# Supplementary information for "Substrate Coupling Suppresses Size Dependence of Thermal Conductivity in Supported Graphene"

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### I. HIGH-TEMPERATURE ANNEALING PROCESS

To construct the amorphous SiO<sub>2</sub> substrate at room temperature, we start with the crystalline form of SiO<sub>2</sub>: alpha-quartz ( $\alpha$ -quartz). We apply Langevin heat bath to equilibrate  $\alpha$ -quartz at 3000 K (above melting point) for 100 ps in order to achieve the amorphous structure. The resultant structure is then annealed to room temperature with a constant cooling rate of 10<sup>13</sup> K/s [1]. The partial pair distribution function [2] for different chemical bonds in the amorphous SiO<sub>2</sub> structure generated in our study (Fig. S1) shows excellent agreement with previous study [3], which highlights the accuracy of the high-temperature annealing process used in our simulation.

### II. NON-EQUILIBRIUM MOLECULAR DYNAMICS SIMULATION

Before non-equilibrium MD simulation, the canonical ensemble MD simulation with Langevin heat bath first runs for  $10^5$  steps to equilibrate the whole system at room temperature. After structure relaxation, fixed boundary condition is used at the two ends of the length (*x*) direction (Fig. 1a). Next to the fixed boundary, Langevin heat baths with different temperature are applied to the two ends of *x* direction to simulate the heat source (red box) and heat sink (blue box) in real

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experiment, respectively. Periodic boundary condition is used in the width (y) direction, and free boundary condition is used in the out-of-plane (z) direction. Both the graphene and substrate are attached to the heat bath with the same temperature at each end (Fig. 1(b)). The non-equilibrium MD simulations are then performed long enough ( $10^7$  time steps) to allow the system to reach the non-equilibrium steady state where the temperature gradient is well established and the heat flux passing through the system is time independent. Thermal conductivity is calculated according to

$$\kappa = -J/\nabla T,\tag{S1}$$

where  $\nabla T$  and *J* is, respectively, the temperature gradient and the heat flux transported in graphene region only. In our simulation, the heat flux is calculated according to the energy injected into / extracted from the heat source / heat sink in the graphene sheets only (exclude the heat flux in substrate) across unit area per unit time. These two rates are equal in the non-equilibrium steady state. For supported graphene, only heat flux transported in graphene region is recorded in our simulations. The cross section area (*S*) of the graphene sheets is defined as S=3.35\*W\*n Å<sup>2</sup> in our calculations, where *W* is the width of graphene, and *n* is the number of layers. The temperature gradient is calculated according to the slope of the linear fit line of the local temperature in graphene along *x* direction.

## III. COMPARE THERMAL CONDUCTIVITY OF SUSPENDED SINGLE-LAYER GRAPHENE WITH LITERATURE VALUE

We notice that  $\kappa$  of suspended SLG found in our study (~1000 W/m-K) is much larger than that reported by Ong *et al.* [4] (256 W/m-K) under the similar sample size (length is about 30 nm and width is about 5 nm). We contribute this discrepancy to the choice of interatomic potential used for graphene. The original Brenner potential was used in Ref. [4], while we use the Tersoff potential with optimized parameters set for graphene developed by Lindsay *et al.* [5], which can describe more accurately the upper optic phonon branches while providing a good fit to the acoustic velocity and phonon frequency. Lindsay *et al.* found that the original Brenner potential failed to accurately represent the zone-center velocities for all the acoustic modes [5], leading to 30% underestimation for longitudinal acoustic (LA) branch and 12% underestimation for transverse acoustic (TA) branch compared to experimental values. Since thermal conductivity depends critically on the group velocity of acoustic phonons based on the Boltzmann transport equation approach, Lindsay *et al.* found the use of original Brenner potential can lead to a large underestimation of thermal conductivity for suspended SLG [5].

#### IV. SPECTRAL ENERGY DENSITY ANALYSIS

The spectral energy density (SED) in our calculation is defined as [4, 6]

$$\Phi(k,\omega) = \frac{1}{4\pi\tau_0 N_x N_y N_z} \sum_{\alpha} \sum_{b=1}^B m_b \bigg| \int_0^{\tau_0} \sum_{n_x=0}^{N_x-1} \sum_{n_y=0}^{N_y-1} \sum_{n_z=0}^{N_z-1} v_{\alpha,b}(n_x, n_y, n_z, t) \exp\left[\frac{2\pi i k n_x}{N_x} - i\omega t\right] dt \bigg|^2$$
(S2)

where k is the wavevector index,  $\omega$  is the angular frequency,  $\tau_0$  is the total simulation time,  $\alpha$  is the Cartesian index, b is the atom index in each unit cell, m and v is the atomic mass and velocity, respectively, and  $N_x$ ,  $N_y$ ,  $N_z$  denotes the number of unit cell in x, y, z direction, respectively. Here we consider a one-dimensional Brillouin zone along the length (x) direction ( $1 \le k \le N_x$ ). We choose the same four-atom unit cell described in Ref. [4], and use a fixed simulation domain of  $N_x=20$  and  $N_y=6$  for the in-plane direction. Periodic boundary condition is used in both x and y (in-plane) direction, and free boundary condition is used in z (out-of-plane) direction. After structure relaxation with Langevin heat bath, we carry out microcanonical ensemble (NVE) MD simulation to the whole system for 3 ns and 1 ns for the suspended and supported graphene, respectively, and record the velocity for each atom in the graphene every 5 fs. We further extend the NVE MD simulation steps and find the calculation results of SED is well converged within the abovementioned total simulation time. To examine the accuracy of our simulation, we calculate the SED for LA phonons near zone-center (k=1) in suspended SLG and find the eigen-frequency at  $f=4.45\pm0.05$  THz (Fig. S2). Using the lattice constant  $L_0$  and group velocity for LA phonons  $v_{LA}$  associated with the optimized Tersoff potential listed in Ref. [5], the theoretical estimation gives rise to  $f=v_{LA}/(L_0N_x)=4.38$  THz, in good agreement with our simulation results.

### V. ESTIMATE THERMAL CONDUCTIVITY OF BULK GRAPHITE

In order to estimate the thermal conductivity in the bulk graphite limit from our existing data, we fit the raw data from MD simulation according to the double exponential function based on the two-stage increase characteristic as

$$\kappa = \kappa_0 + A_1 \left( 1 - e^{-n/B_1} \right) + A_2 \left( 1 - e^{-n/B_2} \right), \tag{S3}$$

where  $\kappa_0$ ,  $A_1$ ,  $B_1$ ,  $A_2$  and  $B_2$  are fitting parameters. The double exponential fitting yields  $\kappa_0$ =417,  $A_1$ =147, and  $A_2$ =398, suggesting thermal conductivity in bulk graphite limit as  $\kappa_{graphite} = \kappa_0 + A_1 + A_2 = 962$  W/m-K.

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FIG. S1: Partial pair distribution function (PDF) for different chemical bonds in amorphous SiO2 at 300 K.(a) Si-Si bond. (b) Si-O bond. (c) O-O bond.



FIG. S2: Normalized spectral energy density (SED) for zone-center LA phonons (k=1) in suspended singlelayer graphene.