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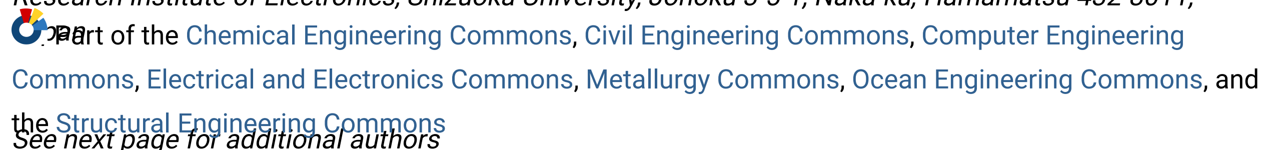
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Construction of a Novel Method of Measuring Thermal Conductivity for Nanostructures

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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.

Abstrak

Penyusunan Metode Baru untuk Mengukur Konduktivitas Termal untuk Strukturnano. Dengan tujuan untuk menggambarkan proses konduksi termal pada bahan-bahan berskala nanometer, kami telah menyusun sebuah metode baru yang didasarkan pada metode kalorimetrik ac. Di dalam metode ini, pemanasan sampel secara periodik dilaksanakan dengan iradiasi sinar, dan suhu periodik yang sejajar dideteksi dengan termometer iradiatif infra merah. Dengan demikian, kami mengukur difusivitas termal tanpa melibatkan kontak dengan sampel objektif. Di dalam kajian ini, kami memutuskan untuk mengukur difusivitas termal dari limbak Si dan Cu menggunakan metode tanpa kontak ini dengan iradiasi lampu halogen. Untuk menentukan difusivitas termal dari hubungan antara deviasi jarak dan waktu tunda, kami menggunakan persamaan gelombang yang paling sederhana, dan angka-angka difusivitas termal dari Si dan Cu yang diperoleh ternyata hampir sama dengan angka-angka yang telah dilaporkan. Oleh karena itu, metode tanpa kontak ini berguna untuk mengevaluasi konduksi termal dan dapat diterapkan untuk bahan-bahan berskala nanometer dengan memperbaiki pemanasan lokal dan sistem-sistem pendeteksian lokal.

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 δL and the delay time δt is measured again. Finally, from the relationship between the deviation δL and delay time δt, we can obtain the information of thermal diffusivity a.

For evaluating the thermal diffusivity from the δt-δL relation, we find a formula among δt, δL and a.

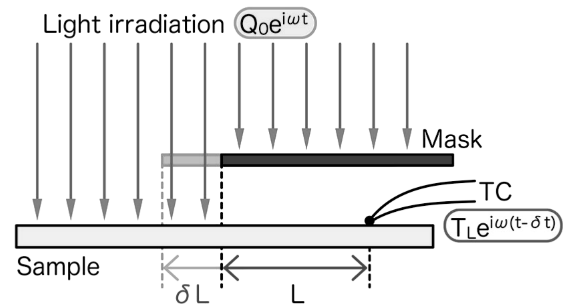

Figure 1. Cross-sectional View of an ac Thermal Diffusivity Measuring System, in which ac Thermal Energy is Supplied Uniformly on the Upper Surfaces of a Sample and a Mask by Light Irradiation. The ac Temperature is Measured by a Thermocouple

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-

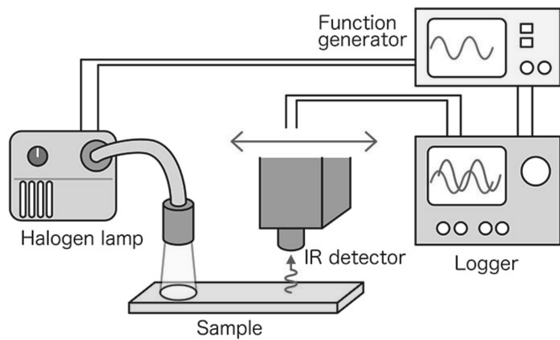

Figure 2. Schematic Diagram of the Apparatus used to Measure the Thermal Diffusivity for Bulk Materials without Contact to the Sample

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 $1x10^{16}$ 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 delay time δt of measured temperature with respect to the periodic heating for the n-type Si is shown in Fig. 3, as a function of distance deviation δL. The measurement frequency is 0.05 Hz. It is found that the plot shows a linear relation, and therefore, the thermal diffusivity is evaluated to be 0.83 cm^2/s from the gradient of the graph.

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.

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.

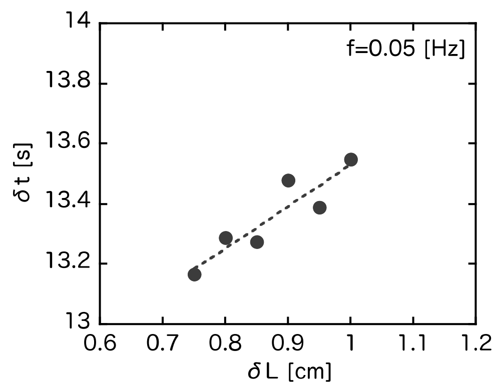

Figure 3. Relationship between Distance Deviation and Delay Time for an n-type Si Wafer with a P Concentration of $1x10^{16}$ cm^{-3}. Measurement Frequency is f=0.05 Hz

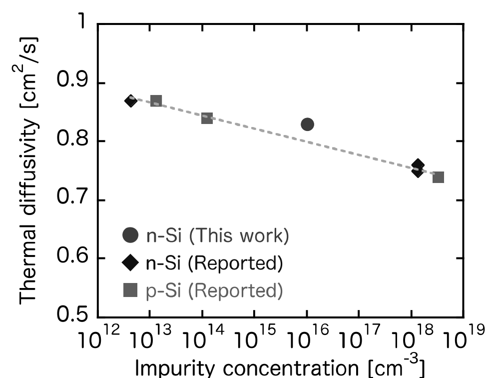

Figure 4. Thermal Diffusivity for Si Wafer as a Function of Impurity Concentration. Reported Values are also Shown and the Broken Line is an Eye Guide

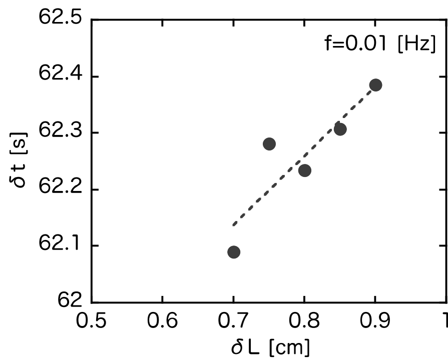

Figure 5. Relationship between Distance Deviation and Delay Time for a Cu Plate. Measurement Frequency is *f*=0.01 Hz

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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