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Two-dimensional $MoSe_2/MoSi_2N_4$ van der Waals heterostructure with high carrier mobility and diversified regulation on electronic properties

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1. Band structures of the units for the monolayer $MoSe_2$ and $MoSi_2N_4$ together with the $MoSe_2/MoSi_2N_4$ heterobilayer

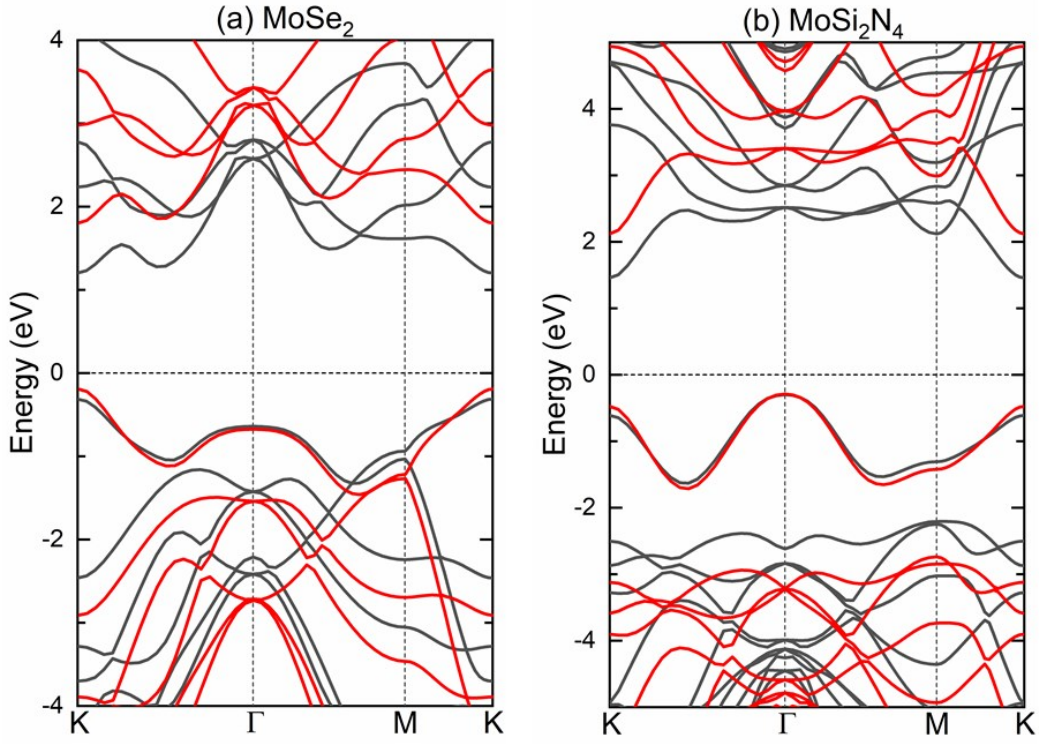

Figure S1. Band structures of the units for the monolayer (a) $MoSe_2$ and (b) $MoSi_2N_4$, where the black and red lines represent the results obtained from the PBE and HSE06 schemes, respectively. Here, the Fermi levels are set to zero.

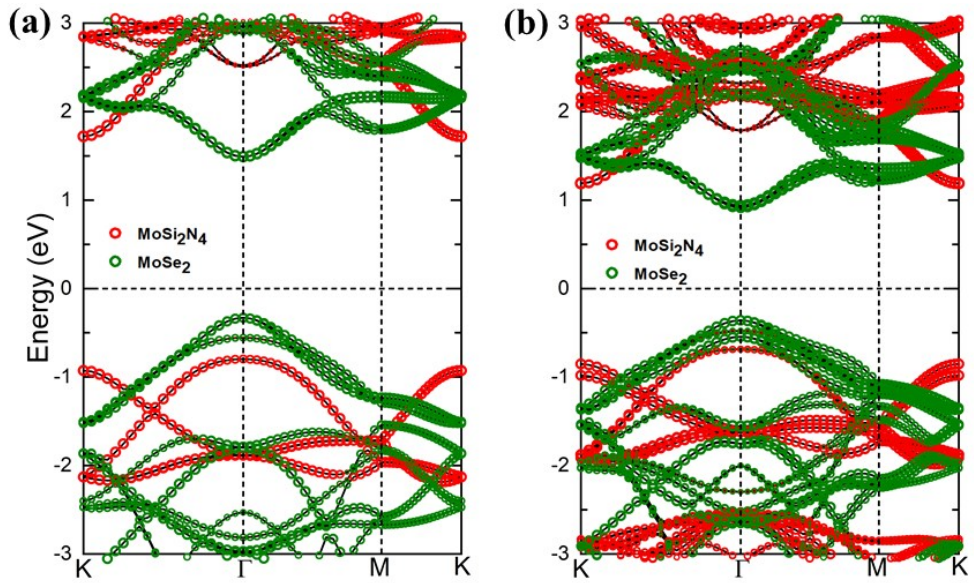

Figure S2. Projected band structures of the $MoSe_2/MoSi_2N_4$ vdWH by the (a) HSE and PBE+SOC functionals, respectively. Here, the Fermi levels are set to zero.

2. Computational details of the carrier mobility of the MoSe2/MoSi2N4 heterobilayer

Using the acoustic phonon limited method, we quantitatively estimate the room-temperature carrier mobility for $MoSe_2$ monolayer and $MoSe_2/MoSi_2N_4$ heterobilayer. The carrier mobility of 2D material is given by $\mu_{2D} = \frac{e\hbar^3 C_{2D}}{k_B T m^* m_d (E_1^i)^2}$, where m^* is the effective mass in the transport direction and m_d is the average effective mass determined by $m_d = \sqrt{m_x^* m_y^*}$. The term E_1 represents the deformation potential constant of the valence-band minimum (VBM) for hole or the conduction-band maximum (CBM) for electron along the transport direction, defined by $E_1^i = \Delta V_i / (\Delta l / l_0)$. Here, ΔV_i denotes the energy change of VBM or CBM when tellurene is compressed or dilated from the equilibrium l_0 by a distance of Δl. The term C_{2D} is the elastic modulus of the longitudinal strain in the propagation directions (*armchair* or *zigzag*, shown in Figure S2) of the longitudinal acoustic wave, which can be derived from $(E - E_0) / S_0 = C(\Delta l / l_0)^2 / 2$: E and S_0 denote the total energy and lattice area of tellurene, respectively. We used $\Delta l / l_0$ ranging from -1.0% to 1.0% to fit the values of C_{2D} and E_1^i for $MoSe_2$ monolayer and $MoSe_2/MoSi_2N_4$ heterobilayer (Figure S3 and Figure S4), respectively.

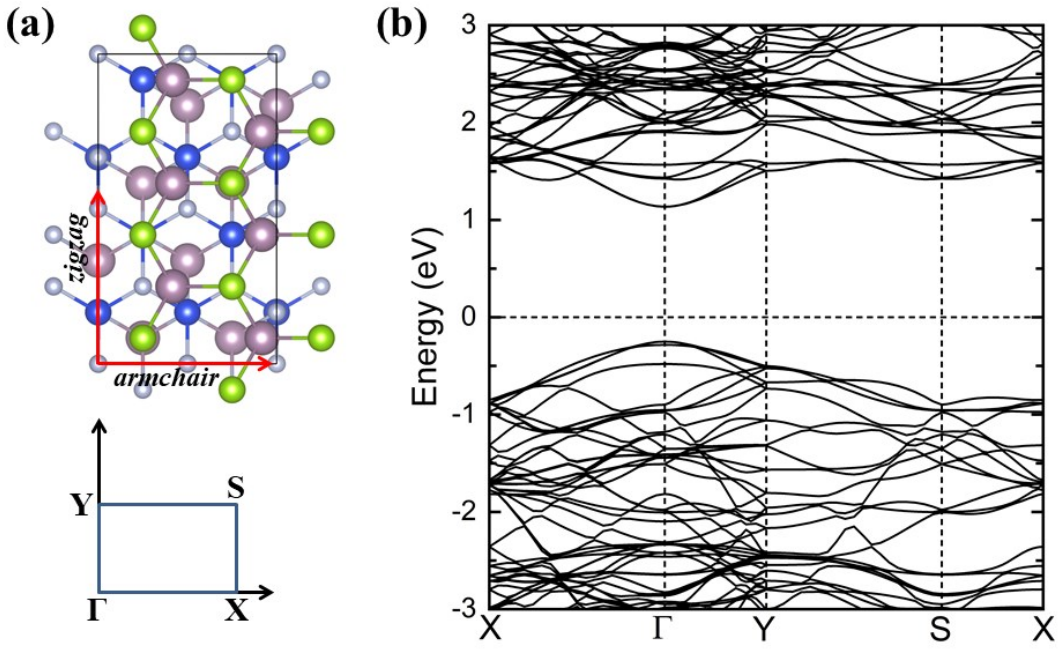

Figure S3. (a) Crystal structure together with the corresponding Brillouin zones and (b) band structure of the rectangular unit cell for the $MoSe_2/MoSi_2N_4$ vdWH. In panel (b), the Fermi level is set to zero.

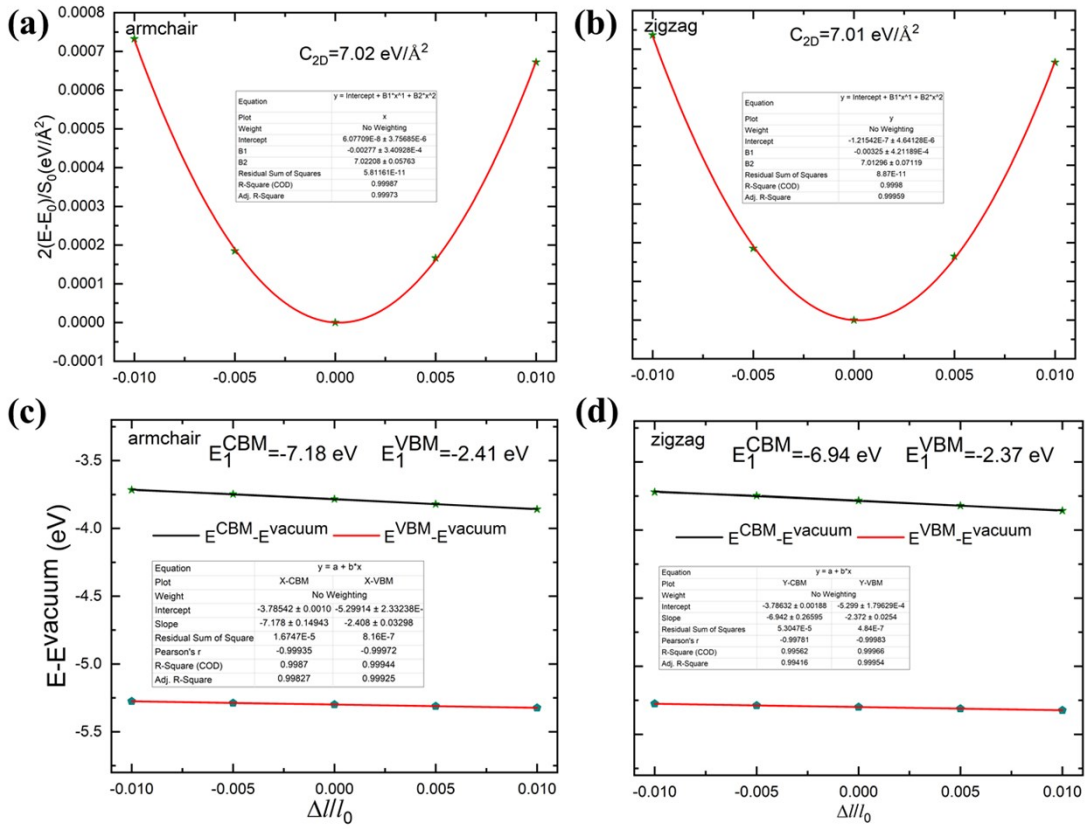

Figure S4. In-plane stiffness and deformation potential of the $MoSe_2$ monolayer obtained from the PBE scheme. (a) and (b) are the elastic moduli along the armchair and zigzag directions, respectively. And (c) and (d) are the corresponding deformation potentials for the CBM and VBM.

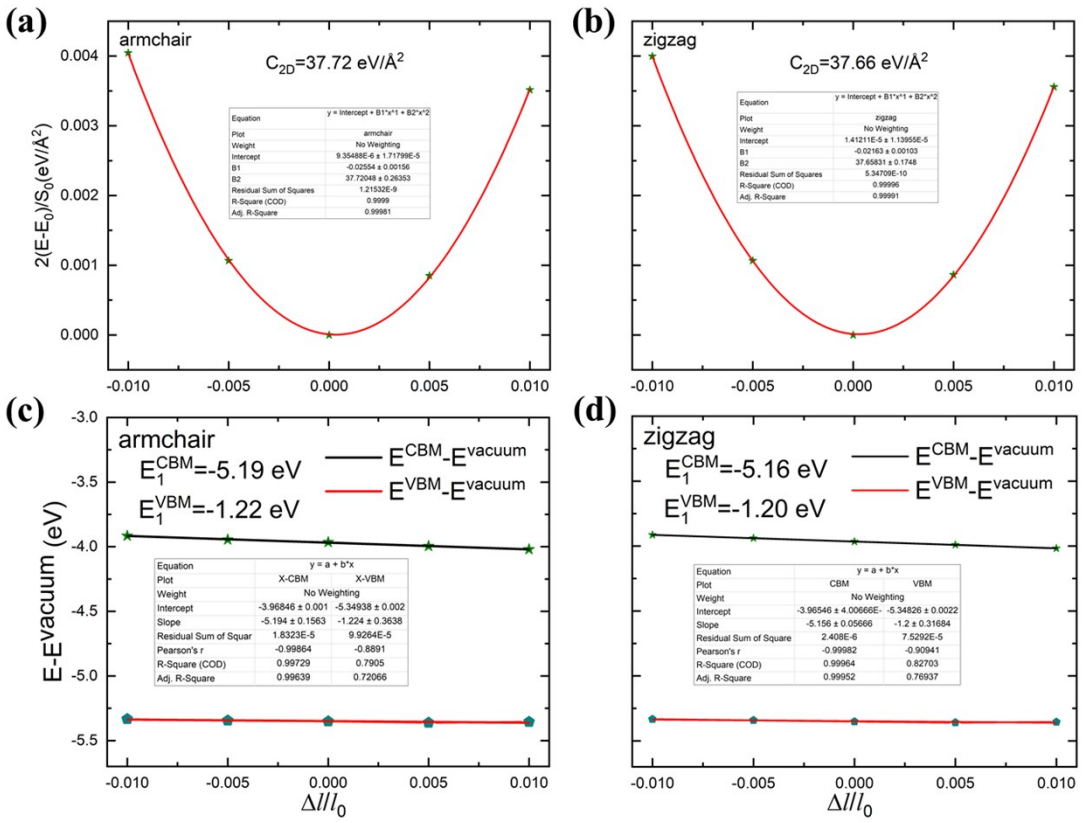

Figure S5. In-plane stifness and deformation potential of the $MoSe_2/MoSi_2N_4$ vdwH obtained from the PBE scheme. (a) and (b) are the elastic moduli along the armchair and zigzag directions, respectively. And (c) and (d) are the corresponding deformation potentials for the CBM and VBM.

3. Strain effect on the $MoSe_2/MoSi_2N_4$ vdWH

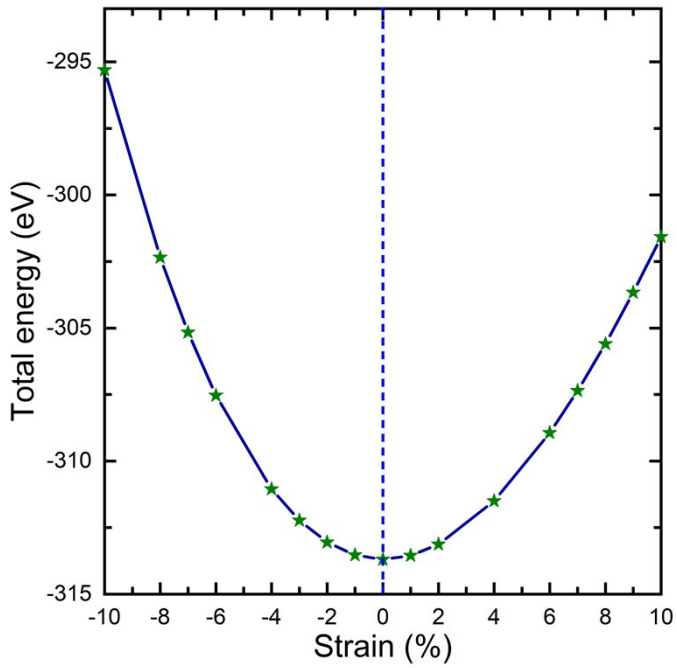

Figure S6. Total energy of the $MoSe_2/MoSi_2N_4$ vdWH as a function of the applied biaxial strain.

Using Bader charge analyses, it is found that Se atoms gain 2.6173 *e* while Mo atoms lose 2.6173 *e* in the isolated $MoSe_2$ monolayer. Further, we calculate the charge transfer of Se and Mo atoms of the component $MoSe_2$ layer in $MoSe_2/MoSi_2N_4$ vdWH under the strain ranging from -10% to +9%, respectively, which relative to those in the isolated $MoSe_2$ monolayer ($^{\Delta Q_{Se}}$ (ΔQ_{Mo})) are shown in Figure S7 (a). Obviously, compared with the isolated $MoSe_2$ monolayer, the charge gain of Se atoms is increased from -0.0601 to 0.3619 *e* in $MoSe_2$ layer of $MoSe_2/MoSi_2N_4$ vdWH under the strain from -3% to +9%, while it is decreased from -0.0129 to -0.3040 *e* under the strain from -4% to -10%. Likewise, the charge loss of Mo atoms has the similar trend with that of Se atoms. A similar case also exists in the component $MoSi_2N_2$ monolayer. Such charge transfer make significant changes in the electronic property of the component $MoSe_2$ and $MoSi_2N_4$ layers, which including the movement of the band edges. Figure S7 (b) shows the variation of ΔQ_{Se} and $-\Delta Q_{Mo}$ with applying strain, whose difference is actually the charge transfer between the component $MoSe_2$ and $MoSi_2N_4$ layers, equivalent to Fig. 6 (a). Evidently, the interlayer charge transfer varies with applying strain, which can lead to the change in the band structure of the vdWH. Therefore, strain induced changes in the electronic property of $MoSe_2/MoSi_2N_4$ vdWH is jointly determined by the changes in the intralayer interactions of the component layers and the interlayer interaction between the component layers. Judging from the magnitude of charge transfer (Figure S7 (b)), intralayer interaction has a greater effect on the electronic property of the $MoSe_2/MoSi_2N_4$ vdWH.

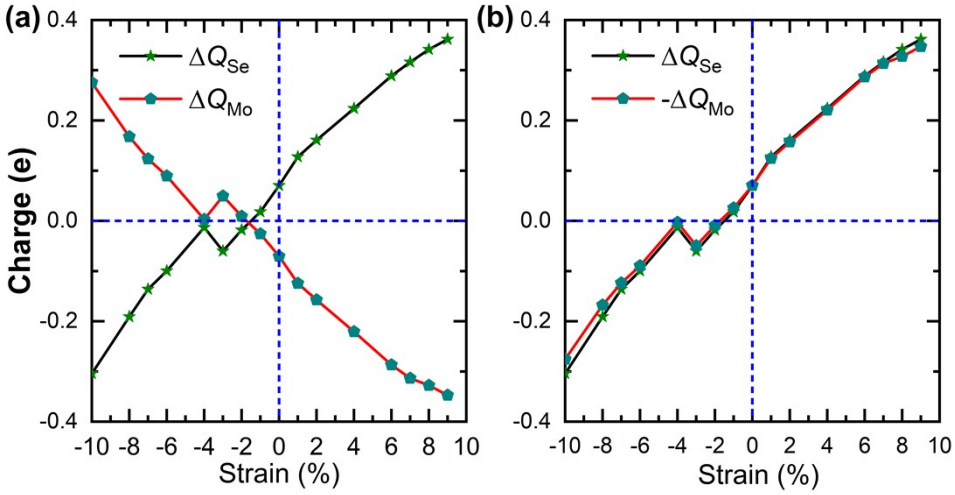

Figure S7. (a) Charge gain and loss of Se and Mo atoms in the component $MoSe_2$ monolayer of the $MoSe_2/MoSi_2N_4$ vdWH under strain, respectively, which are relative to those in the

isolated $MoSe_2$ monolayer. (b) Comparation between ΔQ_{Se} and $-\Delta Q_{Mo}$.