Electric Field Control of Anisotropic Rashba Splitting in Janus Chromium Dichalcogenide Monolayers: A Computational Study based on Density Functional Theory

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Abstract. *The electronic structure of Janus Chromium Dichalcogenide monolayers CrXY (X ≠ Y = S, Se, Te) in the first Brillouin zone was studied using Density Functional Theory. Due to the mirror symmetry break and strong spin-orbit coupling (SOC) in the crystal structure of Janus Chromium Dichalcogenide monolayers, we discover anisotropic Rashba splitting at around Γ point in the first Brillouin zone. We analyse this anisotropic Rashba splitting using linear Rashba model analysis. By giving the effect of the external electric field, we manipulate the characteristics of Rashba splitting on the Janus Chromium Dichalcogenide monolayer. The manipulation of Rashba splitting by applying an external electric field shows that the Janus Chromium Dichalcogenide monolayers have the potential for spintronic devices.*

Keywords: *Density Functional Theory, Janus Chromium Dichalcogenide, Rashba Splitting*

INTRODUCTION

Recently, the research focused on low-dimensional materials for spin-field effect transistor (SFET) devices. It experienced a notable shift with the groundbreaking isolation of graphene in 2004, offering various advantages [1]. Serving as a pioneering wonder material, the interest of researchers has extended to explore other types of low-dimensional materials. One of the most promising materials for potential discoveries in this context is transition metal dichalcogenides (TMDCs). Members of the two-dimensional (2D) transition metal dichalcogenides (TMDs) family, including MoS_2 [2], $MoSe_2$ [3], and WSe_2 [4], have significant potential in nanoelectronics, nanophotonics, and photocatalysis.

In contrast to the theoretical and experimental attention given to tungsten-based and molybdenum-based dichalcogenides, chromium-based dichalcogenides have received limited study. Following the successful synthesis of the Janus MoSSe monolayer by Lu et al. [5], Janus transition metal dichalcogenide (TMD) monolayers exhibit various novel properties due to the broken centrosymmetry and significant spin-orbit coupling (SOC) [6]. These include substantial out-of-plane piezoelectricity for energy harvesting and

significant Rashba spin splitting (RSS) for semiconductor spintronics applications [7].

Furthermore, Janus Chromium Dichalcogenides have been theoretically demonstrated to be promising materials for photocatalytic applications [8]. Moreover, Janus Cr*XY* (*X* = *Y* = S, Se, Te) monolayers demonstrate intrinsically excellent Rashba effects and piezoelectric responses attributed to inversion asymmetry and robust spin-orbit coupling (SOC) effects [9]. The discovery of both properties adds to the usefulness of Janus transition metal dichalcogenides (TMDs).

In this paper, we primarily utilize first-principles calculations to examine the band structure and Rashba spin splitting of the Janus Cr*XY* (*X* ≠ *Y* = S, Se, Te) monolayers. We also analyse how these properties are modulated under an external electric field. Our findings indicate that the intrinsic Janus CrXY monolayers demonstrate Rashba spin splitting at Γ points in the first Brillouin zone. Moreover, the application of an external electric field allows flexible tuning of the band structures, with significant enhancements observed in the Rashba band splitting effect for spintronics application.

COMPUTATIONAL METHOD

In this work, the Density Functional Theory (DFT) calculations were performed using the Generalized Gradient Approximation [10], implemented in OpenMX code [11]. We utilized norm-conserving pseudopotential [12] with an energy cutoff 250 Ry for charge density. The Spin orbit coupling (SOC) was included in our calculation by j-dependent pseudopotentials [13].

The Janus Chromium Dichalcogenide Cr*XY* (*X* ≠ *Y* = S, Se, Te) monolayers system are the primary materials used in the computational calculation, which consists of CrSSe, CrSTe, and CrSeTe. We construct the crystal structure of Janus Chromium Dichalcogenide by a hexagonal structure. The geometry of the Janus Cr*XY* monolayers was shown in Fig. 1(a) and 1(b) for top and side view, respectively. Here, the structure of Cr*XY* monolayers consists of one transition metal atom Cr which bonds two different chalcogen atoms. Consequently, this structure has a polar system indicated by different distance between Cr-*X* and Cr-*Y*. This condition leads the system to have a non-centrosymmetric structure with the C_{3v} point group.

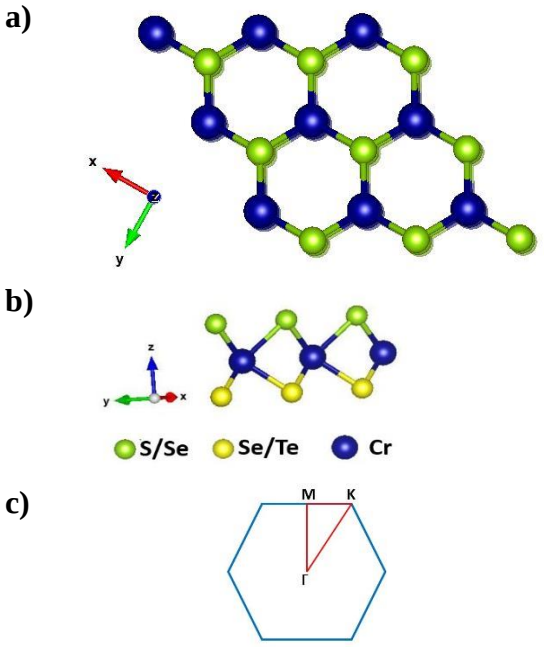

Figure 1. (a) Top and (b) side view of Janus CrXY monolayer which is categorized as a hexagonal structure. (c) The Brillouin zone with path M- K-Q-Γ.

In our calculations, the two-dimensional structure of Cr*XY* monolayers was modelled as a periodic slab with a sufficiently large vacuum layer of 24 Å to eliminate interaction between adjacent layers. In this case, 8×8×1 k-point mesh was applied and the geometries of atomic position were fully

relaxed with force criterion of 1×10^{-5} Hartree/Bohr. The first Brillouin zone of this material system is characterized by several high symmetry points (M, K, Q, Γ) as shown in Fig. 1(c). Then we consider the external electric field with range from 1 to 5 GV/m to know the changes of band structure, especially Rashba splitting due to this effect. However, the electronic structures are calculated after all atoms are fully relaxed for each strains.

RESULTS AND DISCUSSION

We firstly examined the stability of the Cr*XY* monolayers structure before optimizing structural parameters. We found that the optimized lattice constants (a_0) were 3.126 Å (CrSSe ML), 3.300 Å (CrSTe ML), and 3.360 Å (CrSeTe ML). These values have a good agreement with previous research [9].

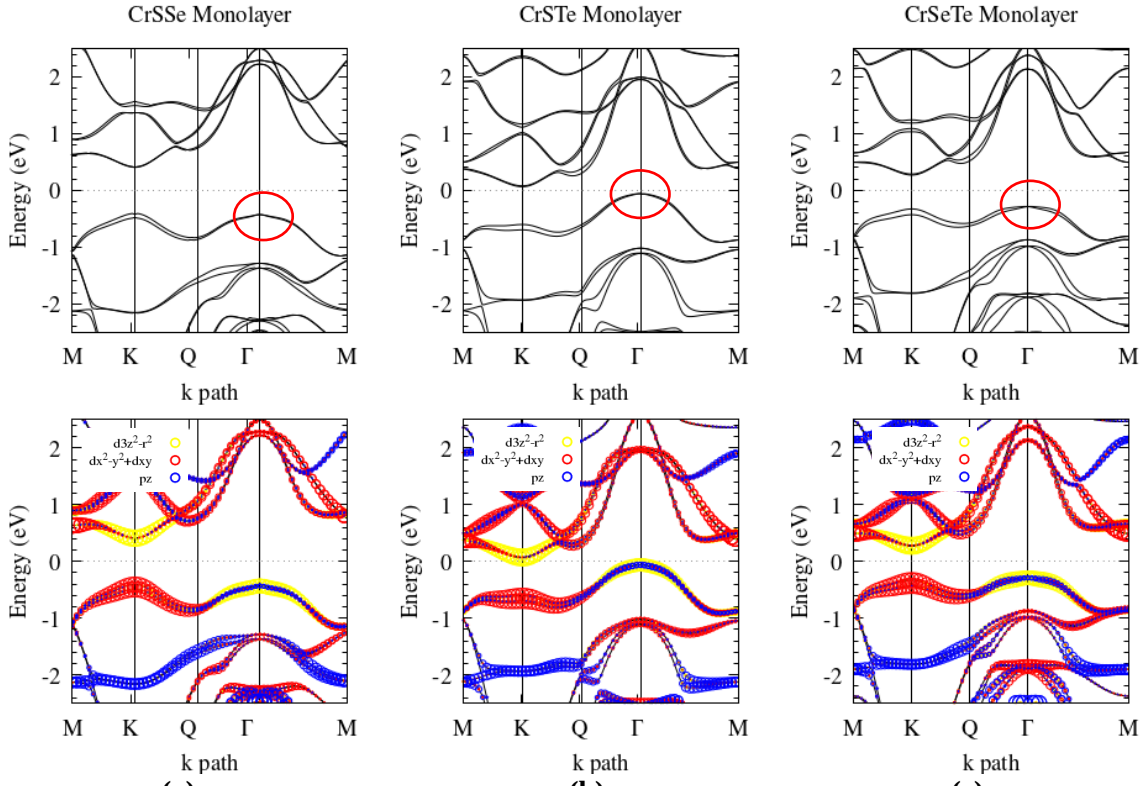

Figure 2. Band Structure (Top) and Unfolding (Bottom) of a) CrSSe, b) CrSTe, and c) CrSeTe Monolayers

Next, the calculated electronic structures of CrSSe, CrSTe, and CrSeTe monolayers are shown in Fig. 2. As shown in Fig. 2 (bottom), valence band maximum (VBM) at Γ point is dominated by the coupling between in-plane atomic orbital contribution $d_{3z^2-r^2}$. By including SOC in the electronic structure calculation, the Rashba splitting occurs around Γ point in VBM for all systems CrSSe ML, CrSTe ML, and CrSeTe ML, due to the time-reversal symmetry breaking (Fig 2a-2c).

Based on the appearance of the Rashba Splitting in all systems, we then focus on characterizing the strength of the splitting using linear Rashba model. The energy dispersion for the Rashba splitting bands is expressed by equation (1) and (2) as follows:

$$E(k) = \frac{\hbar^2}{2m^*} \pm \alpha_R k \quad \ldots\ldots\ldots (1)$$

$$E(k) = \frac{\hbar^2}{2m^*}(|k| \pm k_R)^2 - E_R \quad \ldots\ldots (2)$$

To obtain the Rashba parameter (α_R) we use the relationship expressed in equation (3)

$$\alpha_R = \frac{2E_R}{k_R} \quad \ldots\ldots\ldots\ldots (3)$$

where E_R and k_R are the Rashba energy and the momentum offset, respectively. Based on our calculations we obtain the values of Rashba parameters for CrXY monolayers showed in Table 1.

Table 1. Rashba splitting of CrXY Monolayer around $\Gamma - M$ and $\Gamma\ Q$

System	Rashba Parameter	
	$\alpha_{R,(\Gamma-M)}$	$\alpha_{R,(\Gamma-Q)}$
CrSSe	0.352 eVÅ	0.249 eVÅ
CrSTe	0.019 eVÅ	0.039 eVÅ
CrSeTe	0.568 eVÅ	0.502 eVÅ

The highest Rashba parameter occurs on CrSeTe ML with value 0.568 eVÅ (Γ – M direction) and 0.502 eVÅ (Γ – Q direction). These results are completely consistent with the reference [14]. The Rashba splitting clearly appears in the CrSeTe ML system (as shown in Fig. 3) due to the strong SOC from both Se and Te atoms and also broken inversion symmetry in this system. In contrast, for CrSSe ML and CrSTe ML, the couplings were contributed only by Se or Te atom with little contribution from the counterpart S atom. It makes the Rashba Splitting in these systems are relatively weaker than CrSeTe ML.

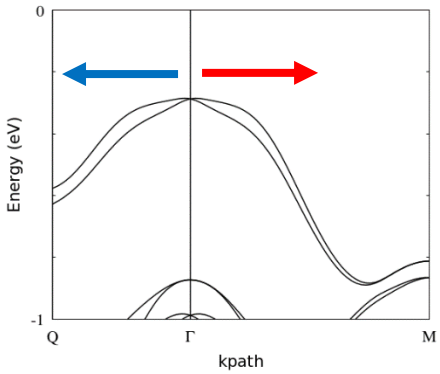

Figure 3. Rashba splitting of CrSeTe ML around $\Gamma - M$ (red) and $\Gamma - Q$ (blue)

Moreover, these findings showed that the Rashba splitting of CrXY Monolayer is typically anisotropic due to different value of Rashba Parameter on Γ – M direction and Γ – Q direction.

We further consider the external electric field effect on the electronic structure of CrSeTe ML with range from 1 to 5 GV/m. Fig. 4 shows the evolution of the band structure of CrSeTe ML respect to the external electric field.

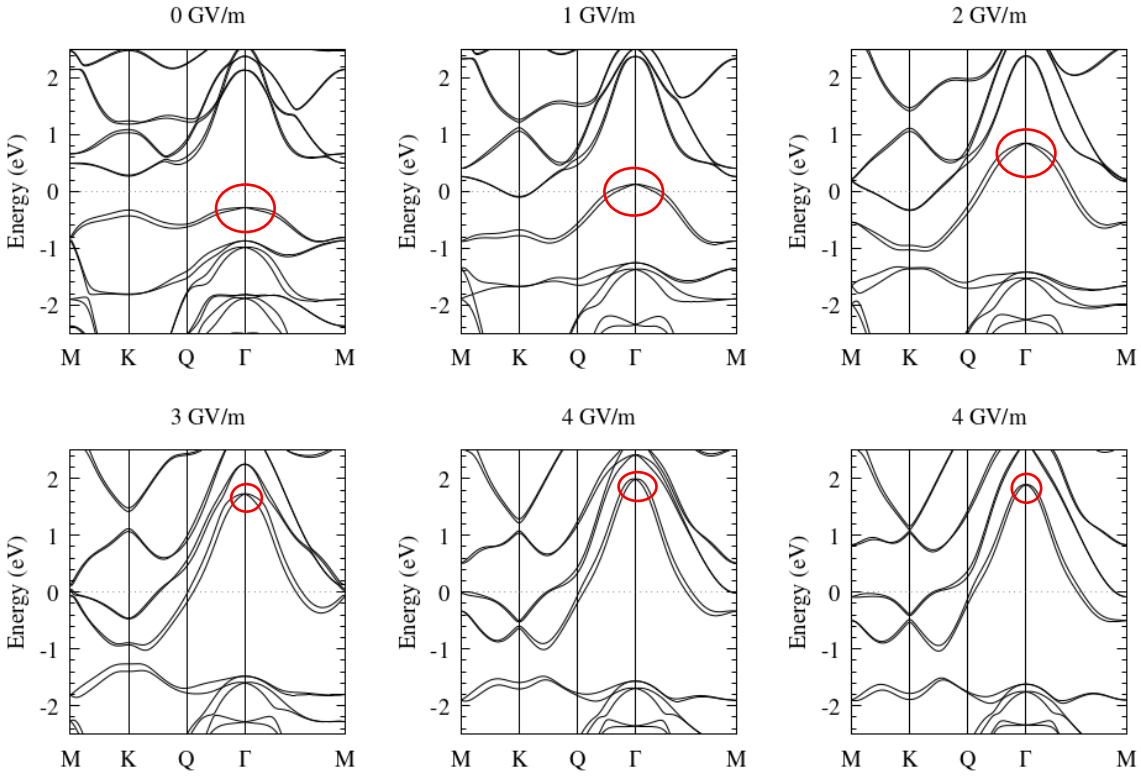

Figure 4. The Electronic Structure of CrSeTe under External Electric Field

The evolution of the electronic structures of the CrSeTe ML by external electric field induces the significant change of the Rashba splitting around Γ point. The Rashba Splitting Parameter of CrSeTe ML under external electric field showed in Table 2.

Table 2. Rashba splitting of CrSeTe Monolayer under External Electric ` Field Effect around $\Gamma - M$ and $\Gamma\ Q$

Electric Field	Rashba Parameter	
	$\alpha_{R,(\Gamma-M)}$	$\alpha_{R,(\Gamma-Q)}$
0 GV/m	0.568 eVÅ	0.502 eVÅ
1 GV/m	0.229 eVÅ	0.555 eVÅ
2 GV/m	0.156 eVÅ	0.625 eVÅ
3 GV/m	0.242 eVÅ	0.424 eVÅ
4 GV/m	0.479 eVÅ	0.697 eVÅ
5 GV/m	0.155 eVÅ	0.368 eVÅ

According to Table 2, the values of Rashba Splitting Parameters of CrSeTe ML are changing both Γ – M direction and Γ – Q direction due to the external electric field. The highest Rashba Splitting Parameter reaches 0.697 eVÅ with 4 GV/m electric field. The Rashba effect in janus CrXY systems generally occurs because of the out-of-plane potential gradient caused by the crystal's non-centrosymmetric nature, meaning it lacks inversion symmetry. This phenomenon doesn't exist in non-Janus systems. By applying external electric field, one can manage the out-of-plane potential gradient. As a result, the Rashba Splitting can be adjusted and regulated using this method.

CONCLUSION

The Density Functional Theory was applied to study the structural and electronic characteristics of Janus CrXY CrXY ($X \neq Y$ = S, Se, Te) monolayer in the first Brillouin zone. Our results reveal the emergence of Rashba splitting at the Γ points in the first Brillouin zone of the Janus Chromium dichalcogenides system. The highest

Rashba Splitting Parameter was found in CrSeTe monolayer. Band structure and Rashba Splitting of CrSeTe monolayer are clearly affected by external electric field applied. The highest Rashba Splitting Parameter reach 0.697 eVÅ under applying 4 GV/m. It suggests that the Janus Chromium Dichalcogenide system could serve as a promising semiconductor material for spintronic applications.

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