Thermal rectification in thickness-asymmetric graphene nanoribbons

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Abstract

Thermal rectification in thickness asymmetric graphene nanoribbons connecting single-layer with multi-layer graphene is investigated by using classical nonequilibrium molecular dynamics. It is reported that the graphene nanoribbons with thickness-asymmetry have a good thermal rectification. The thermal rectification factor depends on temperature as well as the thickness-ratio of the two-segment. Our results provide a direct evidence that the thermal rectifier can be achieved in a nanostructure crossing two- and three-dimension.
In the past decade, the studies of the mechanism of heat conduction lead to potentially interesting applications based on the possibility to control the heat flow. Thermal rectification has potential applications in nanoscale thermal management such as on-chip cooling and energy conversion by controlling the heat transport. It’s also fundamental in several recently proposed schemes of “thermal circuits” or information processing using phonons. Since the thermal rectifier based on Morse potential has been proposed, many thermal rectifiers have been revealed in various structures. For example, the thermal diode by coupling two nonlinear lattices, the asymmetric nanotubes and graphene, anharmonic graded mass crystals and a spin-boson nanojunction model. Inspired by these theoretical studies, Chang and co-workers have produced a microscopic solid-state thermal rectifier based on carbon nanotubes. Correspondingly, for the study of thermal rectifier, an immediate goal is to find a structure with good thermal rectification factor.

Up to now, although the rectification factor in asymmetric nonlinear Frenkel-Kontorova(FK) lattices has been reported to be about 100, the rectification observed in experiment is only 0.03 \( \sim \) 0.07. The rectifying coefficient of oxide thermal rectifier is also only 0.4, which are far smaller than the predicted value. The theoretical rectification of asymmetric carbon nanotubes is about 0.1 \( \sim \) 0.12. Recently, N. Yang et al. and J. Hu et al. reported a significant rectification of 3.0 and 1.2, respectively, in asymmetric single-layer graphene nanoribbons(GNRs). However, the asymmetric GNRs of triangular shapes only contains one or two atoms in one end. Whether this kind of GNRs can be easily fixed on the substrate is still not clear. From the view of experiment, a few-layer graphene can be fabricated more easily than a single-layer graphene. A thermal diode based on the few-layer graphene should be more popular and its the potential applications may be enormous. Furthermore, a thermal rectification based on the few-layer GNRs can be more helpful to understand the thermal mechanism from two- to three-dimensional materials.

In this letter, we build an asymmetric two-segment GNRs, as shown in Fig.1, which is consisting of a multi-layer graphene in one segment and a single-layer graphene in the other segment. In our studies, the right segment is fixed as a single-layer graphene and the thickness of the right segment can be changed through adding the layers. Here we will display that this asymmetric structure also performs a good rectification. Additionally, the thickness-asymmetric GNRs provide more alternative options for controlling the rectification factor through changing the number of layers in the multi-layer segment. The obtained
results are of significance for understanding the asymmetry and thickness effect on the rectification factor. Therefore, this may open up multi-layer graphene’s applications in the nanoscale thermal diode.

In our simulations, we have used the Tersoff-Brenner potential \[18\] for the C-C bonding interactions in intra-plane and the Lennard-Jones potential \[19\] for van der Waals interactions in the inter-plane. The Nosé-Hoover thermostats are applied to the left and right end of GNRs \[6, 7, 20\]. \(T_T\) (\(T_L\) or \(T_R\)), the set temperatures of the heat baths, are placed in the two ends of the graphene. The average temperature is denoted as \(T_0 = (T_L + T_R)/2\), and the temperature difference is \(\Delta T = (T_L - T_R)/2\). In order to avoid the spurious global rotation of the graphene in the simulations, as shown in Fig.1, we use fixed boundary conditions in the two ends of the graphene (L and R). The fixed region and the heat baths occupy one layer and four layers of atoms, respectively. Free boundary conditions are applied to the other boundaries.

![Schematic diagram showing the multi- and single-layer zigzag edge graphene coupling to two heat baths. The two end of the graphene is fixed and the other boundaries are free. The layers can be changed in multi-layer segment and the right part is fixed as a single-layer. The total length of the GNRs is 5nm.](image)

We integrate the equations of motion for atoms by the Verlet method \[21\]. The total heat flux injected from the heat bath to the system can be obtained by \(J = \sum_i [-\Gamma p_i^2/m_i] = -3\Gamma N k_B T(t)\), where the subscript \(i\) runs over all the particles in the thermostat \[7\]. The direction of the flux \(J_{L\rightarrow R}\) (from L to R) is positive and the direction of the flux \(J_{R\rightarrow L}\)
(from R to L) is negative. The thermal conductivity is calculated from the well-known Fourier’s law \( \kappa = Jl^2/(\Delta TV) \), in which \( l \) and \( V \) are the length and the volume of the total GNRs, respectively. The thickness of each layer of the graphene is 0.335nm. The thermal rectification factor is defined as \( R_0 = |J_{L\rightarrow R} + J_{R\rightarrow L}|/|J_{L\rightarrow R}| \times 100\% \). For comparing edge chirality dependent on the thermal rectification, an armchair and a zigzag graphene are investigated in parallel. The widths of the armchair and the zigzag graphene are 1.6 and 2.1nm, respectively. The thickness asymmetry of the GNRs is \( \gamma = N_L/N_R \), where \( N_L \) and \( N_R \) are the numbers of the layers at left segment and right segment, respectively.

As illustrated in Fig. 2, we present the heat current \( J \) versus \( \Delta T \) for both armchair and zigzag graphene. Here we use an asymmetric GNRs with a two-layer graphene at the left segment and a single-layer graphene at the right segment. When \( \Delta T < 0 \), the heat current increases rapidly with \( \Delta T \), while in the region \( \Delta T > 0 \), the heat current increases slowly with \( \Delta T \), so the system behaves as a thermal rectifier. The inset in Fig. 2 shows that the rectification factor increases with the temperature difference. The thermal rectification can also be described from the thermal conductivity in different directions. We have investigated the thermal conductivity of the thickness-asymmetric GNRs and found significant thermal rectification. It is shown in Figure 3 that the thermal conductivity from the multi-layer graphene (L) to the single-layer graphene (R) is less than that from R to L. In calculating the thermal conductivity of thickness-asymmetric GNRs, the thickness \( (h) \) is taken as the average thickness of GNRs, which is given by \( h = V/(wl) \), where \( V, l \) and \( w \) are, respectively, the volume, the length and the width of GNRs.

In Ref. [7], the overlapping of the phonon spectra of the two atoms near the connecting parts is used to explained the thermal rectification in carbon nanotubes. It has been stated that in this real system, the relationship between the overlap area and the absolute value of the heat flux is the same as that in the one dimensional nonlinear lattice systems [1, 2], in which it is found that matching and/or mismatching of the energy spectra near the interface is the underlying mechanism of the rectification. In the final analysis, the rectification origins from the asymmetric scattering of phonons through two opposite directions. As shown in Fig. 1, when the phonons transfer from single-layer (R side) to multi-layer (L side), i.e., from two- to three-dimensional discontinuous structure, they have more channels to go through smoothly. On the contrary, the phonons from multi-layer (L side) to single-layer (R side), have to crush up near the junction, where three-dimension is connected with two-dimension,
FIG. 2: Heat current as a function of the temperature difference for armchair and zigzag edge asymmetric graphene nanoribbons. The inset is the temperature difference dependence of the thermal rectification factor. The average temperature $T_0 = 320K$.

FIG. 3: Temperature dependence of the thermal conductivity in GNRs for two opposite directions (R to L and L to R). The temperature difference $\Delta T = 20K$.

and then the heat current is suppressed significantly.

Figure 3 also shows that thickness-asymmetric GNRs have the rectifying effect in a wide temperature range from 220k to 380k. The rectification factor depends on the average temperature ($T_0$) as well as on the thickness-ratio ($\gamma$).

As shown in Fig. 4, the rectification factor decreases with the average temperature for different temperature differences. This can be explained as the weaken effect of Umklapp
scattering. The scattering of phonons increases steadily with the temperature, and the asymmetry will be broken. When the temperature increases to the value that Umklapp scattering in the single-layer graphene approaches that in the junction, the rectification factor goes to zero.

FIG. 4: Temperature dependence of thermal rectification factor for two temperature difference. $T_0 = 320K$.

At small temperature difference $\Delta T = 20K$, figure 5 shows that the increasing of asymmetry can improve the thermal rectification factor. The rectification effect is usually enhanced as the asymmetry of GNRs increases. Interestingly, at large temperature difference $\Delta T = 60K$, the rectification factor decreases with the asymmetry of GNRs. Here we propose an elementary explanation on this phenomenon. At large temperature difference, when the number of the layers increases, the thickness strongly suppresses the thermal conductivity of the GNRs [20]. The heat current cannot flow through the GNRs easily in the two directions, the asymmetry dependence of GNRs on the current will become weaker, and then the rectification will decrease. This result provides an important illumination that the temperature difference is also a factor for improving thermal rectification.

In summary, we have studied the thermal rectification of thickness-asymmetric GNRs by using classical nonequilibrium molecular dynamics. The calculated significant thermal rectification effect in thickness-asymmetric GNRs is on the same order of asymmetric single-layer graphene in Ref. [6]. We have also demonstrated that armchair and zigzag edges graphene perform similarly thermal rectification effect. The rectification factor decreases
FIG. 5: Thickness ratio $\gamma$ dependence of thermal rectification factor for two temperature differences 20 and 60K, respectively. $T_0=320K$.

with the temperature. The thickness-ratio between the left and the right segment has a negative effect on the thermal rectification factor at large temperature difference and a positive effect at small temperature difference. Different from the previous thermal rectifiers in graphene, we demonstrate another structure crossing two- and three-dimension, which may be useful for clear understanding the thermal transport in nanoscale materials from low- to high-dimension.

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