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## Direct Observation of the Orientational Fluctuations in a Nematic Liquid Crystal with a High-Speed Camera

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The orientational fluctuations in a homogeneously aligned nematic liquid crystal were successfully observed with a high-speed video camera. From the series of images obtained, first the spatial Fourier transformation of each image was computed and then the time correlation function at each wave number was calculated. A theory to analyse the results was developed and by using it the ratios of the elastic constant to the viscosity were obtained.

Keywords: fluctuation; direct observation; nematic liquid crystal; high-speed TV camera

### INTRODUCTION

Orientational fluctuation is one of the most remarkable characteristics of liquid crystals. In the nematic liquid crystal and some kinds of tilted smectic liquid crystals such as SmC, there is a large, slow orientational fluctuation due to the Goldstone mode. So far, the fluctuation dynamics has been studied mainly by the photon correlation spectroscopy, in which the intensity fluctuation of the light scattered by the sample is analyzed with a correlator to calculate the time correlation function [1,2]. This measurement is carried out in reciprocal space.

Quite recently, on the other hand, we have succeeded in directly observing orientational fluctuations of a tilted smectic liquid crystal in the real space with a high speed video camera [3]. From the series of images obtained under a polarizing microscope, the spatial Fourier transformation of each image was computed and then the time correlation function at each wave number was calculated. It was demonstrated that from this analysis the ratio of the elastic constant to the viscosity can be obtained.

In this paper we apply the above-mentioned method, called dynamic image analysis, to a homogeneously aligned nematic liquid crystal. However, the expression of the intensity used for the smectic liquid crystal is not enough for a nematic liquid crystal. Therefore, we derive it on the basis of Maxwell's equations in the next section.

### THEORY

We consider a homogeneously aligned namatic liquid crystal, where the average direction of the director is taken along the x-axis and the incident light is along z-axis, and the lower and upper interfaces between the liquid crystal and the glasses are at z=0 and d, respectively. In nematic liquid crystals the dielectric tensor is given in terms of the director n:

$$\boldsymbol{\varepsilon}_{\alpha\beta} = \boldsymbol{\varepsilon}_{\perp} + \boldsymbol{\varepsilon}_{a} \boldsymbol{n}_{\alpha\beta} , \qquad (1)$$

where  $\varepsilon_u = \varepsilon_{\parallel} - \varepsilon_{\perp}$  is the dielectric anisotropy, and  $\varepsilon_{\parallel}$  is the dielectric constant parallel to the director and  $\varepsilon_{\perp}$  perpendicular to it. In our geometry,

$$\varepsilon = \varepsilon^{(0)} + \Delta \varepsilon \tag{2}$$

with

$$\varepsilon^{(0)} = \begin{pmatrix} \varepsilon_{\parallel} & & \\ & \varepsilon_{\perp} & \\ & & \varepsilon_{\perp} \end{pmatrix} \text{ and } \Delta \varepsilon = \varepsilon_{a} \begin{pmatrix} 0 & \Delta n_{y} & \Delta n_{z} \\ \Delta n_{y} & 0 & 0 \\ \Delta n_{z} & 0 & 0 \end{pmatrix}, \tag{3}$$

where  $\Delta n_y$  and  $\Delta n_z$  are the fluctuation parts of the director. Taking into account that the frequency of light should much higher than that of the director fluctuation, Maxwell's equations become

$$\operatorname{rot} \mathbf{E} = i\omega \mu_0 \mathbf{H} ,$$

$$\operatorname{rot} \mathbf{H} = -i\omega \varepsilon_0 \varepsilon \mathbf{E} , \tag{4}$$

where  $\omega$  is the angular frequency of light and  $\varepsilon_0$  and  $\mu_0$  are respectively the dielectric and magnetic permitivities in vacuum. We can solve Eqs. (4) with Eqs. (2) by using the perturbation method. After some mathematical manipulations, when the angle between the polarizer and the average director (x-axis) is  $\alpha$  and the field strength at the polarizer is  $E_0$ , the field strength on the upper interface observed through the analyzer,  $E_a(x,y,d)$ , is given as

$$E_{a}(x,y,d) = \frac{1}{2} \left( e^{iq_{\parallel}d} - e^{iq_{\perp}d} \right) \sin 2\alpha E_{0} - i \frac{\pi d\varepsilon_{a}}{\lambda}$$

$$\times \sum_{q_{x},q_{y}} \left( \frac{1}{\sqrt{\varepsilon_{\parallel}}} \Delta n_{yq_{*}} e^{iq_{\parallel}d} \sin^{2}\alpha - \frac{1}{\sqrt{\varepsilon_{\perp}}} \Delta n_{yq_{*}} e^{iq_{\perp}d} \cos^{2}\alpha \right) e^{i(q_{x}x + q_{y}y)} E_{0}$$
(5)

with

$$q_{\parallel,\perp} = \sqrt{\varepsilon_{\parallel,\perp}} \omega/c, \ \Delta q = q_{\parallel} - q_{\perp}, \ \boldsymbol{q}_{\pm} = (q_x, q_y, \pm \Delta q), \tag{6}$$

where  $\Delta n_{,q}$  is the Fourier transform of  $\Delta n_{,q}(x,y,z)$ . Therefore, the intensity  $I_a(x,y,d)=|E_a(x,y,d)|^2$  on the upper interface is

$$I_a(x, y, d) = \sin^2 \frac{\Delta q d}{2} \sin^2 2\alpha E_0^2 + \sum_{q_x, q_y} I_{q_x, q_y} e^{i(q_x x + q_y, y)}, \quad (7)$$

with

$$\begin{split} I_{q_{x},q_{v}} &= \frac{\pi d \varepsilon_{a}}{2\lambda} \sin 2\alpha \\ &\times \left( \frac{1}{\sqrt{\varepsilon_{\parallel}}} \sin^{2} \alpha - \frac{1}{\sqrt{\varepsilon_{\perp}}} \cos^{2} \alpha \right) \left\{ (1 - \mathrm{e}^{-i\Delta q d}) \Delta n_{yq_{\perp}} - (1 - \mathrm{e}^{i\Delta q d}) \Delta n_{yq_{\perp}} \right\} E_{0}^{2}. \end{split} \tag{8}$$

Thus, taking into account that  $\Delta n_{yq_z}$  depends on time, the time correlation function of the Fourier coefficient of the observed intensity is given as

$$C_{q_{\tau},q_{\tau}}(t) = \left\langle I_{q_{\tau},q_{\tau}}(t)^{*} I_{q_{\tau},q_{\tau}}(0) \right\rangle = 2 \left( \frac{\pi d \varepsilon_{u}}{\lambda} \right)^{2}$$

$$\times \sin^{2} \frac{\Delta q d}{2} \sin^{2} 2\alpha \left( \frac{1}{\sqrt{\varepsilon_{\parallel}}} \sin^{2} \alpha - \frac{1}{\sqrt{\varepsilon_{\perp}}} \cos^{2} \alpha \right) \left| \left| \Delta n_{yq_{+}} \right|^{2} \right\rangle e^{-t/\tau} E_{0}^{4}$$
(9)

where for  $1/\tau$  along x-axis  $(q_x = 0)$  [4]

$$\frac{1}{\tau} = (K_2 \Delta q^2 + K_3 q_x^2) / \left( \gamma_1 - \frac{\alpha_2^2 q_x^2}{\eta_a \Delta q^2 + \eta_c q_x^2} \right) 
\approx \frac{K_2}{\gamma_1} \Delta q^2 + \frac{K_3}{\gamma_1} \left( 1 + \frac{K_2 \alpha_2^2}{K_3 \gamma_1 \eta_a} \right) q_x^2, \tag{10}$$

for  $1/\tau$  along y-axis  $(q_r = 0)$  [4]

$$\frac{1}{\tau} = \frac{K_2}{\gamma_1} \Delta q^2 + \frac{K_2}{\gamma_1} q_y^2. \tag{11}$$

### EXPERIMENTAL AND RESULTS

The sample used was a nematic liquid crystal 5CB, which was sandwiched between two glass plates with polyimide layers. The film thickness d was 50 $\mu$ m. The homogeneous cell was observed under a polarizing microscope (Olympus, BH-2) with a high-speed video camera (Photoron, Fastcam-Rabbit) at 30°C in the nematic phase. The angle between the rubbing direction and the polarizer was set to be 10°. To obtain a monochromatic light we used an interference filter (550nm).

In the measurement, 1090 frames were recorded at a rate of 240 frames/sec with the high-speed video camera. The captured images were transferred to an image processing system (Carl Zeiss, Vidas Plus) and digitized into  $256 \times 256$  pixels with 256 gray scales, and then FFT of the images was made. Defining the intensity of the image at the position (x,y) and the time t as I(x,y,t), the Fourier coefficient is given as

$$I_{q_x,q_y}(t) = \frac{1}{S} \iint I_a(x,y,d,t) \exp[-i(q_x x + q_y y)] dx dy, \qquad (12)$$

where S is the sample area within the frame. Then, the time correlation function of the Fourier coefficient was calculated as

$$C_{q_{x},q_{y}}(t) = \left\langle I_{q_{x},q_{y}}(t)^{*} I_{q_{x},q_{y}}(0) \right\rangle. \tag{13}$$

The above time correlation function has been theoretically obtained in the previous section.

In Figure 1 we show a typical time correlation function at  $q_x = 0.24 [\mu \text{m}^{-1}]$  along x-axis  $(q_y = 0)$ . As is expected from Eq. (9), the data almost fall on an exponential curve, which was obtained by fitting. Figure 2 shows the dependencies of  $1/\tau$  on the squares of  $q_x$  and  $q_y$ . Equation (11) indicates that we can obtain  $K_2/\gamma_1 \cdot \Delta q^2$  and  $K_2/\gamma_1$  from the intercept and the slope of  $1/\tau$  vs.  $q_x^2$ , respectively, in Figure 2. Furthermore, we can get the coefficient of  $q_x^2$  in Eq. (10) from the slope of  $1/\tau$  vs.  $q_x^2$  in Figure 2. The results are:  $K_2/\gamma_1 = 70 \pm 15$  [ $\mu$ m²/sec],  $\Delta q = 1.6 \pm 0.3$  [ $\mu$ m¹] and  $K_3/\gamma_1 \cdot (1 + \alpha_2^2/\gamma_1 \eta_a) = 250 \pm 50$  [ $\mu$ m²/sec], which are in good agreement with the values calculated from the other measurements [5,6,7], 60, 1.9 and 260, respectively.

### **CONCLUSIONS**

In this paper, we have shown that the orientational fluctuation of a nematic liquid crystal can be observed directly with a high-speed TV camera. The results were analysed by a developed theory and it was found that the dispersion relation is still valid for fluctuations with long wavelength, which are difficult to be observed by the light scattering experiments, and the elastic constant divided by the viscosity is obtainable. The measurement time in this experiment is only 1 minutes, in which we can get two-dimensional data, though we require more time to analyse the data. Thus, we can expect that the direct observation and the analysis presented in this paper will be a powerful method to study the orientational fluctuations with long wavelength in liquid crystals.

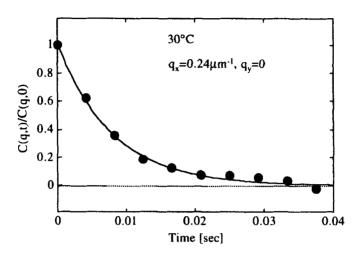


Figure 1 Typical spatial correlation function.

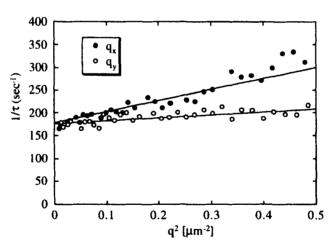


Figure 2 Dependencies of  $1/\tau$  on  $q^2$  along x- and y-axes.

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