Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Tunable Structural Phases and Electronic Properties of Group V MSi_2N_4 (M=V, Nb, Ta) nanosheets via Surface Hydrogenation: A First-Principles Study

Yanli Wang[1, a)] and Yi Ding[2, b)]

[1)] *Department of Physics, Zhejiang Sci-Tech University, Hangzhou, Zhejiang 310018, People's Republic of China*

[2)] *School of Physics, Hangzhou Normal University, Hangzhou, Zhejiang 311121, People's Republic of China*

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[a)] Y. Wang: wangyanli-04@tsinghua.org.cn

[b)] Y. Ding: dingyi2001@tsinghua.org.cn

Figure S1: The AIMD simulations of $NbSi_2N_4H_1$ nanosheets at 300 K.

Figure S2: The COHP analysis of $NbSi_2N_4H_1$ nanosheets.

Figures S3 and S4: The atomic structures of $NbSi_2N_4H_x$ systems.

Figure S5: The strain and electric field effects on the energy differences.

Figure S6: The charge injection influence on the energy differences.

Figure S7: The AIMD simulation of pristine $NbSi_2N_4$ systems.

Figure S8: The band structures of $NbSi_2N_4H_x$ ($x \neq 0, 1, 2$) nanosheets.

Figure S9: The stability of $NbSi_2N_4H_1$ system under the electric fields.

Figure S10: The band structures of $NbSi_2N_4H_x$ ($x = 0, 2$) under the electric fields.

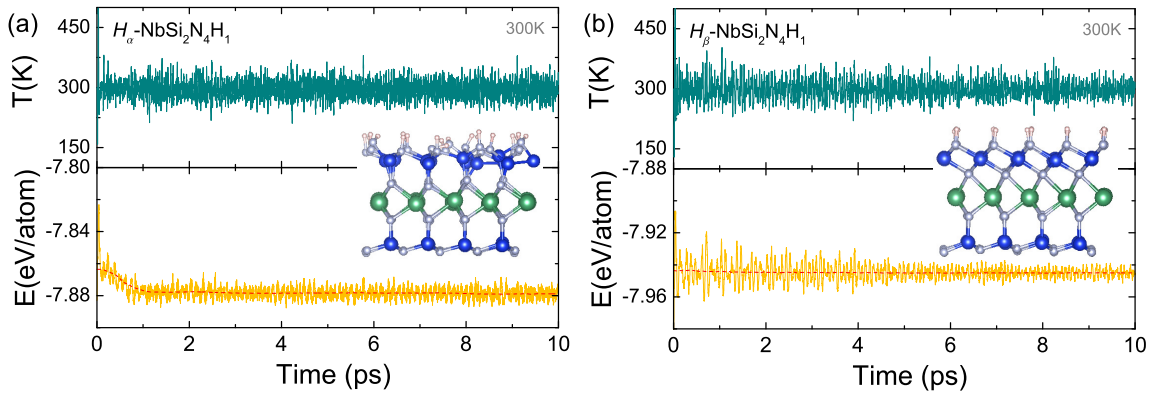

Figure S1: The AIMD simulation results of (a) H_α-$NbSi_2N_4H_1$ and (b) H_β-$NbSi_2N_4H_1$ nanosheets under a temperature of 300 K.

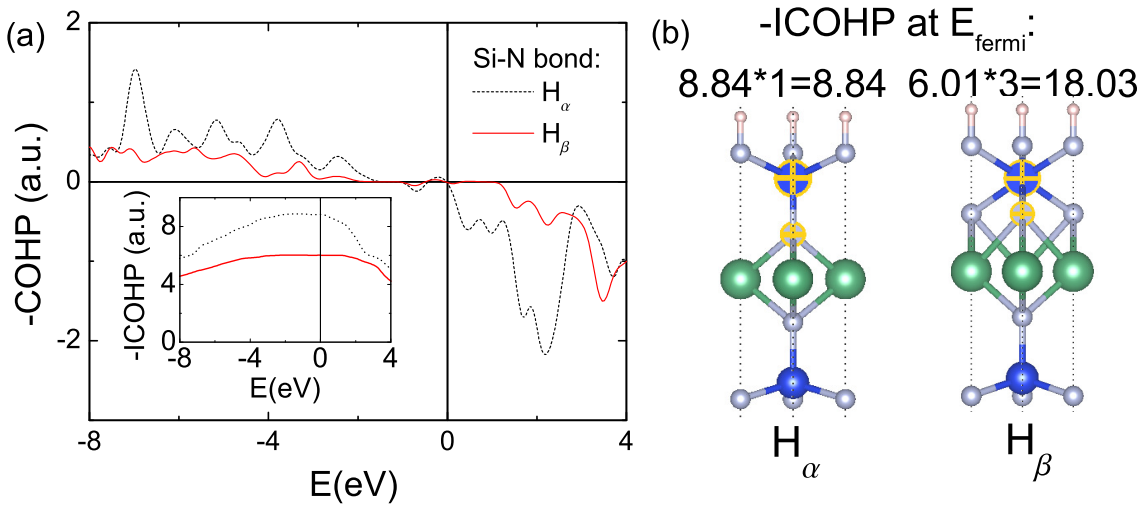

Figure S2: (a) The -COHP functions of Si-N_i bonds that connect the hydrogenated SiN surface layer and the inner NbN_2 part. The integral of -COHP (-ICOHP) is also depicted in the inset. (b) The lateral view of H_α- and H_β-$NbSi_2N_4H_1$ nanosheets, where the Si-N_i bonds are marked. It can be seen that there is one and three Si-N_i bonds per formula unit in the H_α- and H_β-phase structures, respectively. The corresponding -ICOHP values at the Fermi level per formula unit are also noted.

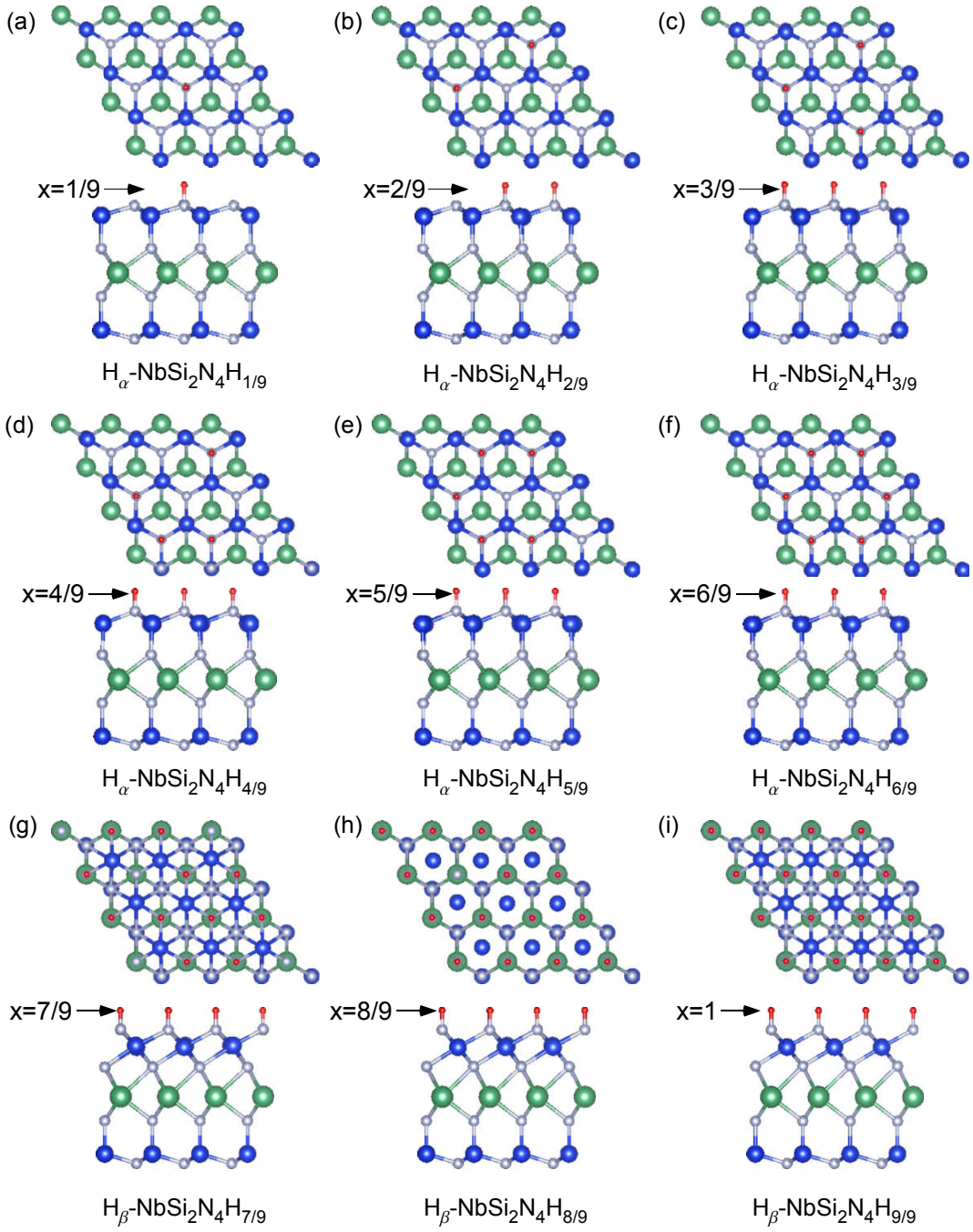

Figure S3: The geometrical structures of $NbSi_2N_4H_x$ nanosheets with x =1/9∼1. The Nb, Si, N and H atoms are represented by green, blue, grey and red balls, respectively.

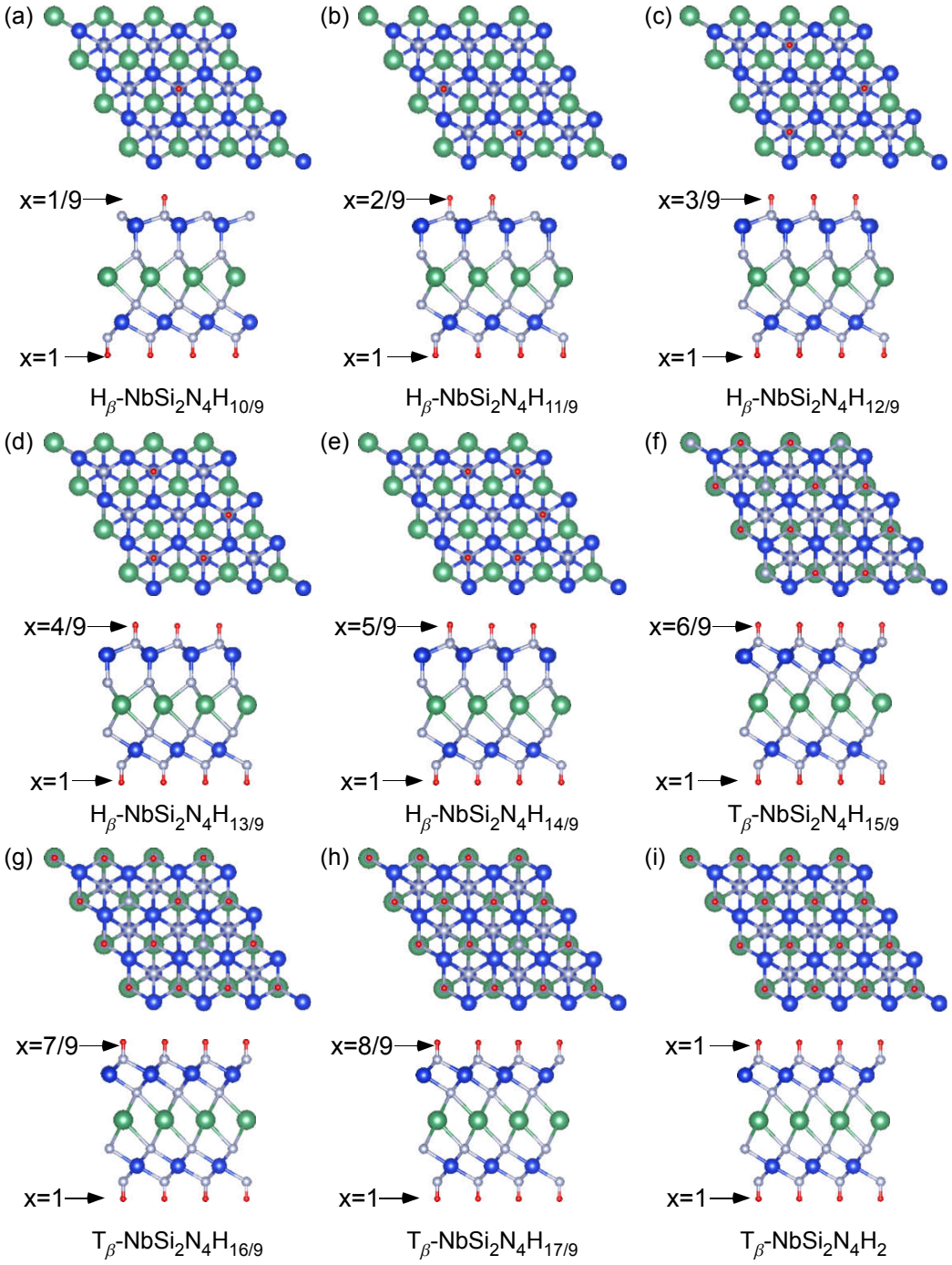

Figure S4: The geometrical structures of $NbSi_2N_4H_x$ nanosheets with x =10/9∼2. The Nb, Si, N and H atoms are represented by green, blue, grey and red balls, respectively.

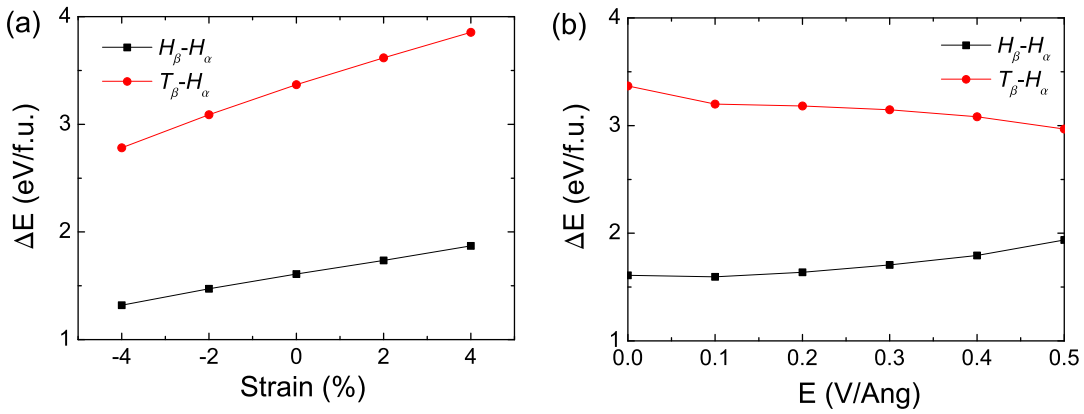

Figure S5: The variations of energy differences between the H_β/T_β- and H_α-$NbSi_2N_4$ nanosheets under the (a) strain and (b) electric field modulations. It can be seen that the H_α-phase geometry is always favored in these $NbSi_2N_4$ nanosheets.

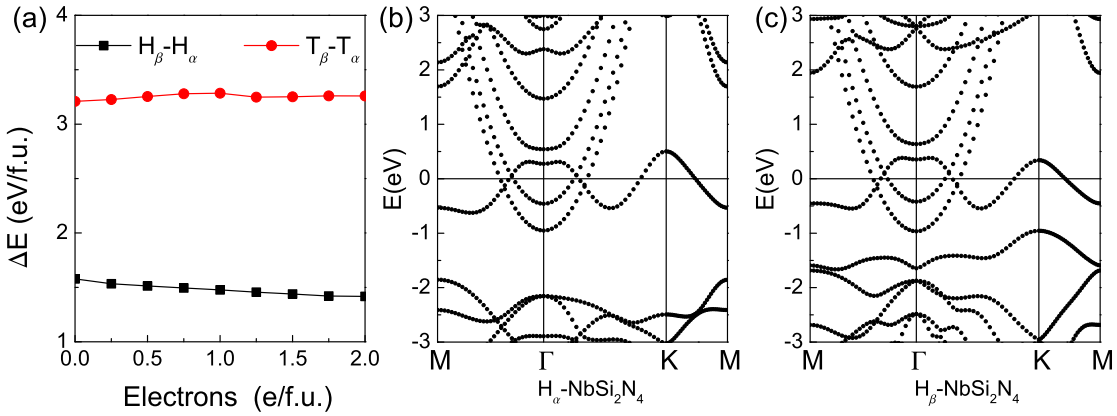

Figure S6: (a) The variations of energy differences between the H_β/T_β- and H_α-$NbSi_2N_4$ nanosheets with the different injected electron amount. The band structures of (b) doped H_α- and (b) H_β-$NbSi_2N_4$ nanosheets with injected electron amount of 1 e/f.u.. The H_α-phase geometry is preferred for the doped $NbSi_2N_4$ nanosheet and it exhibit different band structures from the semihydrogenated $NbSi_2N_4H_1$ one.

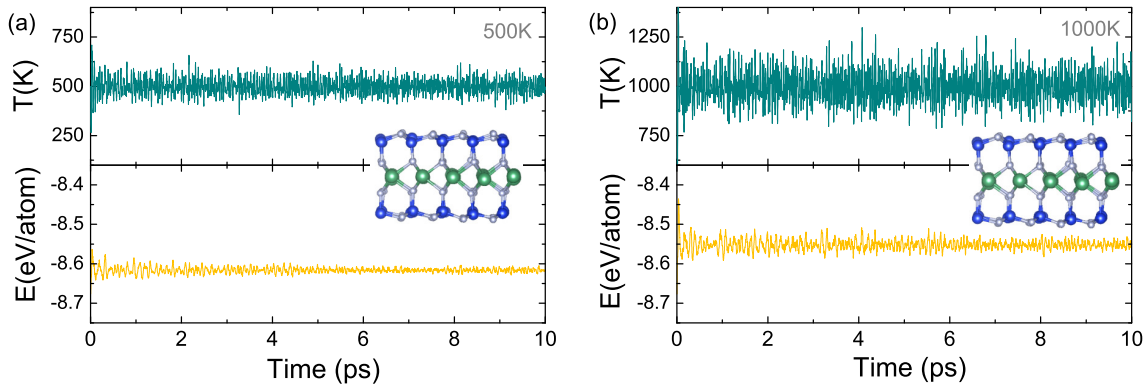

Figure S7: The AIMD simulation results for the pristine H-phase $NbSi_2N_4$ nanosheet under the temperatures of (a) 500 K and (b) 1000 K, respectively.

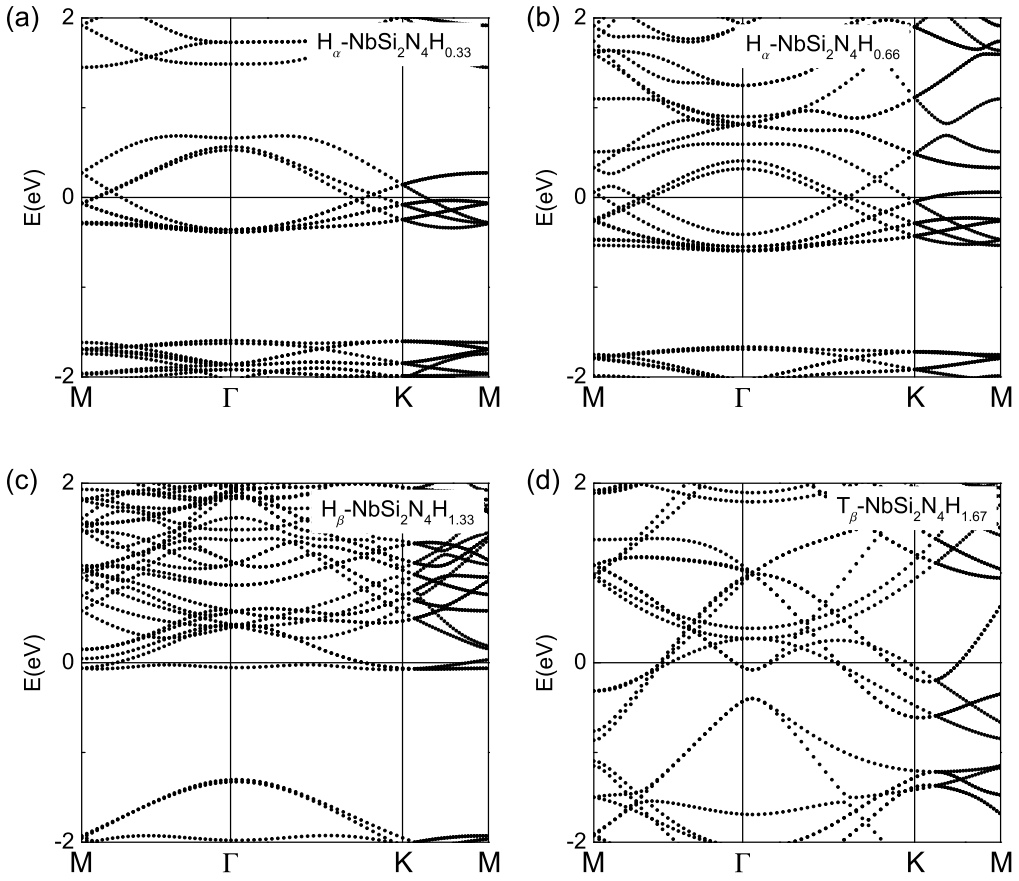

Figure S8: The band structures of (a) H_α-$NbSi_2N_4H_{0.33}$, (b) H_α-$NbSi_2N_4H_{0.67}$, (c) H_β-$NbSi_2N_4H_{1.33}$ and (d) T_β-$NbSi_2N_4H_{1.67}$ nanosheets.

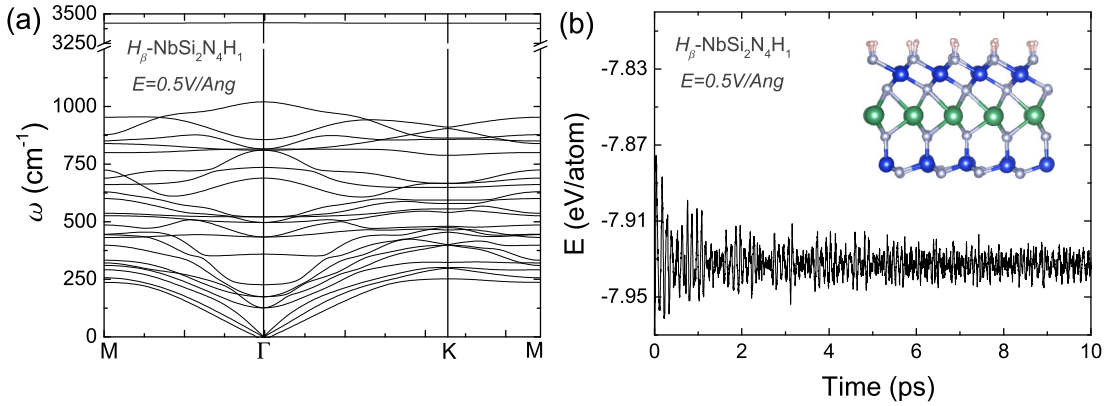

Figure S9: The phonon dispersions and AIMD simulation results of H_β-$NbSi_2N_4H_1$ nanosheet under the electric field of $E = 0.5$ V/Å.

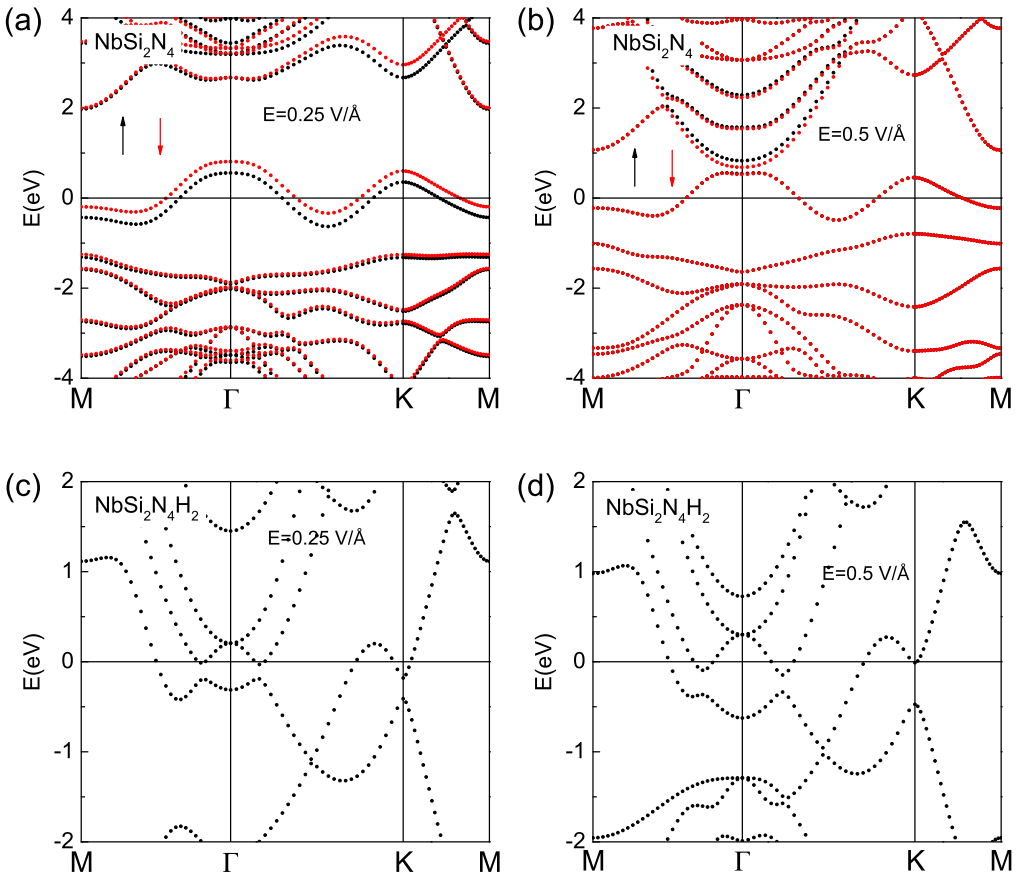

Figure S10: (a)-(b) The band structures of pristine H_{α}-$NbSi_2N_4$ nanosheet at the ferromagnetic state under the electric field of E= 0.25 and 0.5 V/Å. (c)-(d) The band structures of T_{β}-$NbSi_2N_4H_2$ nanosheet under the electric field of 0.25 and 0.5 V/Å.