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Commensurate lattice constant dependent thermal conductivity of misoriented bilayer graphene

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ABSTRACT

Misorientation of two layers of bilayer graphene leaves distinct signatures in the electronic properties and the phonon modes. The effect on the thermal conductivity has received the least attention and is the least well understood. In this work, the in-plane thermal conductivity of misoriented bilayer graphene (m-BLG) is investigated as a function of temperature and interlayer misorientation angle using nonequilibrium molecular dynamics (NEMD). The central result is that the calculated thermal conductivities decrease approximately linearly with the increasing lattice constant of the commensurate m-BLG unit cell. Comparisons of the phonon dispersions show that misorientation has negligible affect on the low-energy phonon frequencies and velocities. However, the larger periodicity of m-BLG reduces the Brillouin zone size to the extent that the zone edge acoustic phonons are thermally populated. This allows Umklapp scattering to reduce the lifetimes of the phonons contributing to the thermal transport, and consequently, to reduce the thermal conductivity. This explanation is supported by direct calculation of reduced phonon lifetimes in m-BLG based on density functional theory (DFT).

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1. Introduction

The record high thermal conductivity of graphene has created widespread interest and may lead to its applications in thermal management [1,2]. The room temperature thermal conductivity (κ) of single layer graphene (SLG) is approximately 3000 – 5000 W/m·K, while the room temperature thermal conductivity of few-layer graphene (FLG) ranges from 1300 W/m·K to 2800 W/m·K [1,3,4]. Similar values have also been obtained from theoretical studies [5–7]. There are many factors that influence the thermal conductivity of graphene such as vacancies, chirality, isotope [8], wrinkles, number of layers, etc. Among them, the effect of interlayer misorientation on the in-plane thermal conductivity of bilayer graphene (BLG) has been the least studied, and it is the focus of this work.

In graphene, or BLG, heat is carried by the low-energy vibrational (phonon) modes [9]. Anything that alters the low-energy phonon spectrum or the phonon scattering can affect the thermal conductivity. Experimentally, Raman spectroscopy has been extensively used to probe the zone-center vibrational properties of graphene, AB-BLG, and misoriented-BLG (m-BLG) [10–17]. It has been used to measure the misorientation angle dependence of the high-energy optical phonons of the G and 2D peaks of m-BLG, and it has also been used to measure the new peaks that appear in the low-energy range of 90–200 cm^{-1} in the vicinity of the original ZO′ breathing mode [13,14,18]. The position, intensity, and width of the Raman 2D peak can be used to identify the m-BLG misorientation angle [12,16]. Misorientation also affects the electron-phonon interlayer and intralayer interactions [19,20], and Raman spectroscopy has very recently been used to distinguish the interlayer from the intralayer interactions [20]. While the Raman studies are useful for understanding the optical phonon branches and their interactions with electrons, heat is carried by the low-energy acoustic modes over a range of wavevectors which the Raman studies do not probe.

Recently, the in-plane thermal conductivities of two suspended BLG samples were experimentally measured over a range of

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temperatures from 300 K to 650 K [21]. One sample was aligned AB-stacked BLG (AB-BLG), and the other sample was m-BLG with a misorientation angle of 32.2°. The average κ values of the m-BLG were uniformly lower than those of the AB-BLG. There was considerable experimental uncertainty of the data, with error bars of up to 40%, and the measurements were taken from a single m-BLG sample. A theoretical study of m-BLG nanoribbons found strong edge effects and an increase of thermal conductivity for misorientation angles of 22.5° and 30.0° compared to that of an AB stacked nanoribbon [22]. Overall, the effect of the misorientation angle on the in-plane thermal conductivity of BLG is still an open question.

The existing computational research on the phonon properties of m-BLG indicates that misorientation only slightly affects the phonon frequencies, density of phonon modes and the specific heat above room temperature [18,23–26]. New low-energy $q = 0$ modes in the m-BLG $\omega - q$ dispersion naturally occur due to zone-folding. Considering the simple expression relating the thermal conductivity, specific heat, velocity, and effective mean-free path, $\kappa = \frac{1}{3} C_v \times v \times l_{\text{eff}}$, one would infer that interlayer misorientation should not significantly affect the in-plane thermal conductivity provided that l_{eff} is not significantly changed. However, zone-folding reduces the size of the Brillouin zone (BZ) and opens up new Umklapp scattering channels that result in increased Umklapp scattering and a reduced mean free path [21].

Usually, Umklapp scattering would be expected to have little effect on the heat transport by low-energy phonons with small wave vector q. The thermal conductivity depends on the low-energy region, while Umklapp processes dominate the high-energy region. However, the periodicity introduced by the moiré pattern or determined by the commensurate unit cell can be very long. The lattice constant of the commensurate unit cell with the smallest misorientation angle that we consider of 13.17° is 1.07 nm, corresponding to a BZ Γ-K path length of 3.91 nm^{-1}. At wave vector K, the phonon frequency of the LA branch is approximately 360 cm^{-1} corresponding to an energy of 45 meV, which is less than 2 $k_B T$ at room temperature [23]. Thus, the severe reduction of the BZ brings the zone edges into the low-energy range where Umklapp processes could play a role in the room temperature thermal transport.

2. Method and computational approach

The starting point of the theoretical investigation is the construction of the misoriented bilayer atomic structures using commensurate rotation angles. These special angles ensure that the overall structure remains periodic, albeit with a much longer periodicity. Commensurate misorientation angles are given by Ref. [27].

$$\cos\theta = \frac{n^2 + 4nm + m^2}{2(n^2 + nm + m^2)}, \tag{1}$$

where m and n are non-negative integers with $m \leq n$. The commensurate unit cell vectors $\mathbf{c}_1$ and $\mathbf{c}_2$ are $\mathbf{c}_1 = n\mathbf{a}_1 + m\mathbf{a}_2$ and $\mathbf{c}_2 = -m\mathbf{a}_1 + (m+n)\mathbf{a}_2$. We will refer to $\mathbf{c}_1$ and $\mathbf{c}_2$ as the primitive lattice vectors, their magnitude as the primitive lattice constant, and a unit cell constructed with $\mathbf{c}_1$ and $\mathbf{c}_2$ as a primitive cell, since it is the smallest periodic cell that can be constructed for a given misorientation angle θ. Due to the 6-fold rotational symmetry of graphene, we only need to consider misorientation angles between 0° and 60°. The misorientation angles considered here are 0° (AB-stacking), 13.17°, 21.78°, and 32.20°. The three angles were chosen since they give the three smallest primitive cells as shown in Fig. 1. A 27.79° primitive cell is the same size as the 32.30° primitive cell, but the 32.30° angle was chosen, since it corresponds to a clockwise rotation of the upper layer in the same sense as the 13.17° and 21.78° angles, and this is also the misorientation angle of the m-BLG sample measured in Ref. [21]. The next larger commensurate primitive cell corresponds to a rotation angle of 17.89° with a primitive lattice constant of 13.69 Å containing 124 atoms. The sizes of the commensurate primitive cells quickly increase from there, and a table of commensurate angles and primitive lattice constants is given in Ref. [28].

The NEMD simulations require a rectangular unit cell, so we define orthogonal unit cell vectors $\mathbf{r}_1$ and $\mathbf{r}_2$ as $\mathbf{r}_1 = \mathbf{c}_1$ and $\mathbf{r}_2 = 2\mathbf{c}_2 - \mathbf{c}_1$. The number of atoms N in the rectangular unit cell is $N = 8(n^2 + nm + m^2)$. A top view of the rectangular unit cells are shown in Fig. 1 underneath their respective primitive cells. The rectangular unit cell of AB-BLG can be made smaller than in Fig. 1a. It is constructed to be the same size as the 21.78° unit cell, so that the thermal conductivities can be compared at a constant width and a constant length. The rectangular unit cell is repeated multiple times in the direction of the heat transport. It is also repeated in the direction perpendicular to the direction of heat flow to ensure that the width is sufficiently large, so that the calculated thermal conductivity values are independent of the width [29].

For the NEMD simulations, the BLG structures are divided into $2N$ identical slabs along the transport direction where N is a positive integer, usually not less than 10 for the accuracy of calculation. We will refer to the slabs as 'NEMD slabs', since they are created purely for the NEMD calculation. For each NEMD slab, we obtain one statistical average temperature. The hot region is at the center in NEMD slab $N+1$, and the cold regions are at the sample ends in NEMD slabs 1 and $2N$. The geometry is illustrated in the inset of Fig. 2b.

The NEMD simulations are implemented in the LAMMPS code [30]. The time step is 0.2 fs. A reactive empirical bond order (REBO) potential [31] is used for the in-plane bonding interactions, and the Lennard-Jones (LJ) potential is included for the interlayer van der Waals (vdW) forces with a well-depth energy of 2.96 meV and an equilibirum distance of 0.334 nm [32].

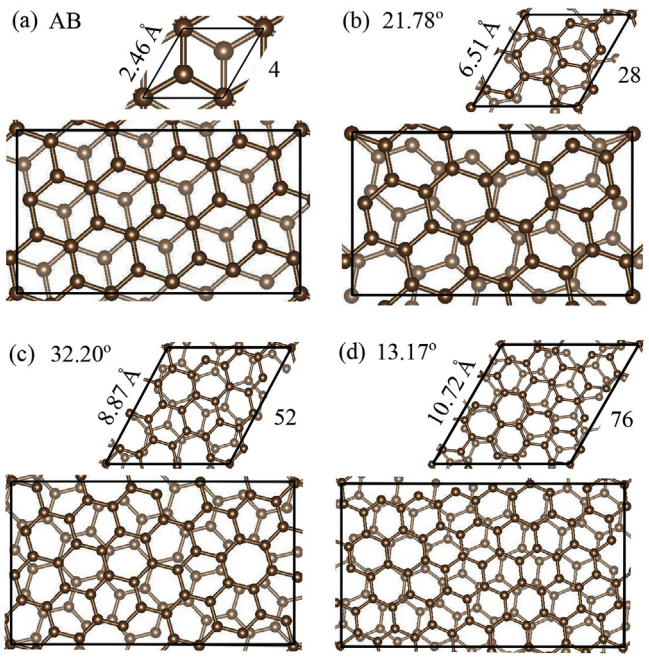

Fig. 1. Top views of the primitive cells and the rectangular unit cells of (a) AB-BLG, (b) 21.78° m-BLG, (c) 32.20° m-BLG, (d) 13.17° m-BLG. The value of the primitive lattice constant is shown along the left edge of each primitive cell, and the number of atoms in each primitive cell is shown to the right of each cell. (A colour version of this figure can be viewed online.)

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