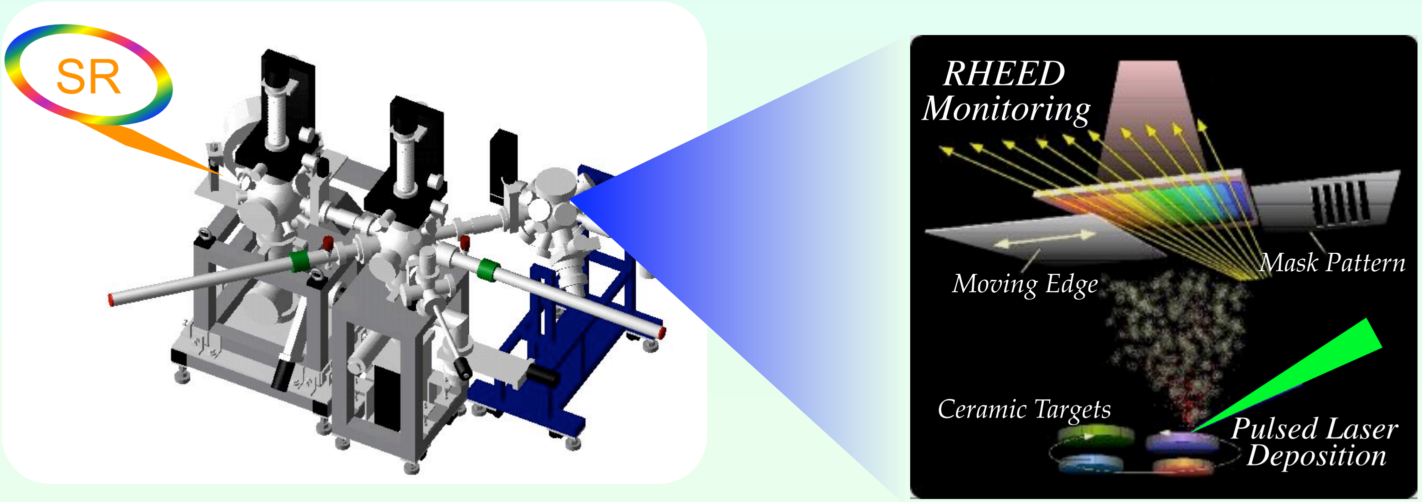
PF研究会「軟X線分光・散乱測定を用いた物性研究の現状と展望」
H23年９月13（13,14日）＠KEK研究本館小林ホール

次元性制御強相関酸化物薄膜の軟X線光電子分光

組頭 広志
KEK−PF（物構研＆構造物性研究センター）
JSTさきがけ



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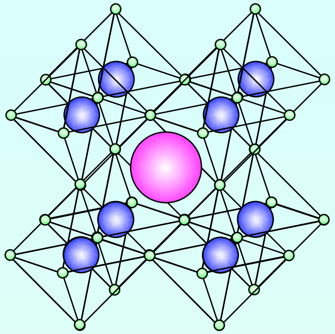
INTRODUCTION

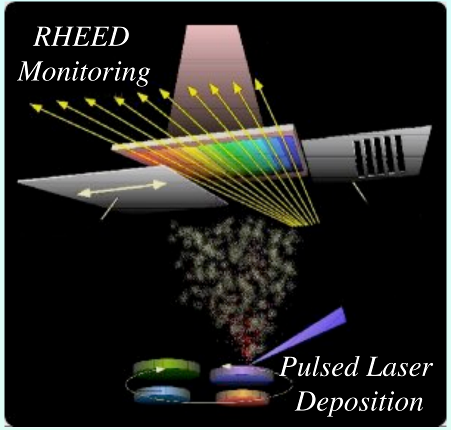
Transition Metal Oxides



High-Tc
CMR
M-I Transiton

Controlling unusual physical properties

Laser MBE



e^-

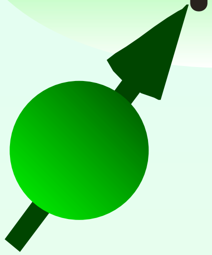
Charge

Charge Transfer

Heterostructure

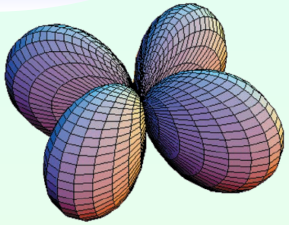
Spin

Spin Exchange



Orbital

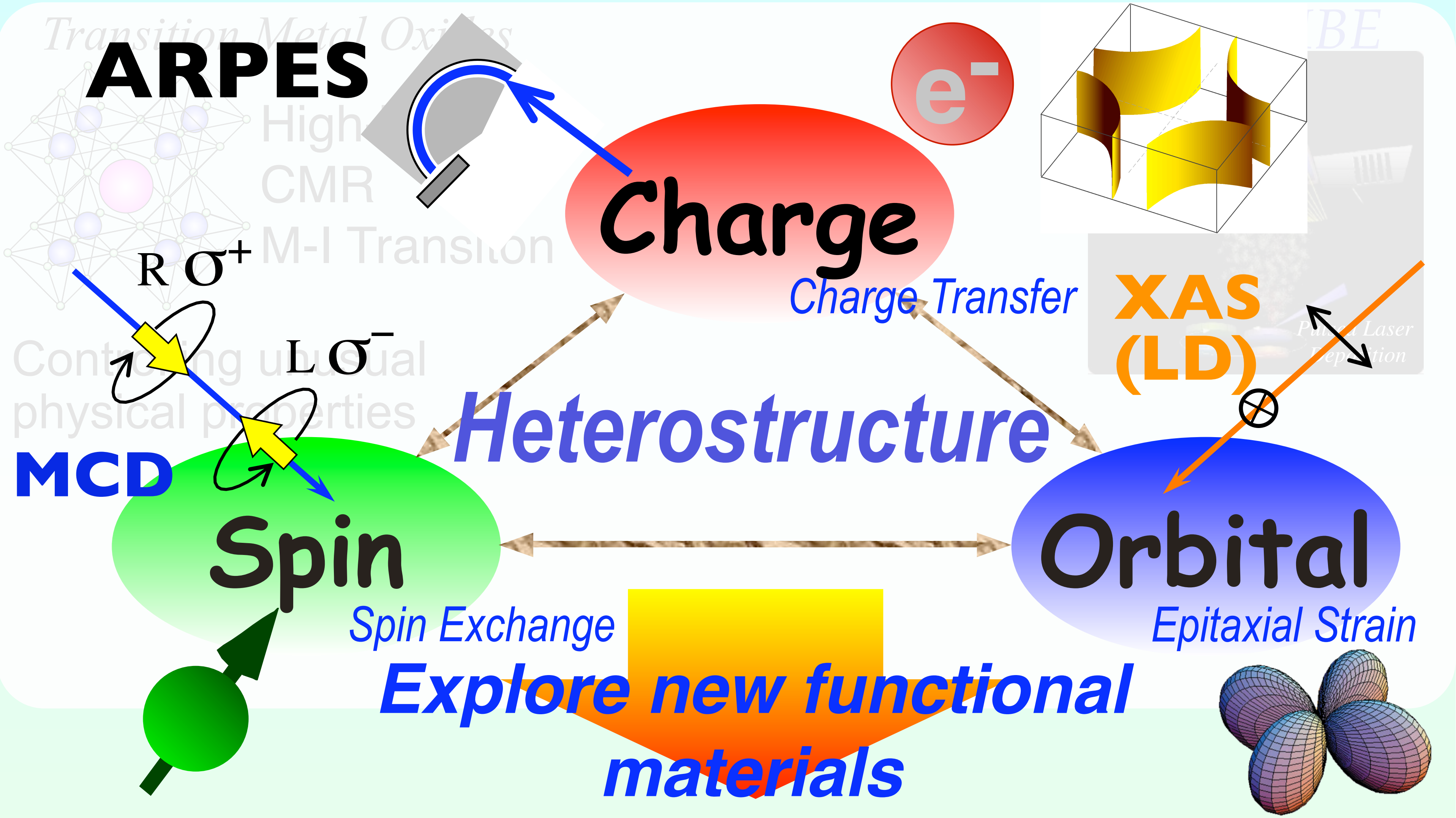
Epitaxial Strain



Explore new functional materials

"in-situ Photoemission"

Oxide Artificial Structures



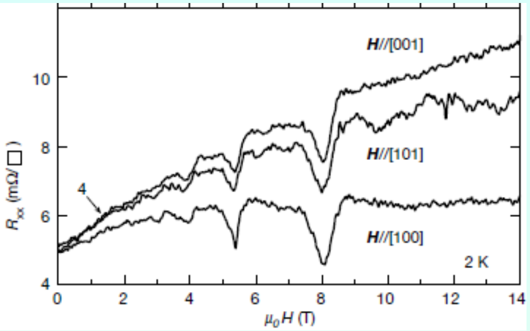
Explore new functional materials

"in-situ Photoemission"

Appearance of metallic conductivity at the interface between the band insulators $LaAlO_3$ and $SrTiO_3$

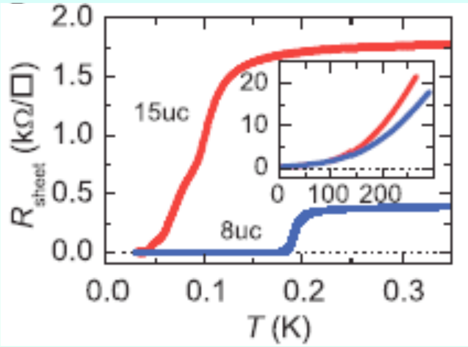
High-mobility electron gas

(A. Ohtomo *et al*, Nature 2004)



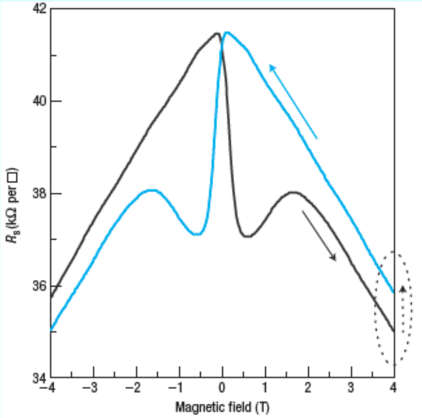
Superconductivity

(N. Reyren *et al*., Science 2007)



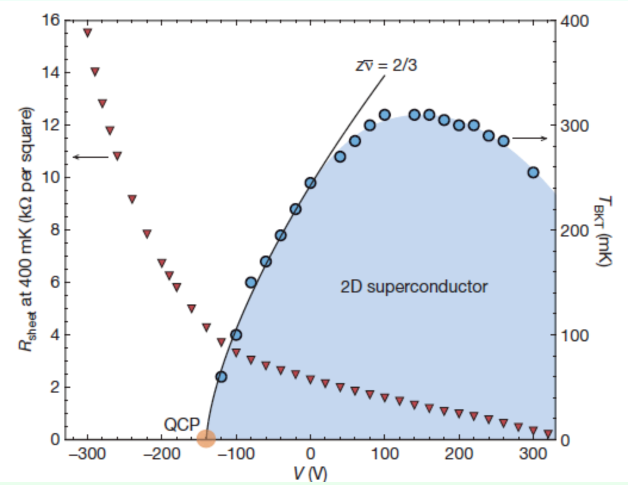
Ferromagnetism?

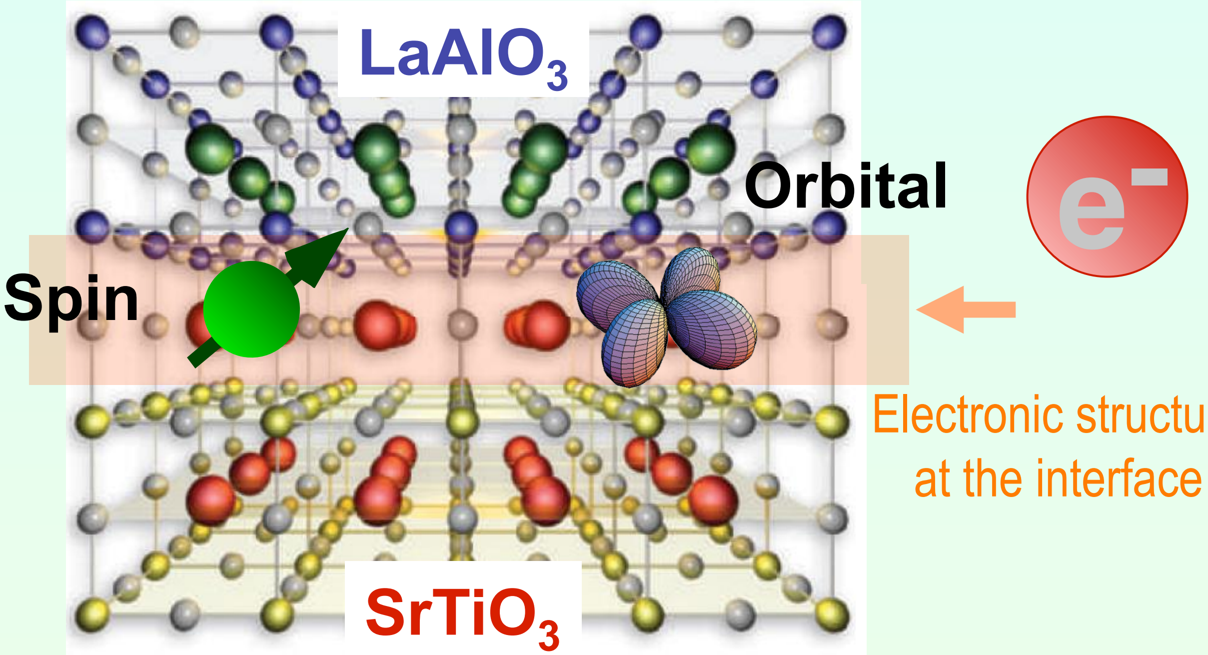
(A. Brinkman *et al*, Nat. Mater. 2007)



Electric field control of the superconductivity

(A.D. Caviglia *et al*, Nature 2008)

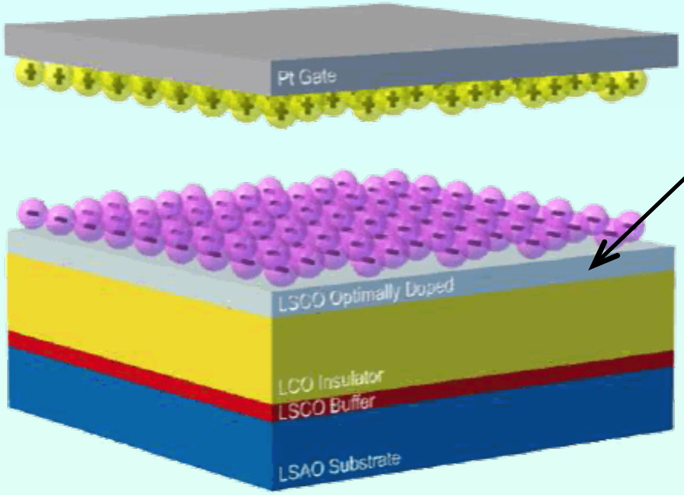


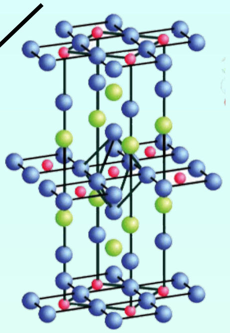


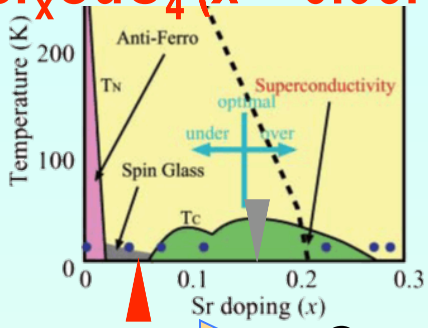
Electronic structure at the interface

Superconductor–Insulator Transition in La$_{2-x}$Sr$_x$CuO$_4$ Driven by Electric Field

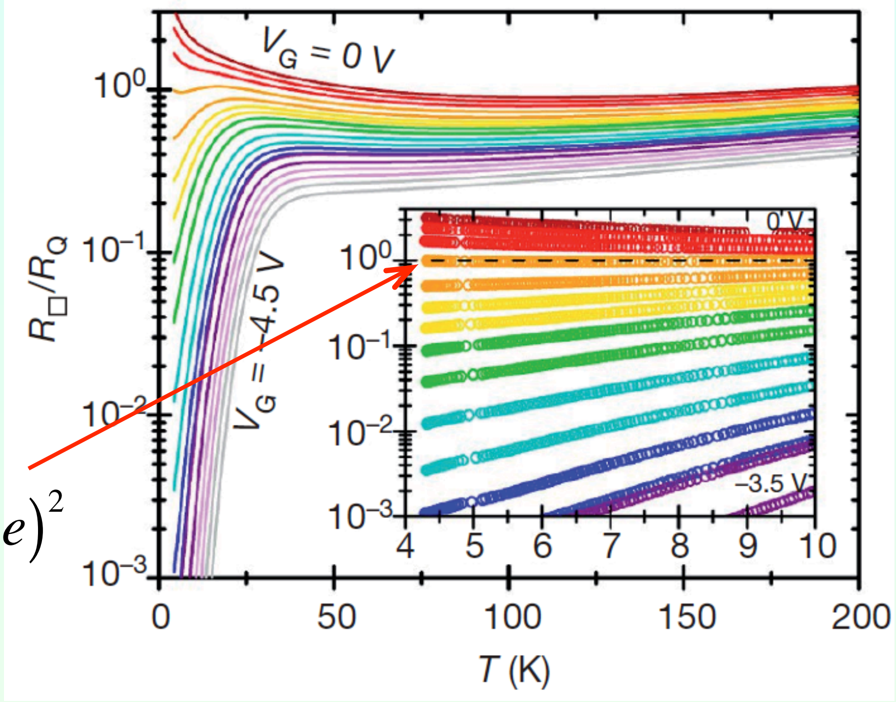
1 u.c. La$_{2-x}$Sr$_x$CuO$_4$ (x = 0.06: Under doped)



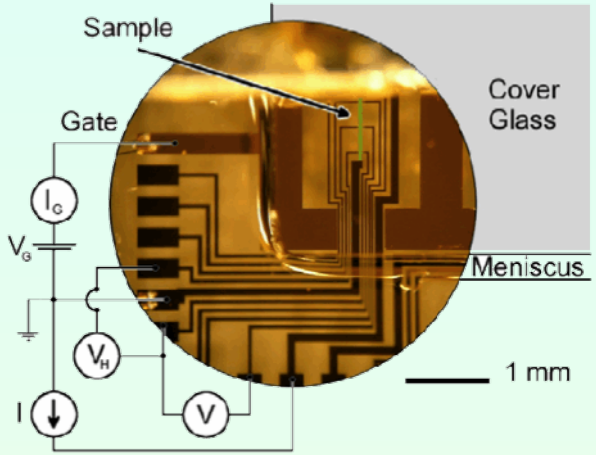




Controlling S.C.



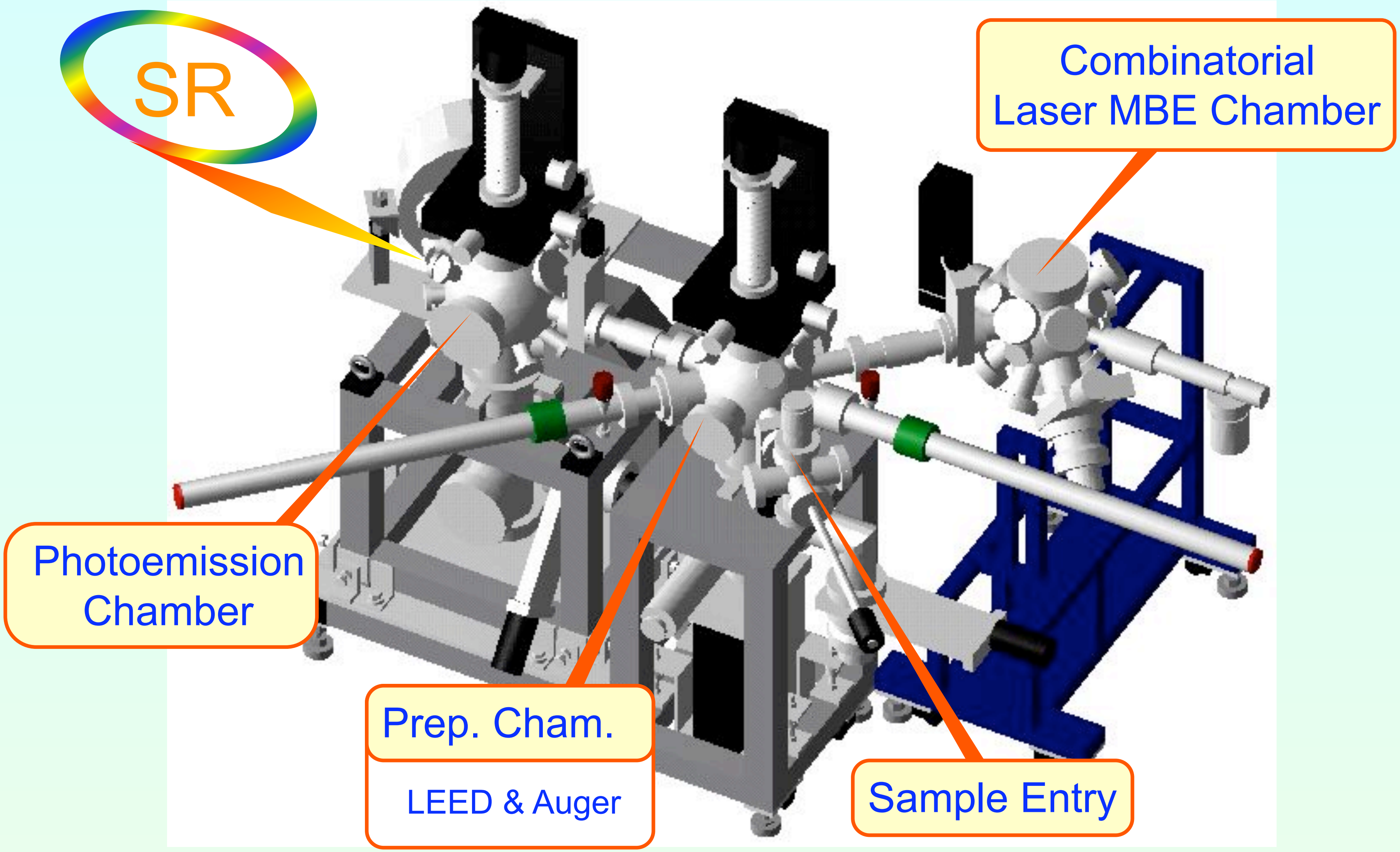
$h/(2e)^2$



Electrolyte Double-Layer Transistor
(Field Effect Transistor)

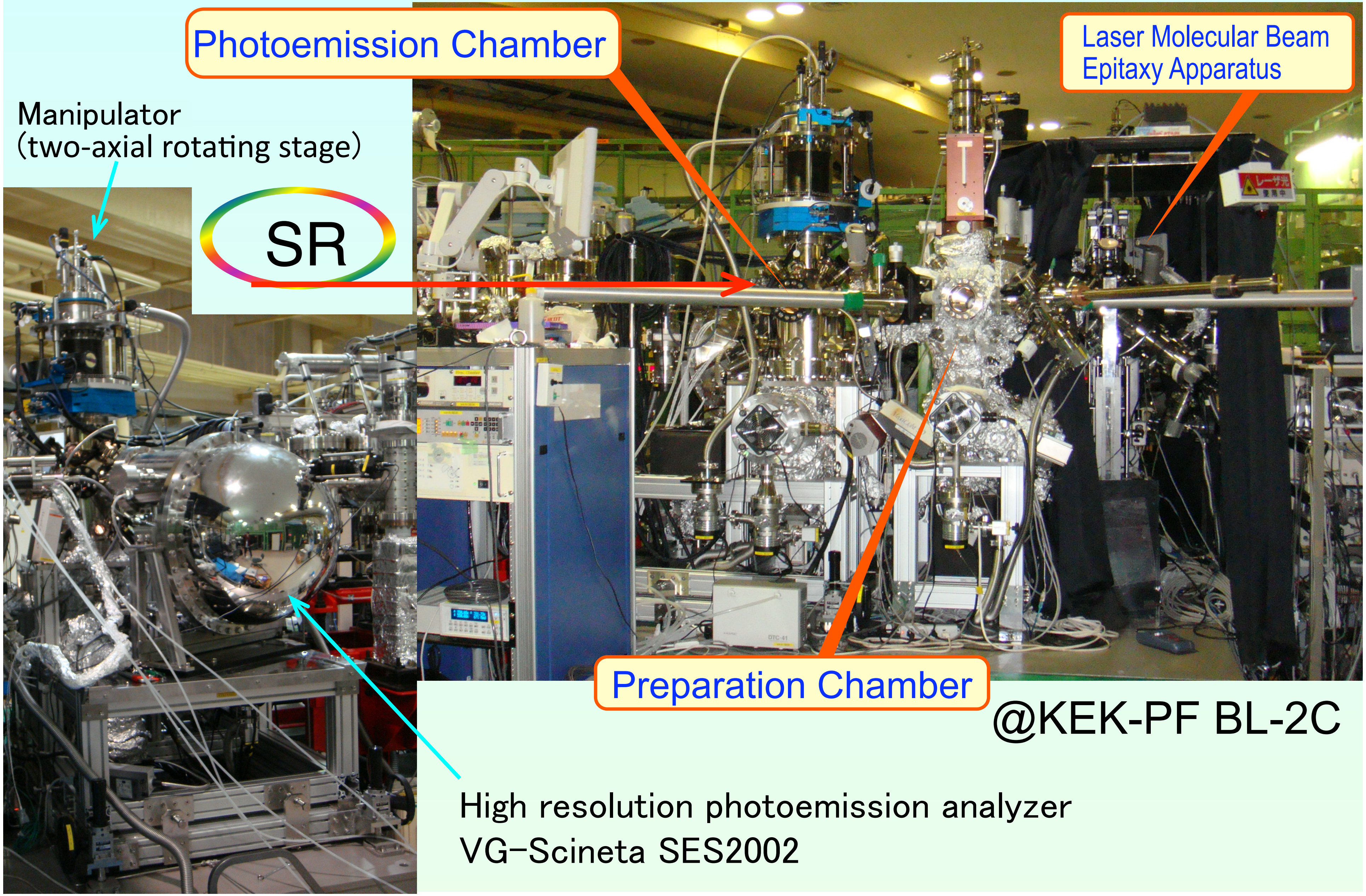
A.T. Bollinger et al., Nature **472**, 458 (2011).

In-situ PES + Laser MBE system



K. Horiba, HK *et al.*, Rev. Sci. Instrum. **74**, 3406 (2003).

New *in-situ* PES + Laser MBE system



Photoemission Chamber

Laser Molecular Beam
Epitaxy Apparatus

Manipulator
(two-axial rotating stage)

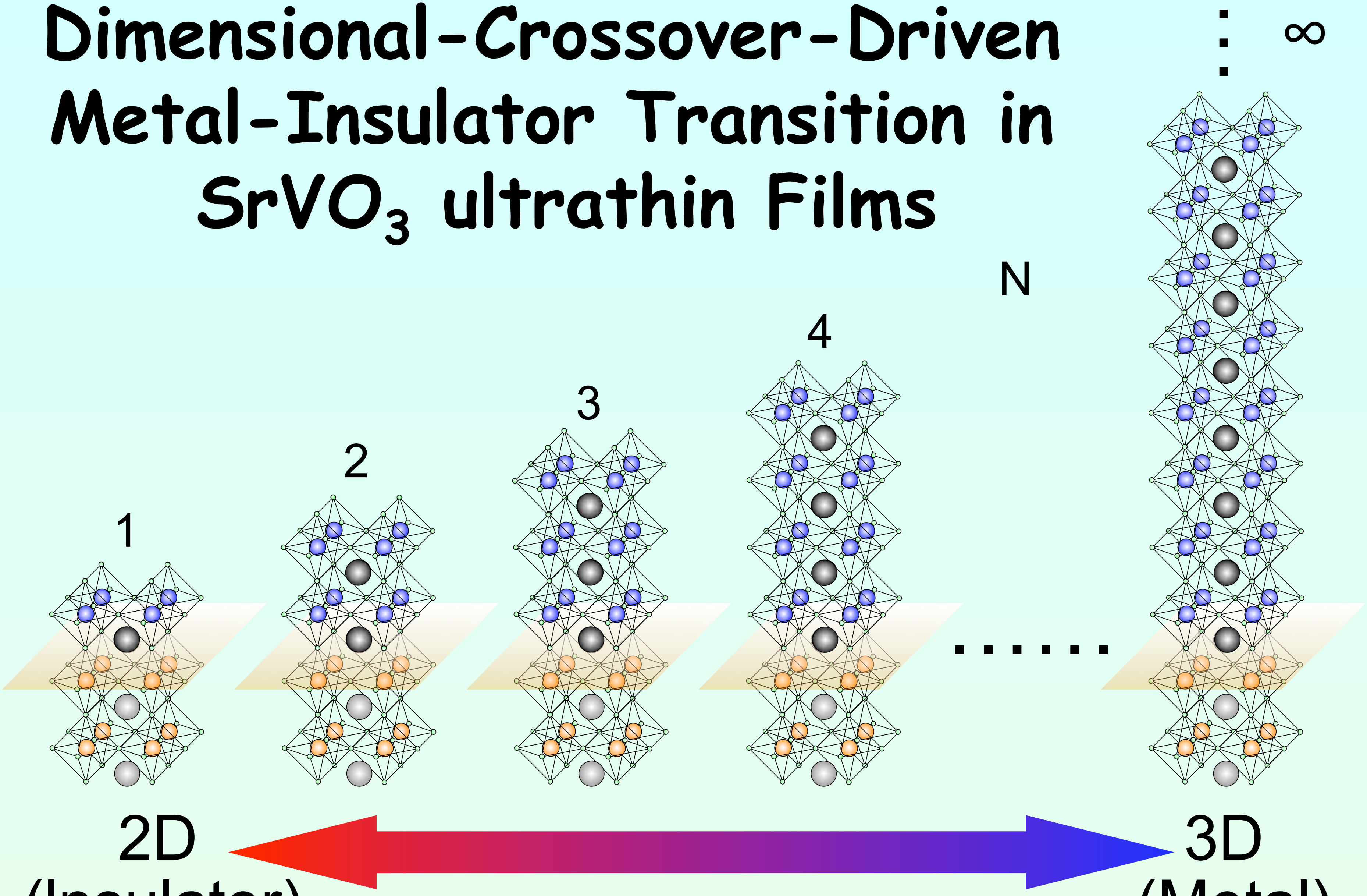
SR

Preparation Chamber

@KEK-PF BL-2C

High resolution photoemission analyzer
VG-Scineta SES2002

Dimensional-Crossover-Driven Metal-Insulator Transition in $SrVO_3$ ultrathin Films



2D
(Insulator)

3D
(Metal)

Metallic Quantum Well

K. Yoshimatsu, H.K., *et al*., *Phys. Rev. Lett.* **104**, 147601 ('10).
K. Yoshimatsu, H.K., *et al*., *Science* **333**, 315 ('11).

Metal Insulator Transition

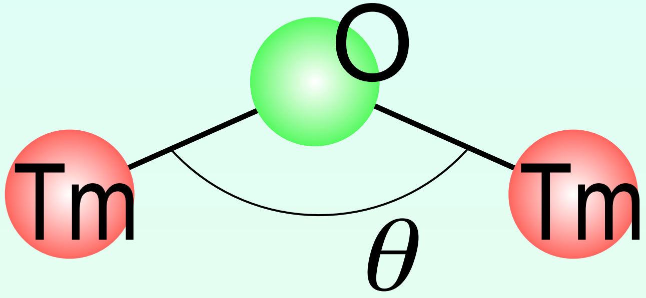
Mott-Hubbard theory

$U >> W$ Insulator

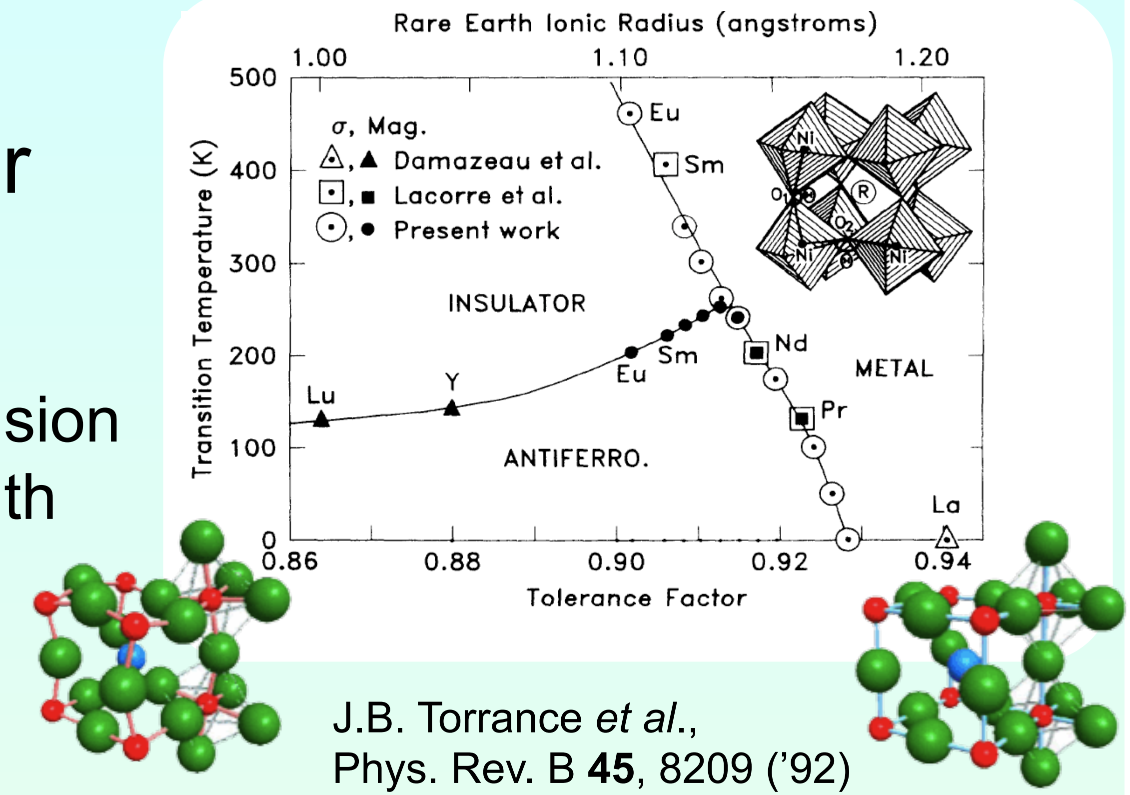
$U << W$ Metal

U : on-site Coulomb repulsion

W: one-electron band width



$W \propto \cos^2\theta$

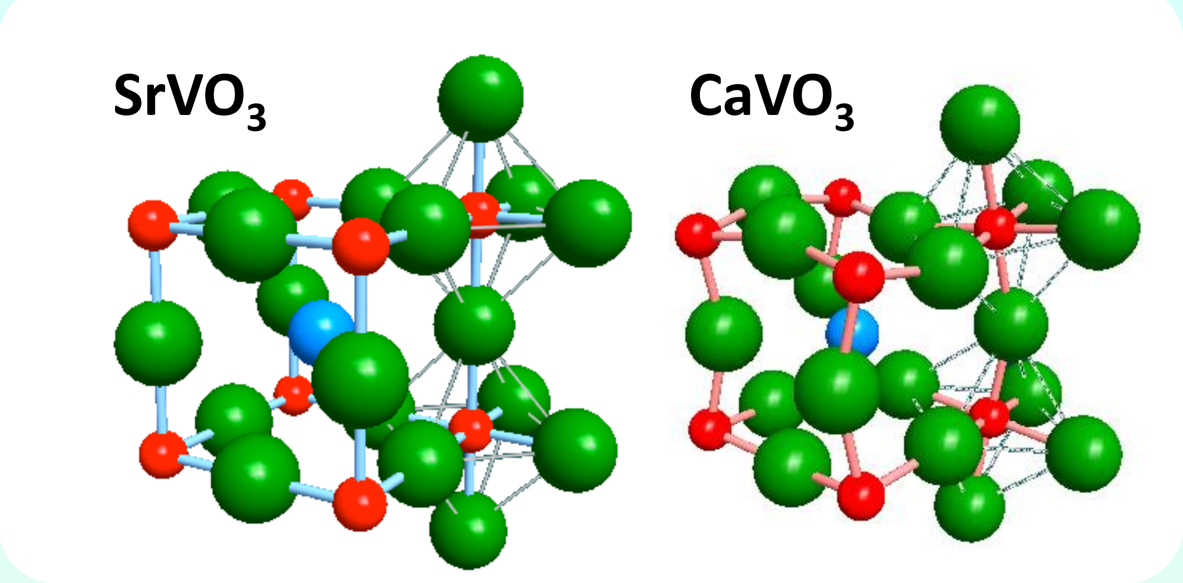


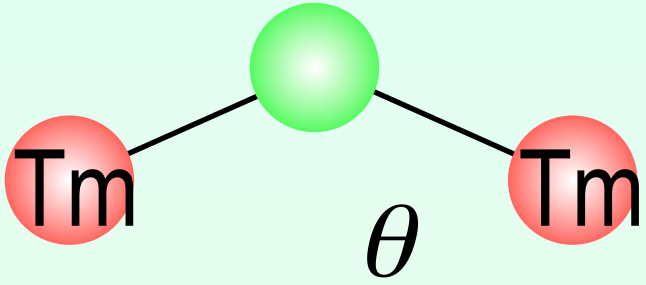
J.B. Torrance *et al.*,
Phys. Rev. B **45**, 8209 ('92)

For bulk materials, MIT has been intensively studied by chemical substitution of constituent ions with ones having a smaller ion radius. However, such chemical substitution always induce randomness in a solid.

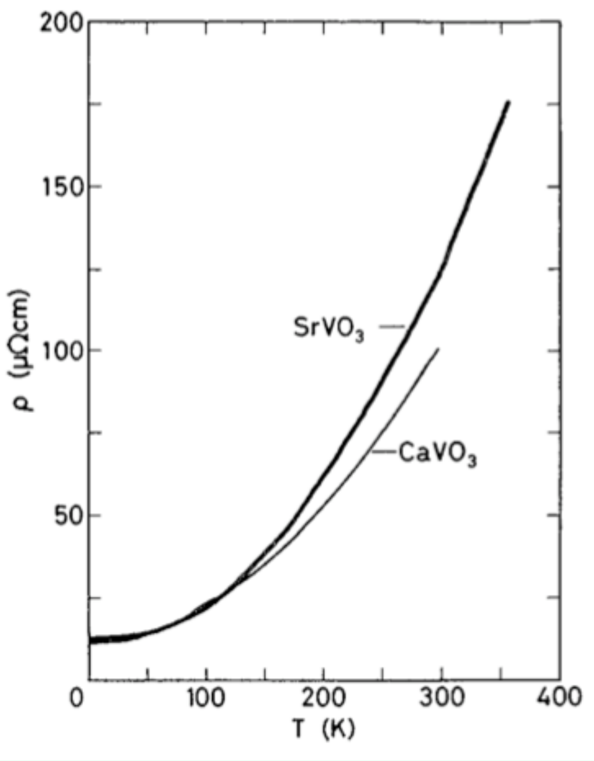
Bandwidth control in $Sr_{1-x}Ca_xVO_3$

$W = 12t$ $W = 10.6t$ $W \propto \cos^2\theta$





●	A	Sr^{2+}, Ca^{2+}
●	B	V^{4+} (d^1)
●	O	O^{2-}



M. Onoda *et al.*, Solid State Comun. **79**, 281 (1991).

θ :
$SrVO_3$ 180°
$CaVO_3$ ~160°

$Sr_{1-x}Ca_xVO_3$ is always metal in the entire *x* range.

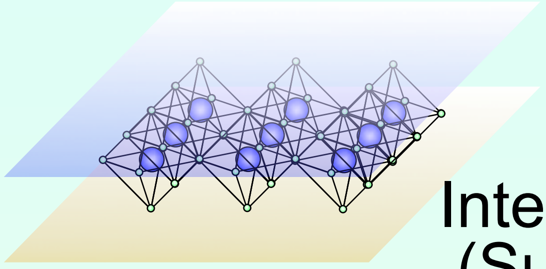
Our Approach: Dimensional crossover occurring in an

Digitally-controlled

2D

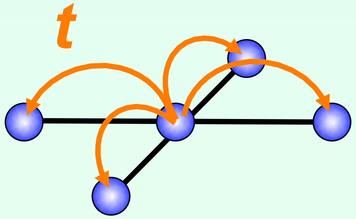
SrVO$_3$: Paramagnetic metal
3d^1 system

1 ML



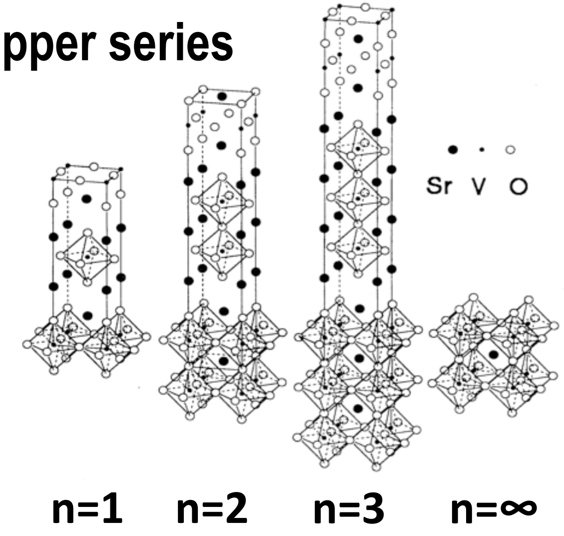
Surface
(Vacuum)

Interface
(Substrate)

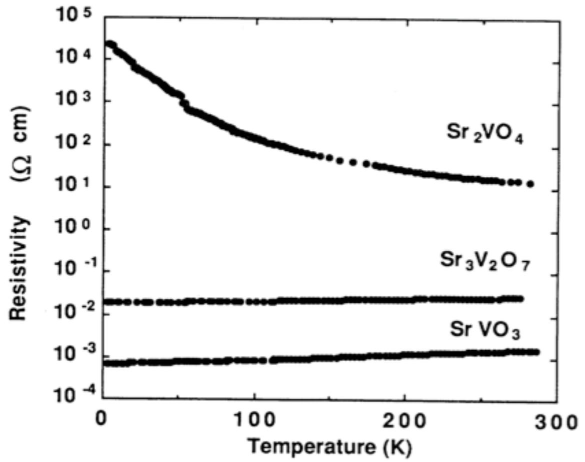


$W = 8t$

$A_{n+1}B_nO_{3n+1}$
Ruddlesden-Popper series



n = 1
Insulator



n > 2
Metal

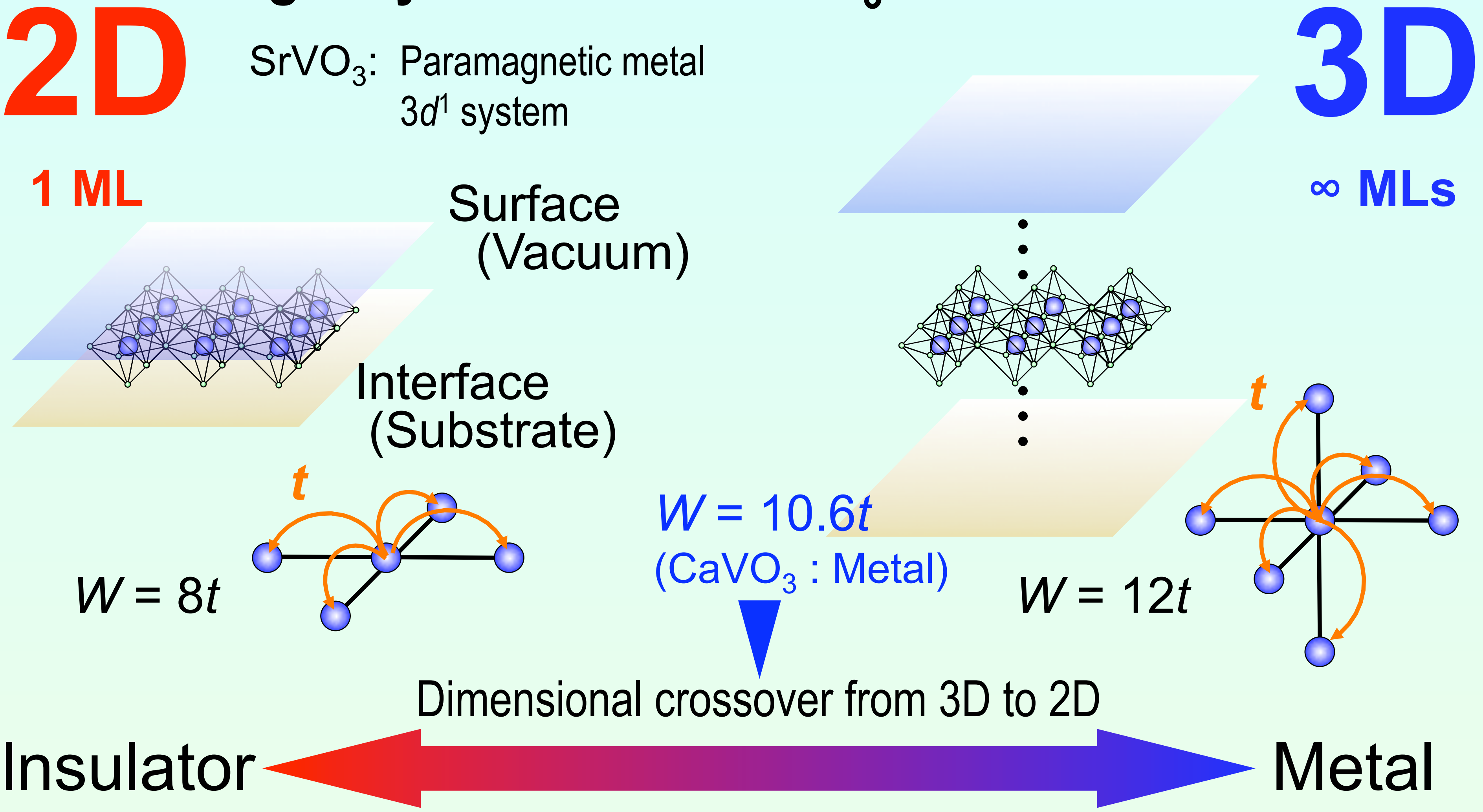
A. Nozaki *et al*., Phys. Rev. B **43**, 181 (1991).

Dimensional crossover from 3D to 2D

Insulator ⟷ Metal

Our Approach: Dimensional crossover occurring in an artificial structure

Digitally-controlled $SrVO_3$ ultrathin films



2D

1 ML

$SrVO_3$: Paramagnetic metal
$3d^1$ system

Surface
(Vacuum)

Interface
(Substrate)

t

$W = 8t$

$W = 10.6t$
($CaVO_3$: Metal)

3D

∞ MLs

t

$W = 12t$

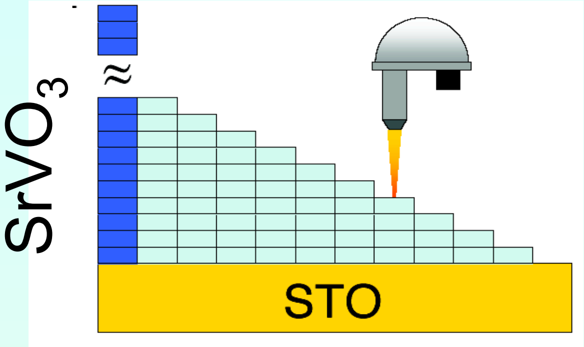
Dimensional crossover from 3D to 2D

Insulator

Metal

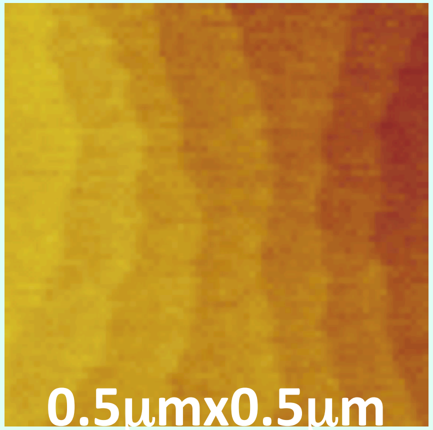
Atomically-flat Surface and Abrupt Interface

Preconditions to thickness dependent experiments

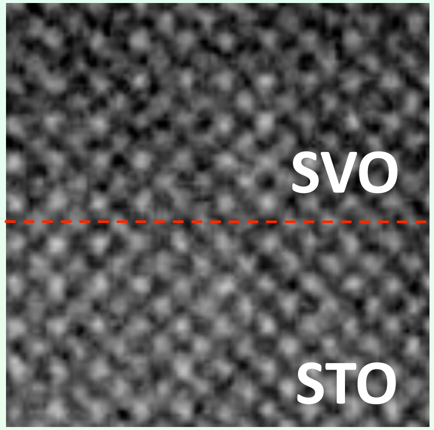


- ✓ **Precise digital control of thickness (RHEED oscillation)**
- ✓ **Atomically flat surface (AFM image)**
- ✓ **Chemically abrupt interface (TEM image)**
- ✓ **Coherent growth of thin film (Reciprocal space mapping)**

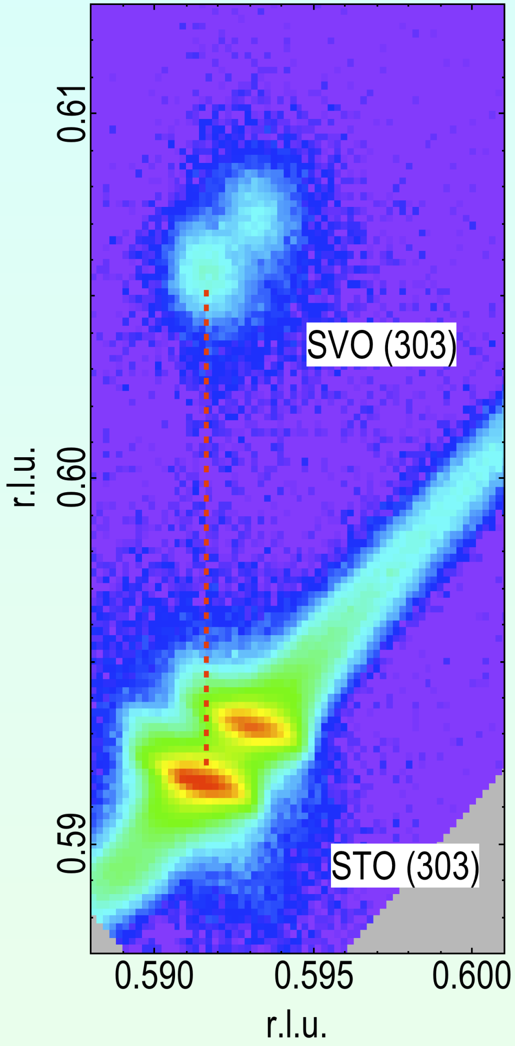
AFM image



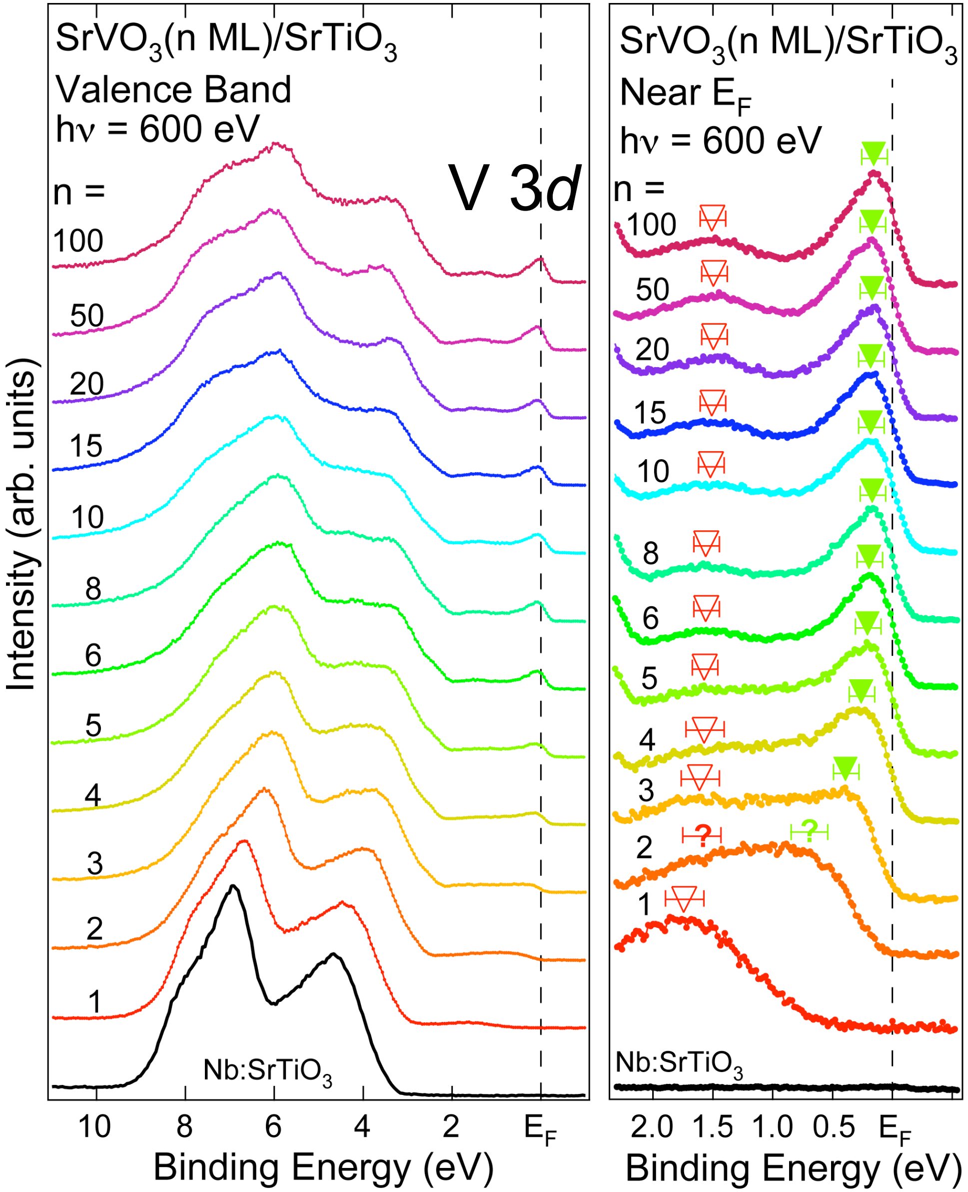
TEM image

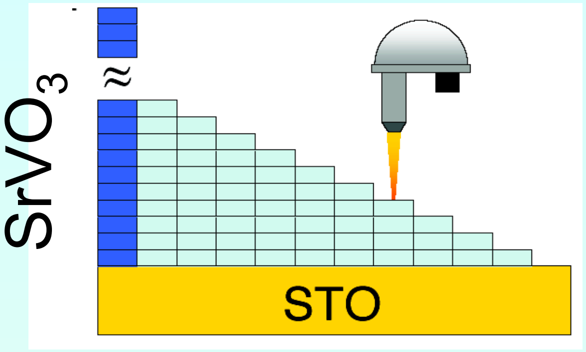


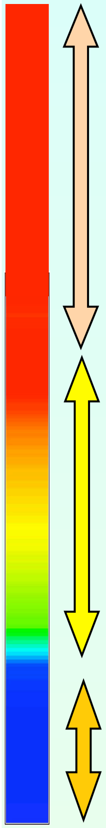
4cXRD RSM around (303)



In situ PES Spectra of $SrVO_3$ thin Films







Spectra remain unchanged down to 6–8 ML

Pseudogap formation at E_F

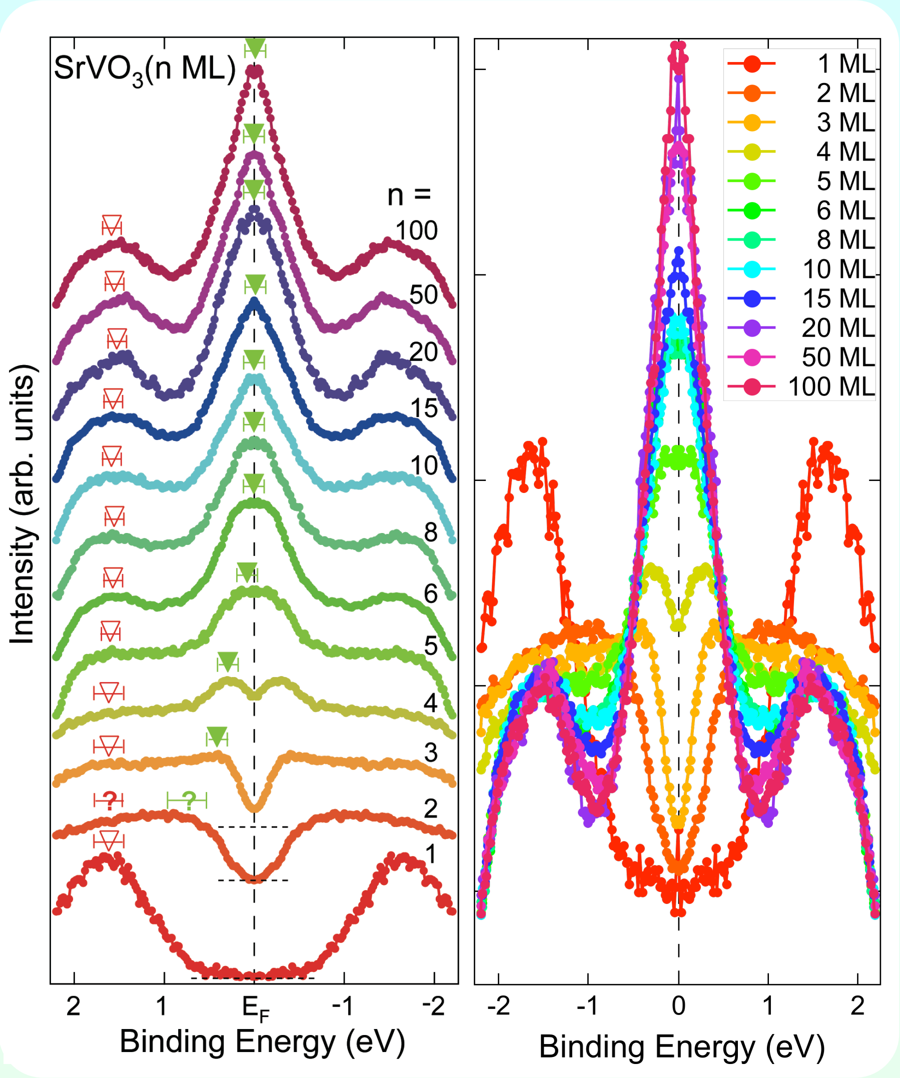
Disppearance of Fermi edge

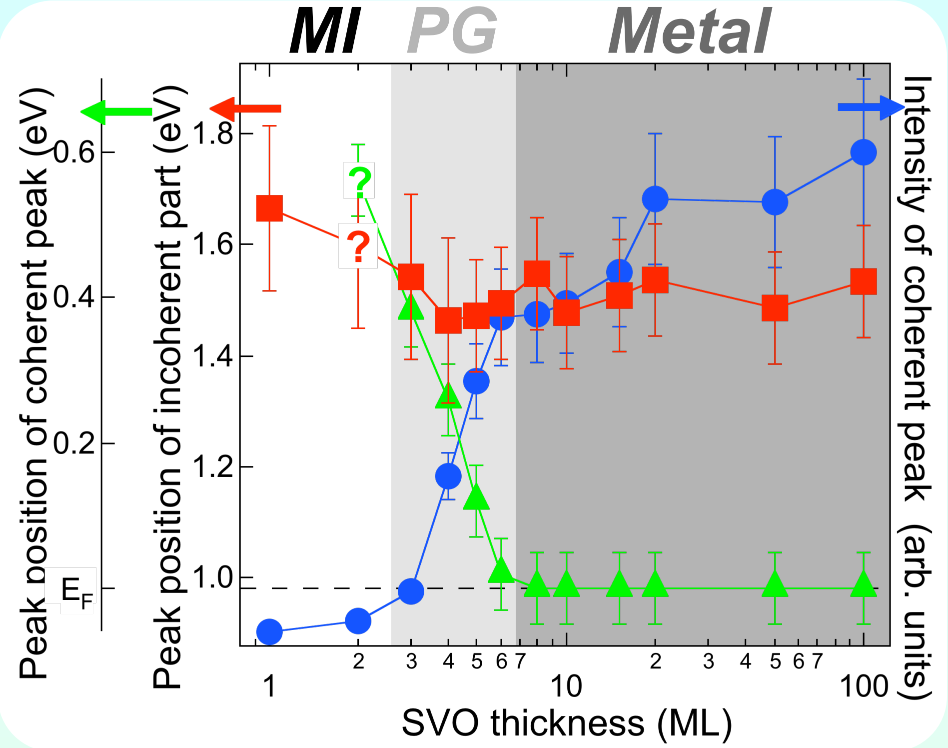
Gap at E_F

MIT@2-3 ML

Metal-insulator transition occurs in the SVO thickness of 2-3 ML.

Dimensional-Crossover-Driven MIT in SrVO$_3$ Films



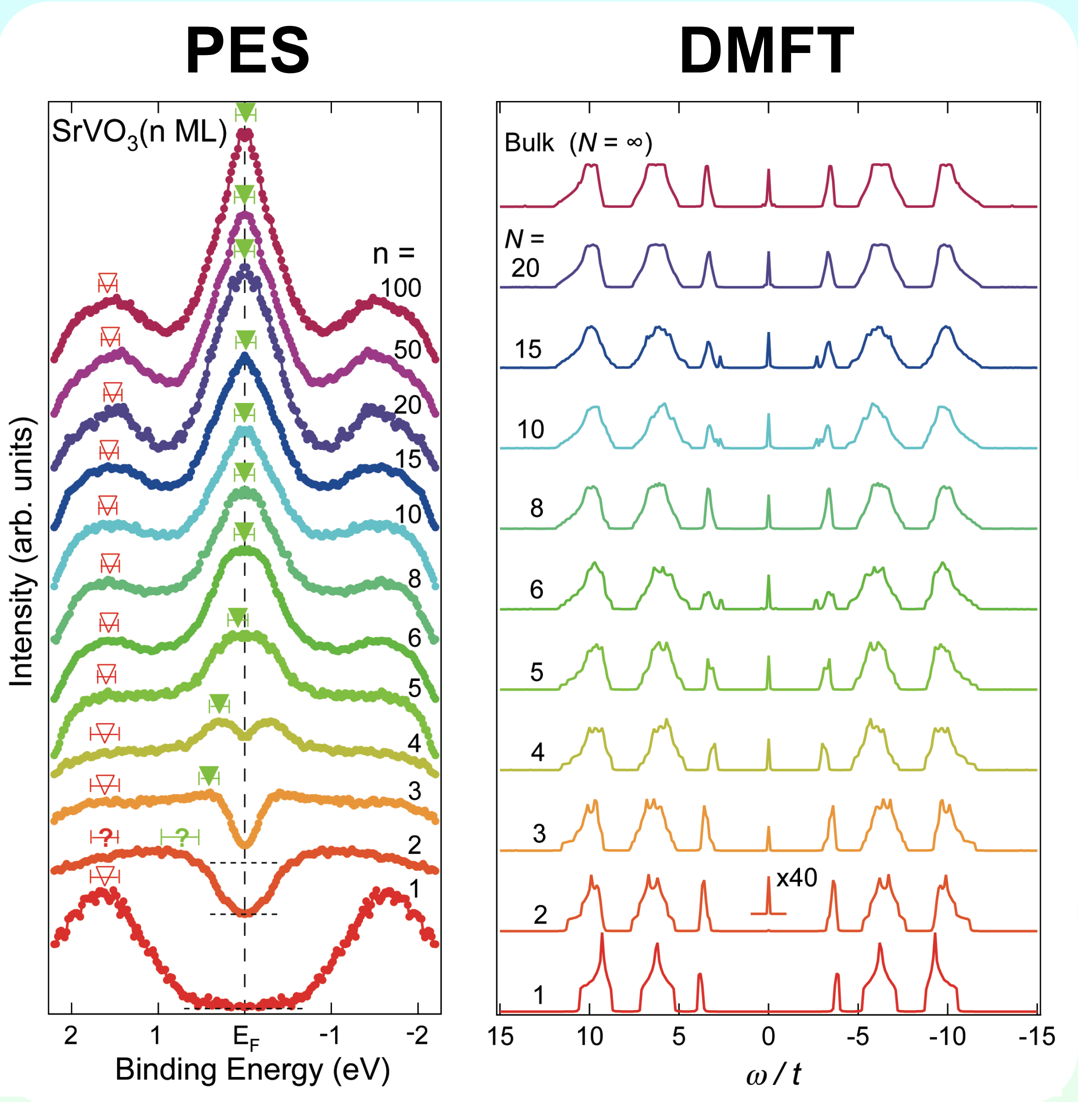


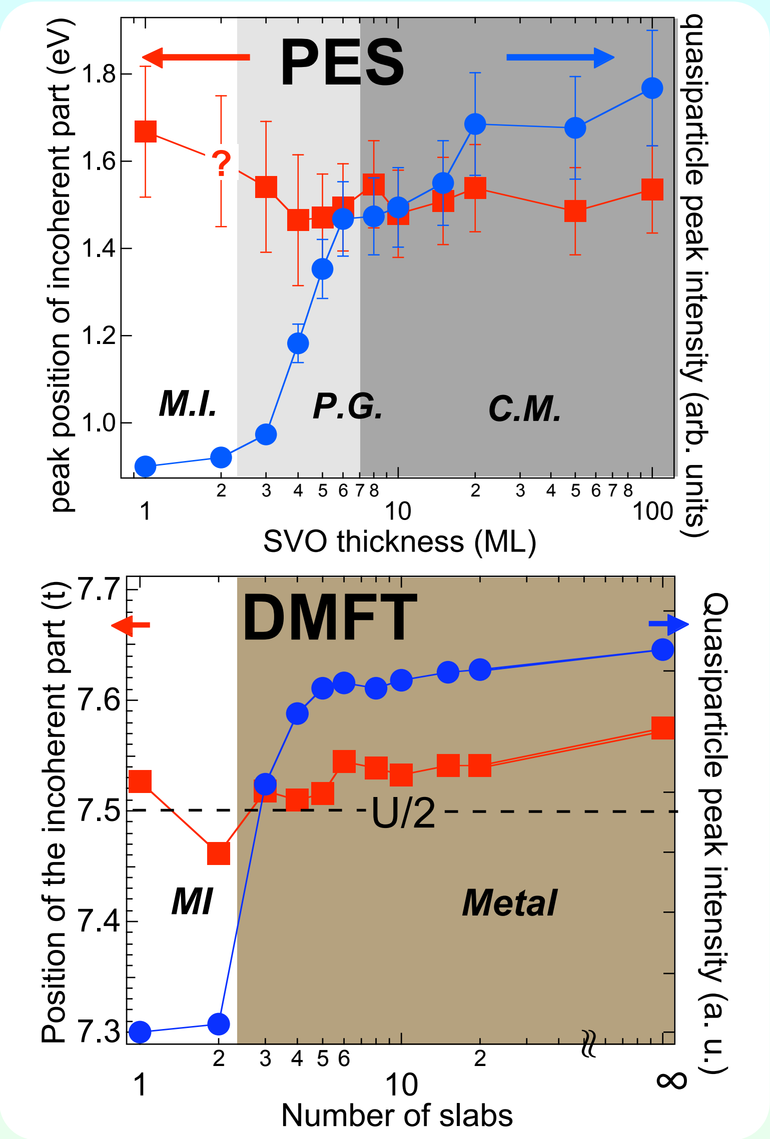
Quasiparticle peak intensity steeply increasing with increasing film thickness (*W* increasing).

Peak position of the incoherent peak remains unchanged (*U* is constant).

Dimensional-Crossover-Driven (from 3D to 2D) MIT in SrVO$_3$ Ultrathin Films

Comparison between PES and Layer DMFT Cal.





K. Yoshimatsu, H.K., *et al*., Phys. Rev. Lett. **104**, 147601 ('10).

Spectral behavior is well reproduced by layer DMFT calculation.

Summary: $SrVO_3$ ultrathin films

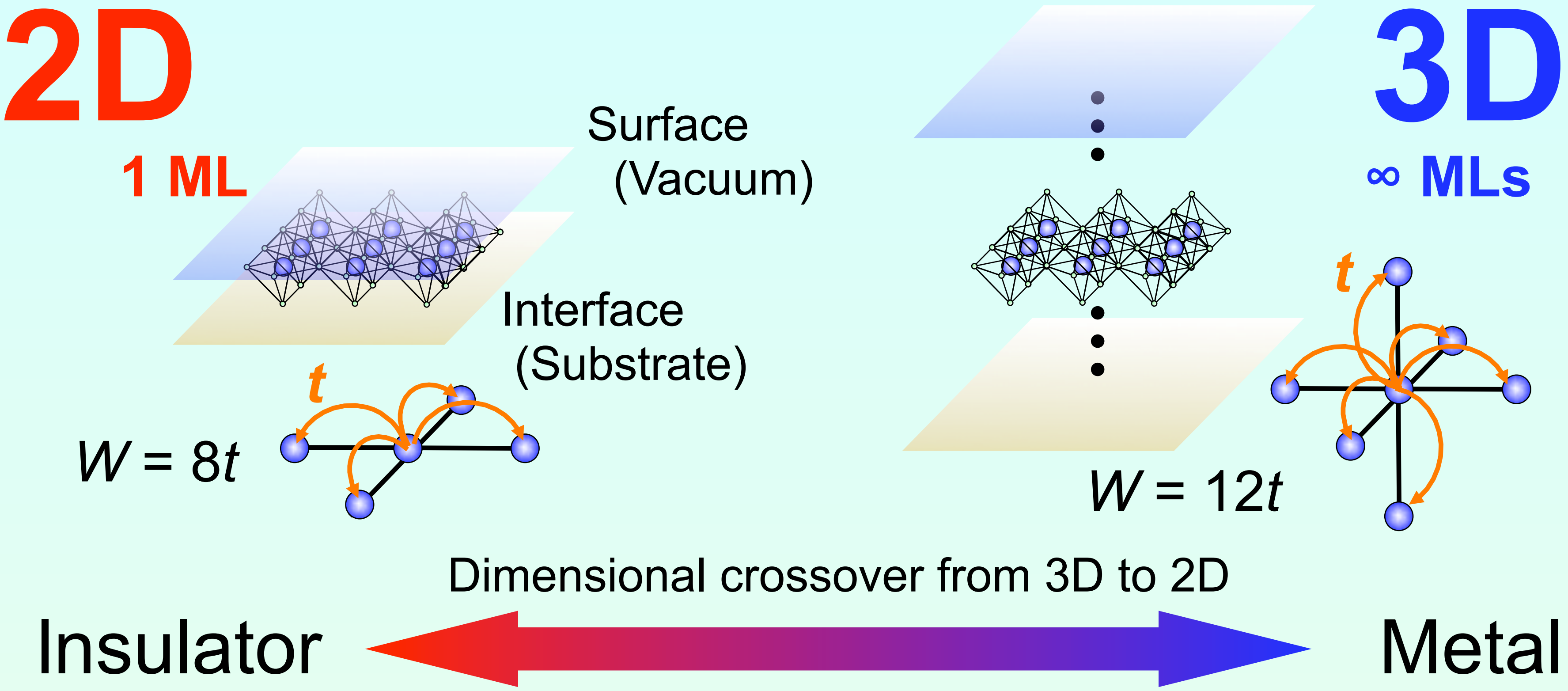
We have investigated the change occurring in the electronic structure of digitally controlled $SrVO_3$ ultrathin films across the dimensional-crossover-driven MIT using *in-situ* photoemission spectroscopy.

We have found

1. The V 3*d* derived states near E_F remain unchanged down to 6–8 ML.
2. In the film thickness of 3–6 ML, a pseudogap is formed at E_F owing to the spectral weight transfer from the coherent part to incoherent part.
3. The pseudogap finally evolves into an energy gap below 2 ML, indicating the occurrence of MIT at a critical thickness of 2–3 ML.
4. These spectral behaviors are well reproduced by layer DMFT calculations.

⇨ The observed MIT is caused by the reduction of bandwidth due to dimensional-crossover from 3D to 2D.

Summary



2D

1 ML

Surface (Vacuum)

Interface (Substrate)

t

$W = 8t$

3D

∞ MLs

t

$W = 12t$

Dimensional crossover from 3D to 2D

Insulator

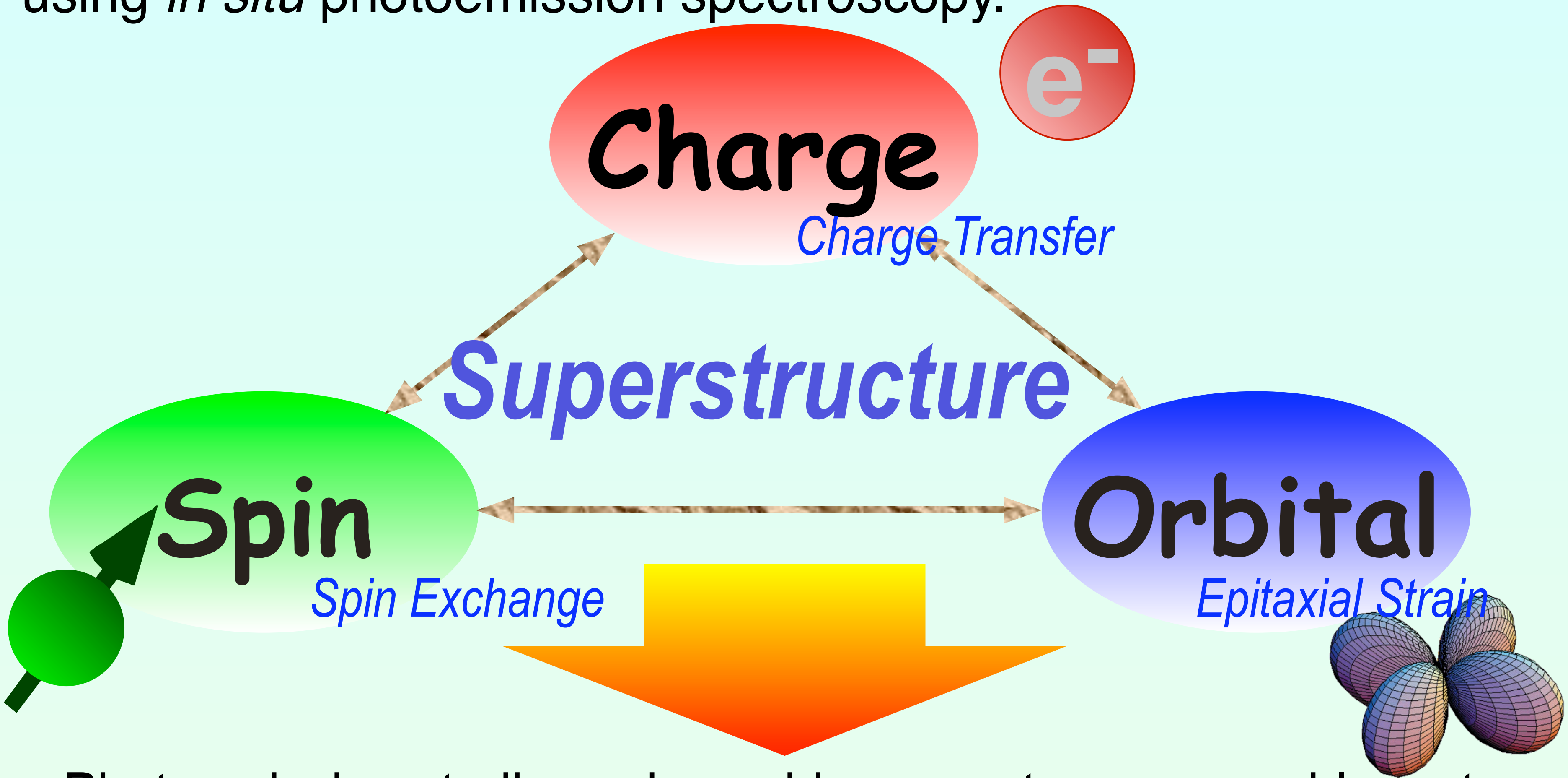
Metal

The observed MIT is caused by the reduction of bandwidth due to dimensional-crossover from 3D to 2D.

Ubiquity of thickness-dependent MIT in conductive oxide films are derived from the dimensional-crossover.

Concluding Remarks

We studied the electronic states of oxide superstructures by using *in situ* photoemission spectroscopy.



e^-

Charge

Charge Transfer

Superstructure

Spin

Spin Exchange

Orbital

Epitaxial Strain

Photoemission studies using oxide superstrucures enable us to pave a new way for the better understanding of the physics of strongly correlated oxides.