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The influence of phonon boundary scattering on the thermal conductivity of a two-dimensional noninteractive phonon system of nanosized structures

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One of the problems that arise when studying the thermal conductivity of low-dimensional phonon systems at low temperatures is the appearance of differences in expressions for the thermal conductivity as a function of sample size, as well as the appearance of unusual dependences of heat fluxes on temperature gradients. For example, in the generally accepted Casimir – Zaiman model, it is assumed that a linear temperature gradient is created on the lateral surface by external sources. Moreover, the Casimir model requires two conditions at the border. This is a diffuse reflection in which the phonon is reflected with an isotropic angular distribution function. The second condition is the presence of redistribution of phonons by energy, so that the distribution of reflected phonons corresponds to the radiation of an absolutely black body - that is, the reflection of phonons must be inelastic. And if the first condition can be achieved, for example, by boundaries with a certain degree of roughness, the second condition can be achieved only in the presence of thermal contact between the side edges of the sample and the thermal medium at a certain temperature distribution. In the case of thermally insulated sample boundaries (for example, when the sample is in vacuum) or at least with imperfect thermal contact, the fulfillment of the second condition is practically impossible.

In this paper, we consider the problem of thermal conductivity of two-dimensional nanostructures - nanobands - in the temperature range, when the interaction between phonons can be neglected. In this ballistic mode, heat fluxes can be limited only by the interaction of phonons with the sample boundaries. A number of types of interaction of phonons with the boundaries of two-dimensional samples are considered: absorption at the boundary, finite number of reflections, absorption inside the sample on defects, impurities, etc. Explicit expressions of thermal conductivity in these cases are derived. Interpolation relations are obtained, which generalize the existing expressions of thermal conductivity in the case of mirror reflection and reflection with losses.

Keywords: thermal conductivity, phonons, heat flux, two dimensional nanoribbons.

Вплив фононного граничного розсіювання на теплопровідність двовимірної неінтерактивної фононної системи нанорозмірних структур

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Однією з проблем, що виникає при дослідженні теплопровідності фононних систем з низькою розмірністю при низьких температурах, є поява розбіжності у виразах для коефіцієнта теплопровідності як функції розміру зразка, а також поява незвичайних залежностей теплових потоків від градієнтів температури. Наприклад, у загальноприйнятій моделі Казимира–

Займана передбачається, що на бічній поверхні зовнішніми джерелами створюється лінійний температурний градієнт. Більше того, в моделі Казимира необхідні дві умови на кордоні. Це дифузне відбиття, при якому фонон відбивається з ізотропною функцією кутового розподілу. Друга умова — наявність перерозподілу фононів за енергією, таким чином, щоб розподіл відбитих фононів відповідав випромінюванню абсолютно чорного тіла — тобто відбиття фононів має бути непружним. І якщо перша умова може бути досягнута, наприклад, за допомогою кордонів з певним ступенем шорсткості, то друга умова може бути досягнута лише за наявності теплового контакту між бічними краями зразка та термічним середовищем за певним розподілом температури. У разі термоізольованих кордонів зразка (наприклад, коли зразок знаходиться у вакуумі) або, принаймні, з недосконалим тепловим контактом, виконання другої умови є практично неможливим.

У статті ми розглядаємо проблему теплопровідності двовимірних наноструктур – нанострічок – в діапазоні температур, коли взаємодією між фононами можна знехтувати. У цьому балістичному режимі теплові потоки можуть бути обмежені лише взаємодією фононів з межами зразків. Розглядається низка типів взаємодії фононів з межами двовимірних зразків: поглинання на межі, кінцеве число відбиттів, поглинання всередині зразка на дефектах, домішках тощо. Виведено явні вирази теплопровідності в цих випадках. Отримано інтерполяційні співвідношення, які узагальнюють існуючі вирази теплопровідності у випадку дзеркального відбиття та відбиття зі втратами.

Ключові слова: теплопровідність, фонони, тепловий потік, двовимірні нанострічки.

INTRODUCTION

One of the problems that arises when studying the thermal conductivity of phonon systems with low dimensionality at low temperatures is the appearance of divergence in the expressions for the thermal conductivity coefficient as a function of sample size [1 – 4], as well as the appearance of unusual dependences of heat fluxes on temperature gradients [2, 5].

For example, in the generally accepted Casimir–Ziman model [2] it is assumed that a linear temperature gradient is created on the lateral surface by external sources. Moreover, in the Casimir model, two conditions at the boundary are necessary. This is a diffuse reflection, in which a phonon is reflected with an isotropic angular distribution function. The second condition is the redistribution of phonons by energy, so that the distribution of reflected phonons corresponds to the radiation of a absolutely black body - that is, the reflection of phonons must be inelastic. And if the first condition can be achieved, for example, by using boundaries with a certain degree of roughness, then the second condition can be achieved only if there is thermal contact between the lateral edges of the sample and thermal media, and with a given temperature distribution. In the case of thermally insulated sample boundaries (for example, when the sample is in vacuum) or, at least, with imperfect thermal contact, the second condition is almost impossible.

In this work, we study the stationary nonequilibrium state of the phonon system, which is provided by the interaction of phonons with the lateral boundaries of the samples. Particularly, we consider the influence on the heat flow of the phonon absorption at the boundaries, the finite number of phonon interactions with the boundaries, as well as the dependence on the finite value of phonon lifetime. As the result we get the exact results for definite cases and the interpolation formulas for intermediate cases.

As the object of study, we consider phonon system of a rectangular two-dimensional strip of width W and length L (Fig. 1). For simplicity we consider just longitudinal phonons and do not include bending vibrations and transverse phonons. For such a system, we introduce two-dimensional densities of energy E and heat capacity C_S:

$$E = \int \hbar \omega n(T) d\Gamma \,,$$

$$\tilde{N}_S = \left.\frac{\partial E}{\partial T}\right|_S = \int \hbar \omega \frac{\partial n(T)}{\partial T} d\Gamma \,, \qquad (1)$$

Here ω = ω(k) is the energy-momentum relation for phonon, $n(T)$ is the distribution function of phonons and $d\Gamma = dk_x dk_z / (2\pi)^2$ is two-dimensional phase volume.

As a heat flux, we consider two-dimensional heat flux

$$\mathbf{Q} = \int \hbar \omega \frac{\partial \omega}{\partial k} \mathbf{k} n(T) d\Gamma \,. \qquad (2)$$

that is the flow of energy per unit of time and through the unit of length and has a dimension W/m.

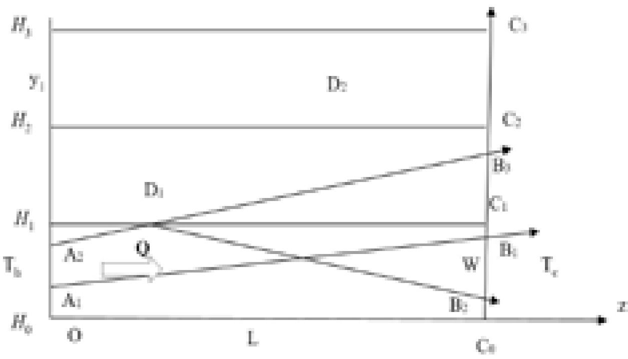

Fig. 1. The geometry of the problem.

As heat sources we consider the heat reservoirs of infinite heat capacity with given temperatures: the hot one with a temperature of T_h and cold one with a temperature of T_c, which are in thermal contact with the end faces of H_0H_1 and C_0C_1 (see Fig.1), respectively. It is assumed, that these reservoirs provide a constant temperature on these sides and interface heat contacts are ideal. In this case, the heat flux that is radiated by these walls will be determined by the law of radiation of an absolutely black body [5].

$$Q_{BBR} = \frac{1}{\pi} V_{eff} E \,. \tag{3}$$

where the effective phonon velocity is introduced as follows

$$V_{eff} = \int \hbar\omega v_g\ (\omega) n(T) d\Gamma \,/ \int \hbar\omega n(T) d\Gamma \,, \tag{4}$$

Now we can consider the different cases of phonon scattering on the lateral sides of the sample and influence of this scatterings on the heat flow.

ABSOLUTELY ABSORBING BOUNDARY

Now we consider the limiting case in which the lateral boundaries of the sample are absolutely absorbing. In this case, the resulting heat flux will be determined only by phonons, which are emitted on the hot wall and fall on the cold one and conversely without reflections from the lateral walls. Then the resulting heat flux density for small temperature differences is determined as

$$Q_0 = \frac{1}{\pi} V_{eff} C_S (T_h - T_c) R_0 \tag{5}$$

Here the value of form-factor R_0

$$R_0 = \frac{\int_0^W \frac{dx}{W} 2 \int_0^{\arcsin(x/L)} \cos(\varphi) d\varphi}{2 \int_0^{\pi/2} \cos(\varphi) d\varphi} = \frac{W}{\sqrt{W^2 + L^2} + L} \tag{6}$$

is equal to the part of the energy emitted by one of the sides, which goes directly to the other side, without reflections in the lateral walls (see, for example, line A_1B_1 at Fig. 1). It is determined by the isotropic nature of the radiation (6) and the geometric dimensions (and shape) of the sample:

Now we can introduce the effective coefficient of thermal conductivity, as the coefficient of proportionality between the heat flux density and the ratio of the temperature difference to the length of the sample:

$$Q_0 = \kappa_{eff} \frac{(T_h - T_c)}{L} \tag{7}$$

The thermal conductivity coefficient turns out to be presented in the form, which is usual for phonon systems:

$$\kappa_{eff} = \frac{1}{2} C_S V_{eff} \Lambda_0 \,. \tag{8}$$

This expression contains information about the dimension of the conductor (two in the denominator), the heat capacity of the phonon gas, the effective velocity and effective path length of the phonons:

$$\Lambda_0 = \frac{2}{\pi} L R_0 = \frac{2}{\pi} \frac{LW}{\sqrt{W^2 + L^2} + L} \,. \tag{9}$$

From this formulae it follows that for large W and (or) small L, the presence of lateral boundaries can be neglected, and the heat flux turns out to be equal to the difference in fluxes (3) from the hot and cold edges and does not depend on the distance L between them.

In the case of small W and (or) large L, the resulting heat flux is determined by the width of the sample, and the mean free path of phonons is determined by a ratio that is similar to the corresponding expression for three-dimensional heat conductors in this limit:

$$\Lambda_0 (W \to 0) = \frac{1}{\pi} W \,. \tag{10}$$

Now consider the opposite case of specular reflection.

ABSOLUTELY SPECULAR BOUNDARY

In the case of absolutely specular lateral boundaries, all phonons emitted by the heater and the cooler reach the opposite ends without loss. This leads to the obvious result [3, 4] that the heat flux will not depend on the length of the conductor. In this case, the thermal conductivity determined by relation (7) will linearly increase with length. A similar result follows from (8) in the limit of small L, but in this case all the phonons fall at opposite ends due to the absence of reflections in the side walls.

Let's obtain the same result analytically, taking into account all phonon reflections in the side walls. To account all the reflections, we use the image method. In this method, a phonon moving, for example, along the trajectory $A_2D_1B_2$ in Fig. 1 is considered as a phonon freely moving along the $A_2D_1B_3$ path, where point B_3 is an image of point B_2 in the lateral wall H_1C_1. In this case, the phonon flux which have exactly n reflections from the lateral walls and was radiated by warm (left) end

$$\begin{aligned} Q_h^{(n)} &= Q(H_0H_1 \to C_nC_{n+1}) + \\ &+ Q(H_0H_1 \to C_{-n}C_{-n+1}) = \\ &= 2Q(H_0H_1 \to C_nC_{n+1}) \end{aligned} \tag{10}$$

and cold (right) end

$$\begin{aligned} Q_c^{(n)} &= Q(C_0C_1 \to H_nH_{n+1}) + \\ &+ Q(C_0C_1 \to H_{-n}H_{-n+1}) = \\ &= 2Q(C_0C_1 \to H_nH_{n+1}) \end{aligned} \,, \tag{11}$$

where n = 1,2,3…, are defined by following relations:

$$Q_h^{(n)} = Q_{BBR}(T_h) \cdot \left(R_n - R_{n-1}\right) \quad \text{and}$$

$$Q_c^{(n)} = Q_{BBR}(T_c)\cdot\left(R_n - R_{n-1}\right) \quad (12)$$

The quantities R_n are determined by an integral similar to (6)

$$R_n = \frac{1}{W}\int_0^W dx \int_0^{\phi_n(x)} \cos(\phi)d\phi \; . \quad (13)$$

but with other limits of the angular variable:

$$\sin\varphi_n(x) = \frac{(n+1)W - x}{\sqrt{(\,n+1)W - x)^2 + L^2}} \; . \quad (14)$$

As a result

$$R_n = \frac{1}{W}\left(\sqrt{(n+1)^2W^2 + L^2} - \sqrt{n^2W^2 + L^2}\right). \quad (15)$$

From this relation, it obviously follows that if the reflections are specular, the corresponding terms are cancelled, if one summarizes the quantities (10) or (11):

$$Q = \sum_0^\infty Q_n = Q_{BBR}\begin{pmatrix} R_0 + \\ +\sum_1^\infty\left[R_n - R_{n-1}\right]\end{pmatrix} = \quad (15)$$
$$= Q_{BBR}R_\infty \equiv Q_{BBR}$$

Here $R_\infty = 1$ is the limiting value of R_n when n tends to infinity.

This answer is fairly obvious, and suggests that in the absence of resistive processes it is impossible to create a temperature gradient inside the sample. Now we can proceed to consider possible resistive processes at the boundaries that will lead to the presence of a temperature gradient inside the conductor and, consequently, to the final value of the thermal conductivity coefficient.

FINITE NUMBER OF REFLECTIONS

The simplest model of such processes is the restriction the reflections number N, that will allow us to consider the nature of the divergences in a number of limiting cases. In this case, after mutual reductions in the values of R_n, only the last of them remains:

$$Q_N = \sum_0^N Q_n = \left[Q_{BBR}(T_h) - Q_{BBR}(T_c)\right]\cdot R_N =$$
$$= \frac{\Delta Q}{W}\left(\sqrt{(N+1)^2W^2 + L^2} - \sqrt{N^2W^2 + L^2}\right) \quad (16)$$

For the convenience of further discussions, a normalized heat flux is introduced that is directly related to the dimensionless effective mean free path of phonons:

$$q = \frac{Q_N}{\Delta Q} = \frac{\pi}{2L}\Lambda \; . \quad (17)$$

Thus, for a finite number of reflections, this heat flux is

$$q_N = \frac{(2N+1)W}{\sqrt{(N+1)^2W^2 + L^2} + \sqrt{N^2W^2 + L^2}} \quad (18)$$

For infinite N, we obtain the limit of completely specular reflections (15)

$$q_{spec} = 1, \quad (19)$$

which does not depend on the sample sizes.

In the case of finite N for short samples, when the contribution of the reflections becomes negligible, the result is again equal to (18)

$$q_{norefl} = 1 \quad (20)$$

For enough narrow and long conductors at finite N we obtain

$$q_{0N} = \frac{2N+1}{2}\frac{W}{L} \; . \quad (21)$$

Thus, for the effective phonon mean free path

$$\Lambda_{0N} = \frac{2N+1}{\pi}W \; . \quad (22)$$

we obtain a value that is determined by the N-fold width of the sample.

THE CASE OF ABSORPTION AT THE BORDER

Another simple resistive process that can be proposed is the absorption at the boundary, in which the angle of reflection of the phonon is equal to the angle of incidence, but the number of phonons decreases p times, that is, the value of p play the role of the absolutely specular reflection probability.

This situation takes place if the conductor does not have free boundaries but borders on another substance. In this case, the phonon with a certain probability p can be reflected back at the boundary, and with a probability $1 - p$ it leaves the sample.

In this case, the heat flux is determined by the sum of the infinite convergent series

$$q_p = q_0 + \sum_1^\infty q_n p^n = \cdot R_0 + \sum_1^\infty (R_n - R_{n-1})p^n \quad (23)$$

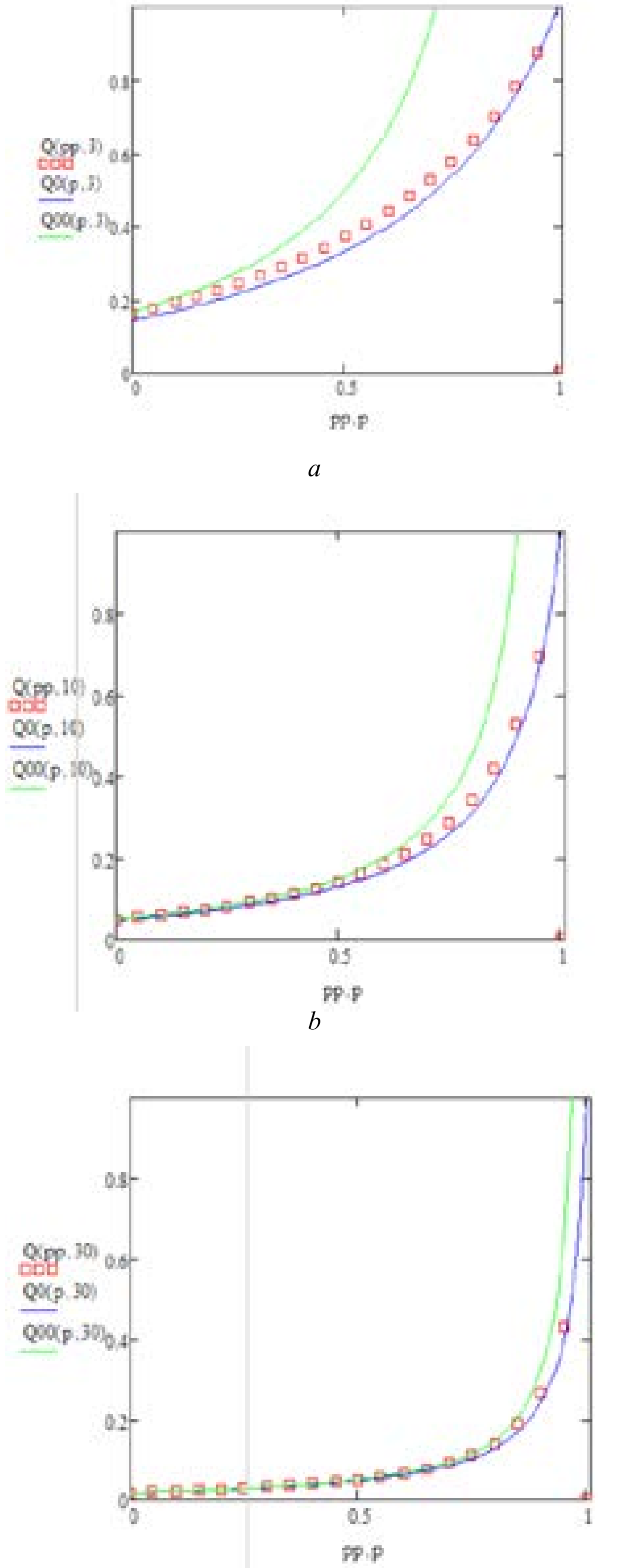

Fig. 2. The comparison of the analytical approximations with the results of calculation (red squares) for various *L*/*W* ratio – a) 3, b) 10, c) 30. Green line refers to limiting case (26). Blue line respects to interpolation (28).

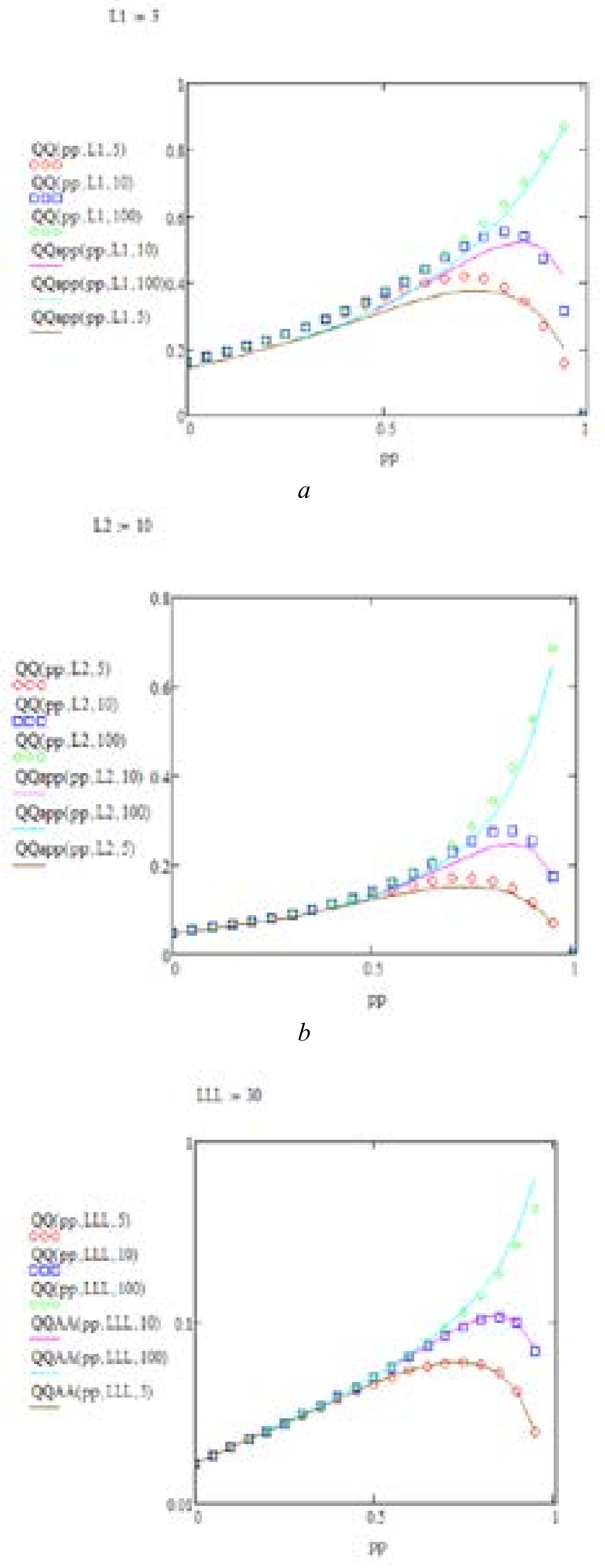

Fig. 3. The comparison of the analytical approximations with the results of numerical calculation by Eq. (30) (points) for various *L*/*W* ratio a) 3, b) 10, c) 30. Line refers to analytical interpolation (32). Different points refers to different number of reflections: red to *N*=5, blue to *N*=10, green to *N*=100.

In sum, we can rearrange the terms and get a simpler expression:

$$q_p = R_0(1-p) + (1-p)\sum_1^{\infty} R_n p^n =$$

$$= (1-p)\left[\begin{array}{l} \dfrac{W}{\sqrt{W^2+L^2}+L} + \\ +\sum\limits_1^{\infty} \dfrac{(2n+1)W}{\sqrt{(n+1)^2W^2+L^2} + \sqrt{n^2W^2+L^2}} p^n \end{array}\right] \quad (24)$$

For great W we get the expectable results

$$q_{p\infty} = (1-p)\left[1+\sum_1^{\infty} p^n\right] = (1-p)\left[1+\frac{p}{1-p}\right] = 1, \quad (25)$$

since reflections do not contribute at the small distance between the heater and the cooler.

For a narrow and long sample, we get

$$q_{p0} = (1-p)\frac{W}{2L}\left[1+\sum_1^{\infty}(2n+1)p^n\right] = \frac{W}{2L}\frac{1+p}{1-p}, \quad (26)$$

and for effective length

$$\Lambda_{p0} = \frac{W}{\pi}\frac{1+p}{1-p} = \Lambda_0\frac{1+p}{1-p}. \quad (27)$$

This result contains a well-known denominator that takes into account the efficiency of specular reflection p and gives the correct limit transition to the case of complete absorption at the boundaries (14) when $p = 0$.

But as it can be seen from Eq. 24, the result (26) is incorrect, because for any small value W/L for enough great n, the product nW/L ceases to be small.

To account this feature, one can use the following interpolation formula

$$q_{p0in} = \frac{W(1+p)}{2L(1-p)+W(1+p)} = \left(1+q_{p0}^{-1}\right)^{-1}, \quad (28)$$

which describes the real sum (24) with high accuracy (see Figure 2).

As a result, for the effective length we propose following result

$$\Lambda_p = \Lambda_0\frac{1}{\dfrac{1-p}{1+p}+\dfrac{W}{2L}}, \quad (29)$$

that can be used for the values of the ratio $W/L < 1$ and in the entire range of value p.

To consider the behavior of the q near the value $p = 1$ we go back to the case of finite number N of reflections and present the partial sum of the infinite series fom (26) as follows:

$$q(p,N) = (1-p)\frac{W}{2L}\left[\begin{array}{l}1+ \\ +\sum\limits_1^{N}(2n+1)p^n\end{array}\right] = \quad (30)$$

$$= \frac{W}{2L}\left[\frac{1+p}{1-p} - pp^N\left\{\frac{2}{1-p}+2N+1\right\}\right]$$

In the case $p < 1$ and $N \to \infty$ this result obviously gives the Eq.(26). In the case if finite N and p close to unity we get the expression that demonstrate the competitive influence of number of reflections N and absorption at one reflection p:

$$q(p \approx 1, N) = \frac{W}{2L}(N+1)^2(1-p). \quad (31)$$

This result shows, as p tends to unity, the contribution of terms with a finite number of reflections R_n decreases in comparison with the limiting value $R_\infty = 1$, which formally corresponds to an infinite number of reflections. Thus, any physical resistive process that limits the phonon mean free path will lead to the elimination of a divergence of the form (27).

Expression (30) can be used for interpolation folrmulae like (26)

$$q_{in}(p,N) = \left(1+q^{-1}(p,N)\right)^{-1} \quad (32)$$

As can be seen from Fig.3, the resulting expression practically coincides with expression (18) for various sizes of the system. Thus, we have obtained expression (19) for a finite number of reflections and use it for description of experimental data.

CONCLUSION

In the paper we studied the stationary nonequilibrium state of the phonon system, which is provided by the interaction of phonons with the lateral boundaries of the samples. Particularly, we consider the influence on the heat flow of the phonon absorption at the boundaries and the finite number of phonon interactions with the boundaries. As the result we get the exact results for definite cases and the interpolation formulas for intermediate cases. Particularly, the exact expression (24) for heat flow in the case of absorption on the lateral boundary was derived and the simple interpolation formula (28) was proposed. For the case of finite number of reflections the general expression (30) was presented and an analytical approximated formula (32) was derived. The comparison of numerical calculations by exact expressions with analytical interpolation formulae was carried out and demonstrated good agreement.

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