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A.C. SHEAR ELASTIC CONSTANT OF A SMECTIC B LIQUID CRYSTAL

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ABSTRACT : We have measured the low frequency A.C. complex shear modulus of a smectic B liquid crystal, parallel to the smectic layers for thin samples. The apparent elastic modulus C_{44} is very small ($\sim 10^6$ dyn/cm^2). It increases monotonically versus the frequency (from 10^{-1} to 5.10^3 Hz), following a logarithmical law. At a given frequency (10^3Hz) the elastic constant is measured close to a transition to a smectic A phase and shows a pretransitionnal softening. These results compare well with ultrasonic data from Thiriet and Martinoty demonstrating a large relaxation effect between 10^4Hz and 10^8 Hz.

Smectic B liquid crystals are layered systems, made of rod like organic molecules. X rays structural [1] data are now available on these materials : the molecules are normal to the layers; in each layer, there exists an hexagonal positional order, long range for most of the materials; there are also in general strong correlations of positional order between layers, so that the smectic B materials appear to be a class of plastic crystals.

To test on large scale the anisotropy of the structure, measurements of the shear modulus parallel to the layers have been made, at ultrasonic frequencies [2], or low frequencies [3]. The results show a very low elastic shear modulus C_{44}, in the range of 10^6 dyn/cm^2, typical of the lamellar structure. At very low frequencies a Newtonian shear creep has been demonstrated [4]. In addition, internal resonances at higher frequencies were explained as local distorted texture vibrations. Using thinner samples, we have been able to suppress almost completely the defects leading to these resonances. In this paper we present the frequency and temperature dependence of the layers parallel shear modulus C_{44} of a smectic liquid crystal in the smectic B phase, and near a higher symetry fluid smectic A phase, where the only remaining positional order is the lamellar order.

The compound we use is the 4.08 (butyloxy benzylidene octylaniline), which presents a smectic B to smectic A transition at 49°C. The sample, of thickness $d = 25$ µm, is placed in between two glass plates, in the "homeotropic" orientation (i.e. with the lamellae parallel to the plates), using a silane coating. The mechanical set up is the same as previously described [3,4]. A sinusoidal displacement $\mathrm{Re}(\delta \exp(i\omega t))$, parallel to the layers, is applied through piezoelectric ceramics to one face of the sample; amplitude and phase of the transmitted shear force are measured on the other face with identical piezoelectric ceramics. The results are described by the ratio $Z(\omega) = \sigma/\varepsilon \exp i\phi$ between the transmitted shear stress $\sigma \exp i(\omega t + \phi)$ and the applied shear strain $\varepsilon \exp(i\omega t)$, equivalent to a complex mechanical impedance. When the wave lengthes associated with shear waves are large compared to the thickness d of the sample, Z is caracteristic of the material. Otherwise, Z could contain a dephasing associated with wave propagation. In all the present work , we remain in the first case. The available frequencies range from 0.1 to 10^4 Hz. For higher frequencies, mechanical resonances of our set up become visible. The liquid crystal sample is temperature controled with a stability better than 10^{-2}°C. Before measurements, we control the sample quality by a routine optical "conoscopic" observation.

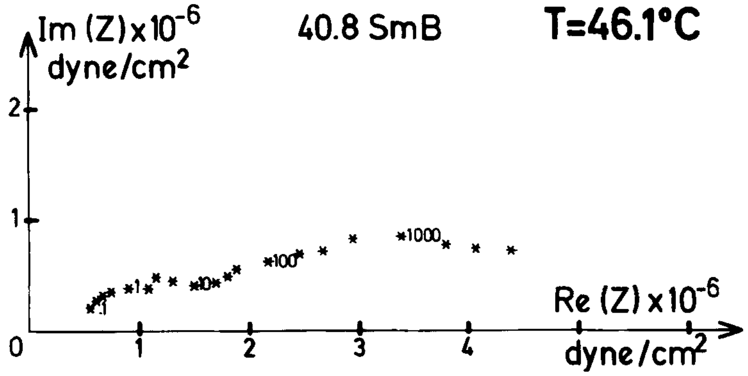

FIGURE 1 : Shear mechanical impedance Z(ν) in smectic B phase.
The numbers are the frequencies in hertz

We fix first the temperature at T = 46.1°C, in the smectic B range and we measure Z (ν) where $\nu = \omega/2\pi$ is the frequency. A typical plot is shown on Fig. 1, which represents Z (ν) in the complex plane, the axis being measured in units of dyn/cm^2. We have not studied in details the very low frequency range previously described [4] . In our range of frequency, we note a continuous increase of the real part of Z, i.e. of the elastic C_{44}. We had previously observed that C_{44} followed a logarithmic law in frequency. To check this behaviour at higher frequencies, we plot (Fig.2) C_{44} versus $\mathrm{Log}_{10}\ \nu$. We find now two logarithmic dependences : for low frequencies, C_{44} increases as cte + 0.57 10^6 $\mathrm{Log}_{10}\ \nu$. Note that this slope 0.57 10^6 is the same as that found previsouly in lower scale of frequencies from 10^{-2} to 10 Hz [4] . At high frequencies, the increase writes as 1.35 10^6 $\mathrm{Log}_{10}\ \nu$. The cross over between the two regimes appears around ν = 100 Hz.

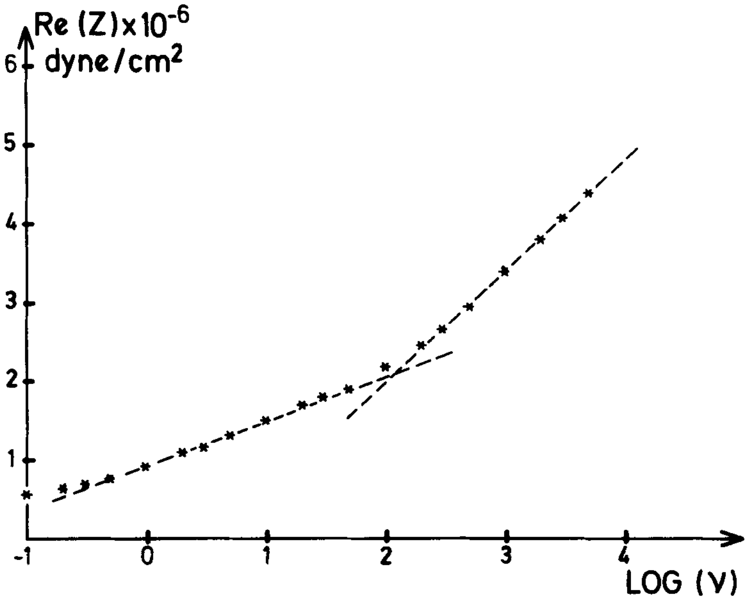

FIGURE 2 : Real part of $Z(\nu)$ versus $\mathrm{Log}_{10}(\nu)$ from the data of Fig.1. The two dashed straight lines represent the logarithmic laws described in the text.

At lower temperature, we observe the same two logarithmic laws with the same slopes; the cross over frequency however decreases when the temperature decreases, down to 20 Hz for T = 40°C. The simplest interpretation is to assume that this increase of C_{44} is due to the progressive freezing of the defects (edge dislocations of layers, a.s.o) responsible for the low frequency plastic response. Above the cross over frequency, a new kind of defects should become frozen. Note finally that with an average $C_{44} \sim 10^6$, the wave length at 1 KHz is about 1 cm, much larger than d = 25 μm, as above assumed.

It is now interesting to compare our results of C_{44} with the ultrasonic data of ref (2), for the same shear mode and the same compound at the same temperature. On Fig. 3, we have plotted the two sets of data versus $\mathrm{Log}_{10}\nu$. The two sets of results are compatible, if we accept the idea, already suggested in ref. (2) that the ultrasonic data are taken at frequencies around a relaxation process. It would be now interesting to cover the gap between 10^4 and 10^7 Hz, to check this point. For the higher frequencies one knows [5], but not for this compound, that C_{44} saturates around 10^8 dyn/cm^2, which reinforce the relaxation idea.

Im (Z) shows also a more or less regular increase in frequency, with small oscillations. The high frequency one is in fact what remains in the thin samples of the large internal resonance previously observed for thick samples, as we have checked by measuring Z (ω) in samples of intermediate thickness. We thus do not believe that these oscillations are really intrinsic properties of the material.

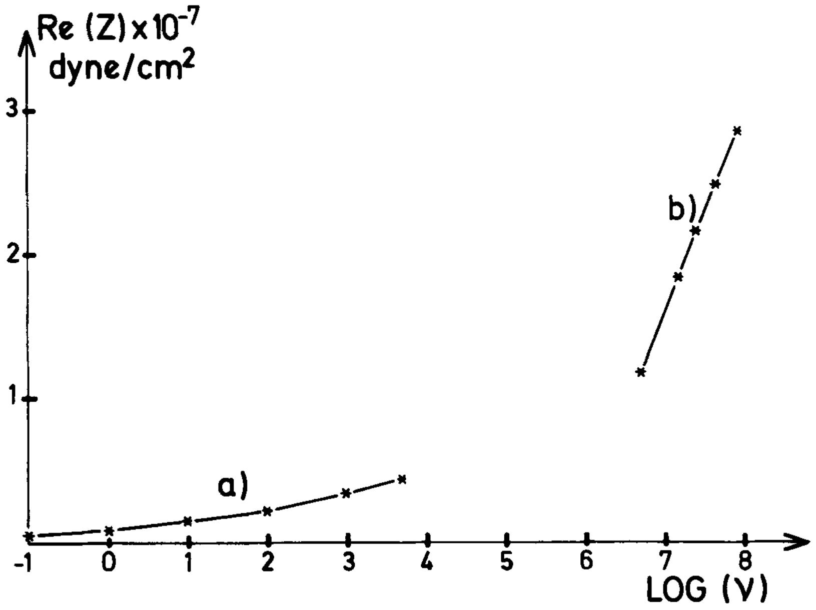

FIGURE 3 : Real part of Z (ν) versus Log_{10} (ν) at 46°C.
a) our present data b) data from ref.(2).

To measure the temperature dependence of the elastic constant C_{44}, we must now fix the frequency since we do not see any saturation of Re (Z(ω)). We have chosen $\nu = 10^3$ Hz, high enough in our available range, but not too close to our spurious mechanical resonances. Our results are plotted on Fig. 4, which represents C_{44} (1000 Hz) versus the temperature T. The transition temperature T_c toward the smectic A phase is 47.2°C, lower than the previously reported 49°C. This is probably indicative of a slight degradation of the material. In the S_A phase, C_{44} is zero as expected for a simple one dimensional piling of fluid layers. In the vicinity of T_c, on a range of 0.7°C, we observe a linear increase of C_{44} above zero. As the $S_A \rightarrow S_B$ transition is slightly first order, we can obviously relate this behavior to the coexistence range of the liquid and solid phases in this slightly impure system. At lower temperatures, we observe a continuous increase of C_{44}. The curvature of C_{44} close to T_c is a typical pretransitional effect. The same pretransitional effect has also been observed at ultrasonic frequencies [2] .

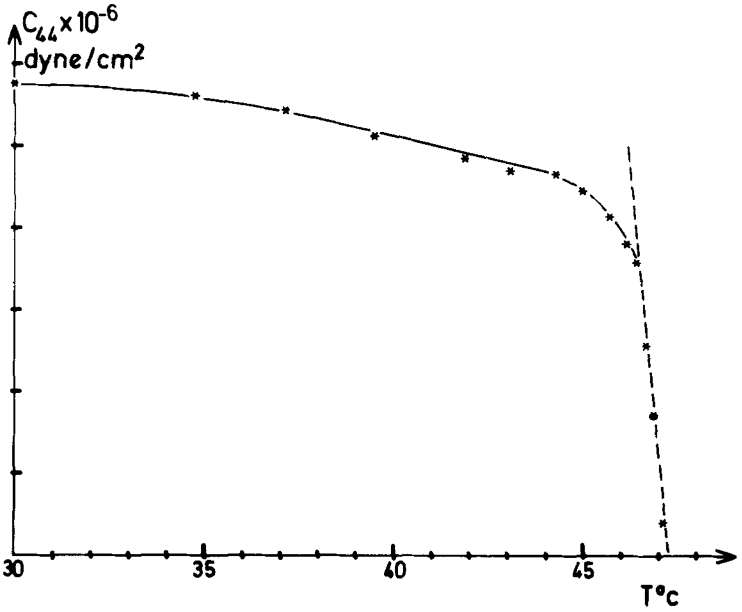

FIGURE 4 : Real part of Z at 1.000 Hz versus temperature. The solid and dashed lines are just a guide for the eye.

In conclusion, we have measured the viscoelastic response of a smectic B liquid crystal submitted to a shear strain parallel to the smectic layers. Our essential observation is a logarithmic increase of the apparent elastic constant C_{44} versus the frequency, in the range 10^{-1} to 10^4 Hz. At a given frequency, the temperature dependence of C_{44} exhibits a pretransitional softening close to the smectic B → smectic A phase transition. This novel flow situation suggest the need for an analytical model.

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