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Thermal conductivity of superlattices

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The lattice thermal conductivity of superlattices is shown to depend on a new kind of umklapp scattering process, called a mini-umklapp, associated with the mini-Brillouin zone of the superlattice. Expressions for three-phonon mini-umklapp-scattering matrix elements are obtained for superlattices consisting of lattice-matched simple-cubic layers. The temperature dependence of the thermal conductivity is evaluated using an extension of Callaway's phenomenological model.

I. INTRODUCTION

Man-made superlattices[1] have attracted considerable scientific interest because of their great potential for use in new electronic devices.[2] For the most part, studies of such superlattices have focused primarily on the electronic and optical properties of technological interest, rather than on the vibrational and mechanical properties.[3] In this paper we report a theory of phonon thermal conductivity in a superlattice consisting of alternating slabs of insulating material.

It is well known[4–6] in the theory of lattice thermal conductivity that the normal three-phonon scattering processes in which the total crystal momentum is conserved cannot by themselves lead to a finite thermal resistance. Only those processes in which crystal momentum is not conserved contribute to the lattice thermal resistance: boundary-scattering, impurity-scattering, and umklapp-scattering processes in which the change of crystal momentum equals some reciprocal-lattice vector $\vec{G}$.

Man-made superlattices consisting of periodically alternating layers of lattice-matched crystalline material have a large superlattice constant and hence a small or mini-Brillouin zone in the direction of layering. The mini-reciprocal-lattice vectors associated with this minizone give rise to minimum-klapp processes which contribute to the thermal resistance.[7]

Here we use a phenomenlogical model of the type proposed by Callaway[8] for bulk thermal conductivity to predict the additional temperature dependence of the thermal resistance of a superlattice. We employ a phenomenological rather than a first-principles approach because (i) the theory of thermal conductivity, as influenced by multiphonon processes, is sufficiently complicated numerically that no successful *a priori* theory has been reported, to our knowledge, and (ii) our purpose is to explore the underlying physics of mini-umklapp processes in the simplest available model.

II. THREE-PHONON-SCATTERING MATRIX ELEMENTS

The basic physics is contained in the three-phonon scattering matrix elements: $\Phi_{\lambda\lambda'\lambda''}^{\vec{k}\,\vec{k}'\,\vec{k}''}$. Here we use the notation of Ref. 9: $\vec{k}$, $\vec{k}'$, and $\vec{k}''$ are the wave vectors of the three involved phonons, and λ, λ', and λ'' are corresponding vibrational branches or polarization indices.

A. 1×1 superlattice

To illustrate how the superlattice affects Φ, we first consider the example of a 1×1 isotopic one-dimensional superlattice.[7] We assume that the two constituents A and B have the same lattice constants and that the interactions between A-A, B-B, and A-B are exactly the same. The only difference between atoms A and B is the mass difference: $m_A \neq m_B$. In this model the unit cell in real space is twice that of a bulk unit cell, and the mini-Brillouin zone of the superlattice is half that of a bulk Brillouin zone (Fig. 1). Hence there are the

two different types of umklapp processes depicted in Fig. 1. Ordinary umklapps occur in three-phonon scattering in the bulk crystals of both constitutents, mini-umklapp processes occur only in a superlattice. We can imagine any bulk material of A as a fictitious "superlattice" with two constitutents that are exactly the same; in this limit mini-umklapp processes must be forbidden, giving zero thermal resistance. Physically it is clear that the mini-umklapps disappear in this limit of $m_A = m_B$; hence the mini-umklapp scattering rate must vanish as some power of $m_A - m_B$.

In order to evaluate $\Phi^{\vec{k}\,\vec{k}'\,\vec{k}''}_{\lambda\lambda'\lambda''}$ we assume there are only nearest-neighbor interactions in our model. In Leibfried and Ludwig's notation, the coupling parameters of third order in a site representation, $\Phi^{0mn}_{\mu\nu\kappa}$, are zero unless two of the site indices, $(0,\mu)$, (m,ν), (n,κ), refer to the same atomic site and the third to a neighboring site. Thus only twelve of the parameters $\Phi^{0mn}_{\mu\nu\kappa}$ are nonzero; all of these have the values $\pm g$, where g is a third-order anharmonic force constant.[10]

The desired three-phonon matrix elements $\Phi^{\vec{k}\,\vec{k}'\,\vec{k}''}_{\lambda\lambda'\lambda''}$ are obtained from $\Phi^{0mn}_{\mu\nu\kappa}$ by the usual Fourier normal-model transformation. For the present model we find

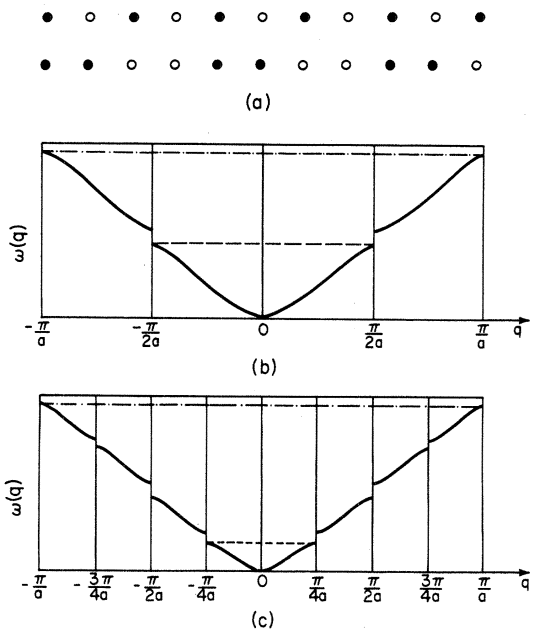

FIG. 1. (a) Schematic illustrations of one dimensional 1×1 (upper) and 2×2 (lower) superlattices. (b) Brillouin zone of 1×1 superlattice, its bulk umklapp process wave vector (dash-dotted line) and its smaller mini-umklapp process wave vector (dashed). (c) Brillouin zone of a 2×2 superlattice, its bulk umklapp process wave vector (dash-dotted line), and the smallest mini-umklapp process wave vectors (dashed).

$$\Phi^{kk'k''}_{\lambda\lambda'\lambda''} = \tfrac{1}{2}\Delta(k+k'+k'')g\frac{1}{(M_A M_B)^{1/2}}$$
$$\times\Big\{[\exp(ik''a)-\exp(-ik''a)]$$
$$\times\left[\frac{1}{M_A^{1/2}}e_1(k\lambda)e_1(k'\lambda')e_2(k''\lambda'')+\frac{1}{M_B^{1/2}}e_2(k\lambda)e_2(k'\lambda')e_1(k''\lambda'')\exp[i(k+k'+k'')a]\right]$$
$$+[\exp(ik'a)-\exp(-ik'a)]\left[\frac{1}{M_A^{1/2}}e_1(k\lambda)e_2(k'\lambda')e_1(k''\lambda'')\right.$$
$$\left.+\frac{1}{M_B^{1/2}}e_2(k\lambda)e_1(k'\lambda')e_2(k''\lambda'')\exp[i(k+k'+k'')a\right]$$
$$+[\exp(ika)-\exp(-ika)]\left[\frac{1}{M_A^{1/2}}e_2(k\lambda)e_1(k'\lambda')e_1(k''\lambda'')\right.$$
$$\left.+\frac{1}{M_B^{1/2}}e_1(k\lambda)e_2(k'\lambda')e_2(k''\lambda'')\right.$$
$$\left.\times\exp[i(k+k'+k'')a]\right]\Big\}. \tag{2.1}$$

Here the wave vectors k are all in the direction of layering, e is a vibrational amplitude, and $\Delta(k)$ is unity if k is a mini-reciprocal-lattice vector, but is zero otherwise. By using (2.1) and Fig. 1 we can discuss two spe-

cial cases: ordinary umklapp processes and mini-umklapp processes. For the ordinary umklapp processes, we have $k+k'+k''=2(2\pi/L)=2\pi/a$, and

$$\Phi^{kk'k''}_{\lambda\lambda'\lambda''}=\tfrac{1}{2}g\frac{1}{(M_AM_B)^{1/2}}$$
$$\times\left[\left[\frac{1}{M_A^{1/2}}e_1(k\lambda)e_2(k'\lambda')e_1(k''\lambda'')+\frac{1}{M_B^{1/2}}e_2(k\lambda)e_1(k'\lambda')e_2(k''\lambda'')\right][\exp(ik'a)-\exp(-ik'a)]\right.$$
$$+\left[\frac{1}{M_A^{1/2}}e_1(k\lambda)e_1(k'\lambda')e_2(k''\lambda'')+\frac{1}{M_B^{1/2}}e_2(k\lambda)e_2(k'\lambda')e_1(k''\lambda'')\right][\exp(ik''a)-\exp(-ik''a)]$$
$$\left.+\left[\frac{1}{M_A^{1/2}}e_2(k\lambda)e_1(k'\lambda')e_1(k''\lambda'')+\frac{1}{M_B^{1/2}}e_1(k\lambda)e_2(k'\lambda')e_2(k''\lambda'')\right][\exp(ika)-\exp(-ika)]\right]. \tag{2.2}$$

This umklapp process always occurs provided the anharmonic coupling exists ($g\neq 0$) even if $M_A=M_B=M$, in which case we have $e_1(k\lambda)=e_2(k\lambda)=e(k\lambda)$:

$$\Phi^{kk'k''}_{\lambda\lambda'\lambda''}=g\frac{1}{M^{3/2}}e(k\lambda)e(k'\lambda')e(k''\lambda'')[\exp(ika)+\exp(ik'a)+\exp(ik''a)$$
$$-\exp(-ika)-\exp(-ik'a)-\exp(-ik''a)]\,. \tag{2.3}$$

Equation (2.3) is exactly the form of the three-phonon umklapp-scattering matrix element for a monoatomic chain. For the mini-umklapp processes we have $k+k'+k''=2\pi/L=\pi/a$ and

$$\Phi^{kk'k''}_{\lambda\lambda'\lambda''}=\tfrac{1}{2}g\frac{1}{(M_AM_B)^{1/2}}$$
$$\times\left[\left[\frac{1}{M_A^{1/2}}e_1(k\lambda)e_1(k'\lambda')e_2(k''\lambda'')-\frac{1}{M_B^{1/2}}e_2(k\lambda)e_2(k'\lambda')e_1(k''\lambda'')\right][\exp(ik''a)-\exp(-ik''a)]\right.$$
$$+\left[\frac{1}{M_A^{1/2}}e_1(k\lambda)e_2(k'\lambda')e_1(k''\lambda'')-\frac{1}{M_B^{1/2}}e_2(k\lambda)e_1(k'\lambda')e_2(k''\lambda'')\right][\exp(ik'a)-\exp(-ik'a)]$$
$$\left.+\left[\frac{1}{M_A^{1/2}}e_2(k\lambda)e_1(k'\lambda')e_1(k''\lambda'')-\frac{1}{M_B^{1/2}}e_1(k\lambda)e_2(k'\lambda')e_2(k''\lambda'')\right][\exp(ika)-\exp(-ika)]\right] \tag{2.4}$$

In the limit of no superlattice, we have $M_A=M_B$ and $e_1(k\lambda)=e_2(k\lambda)$; thus $\Phi^{kk'k''}_{\lambda\lambda'\lambda''}$ for mini-umklapps vanishes, as expected. In the general case of a superlattice ($m_A\neq m_B$), we can solve the equations of motion for $e_2(k\lambda)/e_1(k\lambda)$:

$$\frac{e_2(k\lambda)}{e_1(k\lambda)}=\frac{2\frac{1}{M_B}\cos(ka)}{\frac{1}{M_B}-\frac{1}{M_A}+\left[\left[\frac{M_A+M_B}{M_AM_B}\right]^2-\frac{4\sin^2(ka)}{M_AM_B}\right]^{1/2}}\,. \tag{2.5}$$

If we define

$$\frac{1}{M_0}=\frac{1}{2}\left[\frac{1}{M_A}+\frac{1}{M_B}\right], \tag{2.6a}$$

and

$$\Delta\frac{1}{M}=\frac{1}{2}\left[\frac{1}{M_A}-\frac{1}{M_B}\right], \tag{2.6b}$$

then we find

$$\frac{e_2(k\lambda)}{e_1(k\lambda)}=\frac{\left[\frac{1}{M_0}-\Delta\frac{1}{M}\right]\cos ka}{\left[\left[\frac{1}{M_0}\right]^2\cos^2(ka)+\left[\Delta\frac{1}{M}\right]^2\sin^2(ka)\right]^{1/2}-\Delta\frac{1}{M}}. \tag{2.7}$$

By inserting (2.7) into (2.4) we will have $\Phi^{kk'k''}_{\lambda\lambda'\lambda''}$ as a function of $1/M_0$ and $\Delta(1/M)$, a rather complicated expression.

The three-phonon scattering matrix element simplifies greatly in the case of small isotopic mass difference: $\Delta(1/M)/(1/M_0)<<1$. Then we find

$$\frac{e_2(k\lambda)}{e_1(k\lambda)}=1+C(k)\frac{\Delta\frac{1}{M}}{\frac{1}{M_0}}=1-C(k)\frac{\Delta M}{M_0}, \tag{2.8}$$

where we have

$$C(k)=\frac{1-\cos(ka)}{\cos(ka)}. \tag{2.9}$$

Thus the matrix element becomes

$$\begin{aligned}\Phi^{kk'k''}_{\lambda\lambda'\lambda''}=&\tfrac{1}{2}g\frac{1}{M_0^{3/2}}e_1(k\lambda)e_1(k'\lambda')e_1(k''\lambda'')\frac{\Delta M}{M_0}\\&\times\{[C(k'')-C(k)-C(k')+\tfrac{1}{2}][\exp(ik''a)-\exp(-ik''a)]\\&+[C(k')-C(k'')-C(k)+\tfrac{1}{2}][\exp(ik'a)-\exp(-ik'a)]\\&+[C(k)-C(k')-C(k'')+\tfrac{1}{2}][\exp(ika)-\exp(-ika)]\}.\end{aligned} \tag{2.10}$$

The most important feature of this three-phonon mini-umklapp matrix element is that it is proportional to the fractional "disorder" of the superlattice $\Delta M/M_0$. Thus we expect mini-umklapp-scattering rates to be proportional to the square of the amplitude of the broken symmetry.

B. $N_1\times N_2$ superlattice

An $N_1\times N_2$ superlattice consists of alternating slabs of N_1 layers of material A and N_2 layers of B. Even for the isotopic superlattice discussed above, analytic solutions for the mini-umklapp-scattering matrix elements are not easily obtained. Since our purpose here is to emphasize the phenomena rather than to present specific and detailed calculations, we concentrate on those features of $N_1\times N_2$ superlattice mini-umklapp-scattering that we expect to be general and independent of the specific model. We do note however that even for an $N_1\times N_2$ superlattice there are N_1+N_2-1 different mini-reciprocal-lattice vectors; each of these vectors (and integral multiples of them) corre-

sponds to a mini-umklapp scattering.

The general features we expect of such scattering are that the scattering rate should be proportional to $(\Delta M/M_0)^2$, the square of the amplitude of broken symmetry, and approximately proportional to the square of the smallest mini-reciprocal-lattice vector, i.e., proportional to $(N_1+N_2)^{-2}$.

III. RELAXATION-TIME APPROXIMATION

Even if the mini-umklapp-scattering matrix element can be evaluated, the solution of the Boltzmann equation to determine the resulting thermal conductivity is normally not possible. Thus we follow the established tradition of constructing a phenomenological theory of thermal conductivity, with the nature of the phenomenological elements dictated by the microscopic theory of the three-phonon matrix element (Sec. II).

We employ a generalized version of Callaway's relaxation-time approximation[7] and treat only the three-phonon scattering and boundary-scattering contributions to the thermal conductivity. The scattering rates τ^{-1} for normal and umklapp scattering are assumed to have Callaway's postulated form:

$$\frac{1}{\tau_U}=B_U T^3\omega^2+C/L\ , \tag{3.1}$$

$$\frac{1}{\tau_N}=B_N T^3\omega^2\ . \tag{3.2}$$

Here B_U depends on temperature but B_N does not. The second term on the right side of (3.1) is a boundary-scattering term; the first term is a three-phonon umklapp-scattering term. For bulk umklapp processes, B_U contains a temperature-dependent factor $e^{-\Theta/aT}$, with Θ the Debye temperature, and a a constant.

According to Ziman,[6] a three-phonon umklapp-scattering rate is proportional to the square of the change of total crystal momentum involved. Thus we assume for a $N_1\times N_2$ superlattice that the mini-umklapp processes contribute to the scattering additively:

$$B_U=C_U[e^{-\Theta/aT}+(\Delta M/M)^2(N_1+N_2)^{-2}e^{-\Theta/a(N_1+N_2)T}]\ , \tag{3.3}$$

where C_U is a temperature-independent constant. The factor N_1+N_2 appears in the exponent because crystal momentum conservation (modulo a reciprocal-lattice vector) dictates that the energy-conserving scattering processes involve two phonons of a wave vector approximately half the distance from the center of the Brillouin zone to the minizone boundary; these phonons have an energy or a thermal activation temperature of $\sim\Theta/a(N_1+N_2)$.

Because he could not separate B_N and B_U, Callaway just neglected the temperature dependence of B_U and took the value of B_U+B_N determined at 75 K to be a constant. That is, he actually neglected B_U and took $B_U+B_N=B_N$. Instead of doing this, we assume mini-umklapp processes will dominate the thermal resistance of a superlattice, and take $B_U+B_N=B_U$, with B_U having the temperature dependence shown in Eq. (3.3). We might have overestimated the temperature dependence of B_U+B_N, in contrast to Callaway, who underestimated it.

By using these approximations and the data for C/L, a, Θ, and B_U+B_N,[8] we can calculate the thermal conductivity of a fictitious Ge-like superlattice. Our results are shown in Figs. 2 and 3. Figure 3 displays the temperature dependence of the thermal conductivity for the same kind of superlattice as in Fig. 2, but with a sample size ten times larger. The mini-umklapp-scattering effect becomes more evident in this latter case.

It should be emphasized that the mini-umklapp scattering produces a $\lesssim 25\%$ reduction of the thermal-conductivity peak. Experiments searching for this effect should take steps to guarantee that phonon scattering by defects introduced in superlattice fabrication does not mask the mini-umklapp scattering.

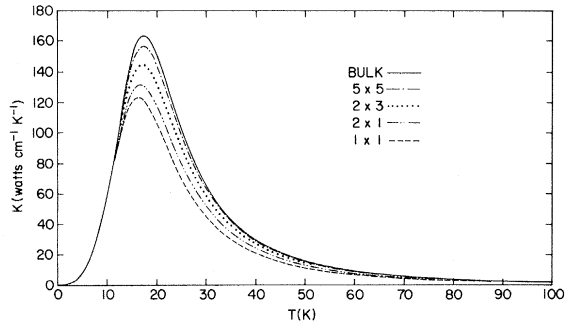

FIG. 2. Temperature dependence of the thermal conductivity for 1×1, 2×1, 2×3, and 5×5 superlattices, compared with the bulk thermal conductivity, after Ref. 7. The model sample size is $L=0.18$ cm; we assume $\Delta M/M=1.0$, $C=3.5\times10^5$ cm/sec, $a=8$, $\Theta=375$ K, $B_U+B_N=2.77\times10^{-23}$ sec deg^{-3} at 75 K. After Ref. 8.

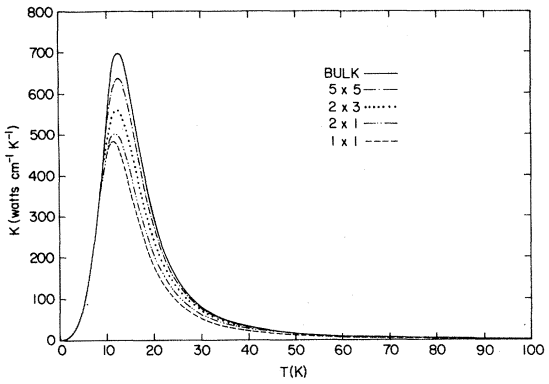

FIG. 3. Temperature dependence of the thermal conductivity for 1×1, 2×1, 2×3, and 5×5 superlattices. The sample size is an order of magnitude larger than in Fig. 2, where $L=1.8$ cm. We assume $\Delta M/M=1.0$.

IV. CONCLUSION

Mini-umklapp three-phonon scattering is predicted to significantly reduce the lattice thermal conductivity of a superlattice below the perfect-crystal value. The reduction is most significant in large samples with small superlattice periods and with large differences between the superlattice layers. We propose that these concepts be tested experimentally.

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[10]A rather complicated and more realistic model can lead to essentially the same result. Suppose we have a simple cubic crystal, but in the x direction this crystal is composed of alternating layers of atoms A and B. There are only nearest-neighbor central forces between atoms, and the interaction potentials between A-A, B-B, and A-B are the same. The only difference between the A and B atoms is their masses. According to Leibfried and Ludwig, in this system the coupling parameters of third order $\Phi^{omn}_{\mu\nu\kappa\, ijk}$ depend on only one independent parameter α_1', and the motion in the x, y, and z directions can be separated.