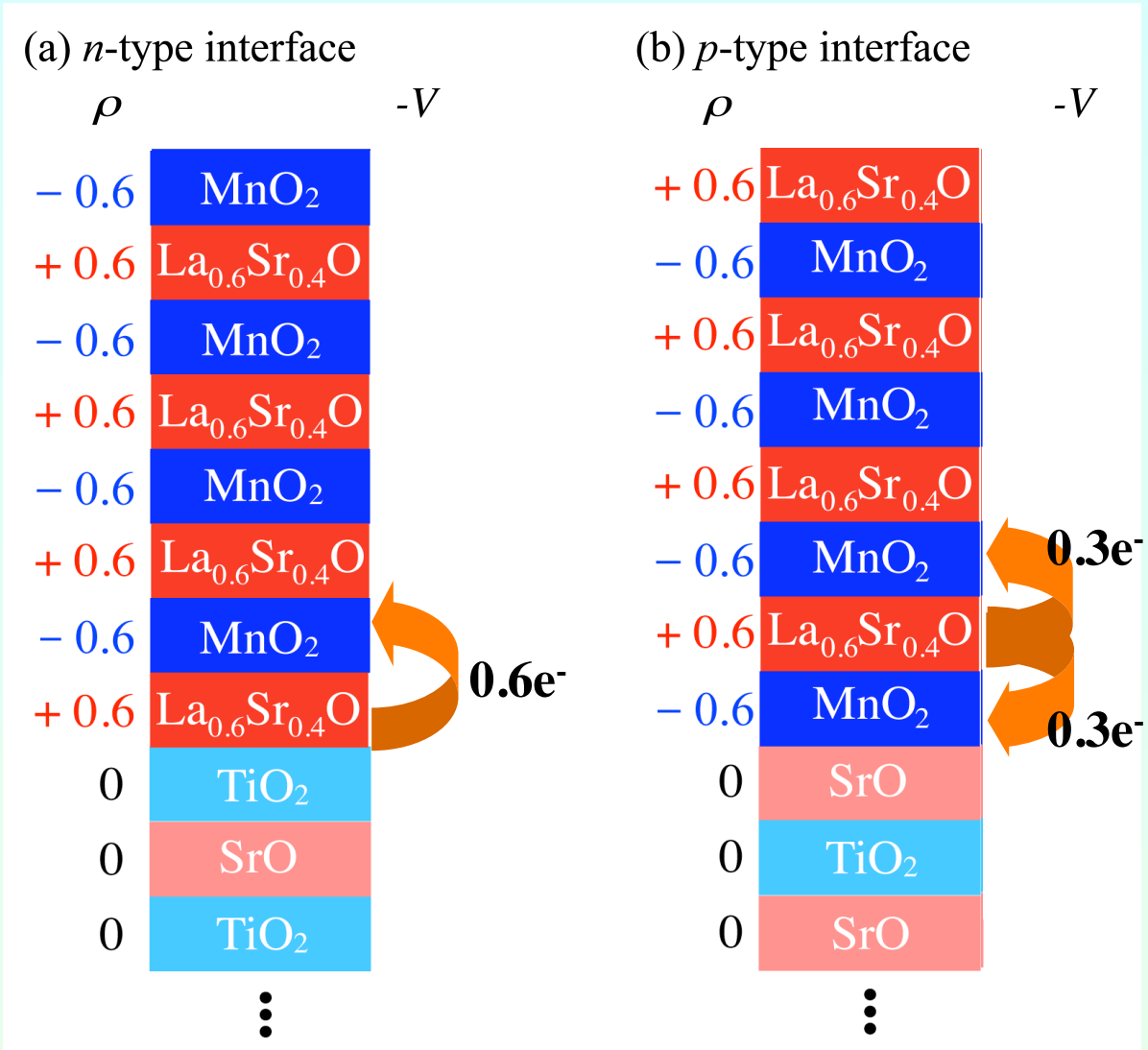




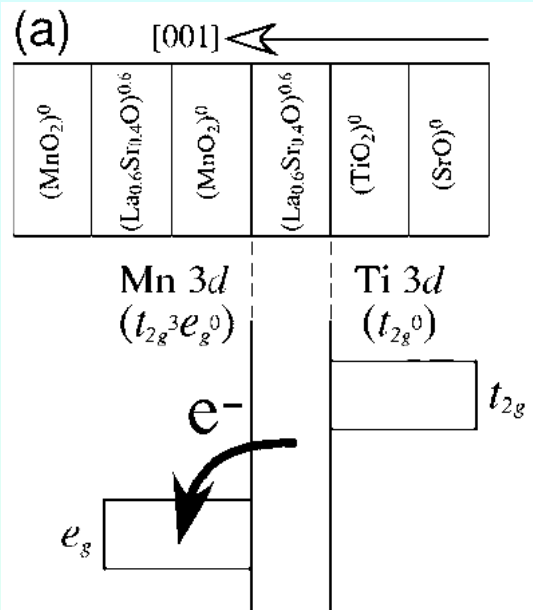
H. Kumigashira *et al.*, Appl. Phys. Lett. **88**, 192504 ('06).

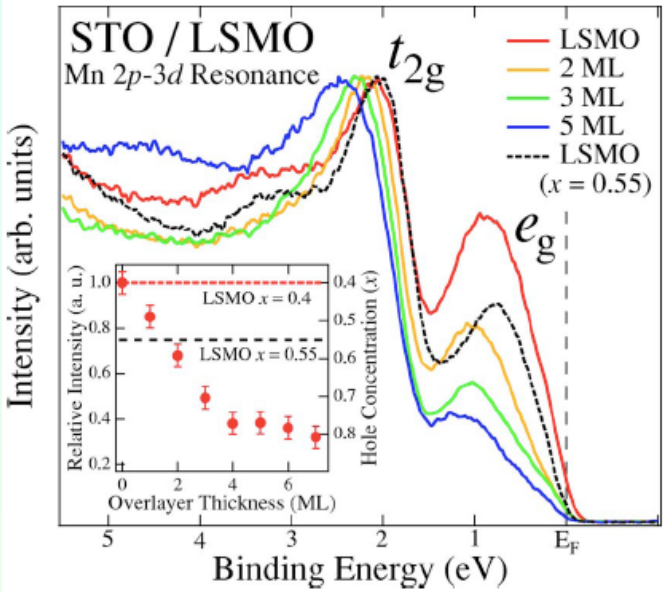
Robust Ti^{4+} states

Origin of the interface dipole



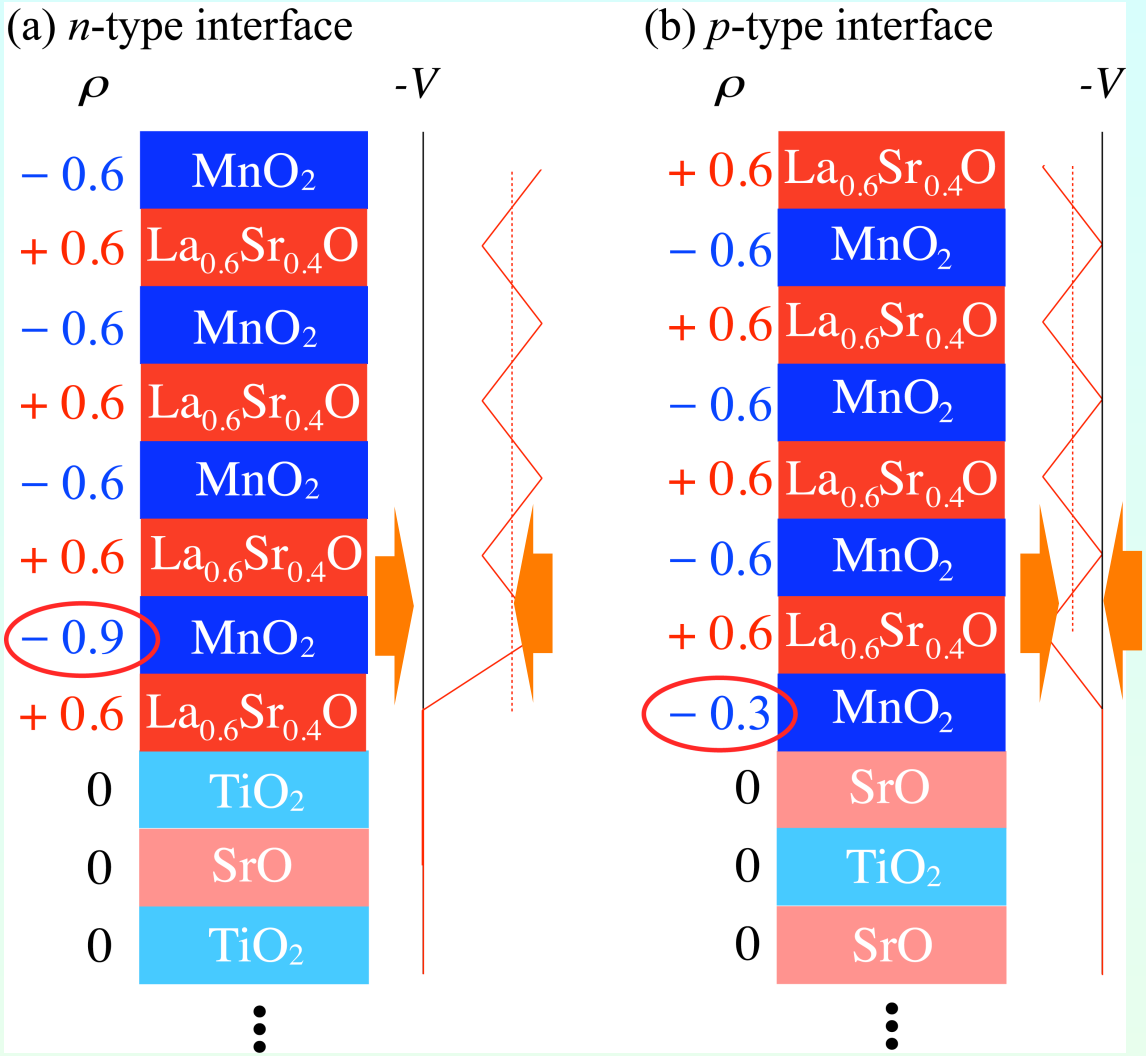
M. Minohara, H.K. *et al.*, *Phys. Rev. B* **81**, 235322 (2010).





H. Kumigashira *et al.*, Appl. Phys. Lett. **88**, 192504 (2006).

Origin of the interface dipole



$$V = \frac{Qd}{\varepsilon_r \varepsilon_0 S}$$

$\varepsilon_r = 30$ [Ref.1]

[1] R. D. Shannon, J. Appl. Phys. 73, 348 (1993).

V = +0.5 eV (*n*-type)
V = -0.2 eV (*p*-type)

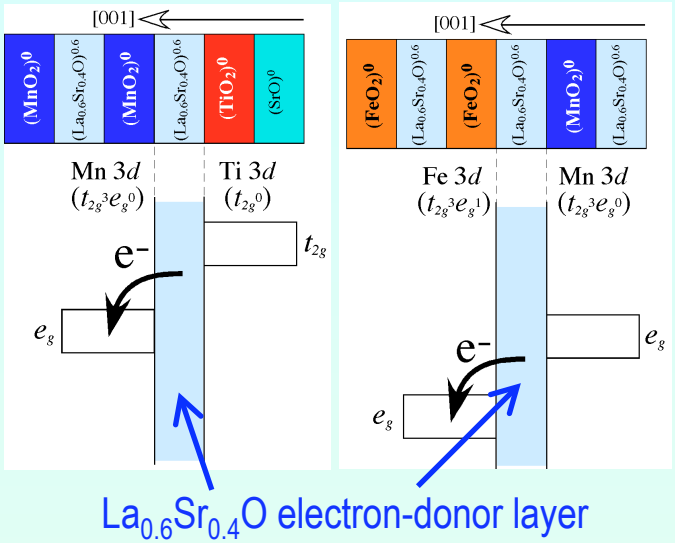
<u>Experimental results</u>

V = +0.5 eV (*n*-type)
V = -0.4 eV (*p*-type)

M. Minohara, H.K. *et al.*, *Phys. Rev. B* **81**, 235322 (2010).

まとめ

ABO_3/LSMO接合界面の電子状態について調べるために放射光電子分光を行った。



ヘテロ界面の化学状態

LSFO/LSMO: FeとMnイオン間の電荷移動

STO/LSMO: TiとMnイオン間の電荷移動は起こらず、Ti^{4+}状態を維持（Mn側が収納）

➡ 遷移金属イオンの3*d*準位の相対位置、およびFillingが重要。

ヘテロ界面のバンドダイアグラム

LSMO/TiO_2-STO: 0.5 eVの界面ダイポール形成
LSMO/SrO-STO: -0.4 eVの界面ダイポール形成（界面ダイポールの反転）

➡ 静電ポテンシャルの発散を抑制するために、Mn側の価数が変調

遷移金属イオンの3*d*準位位置、AO層の電荷量、終端面を考慮した界面設計