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STATISTICAL, NONLINEAR, AND SOFT MATTER PHYSICS

Small-Angle Light Scattering from Polymer-Dispersed Liquid-Crystal Films

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Abstract—A method is developed for modeling and computing the angular distribution of light scattered forward from a single-layer polymer-dispersed liquid-crystal (PDLC) film. The method is based on effective-medium approximation, anomalous diffraction approximation, and far-field single-scattering approximation. The angular distribution of forward-scattered light is analyzed for PDLC films with droplet size larger than the optical wavelength. The method can be used to study field- and temperature-induced phase transitions in LC droplets with cylindrical symmetry by measuring polarized scattered light intensity.

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1. INTRODUCTION

Liquid crystals (LCs) and LC composites have been the subject of ongoing studies due to their wide use in information display devices, telecommunication systems, optoelectronics, and other applications [1–5]. Analysis of light scattering is an effective approach in the study of LCs and polymer/LC composite films.

Polymer-dispersed liquid-crystal (PDLC) films are composite materials consisting of liquid-crystal droplets dispersed in a solid polymer matrix. Controlled light scattering from LC materials is achieved by using electrically, magnetically, or thermally induced change in director orientation and molecular configuration to manipulate their optical properties. Since liquid crystals are optically anisotropic, scattering problems are more difficult to solve for single LC droplets and droplet arrays as compared to optically isotropic particles. For this reason, solutions are generally obtained by approximate methods. The choice of an approximate method depends on the sample parameters and the wavelength of the incident light. For example, the Rayleigh or Rayleigh–Gans approximation is used when the droplet size is smaller than the wavelength [6, 7]. Small-angle light scattering from PDLC droplets much larger than the wavelength of the incident light is analyzed by using anomalous diffraction approximation [6, 8]. However, even approximate analytical or semianalytical solutions can be obtained only for particular distributions of molecules inside a droplet [7, 8].

In this paper, we propose a method for analyzing the angular distribution of light scattered forward from a single-layer PDLC film (monolayer) with droplet size larger than the optical wavelength in the polymer matrix. The method is based on anomalous diffraction approximation and an interference approximation taking into account cooperative scattering effects for an array of anisotropic LC droplets [9–12]. We examine the intensities of the forward-scattered light components polarized parallel (vv) and perpendicular (vh) to the polarization of a linearly polarized plane wave normally incident on a single-layer PDLC film. We consider a single layer of symmetric spherical LC droplets with cylindrical symmetry. The internal structure of the droplets and their orientation in the layer are modeled by using a hierarchy of scalar and tensor order parameters [2, 13, 14], which substantially simplifies solution of direct and inverse scattering problems [15–17].

The results obtained here provide a basis to develop simple techniques for studying field- and temperature-induced phase transitions in nematic LC droplets with the use of measured angular distributions of scattered polarized intensity. They can be used to retrieve the film and droplet orientational order parameters for LC droplets larger than the optical wavelength in the polymer matrix.

2. ANGULAR DISTRIBUTION OF LIGHT SCATTERED FROM A PDLC MONOLAYER: EFFECTIVE-MEDIUM APPROXIMATION FOR LARGE LC DROPLETS

Figure 1 schematizes a PDLC monolayer illuminated with a normally incident linearly polarized plane wave. Here, the laboratory coordinate system xyz is

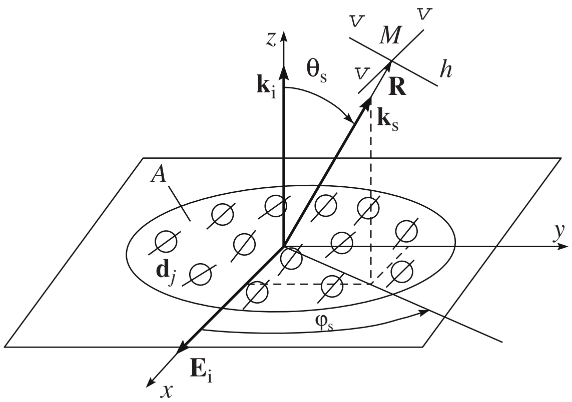

Fig. 1. Schematic representation of illumination of a single-layer PDLC film for analysis of the angular distribution of scattered light: xyz is the laboratory coordinate system; xy is the film plane; $\mathbf{E}_i$ is the incident linearly polarized electric field; $\mathbf{k}_i$ is the incident wavevector; $\mathbf{k}_s$ is the scattered wavevector; lines vv and vh correspond to the scattered polarization components parallel and perpendicular to the incident polarization; $\mathbf{d}_j$ is the director of the jth droplet; A is the illuminated area; and $\mathbf{R}$ is the position vector of observation point M.

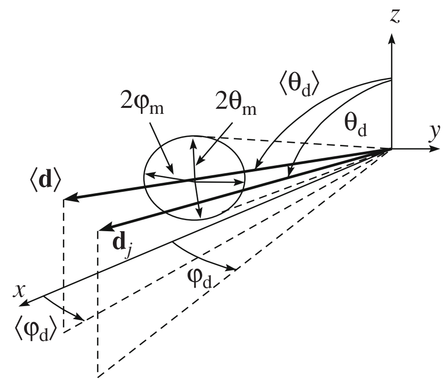

Fig. 2. Schematic representation of LC droplet director orientation in a monolayer: $\langle\mathbf{d}\rangle$ is the average droplet director; angles $\langle\varphi_d\rangle$ and $\langle\theta_d\rangle$ define the orientation of $\langle\mathbf{d}\rangle$; angles φ_d and θ_d define the orientation of the jth droplet director $\mathbf{d}_j$; $2\varphi_m$ and $2\theta_m$ are the director cone angles in the $\langle\mathbf{d}\rangle y$ and $\langle\mathbf{d}\rangle z$ planes, respectively.

defined by the incident polarization (x axis), the incident wave propagation direction (z axis), and the film plane (xy); A is the illuminated area; θ_s and φ_s are the polar and azimuthal scattering angles, respectively; and lines vv and vh correspond to the transmitted polarization components parallel (vv) and perpendicular (vh) to the incident polarization, which can be measured with parallel and crossed polarizers, respectively. In Figs. 1 and 2, the director orientation of the jth LC droplet is represented by the vector $\mathbf{d}_j$ ($j = 1, \ldots, N$, where N is the number of droplets within the area A).

We consider monolayers of droplets whose size is larger than the optical wavelength in the polymer matrix. The liquid-crystal and polymer refractive indices are nearly equal. Under these conditions, multiple scattering between droplets is negligible, and we can use single-scattering approximation, which has also been called interference approximation since it takes into account the far-field interference of waves scattered by the droplets [10]. The multiple-scattering contribution decreases with increasing droplet size as more light is scattered forward. Assuming that the droplets are illuminated only with the incident light and taking into account the far-field interference of waves scattered by the droplets, we write expressions for the intensities of the vv and vh components of incoherent (diffuse) light transmitted through the PDLC film:

$$I_{\text{inc}}^{vv} = \frac{E_i^2 N}{k^2 R^2}\sum_{l=1}^{m} P_l \left|f_l^{vv}(\mathbf{k}_s)\right|^2 + \frac{E_i^2 N}{k^2 R^2}\sum_{l,l'=1}^{m} P_l P_{l'} f_l^{vv}(\mathbf{k}_s) f_{l'}^{vv*}(\mathbf{k}_s)(S_{ll'}(\mathbf{k}_s)-1), \quad (1)$$

$$I_{\text{inc}}^{vh} = \frac{E_i^2 N}{k^2 R^2}\sum_{l=1}^{m} P_l \left|f_l^{vh}(\mathbf{k}_s)\right|^2 + \frac{E_i^2 N}{k^2 R^2}\sum_{l,l'=1}^{m} P_l P_{l'} f_l^{vv}(\mathbf{k}_s) f_{l'}^{vh*}(\mathbf{k}_s)(S_{ll'}(\mathbf{k}_s)-1), \quad (2)$$

where the partial structure factors $S_{ll'}(\mathbf{k}_s)$ are calculated as

$$S_{ll'}(\mathbf{k}_s) = 1 + \Lambda\int_A (W_{ll'}(\mathbf{r})-1)\exp(i\mathbf{k}_s\cdot\mathbf{r})d\mathbf{r}. \quad (3)$$

In the expressions above, E_i is the incident wave amplitude; k is the wavevector magnitude in the polymer; R is the distance from the origin to the observation point M (see Fig. 1); subscripts l and l' refer to LC droplet types that differ in terms of shape, size, internal structure, etc.; m is the number of distinct LC droplet types; P_l and $P_{l'}$ denote the partial surface concentrations of droplets of types l and l'; Λ is the mean surface concentration of LC droplets; the pair distribution function $W_{ll'}(\mathbf{r})$ is the probability that droplets of types l and l' are separated by the relative position vector $\mathbf{r}$ in the xy plane; $f_l^{vv}(\mathbf{k}_s)$ and $f_l^{vh}(\mathbf{k}_s)$ are the vv and vh components of the scattering matrix in the $\mathbf{k}_s$ direction for LC droplets of type l; and the asterisk denotes the complex conjugate.

The effect of far-field interference [9, 10] on the forward-scattered intensities I_{inc}^{vv} and I_{inc}^{vh} is represented by the second terms in expressions (1) and (2). This effect increases with the deviation of $S_{ll'}(\mathbf{k}_s)$ from unity, in proportion with the mean droplet concentration.

According to expressions (1)–(3), to analyze the angular distribution of scattered light, the scattering-

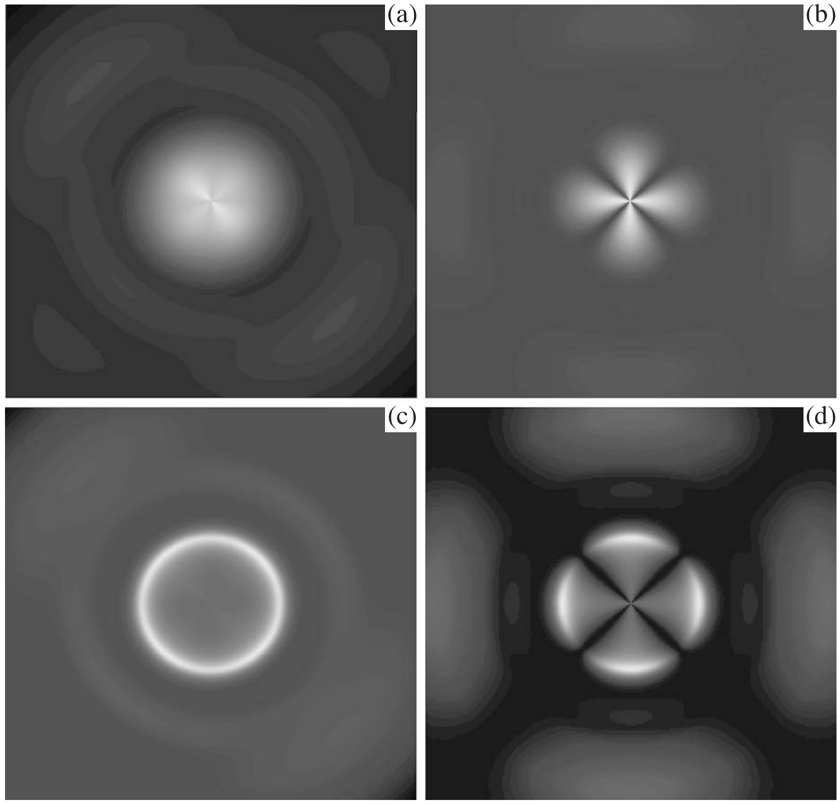

Fig. 3. Forward-scattered intensity distributions $I_{\mathrm{inc}}^{vv}(\theta_s, \varphi_s)$ (a, c) and $I_{\mathrm{inc}}^{vh}(\theta_s, \varphi_s)$ (b, d) for filling factors $\eta = 0.05$ (a, b) and 0.65 (c, d), $\varphi_m = 1°$, $S_d = 0.7$, $S_{fz} = -1/2$, $c = 5$ μm, and $\langle\varphi_d\rangle = 45°$.

matrix components $f_l^{vv}(\mathbf{k}_s)$ and $f_l^{vh}(\mathbf{k}_s)$ must be determined by solving the scattering problem for single LC droplets, and the partial structure factors $S_{ll'}(\mathbf{k}_s)$ must be found by calculating the pair distribution functions $W_{ll'}(\mathbf{r})$. General solution of these problems is a formidable task because of the complexity of external effects on the molecular configuration inside an LC droplet. Consequently, the solutions to inverse scattering problems are also difficult to find [17]. This motivates the use of approximate methods to obtain simplified solutions relating the angular distribution of light scattered from a PDLC monolayer to the orientational structure of the layer and the dispersed LC droplets.

Suppose that the droplet directors $\mathbf{d}_j$ are preferentially aligned, within a cone, along a certain average direction $\langle\mathbf{d}\rangle$, as schematized in Fig. 2. To analyze the angular distribution of light scattered by droplets with cylindrical symmetry, we use an effective-medium approximation [2, 13]. The effective ordinary and extraordinary refractive indices of the droplets, n_{do} and n_{de} [2], are expressed as follows [15, 16]:

$$n_{do} = n_{iso} - \frac{1}{3}\Delta n S_d, \tag{4}$$

$$n_{de} = n_{iso} + \frac{1}{3}\Delta n S_d(1 - 2S_{fz}), \tag{5}$$

where $n_{iso} = (2n_o + n_e)/3$, $\Delta n = n_e - n_o$ (n_o and n_e are the ordinary and extraordinary refractive indices of the LC), S_d is the scalar order parameter of the droplets [2], and S_{fz} is the z component of the tensor order parameter of the PDLC film [14, 16].

The coherent transmittance of a PDLC film evaluated by using the effective-medium approximation is in good agreement with experimental data [15–17]. Satisfactory agreement between theory and experiment can also be expected for small-angle light scattering from PDLC films. To simplify analysis, we consider a monolayer of identical LC droplets with uniform distribution $p(\mathbf{d}_j)$ of their directors within a solid angle $\Delta\Omega$ (see Fig. 2):

$$p(\mathbf{d}_j) = \begin{cases} 1/\Delta\Omega, & \mathbf{d}_j \in \Delta\Omega \\ 0, & \mathbf{d}_j \notin \Delta\Omega. \end{cases} \tag{6}$$

Assuming that the droplets do not coalesce, we use expressions (1)–(3), the effective refractive indices, the anomalous diffraction approximation for large monodisperse spherical LC droplets $2kc(n_{e,o}/n_p - 1) \gg 1$,

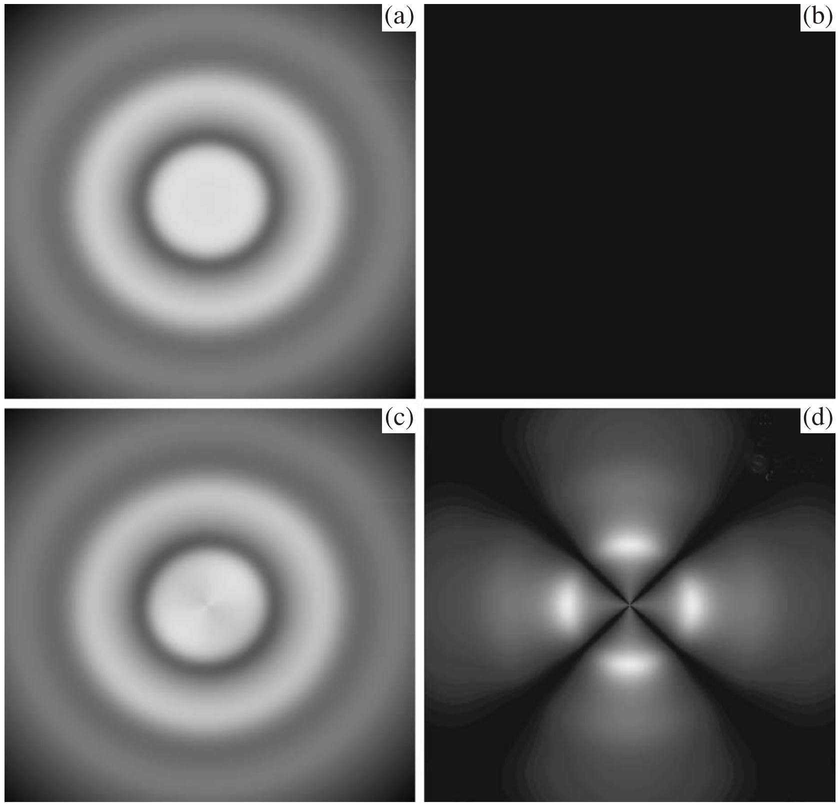

Fig. 4. Forward-scattered intensity distributions $I_{\rm inc}^{vv}(\theta_s, \varphi_s)$ (a, c) and $I_{\rm inc}^{vh}(\theta_s, \varphi_s)$ (b, d) for droplet order parameters $S_d = 0$ (a, b) and 0.01 (c, d), $\varphi_m = 1°$, $\eta = 0.45$, $S_{fz} = -1/2$, $c = 5$ μm, and $\langle\varphi_d\rangle = 45°$.

where c is the droplet radius and n_p is the refractive index of the polymer binder), and the interference approximation to obtain

$$I_{\rm inc}^{vv}(\theta_s, \varphi_s) = C_{vv}\frac{\eta}{\sigma k^2}\langle|f_{vv}(\theta_s, \varphi_s)|^2\rangle S(\theta_s), \quad (7)$$

$$I_{\rm inc}^{vh}(\theta_s, \varphi_s) = C_{vh}\frac{\eta}{\sigma k^2}\langle|f_{vh}(\theta_s, \varphi_s)|^2\rangle S(\theta_s), \quad (8)$$

$$\begin{aligned}\langle|f_{vv}(\theta_s, \varphi_s)|^2\rangle &= \frac{k^4c^4}{4}\{|H_e(\theta_s)|^2\langle\cos^4(\varphi_s-\varphi_d)\rangle \\ &+ |H_o(\theta_s)|^2\langle\sin^4(\varphi_s-\varphi_d)\rangle + \frac{1}{2}({\rm Re}H_e(\theta_s){\rm Re}H_o(\theta_s) \\ &+ {\rm Im}H_e(\theta_s){\rm Im}H_o(\theta_s))\langle\sin^2 2(\varphi_s-\varphi_d)\rangle\},\end{aligned} \quad (9)$$

$$\begin{aligned}\langle|f_{vh}(\theta_s, \varphi_s)|^2\rangle &= \frac{k^4c^4}{16}\{|H_e(\theta_s)|^2 + |H_o(\theta_s)|^2 \\ &- 2({\rm Re}H_e(\theta_s){\rm Re}H_o(\theta_s) \\ &+ {\rm Im}H_e(\theta_s){\rm Im}H_o(\theta_s))\} \\ &\times\langle\sin^2 2(\varphi_s-\varphi_d)\rangle,\end{aligned} \quad (10)$$

$$\begin{aligned}\langle\cos^4(\varphi_s-\varphi_d)\rangle &= \frac{3}{8} + \frac{1}{2}\cos 2(\varphi_s - \langle\varphi_d\rangle){\rm sinc}2\varphi_m \\ &+ \frac{1}{8}\cos 4(\varphi_s - \langle\varphi_d\rangle){\rm sinc}4\varphi_m,\end{aligned} \quad (11)$$

$$\begin{aligned}\langle\sin^4(\varphi_s-\varphi_d)\rangle &= \frac{3}{8} - \frac{1}{2}\cos 2(\varphi_s - \langle\varphi_d\rangle){\rm sinc}2\varphi_m \\ &+ \frac{1}{8}\cos 4(\varphi_s - \langle\varphi_d\rangle){\rm sinc}4\varphi_m,\end{aligned} \quad (12)$$

$$\begin{aligned}&\langle\sin^2 2(\varphi_s-\varphi_d)\rangle \\ &= \frac{1}{2}(1 - \cos 4(\varphi_s - \langle\varphi_d\rangle)\sin 4\varphi_m),\end{aligned} \quad (13)$$

$$\begin{aligned}H_{e,o}(\theta_s) &= 2\int_0^1(1-\exp(i v_{\rm de,do}\sqrt{1-u^2})) \\ &\times J_0(zu)udu.\end{aligned} \quad (14)$$

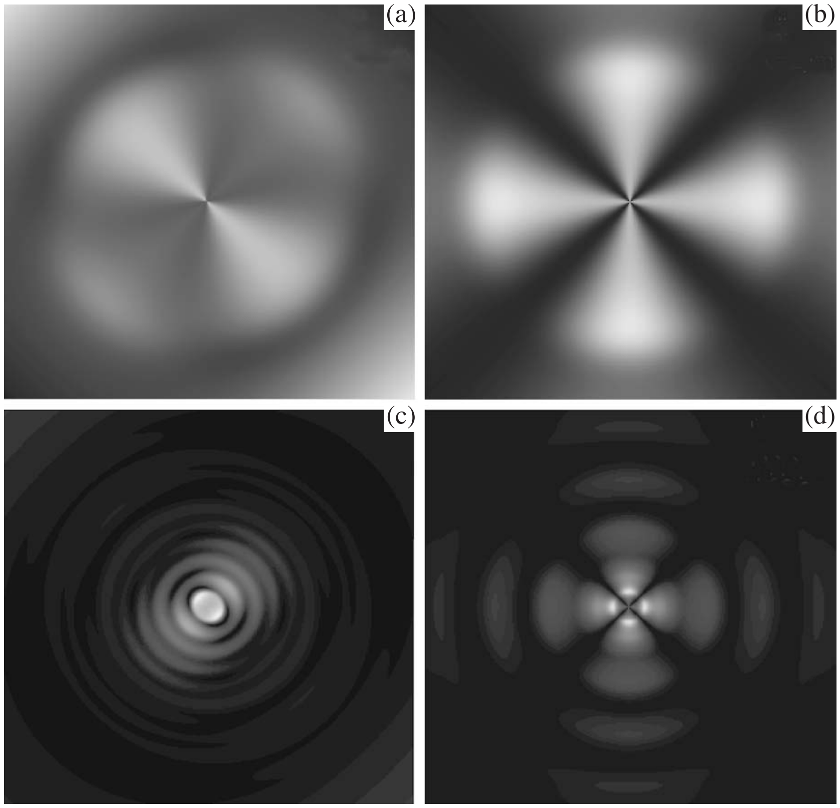

Fig. 5. Forward-scattered intensity distributions $I_{\rm inc}^{vv}(\theta_s, \varphi_s)$ (a, c) and $I_{\rm inc}^{vh}(\theta_s, \varphi_s)$ (b, d) for droplet order radii $c = 2$ (a, b) and 20 (c, d) μm, $\varphi_m = 1°$, $\eta = 0.45$, $S_d = 0.7$, $S_{fz} = -1/2$, and $\langle\varphi_d\rangle = 45°$.

Here, angle brackets denote averaging over droplet director orientation;

$$z = kc\sin\theta_s; \quad v_{\rm de,\,do} = 2kc(n_{\rm de,\,do}/n_p - 1);$$

$$\sigma = \pi c^2;$$

η is the filling fraction defined as the ratio between the cross-sectional area of the droplets projected onto the xy plane and the PDLC film area ($\eta = N\sigma/A$ for monodisperse droplets); the effective ordinary and extraordinary refractive indices of a droplet are given by expressions (4) and (5), respectively; and the parameters C_{vv} and C_{vh} are determined by experimental conditions, depending on the light source intensity, detector sensitivity, and the distance between the examined sample and the detector.

The structure factor $S(\theta_s)$ contained in expressions (7) and (8) can be calculated numerically [9, 11]. We use the Percus–Yevick approximation for monodisperse hard disks proposed in [18]. This approximation provides a relatively simple tool for analyzing first-order concentration effects on the angular distribution of scattered light and simplifies solution of inverse problems [9]. In the general case of a polydisperse PDLC film, a numerical solution of the Ornstein–Zernike equation [10]. Using the Percus–Yevick approximation, we express the structure factor as

$$S(\theta_s) = \left\{1 + \frac{4\eta}{1-\eta}\frac{2J_1(2z)}{2z} + \frac{4\eta^2}{(1-\eta)^2}J_0(z)\frac{2J_1(z)}{z} + \left(\frac{\eta^2}{(1-\eta)^2} + \frac{2\eta^3}{(1-\eta)^3}\right)\left[\frac{2J_1(z)}{z}\right]^2\right\}^{-1}, \quad (15)$$

where J_0 and J_1 are the zeroth- and first-order Bessel functions of the first kind, respectively.

The method developed here can be used to examine the dependence of the angular distribution of light scattered from droplet monolayers and single droplets on droplet composition, structure, and orientation and determine layer and droplet characteristics from the angular distribution of forward-scattered light. The scope of this approach is illustrated by several numerical solutions to the direct scattering problem presented in the next section.

3. RESULTS

Figures 3–6 represent the angular scattered intensity distributions $I_{\rm inc}^{vv}(\theta_s, \varphi_s)$ and $I_{\rm inc}^{vh}(\theta_s, \varphi_s)$ calculated by using expressions (7)–(15) for $0 < \theta_s \le 7°$ and $0 \le \varphi_s \le 360°$. Here, both parallel and perpendicular intensities

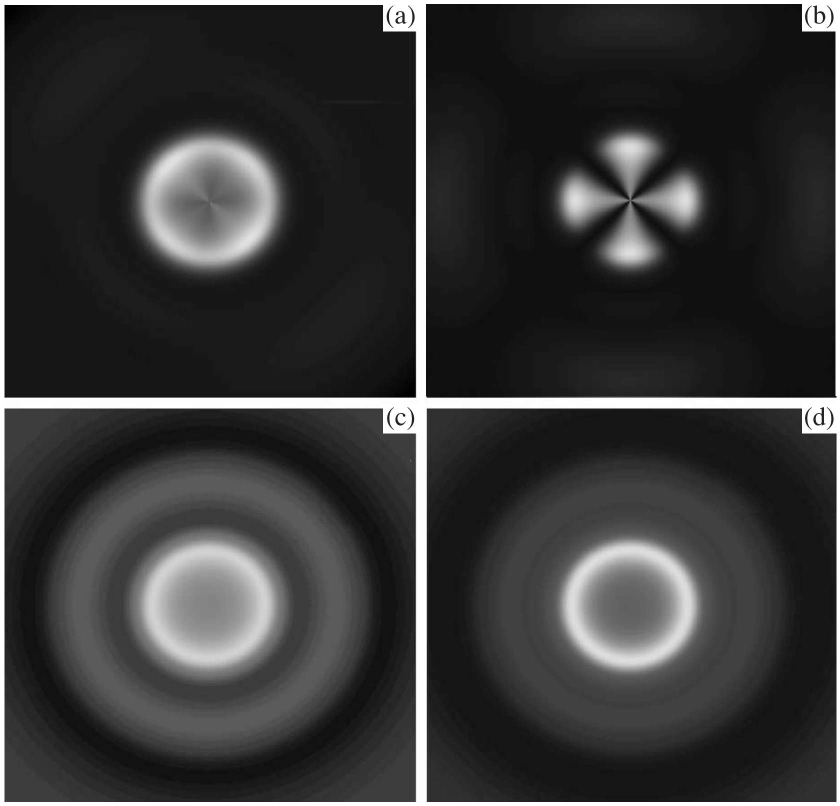

Fig. 6. Forward-scattered intensity distributions $I_{\text{inc}}^{vv}(\theta_s, \varphi_s)$ (a, c) and $I_{\text{inc}}^{vh}(\theta_s, \varphi_s)$ (b, d) for LC droplet director deviation angles $\varphi_m = 1°$ (a, b) and 90° (c, d) μm, $S_{fz} = -1/2$ (a, b) and 0 (c, d), $\eta = 0.45$, $S_d = 0.7$, $c = 5$ μm, and $\langle\varphi_d\rangle = 45°$.

are measured in arbitrary units and the grayscale brightness increases with intensity.

The calculations are performed for an incident wavelength of 0.6328 μm, $n_o = 1.5183$, $n_e = 1.7378$, and $n_p = 1.524$. The distributions presented in Figs. 3–5 are obtained for planar droplet director orientation (when $S_{fz} = -1/2$), director cone with $2\varphi_m = 2\theta_m = 2°$, and azimuthal angle $\langle\varphi_d\rangle = 45°$ of preferred droplet director orientation. (When φ_m is larger, except for the case of random orientation of droplet directors, other components of the tensor order parameter must be taken into account [16].) These images illustrate the dependence of forward-scattered intensity distribution on the filling fraction η (Fig. 3), the droplet order parameter S_d (Fig. 4), and the droplet radius c (Fig. 5). Figure 6 demonstrates the difference in angular scattered intensity distribution between the cases of aligned and randomly oriented directors ($\varphi_m = 1°$ and 90°, respectively).

With increasing droplet concentration, the direction of maximum scattering deviates from the incident wavevector [9]. Figure 3 illustrates the amount of deviation due to the change from $\eta = 0.05$ to $\eta = 0.65$ for the vv and vh components of scattered intensity. In the angular distribution of the vv component, the deviation is observed at all azimuthal angles φ_s. The azimuthal distribution of the vh component is highly nonuniform, and the polar angle of maximum scattered intensity increases with the filling factor around each azimuthal angle of maximum scattering.

Figure 4 illustrates the effect of the molecular configuration inside an LC droplet on the scattered light intensity. It is clear that a small variation of S_d may cause a drastic change in the vh component. The highest sensitivity to the molecular configuration in droplets is observed in the neighborhood of $S_d = 0$. When the LC molecules are randomly oriented ($S_d = 0$), zero transmission of scattered light through crossed polarizers is observed (Fig. 4b). Even a slight ordering of molecules in LC droplets (Fig. 4d) drastically increases transmission through crossed polarizers, which can serve as an indicator of liquid crystal phase transition.

An increase in droplet radius narrows the angular distribution of forward-scattered light. Figure 5 shows the numerical results obtained for monolayers of small and large LC droplets with $c = 2$ and 20 μm, respectively.

The azimuthal anisotropy of scattered light decreases with increasing orientational disorder of droplets in a monolayer. Figure 6 compares the angular distributions of scattered light obtained for films with ordered and random LC droplet orientations ($\varphi_m = 1°$, $S_{fz} = -1/2$ and $\varphi_m = 90°$, $S_{fz} = 0$, respectively).

Note the symmetry of the scattered light distributions about the diagonal axes in Figs. 3–6. It is due to the choice of $\langle\varphi_d\rangle = 45°$ as a mean azimuthal angle of droplet director orientation.

The locations, geometries, and relative areas of dark and bright regions ("rings" and "crosses") are determined by the following factors:

(i) cooperative interference effects leading to redistribution of scattered intensity;

(ii) degree of ordering of droplet directors in a monolayer;

(iii) parameters of the droplets (c and S_d) and the liquid crystal.

The solutions to direct scattering problems obtained here can be used in analyzing inverse scattering problems to determine LC droplet size and orientation, as well as the LC and polymer concentrations required to create films with desired characteristics.

For filling factors and droplet sizes sufficiently small that the Rayleigh–Gans approximation is applicable [6], the results of the proposed analysis of the angular distribution of light scattered by PCLC monolayers are qualitatively consistent with those obtained in [19, 20] for single LC droplets with bipolar and axial configurations.

4. CONCLUSIONS

A method is developed for modeling and computing the angular distribution of light scattered forward from a single-layer polymer-dispersed liquid-crystal (PDLC) film. The calculated results illustrate the dependence of the distribution of forward-scattered light on the droplet concentration and size and the droplet and film order parameters.

The method relies on the use of anomalous diffraction approximation and effective refractive indices of LC droplets and takes into account cooperative scattering effects. The method provides a tool for examining the distribution of molecules in a liquid-crystal droplet by means of polarized optical microscopy. It can be used to study field- and temperature-induced phase transitions in LC droplets with cylindrical symmetry (bipolar, axial and other configurations of molecules in a droplet) by analyzing the angular distribution of forward-scattered light.

Our results can be applied in developing various devices based on polymer-dispersed liquid-crystal films (optical amplitude and phase modulators, polarization converters, displays, etc.), with response due to changes in LC configuration caused by external factors.

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