Signature of cubic and linear Rashba in LaAlO$_3$/KTaO$_3$ (0 0 1) heterostructure

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The LaAlO$_3$/KTaO$_3$ system serves as a prototype to study the electronic properties that emerge as a result of spin-orbit coupling. In this article, we have used first-principles calculations to systematically study two types of defect-free (0 0 1) interfaces, which are termed as Type-I and Type-II. While the Type-I heterostructure produces a two dimensional electron gas, the Type-II heterostructure hosts an oxygen-rich two dimensional hole gas at the interface. Furthermore, in the presence of intrinsic spin-orbit coupling, we have found evidence of both cubic and linear Rashba interactions in the conduction bands of the Type-I heterostructure. On the contrary, there is spin-splitting of both the valence and the conduction bands in the Type-II interface, which are found to be only linear Rashba type. Interestingly, the Type-II interface also harbors a potential photocurrent transition path, making it an excellent platform to study the circularly polarized photogalvanic effect.

1. INTRODUCTION

Spin based electronics has taken the lead in the fast expanding field of quantum technologies for carrying information by spin instead of charge [1]. These energy-efficient devices necessarily require manipulation of local spins for efficient processing of quantum information. In recent years, manipulation of the spin degree of freedom in solid-state materials via spin-orbit coupling, especially the Rashba spin-orbit coupling, has emerged as one of the most promising approach for developing such next generation energy-efficient electronic devices [2, 3]. Dresselhaus and Rashba were the first to observe that the combined effect of the intrinsic spin-orbit coupling (SOC) and the bulk inversion asymmetry could lead to spin-split energy bands in non-centrosymmetric zincblende or wurtzite semiconductors [4, 5]. Later, Bychkov and Rashba [6] discovered that spin-split energy bands can also appear in two-dimensional materials due to structural inversion asymmetry of the confining potential, which is now commonly referred as Rashba spin-orbit (RSO) interaction. The advantage of controlling the RSO interaction in a material by an external electric field holds the promise to resolve the designing issues of the spintronic devices which typically arises due to the inclusion of local magnetic field [7, 8]. The motivation of designing spin-based electronic devices without the ferromagnetic elements has given rise to the rapidly emerging field of spin-orbitronics [9, 10].

The most effective method of creating spin current inside the non-magnetic materials is by spin-charge interconversion. The electrically induced regulation of the spin dynamics through spin-orbit coupling is the most practical and desirable method of achieving this interconversion [11, 12]. The principle of spin-orbitronics was first explored in semiconductors and metals, though within some years oxide-heterostructures have emerged to be the most promising platform with a plethora of unique characteristics [13]. Complex oxide heterostructures are widely studied for their numerous fascinating properties, such as 2D superconductivity, coexistence of ferromagnetism & superconductivity, colossal magnetoresistance etc. [14-17], and their promising applications in the manufacturing of next generation all-oxide solid state devices [18, 19]. The interface of a heterostructure made of complex oxide perovskites breaks the structural inversion symmetry and therefore the electron/hole gas confined at the interface experiences a potential gradient perpendicular to the conduction plane [20]. Oxide interfaces offer a flexible platform for creating, controlling, and detecting spin currents or spin textures. SrTiO$_3$ based surfaces and interfaces are first observed as the promising candidate for creating two dimensional electron gas (2DEG) with large transport properties and highly confined quantum well [21, 22]. As SrTiO$_3$ has strongly correlated 3d-orbital, the effect of intrinsic SOC is expected to be strong in those surfaces and interfaces making the system a suitable platform for generating Rashba spin splitting [24]. The first evidence for RSO interaction in oxide surfaces indeed emerged in the 2DEG generated at the SrTiO$_3$ surface along the (0 0 1) direction [25]. Irrespective of various interesting SO-based properties in LaAlO$_3$/SrTiO$_3$ heterostructure, one of the main difficulty with this system is the presence of ferromagnetism at the interface, and the RSO interaction is dependent on the magnetoresistance tuning [26].

To overcome this situation, there is an ongoing effort to create oxide heterostructure with non-magnetic interface and stronger SOC, and KTaO$_3$ has emerged as an exciting alternative for its novel electronic as well as spintronic properties due to the presence of the 5d-orbital
and high Z-value. The carrier dependence of spin precision length is shorter in KTaO$_3$ than that in SrTiO$_3$. Due to the polar nature of KTaO$_3$, the surface with TaO$_{2-}$ or KO$^-$ termination gives rise to naturally induced 2DEG and its counterpart two-dimensional hole gas (2DHG) at the interfaces. The in-built electric field at the surface of KTaO$_3$ provides an ideal system for studying RSO and recently experimental evidences of spin splitting have been found in the surface bands. In comparison to LaAlO$_3$/SrTiO$_3$ heterostructure, LaAlO$_3$/KTaO$_3$ has larger charge carrier density at interface with high electron mobility as LaAlO$_3$ is also a polar material. Besides, in the bulk state, KTaO$_3$ and LaAlO$_3$ are cubic with space group Pm3m (No. 221). Due to the comparable band gaps and lattice parameters, LaAlO$_3$ and KTaO$_3$ are considered as very good candidates for creating heterostructure with an effective charge accumulation at the interfaces. Recently, there have been theoretical and experimental studies of LaAlO$_3$/KTaO$_3$ heterostructures. However, detailed first-principle study on the effect of intrinsic SOC and the possibility of RSO interaction has remained unexplored.

Motivated by this, in this article, we have investigated the electronic and spintronics properties of the LaAlO$_3$/KTaO$_3$ heterostructure for two possible interfaces, which are TaO$_{2-}$/LaO$^+$ (Type-I) and KO$^-$/AlO$_{2-}$ (Type-II) using density functional theory (DFT) based first-principle calculations. We have found 2DEG in Type-I heterostructure whereas Type-II system exhibit 2DHG in its interface. In the presence of SOC, Type-I system produce both the cubic and linear RSO splitting which has been originated from conduction bands. On the contrary, Type-II heterostructure predominantly produce large k-linear RSO coupling strength which has been produced from both the conduction bands and valence bands. As earlier reported in Ref. [39], the production of circularly polarized photocurrent is possible in KTaO$_3$ slab system where giant RSO interaction is present. Following this, we have found a possible photocurrent transition route for the Type-II system.

In the present report, first we have discussed structural and computational details in section 2. In Section 3 we have discussed on the effective Hamiltonian for the C$_{4v}$ little point group. Following this we have systematically reported the Type-I and Type-II system results in Section 4. The orbital contribution in the absence of SOC has been discussed in subsections and 4.3. The explanation of RSO and spin texture for both type systems is covered in sections 4.2 and 4.4. In section 5 we finally come to a conclusion.

2. STRUCTURAL AND COMPUTATIONAL DETAILS

In Fig. [1] we present the asymmetric slab model along (0 0 1) made of (KTO)$_{6.5}$ and (LAO)$_{2.5}$ with two types of defect-less interfaces. No strain has been applied in the system. The perovskite oxide material with parent formula ABO$_3$ can be stacked as BO$_2$ and AO. KTaO$_3$ substrate comprises of alternative KO$^-$ and TaO$_{2-}$ layer, whereas LaAlO$_3$ epitaxial layers are made of LAO$^+$ and AlO$_{2-}$ sub-layers. To maintain the interface stoichiometry there are two possible kind of interfaces in the heterostructure created by LaAlO$_3$ and KTaO$_3$ along (0 0 1) direction, i.e., TaO$_{2-}$/LaO$^+$ and KO$^-$/AlO$_{2-}$. Due to the asymmetry present in the heterostructures, both the systems correspond to P4mm (No. 99) space group, which does not have spatial inversion symmetry or the mirror symmetry.

The first-principle density functional calculations are performed by Quantum Espresso (QE) package [40]. The exchange-correlation of the electron interactions are taken into account by Perdew-Burke-Ernzerhof (PBE) functional [41]. The projector augmented wave (PAW) basis set has been used to include the interaction between valence electrons and core ions [42]. The on site Coulomb interactions of Ta-5d orbitals are considered by using the standard PBE+U method [43]. The effective value U$_{eff}$ = 3 eV is employed for Ta-5d states in this work, as it is well established that such a value is appropriate to describe the strongly-correlated states of KTO [44]. Using linear response theory implemented in the QE package we have independently verified the U value of the Ta-5d states for our heterostructures. The plane-wave basis with a cut-off energy of 75 Ry is used to expand the electronic wavefunctions. Full geometry optimizations are performed using the Broyden-Fletcher-Goldfarb-Shann (BFGS) algorithm [45]. Here, Γ-centered k-point grids for sampling [46] the first Brillouin zone are set to 8 × 8 × 1 for 1 × 1 × (m + n) slab models of LAO/KTO heterostructures where m and n are the number of sublayers of KTaO$_3$ and LaAlO$_3$. Scalar relativistic effect has been taken for optimization of the structure and later on fully relativistic effect has been included in selected pseudopotentials to study the SOC.
effect. It has been tested that there is no significant effect of SOC in the structure optimization. The bottom layer has been kept fixed to replicate a really thick substrate. As shown in Fig. 1 in the Type-I heterostructure the bottom two atomic sublayers KO and TaO₂ are kept fixed during structural optimization, while only the bottom TaO₂ sublayer is considered to be the fixed in case of the Type-II heterostructure. To minimize the interaction between neighboring slabs, a vacuum layer with a thickness of ≥ 30 Å is applied along the z direction (out of plane). All atoms except those in the fixed sublayer/s are fully relaxed until the force acting on each atom is < 10⁻³ Ry/Bohr. The convergence criteria of the total energy is set to be < 10⁻⁴ Ry/Bohr. Spin-polarized calculations has been done on the slab system to check the collinear magnetism, which has been found to be zero, and hence all the SOC calculations have been conducted in the slab system by setting the initial magnetism on Ta as 0 μB. The experimental lattice parameters of bulk LaAlO₃ and KTaO₃ are 3.790 Å and 3.989 Å [77], whereas by using PBE type pseudopotential we have found the theoretical lattice parameters of KTO and LAO to be 4.02 Å and 3.81 Å, which has also been reported earlier [30]. For the heterostructures, we use an average lattice constant of 4.02 Å for both the KTO and LAO regions. The theoretical band gaps of LAO and KTO with PBE pseudopotential are 3.61 eV and 2.84 eV which are comparable with the experimental band gaps 5.6 eV and 3.5 eV respectively [43]. Unlike LaAlO₃/SrTiO₃ heterostructure, there is no critical thickness of epitaxial layers for insulator-to-metal transition in LaAlO₃/KTaO₃ heterostructure [30].

3. EFFECTIVE HAMILTONIAN

The Rashba effect is a momentum-dependent spin-splitting of an energy band resulting from the combined effect of intrinsic spin-orbit interaction and broken inversion symmetry. Both the heterostructures considered in this work have C₄ᵥ point group symmetry. The little group has three high symmetry points when it is considered for 2D materials, which are Γ (0,0,0), X (0.5,0,0), M(0.5,0.5,0). In these systems, the splitted bands can be described by an effective two band Hamiltonian [49] given by,

\[
H_{c₄ᵥ} = \begin{pmatrix}
\epsilon_1 & \alpha k_x^2 - \alpha R_1 k_x - \alpha R_2 k_y^3 \\
\alpha k_x^2 - \alpha R_1 k_x - \alpha R_2 k_y^3 & \epsilon_2 - \alpha k_x^2 + \alpha R_1 k_x + \alpha R_2 k_y^3
\end{pmatrix}
\]  

where \(\epsilon_1\) and \(\epsilon_2\) are the energy eigenvalues for the first and second band, respectively. \(\alpha R_1, \alpha R_2\) are the coefficient of cubic RSO interaction. To estimate the strength of the linear and cubic RSO interactions in our systems, we consider the Γ – X high symmetry path. Imposing this condition we solve Eq. [1] to obtain the following two eigenvalue equations given by,

\[
\epsilon_1 = \alpha k_x^2 + \alpha R_1 k_x + \alpha R_2 k_y^3\]

The amount of momentum dependent spin-splitting \(\Delta_R\) is given by,

\[
\Delta_R = \epsilon_1 - \epsilon_2 = 2\alpha R_1 k_x + 2\alpha R_3 k_x^3
\]

4. RESULTS

In this section we discuss the aforementioned two types of interface made of LaAlO₃ and KTaO₃. First we present the results of TaO₂⁺/LaO⁺ (type-I) interface which will be followed by KO⁻/AlO₂⁻ (type-II). The role of SOC and the origin of Rashba interactions are presented along with each of the interface.

4.1. Type-I heterostructure without SOC

The TaO₂⁺/LaO⁺ interface is made by two polar end of parent materials which helps the accumulation of electron-like carriers at the interface, thereby giving rise to a 2DEG. Fig. 2(a) presents the band structure in the absence of SOC along M – Γ – X – M high symmetry path. The band structure clearly shows that close to the Fermi energy the atomic contributions are mainly due to the Ta-5d orbitals. However, above the Fermi energy at about 1 eV or more, O-2p orbitals also contribute, albeit tiny, towards the conduction bands. However O-2p orbitals contribute significantly to form the valence bands. The zoomed-in view of the orbital contributions has been presented in Fig. 2(b).

Due to the crystal field splitting (CFS) the degeneracy of the t₂g orbitals of Ta-5d are lifted into a dₓz/dᵧz doublet. At the interface the dₓz band is lower in energy than the dₓz or dᵧz band with a parabolic structure centered around the Γ point. Each dₓz band originates from individual sublayer (marked by open green circle in Fig. 2(b)), whereas all the sublayers contribute to form each of the dₓz/dᵧz bands (marked by solid magenta circle in Fig. 2(b)). O-2p orbital hybridizes with the dₓz/dᵧz orbitals. The angular momentum of dₓz and dᵧz are same with opposite orientation hence the occupation of dₓz and dᵧz orbitals are equal in weightage. The layer-resolved density of states (LRDOS) without spin-orbit interaction are presented in Fig. 2(c), which confirms that the Ta-5d orbitals are predominantly present at the interface that gives rise to the 2DEG.

A visible inter orbital crossing takes place between dₓz and dᵧz in the Γ – X path. The inter-orbital crossing between dₓz and dᵧz bands gives the usual multiorbital (M-O) effect, which has been reported earlier at the interface of SrTiO₃/LaAlO₃ heterostructure [29]. However, in contrast to this earlier reported results, in
Figure 2. (a) Band structure of the Type-I heterostructure. Gray bands represent the states originating from fixed substrate layers, (b) Zoomed-in display of the subbands coming from Ta-5d orbitals, and (c) Layer-resolved density of states for the slab system. Fixed substrate layers are not shown in the picture.

4.2. Type-I heterostructure with SOC

Fig. 3(a) shows the band structure along M−Γ−X−M of the Type-I interface of LAO/KTO heterostructure in the presence of SOC. The spin polarizations are shown (solid red and blue circles) for the s_y component of the spin angular momentum. In Appendix A, the spin polarization of s_x and s_y states are shown in Fig. 3(a) and (b). The spin-splitted Ta-5d orbitals are shown in the left panel. According to the earlier report, the amount of RSO splitting is dependent on the M-O effect present in the system [26]. In Ref. [26], it is found that larger the M-O effect smaller is the RSO splitting. To study the RSO effect, we have confined our study only along the Γ−X path. In the presence of SOC, all the Ta-5d bands break into the Γ_7 and Γ_6 levels. Γ_7 levels have orbital character of d_xy. In Fig. 3(a), we have presented all the Γ_7 levels and two Γ_6 levels. Γ_7 level originates from the d_xy subband of SL-I, and it has no spin-splitting. Systematic study of every Γ_7 level reveals that the spin-splitting gets stronger as we move away from the interface. Γ_7 level has almost no splitting near k=0, whereas for larger k value (around 0.28 Å) it has a splitting at the band crossing region, and it is predominantly cubic like with a RSO coupling strength \( \alpha_{R3} = 80 \text{ meVA}^3 \). Γ_7 level has a similar nature like Γ_7 level. The RSO interaction is cubic like with \( \alpha_{R3} = 0.98 \text{eVÅ}^3 \) at \( k_x \approx 0.13 \text{ Å} \). In the Γ_7 and Γ_7 levels, for \( k_x \lesssim 0.02 \text{ Å} \) the linear RSO interaction strength is one order magnitude higher than the cubic type RSO with \( \alpha_{R1} = 43 \text{ meVA} \) and \( \alpha_{R1} = 256 \text{ meVÅ} \). Γ_7 level shows a spin flipping across the Γ−X path (shown in the left inset of Fig. 3(a)). However, there is no spin-splitting of the bands, and it shows similar behavior to the Γ_7 level. The Γ_6 level, which is a complicated combination of d_yz/d_xz and O-2p_x subbands, shows a spin-splitting. However, this spin-splitting is not Rashba-like. We have estimated the RSO coupling strengths of each spin-splitted levels by fitting Eq. 3 to our DFT data, and the results are presented in Table I.

To study the nature of the spin-splitting we have plotted spin texture of the corresponding bands in the \( k_x − k_y \) plane at a particular energy-cut (light green...
Table I. The table represents the RSO coupling strength obtained by fitting Eqn. with calculated DFT band dispersion data along $\Gamma - X$. Corresponding plots are shown in supplemental material.

| Orbital       | $\alpha_R^1$ (e\AA) | $\alpha_R^3$ (e\AA$^3$) |
|---------------|---------------------|--------------------------|
| $d_{xy}(\Gamma_7)$ | $-7.5$              | $-5.0$                   |
| $d_{xy}(\Gamma_7)$ | $0.8 \times 10^{-3}$ | $0.8$                    |
| $d_{xy}(\Gamma_7)$ | $2.43 \times 10^{-3}$ | $0.98$                   |
| $d_{xy}(\Gamma_7)$ | $-7.5$              | $-5.0$                   |
| $d_{xy}(\Gamma_7)$ | $0.043$             | $39.145$                 |
| $d_{xy}(\Gamma_7)$ | $0.256$             | $113.42$                 |

arrows present the inner band spin orientations and brown arrows present the outer band spin orientations.) Fig. (a) shows the spin texture of $\Gamma_7$ band at iso-energy $E = E_f + 0.19$ eV. It is evident that at lower energy, around $k=0$, there is no spin-splitting of $\Gamma_7$ band. However, as pointed out earlier, at about $E = E_f + 0.96$ eV, there is a lateral shift of up and down spin for inner and outer energy bands (figure not shown). Fig. (b) shows the spin texture of $\Gamma_7$ level at $E = E_f + 1.019$ eV. This spin texture reconfirms the linear RSO interaction in these bands up-to $k_\parallel \approx 0.02$ Å.

![Figure 4](image)

Figure 4. (a) Associated spin-texture of $\Gamma_7$ band in the 2D first Brillouin zone at isoenergy $E - E_f = 0.19$ eV. (b) Spin-texture of $\Gamma_7$, at isoenergy $E - E_f = 1.019$ eV. Brown and green colors distinguish between the two splitted bands with opposite spin orientation. The length of the arrows are in unit of $\hbar$.

### 4.3. Type-II heterostructure without SOC

we have also found Fig. (a) presents the band structure for Type-II system without SOC along M-\Gamma-X-M high symmetry path. O-2p orbitals (marked with solid red circles) are originated from the polar interface created by KO$^-$ and AlO$_2^-$ . Band structure shows that the conduction bands are consisted of Ta-5d subbands (marked by open purple circle) which arise from KTaO$_3$ substrate. The parabolic nature of conduction bands around $\Gamma$ indicates that there is a creation of quantum well, although the conduction band minima does not cross the Fermi level. The gray bands without orbital contributions are shown for the fixed layers of the system which do not have any physical significance. In Fig. (b) we have presented the individual orbital contributions of Ta-5d and O-2p orbitals in the system. It is evident that similar to the Type-I heterostructure, the the degeneracy of the $t_{2g}$ subband of Ta-5d orbital is lifted in an identical manner. Moreover, there is a similar inter-orbital crossing of $d_{xy}$ and $d_{xz}/d_{yz}$ bands in this Type-II hetero-interface. In this oxygen-rich interface CFS also plays a crucial role in O-2p orbital splitting. After orbital separation, the valence bands become an admixture of $p_x + p_z$ orbitals and $p_y + p_z$ orbitals, originated from different epitaxial and substrate sub-layers. LDOS for KO$^-$/AlO$_2^-$ heterostructure reconfirms the formation of O-rich interface, which presented in Fig. (c). Since both the substrate...
and the epitaxial layers at the interface are made of negatively charged surfaces, the occurrence of hole gas is expected. This can be seen from the valence band maxima (VBM) at X (0.5,0,0) (Fig. 11 shown in Appendix B) which is primarily composed of O-2p orbitals. However, this band is a combined effect of contributions from the epitaxial sublayers (EL-I and EL-II) and substrate layer (SL-I). The carrier density in this interface is found to be 3.3 $\times$ 10$^{14}$ cm$^{-2}$. Similar to the type-I system, the last layer of this heterostructure has also been kept fixed, and the density of states (DOS) of the fixed layer has not been shown here. The states of the conduction subbands do not have much contribution at the heterojunction.

**4.4. Type-II heterostructure with SOC**

![Figure 6](image1)

Figure 6. SOC included band structure along M – Γ – X – M contributed by Ta-5d and O-2p orbitals. Red and blue colored circles represent up and down spin polarization along $s_y$. The circle size represent the weight of the spin.

![Figure 7](image2)

Figure 7. (a) Spin-splitted Ta-d$_{xy}$/d$_{xz}$ band along $-X - \Gamma - X$ symmetry. $\Delta E_R$ indicates the maximum energy difference between HS and the extremum of the parabola at $k_x$, where the maximum linear RSO splitting has been found. (b) Associated spin-textures of $\Gamma_6$ band in the $k_x - k_y$ plane at iso-energy $E - E_F = 0.343$ eV. Brown and green colors distinguish between the outer and inner bands with opposite spin orientation.

In this subsection, we have studied Type-II heterostructure in the presence of SOC, and the corresponding band structure is presented in the Fig. 9. The presence of SOC in Type-II system leads to the formation of the $\Gamma_6$ and $\Gamma_7$ levels. The $\Gamma_6$ conduction bands consist of the Ta-5d orbitals, while the valence band consist of O-2p orbitals only. Distinct $\Gamma_6$ levels are indicated in the band structure by the numerals 1, 2, 3 etc. Unlike Type-I system, here all the $\Gamma_6$ levels show RSO spin splitting, whereas $\Gamma_7$ levels has no splitting. This is an oxygen-rich heterojunction, as was previously mentioned, and O-2p orbitals exhibit RSO-splitting in the valence band region. The zoomed in view of the $\Gamma_6$ band, which is a combination of the d$_{yz}$/d$_{xz}$ orbitals, is presented in Fig. 7(a) to demonstrate the k-dependent spin splitting ($\Delta E_R$) (k path from -0.14X to 0.14X). In Fig. 7 we have presented the spin texture corresponding to the $\Gamma_6$ band, and the texture clearly shows a modified linear RSO splitting at iso-energy $E - E_F + 0.343$ eV. The inner band maintains a constant spin projection in the $k_x - k_y$ plane, while in the outer band the amplitude of the spin projection periodically changes with momentum. In order to estimate the RSO coupling strength for Type-II system, the DFT band dispersion data is fitted with Eqn. 4. In Table II the results for all the $\Gamma_6$ levels are presented. For both the $\Gamma_6$ and $\Gamma_6^*$ bands, linear RSO coupling strength is very strong with $\alpha_{R1} \approx 780$ meVÅ and $\alpha_{R1} \approx 800$ meVÅ respectively. In the conduction band region, up to within $k_x \approx 0.05$ Å, $\Gamma_6$ and $\Gamma_6^*$ level shows linear RSO splitting with coupling strength 200 meVÅ and 260 meVÅ respectively. The Type-II system is found to be an ideal system to explore CPGE because it is a member of the $C_{4v}$ point group and possesses a large k-linear RSO splitting with electron spin polarization along $k_x$. Fig. 8 is a schematic representation of the possible electronic arrangement for producing circularly polarized photocurrent in the system which may provide a likely explanation for the recently discovered experimental evidence of CPGE in the LAO/KTO heterostructure [50]. For the right-handed polarized light ($\sigma_+$), the interband transition is possible only for $|j, m_j|=|\frac{1}{2}, \frac{1}{2} \rangle \rightarrow |\frac{1}{2}, \frac{1}{2} \rangle$ and similarly for left handed polarized light ($\sigma_-$) the only possible transition is $|j, m_j|=|\frac{1}{2}, -\frac{1}{2} \rangle \rightarrow |\frac{1}{2}, -\frac{1}{2} \rangle$. In the Type-II system, we have shown schematically the transition takes place between $\Gamma_6$ and $\Gamma_6^*$ levels.

| Band level | $\alpha_{R1}$ (eVÅ) | $\alpha_{R5}$ (eVÅ$^2$) |
|------------|-----------------|-----------------|
| $\Gamma_6$ | 0.78            | -15.3           |
| $\Gamma_6^*$ | 0.8             | -19.4           |
| $\Gamma_6^*$ | 0.2             | -3.24           |
| $\Gamma_6^*$ | 0.26            | -85.41          |

Table II. The table represents the RSO coupling strength for Type-II system obtained by fitting Eqn. 4 with calculated DFT band dispersion data. Corresponding plots are shown in supplemental material [13].
system with the spin polarization along x and z direction. Along z direction there is no spin polarization present in the band dispersion. Along $s_x$, there is spin polarization, although we have not studied this system in detail.

### Appendix B: Spin polarized band-structures of Type-II

![Band structure of Type-II system](image)

Figure 10. (a) and (b) show SOC included band structures of the Type-II heterostructure with the projection of electron spins to $s_x$ and $s_z$, respectively. Red and blue dots denote positive and negative components of spin projections, respectively. The size of the dot shows the relative amplitude of the corresponding spin component.

![Band structure of Type-II system](image)

Fig. 10(a) Fig. 10(b) show the Type-II system’s band structure with spin polarization in the x and z directions. The band dispersion exhibits no spin polarization in the direction of z. Spin polarization exists along $s_x$, however we have not thoroughly examined this system. At the

### Appendix A: Spin polarized band-structures of Type-I

![Band structure of Type-I system](image)

Figure 9. (a) and (b) show SOC included band structures of the Type-I heterostructure with the projection of electron spins to $s_x$ and $s_z$, respectively. Red and blue dots denote up and down components of spin projections, respectively. The size of the dot shows the relative amplitude of the corresponding spin component.

![Band structure of Type-I system](image)

Fig. 9(a) Fig. 9(b) present the band structure of Type-I heterostructure along M-X-M with the projection of electron spins to $s_x$ and $s_z$, respectively. Red and blue arrows show the possible transitions.

### 5. CONCLUSION

In conclusion, TaO$_2$/LaO$_2$ and KO$_2$/AlO$_2$, both of which are referred to as Type-I and Type-II stoichiometric interfaces, has been individually explored in this work. We have investigated the band dispersion characteristics along the $\Gamma-X$ path for both the systems without and with SOC. We have found that, Ta-5d$_{xy}$ bands can produce the strongest k-linear RSO for Type-I systems with a strength up to 256 meVÅ, while the k$^3$-RSO coupling can attain a value up-to 113.42 eVÅ$^3$. In this Type-I heterostructure, no RSO splitting is observed in Ta-5d$_{xy}$/d$_{xz}$ bands. However, Type-II systems predominantly produce k-linear RSO in both the Ta-5d and O-2p orbitals. The maximum RSO coupling strength for this system is estimated to be 800 meVÅ. Interestingly, we have also found that the RSO spin-splitting in Type-II system gives rise to the possibility of circularly polarized photogalvanic effect.

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SUPPLEMENTAL MATERIALS

1. Fitting for RSO parameter in Type-I heterostructure

Figure 12. Hamiltonian fitting by using eqn. 4 for (a) $\Gamma_2$ (b) $\Gamma_3$ (c) $\Gamma_5$ (d) $\Gamma_6$ bands

2. Fitting for RSO parameter in Type-II heterostructure
Figure 13. Hamiltonian fitting by using eqn. 4 for (a) \( \Gamma_{61} \) (b) \( \Gamma_{62} \) (c) \( \Gamma_{63} \) (d) \( \Gamma_{64} \) bands