



Kapitza thermal resistance across individual grain boundaries in graphene



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ABSTRACT

We study heat transport across individual grain boundaries in suspended monolayer graphene using extensive classical molecular dynamics (MD) simulations. We construct bicrystalline graphene samples containing grain boundaries with symmetric tilt angles using the two-dimensional phase field crystal method and then relax the samples with MD. The corresponding Kapitza resistances are then computed using nonequilibrium MD simulations. We find that the Kapitza resistance depends strongly on the tilt angle and shows a clear correlation with the average density of defects in a given grain boundary, but is not strongly correlated with the grain boundary line tension. We also show that quantum effects are significant in quantitative determination of the Kapitza resistance by applying the mode-by-mode quantum correction to the classical MD data. The corrected data are in good agreement with quantum mechanical Landauer-Bütticker calculations.

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1. Introduction

Graphene [1], the famous two-dimensional allotrope of carbon, has been demonstrated to have extraordinary electronic [2], mechanical [3], and thermal [4] properties in its pristine form. However, large-scale graphene films, which are needed for industrial applications are typically grown by chemical vapor deposition [5] and are polycrystalline in nature [6], consisting of domains of pristine graphene with varying orientations separated by grain boundaries (GB) [7–9]. They play a significant or even dominant role in influencing many properties of graphene [10,11].

One of the most striking properties of pristine graphene is its extremely high heat conductivity, which has been shown to be in excess of 5000 W/mK [4,12]. Grain boundaries in graphene act as line defects or one-dimensional interfaces which leads to a strong reduction of the heat conductivity in multigrain samples [13,14]. The influence of GBs can be quantified by the Kapitza or thermal

boundary resistance R . The Kapitza resistance of graphene grain boundaries has been previously computed using molecular dynamics (MD) [15,16] and Landauer-Bütticker [17,18] methods, and has also been measured experimentally [19]. However, these works have only considered a few separate tilt angles, and a systematic investigation on the dependence of the Kapitza resistance on the tilt angle between any two pristine grains is still lacking. The relevant questions here concern both the magnitude R for different tilt angles and possible correlations between the structure or line tension of the GBs and the corresponding value of R .

Modelling realistic graphene GBs has remained a challenge due to the multiple length and time scales involved. Recently, an efficient multiscale approach [20] for modelling polycrystalline graphene samples was developed based on phase field crystal (PFC) models [21,22]. The PFC models are a family of continuum methods for modelling the atomic level structure and energetics of crystals, and their evolution at diffusive time scales (as compared to vibrational time scales in MD). The PFC models retain full information about the atomic structure and elasticity of the solid [22]. It has been shown [20] that using the PFC approach in two-dimensional space one can obtain large, realistic and locally relaxed

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microstructures that can be mapped to atomic coordinates for further relaxation in three-dimensional space with the usual atomistic simulation methods.

In this work, we employ the multiscale PFC strategy of Ref. [20] to generate large samples of tilted, bicrystalline graphene with a well-defined GB between the two grains. These samples are then further relaxed with MD at $T = 300$ K. A heat current is generated across the bicrystals using nonequilibrium MD (NEMD) simulations, and the Kapitza resistance is computed from the temperature drop across the GB. We map the values of $R(\theta)$ for a range of different tilt angles θ and demonstrate how R correlates with the structure of the GBs. Finally, we demonstrate that quantum corrections need to be included in R to obtain quantitative agreement with experiments and lattice dynamical calculations.

2. Models and methods

2.1. PFC models

PFC approaches typically employ a classical density field $\psi(\mathbf{r})$ to describe the systems. The ground state of ψ is governed by a free energy functional $F[\psi(\mathbf{r})]$ that is minimized either by a periodic or a constant ψ , corresponding to crystalline and liquid states, respectively. We use the standard PFC model

$$F = \int d\mathbf{r} \left(\frac{1}{2} \psi \left[\varepsilon + (q^2 + \nabla^2)^2 \right] \psi + \frac{1}{3} \tau \psi^3 + \frac{1}{4} \psi^4 \right), \quad (1)$$

where the model parameters ε and τ are phenomenological parameters related to temperature and average density, respectively. The component $(q^2 + \nabla^2)^2$ penalizes for deviations from the length scale set by the wave number q , giving rise to a spatially oscillating ψ and to elastic behaviour [21,22]. The crystal structure in the ground state is dictated by the formulation of F and the average density of ψ , and for certain parameter values the ground state of ψ displays a honeycomb lattice of density maxima as appropriate for graphene [20].

The PFC calculations are initialized with symmetrically tilted bicrystals in a periodic, two-dimensional computational unit cell. The initial guess for the crystalline grains is obtained by using the one-mode approximation [22].

$$\psi(x, y) = \cos(qx) \cos\left(\frac{qy}{\sqrt{3}}\right) - \frac{1}{2} \cos\left(\frac{2qy}{\sqrt{3}}\right), \quad (2)$$

and by rotating alternately by $\pm\theta$. The tilt angle between two adjacent grains is $\theta - (-\theta) = 2\theta$, which ranges from $2\theta = 0^\circ$ to $2\theta = 60^\circ$ (see Fig. 1 for examples). We consider a subset of the tilt angles investigated in Ref. [20], with the exact values being listed in Table 1. The rotated grains and the unit cell size are matched together as follows: if just one of the rotated grains filled the whole unit cell, it would be perfectly continuous at the periodic edges. Along both interfaces, narrow strips a few atomic spacings wide are set to the average density – corresponding to a disordered state – to give the grain boundaries some additional freedom to find their lowest-energy configuration. We assume non-conserved dynamics to relax the systems in analogy to chemical vapor deposition [23] – the number of atoms in the monolayer can vary as if due to exchange with a vapor phase. In addition, the unit cell dimensions are allowed to vary to minimize strain. Further details of the PFC calculations can be found in Ref. [20]. The relaxed density field is mapped to a discrete set of atomic coordinates suited for the initialization of MD simulations [20].

2.2. NEMD simulations

We use the NEMD method as implemented in the GPUMD (graphics processing units molecular dynamics) code [24–26] to calculate the Kapitza resistance, using the Tersoff [27] potential with optimized parameters [28] for graphene. The initial structures obtained by the PFC method are rescaled by an appropriate factor to have zero in-plane stress at 300 K in the MD simulations with the optimized Tersoff potential [28].

In the NEMD simulations, periodic boundary conditions are applied in the transverse direction, whereas fixed boundary conditions are applied in the transport direction. We first equilibrate the system at 1 K for 1 ns, then increase the temperature from 1 K to 300 K during 1 ns, and then equilibrate the system at 300 K for 1 ns. After these steps, we apply a Nosé-Hoover chain of thermostats [29–31] to the heat source and sink, choosing as two blocks of atoms around the two ends of the system, as schematically shown in Fig. 2. The temperatures of the heat source and sink are maintained at 310 K and 290 K, respectively. We have checked that steady state can be well established within 5 ns. In view of this, we calculate the temperature profile $T(x)$ of the system and the energy exchange rate Q between the system and the thermostats using data sampled in another 5 ns. The velocity-Verlet integration scheme [32] with a time step of 1 fs is used for all the calculations. Three independent calculations are performed for each system and the error estimates reported in Table 1 correspond to the standard error of the independent results.

In steady state, apart from the nonlinear regions around the heat source and the sink intrinsic to the method, a linear temperature profile can be established on each side of the GB, but with an inherent discontinuity (temperature jump) at the GB. An example of this for the system with $2\theta = 9.43^\circ$ is shown in Fig. 3. The Kapitza resistance R is defined as the ratio of the temperature jump ΔT and the heat flux J across the grain boundary:

$$R = \frac{\Delta T}{J}, \quad (3)$$

where J can be calculated from the energy exchange rate Q (between the system and thermostat) and the cross-sectional area S (graphene thickness is chosen as 0.335 nm in our calculations), *i.e.* $J = Q/S$.

3. Results and discussion

It is well known [15,16,33] that the calculated Kapitza resistance depends on the sample length in NEMD simulations. Fig. 4 shows the calculated Kapitza resistance R in the $2\theta = 9.43^\circ$ case as a function of the sample length L_x . Using fixed boundary conditions as described above, R saturates at around $L_x = 400$ nm. On the other hand, using periodic boundaries as described in Ref. [15], R converges more slowly. To this end, we have here used fixed boundary conditions and a sample length of 400 nm for all the systems. The calculated temperature jump ΔT , heat flux J , and Kapitza resistance R in the 13 bicrystalline systems are listed in Table 1.

The Kapitza resistance calculated from the heat flux does not contain any information on the contributions from individual phonon modes. Methods of spectral decomposition of both the heat current (flux) [37–42] and the temperature [43] within the NEMD framework have been developed recently. Here, we use the spectral decomposition formalism as described in Ref. [42] to calculate the spectral conductance $g(\omega)$ of the $2\theta = 9.43^\circ$ system. In this method, one first calculates the following nonequilibrium heat current correlation function (t is the correlation time):

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