Supplemental Material for Modulating the electronic properties of silicene allotropes through the synergistic effect of different geometric configurations and edge hydrogenation

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Here, we supplement the DFTB theory. The geometric structure (S1, S2), band structure and density of states (S3 and S4) of TO-SiNRs during the research process are provided. S1 is the structure diagram of intrinsic TO-SiNRs before and after relaxation. S2 is the structure diagram of TO-SiNRs before and after relaxation under edge hydrogenation. S3 is the band structure and density of states diagram of intrinsic TO-SiNRs; S4 is the band structure and density of states of TO-SiNRs after edge hydrogenation.

DFTB theory

The Density Functional Tight-Binding (DFTB) method is an efficient tight-binding approximation method based on density functional theory (DFT), which combines the quantum mechanical calculation's accuracy and the tight-binding method's computational efficiency. It is especially suitable for structural optimization and long-term dynamic simulation of large-scale systems. This method was first proposed by Elstner *et al.* [1] in 1998, and then the open-source software package DFTB+ was systematically developed by Professor Frauenheim's team, which greatly expanded its application in materials science [2], biomolecular simulation [3] and other fields. Theoretically, the DFTB method is derived from the second-order expansion of the Kohn-Sham (KS) [4-7] energy near the reference charge density. The core assumption is that the charge density at the centre of the atom can be approximated as a neutral spherical symmetry distribution, and the actual charge density of the system is regarded

as a perturbation of this reference density [8]. In the specific implementation, DFTB uses the minimum basis set to expand the Kohn-Sham eigenstate and constructs an effective polyatomic potential field by optimizing the neutral atom potential superposition and related charge density. All integrals are strictly calculated within the DFT theoretical framework. It is worth noting that the self-consistent processing of DFTB is unique: this method performs self-consistent iteration at the Mulliken charge population level [9] rather than the self-consistent cycle of electron density and effective potential in traditional DFT, which significantly improves the computational efficiency.

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Fig. S1.

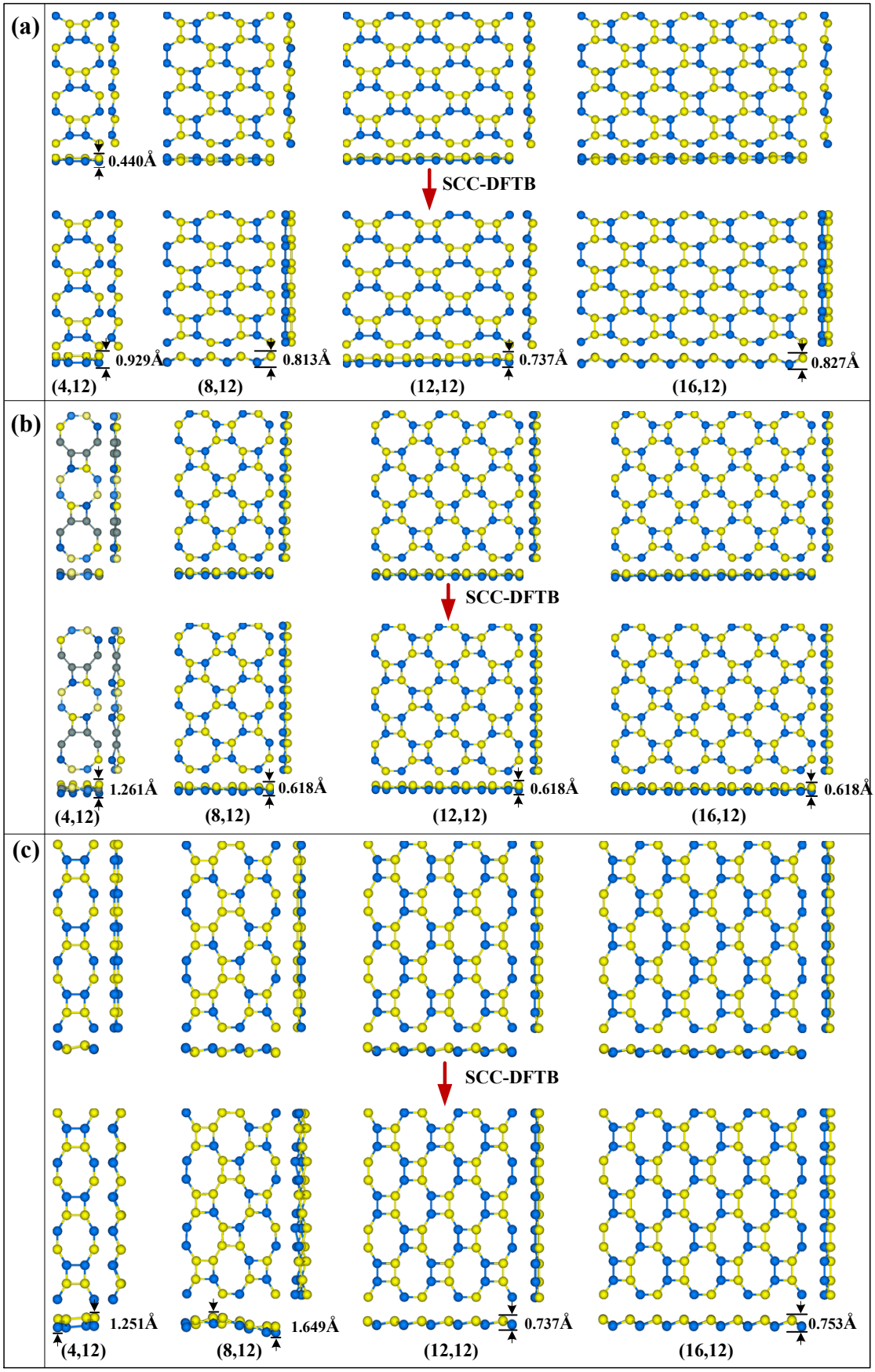
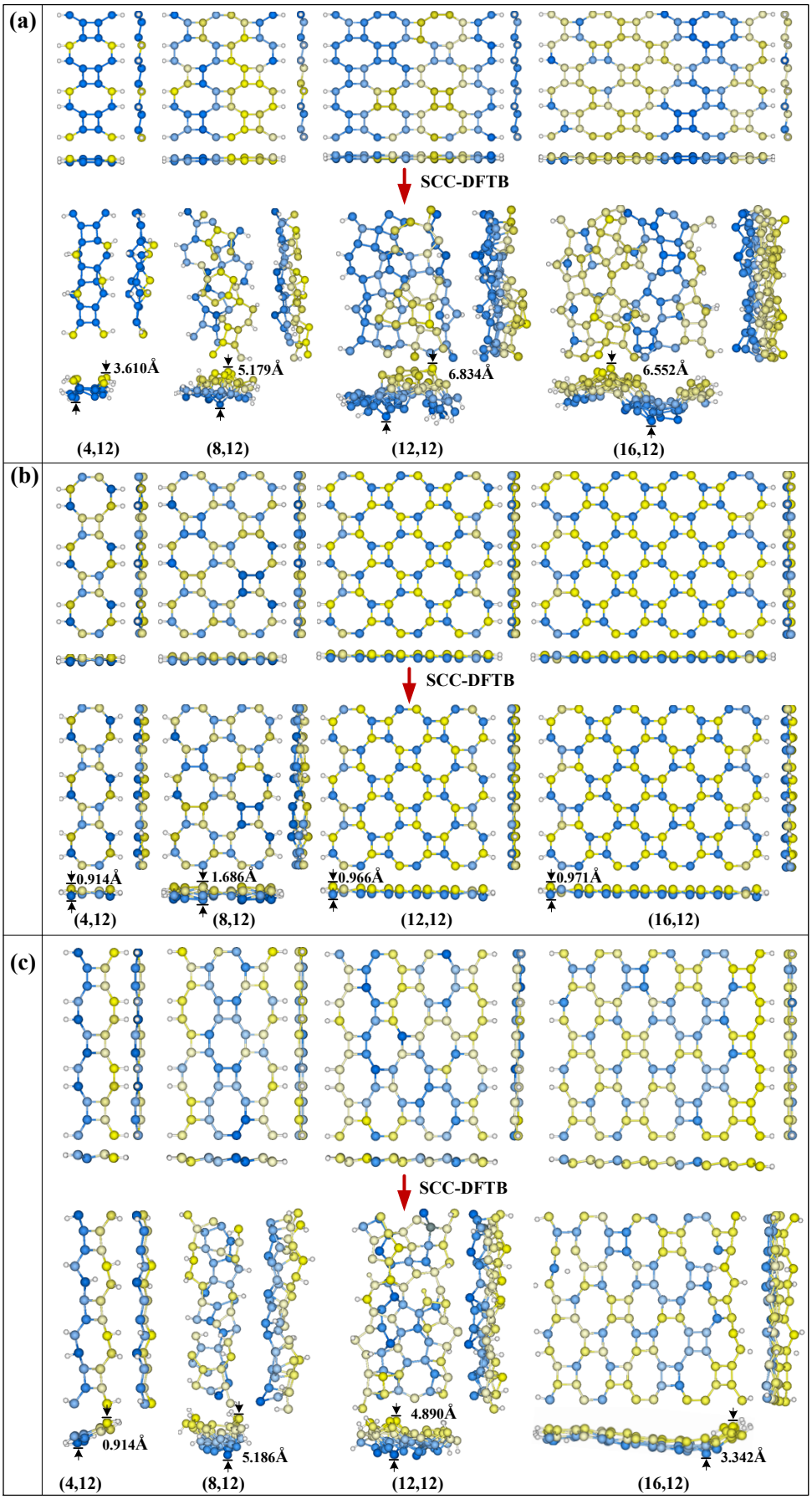

(a)
0.440Å
SCC-DFTB
0.929Å
0.813Å
0.737Å
0.827Å
(4,12)
(8,12)
(12,12)
(16,12)
(b)
SCC-DFTB
1.261Å
0.618Å
0.618Å
0.618Å
(4,12)
(8,12)
(12,12)
(16,12)
(c)
SCC-DFTB
1.251Å
1.649Å
0.737Å
0.753Å
(4,12)
(8,12)
(12,12)
(16,12)

Fig. S2.


(a)
SCC-DFTB
3.610Å
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6.834Å
6.552Å
(4,12)
(8,12)
(12,12)
(16,12)
(b)
SCC-DFTB
0.914Å
1.686Å
0.966Å
0.971Å
(4,12)
(8,12)
(12,12)
(16,12)
(c)
SCC-DFTB
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4.890Å
3.342Å
(4,12)
(8,12)
(12,12)
(16,12)

Fig. S3.

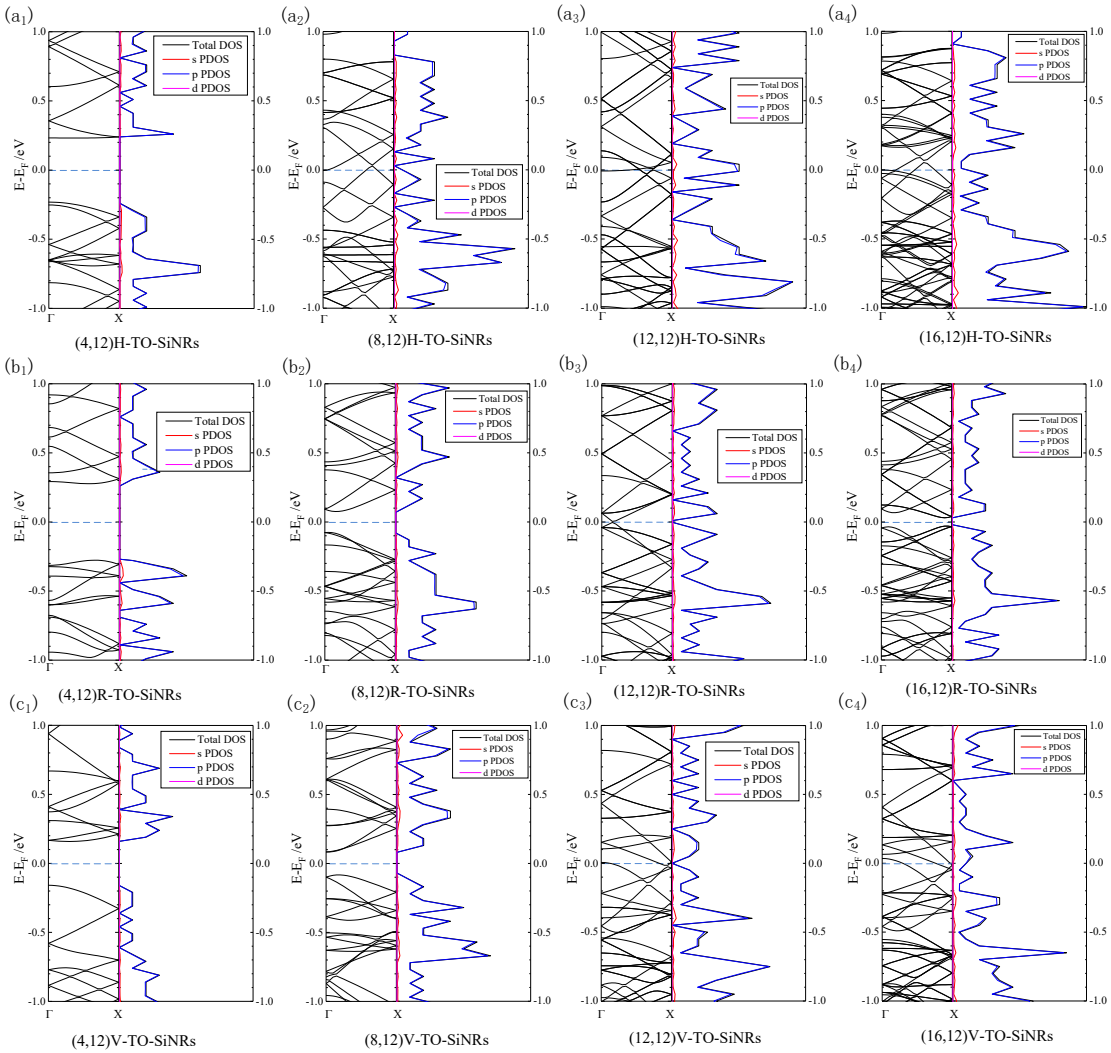
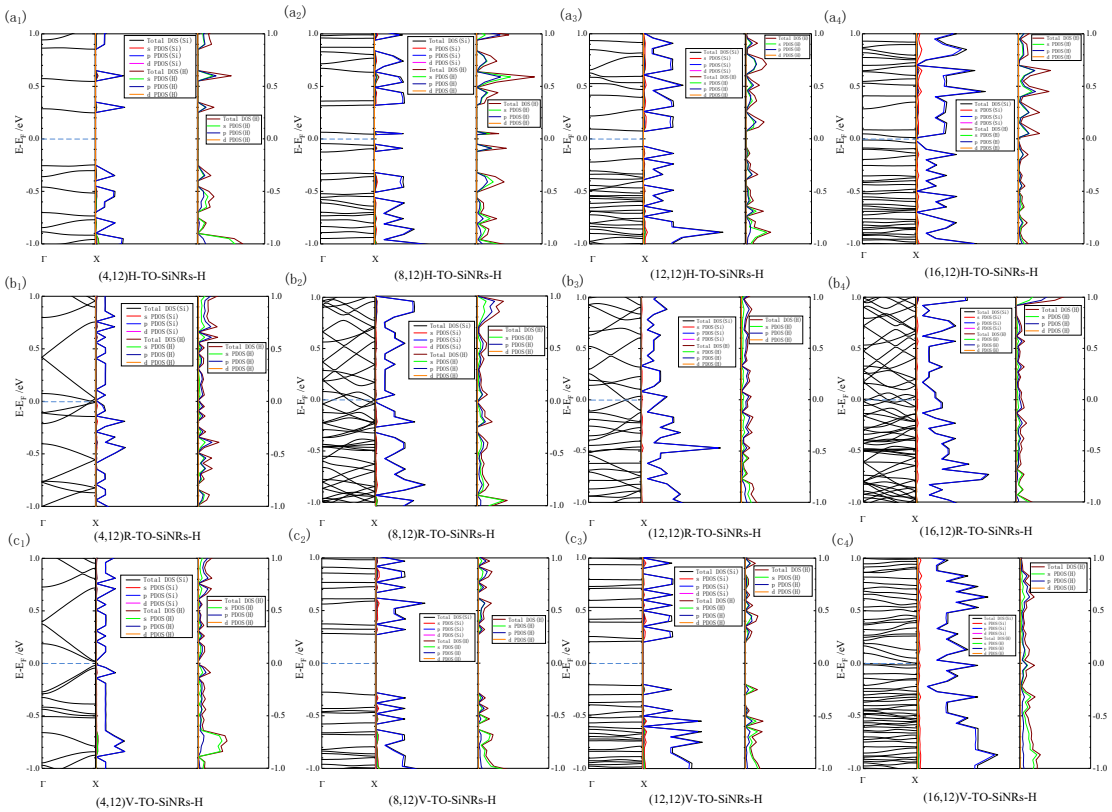

(a1)
(a2)
(a3)
(a4)
(4,12)H-TO-SiNRs
(8,12)H-TO-SiNRs
(12,12)H-TO-SiNRs
(16,12)H-TO-SiNRs
(b1)
(b2)
(b3)
(b4)
(4,12)R-TO-SiNRs
(8,12)R-TO-SiNRs
(12,12)R-TO-SiNRs
(16,12)R-TO-SiNRs
(c1)
(c2)
(c3)
(c4)
(4,12)V-TO-SiNRs
(8,12)V-TO-SiNRs
(12,12)V-TO-SiNRs
(16,12)V-TO-SiNRs
Total DOS
s PDOS
p PDOS
d PDOS
E-E_F /eV
Γ
X

Fig. S4.


(a1)
(a2)
(a3)
(a4)
(4,12)H-TO-SiNRs-H
(8,12)H-TO-SiNRs-H
(12,12)H-TO-SiNRs-H
(16,12)H-TO-SiNRs-H
(b1)
(b2)
(b3)
(b4)
(4,12)R-TO-SiNRs-H
(8,12)R-TO-SiNRs-H
(12,12)R-TO-SiNRs-H
(16,12)R-TO-SiNRs-H
(c1)
(c2)
(c3)
(c4)
(4,12)V-TO-SiNRs-H
(8,12)V-TO-SiNRs-H
(12,12)V-TO-SiNRs-H
(16,12)V-TO-SiNRs-H
E-E_f /eV
Γ
X