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# Theoretical Modeling of Obliquely Crossed Photothermal Deflection for Thermal Conductivity Measurements of Thin Films[1]

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A three-dimensional theoretical model has been developed to calculate the normal probe beam deflection of the obliquely crossed photothermal deflection configuration in samples which consist 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 maxima of the normal deflection amplitude, which occurs 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 the probe beams in the sample, the modulation frequency of the excitation beam, the optical absorption and thickness of the thin films, and the thermal properties of substrates on the thermal conductivity measurement of the thin film, are discussed. The obliquely crossed photothermal deflection technique seems to be well suited for thermal conductivity measurements of thin films with a high thermal conductivity but a low optical absorption, such as diamond and diamond-like carbon, deposited on substrates with a relatively low thermal conductivity.

**KEY WORDS:** photothermal deflection; thermal conductivity; thin film.

## 1. INTRODUCTION

Many experimental methods have been developed in recent years to measure the thermal properties of thin films [1–3]. Among these methods,

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thermal-wave techniques, including photoacoustic (PA) [4, 5], photothermal deflection (PTD) [6, 7], surface displacement [8, 9], thermoreflectance (TR) [10, 11], surface thermal grating [12, 13], interferometry [14], radiometry [15], and/or photopyroelectric (PPE) [16] detections, appear to be very 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 the pulsed excitation case) or frequency [7] (in the modulated cw excitation case) dependence 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 was developed to separate 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 where the sample consists of a thin film and a weakly absorbing substrate, the optical and 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 films with a high conductivity at a relatively low modulation frequency.

In this paper, we describe a theoretical model for thermal conductivity measurements of thin films using the 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 follows:

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

$$\begin{aligned} T_1(r, z, t) = \tfrac{1}{2} \int_0^\infty \delta d\, \delta J_0(\delta r)[E(\delta) \exp(-\alpha_1 z) + C_1(\delta) \exp(-\beta_1 z) \\ + C_2(\delta) \exp(\beta_1 z)] \exp(i\omega t) + \text{c.c.} \end{aligned} \tag{2}$$

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

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

where $T_i(r, z, t)$ denotes the temperature rise within region $i$ ($i = 0, 1, 2, 3$). $\alpha_1$, $L_1$ and $\alpha_2$, $L_2$ 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, and $J_0(\delta r)$ is the zero-order Bessel function of the first kind. And

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

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

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

The coefficients $A$, $B$, $C_1$, $C_2$, $H_2$, $D_1$, and $D_2$ can be determined by the boundary conditions of the temperature and heat flux continuities.

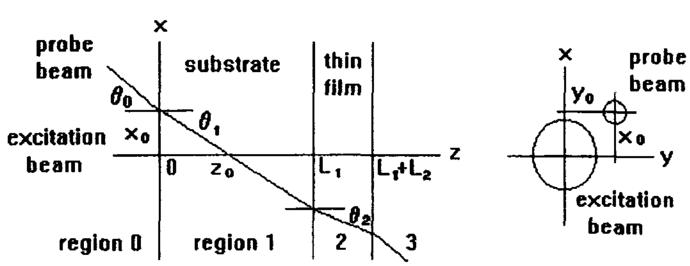


**Fig. 1.** Beam geometry for the theoretical model.

$K_i$ $(i=1,2)$ and $k_{\text{th}\,i}$ $(k_{\text{th}\,i}=K_i/\rho_i c_i)$ are the thermal conductivities and thermal diffusivities, and $R_1$ 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 a Gaussian spatial intensity profile, and $P$ 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 zero 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 $\varphi_{\text{n}}$ and $\varphi_{\text{t}}$, respectively [18]:

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

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

where $n_1$ and $n_2$ are the refractive indices of the substrate and thin film, $dn_1/dT$ and $dn_2/dT$ are the temperature coefficients of the refractive index, respectively, $\theta_0$ is the incident angle of the probe beam at the substrate surface, and $\theta_1$ and $\theta_2$ are the refractive angles of the probe beam at the substrate and the thin film, respectively, which follow Snell's law. The normal or transverse components of the probe beam deflection represent 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 Eq. (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 needs to be considered. For simplicity, the Cartesian coordinates are transformed into cylindrical coordinates, that is, $r=\sqrt{x^2+y^2}$. Then we have

$$
\begin{aligned}
\varphi_{\mathrm{n}} = & -\frac{\cos(\theta_1)}{\cos(\theta)} \frac{1}{n_1} \frac{dn_1}{dT} \int_0^{L_1} dz \int_0^{\infty} \delta\, d\delta \{ \delta J_1(\delta x_1) \\
& \times [E(\delta) \exp(-\alpha_1 z) + C_1(\delta) \exp(-\beta_1 z) \\
& + C_2(\delta) \exp(\beta_1 z)] + \tan(\theta_1) \cdot J_0(\delta x_1) [\alpha_1 E(\delta) \exp(-\alpha_1 z) \\
& + \beta_1 C_1(\delta) \exp(-\beta_1 z) - \beta_1 C_2(\delta) \exp(\beta_1 z)]\} \exp(i\omega t) \\
& - \frac{1}{2} \frac{\cos(\theta_2)}{\cos(\theta)} \frac{dn_2}{dT} \int_{L_1}^{L_2} dz \int_0^{\infty} \delta\, d\delta \{ \delta J_1(\delta x_2) \\
& \times [F(\delta) \exp[-\alpha_2(z-L_1)] + D_1(\delta) \exp[-\beta_2(z-L_1)] \\
& + D_2(\delta) \exp[\beta_2(z-L_1)]] + \tan(\theta_2) \cdot J_0(\delta x_2) \\
& \times [\alpha_2 F(\delta) \exp[-\alpha_2(z-L_1)] + \beta_2 D_1(\delta) \exp[-\beta_2(z-L_1)] \\
& - \beta_2 D_2(\delta) \exp[\beta_2(z-L_1)]]\} \exp(i\omega t) + \text{c.c.}
\end{aligned} \tag{10}
$$

where $x_1=(z_0-z)\tan(\theta_1)$, $x_2=(z_0-L_1)\tan(\theta_1)-(z-L_1)\tan(\theta_2)$, and $J_1$ 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 nonabsorbing or low-absorbing dielectric thin films, such as optical coatings and 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 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.

Figure 2 shows the dependence of the normal deflection amplitude on the cross-point position for different thermal conductivity ratios of the thin film to substrate at an incident angle of 80°. For comparison, the result for a substrate without thin film is also shown in Fig. 2 (curve 1). As shown in Fig. 2, the thin film influences the normal deflection amplitude mainly near the surface of the substrate where the thin film is deposited. At a low thermal conductivity ratio, the thin film has a 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 maxima of the deflection amplitude increases. The ratio of the two maxima of the deflection amplitude approaches saturation for a high thermal conductivity of the thin film. That is, there exists a thermal conductivity ratio range in which the thermal property of the thin film has an apparent effect on the ratio of the two maxima of deflection amplitudes.

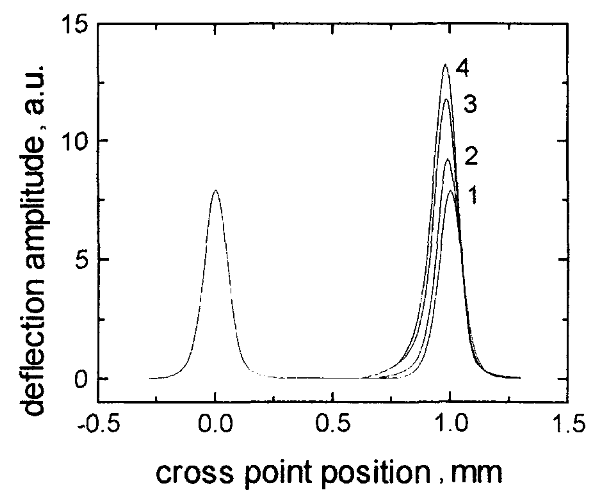


**Fig. 2.** Normal deflection amplitude as a function of cross-point position in the case of a glass substrate. The modulation frequency is 100 Hz and the incident angle is 80°. The curve 1 is the result for a free substrate. The curves 2–4 are the results for the deposited thin films with $K_2/K_1$ of 20, 200, and 2000 respectively, and the thickness of the films is 1 $\mu$m.

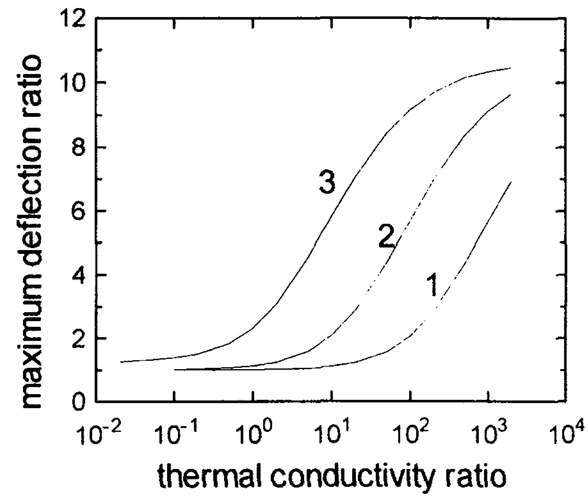


**Fig. 3.** Normalized maximum deflection ratio vs thermal conductivity ratio. The modulation frequency is 100 Hz, the refractive angle is 80°, and the thin-film thickness is (1) 0.1 $\mu$m, (2) 1 $\mu$m, and (3) 10 $\mu$m, respectively.

Figures 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 maxima 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 much lower. Numerical calculations also show that the change in the 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 thin films moves down to the low thermal conductivity region as the thermal conductivity of the substrate decreases.

The above numerical evaluations show that for nonabsorbing thin films deposited on absorbing substrates, the thermal conductivity of the thin film can be determined from the ratio of the deflection maxima, 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 very suited for the measurement of the thermal conductivity of thin films with a high thermal conductivity and thin thickness at a relatively low modulation frequency, which is also possible for the measurement of the thermal conductivity of thin films with a thin thickness and low thermal conductivity at a high modulation

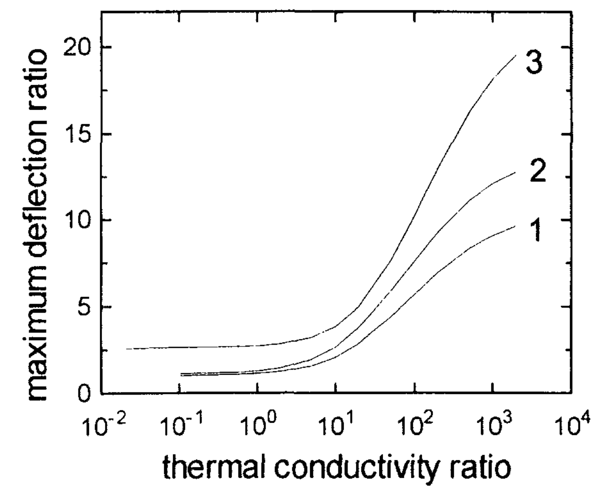


**Fig. 4.** Normalized maximum deflection ratio vs thermal conductivity. The thin-film thickness is 1 $\mu$m, the refractive angle is 80°, and the modulation frequency is (1) 10 Hz, (2) 100 Hz, and (3) 1 KHz, respectively.

frequency. The results also show that a large difference between the thermal conductivity of the thin film and that of the substrate is beneficial to the thermal conductivity measurement of the thin film with the obliquely crossed PTD configuration.

The existence of thin-film absorption considerably complicates the thermal conductivity measurement using the 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 on the cross-point position, the dependence of the total deflection on the cross-point position is very complicated, especially when the absorption of the thin film and that of the substrate are comparable. In this case the determination of the thermal conductivity of the thin film by the ratio of the deflection maxima is very difficult. If the difference between the probe beam deflections caused by the thin film and the substrate is large, it is possible to determine the thermal conductivity of the thin film from the ratio of the deflection maxima. In the case where the absorption of the 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 suitable technique for thermal conductivity measurements of the dielectric thin films. The above numerical evaluation demonstrates that, by employing the ratio of maximum deflections, the obliquely crossed PTD configuration is capable of measuring the thermal conductivity of films over a wide range of values, and the measurement is independent of the substrate absorption. Numerical results further indicate that the obliquely crossed PTD configuration is especially suited for thermal conductivity measurements of thin films with a high thermal conductivity, such as diamond or diamond-like films, which are usually difficult to measure by other PT methods due to the limitations of the modulation frequency, spatial (depth) resolution, sensitivity, etc. However, the amplitude measurement is not always very stable, which may decrease the precision of the method even when the ratio of amplitudes is considered.

## 4. CONCLUSION

A three-dimensional theoretical model for samples consisting of thin films deposited on substrates has been developed to calculate the photothermal deflection with the 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) angles, the thermal conductivity of a transparent or weakly absorbing thin film deposited on an absorbing substrate can be determined. The results demonstrate that the obliquely crossed photothermal deflection technique is well suited for measuring the thermal conductivity of films with a high thermal conductivity and very thin thickness, which is difficult to measure by the transverse PTD scheme (mirage effect) employing the frequency dependences of the amplitude or phase of the deflection signal.

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