Supporting Information For

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

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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.

(1) T. Ozaki and H. Kino, Phys. Rev. B, 2005, **72**, 045121.
(2) K. Lejaeghere, G. Bihlmayer, T. Bjorkman, P. Blaha, S. Blugel, V. Blum, D. Caliste, I. E. Castelli, S. J. Clark, A. Dal Corso, S. de Gironcoli, T. Deutsch, J. K. Dewhurst, I. Di Marco, C. Draxl, M. Dulak, O. Eriksson, J. A. Flores-Livas, K. F. Garrity, L. Genovese, P. Giannozzi, M. Giantomassi, S. Goedecker, X. Gonze, O. Granas, E. K. U. Gross, A. Gulans, F. Gygi, D. R. Hamann, P. J. Hasnip, N. A. W. Holzwarth, D. Iusan, D. B. Jochym, F. Jollet, D. Jones, G. Kresse, K. Koepernik, E. Kucubenli,

Y. O. Kvashnin, I. L. M. Locht, S. Lubeck, M. Marsman, N. Marzari, U. Nitzsche, L. Nordstrom, T. Ozaki, L. Paulatto, C. J. Pickard, W. Poelmans, M. I. J. Probert, K. Refson, M. Richter, G.-M. Rignanese, S. Saha, M. Scheffler, M. Schlipf, K. Schwarz, S. Sharma, F. Tavazza, P. Thunstrom, A. Tkatchenko, M. Torrent, D. Vanderbilt, M. J. van Setten, V. Van Speybroeck, J. M. Wills, J. R. Yates, G.-X. Zhang and S. Cottenier, Science, 2016, **351**, 6280.

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

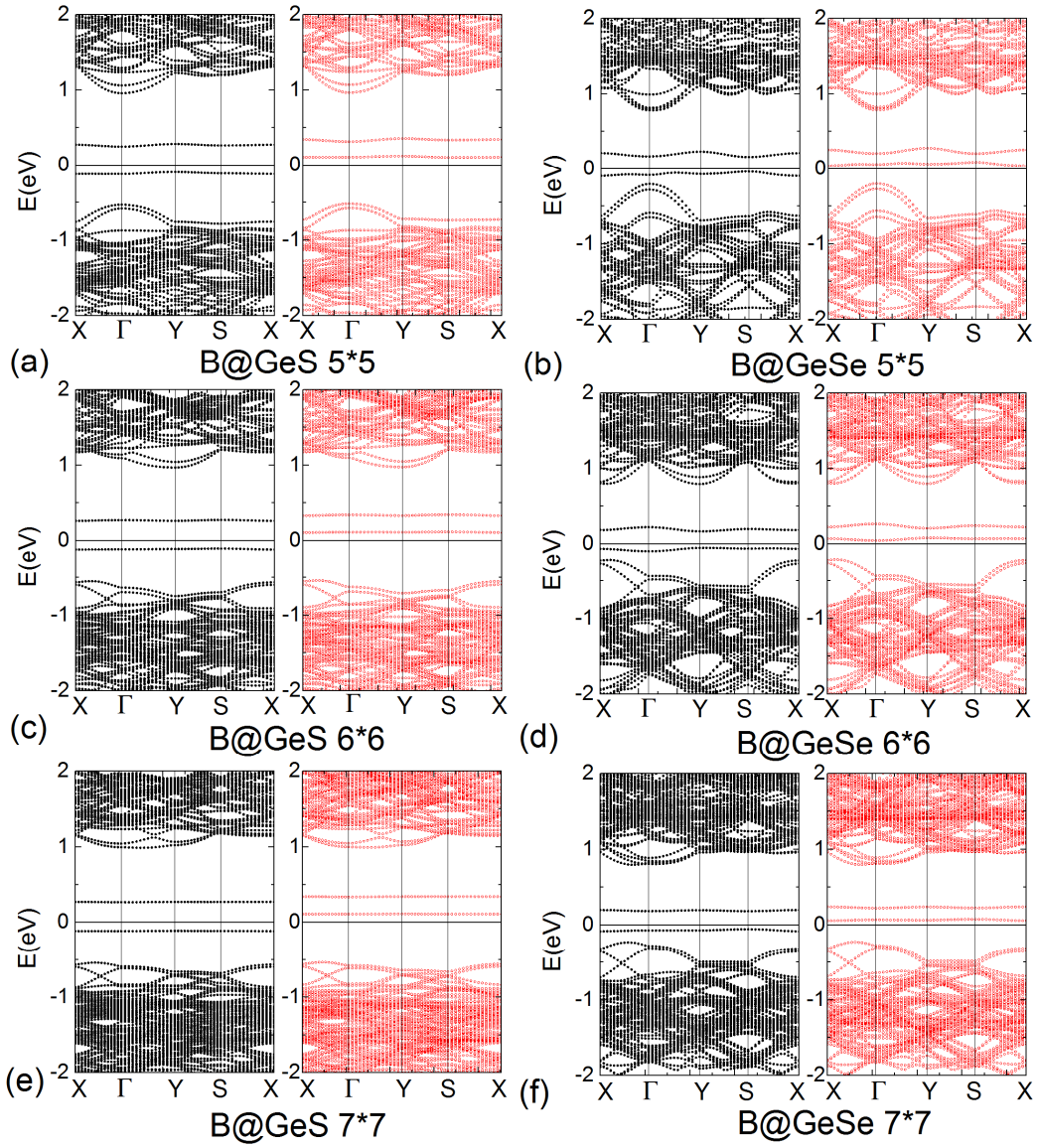

Figure S1: PBE band structures of B adatom on the 5×5, 6×6, and 7×7 supercells of [(a), (c), (e)] GeS and [(b), (d), (f)] GeSe sheets..