Symmetry engineering and novel physics of metallic polar interface

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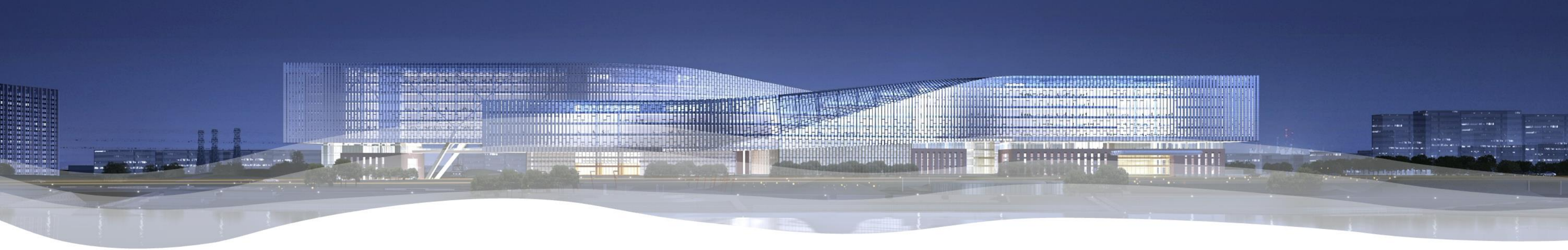


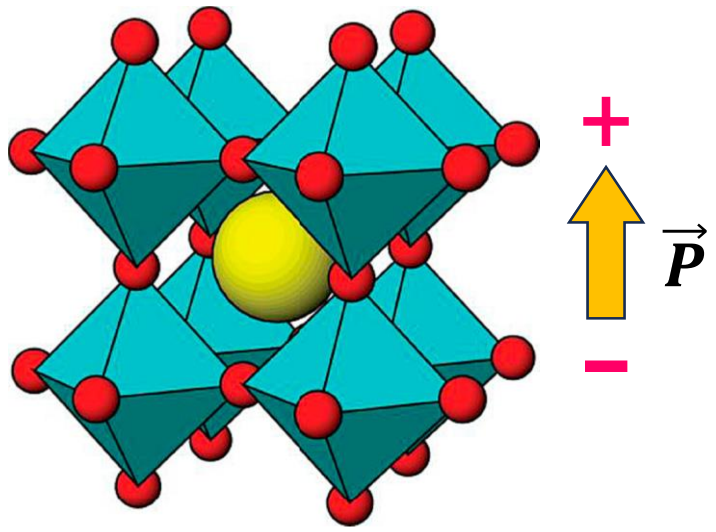
Outline

- Introduction of **Polar metallic interface**
- A novel method to modulate interface symmetry
- Emergent physical effects in polar interface

➢ Polar Metal

✓ Polar materials

- Polar materials are non-centrosymmetric crystals with a symmetry-allowed macroscopic dipole;
- Ferroelectrics are the subset whose dipole is switchable.
- Stablized by ① long-range Coulomb interactions, ② soft polar phonons



For example: $BaTiO_3$

✓ Metals: materials with free carriers

- Free carriers screen electric fields
- Screening usually stiffens polar soft modes

✓ Polar Metals: rare but exist

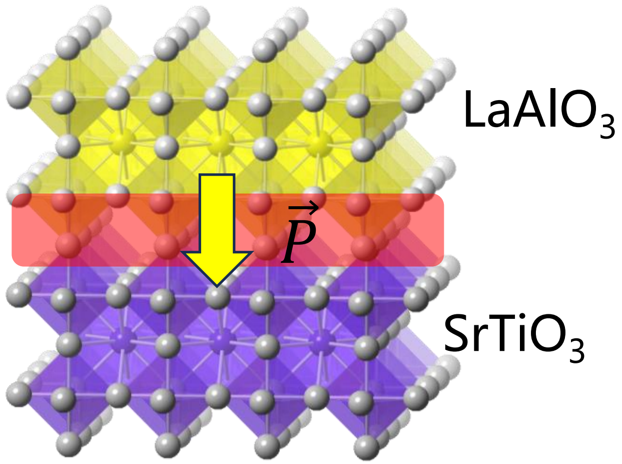
Driven by local chemical or geometrical constraints

➢ Polar metallic Interface: alternative playground

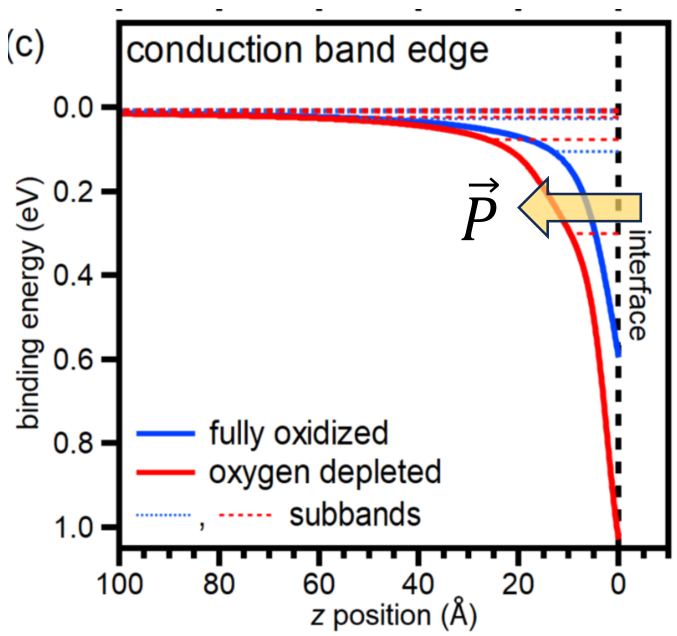
✓ Advantages of interface

- Symmetry breaking is automatic;
- Strain/charge transfer/confinement can stabilize polar distortions while keeping it metallic.

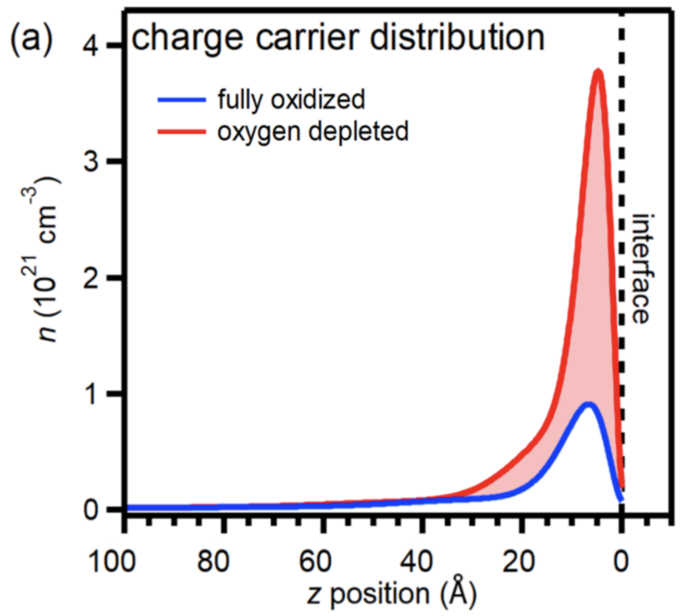
✓ $LaAlO_3/SrTiO_3$ heterostructure



✓ Potential distribution @ interfacial region



✓ Carrier density distribution @ interfacial region



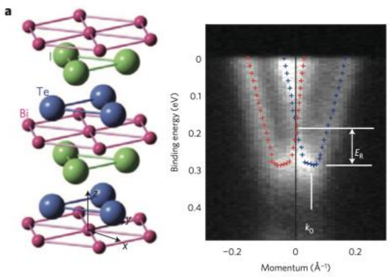
Ohtomo, A. & Hwang, H. Y. Nature 427, 423 (2004). J. Gabel et al., PRB **108**, 045125 (2023)

Research history of Polar Metal

PRL 1965
Anderson&
Blount 提出“铁电金属”概念

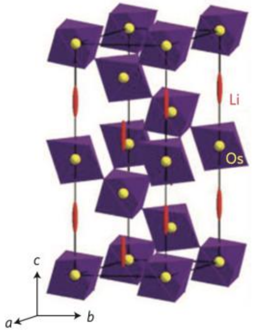
Anderson

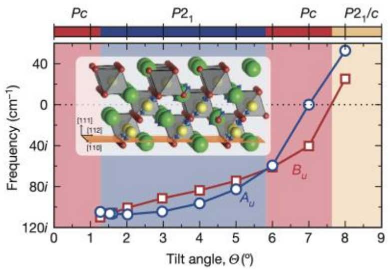
Blount



BiTeI
3m 点群，巨大的
Rashba 自旋劈裂
Nat. Mater. 2011

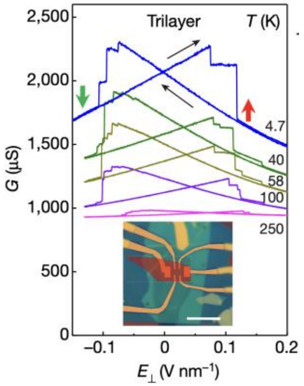
Nat. Mater. 2013
$LiOsO_3$
$R\bar{3}c$ to $R3c$ @ 140 K

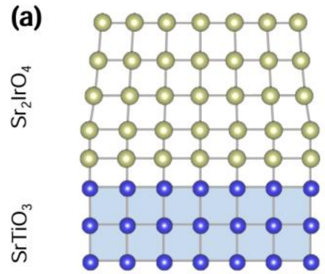




$NdNiO_3$ (111)
衬底约束调控八面体旋转模式，诱导极性
Nature 2016

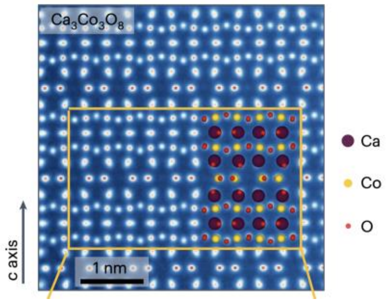
Nature 2018
WTe_2
金属铁电翻转

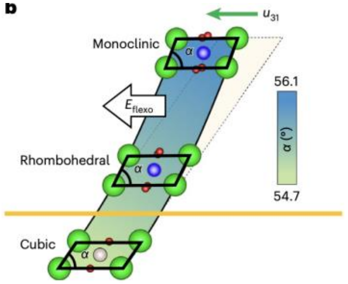




Sr_2IrO_4
应变梯度诱导
极性铁磁
PRL 2024

Nat. Mater. 2024
$Ca_3Co_3O_8$
极性&铁磁金属
≤150K

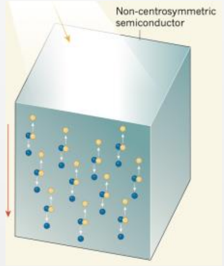




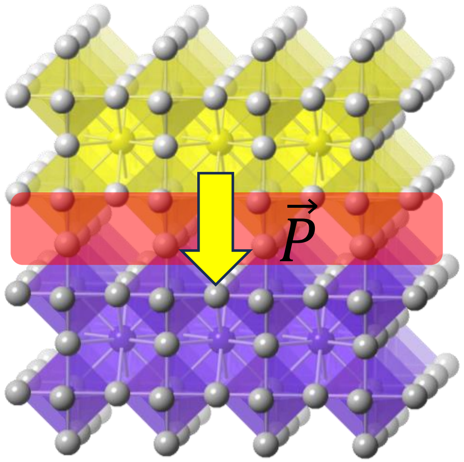
$SrRuO_3$ (111)
剪切应变梯度诱导
极性金属
Nat. Phys. 2024

➢ Rich physics of polar metal

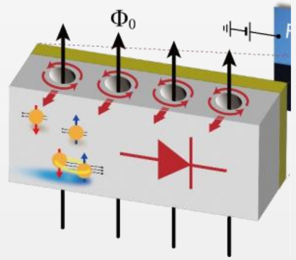
Bulk photovoltaic effect



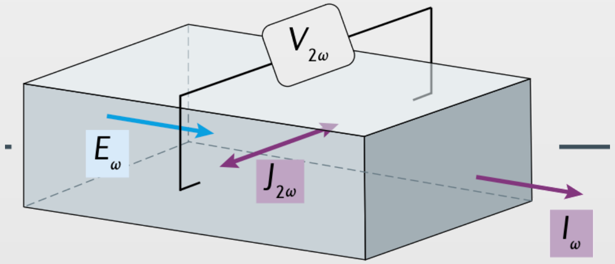
Polar metal



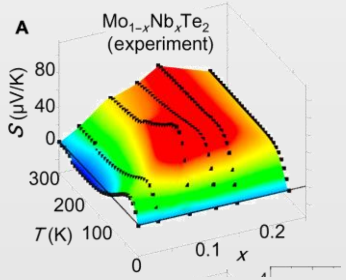
Superconducting diode effect



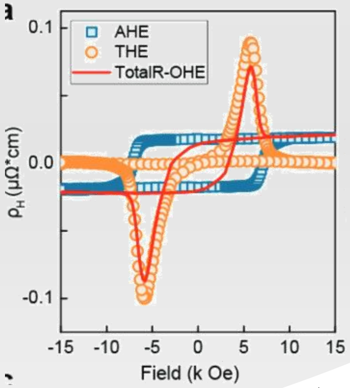
Nonlinear Hall effect



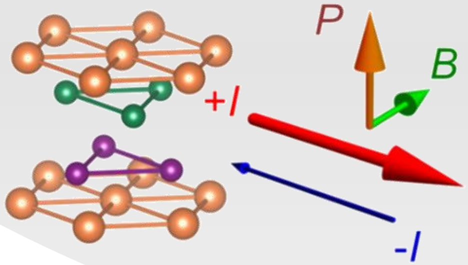
Colossal thermoelectric effect



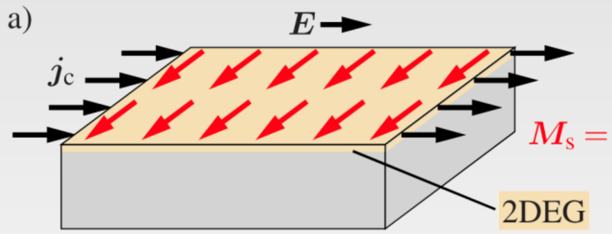
Topological Hall effect



Nonreciprocal transport



Spin-charge interconversion



- ✓ Nature 539, 509(2016)
- ✓ Nature 583, 377 (2020)
- ✓ Nature Nanotech. 9, 851(2014)
- ✓ Rev. Mod. Phys. 89, 025006(2017)

➢ Symmetry breaking induces physical effects

✓ **Inversion symmetry breaking (Polar symmetry)** ⟺ ✓ **Free carriers**

⬇

- ✓ Bulk photovoltaic effect
- ✓ Interfacial charge-spin interconversion
- ✓ Nonlinear Hall effect
- ✓ Nonreciprocal transport
- ✓ Superconducting diode effect
- ✓ Quantum geometry： e.g. Berry curvature diploe

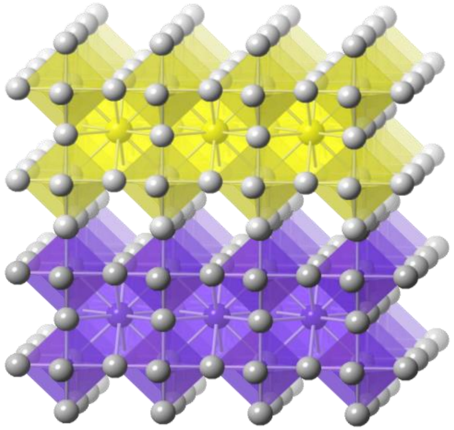
If one can manipulate the symmetry of interface, one can control these effects

➢ Symmetry breaking induces physical effects

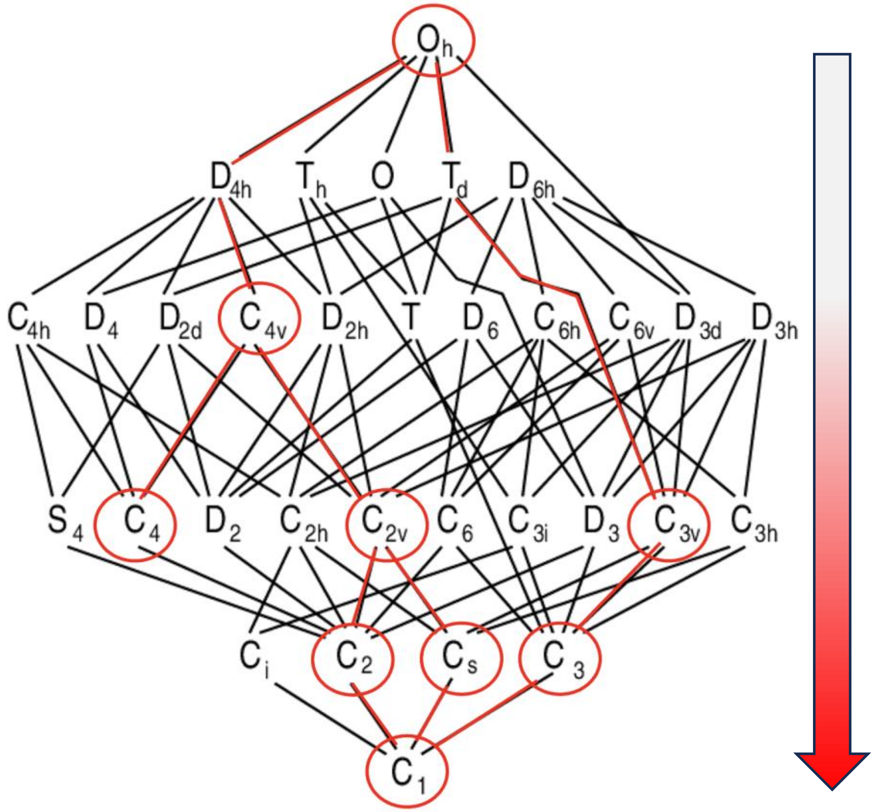
✓ How symmetry breaks @ interface?

① Out-of-plane mirror symmetry is naturally broken

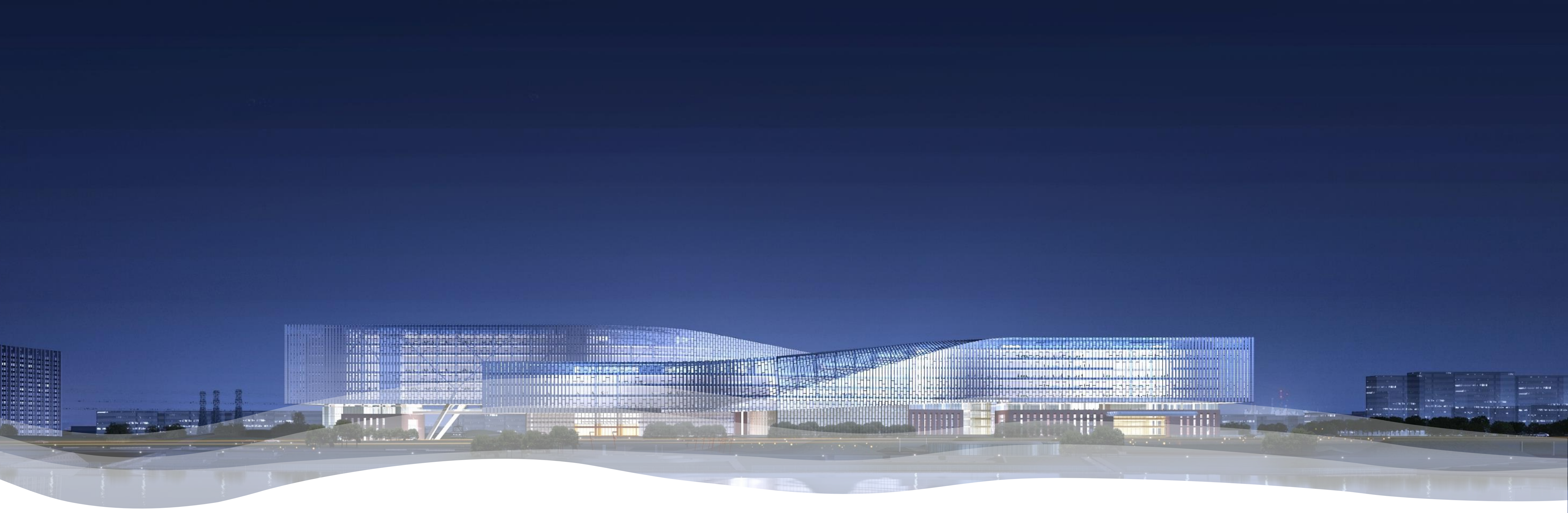
① Symmetry breaking must follow the hierarchy



OOP mirror



- 21 non-centrosymmetry point group
- 10 polar point group

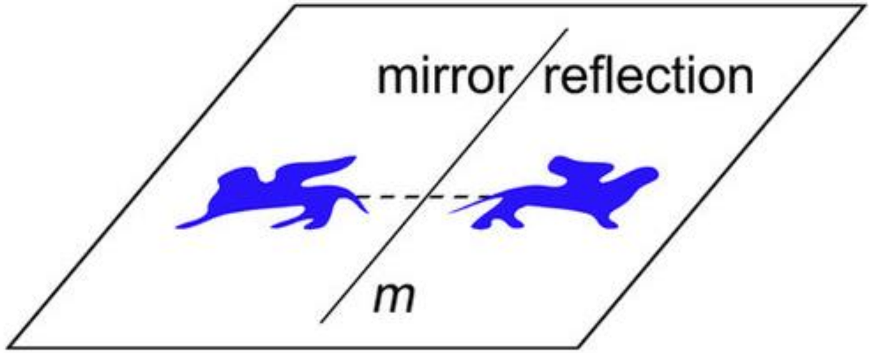


➢ A novel method to modulate the interface symmetry: Orientation

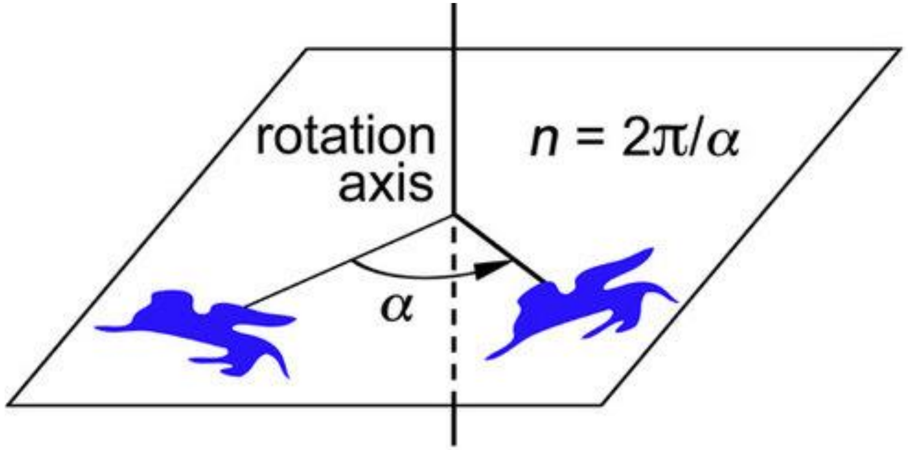
$LaAlO_3/SrTiO_3$ as an example

✓Criteria for controlling interface symmetry：

✓ Mirror symmetry



✓ Rotation symmetry

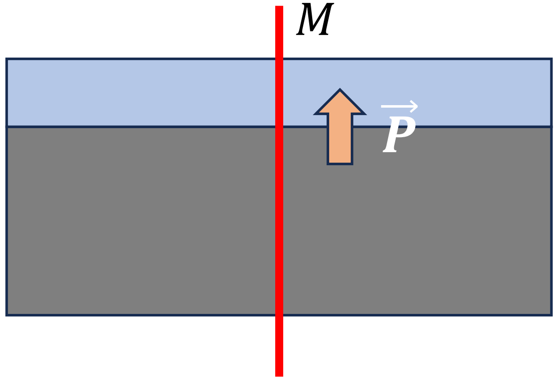


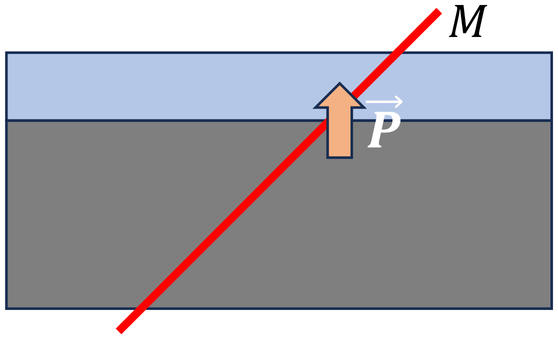
n=1,2,3,4,6

Whether these symmetry elements break or persist at the interface?

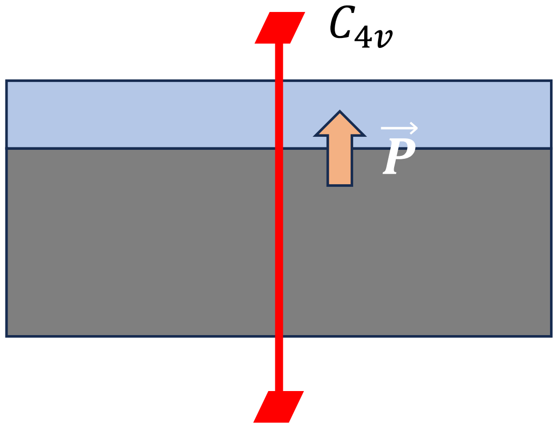
✓Criteria for controlling interface symmetry：

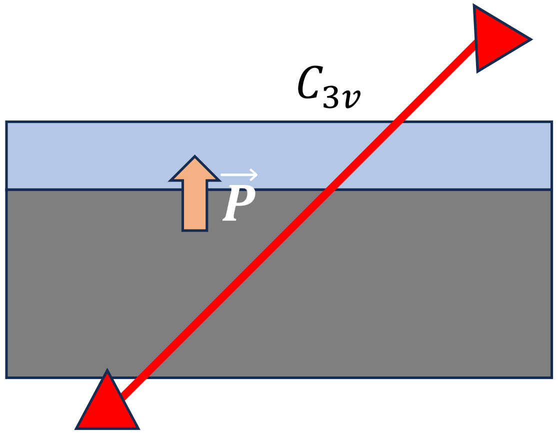
① Mirror symmetry is preserved when it is parallel to the interface's potential gradient, while it breaks when it is at an angle to the potential gradient.





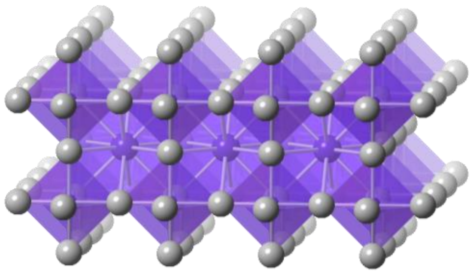
② Rotational symmetry is preserved when its axis is parallel to the interface potential gradient, while it breaks when it is at an angle to the potential gradient.

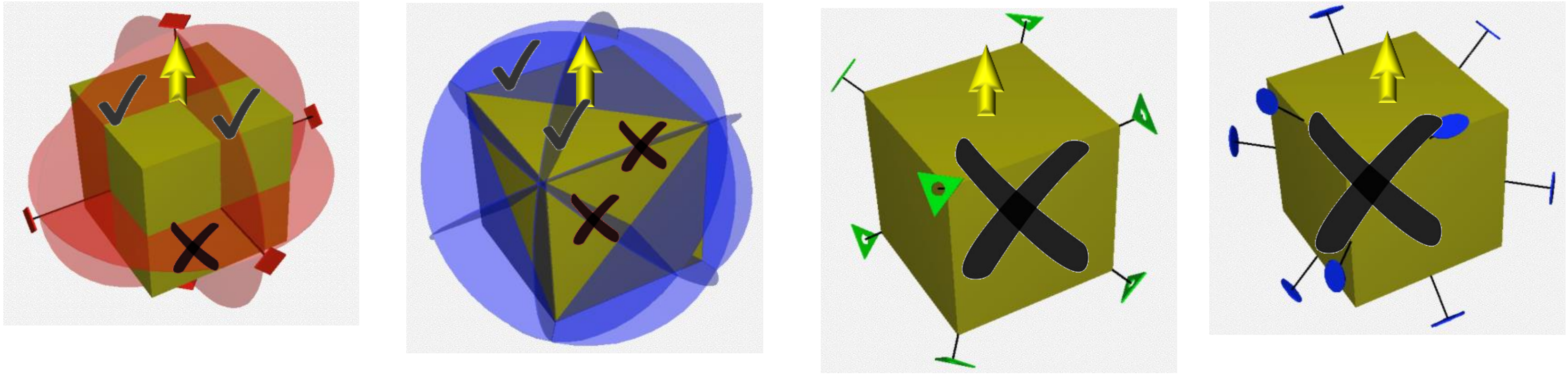




➢ (001)-$LaAlO_3/SrTiO_3$ heterostructure

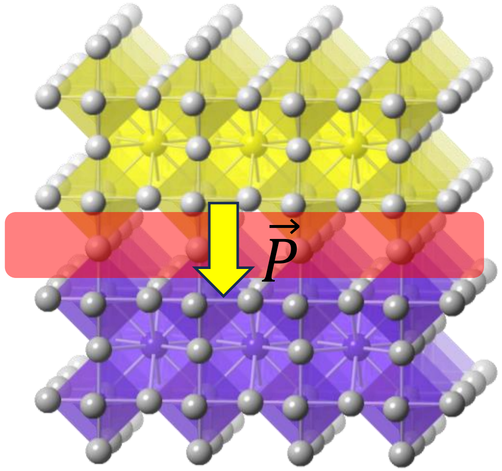
✓ $SrTiO_3$ Crystal: O_h cubic



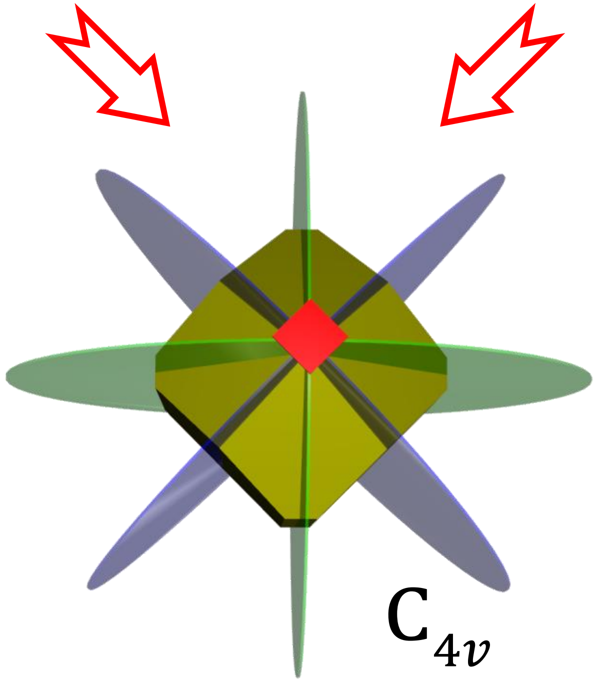


$3\times C_{4h}$ $\quad$ $6\times m_{110}$ $\quad$ $4\times C_{3i}$ $\quad$ $6\times C_2$

$LaAlO_3/SrTiO_3$ heterostructure



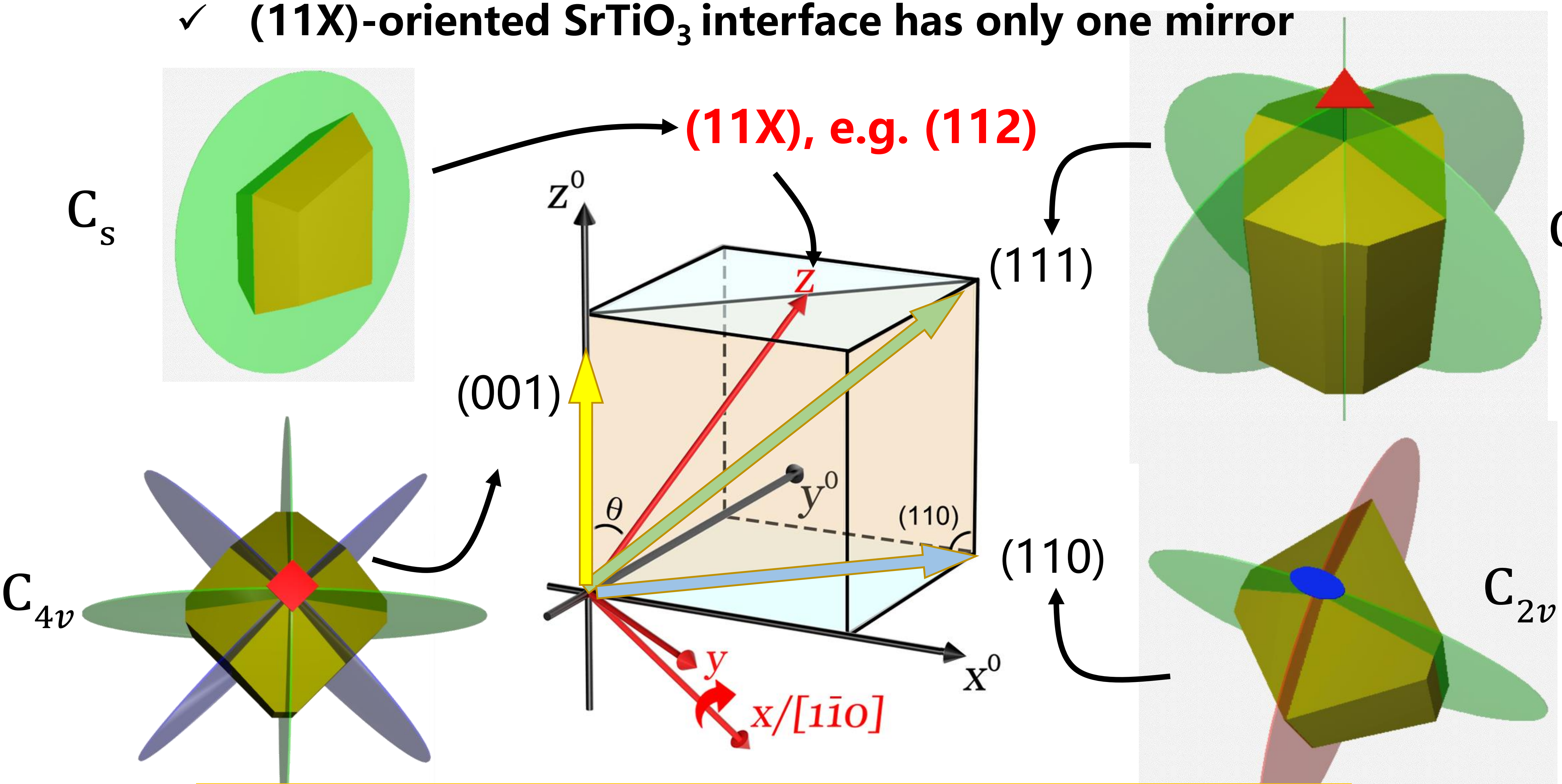
- Conductive
- Polar layer



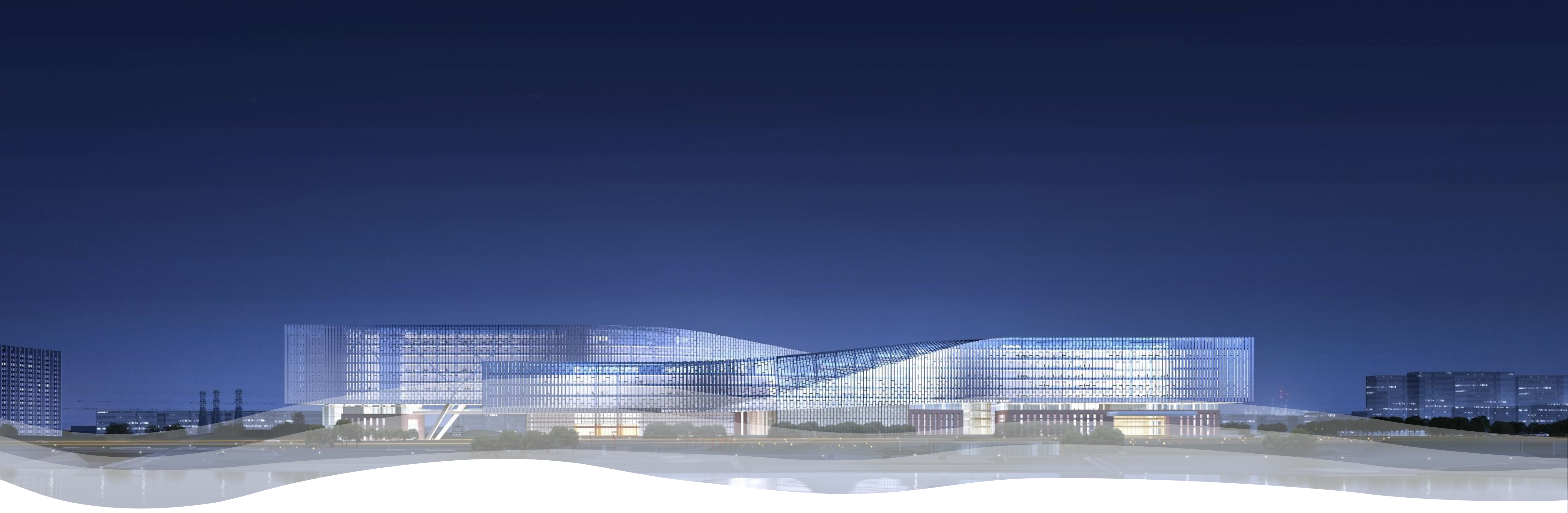
- 4 in-plane mirrors

➢ Interface symmetry modulation via crystal orientation

✓ (11X)-oriented $SrTiO_3$ interface has only one mirror



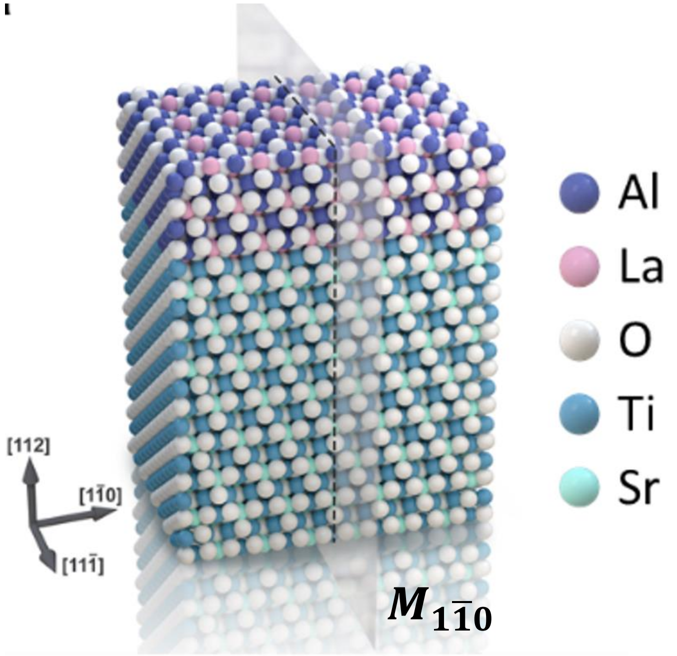
The less mirror planes, the more physical properties.



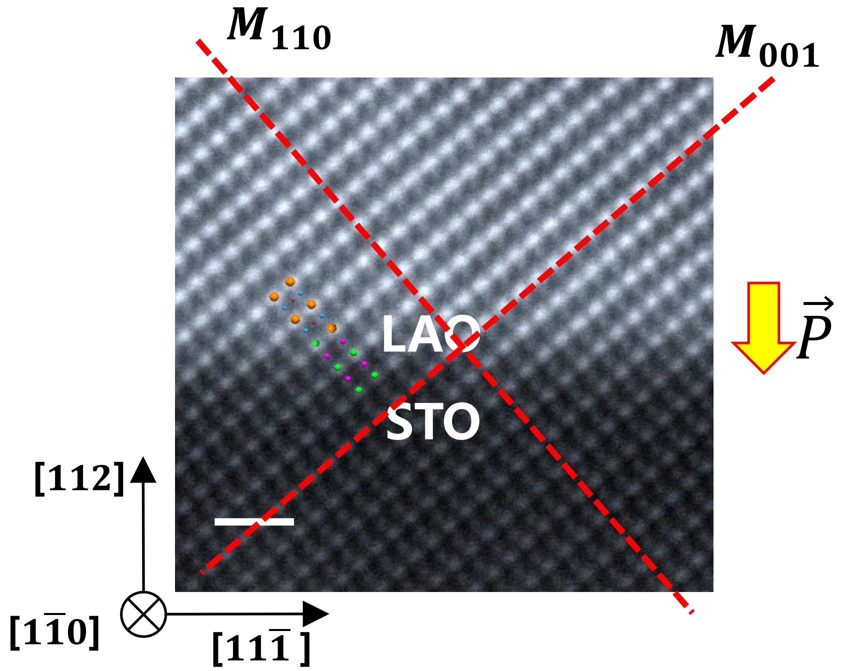
- **(112)-oriented** $LaAlO_3/SrTiO_3$ **interface**

➢ (112)-$LaAlO_3/SrTiO_3$ interfacial atomic structure

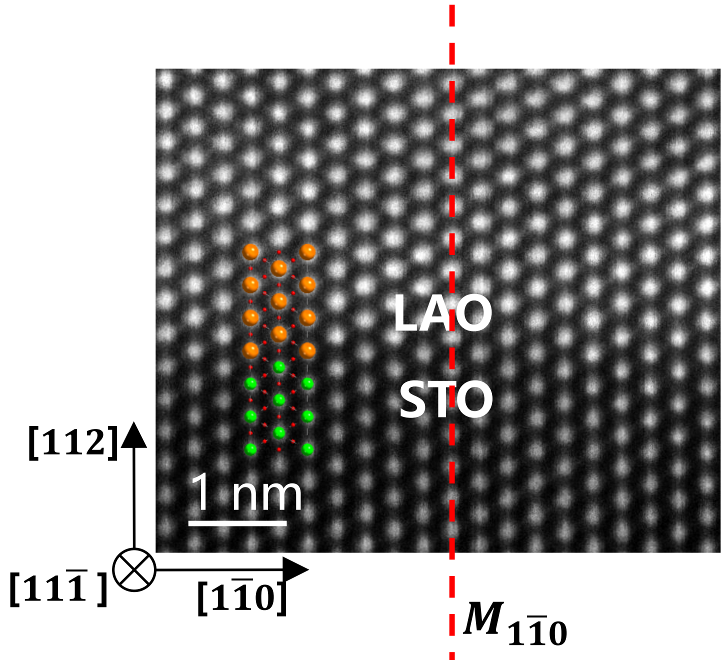
✓ **Schematic of (112)-interface**



✓ **(1$\bar{1}$0) cross section**



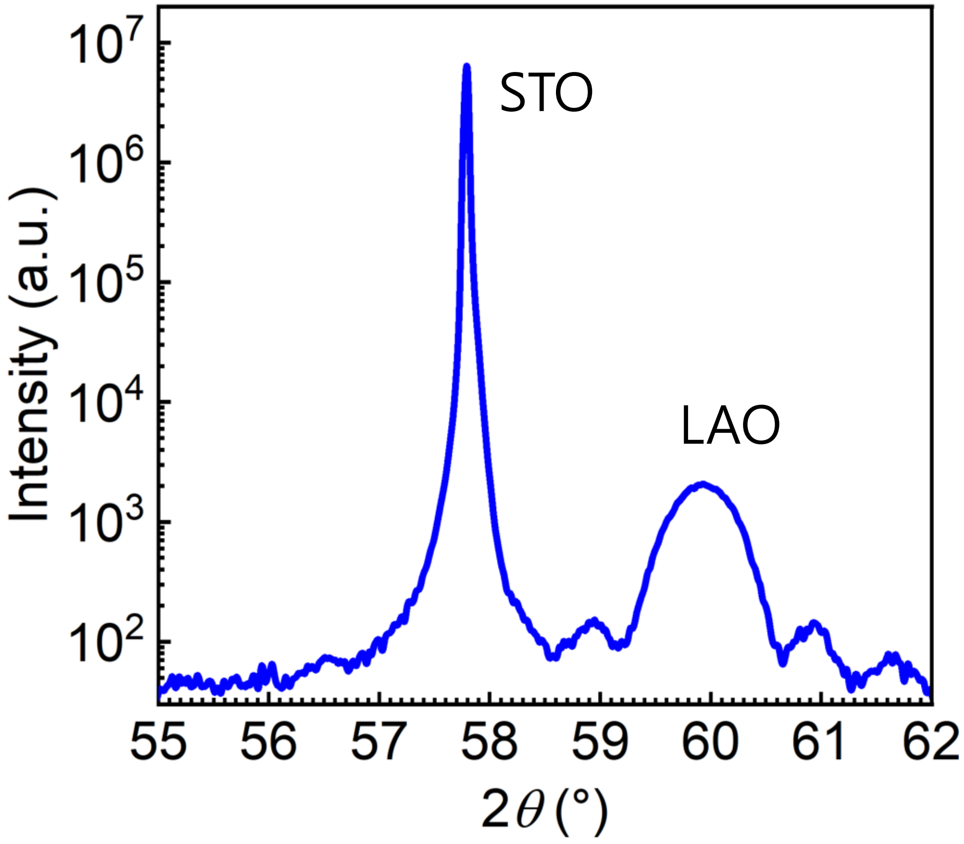
✓ **(11$\bar{1}$) cross section**



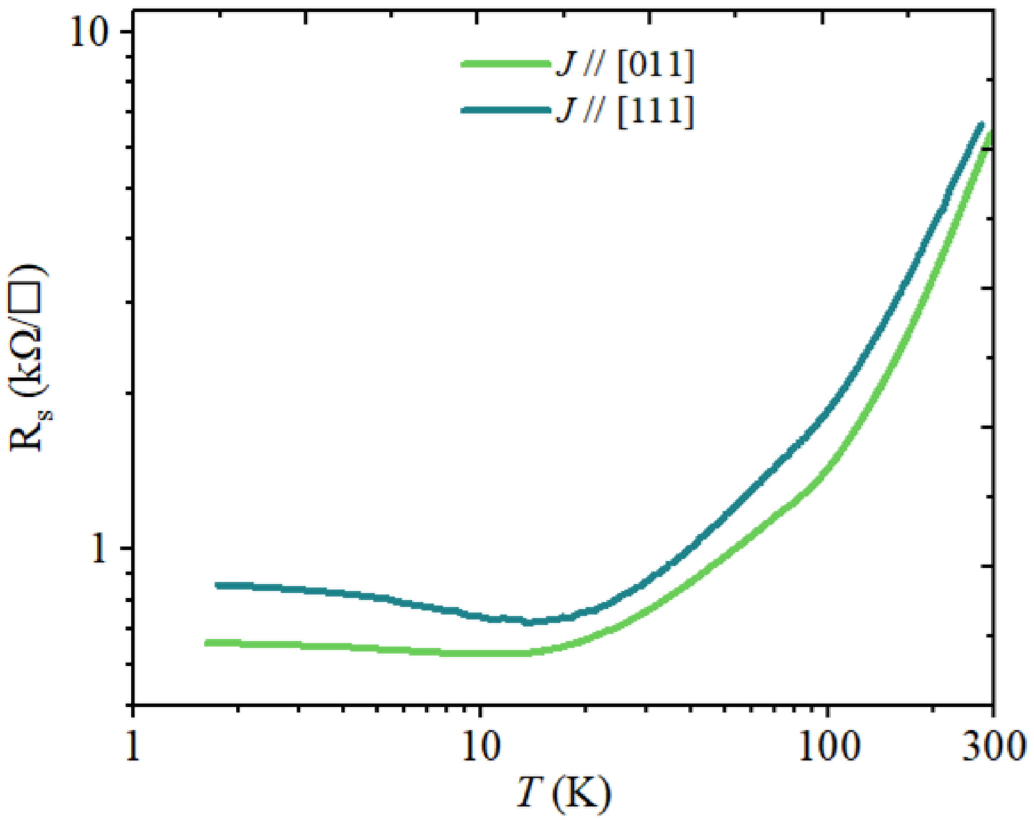
✓ **Only $M_{1\bar{1}0}$ mirror preserves, the point group becomes C_s**

➢ (112)-$LaAlO_3/SrTiO_3$ interfacial metallicity

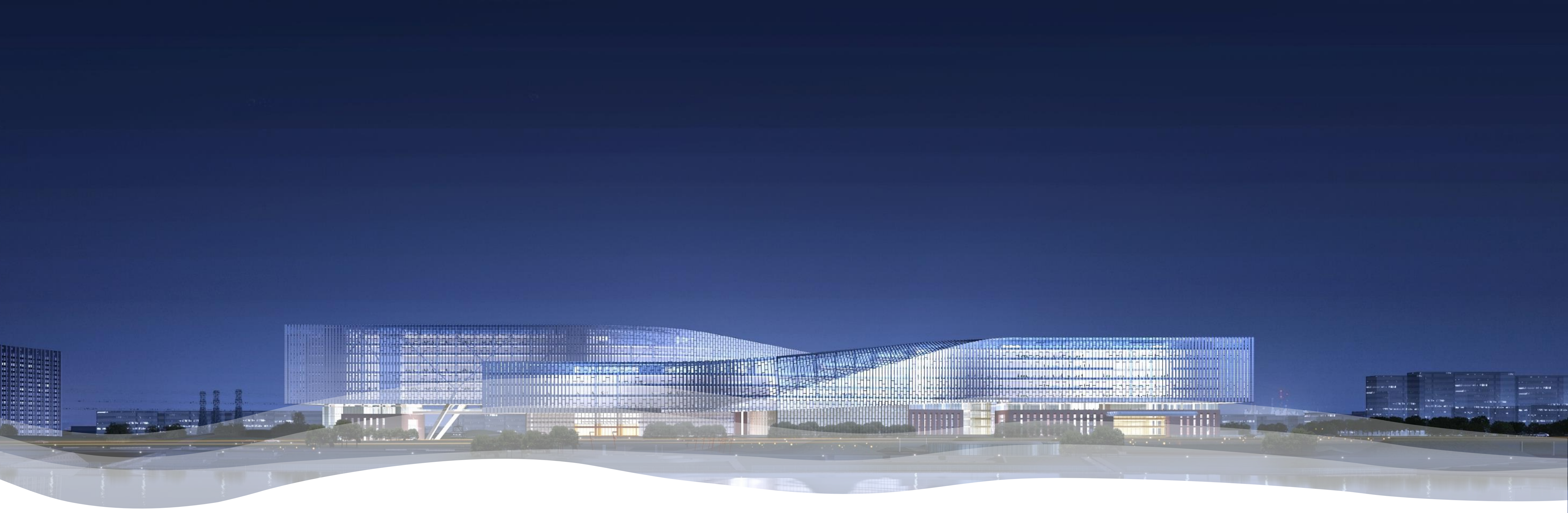
✓ XRD spectrum



✓ Resistance vs temperature

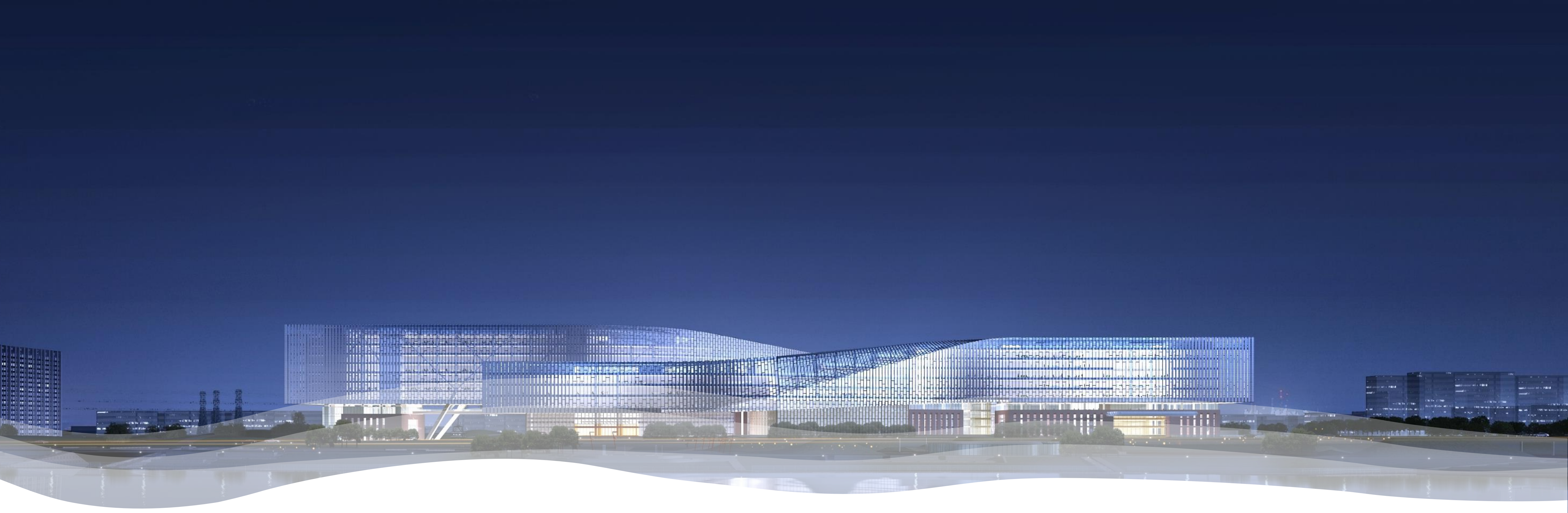


Metallic conduction with in-plane anisotropy



➢ **Novel properties** @ (112)-$LaAlO_3/SrTiO_3$

- ✓ Circular photogalvanic effect
- ✓ Charge-magnetization conversion
- ✓ Nonlinear Hall effect



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- ✓ Nonlinear Hall effect

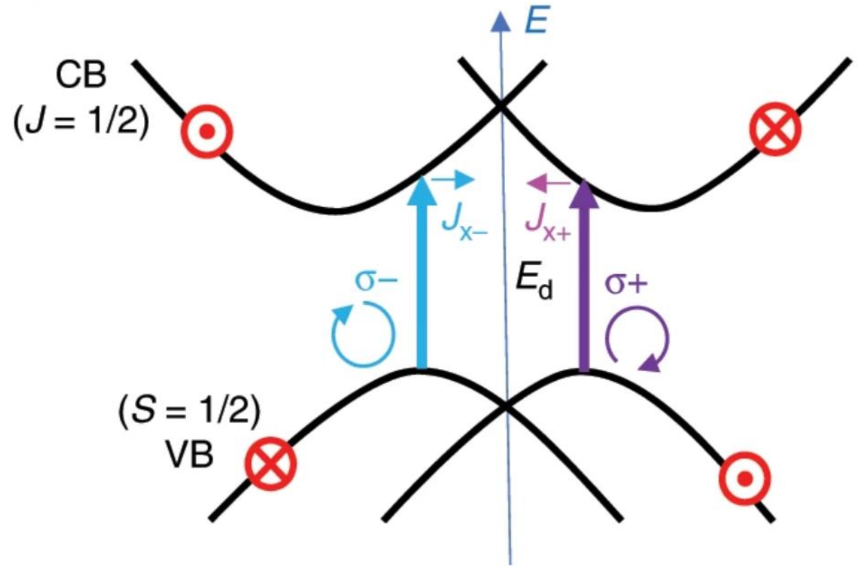
Circular photogalvanic effect（CPGE）to verify the symmetry

- **Circular photogalvanic effect:**

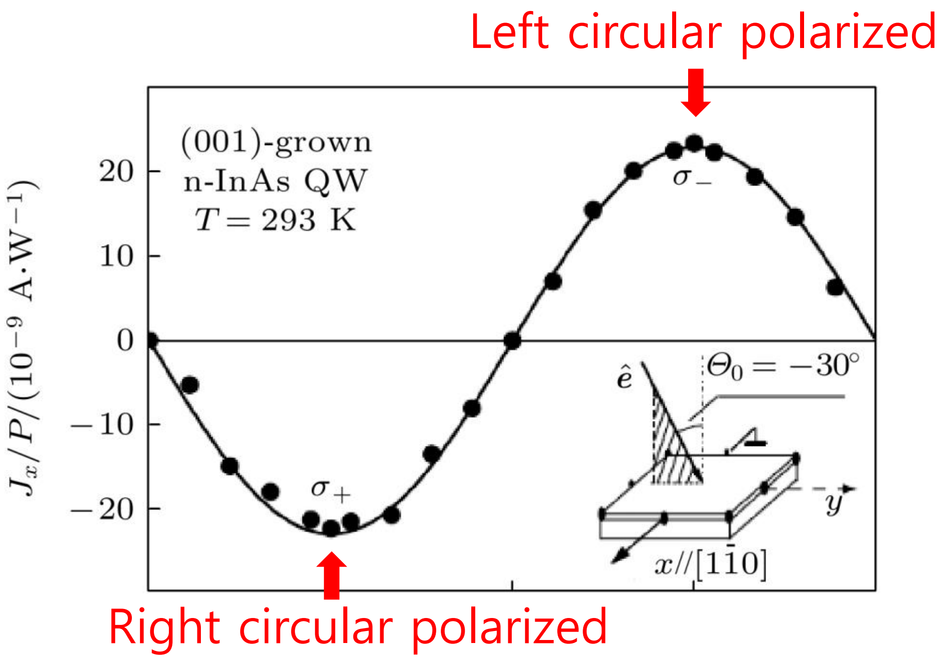
$$J_i = \gamma_{ij}[\vec{e} \times \vec{e}^*]_j$$

γ_{ij}: CPGE 2nd rank tensor; $\vec{e}$: light's electric vector

- Optical selection rule



- Photocurrent dependens on circular polarization



STURMAN et al., The Photovoltaic and Photorefractive Effects in Non-centrosymmetric Materials 1992

➢ Circular photogalvanic effect（CPGE）to verify the symmetry

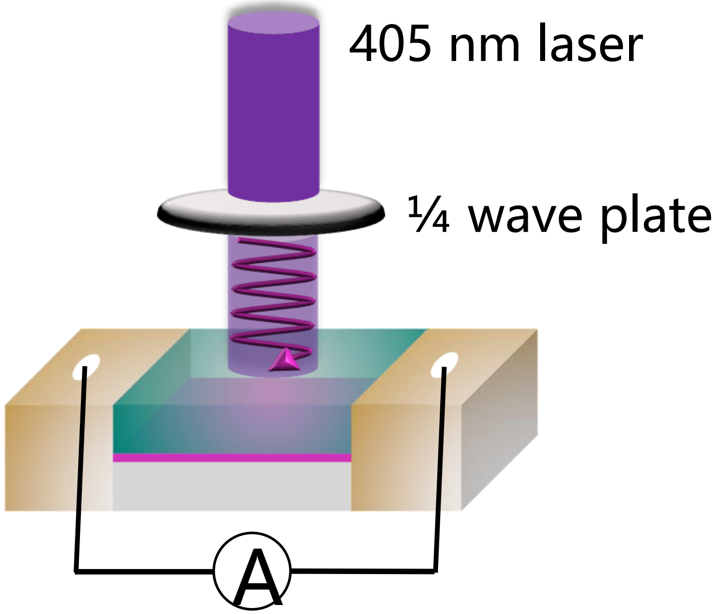
✓ **Circular photogalvanic effect:**

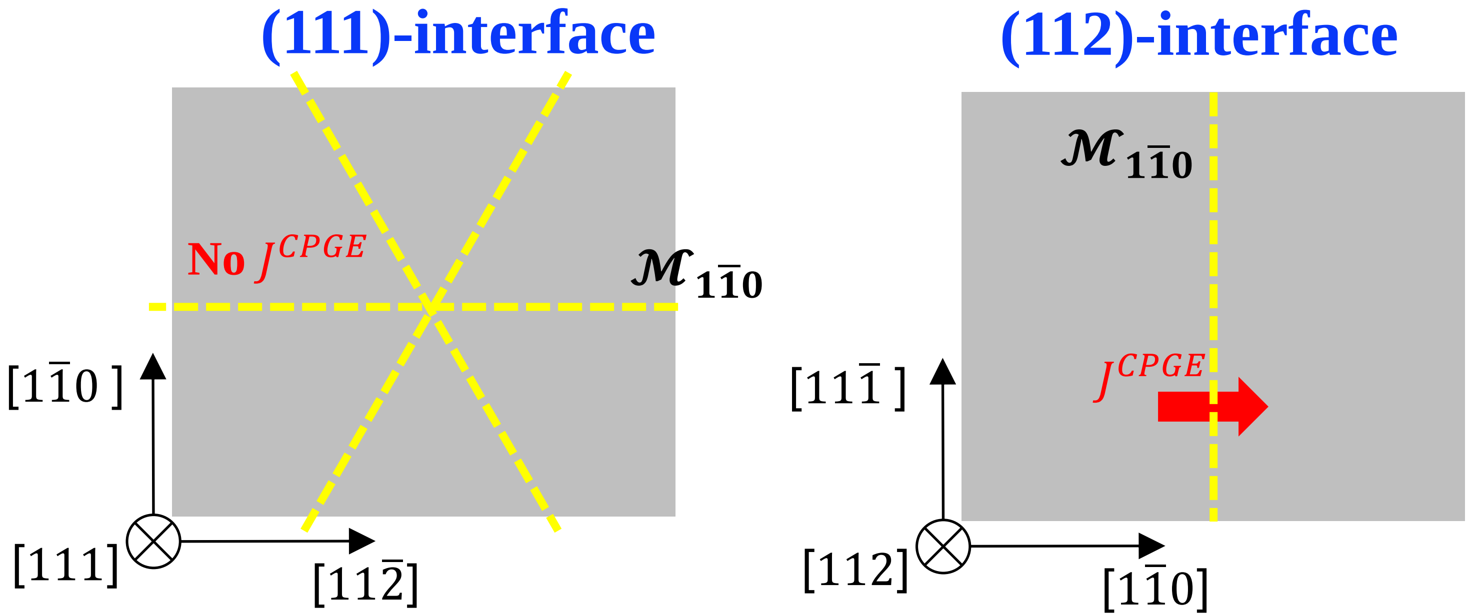
$$J_i = \gamma_{ij}[\vec{e} \times \vec{e}^*]_j$$

γ_{ij}: CPGE 2nd rank tensor； $\vec{e}$：light's electric vector

✓ **Main character**:

When the light is normal to the interface, only **photocurrent in the direction perpendicular to the mirror** can be generated.





STURMAN et al., The Photovoltaic and Photorefractive Effects in Non-centrosymmetric Materials 1992

➢ Circular photogalvanic effect (CPGE) @ (111)-$LaAlO_3/SrTiO_3$

✓ **Circular photogalvanic effect:**

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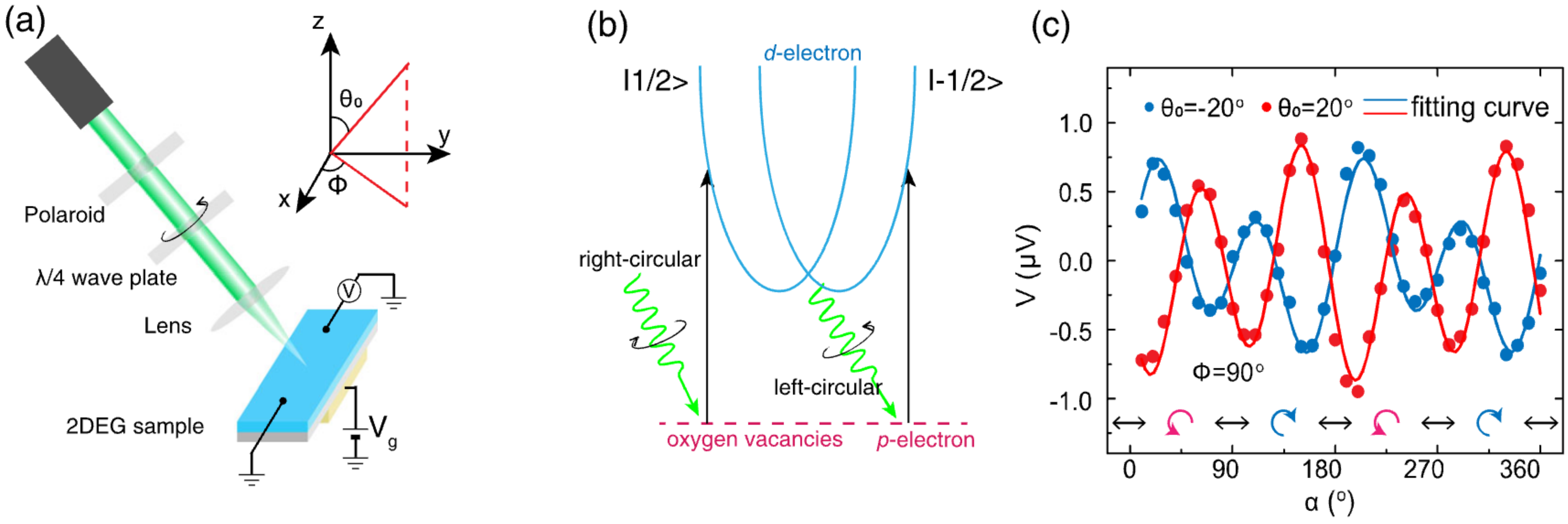


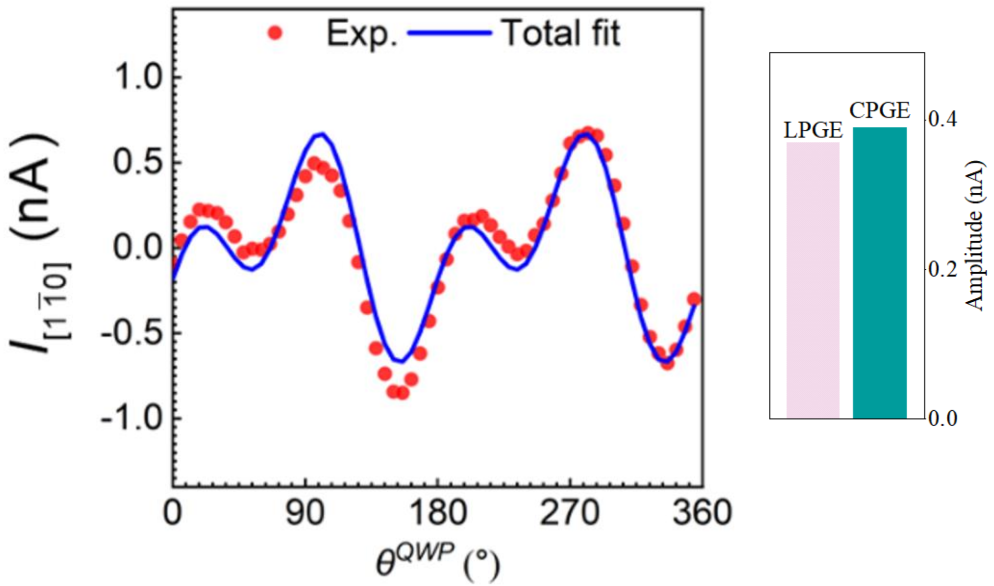
Tilted illumination is required @ (111)-LAO/STO interface

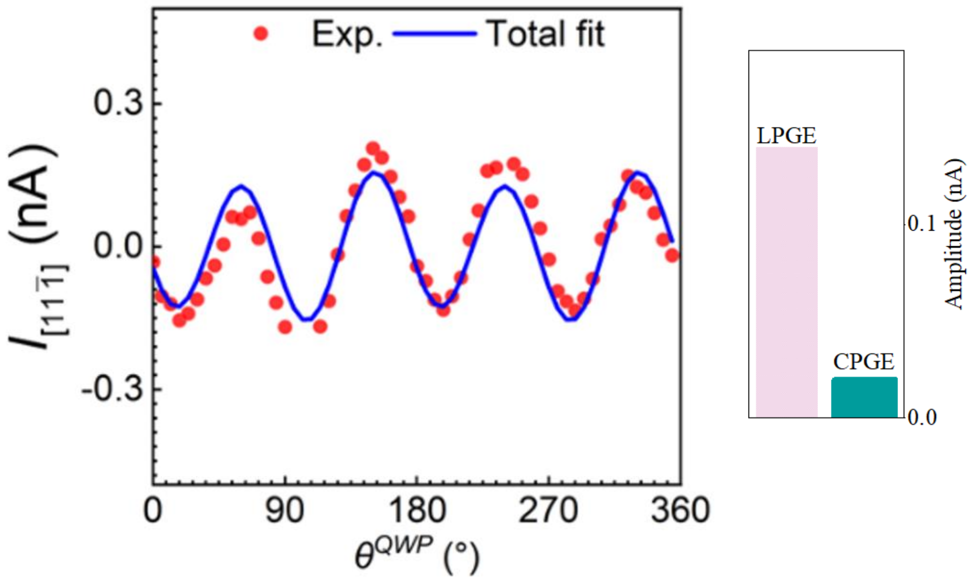
PRL 128, 187401 (2022)

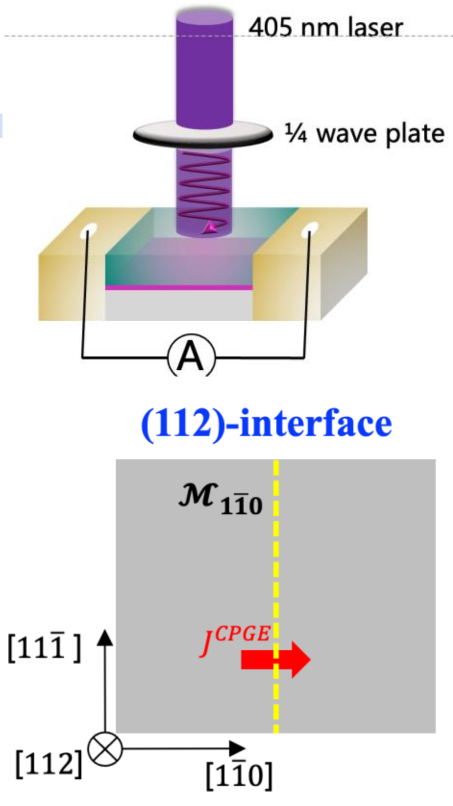
➢ Circular photogalvanic effect @ (112)-$LaAlO_3/SrTiO_3$

✓ $[1\bar{1}0]$ photocurrent

✓ $[11\bar{1}]$ photocurrent

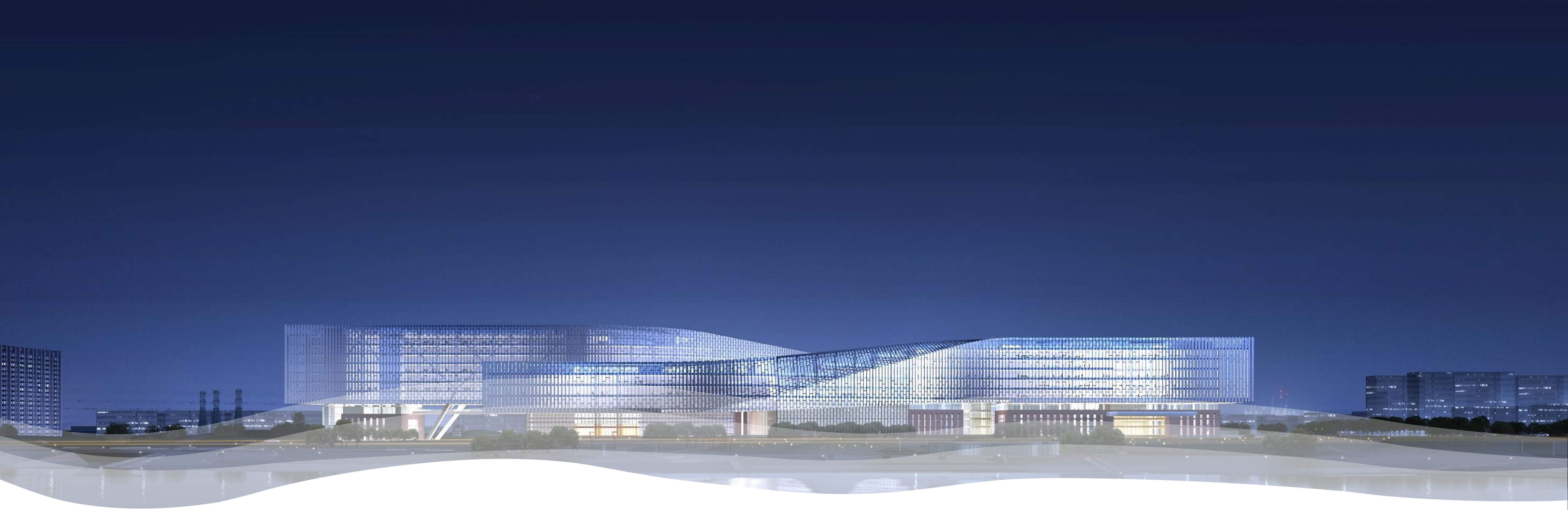






$$J = C\sin(2\theta^{QWP}) + L\sin(4\theta^{QWP})$$

✓ **(112)-** LAO/STO interface has only one mirror symmetry



➢ (112)-$LaAlO_3/SrTiO_3$ **novel properties**

✓ Circular photogalvanic effect

✓ Charge-magnetization conversion

✓ Nonlinear Hall effect

➢ Charge-Magnetism conversion @ polar metal

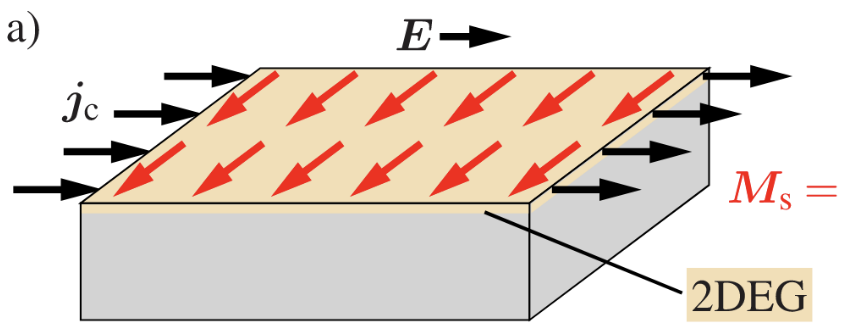
✓ (Inverse) Spin-galvanic effect (*Rashba–Edelstein effect*):

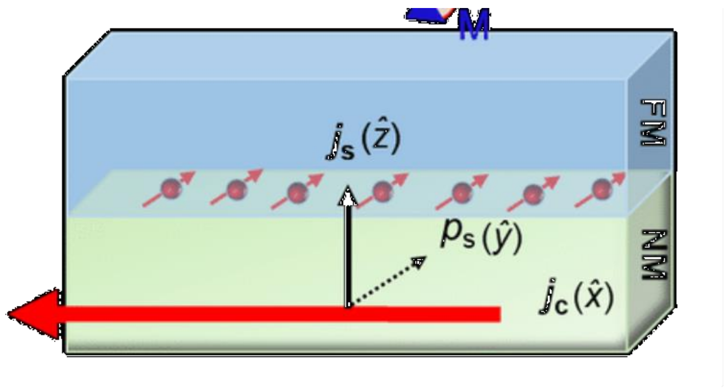
In polar metal, electron conduction induces nonequilibrium magnetism (spin or/and orbital origin)

$$M_i = \beta_{ij} J_j$$

M_i: magnetic moment, β_{ij}: tensor, J_j: current density

- ✓ Current induced spin polarization
- ✓ Spin orbital torque





A. Johansson et al., Phys. Rev. Research 3, 013275 (202

➢ Charge-Magnetism conversion @ polar metal

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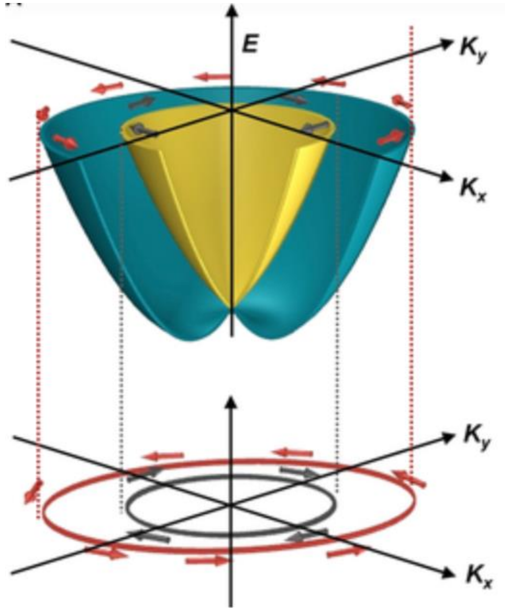
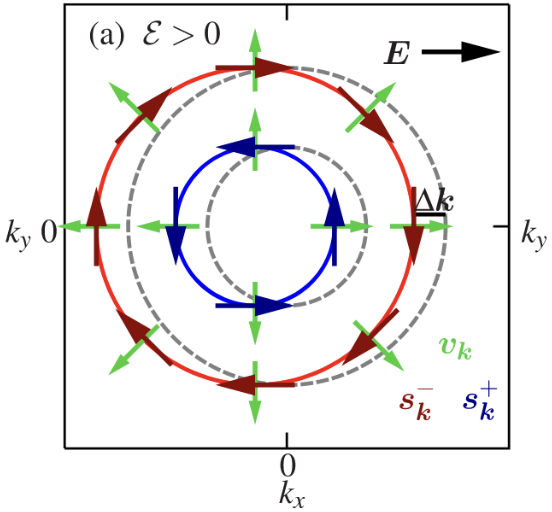
In polar metal, electron conduction induces nonequilibrium magnetism (spin or/and orbital origin)

$$M_i = \beta_{ij} J_j$$

M_i: magnetic moment, β_{ij}: tensor, J_j: current density



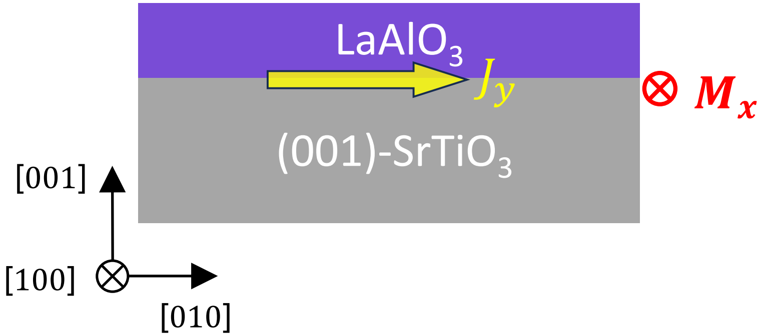


Applied voltage shifts Fermi surface, leading to nonequilibrium spin/orbital distribution

A. Johansson et al., Phys. Rev. Research 3, 013275 (202

➢ Charge-magnetism conversion @ $LaAlO_3/SrTiO_3$ Interface

✓ (001)、(110)、(111)-$LaAlO_3/SrTiO_3$ Interface



$$M_i = \begin{pmatrix} M_x \\ M_y \\ M_z \end{pmatrix} = \begin{pmatrix} 0 & \gamma_{12} & 0 \\ -\gamma_{12} & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} J_x \\ J_y \\ 0 \end{pmatrix} = \begin{pmatrix} \gamma_{12} J_y \\ -\gamma_{12} J_x \\ 0 \end{pmatrix}$$

❖ Current only induces **in-plane** magnetic moment, due to ≥2 in-plane mirror planes

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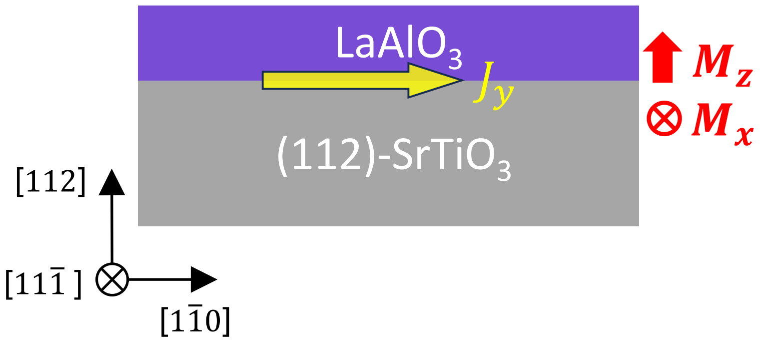
✓ (112)-$LaAlO_3/SrTiO_3$ Interface

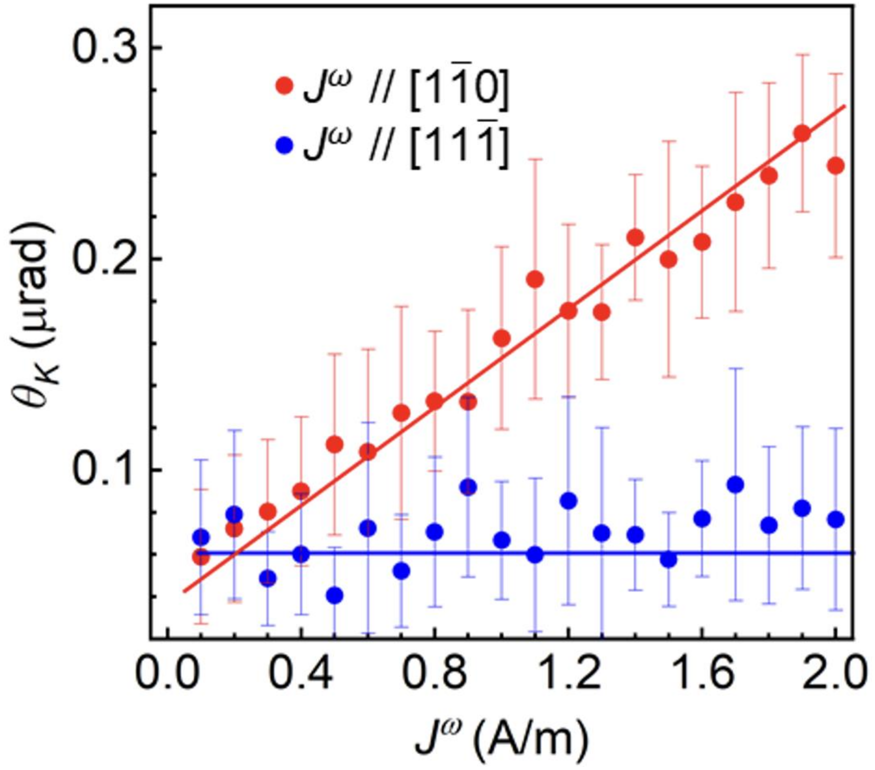


$$M_i = \begin{pmatrix} M_x \\ M_y \\ M_z \end{pmatrix} = \begin{pmatrix} 0 & \gamma_{12} & 0 \\ \gamma_{21} & 0 & \gamma_{23} \\ 0 & \gamma_{32} & 0 \end{pmatrix} \begin{pmatrix} J_x \\ J_y \\ 0 \end{pmatrix} = \begin{pmatrix} \gamma_{12} J_y \\ \gamma_{21} J_x \\ \gamma_{32} J_y \end{pmatrix}$$

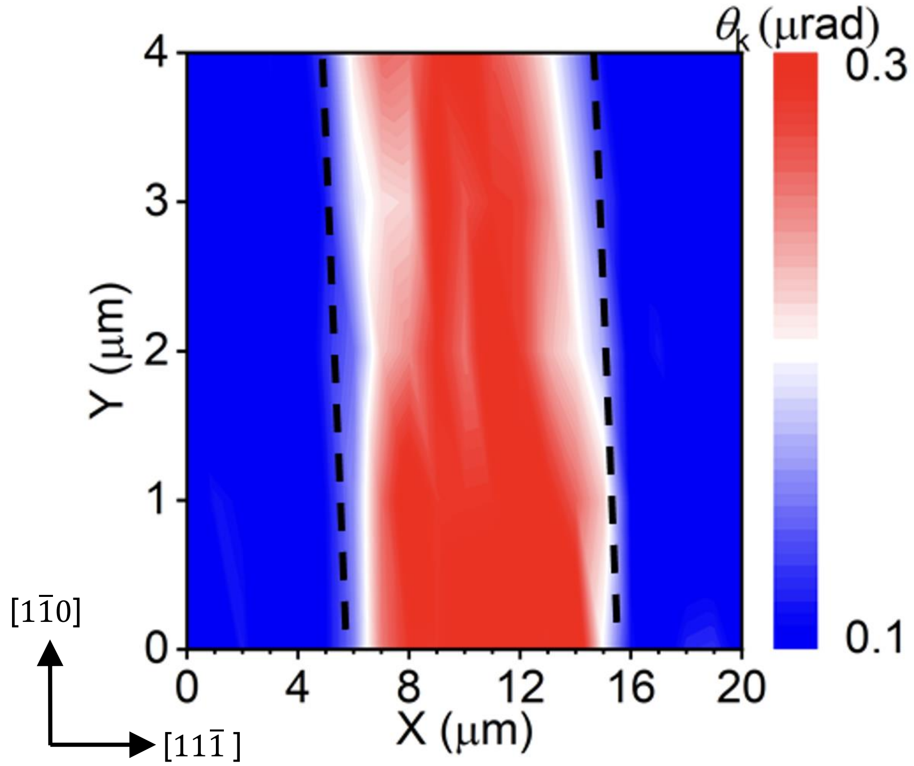
❖ Current can induce both **in-plane and out-of-plane** magnetic moment

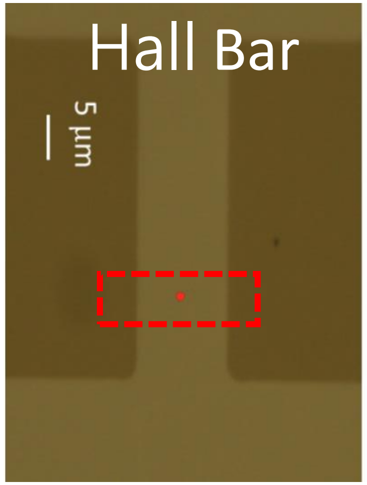
➢ Charge-Magnetism conversion @ (112)-$LaAlO_3/SrTiO_3$

✓ Polar MOKE signal vs input current



✓ Space resolved polar MOKE signal

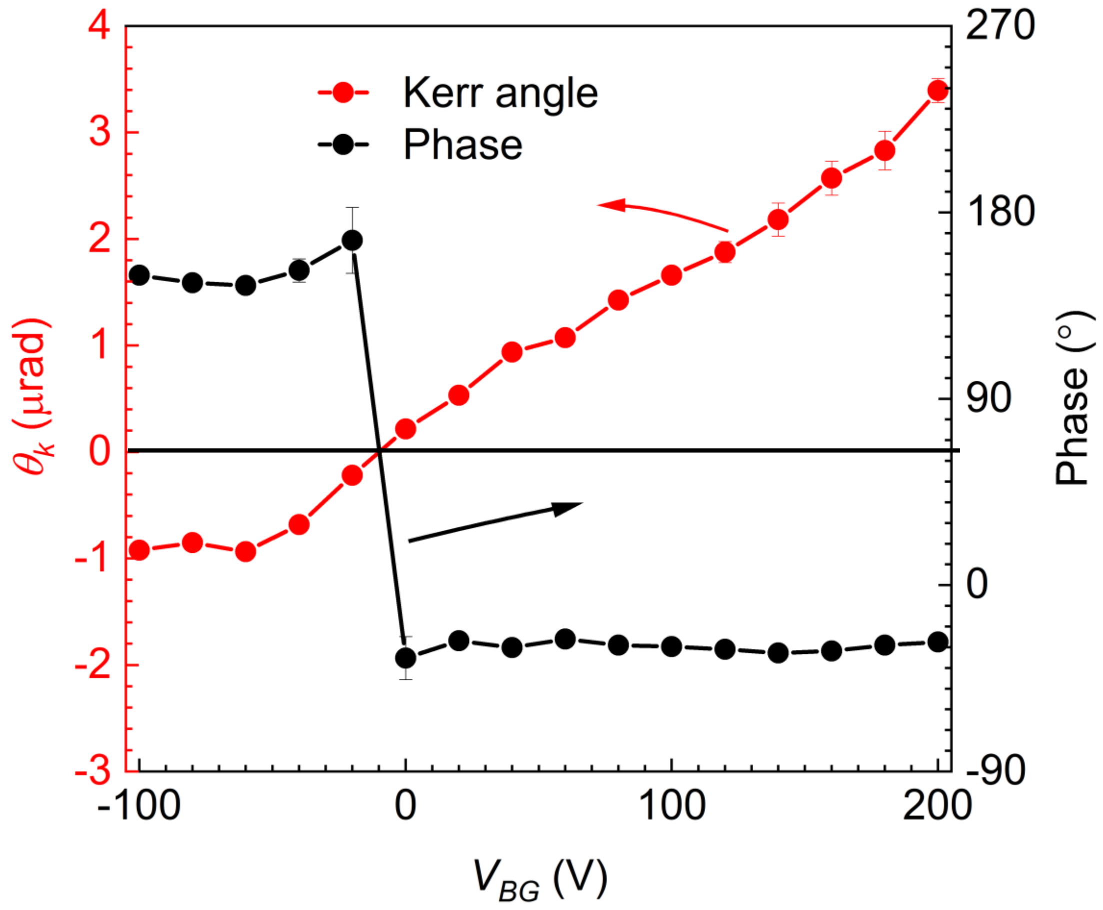


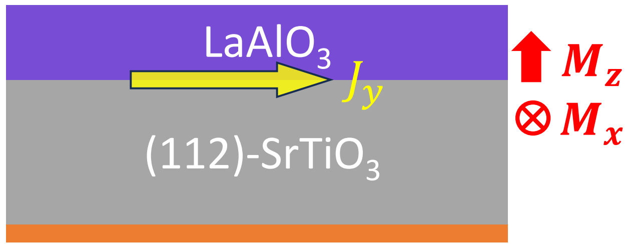


✓ Current along [1$\bar{1}$0] induces out-of-plane magnetization

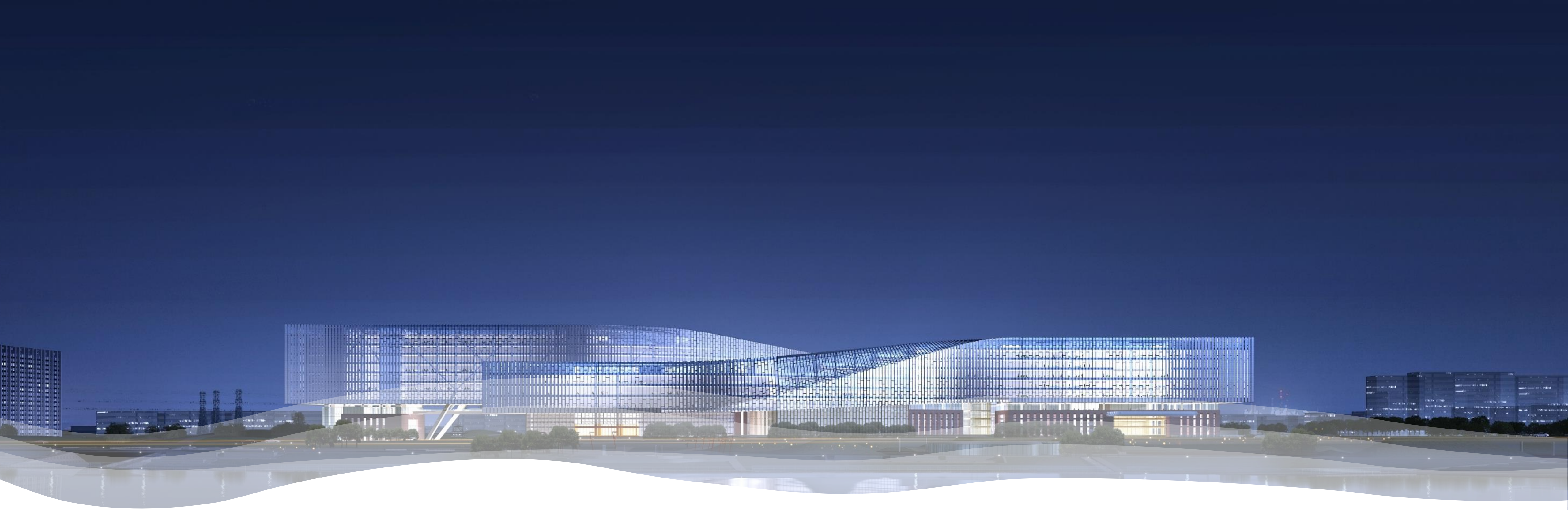
➢ Field effect on interfacial charge-magnetization conversion

✓ Polar MOKE rotation as a function of back gate





✓ **The magnitude and direction of the induced out-of-plane magnetic moment can be controlled by field effects.**



➢ (112)-$LaAlO_3/SrTiO_3$ **novel properties**

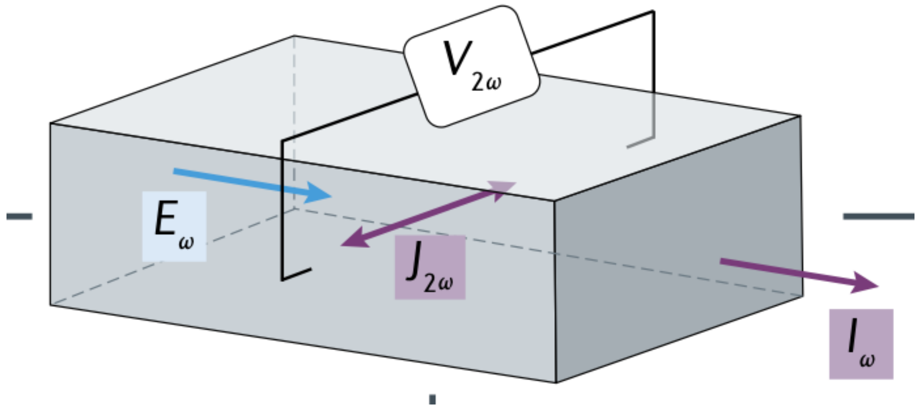
- ✓ Circular photogalvanic effect
- ✓ Charge-magnetization conversion
- ✓ Nonlinear Hall effect

➢ Nonlinear Hall Effect

✓ Hall current driven by longitudinal bias:

$$J_a = \sigma_{ab} E_b + \chi_{abc} E_b E_c + \dots,$$

σ_{ab} ($a \neq b$) : linear Hall conductivity
χ_{abc} :nonlinear conductivity tensor



Non-centrosymmetric materials

Time reversal symmetry

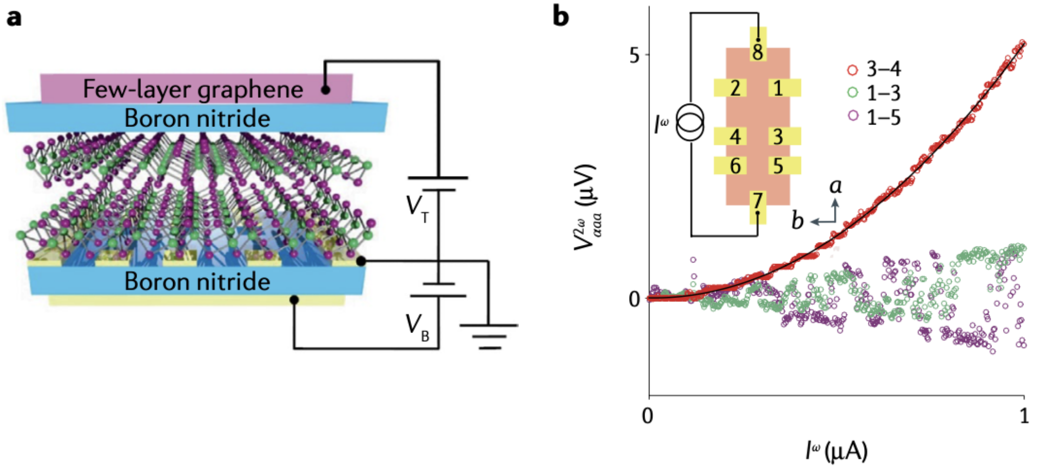
PRL **115**, 216806 (2015) PHYSICAL REVIEW LETTERS week ending 20 NOVEMBER 2015

LETTER

https://doi.org/10.1038/s41586-018-0807-6

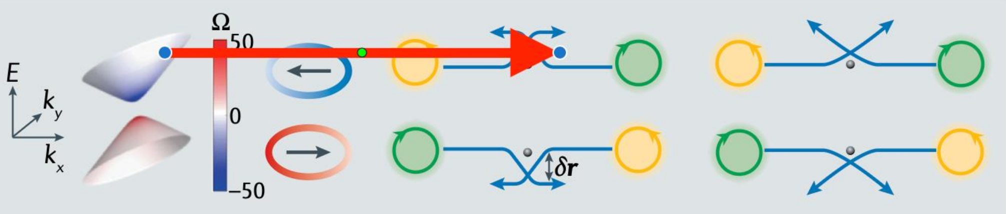
Observation of the nonlinear Hall effect under time-reversal-symmetric conditions

Qiong Ma[1,13], Su-Yang Xu[1,13], Huitao Shen[1,13], David MacNeill[1], Valla Fatemi[1], Tay-Rong Chang[2], Andrés M. Mier Valdivia[1], Sanfeng Wu[1], Zongzheng Du[3,4,5], Chuang-Han Hsu[6,7], Shiang Fang[8], Quinn D. Gibson[9], Kenji Watanabe[10], Takashi Taniguchi[10], Robert J. Cava[9], Efthimios Kaxiras[8,11], Hai-Zhou Lu[3,4], Hsin Lin[12], Liang Fu[1], Nuh Gedik[1]* & Pablo Jarillo-Herrero[1]*



➢ Nonlinear Hall Effect: contributions

	a Intrinsic	b Side-jump		c Skew-scattering	
Mechanisms					



	a Intrinsic	b Side-jump		c Skew-scattering	
Relevant quantities	Berry curvature dipole	Anomalous velocity	Anomalous distribution	Gaussian disorder	Non-Gaussian disorder
Disorder dependence	$(n_iV_0^2)^{-1}$	$(n_iV_0^2)^{-1}$	$(n_iV_0^2)^{-1}$	$(n_iV_0^2)^{-1}$	$\frac{n_iV_1^3}{(n_iV_0^2)^2}$
Symmetry constraints	$\varepsilon_{abd}T_{cd}+\varepsilon_{acd}T_{bd}$	$T_{abc}+T_{acb}$		$T_{abc}+T_{acb}$	

✓ Berry curvature Diploe

$$D_{ab}=\int \frac{d^n\mathbf{k}}{(2\pi)^n}(\partial_a\Omega_b)f_0,$$

$$\chi_{abc}=-\varepsilon_{adc}\frac{e^3\tau}{2(1+i\omega\tau)}D_{ab}$$

*Appears in materials with no more than one mirror symmetry (Cs or C1)

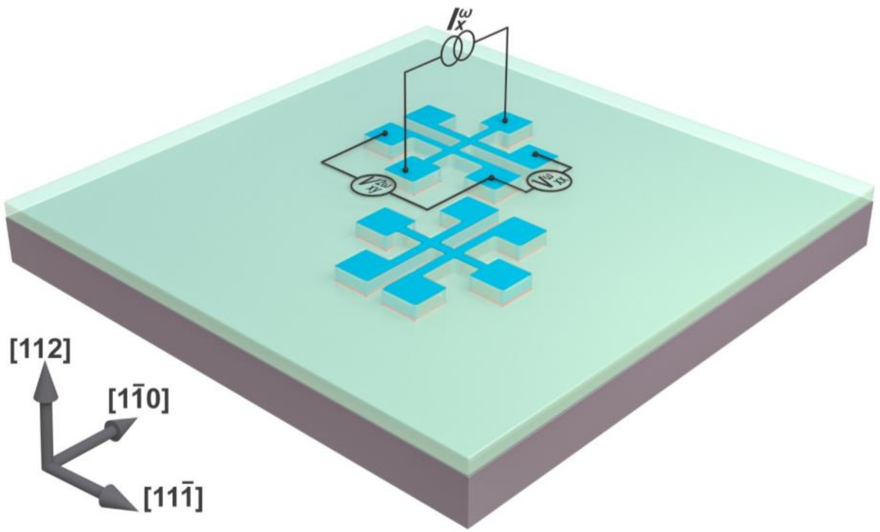
(112)-$LaAlO_3/SrTiO_3$ interface is Cs

Very few conductive materials are of Cs or C1 symmetry !

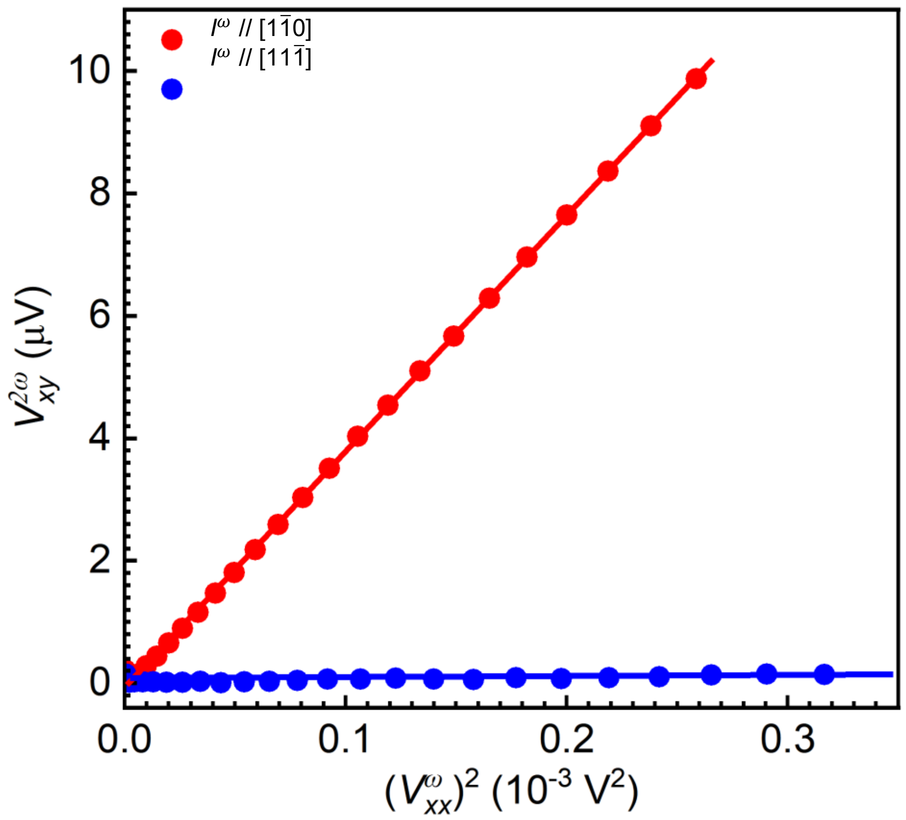
Z. Z. Du et al., Nat. Rev. Phys. 3, 744 (2021)

➢ Nonlinear Hall effect of (112)-$LaAlO_3/SrTiO_3$ interface

✓ Device geometry



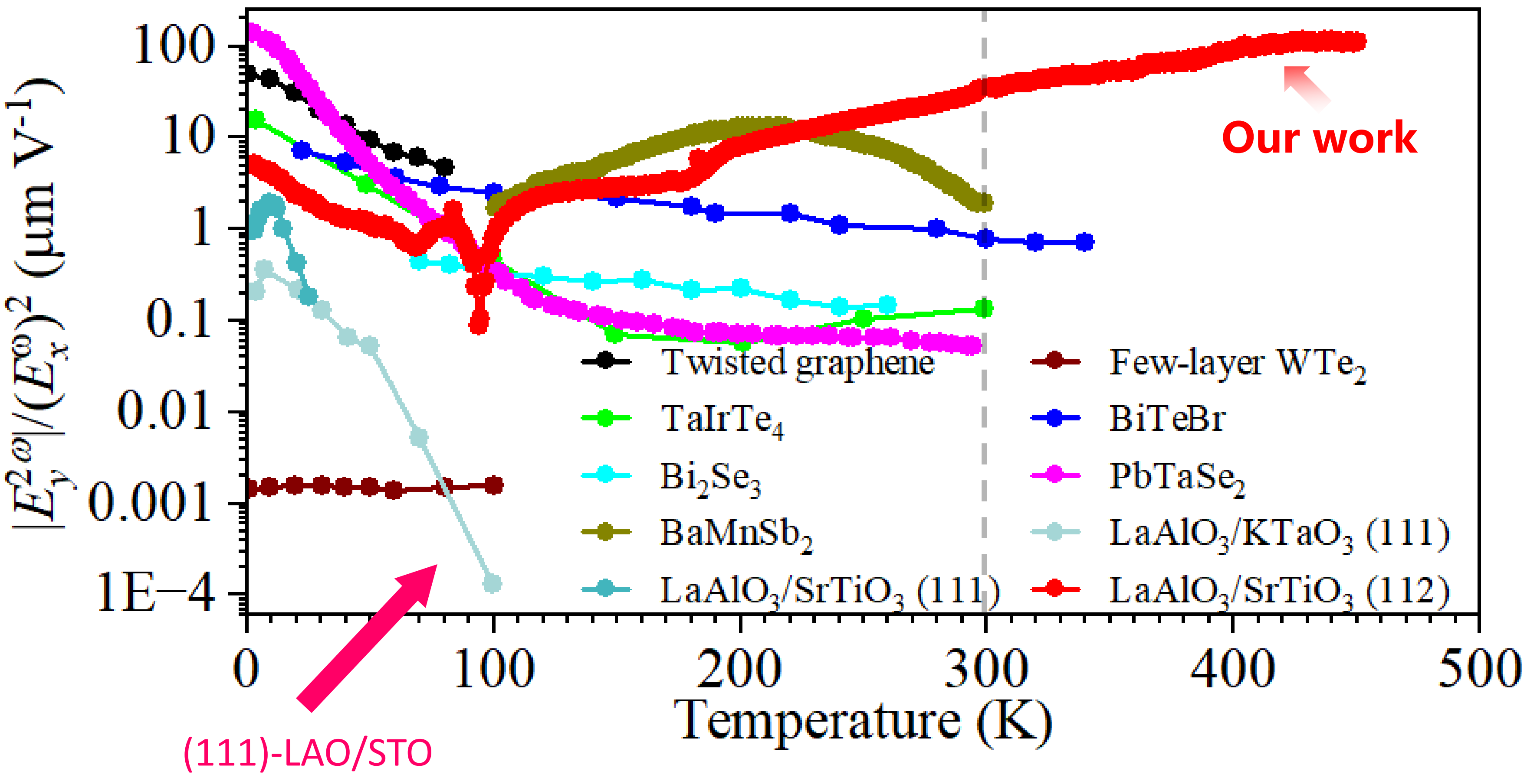
✓ Nonlinear Hall performance



Substantial nonlinear Hall effect in (112)-interface @ room temperature:

~100 larger than (111)-orientation @ low temp.

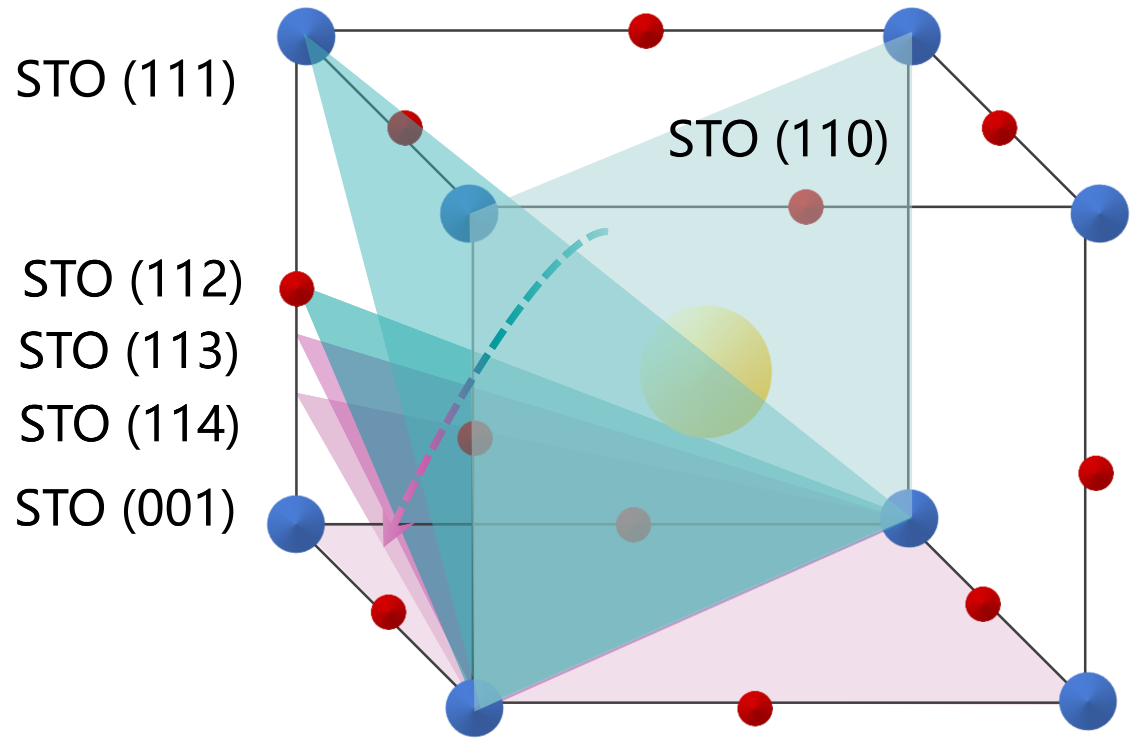
➢ Room temperature NLHE @(112)-$LaAlO_3/SrTiO_3$ interface



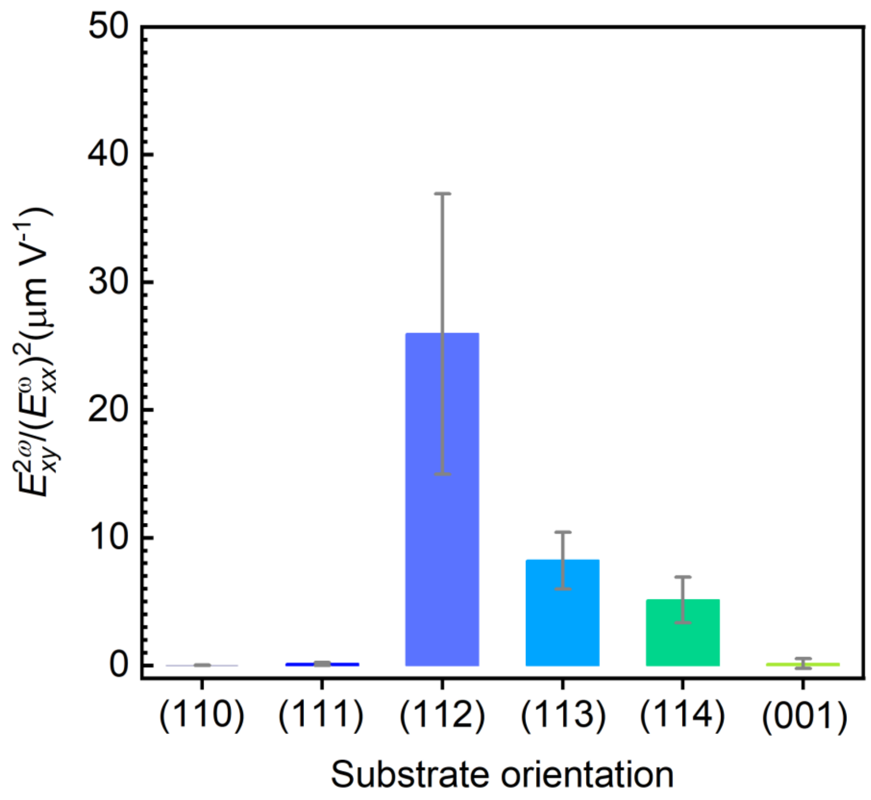
★ large coefficient ★ wide-range working temperature

➢ Crystal orientation modulation on NLHE @LAO/STO interface

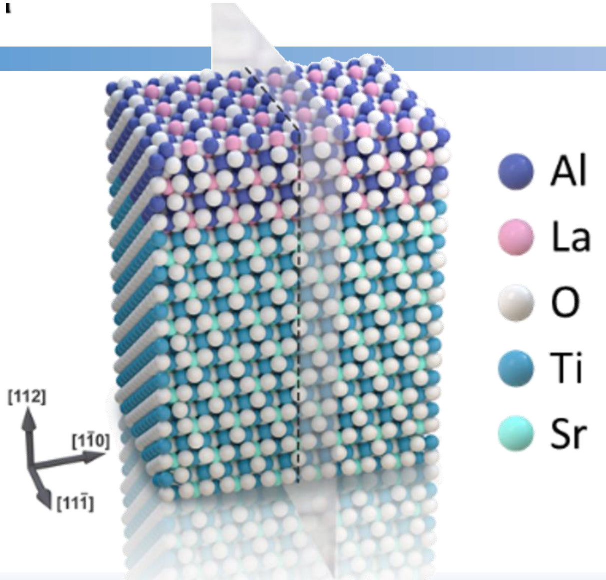
✓ Substrate orientation modulates the symmetry



✓ NLHE varies with the substrate orientation



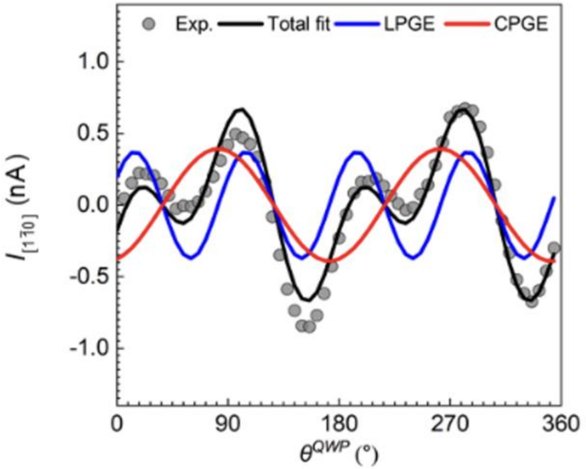
➢ Summary



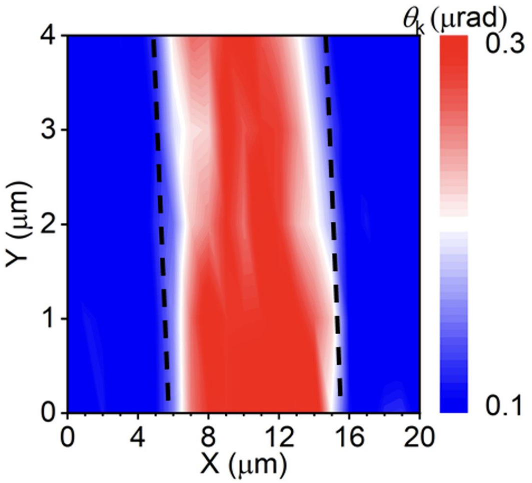
Nat. Commun. 2025 (In press)

The orientation Engineering @ interfaces.

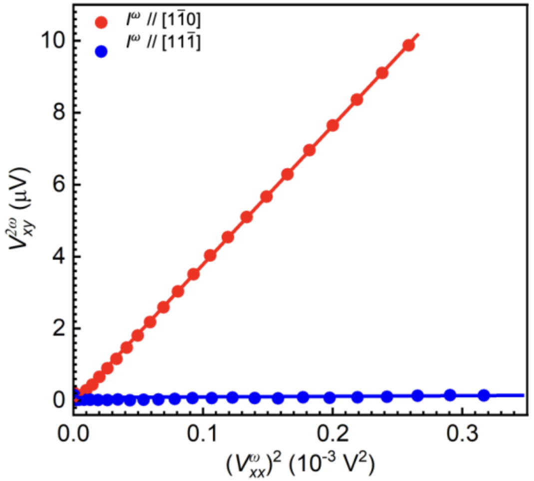
✓ Circular photogalvanic effect



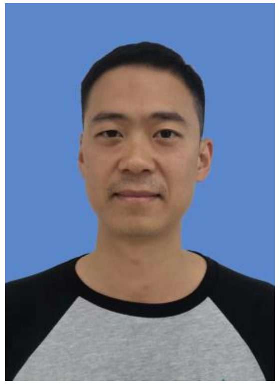
✓ Charge-magnetization conversion



✓ Nonlinear Hall effect



Acknowledgement



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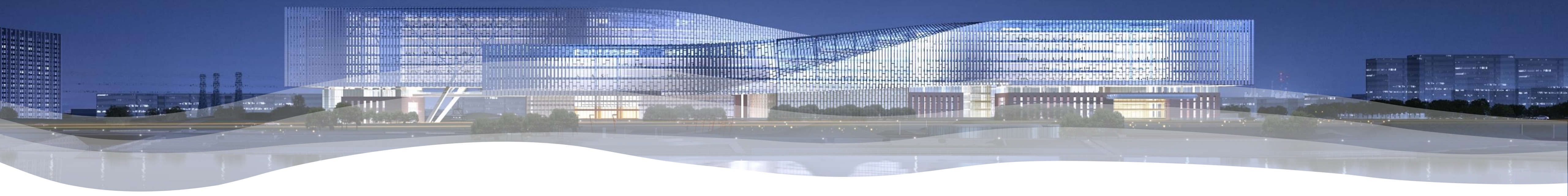

Thank You