Supplementary Material for

Superconductivity and superionic behaviors of Kagome lattice in Li-Si compounds under high pressure

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Contents

The VASP package [S1] is applied to perform density functional theory (DFT) [S2,S3] calculations, with the projected augmented wave pseudopotentials method [S4] to describe the electron-ion interaction, using $1s^2 2s^1$, $2s^2 2p^2$, $2s^2 2p^3$, $3s^2 3p^2$, $3s^2 3p^3$, $4s^2 4p^2$, $4s^2 4p^3$, $5s^2 5p^2$, and $5s^2 5p^3$ valence electronic structures of Li, C, N, Si, P, Ge, As, Sn, and Sb, respectively. The ionic positions and cell parameters are optimized with the criteria of energy and forces being 10^{-5} eV and 0.002 eV/Å, respectively. A sphere with a Wigner-Seitz radius of 1.20 Å is used to obtain the projected density of states on the interstitial position. In *ab initio* molecular dynamics (AIMD) simulations for each temperature, the volume of the simulated system is adjusted to ensure that the pressure varies over a range of less than 1 GPa. The cutoff energy is set 600 eV. We computed the electron-phonon coupling strength and solved the Eliashberg equation. Based on the theory of traditional superconductivity BCS theory and Allen-McMillan equation [S5-S9]:

$$T_c = \frac{\omega_{\log}}{1.2} \exp\left[\frac{-1.04(1+\lambda)}{\lambda(1-0.62\mu^*)-\mu^*}\right]. \quad (1)$$

Typical Coulomb pseudpotential parameter μ^* are set with $\mu^* = 0.1$. The integrated EPC strength λ is given by:

$$\lambda = 2\int_0^\infty \frac{\alpha^2 F(\omega)}{\omega} d\omega. \quad (2)$$

Eliashberg spectral function is obtained by:

$$\alpha^2 F(\omega) = \frac{1}{2\pi N(\varepsilon_F)} \sum_{q\upsilon} \frac{\gamma_{q\upsilon}}{\omega_{q\upsilon}} \delta(\omega - \omega_{q\upsilon}) \quad (3)$$

Phonon wave vector is labeled by **q** and $\omega_{\mathbf{q}\upsilon}$ represent phonon frequency mode υ at **q**. Frequency moments $\omega_{\log}$ are calculated by:

$$\omega_{\log} = \exp\left[\frac{2}{\lambda}\int_0^\infty \frac{d\omega}{\omega} \alpha^2 F(\omega)\right]. \quad (4)$$

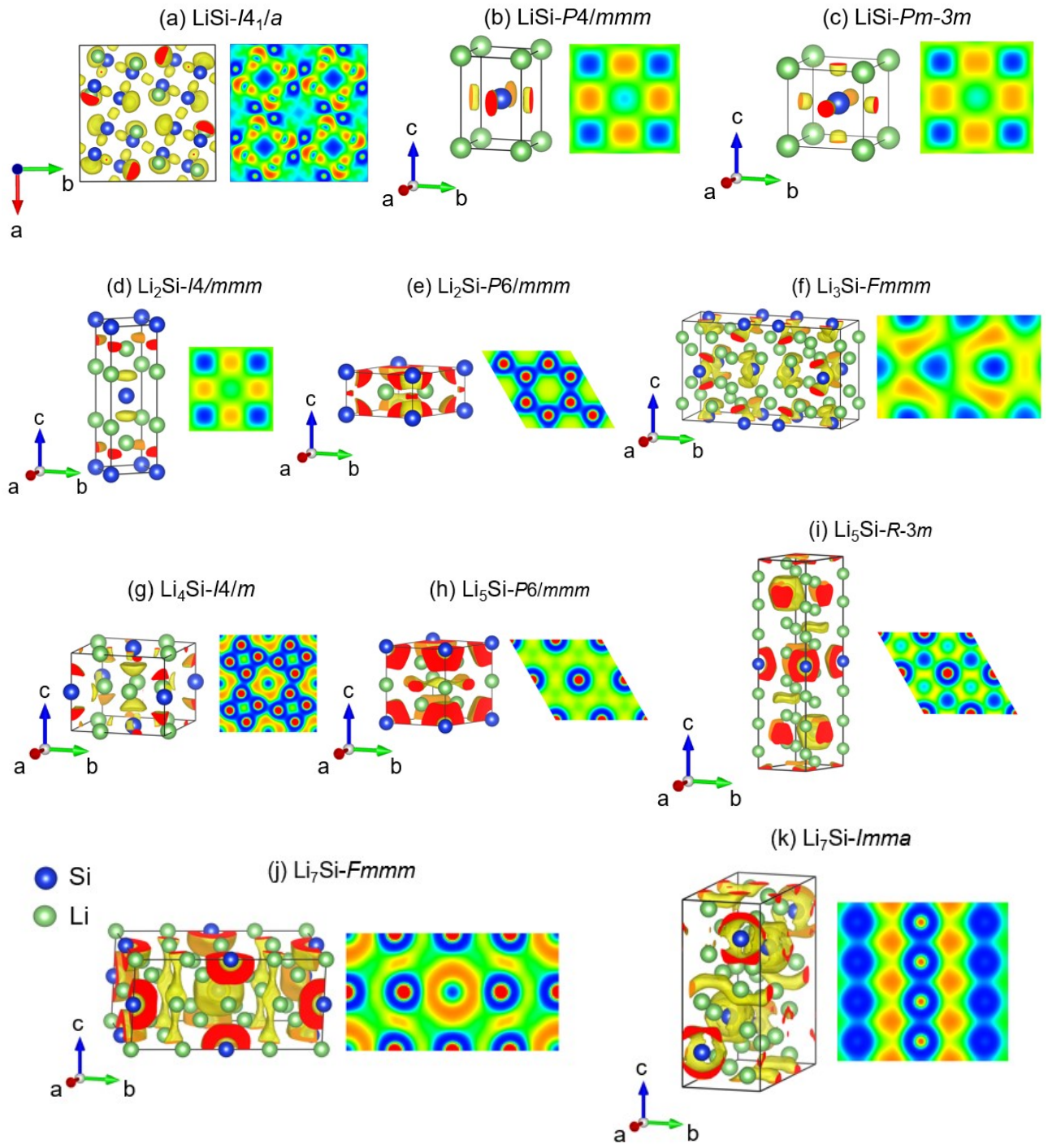
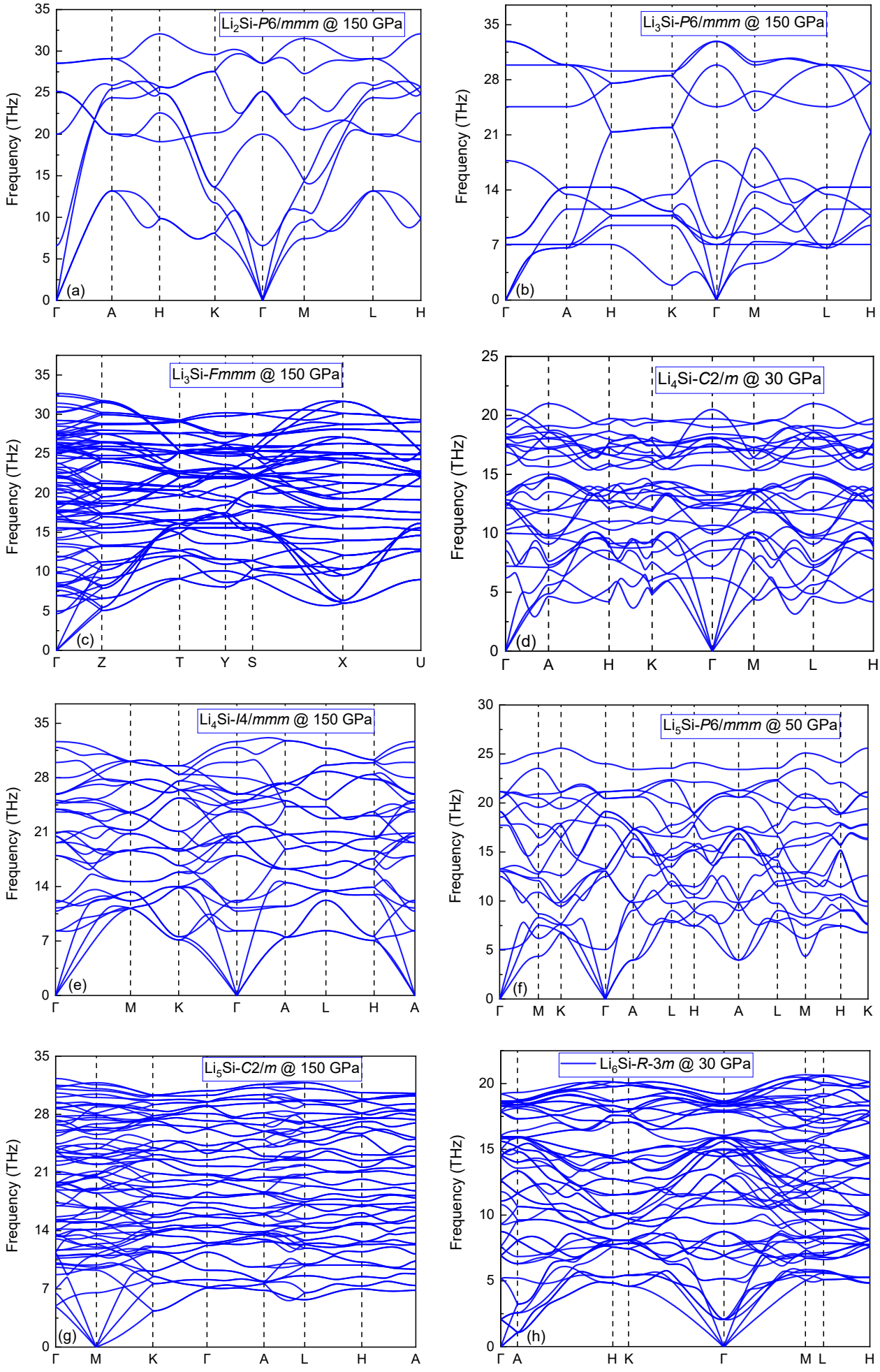


Fig. S1. Charge densities as well as electron localization functions of the Li-Si system: (a) LiSi-$I4_1/a$ at 0 GPa, (b) LiSi-$P4/mmm$ at 20 GPa, (c) LiSi-$Pm\text{-}3m$ at 120 GPa, (d) Li_2Si- $I4/mmm$ at 120 GPa, (e) Li_2Si- $P6/mmm$ at 50 GPa, (f) Li_3Si- $Fmmm$ at 100 GPa, (g) Li_4Si-$I4/m$ at 45 GPa, (h) Li_5Si- $P6/mmm$ at 10 GPa, (i) Li_6Si- $P6/mmm$ at 10 GPa, (j) Li_7Si-$Fmmm$ at 0 GPa, and (k) Li_7Si-$Imma$ at 0 GPa. From the results of these analyses, it can be observed that the dimensionality of the ISQs covers 0D, 1D, and 2D.


Li2Si-P6/mmm @ 150 GPa
(a)
Li3Si-P6/mmm @ 150 GPa
(b)
Li3Si-Fmmm @ 150 GPa
(c)
Li4Si-C2/m @ 30 GPa
(d)
Li4Si-I4/mmm @ 150 GPa
(e)
Li5Si-P6/mmm @ 50 GPa
(f)
Li5Si-C2/m @ 150 GPa
(g)
Li6Si-R-3m @ 30 GPa
(h)
Frequency (THz)
Γ A H K Γ M L H
Γ Z T Y S X U
Γ M K Γ A L H A
Γ M K Γ A L H A L M H K
Γ A H K Γ M L H

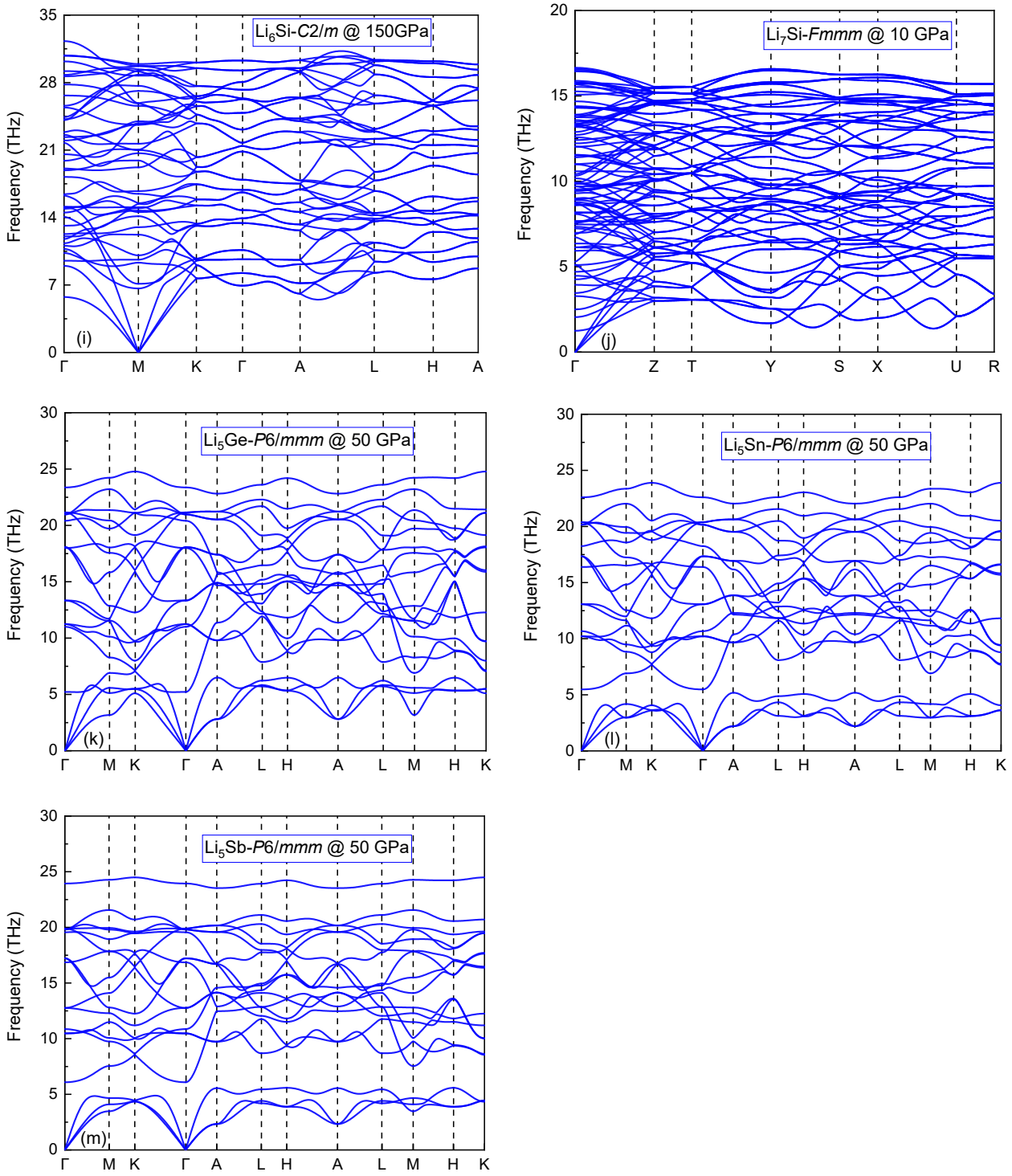


Fig. S2. Phonon dispersion curves for the corresponding structures of Li-Si, Li-Ge, Li-Sn, and Li-Sb systems: (a) Li_2Si-*P*6/*mmm* at 150 GPa, (b) Li_3Si-*P*6/*mmm* at 150 GPa, (c) Li_3Si-*Fmmm* at 150 GPa, (d) Li_4Si-*C*2/*m* at 30 GPa, (e) Li_4Si-*I*4/*mmm* at 150 GPa, (f) Li_5Si-*P*6/*mmm* at 50 GPa, (g) Li_5Si-*C*2/*m* at 150 GPa, (h) Li_6Si-*R*-3*m* at 30 GPa, (i) Li_6Si-*C*2/*m* at 150 GPa, (j) Li_7Si-*Fmmm* at 10 GPa, (k) Li_5Ge-*P*6/*mmm* at 50 GPa, (l) Li_5Sn-*P*6/*mmm* at 50 GPa, and (m) Li_5Sb-*P*6/*mmm* at 50 GPa. The calculated phonon dispersion curves have no imaginary frequencies.

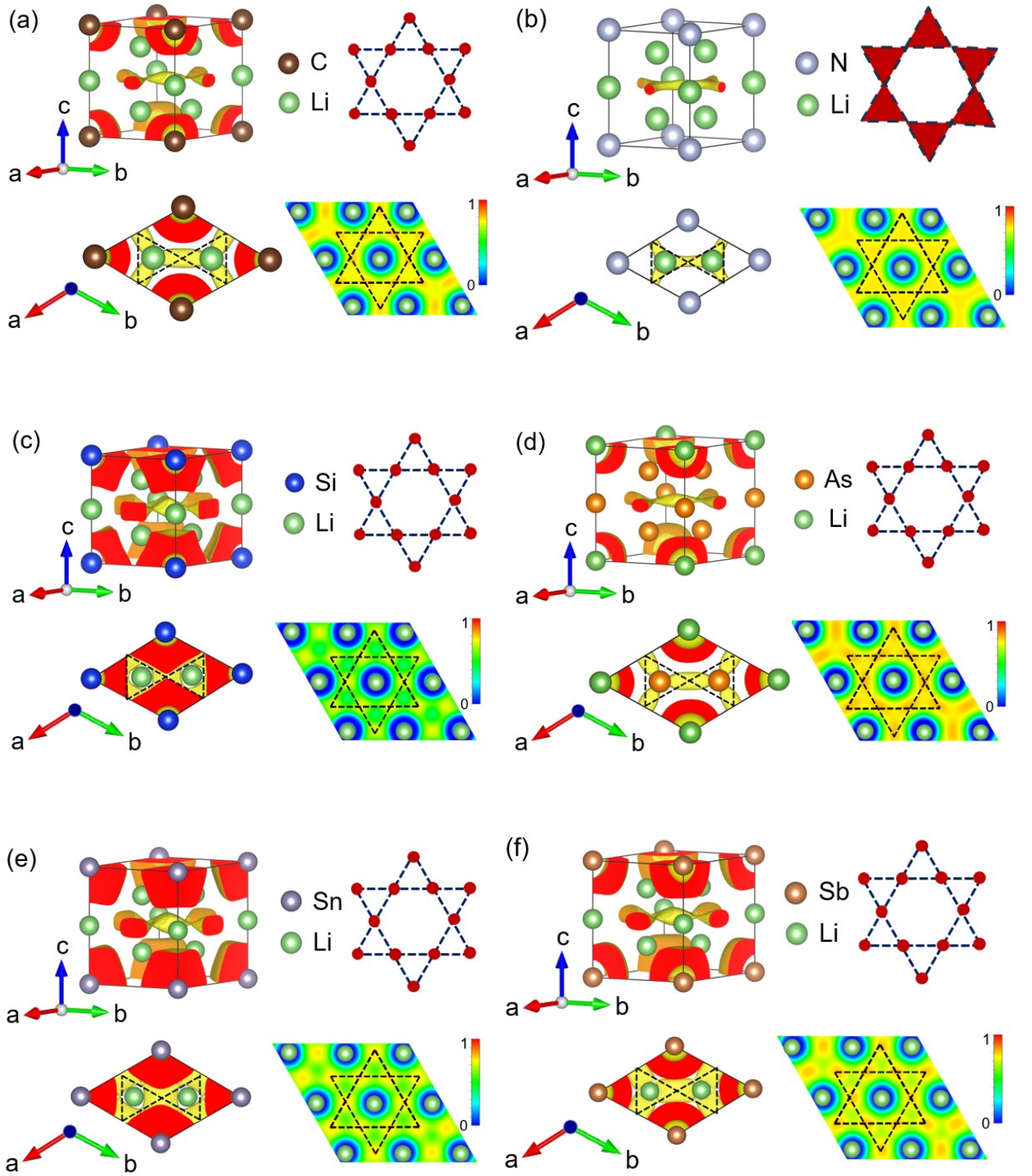


Fig. S3. Atomic structures, charge densities, and electron localization functions of the *P6/mmm* phases: (a) Li_5C-*P6/mmm* at 50 GPa with isovalue = 0.75, (b) Li_5N-*P6/mmm* at 150 GPa with isovalue = 0.89, (c) Li_5Si-*P6/mmm* at 50 GPa with isovalue = 0.55, (d) Li_5As-*P6/mmm* at 40 GPa with isovalue = 0.78, (e) Li_5Sn-*P6/mmm* at 50 Gpa with isovalue = 0.6, and (f) Li_5Sb-*P6/mmm* at 50 GPa with isovalue = 0.65.

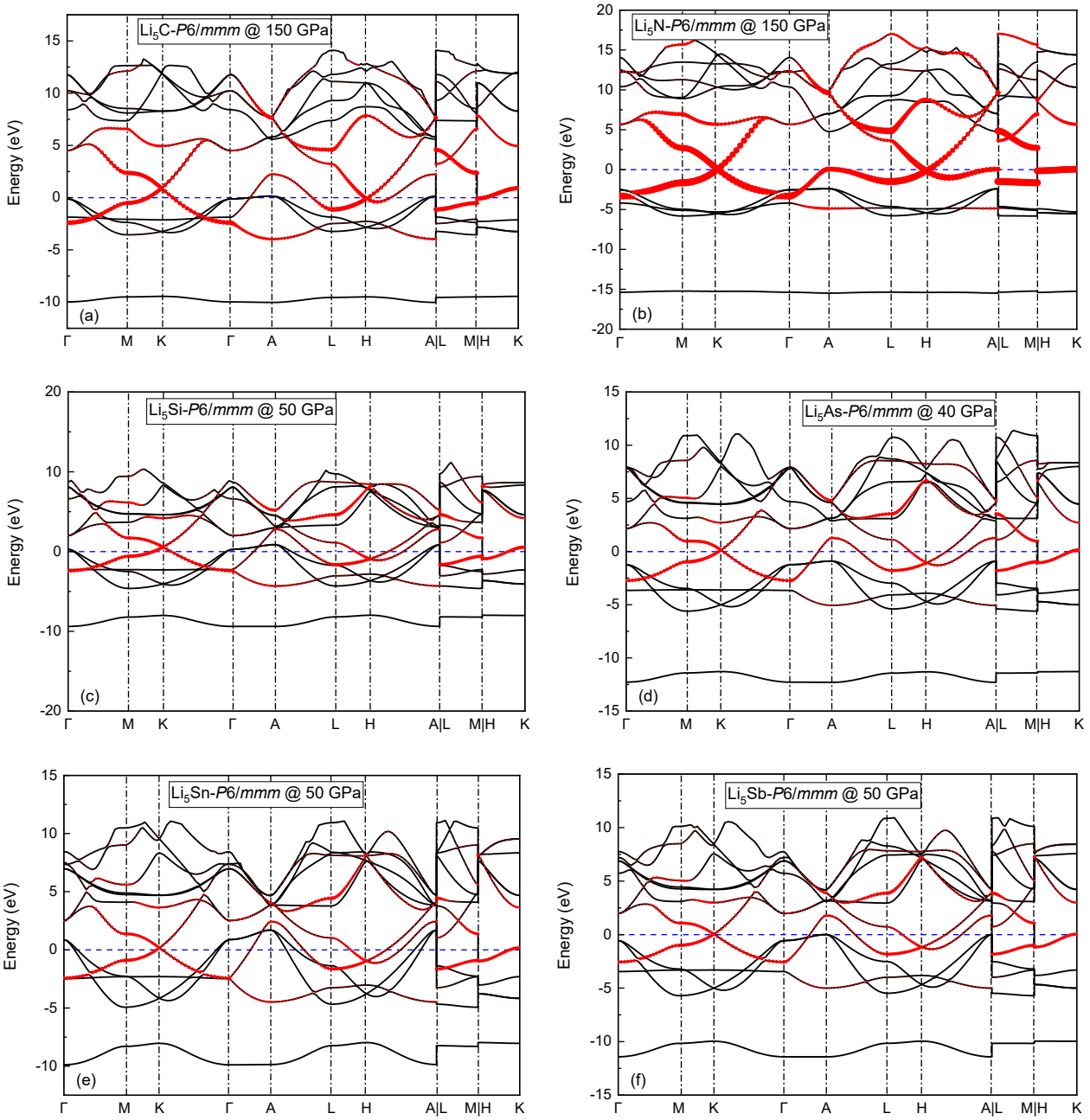


Fig. S4. Energy bands of Kagome lattice formation: (a) Li_5C-*P6/mmm* at 150 GPa, (b) Li_5N-*P6/mmm* at 150 GPa, (c) Li_5Si-*P6/mmm* at 50 GPa, (d) Li_5As-*P6/mmm* at 40 GPa, (e) Li_5Sn-*P6/mmm* at 50 GPa, and (f) Li_5Sb-*P6/mmm* at 50 GPa.

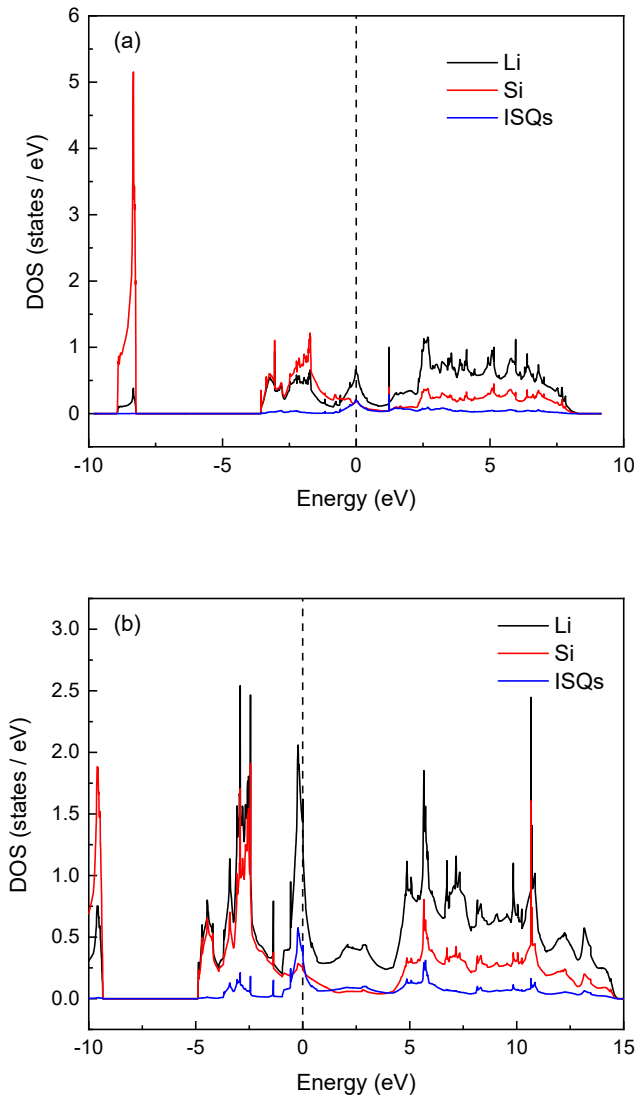
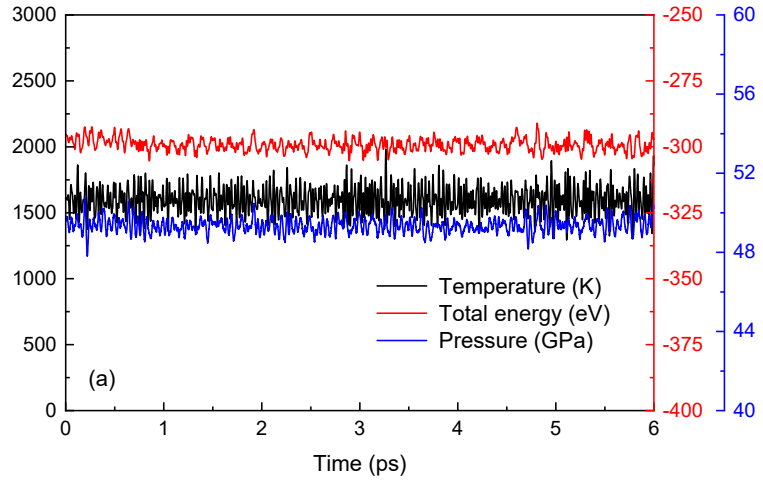
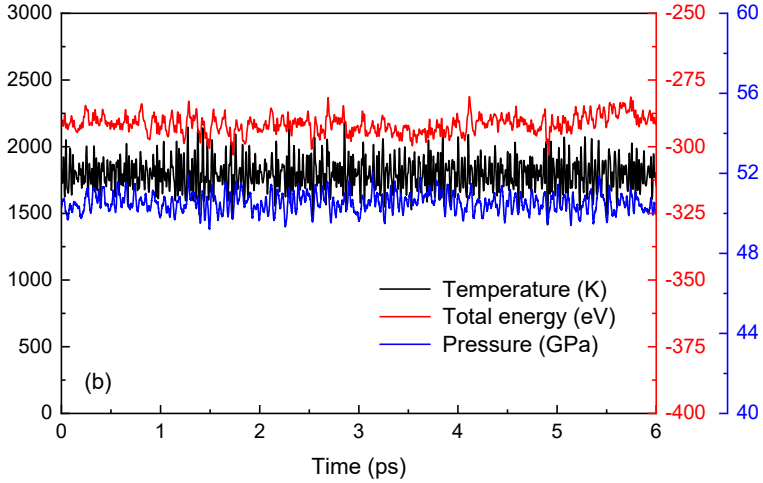
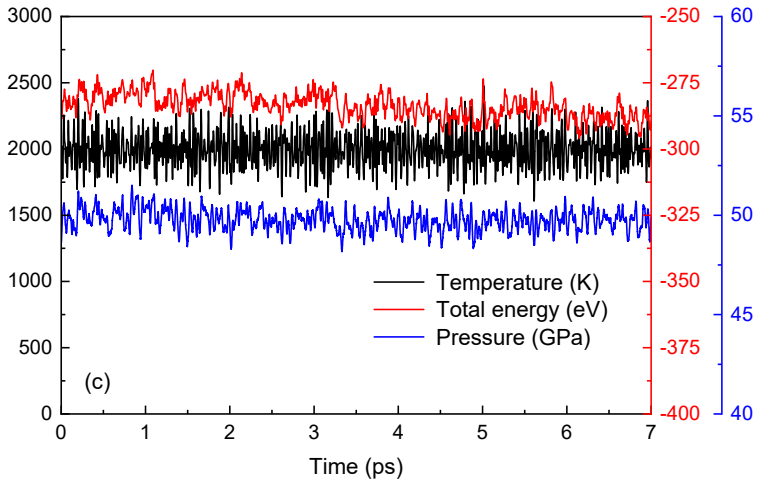


Fig. S5. Contribution of ISQs to the density of states in the Li_5P-*P6*/*mmm* phase: (a) at 0 GPa and (b) at 100 GPa.


3000
2500
2000
1500
1000
500
0
-250
-275
-300
-325
-350
-375
-400
60
56
52
48
44
40
Temperature (K)
Total energy (eV)
Pressure (GPa)
(a)
0
1
2
3
4
5
6
Time (ps)


3000
2500
2000
1500
1000
500
0
-250
-275
-300
-325
-350
-375
-400
60
56
52
48
44
40
Temperature (K)
Total energy (eV)
Pressure (GPa)
(b)
0
1
2
3
4
5
6
Time (ps)


3000
2500
2000
1500
1000
500
0
-250
-275
-300
-325
-350
-375
-400
60
55
50
45
40
Temperature (K)
Total energy (eV)
Pressure (GPa)
(c)
0
1
2
3
4
5
6
7
Time (ps)

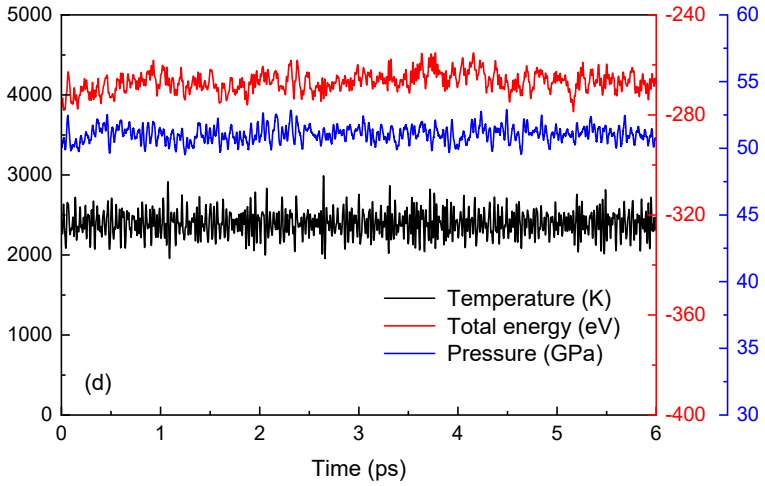
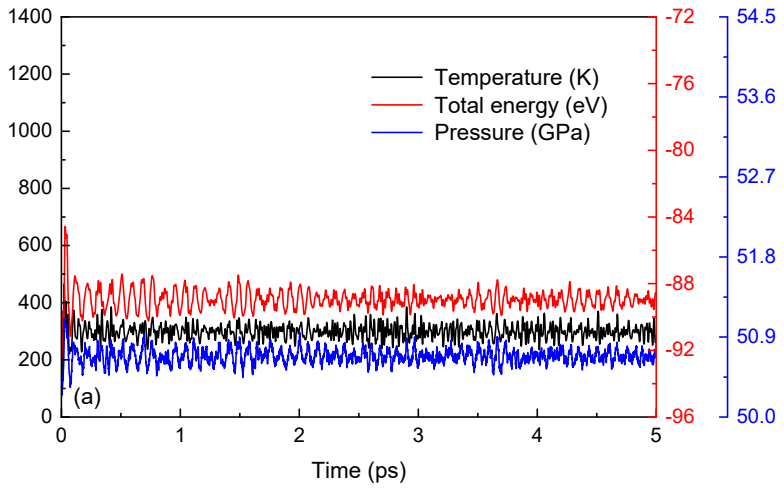
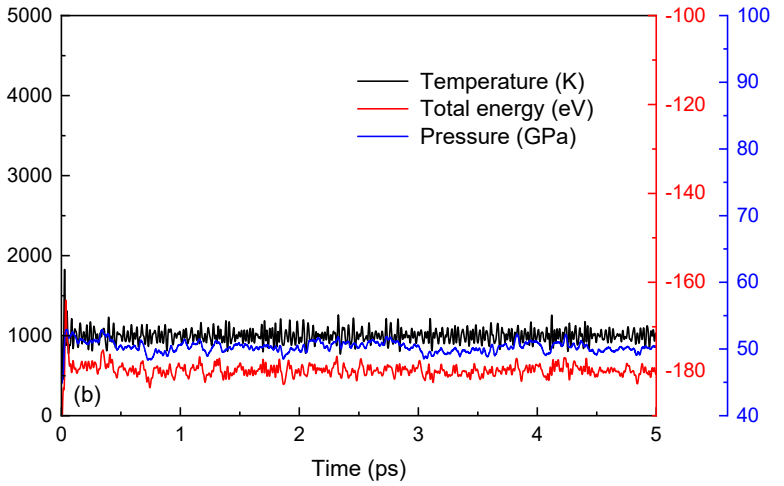


Fig. S6. Temperature, pressure, and energy versus time in the Li_5Si-*P*6/*mmm* phase at (a) 1600 K, (b) 1800 K, (c) 2000 K, and (d) 2400 K. The results indicate that the corresponding systems are highly stable.





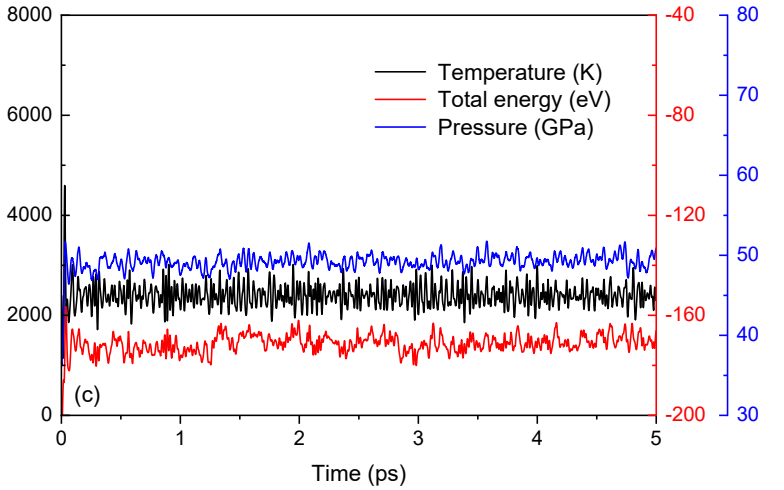


Fig. S7. Temperature, pressure, and energy versus time in the Li_6Si-*C*2/*m* phase at pressure of 50 GPa: (a) 300 K, (b) 1000 K, and (c) 2200 K.

Table S1 Lattice parameters, atomic coordinates, and equivalent isotropic displacement parameters of the predicted stable Li-Si phases.

Compounds & Pressures	Lattice parameters (Å)	Wyckoff positions	*x*	*y*	*z*
Li_3Si	$a = 3.76060$ $\alpha = 90$	Li (3g)	0.50000	0.50000	0.50000
*P*6/*mmm*	$b = 3.76060$ $\beta = 90$	Si (1a)	0.00000	0.00000	0.00000
(150 GPa)	$c = 2.03850$ $\gamma = 120$				
Li_4Si	$a = 7.68420$ $\alpha = 90$	Li1 (4i)	0.40293	1.50000	1.10653
*C*2/*m*	$b = 4.46665$ $\beta = 138.84$	Li2 (4i)	1.22734	1.50000	2.33982
(0 GPa)	$c = 6.79514$ $\gamma = 90$	Si (2d)	0.00000	0.50000	0.50000
Li_4Si	$a = 4.10054$ $\alpha = 90$	Li (8j)	-0.50000	0.20200	0.00000
*I*4/*mmm*	$b = 4.10054$ $\beta = 90$	Si (2a)	-1.00000	-0.00000	-0.00000
(150 GPa)	$c = 3.46897$ $\gamma = 90$				
Li_5Si	$a = 4.09674$ $\alpha = 90$	Li1 (4h)	0.66667	0.33333	0.21845
*P*6/*mmm*	$b = 4.09674$ $\beta = 90$	Li2 (1b)	0.00000	-0.00000	1.50000
(10 GPa)	$c = 5.05114$ $\gamma = 120$	Si (1a)	0.00000	0.00000	0.00000
Li_5Si	$a = 5.57710$ $\alpha = 90$	Li1 (8j)	-0.20659	0.20248	-0.57364
*C*2/*m*	$b = 4.08330$ $\beta = 128.17$	Li2 (4g)	-0.00000	0.79335	-0.00000
(150 GPa)	$c = 7.39580$ $\gamma = 90$	Li3 (4i)	-0.16247	0.50000	-0.70064
		Li4 (4i)	0.05659	0.50000	-0.13898
		Si (4i)	-0.39953	0.50000	-0.21960
Li_6Si	$a = 3.82830$ $\alpha = 90$	Li1 (6c)	1.33333	-0.33333	0.45743
R-3*m*	$b = 3.82830$ $\beta = 90$	Li2 (6c)	0.66667	-0.66667	0.24630
(30 GPa)	$c = 16.3242$ $\gamma = 120$	Li3 (6c)	0.33333	-0.33333	0.03736
		Si (3b)	1.00000	-1.00000	0.50000
Li_6Si	$a = 5.84224$ $\alpha = 90$	Li1 (8j)	0.67289	-0.28609	0.12204
*C*2/*m*	$b = 4.12375$ $\beta = 114.52$	Li2 (4i)	0.09351	-0.50000	-1.61285
(150 GPa)	$c = 3.57250$ $\gamma = 90$	Si (2c)	0.50000	-0.50000	-0.50000
Li_7Si	$a = 11.5614$ $\alpha = 90$	Li1 (16l)	0.10988	0.25000	-0.25000
Fmmm	$b = 6.37239$ $\beta = 90$	Li2 (8g)	-0.27723	0.50000	-0.50000
(10 GPa)	$c = 5.54330$ $\gamma = 90$	Li3 (4a)	-0.00000	0.50000	-0.50000
		Si (4b)	-0.00000	1.00000	-0.50000
Li_7Si	$a = 8.06591$ $\alpha = 90$	Li1 (8i)	-0.21949	-0.25000	0.62665
Fmmm	$b = 3.76992$ $\beta = 90$	Li2 (8i)	0.12269	-0.25000	1.06029
(50 GPa)	$c = 8.72480$ $\gamma = 90$	Li3 (4e)	-0.00000	-0.25000	0.53773
		Li4 (4e)	0.00000	-0.25000	0.80904
		Si (8i)	0.37478	0.25000	0.80231
Li_7Si	$a = 5.89769$ $\alpha = 90$	Li1 (8j)	0.55361	-0.31084	0.38060
*C*2/*m*	$b = 4.06788$ $\beta = 134.03$	Li2 (8j)	0.78201	-0.20446	0.94002
(150 GPa)	$c = 10.3443$ $\gamma = 90$	Li3 (4i)	0.17368	-0.50000	0.15590
		Li4 (4i)	0.89942	-0.50000	0.70442

		Li5 (4i)	0.63762	-0.50000	0.22065
		Si (4h)	0.50000	-0.80071	0.50000
Li_8Si	a = 4.40650 α = 90	Li1 (2c)	1.00000	1.00000	0.69121
R-3*m*	b = 4.40650 β = 90	Li2 (2d)	1.33333	0.66667	0.45634
(0 GPa)	c = 8.43180 γ = 120	Li3 (2d)	0.66667	0.33333	0.17924
		Li4 (2d)	0.66667	0.33333	-0.10385
		Si (1a)	1.00000	1.00000	0.00000
Li_8Si	a = 5.97224 α = 90	Li1 (8j)	0.50867	-0.18891	0.21749
*C*2/*m*	b = 4.01358 β = 144.78	Li2 (4i)	-0.01120	-0.00000	-0.64065
(150 GPa)	c = 7.10957 γ = 120	Li3 (4g)	0.50000	-0.30192	-0.00000
		Si (2d)	0.00000	-0.50000	-0.50000
Li_5Ge	a = 4.35430 α = 90	Li1 (1b)	-1.00000	0.00000	-0.50000
*P*6/*mmm*	b = 4.35430 β = 90	Li2 (4h)	-0.66667	0.66667	-0.78023
(0 GPa)	c = 5.35750 γ = 120	Si (1a)	-1.00000	0.00000	-0.00000
Li_5Sn	a = 4.68550 α = 90	Li1 (4h)	0.66667	0.33333	0.22681
*P*6/*mmm*	b = 4.68550 β = 90	Li2 (1b)	0.00000	0.00000	0.50000
(0 GPa)	c = 5.75500 γ = 120	Si (1a)	0.00000	0.00000	0.00000
Li_5Sb	a = 4.52590 α = 90	Li1 (4h)	0.66667	-0.66667	-0.76225
*P*6/*mmm*	b = 4.52590 β = 90	Li2 (1b)	-0.00000	-1.00000	-0.50000
(0 GPa)	c = 5.50810 γ = 120	Si (1a)	-0.00000	-1.00000	-0.00000

[S1] G. Kresse and J. Furthmüller, Phys. Rev. B **54**, 11169 (1996).

[S2] P. Hohenberg and W. Kohn, Phys. Rev. **136**, B864 (1964).

[S3] W. Kohn and L. J. Sham, Phys. Rev. **140**, A1133 (1965).

[S4] P. E. Blöchl, Phys. Rev. B **50**, 17953 (1994).

[S5] K. Kitazawa, M. Naito, T. Itoh, and S. Tanaka, J. Phys. Soc. JPN. **54**, 2682 (1985).

[S6] L. N. Oliveira, E. K. U. Gross, and W. Kohn, Phys. Rev. Lett. **60**, 2430 (1988).

[S7] M. Lüders, M. A. L. Marques, N. N. Lathiotakis, A. Floris, G. Profeta, L. Fast, A. Continenza, S. Massidda, and E. K. U. Gross, Phys. Rev. B **72**, 024545 (2005).

[S8] M. A. L. Marques, M. Lüders, N. N. Lathiotakis, G. Profeta, A. Floris, L. Fast, A. Continenza, E. K. U. Gross, and S. Massidda, Phys. Rev. B **72**, 024546 (2005).