Vol 13 No 12, December 2004
1009-1963/2004/13(12)/2109-06

Chinese Physics

© 2004 Chin. Phys. Soc.
and IOP Publishing Ltd

Thermal conductivity of GaAs/AlAs superlattices: The Umklapp process*

Yang Yu-Rong(杨玉荣)[a], Yan Xiao-Hong(颜晓红)[a)b)c)†], Cao Jue-Xian(曹觉先)[a],
Xiao Yang(肖 杨)[a], Mao Yu-Liang(毛宇亮)[a],
Xiang Jun(向 君)[a], and Yu Hai-Lin(于海林)[a]

[a] *Department of Physics, Xiangtan University, Xiangtan* 411105, *China*
[b] *Interdisciplinary Centre of Theoretical Studies and Institute of Theoretical Physics, Chinese Academy of Sciences, Beijing* 100080, *China*
[c] *College of Science, Nanjing University of Aeronautics and Astronautics, Nanjing* 210016, *China*

(Received 19 March 2004; revised manuscript received 14 July 2004)

The thermal conductivity of GaAs/AlAs superlattices limited by the three-phonon Umklapp process and boundary scattering has been studied theoretically based on the model of lattice dynamics with force constant matrix. It was found that the Umklapp relaxation rate approximates $BT\omega^2/n$ with a fitting parameter B. The thermal conductivity increases with the increase of temperature at low temperatures, and would show a peak behaviour at about 60K before falling off at high temperatures. In addition, the thermal conductivity increases with the increase of period thickness of the superlattices.

Keywords: thermal conductivity, superlattices, three-phonon Umklapp process
PACC: 6670, 6322, 6320H

1. Introduction

The thermal conductivity of a semiconductor superlattice is of great importance for the performance of thermoelectric coolers and power converters[1−7] as well as optoelectronic devices.[8−13] Low thermal conductivity materials are desired for thermoelectric elements in order to reduce the backflow of heat from the hot to cold junction, while high conductivity materials are necessary for optoelectronic devices in order to dissipate heat. Recently, experimental results on the thermal conductivity of superlattices have been reported for several material systems. The thermal conductivity both parallel[14,15] and perpendicular[16−20] to the interface is found significantly reduced in comparison to their corresponding bulk values. Several models have been developed to explain these findings.

Ren and Dow[21] modelled the thermal conductivity of ideal superlattice structures by combining Callaway's model[22] with a quantum mechanical treatment of the scattering rate. The predicted reduction in thermal conductivity from their model, however, is too low to explain the orders of magnitude reduction in the measured thermal conductivity of semiconductor superlattices. Chen *et al*[23] attributed the large cross-plane thermal conductivity reduction in superlattices to the phonon interface reflection and diffuse scattering, based on the solution of the Boltzmann transport equation. An alternative approach was taken by Hyldgaard[3] and Tamura[24] based on the lattice dynamic calculation of the phonon dispersion in superlattices. They explained that the thermal conductivity is reduced because the phonon group velocity is reduced due to zone folding.

Although a variety of mechanisms for thermal conductivity of superlattices have been developed, it is necessary to describe the phonon characteristic for understanding the physical essence of thermal transport. In this paper, considering the three-phonon Umklapp process, we analyse the physical mechanism of the thermal transport perpendicular to the layers

*Project supported by the Major Programme of Education Bureau of Hunan Province, China (Grant No 03A046), the Science Foundation of Science and Technology Department of Hunan Province, China (Grant No 03JZY3019), and by the Natural Science Foundation of Hunan Province, China (Grant No 03JJY3010).
†E-mail: xhyan@xtu.edu.cn

in GaAs/AlAs superlattices.

2. Principal Theory

Considering the fcc model,[24,25] the superlattice in our study consists of lattice GaAs (A) and lattice AlAs (B) packed alternately along [001] direction as shown in Fig.1. The thickness of lattice A (B) is $d_A = n_A a$ ($d_B = n_B a$), where n_A (n_B) is the number of atomic layers and period thickness is $D = d_A + d_B$. We let $\boldsymbol{u}_{lmn}$ be the displacement of an atom from its equilibrium position $\boldsymbol{r}_{lmn} = (\boldsymbol{x}_{lm}, z_n) = (la, ma, na)$ in one unit period lattice. Based on the force constant model, the dynamic equation for the displacement of the ith atom in the unit period can be written as

$$M_i \ddot{\boldsymbol{u}}_i = \sum_j K^{(i,j)}(\boldsymbol{u}_j - \boldsymbol{u}_i), \ (i = 1, \ldots, n), \qquad (1)$$

where M_i is the mass of the ith atom and $K^{(i,j)}$ represents the 3×3 force constant tensor between the ith and the jth atoms. The sum over j in Eq.(1) is taken over the nearest neighbour sites of the ith atom. The vibrational wave functions of lattice vibration are often taken to be

$$\boldsymbol{u}_i = \boldsymbol{U}_i \exp[\mathrm{i}(\boldsymbol{k}_i \cdot \boldsymbol{r}_i - \omega t)], \qquad (2)$$

where $\boldsymbol{U}_i$ is the amplitude of the ith atom in one unit period of superlattice, and $\boldsymbol{k} = (k_x, k_y, k_z)$ is the wave vector. Substituting wave function (2) into the dynamic equation (1), one can obtain

$$\begin{pmatrix} B & A_1 & 0 & 0 & A_{-1} \\ A_{-1} & B & A_1 & 0 & 0 \\ 0 & A_{-1} & B & A_1 & 0 \\ 0 & 0 & \ddots & \ddots & \vdots \\ A_1 & 0 & 0 & A_{-1} & B \end{pmatrix} \begin{pmatrix} \boldsymbol{U}_1 \\ \boldsymbol{U}_2 \\ \boldsymbol{U}_3 \\ \vdots \\ \boldsymbol{U}_n \end{pmatrix} = 0, \qquad (3)$$

where

$$B = M_i \omega^2 I - 2K \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 2 \end{pmatrix},$$

$$A_1 = K \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 2 \end{pmatrix} \exp(\mathrm{i}k_z a),$$

$$A_{-1} = K \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 2 \end{pmatrix} \exp(-\mathrm{i}k_z a).$$

The dispersion relation for phonons in the cross-plane direction of the superlattices can be determined by solving the equation of the dynamical matrix.

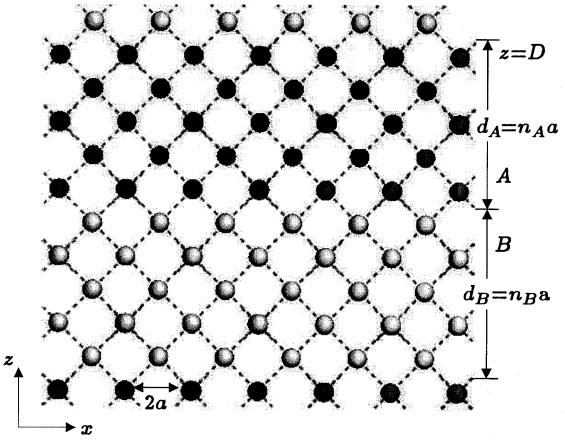


Fig.1. Structure of unit period in the ($n_A \times n_B$) GaAs/AlAs superlattice consisting of two fcc lattices.

As heat in superlattices is mostly carried by phonons, it is reasonable to neglect electronic component of thermal conductivity.[23] Based on the dispersion relation, the lattice thermal conductivity κ at a given temperature T is given by the formula[24,26]

$$\kappa(T) = \sum\nolimits_\lambda \tau_\lambda(T) C_\lambda(T) V_{g,\lambda}^2, \qquad (4)$$

$$C_\lambda(T) = \frac{(\hbar\omega)^2}{k_\mathrm{b} T^2} \frac{\exp(\hbar\omega/k_\mathrm{b}T)}{[\exp(\hbar\omega/k_\mathrm{b}T) - 1]^2}, \qquad (5)$$

$$V_{g,\lambda} = \partial\omega_\lambda / \partial q, \qquad (6)$$

where τ_λ, C_λ, and $V_{g,\lambda}$ are the phonon relaxation time, specific heat, and phonon group velocity of a given phonon state λ, respectively. In superlattices, the phonon relaxation time is mainly controlled by boundary scattering and the three-phonon Umklapp scattering process. The total phonon relaxation time τ is usually given by the Mathiessen rules[27]

$$\frac{1}{\tau} = \frac{1}{\tau_\mathrm{U}} + \frac{1}{\tau_\mathrm{B}}, \qquad (7)$$

with the relaxation time parameters for the three-phonon Umklapp scattering process τ_U and for boundary scattering τ_B.

From the first-order perturbation theory, the relaxation time τ_U in the three-phonon Umklapp process for thermal modes q is given by [28]

$$\frac{1}{\tau_\mathrm{U}} = \sum_{q'} 2 |C_3|^2 \frac{\hbar}{M^3 \omega\omega'\omega''} \pi\delta(\omega + \omega' - \omega'') \times (N_0(\omega') - N_0(\omega'')), \qquad (8)$$

where $|C_3|^2 = (4\gamma^2/3n_a)(M^2/V_g^2)\omega^2\omega'^2\omega''^2$, $\tau_{\rm U}$ is the relaxation time, γ is the Gruneisen parameter, n_a is the number of atoms per unit volume, M is the atom mass. $N_0(\omega')$ and $N_0(\omega'')$ are the equilibrium occupancies of phonon modes q' and q'', respectively. The equilibrium occupancy is given by the Planck distribution $N_0 = 1/[\exp(\hbar\omega/k_{\rm B}T)-1]$. The evaluation of the relaxation rate of the mode q requires the integration over all possible q' modes. The frequencies ω, ω', ω'', and the phonon vector q, q', q'' of phonon modes are connected by the energy and momentum conservation laws expressed as

$$\omega + \omega' = \omega'', \tag{9}$$

$$q + q' = q'' + G, \tag{10}$$

where G is the reciprocal-lattice vector. The restrictions followed from the selection rules require that (I) all three interacting modes cannot belong to the same polarization branch, and (II) the resultant mode should be above the two initial (interacting) modes.[29] It is known that for isotropic semiconductors only two general types of channels are allowed: TA+TA⇔LA or LA+TA⇔LA.[30] In our calculation for GaAs/AlAs superlattices, all phonon branches, including acoustic phonons, and optical phonons, are considered. So, much more possible three-phonon anharmonic decay channels are allowed than those in isotropic semiconductors.

The relaxation time for boundary scattering can be evaluated from the semiempirical relation[22,31]

$$\frac{1}{\tau_{\rm B}} = \frac{V_g}{D}, \tag{11}$$

where D is the characteristic thickness of sublattice GaAs (AlAs) in a unit period of GaAs/AlAs superlattices. More precise description of the boundary scattering can be obtained using the approach of Ref.[23], which takes into account the effects induced by partially diffuse and partially specular interfaces. Equation (8) was derived for bulk and cannot be applied for strictly 2D systems since in this case a reduced zone picture and the Umklapp formalism do not work in the growth direction. In our calculations we will consider a rather thick periodic structure (many atomic layers) so that the formula can still be applied.

3. Numerical results and discussion

In virtue of the parameters taken from Ref.[24], we calculate the phonon dispersion relation in the cross-plane direction of GaAs/AlAs superlattices, as shown in Fig.2. The first three modes of the phonon dispersion relation around Γ point in Fig.2 are acoustic modes, including the doubly degenerated transverse acoustic (TA) mode and the nondegenerate longitudinal acoustic (LA) mode. The curves of the three phonon acoustic modes rise linearly from Γ point to near the X point. As for the remaining optical branches, the roughly linear relations of the phonon frequency versus wave vector are obtained except for the zone near the centre and the boundary. At the centre and the boundary of the folded Brillouin zone, the phonon dispersion curves are flattened because of the zone folding effects. In addition, the band-frequency gaps occur at the centre and the boundary. The band-gap width would have an important influence on the occurrence of the Umklapp process.

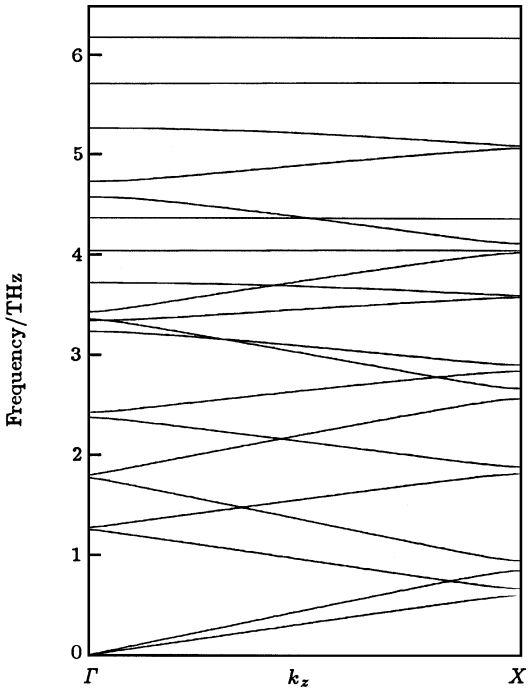


Fig.2. Dispersion relation of phonons in the cross-plane direction of the (5×5) GaAs/AlAs superlattice.

Figure 3 shows the phonon group velocity of (5×5) GaAs/AlAs superlattices, in which the dashed curve and the dotted curve represent the phonon group velocities of the LA mode and TA mode respectively. Both the curves are almost horizontal from the Γ point to near the X point, then rapidly decrease to zero. The remaining solid curves represent the phonon group velocity of the optical modes. Obviously, all of these curves decrease to zero near the centre and the boundary of Brillouin zone because the corresponding phonon dispersion curves are flattened at the centre

and the boundary. It can be seen that the group velocity in the superlattices is reduced at the zone centre and the boundary.

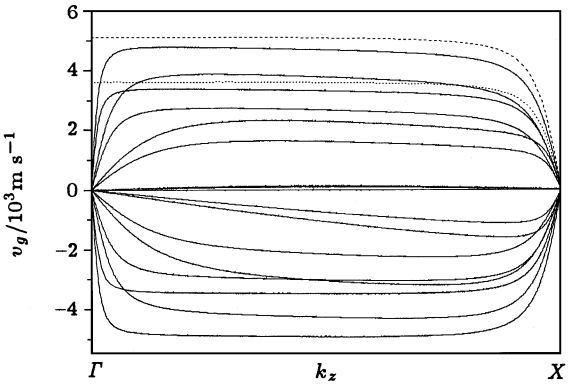


Fig.3. The group velocity of the(5 × 5) GaAs/AlAs superlattice.

In order to obtain the relaxation rate for the three-phonon Umklapp process that goes through all possible channels,[32] it is necessary to substitute the dispersion relation and group velocity calculated for the relevant range of q in Eq.(8), and then sum all together. As an example, we present the Umklapp relaxation rate for mode TA in Fig.4. The relaxation rates for (5 × 5) GaAs/AlAs superlattices at 50, 150, 250, 350, 450 and 550K are shown in Fig.4(a). It is found that the relaxation rates are zero at low frequency at different temperatures. It suggests that phonons with low frequency would transfer comparatively freely in superlattices without any contribution to τ_U^{-1} except to surface scattering. As the phonon frequency increases, the relaxation rates increase and approximate ω^2 as a function of phonon frequency at different temperatures. However, the relaxation rates would break the quadratic dependence and tend to infinity for phonon frequency higher than 12×10^{12} rad s^{-1}. This is because the phonon group velocity decreases to zero near the boundary of the folded Brillouin zone. In addition, the relaxation rates are approximately zero over most frequencies at 50K. The relaxation rates increase linearly with the increase of temperature at phonon frequency $\omega = 2.6 \times 10^{12}$ rad s^{-1}, as shown in the inset of Fig.4(a). These results suggest that the Umklapp scattering freezes out at low temperatures. As temperature increases, phonons with high frequency are thermally populated to dominate the Umklapp process, which leads to the increase of relaxation rates. The relaxation rates at 300K for (n × n) ($n = 1 - 10$) GaAs/AlAs superlattices are given in Fig.4(b). As the number of monolayers n in a period increases, τ_U^{-1} decreases. This means that as the period thickness increases, the probability of phonon scattering decreases. The inset of Fig.4(b) shows that the relaxation rates are inversely proportional to the number n. Consequently, the relaxation rate can be fitted with the formula

$$\tau_U^{-1} = B\omega^2 T/n, \tag{12}$$

where B is a fitting parameter. The formula provides a precise and direct estimate of the relative strengths of the Umklapp phonon scattering mechanism and the thermal resistance.

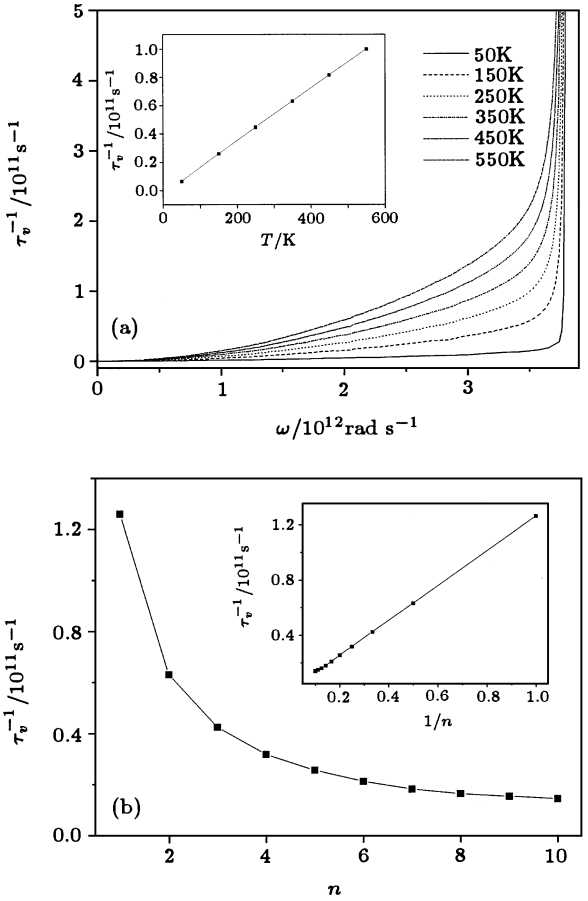


Fig.4. (a) Relaxation rates of the Umklapp process of (5 × 5) GaAs/AlAs superlattices at different temperatures. The inset shows that the relaxation rates of (5×5) GaAs/AlAs superlattices increase linearly as the temperature increases at the phonon frequency $\omega = 2.6\times 10^{12}$ rad s^{-1}. (b) Relaxation rates of Umklapp process of (n × n) ($n = 1 - 10$) GaAs/AlAs superlattices at 300K at the phonon frequency $\omega = 1.8\times 10^{12}$ rad s^{-1}. The inset shows that the relaxation rates are inversely proportional to the number of monolayers n at 300K at the phonon frequency $\omega = 1.8 \times 10^{12}$ rad s^{-1}.

In Fig.5, the thermal conductivity for (n × n) (n=1, 5, 10, 15, 20) GaAs/AlAs superlattices is calcu-

lated by considering the three-phonon Umklapp process with boundary scattering. As the temperature increases at low temperatures, the thermal conductivity of the GaAs/AlAs superlattices increases rapidly. After going through a maximum at about 60K, the $\kappa(T)$ versus temperature curves shows a downward bending. The behaviour of thermal conductivity for GaAs/AlAs superlattices can be understood by the three-phonon Umklapp process. At low temperatures, the Umklapp scattering freezes out. The phonon process is dominated by the boundary scattering, and $\kappa(T)$ simply follows the temperature dependence of specific heat. As T increases, the phonon-phonon Umklapp scattering becomes stronger due to the generation of higher-energy phonons. As the Umklapp scattering becomes comparable to the boundary scattering, the thermal conductivity reaches the maximum. As T further increases, the Umklapp process dominates the phonon process in superlattices and the relaxation rates for the Umklapp process increase rapidly with temperature, resulting in the decrease in κ. The decrease of the thermal conductivity at high temperatures is consistent with the experimental results.[19] In addition, it is demonstrated that superlattices with smaller period thickness show lower thermal conductivity. As shown in the inset of Fig.5, κ at 300K is approximately proportional to the number n: i.e., the thermal conductivity of the superlattices increases with the increase of period thickness. The above results are consistent with the experimental[19] and the theoretical results calculated with rough interfaces samples based on molecular dynamics simulation[33] and a modified lattice dynamic model.[34] The period thickness dependence of the thermal conductivity in superlattices may be understood from the Umklapp process and boundary scattering. As the period thickness in superlattices increases, the probability of the Umklapp process at any given temperature will decrease, as shown in Fig.4(b). The boundary relaxation rate is higher in superlattices with small period thickness than in superlattices with large period thickness (as can be seen from Eq.(11)). Since the three-phonon Umklapp process and boundary scattering both reduce the thermal conductivity, the thermal conductivity in superlattices increases with the increase of the period thickness.

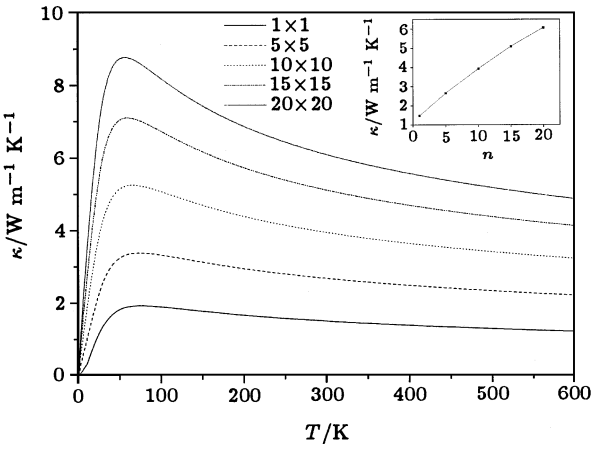


Fig.5. The lattice thermal conductivity as a function of temperature for $(n \times n)$ $(n = 1, 5, 10, 15, 20)$ GaAs/AlAs superlattices. The inset shows that the thermal conductivity at 300K increases linearly as the monolayers number n increases.

4. Conclusion

We have presented a theoretical model to calculate the cross-plane thermal conductivity in superlattices by considering the three-phonon Umklapp process. It was shown that the Umklapp relaxation rate approximates $BT\omega^2/n$ with a fitting parameter B. The thermal conductivity increases with the increase of T at low temperatures, and shows a peak behaviour at about 60K before falling off at high temperatures. The behaviour of temperature dependence of the thermal conductivity can be attributed to the boundary scattering and the Umklapp scattering process in superlattices. At low temperatures, the thermal transport in superlattices is dominated by the phonon boundary scattering. As the temperature increases, the Umklapp scattering process becomes strong and thus the main contribution to the heat transfer. In addition, the thermal conductivity increases with the increase of period thickness of the superlattices. The calculations suggest that both Umklapp scattering and boundary scattering may result in an increase of thermal conductivity.

References

[1] Hicks L D and Dresselhaus M S 1993 *Phys. Rev.* B **47** 12727

[2] Lin-Chung P J and Reinecke T L 1995 *Phys. Rev.* B **51** 13 244

[3] Hyldgaard P and Mahan G D 1997 *Phys. Rev.* B **56** 10754

[4] Venkatasubramanian R, Siivola E, Colpitts T and

O'Quinn B 2001 *Nature* **413** 597
[5] Harman T C, Taylor P J, Walsh M P and LaForge B E 2002 *Science* **297** 2229
[6] Zhu B F 1992 *Phys. Rev.* B **46** 13 619
[7] Jin Y, Hou Y T, Zhang S L, Li J, Yuan S X and Qin G G 1992 *Phys. Rev.* B **45** 12 141
[8] Chen G 1996 *Ann. Rev. Heat Transfer* (Hemisphere, New York) Vol.7
[9] Bhattacharya P 1997 *Semiconductor Optoelectronic Devices* (Prentice-Hall, Upper Saddle River, NJ)
[10] Liu Y *et al* 1994 *Chin. Phys. Lett.* **11** 229
[11] Wei J H, Xie S J and Mei L 2000 *Acta Phys. Sin.* **49** 2254 (in Chinese)
[12] Li S S, Zhu B F and Xia J B 1999 *J. Phys.: Condens. Matter* **11** 2809
[13] Zhang A Z, Zhang P, Duan S Q, Zhao X G and Liang J Q 2001 *Chin. Phys.* **10** 537
[14] Yao T 1987 *Appl. Phys. Lett.* **51** 1798
[15] Yu X Y, Chen G, Verma A and Smith J S 1995 *Appl. Phys. Lett.* **67** 3554
[16] Venkatasubramanian R 2000 *Phys. Rev.* B **61** 3091
[17] Lee S, Cahill D G and Venkatasubramanian R 1997 *Appl. Phys. Lett.* **70** 2957
[18] Capinski W S and Maris H J 1996 *Physica* B **219** 699
[19] Capinski W S, Maris H J, Ruf T, Cardona M, Ploog K and Katzer D S 1999 *Phys. Rev.* B **59** 8105
[20] Huxtable S T *et al* 2002 *Appl. Phys. Lett.* **80** 1737
[21] Ren S Y and Dow J D 1982 *Phys. Rev.* B **25** 3750
[22] Callaway J 1959 *Phys. Rev.* **113** 1046
[23] Chen G and Neagu M 1997 *Appl. Phys. Lett.* **71** 2761
Chen G 1998 *Phys. Rev.* B **57** 14 958
[24] Tamura S I, Tanaka Y and Maris H J 1999 *Phys. Rev.* B **60** 2627
[25] Yang B and Chen G 2001 *Microscale Thermophys. Eng.* **5** 107
[26] Cao J X, Yan X H, Xiao Y, Tang Y and Ding J W 2003 *Phys. Rev.* B **67** 045413
[27] Zou J and Balandin A 2001 *J. Appl. Phys.* **68** 2932
[28] Khitun A and Wang K L 2001 *Appl. Phys. Lett.* **79** 851
[29] Lax M, Hu P and Narayanamurti V 1981 *Phys. Rev.* B **23** 3095
[30] Herring C 1954 *Phys. Rev.* **95** 954
[31] Balandin A and Wang K L 1998 *Phys. Rev.* B **58** 1544
[32] Xiao Y, Yan X H, Cao J X and Ding J W 2003 *J. Phys.: Condens. Matter* **15** L341
[33] Daly B C, Maris H J, Imamura K and Tamura S 2002 *Phys. Rev.* B **66** 02430
[34] Yang B and Chen G 2003 *Phys. Rev.* B **67** 195311