Electronic supplementary information for

Ultrahigh thermal conductivity of carbon allotropes with correlations to the scaled Pugh ratio

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1. Three-phonon scattering channels.

We calculate the scattering channels of the four carbon allotropes to understand the scattering process. The results are shown in Fig. S1-3. Generally, the scattering channels could be divided into two types: the absorption process (as shown in Fig.S1), where two phonons merge into one phonon, and the emission process (as given in Fig. S2 and S3), where one phonon splits into two phonons. For simplicity, "A", "O" and "X" are used to stand for the acoustic, optical and any phonon branches, respectively. Overall, the scattering rates of absorption process are about one magnitude smaller than that of emission process, indicating that the phonons are mainly scattered through emission processes. In the absorption process, the behaviors of four allotropes are quite different. In Bct-C4 and Z-carbon, the scattering channels only contain X+O/A→O, while in lonsdaleite, additional scattering channels like X+ZA/TA→TA/LA also exist. Moreover, the scattering channels like X+LA→TA/LA also exist in diamond, where the scattering rates and frequency width

of X+O→O are extremely small compared to that of the other three materials, indicating the weak coupling between optical and acoustic branches in diamond.

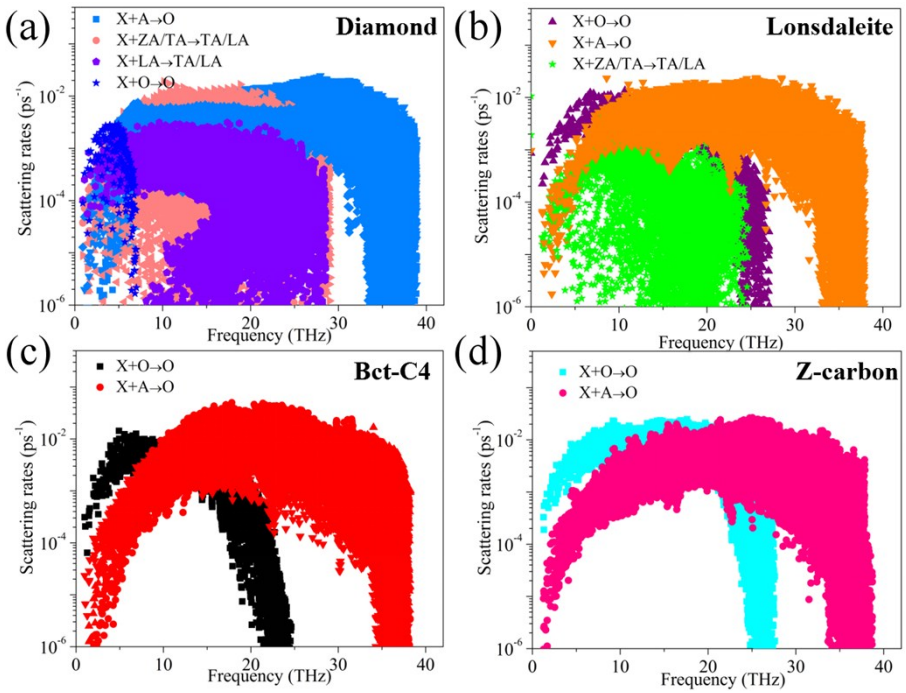

Fig. S1. Calculated three phonon scattering rates in absorption processes for (a) diamond, (b) lonsdaleite, (c) Bct-C4 and (d) Z-carbon, respectively.

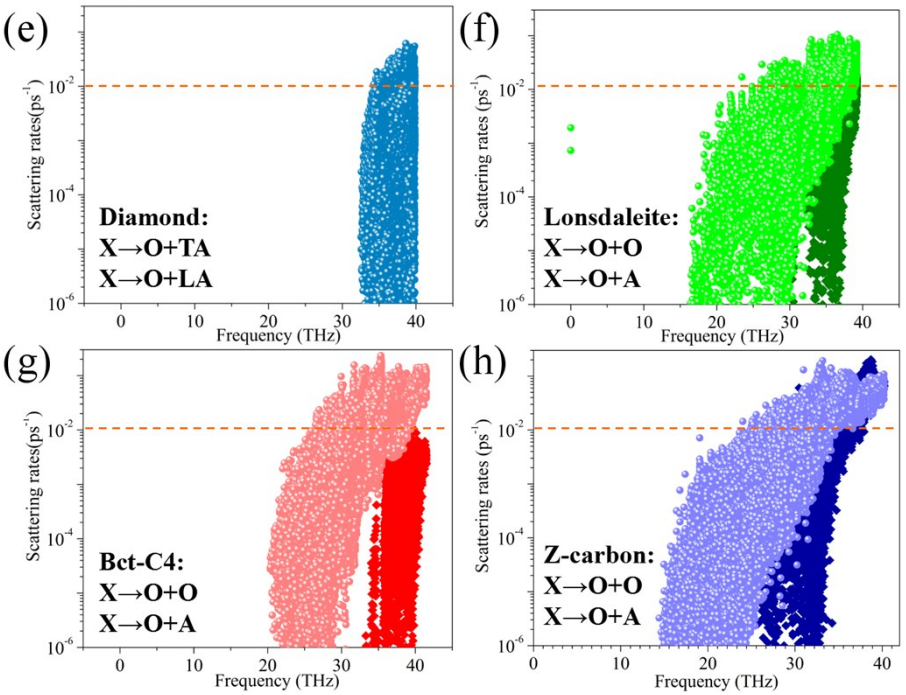

Fig. S2. Calculated three phonon scattering rates in type I absorption processes for (a) diamond, (b) lonsdaleite, (c) Bct-C4 and (d) Z-carbon, respectively. The dashed lines are guide for the eyes.

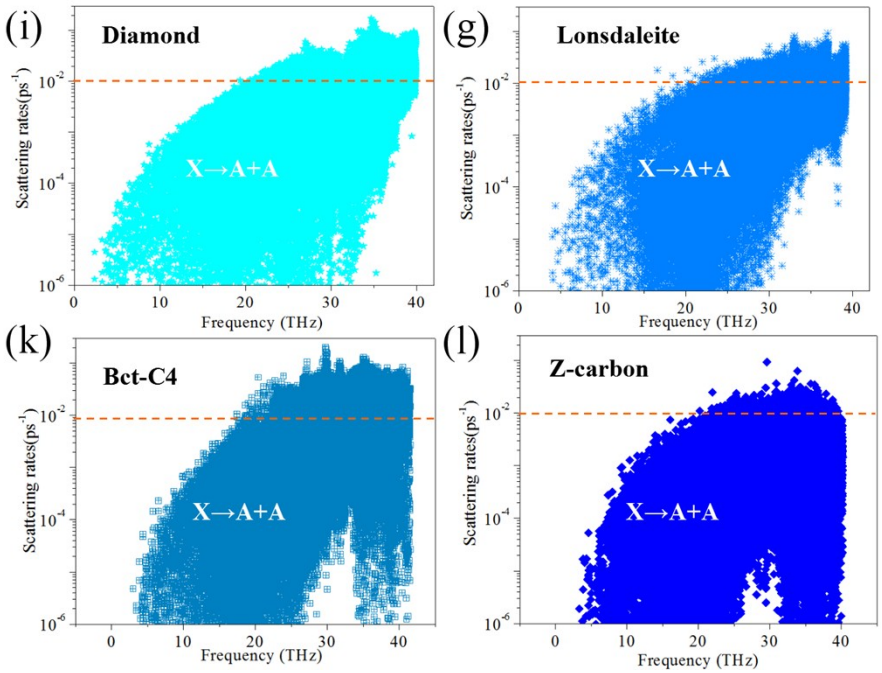

Fig. S3. Calculated three phonon scattering rates in type II absorption processes for (a) diamond, (b) lonsdaleite, (c) Bct-C4 and (d) Z-carbon, respectively. The dashed lines are guide for the eyes.

For the convenience of discussions, we divide the scattering channels of emission processes into two types: type I (containing both acoustic and optical branches, as displayed in Fig. S2), and type II (only involving acoustic branches, as shown in Fig. S3). In the type I process, the main scattering channels for lonsdaleite, Bct-C4 and Z-carbon are X→O+O/A, while only X→O+TA/LA exists in diamond. So, the phonon scattering in diamond is significantly suppressed. One can also clearly see that in the type I emission process, the scattering rates of Bct-C4 and Z-carbon are comparable to each other, and they are larger than that of lonsdaleite and diamond. In the type II emission process, although the four carbon allotropes exhibit the same

scattering channels like X→A+A, the scattering rates of Z-carbon are slightly less than that of diamond, Bct-C4 and lonsdaleite. Therefore, the difference in thermal conductivity of four compounds is mainly due to the type I emission process. So we can conclude that the weak coupling between acoustic and optical branches in diamond leads to low scattering rates in the depressed three phonon scattering channels, thus endowing diamond with ultrahigh thermal conductivity.

2. Elastic properties calculation

Table S1. The calculated shear modulus *G,* bulk modulus *K* and Young's modulus E_H for the carbon allotropes. The data in bracket are from previous studies for comparison.[1-11]

	G	*K*	E_H
Diamond	518.59 (518.7[1],535[5])	435.12 (435.1[1],443[3])	1113.43 (1113.8[1],1178[11])
Lonsdaleite	524.36 (552[3],551.0[10])	435.51 (444[3], 430[4],454.5[10])	1122.55 (1185[11])
Bct-C4	419.76 (419.9[1],431.2[6],428.77[7])	416.23 (403.4[1],409.6[4],401.9[9])	1023.21 (935.5[1],989[11], 1021.1)
Z-carbon	469.25 (470.7[1],468.1[8])	416.23 (409.6[1],412.7[8])	1023.21 (1021.1[1],1081[11])
graphite	157.34	115.31 (294[4])	276.19
T12-carbon	406.76 (422.1[2],479.3[10])	458.73 (451.8[2],424.8[10])	1000.19
AA-T12	361.46	372.96	832.54
T-carbon	159.82 (169[2], 175[4])	45.48 (70[2])	124.46
3D-graphene	147.62	46.05	120.52
BCO-C16	210.07	150.24	362.76

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