Computer Physics Communications 195 (2015) 112–116

Contents lists available at ScienceDirect

Computer Physics Communications

journal homepage: www.elsevier.com/locate/cpc

A coherent phonon pulse model for transient phonon thermal transport

Xiang Chen [a,*], Aleksandr Chernatynskiy [b], Liming Xiong [c], Youping Chen [a]

[a] Department of Mechanical and Aerospace Engineering, University of Florida, Gainesville, FL 32611, United States
[b] Department of Materials Science and Engineering, University of Florida, Gainesville, FL 32611, United States
[c] Department of Aerospace Engineering, Iowa State University, Ames, IA 50011, United States

ARTICLE INFO

Article history:
Received 23 January 2015
Received in revised form
11 April 2015
Accepted 13 May 2015
Available online 21 May 2015

Keywords:
Computational model
Coherent phonons
Ultrashort laser pulse
Transient phonon transport

ABSTRACT

In this work, we present a novel heat source model, the *coherent phonon pulse* (CPP), composed of spatiotemporal Gaussian wave packets to mimic the coherent excitation of a non-equilibrium phonon population by ultrashort laser techniques, for the study of transient phonon thermal transport. Through molecular dynamic simulations of phonon transport in bicrystalline silicon-nanowires containing $\Sigma 3$ and $\Sigma 19$ grain-boundaries (GBs), we demonstrate that the new model facilitates not only a quantitative measurement of phonon-interface scattering, but also a mechanistic understanding of the highly non-equilibrium process of phonon transport with the coherent wave nature being preserved.

1. Introduction

The advances in time-resolved experimental studies of thermal transport, especially in the study of transient non-equilibrium heat flow using pump-probe laser pulse methods, have led to discoveries of many new phenomena, and have produced a wealth of challenges for the simulation community to interpret the experiments [1]. In particular, the pump-probe technique has become a leading transient thermal metrology not only for the measurement of thermal conductivity and interfacial thermal conductance, but also for observation of coherent phonon transport [2]. Despite the rich pool of experimental studies, however, there is currently no well-established simulation method that can mimic either the coherent phonon generation or the process of the coherent phonon pulse propagation and scattering. A key component that is missing is a heat pulse model that can mimic the phonon population excited by laser pulses.

A laser pulse generates a local concentration of non-equilibrium phonons. According to Msall and Wolfe [3], the generated non-equilibrium phonons highly depend on the "excitation density" (or absorbed laser power per area) and the properties of the sample material. Although the process of phonon generation and the resulting complex phonon oscillation are still a topic of study [4], using ultrashort laser techniques, several research groups have demonstrated that the propagation of the light pulses in solids is accompanied by lattice vibrations showing a high degree of spatial and temporal coherence [4–7]. Coherent phonon generations are observed from the Γ- point phonons to the near-Brillouin zone boundary phonons, including acoustic and optical phonons [8–10]. Such excitation and detection of coherent lattice vibrations has been observed in many transparent and opaque materials, such as transition metals [11], semi-metals [12–17], cuprates [18], insulators [19] and semiconductors [20–22]. Several mechanisms were proposed to explain the coherent phonon generation [23,24]. A widely accepted view is that, when the duration of laser pulse is short enough, the phonons are excited coherently. Coherence is a general phenomenon that takes place whenever the ultrashort laser interacts with solid [25].

The atomic-level molecular dynamics (MD) simulation is a powerful tool that can be used to simulate transient non-equilibrium process without the need of a priori information of phonon properties. The most widely used non-equilibrium MD simulation method for the calculation of thermal conductivity or thermal conductance is the direct method, i.e., the heat source–sink method, which is in analogy to the experimental steady-state measurement [26–28]. To determine the thermal conductance (σ_k) of an interface, this method uses a thermal model, $\sigma_k = J/\Delta T$, where J is the steady state heat current and ΔT is the temperature discontinuity at the interface. However, this method

* Corresponding author. Tel.: +1 352 392 8494; fax: +1 352 392 7303.
E-mail address: xiangchen@ufl.edu (X. Chen).

http://dx.doi.org/10.1016/j.cpc.2015.05.008
0010-4655/

cannot provide the fine details that reveal the underlying mechanisms about phonon-interface scattering, which is critically important in the study of nanoscale thermal transport.

In order to gain information about phonon scattering mechanism, the phonon wave-packet dynamics method (PWD) [29,30] has been developed [31], and shown to be instructive in the mechanistic study of an individual phonon with a specific polarization and wavevector. In addition, by calculating the energy transmission coefficient of each phonon, the theoretical thermal conductance can be obtained [27,32]. However, to predict the thermal boundary conductance, many simulations have to be performed to determine the energy-transmission coefficient for each polarization and each wavevector. The contribution of each phonon is then superimposed according to a theoretical model based on the thermodynamic equilibrium Bose distribution [27]. Currently this method is essentially a zero-temperature method, the anharmonicity or phonon–phonon interaction that may accompany the ballistic phonon transport processes is basically ignored [29].

The direct method is very useful for quantitative study of steady-state heat flow, while the wave packet method is the method of choice for the mechanistic study of the scattering of individual phonons by interfaces. Unfortunately, neither method is well suited for the simulation of the generation and propagation of a phonon pulse. Existing attempts to study the transient heat pulse propagation have used the thermostat algorithms to model the heat source [33–35]. However, thermostat algorithms mimic thermalizing events such as damping and dephasing of phonon waves. As a result, the phase information or the phonon coherency is usually completely or partially destroyed [36]. Therefore, they are also not well suited for the simulation of the coherent phonon excitation by ultrashort laser pulses.

Motivated by the need of a heat pulse model to mimic the coherent phonon excitation and inspired by the time-resolved experimental studies, in this paper we propose a heat source model called *coherent phonon pulse* (CPP) model to mimic the coherent phonon excitation in the ultrashort laser pulse experiments.

2. The CPP model

Coherence is the key property of the phonons excitations generated in solids by the ultra-short laser pulse. The spatial and temporal coherence of phonons can be mathematically represented by phonon wave packets [37]. In addition, experiments show that most of the incident pump laser pulses can be described in a Gaussian profile [23,38–40]. Also, a laser pulse generates a local concentration of nonequilibrium phonons, and the number of phonons excited can be linked to the local temperature associated with the heat pulse [41].

Based on the results of the experimental studies as well as considerations mentioned above, in this work, we construct the CPP model from spatiotemporal Gaussian phonon wave packets. The general mathematical description of such wave packets can be given as follows:

$$u_{i\mu\lambda}(r,t) = \sum_{j=1}^{n} A_j \varepsilon_{ji\mu\lambda} \exp[i(k_j(r-r_0) - \omega_j(t-t_0))] \times \exp\left[-\frac{(t-t_0)^2}{\xi_j^2}\right] \exp\left[-\frac{(r-r_0)^2}{\eta_j^2}\right] \quad (1)$$

where $u_{i\mu\lambda}(r,t)$ is the μth component of the displacement of atom i polarized along $\varepsilon_{ji\mu\lambda}$ in the designated unit cell located at position r; the phonon branch is labeled by λ; r_0 and t_0 are the center of the wave packet in spatial and temporal domain, respectively. The summation over j accounts for the combination of n Gaussian waves, which form a specific phonon distribution; k_j, ω_j, η_j and ξ_j are central wavevectors, frequencies and variances of the Gaussian wave packets in spatial and temporal domain. The magnitude A_j of each wave packet can be determined through the temperature and the phonon distribution associated with the heat pulse. The applied transient atomic vibration according to Eq. (1) gives rise to a coherent heat source comprised of Gaussian wave packets in both spatial and temporal domain. Note that when 1D transport is considered without time dependence or a specific distribution, the CPP model reduces to the phonon wave-packet dynamics.

The salient features of the CPP model can be summarized as follows. It mimics the ultrashort laser pulse experiments generating an ensemble of phonons, enables the wave nature of phonon thermal transport to be retained, and allows for the transient characterization in thermal conduction regime where Fourier's Law breaks down. In the experiment, the control of phonons has to be realized through a precise modulation of the light source [25], while using the CPP model with MD simulation we can directly assign the transient atomic motion. In addition, to characterize the material properties, we directly track the atomic trajectories everywhere in the specimen without the need of other assumptions. This also significantly simplifies the data analysis, which is complex in the experiment [38]. At the simplest level, the effect of the grain boundary (GB) can be characterized in terms of phonon transmittance [30]. Thus, for any ensemble of phonons of interest, we calculate the corresponding energy-transmission coefficients of the interfaces. Moreover, the intrinsic scattering, i.e., the phonon–phonon interaction at finite temperature is naturally allowed using the CPP model.

3. Simulation results

We apply this model in molecular dynamics (MD) simulation to study the non-equilibrium transient phonon thermal transport in silicon nanowires (SiNWs) with grain boundaries (GBs). We choose SiNWs because of the coexistence of coherent and incoherent phonon transport during the process of heat transfer due to the intensive surface scattering. GBs are introduced in order to demonstrate the capability of this new model in studying the thermal boundary resistance (TBR) problem.

Since SiNW provides a quasi-1D channel for energy transport, Eq. (1) can be reduced to

$$u(z_0,t) = \sum_{j=1}^{n} A_j \varepsilon_{ji\mu\lambda} \exp[i\omega_j(t-t_0)] \exp\left[-\frac{(t-t_0)^2}{\xi_j^2}\right] \quad (2)$$

where $u(z_0,t)$ is the transient displacement of the 1st atom in the designated unit cell located at z_0, at time t. All the other symbols follow the definition in Eq. (1). In addition, we do not differentiate acoustic and optical phonons since they are no longer relevant in nanostructures due to the phonon dispersion modification [42]. Hence, the displacement of the 2nd atom is not controlled, but solved in the simulation according to the equation of motion. Since the phonon density of state (PDOS) is one way to link the phonon distribution to the temperature within the framework of classical mechanics, in this work, we construct the CPP according to the PDOS of the material.

The resulted displacement of the CPP is plotted in Fig. 1(a). The corresponding frequency domain analysis by Fourier transform is presented in Fig. 1(b), from which, the heat pulse can be identified as four Gaussian waves (the central frequency of the last two Gaussian waves are close). The inset in Fig. 1(b) is the PDOS of single crystal SiNW at 500 K, which is obtained by calculating the Fourier transform of the velocity–velocity autocorrelation from the trajectories of atoms in a MD simulation [43,44]. Comparing Fig. 1(b) with the inset, we conclude that the heat pulse input we constructed is a rough representation of the PDOS.