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Thermal Diffusivity Measurement of Diamond Films[1]

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This paper discusses the short-pulse-flash method developed for thermal diffusivity measurements on thin films. Two kinds of CVD diamond film have been prepared, and their thermal diffusivity in the perpendicular direction has been measured with this method. The measurement errors caused by the surface coating are discussed.

KEY WORDS: diamond; film; flash method; thermal diffusivity.

1. INTRODUCTION

The thermal transport properties (thermal diffusivity and thermal conductivity) of diamond films synthesized by CVD have become increasingly important because of their wide range of applications. One of the important applications of synthetic diamond films is as high-thermal-conductivity substrates in microelectronics.

The measurement of the thermal diffusivity of films is classified into the following two main cases: measurement of the thermal diffusivity parallel to the surface of a thin film, $\alpha_{||}$ and measurement of the thermal diffusivity perpendicular to the surface of a thin film, $\alpha_{\perp}$. For a diamond film, the thermal diffusivity varies with the synthetic process, and the thermal diffusivity in the perpendicular direction $\alpha_{\perp}$ differs from that in the plane direction $\alpha_{||}$ because of its anisotropy. Diamond single crystals have

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the highest thermal conductivity ($2.2 \times 10^3\ \mathrm{W \cdot m^{-1} \cdot K^{-1}}$ for natural crystals), and are also expected to have a high thermal diffusivity as synthetic diamond films. Thus, the measurement of the thermal diffusivity of a diamond film in the perpendicular direction is very difficult due to the fast thermal transport.

The flash method was initially developed by Parker et al. [1] in 1963. The basic formula for thermal diffusivity measurement by the flash method is

$$\alpha = 1.37L^2/(\pi^2 t_{1/2}) \tag{1}$$

where L is the thickness of the specimen and $t_{1/2}$ is the time from the initiation of laser pulse to that when the rear surface temperature rises to half of its maximum value.

The flash method has been widely used for thermal diffusivity measurements and has been extended to the measurement of thermal diffusivity of multilayer materials [2, 3], composite materials [4], porous materials [5], and functionally gradient materials [6].

Recently, this method was applied to the $\alpha_\perp$ measurement of thin films [7–9], although it has not been widely used for this purpose. In this paper, the thermal diffusivity of diamond films in the perpendicular direction is measured using this method.

2. EXPERIMENT

A schematic of the experimental apparatus used here is shown in Fig. 1. The transient temperature rise was detected by a high-speed infrared radiation pyrometer. For this pyrometer, the photoelectric infrared receiving and converting device ((Hg, Cd)Te (PV Model)) has a response time of 0.01 μs and an operating temperature of 77 K. The output electric signal feeds to a low-noise preamplifier with an amplification factor of 1500 and a frequency band of 200 Hz to 5 MHz. The output signals from the pyrometer are received and recorded by a DAS 820M high-speed data acquisition system with a maximum write speed of 0.05 μs/point. A 0.1 μs CO_2 laser pulse was used as a heating source with a wavelength of 10.6 μm and a pulse power of $1 \times 10^4\ \mathrm{J \cdot m^{-2}}$. A cylindrical mirror and Ge lens are used to make the light spot cover the specimen uniformly.

Two kinds of CVD synthetic diamond film samples were prepared at the Hebei Academy of Sciences, China. Each sample has a diameter of 12 mm and a thickness of 340 μm. Sample 1 is a diamond with main crystal direction (111) in the normal direction of the sample plane and has a small crystal grain. Sample 2 is a diamond with main crystal direction (110) in

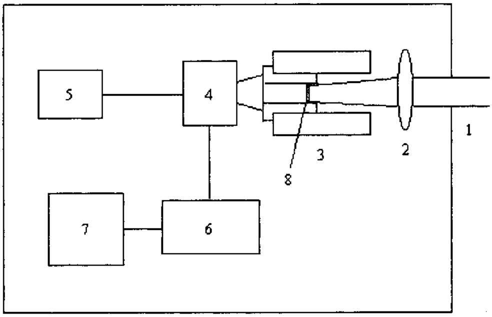

Fig. 1. Schematic of measurement apparatus. (1) CO_2 pulse laser, (2) Ge lens, (3) sample holder, (4) high-speed infrared radiation pyrometer, (5) power source of the pyrometer, (6) DAS-820M high-speed data acquisition system, (7) oscilloscope, (8) sample.

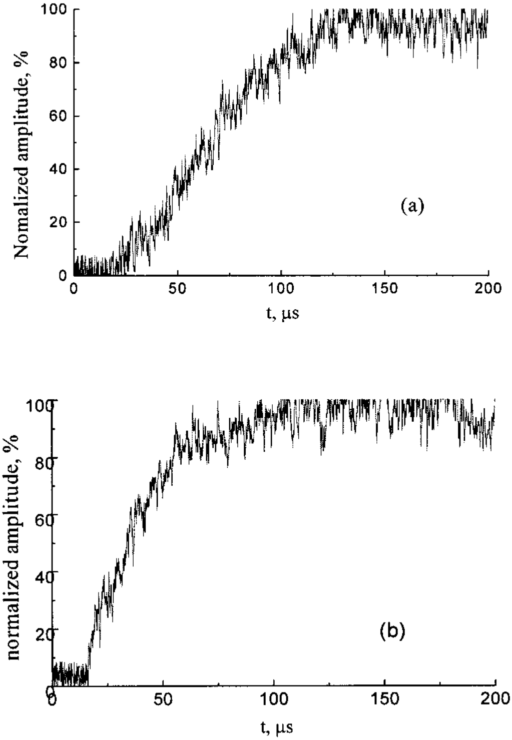

Fig. 2. Rear-surface temperature-rise history of samples. (a) 1 and (b) 2.

the normal direction of the sample plane and has a large crystal grain. A titanium film with a thickness of less than 1 μm was doped in both sample surfaces. In order to increase the radiation absorption of the specimen and its emission to the pyrometer, a colloidal graphite coating layer was applied to both films.

Figure 2 shows the temperature-rise curve at the rear surface of the samples under transient laser heating. The characteristic time $t_{1/2}$ can be determined using the temperature-rise curve.

3. ERROR ANALYSIS, CORRECTION, AND RESULTS

According to Parker's mathematical analysis, some basic assumptions need to be satisfied when one uses the method: (1) uniform distribution of radiation energy on the surface layer, (2) no heat loss, (3) homogeneous material, (4) small depth absorption of the energy pulse, and (5) no finite-pulse-time effect. For the measurement of $\alpha_\perp$ of thin films by the flash method, the measurement errors caused by the pulse-time effect, the infinite absorbed thickness, and the surface coating will be more significant than those for thick samples.

According to Tang et al. [9, 10], for a 0.1 μs CO_2 laser pulse, when the thickness of the sample is larger than 10 μm, the deviation in the measurement caused by infinite absorbed thickness is less than 1%, and the deviation caused by the pulse-time effect is within 1%. Thus, the measurement errors caused by these two factors can be neglected in this experiment.

For the surface coating, because the thickness of the coating layer is comparable with the thickness of the thin-film sample, the deviation should be very large and must be corrected. We treat the film samples with coating on both the front and rear surfaces as a three-layer complex sample (Fig. 3). For simplification, we assume $F(t)$ is a δ function, so we can take

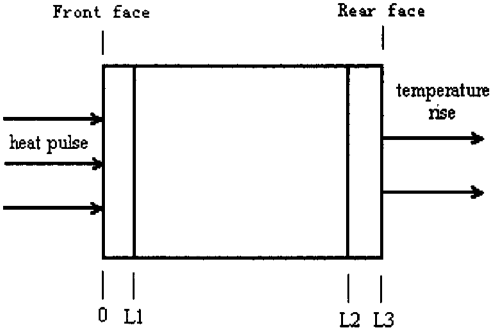

Fig. 3. Schematic of complex three-layer sample.

the rear-surface temperature-rise history of the complex three-layer sample as [11]:

$$V(L_3, t) = 1 + \frac{2}{\rho_1 C_1 L_1 + \rho_2 C_2 (L_2 - L_1) + \rho_3 C_3 (L_3 - L_2)} \times \sum_{k=1}^{\infty} \frac{\exp(-\alpha_3 \beta_k^2 t/(L_3 - L_2)^2)}{h(\beta_k)} \tag{2}$$

In Eq. (2), β_k was calculated from

$$\begin{aligned} &\sqrt{\alpha_1}\, \rho_1 C_1 \sin(\beta_k x_1) \cos(\beta_k x_2) \cos \beta_k \\ &\quad + \sqrt{\alpha_2}\, \rho_2 C_2 \cos(\beta_k x_1) \sin(\beta_k x_2) \cos \beta_k \\ &\quad + \sqrt{\alpha_3}\, \rho_3 C_3 \cos(\beta_k x_1) \cos(\beta_k x_2) \sin \beta_k \\ &\quad - \sqrt{\frac{\alpha_1 \alpha_3}{\alpha_2}} \frac{\rho_1 \rho_3 C_1 C_3}{\rho_2 C_2} \sin(\beta_k x_1) \sin(\beta_k x_2) \sin \beta_k = 0 \end{aligned} \tag{3}$$

$$\begin{aligned} h(\beta_k) = {} & k_1 \cos(\beta_k x_1) \cos(\beta_k x_2) \cos \beta_k - k_2 \cos(\beta_k x_1) \sin(\beta_k x_2) \sin \beta_k \\ & - k_3 \sin(\beta_k x_1) \cos(\beta_k x_2) \sin \beta_k - k_4 \sin(\beta_k x_1) \sin(\beta_k x_2) \cos \beta_k \end{aligned} \tag{4}$$

where

$$\begin{aligned} k_1 &= \rho_1 C_1 L_1 + \rho_2 C_2 (L_2 - L_1) + \rho_3 C_3 (L_3 - L_2) \\ k_2 &= \frac{1}{\sqrt{\alpha_2 \alpha_3}} (\alpha_3 \rho_1 C_1 \rho_3 C_3 L_1/\rho_2 C_2 + \alpha_2 \rho_2 C_2 (L_2 - L_1) + \alpha_3 \rho_3 C_3 (L_3 - L_2)) \\ k_3 &= \frac{1}{\sqrt{\alpha_1 \alpha_3}} (\alpha_1 \rho_1 C_1 \alpha_3 \rho_3 C_3 (L_3 - L_1)/\alpha_2 \rho_2 C_2 + \alpha_1 \rho_1 C_1 (L_3 - L_2) \\ &\quad + \alpha_3 \rho_3 C_3 L_1) \\ k_4 &= \frac{1}{\sqrt{\alpha_1 \alpha_2}} (\alpha_1 \rho_1 C_1 \rho_3 C_3 (L_3 - L_2)/\rho_2 C_2 + \alpha_2 \rho_2 C_2 (L_2 - L_1) + \alpha_1 \rho_1 C_1 L_1) \\ x_1 &= \frac{L_1 \sqrt{\alpha_3}}{(L_3 - L_2) \sqrt{\alpha_1}} \\ x_2 &= \frac{(L_2 - L_1) \sqrt{\alpha_3}}{(L_3 - L_2) \sqrt{\alpha_2}} \end{aligned} \tag{5}$$

α_j is the thermal diffusivity, ρ_j is the density, and C_j is the specific heat of the jth layer.

Using Eqs. (2)–(5), we can calculate and correct the error caused by the colloidal graphite coating. To do this, the density ρ, the specific heat C_p, the thermal diffusivity α, and the thickness L of the coating layer must be known. For this calculation, we take the specific heat C_p of the coating as $1.36 \times 10^3\ \mathrm{J \cdot kg^{-1} \cdot K^{-1}}$, the density ρ as $8 \times 10^2\ \mathrm{kg \cdot m^{-3}}$, and the thermal diffusivity α as $5 \times 10^{-6}\ \mathrm{m^2 \cdot s^{-1}}$ from TPRC [12].

Because it is very difficult to measure accurately the thickness of the coating layer by a direct method, an indirect method was used. We used an Al film with a thickness of 160 μm as a reference sample, coated it by the same method as used for the diamond film, and measured the $\alpha_{||}$ of the reference Al film by ac calorimetry. Because the Al film is homogeneous in both directions, the $\alpha_{\perp}$ should be the same as $\alpha_{||}$. With a knowledge of α of the reference Al film and the measured rear-face temperature history, the thickness of the surface coating was calculated. Then, using Eq. (2), the experimental data were processed and the deviation caused by the coating layer corrected, as shown in Table I.

In addition to the deviation caused by the colloidal graphite coating, there were also errors involved in the calculated correction caused by the errors in C_p, ρ, and α of the coating layer and the nonuniformity of the coating layer.

The errors caused by the errors in C_p, ρ, and α of the coating layer are different for each sample. For sample 1, the error is within 2%, but for sample 2, the error is 5%. The error caused by the nonuniformity of the coating layer is much larger; for a 5% deviation of thickness, the error will be 5% to 7%. Also, errors involved in the measurement of the characteristic time $t_{1/2}$ resulted in a large error in the thermal diffusivity. In this experiment the error is 5% for sample 1 and 15% for sample 2. Thus, the total error (or uncertainty) for the measurement of thermal diffusivity is within 15% for sample 1 and 30% for sample 2.

According to our results for diamond films, it can be seen that for the two samples prepared by different synthetic processes, the thermal diffusivity α differs significantly; the α of sample 2 is much larger than that of sample 1.

Table I. Thermal Diffusivity of Diamond Film Samples

Sample	1	2
$t_{1/2}$ (μs)	61.5 ± 2.5	37.1 ± 2.0
α ($10^{-4}\ \mathrm{m^2 \cdot s^{-1}}$)	4.02 ± 0.24	8.91 ± 0.80

4. CONCLUDING REMARKS

In the present study, we have developed a high-speed heating system and transient radiation temperature measuring system. Two kinds of CVD diamond film were prepared, and the thermal diffusivity of each was measured by the flash method. The measurement error was analyzed, and the error caused by the surface coating has been calculated and corrected. From our measured result, it can be seen that for the samples prepared by difference synthetic processes, the thermal diffusivity α differs considerably.

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