https://doi.org/10.1007/s11467-022-1227-5

RESEARCH ARTICLE

Structural screening of phosphorus sulfur ternary hydride PSH6 with a high-temperature superconductivity at 130 GPa

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Received June 24, 2022; accepted November 3, 2022

Supporting Information

Contents

1. The ternary phase diagram of P, S and H at 130 GPa

High-pressure structural searches were performed from PSO algorithm using the CALYPSO code, whose effectiveness has been confirmed by the successful applications in discovering the structures of solids, surfaces, and clusters at given chemical compositions and external conditions (e.g., pressure). In pressure range of 10-200 GPa, we predicted more than 10 000 structures in the P-S-H system using variable composition structural searches, and the number of predicted structures at the PSH_6 stoichiometry is more than 1000.

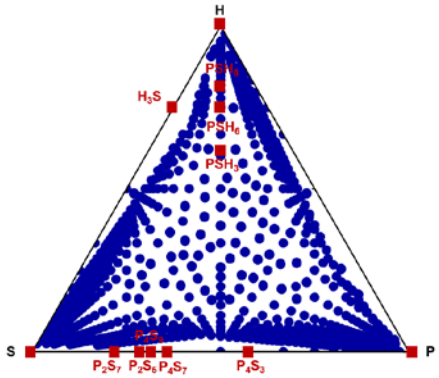

Fig. S1 Ternary phase diagram of P-S-H at 130 Gpa.

2. Crystal structure and Relative formation enthalpy of PSH_6

We selected several structures with the lowest enthalpy of formation from the search results of PSH_6 and showed them in Fig. S2. In addition, we show the change of crystal structure with pressure in Fig. S3. With the increasing pressure, SPH_6 gradually evolves to Pm-3m, a highly symmetrical space group.

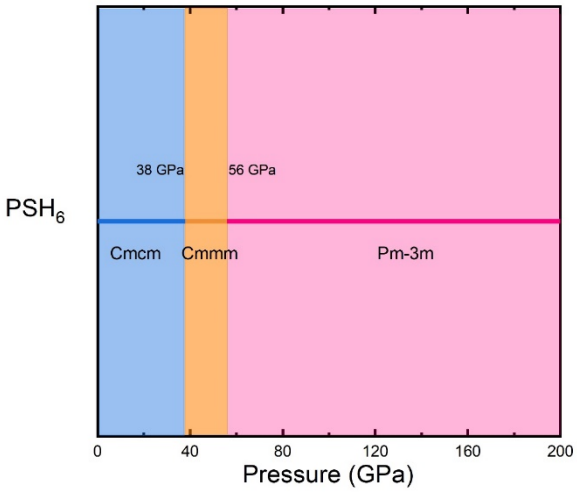

Fig. S2 Phase change with pressure.

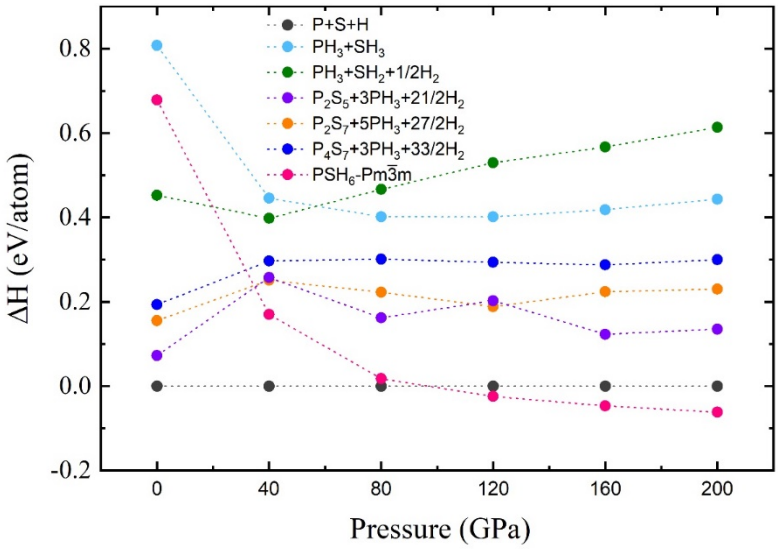

Fig. S3 The decomposition of PSH6 into other stable binary compounds, such as PH_3+SH_3, $2PH_3$ + $2SH_2$ + H_2, P_2S_7 + $5PH_3$ + $27/2H_2$, P_2S_5 + $3PH_3$ + $21/2H_2$, P_4S_7 + $3PH_3$ + H_2, P_4S_9 + $5PH_3$ + $39/2H_2$ and P_4S_3 + SH_3 + $21/2H_2$.

3. RMSD of PSH_6 at 200 GPa and 50 – 500 K

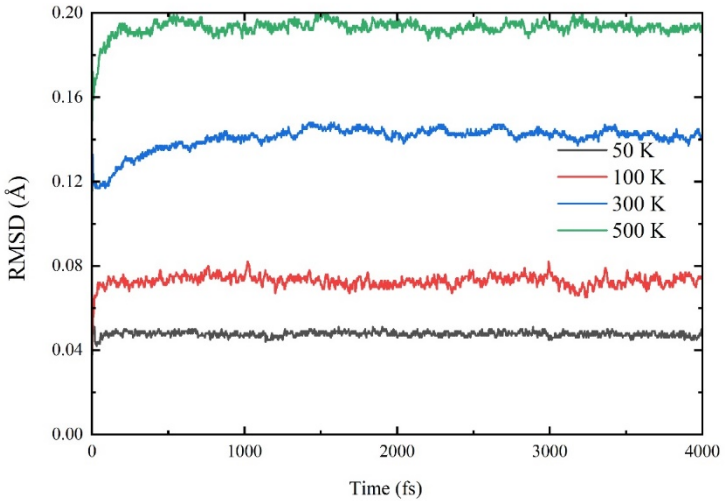

Fig. S4 The RMSD of PSH_6 at 200 GPa and 50 – 500 K.

4. Electronic structures of PSH_6

In order to discuss and analyze the electronic structure of PSH_6, under the premise that all phonons are stable, we calculated the electronic density of states and band structure at each pressure. For geometric optimization, the cutoff energy selected is 600 eV, and the grid density selected in K-space is 32*32*32. To calculate the electronic density of states, we use a denser K-space grid. The path of the high symmetry point selected in the reciprocal space is Γ-X-M-Γ-R-X|M-R, and the interface is selected along the path of the high symmetry point to obtain the band structure.

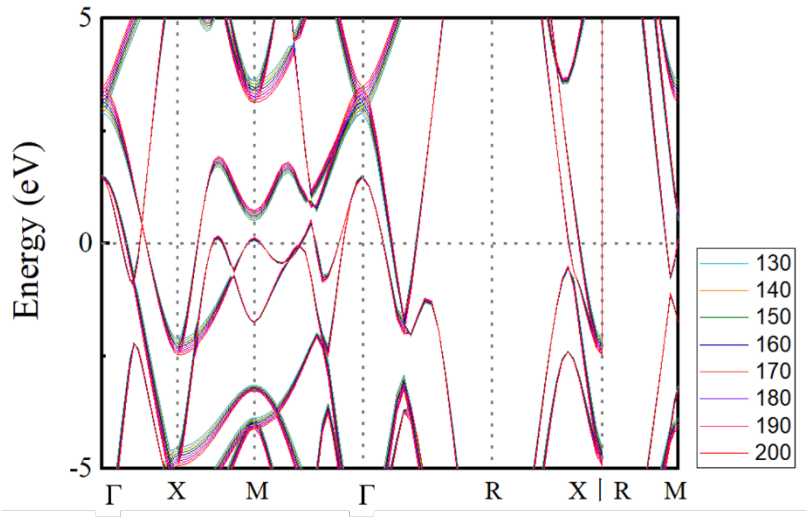

Fig. S5 Band structure of PSH_6 at 130-200 GPa.

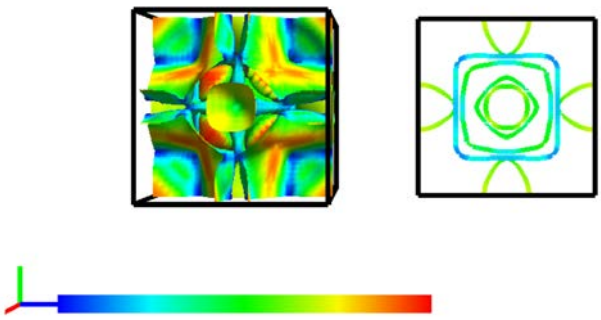

Fig. S6 The 3D Fermi surface of PSH_6 (left) and 2D projection.

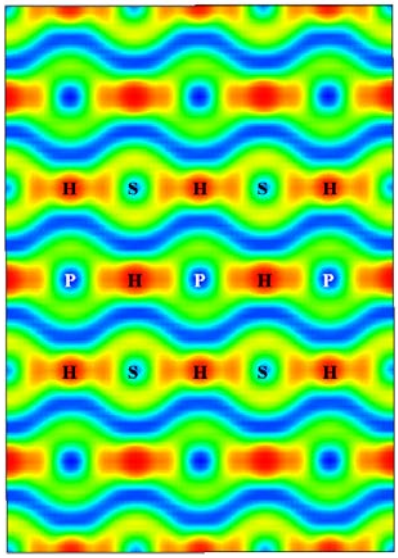

Fig. S7 The ELF of PSH_6 along (0,1,1) with 4*4*4 supercell.

5. Phonon structures of PSH_6

The cut-off energy of 80 Ry is used to calculate all phonon properties. In the calculation of electron-phonon coupling, we select 8 * 8 * 8 q-point grid to ensure the accuracy of the calculation results. The results show that PSH_6 can still maintain the dynamical stability of the structure in the pressure range of 0-200 GPa.

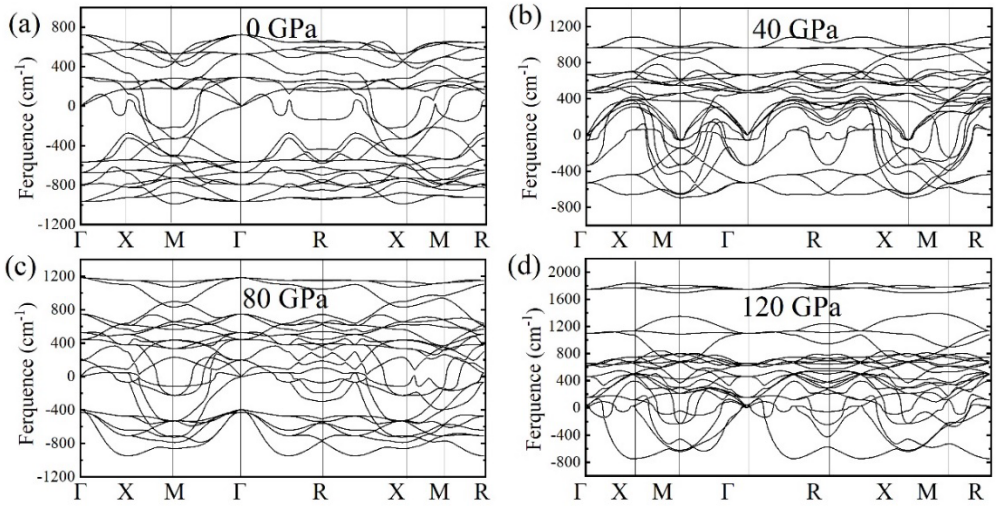

Fig. S8 Phonon spectra of PSH_6 along high symmetrical paths at selected pressures.

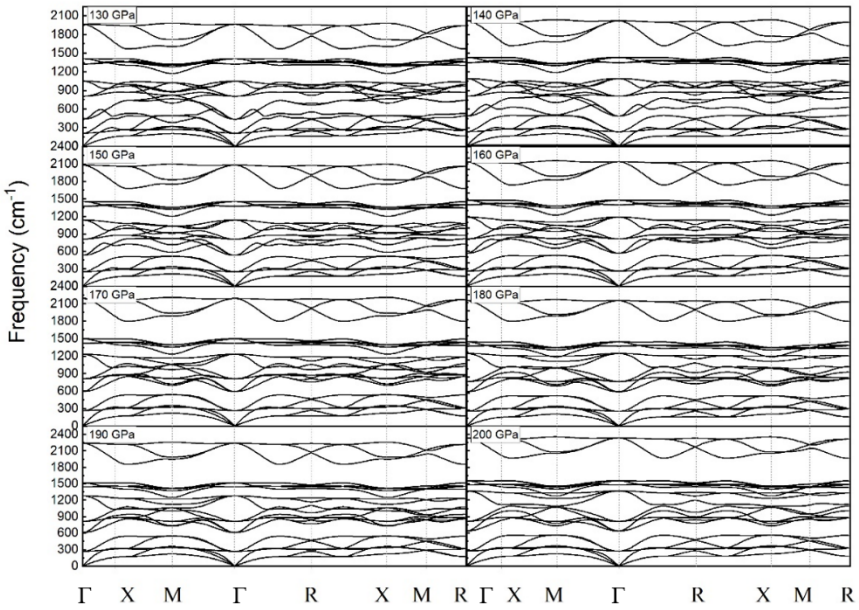

Fig. S9 Phonon spectra of PSH_6 along high symmetrical paths at selected pressures.

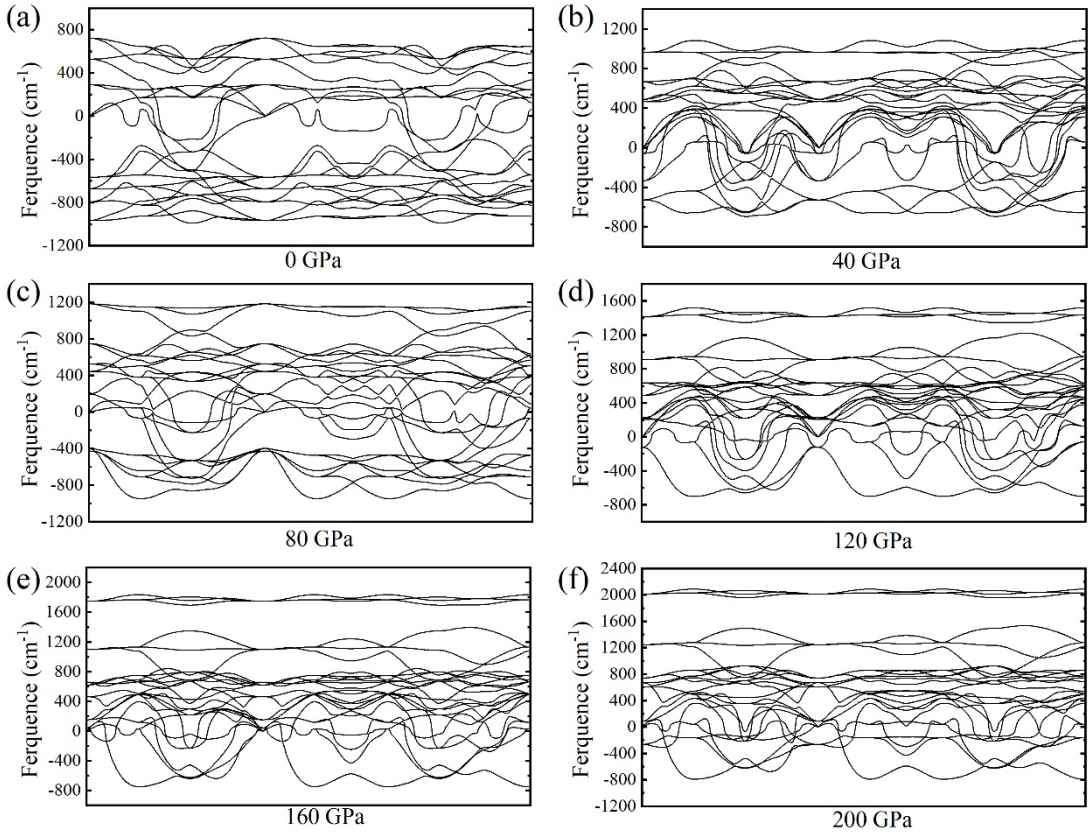

Fig. S10 Phonon spectra of PSH_6-Cmcm along high symmetrical paths "Γ-Y-C-Γ-Z-T-Y-Γ-S-R-Z-T" at selected pressures.

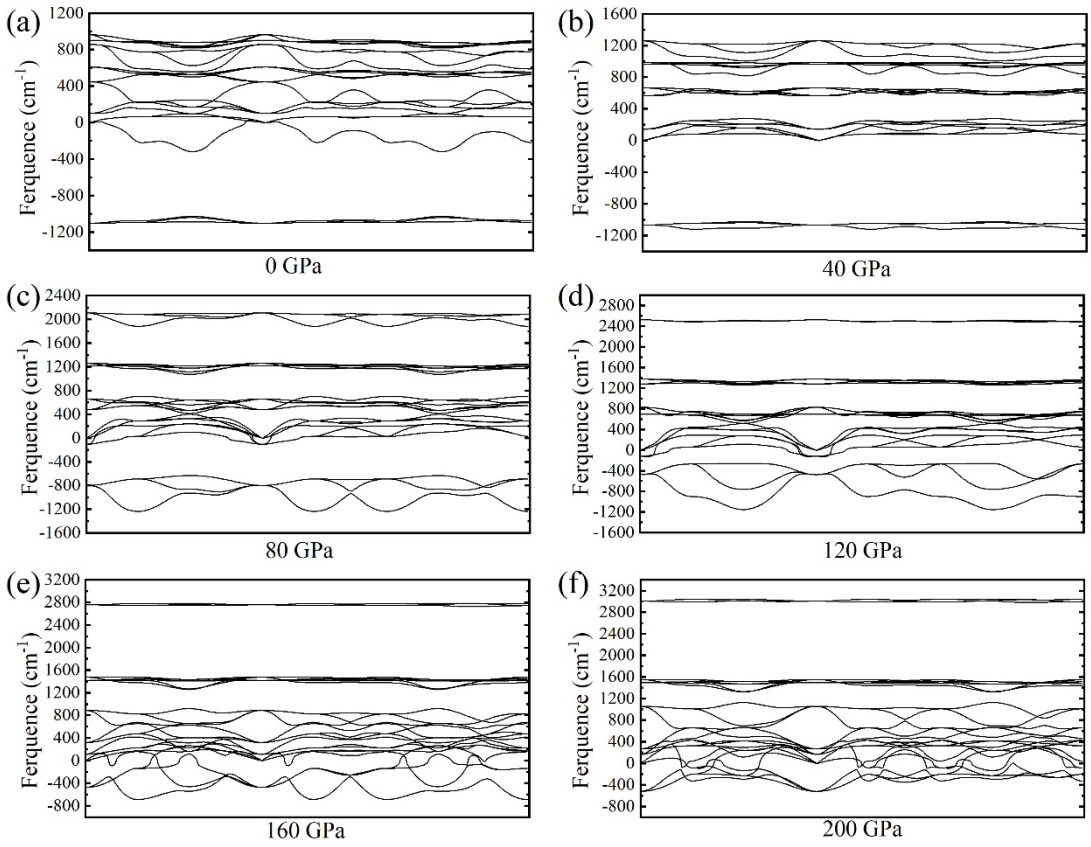

Fig. S11 Phonon spectra of PSH_6-Cmmm along high symmetrical paths "Γ-Y-C-Γ-Z-T-Y-Γ-S-R-Z-T" at selected pressures.

6. Cartesian coordinate of PSH_6

Pm-3m-PSH_6
a=*b*=*c*=2.98
P 0.5000000000000000 0.5000000000000000 0.5000000000000000
S 0.0000000000000000 0.0000000000000000 0.0000000000000000
H 0.0000000000000000 0.5000000000000000 0.5000000000000000
H 0.5000000000000000 0.0000000000000000 0.0000000000000000
H 0.5000000000000000 0.0000000000000000 0.5000000000000000
H 0.0000000000000000 0.5000000000000000 0.0000000000000000
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H 0.0000000000000000 0.0000000000000000 0.5000000000000000

7. T_c, λ, f_1, f_2, ω_{log}(K) of PSH_6

Table S1 T_c, λ, f_1, f_2, ω_{log} (K) of PSH_6.

GPa	T_c μ^*=0.13	f_1	f_2	T_c μ^*=0.1	f_1	f_2	λ	ω_{log} (K)
130	139.92	1.30	1.26	146.53	1.30	1.26	3.03	469.23
140	132.93	1.27	1.23	137.57	1.26	1.23	2.80	478.42
150	130.90	1.26	1.20	136.88	1.26	1.25	2.80	494.70
160	128.97	1.23	1.16	134.44	1.23	1.17	2.71	575.11
170	127.96	1.20	1.14	133.76	1.20	1.19	2.67	584.79
180	126.30	1.17	1.10	132.12	1.17	1.13	2.55	633.72
190	113.15	1.16	1.10	124.59	1.16	1.13	2.08	758.97
200	102.44	1.16	1.08	112.13	1.16	1.10	1.80	787.70

Table S2 Lattice length and bond length of SH_3, PSH_6 and PH_3.

P **(GPa)**	**Lattice length (*a*=*b*=*c*)**			**S-H**	**S-H(P-H)**	**P-H**
	SH_3	**PSH_6**	**PH_3**	**SH_3**	**PSH_6**	**PH_3**
0	3.6303	3.6415	3.6430	1.8210	1.8215	1.8217
40	3.3673	3.3696	3.3717	1.6837	1.6848	1.6858
80	3.2221	3.2242	3.2260	1.6110	1.6121	1.6130
120	3.1222	3.1236	3.1252	1.5611	1.5618	1.5626
160	3.1015	3.1028	3.1045	1.5226	1.5229	1.5237
200	3.1015	2.9835	2.9845	1.4907	1.4917	1.4922