Hydrodynamic Phonon Transport in Graphitic Materials

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Mechanical Engineering and Materials Science / Physics and Astronomy (second appointment)

Project Description

Because of their exceptionally high thermal conductivity coupled with unusual mechanical and electrical properties, graphitic materials including graphite, graphene, and carbon nanotubes (CNTs) have received considerable interest for applications as thermal management and electronic materials [1-4]. In these graphitic materials, phonons (i.e., quanta of lattice vibrations) are the dominant heat carriers. The phonon transport in graphitic materials have been discussed in between the *diffusive* (i.e., Fourier's heat conduction law) and *ballistic* limits (i.e., no internal scattering case). A number of theoretical and experimental efforts have been carried out to investigate the influence of ballistic phonon transport on the thermal transport properties of graphitic materials [5-7]. However, two recent theoretical studies have suggested that another regime of phonon transport, the *hydrodynamic* regime, can be significant in graphene [8,9]. In fact, this hydrodynamic regime was experimentally confirmed more than 50 years ago, but has not drawn much attention for practical applications as it was observed only at extremely low temperature (~ 20 K) [10-12]. However, the two recent papers suggested that the hydrodynamic phonon transport can occur in graphene even at room temperature. The hydrodynamic phonon transport, if confirmed to be significant at room temperature, will provide new opportunities in engineering heat flow in graphitic materials with its features that are not possible within the diffusive and ballistic regimes.

The hydrodynamic regime has been relatively unexplored compared to the diffusive and ballistic regimes, and the details of this new regime still remain unresolved. Past studies on hydrodynamic phonon transport had to rely on empirical formulations due to the lack of available theoretical and numerical methods, and could not reveal detailed characteristics of this new regime of phonon transport [13-15]. The objective of this proposed research is to elucidate the influence of hydrodynamic phonon transport on the thermal transport properties of graphitic materials using first principles based calculations. Achieving this objective will address the following three key questions:

1) What are the fundamental mechanisms of energy and momentum transfer in this new regime of phonon transport? Are the common hydrodynamic phenomena in fluid flow, such as the Knudsen minimum and Poiseiulle flow, possible in the hydrodynamic phonon transport?
2) What are the characteristics of thermal transport in the crossover of three different regimes (i.e., hydrodynamic, diffusive, and ballistic)?
3) In which experimental conditions can we clearly observe and confirm the hydrodynamic phonon transport?

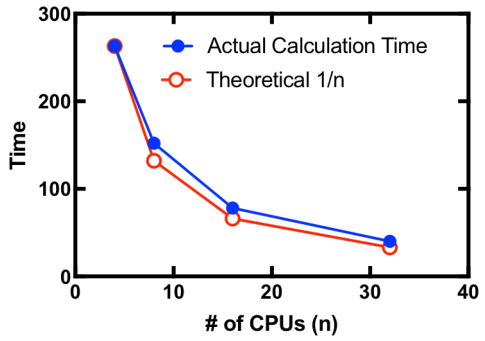
Addressing the above questions require solving the Peierls-Boltzmann transport equation (PBE), the most fundamental equation of phonon transport in the small size scale (down to few nanometers) where the particle description of lattice vibration wave is still valid. Solving the PBE accurately was known to be extremely difficult because i) it is an integro-differential equation in many dimensions, and ii) the input parameters regarding phonon properties, such as phonon dispersion and scattering matrix, could be only calculated using empirical relations. We will overcome these challenges by i) introducing stochastic approach (Monte Carlo method) and ii) calculating phonon properties based on first principles. Regarding the Monte Carlo method, we will particularly use *deviational* Monte Carlo method, where we sample only deviation of phonon

distribution from the equilibrium case instead of the distribution function itself. The deviation is usually much smaller than the distribution, and thus the numerical results from the deviational Monte Carlo simulation can have much reduced statistical uncertainty. Regarding the calculation of phonon properties, we have successfully calculated phonon properties of various materials from first principles and demonstrated its high accuracy and predictability [8,16,17].

If successful, this research will provide better understanding of phonon transport in graphitic materials with a new perspective from the hydrodynamic pictures. Also, this theoretical study will quantitatively guide further experimental efforts to confirm the hydrodynamic regime by providing the best experimental conditions where the hydrodynamic regime is most significant. Moreover, the Monte Carlo solver of the PBE that will be developed in this study will combine the Monte Carlo methods and phonon calculation from first principles, which can be useful for studying phonon transport in various materials not limited to graphitic materials.

Required Computational Resources and Its Justification

We request 1,500,000 service units for the MPI cluster. The simulation we will carry out consists of two steps. First, we calculate phonon properties (phonon dispersion and scattering matrix) using the Quantum Espresso package. The Quantum Espresso package has been developed and maintained by many research groups and already shows a good scalability in MPI environment. This calculation will require 300,000 service units, which is larger than typical DFT calculation of graphitic materials. This is because the scattering matrix calculation requires high order force constants (higher than second order) whose calculation is computationally costly. Second, we will develop and run the Monte Carlo code for solving the PBE to study the hydrodynamic phonon transport in graphitic materials. We already developed the initial version of this Monte Carlo code using Fortran and MPI and confirmed this code runs successfully in the Frank system. The figure on the left proves that our in-house code shows a good scalability in the Frank system. We expect that this calculation will require 1,200,000 service units. Our deviational Monte Carlo method is significantly better than standard Monte Carlo method in terms of computational efficiency, but still we need to sample phonon distribution in total 8 physical dimensions (three for real space, three for wavevector space, one for phonon polarization, and one for time domain), making our calculation still computationally expensive.



The scalability of our in-house Monte Carlo code for parallel computing in MPI environment (Frank)

External Funding & Previous Publication

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References

1. [redacted]