Supporting Information For

Tunable Electronic Structures of Germanium Monochalcogenide Nanosheets by Light Non-metallic Atom Functionalization: A First-principles Study

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Contents:

1. Orbitally decomposed total energies of favourable structures

The discrepancy of favourable structures is attributed to the facts that the O/S adatoms have much weaker O/S-Ge bonds in the $E_1$ conformation than in the $T$ one, and in the $E_1$ conformation hollow-site S/Se atoms will suffer from a stronger repulsion from O/S adatoms. To quantitatively describe these effects, we have performed an OpenMX calculation on the O and N adatoms on GeS sheet, which decomposes the total energy into the contribution associated with each atomic basis function [1,2]. We find that comparing to the $T$ conformation, the O adatom in the $E_1$ case lowers the total energy by 0.576 eV, but the hollow-site S atom raises the total energy by 1.648 eV, which makes the $E_1$ conformation unfavourable for O adatom. On the other hand, the N adatom can lower the total energy greatly by about 6.078 eV, while the hollow-site S atom only raises the total energy by 0.834 eV, which stabilizes the $E_1$ conformation for N adatom.

2. Figure S1. PBE band structures of B adatom on the 5×5, 6×6, and 7×7 supercells of GeS and GeSe sheets.

Figure S1: PBE band structures of B adatom on the 5×5, 6×6, and 7×7 supercells of [(a), (c), (e)] GeS and [(b), (d), (f)] GeSe sheets.