

Minimum Thermal Conductivity of Superlattices

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The phonon thermal conductivity of a multilayer is calculated for transport perpendicular to the layers. There is a crossover between particle transport for thick layers to wave transport for thin layers. The calculations show that the conductivity has a minimum value for a layer thickness somewhat smaller than the mean free path of the phonons.

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The thermal conductivity is a fundamental transport parameter [1]. There has been much recent interest in the thermal conductivity of semiconductor superlattices due to their possible applications in a variety of devices. Efficient solid state refrigeration requires a low thermal conductivity [2]. Preliminary experimental and theoretical work suggests that the thermal conductivity of superlattices is quite low, both for transport along the planes [3,4], or perpendicular to the planes [5–8]. The heat is carried by excitations such as phonons and electrons. Most theories use a Boltzmann equation which treats the excitations as particles and ignores wave interference [7,9]. These theories all predict that the thermal conductivity perpendicular to the layers decreases as the layer spacing is reduced in the superlattice. The correct description using the Boltzmann equation would be to use the phonon states of the superlattice as an input to the scattering, but this has not yet been done by anyone.

We present calculations of the thermal conductivity perpendicular to the layers which include the wave interference of the superlattice. These calculations, in one, two, and three dimensions, always predict that the thermal conductivity *increases* as the layer spacing is reduced in the superlattice. This behavior is shown to be caused by band folding in the superlattice. It is a general feature which should be true in all cases. The particle and wave calculations are in direct disagreement on the behavior of the thermal conductivity with decreasing layer spacing. This disagreement is resolved by calculations which include the mean free path (mfp) of the phonons. For layers thinner than the mfp, the wave theory applies. For layers thicker than the mfp the particle theory applies. The combined theory predicts a minimum in the thermal conductivity, as a function of layer spacing. The thickness of the layers for minimum thermal conductivity depends upon the average mfp, and is therefore temperature dependent.

The particle theories use the interface boundary resistance [10] as the important feature of a superlattice. A superlattice with alternating layers has a thermal resistance for one repeat unit of $R_{\text{SL}} = L_1/K_1 + L_2/K_2 + 2R_B$, where (L_j, K_j) are the thickness and thermal conductivity of the individual layers, and R_B is the thermal boundary resistance. For simplicity assume that $L_1 = L_2 \equiv L$,

which is often the case experimentally. The effective thermal conductivity of the superlattice is then

$$K_{\text{SL}} = \frac{2L}{R_{\text{SL}}} = \frac{2L}{L(1/K_1 + 1/K_2) + 2R_B}. \quad (1)$$

This classical prediction is that the thermal conductivity decreases as the layer thickness L decreases [9].

The wave theory calculates the actual phonon modes $\omega_\lambda(\mathbf{k})$ of the superlattice, where λ is the band index. They are used to calculate the thermal conductivity from the usual formula in d dimensions [1],

$$K(T) = \sum_\lambda \int \frac{d^d k}{(2\pi)^d} \hbar \omega_\lambda(\mathbf{k}) |v_z(\mathbf{k})| \ell_\lambda(\mathbf{k}) \frac{\partial n(\omega, T)}{\partial T}, \quad (2)$$

where $n(\omega, T)$ is the Bose-Einstein distribution function. A rigorous treatment uses Boltzmann theory applied to the transport in minibands to find the mean free path $\ell_\lambda(\mathbf{k})$. At high temperatures, one can approximate $n \sim k_B T / \hbar \omega_\lambda(\mathbf{k})$, which gives the simpler formula

$$K(T) = k_B \sum_\lambda \int \frac{d^d k}{(2\pi)^d} |v_z(\mathbf{k})| \ell_\lambda(\mathbf{k}). \quad (3)$$

The above formula is quite general. There are two important special cases of constant relaxation time (K_τ) and constant mfp (K_ℓ)

$$K_\tau(T) = k_B \tau \sum_\lambda \int \frac{d^d k}{(2\pi)^d} v_z(\mathbf{k})^2, \quad (4)$$

$$K_\ell(T) = k_B \ell \sum_\lambda \int \frac{d^d k}{(2\pi)^d} |v_z(\mathbf{k})|. \quad (5)$$

Both of these formulas can be related to the distribution $P(v_z)$ of phonon velocities perpendicular to the layers

$$P(v_z) = \sum_\lambda \int \frac{d^d k}{(2\pi)^d} \delta(v_z - |v_z(\mathbf{k})|), \quad (6)$$

$$K_\tau = k_B \tau \int dv_z P(v_z) v_z^2, \quad (7)$$

$$K_\ell = k_B \ell \int dv_z P(v_z) v_z. \quad (8)$$

Wave interference leads to band folding [11,12]. Band folding leads to a reduction of the phonon velocities. Both

K_τ and K_ℓ are reduced by wave interference. The case of constant mfp in one dimension can be reduced to a simple formula

$$K_\ell = \frac{k_B \ell}{2\pi} \sum_\lambda \int dk \left| \frac{d\omega}{dk} \right| \quad (9)$$

$$= \frac{k_B \ell}{2\pi} \sum_\lambda [\omega_\lambda^{\max} - \omega_\lambda^{\min}] \quad (10)$$

$$= \frac{k_B \ell}{2\pi} [\omega_{\max} - (\text{energy gaps})]. \quad (11)$$

The integration is eliminated.

These assertions are best illustrated in one dimension, that is with the atomic chain. The simplest model for a superlattice has all spring constants identical, and the layers differ in their masses. Layer one has $N/2$ atoms of mass m_1 and layer two has $N/2$ atoms of mass $m_2 = m_1/\alpha$. The characteristic matrix for phonons is given below for the case $N = 6$

$$\mathbf{M} = \begin{pmatrix} 2 & -1 & 0 & 0 & 0 & -e^{-ikN} \\ -1 & 2 & -1 & 0 & 0 & 0 \\ 0 & -1 & 2 & -1 & 0 & 0 \\ 0 & 0 & -\alpha & 2\alpha & -\alpha & 0 \\ 0 & 0 & 0 & -\alpha & 2\alpha & -\alpha \\ -\alpha e^{ikN} & 0 & 0 & 0 & -\alpha & 2\alpha \end{pmatrix}. \quad (12)$$

Setting the determinant $|\mathbf{M} - \omega^2 \mathbf{1}|$ to zero gives the characteristic equation (the easiest way to derive it is to utilize the similarity of the present problem to the Schrödinger equation with the Kronig-Penney potential) $k(\omega)$ of [13–15]

$$\begin{aligned} \cos(kN) &= \cos(k_1 N/2) \cos(k_2 N/2) \\ &\quad - \frac{1 - \cos(k_1) \cos(k_2)}{\sin(k_1) \sin(k_2)} \\ &\quad \times \sin(k_1 N/2) \sin(k_2 N/2), \end{aligned} \quad (13)$$

where $\cos(k_1) = 1 - \omega^2/2$, $\cos(k_2) = 1 - \omega^2/(2\alpha)$ define the wave vectors (k_1, k_2) in the individual layers in dimensionless units. A typical spectrum is shown in Fig. 1, where $\alpha = 2$ and the superlattice periods are $N = 2, 4, 8, 16$. Modes with frequency $\omega > 2$ are confined within the layer of the lighter atoms, and contribute little to the thermal conduction. As the value of N is increased in Fig. 1, there is more band folding, and the average velocity decreases. The lower curve in Fig. 2 shows the thermal conductivity as a function of superlattice period. The result for constant mfp is normalized to $k_B \ell \omega_{2,\max}$, where $\omega_{2,\max} = 2$ is the maximum phonon frequency in the layer 1 of heavy mass. The heat conduction is highest at small values of N , and rapidly decreases as N increases. It also shows an irregular nonmonotonous behavior which we understand and which will be discussed elsewhere. This result is for one dimension. Similar curves are found for every case

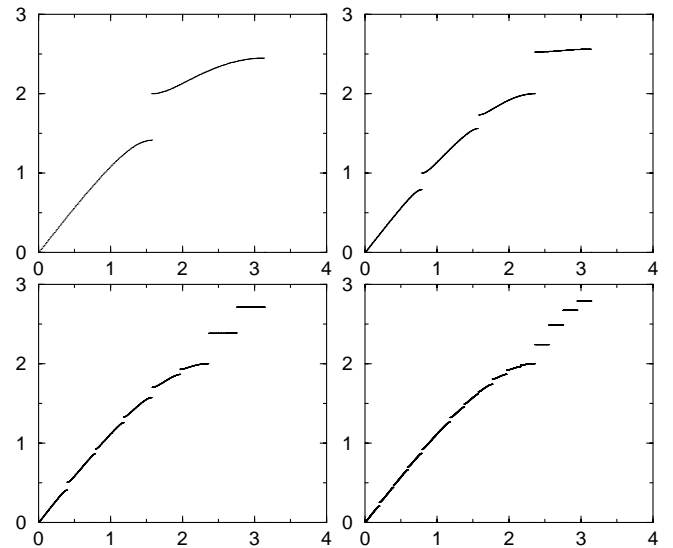


FIG. 1. Spectrum of $N = 2, 4, 8, 16$ superlattices with mass ratio $\alpha = 2$ in the extended zone representation.

which we have calculated: for different values of mass ratio α , and for both K_ℓ and K_τ . Generally, increasing N (i) increases the amount of band folding, (ii) decreases the average velocity in the superlattice, and (iii) decreases the thermal conductivity.

Lower curves in Figs. 3 and 4 show similar calculations of K_ℓ for mass ratio $\alpha = 2$ in two and three dimensions. Including only nearest-neighbor interactions, the characteristic matrix (12) is changed only in its diagonal element, where 2 is replaced by $2[2 - \cos(q)]$ in two dimensions, and $2[3 - \cos(q_x) - \cos(q_y)]$ in three dimensions. Here (q, q_x, q_y) are the wave vectors within the planes. These variables are integrated to find the result for the heat conduction. These cases also have the feature that the thermal

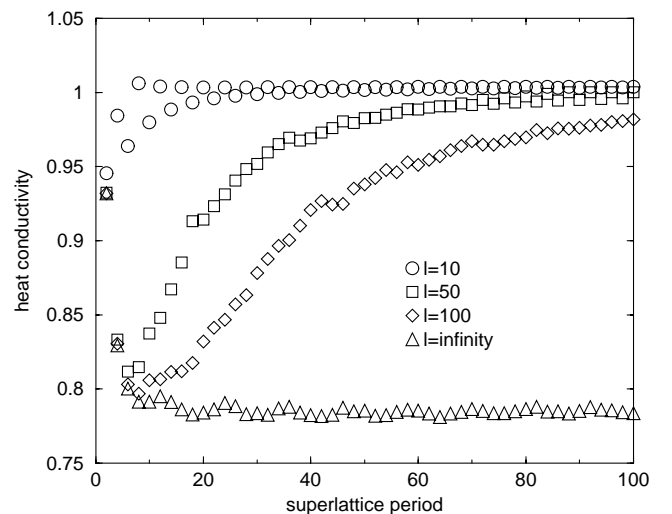


FIG. 2. Heat conductivity in one dimension as a function of the superlattice period for mass ratio $\alpha = 2$ for different values of the phonon mfp which are given in units of lattice periods. Dimensionless units found by dividing (11) by $2k_B \ell$.

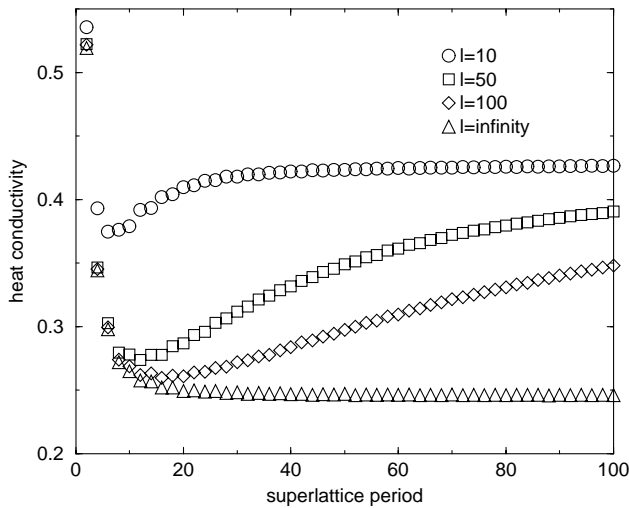


FIG. 3. Thermal conductivity in two dimensions as a function of the superlattice period for mass ratio $\alpha = 2$ for different values of the phonon mfp which are given in units of lattice periods. (In units of thermal conductivity of a uniform system of the heavier atoms.)

conductivity falls with increasing value of layer thickness. The curve is now smooth, due to the averaging over the parallel wave vectors. Similar results are found for all values of α .

In these calculations the thermal boundary resistance seems to have disappeared. Note that our model does predict thermal boundary resistance for a single interface [16]. The scattering of phonon waves at the interface causes the thermal resistance. This scattering is included in the present calculation. However, it also causes band folding, which is a bigger effect than the thermal boundary

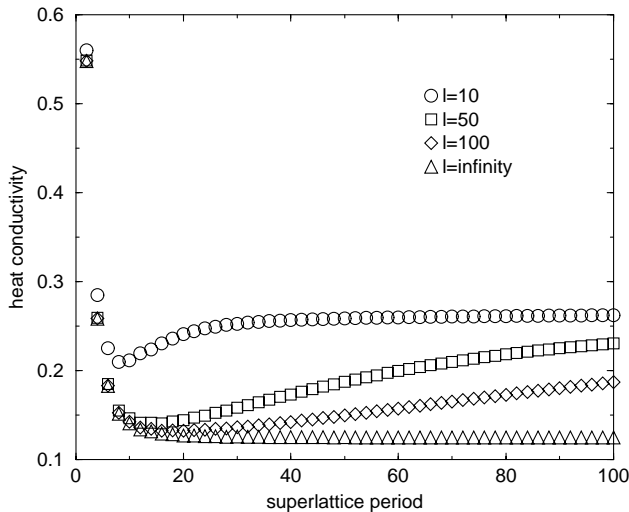


FIG. 4. Thermal conductivity in three dimensions as a function of the superlattice period for mass ratio $\alpha = 2$ for different values of the phonon mfp which are given in units of lattice periods. (In units of thermal conductivity of a uniform system of the heavier atoms.)

resistance. The wave calculation predicts that in three dimensions the thermal conductivity increases as the layer spacing is decreased, which is exactly the opposite of the particle calculation which predicts that it decreases. Since the effect is due simply to band folding, which is a well-documented phenomena, then the prediction must be accurate. Several prior calculations predicted behavior similar to that shown in Figs. 2–4 [17,18]. However, no explanation was given for the behavior.

The missing ingredient in these calculations is the mean free path of the phonons. When the layer thickness exceeds the mfp, then interference effects should diminish, and the particle model should become applicable. Our intuition is that the wave model should apply when $L < \ell$ and the particle model should apply when $L > \ell$. A phenomenological method of including ℓ is to add a complex part to the wave vector k which is i/ℓ . Then recalculate the properties of the superlattice using Eq. (13). This idea came from Pendry [19] who did the same thing for electron energy bands (and provides a more complete justification for this model): energy gaps go away if one includes a finite mfp. A similar result is found for phonons. The band gaps diminish to zero as the mfp is decreased for a fixed value of N .

Upper curves in Figs. 2–4 show the thermal conductivity in one, two, and three dimensions as a function of the superlattice period for four different values of mfp and $\alpha = 2$. The mfp is given in terms of the number of lattice spacings (note: not superlattice spacings). Similar curves are found for other values of α . For large values of ℓ the results are identical to the lower curve (which has $\ell = \infty$). For small values of mfp ($\ell = 10$) the thermal conductivity is nearly independent of superlattice period, except where it falls at small values of N .

The interesting cases have $\ell = 50$ or 100 . Here the thermal conductivity falls as N increases, reaches a minimum, and then starts to increase. This latter behavior is the situation expected in the experiments. At room temperature, in most solids, anharmonic scattering limits the phonon mfp to value in the range of 10–100 lattice constants, which is also the typical value of superlattice parameter in current devices. Therefore we expect the experimental thermal conductivities to behave as the curves marked $\ell = 50, 100$ in Figs. 2–4. The thermal conductivity should have a minimum value when plotted vs superlattice period. The minimum occurs at the crossover between the particle and wave-interference types of transport. One experimental result has this behavior [20].

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