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"Thermal Prism" Method for Measuring Thermophysical Properties of Thin Films

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Abstract Theoretical principles underlying the photothermal method for measuring the thickness and thermal properties of a thin film located between two optical elements ("sandwich") are analyzed. The method is based on the irradiation of the assembly by repetitive pulse laser radiation. Radiation is absorbed in the film and causes heating of the optical elements by heat conduction. The element is monitored by a narrow beam of a second low-power laser propagating through the heated region. The beam is deflected due to the spatial variation of the refractive index, and the magnitude of the deflection angle as a function of time contains the relaxation and "wave" components. It is shown that the phase of the "wave" component depends on the thickness and thermophysical properties of the film. The thermophysical properties of the film can be determined, provided that the analogous properties of the optical element are measured previously or otherwise known, by comparing experimentally measured values of the phase shift with theoretical values obtained from the analytical solution of the non-stationary two-dimensional heat conduction equation.

Keywords Coating · Laser heating · Phase shift · Thermal prism

1 Introduction

A number of photothermal methods are used for measuring thermophysical and optical properties of solids, surfaces, and thin coatings. The photothermal method is based on the irradiation of the investigated sample by the modulated radiation of a laser having a relatively high power. The absorbed electromagnetic radiation causes a local increase of temperature and generation of thermal waves in the illuminated specimen.

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The amplitude and the phase of these thermal waves carry information on the thermal properties of the specimen, the properties of surfaces and interfaces, and the presence of subsurface defects and cracks. There are a number of experimental techniques for detecting thermal waves (an overview of these methods can be found in [1]): photoacoustic spectroscopy [2,3], photothermal displacement method [4,5], photothermal radiometry [6,7], "mirage effect" [8], etc. These techniques are generally used for determining thermal properties of a film deposited on a substrate whereby the front and rear film surfaces are in contact with the surrounding gas (air) and a substrate, respectively.

In the photothermal displacement method, the monitoring beam of a second laser is directed onto the front surface irradiated by a powerful laser beam and is reflected from the sample. In the photothermal method known as the "mirage effect," the monitoring beam passes through the heated air above the front surface of a film. In this study we propose the photothermal method for determining the thermophysical properties of a film embedded between two non-opaque optical elements (plates, lenses, fibers, etc.). The proposed method is based on heating the film by repetitive pulse laser radiation. Due to heat conduction, the temperature distribution in optical elements is inhomogeneous and time dependent. Since the refractive index $n(T)$ of an optical element varies with temperature, it depends on spatial coordinates and time, i.e., $n = n(r, z, t)$. The monitoring laser beam propagates through the substrate at a distance z_0 from the film and is deflected because of a refractive-index gradient in the direction of the z axis (see Fig. 1). Consequently, the substrate operates as a time-dependent quasi-optical deflection system ("thermal prism"). The beam deflection angle can be registered in the experiment and contains the relaxation, $\varepsilon_{\mathrm{rel}}$, and "wave," ε_{w}, components. The first component is caused by the diffusion of thermal energy from the film in the sample (it does not depend on the frequency of modulation of a laser pulse), while the second component is associated with modulated heating of the film. The "wave" component can be extracted (identified) from the beam deflection signal and expanded in a harmonic series using Fourier analysis. Comparing the experimentally measured amplitude and phase data with the predicted values obtained from the analytical solution of the heat conduction equation allows determination of the thickness and the thermophysical properties of a film.

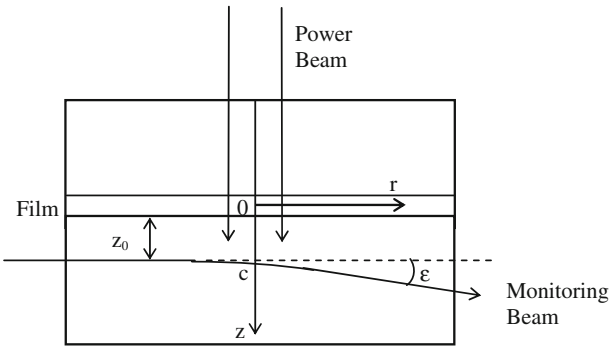

Fig. 1 Schematic of "thermal prism" method

The described quasi-optical deflection system ("thermal prism") can be used not only for determining some properties of the film but also for evaluating the quality of the optical contact between two non-opaque elements, detecting delamination cracks of interfaces [9], controlling cleanliness of the surface of the optical element (in the case of an absolutely clean surface, the "thermal prism" is not formed), and detecting the change in the chemical structure of a film or adhesive exposed to IR laser radiation that is accompanied by the temperature rise [10].

2 Solution of Heat Conduction Problem

The investigated optical element is the "sandwich" composed of two non-opaque substrates (plates, fibers, etc.) and a film embedded between them. The element is irradiated by repetitive pulses of a power laser in a direction normal to the film. The wavelength of the laser radiation is chosen such that the laser radiation is absorbed only in the film. The absorbed radiation energy is transported in the substrates by heat conduction. The theoretical description of the suggested photothermal method is based on the solution of a non-stationary two-dimensional heat conduction equation with the appropriate initial and boundary conditions ($i = 0$ for a substrate and $i = 1$ for a film):

$$c_i \frac{\partial T_i}{\partial t} = \lambda_i \left[\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial T_i}{\partial r} \right) + \frac{\partial^2 T_i}{\partial z^2} \right] + I_0 \sigma_i \exp\left(-\frac{r^2}{r_0^2} \right) f(t), \tag{1}$$

$$T_i|_{t=0} = \left. \frac{\partial T_i}{\partial z} \right|_{r\to\infty} = \left. \frac{\partial T_0}{\partial z} \right|_{z=\pm\infty}, \quad \left. \frac{\partial T_1}{\partial z} \right|_{z=-\Delta/2} = 0,$$

$$T_0 = T_1, \quad \lambda_1 \frac{\partial T_1}{\partial z} = \lambda_0 \frac{\partial T_0}{\partial z} \text{ at } z = 0,$$

$$f(t) = \begin{cases} 1, & 0 < t < \tau_0, \\ 0, & \tau_0 < t < \tau_0 + \tau, \end{cases} \qquad f(t) = f\left[t + n(\tau_0 + \tau) \right], \quad n = 0, 1, 2, \ldots,$$

where c_i is the specific heat, λ_i is the thermal conductivity, I_0 is the laser beam intensity, $\sigma_0 = 0$ (substrate), $\sigma_1 = \sigma$ is a bulk absorption coefficient of the film, r_0 is the laser beam radius, and $f(t)$ is the periodic function with a period $(\tau + \tau_0)$. It is assumed that the film is optically thin, $\sigma\Delta \ll 1$, where Δ is the coating thickness, and, therefore, the temperature field T_i is a symmetric function of the axial coordinate z.

Applying the Laplace–Hankel transform to Eq. 1 yields

$$\frac{\partial^2 \bar{T}_0}{\partial z^2} = \left(\frac{s}{k_0} + p^2 \right) \bar{T}_0,$$

$$\frac{\partial^2 \bar{T}_1}{\partial z^2} = \left(\frac{s}{k_1} + p^2\right)\bar{T}_1 + \frac{I_0 \sigma r_0^2}{2\lambda_1} \exp\left(-\frac{p^2 r_0^2}{4}\right) \bar{f}(s), \tag{2}$$

$$\bar{T}_i\big|_{t=0} = \left.\frac{\partial \bar{T}_i}{\partial z}\right|_{r\to\infty} = \left.\frac{\partial \bar{T}_0}{\partial z}\right|_{z=\pm\infty}, \quad \left.\frac{\partial \bar{T}_1}{\partial z}\right|_{z=-\Delta/2} = 0,$$

$$\bar{T}_0 = \bar{T}_1, \quad \lambda_1 \frac{\partial \bar{T}_1}{\partial z} = \lambda_0 \frac{\partial \bar{T}_0}{\partial z} \text{ at } z = 0, \tag{3}$$

where k_i is the thermal diffusivity,

$$\bar{T}_i(s, p, z) = \int_0^\infty \int_0^\infty T_i(t, r, z) \exp(-st) r J_0(pr) \mathrm{d}t \mathrm{d}r,$$

$$\bar{f}(s) = \frac{1 - \exp(s\tau_0)}{s} \sum_{n=0}^{\infty} \exp[-ns(\tau_0 + \tau)] = \frac{1 - \exp(-s\tau_0)}{s\,\{1 - \exp\,[-s\,(\tau_0 + \tau)]\}}. \tag{4}$$

The general solution of Eq. 2 reads

$$\bar{T}_0 = M_0 \exp(-\chi_0 z), \quad \bar{T}_1 = M_1 \exp(-\chi_1 z) + N_1 \exp(\chi_1 z), \tag{5}$$

where $\chi_i = \sqrt{\frac{s}{k_i} + p^2}$ $(i = 0,\ 1)$ and M_0, M_1, and N_1 are integration constants. Substituting expressions of Eq. 5 into the continuity and boundary conditions of Eq. 3, and after some algebra, we arrive at the expressions for the Laplace–Hankel transform of the temperature distribution in a substrate:

$$\bar{T}_0(s, p, z) = \frac{I_0 \sigma r_0^2}{2\lambda_0 \chi_0} \exp\left(-\frac{p^2 r_0^2}{4}\right) \bar{f}(s) \bar{F}(s, p, z), \tag{6}$$

where

$$\bar{F} = \frac{\exp\,(-\chi_0 z)}{\chi_1} \frac{1}{\frac{\lambda_1 \chi_1}{\lambda_0 \chi_0} + \frac{1}{\tanh(0.5 \chi_1 \Delta)}}. \tag{7}$$

The change in the refractive index n due to the temperature rise T_0 in a substrate is expressed as

$$\Delta n = \left(\frac{\partial n}{\partial T}\right)_0 T_0(t, r, z). \tag{8}$$

The monitoring beam propagates through the substrate at a distance z_0 from the film in the direction perpendicular to the axis of a power laser beam (the axes intersect at point C, see Fig. 1). Spatial variation of the refractive index causes deflection of the monitoring beam. The monitoring beam deflection angle ε is determined by the following relation:

$$\varepsilon = \frac{\partial L}{\partial z} = \frac{2}{n_0}\frac{\partial}{\partial z}\int_0^\infty \Delta n(r, z)\mathrm{d}r = \frac{2}{n_0}\left(\frac{\partial n}{\partial T}\right)_0 \int_0^\infty \frac{\partial T_0}{\partial z}\mathrm{d}r. \tag{9}$$

The Laplace–Hankel transform of the deflection angle is expressed as

$$\bar{\varepsilon}(s, z_0) = \frac{2}{n_0}\left(\frac{\partial n}{\partial T}\right)_0 \int_0^\infty \int_0^\infty \frac{\partial \bar{T}_0}{\partial z} pJ_0(pr)\mathrm{d}p\mathrm{d}r = \frac{2}{n_0}\left(\frac{\partial n}{\partial T}\right)_0 \int_0^\infty \frac{\partial \bar{T}_0}{\partial z}\mathrm{d}p. \tag{10}$$

Equations 6 and 10 yield

$$\bar{\varepsilon}(s, z_0) = \frac{I_0\sigma r_0^2}{n_0\lambda_0}\left(\frac{\partial n}{\partial T}\right)_0 \bar{f}(s)\int_0^\infty \exp\left(-\frac{p^2 r_0^2}{4}\right)\bar{F}(s, p, z_0)\mathrm{d}p. \tag{11}$$

The inverse Laplace transform of Eq. 11 contains the relaxation component, ε_{rel}, which does not depend on the frequency of modulation of a laser heating pulse, and the "wave" component, ε_w, of the monitoring beam deflection angle ε. We are interested in the "wave" component ε_w because the principal measured parameter of the photothermal method is the phase of the deflection angle ε. To determine the inverse Laplace transform of Eq. 11, we use the decomposition theorem:

$$L^{-1}\left[\frac{\Phi(s)}{\Psi(s)}\right] = \sum_{m=1}^{n}\frac{\Phi(s_m)}{(\partial\Psi/\partial s)|_{s=s_m}}\exp(s_m t), \tag{12}$$

where s_m's are the roots of the equation $\Psi(s_m) = 0$. After some algebra, the inverse Laplace transform of Eq. 11 can be written as follows:

$$\begin{aligned}\varepsilon_0(t,\ z_0) = K\int_0^\infty \exp\left(-\frac{p^2 r_0^2}{4}\right)\Bigg[&\frac{\tau_0}{\tau_0+\tau}\bar{F}(0, p, z_0)\\ &+\frac{1}{2\pi i}\sum_{m=1}^{n}\frac{\exp(s_m t)\left[1-\exp(-s_m\tau_0)\right]}{m}\times\bar{F}(s_m,\ p,\ z_0)\Bigg]\mathrm{d}p,\end{aligned} \tag{13}$$

where

$$K = \frac{I_0\sigma r_0^2}{\lambda_0 n_0}\left(\frac{\partial n}{\partial T}\right)_0,\quad i = \sqrt{-1},\quad \omega = \frac{2\pi}{\tau_0+\tau},$$

$s_m = \pm im\omega$ are the roots of the equation $\exp[-s_m(\tau_0 + \tau)] = 1$ (see Eqs. 4, 6, and 11), and $m = 1,\ 2,\ 3, \ldots, n$, except for the case when $\frac{2m\tau_0}{\tau_0+\tau} = N$ is an integer. The first term in the square brackets in Eq. 13 does not depend on time t, while the second term describes oscillations of the deflection angle $\varepsilon(t)$. To determine the phase of oscillations, we substitute the roots s_m into Eq. 13, and after some algebra, we arrive at the following formula for the oscillation term:

$$\frac{\varepsilon_{\mathrm{w}}}{K} = \sum_{m=-\infty}^{\infty} A_m \cos\left[m\omega(t - \tau_0/2) - \Phi_m\right], \tag{14}$$

where

$$A_m = \frac{2}{\pi}\frac{\sin(m\omega\tau_0/2)}{m}\sqrt{I_{\mathrm{c}}^2 + I_{\mathrm{s}}^2}, \quad \Phi_m = \tan^{-1}\left(\frac{I_{\mathrm{s}}}{I_{\mathrm{c}}}\right), \quad I_c = \int_0^{\infty} I(p)\cos\theta \mathrm{d}p,$$

$$I_{\mathrm{s}} = \int_0^{\infty} I(p)\sin\theta \mathrm{d}p, \quad \theta = \frac{pz_0\sin(\varphi_0/2)}{\sqrt{\cos\varphi_0}} + \varphi_1/2 + \tan^{-1}\left(\frac{b_s}{b_c}\right), \tag{15}$$

$$I(p) = \frac{\sqrt{\cos\varphi_1}}{p\sqrt{b_c^2 + b_s^2}}\exp\left[-\frac{p^2r_0^2}{4} - \frac{pz_0\cos(\varphi_0/2)}{\sqrt{\cos\varphi_0}}\right], \quad \varphi_i = \tan^{-1}\left(\frac{m\omega}{k_i p^2}\right).$$

Here A_m, Φ_m are the coefficients of the Fourier spectrum of $\varepsilon_{\mathrm{w}}/K$ and the phase shift of the deflection angle oscillations, respectively. The coefficients b_{c} and b_{s} which depend on the thickness Δ of a film, are determined by the following formulae:

$$b_{\mathrm{c}} = \frac{\lambda_1\sqrt{\cos\varphi_0}}{\lambda_0\sqrt{\cos\varphi_1}}\cos\left[\frac{\varphi_1 - \varphi_0}{2}\right] + R\cos\psi,$$

$$b_s = \frac{\lambda_1\sqrt{\cos\varphi_0}}{\lambda_0\sqrt{\cos\varphi_1}}\sin\left[\frac{1}{2}(\varphi_1 - \varphi_0)\right] + R\sin\psi,$$

$$R = \sqrt{\frac{\cosh(2a_c) + \cos(2a_s)}{\cosh(2a_c) - \cos(2a_s)}},$$

$$\psi = \tan^{-1}[\tan(a_s)\tanh(a_c)] - \tan^{-1}\left[\frac{\tan(a_s)}{\tanh(a_c)}\right], \tag{16}$$

$$a_c = \frac{p\Delta\cos(\varphi_1/2)}{2\sqrt{\cos\varphi_1}}, \quad a_s = \frac{p\Delta\sin(\varphi_1/2)}{2\sqrt{\cos\varphi_1}}.$$

For comparison with Eq. 14, the function $\bar{f}(s)$ (see Eq. 4) that describes the temporal dependence of the repetitive heating laser pulse can be expanded in a Fourier series:

$$\bar{f}(s) = \frac{\tau_0}{\tau_0 + \tau} + \frac{2}{\pi} \sum_{m=1}^{n} \frac{\sin(m\omega\tau_0/2)}{m} \cos[m\omega(t - \tau_0/2)].$$

In a particular case when the thickness of a film is negligibly small, $p\Delta \ll 1$, the obtained formulas can be simplified:

$$\psi = -\varphi_1/2, \quad \sqrt{b_c^2 + b_s^2} = R = \frac{2\sqrt{\cos\varphi_1}}{p\Delta}, \quad \theta = \frac{pz_0 \sin(\varphi_0/2)}{\sqrt{\cos\varphi_0}},$$

$$I_c = \frac{\Delta}{2} \int_0^\infty \exp\left[-\frac{p^2 r_0^2}{4} - \frac{pz_0 \cos(\varphi_0/2)}{\sqrt{\cos\varphi_0}} \right] \cos\theta \, \mathrm{d}p,$$

$$I_s = \frac{\Delta}{2} \int_0^\infty \exp\left[-\frac{p^2 r_0^2}{4} - \frac{pz_0 \cos(\varphi_0/2)}{\sqrt{\cos\varphi_0}} \right] \sin\theta \, \mathrm{d}p. \tag{17}$$

The distance z_0 between the monitoring laser beam and a film is chosen such that the phase shift vanishes,

$$\Phi_1 = \tan^{-1}\left(\frac{I_s}{I_c}\right) = 0, \tag{18}$$

provided that the thickness $\Delta \to 0$. For this purpose we solved numerically the following equation which was obtained from Eq. 17:

$$\int_0^\infty \exp\left[-\frac{\bar{p}^2}{4} - \frac{\bar{p} z_0 \cos(\varphi_0/2)}{r_0\sqrt{\cos\varphi_0}} \right] \sin\left[\frac{\bar{p} z_0 \sin(\varphi_0/2)}{r_0\sqrt{\cos\varphi_0}} \right] \mathrm{d}\bar{p} = 0, \quad \bar{p} = pr_0. \tag{19}$$

Equation 19 has an infinite set of roots z_0 which depend on the modulation frequency, ω, laser beam radius, r_0, and the thermal diffusivity of a substrate, k_0. It is suggested that the monitoring laser beam passes through the heated region at the distance z_0 calculated from Eq. 19. This condition constitutes a normalization procedure that is required to eliminate the system phase shift. The obtained beam deflection signal is registered as a function of time and processed using Fourier analysis to extract the "wave" component ε_w (see Eq. 14) and the phase shift Φ_m. In particular, for the case of a thin film ($\bar{p}\Delta/r_0 \ll 1$), the phase shift $\Phi_1 \to 0$, and it increases with an increase in the thickness of a film. The phase shift depends also on the thermal diffusivity of a film k_1. Therefore, by measuring Φ and comparing the results of measurements with the theoretical predictions, one can determine the thickness and thermal diffusivity of a film.

We also calculated the deflection angle ε of the monitoring laser beam for the case of an assembly composed of a non-opaque substrate and an optically thick film ($\sigma\Delta \gg 1$). In this approximation, the powerful laser radiation is absorbed in the narrow subsurface region of a film. The heat conduction equation with the appropriate boundary condition ($i = 0$ for a substrate and $i = 1$ for a film) is expressed as

$$c_i \frac{\partial T_i}{\partial t} = \lambda_i \left[\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial T_i}{\partial r} \right) + \frac{\partial^2 T_i}{\partial z^2} \right], \quad i = 0,\ 1,$$
$$\lambda_1 \frac{\partial T_1}{\partial z}\bigg|_{z=-\Delta} = I_0 \beta \exp\left(-\frac{r^2}{r_0^2} \right) f(t), \tag{20}$$

where β is a fraction of the absorbed radiation. The remaining initial and boundary conditions are given by Eq. 1. The Laplace–Hankel transform of a temperature field in a substrate is given by the following expression:

$$\bar{T}_0(s, p, z) = \frac{I_0 \beta\, r_0^2}{\lambda_0 \chi_0} \exp\left(-\frac{p^2 r_0^2}{4} \right) \bar{f}(s) \bar{F}(s, p, z), \tag{21}$$

where

$$\bar{F} = \frac{\exp(-\chi_0 z)}{\left[\exp(\chi_1 \Delta) - \exp(-\chi_1 \Delta) \right]} \frac{1}{\frac{\lambda_1 \chi_1}{\lambda_0 \chi_0} + \frac{1}{\tanh(0.5 \chi_1 \Delta)}}.$$

After some algebra, we obtained the oscillation term in the form of Eq. 14 with the following changes in Eq. 15:

$$\theta = \frac{p z_0 \sin(\varphi_0/2)}{\sqrt{\cos \varphi_0}} + \tan^{-1}\left[\frac{\tan(2a_s)}{\tanh(2a_c)} \right] + \tan^{-1}\left(\frac{b_s}{b_c} \right)$$

$$I(p) = \frac{1}{\sqrt{b_c^2 + b_s^2}\sqrt{2[\cosh(4a_c) - \cos(4a_s)]}} \exp\left[-\frac{p^2 r_0^2}{4} - \frac{p z_0 \cos(\varphi_0/2)}{\sqrt{\cos \varphi_0}} \right]. \tag{22}$$

The remaining equations in the set of Eqs. 15 and 16 are not changed.

3 Results and Discussion

The photothermal experiment for measuring the thickness and the thermal properties of a film includes irradiation of an optical element by repetitive pulses of a powerful laser and monitoring the affected region by a second laser beam in a direction parallel to a film (see Fig. 1). Propagating through the heated region, the monitoring beam is

deflected due to the gradient of the refractive index. The modulated beam deflection signal is expanded in a Fourier series to determine the phase shift Φ_m (see Eq. 14) and the coefficient A_m that determines the amplitude of the signal. The phase shift measurement is preferable over the amplitude A_m because the former is independent of the laser beam intensity I_0, the absorption coefficient σ, and the temperature coefficient of the refractive index $(\partial n/\partial T)_0$. Besides, the phase shift measurement is more accurate and sensitive to variations of the thermal properties of a coating in comparison with the amplitude measurement. The thermophysical properties and thickness of a film can be determined by fitting the experimental and the theoretically predicted dependences of the phase shift (see Eqs. 15–17, 19, and 22) versus the film properties.

We used the repetitive laser pulses for heating the film and obtained an expression for the beam deflection angle (signal) in the form of a Fourier series including the harmonic terms with the coefficient A_m, the frequency $m\omega$, and the phase shift Φ_m ($m = 1, 2, \ldots, n$, except for the case when $\frac{2m\tau_0}{\tau_0+\tau} = N$ is an integer (Eq. 14)). Consequently, using the single effect of the repetitive laser pulses on an assembly, we obtained the response of the investigated sample at the modulation frequency ω and, also, at the frequencies 2ω, $3\omega, \ldots$. This allows using not only the amplitude A_1 and the phase shift Φ_1 for determining the thermophysical properties as in the previous studies that have employed harmonic heating (see [1,6]), but also A_m and Φ_m for $m > 1$ which are obtained from Eqs. 15, 16, and 22.

We considered the following two assemblies: weakly absorbing laser radiation polyethylene film located between two AgBrCl substrates and an absorbing metallic (Ni) film deposited on an AgBrCl substrate. These materials differ significantly in their thermal and optical properties. We used the following values of the thermal diffusivity and thermal conductivity of polyethylene, Ni, and AgBrCl: $k = (1.1 \times 10^{-7}, 16.8 \times 10^{-6}$, and $0.5 \times 10^{-6})\ \mathrm{m^2 \cdot s^{-1}}$ and $\lambda = (0.3, 69,$ and $1.1)\mathrm{W \cdot m^{-1} \cdot K^{-1}}$, respectively (see [11]). The parameters of a power laser beam are as follows: $r_0 = 1\,\mathrm{mm}$; $\tau_0 = 3\,\mu\mathrm{s}$; and $\omega = 10$, 20, and 50 Hz. The average rise in temperature over a heated zone T can be estimated by the following formula [4]:

$$T = \frac{\beta I_0}{\lambda}\sqrt{\frac{k}{\omega}},$$

where I_0 is the laser beam intensity ($\mathrm{W \cdot m^{-2}}$) and β is the fraction of laser radiation absorbed by a coating. Calculations show that to attain the temperature rise $\mathrm{T} \approx 10$–$100\,\mathrm{K}$ during irradiation of a polyethylene coating by a CO_2 laser (using the above values of λ, k, r_0, and ω), the required power P of the laser must be of the order of 1–10 W (see [10]). For the case of a metal film with high reflectance (a nickel film exposed to a CO_2 radiation), one can significantly increase the absorptivity of a film by depositing a thin strongly absorbing layer (for example, graphite). Then the required power of the laser $P \approx$ 1–10 W remains unchanged.

Consider the results obtained for the specimens with optically thin polyethylene and optically thick Ni films. Figures 2 and 3 show the dependences of the coefficient A_1 and the phase shift Φ_1 versus the thickness Δ of the polyethylene (Fig. 2) and metallic (Fig. 3) films for different values of the modulation frequency ω (the

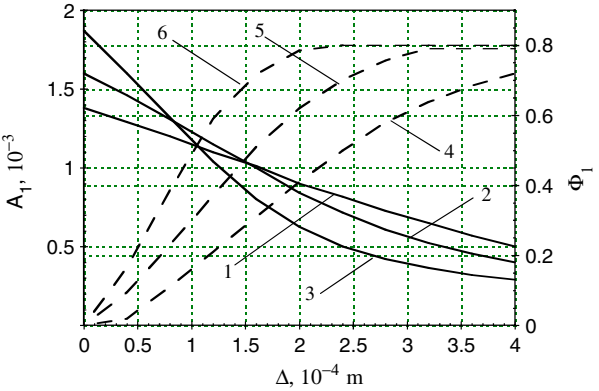

Fig. 2 Dependence of the coefficient A_1 (curves 1–3) and the phase shift Φ_1 (4–6) on the film (polyethylene) thickness Δ for different values of the modulation frequency ω; Curves 1, 4: $\omega = 10\,\text{Hz}$, Curves 2, 5: $\omega = 20\,\text{Hz}$, Curves 3, 6: $\omega = 50\,\text{Hz}$

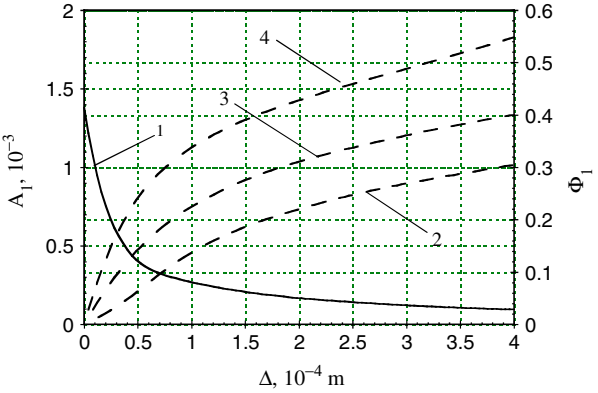

Fig. 3 Dependence of the coefficient A_1 (curve 1) and the phase shift Φ_1 (2–4) on the film (Ni) thickness Δ for different values of the modulation frequency ω; Curve 1: $\omega = 10\,\text{Hz}$, Curve 2: $\omega = 10\,\text{Hz}$, Curve 3: $\omega = 20\,\text{Hz}$, Curve 4: $\omega = 50\,\text{Hz}$

normalized pulse duration $\bar{\tau} = \tau_0/(\tau_0 + \tau) = 0.5$). The distance z_0 between the monitoring laser beam and the film is calculated from Eq. 19 for different values of the modulation frequency ω. In particular, the dimensionless distance z_0/r_0 is equal to 2.03, 1.43, and 0.90 for $\omega = 10$, 20, and 50 Hz, respectively. The coefficient A_1 is the monotonically decreasing function of a film thickness Δ for optically thin and optically thick films, but the sensitivity of A_1 with respect to variations of Δ is significantly higher for the case of the optically thick film (especially, at $\Delta < 50\,\mu\text{m}$). In Eq. 14 the coefficient $A_2 = 0$, and A_3 does not exceed 3% of A_1 for the normalized pulse duration $\bar{\tau} = 0.5$. Calculations showed that the sensitivity $(\frac{\partial \Phi_1}{\partial \Delta})$ of the phase shift Φ_1 with respect to variations of Δ increases as the thickness of a film decreases. For the case of the optically thin film, the phase shift tends to $(\pi/4)$ as the thickness Δ increases, and the sensitivity vanishes, $(\frac{\partial \Phi_1}{\partial \Delta}) \to 0$, especially, in a high frequency range. Consequently, application of the photothermal

method at high modulation frequencies is most efficient for thin films. If the thickness Δ becomes of the order of several hundred microns it is necessary to use the method described above at low-modulation frequencies or apply another photothermal method.

The sensitivity of the phase shift Φ to the variations of the thermal diffusivity k and thermal conductivity λ of a film is characterized by the slope of the curves $\Phi(k)$ and $\Phi(\lambda)$. Figure 4 shows the calculated values of the phase shift Φ_1 as a function of the normalized thermal diffusivity k/k_1 and thermal conductivity λ/λ_1 obtained for the optically thick (Ni, curves 2–4) and thin (polyethylene, curves 1, 5) films. In both cases the thickness of the film is equal to 50 μm, and the calculations have been performed for modulation frequencies of 10 and 50 Hz. The sensitivity of the phase shift to variations of thermal diffusivity is higher in comparison with the sensitivity to variations of thermal conductivity. With the increase of the modulation frequency, the sensitivity of the phase shift increases for the case of an optically thick film. As for an optically thin film, the dependence of the sensitivity on the modulation frequency is more involved because it is also determined by the thickness of a film.

We also determined the Fourier series expansion of the beam deflection angle (see Eq. 14) for different values of the relative pulse duration $\bar{\tau}$. For example, the Fourier series expansion for the beam deflection angle reads ($\Delta = 40\,\mu$m, $\bar{\tau} = 0.333$, $\omega = 10$ Hz):

$$\frac{\varepsilon_w}{K} = A_1 \cos\left[10\left(t - \frac{\tau_0}{2}\right) - \Phi_1\right] + A_2 \cos\left[20\left(t - \frac{\tau_0}{2}\right) - \Phi_2\right] + \cdots, \quad (23)$$

where

$$A_1 = 0.0277,\ A_2 = 0.0036,\ A_3 = 0,\ A_4 < 10^{-4},\quad \Phi_1 = 0.16,\ \Phi_2 = 1.60.$$

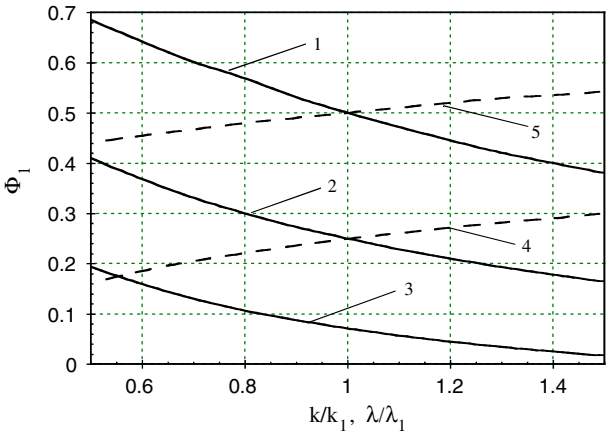

Fig. 4 Phase shift Φ_1 as a function of variations of the thermal properties k/k_1 (curves 1–3 and λ/λ_1 (curves 4, 5) for different values of modulation frequency ω; Curves 1, 2, 5: $\omega = 50$ Hz, Curves 3, 4: $\omega = 10$ Hz

The first two terms in the expansion (Eq. 23) reproduce with high accuracy the Fourier expansion of the angle ε. The measured photothermal deflection signal can be expanded in harmonic series terms using Fourier analysis:

$$\varepsilon_w(t) = B_1 \cos\left[\omega\left(t - \frac{\tau_0}{2}\right) - \Phi_1\right] + B_2 \cos\left[2\omega\left(t - \frac{\tau_0}{2}\right) - \Phi_2\right] + \cdots, \quad (24)$$

where B_m's are the coefficients of expansion depending on the laser beam parameters (power, beam radius), the optical characteristics (absorption coefficient, temperature coefficient of the refractive index), and the thermal properties of a film and a substrate. Comparing the measured values of the phase shift, Φ_1, Φ_2 given by Eq. 24 with the theoretically predicted values (Eqs. 15–17) allows determination of the thickness and one of the thermal properties of a film. The ratio $\bar{B} = B_2/B_1$ (or B_3/B_1 for $\bar{\tau} = 0.5$) depends on the thickness and thermal properties of a film and does not depend on the laser beam parameters, optical characteristics, etc. Expression for $\bar{B}$ can be also found from Eq. 23, e.g., $\bar{B} = A_2/A_1$ (or A_3/A_1 for $\bar{\tau} = 0.5$). Comparing the experimentally obtained value of $\bar{B}$ with the theoretically predicted one (Eq. 23) yields an additional criterion for testing the validity of the theoretical analysis and reliability of the experimental data. Figure 5 shows the dependences of the coefficients A_m and the phase shift Φ_m for $m = 1, 2$ as functions of the thickness Δ for the laser beam radii $r_0 = 1$ and 0.5 mm, the modulation frequency $\omega = 10$ Hz, and the normalized pulse duration $\bar{\tau} = 0.25$. It is found that with a decrease of parameter $\bar{\tau}$, contributions of the terms with $m = 2, 3, \ldots$ increase (see Eq. 23). For example, the ratio $\bar{B} = A_2/A_1$ is equal to 0, 0.13, 0.18, and 0.24 for $\bar{\tau} = 0.5, 0.33, 0.25,$ and 0.10, respectively. Therefore, to use the second harmonics in a Fourier expansion (Eqs. 23, 24) as additional criteria for testing the reliability of the experimental data, it is preferable to maximize the signal magnitude A_2 using the powerful laser with low normalized pulse duration $\bar{\tau}$. Calculations showed that variation of the laser beam radius $r_0$causes a significant change of the distance z_0 (Eq. 19)

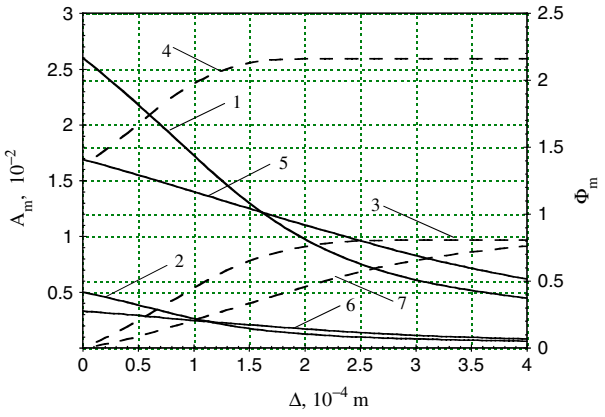

Fig. 5 Coefficients A_m and the phase shift Φ_m versus thickness Δ of the polyethylene film for different values of the laser beam radius r_0 ($\omega = 10$ Hz, $\bar{\tau} = 0.25$); Curve 1: A_1, Curve 2: A_2, Curve 3: Φ_1, Curve 4: Φ_2 ($r_0 = 1$ mm), Curve 5: A_1 Curve 6: A_2 Curve 7: Φ_1($r_0 = 0.5$ mm)

between the monitoring laser beam and a film, namely, z_0 increases as the radius r_0 decreases. For example, for the laser beam with a radius $r_0 = 1$ mm and 0.5 mm, the distance z_0 is equal to 1.03 and 2.13 mm, respectively. Calculations showed that the value of r_0 affects the phase shift Φ_1 (curves 3 and 7 in Fig. 5) and the signal magnitude A_1 (curves 1 and 5 in Fig. 5) at $\Delta < 25\,\mu$m. The magnitudes of Φ_2 and A_2 depend weakly on the laser beam radius r_0 (curves 2, 6).

4 Conclusions

We described the algorithm of the photothermal method ("thermal prism") for measuring the thickness and thermophysical properties of a film deposited on a non-opaque substrate. To this end, a film is irradiated by repetitive pulses of a powerful laser causing a temperature rise in a subsurface region of a substrate. The monitoring beam of another laser propagates over the heated region of a substrate and is deflected due to non-uniform distribution of the refractive index. The beam deflection angle is the principal measured parameter of the "thermal prism" method. We obtained expressions for the beam deflection angle in a form of Fourier series composed of the harmonic functions with the phase shift that depends on the thickness and the thermal properties of a film. Expanding the measured photothermal deflection signal in a Fourier series of harmonic functions and comparing the measured and the theoretically predicted values of the phase shift allows determination of the thickness and thermophysical properties of a film. For the case of a non-opaque film, the phase shift tends to a constant value with an increase in the thickness of a film. Therefore, applicability of the suggested method for a film with a thickness $\Delta > 100\,\mu$m is rather limited. In the case of an opaque film, the phase shift monotonically increases with an increase in the thickness Δ, and the proposed method can be used without any limitations. We also estimated the sensitivity of the phase shift to variations of the thermal properties of a film for different modulation frequencies.

Nomenclature

c	Specific heat
I_0	Laser beam power intensity
$J_i(x)$	i-th-order Bessel function of the first kind
k	Thermal diffusivity
L	Optical path of the monitoring beam in a substrate
$n(T_0)$	Refractive index of a substrate
r	Radial coordinate
r_0	Radius of a laser beam
s, p	Parameters of Laplace–Hankel transforms with respect to t and r, respectively
t	Time
T	Temperature
$\bar{T}(s, p, z)$	Laplace–Hankel transform of temperature
z	Axial coordinate

z_0 Distance between a film and the monitoring laser beam
β Fraction of the absorbed radiation
Δ Coating thickness
Φ_m Phase shift of the monitoring beam deflection angle
ε Laser beam deflection angle
λ Thermal conductivity
σ_i Absorption coefficient
τ_0 Duration of a single laser pulse
ω Cyclic modulation frequency of a powerful laser beam

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