Theory of interface scattering of phonons in superlattices

S. P. Hepplestone and G. P. Srivastava
School of Physics, University of Exeter, Exeter EX4 4QL, United Kingdom
(Received 16 September 2010; published 12 October 2010)

We present an atomic-scale theory of interface scattering of phonons in superlattices. In particular, we describe the scattering as a result of two features, mixing of atoms at interfaces and presence of dislocations at interfaces due to lattice mismatch. We apply the theory to quantitatively explain the thermal conductivity, and its variation with period and temperature, of Si/Ge superlattices.

DOI: 10.1103/PhysRevB.82.144303 PACS number(s): 63.22.Np, 68.35.−p

I. INTRODUCTION

Superlattices represent a subgroup of metamaterials which allow manipulation of the vibrational characteristics to present new properties. The advantages of using metamaterials for engineering properties of systems has been made clear with a range of applications in photonics,[1,2] phononics,[3,4] and thermoelectrics.[5] One of the most important features of superlattices is the ultralow thermal conductivity along growth direction when compared with either of the two bulk constituents. For thermoelectrics formed from superlattices, a sizeable decrease in the phonon contribution to thermal conductivity leads to an effective increase in their conversion efficiency. For Si/Ge superlattices, there is a drop in thermal conductivity of more than a factor of 100 at room temperature and below.[6] What governs the low thermal conductivity of superlattices is not generally well understood but it is clear that considerations of new scattering mechanisms which rely on the formation of a superlattice are required. Considerable experimental and theoretical challenges exist in understanding such interactions. Mechanisms contributing to this have been proposed for periodically embedded nanodots[7,8] in a host material but these rely on the embedding material being considered as a small perturbation. While such a consideration may be defended for embedded nanodots characterized with three-dimensional interfaces, it cannot be applied to superlattice structures where the interfaces are planar (two dimensional) and each material segment plays an equally important role.

For phonon transport in superlattices, a considerable amount of work has been previously carried out. Among the first clear measurements of the thermal conductivity of semiconductor superlattices were made in 1997 by Lee *et al.*[9] for Si/Ge[001] and in 1999 by Capinski *et al.*[10] for GaAs/AlAs[001]. These and later measurements (which are thoroughly reviewed in Ref. 6) show that the thermal conductivity is clearly dependent on the period of the superlattice, its constituent materials, and the temperature. The explanation for these phenomena has been explored in several theoretical papers going as far back as 1959,[11] but no consensus has been reached.

In 1997 Hyldgaard and Mahan[12] applied the kinetic relaxation time theory to predict the conductivity of superlattices. This highly simplified model showed that the conductivity is reduced by up to an order of magnitude due to the acoustic mismatch. In the method presented, the layers of the superlattice were assumed to have bulklike dispersion and the changes to the relaxation time of the phonon modes due to the superlattice presence were ignored. Due to this and other factors, the model was unable to provide a quantitative prediction of the conductivity for any system. The theoretical problem of superlattice conductivity was explored in 1982 by Ren and Dow.[13] In their early paper, they discussed the effects of zone folding on phonon-phonon interactions and showed that "mini-umklapp" processes were possible and would reduce the conductivity but were unable to quantify further the effect. In 1998 Chen[14,15] provided a detailed explanation for the thermal conductivity of the superlattices based upon simple phonon-dispersion relations and using several parameters. This model was among the first to predict the magnitude correctly but was clearly unable to predict the temperature dependence at low or high temperatures.

In 2000 Volz *et al.*[16] performed a series of molecular-dynamics calculations which showed that the thermal conductivity of Si/Ge superlattices decreased when compared to the bulk systems but were unable to reach the low values measured in experiment. In their paper they hypothesized that interface disorder would be necessary to introduce to account for the difference between theory and experiment. A comprehensive study of the effects of a finite sample size on the phonon group velocities within the superlattices was carried out by Imamura *et al.*[17] in 2002. In their paper they showed that there was a small decrease in phonon group velocities when compared to infinite superlattices but this drop was insufficient to account for the discrepancy between theory and experiment. They also reported the intriguing possibility that introducing the effects of finite size to the modeling of superlattices introduces modes with exceptionally high group velocities but very low transmission probability within the stop bands. Also, Abramson *et al.*[18] showed in 2002 using a molecular-dynamics simulation of Ar and Kr that the effects of strain is to reduce the thermal conductivity (by up to 35%) and hence this feature alone could not explain the drop in the conductivity. In 2003 a detailed review was published by Cahill *et al.*[6] which highlighted several of the experimental and theoretical results to date. In this review, it is clearly shown that with the exception of the research performed by Chen[14,15] the modification of the phonon-scattering rate due to the formation of the superlattice has not been considered within relaxation time theory.

More recent efforts by Broido *et al.*[19,20] has focused upon examining in detail the anharmonic contribution to the lifetime of the phonon modes, commonly described as three-phonon interactions. These authors have shown that consideration of more accurate phonon-dispersion relations than the common linear continuum assumption actually reduces the scattering rate, suggesting a deficit within the existing theory. The latter effort[20] clearly shows that mini-umklapp processes and accurate phonon-dispersion relations reduce the thermal conductivity but not sufficiently to account for the experimental results. Chen *et al.*,[21] using molecular dynamics, show that the thermal conductivity of superlattices has a minimum with period occurring near 8–10 monolayers of atoms. While this is at odds with the results of Capinski[10] it agrees with the earlier results of Lee *et al.*[9] using a four-parameter model. However, the elastic constants and masses used in this model are not comparable with real systems. Also, the authors clearly state that disorder effects at the interface need to be taken into account to produce a better picture of what mechanisms are responsible for the low thermal conductivity of these structures. Koh *et al.*[22] in 2009 carried out thermal conductivity measurements of AlN(4 nm)-GaN(X nm) superlattices, where X was varied. In this paper, using a six-parameter model, the thermal conductivity of these samples was accounted for, but not explained in detail. Also an important factor to note is that these samples contain a relatively small thickness of the AlN layers. In effect the thicker the sample the more GaN-like the phonon-dispersion relations become. Also in 2009, Termentdizis *et al.*[23] investigated, using nonequilibrium molecular-dynamics simulations, the effect of interface roughness in a simple manner by changing its shape from a smooth line to a zigzag. While this may not be the most realistic consideration, it is clearly shown that even such a periodic structured roughness decreases the thermal conductivity of superlattices. However, no comparison with experiment is given. The very recent work by Alvarez *et al.*[24] has provided a simple analytic model for the thermal conductivity of superlattices. While not quantitative, the model shows that ballistic transport within the layers of the superlattice, and the bulklike dispersion in each of these layers is not applicable.

While the above is not a comprehensive review of the literature on phonon transport in superlattice structures, it is apparent that assumptions based upon bulk dispersion relations are not suitable for the thin superlattices and that defects at the interface have a key role to play. In this paper we present an atomic-scale theory for the scattering mechanisms of phonons due to the formation of superlattice interfaces. We show that for periodicities at nanometer scale strong scattering of phonon modes arises due to interface mixing. We also show that for superlattices of midranged periods composed of lattice-mismatched layers, the onset of dislocations at interfaces leads to an initial decrease in the thermal conductivity. However, as the period length increases, the thermal conductivity increases again as the scattering due to the combined effects of both mechanisms decreases. Both of these scattering rates are estimated from the application of Fermi's golden rule and using realistic phonon-dispersion relations for the full spectrum of phonon modes. In the treatment provided here, a two-parameter model is applied to explain the measured thermal conductivities of Si/Ge superlattices.

II. THEORY OF INTERFACE SCATTERINGS

The lifetime of phonon modes in the majority of bulk materials is governed generally by four interaction mechanisms: boundary, electron-phonon, phonon-phonon, and defect scatterings,[25] where defect scattering includes several subcategories such as point, isotope, and extended defect scatterings. Superlattices (and other systems with interfaces) must contain two additional defect-related phonon-scattering mechanisms: interface mass-mixing scattering (IMS) due to diffusion or mixing of atoms at the interfaces and interface dislocation scattering (IDS) which results from dislocations or missing bonds present at interfaces. For studying both of these additional scattering mechanisms, we consider a periodic unreconstructed superlattice A(m)/B(n) with m atomic layers of material A and n layers of material B. The superlattice consists of N_0 unit cells and the total number of atoms in the system can be written as $N+M=N_0(n+m)$.

We express the perturbed Hamiltonian of the superlattice with interface mass mixing for IV-IV semiconductors as

$$H'(\mathrm{IMS}) = \frac{1}{2}\sum_{i=1}^{N}(M_i|\mathbf{v_i}|^2 - M_A|\mathbf{v_A}|^2) + \frac{1}{2}\sum_{i=N+1}^{N+M}(M_i|\mathbf{v_i}|^2 - M_B|\mathbf{v_B}|^2), \quad (1)$$

where M_i is the mass of the ith atom, $\mathbf{v}_i = \frac{d\mathbf{u}_i}{dt}$, where $\mathbf{u_i}$ is the relative displacement of the ith atom with respect to its neighbors and t is time. M_A and M_B are isotopic average masses of materials A and B. The first/second term represents all the atoms in the region A/B layers of the superlattice. For superlattices made of III-V materials, additional terms must be added to Eq. (1) to account for the two species in each component material. However, as seen later, these terms become superfluous.

For dealing with IDS at the atomic scale, we consider a dislocation as a series of randomly missing bonds located near the interface within a unit cell. From this one can write the perturbed Hamiltonian as

$$H'(\mathrm{IDS}) = \frac{1}{2}\sum_{i=1}^{N'}(K_0|\mathbf{u_i}|^2 - K_A|\mathbf{u_A}|^2) + \frac{1}{2}\sum_{i=N'+1}^{N'+M'}(K_0|\mathbf{u_i}|^2 - K_B|\mathbf{u_B}|^2), \quad (2)$$

where u_i as the relative displacement between the two neighboring atoms and N'/M' is number of interatomic bonds in material segments A/B. Also, $K_A(K_B)$ represents the interatomic spring constant in the layer A(B) and K_0 represents a spring constant in the dislocation region, i.e., has a value equal to zero, or close to zero, for broken or missing bonds. We assume $K_A \approx K_B$ as the difference between the two values

is insignificant for dealing with IDS and consider a random distribution and orientation of the broken bonds within each superlattice plane (perpendicular to the growth direction). Again, for III-V systems, additional terms are introduced, but like the additional terms for IMS, these become unnecessary.

We apply Fermi's golden rule[26] to obtain the probability of a phonon mode $\omega(\mathbf{q}s)$ being scattered into mode $\omega(\mathbf{q}'s')$ for each scattering mechanism. For IMS we obtain the probability $P_{\mathbf{q}s}^{\mathbf{q}'s'}$ of a phonon mode $\omega(\mathbf{q}s)$ being scattered into mode $\omega(\mathbf{q}'s')$ as

$$P_{\mathbf{q}s}^{\mathbf{q}'s'} = \frac{\hbar\pi}{2N_0^2(n+m)^2}\omega(\mathbf{q}s)\omega(\mathbf{q}'s')\bar{n}_{\mathbf{q}s}(\bar{n}_{\mathbf{q}'s'}+1)\left\{\sum_{i=1,j=1}^{n,n}(e_ie_j'-e_Ae_A')e^{i[(\mathbf{q}-\mathbf{q}')\cdot(\mathbf{r_i}-\mathbf{r_j})]}+\sum_{i=1,j=n+1}^{n,n+m}(e_ie_j'-e_Ae_B')e^{i[(\mathbf{q}-\mathbf{q}')\cdot(\mathbf{r_i}-\mathbf{r_j})]}\right.$$
$$\left.+\sum_{i=n+1,j=1}^{n+m,n}(e_ie_j'-e_Be_A')e^{i[(\mathbf{q}-\mathbf{q}')\cdot(\mathbf{r_i}-\mathbf{r_j})]}+\sum_{i=n+1,j=n+1}^{n+m,n+m}(e_ie_j'-e_Be_B')e^{i[(\mathbf{q}-\mathbf{q}')\cdot(\mathbf{r_i}-\mathbf{r_j})]}\right\}, \tag{3}$$

where $\bar{n}_{\mathbf{q}s}$ is the Bose-Einstein distribution of phonon mode $\omega(\mathbf{q}s)$ at temperature T and e_i is the amplitude of ith atom. Similarly for IDS we obtain the corresponding probability $Q_{\mathbf{q}s}^{\mathbf{q}'s'}$ as

$$Q_{\mathbf{q}s}^{\mathbf{q}'s'} = \frac{\hbar\pi}{2N_0^2(n+m)^2}\frac{\bar{n}_{\mathbf{q}s}(\bar{n}_{\mathbf{q}'s'}+1)}{\omega(\mathbf{q}s)\omega(\mathbf{q}'s')}\sum_{i=1,j=1}^{n+m,n+m}(e_ie_j')\frac{K_0^2}{M_iM_j}. \tag{4}$$

Equations (3) and (4) can be simplified by assuming that only J layers are affected at the interface, thus reducing the number of terms. Finally, by assuming that in the in-plane directions the broken bonds and mass substitutions are random, we can eliminate the off-diagonal components. Thus from the above, it is possible to calculate the phonon relaxation rate for these two mechanisms. For IMS, we derive the following expression:

$$\tau_{\mathrm{IMS}}^{-1}(\mathbf{q}s) = \frac{\alpha\pi}{2N_0(n+m)^2}\times\int d\omega(\mathbf{q}'s')g[\omega(\mathbf{q}'s')]\omega(\mathbf{q}s)\omega(\mathbf{q}'s')\times\frac{\bar{n}(\mathbf{q}'s')+1}{\bar{n}(\mathbf{q}s)+1}\delta[\omega(\mathbf{q}s)-\omega(\mathbf{q}'s')]\times\left[\left(1-\frac{e_Ae_A'}{e_Be_B'}\right)^2+\left(1-\frac{e_Be_B'}{e_Ae_A'}\right)^2\right], \tag{5}$$

where α is regarded as a parameter related to the amount of mixing at the interface (discussed later), $g[\omega(\mathbf{q}s)]$ is the density of states, and e_B/e_A is the ratio of the amplitudes of eigenvectors in materials B and A. Similarly, the relaxation rate due to IDS may be written as

$$\tau_{\mathrm{IDS}}^{-1}(\mathbf{q}s) = \frac{\pi\omega_0^4}{4N_0}\frac{\alpha'}{(n+m)^2}\int d\omega(\mathbf{q}'s')\frac{g[\omega(\mathbf{q}'s')]}{\omega(\mathbf{q}s)\omega(\mathbf{q}'s')}\times\frac{\bar{n}(\mathbf{q}'s')+1}{\bar{n}(\mathbf{q}s)+1}\delta[\omega(\mathbf{q}s)-\omega(\mathbf{q}'s')]\times\left[1+\left(\frac{e_Ae_A'}{e_Be_B'}\right)^2+1+\left(\frac{e_Be_B'}{e_Ae_A'}\right)^2\right], \tag{6}$$

where α' is a measure of dislocation concentration (discussed later), ω_0 can be approximated as the highest zone-center frequency, and the rest of the symbols have their usual meaning. From the above results it is clear that the relaxation rates due to the IMS and IDS depend on several factors: the amplitude ratio e_B/e_A, phonon frequency, superlattice period, and the parameters α and α'. We will discuss these factors below.

The amplitude ratio e_B/e_A, required for both scattering mechanisms, can be approximated, using the diatomic linear chain model along the superlattice growth direction, as

$$\frac{e_B}{e_A} = \frac{\left[\frac{1}{M_0}-\Delta\left(\frac{1}{M}\right)\right]\cos(l_zq_z)}{\left\{\left(\frac{1}{M_0}\right)^2\cos^2(l_zq_z)+\left[\Delta\left(\frac{1}{M}\right)\right]^2\sin^2(l_zq_z)\right\}^{1/2}-\Delta\left(\frac{1}{M}\right)}, \tag{7}$$

with $M_0=\frac{1}{2}(1/M_A+1/M_B)$, $\Delta(1/M)=\frac{1}{2}(1/M_A-1/M_B)$, and l_z as the period along the superlattice growth direction. The ratio of amplitudes increases in magnitude with increasing wave vector, q_z. This leads to the expected result that the interface will weakly scatter long-wavelength modes. When the ratio of amplitudes is very large, then the two layers in each superlattice unit cell act out of phase and hence traveling phonons are scattered more strongly by the presence of the interfaces.

For both IMS and IDS scattering mechanisms optical modes in superlattices will be scattered at the interface more strongly than acoustic modes. Also, as the frequency dependence of IMS is stronger than that of IDS, acoustic modes are more strongly scattered by IMS. Sharp peaks in the density of states in the optical range, due to strongly confined and thus much reduced velocities, result in the scattering rate of optical modes being greater than for their acoustic counterparts. Also, for optical modes, the energy conservation condition implied in Eqs. (5) and (6), coupled with the flatness of the branches, enables these modes to undergo considerably more interactions than the dispersive acoustic modes, resulting in an increased scattering rate for these modes. The frequency dependence of long-wavelength acoustic modes compares favorably with existing similar scattering mechanisms developed within the continuum regime for bulk.[27–29] However, the atomistic treatment presented here is both more appropriate for ultrathin superlattices and also leads to the consideration of the amplitude ratio in Eq. (7), which modifies the dependence of IDS and IMS from the simple frequency power laws that one considers in bulk materials.

The factor $1/(n+m)^2$ in Eqs. (5) and (6) implies that the strengths of these scattering rates are greatest for thin superlattices and decrease as the superlattices become thicker. This is an important result, as both in the case where the period of superlattices tends to infinity and also in the case where the atomic masses in the layers A and B become equal, these interaction strengths become zero. This correctly ensures that these scattering mechanisms would not exist in bulk systems.

The parameter α in Eq. (6) gives an indication of the amount of mixing which occurs at the interface and is also proportional to the interface area (normal to superlattice growth direction). This parameter is expected to increase with increasing period but not by a dramatic amount. The parameter α' in Eq. (6) indicates the number of dislocations and distortions of the crystal at the interfaces and is also proportional to interface area. The ratio of α and α' gives a clear indication of the interface quality and dominant type of disorder present. Both these parameters also take into account the number of layers over which the interface effects are spread. While this can be introduced separately it is an additional parameter that is not easy to predict and so it is merged into α and α'. In the approach given here, these are the only two adjusted/free parameters used to fit the experimental data.

III. THERMAL CONDUCTIVITY

In order to evaluate the effectiveness of the interface scattering term in describing real systems we now apply the theory developed here to explain the low-temperature thermal conductivity of two Si/Ge superlattices, namely, the Si(19)/Ge(5)[001] and Si(72)/Ge(30)[001] superlattices fabricated and measured by Lee *et al.*[9] The thermal conductivity of a superlattice is an anisotropic quantity, requiring a full calculation of the corresponding dispersion relations of the system. The expression for the thermal conductivity tensor

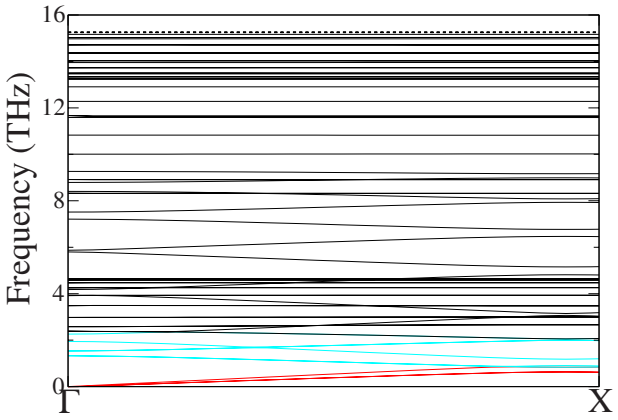

FIG. 1. (Color online) The phonon-dispersion curves for the Si(19)/Ge(5)[001] superlattice along the growth direction. For clarity, the three acoustic branches are highlighted in gray (red online), the folded acoustic in gray (blue online) , and the highest optical is presented as a black dashed line.

for a superlattice within the single-mode relaxation-time approximation can be expressed as

$$\kappa_{ij} = \frac{\hbar^2}{N_0 \Omega_0 k_B T^2} \sum_{\mathbf{q},s} \omega_{\mathbf{q}s}^2 v_{\mathbf{q}s,i} v_{\mathbf{q}s,j} \tau_{\mathbf{q}s} \bar{n}_{\mathbf{q}s} (\bar{n}_{\mathbf{q}s} + 1), \qquad (8)$$

where Ω_0 is the volume of the unit cell, k_B is the Boltzmann constant, $v_{\mathbf{q}s,i}$ is the ith component of the velocity of the phonon mode $(\mathbf{q}s)$, and $\tau_{\mathbf{q}s}$ is the total the lifetime of the phonon: $\tau^{-1} = \tau_{\mathrm{IMS}}^{-1} + \tau_{\mathrm{IDS}}^{-1} + \tau_{\mathrm{MD}}^{-1} + \tau_B^{-1}$ with τ_B^{-1} (boundary scattering[30]) and τ_{MD}^{-1} (isotope scattering[27]) taking their usual forms. The boundary length is taken from experiment as 0.1 μm (Ref. 9) to calculate τ_B^{-1} and the known isotopic ratios for Si and Ge are used to evaluate τ_{MD}^{-1}. The integrations required in Eqs. (6)–(8) were evaluated by employing the special-$\mathbf{q}$ points scheme.[31] In the low-temperature regime, the effect of three-phonon scattering is negligible and can be discarded. However, a more complete form for three-phonon scattering in superlattices and metamaterials has been discussed previously.[32]

The calculation of the scattering of phonon modes in superlattices requires an extensive knowledge of phonon-dispersion curves for the unperturbed structures. Previously we developed an extended bond charge model,[33,34] which accurately described the phonon gaps in a Si/SiGe semiconductor superlattice observed experimentally in Ref. 35. With the application of that model we present in Fig. 1 the full phonon-dispersion relations for the Si(19)/Ge(5)[001] superlattice along the growth direction. Superlattice phonon-dispersion relations, like those shown in Fig. 1, are characterized by shallow dispersive branches, flat confined branches (dispersionless modes at frequencies greater than 9 GHz), band splitting at the zone center and zone edge, known as polarization gaps. Also, as the period increases, due to zone folding, the number of bands increases and the frequency of the lowest zone-center folded mode decreases. Development of disallowed frequency regions along the superlattice growth direction has a direct effect on phonon interactions.

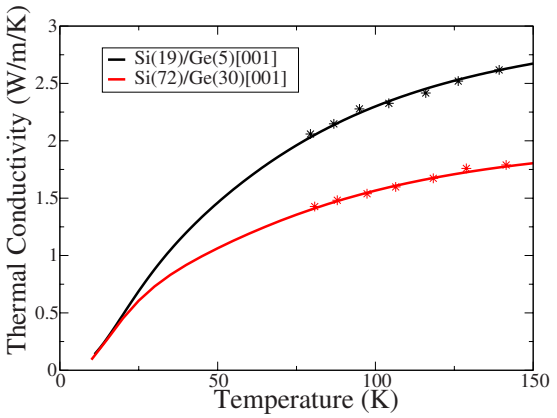

FIG. 2. (Color online) The thermal conductivity, κ_{ZZ}, of two Si/Ge superlattices in the growth direction as a function of temperature. The lines are the theoretical fits and stars show the measured results from Lee *et al.* (Ref. 9).

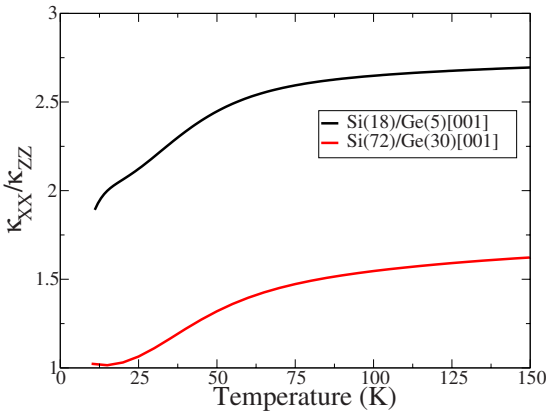

FIG. 3. (Color online) The thermal conductivity ratio κ_{XX}/κ_{ZZ} of two Si/Ge superlattices as a function of temperature.

From Fig. 2 it is clear that our calculated results for κ_{ZZ} agree well with the measured results.[9] In additional to the standard scattering mechanisms, from boundary and isotopic mass defects, it was found that throughout the temperature range considered, the dominant scattering mechanisms are IMS and IDS as presented in Eqs. (5) and (6). The important feature to consider is the ratio of the IDS and IMS scattering strengths. In the thinner superlattice, for the majority of phonon modes across the frequency spectrum, IMS is the dominant scattering mechanism and the contribution of IDS is negligible ($\alpha \approx 550$ and $\alpha' \approx 0.0$). This suggests that the relative number of dislocations is small and the quality of the interfaces between the layers of the thinner superlattice in Ref. 9 is relatively clean. In contrast, for the thicker superlattice, the dominant scattering mechanism is IDS, indicating that the interfaces have several broken or distorted bonds (as $\alpha \approx 10^7$ and $\alpha' \approx 10^{-4}$).

The interplay between the IMS and IDS in controlling the phonon lifetime in such superlattices explains the apparent dip in the thermal conductivity measured by Lee *et al.* as a function of period. The thinner sample can grow as a stable crystal structure with relaxed interatomic bonds, without the interface acquiring many dislocations or missing bonds, consistent with experimental investigations.[36] Thus for these systems the IDS contribution is very weak. For samples with a larger period, each of the alternating material layers mimics its individual bulk lattice constant at the cost of increase in the strain at the interface due to lattice mismatch, thus leading to a greater amount of dislocations and other defects or imperfections. Hence, when compared to thinner samples, the amount of phonon scattering by IDS is much greater, but IMS is relatively the same. This scenario is also consistent with the thermal conductivity results obtained by Capinski *et al.*[10] for GaAs/AlAs[001] superlattices (composed of nearly latticed-matched GaAs and AlAs layers) of various periods. Due to very little lattice mismatch, the number of dislocations in the GaAs/AlAs superlattices is very low, and thus for all samples studied by Capinski *et al.* in the low-temperature region, the scattering will be dominated by the IMS mechanism. However, to fully discuss the results presented by Capinski *et al.* requires inclusion of anharmonic effects.

As expected, our numerical results show clear anisotropic behavior of the thermal conductivity of the superlattice structures studied here. As shown in Fig. 3, at low temperatures, for the thicker superlattices the thermal conductivity ratio κ_{XX}/κ_{ZZ} tends to unity whereas for the thinner superlattice the ratio remains much greater than unity. At low temperatures, only low-lying modes are populated. Thus, in the thicker superlattice the large amounts of zone folding means that at low temperatures phonon modes both at the zone center and the zone edge are populated. Conversely, for thinner superlattices with less zone folding, the zone edge modes are not populated until higher temperatures. One can see, in accordance with Eq. (7), that zone-edge modes are scattered more strongly than zone-center modes, making both IDS and IMS highly anisotropic scattering mechanisms at low temperatures for thinner superlattices, but less so for thicker superlattices.

The scattering mechanisms presented here contain all of the necessary ingredients for relaxation-time theory for such structures as more detailed or additional scattering mechanisms would require additional parameters to be introduced. While strain could be considered as an additional mechanism, we may regard it being incorporated into the IDS scattering by treating α' as an adjustable parameter (as the amount of strain would depend on the mixing at the interfaces). In principle, the strain field will manifest itself as a change in the elastic constants and hence would be difficult to distinguish from IDS. Similarly, the effect of alien species of atoms in the superlattice may be considered included in the parameter α, without requiring the use of an additional parameter as the form of such a mechanism is similar to that of IMS. Hence, our model represents the limitation of relaxation-time theory for superlattices at low temperatures by constraining itself to only two parameters.

IV. SUMMARY

In summary, we have presented an atomic-scale theory of phonon scattering in metamaterials due to atomic mixing and dislocations at interfaces. A successful application of the theory to explain the measured low-temperature thermal con-

ductivity of Si/Ge superlattices[9] has revealed information regarding the relative importance of these two scattering mechanisms, and their joint influence in determining the conductivity along the growth direction. We have shown that, using a simple two-parameter model, the onset of the two scattering mechanisms, inherent to superlattice growth procedure, reduces the conductivity of the superlattices when compared to that of their bulk counterparts, or an alloy of their two constituents materials. Our calculations at 100 K show $1<\kappa_{XX}/\kappa_{ZZ}<3$ with κ_{ZZ} being over two orders of magnitude smaller than the average result for bulk samples of Si and Ge. Our results also show the unsurprising result that the interface quality in thin superlattices is much superior to that in thicker superlattices. This effect accounts for the observed dip in the thermal conductivity of the Si/Ge superlattices as a function of period. We believe that an understanding of the two scattering mechanisms discussed in this work will enable a good understanding of the effects of interfaces in superlattices and will provide a clear route to understanding their low thermal conductivity when compared to either their bulk constituents or their alloys. We hope that relevant modification of the present treatment of interface scattering of phonons in superlattices will help elucidate transport measurements in metamaterials in general.

ACKNOWLEDGMENTS

This work has been carried out with financial support from the Leverhulme Trust (U.K.) through Grant No. F/00 144/AS.

[1] *Metamaterials: Physics and Engineering Explorations*, edited by N. Engheta and R. W. Ziolkowski (Wiley-IEEE Press, New York, 2006).
[2] S. C. Kitson, W. L. Barnes, and J. R. Sambles, Phys. Rev. Lett. **77**, 2670 (1996).
[3] M. Sigalas and E. N. Economou, J. Sound Vib. **158**, 377 (1992).
[4] M. S. Kushwaha, P. Halevi, L. Dobrzynski, and B. Djafari-Rouhani, Phys. Rev. Lett. **71**, 2022 (1993).
[5] H. Bottrier, C. Chen, and R. Venkatasubramanium, MRS Bull. **31**, 211 (2006).
[6] D. G. Cahill, W. K. Ford, K. E. Goodson, G. D. Mahan, A. Majumdar, H. J. Maris, R. Merlin, and S. R. Phillpot, J. Appl. Phys. **93**, 793 (2003).
[7] W. Kim and A. Majumdar, J. Appl. Phys. **99**, 084306 (2006).
[8] J.-N. Gillet, Y. Chalopin, and S. Volz, J. Heat Transfer **131**, 043206 (2009).
[9] S. M. Lee, D. G. Cahill, and R. Vekatasubramanian, Appl. Phys. Lett. **70**, 2957 (1997).
[10] W. S. Capinski, H. J. Maris, T. Ruf, M. Cardona, K. Ploog, and D. S. Katzer, Phys. Rev. B **59**, 8105 (1999).
[11] W. A. Little, Can. J. Phys. **37**, 334 (1959).
[12] P. Hyldgaard and G. D. Mahan, Phys. Rev. B **56**, 10754 (1997).
[13] S. Y. Ren and J. D. Dow, Phys. Rev. B **25**, 3750 (1982).
[14] G. Chen, Phys. Rev. B **57**, 14958 (1998).
[15] G. Chen, T. Zeng, T. Borca-Tasciuc, and D. Song, Mater. Sci. Eng., A **292**, 155 (2000).
[16] S. Volz, J. B. Saulnier, G. Chen, and P. Beachamp, Microelectron. J. **31**, 815 (2000).
[17] K. Imamura, Y. Tanaka, and S. Tamura, Phys. Rev. B **65**, 174301 (2002).
[18] A. R. Abramson, C.-L. Tien, and A. Majumdar, J. Heat Transfer **124**, 963 (2002).
[19] D. A. Broido and T. L. Reinecke, Phys. Rev. B **70**, 081310 (2004).
[20] A. Ward and D. A. Broido, Phys. Rev. B **77**, 245328 (2008).
[21] Y. Chen, D. Li, J. R. Lukes, Z. Ni, and M. Chen, Phys. Rev. B **72**, 174302 (2005).
[22] Y. K. Koh, Y. Cao, D. G. Cahill, and D. Jena, Adv. Funct. Mater. **19**, 610 (2009).
[23] K. Termentzidis, P. Chantrenne, and P. Keblinski, Phys. Rev. B **79**, 214307 (2009).
[24] F. X. Alvarez, J. Alvarez-Quintana, D. Jou, and J. Rodriguez Viejo, J. Appl. Phys. **107**, 084303 (2010).
[25] G. P. Srivastava, in *The Physics of Phonons* (Adam Hilger IOP, Bristol, 1990).
[26] P. Carruthers, Rev. Mod. Phys. **33**, 92 (1961).
[27] P. G. Klemens, Can. J. Phys. **35**, 441 (1957).
[28] W. Fon, K. C. Schwab, J. M. Worlock, and M. L. Roukes, Phys. Rev. B **66**, 045302 (2002).
[29] J. E. Graebner, M. E. Reiss, L. Seibles, T. M. Hartnett, R. P. Miller, and C. J. Robinson, Phys. Rev. B **50**, 3702 (1994).
[30] J. M. Ziman, *Electrons and Phonons* (Clarendon, Oxford, 1960).
[31] H. J. Monkhorst and J. D. Pack, Phys. Rev. B **13**, 5188 (1976).
[32] S. P. Hepplestone and G. P. Srivastava, MRS Spring Meeting **1172E**, 1172–T03–09 (2009).
[33] S. P. Hepplestone and G. P. Srivastava, Phys. Rev. Lett. **101**, 105502 (2008).
[34] S. P. Hepplestone and G. P. Srivastava, J. Appl. Phys. **107**, 043504 (2010).
[35] Y. Ezzahri, S. Grauby, J. M. Rampnoux, H. Michel, G. Pernot, W. Claeys, S. Dilhaire, C. Rossignol, G. Zeng, and A. Shakouri, Phys. Rev. B **75**, 195309 (2007).
[36] Y.-W. Mo, D. E. Savage, B. S. Swartzentruber, and M. G. Lagally, Phys. Rev. Lett. **65**, 1020 (1990); T. Ma, H. Tu, B. Shao, A. Liu, and G. Hu, Mater. Sci. Semicond. Process. **9**, 49 (2006).