

*Supplemental Material*

**Rashba effect modulation in two-dimensional  $A_2B_2Te_6$  (A = Sb, Bi; B = Si, Ge) materials via charge transfer**

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## Structural parameters of original and Janus monolayers

Table SI shows the optimized lattice parameters of original and Janus structures, the lattice parameters of the T-Janus  $A_2B_2X_3Y_3$  monolayers range from 6.59 to 7.15 Å, where the  $Bi_2Ge_2Se_3Te_3$  monolayer having the maximum lattice parameter of 7.15 Å; The lattice parameters of the I-Janus  $A_2BCX_6$  monolayers range from 6.49 to 7.36 Å, where the  $Bi_2SiGeTe_6$  monolayer having the maximum lattice parameter of 7.36 Å; The lattice parameters of the C-Janus  $A_2BCX_3Y_3$  monolayers range from 6.63 to 7.13 Å, where the  $Bi_2SiGeSe_3Te_3$  monolayer having the maximum lattice parameter of 7.13 Å. The variation of lattice parameters can be attributed to the size of atomic radius, therefore the structure with the largest atomic radius in each class of Janus has the largest lattice parameter.

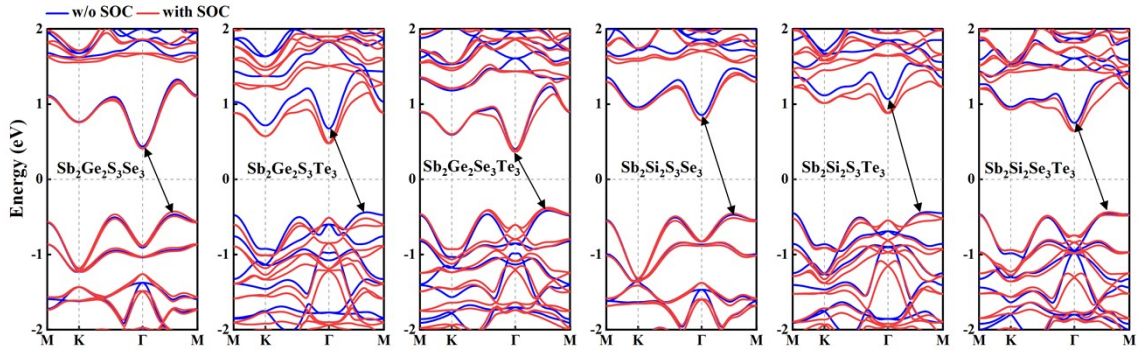
TABLE SI. For original and Janus monolayers: lattice constant ( $a = b$ ), band gap without and with SOC ( $E_{g-PBE}$  and  $E_{g-PBE+SOC}$ ), cohesive energy ( $E_{coh}$ ), position and constant  $\alpha_R$  of Rashba effect ( $C_1$  and  $V_1$  represent the conduction band and valence band closest to the Fermi level, respectively.), dipole moment ( $P$ ).

Classes	Systems	$a=b$ (Å)	$E_{g-PBE}$ (eV)	$E_{g-PBE+SOC}$ (eV)	$E_{coh}$ (eV/atom)	Position ( $\Gamma$ )	$\alpha_R$ (eVÅ)	$P$ (e×Å)
$A_2B_2Te_6$	$Sb_2Si_2Te_6$	7.23	1.04(i)	0.90	-3.18	/	0	0
	$Sb_2Ge_2Te_6$	7.29	0.67(i)	0.58	-2.99	/	0	0
	$Bi_2Si_2Te_6$	7.33	1.38(i)	0.83	-3.22	/	0	0
$A_2B_2X_3Y_3$	$Sb_2Ge_2S_3Se_3$	6.68	0.90(i)	0.84	-3.36	$C_1/V_1$	0.06/0.41	0.089
	$Sb_2Ge_2S_3Te_3$	6.94	1.12(i)	0.99	-3.22	$C_1$	0.57	0.213
	$Sb_2Ge_2Se_3Te_3$	7.06	0.82(i)	0.75	-3.12	$C_1$	0.74	0.129
	$Sb_2Si_2S_3Se_3$	6.59	1.33(i)	1.23	-3.65	$V_1$	0.34	0.093
	$Sb_2Si_2S_3Te_3$	6.87	1.50(i)	1.34	-3.47	$C_1$	0.49	0.223
	$Sb_2Si_2Se_3Te_3$	6.99	1.20(i)	1.08	-3.34	$C_1$	0.64	0.134
	$Bi_2Ge_2S_3Se_3$	6.78	1.24(i)	0.96	-3.42	$V_1$	0.47	0.089
	$Bi_2Ge_2S_3Te_3$	7.04	1.28(d)	0.73	-3.27	$C_1$	0.15	0.211
	$Bi_2Ge_2Se_3Te_3$	7.15	1.12(i)	0.74	-3.16	$C_1$	0.34	0.127
	$Bi_2Si_2S_3Se_3$	6.70	1.65(i)	1.22	-3.70	$V_1$	0.41	0.091
	$Bi_2Si_2S_3Te_3$	6.97	1.76(d)	1.02	-3.52	$C_1$	0.16	0.218
	$Bi_2Si_2Se_3Te_3$	7.09	1.51(i)	1.02	-3.38	$C_1$	0.27	0.131
$A_2BCX_6$	$Sb_2SiGeS_6$	6.49	1.32(i)	1.27	-3.63	$C_1$	0.07	0.022
	$Sb_2SiGeSe_6$	6.78	0.94(i)	0.88	-3.38	$C_1/V_1$	0.11/0.17	0.019
	$Sb_2SiGeTe_6$	7.26	0.88(i)	0.75	-3.08	$C_1$	0.30	0.013
	$Bi_2SiGeS_6$	6.60	1.63(i)	1.28	-3.69	$C_1$	0.14	0.016
	$Bi_2SiGeSe_6$	6.87	1.28(i)	0.97	-3.43	$C_1/V_1$	0.16/0.30	0.014
	$Bi_2SiGeTe_6$	7.36	1.21(i)	0.73	-3.12	$C_1$	0.23	0.011
<u><math>A_2BCX_3Y_3</math></u>	$Sb_2SiGeSe_3S_3$	6.63	1.15(i)	1.07	-3.49	$C_1/V_1$	0.24/0.25	0.070
	$Sb_2SiGeTe_3S_3$	6.90	1.33(i)	1.19	-3.31	$C_1$	0.70	0.202
	$Sb_2SiGeS_3Se_3$	6.64	1.14(i)	1.06	-3.52	$V_1$	0.51	0.111
	$Sb_2SiGeTe_3Se_3$	7.02	1.03(i)	0.93	-3.21	$C_1$	0.86	0.116
	$Sb_2SiGeS_3Te_3$	6.91	1.33(i)	1.18	-3.37	$C_1$	0.27	0.237
	$Sb_2SiGeSe_3Te_3$	7.03	1.04(i)	0.93	-3.24	$C_1$	0.43	0.148

$\text{Bi}_2\text{SiGeSe}_3\text{S}_3$	6.74	1.49(i)	1.13	-3.54	$C_1/V_1$	0.15/0.28	0.075
$\text{Bi}_2\text{SiGeTe}_3\text{S}_3$	7.00	1.59(d)	0.93	-3.36	$C_1$	0.43	0.202
$\text{Bi}_2\text{SiGeS}_3\text{Se}_3$	6.75	1.46(i)	1.11	-3.58	$C_1/V_1$	0.29/0.58	0.105
$\text{Bi}_2\text{SiGeTe}_3\text{Se}_3$	7.12	1.34(i)	0.93	-3.26	$C_1$	0.58	0.116
$\text{Bi}_2\text{SiGeS}_3\text{Te}_3$	7.01	1.51(d)	0.85	-3.42	/	0	0.227
$\text{Bi}_2\text{SiGeSe}_3\text{Te}_3$	7.13	1.34(i)	0.87	-3.29	/	0	0.142

## Band structures of original and Janus monolayers

The band structures of 3 original monolayers and 30 Janus monolayers using PBE method with and without SOC are shown in Fig. S1. When SOC is taken into account, we find that RSS occurs in most Janus monolayers, varying from 0.06 to 0.86 eVÅ. However, for some monolayers, such as  $\text{Sb}_2\text{Si}_2\text{S}_3\text{Te}_3$ , although the RSS also appears at the CBM, it is too weak to be calculated according to the formula, so we do not consider it. In addition, for  $\text{Sb}_2\text{SiGeSe}_6$ ,  $\text{Bi}_2\text{SiGeSe}_6$ ,  $\text{Sb}_2\text{Ge}_2\text{S}_3\text{Se}_3$ ,  $\text{Sb}_2\text{Si}_2\text{S}_3\text{Se}_3$ ,  $\text{Bi}_2\text{Ge}_2\text{S}_3\text{Se}_3$ ,  $\text{Bi}_2\text{Si}_2\text{S}_3\text{Se}_3$ ,  $\text{Sb}_2\text{SiGeSe}_3\text{S}_3$ ,  $\text{Sb}_2\text{SiGeS}_3\text{Se}_3$ ,  $\text{Bi}_2\text{SiGeSe}_3\text{S}_3$  and  $\text{Bi}_2\text{SiGeS}_3\text{Se}_3$  monolayers, their first valence band ( $V_1$ ) exhibits the Rashba effect at the  $\Gamma$  point, but since it is not at the VBM, the conducting holes or electrons will be disturbed by other states near the Fermi level, so we do not further discuss it.



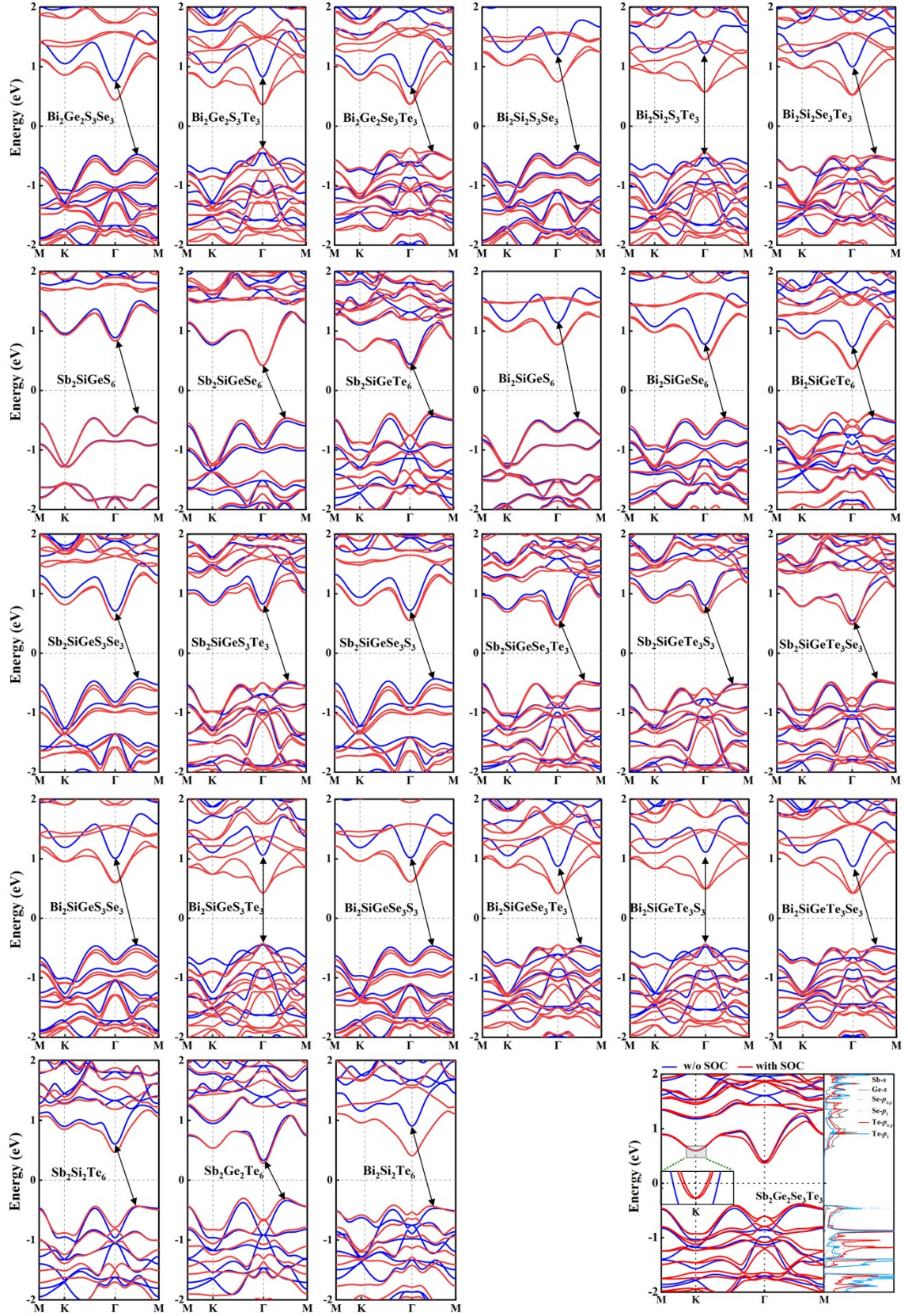


FIG. S1. Band structures of original and Janus monolayers.



## Electric field tuning of the Rashba effect

The electric field ranges from -0.3 to 0.3 VÅ<sup>-1</sup>, which can be achieved in the experiment<sup>1</sup>. The calculated variation of the Rashba constant  $\alpha_R$  with the  $E_{ex}$  is shown in Fig. S2. We find that  $\alpha_R$  is

linearly dependent on the  $E_{ex}$ . Furthermore, the electric field response rate ( $k = \frac{\Delta\alpha_R}{\Delta E}$ ) has been calculated, whose  $|k|$  values are greater than 0.1 eÅ<sup>2</sup>.

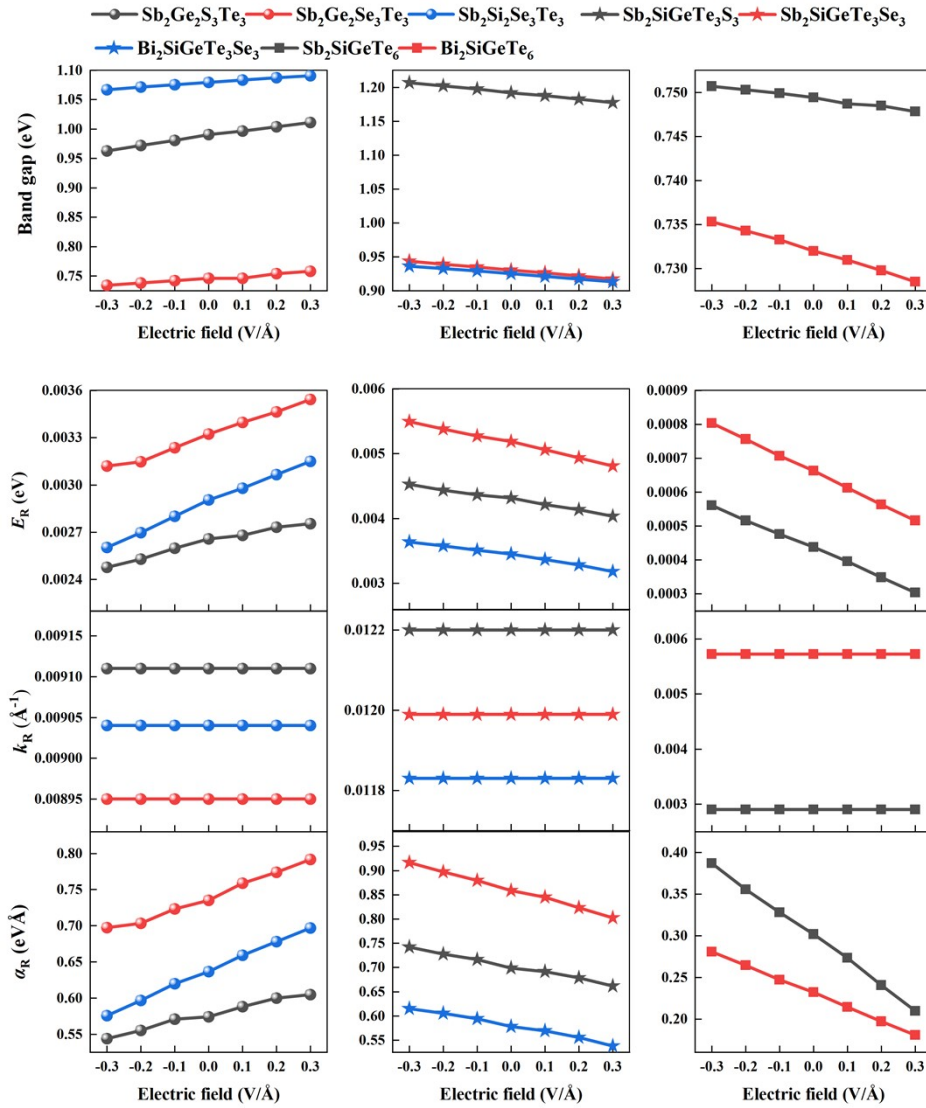


FIG. S2. The band gap, Rashba energy  $E_R$ , momentum offset  $k_R$ , Rashba constant  $\alpha_R$  of T-, I-, and C-Janus monolayers under external electric field ( $E_{ex}$ ).

## Polarization properties of Janus monolayers

Previous studies have shown that the breaking of intrinsic mirror symmetry in Janus 2D materials leads to some fascinating polarization-dependent properties such as different surface work functions, dipole moment, and  $E_{in}^{2,3}$ . Specifically speaking, the electrostatic potential energy peaks of the original systems are symmetric, consistent with the symmetric crystal structures, as listed in Fig. S3(a<sub>1</sub>). However, for the Janus systems with broken structural symmetry, the electrostatic potential distributions are asymmetrical with different peaks in the different positions, which leads to different work functions at X and Y atomic surfaces [Fig. S3(a<sub>2</sub>)]. Moreover, we further investigate the mirror symmetry breaking induced dipole moment to confirm the polarization effect (Table SI). The work function difference  $\Delta\Phi$  between both surfaces is directly affected by the dipole moment  $P$ , which is in agreement with the Helmholtz equation. Considering that the vertical intrinsic electric field  $E_{in}$  is generally the key factor affecting the  $\alpha_R$  of Janus Rashba systems and even its modulation trend under  $E_{ex}$ , which could be directly reflected in aspects of work function and electrostatic potential, dipole moment, or charge transfer, we have carried out analysis on the above physical quantities (Fig. S4-S7).

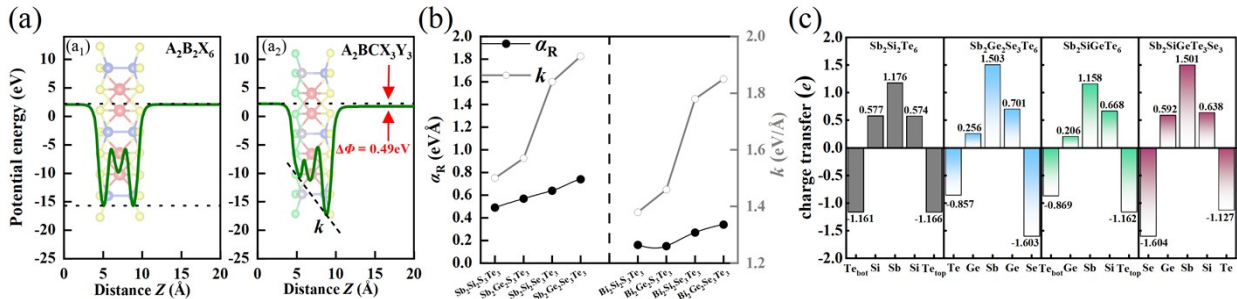
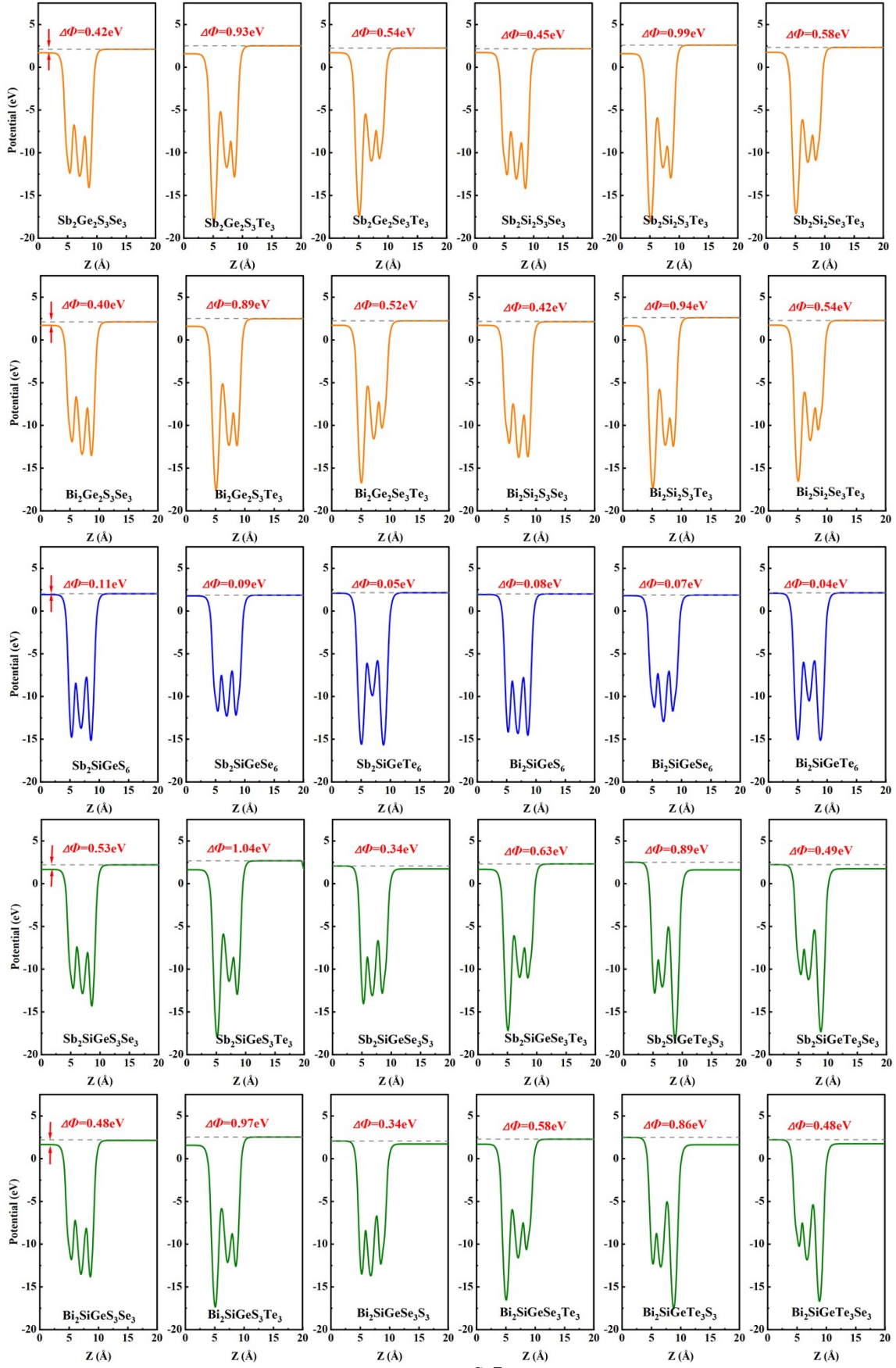


FIG. S3. (a) Planar average of the electrostatic potential of (a<sub>1</sub>) original and (a<sub>2</sub>) Janus monolayers along the  $z$  direction (taking the original  $Sb_2Ge_2Te_6$  and composite Janus  $Sb_2SiGeTe_3Se_3$  monolayer as examples). (b) The electrostatic potential slope  $k$  on both sides and Rashba constant  $\alpha_R$  for the T-Janus  $A_2B_2X_3Y_3$  monolayers. (c) Charge transfer of  $Sb_2Si_2Te_6$ ,  $Sb_2Ge_2Se_3Te_6$ ,  $Sb_2SiGeTe_6$ , and  $Sb_2SiGeTe_3Se_3$  monolayers.



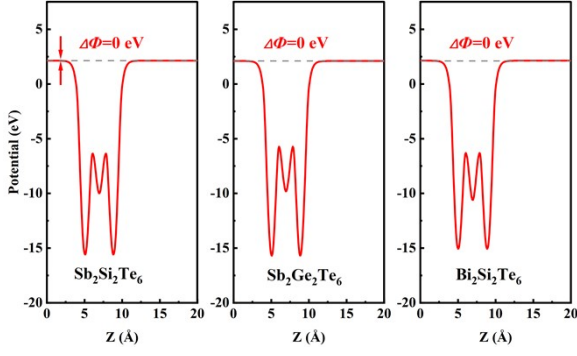


FIG. S4. Planar average of the electrostatic potential of Janus and original monolayers along the  $z$  direction. The yellow, blue, green and red solid lines indicate the configurations of T-, I-, C-Janus monolayers and original monolayers, respectively.

The electric field can be estimated by the “slope  $k$ ” of the potential energy diagram:

$$k = \frac{V(X) - V(Y)}{d(X) - d(Y)}$$

where  $V(X/Y)$  is the vertical planar average potential on X/Y atom,  $d(X/Y)$  is the distance between origin and X/Y atom, as shown in Fig. S5.

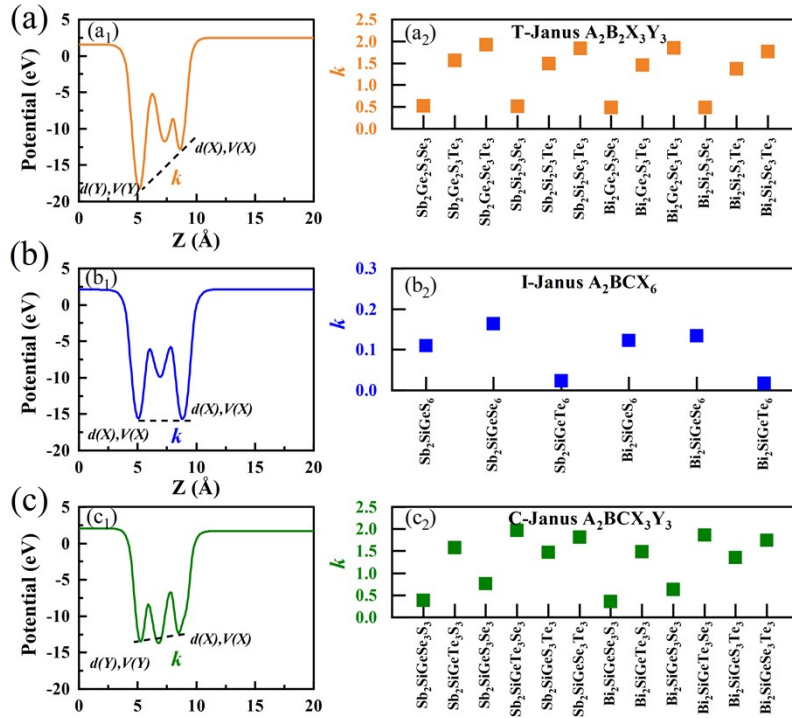


FIG. S5. Slope of potential energy differences for three classes of Janus structures (a) T-Janus  $A_2B_2X_3Y_3$ , (b) I-Janus  $A_2BCX_6$  and (c) C-Janus  $A_2BCX_3Y_3$ .



For T-Janus, due to its characteristic of having only the outer-layer elements differing, T-Janus  $A_2B_2X_3Y_3$  exhibits a distinct work function difference  $\Delta\Phi$  and the slope of potential energy difference  $k$  [Fig. S6(a)]. According to the Helmholtz relation, the work function difference  $\Delta\Phi$  is positively correlated with the dipole moment  $P$ , thus T-Janus possesses a significant dipole moment. However, as observed from the locally magnified potential energy diagrams in Fig. S6(b) and S6(c), there is no significant difference in work function, potential energy between I-Janus  $A_2BCX_6$  and C-Janus  $A_2BCX_3Y_3$ . Similarly, both have smaller dipole moments. In particular, for I-Janus  $A_2BCX_6$ , where the atoms at both ends are identical, the work function and potential energy difference are nearly zero. The method of measuring the  $E_{in}$  through Bader charge transfer analysis is not limited by the type of Janus configuration. In addition, using charge transfer to unify the description can well reflect the changes in local electric fields in subsequent single atom adsorption.

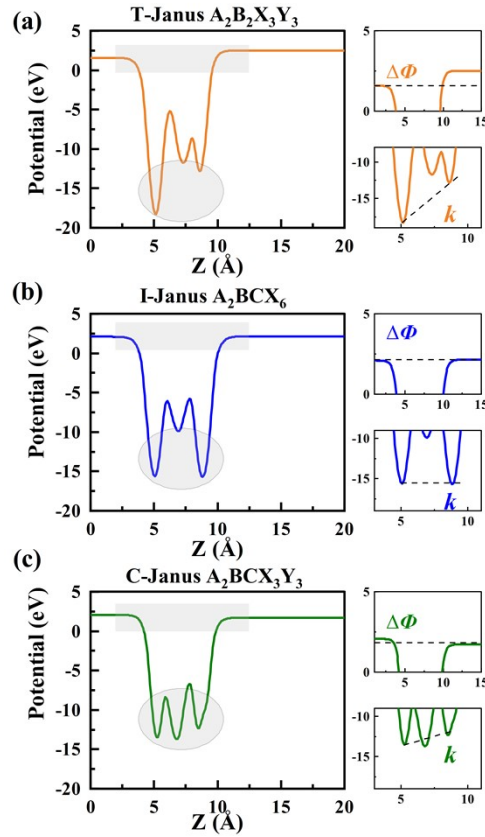


FIG. S6. Images of the local magnified work function difference as well as the slope  $k$  of the potential energy difference for three classes of Janus structures (a) T-Janus  $A_2B_2X_3Y_3$ , (b) I-Janus  $A_2BCX_6$  and (c) C-Janus  $A_2BCX_3Y_3$ .

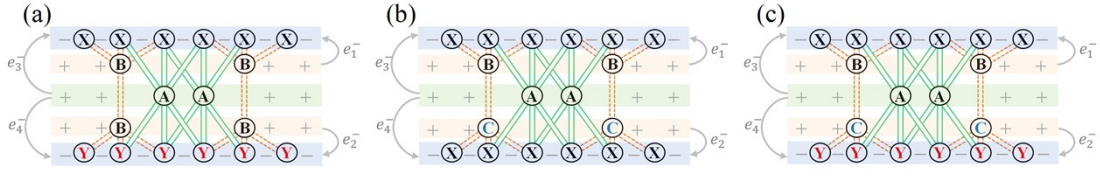


FIG. S7. Schematic illustrations of charge transfer in the (a) T-Janus, (b) I-Janus, and (c) Janus structures.

$$Q_d = |e_1^- + e_3^- - e_2^- - e_4^-|$$

$$Q_t = |e_1^- + e_2^- + e_3^- + e_4^-|$$

	$Q_Y$	$Q_B$	$Q_A$	$Q_B$	$Q_X$	$Q_d$	$Q_t$
$\text{Sb}_2\text{Ge}_2\text{S}_3\text{Se}_3$	1.579	-0.658	-2.069	-0.948	2.096	0.517	3.675
$\text{Sb}_2\text{Ge}_2\text{S}_3\text{Te}_3$	0.841	-0.304	-1.749	-0.907	2.119	1.278	2.96
$\text{Sb}_2\text{Ge}_2\text{Se}_3\text{Te}$	0.857	-0.256	-1.503	-0.701	1.603	0.746	2.46
$\text{Sb}_2\text{Si}_2\text{S}_3\text{Se}_3$	2.265	-1.264	-2.083	-1.855	2.938	0.673	5.203
$\text{Sb}_2\text{Si}_2\text{S}_3\text{Te}_3$	1.163	-0.508	-1.740	-1.865	2.950	1.787	4.113
$\text{Sb}_2\text{Si}_2\text{Se}_3\text{Te}_3$	1.147	-0.585	-1.494	-1.354	2.286	1.139	3.433
$\text{Bi}_2\text{Ge}_2\text{S}_3\text{Se}_3$	1.638	-0.660	-2.175	-0.953	2.149	0.511	3.787
$\text{Bi}_2\text{Ge}_2\text{S}_3\text{Te}_3$	0.945	-0.311	-1.897	-0.916	2.178	1.233	3.123
$\text{Bi}_2\text{Ge}_2\text{Se}_3\text{Te}_3$	0.957	-0.257	-1.671	-0.703	1.674	0.717	2.631
$\text{Bi}_2\text{Si}_2\text{S}_3\text{Se}_3$	2.333	-1.278	-2.187	-1.863	2.994	0.661	5.327
$\text{Bi}_2\text{Si}_2\text{S}_3\text{Te}_3$	1.249	-0.507	-1.886	-1.879	3.023	1.774	4.272
$\text{Bi}_2\text{Si}_2\text{Se}_3\text{Te}_3$	1.256	-0.580	-1.680	-1.362	2.366	1.11	3.622

TABLE SII. The charge transfer of atoms in T-Janus  $A_2B_2X_3Y_3$  monolayer.  $Q_A(e_3^- + e_4^-)$ ,  $Q_B(e_1^- + e_2^-)$ ,  $Q_X(e_1^- + e_3^-)$  and  $Q_Y(e_2^- + e_4^-)$  represent the charge transfer amount of each layer respectively. Negative values represent electron loss of corresponding

	$Q_X$	$Q_C$	$Q_A$	$Q_B$	$Q_X$	$Q_d$	$Q_t$
$\text{Sb}_2\text{SiGeS}_6$	2.101	-0.763	-2.309	-1.998	2.970	0.869	5.071
$\text{Sb}_2\text{SiGeSe}_6$	1.594	-0.502	-1.840	-1.554	2.303	0.709	3.897
$\text{Sb}_2\text{SiGeTe}_6$	0.869	-0.206	-1.158	-0.668	1.162	0.293	2.031
6							
$\text{Bi}_2\text{SiGeS}_6$	2.126	-0.761	-2.387	-1.995	3.016	0.89	5.142
$\text{Bi}_2\text{SiGeSe}_6$	1.646	-0.502	-1.964	-1.549	2.369	0.723	4.015
$\text{Bi}_2\text{SiGeTe}_6$	0.993	-0.212	-1.377	-0.687	1.283	0.29	2.276
	Y	C	A	B	X	$Q_d$	$Q_t$
$\text{Sb}_2\text{SiGeSe}_3\text{S}_3$	2.100	-0.767	-2.086	-1.541	2.293	0.193	4.393
$\text{Sb}_2\text{SiGeTe}_3\text{S}_3$	2.119	-0.859	-1.766	-0.609	1.116	1.003	3.235
$\text{Sb}_2\text{SiGeS}_3\text{Se}_3$	1.578	-0.496	-2.057	-1.999	2.975	1.397	4.553
$\text{Sb}_2\text{SiGeTe}_3\text{Se}$	1.604	-0.592	-1.501	-0.638	1.127	0.477	2.731
3							
$\text{Sb}_2\text{SiGeS}_3\text{Te}_3$	0.855	-0.107	-1.716	-2.003	2.970	2.115	3.825
$\text{Sb}_2\text{SiGeSe}_3\text{Te}$	0.867	-0.186	-1.494	-1.493	2.306	1.439	3.173
3							
$\text{Bi}_2\text{SiGeSe}_3\text{S}_3$	2.146	-0.774	-2.188	-1.551	2.367	0.221	4.513
$\text{Bi}_2\text{SiGeTe}_3\text{S}_3$	2.171	-0.867	-1.907	-0.645	1.248	0.923	3.419
$\text{Bi}_2\text{SiGeS}_3\text{Se}_3$	1.644	-0.497	-2.175	-2.005	3.033	1.389	4.677
$\text{Bi}_2\text{SiGeTe}_3\text{Se}_3$	1.675	-0.596	-1.688	-0.649	1.258	0.417	2.933
$\text{Bi}_2\text{SiGeS}_3\text{Te}_3$	0.953	-0.110	-1.875	-2.021	3.053	2.1	4.006



TABLE SIII. The charge transfer of atoms in I-Janus  $A_2BCX_6$  monolayer.

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TABLE SIV. The charge transfer of atoms in C-Janus  $A_2BCX_3Y_3$  monolayer.

In pristine  $A_2B_2X_6$ , there is no difference in electronegativity between the atoms on both sides of the center, which results in a charge transfer difference  $Q_d$  of 0 and the absence of  $E_{in}$ . For the Janus  $A_2B_2X_3Y_3$ , due to the difference in electronegativity between the upper and lower atomic layers, there will be a non-zero  $Q_d$  and  $E_{in}$ . The schematic diagram of the Janus structures (e.g., T-Janus structure) and the charge transfer model are shown in Fig.S8(a). When applying an  $E_{ex}$ , the charge transfer  $Q_d$  ( $Q_d = |e_1^- + e_3^- - e_2^- - e_4^-|$ ) and  $Q_t$  ( $Q_t = |e_1^- + e_2^- + e_3^- + e_4^-|$ ) combined with its response to the  $E_{ex}$  ( $k_1 = \frac{\Delta Q_d}{\Delta E}$  and  $k_2 = \frac{\Delta Q_t}{\Delta E}$ ), we establish a rough relationship between charge transfer  $Q_{d+t}$  (defined as  $Q_{d+t} = Q_d \times k_1 + Q_t \times k_2$ ) and  $\alpha_R$ . When  $Q_{d+t}$  is greater than 0, the  $\alpha_R$  enhances (weakens) monotonically with the increase of  $E_p$  ( $E_n$ ) strength; When  $Q_{d+t}$  is less than 0, the  $\alpha_R$  weakens (enhances) monotonically with the increase of  $E_p$  ( $E_n$ ) strength.

The Fig. S8(c) and Fig. S8(d) illustrates the linear variation of  $Q_d$  and  $Q_t$  under  $E_{ex}$ , we define

their electric field responses as  $k_1 = \frac{\Delta Q_d}{\Delta E}$  and  $k_2 = \frac{\Delta Q_t}{\Delta E}$ , respectively. Considering that  $Q_d$  can reflect the strength of  $E_{in}$  by generating a potential gradient in the basal plane at both ends of monolayers,  $Q_t$  reflects the strength of the electronegativity difference between atoms by the total amount of charge transfer. To explain the change in  $\alpha_R$  of Janus structures under the action of  $E_{ex}$ , we continue to use the mathematical expression  $Q_d \times k_1 + Q_t \times k_2$  based on  $Q_d$  and  $Q_t$ , which is abbreviated as  $Q_{d+t}$ .

For the charge transfer of T-Janus structures under  $E_{ex}$ , we obtain the relationship between  $\frac{\Delta \alpha_R}{\Delta E}$  and  $Q_{d+t}$  and extend it to I- and C-Janus structures [Fig. S8(e)]. As a result, the numerical values of  $Q_{d+t}$  are 0.34, 0.26 and 0.46 for SGST-1, SGST-2 and SSST monolayers; The C-Janus structures SSGTS-1, SSGTS-2, BSGTS, and I-Janus structures SSGT, BSGT have values of -1.03, -0.86, -0.93, -0.42 and -0.39, respectively (Table SV).

Therefore, when combining the  $Q_d$  with another  $Q_t$  related to element electronegativity, the trend of  $\alpha_R$  increasing or decreasing with the strength of  $E_{ex}$  can be further approximately confirmed based

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$Q_d$	$k_1$	$Q_t$	$k_2$	$Q_{d+t}$
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on the slope of charge transfer change under  $E_{ex}$ .

TABLE SV. The data associated with  $Q_{d+t}$  comprises  $Q_d$ ,  $k_1$ ,  $Q_t$ , and  $k_2$ , where  $k_1$  and  $k_2$  denote the electric field rate of change for  $Q_d$  and  $Q_t$ , respectively.

$\text{Sb}_2\text{Si}_2\text{Se}_3\text{Te}_3$	1.139	0.43	3.433	-0.01	0.46
$\text{Sb}_2\text{SiGeTe}_3\text{S}_3$	1.003	-0.41	3.235	-0.19	-1.03
$\text{Sb}_2\text{SiGeTe}_3\text{Se}$	0.477	-0.44	2.731	-0.23	-0.86
3					
$\text{Bi}_2\text{SiGeTe}_3\text{Se}_3$	0.417	-0.45	2.933	-0.25	-0.93
$\text{Sb}_2\text{SiGeTe}_6$	0.293	0.50	2.031	-0.28	-0.42
$\text{Bi}_2\text{SiGeTe}_6$	0.29	0.52	2.276	-0.24	-0.39



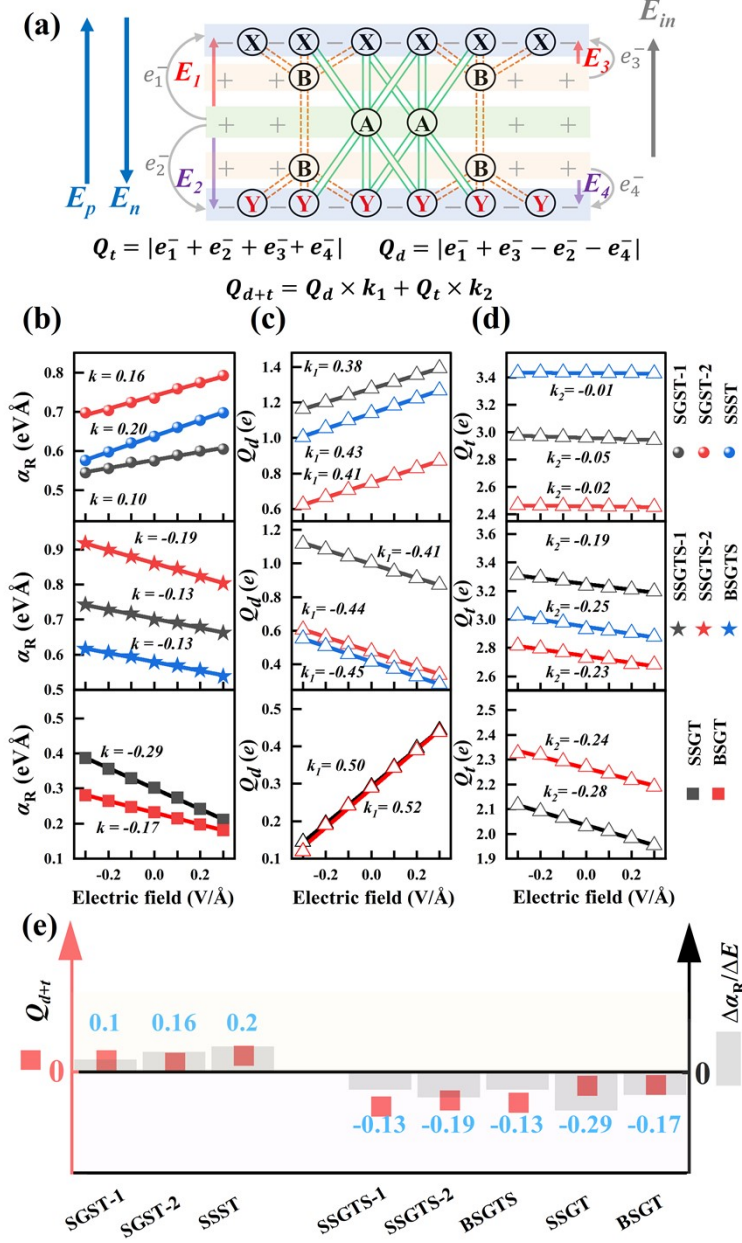


FIG. S8. (a) Schematic diagram of charge transfer of T-Janus  $A_2B_2X_3Y_3$ .  $e_1^-, e_2^-, e_3^-, e_4^-$

represent the flow of electrons between different layers.  $E_1, E_2, E_3, E_4$  represent the localized electric field. The gray and blue arrows represent  $E_{in}$  and  $E_{ex}$ , respectively, where the positive electric field ( $E_p$ ) along the  $z$ -direction, and the negative electric field in the opposite direction ( $E_n$ ). Under an external electric field (b)  $a_R$  response is quantified by parameter  $k$ , (c)  $Q_d$  response is governed by parameter  $k_1$

## Atomic adsorption reconfigures charge transfer

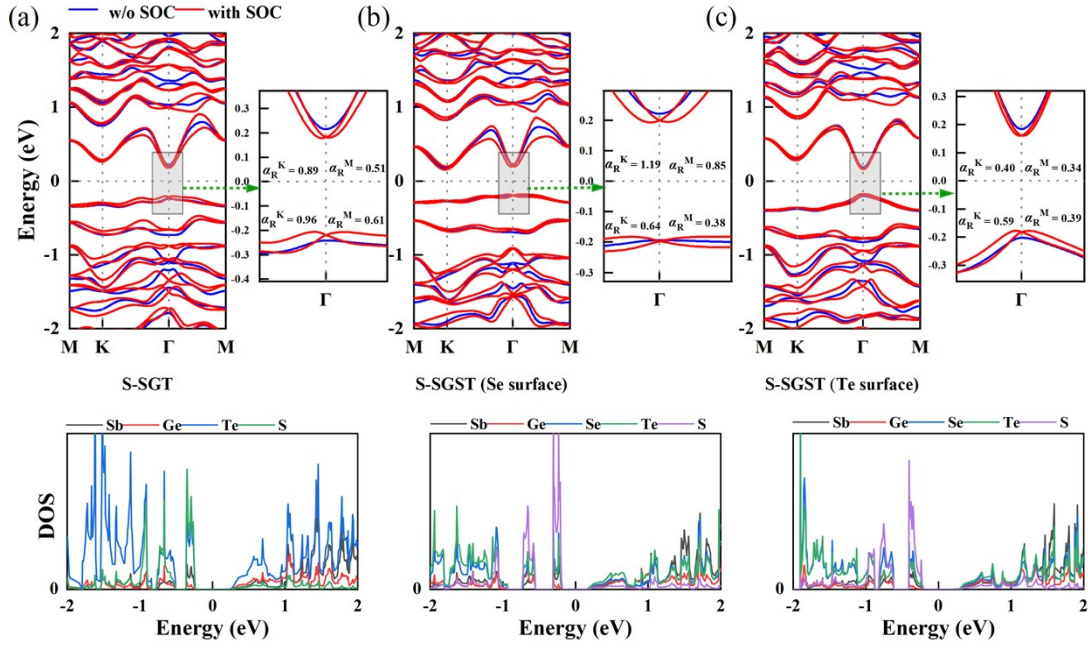


FIG. S9. (a) Band structures and DOS of S adsorbed on SGT. Band structures and DOS of (b) S adsorbed on SGST (Se-surface), and (c) S adsorbed on SGST (Te-surface).

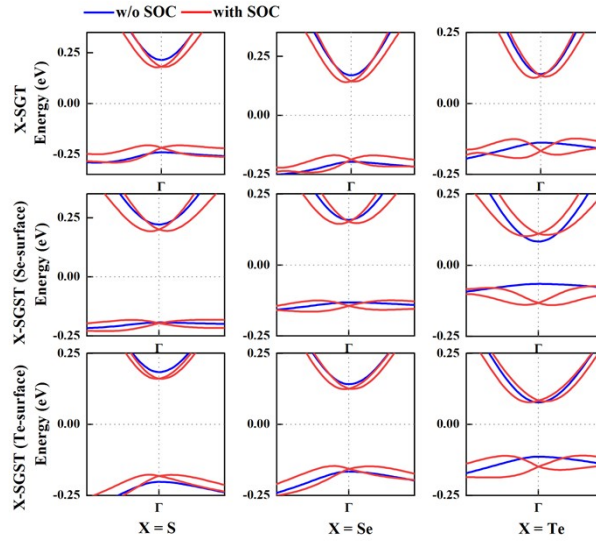


FIG. S10. Band structures of X-SGT and X-SGST systems. Blue and red solid lines indicate methods of PBE and PBE+SOC, respectively.

## BN/SGST and BN/S-SGST heterostructures

In experiments, 2D materials are usually grown on a substrate, and the electronic properties are affected by the substrate. Generally, the insulating substrate materials used for 2D devices are BN, AlN, and GaN. The lattice mismatches for SGST/BN ( $3\times3\times1$ ), SGST/AlN ( $2\times2\times1$ ), and SGST/GaN ( $2\times2\times1$ ) are 6%, 12%, and 9%, respectively. Considering practical feasibility and minimizing lattice mismatch between the two stacks, we construct the heterostructure using a  $3\times3\times1$  supercell of BN and one SGST layer and investigate their electronic properties. Electronic structure calculations show that heterostructures retain the indirect or direct band gap characteristics of their original systems, but the band gap is significantly reduced compared to the original systems. The band gap of the BN/SGST heterostructures decreases to 0.47 eV (Se-surface) and 0.46 eV (Te-surface), and the band gap of the BN/S-SGST heterostructures decreases to 0.05 eV (Se-surface) and 0.03 eV (Te-surface). When SOC is taken into account, the band gap of the BN/SGST heterostructures decreases to 0.42 eV, 0.41 eV, and the band gap of the BN/S-SGST heterostructures decreases to 0.05 eV, 0.05 eV. It is worth noting that for the BN/S-SGST system, significant RSS occurs simultaneously at CBM and VBM, which can be attributed to the presence of Te and Se atomic orbitals at CBM and S atomic orbital at VBM, as shown in the projected DOS diagrams of the two heterostructures [Fig. S11(b)].

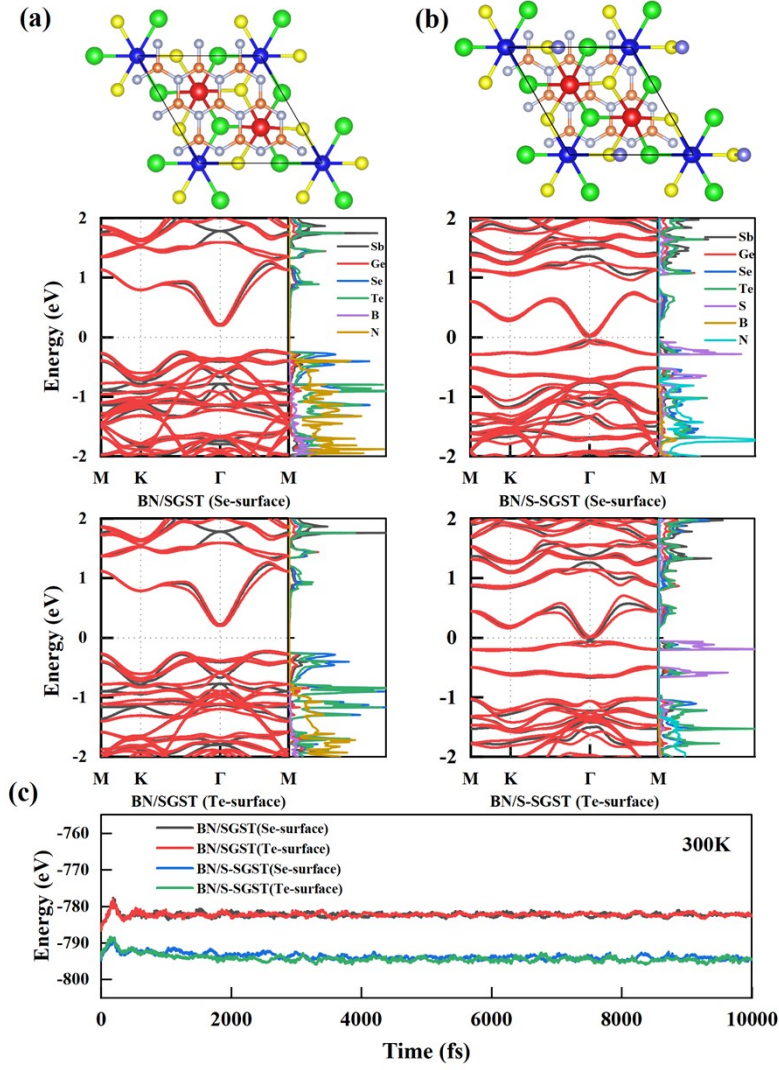


FIG. S11. (a) Band structures and DOS of BN/SGST heterostructures (Se-surface and Te-surface) (b) Band structures and DOS of BN/S-SGST (Se-surface and Te-surface) heterostructures. (c) AIMD simulations of total energy fluctuation under 10 ps at 300K of BN/SGST and BN/S-SGST heterostructures.

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