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Discriminating the role of sample length in thermal lensing of solids

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Thermal lens (TL) is a key effect in laser engineering and photothermal spectroscopy. The amplitude of the TL signal or its dioptric power is proportional to the optical path difference (OPD) between the center and border of the beam, which is proportional to the heat power (Ph). Due to thermally induced mechanical stress and bulging of end faces of the sample, OPD depends critically on the geometry of the sample. In this investigation, TL measurements were performed as a function of the sample length keeping the same Ph. It is experimentally demonstrated that for materials with positive ∂n/∂T OPD increases typically 30 to 50% with the decrease of sample length (from long rod to thin-disk geometry). For materials with negative ∂n/∂T, this variation is much larger due to the cancelation of the different contributions to OPD with opposite signs. Furthermore, the experimental investigation presented here validates a recently proposed unified theoretical description of the TL effect. © 2014 Optical Society of America

The dual-beam mode-mismatched thermal lens (MMTL) spectrometry was developed to improve the sensitivity of TL measurements. It is a very sensitive, accurate, and simple method for the determination of TL induced phase-shifts and thermo-optical properties of laser materials, such as thermal diffusivity and the fluorescence quantum efficiency (η) of laser media, without requiring complicated setups or calibration procedures [11]. This method was used to determine the temperature coefficient of the optical path of several laser and optical materials [12]. It was also applied to determine fth and the thermal load of a diode end-pumped Nd:YAG laser (oscillating at 1.06 and 1.34 μm) [13]. In this study, the MMTL method was used to determine the evolution of the fth with the sample length in order to discriminate the effect of thermal stress. The experiments were performed in four different materials: calcium aluminosilicate (CAS), Q98 from Kigre Inc., and BK7 borosilicate and Zerodur glass from Schott. These materials were selected for their thermo-optical properties (Table 1). CAS is typical oxide glass with positive ∂n/∂T and positive linear thermal expansion coefficient [14,15]. Q98 is a Nd3+ doped phosphate glass with negative ∂n/∂T, which was designed to be an athermal optical material. BK7 is a typical highly transparent borosilicate glass. Zerodur is a glass ceramic with an extremely low coefficient of thermal expansion. It was observed that the TL signals, and consequently fth, are in complete agreement with the theoretical predictions of the recently developed theory [8,9]. Although, the results reach the expected “thin-disk”
Table 1. Physical Properties of the Samples [14,15]. Parameters for Q98, Zerodur, and BK7 are from Vendors.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Units</th>
<th>CAS</th>
<th>Q98</th>
<th>Zerodur</th>
<th>BK7</th>
</tr>
</thead>
<tbody>
<tr>
<td>ρ</td>
<td>kg/m³</td>
<td>2890</td>
<td>3099</td>
<td>2530</td>
<td>2510</td>
</tr>
<tr>
<td>n</td>
<td></td>
<td>1.63</td>
<td>1.55</td>
<td>1.54</td>
<td>1.517</td>
</tr>
<tr>
<td>c</td>
<td>J/kg·K⁻¹</td>
<td>810</td>
<td>800</td>
<td>800</td>
<td>858</td>
</tr>
<tr>
<td>k</td>
<td>Wm⁻¹·K⁻¹</td>
<td>1.35</td>
<td>0.82</td>
<td>1.46</td>
<td>1.114</td>
</tr>
<tr>
<td>∂n/∂T</td>
<td>m⁻¹·K⁻¹</td>
<td>2.517</td>
<td>5.3</td>
<td>-4.5</td>
<td>14.3</td>
</tr>
<tr>
<td>αₜ</td>
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<td>9.9</td>
<td>0.02</td>
<td>7.1</td>
</tr>
<tr>
<td>Aₑ</td>
<td>m⁻¹</td>
<td>62</td>
<td>46</td>
<td>18</td>
<td>0.2</td>
</tr>
<tr>
<td>ν</td>
<td></td>
<td>0.23</td>
<td>0.24</td>
<td>0.24</td>
<td>0.208</td>
</tr>
<tr>
<td>E</td>
<td>Pa⁻¹</td>
<td>0.0136</td>
<td>0.0136</td>
<td>0.0136</td>
<td>0.0136</td>
</tr>
<tr>
<td>q</td>
<td></td>
<td></td>
<td>10⁻¹²</td>
<td>0.09</td>
<td>0.35</td>
</tr>
<tr>
<td>q⊥</td>
<td>10⁻¹²</td>
<td>0.90</td>
<td>1.95</td>
<td>1.86</td>
<td></td>
</tr>
</tbody>
</table>

and “long-rod” values in their respective limits, we found that the thin-disk formula has been used indiscriminately in the literature, sometimes in situations better described by the long-rod case.

The experiments were performed in a mode-mismatched configuration, as shown in Fig. 1, at room temperature. A continuous wave (cw) TEM₀₀ Gaussian excitation laser beam at the wavelength λₑ = 514.5 nm irradiates a weakly absorbing sample of thickness l₀, inducing a time-dependent temperature gradient and a correspondent thermoelastic surface displacement. A weak TEM₀₀ Gaussian beam at λₑ = 632.8 nm, almost collinearly arranged with the excitation laser beam, travels through the sample and probes the TL. The radii of the excitation and probe beams in the sample are wₑ = 55 μm and wᵖ = 377 μm, respectively. The confocal distance of the excitation laser beam is Zₑ = 18.5 mm. The probe beam propagates in the z-direction, and the sample is placed at z = 0. The distance between the sample and the probe beam waist of radius w₀p is Z₁ = 13.3 cm, and the distance between the sample and the detector plane is Z₂ = 276.6 cm. As in most TL analytical approaches, it is assumed that the radial dimensions of the sample are large compared with the excitation beam radius, and the excitation time is short enough to avoid edge effects, i.e., the sample is assumed to be radially infinite.

Figure 2 displays the time-resolved TL transients obtained from the experiments for Zerodur and CAS samples with different thicknesses. For both materials the experiments were designed to compare the TL signal of the different samples keeping the product P₁l₀ constant (P₁ is the excitation beam power). The signals from five different Zerodur samples cannot be distinguished while for CAS the signal amplitude increases as the sample thickness is decreased. The Zerodur data can be interpreted by the standard TL theory [11], where the transient signal amplitude is proportional to θₜ, and the CAS signal cannot be explained by the standard simplified model. This is clearly shown by the fact that the amplitude of the TL signal decreases as l₀ increases.

Fig. 1. Schematic diagram of an apparatus for time-resolved TL experiment. L₁, M₁, P₁, and f stand for lenses, mirrors, photodiodes, and focal distances, respectively.

Fig. 2. Experimental TL transients for (a) Zerodur (l₀ = 0.5, 1.0, 1.9, 3.0, 5.0 mm) and (b) CAS glasses with the product P₁l₀ constant. Open symbols: experimental data; solid lines: best curve fitting.

\[
θₜ = \delta l₀ / T,
\]

where \(δ = P₁Aₑ \phi / (kλₑ)\). \(Aₑ\) is the optical absorption coefficient at the excitation beam wavelength, \(k\) is the thermal conductivity, \(\phi\) is the fraction of absorbed energy converted into heat, and \(χ\) is thermo-optical coefficient. The characteristic TL signal response time is given by \(τₜ = w₀p / 4D\), where \(D = k / (ρc)\) is the thermal diffusivity, in which \(ρ\) is the mass density and \(c\) is the specific heat. The parameters \(θₜ\) and \(τₜ\) were obtained by fitting the experimental transient signals of Fig. 2 to the standard model [11]. For Zerodur, from \(τₜ\) the thermal diffusivity value was obtained as \(D = (8.2 \pm 0.3) \times 10^{-7} \text{ m}^²/\text{s}\), which is in good agreement with the literature (Table 1). Zerodur has no fluorescence under excitation at 514.5 nm, and it can be assumed that all absorbed energy is transformed into heat, i.e., \(\phi = 1\). Therefore, using \(θₜ\) from regression in Eq. 1, we obtained for Zerodur, as in a liquid, that \(χ ≈ (∂n/∂T)\) since this material was designed to have negligible thermal expansion (\(αₜ \approx 0\)). When the factor \(P₁l₀\) is constant, the absorbed power is constant and, by Eq. (1), \(θₜ\) and the TL signal are also expected to remain the same. Hence, Fig. 2(b) demonstrates that the CAS signal cannot be explained by the standard simplified model. This is clearly shown by the fact that the amplitude of the TL signal decreases as l₀ increases. The problem here is the assumption that the phase profile is proportional to the temperature profile by \(Φ(r,t) \propto χT(r,t)\). In fact, due to the thermoelastic effect...
in solid samples, this assumption is only valid in two limiting cases: thin disk and long rod. For thin samples, the so-called plane-stress approximation results in \[ \chi^0 = \frac{\partial n}{\partial T} + (n - 1)(1 + \nu)a_T + \frac{n^3Ea_T}{4}(q_\parallel + q_\perp). \] (2)

where \( \nu \) is the Poisson’s ratio, \( n \) is the refractive index of the sample, \( E \) is the Young’s modulus, and \( q_\parallel \) and \( q_\perp \) refer to the piezo-optic coefficients for stresses applied parallel and perpendicular to the polarization axis, respectively. The other limit, where the sample thickness is larger than the radius of the region affected by the thermoelastic effect, the so-called “long rod” approach, uses the plane strain approximation [5],

\[ \chi^\infty = \frac{\partial n}{\partial T} + \frac{n^3Ea_T}{4(1 - \nu)}(q_\parallel + 3q_\perp). \] (3)

Using the published data for CAS we estimate \( \chi^0 = 11.8 \times 10^{-6} \) K\(^{-1}\) and \( \chi^\infty = 7.9 \times 10^{-6} \) K\(^{-1}\). In fact, if we use the simplified theory to fit the experimental data, we obtain \( \chi^0 = (11.5 \pm 0.9) \times 10^{-6} \) K\(^{-1}\) for the sample with \( l_0 = 0.6 \) mm and \( \chi^\infty = (8.5 \pm 0.8) \times 10^{-6} \) K\(^{-1}\) for \( l_0 = 5.6 \) mm.

In general, samples with arbitrary thickness, can be analyzed using the analytical model recently developed by Malacarne and co-workers [8,9]. In [10] it was shown that the difference introduced in the phase shift by the airglass heat coupling need only be considered in the case of extremely thin samples. Under normal experimental conditions, the adiabatic second-kind (Neumann) boundary condition at the outer surfaces could be considered. Using the unified model for calculating the laser-induced wavefront distortion in optical materials [8,9], the sample contribution for the phase shift in the case of a moderate optical absorbing material, probed by an axially symmetric unpolarized Gaussian beam, is

\[ \Phi(g, t) = \Delta \int_0^\infty l_0 \left(1 - \frac{A_{l0}}{2}\right) \chi(a, l_0)e^{-\frac{a^2}{aw_0^2}} da \times (1 - e^{-\frac{\pi^2aw_0^2}{l_0^2}}) \left[ J_0\left(\frac{aw_0}{\sqrt{mg}}\right) - 1\right]a^{-1} da \] (4)

with \( m = (w_1/aw_0)^2 \), and

\[ \chi(a, l_0) = \frac{\partial n}{\partial T} + \frac{4(n - 1)(1 + \nu)a_Th(a, l_0)}{l_0a} + \frac{n^3Ea_T}{4(1 - \nu)}\left( (q_\parallel + 3q_\perp) - \frac{4(q_\parallel + q_\perp(2 + \nu))h(a, l_0)}{l_0a} \right). \] (5)

where \( h(a, l_0) = [\cosh(l_0a) - 1]/[l_0a + \sinh(l_0a)] \). \( J_0(x) \) is the Bessel function of the first kind. The factor \( \left[ 1 - (A_{l0})/2 \right] \) in Eq. (4) accounts for the effect of moderated optical absorption coefficients. When the sample thickness is smaller than the radius of the region affected by the thermoelastic effect, the above expression reduces to the plane-stress approximation given by Eq. (2), which is indiscriminately used in the literature. In the other limit, for sample thicknesses larger than the radius of the region affected by the thermoelastic effect, Eq. (5) recovers the plane-strain approximation, Eq. (3). We remind the reader that the plane-stress and plane-strain approximations are valid only under the above restrictive conditions.

The thermal characteristic time \( \tau \) and the parameter \( \beta \) were obtained from regression using the Fresnel–Kirchhoff diffraction integral

\[ I(t) = \frac{\int_0^\infty \exp[-(1 + iV)g - i\Phi(g, t)]dg^2}{\int_0^\infty \exp[-(1 + iV)g]dg^2} \] (6)

with Eqs. (2), (3), or (5) in the phase shift. \( V = Z_1/Z_c + Z_1[(Z_1/Z_0)^2]/Z_0 \), \( Z_c = 4.65 \) cm is the confocal distance of the probe beam. Regressions with the models were performed using the “NonlinearModelFit” function in Wolfram Mathematica 7. Using the unified model, it was observed that \( \beta \) presented a linear dependence with \( P_c \), as expected.

Figure 2 presents the parameter \( \beta/P_c \) obtained from the fitted TL transients with the unified and approximated theories normalized by the excitation beam power. It shows that the unified model gives the same \( \beta/P_c \) value regardless of the thickness of \( l_0 \) for both samples CAS and Q98. It should be noticed that the unified model covers the limiting cases, thin disk and long rod, in their respective approaches with \( \beta = \theta_T/\chi(l_0) \). For instance, the “long rod” result is close to the expected value for \( l_0 \) larger than 5.5 mm and the “thin disk” for \( l_0 \) thinner than 0.5 mm. Similar behavior was observed for \( l_0 > 9 \) mm and \( l_0 < 0.5 \) mm in the case of Q98. Moreover, as an athermal glass, Q98 presents negative \( \partial n/\partial T \) that cancels the

![Fig. 3. Parameter \( \beta/P_c \) versus \( l_0 \) obtained by the fit of experimental data for (a) CAS and (b) Q98 glasses using the models: (open circle) unified model, (open square) plane-stress approximation, and (open triangle) plane-strain approximation. Dashed lines are guides for the eye. The standard deviations are smaller than 5% for CAS and smaller than 3% for Q98.](https://example.com/image)
Usually positive contributions of the expansion and stress. In this case, the discrepancy between $\chi^0$ and $\chi^\infty$ is remarkable ($\chi^0 \approx 4\chi^\infty$) due to the relative larger contribution of the stress. The averaged thermal diffusivities were found to be $D = (5.0 \pm 0.4) \times 10^{-7}$ m$^2$s$^{-1}$ for CAS and $D = (3.0 \pm 0.3) \times 10^{-7}$ m$^2$s$^{-1}$ for Q98. These values are within the expected values for these materials [14,15].

Figure 4 shows the amplitude (steady-state signal) of the TL signals normalized by the product $P_d l_0$ as a function of $l_0$ for BK7 samples with thicknesses varying from 1.1 to 30 mm. The amplitude of the TL signal is very small ($\theta_T \approx 10^{-3}$) due to the very small absorbance at 514.5 nm ($\sim 2 \times 10^{-3}$ cm$^{-1}$), and consequently the experimental uncertainty is larger than in Fig. 2. Even though, it can be clearly seen that the signal increases as the thickness decreases, in agreement with $\chi^0 \approx 1.3\chi^\infty$, as expected from calculated published data (Table 1). In this case, the experimental data is compared with transient curves obtained by the unified model (Eq. 5) and is in very good agreement for all thicknesses.

It is important to mention that the simplified and unified theories assume the excitation beam radius, $w_{0e}$, to be constant inside the sample along the z-direction. This approximation has been tested by comparing numerically the intensity signal using $w_e(z) = w_{0e}$ and $w_e(z) = w_{0e}\sqrt{1 + [(z - l_0/2)/z_c]}$, and it was found that the error between the two is less than 0.5%.

In conclusion, we have performed the first experimental validation, to our knowledge, of the recently proposed model to describe the laser-induced wavefront distortion in optical elements using the TL technique. We showed that consistent values are obtained, irrespective to the sample thickness, when we apply the unified model to describe the laser induced wavefront distortion (TL effect). The results allow us to obtain more accurate physical properties from TL experiments. In addition, for the laser source designers, the unified solution for the optical path in analytic terms, regardless of the thickness of the element, allows the calculation of the TL effect in optical materials without the use of special numerical codes.

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References