P24

Thermal Conductivity Measurements of Ultrananocrystalline Diamond/ Hydrogenated Amorphous Carbon Composite Films by Time-Domain Thermoreflectance

Satoshi Takeichi,[1] Takashi Nishiyama,[2] Mitsuru Tabara[3], Shuichi Kawawaki,[3] Masamichi Kohno,[3] Koji Takahashi[2] and Tsuyoshi yoshitake[1]

[1] *Department of Applied Science for Electronics and Materials, Kyushu University, 6-1 Kasuga, Fukuoka 816-8580, Japan*

[2] *Department of Aeronautics and Astronautics, Kyushu University, 744 Motooka, Nishi-ku, Fukuoka 819-0395, Japan*

Kasuga-koen, Kasuga, Fukuoka 816-8580, Japan,

[3] *Department of Mechanical Engineering, Kyushu University, 744 Motooka, Nishi-ku, Fukuoka 819-0395, Japan*

Abstract

The thermal conductivity of ultrananocrystalline diamond/hydrogenated amorphous carbon composite films prepared by coaxial arc plasma deposition (CAPD) was measured by a time-domain thermoreflactance method. The specific heat and density of the films were measured to be 0.891 J/g·K and 1.70 g/cm^3, respectively, as parameters for the estimation of the thermal conductivity. Based on them, the thermal conductivity was estimated to be 2.32 W/m·K. This value is smaller than that of general UNCD/a-C:H films prepared by chemical vapor deposition (CVD). It might be because the CAPD films possesses a huge number of grain boundaries owing to extremely small grains of approximately 2.3 nm as compared with those of CVD films and its structure enhances phonon scattering.

1. Introduction

Ultrananocrystalline diamond (UNCD)/hydrogenated amorphous carbon (a-C:H) composite (UNCD/a-C:H) is a new attractive candidate for thermoelectric devices, because of nanostructured materials comprising carbon that is abundant and harmless for human being, easy processing by lithography, and light.

The thermoelectric properties of nanodiamond films have been studied mainly for films prepared by CVD[1]. However, ZT of B-doped nanodiamond films prepared by CVD is extremely small, because the grain size are approximately 10 nm, which is insufficient for a reduction in the thermal conductivity owing to phonon scattering in grain boundaries[2].

Recently we have realized the formation of UNCD/a-C:H films by coaxial arc plasma deposition (CAPD). UNCD/a-C:H films prepared by CAPD comprises a large number of diamond grains with diameters of approximately 2.3 nm and an a-C:H matrix, and the grain size is smaller than those of UNCD/a-C:H films prepared by CVD [3]. It is expected that its structure is feasible for an enhancement in the Seebeck coefficient and a reduction in the thermal conductivity. The thermal conductivity of UNCD/a-C:H films has rarely been studied experimentally thus far, and there have been no studies on the thermal conductivity of UNCD/a-C:H films prepared by CAPD, to our knowledge. In this study, the thermal con-

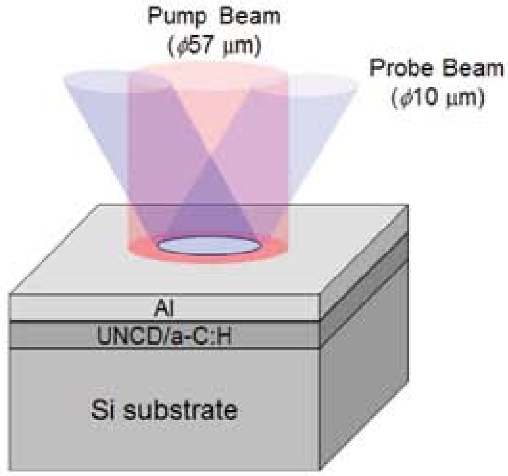

Figure 1. Schematic diagram of time-domain thermoreflectance method.

ductivity
of UNCD/a-C:H films prepared by CAPD were estimated by a time-domain thermoreflectance (TDTR) method, on the basis of the measurements of parameters requited for the thermal conductivity estimation.

2. Chapter Title

UNCD/a-C:H films were deposited on Si substrates at a substrate temperature of 550 °C and a hydrogen pressure of 53.3Pa by CAPD using a coaxial arc plasma gun (Ulvac ARL-300) with a graphite cathode target. The arc plasma gun equipped with a 720 μF capacitance was operated at an applied voltage of 100 V. The repetition rate of arc discharges was 5 Hz. Al films were deposited on the UNCD/a-C:H films by sputtering for the TDTR measurement.

Figure. 1 shows a schematic diagram of TDTR method. The thermal conductivities of the UNCD/a-C:H films were determined by fitting the measured $V_{in}(t)/V_{out}(t)$. Here, $V_{in}(t)$ and $V_{out}(t)$ are the voltages converted from the intensity of incident light and reflected light of a probe laser, respectively. The heat flow was approximated to be one-dimensional since the radius of the focused laser beam on the sample was always much larger than the film thickness.

3. Results and discussion

Figure 2 shows a TDTR spectrum measured for a sample indicated in Fig. 1. The thermal conductivities of the film was estimated to be 2.32 W/m·K from a fitting curve for the $V_{in}(t)/V_{out}(t)$ plot with a thermal model. Here, the time range for the fitting was determined to be 2-50 ps from a sensitivity analysis. The thermal conductivity value, 2.32 W/m·K, is an order of magnitude smaller than that reported for UNCD/a-C:H films prepared by CVD thus far. It might be because of the existence of a huge number of grain boundaries caused by extremely small grain size in the films. As another reason, it is supposed that UNCD/a-C:H films prepared by CAPD contain more amount of a-C:H in the films, as compared with those prepared by CVD.

4. Conclusion

The thermal conductivity of UNCD/a-C:H films deposited on Si substrates by CAPD was measured by TDTR method, and it was estimated to be 2.32 W/m·K. The value is approximately an order of magnitude smaller than that of UNCD/a-C:H films prepared by CVD. It might be because the grain size is smaller than that of films prepared by CVD. A huge number of grain boundaries caused by the existence extremely small grain with diameters of approximately 2.3 nm enhances phonon scattering, which results in a decrease in the thermal conductivity. It was experimentally demonstrated that UNCD/a-C:H films pre-pared by CAPD possess the low thermal conductivity, which is beneficial for their application as thermoelectric materials.

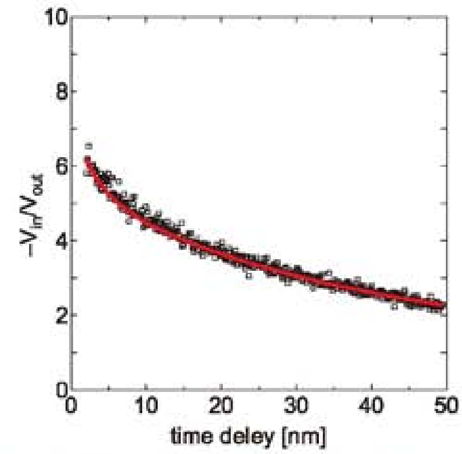

Figure 2. Plot of ratio between incident and reflected light intensities in voltage, measured at room temperature by TDTR, as a function of time. Simulation fitting curve is indicated in red.

Acknowledgment

This work was partially supported by the Advanced Low Carbon Technology Research and Development Program (ALCA) of the Japan Science and Technology (JST), the Yoshida Science and Education Promotion Association, and JSPS KAKENHI Grant Number 15H04127. The first author, S. T. is grateful for financial assistance provided by the Kyushu University Advanced Graduated Program in Global Strategy for Green Asia.

Reference

1) A. Balducci, M. Marinelli, M. E. Morgada, G. Pucella, G. Rodriguez, M. Scoccia, and G. Verona-Rinati, Microsyst. Technol., **12**, 365 (2006).
2) M. Engenhorst, J. Fecher, C. Notthoff, G. Schierning, R. Schmechel, and S. M. Rosiwal, Carbon N. Y., 81, 650 (2015).
3) T. Yoshitake, Y. Nakagawa, A. Nagano, R. Ohtani, H. Setoyama, E. Kobayashi, K. Sumitani, Y. Agawa, and K. Nagayama, Jpn. J. Appl. Phys., 49, Part 1 (2010).

Email: satoshi_takeichi@kyudai.jp