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Theory of the thermal boundary resistance between dissimilar lattices

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Continuum acoustics allows only three different phonons to be reflected and transmitted for each phonon that is incident on an interface between two materials. We show that a lattice-dynamical theory for dissimilar lattices allows more phonons to be generated at the interface since we must introduce a larger unit cell. Numerical examples are given for the thermal boundary conductivity between two fcc crystals. The additional high-frequency phonons are found to carry an appreciable amount of the reflected and transmitted heat.

I. INTRODUCTION

When heat is transported across an interface between two different materials, there will in general be a temperature discontinuity at the interface. This effect was first observed at the interface between a metal and liquid helium[1] but has later also been seen at the interface between two solids.[2] The temperature discontinuity arises because some phonons are reflected at the interface and it is possible to define a thermal boundary resistance (Kapitza resistance), $R = A\Delta T/\dot{Q}$, where A is the area of the interface, $\dot{Q}$ is the heat flow across this area, and ΔT is the temperature difference between the distributions of phonons incident on the two sides of the interface. We have developed the first lattice-dynamical theory for the thermal boundary resistance that can be applied to cases where the two crystals have different lattice constants or even different crystal structure.

Since R is geometry independent it should be called a thermal boundary resistivity. However, we will follow the terminology as it is defined by Swartz *et al.*[2] and use thermal boundary resistance for R, but we will use the term thermal boundary conductivity for its inverse.

Theoretical estimates of the thermal boundary resistance between two solids have mainly been based on the acoustic mismatch theory.[3] In this theory, the two media are regarded as two elastic continua and a perfect junction between the two is assumed, i.e., the interface is "welded." A phonon that is incident on the interface has a certain probability of being reflected or transmitted and the boundary conditions allow three different phonons to be reflected and transmitted, respectively. Another model uses diffuse mismatch to explain the behavior of rough interfaces between a solid and liquid helium at higher temperatures.[2] It is assumed that all phonons are diffusively scattered at the interface, i.e., an incident phonon is reflected and transmitted into all phonon states with the same energy.

These theories work well for solid-solid interfaces at low temperatures, but at high temperatures the experimental thermal boundary resistance is, in some cases, found to be higher than that predicted by the theories. It has been suggested that the excess thermal boundary resistance at high temperatures is due to damage of the surface layer.[4]

Neither the diffusive mismatch theory nor simple versions of the acoustic mismatch theory takes into account phonon dispersion or the actual forces that act across the interface. There is also a possibility that the incident phonon has such a high frequency that there are no phonons in the other material with the same frequency. A lattice-dynamical theory is needed in order to include all these effects which are particularly important at high temperatures.

Several lattice-dynamical calculations have been made on one- and two-dimensional lattices.[5–7] The only calculation on a three-dimensional lattice has been made by Young and Maris.[8] However, all of these calculations have assumed that the lattice constants were the same in the two materials. Here we present a theory for dissimilar lattices.

II. THEORY

A. Similar lattices

First we review the theory of Ref. 8 for similar lattices. This case is drawn in Fig. 1(a), where we denote atoms with different masses with circles of different radii, and we distinguish between different force constants by the thickness of the line joining two atoms. Assume a phonon with wave vector $\mathbf{k}^{\text{in}}$ and frequency ω is incident on the interface from the left in Fig. 1(a). The outgoing phonon has wave vector $\mathbf{k}^{\text{out}}$. All atoms along one side of the boundary are in equivalent positions and the phase difference between incoming and outgoing phonons must therefore be the same at all interface atoms on one side of the boundary. This gives us the boundary condition

$$k_x^{\text{out}} = k_x^{\text{in}} + \frac{n 2\pi}{r} , \qquad (1)$$

where r is the common lattice constant and the only acceptable value for the integer n is zero since we require all wave vectors to lie in the first Brillouin zone.

In a three-dimensional lattice, Eq. (1) would also be valid for the k_y component. The k_z^{out} component is determined from the requirement that the frequencies of the incoming and outgoing phonons must be the same, i.e.,

$$\omega(k_x^{\rm in},k_y^{\rm in},k_z^{\rm in})=\omega'(k_x^{\rm out},k_y^{\rm out},k_z^{\rm out}) \ , \tag{2}$$

where the prime denotes that ω is a different function of $\mathbf{k}$ if the phonon is transmitted. On each side of the interface, there are six values of $k_z^{\rm out}$ that are solutions to Eq. (2). Only three of these are physically acceptable. We choose the solutions that correspond to phonons traveling away from the interface or, in the case of imaginary values of $k_z^{\rm out}$, we choose the evanescent waves whose amplitudes are vanishingly small far from the interface. The ratios of the phonon amplitudes are obtained from the equations of motion for the atoms at the interface.[8]

From the phonon amplitudes we get the transmission coefficient, $t(\mathbf{k},\mathbf{j})$, which is defined as the fraction of the energy that is transmitted when a phonon with wave vector $\mathbf{k}$ and branch index j is incident on the interface. The heat flow across the interface is now obtained from

$$\dot{Q}=\frac{A}{V}\left[\sum_{k_1,j_1}^{+}\hbar\omega(\mathbf{k}_1,j_1)N_0(\omega_1,T_1)v_z(\mathbf{k}_1,j_1)t(\mathbf{k}_1,j_1)\right.$$
$$\left.+\sum_{\mathbf{k}_2,j_2}^{-}\hbar\omega(\mathbf{k}_2,j_j)N_0(\omega_2,T_2)v_z(\mathbf{k}_2,j_2)t(\mathbf{k}_2,j_2)\right] \ , \tag{3}$$

where N_0 is the thermal equilibrium distribution. The plus (minus) sign indicates that we sum only over those phonons that are traveling towards the interface in material 1 (2).

B. Dissimilar lattices

Now we show how to extend the theory to the case of dissimilar lattices. In Fig. 1(b) we have two lattices with $3r_1=2r_2$, where r_1 and r_2 are the lattice constants to the left and to the right of the interface, respectively. The phase differences between incoming and reflected waves are not required to be the same at neighboring atoms since the forces acting on neighboring atoms are different. Interface atoms in equivalent positions in material 1 are separated a distance $3r_1$ and Eq. (1) must therefore be replaced by

$$k_x^{\rm refl}=k_x^{\rm in}+\frac{n2\pi}{3r_1}, \quad n=0,1,2 \ . \tag{4}$$

For the transmitted phonons we find

$$k_x^{\rm trans}=k_x^{\rm in}+\frac{n\pi}{r_2}, \quad n=0,1 \ . \tag{5}$$

The k_z components are again obtained from Eq. (2). For three-dimensional simple cubic lattices the equations for the k_y components will be identical to Eqs. (4) and (5) and there will be 27 reflected waves and 12 transmitted waves. Thus, different lattice constants in the two materials will allow more phonons to be generated at the interface.

In general we can have different crystal structures in the two materials. Therefore we will derive a more general expression for the k_x and k_y components for the outgoing waves. As an example of a more complex case, consider the interface between two fcc crystals with the (100) planes parallel to the interface. The positions of the atoms at the interface in material 1 are shown to the left in Fig. 1(c) and the positions of the corresponding atoms in material 2 are shown to the right. The lattice constant in material 2, r_2, is twice the lattice constant in material 1, r_1. The boundary for a choice of primitive unit cell in material 1 is given by the dashed line. It is easily seen that neighboring unit cells are not in equivalent positions relative to the atoms in material 2. We must define a surface unit cell that is four times larger (solid line) than the unit cell in the bulk in order to include all possible interactions with atoms in the other material.

This larger surface unit cell has translational symmetry along the interface so we can use the arguments for the similar lattice [Fig. 1(a)] to obtain $k_x^{\rm refl}=k_x^{\rm in}$ and $k_y^{\rm refl}=k_y^{\rm in}$, where $\mathbf{k}$ is defined in the surface Brillouin

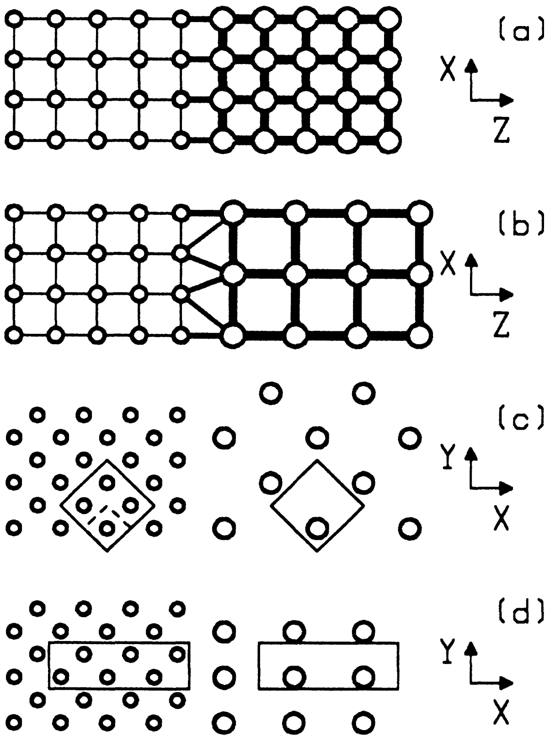

FIG. 1. (a) Location of atoms in a plane perpendicular to the interface between two similar lattices; (b) location of atoms in a plane perpendicular to the interface between two dissimilar lattices; (c) location of atoms in (100) planes parallel to the interface for two dissimilar fcc lattices. The lines mark the boundaries of different unit cells that are defined in the text; (d) left, location of atoms in a (100) plane parallel to the interface; right, location of atoms in a (110) plane parallel to the interface. The lines mark the boundaries of the surface unit cells.

zone. This gives us twelve reflected phonons since we now have four times as many phonon branches. Usually we would like to have the phonons defined in the bulk Brillouin zone and we must then unfold the phonon branches by adding surface reciprocal-lattice vectors.

In the general case where the surface unit cell is n times larger than the bulk unit cell, we need n surface reciprocal-lattice vectors, g^k, to unfold the phonon branches. The n allowed values for $k_x^{\rm refl}$ in the bulk Brillouin zone will be $k_x^{\rm in}, k_x^{\rm in}+g_x^1, \ldots, k_x^{\rm in}+g_x^{n-1}$ and similar for the y components. The same scheme can be used to obtain the allowed values for $k^{\rm trans}$ by determining the corresponding surface unit cell in material 2. In our example in Fig. 1(c) the surface unit cell is equal to the bulk unit cell and we do not get any extra allowed **k** vectors.

The advantage of this last approach is seen when it is applied to a grain boundary. In Fig. 1(d) we have the same material on both sides of the interface but we have a (100) plane parallel to the interface on the left and a (110) plane parallel to the interface to the right. The distance between neighbor atoms in the x direction in the (110) plane is $\sqrt{2}r_1 \approx 1.5 r_1$. The Brillouin zones to the left and to the right have now been rotated with respect to each other, but as soon as the surface unit cell has been identified, it is straightforward to use the same scheme as above. In this case the surface unit cells to the left and to the right contain six and two atoms, respectively. For each incoming phonon there will be eighteen reflected phonons and six transmitted phonons.

III. NUMERICAL EXAMPLES

We have made a numerical calculation of the boundary resistance between two fcc crystals, both of which have their (100) planes parallel to the interface. The model crystals consist of masses, m_i, connected with springs with spring constants K_i. We used the method of Young and Maris[8] to find the solution of Eq. (2). In all cases we have studied, we have $m_1=1$, $K_1=1$, and $r_1=1$. In the first case we use these parameter values also in material 2. In order to compare our theory with the acoustic mismatch theory, we change the mass, spring constant, and lattice constant in material 2 so that it will have the same acoustic impedance and the same critical cone as in the case with $r_2=1$. We get $m_2=8, K_2=2$ for $r_2=2$ and $m_2=3.375, K_2=1.5$ for $r_2=1.5$. We do not allow for relaxation of the atoms at the interface since this would complicate the calculations. An atom in the lattice with the smaller lattice constant interacts with 4–6 atoms on the other side of the boundary, depending on its position. The corresponding spring constants at the interface are all chosen to be equal to $(K_1+K_2)/2$ to ensure a strong bonding across the interface. We note that a simple acoustic mismatch theory would predict identical temperature dependence for these three cases.

In Fig. 2(a) we show the result of our calculations for three different ratios of the lattice constants: 1, 1.5, and 2, shown by the solid, dashed, and dotted lines, respectively. We have plotted thermal boundary conductivity against temperature, scaled with the constants $\Theta_D=2.965(K_1/m_1)^{1/2}\hbar/k_B$ and $\sigma_0=(K_1/m_1)^{1/2}k_B/r_1^2$ (see Ref. 8). At low temperatures we do indeed find identical temperature dependences as is expected with the acoustic mismatch model, but at higher temperatures the thermal boundary conductivity saturates at different values.

It may seem unphysical that we obtain a value for the thermal boundary resistance even when the two materials are identical [solid line in Fig. 2(a)], but with our definition of ΔT this is not a problem as we show in the Appendix. As long as you measure the appropriate temperatures this theory is valid even when the two materials are almost identical.[2,9]

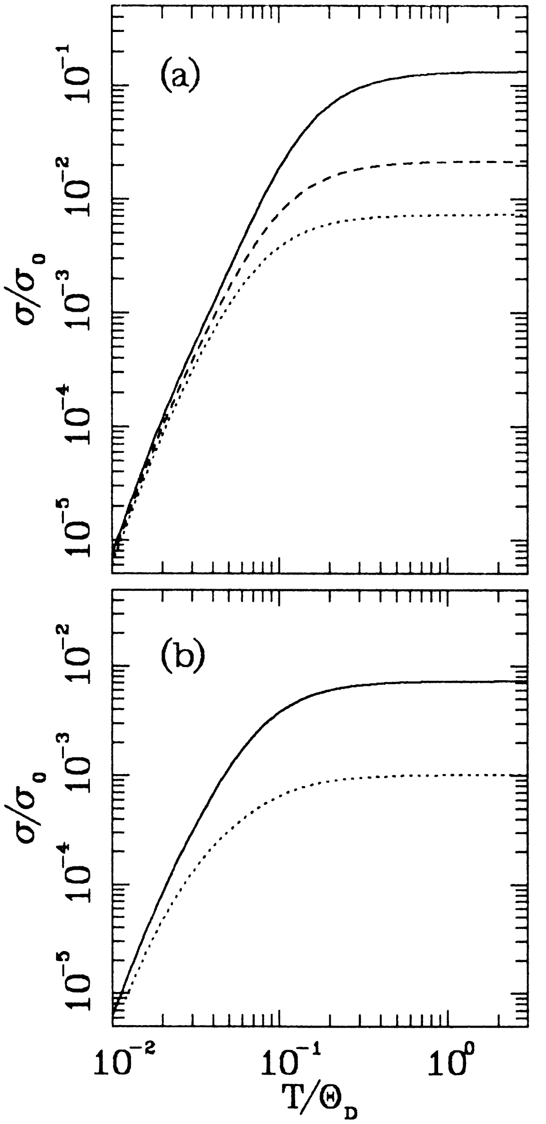

FIG. 2. Thermal boundary conductivity between two different fcc crystals, both with their (100) planes parallel to the interface. $K_1=1$, $m_1=1$, and $r_1=1$ in all cases. (a) ——, $K_2=1$, $m_2=1$, $r_2=1$; — — —, $K_2=1.5$, $m_2=3.375$, $r_2=1.5$; . . . ., K_2, $m_2=8$, $r_2=2$; (b) $K_2=2$, $m_2=8$, and $r_2=2$ for both curves. . . . ., interface spring constants from Lennard-Jones potential. ——, all interface spring constants equal to $(K_1+K_2)/2$.

The saturation of the thermal boundary conductivity in Fig. 2(a) is mainly due to the cutoff in phonon frequency in material 2, but a detailed study shows that the phonon transmission coefficient (i.e., the ratio between transmitted and incident heat flux) starts decreasing well below the cutoff frequency. It is only the very-long-wavelength phonons that have a transmission coefficient near 1. With dissimilar lattices the transmission coefficient is considerably smaller than when the lattices match. There are two possible reasons for this.

The first reason is that the present theory introduces extra outgoing phonons in addition to the three reflected and three transmitted phonons that are allowed for similar lattices. The extra phonons can carry an appreciable part of the outgoing heat, especially for the ratio 1:1.5 of the lattice constants. In this case we find that for 20% of the incoming phonons the extra reflected phonons carry more of the reflected heat than the "ordinary" reflected phonons do. Similar numbers hold for the transmitted heat. The extra phonons carry heat only at high frequencies. At low frequencies, the extra phonons are all evanescent and carry no heat.

The second reason is that the bonding at the interface is weakened as the atoms on opposite sides of the interface no longer match. The thermal boundary resistance is very sensitive to what forces we assume at the interface. An example of this is shown in Fig. 2(b), where the upper curve is calculated with equal spring constants for all interactions across the interface. The lower curve is obtained with these spring constants calculated from a Lennard-Jones potential. In the latter case the spring constants vary by 2 orders of magnitude since the distances between interacting atoms are very different. In fact, one pair of atoms dominates the interactions at the interface. In a real crystal the atoms at the interface would relax to new equilibrium positions and the force constants would be similar in magnitude. On the other hand, we would then have two layers of atoms that do not have the structure of either lattice, which would make it more difficult to transmit phonons.

It is not possible to separate the effect of the "extra" phonons from the effect of the weakly bonded interface due to mismatch of the atoms at the interface. The existence of additional reflected and transmitted phonons requires that there is no one-to-one correspondence of the atoms on the two sides of the interface and vice versa. However, in all cases we have studied the net effect is a smaller transmission coefficient.

We expect that our theory will have its main applications to certain grain boundaries where it is possible to use a relatively small surface unit cell. It will probably be necessary to allow for relaxation so that realistic forces between atoms on opposite sides of the interface can be calculated. The phonon amplitudes must then be obtained from the equation of motion for the atoms in the second plane from the interface.

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APPENDIX

It has been argued[10] that it is not correct to use Eq. (3) since it leads to a finite value of the thermal boundary resistance even when the two materials are identical. The problem is related to the thermal equilibrium distribution that has been used in Eq. (3). If we have a net flow of energy the phonon distribution cannot be the thermal equilibrium distribution. This apparent "paradox" has been resolved[10] by introducing the actual phonon distribution that corresponds to a net heat flow.

However, Simons[10] used the total phonon distribution in his derivation while we have used the phonon distribution for the phonons that are incident on the interface. It is important to distinguish these two cases. When theory is compared with experiment it is important to know what temperature the thermometers are measuring.[2,9] According to Katerberg *et al.*, most experimental setups measure the temperature of the phonons that are incident on the interface.

We will show that Eq. (3) is consistent with the ordinary theory for the thermal conductivity when the two materials are identical. Since the materials are identical and all transmission coefficients are equal to 1, we can write Eq. (3) as

$$\frac{\dot{Q}}{A}=\frac{1}{V}\sum_{\mathbf{k},j}^{+}\hbar\omega(\mathbf{k},j)v_z(\mathbf{k},j)[N_0(\omega,T_1)-N_0(\omega,T_2)]. \qquad \text{(A1)}$$

For a phonon that hits the interface, the mean time back to the last scattering event is $\tau(\mathbf{k},j)$. Phonons that are scattered will have a distribution that corresponds to the temperature at the place where the scattering occurred, i.e., $T=v_z(\mathbf{k},j)\tau(\mathbf{k},j)dT/dz$. This gives

$$\frac{\dot{Q}}{A}=\frac{1}{V}\sum_{\mathbf{k},j}\hbar\omega(\mathbf{k},j)v_z^2(\mathbf{k},j)\tau(\mathbf{k},j)\frac{dN_0}{dT}\left(-\frac{dT}{dz}\right), \qquad \text{(A2)}$$

where the summation now extends over all phonons in the Brillouin zone. The factor in front of the temperature gradient is seen to be the familiar expression for the thermal conductivity. Thus, we have shown that Eq. (3) is consistent with the usual theory for the thermal conductivity when we assume that the two materials are identical.

We have made the derivation in this Appendix to stress the fact that Eq. (3) is not unphysical even when the two materials are the same, as long as the proper temperatures are used. A discussion on which experimental setups measure temperatures appropriate to Eq. (3) has been given by Katerberg *et al.*[9]

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