Analysis of diffusion processes in a phonon gas

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Abstract. Diffusion processes in a phonon gas were studied in a wide temperature range in the application to silicon. To provide this analysis we develop a new method of computation based on the kinetics of phonon gas in combination with Monte Carlo simulation. This model for the first time allows one to obtain the diffusion coefficients, the influence of different types of interaction processes of phonons on diffusion and other information about processes in phonon gas. Presented method is useful in the study of heat transfer in nanostructures.

1. Introduction

Interest in the study of phonon diffusion is associated with the characteristics of phonon interactions with each other. These interactions are multifactorial, which significantly complicates the study of diffusion. In particular, it is necessary to answer the question whether it belongs to Brownian diffusion or not. The characteristic of diffusion is determined by the dependence of the mean square of the displacement from the starting point on time:

\[ \langle r^2(t) - r^2(0) \rangle = Ct^\alpha, \]

where \( C \) is a constant. According to the value of power \( \alpha \), there are three different types of diffusion.

1) The case \( \alpha = 1 \) corresponds to the classical Brownian diffusion (obtained by Einstein in 1905);
2) The case \( \alpha < 1 \) is the so-called subdiffusion;
3) The case \( \alpha > 1 \) is superdiffusion (Levy diffusion). It is characterized by the appearance of very large wavelengths that is called “Levy flights”.

At present there are two well-known works devoted to the phonon diffusion [1, 2] in application to semiconductor alloys. It is shown that the diffusion depends on the time from the moment of observation. At the beginning the diffusion processes belong to the Levy diffusion, and then gradually pass to the Brownian diffusion.

The question of the type of diffusion is reduced to the determination of the value \( \alpha \). As the calculations were carried out, articles [1, 2] tell practically nothing about it and the influence of temperature on the obtained results are not mentioned. Phonon diffusion was also studied in [3] where examples of phonon tracks for a constant temperature are presented. There is also the presence of long flight, which is similar to Levy diffusion but there is no information about how to carry out these calculations.

In this paper, we introduce a diffusion analysis in so called “phonon gas” using the direct Monte Carlo simulation. The main task is to obtain sufficiently detailed information about the processes taking place in a wide temperature range. To solve this problem it is necessary to take into account all the phenomena, which accompanying the interaction of phonons with each other [4]. The fact is that
these interactions are much more complex than, for example, the interactions of atoms with each other. Therefore, a more complex apparatus is required for their description [5].

The importance of developing such apparatus is connected with the following tasks. First one is to identify the influence of various factors on diffusion processes, and second one is to solve the problems arising from the analysis of transfer processes in nanoscale structures. For example, it will give an opportunity to study the thermal boundary resistance that is ordinary considered under the assumption of the acoustic mismatch model [6] and heat transfer calculation in multilayer nanoscale structures [7, 8]. All of the presented calculations refer to the phonon gas in silicon. This means, in particular, that the dispersion relations, the energy dependences of the mean phonon lifetime and etc. were obtained for the silicon lattices.

2. Model of the phonon diffusion

First of all, by definition, phonons are quanta of elastic waves propagating in solids. These are quasiparticles with zero mass, whose energy is \( \hbar \omega \), quasi-momentum is \( \hbar k \), group velocity is \( v = \partial \omega / \partial k \), where \( \omega(k) \) is the dispersion relation that describes the vibrations of the atoms in crystal lattice. In this paper we provide calculations for silicon because its properties are well studied both theoretically and experimentally.

Phonon interaction processes exhibits specific behavior. Firstly, in each material there are at least two types of phonons (usually longitudinal and transverse). Secondly, the main role is played by interactions of three phonons (four phonons is relatively rare and therefore usually this situation is not taken into account). And thirdly, as a result of this, either the initial phonon decays into two phonons, or the phonon merges with another phonon so a new phonon is appeared. Thus in contrast to the classical gas, in this case it is not the diffusion of a single particle that is moving in a gas of the same particles, but the diffusion of quasi-particles that are successively replacing each other. Accordingly, as a result of the interaction of phonons, not only energy and quasi-momentum could be changed, but also the type of phonons could be changed. In silicon and in other materials there are two types of phonons: longitudinal and transverse.

1) First of all, it is necessary to determine the polarization of phonons. In this work only longitudinal and transverse acoustic phonons \( LA \) and \( TA \) are taken into account. Optical phonons are not explored since their contribution to heat transfer processes is negligible. The following processes of phonon interaction with each other are considered [9]:

\[
TA + LA \rightarrow LA, \quad TA + TA \rightarrow LA.
\]

2) The next step is to take into account the processes of phonon-phonon interactions, specifically the so-called normal processes (N-processes) and processes with a flip (U-processes). Normal processes interaction occurs with conservation of energy and quasi-momentum:

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

The existence of processes with a flip is due to the fact that the frequencies and quasi-momentum of phonons are limited by maximum values: \( \omega_{\text{max}} \) and \( k_{\text{max}} \). This restriction arises due to the discreteness of the atomic lattice of solids – the minimum distance that allows wave propagation. This value is called the lattice constant (the distance between neighboring atoms). In this connection then the merger of two phonons formally takes place, the total value of the quasi-momentum \( \vec{k} \) is greater than \( k_{\text{max}} \) in absolute value, but there are no such phonons so the problem is solved as follows. The transfer of momentum is occurred from the interacting phonons to the crystal lattice. This momentum is denoted as \( \vec{G} \) and its absolute value is equal to \( 2\pi/a \) (\( a \) is lattice constant). This solution provides the energy conservation of the system. Therefore the energy conservation equation and the quasi-momentum conservation equation are written in the form:

\[
\omega_1 + \omega_2 = \omega_3, \quad \vec{k}_1 + \vec{k}_2 = \vec{k}_3 + \vec{G}.
\]
Actually, the real phonon has a quasi-momentum equal to \( \vec{k}_3 = \vec{k}_1 + \vec{G} \). In this situation, \( |\vec{k}_3| < k_{\text{max}} \).

3) Thus it is necessary to define a criterion to determine the type of phonon interaction process (N- or U-process). We introduce the value of the angle \( \varphi \) between the vectors \( \vec{k}_1 \) and \( \vec{k}_2 \) of the phonons merging with each other. The critical value of the angle \( \varphi_{cr} \) is entered so if \( \varphi < \varphi_{cr} \), it is the U-process, and if \( \varphi > \varphi_{cr} \), it is the N-process.

4) An important characteristic of phonon interaction processes is the mean free path, moreover in practice we use the average time between successive interactions that is strongly dependent on energy [10]. This is the distinction from the classical gas where an interaction cross sections is the relevant property. A particular question is the free distribution of a phonon in the interval between successive interactions. This procedure is performed according to [11]. This method could determine the time between successive interactions with each other and the corresponding value of the free path.

5) The next important step is the introduction of temperature through the Bose - Einstein distribution function. The corresponding random number generator is tuned to select phonons by energy proportional to the values of this function at a given temperature.

6) We develop algorithms to provide diffusion calculations in two-dimensional and three-dimensional approximations. In the two-dimensional (2D) assumption the analysis of phonon interaction processes is carried out in the plane. In the three-dimensional (3D) assumption for each process of interaction random number generator is used to determine the plane in space where this interaction takes place. Next we realize the transition to this plane and conduct the operations related to the 2D analysis. After that we get back to three-dimensional space and provide the analysis of the free motion of the quasiparticle and to subsequent interaction of phonons.

7) Finally, the last block represents the results of calculations.

3. Results

We implement our unique method and provide numeric calculation using direct Monte Carlo simulation of phonon diffusion in a two-dimensional and three-dimensional assumption for a sample made of pure silicon. Here we present the following important results.

1. Firstly, we introduce the consecutive positions of the points in space where phonon-phonon interaction processes take place (starting from the initial point \([0,0]\)). In figure 1 three-dimensional diffusion is presented; in figure 2 there is two dimensional diffusion.

![Figure 1. Three-dimensional diffusion of phonons. Consecutive positions of \(10^3\) interaction points.](image)
2. We provide calculation of the dependence of the average value of the square of the phonon replacement from the starting point on time, $\langle L^2 \rangle = f(t)$, figure 3. Such dependences indicate the characteristic of diffusion and are represented in power functions $f(t) = C t^\alpha$. The type of diffusion is determined by the value $\alpha$. Figure 3 represents diffusion coefficient and its dependence on time; value $\alpha$ is about 1 that corresponds to the Brownian diffusion.

3. Thus, the factors that could determine the characteristic of phonon diffusion are: the fact that phonon interactions are accompanied by their decay, or merge, different types of phonon interactions, different types of interaction processes, in which phonons are involved, and the dependence of average times between successive interactions on energy. The fraction of various types of phonon interactions is shown in table 1.

| Process  | Formula          | Fraction |
|----------|------------------|----------|
| Merge    | $TA + TA \rightarrow LA$ | 19 %     |
|          | $TA + LA \rightarrow LA$ | 45 %     |
|          | $LA \rightarrow TA + TA$ | 19 %     |
| Decay    | $LA \rightarrow TA + LA$ | 17 %     |
4. Finally, the calculations of the ratio of $N$- and $U$-processes (figure 4) during phonon diffusion are presented.

![Figure 4. Probability of U-processes.](image)

**Conclusion**
In this work we present a new method for calculating the kinetics of phonon gas based on direct Monte Carlo simulation. For the first time this algorithm allows the study of the phonon diffusion in a wide range of temperature and gives opportunity to obtain the diffusion coefficients and other information about diffusion processes. The developed methods will be used in the future to solve practical problems related, primarily, to the processes of heat transfer in nanostructures such as the problem of the Kapitza resistance in multilayer nanoscale structures, scattering of phonons on the boundaries of the sample and etc.

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