Supporting Information for

Pressure-driven significant phonon mode softening and robust superconductivity in layered germanium phosphide

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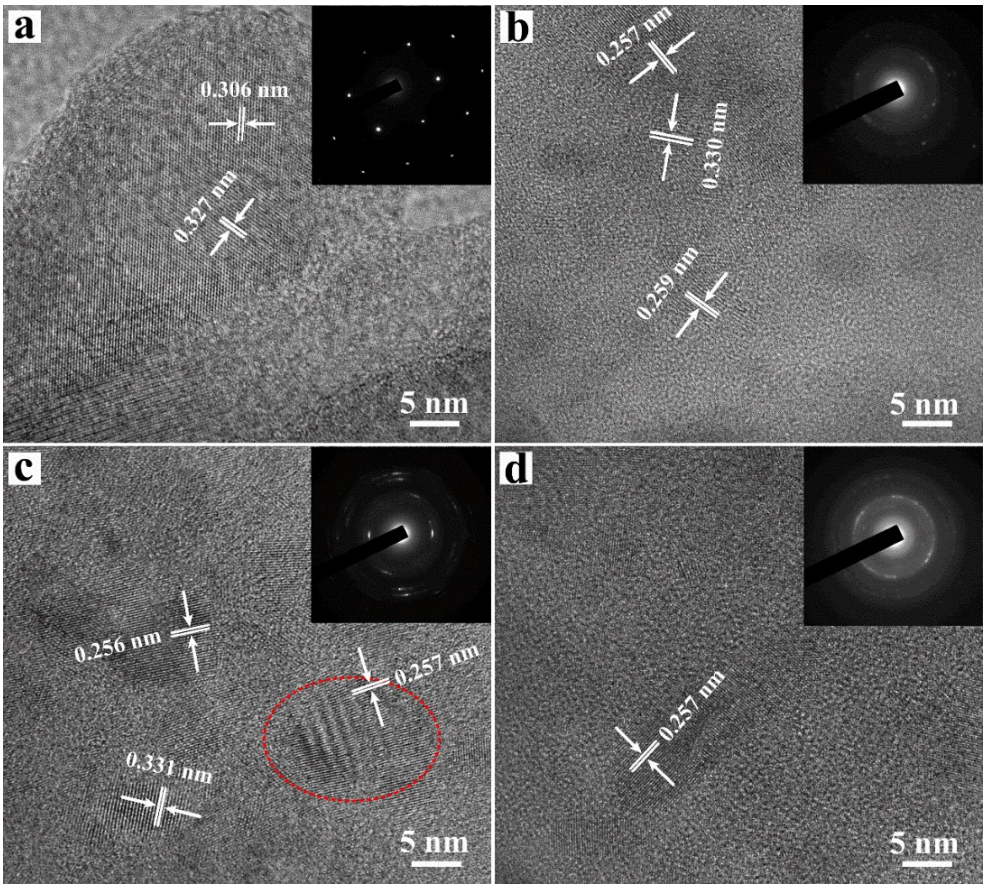

Fig. S1. Typical TEM and SEAD images for GeP_5 at ambient condition (a) and decompressed from 18.5 GPa (b), 53.5 GPa (c) and 60.3 GPa (d), indicating pressure-induced amorphous structures and deformation (shown in the red dashed area in c).

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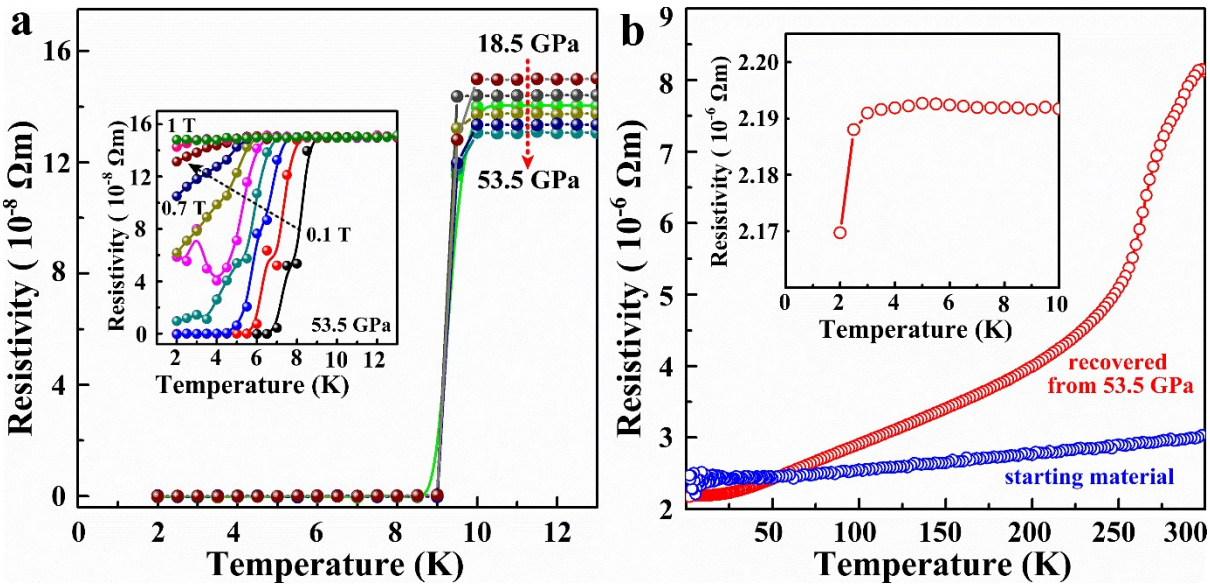

Fig. S2. (a) Electrical resistivity as a function of temperature in the range of 1.8-15 K at pressure from 18.5 GPa to 53.5 GPa. Inset image: temperature dependence of resistivity of GeP_5 under different magnetic fields up to 1 T at 53.5 GPa. (b) Resistance-pressure curves of ranging from 300 to 1.8 K of the starting materials (blue cycles) and the decompressed sample from 53.5 GPa (red cycles). Inset image: enlarged image for the decompressed sample.

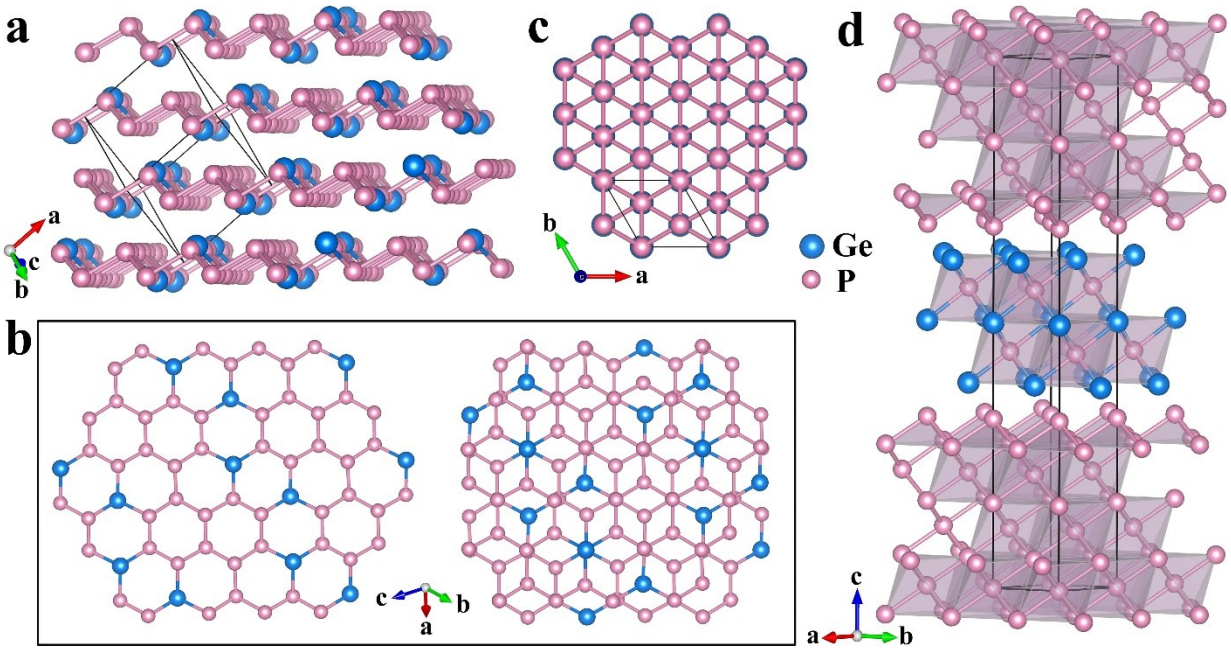

Fig. S3. Crystal structures of GeP_5 with space group $P\bar{1}$ (a and b) and $P\bar{3}m1$ (c and d). (b) shows the top views of one (left panel) and two Ge-P layers (right panel). Large blue and small pink spheres represent Ge and P atoms, respectively.

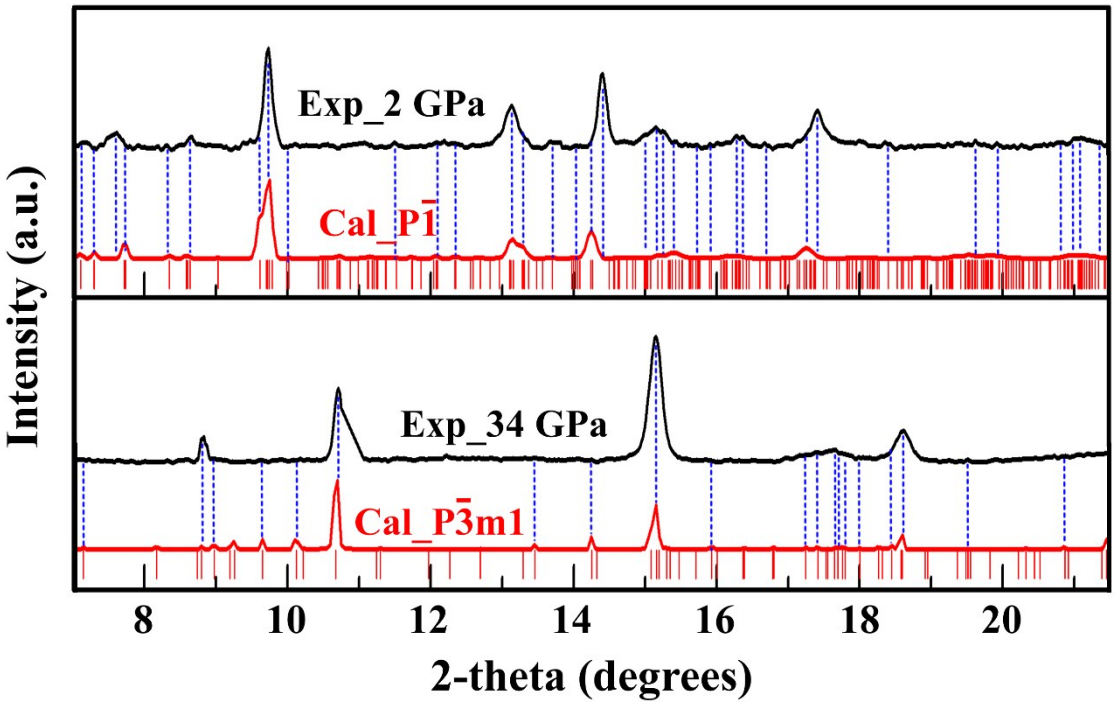

Fig. S4. Comparison of XRD patterns of the $P\bar{1}$ and $P\bar{3}m1$ structures along with the experimental data at 2 GPa and 34 GPa, respectively.

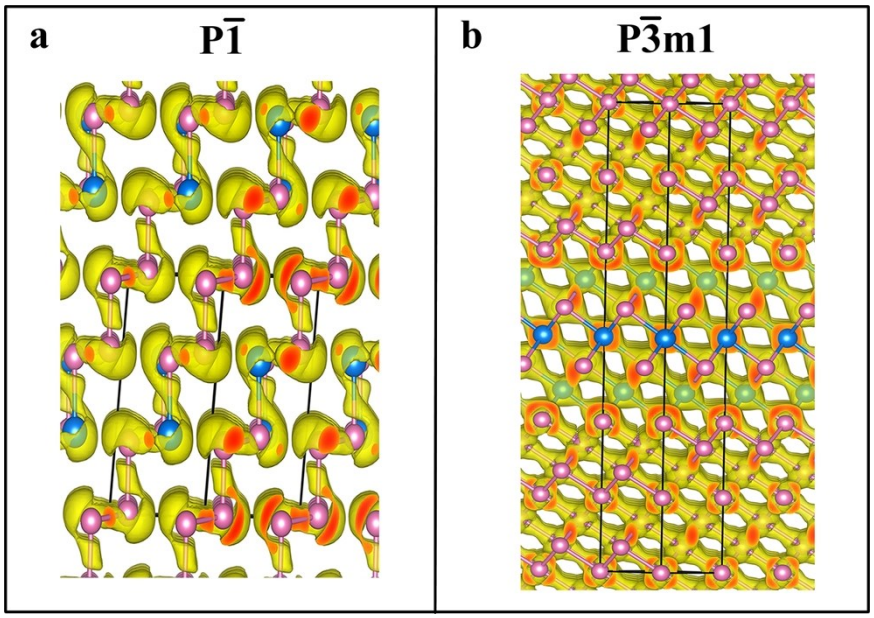

Fig. S5. Calculated valence electron localization functions (ELF) with isosurface value of 0.8 for the P$\bar{1}$ structure (a) at ambient pressure, and of 0.7 for the P$\bar{3}$m1 structure (b) at 16 GPa. The blue and pink spheres represent Ge and P atoms, respectively.

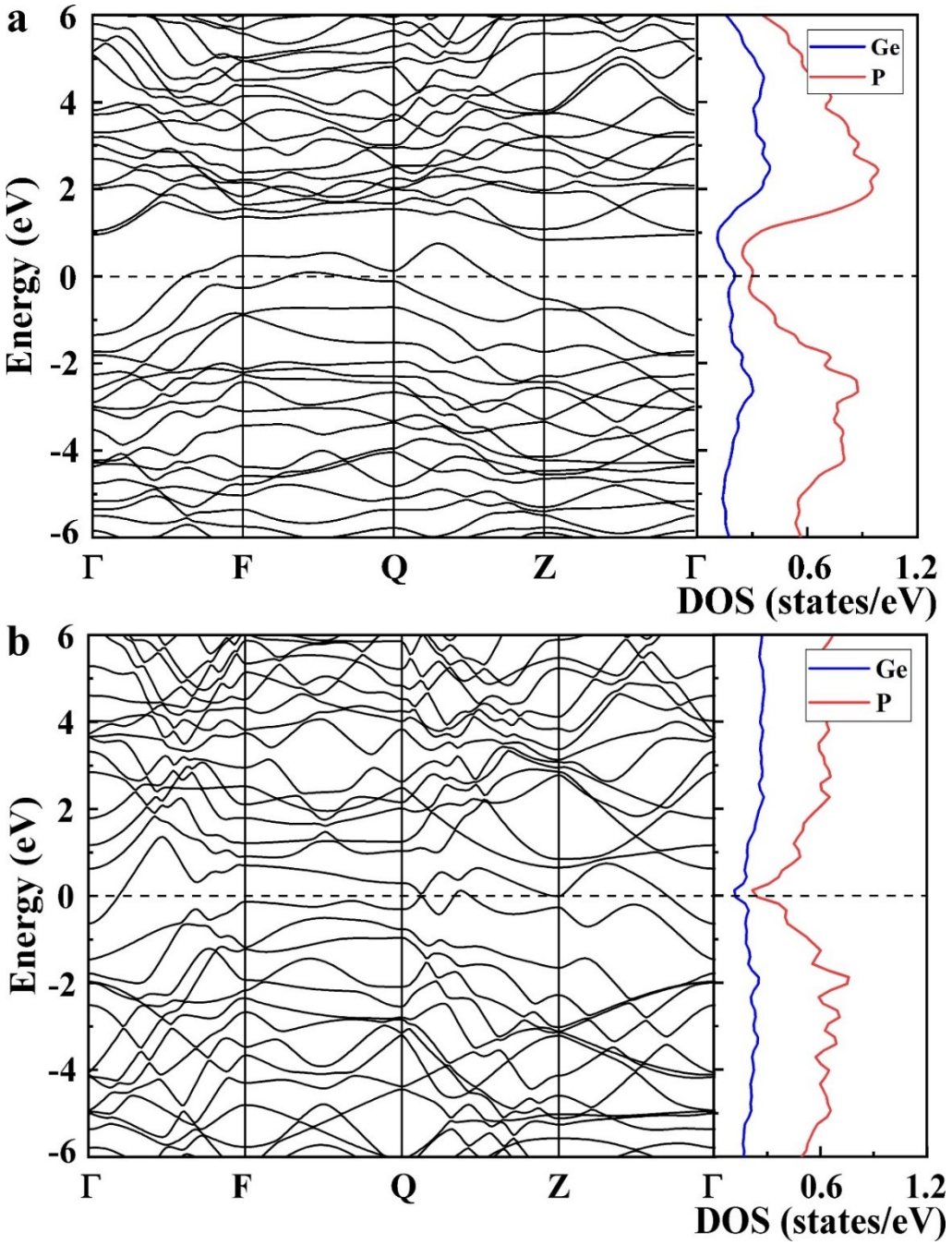

Fig. S6. The calculated band structures and partial electronic density of states (DOS) for low-pressure $P\bar{1}$ phase of GeP_5 at 0 (a) and 14 GPa (b).

Table S1. Calculated electron–phonon coupling (EPC) parameter λ, the logarithmic average phonon frequency ω_{log}, the density of states at the Fermi level per formula unit, $N(E_F)$, and the superconducting transition temperature T_c for the $P\bar{1}$ and $P\bar{3}m1$ structures at selected pressures from 0 to 60 GPa. μ^* of 0.1 was selected for the T_c calculation.

Phase		P (GPa)	λ	ω_{log} (K)	$N(E_f)$	T_c (K)
LP	$P\bar{1}$	0	0.40	223	8.37	0.9
		6	0.45	282	9.89	2.2
		10	0.57	302	10.44	5.8
		14	0.66	299	10.79	9.1
HP	$P\bar{3}m1$	16	0.73	297	12.80	11.5
		20	0.61	351	12.61	8.6
		30	0.54	388	12.39	6.1
		40	0.52	401	12.39	5.5
		60	0.49	401	12.31	4.4