"SECOND SOUND" PROPAGATION AND THE SMECTIC RESPONSE FUNCTION

L. Ricard, Jacques Prost

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Abstract. — We present a study of the smectic response function close to the smectic A to nematic transition, as a function of temperature. The progressive evolution from an overdamped to an underdamped behavior is evidenced, and we can deduce a temperature dependence for the compressional elastic constant of octylcyanobiphenyl \((8\text{ CB}) B \propto (T_{NA} - T)^{2x}\) with \(2x \approx 0.61 \pm 0.05\).

1. Introduction. — In a smectic A phase, two propagating modes may exist [1, 2]. The first one corresponds to the usual sound (essentially a density modulation). The second is closely related to the motion of the layers characterising the \(S_A\) phase. In analogy to the superfluid case, it has been called second sound, but is in fact very similar with shear waves in a solid. The main difference with these last modes arises from its extreme anisotropy: for wave vectors either parallel or perpendicular to the layers the mode is overdamped, whereas for intermediate angles it is underdamped. The two cases have been separately observed [3, 4, 5] with the use of quite distinct techniques. In particular, the propagating behavior has been evidenced only for wave vectors directions at \(\Psi\) angles smaller than 45° (\(\Psi\) defines the angle of the wave vector with respect to \(\hat{z}\) the normal of the layers figure 1). The main difficulty with traditional techniques such as Brillouin scattering and ultrasounds resonance, lies in the very low propagation velocity of this second sound wave \((v \lesssim 300 \text{ m s}^{-1})\).

In the second section, we show how an adaptation of the interdigital electrodes technique provides a suitable tool for studying the shear mode of smectics. This allows us, to present in the third section the first observation of the progressive evolution from an overdamped to an underdamped mode at fixed \(\Psi\) angle \((\Psi \approx 85^\circ)\) (as a function of temperature), and deduce the temperature dependence of the compressional elastic constant of octylcyanobiphenyl \((8\text{ CB})\) in the vicinity of the transition to the nematic phase. The conclusion is devoted to a short comparison of our results with those obtained by low frequency techniques [6, 7].

2. The use of the « interdigital electrodes technique ». — Figure 1 displays the essential characte-
ristics of the shear wave in smectics: for propagation directions tilted with respect to $\tilde{z}$, it involves both compression and tilt of the layers. Because of its low propagation velocity, there is total internal reflection at the solid-liquid crystal interfaces [5]. If one can couple either to the compression or to the tilt, at a suitable wave vector and frequency, one expects to reach a resonance as in any acoustic experiment. The originality of our set up is to use interdigital electrodes, and the flexoelectric coupling to excite the resonance. The wave vector is defined by the sample spacing ($z$ component $q_z = \Pi/D$ in the lower mode) and the electrode geometry ($x$ component $q_x = 2 \Pi/\lambda e$); we refer the reader to reference [8] for a more detailed description of the electric field created by interdigital electrodes, and postpone for a more complete article, the derivation of the smectic response. Let us just remark, that the coupling with electric field gradients has the following form [9]:

$$\Delta E = f \delta n_x \frac{\partial E_z}{\partial z} - f' \theta \frac{\partial E_x}{\partial x}$$

(1)

$\delta n_x$ is the tilt angle of the director (unperturbed direction along $z$) and $\theta$ the bulk dilation (we assume that there is no field along $y$). As in the nematic phase $f = f_\parallel + f_z$: the first term being expected to be more important than the second one, the main effort on the sample is a torque exerted on each molecule; in the hydrodynamic limit it is transmitted to the layers which are consequently excited at the frequency $\nu$ of the applied field, with a wavelength along the $x$ direction corresponding to the period $\lambda_e$ of the field. This limit is defined by: $\nu \ll B_1/2 \Pi y$ in which $B_1$ is an elastic constant describing how difficult it is to tilt the molecules away from the normal of the layers, and $y$ the corresponding friction coefficient. Typical numbers seem to impose $\nu \ll 10^6 \times 10^7$ [7, 13].

The response of the smectic can be then calculated from the hydrodynamical equations of A Smectics [2] ($U$ layers displacement, figure 1):

$$U \propto \frac{1}{1 - \frac{\omega^2}{\omega_x^2} - \frac{j \omega}{\omega_x}}$$

(2)

with:

$$\omega_x^2 \sim \frac{Bq_x^2}{\rho}$$

(3)

and:

$$\omega_t \sim \frac{q_t^2 B}{\eta q_x^2}$$

(4)

under the assumption $q_x^2 \gg q_t^2$ (in our experiment: $q_x^2 > 100 q_t^2$)

$$B = B - \frac{C^2}{A}$$

(5)

$\omega_t/2 \Pi$ is the resonance frequency of the so-called second sound, while $\omega_t$ governs the damping. $A$, $B$ and $C$ are elastic constants corresponding to the bulk compression, the layer compression, and a crossed term between bulk dilation and the layers. $\eta$ is a shear viscosity expected to exhibit no temperature dependence at the smectic A to nematic transition ($\eta = \frac{4 \eta + 2 \rho e}{2}$ in De Gennes notations [10]).

For $\omega_t > \sqrt{2} \omega_r$, the response is overdamped, there is no maximum in $u(\omega)$; on the contrary for $\omega_t < \sqrt{2} \omega_t$ the response exhibits a maximum at the frequency

$$\nu_m = \frac{1}{2 \Pi} \left( \frac{\omega_t^2 - \omega_r^2}{\omega_r^2} \right)^{1/4}.$$  

(6)

The condition for observing the resonance reads in terms of experimental parameters:

$$\frac{q_x^2}{q_t} < \frac{(2 \rho B)^{1/2}}{\eta}$$

If one wants to keep $\omega_t/2 \Pi$ in the $10^5$ Hz range, and at the same time observe a propagating mode (anticipating $B \sim 10^8$ erg cm$^{-3}$ and $q_x = \Pi/D \approx 60$ cm$^{-1}$ imposed by observability requirements), one has to choose a small $q_t$. Our choice $\lambda_e = 2 \Pi q_x = 100$ $\mu$m, is in fact the only difference with our previous experiments in the smectic phase [8], in which we had $\lambda_e = 5$ $\mu$m.

3. Experimental results. — We used a set up similar to the one already described in reference [8]. This is basically a forced Rayleigh scattering technique. The distortion $U(q)$ generates an index of refraction modulation, which scatters light in well defined directions, corresponding to the standard wave vector conservation laws. Whereas for $q_x$ wave vectors larger than a few $10^3$ cm$^{-1}$, depolarized scattering is strongly favored in smectic A phases (and gives rise to the so-called crescent figure), polarized scattering was found to yield the best results with an incidence angle of 20°, in this case ($q_x = 628$ cm$^{-1}$). A 124 Spectra Physics H$_2$N$_2$ laser was used as the light source; the beam irradiating the whole sample area ($\sim 1$ cm$^2$) was focused on the cathode plane of the photodetector (XP 1002 RTC PM tube), and a 100 $\mu$m diameter pinhole allowed to select a coherence area in the direction corresponding to the forced Rayleigh scattering. Optical heterodyning, and a phase sensitive detection (124 A. PAR Lock in amplifier) were used to measure the scattered amplitude as a function of frequency. The sample was kept under Argon atmosphere, in a temperature controled oven stable within 10 mK a day. The good chemical stability of 8 CB is demonstrated by the
nearly constance of the nematic to smectic A transition temperature (70 mK shift over a month). The sample thickness was $D = 545 \mu m \pm 15 \mu m$, thus leading to a $\Psi$ angle of nearly 85°. The interest of large thicknesses is two fold: first the scattered amplitude is larger the thicker the sample is; second, the resonance frequency may be brought down within the detection range of our system.

![Figure 2](image)

**FIG. 2.** The smectic response function of 8 CB versus frequency for a set of different temperatures. The crosses correspond to the measured values of the scattered amplitude at the corresponding frequency (see the text, section 3). The curves are vertically shifted from an arbitrary amount for clarity sake, but the frequency scale is respected. The solid line represents the best fit obtained from equation [2].

Typical experimental results are displayed on figure 2. For temperatures close to $T_{NA}$, $B$ is small, and the inequality (6) cannot be fulfilled: the smectic response is fully overdamped; however the existence of the shear mode, and its basic difference with the nematic behaviour may be inferred from the measurement of the phase lag of the scattered amplitude with respect to the applied electric field: in the nematic case, the phase lag is $\Pi/2$ in the high frequency regime, whereas in this case we can reach a domain $\omega > \omega_s > \omega_c$ for which the phase lag is $\Pi$. For $T - T_{NA} \sim 0.1 K$, the inequality (6), begins to hold and upon further cooling, one clearly sees a maximum developing in the response function. To our knowledge it is the first observation of a resonant behaviour at large $\Psi$ angles, and also the first observation of the progressive shift from a purely overdamped to an underdamped behaviour. The fit of the experimental curves involves two parameters, which directly give $(B/\rho)^{1/2}$ and $B/\eta$. The success of equation (2), confirms the validity of the use of the hydrodynamic limit. The knowledge of $\rho$ from direct density measurements [11] allows us to obtain the $B$ and $\eta$ values. We find $\eta \approx 1.6$ poise basically temperature independent, and $B \approx 2.6 \times 10^6$ dyne cm$^{-2}$ for

$$|T - T_{NA}| = 1 K .$$

We have plotted on figure 3, the temperature dependence of $B^{1/2}$ and $B$ obtained respectively from the

![Figure 3](image)

**FIG. 3.** — $B^{1/2}$ and $B$ temperature dependence: on the left scale, $B^{1/2}$, as calculated from the second sound resonance frequency ($\omega_c/2\Pi$), on the right scale $B/\eta \propto \omega_s$, obtained from the damping. The dashed line corresponds to a least square fit $B^{1/2} \propto (T_{SA} - T)^z$ and the solid line $B \propto (T_{SA} - T)^z$ with $T_{NA} = T_{SA} = 32.81 ^oC$; $x = 0.28 \pm 0.04$ in the first case, $2x = 0.61 \pm 0.05$ in the second.

in the second.

The resonably good agreement between the two values confirms the very weak temperature dependence of $\eta$. These values correspond to the best fit obtained with a least mean square procedure. In particular the best $T_{NA}^*$ value was found to be exactly the experimental transition temperature

$$(T_{NA}^* = T_{NA} = 33.81 ^oC) .$$

However, one knows that the nematic to smectic A transition in 8 CB is first order, which implies that one should have $T_{NA}^* > T_{NA}$; imposing

$$T_{SA} - T_{NA} = 10 mK$$

leads to $2x = 0.66$. Of course this should not be taken too seriously, but it however indicates that our results are in rather good agreement with the helium analogy [12]. This result further agrees with the most recent critical exponent values obtained in the nematic phase of 8 CB [13].

4. Conclusion. — From a theoretical point of view, three different results are possible:

$2x = 0.5$ near a tricritical point,

$2x = 0.66$ in the helium analogy,

$2x = 1$ if mean field theory can be applied.

Our results rule out mean field behaviour and although the helium analogy seems to be appropriate, we feel that further experimentation is needed to clearly assess this point.

An other striking feature, is the marked difference with previous $B$ measurements, which all lead to $\beta$ values typically twice smaller [6, 7, 13]. The first idea would be to invoke the frequency difference between the previous and our measurements. We show in particular on figure 4, how the low frequency response function differs from what it should be, if there was no dislocation motion. However, the
FIG. 4. — The low frequency smectic response function. The crosses are experimental data points. The solid line corresponds to a single relaxation process \( \nu, L \times 50 \text{ Hz} \) superimposed to the high frequency response (presumably, the dislocation motion provokes a renormalization of the elastic constant from a value \( B_{\text{res}} \) to a smaller value \( B^{\text{eff}} \), as described in reference [14].

characteristic time that we estimate for this motion corresponds exactly to the one measured in the instability experiments [6], thus suggesting that in both cases one should be able to get rid of this effect. More basic may be the wave vector at which the two sets of experiments are performed.

In the instability and in the MIT Rayleigh scattering experiments, \( q \) is typically of the order of \( 10^5 \text{ cm}^{-1} \), whereas in this case it is \( 6.28 \times 10^2 \text{ cm}^{-1} \). As a consequence, although the typical frequencies at which both sets of experiments are performed differ only by a little more than a factor of ten, one may be in quite different regimes, with respect to certain particular hydrodynamic modes. This may be true for impurity diffusion, and is quite certain for thermal diffusion: our measurements concerns \( B \) taken at constant entropy, whereas the previous ones were characteristic of \( B \) at constant temperature. It is our opinion that this point should deserve a closer attention.

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