Supporting Information (SI)

Comparative Study of Janus B_2XY (X, Y=S, Se, Te) and F-BNBN-H Monolayers for Water Splitting: Reveal the Positive and Negative Roles of Intrinsic Dipole

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SI-1 The magnetic properties of B_2XY monolayers

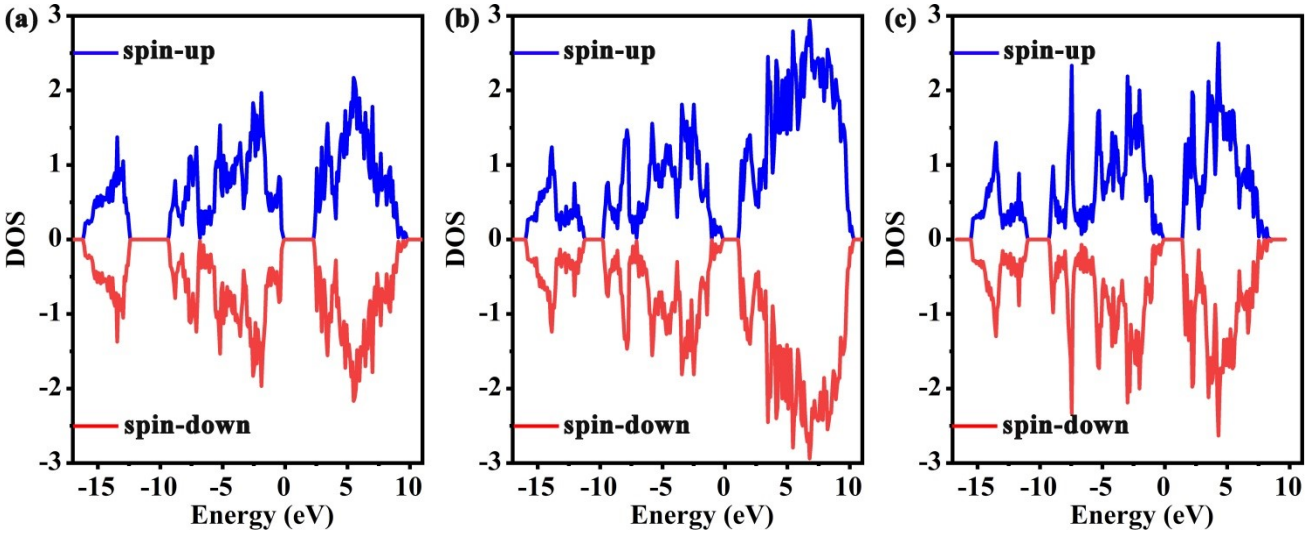

Figure S1 The electronic states of spin-up and spin-down channels for B_2SSe, B_2STe and B_2SeTe monolayers.

The spin-polarized calculations (ISPIN=2) are performed to identify the magnetic properties of B_2XY monolayers. As shown in **Figure S1a-c**, the electronic states in spin-up and spin-down channels are completely symmetric with respect to the zero level, which demonstrates that B_2XY monolayers are non-magnetic.

SI-2 The VBM and CBM charge density of the B_2STe monolayer

The reason for the disappearing VBM charge density of the B_2STe monolayer is that the isosurface value is set as large as 0.0075 e·Bohr^{-3} in **Figure 3** of our main manuscript. As shown in **Figure S2**, when the isosurface is reset as 0.0011 e·Bohr^{-3}, the VBM of the B_2STe monolayer is more visualized and mainly distributed at the top surface.

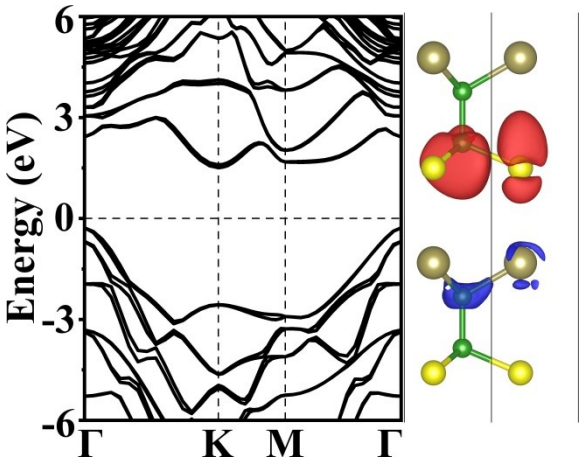

Figure S2 The CBM and VBM charge density of the B_2STe monolayer. The isosurface value is taken as 0.0011 e·Bohr^{-3}.

SI-3 The band structure, VBM and CBM levels with the Fermi level not reset

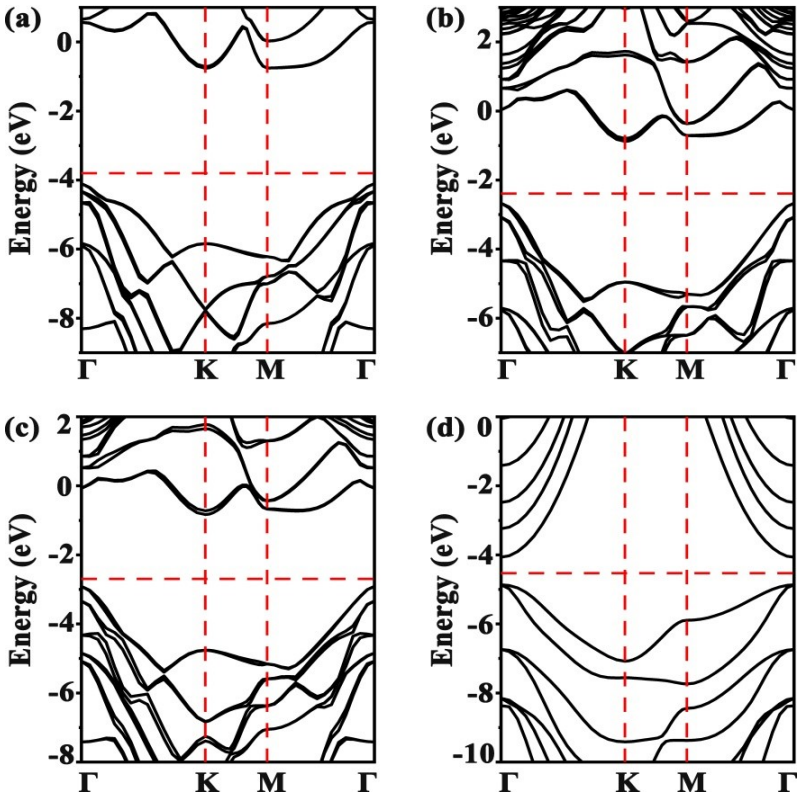

Figure S3 The band structures of (a) B_2SSe, (b)B_2STe and (c) B_2SeTe monolayers at the HSE06+SOC level; (d) The band structure of the F-BNBN-H monolayer at the HSE06 level, with fermi level not reset.

Table S1. The CBM, VBM and Fermi levels of B_2XY and F-BNBN-H monolayers directly extracted from the VASP outcome, with fermi level not reset.

Species	$E_{CBM}^{'}$	$E_{VBM}^{'}$	E_{Fermi}
B_2SSe	-0.75	-4.10	-3.80
B_2STe	-0.81	-2.65	-2.38
B_2SeTe	-0.83	-2.92	-2.69
F-BNBN-H	-4.06	-4.86	-4.53

SI-4 The band alignment of B_2XY and F-BNBN-H monolayers calculated by shifting the redox potentials of water

As shown in **Figure S4a-d**, the band alignments of B_2XY and F-BNBN-H monolayers are also obtained by shifting the redox potentials [1, 2]. For B_2XY monolayers, their CBM is located on the bottom $(00\bar{1})$ surface, while their VBM is distributed on the top surface. The CBMs of B_2XY monolayers are higher than the shifted reduction potential of water for the bottom $(00\bar{1})$ surface. The VBM of B_2SSe monolayer is just below the shifted oxidation potential of water for the top (001) surface, and thus the B_2SSe monolayer has very limited photocatalytic ability. The VBMs of B_2STe and B_2SeTe monolayers are higher than the shifted oxidation potential of water for the top (001) surface, which indicates the vanishing photocatalytic ability for B_2STe and B_2SeTe monolayers. Summarily, the B_2SSe monolayer has limited photocatalytic ability, while B_2STe and B_2SeTe monolayers possess no photocatalytic ability.

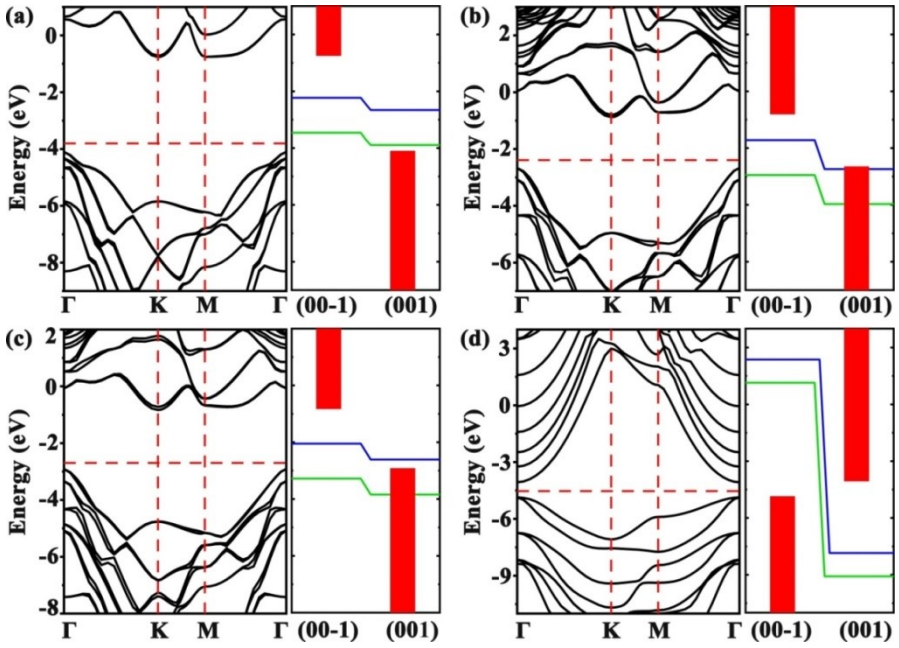

Figure S4 The band alignment of (a) B_2SSe, (b) B2STe, (c) B2SeTe and (d) F-BNBN-H monolayers calculated by shifting the redox potential of water. The blue lines indicate the shifted reduction potential of water, while the green lines represent the shifted oxidation potential of water.

On the other side, the CBM of F-BNBN-H monolayer is much higher than the shifted reduction potential of water for the top (001) surface, and the VBM is much lower than the shifted reduction potential of water for the bottom $(00\bar{1})$ surface. Hence, the F-BNBN-H monolayer has good

photocatalytic ability for water splitting. These analyses are well agreement with that in the main manuscript.

Reference

[1] C.F. Fu, J. Sun, Q. Luo, X. Li, W. Hu, J. Yang, Intrinsic Electric Fields in Two-dimensional Materials Boost the Solar-to-Hydrogen Efficiency for Photocatalytic Water Splitting, **Nano Lett.** 18 (**2018**) 6312.

[2] G. Wang, W. Tang, W. Xie, Q. Tang, Y. Wang, H. Guo, P. Gao, S. Dang, J. Chang, Type-II CdS/PtSSe heterostructures used as highly efficient water-splitting photocatalysts, **Appl. Surf. Sci.** 589 (**2022**) 152931.