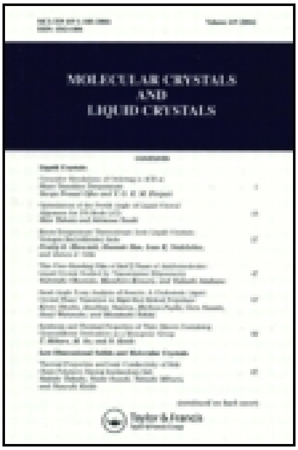

Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl19

Transient Characteristics of Photothermal Self-Phase Modulation in Guest Host Liquid Crystals

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Version of record first published: 24 Sep 2006

Mol. Cryst. and Liq. Cryst., 2001, Vol. 368, pp. 377-386
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Transient Characteristics of Photothermal Self-Phase Modulation in Guest Host Liquid Crystals

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Transient properties of self-phase modulation in guest host liquid crystals (GHLCs) have been quantitatively by both experimental and theoretical approaches. By theoretical calculations using time-dependent heat-conduction analysis, we have time constants in good agreement with the experimental observation for two kinds of GHLC cells with glass and acrylate substrates.

Keywords: self-phase modulation; heat conduction analysis; guest host liquid crystal; nonlinear optics

1. INTRODUCTION

Nematic liquid crystals (NLCs) posses an abundance of useful characteristics, including very broadband large birefringence, transparency, unusually large susceptibility to ac, dc and optical fields, and ease of fabrication of large areas [1]. It is known that optically induced NLC director reorientation results in extraordinary large nonlinear refractive index change, which can be enhanced by doping appropriate dye molecules (guest host NLC, GHLC) [1]. On the other hand, it is known that NLCs have large gradients $\partial n_e / \partial T$ and $\partial n_o / \partial T$, where n_e and n_o are the extraordinary and ordinary refractive indices, respectively. Even though the director reorientation of NLCs does not occur, photosensitivity (photothermal effect) is achieved by absorbing the laser beam and heating NLC [1-21]. Photothermal effect is most fundamental phenomenon in laser-induced refractive index change in NLCs and self-phase modulation is typical of the type of nonlinear wave propagation which is caused by a photothermal refractive index change in GHLC. It is very important to characterize the photothermal effects in GHLC in order to apply other photophysical and/or photochemical properties to optically active devices.

The first purpose of this study is to present the characterization and analyzing method for the transient properties of photothermal effects in GHLC. The second purpose is to clarify the effect of kind of LC cell substrate by means of developed characterization method.

2. SELF-PHASE MODULATION IN GHLC

In the present paper, we treat homogeneously aligned GHLC and the polarization direction of the pump laser beam is set to be parallel to the director of GHLC. In case of a beam polarized parallel to the GHLC director, the gradient $\partial n_e / \partial T$ is negative.

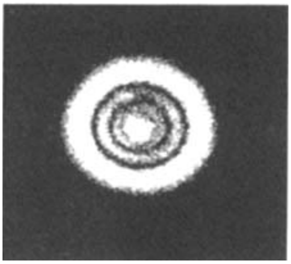

FIGURE 1 Profile of the diffraction pattern from GHLC.

When the intensity of pump beam is sufficiently large and the maximum phase increment due to the photothermal effect is much larger than 2π, the number of the bright rings is appeared as shown in Figure 1. Since the thermal conductivity of GHLC shows strong anisotropy and the elliptical distribution of the heat is generated by photothermal effects, the diffraction pattern from GHLC shows the elliptical shape as shown in Figure 1.

We use the single-mode laser beam with the Gaussian transverse profile propagating into the GHLC film and we have

$$I(x) = I_0 \exp\left(-\frac{2x^2}{\omega^2}\right) \quad , \tag{1}$$

where, I_0 is the on-axis intensity, ω is the beam waist, and x is the coordinate parallel to the LC director. According to Kirchhoff' diffraction theory, a far-field diffraction intensity distribution of the Gaussian beam transmitted through the GHLC can be given as

$$I(\rho,\ Z) = \left(\frac{2\pi}{\lambda Z}\right)^2 I_0 \left|\int_0^\infty x dx J_0\left(k_0 \frac{x\rho}{2Z}\right) \exp\left[-2\frac{x^2}{\omega^2} - i\phi(x)\right]\right|^2 \quad , \tag{2}$$

where, ρ is the distance from the center of the far-field pattern at the observation point, Z is the distance from the GHLC film to the observation point, $J_0(x)$ is the first-order Bessel function of the first kind, k_0 is the wavenumber in free space. The phase factor, $\phi(x)$, consists of the Gaussian phase depending on the wave-front curvature and the phase shift of the beam transmitted through the medium of the thickness d, given as

$$\phi(x) = \phi_L(x) + \phi_{NL}(x), \tag{3}$$

where $\phi_L(x)$, and $\phi_{NL}(x)$ are the intensity-independent and the intensity-dependent (nonlinear) phases, respectively, and are given by

$$\varphi_L(x) = k_0\left(\frac{x^2}{2Z} + \frac{x^2}{2f}\right) \tag{4}$$

$$\varphi_{NL}(x) = k_0\int_0^d \Delta n(x, z)dz \equiv k_0 d\Delta n(x) \tag{5}$$

where $\Delta n(x, z)$ is the distribution of the refractive index change, which is negative in our experimental condition. According to our previous works, the higher-order intensity dependence of the refractive index is given as

$$\Delta n(x) = n_2 I(x) - n_4 [I(x)]^2. \tag{6}$$

where, n_2 and n_4 are nonlinear coefficients.

3. SAMPLES

The nematic LC, 4'-pentyl-4'-cyanobiphenyl (5CB), was obtained from Merck Japan Ltd. Commercially available Disperse Blue 14 (DB14) was obtained from Aldrich Co., Ltd. as guest dye and was used without further purification. GHLC was sandwiched between two parallel unidirectionally rubbed poly(vinyl alcohol) coated with transparent substrates. The dye concentration was 0.075 wt% and the GHLC thickness was 100 μm. In order to clarify validity of our characterization method, we used two kinds of substrates, glass and acrylate, which show the different thermal conductivities. Since the GHLC was homogeneously aligned due to the guest-host effect, the GHLC showed strong anisotropy. The thickness of glass and acrylate substrates were 1 and 2 mm, respectively.

4. TRANSIENT DIFFRACTION PATTERNS

Figure 2 shows experimental setup for transient self-phase modulation in GHLC. A linearly polarized He-Ne laser (632.8 nm) with an intensity of 3 mW was used for exciting the self-phase modulation in GHLC. The polarization direction of the He-Ne laser beam was controlled by a half-wave plate and set to be parallel to the director of GHLC.

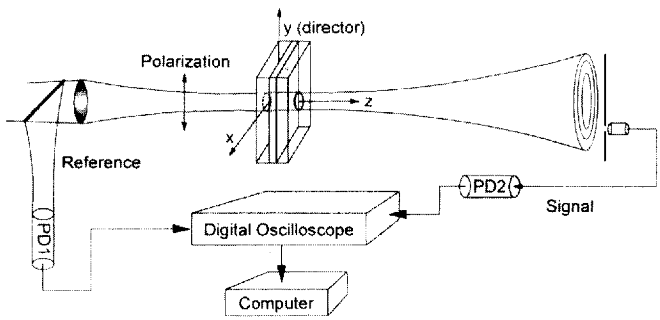

FIGURE 2 Experimental setup for measuring transient self-phase modulation.

The He-Ne laser beam was focused to an e^{-2} diameter of 134 μm at the GHLC cell by a focusing lens ($f = 30$ cm). A part of the traverse diffraction beam was cut using a pinhole of 25 μm radius and guided to a photodiode (PD2). In order to characterize the transient properties of He-Ne laser beam was separated into excited and reference beams before the beam incident the GHLC cell. The laser beam was switched by a mechanical shutter and the reference beam was monitored by another photodiode (PD1) in order to determine the time when the beam was started to incident the sample. Both transient signals from PD1 and PD2 were monitored by a Kikusui K-7101A digitizing oscilloscope and typical signals are shown in Figure 3.

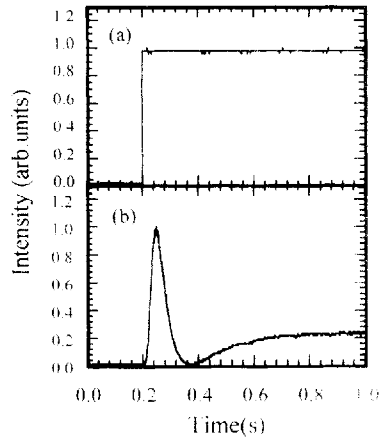

FIGURE 3 Typical transient signals monitored by (a) PD1 (reference) and (b) PD2 (fringe signal).

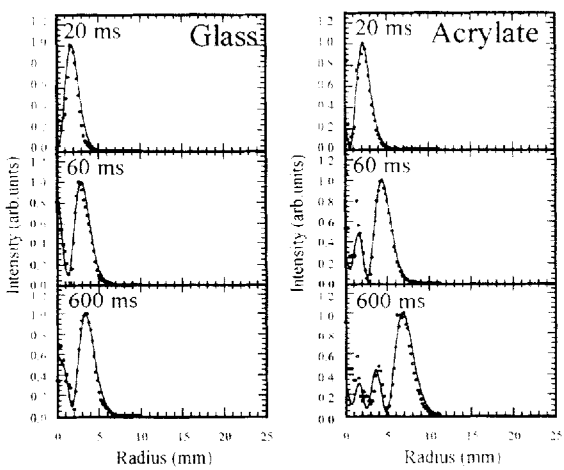

FIGURE 4 Transient radial intensity distributions of the diffraction pattern from GHLC. Filled circles, experimental data; solid curves, theoretical fitted curves.

The pinhole was set on the pulse stage and one-dimensional scans was performed. The same transient signals as shown in Figure 3 were obtained by repeating the same transient experiment at each points. A series of signals is reorganized into the transient fringe signals at each time as described in Figure 4. As the time is passed, the power from the central region appears to be pushed out into the wing region. The larger number of diffraction fringe from GHLC cell with acrylate substrates were observed in comparison with those from GHLC with glass substrates because thermal conductivity of the acrylate substrate is much lower than that of the glass substrate, which will be quantitatively discussed in the next section.

5. TRANSIENT HEAT CONDUCTION ANALYSIS

In order to clarify the quantitative mechanism for transient self-phase modulation obtained from experiments, time-dependent heat conduction analysis was performed by numerically solving the heat conduction equation. According to Biot-Fourier phenomenological assumption, we obtain the following expression: [19, 21]

$$\rho_d c \frac{\partial T}{\partial t} = \frac{\partial}{\partial x}\left(k_x \frac{\partial T}{\partial x}\right) + \frac{\partial}{\partial y}\left(k_y \frac{\partial T}{\partial y}\right) + \frac{\partial}{\partial z}\left(k_z \frac{\partial T}{\partial z}\right) + Q(x, y, z) \tag{7}$$

where k_x, k_y, and k_z denote the three-dimensional thermal conductivities, ρ_d denotes the density of the media, c is specific heat, and $Q(x, y, z)$ is a heat source which is supplied from the laser beam. Equation (7) was solved by means of finite-element method and time-dependent temperature distribution $T(x, y, z, t)$ was obtained. Figure 5 shows a typical of temperature distributions at the boundary between substrate and GHLC layer.

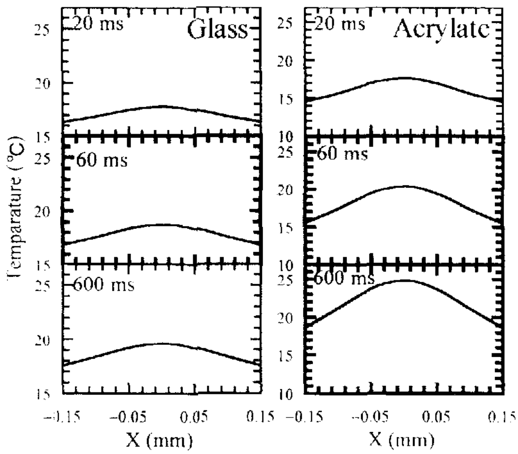

FIGURE 5 Calculated time-dependent temperature distributions. In the case of glass substrates, $T(x,0,1,t)$ at $y=0$ and $z=1$ mm were described in the figure, while $T(x,0,2,t)$ for acrylate substrates.

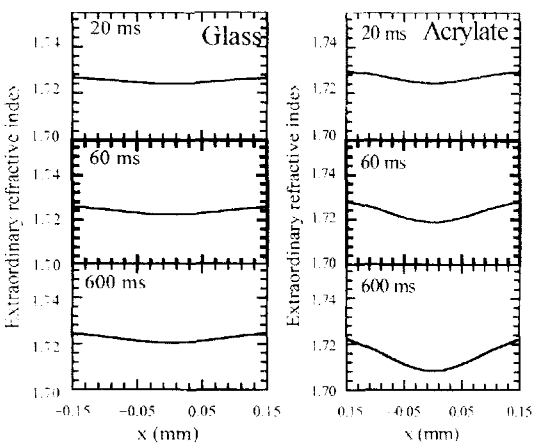

FIGURE 6 The calculated transient refractive index distributions due to the photothermal effects.

6. COMPARISON EXPERIMENTS WITH THEORY

In order to discuss transient self-phase modulation, refractive index distributions have to be estimated from the calculated temperature distributions. We can calculate the index distributions by the temperature distributions and the temperature variation of the refractive index. Figure 6 shows the index distributions in the x-axis direction.

Figure 7 shows the time-dependent refractive index change [$\Delta n(0)$].

The data are normalized at the saturated value of refractive index change. Although the slight difference between the theory and experiment for the GHLC cell with acrylate substrates was observed, we can obtain the similar time constant from both experimental observation and theoretical estimation.

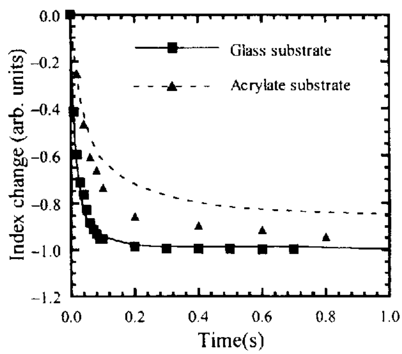

FIGURE 7 Time-dependent refractive index change. Filled squares and triangles were obtained from experiments for GHLC cells with glass and acrylate substrates, respectively. The solid and dotted curves were obtained from theoretical calculations.

7. CONCLUSIONS

We quantitatively characterize the photothermal self-phase modulation including the time constant. The theoretical calculations by means of time-dependent heat conduction analysis were very useful for characterizing the photothermal effects in GHLC. By using characterizing method described here, we also explained the difference between the photothermal effects in GHLC with glass substrates and those in GHLC with acrylate substrates.

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