Interference patterns of scattering light induced by orientational fluctuations in an electric-field-biased nematic liquid-crystal film

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Received March 2, 1998

A new light-scattering phenomenon from a planar aligned nematic liquid-crystal film is observed and studied. This new phenomenon exhibits ring patterns in the orthogonal polarization. A simple model based on optical interference has been developed, and its predictions agree well with experimental observation. © 1998 Optical Society of America

OCIS codes: 160.3710, 290.5840, 290.5820, 160.4760, 310.6860.

Light-scattering phenomena of liquid-crystal films have attracted much research interest because they are useful tools for diagnosing the properties of liquid-crystal materials and may have some other applications.[1–5] As an example, diffraction ring patterns caused by the spatial self-phase modulation induced by the optical Gaussian intensity profile in a planar aligned nematic liquid-crystal film have been reported and studied extensively.[6–10] It has also been proposed that optical intensity-limiting devices based on such a phenomenon can be made.[6,7] Here we report and study a new light-scattering phenomenon from the same planar aligned nematic liquid-crystal film structure. This new phenomenon exhibits ring patterns in the orthogonal polarization that are caused by interference of the scattering light. Our aim here is thus to investigate the influences of the applied electric field on these interference patterns both experimentally and theoretically.

Our experimental setup is shown in Fig. 1(a). The liquid-crystal film that we use is composed of the nematic liquid crystal E7 sandwiched between two indium tin oxide–coated glass windows that have been treated with polyvinyl alcohol for planar alignment. A sample thickness d of $\sim 150\ \mu m$ is determined by a Mylar spacer. A 1-kHz electric field generated by a function generator (WaveTek Inc., Model 23) is applied normally to the sample's glass windows. The 514.5-nm-wavelength light beam from an Ar^+ laser is normally incident onto the sample with a spot diameter of ~ 1.9 mm, and its intensity is $\sim 4.55\ W/cm^2$. The sample is fixed on a sample holder, which has a cooling system to eliminate laser heating effects and keep the cell temperature in the nematic phase range. The polarization of the incident beam is in the plane with the direction of the molecular director $\hat{n}$ (the e-wave). The scattering light in the orthogonal polarization (the o-wave) is observed through an analyzer in front of the screen. At first sight it may seem that we should not observe anything in the o-wave direction. However, because there are thermal fluctuations of molecular orientation at room temperature, from the bulk liquid-crystal theory of De Gennes and Prost,[4] light scattering caused by orientational fluctuations can be observed in the orthogonal polarization. Photographs of o-wave scattering light under various bias voltages are shown in Fig. 1(b). From Figs. 1(b-1)–1(b-5) the bias voltage increases monotonically; the maximum bias voltage is <1.8 V. From these figures we can make the following observations: First, there is a dark circular ring fringe in these photographs. Second, the patterns of Figs. 1(b-1)–1(b-4) periodically appear when the bias voltage increases, and the diameter of the dark circular fringe gradually increases with the increase of the bias voltage. Although these circular ring patterns may look like the diffraction rings caused by a thermal effect[6,7] or a molecular reorientation effect[8–10] in the presence of a Gaussian-profile optical intensity distribution, in what follows we prove that such is not the case. In our experiment, no dark ring fringe was observed in the

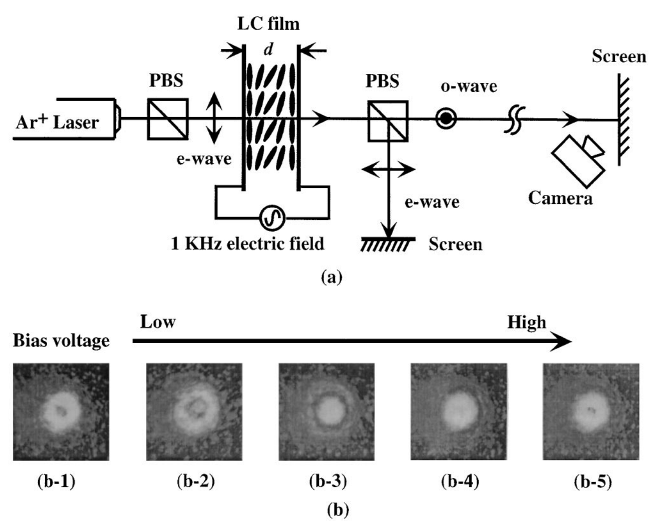

Fig. 1. (a) Experimental setup: PBS, polarizing beam splitter; LC, liquid crystal; d, sample thickness; o-wave, ordinary wave; e-wave, extraordinary wave. (b) Photographs of o-wave scattering light under different bias voltages. From (b-1) to (b-5) the bias voltage is increasing monotonically.

0146-9592/98/120891-03$15.00/0

e-wave polarization (the input polarization), which quickly rules out the possibility that the patterns that we observed are diffraction rings caused by spatial self-modulation. We believe that the formation of these dark circular ring patterns is due to the destructive interference of scattering light. Based on the theory of optical interference, we developed the following simple model to explain our experimental observations. A summary of the differences among the circular ring fringe patterns caused by different physical mechanisms is given in Table 1 for a clear comparison.

To obtain the distribution of effective refractive index in a liquid-crystal film, we must first calculate the distribution of the molecular orientation. Figure 2 is a schematic diagram of the incident and scattered laser light propagating in a planar aligned liquid-crystal film. The wave vectors of the incident and the scattered light are $\mathbf{k}_i$ and $\mathbf{k}_f$, respectively, and the angle between them is ϕ. The angle between the molecular director $\hat{n}$ and the y axis is θ. Consequently the director $\hat{n}$ can be expressed as $\hat{n} = (0, \cos\theta, \sin\theta)$. From the continuum theory,[4] which is a macroscopic phenomenological theory of liquid crystals dealing with a slowly varying director field, we can obtain the distribution function $\theta(z)$ by minimizing Frank free energy F.

Based on previous research[11,12] the Frank free-energy density $\mathcal{F}$ for an electric-field-biased nematic liquid-crystal film is given by

$$\mathcal{F} = 1/2[K_{11}(1 - K\sin^2\theta)(\partial\theta/\partial z)^2] - \frac{D_z{}^2}{8\pi\epsilon_\perp(1 - W\sin^2\theta)} - \frac{In_e}{c(1 - \mu\sin^2\theta)^{1/2}}, \quad (1)$$

where $K = 1 - K_{33}/K_{11}$, $W = 1 - \epsilon_{/\!/}/\epsilon_\perp$, $\mu = 1 - (n_e/n_o)^2$, D_z is the z component of the electric displacement, c is the velocity of light in vacuum, I is the input optical intensity, and K_{11} and K_{33} are splay and bend elastic constants, respectively. Instead of solving for the distribution function $\theta(z)$ directly, we assume a trial solution of $\theta(z)$ as follows:

$$\theta(z) = \theta_m \sin(\pi z/d). \quad (2)$$

Here we have imposed the hard boundary conditions $\theta(z = 0) = \theta(z = d) = 0$, and θ_m represents the maximum orientation angle at $z = d/2$. Substituting the trial solution into expression (1) and then integrating the free-energy density over the whole volume of the liquid-crystal film, we obtain the total free energy $F = \int_V \mathcal{F}\mathrm{d}V$ as a function of θ_m. In equilibrium the free energy is at a minimum, and thus θ_m must satisfy $\partial F/\partial\theta_m = 0$, which gives rise to the following equation for θ_m:

$$2\theta_m - K[\theta_m - \theta_m J_0(2\theta_m)] - \left[\left(\frac{V}{V_{\mathrm{th}}}\right)^2 - \frac{I}{I_{\mathrm{th}}}\right][2J_1(2\theta_m)] = 0. \quad (3)$$

Here J_i is a Bessel function of the first kind of order i, I is the incident optical intensity, V is the bias voltage, $I_{\mathrm{th}} = cK_{11}(\pi/d)^2/(-n_e\mu)$ is the threshold optical intensity, and $V_{\mathrm{th}} = 2\pi[\pi K_{11}/(\epsilon_{/\!/} - \epsilon_\perp)]^{1/2}$ is the threshold voltage. Under the small-angle approximation, Eq. (3) can be solved to yield

$$\theta_m \cong \left(\frac{2b}{b + 1 - K}\right)^{1/2}, \quad (4)$$

where the effective field is $b = (V/V_{\mathrm{th}})^2 - (I/I_{\mathrm{th}}) - 1$. Substituting Eq. (2) and relation (4) into the expression for the effective refractive index for the e-wave optical field, $n_{\mathrm{eff}}(\theta) = n_e/(1 - \mu\sin^2\theta)^{1/2}$, we have

$$n_{\mathrm{eff}}(\theta) \cong n_e - \frac{n_e(-\mu)b}{b + 1 - K}\sin^2(\pi z/d). \quad (5)$$

We are now ready to explain our experimental observations based on the theory of optical interference. From the schematic diagram shown in Fig. 2, the phase difference $\Delta\delta_{\max}$ between beams 1 and 2 can be expressed as

$$\Delta\delta_{\max} = \frac{2\pi}{\lambda}\left[\int_0^d n_{\mathrm{eff}}(\theta)\mathrm{d}z - n_o d\cos\phi\right], \quad (6)$$

Table 1. Comparison of Circular Ring Fringe Patterns from Different Physical Mechanisms

Type of Wave	Type of Physical Mechanism: Diffraction Ring: Thermal Effect	Diffraction Ring: Molecular Reorientation	Interference of Scattering Light
o	Yes	No	Yes
e	Yes	Yes	No

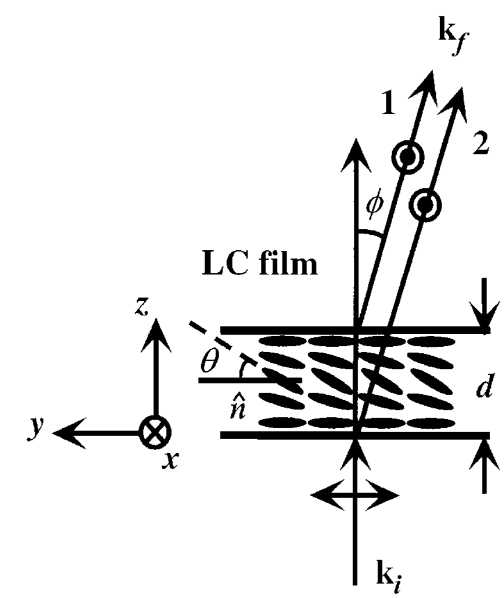

Fig. 2. Schematic diagram of the incident and scattered light in a planar aligned nematic liquid-crystal film: $\mathbf{k}_i$, $\mathbf{k}_f$, wave vectors of the incident and the scattered light, respectively; ϕ, scattering angle; $\hat{n}$, molecular director; θ, molecular orientation angle.

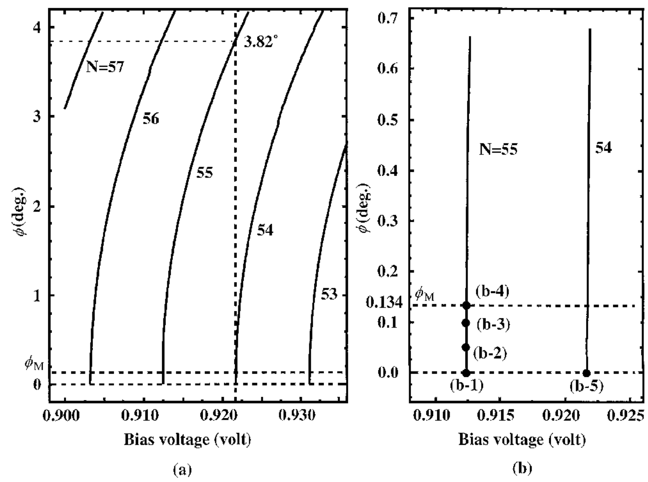

Fig. 3. (a) Numerical results of angle ϕ_N versus bias voltage. (b) Resized version of (a). Points, operating points corresponding to Figs. 1(b-1)–1(b-5). ϕ_M is the maximum observable scattering angle and is ~0.134°.

where ϕ is the scattering angle and λ is the optical wavelength in vacuum. Substituting relation (5) into Eq. (6) and performing the integration, we obtain

$$\Delta\delta_{\max} \cong \frac{2\pi d}{\lambda}\left[n_e - \frac{n_e(-\mu)b}{2(b+1-K)} - n_o \cos\phi\right]. \quad (7)$$

This is the maximum phase difference among the scattering lights that occur at different positions inside the liquid-crystal film. From the viewpoint of optical interference, the condition for the occurrence of destructive interference is $\Delta\delta_{\max} = N \times 2\pi$, where N is an integer. When this condition is satisfied, the scattering lights that occur at different positions inside the liquid-crystal film cancel one another, just as in the case of the single-slit optical interference experiment. As we shall see, predictions from relation (7) are in good agreement with our experimental results.

From relation (7), the formation of interference patterns with a dark circular fringe can be explained and the characteristics of the interference patterns under different bias voltages can be obtained. First, if an external electric field is present, there is a minimum positive integer N for the occurrence of destructive interference; this means there is a minimum scattering angle ϕ_N for observation of a dark circular fringe. Second, as the term $n_e(-\mu)b/[2(b+1-K)]$ in relation (7) increases with the bias voltage, angle ϕ_N must behave in the same way to satisfy the interference condition. Therefore the diameter of the dark fringe increases with the bias voltage for a fixed integer N. Third, because the angle distribution of the scattering light is finite, our observation window is also finite. Therefore fringes that move outside the window can no longer be observed. However, the interference fringe that corresponds to a smaller N may become visible within the observation window, which explains why interference patterns periodically occur when the bias voltage is increased. Finally, angle ϕ_N under different bias voltages can be calculated numerically from relation (7). Using typical values[13] $K = -0.36$, $n_e = 1.712$, $n_o = 1.52$, $\mu = -0.27$, $V_{\text{th}} = 0.9$ V, $I_{\text{th}} = 592$ W/cm^2 for nematics E7, $\lambda = 514.4$ nm, $d = 150$ μm, and $I = 4.55$ W/cm^2 yields the calculated results shown in Fig. 3. The difference between angle ϕ_N for $N = 55$ and for $N = 54$ at a biased voltage of 0.9217 V is ~3.82°, as can be seen from Fig. 3(a). However, in our experiment the maximum observable scattering angle ϕ_M was only ~0.134°, which is much smaller than 3.82°. This explains why only one dark circular fringe is observed in Fig. 1(b). Moreover, in Fig. 3(b) the dots labeled (b-1)–(b-5) are the operating points that correspond to the photographs in Fig. 1(b). The numerical results are in good agreement with the experimental data.

In conclusion, we have observed and studied a new light-scattering phenomenon from a planar aligned nematic liquid-crystal film. Circular dark fringe patterns were observed in the orthogonal polarization; they periodically appeared when the bias voltage was increased. A model based on optical interference was developed, and its predictions agree well with our experimental observations. We are currently investigating how the patterns will change when the bias voltage, the input optical intensity, or both are high.

This research is supported, in part, by the National Science Council of Taiwan under contract NSC87-2112-M009-012,

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