Modeling of phonon diffusion using a Monte-Carlo method based on physics of phonon

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Abstract. Modern studies of heat transfer in semiconductor and dielectric structures (integrated circuits, etc.) are based on the analysis of the interaction of quasi-particles called phonons – quanta of elastic waves propagating inside the crystal lattice. In this paper, it is shown that to describe the diffusion it requires a detailed analysis of the physics of phonons interactions. Firstly, we formulated a method to perform a phonon diffusion calculations which takes into account the peculiarities of phonon interaction. Secondly, we developed a brand-new algorithm, which allow to carry out a computer simulation of phonon diffusion. Obtained results are necessary both for a deeper understanding of the processes of heat transfer, and for future calculations of the contact thermal resistances and phonon scattering on the boundaries – the relevant problems of heat transport in nanostructures.

1. Introduction

The thermophysical properties of semiconductors and dielectrics are defined by the properties of phonon gases. Phonons are the quants of elastic waves propagating in solids – quasi-particles with zero-mass, whose energy is equal to ω, quasi-momentum is k (where k is the wave vector), the phonon group velocity v = dω/dk, is determined from the dispersion relations ω(k) [1]. Each material requires special studies to establish its most important properties. These are, firstly, thermodynamic properties, for example, the heat capacity of materials, their thermal expansion, elastic constants, and others, depending on temperature. Secondly, the kinetic properties, for example, thermal conductivity, electrical conductivity of materials (for semiconductors), and others, also depending on temperature.

The properties of phonon gases are depended on the features of the lattices of solids.

1) In particular, different solids contain different sets of phonons. For example, silicon contains a double set of transverse (TA) and a single set of longitudinal (LA) acoustic phonons; in comparison graphene contains set of longitudinal (LA), transverse (TA), and flexural (ZA) acoustic phonons. In addition, there are optical phonons (TO, LO), which are usually not taken into account when calculating the thermal conductivity.

2) Another important feature of phonon gas is the range of frequencies and wave numbers limited by maximum values ωmax and kmax. The properties of phonon and phonon gas depend on the geometry of the atomic lattices of solids, namely, they are determined by the vibration spectra of these systems,
and, to fully determine these properties, both in harmonic and in anharmonic approximations, an analysis of the vibrations is necessary.

3) Phonon interactions have a list of principle differences from classical gases.

First of all, in phonon gas the mainly interactions are not a pair but the triple one (figure 1). They are accompanied either by the decay of one phonon into two, or by the merging of two phonons into one:

a) the merge of the phonons

\[ TA + TA \rightarrow LA \quad \text{and} \quad TA + LA \rightarrow LA \]

\[ k_1 + k_2 = k_3 ; \]

(1)

b) the decay of the phonon (the reverse processes)

\[ k_1 = k_2 + k_3 . \]

(2)

Figure 1. Merge and decay of phonons.

Secondly, in contrast to classical gases, for phonons there is no concept of cross section of interaction. Instead, the analysis of the anharmonic properties of the solid lattice determines the average times \( \tau \) of phonon interaction with each other that is depended on the temperature and frequency (energy) of the phonon. These times are determined from the analysis of lattice vibrations of a solid in anharmonic approximation. Therefore, the mean free path of the phonon is defined as

\[ l(\omega, T) = v(\omega) \cdot \tau(\omega, T) . \]

Finally, there are two types of phonon interaction. The first type is the so-called normal processes (N-processes). They are processes, by which energy and momentum are conserved during interaction. And they are realized in the region of any energy (frequency) and temperature of phonon. Another type of processes are the so-called Umklapp processes (U-processes), or processes with flip. They are realized in the region of temperature more than 50 Kelvin and only when merge of phonons happened.

In the case of N-processes the so-called vector of the reciprocal lattice, \( \mathbf{k} \), is equal to zero,

\[ \mathbf{k}_1 + \mathbf{k}_2 = \mathbf{k}_3 + \mathbf{K} \]

(3)

in the case of U-processes, \( \mathbf{K} \) is a constant and the absolute value equal to \( 2\pi / a \), there \( a \) is the lattice constant. The most important feature of the vector \( \mathbf{K} \) is that it presence in the quasi-momentum conservation equation simultaneously conserves energy of the system.

Thus there are numerous factors which influence on the interaction of phonons with each other. The multifactorial nature of phonon gas makes the study of the kinetic properties significantly complicates, especially when it comes to solve the problem using Monte Carlo method. In many cases, presented task needs substantial simplifications for the interaction processes. The main object of this work is to develop the Monte Carlo method which takes into account, if possible, all the above processes. In addition, takes into account phonon scattering processes on impurity atoms in the lattice.

In regard to these, in this work we consider the relatively simple task of studying the diffusion of phonons in its own gas, which allows to strictly take into account all the main features of the interaction of phonons with each other. As an example, we consider the phonon gas in silicon that is a widespread semiconductor material in electronics.

Here we discuss a new method of calculation and show the results in a wide range of temperature. Presented model takes into account the temperature dependence of diffusion coefficient of phonon gas, the frequency (energy) dependence of different interaction processes (N- and U-processes).

Next task is to determine the type of diffusion processes that happened in phonon gas. This problem arises precisely in connection with a large number of factors that represent the interaction of phonons. At present the classification of diffusion processes is based on the time dependence of the standard deviation of a particle from its initial position that generally has the form [2, 3]:
\[
\left\langle (r(t) - r(0))^2 \right\rangle = Cr^2, \\
\]
where \( C \) is a constant.

a) case \( \alpha = 1 \) corresponds to the classical Brownian diffusion obtained by Einstein in 1905 [4];

b) \( \alpha < 1 \) is the so-called subdiffusion;

c) \( \alpha > 1 \) is superdiffusion (Levy diffusion). It is characterized by the appearance of very large wavelengths – “Levy flights”.

The task to determine the type of diffusion is reduced to calculation of \( \alpha \).

Noted that, at present there are several works dedicated to analysis of phonon diffusion [5, 2, 3]. In article [4] there is no data on how the calculation was performed. It is clear that the calculations were carried out for silicon only for \( U \)-processes of phonon interaction, as well as interactions with impurities and boundaries of solids. In the second work, only phonon-phonon interactions for semiconductor alloys are taken into account. In both works calculations were performed for one temperature. However, very long free path of phonons was relatively rare observed, which resemble the so-called “Levy flights” characterized of Levy diffusion. In the work [2, 3] an approximation was proposed within the framework of which a restriction on the phonon mean free path was introduced. In this work it is argued that for semiconductor alloys Levy diffusion occurs at the initial stage which transfers into Brownian diffusion by time (“truncated diffusion” of Levi [6, 7]).

The main task of the work is to determine the nature of phonon diffusion in silicon depending on time and temperature of a solid body. Formally, phonon interactions between sequential scatterings at the boundaries were considered 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, 13].

2. Calculation method

In this work we take into account the following features of the physics of phonon.

1. Three-phonon interactions: merge (1) and decay (2).
2. \( N \)- and \( U \)-processes (3).
3. Phonon interactions with impurities.
4. Dispersion equations in the quasi-isotropic approximation for lattice direction \([100]\).
5. Average times between different interaction processes in phonon gas.
6. The calculations are performed in two-dimensional approximation in a wide range of temperatures.

The whole procedure of phonon diffusion can be conveniently represented in the form of a block structure that consist of the following units.

2.1. Initialization unit

The first block includes the initial data for calculations. Data, such as the total number of steps, time steps, enumeration of types of interactions taken into account and interaction processes, are given.

To define the properties of each phonon we create a special random generator that allows to take into account the temperature of a phonon gas. The core of the generator is based on the density of states (DOS) and Bose-Einstein distribution function. It help us to get a random frequency of phonon for particular type of polarization.
2.2. Lattice dynamic unit
This block contains the properties of phonons including the dispersion relations for longitudinal and transverse phonons, as well as the average times \( \tau(\omega, T) \) of phonons between successive interactions for different processes and different polarization [14]:

\[
\tau_{N,j}^{-1}(\omega, T) = A_j^N \omega^{-2} f(T), \quad \tau_{U,j}^{-1}(\omega, T) = A_j^U \omega^{-2} f(T), \quad f(T) = T \left[ 1 - \exp \left( \frac{1}{\theta_\omega} \right) \right]
\]

where \( A_j^N \) and \( A_j^U \) – coefficients for \( N \)- and \( U \)-processes for \( j \)-polarization of phonons (TA and LA), \( \theta_\omega \) – Debye temperature.

2.3. Conservation law unit
This part includes the random selection of phonon interaction processes (\( N \)- and \( U \)-process or impurity scattering) and further calculations in two-dimensional assumption.

2.3.1. Block for \( N \)-processes
Block represents information on the processes of merge (1) and decay (2) of longitudinal and transverse phonons, including the corresponding laws of conservation of energy and quasi-momentum. In addition, a random number generator is included, that is necessary to select the energy of one of the interacting phonons and determine the absolute value of its quasi-momentum. Finally, the means for determining the directions of the wave vectors of two phonons by constructing the corresponding triangle.

2.3.2. Block for \( U \)-processes
This part of algorithm contains all the positions of the previous block 2.3.1, as well as additional data – a special form of the law of conservation of quasi-momentum which includes the reciprocal lattice momentum (3).

2.3.3. Block for impurity scattering
The fourth block is the calculation of two-dimensional phonon interactions with impurity atoms. The possibility of interaction processes of this type and the corresponding law of conservation of quasi-momentum is included in the random number generator (the law of energy conservation in this case is performed automatically).

2.4. Position recalculation unit
This block calculates the mean free path of phonons between interactions using the follow expression:

\[
l_i = v_i(\omega_i) \cdot \tau_i(\omega_i), \quad l_{ij} = l_i \cos\alpha_{ij}, \quad l_i = l_i \sin\alpha_{ij},
\]

here \( v_i(\omega_i) \) – velocity for a given phonon frequency and polarization obtained from the dispersion relation; \( \tau_i(\omega_i) \) – time between successive phonon interactions. Using the law of cosines we determine the direction of wave vectors that relatives to the first vector, \( \mathbf{k}_i \):

\[
\cos\alpha_{ij} = \frac{k_{ij}^2 + k_i^2 - k_j^2}{2k_i k_{ij}}
\]

We apply a special random generator [15] and determine the real time of the flight of a phonon between its successive interactions.

The general algorithm that represents the corresponding units is shown on figure 2.

3. Results
We apply our unique algorithm (figure 2) and develop the program code that performed computer simulation of phonon diffusion in a two-dimensional sample of pure silicon. Here we present some important results.

1. Position of phonon-phonon interaction processes – the displacement in space relative to the position of the starting point \([0,0]\) at equal intervals of time (figure 3).
Figure 2. A general algorithm of direct Monte-Carlo simulation of phonon diffusion.

Figure 3. Diffusion of phonons. Trajectories of phonons that have interactions with each other.

2. Calculation of phonon diffusion coefficients depending on temperature (figure 4).
3. The fraction of merge and decay processes of phonon:
   a) merge (64%)
      \[ T_A + T_A \rightarrow L_A - 19\% \quad \text{and} \quad T_A + L_A \rightarrow L_A - 45\% \]
   b) decay (36%)
      \[ L_A \rightarrow T_A + T_A - 19\% \quad \text{and} \quad L_A \rightarrow T_A + L_A - 17\% \]

4. Calculation the fractions of N- and U-processes (figure 5), fractions of various types of phonon interactions, contributions of phonons with different energies to these processes and etc.

4. Conclusion
   The paper presents a new method of calculating the diffusion processes in phonon gas, and formulate unique algorithm (figure 2) that allows to make a computer simulation of phonon diffusion. And finally we obtain the following results.
   1. The general picture of phonon diffusion that represents the changes in displacement with time.
   2. Dependence of \[ \langle [r(t) - r(0)]^2 \rangle = f(t) \].
   3. Statistics of N- and U-process.
   4. Effect of temperature on the characteristics of phonon diffusion.
   5. Statistical data on mean free paths, different types of interactions and others.

The presented algorithm for phonon diffusion modeling can be used to solve the 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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**Acknowledgment**
The work is supported by Ministry of Science and Higher Education of the Russian Federation, project 16.8107.2017/6.7.