Development of Seebeck-Coefficient Measurement Systems Using Kelvin-Probe Force Microscopy

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Abstract

Thermoelectric device is investigated by a number of researchers in order to enhance the thermoelectric efficiency. It is known that the efficiency can be improved by quantum effect. However, it is difficult to measure the thermoelectric characteristics of nanometer-scale structures. Thus a new measurement method is expected to be developed. We propose to apply Kelvin-probe force microscopy (KFM) to characterization of thermoelectric materials. KFM can locally observe surface potential of Fermi energy of a sample without touching the sample surface. In the present paper, we estimate the Seebeck coefficient of thin Si-on-insulator layers using KFM.

Abstrak

Pengembangan Sistem Pengukuran Koefisien Seebeck dengan Mikroskopi Gaya Pengamatan Kelvin. Sejumlah peneliti mengkaji alat termoelektrik untuk meningkatkan efisiensi termoelektrik. Telah diketahui bahwa efisiensi dapat ditingkatkan dengan efek kuantum. Namun, pengukuran ciri-ciri termoelektrik sulit dilaksanakan pada struktur-struktur berskala nanometer. Oleh karena itu, sebuah metode pengukuran baru harus dikembangkan. Kami menyarankan penggunaan mikroskopi gaya pengamatan Kelvin (*Kelvin-probe force microscopy* atau KFM) di dalam proses karakterisasi materi-materi termoelektrik. KFM dapat secara lokal mengamati potensi permukaan energi Fermi dari sebuah sampel tanpa menyentuh permukaan sampel itu. Di dalam makalah ini, kami memperkirakan koefisien Seeback dari lapisan-lapisan insulator Si-on menggunakan KFM.

Keywords: Fermi energy, Kelvin-probe force microscopy, Seebeck coefficient

1. Introduction

Recently, intensive research has been conducted on the development of power generators that make use of renewable energy. Thermoelectric power generation is a key technology for achieving the low-carbon society. However, the use of thermoelectric power generation has not been widespread because its efficiency is insufficient compared with other generators such as solar cell. Therefore, breakthroughs to drastically enhance the thermoelectric efficiency are necessary.

The thermoelectric efficiency increases monotonically with the dimensionless figure-of-merit ZT, where Z is the figure-of-merit and T is the absolute temperature. The figure-of-merit is defined as follows:

$$Z = \frac{S^2 \sigma}{\kappa}, \tag{1}$$

where S is the Seebeck coefficient, σ is the electric conductivity, and κ is the thermal conductivity. Improvement in efficiency may be achieved by introducing nanometer-scale structures, such as nanowires and superlattices [1-4]. A number of researchers are currently engaged in experimentally characterizing nanometer-scale thermoelectric materials [5-8]. However, it is difficult to perform such measurements because of the very small dimensions. In order to characterize nanostructures, electrodes and heaters for measurement must be fabricated on the sample by microelectromechanical systems (MEMS). In addition, external disturbances, such as lead-wire contacts, affect the accuracy of the evaluation. On the other hand, a few methods using probe microscopy techniques have been developed to measure the characteristics of nanostructured materials [9-10]. In these techniques, a probe touches the sample surface, and thermoelectric characterization is performed in the

vertical direction (i.e., normal to the sample surface). Therefore, contact between the metallic probe and the sample surface is not avoidable and specially-customized equipment is required.

In order to measure the Seebeck coefficient on a nanometer scale, we propose a new technique using Kelvin-probe force microscopy (KFM), which allows non-contact measurement. Another advantage of our technique is that, if the sample holder is only adjusted, commercial KFM equipment can be used. A full detail of the technique is given in the next section. In this paper, we estimate the Seebeck coefficient of thin Si-on-insulator (SOI) layer by our new technique.

Kelvin-probe force microscopy. Kelvin-probe force microscopy is a powerful tool for measuring the electric potential of nanometer-scale structures. A number of researchers have demonstrated local potential characterization for a variety of materials by KFM [11-13]. Fig. 1 describes the measurement principle of the Fermi energy of a sample by KFM. When a cantilever is in close proximity to the sample surface and is electrically connected to the sample, the Fermi energy of the cantilever coincides with that of the sample. Then, a Coulomb force will occur between the cantilever and the sample due to the work-function difference $q\Delta\Phi$, as shown in Fig. 1(a). The Coulomb force is expressed as follows:

$$F = -\frac{1}{2}V^2\frac{\partial C}{\partial z},\qquad(2)$$

where V is the potential difference between the cantilever and the sample (or $\Delta\Phi$), C is the cantilever-sample capacitor, and z is the cantilever-sample distance.

Note that KFM nullifies this force by applying an external bias between the cantilever and the sample (V_{DC} in Fig. 1(b)). The magnitude of this bias corresponds to the potential difference. Using the above procedure, the Fermi energy of the sample with respect to that of the cantilever can be obtained from the applied bias.
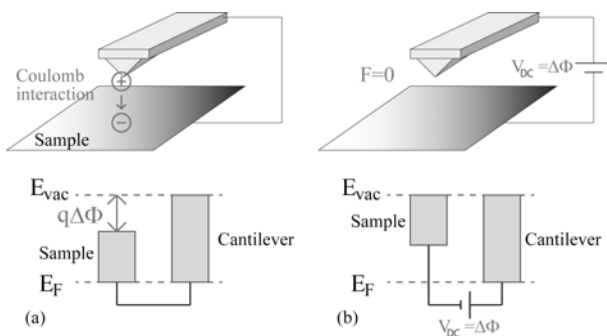

Fig. 1. Band Structure of the Cantilever and the Sample at (a) $V_{DC} = 0$ and (b) $V_{DC} = \Delta\Phi$

Band structure. The Seebeck coefficient S is defined as follows:

$$S = -\frac{\Delta V}{\Delta T},\qquad(3)$$

where ΔV is the thermoelectromotive force, and ΔT is the temperature difference. Hence, S can be obtained by measuring the thermoelectromotive force for a given temperature difference across the sample. Equation (3) is converted to where T_H and T_L are the temperatures in the high- and low-temperature regions, respectively, E_{FTH}, E_{FTL}, and E_{FAu} are the Fermi energies of each of these regions and of the cantilever, respectively. Φ_{TH} and Φ_{TL} are the surface potentials in the high- and low-temperature region, respectively. Therefore, S can be estimated from the measurement of the Fermi energy for

$$S = -\frac{1}{q}\frac{E_{FTH} - E_{FTL}}{T_H - T_L}$$

$$= -\frac{1}{q}\frac{(E_{FTH} - E_{Au}) - (E_{FTL} - E_{Au})}{T_H - T_L}$$

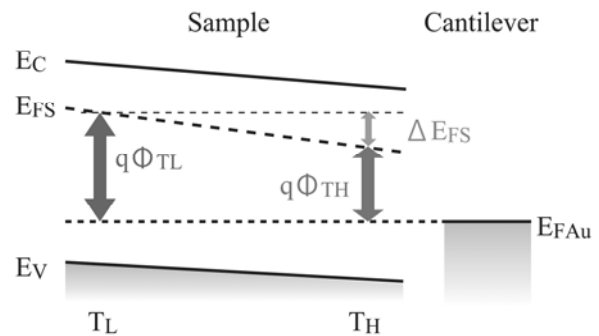$$= \frac{\Phi_{TH} - \Phi_{TL}}{T_H - T_L},\qquad(4)$$

Fig. 2. Fermi Energy Positions when a Temperature Difference is Applied to the Sample
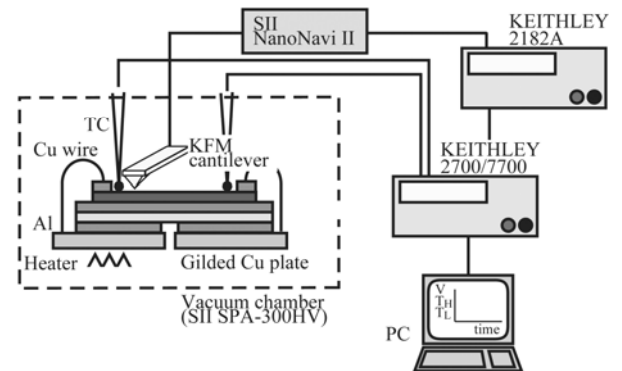

Fig. 3. Diagram of the Apparatus Used to Measure S by KFM

each region. Fig. 2 shows the band structure of the cantilever and the n-type Si sample under a temperature difference. When a temperature difference is applied to the sample, the Fermi energy of the sample will be spatially distributed. Generally, E_{FTH} is lower than E_{FTL} in n-type Si. Since KFM can measure Φ_{TH} and Φ_{TL} as shown in Fig. 2, we can evaluate the Seebeck coefficient.

2. Experiments

Experimental equipment. Fig. 3 shows a schematic diagram of the experimental setup. In this study, we used the new KFM control unit (Seiko Instruments Inc., NanoNavi II) and the measurement devices. The surface potential of the sample was monitored using a nanovoltmeter (KEITHLEY, 2182A), and the temperatures of the sample were measured using a digital multimeter (KEITHLEY, 2700). A sample was bridged over two gilded Cu plates separated by a gap of 3 mm, and Cu wires were connected between the plates and Al electrodes on the SOI layer. A joule heater was attached to one of the Cu plates. The temperature difference was applied to the sample by flowing a current to the heater. Two T-type thermocouples were directly attached to both ends of the sample surface in order to measure the temperature. At the high- and low-temperature regions of the sample surface, the time evolutions of temperature and surface potential were measured simultaneously. Measurements were performed in a vacuum chamber (Seiko Instruments Inc., SPA-300HV) at a pressure of 3.2×10^{-5} Pa.

Sample preparation. In this study, we measured a SOI wafer. The SOI layer was 85nm in thickness and was doped with a phosphorus concentration of 1.1×10^{17} cm^{-3}. The SOI layer was patterned, on which Al electrodes were deposited, as shown in Fig. 4. Another Al electrode was deposited onto the bottom of the Si sample. Finally, the SOI sample was annealed in ambient N$_2$.
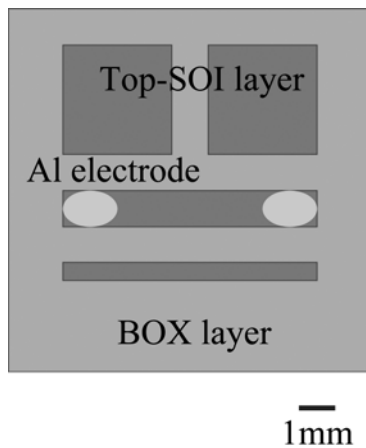

Fig. 4. **Schematic Diagram of Patterned SOI Wafer**

3. Results and Discussion

Fig. 5 shows the time evolution of the temperature on the SOI surface and the temperature difference $\Delta T = T_H - T_L$. The temperature difference was found to increase with increasing sample temperature. In this case, the temperature difference was more than 3 K.

Fig. 6 shows the time evolution of the surface potential in the low-temperature region obtained simultaneously with the temperatures shown in Fig. 5. The surface potential was found to decrease with increasing time (or ΔT). There are some disturbances in the surface-potential data, as pointed by arrows in this figure. At these points, we had to adjust a distance between the sample and the cantilever because the thermal expansion of the sample caused on overflow in the z-gain, that is, an excess of a limitation in the piezoelectric system. Therefore, an analysis was performed after removing with the exception of the data during the adjustment.
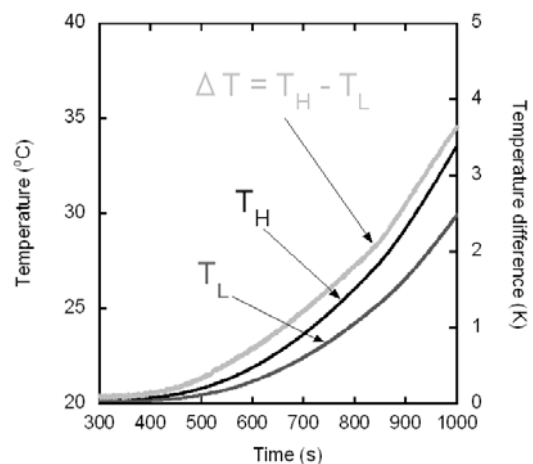

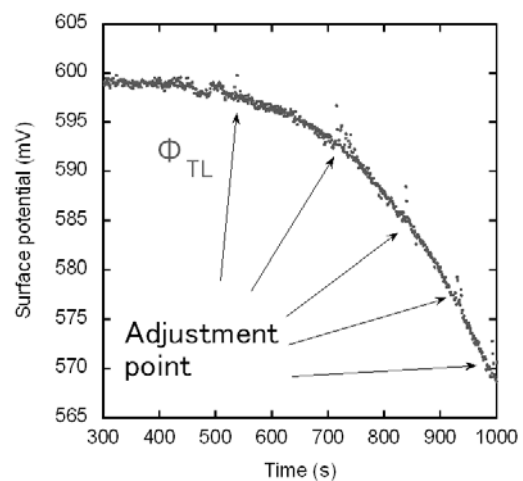Fig. 5. **Time Evolution of Temperatures and Temperature Difference**

Fig. 6. **Time Evolution of Surface Potential at Low-temperature Region**

Fig. 7 is the relationship between surface potential shift and temperature difference obtained from Figs. 5 and 6. The thermoelectromotive force is given by $\Phi_{TH} - \Phi_{TL}$ at the same temperature difference. According to (3), the Seebeck coefficient is estimated from fitted lines in Fig. 7. The result of the estimated Seebeck coefficient is drawn in Fig. 8, as a function of average temperature. The circle and square data are the value estimated using KFM, and the diamond data are obtained by a conventional method. The Seebeck coefficient is -2.8 mV/K for the KFM technique, which is about 2.5 times as large as the values for the conventional method, although the reproducibility in the KFM estimation
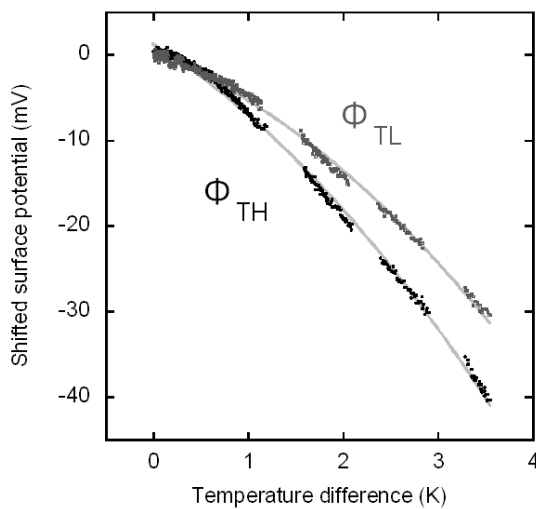
seems to be good. Unfortunately, the reason of the disagreement is not clear yet. Thus, we are now making sure of the certainty and the validity of estimated Seebeck coefficient.

4. Conclusions

We proposed a new technique for estimating the Seebeck coefficient on a nanometer scale using KFM. The Seebeck coefficient of a thin SOI film by this technique was estimated to be -2.8 mV/K, which was observed reproducibly. There was disagreement between the Seebeck coefficients obtained by a KFM technique and a conventional method. Therefore, the certainty and the validity of the KFM estimation is now under investigation.

Acknowledgment

References

[1] L.D. Hicks, M.S. Dresselhaus, Phys. Rev. B. 47 (1993) 12727.

[2] L.D. Hicks, M.S. Dresselhaus, Phys. Rev. B. 47 (1993) 16631.

[3] A.A. Balandin, O.L. Lazarenkova, Appl. Phys. Lett. 82 (2003) 415.

[4] M.V. Simkin, G.D. Mahan, Phys. Rev. Lett. 84 (2000) 927.

[5] T.C. Harman, P.J. Taylor, M.P. Walsh, B.E. LaForge, Science. 297 (2002) 2934.

[6] D. Li, Y. Wu, P. Kim, I. Shi, P. Yang, A. Majumdar, Appl. Phys. Lett. 83 (2003) 2934.

[7] A.I. Hochbaum, R. Chen, R.D. Delgado, W. Liang, E.C. Garnett, M. Nafarian, A. Majumdar, P. Yang, Nature. 451 (2008) 163.

[8] A.I. Boukai, Y. Bunimovich, J.T. Kheli, J.K. Yu, W.A. Goddard. III, J.R. Heath, Nature. 451 (2008) 168.

[9] M. Koyano, N. Akashi, J. Elecron. Mater. 38 (2009) 1037.

[10] Y. Zhang, C.L. Hapenciuc, E.E. Castillo, T. Borca-Tasciuc, R.J. Mehta, C. Karthik, G. Ramanath, Appl. Phys. Lett. 96 (2010). 062107-1-3.

[11] Y. Leng, C.C. Williams, L.C. Su, G.B. Stringfellow, Appl. Phys. Lett. 66 (1995) 1264.

[12] A. Kikukawa, S. Hosaka, R. Imura, Appl. Phys. Lett. 66 (1995) 3510.

[13] A. Chavez-Pirson, O. Vatel, M. Tanimoto, H. Ando, H. Iwamura, H. Kanbe, Apple. Phys. Lett. 67 (1995) 3069.

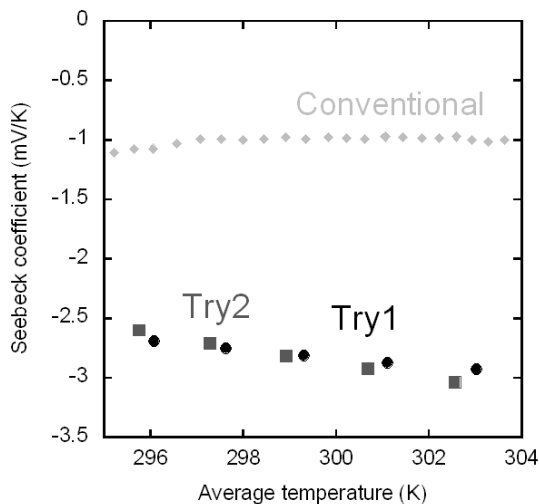**Fig. 7. Relationship between Surface Potential Shift and Temperature Difference**

Fig. 8. Seebeck Coefficient as a Function of Average Temperature. The Measurement by KFM System was Performed Two Times (Try1 and 2). Seebeck Coefficient Obtained by a Conventional Method is also Shown