Modified callaway model calculations for lattice thermal conductivity of a 20 nm diameter silicon nanowire

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The modified Callaway model is used to calculate Lattice Thermal Conductivity (LTC) for (20-nm) silicon nanowires diameter in the temperature range from 2K to 800K. Acoustic phonon mode and group velocity in the calculations are modified by spatial confinement of phonons with that of the boundary effects. All important scattering rates such as Umklapp, Mass difference, Resonance, and Boundary are calculated at room temperature. Room temperature LTC for this diameter is about only 10.23% of its bulk value. Numerical evaluation is also investigated and the results are compared to that of the reported experimental as well as theoretical data.

Keywords: Silicon nanowires; thermal conductivity; spatial confinement; phonon scattering.

1. INTRODUCTION

Thermal properties of semiconductor nanostructures are beginning to attract researchers in the last decades. It begins with quantum confined structure explanations for a continuous scaling down of the size in microelectronics devices and circuits. The case leads to an increase in power dissipation per unit area of a semiconductor chip to that of nanometers [1,2]. Scientists also attracts to the field of thermoelectric materials. Particularly the nanowires for their interesting technological applications, including visible and infrared light emitting diode, optical sensors, and solar cells. [3,4]. There exists a large amount of literature on thermal properties of nanomaterials, for example, Zhang et al. [5] studied the effect of length, diameter, orientation, shape and cross-section effects on thermal conductivity of silicon nanowires. Recently Guofeng et al. [6] studied the effect of Phonon coherence on lattice thermal conductivity of both superlattice and core–shell nanowires. Ibrahim et al. [7] showed the Carrier concentration effect and other structure-related parameters on lattice thermal conductivity of Si nanowires. Zhongwei et al. [8] investigated theoretically and experimentally the size-Dependent Phononic Thermal Transport in Low-dimensional Nano-materials. Researchers confirm that thermal conductivity in nanowires is suppressed due to two reasons, first as the diameter of the wire reduces to the order of phonon mean free path in the originally bulk materials, e.g. on the order of 10-100nm, phonon scattering by the boundary reduces the thermal conductivity, the second reason is that due to the size
confinement, that the phonon frequency $\omega$ versus wave vector $q$ depression is modified from that of the bulk [9] 
In this work, attempts will be made to calculate lattice thermal conductivity of 20 nm diameter silicon nanowires by using a relation applicable for bulk state with a modifications of acoustic phonon dispersions and group velocity under the phonon confinement effects.

2. THEORY

2.1. Phonon Boltzmann transport equation

The general form of thermal current is:

$$J_Q = -K \nabla T$$  

(1)

The phonon heat current under $\nabla T$ which is a temperature gradient is given by:

$$J_Q = \sum \tilde{N}_{q,s} \hbar \omega_S(q) V_S(q)$$  

(2)

where $\hbar \omega_S(q)$ and $V_S(q)$ are phonon energy and its group velocity in the direction of $q$, which contributes $\hbar \omega_S(q) V_S(q)$ to the heat current, $(s)$ refers to a particular phonon polarization type.

$\tilde{N}_{q,s} = N_{q,s}^0 - N_{q,s}$ is the deviation of the phonon distribution, from its equilibrium value , and its given by the Bose-Einstein distribution [2,10,11];

$$N_{q,s}^* = \frac{1}{e^{(\hbar \omega_S(q)/k_B T) - 1}}$$  

(3)

In order to obtain LTC, Boltzmann equation for $N_{q,s}$ to be solved as follows;

For a steady state case, the phonon Boltzmann equation can be written as [2,3,10];

$$\frac{\partial N_{q,s}}{\partial t}_{\text{drif}} + \frac{\partial N_{q,s}}{\partial t}_{\text{scatt}} = 0$$  

(4)

where $(\frac{\partial N_{q,s}}{\partial t}_{\text{drif}})$ represents the change of the phonon distribution according to $\nabla T$, in the form:

$$\frac{\partial N_{q,s}}{\partial t}_{\text{drif}} = -V_i(q) \cdot \nabla N_{q,s} = -(V_i(q) \cdot \nabla T) \frac{\partial N_{q,s}}{\partial T}$$  

(5)

However Values of $N_{q,s}$ are affects by scattering with other phonons, impurities, charge carriers, interfaces, boundaries, etc. all denoted to eq (4) by $(\frac{\partial N_{q,s}}{\partial t}_{\text{scatt}})$. In the relaxation-time approximation this can be written as [2,10,13];

$$\left(\frac{\partial N_{q,s}}{\partial t}\right)_{\text{scatt}} = \frac{N_{q,s}^0 - N_{q,s}}{\tau_{Cs}(q)}$$  

(6)

where $\tau_{Cs(q)}$ is the combined phonon relaxation time. Substituting eq. (5) and eq (6) in eq. (4) will give the phonon Boltzmann equation to a form:

$$-V_x \frac{\partial N_{q,s}}{\partial x} - V_y \frac{\partial N_{q,s}}{\partial y} - V_z \frac{\partial N_{q,s}}{\partial z} + \frac{\tilde{N}_{q,s}}{\tau_{Cs}(q)} = 0$$  

(7)

where $V_x$, $V_y$, and $V_z$ are the three components of phonon group velocity along the x,y, and z axis, respectively.

For free standing nanowires under a temperature gradient along z axis, eq. (7) becomes:

$$V_x \frac{\partial \tilde{N}_{q,s}}{\partial x} + V_y \frac{\partial \tilde{N}_{q,s}}{\partial y} + \frac{\tilde{N}_{q,s}}{\tau_{Cs}(q)} = V_z \frac{\partial T}{\partial z} \frac{\partial N_{q,s}}{\partial T}$$  

(8)

For a small phonon distribution deviation from its equilibrium,
\( \hat{c} N_q, s \hat{T} \) can be changed to \( \hat{c} N^s_q, s \hat{T} \). Then the linearized phonon Boltzmann equation of eq. (8) takes the form [2,3]:

\[
V_x \frac{\partial N_{q,s}}{\partial x} + V_y \frac{\partial N_{q,s}}{\partial y} + \frac{\partial N_{q,s}}{\partial z} = \frac{\partial T}{\partial z} \tau_{c_s}(q) \frac{\partial N^s_{q,s}}{\partial T}
\]

(9)

2.2. Calculations of the lattice thermal conductivity

For simplicity, the subscript \( q \) and \( s \) will be omitted, in the bulk state; the solution for Eq(9) will have the form:

\[
\bar{N} = \frac{\partial N^s}{\partial T} \nabla T \nabla \tau_c
\]

(10)

\( N \) can have further form of:

\[
\bar{N} = \frac{\partial N^s}{\partial T} \frac{\partial T}{\partial z} V \tau_c
\]

(11)

Equations (11), (3) and (2) gives a regular bulk formula for lattice thermal conductivity in the form [10],

\[
k = \left( \frac{K_B}{\hbar} \right)^3 \left( \frac{K_B}{2\pi^2 v_g} \right) T^3 \int_0^{\theta_D} \tau_c e^x \left( e^x - 1 \right)^2 dx
\]

(12)

Equation (12) is the Klemens-Galloway's expression for LTC in the bulk state, where \( x = \hbar \omega/K_BT \), \( h \) is Dirac constant, \( \tau_c \) is the combined relaxation time, and \( v_g \) and \( \theta_D \) is the phonon group velocity and Debye temperature respectively [2,3,10]. Callaway considered this phenomenological model in which he treated the speed of sound as a constant parameter. His calculations contained acoustic branches, while the contribution of the optical modes is neglected [12]. For nanowires eq (12) is used with an appropriate modification of the phonon group velocity with that of the combined relaxation time. Since phonon waves have different velocity and different energy [2,10].

2.3. Phonon dispersion and group velocity

The acoustic phonon dispersion in free standing nanowires are calculated by considering only the in-plane transport in the well and the main contributions, that comes from the modified longitudinal acoustic phonons.

For nanowires with a diameter \( D \), the confined phonons dispersion relation can be written as [10]:

\[
\left( q^2 - q_t^2 \right) \left( q_d D/2 \right) J_s(q_d D/2) - 2 q_d^2 (q^2 + q_t^2) + 4 q^2 q_d^2 (q_d D/2) J_s(q_d D/2) = 0
\]

(13)

where \( J_0 \) and \( J_t \) are ordinary Bessel functions and \( q \) is the phonon wave vector, the two parameters \( q_t \) and \( q_d \) are related by:

\[
q^2 = \frac{q_t^2}{v_{t,d}^2} - q_t^2
\]

(14)

Here \( v_t = \sqrt{(\lambda + 2\mu)/\rho} \) and \( v_t = \sqrt{\mu/\rho} \) are the longitudinal and transverse sound velocity in the materials bulk state respectively, \( \lambda, \mu \), is lame constant and \( \rho \) is the density. The phonon dispersion relation is written as:
\[ \omega_n = v_{d,t} \left( q^2 + q_{d,n}^2 \right)^{1/2} \]  

(15)

where \( \omega_n \) is the phonon frequency for the nth branch. Numerically solving this equation with that of Eq. (13) give the phonon dispersion curve for five values of \( q \) as shown in figure (1):

![Figure 1](image)

**Figure 1** Acoustic phonon dispersion relation for five lowest confined branches in a free-standing silicon nanowire having a diameter 20 nm, can be seen that spatial confinement leads to flattening the phonon branches.

The phonon group velocity is calculated for each branch by using the relation,

\[ v_n = \frac{d\omega_n}{dq} \]  

(16)

Since different branch have different group velocities, then this averaged overall contributing branch and calculated by using the following population averaged phonon group velocity equation [2,10,12,13]:

\[
V'(\hbar\omega) \equiv \sum_n v_{g,n}(\hbar\omega) \frac{N_n(\omega)}{\sum_n N_n(\omega)} = \sum_n v_{g,n}(\hbar\omega) e^{\frac{\hbar \omega}{k_BT}} \]

\[
\sum_n e^{\frac{\hbar \omega}{k_BT}}
\]

(17)

Hence \( v_{g,n} \) and \( N_n(\omega) \) are group velocity, number of the oscillator having frequency \( \omega \) for the nth mode respectively, index \( n \) refer to different s (mode) branches. According to this equation, the average group velocity versus phonon energy is obtained for 20nm nanowires diameter drawn as shown in figure (2), and the results are used to calculate its LTC.
Population averaged phonon group velocity as a function of phonon energy. The overall value of the average phonon group velocity is \((5.76 \times 10^5 \text{ cm/sec})\) which is about half of the phonon group velocity in the bulk \((8.47 \times 10^5 \text{ cm})\) (violet line).

### 2.4. Phonon relaxation times

The combined phonon relaxation time \(\tau_C\) is obtained from the Mathiessens rule [8]:

\[
\frac{1}{\tau_C} = \frac{1}{\tau_u} + \frac{1}{\tau_B} + \frac{1}{\tau_M} + \frac{1}{\tau_R}
\]  
(18)

Here, \(\tau_u\) is the three-phonon Umklapp scattering and is due to the inharmonic nature of the crystal potential energy, \(\tau_M\) is the mass- difference scattering of phonons, \(\tau_R\) is resonance scattering and \(\tau_B\) is relaxation times due to boundary [13].

However, Phonon spatial confinement lead is to modification of the phonon dispersion and phonon group velocity, and hence to a change of phonon scattering rate.

The relaxation time for Umklapp scattering given by Klemens as:

\[
\frac{1}{\tau_u} = 2\gamma^2 \left( \frac{K_BT}{\mu V_0} \right) \left( \frac{\omega_D^2}{\omega_D^2} \right)
\]  
(19)

Hence \(\gamma\) is the Gruneisen anharmonicity parameter, \(\mu\) is the shear modulus, \(V_0\) is the lattice volume, and \(\omega_D\) is the Debye frequency [2,10,13].

The relaxation due to the mass- difference scattering is calculated according to the following expression,

\[
\frac{1}{\tau_M} = \left( \frac{V_0 \omega_D^4 \Gamma_m}{4\pi^3} \right)
\]  
(20)

where \(\Gamma_m\) is the measure of the effect of the mass-difference scattering defined as:

\[
\Gamma_m = \sum f_i \left( \frac{(\Delta M_i - M^*)^2}{M^*} \right)
\]  
(21)

where, \(f_i\) is the fractional content of mass \(M_i\), it is different from \(M\), the mass of the main atom, and \(\Delta M_i = M - M_i\), for \(M^*\) to given by: \(M^* = \sum f_i M_i\), which is the average atomic mass. Since natural silicon contains a mixture of three main isotopes of 92% of Si\(^{28}\), 4.9% of Si\(^{29}\), and 0.1% of Si\(^{30}\),...
and 3.1% of $\text{Si}^{30}$, then the isotope scattering significantly contributes to the thermal resistance of the material [2,13].

Resonance scattering $\tau_R$ which is the process of phonon interaction with point defects that have some internal frequency of oscillation $\omega_0$, and can be calculated according to the following relation, [14]

$$\frac{1}{\tau_R} = \frac{R \omega^2}{(\omega_0^2 - \omega^2)^2 + \Omega \omega^2 \omega^2}$$

(22)

where $R$ depends on the point defects concentration, and $\Omega$ is damping related parameters, it is chosen to be zero, since, $\Omega \geq 0$ does not affect the calculation results [14,15].

$\tau_B$ which is the Boundary scattering rate is calculated by the following modified relations [10]:

$$\frac{1}{\tau_B} = \frac{v_g}{D} (1 - p)$$

(23)

Here $p$ is the roughens parameter. it represents the probability that the phonon is undergoing a specular scattering event at the interface, $p = 1$, means purely specular scattering. For purely diffuse scattering the equation reduces to;

$$\frac{1}{\tau_B} = \frac{v_g}{D}$$

(24)

This is the well-known formula in the Casimer limit [13,16]. Figure (3) shows all scattering rate of phonon as a function of frequency obtained from equation (18) to eq. (24).

![Figure 3](image)

**Figure 3** The phonon relaxation rate in a silicon nanowires of diameter $D=20$nm, due to different scattering mechanisms as a function of the phonon frequency such as; Umklapp (U), Mass- difference (M), resonance scattering (R) and Boundary scattering as a function of frequency at (300K).

3. RESULTS AND DISCUSSION
3.1. Calculation lattice thermal conductivity and different type of scattering

First, the dispersion relations of confined acoustic phonon modes in a free-standing cylindrical nanowire with a particular diameter. The numerical parameter used in the numerical solution was:

\[ v_l = 8.47 \times 10^5 \text{ cm/sec} \quad \text{and} \quad v_t = 5.84 \times 10^5 \text{ cm/sec} \],

are both obtained from the relations:

\[ v_l = \sqrt{\frac{C_{11}}{\rho}} \quad \text{and} \quad v_t = \sqrt{\frac{C_{44}}{\rho}}, \]

where \( C_{11} \) and \( C_{44} \) are elastic constant [17]. Figure (1) shows that only the first branch has a linear dispersion relation particularly for very small values of \( q \). \( q = 0 \) gives \( \omega = 0 \), that is the lowest mode for the confined phonon without having cut-off frequency unlike other higher mode. Due to the phonon confinement in nanowires extra branches of dispersion and velocities are produces for each polarization type in compared with the bulk state [10,13]. Theses phonon confinement, lead also to flattening the phonon dispersion branches and decrease the phonon group velocity in the nanowire. However, spatial confinement of the phonon can cause its quantized energy. Each \( q \) of the phonon give different values of energy, the latter is because the reduction in the dimension of sample leads to increase the effect of surface on the internal unit cells. In this case distortion in the potential periodicity at the surface, distorts the potential of the unit cells. The closer the unit cells to the surface the more influenced by the surface effect [23,18]. Phonon confinement cause changes in the dispersion relation. Such changes can modify the group velocity to a form suitable to obtain LTC in nanoscale solids. This means a lower values of effective phonon group velocity for a low dimensional structure compared to the corresponding bulk state material [19].

The overall value of the average phonon group velocity which is obtained, equal to \( 5.76 \times 10^5 \text{ cm/sec} \), where it is about half of the phonon group velocity compared to that of the bulk. This result is due to phonon spatial confinement that led to a modified dispersion relation and consequently changes the group velocity in the nanowires. The longitudinal sound velocity in the bulk Si is equal to \( 8.47 \times 10^5 \text{ cm/sec} \) [2,12]. The higher values of the phonon energy it oscillates about a constant value, and asymptotically reaches to a value of \( 5.2 \times 10^5 \text{ cm/sec} \).

Combined phonon relaxation rates calculated by using eq. (18). Following material parameters that have been used in the calculations: Gruneisen parameter \( \gamma = 0.56 \), density \( \rho = 2.329 \times 10^3 \text{ (kg/m}^3) \). The shear modulus which was estimated from the formula, \( (\mu = v_t^2 \rho) \) and was \( (\mu = 7.96 \times 10^{10} \text{ Kg/sec}^2 \text{.m}) \), isotopic factor \( \Gamma = 2.5 \times 10^{-4} \), lattice volume \( V_0 = 2.002 \times 10^{-29} \text{m}^3 \) and cut-off frequency \( \omega_c = 5.43 \times 10^{12} \text{ (1/sec)} \) [13]. For bulk state, the Umklapp scattering dominate over all scattering rates [13], while in nanowires the boundary scattering is dominates over all the scattering rate as shown in Fig(3). Perils point out that boundary scattering will become dominated when the phonon mean free path approach crystal dimension. For wires having a diameter of \( (D \leq 30\text{-nm}) \) the boundary scattering will dominate over all scattering process. [4,13,16].

For bulk material, the boundary scattering regime occurs only at low temperature, when the phonon mean free path becomes comparable to the sample size [21]. For nanoscale size materials, the phonon mean free path may reach the sample dimension even at room temperature. Resonance scattering mechanism can lead to a significant increase in the relaxation time but its influence is localized in the vicinity of the resonance frequency \( \omega_0 \) [10, 17]. However, for higher carrier concentration, resonance scattering becomes more important, and its effects reaches to the level of that of other relaxation mechanisms, the boundary scattering is dominating over all the scattering process until it reaches to a frequency equal to \( 8 \times 10^{12} \text{ (1/sec)} \) as shown in Fig (3), and this is due to the long wave length of phonons at this range of frequency, therefore, phonon wave does not sense of the impurity at this range.
After group velocity is found, the LTC calculated in the temperature range of 2-800K by using eq (12), and the results compared with that the bulk value. In order to illustrate the contribution of different scattering mechanisms to the thermal conductivity, it is limited only by the scattering process of Umklapp, mass-difference, and boundary scattering. Umklapp scattering effect on LTC at low and high temperature. The lattice thermal conductivity in silicon Nanowires at 300K is reduced to about 40% of its bulk value [13]. i.e, Umklapp process cases to drop the lattice thermal conductivity to about 2.5 times of the bulk values due to the spatial confinement of phonons as shown in Fig (4).

Figure 4 Lattice thermal conductivity as a function of temperature for silicon free-surface nanowire of diameter D=20nm. Calculated using effectively bulk formula in a cylindrical nanowires and bulk silicon at; A) low temperature, B) high temperature. According to this model the lattice thermal conductivity in the wire is reduced to about 25% of it is bulk value.
at 300K. The effect of Umklapp scattering on thermal conductivity (U), the Umklapp and mass-difference scattering only (U+M), and by all processes including boundary scattering (U+M+B).

For nanowires, the boundary scattering will become much more significant even at a higher temperature. Phonon transport in nanowires will experience stronger boundary scattering and depending on the boundary properties [12,13,23].

3.2. Comparison result with other models

Because the experimental and theoretical data for the 20 nm diameter Nanowires of silicon was not available to compare. In order to have comparable results to this work. Values of LTC for Si nanowires (20 nm) were calculated by using empirical relations obtained from the experimental data reported by Deyu Lie [22] as shown in figure (5).

Figure 5 Temperature dependence lattice thermal conductivity for Si nanowires, theoretical result represents by (solid line) and dashed lines ▲ and ■ represent Experimental result, from Deyu Lie work [22]. The numbers above lines denotes the corresponding different wire diameters.

To get values for such diameter (D = 20 nm) of silicon Nanowires, the lattice thermal conductivity for all diameters reported were taken at certain temperature 300K as an example in figure (5), and also for other temperatures then the LTC as a function of nanowires diameter was obtained and drawn as shown in Fig (6). The least square fitting to this dependence gives an empirical relation in the form of:

$$k=2\times10^{-5}D^{3}-0.0049D^{2}+0.66D-0.0445(W/m.K)$$

Equation (25), gives the lattice thermal conductivity at 300K for 20-nm nanowire diameters. The processes were repeated for all other temperature as shown in Fig. (7).
Figure 6 Lattice thermal conductivity as a function of Si nanowires diameter calculated using equation (25) at 300 K.

Figure 7 Values of thermal conductivity taken from Eq.(25): (A) Experimental curve, (B) Theoretical curve. The number beside each curve denotes the corresponding wire diameter. Values for D-20 nm are calculated by empirical relation formed for each temperature.
Figure 8 Comparison of the present work data (solid line) with experimental and theoretical results obtained from the Deyu Li data for thermal conductivity for a diameter 20-nm Silicon Nanowires.

The comparison between results of this work and Deyu Lie for 20 nm nanowire diameters of silicon are given in figure (8). Figure (9), show LTC calculated by different models and the one calculated by this work. LTC for a 20-nm diameter Nanowires was calculated from the data taken from theoretical curves calculated by methods of Callaway, Holland and (Dames & Chen) models compared to that of Deuy Li work [20-24].

Figure 9 Lattice thermal conductivity of a 20-nm diameter silicon Nanowires, from different models of : a) Callaway, b) Holland, c) Dames, d) Deyu Li [20,24], the solid line represent the present work.

The lattice thermal conductivity calculated by this work is similar to the results from the data taken by Dames to comparing with the experimental curves shown in Fig. (9).

4. CONCLUSIONS

Lattice thermal conductivity of Si nanowires is differing significantly from that as macrostructures. Strong modification of the phonon dispersion and group velocities due to spatial confinement of phonon lead to a significant increase of the phonon scattering and as a
result to the decreases of the lattice thermal conductivity. Boundary scattering dominate over all other scattering process. Numerical evaluation is a successful method to calculate lattice Thermal conductivity for Si nanowires. The model predications are in good agreement with available experimental and theoretical data.

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