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Thermal-lens model compared with the Sheik-Bahae formalism in interpreting Z-scan experiments on lyotropic liquid crystals

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We carried out Z-scan measurements on lyotropic liquid crystals and on lyotropic liquid crystals doped with ferrofluid. In these experiments, the sample is translated through the focal region of a focused Gaussian laser beam. The dependence of the far-field intensity on sample position due to intensity-dependent optical nonlinearities has been analyzed on the basis of the thermal-lens model of Gordon *et al.* and the Gaussian decomposition analysis of Sheik-Bahae *et al.* The thermal-lens model is nonlocal in space and time, whereas the Gaussian decomposition is predicated on a strictly local response. We compare the goodness of fit of the predictions of these models to experimental data, and we discuss limitations of these models in describing Z-scan experiments on systems with nonlocal response. © 2002 Optical Society of America

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1. INTRODUCTION

In 1989, Sheik-Bahae *et al.*[1,2] proposed an elegant method for determining the sign and amplitude of the nonlinear refractive index n_2 using the so-called Z-scan technique. This technique uses a single focused Gaussian laser beam propagating along the z axis. The sample is moved through the focal region of the beam, along the direction of beam propagation. The transmitted light intensity in the far field is measured as a function of sample position. The intensity-dependent optical nonlinearities that give rise to diffraction and absorption can be determined from the dependence of transmittance on sample position, or the Z-scan curve. This technique has been used in a wide variety of experiments to determine the nonlinear susceptibilities of solids, ordinary liquids,[2–7] and liquid crystals.[8–14]

Sheik-Bahae *et al.* used a novel application of Gaussian decomposition to analyze the effects of the nonlinear response of the sample on the far-field intensity. We refer to this method as the Sheik-Bahae formalism (SBF). An essential feature of the SBF in analyzing the transmittance is the assumption of a local interaction between the radiation field and the sample; it is assumed that the susceptibility is a function of only the local intensity. In general, however, a variety of mechanisms may contribute to the nonlinearity, some of which may be nonlocal. In nematic liquid crystals, for example, optical-field-induced reorientation of the director is nonlocal in space, as are processes in all systems where thermal diffusion plays a dominant role. The thermal-lens model[15–18] (TLM), which considerably predates the Z-scan technique, can be successfully used to interpret Z-scan experiments.[19] In this application, the sample is considered as a weakly absorbing medium, where energy absorbed from the laser beam is immediately converted into heat. The nonlinearity arises from the dependence of the refractive indices on temperature. Due to the diffusion of heat, the spatial temperature profile can differ significantly from the laser intensity profile; hence the mechanism is nonlocal.

Z-scan experiments probe the intensity-dependent nonlinear susceptibility. In the case of laser heating, this depends on the thermo-optic coefficient $\partial n/\partial T$, where n is the refractive index and T is the temperature, and on the thermal diffusivity. We therefore expect Z-scan experiments to furnish information on these quantities.

It is interesting to ask whether these two different approaches have a common limit. The aim of this paper is to discuss, from the theoretical point of view, the common limit of the different TLM and SBF models and to use a ferronematic lyotropic liquid crystal[20] as a working sample. This material is particularly useful for this purpose, since its absorption can be controlled by the ferrofluid doping.

We present a brief overview of SBF and TLM and discuss their salient features. We present experimental Z-scan results on lyotropic liquid crystals and compare these with model predictions.

2. SHEIK-BAHAE FORMALISM

We consider a medium with inversion symmetry illuminated by a monochromatic Gaussian laser beam, with electric field **E** and frequency ω. The nonlinear refractive index, n_2, and the nonlinear absorption coefficient, β, in a system whose response is local both in space and time, is defined as

$$n = n_0 + \frac{1}{2} n_2 E^2 = n_0 + \gamma I,$$

$$\alpha = \alpha_0 + \alpha_2 E^2 = \alpha_0 + \beta I, \tag{1}$$

where n is the refractive index and α is the absorption coefficient.

In a Z-scan experiment, the far-field on-axis intensity $I(z)$ is measured as a function of sample position. For a sample where nonlinear absorption is negligible, the on-axis normalized transmittance T_N^{SBF} as function of the sample position z is given approximately by[2,8]

$$T_N^{SBF}(z) \simeq 1 + \Delta\Phi_0 \frac{4x}{(1 + x^2)(9 + x^2)}, \tag{2}$$

where $\Delta\Phi_0$ is the on-axis nonlinear phase shift at focus, x is the dimensionless sample position $x = z/z_0$, and $z_0 = \pi w_0^2/\lambda$ is the Rayleigh range of the Gaussian beam with waist w_0. Sample position is measured from the focus at $z = 0$; the positive z direction is toward the laser source.

The nonlinear phase shift is given by

$$\Delta\Phi_0 = \frac{2\pi\Delta n d}{\lambda} = \frac{2\pi\gamma I_0 d}{\lambda} = \frac{2\pi d}{\lambda} \frac{n_2}{\varepsilon_0 c n_0} I_0, \tag{3}$$

where ε_0 is the permittivity of free space, λ is the wavelength, I_0 is the on-axis intensity at focus, Δn is the intensity-dependent change of the refractive index, and d is the sample thickness. This expression [Eq. (2)] approximates the exact result well if $\Delta\Phi_0$ is small. The Z-scan curve corresponding to Eq. (2) is shown in Fig. 1(a).

Extrema in the transmittance curve occur at $x = 0.8585$ where $T_N^{SBF} = 1 \pm 0.203\Delta\Phi_0$; the nonlinear phase shift can therefore be determined from the difference between the transmittance peak and valley; the peak–valley difference ΔT_{p-v}^{SBF} is simply

$$\Delta T_{p-v}^{SBF} = 0.406|\Delta\Phi_0|. \tag{4}$$

The chief advantage of the Z-scan (SBF) method is that it allows straightforward determination of the magnitude and sign of the nonlinear phase shift $\Delta\Phi_0$ from the peak–valley difference in the Z-scan curve. The separation Δz_{pv}^{SBF} of the transmittance peak and valley in the z direction is

$$|\Delta z_{pv}^{SBF}| = 1.717\ z_0,$$

and measurement of Δz_{pv} could provide one check of the applicability of the SBF method.

The response given by Eq. (2) also describes systems nonlocal in time, once they have reached steady state.[2] We note that, recently, D. I. Kovsh *et al.*[7] have studied thermal refraction using the Z-scan method and developed extensive numerical modeling. This research, however, focuses on the nanosecond regime, where the transit time of sound waves across the beam waist is comparable to the pulse width. Our interest is in the long time regime, where the refractive-index change is proportional to the change in temperature.

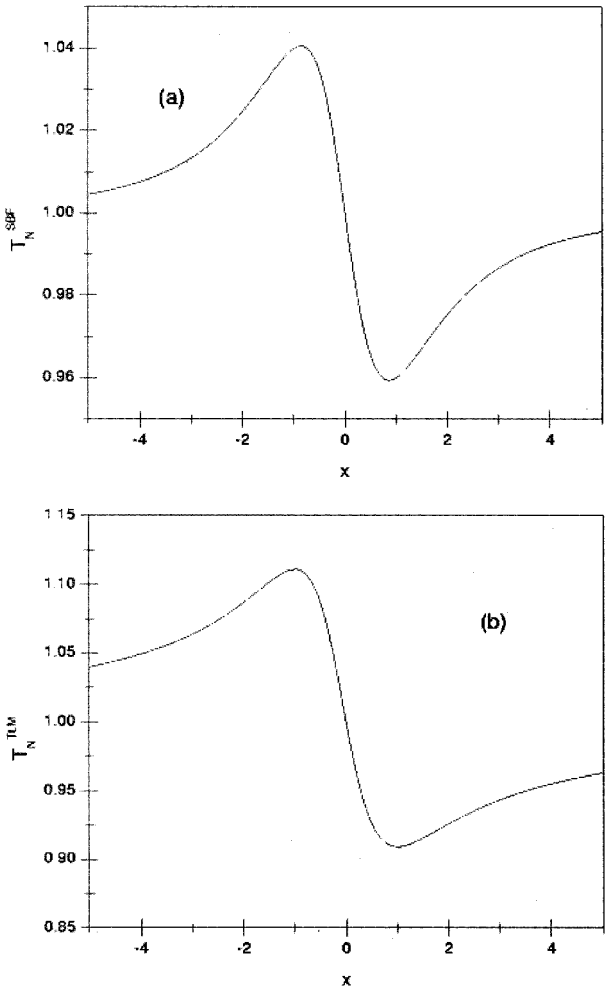

Fig. 1. (a) Z-scan curve from Eq. (2), the Sheik-Bahae formalism (SBF) with $\Delta\Phi_0 = -0.2$; (b) Z-scan curve from Eq. (14), the Thermal-lens model (TLM). $\theta = 0.1$.

In terms of the measured far-field on-axis intensity $I(z, t)$, with sample at position z at time t, the normalized transmittance can be written as

$$T_N^{SBF}(z) = \frac{I(z, \infty)}{I(\infty, \infty)}. \tag{5}$$

Assuming that, at $t = 0$ the measured intensity is independent of z, we have

$$I(\infty, \infty) = I(z, 0), \tag{6}$$

and Eq. (5) can be written as

$$T_N^{SBF}(z) = \frac{I(z, \infty)}{I(z, 0)}. \tag{7}$$

This expression allows comparison of the SBF model with the thermal lens (TLM) model considered below.

3. THERMAL LENS MODEL

The effects of local heating due to the absorption of light on the propagation of a Gaussian beam were studied by

Gordon *et al.*[16] in 1964; this study forms the foundation of the thermal-lens model (TLM). In this application, as the Gaussian beam propagates through a weakly absorbing sample, light is absorbed and immediately gives rise to local heating. Diffusion of heat is assumed to take place in the radial direction (perpendicular to the z direction), giving rise to a spatially varying temperature field that is not proportional to the local light intensity. The refractive index is assumed to depend linearly on temperature, and the nonlinearity is due to this nonlocal dependence of the refractive index on the light intensity. In their 1984 paper comparing models for the thermal-lens effect, Carter and Harris summarize earlier results[16–18] describing the effects of laser heating by a Gaussian beam on its far-field intensity. The analysis is predicated on a parabolic approximation to the temperature field[16] and on approximating the optical effects due to temperature as being the same as those of a simple lens. Their time-dependent expression[19] for the far-field on-axis transmittance is

$$T_{\mathrm{N}}^{\mathrm{TLM}}(z,\,t) = \frac{I(z,\,t)}{I(z,\,0)} = \frac{1}{1 + \left(\dfrac{\theta}{1 + t_c/2t}\right)\dfrac{2x}{1 + x^2} + \left(\dfrac{\theta}{1 + t_c/2t}\right)^2 \dfrac{1}{1 + x^2}}, \tag{8}$$

where $x = z/z_{\mathrm{o}}$ as before, the thermal-diffusion time t_c is given by

$$t_c = \frac{w^2}{4D}, \tag{9}$$

where $w = w_{\mathrm{o}}\sqrt{1 + x^2}$ is the beam waist, $D = \kappa/C_p$ is the thermal diffusivity, C_p is specific heat per volume at constant pressure, and κ is the thermal conductivity. The on-axis phase shift (the phase difference between the center and the outer edge of the beam)[17] is given by

$$\theta = \frac{\alpha_{\mathrm{o}} P d}{\lambda \kappa}\frac{\partial n}{\partial T}, \tag{10}$$

where α_{o} is the linear absorption coefficient, P is the power of the Gaussian beam, and T is the temperature.

Assuming that the nonlinear phase shift $\theta \ll 1$, the second term in the denominator of Eq. (8) can be neglected, and the far-field transmittance is

$$T_{\mathrm{N}}^{\mathrm{TLM}}(z,\,t) \simeq \frac{1}{1 + \left(\dfrac{\theta}{1 + t_c/2t}\right)\dfrac{2x}{1 + x^2}}. \tag{11}$$

Equation (11) thus gives the on-axis normalized transmittance $T_{\mathrm{N}}^{\mathrm{TLM}}(z,\,t)$ as a function of the sample position z and time t. We point out that the thermal-diffusion time is position dependent. It follows that

$$t_c = \frac{w_{\mathrm{o}}^2(1 + x^2)}{4D} = t_{\mathrm{co}}(1 + x^2), \tag{12}$$

where $t_{\mathrm{co}} = w_{\mathrm{o}}^2/4D$. The normalized transmittance then, with explicit sample position dependence, is

$$T_{\mathrm{N}}^{\mathrm{TLM}}(z,\,t) = \left\{1 + \left[\frac{\theta}{1 + (1 + x^2)t_{\mathrm{co}}/2t}\right]\frac{2x}{1 + x^2}\right\}^{-1}. \tag{13}$$

At times $t \gg t_c$, the steady state has been reached, and here

$$T_{\mathrm{N}}^{\mathrm{TLM}}(z) \simeq \frac{1}{1 + \theta\dfrac{2x}{1 + x^2}}. \tag{14}$$

The Z-scan curve corresponding to Eq. (14) is shown in Fig. 1(b). Extrema in the transmittance curve occur at $x = \pm 1$, where $T_{\mathrm{N}}^{\mathrm{TLM}} = 1/(1 \pm \theta) \cong 1 \mp \theta$. The nonlinear phase shift can therefore be determined from the difference between the transmittance peak and valley; the peak–valley difference $\Delta T_{\mathrm{p-v}}^{\mathrm{TLM}}$ is simply

$$\Delta_{\mathrm{p-v}}^{\mathrm{TLM}} \simeq 2|\theta|. \tag{15}$$

The separation $\Delta z_{\mathrm{pv}}^{\mathrm{TLM}}$ of the transmittance peak and valley in the z direction is

$$|\Delta z_{\mathrm{pv}}^{\mathrm{TLM}}| \approx 2z_{\mathrm{o}}. \tag{16}$$

Measurement of Δz_{pv} could provide one check of the applicability of the TLM method.

4. DISCUSSION

The Z-scan curves from the Sheik-Bahae formalism (SBF) and the thermal-lens model (TLM) as shown in Figs. 1(a) and 1(b) are remarkably similar. The peak–valley separations predicted by the two schemes are very close; $\Delta z_{\mathrm{pv}}^{\mathrm{SBF}}| = 1.7z_{\mathrm{o}}$, and $\Delta z_{\mathrm{pv}}^{\mathrm{TLM}}| = 2z_{\mathrm{o}}$. One significant discrepancy is that a given peak–valley difference $\Delta T_{\mathrm{p-v}}$ corresponds to a nonlinear phase shift of

$$|\Delta\Phi| = \frac{\Delta T_{\mathrm{p-v}}}{0.406} \tag{17}$$

in the SBF, and it corresponds to a nonlinear phase shift of

$$|\theta| = \frac{\Delta T_{\mathrm{p-v}}}{2} \tag{18}$$

in the TLM. Some discrepancy is expected, since the refractive-index profiles are different in the two cases. Approximations made in estimating the far-field intensity in the TLM likely also contribute to this discrepancy.

Distinguishing features of the two models are associated with the nonlocal aspects of the thermal response. Time dependence associated with the thermal-diffusion time t_{o} is explicit in the TLM. This is easily observable experimentally; typically, $D = 10^{-7}$ m^2/s, $\omega \simeq \omega_{\mathrm{o}} = 2 \times 10^{-5}$ m, and

$$t_{\mathrm{o}} = \frac{w^2}{4D} \approx 1\ \mathrm{ms}. \tag{19}$$

The nonlinear phase shift in both cases can be written in terms of the nonlinear index n_2. In the local SBF, this is

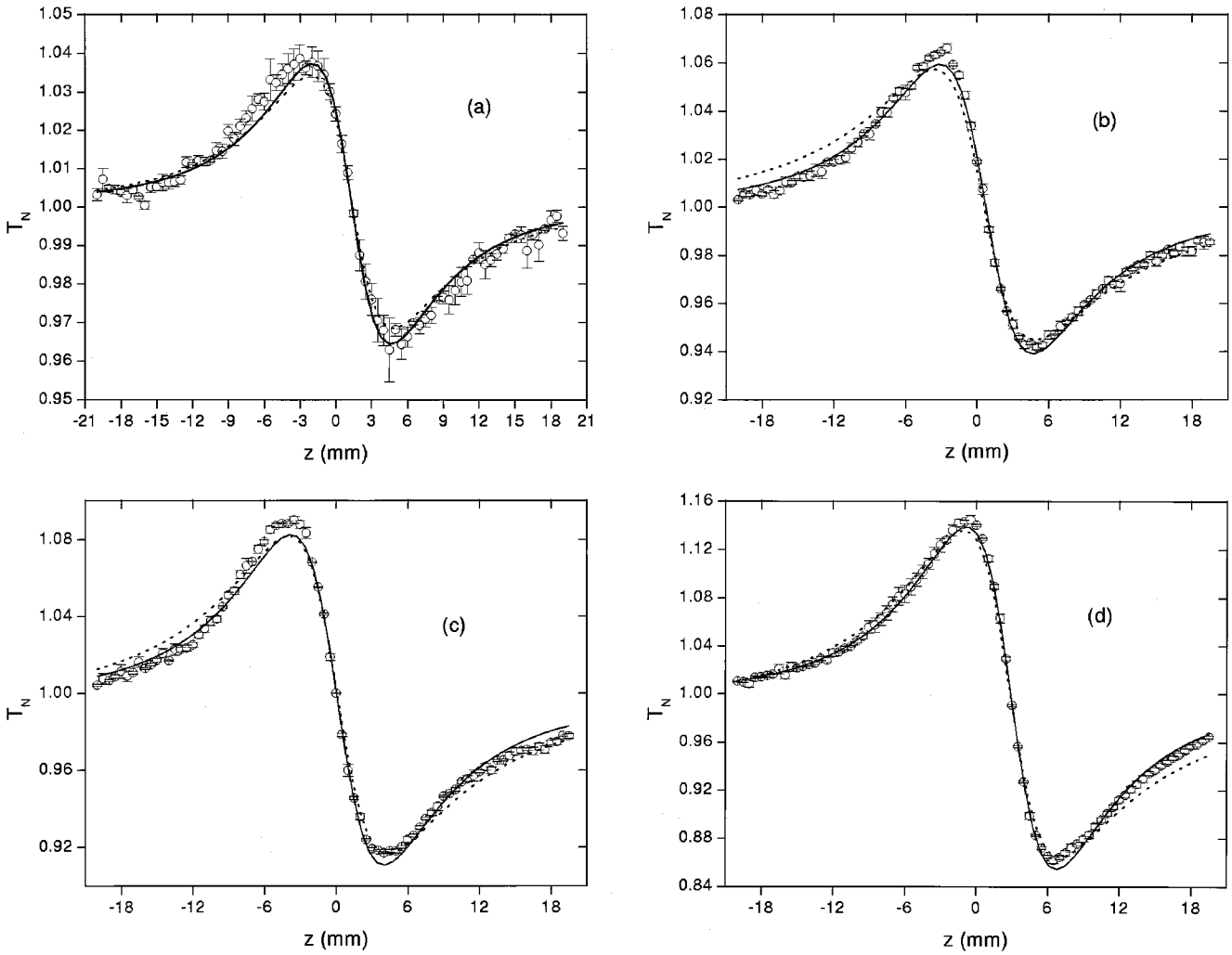

Fig. 2. Typical Z-scan results with the different lyotropic mixtures. The solid curve corresponds to the SBF fitting [Eq. (2)] and the dotted curve to the TLM [Eq. (13) with $t \approx 10t_{co}$]. (a) Undoped mixture; (b) ferrofluid-doped mixture (c_1); (c) ferrofluid-doped mixture (c_2); (d) ferrofluid-doped mixture (c_3).

$$\Delta\Phi = \frac{2\pi d}{\lambda}\frac{n_2}{\varepsilon_0 c n_0} I_0. \tag{20}$$

A signature of spatial nonlocality is the dependence of the nonlinear phase shift on the beam waist; writing θ in terms of the intensity, TLM gives

$$\theta = \frac{2\pi d}{\lambda}\frac{n_2}{\varepsilon_0 c n_0} I_0 = \frac{\pi \alpha_0 w_0^2 d}{2\lambda\kappa}\frac{\partial n}{\partial T} I_0. \tag{21}$$

The dependence of the ratio of the nonlinear phase shift to intensity I_0, or of the nonlinear index n_2, on beam waist is therefore clear evidence of the spatially nonlocal response.

A subtler effect distinguishing the local SBF and nonlocal TLM is the dependence of the thermal-diffusion time on the beam waist. Since $T_N^{TLM}(z, t)$ is given by Eq. (13), if the response is measured at times comparable to t_{co}, the shape of the Z-scan curve can vary significantly.

It is worth noting, however, that if the laser pulse is long enough for the thermal lens to develop, but short enough to neglect the thermal diffusion ($t \ll t_{co}$), the complications caused by the time-varying beam size (due to the thermal diffusion) can be avoided. In this adiabatic limit, the response is spatially local, and the "instantaneous" thermal lens can be simply characterized. The refractive-index change is proportional to the temperature change, which is given by

$$\Delta T = \frac{\alpha_0 I t}{C_p}, \tag{22}$$

and the corresponding index change is

$$\Delta n = \frac{\alpha_0}{C_p}\frac{\partial n}{\partial T} I t, \tag{23}$$

and the nonlinear phase shift is everywhere proportional to the fluence. We would expect the TLM to reduce in this case essentially to the SBF. It does not, however; at least in part this is due to the approximations made in the parabolic description of the temperature field. The expression for the temperature used by Gordon *et al.*,[16] before the parabolic approximation, is

$$\Delta T(r, t) = \frac{A\omega_o^2}{8k}\left\{\mathrm{Ei}\left(-\frac{2r^2}{w_o^2}\right) - \mathrm{Ei}\left(-\frac{2r^2}{8Dt + w_o^2}\right)\right\}, \tag{24}$$

which in fact reduces to the appropriate Gaussian form in the short-time limit.

We finally note that one serious concern regarding the applicability of the TLM is its assumption that thermal diffusion occurs only in the radial direction. In thin liquid samples, which are usually between glass plates, even though the absorption may only be in the liquid, a considerable amount of heat flows into the glass. Heat diffusion in the z direction therefore cannot be neglected, and the temperature profile will differ significantly from that in Eq. (16). The current TLM is not expected to give a good description of the response in such cases.

5. Z-SCAN MEASUREMENTS ON LYOTROPIC LIQUID CRYSTALS

The lyotropic liquid crystal[20] used was a mixture of potassium laurate (KL)/decanol (DeOH)/water, in concentrations 26.02/6.72/67.26 wt. %. Additional samples doped with a water-base ferrofluid[20] were also prepared, with ferrofluid concentrations of $c_1 = 1.9 \times 10^{11}$, $c_2 = 3.7 \times 10^{11}$, and $c_2 = 7.4 \times 10^{11}$ grains/cm^3. The addition of ferrofluid increases the sample absorptivity and consequent heating of the liquid-crystal medium.[14]

Samples of thickness 200 μm were prepared with glass microslides, with each wall having a thickness of 1 mm. The measurements were carried out at the temperature of $T = 23\,^{\circ}\mathrm{C}$, when the samples are in the isotropic phase. A solid-state cw Millennia laser was used at $\lambda = 532\,\mathrm{nm}$. A mechanical chopper was used to modulate the intensity, providing nearly square pulses with a pulse width of 25 ms. The power of the beam ranged from 90 to 400 mW; a typical beam waist at the sample was 30 μm. A signal-acquisition scheme with temporal resolution was used to eliminate linear effects.[4,21] Details of the setup and sample preparation are given elsewhere.[14]

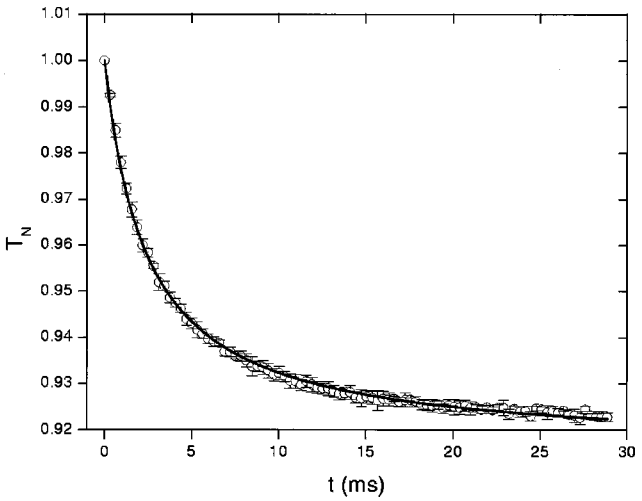

Fig. 3. Typical time-dependent transmittance at a fixed z (ferrofluid-doped mixture with c_2). The solid curve is a fitting to Eq. (13).

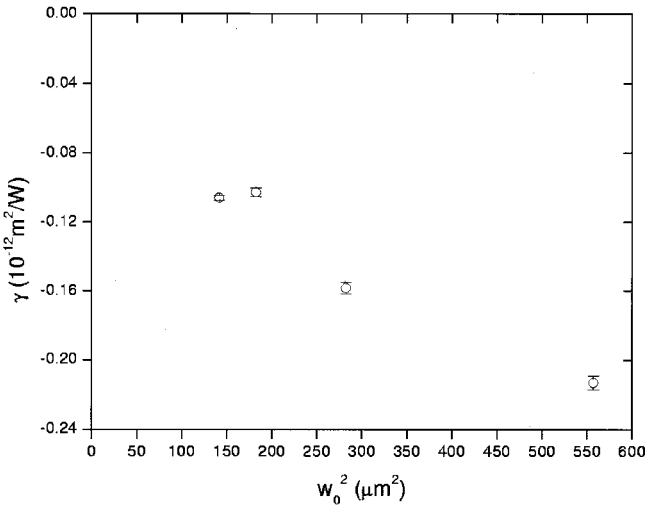

Fig. 4. Beam-waist dependence of the nonlinear index γ. Undoped mixture. Incident power of 238 mW.

Figure 2 shows the typical Z-scan experimental results from the different lyotropic mixtures investigated. The experimental data are analyzed according to the SBF [Eq. (2)] and TLM [Eq. (13), with $t = t_p \approx 40$ ms, where t_p is the pulse width].

In the case of the SBF, the Z-scan fitting procedure with Eq. (2) furnishes $\Delta\Phi_o$. In the case of the TLM, however, a self-consistent fitting procedure has to be used. At a given position z and pulse width $t_p \approx 40$ ms, the time-dependent transmittance is measured. This function [Eq. (13)] has two fitting parameters: θ and t_{co}. The Z-scan curve [Eq. (13), with $t = t_p \approx 40$ ms] also has these two fitting parameters. Both parameters are obtained by means of a self-consistent fitting procedure that takes into account all the time-dependent transmittances at each spatial position z and the Z-scan curve. The fit gives $t_{co} \approx 3\,\mathrm{ms} \approx t_p/10$. The temporal response, together with the fit from Eq. (13), are shown in Fig. 3. The diffusion time t_{co} allows experimental determination of the thermal diffusivity D, since

$$D = \frac{w_o^2}{4t_{co}}. \tag{25}$$

Since the beam waist is $w_o = 30\,\mu$m, we obtain $D \approx 10^{-7}\,\mathrm{m^2/s}$. This is in reasonable agreement with the value of D of water. Assuming $C_p^{\mathrm{water}} = 4.18 \times 10^7\,\mathrm{J/kg\,K}$ and $\kappa^{\mathrm{water}} = 0.6\,\mathrm{J/s\,K\,m}$,[22] $D^{\mathrm{water}} = \kappa/\rho C_p \simeq 1.5 \times 10^{-7}/\mathrm{s}$, as expected, since our lyotropic sample is mainly water. Our results also agree with the one obtained by Bento *et al.*[23]

The TLM predicts that, due to the nonlocal response, the nonlinear refractive index n_2 (or γ) depends on the beam waist. From Eq. (21), we obtain

$$n_2 = cn_o\gamma = \frac{\varepsilon_o c n_o \alpha_o}{4\kappa}\frac{\partial n}{\partial T}w_o^2, \tag{26}$$

and expect n_2 (or γ) to be proportional to w_o^2. Measuring the peak–valley difference allows determination of the nonlinear phase shift, and from this, by Eq. (20) or (21), n_2 (and γ) can be determined. The TLM predicts a de-

pendence of n_2 on the beam waist, which, in fact, is observed. The beam-waist dependence of the nonlinear index γ is shown in Fig. 4. Fitting a straight line through the origin to the data gives the slope $\partial n_2/\partial w_0^2 = -(2.7 \pm 0.5) \times 10^{-4}$ W^{-1}, and we get, from Eq. (26),

$$\frac{\alpha_0}{\kappa}\frac{\partial n}{\partial T} = \frac{\varepsilon_0 c n_0}{4}\frac{\partial n_2}{\partial w^2}. \tag{27}$$

Thus analysis of the data with the TLM allows determination of the thermal diffusivity and the thermo-optic coefficient if κ and α_0 are known.

Table 1 presents the different values of n_2, $\partial n/\partial T$, and κ obtained by means of least-square fits of Eqs. (2) and (13) (with $t = t_p$) to the experimental data. The thermal conductivity measured increases with the ferrofluid doping concentration. The nonlinear refractive index increases and the thermo-optical coefficient decreases, in absolute values, with increasing ferrofluid concentration. Table 2 presents the nonlinear optical phase shift in both SBF and TLM obtained from the fitting procedure; the ratio $\theta/|\Delta\Phi_0| = 0.203$ is in agreement with Eqs. (17) and (18). Table 3 shows the quality of the fit of the experimental Z-scan data to theory in terms of the χ^2 criterion. The SBF expression of Eq. (2) consistently provides a better fit than the TLM expression of Eq. (13).

The dominant physical process that is assumed to take place in these samples is density change due to laser heating, which in turn leads to a change of the refractive index. The thermal expansivity α_T of the lyotropic liquid crystal[24] without the ferrofluid doping is ~3 times that of water. When ferrofluid is added, the doped sample becomes more absorptive; the absorption depends on the concentration of the magnetic grains. At low doping levels, a linear dependence on concentration is expected. This was observed in our experiments.

Measurements done with the open-aperture Z-scan setup did not indicate, within the sensitivity of our experiment, nonlinear absorption. The Z-scan results obtained with a closed aperture were always symmetric with respect to the asymptotic value of the normalized transmittance for $z \to \pm\infty$. This implies that the absorption is linear, as expected for the low light intensities in the experiments. The lyotropic liquid-crystal sample, without the ferrofluid doping, consists primarily of water (~94 mol.%), and its linear absorption coefficient is essentially that of water (~10^{-3} cm^{-1}). The addition of ferrofluid, however, can increase the absorption coefficient by many orders of magnitude.

Table 1. Nonlinear Refractive Index (n_2) in the SBF Framework, Thermal Conductivity (κ), and Thermo-optic Coefficient ($\partial n/\partial T$) in the TLM Framework, of the Different Lyotropic Samples Investigated

c(10^{11}grains /cm^3)	$-n_2$(10^{-7} esu)	κ(W m^{-1} °C^{-1})	$-$dn/dT (10^{-5} °C^{-1})
0	3.4 ± 0.2	0.249 ± 0.009	9.4 ± 0.6
1.9	5.7 ± 0.3	0.297 ± 0.018	4.0 ± 0.3
3.7	8.3 ± 0.9	0.308 ± 0.009	2.7 ± 0.2
7.4	13.6 ± 0.4	0.332 ± 0.009	1.38 ± 0.08

Table 2. Nonlinear Optical Phase Shifts ($\Delta\Phi_0$ and θ) and Their Ratio

c(10^{11} grains /cm^3)	θ(10^{-2} rad)	$-\Delta\Phi_0$ (rad)	$\theta/\lvert\Delta\Phi_0\rvert$
0	3.8 ± 0.1	0.1722 ± 0.0022	0.209 ± 0.001
1.9	6.1 ± 0.2	0.273 ± 0.006	0.207 ± 0.001
3.7	9.0 ± 0.3	0.429 ± 0.005	0.207 ± 0.001
7.4	14.9 ± 0.5	0.8896 ± 0.0015	0.209 ± 0.001

Table 3. Fit χ^2 of SBF and TLM Expressions to Experimental Z-Scan Data

c(10^{11} grains /cm^3)	χ^2_{SBF}	χ^2_{TLM}
0	2.97	4.75
1.9	8.76	33.86
3.7	31.51	66.12
7.4	4.95	24.27

As a final remark, we note that, in thick samples with local response, the errors introduced by the parabolic phase approximation can be corrected, to lowest order, by decreasing the curvature of the parabolic index profile by a numerical factor (a factor).[25] In our experiments on thin samples with nonlocal response, this approach did not improve the agreement between the SBF model and the experimental data.

6. CONCLUSIONS

Z-scan experiments were carried out on lyotropic liquid-crystal samples with and without ferrofluid doping. The experimental results were analyzed with the SBF and the TLM. The TLM describes well features of the nonlocal nonlinearity, such as the time dependence of the transmittance and the dependence of the nonlinear refractive index on the beam waist. The time and beam-waist dependence can be used to obtain information about thermal diffusivity and the thermo-optic coefficient. Interestingly, the shape of the experimental Z-scan curve is better described by the SBF than by the TLM. This is likely due to approximations made in the TLM that are not well obeyed. These include the assumption that no thermal diffusion takes place in the z direction, the parabolic approximation to the temperature profile, and the assumption that the far-field intensity profile is the same as what would be produced by a simple lens whose focal length is estimated. We expect that, if the dominant heat flow was into the glass cell walls, then the temperature profile would more closely resemble the intensity, and in this case the local SBF model would provide a better description than TLM. It is also possible that mechanisms other than laser heating are contributing to the observed nonlinearity: electrostriction and the Soret effect could be such mechanisms.

We have found that, in addition to the SBF, the TLM offers important valuable insights into the intensity-dependent optical response of complex fluids such as liquid crystal. The simplicity of the steady-state analysis

presented here may make this approach particularly useful for applications such as the measurement of small absorption coefficients in samples where other material parameters are known. Much challenging research in more accurately modeling the thermal and optical response remains to be done.

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