Effects of vacancy and magnetic field on thermoelectric properties of straight and kinked graphene nanoribbons

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INTRODUCTION

Low-dimensional systems and structures are promising for thermoelectric applications due to the quantum confinement effects, such as graphene nanoribbons (GNRs) [1]. The efficiency of thermoelectric effects can be represented by the dimensionless figure of merit, $ZT = S^2\sigma T/\kappa$, where S, σ, T and κ are the Seebeck coefficient, electrical conductivity, absolute temperature, and thermal conductivity respectively [2]. Theoretical study has shown that thermoelectric performance of GNR is improved compared to 2D graphene, and surface roughness and defects can decrease the thermal conductivity [3]. Moreover, magnetic B-field can modulate the electron transport [4]. Recent studies have also shown that various kinked GNRs have better thermoelectric performance than straight GNRs [5]. So it is interesting to study the effects of defects and magnetic field on thermoelectric properties of various GNR structures.

In this work, Landauer formulism in linear response based on nonequilibrium Green's function (NEGF) is used to calculate the thermoelectric properties. Hamiltonian and force constant for electron and phonons are obtained by π orbital tight-binding method and the fourth-nearest-neighbour interactions approach, respectively [6]. When an external magnetic field is applied in the unit of flux quantum, $\varphi_0 = h/q$, the hopping energy is modified following Peierls phase approximation [7]. Below is a list of key equations.

$$t(\varphi) = t(0)\exp(iq/h\int_{l_i}^{l_j} A(\varphi)d\vec{l}), \qquad t(0) \approx 3eV \tag{1}$$

$$L_n(\mu,T) = 2/h\int dET_E(E-\mu)^n[-\partial f(E,\mu,T)/\partial E] \tag{2}$$

$$G(\mu,T) = e^2 L_0(\mu,T) \tag{3}$$

$$S(\mu,T) = L_1(\mu,T)/[qTL_0(\mu,T)] \tag{4}$$

$$\kappa_e(\mu,T) = (1/T)[L_2(\mu,T) - L_1^2(\mu,T)/L_0(\mu,T)] \tag{5}$$

$$\kappa_{ph} = 1/h\int d\hbar\omega T_{ph}(\omega)\hbar\omega[\partial n(\omega,T)/\partial T], \quad E_{ph} = \hbar\omega \tag{6}$$

$$ZT = S^2GT/(\kappa_e + \kappa_{ph}) \tag{7}$$

DISCUSSION

First, we investigate the thermoelectric properties of 9-AGNRs and 1nm kinked AA-GNRs with smooth and defected structures as shown in Fig. 1, under room temperature T = 300K. The peak values of ZT can be observed near the zero reduced Fermi-level ($\eta_F = E_F - \varepsilon_1$, ε_1 is the first conduction or valance band) as shown in Fig. 2. Considering GNRs without vacancies, kinked AA-GNR has a higher ZT than straight 9-AGNR mainly due to the reduction in κ_{ph}. Now, comparing defected with smooth GNRs, S values are almost the same while defected GNRs only have slightly higher values than smooth ones. Defected GNRs have smaller κ_{ph}, since phonon transmission has more scattering but lower values as shown in Fig. 3. However smooth GNRs have much higher G. This can be explained through the electron transmission as shown in Fig. 4. With vacancies in the center of the channel, transmission becomes smaller whose value is upper-bounded by those of smooth cases, and no longer in steps of units. Hence, even though κ_{ph} is decreased for smooth 9-AGNR and kinked AA-GNRs, ZT is decreased since the decrease in G outweighs the decrease in κ_{ph} and the increase in S.

Next, we investigate the thermoelectric performance when an external magnetic field is applied perpendicular to the channel. Fig. 5 shows that the first peak value of ZT (ZT_{max}) is changing as B-field varies. Under B-field, ZT_{max} of smooth 9-AGNR can reach 0.33, almost twice as when $\varphi = 0$. For defected 9-AGNR, at $\varphi = 22\varphi_0$, ZT_{max} is around six times as large as when $\varphi = 0$, and the value is larger than that of smooth 9-AGNRs under B-field from 0 to $40\varphi_0$. This is because electron transmission is greatly improved when B-field of $\varphi = 22\varphi_0$ is applied, which is shown in Fig. 4(a). The bandgap is smaller, and the transmission is larger than those for smooth 9-AGNR. However, for defected kinked AA-GNRs, varying B-field can improve ZT_{max}, but it doesn't exceed the value of smooth AA-GNRs under different B-fields.

Last, other types of hybridized kinked GNRs with segment width of 1 nm are studied as shown in Fig. 6. For defected ones, κ_{ph} are all smaller than those for smooth ones. Similarly to 9-AGNR and kinked AA-GNR, defected GNRs have much smaller ZT_{max} than smooth ones, with one exception for defected kinked ZZ-GNR whose ZT_{max} is improved due to a unique electron transmission peak near the first conduction band which improves S and G. Applying same B-field $\varphi = 22\varphi_0$ to all defected kinked GNRs, ZT_{max} of kinked AA- and AZZ-GNRs are enhanced, but not larger than smooth ones.

CONCLUSION

In summary, vacancies in various GNR structures can decrease κ_{ph}, but ZT also decreases for various GNR structures except for kinked ZZ-GNR. With an external B-field, electron transport can be tuned to obtain an enhanced thermoelectric performance.

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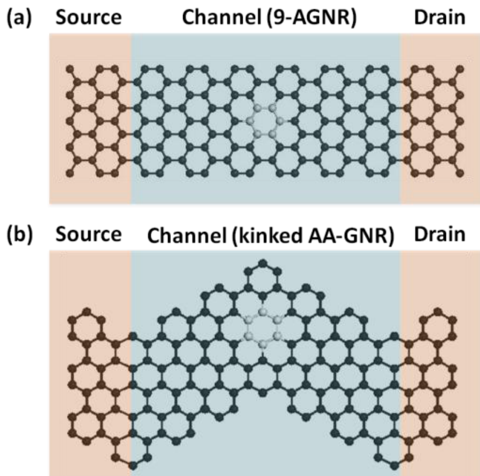

Fig. 1. (a) 9-AGNR and (b) kinked AA-GNR with segment arm width of 1 nm as the channel for electron and phonon transport calculations. The defect-engineered GNRs are constructed by removing the center (6 atoms shown in grey color) to create a vacancy.

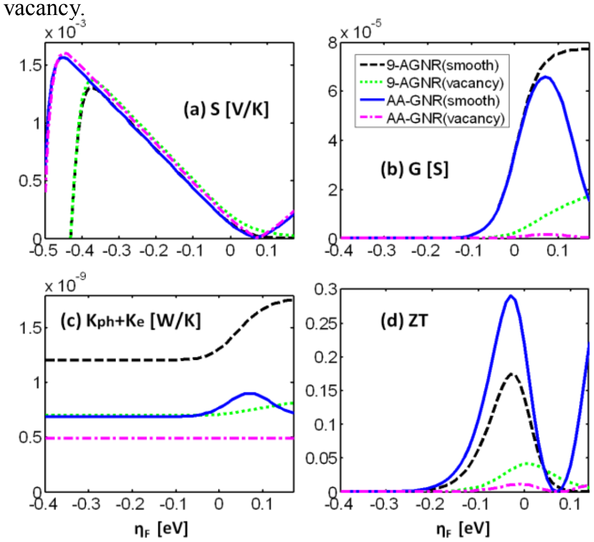

Fig. 2. Thermoelectric properties, (a) S, (b) G, (c) $\kappa_{ph}+\kappa_e$ and (d) ZT as a function of η_F for smooth and defected 9-AGNR and 1 nm kinked AA-GNR.

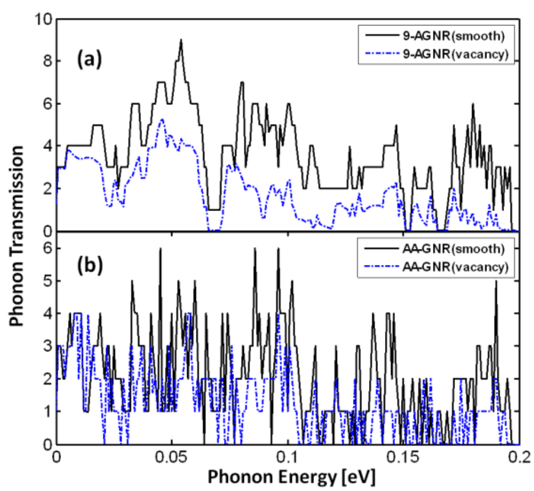

Fig. 3. Phonon transmission as a function of energy for (a) 9-AGNR and (b) 1 nm kinked AA-GNR with smooth and defected structures.

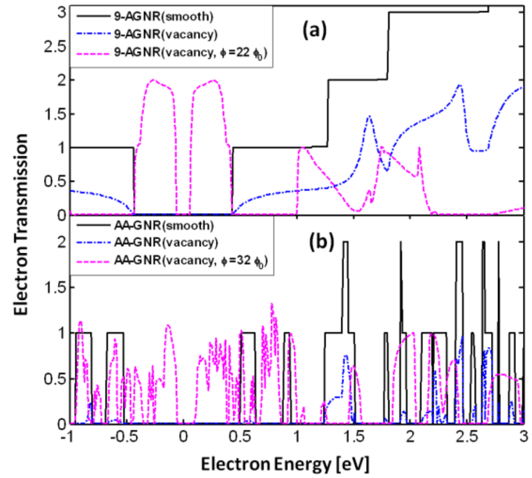

Fig. 4. Electron transmission as a function of energy for (a) 9-AGNR and (b) 1 nm kinked AA-GNR with smooth, defected structures and under magnetic flux $\varphi = 22\varphi_0$.

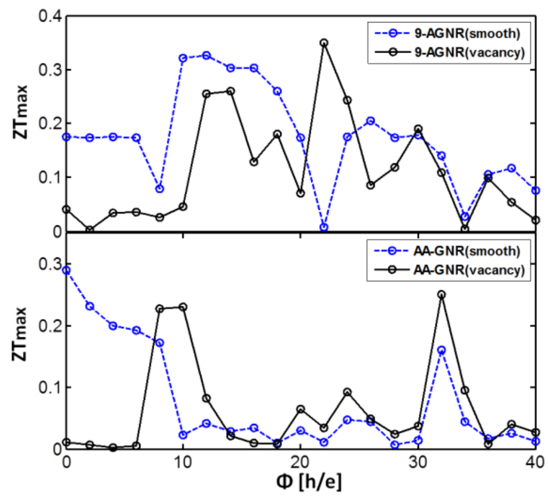

Fig. 5. First peak values of ZT for smooth and defected (a) 9-AGNR and (b) 1 nm kinked AA-GNR under different B-fields.

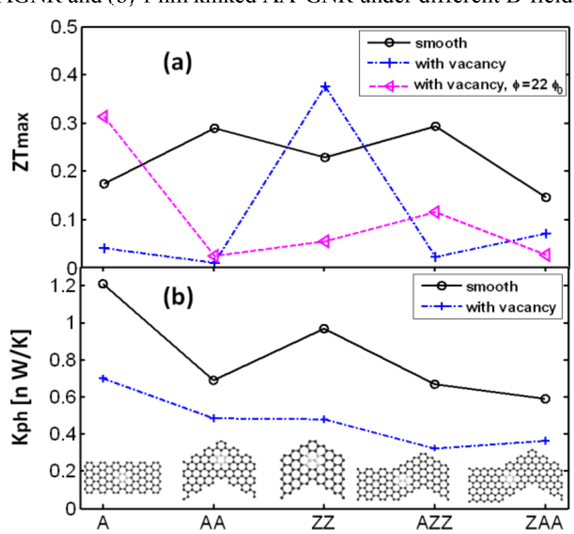

Fig. 6. (a) First peak values of ZT for different kinked GNRs with segment arm width of 1 nm as the transport channel with smooth, defected structures and under magnetic flux $\varphi = 22\varphi_0$. (b) Lattice thermal conductance κ_{ph} for various smooth and defected kinked GNR structures.