A review to the specific problems of nano thermal physics

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Abstract. The paper is devoted to three current problems that related to thermal transfer in the solid-state nanostructures. The first task is the analysis of interaction of phonons with rough solid boundaries, the presence of which leads to a significant increase in the resistance to heat transfer in nanostructures. This fundamental problem is discussed in a connection to the theoretical formulation of rough interface between solids. The second task is exploring methods to calculate diffusion and heat transfer in solids within the framework of detailed consideration of phonon interaction processes. The development of the computational model that adequately represents the physics of interaction processes and allows to calculate thermal properties that are in a good agreement with experimental data remains open. The third task is to calculate the thermal boundary conductance. The problem is solved by considering the processes of scattering and refraction of elastic waves at the interface. For the first time, the dispersion properties of elastic waves are taken into account in the problems of this kind. The result is the applicability of the new model in a much wider temperature range than in the existing models.

1. Introduction
There are a lot of problems that are characteristic of nanothermophysics, but are not of interest in classical thermophysics, which studies the processes occurring in macroscopic objects. It is important to emphasize that at present the resolution of these problems is still in its infancy; This area is developing intensively, but there are many “white spots” that require fruitful analysis. Thus, in this paper, we consider three urgent problems characteristic of solid-state nanostructures — semiconductors and dielectrics, in which heat transfer is carried out by phonons [1]. These materials are of particular interest in connection with the development of nanoscale electronics, optics, and acoustics.

1) The first task is the interaction of phonons with the boundaries of nanostructures. It should be noted that there are two types of problems on the interaction of phonons with rough surfaces. The first type is interactions with the boundaries of solids separating phonons from the gaseous medium, in which phonons can no longer propagate. In these cases, the problem is reduced to the complete reflection of phonons. The second type is the interaction of phonons with boundaries separating them from other solids or liquids. In these cases, part of the energy of the phonons (or elastic waves) propagating in the solid transfer will be transferred to another solid or liquid through the surface. Therefore, the problem of co-interaction with a rough surface changes fundamentally.

2) The second task is to study the kinetics of phonon gas. The processes of interaction of phonons with each other are very complex, because of which it is not yet possible to ensure their full and detailed consideration in the simulation of transfer processes. Therefore, existing models of heat transfer are quite crude [2]. In this regard, in the framework of the relatively simple problem of phonon diffusion, it
was possible to take into account almost exactly phonon interaction processes [3]. This allows to estimate the accuracy of modern methods for calculating kinetic processes. In principle, this problem is common to both nano- and macroscale thermal physics. It should be emphasized that the Mattissen rule is also used in ab initio (“first-principles”) models of thermal conductivity [4].

3) The third task is to improve the method of calculating Kapitza resistance. The appearance of contact thermal resistance at the boundary between two solids has a significant effect on a heat transfer in multilayer nanostructures (electronic circuits, nanolasers, superlattices), because of its limitation of heat transfer between individual layers.

2. The interaction of phonons with the boundaries of nanostructures

2.1. Free boundary
The interaction of phonons with the free boundary of a sample has been the part of the theory of phonon heat transfer when calculating the thermal conductivity of macroscopic samples. The first fundamental point is how the scattering processes at the boundary contribute to thermal conductivity and under what conditions this contribution becomes significant. To answer this question, a dimensionless parameter is used, which is equal to the ratio of the mean free path $l_\infty$ to the characteristic size of the sample $l$ — the Knudsen number, $Kn = l_\infty / l_{\text{char}}$. For values of $Kn \sim 1$ and $Kn \gg 1$ we obtain, respectively, the diffusion-ballistic and ballistic heat transfer regimes; that is, the conditions under which the interaction of phonons with a boundary plays an essential or decisive role in the thermal conductivity.

To visualize this dependence, we analyze the heat transfer regime for Silicon. Since the mean free path is a function of temperature, the heat transfer mode (Knudsen number) will depend on the temperature and also on the size of the sample.

![Figure 1. Assessment of the heat transfer regime for Silicon: $Kn \ll 1$ – diffusion; $Kn \sim 1$ – diffusion ballistic; $Kn \gg 1$ – ballistic mode.](image-url)

Therefore, Figure 1 makes it possible to determine the heat transfer regime for a given structure with a characteristic size $l$ and at a given operating temperature $T$. It can be seen that the influence of scattering at the boundaries becomes decisive at room temperatures for micro-structures, and at higher temperatures – mesoscale and nanoscale structures.

The second fundamental point is the factors influencing the scattering of phonons at the free boundary. At present, the only parameter that reduces the analysis of the interaction of phonons with boundaries is the specularity parameter $p$, $0 \leq p \leq 1$, which characterizes the probability of specular scattering of phonon. To determine $p$ the expression [5] is used:

$$ p(\zeta, \theta) = \exp(-\zeta^2 \cos^2 \theta), $$

(1)
taking into account only \( \zeta = 4\pi\sigma_{rms}/\lambda \) – the ratio between the value of the root mean square roughness, \( \sigma_{rms} \), and the characteristic wavelength of the phonon, \( \lambda \); \( \theta \) – the angle between the normal to the surface and the incident phonon. Two limiting regimes of phonon scattering at the boundary are distinguished depending on \( \zeta \): 1. \( \zeta \gg 1 \) and \( p = 0 \) – diffusion scattering (Casimir), 2. \( \zeta \ll 1 \) and \( p = 1 \) – specular scattering (reflection).

In this case, the influence of temperature, features of surface geometry, directions of heat distribution and etc. are not taken into account. Moreover, the specular reflection parameter is used as a “fitting” parameter to achieve good agreement with the experimental data. Of fundamental importance is the multi-parameter nature of the problem being solved and the need for a detailed analysis of the influence of various factors on dispersion at the boundary. In this regard, we have developed a numerical method for estimating this quantity depending on temperature and the morphology of the sample.

To take into account all possible wavelengths in solid body and its distribution, we use the average wavelength of phonons \[ \lambda_{ph} \] depends on the polarization, \( j \), and temperature:

\[
\bar{\lambda}(j, T, \sigma_{rms}) = \frac{4\pi\sigma_{rms}}{\lambda_{ph}(j, T)}. 
\] (2)

For the average wavelength of phonons \( \bar{\lambda} \), we use the value \( \lambda_{50} \) (wavelengths of phonons that contribute 50% to thermal conductivity) and consider the Silicon as an example. According to relation (1), we determine the specularity parameter as a function of temperature (taking into account averaging over the angle of incidence \( \theta \)):

\[
\bar{p}(j, T, \sigma_{rms}) = \frac{2}{\pi} \int_{0}^{\pi/2} \exp\left[-\zeta^{2}(j, T, \sigma_{rms})\cos^{2}\theta\right] d\theta.
\] (3)

The calculation results are presented in figure 2. From the analysis of the diagram it can be concluded 1) phonons at low temperatures that have the large average wavelengths \( \bar{\lambda}_{ph} \gg \sigma_{rms} \) are scattered mainly specularly; 2) as the temperature increase and average wavelength is getting lower, the scattering regime becomes diffuse; 3) when \( \sigma_{rms} \) of the surface is getting higher, the parameter \( \zeta \) increase and the scattering becomes more diffusive, \( \bar{p} \ll 1 \).

Figure 2. Calculation of the probability of specular reflection (10) for Silicon using the average wavelength of phonons \( \bar{\lambda} = \lambda_{50}(T) \) for values of root mean square roughness \( \sigma_{rms} \): 0.1 nm, 0.5 nm, 1 nm, 5 nm, 10 nm, 50 nm and 0.5 \( \mu \)m. The solid lines correspond to transverse waves (TA), the dashed lines to longitudinal (LA). The numbers show the parameter values \( \bar{p} \) at \( T = 150 \) K.
Therefore, based on the analysis of heat transfer in micro- and nanoscale structures in which the phonon quasiparticles are the heat carriers, it is shown that the heat transfer mode is determined by the temperature $T = 150K$ and a set of characteristic linear scales (mean square roughness, characteristic sample size, average phonon wavelength and average mean free path). Methods are formulated for estimating the heat transfer regime and the type of phonon scattering at the boundary, depending on the magnitude of the corresponding characteristic size and temperature. As an example, calculations were performed for Silicon and the corresponding diagrams were obtained, which can be used in practice.

2.2. Interface

Interface scattering play the main role in heat transfer in structures where the characteristic size of sample is comparable or less than mean free path of phonons in bulk material. Interface scattering causes thermal resistance and is highly dependent upon surface roughness, structure, and scattering processes around the interface. Several experimental studies have investigated the effects of interfacial disorder and roughness on thermal boundary resistance (TBR), according to which roughness significantly affects the TBR [7]: resistance increases with an increase in the interface roughness. To date, there are no theoretical estimates of these processes. However, for nanostructures, the analysis of such processes is extremely important. It should be noted the variety of related tasks. In determining TBR (also called Kapitza resistance), the contact resistance of an infinitely thin interface is most often considered. But it is also possible the presence of thin films between two solids and transition layers in which the atoms of one material are gradually replaced by another material [8].

The theoretical formulation of such problems is still missing. It should differ significantly from the problems of the interaction of phonons with a free boundary.

3. The study of the kinetics of phonon gas

The second task is to study the kinetics of phonon gas. The processes of interaction of phonons with each other are very complex, because of which it is not yet possible to ensure their full and detailed consideration in the simulation of transfer processes. Therefore, existing models of heat transfer are quite crude [2]. In this regard, in the framework of the relatively simple problem of phonon diffusion, it was possible to take into account almost exactly phonon interaction processes. The problem posed corresponds to the problem of self-diffusion of atoms in their own gas — the diffusion of a fixed atom, which is preserved from the very beginning to the end. However, when it comes to phonons, such a variant is impossible to implement, since the phonon entering into interaction disappears as a result of it. The next span is realized by another phonon. But the main and most important feature of this dress is a strict accounting of phonon interactions, taking into account the laws of conservation of energy and quasimomentum. This allows to estimate the accuracy of modern methods for calculating kinetic processes.

As an example, we consider the situation with the Mattissen rule, which is used to solve the Boltzmann equation for determining the thermal conductivity of solids. To determine the relaxation times of a phonon gas in the presence of multiple types of interactions, the Mattissen formula is based on a dubious analogy with electrical resistance, which summarized in the presence of several different physical causes that resistance. These resistances are summed, and each is inversely proportional to the corresponding average times between interactions [9]. However, these two formulas are significantly different for electrical resistance and thermal conductivity. Therefore, doubts arise and large errors are allowed when using the Mattissen rule for determining thermal conductivity [10]. An analysis of phonon diffusion in the approximation under consideration, when all the interaction processes are taken into account separately and strictly, and are not questionably combined using the Mattissen rule, allows us to obtain rigorous results and first compare with what the Matthiessen’s rule gives [3].

The table below presents data on the determination of the times between successive interactions obtained using the Mattissen rule and as a result of analysis obtained from the study of phonon diffusion. U – processes are compared. It can be seen from the presented data that the use of Mattissen data provides higher values of the time between successive interactions, and hence, higher values of thermal conductivity than the results obtained from calculations of phonon diffusion. In this regard, we point out
the data of [11], [12], which indicate that the use of the Mattisson rule gives overestimated values of thermal conductivity.

**Table 1.** Times between successive interactions using the Mattissen rule and phonon diffusion.

| Temperature | Diffuse, s. | Mattissen, s. |
|-------------|-------------|---------------|
| 100 K       | $2.77 \cdot 10^{-10}$ | $3.66 \cdot 10^{-10}$ |
| 200 K       | $9.43 \cdot 10^{-11}$ | $9.87 \cdot 10^{-11}$ |
| 300 K       | $4.77 \cdot 10^{-11}$ | $5.17 \cdot 10^{-11}$ |
| 400 K       | $2.96 \cdot 10^{-11}$ | $3.33 \cdot 10^{-11}$ |
| 500 K       | $1.88 \cdot 10^{-11}$ | $2.31 \cdot 10^{-11}$ |

4. **Combination of methods for calculating Kapitza resistances**

There are several methods for calculating Kapitza resistance, and from the physical point of view the clearest one is the acoustic mismatch model (AMM). This method firstly proposed by Khalatnikov [13] in 1952 for calculating the phenomena that occur on the contact surfaces between two materials when the heat flow through. The essence of this model is that when an elastic wave passes through the interface of two contact objects, it transfers energy from one of them to another accompanied by reflection and refraction. The reflection of waves leads to heat resistance and occurs the temperature jump. It gave impetus to the development of AMM [13,14]. These models are based on the analysis of heat transfer processes through the interface, and scattering of elastic waves propagating in solids or liquids on two sides of the interface. When evaluating these types of models, the following circumstances must be kept in mind. First, Kapitza resistances play a significant role in the heat transfer of only nanostructures. In macroscopic systems, their contribution is negligible. Secondly, when considering AMM models, in their current form they are suitable only for conditions when the scale of the roughness of the interfaces is of the order of atomic sizes (that is, no more than two nanometers). These features correspond to each other; therefore, the AMM models are suitable for calculating the Kapitza resistances in nanostructures. The results of calculations based on existing AMM models [14, 15] show that they have good agreement with experimental data, only in the low-temperature region (no more than 50 K).

![Figure 3](image-url). Predicted and experimental values of Kapitza conductance at the Al-Si interface as a function of temperature. Triangles are measured values from [17]. The solid lines represent the Kapitza conductance obtained using the presented model EWMD (Elastic wave model with dispersion) [16], and the dashed lines show the theoretical results of the most commonly used conventional AMM [12,13,14] и DMM [18].

However, in the modern version, the model is not complete – it does not take into account the dispersion of acoustic waves and the presence of maximum frequencies [14, 15]. As shown in Fig.3, the theoretical results of our model (EWMD), AMM and DMM are compared with the experimental data.
It was found that proposed considerations (the dispersion of waves in combination with the limitation of the maximum frequencies of the incident wave, reflected and refracted waves) can significantly improve the agreement between theory and experiment. We use a model based on the consideration of energy transfer through an interface by elastic waves, and firstly obtain a good agreement with experiment data in a wide temperature range from 0 to 300 K. At the Al–Si interface, the maximum discrepancy between theory and experiment at 300 K [17] is only 10%. The roughness in the samples was 0.5 nm.

Further, it is noted that the results of AMM agrees well with the experimental data in the temperature range from 0 to 50 K. It is necessary to add an important explanation about the applicability of AMM theory to Kapitza resistances analysis on nanostructures. The usual view that AMM is only suitable within low temperatures is incorrect for nanoscale materials. This is due to two circumstances. First, the roughness of nanostructures is significantly less than that of macroscopic samples; they are about tenths of a nanometer. Second, the frequency of elastic waves is limited by the maximum value based on the dispersion relations of materials on both side of the interface. A detailed analysis shows that these two circumstances ensure that up to a temperature of the order of 1000 K, the ratio of the elastic wavelength is sufficient to reach the average roughness. We considered these factors and the results showed that taking into account the dispersion gives a good agreement with experimental data for nanostructures in a wide temperature range, at least up to 350 K [16].

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