The effect of phonon diffusion on heat transfer

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Abstract. The work is devoted to the development of a tool that allows simulating diffusion in structures where phonons play the main role in the heat transfer (dielectrics, semi-conductors). It is shown that the nature of phonon interaction is fundamentally different from the interactions in classical gases. For the first time, a method for calculating the diffusion of phonons, that takes into account all the important features of their interaction with each other, is presented. A computer simulation of phonon diffusion for a silicon sample in a two-dimensional formulation was carried out. It is shown that the nature of phonon diffusion differs from the classical Brownian diffusion.

1. Introduction

Diffusion processes play a significant role in the heat and mass transfer of matter (except for the so-called ballistic mode of heat transfer in nanostructures). For example, in ordinary gases diffusion is the result of step-by-step random processes of particles interaction with the surrounding atoms (molecules), that is called Brownian diffusion. Einstein in 1905 [1] had shown the following relation is fulfilled for Brownian motion

\[ (x(t) - x(0))^2 = 2Dt. \] (1)

Here \( x \) – displacement of the particle, \( t \) – time, \( D \) – diffusion coefficient. The value \( (x(t) - x(0))^2 \) is the mean square value of the displacement of the particle coordinate over time \( t \). The diffusion process under these conditions is characterized by the probability of distribution in time and space \( p(x,t) \), which satisfies the Fokker-Planck equation in a one-dimensional approximation:

\[ \frac{\partial p}{\partial t} = D \frac{\partial^2 p}{\partial x^2}. \] (2)

The solution of equation (2) is looks as follows

\[ p(x,t) = \frac{1}{\sqrt{4\pi Dt}} \exp \left( -\frac{x^2}{4Dt} \right). \] (3)

It follows from the Einstein relation (3) that eventually the diffusion package is propagated proportionally to \( t^{0.5} \), and from the presented solution – that the “tails” of the distribution exponentially decrease in space. The main feature is that Brownian diffusion is a Markov process (a system without memory) – the next spatial step depends only on time. In contrast, in the case of phonon diffusion, as will be shown below, the next step depends on the energy (frequency).
In addition to the classical Brownian diffusion, there are anomalous types of diffusion, so-called superdiffusion and subdiffusion [2, 3]. The difference from classical diffusion (1) is in laws of phonon propagation. Precisely, they are expressed as follows:

\[ \langle x(t) - x(0) \rangle^2 = \text{const} \cdot t^\gamma, \]

where \( \gamma \) is not equal to one. When \( \gamma < 1 \) there is a subdiffusion, and for \( \gamma > 1 \) is a superdiffusion that is often referred to a Levi diffusion. The main feature of such diffusion is that the dispersion of mean free path can reach infinity. It is characterized by the appearance of a moving particle to travel for a very long distance - the so-called “Levy flight”. In articles [4, 5] superdiffusion applied to a phonon gas in semiconductor alloys within the framework of the truncated diffusion of Levi [3, 6, 7]. This model has a finite value of the dispersion of the free path lengths. As a result, the calculations of thermal conductivity of InGaAs using the value \( \gamma = 1.67 \) and SiGe using \( \gamma = 1.69 \) are in a good agreement with the experiment. Surprisingly, these works said nothing about the dependence of diffusion on temperature.

This work is devoted to the study of phonon diffusion based on the Monte Carlo method. The main task is to establish the nature of time dependence in expression (4) and to determine the nature of phonon diffusion in silicon depending on temperature of a solid body. Formally, phonon interactions between sequential scattering at the boundaries were taken into account in [8]. However, the changes in the direction of the velocity vector and in the phonon frequency were not taken into account.

This task is necessary to get more information about the complex processes of heat transfer in solids [9]. However, this study has not only a fundamental nature, but also an important engineering application. It can give a tool to solve relevant thermal transport problems in nanostructures. Such as the study of diffusion-ballistic (quasi-ballistic) regime of heat transfer [10, 11]. Currently there is no justified formula for calculating the time between the sequential interactions of phonons with surface in conditions when it is necessary to take into account the interaction of phonons with each other and other possible processes [12].

2. Basics of phonon diffusion

Phonons are quasiparticles that describes a collective excitation of particles (atoms, molecules) in a crystal lattice. The physics of phonon diffusion is not similar with the Brownian one. There are a number of reasons for this.

1. Elastic waves in solid body has different modes of vibrations such as transverse and longitudinal, that corresponds to the same polarization types of phonons. While in a classical gases only one type of particles is considered.

2. The atoms and molecules in classical gases undergo pair interactions, contrariwise, in phonon gas there are triple interactions. Moreover, for high temperature there exists four-phonon interactions.

3. Each collision in phonon gas represents decay or merge of quasiparticles. There are two types of interaction processes: normal (N-processes) and Umklapp (U-processes). The relative contribution of each process to heat transfer is highly dependent on the temperature of the phonon gas, so diffusion processes must depend on temperature. In the case of Brownian motion, this dependence is determined by the value of the diffusion coefficient \( D \).

4. The mean free path of phonons depends significantly on their energy, which usually is not taken into account in Brownian diffusion analysis. Another important feature of phonon-phonon interactions is that the energy of a new particle is much different from the energy of the initial particles. Also the time between phonon-phonon interactions and the speed of phonons essentially depend on their energy (frequency) and polarization type. The consequence of this fact is a strong dependence of the mean free path on the frequency.
5. As the size of real crystalline structure and the lattice constant are finite, it must be emphasized that the dispersion of mean free path is limited. These distinctive features should be taken into account when developing methods of phonon diffusion calculation to provide a real nature of phonons.

3. The calculation procedure

This work is devoted to the study of phonon diffusion based on the unique author’s Monte Carlo method. We explore the propagation of single particle, and after each interaction with another particle (three-phonon processes), we select one of the produced particles for further calculations. The whole procedure of phonon diffusion can be conveniently represented in the form of a block structure that consist of the following elements: initialization unit, conservation law unit, position recalculation unit. Let us discuss each of them.

3.1. Initialization unit

First of all, it necessary to prepare the input data for calculation.

1. We need to divide the frequency spectra of LA (longitudinal) and TA (transverse) phonons into segments. It is proposed to divide it into \( N \) parts:

\[
\Delta \omega_{LA} = \frac{\omega_{LA,\text{max}}}{N}, \quad \Delta \omega_{TA} = \frac{\omega_{TA,\text{max}}}{N}.
\]

Thus, a set of fixed frequency values is introduced \( \omega_{LA,i}, \omega_{TA,i} \) where \( i = 1, 2, 3, \ldots, N \). The value \( \omega_i \) is defined as \( \omega_i = (i - 0.5)\Delta \omega \). Accordingly, the group speed of phonons for the value \( \omega_i \) is given by expression \( v_{g,i} = \partial \omega_i / \partial k \).

2. Setting the temperature \( T \) of the solid structure.

3. Preparing a random generator for phonons.

We had designed a special random number generator that allows us to take into account the temperature of a solid body. The core of the generator is based on the density of states (DOS) and Bose-Einstein distribution function. On the output we get a random frequency of phonon for particular type of polarization, that is satisfy to distribution of phonons in real solid structure for specific temperature.

4. It is necessary to write down analytical dependences for longitudinal and transverse elastic waves – dispersion relations – for wave number \( k_j = f(\omega) \). In this work we use an approximation of experimental data with polynomials of the fifth degree.

3.2. Conservation law unit

Methods for calculating the conservation laws of the quasi-momentum and phonon energy for \( N \)-processes and \( U \)-processes differ from each other. Therefore, the calculation procedure for each process is presented below.

\( N \)-processes. For normal processes there are several ways how phonon can be involved in interactions: \( TA + TA = LA \) and \( TA + LA = LA \).

In this case we have two variants of the systems of equations of energy \( \omega \) and momentum \( \vec{k} \) conservation:

- the merge of the phonons:

\[
\omega_1 + \omega_2 = \omega_3, \quad \vec{k}_1 + \vec{k}_2 = \vec{k}_3;
\]

- the decay of the phonon:

\[
\omega_1 = \omega_2 + \omega_3, \quad \vec{k}_1 = \vec{k}_2 + \vec{k}_3.
\]

Below we present the algorithm that ensure the conservation law of energy and momentum, and allow to determine the direction of the vector \( \vec{k} \).
Step 1. Preparing value $\omega_1$ for the corresponding absolute value $k_1$ and direction of the vector $\vec{k}_1$ from the previous interaction.

Step 2. We get $\omega_2$ and $k_2$ using a random number generator presented in Initialization unit.

Step 3. Calculating $\omega_3$ from the corresponding conservation equation, and after that we get $k_3$ from dispersion relation for corresponding polarization of phonon 3.

Step 4. Using the law of cosines we determines the direction of wave vectors that relatives to the first vector $\vec{k}_1$:

$$\cos(\alpha_i) = \frac{k_1^2 + k_2^2 - k_3^2}{2k_1k_2}.$$ 

Here it remains uncertain which side of $\vec{k}_1$ (left or right) the $\vec{k}_2$ and $\vec{k}_3$ vectors should be placed. The position can be set, for example, using a random number generator. Hence the problem is completely solved.

$U$-processes. At the beginning, we will make two preliminary remarks. First, it is obvious that in $U$–processes only interactions with the merge of particles are realized (6). Second, after determining the value can be set in two ways: either on the branch of the dispersion relation within the Brillouin zone (N-process), or outside of the Brillouin zone (U–process). In first case, we go to unit of N-processes calculation. In second case our further actions are unambiguous (we will get Umklapp processes).

Let us consider step-by-step actions in case of U–process analysis. The first three steps are similar to previously presented algorithm for N-processes.

Step 4. Us the end of the wave vector $\vec{k}_3$ is outside the Brillouin zone, we use the inverse lattice vector $\vec{G}$ to determine a new value of the third vector $\vec{k}_3'$ which is inside the Brillouin zone: $\vec{k}_3' = \vec{k}_3 - \vec{G}$, instead of $\vec{k}_3$.

Step 5. Now we have a set of absolute values of wave vectors $k_1$, $k_2$, $k_3'$, and using them, we perform calculations that similar to N-processes algorithm. So we know the direction and absolute value of the vector $\vec{k}_1$. Direction of vectors $\vec{k}_1$ and $\vec{k}_3'$ are determined using well-known equations. Thus, we ensure the implementation of the law of conservation of momentum.

3.3. Position recalculation unit
Using the absolute value and the direction of the wave vector, as well as the value of the phonon energy obtained in the previous step, we determine a new position where the next process of three-phonon interaction will occur. To calculate the displacement the follow expression is used:

$$l_i = v_{g,j}(\omega_i)\tau_j(\omega_i), \quad l_{x,i} = l_i \cos(\alpha_i), \quad l_{y,i} = l_i \sin(\alpha_i),$$

here $v_{g,j}$ – velocity for a given phonon frequency and polarization $j$ obtained from the dispersion relation; $\tau_j$ – time between successive phonon interactions. We use relaxation time of phonon from Ward-Broido ab initio calculations [13]:

$$\tau_{N,j}^{-1}(\omega,T) = A_{N,j}\omega^2 f(T),$$
$$\tau_{U,j}^{-1}(\omega,T) = A_{U,j}\omega^4 f(T),$$
$$f(T) = T \left[ 1 - \exp \left( \frac{3T}{\theta_D} \right) \right],$$

where $A_{N,j}$ and $A_{U,j}$ – coefficients for N- and U-processes for $j$-polarization of phonons, $\theta_D$ – Debye temperature.

Figure 1 illustrates how the times $\tau_{N,j}$ and $\tau_{U,j}$ for different processes and polarization types $j$ correspond with each other for $T = 300$ K. It is seen that in the low frequency region N-processes dominate, and in the high-frequency region U–processes prevail. The essential feature is a strong
scatter of values $\tau_j(\omega_i)$ (by several orders of magnitude) when the frequency changes from a minimum value to a maximum (paragraph 5 of section 2). In comparison the dependance $\tau$ from frequency $\omega$ for $T = 15$ K looks the same as on figure 1 but the value is about $10^2$ above.

4. Results

Using our unique algorithm of phonon diffusion calculation from section 2 we developed the program code and performed computer simulation of phonon diffusion in a two-dimensional sample of pure silicon. The phonon velocity was determined from the dispersion relations obtained for the direction [100] in the isotropic approximation [14].

Figure 2 (a) shows the results of phonon diffusion simulation at a temperature 15 K when normal processes are dominated. At this regime the mean free path of phonons are large (phonons move in body without collisions).

In figure 2 (b), at 300 K Umklapp processes is dominated, so the mean free path is several orders of magnitude lower then for 15 K (figure 2) (a).

As can be seen from the figure 2 (a,b) there are large clusters of small track lengths, which are connected by lines representing large free path lengths of phonons. It can be seen that there are two types of behavior of diffusing particles. First one is represented displacement with a small free path length which coincides with the Brownian motion. These clusters are connected by sets of tracks with large free path lengths, which are similar to so-called “Levi flights” relating to the diffusion of Levi. They appear because our model took into account the dependence of time between sequential interactions of phonons with each other on frequency. Therefore, at random transitions from the region of high frequencies (small mean free paths) to the region of low frequencies (large mean free paths), we obtain areas corresponding to small and large free path lengths.

Diffusion calculations have shown that at a temperature of 15 K (low temperatures) only normal processes take place, and at a temperature of 300 K (room temperatures) the greatest role is played by the Umklapp processes (the number of processes reaches 40%) because they rapidly limit the mean free path.

At the same time, the number of U-processes cannot be more than half, since all processes of phonon decay are N-processes.
Figure 2. Diffusion of phonons at $T = 15$ K (a) and at 300 K (b). Trajectories of phonons that have $10^4$ interactions with other phonons.

5. Conclusion
The paper presents a new method for calculating diffusion processes in phonon gas, and a unique algorithm that allows to make a computer simulation of phonon diffusion. Obtained solution is associated with the heat transfer in solid nanostructures.

The developed method takes into account simultaneously all the main physical features that determine the nature of phonon diffusion: the influence of N- and U-processes, solid temperature, dispersion of phonons, the presence of different polarizations of phonons, the time dependence between successive interactions of phonons on the frequency. Analogous to such kind of works wasn’t found.

The results show that phonon diffusion is the qualitatively different from the classical Brownian diffusion. The main reason is associated with the fact that relaxation time of phonons differs by several orders of magnitude with frequency (energy), which is absent in the case of Brownian diffusion. This circumstance is determined by the fact that phonon-phonon interactions depend on the energy of quasiparticles, as well as by the fact that these interactions are accompanied by the decay or merge of phonons.

The presented tool for modeling can be used in the future to solve specific problems of heat transfer in solids, such as phonon scattering at the boundaries, the calculation of the Kapitza resistance (thermal contact resistance) between interfaces in superlattices.

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