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Thermal conductivity of boron nitride nanoribbons: Anisotropic effects and boundary scattering

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ABSTRACT

Previous studies on the thermal conductivity of single-layer boron nitride nanoribbons (BNNRs) are focused on ribbons along the zigzag (ZZ) and the armchair (AC) directions. In this study, we model the thermal conductivity of BNNRs in various transport directions by means of the non-equilibrium molecular dynamics (NEMD) simulations and the Boltzmann transport equation (BTE) under the relaxation time approximation (RTA). In particular, the values of the edge specularity for the boundary scattering, which is closely related to the edge roughness, are obtained at different chiral angles, thereby estimating the anisotropic effects of boundary scattering. It is found that the thermal conductivity has a local maximum at the chiral angle of 19.11°, at which the edge specularity also attains a local maximum. The thermal conductivity generally increases with increasing the ribbon length, yet its value is saturated at a certain length, which significantly depends on the chiral angle. These unusual thermal properties suggest that we may choose optimized structures to achieve better applications of BNNRs in electronic industry.

1. Introduction

Since the successful isolation of single layer graphene, two dimensional materials, such as hexagonal boron nitride (h-BN), Molybdenum disulfide (MoS_2) and Tungsten disulfide (WS_2), have received widespread attention [1–4]. Among these materials, the single-layer boron nitride shares a similar honeycomb lattice structure with graphene, and has also been obtained in experiments [5]. In both graphene and h-BN structures, atoms are bounded by strong sp^2 covalent bonds, leading to their superior in-plane transport properties. Because of their high electrical conductivity and thermopower, graphene nanoribbons (GNRs) could have extremely high thermoelectric figure of merit ZT at room temperature [6–10]. The electric properties of BN, however, is limited by its intrinsic electronic band gap (5 to 6 eV) [11,12]. However, recent studies show that the composite material made of few-layer BN nanosheets and polymethyl methacrylate has remarkable enhancement (17 times) of the thermal conductivity and (2.5 times) of the dielectric constant comparing to the polymer itself, which features the application of high-performance, durable packaging materials for electrical circuits [13,14]. Experimental study by Dean et al. [15] also shows that the electronic transport properties of graphene nanodevices can be tuned via replacing the substrate from SiO_2 to h-BN—the electron mobility of BN-supported graphene (15000–60000 cm^2/V) are remarkably better than that of SiO_2-supported graphene (2000–20000 cm^2/V).

Thermal transport of graphene and boron nitride has also been systematically studied [16–26]. It has been reported that the thermal conductivity of graphene could reach over 5000 W/mK at room temperatures [17,18]. Recently, the thermal conductivity of few-layer h-BNs has also been measured [27]. The results show that the 11-layer h-BN sample could have the thermal conductivity of 360 W/mK, which is slightly smaller than that of the bulk h-BN [28] (~400 W/mK). Despite sharing similar lattice structure with graphene, the single layer h-BN has thermal conductivity significantly lower than graphene. This is often attributed to the stronger phonon–phonon scattering and isotope effect in h-BNs [29,30]. Lindsay et al. [30] showed that by using isotopically pure h-BNs, the thermal conductivity could be enhanced by up to 200 %. Ouyang et al. [31] studied the thermal transport of h-BN nanoribbons (BNNRs) with

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two typical edges and reported that the thermal conductivity of the zigzag (ZZ) BNNR is about 20 % greater than that of the armchair (AC) BNNR. Aksamija et al. [32] investigated the effect of edge roughness scattering on the thermal conductivity of GNRs. They used the Boltzmann transport equation (BTE) with full phonon dispersion relations to calculate the thermal conductivities along all directions.

In the previous BTE studies, however, the boundary scattering is either included only along the transport direction or modeled with using a constant edge roughness. In this study, we employ both the non-equilibrium molecular dynamics (NEMD) simulations and the Boltzmann transport equation (BTE) to investigate the anisotropic behaviors of finite-size BNNRs in the full range of chiral angles. We take into account the full phonon dispersion relations in the first Brillouin zone (FBZ), and consider the phonon–phonon, isotope and boundary scattering. The boundary scattering rate is found by matching the NEMD results with the BTE calculations which requires the input of an edge specularity. The scattering rate is heavily dependent upon the edge roughness, which could be estimated from the distributions of the atoms at the edges. It is shown that the boundary scattering effectively suppresses the thermal conductivity of longer BNNRs. This phenomenon, however, is strongly anisotropic. The thermal conductivities of the BNNRs in between the chiral angle 19.11° and the armchair (AC) direction are 30 % as low as that of the BNNR oriented in the zigzag (ZZ) direction.

2. Model and method

2.1. NEMD method for thermal conductivity

The thermal conductivities of BNNRs are calculated by using the NEMD and employing the BTE. For the NEMD calculations, we use the method developed by Müller-Plathe [33], where the heat flux is imposed on the system by exchanging atoms between hot and cold slabs. The temperature gradient is then extracted from the simulation, and the thermal conductivity can be calculated by Fourier's law

$$\kappa = -\frac{1}{2tA}\left\langle \frac{dT}{dx}\right\rangle^{-1} \sum \frac{m}{2}\left(v_h^2 - v_c^2\right)_{exchange}, \tag{1}$$

where v_h and v_c denote the velocities of the exchanged atoms, t is the simulation time, A is the cross section area, and dT/dx is the average temperature gradient along the heat flux direction. The simulations are performed on BNNRs of the dimensions around 55 × 2000 Å^2 oriented at 16 different chiral angles from $\theta = 0°$ to 30°, as shown in Fig. 1(a). We use the Tersoff-type three-body potential to describe the motion of the atoms [34]. The system is equilibrated to 300 K in a NVE ensemble with the Langevin thermostat before exchanging atoms. The system is divided into 50 slabs, the simulation time step is 1 fs, and the kinetic energies are exchanged every 10 steps.

2.2. BTE analysis for thermal conductivity

The thermal conductivity of lattice vibrations can be modeled by using the BTE under the relaxation time approximations (RTA) [35].

$$\kappa(T) = \sum_j \int_0^{\omega_{max,j}} C_j(\omega, T) v_j^2(\omega) \tau_j(\omega, T) d\omega, \tag{2}$$

where j denotes the phonon mode, v_j is the group velocity, and τ_j is the relaxation time. The heat capacity is defined as $C_j = (\partial f_0/\partial T)\hbar\omega_j D_j(\omega)/V$, where $D_j(\omega)$ is the normalized phonon density of state, V is the volume of unit cell, and $f_0 = [exp(\hbar\omega_j/k_BT)]^{-1}$ is the Bose-Einstein distribution. In this study, only the acoustic normal modes (LA, TA and ZA) are taken into account in BTE calculations, for high-order modes are negligible because of their small phonon group velocities and intensive phonon–phonon scattering. This approach has been employed to calculate the thermal conductivity of graphene [32] and MoS_2 [36], and the results show good agreement with the experimental values.

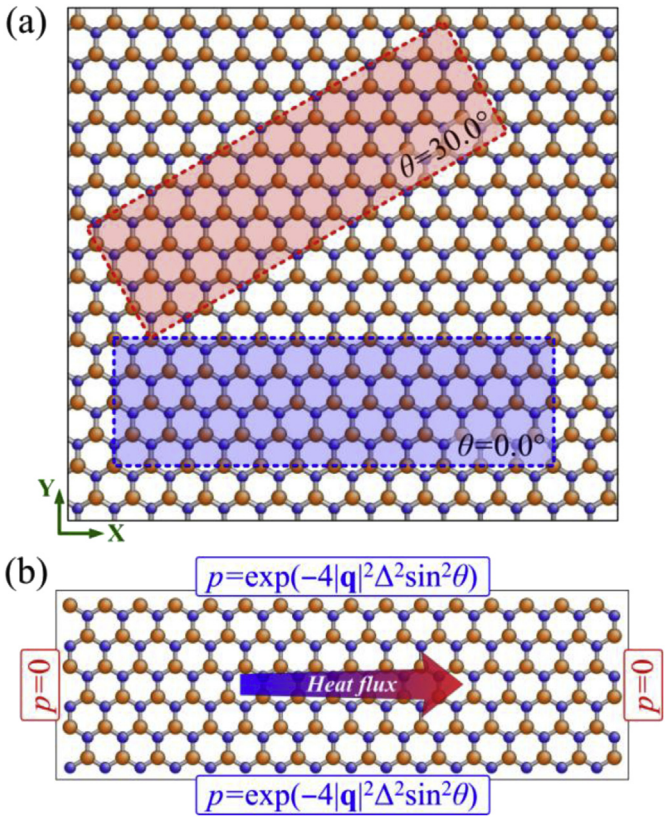

Fig. 1. (a) Lattice structures of the BNNRs (not to scale). In the NEMD simulations, we used 16 BNNRs with different chiral angles (between 0° and 30°). The contour colored by blue denotes the ZZ BNNR ($\theta = 0°$), and by red the AC BNNR ($\theta = 30°$). (b) Scheme of boundary scatterings in BTE analysis. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

In the RTA, phonon collision processes are modeled by means of the effective relaxation time τ_j, which includes various scattering mechanisms:

$$\tau_j^{-1} = \tau_{j,N}^{-1} + \tau_{j,U}^{-1} + \tau_{j,I}^{-1} + \tau_{j,B}^{-1}, \tag{3}$$

where $\tau_{j,I}$ and $\tau_{j,B}$ are associated with the isotope scattering and the boundary roughness scattering, respectively. $\tau_{j,N}$ and $\tau_{j,U}$ stand for the relaxation times of the normal (conservation of crystal momentum) and umklapp (non-conservation of crystal momentum) scattering [37], respectively. In this study, we use the approach proposed by Morelli et al. [38] associated with Matthiessen's rule to obtain the combined relaxation time, which has been employed to calculate the lattice thermal conductivity of group IV and group III–V semiconductors [38], and of graphene nanosheets [39]. The closed form of relaxation time of normal process is given by

$$\tau_{j,N}^{-1} = \frac{Vk_B^3\gamma_j^2\omega_j^2}{m\hbar^2 v_j^5}T^3, \tag{4}$$