Electronic supplementary information for

Ultrahigh thermal conductivity of tetrahedral carbon allotropes with non-simple structure

Qiang Chen[1,†], Pei Zhang[2,†], Tao Ouyang[2*], Xiaoliang Zhang[3*], and Guangzhao Qin[1*]

[1]*State Key Laboratory of Advanced Design and Manufacturing for Vehicle Body, College of Mechanical and Vehicle Engineering, Hunan University, Changsha 410082, P. R. China*

[2]*Hunan Key Laboratory for Micro-Nano Energy Materials & Device and School of Physics and Optoelectronics, Xiangtan University, Xiangtan 411105, Hunan, China*

[3] *Key Laboratory of Ocean Energy Utilization and Energy Conservation of Ministry of Education, Institute of Energy and Environmental Engineering, School of Energy and Power Engineering, Dalian University of Technology, Dalian 116024, China*

[†] Equal contribution.
[*] Author to whom all correspondence should be addressed. E-Mail: T.O. <ouyangtao@xtu.edu.cn>, X.Z. <zhangxiaoliang@dlut.edu.cn>, G.Q <gzqin@hnu.edu.cn>

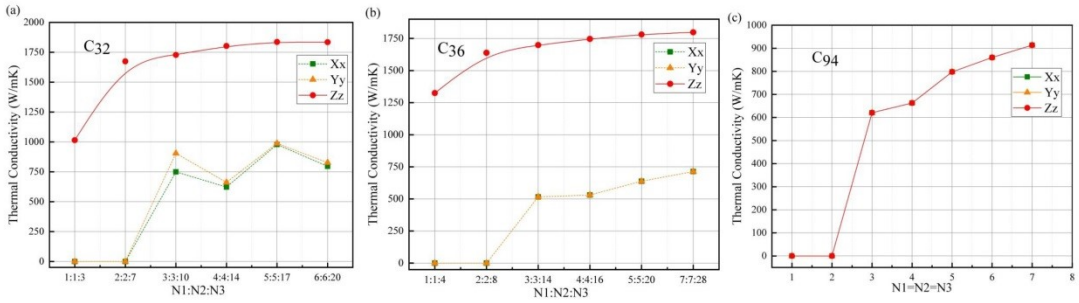

Figure S1. The convergence of thermal conductivity with respect to the grid sampling in *Q* space.

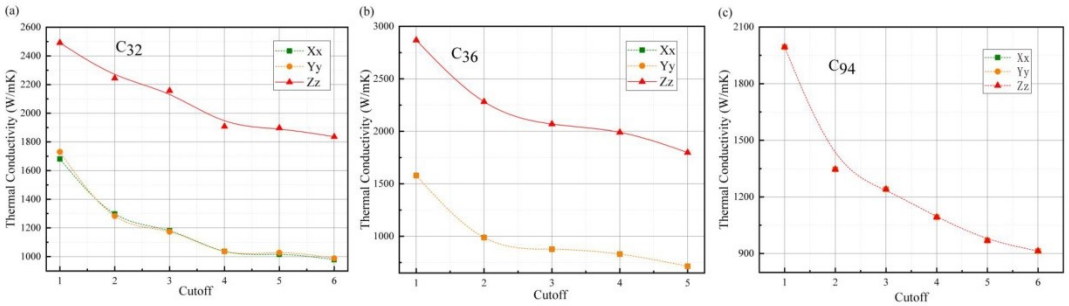

Figure S2.The convergence of thermal conductivity with respect to the cutoff distance of all cases.

The tests of grid convergence of C_{32} and C_{94} in *Q* space both are under the sixth neighbor, but the test of C_{36} is under the fifth neighbor. It is found in Figure S1 that the *z* direction of C_{32} and C_{36} converge rapidly in *Q* space, the grid convergence is *5×5×17* and *7×7×28* respectively, and the structure of C_{94} converges slowly.

Subsequently, as shown in Figure S2, we carry the tests of the cutoff distance of C_{32} *(5×5×17),* C_{36} *(7×7×28)* and C_{94} (*7×7×7)*, both C_{32} and C_{36} converge well near the fifth neighbor except for C_{94}. The difference of thermal conductivity of C_{36} between the third and the fourth neighbor accounts for 3.97% of that of the fourth neighbor in the *z* direction, and similarly accounts for 5.60% in the *x*, *y* direction. The thermal conductivity difference of C_{94} between the fifth and the sixth neighbor is 6.03% of that of the fifth neighbor in the *x*, *y* and *z* direction. Generally, convergence can be considered to be achieved with the difference within 10%, which ensures the accuracy of our calculation results.

Table S1. The convergence test of thermal conductivity (W/mK) with respect to smear parameter.

Scalebroad	**0.1**	**0.2**	**0.3**	**0.5**	**0.7**	**0.9**	**1.0**	**2.0**
C_{32}	1267.59	1267.99	1269.06	1269.64	1269.26	1268.42	1267.94	1264.16

All the calculations by solving the phonon BTE utilizing the ShengBTE package are executed on scalebroad settings with the default value of 1.0. The default value is theoretically guaranteed to work according to the convergence test of thermal conductivity (W/mK) with respect to smear parameter in Table S1.

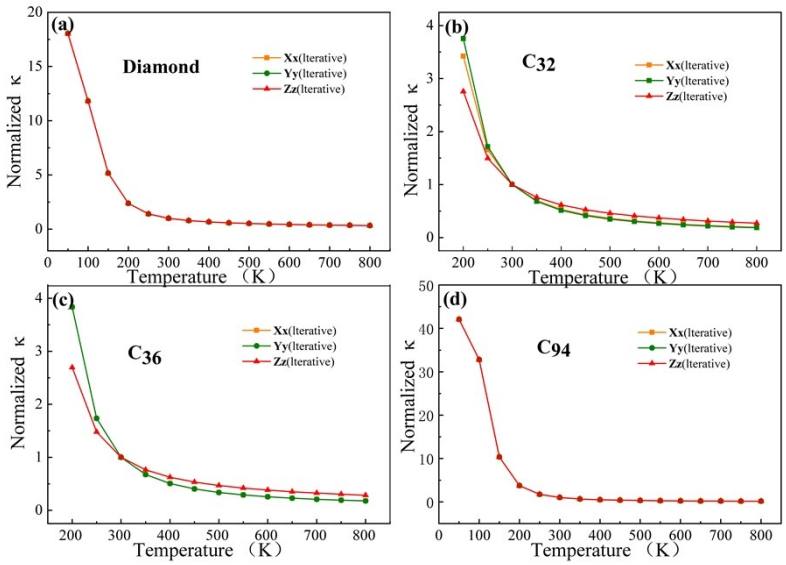

Figure S3.The thermal conductivity which is normalized at 300 K

varies with temperature in different directions.

It can be seen from Figure S3 that the thermal conductivity of *x* ,*y* and *z* directions of Diamond, C_{32}, C_{36} and C_{94} gradually decreases with the increasing of temperature, and tends to be flat after reaching a certain temperature. Where, the thermal conductivity of Diamond and C_{94} remains consistent in three directions, which shows they are isotropic materials. On the contrary, C_{32} and C_{36} are anisotropic materials.

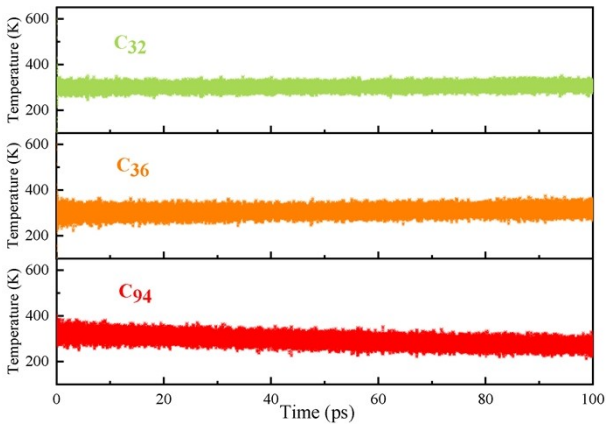

Figure S4. The evolution of temperature with time during the AIMD simulation of C_{32} (green), C_{36} (orange) and C_{94} (red).

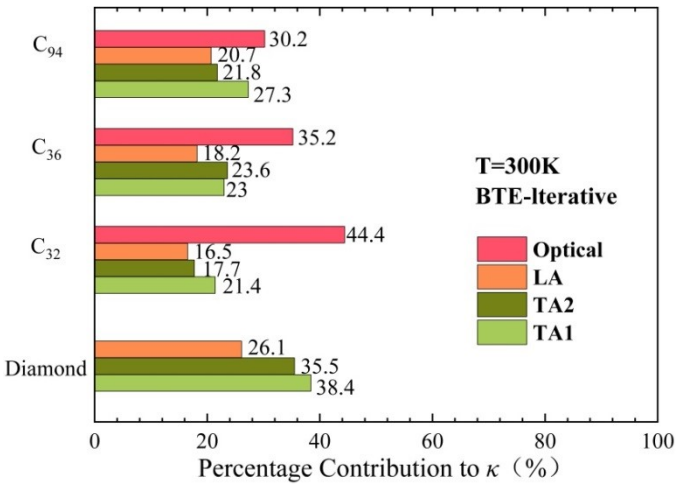

Figure S5.Percentage contribution to κ from each phonon branch (Optical, LA, TA2, and TA1).

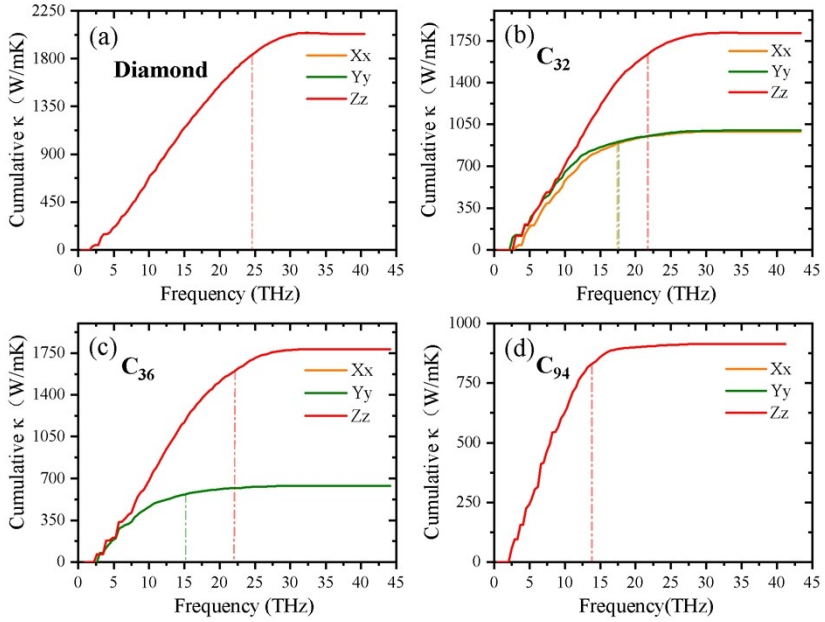

Figure S6. The cumulative thermal conductivities as a function of phonon frequencies. The dotted lines mark the 90% cumulated contribution to κ.

Traditionally, acoustic phonon branches with lower frequencies contribute most to the total κ. To make it clear in the systems as studied in this work, we present the cumulative κ as a function of frequency in Figure S6. One can note that the main contribution (90%) to the total κ (labeled as $\kappa_{0.9}$ by dotted line) indeed comes from the low-frequency phonons.

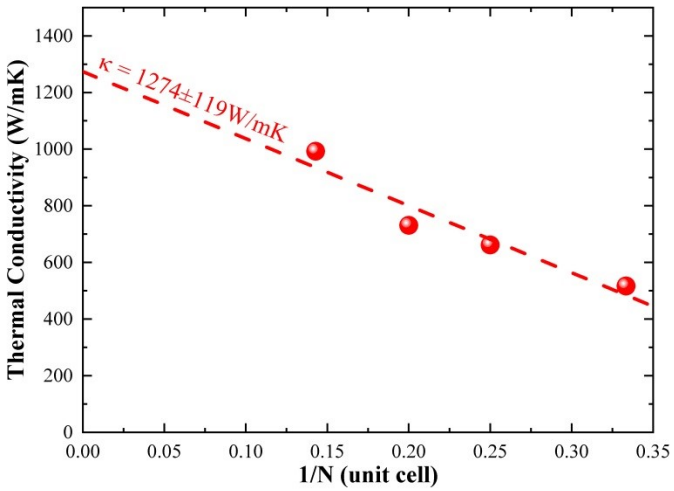

Figure S7. The dependence of thermal conductivity of C_{32} on the size of supercell for SED calculations. The fitted thermal conductivity of C_{32} is presented onsite.

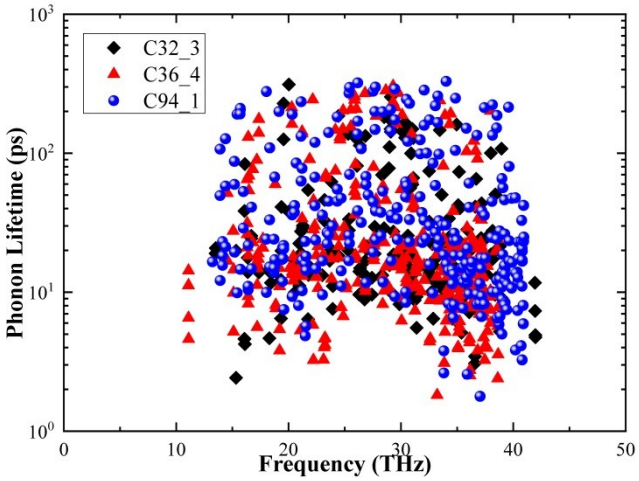

Figure S8. The comparison of the frequency dependent phonon lifetimes of C_{32}, C_{36} and C_{94}.

We carried out the phonon spectral energy density (SED)[1] analysis to obtain the phonon lifetime. First, the phonon normal modes can be obtained by the following equation[1]:

$$\dot{Q}(\vec{\kappa},\mu,t) = \sum_{jl} \sqrt{\frac{m_j}{N}} v_{jl}(t) \cdot \vec{e}_j * (\vec{\kappa},\mu) exp(-2\pi i \vec{k} \cdot \vec{r}_l) \quad (1)$$

Second, the spectral energy density can be calculated by the following equation[2]:

$$\Phi(\vec{\kappa},\mu,f) = |\int \dot{Q}(\vec{\kappa},\mu,t)exp(-2\pi if)d_t|^2 \quad (2)$$

and the phonon linewidth can be obtained by fitting the Lorentzian function[2] as follows:

$$\Phi(\kappa,\mu,f) = \frac{I}{1+\left[\frac{2\pi(f-f_0)}{\gamma}\right]^2} \quad (3)$$

where I is the peak magnitude, f_0 is the frequency at the peak center, and γ is the half-width at half-maximum. Finally, the phonon lifetime can be calculated as below[3]:

$$\tau = \frac{1}{2\gamma} \quad (4)$$

Table S2. The statistics of bond lengths (Å) of diamond, C_{32}, C_{36} and C_{94}.

Diamond	**Number**	**C_{32}**	**Number**	**C_{36}**	**Number**	**C_{94}**	**Number**
1.527936	8	1.546805	32	1.543387	48	1.499317	144
		1.528828	16	1.526269	24	1.527902	48
		1.533526	16	1.535369	24	1.530220	48
		1.559770	16	1.537883	24	1.535118	48
		1.508311	8	1.579293	12	1.599471	48
		1.522193	8	1.611671	12	1.498143	24
		1.565628	8			1.643112	16
		1.573340	8				
		1.577869	8				
		1.460535	4				
		1.586793	4				

1. Zhang, X., Bao, H. & Hu, M. Bilateral substrate effect on the thermal conductivity of two-dimensional silicon. *Nanoscale* **7**, 6014–6022 (2015).

2. Larkin, J. M., Turney, J. E., Massicotte, A. D., Amon, C. H. & McGaughey, A. J. H. Comparison and Evaluation of Spectral Energy Methods for Predicting Phonon Properties. *Jnl of Comp & Theo Nano* **11**, 249–256 (2014).

3. Wei, Z., Yang, J., Bi, K. & Chen, Y. Mode dependent lattice thermal conductivity of single layer graphene. *Journal of Applied Physics* **116**, 153503 (2014).