Supplementary Materials for

Strong Electron-Phonon Interaction Retarding Phonon Transport in Superconducting Hydrogen Sulfide at High Pressures

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S1. Lattice Structure

Fig. S1 shows the $Im\bar{3}m$ lattice structure of high-$T_c$ hydrogen sulfide (H$_3$S) at high pressures which contains two interpenetrating SH$_3$ perovskite sublattices. The unit cell contains 8 atoms and the relaxed lattice constant at 200 GPa is 2.983 Å, slightly smaller than the literature value of 3.089 Å.$^1$ The contact distance between hydrogen atoms is approximately 1.5 Å and very short compared with the van der Waals radii sum of 2.4 Å.

S2. Computational Details

We employed the Vienna \textit{ab initio} simulation package (VASP)$^2$ using the projector augmented wave (PAW) method$^3$ to compute harmonic as well as anharmonic interatomic force constants (IFCs) in real space and then calculate phonon dispersion relation and phonon relaxation time. The Perdew-Burke-Ernzerhof (PBE) exchange-correlation functional$^4, 5$ was adopted and the energy cutoff was set as 500 eV. For the harmonic IFCs calculations, a large $3\times3\times3$ supercell containing 216 atoms was built and atoms were displaced from the equilibrium position by 0.02
Å. During the computations of cubic IFCs, the large 4×4×4 supercell containing 512 atoms was chosen and a cutoff radius of 0.54 nm was introduced which is equivalent to up to the 12th nearest neighbor. After collecting the harmonic and cubic IFCs, the phonon relaxation time ($\tau_{ph}$) and lattice thermal conductivity ($\kappa_L$) were computed with the ShengBTE package. During the calculations of $\tau_{ph}$ and $\kappa_L$, the 15×15×15 $q$ grid was employed and the Gaussian smearing with breadth of 1 was applied to regularize the Dirac delta function to reduce computation cost but keep reasonable accuracy. To perform the electron-phonon interaction calculations, we used the all-electron full-potential linearised augmented-plane wave (FP-LAPW) ELK code. Since there exists strong anharmonicity in the hydrogen sulfide at high pressures, the supercell method was applied to include anharmonicity into phonon dispersion. The phonon calculations were performed with the $q$-mesh of 4×4×4 and $k$-mesh of 72×72×72.

S3. Phonon Transport by Phonon-Phonon Interaction

To explain why there exist large difference in the $\kappa_L$ of H$_3$S at varying high pressures, the important quantities of specific heat, the averaged square group velocity and phonon relaxation time in Eq. (1) of the main text for H$_3$S at varying pressures are fully compared and analyzed. In Figs. S2 and S3, we do not observe a noticeable change in the specific heat and averaged square group velocity at varying pressures. However, in Fig. S4, taking the temperature of 200 K for instance, it is clearly demonstrated that the phonon relaxation time for phonons below 10 THz at 175 GPa is almost one order of magnitude smaller than that of 200 GPa, 225 GPa and 250 GPa. Further analysis on the cumulative $\kappa_L$ with respect to frequency, as presented in Fig. S5, shows that the phonons below 10 THz contribute approximately 80% to the total phonon transport and are thus the major heat-carrying phonons. At 175 GPa, the $T_c$ of H$_3$S is smaller than 100 K and it
is in the normal state at high temperatures as much as 200 K. However, at 200 GPa or higher, the
$T_c$ is larger than 200 K and H$_3$S is in the superconducting state at 200 K. Thus, we can conclude
that the phonon-phonon scattering in the superconducting state is much weaker than that of
normal state.

FIG. S2. The temperature-dependent specific heat of H$_3$S at high pressures varying from 175
GPa to 250 GPa.
FIG. S3. The averaged square group velocity of H$_3$S at high pressures varying from 175 GPa to 250 GPa.
FIG. S4. The phonon relaxation time of H$_3$S due to only PPI at 200 K at high pressures varying from 175 GPa to 250 GPa. The red circle indicates that the relaxation time for phonons below 10 THz at 175 GPa is one order of magnitude smaller than that at higher pressures.
FIG. S5. The cumulative $\kappa_L$ of H$_3$S with respect to frequency at 200 K and high pressures varying from 175 GPa to 250 GPa.

S4. Electron-Phonon Interaction under External Magnetic Field

The inclusion of external magnetic field is achieved within the framework of spin density functional theory.$^9,^{10}$ In presence of external magnetic field $B_{\text{ext}} = \nabla \times A_{\text{ext}}$, the Kohn-Sham equation describing the non-interacting electrons states as$^9,^{10}$

$$\left(-\frac{1}{2}\nabla^2 + v + \mu \sigma \cdot B\right)\Phi_i(r) = \varepsilon_i \Phi_i(r).$$
The noncollinear electron density $\rho$ and magnetization density $m$ are calculated to describe the total energy $E[\rho, m]^{9,10}$

$$E[\rho, m] = T_s[\rho, m] + V_{ext}[\rho] + \mathbf{B}_{ext} \cdot \mathbf{m} + U[\rho] + E_{xc}[\rho, m], \ \text{\textcopyright S2}$$

where $T_s$, $V_{ext}$ and $U$ is the kinetic energy, external potential and Hartree energy, respectively, $\mu_B$ is magnetic permeability and $\sigma$ is the vector of Pauli matrices and. To solve this Kohn-Sham equation, a two-step variational process was proposed: (1) at the first step, the Hamiltonian containing only the scalar potential $v_s$ is solved and the scale states serve as the basis for the second step; (2) at the second variational step, the spin-orbital coupling and magnetic terms are included. By solving those equations, the two-component spinors ($\Phi$), noncollinear density ($\rho$) and magnetization density ($m$) under external magnetic field can be obtained and further applied to compute relevantly physical quantities.

Fig. S6 shows the magnetic field dependent $\alpha^2 F$ of superconducting hydrogen sulfide at 200 GPa. As external magnetic field is strengthened, it is demonstrated that the electron-phonon coupling strength for the high-frequency phonons slightly decreases. As is known, the strong electron-phonon coupling among high-frequency phonons is the essential condition for the high $T_c$ of hydrogen sulfide at high pressures. Thus, the decreased electron-phonon interactions for high-frequency phonons will inevitably reduce $T_c$. 
FIG. S6. The external magnetic field dependent $\alpha^2 F$ of superconducting H$_3$S at 200 GPa.
References


