

Spanish-German WE-Heraeus-Seminar: Emergent Topology and Unconventional Order in Quantum Matter
September 8-11th 2026

Nontrivial Equilibrium and Excited States of Quantum Materials from First Principles

A. Darmawan, M. E. Gruner, O.Köksal, D. Doennig, S. Bandyopadhyay and Rossitza Pentcheva

Department of Physics & CENIDE, University of Duisburg-Essen

Deutsche Forschungsgemeinschaft DFG

CRC/TRR80

SFB 1242

CRC/TRR 247
HETEROGENEOUS OXIDATION CATALYSIS IN THE LIQUID PHASE

2D-MATURE
SCALABLE 2D-MATERIALS ARCHITECTURES

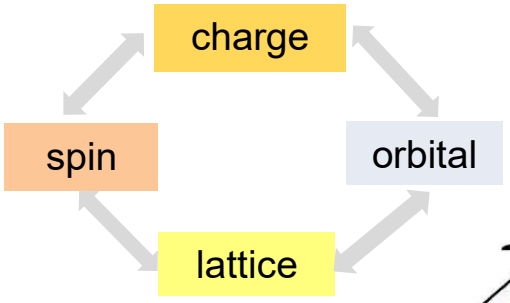
lrz magnitUDE

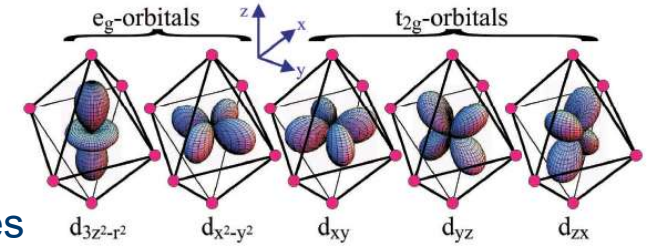
UNIVERSITÄT DUISBURG ESSEN
Offen im Denken

Motivation: Transition metal oxides (TMO)

UNIVERSITÄT DUISBURG ESSEN
Offen im Denken

Bulk

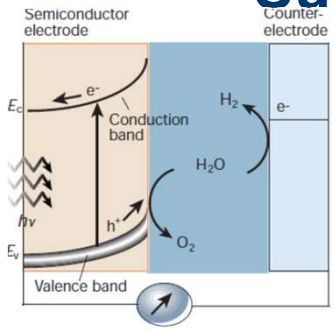




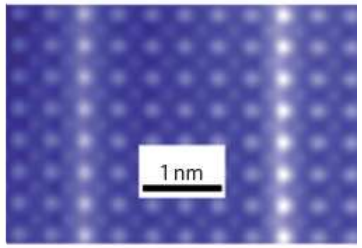
- tunability of properties
- collective phenomena: ferroelectricity, magnetism, superconductivity
- chemically&thermally stable
- environmentally benign
- cheap and abundant

Tokura&Nagaosa, Science 2000

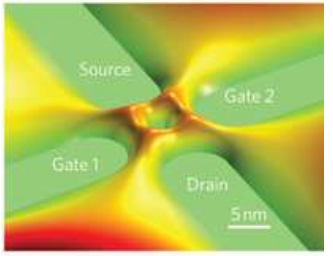
Surfaces & Interfaces



Grätzel (2000)



Ohtomo et al., Nature 419, 378 (2002)



G. Cheng et al., Nature Nano 6, 343 (2011)

Novel electronic phases / emergent phenomena not available in the bulk

- 2DEG, magnetism, superconductivity
- orbital reconstructions
- Metal-to-insulator transitions

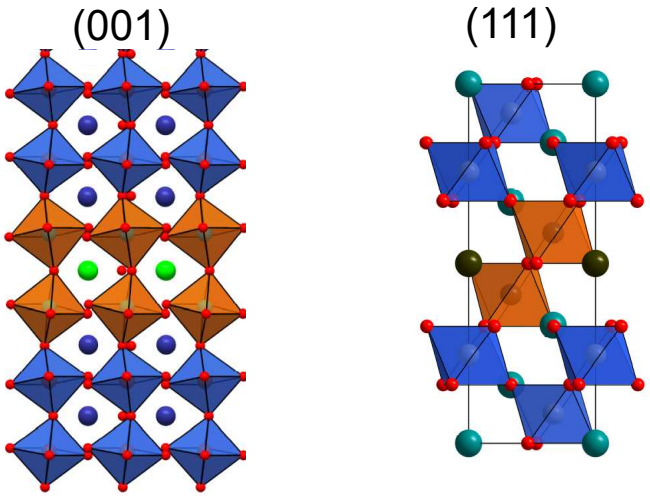
Applications: Electronics and spintronics devices
Oxide Nanoelectronics and Spintronics (Mannhart&Schlom 2010)

Fundamental understanding required

Oxide Interfaces: Control Parameters

UNIVERSITÄT DUISBURG ESSEN
Offen im Denken

- **Symmetry breaking**
- **strain**
- **Polarity**
- **Dimensional confinement**
- **d-band filling**
- **Crystallo-graphic orientation**
- **Nonequilibrium dynamics**



- fundamental understanding of the physics in nanoscale oxide materials
- develop concepts to design functionality

→ metal-to-insulator transitions (MIT)
→ charge, orbital and spin reconstructions
→ topologically nontrivial phases

J. Phys. D: Appl. Phys. **49**, 433001 (2016)
Phys. Status Solidi B **259**, 2100270 (2022)

Outline

UNIVERSITÄT DUISBURG ESSEN

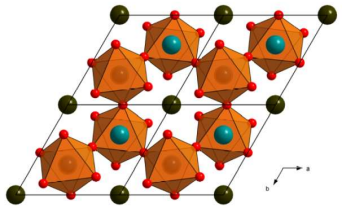
Offen im Denken

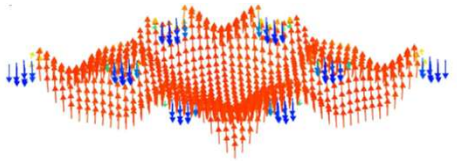
Novel electronic & topological phases in perovskite- and corundum superlattices with honeycomb pattern

PRB 93, 165145 (2016), npj Quantum Mat. 2017, Sci. Rep. **9**, 17306 (2019)

PRB **97**, 035126 (2018), J. Phys. & Chem. Solids **128**, 301 (2019)

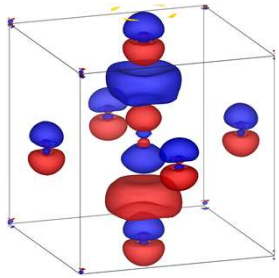
PRB **103**, 045135 (2021)

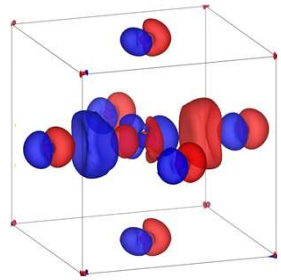




Uncovering the origin of laser-induced transient magnetiszation in $SrTiO_3$ from RT-TDDFT

arXiv.2602.20693





Methods

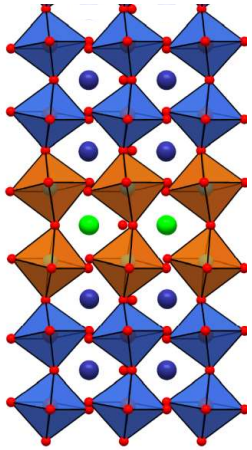
- DFT calculations: FP-LAPW in WIEN2k, PAW in VASP, QE
- topological analysis: Wannier90; TE properties: BoltzTrap
- LDA/GGA+*U, hybrid functionals, GW+BSE, RT TDDFT*

n-type $LaAlO_3/SrTiO_3$(001) vs (111): polar discontinuity

UNIVERSITÄT DUISBURG ESSEN
Offen im Denken

n-type (001)

extra ½ electron/IF

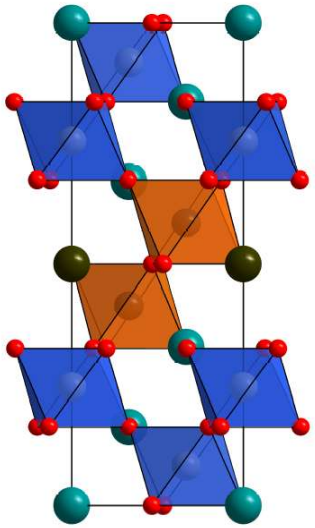


AlO_2	-1
LaO	+1
AlO_2	-1
LaO	+1
TiO_2	?
SrO	0
TiO_2	?
LaO	+1
AlO_2	-1
LaO	+1
AlO_2	-1

- stacking: AO - BO_2 - AO - BO_2
- polar discontinuity across IF
- 2DEG, magnetism, superconductivity…

n-type (111)

extra ½ electron/IF

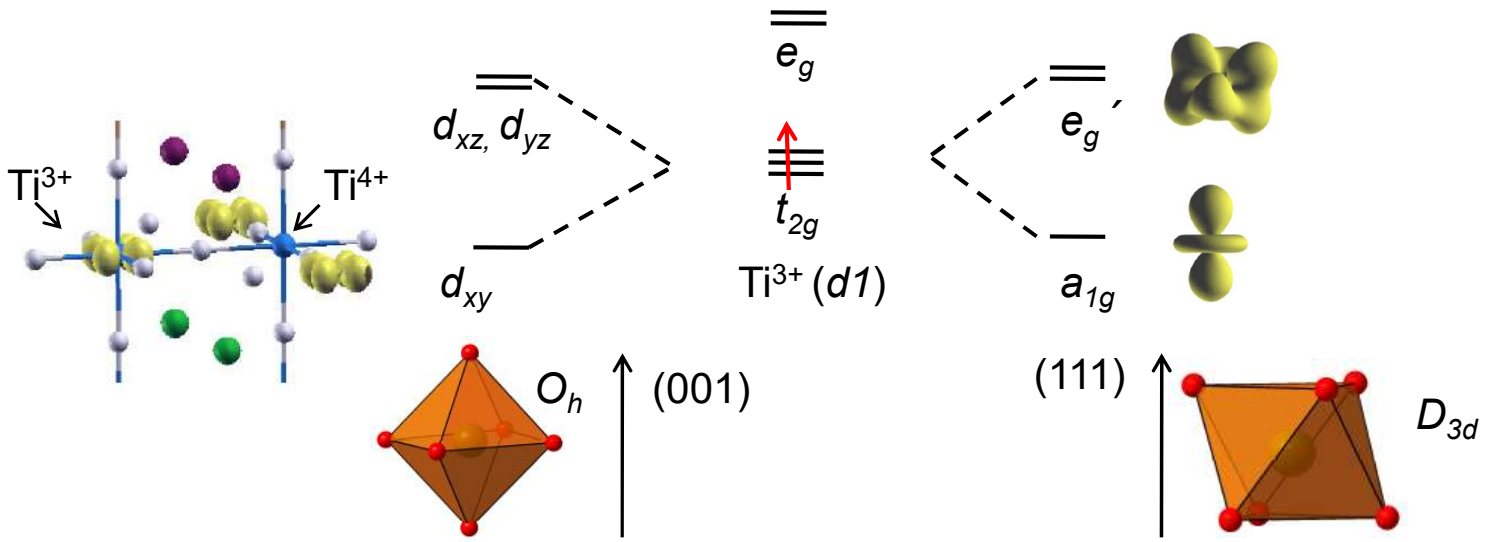


LaO_3	-3
Al	+3
LaO_3	-3
Al	+3
LaO_3	-3
Ti	?
SrO_3	-4
Ti	?
LaO_3	-3
Al	+3
LaO_3	-3
Al	+3
LaO_3	-3

- stacking: AO_3 - B - AO_3 – B
- planes have highly polar character
- charge mismatch @IF same as in (001)
  - ➢ 2DEG?

[1] A. Ohtomo and H.Y. Hwang, *Nature* **2004**, 427, 423.
[2] A. Brinkmann *et al.*, *Nat. Mater.* **2007**, 6, 493.
[3] N. Reyren *et al.*, *Science* **2007**, 317, 1196.

n-type $LaAlO_3/SrTiO_3$(001) vs (111): orbital reconstruction



(001)-oriented interfaces: [1-4]

- t_{2g} degeneracy lifted
  → d_{xy} orbital occupation (Ti^{3+} sites)
- charge ordered state @IF

(111)-oriented interfaces:

- symmetry lowered to D_{3d}
- t_{2g} → e_g´ + a_{1g}

➢ what orbital polarization can emerge?

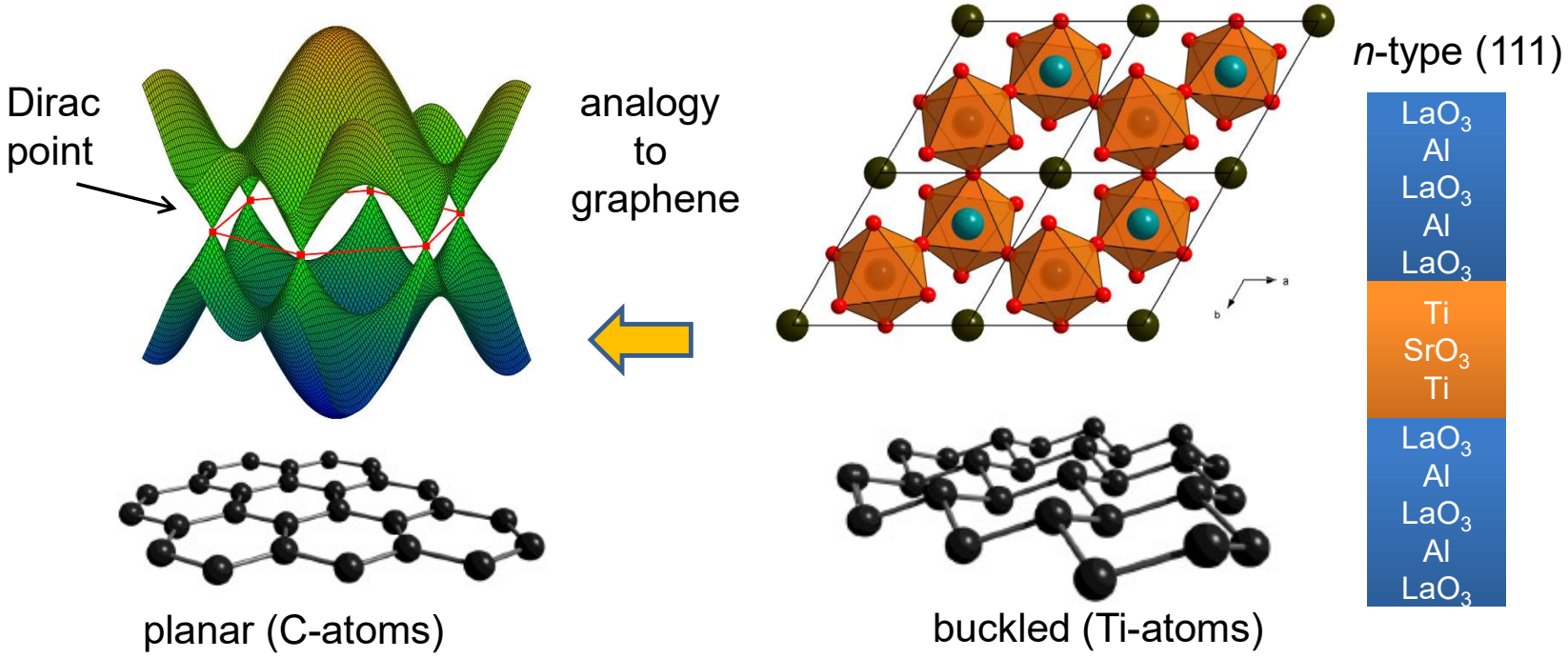
[1] RP and W.E. Pickett, *Phys. Rev*. B **78**, 205106 (2008).
[2] Z. Zhong and P.J. Kelly, *Eur. Phys. Lett.* **84**, 27001 (2008).
[3] M. Salluzzo *et al*., *Phys. Rev. Lett*. **102**, 166804 (2009).
[4] D. Doennig & RP, Sci. Rep. **5,** 7909 (2015).

$(ABO_3)_2/(A´B´O_3)_n(111)$: buckled honeycomb layer

UNIVERSITÄT DUISBURG ESSEN
Offen im Denken

Bilayer: buckled honeycomb lattice of B-site cations

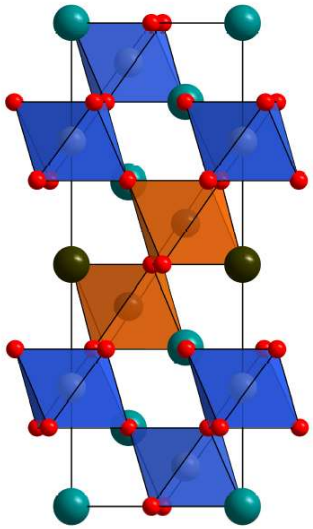
→ can support nontrivial topology: QAHI [1-3]

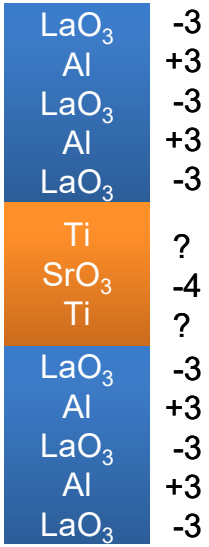


[1] F. D. M. Haldane, *Phys. Rev. Lett.* **61**, 2015 (1988).
[2] D. Xiao *et al.*, *Nat. Commun.* **2**, 596 (2011).
[3] A. Rüegg and G. A. Fiete, *Phys. Rev.* B **84**, 201103 (2011).

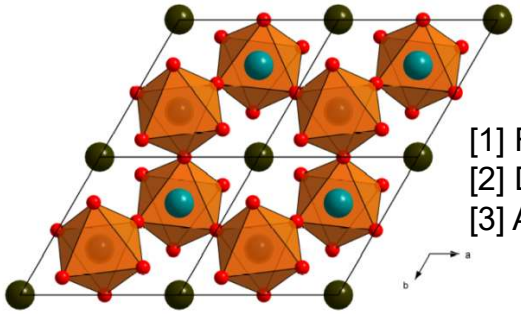
Novel electronic/topological phases in oxide superlattices with honeycomb pattern

UNIVERSITÄT DUISBURG ESSEN
Offen im Denken





$(LaAlO_3)_4$ / $(SrTiO_3)_2$ (111) SL



[1] F. D. M. Haldane, *Phys. Rev. Lett.* **61**, 2015 (1988).
[2] D. Xiao *et al*., *Nat. Commun.* **2**, 596 (2011).
[3] A. Rüegg and G. A. Fiete, *Phys. Rev.* B **84**, 201103 (2011).

Methods

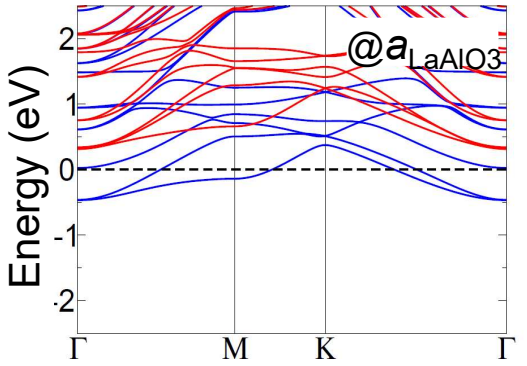
- DFT calculations,
- FP-LAPW method in WIEN2k, PAW in VASP
- LDA/GGA+*U*:
- wien2wannier, wannier90

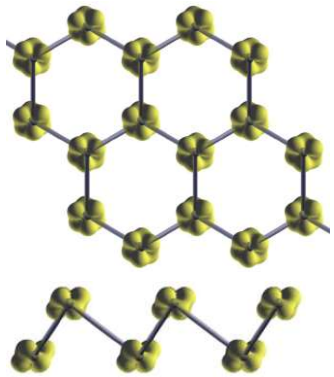
PRL111, 126804 (2013).

$(LaAlO_3)_4/(SrTiO_3)_2(111)$ SL: from Dirac cones to flat bands

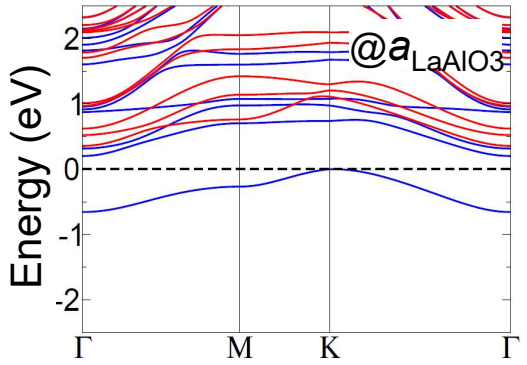
UNIVERSITÄT DUISBURG ESSEN
Offen im Denken

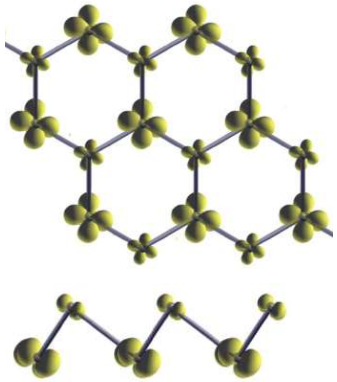
P321





P1



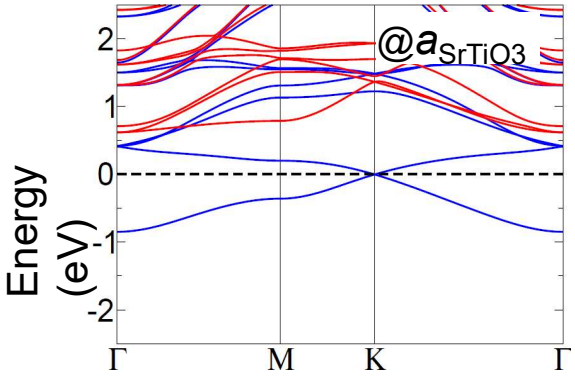


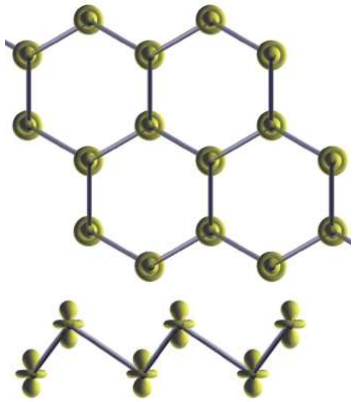
equivalent sublattices: e_g' vs a_{1g} orbital polarization as a function of strain

Relaxation leads to symmetry breaking of the two sublattices

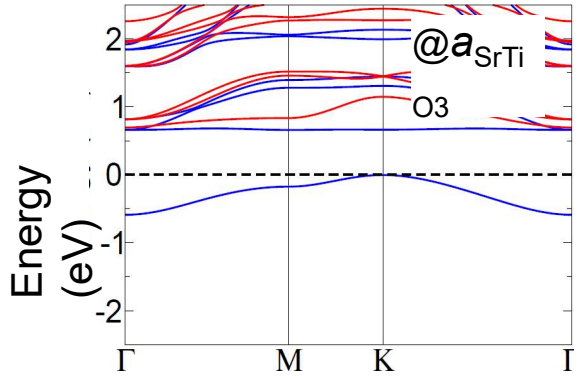
- insulating, flat bands, Ti^{3+},Ti^{4+} CO state
- a_{LaAlO3}: d_{xy} orbital @Ti^{3+}
- a_{SrTiO3}: a_{1g} orbital @ Ti^{3+}

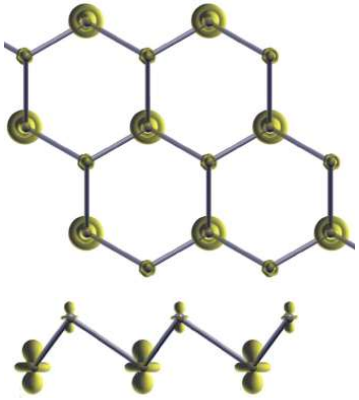
P321





P3

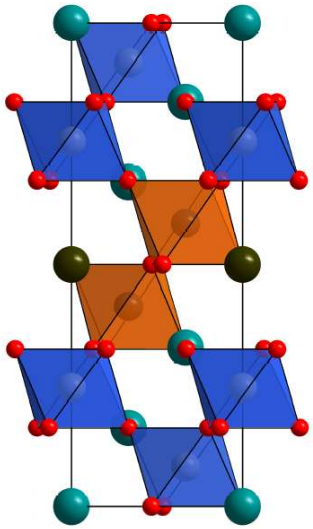


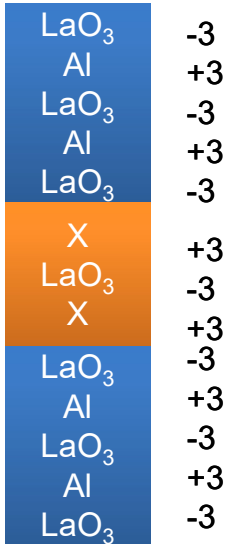


Doennig, Pickett & RP, PRL**111**, 126804 (2013).

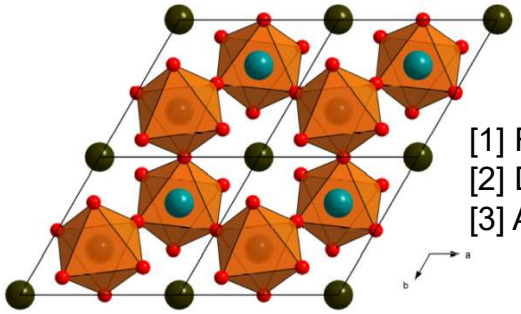
Novel electronic/topological phases in oxide superlattices with honeycomb pattern

UNIVERSITÄT DUISBURG ESSEN
Offen im Denken





$(LaXO_3)_2$ / $(LaAlO_3)_4$ (111) SL, *X=3d, 4d and 5d ion*



[1] F. D. M. Haldane, *Phys. Rev. Lett.* **61**, 2015 (1988).
[2] D. Xiao *et al*., *Nat. Commun.* **2**, 596 (2011).
[3] A. Rüegg and G. A. Fiete, *Phys. Rev.* B **84**, 201103 (2011).

Methods

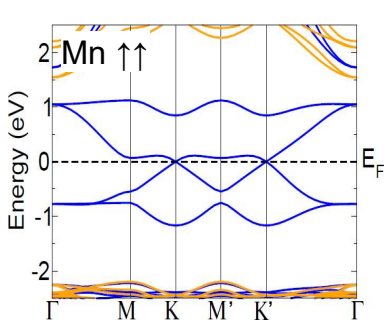
- DFT calculations,
- FP-LAPW method in WIEN2k, PAW in VASP
- LDA/GGA+*U*:
- wien2wannier, wannier90

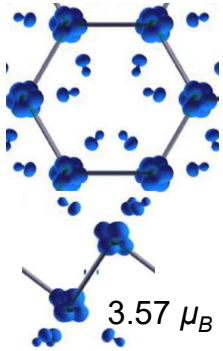
PRB 93, 165145 (2016)
npj Quantum Mat. 2, 4 (2017)
Sci. Rep. **9**, 17306 (2019)

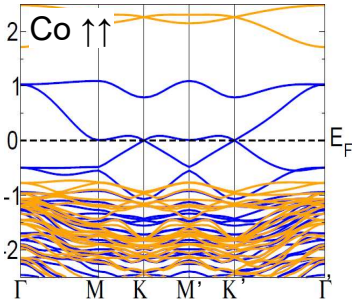
$(LaXO_3)_2/(LaAlO_3)_4(111)$ SL: e_g *systems*

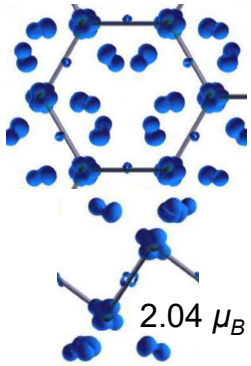
UNIVERSITÄT DUISBURG ESSEN
Offen im Denken

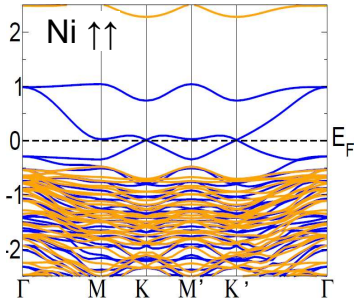
P321 symmetric case: similar band structure: four bands, two nearly flat and two linearly crossing at K, K'

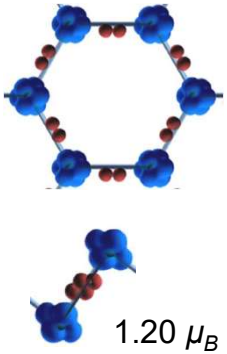




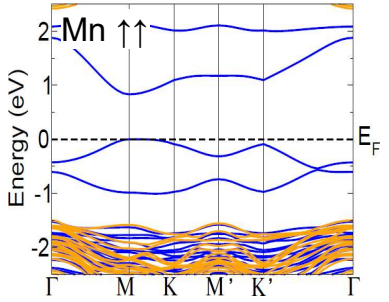


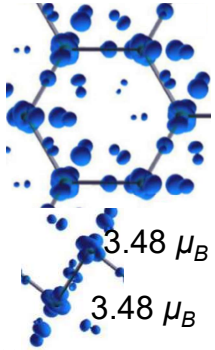




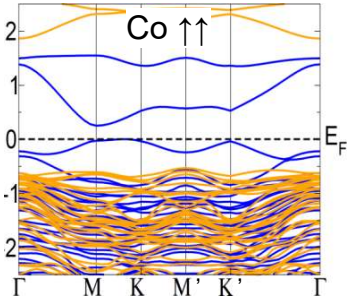


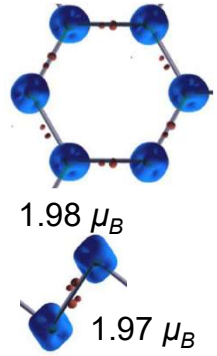
different symmetry-breaking mechanisms open a gap:



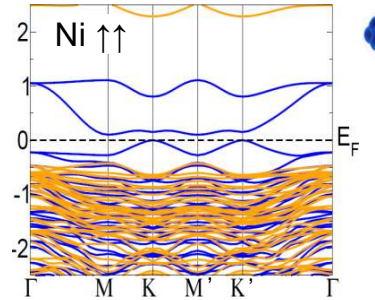


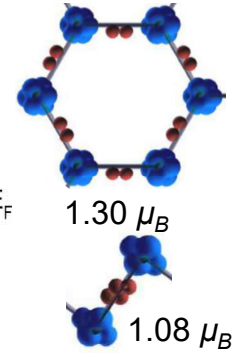
X=Mn: Jahn-Teller effect





X=Co: orbital ordering





X=Ni: site disproportionation

PRB 93, 165145 (2016)

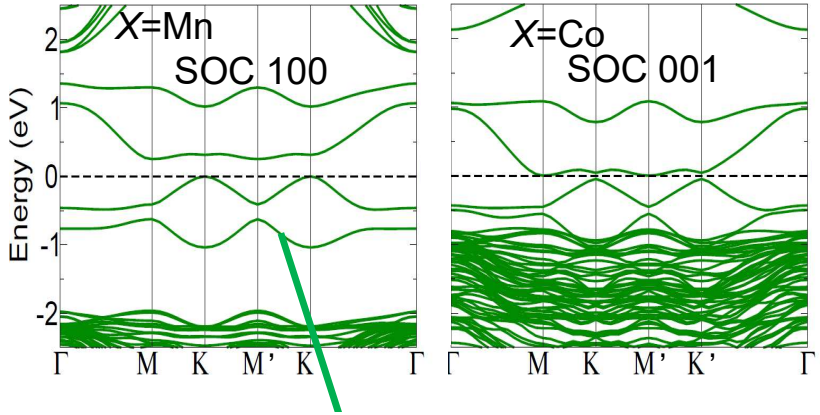
See also

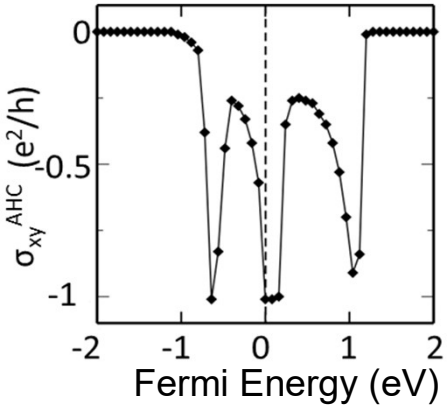
[1] Rüegg and G.A. Fiete, PRB 84, 201103R (2011).
[2] K.-Y. Yang et al., PRB 84, 201104R (2011).
[3] D. Doennig, W.E. Pickett & RP, PRB 89, 121110R (2014).

$(LaXO_3)_2/(LaAlO_3)_4(111)$: Effect of spin-orbit coupling

UNIVERSITÄT DUISBURG ESSEN
Offen im Denken

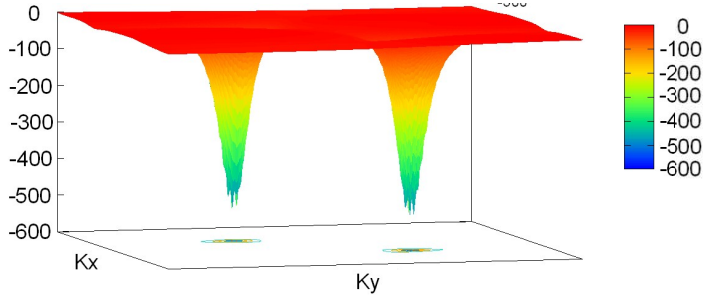
SOC: band gap opening & lifting of degeneracy @Γ



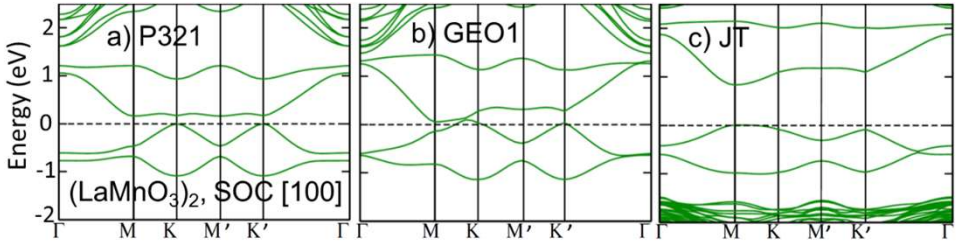


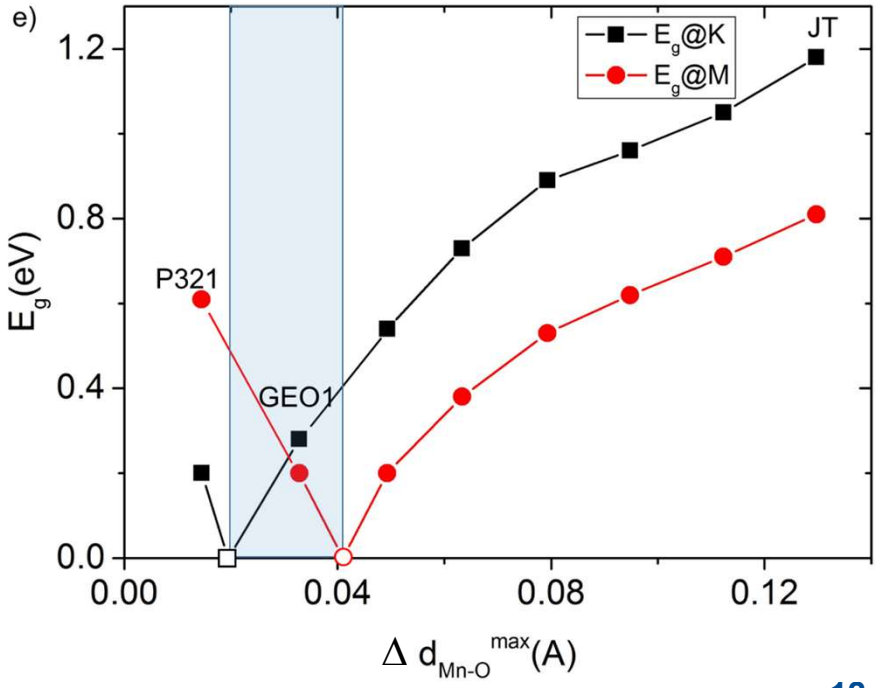
Anomalous Hall conductivity (wannier90):

X=Mn: QAHI with C=-1



Topological transition





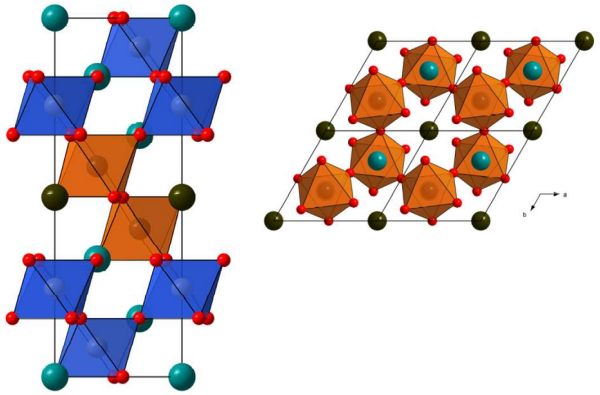
PRB 93, 165145 (2016)

Summary: topologically nontrivial phases in oxide superlattices

UNIVERSITÄT DUISBURG ESSEN
Offen im Denken

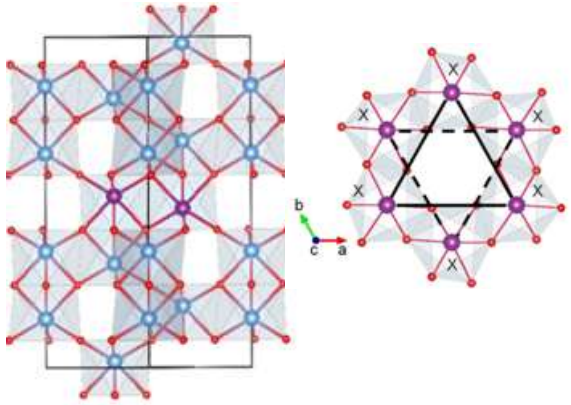
Perovskite-derived

$(LaXO_3)_2/(LaAlO_3)_4$ (111)

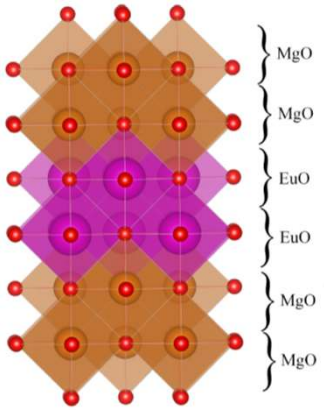


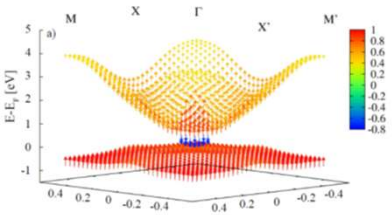
Corundum-derived honeycomb lattices

$(X_2O_3)_1/(Al_2O_3)_5$ (0001)



Rocksalt $(EuO)_1/(MgO)_3$(001) QW





- Candidates for **Chern** Insulators

X=Mn, Co, Ru, Os, Pt, Tc *X*= Pt, Tc

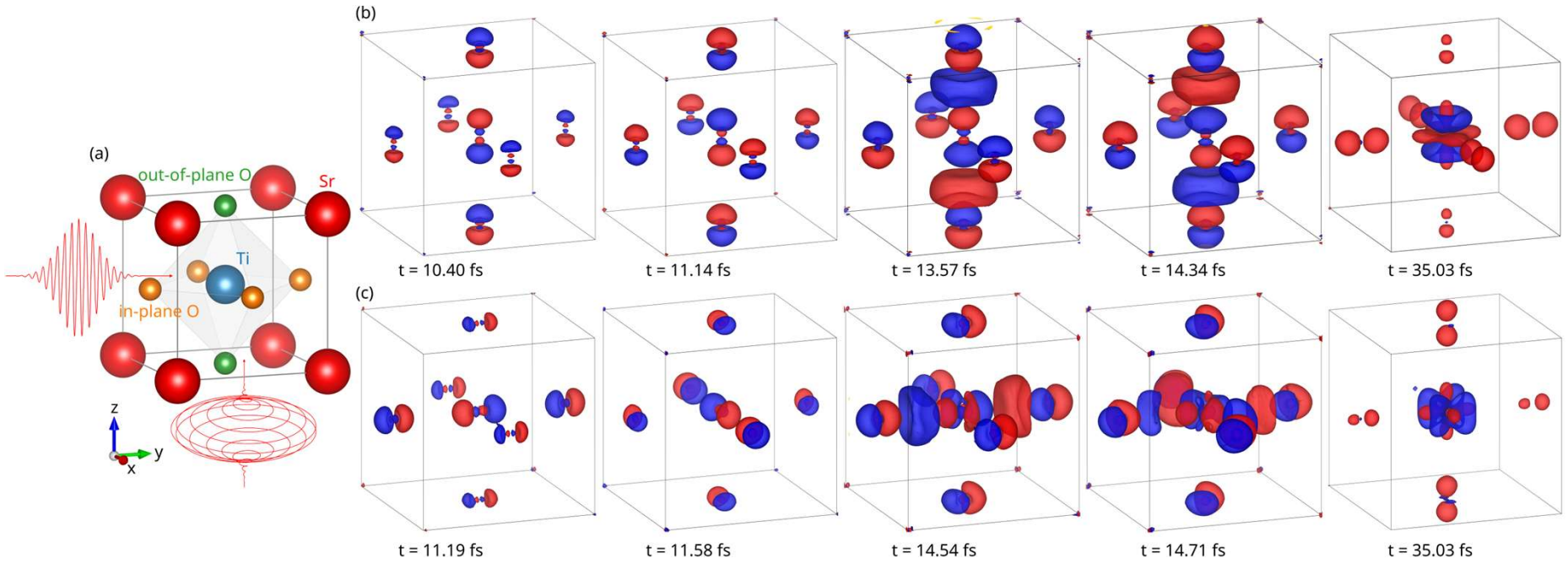
- FM coupling and symmetry of the two sublattices essential
- Tensile strain can stabilize the CI state
- Symmetry breaking and AFM coupling leads to trivial **Mott** insulating states: rich set of orbital/magnetic reconstruction unattainable in bulk
- Flat bands Strong SOC*: X*= Ti, Os

See also J. Chakhalian et al, APL Materials 8, 050904 (2020)

SOC induced band inversion between Eu 4f and 5d states for a_{MgO}

Enhanced thermoelectric response

PRB 93, 165145 (2016),
npj Quantum Mat. (2017)
PRB **97**, 035126 (2018),
J. Phys. & Chem. Solids **128**, 301 (2019)
Sci. Rep. **9**, 17306 (2019)
PRB **103**, 045135 (2021)



UNIVERSITÄT DUISBURG ESSEN

Offen im Denken

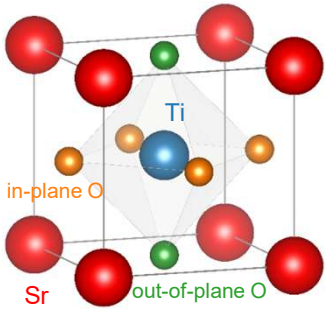
Uncovering the origin of laser-induced transient magnetiszation in $SrTiO_3$ from RT-TDDFT

with Andri Darmawan, Markus E. Gruner

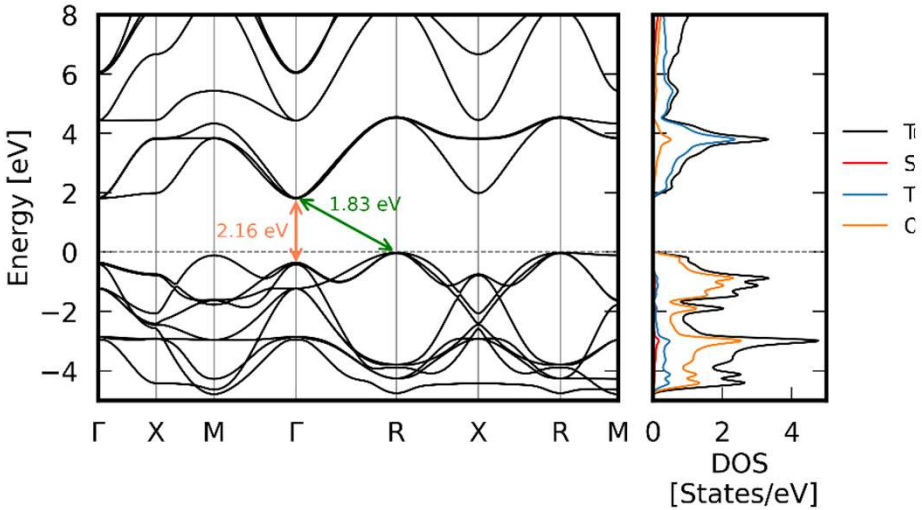
arXiv.2602.20693

$SrTiO_3$: Ground state properties:

- $SrTiO_3$
  - Quantum paraelectric
  - Cubic to tetragonal transition @105K[1]
  - Diamagnetic Ti 3*d* (d^0) band insulator
  - Experimental band gap [2]
    - $E_g^{exp}\,(\Gamma - \Gamma) = 3.78\,\text{eV}$
    - $E_g^{exp}\,(\Gamma - \mathrm{R}) = 3.20\,\text{eV}$
  - Optical and x-ray absorption spectra of bulk $SrTiO_3$ (GW+BSE) [3-4]

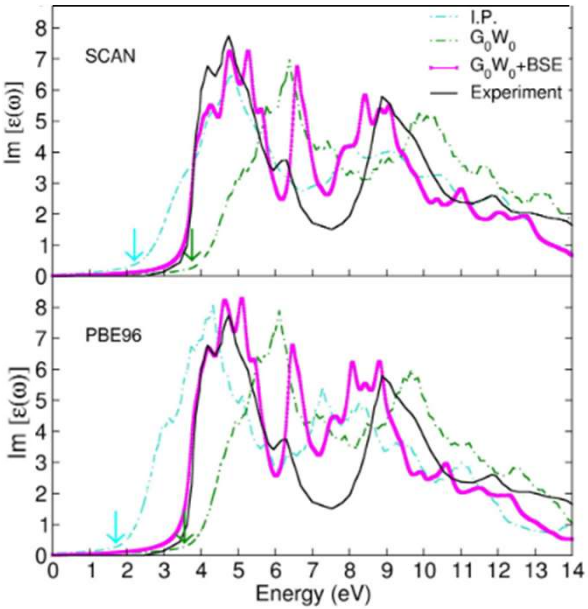


DFT band structure of STO: GGA-PBEsol



VBM O 2*p*, CBM Ti 3d

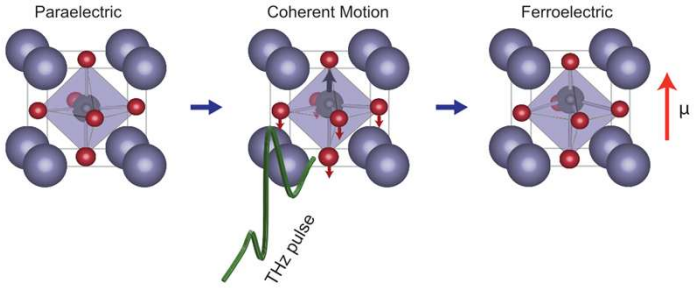
Optical spectrum of STO



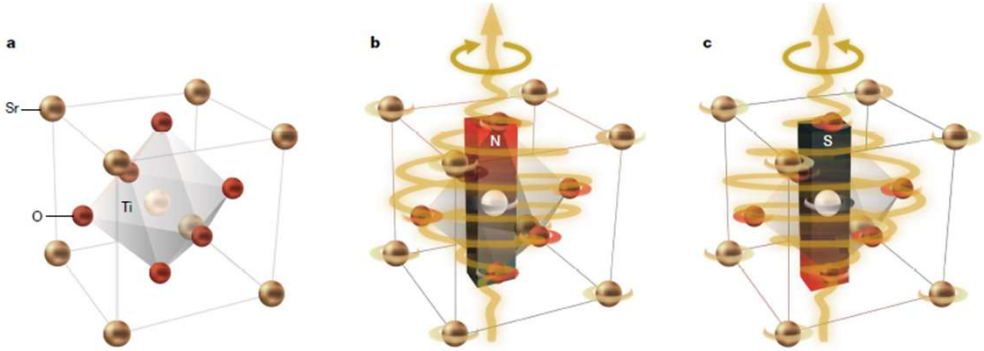
[1] Fleury et al., Phys. Rev. Lett. 21, 16 (1968)
[2] Gogoi and Schmidt. Phys. Rev. B. **93**, 075204 (2016)
[3] Begum et al., Phys. Rev. Mater. 3, 065004 (2019)
[4] Begum-Hudde et al., Phys. Rev. Res. 5, 013199 (2023)

Motivation

- Paraelectric to ferroelectric transition upon excitation with IR or THz pulses [1-5]



- Dynamic multiferroicity driven by circularly polarized THz electric-field



[1] T. F. Nova et al., Science 364, 1075 (2019)
[2] X. Li et al., Science 364, 1079 (2019)
[3] D. Shin et al., Phys. Rev. Lett. 129, 167401 (2022)
[4] C. Song et al, J. Phys. Chem. Lett.14, 576 (2023)
[5] G. Orenstein et al, Nat. Phys (2025)

Possible origin of induced magnetization:

- Barnet effect: coherent phonon rotation induces a magnetization and orbital angular momentum (OAM) in electronic subsystem [6,7]
- electron-phonon coupling [7]
- non-Maxwellian fields [8,9]
- electron-nuclear quantum geometry [10]

- Previous studies THz & IR excitation, excitation of phonons
- here: optical pulse, electronic excitations
- Comparison of linear vs circularly polarized light to induce magnetization in $SrTiO_3$

[6] M. Basini et al., Nature 628, 534 (2024)
[7] N. Shabala and R. M. Geilhufe, Phys. Rev. Lett. 133, 266702 (2024)
[8] R. Merlin, Phys. Rev. B 110, 094312 (2024)
[9] R. Merlin, PNAS nexus 4 (1), pgaf002 (2025)
[10] L. Klebl et al., Phys. Rev. Lett. 134, 016705 (2025)

Real-time time-dependent DFT

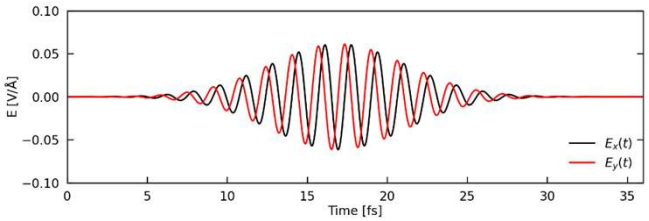
- Non-collinear spin-dependent time-dependent Kohn-Sham equations

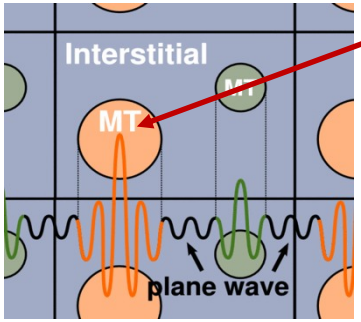
$$i\frac{\partial\psi_j(\boldsymbol{r},t)}{\partial t}=\left[\frac{1}{2}\left(-i\nabla+\frac{1}{c}\boldsymbol{A}_{ext}(t)\right)^2+v_s(\boldsymbol{r},t)+\frac{1}{2c}\,\boldsymbol{\sigma}\cdot\boldsymbol{B_s}(\boldsymbol{r},t)+\frac{1}{4c^2}\,\boldsymbol{\sigma}\cdot\left(\nabla_s(\boldsymbol{r},t)\times(-i\nabla)\right)\right]\psi_j(\boldsymbol{r},t)$$

- Vector potential $\boldsymbol{A}_{ext}(t)$ describes the laser pulse

$$\boldsymbol{A}_{ext}(t)=\boldsymbol{A}_0\exp\left[-\frac{(t-t_0)^2}{2\sigma^2}\right]\sin[\omega(t-t_0)+\phi_i]\quad\text{where }\sigma=\frac{FWHM}{2\sqrt{2\ln 2}}.$$

- Electric field: $\boldsymbol{E}=-\frac{1}{c}\frac{\partial\boldsymbol{A_{ext}}}{\partial t}$
- Electric field amplitude: $E_0\approx 0.06$ V/Å
  - Peak power intensity $S_{peak}=10^{11}$ W/cm²
  - Fluence ~ 0.38 mJ/cm²





Q(t): electronic charge Muffin-tin sphere centered around each nucleus

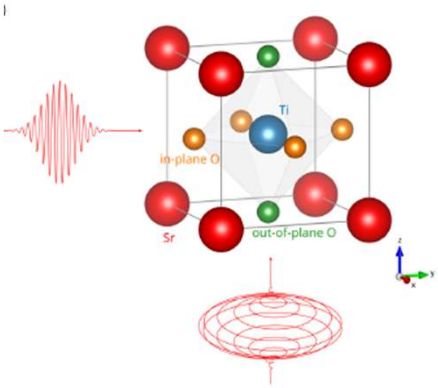
[1] J.K. Dewhurst, S. Sharma, "Development of the Elk LAPW Code," Max Planck Institute of Microstructure Physics, Theory Department

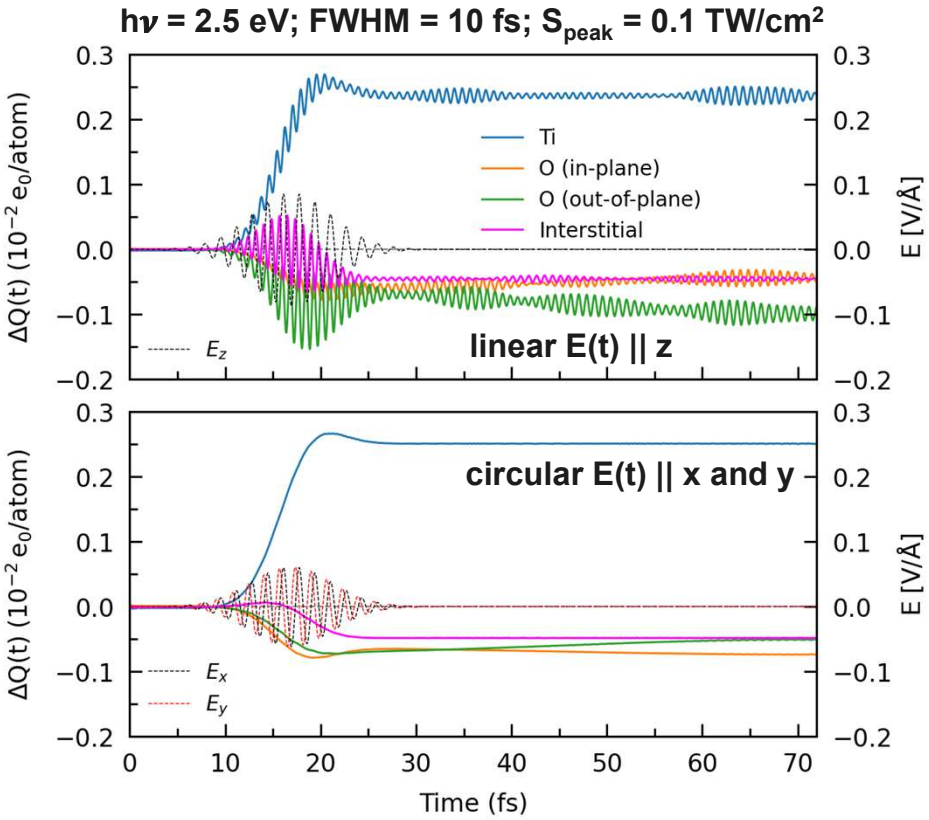
All-electron full-potential linearized augmented plane wave (FP-LAPW) implemented in the Elk code (https://elk.sourceforge.io)

- *K*-mesh of $10\times10\times10$
- Plane-wave $R_{min}^{MT}|\boldsymbol{G}+\boldsymbol{K}|_{max}=8$
- Exchange-correlation functional: PBEsol
- SOC taken into account
- Time step: $dt=0.726\,as$
- magnitUDE and amplitUDE HPC facilities

$SrTiO_3$: Linear vs. circular polarization

Charge excitation



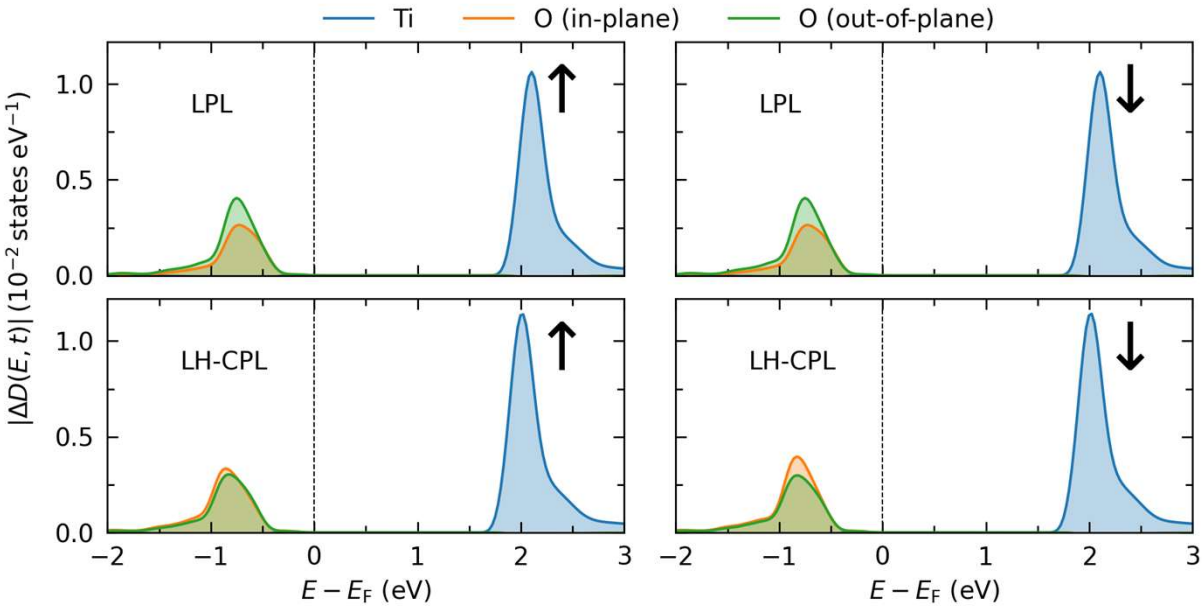


- Charge transfer from O (and interstitial) to Ti sites
- Similar excitation pattern for linearly and circularly polarized pulses
- Difference between in-plane and out-of-plane O

$SrTiO_3$: Linear vs. circular polarization

Differential time-dependent DOS

$$\Delta D(E,t) = D(E,t) - D(E,0)$$



$\hbar\omega$ = 2.5 eV; FWHM = 10 fs; S_{peak} = 0.1 TW/cm²

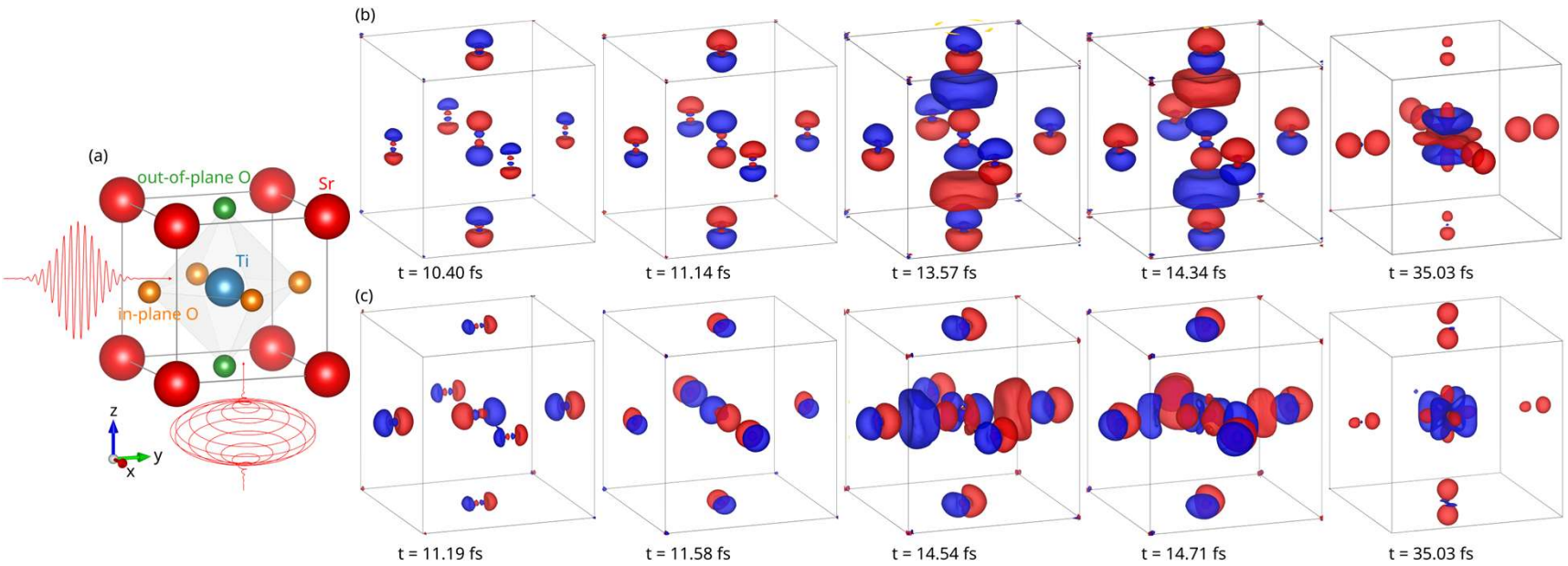
- Excitations from O 2*p* states (−1.0 to −0.5 eV) to empty Ti 3*d* states (1.7 − 2.3 eV)
- Depletion distinct for in- & out-of-plane O 2*p*
- CPL: difference in majority/minority states @ in-planeO 2*p*

$SrTiO_3$

Time-dependent electron density redistribution

$$\Delta\rho(\mathbf{r},t) = \rho(\mathbf{r},t) - \rho(\mathbf{r},0)$$

hν = 2.5 eV



LPL

Left-handed (LH)-CPL

- LPL: Oscillating positive and negative lobes at Ti and O sites with opposite orientation along *E*
- Dynamical pattern of the electron clouds resembles the opposite movement of Ti and O in the soft transverse optical TO_1 mode → dynamical breaking of inversion symmetry

Red: Depletion of charge

Blue: Accumulation of charge

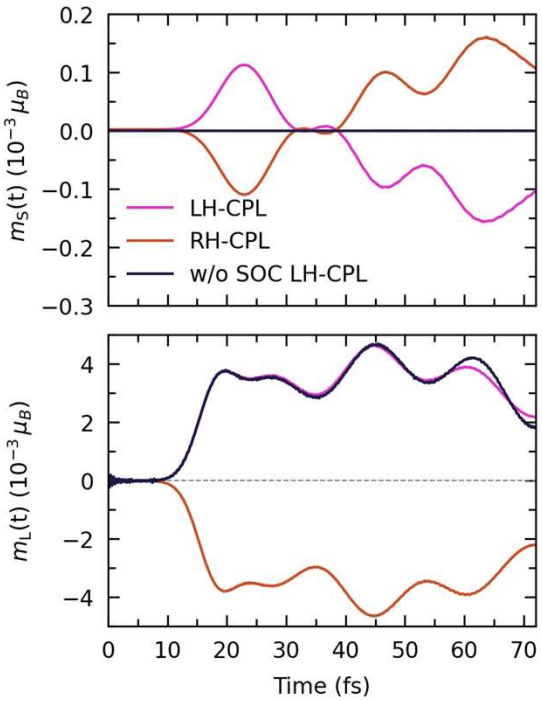
Isosurface level of $\pm 2.02 \times 10^{-3} \frac{e}{\text{Å}^3}$

- LH-CPL: Rotation of positive and negative electron density lobes around O sites → induce circular current
- RH-CPL: opposite rotation of electron cloud
- Transfer of orbital angular momentum (OAM) to electronic subsystem

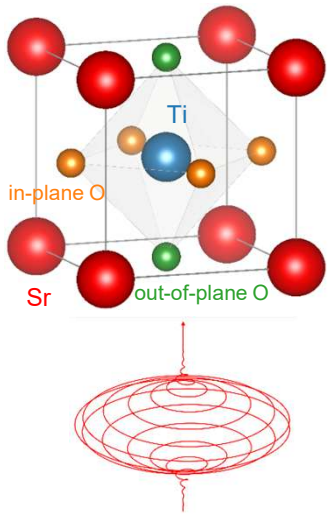
Darmawan, Gruner&RP., arXiv:2602.20693

$SrTiO_3$

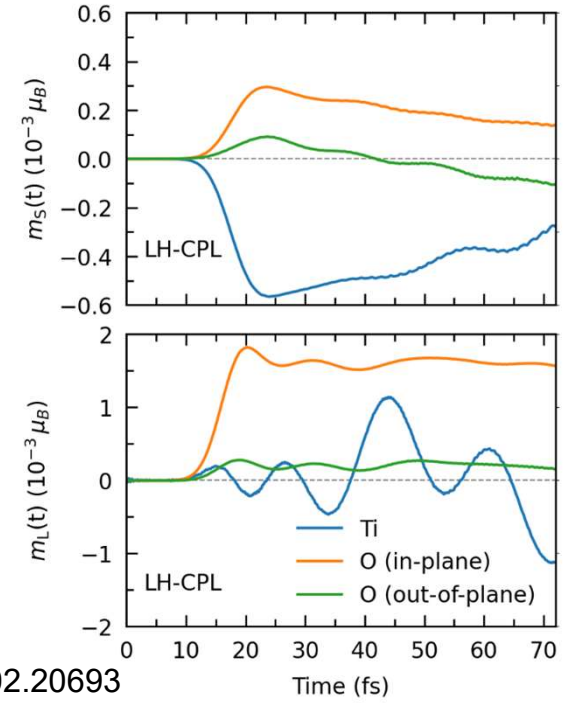
Total light-induced spin and orbital moment



$\hbar\omega$ = 2.5 eV; fluence ~ 0.38 mJ/cm^2



Element-resolved light-induced spin and orbital moment



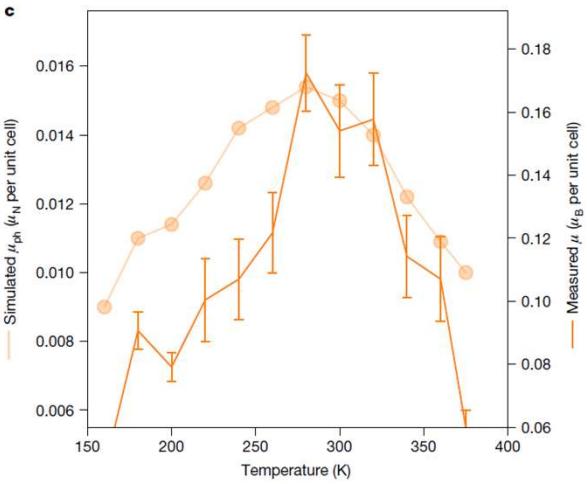
Darmawan, Gruner, and Pentcheva, arXiv:2602.20693

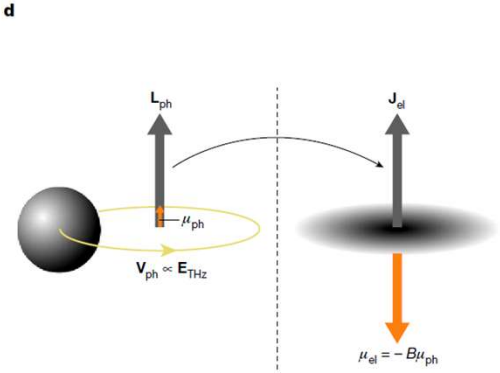
- Spin and orbital momentum is helicity-dependent
- Orbital moment independent of spin-orbit coupling (SOC)
- SOC key for the transfer from orbital to spin AM

- Spin IFE: Opposite direction between in-plane O and Ti
- Orbital IFE:
  - prominent contribution due to in-plane O
  - an order of magnitude larger than spin IFE

Comparison to previous experiment and theoretical results

Measured Kerr Rotation & simulation upon THz excitation in $SrTiO_3$





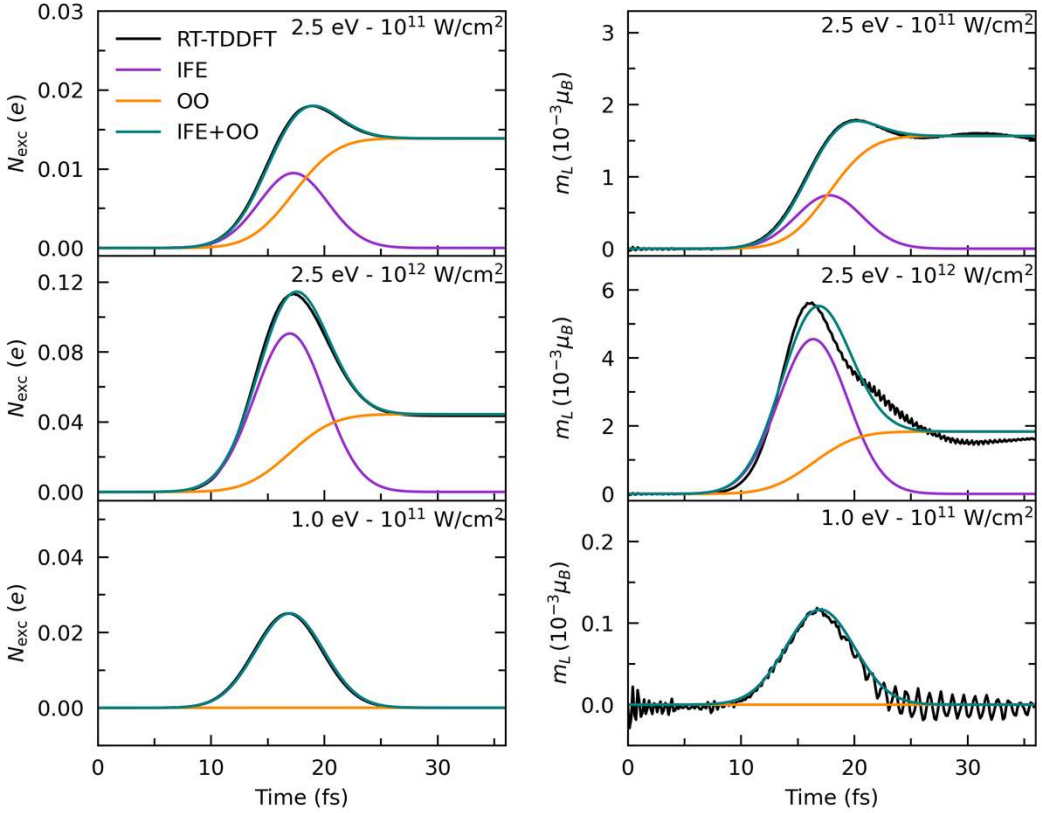
	Spin moment	Orbital moment	Total moment
$SrTiO_3$ (This work)	$0.5 \times 10^{-3}\mu_B$	$4.5 \times 10^{-3}\mu_B$	$5.0 \times 10^{-3}\mu_B$
$SrTiO_3$ (Dyn. Multiferroicity prediction) [1]	-	-	$8.2 \times 10^{-6}\mu_B$
$SrTiO_3$ (Exp. measured) [1]	-	-	$1.6 \times 10^{-1}\mu_B$

[1] Basini et al., Nature 628, 534 (2024)

Inverse Faraday effect vs. optical orientation

Inverse Faraday effect

- Nonlinear process mediated by virtual electronic states
- Scales with instantaneous intensity



Optical orientation

- Direct interband transitions between valence and conduction band
- Grows with time-integrated intensity (pulse fluence)

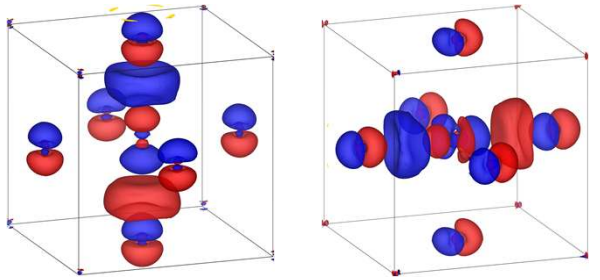
$$N_{exc}(t) = N^{IFE} \exp\left[-\frac{(t-t_0)^2}{2\sigma^2}\right] + \frac{N^{OO}}{2}\left[1 + \frac{\mathrm{erf}(t-t_0)}{\sqrt{2}\sigma}\right]$$

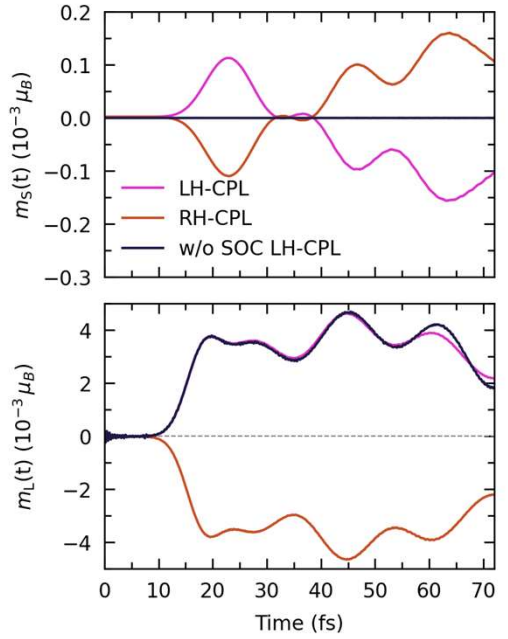
$$m_L(t) = m_L^{IFE} \exp\left[-\frac{(t-t_0)^2}{2\sigma^2}\right] + \frac{m_L^{OO}}{2}\left[1 + \frac{\mathrm{erf}(t-t_0)}{\sqrt{2}\sigma}\right]$$

- below gap excitation (1.0 eV): pure IFE contribution
- above gap excitation (2.5 eV):
  - IFE predominant during pulse,
  - OO develops later and saturates after the pulse
  - 10^{11} W/cm^2 → OO contribution > IFE contribution
  - 10^{12} W/cm^2 → IFE contribution > OO contribution

Conclusion

- LPL:
  - Formation of oscillating dipoles at Ti and O sites with significant orbital polarization and opposite sign
  - Dynamical breaking of inversion symmetry
- CPL: transfer of angular momentum from light - rotation of electron density around atomic positions
- Laser-induced orbital moment in $SrTiO_3$ an order of magnitude higher than spin moment
- Spin-orbit coupling plays a key role for the conversion of orbital angular momentum to spin angular momentum
- Disentanglement of IFE and OO contribution to the induced orbital magnetization





Darmawan, Gruner, and RP, arXiv:2602.20693

Acknowledgement

- A. Darmawan, M.E. Gruner (UDE)
- V. Begum-Hudde (U Urbana-Champaign)
- O.Köksal (U Gent), D. Doennig (LMU),
- W.E. Pickett (UC Davis)
- J. Chakhalian (Rutgers U)
- J. Freeland (Argonne NL)
- A. Gray (Temple U)
- M. Basini (ETH Zürich)
- D. Jüraschek, (U Eindhoven)
- A. Eschenlohr, K. Ollefs (UDE)

