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Research Paper

Ballistic thermal wave propagation along nanowires modeled using phonon Monte Carlo simulations

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HIGHLIGHTS

- Ballistic thermal wave propagation along nanowires is investigated using a phonon-traced Monte Carlo method.
- The effects of boundary scattering on thermal wave propagation differ for ballistic-diffusive and diffusive phonon transport.
- Different phonon transport regimes will be measured using different temporal resolution, and their dependencies on phonon scattering regimes are different.

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ABSTRACT

The propagation of ballistic thermal waves when the phonon transport is in the ballistic-diffusive regime is markedly affected by the boundary. This work simulates ballistic thermal wave propagation in nanowires with a phonon-traced Monte Carlo method to investigate the effects of the nanowire characteristics including the radial Knudsen number and the specularity parameter, and the effects of the temporal resolution of the measurements. The phonon boundary scattering accelerates the evolution of the phonon transport from ballistic to ballistic-diffusive and finally to diffusive transport and increases the thermal conduction resistance by reducing the effective thermal conductivity. High heat pulse frequencies lead to thermal wave propagation in ballistic regime, moderate heat pulse frequencies lead to thermal wave propagation in ballistic-diffusive regime and very low heat pulse frequencies lead to purely diffusive thermal wave propagation, i.e. the Fourier thermal conduction. The ballistic-diffusive thermal wave propagation relies heavily on the specific type of the dominated phonon scattering mechanism while the purely ballistic and diffusive propagations do not. Thus, ballistic-diffusive thermal wave propagation should be modeled by a new constitutive equation with new characteristic parameters.

1. Introduction

Rapid developments of micro-electromechanical systems (MEMS) and nano-electromechanical systems (NEMS) [1–4] are placing higher demands on micro/nano devices with the need for excellent physical properties. Semiconductor nanowires and nanowire arrays are being developed as micro/nano devices due to their novel physical optical [5,6], electrical [7,8] and thermoelectric properties [9–11]. The ultra-short pulse laser technique has become an effective tool in micro device machining due to its high precision [12–17]. The interactions between the ultra-short laser pulse and the materials involve a complex set of optical, electrical and thermal processes [12–17] such as photon-electron interactions, electron-electron interactions and transport, electron-phonon interactions, and phonon-phonon interactions and transport. Since the interactions are very fast, the carriers do not relax diffusively but have non-diffusive characteristics. Another area needed in the development of micro/nano devices is thermophysical properties measurements [18]. The small scales require ultra-high temporal and spatial resolution. Ultra-short pulse lasers offer the necessary resolution for the nanomaterial measurements in a non-contact method called pump-probe optical measurements [19–22] which can be used to measure the properties of thin films [19–23] and nanowire arrays [24]. However, for semiconductor nanostructures like nanowires, the special nature of these nanoscale devices requires more detailed information about the nanowires such as the mean free path (or relaxation time) distributions which require ultrafast carrier dynamics measurements [8,25,26] in addition to their macroscopic thermal physical

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properties to tune and optimize their optical, electrical and thermal properties.

In both ultra-short pulse laser machining and laser measurements, theoretical research on ultrafast heat conduction is needed to guide process analyses. In semiconductor materials, the phonons dominate the thermal conduction as the main carrier [27]. When the characteristic thermal time is on the order of picoseconds, comparable with the phonon relaxation time, the classical Fourier's law fails to predict the phenomena because the heat is transported in the phonon ballistic-diffusive regime [28–33] as thermal waves [34,35]. As a result, more precise modeling and analyses for measurements should start from a more fundamental basis (e.g. the phonon Boltzmann transport equation) instead of the macroscopic models (e.g. Fourier's law). Classical phonon transport theory has two kinds of thermal waves including ballistic thermal waves from the phonon ballistic transport and the second sound from phonon normal scattering dominated transport [34,35]. This research focuses on the ballistic thermal waves occurring in phonon ballistic-diffusive transport. Previous theoretical and numerical analyses have mostly focused on one-dimensional models [36,37] with few studies of three-dimensional effects that include lateral boundaries. However, research on thermal wave propagation in three-dimensional nanostructures is essential as there are more and more measurements of complicated nanostructures such as nanoporous silicon [23] and nanowire arrays [24]. In one-dimensional cases, the thermal wave is mostly affected by the phonon boundary emissions and is dissipated by the intrinsic phonon scattering [34,35], with more phonon boundary scattering involved in three-dimensional cases. Typical boundaries and interfaces include the lateral boundaries in nanowires [38–41], emission and reflection boundaries in nanofilms [34,35] and the interfaces in superlattice nanofilms [42] and polycrystals [43,44]. Unlike steady state heat conduction [38–44], ultrafast heat conduction is a non-equilibrium process with both spatial and temporal variations, making it more complicated and more easily influenced by the properties and spatial distribution of the boundaries in addition to the internal properties of the materials [33].

Although many studies have carried out to develop the macroscopic constitutive heat conduction equation using the phonon Boltzmann transport equation [29,37], especially for cross-plane ultrafast heat conduction, modeling on the boundaries both in microscopic and macroscopic levels which dominate for large Knudsen number conditions are few. Analyses of thermal transport problems that are heavily affected by the boundaries need a comprehensive understanding of the effect of the boundaries to develop a practical constitutive heat conduction equation and make reasonable extensions. Finally, since the effective thermal physical properties of nanomaterials are closely related to the measurement characteristics including the measurement parameters [45] and methods [46], development of a new constitutive equation must carefully considers experimental and engineering practices. Consequently, the characteristic parameters in thermal applications and measurements should be taken into consideration in establishing a new macroscopic constitutive equation with new characteristic parameters to describe the thermal processes and guide the data analyses in experiments.

This work investigates ballistic thermal wave propagation in nanowires using a phonon-traced Monte Carlo (MC) method. Cases with different radial Knudsen numbers, Kn_r and specularity parameters, p, are simulated to study the effects of the general characteristics of the nanowires in both phonon ballistic-diffusive transport and diffusive transport. Heat pulses with various periods are simulated to investigate the effects of the temporal resolution of the measurements. The results give a comprehensive understanding of ballistic thermal wave propagation in nanowires to guide further experimental analyses.

2. Phonon Monte Carlo methods

A phonon-traced Monte Carlo simulation method is used to model the ballistic-diffusive phonon transport [32,47] to simulate the ballistic thermal wave propagation in nanowires by directly solving the phonon Boltzmann transport equation in the nanowires,

$$\frac{\partial f}{\partial t} + \mathrm{v}_g \cdot \nabla f = -\frac{(f - f_0)}{\tau_R}, \tag{1}$$

in which f and f_0 are the phonon distribution function and the phonon equilibrium distribution function, τ_R is the relaxation time for phonon resistive scattering, t is the time and v_g is the phonon group velocity. The heat pulse trace and the nanowire model with a circular cross section are shown in Fig. 1. The heat pulse is input from the left end ($x = 0$) of the nanowire and propagates in the x direction, i.e. the axial direction. The radial direction is described by the radius r and the circumferential direction by the angle φ. The initial temperature of the nanowire is set to be 300 K, room temperature. Phonons are emitted according to the Lambert cosine law at the emission boundary. As Eq. (1) shows, phonon intrinsic scattering is treated with the relaxation time approximation. Phonons interact with the lateral boundary and are scattered back into the nanowire. The phonon boundary scattering is modeled using a specularity parameter, p, according to the formula proposed by Ziman [27],

$$p = \exp(-16\pi^3\delta^2/\lambda^2), \tag{2}$$

in which δ is the root mean square surface roughness and λ is the phonon wavelength. Generally, when the lateral boundaries of the nanowires and nanofilms are flat and the surface roughness is smaller than the phonon wavelength, this classical formula, i.e. Eq. (2), is applicable. In this work, we study the phonon boundary scattering effect using the classical framework in which the boundary roughness is described by the specularity parameter, p. The boundary condition for the lateral boundary in the nanowire is [27]

$$g(\vec{k}, r_B)_{v_n} = pg(\vec{k}', r_B)_{-v_n}, \tag{3}$$

in which g is the deviation distribution function $(f - f_0)$, $\vec{k}'$ and $\vec{k}$ are the wave vectors for the incident and reflected phonons at the boundary, and r_B is the phonon boundary scattering location. Eq. (3) means that $p \times 100\%$ of the phonons colliding with the boundary are scattered specularly with the rest scattered diffusively. The specular scattering conserves the phonon momentum and the non-equilibrium part of the distribution function while the diffuse scattering does not. For a nanowire, the radius of the circular cross section is R and the thickness of the nanowire is described by the radial Knudsen number, Kn_r, defined as $Kn_r = l/R$ where l (56 nm) is the mean free path of the phonon intrinsic scattering [29]. Here, the phonon group velocity, v_g, is 5000 m/s [29]. The heat pulse period, t_p, is 0.2, 2, or 20 ps for the investigations. The gray model is used for the phonon frequency spectrum and the Debye approximation is used for the dispersion [30].

3. Results and discussion

3.1. General discussion

There are several key factors that affect the propagation of the ballistic thermal wave in nanostructures including the phonon emissions at the boundary, the characteristics of the heat pulse (especially the period, t_p), the time, t, of the thermal wave propagation, the relaxation time, τ, for the phonon intrinsic scattering (including τ_N for normal scattering, τ_R for resistive scattering and τ_i for isotopic scattering), the specularity parameter, p, the radial

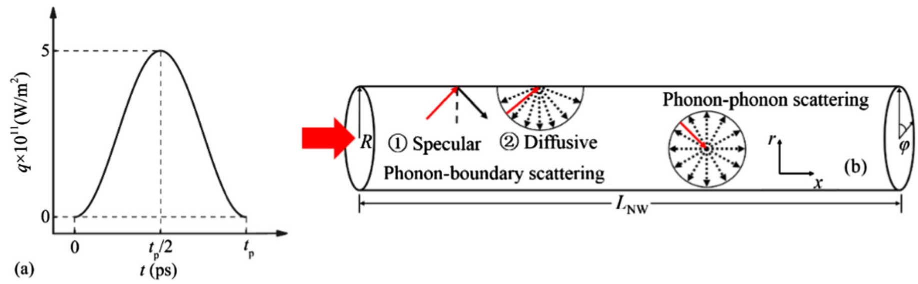

Fig. 1. Schemes for the (a) heat pulse and (b) simulation systems.

Knudsen number, Kn_r (for the lateral boundary in nanowires or nanofilms), and the geometrical constraint equation for the boundary, r_B (especially the boundary shape and the direction of the heat flux vector relative to the boundary).The specularity parameter and the radial Knudsen number determine the relaxation time, τ_B, for the phonon boundary scattering in nanowires which is always diffuse since the specular boundary conserves the non-equilibrium part of the phonon distribution function which makes τ_B infinite. The propagation time, t, commonly has the same order of magnitude as the heat pulse period, t_p. A sine function heat pulse shape is generally used since the period has the greater influence. Previous studies have investigated the effect of the phonon emission at the boundary [34,35]. There are several kinds of phonon intrinsic scattering regimes in a solid, but there is always a dominate one [29]. The phonon resistive scattering dominates at room temperature while normal scattering can be ignored in semiconductors such as silicon [48]. The relaxation time, τ_R, for phonon resistive scattering derived with gray approximation is then a constant. For nanowires, the boundary is assumed to be flat with the phonon boundary scattering then analyzed using the classical framework [27]. A circular cross section is chosen as a general case. The direction relation is determined by the heat flux being parallel to the main boundary, i.e. the lateral boundary. The effects of the boundary roughness and the boundary shape are separated here even though the boundary surfaces are irregular [49] and there is no explicit distinction between the roughness and the shape. Saha et al. [50] and Moore et al. [51] explained the ultralow measured thermal conductivities in nanowires by proposing that the phonon backscattering regime is similar to gas molecules backscattering in rarefied gas transport and that the rough nanowire surface has a sawtooth boundary shape [52,53]. Recently, He et al. [54] proved the need to consider the surface ripples to explain the reduction of the effective thermal conductivity. Phenomenologically, various boundary conditions have been studied for smooth [55] and rough lateral [56,57] boundaries using the phonon hydrodynamics framework in which the diffuse phonon boundary scattering and the phonon backscattering are modeled separately. We prefer to treat the backscattering effect as the effect of the shape of the boundary and treat the lateral boundaries in a general way using the classical framework.

Thus, under the classical framework, the most important factors are the specularity parameter and the radial Knudsen number, the heat pulse period, and the thermal wave propagation time. The effects of these four factors can be divided into the effect of the nanowire characteristics (the first two factors) and effect of the temporal resolution of the measurements (the last two factors).

3.2. Effect of the nanowire characteristics: The passive effect

3.2.1. Comparisons with nanofilms: A validation

In principle, the phonon boundary effects should be taken into consideration only when the characteristic size of the nanostructure is comparable with the characteristic length of the heat carrier motion, i.e. the phonon motion in this work. The specular phonon boundary scattering is normally assumed to not reduce the phonon mean free path and cause a size effect. Consequently, the heat pulse propagation in nanowires with small radial Knudsen numbers or specular boundaries should be the same as those in nanofilms in both the phonon diffusive and ballistic-diffusive regimes. Comparisons of the simulation results in Fig. 2a and b show that this conclusion is correct, which also validates our simulations for nanowires. As a result, the following results for nanowires with specular boundaries and small radial Knudsen numbers can also be applied approximately to nanofilms.

3.2.2. Diffusive regime

Since the characteristics of the circular nanowires, including the specularity parameter, p, of the lateral boundary and the radial Knudsen number, Kn_r, are determined once the sample is prepared and placed in a specific measurement experiment, this effect is referred to as a passive effect. The classical size effect caused by the phonon boundary scatterings reduces the effective thermal conductivity of the nanowires with increasing radial Knudsen number Kn_r and decreasing specularity parameters for steady state heat conduction [48]. This section describes long-time simulations (200 ps) of the propagation of heat pulses in nanowires for different radial Knudsen numbers, and specularity parameters to study how the size affects transient heat conduction in the diffusive regime.

As seen in Fig. 3a for Kn_r equal to 0.2, the differences between the temperature profiles for different specularity parameters can be neglected since the phonon boundary scattering events are too few to significantly influence the thermal wave propagation. In this case, the resistive phonon scattering dominates the phonon transport and the heat pulse propagates in the diffusive regime obeying the predictions of the classical Fourier's law without modifications. When Kn_r increases to 1.0, the temperature profiles for the various specularity parameters differ as seen in Fig. 3b with the temperature peaks reduced for specularity parameters smaller than 1.0. When the radial Knudsen number increases to 4.0, the temperature profiles are quite different even though they are all still in the diffusive regime. The temperature profile in a nanowire with a smaller specularity parameter develops more slowly than with a larger specularity parameter. The temperature profiles for the larger radial Knudsen numbers and the same specularity parameter develop more slowly than with the smaller radial Knudsen number for $p < 1.0$. These results for transient conduction are similar to results for steady state thermal conduction [58] in that nanowires with larger radial Knudsen numbers and smaller specularity parameters have smaller effective thermal conductivities, indicating that the phonon boundary scattering acts as a thermal resistance for transient thermal conduction in nanowires.

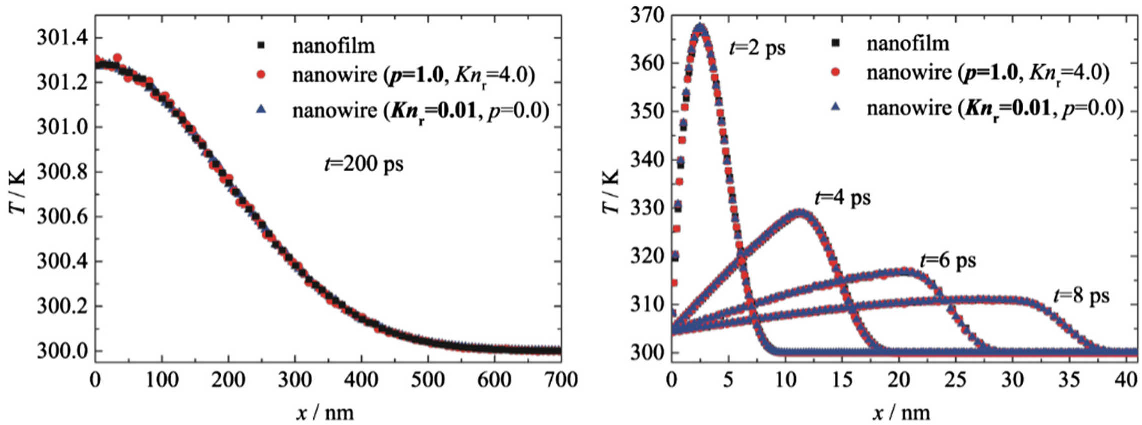

Fig. 2. Temperature profiles in nanofilms and nanowires in the diffusive and ballistic-diffusive regimes: comparison and validation.

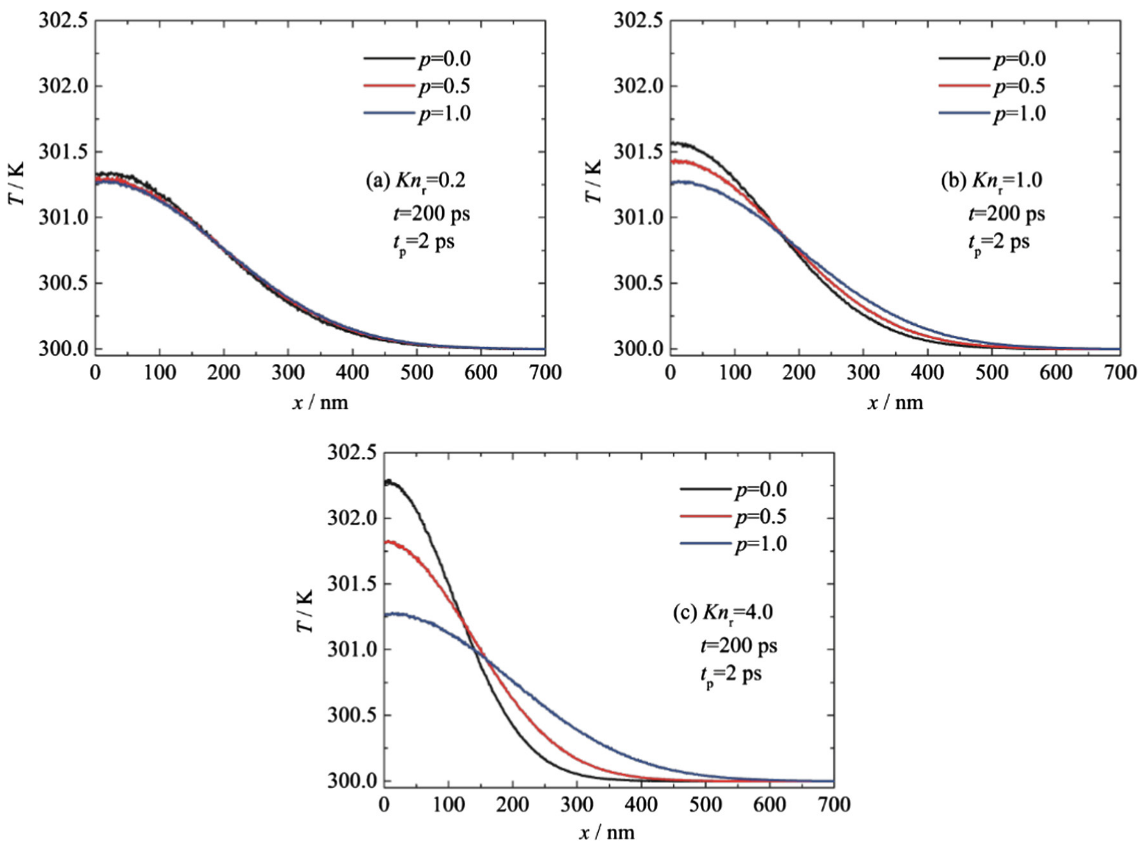

Fig. 3. Temperature profiles in nanowires (t_p = 2 ps, t = 200 ps, p = 0, 0.5, 1) (a) Kn_r = 0.2, (b) Kn_r = 1.0, (c) Kn_r = 4.0.

3.2.3. Ballistic-diffusive regime

Although phonon boundary scattering causes a size and roughness dependent effective thermal conductivity in nanowires, the classical Fourier's law still works for transient heat conduction in the diffusive regime with the modified effective thermal conductivity. However, the thermal processes are quite different when the phonon transport is in the ballistic-diffusive regime. This section describes a series of cases for different Kn_r and p to give a comprehensive description of thermal wave propagation in the ballistic-diffusive regime in nanowires. As shown in Fig. 4a, b and c for Kn_r equal to 0.2, the temperature profiles for $p = 0$, 0.5 and 1 are nearly the same, indicating little phonon boundary scattering so the different specularity parameters have no effect. With increasing Kn_r, the temperature profiles for the different specularity parameters differ as seen in Fig. 4b and c for Kn_r of 1.0 and 4.0. In Fig. 4c, for $p = 1.0$, the thermal wave propagates with a strong peak; for $p = 0.5$, the peak decreases and the temperature near the emission boundary increases while for $p = 0$, i.e. a diffuse boundary, the peak disappears and the temperature distributions become more uniform with the maximum temperature located at the emission boundary, which is very close to the purely diffusive result. The results for the three different Kn_r, also show that the temperature profiles in the nanowires with $p = 1.0$ do not change with Kn_r, indicating that the specular boundary does not affect the ballistic thermal wave propagation, which is consistent with theoretical analyses for steady state cases. Thus,

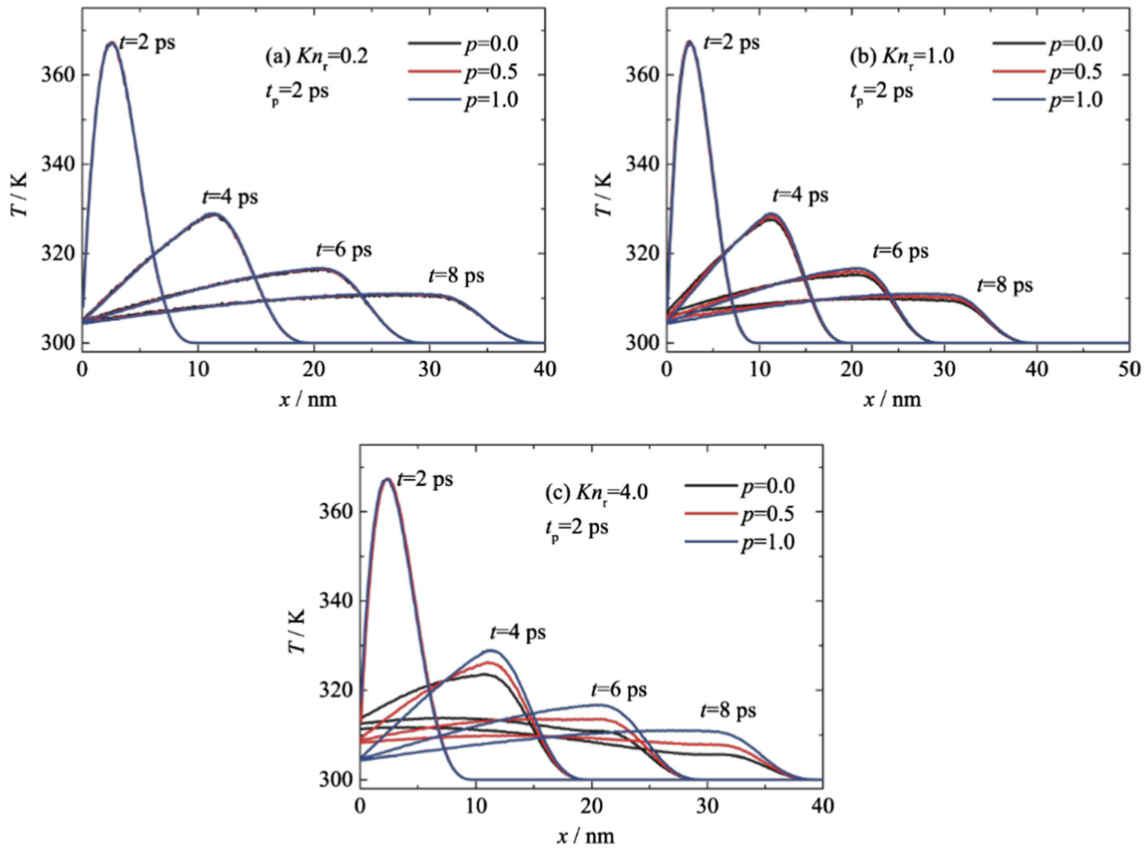

Fig. 4. Temperature profiles in nanowires (p = 0, 0.5, 1) with (a) Kn_r = 0.2, (b) Kn_r = 1.0 and (c) Kn_r = 4.0.

ballistic thermal wave propagation in nanowires is weakened and the phonon transport changes faster from the ballistic regime to the ballistic-diffusive regime by the extra phonon scattering by the boundaries, particularly for large radial Knudsen numbers and small specularity parameters.

3.3. Effect of the heat pulse period: The active effect

A specific nanowire at room temperature has a fixed radial Knudsen number and specularity parameter. Thus, the characteristic phonon relaxation times are constant. Then, the temporal resolution, i.e. the heat pulse period, has the dominate effect on the thermal wave propagation and measurements [59,60]. Consequently, this effect is called the active effect since the heat pulse period can be set arbitrarily in experiments. This section describes investigations of the active effects in nanowires for both specular and diffusive boundaries.

Fig. 5a, b and c shows the temperature profiles for ballistic thermal wave propagation with different heat pulse periods for p = 1.0 where only phonon intrinsic scattering dominates. When heat pulse period is relatively fast, i.e. the heat pulse period, t_p, (0.2 ps) is much shorter than τ_R, the thermal wave propagates in the ballistic regime, as evidenced by the small temperature rise near the emission boundary and the strong peaks. When t_p (20 ps) is larger than τ_R as in Fig. 5c, the peak gradually disappears and the temperature distribution loses its wave characteristics and approaches the prediction of Fourier's law. In the intermediate case where t_p is 2 ps (comparable with the relaxation time, τ_R), the thermal wave propagates in the ballistic-diffusive regime with both wave characteristics and diffusive dissipation characteristics.

Next, effect of the heat pulse period on the ballistic thermal wave propagation in nanowires with p = 0.0 is seen in the results shown in Fig. 6a, b and c. Unlike with p = 1.0, there are two characteristic phonon relaxation times including the relaxation time for phonon boundary scattering, τ_B, and the relaxation time for phonon intrinsic scattering, τ_R. For fast heat pulse periods where t_p (0.2 ps) is much shorter than these two characteristic relaxation times, the thermal wave propagation is nearly the same as in nanowires with p = 1.0 in the nearly complete ballistic regime. In Fig. 6b, the heat pulse period is comparable with both τ_R and τ_B, with the temperature distributions then determined by both the phonon intrinsic scattering and the phonon boundary scattering. Even though the phonon transport shown in Figs. 5b and 6b are both in the ballistic-diffusive regime, they differ due to the different dominated phonon scattering regimes. The phonon transport in Fig. 5b can be called intrinsic scattering dominated ballistic-diffusive transport, while that in Fig. 6b can be called intrinsic and boundary scattering dominated ballistic-diffusive transport.

In measurements, when the heat pulse period is relatively long, although the size affects the effective thermal conductivity, the temperature profiles can still be predicted by Fourier's law with a modified effective thermal conductivity. Generally, this modified effective thermal conductivity is always smaller than the intrinsic thermal conductivity since the phonon boundary scattering reduces the phonon mean free path. However, when the heat pulse period is very short, the ballistic thermal wave propagation is independent of the scattering regime and is quite different from that predicted by Fourier's law. The temperature distributions then need to be predicted using the one-dimensional phonon Boltzmann transport equation or the phonon radiation transport equation since an effective thermal conductivity cannot be defined for these conditions for the classical framework. When the heat pulse period is moderately long, temperature profile predictions are difficult since they rely strongly on the dominant phonon scattering

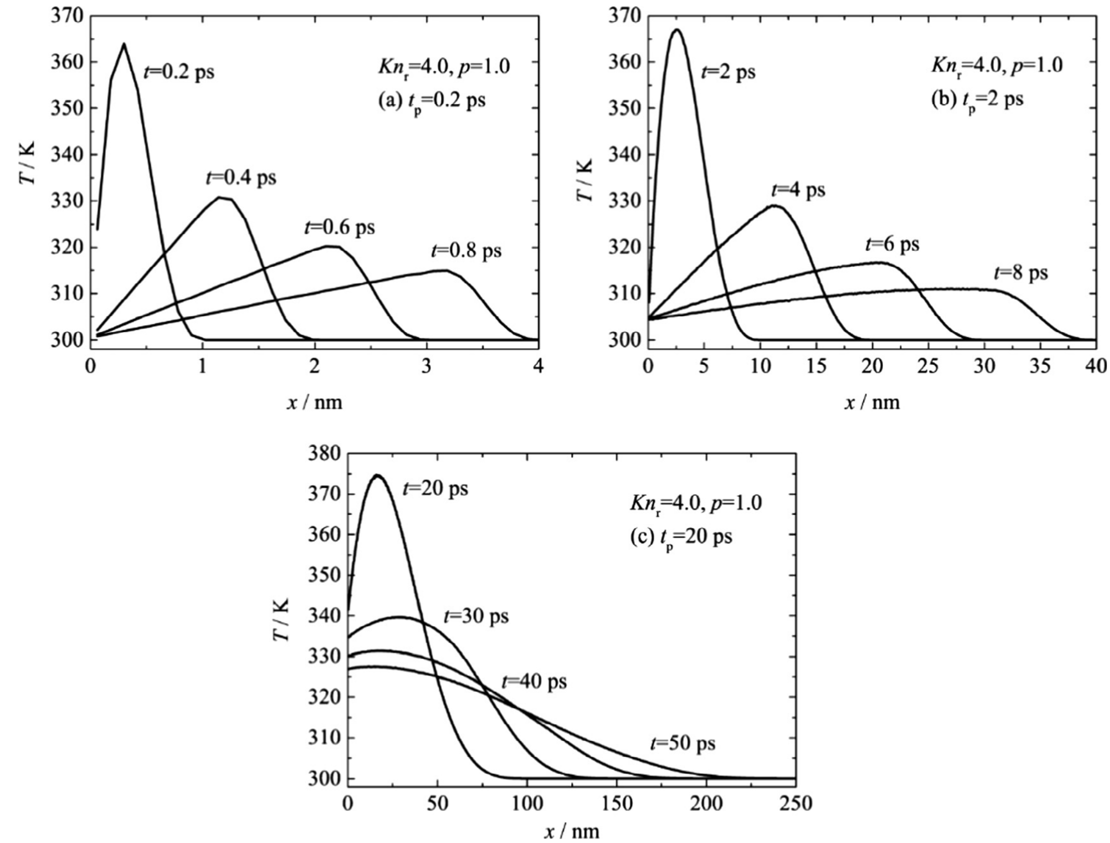

Fig. 5. Temperature profiles in nanowires (Kn_r = 4.0, p = 1.0) with various heat pulse periods (a) t_p = 0.2 ps, (b) t_p = 2 ps, (c) t_p = 20 ps.

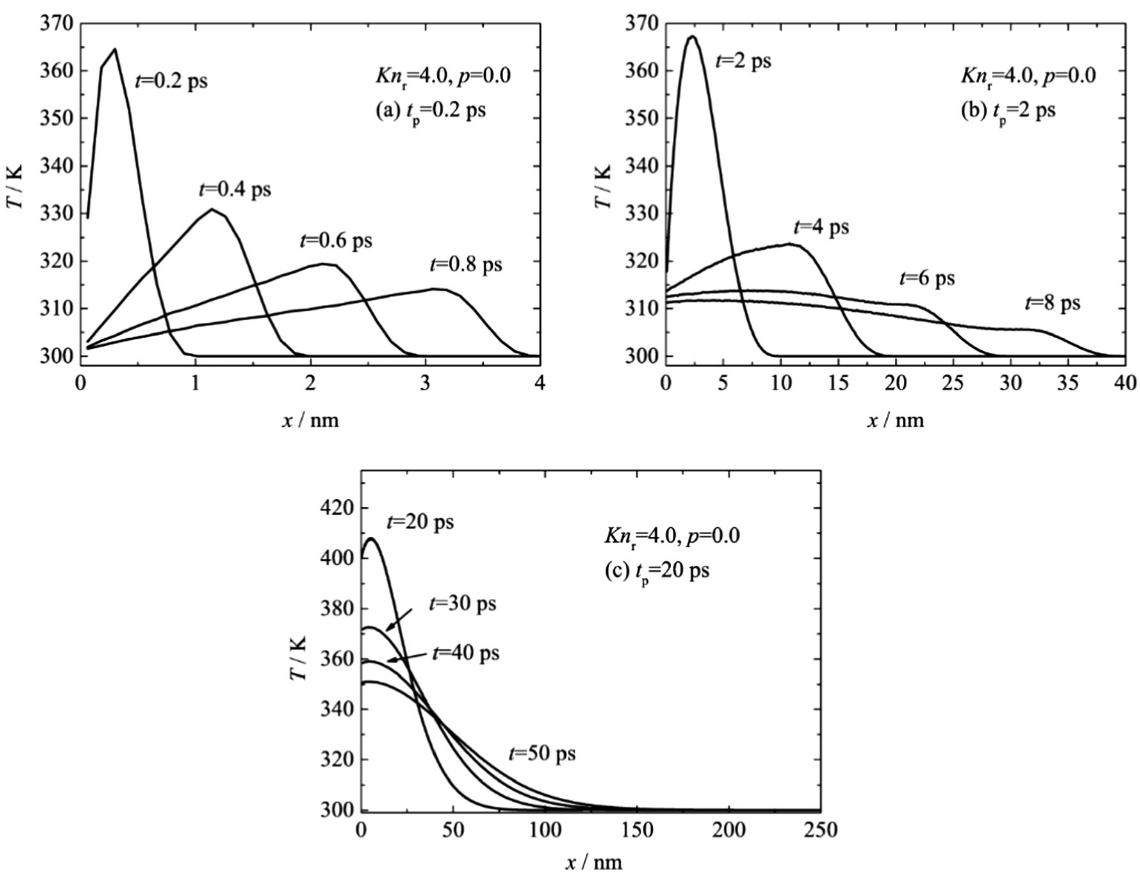

Fig. 6. Temperature profiles in nanowires (Kn_r = 4.0, p = 0.0) with various heat pulse periods (a) t_p = 0.2 ps, (b) t_p = 2 ps, (c) t_p = 20 ps.

regime. Different phonon transport regimes will then be measured in experiments using different heat pulse periods and different heat conduction laws with the appropriate characteristic parameters should be used to analyze the experimental data.

4. Conclusions

Ballistic thermal wave propagation in nanowires is influenced by the characteristic parameters of the boundary including the radial Knudsen number, Kn_r, and the specularity parameter, p. A fully specular boundary does not have any substantial effect on the thermal wave propagation. For large radial Knudsen numbers and small specularity parameters, the phonon boundary scattering causes quicker damping of the wave peaks and more uniform temperature distributions (i.e. close to the diffusive condition) for phonon ballistic-diffusive transport. Thus, the phonon boundary scattering accelerates the change from the phonon ballistic-diffusive regime to the diffusive transport regime, but the phonon boundary scattering acts as a thermal resistance in transient phonon diffusive transport by causing slower development of the thermal conduction with a smaller effective thermal conductivity.

Ballistic thermal wave propagation in nanowires is also significantly influenced by the temporal resolution, i.e. the heat pulse period, of the measurements. Different phonon transport regimes will be measured in experiments using different heat pulse periods, and the corresponding temperature results should be modeled using different constitutive equations. When the heat pulse period is very long, the temperature profiles can be modeled by Fourier's law with a modified effective thermal conductivity, even though the modified effective thermal conductivities differ in nanowires with different physical characteristics since the effective thermal conductivity is size and roughness dependent. When the heat pulse period is quite short, the measured temperatures are quite different from the predictions of Fourier's law and are strong functions of the dominated phonon scattering types, so an effective thermal conductivity for the classical framework cannot be easily defined and new constitutive equations with new parameters are needed to describe the thermal processes and guide the experimental data analyses.

Acknowledgements

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