Temperature gradient and Fourier's law in gradient-mass harmonic systems

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The heat flow and thermal profile in a one-dimensional (1D) harmonic lattice with coordinate-dependent masses have been calculated in the thermodynamic limit. It is shown in the particular example of a 1D harmonic lattice with linearly increasing masses that in standard Langevin conditions of contact, a temperature gradient can form, and Fourier's law can be obeyed.

I. INTRODUCTION

Raising the efficiency of heat dissipation in nanodimensional systems is presently a major problem whose solution will define the potential for further miniaturization of electronic devices. This problem is complicated by the emergence of a host of effects which are specific to heat transport in nanodimensional systems. To cite an example, heat transport is affected noticeably not only by the heat resistance at interfaces, which plays a dominant part in heat transport in nanostructures [1], but also by variations of the pattern of the heat transport itself [2,3], when the phonon mean free path becomes comparable with the sample size. This results in the heat transport becoming anomalous: more specifically, the Fourier law no longer holds; in other words, the heat flux through the system becomes dependent now not on the gradient but on the temperature difference, a phenomenon which has recently been demonstrated experimentally in nanotubes [4,5].

The pursuit of this goal has been a major motivation for studying not only methods that could reduce the heat resistance at interfaces, with recent progress in this direction being reported in Ref. [6], but ways that could lead to development of thermal rectifiers, i.e., to the possibility of varying the modulus of the heat flux by changing the sign of the temperature difference applied to the system [7]. Considerations of a general nature seem to lead to an obvious assumption that in order to observe such a "heat rectification effect," one should produce asymmetry in a system. A theoretical analysis [8,9] and an experimental study of a nonuniformly mass-loaded nanotube [10] have demonstrated that one-dimensional structures with increasing masses are possible candidates for realization of the effect.

Obviously enough, development of such gradient-mass structures is technologically anything but a simple problem [11], and we are witnessing presently only the beginning of this process, with the effect of heat rectification not yet realized in full measure. This is why a search for its realization is being pursued along more than one direction. It has been proposed [12] to use filaments of doped silicon. In place of a spatially varying mass, the idea was also advanced of subjecting a nanotube to nonuniform tension [13], which is obviously equivalent to variation of mass [14,15]. One could apparently employ for this purpose nanodiamond-decorated

carbon nanotubes [16]. The possibility is also being discussed of using asymmetric graphene and silicon structures [17,18].

It appears a plausible assumption that, similar to the *p-n* junction which has become a basis of electronics, development of gradient-mass materials will form a foundation for progress in a new domain—phononics [19]. As a weighty argument for this conclusion there are proposals for a number of nanodevices based on such materials, to wit, heat diodes [20,21], heat transistors [22], heat logical elements [23], memory devices [24], and heat limiters [25].

The above illustrates an increasing interest in studies of the thermal properties of gradient-mass systems. Numerous attempts are being undertaken to investigate such materials by both numerical [8,12,26,27] and analytical methods [9,28]. Many theoretical aspects remain, open, however, even without inclusion of anharmonic effects into consideration. To cite an example, it was shown numerically that a temperature gradient forms in systems with linearly [13] or exponentially [29] varying masses, even in a harmonic case. This phenomenon is surprising in itself. Indeed, despite numerous attempts, a rigorous analytical microscopic foundation of Fourier's law is still lacking [30,31]. Only for several systems has one managed to obtain an analytical result, more specifically, systems with identical [32], alternating [33–36], or random masses [37–39]. It should be stressed, however, that in neither of these cases does the Fourier law hold and a temperature gradient does not form. The temperature profile is linear and Fourier's law holds only in effective models such as harmonic chains with self-consistent stochastic reservoirs at each site [34,40,41].

We are going to show below that gradient-mass systems possess truly unique properties; indeed, by now these are the only systems in which one can obtain a temperature gradient analytically and make the Fourier law hold without self-consistent stochastic reservoirs. Significantly, such functionally graded materials, i.e., nonuniform gradient-mass systems, can be met in nature [42]. Note that the optical properties of systems with graded dielectric permittivity match those of gradient-mass systems [43,44]. And it is these systems that are attracting great current interest [45,46] as effective media for application of the Faraday effect [47].

II. MODEL

Consider a one-dimensional chain of $N + 1$ particles which interact harmonically with their nearest neighbors with a spring constant K. The momentum and displacement n of a particle of mass m_n will be denoted by p_n and x_n, respectively. The
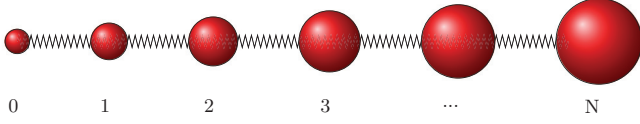
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FIG. 1. (Color online) Schematic of a harmonic system with coordinate-dependent masses.

Hamiltonian of this system (Fig. 1) can be written in the form

$$H = \sum_{n=0}^{N} \left(\frac{p_n^2}{2m_n} + \frac{1}{2}K(x_{n+1} - x_n)^2 \right). \tag{1}$$

The equations of motion for such a system become

$$x_n \cos \beta_n = \frac{1}{2}(x_{n+1} + x_{n-1}), \tag{2}$$

where $\cos \beta_n = (1 - \omega^2 m_n/2K)$.

In the thermodynamic limit the solution of this system of equations can be obtained in analytical form. To do this, we use Fourier's method by writing x_n in the form

$$x_n = \int_{k_1}^{k_2} f(k) \exp(ikn)dk. \tag{3}$$

We choose the limits of integration k_1 and k_2 such that the function $f(k)$ and all its derivatives tend to zero at these points. In this case we come to the equality

$$(-in)^p x_n = \int_{k_1}^{k_2} \frac{d^p f(k)}{dk^p} \exp(ikn)dk.$$

We further assume that the mass m_n and β_n are functions of n/N. Now the function $\cos \beta(n/N)$ can be expanded in a Taylor series with respect to the variable n/N, with Eqs. (2) converting into a differential equation for the function f:

$$\cos \beta \left(i \frac{1}{N} \frac{d}{dk} \right) f = f \cos(k). \tag{4}$$

Only in rare cases, such as a linear dependence of the particle masses on the index,

$$m_n = M_0 + \frac{M_1 - M_0}{N} n, \tag{5}$$

does this differential equation allow an exact solution.

Because we are going to revert to this case more than once, we note that Eqs. (2) are solved in terms of Hankel functions: $x_n = H_{n+\Delta n}^{(1)}(z)$, $H_{n+\Delta n}^{(2)}(z)$, where $\Delta n = z \cos \beta(0)$, $z = N/[\cos \beta(1) - \cos \beta(0)]$. This can be verified by direct substitution using the recurrence relation $2n/z Z_n(z) = Z_{n+1}(z) + Z_{n-1}(z)$, $Z_n(z) = H_n^{(1)}(z)$, $H_n^{(2)}(z)$ [48].

In the general case, we are interested in the solution in the thermodynamic limit. We are going to look for the solution to Eq. (4) subject to the condition $N \to \infty$ in the form

$$f(k) = \varphi(k)e^{ig(k)N}. \tag{}$$

In this case we come to the following relations for g and φ:

$$g = -\int^k \beta^{-1}(z)dz, \tag{6}$$

$$\varphi(\beta(k)) = \exp \left[-\int^k \frac{dx}{M'(x)} \left(\frac{M(x) - M(0)}{x} \right)' \right], \tag{7}$$

where β^{-1} is a function inverse to β. Note that φ does not depend on frequency.

Next we choose the path of integration in Eq. (3) such that it will descend most steeply from the saddle point. Substituting the expressions for g and φ, we come in the limit that $N \to \infty$ to a particular solution to Eqs. (2):

$$y_n = \frac{1}{\sqrt{\sin \beta(a)}} \phi(a)e^{ib\beta(a)}e^{iN \int_0^a \beta(x)dx}, \tag{8}$$

where n is taken in the form $n = aN + b$, and $\phi(k) = \varphi(\beta(k))\sqrt{|M'(k)|}$.

We finally come to the general solution of Eqs. (2) in the form $x_n = Ay_n + By_n^*$.

III. FLUX AND THE TEMPERATURE PROFILE

Using the standard nonequilibrium Green's function method [12,30,49] and assuming the temperature on the left to be $T_L = T + 1/2\Delta T$ and that on the right, $T_R = T - 1/2\Delta T$, with $\Delta T, \omega_n \ll T$, we can obtain both the heat flux J in the system, and the temperature profile T_n:

$$J = \frac{\gamma}{m_N} \Delta T I_N,$$

$$T_n = T + \frac{1}{2}\Delta T(I_n - \bar{I}_n), \tag{9}$$

where the quantities

$$I_n = \frac{m_n \gamma}{\pi} \int_{-\infty}^{\infty} d\omega \omega^2 |G_{0n}|^2,$$

$$\bar{I}_n = \frac{m_n \gamma}{\pi} \int_{-\infty}^{\infty} d\omega \omega^2 |G_{nN}|^2 \tag{10}$$

are expressed in terms of the Green's function:

$$G(\omega) = [K - M\omega^2 - \Sigma_L(\omega) - \Sigma_R(\omega)]^{-1}$$

with M being a diagonal matrix with elements corresponding to the particle masses, and K the dynamical matrix for the system. The function $\Sigma(\omega)$ specifies the conditions of contact of the system under consideration with heat reservoirs. For the standard (Langevin) contact, $\Sigma_L(\omega) = i\omega\gamma\delta_{i,0}\delta_{j,0}$ and $\Sigma_R(\omega) = i\omega\gamma\delta_{i,N}\delta_{j,N}$, where γ is the dissipation constant. After a few straightforward transformations similar to those made in Ref. [50], we come to

$$G_{0,k} = \frac{|D_{k+1,N} - \Sigma D_{k+1,N-1}|^2}{|D_{0,N} - \Sigma(\omega)(D_{1,N} + D_{0,N-1}) + \Sigma(\omega)^2 D_{1,N-1}|^2}, \tag{11}$$

where $D_{l,m}$ is defined to be the determinant of the submatrix of $K - \omega^2 M$ beginning with the lth row and column and ending with the mth row and column. This determinant can be readily derived, because we know the general solution for Eqs. (2):

$$D_{lm} = \frac{\text{Im } y_{l-1} y_{m+1}^*}{\text{Im } y_{l-1} y_l^*}. \tag{12}$$

The solution of the equation $D_{0N} = 0$ with respect to the frequencies ω yields the dependence of the wave vector k on the frequency of vibrations in the system under
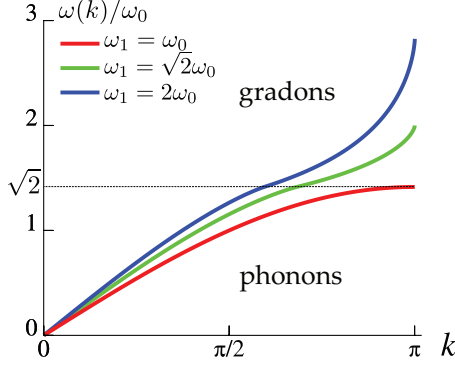
FIG. 2. (Color online) Dispersion relation of the vibration energy in a harmonic chain with linearly increasing masses (5) plotted vs the wave vector for different relations between the boundary masses $\omega_{0,1} = \sqrt{2K/M_{0,1}}$. For energies $\omega < \sqrt{2}\omega_0$, the system operates with delocalized phonons, and for $\sqrt{2}\omega_0 < \omega < \sqrt{2}\omega_1$, with localized gradons.

consideration, $k(\omega)$:

$$\text{Re} \int_0^1 \beta(x)dx = k(\omega). \tag{13}$$

In particular, for the case of linear mass distribution (5) we come to

$$k(\omega) = \text{Re} \frac{f(\beta(0)) - f(\beta(1))}{\Delta_{\cos}},$$

where $f(\beta) = (\sin\beta - \beta\cos\beta)$. This dispersion relation is displayed in Fig. 2. This result correlates with those of numerical simulations [51].

As seen from Fig. 2, in systems with linearly increasing masses one can identify two kinds of vibrations, more specifically, delocalized phonons with frequencies $\omega < \sqrt{2}\omega_0$ and localized "gradons" with frequencies $\sqrt{2}\omega_0 < \omega < \sqrt{2}\omega_1$, $\omega_{0,1} = \sqrt{2K/M_{0,1}}$.

In the general case, there exist also phonons with frequencies $\omega < \sqrt{2}\omega_{\min}$ and gradons with frequencies $\omega > \sqrt{2}\omega_{\min}$, with $\omega_{\min}$ being determined by the maximum mass in the system. Incidentally, for gradons β becomes imaginary, with the determinant starting to grow exponentially. As a result, frequencies $\omega > \sqrt{2}\omega_{\min}$ do not contribute to the integrals in Eqs. (10).

Standard methods of averaging [33] permit reducing the integrands in Eq. (10) in the $N \to \infty$ limit to the forms

$$|G_{0N}|^2 = \left(\frac{\phi(0)}{\phi(1)}\right)^2 \frac{\sin\beta(0)\sin\beta(1)}{\gamma\omega(1 + \gamma^2\omega^2)[\sin\beta(0) + \sin\beta(1)]},$$

$$|G_{0n}|^2 = \frac{1}{2\gamma\omega}\left(\frac{\phi(0)}{\phi(x)}\right)^2 \frac{\sin\beta(0)}{[\sin\beta(0) + \sin\beta(1)]\sin\beta(a)}, \quad (14)$$

$$|G_{nN}|^2 = \frac{1}{2\gamma\omega}\left(\frac{\phi(x)}{\phi(1)}\right)^2 \frac{\sin\beta(1)}{[\sin\beta(0) + \sin\beta(1)]\sin\beta(a)},$$

where n is used in the form $n = aN$. To preclude misunderstanding, we note that these relations do not go one into another at $a = 1$ and $a = 0$, because $N \to \infty$ is a nonuniform limit.

Putting relations (14) into (9), we come to a general answer for the heat flux and temperature profile in a system with an arbitrary mass distribution. While these integrals cannot be performed in the general case, they permit a number of conclusions.

To begin with, it turns out that a flow in a system depends only on the boundary masses and the maximum mass in the chain (which specifies the minimum frequency). We readily see that if the masses are constant, we come to the standard answer for the heat flow [32]. In the reverse approximation, when $M_1 \gg M_0$, we obtain

$$J = \frac{2\sqrt{2}\Delta T}{\pi\omega_0\gamma^2}\left(\frac{\phi(0)}{\phi(1)}\right)^2 [\sqrt{2}\omega_{\min}\gamma - \arctan(\sqrt{2}\omega_{\min}\gamma)].$$

Up to this point, we assumed that the boundary masses do not depend on N. We can include this dependence in the case of linearly increasing masses. Setting $M_1 \sim M_0 N^s$, the heat flux will acquire the form

$$J = \frac{8\Delta T}{3\pi}\omega_0^2\gamma \frac{1}{N^{3s/2}}.$$

It is easy to verify the correctness of the assumptions we have made here, because we know the exact solution for the linear case.

As follows from the expression for the flux J, for a constant gradient $s = 1$ the system behaves as a thermal insulator, $J \sim N^{-3/2}$, and for $s = 0$ the flux does not depend on system size. In the intermediate case of $s = 2/3$, the system conforms to Fourier's law $J \sim N^{-1}$. Note that the dependence of heat flux on the size of a harmonic system is specified by the boundary conditions [6,50], and it is possible to select them such that Fourier's law in the system will be obeyed. In the $s = 2/3$ case we selected the system itself appropriately while leaving unchanged the standard boundary conditions. Thus a system with linearly increasing masses can be adjusted such that it will conform to Fourier's law.

Let us turn now to the temperature profile. First, as seen from the relations (14), if the masses are not constant within the chain, the temperature at the points at which $M'(a) = 0$ should rise strongly.

Second, it can be shown that in the case of constant masses the thermal profile within the system $T(a) = T$ is constant, a point well enough known. In the reverse approximation, in the case of linearly increasing masses, when $M_1 \gg M_0$, we come to

$$T_a = T - \frac{1}{\pi}\Delta T \arcsin(\sqrt{a}).$$

We see immediately that the temperature in the system is determined by the more massive end. Now at the center of the system under consideration a temperature gradient $-\Delta T/\pi$ will appear.

This conclusion correlates well with available numerical simulations [26]. A temperature gradient is built up in the graded harmonic chain, but Fourier's law does not hold because the mass difference does not grow with the system size N, as happens in the present work.

In conclusion, we have presented a solution to the system of linear equations (2) describing systems of functional-gradient materials. In particular, we analyzed the problem of a nonequilibrium steady state of a harmonic variable-mass system connected with heat reservoirs which are maintained at different temperatures.

We have shown that in the particular case of linearly increasing masses the heat flux depends on system size, $J \sim 1/N^{3s/2}$, with the exponent in this relation being determined by the law governing the increase of the boundary mass, $M \sim N^s$. This result finds ready explanation when we turn to Fig. 2. The systems under consideration maintain vibrations of two types, delocalized phonons which transport heat ballistically and localized gradons. By properly varying the boundary mass as a function of N, we modulate in this way the number of phonons, to arrive finally at the dependence of the heat flux J on the system size N. A similar effect accounts for the appearance of a thermal gradient in a system with linearly increasing masses. We have demonstrated in the particular example of a system with linearly increasing masses that a harmonic system can both sustain formation of a temperature gradient and conform to Fourier's law.

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