Thermal mirror and thermal lens techniques for semitransparent material characterization

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Abstract. The mode-mismatched thermal lens technique (TL) has been used to study many semitransparent materials. Its theoretical development considers weakly absorbing materials, which introduce restrictions on the sample's optical thickness. However, the same equipment required by TL can be used to perform the thermal mirror (TM) experiment, which is useful to characterize materials with any optical absorption coefficient. In this work, we investigate a simple correction to be used in the TL model, making it possible to apply TL to a wide range of materials. Using TL and TM, we have determined the temperature coefficient of the optical path length (ds/dT) of a glass.

1. Introduction

Many photothermal techniques have been developed to investigate thermal, optical and mechanical properties of materials. The nondestructive characteristic of these techniques is a great attractive. Particularly, the mode-mismatched thermal lens [1] (TL) technique is useful to determine properties of glasses [2], oils [3], crystals [4] and any other semitransparent material, provided that certain conditions are obeyed. While the sample is monitored by a probe beam, the sample is warmed using the excitation beam. The temperature rise in the sample modifies the optical path of the first beam, which is interpreted as a phase shift in its electric field. Measuring the intensity of the probe beam center in function of the time, some sample properties can be obtained, such as: thermal diffusivity (D), the temperature coefficient of the optical path length (ds/dT), optical absorption coefficient (Ae) and luminescence quantum efficiency η.

The complex electric field of the center of a Gaussian probe beam, after crossing the sample, is [1]

\[ U(Z_1 + Z_2, t) = C_I \int_0^g \exp \left[ (iV - 1)g - i\Phi(g, t) \right] dg, \]  

in which \( g = r/\omega_{hp} \), \( C_I = B(1/2\pi\omega_{hp}^{1/2}(\lambda_pZ_0)) \exp(2i\pi Z_2/\lambda_p) \), with \( B = \omega_{hp}^{1/2}(2P_J/\pi)^{1/2} \exp(-2i\pi Z_2/\lambda_p) \), \( V = Z_1/Z_C + Z_2/Z_C[1 + (Z_1/Z_C)^2] \). Z_1 is the distance between the probe beam focus and the sample, Z_C is the confocal distance and Z_2 is the distance from the sample to the photodiode. \( \omega_{hp} \) and \( \lambda_p \) are the...
probe beam radius in the sample center and probe beam wavelength, respectively. In the expression (1), the phase $\Phi$ is the perturbation induced by thermal lens in the probe beam’s electric field, which is obtained by equation[1]

$$\Phi (r, t) = \left( \frac{2\pi}{\lambda_p} \right) \left( \frac{ds}{dT} \right) \int_0^L \left[ T(r, z, t) - T(0, z, t) \right] dz. \tag{2}$$

To calculate this phase, we need to describe the temperature profile in the sample when the excitation beam passes through it. After this step, we can calculate the phase, and the electric field amplitude, which is used to obtain the intensity of the probe beam center, using the following equation

$$I(t) = |U(Z_l + Z_s, t)|^2. \tag{3}$$

Thermal mirror[5-7] (TM) is an interesting technique to be used combined with TL. Both can be mounted using the same equipment, making it possible to determine thermal, optical and mechanical properties of samples. While TL monitors the transmitted probe signals, TM is based on the reflected probe signal on the first sample’s surface. In this case the probe beam phase shift induced by the temperature rise in the sample is

$$\Phi = \left( \frac{2\pi}{\lambda_p} \right) 2u_z(r, 0, t), \tag{4}$$

where $u_z(0, 0, t)$ is axial component of the displacement vector in the sample surface, which is obtained solving the thermo-elastic equation, in the quasi-static approximation[8],

$$(1 - 2\nu) \nabla^2 u + \nabla (u \cdot \nabla) = 2(1 + \nu) \alpha \nabla T(r, z, t), \tag{5}$$

in which the temperature dependency appears. In both techniques, the theoretical development starts looking for a temperature rise description.

2. Temperature profile and probe beam intensity

The temperature profile of a sample, reached by a Gaussian beam, is obtained solving the following diffusion equation

$$\frac{\partial}{\partial t} T(r, z, t) - \frac{k}{c\rho} \nabla^2 T(r, z, t) = Q(r, z). \tag{6}$$

The source profile for a Gaussian laser beam illumination is given by

$$Q(r, z) = Q_0 \exp \left( -2r^2 / \omega_{0z}^2 \right) Q(z), \tag{7}$$

with $Q_0=2P_cA_c/\phi(\pi\omega_0^2\omega_{0z})$, in which $A_c$ is the optical absorption coefficient at the excitation beam wavelength, $P_c$ is the excitation beam power, $\rho$ is the mass density, $c$ is the specific heat, and $\omega_{0z}$ is the excitation beam radius. $\phi=1 - \eta <\lambda_m>$, in which $\lambda_m$ is the excitation beam wavelength, $<\lambda_m>$ is the average wavelength of the fluorescence emission, and $\eta$ is the fluorescence quantum efficiency. For the case that all the absorbed energy is totally converted into heat, $\eta=0$. Let us consider a sample with thickness $L$, and that in its surfaces the heat flux follows the condition[9]

$$\frac{\partial T(r, z, t)}{\partial z} \bigg|_{z=0} = h T(r, z, t), \tag{8}$$

where $h=k/k_s$, with $k_s$ being the thermal conductivity of the surround and $b_s$ a characteristic distance, where the temperature rise in the air is zero. The solution of this problem is[10]

$$T_{3D}(r, z, t) = \frac{P_c A_c \phi}{\pi T c K} \sum_{n=1}^{\infty} \left[ \frac{\alpha_n \cos(\alpha_n z) + h \sin(\alpha_n z)}{L(\alpha_n^2 + h^2) + 2h} \right] F_{\alpha_n} \int_0^{2\pi/\omega_{0z}} e^{-2\pi^2/\omega_{0z}^2} d\tau, \tag{9}$$

in which $\alpha$ represents the positive roots of $\tan(\alpha L)=2ah/(\alpha^2-h^2)$ and
The characteristic time of the experiment \( t_c = \frac{w_0^2 e}{4D} \).

To develop the TL model, the authors\[1,2\] made some approximations that simplify the solution of equation (6). The first one is that there is no heat flux on sample surfaces, what is verified as a good approximation because the experiment takes a short time, and usually the thermal conductivity of the sample is bigger than the environment conductivity. Another approximation used is that the heat source is constant through the sample thickness. Considering the Beer’s law, \( Q(z) = \exp(-A \varepsilon z) \), we have this condition when the product \( A \varepsilon L \ll 1 \), so the Beer’s law in the diffusion equation becomes \( \exp(-A \varepsilon z) \), and the solution is independent of \( z \).

Making these considerations the problem becomes bi-dimensional, and the solution to describe the temperature profile is\[1\]

\[
T_{2D}(r,t) = P_s \phi A_r / (4 \pi K) \left[ Ei\left(-2r^2 / \omega_0^2\right) - Ei\left(-2r^2t_c / \left[\omega_0^2 (2t_c + t_c)\right]\right)\right].
\]

To eliminate the restriction on the optical absorption, we investigated the following approximation

\[
T_{approx}(r,z,t) \approx e^{-A \varepsilon} T_{2D}(r,t),
\]

in which the Beer’s law was introduced ad hoc. With this, we are using a function that describes the heat source decay inside the sample to describe the temperature rise decay. In figure (2), we compare the temperature profile suggested above with the 3D solution.

\[\text{Figure 1. (a)} \text{ Temperature profile in } r=0, \text{ with the 3D solution (solid symbols) and the approximation (open symbols), with } b_s=0.5 \text{mm, } k_s=0.03 \text{W/mK, } t=100t_c, \phi=1. \text{ (b)} \text{ Phase shift calculated using 3D solution (solid symbols) and the approximation (open symbols) for } A_s=10 \text{cm}^{-1}.\]

The figure (1.a) shows that the correction produces a different description of the temperature profile, containing errors mainly near the sample surfaces. On the left the approximation produces higher temperatures, and on the right lowers ones. However, the thermal lens signal depends on an integral on temperature from \( z=0 \) until \( z=L \); in other words, it depends on the area under the curves in figure (1.a). Looking at this, we can see that the areas are approximately equal. Using the approximation (12), instead of the 3D solution (9), to calculate the phase, we add a quantity in the region of the first surface and take off about the same quantity in the rest of the thickness, in a way that the error on the phase is small. In figure (1.b), we show the phase calculated using both temperature profiles. Calculating the intensity using the approximation (12), we obtain the expression...
in which \( m = \omega T_{1P}/\omega_0 \) is the degree of mode mismatching of the probe and the excitation beams in the sample, and \( \theta_{TL} = P_e A_e L (ds/dT) \phi/(K \lambda_0) \). In this expression for intensity, comparing with the original expression obtained by Shen[1-2], there is only an additional term, written in brackets. In the low absorption limit this term becomes equal to one and we recover the original expression.

The TM model was already developed for high absorbing materials[6,7]. Taking the Beer's law, the approximations of null flux and semi-infinity thickness into account, the resulting equation for the phase shift and the TM intensity signal is given [6,7], with the parameter defined by

\[
\theta_{TM} = P_e A_e \alpha \nu \phi / (K \lambda_0).
\]  

3. Results and discussions

Using TM and TL concurrently, we have an alternative method to determine \((ds/dT)\) from the ratio of \( \theta_{TL} \) by \( \theta_{TM} \),

\[
\left( \frac{ds}{dT} \right) = \frac{\alpha \nu}{L} \frac{\theta_{TL}}{\theta_{TM}}.
\]

Our results are compared with the values measured by an interferometric technique[10], where the coefficient is obtained using the relation[11]

\[
\left( \frac{ds}{dT} \right) = \left( \frac{ds}{dT} \right)_{INT} + \alpha \nu \phi (n v - 1 - \nu),
\]

in which \((ds/dT)_{INT}\) is the temperature coefficient of the optical path length in the interferometry, where unlike TL there isn't surface bulging and stress. Therefore, we perform TL and TM in a calcium aluminosilicate glass doped with 4% of \( \text{Er}_2\text{O}_3 \). To calculate \((ds/dT)\), we use[11,12] \( n = 1.629 \), \((ds/dT)_{INT} = 16.1 \times 10^{-6} \text{K}^{-1} \), \( \nu = 0.29 \) and \( \alpha = 6.7 \times 10^{-6} \text{K}^{-1} \), resulting in \((ds/dT) = 10.6 \times 10^{-6} \text{K}^{-1} \) with an error estimated of \( 1 \times 10^{-6} \text{K}^{-1} \).

The results of a TL and TM measurement are showed in figure (2), where we also compare the fits with expression (13) and the original expression. We find that the correction for high absorbing materials increases \( \theta_{TL} \) in about 15% for this sample, and \( t_e \) is exactly the same with or without the correction.
Results

Using the original model (Ae=0)

θL=-0.321615

tc=1.748 ms

Using the correction

θL=-0.37983

tc=1.748 ms

Intensity (V)

Time (ms)

Experimental data

Best fit

Figure 2. (a) Experimental data and the best fit of thermal lens signal with equation. (b) Experimental data and the best fit of thermal mirror signal. The optical absorption coefficient of this sample is Ae=2.5 cm⁻¹, for an excitation wavelength of 488nm and the thickness is L=0.137 cm (m=37.9, V=4.29, ae₀e=56μm and Pe=221 mW).

Calculating (ds/dT) using the original TL model we obtain 8.3x10⁻⁶K⁻¹; while, including the high absorbing correction, the value obtained was 9.9x10⁻⁶K⁻¹. Comparing these results with the value obtained by interferometry, we can conclude that, employing the correction (9), the results are in good agreement. So, both TL and TM models are able to extract quantitative information about high absorbing materials’ properties, including (ds/dT).

4. Conclusions

We conclude that, by introducing a simple correction, TL technique can be extended to be applied in high absorbing materials, with good accuracy. Some authors have already used a correction[4,5,12,13] that is equivalent to the one proposed in this work in equation (12). However, here we have introduced a theoretical interpretation and performed experimental measurements that show the robustness of this correction.

The authors are thankful to Brazilian agency CNPq for partial financial support.

References

[1] J. Shen, R. D. Lowe, and R.D. Snook, Chem. Phys. 165 (1992) 385. D. Snook and R. D. Lowe, Analyst 120, 2051-2068 (1995).
[2] M. L. Baesso, J. Shen, R. D. Snook, Chemical Physics Letters 197 (1992), 255.
[3] S. M. Lima, E. S. Bannwart, R. G. Oliveira, L. H. C. Andrade, P. V. Del Re, N. Jorge, F. Pedrochi, R. Constantini, A. N. Medina, M. L. Baesso. European physical journal-special topics 153 (2008) 531.
[4] M.R.R. Gesualdi, C. Jacinto, T. Catunda, M. Muramatsu, V. Pilla. Appl Phys B 93 (2008) 879.
[5] N. G. C. Astrath, L. C. Malacarne, P. R. B. Pedreira, A. C. Bento, M. L. Baesso, and J. Shen, Appl Phys. Lett. 91 (2007) 191908.
[6] L. C. Malacarne, F. Sato, P. R. B. Pedreira, A. C. Bento, R. S. Mendes, M. L. Baesso, N. G. C. Astrath and J. Shen. Applied physics letters 92, (2008) 131903
[7] F. Sato, L. C. Malacarne, P. R. B. Pedreira, M. P. Belancon, R. S. Mendes, M. L. Baesso, N. G. C. Astrath and J. Shen, Journal of applied physics 104, (2008) 053520.
[8] J. Cheng, L. Wu, and S. Zhang, J. Appl. Phys. 76 (1994) 716.
[9] A. Steimacher, A.N. Medina, A.C. Bento, J.H. Rohling, M.L. Baesso, V.C.S. Reynoso, S.M. Lima, M.N. Petrovich and D.W. Hewak. Journal of Non-Crystalline Solids 348 (2004) 240.
[10] J. Shen, M. L. Baesso, and R. D. Snook, J. Appl. Phys. 75 (1993), 3738.
[11] N. G. C. Astrath, A. Steimacher, J. H. Rohling, A. N. Medina, A. C. Bento, M. L. Baesso, C. Jacinto, T. Catunda, S. M. Lima and B. Karthikeyan. Optics Express 16 (2008) 21248.
[12] A. Steimacher, N.G.C. Astrath, A. Novatski, F. Pedrochi, A.C. Bento, M.L. Baesso and A.N. Medina. Journal of Non-Crystalline Solids 352 (2006) 3613.
[13] N. G. C. Astrath, J. H. Rohling, A. N. Medina, A. C. Bento, M. L. Baesso, C. Jacinto, T. Catunda, S. M. Lima, F. G. Gandra, M. J. V. Bell and V. Anjos, Phys. Rev. B. 71 (2005) 214202.