A new technique for modelling phonon scattering processes at rough interfaces and free boundaries of solids

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Abstract. Scattering processes at interfaces and free boundaries of solids strongly affect heat transfer in micro- and nanostructures such as integrated circuits, periodic nanostructures, multilayer thin films, and other nanomaterials. Among many influencing factors, surface roughness due to atomic disorder plays a significant role in the rate of thermal transport. Existing approaches have been developed only for the limiting cases of smooth or completely diffuse surfaces. We have developed a new effective and simple method based on a direct consideration of the scattering of elastic waves from a statistically random profile (using a normal Gaussian surface as an example). This approach, first, allows to generalize common methods for determining the thermal properties of a real random rough surface using simple modifications, and, second, provides a tool for calculating the Kapitza conductance and the effective longitudinal thermal conductivity and studying the influence of roughness on heat transfer.

1. Introduction

The study of energy transfer processes in nanoscale electrical circuits with powerful specific heat generation is a wide range of complex problems, whose solution is extremely relevant and absolutely necessary, from both theoretical and practical points of view [1, 2]. Unlike macroscopic bodies, for which the classical laws of heat transfer are applicable, for micro and nanoscale structures, it is necessary to use completely different methods based on the statistics of quasiparticles called phonons—quanta of lattice vibrations. Over the past decades, there have been great advances in this area [3], but there are also a number of blind spots in fundamental the research that hinder the development of new thermal physics and hence nanoelectronics. In this paper we consider two modern problems of thermal transport of micro- and nanostructures:

1) heat transfer through the contact surfaces between two solids (through interfaces). A quantitative measure of the rate of heat transfer through interfaces is the Kapitza conductance (the reciprocal is the thermal boundary resistance (TBR).

2) Scattering of heat carriers (phonons) at the free boundary of a sample during the propagation of heat along the films (tubes). Quantitatively, this process is characterized by the effective longitudinal thermal conductivity (ETC).

In this case, the boundary of the sample is rough, i.e., close to the real one, and not smooth, as is usually assumed in classical thermal physics. At the moment, there are no reliable methods for calculating thermophysical properties (such as TBR and ETC) that would provide sufficient predictive modeling and control of the thermal regime of promising devices. To substantiate this thesis, we will focus on each of the above problems in more detail.
1.1. Thermal boundary resistance (of interface)

The analysis of heat transfer through the contact surface of two solid bodies is a continuation of the classical problem of determining the Kapitza conductance (resistance) [4]. The task is extremely important, because, first, nanoelectronic circuits contain millions of contacts, and second, estimates show that thermal boundary contacts create the greatest total resistance to heat removal [5]. The existing theory of TBR calculation is based on two models: the acoustic mismatch model (AMM) [6] and the diffusion mismatch model (DMM) [7]. At the same time, it is important that in the AMM the interface is considered in the limiting case of a smooth surface, and in the DMM it is completely diffuse, whereas real interfaces have a complex structure, including roughness.

The effect of interface roughness on TBR has been investigated experimentally [8–11] and more recently using molecular dynamics [12–14] and ab initio [15] methods. Note, first, that these methods do not allow calculations for a previously known surface structure. Second, depending on roughness, phonons with different wavelengths will scatter differently: some diffusely, and some specularly. Therefore, further development of the computational model obviously requires the use of statistical methods to analyze the mechanism of phonon scattering on rough surfaces, and we have taken the first steps in the development of this method [16].

1.2. Effective thermal conductivity (films and tubes, in-plane)

Experimental studies of nanostructures (e.g., nanowires) have shown that roughness has a significant effect on heat transfer [17] due to the interaction of phonons with free surfaces. This is a decisive factor in the development of reliable nanotransistors, nanolasers, superlattices, and other devices.

The conditions in nanostructures are such that the mean free path (MFP) of phonons from one surface of films to another is significantly less than the average MFP of internal interaction processes in the sample. This is the so-called ballistic heat transfer regime [3]. The modern theory of thermal conductivity is based on the consideration of two limiting cases: completely diffuse scattering at the sample boundary and specular scattering, where the MFP is limited only by internal processes (phonon-phonon, on impurities, etc.) [18]. In this case, the transition from a purely diffuse regime to a specular one is carried out through the specular parameter $p$, which allows one to relate the MFPs of ballistic phonons for the two specified limiting cases [18].

The above method does not allow predicting the thermal properties of such structures, since the specular reflection parameter is used as a fitting parameter to ensure better agreement between theory and experiment [19, 20]. However, it is worth noting that in [21, 22], it is proposed to evaluate the parameter $p$ as an integral (average) parameter for the sample. At the same time, it is not considered how the wavelengths of phonons, their polarization, and the temperature, geometry, and size of the sample will affect the nature the phonon scattering at a rough boundary (diffuse or specular reflection). Analysis shows that the question of the further development of the computational model remains open.

2. Mathematical model

Despite the fundamental differences between phonon scattering at rough interfaces (problem 1) and the interaction of phonons with free rough boundaries (problem 2), the method we propose allows us to develop a unified computational technique.

First, the method is based on the consideration of phonons as quasiparticles that obey Bose–Einstein (BE) statistics. Therefore, a number of properties of the carrier medium (crystal lattice) are required to describe phonons: dispersion relations and the density of states (DOS). In this paper, we use a well-proven approach to the application of polynomial approximation of experimental data [19]. The result is a nonlinear dispersion relation that characterizes the elastic properties of a real medium. We also note that the use of phonon statistics makes it possible to calculate the average values of the required thermal properties (thermal conductivity, Kapitza conductance) as a function of temperature.

First, we use the Landauer formalism, according to which the Kapitza conductance (thermal boundary resistance) is given by the expression
and the effective thermal conductivity is
\[ \kappa = \frac{1}{3} \sum_j \int_0^{\omega_{\mathrm{ph}}} \int_0^{\omega_{\mathrm{ph}}} C_{\mathrm{ph}}(\omega, T) \Phi_j(\omega) \nu_{g,j}(\omega) \alpha_{1-2,j}(\omega, \theta) \cos \theta \sin \theta \, d\omega \, d\theta, \]  
where \( \omega \) is the phonon frequency, \( j \) is the phonon polarization, \( C_{\mathrm{ph}} \) is the phonon capacity, \( f_{\mathrm{BE}} \) is the Bose–Einstein distribution function, \( f_{\mathrm{BE}}(\omega) = \left[ \exp(\hbar \omega/k_\mathrm{B} T) - 1 \right]^{-1} \), \( \Phi_j \) is the density of state \([19]\); \( \nu_{g,j} \) is the phonon group velocity; \( \theta \) is the angle between the direction of incidence of a phonon and the normal to the surface; \( \alpha_{1-2,j} \) is the coefficient of energy transfer from material 1 to material 2, and \( l_j \) is the mean free path of phonons. Note that expression (2) is given in a quasi-isotropic approximation \([19]\); in this case, the heat flux propagates in the film (tube) or in the so-called in-plane direction.

Thus, when calculating thermophysical properties, roughness is taken into account through the transfer coefficient \( \alpha_{1-2,j} \) in expression (1) and through the mean free path \( l_j \) in equation (2). Next we will talk about the features of the calculation of \( \alpha_{1-2,j} \).

Second, both problems are qualitatively different from each other. In problem 1, part of the energy from medium 1 is transferred through the interface to medium 2 (see Figure 1), and in problem 2, the energy does not leave the carrier medium. Figure 1 shows a schematic image of scattering on a rough interface between two bodies (solid 1 and solid 2) \([16]\). According to the theory of elasticity, a mode transformation occurs at the boundary \([23]\): an incident wave with index (0) is transformed into two reflected (1) and (2) and two refracted (3) and (4) waves. Letters denote polarizations: longitudinal (P) or transverse, where oscillations occur in the plane of propagation (SV) or across it (SH).

Figure 1. Scattering of elastic waves on a rough interface. The left picture corresponds to P or SV waves, and the right one to SH waves. Here the superscript (0) corresponds to the incident wave, (1) to the reflected longitudinal wave, (2) to the reflected transverse wave, (3) to refracted longitudinal wave, and (4) to the refracted transverse wave. The dashed line "x" is the mean line of the profile, and the solid line "h" shows the heights of the real (rough) profile.

Here we use the elastic wave model with dispersion (EWMD) to calculate the Kapitza conductance. The EWMD is a further development of the acoustic mismatch model (AMM), and it can be found in detail in \([5, 16, 23]\).
Third, a distinctive feature of this method is that the description of the rough boundary of a sample is based on statistical accounting for the slopes of the real surface profile. For example, we consider a model of a two-dimensional random Gaussian normal surface $z = \zeta(x, y)$, which is characterized by two mean values: the root-mean-square roughness $\sigma$ and the correlation length (interval) $L$. At the same time, unlike the well-known methods [18, 21, 22], the distribution density is used not for the profile heights $z$, but for profile slope gradients $n = \nabla \zeta$ [24]:

$$w(n) = \frac{1}{\sqrt{2\pi}} \exp \left(-\frac{n^2}{2\gamma^2}\right), \quad (0 \leq n < \infty),$$

where is the variance of the first derivative, $\gamma = \frac{\sigma}{l}$, which in the special case is given by the expression $\gamma = \sigma / l$. Note that in further calculations, as an argument we use not the gradient of the profile, but the angle of inclination $\phi$ (see Figure 1) between the mean line and the tangent plane, which are related as $n = \tan \phi$.

Next, the problem of the wave propagation through the interface is solved relative to the tangent plane (see Figure 1). First, the transfer coefficient $\alpha_{1-2}(\theta_0)$ is determined as a function of the local incidence angle $\theta_0$ using the well-known relations from the theory of elasticity [23]. Then using the relationship between $\theta_0$, the angle of inclination of the profile $\phi$ at the scattering point, and the actual angle of incidence $\theta$ of the wave on the interface, we can pass from the local value $\alpha_{1-2}(\phi, \theta)$ to the effective transfer coefficient $\alpha_{1-2,\text{eff}}(\phi)$ of the rough surface [16]:

$$\alpha_{1-2,\text{eff}}(\phi) = \int_{\phi_{\min}}^{\phi_{\max}} \alpha_{1-2}(\phi, \theta)w(\phi)d\phi$$

It is due to the transition from $\alpha_{1-2}$ for the tangent plane to $\alpha_{1-2,\text{eff}}$ for the interface in expression (1) that the well-known relations of the theory of elasticity [23] can be naturally applied to the problem of reflection-refraction of elastic waves at a rough interface between two solids [16]. It should also be noted that when averaging over different angles of inclination of the profile (4), we need to take into account the restrictions on the limits of integration $\phi_{\min}$ and $\phi_{\max}$ [23] that are imposed by the phenomenon of total internal reflection and change the pattern of reflected-refracted waves.

The interaction of phonons with the rough wall of a sample during in-plane heat flux propagation is considered in a similar manner. The difference is that in expression (4), $\alpha_{1-2}$ is replaces with the MFP $l_b$ between successive collisions with the boundary:

$$l_{b,\text{eff}}(\theta, h) = \int_{\phi_{\min}}^{\phi_{\max}} l_b(\phi, \theta, h)w(\phi)d\phi$$

Details of the calculation of $l_b$ can be found in the original work [18] or in our previous analysis [19]. After that, the average MFPs $l_{b,\text{eff}}$ are used to determine the effective longitudinal thermal conductivity of films (heat transfer rate) as a function of the size of the sample, its roughness, and temperature. The equation for ETC is similar to (2) but must include integration over k-space (reciprocal space of wavevectors) instead of integration over frequency. Moreover, to take into account the size effect in films, it is necessary to perform summation in the cross-plane direction and integration in the in-plane direction (for more information, see [25]).
3. Results and discussion
As an example, the problem of determining the Kapitza conductance through a rough aluminum-silicon interface is considered. The necessary properties and parameters of the materials are given in [16]. Here we focus on the calculation results presented in Figure 2. The initial conductance is the Kapitza conductance for a smooth interface (black line, "smooth"), which is obtained using the elastic wave model with dispersion (EWMD) [23]. Further, with an increase in roughness, a significant decrease in conductivity is observed. This is due to the fact that the proportion of waves capable of overcoming the interface and moving from medium 1 to medium 2 decreases, and, as a consequence, the effective transfer coefficient (4) decreases. For more information on the influence of wave polarization and interface roughness on the transfer coefficients see the article [16]. Note that here we are talking about inhomogeneities of an atomic-scale surface—the so-called atomic roughness.

![Figure 2. Kapitza conductance for various Al/Si interfaces: smooth and with a root-mean-square roughness of 0.1, 0.3, 0.5, 1, 5, and 10 nm. Circles represent experimental data [26].](image)

Thus, we have developed a new technique for studying the effect of real surface roughness of a sample on the rate of heat transfer. Our method is the first to use the statistics of profile slopes of a random surface (using, as an example, the Gaussian distribution of slopes). The presented methodology, first, made it possible to formulate a statistical model for calculating the Kapitza conductance taking into account the roughness of the interface [16] and use it to calculate the silicon-aluminum contact (Figure 2). Second, it allowed us to develop a new method for calculating the effective thermal conductivity in the ballistic regime of heat transfer taking into account the multiparametric and statistical nature of phonons and a random (rough) surface.

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