Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supplementary Information

Pressure-induced superconducting CS_2H_{10} with H_3S framework

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FIGURES

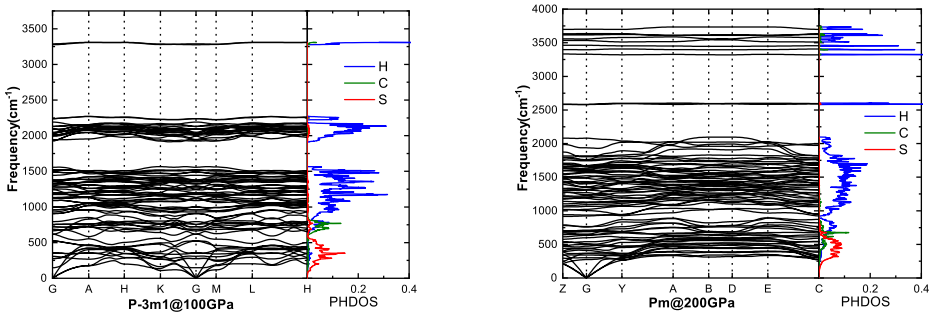


Fig. S1 The phonon dispersion curves and phonon density of states (PHDOS) of *P*-3*m*1 and *Pm* at different pressures.

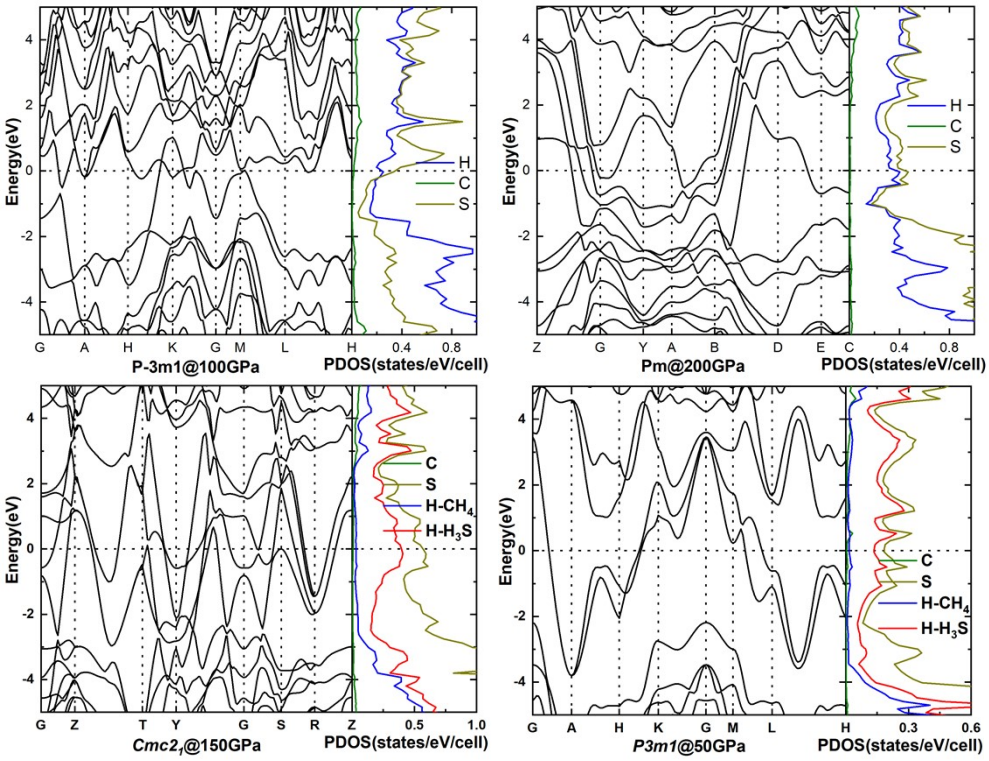


Fig. S2 The electronic band structures and PDOS of *P*-3*m*1, *Pm*, *Cmc*2$_1$ and *P*3*m*1 at different pressures.

TABLES

Table S1 Lattice parameters and atomic positions of $Cmc2_1$, $P3m1$, $P\text{-}3m1$ and Pm phases at different pressures.

Structure of CS_2H_{10}	Parameters (Å, deg)	Atom	x	y	z
$Cmc2_1$ (150 GPa)	a=b= 5.0732	H	-0.53941	0.06501	-0.43604
	c= 4.2955	H	-0.15952	0.33346	-0.84568
	α=β= 90	H	-0.33996	0.16820	-0.84831
	γ= 49.8075	H	-0.12439	0.87561	-0.10929
		H	-0.55998	0.44002	-0.59198
		H	-0.23214	0.76786	-0.61037
		H	-0.29158	0.70842	-0.28471
		C	-0.23823	0.76177	-0.06931
		S	-0.41412	0.58588	-0.60062
		S	-0.07965	0.92035	-0.59928
$P3m1$ (50 GPa)	a=b= 4.7290	H	0.85970	0.14030	0.95335
	c= 2.8440	H	0.21072	0.78928	0.20212
	α=β= 90	H	0.53725	0.07449	0.62299
	γ= 120.0000	H	0.33333	0.66667	0.69066
		C	0.33333	0.66667	0.06473
		S	0.66667	0.33333	0.30778
		S	1.00000	1.00000	0.63792
$P\text{-}3m1$ (100 GPa)	a=b= 5.8103	H	0.08312	0.37174	0.31215
	c= 2.7549	H	0.22373	0.11187	0.28652
	α=β= 90	H	0.66667	0.33333	0.58654
	γ= 120.0000	C	0.66667	0.33333	0.20404
		S	0.50000	0.00000	0.00000
		S	0.00000	0.00000	0.00000
Pm (200 GPa)	a= 6.9823	H	0.81047	0.69036	0.04784
	b= 4.4790	H	0.88363	0.74296	0.55578
	c= 3.1308	H	0.17630	0.70927	0.70906
	α=γ= 90	H	0.20833	0.24000	0.21629
	β= 102.9245	H	0.49729	0.19383	0.83501
		H	0.56975	0.21541	0.39789
		H	0.36898	0.50000	0.78034
		H	0.03059	0.50000	0.12223
		H	0.59465	0.50000	0.07694
		H	0.68220	0.50000	0.59743
		H	0.72399	0.00000	0.93378
		H	0.29172	0.00000	0.57489
		H	0.36428	0.00000	0.14275

		H	0.05633	0.00000	0.61830
		C	0.72193	0.50000	0.94374
		C	0.41233	0.00000	0.84683
		S	0.36374	0.50000	0.30124
		S	0.03226	0.50000	0.62806
		S	0.71467	0.00000	0.45929
		S	0.04795	0.00000	0.13040

Equations for calculating T_c and related parameters

(1) The Allen−Dynes-modified McMillan equation

T_c can be estimated by the McMillan equation[1]:

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

where λ and ω_{log} are the electron−phonon coupling constant and the logarithmic-averaged phonon frequency, respectively, and μ^* is the Coulomb pseudopotential, for which we use the widely accepted range of 0.1-0.13. λ and ω_{log} are given by

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

and

$$\omega_{log} = exp\left(\frac{2}{\lambda} \int_0^\infty \frac{d\omega}{\omega} \alpha^2 F(\omega) \ln \omega \right) \tag{3}$$

The parameter ω denotes the phonon frequency, and $\alpha^2F(\omega)$ is the Eliashberg spectral function

$$\alpha^2 F(\omega) = \frac{1}{2\pi N(\varepsilon_F)} \sum_{q\upsilon} \frac{\gamma_{q\upsilon}}{\omega_{q\upsilon}} \delta(\omega - \omega_{q\upsilon}) \tag{4}$$

The line width $\gamma_{q,\upsilon}$ is written as

$$\gamma_{q\upsilon} = \pi \omega_{q\upsilon} \sum_{mn} \sum_{k} \left| g_{mn}^{\upsilon}(k,q) \right|^2 \delta(\varepsilon_{m,k+q} - \varepsilon_F) \times \delta(\varepsilon_{n,k} - \varepsilon_F) \tag{5}$$

where $\varepsilon_{n,\mathbf{k}}$ is the energy of the bare electronic Bloch state, ε_F is the Fermi energy, and $g_{mn}^{\upsilon}(k,q)$ is the electron−phonon matrix element.

When the value of λ larger than 1.3, strong-coupling corrections begin to appear. Therefore, P. B. Allen and R. C. Dynes use two separate correction factors (f_1 and f_2) to describe these two effects. Then we can further obtain the Allen−Dynes-modified McMillan equation[2]:

$$T_c = \frac{f_1 f_2 \omega_{log}}{1.2} exp\left[-\frac{1.04(1+\lambda)}{\lambda - \mu^*(1+0.62\lambda)} \right] \tag{6}$$

f_1 and f_2 are given by

$$f_1 = \sqrt[3]{\left[1 + \left(\frac{\lambda}{2.46(1+3.8\mu^*)} \right)^{\frac{3}{2}} \right]} \tag{7}$$

and

$$f_2 = 1 + \frac{\left(\frac{\omega_2}{\omega_{log}} - 1 \right) \lambda^2}{\lambda^2 + \left[1.82(1+6.3\mu^*) \frac{\bar{\omega}_2}{\omega_{log}} \right]} \tag{8}$$

average frequencies $\bar{\omega}_2$ is given by

$$\bar{\omega}_2 = \sqrt{\frac{2}{\lambda}\int_0^{\infty}\frac{d\omega}{\omega}\alpha^2F(\omega)\omega d\omega} \tag{9}$$

(2) Self-consistent solution of the Eliashberg equation

For strong-coupling system, it can be better described with Eliashberg equation[3]:

$$Z(i\omega_n)\Delta(i\omega_n) = \frac{\pi T}{N_F}\sum_{n'}\frac{\Delta(i\omega_{n'})}{\sqrt{\omega_{n'}^2 + \Delta^2(i\omega_{n'})}} \times [\lambda(\omega_n - \omega_{n'}) - N_F\mu^*]\delta(\epsilon) \tag{10}$$

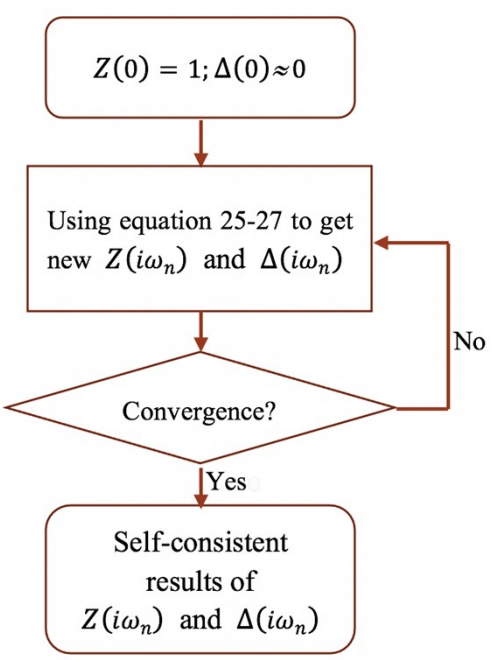
$$Z(i\omega_n) = 1 + \frac{\pi T}{N_F\omega_n}\sum_{n'}\frac{\omega_{n'}}{\sqrt{\omega_{n'}^2 + \Delta^2(i\omega_{n'})}}\lambda(\omega_n - \omega_{n'})\delta(\epsilon) \tag{11}$$

where functions $Z(i\omega_n)$ and $\Delta(i\omega_n)$ are the renormalization function and pairing order parameter, respectively. N_F is the density of electronic states at the Fermi level, and $\delta(\epsilon)$ is the Dirac delta function. $i\omega_n = i(2n+1)\pi T_c$ are the fermion Matsubara frequencies. μ^* is the Coulomb pseudopotential, for which we use the widely accepted range of 0.1 - 0.13. $\lambda(\omega_n - \omega_{n'})$ contains the electron-phonon coupling matrix, phonon propagator, and the phonon density of states, and is given by:

$$\lambda(\omega_n - \omega_{n'}) = \int_0^{\infty} d\omega\frac{2\omega}{(\omega_n - \omega_{n'})^2 + \omega^2}\alpha^2F(\omega) \tag{12}$$

The equations for the $Z(i\omega_n)$ and $\Delta(i\omega_n)$ form a coupled nonlinear system and are solved self-consistently. We evaluated renormalization function and the order parameter for each Matsubara frequency along the imaginary energy axis. After calculating $Z(i\omega_n)$ and $\Delta(i\omega_n)$, an analytic continuation is performed to the real axis using Pade' functions.

The specific process is as follows:



References

1. W. L. McMillan, *Physical Review*, 1968, **167**, 331.
2. P. B. Allen and R. C. Dynes, *Phys. Rev. B*, 1975, **12**, 905-922.
3. G. M. Eliashberg, *Sov Phys Jetp*, 1960, **11:3**, 696-702.