Supporting information for

High Bond Difference Parameters-Induced Low Thermal Transmission in Carbon allotropes with sp^2 and sp^3 hybridization

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- **Figure S3** Calculated heat capacities for T6-carbon, T10, 3D-C5, diamond and graphene at different temperatures, the heat capacities at 300 K has been circled.

Discussion 1: Thermal conductivity calculation by non-equilibrium molecular dynamics (NEMD) simulation

Thermal conductivity of T6-carbon, T10, 3D-C5, as well as diamond and graphene, is calculated using NEMD simulation in this work, which is conducted by introducing a heat flux through the simulation system, and then measuring the induced temperature gradient[1]. Figure S1 presents the thermal transmission in diamond as an illustration. In Figure S1(a), it can be seen that the heat flux is introduced by continuously transferring energy from 'cold' regions to 'hot' regions, here, the cold region is located at the ends of simulation system, and the hot region is in the middle. By exchanging the velocities of the hottest atoms in the cold region with the coldest atoms in the hot region, the energy transmission is accomplished, and then thermal conductivity can be calculated from the heat flux and temperature gradient shown in Figure S1(b) by following the Fourier's heat conduction equation[2]

$$J = -KdT / dx, \tag{1}$$

where K is the thermal conductivity, dT/dx is the temperature gradient averaged over time and space, and J is the heat flux density, which is given by the sum of exchanged energy per unit time and area,

$$J = \frac{1}{2tA} \sum_{transfer} \frac{m}{2} (v_{hot}^2 - v_{cold}^2), \tag{2}$$

where t is the simulation time, A is the cross-sectional area of the simulation system perpendicular to the direction of the heat flux, m is the atom mass, v_{hot} and v_{cold} denote the velocities of the hottest and coldest atoms to be exchanged at each step, and the factor 2 arises from the periodicity.

Discussion 2: Calculation of phonon dispersion and phonon density of states

In this work, the phonon dispersion and phonon density of states for T6-carbon, T10, 3D-C5, diamond and graphene are all calculated with the density functional theory (DFT) as implemented within the Vienna Ab initio Simulation Package (VASP)[3,4], which employs the plane-wave basis. The phonon dispersions are calculated by using the supercell approach with the finite displacement method implemented in the Phonopy package[5,6]. The plane-wave energy cutoff is set to be 770 eV, and the electronic energy convergence is 10^{-6} eV. In structure relaxations, the force convergence has been set to be 10^{-3}eV/A. In phonon calculations, the unit cells of the five carbon allotropes are used, for T6-carbon, the unit cell contains 6 atoms, for T10, 10 atoms, for 3D-C5, 20 atoms, for diamond, 8 atoms, and for graphene, only 2 atoms. For diamond with all sp^3 hybridization, the k points sampling

meshes are adopted as 9×9×9, and a mesh of 10×10×1 for graphene with all sp^2 hybridization, and 9×9×3 for T6-carbon, T10 and 3D-C5 with sp^2 and sp^3 hybridization.

References

[1] Bagri A, Kim SP, Ruoff RS, Shenoy VB. Thermal transport across twin grain boundaries in polycrystalline graphene from nonequilibrium molecular dynamics simulations. Nano Lett., 2011, 11(9): 3917-3921.

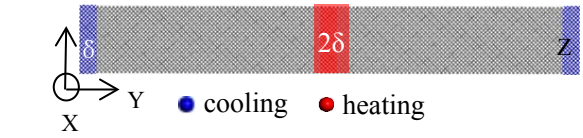
[2] Ju, SH, Liang XG. Thermal conductivity of nanocrystalline silicon by direct molecular dynamics simulation. J. Appl. Phys., 2012, 112: 064305.

[3] Dong HC, Xiao JW, Melnik R, Wen B. Weak phonon scattering effect of twin boundaries on thermal transmission. Sci. Rep., 2016, 6: 19575.

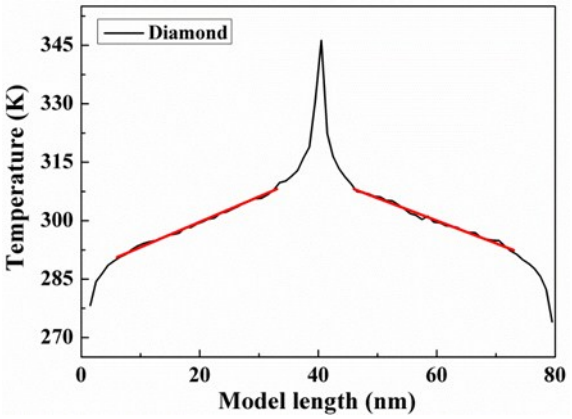
[4] Dong HC, Wen B, Zhang YW, Melnik R. Low thermal conductivity in Si/Ge hetero-twinned superlattices. RSC Adv., 2017, 7(48): 29959-29965.

[5] Kresse G, Hafner J. Ab initio molecular dynamics for liquid metals. Phys. Rev. B, 1993, 47(1): 558.

[6] Kresse G, Furthmüller J. Efficient iterative schemes for ab initio total-energy calculations using a plane-wave basis set. Phys. Rev. B, 1996, 54(16): 11169.



(a)



(b)

Figure S1 Diagram of NEMD process for calculating thermal conductivity of diamond. (a) Simulation model for thermal conductivity calculation, the hot region is located in the middle, and the cold region is located at the end. (b) Obtained temperature profile along the heat transmission direction.

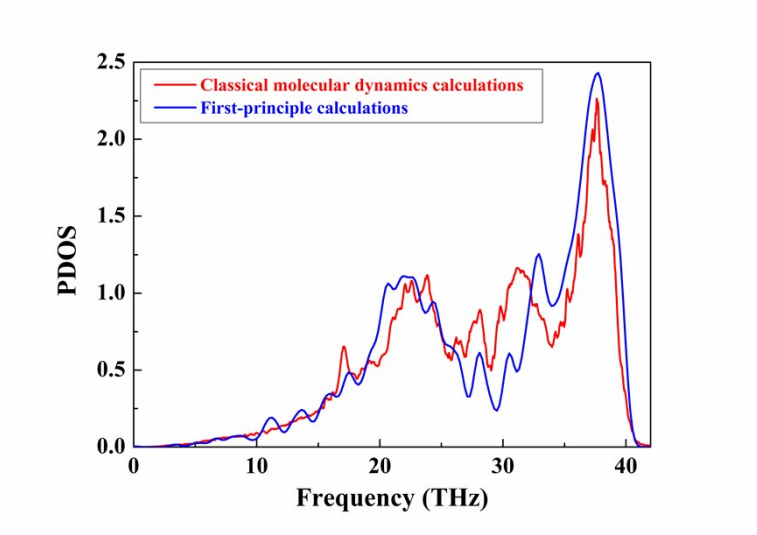


Figure S2 Comparison of phonon density of states for diamond between classical molecular dynamics calculations and first-principle calculations.

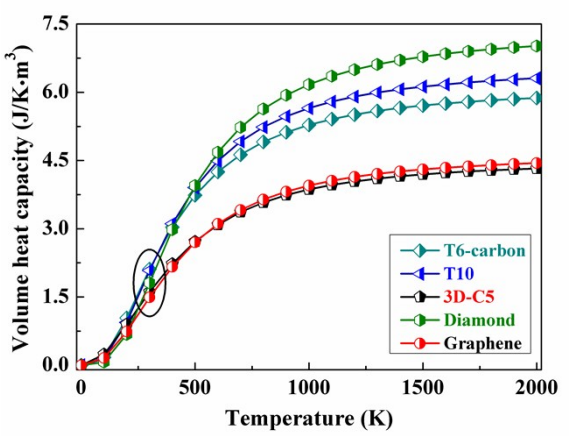


Figure S3 Calculated heat capacities for T6-carbon, T10, 3D-C5, diamond and graphene at different temperatures, the heat capacities at 300 K has been circled.