Rashba spin-orbit coupling in the oxide 2D structures: The $KTaO_3$ (001) Surface

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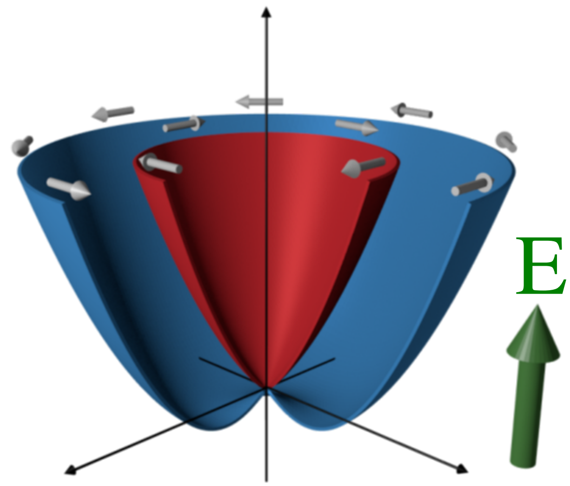
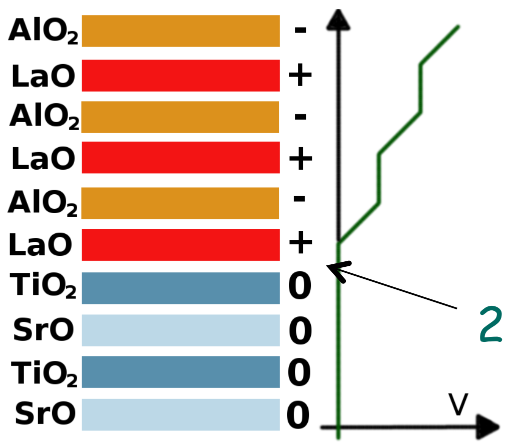


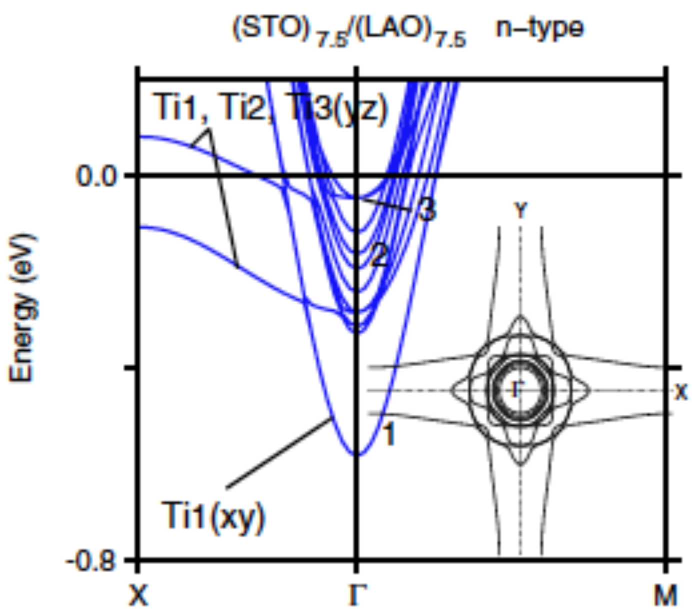
Ref: K. V. Shanavas and S. Satpathy, Phys. Rev. Letts. 112, 086802 (2014) ; Also: Supplementary section

U.S. DEPARTMENT OF ENERGY

Correlated Oxides 2014, Minneapolis
May 3, 2014

Gate control of the Rashba SOI in $LaAlO_3/SrTiO_3$ interface

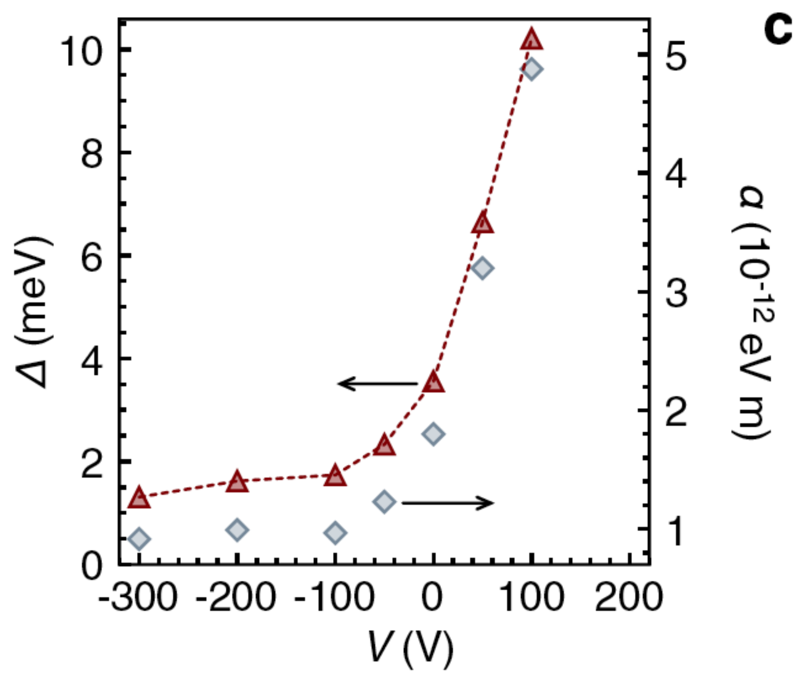




Ohtomo and Hwang, Nature (2004)

Popovic, SS, Martin, PRL (2008)

Measure Rashba coefficient from spin relaxation rates



Ben Shalom et al, PRL (2010)

Caviglia, Gabay, et al., PRL (2010)

Free-electron result : $\alpha_R = \dfrac{\hbar^2 E}{2m^2c^2}$

TB result : $\alpha_R = \dfrac{2\beta\xi}{\varepsilon}, \quad \beta \approx E$

PRL **108,** 117602 (2012) PHYSICAL REVIEW LETTERS week ending 16 MARCH 2012

Subband Structure of a Two-Dimensional Electron Gas Formed at the Polar Surface of the Strong Spin-Orbit Perovskite $KTaO_3$

P. D. C. King,[1] R. H. He,[2] T. Eknapakul,[3] P. Buaphet,[3] S.-K. Mo,[2] Y. Kaneko,[4] S. Harashima,[5] Y. Hikita,[6,7] M. S. Bahramy,[8] C. Bell,[6,7] Z. Hussain,[2] Y. Tokura,[4,5,8] Z.-X. Shen,[6,7] H. Y. Hwang,[6,7,8] F. Baumberger,[1,*] and W. Meevasana[3,9,†]

[1]*SUPA, School of Physics and Astronomy, University of St. Andrews, St. Andrews, Fife KY16 9SS, United Kingdom*

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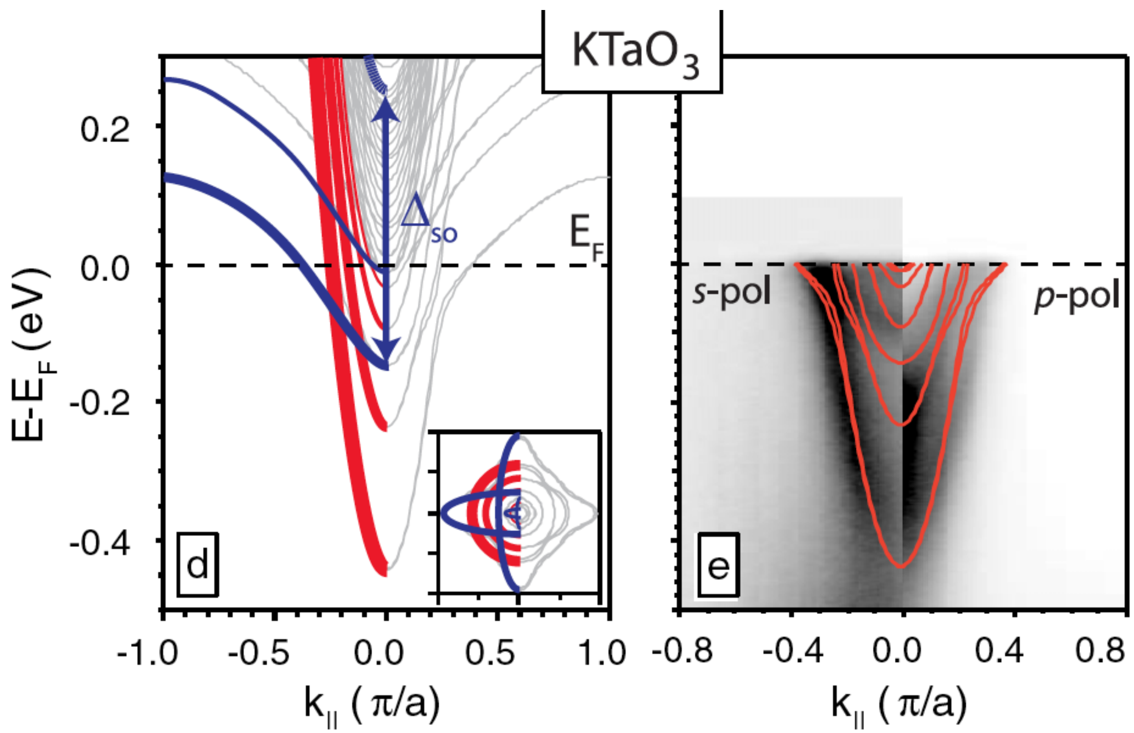
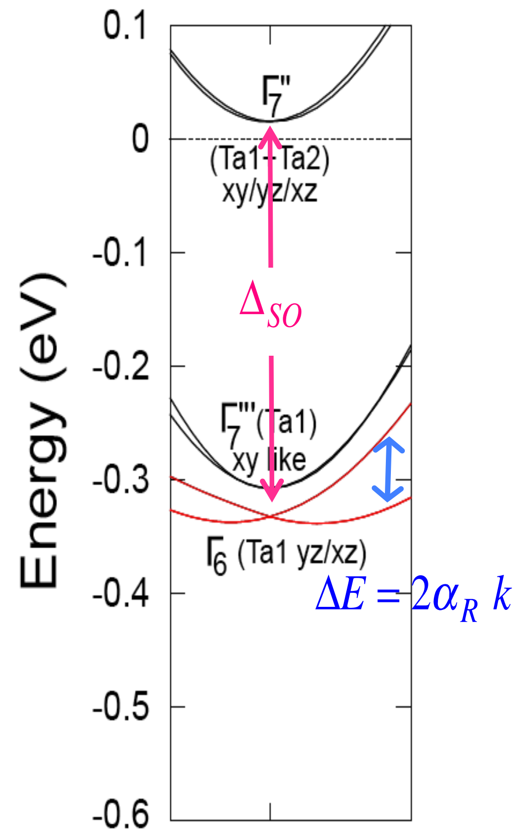




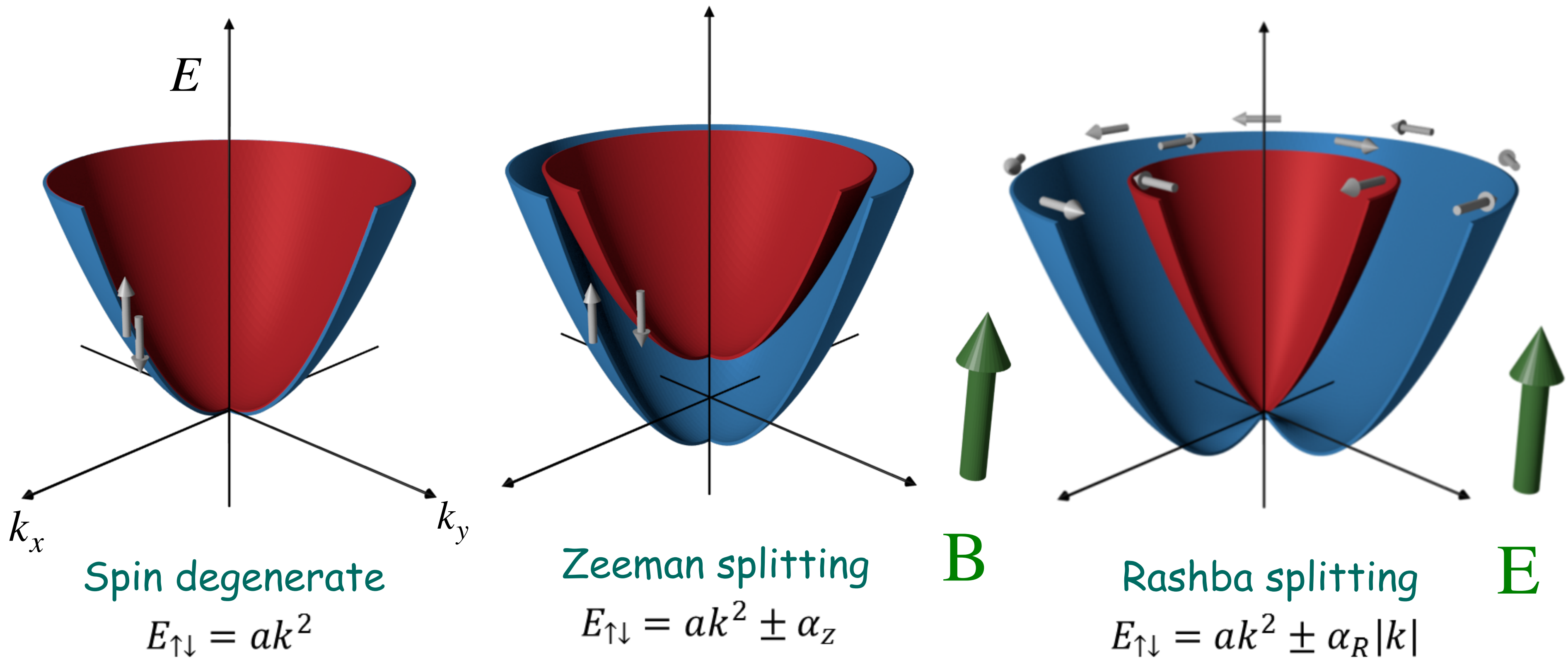
Looked for Rashba: Found small $\Delta k_{\|} \sim 0.02$ Å^{-1}

Emmanuel I. Rashba (1927-)

The Rashba Effect

Momentum-dependent splitting of the spin bands in 2D systems (surfaces, interfaces, heterostructures)

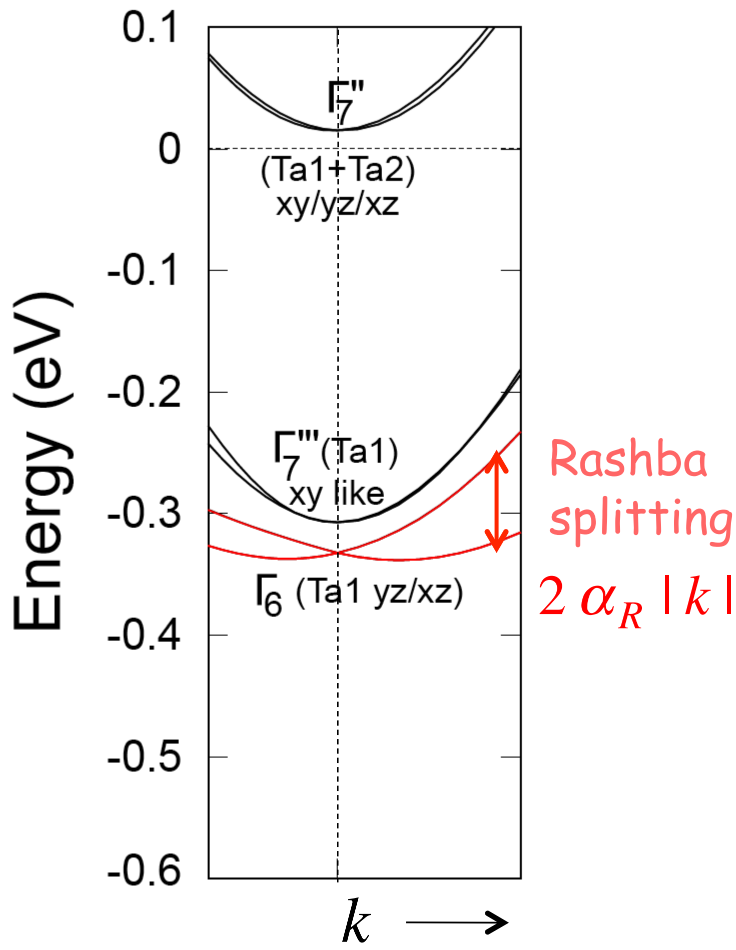
Rashba Hamiltonian: $H_R = \alpha_R(\vec{\sigma} \times \vec{k}) \cdot \hat{z} = \alpha_R(\sigma_x k_y - \sigma_y k_x)$



Ref: Rashba, Sov. Phys. Solid State (1960)

Manifestation of the Rashba effect

Theory: band structure

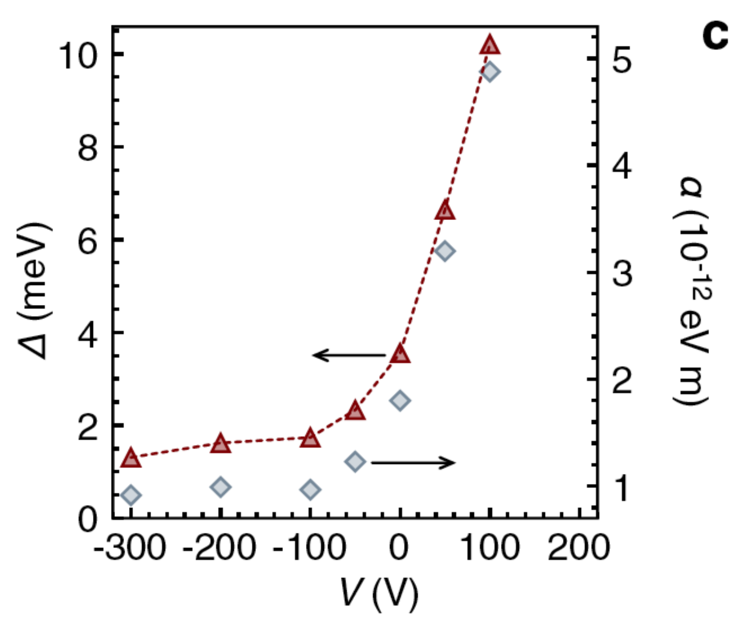


$$E_{\uparrow\downarrow} = ak^2 \pm \alpha_R |k|$$

Experiment

Can drive a wide range of novel phenomena

- ✓Spin tunneling, Spintronics, spin relaxation rates
- ✓Fulde-Ferrel-Larkin-Ovchinnikov (FFLO) state in superconductors
- ✓Topological p-wave superconductors
- ✓Cold atoms



Expt: spin relaxation rates in STO/LAO: Caviglia et al., PRL (2010)

Rashba effect: naïve derivation within the free electron model

1. Apply Electric field $\vec{E} = E\,\hat{z}$

2. Electron sees the magnetic field in its rest frame: $\vec{B} = (\vec{v} \times \vec{E}) / c^2$

3. B field couples to the magnetic moment of the electron spin:

$$H_{SO} = -\vec{M} \cdot \vec{B} = (\frac{g\mu_B \vec{\sigma}}{2}) \cdot (\frac{\vec{v} \times \vec{E}}{c^2}) = \alpha\, \vec{\sigma} \cdot (\vec{k} \times \vec{E})$$

4. Rashba Hamiltonian: $H_R = \alpha_R \vec{\sigma}.(\vec{k} \times \hat{z}) = \alpha_R(\sigma_x k_y - \sigma_y k_x)$

$$= \alpha_R\, (\vec{k} \times \vec{z}) \cdot \vec{\sigma}$$

$$= \vec{H}_k \cdot \vec{\sigma}$$

$$\vec{H}_k = \frac{\hbar^2 E}{2m^2 c^2} (\vec{k} \times \hat{z})$$

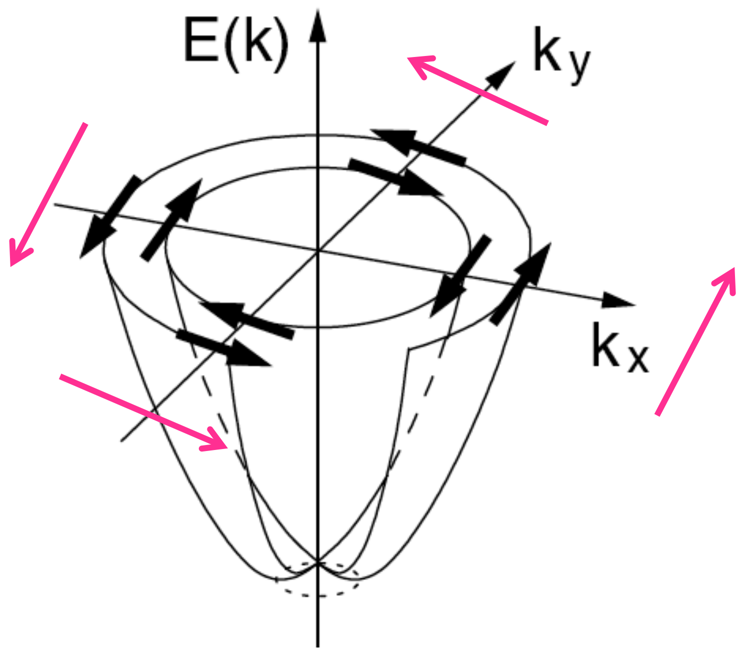
The electric field has been converted into a momentum-dependent magnetic field!

Rashba effect => k-dependent magnetic field

Rashba Hamiltonian: $$H_R = \alpha_R\,(\vec{k} \times \vec{E}) \cdot \vec{\sigma} = \vec{H}_k \cdot \vec{\sigma}$$

$$\vec{H}_k = \frac{\hbar^2 E}{2m^2c^2}(\vec{k} \times \hat{z})$$

$$\vec{E} = E\,\hat{z}$$

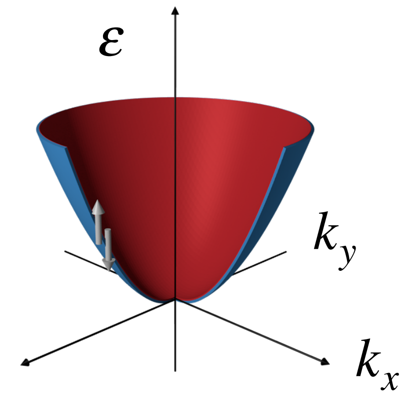


$\vec{H}_k$ Rashba Magnetic field

Rashba splitting

$$E_{\uparrow\downarrow} = ak^2 \pm \alpha_R\,|k|$$

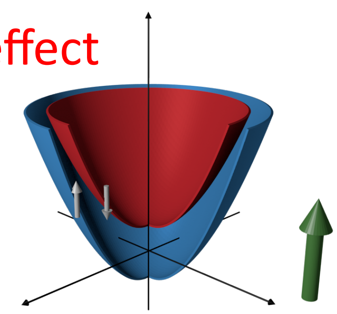
Broken symmetry and Rashba effect



Crystal inversion symmetry $\varepsilon(\vec{k}\uparrow) = \varepsilon(-\vec{k}\uparrow)$

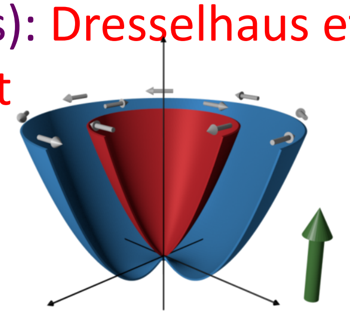
Time-reversal symmetry (Kramers Degeneracy) $\varepsilon(\vec{k}\uparrow) = \varepsilon(-\vec{k}\downarrow)$ ⇨ $\varepsilon(\vec{k}\uparrow) = \varepsilon(\vec{k}\downarrow)$

✓ Broken time-reversal symmetry (e.g. magnetic field) Zeeman effect



✓ Broken inversion symmetry

1. Bulk inversion symmetry broken (zinc blende AlAs): Dresselhaus effect
2. Surface inversion symmetry broken: Rashba effect



Spin-Orbit coupling is essential to couple the electron motion with spin

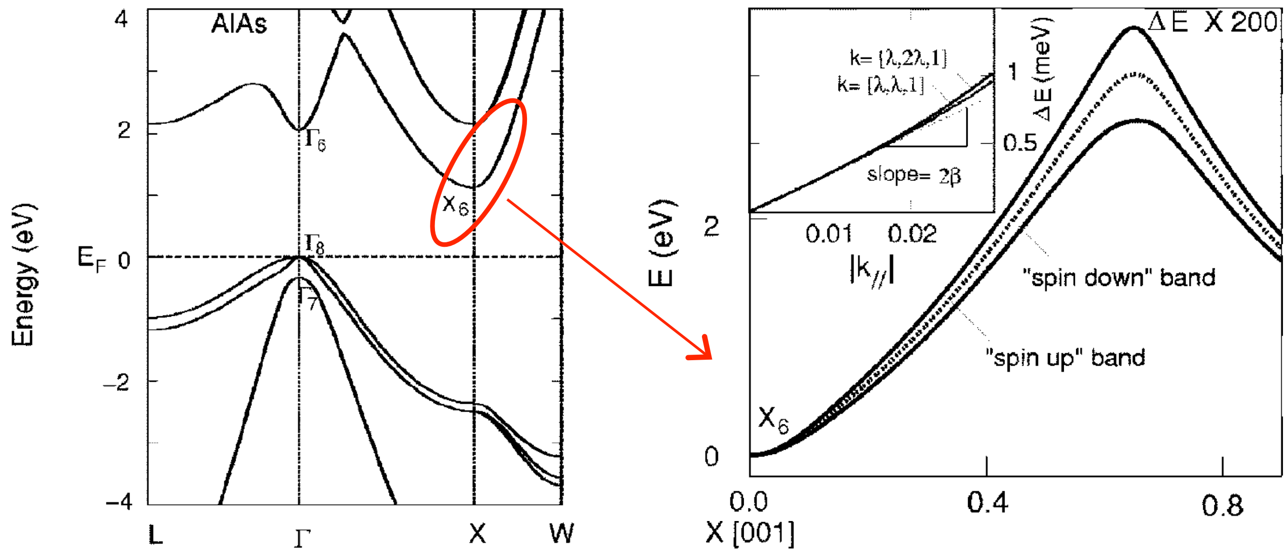
Bulk Inversion symmetry broken: Dresselhaus Effect

Dresselhaus Hamiltonian for the X point in zinc-blende crystals: $H_X = \beta\,(\sigma_x k_x - \sigma_y k_y)$

$\varepsilon_k = \pm\beta k$

$\beta = 10.9\ \text{meV}\cdot\mathring{A}$

AlAs zinc blende



DFT with Spin-orbit interaction

Material	AlP	AlSb	AlAs	GaP	Si
β (meV Å)	10.3	6.2	10.9	72.4	0
$m_\perp/m$	0.212	0.26	0.19	0.21	0.19
$m_\parallel/m$	3.67	1.0	1.1	7.25	0.92
Indirect gap (eV) (Γ_v-X_c)	2.5	1.69	2.23	2.35	1.17
Direct gap (eV) (Γ_v-Γ_c)	3.63	2.38	3.13	2.90	4.19
Conduction minimum	X	Δ	Δ	Δ	Δ

Mishra, Thulasi, SS, PRB 72, 195347 (2005)

G. Dresselhaus, Phys. Rev. 100, 580 (1955)

Surface Inversion symmetry broken: Rashba effect

The Rashba Hamiltonian: Naïve derivation

Electric field $-\vec{\nabla} V = E\,\hat{z}$

Spin-orbit coupling $H_{SO} = \dfrac{\hbar^2}{2m^2c^2}(\vec{\nabla} V \times \vec{k}) \cdot \vec{\sigma}$

$$H_R = \frac{\hbar^2 E}{2m^2c^2}(\hat{z} \times \vec{k}) \cdot \vec{\sigma} \qquad \varepsilon_k = \frac{\hbar^2 k^2}{2m} \pm \alpha_R k$$

$$\alpha_R = \frac{\hbar^2 E}{2m^2c^2} = \frac{(3.8 \text{ eV}\cdot\mathring{A}^2) \times (1 \text{ eV} / \mathring{A})}{0.5 \text{ MeV}} \sim 10^{-5} \text{ eV}\cdot\mathring{A}$$

1. Too small by two to three orders of magnitude of observed spin splitting

2. Violates Ehrenfest Theorem (for bound electrons)

Tight-Binding model for the Rashba splitting

- Hamiltonian has three parts:

$$H = H_{KE} + H_{SO} + H_E$$

Kinetic energy: $H_{KE} = \sum_{\langle ij\rangle\sigma} t_{ij} c^{\dagger}_{i\sigma} c_{j\sigma}$

Spin orbit coupling: $H_{SO} = \xi \boldsymbol{L}\cdot\boldsymbol{S}$

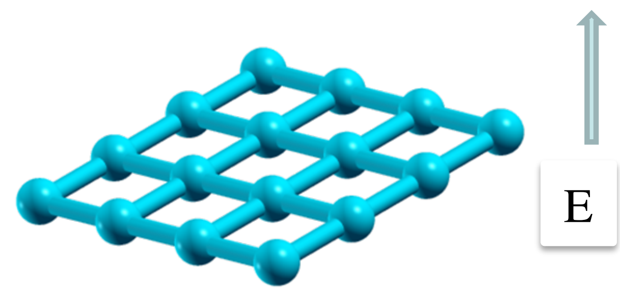
$$= \frac{\xi}{2}(L^{+}\sigma^{-} + L^{-}\sigma^{+} + L^{z}\sigma^{z})$$

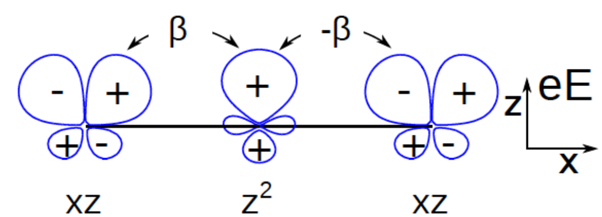
External electric field can cause finite hopping between orbitals that would be otherwise zero.

$$H_E = \begin{pmatrix} 0 & i\alpha_a sx & 0 & i\alpha_a sy & 0 \\ -i\alpha_a sx & 0 & i\alpha_b sy & 0 & i\alpha_c sy \\ 0 & -i\alpha_b sy & 0 & -i\alpha_b sx & 0 \\ -i\alpha_a sy & 0 & i\alpha_b sx & 0 & -i\alpha_c sx \\ 0 & -i\alpha_c sy & 0 & i\alpha_c sx & 0 \end{pmatrix}$$

H is a 10×10 matrix $\quad sx \equiv \sin x\,;\quad sy \equiv \sin y$

Cubic lattice with a surface and d electrons





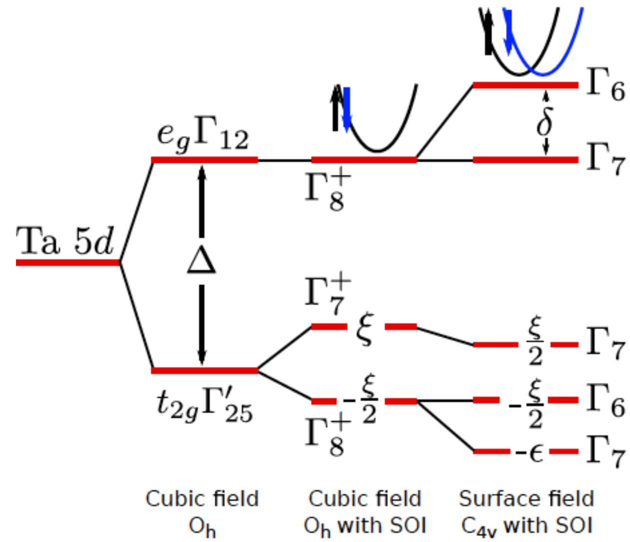
See also: Zhong, Toth, Held, PRB 87, 161102 (R) (2013);
Khalsa, Lee, MacDonald, PRB 88, 041302 (R) (2013)

Tight-Binding model for the Rashba splitting (focus on d electrons)

$$\mathcal{H} = \mathcal{H}_{\rm TB} + \mathcal{H}_{\rm SO} + \mathcal{H}_{\rm E}.$$

$$\mathcal{H}_{\rm TB}(\vec{k}) = \begin{pmatrix} V_\sigma(c_x+c_y+4c_z)/2+\Delta+\delta & -\sqrt{3}\,V_\sigma(c_x-c_y)/2 & 0 & 0 & 0 \\ -\sqrt{3}\,V_\sigma(c_x-c_y)/2 & 3\,V_\sigma(c_x+c_y)/2+\Delta & 0 & 0 & 0 \\ 0 & 0 & 2\,V_\pi\,(c_x+c_y)+\epsilon & 0 & 0 \\ 0 & 0 & 0 & 2\,V_\pi\,(c_x+c_z) & 0 \\ 0 & 0 & 0 & 0 & 2\,V_\pi\,(c_y+c_z) \end{pmatrix} \otimes \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}$$

$$\mathcal{H}_{\rm SO}(\vec{k}) = \frac{\xi}{2} \begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 0 & 0 & -\sqrt{3} & 0 & \sqrt{3}\,i \\ 0 & 0 & 0 & 0 & 0 & 0 & \sqrt{3} & 0 & \sqrt{3}\,i & 0 \\ 0 & 0 & 0 & 0 & -2\,i & 0 & 0 & 1 & 0 & i \\ 0 & 0 & 0 & 0 & 0 & 2\,i & -1 & 0 & i & 0 \\ 0 & 0 & 2\,i & 0 & 0 & 0 & 0 & -i & 0 & 1 \\ 0 & 0 & 0 & -2\,i & 0 & 0 & -i & 0 & -1 & 0 \\ 0 & \sqrt{3} & 0 & -1 & 0 & i & 0 & 0 & -i & 0 \\ -\sqrt{3} & 0 & 1 & 0 & i & 0 & 0 & 0 & 0 & i \\ 0 & -\sqrt{3}\,i & 0 & -i & 0 & -1 & i & 0 & 0 & 0 \\ -\sqrt{3}\,i & 0 & -i & 0 & 1 & 0 & 0 & -i & 0 & 0 \end{pmatrix}$$



$$\mathcal{H}_{\rm E}(\vec{k}) = 2i \begin{pmatrix} 0 & 0 & 0 & -\beta\sin k_x & -\beta\sin k_y \\ 0 & 0 & 0 & -\gamma\sin k_x & \gamma\sin k_y \\ 0 & 0 & 0 & \alpha\sin k_y & \alpha\sin k_x \\ \beta\sin k_x & \gamma\sin k_x & -\alpha\sin k_y & 0 & 0 \\ \beta\sin k_y & -\gamma\sin k_y & -\alpha\sin k_x & 0 & 0 \end{pmatrix} \otimes \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}$$

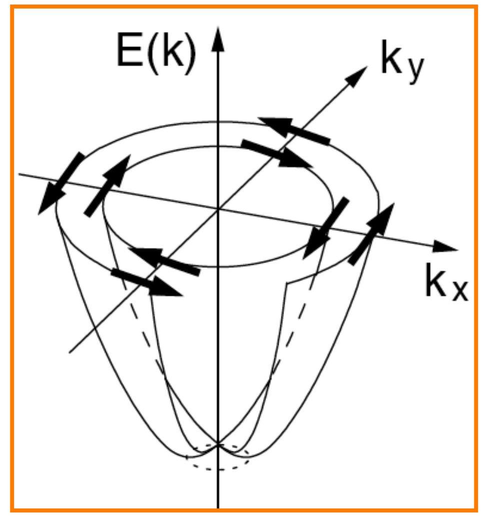
Use Löwdin downfolding to get an effective 2 x 2 Hamiltonian (Rashba) for each state such as Γ_6

$$h' = \begin{pmatrix} h'_{\uparrow\uparrow} & h'_{\uparrow\downarrow} \\ h'_{\downarrow\uparrow} & h'_{\downarrow\downarrow} \end{pmatrix} = \begin{pmatrix} \Delta & \alpha_R(k_y+ik_x) \\ \alpha_R(k_y-ik_x) & \Delta \end{pmatrix}$$

$$h' = \Delta + \alpha_R\,\vec{\sigma}.(\vec{k}\times\hat{z})$$

TB model: Rashba splitting is orbital dependent

Cubic Field (O_h)	Surface Field (C_{4v}) Symmetry	Partner functions	Rashba α_R/a
e_g $\Gamma_7^+(\Delta)$	$\Gamma_7(\Delta+\delta)$	$z^2\uparrow, z^2\downarrow$	$-2\sqrt{3}\beta\xi/\Delta$
	$\Gamma_6(\Delta)$	$x^2-y^2\uparrow, x^2-y^2\downarrow$	$-2\gamma\xi/\Delta$
t_{2g} Case 1. Weak SOI, $\xi \ll \lvert\varepsilon\rvert$			
Γ_7^+ (ξ)	$\Gamma_7(-\varepsilon)$	$xy\uparrow, xy\downarrow$	$2\alpha\xi/\varepsilon$
Γ_8^+ $(-\xi/2)$	$\Gamma_7'(\xi/2)$	$yz\downarrow + ixz\downarrow, yz\uparrow - ixz\uparrow$	$2\alpha\xi/\varepsilon$
	$\Gamma_6(-\xi/2)$	$yz\downarrow - ixz\downarrow, yz\uparrow + ixz\uparrow$	$2\sqrt{3}\beta\xi/\Delta$
t_{2g} Case 2. Strong SOI, $\xi \gg \lvert\varepsilon\rvert$, weak electric field $\lvert\alpha\rvert \ll \lvert\varepsilon\rvert$			
Γ_7^+ (ξ)	$\Gamma_7''(\xi-\varepsilon/3)$	$xy\uparrow + yz\downarrow + ixz\downarrow$, $xy\downarrow - yz\uparrow + ixz\uparrow$	$-4\alpha/3$
Γ_8^+ $(-\xi/2)$	$\Gamma_7'''(-\frac{3\xi+4\varepsilon}{6})$	$2xy\uparrow - yz\downarrow - ixz\downarrow$, $2xy\downarrow + yz\uparrow - ixz\uparrow$	$4\alpha/3$
	$\Gamma_6(-\xi/2)$	$yz\downarrow - ixz\downarrow, yz\uparrow + ixz\uparrow$	$2\sqrt{3}\beta\xi/\Delta$



Pseudo spins

$\Delta = t_{2g} - e_g$ splitting

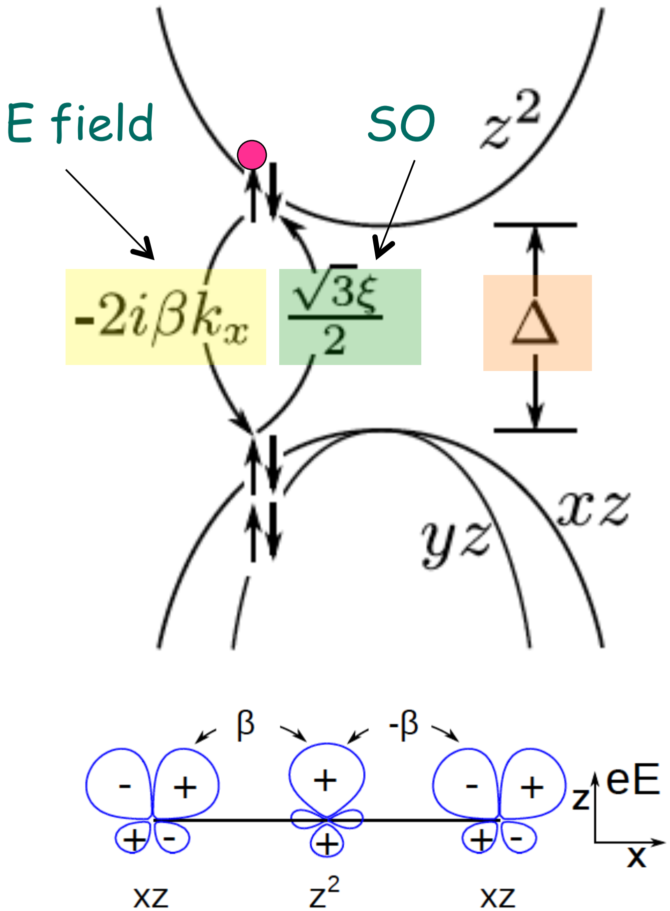
$\xi =$ SOC strength

$\alpha, \beta, \gamma =$ electric field induced hoppings

$\varepsilon = \varepsilon_{xy} - \varepsilon_{xz/yz} =$ surface asymmetry

$$H_R = \alpha_R (\vec{k} \times \hat{z}) \cdot \vec{\sigma}$$

Rashba Hamiltonian from 2nd order Perturbation Theory



For the conduction band state

$$h'_{\uparrow\downarrow} = \sum_i \frac{V_{\uparrow i} V_{i\downarrow}}{\Delta} = \frac{(-2\beta i k_x)(\sqrt{3}\xi/2)}{\Delta} + \frac{(-2\beta i k_y)(i\xi)}{\Delta}$$

$$= \alpha_R (k_y + i k_x)$$

$$\alpha_R = -2\sqrt{3}\beta\xi / \Delta$$

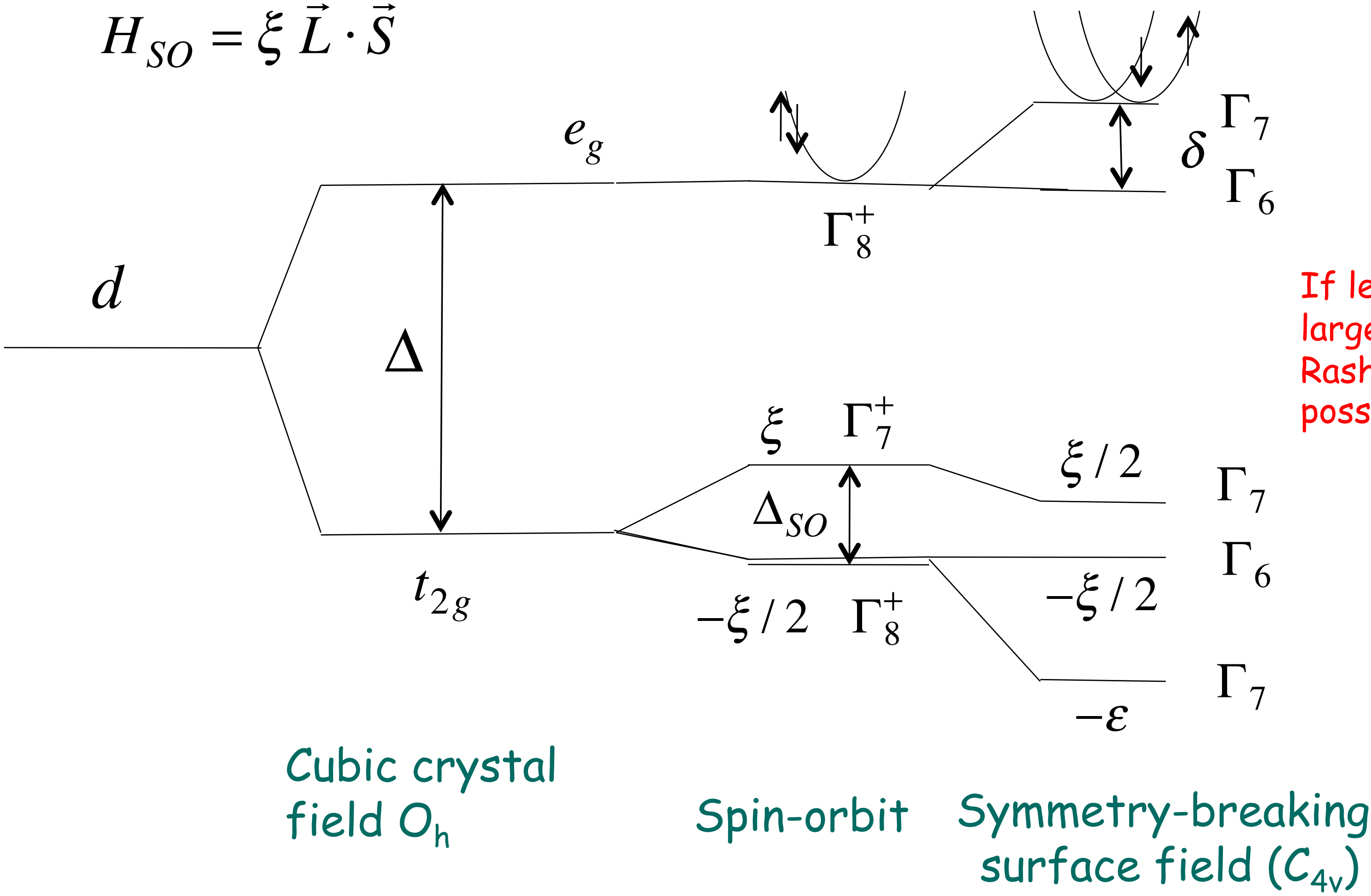
$$h' = \begin{pmatrix} h'_{\uparrow\uparrow} & h'_{\uparrow\downarrow} \\ h'_{\downarrow\uparrow} & h'_{\downarrow\downarrow} \end{pmatrix} = \begin{pmatrix} \Delta & \alpha_R(k_y + i k_x) \\ \alpha_R(k_y - i k_x) & \Delta \end{pmatrix}$$

$$h' = \Delta + \alpha_R\, \vec{\sigma}.(\vec{k} \times \hat{z})$$

Both the broken inversion symmetry (β) and
the SOC term (ξ) must be present for the Rashba effect.

Rashba form of the Hamiltonian not always good

$$H_{SO} = \xi \vec{L} \cdot \vec{S}$$



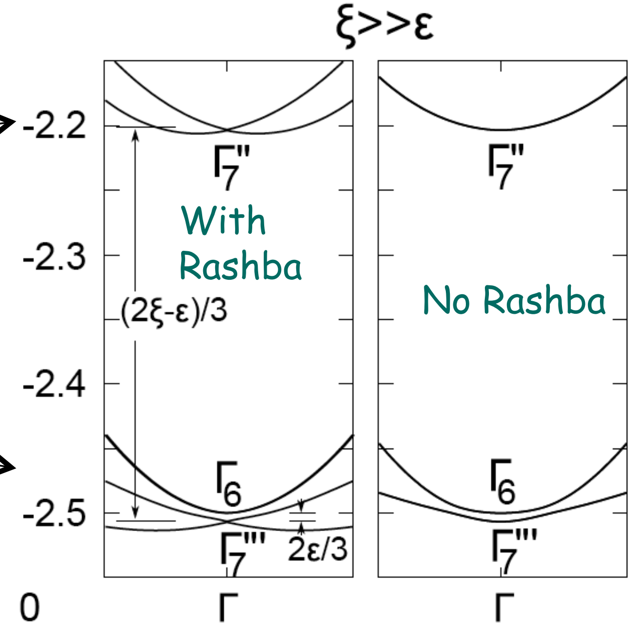
If level separation is not large enough, a 2 x 2 Rashba Hamiltonian not possible.

Rashba splitting in nearly-degenerate case

$h' = \Delta + \alpha_R \, \vec{\sigma}.(\vec{k} \times \hat{z})$

Rashba 2 x 2 Hamiltonian

Rashba 4 x 4 Hamiltonian



Case : $\varepsilon \sim \alpha$

ξ = SOC strength

$\varepsilon = \varepsilon_{xy} - \varepsilon_{xz/yz}$

α = E field induced hopping

$$\mathcal{H}_R = \frac{2}{3}\begin{pmatrix} ak^2+\varepsilon & 2\alpha\, k_+ & c\bar{k}^2 & -\sqrt{3}\alpha\, k_+ \\ 2\alpha\, k_- & ak^2+\varepsilon & \sqrt{3}\alpha\, k_+ & -c\bar{k}^2 \\ c\bar{k}^2 & \sqrt{3}\alpha k_- & bk^2 & \frac{3\sqrt{3}\beta\xi}{\Delta}k_+ \\ -\sqrt{3}\alpha\, k_+ & -c\bar{k}^2 & \frac{3\sqrt{3}\beta\xi}{\Delta}k_- & bk^2 \end{pmatrix}$$

Subband Structure of a Two-Dimensional Electron Gas Formed at the Polar Surface of the Strong Spin-Orbit Perovskite $KTaO_3$

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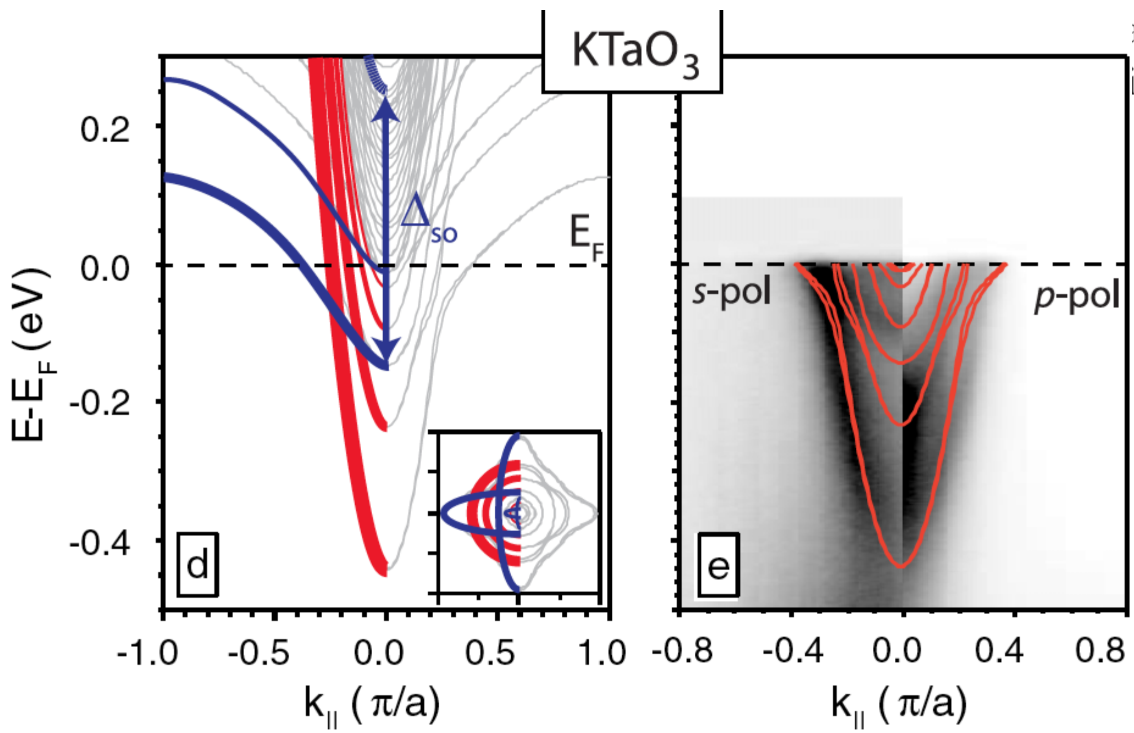
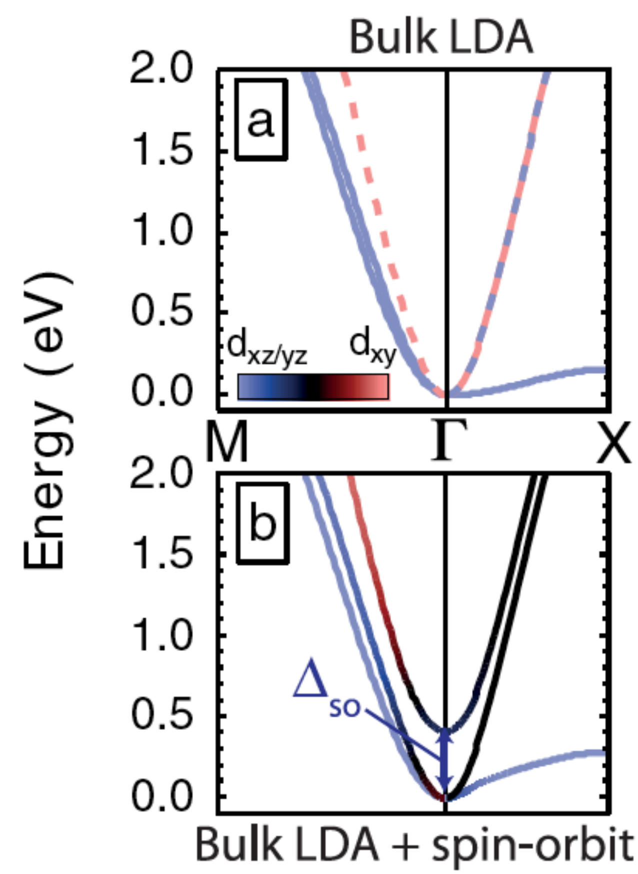
[1]*SUPA, School of Physics and Astronomy, University of St. Andrews, St. Andrews, Fife KY16 9SS, United Kingdom*
[2]*Advanced Light Source, Lawrence Berkeley National Lab, Berkeley, California 94720, USA*

as (2DEG) ...
ion,





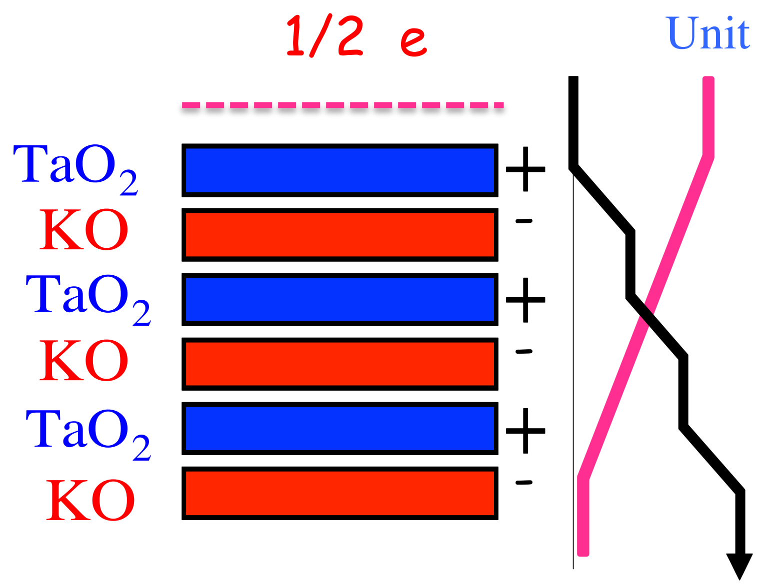
Looked for Rashba: Found small $\Delta k_{\|} \sim 0.02\ \text{Å}^{-1}$

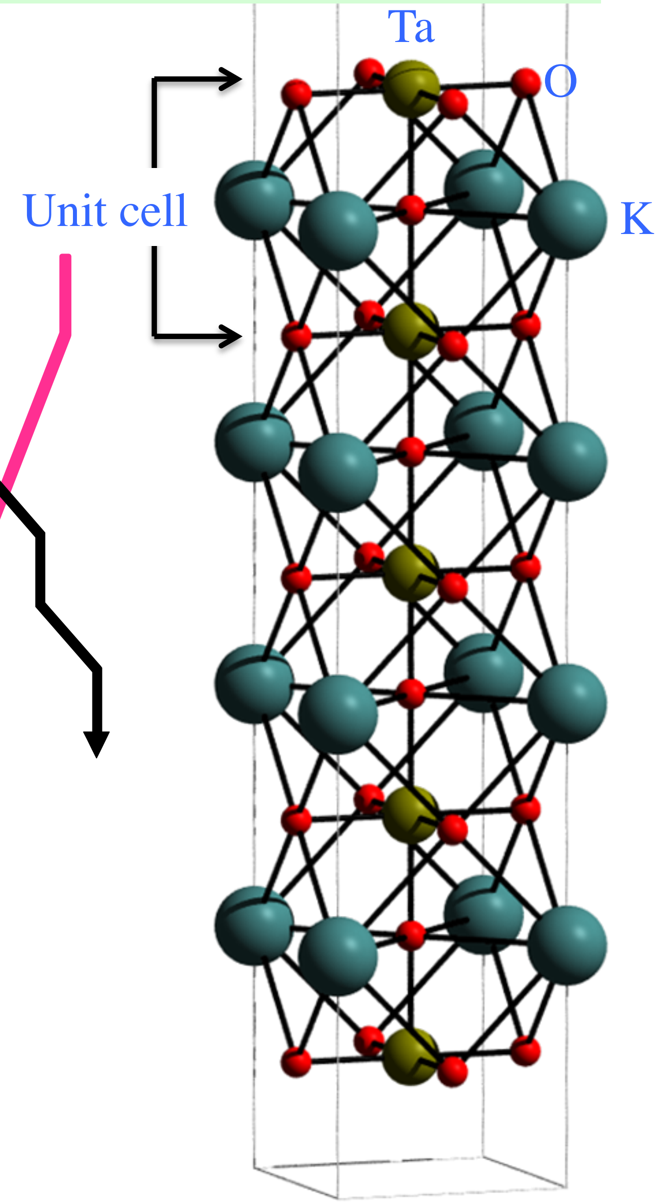
Polar catastrophe and the origin of the 2DEG at the $KTaO_3$ surface

Polar Catastrophe

Chemical valence $K^{+}Ta^{5+}O_3^{6-}$ => TaO_2^{+} and KO^{-} layers. This again leads to diverging potential.

The polar catastrophe is satisfied by inserting ½ e^{-} at the surface.

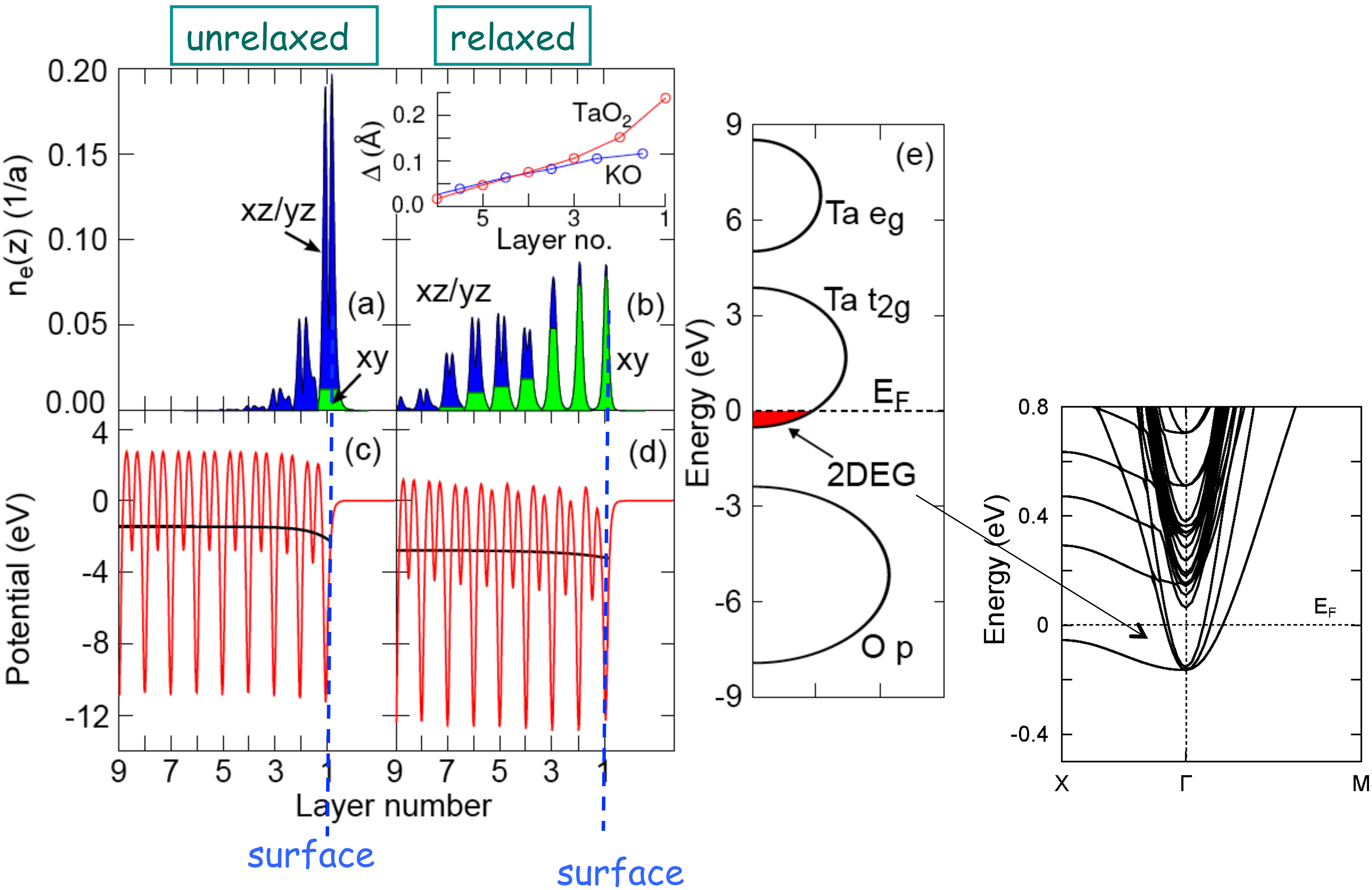




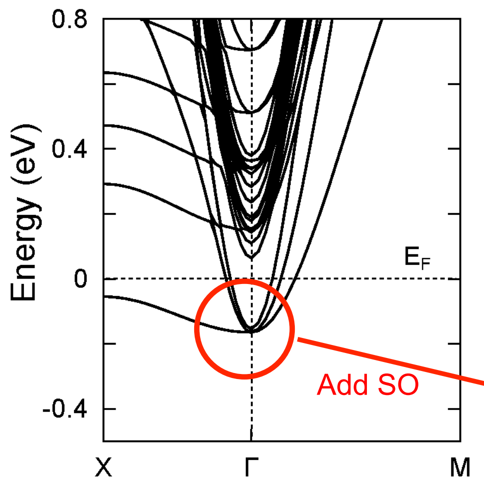
W. Harrison (1978)

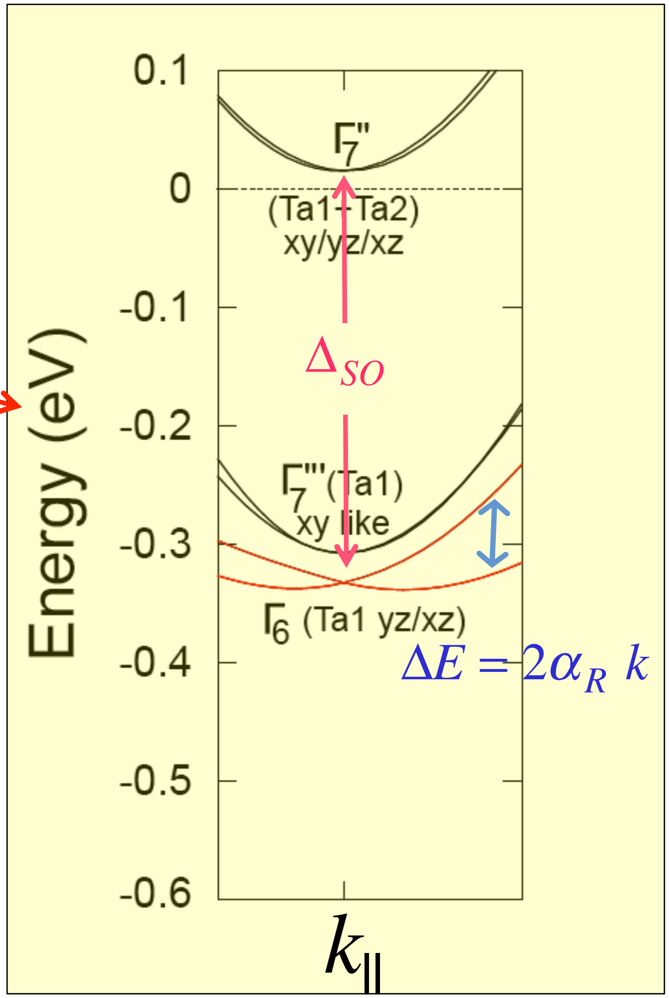
Origin of 2DEG at LAO/STO oxide interfaces:
Ohtomo and Hwang, Nature (2004)
Popovic, SS, Martin, PRL 101, 256801(2008)

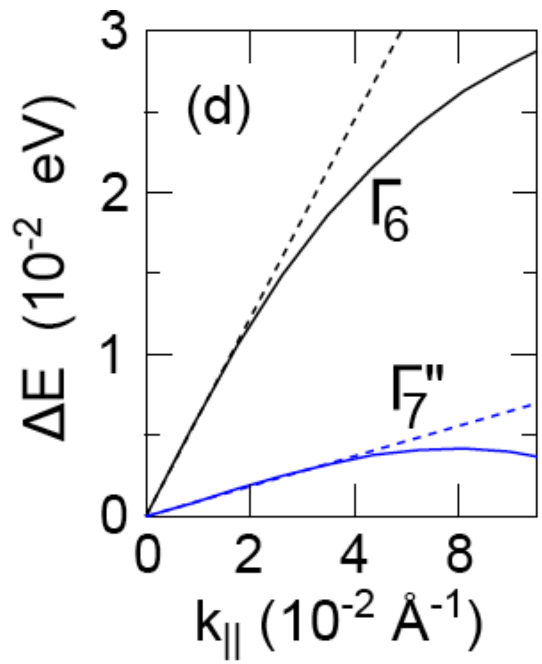
2DEG at the KTO surface: DFT results



Rashba Splitting of the KTO surface bands





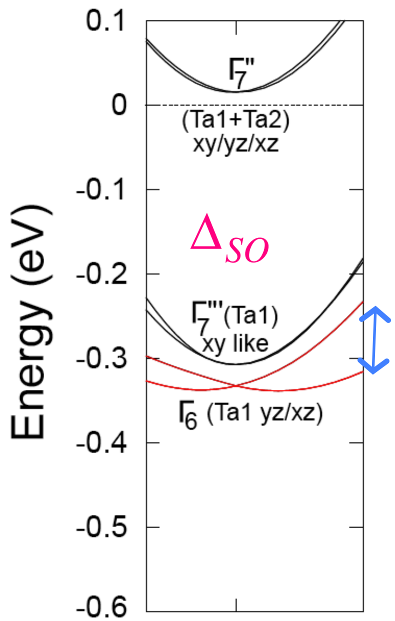


$$\varepsilon_k = \frac{\hbar^2 k^2}{2m} \pm \alpha_R k$$

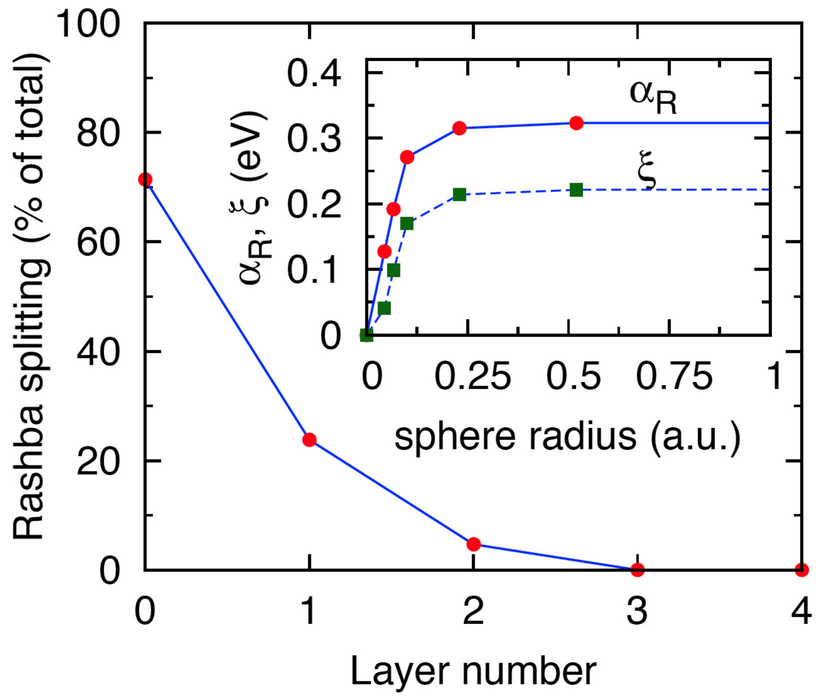
Where does the Rashba effect come from? : The nuclear region in the surface layers

$$H_{SO} = \frac{1}{m^2 c^2 r} \frac{\partial V}{\partial r} \vec{L} \cdot \vec{S} = \xi \, \vec{L} \cdot \vec{S}$$

Retain SO coupling only on a specific surface layer or only within radius r from the nucleus



$$\Delta E = 2\alpha_R \, k$$

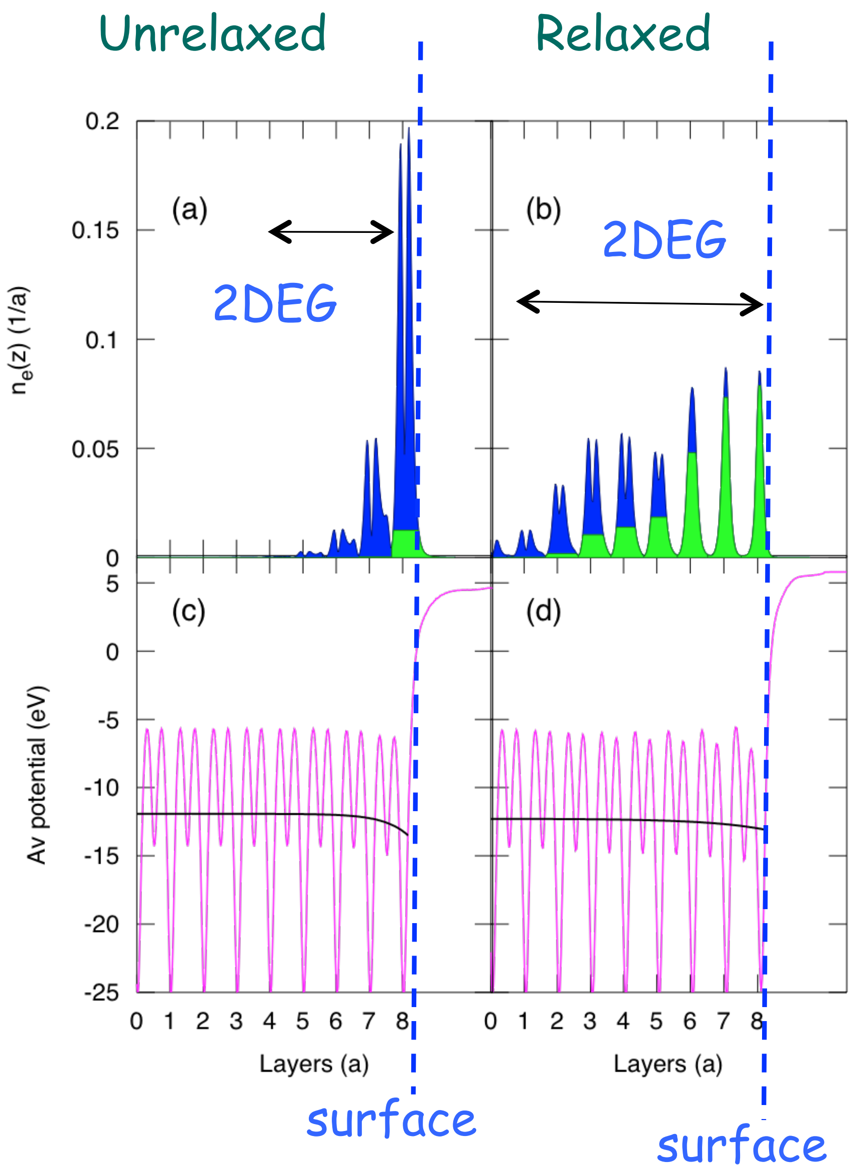


Origin of Rashba Splitting

(a) From electric fields near the nucleus and

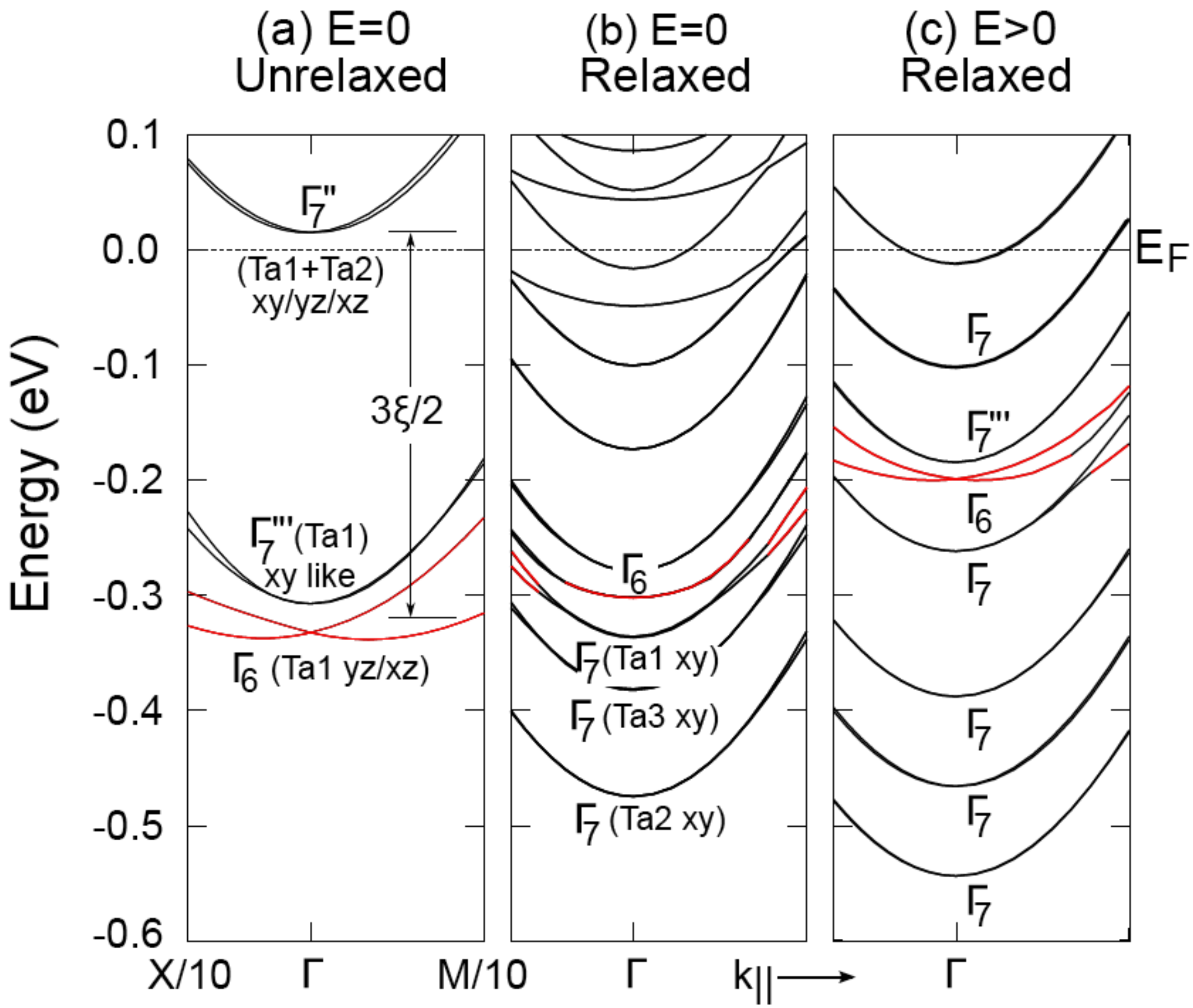
(b) Broken inversion symmetry in the first few surface layers

2DEG profile due to surface relaxation

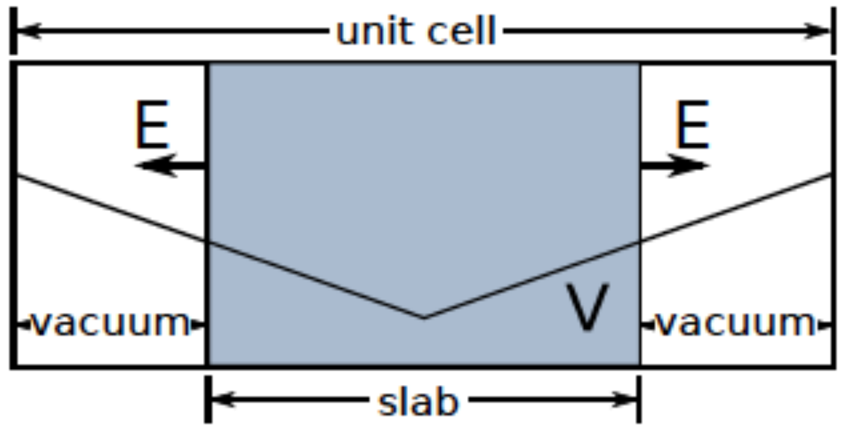


Relaxed structure: Surface electric field becomes weaker due to ionic screening and the 2DEG spreads deeper into the bulk

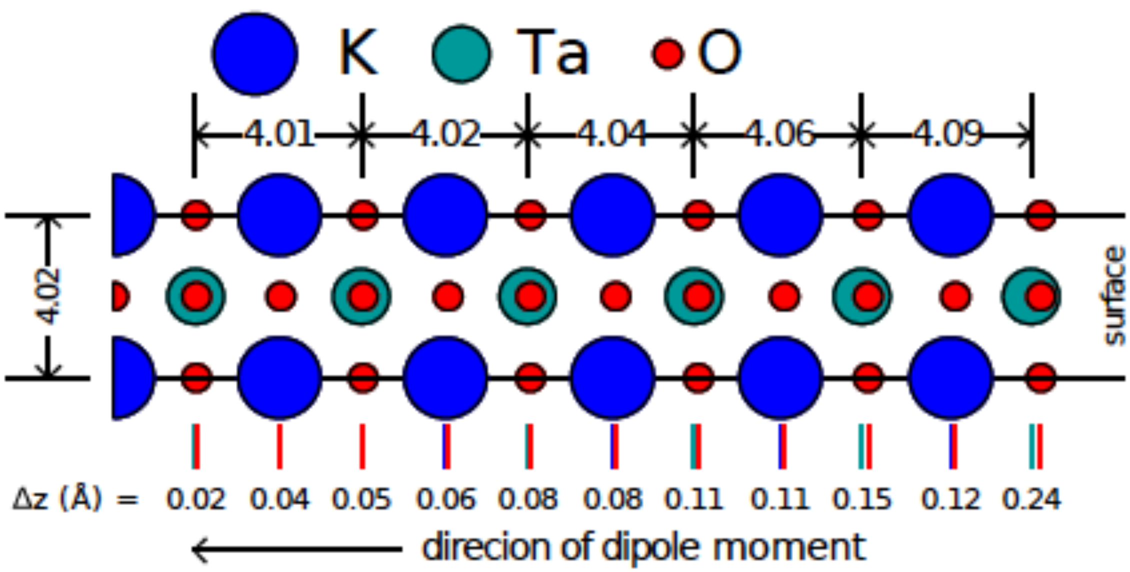
Tuning of the Rashba effect by E field: DFT results



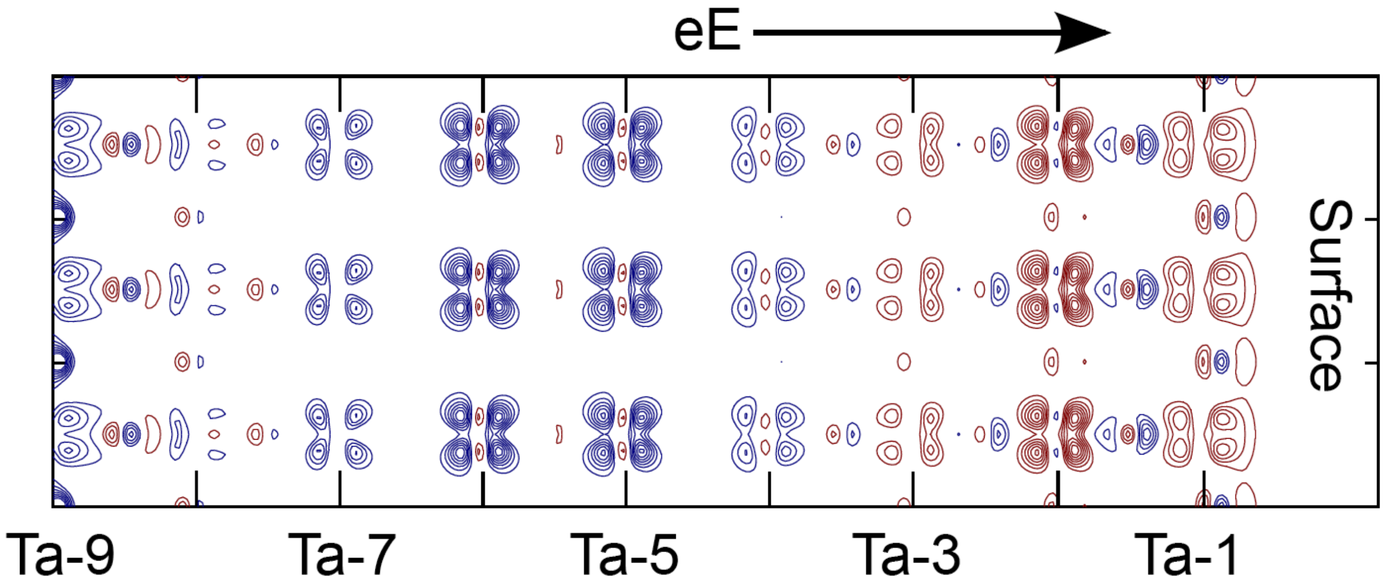
DFT Calculations of the Rashba Effect with Electric field



17 KTO layers slab
Two identical surface

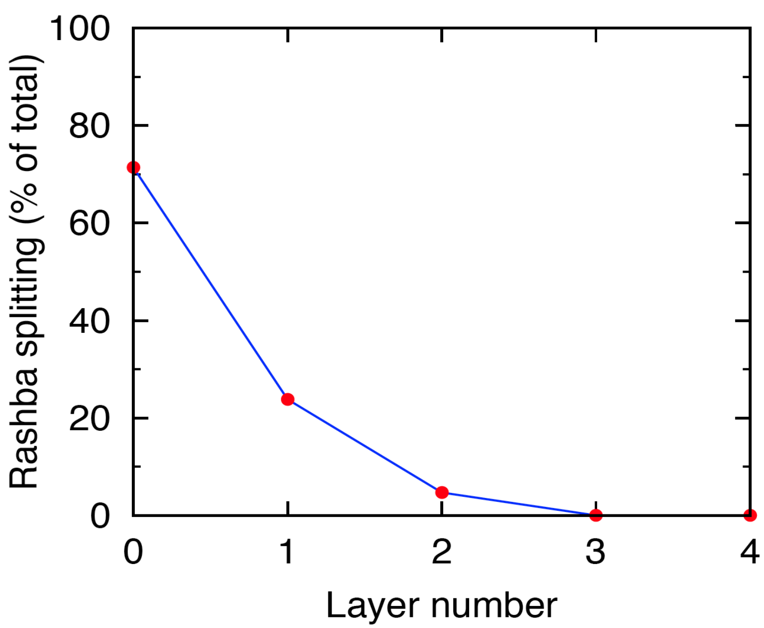


Control of Rashba Effect by an electric field



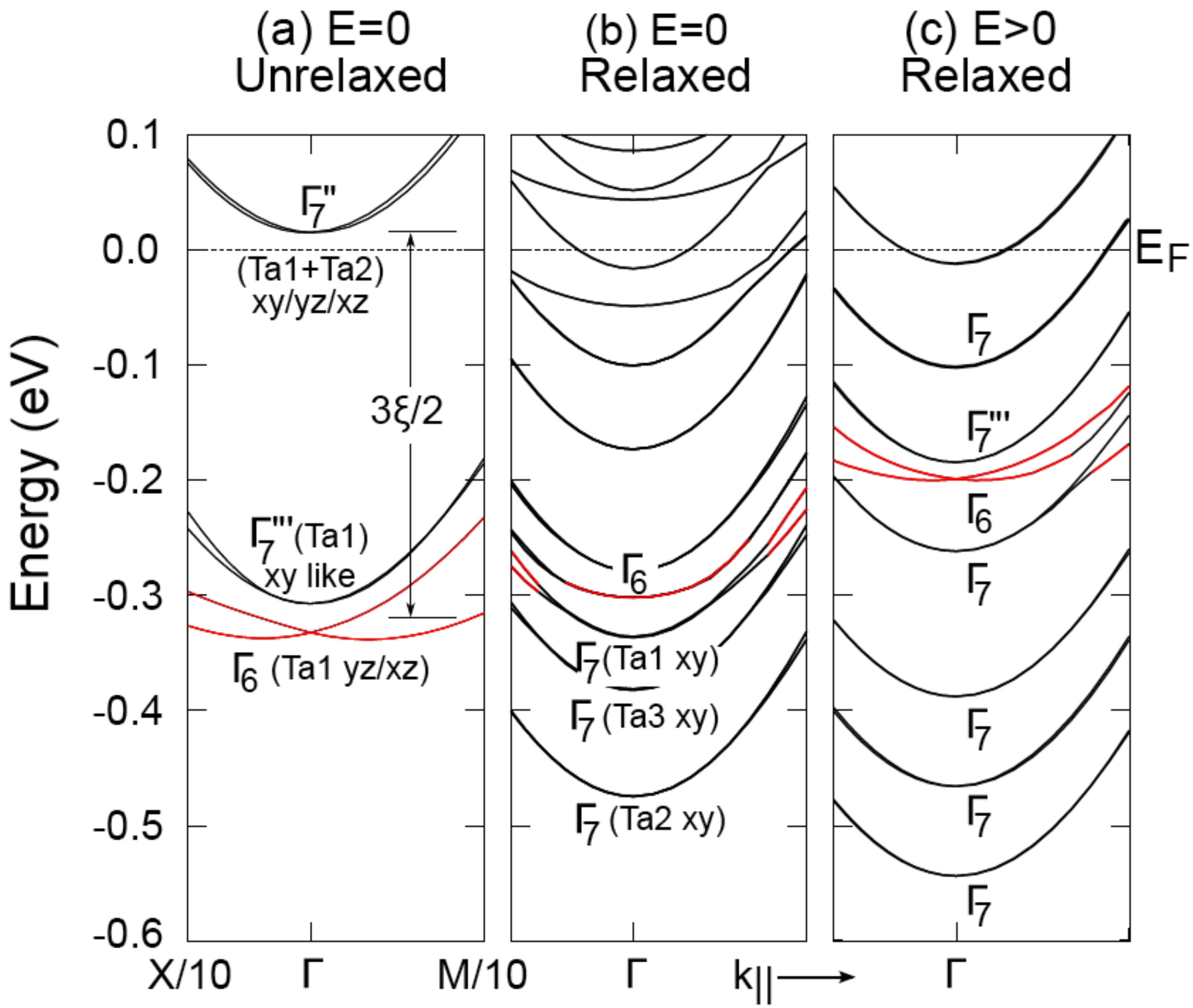
Violet: increase in electron density
blue: decrease

2DEG at the surface can be moved in and out by an applied electric field

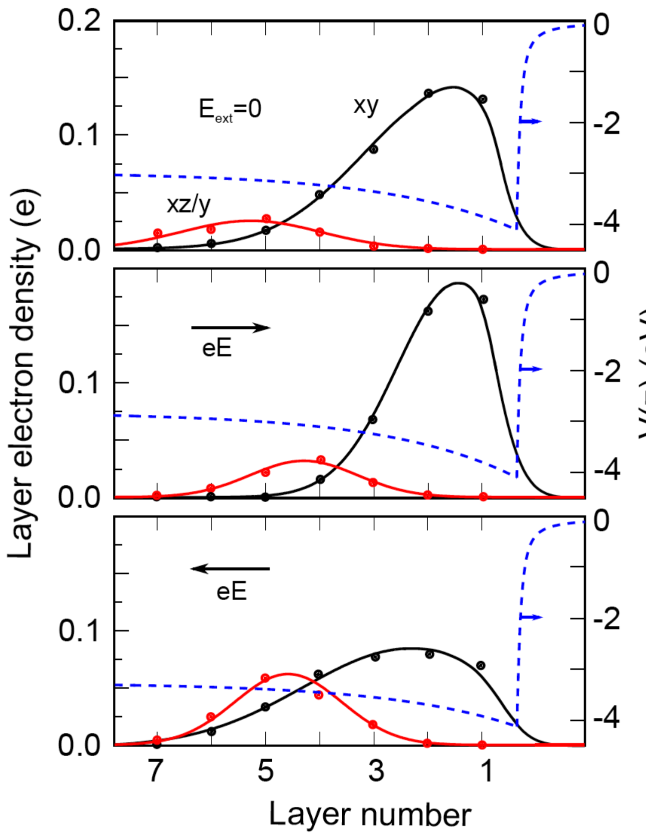


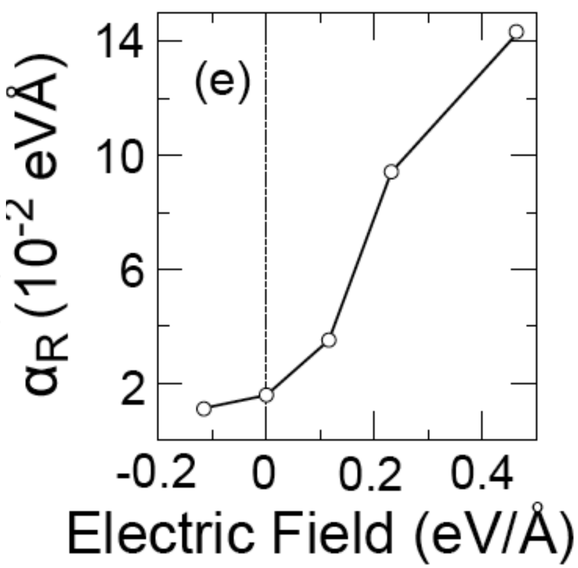
Rashba effect comes from the first couple of surface layer electrons

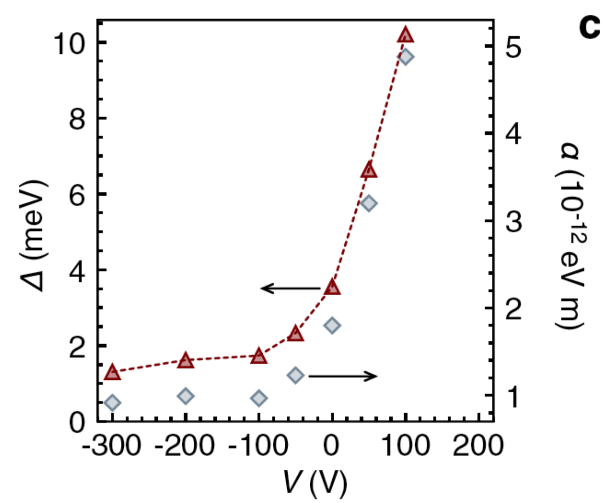
Tuning of the Rashba effect by E field: DFT results



Tuning of the Rashba effect by the electric field



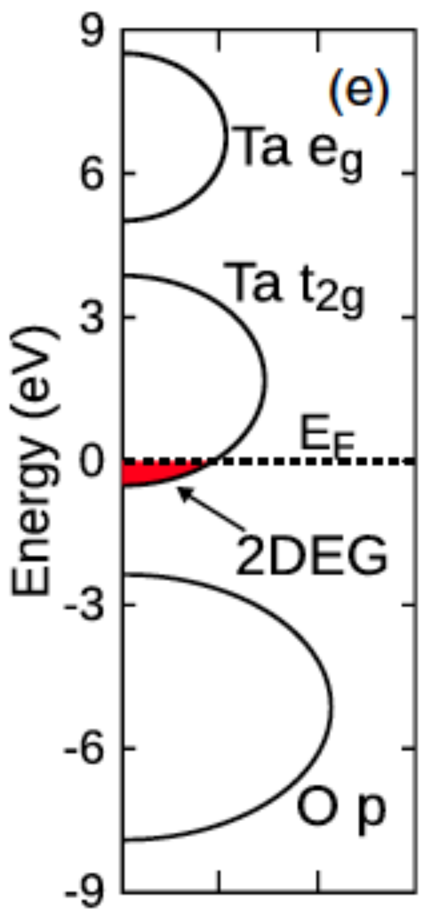


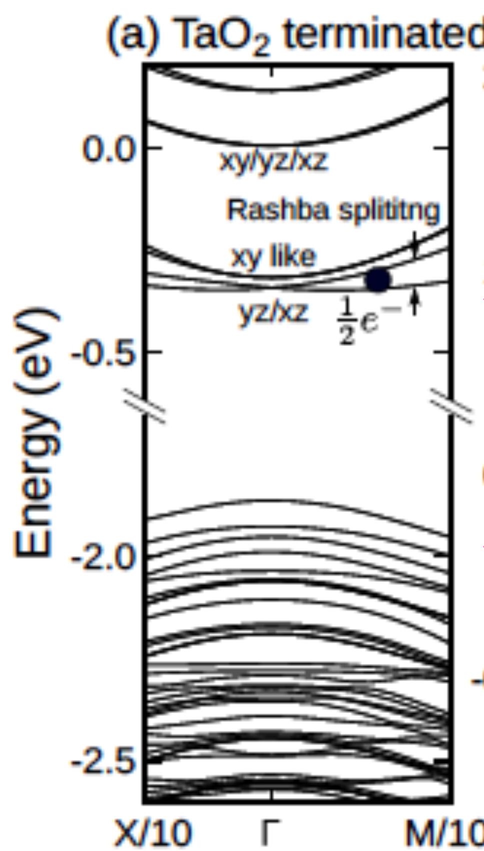


Expt: STO/LAO: Caviglia et al., PRL (2010)

Measured α from electron spin relaxation rates

So far: TaO_2-terminated Surface

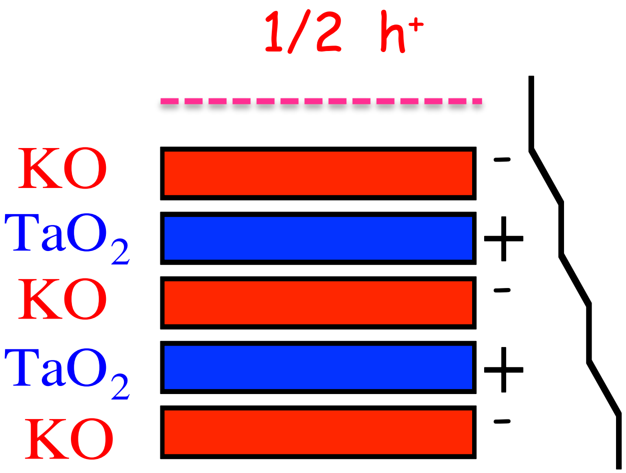


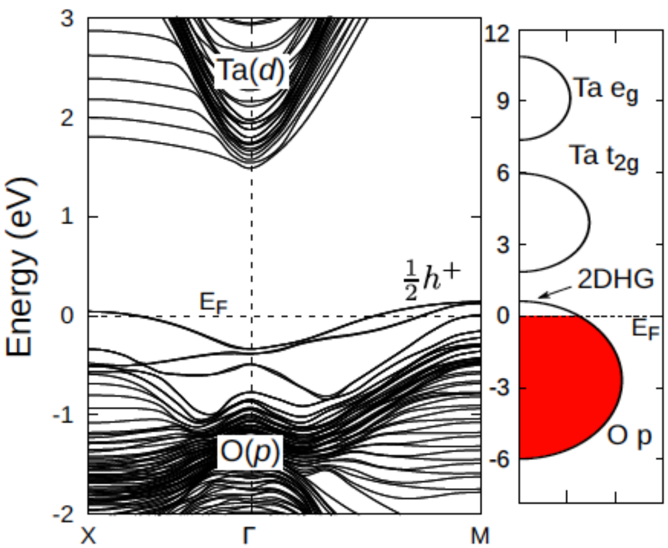


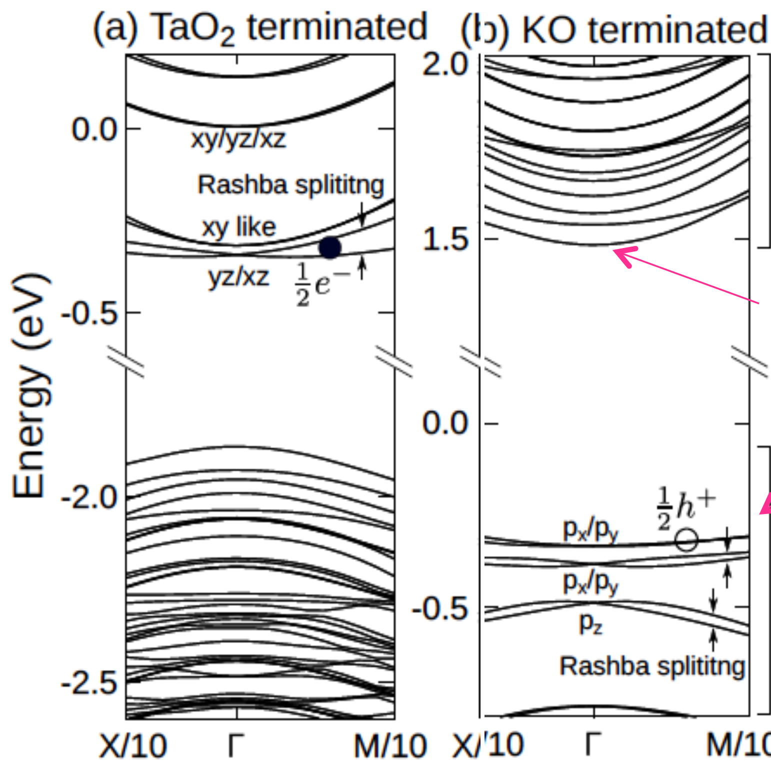
Strong Rashba (2DEG confined to surface)

Weak Rashba (weak SOI, O (p) states delocalized into the bulk)

KO-terminated Surface (2D Hole gas)







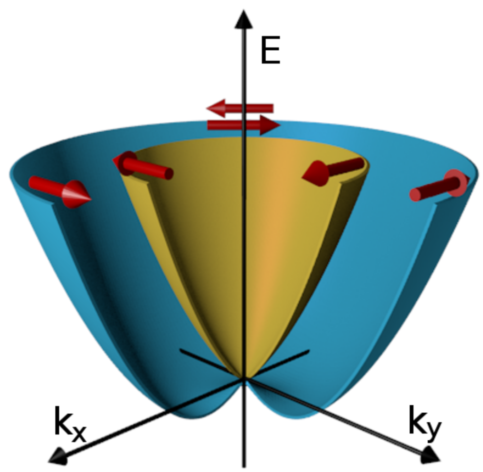
Rashba splitting much smaller, as conduction states not confined to the surface.

Rashba splitting much stronger, as the O(p) holes are confined to the surface

DFT bands: Ideal surface

Summary

- ✓ Rashba spin splitting: TB model serves as a framework for undersanding of the Rashba effect.



- ✓ Origin of the effect: First few surface layers and mostly from spin-orbit coupling in the nuclear region

- ✓ Controlled by several factors
  - ➢ spin-orbit coupling strength
  - ➢ Strength of the broken-inversion symmetry
  - ➢ Gate control achieved via

    (i) the 2DEG moving in and out of the surface region

    (ii) Reorganization of the xy/yz/zx energies at the surface.

- ✓ DFT results for $KTaO_3$ surface => Oxides are rich systems for manipulating the Rashba effect with the electric field