On Unsteady Heat Conduction in a Harmonic Crystal

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An analytical model of unsteady heat transfer in a one-dimensional harmonic crystal is presented. A nonlocal temperature is introduced as a generalization of the kinetic temperature. A closed system of differential-difference equations determining unsteady thermal processes is derived. For an instantaneous heat perturbation a time-reversible thermal wave equation is derived and solved. The resulting constitutive law for the heat flux in the considered system is obtained. This law significantly differs from Fourier's law and it predicts a finite velocity of the heat front and independence of the heat flux on the crystal length. The analytical results are confirmed by computer simulations.

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An understanding of heat transfer at microlevel is essential to obtain link between microscopic and macroscopic description of solids [1, 2]. As far as macroscopic scale level is concerned the Fourier law of heat conduction is widely and successfully used to describe heat transfer processes. At microscopic level, however, analytical and numerical investigations have shown substantial deviations from Fourier's law [3–5]. These inadequacies can be on principle addressed by using special laws of particles interactions [6–8] or complex enough structures [9, 10]. Recent experimental data however showed that Fourier's law is indeed violated in low-dimensional nanostructures [11–13]. This motivates interest to the simplest lattice models, in particular harmonic one-dimensional crystals (chains), where these anomalies are most prominent [14, 15]. Problems of this kind previously have been mainly addressed in the context of the steady state heat conduction [3–5]. The present work focuses on unsteady conduction regimes [10, 16, 17].

Here we suggest an approach that allows direct derivation of macroscopic heat conduction equations for harmonic systems in a one-dimensional, non-quantum case. The obtained equations differ substantially from the earlier suggested heat transfer equations [18, 19], however they are in excellent agreement with molecular dynamics simulations and previous analytical estimations [16]. The approach presented here is developed in context of one-dimensional systems, however the same ideas can be applied to systems of higher dimensions.

Thus we consider a one-dimensional crystal, described by the following equations of motion:

$$\ddot{u}_i = \omega_0^2(u_{i-1} - 2u_i + u_{i+1})\,, \qquad \omega_0 = \sqrt{C/m}, \tag{1}$$

where u_i is the displacement of the ith particle, m is the particle mass, C is the stiffness of the interparticle bond. The crystal is infinite: the index i is an arbitrary integer. The initial conditions are

$$u_i|_{t=0} = 0\,, \qquad \dot{u}_i|_{t=0} = \sigma(x)\varrho_i, \tag{2}$$

where ϱ_i are independent random values with zero expectation and unit variance; σ is variance of the initial velocities of the particles, which is a slowly varying function of the spatial coordinate $x = ia$, where a is the lattice constant. These initial conditions correspond to an instantaneous temperature perturbation, which can be induced in crystals, for example, by an ultrashort laser pulse [20]. The displacements as functions of time $u_i = u_i(t)$ can be found as a solution of the Cauchy problem (1)–(2). In addition, these functions are linearly dependent on the integration constants, which are random due to the random nature of the initial conditions (2).

The first analytical solution of a steady heat conduction problem for a harmonic chain was obtained in [21] using a covariance matrix for coordinates and momenta. In this paper a somewhat similar approach based on analysis of covariances for velocities [22–24] is used. We introduce a *nonlocal temperature* $\theta_n(x)$ as

$$k_B(-1)^n\,\theta_n(x) \stackrel{\text{def}}{=} m\langle \dot{u}_i \dot{u}_j\rangle, \tag{3}$$

where k_B is the Boltzmann constant, $n = j - i$ is the covariance index, $x = \frac{i+j}{2}a$ is the spatial coordinate, angle brackets stand for mathematical expectation: $\langle \dot{u}_i\rangle \equiv 0$, $\langle \dot{u}_i \dot{u}_j\rangle$ is the velocity covariance. If $n = 0$ then $i = j$ and quantity θ_n coincides with the kinetic temperature T: $\theta_0(x) = T(x) = \frac{m}{k_B}\langle \dot{u}_i^2\rangle$, where $i = x/a$. According to its definition, the introduced nonlocal temperature reflects a nonlocal nature of thermal processes in harmonic crystals and can be considered as a generalization of the kinetic temperature.

Let us calculate the second time derivative of θ_n using the dynamics equations (1) and the following two approximations.

1. The nonlocal temperature $\theta_n(x)$ is a slowly varying function of the spatial coordinate x (on the distances of order of the lattice constant a). This allows replacing the finite differences by spatial derivatives [25]. The approximation is adequate for processes that are sufficiently smooth in space, e. g. for spatial temperature profiles in a form of waves that are much longer then a.

2. The virial approximation [1]: time or spatial derivatives of mathematical expectations are small with respect to quantities that have non-zero values in thermodynamic equilibrium. In particular, this allows to express covariances of the bond strains in terms of the nonlocal temperature. The approximation is adequate for processes that are not too far from thermodynamic equilibrium.

Then after some transformations we obtain a differential-difference equation for the nonlocal temperature

$$\ddot{\theta}_n + \tfrac{1}{4}c^2(\theta_{n-1} - 2\theta_n + \theta_{n+1})'' = 0, \tag{4}$$

where $c = \omega_0 a$ is the speed of sound. Equation (4) can be also interpreted as an infinite system of partial differential equations. The initial condition for equation (4) follows from (2):

$$\theta_n|_{t=0} = T_0(x)\delta_n\,, \qquad \dot{\theta}_n|_{t=0} = 0, \tag{5}$$

where $T_0(x) = \frac{1}{2k_B}m\sigma^2(x)$ is the initial temperature distribution; $\delta_n = 1$ for $n = 0$ and $\delta_n = 0$ for $n \neq 0$. The initial conditions are taken after a fast transition process, which results, according to the virial theorem, in a double reduction of the initial kinetic temperature [23].

Thus, a slow long-wave dynamics of the nonlocal temperature is described by initial value problem (4)–(5). Note that this problem is deterministic, since it is expressed in terms of mathematical expectations. The integral Fourier transform of (4)–(5) in the spatial coordinate x gives for the Fourier image $\hat{\theta}_n(t,k)$ the initial value problem

$$\begin{aligned} &\ddot{\hat{\theta}}_n = \tfrac{1}{4}c^2k^2(\hat{\theta}_{n-1} - 2\hat{\theta}_n + \hat{\theta}_{n+1}),\\ &\hat{\theta}_n|_{t=0} = \hat{T}_0(k)\delta_n\,, \qquad \dot{\hat{\theta}}_n|_{t=0} = 0, \end{aligned} \tag{6}$$

where k is the spatial frequency, $\hat{T}_0(k)$ is the Fourier image of the initial temperature distribution $T_0(x)$. Let us note the similarity between (1)–(2) and (6). Indeed, initial value problem (6) can be interpreted as a motion of a harmonic chain having an initial shift of the central particle. This problem has analytical solution in terms of the Bessel functions of the 1st kind [26]: $\hat{\theta}_n(t,k) = \hat{T}_0(k)J_{2n}(ckt)$. From the practical point of view the most interesting case is $n = 0$, which gives Fourier image $\hat{T}(t,k)$ of the kinetic temperature distribution:

$$\hat{T}(t,k) = \hat{T}_0(k)J_0(ckt). \tag{7}$$

From (7) it follows that the image $\hat{T}(t,k)$ satisfies the Bessel differential equation

$$\ddot{\hat{T}} + \frac{1}{t}\dot{\hat{T}} = -c^2k^2\hat{T}. \tag{8}$$

Fourier inversion of (8) gives a partial differential equation for the temperature field

$$\ddot{T} + \frac{1}{t}\dot{T} = c^2T''. \tag{9}$$

The corresponding initial conditions follow from (5):

$$T|_{t=0} = T_0(x)\,, \qquad \dot{T}|_{t=0} = 0. \tag{10}$$

Fourier inversion of the representation (7) gives an analytical solution of the initial value problem (9)–(10):

$$T(t,x) = \frac{1}{\pi}\int_{-t}^{t}\frac{T_0(x-c\tau)}{\sqrt{t^2-\tau^2}}\,d\tau. \tag{11}$$

Thus, the evolution of the temperature field in a one-dimensional crystal after an instantaneous thermal perturbation is described by partial differential equation (9) with initial conditions (10) or by integral formula (11). According to (11) the thermal front propagates with the sound speed c (in contrast to the thermal conductivity based on Fourier's law where an unphysical instantaneous signal propagation is realized). The obtained wave behavior of the heat front is similar to predictions of the wave theories of heat conduction [18, 19]. However, the obtained solution has important differences, which will be shown in the text to follow.

Let us consider the heat flux. For the considered system it can be represented [4, 5, 27] as

$$q = \tfrac{1}{2}C\langle(u_i - u_{i+1})(\dot{u}_i + \dot{u}_{i+1})\rangle. \tag{12}$$

The heat flux q satisfies the energy balance equation

$$\rho k_B\dot{T} = -q', \tag{13}$$

where $\rho = 1/a$ is the density (number of particles per unit volume). Joint consideration of equations (9) and (13) gives the constitutive law for the heat flux

$$\dot{q} + \frac{1}{t}q = -k_B\rho c^2T', \tag{14}$$

which replaces Fourier's law in the considered system. Alternatively, the law (14) can be derived directly, in the same way as equation (9) is derived. An integral representation for the heat flux follows from (11) and (14):

$$q(t,x) = \frac{k_B\rho c}{\pi t}\int_{-t}^{t}\frac{T_0(x-c\tau)}{\sqrt{t^2-\tau^2}}\,\tau d\tau. \tag{15}$$

Comparison of the obtained heat transfer equation (9) with the heat equation based on Fourier's law and the thermal wave equation based on the Maxwell-Cattaneo-Vernotte (MCV) law is given in table 1. The latter equation and equation (9) have similar form and somewhat similar behavior (e. g. a finite velocity of the heat front propagation). However, there are significant differences. First is that τ, a material constant, is replaced in (9)

	Heat equation (Fourier)	Thermal wave equation (MCV)	Equation (9) (present paper)
a)	$\dot{T} = \beta T''$	$\ddot{T} + \frac{1}{\tau}\dot{T} = \frac{\beta}{\tau}T''$	$\ddot{T} + \frac{1}{t}\dot{T} = c^2 T''$
b)	$q = -\kappa T'$	$\dot{q} + \frac{1}{\tau}q = -\frac{\kappa}{\tau}T'$	$\dot{q} + \frac{1}{t}q = -k_B \rho c^2 T'$
c)	$e^{-\beta k^2 t}$	$\approx e^{-\frac{t}{2\tau}} \cos(kct)$	$J_0(kct) \approx \frac{\cos\left(kct - \frac{\pi}{4}\right)}{\sqrt{\frac{\pi}{2}kct}}$

TABLE I. (a) heat transfer equations, (b) equations connecting heat flux and temperature, (c) decay laws for the sinusoidal heat perturbation. Notations: t is time (variable), τ is the relaxation time (constant), β is the thermal diffusivity, κ is the thermal conductivity, c is the sound speed, ρ is the density, k_B is the Boltzmann constant, k is the spatial frequency. Approximation (c) for the thermal wave equation is valid for large k, approximation for J_0 is obtained for relatively large t.

by the physical time t. Second is that equation (9) is time-reversible: it is not changing when t is replaced by $-t$, same as the original microscopic equation (1). On contrary, both classical and wave equations of heat conduction are irreversible. The contradiction between time-reversibility of the classical microscopic equations and irreversibility of the corresponding macroscopic continuum equations is one of the opened questions of the modern physics [1]. The obtained reversible macroscopic equation of heat conduction may be a step towards solution of this problem.

We consider now the problem of a sinusoidal perturbation of a temperature field [10, 28]. The initial temperature is

$$T_0(x) = A_0 \sin kx + B, \tag{16}$$

where A_0 and B are temperature constants, $k = 2\pi/\lambda$ is the spatial frequency, λ is the wavelength of the perturbation. Formulas (11) and (15) give exact analytical solution for the temperature and heat flux

$$\begin{aligned} T(t,x) &= A_0 J_0(kct) \sin kx + B, \\ q(t,x) &= -k_B \rho c A_0 J_1(kct) \cos kx, \end{aligned} \tag{17}$$

where J_0 and J_1 are the Bessel functions of the 1st kind.

To justify the assumptions in derivation of the analytical solution we compare it with results of molecular dynamics (MD) simulations. Equations (1) are solved by the central differences method, the time step is $0.01\tau_0$, where $\tau_0 = 2\pi/\omega_0$. The initial conditions (16) are set by a random number generator, the wavelength λ is equal to the length of the chain containing 10^4 particles. To provide correspondence with the analytical approach used above, 10^4 realizations of such chain with an independent random initiation are computed. To optimize the computations all chains are joined at end-points to form a long chain (10^8 particles) with periodic boundary conditions. The results of the computations are compared with analytical solution (17) in Fig. 1. The horizontal axis

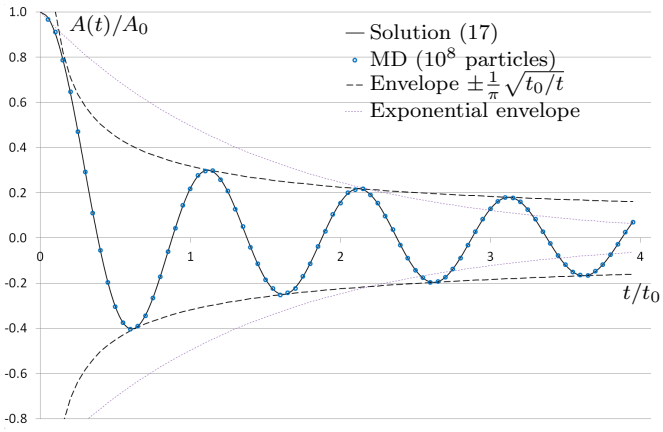

FIG. 1. Oscillational decay of the thermal perturbation amplitude for 1D harmonic crystal. Comparison of the analytical solution (17) with the MD modeling results (10^4 linked chains containing 10^4 particles each). Dashed lines show the envelope proportional to $1/\sqrt{t}$ and also an exponential envelope inherent to the thermal wave equation.

in Fig. 1 represents the dimensionless time t/t_0, where $t_0 = \lambda/c$; the vertical axis stands for the oscillation amplitude $A(t)$, which is computed as the first coefficient of a spatial Fourier expansion of the temperature field. According to Fig. 1 there is an excellent agreement between the computational results and the analytical curve.

Due to the Bessel function properties [26], the temperature and heat flux (17) have an oscillational decay, where the oscillation amplitude is asymptotically proportional to $1/\sqrt{t}$. The same asymptotics has been obtained in [16] for one-dimensional harmonic crystals. In Fig. 1 the envelope proportional to $1/\sqrt{t}$ is shown by the dashed lines, perfectly bounding both analytical and computational graphs. The existing theories of heat conduction [18, 19], such as Fourier's, Maxwell-Cattaneo-Vernotte (MCV), dual-phase-lag [29], and spacetime-elasticity [17] predict an exponential decay of the sinusoidal perturbation amplitude, since all of them result in linear differential equations with constant coefficients. In table 1 a comparison of the analytically obtained decay law for $A(t)/A_0$ with the results based on some other theories is demonstrated, an exponential envelope inherent to the thermal wave model is also shown in Fig. 1. Thus, among all mentioned theories only the current one gives an analytical solution, which agrees with the MD simulations and asymptotic estimations for harmonic chains [16].

Let us consider now a stepwise initial temperature distribution, modelling heat transfer between a hot and a

cold bodies:

$$x<0: \quad T(x)=T_2\,, \qquad x>0: \quad T(x)=T_1, \tag{18}$$

where $T_2>T_1$. In this case the integral representations (11), (15) yield for $|x|\le ct$ an exact analytical solution

$$\begin{aligned} T(t,x) &= T_1+\tfrac{\Delta T}{\pi}\arccos\tfrac{x}{ct}, \\ q(t,x) &= \tfrac{k_B\rho c\Delta T}{\pi}\sqrt{1-\left(\tfrac{x}{ct}\right)^2}, \end{aligned} \tag{19}$$

where $\Delta T=T_2-T_1$; for $x>ct$ the original temperature distribution remains and the heat flux is zero. According to (19) the heat front propagates with the sound speed c and the heat flux through cross-section $x=0$ is constant and equal to $\frac{1}{\pi}k_B\rho c\Delta T$. In contrast, use of Fourier's law for the same problem gives the heat flux proportional to $t^{-1/2}$, which is infinite at $t=0$ (an unphysical consequence of Fourier's law). From (19) it follows that the temperature difference ΔT is realized on the interval $x\in[-ct,ct]$ and the length of this interval is increasing with time as $2ct$. Thus, the heat flux depends on the temperature difference rather than on the temperature gradient. This is in qualitative agreement with the known phenomenon of thermal superconductivity: the heat flux through a one-dimensional harmonic crystal placed between two thermal reservoirs does not depend on the length of the crystal [5, 21].

In Fig. 2 the analytical solution (19) is compared with computer simulations for $T_2=2T_1$. The above described

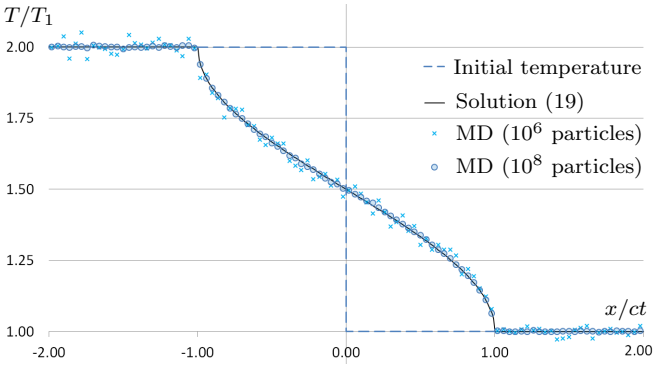

FIG. 2. Heat transfer between hot (left) and cold (right) areas of 1D harmonic crystal. The analytical solution (19) is compared with the computer simulation (MD): 10^3 chains containing 10^3 particles each (cross is an average over 10 particles); 10^4 chains containing 10^4 particles (circle is an average over 100 particles).

computation procedure is used. Fig. 2 shows the initial temperature distribution, the analytical solution, and the computation results obtained at $t=t_0/8$ using 10^6 and 10^8 particles (only half of the chain is shown). Convergence to the analytical solution with the increase of the system size is clearly seen.

Fig. 3 shows a part of Fig. 2 corresponding to positive x. For symmetry reasons this case can be interpreted as a problem of a half-space heating: the initial temperature for $x>0$ is T_1 and the boundary condition at $x=0$ is $T=(T_2+T_1)/2>T_1$. The advantage of this formula-

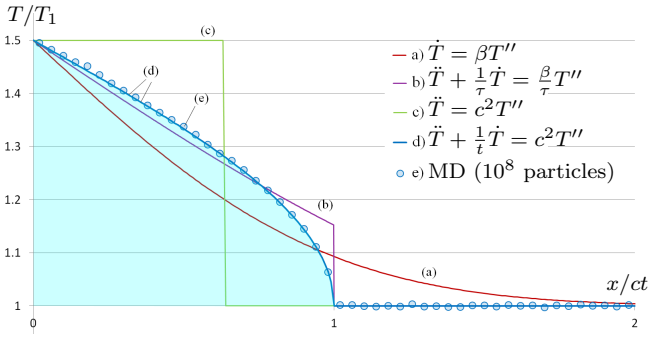

FIG. 3. Heat propagation for different 1D models: a) heat equation, b) thermal wave equation, c) wave equation, d) equation (9), e) computer simulation for 1D harmonic crystal (10^4 chains containing 10^4 particles each).

tion is that the constant boundary temperature is maintained without any thermostat. This is important since the heat transfer can substantially depend on the thermostat properties [30]. Solutions of the considered problem using four different continuum equations are compared in Fig. 3 with the simulation results. Parameters are chosen in such a way that the total heat quantity transferred through the cross-section $x=0$ (area under each curve) is equal for all models and the heat front (when it exists) propagates with the sound speed c. According to Fig. 3 the computation results almost coincide with the analytical solution of equation (9) and significantly differ from the solutions based on the other theories of thermal conduction. Indeed, the classical heat equation predicts no heat front, the thermal wave equation gives a stepwise front, while the real heat front is described by a smooth curve having a vertical tangent at $x=ct$. Note that the thermal wave (MCV) equation behaves as wave equation at small times and as heat equation at large times [31]. However, according to the analytical solution (19) and the presented computer simulations, the heat transfer in a one-dimensional harmonic crystal is self-similar, i. e. $T=T(\frac{x}{ct})$.

Thus, a nonlocal temperature (a generalization of the kinetic temperature) is introduced in this work to obtain closed description of thermal transfer in a one-dimensional harmonic crystal. Finally this yields to a partial differential equation (9) for the kinetic temperature, which can be referred to as the time-reversible thermal wave equation. The resulting macroscopic constitutive law (14) (an alternative of Fourier's law for the considered system) predicts a finite velocity of the heat front and independence of the heat flux on the crystal length. The analytical findings are in excellent agreement with

the molecular dynamics simulations and previous analytical estimations. The obtained results are relevant to aspects of nanotechnology that involve heat transfer processes in high purity nanostructures [11, 12, 32].

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