Quasi-static electric-field-enhanced degenerate four-wave mixing in a nematic liquid-crystal film

Shu-Hsia Chen, C.-L. Kuo, and Ming-Chih Lee

Institute of Electro-Optical Engineering, National Chiao Tung University, Hsinchu, Taiwan 30050, China

Received June 20, 1988; accepted November 5, 1988

Degenerate four-wave mixing can be induced or enhanced dramatically when two weak overlapping laser beams are incident upon a nematic liquid-crystal film that is biased by a quasi-static electric field. This phenomenon is shown to be the result of the critical behavior at the Freedericksz transition. The experimental results show that the diffracted intensity is proportional to the third power of the laser intensity, as expected for a four-wave mixing process, despite the strength of the electric-field bias voltage above threshold.

Nonlinear optical processes in liquid crystalline media have received considerable attention recently.[1-4] One interesting application is the phase grating,[5-7] which is made possible by the molecular reorientation that contributes large optical nonlinearity in liquid crystals. It was suggested by Herman and Serinko[5] that a phase grating can be obtained easily in a nematic liquid-crystal film if one uses a dc field to bias it near the critical orientational Freedericksz transition (FT). Fuh *et al.*[8] reported a three-dimensional model and experimental results of purely optically induced phase gratings. Khoo and Zhuang[9] and Khoo[10,11] reported observations of degenerate four-wave mixing in a nematic liquid-crystal film with and without a biased magnetic field.

We use the molecular reorientation mechanism in a degenerate four-wave mixing (DFWM) scheme to obtain the dependence of the diffracted beam's intensity on the incident beam's intensity and the strength of the electric-field bias. The DFWM is dramatically enhanced by the coupling of the electric field to the optical field owing to the critical behavior of the FT. The results of detailed measurements show that, for weak laser beams (<1.6 W/cm^2), the diffracted intensity is proportional to the third power of the incident intensity. This is expected for a DFWM process, in spite of the strength of the biased voltage above threshold. There is also a peak diffracted intensity near the FT voltage as predicted by theory.

We consider a homeotropically aligned nematic liquid-crystal film having the geometry shown in Fig. 1. Following the derivations by Herman and Serinko,[5] we minimize the free energy with the variational principle and obtain an approximate solution to the sine–Gordon equation for a small molecular reorientation angle given by

$$\theta(x, z) = \theta_M(x)\sin(\pi z/d), \tag{1}$$

where $\theta_M(x)$ is the maximum reorientation angle and is given by

$$\theta_M(x) = \begin{cases} 0 & E_{\text{eff}}(x) \le E_c \\ 2[(E_{\text{eff}}(x) - E_c)/E_c]^{1/2} & E_{\text{eff}}(x) \ge E_c \end{cases}. \tag{2}$$

E_{eff} and E_c are the effective and critical fields, respectively, and are given by

$$E_{\text{eff}}^2(x) = E_{\text{op}}^2(x)[2\Delta\epsilon_{\text{op}}/|\Delta\epsilon_{\text{dc}}|] + E_{\text{dc}}^2 \tag{3}$$

and

$$E_c = \frac{\pi}{d}\left|\frac{4\pi k}{|\Delta\epsilon_{\text{dc}}|}\right|^{1/2}. \tag{4}$$

In these expressions x and z denote the transverse and longitudinal coordinates, respectively, inside the liquid-crystal film of thickness d, k is the elastic constant, E_{dc} is the quasi-static electric-field bias, and $\Delta\epsilon_{\text{op}}$ and $\Delta\epsilon_{\text{dc}}$ are the dielectric anisotropies of the optical and electric fields, respectively. When two mutually coherent laser beams (with intensities $I_1 = I_2 = I_0$) having a small intersection angle α overlap and are normally incident upon the surface of an electric-field-

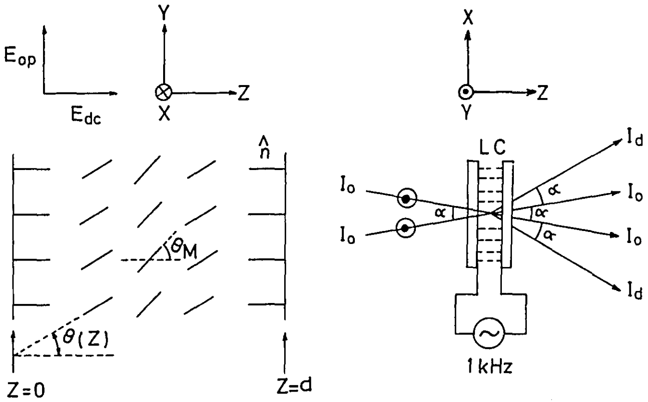

Fig. 1. Experimental geometry of the nematic liquid-crystal film. LC, liquid crystal.

biased nematic liquid-crystal film, a sinusoidally varying intensity pattern is produced in the sample. The optical field is coupled to the biasing electric field and induces molecular reorientation, which then gives rise to a spatially modulated refractive-index grating as long as $E_{\mathrm{eff}} > E_c$. The grating period is given by $\Lambda = \lambda/[2\sin(\alpha/2)]$, where λ is the optical wavelength. The corresponding θ_M at the optical interference peak is given by

$$\theta_{\mathrm{MP}} = 2\left[\left(\frac{8\Delta\epsilon_{\mathrm{op}}}{|\Delta\epsilon_{\mathrm{dc}}|}\frac{E_0^{\,2}}{E_c^{\,2}} + \frac{E_{\mathrm{dc}}^{\,2}}{E_c^{\,2}}\right)^{1/2} - 1\right]^{1/2}, \qquad (5)$$

where E_0 is the field amplitude of each incident laser beam. The phase modulation $\delta(x)$ experienced by a normally incident laser beam should be considered in two regimes. When $E_{\mathrm{dc}} \geq E_c$, $\delta(x)$ is a well-behaved sinusoidally varying function of x. On the other hand, when $E_{\mathrm{dc}} < E_c$, $\delta(x)$ is a periodic function with a flat segment instead of a minimum point in each period. In this case, it is reasonable to approximate $\delta(x)$ as a sinusoidal function if E_{dc} is close to E_c. Therefore, for $|E_{\mathrm{dc}} - E_c|/E_c \ll 1$, the amplitude of the phase modulation, δ_0, can be expressed as

$$\delta_0 = \begin{cases} \dfrac{4\pi n_o d(\Delta\epsilon_{\mathrm{op}})^2 E_0^{\,2}}{\lambda n_e^{\,2}|\Delta\epsilon_{\mathrm{dc}}|E_{\mathrm{dc}}E_c} & E_{\mathrm{dc}} \geq E_c \\ \dfrac{\pi n_o \Delta\epsilon_{\mathrm{op}} d}{\lambda n_e^{\,2}}\left[\left(\dfrac{8\Delta\epsilon_{\mathrm{op}}}{\Delta\epsilon_{\mathrm{dc}}}\dfrac{E_0^{\,2}}{E_c^{\,2}} + \dfrac{E_{\mathrm{dc}}^{\,2}}{E_c^{\,2}}\right)^{1/2} - 1\right] & E_{\mathrm{dc}} \leq E_c \end{cases}, \qquad (6)$$

where n_o and n_e are, respectively, the ordinary and maximum extraordinary refractive indices of the sample. It is obvious from the above equations that for weak laser beams, the phase grating can be induced only by a large E_{dc}, namely, near E_c. The enhancement increases with increasing E_{dc} when $E_{\mathrm{dc}} \leq E_c$, and then decreases when $E_{\mathrm{dc}} \geq E_c$. Consequently, the intensity of the diffracted beam can be given by

$$I_d = \begin{cases} \dfrac{64\pi^4 d^2(\Delta\epsilon_{\mathrm{op}})^4}{c^2\lambda^2 n_e^{\,4}|\Delta\epsilon_{\mathrm{dc}}|^2 E_{\mathrm{dc}}^{\,2}E_c^{\,2}} I_0^{\,3} & E_{\mathrm{dc}} \geq E_c \\ \dfrac{\pi^2 n_o^{\,2}\Delta\epsilon_{\mathrm{op}}^{\,2} d^2}{4\lambda^2 n_e^{\,4}}\left[\left(\dfrac{32\pi\Delta\epsilon_{\mathrm{op}}}{n_o c|\Delta\epsilon_{\mathrm{dc}}|E_c^{\,2}} I_0 + \dfrac{E_{\mathrm{dc}}^{\,2}}{E_c^{\,2}}\right)^{1/2} - 1\right]^2 I_0 & E_{\mathrm{dc}} < E_c \end{cases}, \qquad (7)$$

where c is the velocity of light in a vacuum. From Eqs. (6) and (7) one can see that the effect of diffraction enhancement due to the electric-field bias is identical to the effect of the phase grating.

The sample preparation was essentially the same as that described in Ref. 12. The nematic liquid crystal *p*-methoxybenzylidene-*p*-*n*-butylaniline was sandwiched between two glass slides that were first coated with indium–tin–oxide as transparent electrodes and then treated with octadecyldimethyl(3-trimethoxysilyl-propyl)ammonium chloride for homeotropic alignment. The sample film was 75 μm thick as determined by a calibrated Mylar spacer and was kept at 26°C to avoid any thermal effects. Since $\Delta\epsilon_{\mathrm{dc}}$ is negative at low frequencies, a quasi-static electric field (1 kHz) was applied along the unperturbed molecular direction to achieve the enhancement effect. A Spectra-Physics 2020 Ar$^+$ laser (514.5 nm) was used. A small fraction (~8%) of its output intensity was split off as the reference beam. A 50% beam splitter separated the rest of the output beam into two equally intense beams that were directed onto the same spot (~1.6 mm in diameter as measured to e^{-2} intensity) on the sample. To induce the phase grating and achieve the diffraction effect, the beams were normally incident to the film with a small crossing angle ($\alpha \approx 0.4°$). Both the reference and diffracted intensities were detected by photocells (Hamamatsu S1223-01) so that the proper normalization could be made to correct for any power drift or fluctuation.

Because background scattering is significant and increases with increasing pump intensity, subtraction of it is necessary to obtain the true diffracted intensity. We have verified that the intensity of background scattering is additive and linearly proportional to that of the incident beam. Therefore the background scattering at the diffraction spot when two pumps are simultaneously turned on can be taken as the sum of the scattered intensity for both beams. This background contribution has been subtracted from the diffracted intensity in our experimental results. To determine the FT critical field, the electrocontrolled birefringence of our samples was measured with a He–Ne laser by the modulation technique originally devised by Lim and Ho.[13]

The experimentally measured electrocontrolled birefringence and the intensity (I_d) of the diffracted beam for a weak ($I_0 = 1$ W/cm^2) incident laser versus the electric-field bias voltage (V_{dc}) of our sample are shown in Fig. 2. With the laser beam on by itself, no diffraction was detected, which ensures that the thermal grating is not significant in this experiment. The electrocontrolled birefringence curve shows a typical critical behavior near the FT critical voltage (field) when $\mathrm{d}\theta/\mathrm{d}E_{\mathrm{dc}}$ is large. The curve reaches its maximum when $V_c(E_c) = 3.7$ V. From the diffracted intensity curve, one can easily see the enhancement effect. There is no significant diffraction intensity in the low-voltage (field) regime, i.e., $V_{\mathrm{dc}} \ll V_c$ ($E_{\mathrm{dc}} \ll E_c$). However, I_d appears near the FT critical voltage, corresponding to the induced DFWM. It increases dramatically and reaches its maximum at 4.38 V, illustrating the strong enhancement of DFWM due to the critical behavior of the FT. This enhancement

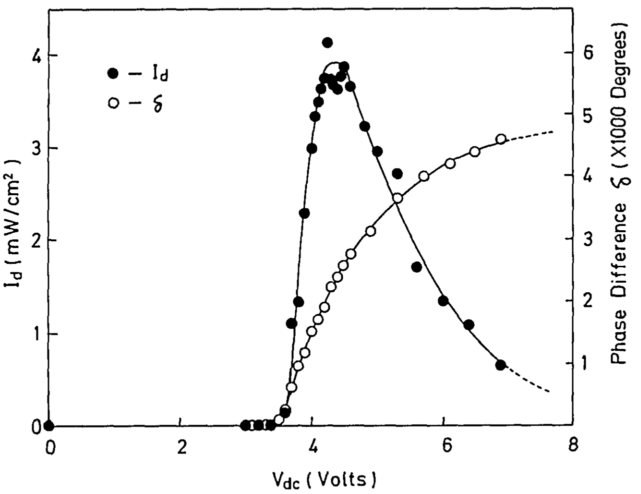

Fig. 2. Quasi-static electric-field-induced birefringence and the diffracted intensity versus the electric field for an incident laser at 1 W/cm². The solid lines are to aid in visualization of the data.

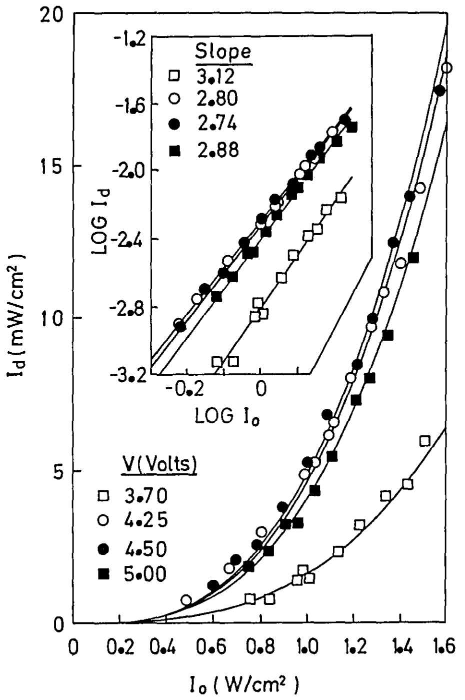

Fig. 3. Diffracted intensity versus the incident intensity for various field strengths. The solid curves are fit to the cubic dependence. The inset shows $I_d \propto I_0^3$ as predicted by Eq. (7).

effect then decreases with further increases in the electric-field bias voltage, E_{dc} (V_{dc}), as predicted by Eq. (7). The diffraction curve eventually decays as V_{dc} enters the saturation regime of the electrocontrolled birefringence curve.

According to our theory, the diffracted intensity is proportional to the square of the amplitude of the phase modulation. Therefore, a peak must occur at the FT field (voltage), E_c (V_c), where the phase modulation reaches its maximum. From our experimental results, however, the maximum phase modulation occurs at 3.7 V, while the peak diffracted intensity occurs at 4.38 V. The difference is likely due to our assumption that the finite beam size effect can be neglected.[14] In other words, the twisting effect[15] could be significant owing to the small width of the grating period (~73 μm) since the polarization of the optical field is perpendicular to the grating direction.

The diffracted intensity versus the incident beam intensity for various external voltages is shown in Fig. 3. The experimental data follow the DFWM cubic dependence well for all four chosen voltages. This is shown more clearly in the inset of Fig. 3, in which the slopes are close to 3. Although Eq. (7) predicts only the cubic dependence that characterizes the DFWM process near the FT critical voltage for weak laser beams, it is true for any bias voltage that induces nonsaturating molecular reorientation, provided that the intensity of the incident beam is weak enough.

In conclusion, we have shown both theoretically and experimentally that the DFWM can be induced or enhanced dramatically by a quasi-static electric field with two overlapping normally incident weak laser beams on a nematic liquid-crystal film. This is due to the critical behavior of FT. The cubic dependence characterizing the DFWM process is obtained for $I_0 <$ 1.6 W/cm², independent of the bias voltage. The I_d-versus-V_{dc} curve shows a peak at $V_{dc} = 4.38$ V rather than at $V_c = 3.7$ V, as predicted by our theory, where $d\theta/dE_{dc}$ is maximum. We attribute the difference to the simplification in our calculation.

This research was supported partially by the Chinese National Science Council under contract NSC-74-0608-E009-04R.

References

1. S. D. Durbin, S. M. Arakelian, M. M. Cheung, and Y. R. Shen, J. Phys. **44**, 161 (1983).
2. N. V. Tabiryan and B. Ya. Zel'dovich, Mol. Cryst. Liq. Cryst. **62**, 237 (1980).
3. I. C. Khoo and R. Normandin, Opt. Lett. **9**, 285 (1984).
4. S.-H. Chen and J. J. Wu, Appl. Phys. Lett. **52**, 1998 (1988).
5. R. M. Herman and R. J. Serinko, Phys. Rev. A **19**, 1757 (1979).
6. I. C. Khoo, Phys. Rev. A **27**, 2747 (1983).
7. S. D. Durbin, S. M. Arakelian, and Y. R. Shen, Opt. Lett. **7**, 145 (1982).
8. Y. G. Fuh, R. F. Code, and G. X. Xu, J. Appl. Phys. **54**, 6368 (1983).
9. I. C. Khoo and S. L. Zhuang, Appl. Phys. Lett. **37**, 3 (1980).
10. I. C. Khoo, Appl. Phys. Lett. **38**, 123 (1981).
11. I. C. Khoo, Phys. Rev. A **23**, 2077 (1981).
12. C.-L. Pan, S.-H. Chen, and H.-H. Liao, Phys. Rev. A **33**, 14312 (1986).
13. K. C. Lim and J. T. Ho, Mol. Cryst. Liq. Cryst. **47**, 173 (1978).
14. I. C. Khoo, T. H. Liu, and P. Y. Yan, J. Opt. Soc. Am. B **4**, 115 (1987).
15. I. C. Khoo, Phys. Rev. A **27**, 2747 (1983).