Understanding Bond Relaxation and Electronic Properties of T-Type WTe$_2$/MoS$_2$ Heterostructure using Binding Energy and Bond Charge Models

Hongrong Qiu, Hanze Li, Jiannan Wang, Yunhu Zhu, and Maolin Bo*

Binding energy and bond charge models and the topological concept to obtain the nonbonding, bonding, and antibonding states of the T-type WTe$_2$/MoS$_2$ heterostructure are combined. It is found that the electronic probability and electronic dispersion in the valence band of the WTe$_2$/MoS$_2$ heterostructure can be precisely determined based on electronic entropy. The energy-band projection method and electronic entropy are remarkable approaches for analyzing the electronic properties of various structures based on density functional theory calculations. Hence, a new method is provided to describe the electronic properties of T-type heterostructures, and the electron and bonding state probabilities are calculated.

1. Introduction

2D transition metal sulfides (TMDs) have attracted considerable attention owing to their advantageous properties, such as high current-carrying mobility and wide adjustable bandgaps.[15] These TMDs are used in numerous applications, including semiconductor devices and photocatalysis.[8] A sandwich-like structure is exhibited by TMDs with a transition metal element layer in the middle and a chalcogen element on each side.[8] Various TMD material types have different electrical and optical properties,[9,10] and specific properties can be obtained by forming TMD heterostructures.[11–13] Ceballos et al. reported the formation of long-lived indirect excitons in an MoS$_2$/MoSe$_2$ heterostructure.[14] Yelgel et al. showed that the WS$_2$/MoS$_2$ heterostructure has a stable structure and excellent electronic properties.[15] Compared with the 2 H heterostructure,[14,15] the vertical T-type heterostructure can better satisfy electron migration and provide a larger working area for optoelectronic devices.[16]

Vertical heterostructures offer new opportunities to design novel optoelectronic devices, which present a rich collection of physics and electronics.[17–20] For example, a graphene and MoS$_2$ vertical heterojunction was used to realize phototransistors with ultrahigh photoresponsivity exceeding 10$^7$ A W$^{-1}$.[21] Shin et al. reported that Si/MoS$_2$ vertical heterojunctions have high-responsivity photodetectors.[22] Zhan et al. found that vertical Cu$_2$S/CdS heterojunctions exhibit a higher photoelectric conversion capability compared with devices based on single-component CdS or Cu$_2$S junctions.[23] Moreover, the electronic properties of vertical T-type heterostructures are related to interface bond formation.[24,25] Understanding the fundamental nature of the interface bond formation and its consequences for the electronic binding energy, as well as determination of the relevant energetics, presents a great challenge. A method for purifying the surface and interface information about atomic bond and electronic is highly desired.

In this work, we constructed a T-type WTe$_2$/MoS$_2$ heterostructure and modeled its structure through density functional theory (DFT) calculations.[26] We combined the binding energy and bond charge (BC) models and the topological concept to obtain the nonbonding, bonding, and antibonding states of the T-type WTe$_2$/MoS$_2$ heterostructure. Subsequently, we investigated the electronic properties of the T-type WTe$_2$/MoS$_2$ heterostructure using an energy-band projection method. We analyzed the density of state (DOS) and band structure in each energy range along each Brillouin path. We used the concept of electronic entropy to express the electronic dispersion in the T-type WTe$_2$/MoS$_2$ heterostructure valence band. This energy-band projection method and the concept of electronic entropy provide a new approach for examining the electronic properties of various structures.

2. Methods

2.1. DFT Calculation

We present the process of building T-type WTe$_2$/MoS$_2$ heterostructure in Figure S1, Supporting Information. The structure building is assured via the used Cambridge Sequential Total Energy Package (CASTEP) code, and the DFT is used for the electronic feature’s calculations of heterostructure.[27] The interaction between ion and valence electrons is described by the projector augmented wave method. The crystal structures were geometrically optimized using the Broyden–Fletcher–Goldfarb–Shanno method with fixed basis quality as in CASTEP.[28] We used the hybrid exchange–correlation functional (HSE06) to
describe the electron exchange and correlation potential,[29] the cut-off energy of the plane-wave basis set was 650 eV, and \(3 \times 2 \times 1\) k-point grids were used. In the calculations, the energy converged to \(10^{-6}\) eV, and the force on each atom converged to <0.01 eV Å\(^{-1}\).

### 2.2. Binding Energy Model

The Hamiltonian is given by

\[
H = -\frac{\hbar^2 V^2}{2m} + V_{\text{atom}}(r) + V_{\text{cr}}(r)(1 + \Delta \eta)
\]  

(1)

Here, \(V_{\text{atom}}(r)\) is the intra-atomic potential of the atom, \(V_{\text{cr}}(r)\) is the potential of the crystal, and the interaction potential changes with the coordination environment and during chemical reactions. The electronic binding energy (BE) of the \(v\)th energy band \(E_v(0)\) and \(E_{v}(B)\) are

\[
E_v(0) = -\langle v, i | -\frac{\hbar^2 V^2}{2m} + V_{\text{atom}}(r) | v, i \rangle
\]

(2)

\[
E_v(x) - E_v(0) = -\alpha_v \sum_j f(k) \cdot \beta_j - \alpha_v \left(1 + \sum_j f(k) \cdot \beta_j \right)
\]

\[
\equiv -\alpha_v (1 + \Delta \eta) \propto \langle E_v \rangle
\]

(3)

Here, \(E_v(x)\) and \(E_v(0)\) are the energy levels of atoms and an isolated atom, respectively, and \(\alpha_v\) and \(\beta_j\) contribute to the energy band width. In the localized band of core levels, \(\beta_j\) is considerably small, so \(\alpha_v\) determines the energy band of the core levels.

\[
\alpha_v = -\langle v, i | V_{\text{cr}}(r) | v, i \rangle \propto \langle E_{v} \rangle
\]

(4a)

\[
\beta_j = -\langle v, i | V_{\text{cr}}(r) | v, j \rangle \sum_j f(k) \beta_j \propto \langle E_v - E_b \rangle
\]

(4b)

Here, \(|v, i\rangle\) represents the wave function, and the periodic factor \(f(k)\) is in the form of \(e^{\imath k \cdot r}\), where \(k\) is the wave vector. Further, \(\beta\) is dependent on the overlap between orbitals centered at two neighboring atoms.

The bond energy \(E_b\) uniquely determines the core-level BE shift.[30]

\[
z_x = \frac{12}{8 \ln(2 \Delta E_v(x) / \Delta E_{v}(B)) + 1} (\Delta E_v(x) \geq 0)
\]

(5)

where \(z_x\) is the atomic coordination number of an atom in the \(x\)th atomic layer from the surface.

\[
\Delta E_v(x) = \Delta E_v(x) + \Delta E_v(B) / \Delta E_{v}(B)
\]

(6)

\[
\frac{\Delta E_v(w_b)}{\Delta E_{v}(w_b)} \propto \frac{E_v}{E_b} = \gamma = 1 + \Delta \eta = e^{-m} = \left(\frac{d_x}{d_b}\right)^{-m}
\]

(7)

Here, \(Z\) is the atomic charge parameter \((Z = -1)\) (isolated atom), \(Z = 0\) (bulk atom), \(Z = +\Delta \eta\) \((\gamma > 0)\) (charged positive atoms), and \(Z = -\Delta \eta\) \((\gamma < 0)\) (charged negative atoms). Thus, the core-electron BE shifts are 0, \(-\sum_j \langle v, i | V_{\text{cr}}(r) | v, j \rangle \propto \langle E_v \rangle\), \(-\sum_j \langle v, i | V_{\text{cr}}(r) | v, i \rangle \propto \langle E_v \rangle\), respectively.[31] Equation (16) provides estimates for the bond energy \(E_b\) and bond length \(d_x\) and \(\Delta E_v(w_b) \propto \Delta E_v(B)\) is the spectral full width of the bulk component \((w_b)\) of the \(v\)th energy level; the BE shift width for the \(v\)th energy level surface component \((w_s)\) is \(\Delta E_s(w_s) = \Delta E_s(w_b) + \Delta E_s(x)\). However, the actual spectral intensities and shapes are subject to polarization effects and measurement artifacts. We can calculate the chemisorption and defect-induced interface bond energy ratio \(\gamma\) with the known reference value of \(\Delta E_v(x) = E_v(x) - E_v(B), \Delta E_v(B) = E_v(B) - E_v(0),\) and \(\Delta E_v(x) = E_v(x) - E_v(0)\) derived from the surface via DFT calculations and X-ray photoelectron spectroscopy analysis. Hence, we obtain

\[
\delta \eta = \frac{\Delta E_v(x) + \Delta E_v(B)}{\Delta E_v(B) - 1} = \gamma - 1 \quad \text{(RBER)}
\]

(11)

\[
\delta E_b = (d_x/d_b) - 1 = \gamma^{-1} - 1 \quad \text{(RLBS)}
\]

(12)

\[
\delta E_d = (E_i/d_i^4)/(E_b/d_b^4) - 1 = \gamma^4 - 1 \quad \text{(RBED)}
\]

(13)

\[
\Delta E_v(x) \propto -\gamma \langle v, i | V_{\text{cr}}(r) | v, i \rangle \propto \langle E_v - E_b \rangle
\]

(14)

Thus, one can drive the interface binding energy ratio parameter \(\delta \eta\). If \(\delta \eta < 0\), then the binding energy is reduced, and the crystal and the bond potential are weakened. Conversely, if \(\delta \eta > 0\), then the binding energy increases, and the crystal and the bond potential become stronger. The relative local bond strain \(\delta E_b\) indicates the relative contraction of the atomic bond length \(d_x\). The relative bond energy density \(\delta E_d\) is the energy density of the atomic bond with energy \(E_b\).

### 3. Results and Discussion

#### 3.1. Electronic Properties of the T-Type WTe\(_2\)/MoS\(_2\) Heterostructure

We aimed to analyze the energetics, electronic properties, and atomic structure of the T-type WTe\(_2\)/MoS\(_2\) heterostructure. Figure 1 presents the structure of the T-type WTe\(_2\)/MoS\(_2\) heterostructure, which has a T-shape. The T-type WTe\(_2\)/MoS\(_2\)
heterostructure lattice parameters are presented in Table 1 (a = 6.235 Å, b = 12.980 Å, c = 14.638 Å), and the optimized atomic coordinate results are presented in Table 2. The band structure of the WTe2/MoS2 heterostructure is shown in Figure 2. The bandgap of the T-type WTe2/MoS2 heterostructure is 0.328 eV. Zheng et al. reported that the bandgap of the strained 2H-type MoTe2/WTe2 heterostructure is 0.29 eV.[32] This result is consistent with our reported result. In addition, we observed a flat band in the bandgap of the T-type WTe2/MoS2 heterostructure. In Figure 2, a quite distinct horizontal band with almost no dispersion appears when the Fermi level (E_f = 0) is near zero. Therefore, the flat band is able to adjust the electron states of the band structure.

The total DOS of the T-type WTe2/MoS2 heterostructure is shown in Figure 2. The figure shows that the zero domain appears at the Fermi level (E_f = 0). The figure also presents the electronic distribution of the valence band maximum of the T-type WTe2/MoS2 heterostructure that has an energy of 0 eV and is primarily distributed on the Fermi level E_f. The electron in the conduction band minimum is at an energy of ≈0.328 eV. The partial density of states (PDOS) is shown in Figure 3. From the PDOS, we can observe that the electron density near the valence band Fermi level is primarily concentrated in −0.236 and −1.842 eV, and the electrons in the conduction band are concentrated in 1.842 and 2.291 eV. We calculate the atomic local DOS of the WTe2/MoS2 heterojunction of Mo (4p4d5s), S (3s3p), W (6s5d), and Te (5s5p) atoms. As shown in Figure 3, the WTe2/MoS2 heterostructure of main electron contributions to the conduction band comes from Mo 4d and S 3p orbitals. Moreover, the W and Te atoms have less electron contribution to the Fermi level.

### Table 1. Lattice parameters of the t-type WTe2/MoS2 heterostructure.

| Structure    | Angle [deg] | Lattice parameter [Length] |
|--------------|-------------|----------------------------|
|              | α [deg] | β [deg] | γ [deg] | a [Å] | b [Å] | c [Å] |
| WTe2/MoS2    | 90.00    | 90.00   | 76.10   | 6.235 | 12.980 | 14.638 |

### Table 2. T-type WTe2/MoS2 heterostructure atomic fractional coordination after optimization.

| Atom | x   | y   | z   |
|------|-----|-----|-----|
| Te1  | 0.687 | 0.000 | 0.111 |
| Te2  | 0.114 | 0.140 | 0.101 |
| Te3  | 0.548 | 0.280 | 0.099 |
| Te4  | 0.972 | 0.426 | 0.109 |
| Te5  | 0.398 | 0.574 | 0.110 |
| Te6  | 0.827 | 0.719 | 0.099 |
| Te7  | 0.254 | 0.859 | 0.101 |
| Te8  | 0.338 | 0.000 | 0.229 |
| Mo22 | 0.025 | 0.504 | 0.501 |
| Mo23 | 0.479 | 0.496 | 0.501 |
| Mo24 | 0.284 | 0.373 | 0.556 |
| Mo25 | 0.795 | 0.372 | 0.555 |
| Mo26 | 0.168 | 0.627 | 0.556 |
| Mo27 | 0.657 | 0.627 | 0.556 |
| Mo28 | 0.228 | 0.500 | 0.678 |
| Mo29 | 0.728 | 0.499 | 0.678 |
| S30  | 0.041 | 0.375 | 0.742 |
| S31  | 0.541 | 0.373 | 0.739 |
| S32  | 0.418 | 0.624 | 0.743 |
| S33  | 0.914 | 0.626 | 0.739 |
| Mo34 | −0.022 | 0.503 | 0.863 |
| Mo35 | 0.481 | 0.495 | 0.863 |
| S36  | 0.287 | 0.375 | 0.924 |
| S37  | 0.789 | 0.372 | 0.921 |
| S38  | 0.162 | 0.626 | 0.922 |
| S39  | 0.663 | 0.623 | 0.925 |

### 3.2. Energy-Band Projection Method

We used a new method to analyze the energy distribution near the Fermi surface of the T-type WTe2/MoS2 heterostructure. We can also use this method to analyze the probability of electrons appearing in various energy ranges along each Brillouin path.
In the following section, we will use electronic entropy to explain the reason. First, we normalized the DOS, extracted the points with energy between $-3$ and $3$ eV in the DOS, and listed the extracted energy as the basis. Simultaneously, we extracted the energy points and equally divided the corresponding points into 3000 points; after the equalization, the point sequence was integrated and normalized. Second, we stratified the normalized DOS. The point list was integrated piecewise, sorted with a unit of 0.2 eV, and divided into 11 levels (from $-3$ to $3$ eV). Different levels are represented by various colors for probability of electrons. Third, we conducted hierarchical processing on the band structure. The points with energy between $-3$ and $3$ eV, and the corresponding data were extracted completely, and the energy range obtained in the second step was matched with the color. The difference between the energy-band projection method and Fatband software for 2D band projection is that the latter uses the $k$-point projection, while the former takes the energy projection for subsequent analyses.

Figure 4 displays a projection flow picture; here, the DOS is projected on the band structure diagram based on energy, and different colors are used to represent the electron probabilities. According to Shannon, for a discrete probability distribution, the entropy is defined as follows:

$$H(X) = -\sum_{x\in X} P(x_i) \log P(x_i)$$  \hspace{1cm} (15)

For the entropy of electrons, the random variable $X$ represents the energy range corresponding to a point on the Brillouin region path, $x_i$ represents one of the energy ranges, and $P(x_i)$ represents the probability of the electron appearing in a certain energy range. The electronic entropy corresponding to each energy range can be obtained using the data displayed in Table 3. Near the valence band (from $-3$ to $0$ eV), the electron entropy was 0.116 bit. The entropy near the shallow energy level of the valence band (from $-0.6$ to $0$ eV) was calculated as 0.018 bit. Furthermore, for the conduction band (0–$3$ eV), the electron...
entropy was 0.090 bit. Meanwhile, the entropy near the shallow energy level of the conduction band (0–1.2 eV) is 0.013 bit.

The order of the electronic entropy values in each range was consistent with that of the corresponding electron appearance probability. Figure 5 presents the energy band structure of the processed DOS projection. We can observe that the color is closer to black (0 bit). Thus, the smaller the entropy value, the smaller the probability of electrons appearing; meanwhile, the color is closer to red (0.055 bit), the greater the entropy value, and the greater the probability of electrons appearing. Noticeably, around the Fermi level, especially the energy range of the valence band (from −0.6 to 0 eV), the probability of the occurrence of electrons is generally low. This indicates that the shallow energy level of the valence band is very close to the cavitation-bound state of the valence band in the range from −0.6 to 0 eV. The relationship between the valence band cavitation-bound state and the conduction band cavitation-bound state is

\[
E_C - E_f = \frac{kT}{2n_c} \ln \left( \frac{N_c}{N_D} \right) - \frac{\sum_{x \in X} x_c \log(x_c)}{\sum_{x \in X} x_c} \frac{H_c(x_c)}{H_v(x_c)}
\]

(16)

Here, \(E_C\) is the energy near the minimum conduction band value, \(E_v\) is the energy near the maximum valence band value, \(E_f\) is the Fermi level energy, \(N_c\) is the effective state density of electrons at the bottom of the conduction band, \(N_v\) is the effective state density of electrons at the top of the valence band, \(N_D\) is the concentration of electrons after complete ionization, and \(N_A\) is the concentration of holes after complete ionization. The electrons or holes in the shallow valence band energy level are ionized at a slightly higher temperature to become free electrons in the conduction band and free holes in the valence band, which promotes electricity conduction.

3.3. Bonding States of the T-Type WTe₂/MoS₂ Heterostructure

We considered a free quantum particle of charge \(e\). We confined charge \(e\) in one dimension and subjected it to periodic boundary conditions (i.e., a particle on a ring). Thus, the free Hamiltonian of the system takes the following forms:\(^{[36]}\)

\[
H = -\frac{\hbar^2}{2m} \left( \nabla - \frac{q}{\hbar c} A(\vec{r}) \right)^2 + \frac{1}{2} \left( -i \partial_\phi - A(\vec{r}) \right)^2
\]

(17)

\[
\psi_\phi(\phi) = \frac{1}{\sqrt{2\pi}} \exp(i\phi)
\]
where $\hbar$ is the Planck’s constant, $q$ is the amount of charge, $c$ is the speed of light, $r$ is the electron radius, and $\phi$ is the field.

To understand a particle on a ring, we express the field $\phi$ as mapping

$$\phi: S_1 \to S_2, \tau \to \phi(\tau)$$

from the unit circle $S_1$ into another circle. Mappings of this type can be assigned to a winding number $W$ (see Figure 6). The number of times $\phi(\tau)$ winds around the unit circle as $\tau$ progresses

Table 3. Probability of the energy range and corresponding color.

| Probability (Integral value) | Energy range | Color |
|------------------------------|--------------|-------|
| 0.038                        | (-3.0 to -2.8 eV) |       |
| 0.040                        | (-2.8 to -2.6 eV) |       |
| 0.041                        | (-2.6 to -2.4 eV) |       |
| 0.035                        | (-2.4 to -2.2 eV) |       |
| 0.046                        | (-2.2 to -2.0 eV) |       |
| 0.053                        | (-2.0 to -1.8 eV) |       |
| 0.042                        | (-1.8 to -1.6 eV) |       |
| 0.030                        | (-1.6 to -1.4 eV) |       |
| 0.041                        | (-1.4 to -1.2 eV) |       |
| 0.042                        | (-1.2 to -1.0 eV) |       |
| 0.042                        | (-1.0 to -0.8 eV) |       |
| 0.037                        | (-0.8 to -0.6 eV) |       |
| 0.020                        | (-0.6 to -0.4 eV) |       |
| 0.013                        | (-0.4 to -0.2 eV) |       |
| 0.023                        | (-0.2 to 0.0 eV) |       |
| 0.003                        | (0.0 to 0.2 eV)  |       |
| 0.001                        | (0.2 to 0.4 eV)  |       |
| 0.014                        | (0.4 to 0.6 eV)  |       |
| 0.018                        | (0.6 to 0.8 eV)  |       |
| 0.019                        | (0.8 to 1.0 eV)  |       |
| 0.023                        | (1.0 to 1.2 eV)  |       |
| 0.043                        | (1.2 to 1.4 eV)  |       |
| 0.034                        | (1.4 to 1.6 eV)  |       |
| 0.037                        | (1.6 to 1.8 eV)  |       |
| 0.046                        | (1.8 to 2.0 eV)  |       |
| 0.050                        | (2.0 to 2.2 eV)  |       |
| 0.035                        | (2.2 to 2.4 eV)  |       |
| 0.037                        | (2.4 to 2.6 eV)  |       |
| 0.042                        | (2.6 to 2.8 eV)  |       |
| 0.030                        | (2.8 to 3.0 eV)  |       |

Only the index of topological sector is the field $\phi$. $S_{top}$ is the topological sector of the field contribution.

The atomic bond relaxation is expressed based on the BC model, as follows:

$$E_i \propto V_{cry}(r_g) = qA(\tau_g) = qS_{top}[\phi]/i2\pi W$$

(20)

$$\Delta E_s(x) \approx -\sum_j f(k)\langle v, i|qA(\tau_g)|v, j\rangle$$

$$= -\sum_j f(k)\langle v, i|qS_{top}[\phi]/i2\pi W|v, j\rangle \propto \Delta E_i$$

(21)
Here $E_i$ is the single bond energy, $\Delta \rho(r_{ij})$ is the deformation charge density, $d_i$ is the atom bond length, $V_{cry}(r_{ij})$ is the crystal potential, $B$ represents bulk atoms, and $m$ is the bond nature indicator. In Equation (6) and (7), chemical bonds are associated with the topological effects and quantum indicator. In Equation (8), the potential function $\gamma V_{cry}(r_{ij})$ may become deeper ($\gamma > 1$) or shallower ($\gamma < 1$) than the corresponding $V_{cry}(r_{ij})$ of the specific constituent. Equation (9) describes the relationship between the deformation charge density $\Delta \rho(r_{ij})$ and the crystal potential $V_{cry}(r_{ij})$. This method is generally applicable to the heterostructure bonding state. The method has been applied in a variety of structure analytical bonding states, such as Sb/MoSe$_2$ and BN/SiC heterostructures. This method is suitable for the bonding, antibonding, and nonbonding states of s, p, and d orbital electron hybridization.

Figure 7 depicts the deformation charge density. We believe that the formation of the WTe$_2$/MoS$_2$ heterostructure is primarily because of the contributions of bonds, electrons, and charges. Using Equation (7), we calculated crystal potentials of the antibonding, nonbonding, and bonding states of the WTe$_2$/MoS$_2$ heterostructure. Table 4 presents the electronic states of the three chemical bonds calculated by DFT. The nonbonding, bonding, and antibonding states are displayed in the deformation charge density. The dark-red area indicates the bonding state. Bonding states indicate that the electron density increases in the region, but the electron density overlaps with the other atomic electron densities, indicating that the atom is in the bonding state. The light-red area is a nonbonding state. Nonbonding states indicate that the electron density increases in the region, but the surrounding electron density does not overlap with the electron density of the other atoms. The white area represents the antibonding state. The antibonding electrons will displace the surrounding electrons, creating a cloud of positively charged "holes" that move along with the electrons. The antibonding electrons call these correlated holes, and the correlation effect is shown as a shielding effect on electrons. Furthermore, we analyzed the deformation charge density to define the atomic bonding information.

4. Conclusion

We used DFT to calculate the constructed T-typed WTe$_2$/MoS$_2$ heterostructure and combined the BC model and the topological concept to obtain the nonbonding, bonding, and antibonding states of the T-typed WTe$_2$/MoS$_2$ heterostructure. Furthermore, the energy-band projection method was used to study the electronic properties of the WTe$_2$/MoS$_2$ heterostructure. Using the formula of electronic entropy, the valence band (from $-3$ to 0 eV), shallow energy level of the valence band (from $-0.6$ to 0 eV), conduction band (0–3 eV), and shallow energy level of the conduction band (0–1.2 eV) were calculated to represent the electronic probability of the WTe$_2$/MoS$_2$ heterostructure. The energy-band projection method and the concept of electronic entropy provide a new approach for studying the electronic properties of various structures.

Table 4. Chemical bonding states, deformation charge density $\Delta \rho(r_{ij})$, and potential functions $V_{cry}(r_{ij})$ of van der Waals heterojunctions obtained using the calculated BC model ($e_0 = 8.85 \times 10^{-12}$ C$^2$ N$^{-1}$ m$^{-3}$, $e = 1.60 \times 10^{-19}$ C).

| Bonding State | $\Delta \rho(r_{ij})$ (e$^{-3}$) | $V_{cry}(r_{ij})$ (eV) |
|-------------|-----------------|----------------|
| Nonbonding  | $-0.976$         | $-2.112$      |
| Bonding     | $1.298$          |                |

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements

This work was supported by the Scientific and Technological Research Program of Chongqing Municipal Education Commission.
Conflict of Interest
The authors declare no conflict of interest.

Data Availability Statement
Research data are not shared.

Keywords
band structure, bond relaxation, density of states, electronic properties, T-type heterostructures, WTe₂/MoS₂

Received: August 18, 2021
Revised: November 8, 2021
Published online: December 16, 2021

[1] J. Bonde, P. G. Moses, T. F. Jaramillo, K. Nørskov, I. Chorkendorff, Faraday Discuss. 2009, 140, 219.
[2] A. Litke, T. Weber, J. P. Hofmann, E. J. M. Hensen, Appl. Catal. B 2016, 198, 16.
[3] T. A. Pecoraro, R. R. Chianelli, J. Catal. 1981, 67, 430.
[4] S. Das, J. Appenzeller, Phys. Status Solidi RRL 2013, 7, 268.
[5] H. Ning, Y. Zeng, S. Zuo, S. V. Kershaw, Y. Hou, Y. Li, X. Li, J. Zhang, Y. Yi, L. Jing, J. Li, M. Gao, ACS Nano 2021, 13, 873.
[6] A. Splendiani, L. Sun, Y. Zhang, T. Li, J. Kim, C. Y. Chim, G. Galli, F. Wang, Nano Lett. 2010, 10, 1271.
[7] Q. Zhang, Z. Liu, Y. Sun, H. Yang, J. Jiang, S.-K. Mo, Z. Hussain, X. Qian, L. Fu, S. Yao, M. Lu, C. Felser, B. Yan, Y. Chen, L. Yang, Phys. Status Solidi RRL 2017, 11, 1700209.
[8] X. Huang, J. Tang, B. Luo, R. Knibbe, T. Lin, H. Hu, M. Rana, M. Hu, X. Zhu, Q. Gu, D. Wang, L. Wang, Adv. Energy Mater. 2019, 9, 1901872.
[9] J. He, K. Hummer, C. Franchini, Phys. Rev. B 2014, 89, 075409.
[10] G. Eda, H. Yamaguchi, D. Voiry, T. Fujita, M. Chen, M. Chhowalla, Nano lett. 2011, 11, 5111.
[11] J. Zhang, L. Zhang, X. Ma, Z. Ji, Appl. Surf. Sci. 2018, 430, 424.
[12] H. Croy-Diaz, F. Bertran, C. Chen, J. Avila, J. Rault, P. Le Fèvre, M. C. Asensio, M. Batzill, Phys. Status Solidi RRL 2015, 9, 701.
[13] M.-Y. Qian, Z.-L. Yu, Q. Wan, P.-B. He, B. Liu, J.-L. Yang, C.-M. Xu, M.-Q. Cai, Phys. Status Solidi RRL 2020, 14, 2000016.
[14] F. Ceballos, M. Z. Bellus, H.-Y. Chiu, H. Zhao, ACS Nano 2014, 8, 12717.
[15] C. Yelgel, O. C. Yelgel, O. Gürleren, J. Appl. Phys. 2017, 122, 065303.
[16] J. Shim, D.-H. Kang, Y. Kim, H. Kum, W. Kong, S.-H. Bae, I. Almansouri, K. Lee, J.-H. Park, J. Kim, Carbon 2018, 133, 78.
[17] R. Frisenda, A. J. Molina-Mendoza, T. Mueller, A. Castellanos-Gomez, H. S. J. van der Zant, Chem. Soc. Rev. 2018, 47, 3339.
[18] Y. Xue, Y. Zhang, Y. Liu, H. Liu, J. Song, J. Sophia, J. Liu, Z. Xu, Q. Xu, Z. Wang, J. Zheng, Y. Liu, S. Li, Q. Bao, ACS Nano 2016, 10, 573.
[19] J. H. Yu, H. R. Lee, S. S. Hong, D. Kong, H. W. Lee, H. Wang, F. Xiong, S. Wang, Y. Cui, Nano Lett. 2015, 15, 1031.
[20] W. J. Yu, Y. Liu, H. Zhou, A. Yin, Z. Li, Y. Huang, X. Duan, Nat. Nanotechnol. 2013, 8, 952.
[21] W. Zhang, C.-P. Chuu, J.-K. Huang, C.-H. Chen, M.-L. Tsai, Y.-H. Chang, C.-T. Liang, Y.-Z. Chen, Y.-L. Chueh, J.-H. He, M.-Y. Chou, L.-J. Li, Sci. Rep. 2014, 4, 3826.
[22] G. H. Shin, J. Park, K. J. Lee, G.-B. Lee, H. B. Jeon, Y.-K. Choi, K. Yu, S.-Y. Choi, ACS Appl. Mater. Interfaces 2019, 11, 7626.
[23] Y. Zhan, Z. Shao, T. Jiang, J. Ye, X. Wu, B. Zhang, K. Ding, D. Wu, J. Jie, J. Mater. Chem. A 2020, 8, 789.
[24] M. Bo, H. Li, A. Deng, L. Li, C. Yao, Z. Huang, C. Peng, Mater. Adv. 2020, 1, 1186.
[25] M. Bo, H. Li, Z. Huang, L. Li, C. Yao, AIP Adv. 2020, 10, 015321.
[26] C. J. Cramer, D. G. Truhlar, Phys. Chem. Chem. Phys. 2009, 11, 10757.
[27] F. Stéglich, S. Wirth, Rep. Prog. Phys. 2016, 79, 084502.
[28] J. D. Head, M. C. Zerner, Chem. Phys. Lett. 1985, 122, 264.
[29] J. Heyd, G. E. Scuseria, M. Ernzerhof, J. Chem. Phys. 2003, 118, 8207.
[30] J. Liao, B. Sa, J. Zhou, R. Ahuja, Z. Sun, J. Phys. Chem. C 2014, 118, 17594.
[31] S. Peredkov, S. L. Sorensen, A. Rosso, G. Ohrwall, M. Lundwall, T. Rander, A. Lindblad, H. Bergersen, W. Pokapanich, S. Svensson, O. Björnholm, N. Märtensson, M. Tchaplyguine, Phys. Rev. B 2007, 76, 081402.
[32] Z. Zheng, X. Wang, W. Mi, J. Phys.: Condens. Matter. 2016, 28, 505003.
[33] A. E. Cocke, P. Z. Fulé, J. E. Crouse, Int. J. Wildland Fire 2005, 14, 189.
[34] A. E. Cocke, P. Z. Fulé, J. E. Crouse, Int. J. Wildland Fire 2005, 14, 189.
[35] M. C. Payne, M. P. Teter, D. C. Allan, T. A. Arias, J. D. Joannopoulos, Rev. Mod. Phys. 1992, 64, 1045.
[36] A. Mohajeri, M. Alipour, Chem. Phys. 2009, 360, 132.
[37] M. A. Omar, Elementary Solid State Physics: Principles and Applications, Addison-Wesley, Boston, MA, USA 1993.
[38] C. Q. Sun, Relaxation Of The Chemical Bond: Skin Chemisorption Size Matter ZTP Mechanics H₂O Myths (Ed: C. Q. Sun), Springer Singapore, Singapore 2014, pp. 203–221.