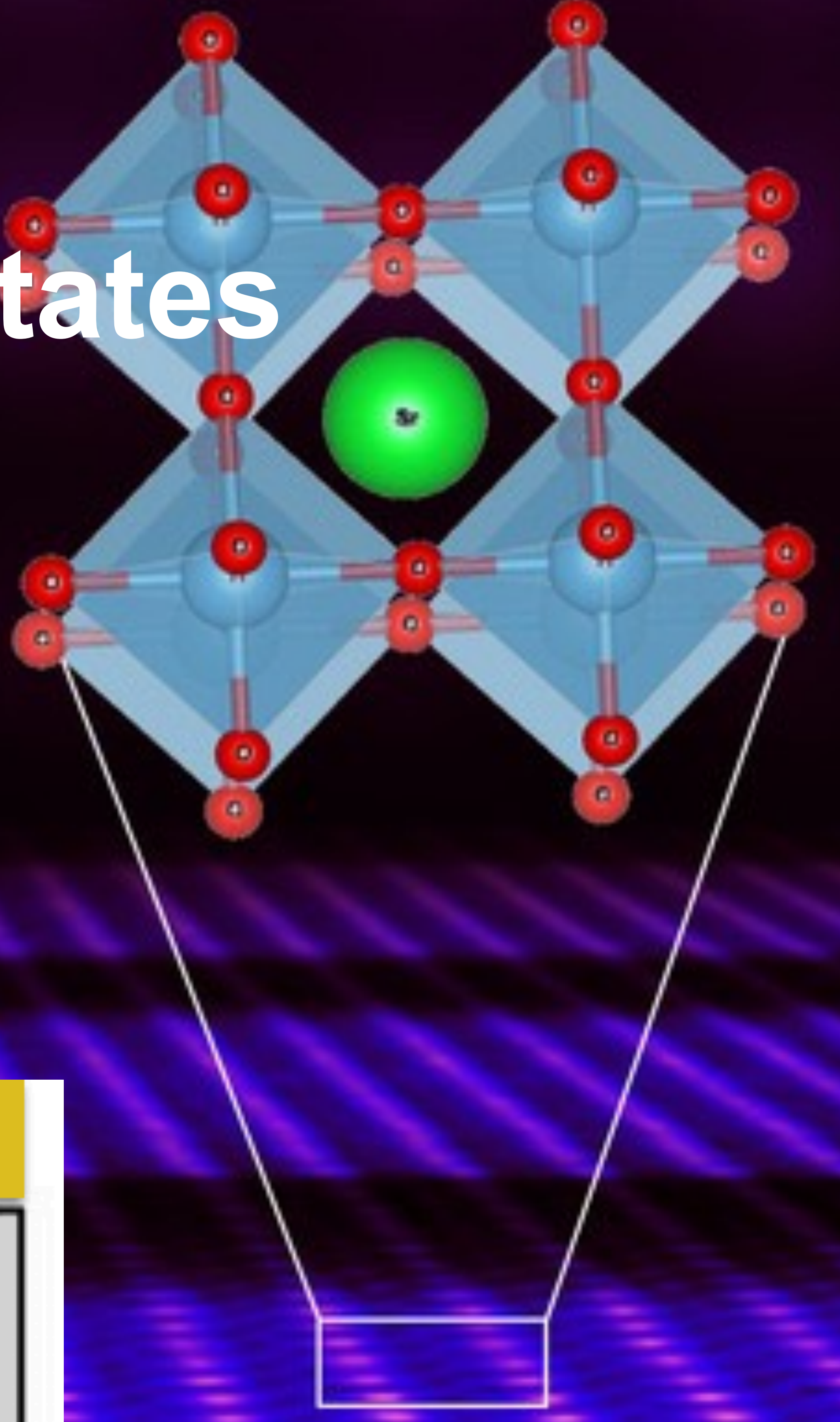
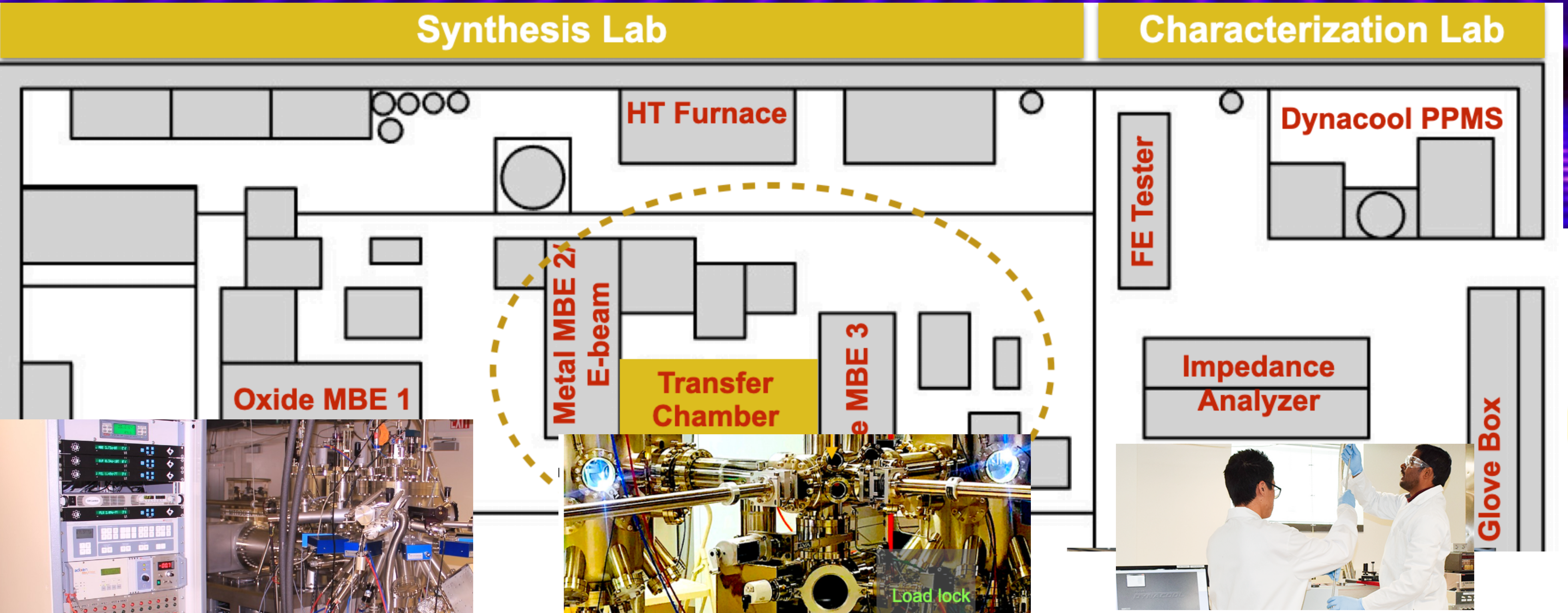
Emerging Altermagnetism and Polar Metallic States in Epitaxial RuO_2 Films

Bharat Jalan
Professor

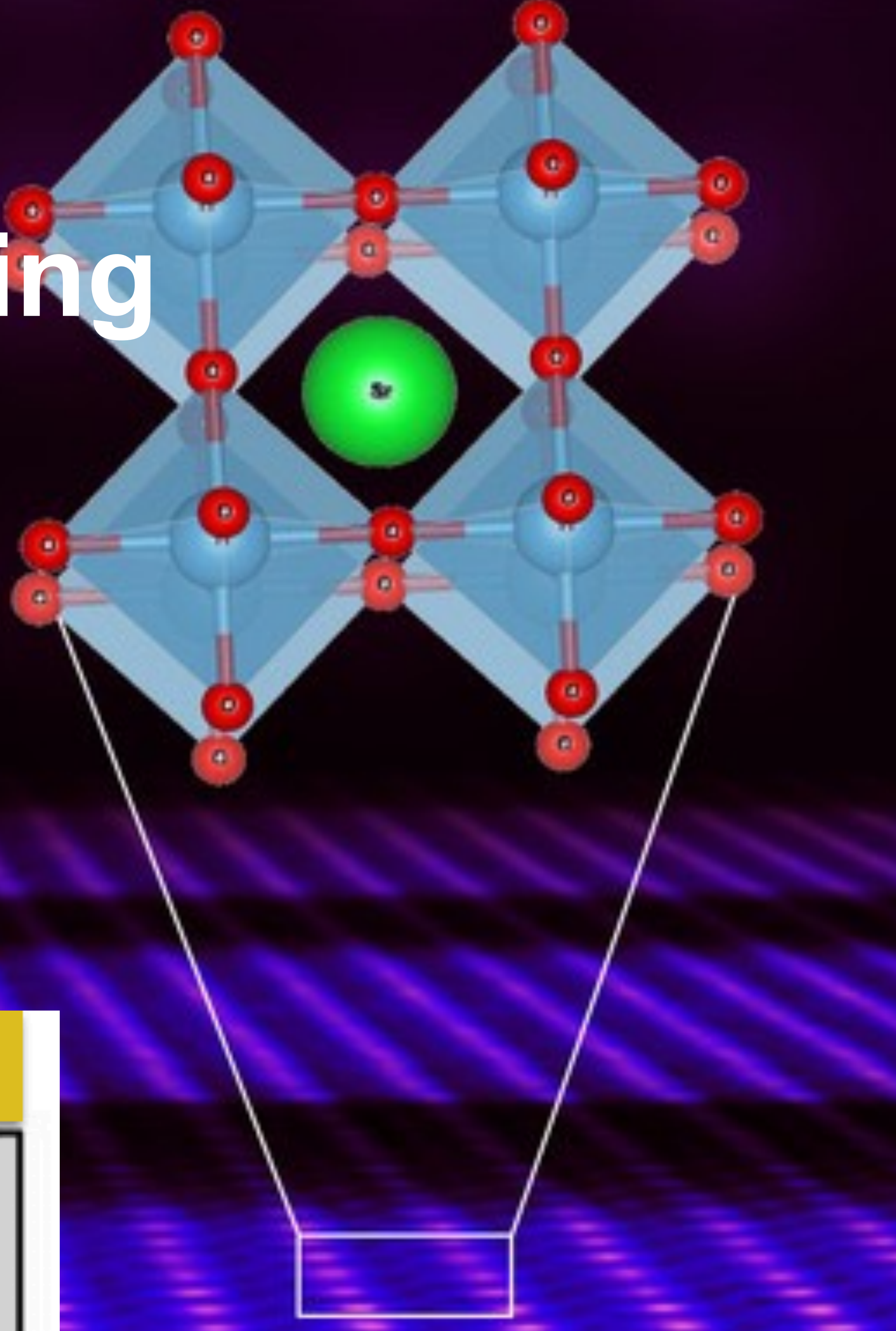
Department of Chemical Engineering and Materials Science
University of Minnesota, Twin Cities





Acknowledgements

Adding materials-specific reality to the ongoing discussion of magnetism in RuO_2



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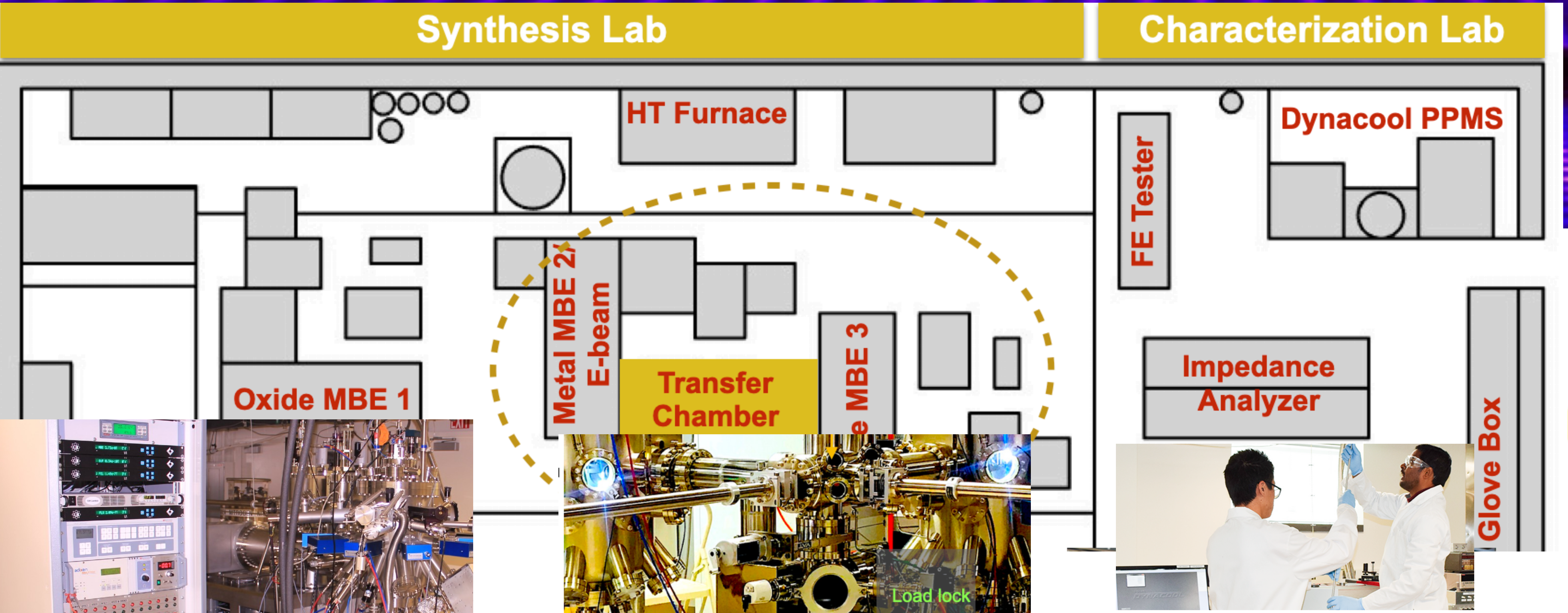
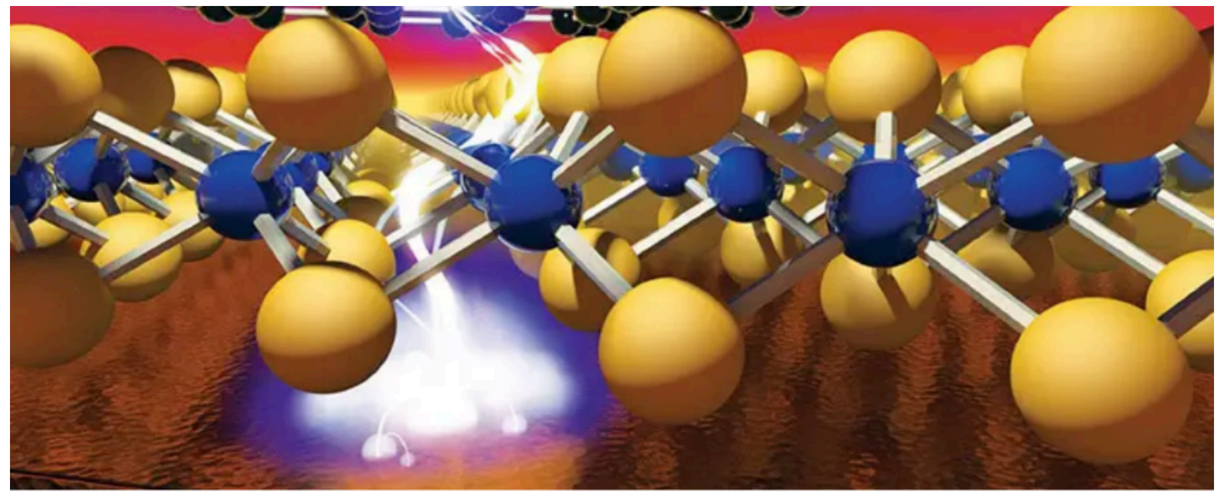


Acknowledgements

RuO_2: An Emerging Quantum Material



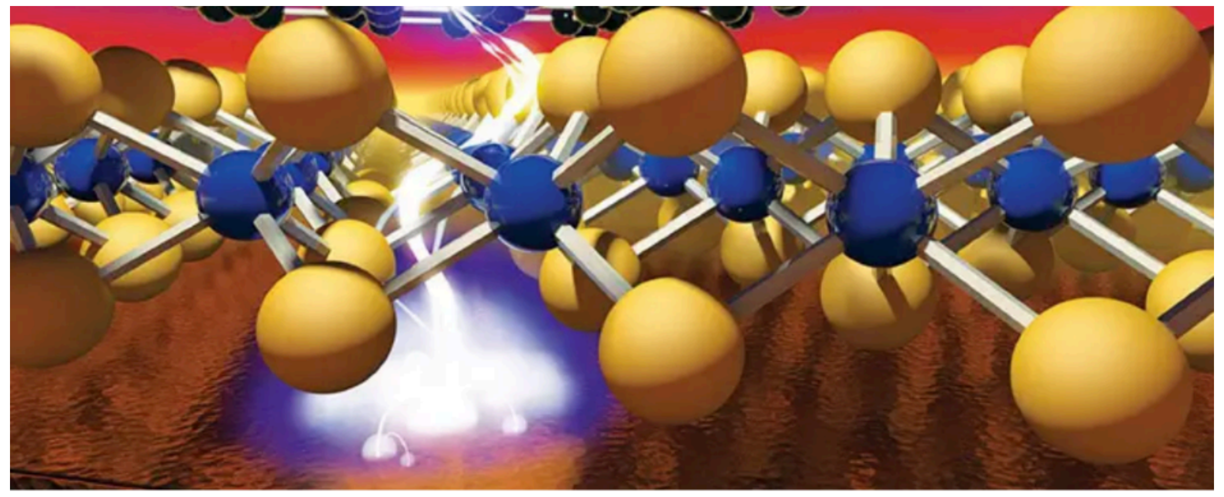
Bulk

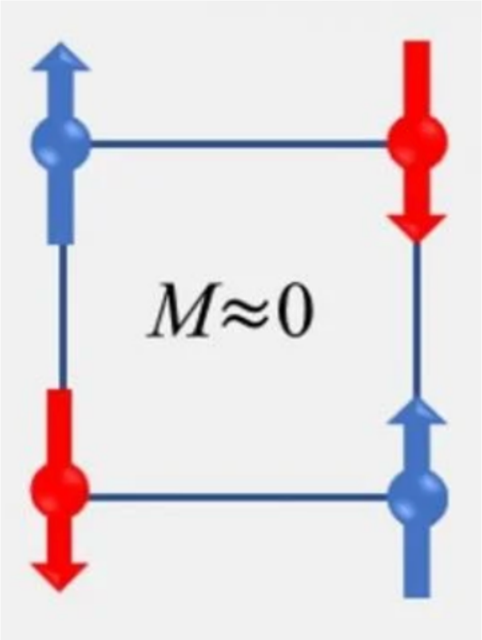
- Catalysis
- Stable electrodes

20th century

Film

RuO_2: An Emerging Quantum Material





Bulk

- 20th century: Catalysis; Stable electrodes
- 2017: Antiferromagnetism (Neutron diffraction)
- 2018: Dirac nodal lines
- 2019
- 2020: Crystal Hall effect (Theory)
- 2021
- 2022
- 2023

Film

Bulk studies:
J. Chem. Phys. 52, 6058 (1970)
Electrochim. Acta. 22, 513 (1977)
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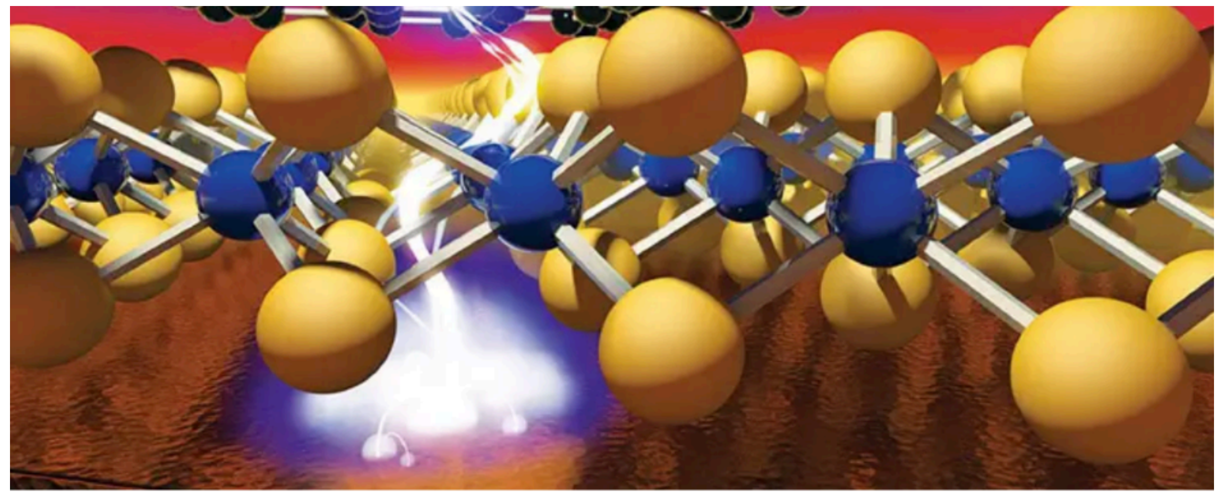
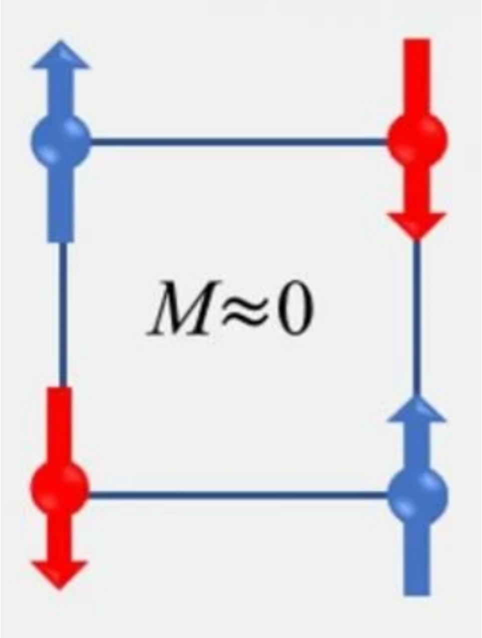
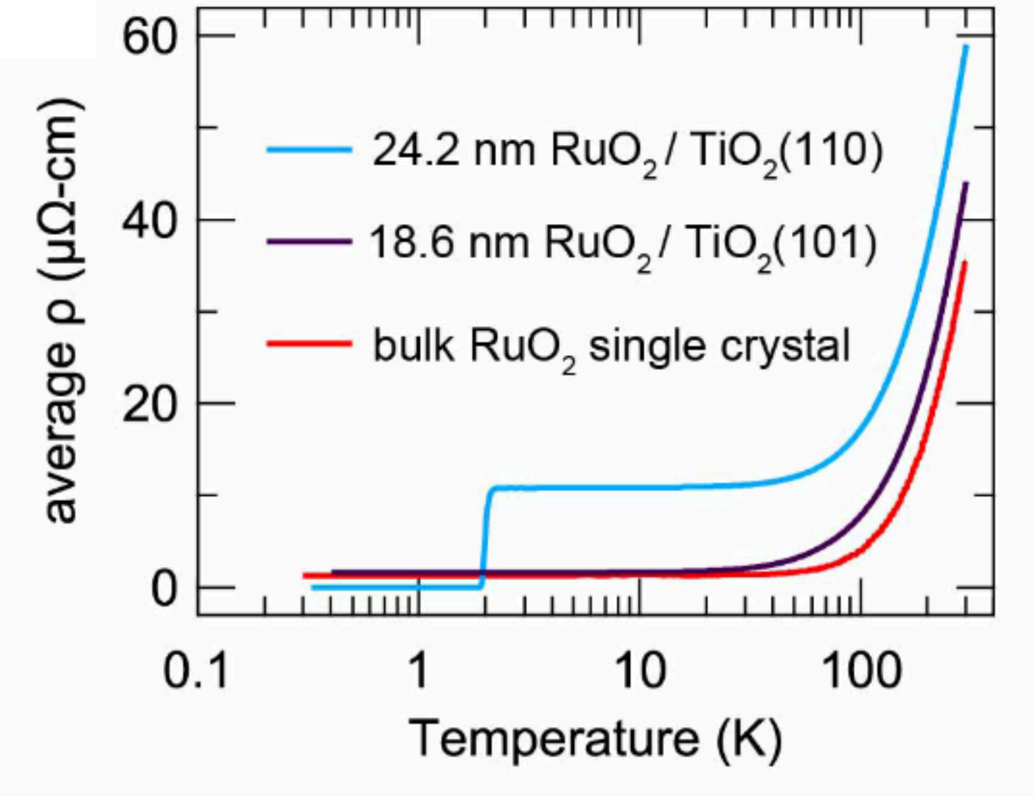




Schlom group, Cornell and Kawasaki group, U Tokyo



Bulk

- Catalysis
- Stable electrodes

20th century

Antiferromagnetism (Neutron diffraction) — 2017

Dirac nodal lines — 2018

2019 — Antiferromagnetism (X-ray scattering)

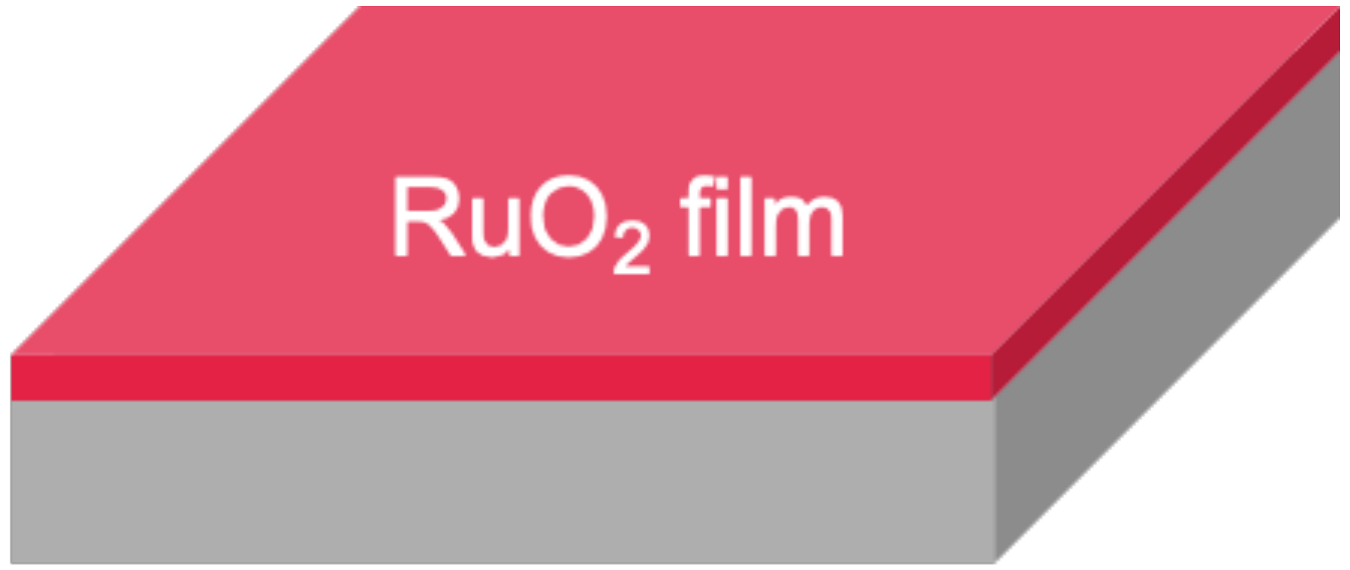
Crystal Hall effect (Theory) — 2020 — Superconductivity

2021

2022

2023

Film



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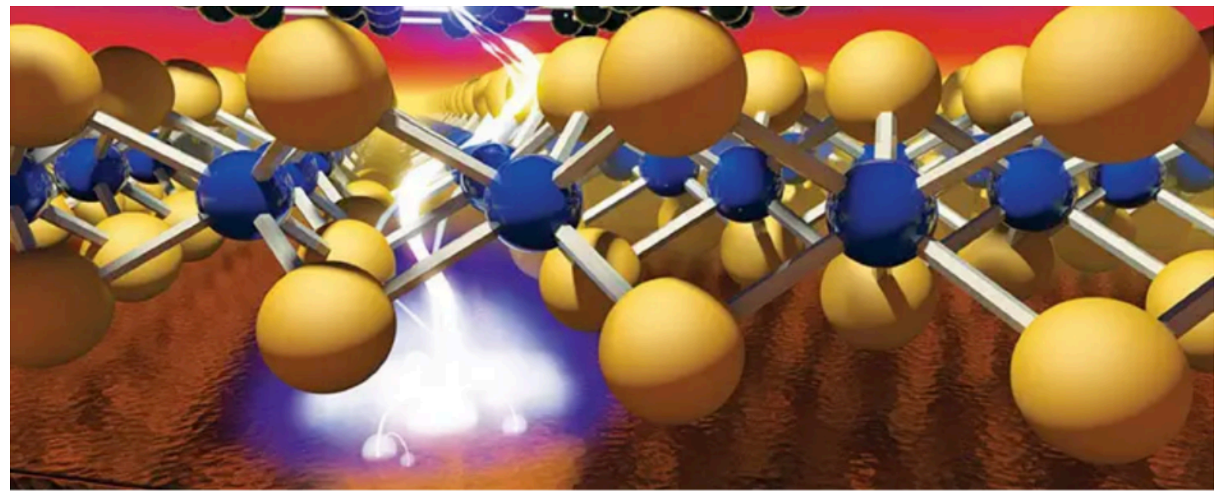

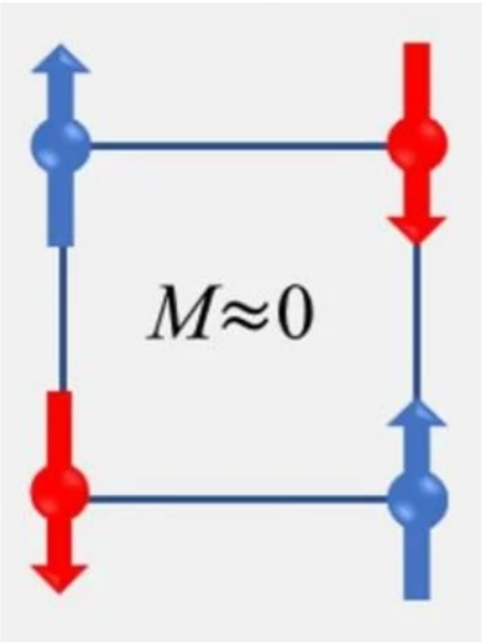

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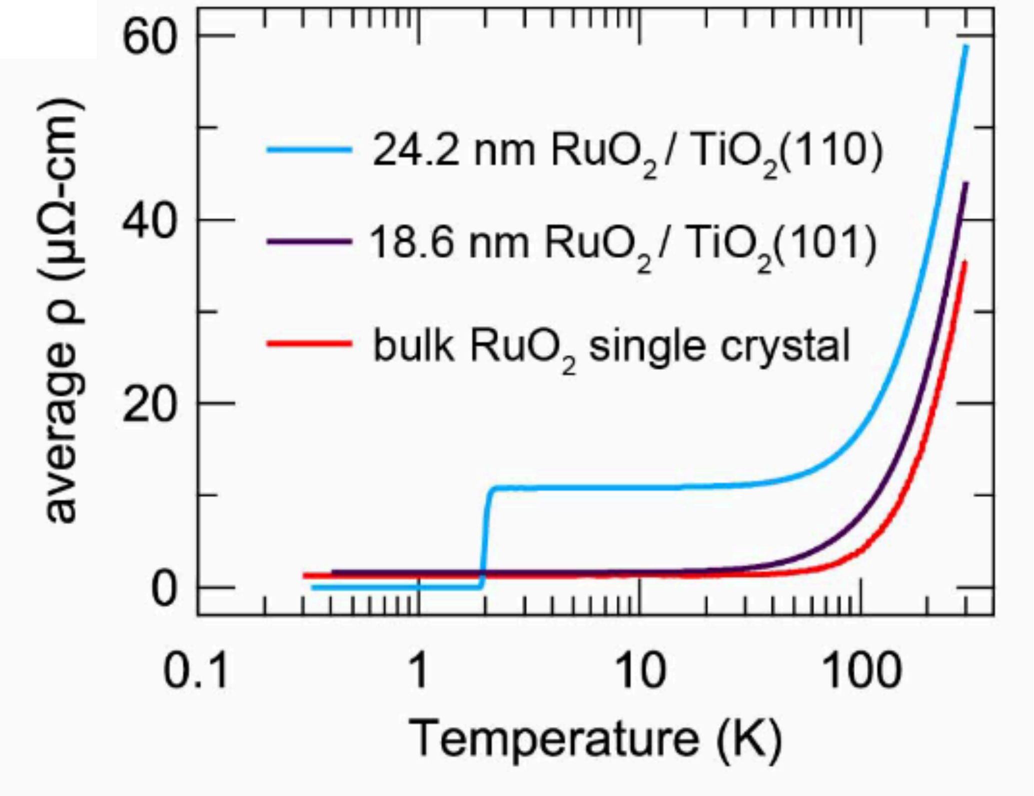

Bulk

- 20th century: Catalysis; Stable electrodes
- 2017: Antiferromagnetism (Neutron diffraction)
- 2018: Dirac nodal lines
- 2020: Crystal Hall effect (Theory)
- 2021: Large tunnel magneto-resistance (Theory)

Film

- 2019: Antiferromagnetism (X-ray scattering)
- 2020: Superconductivity
- 2022: Anomalous Hall effect; Spin-spitter torque
- 2023: Spin Seebeck effect
- 2024: Exchange bias; Spin density wave; Spin splitting band; Magnetic circular dichroism; Ultrafast magneto-optical Kerr effect; Magnetic octupole

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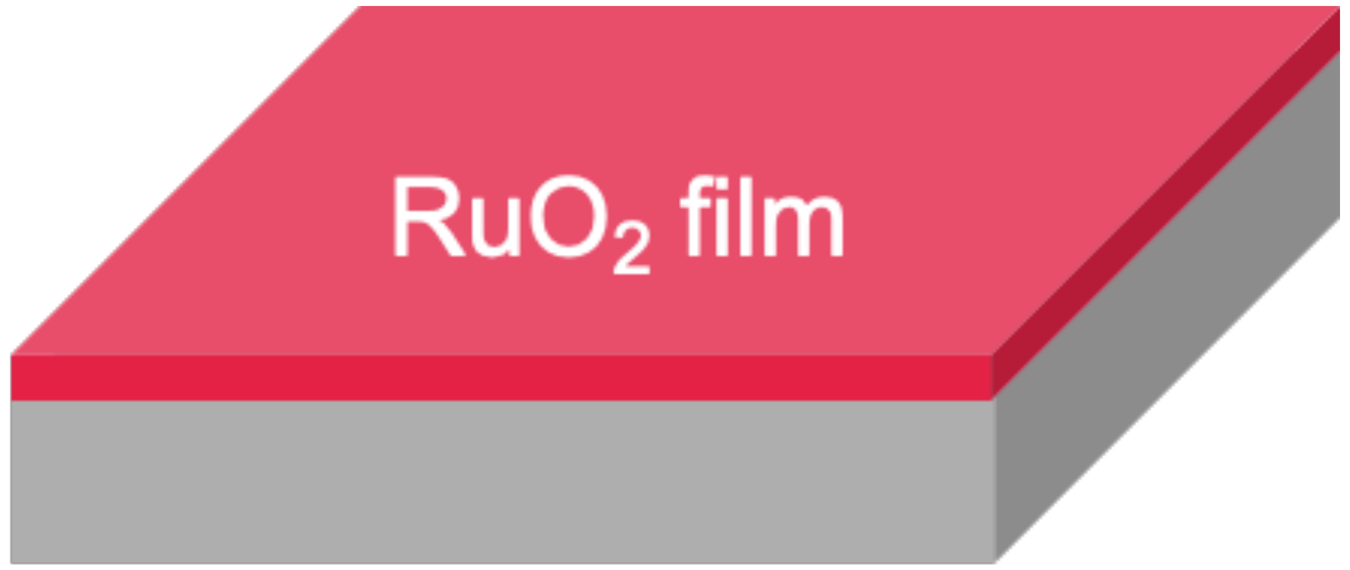

RuO_2: An Emerging Quantum Material

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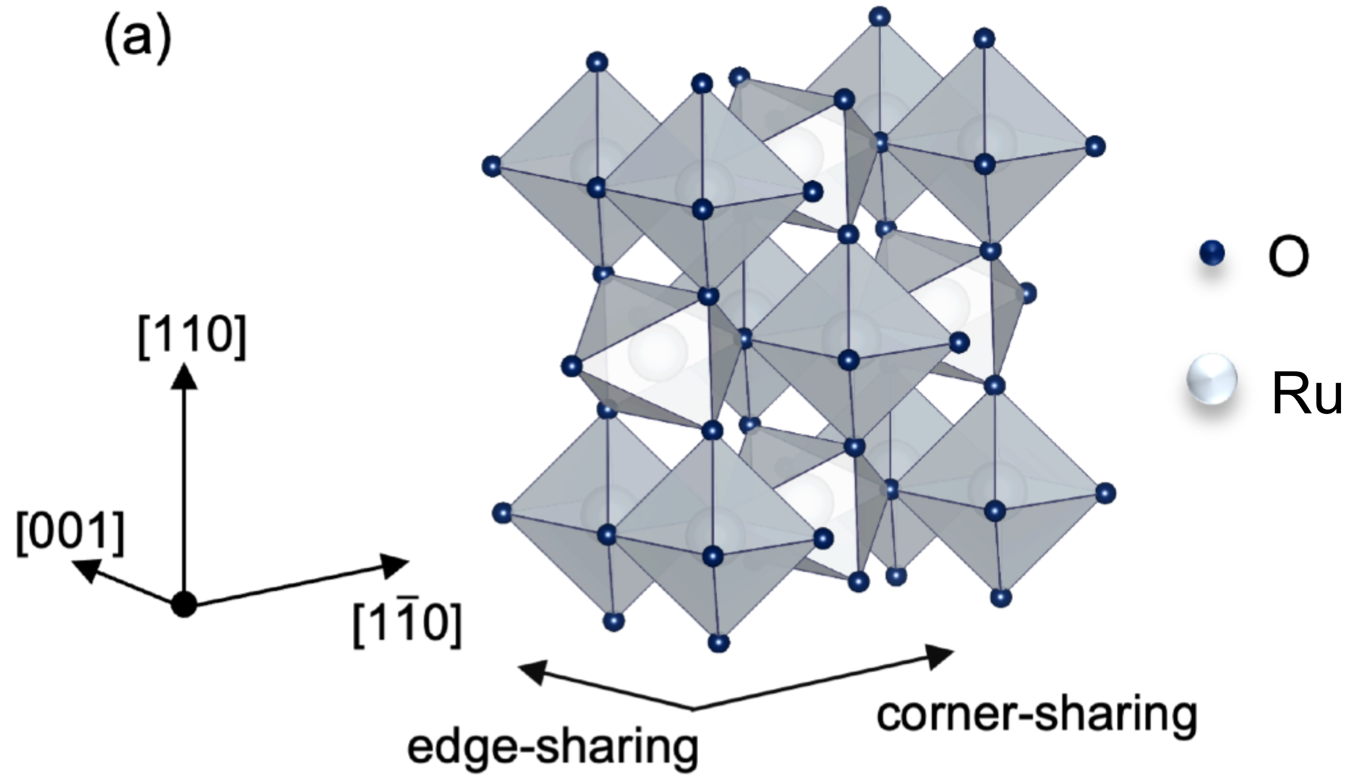

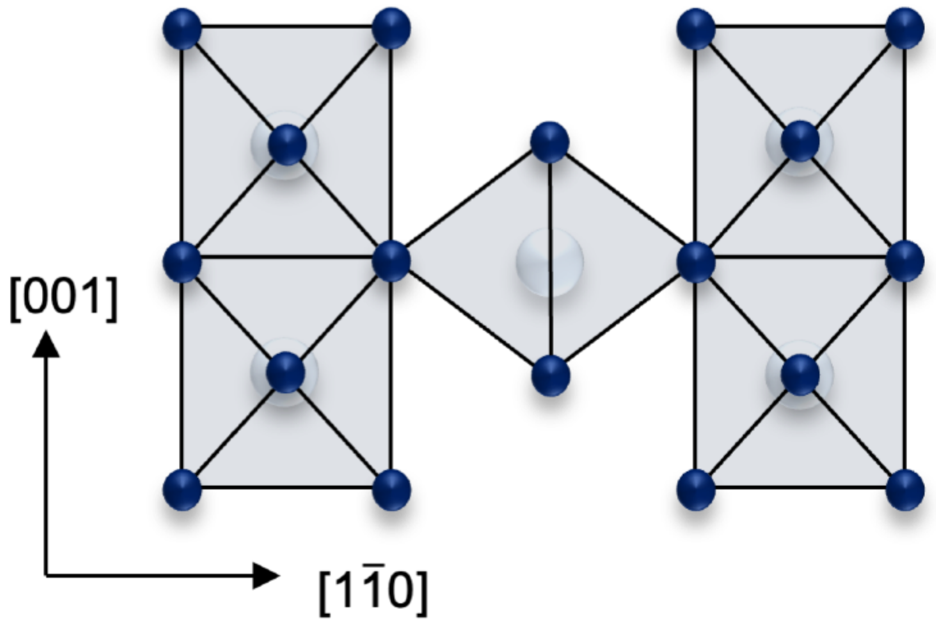

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RuO_2: An Emerging Quantum Material

- Nonmagnetic RuO_2
- Magnetic RuO_2

Bulk

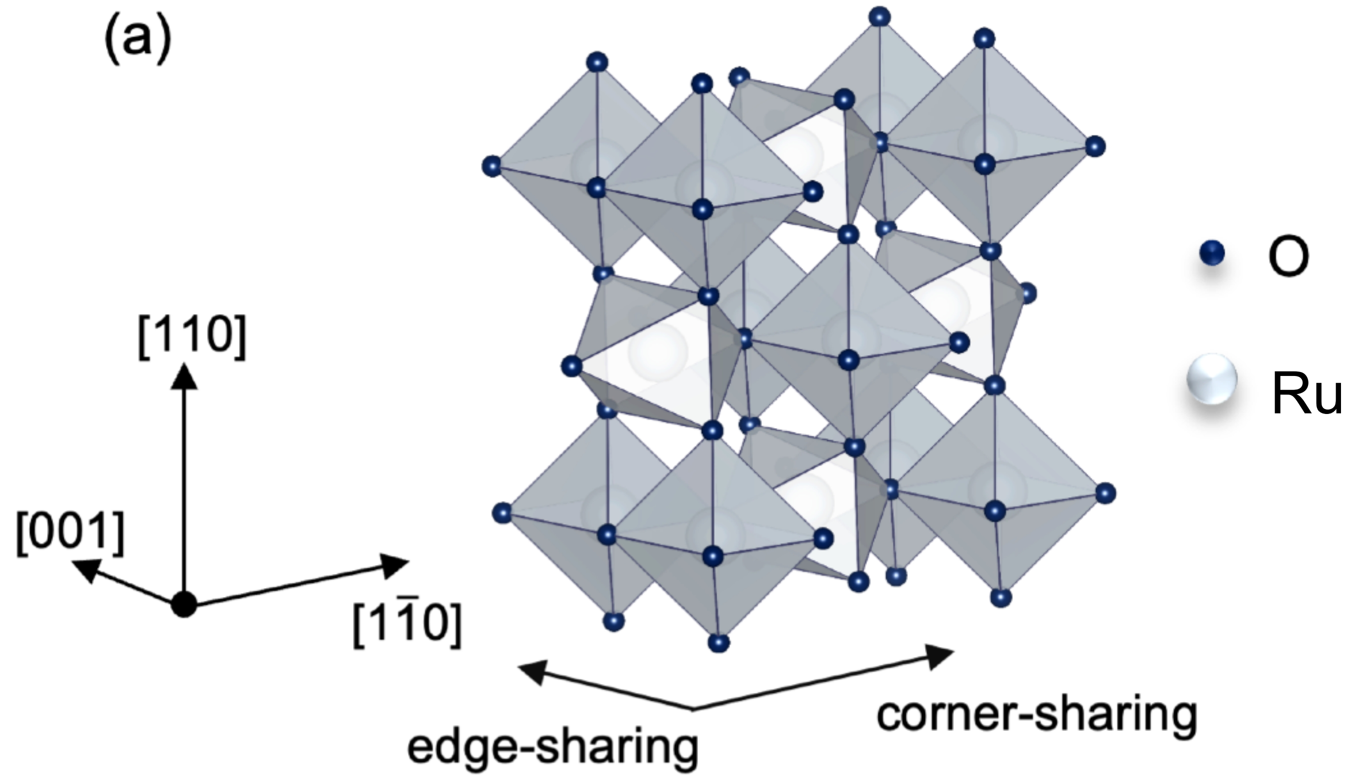
- 20th century — Paramagnetic RuO_2
- 2017 — Antiferromagnetism (Neutron diffraction)
- 2019 — Antiferromagnetism (X-ray scattering)
- 2020 — Crystal Hall effect (Theory)
- 2022
- 2023
- 2024

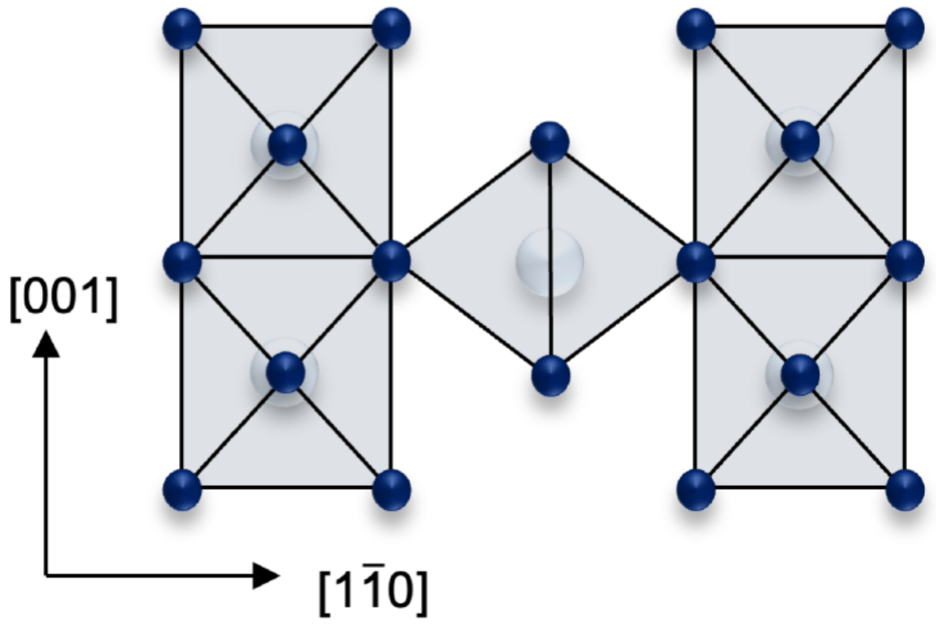
Film

- 2019 — Antiferromagnetism (X-ray scattering)
- 2020
- 2022
  - Anomalous Hall effect
  - Spin-spitter torque
- 2023 — Spin Seebeck effect
- 2024
  - Exchange bias
  - Spin density wave
  - Spin splitting band (ARPES)
  - Magnetic circular dichroism
  - Ultrafast magneto-optical Kerr effect
  - Magnetic octupole (SHG)

Altermagnetism

(a)





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Altermagnetism.

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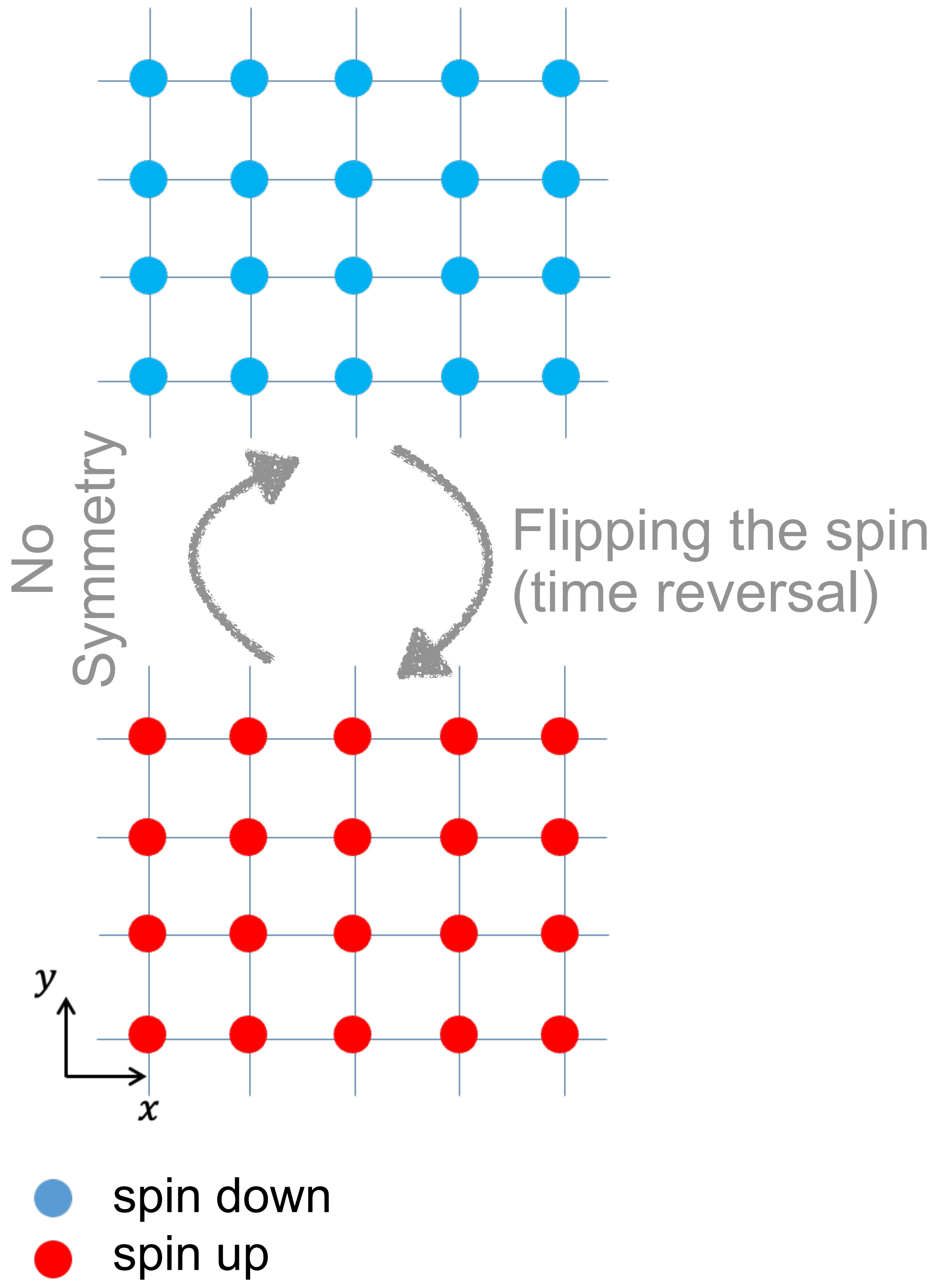
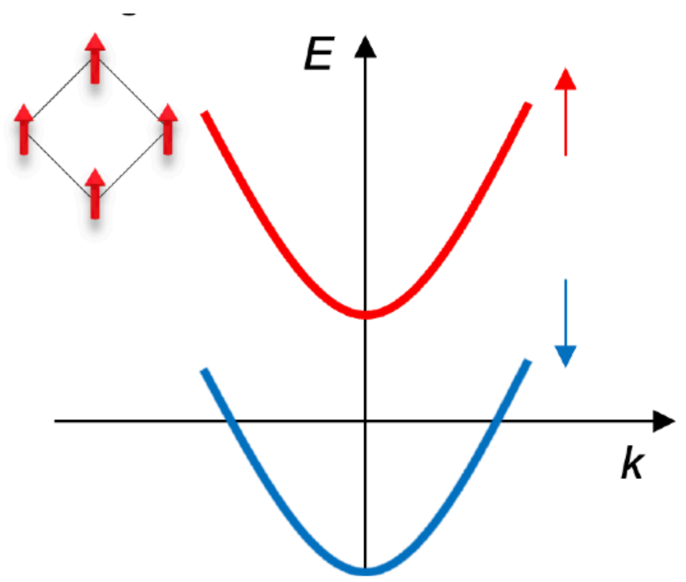
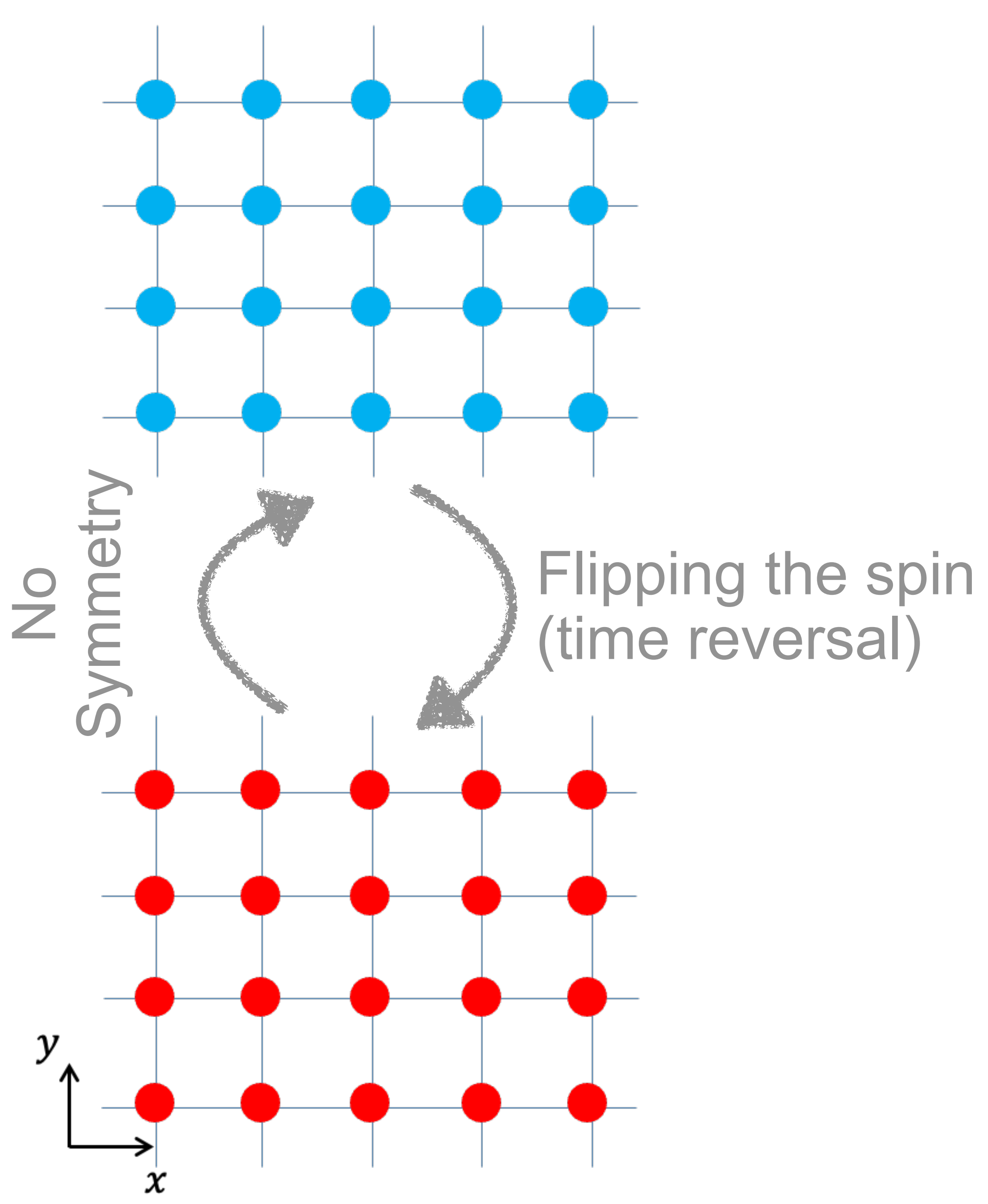
Ferromagnetism (FM)



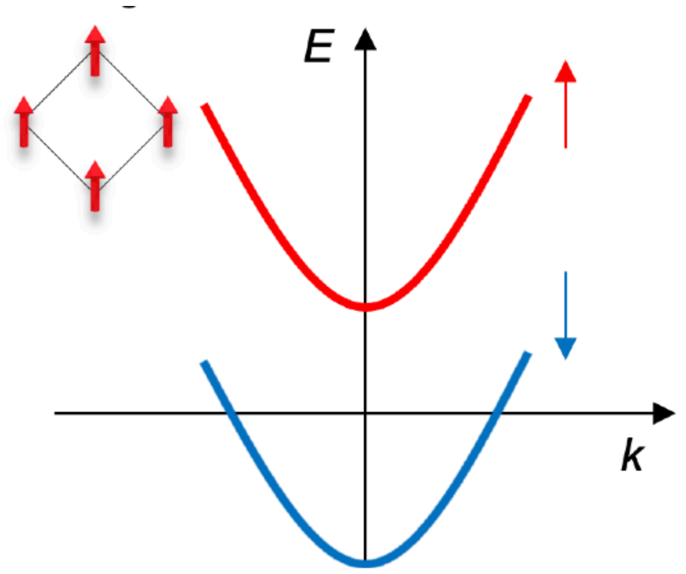


Altermagnetism.

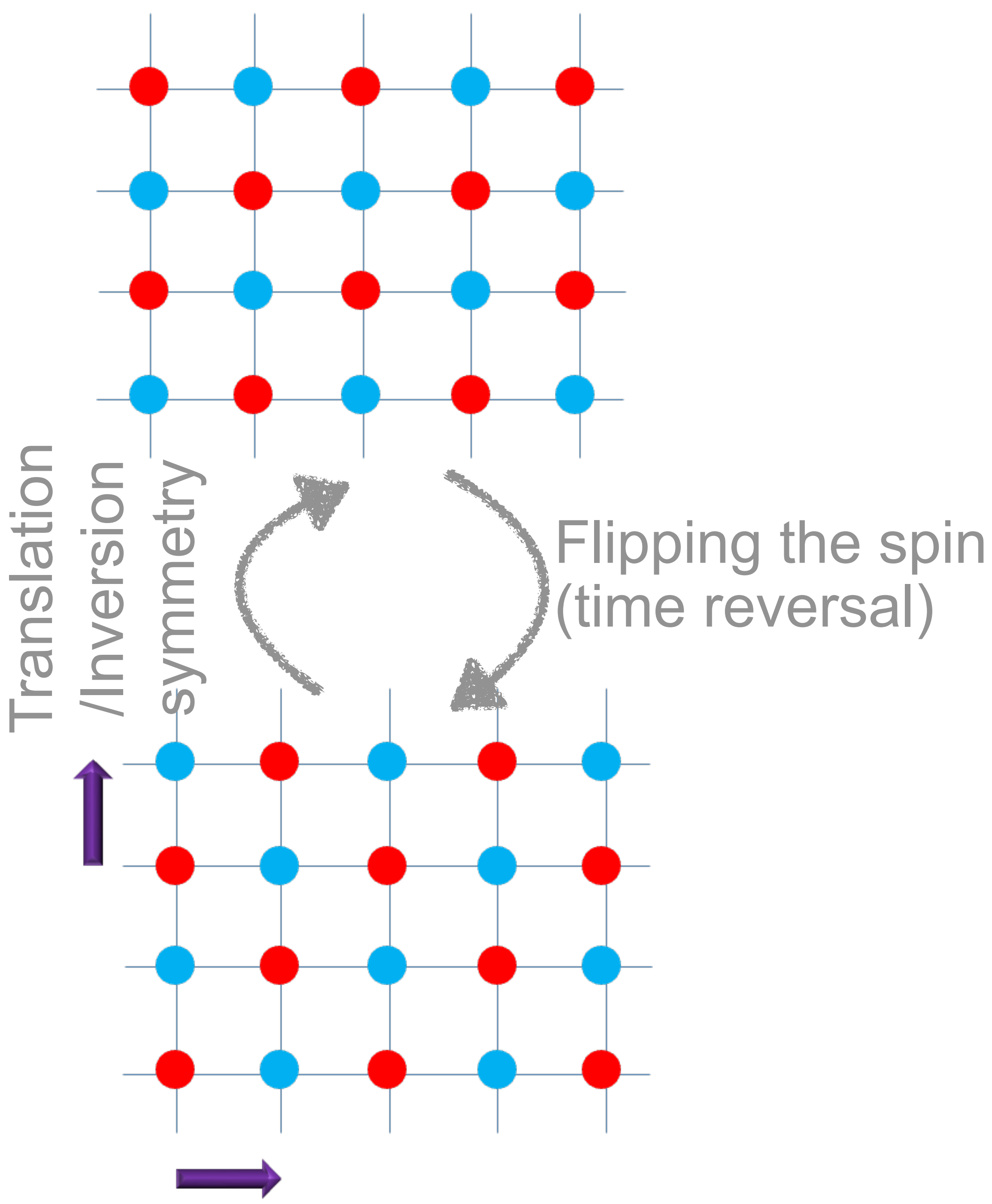
Ferromagnetism (FM)

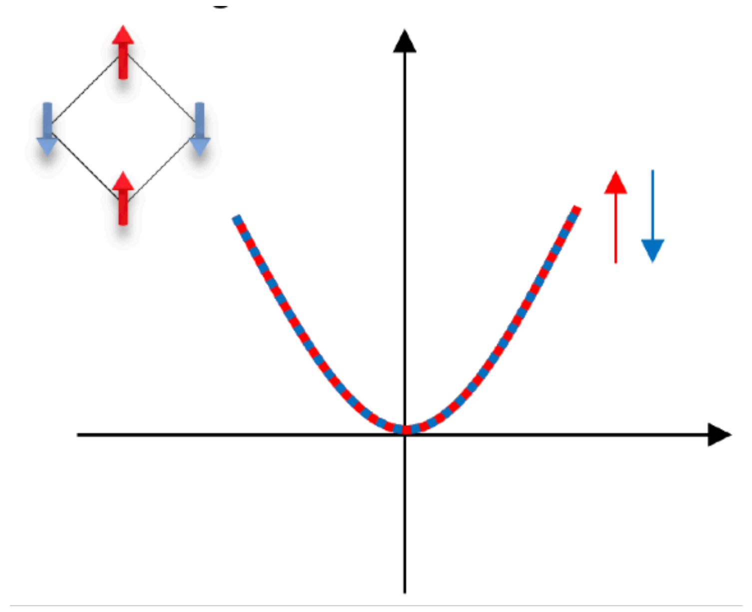


spin down
spin up



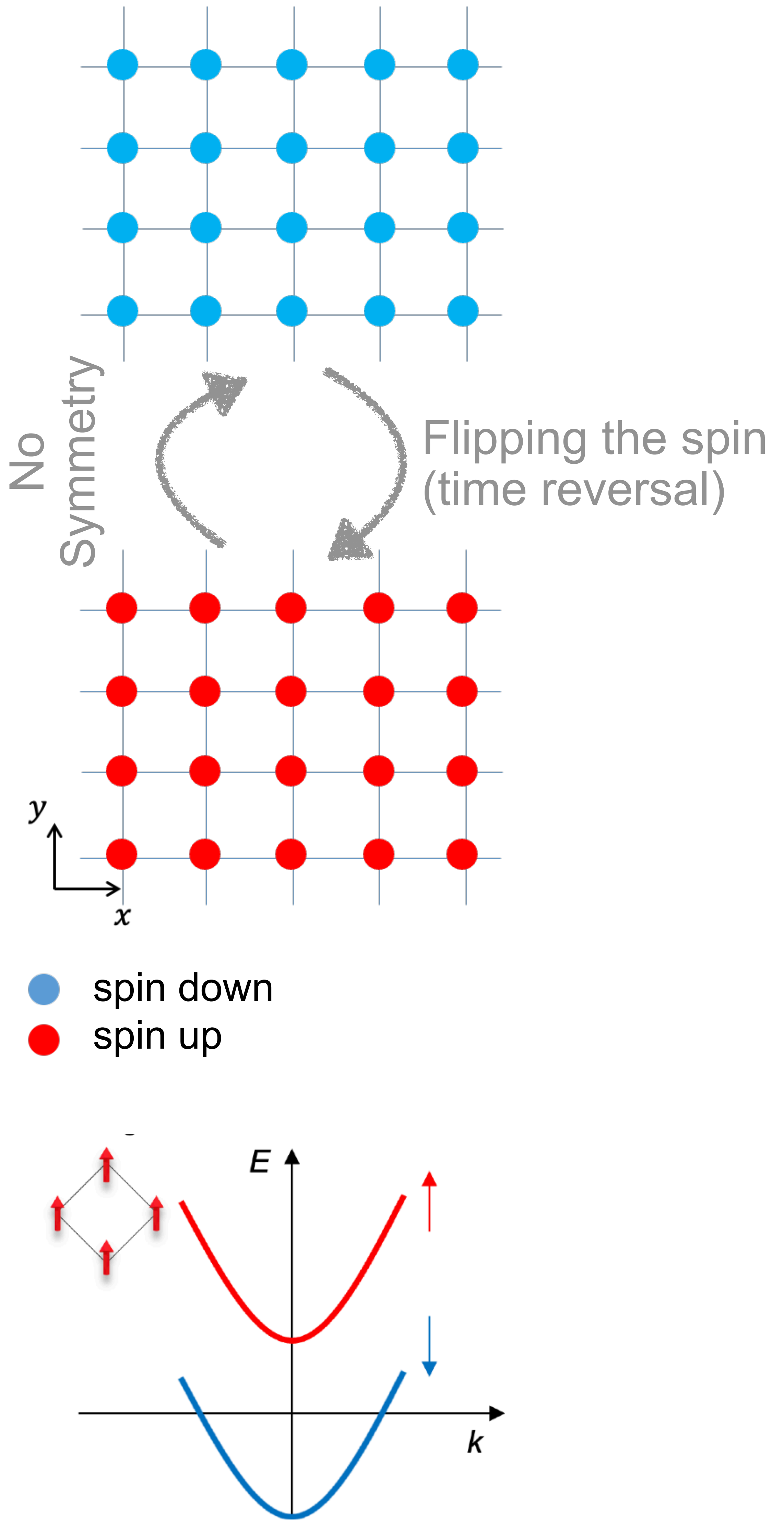
Antiferromagnetism (AF)



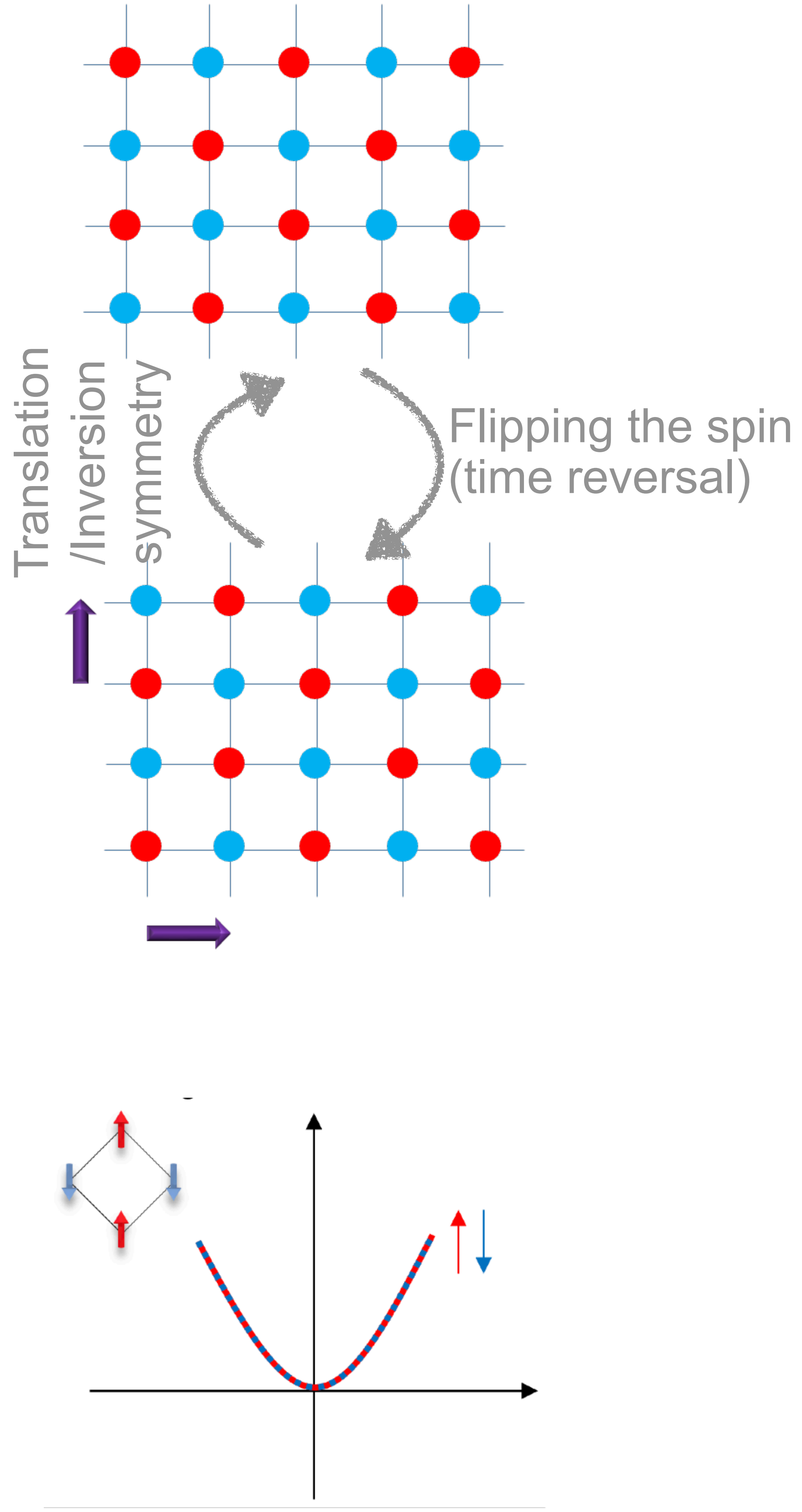


Altermagnetism.

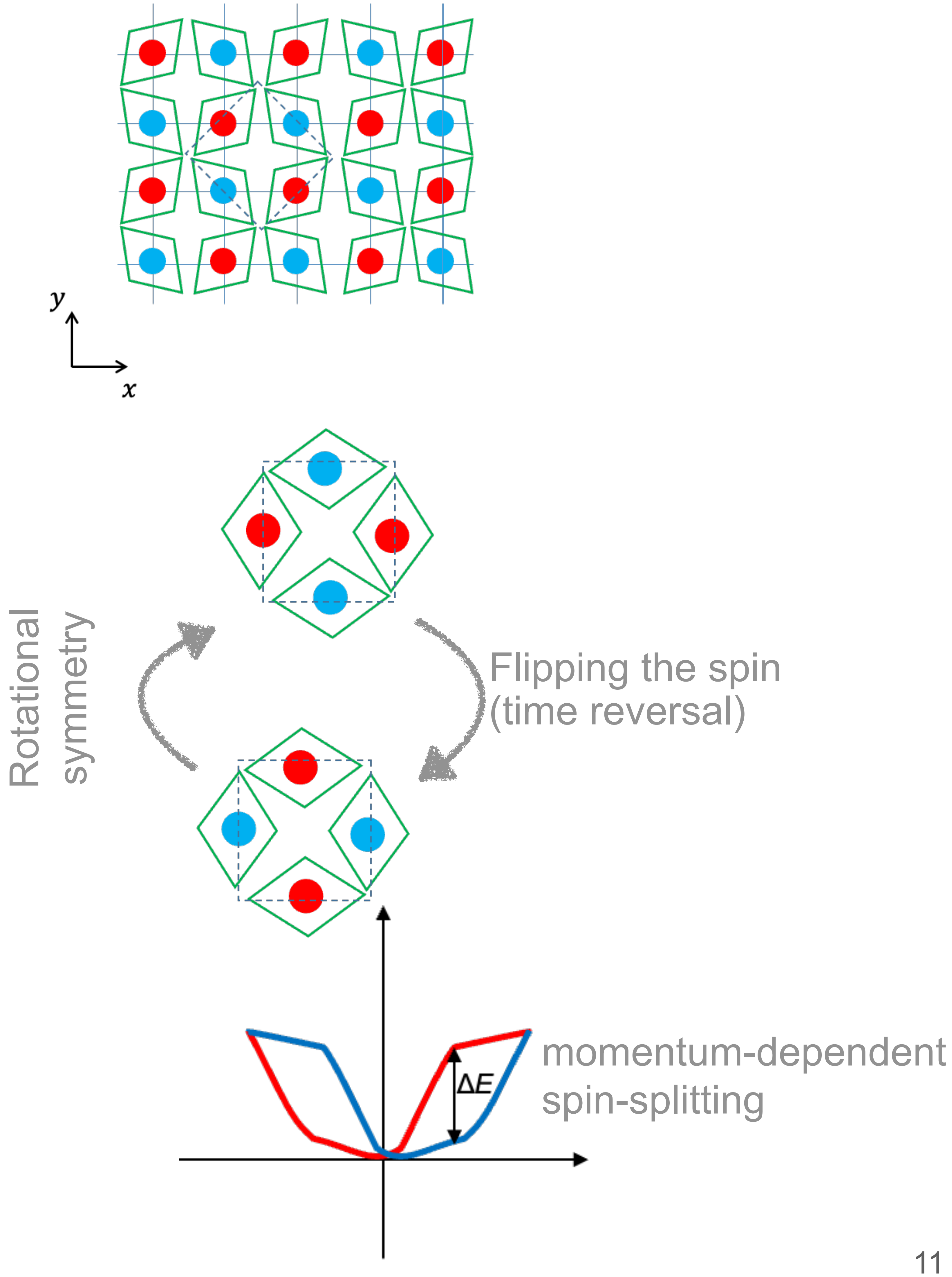
Ferromagnetism (FM)



Antiferromagnetism (AF)



Altermagnetism (AF + local environments)



A Controversial Topic in Condensed Matter Physics

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Nonmagnetic RuO_2
Magnetic RuO_2

Bulk

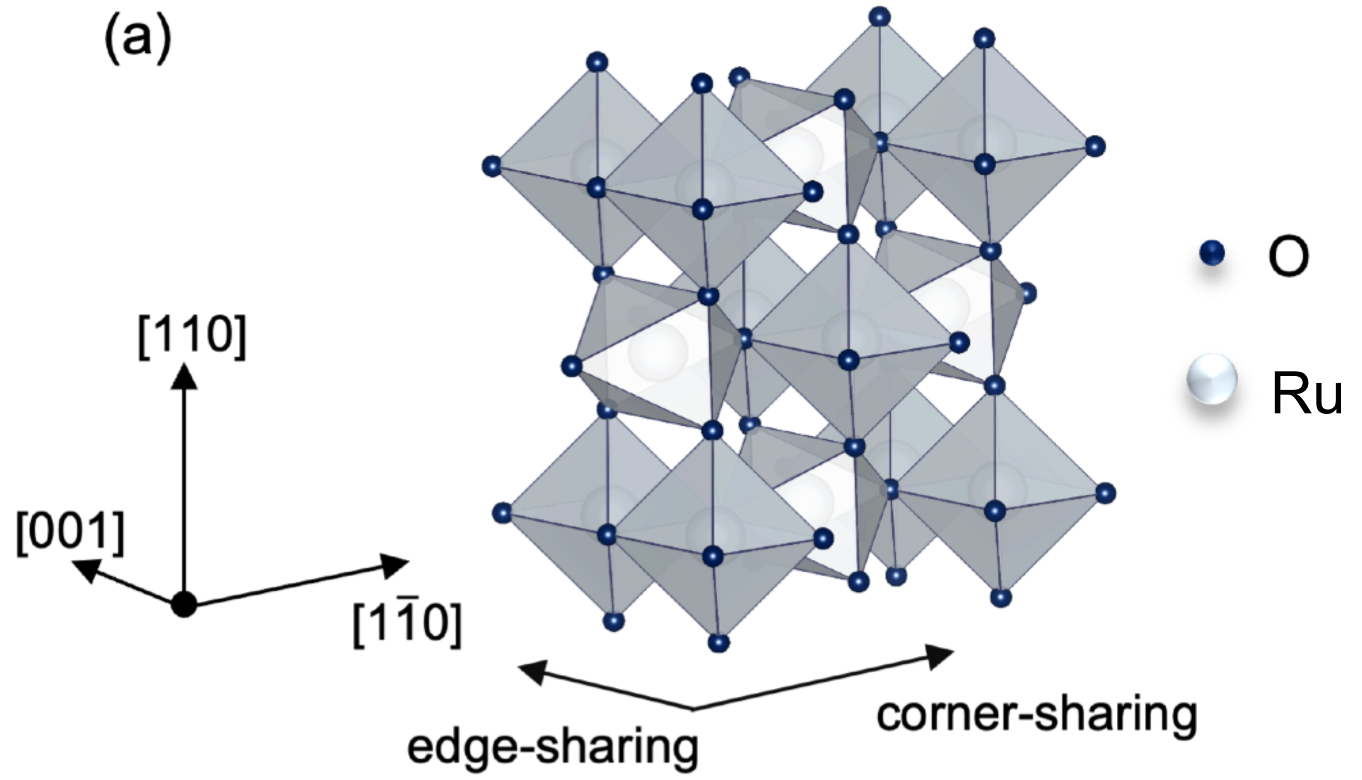
- 20th century: Paramagnetic RuO_2
- 2017: Antiferromagnetism (Neutron diffraction)
- 2019: Antiferromagnetism (X-ray scattering)
- 2020: Crystal Hall effect (Theory)
- 2023/2024:
  - Absence of magnetization (μSR spectroscopy and neutron diffraction)
  - Absence of spin splitting band (ARPES)
  - Nonmagnetic phase in optical conductivity
  - Multicarrier transports

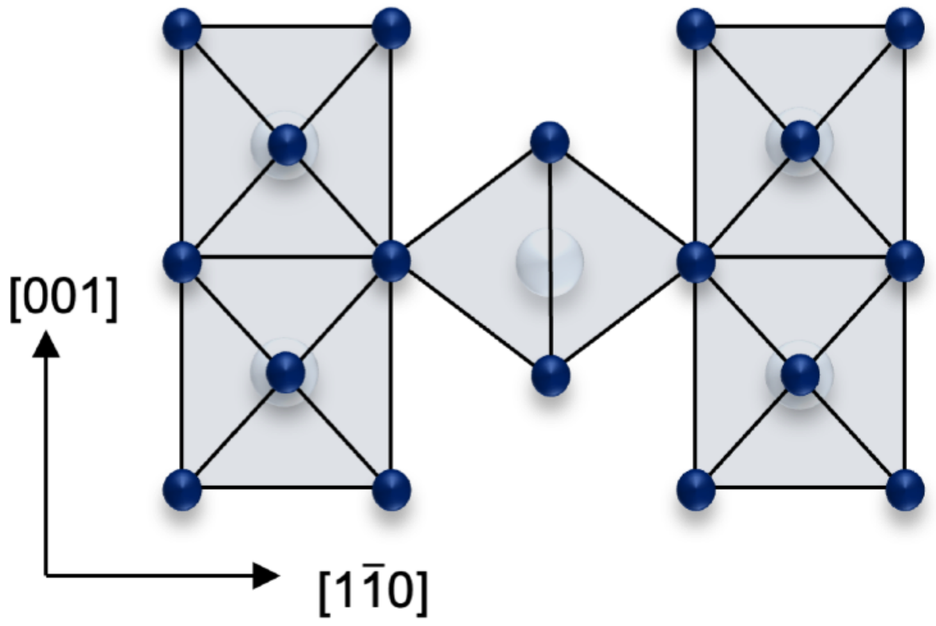
Film

- 2019: Antiferromagnetism (X-ray scattering)
- 2022:
  - Anomalous Hall effect
  - Spin-spitter torque
- 2023: Spin Seebeck effect
- 2024:
  - Exchange bias
  - Spin density wave
  - Spin splitting band (ARPES)
  - Magnetic circular dichroism
  - Ultrafast magneto-optical Kerr effect
  - Magnetic octupole (SHG)
  - Absence of magnetization (μSR spectroscopy)
  - Absence of spin splitting band (ARPES)

Altermagnetism (2020–2024)

(a)





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Looking at RuO_2 through Materials Science Lenses..

Ru can take multiple valence states..

Charge	C.N.	Spin	I.R./Å
+3	6		0.68
+4	6		0.620
+5	6		0.565
+7	4		0.38
+8	4		0.36

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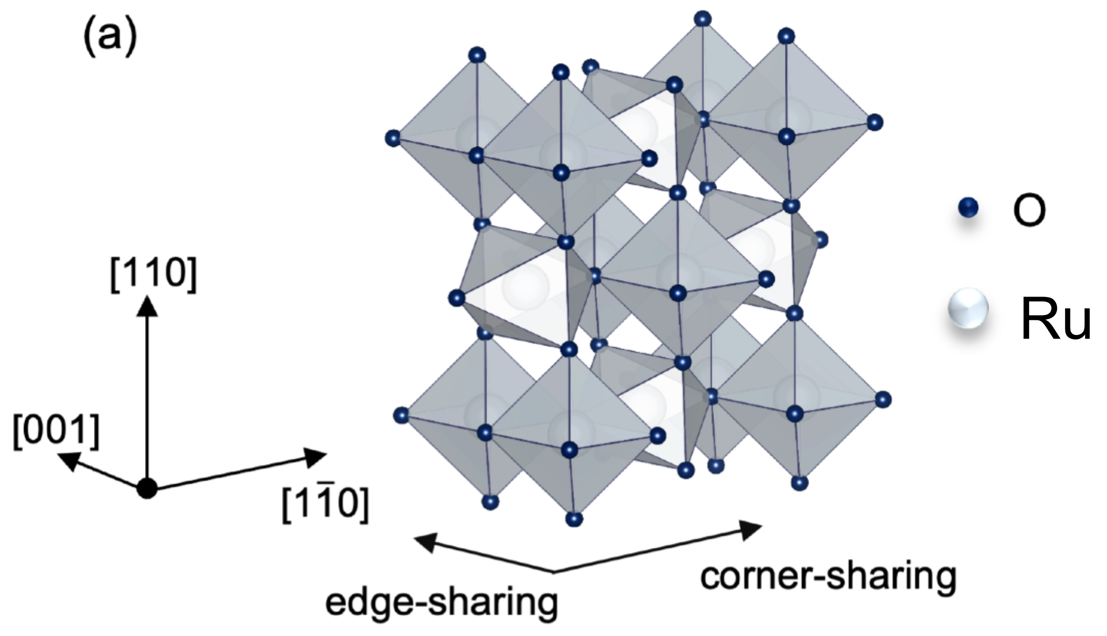
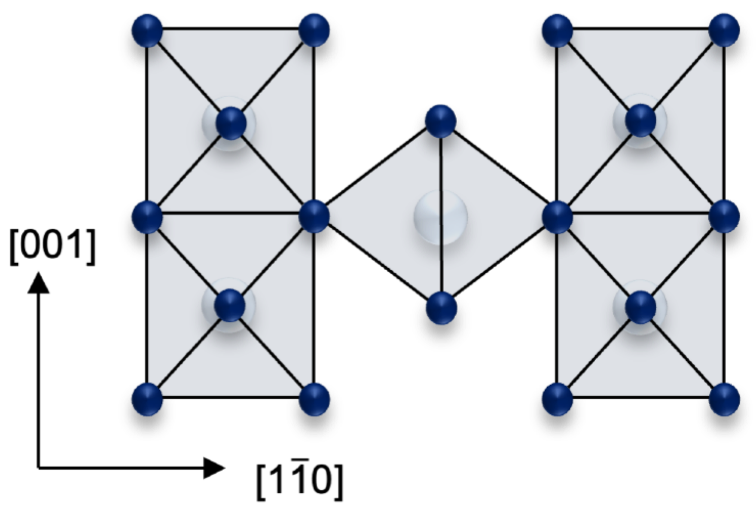
Anisotropic crystal structure/bonding

(a)





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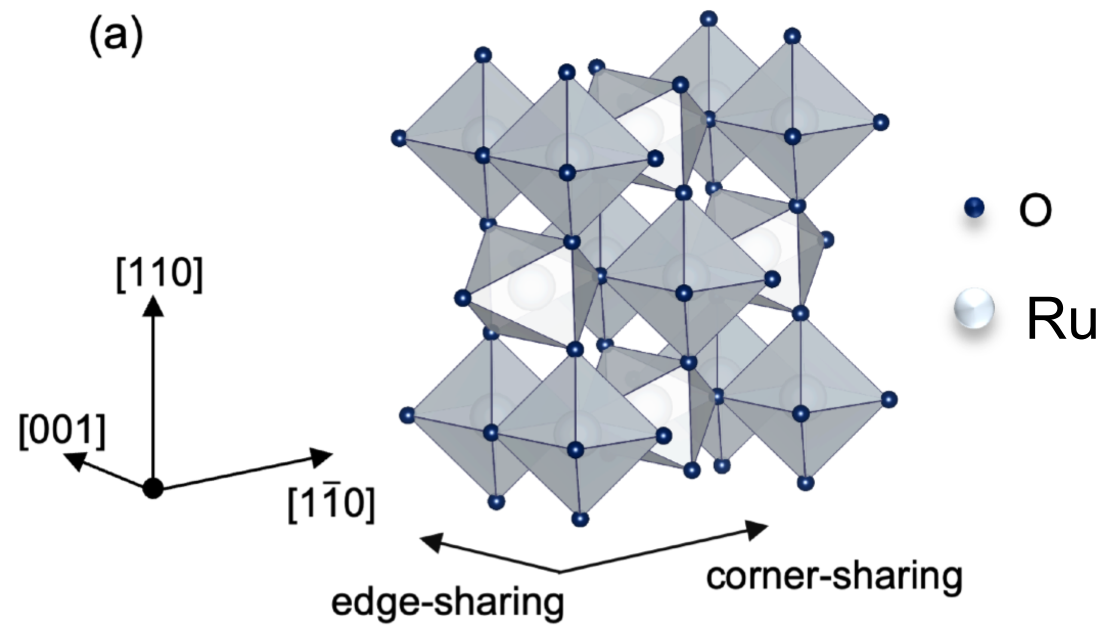
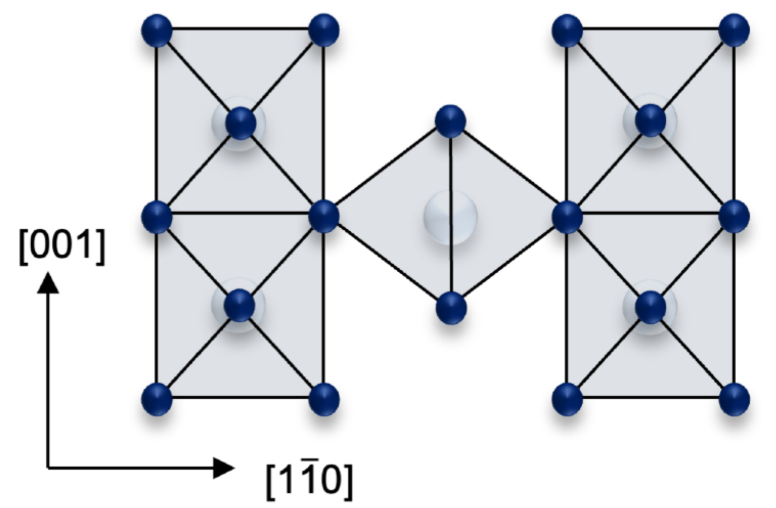
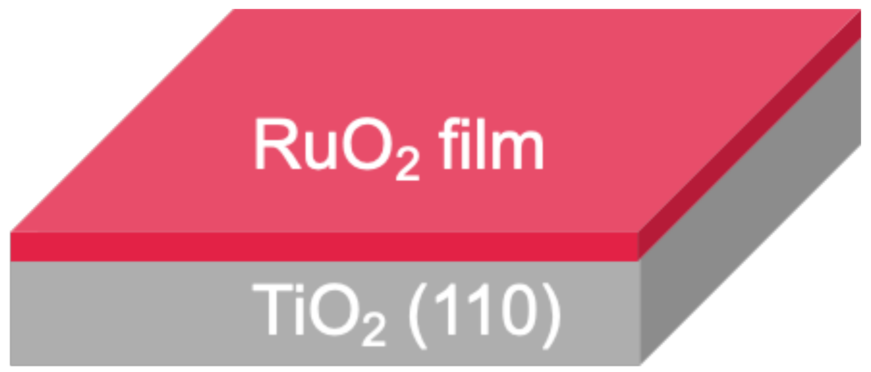
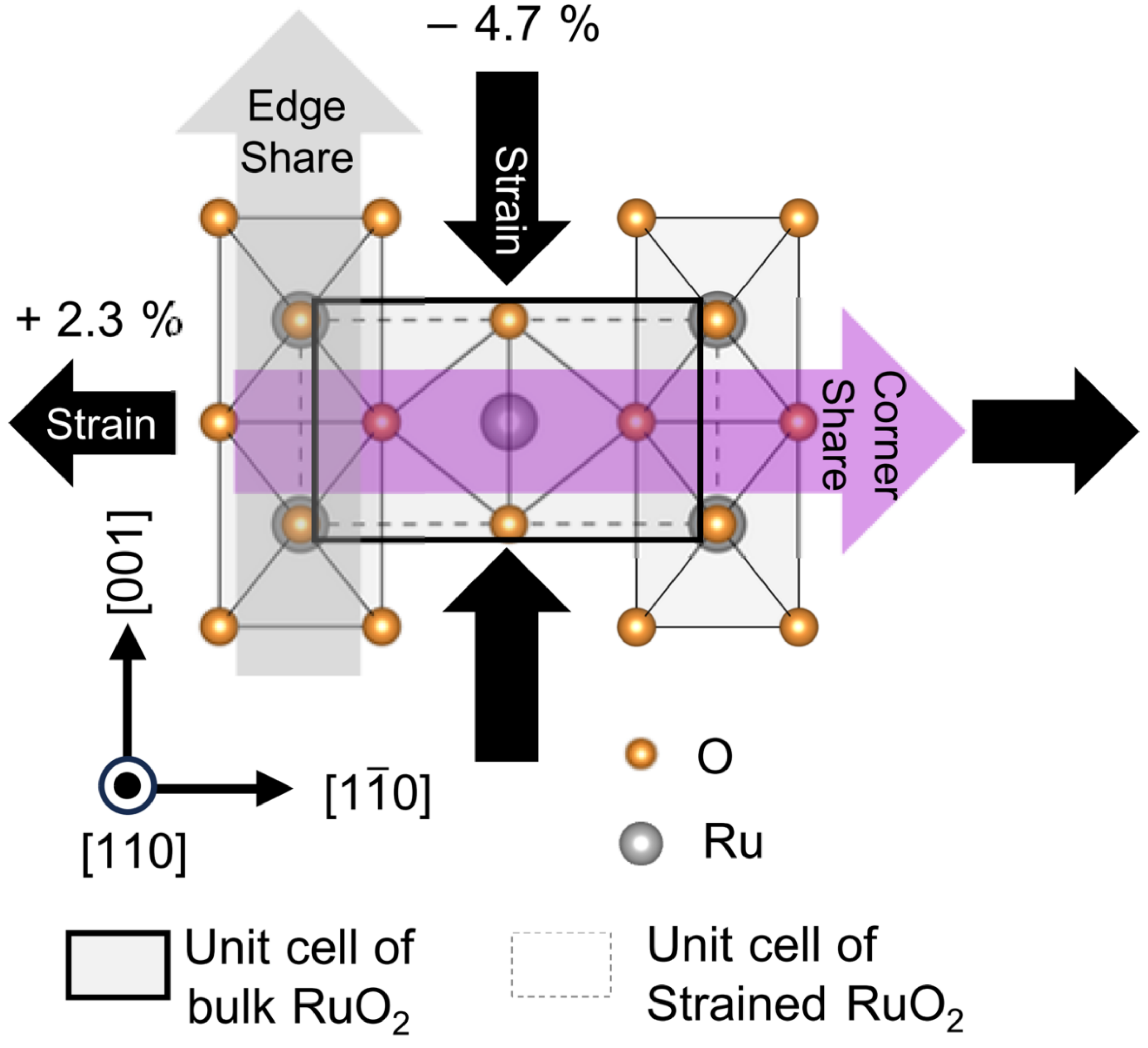




Thin films: Epitaxial strain (anisotropy)..





Synthesis of atomically-precise thin films of RuO_2

- Low vapor pressure (harder to evaporate/sublimate)
- Low oxidation potential (harder to oxidize)

R. Choudhary, B. Jalan, Atomically precise synthesis of oxides with hybrid molecular beam epitaxy, Device (cell press) **3** 100711 (2025)

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Synthesis of atomically-precise thin films of RuO_2

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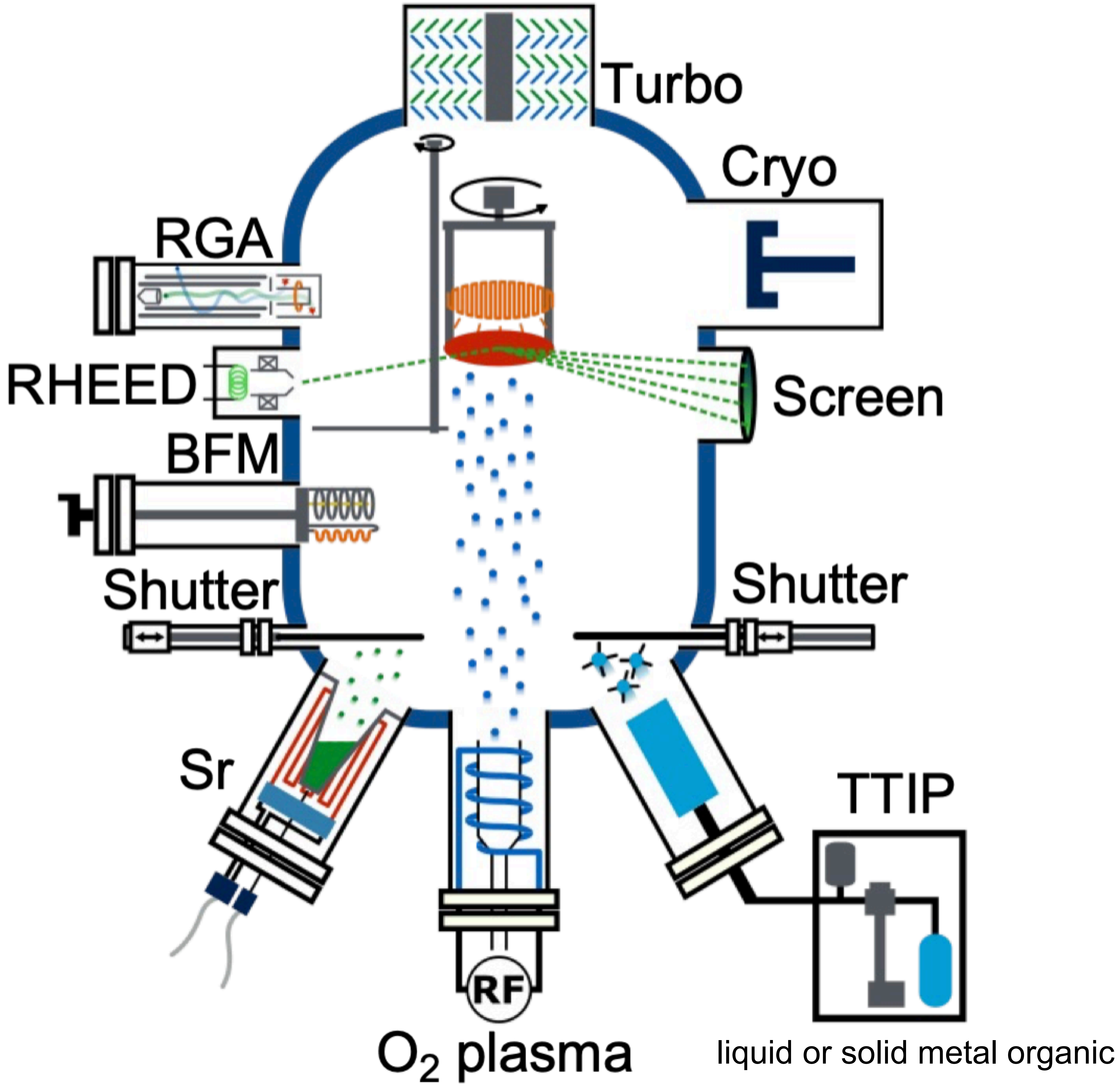
Hybrid MBE addresses these challenges!



W. Nunn, A. K. Manjeshwar, …, B. Jalan, *PNAS*, 118(32) (2021)

Hybrid MBE addresses these challenges!

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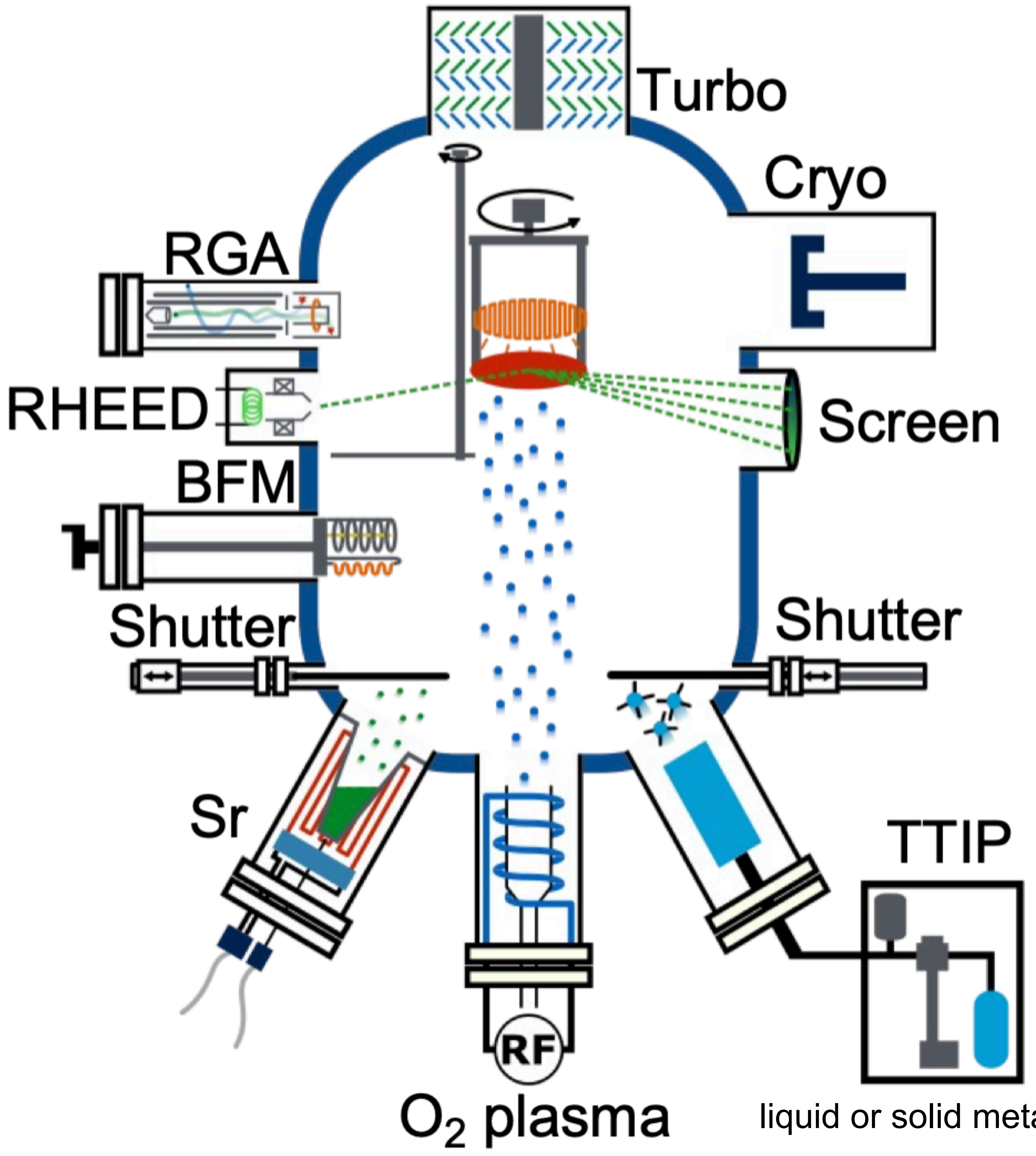
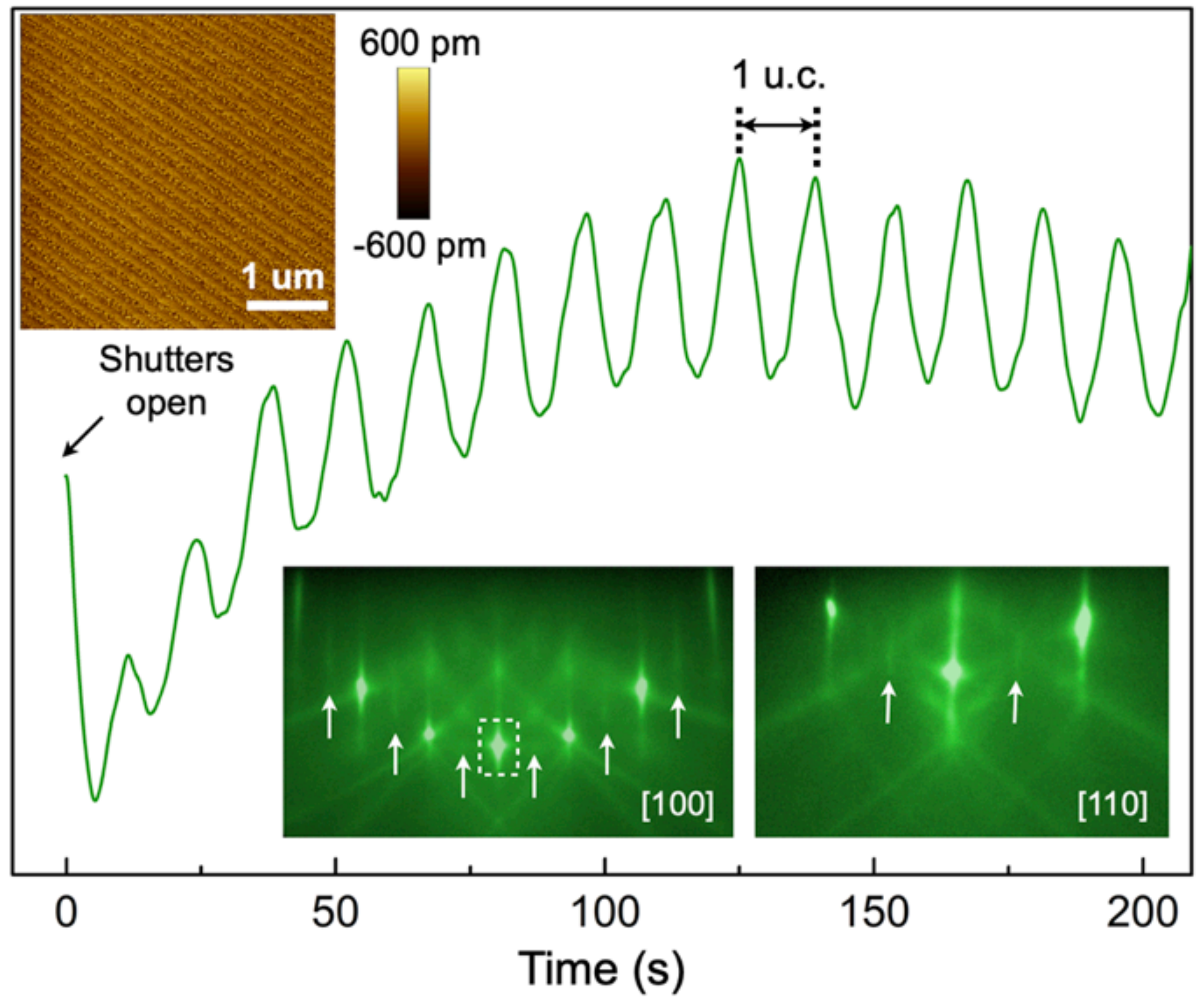
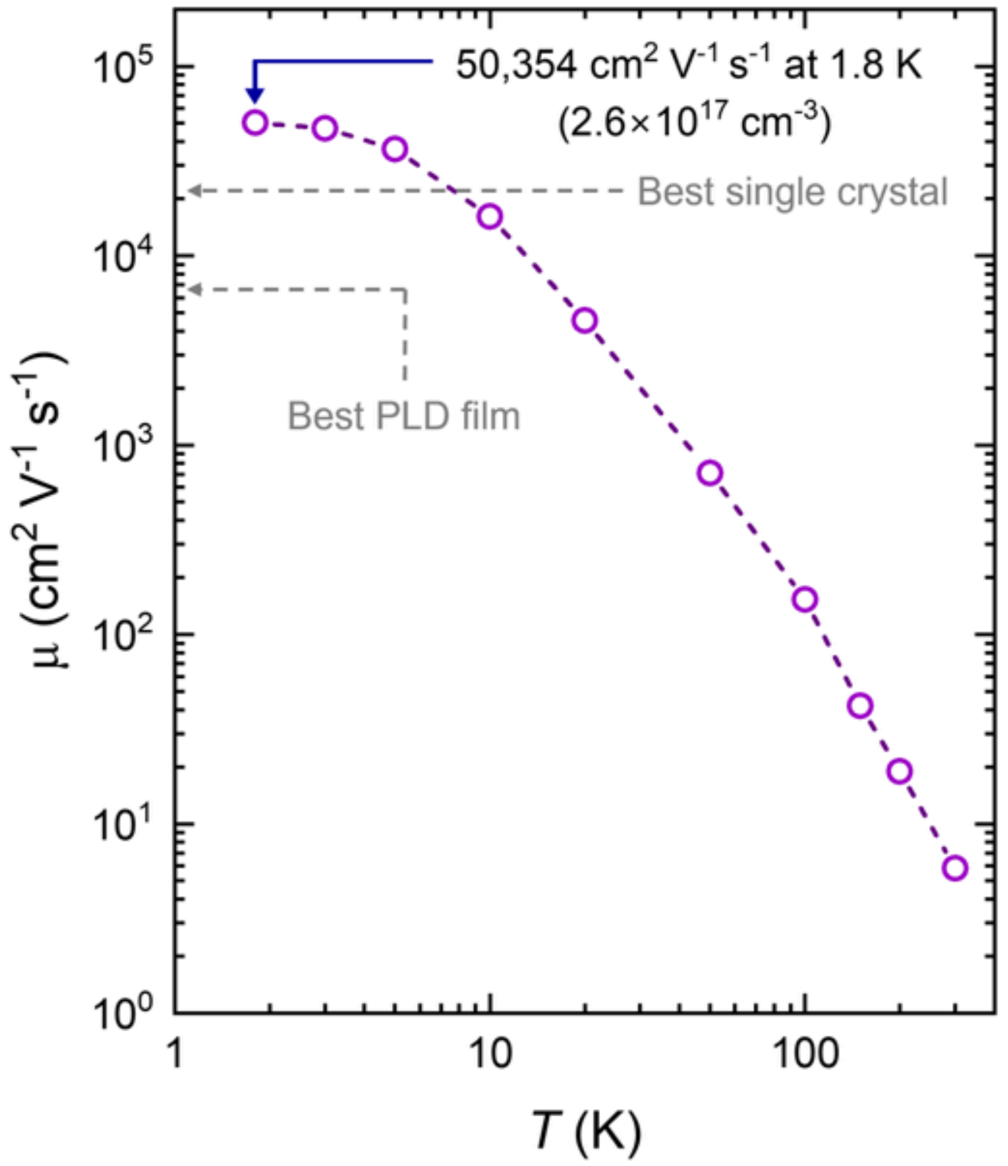


W. Nunn, A. K. Manjeshwar, …, B. Jalan, *PNAS*, 118(32) (2021)

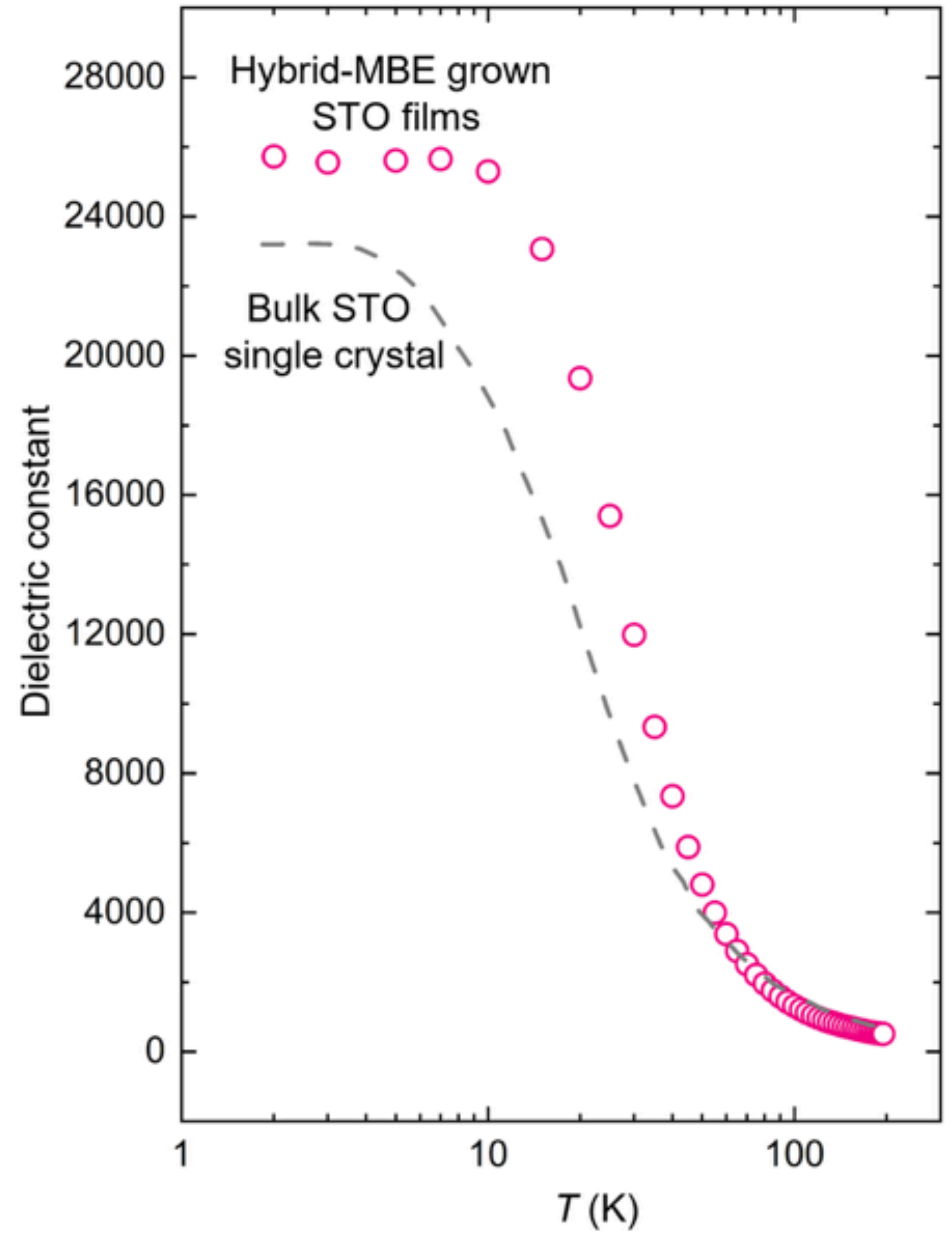
Atomic layer-by-layer growth



Higher mobility than Bulk single crystals



Record-high dielectric constant to-date



Record-high mobility and dielectric constant in $SrTiO_3$ to-date!

J. Yue, Y. Ayino, T. K. Truttmann, M. N. Gastiasoro, E. Persky, A. Khanukov, D. Lee, L. R. Thoutam, B. Kalisky, R. M. Fernandes, V. S. Pribiag, and B. Jalan, **Sci. Adv. 8, eabl5668 (2022)**

Z. Yang, D. Lee, J. Yue, J. Gabel, T.-L Lee, R. D. James, S. Chambers, and B. Jalan, **Proc. Natl. Acad. Sciences 119, e2202189119 (2022)**

Hybrid MBE addresses these challenges!

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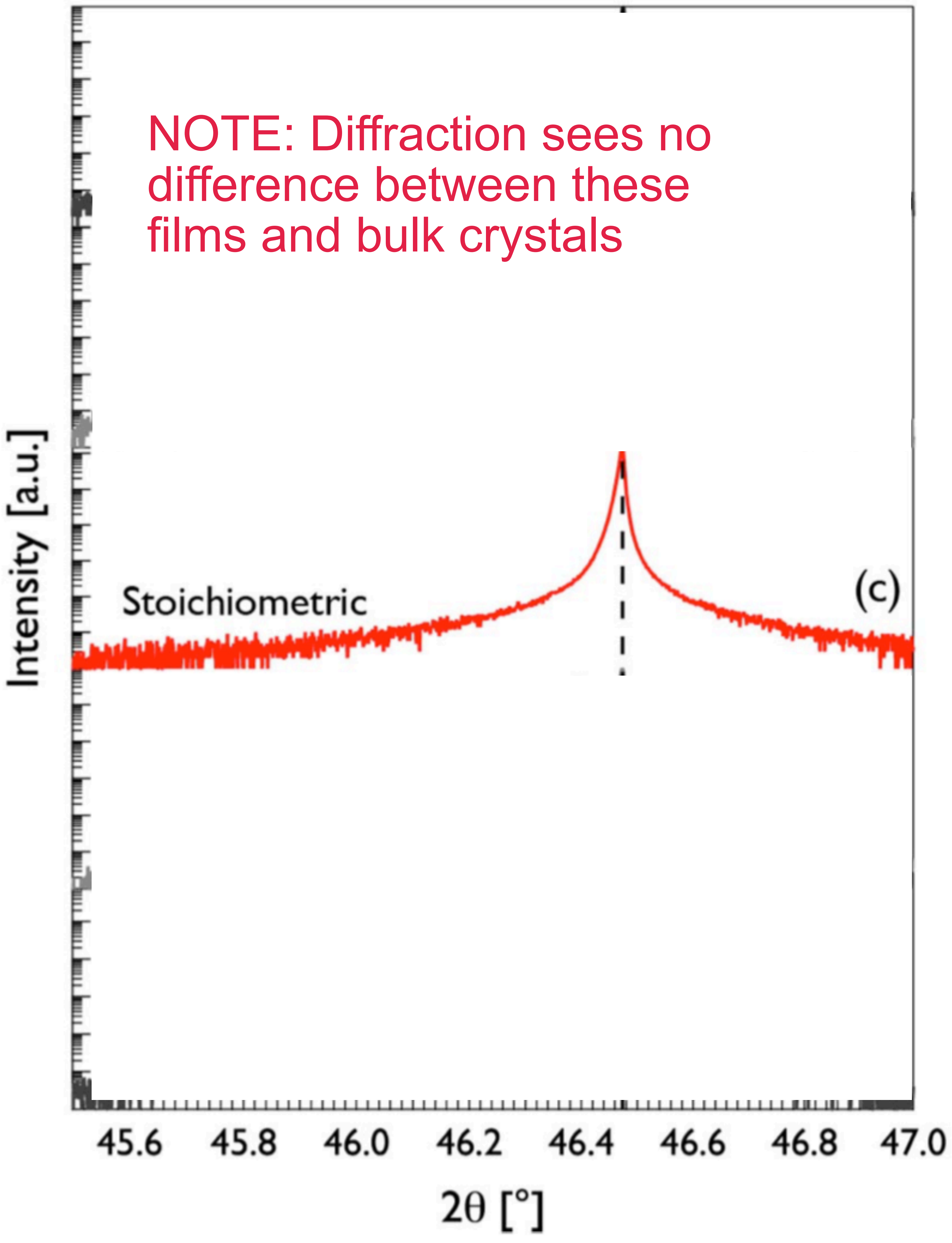
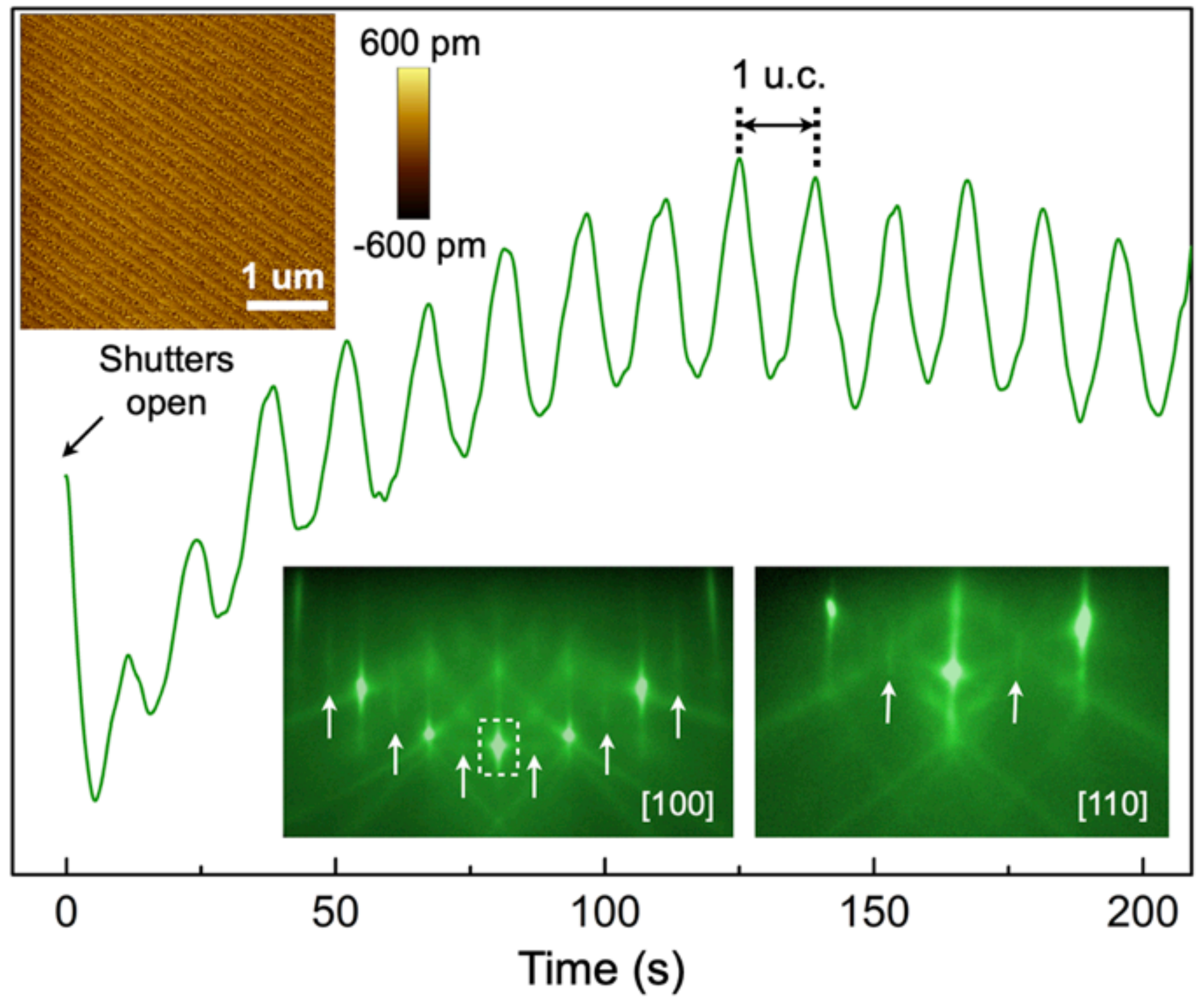
NOTE: Diffraction sees no difference between these films and bulk crystals



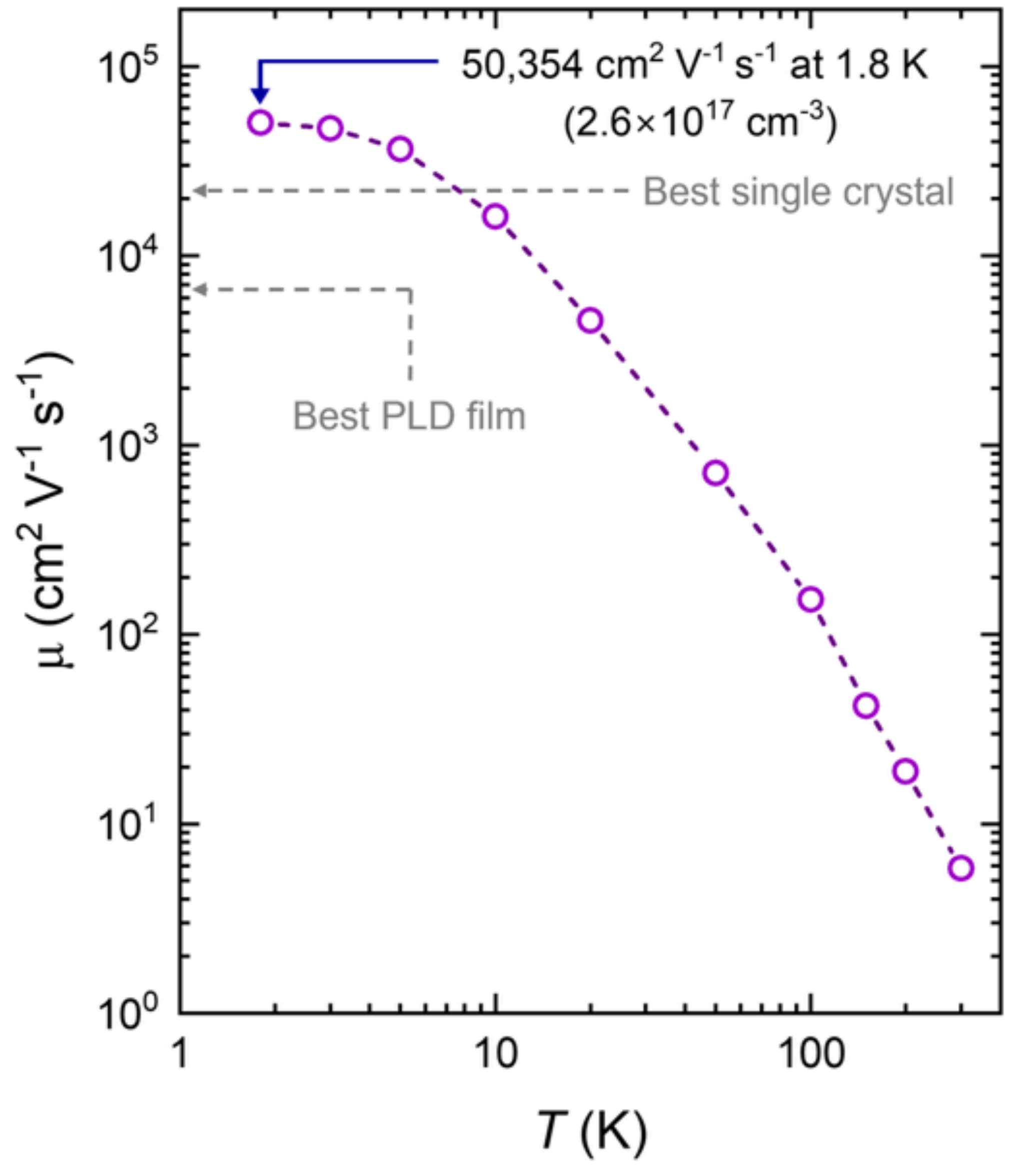
tal organic

W. Nunn, A. K. Manjeshwar, …, B. Jalan, *PNAS*, 118(32) (2021)

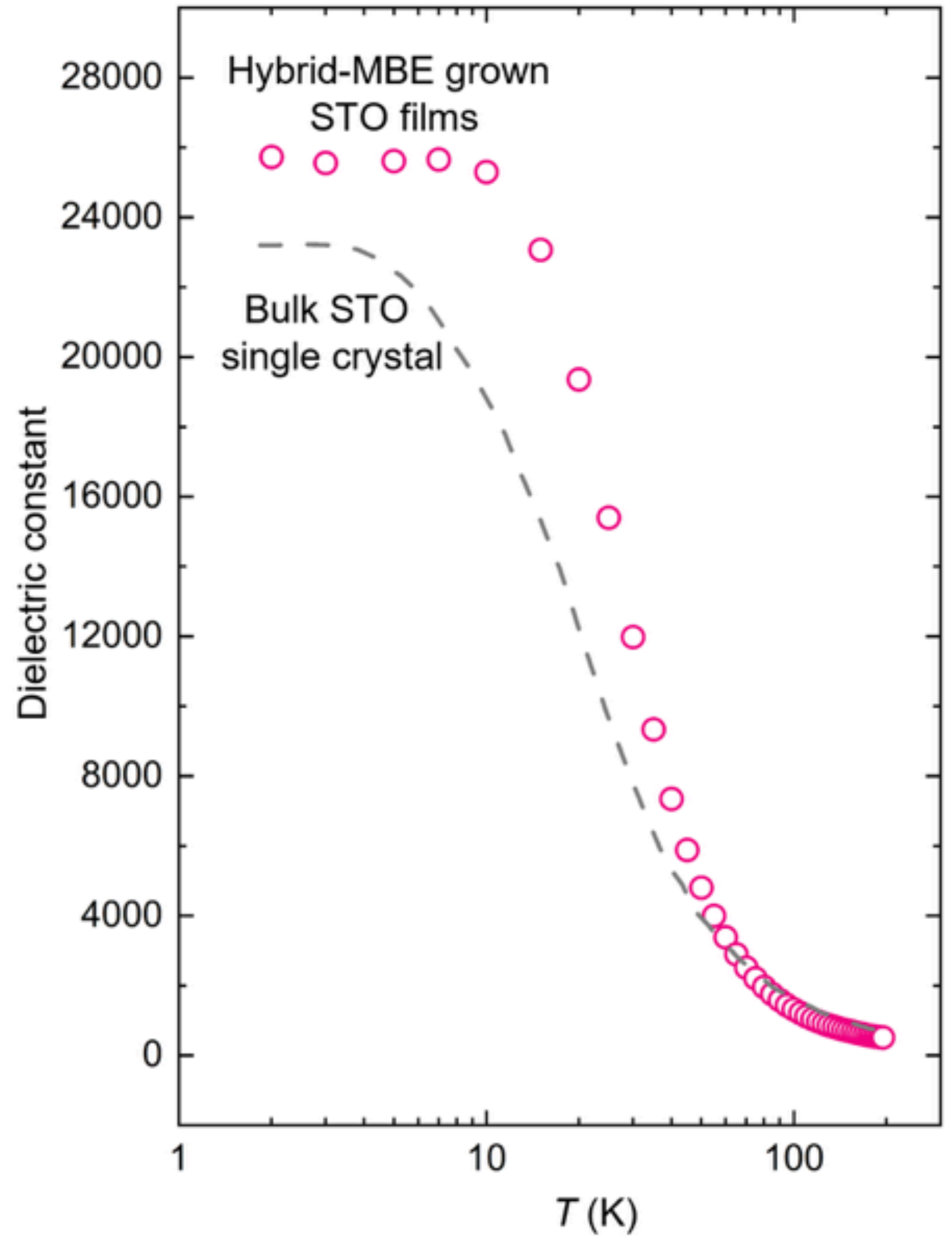
Atomic layer-by-layer growth



Higher mobility than Bulk single crystals



Record-high dielectric constant to-date



Record-high mobility and dielectric constant in $SrTiO_3$ to-date!

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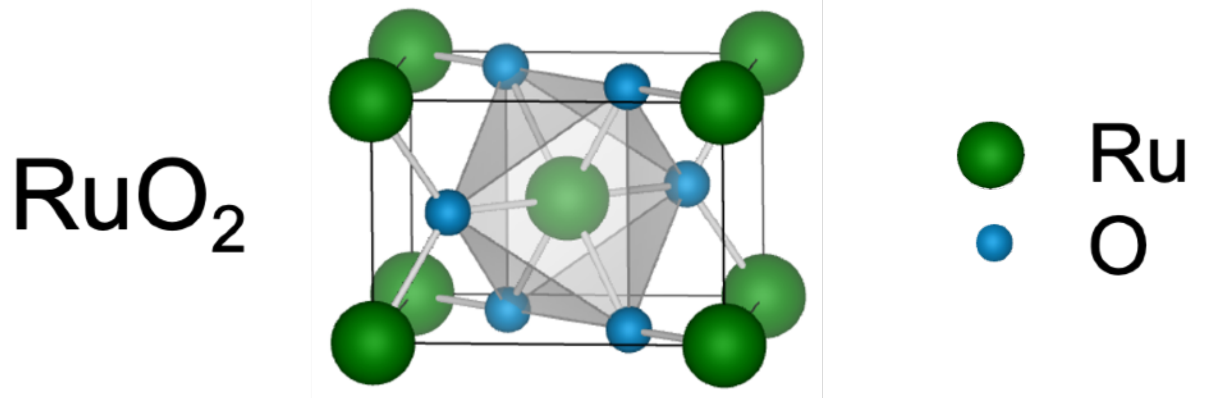
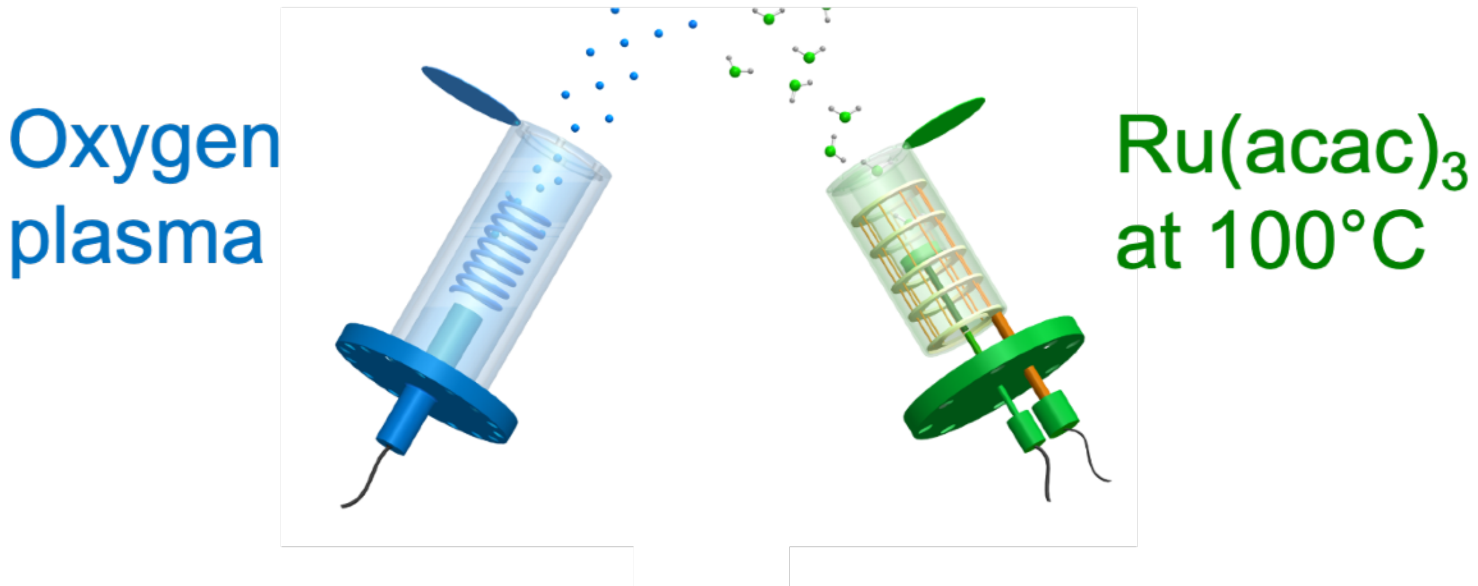
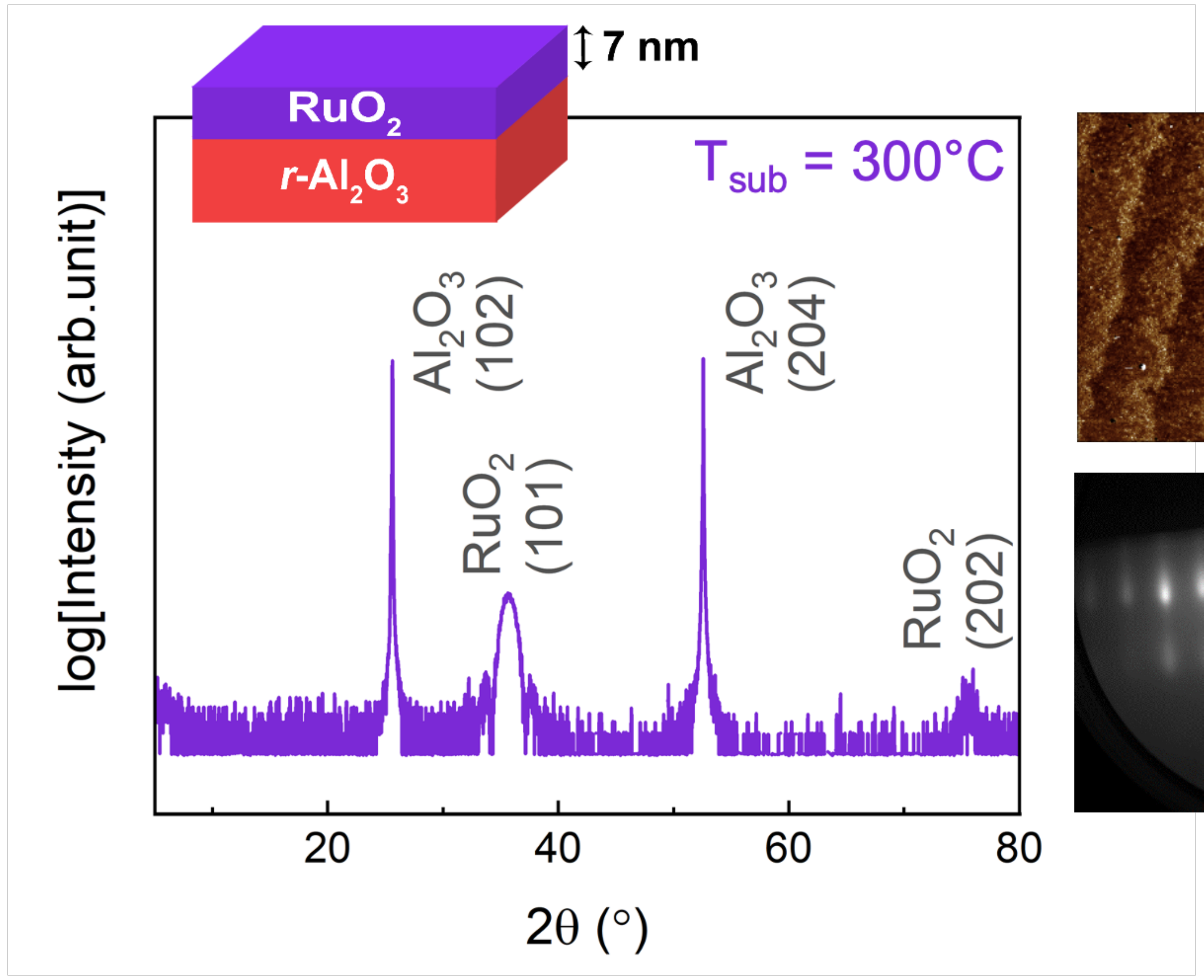
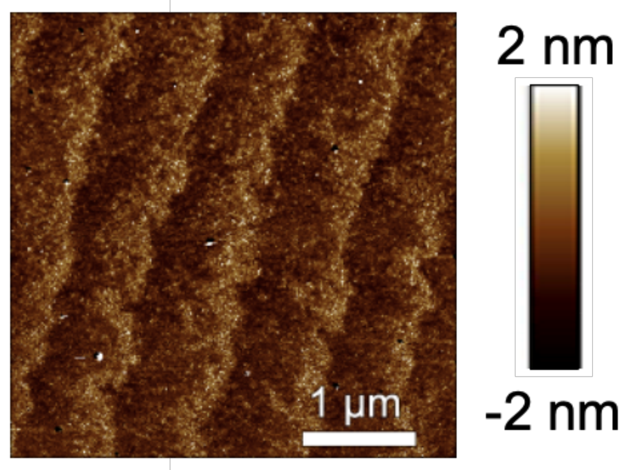
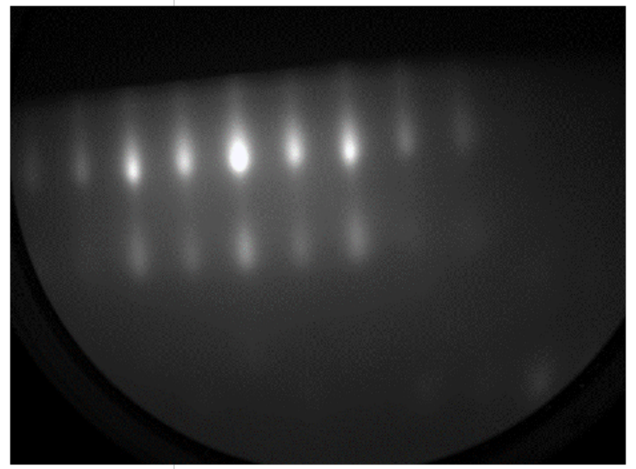
Phase-pure, Epitaxial RuO_2 films

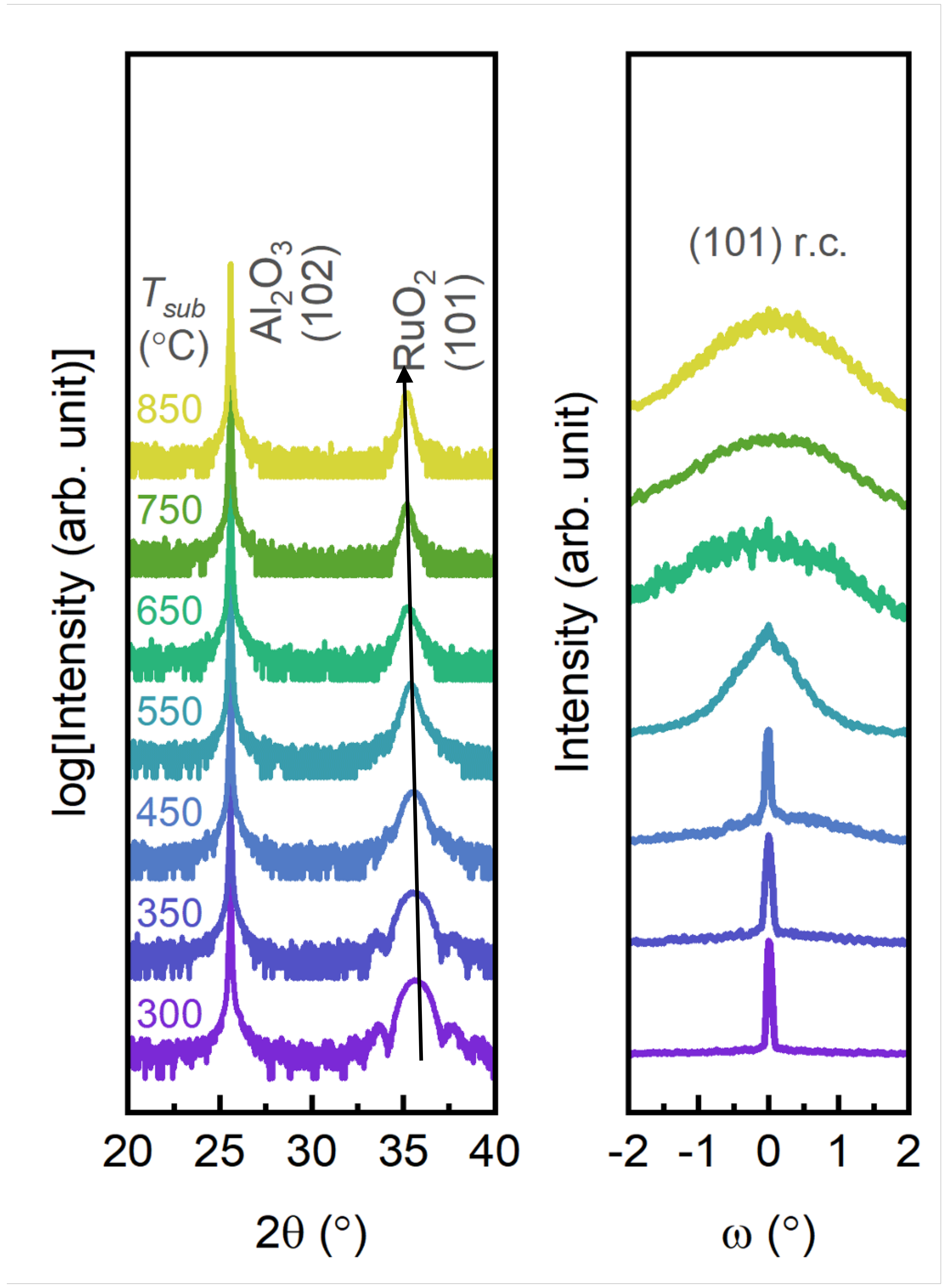


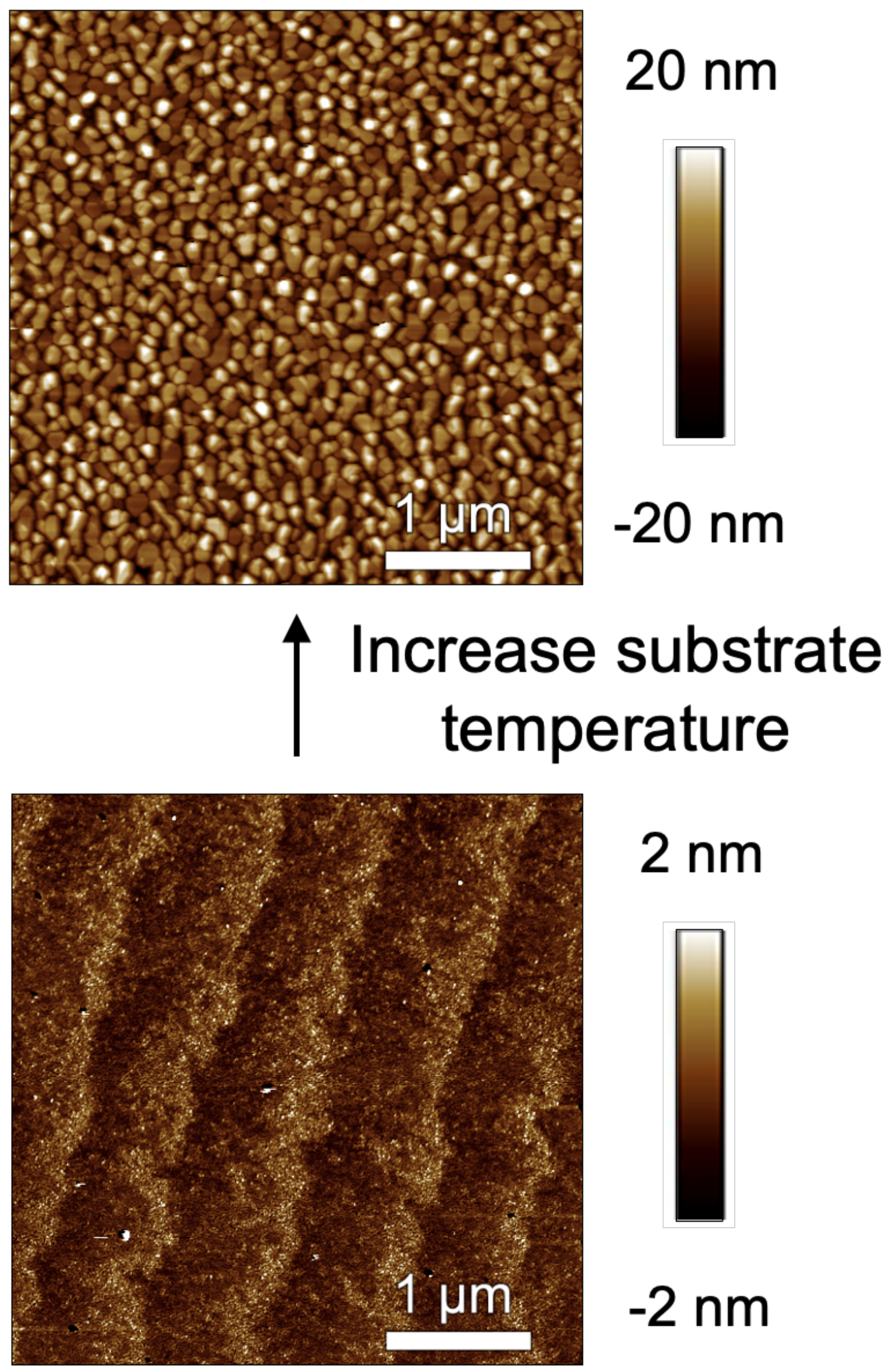












T_{sub} ↑ : crystal quality ↓ ; absence of thickness fringes —
apparent change in structural quality!

W. Nunn, S. Nair, H. Yun, A. K. Manjeshwar,…and B. Jalan, *APL Mater.* 9, (2021) [Editors' Pick][Cover]

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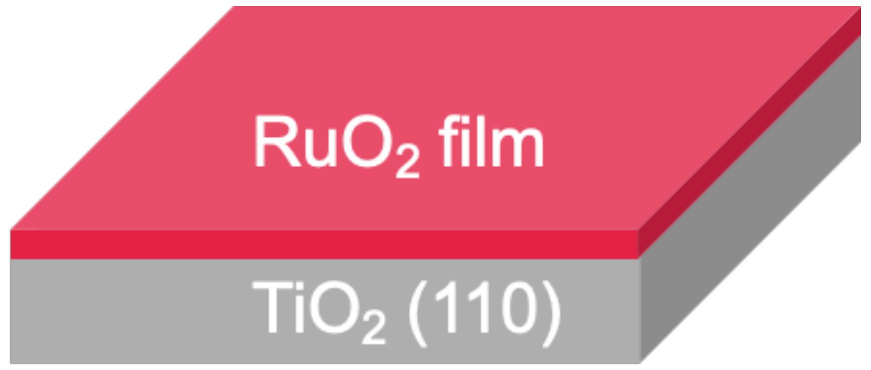
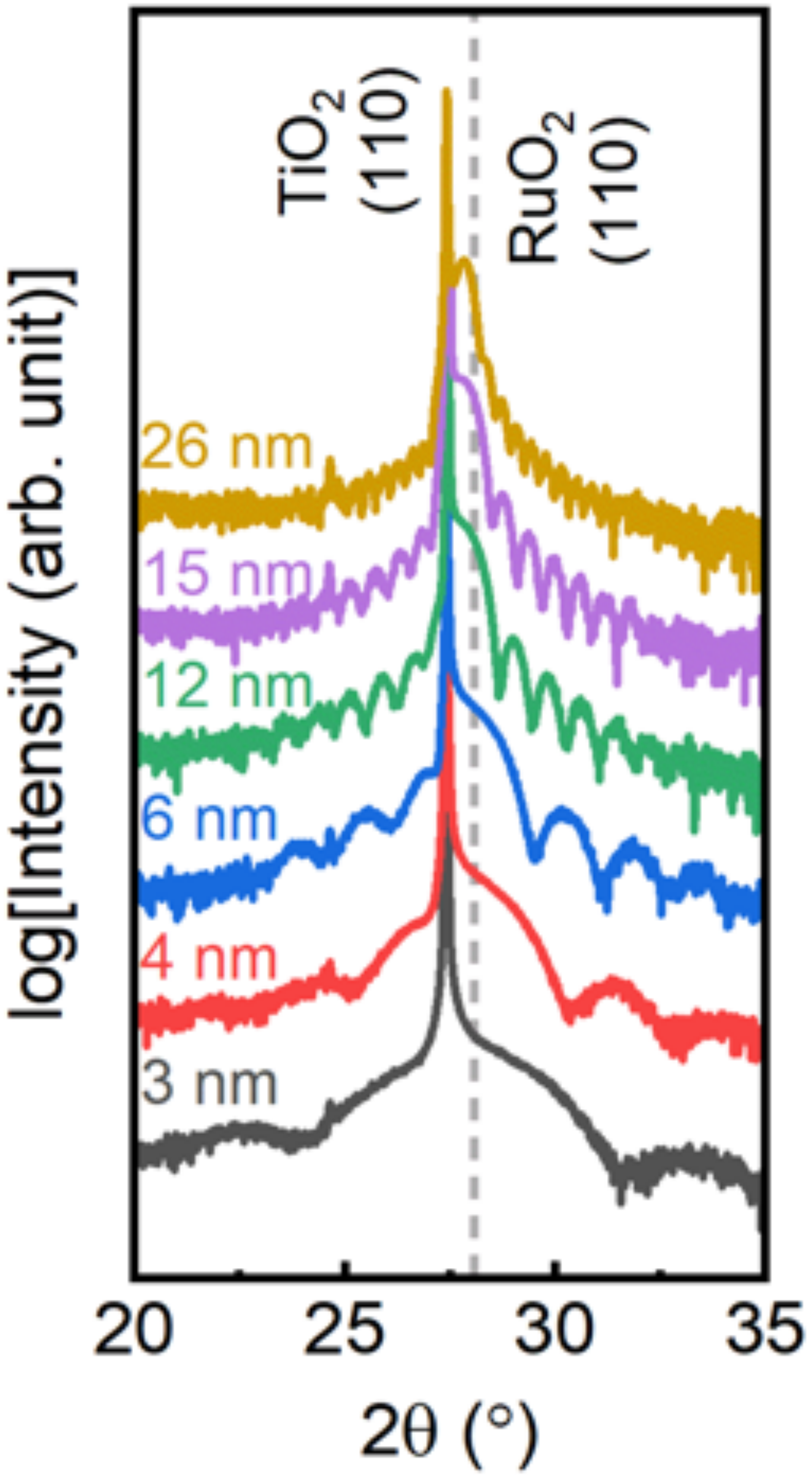


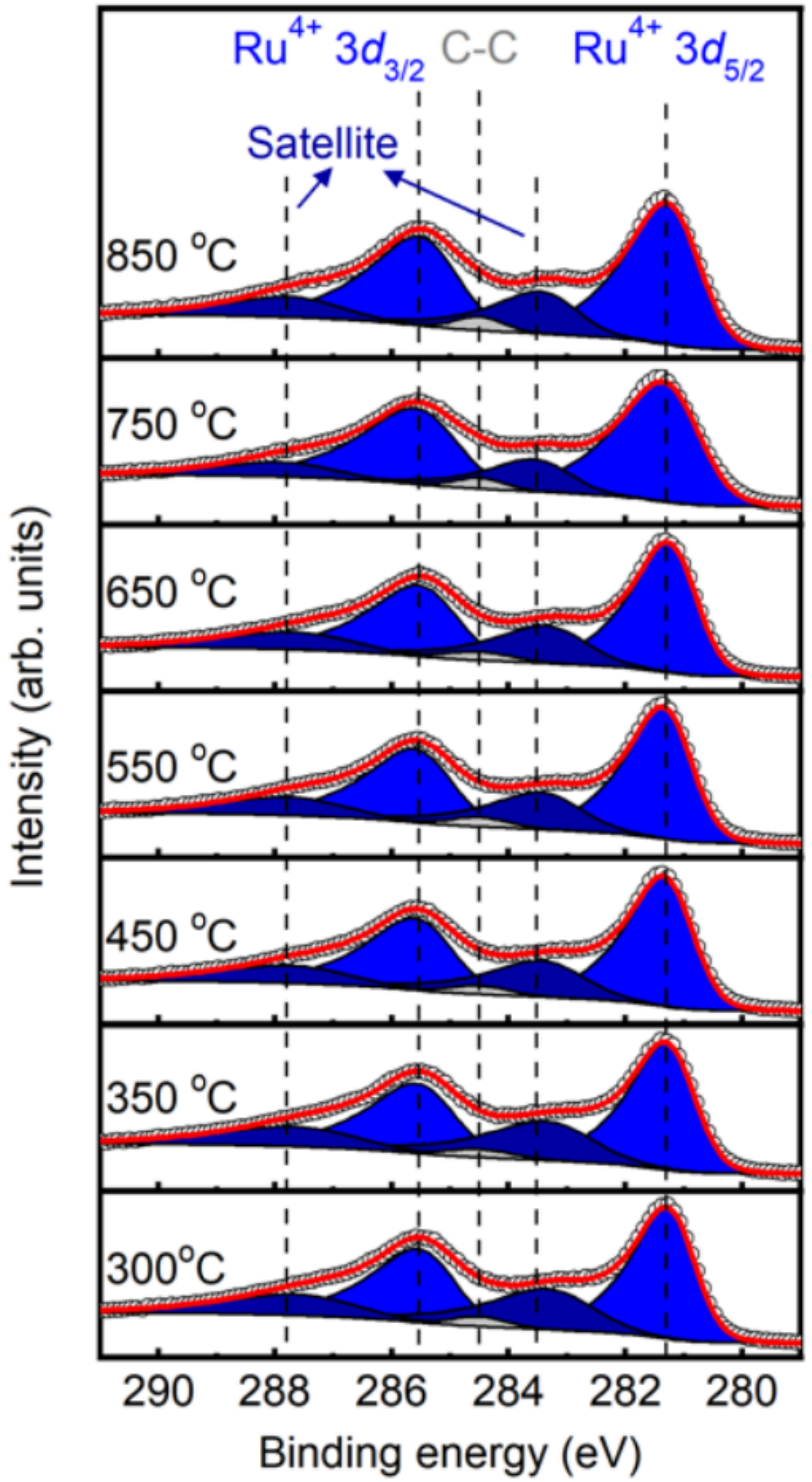
T_{sub} = 300 °C

Phase-pure, epitaxial films with Laue oscillations - evidence of excellent structural quality!

Epitaxial, phase-pure films



XPS confirms Ru^{4+}
(within the XPS resolution!)



W. Nunn, S. Nair, H. Yun, A. K. Manjeshwar,…and B. Jalan, *APL Mater.* 9, (2021) [Editors' Pick][Cover]

Residual Resistivity as a Sensitive Proxy to Disorder

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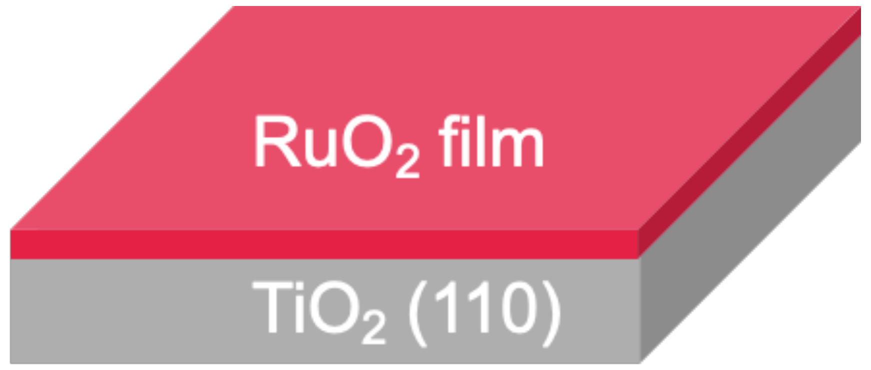
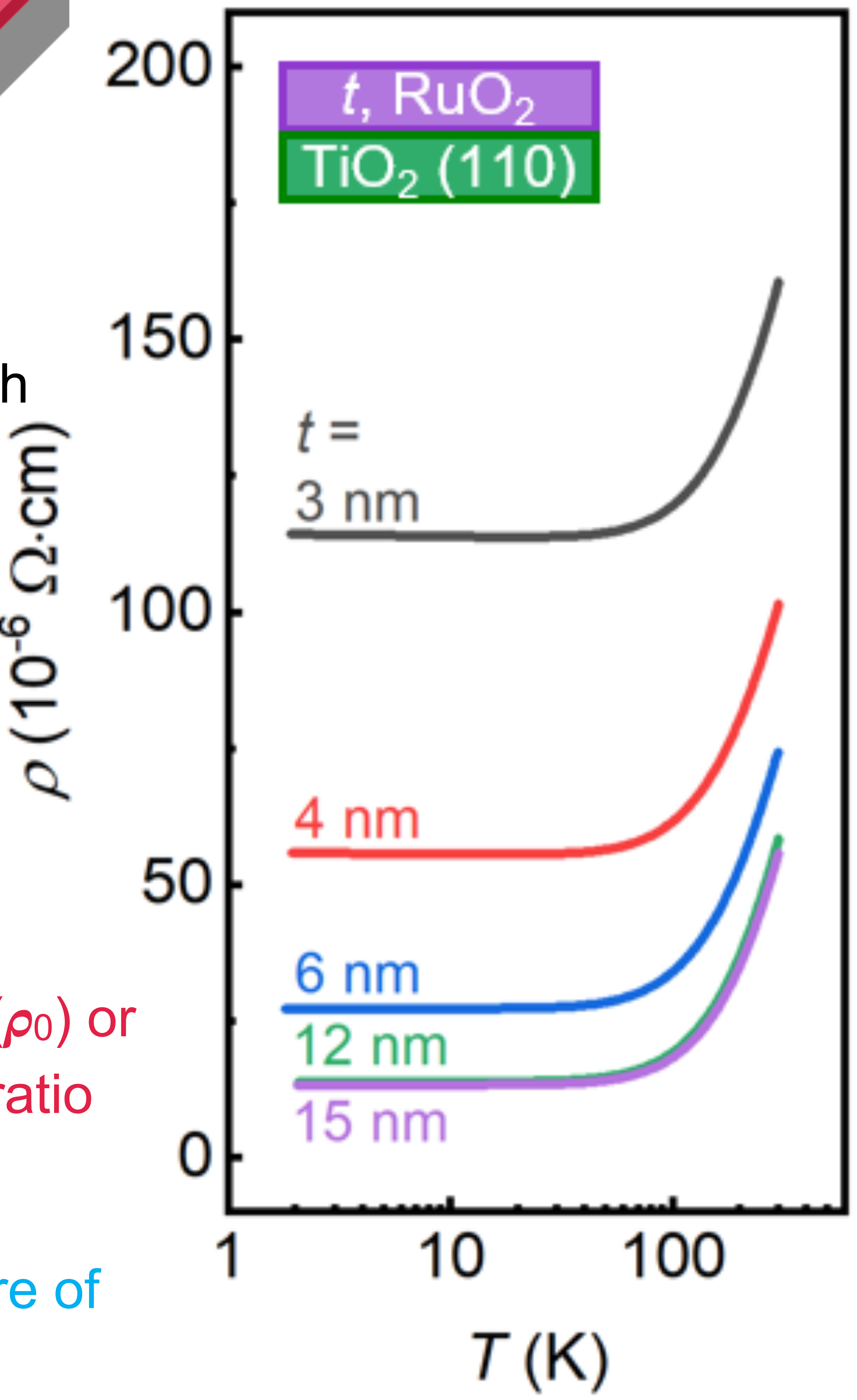


T_{sub} = 300 °C

Resistivity increases with decreasing film thickness!



residual resistivity (ρ_0) or residual resistivity ratio (RRR)

a sensitive measure of disorder!

W. Nunn, S. Nair, H. Yun, A. K. Manjeshwar,…and B. Jalan, *APL Mater.* 9, (2021) [Editors' Pick][Cover]

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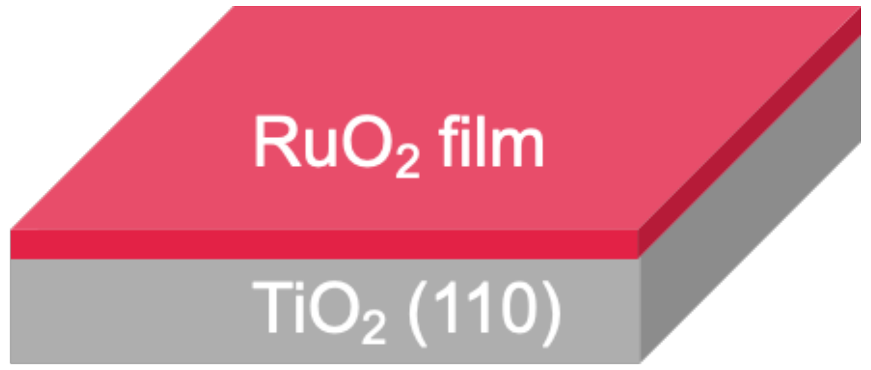

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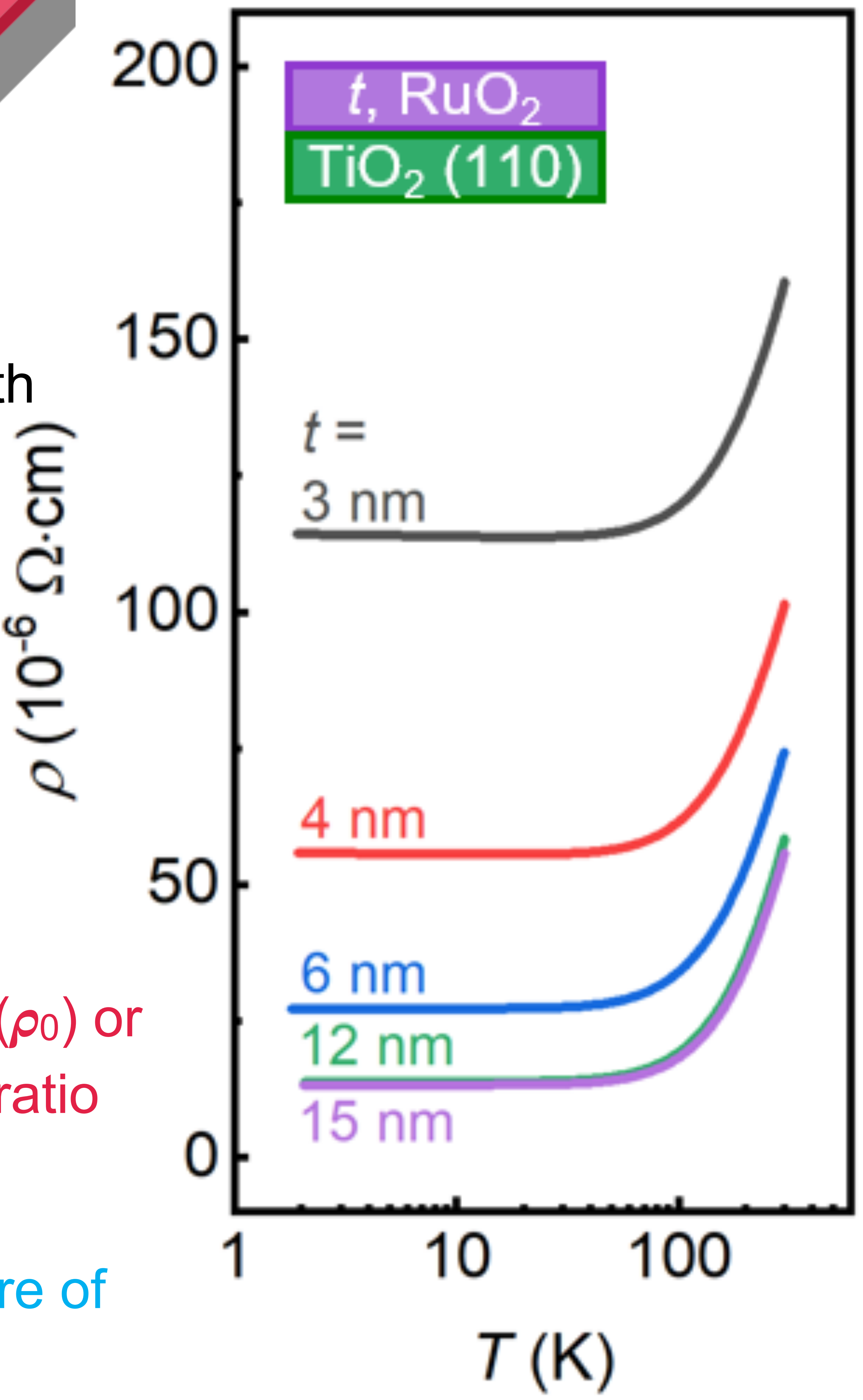
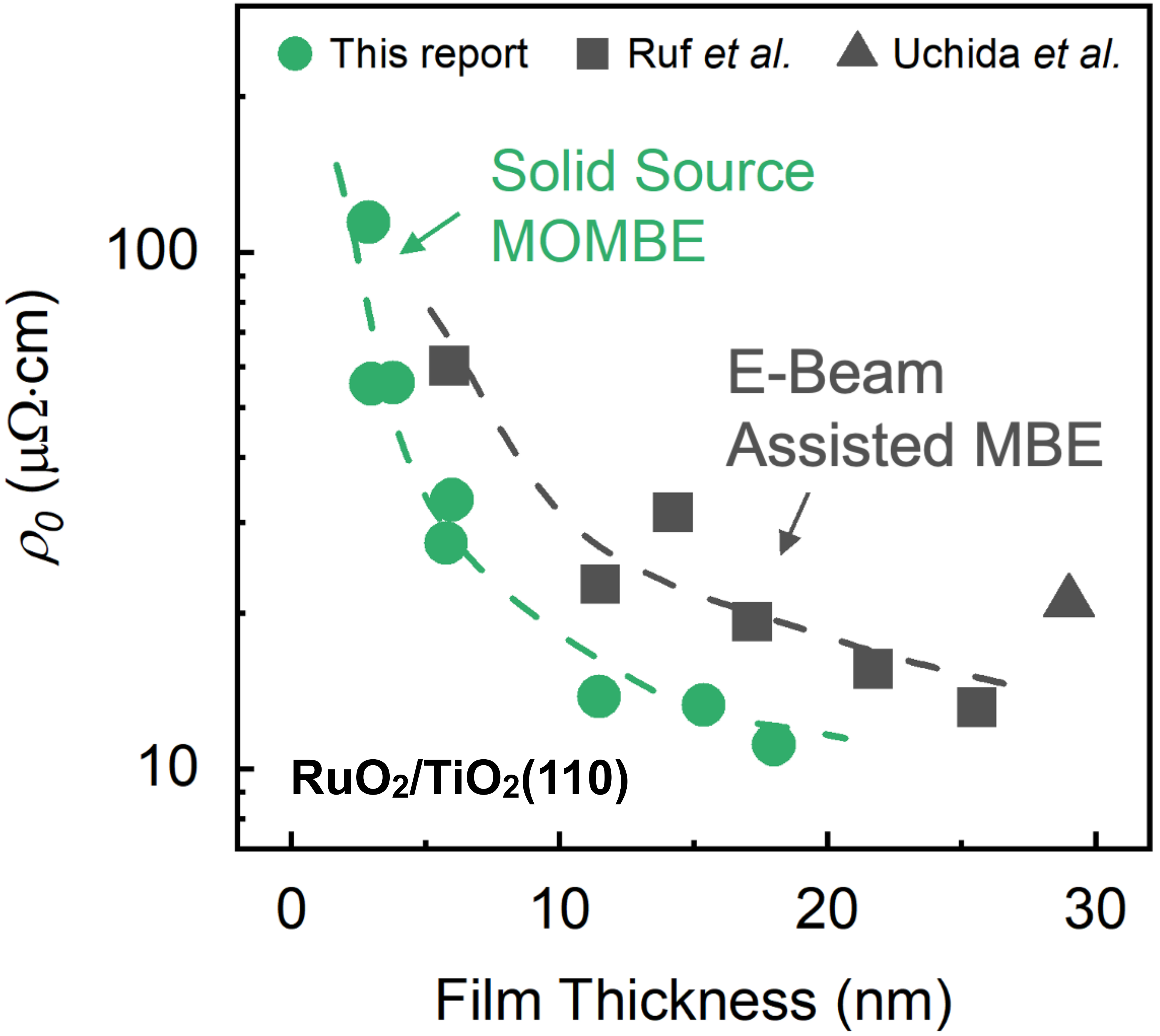


residual resistivity (ρ_0) or residual resistivity ratio (RRR)

a sensitive measure of disorder!

Comparable residual resistivity compared to the films grown using ozone-assisted MBE



W. Nunn, S. Nair, H. Yun, A. K. Manjeshwar,…and B. Jalan, *APL Mater.* 9, (2021) [Editors' Pick][Cover]

Anisotropic Strain Relaxation in RuO_2/TiO_2 (110)

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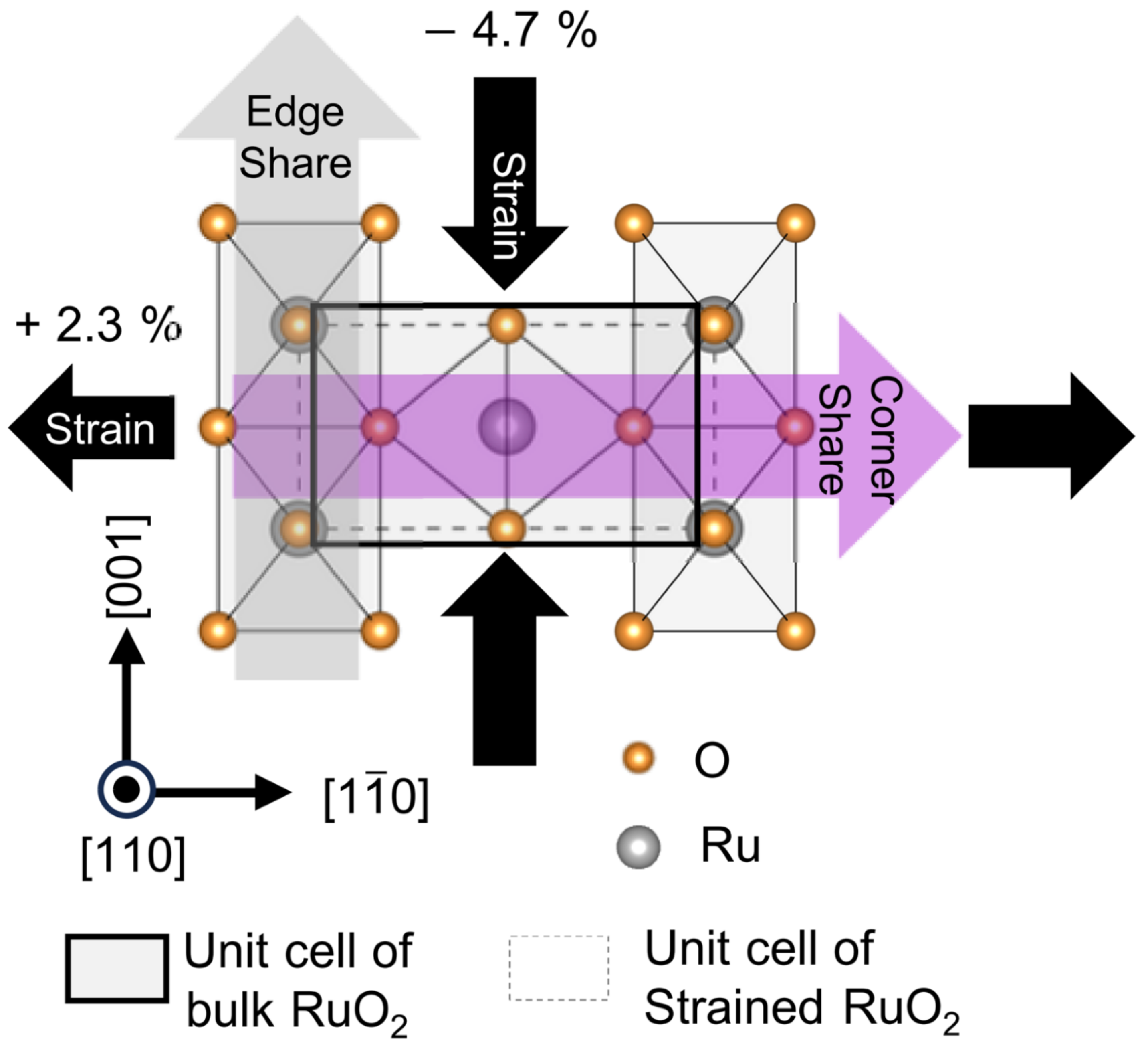

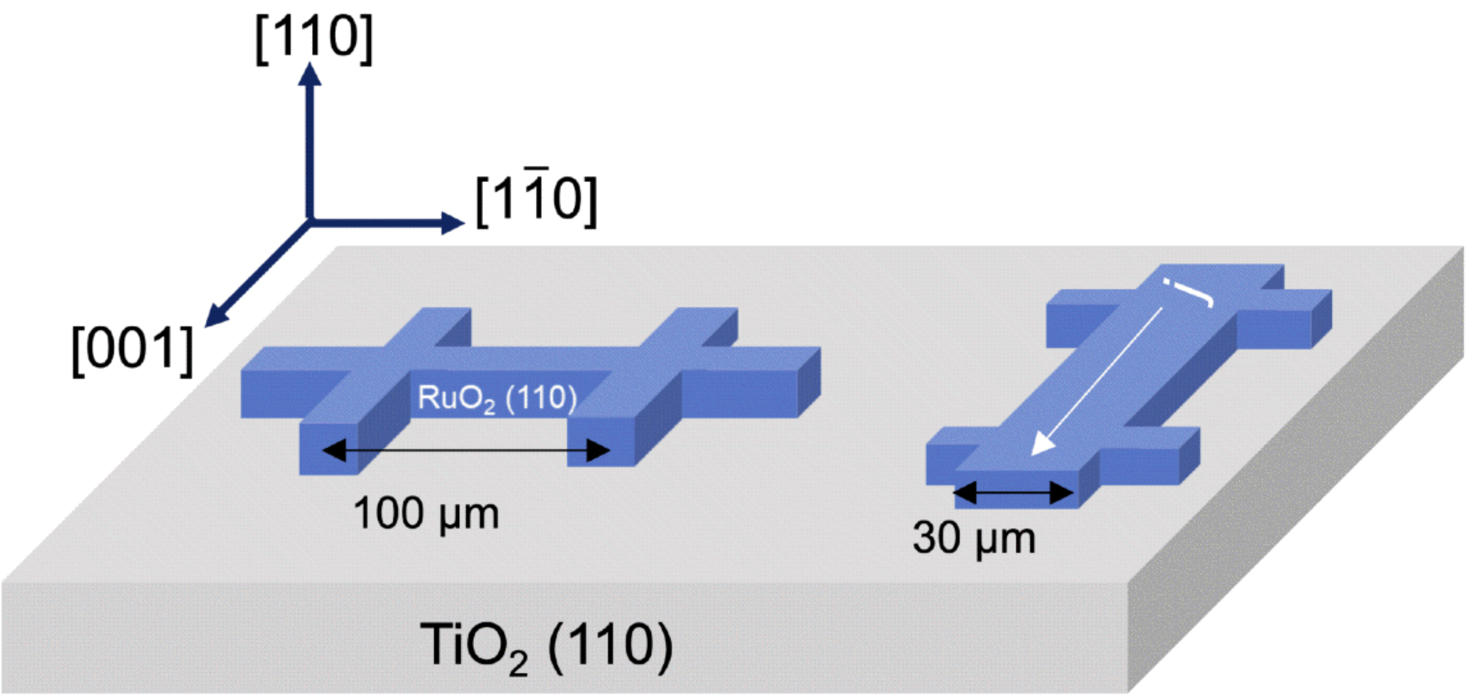

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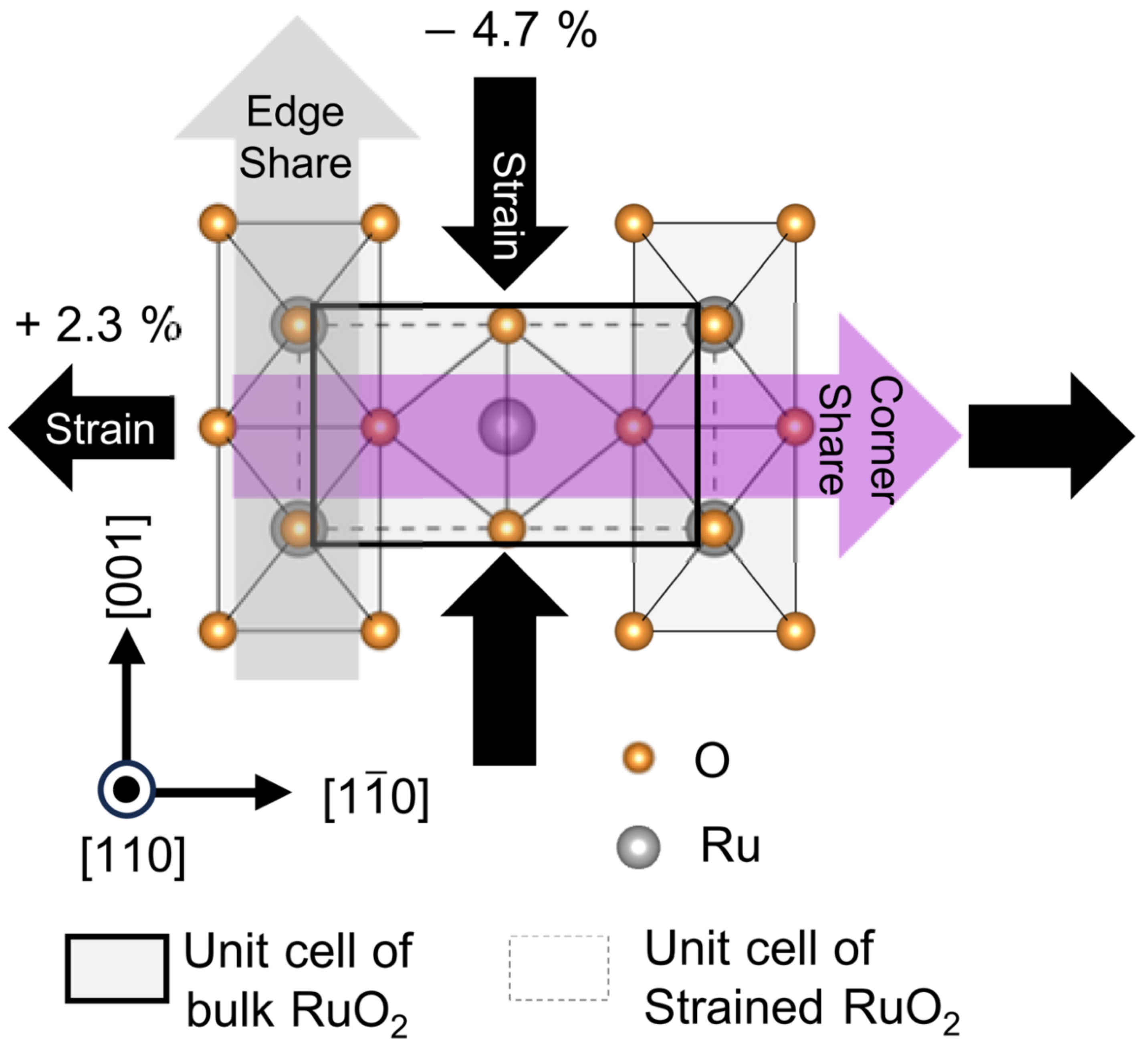
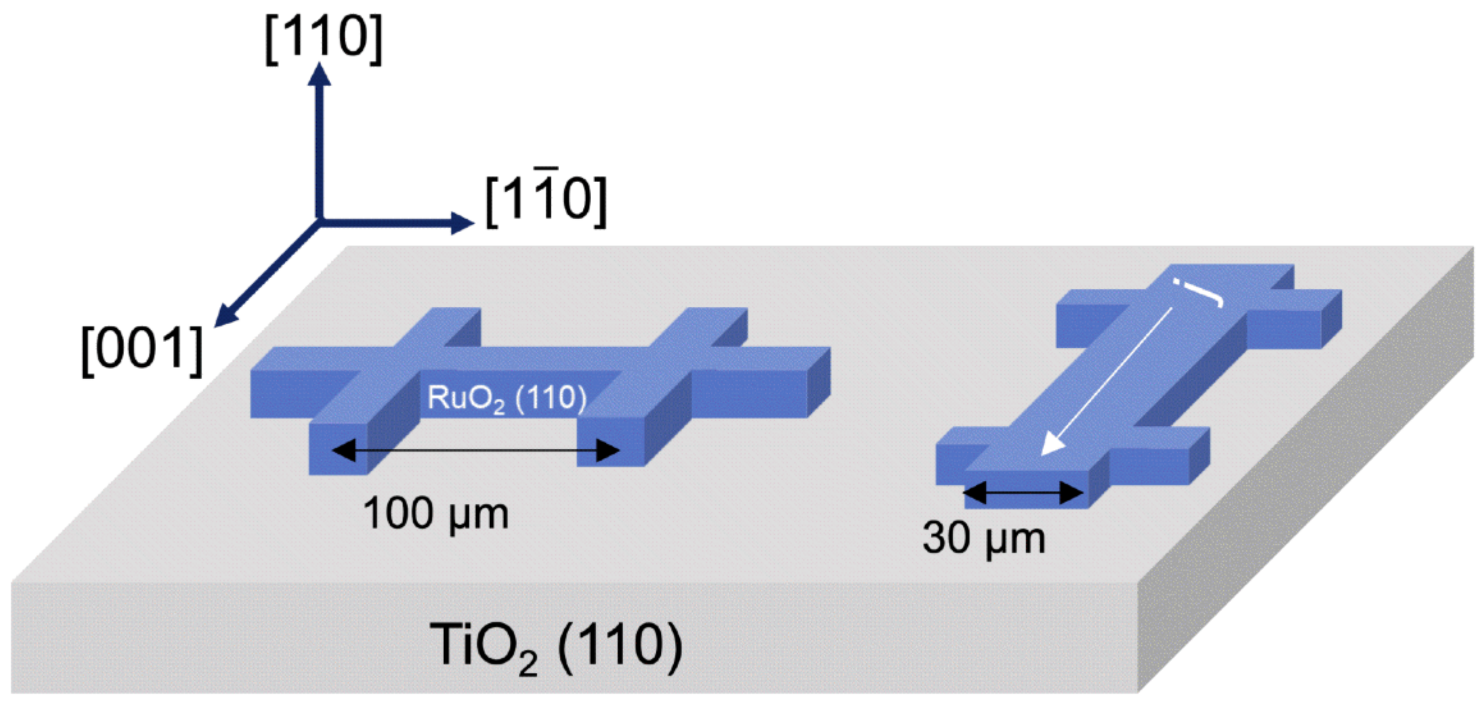
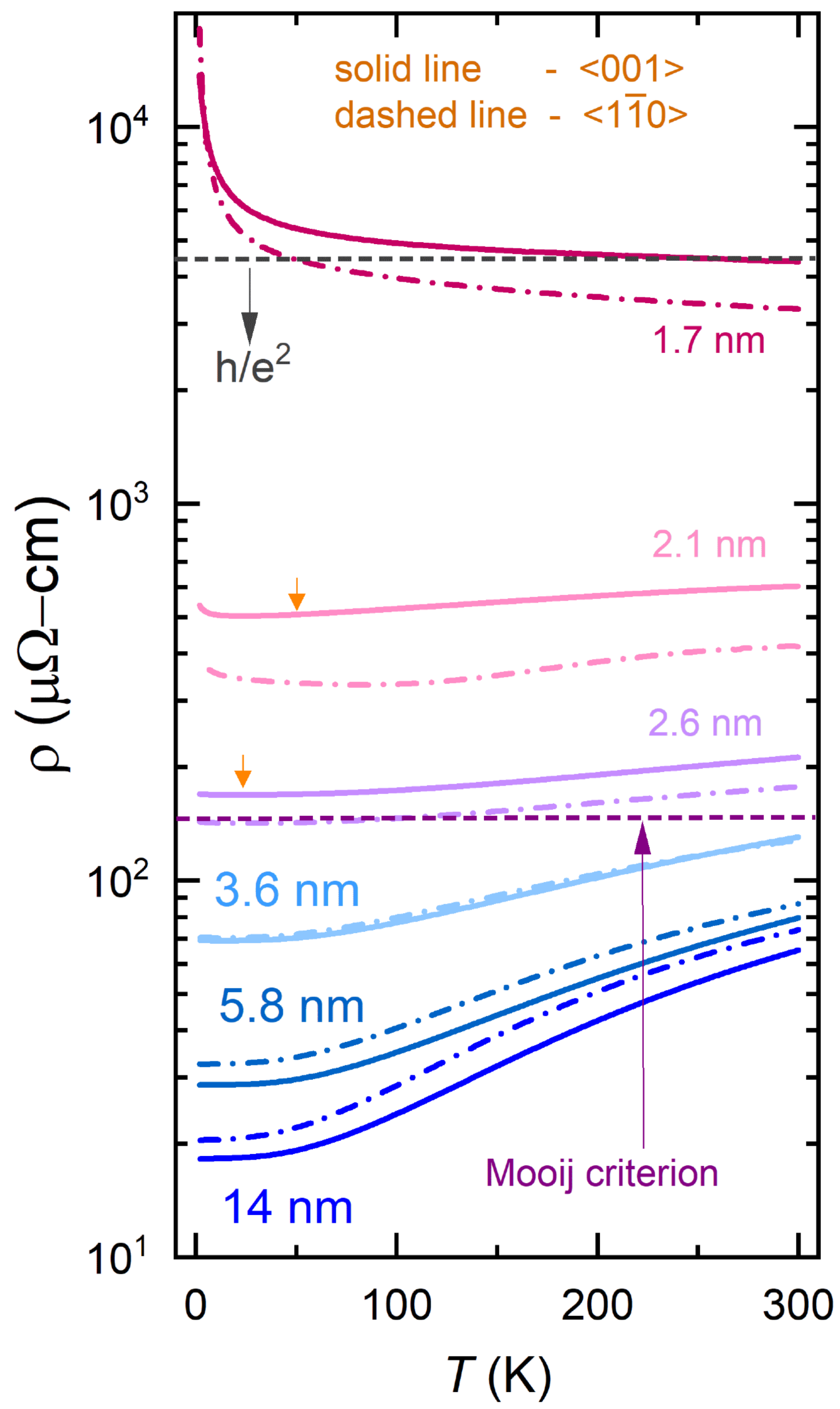




Metal-to-insulator transition as t_{film} ↓



A. K. Rajapitamahuni, S. Nair, Z. Yang, A. K. Manjeshwar, S. G. Jeong, W. Nunn, and B. Jalan, Phys. Rev. Mater. 8, 075002 (2024)

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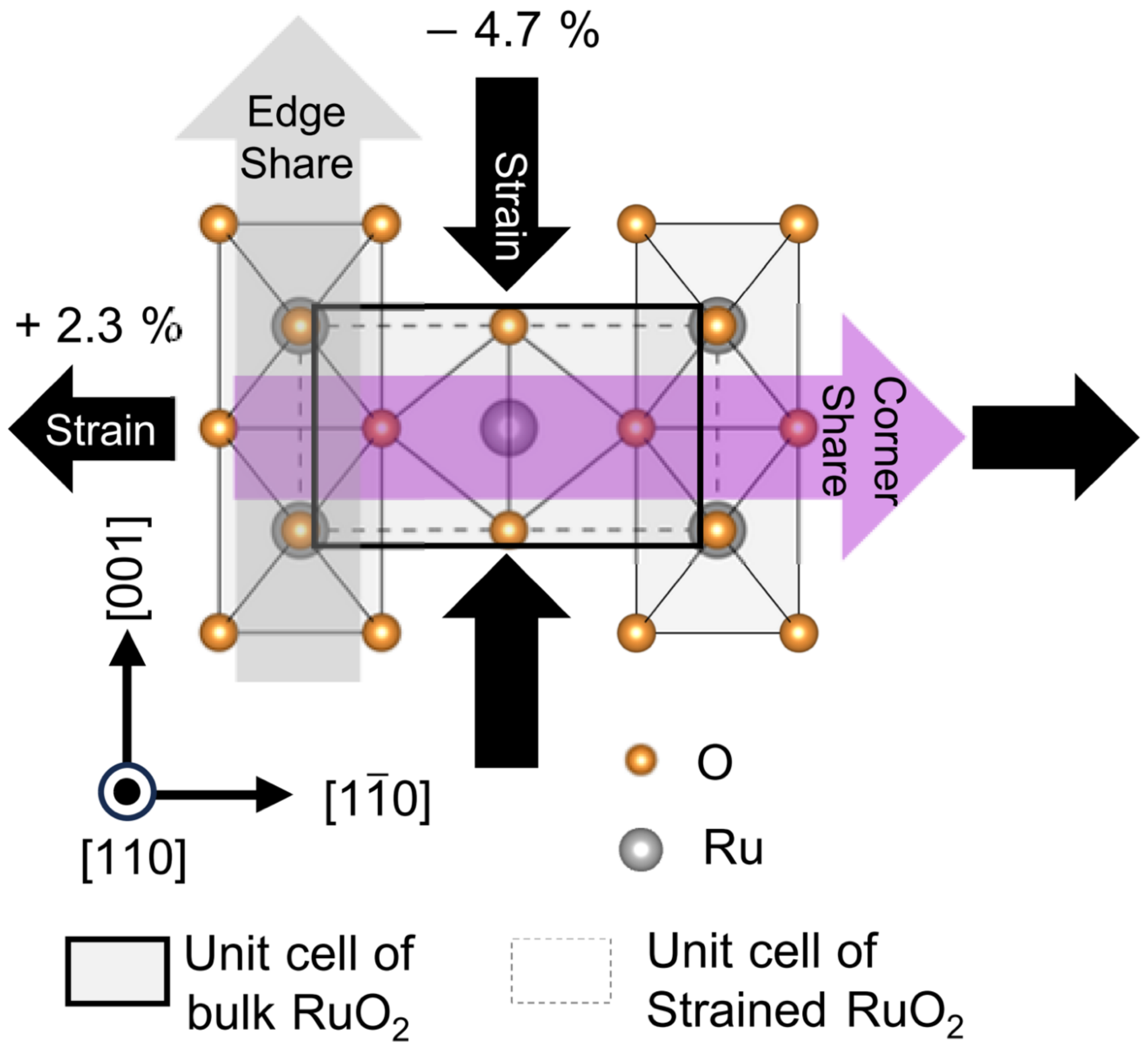
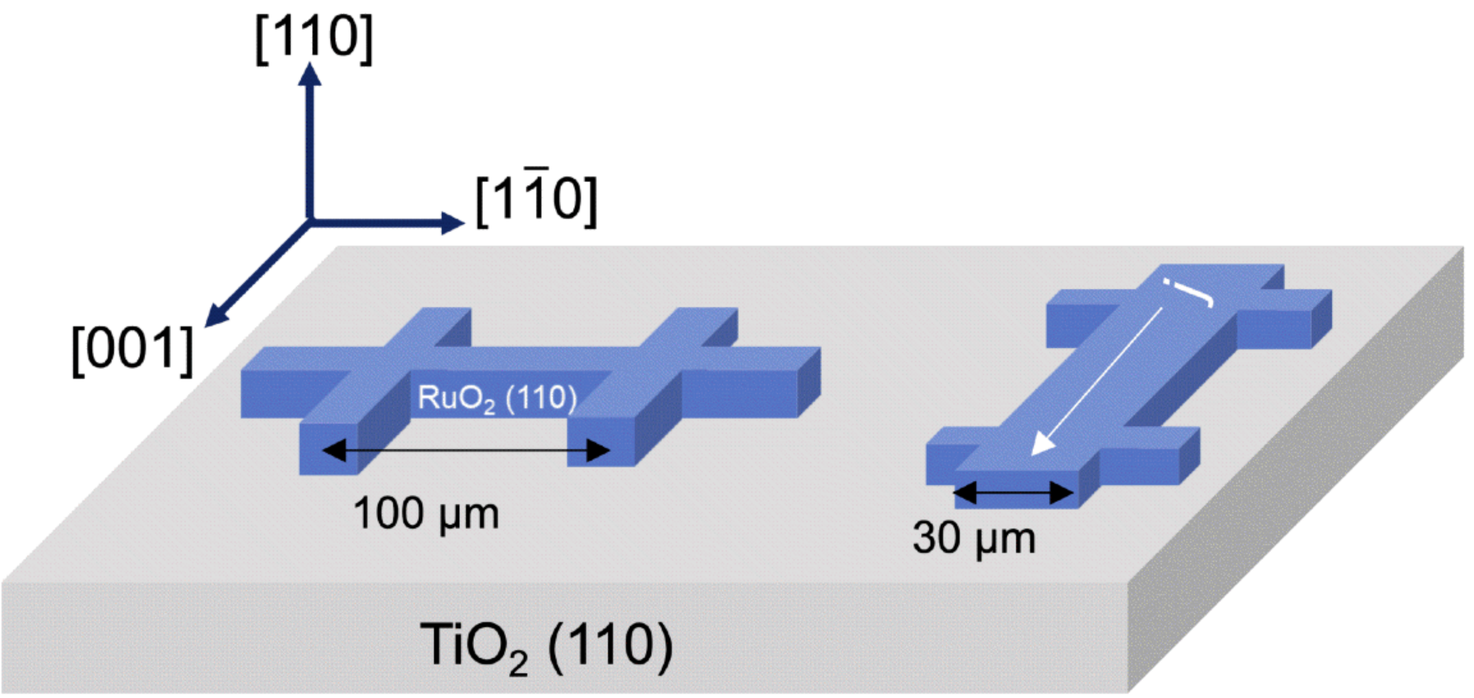
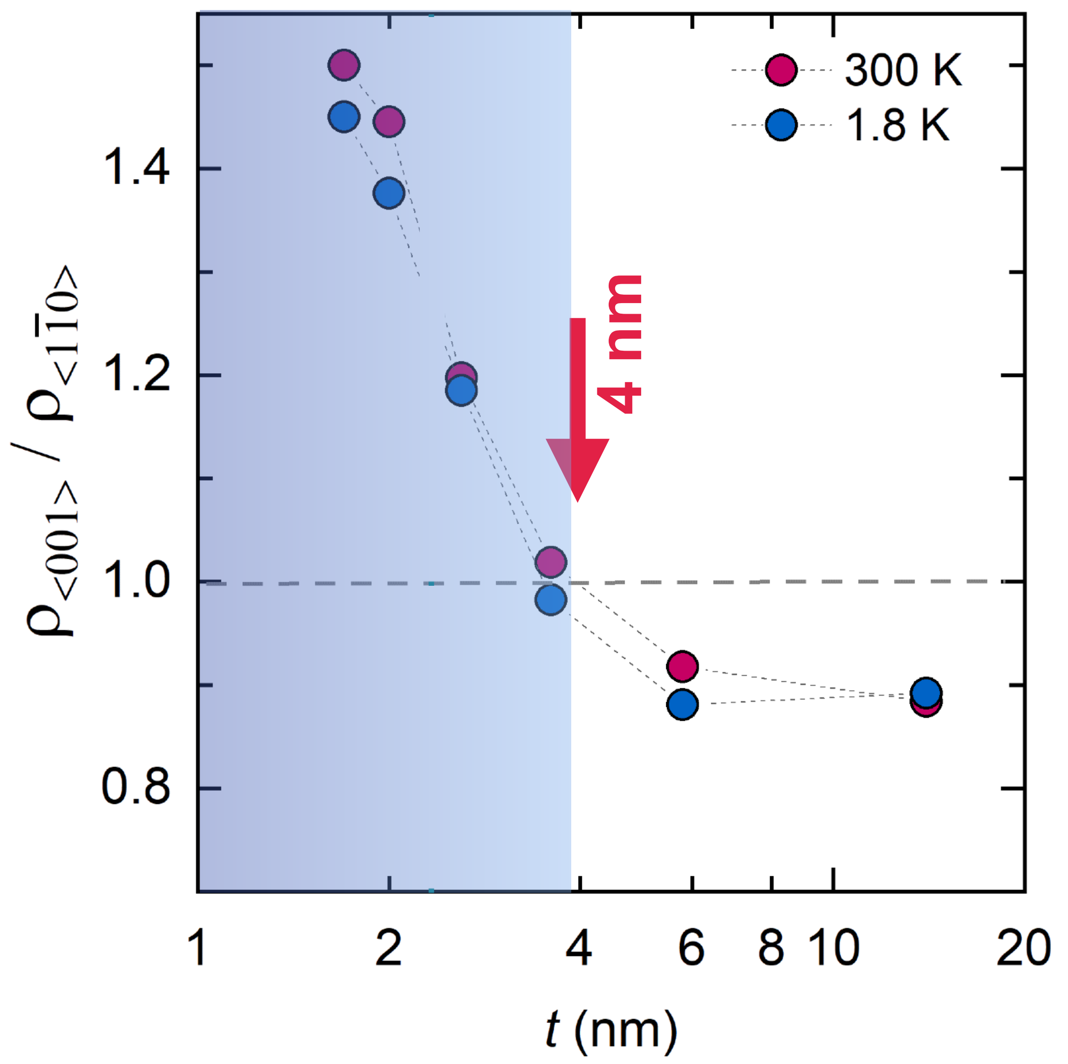




ρ ([1$\bar{1}$0] exceeds ρ [001] at t_{film} > 4 nm)



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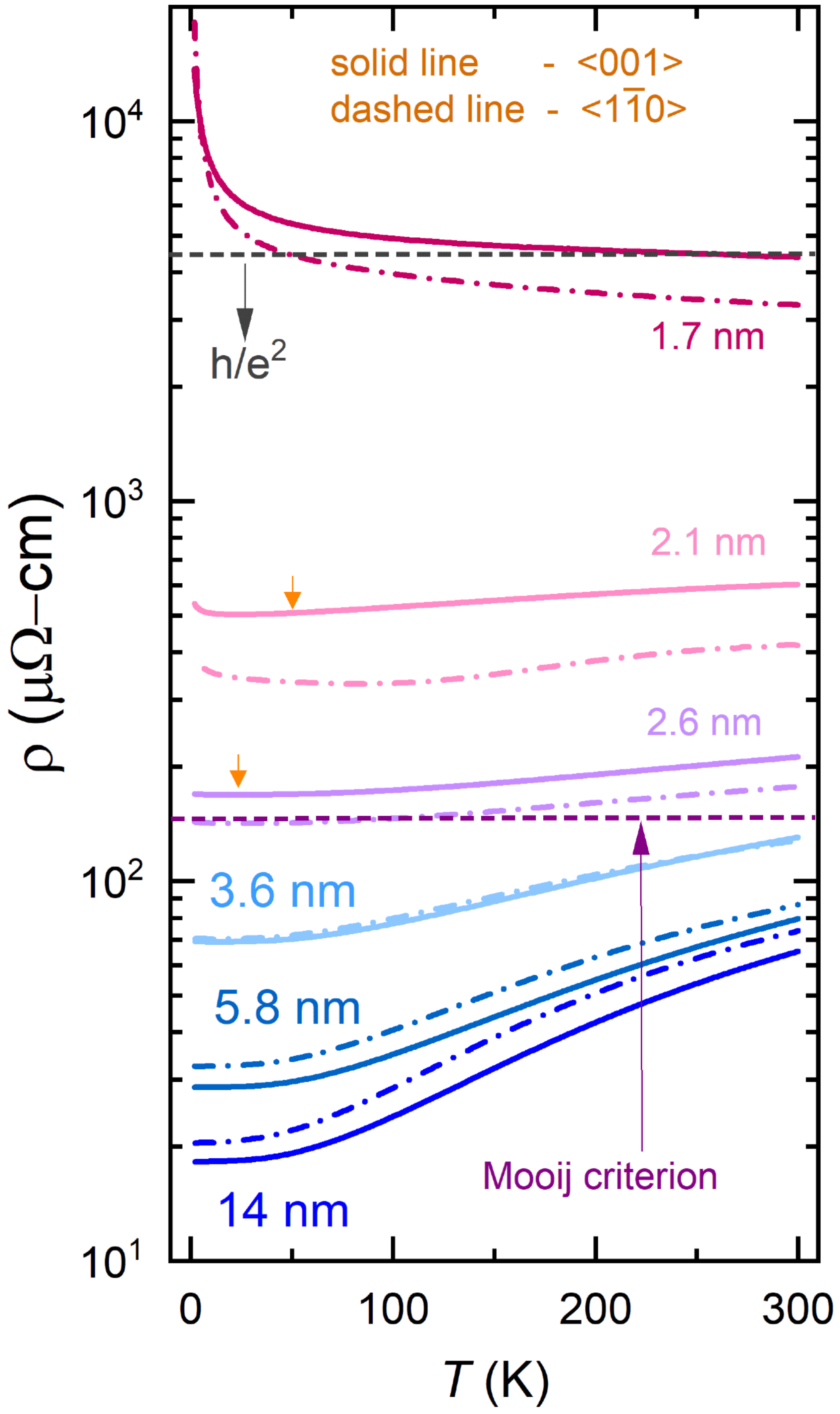

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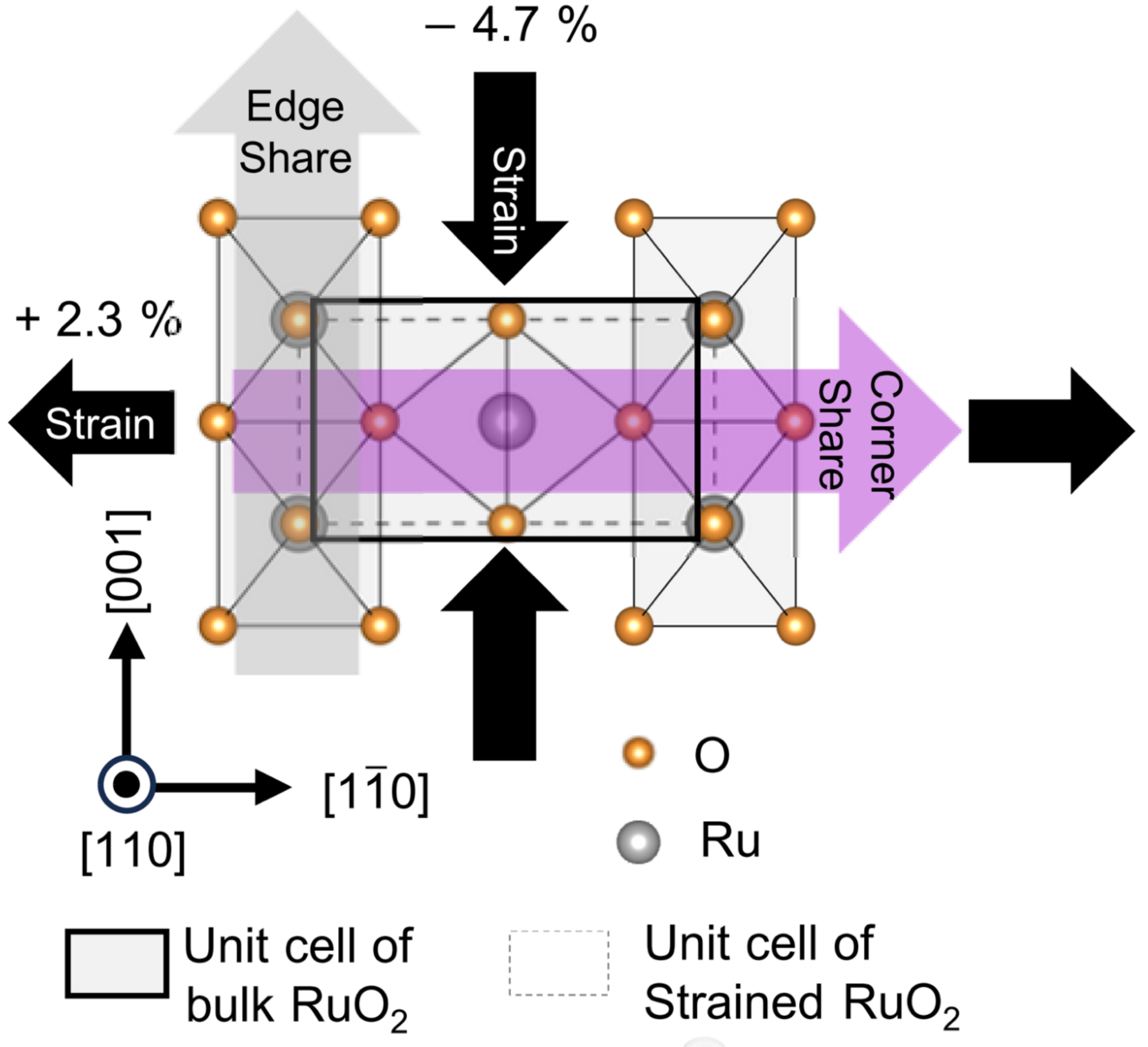
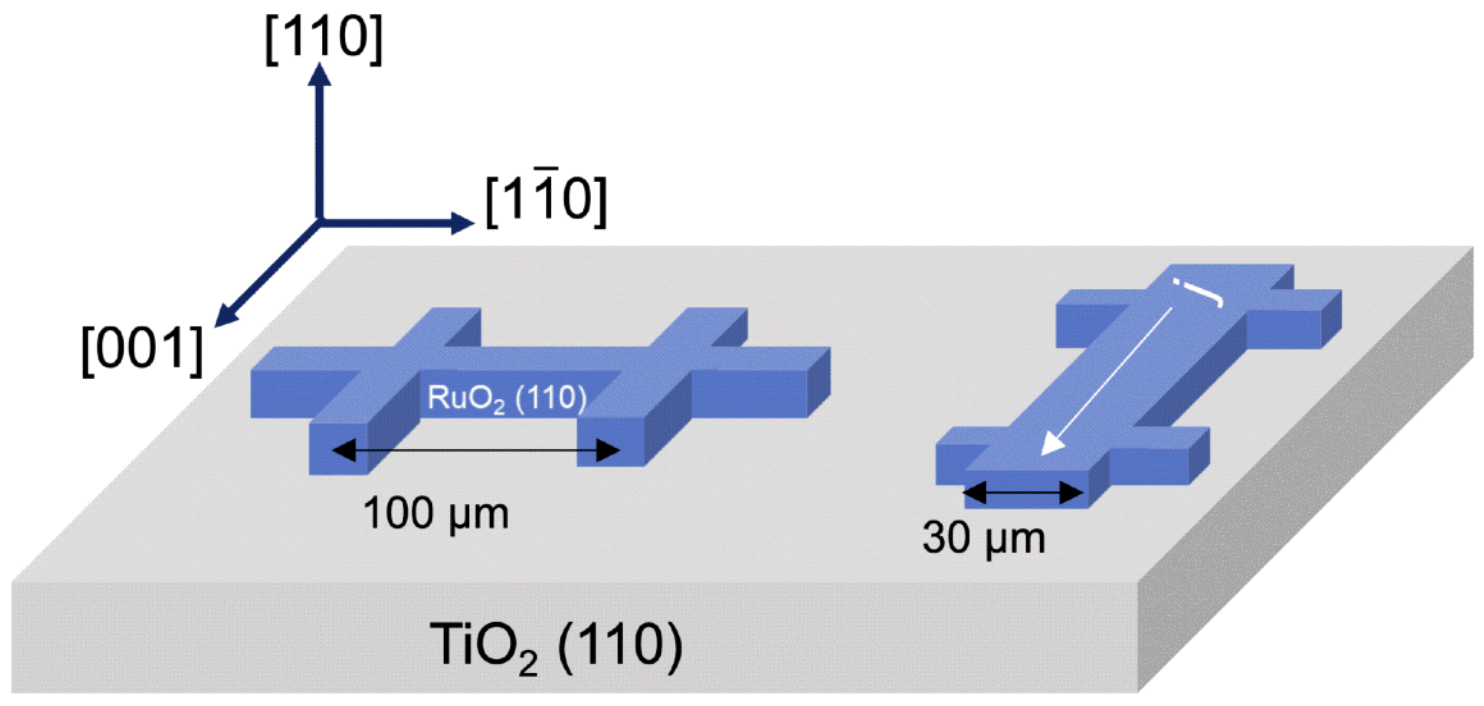
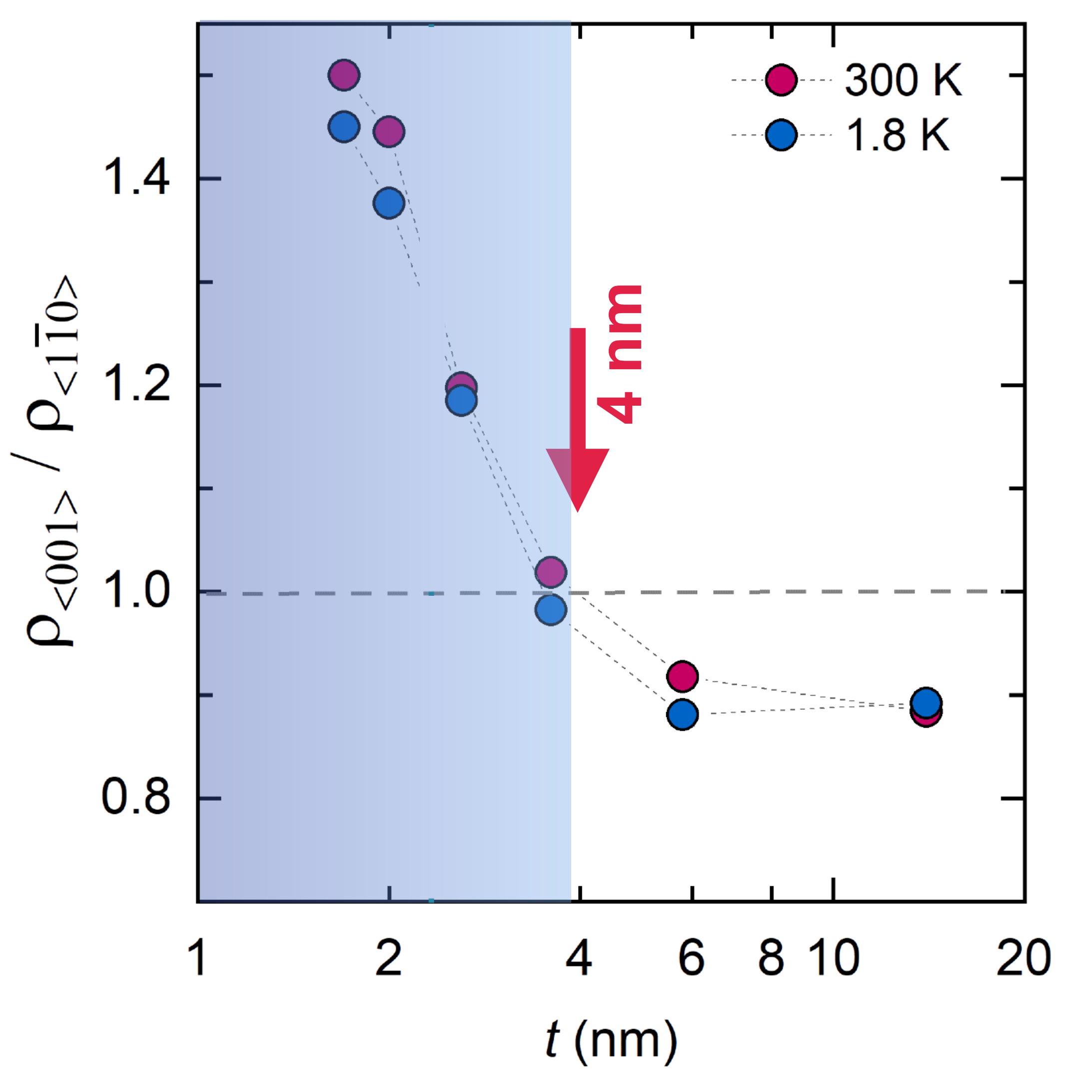
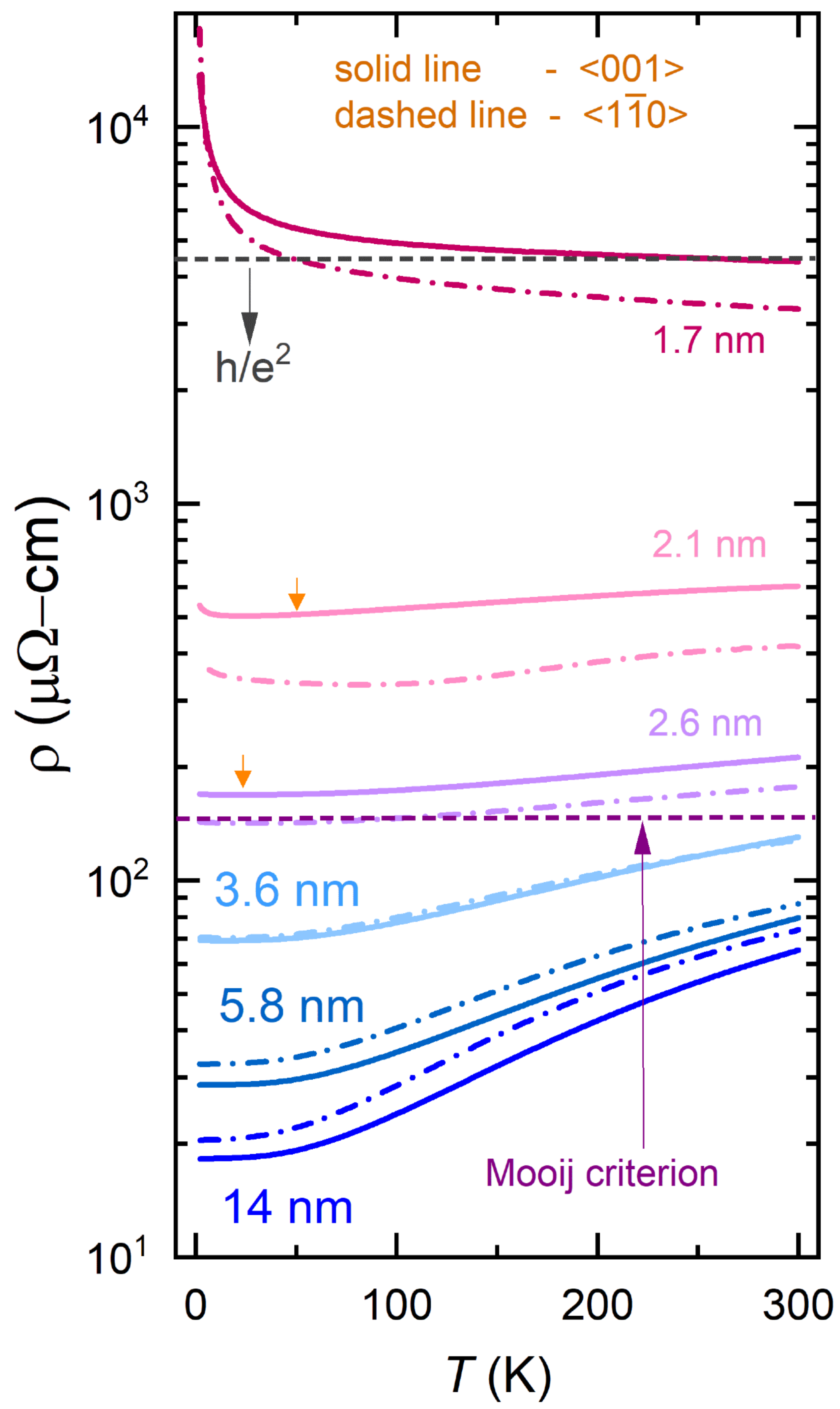




ρ ([$\bar{1}\bar{1}$0] exceeds ρ [00$\bar{1}$] at t_{film} > 4 nm)



Metal-to-insulator transition as t_{film} ↓



Open Questions

1. *Directional crossover in resistivity at 4 nm: Is it related to strain-relaxation?*

2. As t_{film} ↓, resistivity increases leading to M → I transition?
   structural disorder, surface, interface, or dimensionality?

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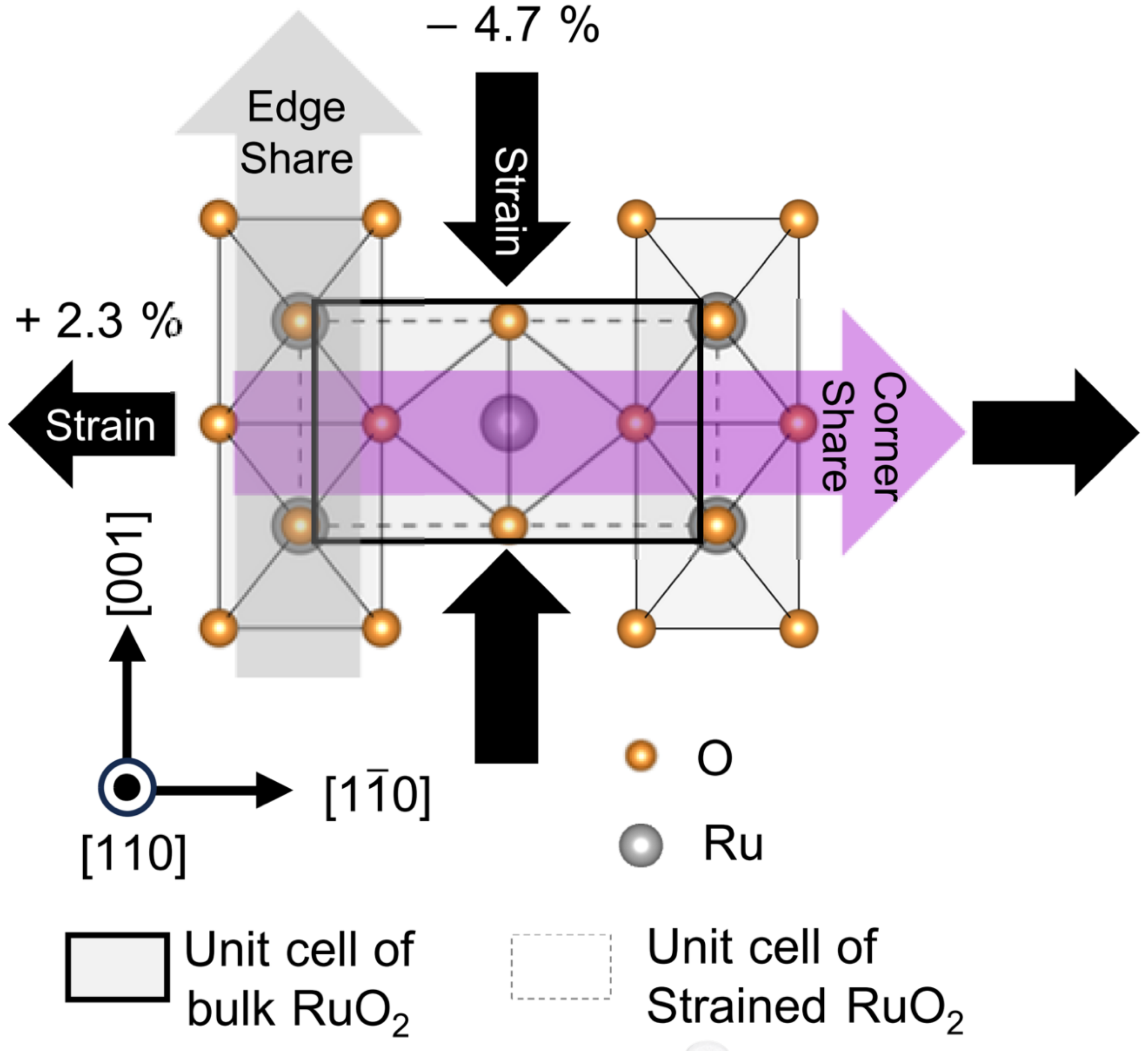

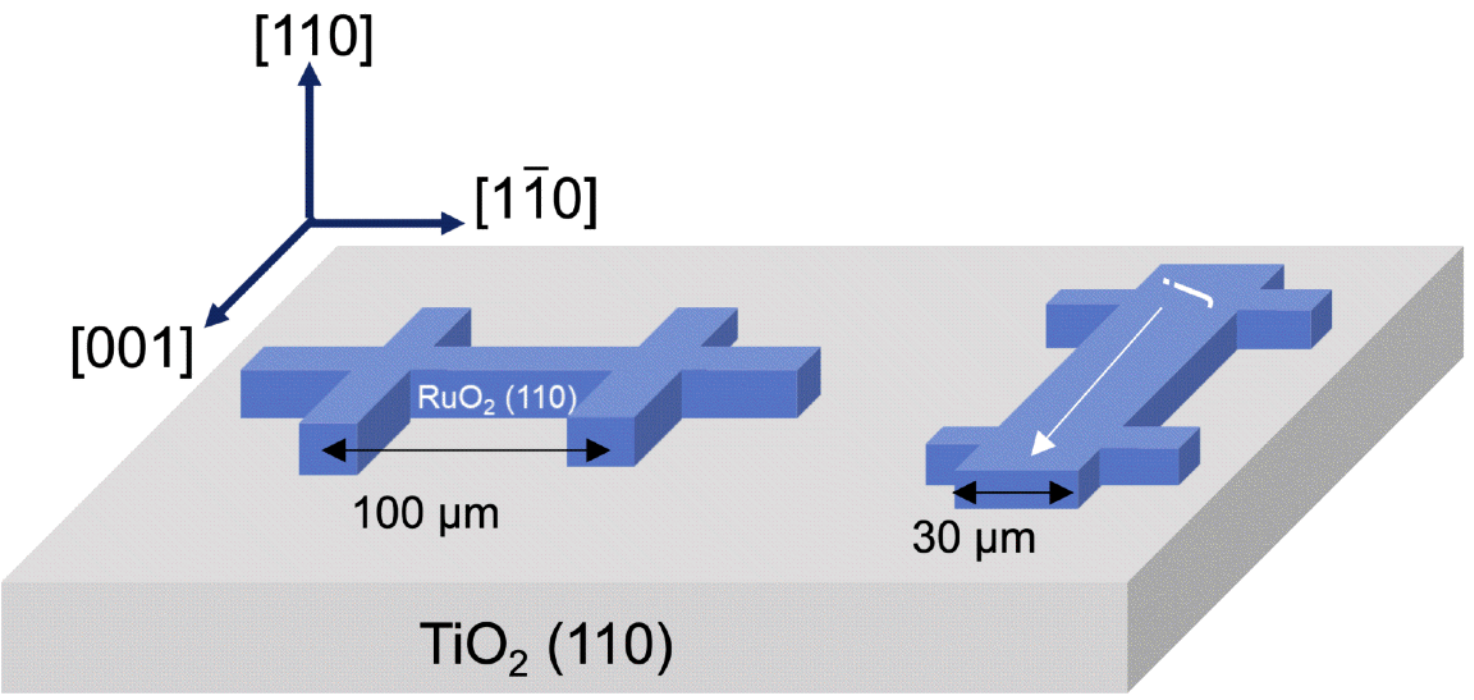

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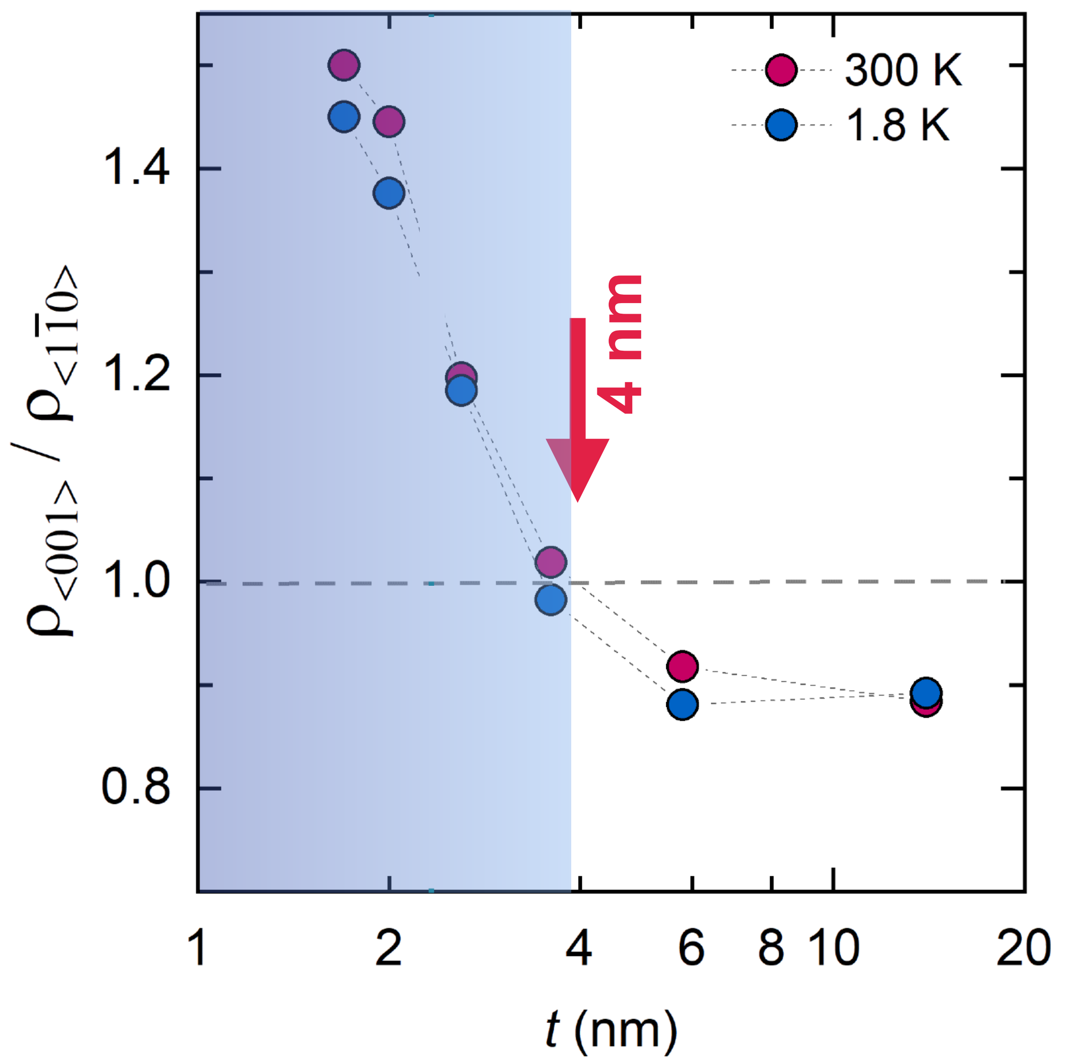
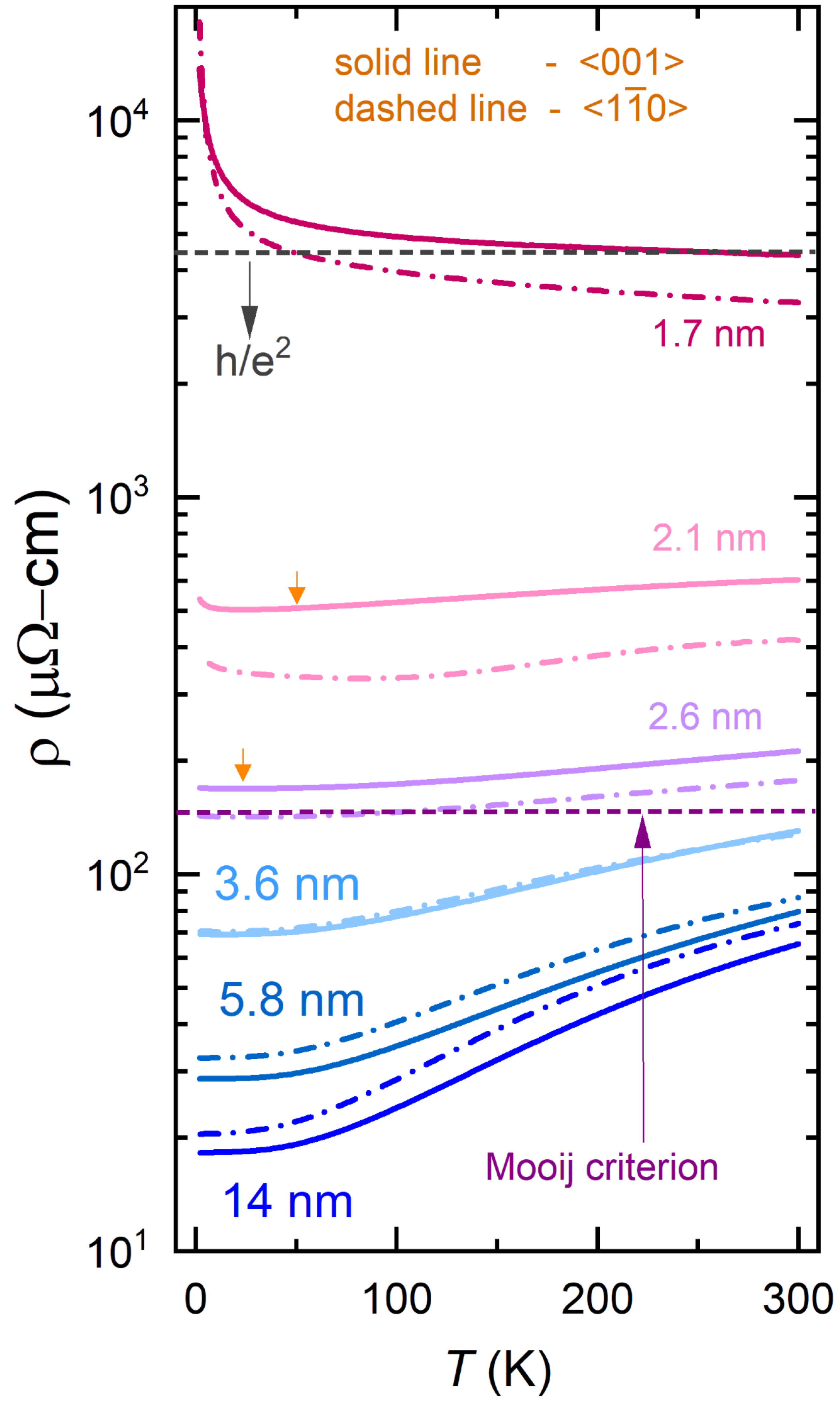


Metal-to-insulator transition as t_{film} ↓



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   structural disorder, surface, interface, or dimensionality?
   ✓Interface effect

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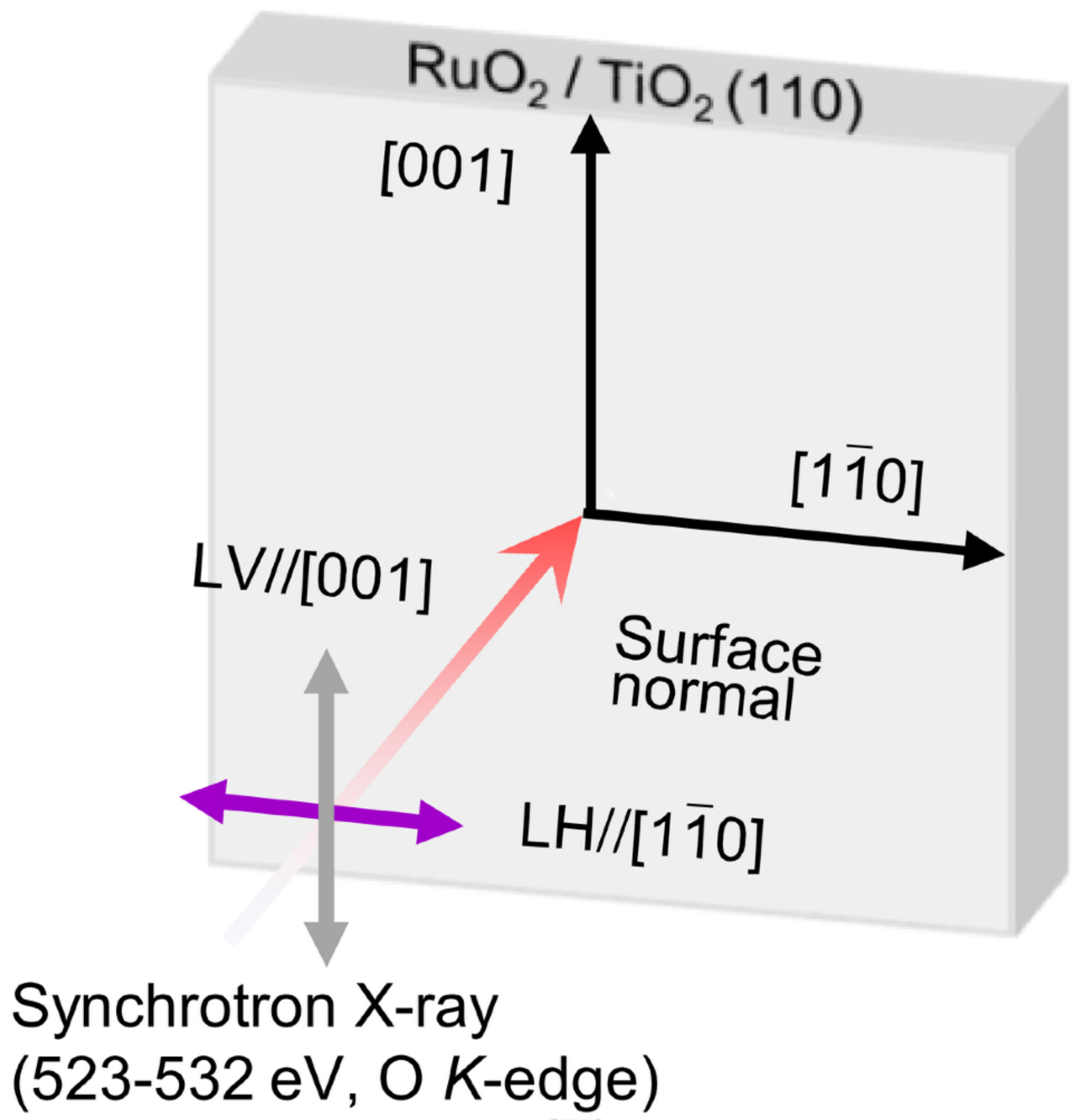

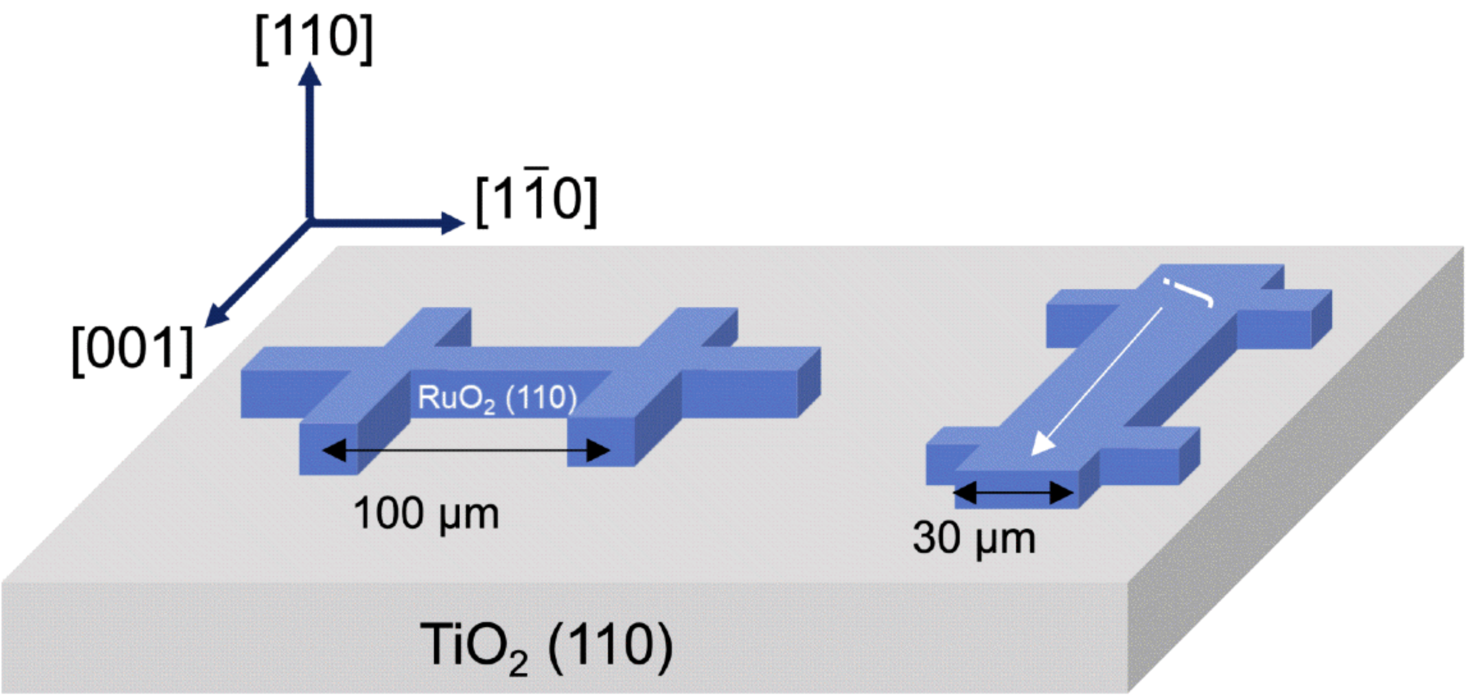

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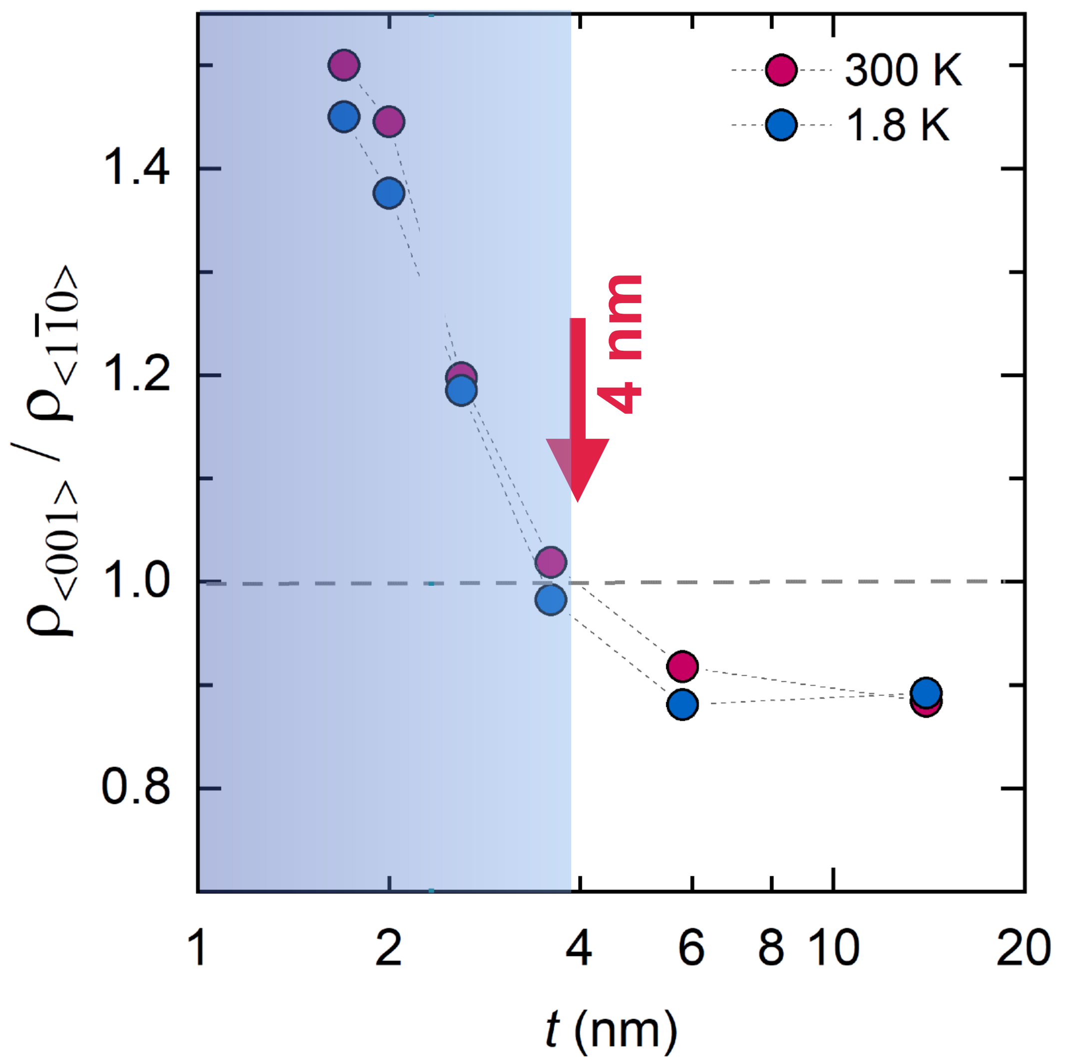
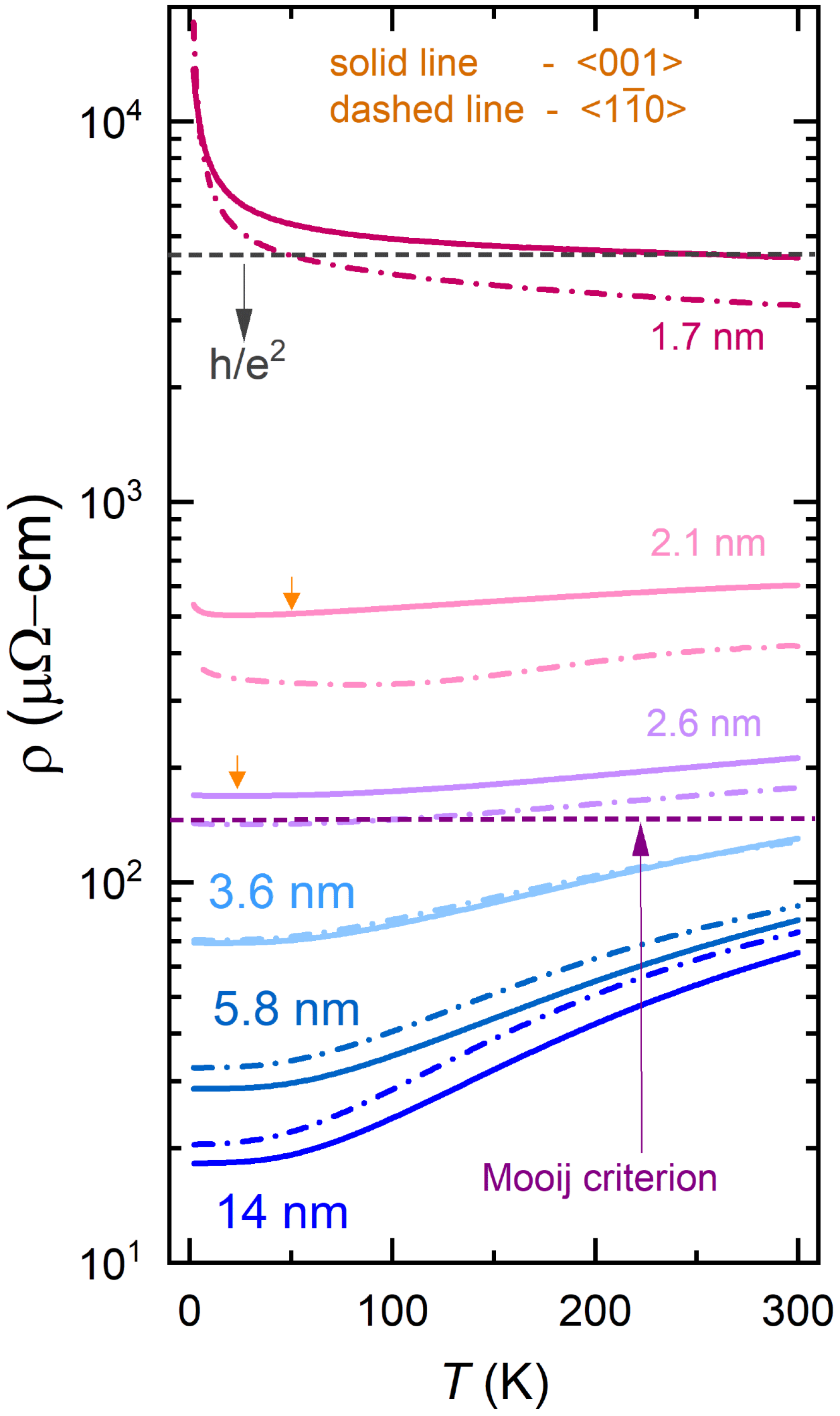


Metal-to-insulator transition as t_{film} ↓



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   structural disorder, surface, interface, or dimensionality?
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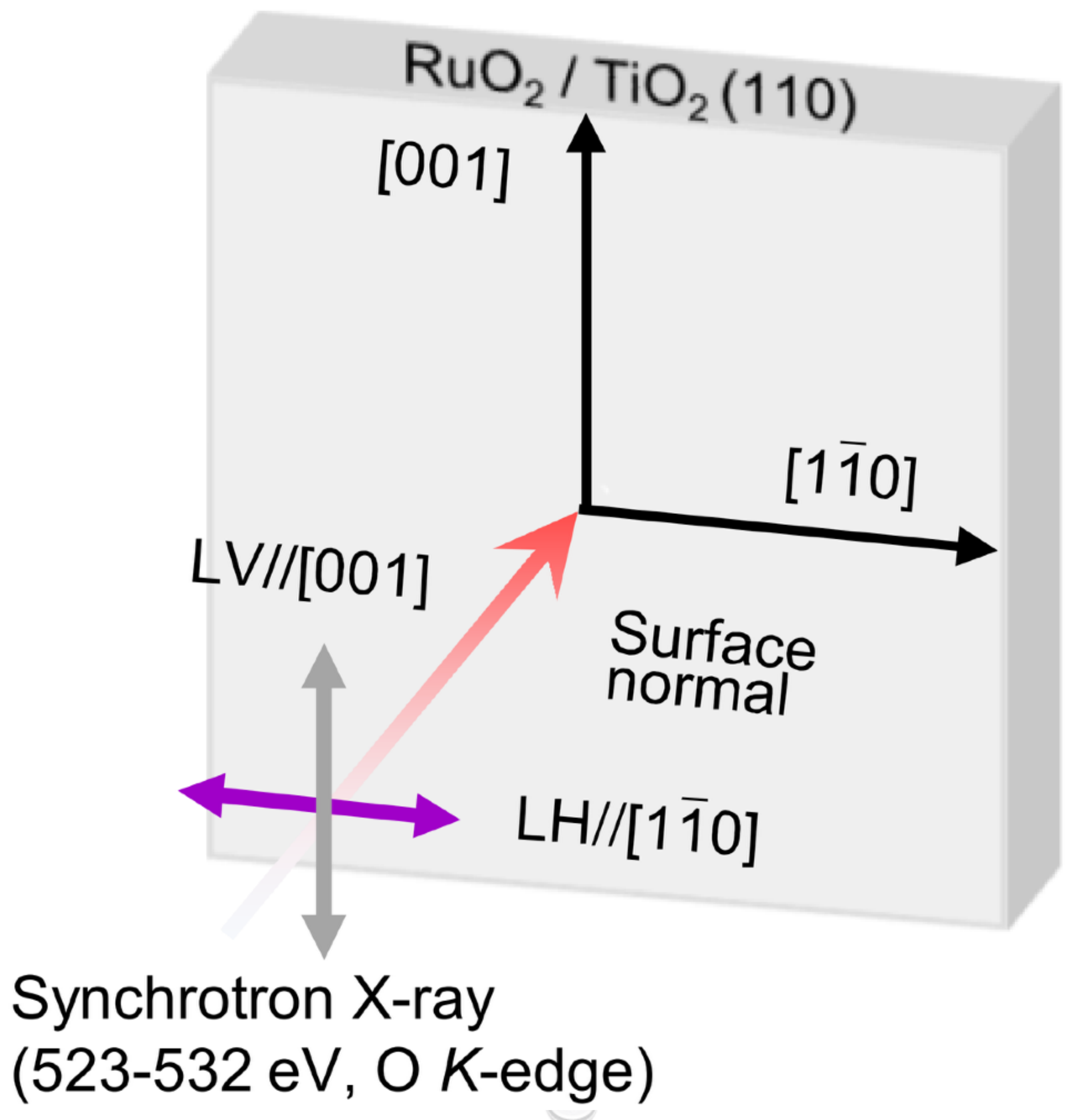

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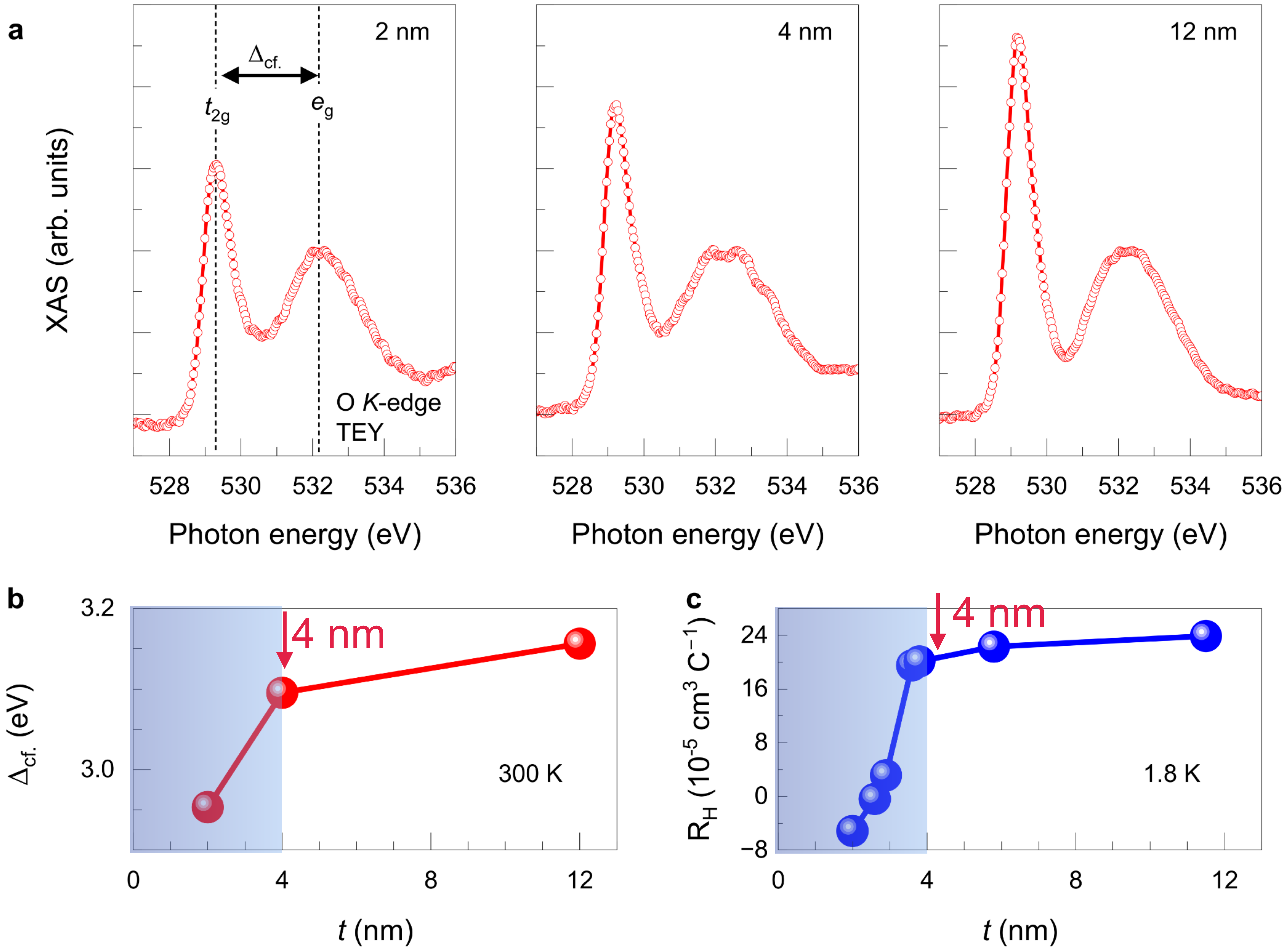
   ✓*Interface*

Significant change in crystal field energy at t_{film} < 4 nm (change in electronic band structure)

X-ray Absorption Spectroscopy (XAS)
courtesy: Prof. W. S. Choi, SKKU



S. G. Jeong, I. H. Choi, S. Lee, J. Y. Oh, S. Nair, J. H. Lee, C. Kim, A. Seo, W. S. Choi, T. Low, J. S. Lee, and B. Jalan, Anisotropic Strain Relaxation-Induced Directional Ultrafast Carrier Dynamics in RuO2 Films, Sci. Adv. 11, eadw7125 (2025)

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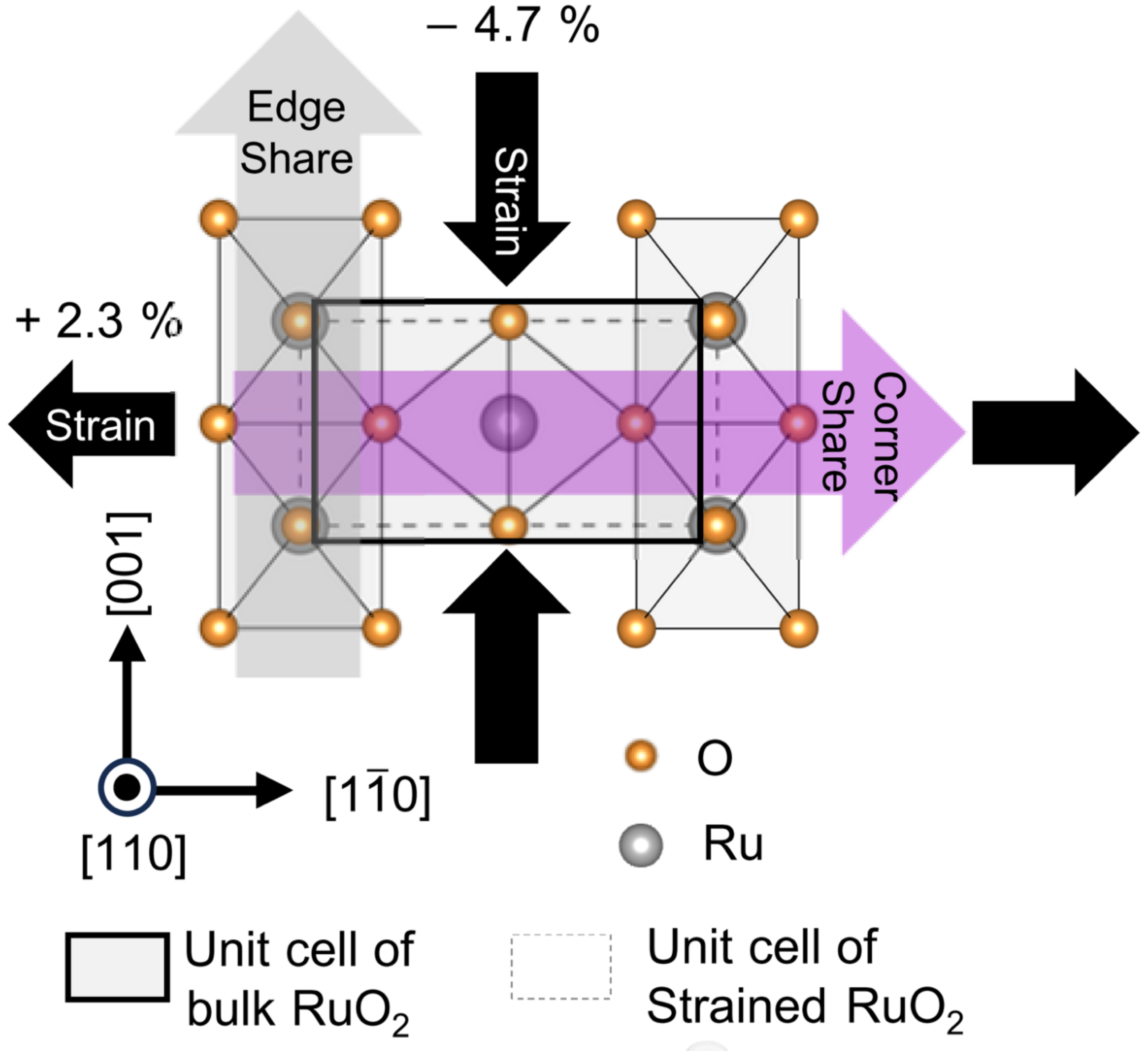

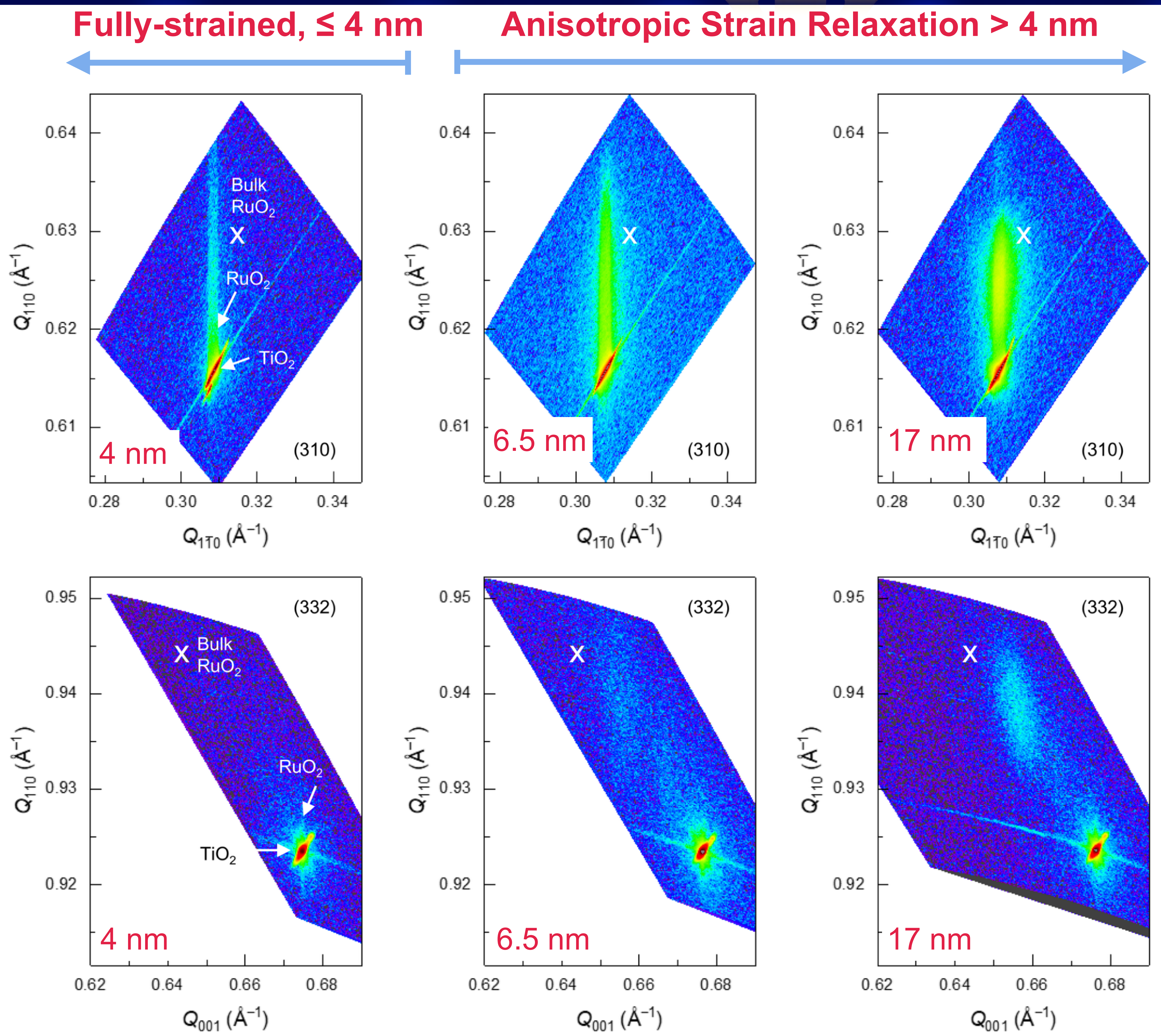

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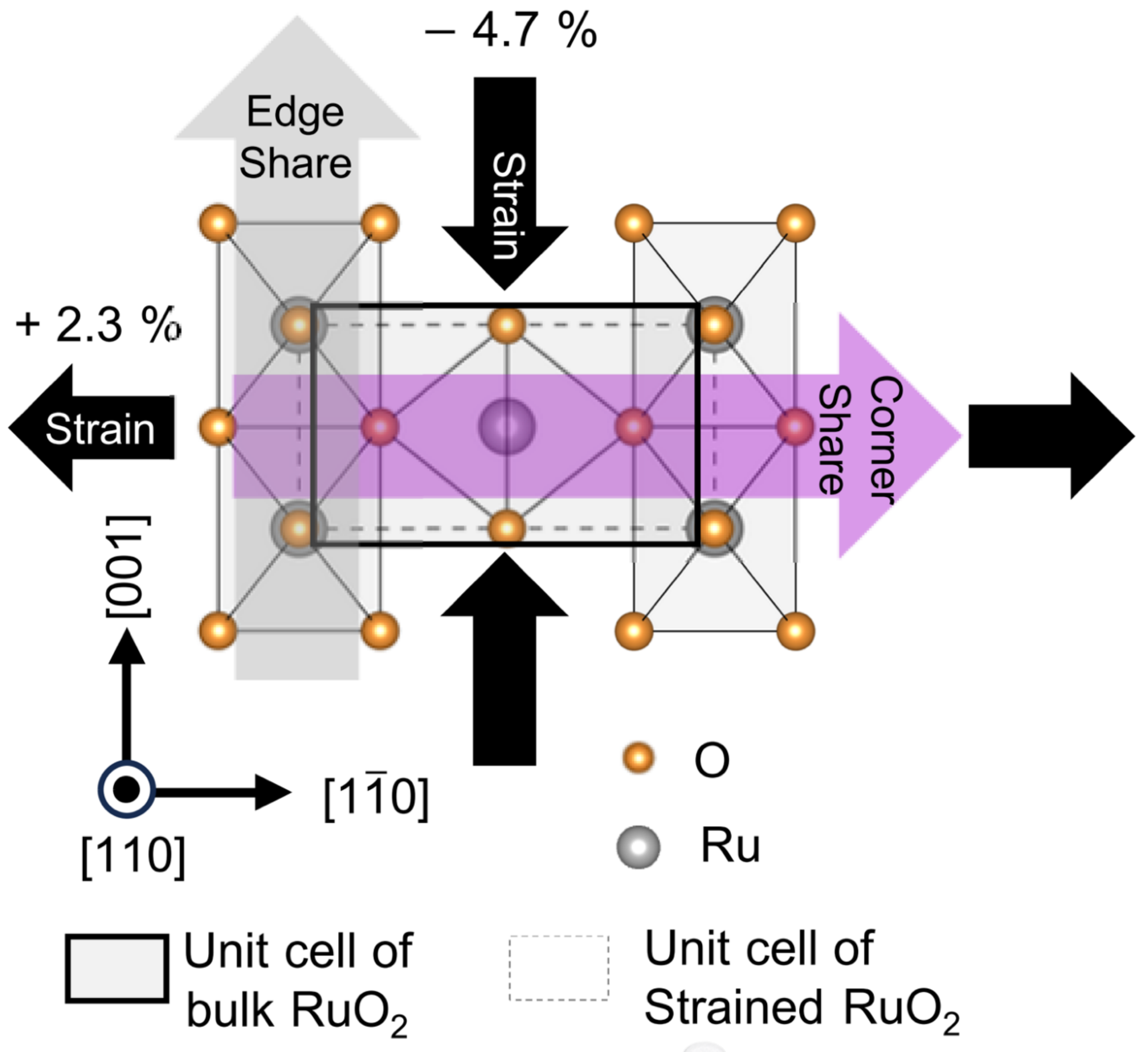

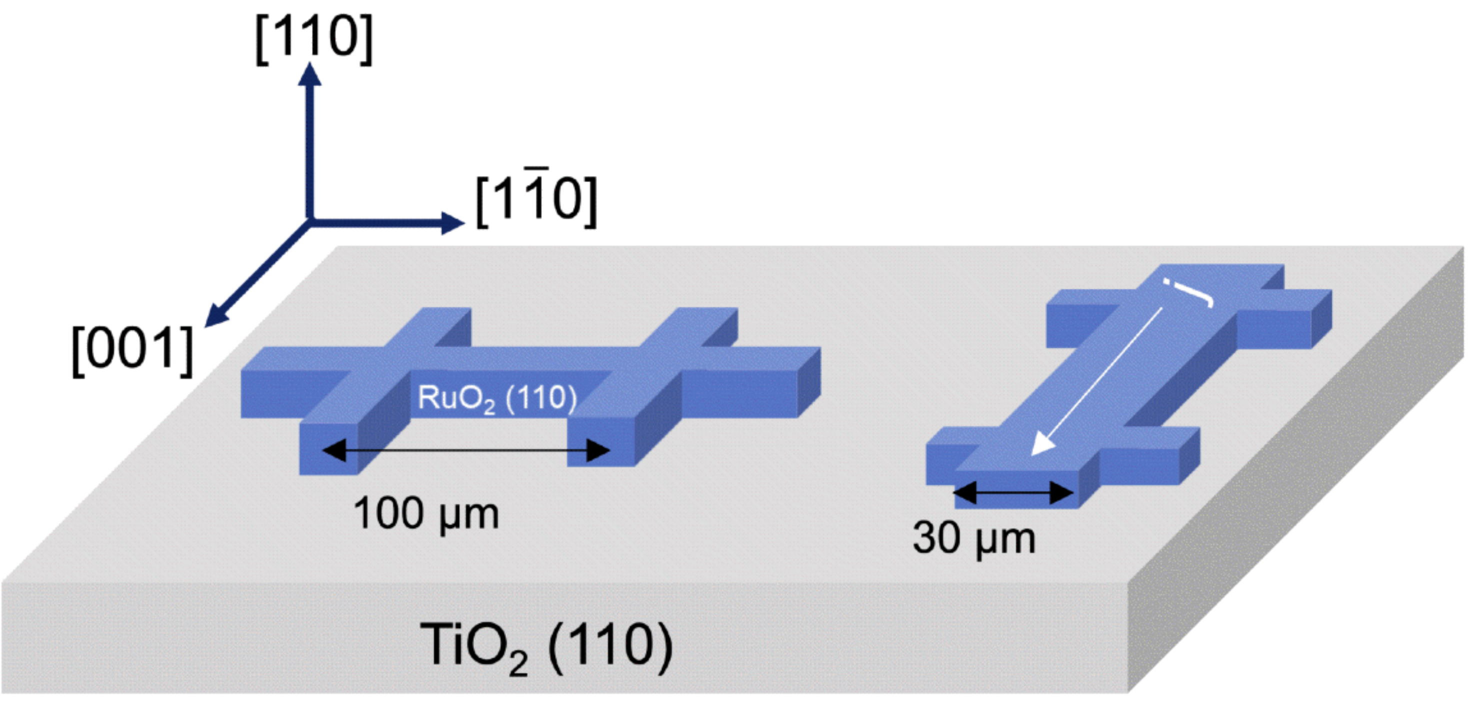

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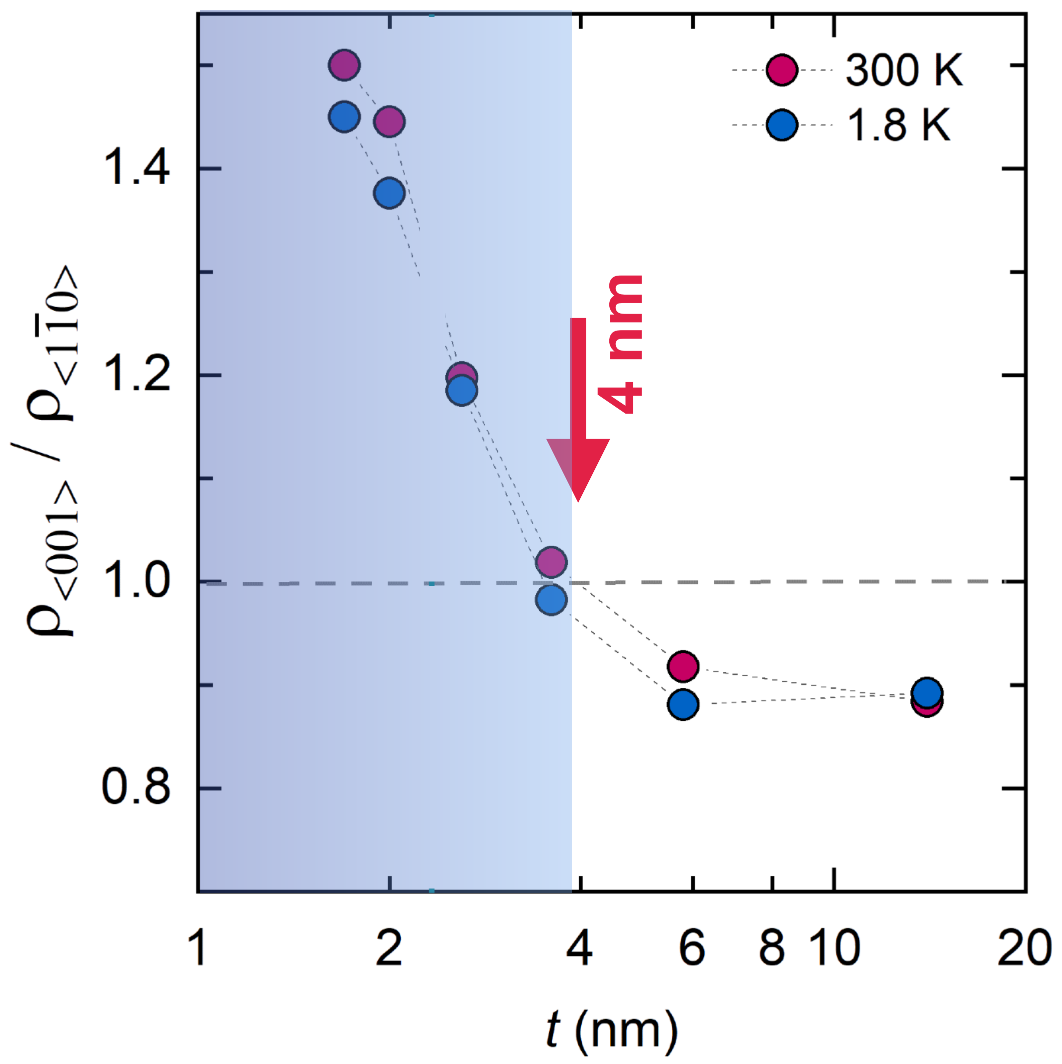
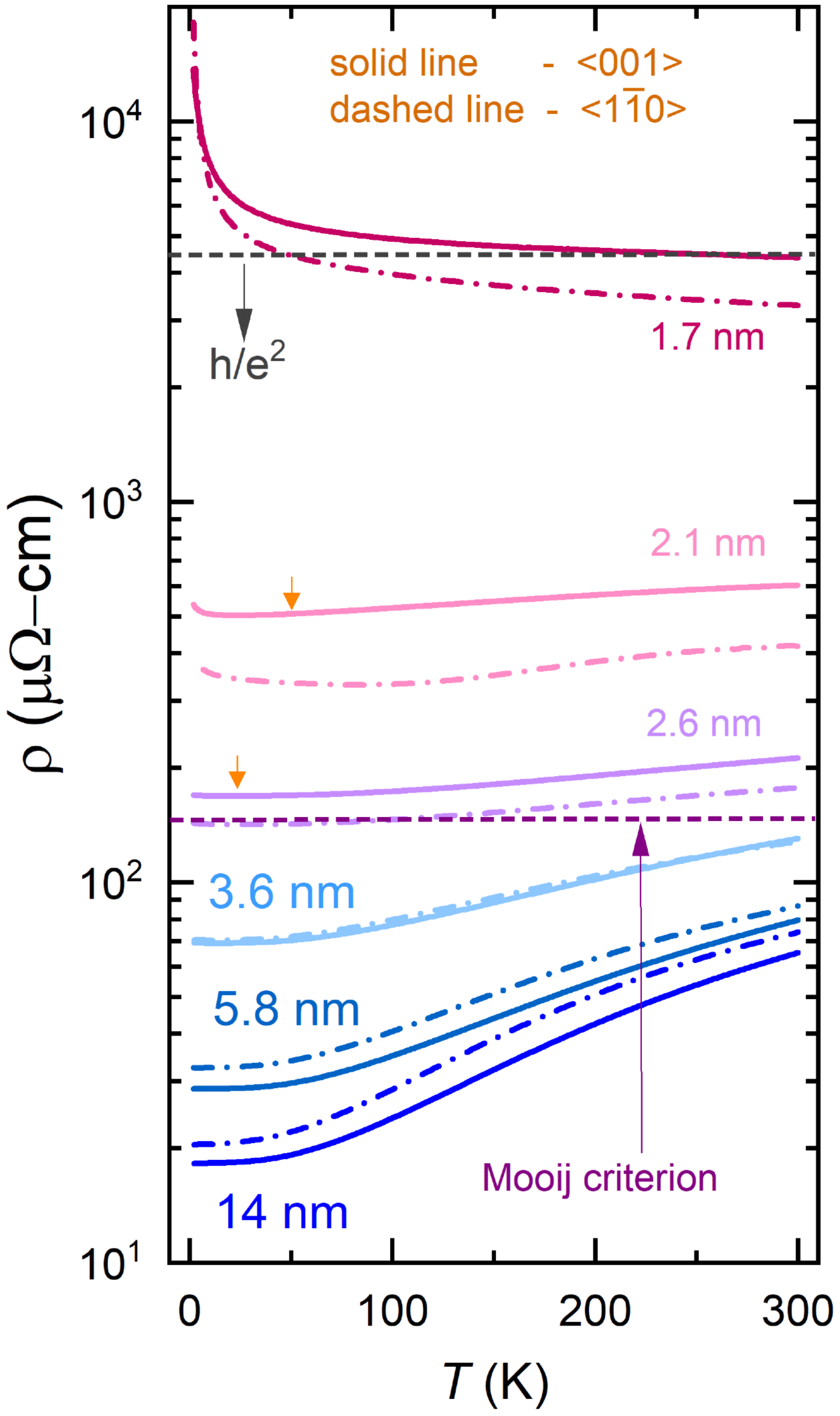


Metal-to-insulator transition as t_{film} ↓



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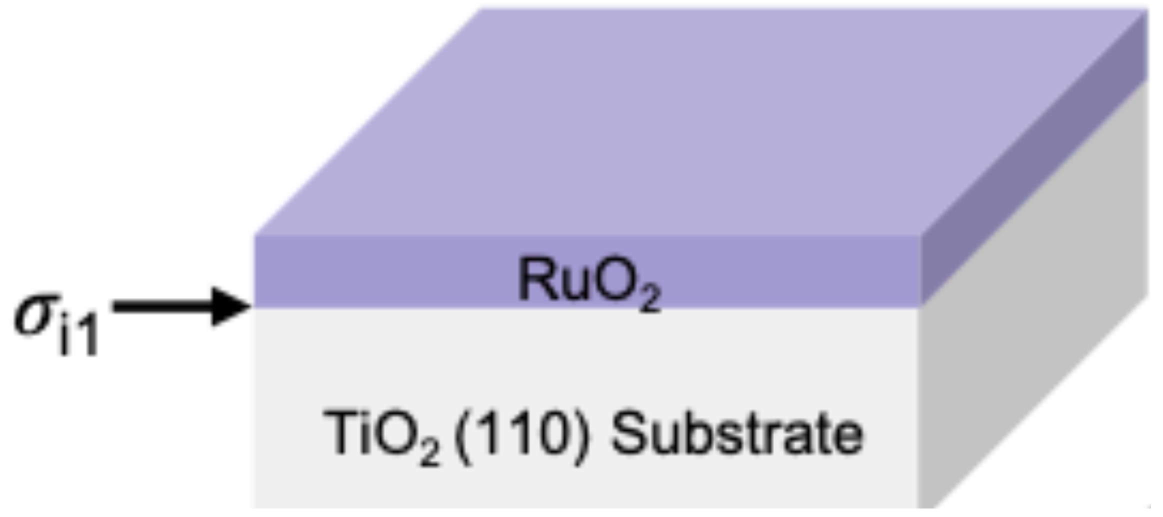
A. K. Rajapitamahuni, S. Nair, Z. Yang, A. K. Manjeshwar, S. G. Jeong, W. Nunn, and B. Jalan, Phys. Rev. Mater. 8, 075002 (2024)

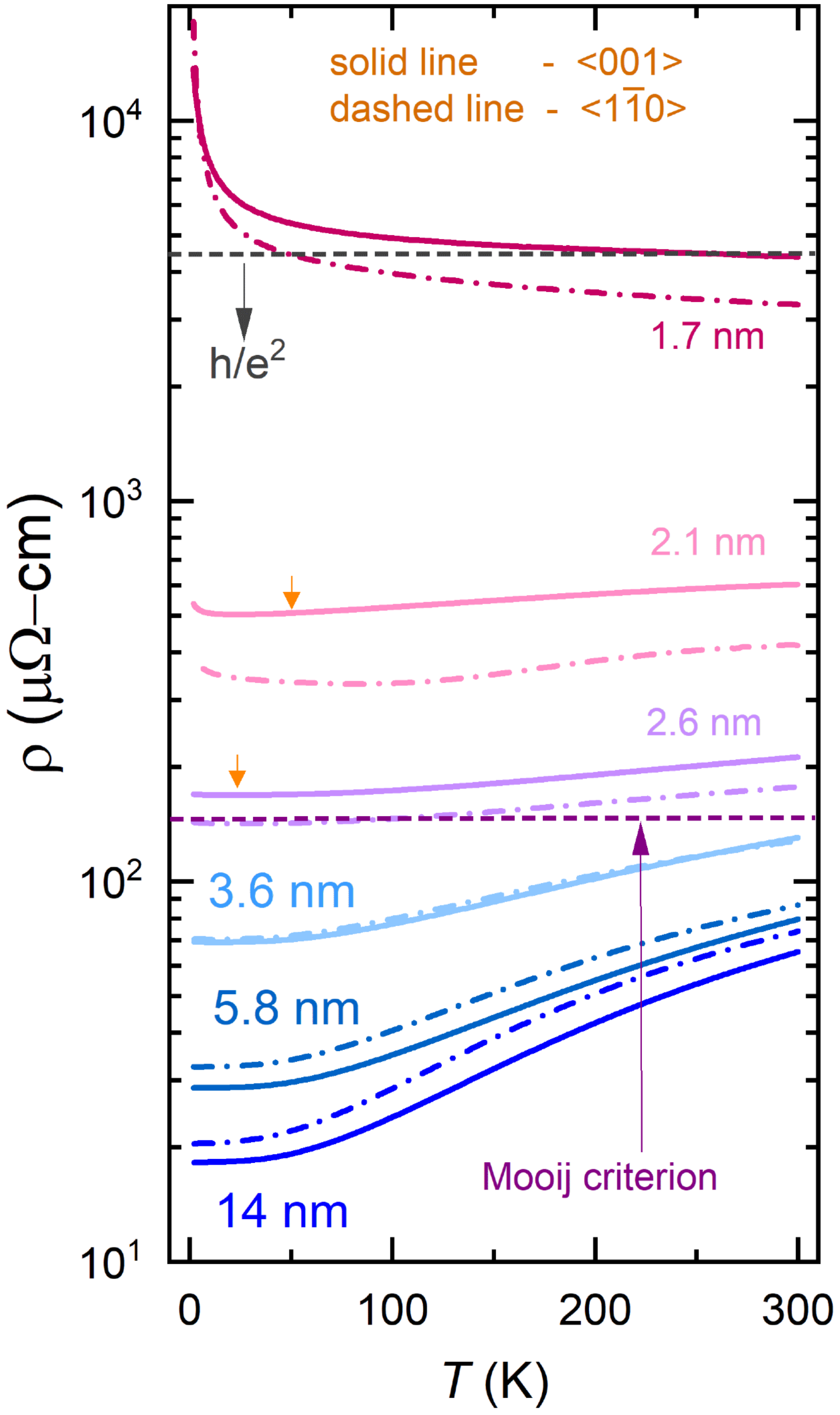
Origin of thickness-dependent M→ I Transition

Interfacial compensation as the origin of Metal-to-insulator transition as t_{film} ↓



$$\sigma_{2D} = (t_{RuO_2} - t_{i1})\cdot\sigma_b + t_{i1}\cdot\sigma_{i1}$$
$$= t_{RuO_2}\cdot\sigma_b - t_{i1}\cdot(\sigma_b - \sigma_{i1})$$

Metal-to-insulator transition as t_{film} ↓



Origin of thickness-dependent M→ I Transition

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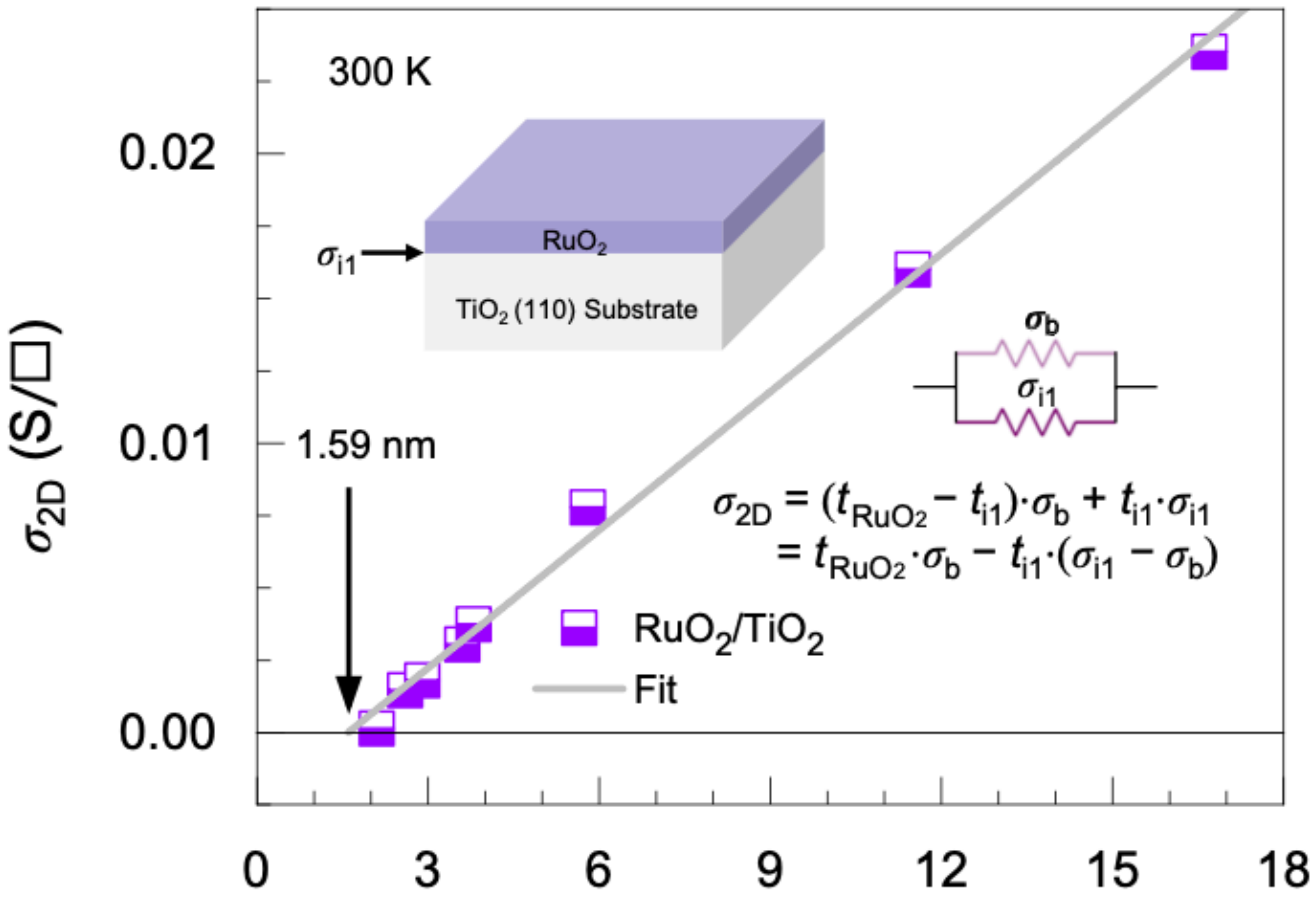
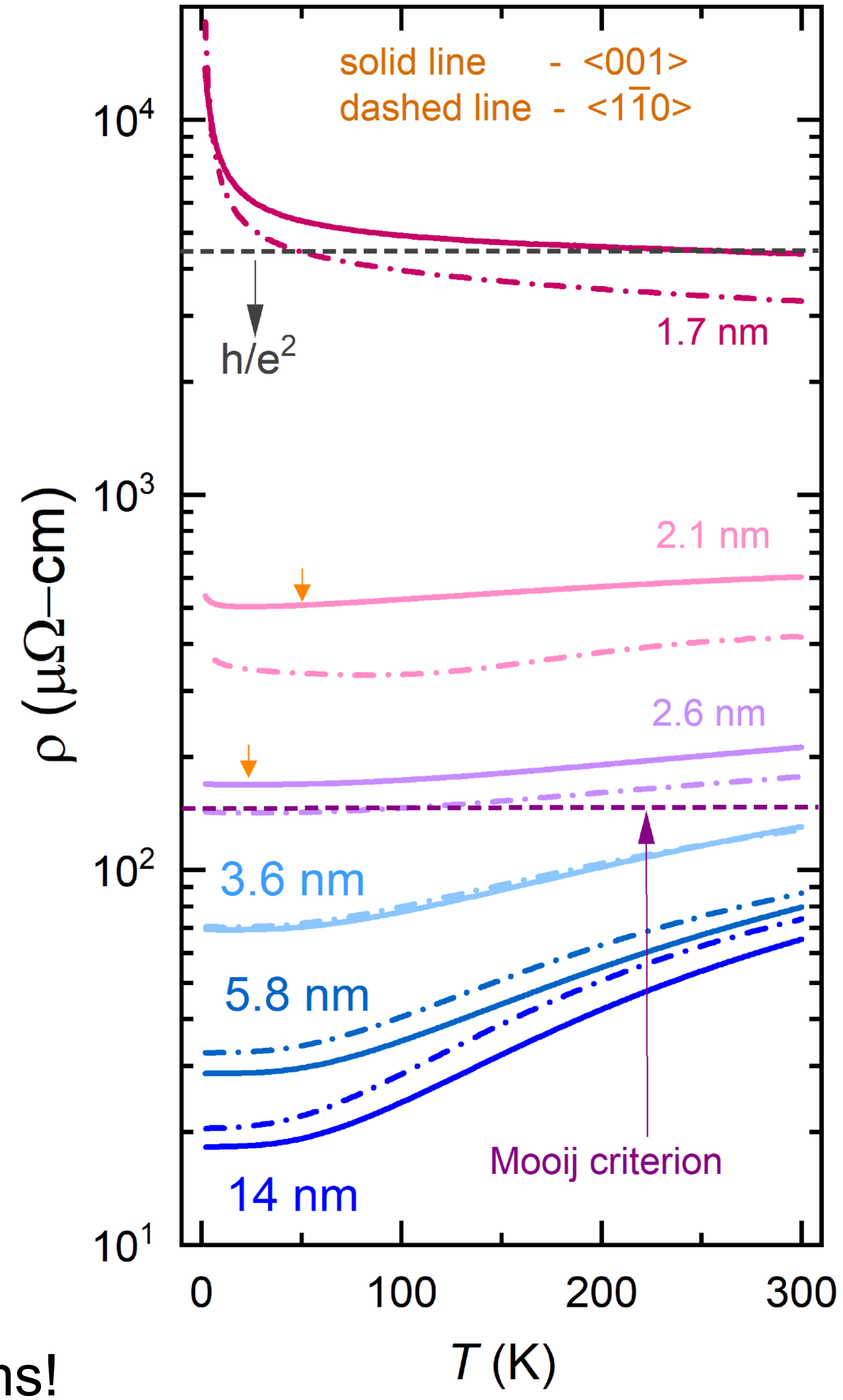
Interfacial compensation as the origin of Metal-to-insulator transition as t_{film} ↓



Evidence of an interfacial charge compensation layer between TiO_2 and RuO_2 films!

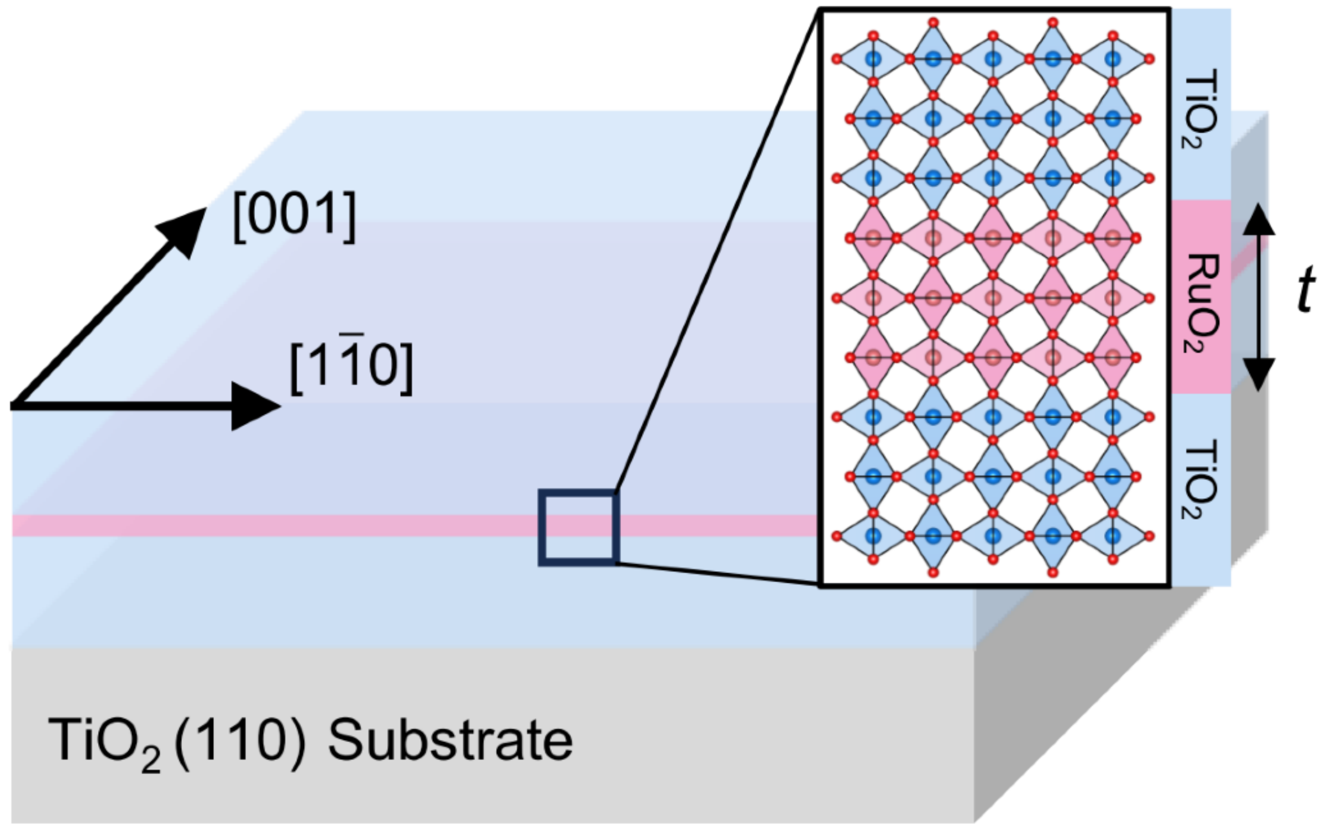
But, what about the surface effect such as contamination?

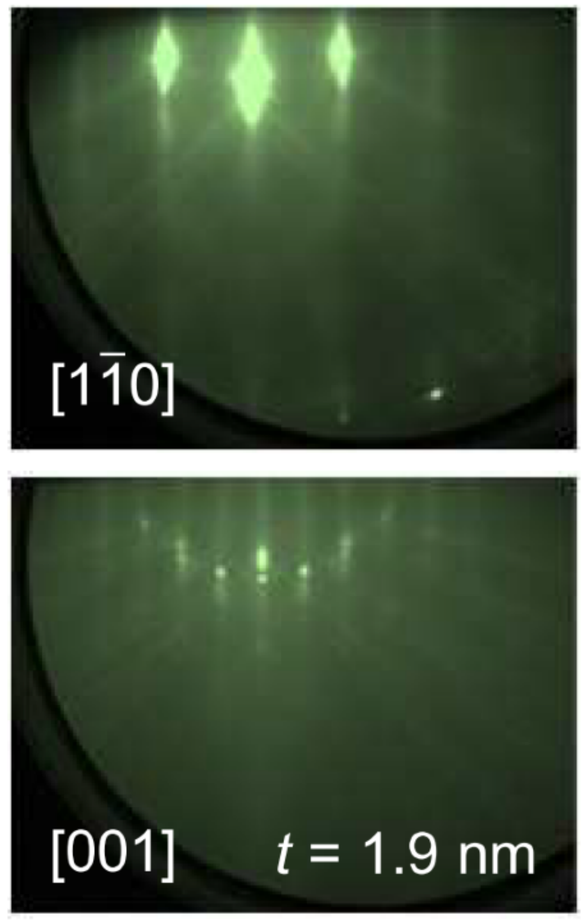
Metal-to-insulator transition as t_{film} ↓

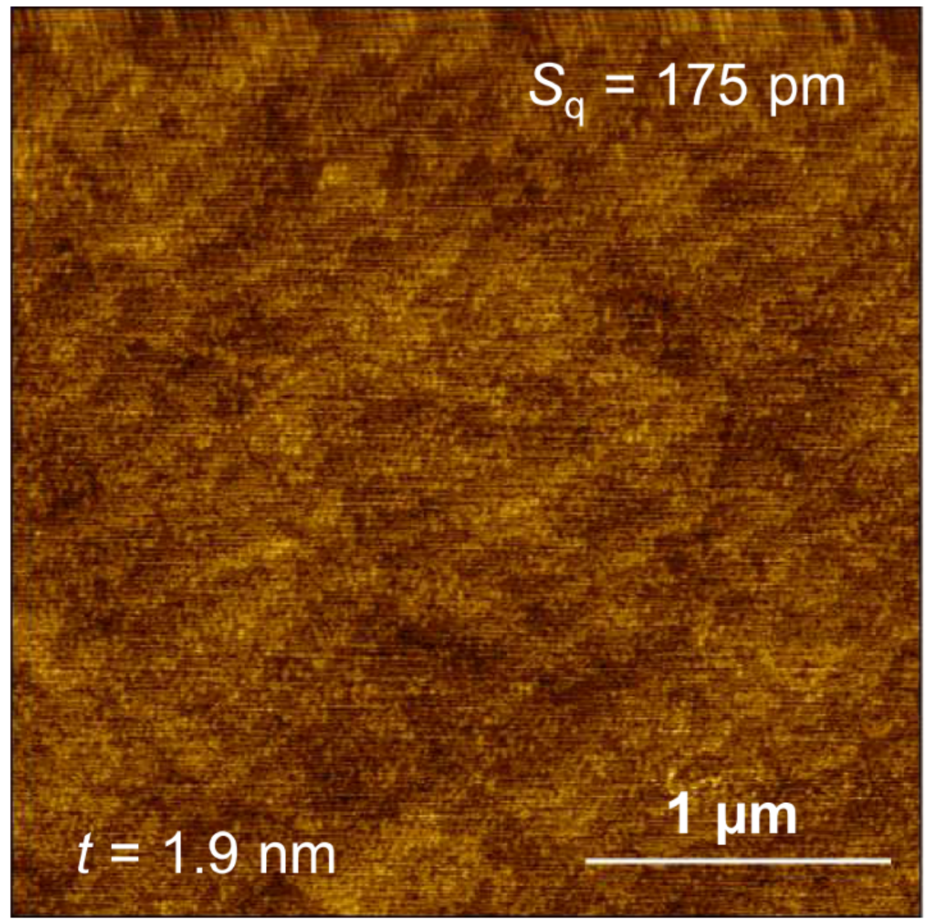


S. G. Jeong, ... J. M. LeBeau, and B. Jalan, Nat. Commun. (accepted) (2026)

Origin of thickness-dependent M→ I Transition







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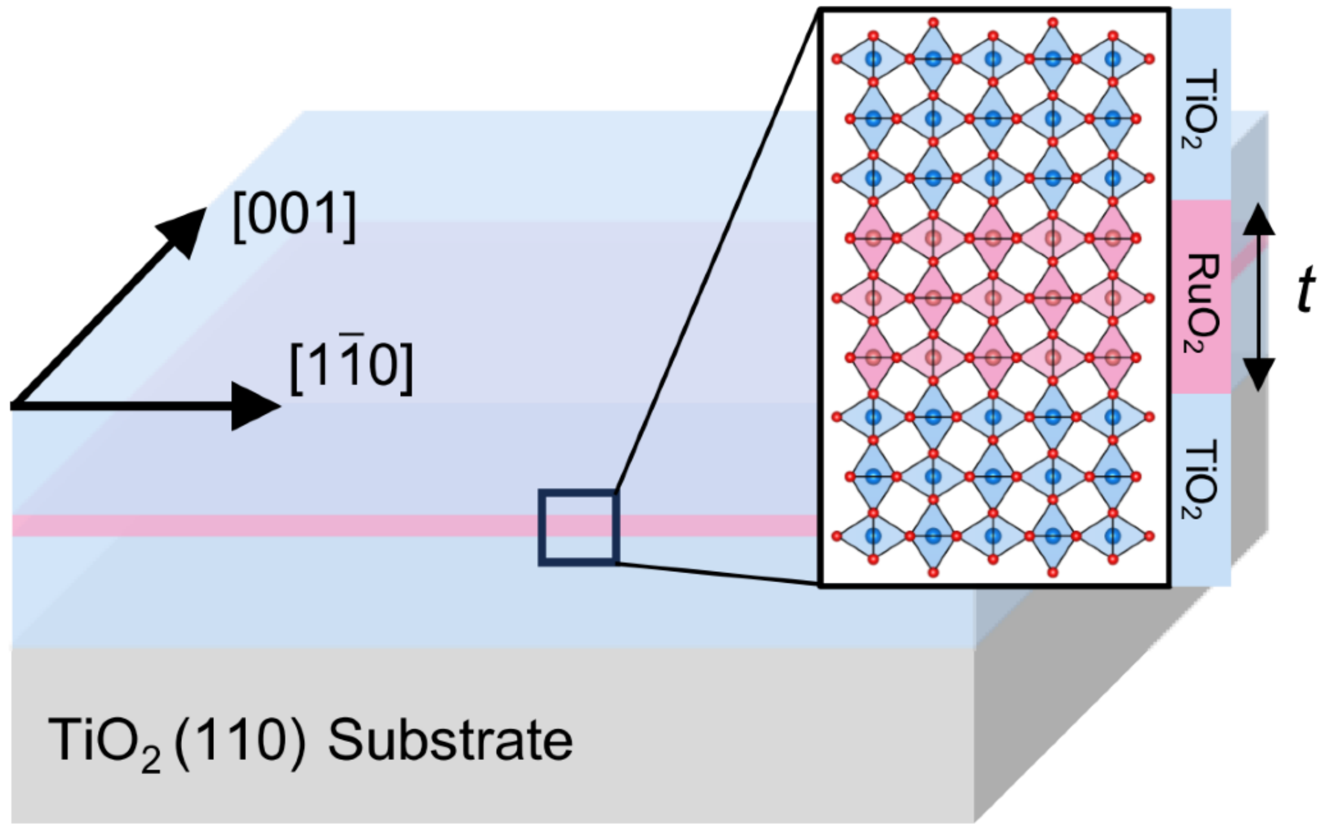

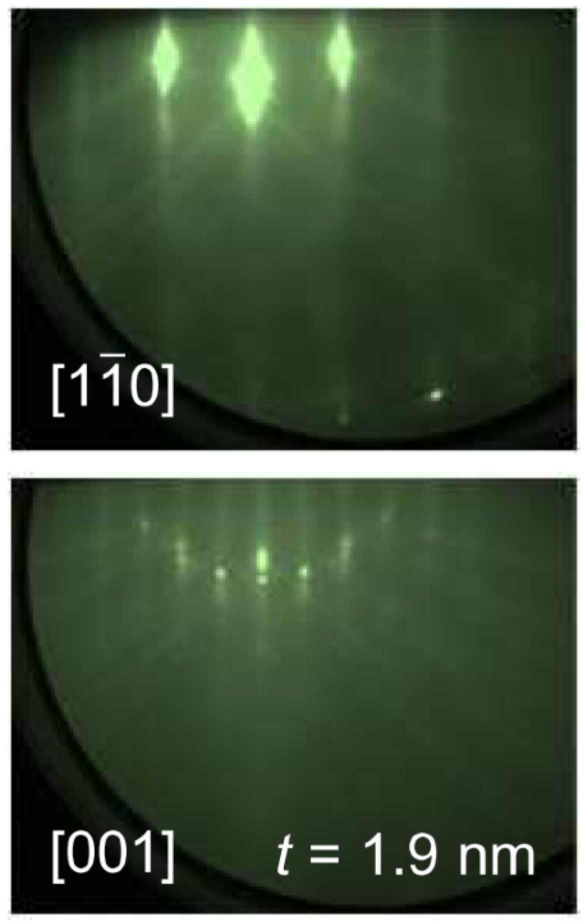

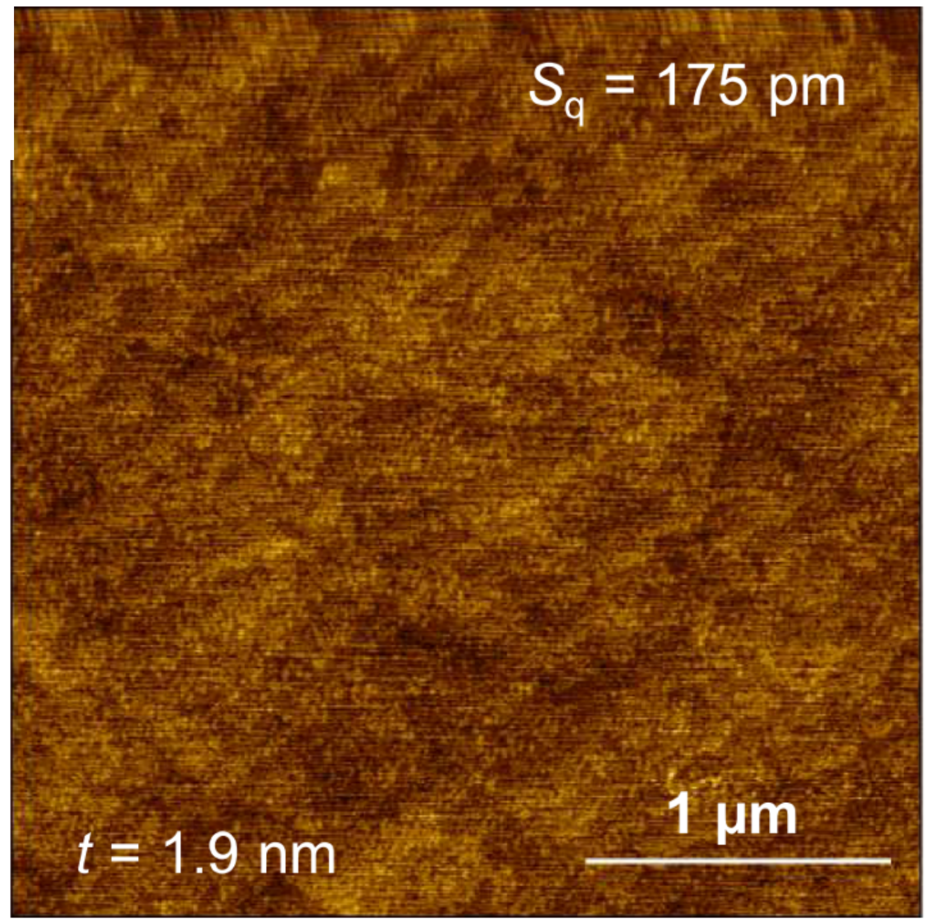

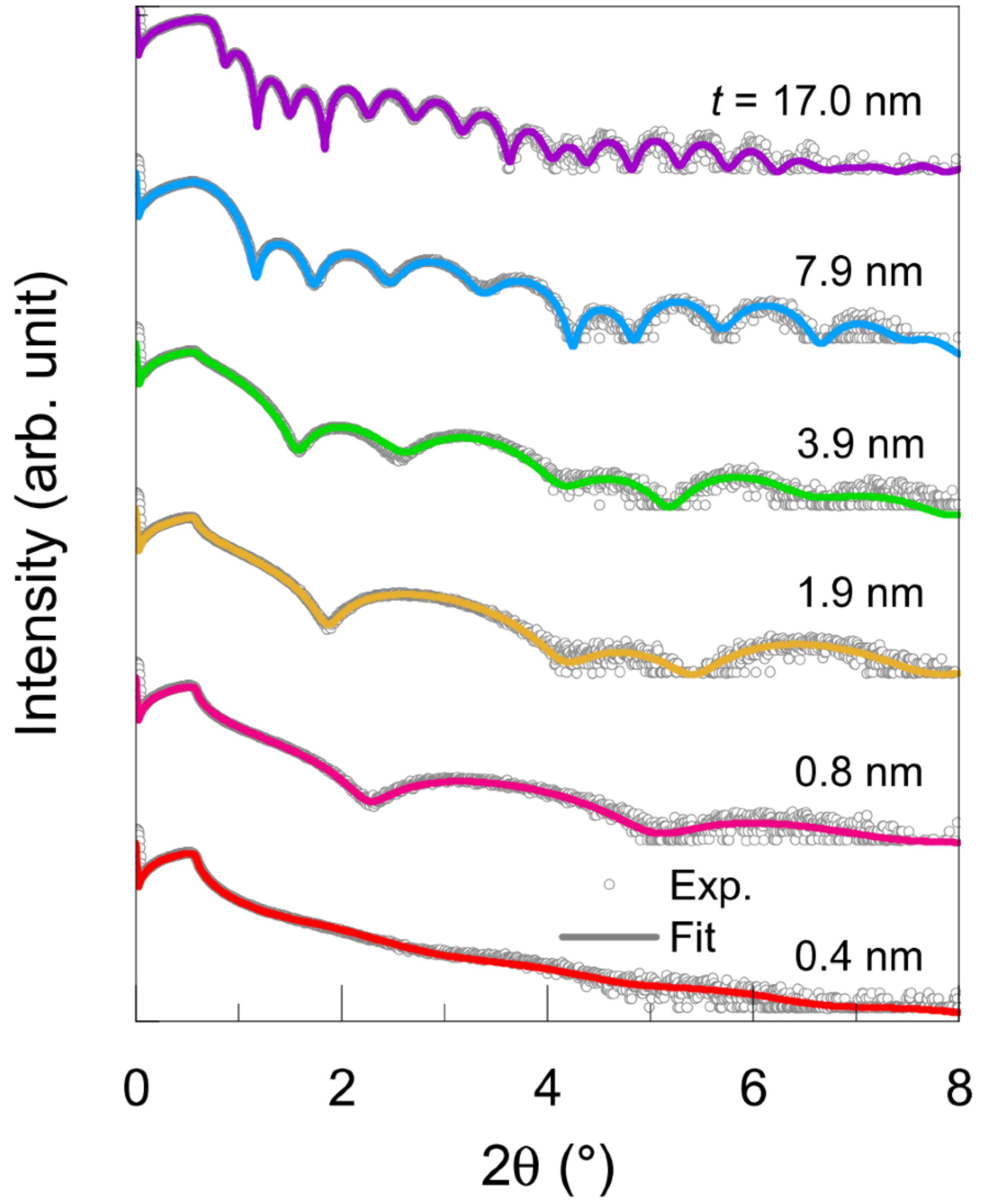

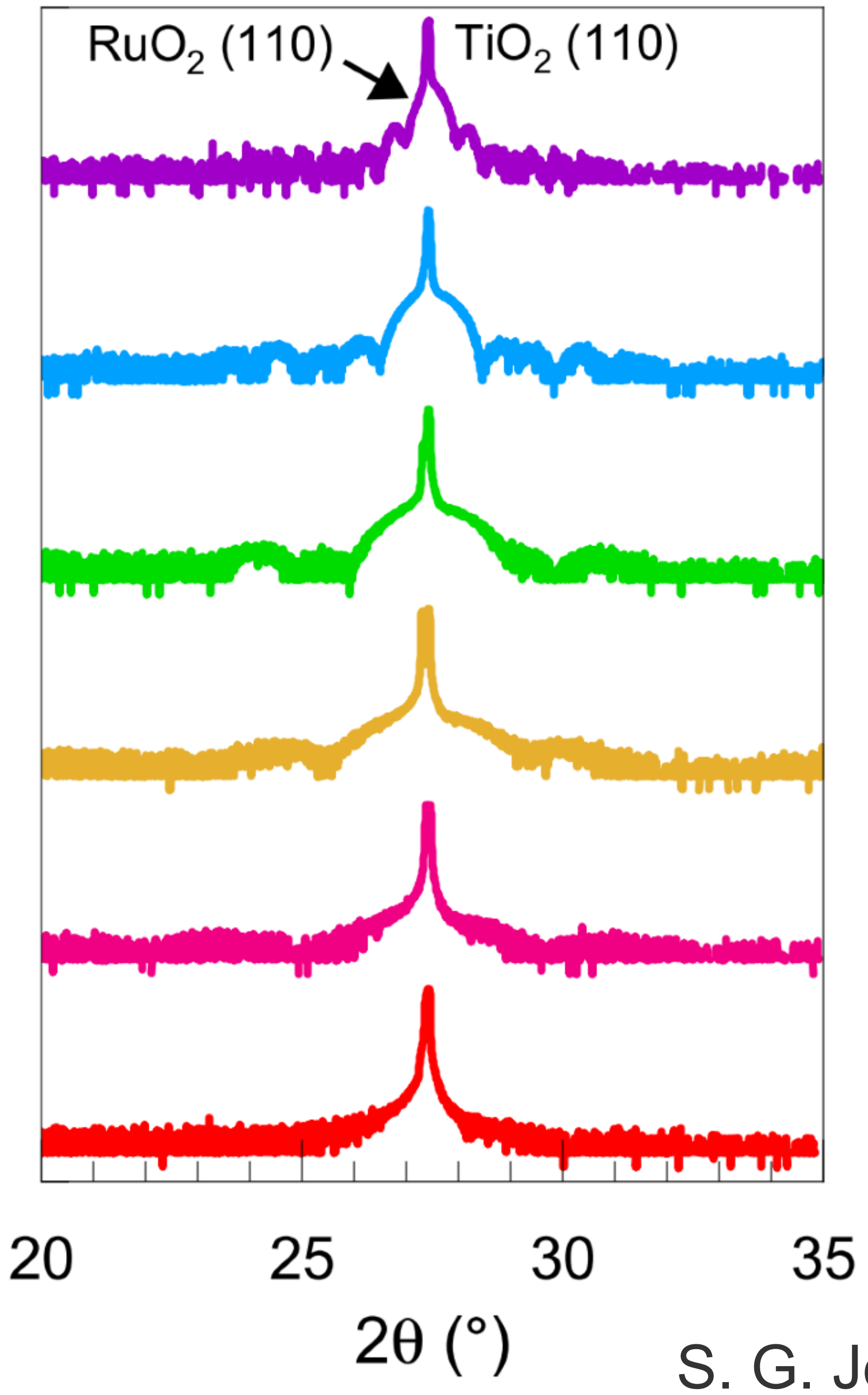

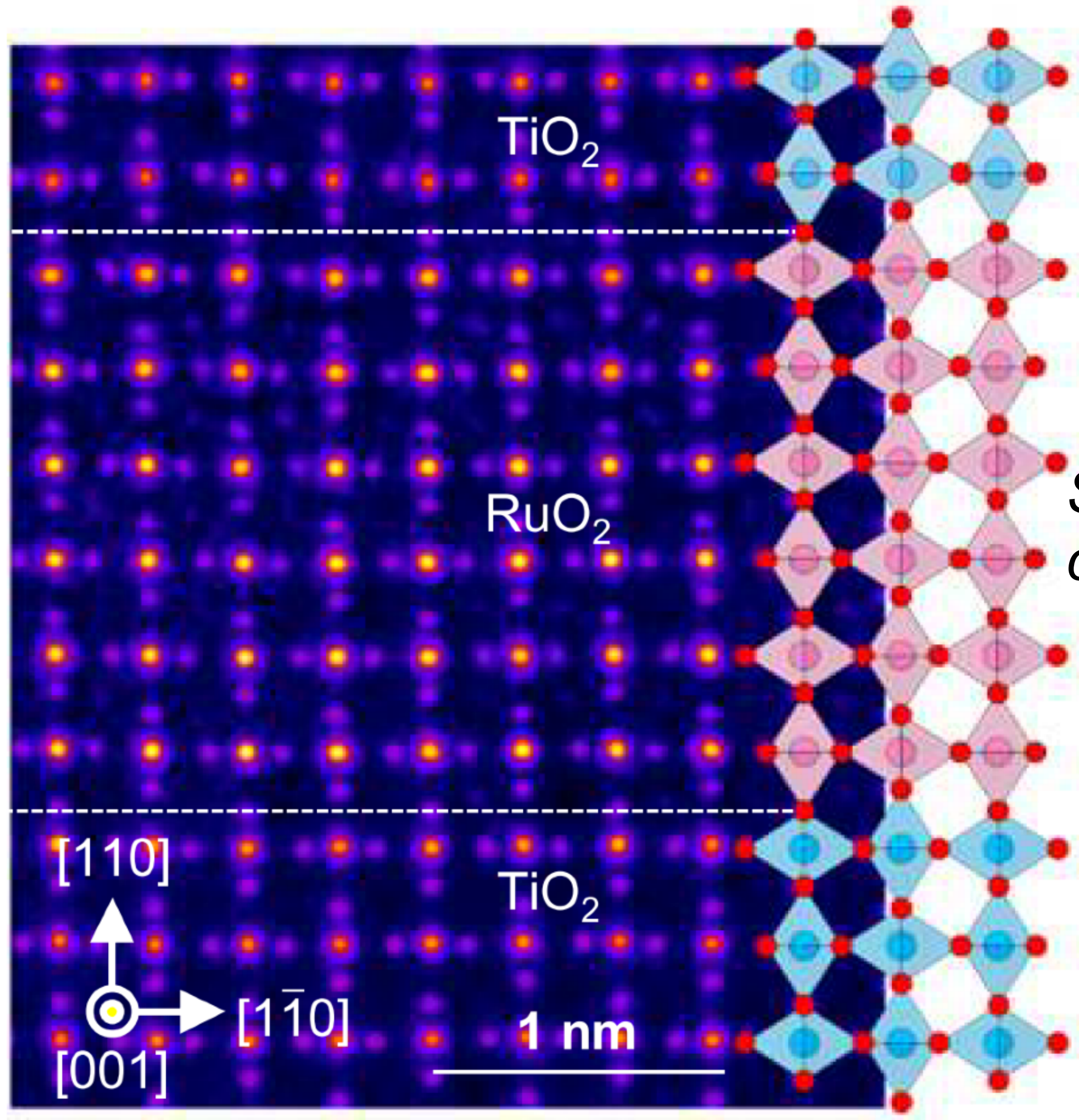

STEM Ptychography In collaboration with J. Lebeau, MIT

S. G. Jeong, …. J. M. LeBeau, and B. Jalan, Nat. Commun. (accepted) (2026)

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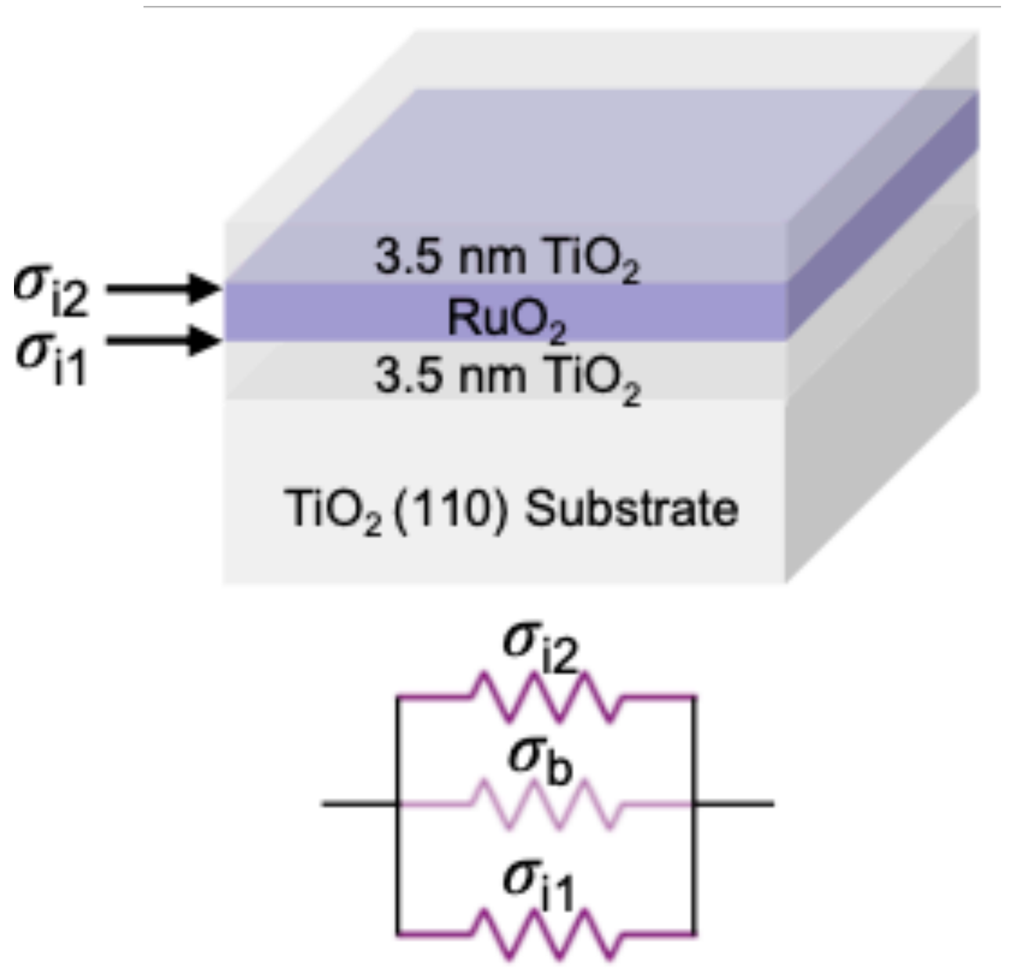

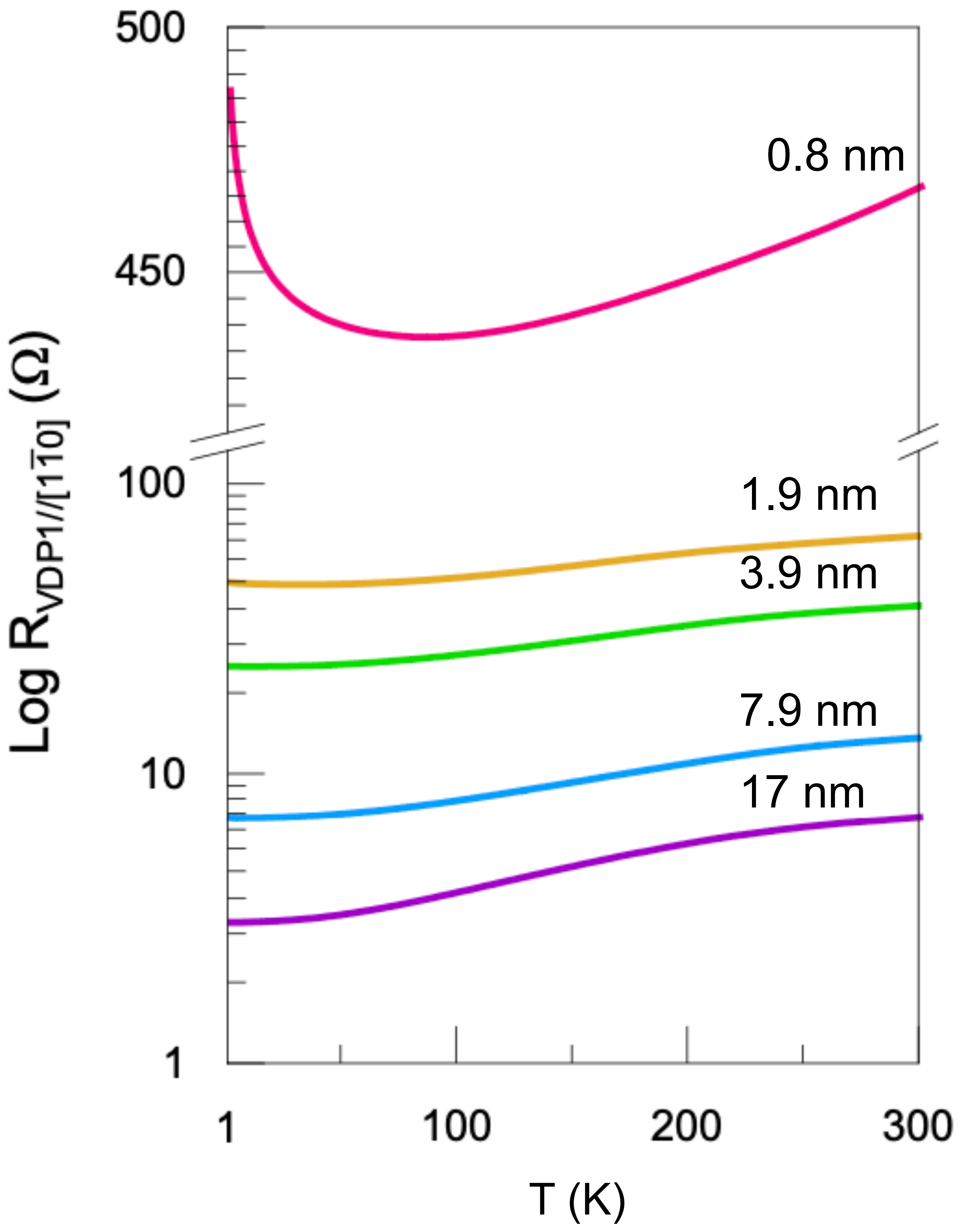

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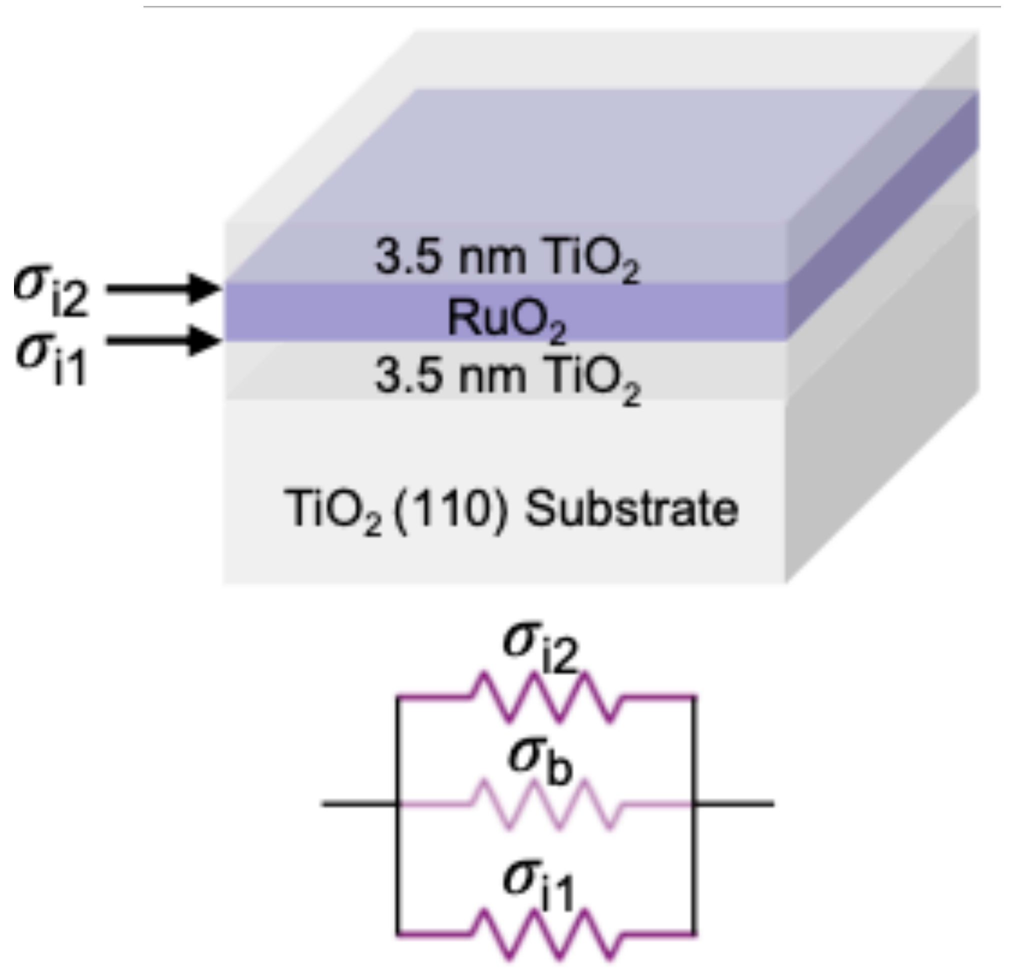
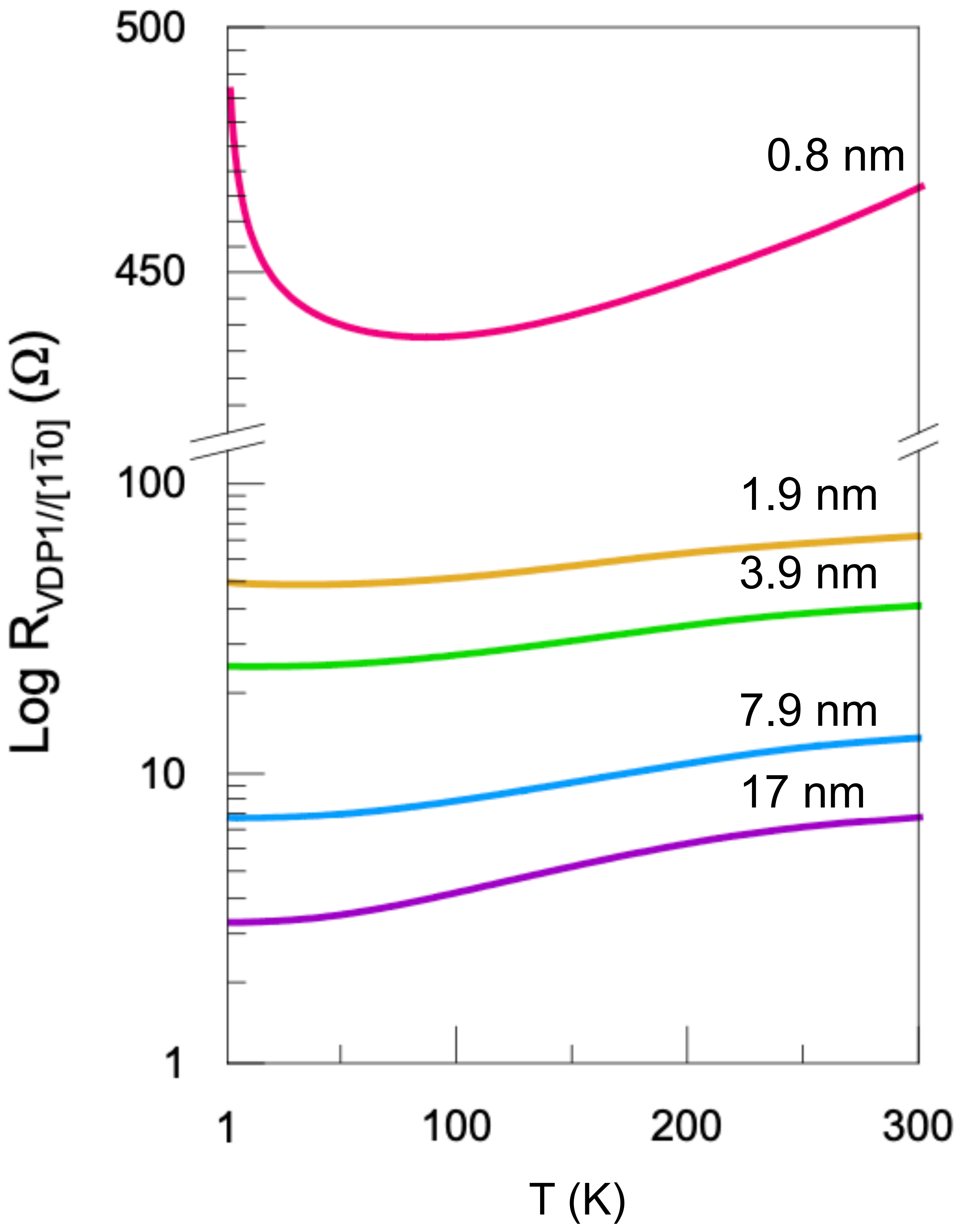


$$\sigma_{2D} = t_{RuO_2} \cdot \sigma_b - (t_{i1} + t_{i2}) \cdot \{(\sigma_{i1} + \sigma_{i2}) - \sigma_b\}$$



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Origin of thickness-dependent M→ I Transition

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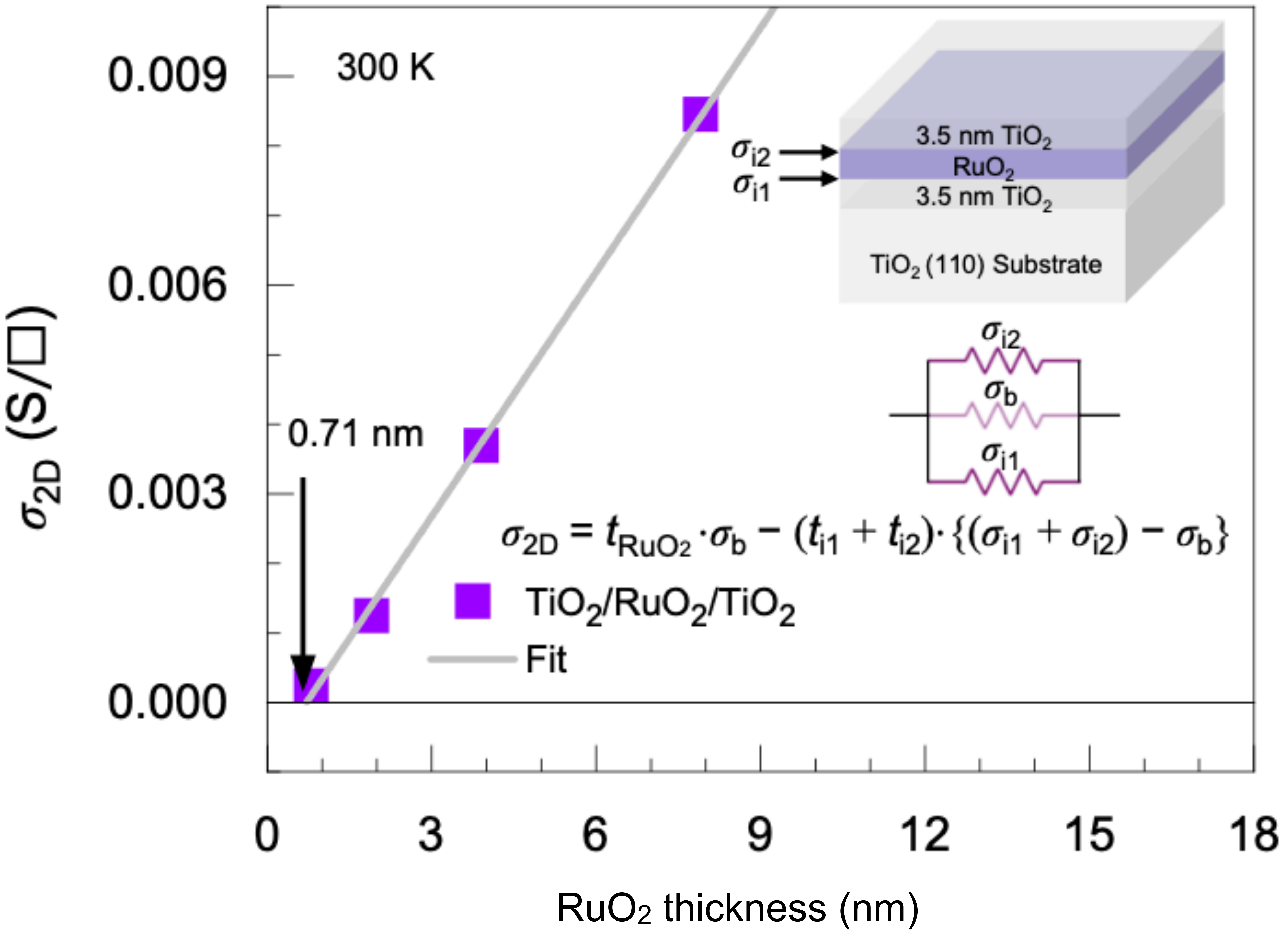

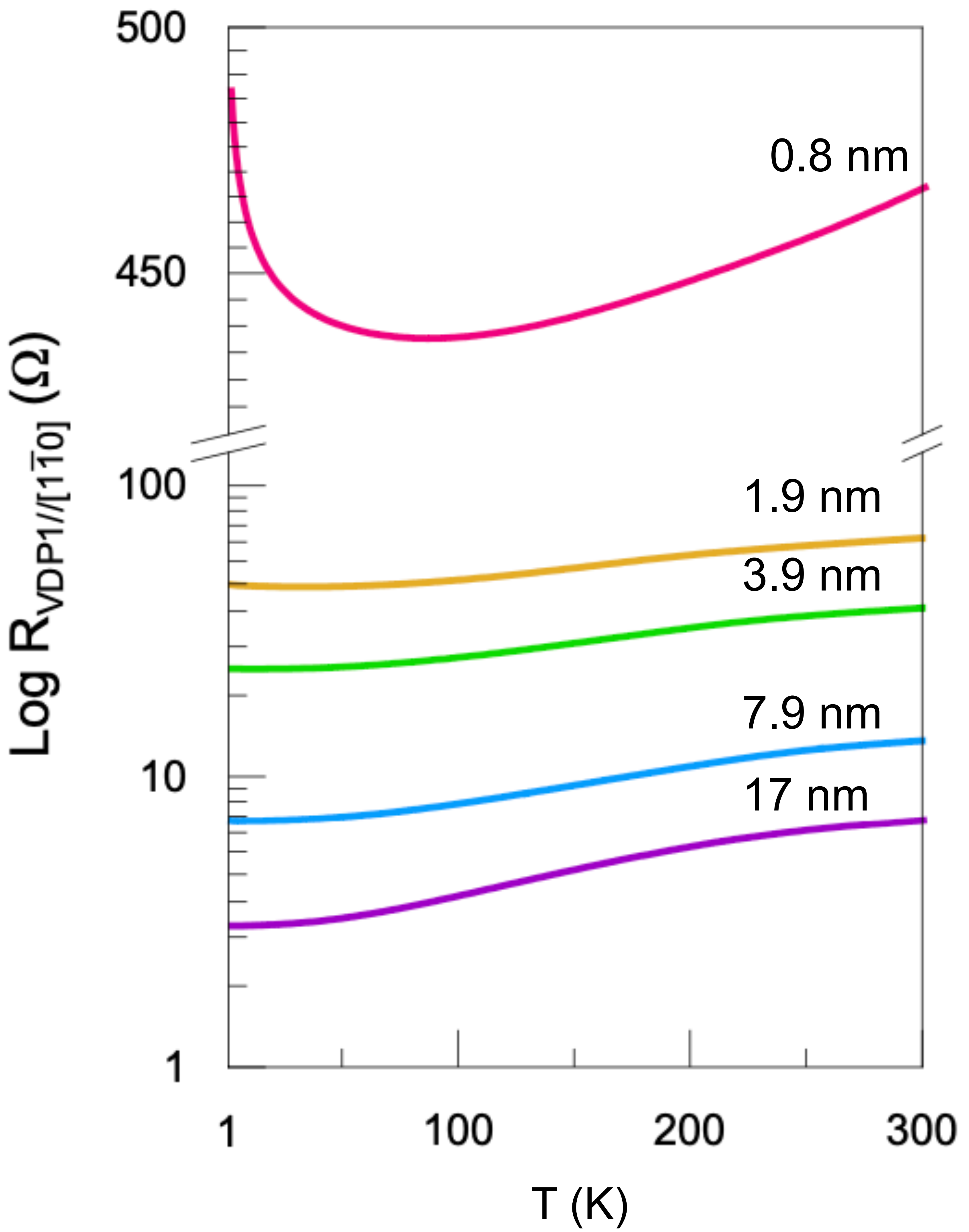

✓ Thickness-dependent M-I transition is associated with the interface!

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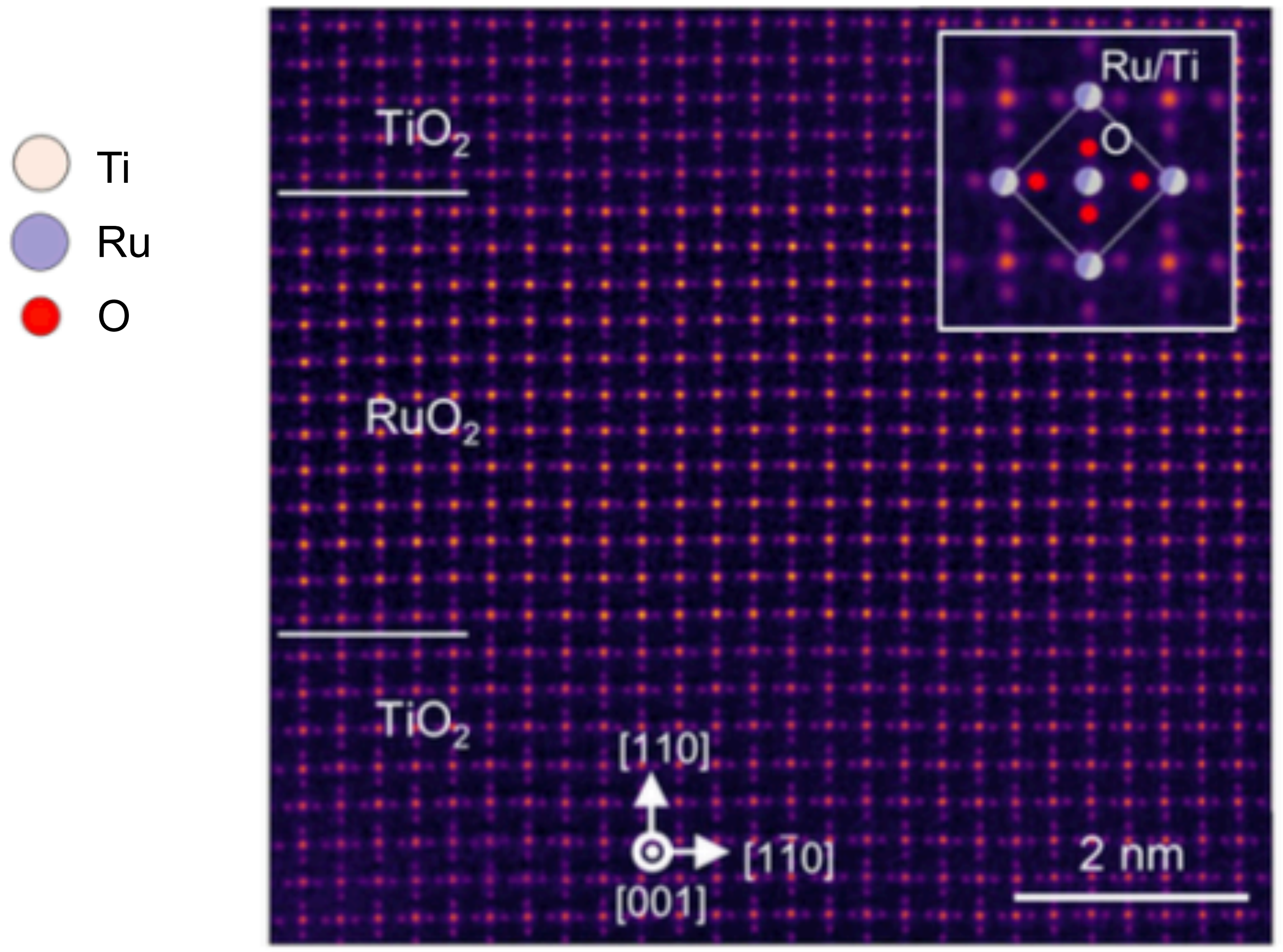
What is the Origin of Interfacial Dead Layer?



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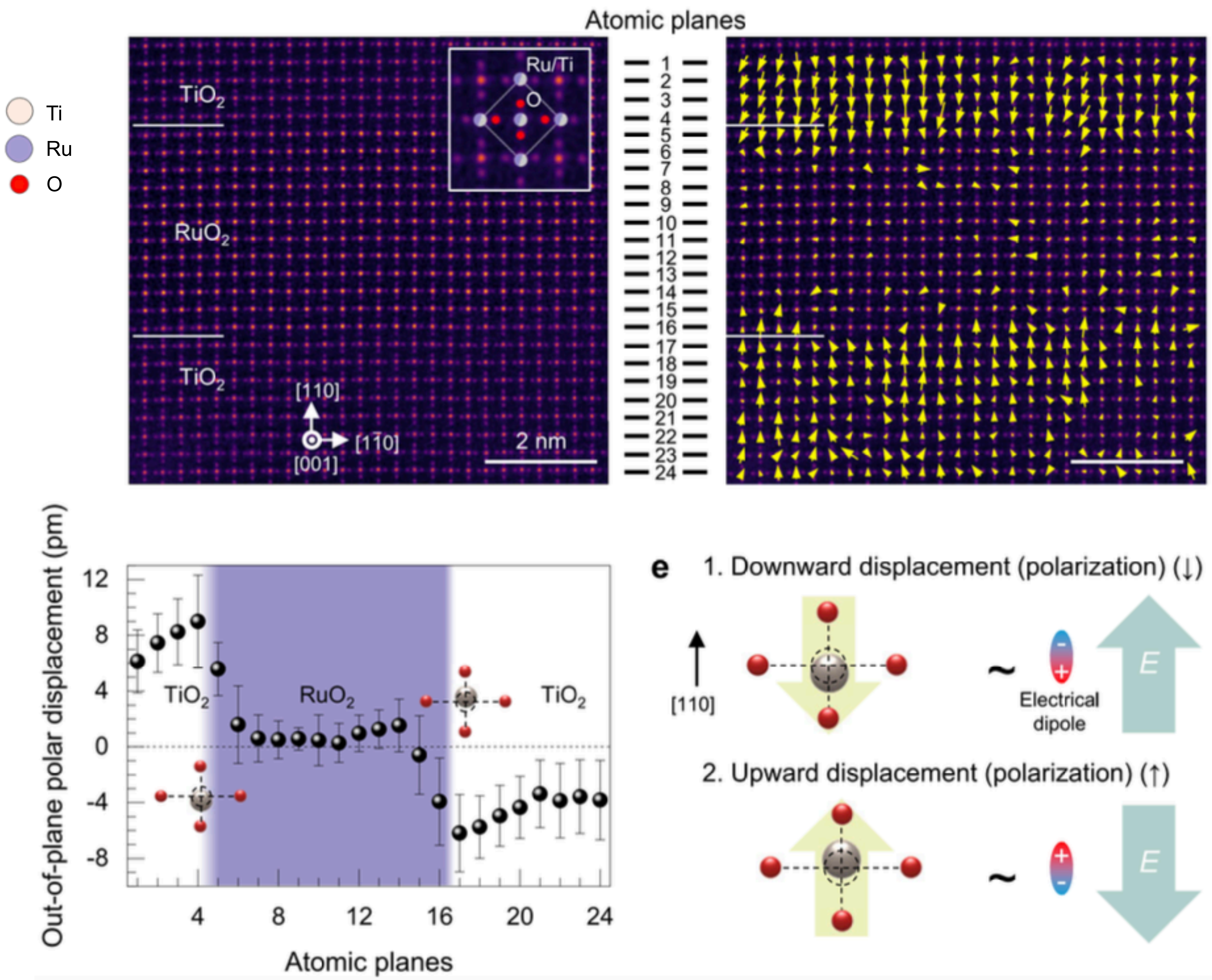

✓*Polarization at the interface between TiO_2/RuO_2 - likely due to the difference in rigidity and large epitaxial strain*

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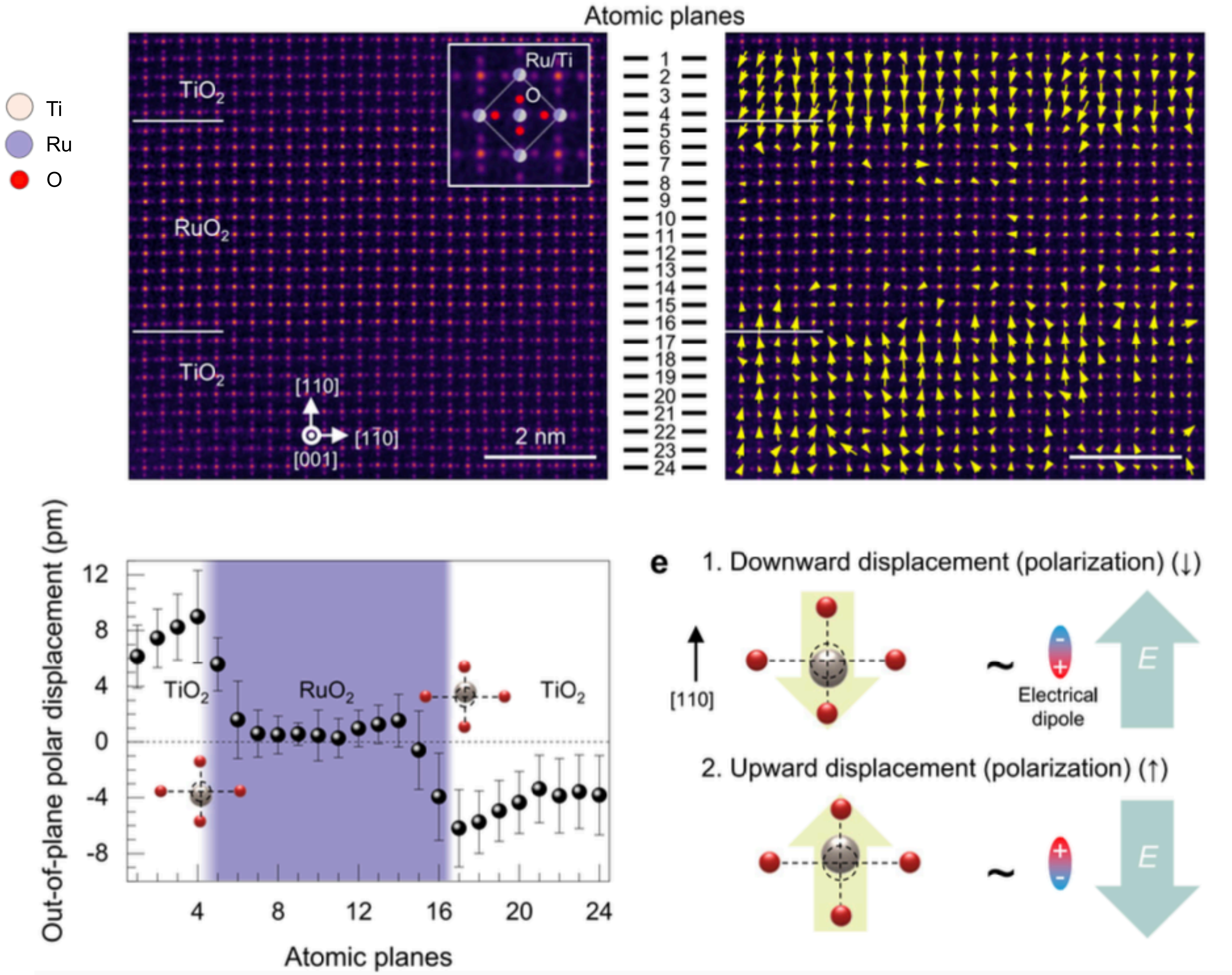

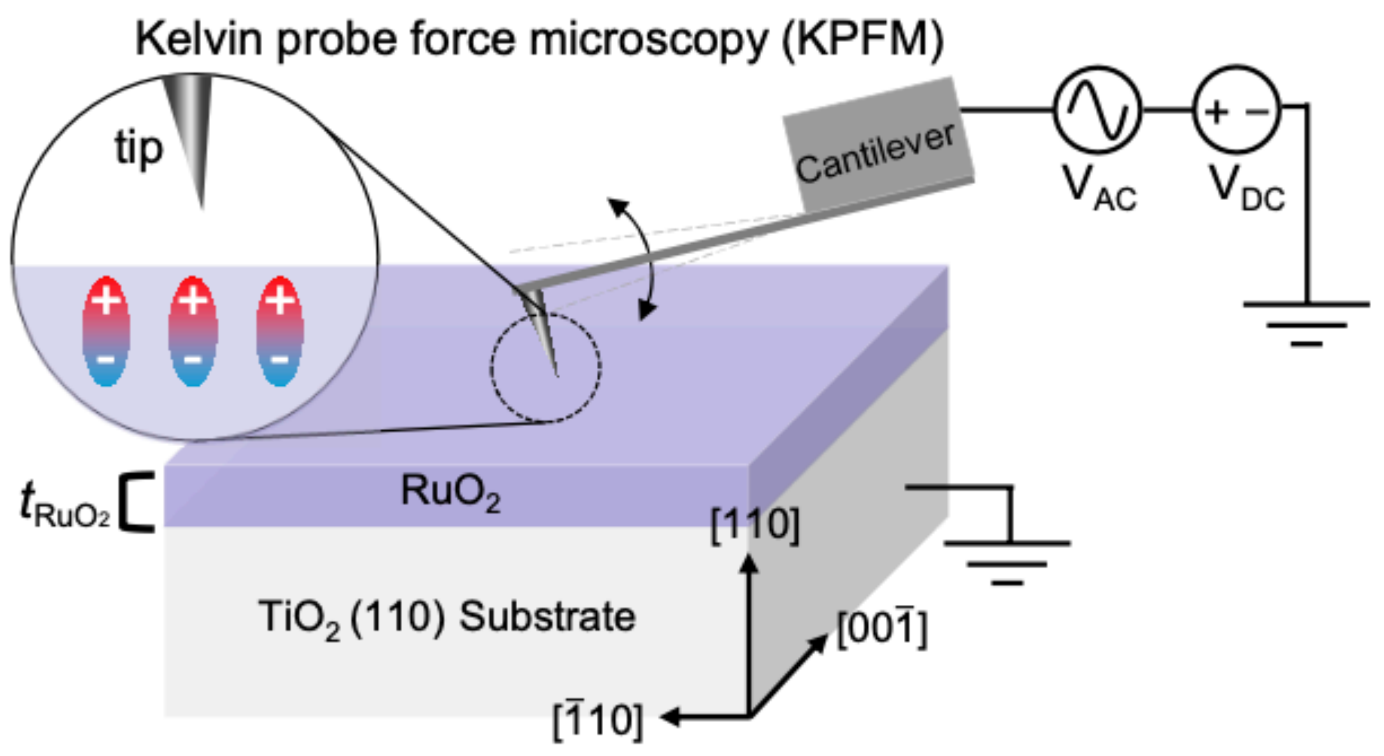

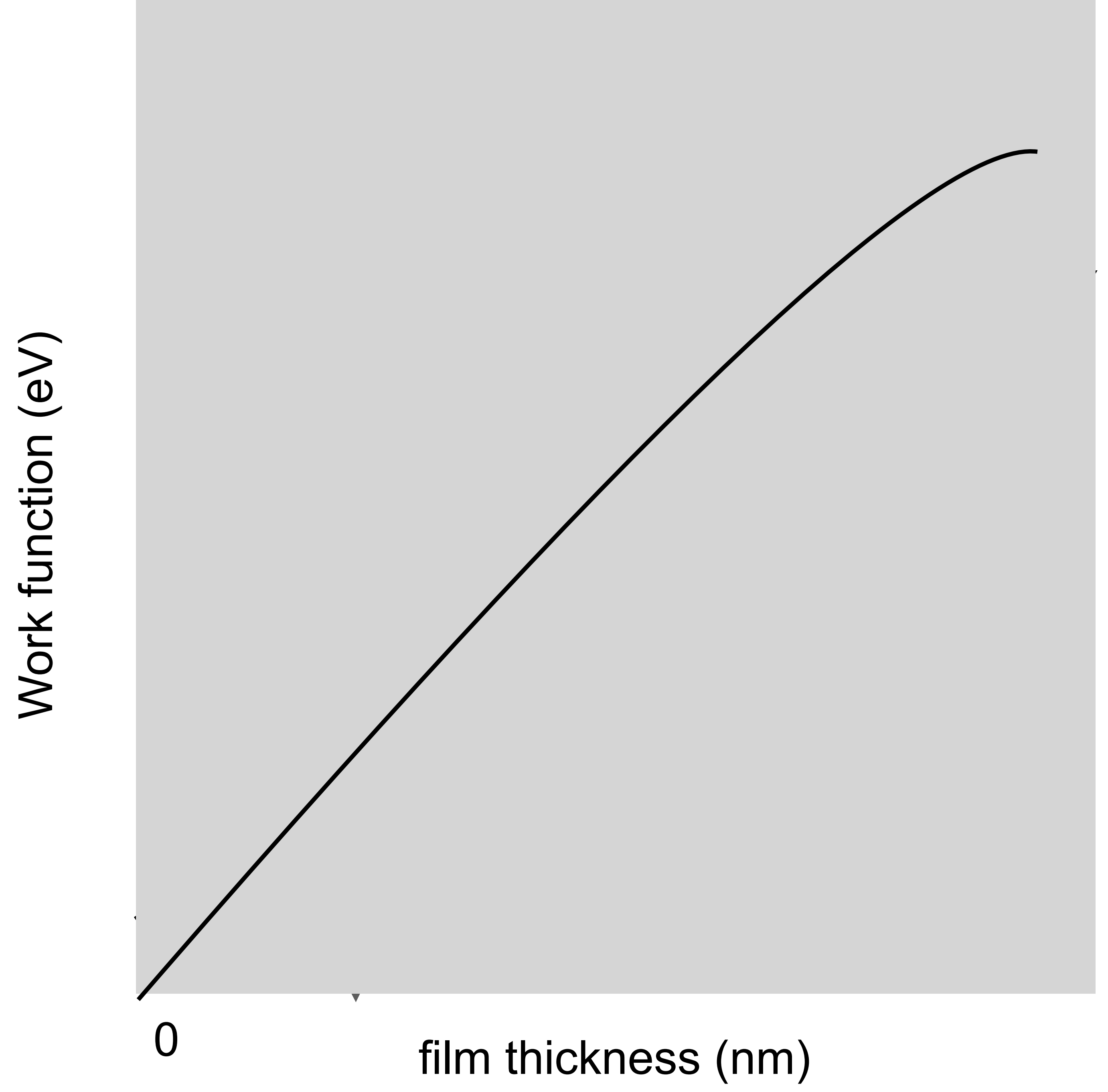

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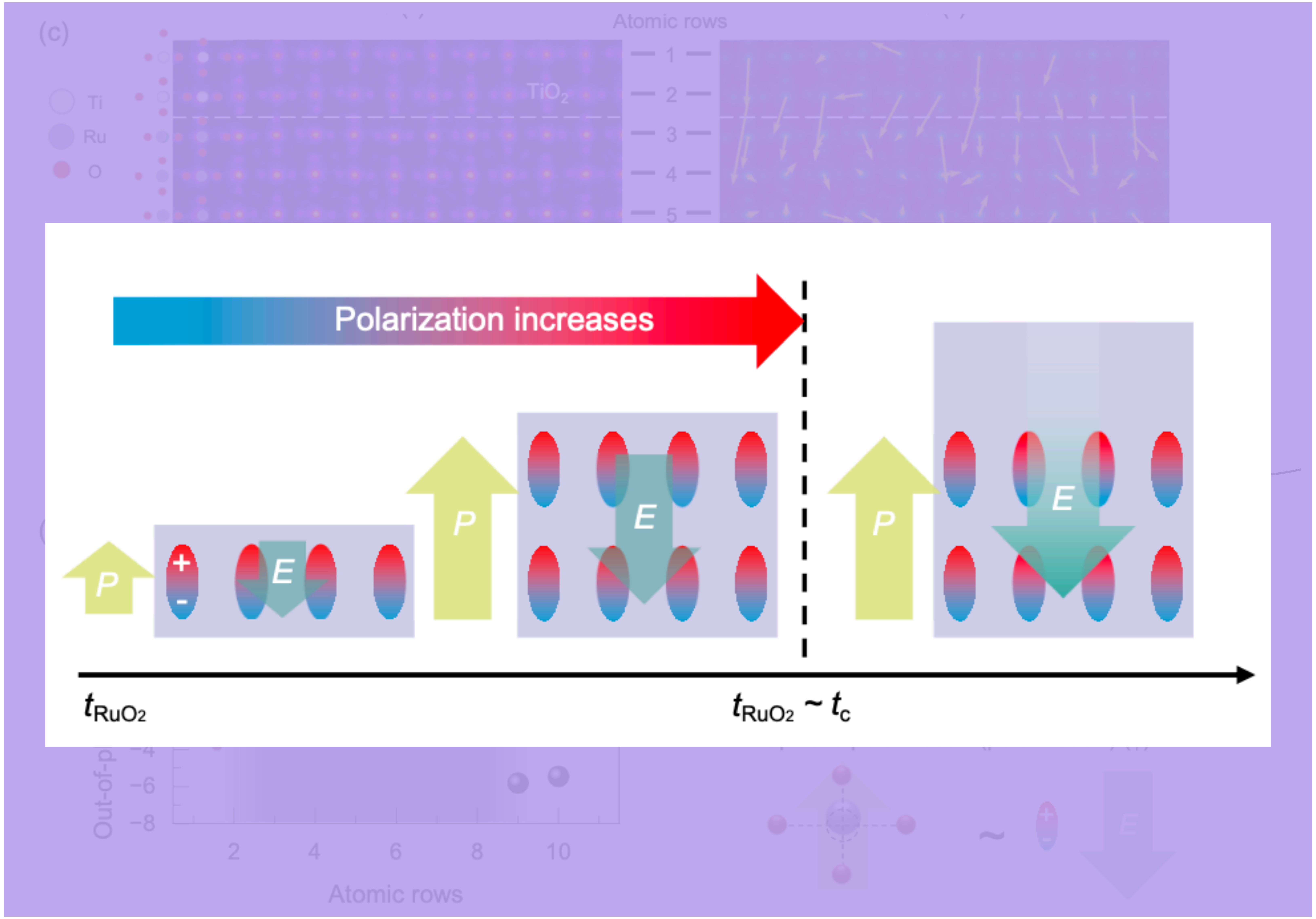

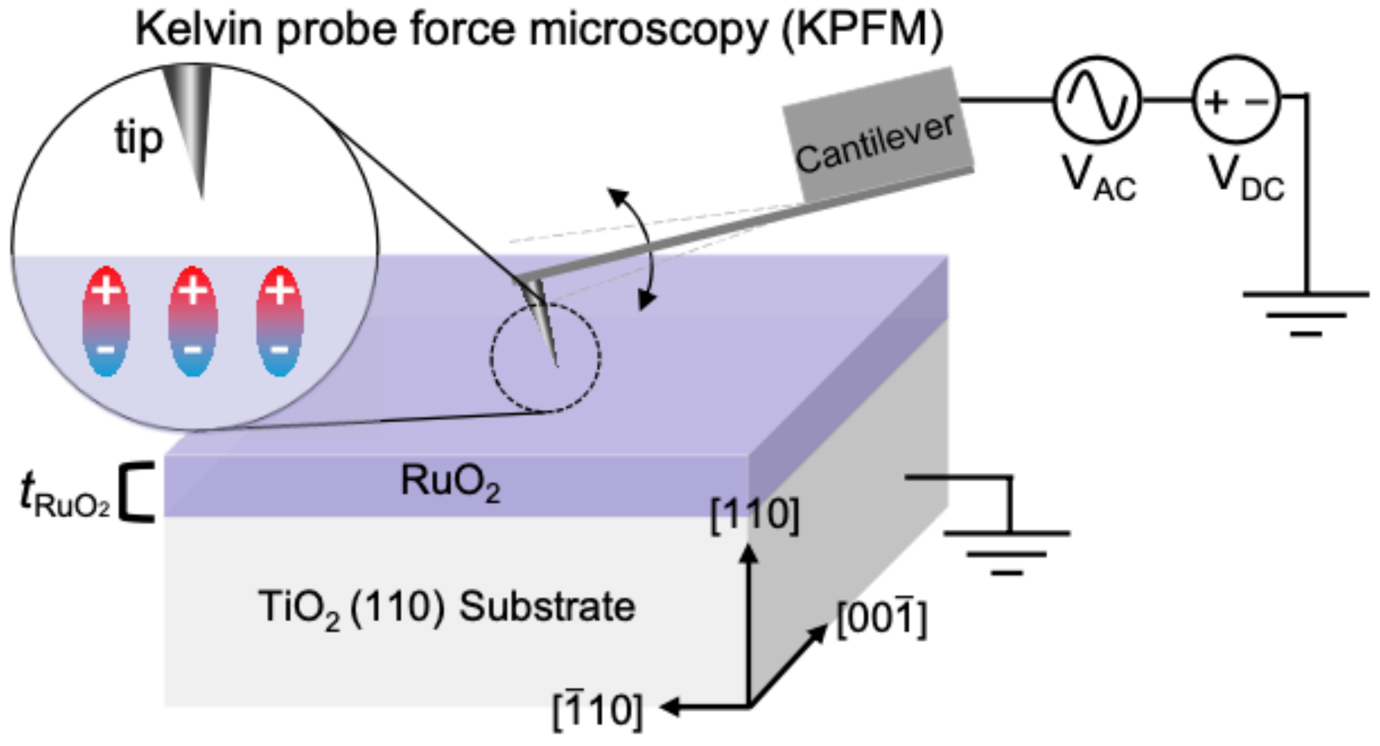

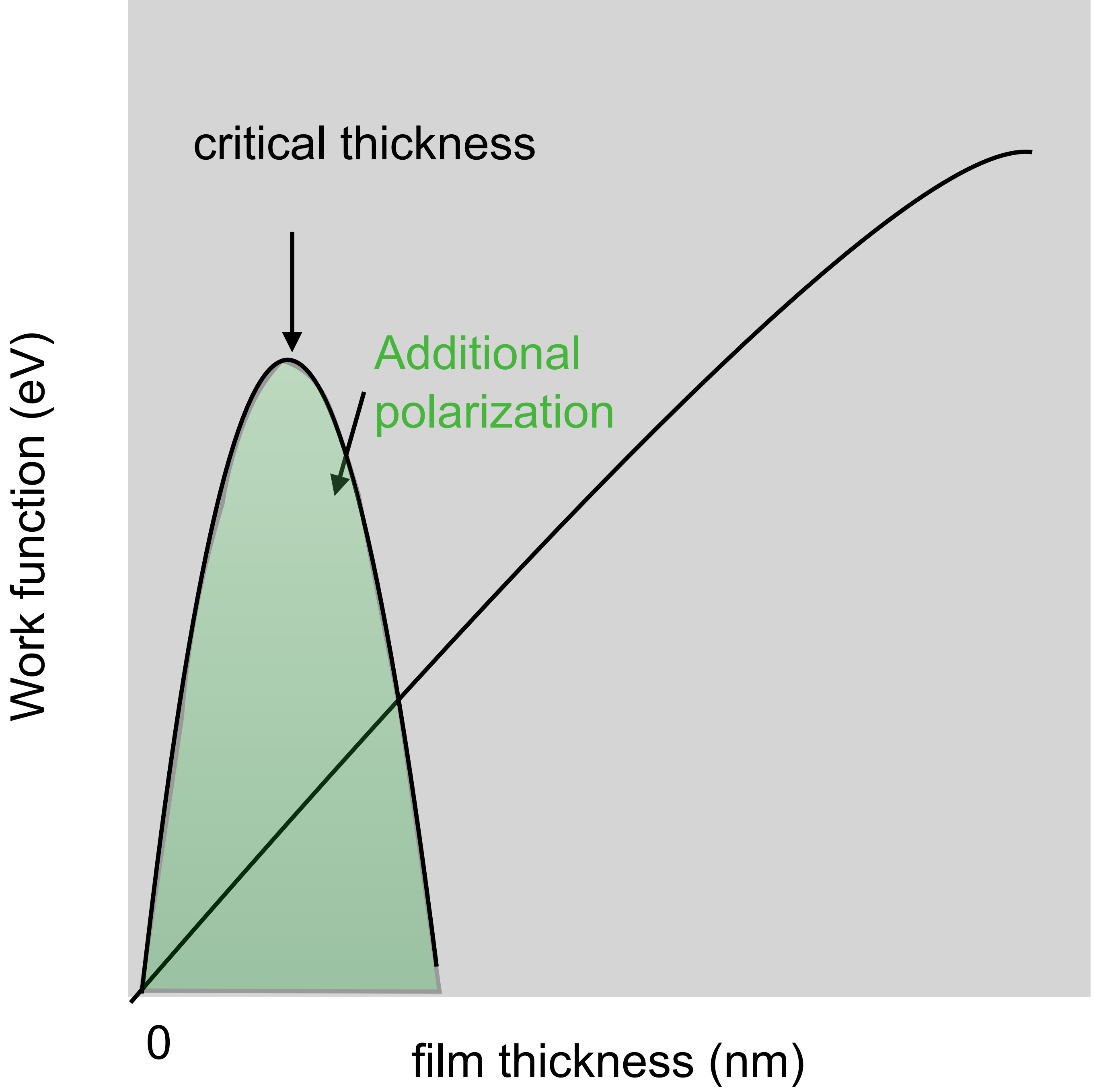

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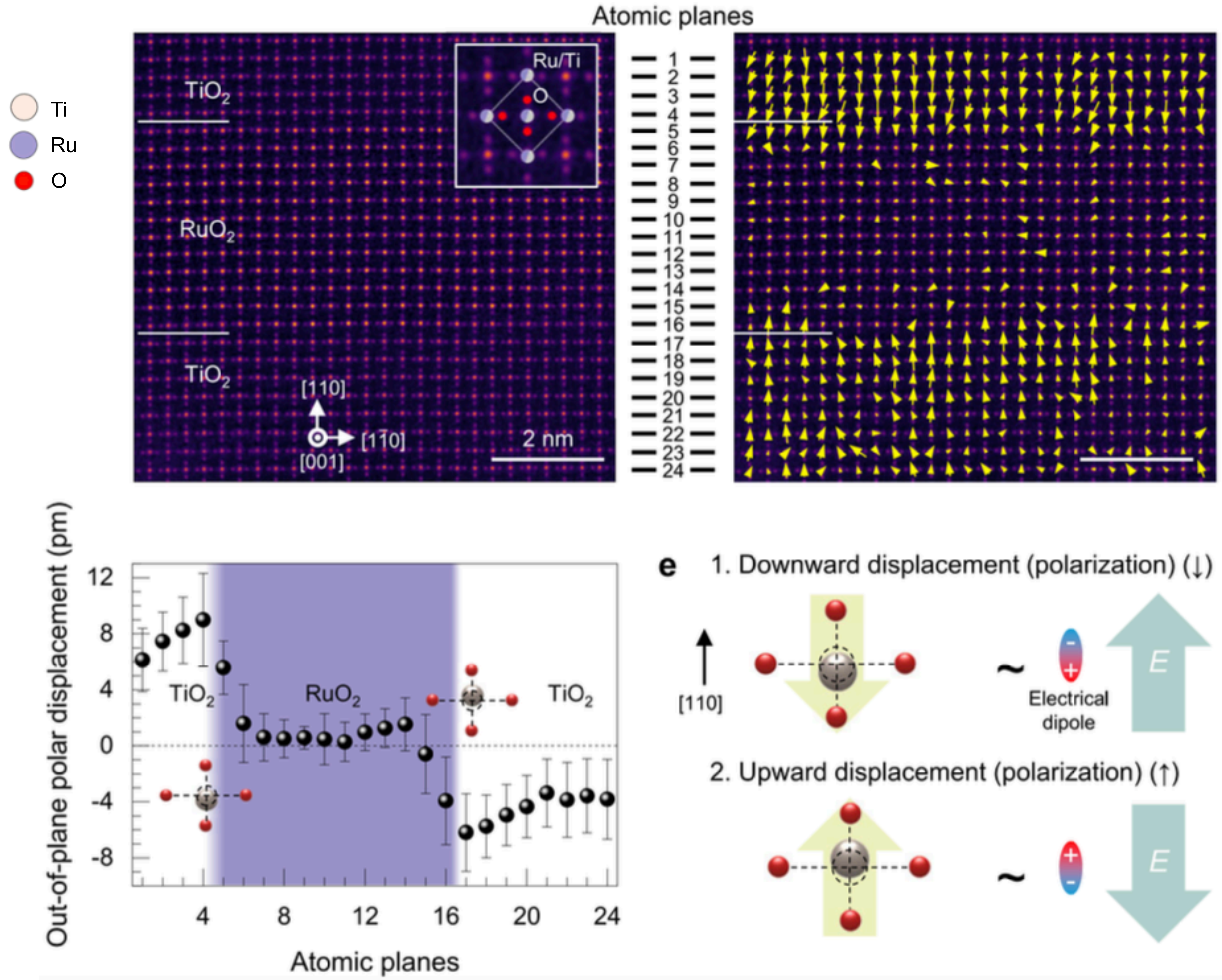

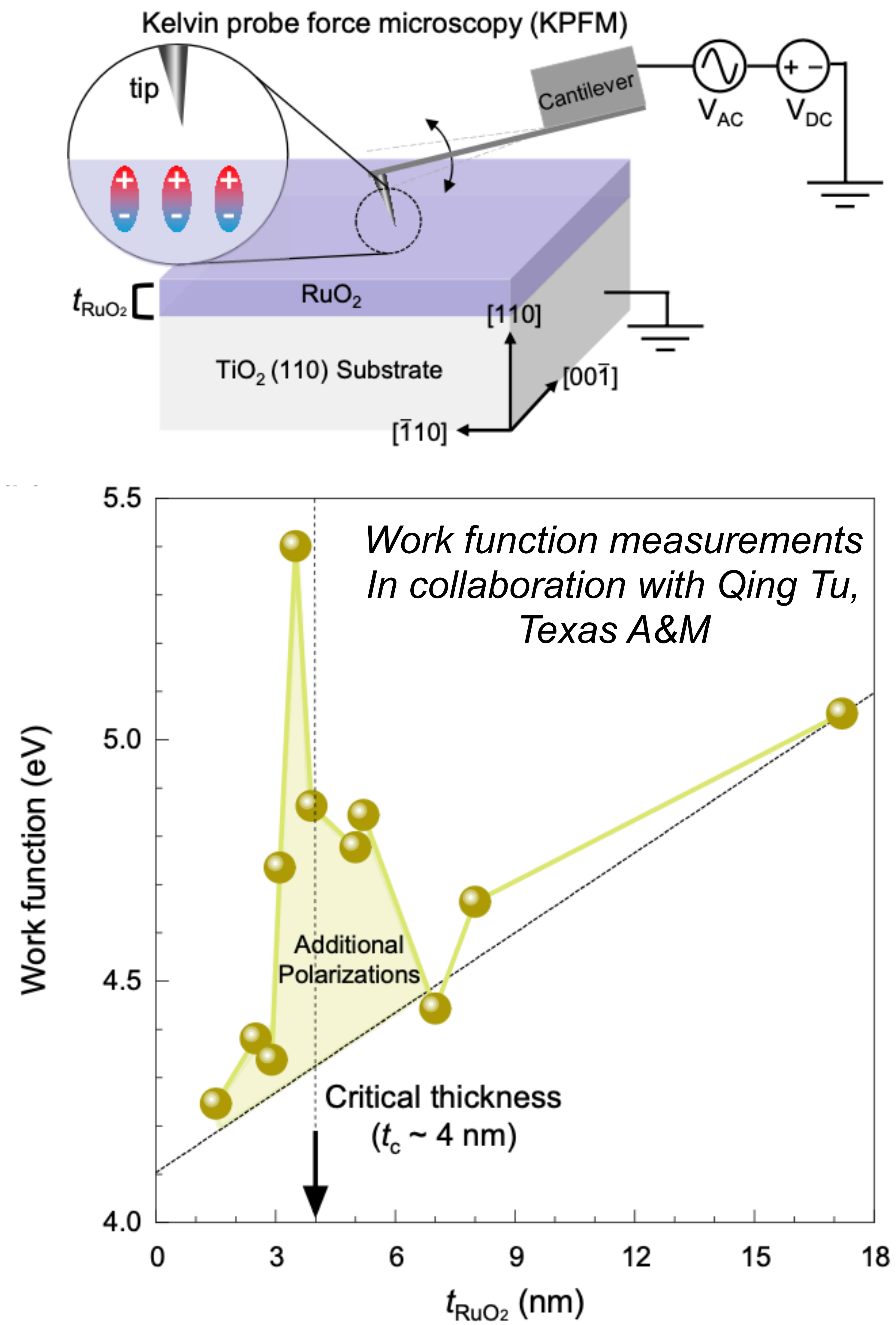

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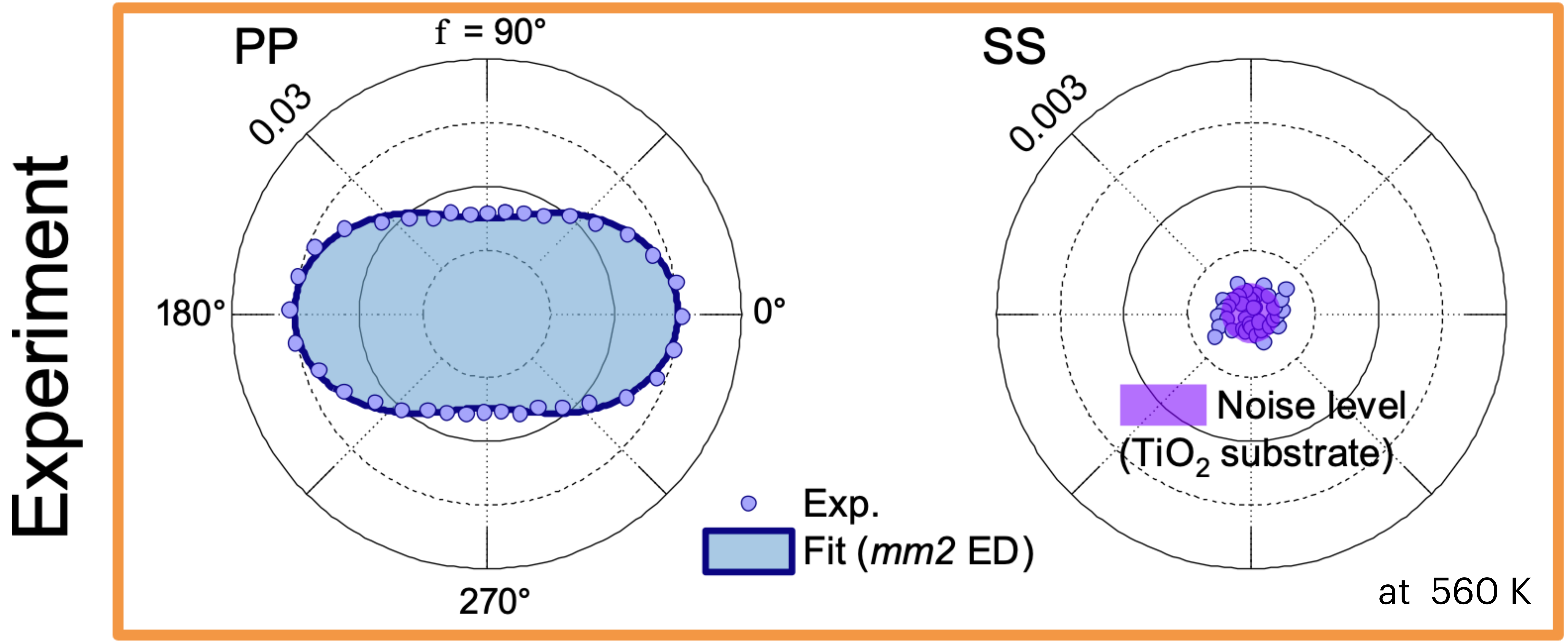
S. G. Jeong, …. J. M. LeBeau, and B. Jalan, Nat. Commun. (accepted) (2026)

Polarization consistent with the SHG point group..

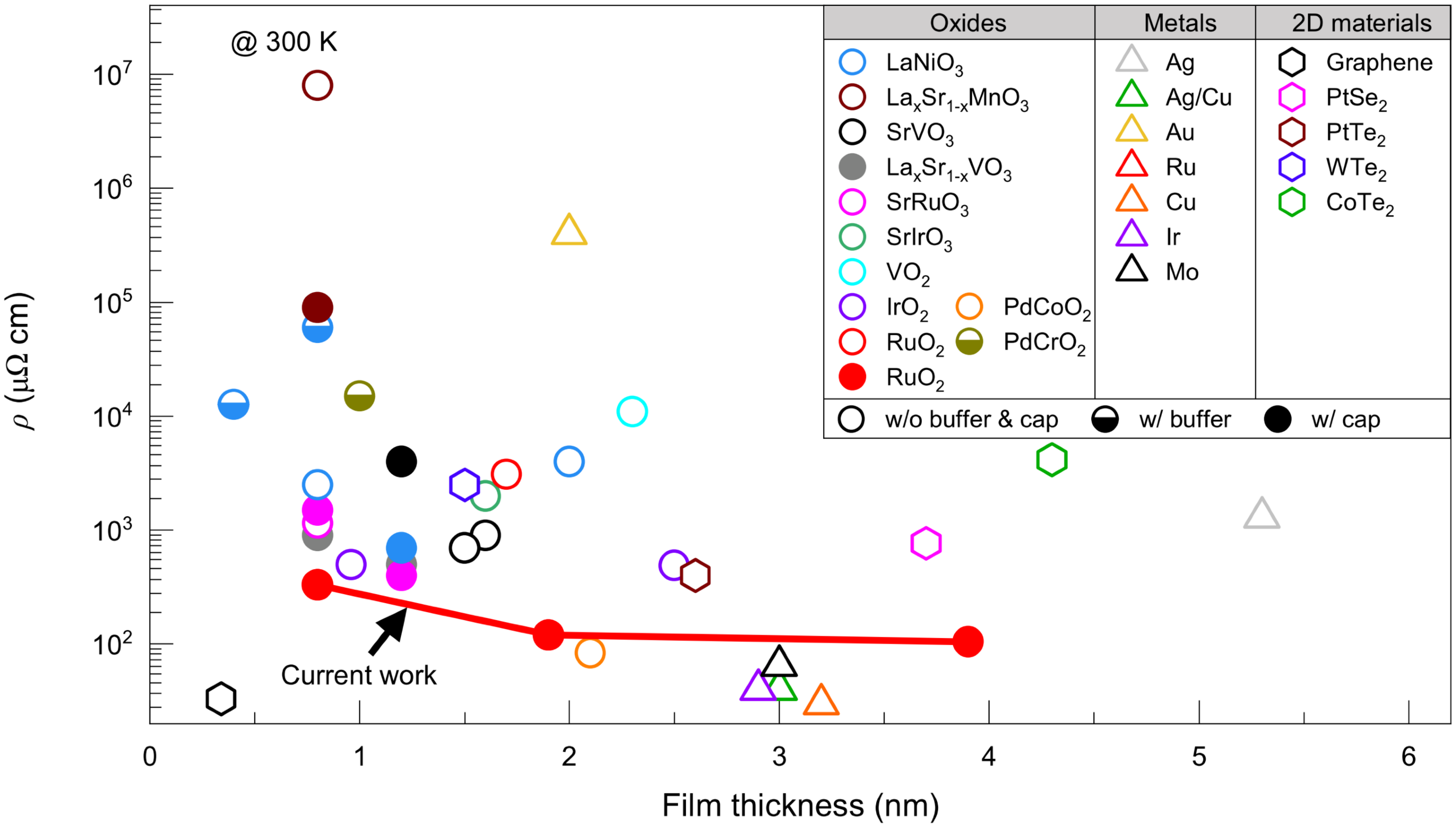


SHG measurements In collaboration with Prof. Jeongseok Lee, GIST, S. Korea

SHG analyses suggest *mm2* point group in fully-strained RuO_2/TiO_2 (110)

S. G. Jeong, I. H. Choi, S. Nair, L Buiarelli, B. Pourbahari, J. Y. Oh, N. Bassim, A. Seo, W. S. Choi, R. M. Fernandes, T. Birol, L. Zhao, J. S. Lee, and B. Jalan, Altermagnetic polar metallic phase in ultra-thin epitaxially-strained RuO2 films, PNAS (accepted) (2026)

Most conducting metal at atomic scale.



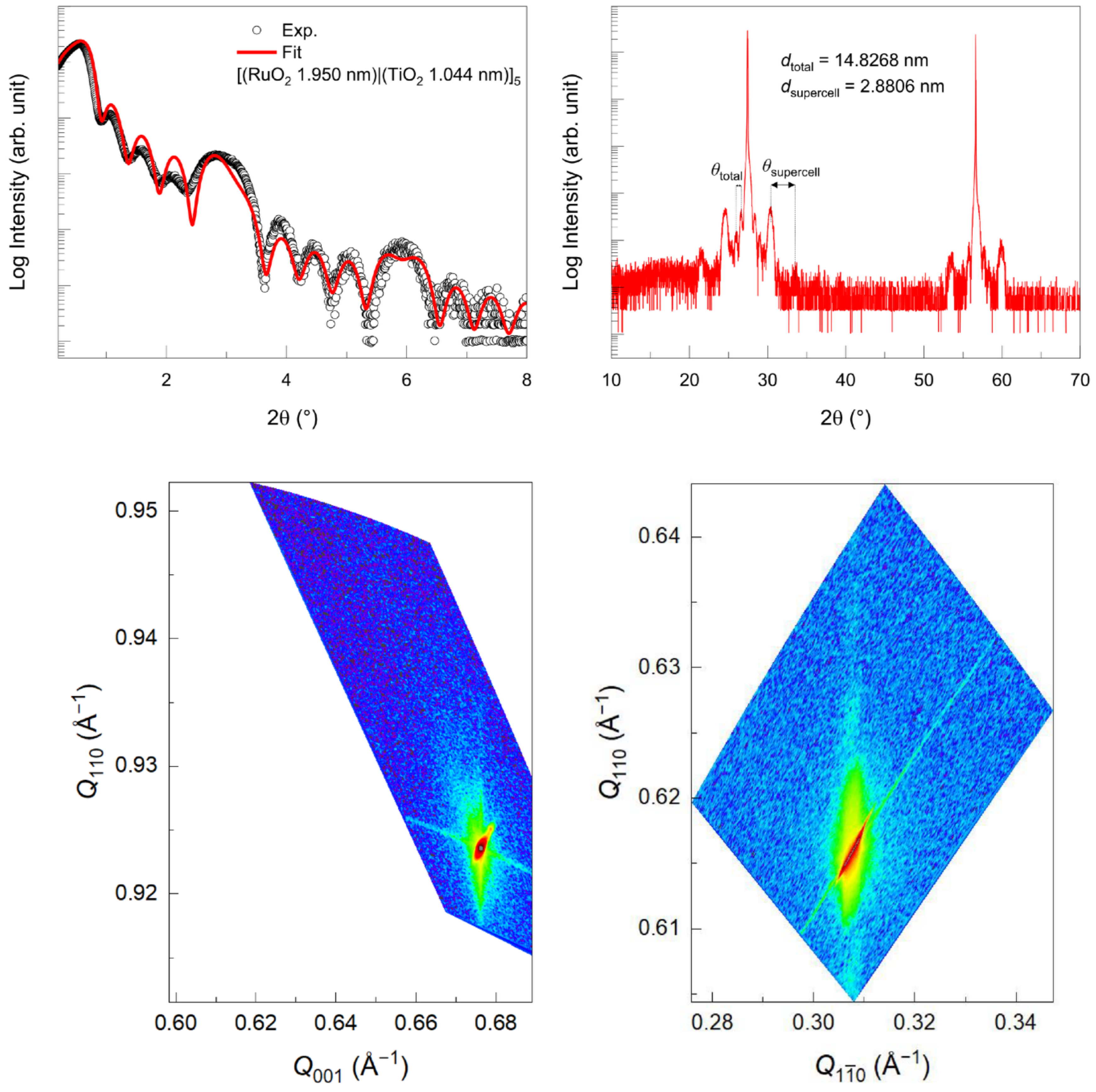
- *Conductivity down to 2-3 u.c.!*
- *It reveals the lowest resistivity among all known materials (except graphene) at nano-scale!*

S. G. Jeong, S. Lee, B. Lin, Z. Yang, I. H. Choi, J. Y Oh, S. Song, S. W. Lee, S. Nair, R. Choudhary, J. Parikh, S. Park, W. S. Choi, J. S. Lee, J. M. LeBeau, T. Low, and B. Jalan, Metallicity and Anomalous Hall Effect in Epitaxially-Strained, Atomically-thin RuO2 Films, PNAS 122 (24) e2500831122 (2025)

Is the hybrid MBE-grown RuO_2 film magnetic?

Magnetism is further evident in MOKE & Linear Dichroism

[2 nm RuO_2/1 nm TiO_2]$_5$ superlattice



- Superlattice with excellent interfaces as evident from persistent Kiessig fringes
- Both MOKE and LD confirms magnetism.
- AFM-like transition observed at ~ 500 K

MOKE and LD measurements In collaboration with Prof. Jeongseok Lee, GIST, S. Korea

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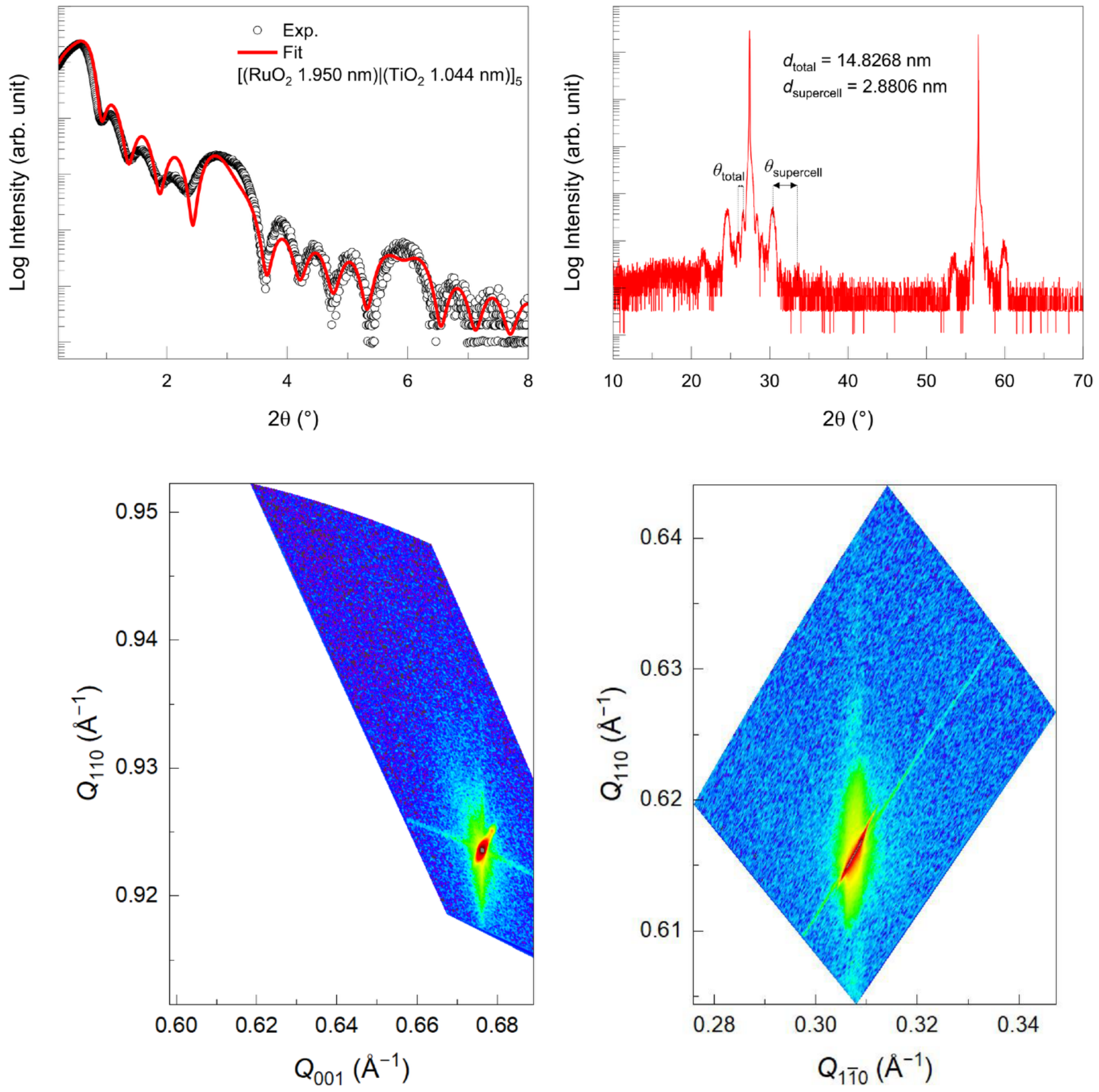

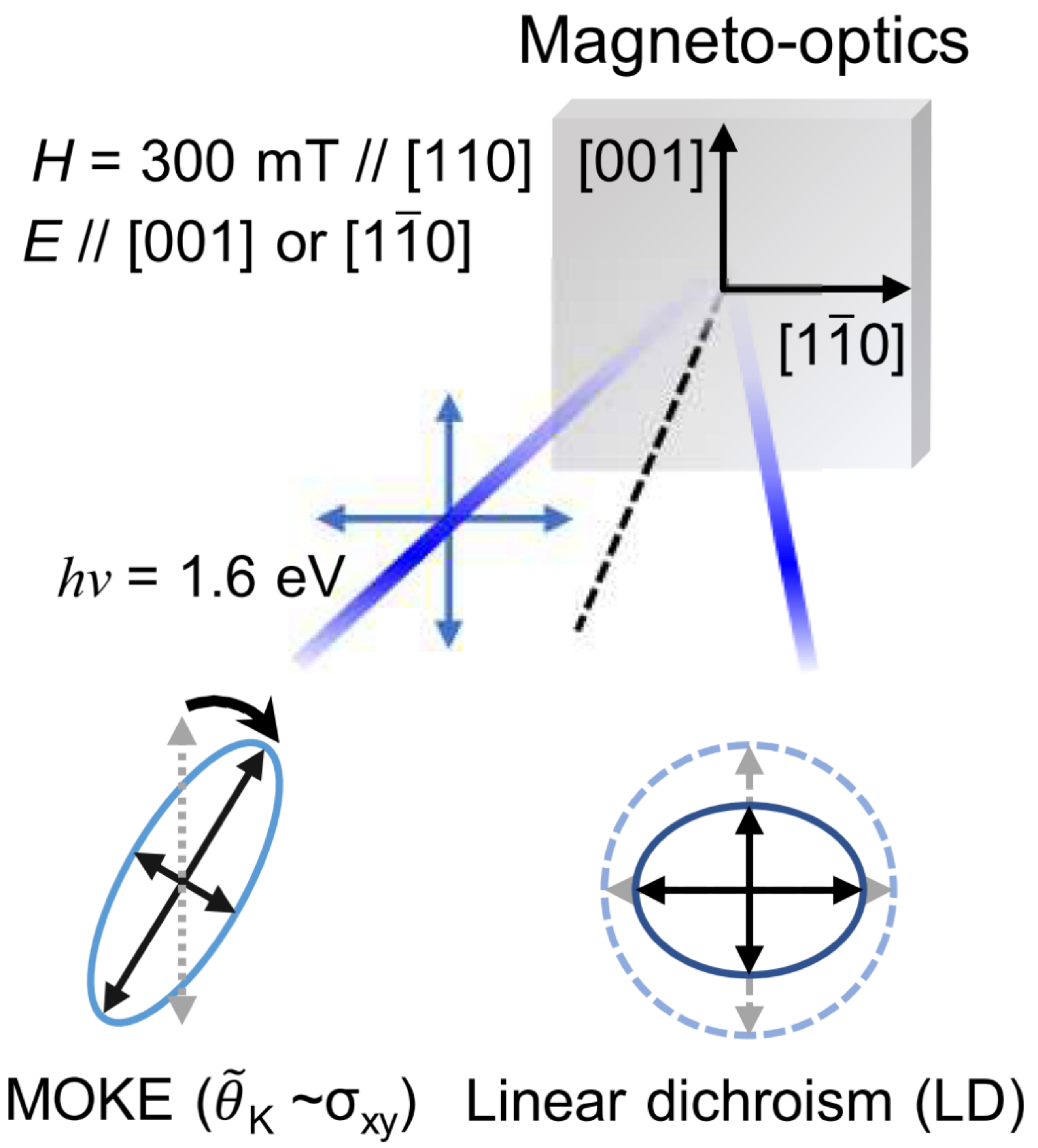

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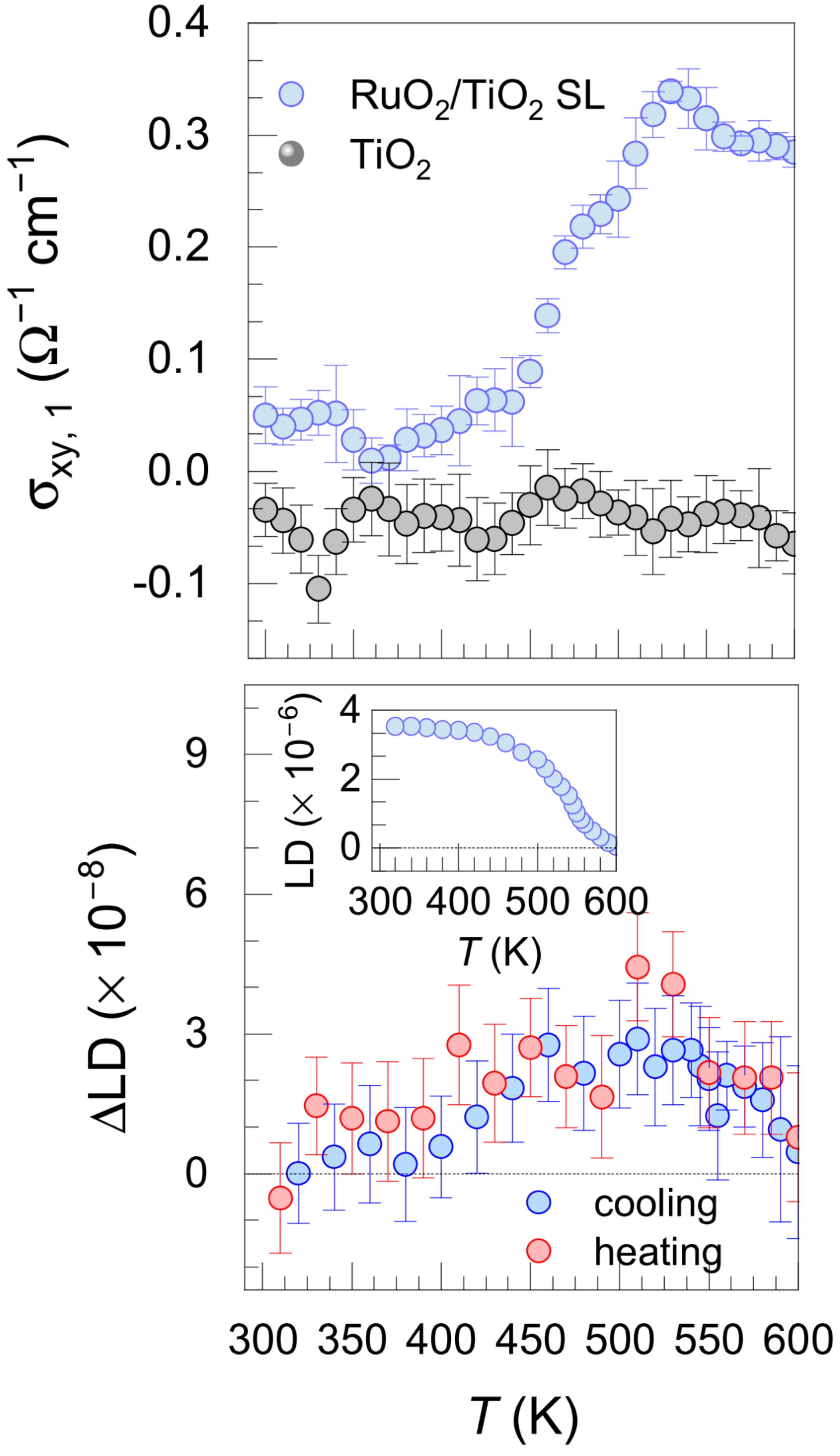

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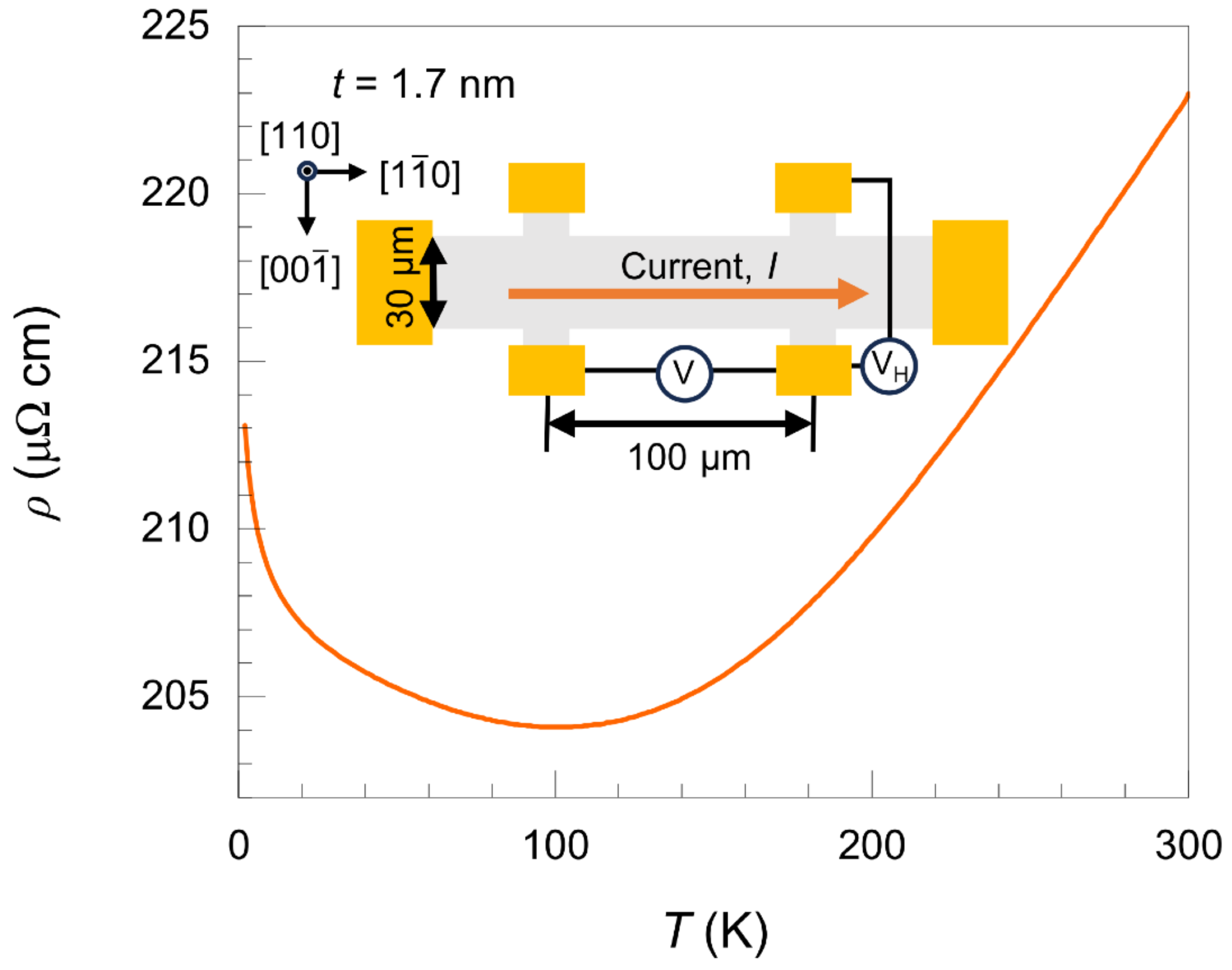
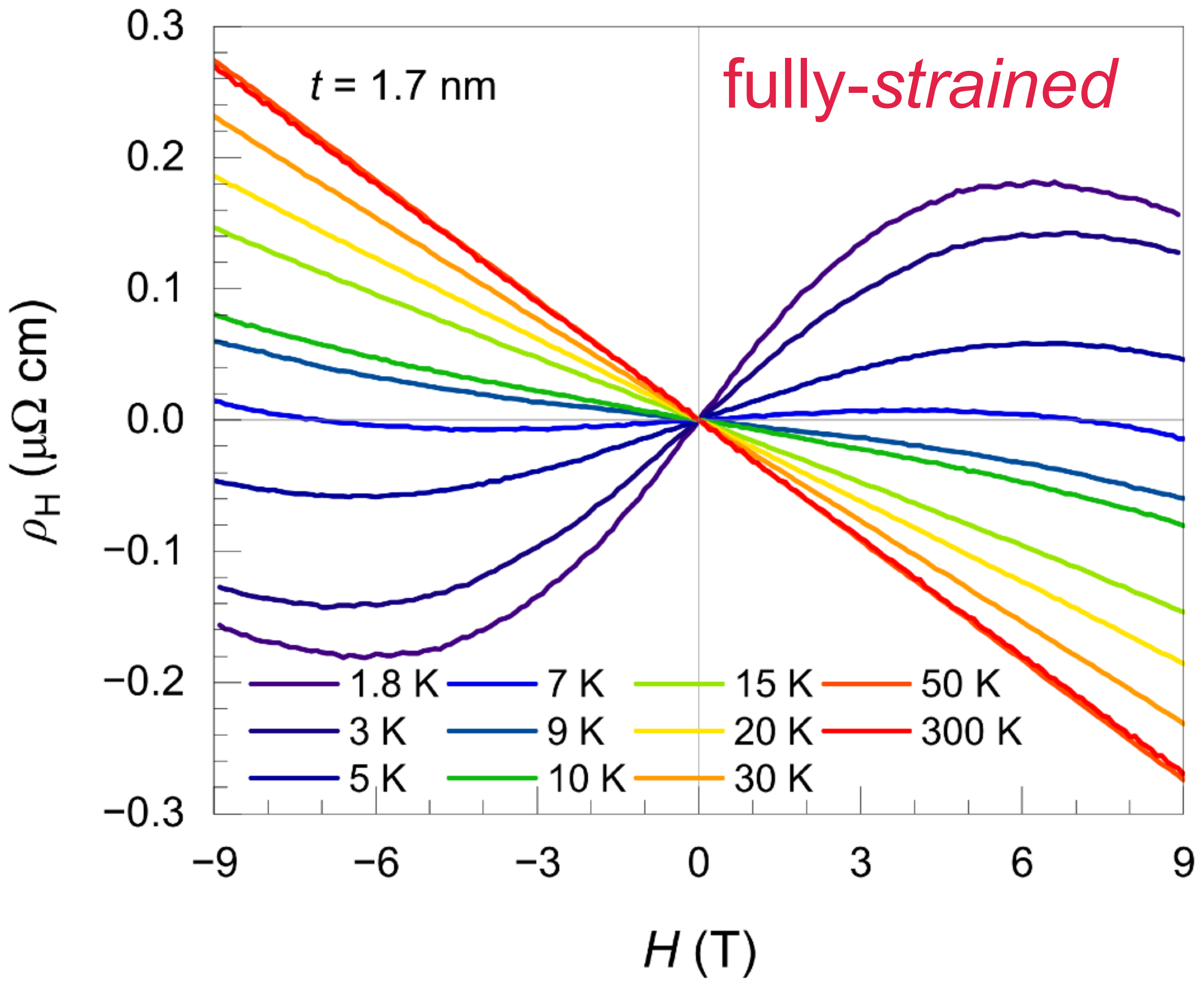
Non Linear Hall Effect in Strained RuO_2 Films





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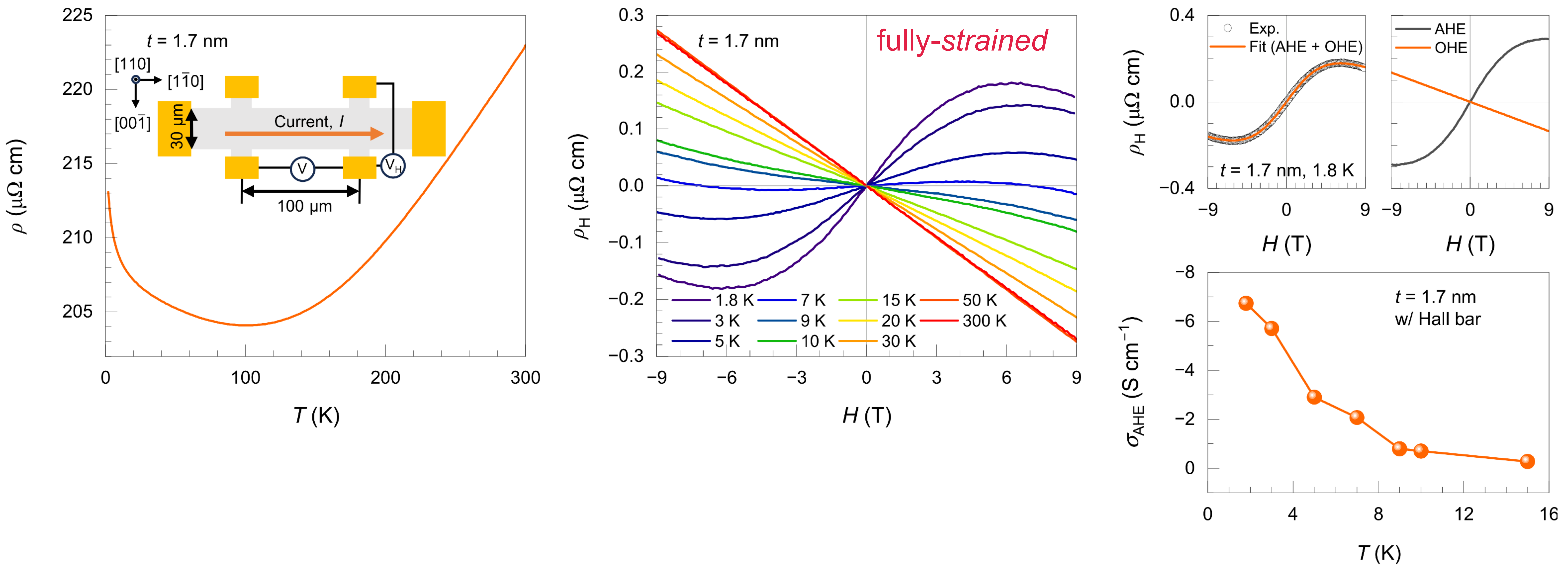

- Non-linear Hall owing to anomalous Hall effect
- Fits yield AHE conductivity which rapidly increases < 13 K

S. G. Jeong, S. Lee, B. Lin, Z. Yang, I. H. Choi, J. Y Oh, S. Song, S. W. Lee, S. Nair, R. Choudhary, J. Parikh, S. Park, W. S. Choi, J. S. Lee, J. M. LeBeau, T. Low, and B. Jalan, Metallicity and Anomalous Hall Effect in Epitaxially-Strained, Atomically-thin RuO2 Films, PNAS 122 (24) e2500831122 (2025)

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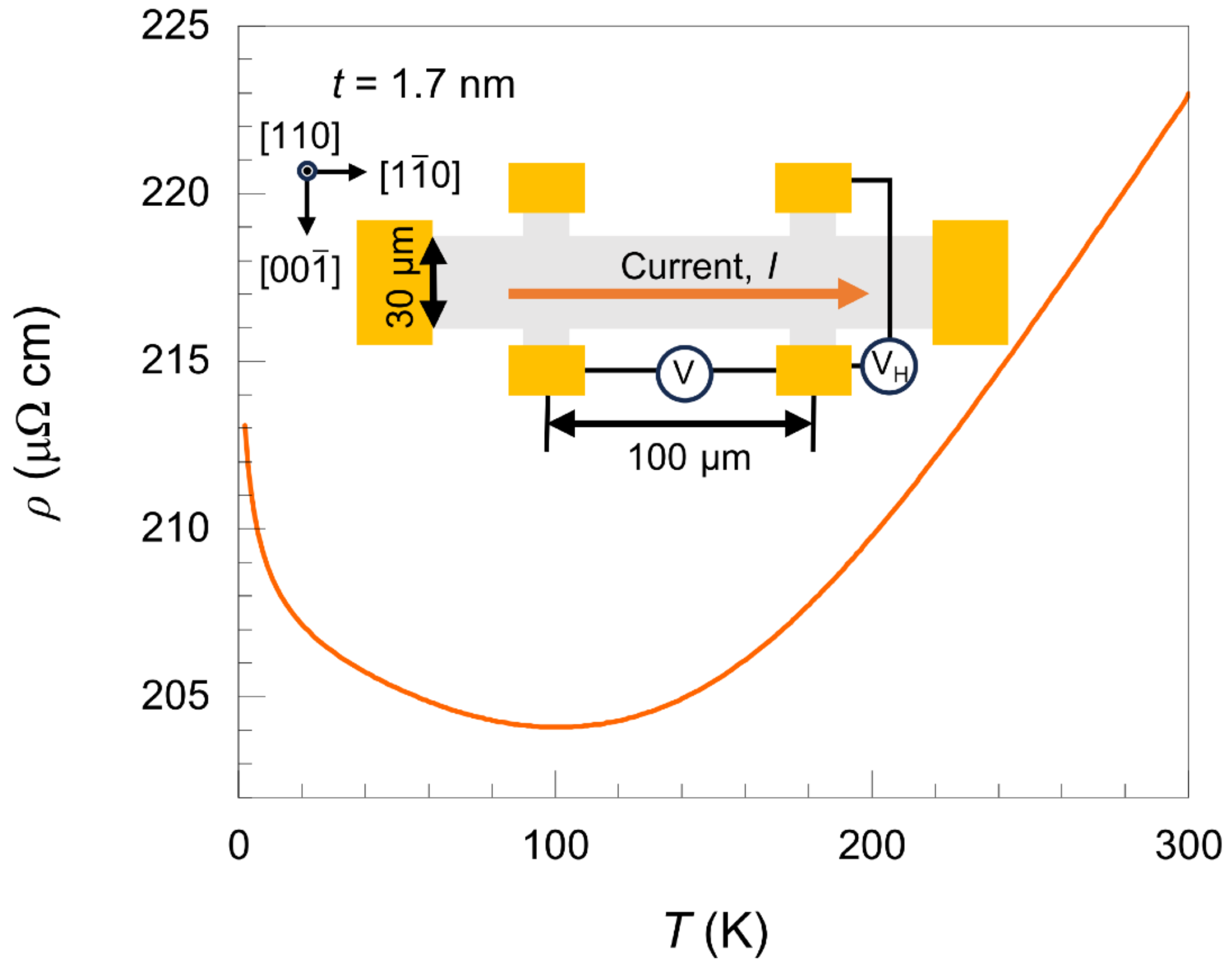

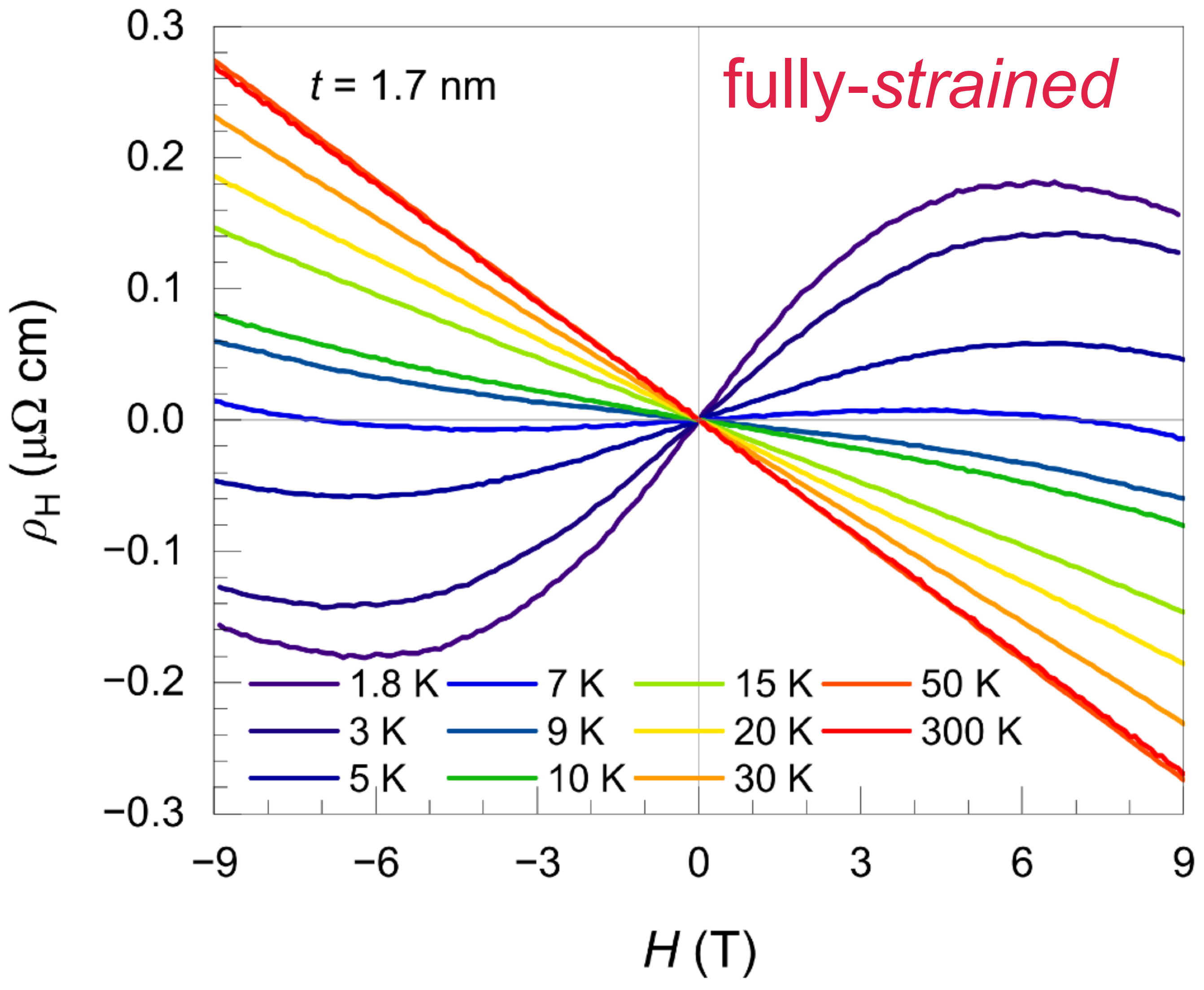

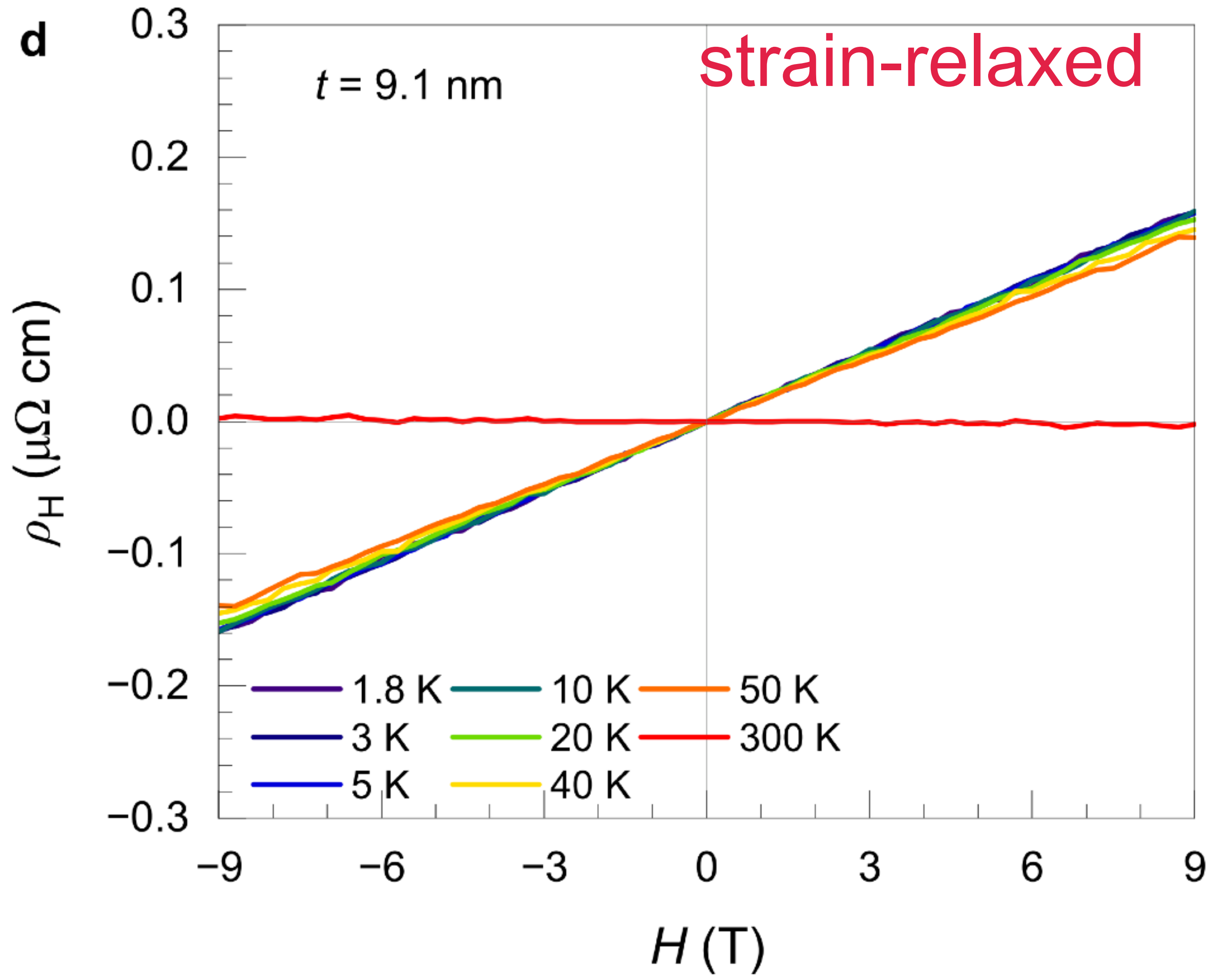

- No AHE in relaxed films

- Non-linear Hall owing to anomalous Hall effect
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S. G. Jeong, S. Lee, B. Lin, Z. Yang, I. H. Choi, J. Y Oh, S. Song, S. W. Lee, S. Nair, R. Choudhary, J. Parikh, S. Park, W. S. Choi, J. S. Lee, J. M. LeBeau, T. Low, and B. Jalan, Metallicity and Anomalous Hall Effect in Epitaxially-Strained, Atomically-thin RuO2 Films, PNAS 122 (24) e2500831122 (2025)

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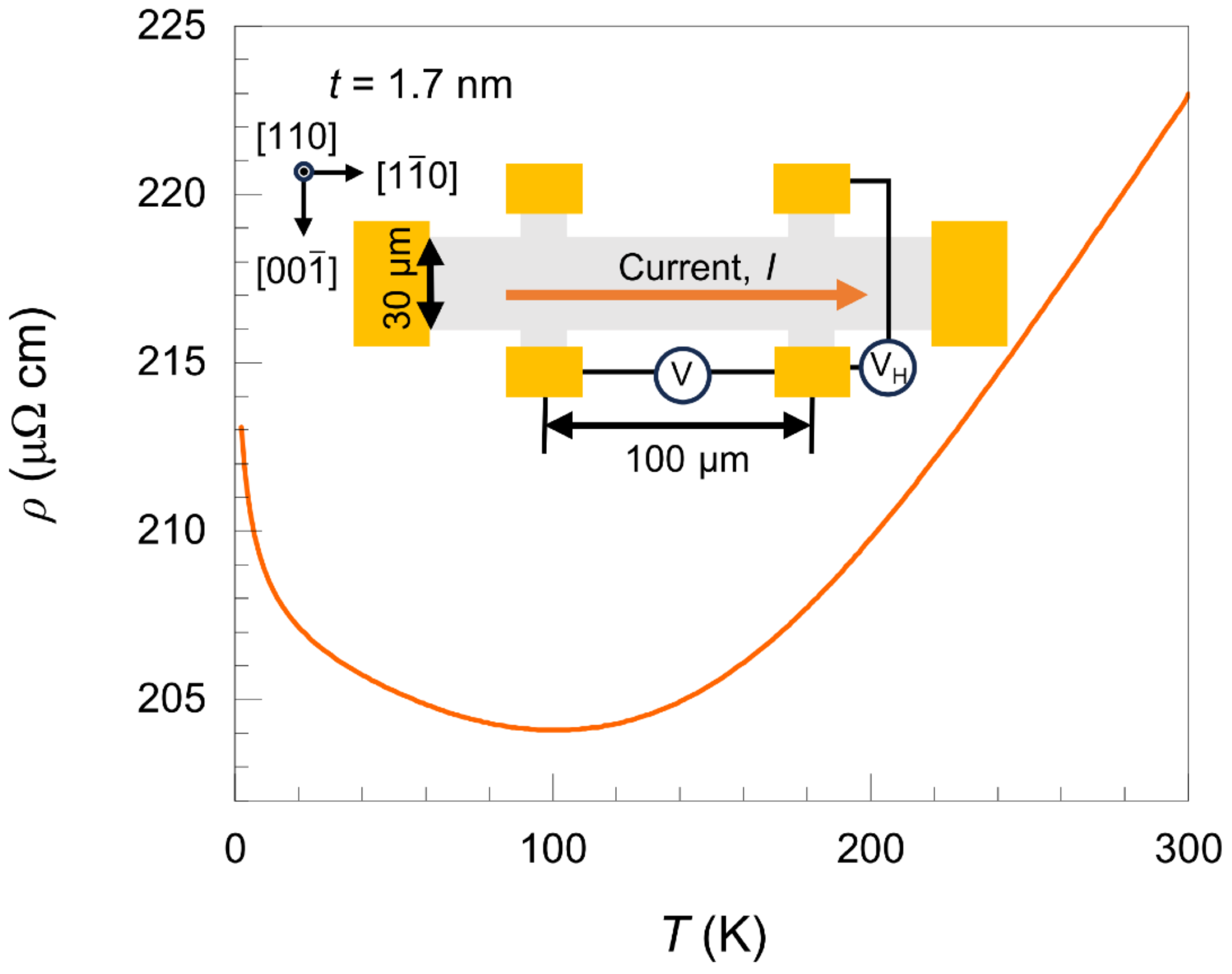

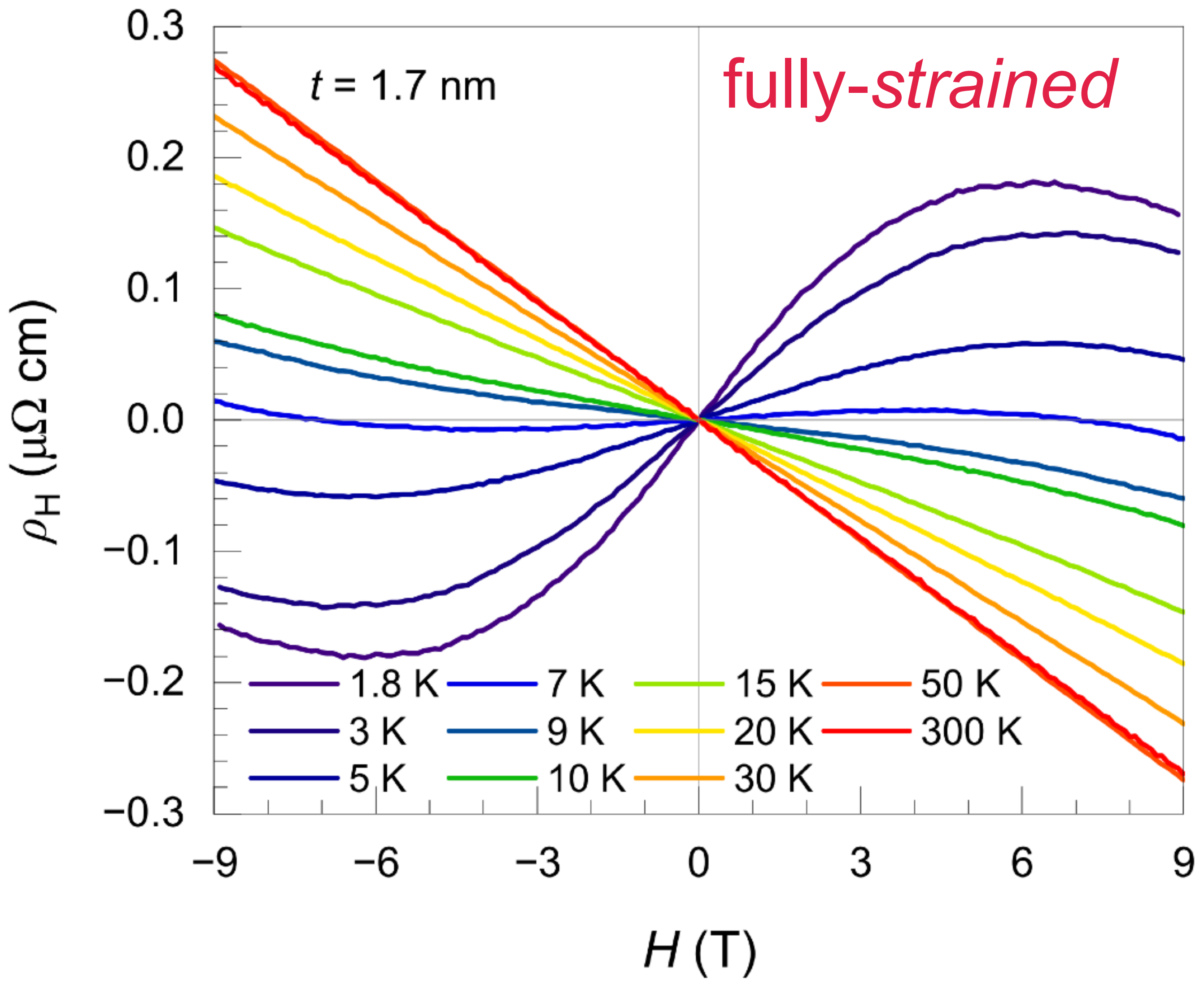

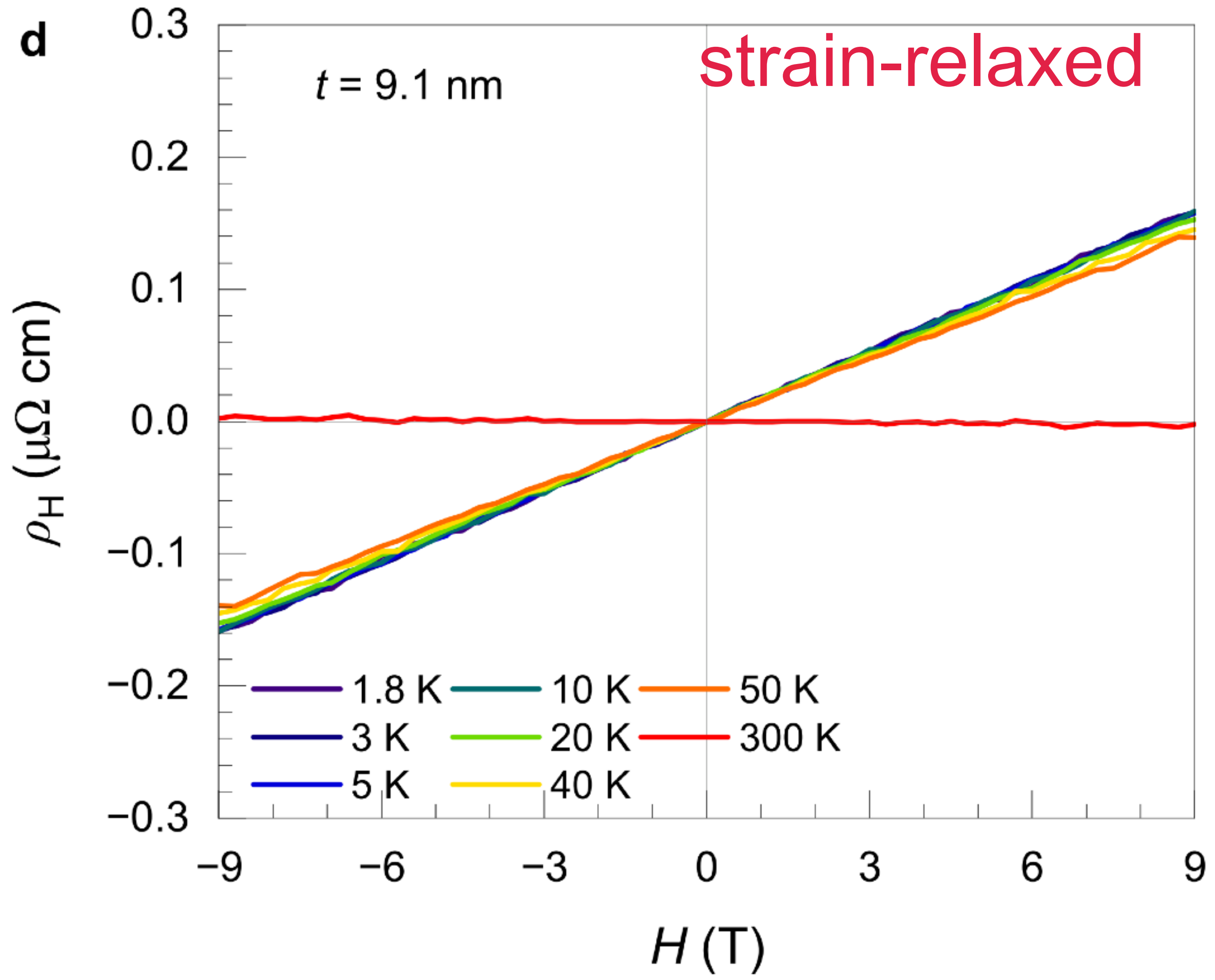

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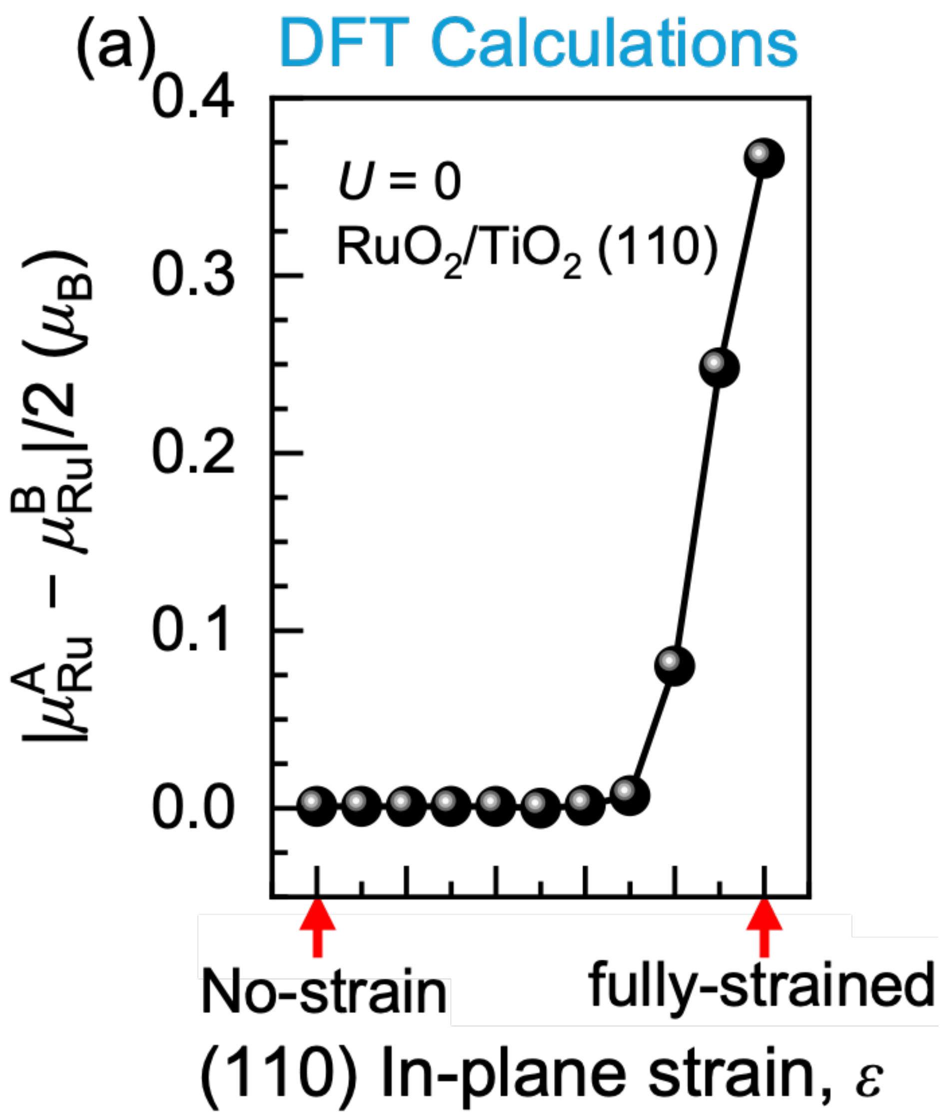

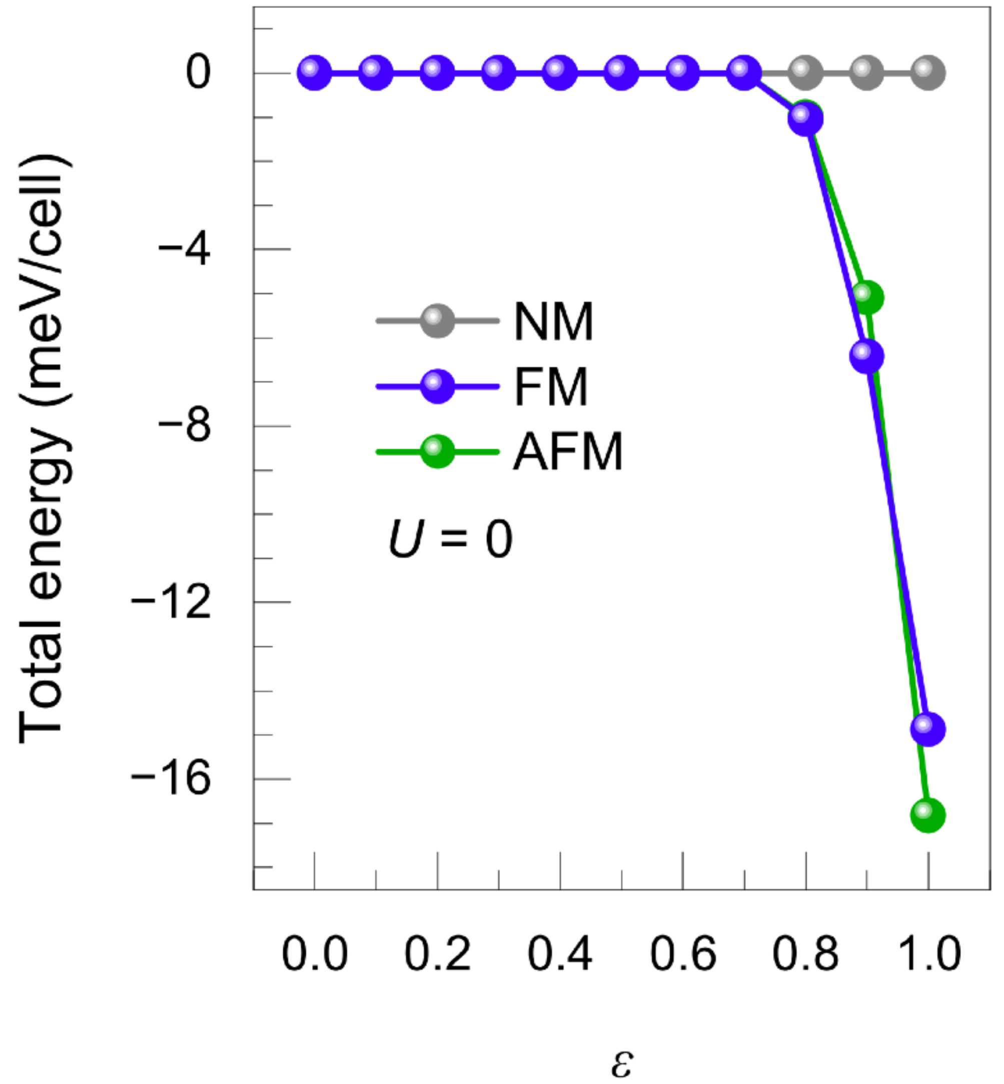

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- Fits yield AHE conductivity which rapidly increases < 13 K
- DFT w/o +*U* reveals net magnetic moments in addition to revealing lower energy for altermagnetic (AFM) phase

S. G. Jeong, S. Lee, B. Lin, Z. Yang, I. H. Choi, J. Y Oh, S. Song, S. W. Lee, S. Nair, R. Choudhary, J. Parikh, S. Park, W. S. Choi, J. S. Lee, J. M. LeBeau, T. Low, and B. Jalan, Metallicity and Anomalous Hall Effect in Epitaxially-Strained, Atomically-thin RuO2 Films, PNAS 122 (24) e2500831122 (2025)

Is the hybrid MBE-grown RuO_2 film magnetic?

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Magnetism is further evident in Polarized Neutron Reflectometry

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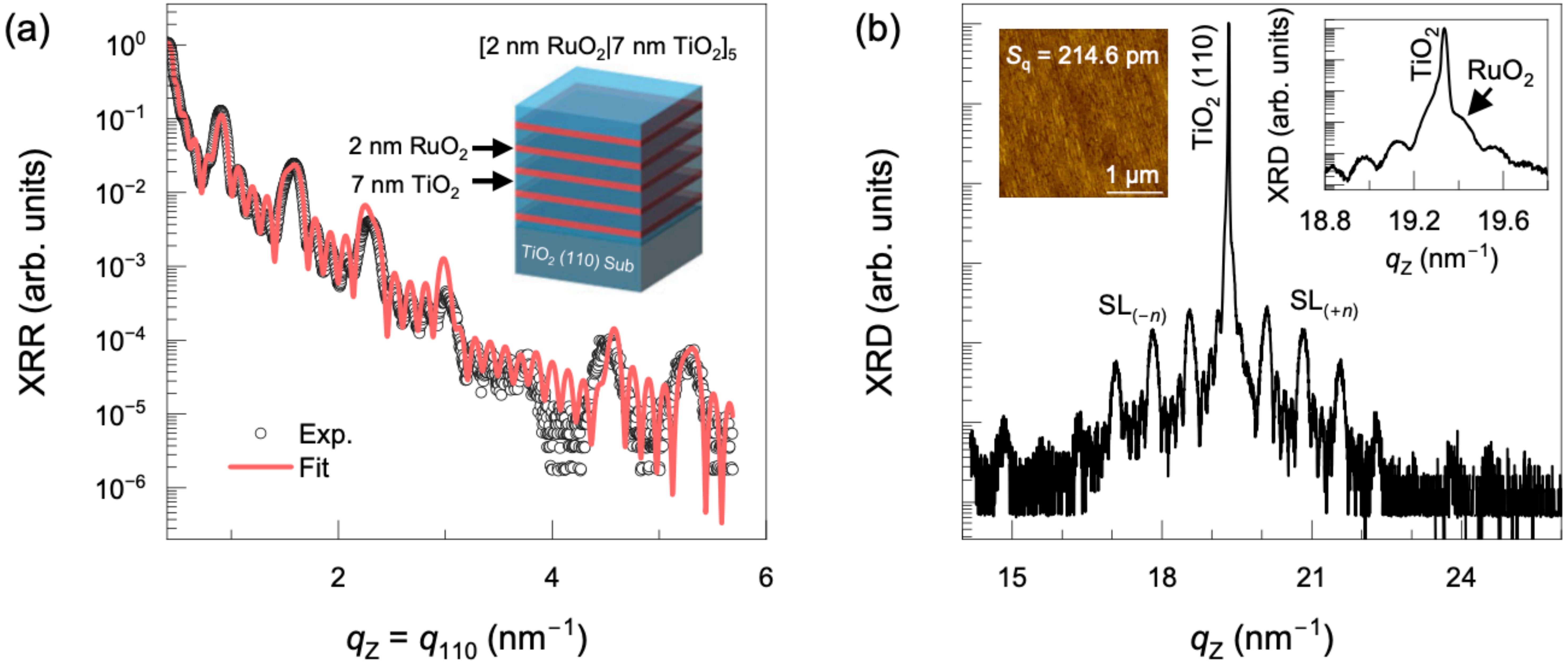

PNR: in collaboration with Valeria Lauter, SNS, ORNL

- Superlattice with excellent interfaces as evident from persistent Kiessig fringes
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S. G. Jeong, S. Lee, ……., Valeria, and B. Jalan, submitted

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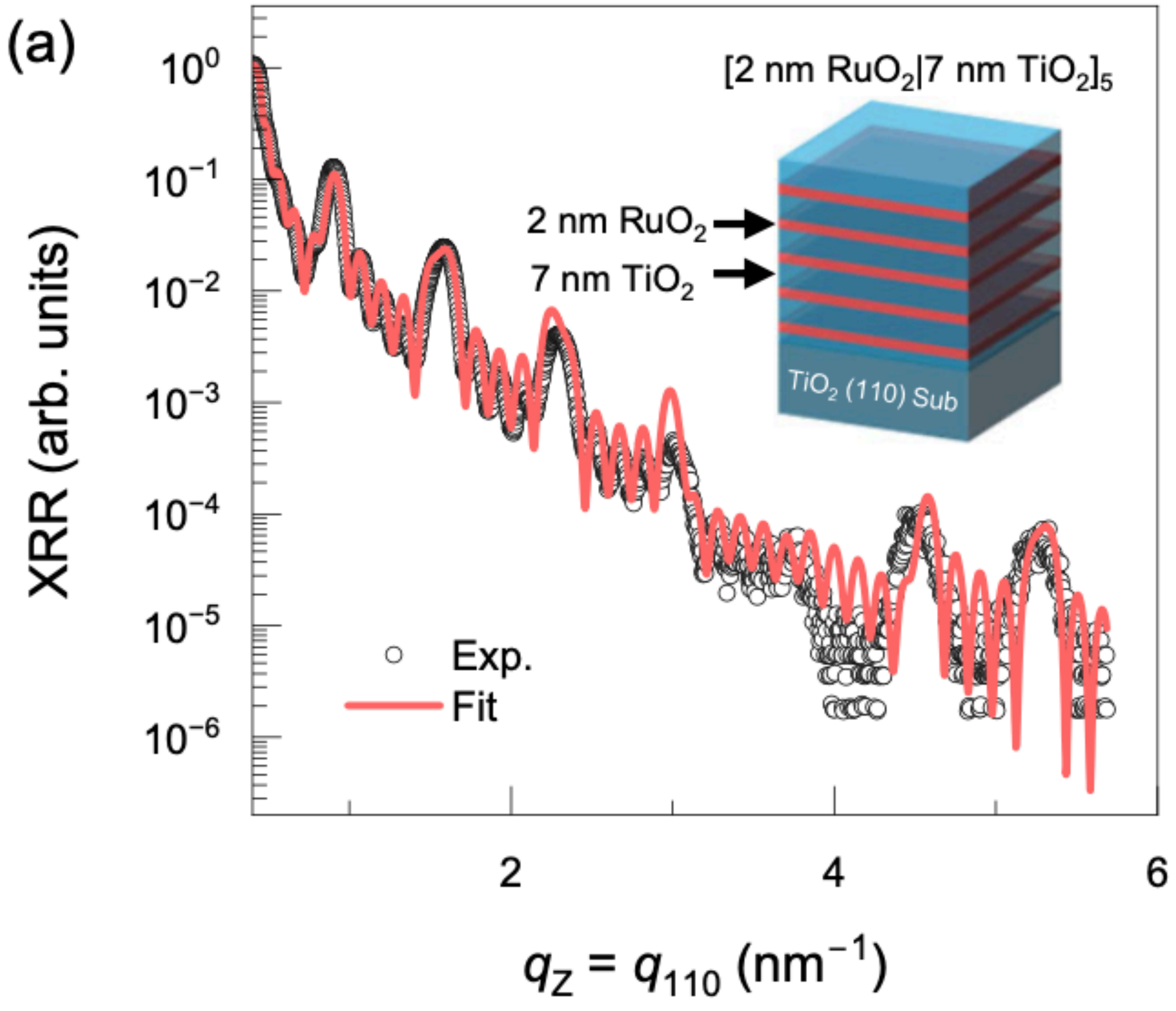
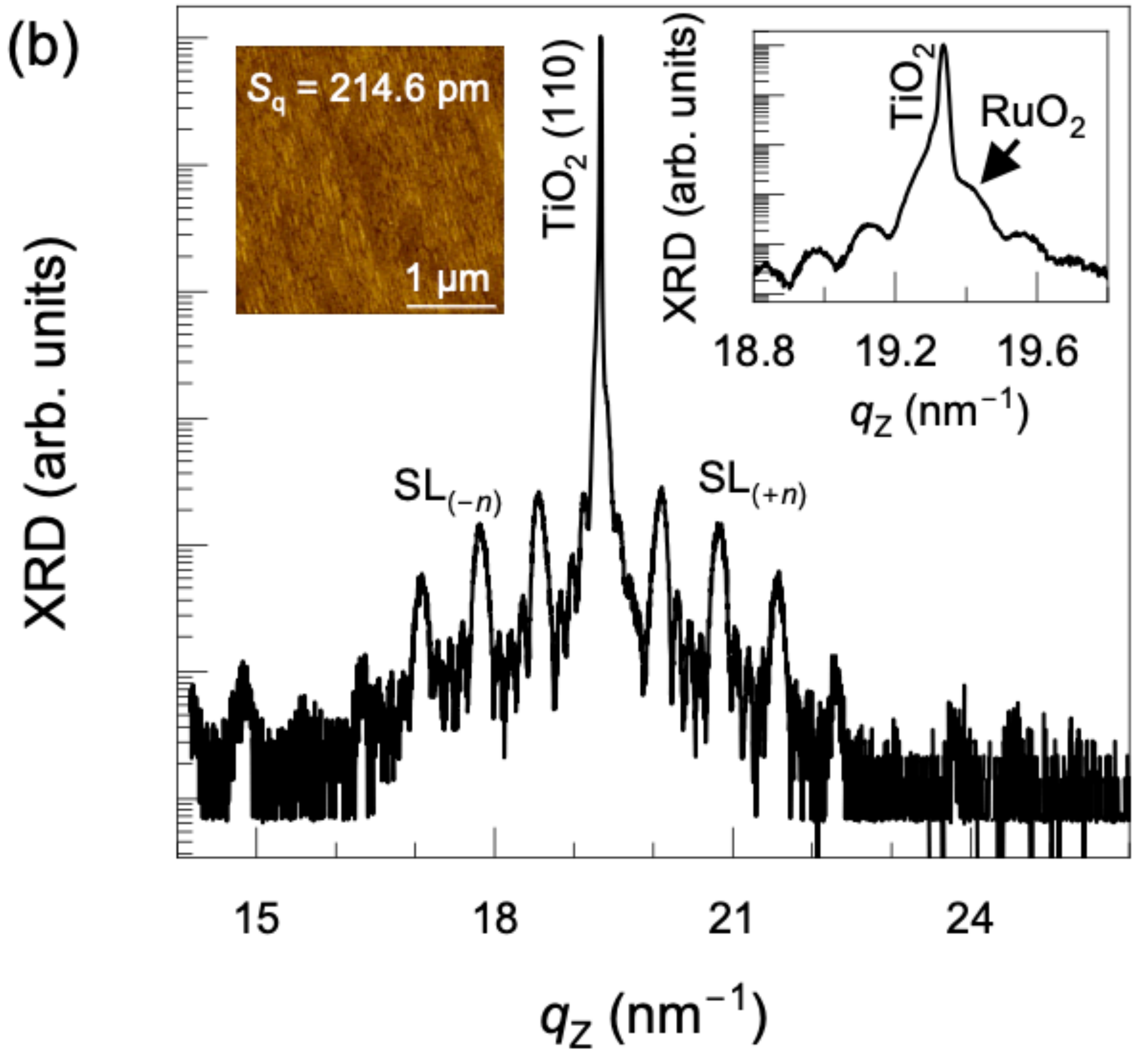
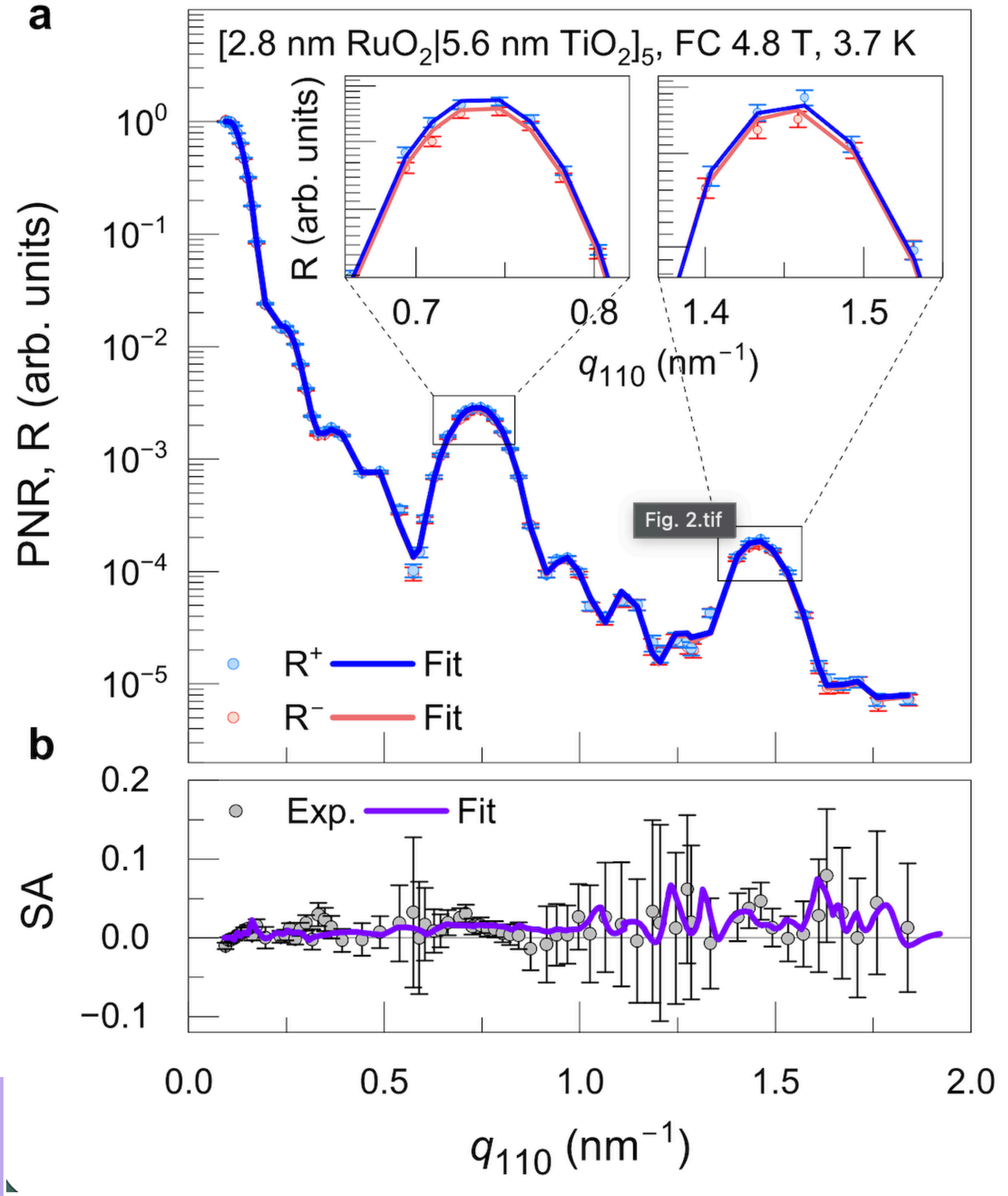




PNR: *in collaboration with Valeria Lauter, SNS, ORNL*

Polarized Neutron Reflectometry (PNR)



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S. G. Jeong, S. Lee, ……., Valeria, and B. Jalan, submitted

Summary: The trick is strain, the treat is emerging phases

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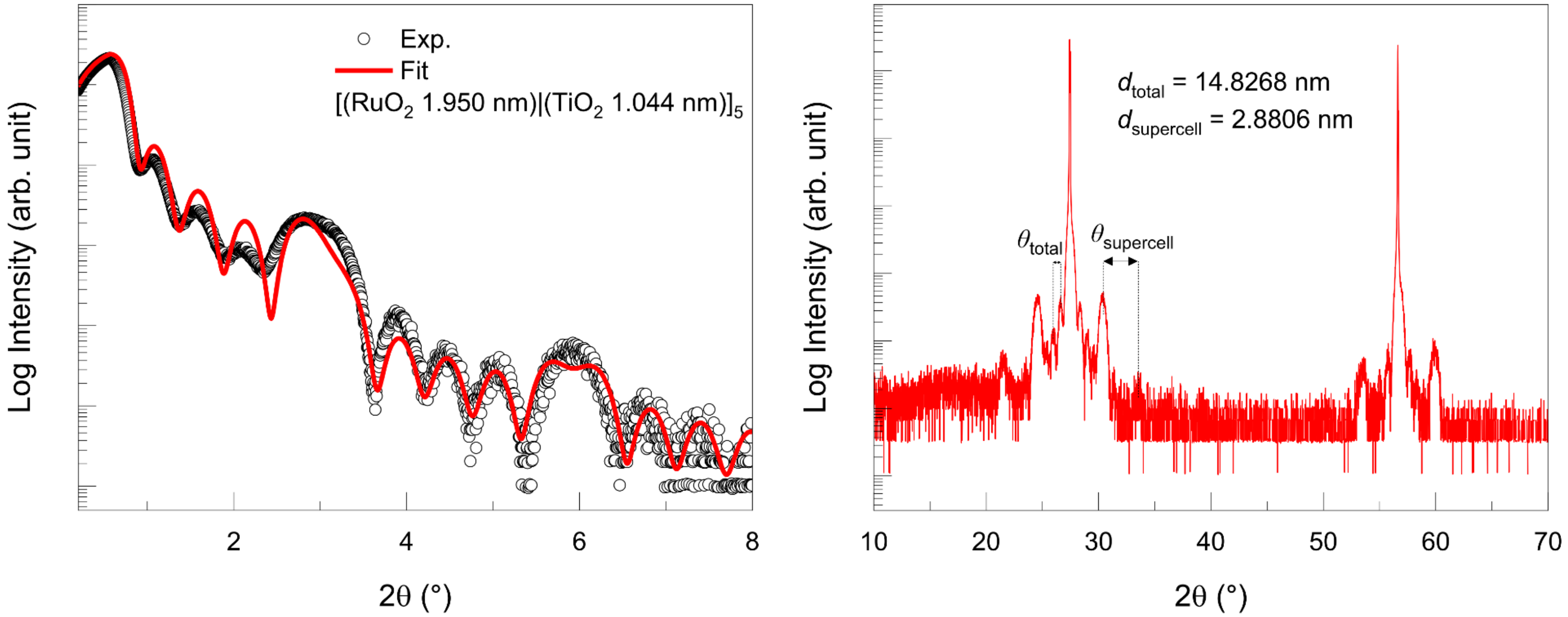
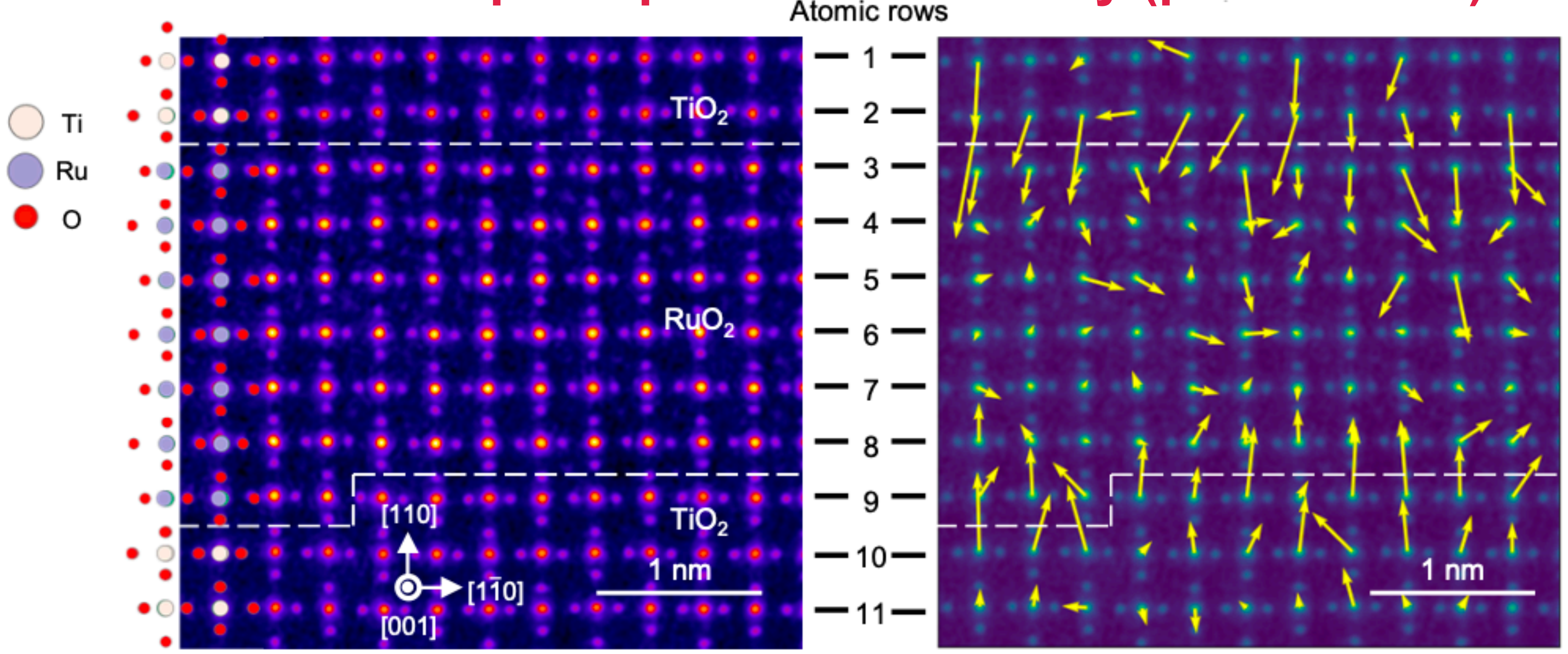
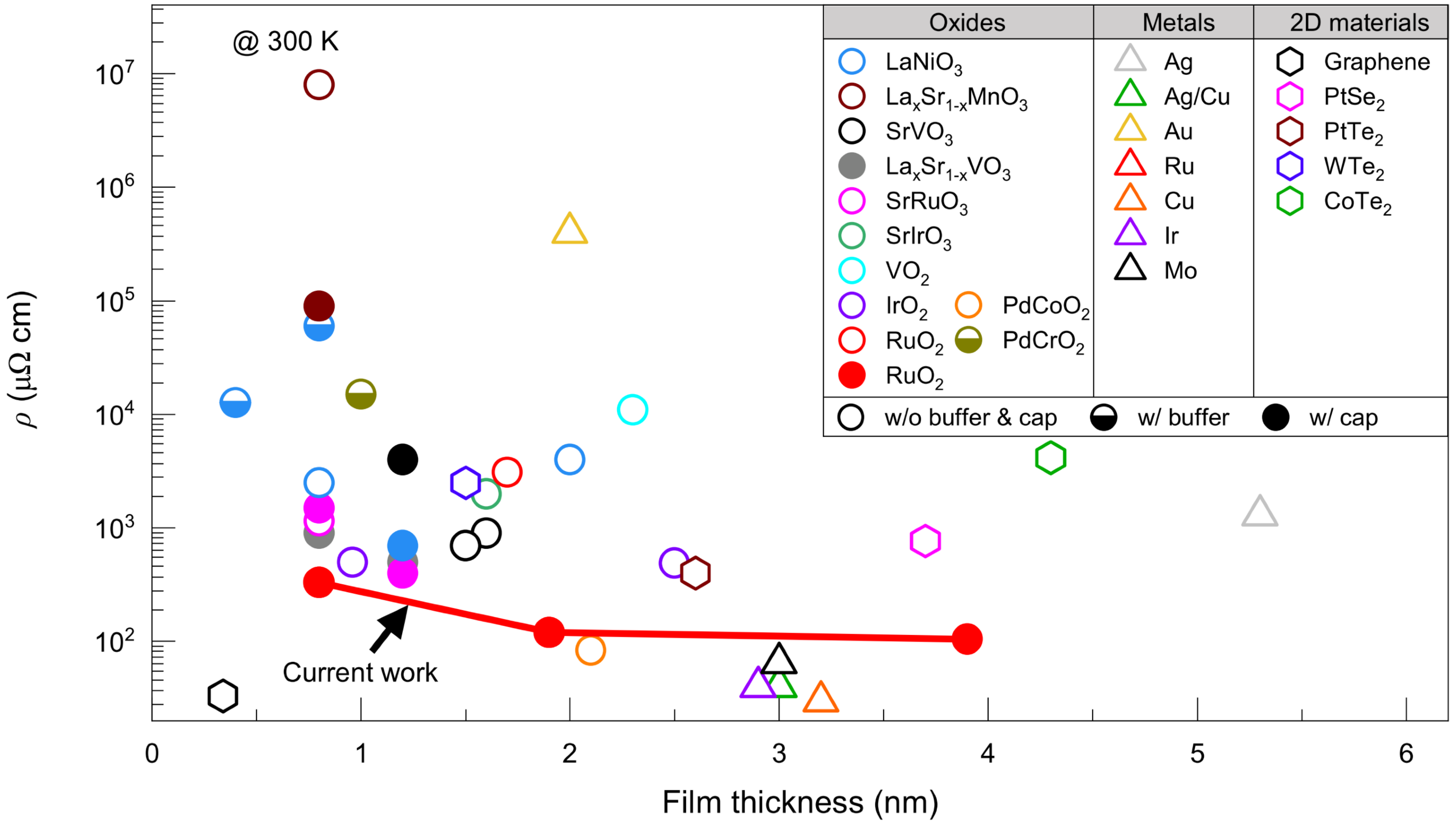
Excellent structural quality using Hybrid MBE



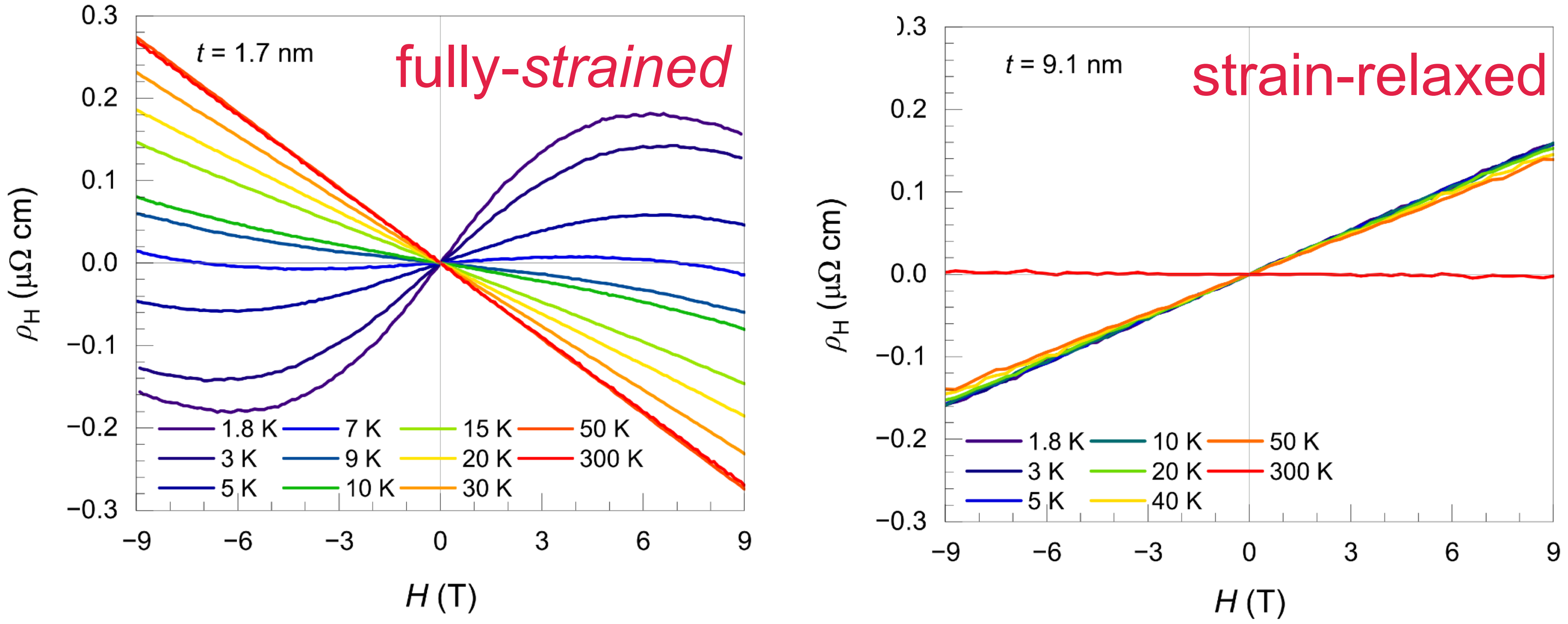
Interfacial polar phase + metallicity (polar metal!)



Excellent Metallicity down to 2-3 u.c. level (second to graphene)



Magnetism in RuO_2 film owing to *epitaxial strain*



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Thank you!

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