Electronic Supplementary Material (ESI) for RSC Advances.

Supplementary information for 'Exotic thermoelectric behavior in nitrogenated holey graphene'

Yinchang Zhao,[1,*] Zhenhong Dai,[1,†] Chao Lian,[2,‡] and Sheng Meng[2,3,§]

[1] *Department of Physics, Yantai University, Yantai 264005, People's Republic of China*

[2] *Beijing National Laboratory for Condensed Matter Physics and Institute of Physics, Chinese Academy of Sciences, Beijing, 100190, People's Republic of China*

[3] *Collaborative Innovation Center of Quantum Matter, Beijing 100084, People's Republic of China*

(Dated: March 21, 2017)

S1. Phonon group velocities for nitrogenated holey graphene

Besides phonon scattering rates, another important factor determining thermal transport is phonon group velocity $v_g = d\omega_\nu/dq$, where ν represents the νth branch within the whole first Brillouin zone. The phonon group velocity of all phonon modes within the first Brillouin zone as a function of frequency for nitrogenated holey graphene is plotted in Fig. S1. The maximum group velocities for three acoustic branches are about 2.50, 17.76, and 18.48 km/s, respectively. For most of optical modes, the maximum group velocities are in the range of 1.55 to 13.16 km/s. These values are much higher than those of $CoSb_3$ and IrSb. In view of $\kappa_L \sim v_g^2$ in RTA, these relatively high group velocities support that the values of the re-scaled κ_L for nitrogenated holey graphene are two or three times higher than those of $CoSb_3$ and $IrSb_3$, although the larger anharmonic three-phonon scattering rates are observed in nitrogenated holey graphene.

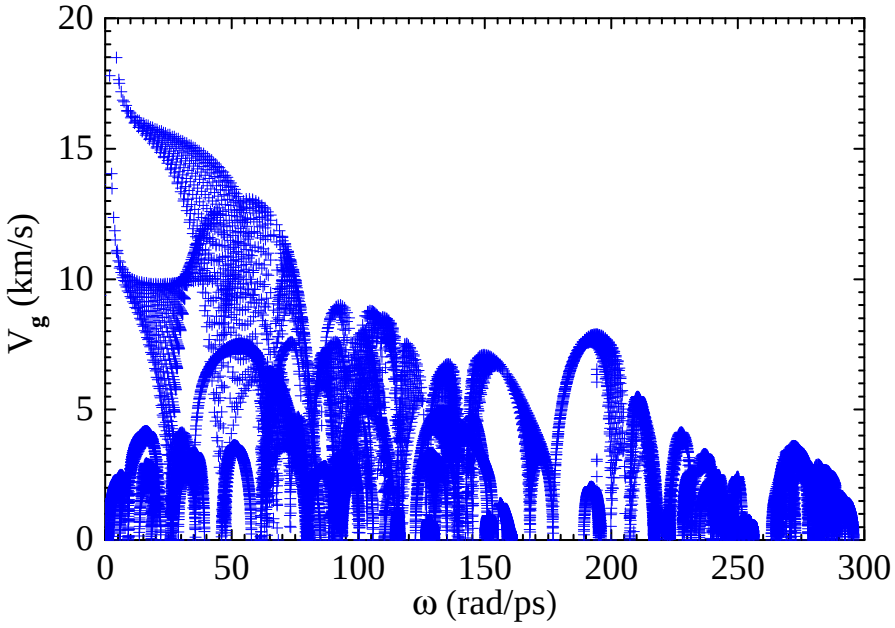

FIG.S1. (Color online). Phonon group velocities of all phonon modes within the first Brillouin zone as a function of frequency for nitrogenated holey graphene.

S2. The doping concentration n, average electron-electron distance d, electron velocity v, and scattering time τ for nitrogenated holey graphene

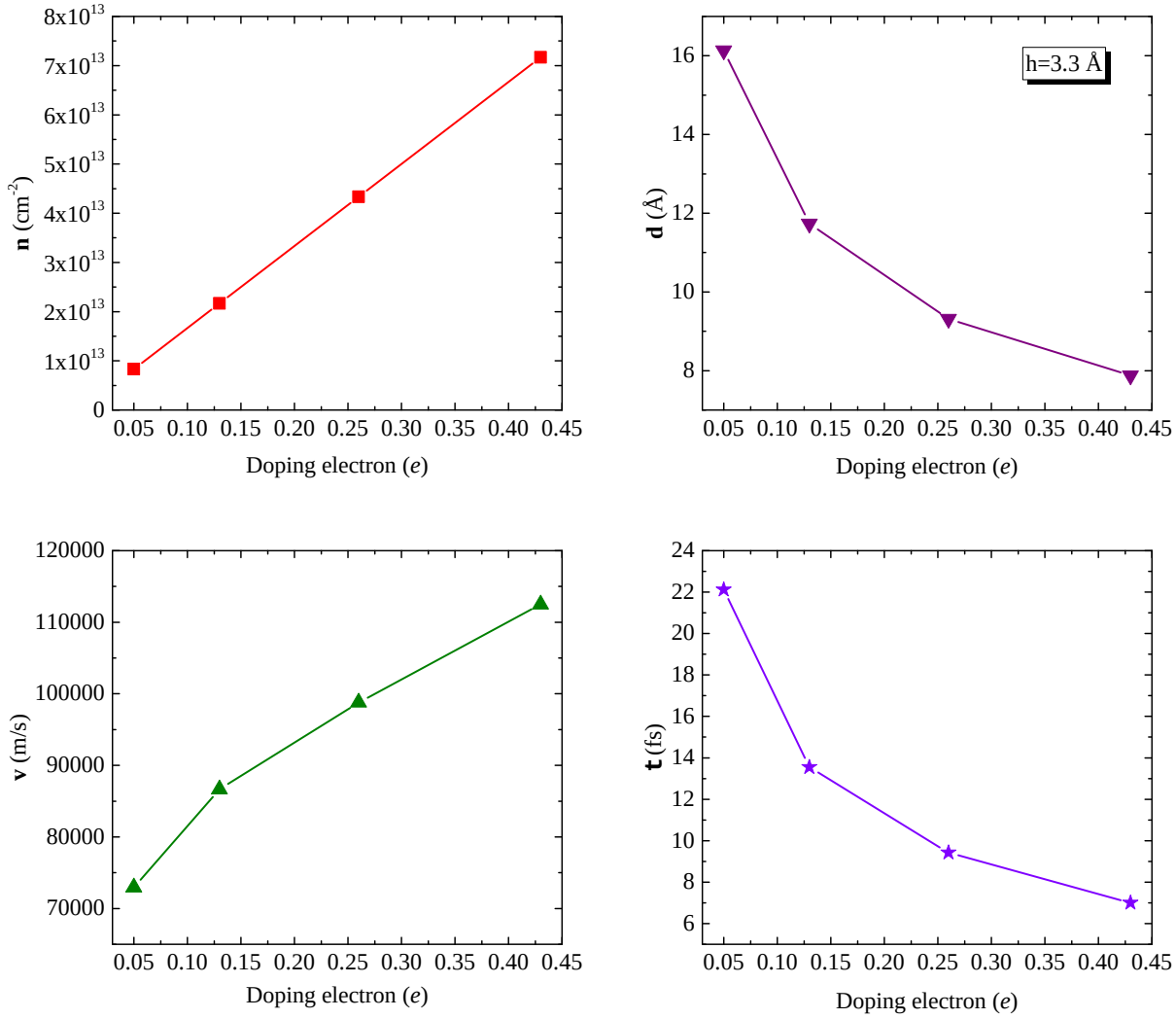

FIG.S2. (Color online). The doping concentration n, average electron-electron distance d, electron velocity v, and scattering time τ as a function of doping electron e for nitrogenated holey graphene.

The model $\tau \sim d/v$ is used to assess the amplitude of scattering time τ for nitrogenated holey graphene, where d is the average electron-electron distance estimated by the doping concentration n and the effective thickness $h \sim 3.30$ Å, $v = \frac{1}{\hbar}\frac{\partial E(k)}{\partial k}$ represents the corresponding electron velocity. As shown in Fig.S2, in the n-type doping range of 0.05 to 0.43 e in each primitive cell, $n \sim 8.33\times10^{12}$ to 7.17×10^{13} cm^{-2}, $d \sim 16.13$ to 7.87 Å, $v \sim 7.30\times10^{4}$ to 11.25×10^{4} m/s, and $\tau \sim 23$ to 6 fs. At $n \sim 2.00\times10^{13}$ cm^{-2}, $d \sim 12$ Å, $v \sim 9.30\times10^{4}$

m/s, and thus $\tau \sim 13$ fs. Therefore, it suitable to select the value of $\tau = 13$ fs to calculate the electrical conductivity σ and electronic thermal conductivity κ_{el}. The values calculated with $\tau = 6$ and 23 fs can be taken as the lower and upper limits of these parameters.

S3. Structural stabilities of nitrogenated holey graphene

In order to study the structural stabilities of nitrogenated holey graphene with n-type doping at high temperature, *ab initio* molecular dynamics (MD) calculations are carried out. After 5000 steps (10 ps) MD at 1500 K, the nitrogenated holey graphene with doping concentration $n \sim 2.00 \times 10^{13}$ cm^{-2} keeps quite intact and have no any lattice destruction. In Fig.S3, we show the bond length evolution of nitrogenated holey graphene with $n \sim 2.00 \times 10^{13}$ cm^{-2} at 1500 K. The C-N, C-C1, and C-C2 bonds keep small fluctuations around their balanceable bond lengths.

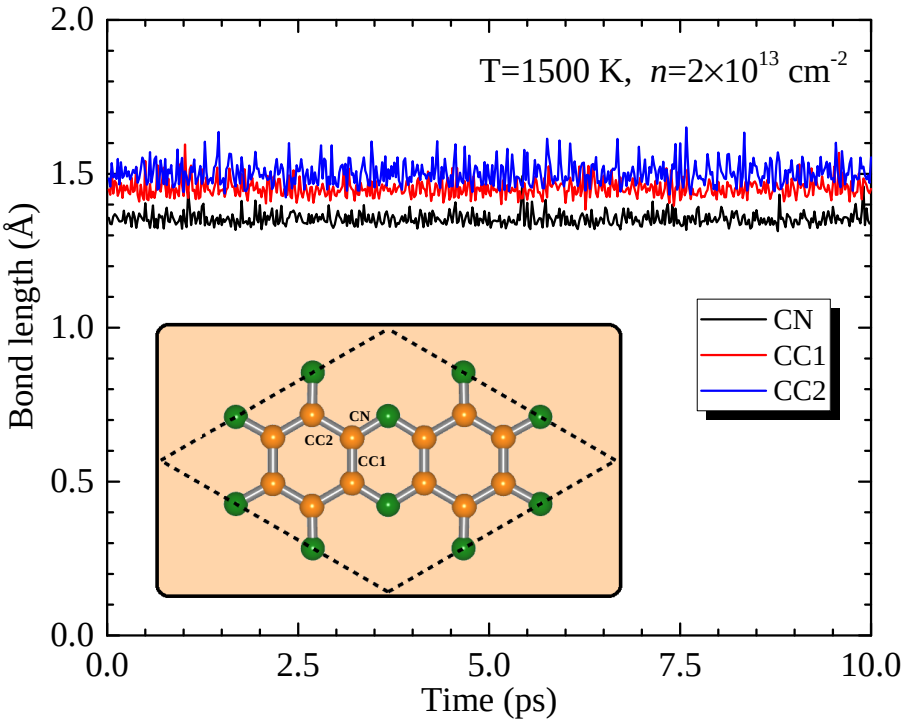

FIG.S3. (Color online). Bond length evolution at 1500 K for nitrogenated holey graphene with n-type doping concentration $n \sim 2.00 \times 10^{13}$ cm^{-2}. Inset shows the primitive cell labeled the C-N, C-C1, and C-C2 bonds.

* y.zhao@ytu.edu.cn

† zhdai@ytu.edu.cn

‡ chaolian@iphy.ac.cn

§ smeng@iphy.ac.cn