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Optical phase grating diffraction in a quasistatic electric field biased nematic liquid crystal film

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It is known that an optical phase grating can be obtained when two mutually coherent laser beams overlap in a nematic liquid crystal. This is mainly due to director reorientation which contributes to a large optical non-linearity. It has been suggested by Herman and Serinko that a phase grating could be obtained in nematic liquid crystals if a D.C. field is used to bias it near the critical orientational Freedericksz transition. A homeotropic MBBA film biased by an electric field at 1 kHz has been studied. Two weak Ar^+ laser beams were incident normally to the film with a small intersection angle ($\approx 0{\cdot}4°$). Using the picture of a director reorientation mechanism and a degenerate four wave mixing scheme, we have obtained the dependence of the diffraction beam intensity on that of the incident beam and the strength of the biased electric field. The theoretical prediction and experimental results both show that the phase grating diffraction can be dramatically enhanced by the coupling of the electric field to the optical field in the Freedericksz transition region. This is due to the critical behaviour of the sample at that transition. The prominently improved signal-to-noise ratio is discussed.

1. Introduction

In recent years, non-linear optical effects based on optical field-induced refractive index changes have attracted considerable attention. Because of their high molecular anisotropy, liquid crystal media exhibit very large optical non-linearities [1]. The optical field-induced Freedericksz transition and associated non-linear optical effects in a nematic liquid crystal film such as self-focusing [2–5], self-phase modulation [6–8] and optical bistability [9–16] have been studied extensively. In particular, interesting highly non-linear optical effects, e.g. phase grating diffraction [17–23], have been observed. It has been suggested by Herman and Serinko [23] that the phase grating can be easily obtained for two intersecting normally incident coherent laser beams in a nematic film if a D.C. field is used to bias it near the critical orientational Freedericksz transition. The director reorientation attributing phase grating diffraction aided by a static magnetic field have been observed by Khoo [24] and Shen's [22] group, separately. In our previous work [25], it has been shown theoretically by a simple physical model and verified by experiment that the degenerate four-wave mixing can be enhanced dramatically by a quasistatic electric field due to the critical behaviour of the sample at the Freedericksz transition. In this paper we present result of a detailed investigation of crossed-beam experiments with nematic films. In particular, we have measured the diffraction from the phase grating generated by the crossed beams as a function of the optical and the electric fields. In § 2 we state the essential results from an approximate theory for field-induced director reorientation and phase grating diffraction. In §3 the experimental procedures are described. In §4

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the experimental results are summarized and shown to be in good agreement with the theoretical predictions. In addition, detailed studies of the background scattering are reported. The prominent improvement of the signal-to-noise ratio by the electric field is also discussed in the view of its potential applications.

2. Theory

Our derivation, based on a continuum model of the liquid crystal, is essentially the same as that by Herman and Serinko [23]. For simplicity, a homeotropically aligned nematic film of thickness d is considered. Its unperturbed director $\hat{\mathbf{n}}_0$ is along the z axis; this is illustrated in figure 1. The normally incident light beams are in the (x, z) plane with the polarization along the y axis. The quasistatic electric field is along the z axis. Under the action of the superimposed fields, the local director $\hat{\mathbf{n}}(\mathbf{r})$ will attain a director reorientation angle θ, i.e., the angle between $\hat{\mathbf{n}}(\mathbf{r})$ and $\hat{\mathbf{z}}$ at (x, z). Thus $n_x = 0$, $n_y = \sin\theta(x, z)$ and $n_z = \cos\theta(x, z)$. The angle $\theta(x, z)$ can be calculated by minimizing the total free energy of the system, $F = \int \mathscr{F}\, dv$. The free energy density $\mathscr{F}$ is given by

$$\begin{aligned} &\frac{K}{2}\{[\nabla \cdot \hat{\mathbf{n}}(\mathbf{r})]^2 + [\nabla \times \hat{\mathbf{n}}(\mathbf{r})]^2\} - \frac{\Delta\varepsilon_{dc}}{8\pi}[\mathbf{E}_{dc} \cdot \hat{\mathbf{n}}(\mathbf{r})]^2 \\ &\qquad - \frac{\Delta\varepsilon_{op}}{4\pi}[\mathbf{E}_{op} \cdot \hat{\mathbf{n}}(\mathbf{r})]^2 \\ &= \frac{K}{2}\left(\frac{d\theta}{dz}\right)^2 - \frac{\Delta\varepsilon_{dc}}{8\pi}E_{dc}^2\cos^2\theta - \frac{\Delta\varepsilon_{op}}{4\pi}E_{op}^2\sin^2\theta. \end{aligned} \tag{1}$$

In this equation, K is the elastic constant in the so-called one constant approximation [4]. E_{dc} and E_{op} are the quasistatic electric and optical fields inside the sample; $\Delta\varepsilon_{dc}$ and $\Delta\varepsilon_{op}$ are the dielectric anisotropies, $\Delta\varepsilon = \varepsilon_{\parallel} - \varepsilon_{\perp}$, due to the quasistatic electric and the optical field; and $\varepsilon_{\parallel}$ and $\varepsilon_{\perp}$ are the dielectric constants parallel and perpendicular to the director, respectively.

Application of the Euler–Lagrange equation yields the familiar sine–Gordon equation for the torque balance,

$$\xi^2\frac{d^2\theta}{dz^2} + \sin\theta\cos\theta = 0, \tag{2}$$

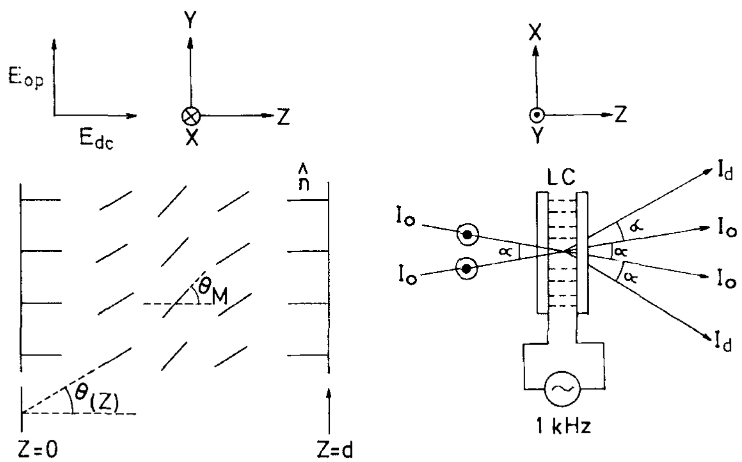

Figure 1. Experimental geometry.

where

$$\xi^2 = \frac{4\pi K}{|\Delta\varepsilon_{dc}|E_{eff}^2} \quad \text{and} \quad E_{eff}^2 = E_{dc}^2 - \frac{2\Delta\varepsilon_{op}}{\Delta\varepsilon_{dc}} E_{op}^2. \tag{3}$$

With the boundary conditions $\theta = 0$ at $z = 0$ and $z = d$, in the region $0 < z < d/2$ equation (2) yields

$$z\frac{\sin\theta_m(x)}{\xi} = \int_0^{\theta(x,z)} \left[1 - \frac{\sin^2\theta'}{\sin^2\theta_m(x)}\right]^{1/2} d\theta'. \tag{4}$$

The value of $\theta_m(x)$ is obtained by equating θ to θ_m at $z = d/2$ in this equation. Typically, numerical integration of equation (4) is required to solve for $\theta(x, z)$. The effect of superimposed quasistatic electric and optical fields on the director reorientation is most evident by examining equation (2) for small reorientation angles, i.e. $\theta \ll 1$. In this limit, we have

$$\theta(x, z) = \theta_m(x)\sin(\pi z/d) \tag{5}$$

and

$$\theta_m(x) = 2\{[E_{eff}(x) - E_c]/E_c\}^{1/2} \quad \text{for} \quad 1 \gg \frac{E_{eff}}{E_c} - 1 > 0 \tag{6}$$

where

$$E_c = \frac{\pi}{d}\left[\frac{4\pi K}{|\Delta\varepsilon_{dc}|}\right]^{1/2}$$

is the critical electric field for the Freedericksz transitions.

When two mutually coherent laser beams with wavevectors $\mathbf{k}_1$, $\mathbf{k}_2$, and equal intensities ($I_1 = I_2 = I_0$), overlapped with a small intersection angle α, incident normally on the sample film, the interference of these two laser beams produces a sinusoidally varying intensity pattern in the sample. This optical field is superimposed on the applied electric field and induces the director reorientation which then gives rise to a spatially modulated refractive index grating provided $E_{eff} > E_c$. The resulting optical field is given by

$$E_{op}(x)^2 = 2E_0^2[1 + \cos(k_x x)], \tag{7}$$

where E_0 is the electric field amplitude of one beam;

$$k_x = |k_2 - k_1| = 2\pi/\Lambda; \; \Lambda = \lambda/[2\sin\alpha/2)]$$

is the grating period and λ is the optical wavelength. The corresponding θ_m at the optical interference peak is

$$\theta_{mp} = 2\left\{\left[\frac{8\Delta\varepsilon_{op}}{|\Delta\varepsilon_{dc}|}\frac{E_0^2}{E_c^2} + \frac{E_{dc}^2}{E_c^2}\right]^{1/2} - 1\right\}^{1/2}, \tag{8}$$

for $\theta(x, z) \ll 1$, the laser beam should experience an induced phase difference

$$\delta(x) = \frac{2\pi}{\lambda}\frac{n_0\Delta\varepsilon_{op}}{2n_e^2}\int_0^d \theta^2(x, z)dz, \tag{9}$$

where n_0 and n_e are, respectively, the ordinary and maximum extraordinary refractive indices of the sample. The phase grating should be considered in two regimes. For

$E_{dc} > E_c$. $\delta(x)$ is a well behaved sinusoidally varying function of x. On the other hand, for $E_{dc} < E_c$, $\delta(x)$ 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 very close to E_c. Therefore from equation (5), in the limit of $|E_{dc} - E_c|/E_c \ll 1$, the phase shift can be written as

$$\delta(x) = A + \delta_0\{1 + \tfrac{1}{2}[(\exp(ik_x x) + \exp(-ik_x x))]\}, \qquad (10)$$

where A is a constant independent of x and δ_0 is the amplitude of the phase modulation. Form equation (6) we have

$$\delta_0 = \begin{cases} \dfrac{4\pi n_0 d(\Delta\varepsilon_{op})^2 E_0^2}{\lambda n_e^2 |\Delta\varepsilon_{dc}| E_{dc} E_c}, & E_{dc} \geqslant E_c \\ \dfrac{\pi n_0 \Delta\varepsilon_{op} d}{\lambda n_e^2}\left[\left(\dfrac{8\Delta\varepsilon_{op}}{\Delta\varepsilon_{dc}}\dfrac{E_0^2}{E_c^2} + \dfrac{E_{dc}^2}{E_c^2}\right)^{1/2} - 1\right], & E_{dc} \leqslant E_c. \end{cases} \qquad (11)$$

It is obvious from these equations that for weak laser beams, the phase grating can be induced only by large E_{dc}, namely near E_c. The enhancement of the phase grating by the applied electric field increases and then decreases with increasing E_{dc} for $E_{dc} \leqslant E_c$ and $E_{dc} \geqslant E_c$, respectively.

The wavefront of the plane wave, $\mathbf{E}_2$, propagating through the sample film can be written as

$$E_0 \exp\{i[-\mathbf{k}_2 \cdot \mathbf{r} - \delta(x)]\} = \exp[-i(A + \delta_0)]\{E_0 \exp[-i\mathbf{k}_2 \cdot \mathbf{r}] - i\frac{E_0\delta_0}{2}\exp[-i(\mathbf{k}_2 + \mathbf{k}_x)\cdot\mathbf{r}] - i\frac{E_0\delta_0}{2}\exp[-i(\mathbf{k}_2 - \mathbf{k}_x)\cdot\mathbf{r}]\}, \qquad (12)$$

those terms on right hand side represent the transmission beam, the first order diffraction beam and one other diffraction beam coincide with the transmission beam of $\mathbf{E}_1$, respectively. Consequently, the intensity of the diffraction beam can be expressed as

$$I_d = \begin{cases} \dfrac{64\pi^4 d^2 (\Delta\varepsilon_{op})^4}{c^2\lambda^2 n_e^4 |\Delta\varepsilon_{dc}|^2 E_{dc}^2 E_c^2} I_0^3, & E_{dc} \geqslant E_c \\ \dfrac{\pi^2 n_0^2 \Delta\varepsilon_{op}^2 d^2}{4\lambda^2 n_e^4}\left\{\left[\dfrac{32\pi\Delta\varepsilon_{op}}{n_0 c|\Delta\varepsilon_{dc}|E_c^2} I_0 + \dfrac{E_{dc}^2}{E_c^2}\right]^{1/2} - 1\right\}^2 I_0, & E_{dc} < E_c \end{cases} \qquad (13)$$

where c is the velocity of light in vacuum. We can see that the diffraction enhancement effect due to the biased electric field is the same as the behaviour of the phase grating.

3. Experimental

The liquid crystal used was 4-methoxybenzylidene-4′-*n*-butylaniline (MBBA). The sample was prepared by sandwiching the nematic between two glass windows which were coated first with indium-tin oxide as transparent electrodes and then treated with octadecyldimethyl [3-trimethoxysilyl-propyl] ammonium chloride (DMOAP) for homeotropic alignment. The alignment was checked with conoscopy. The two samples used were 75 μm thick, as determined by the calibrated mylar spacer. They were kept at 26°C to avoid thermal effects during the measurement.

The experimental setup is shown in figure 2. A quasistatic electric field at 1 kHz was applied normal to the glass windows. A single line (514·5 nm) argon laser was used; a small fraction ($\approx$ 8 per cent) of its output intensity was split off as the reference beam. The rest of the output beam was separated into two equally intense beams by a 50 per cent beam splitter and recombined at a small crossing angle α ($\approx 0{\cdot}4°$) in the sample. The diameter of the overlapped beams was 1·6 mm as measured to e^{-2} intensity. Both the reference and diffraction intensities were detected by photodiodes so that normalization was made to correct for any power drift or fluctuation. The background scattering with respect to the pump laser intensity was measured at a neighbouring point of the diffraction spot first with one pump beam on only and then both. The background contribution was subtracted from the diffraction intensity in our results. To see the Freedericksz transition behaviour the electrocontrolled birefringence of our samples was measured with a He–Ne probe laser by using a modulation technique orginally devised by Lim and Ho [26]. The diffraction intensity vesus the applied voltage was measured at two incident beam intensities, namely 1·0 and 3·85 W/cm^2. Two sets of data were taken for the intensity dependence study. One is in the weak beam regime ($<$ 1·6 W/cm^2) to show the cubic dependence characteristic of a degenerate four-wave mixing process. The other is in the higher intensity regime ($<$ 4 W/cm^2).

4. Results and discussion

The background scattering with both pump beams on, at a neighbouring point of the diffraction spot is shown in figure 3. It is obvious that the background scattering is linearly dependent on the incident intensity of the pump beam. In figure 4, the background scattering of both beams on versus the sum of that of each beam on alone is plotted. We can see the resultant data are very close to a straight line with slope of one. In other words, we have verified that the intensity of the background scattering is linearly proportional to that of the incident beam and that it is additive. Therefore

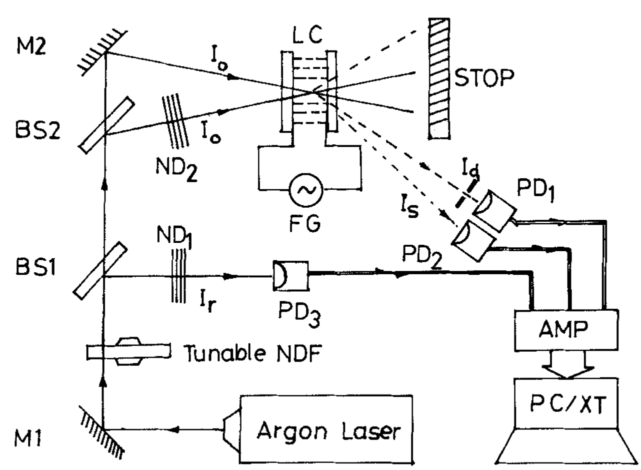

Figure 2. Experimental setup. NDF, neutral density filter; BS, beam splitter, PD, photodiode; M, mirror; FG, function generator.

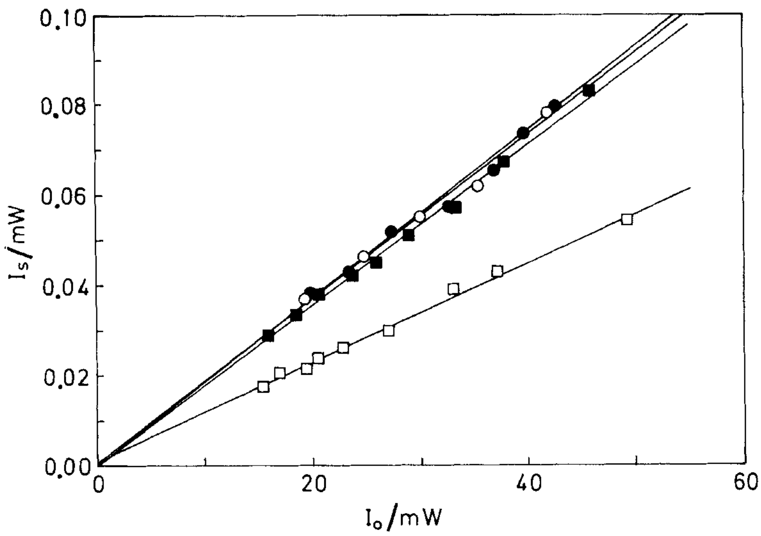

Figure 3. Intensity of background scattering (I_s) versus incident laser intensity (I_0) with both beams on for various voltages; □ = 3·70 V, ○ = 4·25 V, ● = 4·50 V and ■ = 5·00 V.

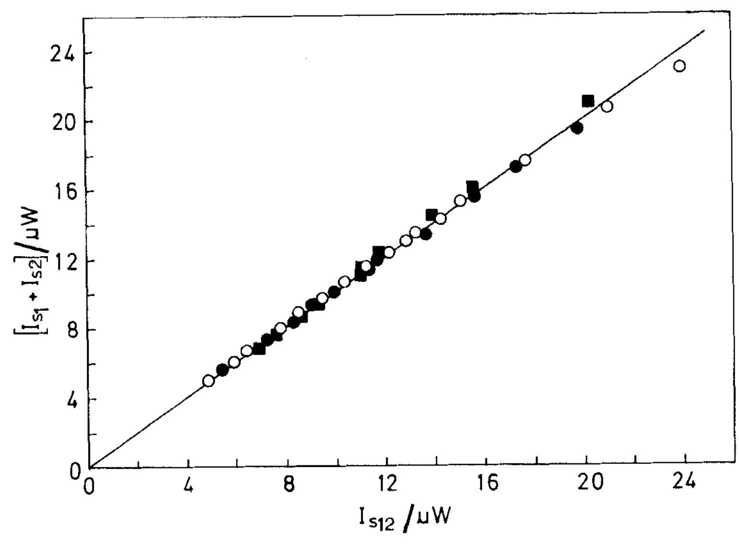

Figure 4. The collective background scattering (I_{s12}) versus the sum of individual background scattering ($I_{s1} + I_{s2}$) shows that the background scattering is additive. ○ indicates a voltage of 4·25 V, ● 4·50 V and ■ 5·00 V.

the intensity of the background scattering at the diffraction spot, when two pumps are turned on simultaneously, can be taken as the sum of the scattering intensities of the two individual beams. The scattering due to the beams was measured at the diffraction spot as a function of the incident intensity. This was used to correct the diffraction intensity in our experimental results.

The measured electrocontrolled birefringence and the diffraction intensity (I_d) for a weak (I_0 = 1 W/cm^2) incident laser versus the biased voltage (V_{dc}) of our sample are

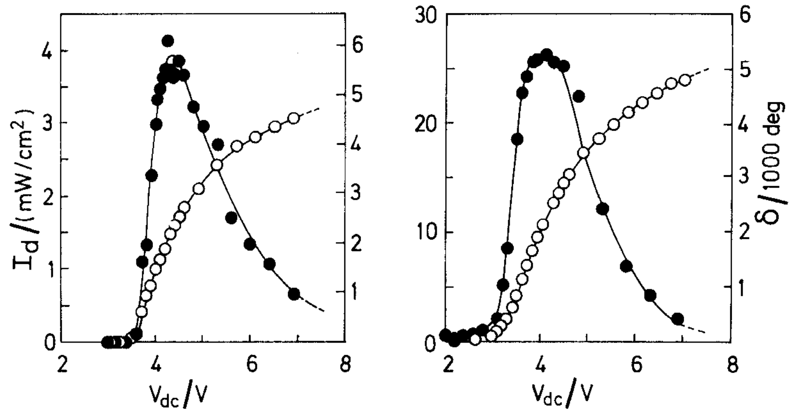

Figure 5. (*a*) The quasistatic electric field induced birefringence indicated by ○ and the diffraction intensity denoted by ● versus electric field with an incident laser at 1 W/cm^2. (*b*) The results for the incident intensity at 3·85 W/cm^2. Solid lines were drawn to aid visualization of the data. ○ indicates the electrocontrolled birefringence and ● denotes the diffraction intensity.

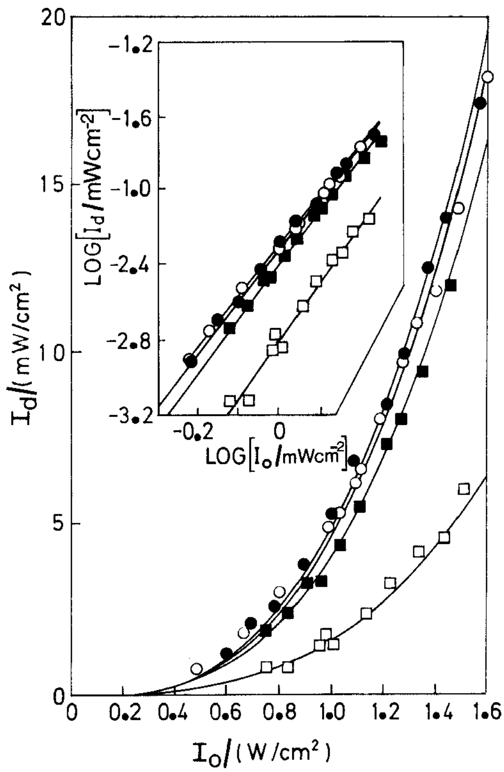

Figure 6. Diffraction intensity versus incident intensity ($I_0 < 1{\cdot}6\,\mathrm{w/cm^2}$) at four field strengths; □ = 3·70 V, ○ = 4·25 V, ● = 4·50 V and ■ = 5·00 V. Solid curves are the fitting lines for the cubic dependent; inset shows $I_d \alpha I_0^3$ as predicted by equation (13), □ represents a slope of 3·12, ○ 2·80, ● 2·74 and ■ 2·88.

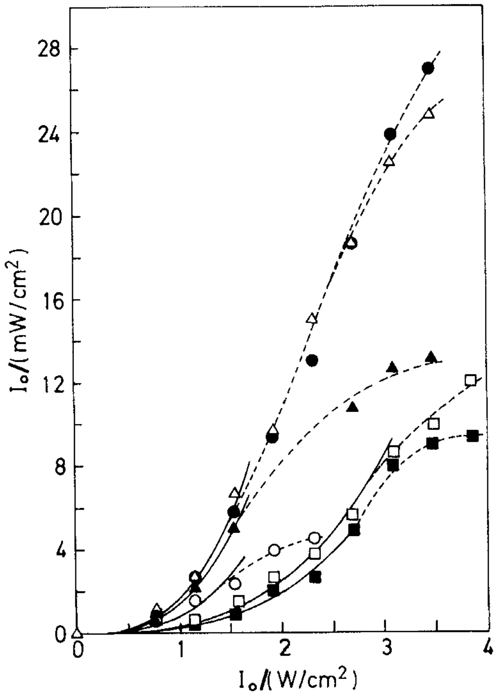

Figure 7. Diffraction intensity versus incident intensity ($I_0 < 4\,W/cm^2$) for various field strengths; ■ = 3·0 V, ● = 3·5 V, △ = 4·0 V, ▲ = 4·5 V, ○ = 5·0 V and □ = 0 V at 45°. Solid curves are lines for the cubic dependence. Broken lines are drawn to aid visualization of the data.

shown in figure 5 (*a*); the slope of the curve reaches its maximum at 3·7 V. There is no significant diffraction intensity in the low field ($E_{dc} \ll E_c$) regime. However the enhancement effect is readily seen once the voltage is raised to the Freedericksz transition critical value. The diffraction intensity appears corresponding to the induced degenerate four-wave mixing and reaches a maximum at 4·38 V. The enhancement effect then decreases with further increase of E_{dc}, as predicted by equation (13). The diffraction intensity decays eventually as V_{dc} goes into the saturation regime of the electrocontrolled birefringence curve. Figure 5 (*b*) shows the applied voltage dependence of the electrocontrolled birefringence and diffraction intensity at the higher incident intensity of 3·85 W/cm^2. The general behaviour is essentially the same as that in figure 5 (*a*). The slope of the curve reaches its maximum at 3·63 V and the diffraction intensity has a peak at 4·0 V. According to our theory, the diffraction intensity is proportional to the square of the amplitude of the phase modulation, a peak must occur at the Freedericksz transition critical field E_c where the slope of the electrocontrolled birefringence curve and so the phase modulation reaches its maximum. In our results, however, for both samples the fields for peak diffraction intensity are larger than the field for maximum phase modulation. This discrepancy is presumably due to the simplifications in our calculation, such as neglecting the finite beam size effect.

Figure 6 shows the diffracted intensity as a function of the weak incident laser intensity. In this low optical intensity limit, the diffracted intensity obeys a degenerate four-wave mixing picture very well, varying as the cube of I_0. This can also be seen

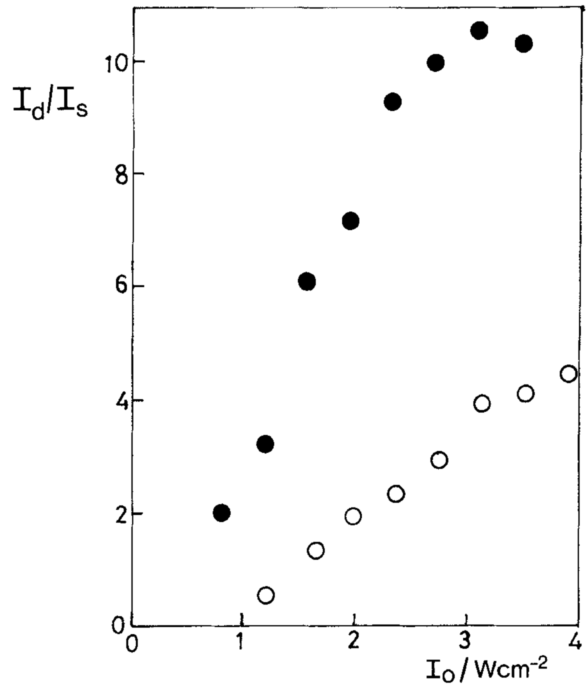

Figure 8. Signal-to-noise ratio (Id/Is) versus incident intensity (I_0 < 4 W/cm^2). ● = 4 V at 0° and ○ = 0 V at 45°

in the inset of figure 6. The slopes are very close to three for all four voltage chosen. Although equation (13) only predicts the cubic dependence, characterizing the degenerate four-wave mixing process, near the Freedericksz transition critical voltage for a weak laser beam, it is true for any applied voltages which induce non-saturating director reorientation, as long as the intensity of the incident beam is weak enough. Deviations from the cubic dependence are observed for a sample biased by the electric field to just above the Freedericksz transition threshold even for a moderate optical intensity (a few W/cm^2), as shown in figure 7.

To induce a phase grating and obtain the diffraction beam for a low optical intensity one can shine the pump beams in at an angle. However this process will be accompanied by high background scattering which is undesirable for applications. In this respect, applying a quasistatic electric field will have some advantages. To illustrate this, the signal-to-noise ratio for the normally incident beams with a voltage of 4 V is compared to that of 45° obliquely incident beams without external electric field. We can see in figure 8 that the signal-to-noise ratio, the ratio of the diffraction intensity I_d to the corresponding background scattering I_s, is improved by about four fold by the applied quasistatic electric field.

5. Conclusions

We have investigated both theoretically and experimentally a quasistatic electric and optical field-induced reorientational phase grating and its diffraction in a nematic film. It was found that a properly biased quasistatic electric field can enhance the phase grating diffraction. The enhancement effects are attributed to the critical behaviour of the Freedericksz transition. The cubic dependence characterizing the degenerate four-wave mixing process is obtained for $I_0 < 1\cdot6\,W/cm^2$ in spite of the strength of the bias voltage. Experimental results are in good agreement with theoretical prediction in the low optical field regime. Deviation from the cubic dependence is observed for pump beam intensity of a few W/cm^2 for a quasistatic electric field biased sample. The dependence of the diffraction intensity on the electric field shows a peak at an electric field greater than the electric field of maximum phase modulation. This discrepancy is attributed to the simplifications used in our calculation. We have also confirmed that the intensity of background scattering is linearly proportional to that of the incident beam and that it is additive.

There are a number of potential applications for the observed effects. First the optically induced phase grating diffraction can be controlled by a properly D.C. field biased nematic cell. The improvement of signal-to-noise ratio further suggests its possible applications in degenerate four-wave mixing based optical phase conjugation, such as aberration compensation in imaging [27]. Other properties, for example, D.C. field biased dependent diffraction efficiency also promises it as a beam splitting ratio tunable element.

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