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equation, $\hbar\omega_0 = 41A^{-1/3}$, the distributions of the extra protons for the isotone pairs ^{90}Zr–^{89}Y and ^{92}Mo–^{90}Zr have been computed and are compared with the experimental predictions in figures 4 and 5 respectively.

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Light scattering by electrohydrodynamic fluctuations in nematic liquid crystals†

Abstract. We present some measurements on light scattered by a liquid crystal under a dc applied electric field. The results are interpreted in terms of radiation diffused by single scattering centres, which suffer electrohydrodynamic velocity fluctuations.

Hydrodynamic instabilities in nematic liquid crystals subjected to a dc electric field have been recently investigated (Heilmeier *et al.* 1968). This kind of effect can be directly observed by a microscope only when the applied electric field is low enough to ensure the presence of domain patterns (Durand *et al.* 1970). A slightly larger range of electric field intensities (some 10^3 V cm^{-1}) has been explored by measuring the 'rise time' of light scattering associated with induced instabilities

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(Koelmans and Van Boxtel 1970, Koelmans and Van Boxtel 1971). The experimental results of Durand *et al.* (1970) and Koelmans and Van Boxtel (1970, 1971) put into evidence the optical turbidity associated with turbulent mass motion in the sample. Such material flow has been predicted either by means of an anisotropic conductance model (Carr 1969, Helfrich 1969) or in terms of hydrodynamic instabilities produced by space charges (Koelmans and Van Boxtel 1970, 1971).

The aim of this paper is to study the turbulent motion in the nematic phase under a dc applied electric field by measuring the spectrum of scattered light. In this frame, we remember that the presence of scattering centres in the nematic phase has been well established by measurements of the angular radiation distribution (Deutsch and Keating 1969). Therefore, our starting point can be based on the theoretical analysis of the frequency spectrum of the radiation diffused by particles suspended in a turbulent fluid (Bertolotti *et al.* 1969, Di Porto *et al.* 1969). We remember that the electric field scattered by N identical optical dishomogeneities in the single scattering approximation reads

$$\mathscr{E}(\boldsymbol{R}, t) = \boldsymbol{\xi}(\boldsymbol{k}) \exp(\mathrm{i}\omega_0 t) \sum_{j=1}^{N} \exp\{-\mathrm{i}\boldsymbol{k} \cdot \boldsymbol{r}_j(t)\} \tag{1}$$

where $\boldsymbol{\xi}(\boldsymbol{k})$ is the scattering vector associated with the single centre whose trajectory is represented by $\boldsymbol{r}_j(t)$, ω_0 is the frequency of the incident plane wave of wavenumber $\boldsymbol{k}_0$ and $\boldsymbol{k} = \boldsymbol{k}_0 - k_0 \boldsymbol{R}/R$. While equation (1) is valid for rigid particles, we can take into account temporal size modifications of the diffusing centres by modifying equation (1) into

$$\mathscr{E}(\boldsymbol{R}, t) = \boldsymbol{\xi}(\boldsymbol{k}) \exp(\mathrm{i}\omega_0 t) \sum_{j=1}^{N} \exp\{-\mathrm{i}\boldsymbol{k} \cdot \boldsymbol{r}_j(t)\} f_j(t) \tag{2}$$

with

$$f_j(t) = 1 + f_j'(t) \tag{3}$$

where $f_j'(t)$ is a stochastical centred function depending upon the temporal size variations of the jth centre. (For the sake of simplicity, we omit all dependence of $f_j(t)$ upon $\boldsymbol{k}$ and E. Furthermore, we observe that, strictly speaking, equation (2) applies to a case in which spherical centres undergo a temporal modification in their radius. If one is interested in more general variations, it is necessary to introduce a stochastical tensor $\overleftrightarrow{f}_j(t)$.) The spectrum of the scattered light

$$I(\boldsymbol{k}, \omega) = \int_{-\infty}^{+\infty} \exp(-\mathrm{i}\omega t) \langle \mathscr{E}(R, t) \mathscr{E}^*(R, 0) \rangle \, \mathrm{d}t \tag{4}$$

can be now easily evaluated under the joint-Gaussian distribution hypothesis for the velocity field. In effect, if we put

$$\langle f_j'(t) f_j'(0) \rangle = \langle f'^2(0) \rangle \exp(-Ft^2) \tag{5}$$

use of equations (2) to (5) (see also the paper of Di Porto *et al.* 1969) yields

$$\begin{aligned} A^2(\boldsymbol{k}, \omega) &= I(\boldsymbol{k}, \omega) \\ &= C(\boldsymbol{k}, E) \left\{ \exp\left(- \frac{(\omega_0 - \omega)^2}{2k^2 \overline{U_1^2}(E)} \right) \right. \\ &\quad \left. + \langle f'^2(0) \rangle \exp\left(- \frac{(\omega_0 - \omega)^2}{2\{k^2 \overline{U_1^2}(E) + 2F\}} \right) \right\} \end{aligned} \tag{6}$$

where the value of $C(\boldsymbol{k}, E)$ is inessential for our further considerations, $\overline{U_1^2}(E)$ represents the mean square value of velocity fluctuations along the direction of $\boldsymbol{k}$, and $A(\boldsymbol{k}, \omega)$ labels the amplitude spectrum of the diffused radiation. It is worthwhile remembering that equation (6) holds whenever the characteristic correlation time of turbulence t^* fulfills the relation

$$t^* \gg (k^2 \overline{U_1^2})^{-1/2} \tag{7}$$

which can be put in the simpler form

$$l^* \gg \frac{1}{k} = \frac{1}{2k_0 \sin(\theta/2)} \tag{8}$$

where θ is the scattering angle and l^* represents a characteristic length of the turbulent field (Bertolotti *et al.* 1969).

In order to test the validity of equation (6) we have performed a scattering experiment by measuring the amplitude spectrum of light diffused by a thin film of anisylidene-para-amino-phenylacetate (APAPA) in the nematic mesophase. The radiation obtained from a Spectra Physics 120 He–Ne laser was made to impinge normally on two samples of APAPA with thicknesses of 6 μm and 25 μm. The samples were placed between SnO_2 coated glass plates, which were used as electrodes in order to apply a dc electric field. While the electric field intensities ranged up to 5×10^4 V cm^{-1}, the measurements were performed at various values of temperature in the nematic range (82 °C–110 °C) with thermal stability of 0·1 °C, and the angular region 0 °–22 ° was explored. The spectrum of the scattered light was measured by optical heterodyning with a frequency shifted reference laser beam. The beat current from the photomultiplier was sent to a high resolution spectrum analyser and many sweeps were averaged.

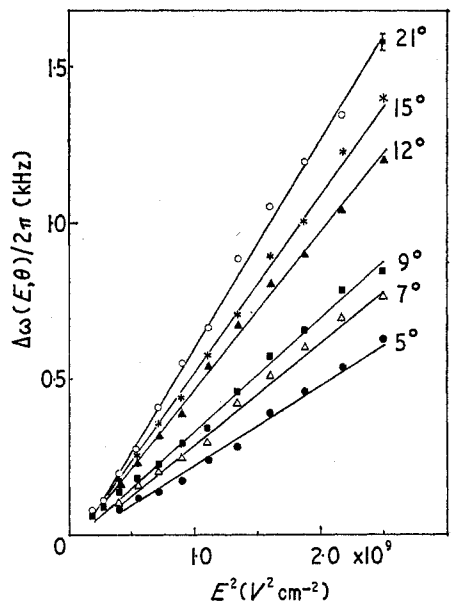

Figure 1. Half-height width of scattered amplitude distribution as a function of E for various values of θ. Sample thickness, 6 μm; T = 93·5 °C.

We report in figure 1 the half-height width $\Delta\omega(E, \theta)$ of the amplitude distribution as a function of E for various values of θ. It is worthwhile to observe that the Gaussian behaviour of $A(k, \omega)$ predicted by equation (6) is verified for all field amplitudes greater than the threshold of dc instabilities and for θ greater than approximately 2°. The Gaussian behaviour cannot be verified for θ less than about 2°; this is due to the finite instrumental resolving power and to the presence of weak contributions of multiple scattering at high applied electric fields. In all cases, the linewidth of $A(\boldsymbol{k}, \omega)$ for $\theta \simeq 0$ is small enough, allowing one to put $\langle f'^2(0)\rangle = 0$ in equation (6).

The main conclusions which can be drawn from the previous considerations and from an accurate inspection of figure 1 can be summarized in the following way: (i) the typical correlation length l^* of the turbulent velocity field fulfills in our experimental conditions the relation $l^* > 3\mu$, as one can see by putting $k_0 \simeq 10^5\ \mathrm{cm}^{-1}$, $\theta \simeq 2°$ in equation (8). This result can be compared with the typical values of the 'coherence distance' for a liquid crystal film of greater thickness obtained by Deutsch and Keating (1969); (ii) the temporal size modifications of the scattering centres are slow enough to allow us to neglect their contribution to the line broadening for θ greater than about 2°; (iii) the linearity of $\Delta\omega(E, \theta)$ as a function of k implies the independence of $\overline{U_1^2}$ from the direction of $\boldsymbol{k}$. This in turn ensures the isotropy of the turbulent velocity field in our experimental situation, which is mainly due to the boundary conditions imposed on the sample. In fact, we have observed remarkable anisotropy effects with different geometrical arrangements; (iv) the linear behaviour of $\Delta\omega(E, \theta)$ against E^2, as directly reported in figure 1, implies the relation $\{\overline{U_1^2}(E)\}^{1/2} = BE^2$, where B is a constant depending on the dynamical properties of our crystal at the considered temperature. This extends in a sense to the turbulent regime, the quadratic dependence already found in the domain regime (Durand *et al.* 1970).

The measurement of $\Delta\omega_T$ at various temperatures for fixed values of E and θ is shown in figure 2. If we assume for the viscosity coefficient η a behaviour of the type

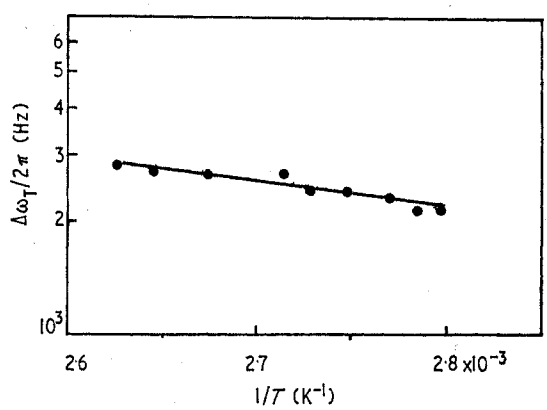

Figure 2. Half-height width of scattered amplitude distribution as a function of T, for $E = 2{\cdot}4 \times 10^4\ \mathrm{V\ cm^{-1}}$ and $\theta = 22°$. The sample thickness is 25 μm.

$\partial(\ln\eta)/\partial(1/T) =$ constant (Haller and Litster 1970), we find a rough agreement with the relation

$$\Delta\omega_T \propto \frac{E^2}{\eta(T)} \tag{9}$$

which can be compared with the relation $t_r \propto \eta(T)/E^2$ for the rise time of light scattering (see Koelmans and Van Boxtel 1970, 1971).

On the whole, we may conclude that our measurements provide a reasonable test of the behaviour of strongly scattering optical dishomogeneities associated with turbulence induced by a continous electric field in a nematic liquid crystal.

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