

Laser-heating-induced self-phase modulation, phase transition, and bistability in nematic liquid crystals

Peng-ye Wang, Hong-jun Zhang, and Jian-hua Dai

Institute of Physics, Academia Sinica, P.O. Box 603, Beijing, China

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A quantitative discussion of the laser-heating-induced self-phase modulation in nematic liquid crystals is given. The process of the laser-heating-induced nematic-isotropic transition is analyzed. The intrinsic optical bistability due to the nematic-isotropic phase transition is observed and analyzed.

It is well known that a liquid crystal is a type of nonlinear medium with a strong temperature dependence.¹ Consequently a number of nonlinear optical phenomena can be readily induced by laser-heating effects, such as the thermal lens effect² and the formation of the isotropic holes in nematic liquid crystals.^{3,4} But only a qualitative description of these phenomena has been given. In this Letter we present a quantitative study of the laser-heating-induced self-phase modulation and the nematic-isotropic phase transition. The intrinsic optical bistability due to the nematic-isotropic (N-I) phase transition is observed and analyzed in a pure nematic liquid crystal for the first time to our knowledge. Hysteresis and discontinuous changes of the diffraction light are observed. Then the liquid-crystal intrinsic optical bistability is performed without the Fabry-Perot interferometer⁵ or the feedback mirror.⁶ Similar phenomena were observed in a liquid crystal containing dichroic dye.⁷

The experiment was performed at room temperature using E7, which is nematic in the temperature range of -10 – 62.5°C . The parallel-aligned nematic film was sandwiched between two glass plates coated with transparent conducting electrodes. The linearly polarized 514.5-nm input beam from an argon laser was nearly normally incident upon the liquid-crystal cell. The nematic director could be parallel or perpendicular to the laser polarization. The optical field-induced reorientation of the molecules does not exist in this case.⁸ Thus the effect of self-phase modulation due to the reorientation⁹ is eliminated. Therefore only the laser-heating effect is considered.

The argon laser was operated in a TEM₀₀ mode. The laser beam was Gaussian. The transverse distribution of the intensity can be written as

$$I(r) = I_0 \exp(-2r^2/r_0^2), \quad (1)$$

where r is the radial distance, r_0 is the beam radius, and I_0 is the central intensity.

Laser heating can result in a temperature rise in the liquid-crystal cell. We neglect the effect of the heat diffusion in the transverse plane and the temperature distribution in the cell along the direction of the laser beam, because the beam diameter is much larger than the thickness of the cell. Then the transverse distribution of the temperature can be written as

$$T(r) = \alpha I(r) + T_1, \quad (2)$$

where α is a constant and T_1 is the room temperature. We then have

$$T' = T_0 \exp(-2r^2/r_0^2), \quad (3)$$

where $T_0 = \alpha I_0$ and $T' = T - T_1$.

The temperature dependence of the refractive indices of E7 was obtained from the literature.¹⁰ We use the following approximation:

$$n_i = A_0 + A_1(T_c - T) + A_2(T_c - T)^2, \quad (4)$$

where $n_i = n_e$ or $n_i = n_o$; A_0 , A_1 , and A_2 are constants; and T_c ($=62.5^\circ\text{C}$) is the critical temperature. We get $A_0 = 1.660$, $A_1 = (4.979 \times 10^{-3})^\circ\text{C}^{-1}$, and $A_2 = (-5.459 \times 10^{-5})^\circ\text{C}^{-2}$ for n_e and $A_0 = 1.544$, $A_1 = (-1.517 \times 10^{-3})^\circ\text{C}^{-1}$, and $A_2 = (2.332 \times 10^{-5})^\circ\text{C}^{-2}$ for n_o from the least-squares fit to the data in Ref. 10. Inserting Eq. (3) into Eq. (4), we obtain

$$n_i = A_0 + A_1[T_c' - T_0 \exp(-2r^2/r_0^2)] + A_2[T_c' - T_0 \times \exp(-2r^2/r_0^2)]^2, \quad (5)$$

where $T_c' = T_c - T_1$. The transverse distribution of the refractive indices when the central temperature $T_{\text{max}} = T_0 + T_1$ is near T_c is shown in Fig. 1. The room temperature was $T_1 = 16.5^\circ\text{C}$ under our experimental conditions. The maximum changes of the refractive indices from Eq. (5) are $\Delta n_e = 0.1135$ and $\Delta n_o = 0.0206$. As with the formation of the ring patterns due to molecular reorientation,⁹ the total number of rings N can be estimated from the relation $N = \Delta\phi_0/2\pi$, where $\Delta\phi_0$ is the maximum phase difference. The thickness of the cell was $d \approx 15 \mu\text{m}$. We get the numbers of rings $N_e = 3$ and $N_o = 1$ for the extraordinary and ordinary light, respectively. The experimental results are shown in Fig. 2, which are in agreement with the theoretical estimation. A more accurate theory to describe the number of rings and ring formation may be found in Ref. 11.

The case discussed above is $T_{\text{max}} < T_c$. But if we continue to increase the input intensity, the temperature T_{max} will become higher than T_c . The liquid crystal in the central part of the laser beam will transform to the isotropic phase, i.e., a small isotropic hole

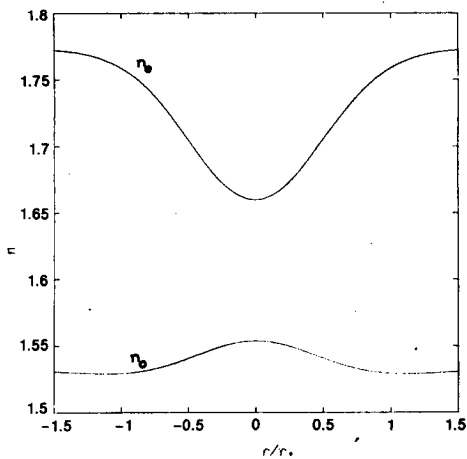


Fig. 1. Transverse distribution of the refractive indices in the liquid-crystal cell due to the laser-heating effect.

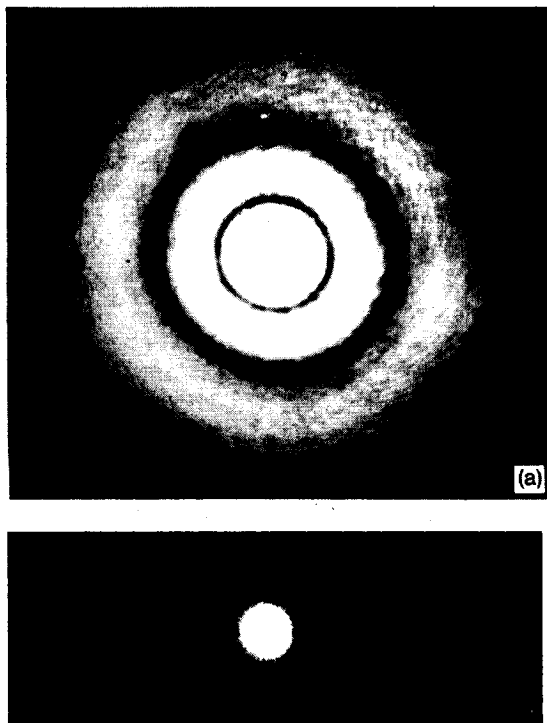


Fig. 2. Self-phase modulation (a) for extraordinary light and (b) for ordinary light. A parallel cell was used.

is formed in the liquid crystal owing to laser heating. The radius r_h of the hole can be obtained from Eq. (3):

$$2r_h^2/r_0^2 = \ln(T_0/T_c'). \tag{6}$$

The central temperature is equal to T_c or $T_0 = T_c'$ at the critical point. Since $T_0 = \alpha I_0$ and I_0 is proportional to the input intensity I , the relation between the radius of the isotropic hole and the input intensity is

$$2r_h^2/r_0^2 = \ln(I/I_c). \tag{7}$$

I_c is defined as T_c'/α , where α is determined below.

When the laser beam propagates through the small hole, Fraunhofer diffraction can be observed. A typical far-field diffraction pattern is shown in Fig. 3. For

higher-order diffraction rings, the separation between two neighboring rings approaches the value¹²

$$\Delta\theta = \lambda/2r_h. \tag{8}$$

This gives

$$r_h = \lambda/2\Delta\theta. \tag{9}$$

In the experiment the nematic director was parallel to the laser polarization. The size of the light spot on the liquid-crystal cell was about 100 μm . The diffraction patterns projected onto a screen were photographed with a camera after a 20-sec exposure time for each different input intensity. The radius of the small isotropic hole was calculated with Eq. (9). The results are shown in Fig. 4. The solid line is the least-squares fit with Eq. (7). We obtained $r_0 = 137 \mu\text{m}$ and $I_c = 1.28 \text{ W}$. The value of r_0 is larger than the radius of the spot of the laser beam. This is presumably due to the transverse heat diffusion and the laser heating of the glass plates of the liquid-crystal cell because of the longer exposure time.

We assumed in the above analysis that the temperature is linearly dependent on the input intensity. But the case will be different around the nematic-isotropic phase transition point. Bistability and hysteresis emerge in this case. To explain the observed bistable effect of the system, we assume that the proportion coefficient α after the N-I phase transition is larger

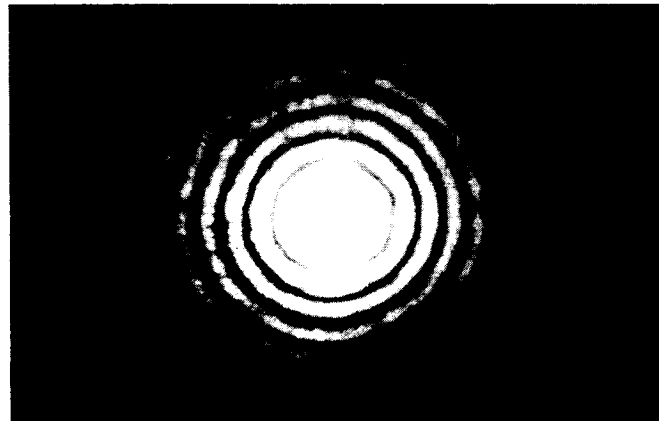


Fig. 3. Typical Fraunhofer diffraction pattern of the isotropic hole in a twisted cell.

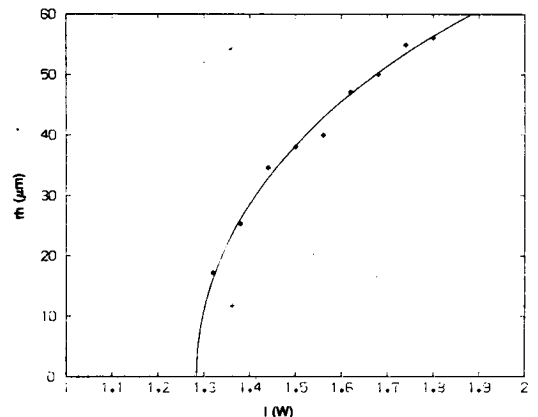


Fig. 4. Radius of the isotropic hole versus the input intensity. A parallel cell was used.

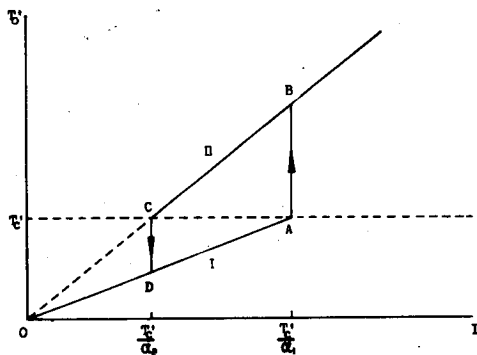


Fig. 5. Plot of the discontinuous change and hysteresis of the temperature T_0 versus the input intensity about the N-I phase transition point.

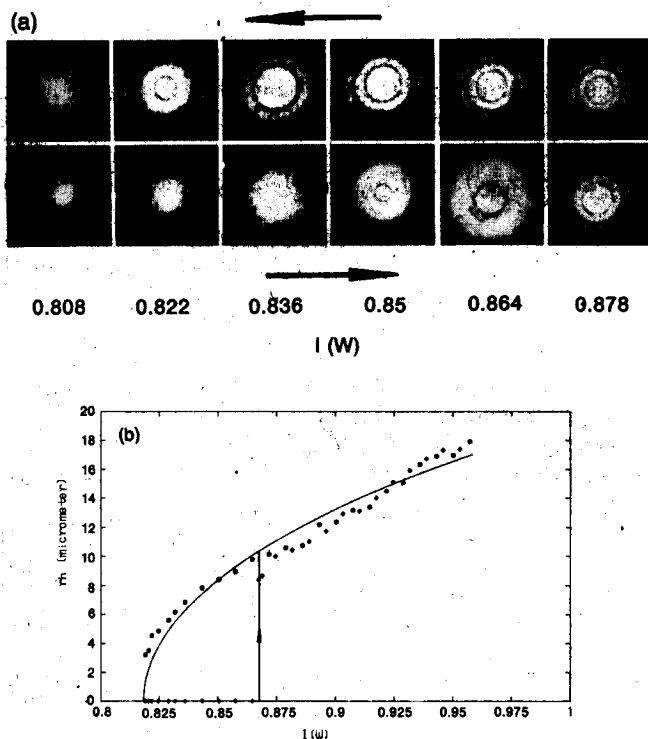


Fig. 6. Experimental results of the discontinuous change and hysteresis of (a) the diffraction pattern and (b) the radius of the isotropic hole. The arrows indicate the changing direction of the intensity. A twisted cell was used.

than that before the N-I phase transition. One important reason for this assumption is that the strong turbidity of the circular region around the isotropic hole will induce stronger absorption because of the strong scattering. Let α_1 and α_2 be the α values before and after the N-I phase transition, respectively. Then we will have $\alpha_2 > \alpha_1$. This can be written as

$$\alpha = \alpha_1 + (\alpha_2 - \alpha_1)H(T_0 - T_c), \quad (10)$$

where H is a Heaviside function.

A plot of the temperature T_0 versus the beam intensity is shown as Fig. 5. The slopes of line I and line II are α_1 and α_2 , respectively.

In the experiment we used a 90° twisted nematic liquid crystal as the sample in order to make the diffraction light of the isotropic hole clear. A polarizer was placed between the liquid crystal and the observing screen. The direction of the polarizer was parallel to the input-laser polarization. The nematic director in the front surface of the cell was perpendicular to the polarization of the input radiation in order to minimize the self-phase modulation. And, in this case, the polarization of the light transmitted through the twisted nematic liquid crystal was rotated perpendicularly to the polarizer so that the light field outside the isotropic hole ended up with its polarization at 90° with respect to the analyzer. Therefore the diffraction pattern could be observed clearly. The diffraction patterns were photographed continuously with a cinecamera. The round-trip changing time of the intensity was 25 sec. The room temperature was 17.5°C . The size of the light spot upon the liquid crystal was about $100\ \mu\text{m}$.

The experimental results are shown in Fig. 6. It can be seen from Fig. 6(a) that the diffraction rings appeared suddenly with increasing light intensity. The switching time was less than 0.1 sec. The size of the isotropic hole was calculated from the separation of rings with Eq. (9). The solid line in Fig. 6(b) is the least-squares fit of the experimental data in the decreasing direction of the intensity with Eq. (7), where $r_0 = 61\ \mu\text{m}$ and $I_c = 0.818\ \text{W}$. Letting the input intensity indicate I_0 , we obtained $\alpha_1 = 51.87^\circ\text{C/W}$ and $\alpha_2 = 54.99^\circ\text{C/W}$ from the jump-up intensity $I_c' = 0.867\ \text{W}$ and the intensity $I_c = 0.818\ \text{W}$, and then $\alpha_2 > \alpha_1$. The results are in agreement with the analysis.

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