Study of the velocity and absorption of second sound in smectic liquid crystals

A. A. Tabidze, G. P. Abramkin, and N. I. Koshkin
N. K. Krupskaya Pedagogical Institute, Moscow

(Submitted 15 June 1987; resubmitted 2 March 1988)
Pis'ma, Zh. Eksp. Teor. Fiz. **47**, No. 8, 386–389 (25 April 1988)

An acoustic method was used to measure, at a frequency of 3 MHz, the orientational dependence (0–90°) and the temperature dependence of the biasing impedance of smectic *A*, *C*, *B*, *F*, and *G* mesophases of pentyloxybenzylidene hexyleniline. The orientational dependence of the absorption of orthogonal smectic mesophase was found to differ from that of the oblique smectic mesophase.

Considerable interest has recently been shown in theoretical and experimental studies of the dynamics of smectic liquid crystals. No information, however, is available in the literature on the acoustic studies of the propagation velocity and absorption of shear waves in smectic liquid crystals, which must be known in order to verify

 0021-3640/88/080461-05$01.00

hydrodynamic theories, on the study of the nature of mesophase transitions, and on the estimates of the values of C_{44}. In this letter we present experimental results on the anisotropic propagation of second sound in smectic liquid crystals of the *A*, *C*, *B*, *F*, and *G* mesophases.

As the object to be studied we chose a single-layer smectic liquid crystal pentyloxybenzylidene hexylaniline (PBHA) with an array of smectic phases with the following mesophase transition temperatures: I–346.2 K–N–334.6 K–S_A–326 K–S_C–323.8 K–S_B–315.2–S_F–312.6–S_G–308.2 K–Cr. In the experiment we used a method of measuring a biasing acoustic impedance,[1] $Z = R + jX$. The components of the impedance R and X were measured within $\pm 3\%$. The propagation velocity v_S, the absorption coefficient α_S of the shear waves, and the dynamic shear modulus $G^* = G' + j\omega\eta'$ were calculated from the standard relations

$$v_S = (R^2 + X^2)/\rho R, \quad \alpha_S = \rho\omega X/(R^2 + X^2), \quad G' = (R^2 - X^2)/\rho, \quad \eta' = 2RX/\omega\rho, \tag{1}$$

where ρ is the density of the liquid crystal, $\omega = 2\pi f$, and f is the ultrasonic velocity.

The shear waves were excited at a frequency of 3 MHz by means of a quarz resonator with an *AT* cut. The liquid crystal molecules were oriented by a 2.2-*T* magnetic field in the nematic phase. The measurements were carried out after cooling the liquid-crystal sample through a N-S_A junction for each angle θ in the range from 0° to 90° in 5° increments; θ is the angle between the wave vector **k** and the normal to the smectic layer **n**. The wave-shift vector **u** lies in the **k**,**n** plane. For the magnetic field used in the experiment the magnetic coherence length $\xi(H)$, which characterizes the orienting effect of the surface of the measuring element, is $\sim 10^{-6}$ m, which is much smaller, for the angles $0° < \theta < 90°$, than the depth at which the shear waves propagate. In other words, the condition for bulk orientation of the samples was satisfied completely. In setting up the experiment special care was taken in orienting the angles $\theta = 0°$ and 90°. The working surface of the resonators was treated with various surface-binding substances: silanes which produce on the surface of the resonators a homeotropic orientation ($\theta = 0$) and planar orientation ($\theta = 90°$) of the liquid-crystal molecules.[2]

The orientational behavior of the active component $R(\theta)$ and the reactive component $X(\theta)$ of the biasing acoustic impedance in various PBHA phases is shown in polar coordinates in Figs. 1 and 2. The impedance components R and X are anisotropic in the smectic *A*, *C*, *B*, *F*, and *G* mesophases. For the angles $\theta = 0$ and $\theta = 90°$ the impedance components in the smectic *A* and *C* mesophases exhibit Newtonian properties, $R \approx X$ within 10%. In the smectic *B*, *F*, and *G* phases $R > X$ for the angles $\theta = 0$ and $\theta = 90°$. This result shows that these phases are not true smectic liquid crystals but are rather multiple-layer crystals with a C_{44} modulus.

At the angles $0° < \theta < 90°$ the elastic properties increase rapidly, $R \gg X$. In all the mesophases the orientational $R(\theta)$ curve goes through a maximum near the angle of 45°. Worth noting here is the fact that mesophases with different symmetries have qualitatively different orientational $X(\theta)$ curves. In the orthogonal *A* and *B* phases the $X(\theta)$ curve goes through a minimum. In the oblique *C*, *F*, and *G* phases the $X(\theta)$

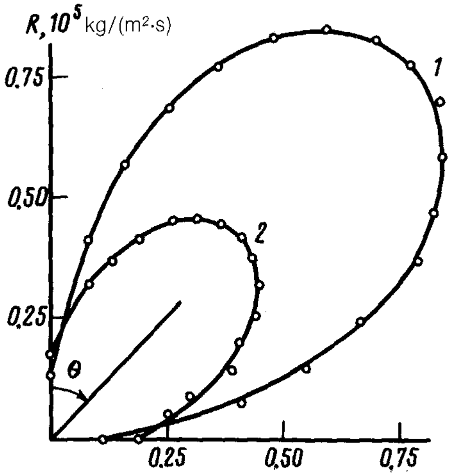

FIG. 1. Angular dependence (0–90°) of the active component of the impedance $R(\theta)$ in S_A (329 K) (curve 1) and S_C (324 K) (curve 2) of PBHA at a frequency of 3 MHz. Solid curves—Theoretical curves based on Eq. (5).

curve is similar in shape to the $R(\theta)$ curve, i.e., the $X(\theta)$ curve goes through a maximum near the angle of 45°.

According to the classical hydrodynamic theory, rapid enhancement of the elastic properties of smectic liquid crystals occurs as a result of the contribution of the second-sound mode. The orientational behavior of the velocity, $v_2(\theta)$, and the absorption, $\alpha_2(\theta)$, of second sound can be described as[3,4]

$$v_2(\theta) = 0.5(B_0/\rho)^{0.5}\sin 2\theta\,, \tag{2}$$

$$\alpha_2(\theta) = \frac{\omega^2}{2\rho v_2^3(\theta)}\,[\,\eta_3\cos^2 2\theta + \eta_{(2)}\sin^2 2\theta\,]\,. \tag{3}$$

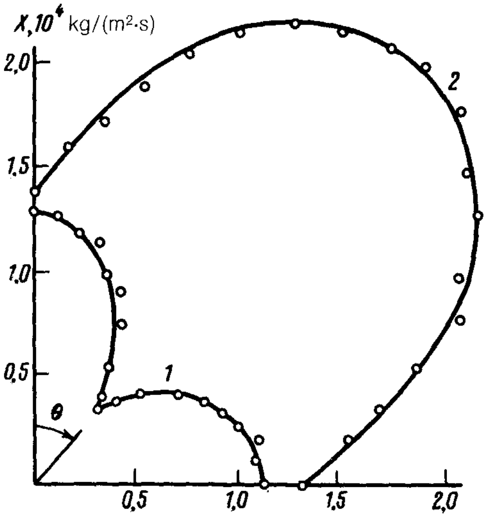

FIG. 2. Angular dependence (0–90°) of the reactive component of the impedance $X(\theta)$ in S_A (329 K) (curve 1) and S_C (324 K) (curve 2) of PBHA at a frequency of 3 MHz. Solid curves—Theoretical curves based on Eq. (6).

Here $\eta_{(2)} = 0.25(\eta_1 + \eta_2 + \eta_4 - 2\eta_5)$, where $\eta_1 - \eta_5$ are the coefficients of viscosity in the notation used in Ref. 3, and B_0 is the transverse-contraction modulus of the smectic layers.

We can thus write the expression for the orientational behavior of the complex shear modulus in the form

$$G^*(\theta) = 0.25B_0 \sin^2 2\theta + j\omega(\eta_3 \cos^2 2\theta + \eta_{(2)} \sin^2 2\theta). \tag{4}$$

From (1) and (4) we find the relations for the orientational behavior of $R(\theta)$ and $X(\theta)$

$$R(\theta) = (R_g^2 \cos^2 2\theta + R_{(2)}^2 \sin^2 2\theta)^{0.5} \tag{5}$$

$$X(\theta) = (X_g^2 \cos^2 2\theta + X_{(2)}^2 \sin^2 2\theta)^{0.5}, \tag{6}$$

where

$$R_{(2)} = 0.5(B_0 \rho)^{0.5}, \quad X_g = (\pi\rho f \eta_3)^{0.5}, \quad X_{(2)} = (\pi\rho f \eta_{(2)})/R_{(2)}.$$

Although (2) and (3) in the given form can be used for S_A, they can nevertheless be used also for the other phases in the first approximation. The exception here is the consideration of the angles $\theta = 0°$ and $\theta = 90°$ for S_B, S_F, and S_G.

In the *A*, *C*, *B*, and *F* phases the experimental values of $R(\theta)$ and $X(\theta)$ are described within 5%, by expressions (5) and (6). For smectic G the agreement is within $\sim 10\%$.

From (5) and (6) we see that the second-sound mode becomes a diffusion mode as a result of a change in the angle θ.

At the angle $\theta = 0$, $R \approx X$ in S_A and S_C within 10%. We can then write the impedance components of the diffusion mode in the form

$$R_g = X_g = (\pi\rho f \eta_3)^{0.5}. \tag{7}$$

A sharp minimum on the $X(\theta)$ curve for S_A and S_B indicates that $X_g > X_{(2)}$ in these planes, whereas the converse is true for S_C, S_F, and S_G (the smectic crystals with an oblique orientation of molecules in the layer): $X_{(2)} > X_g$.

The second-sound velocities v_2 and the dissipation coefficients η_3 and $\eta_{(2)}$ were calculated from the experimental values of $R(\theta)$ and $X(\theta)$ We found the following values: $v_2(S_A$, 329 K$) = 110$ m·s^{-1}, $\eta_3 = 17$ mPa·s, $\eta_{(2)} = 97$ mPa·s, $v_2(S_C$, 324 K$) = 71$ m·s^{-1}, $\eta_3 = 22$ mPa·s, $\eta_{(2)} = 170$ mPa·s; $v_2(S_B$, 318 K$) = 370$ m·s^{-1}, $v_2(S_F$, 314 K$) = 209$ m·s^{-1}, $v_2(S_G$, 309 K$) = 225$ m·s^{-1}.

The values of the coefficient η_3, which were found from the analysis of the measurements of the acoustical and diffusion modes, are in agreement within 5%. The effect of the first-sound mode v_1, which is proportional to v_2^2/v_1^2, on the results of the measurements is no greater than 4%. We have therefore disregarded it.

Calculation of the interlayer shear modulus C_{44} for S_B yields the value $(7 \pm 1) \times 10^5$ Pa ($T = 318$ K).

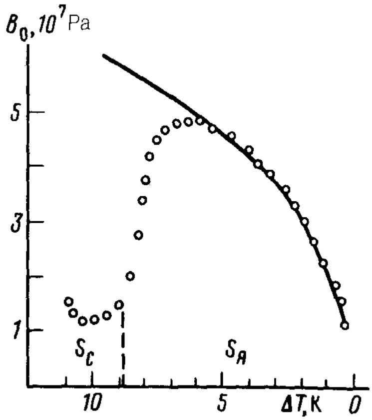

FIG. 3. Temperature dependence of the modulus B_0 in smectic A and smectic C phases of PBHA. Solid line—Theoretical curve.

The critical behavior of the transverse-contraction modulus B_0 of the smectic layers can be studied on the basis of the measurements of the temperature dependence of the impendance components. Figure 3 shows the temperature dependence of B_0 for smectic crystal A. The behavior of $B_0(T)$ in S_A can be represented by the function $B_0(T) = B_1(T_{NA} - T)^{\phi}$, where ϕ is a critical index, and B_1 is the value of B_0 at $\Delta T = T_{NA} - T = 1$ K. In our experimental study we found the values $B_1 = (2.3 \pm 0.1) \times 10^7$ Pa and $\phi = 0.44 \pm 0.2$. The values of ϕ are in agreement with the results obtained by Fisch *et al.*[5] for certain single-layer smectics A.

We wish to thank E. I. Kats, V. V. Lebedev, and É. V. Gevorkyan for a discussion of the results and for valuable comments. We also thank P. Adomenas for furnishing PBHA.

[1] A. A. Tabidze and R. Kh. Kazakov, Izm. tekh., No. 1, 34 (1983).
[2] A. A. Tabidze and N. I. Koshkin, Zh. Fiz. Khim. **60**, 1501 (1986).
[3] P. S. Martin, O. Parodi, and P. S. Pershan, Phys. Rev. **A6**, 2401 (1984).
[4] K. Miyano and J. B. Ketterson, Phys. Acoust. Principles and Methods, New York, **14**, 93 (1979).
[5] M. R. Fisch, P. S. Pershan, and L. B. Sorensen, Phys. Rev. **A29**, 2741 (1984).

Translated by S. J. Amoretty