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Interfacial effect on thermal conductivity of diamond-like carbon films [†]

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Abstract

Diamond-like carbon (DLC) has been of interest as a promising coating for protection and insulating layer in micro-electromechanical systems due to high hardness, wear resistance, transparency in IR range, chemical inertness and biocompatibility. The interfacial effect on thermal transport is studied for DLC films deposited on Al_2O_3 substrates with an ion gun method. Thermal conductivity of DLC thin films is measured with a 3ω method. DLC films show the thickness-dependent thermal conductivity, which is understood with the interfacial thermal resistance between DLC thin film and Al_2O_3 substrate. The interfacial thermal resistance and thermal conductivity of bulk DLC are determined with the measured thickness-dependent thermal conductivity of DLC films.

Keywords: Diamond-like Carbon; Hydrogenated amorphous carbon; Thermal conductivity; 3 omega method

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1. Introduction

Diamond-like carbon (DLC) is an amorphous carbon with sp^3 bonding of carbons. DLC containing hydrogen is called hydrogenated amorphous carbon (a-C:H) [1]. DLC has similar properties to diamond such as high hardness, high wear resistance, high transparency in the infrared range, chemical inertness, and biocompatibility. Due to these properties, the interest in DLC has grown for various applications in modern industries such as protective coatings, microelectronic systems, optical coatings and biomedical applications [1-3].

Since devices have become miniaturized rapidly, the interfacial effect becomes more important in determining the physical properties of devices. There have been reports on the reduction in thermal conductivity of dielectric materials consisting of nanosized grains and films [4-6]. Yang *et al.* [4] reported the strong grain size dependence of the thermal conductivity of nanocrystalline yttria-stabilized zirconia (YSZ) and the interfacial thermal resistance of grain boundaries at temperatures between 6 K and 480 K. The film thickness-dependent thermal conductivity of Y_2O_3 thin films deposited on an Al_2O_3 substrate and the interfacial thermal resistance between Y_2O_3 and Al_2O_3 have also been reported [5].

Thermal conductivities of DLC thin films were reported by Shamsa et al. [7]. They studied various DLC film with such as hydrogenated amorphous carbons (a-C:H) and tetrahedral amorphous carbon (ta-C) films. This work reports the interfacial effect on the thermal conductivity of a-C:H thin films deposited on Al_2O_3 substrates. a-C:H thin films with thicknesses of 200 nm, 600 nm, 1200 nm, and 1800 nm are prepared with an ion gun method. The thermal conductivity of the a-C:H films is measured with the 3ω method [6].

2. Experimental details

a-C:H films are deposited on an Al_2O_3 substrate using a linear ion beam source system (J&L Tech Co., LTD.). Fig. 1 shows schematically a linear ion beam source system that consists of gas feed manifolds, steel-case grounded cathodes, anodes, and magnets. C_2H_2 gas is fed through the cavity between the cathodes and anodes. Plasma is produced by ionizing the feed gas with electrical energy. The ions are bombarded in the form of an ion beam, directed toward a substrate, and form in carbon films on the substrate. Table 1 shows the deposition conditions for the a-C:H thin films. The thickness of films is controlled by varying the deposition time, and 200 nm, 600 nm, 1200 nm, and 1800 nm thick a-C:H films are prepared for this study. The structure of the a-C:H films is analyzed by X-ray diffraction (XRD: Rigaku GDX-11P3A). The XRD patterns of the a-C:H films show no peaks except for the peaks of the Al_2O_3 substrate as expected because carbons form in the amorphous a-C:H films.

The thermal conductivity of the a-C:H thin films is measured with the 3ω method which needs a metal pattern of four

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Table 1. Deposition conditions for the growth of a-C:H films.

Substrate	Al_2O_3
Gas	C_2H_2 (35 sccm)
Substrate-source distance	10 cm
Initial pressure	2.6×10^{-5} torr
Process pressure	0.8×10^{-3} torr
Source power	1100 V/0.2 A
Bias power	100 V/0.2 A/350 kHz/1.3 μs
Deposition rate	12.5 nm/min

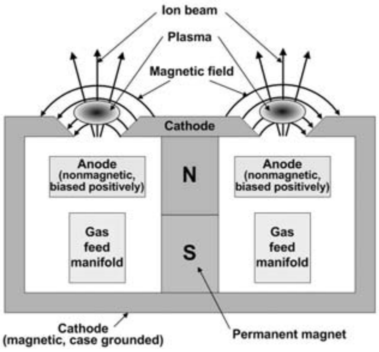

Fig. 1. Schematic diagram of a linear ion beam source system used for the deposition of hydrogenated amorphous carbon films. This system was developed by J&L Tech co., LTD.

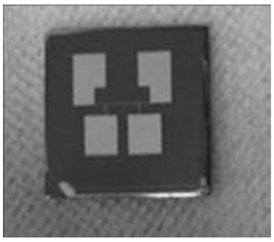

Fig. 2. An image of surface view of a metal pattern fabricated on a-C:H film.

probes connected with a line on the top of the film. Gold and chromium are deposited by electron beam evaporation on the sample for the metal pattern. Chromium is an intermediate layer for improving the adhesion of gold to the a-C:H thin film. The pattern shown in Fig. 2 is obtained by following the procedures of photolithography and etching, where the width and length of metal line are 30 μm and 3.074 mm, respectively.

The metal line behaves as both heater and thermometer. By driving an AC current of frequency ω to the metal line, joule heating of frequency 2ω is generated. Since the resistance of the metal line is proportional to the temperature, the voltage drop across the metal line has a 3ω component. Therefore, the frequency-dependent temperature oscillation of the metal line can be obtained from the 3ω component in the voltage. A lock-in amplifier (SR850, Stanford Research System) separates the 3ω term from other ω terms with a cancellation circuit shown in Fig. 3. The measurement of the thermal conductivity is performed at room temperature, and the measurement of each thin film is repeated three times in order to minimize the experimental error.

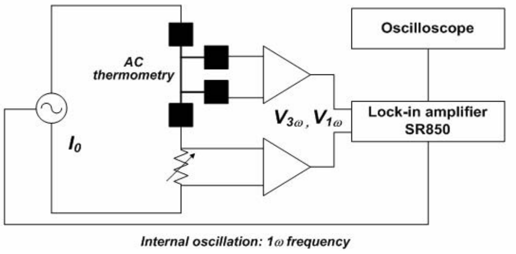

Fig. 3. Schematic diagram of cancellation circuit for 3ω measurement.

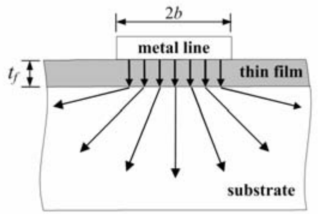

Fig. 4. Schematic diagram of heat flow through thin film and substrate in 3ω measurement.

3. Results and discussion

The thermal conductivity of the a-C:H films is measured with the differential 3ω method [6]. If the film thickness is smaller than the thermal penetration depth of the film, the heat flow in the film can be given by a one-dimensional Fourier's equation. Fig. 4 shows schematically the heat flow through a thin film and substrate using the 3ω method, where a thin film shows one-dimensional heat flow while the substrate is characterized by radial heat flow.

Since the thickness of a substrate is much larger than the thermal penetration depth of the substrate, the temperature fluctuation of a substrate depends on the frequency. The temperature oscillation of a metal line on a thin film, ΔT, is represented by the summation of temperature oscillations of the substrate, ΔT_s and film, ΔT_f [6]:

$$\begin{aligned}\Delta T(\omega) &= \Delta T_s(\omega) + \Delta T_f \\ &= \frac{P}{l\pi k_s}\left(\frac{1}{2}\ln\frac{D_s}{b^2} + \eta - \frac{1}{2}\ln(2\omega) - \frac{i\pi}{4}\right) + \frac{Pt_f}{2lbk_f}\end{aligned} \quad (1)$$

where D_s is the thermal diffusivity of the substrate, ω is the frequency of the driven current, P is the power, l and b the length and half-width of the heater, respectively, t_f the film thickness, and k_s and k_f are thermal conductivity of the substrate and the film, respectively. We can obtain temperature oscillations of the metal line from measurements of the 3ω term and can also calculate the temperature oscillations of the substrate. From the difference between temperature oscillations of the metal line and the substrate, the thermal conductivity of the thin film can be obtained as follows:

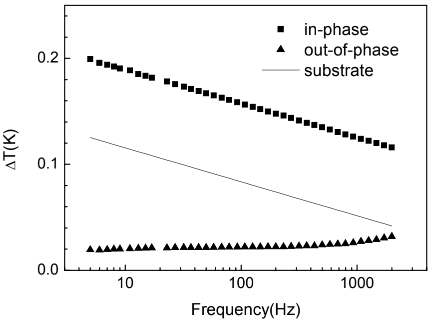

Fig. 5. Amplitude of the temperature oscillations of a heater on a 1200 nm thick a-C:H film as a function of the frequency of driven current.

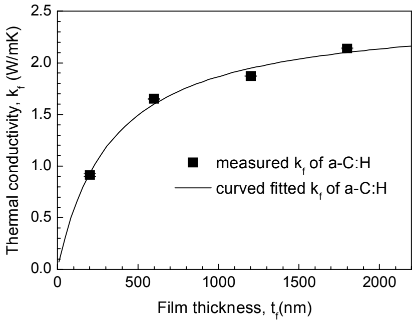

Fig. 6. Thermal conductivity of a-C:H films on Al_2O_3 substrate as a function of film thickness denoted by closed squares (■).Results are fitted by Eq. (4) and represented by a solid line.

$$k_f = \frac{Pt_f}{2lb\Delta T_f} \tag{2}$$

Fig. 5 shows the measured amplitude of the temperature oscillations of the metal line on a 1200 nm thick a-C:H film on the Al_2O_3 substrate. Temperature oscillations of the metal line and substrate are denoted with closed squares and solid line, respectively. The figure shows that differences between the two temperature oscillations are independent of frequency of the driven current. The thermal conductivity of the 1200 nm thick DLC film at room temperature, 1.8715 W/m/K, is calculated from Eq. (2). Thermal conductivities of a-C:H films of 200 nm, 600 nm, 1200 nm, and 1800 nm are obtained following the same procedure and shown in Fig. 6.

A a-C:H thin film does not show a dramatic change in thermal conductivity between 600 nm and 1800 nm, but the thermal conductivity decreases dramatically when the thickness is less than 500 nm. The thermal conductivity of 200 nm a-C:H films is almost half that of 600 nm a-C:H films. The reduction in thermal conductivity is directly related to the interfacial resistance between the thin film and substrate because the obtained thermal conductivity is the overall value of the thin film. Since all samples used in this study are prepared at deposition conditions, we assume that all samples have the same interface with one another and, therefore, the interfacial resistance is constant for all films. The overall thermal resistance of the thin film, the inverse of the thermal conductance, is represented as the thermal resistance of the interior part of the film and that of the interface as follows:

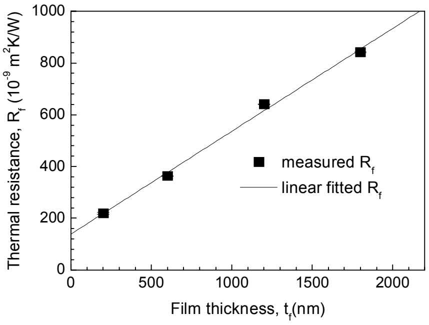

Fig. 7. Thermal resistance of a-C:H films on Al_2O_3 substrate as a function of film thickness denoted by closed squares (■).Results are fitted by Eq. (3) and represented by a solid line.

$$\frac{t_f}{k_f} = \frac{t_f}{k_i} + R_k \tag{3}$$

where k_i is the thermal conductivity of bulk, t_f is the film thickness, and R_k is the interfacial thermal resistance.

Fig. 7 shows the overall thermal resistance of the films. The closed squares and solid line represent the experimental data and a fit to the data with Eq. (3), respectively. The intercept on the y-axis gives the interfacial resistance, 13.443×10^{-8} m^2K/W. Eq. (4) can be represented for the overall thermal conductivity, k_f, as follows [4, 5, 8]:

$$k_f = \frac{k_i}{1 + k_i R_k / t_f} \tag{4}$$

The solid line in Fig. 6 is a fit to the experimental values with Eq. (4), which gives the interfacial thermal resistance between the a-C:H film and Al_2O_3 substrate, 13.443×10^{-8} m^2K/W, and the thermal conductivity of bulk, 2.491 W/m/K. Eq. (4) illustrates that the overall thermal conductivity, k_f, of the thin film is less than the thermal conductivity of bulk, k_i, as the interfacial effect term, k_iR_k/t_f, becomes significant, but k_f becomes close to k_i as the film thickness increases. In a-C:H thin films, the interfacial effect is significant for a thickness less than 500 nm.

4. Conclusions

a-C:H films are deposited on Al_2O_3 substrates using an ion gun method. The thin films of 200 nm, 600 nm, 1200 nm, and 1800 nm thicknesses are prepared by controlling the deposition time. The differential 3ω method is used to measure the thermal conductivity of the DLC thin films. The thermal conductivity of the a-C:H films shows a film thickness dependence, which is directly related to the interfacial thermal resistance. The interfacial thermal resistance between a a-C:H film and Al_2O_3 substrate and the thermal conductivity of bulk a-

C:H are determined as 13.443×10^{-8} m^2K/W and 2.491 W/m/K, respectively, along with the measured thickness-dependent thermal conductivity of a-C:H films.

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Nomenclature

b : Half-width of heater
D_s : Thermal diffusivity of substrate
k_f : Thermal conductivity of film
k_i : Intrinsic(or bulk) thermal conductivity
k_s : Thermal conductivity of substrate
l : Length of heater
P : Power of heater
t_f : Thickness of film
R_k : Interfacial thermal resistance or Kapitza resistance
ω : Frequency of driven current of heater

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