

Predicting the phonon spectra of coupled nonlinear chains using effective phonon theory

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Abstract

In general one-dimensional nonlinear lattices, extensive studies have discovered the existence of renormalized phonons due to nonlinear interactions and found these renormalized phonons, as the energy carriers, are responsible for heat transport. Within the framework of renormalized phonons, a generic form of renormalized phonon spectrum has been derived and effective phonon theory (EPT) has been developed to explain the heat transport in general 1D nonlinear lattices. Our attention is dedicated to generalizing the EPT for two-layer nonlinear lattices and deriving the analytic expression of phonon spectra. By calculating the phonon spectra of different coupled models with EPT, it is found that the phonon dispersion relation is in good agreement with the result obtained from the spectral energy density method. It is demonstrated that the EPT of a coupled system can predict the phonon spectra of two-layer nonlinear lattices well. Thus, this finding may shed light on the prediction of heat conduction behavior in a coupled system, qualitatively, and provide a useful guide for designing thermal devices.

Keywords: renormalized phonons, effective phonon theory, phonon spectra, heat transport, nonlinear lattices, spectral energy density

(Some figures may appear in colour only in the online journal)

1. Introduction

The search for novel structures and materials with the desired thermal properties, especially thermal conductivity, has been attracting increasing attention in recent years because it is important for the development of nanostructured microelectronic and optoelectronic devices [1–20]. To better guide the designs and applications, it is vital to develop a fundamental understanding of the physics of carrier transport. In non-metallic and semiconductor structures, the phonon is the dominant heat carrier. Thus, an exact knowledge of phonon spectra is needed before investigating the thermal conductivity of these structures.

Many methods have been proposed and applied to predict the phonon properties in the past few decades. It has been reported that using molecular dynamics (MD) simulation and normal mode analysis can predict the phonon spectra [21–24], but this method requires *a priori* knowledge of the phonon frequencies and polarization vectors and is time-intensive. Spectral energy density (SED) analysis is another version of normal mode analysis which is implemented in the frequency domain and it has recently been employed to predict the phonon properties [25–33]. Using this method, we can get an exact knowledge of phonon spectra, but it is always a tedious process to fit every phonon mode, especially for long nonlinear chains. In addition, it is difficult to avoid the uncertainty of the phonon lifetime [31]. Recently, based on generalized equipartition theorem, Li *et al* proposed the so-called effective phonon theory (EPT) which opens up a new way to obtain knowledge of phonon spectra for general 1D nonlinear lattices [34–39]. The analytic expression of phonon spectra by EPT makes it possible to predict the heat conduction behavior qualitatively. The formula for heat conductivity based on effective phonon theory has been applied to explain the anomalous and normal heat conduction in lattices with or without on-site potential very well [34, 35]. Moreover, the size-dependent and temperature-dependent thermal conductivities of 1D nonlinear lattices have also been successfully explained by effective phonon theory [36–39]. Therefore, using EPT we can gain a better understanding of the heat conduction in 1D nonlinear lattices. However, EPT is only applicable to isolated 1D nonlinear lattices. With the increasing emergence of assembled nanomaterial in real applications, research on the thermal conductivity of coupled systems become more important [40–42], so it is necessary to develop the EPT to extend to two-layer lattices.

In this paper, we generalize the EPT to coupled nonlinear lattices and an analytic expression of phonon spectra is derived based on generalized equipartition theorem. To confirm the theory, the SED method is employed to verify the EPT calculations. It is found that the phonon dispersion relation obtained by these two methods agrees well with each other.

The rest of the paper is organized as follows. In section 2, we describe the coupled nonlinear chains model and derive the renormalized phonon spectrum for general coupled 1D nonlinear lattices. Section 3 is devoted to verifying the EPT of a coupled system by comparing the phonon dispersion derived from EPT with the prediction by SED method. Then, we conclude our paper in section 4.

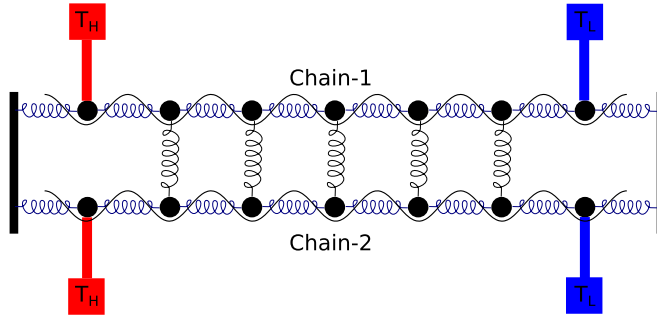


Figure 1. Schematics of the coupled nonlinear chains.

2. Coupled nonlinear lattices and effective phonon theory

In the present paper, we consider two identical coupled 1D anharmonic (nonlinear) lattices (see figure 1) with the Hamiltonian

$$H_n = \sum_{j=1}^N \left[\frac{p_n^j{}^2}{2m_n} + V_n(x_n^j - x_n^{j+1}) + U_n(x_n^j) \right],$$

for $n = 1, 2$ (1)

$$H_c = \sum_{j=2}^{j=N-1} V_{12}(x_1^j - x_2^j). \quad (2)$$

Here, H_1 , H_2 and H_c denote the Hamiltonians of chain 1, chain 2 and the coupling term, respectively. x_n^j is the displacement from the equilibrium position of the j th particle in n th chain; p_n^j is the momentum of this particle. N is the total number of particles and m_n is the mass of the particles in each chain. Here, we set all masses $m_n = 1$ for each lattice. Without loss of generality, we write the interparticle potential $V_n(x_n^j - x_n^{j+1})$, the on-site potential $U_n(x_n^j)$, and the interchain interaction $V_{12}(x_1^j - x_2^j)$ as

$$V_n(x_n^j - x_n^{j+1}) = \sum_{s=2}^{\infty} g_s \frac{(x_n^j - x_n^{j+1})^s}{s}, \quad (3)$$

$$U_n(x_n^j) = \sum_{s=2}^{\infty} \sigma_s \frac{x_n^j{}^s}{s}, \quad (4)$$

$$V_{12}(x_1^j - x_2^j) = \sum_{s=2}^{\infty} l_s \frac{(x_1^j - x_2^j)^s}{s}. \quad (5)$$

By diagonalizing the harmonic Hamiltonian of chain n , the position space can be transformed into normal mode space as follows:

$$\begin{bmatrix} x_n^j \\ p_n^j \end{bmatrix} = B_n \begin{bmatrix} q_n^k \\ p_n^k \end{bmatrix},$$

where $B_1^{k,j} = B_2^{k,j} = B_{k,j} = 1/\sqrt{N} \left(\sin \frac{2\pi kj}{N} + \cos \frac{2\pi kj}{N} \right)$ ($i, k = 1, \dots, N$). For a single free-standing harmonic lattice, its phonon spectrum is $\omega_k = 2 \sin k/2$. Here, k is the wave vector with replacement $k \rightarrow 2k\pi/N$. Under ergodic hypothesis, the coupled lattices obey the generalized equipartition theorem

$$k_B T = \left\langle p_n^k \frac{\partial H}{\partial p_n^k} \right\rangle = \left\langle q_n^k \frac{\partial H}{\partial q_n^k} \right\rangle. \quad (6)$$

The force of each lattice in k space can be written in the form

$$-F_n^k = \frac{\partial H}{\partial q_n^k} = \sum_{j=1}^N \sum_{s=2}^{\infty} [\omega_k g_s (x_n^j - x_n^{j+1})^{s-1} \gamma_{j,k} + B_{j,k} \sigma_s x_n^{js-1}] + \xi_n^k, \quad (7)$$

where

$$\xi_1^k = \sum_{j=2}^{N-1} \sum_{s=2}^{\infty} B_{j,k} l_s (x_1^j - x_2^j)^{s-1}, \quad (8)$$

$$\xi_2^k = - \sum_{j=2}^{N-1} \sum_{s=2}^{\infty} B_{j,k} l_s (x_1^j - x_2^j)^{s-1}. \quad (9)$$

We define the transformation matrix $\gamma_{j,k}$ in equation (7) as $\gamma_{j,k} = 1/\omega_k (B_{j,k} - B_{j+1,k})$, which satisfies $\sum_{j=1}^N (x_n^j - x_n^{j+1}) \gamma_{j,k} = \omega_k q_n^k$ [43].

By substituting equation (7) into equation (6) and using the property of transformation matrix $\gamma_{j,k}$, the generalized equipartition theorem becomes

$$\begin{aligned} k_B T &= \left\langle q_n^k \frac{\partial H}{\partial q_n^k} \right\rangle \\ &\approx \sum_{s=2}^{\infty} \left[g_s \frac{\left\langle \sum_{j=1}^N (x_n^j - x_n^{j+1})^s \right\rangle}{\left\langle \sum_{j=1}^N (x_n^j - x_n^{j+1})^2 \right\rangle} \omega_k^2 + \sigma_s \frac{\left\langle \sum_{j=1}^N x_n^{js} \right\rangle}{\left\langle \sum_{j=1}^N x_n^{j2} \right\rangle} \right. \\ &\quad \left. \pm l_s \frac{\left\langle \sum_{j=2}^{N-1} (x_1^j - x_2^j)^s \right\rangle}{\left\langle \sum_{j=2}^{N-1} (x_1^j - x_2^j) x_n^j \right\rangle} \right] \langle q_n^{k2} \rangle \\ &\equiv \alpha_n (\omega_k^2 + \gamma_n) \langle q_n^{k2} \rangle. \end{aligned} \quad (10)$$

For the third item arising from the interchain interactions in the above expression, we take ‘+’ for $n = 1$ (lattice 1) and ‘-’ for $n = 2$ (lattice 2). Additionally, the two renormalized coefficients α_n and γ_n are defined as follows:

$$\alpha_n = \frac{\sum_{s=2}^{\infty} g_s \left\langle \sum_{j=1}^N (x_n^j - x_n^{j+1})^s \right\rangle}{\left\langle \sum_{j=1}^N (x_n^j - x_n^{j+1})^2 \right\rangle}, \quad (11)$$

$$\begin{aligned} \gamma_n &= \frac{1}{\alpha_n} \left[\frac{\sum_{s=2}^{\infty} \sigma_s \left\langle \sum_{j=1}^N x_n^{js} \right\rangle}{\left\langle \sum_{j=1}^N x_n^{j2} \right\rangle} \right. \\ &\quad \left. \pm \frac{\sum_{s=2}^{\infty} l_s \left\langle \sum_{j=2}^{N-1} (x_1^j - x_2^j)^s \right\rangle}{\left\langle \sum_{j=2}^{N-1} (x_1^j - x_2^j) x_n^j \right\rangle} \right]. \end{aligned} \quad (12)$$

Comparing equation (10) with $k_B T = \omega_k^2 q_k^2$ of a harmonic lattice, an analytic expression of the renormalized phonon spectrum can be derived for a general 1D coupled system with or without on-site potential. It can be identified through the contribution of differences in the displacements of two chains for which the above renormalized phonon spectrum gives the optical phonon branch

$$\omega_{n,k,O}^2 = \alpha_n(\omega_k^2 + \gamma_{n,O}), \quad (13)$$

where $\gamma_{n,O} = \gamma_n$. The acoustic phonon branch for the coupled system should just be the phonon dispersion relation for the corresponding single free-standing chain, which can be given as

$$\omega_{n,k,A}^2 = \alpha_n(\omega_k^2 + \gamma_{n,A}), \quad (14)$$

where

$$\gamma_{n,A} = \frac{1}{\alpha_n} \left[\frac{\sum_{s=2}^{\infty} \sigma_s \langle \sum_{j=1}^N x_n^{js} \rangle}{\langle \sum_{j=1}^N x_n^{j2} \rangle} \right]. \quad (15)$$

3. Comparison between EPT and the SED method

In order to confirm the above theory, we employ the SED [25–31] method to verify the EPT calculations. The SED method can predict fully anharmonic phonon properties using atomic velocities obtained from the MD simulations and its expression reads

$$\Phi_n(\omega, q) = \frac{m_n}{4\pi\tau_0 N} \times \left| \int_0^{\tau_0} \sum_{j=1}^N p_n^j \times \exp\left[i2\pi\frac{j}{N}q - i\omega t\right] dt \right|^2, \quad (16)$$

Here, $\Phi_n(\omega, q)$ is the function of wave vector k ($k = 2\pi q/N$, $q = -\frac{N}{2}, -\frac{N}{2} + 1, \dots, \frac{N}{2} - 1, \frac{N}{2}$) and frequency ω . i is the imaginary unit and τ_0 is the total simulation time. In the following study we consider both cases with harmonic interface interactions and the cases with nonlinear interface couplings for the different models, such as the FPU model [44–46] (without on-site potential), the FK model [45, 47, 48] and the ϕ^4 model [45, 49–51] (with on-site potential).

3.1 The coupled FPU model

In the coupled FPU model, the lattice with Hamiltonian $V_n(x_n^j - x_n^{j+1}) = \frac{g_2}{2}(x_n^j - x_n^{j+1})^2 + \frac{g_4}{4}(x_n^j - x_n^{j+1})^4$ (here $g_2 = 1.0$ and $g_4 = 0.5$), and $V_{12}(x_1^j - x_2^j) = \frac{l_2}{2}(x_1^j - x_2^j)^2 + \frac{l_4}{4}(x_1^j - x_2^j)^4$ is considered, the coefficients α_n , $\gamma_{n,A}$ and $\gamma_{n,O}$ can be derived from equations (11), (12) and (15)

$$\alpha_n = 1.0 + \frac{0.5 \langle \sum_{j=1}^N (x_n^j - x_n^{j+1})^4 \rangle}{\langle \sum_{j=1}^N (x_n^j - x_n^{j+1})^2 \rangle}, \quad (17)$$

$$\gamma_{n,A} = 0, \quad (18)$$

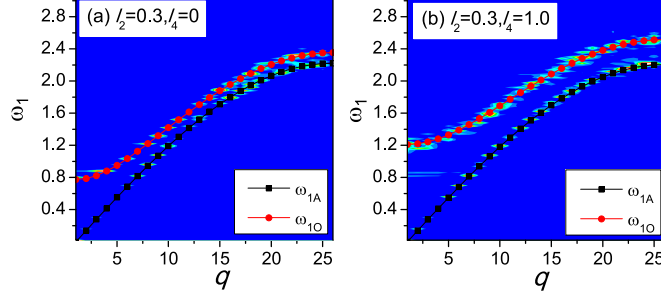


Figure 2. The phonon dispersion relation of chain 1 with different interfacial parameters for coupled FPU chains: (a) $l_2 = 0.3, l_4 = 0$; and (b) $l_2 = 0.3, l_4 = 1.0$. The contour plots are the results by SED method and the red and black dotted lines are results of EPT. ω_{1A} and ω_{1O} denote the acoustic phonon branch and optical phonon branch of chain 1, respectively.

$$\gamma_{n,O} = \pm \frac{1}{\alpha_n} \left[\frac{l_2 \left\langle \sum_{j=2}^{N-1} (x_1^j - x_2^j)^2 \right\rangle}{\left\langle \sum_{j=2}^{N-1} (x_1^j - x_2^j) x_n^j \right\rangle} + \frac{l_4 \left\langle \sum_{j=2}^{N-1} (x_1^j - x_2^j)^4 \right\rangle}{\left\langle \sum_{j=2}^{N-1} (x_1^j - x_2^j) x_n^j \right\rangle} \right]. \quad (19)$$

$N = 50$ and $T = 0.25$ in our MD simulations. In figure 2, we show the dispersion relation of coupled FPU chains derived from effective phonon theory by the dotted lines. It is clear that there are two phonon branches, i.e., the acoustic phonon branch ω_A and optical phonon branch ω_O . For the optical branch, The bundled j th particle pair moves in the opposite directions. In contrast, the two bundled particles (with the same subscript j) move together with the same phase for the acoustic branch, i.e., they behaves in the manner of the center of mass motion, thus the acoustic dispersion relation is exactly the same with the case of the corresponding isolated chain [30]. In figure 2 the contour plots denote the SED patterns and it can be clearly observed that the phonon spectra by EPT are in good agreement with the predictions by SED.

3.2 The coupled FK model

In the coupled FK model $V_n(x_n^j - x_n^{j+1}) = \frac{g_2}{2}(x_n^j - x_n^{j+1})^2$, $U_n(x_n^j) = -\frac{V_{\text{sub},n}}{2\pi^2} \cos(2\pi x_n^j)$, and $V_{12}(x_1^j - x_2^j) = \frac{l_2}{2}(x_1^j - x_2^j)^2 + \frac{l_4}{4}(x_1^j - x_2^j)^4$. Here, we set $g_2 = V_{\text{sub},n} = 1.0$. At the low-temperature regime, the on-site potential $U_n(x_n^j)$ can be expanded in a Taylor series. The renormalization coefficients α_n , $\gamma_{n,A}$ and $\gamma_{n,O}$ are defined as follows:

$$\alpha_n = g_2 = 1.0 \quad (20)$$

$$\gamma_{n,A} = \frac{1}{\alpha_n} \left[\frac{\sum_{s=2}^{\infty} \sigma_s \left\langle \sum_{j=1}^N x_n^{js} \right\rangle}{\left\langle \sum_{j=1}^N x_n^{j2} \right\rangle} \right] \quad (21)$$

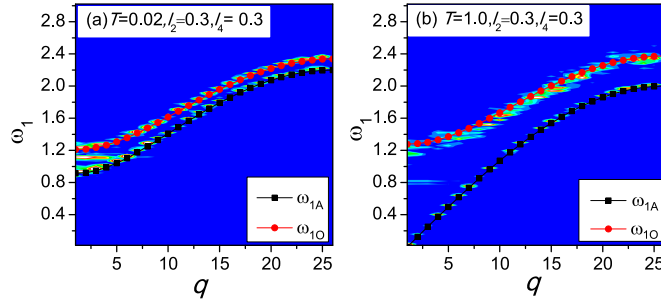


Figure 3. The phonon spectra of chain 1 at low temperature and high temperature for coupled FK chains: (a) $T = 0.02$; and (b) $T = 1.0$ when interface parameter $l_2 = 0.3$, $l_4 = 0.3$. The contour plots are the results by SED method and the red and black dotted lines are results of EPT. ω_{1A} and ω_{1O} denote the acoustic phonon branch and optical phonon branch of chain 1, respectively.

$$\sigma_s = (-1)^{s/2+1} \frac{(2\pi)^{s-2}}{(s-1)!} \quad (s \text{ is even}) \quad (22)$$

$$\gamma_{n,O} = \gamma_{n,A} \pm \frac{1}{\alpha_n} \left[\frac{l_2 \left\langle \sum_{j=2}^{N-1} (x_1^j - x_2^j)^2 \right\rangle}{\left\langle \sum_{j=2}^{N-1} (x_1^j - x_2^j) x_n^j \right\rangle} + \frac{l_4 \left\langle \sum_{j=2}^{N-1} (x_1^j - x_2^j)^4 \right\rangle}{\left\langle \sum_{j=2}^{N-1} (x_1^j - x_2^j) x_n^j \right\rangle} \right] \quad (23)$$

At the high-temperature regime, the on-site potential can be negligible due to the large kinetic energy of the particles, and FK chains recover to harmonic chains (as shown in figure 3(b)). Thus, the coefficient $\gamma_{n,A}$ and $\gamma_{n,O}$ become

$$\gamma_{n,A} = 0 \quad (24)$$

$$\gamma_{n,O} = \pm \frac{1}{\alpha_n} \left[\frac{l_2 \left\langle \sum_{j=2}^{N-1} (x_1^j - x_2^j)^2 \right\rangle}{\left\langle \sum_{j=2}^{N-1} (x_1^j - x_2^j) x_n^j \right\rangle} + \frac{l_4 \left\langle \sum_{j=2}^{N-1} (x_1^j - x_2^j)^4 \right\rangle}{\left\langle \sum_{j=2}^{N-1} (x_1^j - x_2^j) x_n^j \right\rangle} \right] \quad (25)$$

In figure 3, we plot the dispersion relation of coupled FK chains. The dotted lines also denote the result derived from effective phonon theory and the contour plots denote the SED patterns. It is clear that the phonon spectrum using EPT agrees well with the SED calculation.

3.3 The coupled ϕ^4 model

Another commonly studied model is the ϕ^4 model which has intrachain interactions $V_n(x_n^j - x_n^{j+1}) = \frac{g_2}{2}(x_n^j - x_n^{j+1})^2$ and an on-site potential $U_n(x_n^j) = \frac{1}{4}\sigma_4 x_n^{j4}$. For simplicity,

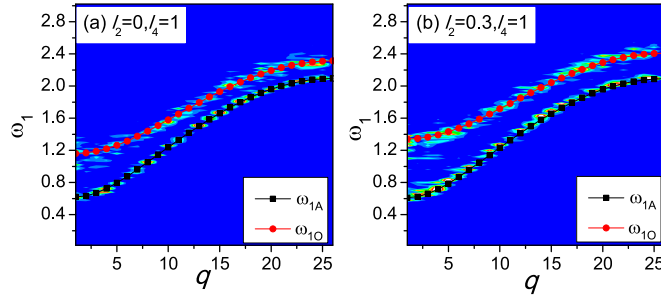


Figure 4. The phonon spectra of chain 1 with interchain couplings for the coupled ϕ^4 model: (a) $l_2 = 0.0, l_4 = 1.0$; and (b) $l_2 = 0.3, l_4 = 1.0$. The contour plots are the results using SED method and the red and black dotted lines are results of EPT. ω_{1A} and ω_{1O} denote the acoustic phonon branch and optical phonon branch of chain 1, respectively.

we set $g_2 = \sigma_4 = 1.0$. We consider the interchain couplings $V_{12}(x_1^j - x_2^j) = \frac{l_2}{2}(x_1^j - x_2^j)^2 + \frac{l_4}{4}(x_1^j - x_2^j)^4$. The coefficients $\alpha_n = 1.0, \gamma_{n,A}$ and $\gamma_{n,O}$ become

$$\gamma_{n,A} = \frac{1}{\alpha_n} \left[\frac{\langle \sum_{j=1}^N x_n^{j4} \rangle}{\langle \sum_{j=1}^N x_n^{j2} \rangle} \right] \quad (26)$$

$$\gamma_{n,O} = \gamma_{n,A} \pm \frac{1}{\alpha_n} \left[\frac{l_2 \langle \sum_{j=2}^{N-1} (x_1^j - x_2^j)^2 \rangle}{\langle \sum_{j=2}^{N-1} (x_1^j - x_2^j) x_n^j \rangle} + \frac{l_4 \langle \sum_{j=2}^{N-1} (x_1^j - x_2^j)^4 \rangle}{\langle \sum_{j=2}^{N-1} (x_1^j - x_2^j) x_n^j \rangle} \right]. \quad (27)$$

In our MD simulations, $T = 0.25$. Figure 4 shows the phonon spectrum of the coupled ϕ^4 chains. The result derived from EPT is also in good agreement with the prediction by SED method. Thus, the perfect agreement of the dispersion relation of different models between EPT and SED method confirms the EPT of coupled systems. For the general realistic solids, the interparticle potential can always be expanded as a polynomial form, so we are able to predict the phonon spectrum for the more complicated potential models in practical problems by EPT.

4. Conclusions

In conclusion, we have presented the effective phonon theory of coupled nonlinear lattices. Specifically, we obtain a generic form of renormalized phonon spectrum for a coupled system. It is found that for lattices with a harmonic interface or nonlinear interface, the dispersion relations predicted using the effective phonon theory are all in perfect agreement with the result obtained by SED method. Thus, the effective phonon theory of a coupled system is universal in its application to predict the phonon spectrum of a two-layer lattice structures. The findings in the present paper highlight the possibility of predicting the heat

conduction behavior of coupled lattices qualitatively and are very helpful for the design of novel thermal devices.

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