

- 放射光を用いたキャラクターゼーション -

# マンガン系人工超格子における 電荷・磁気状態の研究

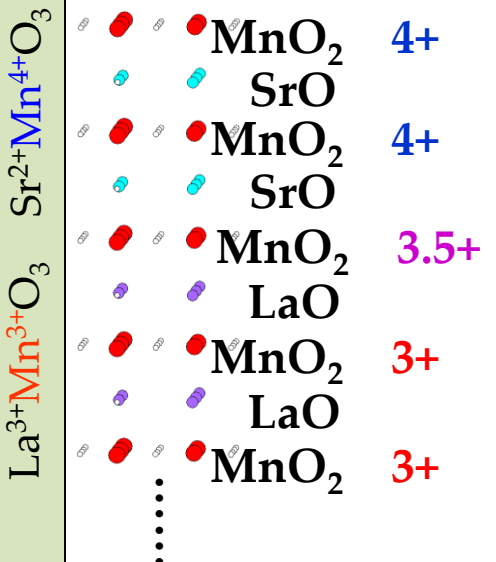
Condensed matter research center (CMRC)  
Photon Factory, IMSS, KEK

中尾裕則

## Outline

- ・研究背景
- ・放射光を用いた薄膜、人工格子の研究について
- ・中性子磁気散乱による磁気構造の研究の現状

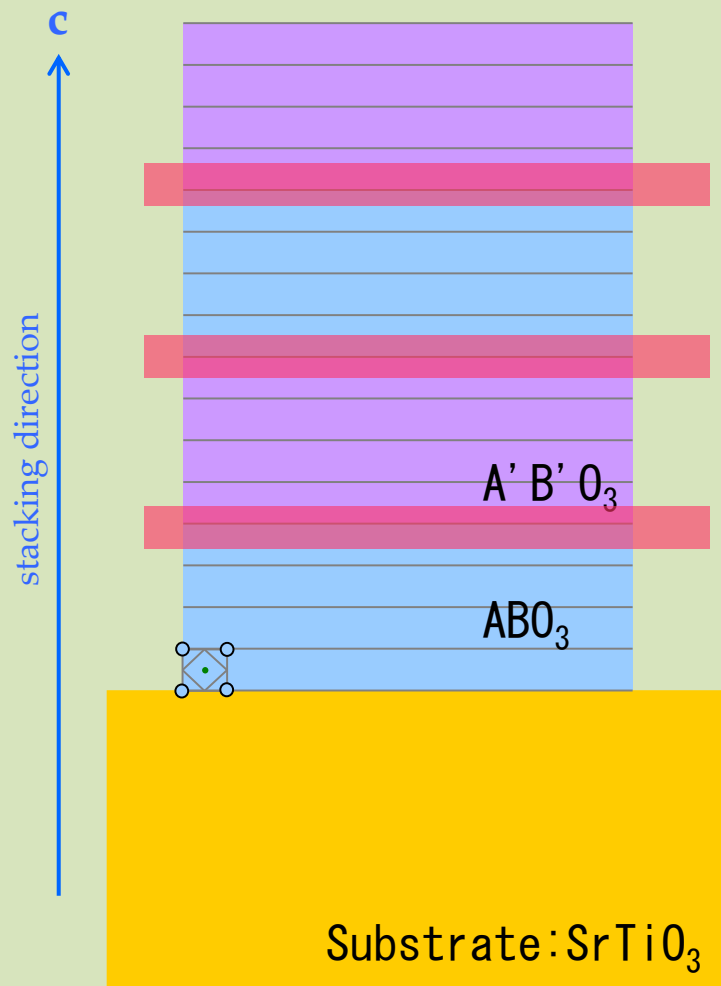
$(\text{LaMnO}_3)_2(\text{SrMnO}_3)_2$  の巨大磁気抵抗効果



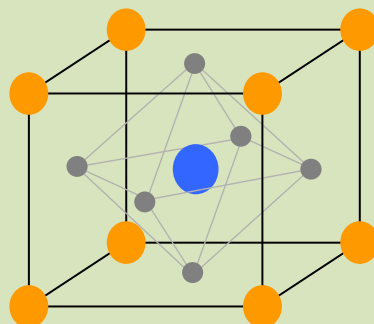
Substrate



# 人工超格子：自由な積層構造の設計による物性制御



結晶 ( $\text{ABO}_3$ )



ペロブスカイト構造

多様な物性

誘電体  
強磁性体  
反強磁性体  
超伝導体

巨大磁気抵抗効果  
電気磁気効果

任意の積層構造の作製

電荷・スピン・軌道・格子の制御

新奇物性の探索や、  
自由なデバイス設計の可能性

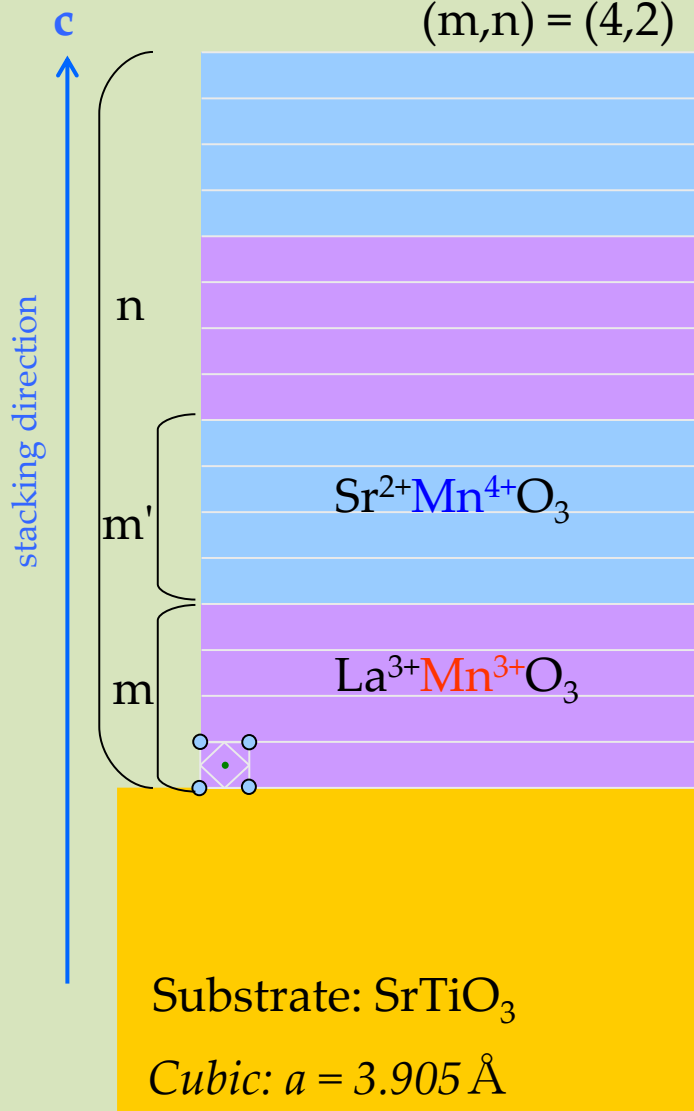
ヘテロ界面で作り出される新たな電子状態



# 人工超格子: $[(\text{LaMnO}_3)_m(\text{SrMnO}_3)_{m'}]_n$

Perovskite structure

$(m,n) = (4,2)$



Substrate:  $\text{SrTiO}_3$

Cubic:  $a = 3.905 \text{ \AA}$

$\text{Sr}^{2+}\text{Mn}^{4+}\text{O}_3$  G-type AF

$e_g$

$t_{2g}$

$\text{Mn}^{4+}$

Band insulator

$\text{La}^{3+}\text{Mn}^{3+}\text{O}_3$  A-type AF

$e_g$

$t_{2g}$

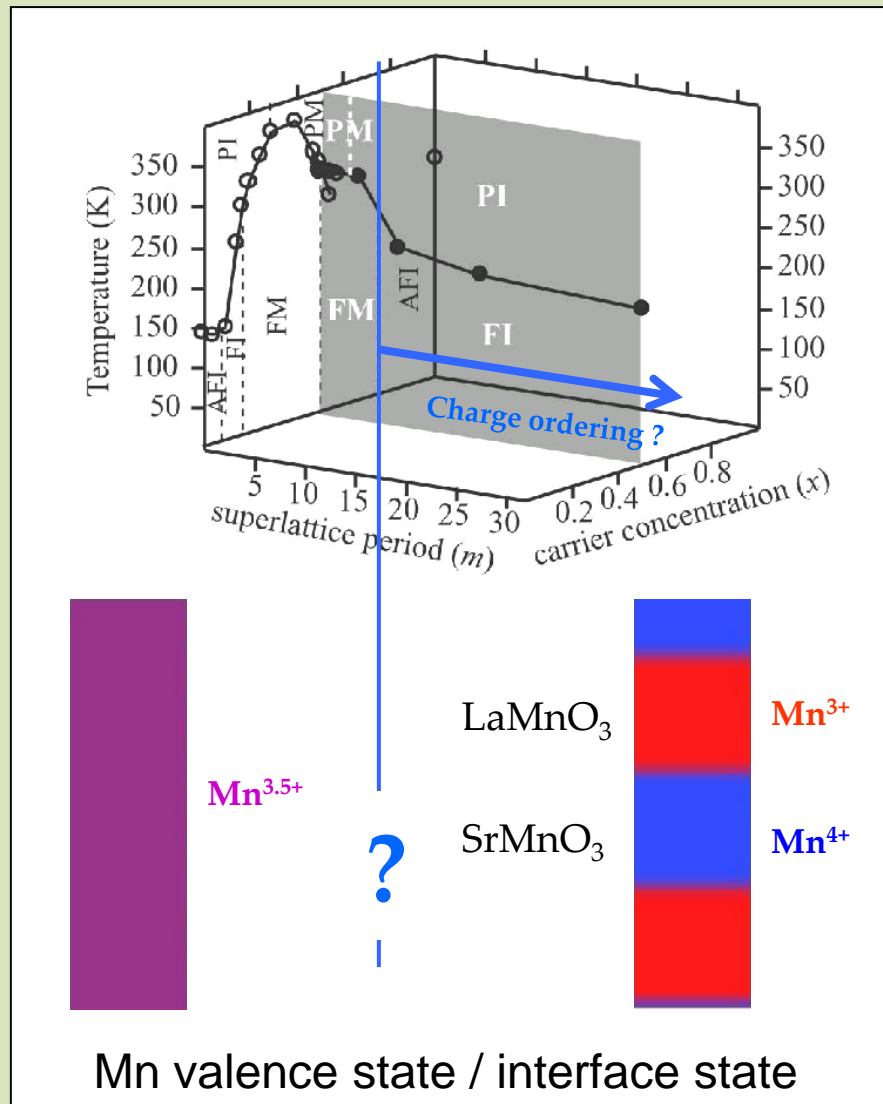
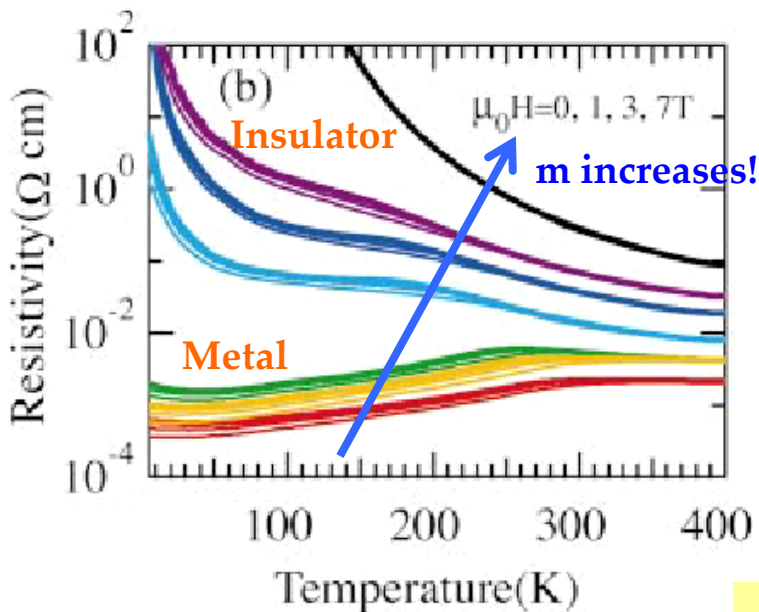
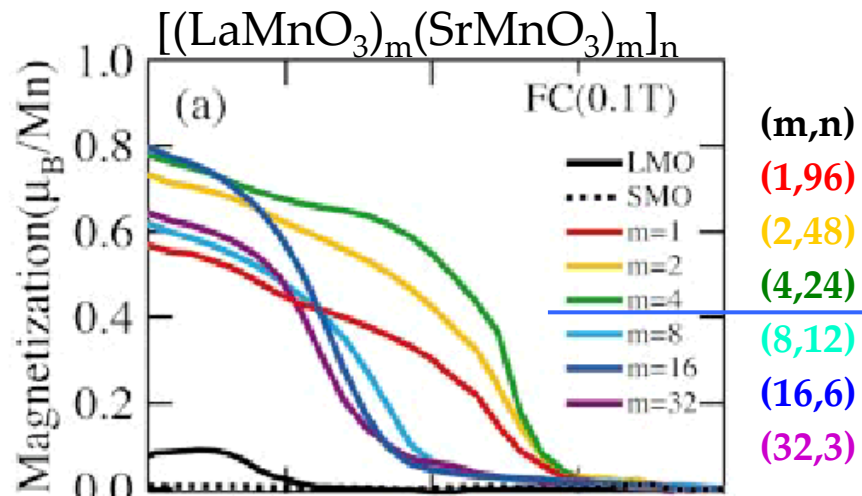
$\text{Mn}^{3+}$

Mott insulator

電荷変調構造の制御 → 新奇物性?



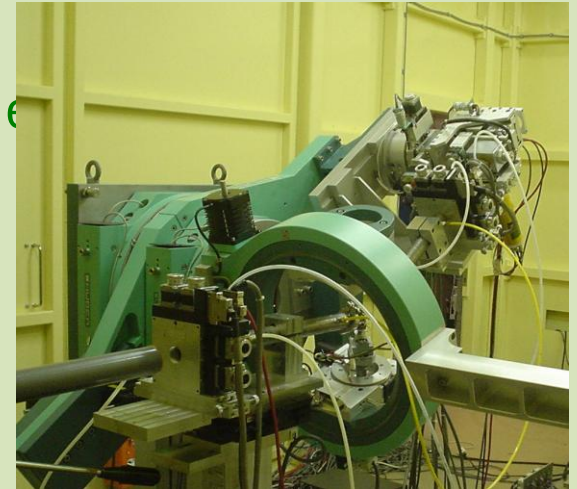
# 人工超格子: $[(\text{LaMnO}_3)_m(\text{SrMnO}_3)_m]_n$



Specific character in superlattice



- Resonant x-ray scattering study of the superlattices to clarify the charge distribution of the Mn ion
- Evaluation of the superlattice structure



## Experiments

- Synchrotron x-ray scattering
- Diffraction pattern along the stacking direction at 6.52 keV  
→ Superlattice structure (atomic coordinations)
- Energy dependence of the scattering intensity near Mn K-edge (~ 6.55 keV)

→ Charge distribution of Mn ion



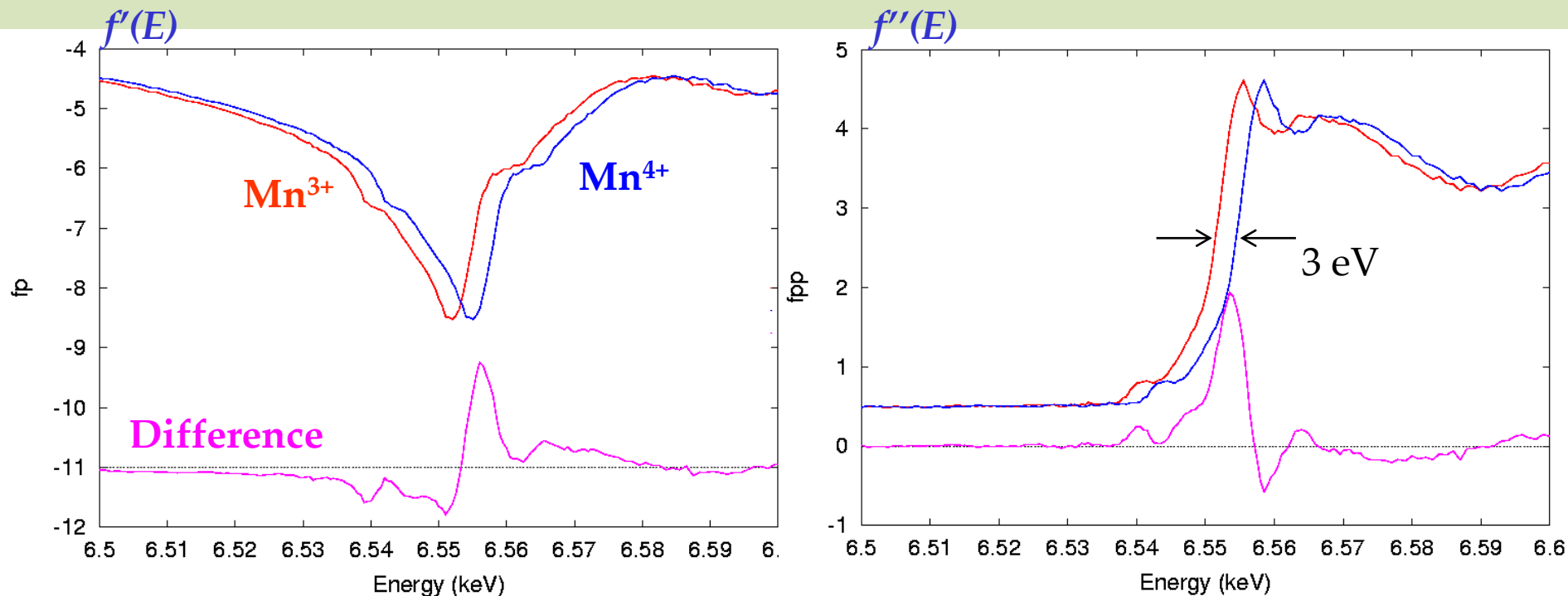
$$I(E, hkl) = |F(E, hkl)|^2$$
$$F(E, hkl) = \sum_i f_i(E) \exp[-2\pi i(hx_i + ky_i + lz_i)]$$

$$f(E) = f_0 + f'(E) + i f''(E)$$

X-ray anomalous scattering factor



# X-ray anomalous scattering factor of Mn ion



- Energy dependence of the scattering intensity near Mn K-edge ( $\sim 6.55$  keV)

→ Charge distribution of Mn ion



$$F(E, hkl) = \sum_i f_i(E) \exp[-2\pi i(hx_i + ky_i + lz_i)]$$

$$f(E) = f_0 + f'(E) + if''(E)$$

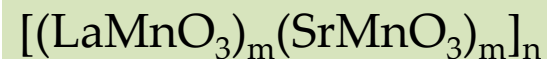
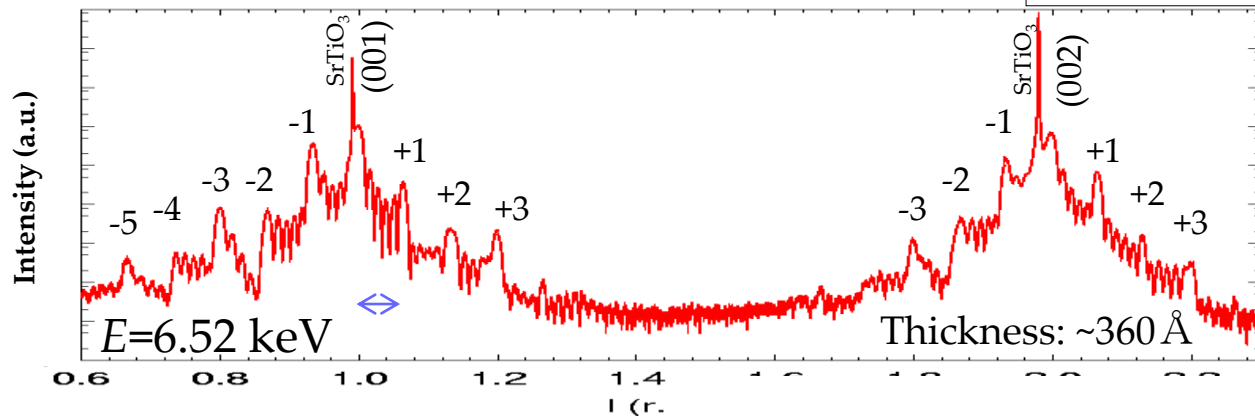
X-ray anomalous scattering



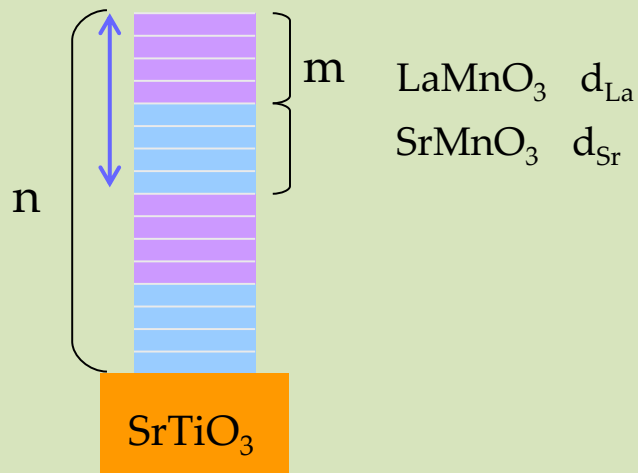
# Crystal structure of superlattice

$$I(E, hkl) = |F(E, hkl)|^2$$

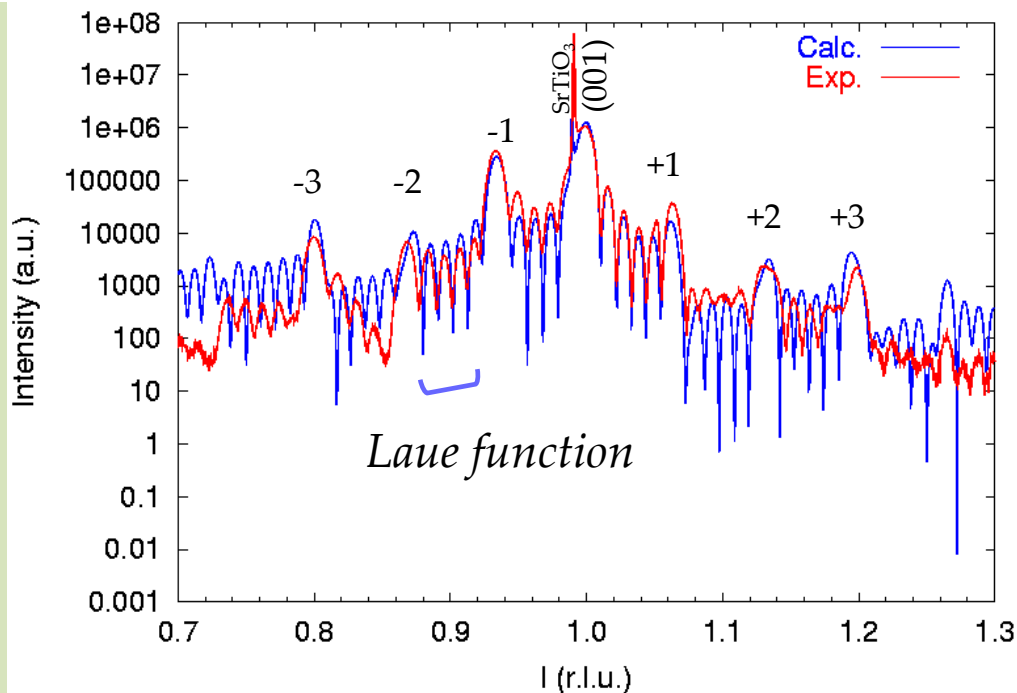
$$F(E, hkl) = \sum_i f_i(E) \exp[-2\pi i(hx_i + ky_i + lz_i)]$$



$$(m, n) = (8, 6)$$

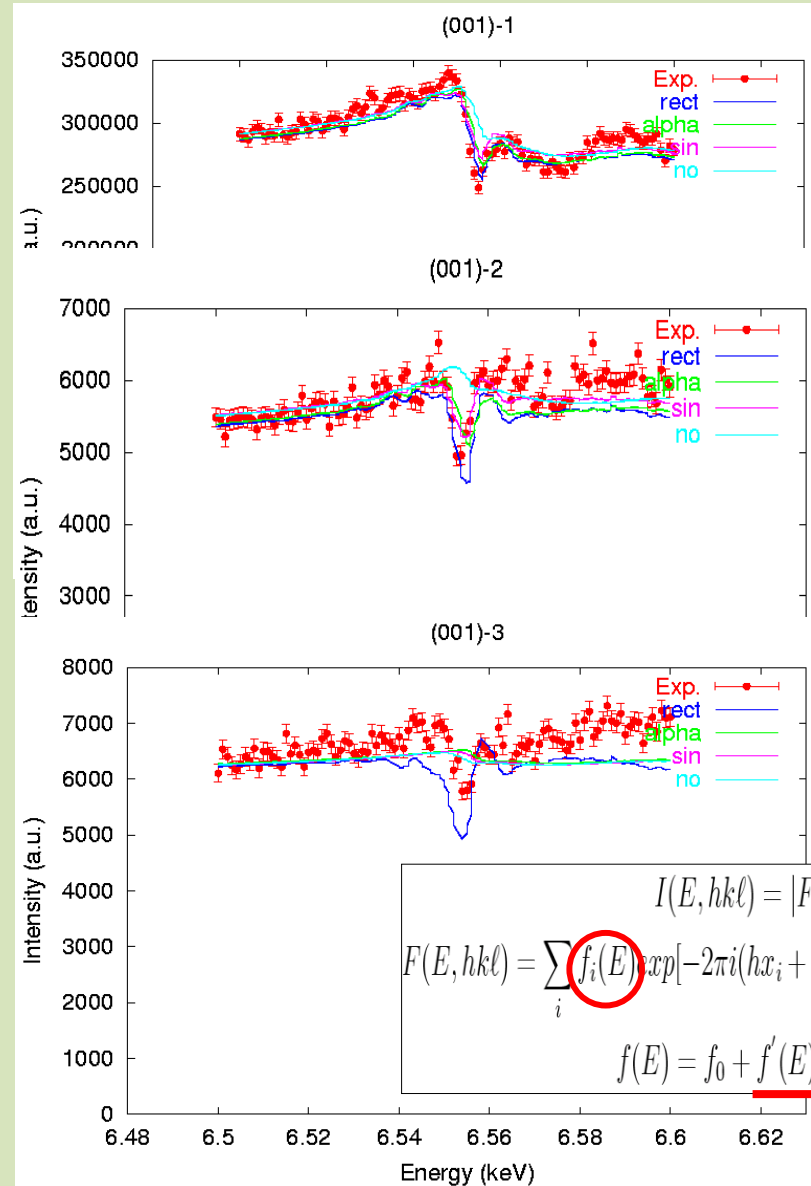
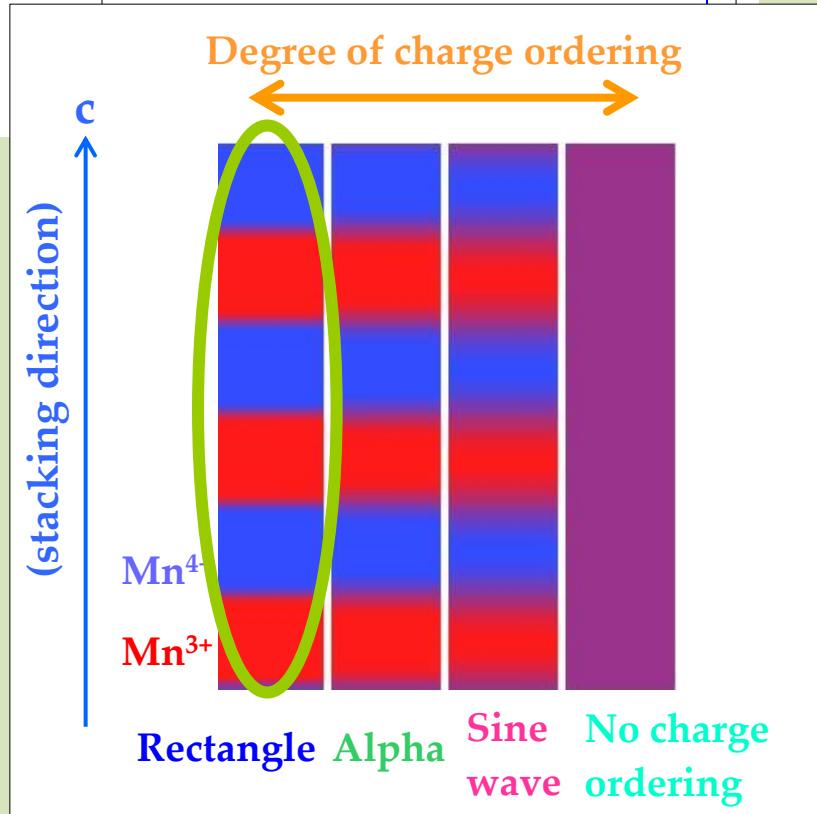
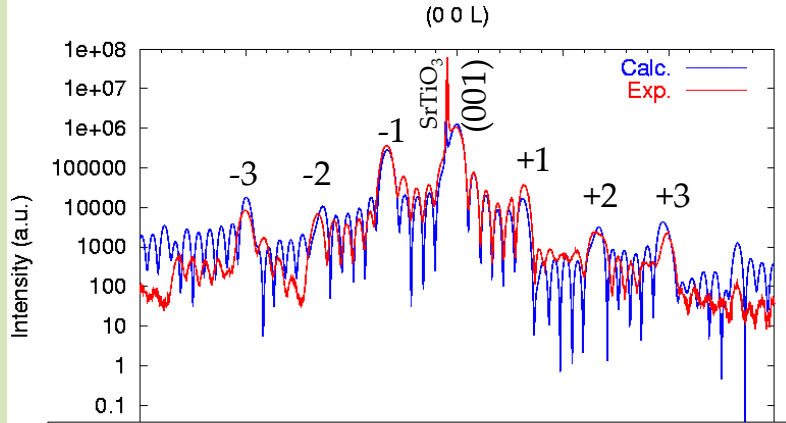


Structural deviation near interface  
Substrate scattering





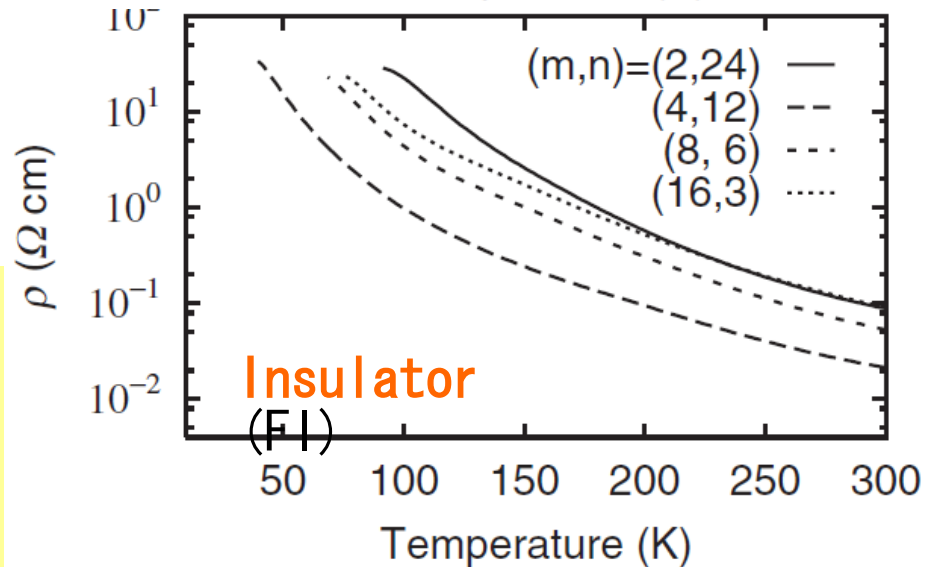
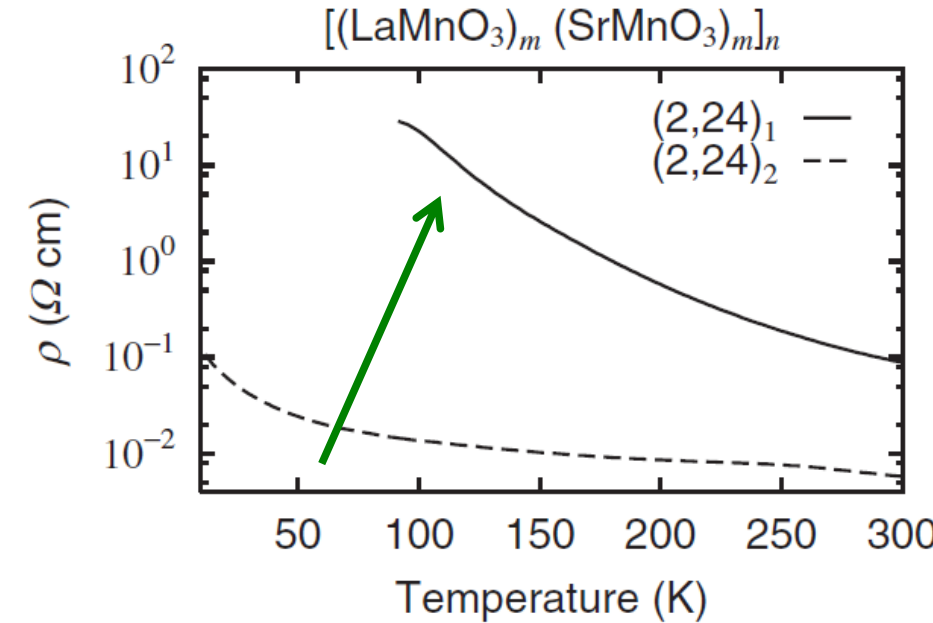
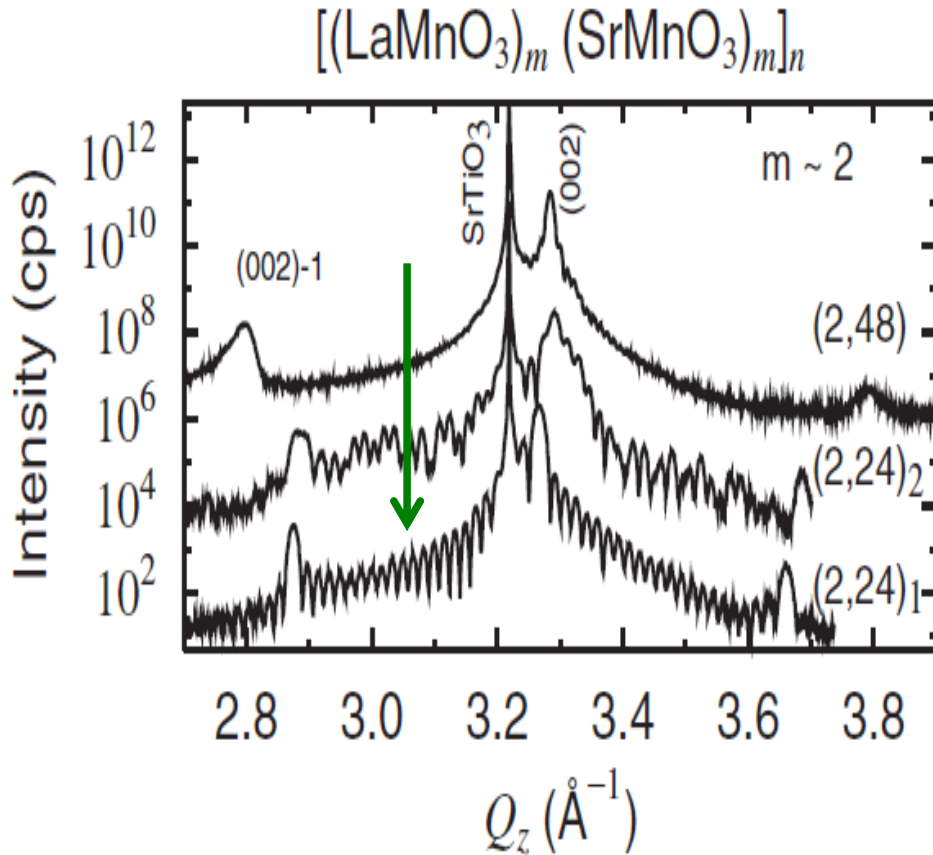
# $[(\text{LaMnO}_3)_m(\text{SrMnO}_3)_m]_n$ (m,n) = (8,6)








# Insulator vs. Metal

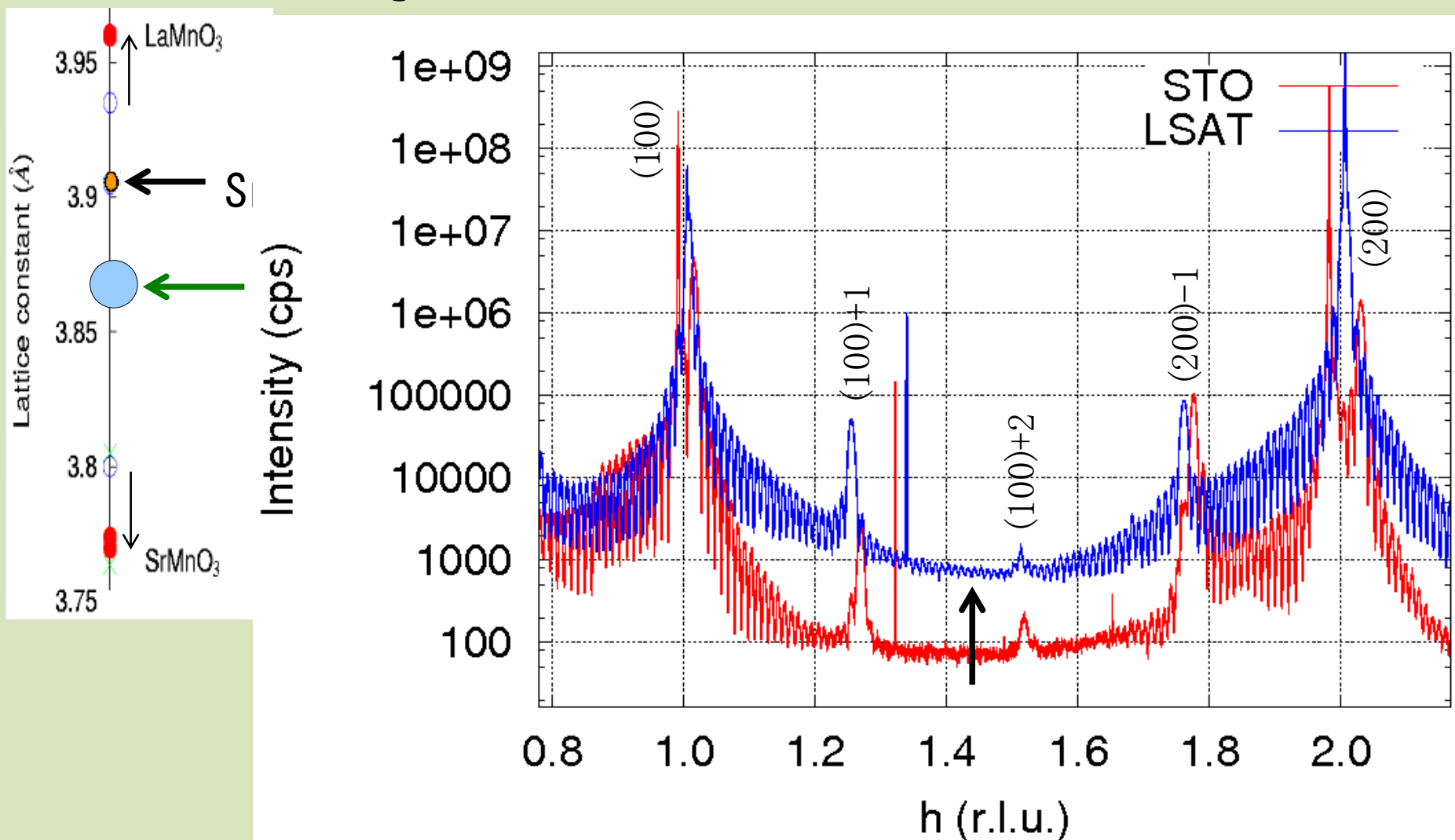


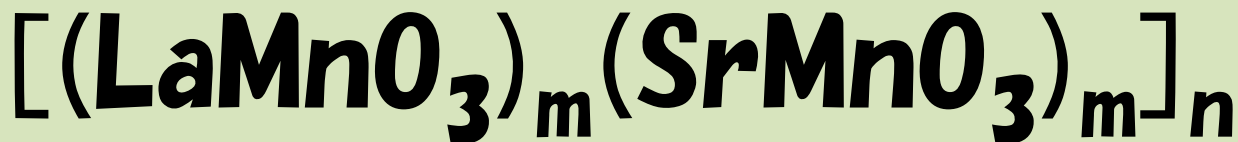
Quality of stacking structure  
  
 Conductivity



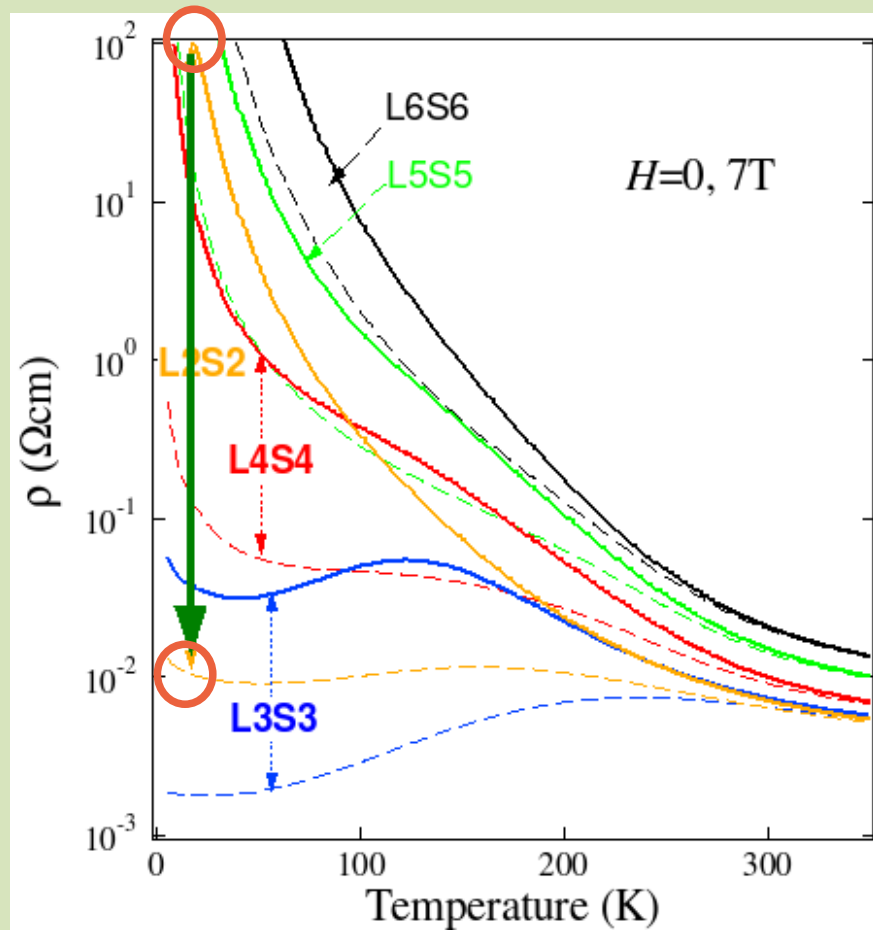
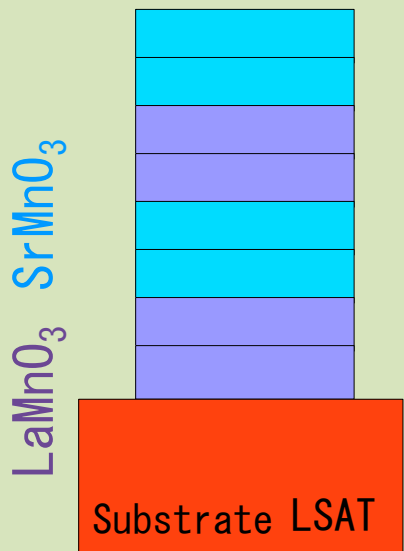
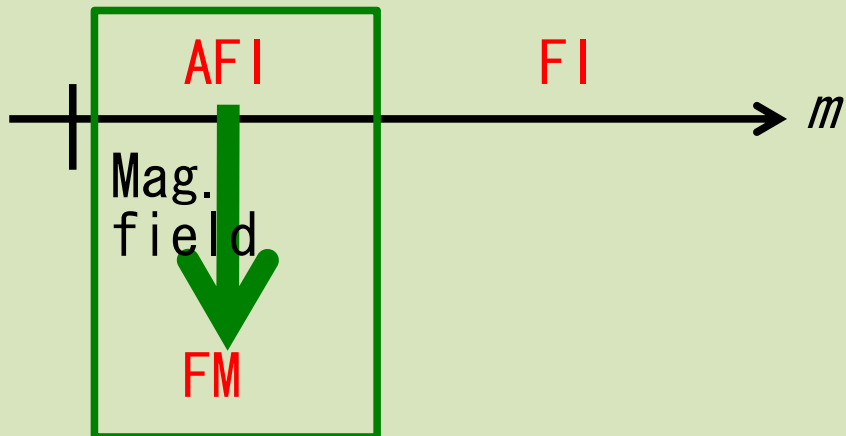
# Sample quality: $\text{SrTiO}_3$ vs. LSAT

Lattice matching between film and substrate





$[(\text{LaMnO}_3)_m(\text{SrMnO}_3)_m]_n$  LSAT



$m \leq 4$ :

Large negative magneto-resistance

This is new phenomenon in film system.

not reported in the bulk



# Study project in CMRC

- Synchrotron radiation

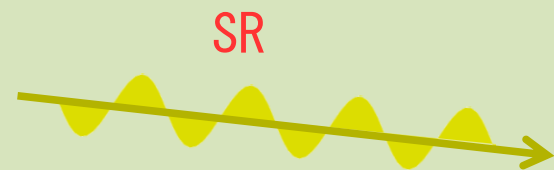
Resonant and non-resonant x-ray scattering

- Mn valence distribution
- Stacking structure

Resonant soft x-ray scattering

- Resonant magnetic scattering
- Mn3d - O2p orbital hybridization
- Modulation of electronic structure

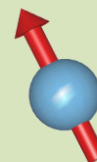
Depth resolved XAS, XMCD (雨宮)...



- Neutron

- Magnetic scattering (東北大 岩佐)
- Reflectivity (JAEA 武田)

→ J-PARC



Neutron

- Muon

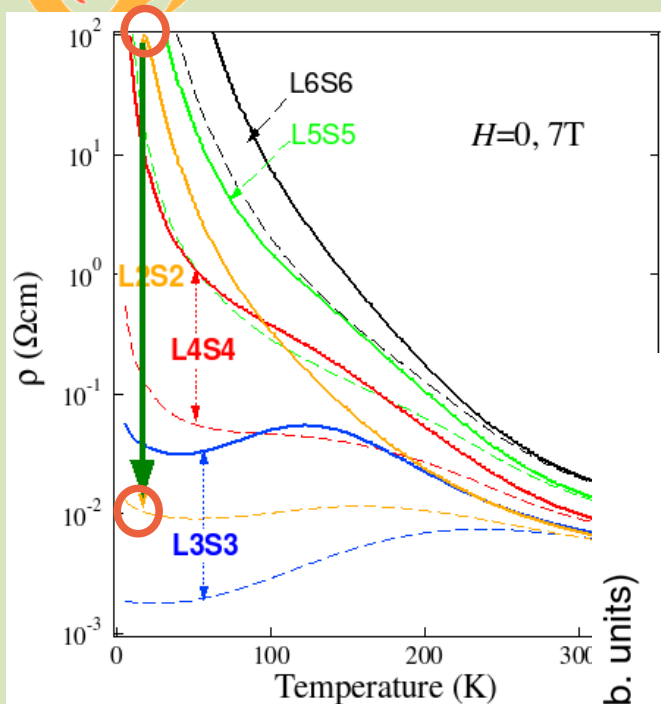
Ultra-slow muon



Muon



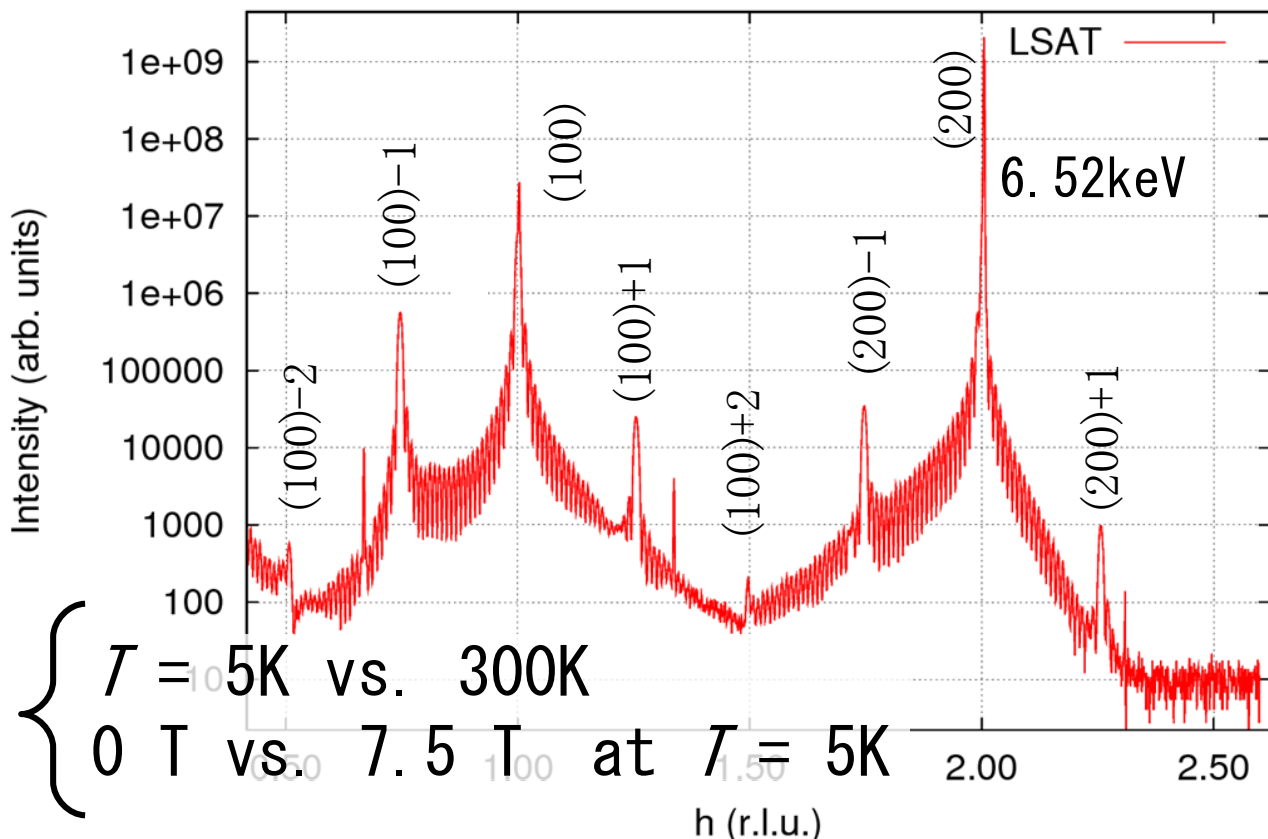
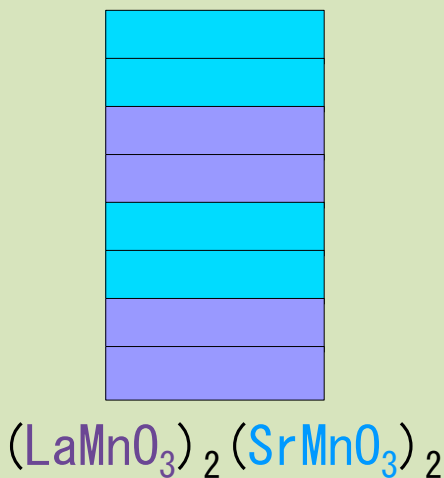
# Crystal structure in superlattice



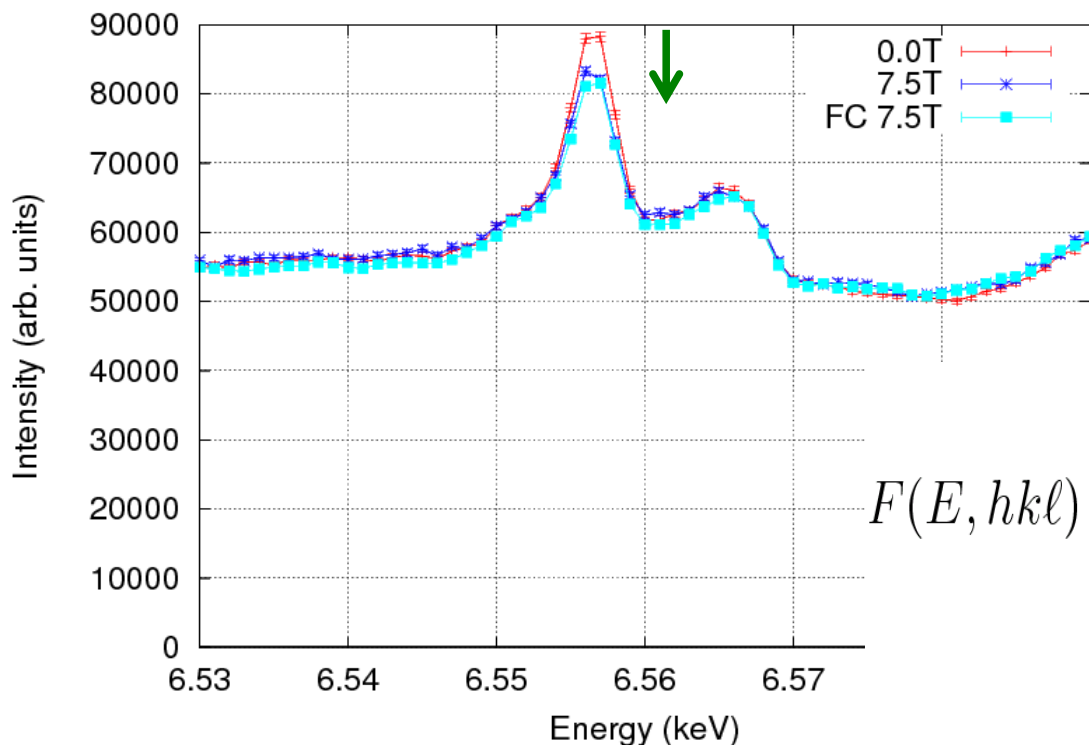
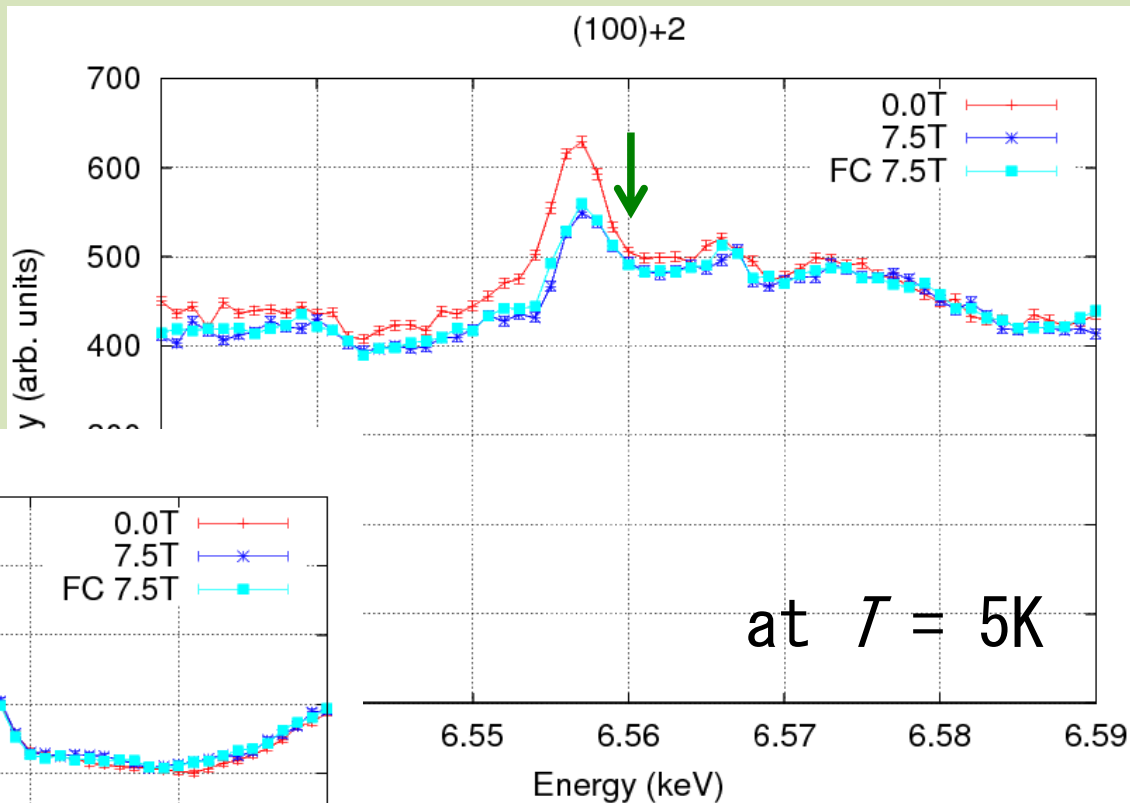
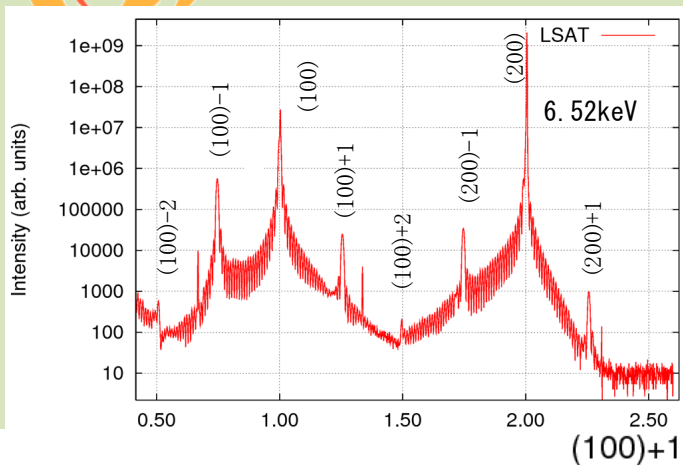
$$I(E, hkl) = |F(E, hkl)|^2$$

$$F(E, hkl) = \sum_i f_i(E) \exp[-2\pi i(hx_i + ky_i + lz_i)]$$

$$f(E) = f_0 + f'(E) + if''(E)$$



# Magnetic effect for Mn valence state studied by resonant x-ray scattering



$$I(E, hkl) = |F(E, hkl)|^2$$

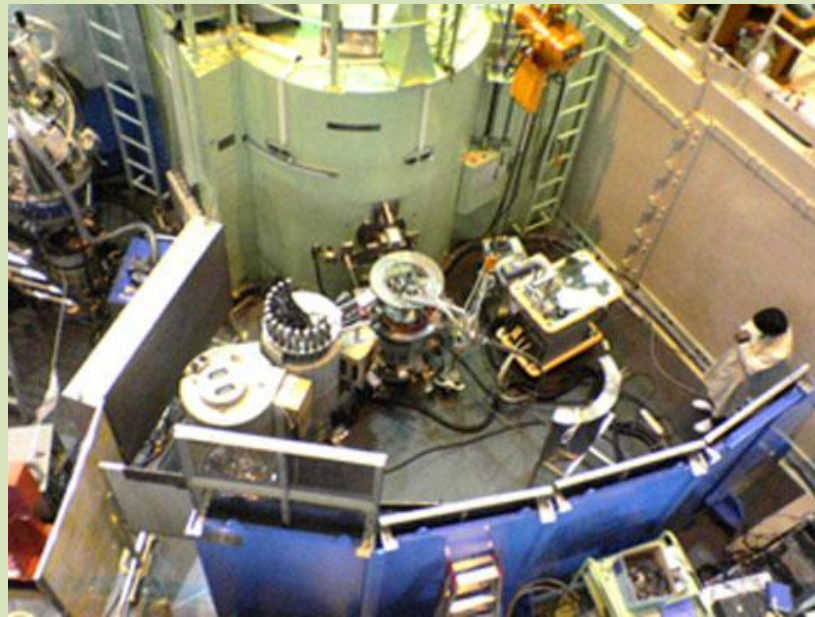
$$F(E, hkl) = \sum_i f_i(E) \exp[-2\pi i(hx_i + ky_i + lz_i)]$$

$$\underline{f(E) = f_0 + f'(E) + if''(E)}$$





# 中性子磁気散乱による 人工格子の磁性研究の可能性

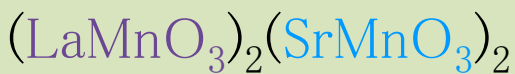


TOPAN (6G) at JRR-3 in Tokai

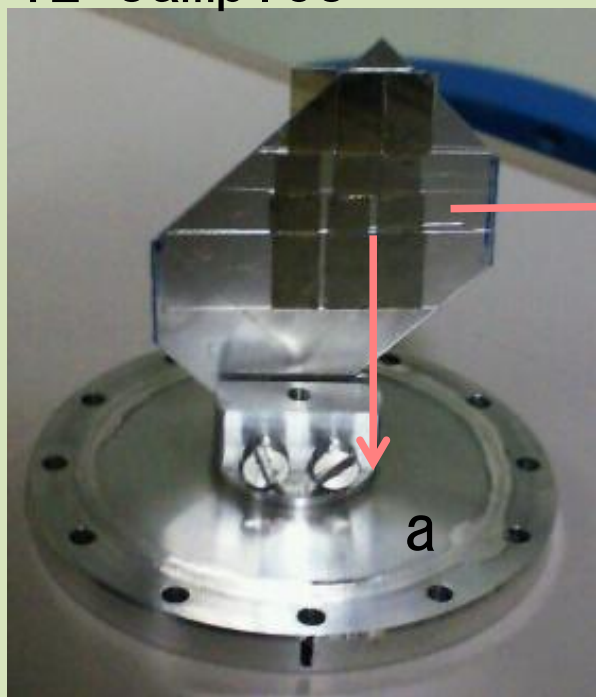
Tohoku Univ. : K.  
Iwasa



# L2S2 / LSAT

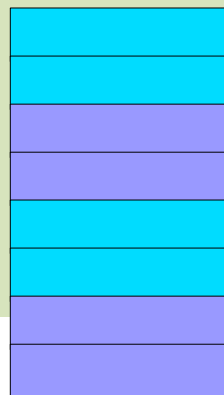


12 samples



b

a

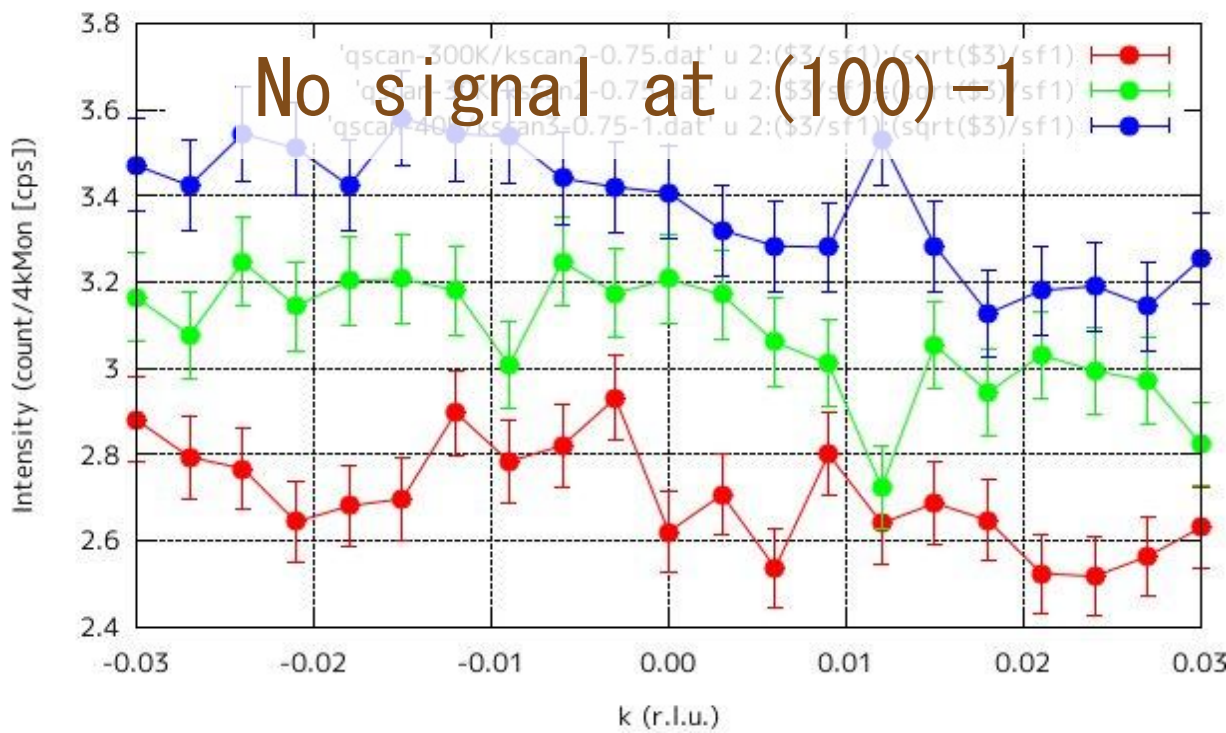


Magnetic scattering at  $(100) \pm 1$

B-PG-Sap-100-Sample-PG-100-B  $E_i=14.7\text{meV}$

$3\text{cm} \times 1.5\text{cm} \times 400 \text{ \AA}$

$< 0.3 \times 0.3 \times 0.3 \text{ mm}^3$

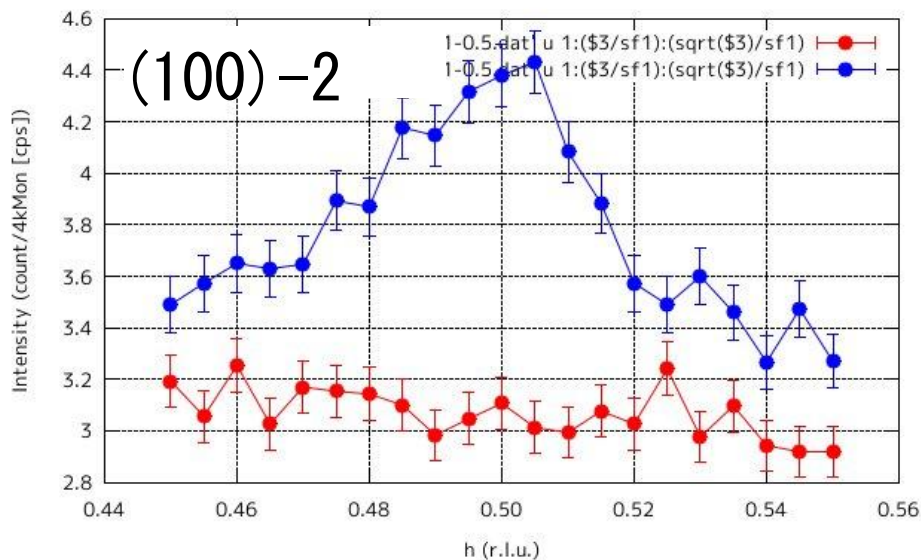




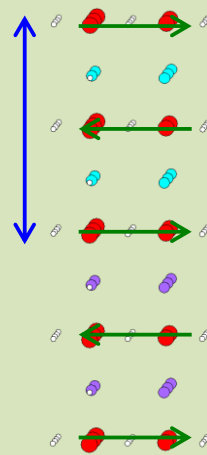
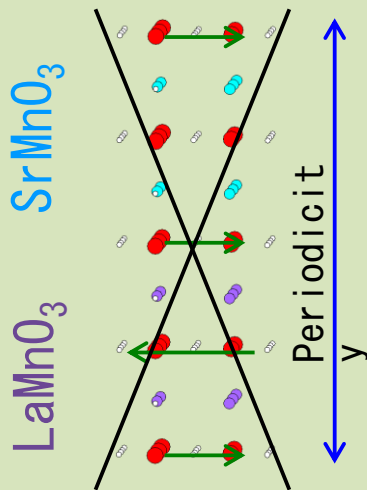
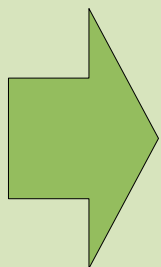
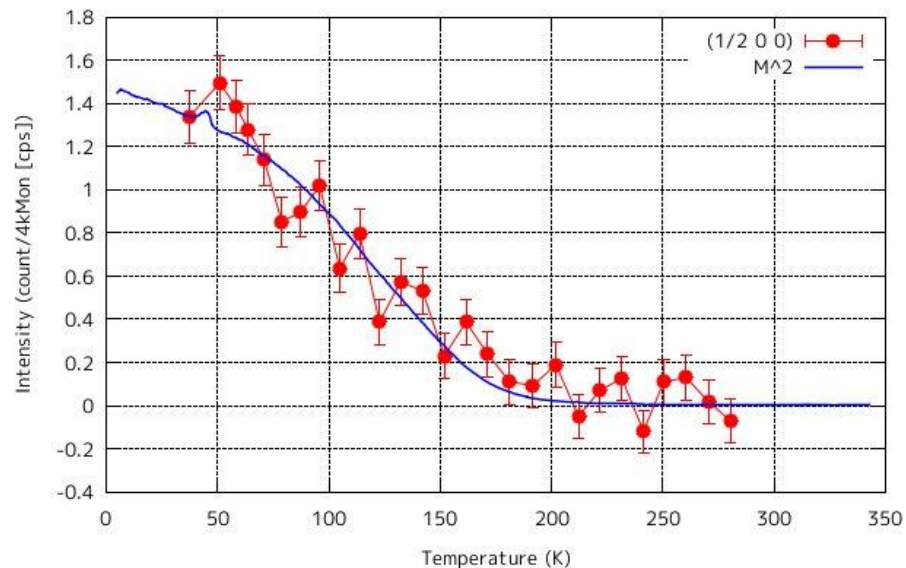


# Magnetic scattering: L2S2/LSAT

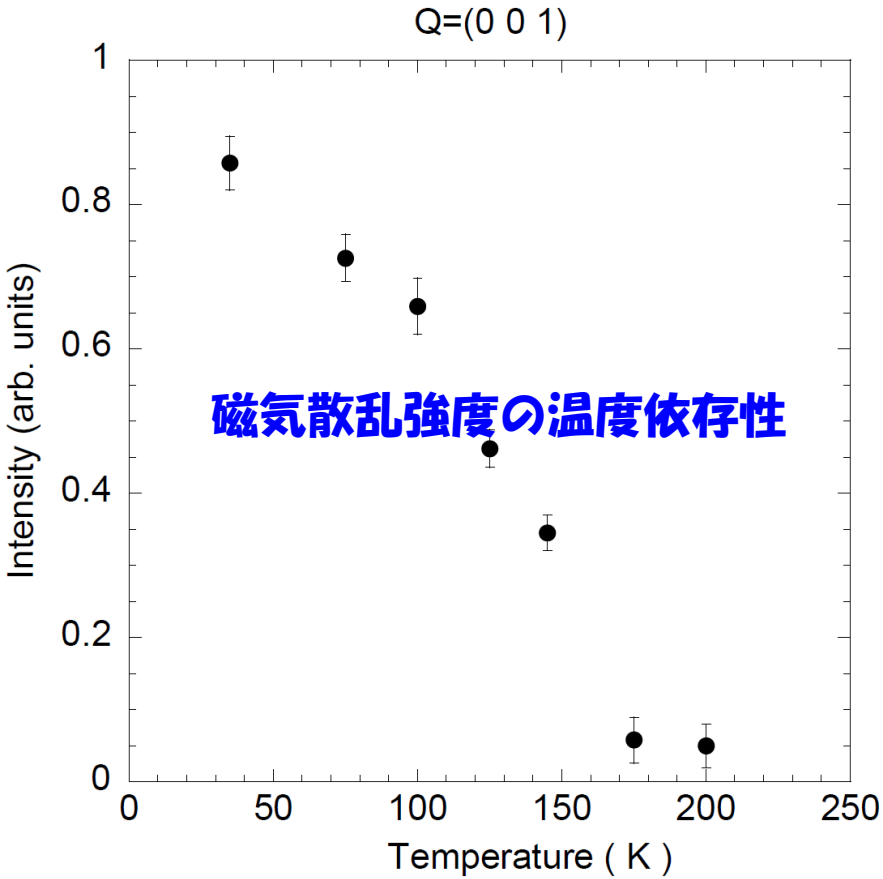
B-PG-Sap-100-Sample-PG-100-B  $E_i=14.7\text{meV}$



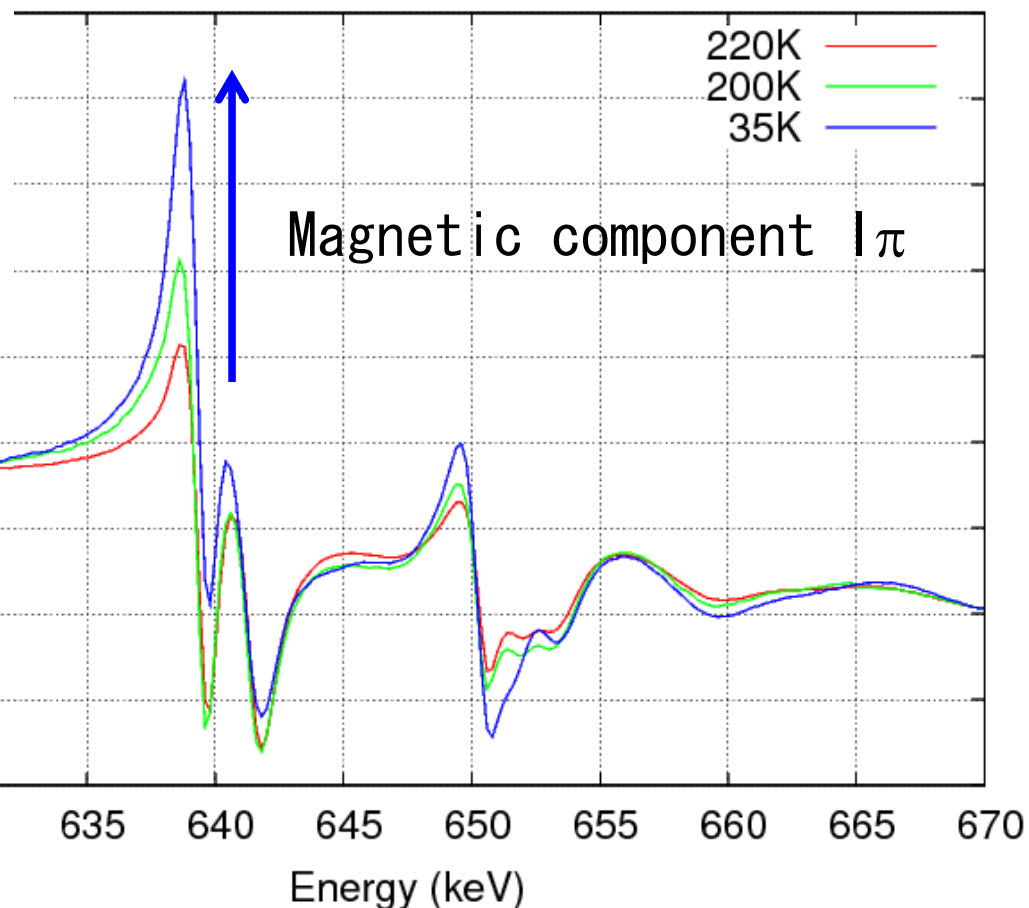
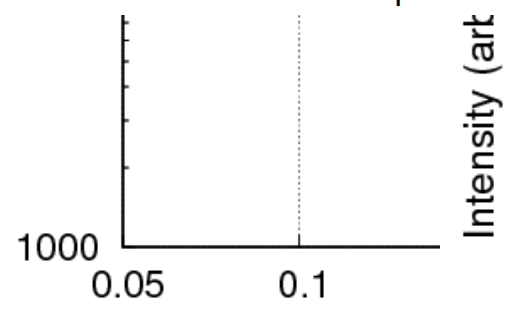
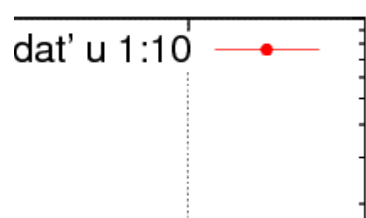
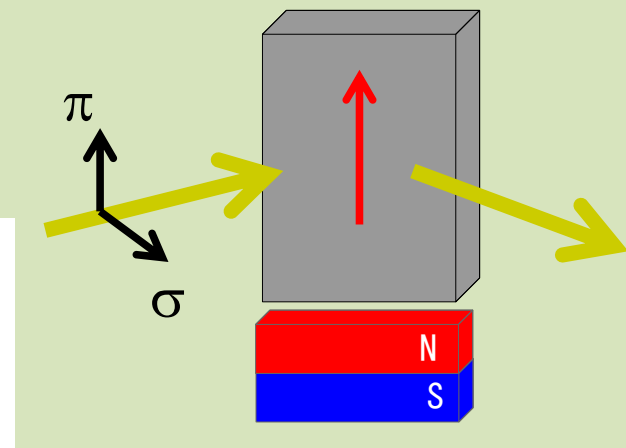
B-PG-Sap-100-Sample-PG-100-B  $E_i=14.7\text{meV}$



Simple AFM



dge: L5S5



久保田  
(JAEA)



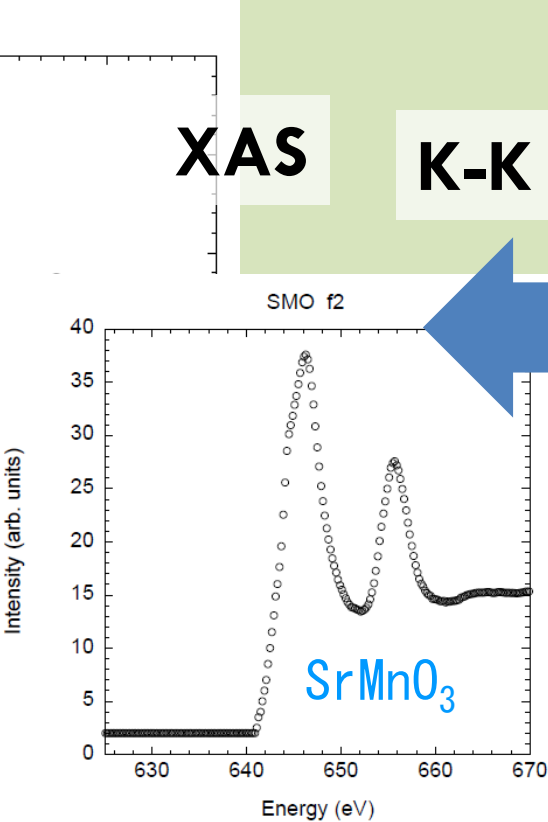
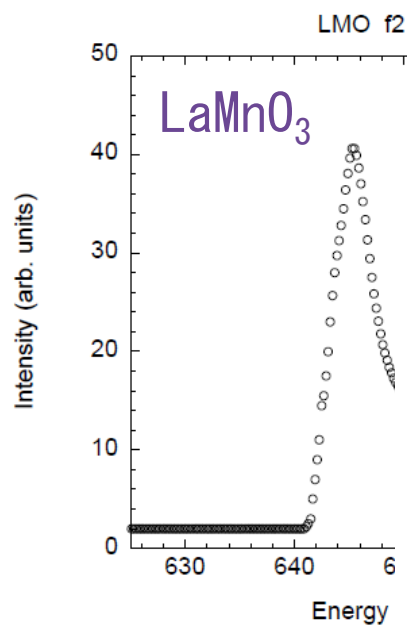
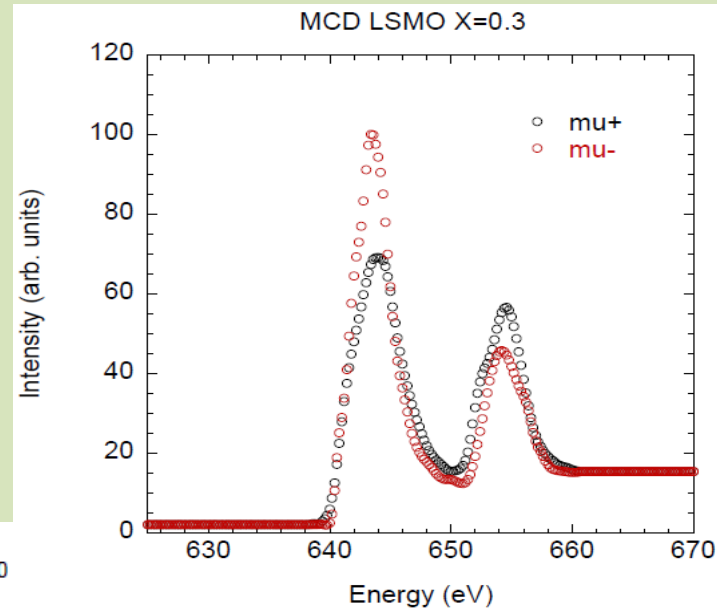
# Form factor

$$I(E, hkl) = |F(E, hkl)|^2$$

$$F(E, hkl) = \sum_i f_i(E) \exp[-2\pi i(hx_i + ky_i + lz_i)]$$

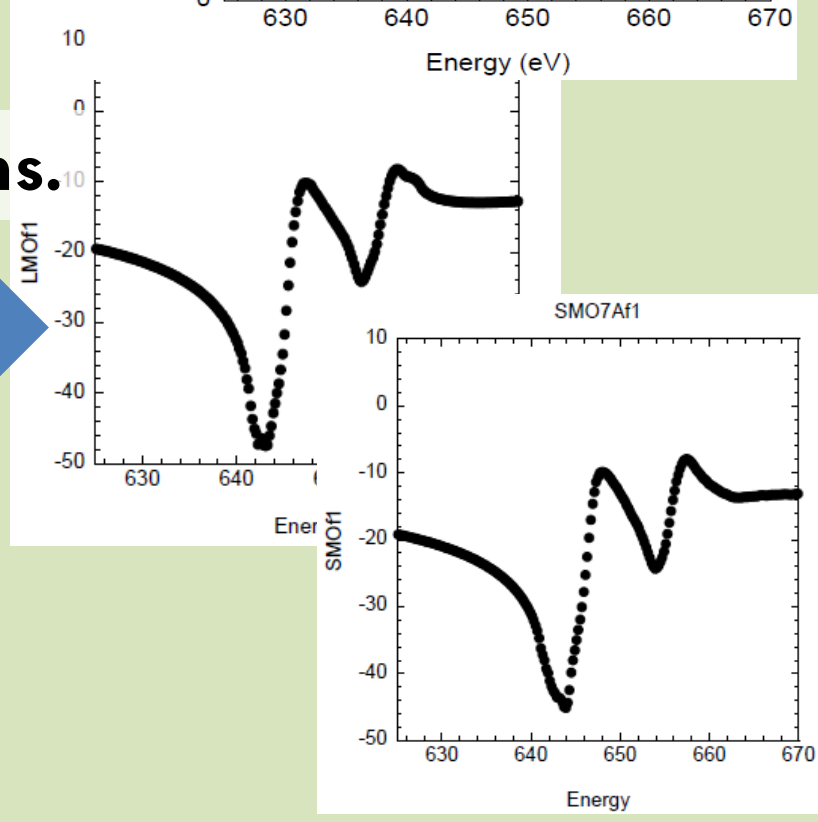
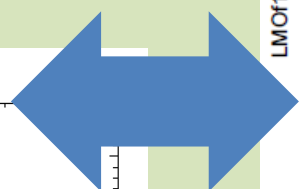
$$f(E) = f_0 + \underline{f'(E)} + i f''(E)$$

X-ray anomalous scattering factor



XAS

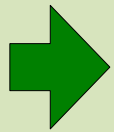
K-K trans.





# Study of artificial superlattice

- { Mn valence distribution  
Stacking structure (crystal structure)



Resonant and non-resonant x-ray scattering

- Magnetic structure



{ Neutron magnetic scattering / Reflectivity  
Resonant magnetic x-ray scattering: Mn L-edge



L2S2

L5S5

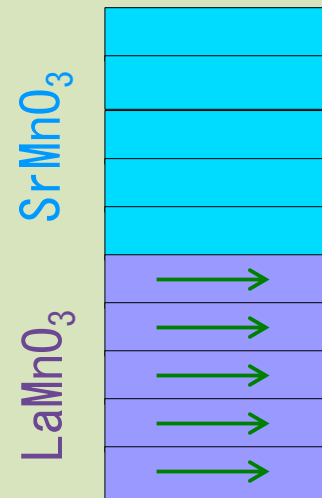
AFI

FI



Mag.  
field

FM





# Collaborator

**Y. Yamasaki, J. Okamoto, T. Sudayama, Y. Murakami**

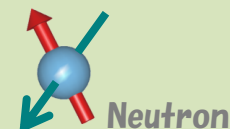
*Condensed Matter Research Center / Photon Factory, IMSS, KEK*



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**M. Kubota, Y. Takeda** *Japan Atomic Energy Agency*



**J. Nishimura, A. Ohtomo, T. Fukumura, and M. Kawasaki**

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**H. Yamada, A. Sawa**

*Nanoelectronics Research Institute, AIST*