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Speed-distribution analysis of the convective flow in electrohydrodynamic instabilities of nematic liquid crystals

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Convective velocity analysis of the electrohydrodynamic instabilities has been carried out by dynamic light scattering. Evidences of Doppler shift caused by the random motion of disclination lines in the convective flow are proposed. Based on that fact, speed distribution of the flow is analyzed. The validity of the analysis is confirmed measuring the real speed of moving disclination lines by serial photographs.

INTRODUCTION

When the voltage applied on the nematic liquid crystal is increased, we observe the successive transitions to fully developed turbulence from the laminar flows.[1,2] Many interests have been focused on this system from the point of view of "the bifurcation problem."[3,4] The analyses of static and dynamic characteristics have been performed using the optical technique. However, very little attention has been paid on the convective velocity of the fluid, one of the important parameters in these problems. In a previous paper, we reported the results of light scattering, where the power spectrum of scattered light could well describe behavior of forced director oscillation under high ac voltage.[5] However, the information about the convective velocity has not been satisfactory, because the laser light is parallel to the flow direction. In this study, we report a measurement of the velocity of the convective flow using the dynamic light scattering technique.

EXPERIMENT

A transverse cell was prepared as shown in Fig. 1; the nematic liquid-crystal MBBA [*p*-methoxybenzilidene *p*-(*n*-butyl)aniline] is enclosed in sandwiched glass cell with 0.5-mm-thick aluminum spacer electrodes, whose separation is 1.0 mm. The experimental setup is also shown in the figure. The incident laser light (He-Ne laser with about 0.8-mm cross-sectional diameter beam size) is perpendicular to the convective flow and the scattering vector **K** is almost parallel to the flow direction in a small scattering angle region. In this alignment, it is expected that convective flow brings large Doppler shift [see Eq. (2)]. The signal detected by the photodiode is analyzed by a fast Fourier transformation (FFT) spectral analyzer (Ono Sokki, CF 300). Averaged (1024 times) power spectra of the scattered light are obtained. The ac voltage of 25 Hz is applied on the electrodes. The critical voltage E_c of the laminar flow is 27 V_{rms}. The measurement is carried out at a temperature of 30 °C controlled within ±0.2 °C.

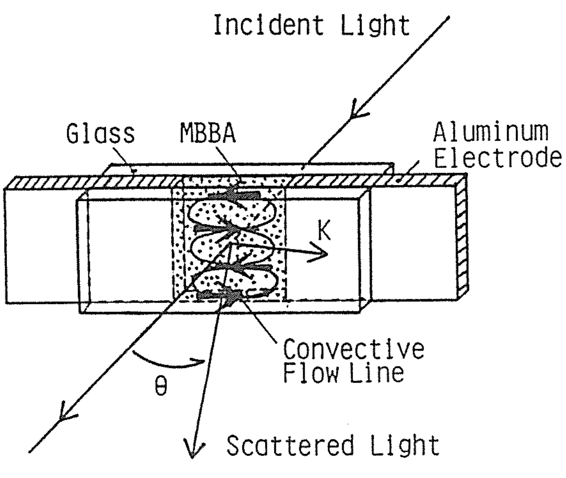

FIG. 1. Geometrical setup of the experiment. The incident light is perpendicular to the convextive flow. The scattering vector **K** is approximately parallel to the flow in a small scattering angle region.

EXPERIMENTAL RESULTS

The voltage dependence of the power spectrum is shown in Fig. 2. Here, the corresponding flow patterns photographed at each step are also shown. The zero voltage curves are superposed to clear the effect of the voltage in all figures. Above the threshold E_c below which no convective flow is observed, the difference between two power spectra increases with the voltage. Here, the lines known as "disclination lines," considered to be scatterers, appear. We can observe many scatterers in the scattering volume. In a higher voltage region above 70 V_{rms}, the flow patterns are complicated and random motion of disclination lines is clearly seen. We suppose that the movement of disclination lines corresponds to the convective motion.

At a lower-frequency region the spectra show exponential decay, the slope of which becomes more gentle with the applied voltage. It seems to be correlated to dynamic characteristics of the flow. We consider that the differences between two spectra S_{dif} are brought by the convective flow and define them as

$$S_{\mathrm{dif}} = S_E - S_0 \ , \qquad (1)$$

where S_E and S_0 are the power spectra under the voltage E and 0, respectively. In the following, using the difference spectrum S_{dif}, we carry out the convective velocity analysis by dynamic light scattering theory.

ANALYSIS BY DYNAMIC LIGHT SCATTERING

Theoretical background

Dynamic light scattering has been shown to be a powerful technique for analyzing a motility of microorganisms such

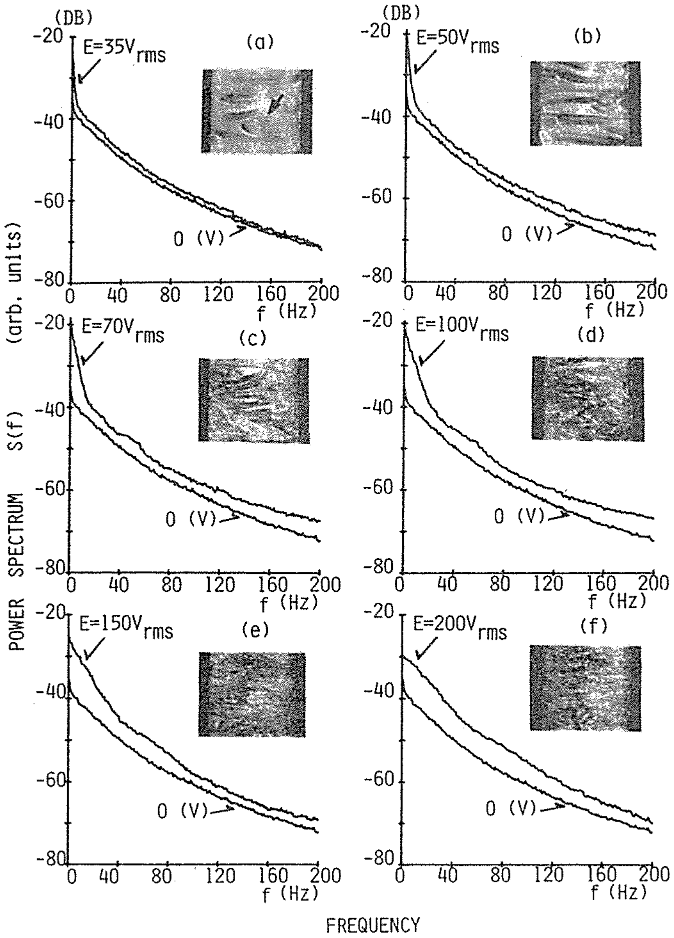

FIG. 2. Voltage dependence of the power spectrum. Zero voltage spectrum is superposed for comparison. The photograph is the convective flow patterns at each step. Arrow show "disclination lines" moving with the convective flow.

as swimming spermotozoa and motile bacteria.[6,7] Moving scatterers cause a Doppler frequency shift ω in the scattered light. If the scatterers move with velocity $\mathbf{v}$, the shift frequency ω is given by

$$\omega = \mathbf{K} \cdot \mathbf{v} \ , \tag{2}$$

where $\mathbf{K}$ is the scattering vector. The magnitude of $\mathbf{K}$ is given by

$$K = 4\pi n \sin(\theta/2)/\lambda \ , \tag{3}$$

where n is the refractive index of the scattering medium, θ the scattering angle, and λ the wavelength of the incident light. The distributed velocity of each scatterer causes a dispersion in the spectrum of scattered light. It has been shown that the correlation function $F(t)$ and the heterodyne photocurrent spectrum $S(\omega)$ are represented by[7]

$$F(t) = \int_0^\infty dv\, p(v) \sin(Kvt)/(Kvt) \ , \tag{4}$$

$$S(\omega) = BN_m \int_{\omega/K}^\infty p(v)\, dv/(Kv) \ , \tag{5}$$

where $p(v)$ is the speed distribution of scatterers in the scattering volume, B the proportional constant, and N_m the number of motile scatterers. Note that $F(t)$ obeys Kt scaling. When $F(t)$ or $S(\omega)$ is measured, we can estimate the speed distribution $p(v)$. Taking the inverse transform of Eqs. (4) and (5), $p(v)$ is obtained as

$$p(v) = (2v/\pi) \int_0^\infty d(Kt)\, KtF(Kt) \sin(Ktv) \ , \tag{6}$$

$$p(v) = -(K/BN_m)\omega(d/d\omega)S(\omega) \ . \tag{7}$$

It is well known that the measured power spectrum of motile organisms has the form[8]

$$S(\omega) = A \exp(-\omega/Kv_c) \ , \tag{8}$$

where A is a constant and v_c the characteristic velocity of the scatterers. From the Fourier transformation relationship, the normalized autocorrelation function $F(t)$ will be of Lorenzian form

$$F(t) = 1/[1 + (t/t_c)^2] \ . \tag{9}$$

The half-height time t_c is related to v_c as

$$t_c = 1/(Kv_c) = \lambda/[4\pi n \sin(\theta/2) v_c] \ . \tag{10}$$

The speed distribution $p(v)$ calculated using Eq. (7) is normalized as

$$p(v) = (v/v_c^2)\exp(-v/v_c) \ . \tag{11}$$

This is called the "Saclay distribution."[7]

Evidence of Doppler shift

As noted above, we suppose that the difference spectrum S_{dif} is caused by the convective flow. The evidences of Doppler shift caused by the motion of disclination lines are shown in the following.

The autocorrelation function $F(t)$, which expresses the dynamic nature of scatterers, is calculated from S_{dif} in Fig. 2. Its temporal change is shown in Fig. 3 at various voltages; they have Lorenzian form. The behavior has some analogy to Eq. (9). This fact confirms that the moving disclination lines have the same role as motile organisms in the motion of scatterers. Namely, it is considered that S_{dif} is caused by the Doppler shift due to the random motion of scatterers—the disclination lines. In the figure, the half-height time t_c decreases with the voltage. According to Eq. (10) the decrement of t_c is correlated to the increment of the speed of moving scatterers.

We have other evidence. The angular dependence of the

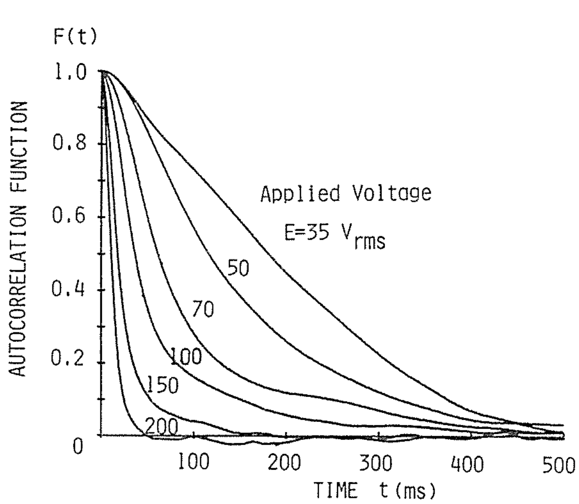

FIG. 3. Normalized autocorrelation function calculated from S_{dif}.

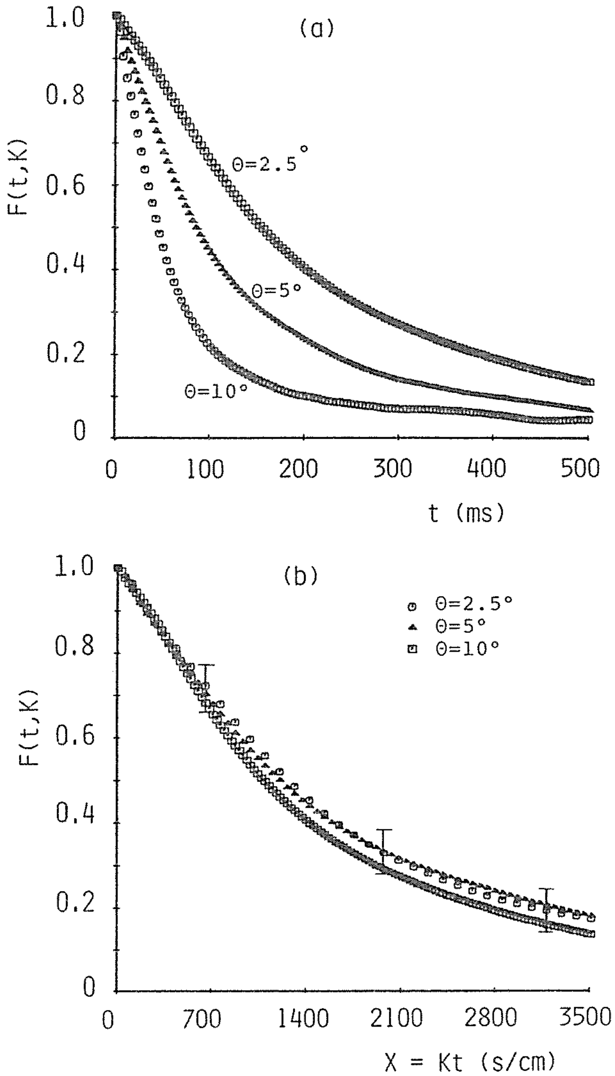

FIG. 4. Autocorrelation function as the functions of (a) the scattering angle and (b) $X=Kt$ in the turbulent state ($E=70\,V_{rms}$).

autocorrelation function was examined at the scattering angles of 2.5, 5, and 10 deg in the turbulent state ($70\,V_{rms}$) shown in Fig. 2(c). The results are shown in Fig. 4(a). They also have Lorenzian form with different t_c. It is noted that t_c has nearly an inverse proportion to the scattering angle. This behavior is expected from Eqs. (9) and (10) in the small scattering angle region. Furthermore, autocorrelations are replotted as a function of $X=Kt$ in Fig. 4(b). Their dependences are the same within experimental error. Namely, $F(t)$ obeys Kt scaling. This behavior is consistent with Eq. (4) and is well known to be evidence of the existence of the Doppler shift by moving scatterers with random velocity and constant speed in the scattering volume.[7-9]

Speed-distribution analysis

Here, we try to analyze the velocity information of the convective flow based on the fact that S_{dif} is caused by the Doppler shift due to the moving disclination lines. Therefore, tentatively using S_{dif} for $S(\omega)$, we carried out the analysis of the speed distribution of the scatterers in the scattering volume. Figure 5 shows the change of the speed

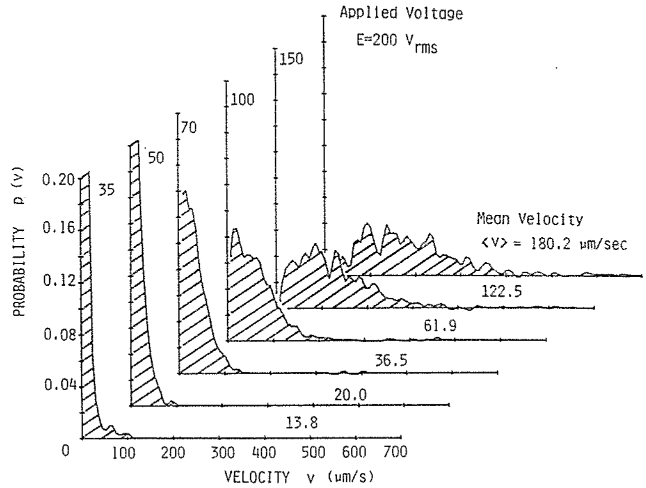

FIG. 5. Change of speed distribution $p(v)$. The profile changes from Gaussian-like to Saclay types with increase of the voltage.

distribution $p(v)$ with the voltage calculated from Eq. (7); the profile changes from Gaussian-like to Saclay type with increasing voltage. So far as we know, this is the first demonstration of the speed distribution in the electrohydrodynamic (EHD) instability of nematic liquid crystals.

The mean velocity $\langle v\rangle$ is also calculated by

$$\langle v\rangle=\int_0^\infty vp(v)\,dv \quad . \tag{12}$$

The voltage dependence of $\langle v\rangle$ is shown in Fig. 6. Horizontal axis is plotted as a normalized voltage $k(=E/E_c)$. As seen in the figure, two regions are distinguished. In the lower voltage region, $\langle v\rangle$ increases linearly above E_c. A kink point is seen at about $k=4.3$. This behavior is consistent with the experimental result obtained by Kai, Yoshitsune, and Hirakawa.[10] They have reported the measurement of mean flow velocity through a motion of small bubbles in the nematic liquid crystal. Absolute values of

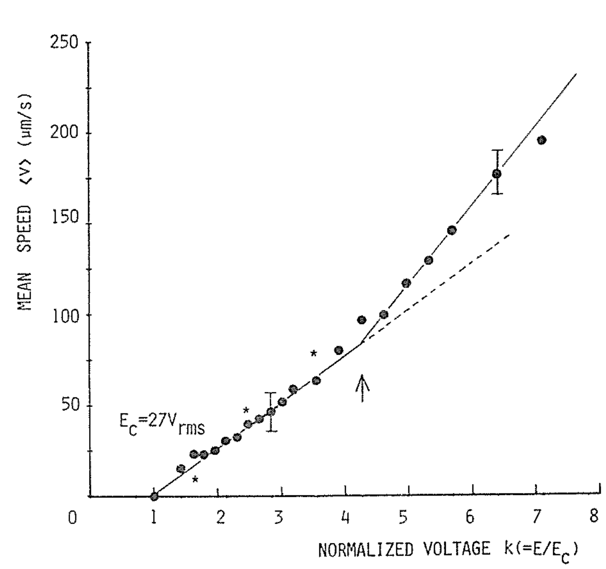

FIG. 6. Voltage dependence of the mean velocity $\langle v\rangle$. Arrow indicates the kink point ($k=4.3$) of the velocity. Asterisks show the speed of the disclination lines measured on the serial photographs.

the mean velocity and the normalized voltage of the kink point are slightly different from our results. This discrepancy may be due to the differences in measuring methods and the aspect ratios—a typical horizontal dimension of the layer of convective fluid divided by its thickness. The aspect ratio of our cell is about 20. Kai *et al.*[10] have also suggested the occurrence of the phase-transition-like phenomenon at the kink point. In a recent experiment Yamazaki, Kai, and Hirakawa[4] clarified the existence of the inverted bifurcation (the first-order-like transition) at $k=5.14$. The aspect ratio of their cell is about 170. Considering the difference between the ratios, the transition point would correspond to the kink point in our experiment. Anyway, consistent behavior of the mean velocity confirms the validity of our analysis.

The real speed of the disclination lines is measured on the serial photographs taken every 0.7 sec. The results are shown by asterisks in the Fig. 6. They agree with those obtained from Eq. (12) within experimental error. This fact also confirms the validity of the speed-distribution analysis by dynamic light scattering.

CONCLUDING REMARKS

In this Rapid Communication we have reported the dynamic light scattering study on the EHD instability of nematic liquid crystals. The important findings are as follows.

(1) The power spectrum of scattered light above E_c is different from that at zero voltage.

(2) It is shown that the difference spectrum S_{dif} is caused by the Doppler shift due to the moving disclination lines with the convective flow.

(3) Based on the above facts, the speed-distribution analysis is carried out by dynamic light scattering. The distribution changes from Gaussian-like to Saclay type.

(4) The validity of the analysis is confirmed measuring the typical speed of disclination lines by serial photography.

In the sense of the first-order approximation, the analysis practiced here would be valid. However, the motion of disclination lines is not perfectly random and independent. The motion is fairly oriented and correlated. The orientational motion becomes more remarkable at the higher voltage region. As a result, the Kt scaling is limited within smaller scattering angles. For closer and more detailed analysis, the theoretical treatment should be improved.

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