Supporting information for

Pressure-induced superhard $SiCN_4$ compound uncovered by first-principles calculation

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Table S1. Structure parameters and atomic positions (Wyckoff positions) of the predicted $P4_12_12$ and *Fdd*2 structure at 0 GPa, and *R*-3 structure at 300 GPa, respectively.

Structure	Lattice parameters (Å)	Atomic position (Wyckoff positions)				
$P4_12_12$	a = b = 4.946 c = 7.229 α = β = γ = 90°	Si	4a	0.2227	0.2227	0.0000
		C	4a	0.7768	0.7768	0.0000
		N1	8b	0.9274	0.2586	0.5498
		N2	8b	0.6172	0.7236	0.1655
*Fdd*2	a = 7.725 b = 7.292 c = 5.873 α = β = γ = 90°	Si	8a	0.7500	-0.7500	-0.5482
		C	8a	0.5000	-0.5000	-0.7518
		N1	16b	0.4141	-0.2979	-0.1171
		N2	16b	0.4657	-0.3445	-0.5864
R-3	a = b = 5.270 c = 13.062 α = β = 90° γ = 120°	Si1	3b	0.0000	0.0000	0.5000
		Si2	9d	-0.1667	-0.3333	-0.1667
		C1	9e	0.1667	0.3333	0.3333
		C2	3a	0.0000	0.0000	0.0000
		N1	6c	-0.0000	0.0000	0.3039
		N2	6c	-0.0000	0.0000	0.7948
		N3	18f	-0.2958	-0.3992	0.3942
		N4	18f	-0.2377	0.0255	0.4257

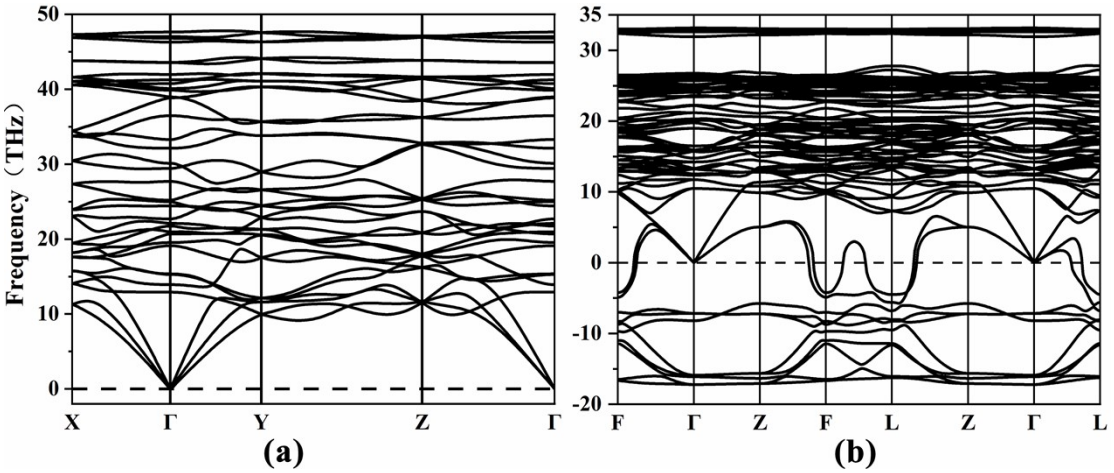


Fig. S1. The phonon dispersion curves of (a) *Fdd*2 at 200 GPa and (b) is *R*-3 at 0 GPa.

Table S2. The bond type and bond length of $P4_12_12$ and *Fdd*2 at 0 GPa, *R*-3 at 300 GPa.

Structure	Chemical Bonds	Bond Length(Å)
$P4_12_12$	Si-N1	1.76877
	Si-N2	1.78843
	C -N1	1.51672
	C -N2	1.45734
	N1-N2	1.41072
*Fdd*2	Si-N1	1.93438
	Si-N2	1.81727
	Si-N2	2.05466
	C -N1	1.47930
	C -N2	1.51608
	N1-N2	1.40499
R-3	Si1-N4	1.64219
	Si2-N2	1.60225
	Si2-N3	1.66726
	Si2-N4	1.63863
	C1-N1	1.56918
	C1-N3	1.55413
	C1-N4	1.49167
	C2-N3	1.54211
	N1-N2	1.28958
	N3-N4	1.30044

Table S3. Atomic charges obtained from a purely ionic model (Q^i), Bader analysis (Q^B) and their differences ($\Delta Q = Q^B - Q^i$). The polyhedrons formed in each structure are also shown.

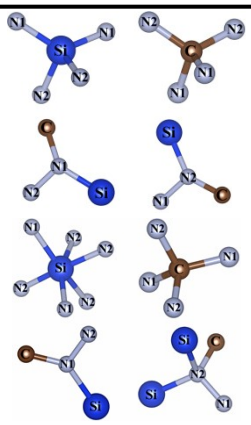
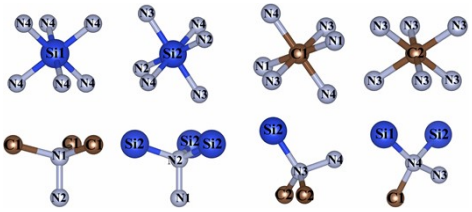
Structure	Atoms	Q^B	Q^i	ΔQ	Atomically-bonded polyhedrons
$SiCN_4$-$P4_12_12$ (0GPa)	Si	1.05	4	-2.95	
	C	2.83	4	-1.17	
	N1	6.02	5	+1.02	
	N2	6.04	5	+1.04	
$SiCN_4$-$Fdd2$ (0GPa)	Si	1.06	4	-2.94	
	C	2.93	4	-1.07	
	N1	5.91	5	+0.91	
	N2	6.09	5	+1.09	
$SiCN_4$-R-3 (300 GPa)	Si1	0.82	4	-3.18	
	Si2	0.83	4	-3.17	
	C1	2.74	4	-1.26	
	C2	2.80	4	-1.20	
	N1	5.56	5	+0.56	
	N2	6.65	5	+1.65	
	N3	5.88	5	+0.88	
	N4	6.33	5	+1.33	
γ-Si_3N_4-Fd-3m (0 GPa)	Si1	0.99	4	-3.01	
	Si2	1.05	4	-2.95	
	N	7.24	5	+2.24	

Fig. S2. MD simulation at 300, 1000 and 1500 K and a volume corresponding to cold pressure of 0 GPa for $P4_12_12$ and *Fdd*2 structures. (a), (b) and (c) are $P4_12_12$ Energy-Time steps. (d), (e) and (f) are *Fdd*2 Energy-Time steps.

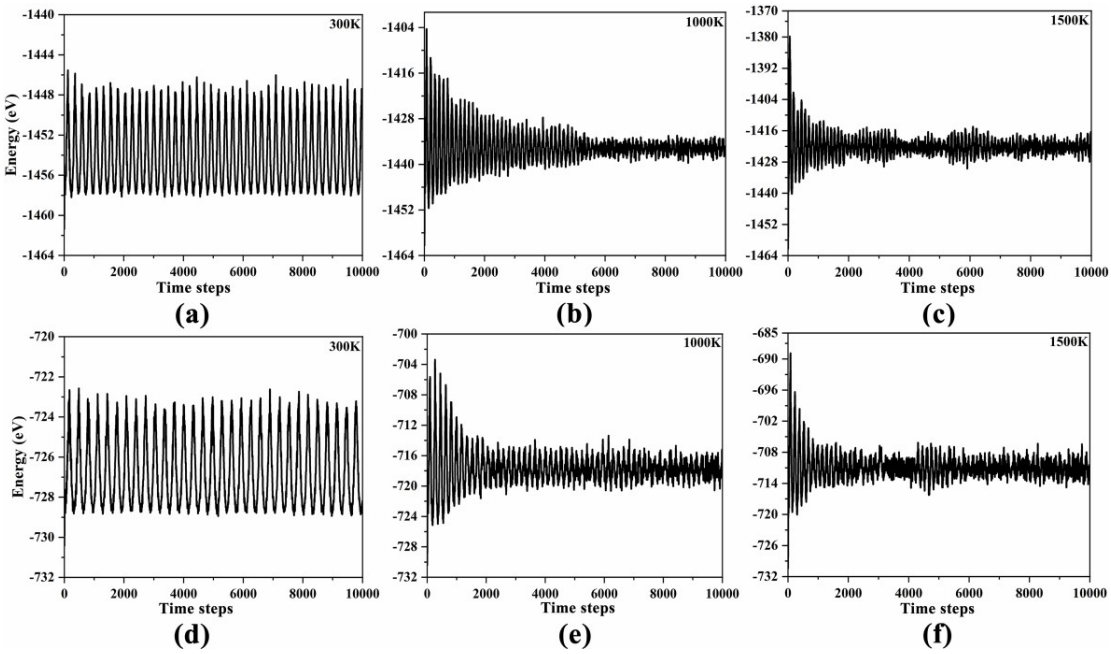


Fig. S3. MD simulation at 300, 1000 and 1500 K and a fixed volume correspond a cold pressure of 200 GPa for *Fdd*2 and 300 GPa for *R*-3 structure. (a), (b) and (c) are *Fdd*2 Energy-Time steps. (d), (e) and (f) are *R*-3 Energy-Time steps. (g), (h) and (i) are snapshots of the final frame of *Fdd*2 at 300 K, 1000 K and 1500 K at the end of 10 ps MD simulations, and (j), (k) and (l) are that of *R*-3.

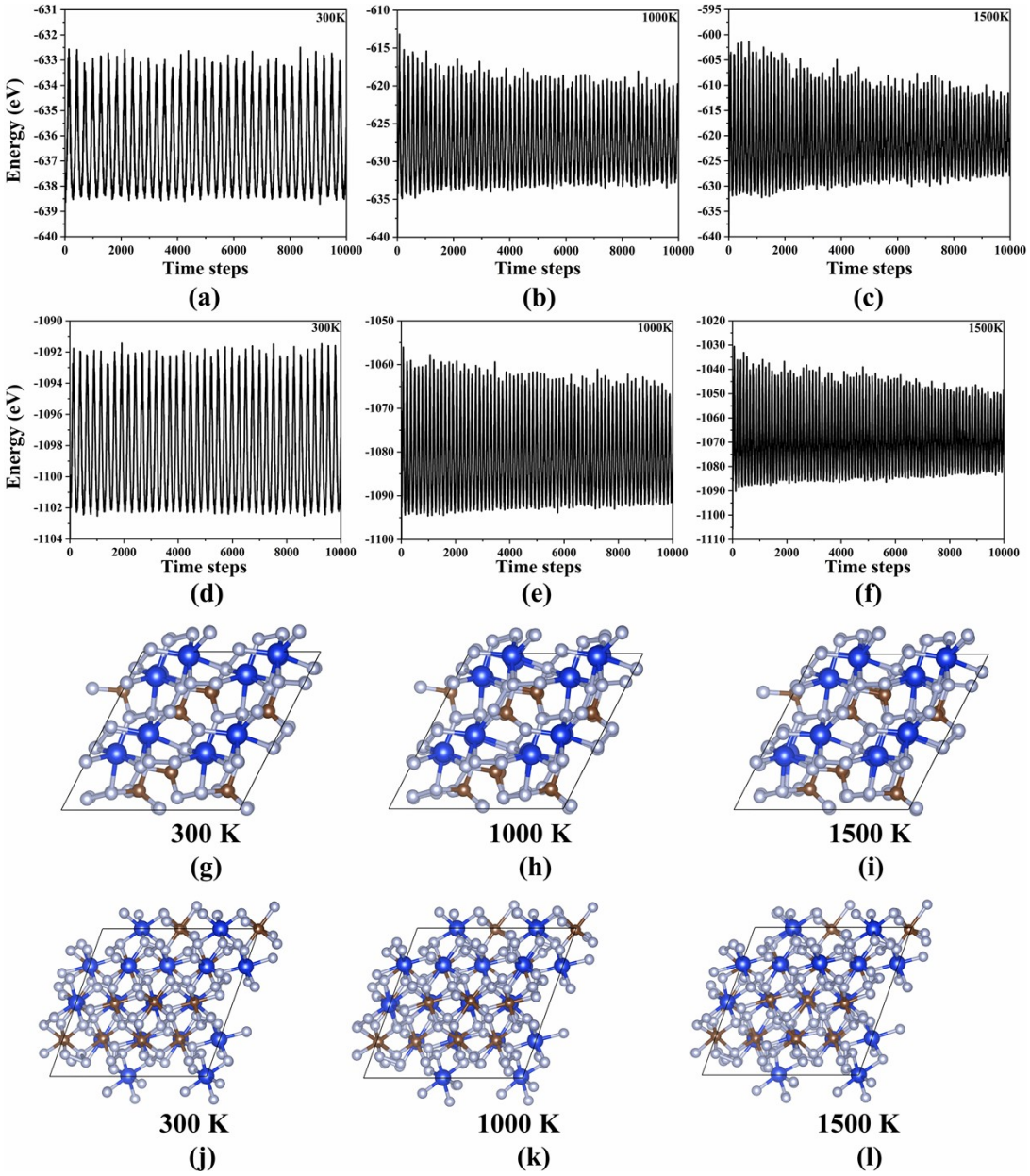


Table S4. The calculated elastic constants C_{ij} (GPa) of $P4_12_12$, *Fdd*2 and *R*-3 structures at 0 GPa.

structure	C_{11}	C_{12}	C_{13}	C_{14}	C_{15}	C_{22}	C_{23}	C_{33}	C_{44}	C_{55}	C_{66}
$P4_12_12$	654	59	179	—	—	—	—	836	365	—	198
*Fdd*2	733	129	125	—	—	952	97	771	351	323	396
R-3	863	115	45	100	3	—	—	909	284	—	—