Tunable Rashba spin splitting in Janus transition-metal dichalcogenide monolayers via charge doping

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I. STRUCTURE CONSTANTS AND BAND GAPS OF SIX JANUS MXY MONOLAYERS

Table S1: Calculated lattice constant ($a = b$), bond length (l_{M-X}) and (l_{M-Y}), distance between the sublayers (d_{M-X}) and (d_{M-Y}), electrostatic potential difference $\Delta\phi$ and band gaps with different functionals (PBE and PBE+SOC) are displayed. The band-gap values are labeled by letter D or I, which represents the direct or indirect band gap, respectively.

Structures	a, b (Å)	l_{M-X} (Å)	l_{M-Y} (Å)	d_{M-X} (Å)	d_{M-Y} (Å)	$\Delta\phi$ (eV)	PBE+vdW (eV)	PBE+vdW+SOC (eV)
MoSSe	3.230	2.417	2.533	1.538	1.713	0.351	1.63 (D)	1.53 (D)
MoSTe	3.344	2.435	2.714	1.483	1.907	0.703	1.16 (I)	1.12 (I)
MoSeTe	3.411	2.553	2.715	1.625	1.869	0.353	1.33 (D)	1.20 (D)
WSSe	3.231	2.422	2.539	1.545	1.722	0.337	1.77 (D)	1.44 (I)
WSTe	3.344	2.438	2.720	1.490	1.916	0.673	1.34 (I)	1.21 (I)
WSeTe	3.412	2.559	2.722	1.634	1.878	0.338	1.42 (D)	1.10 (I)

II. TUNABLE RASHBA SPIN SPLITTING IN MXY MONOLAYERS VIA CHARGE DOPING

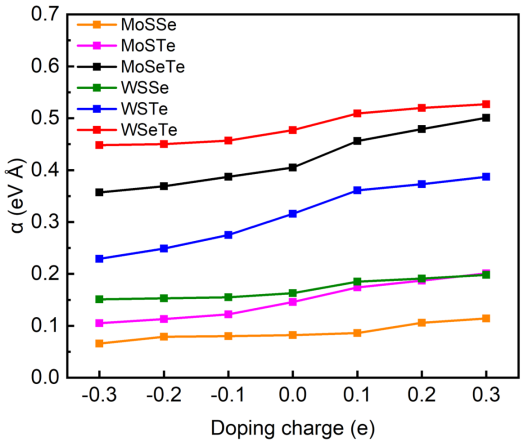

Fig. S1: Rashba constants α with charge doping of six Janus MXY monolayers.

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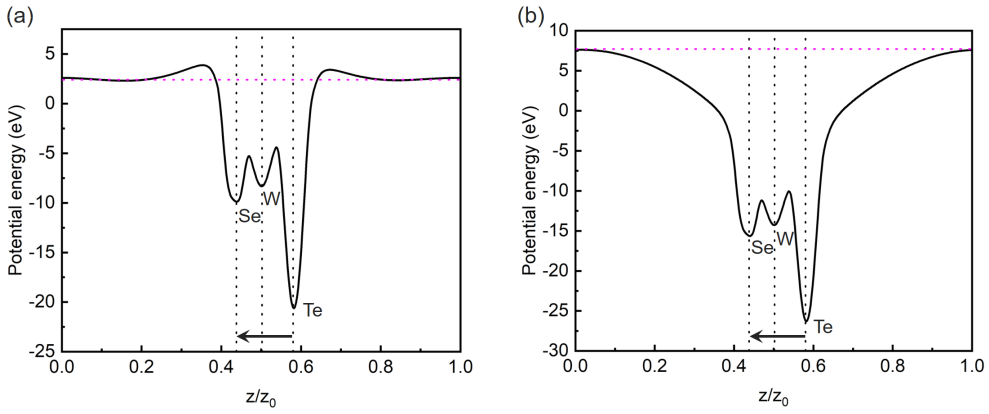

Fig. S2: Planar average of the electrostatic potential energy of the WSeTe monolayer with (a) electron doping (+0.3 e) and (b) hole doping (-0.3 e).

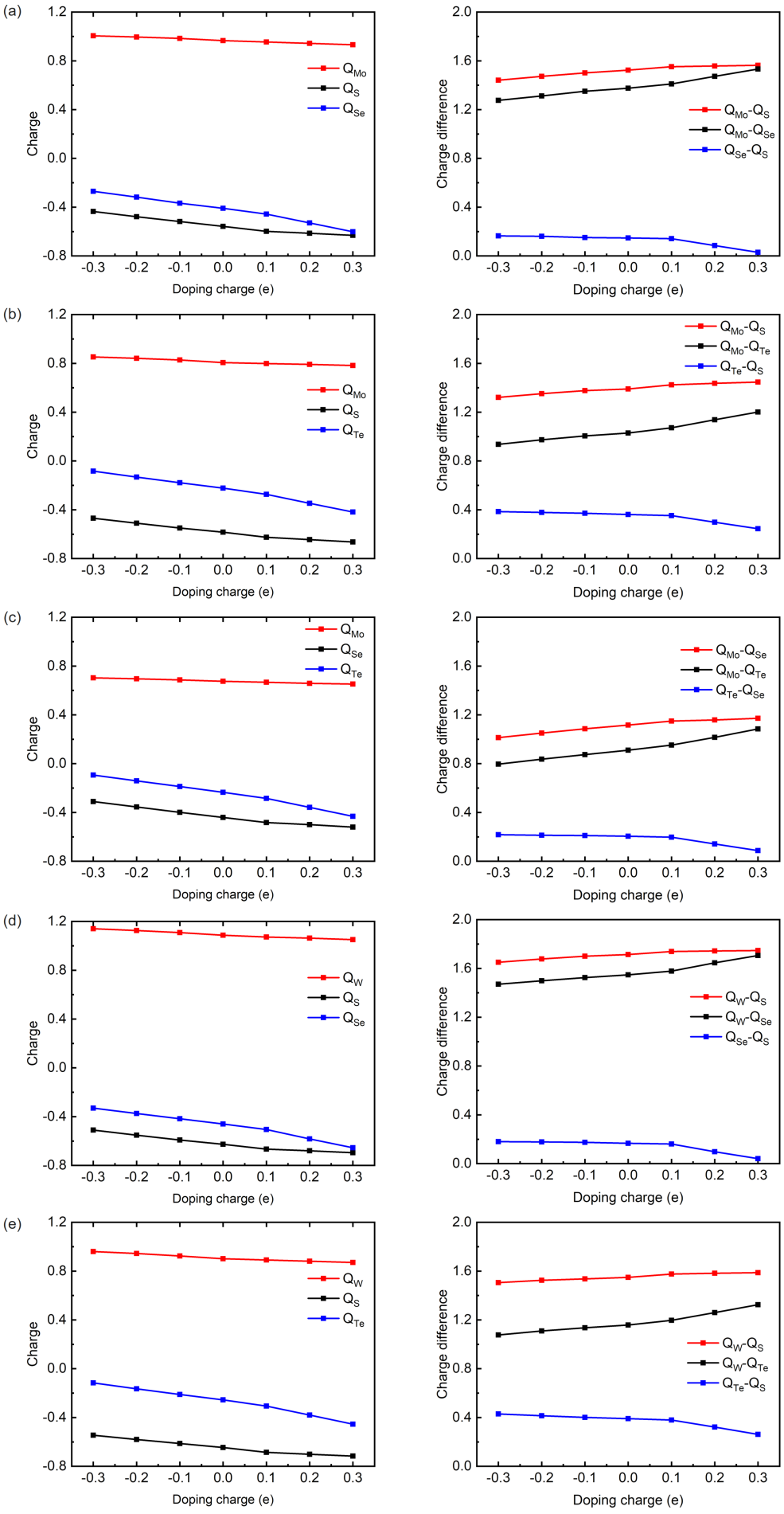

Fig. S3: The charge of M, X and Y and (b) Charge difference between M, X and Y in (a) MoSSe, (b) MoSTe, (c) MoSeTe, (d) WSSe and (e)WSTe monolayers.

III. MECHNISM OF TUNABLE RASHBA EFFECT VIA CHARGE DOPING

In Section 2, Rashba constant α is written as

$$\alpha = k_M Q_M + k_X Q_X + k_Y Q_Y \tag{1}$$

After some linear transformations

$$\begin{aligned} k_M &= k_{MX} + k_{MY} \\ k_X &= -k_{MX} - k_{YX} \\ k_Y &= -k_{MY} + k_{YX} \end{aligned} \tag{2}$$

α is transformed into the following expression as in Section 3:

$$\alpha = k_{MX}(Q_M - Q_X) + k_{MY}(Q_M - Q_Y) + k_{YX}(Q_Y - Q_X) \tag{3}$$

In order to get coefficients k_{MX}, k_{MY} and k_{YX}, we solve the nonhomogeneous linear equation $Ax = b$ with singular value decomposition (SVD)[1] by MATLAB software[2]. A consists of the charge differences between M and X, M and Y, Y and X ($Q_M - Q_X$, $Q_M - Q_Y$ and $Q_Y - Q_X$), b consists of corresponding Rashba constants, and x consists of the coefficients α' we want.

The MATLAB code for WSeTe monolayer is

```
A=[1.155  0.913  0.242
   1.190  0.955  0.234
   1.218  0.991  0.227
   1.248  1.024  0.224
   1.280  1.062  0.219
   1.289  1.129  0.160
   1.298  1.189  0.109];

b=[0.448
   0.450
   0.457
   0.477
   0.509
   0.520
   0.527];

[U S V] = svd(A);
B=U'*b;
for i=1:3
    Y(i)=B(i)/S(i,i);
end
x=V*Y'
```

So, k_{MX}, k_{MY} and k_{YX} equal to -8.330 V·Å, 8.755 V·Åand 8.541 V·Å, respectively.

IV. REASON FOR USING OPTB86B-VDW FUNCTIONAL IN JANUS TMD MONOLAYERS

We use a vdW functional for our DFT calculations, due to previous researches that MoSSe monolayer has been successfully synthesized in experiment and the DFT calculations are performed including the effects of long-range van der Waals interactions[3,4]. One of them uses van der Waals corrections with optB86b-vdW[5,6]. The theoretical lattice constants (3.23 Å) for the monolayers are consistent with the experimental value (3.22 Å), which is taken as the average of experimental lattice constants of MoS_2 and $MoSe_2$[4]. Other researchers[7] also investigate the Janus TMD monolayers with optB86b-vdW functional.

Besides, if the layer distance is larger than 3 Å, van der Waals interaction should be considered, which is proved in bilayer grephene[8,9]. In Janus TMD MXY monolayers, the layer distance between X atom and Y atom is larger than 3 Å. Hence, van der Waals interaction should be used in Janus TMD monolayers.

Our lattice parameter of MoSSe with optB86b-vdW functional (3.230 Å) matches well with experimental lattice parameter. The MoSSe monolayer without vdW interaction is also be optimized and the lattice parameter is equal to 3.250 Å. Thus, the optB86b-vdW functional performs well for our Janus TMD monolayers.

V. RASHBA CBM BANDS AROUND M POINT AND CORRESPONDING RASHBA CONSTANTS

The Rashba constants around the M point in the M-K direction is too steep to construct a quadratic curve for several Janus TMD monolayers.The Rashba CBM bands around M point along M-Γ direction of WSeTe monolayer is shown in Fig. S4. Rashba constants of Rashba CBM band around the M point in the M-Γ direction of WSSe, WSTe and WSeTe monolayers are listed in Table.S2. Rashba constants of other three Janus TMD nonolayers are too small to be detected.

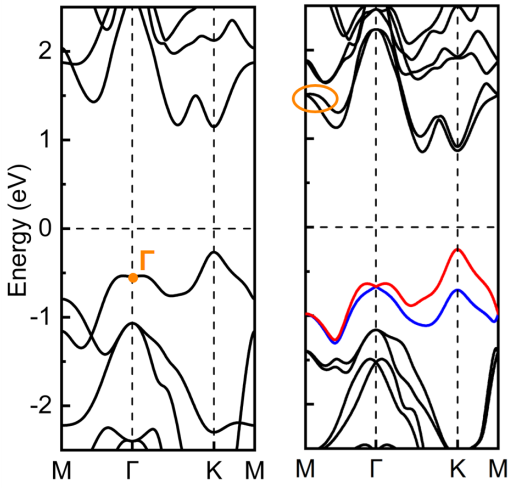

Fig. S4: Band structures without and with SOC of WSeTe monolayer. The orange ellipse indicates the Rashba CBM bands around M point along M-Γ direction.

Table S2: Rashba constants of Rashba CBM band around the M point in the M-Γ direction of WSSe, WSTe and WSeTe monolayers.

Doping charge (e)	Rashba constants (eV·Å)		
	WSSe	WSTe	WSeTe
-0.3	0.133	0.615	0.121
-0.2	0.144	0.651	0.128
-0.1	0.145	0.691	0.194
0	0.169	0.750	0.241
0.1	0.229	0.954	0.323
0.2	0.252	1.007	0.355
0.3	0.280	1.058	0.396

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