Quasi-static electric-field-enhanced diffraction effects in a nematic liquid-crystal film

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A quasi-static electric field can enhance laser-induced diffraction rings from a nematic liquid-crystal film. This phenomenon is shown to be the combined result of the critical behavior of the sample at the Freedericksz transition and the nonlinear coupling of the optical and quasi-static electric fields.

Nonlinear optical propagation effects leading to pronounced changes in the transverse profiles of laser beams are of current interest in optical physics. Far-field diffraction rings, for example, have been observed in a number of systems.[1] In particular, a large number of rings have been observed in nematic liquid-crystal (NLC) films because of the large optical nonlinearity associated with molecular reorientation in this medium.[2–4] This phenomenon has been described as the spatial analog of the well-known self-phase modulation of light as the laser beam traverses the sample. The total number of rings, N, is a function of the laser's beam radius and its radius of curvature.[5] In a first approximation, N can be estimated from the optical-field-induced phase shift, $\Delta\psi(\rho)$, by[4]

$$N \simeq \frac{\Delta\psi(\rho = 0) - \Delta\psi(\rho = \infty)}{2\pi}, \qquad (1)$$

where ρ is the transverse position in the beam. Quasi-static electric and magnetic fields can also induce molecular reorientation in NCL films.[6] It is thus expected that optical propagation effects in an NLC film can be influenced by applying such fields on the sample. Indeed, Khoo[7] has shown that a dc magnetic field can be used to enhance the efficiency of degenerate four-wave mixing, while Csillag *et al.*[8] reported ring patterns in NLC films by superposed optical and quasi-static electric fields. It is not immediately obvious from Eq. (1), however, that the quasi-static electric field would increase the number of laser-induced rings because this field only produces a uniform phase change across the laser-beam profile. In this Letter, we clarify this question by presenting quantitative measurements on field-induced birefringence and far-field diffraction rings in a thin-film *p*-methoxy-benzylidene-*p*-*n*-butylaniline (MBBA) NLC. The results are in excellent agreement with our proposed physical picture.

We consider a homeotropically aligned NLC film with the geometry shown in the inset of Fig. 1. The reorientation angle of the director, $\theta(z)$, can be calculated by using a free-energy minimization procedure[3,9] that yields

$$z[2A\sin^2(\beta + \theta_m) + 2B\sin^2\theta_m]^{1/2} = \int_0^{\theta(z)} \left[1 - \frac{A\sin^2(\beta + \theta') + B\sin^2\theta'}{A\sin^2(\beta + \theta_m) + B\sin^2\theta_m}\right]^{-1/2} d\theta', \qquad (2)$$

where $A = (\Delta\epsilon_{op}/8\pi K)E_{op}{}^2$ and $B = -(\Delta\epsilon_{ac}/8\pi K)E_{ac}{}^2$. E_{ac} and E_{op} are quasi-static and optical electric fields; θ_m is the maximum molecular reorientation angle in the liquid-crystal sample; $\Delta\epsilon_{ac}$ and $\Delta\epsilon_{op}$ are the dielectric anisotropies, $\Delta\epsilon = \epsilon_\parallel - \epsilon_\perp$, that are due to the applied fields along and perpendicular to the director; and K is the elastic constant of the NLC sample in the so-called

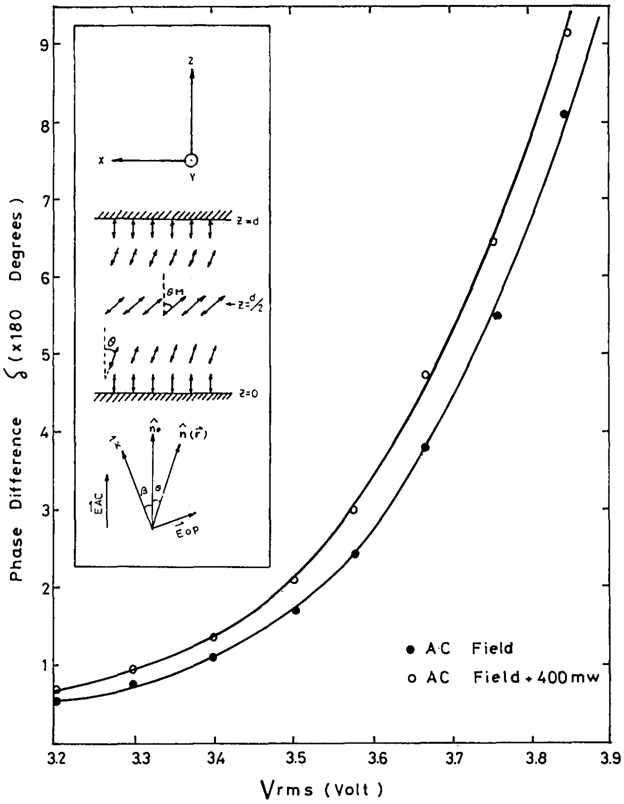

Fig. 1. ECB and superposed quasi-static and optical electric-field-induced birefringence in the Freedericksz-transition region. Inset shows experimental geometry.

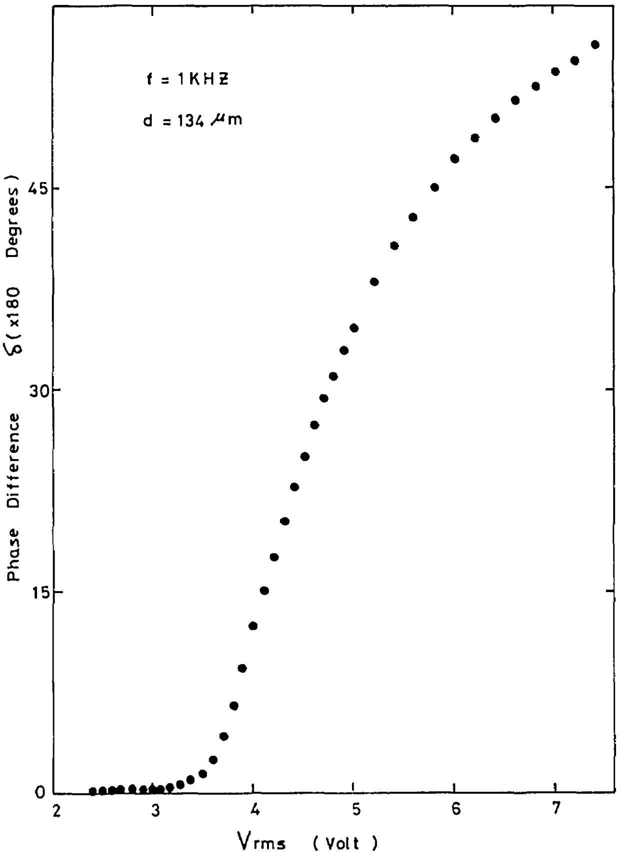

Fig. 2. Experimental values for the quasi-static-field-induced birefringence (ECB).

one-constant approximation.[3] The enhancement effect is evident by examining Eq. (2) in the limit of weak fields, i.e., $E_{ac} \ll E_{op} \ll E_c$, where E_c is the critical field for the Freedericksz transition:

$$\theta(z) \simeq \frac{\Delta\epsilon_{op} \sin 2\beta}{16\pi K} E_{op}{}^2 \left\{ (zd - z^2) + \left[\frac{-z^3 d}{6} + \frac{z}{12} d^3 + \frac{z^4}{12} \right] + \left[\frac{2\cos^2\beta \Delta\epsilon_{op} E_{op}{}^2 - 2\Delta\epsilon_{ac} E_{ac}{}^2}{8\pi K} \right] \right\}. \tag{3}$$

Both terms enclosed in the square brackets are positive. For MBBA, $\Delta\epsilon_{op} > 0$, whereas $\Delta\epsilon_{ac} < 0$. Thus the quasi-static electric field should help to reorient the director such that it is aligned with the pump-laser polarization direction in this case. With the field-induced $\theta(z)$, a normally incident laser beam with wavelength λ_p should have its extraordinary component experience an induced phase change

$$\Delta\psi = \frac{4\pi}{\lambda_p} \int_0^{d/2} \left[\frac{n_0 n_e}{(n_e{}^2 \cos^2\theta + n_0{}^2 \sin^2\theta)^{1/2}} - n_o \right] dz, \tag{4}$$

where n_o and n_e are, respectively, the ordinary and maximum extraordinary refractive indices of the sample.

In our experiment, the NLC cells were 135 μm thick with glass windows coated first with indium oxide as transparent electrodes and then obliquely evaporated with SiO to prevent E_{ac}-induced space charges in the nematic medium. Finally, the samples were treated with octadecyldimethyl [3-(trimethoxysilyl)-propyl] ammonium chloride (DMOAP) for homeotropic alignment. A quasi-static electric field at 1 kHz was applied normal to the glass window. An unfocused Ar^+ laser (beam diameter ~2.25 mm) incident at 30° provided the pump beam. Between the phase measurements at two different voltages, the laser beam was blocked to reduce the heating effect. The sample was maintained at 23 ± 0.1°C. The field-induced birefringence at the Ar^+ laser-beam center was measured by a tightly focused He–Ne probe laser by using a modulation technique originally devised by Lim and Ho.[10] Details will be presented elsewhere.

The experimentally measured electrocontrolled birefringence (ECB) of our sample is given in Fig. 2. It shows a typical critical behavior near the Freedericksz-transition critical field for the quasi-static electric field, i.e., $d\theta/dE_{ac}$ is large. The ECB and the phase difference, $\Delta\psi(0)$, that is due to the superposed fields at a laser intensity of 10 W/cm^2 (400 mW) are illustrated for the transition region in Fig. 1. At this intensity level, Eq. (3) predicts a phase change of 1.28° without E_{ac} and 2.36° at 1 V. This is in good agreement with experimentally measured values (not shown in the figures) of 1 ± 0.3° and 2.7 ± 0.4°, respectively. This phase difference increases dramatically to 210° at 3.86 V, near the Freedericksz-transition critical field, illustrating the strong enhancement in the transition region (Fig. 1). In Fig. 3, we have plotted the maximum phase

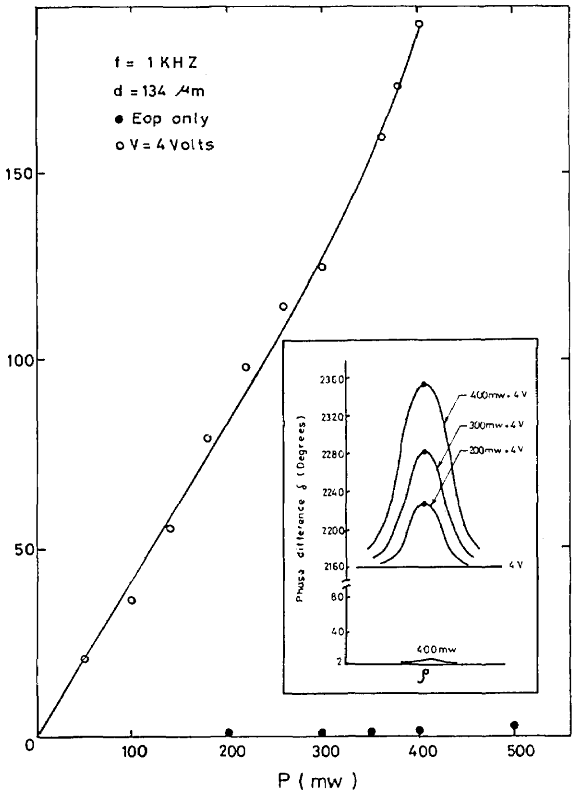

Fig. 3. Optical-field-induced birefringence with and without E_{ac} as a function of incident laser power. Inset shows the proposed physical picture.

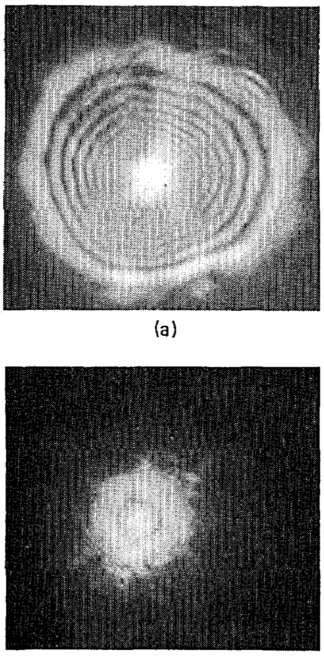

Fig. 4. Far-field pattern of the pump-laser beam at 600 W/cm^2: (a) with and (b) without superposed quasi-static electric potential at 4.1 V.

change across the beam profile, $\Delta\delta = \Delta\psi(0) - \Delta\psi(\infty)$, as a function of pump-laser power. As can be seen in Eq. (3), there is a nonlinear coupling between E_{op} and E_{ac}. This is also expected to enhance the phase difference, and the enhancement would be nonuniform because of the Gaussian beam profile and nonlinear coupling. For example, $\Delta\delta$ increases from 2.8° at 400 mW (~10 W/cm^2) to 192° when a superposed quasi-static electric potential of 4 V is applied. This physical picture is shown in the inset of Fig. 3. An enhanced phase change of multiples of π, presenting itself as alternating bright and dark rings in the far-field pattern, could thus be realized with superposed optical and quasi-static electric fields. As many as 12 rings were observed for $V_{\mathrm{ac}} = 4.1$ V and pump-laser intensity as low as 600 W/cm^2. This is a sixfold enhancement in the number of diffraction rings compared with the case without the quasi-static electric field (Fig. 4).

In conclusion, we have shown that the quasi-static electric field can enhance far-field diffraction rings in a NLC film. Our quantitative measurements show that this is caused by the combined effects of the critical behavior of the nematic material near the quasi-static electric-field-induced Freedericksz-transition critical field and the nonlinear coupling of the optical and quasi-static electric fields. This enhancement effect should be of interest for many applications, notably optical bistable devices and wave-front conjugation.

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