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Intensity and Anisotropy of the Dynamic Light Scattering in Nematic Liquid Crystals

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The dependence on voltage of the correlation length of turbulence in the dynamic scattering mode in an ac excited nematic liquid crystal has been measured and discussed. The influence of a high frequency stabilizing field has been shown. The total anisotropy of the turbulent medium has been demonstrated, using measurements on a wedge-shaped sample.

I INTRODUCTION

A nematic liquid crystal in an external electric field gives rise to the so-called dynamic light scattering^{1,2} (cf. Refs. 3 and 4 also) with very important practical uses;⁴ however, no adequate attention has been devoted to physical processes occurring in the crystal structure.

Turbulent fluctuations found to occur in the dynamic scattering mode (DSM) are due mainly to the diffusion of orientation and not of vorticity as in isotropic liquids,⁵ the ratio of the two respective diffusion coefficients being $\sim 10^4$. Experimental dependence of the intensity of scattered light I_f on the wave vector \mathbf{q} can be adequately approximated by the function (cf. Refs. 3 and 5).

$$I_f(q) = I_i \exp(-|\mathbf{q}|l) \quad (1)$$

where I_i is the intensity of incident light and $|\mathbf{q}| = (4\pi/\lambda)\sin(\varphi/2)$ (φ is the scattering angle, λ the wavelength of incident light). Using dimensional analysis, l can be interpreted as a correlation length (at which turbulent fluctuations in the liquid crystal are still sufficiently connected with each other) and can be calculated from the angular dependence of the intensity

of scattered light. The existence of a coherence length is a general and characteristic property of the turbulent state. Fluctuations can be stabilized with an additional electric field whose frequency is so high that it cannot produce instabilities⁷ in the medium. We expect that in such a case it will be more difficult to produce turbulence and that the correlation lengths will be larger. Some results obtained from investigation of the voltage and frequency dependences of the intensity of dynamic light scattering (cf. also Ref. 5) and of the anisotropy in the disordered state will be discussed in Section III.

II EXPERIMENTAL

Sample

The liquid crystal used was *p*-*n*-methoxybenzylidene butylaniline (MBBA) which is nematic between 21–47°C. It has been sandwiched either in a classical thin cell with 50 μm teflon spacers or in a wedge-shaped cell, both with conducting inner surfaces (SnO_2); the crystal was oriented by the rubbing technique. The sample in the plane and in the wedge-shaped cell was placed in the light scattering instrument and polarizing microscope respectively, and excited by an amplified ac current from a sinusoidal generator (Tesla BM 365 U). If desired, its signal was mixed in a resistance mixer with the stabilizing high frequency signal (5 kHz) from a second generator.

Light scattering

Dutch Blecker instrument (with photomultiplier RCA 1P21, Hg lamp and 546 nm interference filter) was used for measuring the angular dependence of the intensity of scattered light. The correlation lengths have been then calculated with a computer following Eq. (1). Experimental arrangement HHH and HHV (in the notation adopted previously⁵) was chosen for this measurement: the polarizer gave horizontally polarized incident light, optical axis of the crystal in the unperturbed state was parallel to it, and the analyzer selected either the horizontal or the vertical component of the scattered light.

Birefringence

Integral birefringence was measured interferometrically using a wedge-shaped cell in a polarizing microscope. Its use is based on the observation⁷ that the instabilities near the threshold in the low-frequency region (the

Williams domains first and turbulence second) depend on the electrode voltage and not on the electric field intensity (and thus are independent of the sample thickness). In crossed polarizers there appear interference maxima and minima whose distance is related to the birefringence of the crystal, Δn , by the formula

$$\Delta n = (2k + 1)(\lambda/2)(b/xh) \quad (2)$$

where k is the number of extrema considered and x is the length covered by them; b is the length of the cell and h its height. If there remains some order in the turbulent state, the interference lines will not completely disappear, but only move (and certainly become less clear), characterizing thus the magnitude of birefringence in the turbulent state. Using the Fresnel equation for propagation of light in an anisotropic medium (cf. Ref. 8), we can calculate the average angle $\bar{\vartheta}$ between the optical axis and the direction of the incident light beam according to Eq. (3)

$$\cos^2 \bar{\vartheta} = (n_{\text{ef}}^{-2} - n_e^{-2}) / (n_o^{-2} - n_e^{-2}) \quad (3)$$

where n_o , n_e are the refractive indices of ordinary and extraordinary beams respectively, $n_{\text{ef}} = c/v$ being the effective refractive index for a beam propagating (at a velocity v) at an oblique angle with respect to the optical axis; c is the velocity of light in vacuum. The average angle ϑ of inclination of the molecules from their unperturbed position is given simply by $\vartheta = \pi/2 - \bar{\vartheta}$. In the turbulent state, ϑ of course is a time and space average.

III RESULTS AND DISCUSSION

Light scattering

Effects of the exciting voltage at constant frequency on the dependence of the scattered light has been examined for $U = 30\text{--}70$ V, $\omega = 70$ Hz and arrangement HHH (Figure 1a) and HHV (Figure 1b). We see that the higher the exciting voltage, the greater part of the incident light is scattered in the depolarized beam; at the highest voltages used, more than a half of the scattered intensity has opposite polarization.

Effect of the frequency and voltage on the corresponding correlation lengths l (calculated from Eq. (1)) has been studied in the HHH situation (Figure 2). We note that at higher frequencies the correlation lengths are larger for a given value of the exciting voltage. This can be explained by the mechanism of formation of instabilities described by Heilmeyer.¹ When ions of impurities oscillate with a higher frequency, crystal molecules are not able to move with an amplitude as large as at lower frequencies; as a result,

the orientations of the individual molecules are related to each other at a larger distance. With increasing exciting voltage U the correlation length diminishes for all frequencies (Figure 2a). The three curves seem to approach a certain limiting value l_0 , as predicted by the theory of turbulence:⁶ at constant temperature, the least dimension of the turbulent eddy, on whose level energy is dissipated by transformation into heat, is determined only by the viscosity of the medium. However, this could not be verified experimentally as the crystal broke down at high electric field intensities.

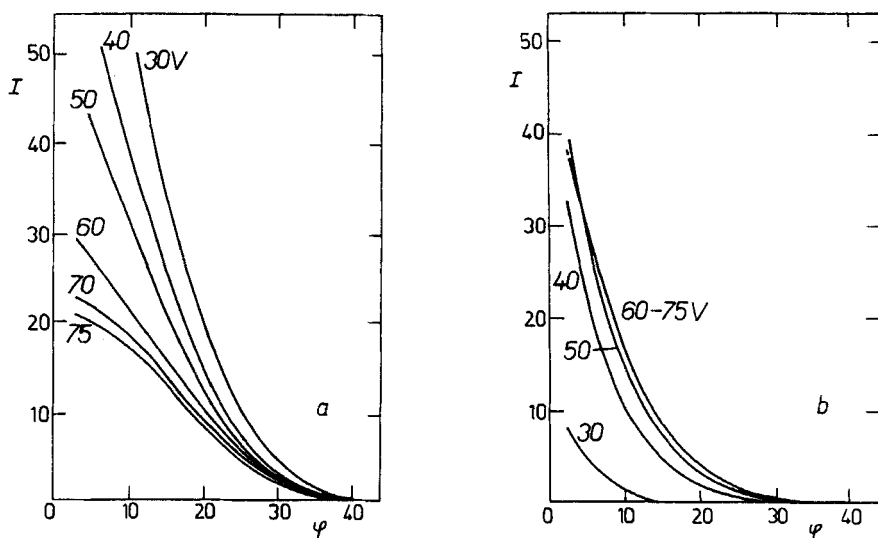


FIGURE 1 The angular dependence of the intensity of scattered light at $\omega = 70$ Hz in arrangement (a) HHH, (b) HHV (depolarized light). The parameter denotes the exciting voltage U .

Effect of the stabilizing electric field on the correlation lengths is shown in Figure 2b. As expected, the correlation length increases with increasing stabilizing voltage U_s (at $\omega_s = 5$ kHz, $U = 30$ V, $\omega = 70$ Hz). The last value corresponds to a moment when turbulent instabilities disappear changing into the Williams domains. The stabilizing electric field, E_s , increases the bulk energy by the term⁹ $F_s \sim E_s^2$, which is in fact equivalent to an increase in the elastic energy of the medium. This means that, under the same electrical conditions, the smaller elastic constants of the material the smaller turbulent eddies and, as a result, the larger the solid angle of scattered light. This is interesting in the case of a practical application of the dynamic light scattering, when we want the light to be scattered within a large solid angle.

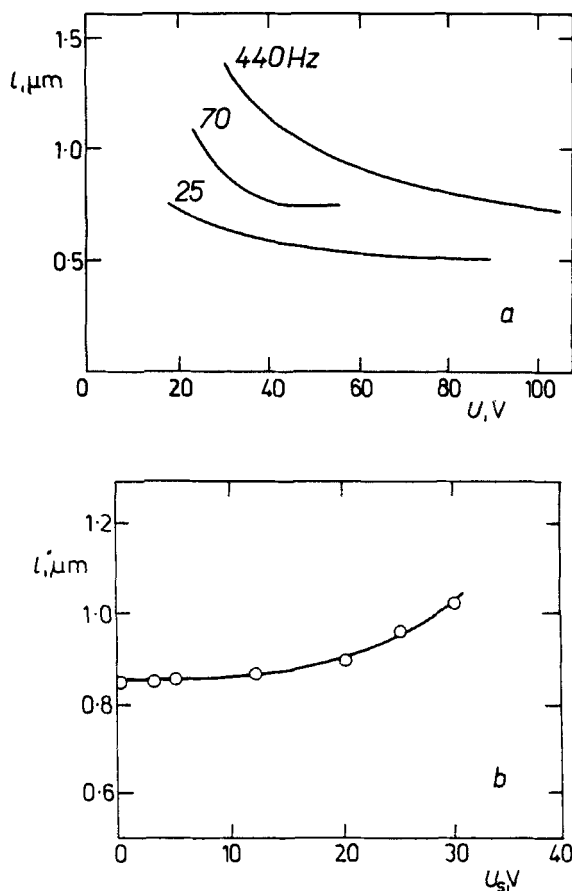


FIGURE 2 The dependence of correlation lengths l_c of turbulence (a) on the exciting voltage U (parameter: frequency ω); (b) on the stabilizing voltage U_s ($U = 30$ V, $\omega = 70$ Hz; $\omega_s = 5$ kHz).

Birefringence

The results of experiments with a wedge-shaped cell are shown in Figure 3; with increasing exciting voltage U (at $\omega = 70$ Hz), the birefringence of the medium decreases, but there remains a detectable Δn (at voltages $U \sim 42$ V the interference lines are too diffuse for a correct determination of Δn). We can conclude that even in the turbulent state the nematic crystal remains anisotropic, though less so than in the zero external field. The average angle β of the inclination of molecules out of their unperturbed positions can be extracted from these data by using Eq. (2) (see Figure 3). The inclination increases with increasing voltage starting with 22° at lower U (in accordance

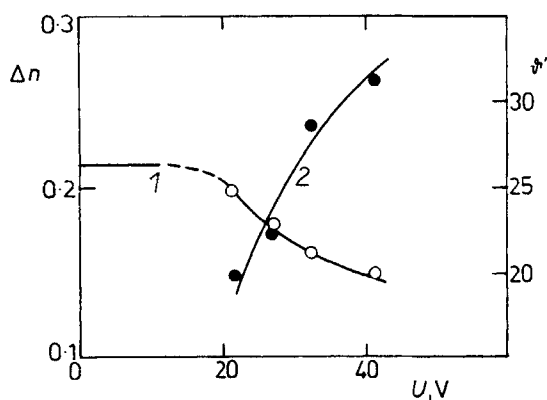


FIGURE 3 birefringence Δn of the nematic crystal (1) and average inclination β' of molecule in turbulent motion (2) plotted against the exciting voltage at $\omega = 70$ Hz.

with a value of 18° found by Penz for the average angle of inclination in the Williams domains) and moves to higher values expected for turbulence.

CONCLUSION

We have predicted and observed the dependence of the intensity of turbulence during the dynamic scattering mode (measured by the correlation length) on the exciting voltage, and explained the shape of the curves at various frequencies in one experimental arrangement (situation HHH). The stabilizing effects of a high-frequency field showed the role played by the elastic constants in the strength of turbulence. In measuring the birefringence interferometrically, we observed that the medium conserves some of its uniaxial character even in the turbulent state, i.e. when the structure of the nematic crystal is highly disordered.

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