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Discrete space-time model for heat conduction: Application to size-dependent thermal conductivity in nano-films

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ABSTRACT

An analytical discrete-variable model has been developed to describe heat conduction in nano-sized systems. The model assumes that the system consists of a homogeneous array of cells with characteristic size h; each cell interacts with the nearest neighbors in discrete time step τ and all the cells compute their new state simultaneously. In the continuum limit $h \to 0$ and $\tau \to 0$, the model reduces to classical heat diffusion equation of parabolic type or heat conduction equation of hyperbolic type, depending on the choice of scaling invariant. The model is applied to heat conduction in nano-films with emphasis on the transition from the diffusive to ballistic heat transport, which occurs with decreasing film thickness. This model provides a simple method for predicting in a self consistent manner the effective cross-plane thermal conductivities, the temperature jump at the boundaries, the heat flux across the film, and the temperature gradient within the film as functions of the film thickness. The results are in good agreement with molecular dynamic and Monte Carlo simulations.

1. Introduction

The classical heat conduction theory, which is based on the local equilibrium assumption, leads to the linear relation $q = -\lambda \nabla T$ between the heat flux q and the temperature gradient ∇T, where λ is the bulk thermal conductivity [1]. This relation known as Fourier law suggests that the heat flux $q(x,t)$ at a space point x and at a time moment t depends on the temperature gradient at the same space-time point, i.e. $\nabla T(x,t)$. In other words, Fourier law is local both in time and space. Strictly speaking heat transport is an inherently nonlocal phenomenon [1–12]. The heat flux at a point depends on the history of the heat carriers reaching the point at time t and the carriers arrive at the point in space having brought the energy from other points. Thus, there are essentially two important non-Fourier effects: (i) the one is related to the time lag between the heat flux and the corresponding temperature gradient - it can be called as a *time* non-local effect, which describes relaxation to local equilibrium, (ii) the other is a *space* non-local effect, which takes into account that the carriers come to a point from another distant point. Several theoretical methods have been proposed to study the local nonequilibrium effects [1–20]. Primarily, more attention has been paid to study the time-nonlocal (or relaxation) effects (see [1–6] and references therein), which, in particular, have been observed in metals under ultra-short laser irradiation [8] or during ultrafast phase transformations [6,17,18,20]. Recently, the trend towards miniaturization of electronic devices has increased the interest in space nonlocal effects during nano-scale heat conduction [1,2,7–47]. One of the most important characteristics of nano-scale heat conduction is that the thermal conductivity of nanostructures such as thin films, superlattices, nanowires and nanotubes is reduced significantly from that of the corresponding bulk materials depending on the sizes of nanostructures [1,2,9,11,13–16,19–43]. This effect has been observed, for example, in silicon and germanium films [14,23–25,27,29,31,32,36,38,41], two-dimensional black phosphorus [43], polycrystalline aluminum nitride [35], graphene and ultrathin graphite [22,28,40,42]. Molecular dynamic (MD) [24–26] and Monte Carlo (MC) [27] or simulations also demonstrate that the effective thermal conductivity of nano-systems is significantly lower than the bulk value and decreases with the system size. The size-dependent thermal conductivity implies the breakdown of the Fourier law in nanoscale heat transfer where the mean-free-path (MFP) of phonons h is comparable or even much larger than the material length scale L. As a consequence, the heat transport is no longer diffusive (i.e. dominated by collisions amongst the particles of the system) but becomes ballistic (i.e. dominated by collisions with the walls). The size-dependent thermal conductivity has been considered on the bases of Boltzmann's equation [9,11], EIT [1,14–16], phonon hydrodynamic equation [29], Landauer approach [23]. Usually, the non-Fourier heat conduction

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effects are considered by expanding in Taylor's series distribution function or corresponding thermodynamic functions in powers of small parameter $Kn = h/L \ll 1$ (the Knudsen number) around local equilibrium state. In such a case the non-local corrections are of the order of h/L, which implies that the transition from the nearly-Fourier heat conduction $h/L \ll 1$ to the non-local (ballistic) heat conduction $h/L \sim 1$ described on the basis of the expansion around local equilibrium state needs careful justification.

An alternative approach, which consider the problem of non-local heat conduction under a completely different angle, is based on the discrete variables (DV) formalism [6–8,12,17]. The DV model is an algorithm that describes discrete spatial and temporal evolution of complex dynamic systems, which consists of nano-sized elemental cells, by applying local interaction rules to the cells on a regular (or non-regular) lattice.

In this paper we use the DV model to study steady-state heat conduction in nano-films with emphasis on the transition from the diffusive to ballistic heat transport, which occurs with decreasing film thickness. In fact, the size-dependent thermal conductivity obtained earlier by more elaborated and computationally expensive methods is repeated in the results section as a validation for the DV formulation in the present work. In addition to the size-dependent thermal conductivity, the DV model allows us to calculate in a self-consistent manner the heat flux across the film, the temperature jumps at the boundaries between the film and the thermal reservoirs, and the internal temperature gradient within the film as functions of the film thickness.

2. Discrete heat conduction model

2.1. General formulation

The idea to describe the non-local heat conduction with space-time discrete variables is closely related to the concept of random walks on a lattice and cellular automata (see [6–8] and references therein). The discrete approach assumes that space and time are discrete variables and a media can be divided into cells of a characteristic length h and states of cells evolve simultaneously in discrete intervals of time τ. The characteristic length h is the minimum size of the cell, to which the local temperature $T(x, t)$ can still be assigned. The idea of the minimum space and time scales employed by the discrete model to describe far from local equilibrium processes [6–8,12,17] corresponds to the conclusion of Majumdar [9] that "since temperature at a point can be defined only under local thermodynamic equilibrium, a meaningful temperature can be defined only at points separated on an average by the phonon mean free path". The idea is also consistent with the concept of minimum heat-affected region suggested by Chen [11], which assumes that during phonon transport from a nanoscale heat source the minimum size of the heat affected region is of the order of the phonon mean free path [11,34]. Thus, in solids h is of the order of the phonon mean free path (MFP). For a sake of simplicity it is assumed here that MFP is frequency-independent, however, the proposed approach allows us to generalize the model for frequency-dependent MFP.

In a one-dimensional description (1D), the discrete approach gives a heat conduction equation as follows [6–8,12,17]

$$T(t+\tau,x) = \frac{1}{2}[T(t,x+h) + T(t,x-h)] + Q(t+\tau/2,x)/C \tag{1}$$

where Q is the heat source, C is the heat capacity, T is the kinetic temperature [13], which is usually used for nonequilibrium situations, although the most suitable definition of the nonequilibrium temperature is still an open problem [1,15]. Eq. (1) assumes that the temperature $T(t,x)$ is assigned to a discrete cell $(x-h/2, x+h/2)$, which center has a coordinate x. Within the cell, i.e. on the space interval $(x-h/2, x+h/2)$, the temperature does not change with x. $T(t,x+h)$ and $T(t,x-h)$ are the temperatures of the neighboring cells.

The discrete formalism implies that the energy exchange between the cells occurs on the border between the neighboring cells, which allows us to present the heat flux q as follows [6,10,12]

$$q(t+\tau/2,x) = \frac{Cv}{2}[T(t,x-h/2) - T(t,x+h/2)] \tag{2}$$

where $v = h/\tau$ is the heat-carrier (phonon) velocity, which assumed to be frequency-independent. Strictly speaking, this assumption is valid in the Debye approximation, while in a general case there are dispersive effects that make v itself depends on the phonon frequency. Note that in Eq. (2) x is a coordinate of the border between the neighboring cells with coordinates $x - h/2$ and $x + h/2$, respectively (see Fig. 1), i.e. there is a coordinate shift $x \to x + h/2$ in comparison with Eq. (1). This reflects the non-local property of heat conduction under far from equilibrium conditions that there is not only the time but also the space lag between the heat flux and the temperature. The Taylor expansions of Eqs. (1) and (2) allows one to rewrite them in an operator form as follows [12,17]

$$[\exp(\tau\partial_t) - \cosh(h\partial_x)]T = [\exp(\tau/2)\partial_t]Q/C \tag{3}$$

$$\exp\left(\frac{\tau}{2}\partial_t\right)q = -\frac{Cv}{2}\sinh\left(\frac{h}{2}\partial_x\right)T \tag{4}$$

Eqs. (1) and (2) can be used for computer simulation directly in the discrete form [7]. Now we turn to the question of the transition from the discrete Eqs. (1) and (2) to usual continuum description in the form of partial differential equations. Taylor expansions of Eqs. (3) and (4) in the continuum limit $h \to 0$ and $\tau \to 0$ contain an infinite number of terms with two small parameters h and τ. To obtain equations with a finite number of terms one should first specify an invariant of the continuum limit, which conserves a desirable property of the discrete model [6–8,12,17].

2.2. Diffusive continuum limit $h^2/2\tau = const > 0$

When $h^2/2\tau > 0$ at $h \to 0$ and $\tau \to 0$, Eq. (3) gives the classical heat conduction equation of parabolic type

$$\frac{\partial T}{\partial t} = \frac{\lambda}{C}\frac{\partial^2 T}{\partial x^2} + \frac{Q}{C}$$

where $\lambda = Ch^2/2\tau = Chv/2$ is the bulk thermal conductivity. In 2D and 3D, the discrete model gives $\lambda = Chv/4$ and $\lambda = Chv/6$, respectively. Note that the discrete model assumes that the heat carriers, which cross a plane x and contribute to the heat flux $q(x)$, come from distance $(x+h)$ and $(x-h)$ with average temperatures $T(x+h/2)$ and $T(x-h/2)$, respectively. Kinetic theory usually assumes that the heat carriers have temperatures $T(x+h)$ and $T(x-h)$, respectively, which in 3D gives $\lambda = Chv/3$. Thus, the result of the present model $\lambda = Chv/6$ takes into account that the temperature $T(x)$ may change on the intervals $(x, x-h)$ and $(x, x+h)$, which seems to give a more adequate value for the bulk thermal conductivity λ.

The requirement that the heat diffusivity $h^2/2\tau$ has a finite value at $h \to 0$ and $\tau \to 0$ implies that $v \to \infty$. Indeed, representing v as $v = \lambda/Ch$ we obtain that $v \to \infty$ at $h \to 0$. This is the so-called 'paradox' of propagation of thermal signals with infinite speed, which corresponds to the usual parabolic description [1]. This implies that the diffusive continuum limit is appropriate for processes with a relatively low characteristic velocity $V \ll v$ [6,20]. Corresponding continuum limit of the discrete equation for the heat flux q, Eq. (4), gives the classical Fourier law $q = -\lambda\nabla T$.