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Development of A Combined Thermal Conductivity Prediction Simulator Including Conduction Electron and Lattice Vibration Effects

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1. Introduction

Evaluation of the thermal conductivity of materials provides extremely relevant information for the development of nano-scale devices, particularly those including impurities, defects, interfaces, etc. This is because a large amount of heat is generated in electronic devices involving high frequency transistors, light emitting diodes and display devices. Consequently, technologies oriented to deal with heat sinking are required.

In general, thermal conductivity can be classified in terms of two kinds of effects, i.e. lattice vibration and conduction electron. Lattice vibration plays the dominant role in semiconductors and insulators. For metallic materials, conduction electron is the dominant one. In order to evaluate the thermal conductivity of materials, several simple methods have been reported [1-3]. The design of nano-scale devices which are composed of complex materials including semiconductors, insulators and metals with impurities defects dopants etc.; however, require the evaluation of both conduction electron and lattice vibration effects. Therefore an estimation method of both effects is required.

In this work, we have succeeded in the development of a combined thermal conductivity predictor based on two kinds of evaluation methods i.e., classical molecular dynamics (classical MD) and tight-binding quantum chemical molecular dynamics (tight-binding QCMD). To evaluate the thermal conductivity of complex materials, first of all, we estimated the lattice vibration effect using classical MD simulator. To estimate thermal conductivity by conduction electron, the Wiedemann-Frantz law is applied using a recent method proposed for electrical conductivity prediction based on our tight-binding QCMD system [4].

2. Computational Method

We have developed combined methods for estimating the thermal conductivity of materials. For the evaluation of thermal conductivity by lattice vibration, our original thermal conductivity evaluation method "THERMOSIM" which is based on classical molecular dynamics for solids and liquids was applied. The model is illustrated in Fig. 1. A measurement area is placed between fixed layers. In this method, the upper part of the measurement region is set at a constant high temperature and the lower region is set at constant low temperature, then the amount of energy transfer from the high temperature region to the low temperature region through the measurement region is monitored. The thermal conductivity λ is evaluated using the average of the transferred energy, as:

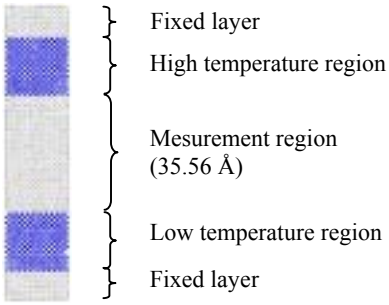

Fig. 1 Calculation model of diamond for lattice vibration evaluation method based on classical molecular dynamics simulation.

$$\lambda = QL/\Delta T\, S\Delta t \quad (1)$$

where Q is the transferred energy per MD step, L is the distance between the high and low temperature regions, S is the cross section of the model, ΔT is the difference in temperature between high and low temperature regions. Δt is the integration time of 1 simulation step.

For effect of conduction electron, where thermal conductivity by conduction electron plays the dominant role on metallic materials, the spatial distribution of probability density and energy levels of molecular orbital are firstly calculated using our original tight-binding QCMD program, "Colors". Based on this information, the electrical conductivity is calculated using our original electrical conductivity evaluation method. Our original tight-binding QCMD method can be applied to calculate the electronic structure of large-scale and complex systems, which cannot be calculated with conventional first-principles methods that are computationally expensive. Thermal conductivity λ is evaluated as follows:

$$\lambda = LT\sigma \quad (2)$$

where L is Lorentz ratio, T is the temperature, and σ is electrical conductivity.

3. Results and Discussion

For evaluation of the thermal conductivity caused by lattice vibration, a diamond model consisting of five layers was prepared, the fixed upper and lower layers, high and low temperature layers, and measurement region. A preliminary relaxation calculation was executed at 298 K. The temperatures at the high and low temperature regions were set to 323 and 273 K respectively. The transferred energy from high to low temperature regions through the measurement region was evaluated using a classical MD method. Using the transferred energy for one simulation step, the thermal conductivity was evaluated according to equation (1). Computed and experimental values for diamond are shown in Table I [5]. Values for graphite, amorphous SiO_2, borosilicate glass, cubic ZrO_2, ice, hydrate and water are also shown in Table I. The table shows excellent agreement of the lattice vibration based values with experimental data.

Table I Evaluated results of thermal conductivity by lattice vibration effect based on classical molecular dynamics method and experimental data.

	Thermal conductivity (W/mK)	
	Calc.	Exp.
Diamond	2935	900-2300
Graphite	1301	600-800
Amorphous	1.95	1.38
Borosilicate	1.11	1.1
Cubic ZrO_2	3.05	3.3
Ice	1.14	2.47
Hydrate	0.26	0.45
Water	0.38	0.61

On the other hand, thermal conductivity caused by conduction electron that plays a dominant role in metallic materials was evaluated according to the Wiedemann-Frantz law using our original electrical conductivity evaluation method based on our original tight-binding QCMD calculation for transition metal Ti and a typical metal such as Sn. The calculation model under three-dimensional periodic boundary condition is hexagonal, the closest packing cell includes 64 Ti atoms and the diamond structure includes 64 Sn atoms.

In this method, firstly, we calculated the electrical conductivity according to Drude's model, using our original electrical conductivity evaluation simulator. This simulator has two calculation steps. The first is the calculation of the electronic structure using our original program "Colors". For instance, the calculated spatial distribution of probability density for the lowest unoccupied molecular orbital of Ti is shown in Fig. 2.

The second step is the estimation of the values corresponding to the mobility using Monte Carlo simulation based on the spatial distribution of probability density and the values corresponding to the number of carriers based on Fermi distribution considering thermal excitation. In order to compute values corresponding to carrier mobility using the Monte Carlo simulation, the calculation models were divided at 0.3 Å intervals and the probability density was allocated to each lattice point. The frequency of passes the between starting point and the terminal in 1,000,000 steps was counted. Moreover, the number of thermal excited electrons was counted based on 300 K Fermi distribution. According to Drude's model, the electrical conductivity is equal to the number of carriers multiplied by the elementary electrical charge and the carrier mobility. The electrical conductivity of each molecular orbital was evaluated. Then the electrical conductivity of the material was evaluated by summation of all molecular orbitals. Result of electrical conductivity estimation is shown in Table II. From Table II, it can be appreciated that these simulation results are in excellent agreement with experimental values.

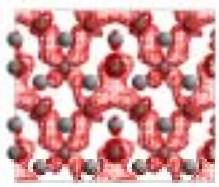

Fig. 2 The calculated spatial distribution of probability density for LUMO of Ti.

Table II Evaluated results of electrical conductivity for Ti and Sn with experimental data.

	Electrical conductivity	
	Calc.	Exp.
Ti	2.9×10^4	2.4×10^4
Sn	1.6×10^5	0.9×10^5

Using the computed electrical conductivity values, thermal conductivities were evaluated according to equation (2). Here, 2.44×10^{-8} (V^2/K^2) was adopted as Lorentz ratio. Calculated results were shown in Table III. Table III illustrates the good agreement of our calculated values with experimental data.

Table III Evaluated results of thermal conductivity for Ti and Sn with experimental data.

	Thermal conductivity	
	Calc.	Exp.
Ti	21.2	21.9
Sn	118	67

4. Conclusions

We have succeeded in the development of a combined thermal conductivity predictor considering both conduction electron and lattice vibration. For various kinds of semiconductors, insulators and metals, it was shown that classical MD and our new method using Wiedemann-Frantz law and our electrical conductivity evaluation method is effective. From these results, using these methods, evaluating the thermal conductivity of many kinds of materials has become possible.

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