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Full paper

Theoretical prediction of thermal transport in BC_2N monolayer

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ABSTRACT

The hybrids of hexagonal boron nitride and graphene have drawn great attentions recently, owing to their many desirable electronic properties, such as a layer-dependent direct bandgap and high carrier mobility. However, the thermal transport properties of them are less investigated and almost unknown. Herein, we implement molecular dynamics simulation to study the thermal transport in one of these hybrids, BC_2N at the first time, including size, temperature and strain effects on the thermal conductivity of BC_2N monolayer. We found that BC_2N owns a strong anisotropy of in-plane thermal transport and the in-plane phonon modes dominate the heat transport, contributing more than 80% in the unstrained BC_2N monolayer at room temperature. Furthermore, for some two-dimensional materials like silicene, the buckled structure is considered as the main reason for the enhanced thermal conductivity resulted from the tensile strain; however, as a planar two-dimensional material without the buckled structure, the enhancement of thermal conductivity of BC_2N is also observed when applying a small tensile strain, which is very interesting and suggests that the buckled structure is not the only mechanism for the tensile strain induced thermal conductivity enhancement. Our findings supplement the influence of strain on phonon transport at nanoscale, and show the BC_2N as a competent candidate for energy devices and electronic thermal management.

1. Introduction

Advances in nanotechnology have enabled engineers to design and tune the electronic, optical, mechanical and thermal properties of materials at nanoscale [1]. The discovery and first exfoliation of graphene from experiments [2] have triggered great interests on finding isomorphic nanostructured materials, and increasingly more similar two-dimensional (2D) materials (e.g. silicene [3–5], hexagonal boron nitride [6,7], monolayer molybdenum disulfide [8,9] and black phosphorus [10,11]) have been reported and synthesized one after another. Due to their many desirable physical properties, these nanostructured materials have been showing great potential for the next-generation electronic, optical and energy conversion devices. However, it is very hard to find an ideal semiconductor or material that satisfies all the technical demands with its intrinsic properties. Owing to a wide bandgap [12] and low carrier mobility [13] in hexagonal boron nitride (h-BN), recently many efforts have been devoted to investigate the hybrids of graphene and h-BN to tailor the electronic properties by utilizing the ultrahigh carrier mobility in graphene [14]. One of the honeycomb structure hybrids called BC_2N, which is composed of commutative C-C and B-N zigzag chains (see the top panel of Fig. 1), comes to our attention. Theoretical calculation from first principles shows that it possesses high carrier mobility and a layer-dependent direct bandgap, which enables the potential applications in nanoelectronics and optoelectronics [15]. In addition, it has been successfully synthesized by chemical vapor deposition method [16] in experiment and holds a 2.0 eV direct bandgap [17] for monolayer from experimental measurement. Because of prior studies almost focusing on the electronic properties of BC_2N and lacking the knowledge of its thermal transport properties, both theoretical calculations and experimental studies should be performed to investigate the thermal properties of BC_2N, if driving its applications in energy conversion such as thermal management and thermoelectrics. These work are important as well as developing new materials to the continued technological advancements, which are strongly in line with the goals of the Materials Genome Initiative [18–20].

Compared with their bulk counterparts, the thermal properties of nanostructured materials can be also easily tuned by introducing defects [21], doping [22], changing the geometry shapes [23,24] and applying external strains [25,26]. Due to the flexibility and robustness of strain engineering, we continue to study the thermal response of BC_2N to external strains. From theoretical calculations, many

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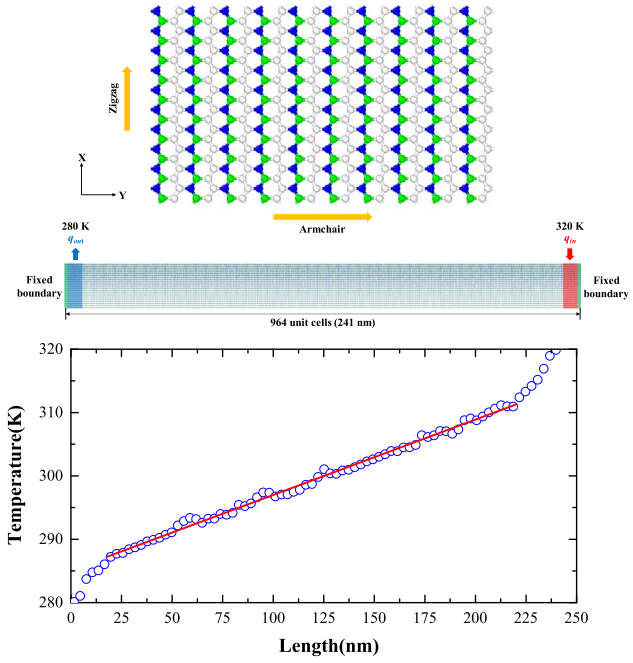

Fig. 1. (Top) Configure of BC_2N monolayer. The zigzag and armchair directions are placed along the x and y directions, respectively. Color code: white, C; green, B; blue, N. (Middle) Snapshot of typical zigzag SLBCN in NEMD simulation. The heat flux is along the zigzag direction. The left and right ends of the system are fixed during the simulation and are highlighted in green. (Bottom) The corresponding temperature profile of SLBCN in NEMD simulation and the red line represents the linear fitting.

researches have observed that the thermal conductivities of nanostructured materials like graphene [27], monolayer MoS_2 [28], h-BN nanotube [29], carbon nanotube, silicon and diamond thin film or nanowire [30] decrease significantly when applying a tensile strain. They believed that stretching and tensile strain leads to the phonon softening, causing a great reduction of phonon group velocity. However, this is not always the case. Xie et al. [31] investigated the effects of biaxial tensile strain on thermal conductivity of single-layer silicene from first principle calculations, finding that within 10% tensile strain the thermal conductivity of silicene can increase dramatically. Using the classical molecular dynamics (MD), similar conclusions were drawn by Hu et al. [26] and Pei et al. [32] that silicene shows an unusual thermal response to uniaxial tensile strain. They attributed this anomalous behavior to the stiffening of flexural phonon modes, reduced scatterings of longitudinal acoustic (LA) and transverse acoustic (TA) phonons with flexural acoustic (ZA) phonons, and enhanced lifetime of ZA phonons after the tensile strain because the buckled structure of silicene becomes more planar, leading to the reflectional symmetry which limits the phonon scattering involving flexural phonons with odd number [33]. Besides, recently molecular dynamics simulation [34] has shown that the thermal conductivity of single-layer black phosphorus (phosphorene) may increase under a small tensile strain, while Jain and McGaughey [35] assumed that owing to the reduced sound velocity in the single-layer black and blue phosphorus after applying tensile strains from first principle calculation, the thermal conductivities of them might decrease. Combined with first principle calculation, Ong et al. [36] also investigated the thermal conductance of single-layer phosphorene by using the nonequilibrium Green's function and found that the armchair-oriented thermal conductance always decreases whatever a zigzag- or an armchair-oriented strain is applied. This indicates that the case of phosphorene with the puckered structure becomes much more complicated and the strain effects on phonon transport are still obscure.

In this work, all from classical molecular dynamics and lattice dynamics, we investigate the thermal transport in BC_2N monolayer. Length-, temperature- and strain-dependent thermal conductivities are then calculated with analyzing the corresponding phonon properties. Like silicene, the in-plane phonon modes dominate the heat transport in BC_2N. Our work reveals a strong anisotropic in-plane thermal conductivity and a strain-tuned high thermal conductivity of BC_2N, demonstrating an ideal candidate for electronic heat dissipation and thermal management.

2. Methodology

All the MD simulations are performed by using Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) package [37] with partial modification to study the thermal transport properties of BC_2N monolayer. Periodic boundary condition is applied in the x (zigzag) and y (armchair) directions, while non-periodic boundary condition is used in z (out-of-plane) direction to simulate the BC_2N monolayer as a 2D material. To describe the atomic interactions in single-layer BC_2N (SLBCN), a Tersoff potential [38,39] developed by Kınacı et al. [40] is used to mimic the heat transport in SLBCN and thermal conductivity is calculated by using the non-equilibrium molecular dynamics (NEMD) [41]. We first use the lattice constants which are predicted from first principle calculation [15] to construct a 10 × 10 × 1 supercell, then perform the energy minimization to obtain the lattice constants under the Tersoff potential, to remove the difference between the fitting semi-empirical force field and full quantum mechanics description based on density functional theory. Since a very long ribbon-like model for NEMD prediction cannot suffer one bar pressure, we also compute the lattice constants of BC_2N at room temperature (300 K) and one bar pressure by using a 10 × 10 × 1 supercell, from which the subsequent modelling systems for MD simulation are constructed to mimic the real one as closely as possible.

The thermal conductivities are predicted from NEMD, specifically the constant temperature gradient method. At the beginning, choosing a timestep as 1.0 fs, we relax the model at 300 K in an NVT (constant particles, volume and temperature) ensemble for 500 ps by using a Nosé-Hoover thermostat [42] then switch into an NVE (constant particles, volume and no thermostat) ensemble for 500 ps to both reach a thermal equilibrium. When calculating the thermal conductivity, as shown in the middle panel of Fig. 1, the left and right ends subtracting several fixed atoms (walls) are chosen as the heat sink and source, controlled as the constant temperature (280 K and 320 K respectively) in two NVT ensembles, while the rest of atoms run in an NVE ensemble to conserve energy. To establish a steady temperature gradient, the total running time for NEMD simulation is about 20 ns with a 200 ps for every individual spatial average. For computing the heat flux, the thickness of BC_2N monolayer is chosen as 3.6 Å which is the Van Der Waals diameter of boron atom. The thermal conductivity κ of SLBCN from NEMD can be obtained by Fourier's Law

$$\kappa = \frac{J}{\partial T/\partial L}, \tag{1}$$

where J is the heat flux and $\partial T/\partial L$ denotes the temperature gradient along the heat flux from a linear fitting (see the bottom panel of Fig. 1). Two very small nonlinear regions could be observed near the heat source and sink due to the phonon scattering with heat source and sink. Additionally, when investigating the temperature- and strain-dependent thermal conductivities, the selected timestep should be reduced to 0.5, 0.3 or 0.25 fs because the SLBCNs at some high temperatures and large strains are unstable.

3. Results and discussion

3.1. Size effect

The length-dependent zigzag- and armchair-oriented thermal conductivities of SLBCN at room temperature are first calculated from