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LIQUID CRYSTALS.

LIGHT SCATTERING BY FLUCTUATIONS INDUCED BY AN APPLIED ELECTRIC FIELD IN A NEMATIC LIQUID CRYSTAL (APAPA)

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1. Introduction. — Nematic crystals under an applied electric field scatter light. This scattering can be explained as due to two contributions.

The first arises from the circumstance that in a nematic liquid crystal a situation can exist in which the molecules tend to be aligned parallel to each other, however thermodynamic fluctuations exist in the alignment. They give rise to scattering of light. This scattering is always present also in absence of an external field.

Another contribution to scattering is given by the scattering due to velocity instabilities induced by the electric field. The application of a low electric field in fact gives rise to regular orientation patterns as observed by many authors [1]. By increasing the field, at strong fields, this regularity disappears giving rise to a turbulence as observed by Heilmeyer [2].

As regards to the first contribution the small motions of an incompressible nematic liquid crystal with coupled flows and molecular rotations have been analyzed in term of the Leslie eq. [3]. The presence of a magnetic field was also accounted for.

One of the major results of ref. [3] is that the power spectrum of the scattered light is entirely controlled by the slow mode (eq. (IV.23)). This mode corresponds to a slow relaxation of a twisted nematic structure. The linewidth of the resulting frequency spectrum of scattered light is very large of the order for example of $10^8$ Hz at an angle of 1 rad.

When account is taken for the presence of an electric field in the free energy expression for the nematic crystal, the same orders of magnitude are obtained as in the magnetic case. Moreover the intensity of the molecular scattered light is very low.

The other contribution that we will consider here is the scattering by velocity fluctuations.

The presence of an hydrodynamic current flow in nematic crystals under electric fields has been observed by Koelmans [4] and Durand [5].

Molecules or molecular aggregates in turbulent motion scatter light due to index of refraction fluctuations connected to their anisotropy and their motion.

A typical « coherence distance », i.e. the distance over which there is appreciable correlation of the turbulence in the liquid crystal, was measured for APAPA by Deutsch and Keating [6] of the order of the micron, and depending on the applied electric field.

The scattered light by the turbulent motion of these molecular aggregates will suffer a frequency shift proportional to the fluctuating velocity component in the transferred momentum direction.

The scattered light intensity must in this case be very large as compared to molecular scattering due to the large difference between the two extreme
values of the index of refraction of the anisotropic molecules.

Accordingly it must be expected that in an experiment of light scattering in the presence of strong electric fields, when the turbulent motion is established, the observed light intensity is mainly due to these hydrodynamical velocity instabilities. We have applied these considerations to the calculation of the spectrum of scattered light [7] and have found that in the case in which the centers of scattering do not change in dimensions, the spectral intensity is given by

\[ I(k, \omega) = C(k, E) \exp \left\{ - \frac{(\omega - \omega_0)^2}{2k^2 \bar{\nu}^2(E)} \right\} \]

where the value of \( C(k, E) \) is inessential for our considerations, \( \omega_0 \) is the frequency of the incident light, and \( \bar{\nu}^2(E) \) represents the mean square value of velocity fluctuations along the direction of \( k \).

The case in which the centers of scattering suffer changes in dimensions was also examined in ref. [7], where it was shown that the contribution to the spectrum of this effect can be neglected, in our experimental conditions.

2. Experimental set-up. — The spectrum of the scattered light was measured by means of an heterodyne technique. A block diagram of the method is shown in figure 1. An ultrasonic cell of frequency \( v_m = 21 \text{ MHz} \) diffracts a portion of the light from a He-Ne laser \( (\lambda = 6.332 \text{ Å, power 5 mW}) \) which results modulated at the frequency \( v_m \). This beam represents the local oscillation with which the scattered light is made to beat. The beating signal, as revealed by means of a photomultiplier, is centered at the frequency \( v_m \), and is analyzed, after filtering of the d. c. component, by a spectrum analyzer \( (0-110 \text{ MHz}) \). It is subsequently stored in the memory of a 1 024 Laben correlatron.

To avoid possible zero drifts of the spectrum analyzer, a square wave is added to the signal, as a reference signal at frequency \( \Delta v \). One of the two frequency peaks at frequency \( v_m \pm \Delta v \) is then used as a trigger for the storage sweeps of the correlatron.

Undesired signals at frequency \( v_m \) due to a possible amplitude modulation of the laser light in the ultrasonic cell are avoided by making the correlatron to work in sum and subtraction in alternate sweeps. To the signal spectrum stored during one sweep we subtract in the next sweep the eventual spurious signal at \( v_m \) by stopping the incident beam in the liquid crystal sample by means of a mechanical chopper. The whole system was completely automatized and gave the results over a writer. The data have been subsequently analyzed by a computer.

We used two cell geometries for the liquid crystal. In one disposition the sample was put between two optical glasses on one of which two chromium electrodes were evaporated at a distance from each other of 1 mm. The resulting d. c. electric field \( E \) was therefore normal to the incident light wavevector \( k_0 \). The cell thickness, by using mylar spacers, was 25 \( \mu \).

In the other disposition the two glasses were covered with a tin-oxide so to realize two transparent electrodes. In this case the d. c. electric field was parallel to \( k_0 \). The used thicknesses of the cells were 6 and 25 \( \mu \).

Temperature to take APAPA in the mesophase range \( (82 \text{ to } 110 \text{ °C}) \) was provided by an external heater, temperature controlled with a stability of \( 0.1 \text{ °C} \).

The used material was APAPA purified by means of successive crystallizations down to resistivity values of the order of \( 10^9 \) to \( 10^11 \text{ Ω} \text{ cm} \).

3. Experimental results. — The frequency spectra of the amplitude of the scattered light obtained for values of the applied electrical field larger than the threshold value for the onset of turbulence, resulted to be always gaussian, within the experimental errors. The scattered light intensity is so high that a small number of sweeps in sum and subtraction is sufficient to obtain good quality spectra. In figure 2 a
typical spectrum is shown referring to the case $\theta = 21^\circ$, $E = 3.3 \times 10^4$ V/cm, $T = 93.7^\circ$C. The gaussian curve that best-fits the experimental points is shown as a dotted line.

We have measured the half-height linewidth as a function of the scattering angle $\theta$, applied d. c. electrical field $E$, and temperature $T$.

We will first consider the results obtained in the geometry in which $E \parallel k_o$.

The scattered light has spatial distribution with a cylindrical symmetry around the $E$ direction. This gives indication that this direction is a preferential one for the electrodynamical turbulence process which results isotropic in the cell plane (orthogonal to $E$).

The isotropy of this process is clearly imposed by the boundary conditions produced by the presence of walls.

The behaviour of $\Delta\nu$ as a function of $E^2$, for various values of $\theta$, in the case $E \parallel k_o$ is shown in figure 3 for a cell 6 $\mu$ thick.

The dependence of $\Delta\nu$ on $k$ is linear with good approximation, starting from $k$ values very near to zero, when the applied electric field $E$ is not very large. At large $E$ values, $\Delta\nu$ does not tend to zero as $k \to 0$. The presence of a finite value of $\Delta\nu$ for $k \to 0$ is more evident for samples 25 $\mu$ thick (s. Fig. 5 taken for one chosen value of $E$). In this case the behavior of $\Delta\nu$ vs $k$ is again linear but shifted towards higher values of frequencies.

A linear relation is evident up to the highest $E$ value considered.

It is of interest to see if for all the considered values of $E$ in figure 3 we are in single scattering conditions. To this purpose some information can be gained by considering the behavior of $\Delta\nu$ as a function of the transferred momentum $k$, as shown in figure 4 for a 6 $\mu$ thick cell.
The linear behavior of $\Delta v$ in $k$ is in agreement with our Doppler scattering model and shows that the scattering is due to velocity fluctuations. The presence of a finite value of $\Delta v$ for $k \rightarrow 0$, for large $E$ values can be understood by assuming that in this case the scattering is no more single.

A measurement of the transmitted light intensity over the incident one as a function of $E$ is shown in figure 6 on a semi-logarithmic plot. It is seen that starting from the threshold value of $E$ for the onset of turbulence up to values of $E$ of the order of $2.6 \times 10^4$ V/cm, the kind of absorption is the same, and we can assume to be in the single scattering region. In figure 4 the $3 \times 10^4$ V/cm curve approaches nearly zero, while the $4 \times 10^4$ V/cm curve gives $\Delta v \approx 270$ Hz.

Multiple scattering seems not to affect the quadratic behavior of $\Delta v$ with $E$. This is also found by using 25 $\mu$ thick cells as is shown in figure 7.

The behavior of $\Delta v$ as a function of temperature $T$ for fixed values of $\theta$ and $E$ is shown in figure 8 for a sample 25 $\mu$ thick.

Figure 8 shows $\ln \Delta v$, measured for increasing temperature (triangles) and decreasing temperature (black dots), as a function $1/T$. The resulting points fit well a rectilinear behavior. We wish to put out that this behavior is similar to the one of viscosity in crystal liquids [8] and suggests a linear dependence of $\Delta v$ on the inverse of viscosity.

We will now consider the case $E \perp k_0$. In this case the interelectrode space in which the electric field is present is much larger than in the preceding geometry (1.2 mm). Moreover we have a strong anisotropy in the space distribution of the scattered light, as already reported [9]. In fact nearly all the scattering is in a plane orthogonal to $E$.

This means that velocity fluctuations are relevant only in a plane orthogonal to the $E$ direction.

By simple inspection it can be seen that there is a matter transport and the liquid crystal moves in the whole cell also out of the electric field.

For the light spectrum study in this geometry the volume of scattering must be chosen far enough from
the electrode so to avoid dishomogeneity problems of the electric field.

The measured behavior of $\Delta v$ vs $E$ at a fixed value of $\theta$ for different $T$ values is shown in figure 9. In this case the behavior of $\Delta v$ is quadratic in $E$ for low temperatures in the mesophase, while a saturation is clearly shown at high temperature.

This can be understood by observing that in this case the fluctuations that produce scattering are in the normal direction to the mean velocity. A decrease in viscosity by increasing temperature will therefore produce a decrease of the friction force responsible for fluctuations.

4. Conclusions. — We have measured the spectrum and linewidth of light scattered by a nematic liquid crystal (APAPA) under an applied d. c. electric field by means of an heterodyning technique. It results that the spectrum of scattered light is within the experimental errors well fitted by a gaussian curve, and that the linewidth increases with $E^2$ in the geometry in which $E \parallel k_0$.

When $E \perp k_0$ a saturation value of linewidth for increasing $E$ values is found, by increasing temperature in the mesophase range.

The results are in agreement with a simple model of scattering by velocity fluctuations of scattering centers in the liquid crystal due to turbulence produced by the applied electric field.

References