Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Effect of Ge doping concentration on the electrochemical performance of silicene anode for lithium-ion batteries: a first-principles study

Jun Song [a, b] *, Mingjie Jiang [a], Jodie A. Yuwono [b], Sailin Liu [b], Jingxiu Wang [b], Qi Zhang [a], Yuhui Chen [a], Jun Zhang [a], Xuehong Wu [a], Juanfang Liu [c, *]

[a] *College of Energy and Power Engineering, Zhengzhou University of Light Industry, Zhengzhou 450000, Henan, China*

[b] *School of Chemical Engineering& Advanced Materials, The University of Adelaide, Adelaide, SA 5005, Australia*

[c] *School of Energy and Power Engineering, Chongqing University, Chongqing 400044, Chongqing, China*

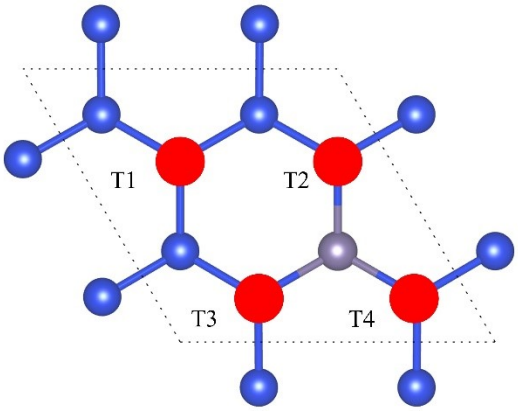

Figure S1 Possible Li adsorption on the T sites of Si_7Ge_1 on the upper surface.

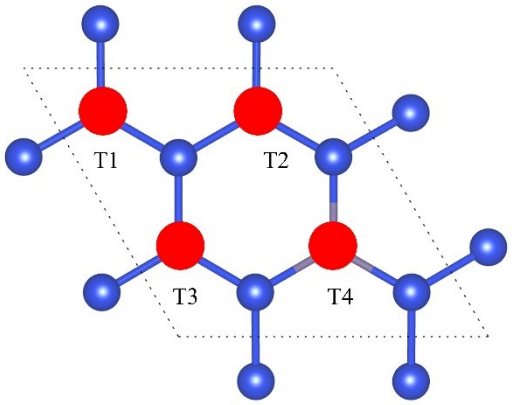

Figure S2 Possible Li adsorption on the T sites of Si_7Ge_1 on the lower surface.

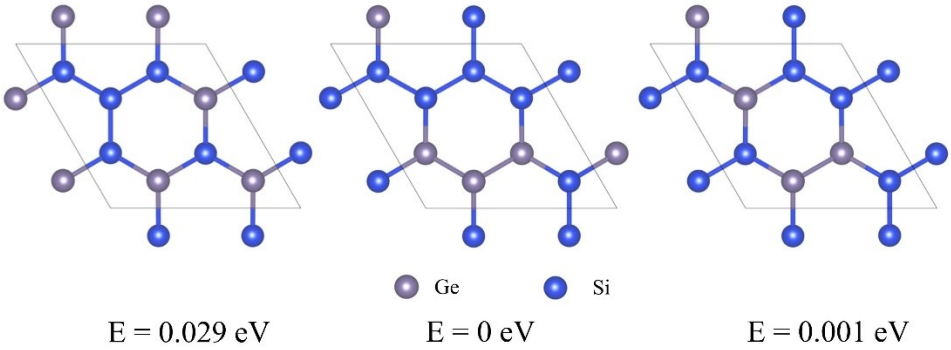

Figure S3 Top view of three possible configurations of Si_5Ge_3.

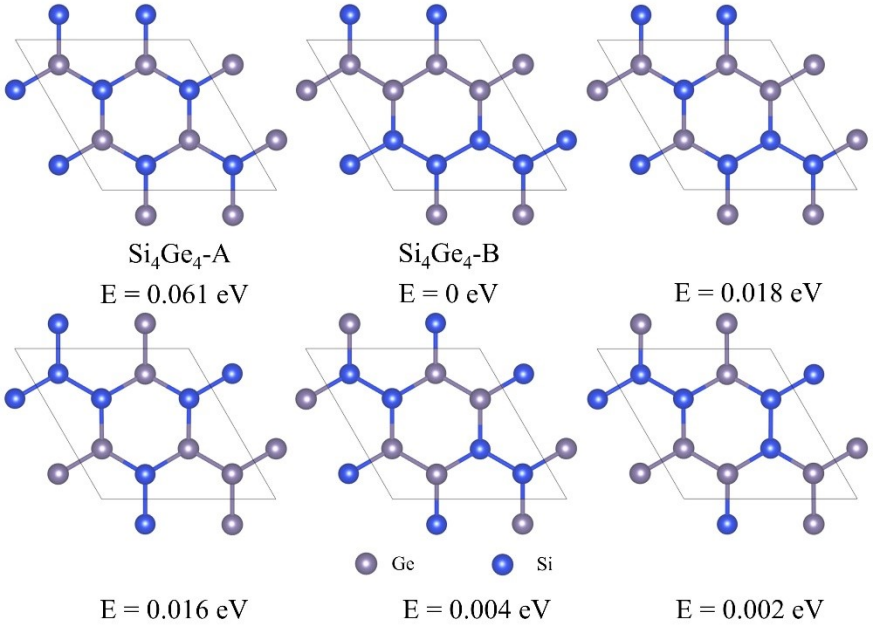

Figure S4 Top view of six possible configurations of Si_4Ge_4.

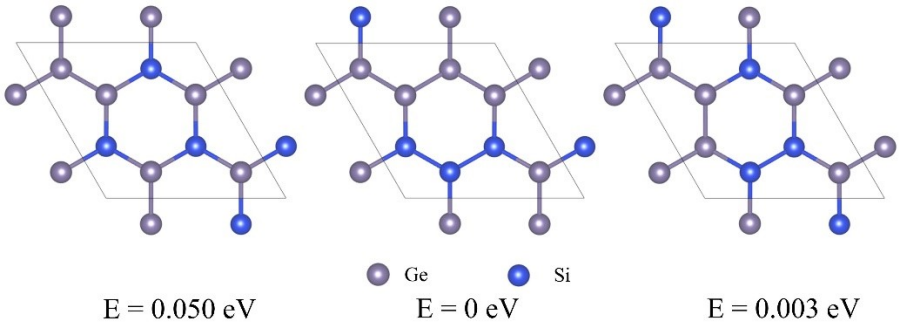

Figure S5 Top view of three possible configurations of Si_3Ge_5.

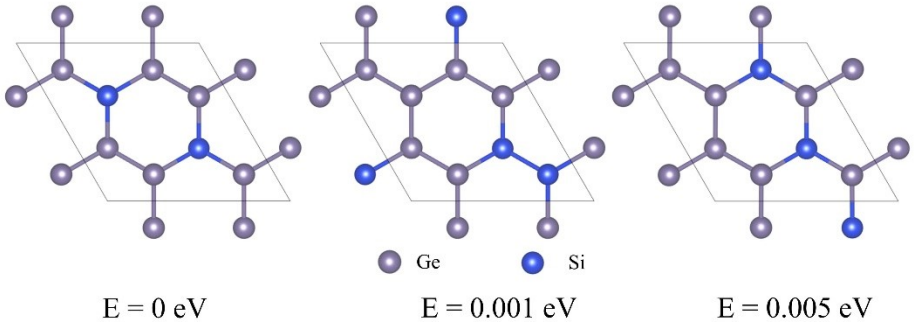

Figure S6 Top view of three possible configurations of Si_2Ge_6.

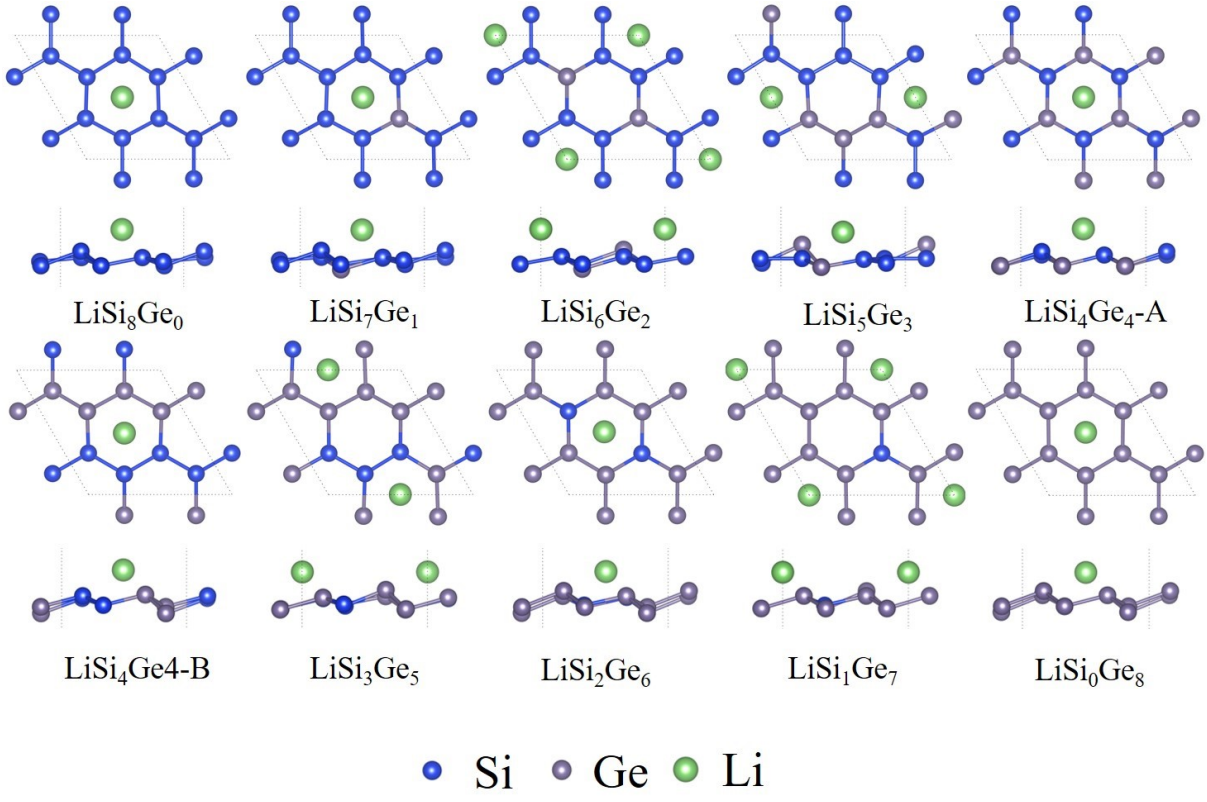

Figure S7 Top view and side view of Li adsorption on all 2D Si_xGe_y investigated here.

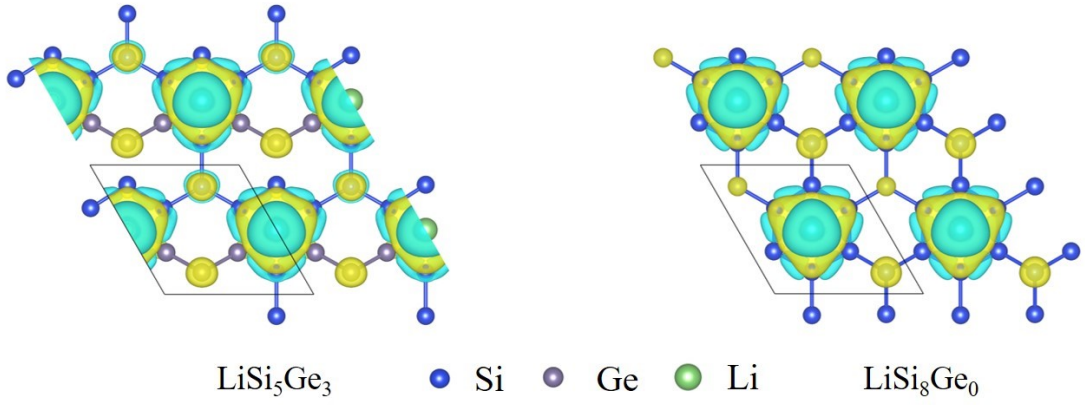

Figure S8 Charge density difference of $LiSi_5Ge_3$ (left) and $LiSi_8Ge_0$ (right).

Yellow and blue color isosurfaces denotes electron loss and gain, respectively.

Table S1 Total electronic energies without and with spin polarization correction.

Structures	Energy (eV) ISPIN=1	Energy (eV) ISPIN=2	△Energy
Si_8Ge_0	-38.7747	-38.7747	0.00%
Si_7Ge_1	-37.9633	-37.9633	0.00%
Si_6Ge_2	-37.1761	-37.1761	0.00%
Si_5Ge_3	-36.4005	-36.4005	0.00%
Si_4Ge_4-A	-35.5916	-35.5916	0.00%
Si_4Ge_4-B	-35.6527	-35.6527	0.00%
Si_3Ge_5	-34.9149	-34.9149	0.00%
Si_2Ge_6	-34.1827	-34.1827	0.00%
Si_1Ge_7	-33.5084	-33.5084	0.00%
Si_0Ge_8	-32.8419	-32.842	0.01%

Table S2 Li adsorption energy on the T sites of Si_7Ge_1 in the upper surface.

Adsorption site	Eads (eV)
T1	-1.81
T2	-1.86
T3	-1.86
T4	-1.86

Table S3 Li adsorption energy on the T sites of Si_7Ge_1 in the lower surface.

Adsorption site	Eads
T1	-1.84
T2	-1.84
T3	-1.84
T4	-1.97

Table S4 Li adsorption energy on 2D Si_xGe_y.

System	E_{ads} (eV)
Silicene	-2.01
Si_7Ge_1	-2.04
Si_6Ge_2	-2.01
Si_5Ge_3	-2.15
Si_4Ge_4-A	-2.01
Si_4Ge_4-B	-2.00
Si_3Ge_5	-2.03
Si_2Ge_6	-2.00
Si_1Ge_7	-2.01
Germanene	-2.01

Table S5 The relationship between atomic ratio used in our DFT model of 2D Si_yGe_z and the mass ratio used in the experiment.

Atomic ratio	Si_7Ge_1	Si_6Ge_2	Si_5Ge_3	Si_4Ge_4-A	Si_4Ge_4-B	Si_3Ge_5	Si_2Ge_6	Si_1Ge_7
Mass ratio	$Si_{0.73}Ge_{0.27}$	$Si_{0.54}Ge_{0.46}$	$Si_{0.39}Ge_{0.61}$	$Si_{0.28}Ge_{0.72}$	$Si_{0.28}Ge_{0.72}$	$Si_{0.19}Ge_{0.81}$	$Si_{0.11}Ge_{0.89}$	$Si_{0.05}Ge_{0.95}$

Table S6 Maximum storage capacity and Li migration energy barrier of 2D anode materials.

2D Materials	Theoretical capacity (mhA/g)	Energy barrier (eV)
GeS [1]	256	0.19
V_2C [2]	941	0.05
Nb_2C [3]	542	0.03
Mo_2C[4]	526	0.04
Ti_3C_2 [5]	319	0.07
graphite [6]	372	0.40
BC_7 [7]	283	1.17
g-CN[8]	813	-
BC_3[9]	714	0.40
C_2N[10]	672	0.03
Stanene[11]	226	0.25
TiC [12]	447	0.03
TiN [12]	433	0.27
SiS[13]	446	0.17
SiSe[13]	250	0.12
ZnP [14]	556	0.56
Antimonene[15]	208	0.34

Reference

[1] ZHOU Y. MX (M = Ge, Sn; X = S, Se) sheets: theoretical prediction of new promising electrode materials for Li ion batteries [J]. J Mater Chem A, 2016, 4(28): 10906-13.

[2] HU J, XU B, OUYANG C, et al. Investigations on V2C and V2CX2 (X = F, OH) Monolayer as a Promising Anode Material for Li Ion Batteries from First-Principles Calculations [J]. J Phys Chem C, 2014, 118(42): 24274-81.

[3] HU J, XU B, OUYANG C, et al. Investigations on Nb2C monolayer as promising anode material for Li or non-Li ion batteries from first-principles calculations [J]. RSC Adv, 2016, 6(33): 27467-74.

[4] ÇAKıR D, SEVIK C, GüLSEREN O, et al. Mo2C as a high capacity anode material: a first-principles study [J]. J Mater Chem A, 2016, 4(16): 6029-35.

[5] TANG Q, ZHOU Z, SHEN P. Are MXenes Promising Anode Materials for Li Ion Batteries? Computational Studies on Electronic Properties and Li Storage Capability of Ti3C2 and Ti3C2X2 (X = F, OH) Monolayer [J]. J Am Chem Soc, 2012, 134(40): 16909-16.

[6] YU T, ZHANG S, LI F, et al. Stable and metallic two-dimensional TaC2 as an anode material for lithium-ion battery [J]. J Mater Chem A, 2017, 5(35): 18698-706.

[7] DAS D, HARDIKAR R P, HAN S S, et al. Monolayer BC2: an ultrahigh capacity anode material for Li ion batteries [J]. Phys Chem Chem Phys, 2017, 19(35): 24230-9.

[8] HANKEL M, SEARLES D J. Lithium storage on carbon nitride, graphenylene and inorganic graphenylene [J]. Phys Chem Chem Phys, 2016, 18(21): 14205-15.

[9] LIU Y, ARTYUKHOV V I, LIU M, et al. Feasibility of Lithium Storage on Graphene and Its Derivatives [J]. J Phys Chem Lett, 2013, 4(10): 1737-42.

[10] ZHANG J, LIU G, HU H, et al. Graphene-like carbon-nitrogen materials as anode materials for Li-ion and mg-ion batteries [J]. Appl Surf Sci, 2019, 487: 1026-32.

[11] MORTAZAVI B, DIANAT A, CUNIBERTI G, et al. Application of silicene, germanene and stanene for Na or Li ion storage: A theoretical investigation [J]. Electrochim Acta, 2016, 213: 865-70.

[12] FAN D, LU S, GUO Y, et al. Two-Dimensional Tetragonal Titanium Carbide: a High-Capacity and High-Rate Battery Material [J]. J Phys Chem C, 2018, 122(27): 15118-24.

[13] KARMAKAR S, CHOWDHURY C, DATTA A. Two-Dimensional Group IV Monochalcogenides: Anode Materials for Li-Ion Batteries [J]. J Phys Chem C, 2016, 120(27): 14522-30.

[14] MORTAZAVI B, BAFEKRY A, SHAHROKHI M, et al. ZnN and ZnP as novel graphene-like materials with high Li-ion storage capacities [J]. Materials Today Energy, 2020, 16: 100392.

[15] SENGUPTA A, FRAUENHEIM T. Lithium and sodium adsorption properties of monolayer antimonene [J]. Materials Today Energy, 2017, 5: 347-54.