Sufficient conditions for thermal rectification in general graded materials

Emmanuel Pereira*
Departamento de Física–ICEx, UFMG, CP 702, 30.161-970 Belo Horizonte MG, Brazil
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We address a fundamental problem for the advance of phononics: the search of a feasible thermal diode. We establish sufficient conditions for the existence of thermal rectification in general graded materials. By starting from simple assumptions satisfied by the usual anharmonic models that describe heat conduction in solids, we derive an expression for the rectification. The analytical formula shows how to increase the rectification, and the conditions to avoid its decay with the system size, a problem present in the recurrent model of diodes given by the sequential coupling of two or three different parts. Moreover, for these graded systems, we show that the regimes of nondecaying rectification and of normal conductivity do not overlap. Our results indicate the graded systems as optimal materials for a thermal diode, the basic component of several devices of phononics.

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I. INTRODUCTION

The study of the macroscopic laws of thermodynamic transport from the underlying microscopic models is still a challenge in statistical physics. In particular, the investigation and control of the energy transport, which mainly involves conduction of heat or electricity, is a fundamental problem of huge theoretical and practical interest. The invention of transistor and other devices used to control the electric charge flow has led to the well-known development of modern electronics. Its much less developed counterpart—the study and control of heat current—has, recently, presented interesting progress, promising to establish, in addition to electronics, a new physical branch in energy and information processing—the phononics [1,2]: researchers have proposed nanodevices such as thermal diodes or rectifiers [3–5] (already built in practice [6]), thermal transistors [7], thermal logic gates [8], and memories [9]. The most fundamental component of these instruments is the thermal diode, a device in which heat flows preferably in one direction. In a short analysis, we may say that this promising advance of phononics is directly dependent on the development of its basic component: a thermal diode with suitable properties.

There are analytical attempts to investigate the phenomenon of thermal rectification such as the works on spin-boson junctions [10], billiard systems [11], etc., but most of the results are by means of computer simulations, see, e.g., the work of Li and collaborators [12]. The most common and recurrent design of diodes is given by the sequential coupling of two or more chains with different anharmonic potentials [3–5]. Although frequently studied, this procedure is criticized [5] due to the difficulty to construct such diode in practice, and due to the significative decay of the rectification with the system size. Recently, a different procedure was considered by Chang *et al.* [6], who built, in an experimental work, the first microscopic solid-state thermal rectifier by using a graded material: nanotubes externally and inhomogeneously mass-loaded with heavy molecules. It is worth to recall that graded materials, i.e., inhomogeneous systems whose composition and/or structure change gradually in space, are abundant in nature, can also be manufactured, and have attracted great interest in many areas [13]: there are many works devoted to the study of the electric, optical, mechanical, and other properties of graded materials, but there are few studies in relation to their heat conduction investigation.

In the present work, we address this fundamental problem of phononics: the build of an appropriate thermal diode, namely, a simple system that may be constructed in practice, and with a rectification that does not decay with the system size. We start from simple conditions for the local thermal conductivity, conditions that are quite general and that are satisfied by anharmonic crystal models used to describe heat conduction in solids, and then we show that they are sufficient to lead to rectification in graded models. Moreover, we derive an expression for such rectification that allows us to see how to make it larger, and how to avoid its decay with the system size. In short, we show that properly manipulated graded materials have suitable properties of rectification, and so, they shall play a central role in the building of thermal nanodevices. The simplicity of the initial conditions and of the arguments to establish the results shows the ubiquity of thermal rectification in graded systems. Moreover, the existence of simple ingredients for the rectification, as described here, deserves attention: as well known, in the literature, the mechanism behind rectification in graded models is far from being clear, e.g., we recall the comment of Casati [2] on the explanation of Chang *et al.* [6]: "The authors speculate that solitons might be involved in the rectification process, but this is still to be confirmed." Here, we do not have to make any speculation about the vibrational spectra or other intricate property.

II. EXISTENCE OF THERMAL RECTIFICATION

Let us introduce our assumptions and derive our results.

We consider a chain with N sites, where the first site is connected to a thermal bath at temperature T_1, and the last site is connected to a bath at temperature T_N. It is possible to extend our analysis also for a d-dimensional lattice with two thermal baths at the boundaries: the chain structure is represented by the axis (direction) of the heat flow. We assume that it is possible to build a temperature gradient in the system.

*emmanuel@fisica.ufmg.br

Such condition always happens if Fourier's law holds, but we do not demand this law here (anyway, we will study cases where Fourier's law holds). Precisely, we assume that the heat flow from site j to $j+1$ is given by

$$\mathcal{F}_{j,j+1} = -\mathcal{K}_j(\nabla T)_j = \frac{1}{\mathcal{C}_j T_j^\alpha + \mathcal{C}_{j+1} T_{j+1}^\alpha}(T_j - T_{j+1}), \quad (1)$$

i.e., with, say, the local thermal conductivity given by the average of a function of the local temperatures and other parameters of the system. For the homogeneous model, such expression reads

$$\mathcal{F}^H_{j,j+1} = \frac{1}{\mathcal{C}\left(T_j^\alpha + T_{j+1}^\alpha\right)}(T_j - T_{j+1}) = \frac{1}{\mathcal{C}'\bar{T}_j^\alpha}(T_j - T_{j+1}),$$

where $\bar{T}_j^\alpha = (T_j^\alpha + T_{j+1}^\alpha)/2$, $\mathcal{C}' = 2\mathcal{C}$, which is exactly the formula described by several results (on homogeneous models) from the literature, e.g., in Ref. [14], we have $\alpha = 2$, $\mathcal{C}'T^2 = 1/\mathcal{K} = \lambda^2 T^2/\omega^9\mu^3$, where λ is the coefficient of the quartic anharmonic potential, ω is the coefficient of the interparticle quadratic interaction, and μ is the harmonic pinning coefficient. Still for this ϕ^4 model, in different conditions and methods, Bricmont and Kupiainen [15] and Spohn *et al.* [16] found $\mathcal{K}_j = T_j^{-2}$. And, in reference to works with detailed computer simulations, Aoki and Kusnezov [17] obtain for this one-dimensional ϕ^4 model, $\mathcal{K} \propto T^{-1.35}$; similarly, N. Li and B. Li [18] obtain $\mathcal{K} \propto T^{-1.5}$, with slight changes in the exponent that depend on the values of the pinning and anharmonicity. It is also worth to recall that, by using an analytical simplified scheme (derived from a rigorous and more intricate approach [19]), we obtain a similar formula for the local thermal conductivity of the graded anharmonic self-consistent chain [20], i.e., of the anharmonic, inhomogeneous model given by a chain of oscillators with quartic on-site potential, quadratic nearest-neighbor interparticle interaction, particles with different masses and inner stochastic reservoirs connected to each site.

Let us now prove the existence of thermal rectification for a graded anharmonic system with a temperature gradient in the bulk, and whose local thermal conductivity depends on temperature (which does not follow in the harmonic case), and changes as we run the chain.

From the fact that the heat current comes into the system by the first site, passes trough the chain and goes out by the last site, we have

$$\mathcal{F}_{1,2} = \mathcal{F}_{2,3} = \cdots = \mathcal{F}_{N-1,N} \equiv \mathcal{F}. \quad (2)$$

These equations together with Eq. (1) give us

$$\mathcal{F}\left(\mathcal{C}_1 T_1^\alpha + \mathcal{C}_2 T_2^\alpha\right) = T_1 - T_2,$$
$$\mathcal{F}\left(\mathcal{C}_2 T_2^\alpha + \mathcal{C}_3 T_3^\alpha\right) = T_2 - T_3,$$
$$\cdots = \cdots,$$
$$\mathcal{F}\left(\mathcal{C}_{N-1} T_{N-1}^\alpha + \mathcal{C}_N T_N^\alpha\right) = T_{N-1} - T_N.$$

Summing up the equations, we find

$$\mathcal{F} = \mathcal{K}\frac{(T_1 - T_N)}{N-1},$$

where

$$\mathcal{K} = \left\{\mathcal{C}_1 T_1^\alpha + 2\mathcal{C}_2 T_2^\alpha + \cdots + 2\mathcal{C}_{N-1} T_{N-1}^\alpha + \mathcal{C}_N T_N^\alpha\right\}^{-1}(N-1), \quad (3)$$

that is Fourier's law for the case of the thermal conductivity $\mathcal{K}$ remaining finite as $N \to \infty$. From Eqs. (1) and (2), it follows that

$$\frac{T_1 - T_2}{\mathcal{C}_1 T_1^\alpha + \mathcal{C}_2 T_2^\alpha} = \frac{T_2 - T_3}{\mathcal{C}_2 T_2^\alpha + \mathcal{C}_3 T_3^\alpha} = \cdots = \frac{T_{N-1} - T_N}{\mathcal{C}_{N-1} T_{N-1}^\alpha + \mathcal{C}_N T_N^\alpha}. \quad (4)$$

Thus, given the temperatures of the baths T_1 and T_N, by using the equations above we determine the inner temperatures T_2, $T_3, \ldots, T_{N-1}$. For ease of computation, let us consider the system submitted to a small gradient of temperature: $T_1 = T + a_1\epsilon$, $T_N = T + a_N\epsilon$, ϵ small. Hence, $T_k = T + a_k\epsilon + \mathcal{O}(\epsilon^2)$. We will carry out the computations only up to $\mathcal{O}(\epsilon)$. And so, up to $\mathcal{O}(\epsilon)$, we have $T_k^\alpha = T^\alpha + \alpha T^{\alpha-1}\epsilon a_k$ (that comes from the Taylor series), and

$$\frac{T_k - T_{k+1}}{\mathcal{C}_k T_k^\alpha + \mathcal{C}_{k+1} T_{k+1}^\alpha} = \frac{(a_k - a_{k+1})\epsilon}{(\mathcal{C}_k + \mathcal{C}_{k+1})T^\alpha},$$

as said, up to $\mathcal{O}(\epsilon)$. From this equation and Eq. (4), we obtain

$$\frac{a_1 - a_2}{\mathcal{C}_1 + \mathcal{C}_2} = \frac{a_2 - a_3}{\mathcal{C}_2 + \mathcal{C}_3} = \cdots = \frac{a_{N-1} - a_N}{\mathcal{C}_{N-1} + \mathcal{C}_N}. \quad (5)$$

We may rewrite these equations as

$$\frac{a_1 - a_2}{\mathcal{C}_1 + \mathcal{C}_2} = \frac{a_1 - a_2}{\mathcal{C}_1 + \mathcal{C}_2},$$
$$\frac{a_1 - a_2}{\mathcal{C}_1 + \mathcal{C}_2} = \frac{a_2 - a_3}{\mathcal{C}_2 + \mathcal{C}_3},$$
$$\cdots = \cdots,$$
$$\frac{a_1 - a_2}{\mathcal{C}_1 + \mathcal{C}_2} = \frac{a_{N-1} - a_N}{\mathcal{C}_{N-1} + \mathcal{C}_N}.$$

Summing them up, we obtain

$$\frac{a_1 - a_2}{\mathcal{C}_1 + \mathcal{C}_2}(\mathcal{C}_1 + 2\mathcal{C}_2 + \cdots + 2\mathcal{C}_{N-1} + \mathcal{C}_N) = a_1 - a_N$$
$$\Rightarrow a_2 = a_1 + \frac{(a_N - a_1)}{\tilde{\mathcal{C}}(N)}(\mathcal{C}_1 + \mathcal{C}_2),$$

where $\tilde{\mathcal{C}}(N) \equiv (\mathcal{C}_1 + 2\mathcal{C}_2 + \cdots + 2\mathcal{C}_{N-1} + \mathcal{C}_N)$. Similarly, writing $(a_{k-1} - a_k)/(\mathcal{C}_{k-1} + \mathcal{C}_k)$ instead of $(a_1 - a_2)/(\mathcal{C}_1 + \mathcal{C}_2)$ in the left-hand side of the list of equations above, we obtain

$$a_k = a_1 + \frac{(a_N - a_1)}{\tilde{\mathcal{C}}(N)}\tilde{\mathcal{C}}(k), \quad (6)$$

for $k = 2, \ldots, N-1$. And so, for the thermal conductivity (3) it follows that

$$\mathcal{K} = (N-1)\{T^\alpha\tilde{\mathcal{C}}(N) + \alpha T^{\alpha-1}\epsilon(a_1\mathcal{C}_1 + 2a_2\mathcal{C}_2 + \cdots + 2a_{N-1}\mathcal{C}_{N-1} + a_N\mathcal{C}_N)\}^{-1}. \quad (7)$$

To investigate the existence or absence of rectification, we need to analyze the heat flow for the system with inverted thermal baths, that is, we compute the new thermal conductivity for the same system, but with temperatures T', where $T'_1 = T_N$ and $T'_N = T_1$. Following the previous manipulations, we see that, in the system with inverted baths, the new temperature for the site k is $T'_k = T + a'_k\epsilon$, where, for $k = 2,3,\dots,N-1$,

$$a'_k = a_N - \frac{(a_N - a_1)}{\tilde{\mathcal{C}}(N)}\tilde{\mathcal{C}}(k). \tag{8}$$

Obviously $a'_1 = a_N$, and $a'_N = a_1$. Hence, the expression for the "inverted" thermal conductivity becomes

$$\mathcal{K}' = (N-1)\{T^\alpha\tilde{\mathcal{C}}(N) + \alpha T^{\alpha-1}\epsilon(a'_1\mathcal{C}_1 + 2a'_2\mathcal{C}_2 + \cdots + 2a'_{N-1}\mathcal{C}_{N-1} + a'_N\mathcal{C}_N)\}^{-1}. \tag{9}$$

And, with simple manipulations, we get

$$\frac{1}{\mathcal{K}} - \frac{1}{\mathcal{K}'} = \frac{\alpha T^{\alpha-1}\epsilon(a_1 - a_N)}{(N-1)\tilde{\mathcal{C}}(N)} \times\{\tilde{\mathcal{C}}(N)^2 - 4\tilde{\mathcal{Q}}(N) - 2\mathcal{C}_N\tilde{\mathcal{C}}(N)\}, \tag{10}$$

where $\tilde{\mathcal{Q}}(N) \equiv \tilde{\mathcal{C}}(2)\mathcal{C}_2 + \cdots + \tilde{\mathcal{C}}(N-1)\mathcal{C}_{N-1}$. As a simple test for the expression above, note that it vanishes (as expected) in the case of a homogeneous system ($\mathcal{C}_1 = \mathcal{C}_2 = \cdots = \mathcal{C}_N$).

To continue the analysis, we take a chain with three sites (say, the smallest possible system). A direct computation gives us

$$[\tilde{\mathcal{C}}(3)]^2 - 4\tilde{\mathcal{Q}}(3) - 2\mathcal{C}_3\tilde{\mathcal{C}}(3) = \mathcal{C}_1^2 - \mathcal{C}_3^2.$$

Now we prove, by induction, that such relation is valid for any number of sites: we assume that it is valid for k sites (i.e., for k replacing 3 in the relation above), and then we show that it follows for $k+1$. In fact, by using the definitions we see that

$$\tilde{\mathcal{C}}(k+1) = \mathcal{C}_1 + 2\mathcal{C}_2 + \cdots + 2\mathcal{C}_k + \mathcal{C}_{k+1} = \tilde{\mathcal{C}}(k) + \mathcal{C}_k + \mathcal{C}_{k+1},$$

$$\tilde{\mathcal{Q}}(k+1) = \tilde{\mathcal{C}}(2)\mathcal{C}_2 + \cdots + \tilde{\mathcal{C}}(k)\mathcal{C}_k = \tilde{\mathcal{Q}}(k) + \tilde{\mathcal{C}}(k)\mathcal{C}_k.$$

Then, a direct computation shows that

$$[\tilde{\mathcal{C}}(K+1)]^2 - 4\tilde{\mathcal{Q}}(k+1) - 2\mathcal{C}_{k+1}\tilde{\mathcal{C}}(k+1) = \mathcal{C}_1^2 - \mathcal{C}_{k+1}^2.$$

Hence, for the difference between the thermal conductivities of the system with N sites, we obtain

$$\frac{1}{\mathcal{K}} - \frac{1}{\mathcal{K}'} = \frac{\alpha T^{\alpha-1}\epsilon(a_1 - a_N)}{(N-1)\tilde{\mathcal{C}}(N)}\left[\mathcal{C}_1^2 - \mathcal{C}_N^2\right], \tag{11}$$

where, we recall, $\alpha T^{\alpha-1}\epsilon(a_1 - a_N)$ in the numerator above is $T_1^\alpha - T_N^\alpha$ up to $\mathcal{O}(\epsilon)$. Thus, the existence of thermal rectification for anisotropic, e.g., graded, materials is transparent.

III. RECTIFICATION PROPERTIES

Now, let us examine the rectification in details and search for conditions leading to suitable properties. First, we write the expression for the rectification factor f_r

$$f_r \equiv \frac{|\mathcal{K} - \mathcal{K}'|}{\mathcal{K}'} \approx \frac{|T_1^\alpha - T_N^\alpha|}{T^\alpha}\frac{|\mathcal{C}_1^2 - \mathcal{C}_N^2|}{[\tilde{\mathcal{C}}(N)]^2}.$$

Hence, fixed the temperatures at the boundaries, the behavior of f_r with N is given by $|\mathcal{C}_1^2 - \mathcal{C}_N^2|/[\tilde{\mathcal{C}}(N)]^2$. We recall that

$$\tilde{\mathcal{C}}(N) = \mathcal{C}_1 + 2\mathcal{C}_2 + \cdots + 2\mathcal{C}_{N-1} + \mathcal{C}_N \approx 2\int_1^N \mathcal{C}_x dx.$$

And, for a small gradient of temperature in the system,

$$\mathcal{K} = (N-1)/\{T^\alpha\tilde{\mathcal{C}}(N) + \mathcal{O}(\epsilon)\}.$$

Thus, to get a normal conductivity (Fourier's law) we must have $\tilde{\mathcal{C}}(N) \sim N$, i.e., $\mathcal{C}_N \sim$ constant. That is, for these graded systems, at least at small temperature gradients, if the conductivity is normal then the rectification factor decays to zero as $N \to \infty$. To avoid the decay of the rectification factor, for example, to make it finite and nonzero as $N \to \infty$, we need to take $\mathcal{C}_N \sim c\exp(\gamma N)$. And so, $\tilde{\mathcal{C}}(N) \sim c[\exp(\gamma N) - 1]/\gamma$. For $\gamma > 0$, $\tilde{\mathcal{C}}(N)$ has exponential growth and $\mathcal{K}(N) \to 0$ as $N \to \infty$. For $\gamma < 0$, $\tilde{\mathcal{C}}(N) \to$ constant and $\mathcal{K} \sim N$, i.e., we have an abnormal conductivity. That is, the regimes of nondecaying rectification and of normal conductivity do not overlap. The possibility of a nondecaying rectification is a very important property: as recalled before, the decay of rectification is a problem for the usual diodes given by the sequential coupling of different parts.

Moreover, still from the previous expression (take $T_1 > T_N$ and $\mathcal{C}_N > \mathcal{C}_1$), we see that the thermal conductivity is smaller when the heat flows from the sites with larger $\mathcal{C}$ to the sites with smaller $\mathcal{C}$.

In short, we have shown that in a lattice system where it is possible to build a temperature gradient, i.e., with the heat flow from site j to $j+1$ given by Eq. (1), with graded structure (i.e., graded $\mathcal{C}_j$) and with local thermal conductivity dependent on temperature [see Eq. (1)], we will always have thermal rectification. To be precise, we need to recall that in our proof, for ease of computation, we have assumed a system with small temperature gradient (however, we believe that it is not a necessary condition—more comments ahead). It is interesting to note that such conditions– temperature gradient in the bulk, local conductivity dependent on temperature, and a graded structure—appear in the quantum harmonic self-consistent chain of oscillators [21], a system that presents rectification in opposition to its classical version (with a conductivity that does not depend on temperature).

To give a concrete example, we turn to the chain with homogeneous anharmonic potential, homogeneous interparticle interactions, etc., but with graded masses. For the model with inner self-consistent reservoirs, weak nearest-neighbor interactions, quartic anharmonicity, in an approximate calculation [20], we have

$$\mathcal{F}_{j,j+1} = \frac{C}{\left(m_{j+1}T_j^{1/2} + m_jT_{j+1}^{1/2}\right)}(T_j - T_{j+1}),$$

where C involves the coefficients for the anharmonicity, interparticle interaction, etc. The denominator of the expression above may be written as $[(T_j^{1/2}/\rho_{j,j+1}) + (T_{j+1}^{1/2}/\rho_{j+1,j})]$, where $\rho_{j,j+1} = m_j/m_{j+1}m_j$, $\rho_{j+1,j} = m_{j+1}/m_{j+1}m_j$. To follow, we define

$$\bar{\rho}_j \equiv \frac{\rho_{j,j-1} + \rho_{j,j+1}}{2} = \frac{1}{2}\frac{(m_{j+1} + m_{j-1})}{(m_{j-1}m_{j+1})},$$

i.e., $\bar{\rho}_j$ is proportional to the inverse of a reduced mass. Hence, considering the entire system $j = 1,\dots,N$, we approximately have $\mathcal{F}_{j,j+1}$ given by Eq. (1) with $\mathcal{C}_j = 1/\bar{\rho}_jC$. And the analysis follows as previously described: now with the bigger flow in the direction from the larger to the smaller masses. It is worth to recall that such property, a bigger heat flow from the larger to smaller densities, as described here, has been already experimentally described [6].

Similar properties appear in a system with homogeneous particle masses, but graded anharmonic on-site potentials or graded interparticle interactions.

IV. FINAL REMARKS

We have some remarks. First, we stress that we have presented here sufficient, not necessary, conditions for manifesting thermal rectification in anisotropic systems. In Ref. [22], by computer simulations, the authors describe rectification in a graded mass Fermi-Pasta-Ulam chain, a model with an invariant translational potential and abnormal conductivity (even for the case of homogenous masses). We also recall that, for the (very different) case of a system of two-terminal junctions, sufficient conditions for rectification have been described in a recent work by Wu and Segal [23].

A further investigation of great interest is the behavior of the graded system as submitted to a large gradient of temperature: we believe that it shall lead to a significative rectification. In Ref. [24], for some specific models given by chaotic billiard systems, the authors claim that there is a significative rectification “provided the temperatures (of the two sides of the system) are strongly different... .”

To conclude, we emphasize that due to their simplicity, the assumptions and arguments described here follow for many of the usual systems modeling heat conduction in solids: it shows the ubiquity of rectification in graded systems. Moreover, the existence of simple conditions for the existence of an efficient rectification, and the fact that graded systems may be constructed in practice (and are even abundant in nature) indicate that they are optimal material to be used in the construction of a thermal diode (and also thermal transistors, etc.), and so, their use shall certainly contribute to the advance of phononics.

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