The electrical and thermal transport properties of hybrid zigzag graphene-BN nanoribbons

Song Gao¹, Wei Lu¹, Guo-Hui Zheng¹, Yalei Jia¹, and San-Huang Ke¹,², ∗¹

¹MOE Key Laboratory of Microstructured Materials, School of Physics Science and Engineering, Tonji University, 1239 Siping Road, Shanghai 200092, P.R. China
²Beijing Computational Science Research Center, 10 Dongbeiwang West Road, Beijing 100094, P.R. China
E-mail: ∗shke@tongji.edu.cn

Abstract. The electron and phonon transport in hybrid graphene-BN zigzag nanoribbons are investigated by the nonequilibrium Green’s function method combined with density functional theory calculations. A 100% spin-polarized electron transport in a large energy window around the Fermi level is found and this behavior is independent of the ribbon width as long as there contain 3 zigzag carbon chains. The phonon transport calculations show that the ratio of C-chain number to BN-chain number will modify the thermal conductance of the hybrid nanoribbon in a complicated manner.

1. Introduction
Graphene and monolayer hexagonal boron nitride (h-BN) have been attracted much attention for their potential applications in next-generation electronics[1, 2, 3, 4, 5] after they were successfully synthesized in experiments by different methods[6, 7, 8, 9]. On the other hand, the experimental realization of hybrid graphene and h-BN provides a new way to modify the electronic and transport properties of graphene-based materials. In the past few years the electronic and transport properties of graphene nanoribbons (GNRs) modified with BN domains or BN nanoribbons (BNNRs) were investigated extensively and several approaches have been proposed to control the spin transport in GNR/BNNR hybrid systems. For instance, Dong et al. found that a BNNR seamlessly connected between two pieces of GNRs can result in highly spin-polarized transport[10]. He et al. found that the currents with different spin configurations display different behavior in hybridized zigzag BNNRₙ-GNRₘ systems with certain n [11], though they did not find a way to achieve 100% spin polarization of current. While the electronic properties of BNNRs and GNRs have been extensively explored and the thermal transports in BNNRs and GNRs have also got much attention[12, 13, 14], the investigation of thermal transport properties of hybrid GNR/BNNR systems is still limited to our best knowledge. In this work, we investigate the electron and phonon transport properties of parallel hybrid zigzag-edged GNR/BNNR (zGNR/zBNNR) systems with different widths by performing first-principles calculations based on the nonequilibrium Green’s function (NEGF) method combined with density functional theory (DFT).

2. Models and Computation
We first consider a zGNR consisting of 8 zigzag carbon chains (8-zGNR). Then we gradually substitute these carbon chains with BN chains to construct parallel zGNR/zBNNR hybrid nanoribbon systems. The connection is made via the B atoms (system A) or N atoms (system B). The two systems are denoted by
$mBn$ and $mNn$, respectively, with $m$ and $n$ being the numbers of C chains and BN chains, respectively. The two edges of the hybrid nanoribbons are saturated by H atoms. As an example, the structures of the 3B5 and 3N5 nanoribbons are shown in Fig. 1. The majority spin of the spin-polarized edge state localized around the zGNR edge is denoted by $\alpha$-spin.

To investigate the electron and phonon transport through the nanoribbon systems, we adopt the NEGF method combined with first-principles DFT calculations\[15\]. The periodic nanoribbon systems are divided artificially into three subsystems: left lead, scattering region, and right lead (see Fig. 1). For the electron transport, the self-consistent Kohn-Sham Hamiltonian of the scattering region and the self-energies of two semi-infinite leads are used to construct a single-particle Green’s function from which the transmission coefficient at any energy is calculated. The computational techniques have been described in details previously\[15\]. For the phonon transport calculations, the similar NEGF framework is adopted, where the phonon Green’s function of the scattering region can be expressed in terms of the force constant matrix $K_{ij} = \frac{\partial^2 E_{\text{total}}}{\partial u_i \partial u_j}$ and the phonon self-energy $\Sigma_p^{L}$ ($\Sigma_p^{R}$) of the left (right) thermal lead:

$$G_p = \left[ (\omega + i\eta)^2 I - K - \Sigma_p^{L} - \Sigma_p^{R} \right]^{-1}. \tag{1}$$

Then the phonon transmission function can be determined by the Caroli equation: $T(\omega) = Tr[\Gamma_p^L(\omega)G_p(\omega)\Gamma_p^R(\omega)G_p^\dagger(\omega)]$, and the lattice thermal conductance can be calculated by

$$\kappa_p = \frac{1}{2\pi k_B T^2} \int_0^{\infty} (\hbar\omega)^2 \frac{e^{\hbar\omega/k_BT}}{(1 - e^{\hbar\omega/k_BT})^2} T(\omega)d\omega. \tag{2}$$

For the DFT electronic structure calculations, we employ a double-$\zeta$ plus polarization (DZP) numerical basis set for all atomic species\[16, 17\]. The optimized Troullier-Martins pseudopotentials\[18\] are adopted to describe the atomic cores and the generalized gradient approximation (GGA)\[19\] is used for the electron exchange and correlation. The atomic structures of the systems studied are fully optimized by minimizing the forces on each single atom to be smaller than 0.005 eV/Å.

3. Results and Analyses

We first present in Figs. 2 (a) and (b) the transmission functions of pristine 8-zGNR with ferromagnetic (FM) and antiferromagnetic (AFM) spin configurations, respectively. These results agree well with a previous work about the transport in pristine 8-zGNRs\[20\]. Now, let us look at the transmission function of 3B5, as shown in Fig.2(c). One can see that the $\alpha$-spin shows a similar behavior to the FM 8-zGNR while the $\beta$-spin shows a similar behavior to the AFM 8-zGNR. To understand this result, we calculate the band structure of 3B5 (Fig.2(g)) and compare it to that of FM and AFM 8-zGNR (Figs.2(e) and(f)). It can be seen that the $\alpha$-spin bands and $\beta$-spin bands around the Fermi level show indeed similar behavior to those of the FM and AFM 8-zGNR, respectively. As a result, the 3B5 system possesses a complete (100%) spin polarization in a large energy range around the Fermi level. Next, let us look at the transmission function of 3N5, as shown in Fig.2(d). It is quite different from that of 3B5: Now the $\alpha$-spin shows a similar behavior to the AFM 8-zGNR while the $\beta$-spin shows a similar behavior to
given in Fig. In all the cases there exists an energy window of 100% spin polarization around the Fermi level.

Our results show that both the 3B5 and 3N5 systems can provide a large energy window around the Fermi level where the electron transport is 100% spin polarized. To check whether or not this behavior is general for hybrid nanoribbons consisting of 3 zigzag carbon chains, we further calculate other systems with different ribbon widths: 3B3, 3B7, and 3N3, 3N7. The transmission functions are given in Fig. 3. In all the cases there exists an energy window of 100% spin polarization around the Fermi level, indicating that this behavior is general as long as the system contains 3 zigzag carbon chains. Our further calculations show that this behavior is weak or absent for other hybrid nanoribbons whose number of carbon chain is not three.

Finally, let us look at the phonon transport properties of the hybrid nanoribbons. Fig. 4(a) shows the calculated phonon transmission functions (T(E)) of the 8-zGNR (8CC), 8-zBNNR (8BN), 3B5, and 3N5. We note that T(E) of 8CC is overall larger than that of 8BN and those of 3B5 and 3N5 are basically in between. To have a systematic comparison, we give the thermal conductance as a function of the number of carbon chain for system A (Fig. 4(b)) and system B (Fig. 4(c)) under a series of temperatures.

At low temperatures (< 200K) the variation of thermal conductance is very flat for both system A and B. When the temperature is higher than 300K, the \( \kappa_p \) of system A first increases and then decreases for 8BN to 3B5, and then increases linearly for 3B5 to 8CC. As a result, the 3B5 has the minimum thermal conductance. System B has the similar behavior from 8BN to 4N4, but the increase after 4N4 is not linear, being different from system A. The ratio of the C-chain number to the BN-chain number provide a way to modifying the thermal transport properties of the hybrid nanoribbons.

Figure 2. (Color online) Transmission functions of (a) pristine 8-zGNR with FM spin configuration, (b) pristine 8-zGNR with AFM spin configuration, (c) 3B5, and (d) 3N5. (e)-(f) Band structures of the FM 8-zGNR, AFM 8-zGNR, 3B5, and 3N5, respectively.

Figure 3. (Color online) Transmission functions of (a) 3B3, (b) 3B7, and (c) 3N3, (d) 3N7.

FM 8-zGNR, just being opposite to 3B5. This can also be seen in the band structure of 3N5 shown in Fig. 2(h). The result is also a 100% spin polarization around the Fermi level.

Our results show that both the 3B5 and 3N5 systems can provide a large energy window around the Fermi level where the electron transport is 100% spin polarized. To check whether or not this behavior is general for hybrid nanoribbons consisting of 3 zigzag carbon chains, we further calculate other systems with different ribbon widths: 3B3, 3B7, and 3N3, 3N7. The transmission functions are given in Fig. 3. In all the cases there exists an energy window of 100% spin polarization around the Fermi level, indicating that this behavior is general as long as the system contains 3 zigzag carbon chains. Our further calculations show that this behavior is weak or absent for other hybrid nanoribbons whose number of carbon chain is not three.
4. Conclusions
By performing first-principles calculations we have studied the electron and phonon transport properties of hybrid zGNe/BNR nanoribbons with different connection atoms and ribbon widths, which can be now fabricated in experiments. It is found that nanoribbons consisting of 3 zigzag carbon chains will provide a large energy window around the Fermi level where the electron transport is 100% spin polarized, regardless of the ribbon width and connection atom. The phonon transport calculations show that the ratio of C-chain number to BN-chain number will modify the thermal conductance of the hybrid ribbon under different temperatures in a complicated manner.

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