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Publisher: Taylor & Francis

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Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:

http://www.tandfonline.com/loi/gmcl19

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To cite this article: I. P. Pinkevich, V. Yu. Reshetnyak & L. G. Grechko (1994): The Ifluence of Nematic Droplet Optical Axes Ordering on Light Scattering in Polymer Matrix, Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals, 251:1, 283-287

To link to this article: http://dx.doi.org/10.1080/10587259408027211

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Mol. Cryst. Liq. Cryst. 1994, Vol. 251, pp. 283-287 Reprints available directly from the publisher Photocopying permitted by license only © 1994 Gordon and Breach Science Publishers S.A. Printed in the United States of America

THE IFLUENCE OF NEMATIC DROPLET OPTICAL AXES ORDERING ON LIGHT SCATTERING IN POLYMER MATRIX

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<u>Abstract</u>. The dependence of the light scattering cross-sections by spherical and ellipsoidal droplets on the droplet optical axes ordering is considered in Rayleigh - Gans approximation.

INTRODUCTION

Investigation of nematic liquid crystals (NLC) dispersed in a polymer matrix are steady intensificating [1]. It is connected with two basic circumstances. First, polymer dispersed liquid crystals (PDLC) form a new class of heterogeneous LC-systems with very developed surface and thus new interesting effects are waited in the future. Second, PDLC present a material of large importance for electrooptics due to the possibility to control its scattering properties.

The scattering properties of spherical nematic droplet were considered in Rayleigh-Gans approximation in paper [2]. The optical anisotropy of the droplet was taken into account and expressions for the scattering matrix, the differential and light scattering cross-sections were obtained. Light scattering by spherical nematic droplet of micron size was studied in paper [3] in anomalous-diffraction approach. The several director configuration in the droplet were considered. It was determined the strong dependence of scattering cross-sections on the director configuration. In paper [4] the light scattering by a set of spherical nematic droplets was investigated taking into account the correlation in droplet space distribution. In the work [5] it was noted that characteristics of opaque-transparent transition should be strongly dependent on the mutual orientation of a director in separate LC-droplets. In the present work it is theoretically analyzed the influence of ordering of the nematic droplet optical axes on the light scattering cross-section in polymer matrix. In the Rayleigh-Gans approximation it is obtained the analytical expressions for scattering cross-section of light by nematic spherical droplets with homogeneous distribution of director inside droplets and ellipsoidal droplets containing isotropic liquid.

GENERAL EXPRESSION FOR SCATTERING CROSS-SECTION

In Rayleigh-Gans approximation the light scattering cross-section by nematic droplet in a homogeneous isotropic medium can be written as

$$\left(\frac{\partial \sigma}{\partial \Omega}\right)_{\parallel,\perp} = \frac{q^4}{16\pi^2} \left| \overrightarrow{i}_{\parallel,\perp} \widehat{D} e \right|^2 \tag{1}$$

where

$$\widehat{D} = \int dV \left(\frac{\widehat{\epsilon}(\overrightarrow{r})}{\epsilon_p} - \widehat{I} \right) e^{-i\overrightarrow{q}_s \overrightarrow{r}}, \ \overrightarrow{q}_s = \overrightarrow{q}' - \overrightarrow{q}$$
 (2)

Here \overrightarrow{i}' , \overrightarrow{e} are the unit vectors denoting the polarization of the scattered and incident light waves, \overrightarrow{q}' , \overrightarrow{q} are the wave vectors of these waves, $\widehat{\epsilon}$, ϵ_p are the dielectric susceptibilities of the droplet substance and isotropic medium (polymer matrix), \widehat{I} is the unit tensor, symbols \parallel,\perp denote the scattered wave polarization in the scattering plane (wave vectors \overrightarrow{q}' , \overrightarrow{q} plane) and perpendicular to it. In order to have the tensor \widehat{D} in the form more convenient for analysis we pass from the Cartesian components of tensor $\widehat{\epsilon}$ to its spherical counterparts (in a laboratory coordinate system):

$$\epsilon_{ij} = \sum_{\sigma,\rho} A_{ij}^{\sigma\rho} \epsilon_{lab}^{\sigma\rho} \tag{3}$$

We take into account the connection between the spherical components of tensor $\hat{\epsilon}$ in the laboratory and own droplet coordinate systems:

$$\epsilon_{lab}^{\sigma\rho} = \sum_{\rho'} D_{\rho\rho'}^{\sigma}(\Omega) \, \epsilon^{\sigma\rho'}, \tag{4}$$

where $D^{\sigma}_{\rho\rho'}(\Omega)$ is the Wigner's -function, Ω is the set of Euler angles; in the own droplet coordinate system the tensor $\hat{\epsilon}$ is diagonal. In addition we expand $\exp(-i\overrightarrow{q}_s\overrightarrow{r})$ into a spherical functions series:

$$e^{-i\overrightarrow{q}_{s}\overrightarrow{r}} = 4\pi \sum_{lm} (i)^{l} I_{l}(q_{s}r) Y_{lm}(\Omega_{q_{s}}) Y_{lm}(\Omega_{r}), \qquad (5)$$

where I_l is Bessel's spherical function, $Y_{lm}(\Omega_a)$ is the spherical function of the vector \overrightarrow{a} angles in a laboratory coordinate system. Substituting expressions (3),(4),(5) into formula (2) we obtain:

$$D_{ij} = 4\pi \sum_{lm} \sum_{\sigma \rho \rho'} (i)^l \int \int \int I_l(q_s r) Y_{lm} (\Omega_q) Y_{lm} (\theta_r, \phi_r)$$

$$\cdot \left[A_{ij}^{\sigma\rho} D_{\rho\rho'}^{\sigma} \epsilon_{p}^{-1} \epsilon^{\sigma\rho'} - \delta_{ij} \right] r^{2} dr \sin \theta_{r} d\theta_{r} d\phi_{r} \tag{6}$$

We use then expression (6) for calculating the differential light scattering crosssection by NLC droplets of given form

LIGHT SCATTERING BY ANISOTROPIC SPHERICAL DROPLET

Let we have a spherical NLC droplet wherein the director distribution is uniform due to the boundary conditions on its surface. Besides, in Rayleigh-Gans approach we may approximately consider the orientational order parameter inside droplet to be constant, [2]. In this case NLC dielectric susceptibility tensor is uniaxial and coordinates-independent. We assume all the droplets to be of the same radius R but of different orientation of own coordinate system relatively to laboratory one. Since the tensor ϵ_{ij} is uniaxial, then the own droplet coordinate system distribution function depends only on the angle θ between the droplet optical axis and the preferred direction of the axes. We assume the axis OZ of the laboratory coordinate system to be directed along the latter.

Substituting expression (6) in formula (1), (2) and realizing the integration we average then the differential scattering cross-section over all orientations of the droplet optical axis with distribution function $f(\theta)$. As result, the averaged value $\langle \frac{d\sigma}{d\Omega} \rangle$ is determined by the expression (dropping out in (1) the factor $\frac{q^4}{16\pi^2}$ identical for all the droplets):

$$\langle D_{ij}D_{kl} \rangle = \left(\frac{16\pi^{2}}{q^{3}}\right)^{2} \left(\sin q_{s}R - q_{s}R\cos q_{s}R\right)^{2}$$

$$\cdot \left\{\delta_{kl}\delta_{ij}\left(\frac{1}{3\epsilon_{p}}Sp\hat{\epsilon} - 1\right) \left[\left(\frac{1}{3\epsilon_{p}}Sp\hat{\epsilon} - 1\right) + \frac{1}{3\epsilon_{p}}\left(\epsilon_{3} - \epsilon_{1}\right) \langle P_{2}(\cos\theta) \rangle \right] \right\}$$

$$\cdot \left(2\delta_{iz} + 2\delta_{kz} - \delta_{ix} - \delta_{kx} - \delta_{iy} - \delta_{ky}\right) + \frac{2}{3\epsilon_{p}^{2}}\left(\epsilon_{3} - \epsilon_{1}\right)^{2} \left[\frac{1}{6}\delta_{kl}\delta_{ij}\left(2\delta_{iz} - \delta_{ix} - \delta_{iy}\right) \right]$$

$$\cdot \left(2\delta_{kz} - \delta_{kx} - \delta_{ky}\right) \langle P_{2}(\cos\theta) \rangle + \sum_{\rho=-2}^{2} A_{ij}^{2\rho}A_{kl}^{2\rho}\sum_{L=0,2,4}\left(22 - \rho\rho \mid L0\right)$$

$$\cdot \left(2200 \mid L0\right) \langle P_{L}(\cos\theta) \rangle \right]$$

$$(7)$$

where symbol < ... > denotes averaging with function $f(\theta)$, ϵ_i is a main value of dielectric susceptibility tensor, $P_L(\cos \theta)$ is Legendre's polynomial, $(22 - \rho \rho \mid L0)$ are the Clebsh-Giordane coefficients, δ_{ij} is Kroneker's symbol. The coefficients $A_{ij}^{\sigma\rho}$ may be calculated, for example, as in paper [6].

In the case of isotropic droplets $\epsilon_3 = \epsilon_1$ and formula (7) reduces to the well-known expression for scattering by a homogeneous isotropic sphere.

Thus, as it follows from expression (7), the orientational ordering of the droplet optical axes leads to the additional contribution to the differential light scattering cross-section. This contribution is proportional to dielectric susceptibility anisotropy $\epsilon_a = \epsilon_3 - \epsilon_1$ and the optical axes ordering degree, described by the parameters $\langle P_L(\cos\theta) \rangle$. As the result, the ratio between the values of differential scattering cross-section of differently polarized light beams changes. So, if the light scattering plane is perpendicular to the preferred direction of the droplet optical axes then in the case $\epsilon_a \ll |\epsilon_p - \epsilon_i|$ we obtain:

$$\frac{\left\langle \left(\frac{d\sigma}{d\Omega}\right)_{\parallel} \right\rangle}{\left\langle \left(\frac{d\sigma}{d\Omega}\right)_{\parallel} \right\rangle} = \frac{\left\langle D_{xx}^{2} \right\rangle}{\left\langle D_{zz}^{2} \right\rangle} = 1 - 2\frac{\epsilon_{a}}{\epsilon_{p}} \frac{\left\langle P_{2}(\cos\theta) \right\rangle}{\frac{1}{3\epsilon_{p}} Sp\hat{\epsilon} - 1}$$
(8)

If NLC anisotropy is not small ($\epsilon_a \sim |\epsilon_p - \epsilon_i|$) then in expression (7) the last term must be taken into account. In this case, the scattering cross-section of light with change of its polarization differs from zero, as can be easily derived from expressions (1), (7). In particular, if the incident light is polarized along the OZ axis perpendicular to the scattering plane and the scattered light polarization vector lies in the scattering plane (XOY plane) then the differential cross-section equals:

$$<\left(\frac{d\sigma}{d\Omega}\right)_{\parallel}> = \frac{16\pi^2q^4}{q_s^6}\left(\sin q_s R - q_s R\cos q_s R\right)^2 \frac{2}{3}\left(\frac{\epsilon_a}{\epsilon_p}\right)^2 \sum_{\rho=1,-1} \left|A_{xz}^{2\rho}\right|$$

As it is seen from (7), (9), if $\epsilon_a \sim |\epsilon_p - \epsilon_i|$ then the scattering cross-section of light with change of its polarization can be comparable in value with that of light scattered without change of polarization.

The expressions obtained above may be used for determining the degree of influence of droplet optical axes ordering on the PDLC transparence at the switching between its opaque and transparent states. The voltages necessary for switching ($\sim\!10\mathrm{V}$) change the value of parameters $< P_L(\cos\theta)>$ but don't affect practically the value of ϵ_i . The dependence of light scattering cross-sections in the PDLC on droplet optical axes ordering was noted in paper [5] and observed experimentally also in [7].

LIGHT SCATTERING BY ISOTROPIC ELLIPSOIDAL DROPLET

In this case droplet dielectric susceptibility is scalar quantity ϵ . Using the expressions (2), (5) and taking integration in (2) one can obtain

$$\langle D_{ij}D_{kl} \rangle = \left[16\pi^2 \left(\frac{\epsilon}{\epsilon_p} - 1\right)\right]^2 \delta_{ij}\delta_{kl} \langle \left[\frac{a_1a_2a_3}{\tilde{q}_s^3} \left(\sin\tilde{q}_s - \tilde{q}_s\cos\tilde{q}_s\right)\right]^2 \rangle \tag{10}$$

where $\tilde{q}_s = \left[(q_{sx'}a_1)^2 + (q_{sy'}a_2)^2 + (q_{sz'}a_3)^2 \right]^{1/2}$, $q_{si'}$ are the Cartesian components of vector \overrightarrow{q}_s in the coordinate system of the ellipsoid, a_1 , a_2 , a_3 denote the length of ellipsoid appropriate semiaxes.

Restricting ourselves to the ellipsoids of rotation $(a_1 = a_2)$ and assuming $q_s a_3 \ll 1$ we can realize averaging in formula (10) and obtain the following expression:

$$\langle D_{ij}D_{kl} \rangle = \left[\frac{16\pi^2}{3}\left(\frac{\epsilon}{\epsilon_p} - 1\right)\right]^2 \delta_{ij}\delta_{kl}a_1^2a_3$$

$$\cdot \left\{ 1 - \frac{(a_3 q_s)^2}{5} \left[\sigma^2 + \left(1 - \sigma^2 \right) < \cos^2 \beta > \right]^2 \right\}$$
 (11)

where $\sigma = \frac{a_1}{a_3}$, β is the angle between the ellipsoid rotation axis and the direction of scattering vector \overrightarrow{q}_s .

In order to find the total scattering cross-section it is convenient to direct the axis OZ of laboratory coordinate system along the incident light wave vector \overrightarrow{q} . Then, averaging expression (1) over the ellipsoid long axis orientations, we substitute expression (11) into it and sum up over scattered light polarization. Assuming the condition $q_s a_3 \ll 1$ to be fulfilled for all the q_s , we integrate the resulting expression with respect to the scattering angles. As the result, we have the following expression for the total scattering cross-section of the incident light polarized along the axis OX:

$$\sigma_t = 2\pi q^4 \left[\frac{4\pi}{3} \left(\frac{\epsilon}{\epsilon_p} - 1 \right) \right]^2 a_1^2 a_3 \left\{ 1 - \frac{1}{5} \left(q_s a_3 \sigma \right)^2 + \frac{1}{10} \left(1 - \sigma^2 \right) \left[\frac{7}{3} + \langle \cos^2 \beta \rangle \right] \right\}$$
(12)

If the incident beam is unpolarized, then the similar calculations yield

$$\sigma_t = 2\pi q^4 \left[\frac{16\pi}{9} \left(\frac{\epsilon}{\epsilon_p} - 1 \right) \right]^2 a_1^2 a_3 \left\{ 1 - \frac{\sigma^2}{4} \left[1 + \frac{3}{5} q_s^2 a_3^2 \right] \right\}$$
 (13)

Thus, the light scattering cross-sections are essentially dependent on the shape of ellipsoidal droplets and in the case of polarized incident light on the orientational ordering of droplet long axes, described by the parameter $<\cos^2\beta>$, too.

ACKNOWLEDGEMENT

The research described in this publication was made possible in part by Grant from the International Science Foundation.

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