Optical phase conjugation in a nematic liquid-crystal film modulated by a quasi-static electric field

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A quasi-static field can play an important role in molecular reorientation of the optical nonlinearity in liquid crystals because of the combination of the critical behavior of the sample at the Freedericksz transition and of nonlinear coupling of the optical and quasi-static fields. The nonlinear-optical phenomenon, optical phase conjugation, was observed in an electric-field-biased nematic liquid-crystal film and can be predicted by molecular reorientation calculated with continuum theory. The external field-modulated intensities of the phase-conjugation beams were obtained by both numerical calculation and experimental measurement. At the same time, the rise times of the intensities of phase-conjugation beams were measured for various external fields. © 1997 Optical Society of America [S0740-3224(97)02507-1]

1. INTRODUCTION

Previously we and others reported that in a biased quasistatic electric field degenerate four-wave mixing (DFWM) can be induced or enhanced dramatically when two coherent laser beams overlap in a nematic liquid-crystal film. The enhancement effects were attributed to the critical behavior of the sample at the Freedericksz transition. Our results showed that the first-order diffraction intensity is proportional to the cube of the laser intensity in the low-optical-field regime. 1,2 We have since found that the crucial factor influencing the first-order diffraction efficiency peak shift from the Freedericksz threshold voltage is twist deformation in the molecular reorientation grating. The first-order DFWM diffraction efficiency reaches its maximum at a biased ac voltage that depends on the elastic constant, the sample thickness, and the grating period.^{3,4} In other words, the diffraction efficiency can be modulated by the biased voltage. This is important in applications utilizing DFWM, such as optical phase conjugation (OPC). However, of the reports⁵⁻⁷ that involved the study of OPC in liquid crystals, in only one was the external biased discussed. Those researchers⁵ used a biasing magnetic field. In this paper we report using a molecular reorientation mechanism and a DFWM scheme to study OPC in a quasi-static electric-field-biased nematic liquid-crystal film. The intensity of the phaseconjugation beam can be modulated by the quasi-static electric field. This has been shown for various incident beam intensities by both numerical calculation and experimental measurement in planar-aligned nematic liquid-crystal films. At the same time, we measured the rise times of intensity of the phase-conjugation beam for various biased voltages at a fixed total incident laser power. The reflected beam, the phase conjugation beam, is also shown, by observation of phase-correction behavior with a cylindrical lens used as a phase aberrator, to be conjugate with the incident beam.

2. THEORY AND NUMERICAL RESULTS

A schematic diagram of the DFWM experimental apparatus and the geometry of a planar-aligned nematic liquidcrystal cell with thickness d are shown in Fig. 1. The nematic liquid crystal is assumed to have positive optical and dielectric anisotropies, namely, $n_e > n_o$ and ϵ_{\parallel} $> \epsilon_{\perp}$, where n and ϵ , respectively, denote the refractive indices and the dielectric constants and ϵ_{\parallel} and ϵ_{\perp} refer to the directions parallel and perpendicular, respectively, to the director \hat{n} . The incident laser beams have the same wavelength, λ . Pump beam I_2 and probe beam I_1 overlap in the nematic liquid-crystal film and intersect at a small angle α . They are nearly normally incident upon the cell and are linearly polarized in the y-axis direction. Another pump beam, I₃, propagates in a direction counter to that of beam I2 and has the same polarization. A quasistatic electric field (1 kHz) is applied perpendicular to the unperturbed molecular director \hat{n} . The optical field is superposed upon the applied electric field and induces the molecular reorientation that then gives rise to a spatially modulated refractive-index grating. Pump beam I3's first-order diffraction beam from this phase grating corresponds to phase-conjugation beam I4 in the nonlinear DFWM process.

Following the derivation in our previous studies, 3,4,8 we can obtain the local molecular orientation angle $\theta(x,z)$ with respect to the y axis by minimizing the total Frank free energy F, $F = \int_v \mathscr{F} \mathrm{d}v$. The Frank free-energy density \mathscr{F} is given by

$$\begin{split} \mathscr{F} &= \frac{1}{2} \left[K_{11} (1 - K \sin^2 \theta) \left(\frac{\partial \theta}{\partial z} \right)^2 + K_{22} \left(\frac{\partial \theta}{\partial x} \right)^2 \right] \\ &- \frac{D_z^2}{8 \pi \epsilon_{\perp} (1 - W \sin^2 \theta)} - \frac{I n_e}{c (1 - \mu \sin^2 \theta)^{1/2}}, \end{split}$$
(1)

where $K=1-K_{33}/K_{11}$, $W=1-\epsilon_{\parallel}/\epsilon_{\perp}$, $\mu=1-(n_e/n_o)^2$, D_z is the z component of the electric displacement, I is the optical intensity, c is the velocity of light in vacuum, and K_{11} , K_{22} , and K_{33} are the splay, twist, and bend elastic constants, respectively. Instead of solving for $\theta(x,z)$ by using the Euler–Lagrange equation, in the first-order approximation we assume that

$$\theta(x, z) = \{\theta_1 + \theta_2[\cos(2\pi x/\Lambda)]\}\sin(\pi z/d), \qquad (2)$$

with the boundary condition $\theta(z=0)=\theta(z=d)=0$. Here θ_1 corresponds to the spatial average reorientation angle, θ_2 is the amplitude of the grating modulation angle at z=d/2, and Λ is the grating period. We can calculate equilibrium values of constants θ_1 and θ_2 from the minimization of F by letting $\partial F/\partial\theta_1=0$ and $\partial F/\partial\theta_2=0$.

If the local reorientation angle θ is known, the effective refractive index $n_{\rm eff}(\theta)$ for a uniaxial medium can be expressed as

$$\begin{split} n_{\text{eff}}(\theta) &= n_e / [(1 - \mu \sin^2 \theta)]^{1/2} \\ &\cong \bar{n} + \Delta \bar{n}_{\text{NL}} \cos(2\pi x / \Lambda), \end{split} \tag{3}$$

where $\bar{n}=n_e+\mu n_e[1-J_0(2\,\theta_1)]/4$ is the spatially uniform refractive index, $\Delta\bar{n}_{\rm NL}=\mu n_e\theta_2J_0(2\,\theta_1)/2$ is the modulation index of the grating, and $J_0(2\,\theta_1)$ is the zero-order Bessel function. Consequently, the phase modulation experienced by a normally incident laser beam can be expressed as

$$\delta(x) \cong \delta_0 + \delta_1 \cos(2\pi x/\Lambda),$$
 (4)

where $\delta_0 \cong (\phi/2)[1-J_0(2\,\theta_1)]$, the first-order phase-modulation amplitude is $\delta_1 \cong \phi\,\theta_2 J_1(2\,\theta_1)$, and $\phi=\pi\mu n_e d/\lambda$. The intensity of phase-conjugation beam I_4 , which is diffracted from pump beam I_3 , is derived as

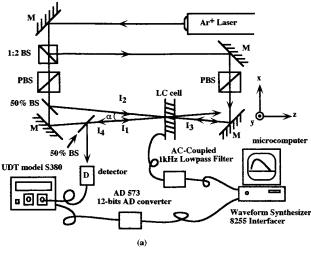
$$I_4 = I_3 J_1^2(\delta_1), (5)$$

and the phase-conjugation reflectivity R can be expressed as

$$R = I_4/I_1 = (I_3/I_1)J_1^2(\delta_1), \tag{6}$$

where $J_1(\delta_1)$ is a first-order Bessel function.

Numerical calculations were made for $\partial F/\partial \theta_1 = 0$, $\partial F/\partial \theta_2 = 0$, and relations (1)–(6). The parameters of the nematic liquid crystal E7 used⁹ are $n_e = 1.7464$, n_o $\begin{array}{l} = 1.5211, \;\; \mu = -0.31817, \;\; \epsilon_{\parallel} - \epsilon_{\perp} = 13.8, \;\; K = -0.54, \\ V_{\rm th} = 1.05 \; {\rm V}, \;\; I_{\rm th} = 335.15 \;\; {\rm W/cm^2}, \;\; {\rm and} \;\; K_{22}/K_{11} = 0.51. \end{array}$ The experimental parameters used are $\Lambda = 100 \, \mu \text{m}$, λ $d = 200 \, \mu \text{m}$ = 514.5 nm,and beam $I_1:I_2:I_3=1:2.84:3.90.$ The intensities of conjugation beam I4 versus the bias voltage were calculated for various total incident laser powers of I_1 , I_2 , and I_3 from 155.6 to 304.7 mW. The numerical results are shown in Fig. 2. It is obvious that the intensity of phase conjugation beam I4 can be modulated by a quasi-static electric field. There is an optimum biasing voltage for a fixed total incident laser power, and this optimum biasing voltage increases monotonically with the total incident laser power.



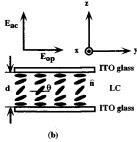


Fig. 1. DFWM geometry and a planar-aligned nematic liquid-crystal cell: LC, liquid crystal; BS's, beam splitters; PBS's, polarizing beam splitters; M's, mirrors; AD, analog to digital; UDT, united detector technology; θ , molecular reorientation angle; $E_{\rm op}$, optical field; $E_{\rm ac}$, quasi-static electric field; n, the molecular director, d, sample thickness. ITO, indium thin oxide.

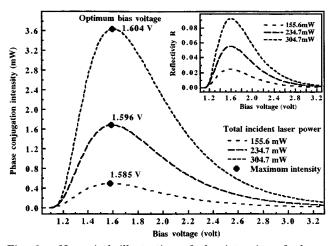


Fig. 2. Numerical illustration of the intensity of phase-conjugation beam I_4 versus bias voltage for various incident total laser powers. The beam ratio is $I_1:I_2:I_3=1:2.84:3.90$ The filled circles show the maximum intensity, and the corresponding voltages are the bias voltages.

3. EXPERIMENTAL RESULTS AND DISCUSSIONS

We prepared the liquid-crystal sample by sandwiching nematic *E*7 between two indium tin oxide-coated glass windows that had been treated with polyvinyl alcohol for planar alignment. A 1-kHz electric field generated by a

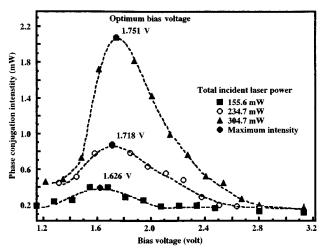


Fig. 3. Experimental results of measurement of the intensity of phase-conjugation beam I_4 versus bias voltage for various incident total laser powers. The beam ratio $I_1{:}I_2{:}I_3$ is approximately 1:2.84:3.90. The filled circles show the maximum intensity obtained by the least-mean-squares fitting method, and the corresponding voltages are the bias voltages. The dashed curves show the results from the least-mean-squares fitting calculation

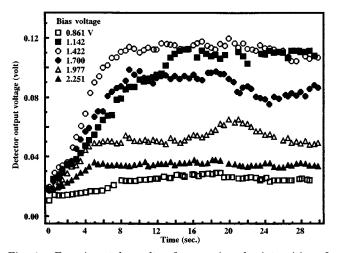


Fig. 4. Experimental results of measuring the intensities of phase-conjugation beam I_4 versus time for various bias voltages at a fixed total incident laser power of 269 mW.

microcomputer's waveform synthesizer (Quatech, Inc., WSB-A12M) was applied normally to the sample's glass windows. The laser light was separated into three beams, with beam ratio $I_1{:}I_2{:}I_3{=}1{:}2.84{:}3.90,$ by beam splitters and mirrors. Probe beam I_1 and pump beam I_2 recombined at the small intersection angle $\alpha~({\approx}5\times10^{-3}~\text{rad})$ on the cell; beams I_2 and I_3 were counterpropagating. The experimental measurements were carried out with a microcomputer. The intensity of phase-conjugation beam I_4 was recorded at various times while incident laser beams impinged upon the sample with a biased quasi-static electric field. The steady-state measurements were recorded while the samples were in equilibrium.

The intensities of phase-conjugation beam I_4 versus the bias voltage for various total incident laser powers is shown in Fig. 3. It is obvious that the intensity of beam

 $\rm I_4$ can be modulated significantly by the biasing voltage. There is also an optimum biasing voltage for a fixed total incident laser power. The maximum intensity biasing voltage, which was found by the least-mean-squares fitting method, increases monotonically with the total incident laser power, as predicted by numerical calculation.

The intensities of I_4 versus time for various biased voltages from 0.86 to 2.25 V at a fixed total incident laser power (269 mW) are shown in Fig. 4. Figure 5 shows the rise time, which is the time interval from 10% to 90% of maximum intensity, obtained by the least-mean-squares fitting method from Fig. 4. It is obvious that the rise time decreases with the biasing voltage. The result is the same as the prediction in Ref. 10.

Figure 6 is a schematic diagram of the experimental setup for confirming that beam I_4 is a phase-conjugation beam. When we observed the OPC reconstruction property, the cylindrical lens was used as an aberrator that changed the width of probe beam I_1 along the direction of the y axis. Phase-conjugation beam I_4 was split off by two 50% beam splitters at positions 1 and 2 then shone on

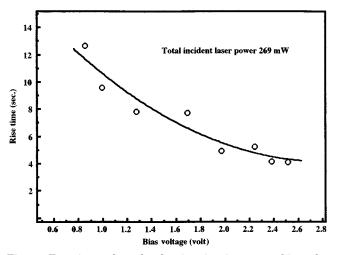


Fig. 5. Experimental results showing rise time versus bias voltage. The results were obtained by the least-mean-squares fitting method from Fig. 4. The solid curve is a guide to the eye.

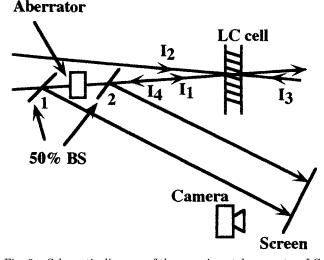


Fig. 6. Schematic diagram of the experimental apparatus: LC, liquid crystal; BS, beam splitter.

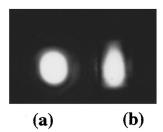




Fig. 7. (a) Optical pattern of probe beam I_1 . (b) Optical pattern of beam I_1 with a cylindrical lens in the way. (c) Optical pattern of phase-conjugation beam I_4 reflected onto the screen by a beam splitter at position 1 as phase-conjugation beam I_4 passes through the lens; (d) optical pattern of beam I_4 reflected onto the screen by the beam splitter at position 2. Positions 1 and 2, the locations of the beam splitters, are shown in Fig. 6.

the screen. Figure 7 shows the results of experimental observation. Figure 7(a) shows a circular optical pattern from probe beam I_1 at position 1 without the cylindrical lens in the way. With a cylindrical lens in the way, the optical pattern of beam I1 describes an elliptical shape at position 2, as shown in Fig. 7(b). The optical patterns of beam I₄ on the screen are shown in Figs. 7(c) and 7(d). Figure 7(d) shows the optical pattern from beam I₄, which was reflected onto the screen by the beam splitter at position 2. We found that this optical pattern had a generally elliptical shape, with the long axis in the same direction as that of beam I_1 in Fig. 7(b). Figure 7(c) shows the generally circular-shaped optical pattern of beam I₄, which was reflected onto the screen by the beam splitter at position 1. It also shows that the aberration of incident beam I₁ that was caused by the cylindrical lens was reconstructed as reflected beam I₄ passed through the aberrator. In other words, it is shown unambiguously that reflected beam I₄ is conjugate to beam I₁.

4. CONCLUSIONS

We have demonstrated unambiguously the nonlinear-optical phase-conjugation phenomenon in a nematic liquid-crystal film by examining the reconstruction properties of the phase-conjugation beam. Molecular reorientation is the mechanism for this phenomenon and can be predicted by continuum theory. We found that a biasing electric field not only can induce but can also modulate the conjugation beam with respect to the biasing voltage, which agrees with the calculation prediction very well. At the same time, we found that the rise time of the phase-conjugation beam intensity decreases with increasing biased electric field.

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