Supporting Information for

Interfacial Electronic States and Self-Formed *p-n* Junctions in Hydrogenated MoS_2/SiC Heterostructure

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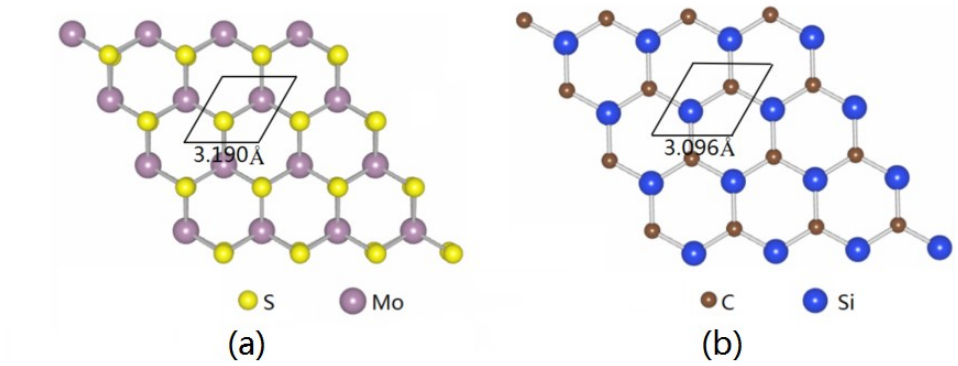

Fig. S1. Optimized geometries for 4×4×1 supercell of monolayers (a) MoS_2 and (b) SiC.

Table S1 Bond lengths of Si-C (L_{Si-C}), C-H (L_{C-H}), and Si-H (L_{Si-H}), and buckling height h (Å) of SiC sheet with or without hydrogenation.

System	L_{Mo-S} (Å)	L_{Si-C} (Å)	L_{Si-H} (Å)	L_{C-H} (Å)	h (Å)
SiC		1.787			
H-SiC		1.858	1.524		0.379
SiC-H		1.878		0.903	0.473
H-SiC-H		1.891	1.108	1.496	0.574

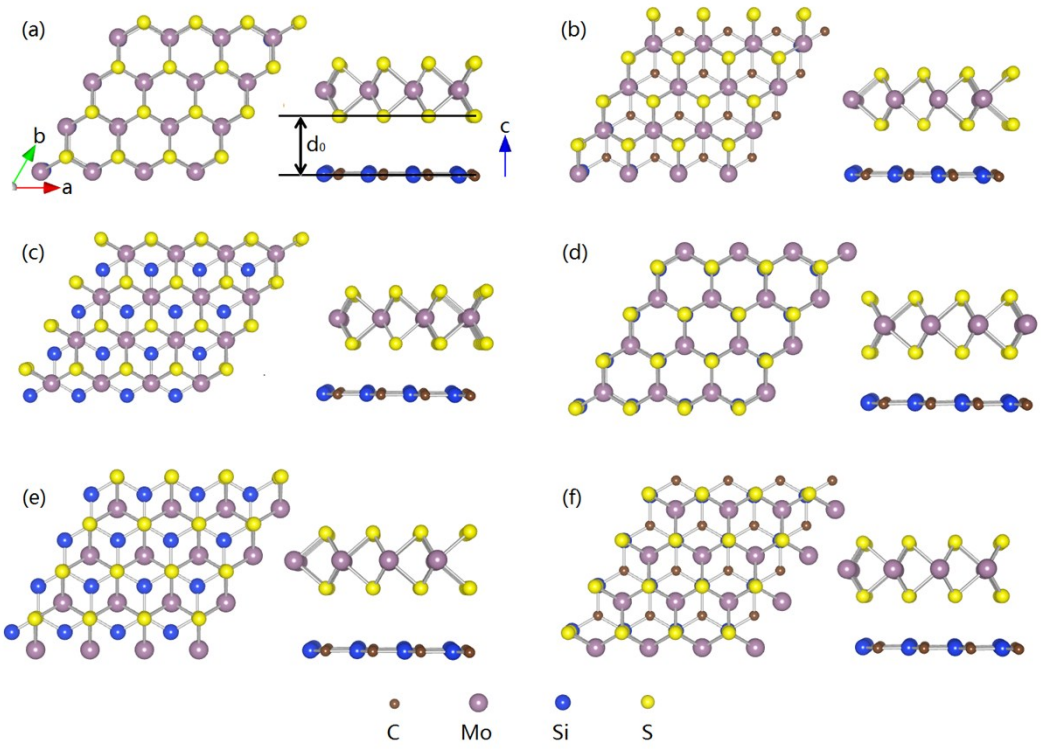

Fig. S2. Top and side views of the MoS_2/SiC heterostructure with different stacking configurations: (a) Mo and S atoms over Si and C atoms, respectively; (b) Mo and S atoms over Si atoms and hollow sites, respectively; (c) Mo and S atoms over C atoms and hollow sites, respectively; (d) Mo and S atoms over C and Si atoms, respectively; (f) Mo and S atoms over hollow sites and Si atoms, respectively; (f) Mo and S atoms over hollow sites and C atoms, respectively.

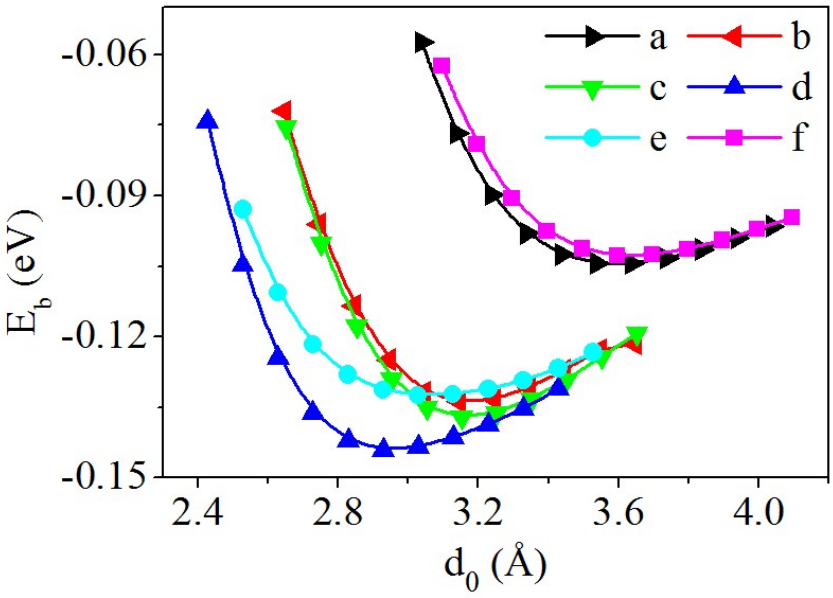

Fig. S3. Binding energy per S atom of MoS_2/SiC as a function of the interlayer distance (d_0) between the topmost S atom of MoS_2 and SiC in the MoS_2/SiC heterostructure.

To evaluate the interfacial interaction, the binding energy between monolayer MoS_2 and the SiC substrate is calculated as $E_b=(E_{MoS_2/SiC}-E_{MoS_2}-E_{SiC})/N_S$, in which $E_{MoS_2/SiC}$, E_{MoS_2}, and E_{SiC} are energies of the MoS_2/SiC heterostructure, isolated monolayer MoS_2, and monolayer SiC, respectively, N_S is the number of S atoms in the supercell of MoS_2/SiC heterostructure. If E_b is negative, the configuration should be stable, and the larger the absolute value is, the stronger is the interaction between monolayer MoS_2 and SiC. As shown in Fig. S3, all the six stacking configurations have the negative E_b value, indicating that the MoS_2/SiC heterostructure is energetically favorable. Although the absolute value of binding energy of the MoS_2/SiC heterostructure is slightly larger than that of monolayer MoS_2 on silicene (-64 meV per S atom) and on germanene (-71 meV per S atom),[1] MoS_2 is bonded onto SiC via van der Waals interaction. The interlayer distance (d_0) changes in the range of 2.928-3.637 Å, characteristic of physical adsorption between monolayer MoS_2 and SiC, and the in–plane structure changes little.

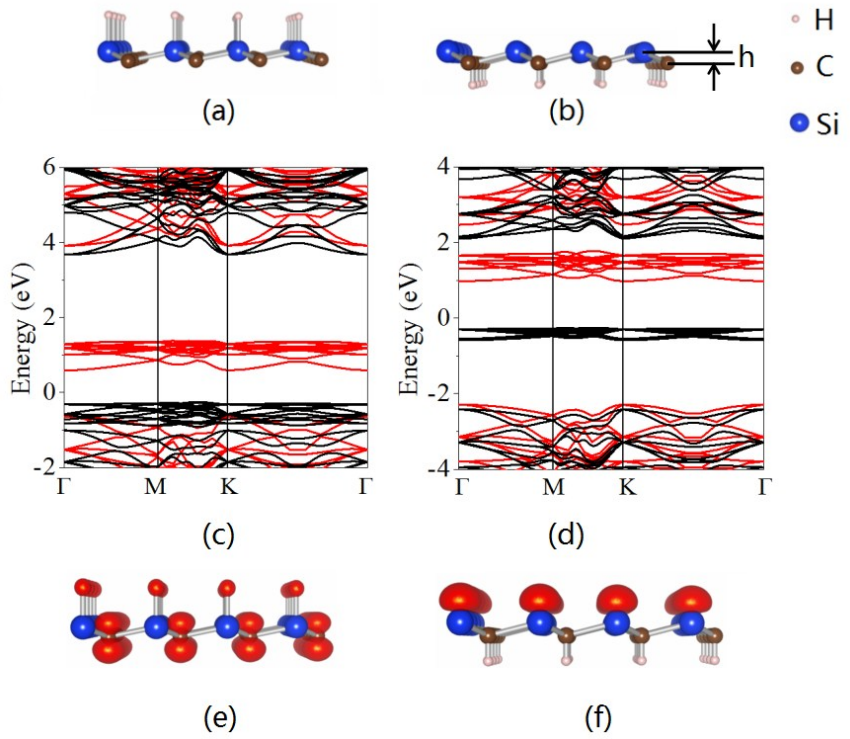

Fig. S4. Optimized structures of partially hydrogenated SiC sheet: (a) H-SiC and (b) SiC-H, and (c) and (d) the band structures, (e) and (f) the magnetic densities. Black and red lines are used to indicate the spin-up and spin-down channels, respectively. The Fermi level is set at 0 eV. The red surface corresponds to the magnetic density isosurface of 0.02 eÅ^{-3}.

SiC is partially hydrogenated on Si sites (labeled as H-SiC) or C sites (labeled as SiC-H), the optimized structures and structural parameters are given in Fig. S4 and Table S1, respectively. The lattice constants of H-SiC and SiC-H are a=b=3.151 Å and a=b=3.147 Å, respectively, that is, they are enlarged upon hydrogenation. Moreover, the hybridization states are changed from sp^2 to sp^3. The optimized Si-H/C-H bond length in H-SiC/ SiC-H is 1.524 Å/0.903 Å, and the buckling heights on H-SiC and SiC-H are 0.379 Å and 0.473 Å, respectively.[2] The partially hydrogenated SiC is still a semiconductor, and spin polarization emerges (Fig. S4c,d). In essential, hydrogenation on Si (C) atoms produces Si-H (C-H) bonds and the dangling bonds of C (Si) atoms on surface are spin unpaired. The spin polarization is localized on Si and H in SiC-H and H-SiC, respectively (Fig. S4e,f).

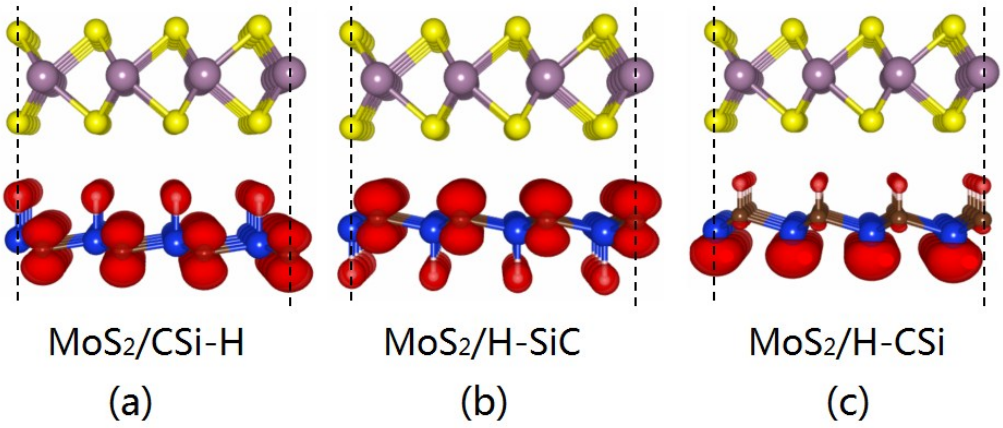

Fig. S5. Magnetization density of MoS_2 on the half-hydrogenated SiC. The red surface corresponds to the spin density isosurface of 0.01 /Å^3.

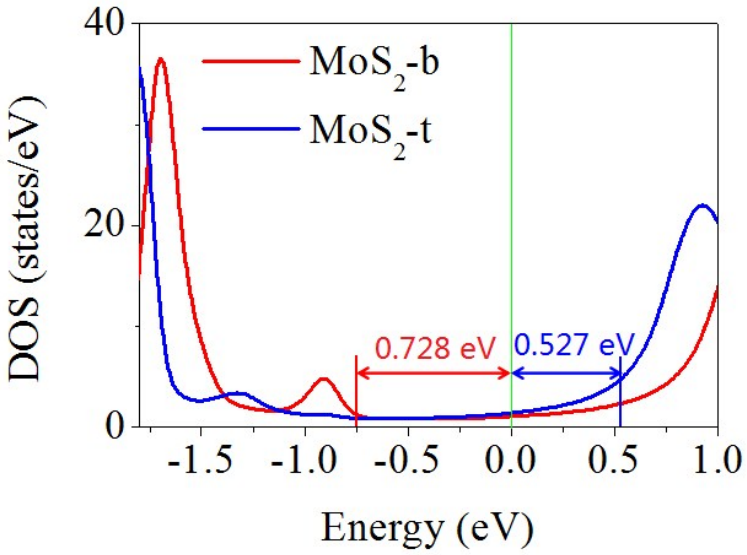

Fig. S6. The density of states of the MoS_2-b and MoS_2-t in MoS_2-H-SiC-MoS_2-based device with Au as the electrode.

References

1 M. Brandbyge, J. -L. Mozos, P. Ordejón, J. Taylor and K. Stokbro, *Phys. Rev. B: Condens. Matter Mater. Phys*., 2002, **65**, 165401.

2 Y. Ma, L. Kou, A. Du and T. Heine, *Nano Res*., 2015, **8**, 3412-3420.