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Fast-Converging and Asymptotic-Preserving Simulation of Frequency Domain Thermoreflectance

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Abstract. The heat conduction under fast external excitation exists in many experiments measuring the thermal conductivity in solids, which is described by the phonon Boltzmann equation, i.e., the Callaway's model with dual relaxation times. Such a kinetic system has two spatial Knudsen numbers related to the resistive and normal scatterings, and one temporal Knudsen number determined by the external oscillation frequency. Thus, it is a challenge to develop an efficient numerical method. Here we first propose the general synthetic iterative scheme (GSIS) to solve the phonon Boltzmann equation, with the fast-converging and asymptotic-preserving properties: (i) the solution can be found within dozens of iterations for a wide range of Knudsen numbers and frequencies, and (ii) the solution is accurate when the spatial cell size in the bulk region is much larger than the phonon mean free path. Then, we investigate how the heating frequency affects the heat conduction in different transport regimes.

AMS subject classifications: 80M20

Key words: Frequency domain thermoreflectance, phonon Boltzmann equation, general synthetic iterative scheme.

1 Introduction

The heat conduction is critically important to the functionality and reliability of materials and devices, which is usually described by the Fourier conduction law, i.e., the heat flux ($\boldsymbol{q}$) is proportional to the product of negative temperature gradient ($-\nabla T$) and thermal conductivity (κ). However, numerous non-Fourier phenomena [1,2] have been observed

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in either nanostructured materials (e.g., heat dissipation on chip cooling [3], thermoelectric energy conversion [4], and nanomedicine [5]) or ultrafast process [6,7], which pose great challenges in investigating thermal properties of solid materials, especially when the ultrafast laser-based thermoreflectance techniques, including the time-domain thermoreflectance (TDTR) and the frequency-domain thermoreflectance (FDTR) techniques, are adopted.

Ultrafast thermoreflectance techniques often employ subpicosecond lasers [9]. Repeated laser pulses are divided into two beams, where the pump beam thermally excites a sample surface and the probe beam measures the time-resolved reflectivity or diffraction. In TDTR method, probe beam arrives at the sample surface at different time intervals after the pump beam through a mechanical delay stage. Various thermal properties, such as the cross-plane thermal conductivity, in-plane thermal conductivity, interfacial thermal conductance, and heat capacity, can be evaluated by fitting the temporal decay of measured signals to a thermal transport model, with unknown thermal properties as the fitting parameters. The TDTR technique has been applied to measure the thermal properties of thin films [10], multilayers [11], bulk materials [12], and their interfaces [13]. However, imperfections of the mechanical moving stage can introduce measurement errors, and ultrafast pulsed lasers are expensive. FDTR [7] is a variation of TDTR, where the thermoreflectance signals are measured by varying the modulation frequency of the pump beam instead of the delay time between the pump beam and the probe beams. FDTR can measure the same thermal properties as TDTR without a moving stage and ultrafast pulsed laser, thus eliminating the disadvantages of TDTR. There are two possible experimental setups for FDTR: one is based on pulsed lasers, almost the same setup as TDTR, with the modulation frequency ranging from 0.1 MHz to 80 MHz. In continuous-wave laser FDTR, the frequency of the pump beam is modulated by the electro-optical modulator and generates heat flux on the sample surface. Because it is not necessary to use an ultrafast pulsed laser, the continuous-wave laser FDTR can be configured at a low cost. Theoretically, in contrast to pulsed TDTR/FDTR, a pump beam of continuous-wave laser FDTR can be modulated at an infinite frequency. However, in practice, the modulation frequency is limited to less than 80 MHz due to the decreasing signal intensity and the presence of noise at high frequencies. To investigate the size effect of the thermal conductivity in nanoscale materials, wide-range modulation of the heating frequency is needed. Broadband FDTR has been implemented to overcome this frequency limitation and extend it to 200 MHz using heterodyne detection [8,14], seeing the schematics in Fig. 1.

Note that the diffusion model based on the Fourier law is often used to extract the thermal conductivity from the experimental data. However, as the problem involves multiscale spatial-temporal heat transfer, the macroscopic conduction model cannot capture the dynamic thermal properties; meanwhile, mainstream methods, such as the molecular dynamics and non-equilibrium Green functions are computationally expensive. Thus, the mesoscopic method based on the phonon Boltzmann equation (PBE), which bridges the microscopic and macroscopic dynamics of heat carriers (phonon), shows great advan-