

Numerical modeling of thermal refraction in liquids in the transient regime

Dmitriy I. Kovsh, David J. Hagan and Eric W. Van Stryland

*School of Optics/CREOL, University of Central Florida
4000 Central Florida Blvd., Orlando, Florida
kovsh@creol.ucf.edu*

Abstract: We present the results of modeling of nanosecond pulse propagation in optically absorbing liquid media. Acoustic and electromagnetic wave equations must be solved simultaneously to model refractive index changes due to thermal expansion and/or electrostriction, which are highly transient phenomena on a nanosecond time scale. Although we consider situations with cylindrical symmetry and where the paraxial approximation is valid, this is still a computation-intensive problem, as beam propagation through optically thick media must be modeled. We compare the full solution of the acoustic wave equation with the approximation of instantaneous expansion (steady-state solution) and hence determine the regimes of validity of this approximation. We also find that the refractive index change obtained from the photo-acoustic equation overshoots its steady-state value once the ratio between the pulsewidth and the acoustic transit time exceeds a factor of unity.

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OSIC codes: (190.4870) Optically induced thermo-optical effects; (190.5940) Self-action effects; (190.5530) Pulse propagation and solitons; Optical limiting.

References and links

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1. Introduction

Thermally induced refractive index changes caused by absorption of light in a material have been intensively investigated both experimentally [1,2,3] and theoretically [4-10] for various time scales of the input laser pulses. Most of these studies addressed the problem of thermal lensing produced inside the material on microsecond and longer (up to CW) time scales [4-6]. The shape of the thermal lens and its impact on the near and far field spatial distributions of the laser beam were analytically estimated for these time scales in references [9-12]. Several authors have considered shorter (nanosecond) pulses to be a source for the refractive index change. In this case the effect of thermally induced index changes can be highly transient for focused beams. The theoretical analysis of such a situation was offered in Refs. [7,8], where the authors derived the coupled hydrodynamic equations defining the local changes in density, pressure and temperature due to the presence of laser radiation. Under certain approximations on the laser beam temporal and spatial profiles (namely Gaussian) the solutions for those equations were obtained. Detection of the sound waves generated by the rapid thermal expansion in liquids is the basis for photo-acoustic spectroscopy that is used to measure weak optical absorption. In Refs. [13,14] the results of experiments and analysis of this effect were given. The strong beam self-action due to the thermal lens formed on a nanosecond scale was also considered by Brochard et. al. [15]. The simplified model describing such self-action and comparison to the steady state case was offered and tested against the results of Z-scan and pump-probe experiments. The main difficulty one experiences when trying to analyze thermal

refraction in the transient regime is the fact that both wave equations for electromagnetic laser field and acoustic equation must be solved simultaneously. Unless certain simplifications are made it is an extremely computationally intense task. However, with the recent increase of computing power of modern PCs and workstations it becomes possible to model the dynamics of the nonlinear media response to pulsed laser light including spatial beam size changes within the medium from external focusing as well as nonlinear self action.

The motivation for this research is our need to model the behavior of passive optical limiters for nanosecond input laser pulses. An optical limiter is a device that is highly transparent for incident light of low intensity, while it strongly attenuates input light of high intensity, thus protecting sensitive optical components from laser-induced damage (see, for example, Ref. [16]). Passive limiters perform this task without any additional switches or controlling circuits, but instead use the nonlinear optical response of certain materials. One of the common designs of passive optical limiters uses materials exhibiting strong excited-state absorption to attenuate high-energy input signals [17-19]. These are often referred to as reverse saturable absorber (RSA) materials. The current state-of-the-art in nonlinear optical materials requires that nonlinear elements be placed at or near a focal plane in the imaging system in order to have sufficient energy density to induce a large enough nonlinear optical effect. Designs for optimized limiters using RSA materials require a distribution of the nonlinear materials over distances of many diffraction lengths along the propagation path. Hence the beam size through the nonlinear materials may vary by more than one order of magnitude (irradiance varying by $> 10^2$), requiring numerical beam propagation algorithms to model limiter response. Modeling the nonlinear absorption itself is a nontrivial task; for instance, a system of rate equations must be solved to compute the excited-state absorption. Inclusion of thermally induced nonlinear index change makes the model even more complicated.

Upon heating of the medium by absorption of the laser pulse, the medium begins to expand. This expansion propagates outward as an acoustic disturbance. The method of analyzing this problem depends very much on the relative time scales for the acoustic expansion and the optical pulse width. If the laser pulse is much longer than the acoustic transit time (the time for the optical expansion to propagate across the laser beam) the thermal lens is essentially instantaneous. The refractive index change in this case is linearly proportional to the temperature change and hence the pulse fluence (if the medium exhibits only linear absorption), which greatly simplifies the analysis. This can be used to measure the thermal optical properties of the material [20-22] or to detect nonlinear absorption [13]. However, if the laser beam size is larger than or comparable to the acoustic transit time, then only later portions of the laser pulse can experience index changes induced by earlier portions. Such a situation has been referred to as the transient regime [15].

In this paper we investigate the dynamics of the refractive index change of the liquid absorptive media due to thermal expansion in this transient regime. We compute the refractive index change as a solution to the acoustic wave equation and compare the results to the instantaneous thermal lensing approximation. The comparisons of the results obtained with and without this approximation allow us to conclude the range of its validity.

In the next section the basic theory of the photo-acoustic effect in liquid media is presented, including the thermal lensing approximation. In later sections, we introduce the numerical algorithm used to solve the equations describing the propagation of an optical pulse inside thick nonlinear media and then show the results of this modeling.

2. Thermally-induced index change

While propagating through absorptive liquid media, a laser pulse induces temperature and density gradients that change the refractive index profile. This process is often called the

thermal lensing effect, since the change in refractive index approximately follows the spatial beam profile, thus forming a lens within the medium. For various time scales of the input pulse, the thermal lensing has different manifestations. As stated above, if the pulse width is longer than a few microseconds, density changes occurring via acoustic propagation may be considered instantaneous for beams having radii of one hundred microns or less (sound velocity $\cong 1\sim 2 \mu\text{m/ns}$). In this case, the density changes (and hence refractive index changes, Δn) follow the temperature change, ΔT . Hence, it is the shape of the beam, coupled with thermal diffusion, which dictates the temperature gradient. Heating the material in this case can be described by the following equation [23]:

$$\rho c_p \frac{\partial T}{\partial t} - \kappa \nabla^2 T = Q, \quad (1.1)$$

where ρ is the density of the medium, c_p is the specific heat at constant pressure and κ the thermal conductivity. The source for the temperature change is Q , the absorbed power of the laser beam per unit volume. In the simplest case where linear absorption is the primary absorption mechanism Q is the product of the beam Irradiance, I , and linear absorption coefficient, α_l ($Q = \alpha_l I$). The refractive index change is, in general, a function of temperature and density changes inside the material [14]:

$$\Delta n = \left(\frac{\partial n}{\partial \rho} \right)_T \Delta \rho + \left(\frac{\partial n}{\partial T} \right)_\rho \Delta T. \quad (1.2)$$

Here, $(\partial n / \partial \rho)_T$ describes the index changes due to thermal expansion or electrostriction, while $(\partial n / \partial T)_\rho$ is due to other temperature-dependent changes in index, which are of less importance in liquids [24]. On a time scale much longer than the acoustic transit time, τ_{ac} (here defined as w/C_s where w is the $\text{HW1/e}^2\text{M}$ beam size and C_s is the sound velocity), refractive index changes become linearly proportional to the change in temperature with the coefficient of proportionality called the thermo-optic coefficient. This occurs due to the fact that for later times $\Delta \rho = (\partial \rho / \partial T)_p \Delta T$ and $(\partial \rho / \partial T)_p$ becomes constant. In this regime the refractive index change is determined by thermal diffusion (Eq. 1.1). (Generally, the density changes due to electrostriction must be included, but in this work we concentrate on the thermally induced index change, as discussed later). The same long-term effect may also be observed for shorter pulses if the input laser has a high repetition rate (e.g. a modelocked laser pulse train). In this case the source term in Eq. (1.1) becomes a series of impulses causing local heating which subsequently undergoes thermal diffusion.

However, for the single pulses of shorter duration (nanosecond time scale), the refractive index changes with the acoustic expansion of the medium generated by local heating (absorptive mode) or by its compression due to the electromagnetic field of the laser beam (electrostrictive mode) [13,14]. Hence, although the refractive index changes linearly with the density, it does not follow the spatial variation of the temperature. If the pulsewidth is in the picosecond regime, the acoustic waves do not have time to propagate, and therefore the density and index cannot change significantly resulting in no lensing effect during the short pulse.

Refractive index variations due to thermal expansion in solid media also occur, but $(\partial \rho / \partial T)_p$ is usually an order of magnitude smaller than in liquids so the effect is often masked by the electrostrictive effect [14] or other sources for $(\partial n / \partial T)_\rho$ [24]. In liquids the

effect of electrostriction is generally one or several orders of magnitude smaller than the thermal expansion (as long as the absorption of the liquid is significant), and therefore can often be neglected. From now on we concentrate on the index changes in liquids that occur on the nanosecond scale. This analysis is appropriate for the heating caused by, for example, single nanosecond pulses generated by a Q-switched Nd:YAG laser.

Combining the three main equations of hydrodynamics: continuity, Navier-Stokes and energy transport equations [23], and assuming small changes of temperature, ΔT , density, $\Delta\rho$, and pressure, Δp , we obtain the acoustic equation defining the density perturbation of the material [13-15]:

$$\frac{\partial}{\partial t} \left[\frac{\partial^2(\Delta\rho)}{\partial t^2} - C_s^2 \nabla^2(\Delta\rho) \right] = \frac{C_s^2 \beta}{c_p} \nabla^2(\alpha_L I) - \frac{\gamma^e}{2nc} \frac{\partial}{\partial t} \nabla^2 I. \quad (1.3)$$

Here the liquid is assumed non-viscous with thermal expansion coefficient $\beta = -(1/V)(\partial V/\partial T)_p$, specific heat c_p and index of refraction n . $\gamma^e = \rho(\partial n^2/\partial \rho)_T$ is the electrostrictive coupling constant, which can be estimated using the Lorentz-Lorenz law as $\gamma^e = (n^2-1)(n^2+2)/3$. The two source terms on the right-hand side of the equation represent two phenomenon discussed earlier – absorption of light energy and the electrostrictive effect. We further assume that the light energy absorbed is converted into kinetic energy of the molecules on a time scale shorter than the pulse itself. This is usually true for relaxation and vibration transitions [14]. The formalism developed here is being used to model refractive index variations in the highly absorptive liquid solutions used for optical limiters. For this type of liquid media the impact of the electrostrictive effect (second source term in Eq. 1.3) can be ignored compared to the density changes due to local heating of the media by the laser pulse. The refractive index is related to the density and temperature changes as given in Eq. (1.2) where $(\partial n/\partial \rho)_T = \gamma^e/(2n\rho)$. Thus, the wave equation for the index change due to the thermal expansion can be written as

$$\frac{\partial^2(\Delta n)}{\partial t^2} - C_s^2 \nabla^2(\Delta n) = \frac{\gamma^e}{2n\rho} \frac{\beta C_s^2}{c_p} \int_{-\infty}^t \nabla^2(\alpha_L I(r, t')) dt'. \quad (1.4)$$

The source term in Eq. (1.4) can be expressed in terms of the temperature change using Eq. (1.1), neglecting diffusion, which does not occur on the nanosecond time scale:

$$\Delta T(r, t) = \frac{1}{\rho c_p} \int_{-\infty}^t \alpha_L I(r, t') dt'. \quad (1.5)$$

Hence we obtain the final form of the acoustic wave equation for the refractive index change used in the numerical analysis:

$$\frac{\partial^2(\Delta n)}{\partial t^2} - C_s^2 \nabla^2(\Delta n) = \frac{\gamma^e \beta C_s^2}{2n} \nabla^2(\Delta T). \quad (1.6)$$

If we have a few nanosecond duration pulse focused to a spot diameter of 10~20 μm , for typical values of the sound velocity in liquids (1~2 $\mu m/ns$) the acoustic wave generated by the front of the pulse traverses the beam and creates an index change affecting the tail end of the pulse. If the pulse width, τ_p , is longer than the acoustic transit time $\tau_p > w/C_s$, we simplify the

numerical modeling of the photo-acoustic effect by parameterizing the index change close to the propagation axis [7] as:

$$\Delta n \cong \left(\frac{dn}{dT} \right) \Delta T, \quad (1.7)$$

where $(dn/dT) = \gamma \beta / (2n)$ is the thermo-optic coefficient. Equation (1.7) is a commonly used approximation called the thermal lensing effect [2], which is usually applied for much longer time scales (microseconds up to CW) where the index change is governed by thermal diffusion. As we show below the actual thermal lens introduced by acoustic density perturbations in the material is a very aberrated replica of the one in Eq. (1.7), since this approximation only can be made for near axis index changes because it ignores the acoustic wave propagation. However, such an approximation is attractive because we can significantly reduce the computational time required to numerically solve the acoustic wave equation for each time slice of the pulse. In fact, there are experimental results in the literature where (dn/dT) was calculated in this approximation for different liquids [20-22]. As shown below, the last approximation requires a certain care to apply and may lead to erroneous results.

3. Beam Propagation Algorithm

The propagation of light through an optical medium can be described by the solution to the vector wave equation [25]:

$$\nabla \times \nabla \times \vec{E}(\vec{r}, t) + \frac{1}{c^2} \frac{\partial^2 \vec{E}(\vec{r}, t)}{\partial t^2} = -\mu_0 \frac{\partial^2 \vec{P}(\vec{r}, t)}{\partial t^2}, \quad (2.1)$$

where $\vec{E}(r_{\perp}, z, t)$ and $\vec{P}(r_{\perp}, z, t)$ are the electric field and the medium polarization. Making the Slowly Varying Envelope Approximation and assuming that group velocity dispersion can be neglected, this equation can be greatly simplified; and for slow (i.e. large F-number) systems rewritten in a scalar paraxial form:

$$2jk \frac{\partial \Psi(r_{\perp}, z, t)}{\partial z} = \nabla_{\perp}^2 \Psi(r_{\perp}, z, t) + (k_0^2 \chi_{NL}(r_{\perp}, z, t) - jk\alpha_L) \Psi(r_{\perp}, z, t), \quad (2.2)$$

where we used the expression for the electric field $E(r_{\perp}, z, t) = \Psi(r_{\perp}, z, t) e^{j\alpha z - jkz}$. Here ∇_{\perp}^2 and r_{\perp} denote the transverse Laplace operator and radial spatial coordinate, while $k = n_0 k_0 = n_0 \omega / c$ is the wave vector in the media with linear index of refraction $n_0 = \sqrt{1 + \text{Re}\{\chi_L\}}$ and linear absorption coefficient $\alpha_L = -(k_0/n_0) \cdot \text{Im}\{\chi_L\}$. $\chi_{NL}(r_{\perp}, z, t)$ is the nonlinear susceptibility of the material, which may consist of instantaneous and cumulative parts:

$$\chi_{NL}(r_{\perp}, z, t) = \chi_{NL}^{ins}(r_{\perp}, z) + \chi_{NL}^{cum}(r_{\perp}, z, t). \quad (2.3)$$

The nonlinear susceptibility is related to the nonlinear refractive index change, Δn , and the nonlinear absorption, α_{NL} , of the material as:

$$\text{Re}\{\chi_{NL}\} = 2n_0 \Delta n \quad (2.4)$$

$$\text{Im}\{\chi_{NL}\} = -\frac{n_0}{k_0} \alpha = -\frac{n_0}{k_0} (\alpha_L + \alpha_{NL}). \quad (2.5)$$

Due to the fact that there is no explicit time dependence in Equation (2.2) (although the field amplitude as well as the nonlinear susceptibility are, in general, functions of time), the modeling of the laser pulse propagation in the nonlinear medium can be split into two separate numerical tasks. The first one is dividing the pulse into a number of time slices and propagating each slice as a CW beam. The second one is computing and storing the cumulative part of the nonlinear susceptibility induced by each slice. Therefore, the solution to the original time-dependent wave equation (2.2) becomes a CW propagation problem. To reduce the computation time and storage memory we assume cylindrical symmetry, thus reducing the spatial 3D problem to 2D. We then implement the so-called Beam Propagation Method (BPM), which requires recomputing the transverse field distribution along the direction of propagation, z , using the formal solution to Eq. (2.2). The unitary, finite difference Crank-Nicholson scheme was chosen [26] for its high efficiency when dealing with propagation through distances of more than a few Rayleigh ranges [27,28].

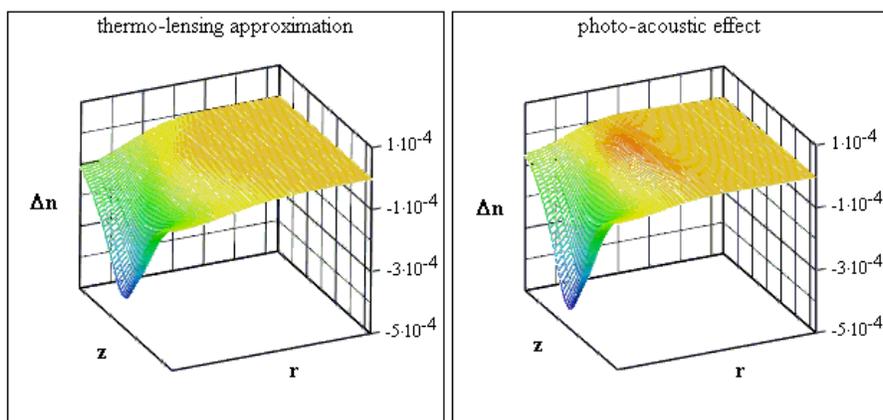


Fig. 1. Spatial distribution of the thermally induced refractive index change ($\tau_p = 4$ ns, $w_0 = 6$ μm , $T_L = 80\%$, $L = 1$ mm, $E_{IN} = 5$ μJ).

4. Results and discussion

To demonstrate our model, we choose a situation where a 4-nanosecond (HW1/eM of irradiance) pulse with a Gaussian spatial and temporal profile propagates through an aqueous solution of nigrosine dye. Nigrosine is chosen since it shows very little nonlinear response other than thermal refraction from linear absorption for nanosecond input pulses. The thermo-optic coefficient of water is equal to $-5.7 \times 10^{-4} \text{ K}^{-1}$ [15]. To study the validity of the approximation of Eq. (1.7), we first choose cases where the linear-optics beam waist inside the sample w_0 is (i) 6 μm and then (ii) 30 μm (HW1/e²M of irradiance). The acoustic transit times in cases (i) and (ii) are $\tau_{ac} = 4$ ns and 20 ns respectively. The sample is chosen to be 1 mm thick in each case. Figure 1 shows an animation of the dynamics of the thermally induced refraction index change in case (i) as the laser pulse passes through the sample. We first include modeling of the acoustic equation (1.6) coupled with propagation and then apply the approximation (1.7), i.e. thermal lensing. Although the approximation (1.7) ignores the small index disturbances on the wings of the pulse, which are due to the acoustic wave propagation,

it correctly predicts the changes of the refractive index close to the axis where most of the beam energy is concentrated. Moreover, the thickness of the sample was chosen to be several Rayleigh ranges, thus complicating the modeling since beam diffraction is included along with the effect of nonlinear self-action. In order to compare the results obtained with and without the approximation (1.7) we also look at the near and far field profiles of the beam. As seen in Fig. 2 the near and far field radial fluence distributions remain the same if computed with and without the approximation (1.7). This is as expected, since the pulse width in case (i) is equal to the acoustic transit time.

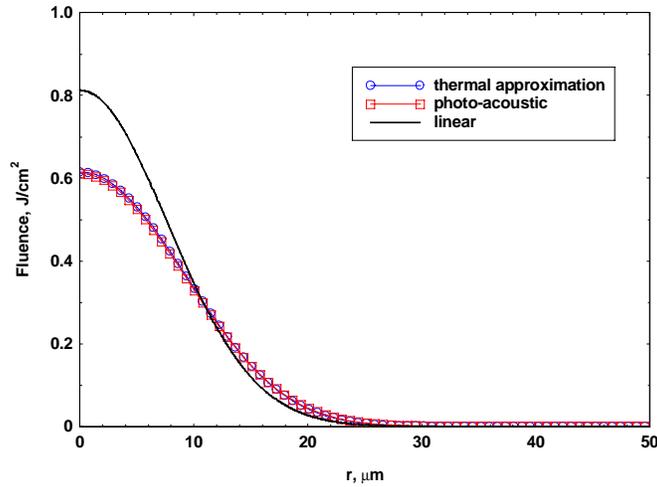


Fig. 2(a). Radial fluence distribution on the back surface of the sample (near field).

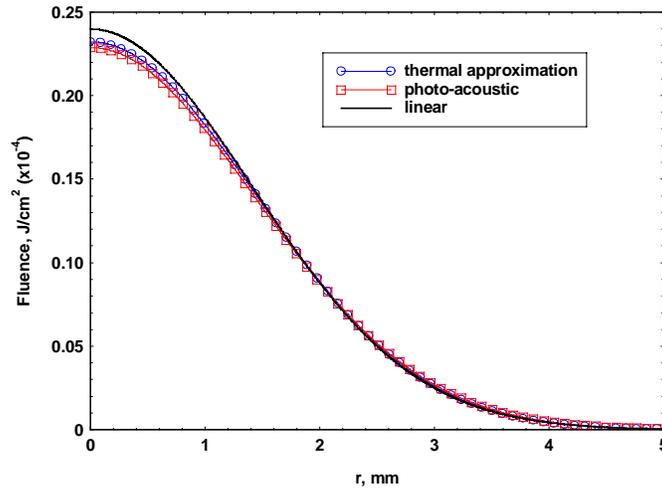


Fig. 2(b). Radial fluence distribution on the detector (far field).

The situation significantly changes once we increase the size of the focused beam in case (ii) to $30\ \mu\text{m}$, leaving the pulsewidth the same (the energy was also increased by a factor of 25 to keep the same value of maximum irradiance at the focal plane and fluence). Here, the acoustic transit time is longer than the pulse width. As shown in the animation of Fig. 3, the acoustic wave does not have enough time to propagate and, therefore, Eq. (1.7) overestimates the effective index change. This can be also seen from the differences in the near and far field radial fluence profiles shown in Fig. 4. It is important to notice that qualitatively the index change developed as a result of the photo-acoustic effect reaches the same value as the one predicted by Eq. (1.7), but it does so for later times, thus diffracting less energetic parts of the pulse (see Fig. 3).

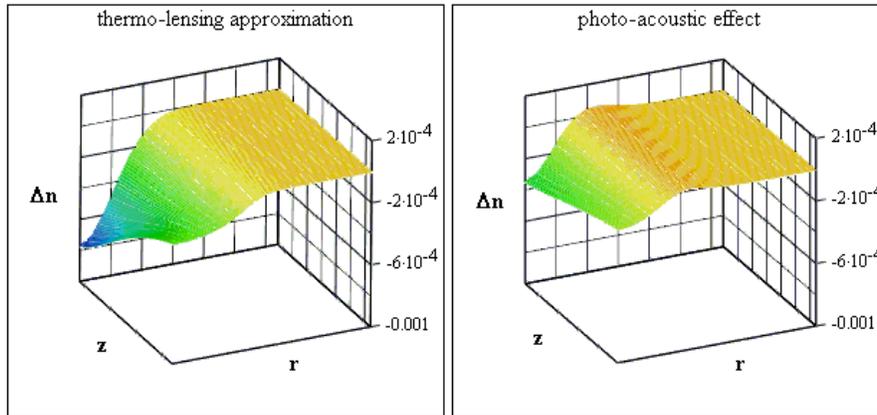


Fig. 3. Spatial distribution of the thermally induced refractive index change ($\tau_p = 4\ \text{ns}$, $w_0 = 30\ \mu\text{m}$, $T_L = 80\%$, $L = 1\ \text{mm}$, $E_N = 125\ \mu\text{J}$).

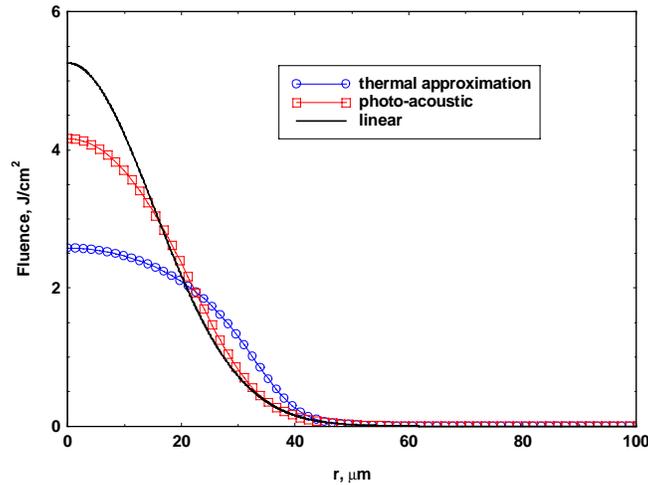


Fig. 4(a). Radial fluence distribution on the back surface of the sample (near field).

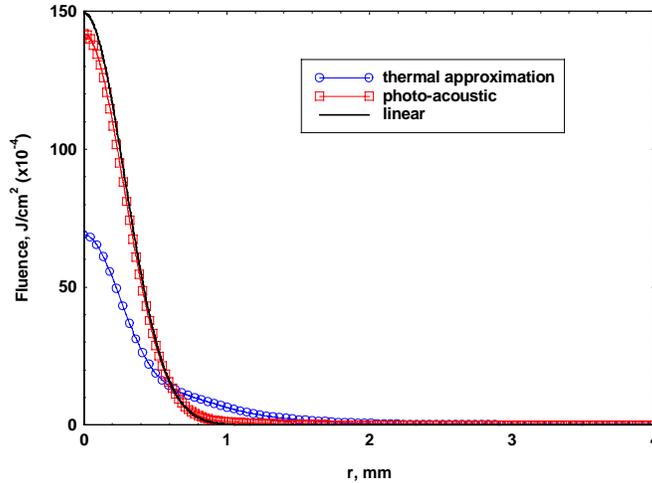


Fig. 4(b). Radial fluence distribution on the detector (far field).

The above results indicate that if the laser pulse width is comparable or larger than the acoustic transient time, the approximation (1.7) may be used, but it will predict incorrect results otherwise. In order to analyze the range of validity in more detail, we model the closed-aperture Z-scan experiment [29] for thermally induced refraction in the aqueous solution of nigrosine. Figures 5 and 6 show the calculated Z-scan signals using both the full solution of the acoustic equation (1.6) and the approximation (1.7). To speed up calculations, the thickness of the sample was chosen to be 200 microns so that the “thin-sample” Z-scan formalism commonly used to analyze experimental data is valid [29]. Choosing the pulse width to be $\tau_p = 10$ ns (HW1/eM) and beam waists of $6 \mu\text{m}$ and $30 \mu\text{m}$, we define the ratio between the pulsewidth and transit acoustic time (τ_p/τ_{ac}) to be 2.5 (Fig. 5) and 0.5 (Fig. 6) respectively. Although the use of thermal approximation results in a good agreement for the former case it clearly produces the erroneous result for the later.

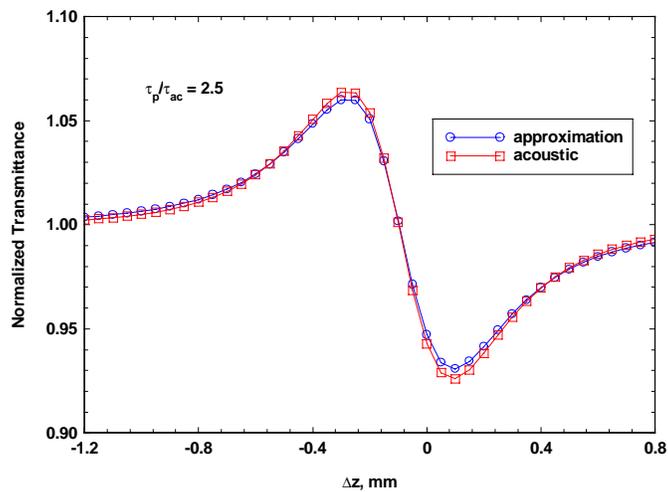


Fig. 5. Closed-aperture Z-scan of nigrosine solution in water ($\tau_p = 10$ ns, $w_0 = 6 \mu\text{m}$, $T_L = 90\%$, $L = 200 \mu\text{m}$, $E_{IN} = 2 \mu\text{J}$).

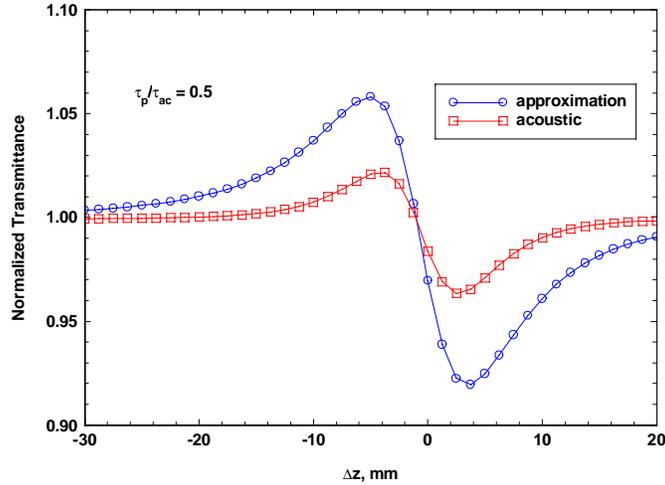


Fig. 6. Closed-aperture Z-scan of nigrosine solution in water ($\tau_p = 10$ ns, $w_0 = 30$ μm , $T_L = 90\%$, $L = 200$ μm , $E_{IN} = 50$ μJ).

In order to explore the range of validity of the approximation (1.7) even further, we model the closed-aperture Z-scan experiment for different values of the parameter τ_p/τ_{ac} comparing the peak-to-valley change in transmittance ΔT_{p-v} obtained with and without this approximation. For two values of the pulse width ($\tau_p = 4$ and 8 ns HW1/eM), we varied τ_p/τ_{ac} by changing the sound velocity. Changing τ_p/τ_{ac} in this way is convenient as it allows us to keep the geometry of the experiment the same. For both pulse widths the value of on-axis fluence at focus and therefore the closed-aperture Z-scan signal are the same [29]. Both sets of parameters produce the curve shown in Fig. 7, indicating that the behavior is determined solely by τ_p/τ_{ac} . This dependence remains unchanged even after we varied the value of input energy to have ΔT_{p-v} in the range of 4-25% producing the same curve (Fig. 7). The estimation of the thermally induced refractive index change using Eq. (1.7) produces accurate results for values of the ratio between the HW1/eM pulse width and acoustic transit time larger than unity (or > 1.6 if FWHM value of the pulse width is used). Hence, $\tau_p/\tau_{ac} = 1$ defines the limit of validity of Eq. (1.7) beyond which the acoustic wave propagation must be included in the analysis. Figure 8 shows the on-axis distribution of the thermally induced refractive index change as a function of time. Note that with the increase of the parameter τ_p/τ_{ac} the value of the index change (and consequently Z-scan signal in Fig. 7) first reaches the value predicted by the steady state solution (1.7) and then surpasses this value (Fig. 8). This is because the index change initially overshoots the steady state solution of this equation. This can also be seen in Fig. 3. Although the solution of Eq. (1.6) reaches the steady state value of Δn for later times, it gives a larger index change for the central parts of the pulse where the irradiance is the largest. This effect was mentioned in the Ref. [15] although no explanation was given.

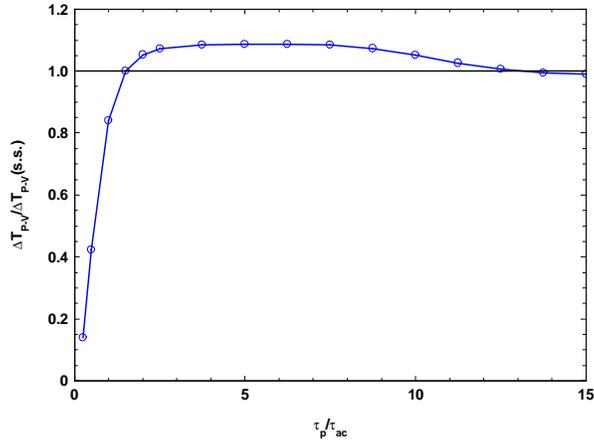


Figure 7. Sensitivity ($\Delta T_{P,V}$) of the closed-aperture Z-scan as a function of ratio between pulse width, τ_p , and acoustic transit time $\tau_{ac} = w_0/C_S$. $\Delta T_{P,V}$ is normalized to the value obtained for the steady state solution (s.s.).

Hence, considerable care must be exercised in applying the thermal lensing approximation to assure that significant errors do not occur. In particular, one has to be careful conducting experiments defining the thermal optical properties of the materials (Refs. [20-22]) or their absorption characteristics (Refs. [13]). Also we see that when modeling the performance of a liquid based optical limiter on a nanosecond time scale [16,17] we can usually apply the thermal lensing approximation if the limiting element is placed near the focus (where the beam size is smaller than the characteristic length of the acoustic wave). However, if the limiter is located far from focus, where the beam size is large (e.g. as in tandem limiters [16]), the photo-acoustic equation (1.3) must be solved in order to correctly model the thermal refraction.

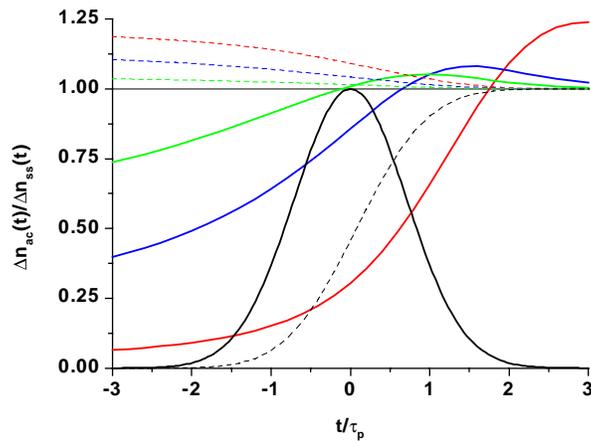


Figure 8. On-axis refractive index change computed as a solution to the acoustic wave equation, $\Delta n_{ac}(t)$, normalized to the steady state index distribution, $\Delta n_{ss}(t)$. Values of the parameter τ_p/τ_{ac} were chosen to be 0.5 (solid red), 1.5 (solid blue), 2.5 (solid green), 5.0 (dash red), 10 (dashed blue) and 15 (dashed green). The normalized $\Delta n_{ss}(t)$ (with negative sign) is shown with dashed black line. The normalized intensity distribution (solid black) is plotted to show the time scale of the index changes.

5. Conclusions

We presented analyses and results of computer modeling of the photo-acoustic effect induced by local heating of linear absorbing liquid media. The thermal lensing approximation is discussed and it can significantly simplify the modeling of nanosecond laser pulses propagating through such media. This is of particular importance when such phenomena are encountered in cases where numerically intensive beam propagation is required [28]. We investigate the range of validity of this approximation and find that for pulses having duration larger than the acoustic transient time across the beam ($\tau_p/\tau_{ac} > 1$), the thermally induced refractive index changes essentially instantaneously throughout the pulse. However, our results show that even in this case, the approximation may yield small (~10%) errors in estimating the index change due to an “overshoot” of the thermally induced density change. This knowledge helps us to understand the results of experiments performed to measure the thermal optical and absorptive characteristics of liquid materials.

Acknowledgments

This work was carried out under the support of the Office of Naval Research (grant number N00014-97-1-0936) and the Naval Air Warfare Center Joint Service Agile Program (contract number N00421-98-C-1327). The authors gratefully acknowledge useful discussions with T. Xia and M. Bolshtyansky.