Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Prediction of extremely long exciton lifetime in Janus-MoSTe monolayer

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The Janus-MoSTe can be produced by well-controlled sulfurization of monolayer $MoTe_2$. Firstly, $MoTe_2$ monolayer is grown by the chemical vapor deposition (CVD) method. The $MoTe_2$ monolayer is then put on the SiO_2/Si substrate and heated in the center of the heating zone. Similar with the production process of Janus-MoSSe, when the gaseous sulfur molecules approach, the top layer Te atoms directly contact with S and can be quickly substituted while the bottom Te atoms remaining intact.[1,2] By such controlled substitution reaction with vaporized sulfur in a typical CVD setup, Janus-MoSTe monolayer shall be synthesized.

The stability of the proposed Janus-MoSTe monolayer can be tested by calculating their vibrational spectra. Dynamically stable structures are heralded when calculated dispersions of phonon modes have positive square of frequency throughout the Brillouin zone. In Fig. S1, we present the dispersion of phonon modes of Janus-MoSTe monolayer. Clearly, there are no imaginary frequencies in the phonon dispersion curves indicating that the considered structures are stable.

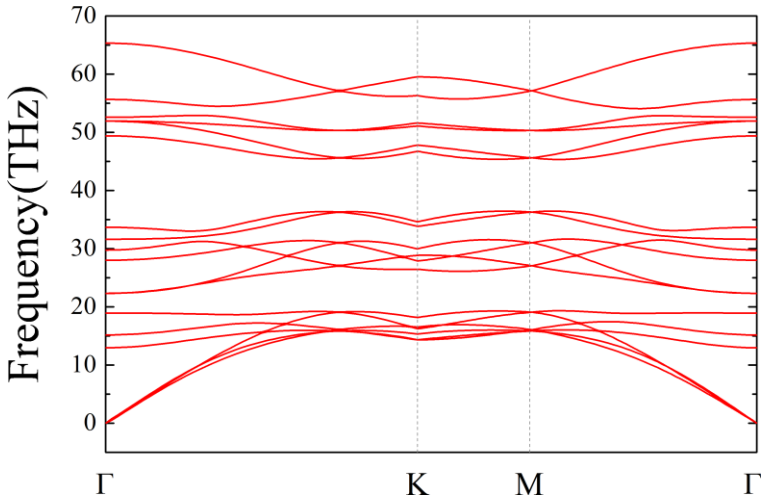

Figure S1: Phonon dispersions of Janus-MoSTe monolayer.

In Fig. S2, we plot the evolution of free energy for the Janus-MoSTe monolayer during *ab initio* molecular dymanics (AIMD) simulation, which are performed in the canonical ensemble using a standard Nosé thermostat. The system is heated to 300 K for 5 ps with a 1 fs time step, which clearly shows that the variation in free energy of the system is considerably small. These results demonstrate that the studied Janus-MoSTe monolayer is stable even at temperatures up to

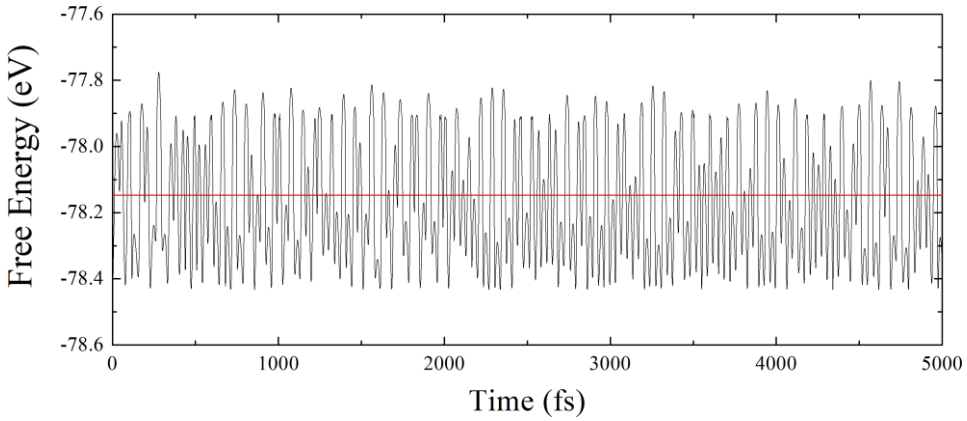

Figure S2: Variation of the free energy during molecular dynamics simulation at 300 K.

300 K.

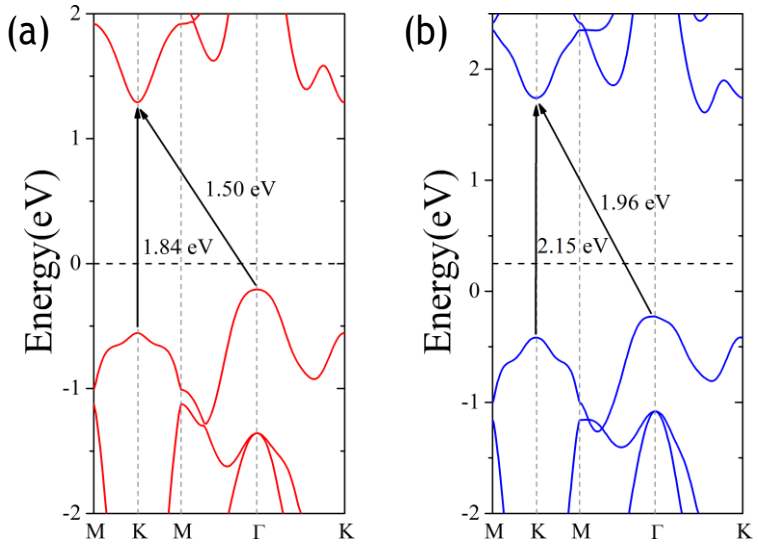

Figure S3: Band structures of Janus-MoSTe monolayer at (a) HSE06 level, and (b) G_0W_0 level. The Fermi level is set to be zero.

To obtain more accurate band structure, we apply the HSE06 method. The band structure of Janus-MoSTe monolayer with HSE06 method is depicted in Fig. S3(a). Obviously, the bandgap at the HSE06 level (1.84 eV) is larger than that from PBE (1.40 eV) at K point. In comparison with previous results at PBE and HSE06 levels, the band structure at G_0W_0 level is shown in Fig. S3(b). A larger bandgap up to 2.15 eV is observed. However, despite the bandgaps, the dispersions of the

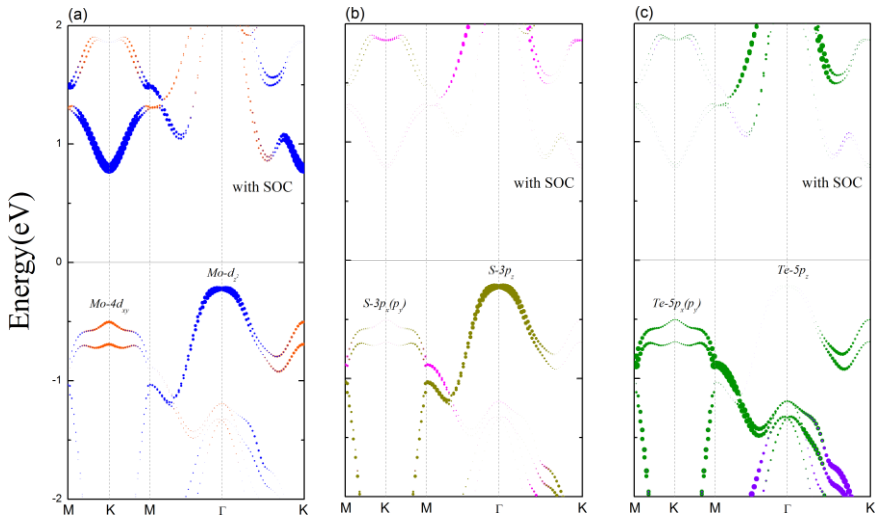

Figure S4: Orbit-projected band structures of Janus-MoSTe monolayer with spin orbit coupling (SOC). The Fermi level is set to be zero. A large valence band splitting can be observed at K-point, with the value up to 188 meV.

band structures with three methods exhibit the similar tendency in both VBM and CBM.

We also calculate the band structures of Janus-MoSTe monolayer with spin orbit coupling (SOC) at PBE level. In analogy with the valence band splitting at K point of the MoS_2, a large valence band splitting up to 188 meV is observed in Janus-MoSTe monolayer. The corresponding orbit-projected band structures of Janus-MoSTe monolayer is shown in Fig. S4.

In Fig. S5, we plot the band structure of Janus-MoSTe monolayer with SOC at G_0W_0 level. The results indicate that the valence band splitting is 185 meV, which agrees well with our PBE results, i.e. 188 meV.

It should be indicated that TD-DFT usually refers to time-dependent DFT, which is formally established based on the Runge-Gross theorem.[3] However, the state-of-the-art TDDFT calculations scales as $O(N^3)$ which makes TDDFT a relatively expensive numerical method. Therefore, several methods have been developed to deal with larger and more complex systems. In this work, we employ Prezhdo's method, in which the classical path approximation (CPA), fewest-switches surface hopping (FSSH), decoherence-induced surface hopping (DISH), and other advanced functionalities are used. In their paper, they defined this approach as the time-domain DFT. A detailed

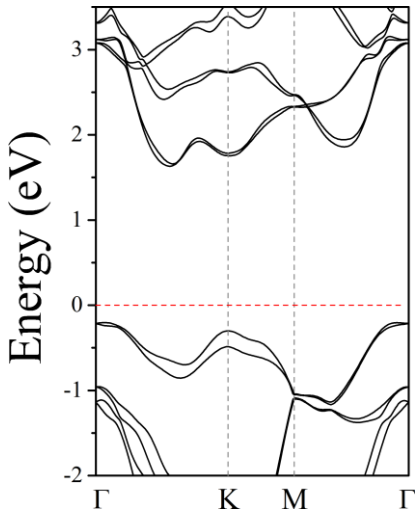

Figure S5: Band structure of Janus-MoSTe monolayer at G_0W_0 level with SOC. The Fermi level is set to be zero.

description of this method can be found in the original paper.[4]

References

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