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Transient and steady state heat transport in layered materials from molecular dynamics simulation

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ABSTRACT

In this paper, both transient and steady state heat transfer in graphite are investigated with molecular dynamics (MD) simulations. The simulation results demonstrate that elastic anisotropy controls heat transfer in the transient state, which makes the outermost isothermal surface of temperature distribution similar to the phonon group velocity surface that has a shape of very flat ellipse in layered materials. In steady state, with the help of phonons engaging in sufficient scattering, the basal plane phonon modes determine the thermal conductivity along all directions except those very close to the cross plane. Our simulation results confirm that the classical theoretical model can accurately predict the thermal conductivity along arbitrary directions in layered materials.

1. Introduction

The properties of layered materials such as graphite and molybdenum disulfide are typically anisotropic, including mechanical [1–4], electrical [5–8] and thermal properties [9–12]. Layered materials often show a variety of new physical properties that cannot be observed in isotropic materials, such as phonon focusing phenomena [13,14] and bending of the heat flux [15]. Due to anisotropy in the thermal properties, heat can be directed in a different direction from that in which the thermal gradient is applied, which offers more opportunities for layered materials in new applications such as thermal cloaks [16], heat management [17,18], thermoelectric devices [19] and building envelopes [20]. For example, graphite and few-layer graphene can be used as not only heat spreaders [18] due to their high basal plane thermal conductivity but also good thermal insulators due to the extremely low cross-plane thermal conductivity [21]. Thus, investigating the anisotropic properties of layered materials is of fundamental and practical importance.

As a widely used layered material, the anisotropic thermal properties of graphite have been extensively explored by experimental measurements [11,22–24], molecular dynamics (MD) simulations [14,25] and lattice dynamics [26–28] (LD) methods. All results showed that the basal plane thermal conductivity is about two orders of magnitude higher than the cross-plane value, an indicator of strong anisotropy. It is worth mentioning that those investigations were carried out in steady state, i.e. phonons in the system undergo sufficient scattering to smooth the temperature. However, many devices experience transient thermal loads in various applications [18,29,30]. For example, with the continued decreasing of microelectronic device size, the rate of local heat generation increases [31] and the device switching time decreases [32], leading to large transient thermal loads. Transient phonon transport is also an important thermal behavior in pulsed-laser processing of materials [33], transient heat flux control of thermal cloaks [29] and transient thermal grating techniques [34]. Therefore, given the potential use of layered materials in these applications, investigating the transient phonon transport in layered materials is significant and important.

Different from steady state transport, transient phonon transport lacks sufficient scattering and the transport velocity is determined by the elastic constant in the corresponding directions. For example, due to the weak interlayer coupling strength in graphite, the phonon velocity along the cross-plane direction is quite slow [35,36], In addition, since the elastic anisotropy of solid is related to the phonon velocity surface of acoustic waves [37], based on the weak interlayer coupling strength and anisotropic dispersion [38], it can be inferred that the phonon velocity along all other directions should be far smaller than that along the graphite basal plane direction. Thus, intuitively, the transient heat transfer speed along the basal plane direction should be far larger than that along any other directions in layered structures. According to the widely accepted theoretical model [1,15], thermal conductivity along the lattice plane with an intersection angle of θ with respect to the basal plane in anisotropic materials can be predicted by

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$$k_\theta = k_c \sin^2\theta + k_{in}\cos^2\theta \quad (1)$$

where k_c and k_{in} are the thermal conductivities along the cross-plane and basal plane directions respectively. The intersection angle θ between the y-z plane and the basal plane is depicted in Fig. 1. This formula implies that the thermal conductivity along the lattice plane having a large intersection angle θ with respect to the basal plane still has a large value, in contrast with the previously described physical picture for the heat transfer in the transient state. Given that the theoretical model is formulated for steady state conditions in which phonons experience sufficient scattering, we believe this difference is due to phonon scattering. Therefore the heat transfer in both transient and steady states should be investigated. Here, we refer to this classical theoretical model as the projection model since it is derived based on the projection relation [15].

In this paper, transient and steady phonon transport is explored using MD simulations. The transient simulation results show that phonon transport is determined by the elastic anisotropy. In steady state, however, both the elastic anisotropy and phonon scattering affect the heat transfer, causing the thermal conductivity along the lattice plane having a large intersection angle with respect to the basal plane to maintain a large value. Moreover, it is found that the basal plane phonon modes are the main contributors to heat transfer along all directions except those very close to the cross plane along which the cross-plane phonon modes are dominant, similar to the prediction by formula (1). Although the MD simulation uses graphite as an example, the conclusion is applicable to other layered materials, like MoS_2 [39], WSe_2 crystal [40], black phosphorus [41], layered muscovite crystal [42], boron nitride [43], and multilayer films [44]. Moreover, the non-equilibrium MD process is similar to what is done in experimental research. Therefore, our work provides a model to investigate the heat transport along arbitrary directions in layered materials.

2. Simulation method

Fig. 1 shows the simulated structure. The structure is constructed in three steps. First, two-layer graphene sheets in the x-y plane are generated, followed by a rotation along the y-axis with an angle ϕ. Then we replicate the rotated structure in the x-z plane to create a graphite region with size large enough. Finally, the graphite region is carefully cut to be orthogonal domain to simplify the simulation.

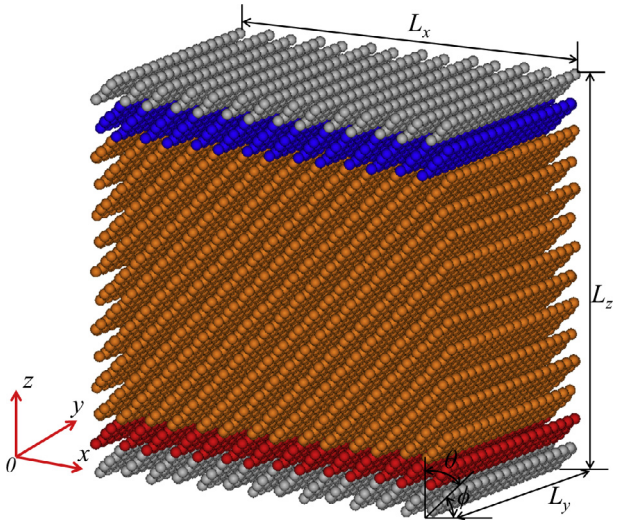

Fig. 1. The model used in the non-equilibrium molecular dynamics simulation. L_x, L_y, L_z are defined as the system size in the x, y and z directions respectively. The angle ϕ is defined as the graphite basal plane rotating along the y axis. The angle θ is the intersection angle between the y-z plane and the basal plane. The two grey slabs with film thickness 0.5 nm are fixed working as the solid wall in the NEMD simulation. The slab composed of the red atoms is defined as the hot reservoir while the cold reservoir is plotted with blue. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

In this paper, both transient and steady state MD simulations are carried out. In the steady state simulation, as shown in Fig. 1, the grey atoms in the top and bottom slabs with a thickness of 0.5 nm are fixed without thermal vibration. The adjacent two 0.5 nm thick slabs are set as the hot and cold reservoirs, respectively. All simulations here are implemented using the LAMMPS package [45]. Each simulation is equilibrated with a constant particle number, volume and energy (NVE) ensemble at 300 K for 1 ns, followed by another 1 ns to adjust the temperature from 300 K to 320 K and 280 K in the hot and cold reservoirs, respectively. The system is then evolved for 26–60 ns depending on the system size. Larger systems need longer simulation time to reach steady state. Periodic boundary conditions are imposed along both the x and y directions. The system size L_z in z direction is varied in order to use the linear extrapolation method [25,46,47] to extract the thermal conductivity along the z direction. Newton's equations of motion are solved through the velocity-Verlet integral algorithm with a time step of 1 fs. The optimized Tersoff potential [48] is chosen to describe the intralayer C-C interactions, and Lennard-Jones potential [49] for interlayer C-C interactions. After the system reaches steady state, the heat flux J and temperature gradient ∇T can be obtained. Using the Fourier's law:

$$k_z = -\frac{J_z}{\nabla T_z} \quad (2)$$

The thermal conductivity of graphite can be calculated from formula (2) (see supporting materials). Here, the subscript z means the value is along the z direction.

In the transient simulation, we only deal with the case $\phi = 0^0$, where the x-y plane stands for the basal plane of the graphite. A small three-dimensional domain with 2 nm × 2 nm in the x-y plane and 2 layer graphene in z-axis direction located in the center of the simulation system is initialized with temperature at 300 K while the temperature in other part is set at 0 K. The three-dimensional domain is located in the center causing the heat of largest velocity to approach all the boundaries at nearly the same time, which can help us better observe and understand the transient heat transfer. Then the system is evolved under the NVE ensemble without temperature control. Snapshots of the temperature distribution at each step during the evolution are stored in order to analyze the transient heat transfer.

3. Results

3.1. Transient state simulation

Fig. 2(a) and (b) show the temperature cloud images in transient state at $t = 0.45$ ps in the x-y (basal plane) and x-z (cross-plane) planes, respectively. Here the logarithm of the temperature is plotted. In the short time scale, phonons with the largest velocity transport the longest distance without scattering, resulting in the wavefront. Phonons along different directions have different velocities, thus the wavefronts, i.e. outermost isothermal surface of temperature distribution, reflect the velocity surface. Fig. 2(a) presents the transient temperature distribution in the graphite basal plane. The circular temperature distribution in the x-y plane is reasonable because the acoustic phonon velocity is approximately equal in the Γ-M and Γ-K directions based on the graphite dispersion relation [38]. It is interesting to observe the transient temperature cloud for the x-z plane in Fig. 2(b), which has an ellip-