Supporting Information

Sodalite-like carbon based superconductors with T_c about 77 K at ambient pressure

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1 Computational details

A. Details on Eliashberg equation

Because the superconducting pairing is only related to the electron bands near the Fermi energy, the energy shift $\chi(\overrightarrow{k}, i\omega_n)$ becomes 0, the T_c merely involves the determination of the renormalization function $Z(i\omega_n)$ and the order parameter $\phi(i\omega_n)$, which are calculated by the following Migdal-Eliashberg equations:[1,2]

$$Z(i\omega_n) = 1 + \frac{\pi T}{\omega_n N_F} + \sum_m \frac{\omega_m}{\sqrt{\omega_m^2 + \Delta^2(i\omega_m)}} \times \lambda(\omega_n - \omega_m)\delta(\varepsilon) \tag{S1}$$

$$\phi(i\omega_n) = Z(i\omega_n)\Delta(i\omega_m) + \frac{\pi T}{N_F}\sum_m \frac{\Delta(i\omega_n)}{\sqrt{\omega_m^2 + \Delta^2(i\omega_m)}} \times \left[\lambda(\omega_n - \omega_m) - N_F\mu^*\right]\delta(\varepsilon) \tag{S2}$$

In Eqs. S(1) and S(2), the N_F is the density of electronic states at the Fermi energy, $\Delta(i\omega_n)$ is superconducting gap, ε is electron and phonon energies, μ^* is the Coulomb pseudopotential, T is the absolute temperature, ω is the frequency, $i\omega_n$ is the nth fermion Matsubara frequencies with n=0, ±1,±2, ..., and $i\omega_n$ is given by

$$i\omega_n = i(2n+1)\pi T \tag{S3}$$

The function $\lambda(\omega_n\text{-}\omega_m)$ describes the effective electron-electron attraction via exchange of phonons, which is connected to Eliashberg EPC spectral function $\alpha^2F(\omega)$ through the relation:[3–5]

$$\lambda(\omega_n - \omega_m) = 2\int_0^\infty \frac{\alpha^2F(\omega)}{(\omega_n - \omega_m)^2 + \omega^2}\omega d\omega \tag{S4}$$

where $\alpha^2F(\omega)$ is defined as[3,5]

$$\alpha^2F(\omega) = \frac{1}{2\pi N_F}\sum_{qv}\frac{\gamma_{qv}}{\omega_{qv}}\delta(\omega - \omega_{qv}) \tag{S5}$$

where q is the wave vector, ω_{qv} is the screened phonon frequency and γ_{qv} is the phonon linewidth, which can be determined by[3,5]

$$\gamma_{qv} = 2\pi\omega_{qv}\sum_{mn}\sum_k \omega_k |g^v_{mn}(\mathbf{k},\mathbf{q})|^2 \delta(\varepsilon_{m,\mathbf{k}+\mathbf{q}} - \varepsilon_F) \times \delta(\varepsilon_{n,\mathbf{k}} - \varepsilon_F) \tag{S6}$$

The electron-phonon matrix elements $g^v_{mn}(\mathbf{k},\mathbf{q})$ are defined as[3,5]

$$g^{v}_{mn}(\mathbf{k},\mathbf{q}) = (\frac{\hbar}{2M\omega_{qv}})^{\frac{1}{2}}\langle m,\mathbf{k}+\mathbf{q}|\delta_{qv}V_{SCF}|n,\mathbf{k}\rangle \tag{S7}$$

In Eq. (S7), V_{SCF} is the self-consistent potential. The bare electronic Bloch state and ionic mass are labeled $|n,\mathbf{k}\rangle$ and M. Eqs. S(1) and S(2) established a coupled nonlinear system, from which we identify the renormalization function $Z(i\omega_n)$ and superconducting gap $\Delta(i\omega_n)$ by self-consistently calculations in Matsubara space with Coulomb pseudopotential μ^* =0.1. Then, the solutions of the Eliashberg equations on the real energy axis are performed by analytic continuation based on pade functions. The T_c is determined as the temperature at which the gap is equals to zero.

B. Details on Allen-Dynes modified McMillan equation

Based on a reanalysis of Eliashberg theory and newly available computational checks in superconductors, Allen and Dynes[3,5] developed and improved the McMillan equation. For $\lambda\leq$ 1.5 they point out that the T_c can be obtained by solving the following equation:

$$T_c = \frac{\omega_{log}}{1.2}\exp\left[-\frac{1.04(1+\lambda)}{\lambda-\mu^*(1+0.62\lambda)}\right] \tag{S8}$$

where ω_{log} is the logarithmic averaged frequency, can be calculated as

$$\omega_{log} = \exp[\frac{2}{\lambda}\int_0^\infty \frac{d\omega}{\omega}\alpha^2F(\omega)\ln(\omega)] \tag{S9}$$

For λ>1.5, the correction factors f_1 and f_2 were introduced and the Eq. (S8) was modified as:

$$T_c = \frac{f_1 f_2\omega_{log}}{1.2}\exp\left[-\frac{1.04(1+\lambda)}{\lambda-\mu^*(1+0.62\lambda)}\right] \tag{S10}$$

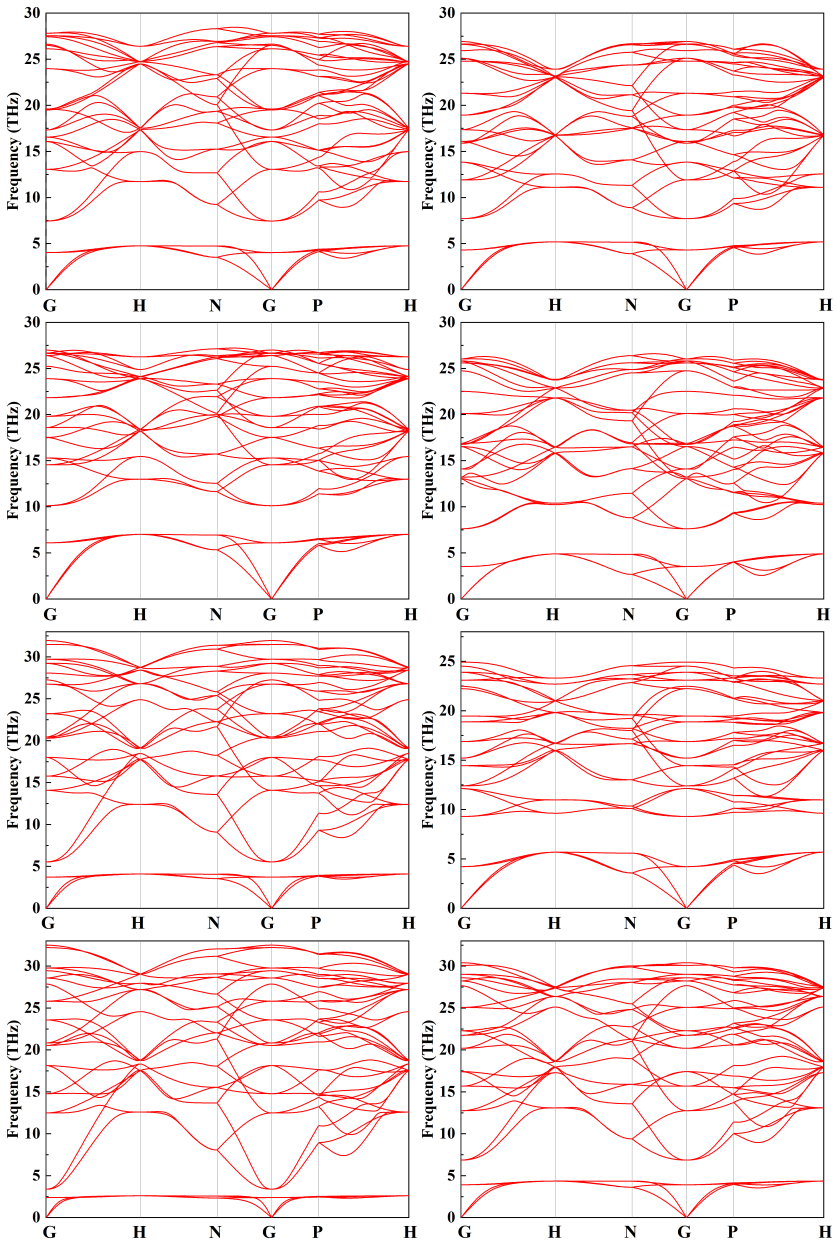
where the correction factors f_1 and f_2 are given by:

$$f_1 = \sqrt[3]{\left[1+(\frac{\lambda}{2.46(1+3.8\mu^*)})\right]^{3/2}}, \qquad f_2 = 1+\frac{(\frac{\bar{\omega}^2}{\omega_{log}}-1)\lambda^2}{\lambda^2+\left[1.82(1+6.3\mu^*)\frac{\bar{\omega}^2}{\omega_{log}}\right]^2} \tag{S11}$$

In Eq. (S11), the mean square frequency is defined as:

$$\bar{\omega} = \sqrt{\frac{2}{\lambda}\int\alpha^2F(\omega)\omega d\omega} \tag{S12}$$

2 Supplemental Figures and Tables



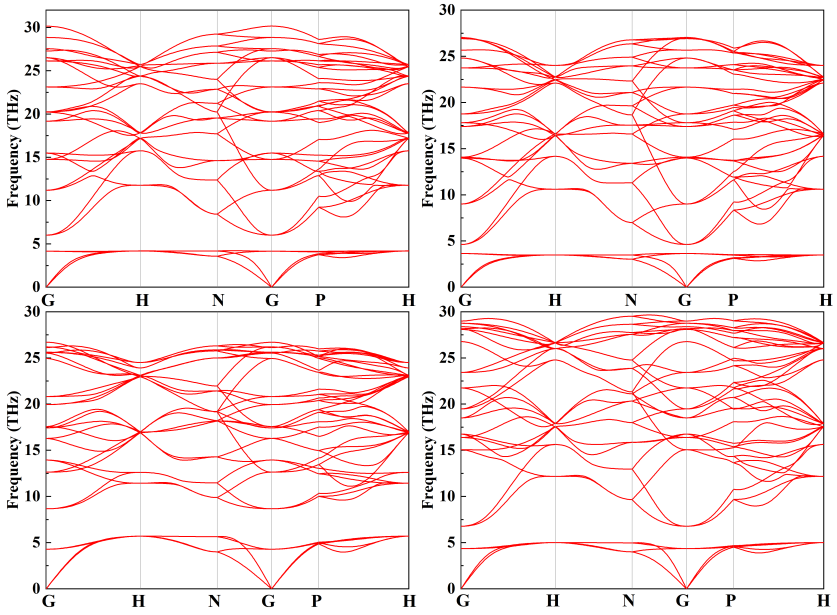


Fig. S1 Calculated phonon dispersion curves of XC_6 (X = Ag, As, Br, Cd, Cu, I, Ni, Pd, Rh, Ru, Se and Zn).

Table S1: The bond length of C-C and X-C (X = Ga and Ge), Bader charge analysis for C and X, negative integrated COHP (-ICOHP) of C-C and X-C, and electronic density of states at the fermi level $N(E_f)$ of GaC_6 and GeC_6.

Parameter		GaC_6	GeC_6
Distance (Å)	C-C	1.64	1.65
	X-C	2.59	2.60
Bader charge	C	4.12	4.13
	X	2.26	3.20
-ICOHP	C-C	7.44	7.85
(eV/atom pair)	X-C	0.61	0.56
$N(E_f)$		2.75	4.55
Lattice constants		a=4.63896Å	a=4.65532Å

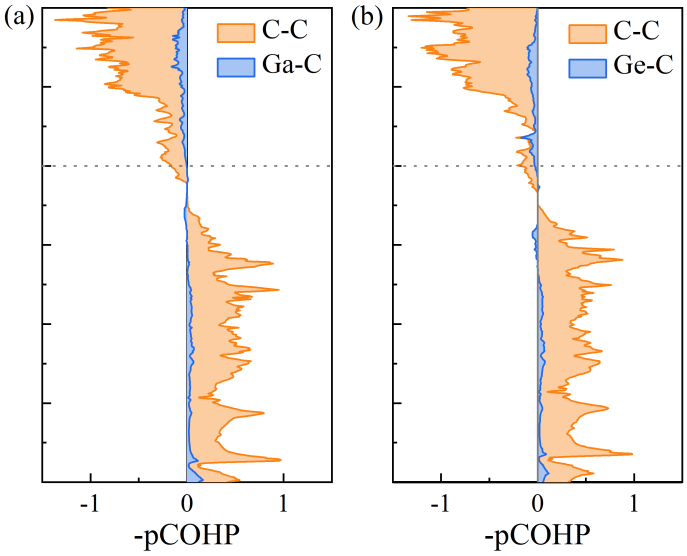


Fig. S2 Negative projected crystal orbital Hamiltonian population (-pCOHP) of (a) GaC_6 and (b) GeC_6.

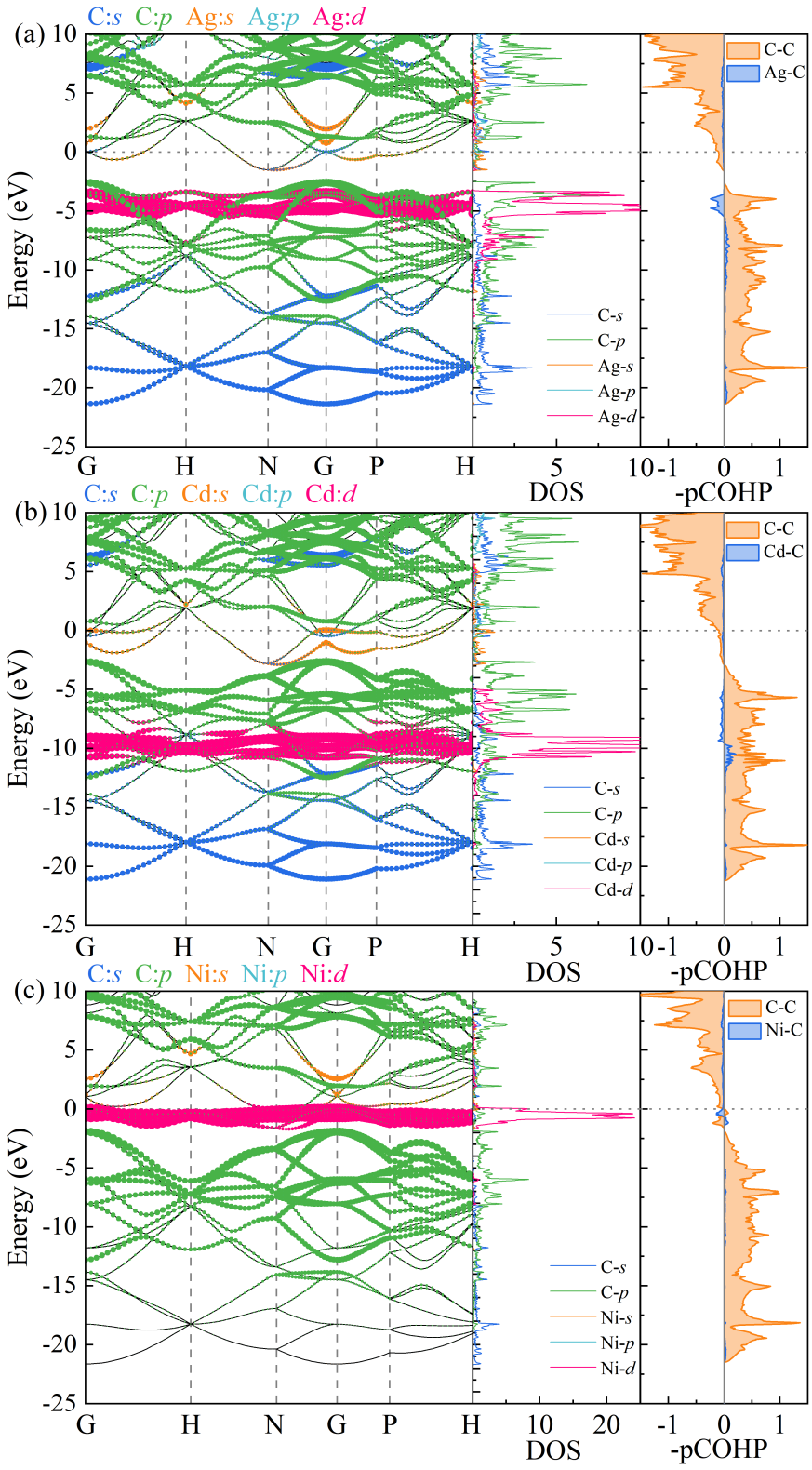


Fig. S3 Calculated band structures, electronic density of states, COHP analysis of (a) AgC_6, (b) CdC_6 and (c) NiC_6.

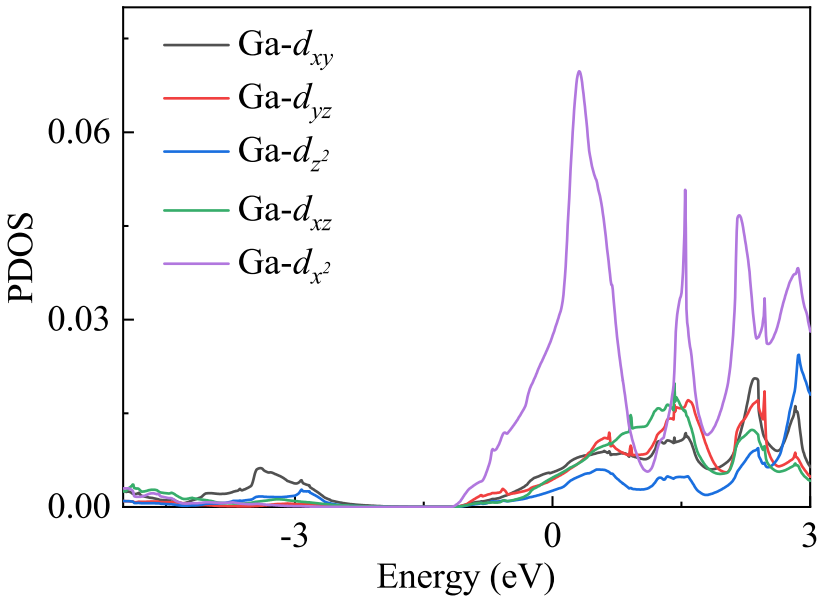


Fig. S4 The projected DOS (PDOS) for Ga *d*-orbitals.

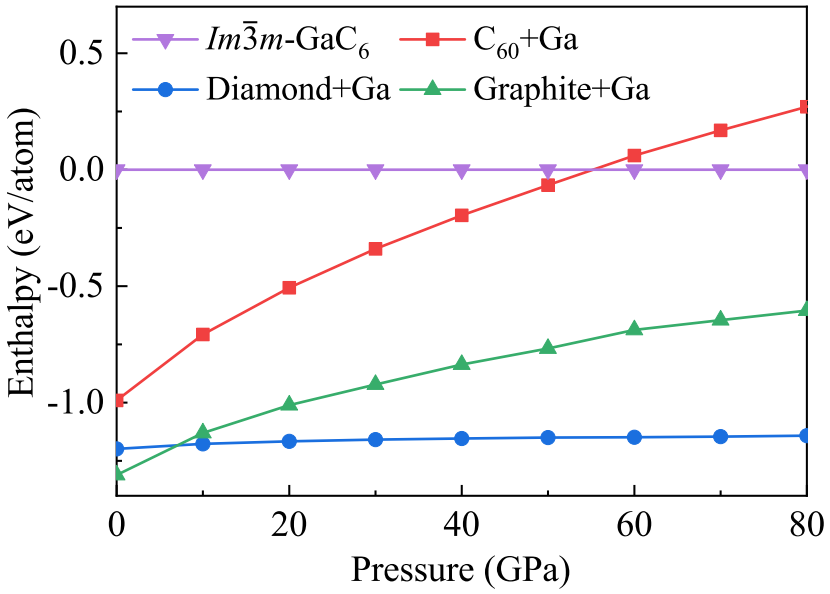


Fig. S5 The enthalpies per atom and decomposition enthalpies as a function of pressure for $Im\bar{3}m$-GaC_6.

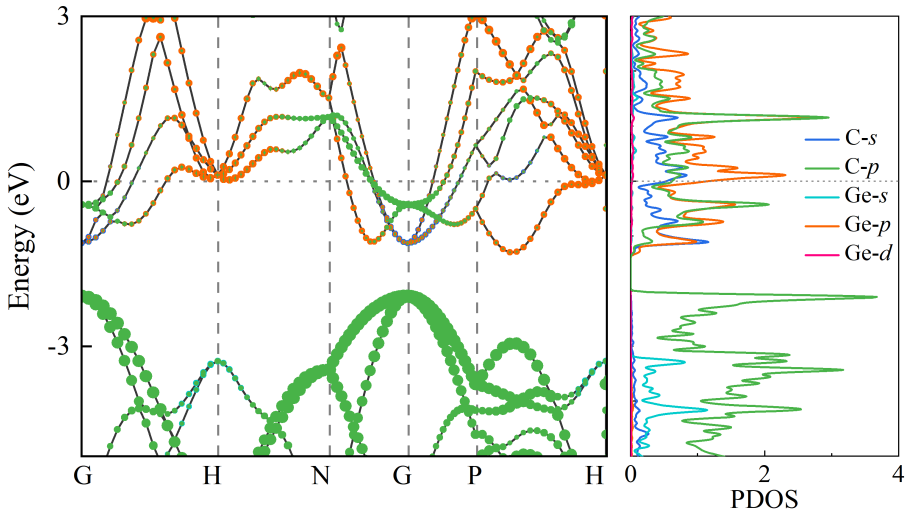


Fig. S6 Calculated band structures and electronic density of states of GeC_6.

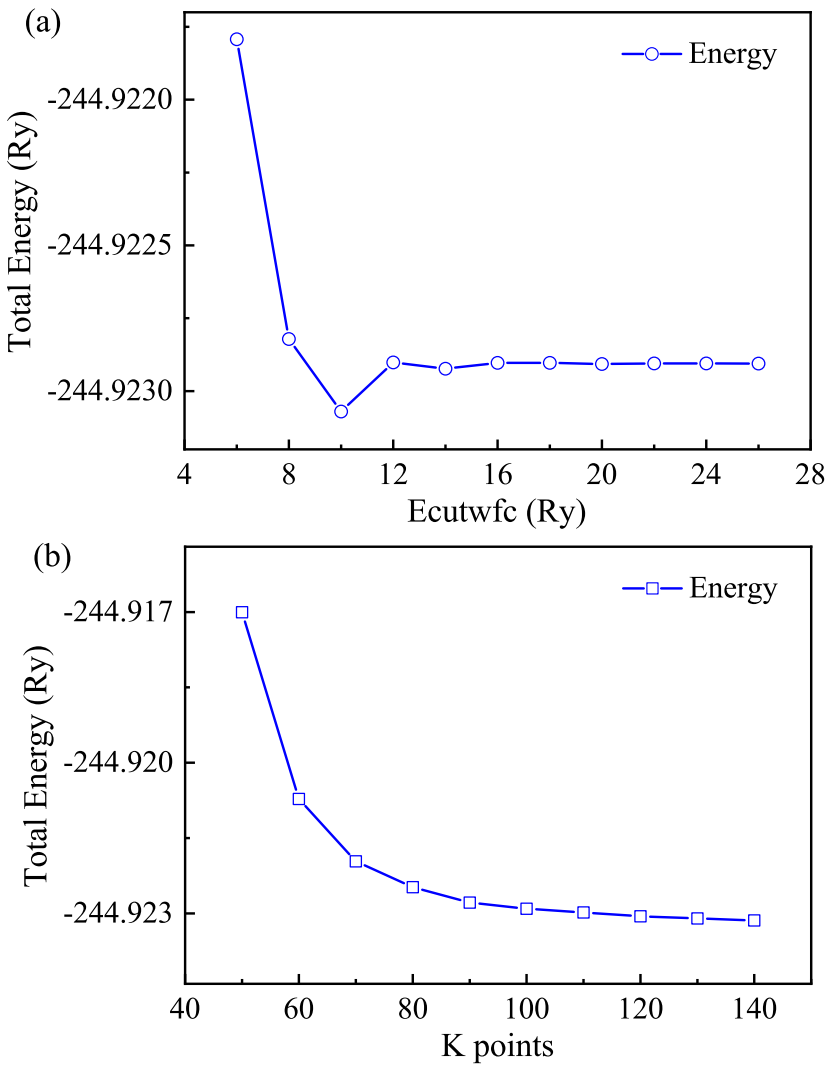


Fig. S7 The convergence tests for the energy of $Im\bar{3}m$-GaC_6 at ambient pressure with (a) the cutoff energy and (b) the k-points sampling, respectively.

Table S2: The bond length, Bader charge, electronic density of states at the fermi level N(E_f), electron-phonon coupling (EPC) parameters λ, logarithmic average phonon frequency ω_{log}, superconducting critical temperatures T_c, negative integrated COHP (-ICOHP) and lattice constants of XC_6 (X = Ag, Se, Br, I, Ag and Zn). The Mc-A-D, G-K and IE correspond to the T_c obtained by the Allen-Dynes modified McMillan equation, the Gor'kov and Kresin equation and the isotropic Eliashberg equation, respectively.

		AsC_6	SeC_6	BrC_6	IC_6	AgC_6	ZnC_6
Distance (Å)	C-C	1.65	1.66	1.66	1.71	1.64	1.63
	X-C	2.61	2.62	2.63	2.71	2.60	2.57
Bader charge	C	4.09	4.04	4.01	4.08	4.08	4.09
	X	4.43	5.73	6.92	6.55	10.5	11.46
N(E_f)		3.55	3.94	11.84	7.12	1.95	1.98
EPC	ω_{log}, K	420.87	516.35	701.03	524.11	646.34	755.70
	λ	1.02	0.61	0.45	0.60	0.74	0.68
	T_c (Mc-A-D), K	30	12	5	12	26	24
	T_c (G-K), K	46	14	4	10	38	37
	T_c (IE), K	35	13	6	14	37	35
-ICOHP	C-C	7.94	7.58	8.00	7.22	7.79	7.72
(eV/atom pair)	X-C	0.46	0.33	0.25	0.34	0.19	0.10
Lattice constants (Å)		a=4.66200	a=4.68112	a=4.70170	a=4.84309	a=4.64989	a=4.59934

Table S3: The bond length, Bader charge, electronic density of states at the fermi level N(E_f), electron-phonon coupling (EPC) parameters λ, logarithmic average phonon frequency ω_{log}, superconducting critical temperatures T_c, negative integrated COHP (-ICOHP) and lattice constants of XC_6 (X = Cd, Ni, Cu, Ru, Rh and Pd). The Mc-A-D, G-K and IE correspond to the T_c obtained by the Allen-Dynes modified McMillan equation, the Gor'kov and Kresin equation and the isotropic Eliashberg equation, respectively.

		CdC_6	NiC_6	CuC_6	RuC_6	RhC_6	PdC_6
Distance (Å)	C-C	1.67	1.59	1.61	1.62	1.62	1.63
	X-C	2.64	2.52	2.54	2.57	2.57	2.57
Bader charge	C	4.11	4.09	4.09	4.1	4.08	4.07
	X	11.33	9.45	10.49	7.37	8.52	9.60
N(E_f)		2.82	10.87	0.80	9.10	3.65	
EPC	ω_{log}, K	670.64	241.21	529.10	240.62	449.31	
	λ	0.58	0.86	0.60	0.86	0.63	
	T_c (Mc-A-D), K	14	13	12	13	12	
	T_c (G-K), K	14	15	11	11	13	
	T_c (IE), K	16	14	10	12	15	
-ICOHP	C-C	7.29	8.04	8.48	7.73	7.75	7.39
(eV/atom pair)	X-C	0.14	0.21	0.17	0.44	0.34	0.11
Lattice constants (Å)		a=4.71477	a=4.51025	a=4.54181	a=4.59235	a=4.59384	a=4.60279

References

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[4] I. A. Kruglov, D. V. Semenok, H. Song, R. Szczesniak, I. A. Wrona, R. Akashi, M. M. Davari Esfahani, D. Duan, D. Cui, A. G. Kvashnin and A. R. Oganov, *Phys. Rev. B*, 2020, **101**, 024508.

[5] P. B. Allen, *Phys. Rev. B*, 1972, **6**, 2577.