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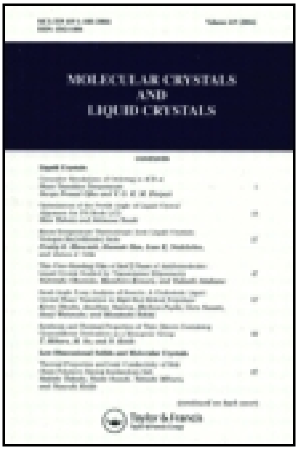

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QUÉTELET-TYPE INTERFERENCE FROM LIQUID CRYSTAL POLYMER DISPERSION FILMS

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The Quételet-type ring pattern is observed in liquid crystal polymer dispersion (LCPD) films. The clusters of the polymer network and LC domains with different director axes in the LCPD films serve as scatterers. Changes in the first-order ring intensity are probed with a linearly polarized laser beam. Experimental results show that the first-order ring intensity of the Quételet-type interference peaks when the polarization of the probe beam is parallel to the director axis of the LC molecules. Also, it is shown that the number of the scatterers, the order parameter of the liquid crystals and the applied electric field affect the ring intensities. A simple model is proposed to explain these observations.

Keywords: interference; liquid crystals; Quételet; scattering

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1. INTRODUCTION

Liquid crystal-polymer dispersion films have recently received increasing attention due to not only their potential uses in flat-panel displays, but also their interesting physical phenomena [1–5]. The potential uses of the LCPD film in electro-optical and flat-panel applications have been the principal driving force behind the extensive researches on the structure and the morphology of the liquid crystals-polymer composite system. The polymer-dispersed liquid crystals (PDLCs) are one type of LCPD film [6–7]. They contain approximately equal amounts of nematic liquid crystals and polymer. A final cured PDLC sample consists of liquid crystal droplets embedded in the continuous polymer matrix. The directors of the liquid crystals inside a droplet are almost along one direction, which is referred to as the droplet director. In the field-on condition, the droplet directors are aligned along the applied field, if a positively anisotropic LC is employed. The sample is transparent. In the field-off condition, the droplet directors are randomly orientated in the sample. Accordingly, the refraction indices of the liquid crystals and the polymer are not matched, and the sample becomes opaque. One problem exhibited by the PDLCs is that in the field-on condition, the film is transparent only for light incident at a particular angle and becomes hazy for light incident at other angles [8].

Another LCPD material called the Polymer-Stabilized Cholesteric Texture (PSCT) [9–10] was developed to solve the refractive mis-matching problem in the PDLCs. In this system, a small amount of monomer (usually below 10 wt%) is dissolved in a cholesteric liquid crystal medium. Fiber-like anisotropic polymer networks are formed after the monomer is polymerized. The surface alignment or the external applied field determines the orientation of the liquid crystals, which, in turn, determines the direction of the polymer network [11]. In such a system, the polymer network imposes an aligning force on liquid crystal molecules, but an external electric field can overcome this force. The refractive mis-matching problem is eliminated because a low concentration of polymer is used in PSCTs. As a result, PSCTs are haze-free at wide viewing angles in the clear mode and have been reported to show great potential in display applications.

The earliest study of the interference fringes arising from light scattering was probably performed by Newton and can be found in Isaac Newton's Optiks (1704, Book Two, Part IV). Most interference experiments require a careful adjustment of the apparatus and special conditions of light illumination. Contrarily, the light scattering interference does not need such strict conditions. In fact, it is almost unavoidable in any optical systems that contain reflecting surface. Thank to the modern design of optical instruments, which almost have eliminated surface-induced light interference.

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Therefore, the effect no longer appears to have practical interest, but remains only historical and pedagogical significance [12]. This work presents the scattering light interference results obtained from the LCPD films, and gives the classical scattering light theory a new significance in the liquid crystal research. Further, it is motivated that another technique to study the LCPD films. The concentric rings pattern observed in these samples are due to the Quételet-type interference [12–13]. The polymer clusters and LC domains with different director axes in the LCPD films serve as scatterers. The polymer clusters possess a dielectric constant, which differ from that of the neighboring anisotropic liquid crystal domains. Samples with homogeneous, hybrid and untreated cells are fabricated. Changes in the first-order ring intensity are measured with a polarized laser light. Experimental results show that the first-order ring intensity of the Quételet-type scattering interference peaks when the polarization of the probe beam is parallel to the director axis of the LC molecules. Also, it is shown that the number of the scatterers, the order parameter of the liquid crystals and the applied electric field affect the ring intensities. A simple model is proposed to explain these observations.

2. EXPERIMENTAL

The nematic liquid crystal used in this experiment was E7 (E. Merck). $\sim 2.5\,\mathrm{wt}\%$ of laboratory synthesized monomer Bis [6-(acryloyloxy) hexyloxy]-1,1′biphenylene (BAB6) and a small amount of photoinitiator Benzyl methylether (BME; from Polyscience) were added to the LC. The monomer BAB6 has a reactive double bond at each end. Under UV radiation, the monomer is polymerized to form a cross-linked anisotropic polymer network [11]. When the UV intensity is low, there are fewer free radicals produced in the LCPD mixture. The free radicals not only initiate chain reaction but also terminate it. With fewer free radicals, there are fewer polymerization sites and the polymer chains grow longer. However, when the UV intensity is high, the free radicals increase and rapidly terminate the polymerization. Thus, the polymer network with large clusters and tiny networks are formed [11].

Cleaned ITO glasses were coated with PVA (Polyvinyl alcohol), and then rubbed with a cloth, to promote homogeneous alignment. The homogeneous cells were constructed using two identical but anti-parallel PVA-treated alignment substrates. For the homeotropical substrates, the ITO glasses were treated with DMOAP ([3-(trimethoxysilyl)propyl]octadecyl-dimethylammonium chloride). The hybrid cells were constructed with a homogeneously treated substrate and a homeotropically treated substrate. The untreated cells were fabricated with two untreated ITO glasses.

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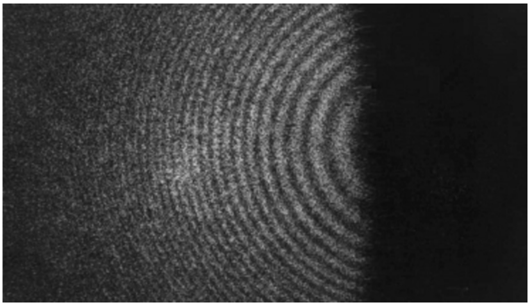

FIGURE 1 The observed half pattern of the Quételet-type ring pattern of a PSCT normal-mode cell in a field-off condition.

Afterwards, the cells were filled capillarily with the LC-monomer mixture. A 15□m-thick spacer was used to control the cell thickness. Finally, the filled cells were polymerized using a Phillips model 400/30 S metal halide lamp. The curing UV intensity was 4 mW/cm^2 and the UV curing time was about 20 minutes.

Figure 1 shows the Quételet-type concentric rings pattern observed from a normal-mode PSCT cell on a separate experiment [13]. The screen was placed just in front of the photodiode. This sample had a monomer concentration of 2.5 wt% and the UV curing intensity was about 2.6 mW/cm^2.

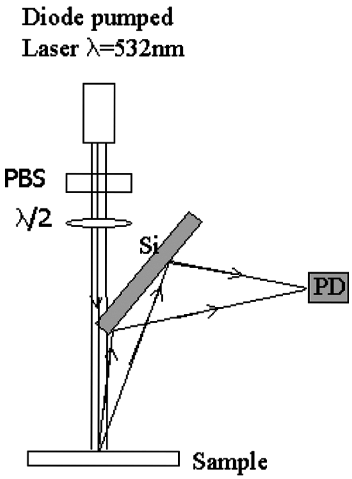

FIGURE 2 A schematic diagram of the experimental setup used to study Quételet-type ring pattern. $\lambda/2$: half waveplate; PBS: polarized beam splitter cube; PD: photodiode; Si: silicon wafer.

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The scatterers are the cluster parts of the polymer networks formed during polymerization and the focal conic LC domains. It should be noted that the silicon wafer shielded half of the incident laser beam, thus we could only observe half of the scattering ring pattern on the screen only.

Figure 2 depicts the experimental setup. A diode pumped CW Nd-YVO$_4$ laser light with a wavelength of 532 nm was incident onto the sample at an angle of $\sim 1^\circ$, and backward Quételet-type concentric rings, due to micron-sized "scatterers" in the LCPD sample, were observed. The polarization of the incident light was controlled using a polarized beam splitter (PBS) cube and a $\lambda/2$ waveplate. The scattering beam was reflected by a silicon wafer and then detected by a photodiode. The electrical signal from the photodiode was fed into a photometer for calculation.

3. RESULTS AND DISCUSSION

Witte [12] reported that particles with radii of $0.3 \sim 30\,\mu\text{m}$ could be used as scatterers in the Quételet-type ring experiment. Also, the scatterers are not limited on the glass surface. They can be dispersed randomly in three dimensions in the sample. In this paper, the LCPD films were fabricated in the condition of high UV intensities; large clusters with tiny networks were formed. For simplicity of discussion, we neglected the tiny networks in the LCPD films and assumed that the clusters were the only scatterers in the LCPD films. Assume that small polymer clusters with radius a and dielectric constant ε_p are surrounded by the liquid crystal domains with dielectric constant $\varepsilon_{\mathit{eff}}(\theta)$, where θ is the angle between the polarization of the incident light and the long axes of the LC molecules. The electric dipole moment induced in a polymer cluster by the incident laser beam is written as [14]

$$\vec{p} = 4\pi\varepsilon_0\varepsilon_{\mathit{eff}}(\theta)\left(\frac{\varepsilon_p - \varepsilon_{\mathit{eff}}(\theta)}{\varepsilon_p + 2\varepsilon_{\mathit{eff}}(\theta)}\right)a^3\vec{E}_{inc} \tag{1}$$

where $\vec{E}_{inc}$ is the incident field and $\varepsilon_{\mathit{eff}}(\theta)$ is the effective dielectric constant of LC molecules along the incident field polarization. Assuming that no magnetic dipole moment is present, i.e. $\vec{m} = 0$, the induced scattering electric field $\vec{E}_{Sc}$ becomes [15],

$$\begin{aligned}\vec{E}_{sc} &= \frac{1}{4\pi\varepsilon_0}k^2\frac{e^{ikr}}{r}[(\vec{n}\times\vec{p})\times\vec{n}]\\ &= k^2a^3\frac{e^{ikr}}{r}\varepsilon_{\mathit{eff}}(\theta)\left(\frac{\varepsilon_p - \varepsilon_{\mathit{eff}}(\theta)}{\varepsilon_p + 2\varepsilon_{\mathit{eff}}(\theta)}\right)[(\vec{n}\times\vec{E}_{inc})\times\vec{n}],\end{aligned}$$

where $\hat{n}$ is a unit vector in the direction of observation and r is the distance from the scatterer, k is the absolute value of the wave vector of the incident light. The effective dielectric constant of LC can be approximated as [16]

$$\varepsilon_{eff}(\theta) = \varepsilon_{\|}\cos^2\theta + \varepsilon_{\perp}\sin^2\theta, \tag{3}$$

where $\varepsilon_{\|}$ and $\varepsilon_{\perp}$ are dielectric constants of LC molecules parallel and perpendicular to the director axis, respectively. The scattering system of an LCPD film consists of many small scatterers, thus, the first-order ring intensity of the Quételet-type scattering light interference resulting from N individual scatterer is given by,

$$I \propto N^2\left[\varepsilon_{eff}(\theta)\left(\frac{\varepsilon_p - \varepsilon_{eff}(\theta)}{\varepsilon_p + 2\varepsilon_{eff}(\theta)}\right)\right]^2, \tag{4}$$

Equation (4) reveals that the first-order ring intensity correlates with the number of the scatterers, ε_p, $\varepsilon_{eff}(\theta)$, and the difference between ε_p and $\varepsilon_{eff}(\theta)$.

Also, in the nematic phase, refractive indices and dielectric constant are mainly determined by the order parameter and the density of the liquid crystals [17]. According to the semi-empirical model developed by Khoo and Normandin, the dielectric constants of the liquid crystals can be expressed as [18]

$$\begin{aligned} \varepsilon_{\|}(T) &= 1 + c_1\rho(T) + \frac{2}{3}c_2\rho(T)S(T), \\ \varepsilon_{\perp}(T) &= 1 + c_1\rho(T) - \frac{2}{3}c_2\rho(T)S(T), \end{aligned} \tag{5}$$

where c_1 and c_2 are parameters that depend on the individual molecular structures and the internal fields, S(T) is the order parameter, ρ(T) is the density and T is the ambient temperature. Substituting Eqs. (5) and (3) into Eq. (4), the qualitative first-order ring intensity of an homogeneous LCPD film were simulated and shown in Figure 3, as a function of the polarization of the probe beam. The simulated first-order ring intensity is seen to vary periodically with θ made between the polarization of the probe beam and the rubbing direction, peaks at 0°, i.e., along the director axis of the LC molecules. In this work, ε_p is assumed to be a constant. As is shown in Figure 3(a), the first-order ring intensity, at a given θ, decreased with decreasing the number of the scatterers. In the figure, number of the scatterers have the normalized ratio of 1:0.85:0.70. Moreover, the first-order ring intensity decreased with decreasing order parameter of the liquid crystals at an angle around 0°, but increased at an angle near 90° as shown in Figure 3(b). In the figure, the normalized order parameters

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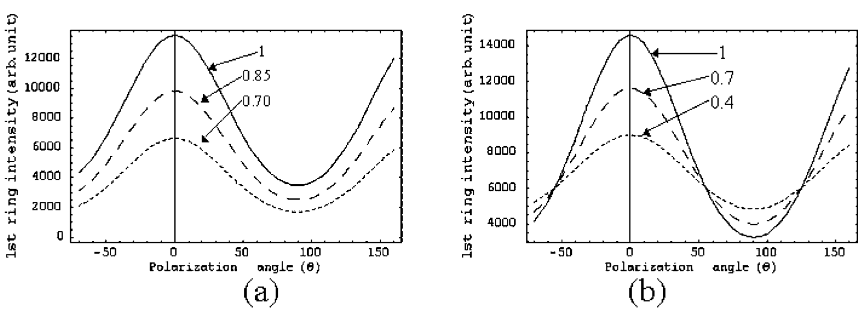

FIGURE 3 (a) The qualitatively simulated first-order ring intensity from an homogeneous LCPD film with various normalized ratios of the number of scatterers. (b) The qualitatively simulated first-order ring intensity from a homogeneous LCPD film with various order parameters of the liquid crystals.

of the three curves are 1, 0.7 and 0.4, respectively. As the order parameter decreased, the dielectric constant of LCs $\varepsilon_{\parallel}$ and $\varepsilon_{\perp}$ were known to decrease and to increase, respectively. Therefore, the simulated result is reasonable.

Figure 4 shows the variation of the measured first-order ring intensity of homogeneous and untreated samples with the polarization angle θ. As is seen in the figure, it peaks at an angle ∼0□, for the homogeneous cell, i.e., along the director axis of the LC molecules. The result is consistent with the simulated one as shown in Figure 3(a). Accordingly, the orientation of the LC molecules can be readily obtained using the Quételet-type interference method. Also, the difference between the maximum and the minimum of the first-order ring intensity of the homogeneous cell is larger

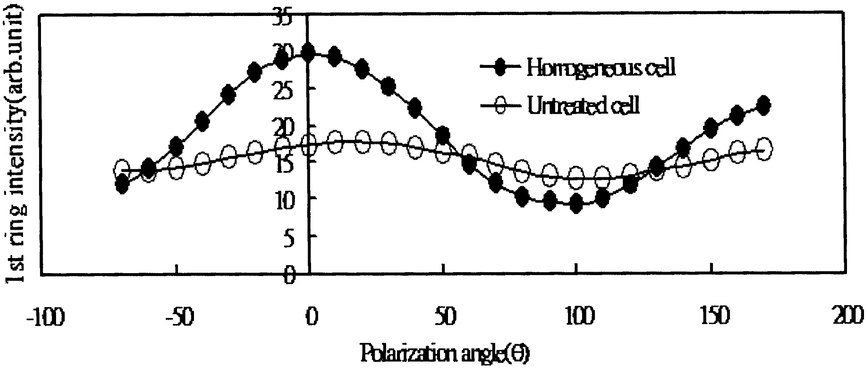

FIGURE 4 The variations of the measured first-order ring intensities of an homogeneous cell and an untreated cell with respect to the polarization angle.

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than that of the untreated cell. The reason is that the homogeneous cell has a higher order parameter than that of the untreated cell, due to the strong surface aligning anchoring force provided in the homogeneous cell. The slight variation of the untreated cell is believed to be due to the flowing direction during filling. Therefore, the observed results are reasonable.

Figure 5 shows the variations of the measured first-order ring intensities of an homogeneous LCPD film with the polarization angle θ for the light being incident from the front and back surfaces after curing. No voltage is applied to the cell in this experiment. The results show that the change of the first-order ring intensity from the minimum to the maximum measured from the front surface is slightly greater than that from the back surface. This can be understood as follow [13]. As a film of LC/monomer mixture is cured using UV light, the photoinitiator decomposes into free radicals upon UV light absorption. The generated free radicals attach to successive monomers and polymerization continues to form polymer networks, due to the overshadowing effect from the curing light, which propagates through the sample. The polymer networks grow denser on the front surface, which face the incident curing light. Therefore, the measured first-order ring intensity from the front surface should have a higher amplitude than that from the back surface. However, the sparser polymer clusters nearing the back surface result in not only lower density of scatterers, but also a less ordered LC alignment due to the weaker anchoring force provided from the sparser polymer networks [1]. Therefore, the first-order ring intensity nearing 90° does not vary significantly.

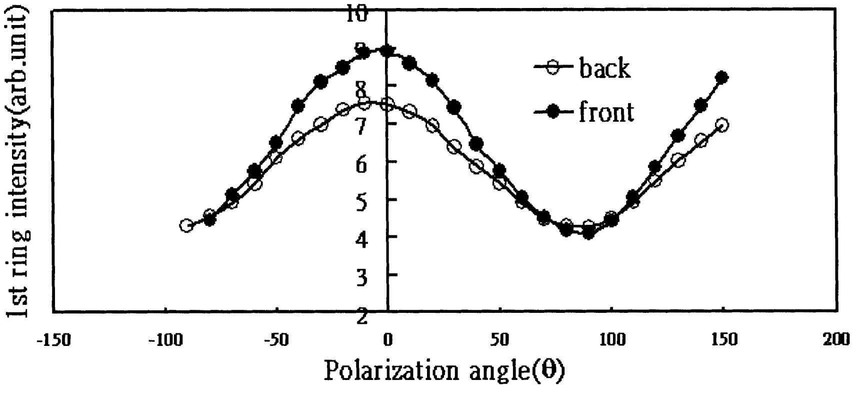

FIGURE 5 Measured first-order ring intensities of an homogeneous LCPD film, with light incident from the front, and the back surfaces after curing. No voltage is applied across the cell.

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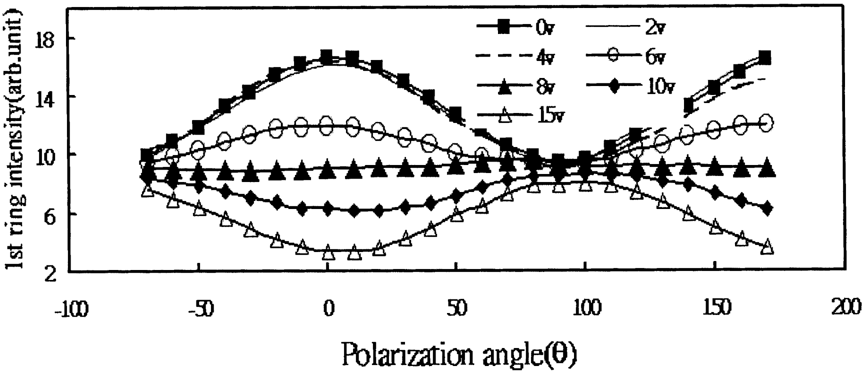

FIGURE 6 Measured first-order ring intensities of a homogeneous cell as a function of the polarization angle for various applied AC voltages (V_{rms}1 kHz).

Figure 6 plots variation in the measured first-order ring intensities of a homogeneous cell with the polarization angle, for various applied AC voltages (V_{rms}, 1 kHz). At low applied voltages (0∼4 V), the cell is homogeneous; the measured curves of the first-order ring intensity are cosine-like, and the magnitude of the change from the minimum to the maximum is high. The signal is attributed to the polymer clusters that are immersed in the LC molecules at these voltages. Some of the LC domains become reorientated as the applied voltage exceeds ∼6 V. The LC domains adjacent to the polymer networks remain uninfluenced by the applied field, due to the overwhelming aligning force of the polymer network. Thus, LC domains with various director axes are generated and scatter the incident light. Notably, the incident light is strongly scattered at angles around 0°, due to a large variation in the refraction index in this direction [1]. Hence, the measured curve of the first-order ring intensity again becomes negatively cosine-like upon increasing the voltage, due to the increase in the number and size of the LC domains.

4. CONCLUSION

The Quételet-type ring pattern is observed in liquid crystal polymer dispersion (LCPD) films. The clusters of the polymer network and LC domains with different director axes in the LCPD films serve as scatterers. Changes in the first-order ring intensities are measured with a polarized laser light. Experimental results show that the first-order interference intensities of the Quételet-type rings peak when the polarization of the incident light is parallel to the director axes of the LC molecules. Thus, the orientation of the LC molecules in a LCPD film can be identical readily using the Quételet-type ring method. Also, it is shown that the density of the scatterers, the order parameter of the liquid crystals and the applied electric field affect the ring intensities. This work measures the Quételet-type interference effect, the observed results can be adequately explained with a simplified model. The real scattering process in the cell is multiple and more complex, goes beyond the scope of this article. The morphology and the size of the polymer clusters are ignored in this work. However, they may play important roles in this system. More detailed experimental and theoretical works are underway.

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