Construction of a Novel Method of Measuring Thermal Conductivity for Nanostructures

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Abstract

With the aim of characterizing the thermal conduction in a nanometer-scaled materials, we have constructed a novel method on the basis of an ac calorimetric method. In this method, periodic sample heating is performed by light irradiation and the corresponding periodic temperature is detected by infrared irradiative thermometer. This makes us measure the thermal diffusivity out of contact with the objective sample. In the present study, we confirm to measure the thermal diffusivity of bulk Si and Cu by this non-contact method with halogen-lamp irradiation. In determining the thermal diffusivity from the relationship between distance deviation and delay time, the simplest wave equation is used, and the obtained values of thermal diffusivity for Si and Cu are close to those reported. Therefore, this non-contact method is useful for evaluating the thermal conduction and applicable for nanometer-scaled materials by improving local heating and local detecting systems.

Keywords: thermoelectrics, thermal conduction, nanostructure, ac calorimetry, thermal diffusivity

1. Introduction

Recently, intensive researches have been conducted for the development of power generator that makes use of renewable energy. Thermoelectric power generation is a key technology for achieving a low-carbon society. However, the use of thermoelectric power generation is not widespread because it is less efficient compare to those of other generators such as solar cells. Therefore, breakthrough to drastically enhance thermoelectric efficiency are necessary.

Thermoelectric efficiency increases monotonously with the dimensionless figure of merit $ZT$, where $Z$ is propor-
tional to the electrical conductivity and the square of the Seebeck coefficient, and inversely proportional to the thermal conductivity, and $T$ is the absolute temperature. The introduction of nanoscale structures into thermoelectric materials is expected to lead to a breakthrough in enhancing the thermoelectric figure of merit [1-4]. A number of researchers are engaged in characterizing nanoscale thermoelectric materials [5-8]. However, owing to the size scale of nanostructured materials, it is very difficult to measure their thermoelectric characteristics.

We have investigated the measurement of the Seebeck coefficient on a nanometer scale by a new technique using Kelvin-probe force microscopy (KFM), which allows a non-contact measurement. It has been demonstrated that the Seebeck coefficient for an n-type Si wafer measured by KFM has a value similar to that obtained by a conventional method, indicating that the KFM technique is a powerful tool for evaluating the Seebeck coefficient of thermoelectric nanostructures [9-12].

On the other hand, it is more difficult to measure the thermal conductivity since the evaluation of heat or temperature is not so easy even for bulk materials. In this study, with the aim of measuring the thermal conductivity on a nanometer scale, as a first step, we construct a non-contact measurement system based on an ac calorimetric method. The non-contact method using halogen-lamp heating and infrared (IR) irradiative thermometer are applied to measure the thermal diffusivity of bulk Si and Cu samples, and the usefulness is successfully demonstrated.

2. Methods

Principle of ac calorimetry. The so-called ac calorimetry technique can measure thermal diffusivity parallel to the broad surface in a thin material [13,14]. In the measurement system, a part of the thin material is shadowed by a mask on the surface, as shown in Fig. 1. By periodic light irradiation, ac thermal energy of $Q_0 e^{i\omega t}$ is supplied over all the sample surface and mask. The origin of the one-dimensional axis at the border of the shadow is set. When ac thermal energy is applied, ac heat propagates in the shadowed region and ac temperature is measured at a distance of $L$ using a thermocouple. The ac temperature has a delay time with respect to the ac thermal energy. Then, the mask position is shifted by $\delta L$ and the delay time $\delta t$ is measured again. Finally, from the relationship between the deviation $\delta L$ and delay time $\delta t$, we can obtain the information of thermal diffusivity $a$.

For evaluating the thermal diffusivity from the $\delta t$-$\delta L$ relation, we find a formula among $\delta t$, $\delta L$ and $a$.

The simplest equation of heat conduction, in which any heat transfer between sample and atmosphere is not taken into account, is expressed by

$$\frac{\partial T(x,t)}{\partial t} = a \frac{\partial^2 T(x,t)}{\partial x^2}$$

Under the condition that one edge of the sample is periodically heated, temperature at the edge is given by

$$T(x=0,t) = T_0 e^{i\omega t}$$

Using this boundary condition, the equation can be analytically solved to

$$T(x,t) = T_0 e^{-kx} e^{i(\omega t-kx)}$$

where

$$k = \sqrt{\frac{\omega}{2a}}$$

According to the solution, we obtain the thermal diffusivity $a$ from the relation

$$\frac{\delta L}{\delta t} = \sqrt{2a \omega}$$

In this paper, we use this equation for determining the thermal diffusivity from the observed relationship between distance deviation and delay time for bulk samples.

Experimental procedure. Figure 2 shows a schematic diagram of the experimental setup. In the present study, we used a halogen lamp (Tokina KTX-100) for periodic heating. An IR irradiative thermometer (CHINO IR-
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Figure 2. Schematic Diagram of the Apparatus used to Measure the Thermal Diffusivity for Bulk Materials without Contact to the Sample

BAT2A) was used for temperature detecting, whose monitored area was about 5 mm in diameter. The halogen lamp was alternately powered by a function generator (Tektronix AFG3022B) and the IR thermometer was movable along the sample. The signals of the function generator and the temperature were monitored by a logger (HIOKI 8430).

Bulk n-type Si with a P concentration of $1 \times 10^{16} \text{ cm}^{-3}$ and Cu were cut to a rectangle 10 mm in width and 50 mm in length. They were cleaned by ethanol before being set to a sample holder. The measurement was performed in an atmospheric pressure at room temperature.

The observed temperature was fitted by a sine curve. The delay time was determined from time shift of the fitted sine curve with respect to the sine signal applied to the halogen lamp.

3. Results and Discussion

The relationship between the distance deviation and the delay time for a Cu plate is shown in Fig. 5, which was measured with a heating frequency of 0.01 Hz. From this graph, the thermal diffusivity of Cu is evaluated to be 1.05 cm$^2$/s, which is close to the reported value of 1.09 cm$^2$/s [17]. The thermal conductivity is obtained at 361 W/mK by using specific heat and density of bulk Cu [17]. Consequently, the non-contact ac calorimetric method used in this study can be applied to characterize the thermal diffusivity of bulk samples.

However from Figs. 3 and 5, the plotted data are slightly scattered, which leads to a lack of precision in the thermal diffusivity evaluation. In order to make more precise, the measurement system will be in a shield box and the logger will be changed into a higher specification for measuring the voltage.

Furthermore, in applying to nanomaterials, the areas of heating and temperature detecting will have to be much smaller on a nanometer scale. In addition, the specific heat of the nanomaterials is essential for evaluating the thermal conductivity from the thermal diffusivity.

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The obtained value of thermal diffusivity for the Si is plotted in Fig. 4 together with reported values for n- and p-type Si [15], as a function of impurity concentration. The reported values are on a straight line, independent of the type of semiconductor. The thermal diffusivity value obtained by our non-contact measurement system also seems to be on the reported line. This result indicates that our measurement system is useful for characterization of thermal diffusivity. Using specific heat and density of bulk Si [16], moreover, thermal conductivity is evaluated to be 136 W/mK.

Figure 3. Relationship between Distance Deviation and Delay Time for an n-type Si Wafer with a P Concentration of $1 \times 10^{16} \text{ cm}^{-3}$. Measurement Frequency is $f=0.05 \text{ Hz}$

Figure 4. Thermal Diffusivity for Si Wafer as a Function of Impurity Concentration. Reported Values are also Shown and the Broken Line is an Eye Guide
4. Conclusions

In order to construct a novel thermal conductivity measurement technique applicable to nanometer-scaled materials, in this study, we set a non-contact system based on an ac calorimetric method and measured the thermal diffusivity of bulk Si and Cu. Although there is a slight lack of precision, the measured values of thermal diffusivity were close to the reported values. Consequently, our non-contact technique is useful for bulk materials and can be a basis of a system that can characterize thermal conductivity for nanomaterials.

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References