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ANOMALOUS HEAT TRANSFER IN ONE-DIMENSIONAL DIATOMIC HARMONIC CRYSTAL

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Abstract. The work is devoted to description of unsteady thermal processes in low-dimensional materials. One-dimensional harmonic crystals with alternating masses and stiffnesses are considered. Analytical solution demonstrates the ballistic nature of heat propagation, which is confirmed by numerical simulations based on the particle dynamics method. It is shown that temperature distribution propagates as two consecutive thermal fronts with finite speed, and its initial shape is preserved.

Keywords: mathematical modeling, low-dimensional materials, discrete media, thermal processes, heat transfer, lattice dynamics, harmonic crystal, polyatomic lattice

1. Introduction

The relevance of this study is connected with the active development of new technologies for creating materials that allow to regulate the material composition and structure at the atomic level [1-3]. The properties of low-dimensional materials are often unique, which opens up promising opportunities for their application [4]. For example, the hexagonal boron nitride has high stability, chemical resistance, hardness, strength and thermal conductivity [5, 6]. In general, low-dimensional materials have a complex crystal structure. For example, two-dimensional graphene lattice consists of two sublattices formed by carbon atoms, and the sublattices of hexagonal boron nitride, binary boron and nitrogen compound, are formed by two different kinds of atoms. Filamentary nanocrystals (nanowires, nanowhiskers) can be formed by either one type of atoms (silicon, carbon-carbine), or several ones (gallium arsenide, indium phosphide). Hence, the development of models that would correctly describe the physical and mechanical properties of such media and structures, including non-stationary thermal processes, becomes particularly important. It should be noted that the existing mathematical models are often not applicable to low-dimensional structures. For example, recent experimental studies have shown that heat propagation at the nanoscale has peculiar properties [6-8]. In particular, the Fourier law, which implies the diffusive type of heat spread, is not fulfilled for low-dimensional structures; in contrast, the heat propagation in nanostructures is of a ballistic nature [7, 9]. The analytical solution demonstrating the anomalous heat propagation in one-dimensional harmonic chain was first presented in [10]. The solution was obtained for the stationary problem of heat propagation between two thermal reservoirs with different temperatures, and it was shown that the thermal resistance does not depend on the length of the chain, which contradicts the Fourier law; harmonic crystals consisting of particles with different masses were considered, for example, in [11, 12].

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In recent works [13-17], a method that allows analytical description of thermal processes in harmonic crystals has been developed. This paper is devoted to application of this method to one-dimensional harmonic crystals with alternating masses and stiffnesses. The object of investigation is a harmonic crystal, which is a crystal lattice consisting of material points interacting via linearized forces. The principle of separation of fast and slow thermal processes is applied. Characteristic time for a fast process is of order of several periods of atomic vibrations. Fast motions refer to fluctuations in the kinetic temperature associated with the partial transfer of the kinetic energy to thermal energy; in polyatomic crystals, it is accompanied by the redistribution of kinetic energy over the unit cell's degrees of freedom. Characteristic time for a slow process is much larger than a period of atomic vibrations [18, 19]; heat transport, i.e. time evolution of the spatial distribution of kinetic temperature, is a slow process. In this paper, analytical solutions are given for two unsteady heat transfer problems: (i) cold and hot half space contact and (ii) propagation of an initially rectangular thermal perturbation. Analytical results are verified by numerical simulation based on the particle dynamics method.

2. Problem statement

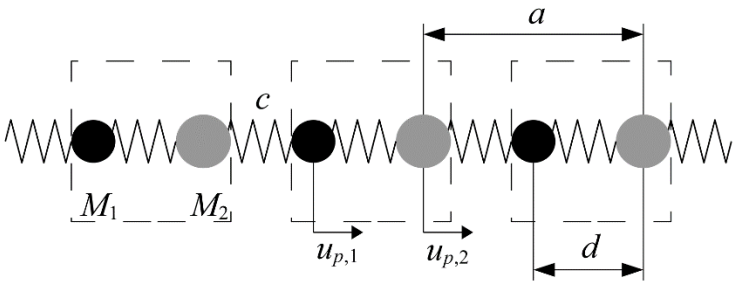

Fig. 1. One-dimensional harmonic crystal with alternating masses

Lattice dynamics equations. Initial conditions. Particle dynamics equations for an infinite one-dimensional harmonic crystal with alternating masses (Fig. 1) have the form:

$$\ddot{u}_{p,1} = \omega_1^2\left(u_{p-1,2} - 2u_{p,1} + u_{p,2}\right), \quad \ddot{u}_{p,2} = \omega_2^2\left(u_{p,1} - 2u_{p,2} + u_{p+1,1}\right). \tag{1}$$

Here $u_{p,1}$ and $u_{p,2}$ are displacements of the particles with masses M_1 and M_2 which belong to the pth unit cell; c is the bond stiffness, and the respective frequencies are $\omega_1 = \sqrt{c/M_1}$ and $\omega_2 = \sqrt{c/M_2}$. Following [20], we introduce the equilibrium interparticle distances: d is the one inside the unit cell, and $a - d$ is the distance between the neighboring particles from different cells. Hence, the so-called length of the unit cell is equal to a. Consequently, for the case of $\omega_1 = \omega_2$ the system yields to one-dimensional harmonic chain with the unit cell length equal to $a/2$.

In order to reduce the number of unknowns, let us introduce a parameter $\alpha \in (0, \infty)$:

$$M_1 = \alpha M, \quad M_2 = M/\alpha \quad \Longrightarrow \quad \omega_1 = \omega\sqrt{1/\alpha}, \quad \omega_2 = \omega\sqrt{\alpha}. \tag{2}$$

Taking the symmetry of its definition (2) into account, we can restrict ourselves to $\alpha \in (0,1)$.

The initial conditions are written as follows

$$u_{p,1}|_{t=0} = 0, \quad u_{p,2}|_{t=0} = 0, \quad \dot{u}_{p,1}|_{t=0} = v_{p,1}, \quad \dot{u}_{p,2}|_{t=0} = v_{p,2}, \tag{3}$$

where $v_{p,i}$ are random velocities with zero mean, i.e. their mathematical expectations are equal to zero. Such initial conditions are used, for instance, to model an ultrashort laser

impact [21, 22]. Note, that the solution of the resulting system (1)-(3) will be a set of random values.

Dispersion relation. Let us seek the solution of (1)-(2) in the wave form with frequency Ω and one-dimensional wave vector K:

$$u_{p,1} = C_1 e^{-I(\Omega t + Kpa)}, \quad u_{p,2} = C_2 e^{-I(\Omega t + Kpa)}. \tag{4}$$

Hence, the dispersion relation is determined as

$$\Omega_{1,2}^2 = \omega^2 \left[\left(\alpha + {}^1\!/_{\alpha}\right) \pm \sqrt{\alpha^2 + {}^1\!/_{\alpha^2} + 2\cos Ka} \right]. \tag{5}$$

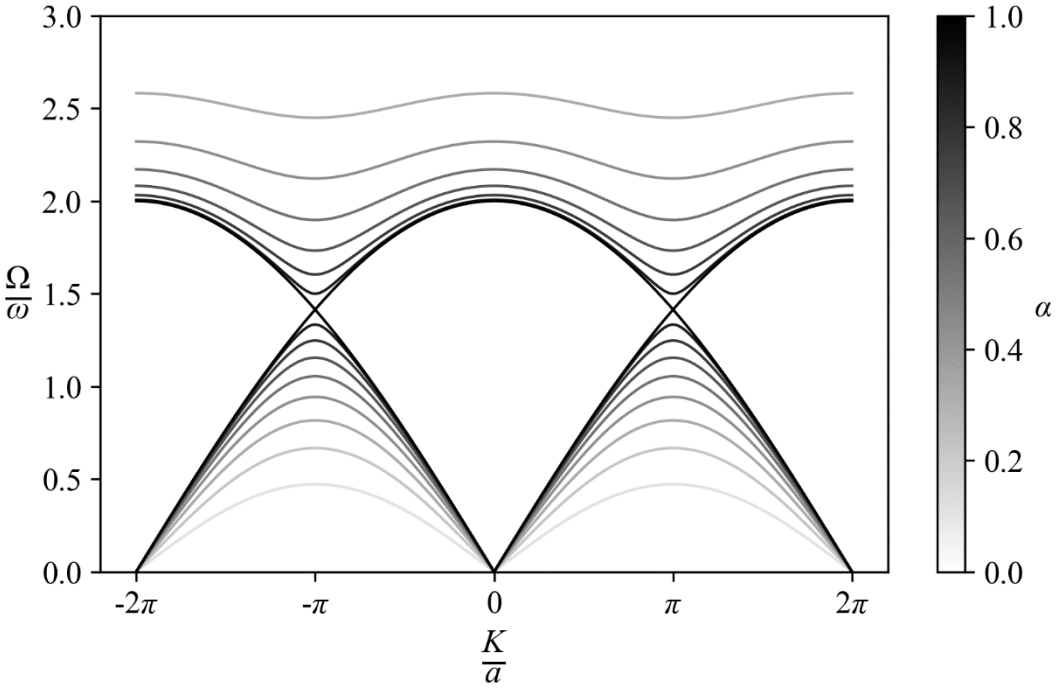

Fig. 2. Dispersion curves for one-dimensional diatomic harmonic crystals with various mass ratios $0 < \alpha < 1$. Black line ($\alpha = 1$) corresponds to the one-dimensional harmonic chain

Figure 2 shows the branches of dispersion relation for one-dimensional diatomic harmonic crystal at different values of the parameter $\alpha \in (0,1)$. For example, the curves for $\alpha = 0.8$ correspond to the mass ratio for the two-dimensional hexagonal boron nitride (the exact value is 0.772), which possesses unique physical and mechanical properties [5, 6].

If the masses differ slightly, i.e. $\alpha \to 1$, optic and acoustic branches Ω_1 and Ω_2 merge. Consequently, we obtain two dispersion curves for monoatomic chain, shifted by 2π along the horizontal axis relative to each other. The appearance of the two curves is due to the change in the translational symmetry; in this case we still solve the system (1) of the two equations for two neighboring particles.

Differentiation of (5) with respect to the wave vector leads to the following representation of group velocities

$$c_{g_{1,2}} = {}^{d\Omega_{1,2}}\!/_{dK} = \mp sign(Ka) \left({}^{a\omega^2 \sin Ka}\!/_{2\Omega_{1,2}\sqrt{\alpha^2 + {}^1\!/_{\alpha^2} + 2\cos Ka}} \right). \tag{6}$$

Note, that the dispersion curves (Fig. 2) will look the same for the system with alternating stiffnesses provided that the ratio of its parameters is kept ${}^{\omega_2}\!/_{\omega_1} = \alpha$.

3. Heat propagation

Kinetic temperature. Let us now follow [13-17] and consider the transfer from the stochastic problem for particle displacements (1)-(3) to closed deterministic one for the statistical characteristics of pairs of particles.

First, we introduce a spatial coordinate $x = pa$, which is one of the convenient ways to identify a pth unit cell. Then, we introduce the kinetic temperature $T(x,t)$ proportional to the sum of the kinetic energies of the particles in the unit cell:

$$k_B T(x,t) = {}^{1}\!/_{2}\left(M_1\langle \dot{u}_{p,1}^2\rangle + M_2\langle \dot{u}_{p,2}^2\rangle\right), \quad (7)$$

where k_B is Boltzmann constant, and brackets $\langle ... \rangle$ denote mathematical expectation. Consequently, the velocities $v_{p,i}$ (see initial conditions (3)) are the random velocities with the variance $\langle v_{p,i}^2\rangle = {}^{2k_B T_0(x)}\!/_{M_i}$, where $T_0(x) = T(x,0)$.

Thus, the conditions (3) mean that at the initial time, the particles have random velocities corresponding to a certain temperature field. This field correlates with the initial kinetic temperature of the system, while the potential energy is initially zero. This, in turn, means that, according to the virial theorem [13, 16, 23], after a certain period of time, the kinetic and potential energies will equilibrate. Characteristic time of this process is of order of several periods of atomic vibrations, and it is referred to as fast process. Also, at such times the kinetic energy is redistributed over the degrees of freedom inside the unit cell [17]. On the contrary, heat transport is a slow process, for which the characteristic time is much larger.

It has been demonstrated that the propagation of the thermal perturbation $T_0(x)$ in a monoatomic chain is described by the formula [16]:

$$T(x,t) = T_F + T_S$$
$$T_F = {}^{T_0(x)}\!/_{4\pi}\int_{-\pi}^{\pi}\cos 2\Omega t\, d(Ka) \quad (8)$$
$$T_S = {}^{1}\!/_{8\pi}\int_{-\pi}^{\pi}\left[T_0\left(x + c_g(Ka)t\right) + T_0\left(x - c_g(Ka)t\right)\right]d(Ka)$$

where Ω is frequency, determined by the dispersion relation, K is one-dimensional wave vector, and c_g is group velocity. At large times fast processes T_F vanish, and temperature field is a superposition of thermal waves travelling with the speed c_g and having the shape of the initial perturbation $T_0(x)$.

Analysis of fast processes for one-dimensional diatomic crystal is given in [17]. Specifically, it is demonstrated that the solution oscillates around the half of the initial temperature T_0, and its amplitude decays as ${}^{1}\!/_{\sqrt{t}}$. At larger times when fast processes are negligible, formulae (8) for polyatomic chain can be rewritten

$$T(x,t) \approx T_S = {}^{1}\!/_{8\pi N}\int_{-\pi}^{\pi}\sum_{j=1}^{N}\left[T_0\left(x + c_{g_j}(Ka)t\right) + T_0\left(x - c_{g_j}(Ka)t\right)\right]d(Ka), \quad (9)$$

where N is the number of atoms in the unit cell; for diatomic chain $N = 2$; c_{g_j} are the respective group velocities (6). Further, several solutions will be constructed and compared with the results of numerical simulation using the particle dynamics method. Formula (9) means that the heat propagation has ballistic character, which differs from the Fourier law. In the case of $N = 1$, formula (9) was derived for scalar lattices [16].

Cold and hot half space contact. Let the initial thermal perturbation have the form of a Heaviside function:

$$T_0(x,0) = T_0 H(x), \quad \dot{T}(x,0) = 0, \quad (10)$$

where T_0 is the temperature of $x > 0$ half-space before kinetic and potential energy equilibrate. Then, for the considered diatomic system, formula (9) yields to

$$T(x,t) = {}^{T_0}/_{8\pi} \int_{-\pi}^{\pi} \big[H\big(x + c_{g_1}(Ka)t\big) + H\big(x - c_{g_1}(Ka)t\big) + H\big(x + c_{g_2}(Ka)t\big) + H\big(x - c_{g_2}(Ka)t\big)\big] d(Ka). \quad (11)$$

Taking into account, that $H(x) = H(\lambda x)$ we finally obtain

$$T\left({}^{x}/_{t}\right) = {}^{T_0}/_{8\pi} \int_{-\pi}^{\pi} \left[H\left({}^{x}/_{t} + c_{g_1}(Ka)\right) + H\left({}^{x}/_{t} - c_{g_1}(Ka)\right) + H\left({}^{x}/_{t} + c_{g_2}(Ka)\right) + H\left({}^{x}/_{t} - c_{g_2}(Ka)\right)\right] d(Ka). \quad (12)$$

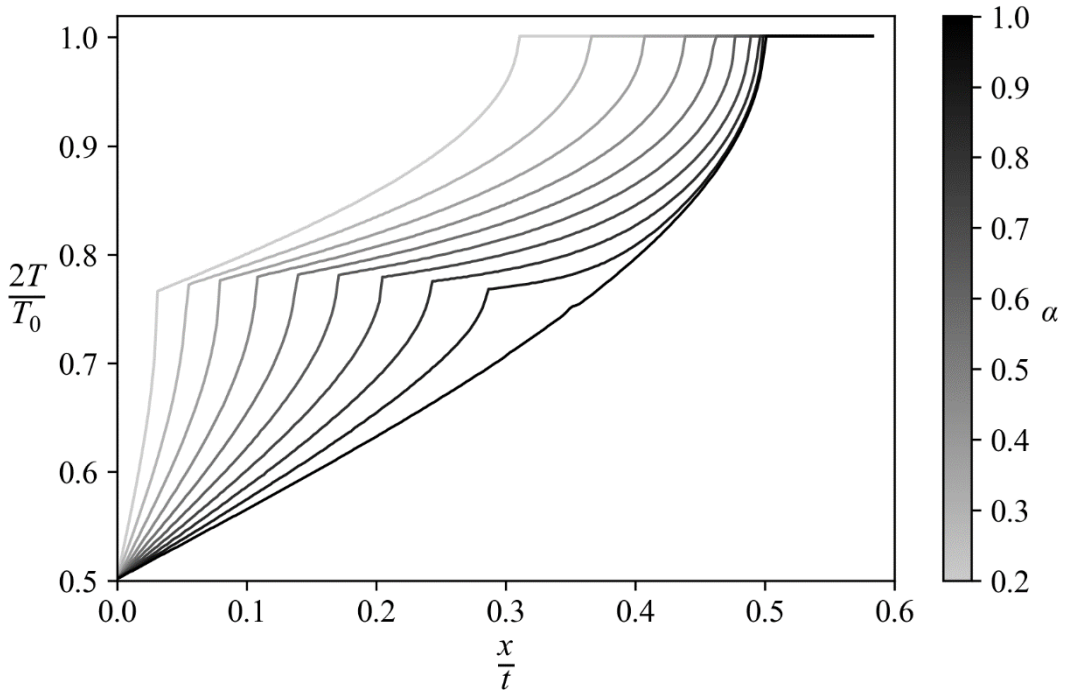

Fig. 3. Propagation of a Heaviside function in one-dimensional diatomic chain with different mass ratios $0 < \alpha < 1$

Figure 3 shows the result of integration of (12) for different values of the parameter $\alpha \in (0,1)$. For $\alpha \neq 0$ and $\alpha \neq 1$ there are two thermal fronts travelling with finite speeds [16], i.e. maximum values of group velocities c_{g_1} and c_{g_2} respectively (6). Note, that $c_{g_2}^{max}$, corresponding to the acoustic branch, is always larger than $c_{g_1}^{max}$.

If $\alpha \to 1$, the two fronts merge and the solution coincides with the solution for the monatomic chain with the unit cell length equal to ${}^{a}/_{2}$ [13]:

$$T\left({}^{x}/_{t}\right) = \begin{cases} {}^{T_0}/_{2} - {}^{T_0}/_{2\pi} \arccos\left({}^{2x}/_{a\omega t}\right), & 0 \le x \le {}^{a\omega t}/_{2} \\ {}^{T_0}/_{2}, & x > {}^{a\omega t}/_{2} \end{cases}, \quad (13)$$

To verify the obtained solution (12), a numerical simulation based on the particle dynamics method was carried out. A sample consisting of 1000 particles was considered, for which equations (1) were solved with initial conditions:

$$u_{p,1}|_{t=0} = 0, \quad u_{p,2}|_{t=0} = 0, \quad \begin{cases} \dot{u}_{p,1}|_{t=0} = v_{p,1}, & \dot{u}_{p,2}|_{t=0} = v_{p,2}, & p \ge 0 \\ \dot{u}_{p,1}|_{t=0} = 0, & \dot{u}_{p,2}|_{t=0} = 0, & p < 0 \end{cases}, \quad (14)$$

and periodic boundary conditions. The kinetic temperature was calculated using formula (7), where the mathematical expectation was approximated by averaging over 20 realizations with

various random initial conditions. Comparison of the analytical solution (12) with the numerical solution has shown a good agreement up to small thermal oscillations near $x \approx 0$.

Propagation of rectangular thermal perturbation. For further analysis of the limiting cases when $\alpha \to 0$ and $\alpha \to 1$, let us consider the following initial temperature distribution

$$T_0(x,0) = T_0\big(H(x+l) - H(x-l)\big), \quad \dot{T}(x,0) = 0, \tag{15}$$

where l is a half of the interval with nonzero temperature.

The solution of this problem is a sum of solutions (12), according to superposition principle. Temperature distribution for a diatomic chain with $\alpha = 0.8$ at several consequent times is shown in Fig. 4. Comparison of the analytical and numerical solutions (see Fig. 5) has also shown quite a good agreement. The deviations in the vicinity of $x \approx 0$ are caused by the residual fast processes, which have not fully decayed [17]. This effect decreases with the increase of the sample size.

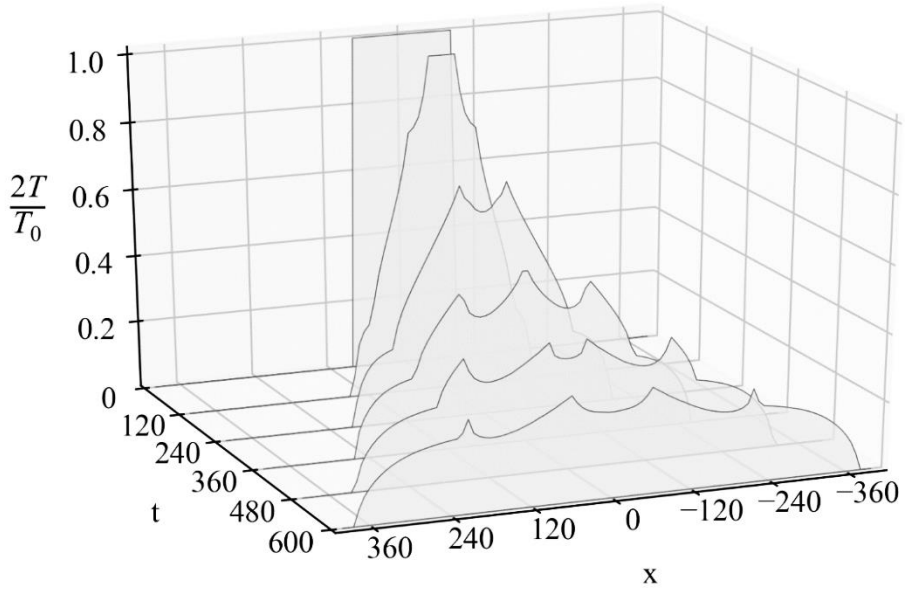

Fig. 4. Evolution of initially rectangular thermal perturbation ($\alpha = 0.8$), analytical solution

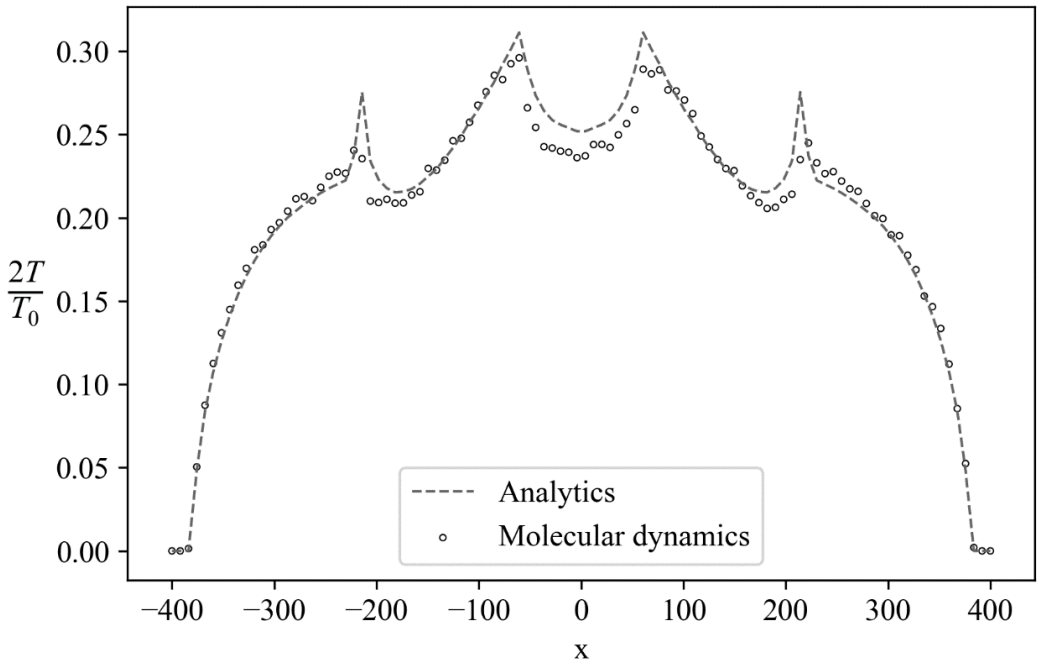

Fig. 5. Comparison of analytical and numerical solutions at t = 600s

According to the classical heat conduction theory, a maximum would be observed at $x = 0$ and it would decay exponentially. In the case of anomalous thermal conductivity, the solution decays faster near zero, forming four fronts that propagate two by two in opposite directions with constant speeds.

Note that in a system with alternating masses or stiffnesses, the thermal front at large times, when the "peaks" become less prominent, looks in a way similar to the solution of a similar problem based on the Fourier thermal conductivity law, whereas the solution for a monatomic chain demonstrates a fundamentally different behavior [13].

If the width of the initial perturbation l decreases and its amplitude T_0 increases, the solution tends to fundamental solution, i.e. solution with perturbation in the form of a delta function. Numerical simulation for $\alpha \neq 0$ and $\alpha \neq 1$ has demonstrated that the thermal fronts stay at a finite distance from each other, and the speed of the first ones ($c_{g_2}^{max}$, acoustic branch) always turns out to be greater than the speed of the second ones ($c_{g_1}^{max}$, optic branch). If one of the masses is negligible in comparison to another, i.e. $\alpha \to 0$, most of the heat is transferred with the speed $c_{g_1}^{max}$, whereas if $\alpha \to 1$ almost all the heat propagates with $c_{g_2}^{max}$. These observations can be further used to test the presented theory in future experiments.

4. Conclusions

In the present work, the method that allows analytical description of heat propagation in harmonic crystals [13-17] is applied to one-dimensional harmonic crystals with alternating masses (Fig. 1). Solution (9) was obtained by analogy with (8) for scalar lattices [16]. Slow motions are considered: the temperature field is a superposition of waves moving with group velocities (6), and has the form of the initial temperature distribution.

Analytical solutions are given for two problems: (i) cold and hot half space contact and (ii) propagation of an initially rectangular thermal perturbation. It is shown, that if the masses differ slightly, all the solutions tend to the profiles for a monoatomic chain.

For the problem (i), it is demonstrated that, for any ratios between the masses, the initial thermal perturbation propagates in the form of two successive thermal fronts having finite speeds and repeating the form of the initial perturbation. The speed of the first, faster front corresponds to the acoustic branch of the dispersion relation (5), and the speed of the second front corresponds to the optical one. In turn, rectangular thermal perturbation (ii) splits into four thermal fronts, which propagate two by two in opposite directions with constant velocities.

Comparison of the analytical results with numerical simulation shows that the presented theory describes the distribution of heat in a diatomic chain with high accuracy. It is demonstrated numerically that if the masses differ slightly, the main part of the initial perturbation propagates at a speed corresponding to the acoustic branch, but the velocities of the fronts corresponding to the optical branch remain finite. The latter means that up to the achievement of exact equality of the masses all fronts exist, and they are located at a finite distance from each other.

The dispersion relation (5) is the same both for a chain with alternating masses and for a chain with alternating stiffnesses, if the ratio of the respective parameters is the same as well. Thus, from the point of view of the heat transfer problem, these systems are equivalent. Note that this is true only when considering the average temperature in the unit cell (7). The propagation of heat waves corresponding to different degrees of freedom will be significantly different for the two systems [17].

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