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SHEAR WAVE ATTENUATION IN SMECTIC-A LIQUID CRYSTALS (*)

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Résumé. — Nous avons étudié le comportement de deux smectiques-A soumis à une contrainte de cisaillement ultrasonore dans le cas où le vecteur d'onde est parallèle à la normale aux couches. Nous avons mis en évidence un processus de relaxation que nous discutons en supposant l'existence d'un couplage entre les fluctuations du directeur et l'écoulement local.

Abstract. — We have studied the shear impedance of two smectics-A when the propagation of the shear wave is along the normal to the layers. We observed that the real part and the imaginary part of the shear impedance differ significantly, indicating a relaxation process. We analysed our results on the basis of a coupling between the director fluctuations and the shear flow.

Impedance methods are traditionally used to study the shear properties of liquids. An indication of a relaxation process is given when the imaginary part X is not equal to the real part R of the shear mechanical impedance, a quantity which is defined as the negative ratio of shear stress to the rate of change of shear displacement.

Using the so-called impedometer method [1] we have recently studied at ultrasonic frequencies the shear impedance of a liquid crystal which has only nematic [2] and isotropic [3] liquid phases. In the isotropic phase we observed the relaxation process predicted by de Gennes [4]. In the nematic phase we found $R \simeq X$ giving no indication of relaxation. This last result enabled us to deduce three effective viscosity coefficients of the nematic.

In this paper we are dealing with smectic-A phases and we present evidence of a relaxation effect when the propagation of the shear wave is along the normal to the layers [5].

The determination of the shear impedance is obtained from measurements of the reflection coefficient of a shear wave at the solid-liquid interface between a fused-quartz bar and a smectic liquid crystal. Oblique and normal incidence can be used, the

result being basically identical for both cases [6-7]. We used oblique incidence because the sensitivity of the method is increased by reflecting the shear wave from the interface at large angles of incidence [8]. Our experimental set-up has been described in detail previously [2]. The homeotropic alignment was obtained by coating the reflecting surface of the bar and the cover glass with a layer of lecithin. The layer was so thin that it did not detectably affect the received pulses. The orientation was examined in polarized light between crossed polarizers. A schematic diagram of the experiment is shown in figure 1.

Diheptylazoxybenzene has two mesomorphic phases, smectic A from 34° to 54 °C and nematic from 54° to 71 °C. It is known to have a nearly-second-order nematic to smectic A phase transition. Our measurements of R and X at 15 MHz are reported in figure 2. In the smectic-A phase we observe that R and X are quite different indicating a relaxation process. In the

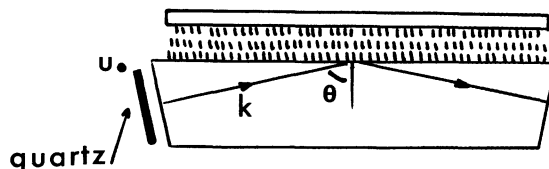


FIG. 1. — Schematic diagram of the acoustic system : the vibration u is parallel to the reflecting surface. k is the wave vector.

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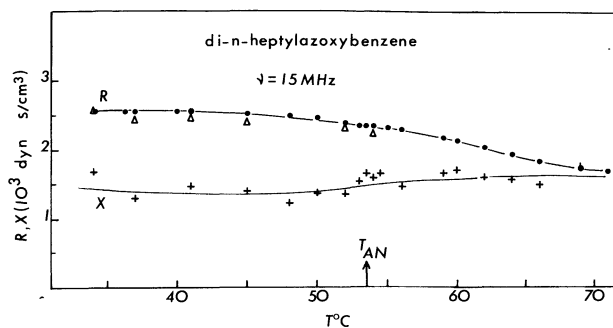


FIG. 2. — The real R and imaginary X parts of the shear impedance in the nematic and smectic-A phases of di-n-heptylazoxybenzene. The symbols \bullet and Δ are for two different samples.

nematic phase there is also evidence of a relaxation process which becomes more and more apparent as the phase transition is approached. Similar behaviour was observed at 25 MHz.

Since the transition is of second order we expect to observe critical viscosities which show a relaxation behaviour at finite frequencies [9]. In fact it has been predicted that the viscosity η_3 (in the MPP notation [10]) which is measured here is not critical. This was verified by capillary [11] and light scattering measurements [12].

a) *The smectic phase.* — The simplest model to explain these results is to assume that the director is disturbed from its equilibrium value by the shear stress (of angular frequency ω) and relaxes with a single characteristic time τ [13]. A simple form for the relaxation behaviour of the viscosity η_3 is given by :

$$\eta_3(\omega) = \eta_3^0 - \delta\eta_3 \frac{i\omega\tau}{1 + i\omega\tau} \quad (1)$$

η_3 goes from η_3^0 (for $\omega\tau \ll 1$) to $\eta_3^0 - \delta\eta_3$ (for $\omega\tau \gg 1$).

The relaxation time τ is given by

$$\tau^{-1} = \frac{B_{\perp}}{\gamma} \quad (2)$$

where B_{\perp} is the force maintaining the molecules normal to the layers and γ is a viscosity coefficient ; τ^{-1} is expected to vanish when the A-N transition is second order [14].

The real and imaginary parts of the viscosity η_3 are given by

$$\eta_3'(\omega) = \eta_3^0 - \delta\eta_3 \frac{\omega^2 \tau^2}{1 + \omega^2 \tau^2} \quad (3)$$

$$\eta_3''(\omega) = \delta\eta_3 \frac{\omega\tau}{1 + \omega^2 \tau^2}. \quad (4)$$

Since the viscosity is a complex number the liquid crystal presents an apparent rigidity coefficient G' given by :

$$G'(\omega) = \omega\eta_3''(\omega). \quad (5)$$

From our measurements we computed the shear modulus G' and the real part η_3' of the viscosity using the formulae :

$$\eta_3' = \frac{2RX}{\rho\omega}, \quad G' = \frac{R^2 - X^2}{\rho} \quad (6)$$

where ρ is the density. In what follows we have assumed $\rho = 1$.

The variations of η_3' and G' with temperature are shown in figure 3. We observe that

$$G'(25 \text{ MHz}) > G'(15 \text{ MHz})$$

and $\eta_3'(15 \text{ MHz}) > \eta_3'(25 \text{ MHz})$. By making use of the curves which represent eq. (3) and eq. (5), these results show that the relaxation frequency $(2\pi\tau)^{-1}$ is far lower the frequencies used in this study. Thus a reasonable estimation of the high frequency limit $\eta_3^0 - \delta\eta_3$ of the viscosity $\eta_3(\omega)$ is given by the value of η_3' at 25 MHz.

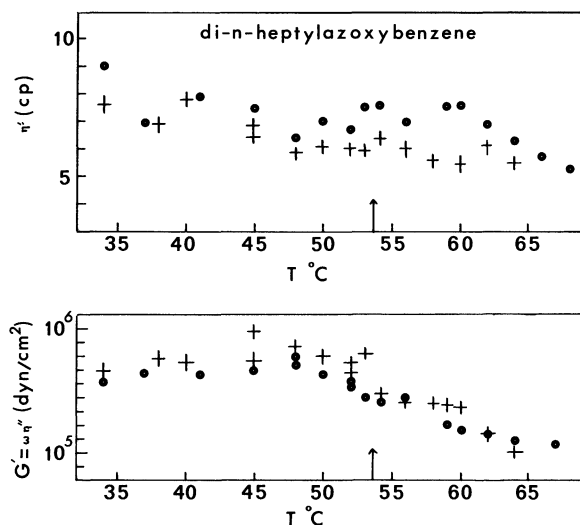


FIG. 3. — The apparent shear modulus G' and the real part η_3' of the viscosity in the nematic and smectic-A phases of di-n-heptylazoxybenzene. The symbols \bullet and $+$ are respectively for 15 MHz and 25 MHz.

In principle the relaxation frequency $(2\pi\tau)^{-1}$ (and the steady flow viscosity η_3^0) could be deduced from these measurements since it follows from eq. (3), eq. (4) and eq. (5) that :

$$\tau^{-1} = \frac{G'(\omega)}{\eta_3^0 - \eta_3'(\omega)} \quad (7)$$

which leads to :

$$\tau^{-1} = \frac{G'(\omega_2) - G'(\omega_1)}{\eta_3'(\omega_1) - \eta_3'(\omega_2)} \quad (8)$$

where $\omega_1/2\pi = 15 \text{ MHz}$ and $\omega_2/2\pi = 25 \text{ MHz}$.

However, it is clear from figure 3 that the differences involved in formula 8 are too small compared with experimental errors to deduce a reliable value of τ^{-1} .

On the other hand, it is hopeless to use the-impedometer method to check formulae (3) and (4) directly since the available frequencies which are studied by shear wave attenuation are much higher than the relaxation frequencies which characterize liquid-crystals in the nematic-smectic A phase transition. In this case light scattering provides more useful information [15-16].

b) *The nematic phase.* — In a usual nematic the director follows the shear at ultrasonic frequencies [2]. Since the transition here is nearly of second order we believe that the relaxation is due to the correlated regions of size $\xi(T)$ which cannot follow the ultrasonic wave. This effect is the same as the one described by de Gennes in the isotropic phase near the transition [4].

Diethylazoxybenzene has two mesomorphic phases, smectic A from 39° to 64.6 °C and nematic from 64.6° to 66.9 °C. The nematic to smectic-A phase transition is first order. Our measurements of R and X at 15 MHz are reported in figure 4. In the smectic-A phase we observe that R and X are quite different, as seen previously for the diheptyl-compound. In the nematic phase, we found $R \simeq X$, as expected. Since the A-N transition is first order, the relaxation frequency $(2\pi\tau)^{-1}$ is expected to be higher than that of the diheptyl-compound [17]. However, since the values of G' and η'_3 reported in figure 5 are of the same order of magnitude as those of the diheptyl-compound, the only thing which can be said is that the relaxation frequency is below 15 MHz. Thus the value of η'_3 at 15 MHz can be reasonably considered as an estimation of the high frequency limit $\eta'_3 - \delta\eta_3$.

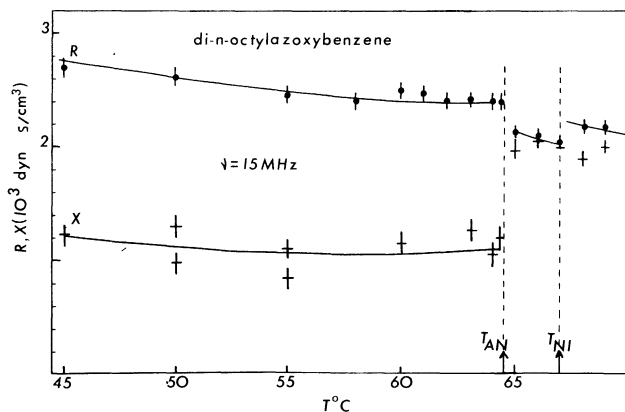


FIG. 4. — The real R and imaginary X parts of the shear impedance of di-n-octylazoxybenzene at 15 MHz.

We wish to conclude this letter with a remark on the ultrasonic behaviour of smectic-B phases. They are layered systems like smectic-A phases. Moreover they

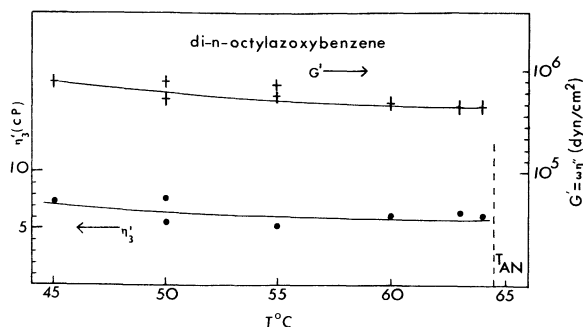


FIG. 5. — The apparent shear modulus G' and the real part η'_3 of the viscosity in the smectic-A phase of di-octylazoxybenzene at 15 MHz.

present an ordering inside the layers and some correlations between the layers. As suggested by de Gennes and Sarma [18], the B phase may correspond to a three-dimensional crystal with rotational freedom or to a system of decoupled layers which might slip on each other. The shear modulus c_{44} is non-zero in the first model and vanishes in the second model.

The observation of an apparent rigidity coefficient in smectic-A phases suggests, following the idea of a coupling between the shear flow and the director fluctuations, that the smectic-B phases which correspond to the second model ($c_{44} = 0$) would present in fact an apparent rigidity coefficient $G' = c_{44} \neq 0$ at ultrasonic and at higher frequencies. This is supported by our earlier study of the smectic-B phase of N-p-butyloxybenzylidene-p-n-octylaniline (BBOA) [19] where we have observed an apparent rigidity coefficient $G' = c_{44}$ whereas it was expected that we would observe the behaviour predicted by the second model ($c_{44} = 0$) since the interlayer correlation is weak (< 2 layers) [20] compared to the ultrasonic penetration depth.

An apparent rigidity coefficient c_{44} has also been observed on other compounds at 110 MHz [21] and at Brillouin frequencies [22]. The values of G' quoted in reference [21] are a hundred times higher than those found here. These high values may be caused by important correlations between layers or alternatively by director fluctuations since formulae (4) and (5) show that G' increases with the frequency. More detailed comparisons are not possible since the phases and the compounds are not the same. Moreover the director fluctuations may vary from the A to the B phase.

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