Obliquely Crossed Photothermal Deflection for Thermal Conductivity Measurements of Thin Films

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Abstract

A three-dimensional theoretical model has been developed to calculate the normal probe beam deflection of obliquely crossed photothermal deflection configuration in samples, which is consisted of thin films deposited on substrates. Utilizing the dependence of the normal component of probe beam deflection on the cross-point position of the excitation and probe beams, the thermal conductivity of the thin film can be extracted from the ratio of the two maximums of normal deflection amplitude, which occur when the cross-point is located near both surfaces of the sample. The effects of other parameters, including the intersect angle between the excitation and probe beams in the sample, the modulation frequency of the excitation beam, the optical absorption and thickness of the thin films, the thermal properties of substrates, on the measurement of the thin film thermal conductivity are discussed. The obliquely crossed photothermal deflection technique seems to be well suited for thermal conductivity measurements of thin films with high thermal conductivity but low optical absorption, such as diamond or diamond-like carbon, deposited on substrates with relatively low thermal conductivity.

Key words: photothermal deflection, thermal conductivity, thin films, three-dimensional theoretical model

1 . Introduction

Many experimental methods have been developed in recent years to measure the thermal properties of thin films[1,2,3]. Among these methods thermal-wave techniques, including photoacoustic (PA)[4,5], photothermal (PT)[6,7], surface displacement[8,9], thermo-reflectance (TR)[10,11], surface thermal grating[12,13], optical interferometry[14], radiometry[15], and/or photopyroelectric (PPE)[16] detections appear to be well suitable for characterizing thin film materials in situ. The thermal characterization (thermal conductivity or diffusivity measurements) of thin films is usually achieved by monitoring the time [17] (in pulsed excitation case) or frequency [7] (in modulated cw excitation case) dependences of the photothermal signal. The modulated photothermal techniques used in thermal conductivity or diffusivity measurements of thin films usually employ the frequency dependence of the amplitude, or more often the phase, of the periodically changed photothermal signal induced by a modulated cw excitation.

Recently, a differential configuration based on the conventional obliquely crossed PTD geometry is developed to separate the thin film and substrate absorptions[18]. This obliquely crossed PTD configuration can be used to measure the thermal conductivity or thermal diffusivity of a thin film deposited on a weakly absorbing substrate. In the case of the sample consists of thin film and weakly-absorbing substrate, the optical, thermal properties and the geometric parameters of the thin film will influence the temperature gradient within the substrate along mainly the vertical direction, which results in changes in the dependence of the normal component of the probe beam deflection on the cross-point position of the two beams. If the optical and geometric parameters of the thin film are previously known, the thermal conductivity or thermal diffusivity of the thin film can be

extracted from the dependence of the deflection signal on the position of the cross-point of the excitation and probe beams. This obliquely crossed PTD scheme can be used for thermal characterization of thin film with high conductivity at relatively low modulation frequency.

In this paper, we describe a theoretical model for thermal conductivity measurements of thin films by using obliquely crossed PTD configuration.

2. Theoretical Treatment

2.1. Temperature Distribution

Consider the geometry shown in Fig. 1, in which the system consists of four regions. The temperature distributions within the four regions given by solving differential thermal conduction equations with standard Hankel transform techniques[19] are shown as following

$$T_0(r,z,t) = \frac{1}{2}\int_0^\infty \delta d\delta J_0(\delta r) A(\delta) \exp(\beta_0 z) \exp(i\omega t) + c.c. \tag{1}$$

$$T_1(r,z,t) = \frac{1}{2}\int_0^\infty \delta d\delta J_0(\delta r)\left[E(\delta)\exp(-\alpha_1 z) + C_1(\delta)\exp(-\beta_1 z) + C_2(\delta)\exp(\beta_1 z)\right] \times \exp(i\omega t) + c.c. \tag{2}$$

$$T_2(r,z,t) = \frac{1}{2}\int_0^\infty \delta d\delta J_0(\delta r)\left\{F(\delta)\exp\left[-\alpha_2(z-L_1)\right] + D_1(\delta)\exp\left[-\beta_2(z-L_1)\right] + D_2(\delta)\exp\left[\beta_2(z-L_2)\right]\right\}\exp(i\omega t) + c.c. \tag{3}$$

$$T_3(r,z,t) = \frac{1}{2}\int_0^\infty \delta d\delta J_0(\delta r) B(\delta)\exp\left[-\beta_0(z-L_1-L_2)\right]\exp(i\omega t) + c.c. \tag{4}$$

where $T_i(r,z,t)$ denotes the temperature rise within region i $(i=0,1,2,3)$. α_1, L_1 and α_2, L_2 are the optical absorption coefficients and thicknesses of the substrate and the thin film,

respectively, $\varpi = 2\pi f$, f is the modulation frequency of the excitation beam, $J_0(\delta r)$ is the zero-order Bessel function of the first kind. And,

$$\beta_i^2 = \delta^2 + i\omega / k_{thi} \qquad \textit{(subscript i=0, 1,2)} \tag{5}$$

$$E(\delta) = \frac{\alpha_1(1-R_1)P}{2\pi K_1} \cdot \frac{\exp\left(-\delta^2 a^2 / 4\right)}{\beta_1^2 - \alpha_1^2} \tag{6}$$

$$F(\delta) = \frac{\alpha_2(1-R_1)(1-R_2)P}{2\pi K_2} \cdot \frac{\exp\left(-\delta^2 a^2 / 4\right)}{\beta_2^2 - \alpha_2^2} \cdot \exp(-\alpha_1 L_1) \tag{7}$$

The coefficients A, B, C_1, C_2, H_2, D_1, and D_2 can be determined by boundary conditions of the temperature and heat flux continuities. K_i $(i=1, 2)$, k_{thi} $(k_{thi}=K_i/\rho_i c_i)$ are the thermal conductivities, thermal diffusivities, R_1 and R_2 denote the reflection coefficients at the region 0/1 and 1/2 interfaces, respectively. a is the (1/e) radius of the excitation beam with Gaussian spatial intensity profile, P is the power of the excitation beam illuminating the sample surface. We have assumed above that the sample extends infinitely in the radial direction and the thermal effusivities of the substrate and the thin film match perfectly. The thermal convection and radiation are neglected. The excitation beam waist does not change over the propagation path within the sample.

From the equations above we know that the thermal parameters and the optical absorption coefficient of the thin film influence the temperature distribution within the substrate. For thin films with low or without absorption, the influence of the thin film on the temperature distribution within the substrate is determined by the thermal properties of the substrate and thin film. The changes in temperature distribution caused by the thermal

diffusion can be detected by the sensitive photothermal deflection with obliquely crossed geometry, which in turn can be used to determine the thermal properties of the thin film.

2.2. Probe Beam Deflection

After taking the probe beam refraction at the interfaces of the sample into consideration, the normal and transverse components of probe beam deflection in air in the obliquely crossed PTD configuration are φ_n and φ_t, respectively[18]:

$$\varphi_n = \frac{\cos(\theta_1)}{\cos(\theta_0)}\frac{dn_1}{dT}\int_0^{L_1}\left[\frac{\partial T_1}{\partial x} + \frac{\partial T_1}{\partial z}\tan(\theta_1)\right]dz + \frac{\cos(\theta_2)}{\cos(\theta_0)}\frac{dn_2}{dT}\int_{L_1}^{L_2}\left[\frac{\partial T_2}{\partial x} + \frac{\partial T_2}{\partial z}\tan(\theta_2)\right]dz \tag{8}$$

$$\varphi_t = \frac{dn_1}{dT}\frac{1}{\cos(\theta_1)}\int_0^{L_1}\frac{\partial T_1}{\partial y}dz + \frac{dn_2}{dT}\frac{1}{\cos(\theta_2)}\int_{L_1}^{L_2}\frac{\partial T_2}{\partial y}dz \tag{9}$$

Where n_1, n_2 are the refractive indices of the substrate and thin film, $\frac{dn_1}{dT}, \frac{dn_2}{dT}$ are their temperature coefficients of refractive index, respectively, θ_0 is the incident angle of probe beam at the substrate surface, and θ_1, θ_2 are the refractive angles of the probe beam at the substrate and the thin film, respectively, which follow Snell law. The normal or transverse components of the probe beam deflection is the sum of that caused by the temperature gradients within the substrate and the thin film, and can be obtained by substituting the temperature distributions within the substrate and the thin film into equations (8) or (9). In the obliquely crossed PTD configuration used to measure the thermal conductivity of the thin film, we set the y-directional offset of the probe and excitation beams to be zero. Then the transverse component of the probe beam deflection equals zero, and only the normal

component of the probe beam deflection needed to be considered. For simplicity, the Cartesian coordinate is transferred into cylindrical system, that is $r = \sqrt{x^2 + y^2}$, we have

$$\varphi_n = -\frac{\cos(\theta_1)}{\cos(\theta)}\frac{1}{n_1}\frac{dn_1}{dT}\int_0^{L_1} dz \int_0^{\infty} \delta d\delta \Big\{\delta J_1(\delta x_1)\big[E(\delta)\exp(-\alpha_1 z) + C_1(\delta)\exp(-\beta_1 z)$$

$$+C_2(\delta)\exp(\beta_1 z)\big] + \tan(\theta_1)\cdot J_0(\delta x_1)\big[\alpha_1 E(\delta)\exp(-\alpha_1 z) + \beta_1 C_1(\delta)\exp(-\beta_1 z)$$

$$-\beta_1 C_2(\delta)\exp(\beta_1 z)\big]\Big\}\exp(i\omega t)$$

$$-\frac{1}{2}\frac{\cos(\theta_2)}{\cos(\theta)}\frac{dn_2}{dT}\int_{L_1}^{L_2} dz \int_0^{\infty} \delta d\delta \Big\{\delta J_1(\delta x_2)\big[F(\delta)\exp\big[-\alpha_2(z-L_1)\big] + D_1(\delta)\exp\big[-\beta_2(z-L_1)\big]$$

$$+D_2(\delta)\exp\big[\beta_2(z-L_1)\big]\big] + \tan(\theta_2)\cdot J_0(\delta x_2)\big[\alpha_2 F(\delta)\exp\big[-\alpha_2(z-L_1)\big]$$

$$+\beta_2 D_1(\delta)\exp\big[-\beta_2(z-L_1)\big] - \beta_2 D_2(\delta)\exp\big[\beta_2(z-L_1)\big]\big]\Big\}\exp(i\omega t) + \text{c.c.} \qquad (10)$$

Where $x_1 = (z_0 - z)\tan(\theta_1)$ and $x_2 = (z_0 - L_1)\tan(\theta_1) - (z - L_1)\tan(\theta_2)$, J_1 is the first-order Bessel function of the first kind.

3. Numerical Results and Discussions

Equation (10) is used to evaluate the feasibility of thermal conductivity measurements of thin films using the obliquely crossed PTD configuration. We restrict our calculation to the cases of non-absorbing or low absorbing dielectric thin films, such as optical coatings, diamond or diamond-like carbon thin films, deposited on glass or other low-absorbing material substrates. Since the temperature coefficient of the refractive index of the air is at least one order of magnitude smaller than that of the glass or other solid

samples, we neglect the contribution of the probe beam deflection in air to the total probe beam deflection in our calculations.

The deposition of thin films induces changes in the amplitude and phase of the normal deflection when the probe beam passes through the heating region of the sample near the thin film/substrate interface. Numerical calculations show that the probe beam deflection produced in the thin film is much smaller than that in the substrate, therefore the contribution of the film to the total probe beam deflection is negligible.

Fig.2 shows the dependence of the normal deflection amplitude on the cross point position for different thermal conductivity ratio of the thin film to substrate at incident angle of 80°. For comparison, the result for substrate without thin film is also shown in Fig.2 (curve 1). As seen from Fig.2, the thin film influences the normal deflection amplitude mainly near the surface of the substrate where the thin film is deposited on. At low thermal conductivity ratio, the thin film has negligible effect on the normal deflection amplitude even near the surface. As the thin film thermal conductivity increases, its effect on the deflection amplitude gradually becomes apparent and the difference between the two maximums of the deflection amplitude increases. The ratio of the two maximums of the deflection amplitude approaches saturation at high thermal conductivity of the thin film. That is, there exist a thermal conductivity ratio range in which the thermal property of the thin film has apparent effect on the ratio of the two maximums of deflection amplitudes.

Figs.3 and 4 show the effects of the thin film thickness and the modulation frequency of the excitation beam on the measurement. The amplitude ratio of two deflection maximums has been normalized. In general, as the thin film thickness and the modulation frequency increase, the measurable range of the thermal conductivity ratio can

be extended to much lower. Numerical calculations also show the change of thermal conductivity of the substrate has no apparent effect on the relationship between the maximum deflection amplitude ratio and the thermal conductivity ratio. Therefore the measured thermal conductivity of the films extends down to low thermal conductivity region as the thermal conductivity of the substrate decreases.

The above numerical evaluations show that for non-absorbing thin films deposited on absorbing substrates, the thermal conductivity of the thin film can be determined from the ratio of the deflection maximum, and the measurement range can be adjusted by changing the modulation frequency of the excitation beam or choosing different substrate materials. The obliquely crossed PTD configuration is well suitable for the measurement of thermal conductivity of thin films with high thermal conductivity and thin thickness at relatively low modulation frequency, which also be possible for the measurement of thermal conductivity of thin film with thin thickness and low thermal conductivity at high modulation frequency. The results also show that large difference between the thermal conductivities of the thin film and substrate is beneficial to the thermal conductivity measurement of the thin film with obliquely crossed PTD configuration.

The existence of thin film absorption considerably complicates the thermal conductivity measurement using obliquely crossed PTD configuration. The probe beam deflection within the film/substrate interfacial region of the substrate is the vectorial sum of the deflections caused by the substrate and thin film absorptions. As the amplitude and phase of the probe beam deflection caused by the thin film and substrate absorptions have different dependences of the cross-point position, the dependence of the total deflection on the cross-point position is very complicated, specially when the absorption of the thin film

and that of substrate are comparative. In this case the determination of the thermal conductivity of thin film by the ratio of deflection maximum is very difficult. If the difference between the probe beam deflections caused by the thin film and substrate is large, it is possible to determine the thermal conductivity of the thin film from the ratio of the deflection maximum. In the case of the absorption of thin film is much lower than that of the substrate, the effect of thin film absorption is negligible.

Since most dielectric films are optically transparent or weakly absorbing, the obliquely crossed PTD scheme seems to be a well suitable technique for thermal conductivity measurements of the dielectric thin films. The above numerical evaluation demonstrates that by employing the ratio of the maximum deflection , the obliquely crossed PTD configuration is capable to measure the thermal conductivity of films, with a wide range, and the measurement is independent on the substrate absorption. Numerical results further indicate that the obliquely crossed PTD configuration is especially suited for thermal conductivity measurements of thin films with high thermal conductivity, such as diamond or diamond-like films.

4. Conclusion

A three-dimensional theoretical model considering the sample including thin film and substrate has been developed to calculate the photothermal deflection with obliquely crossed configuration, which is applied to thermal conductivity measurements of thin films. By experimentally measuring and theoretically calculating the ratio of the two maximum deflection amplitudes, which occur near both surface regions at large incident(or refractive) angle , the thermal conductivity of a transparent or weakly absorbing thin film deposited on

an absorbing substrate can be determined. The results demonstrate the obliquely crossed photothermal deflection technique is well suited for measuring the thermal conductivity of film with high thermal conductivity and very thin thickness, which is difficult to measure by transverse PTD scheme (mirage effect) employing the frequency dependences of the amplitude or phase of the deflection signal.

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Captions of Figures

Fig.1 Beam geometry for theoretical model

Fig.2 Normal deflection amplitude as function of cross point position in the case of a non-absorbing thin film deposited on a glass substrate. The incident angle of the probe beam is 80°. Curve 1 is the result for free substrate case. Curves 2-4 are the results for thin film thickness of 1 μm, and K_2/K_1 of 20, 200, 2000, respectively.

Fig.3 Normalized maximum deflection ratio vs. thermal conductivity ratio. The modulation frequency is 100 Hz, the refractive angle is 80°, and the thin film thickness is (1) 0.1 μm, (2) 1 μm, (3) 10 μm, respectively.

Fig.4 Normalized maximum deflection ratio vs. thermal conductivity ratio. The thin film thickness is 1 μm, the refractive angle is 80°, and the modulation frequency is (1) 10 Hz, (2) 100 Hz, and (3) 1 KHz, respectively.

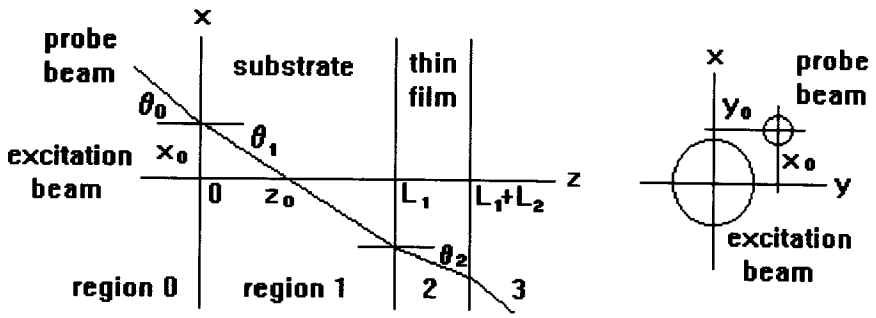

Figure 1

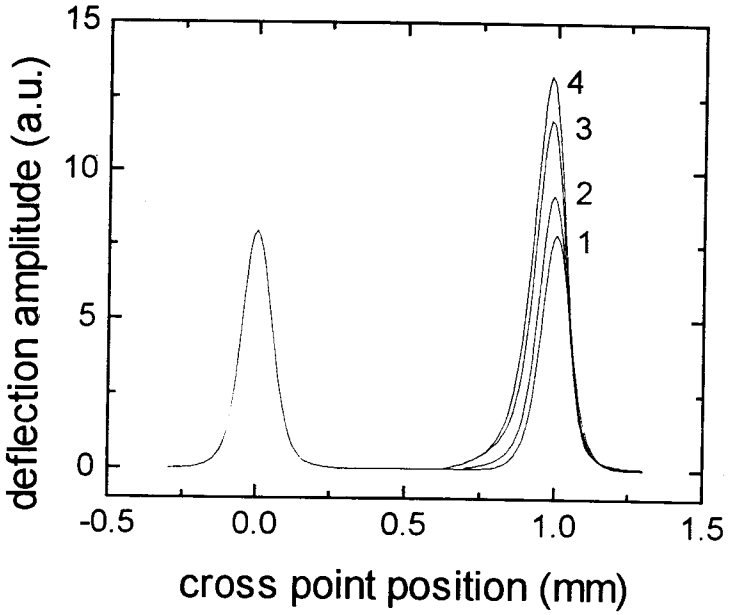

Figure 2

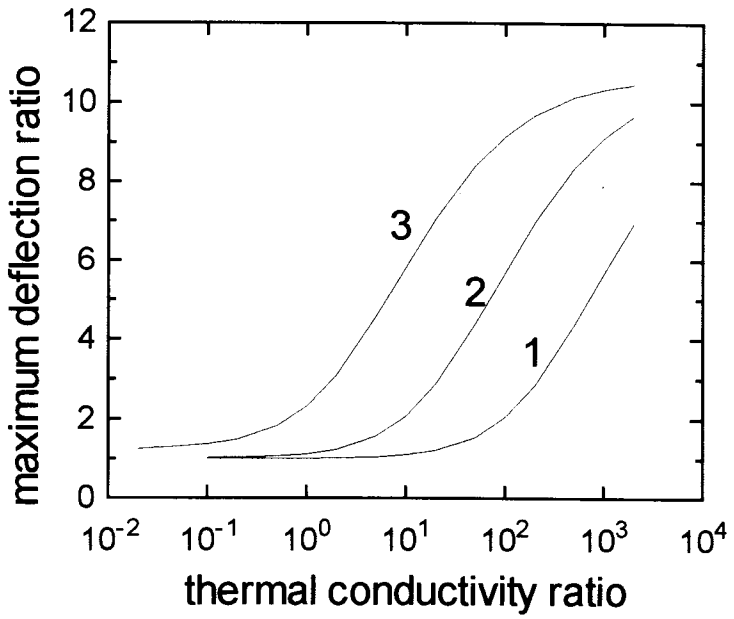

Figure 3

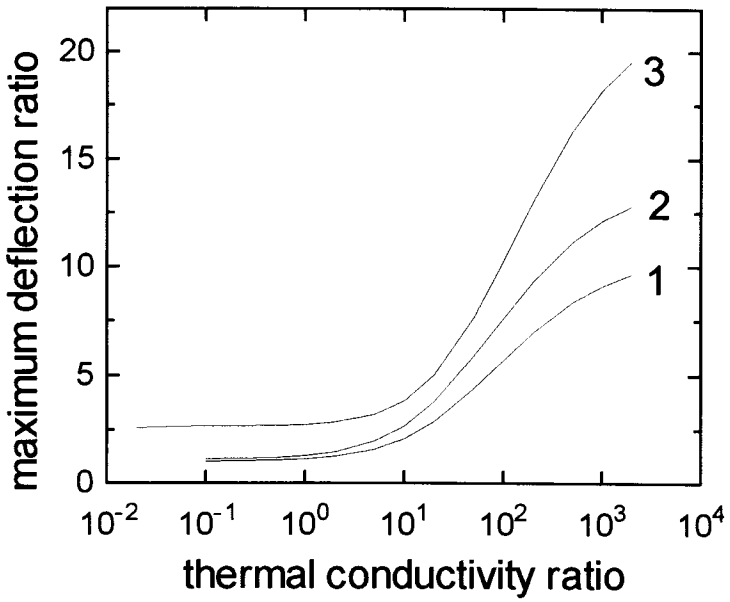

Figure 4