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An ultrasonic investigation of the critical behaviour of the elastic moduli near the smectic C–smectic A phase transition

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Results for the frequency–temperature dependence of the elastic moduli and the longitudinal sound velocity near the S_C–S_A transition are reported. The measurements were made at frequencies in the range 0·15–27 MHz. The interpretation of the results obtained was made using the theory of critical dynamics taking into consideration the bare anisotropy.

1. Introduction

The experimental study of the critical dynamics in smectic liquid crystals is of great interest. This is due primarily to the strong initial anisotropy which serves as a background for critical phenomena in liquid crystals. This anisotropy could not be observed in common liquids in which critical phenomena have been studied fully. This feature leads to a number of peculiarities of both an experimental and theoretical character. First, the elastic properties of smectic phases are anisotropic. They are described not by a single elastic modulus but by several constants. The longitudinal sound velocity possesses a very complicated dependence upon the propagation direction. Thus, the experimental study of critical phenomena in liquid crystals demands a monodomain sample with a large volume, the precise mutual orientation of the initial wavevector and director, and an accurate account of the director surface distortions. Second, due to the strong anisotropy of smectics, the universal models with the multicomponent order parameters, which are traditional for the critical phenomena theory, considered in isotopic space, are not relevant for the description of the smectic C–smectic A phase transition. The point is that a totally universal behaviour takes place over a very narrow temperature range near the transition on the infrafrequencies. This temperature–frequency range is not achievable experimentally. Real experiments are made in the crossover regime. The fluctuations in this case are not strong enough to lead to the whole isotropization of the smectic. That is why, to interpret the experimental data in this case, we need an exact analysis of the critical behaviour at the strong bare anisotropy background.

In [1] the results of an ultra-acoustic investigation of the critical behaviour of the viscocity coefficients were described. In this work the original results concerning the frequency–temperature dependences of the elastic moduli and the longitudinal sound velocity near the smectic C–smectic A transition are reported. The measurements were made in the frequencies range 0·15–1·3 MHz by means of the resonator method [2] and the modified pulse–phase method with an alternating frequency [3]. The interpretation of the results obtained was made using the theory of critical dynamics [5], taking into account the initial anisotropy.

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2. Theory; critical behaviour of the elastic moduli and the longitudinal sound velocity

Here we briefly describe the critical behaviour of liquid crystal near the smectic C–smectic A phase transition. The problems not referring directly to the elastic moduli and to the first sound velocity will be omitted. The details of the calculation can be found in [4, 5], which are specially dedicated to the theoretical investigation of the S_A–S_C transition.

It is well known that in the equilibrium state smectics consist of a system of equidistant layers, situated at a distance l one from the other. The different smectics phases vary in the manner of location of the anisotropic molecules inside the smectic layer. The average direction of the principal molecular axis in the liquid crystal is defined by the unit vector **n** (director). In the smectic A phase the director is perpendicular to the layer. So, the director n and the unit vector of the layer normal v coincide. In a smectic C phase the director is tilted by some angle to the layer normal. The relevant order parameter for the phase transition between smectic A and smectic C phases is the vector [5]

$$\psi = [\mathbf{n}\mathbf{v}]. \tag{1}$$

Its modulus is identically zero in the smectic A phase and differs from zero in smectic C. In the smectic C phase the director **n** is defined by the layer normal vector **v** and the order parameter vector ψ.

In addition to this order parameter the whole list of the long wavelength (Goldstone) variables of the smectic also includes: the specific entropy σ, the density ρ and the smectic variable u, which plays the role of the shift of the smectic layer along the vector normal **v** [6]. In the vicinity of the transition only the order parameter ψ fluctuates strongly. All of the other variables fluctuate weakly. The energy density of smectic near the S_C–S_A transition is a function of the long wavelength variables. To investigate the peculiarities of the phase transition it is sufficient to keep in the energy expansion only the lowest order terms in ψ. For the terms describing the interaction of the order parameter with the weakly fluctuating variables, it is sufficient to keep those which are linear in the latter. In the energy part, which does not contain ψ, it is sufficient to restrict this to terms quadratic in the weakly fluctuating variables. As a result, the energy density of liquid crystal near the S_C–S_A phase transition is

$$E = E_1 + E_2 + E_3 + E_4, \tag{2}$$

$$E_1 = \frac{A\psi^2}{2} + \frac{U\psi^4}{4}, \tag{3}$$

$$E_2 = \frac{K_1(\mathbf{v}(\nabla\psi))^2}{2} + \frac{K_2(\nabla\psi)^2}{2} + \frac{K_3((\mathbf{v}\nabla)\psi)^2}{2} + \frac{K(\nabla^2 u)^2}{2}, \tag{4}$$

$$E_3 = \psi^2 [D_\rho \quad D_\sigma \quad D_u] \begin{bmatrix} \delta\rho/\rho \\ \delta\sigma/\sigma \\ \nabla_z u \end{bmatrix}, \tag{5}$$

$$E_4 = \begin{bmatrix} \delta\rho/\rho \\ \delta\sigma/\sigma \\ \nabla_z u \end{bmatrix} \begin{bmatrix} g_{\rho\rho}^- & g_{\rho\sigma}^- & g_{\rho u}^- \\ g_{\sigma\rho}^- & g_{\sigma\sigma}^- & g_{\sigma u}^- \\ g_{u\rho}^- & g_{u\sigma}^- & g_{uu}^- \end{bmatrix} \begin{bmatrix} \delta\rho/\rho \\ \delta\sigma/\sigma \\ \nabla_z u \end{bmatrix}^{\mathrm{T}}. \tag{6}$$

Here $\delta\rho$, $\delta\sigma$, u are the deviations from the equilibrium values for the density, the specific entropy and the smectic variable, respectively. In the energy density (2), the terms in expansion (3) determine the connection between the director and the layer normal ν. The phase transition takes place depending on the A parameter: when A is positive, then the S_C phase is realised; when A is negative the S_A phase is obtained. Item (4) in the energy density is connected with the spatial deformation of the director. The parameters K_1, K_2, K_3 and K in equation (4) are the Frank moduli. The terms in equation (5) describe the contribution of the order parameter interacting with the weakly fluctuation variables to the energy density. The last item (6) is associated with the deviation of the weakly fluctuating variables ρ, σ, u from their equilibrium values. Equations (5)–(6) are written in the matrix form.

In the equation (5) the vector

$$\begin{pmatrix} D_\rho \\ D_\sigma \\ D_u \end{pmatrix} = \begin{pmatrix} \dfrac{\partial A}{\partial \rho}\rho \\ \dfrac{\partial A}{\partial \sigma}\sigma \\ \dfrac{\partial A}{\partial(\nabla_z u)} \end{pmatrix} \tag{7}$$

is introduced. As it follows from equation (5), the components of this vector characterize the extent of the order parameter interaction with the fluctuations in the density $\delta\rho$, the specific entropy $\delta\sigma$ and the layer shift u.

In equation (6) the elastic adiabatic moduli

$$\left.\begin{aligned} g_{\rho\rho}^{-} &= \rho^2 \frac{\partial^2 E}{(\partial\rho)^2_{u,\sigma}} = \rho\frac{\partial P}{\partial\rho_{u,\sigma}}, \\ g_{uu}^{-} &= \frac{\partial^2 E}{(\partial \nabla_z u)^2_{\rho,\sigma}} = B, \qquad g_{u\rho}^{-} = \frac{\partial^2 E}{\partial\rho\partial\nabla_z u} = -B\frac{\partial(\ln l)}{\partial(\ln\rho)_\sigma}. \end{aligned}\right\} \tag{8}$$

were introduced. Here P is the pressure, B is the smectic layer compression modules, $g_{\rho\rho}^{-}$ is the elastic compression modulus for a constant interlayer distance. For nomal liquids the last term equals the reversed compressibility. The terms

$$-\frac{\partial(\ln l)}{\partial(\ln\rho)_\sigma} \sim -\frac{\partial(\ln l)}{\partial(\ln\sigma)_\rho} \sim 1, \tag{9}$$

appearing in equation (8) are the order of unity. These terms determine the interlayer distance as a function of the density (pressure) and the specific entropy (temperature).

We note that the relation of the elastic moduli, associated with the smectic layer compression, is

$$\frac{g_{uu}^{-}}{g_{\rho u}^{-}} = -\frac{\partial(\ln l)}{\partial(\ln\rho)_\sigma}. \tag{10}$$

In general, we ought to add to the elastic moduli in equation (8) the moduli associated with the entropy fluctuation $g_{\sigma\nu}^{-}$. However, in the following discussion these parameters do not play any role when ultrasound is studied and so we shall not discuss them further.

Before describing the critical behaviour of the parameters, introduced by equations (7)–(10), we shall consider some typical values for the material parameters of a smectic. The compressibility of a smectic is of the same order as in a normal liquid. Thus, the elasticity is

$$g^{-}_{\rho\rho} \sim 10^3 \,\mathrm{J\,cm^{-3}}. \tag{11}$$

The compression modulus of the smectic layers has the typical value

$$g^{-}_{uu} \sim g^{-}_{u\rho} \sim 10 \,\mathrm{J\,cm^{-3}}. \tag{12}$$

It is obvious from these values that there is the following small parameter for smectic phases

$$\frac{g^{-}_{uu}}{g^{-}_{\rho\rho}} \sim \frac{g^{-}_{u\rho}}{g^{-}_{\rho\rho}} \sim 10^{-2}. \tag{13}$$

This small value reflects the weakness of the smectic density modulation due to the vicinity of the smectic to the nematic phase. The existence of this small parameter greatly simplifies the study of the smectic dynamics. The parameters D_ρ, D_σ, D_u, appearing in equation (2) for the energy density, describe the contribution to the energy, coming from the interaction of the order parameter with the weakly fluctuation variables. The parameters are of the order of the compression modulus for smectic layers

$$D_{\rho,\sigma,u} \sim 10 \,\mathrm{J\,cm^{-3}}, \tag{14}$$

$$U \sim 10 \,\mathrm{J\,cm^{-3}}. \tag{15}$$

As shown by a previous analysis [5], taking into account the order parameter fluctuations does not lead to any corrections to the gradient terms (3) in the energy. This is true for the mean field theory as well as for the broad area of advanced fluctuations. The Frank moduli renormalization, which is adequate for the completely universal behaviour of the ψ^4-model with the two component order parameter, will occur only in a very narrow region near the transition. This region is not achievable experimentally, and it will not be considered in this paper. Thus the term the advanced fluctuations area will mean the area of non-universal critical behaviour. The critical behaviour in this area is described [4] by the non-universal indices, which are dependent on the bare relation for the Frank moduli K_1/K_3, K_2/K_3. The value of these indices, as a function of the Frank moduli relation, were calculated in the one-loop approximation in [4].

Unlike the Frank moduli, the elastic moduli are very sensitive to the vicinity of the smectic C–smectic A transition. In the mean field theory they undergo a jump during the transition

$$\left.\begin{aligned} g^{-}_{\rho\rho} &= -D_\rho^2/2U, \qquad g^{-}_{uu} = -D_u^2/2U, \\ g^{-}_{\rho u} &= -D_\rho D_u/2U. \end{aligned}\right\} \tag{16}$$

It is obvious from the estimates (11), (13) and (14), that these moduli decrease by a value of the order of the modulus g^{-}_{uu} in the S_C phase. Near the phase transition the critical fluctuations of the order parameter renormalize these moduli. They start to be dependent on the frequency ω and the vicinity τ to the transition point.

$$\tau = (T - T_{S_CS_A})/T_{S_CS_A}. \tag{17}$$

As shown by the analysis [5], taking into account the critical fluctuations in the dynamics leads to imaginary corrections to the bare values g^-. The renormalized dynamical moduli, in contrast to the bare g^-, will be designated later on by the symbol $\tilde{g}^-$. The renormalized elastic moduli possess the following form:

$$\tilde{g}_{\mu\nu}^- = g_{\mu\nu}^- - \mathrm{Re}\left\{\frac{D_\mu D_\nu F}{1+(D_\kappa g_{\kappa\gamma} D_\gamma)F}\right\}. \tag{18}$$

Here g^- is the matrix of the bare elastic moduli (see equations (6) and (9)), g is the matrix of the initial compressibilities

$$g_{\mu\nu} g_{\nu\gamma}^- = \delta_{\mu\gamma}. \tag{19}$$

As usual, in the matrix form, the sum is taken over the repeated indices (the dummy Greek indices run over the values u, ρ, σ). The vector $\mathbf{D}$, appearing in equation (18), was determined from equation (7). The first term on the right-hand side of equation (18) corresponds to the bare value, and the second to the fluctuational correction. The function F introduced in equation (18), is determined by the correlator

$$F = F_1 + iF_2 = \int \exp(i\omega t) \frac{\langle \psi^2(\mathbf{r}, t)\psi^2(0,0)\rangle}{4T} \,\mathrm{d}t\,\mathrm{d}\mathbf{r}^3, \tag{20}$$

where F_1, F_2 are real and imaginary parts of the correlator, respectively. These functions possess critical peculiarities in the vicinity of the transition. In the hydrodynamic and in the fluctuational regimes they behave as

$$\left.\begin{aligned} &F_1 \sim \tau^{-\alpha}, \quad F_2 \sim \omega \tau^{-(z\nu+\alpha)} \\ \text{and} \quad &F_1 \sim \omega^{-\alpha/z\nu}, \quad F_2 \sim \omega^{-\alpha/z\nu}. \end{aligned}\right\} \tag{21}$$

Here α is the heat capacity index, ν is the correlation length index and z is the dynamic critical index. According to the results of [4, 7] the critical indices possess the values

$$\alpha \sim 0{\cdot}06\text{–}0{\cdot}14, \quad z\nu + \alpha \sim 1{\cdot}06\text{–}1{\cdot}2. \tag{22}$$

We note that the functions F_1, F_2, dependent on the frequency ω and the vicinity parameter τ, take extreme values at the real phase transition point (when $\tau = 0$).

The structure of the elastic matrix, renormalized by the critical fluctuations in the dynamics is analogous to the structure of that in thermodynamics. However, in this case there are considerable differencies. In thermodynamics the correlator (19) is real, but in dynamics it is complex. Its imaginary part determines the critical corrections to the viscosity coefficients. When the applied frequency is zero, the imaginary part of the fluctuational corrections (see equation (19)) disappears. The remaining real part, certainly, reproduces the thermodynamic limit. For example, in the area of advanced fluctuations, when the critical corrections are much larger than the initial moduli g^-, we shall obtain, in the low frequency limit, the following equations for the renormalized compressibilities $\tilde{g}$

$$\tilde{g}_{\rho\rho}, \tilde{g}_{\rho u}, \tilde{g}_{uu} \sim \tau^{-\alpha}. \tag{23}$$

We now consider the temperature–frequency behaviour of the smectic elastic moduli in the vicinity of the transition. The universal behaviour (see equation (22)) takes place only under the infrafrequencies in a very narrow region near the transition. The analysis of the experimental data obtained in this work and in [1, 8] shows that the universal behaviour is realized for frequencies, lower than $10^4\,\mathrm{s}^{-1}$, whereas the common frequency for the ultra-acoustic experiments is about $10^6\,\mathrm{s}^{-1}$. Thus, in a real experimental situation we work in the crossover region. The critical corrections in this area are comparable with the bare values

$$D_\mu D_\nu F \sim g^-_{\mu\nu}. \tag{24}$$

After some lengthy, but not very complicated calculations from equations (18)–(19), we obtain the dynamic elastic moduli of the smectic in the crossover region

$$\tilde{g}^-_{uu} = g^-_{uu} + D_u D_u M, \tag{25}$$

$$\tilde{g}^-_{u\rho} = g^-_{u\rho} + D_u D_\rho M, \tag{26}$$

$$\left.\begin{aligned} &\tilde{g}^-_{\rho\rho} = g^-_{\rho\rho} + D_\rho D_\rho M, \\ &M = -\frac{F_1((D_\mu g_{\mu\nu} D_\nu)^{-1} + F_1) + F_2^2}{((D_\mu g_{\mu\nu} D_\nu)^{-1} + F_1)^2 + F_2^2}(D_\mu g_{\mu\nu} D_\nu)^{-1}. \end{aligned}\right\} \tag{27}$$

These moduli describe the elastic properties of a liquid crystal in the area of advanced fluctuations at frequency ω.

For weak fluctuations the critical corrections are sufficiently small not only when compared with the elastic module $g^-_{\rho\rho}$ but also with the initial value of the smectic layers compressibility module g^-_{uu}

$$FD_u^2 \ll g_{uu}, \quad FD_\rho^2 \ll g_{\rho\rho}. \tag{28}$$

In this case the critical corrections to the elastic moduli take the form

$$\left.\begin{aligned} \delta g^-_{uu} &= -D_u D_u F_1, \\ \delta g^-_{\rho u} &= -D_\rho D_u F_1, \\ \delta g^-_{\rho\rho} &= -D_\rho D_\rho F_1. \end{aligned}\right\} \tag{29}$$

Far from the transition the elastic moduli, naturally, become equal to their initial values (see equation (8)).

We note the following interesting circumstance. At all temperatures the critical corrections to the smectic elastic moduli are connected by the relations

$$\delta g^-_{\rho\rho} : \delta g^-_{\rho u} : \delta g^-_{uu} = \frac{D_\rho^2}{D_u^2} : \frac{D_\rho}{D_u} : 1. \tag{30}$$

Finally, we turn our attention directly to the longitudinal (first) sound spectrum. The successive description of dynamic effects, associated with the order parameter fluctuations, was given in [5]. The system of non-dissipative hydrodynamic equations for the long wavelength variables (the velocity c, the order parameter ψ, the specific entropy σ, the density ρ and the smectic variable u) was obtained with the help of Poisson brackets [6, 9]. Then by a standard procedure some kinetic terms were added to take into account the dissipative processes. From these equations by means of a diagrammatic technique [10, 11] the sound-mode spectrum near the phase transition was obtained. We now write down the results of the calculations. The velocity of the first sound is

$$\rho c^2 = \tilde{g}^-_{\rho\rho} + 2\tilde{g}^-_{u\rho}(\mathbf{q}\mathbf{v})^2/q^2 + \tilde{g}^-_{uu}(\mathbf{q}\mathbf{v})^4/\mathbf{q}^4. \tag{31}$$

Here ρ is the smectic density, $\mathbf{v}$ is the unit vector along the smectic layer normal, $\mathbf{q}$ is the wavevector of propagating sound. The renormalized dynamic elastic moduli $\tilde{g}^-$, dependent on the vicinity of the phase transition τ and the frequency ω, are described by equations (8), (25)–(27) and (29).

3. Experimental and results

In this work the temperature–frequency dependences of ultrasound velocity (c) in the smectic A and C phases of the 4-(n-hexyloxy)phenyl ester of 4-(n-decyloxy)benzoic acid were investigated. The phase behaviour of this compound is [14]

$$\mathrm{S} \xrightarrow{317\,\mathrm{K}} \mathrm{S_C} \xrightarrow{350{\cdot}8\,\mathrm{K}} \mathrm{S_A} \xrightarrow{356{\cdot}7\,\mathrm{K}} \mathrm{N} \xrightarrow{362{\cdot}3\,\mathrm{K}} \mathrm{I}.$$

The measurements were carried out on a sample, cooled from nematic phase under a constant magnetic field of 0·3 T. The angle between the wavevector and the field direction was varied during the measurements. The temperature deviations of the ultrasound velocity $(c-c_0)$ with the respect to the value, c_0, in nematic and isotropic phases, were evaluated during the experiment. The experimental error for $(c-c_0)$ was less than 1 per cent. The measurements of the ultrasound velocity in the frequency range 0·15–1·3 MHz were carried out by the resonator method [2]. In the frequency range 3–27 MHz the measurements were performed by the modified pulse–phase method with an alternating frequency [3].

The c_0 values were estimated at various frequencies by means of the simultaneous numerical treatment of experimental results for the velocity and the absorption coefficient of the ultrasound, obtained by the pulse–phase method with an alternating distance. The procedure for the evaluation of c_0 does not exclude the existence of a systematic error (not more than 0·5 per cent). This error, however, possesses the same value for all the temperatures and frequencies investigated, and it does not influence our conclusions.

The acoustic camera allowed us to realize both the alternating distance method and the alternating frequency method (see figure 1). The basic element of the construction are the cassettes (1) with four pairs of piezo-transformers. One of the cassettes is rigidly attached to the camera body (2). The second is situated axially-symmetrically to the first and can be shifted along the director-rails (3) by means of the wedge (4), which is moved by a micrometer screw. The set, consisting of four pairs of piezo-transformers, allowed us to investigate the acoustic parameters in the 3–27 MHz frequency interval. The electronic part of our experimental set-up allowed us to automate the measuring process and is described in [15].

The temperature dependence of the ultrasound velocity for frequencies 0·3 and 27 MHz and for values of the angle θ of 0°, 30°, 60°, 90° are shown in figures 2 and 3. Here and elsewhere θ is the angle between the layer normal ν and the wavevector direction $\mathbf{q}$. The velocity minimum at the frequency 0·3 MHz in the vicinity of the S_C–S_A phase transition is observed when θ is 0° and 30°. This minimum decreases, with increasing and at 5 MHz practically vanishes. At the same time the phase transition is

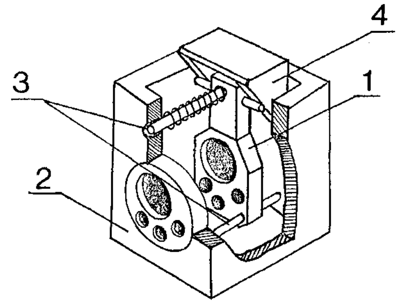

Figure 1. The construction of the acoustic camera.

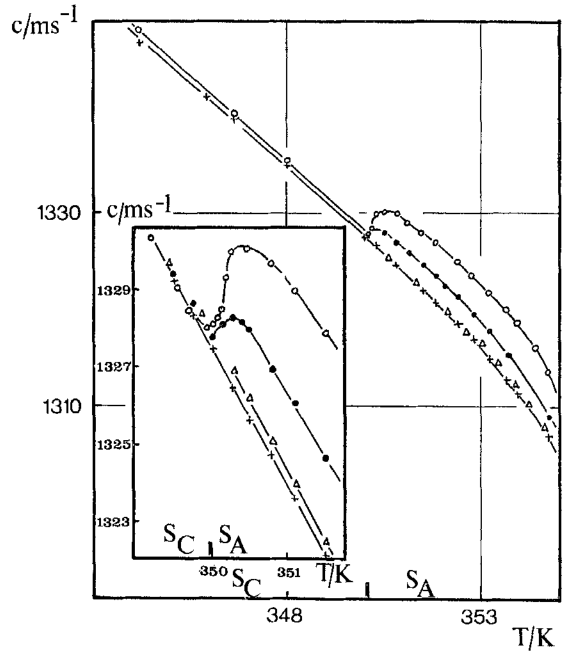

Figure 2. The temperature dependence of the longitudinal sound velocity in smectic C and A phases at 0·3 MHz for different values of the angle θ between the wavevector and the normal to the smectic layers: ○, $\theta=0°$; ●, $\theta=30°$; △, $\theta=60°$; +, $\theta=90°$.

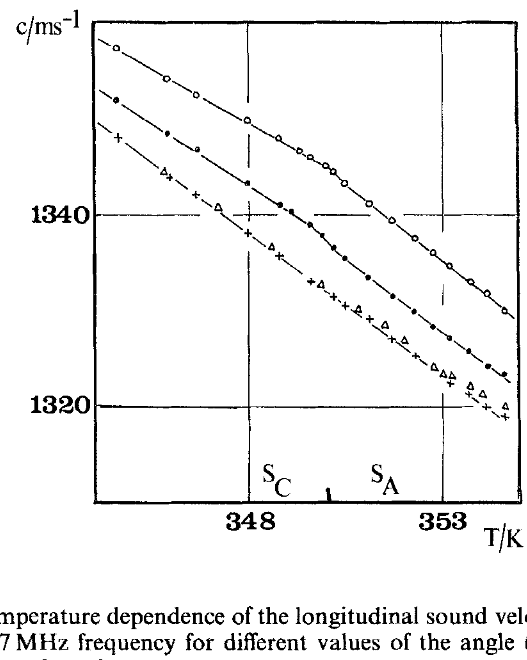

Figure 3. The temperature dependence of the longitudinal sound velocity in smectic C and A phases at 27 MHz frequency for different values of the angle θ: ○, $\theta=0°$; ●, $\theta=30°$; △, $\theta=60°$; +, $\theta=90°$.

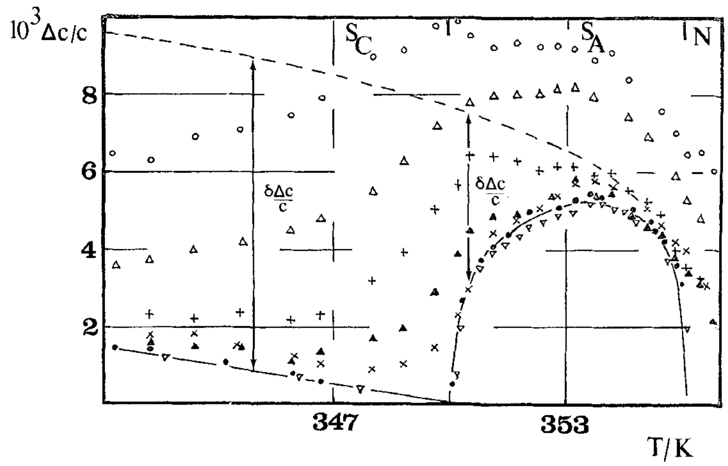

Figure 4. The temperature–frequency dependence of the relative anisotropy of the longitudinal sound veclocity: ▽, 0·15; ●, 0·3; ×, 3; ▲, 5; +, 9; △, 15; ○, 27 MHz. (The dashed lines correspond to the regular character of the ultrasound velocity anisotropy, extrapolated from the smectic A–nematic phase transition region, accordingly to equation (35).)

accompanied by a change in the velocity temperature coefficient (see figure 3). When θ is 60° and 90° we do not observe any significant changes in the character of the ultrasound velocity temperature dependence over the complete frequency range. The exception is the very low frequency regime, where only small changes in the temperature velocity coefficient are visible. Very high velocity sensitivity at $\theta = 0°$ and very low sensitivity at $\theta = 90°$ to the vicinity of the transition demonstrate the monodomain structure of the sample.

At all frequencies the ultrasound velocities when $\theta = 90°$ and 60° were very similar and their difference did not exceed 1 m/s. When θ is 0°, a stronger frequency dependence of the first sound velocity (stronger than for $\theta = 90°$ and for both S_C and S_A phases) was observed. It should be noted that with decreasing frequency the ultrasound anisotropy in the S_C phase diminishes. This dependence practically disappears at low frequencies in the vicinity of the S_{A-N} phase transition. The temperature dependence of the relative anisotropy

$$\Delta c/c = \frac{c(0°) - c(90°)}{c(90°)},$$

is shown in figure 4, for the total frequency range. We note that the frequency dependence of the relative anisotropy $\Delta c/c$ occurs in both smectic phases. At the same time, outside the transition area ($T - T_{S_CS_A} > 3$ K) at frequencies lower then 5 MHz, there is no frequency dependence of this parameter. This shows that the low frequency (hydrodynamic) limit is achieved.

4. Numerical treatment of the experimental results and discussion

Using the frequency-temperature dependences of the first sound velocity (see figures 2 and 3) for four different types of orientation, we can calculate the smectic elastic moduli, occurring in equation (31).

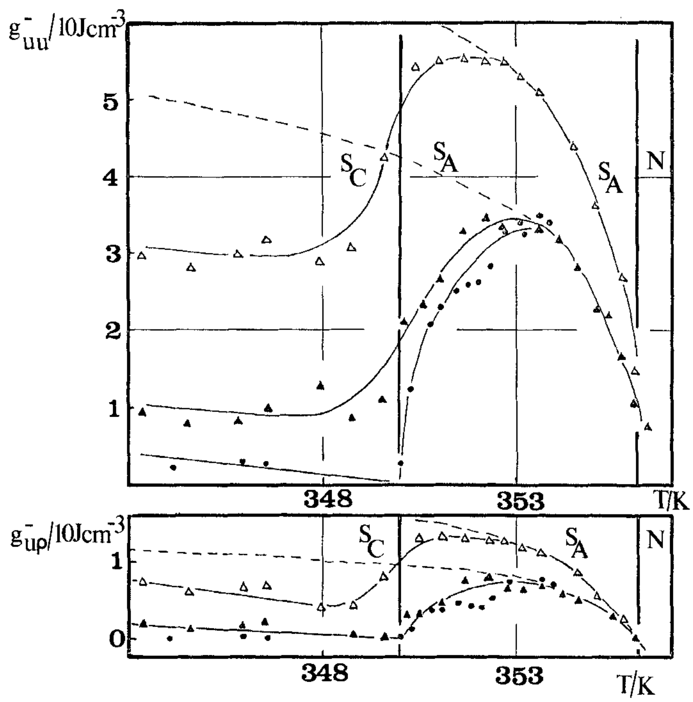

Figure 5. The temperature dependence of the elastic moduli g^-_{uu}, g^-_{pu} in the smectic C and smectic A phases: ●, 0·3; ▲, 3; △, 15 MHz. (The dashed lines correspond to the regular character of the corresponding parameters, extrapolated from the smectic A–nematic transition region, accordingly to equation (35).)

The temperature dependences of the smectic elastic moduli, associated with the smectic layer compressibility, are shown in figure 5. It is obvious that these moduli are approaching zero when heated to the nematic phase at low frequencies. Due to the narrowness of the smectic phase interval, these moduli remain quite small. In the whole frequency interval investigated, they possess the value of an order of 10 J cm^{-3}, which is in agreement with our estimates (see equations (12)–(15)).

The critical behaviour of the elastic modulus g^-_{pp} is shown in figure 6. This elastic modulus is 10^3 J cm^{-3}, which is in agreement with our estimate (11). Thus, in the whole frequency–temperature region, which was investigated, the parameter (12) remained small.

The frequency dispersion of the elastic moduli g^-_{uu} and g^-_{pu}, far from the transition, is, probably, due to of the non-critical molecular dissipation mechanism at high frequencies. Moreoever, in the smectic A phase the frequency behaviour of the elastic moduli is complicated due to the vicinity of the smectic A–nematic transition. The advanced fluctuations areas for both transitions (S_C–S_A, S_A–N) overlap, complicating the analysis of the dispersion properties. Nevertheless, the frequency dependence, as observed from figure 5, vanishes for low frequencies. This indicates that the hydrodynamic limit is achieved.

We can easily calculate, using equation (5), the interlayer distance dependence upon the density (pressure P) in both smectic phases. The results of these calculations are

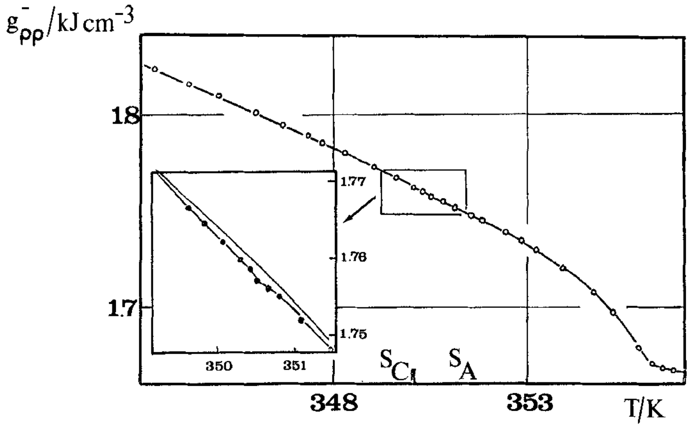

Figure 6. The temperature dependence of the elastic modulus $g_{\rho\rho}^-$ in the critical area at a frequency of 0·15 MHz.

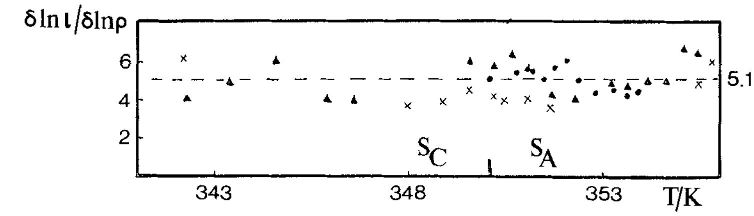

Figure 7. The temperature dependence of the interlayer distance upon the density ρ, calculated from equation (9): ●, 0·3; ×, 3; ▲, 5 MHz.

shown in figure 7. With increasing density the interlayer distance l decreases. (Generally speaking, the situation, when with increasing density the interlayer distance also increases, is not excluded.) In our case in the smectic phases

$$-\frac{\partial(\ln l)}{\partial(\ln \rho)_\sigma} \sim 5, \tag{32}$$

this value was a small temperature dependence.

In the absence of the smectic C–smectic A phase transition, the elastic moduli behaviour would be described by some regular temperature dependences. To determine these dependences (dashed lines in the figures 4 and 5) we have used the experimental data for the anisotropy in the velocity (see figure 4). Making use of the fact, that the parameter (13) is small, not for only the bare g^-, but also for renormalized elastic moduli

$$\frac{\tilde{g}_{uu}^-}{\tilde{g}_{\rho\rho}^-} \sim \frac{\tilde{g}_{u\rho}^-}{\tilde{g}_{\rho\rho}^-} \sim 10^{-2}, \tag{33}$$

we can obtain from equation (31) the proportionality law for the total anisotropy of the velocity

$$\frac{\Delta c}{c} \sim \frac{g_{uu}^- + 2g_{u\rho}^-}{g_{\rho\rho}^-}. \tag{34}$$

In the vicinity of the S_A–N transition the critical behaviour of the elastic moduli g_{uu}^-, $g_{u\rho}^-$ and of the anisotropy of velocity is obeyed with the same law. However, in contrast to the calculated values of the elastic moduli (see figure 5), the total anisotropy of the velocity can be measured more precisely. As we can see from figure 8, with decreasing temperature the velocity anisotropy increases, according to the power law

$$\frac{\Delta c}{c} \sim (T - T_{S_A N})^{-0\cdot 25}. \tag{35}$$

The value of the critical index $-0\cdot25$ is in accord with the experimental result [12, 13].

According to equation (34) the elastic moduli g_{uu}^- and $g_{u\rho}^-$ are described by equation (35) in the vicinity of the S_A–N phase transition. Their regular behaviour in the S_A phase is given by the law (35) extrapolated from the nematic phase (see, the dashed line in figure 5). We stress that the critical behaviour of the velocity anisotropy and the elastic moduli g_{uu}^- and $g_{u\rho}^-$, determined by equation (33), is associated with the S_A–N phase transition.

We come, finally, to the investigation of the critical behaviour of the smectic elastic moduli in the vicinity of the S_C–S_A phase transition. In the area of small fluctuations we can use the results of mean field theory. As we can see from figure 5, the elastic moduli

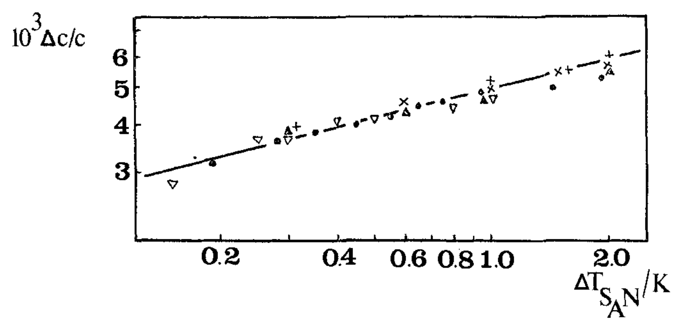

Figure 8. The critical behaviour of the relative anisotropy of the ultrasound velocity in the vicinity of the smectic A–nematic phase transition. (The variable marks are the same as those in figure 4. The unbroken lines correspond to equation (35).)

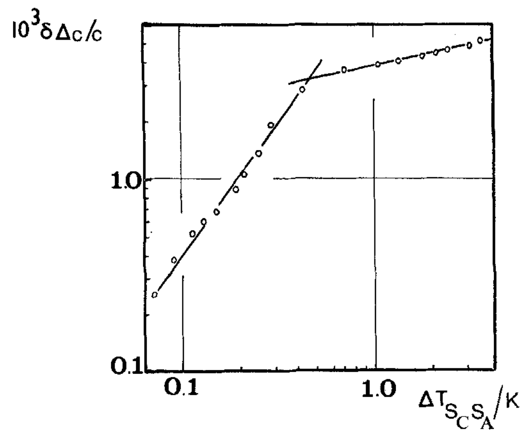

Figure 9. The temperature dependence of the fluctuational correction to the total anisotropy of the ultrasound velocity near the smectic C–smectic A phase transition.

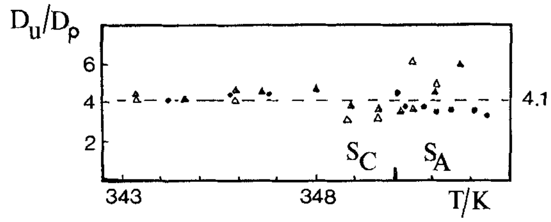

Figure 10. The universal relation of the fluctuational corrections to the critical moduli $g_{\rho u}^{-}$, g_{uu}^{-} in the vicinity of the transition: ●, 0·3; ▲, 5; △, 15 MHz.

g_{uu}^{-} and $g_{u\rho}^{-}$ decrease abruptly at the S_C–S_A transition; this jump is especially clear at low frequencies. The difference between the regular curve (dashed line) and the experimental points for the elastic moduli are of the order of the modulus g_{uu}^{-}. This fact, according to equation (14), confirms our estimates (13) for the numerical values of D and U. Moreover, from the experimental results we can see that the jumps (16) are connected by the relation

$$\frac{\Delta g_{uu}^{-}}{\Delta g_{u\rho}^{-}}=\frac{\Delta g_{u\rho}^{-}}{\Delta g_{\rho\rho}^{-}}=\frac{D_u}{D_\rho}\sim 4. \tag{36}$$

This value for the vector components (see equation (5)) is in agreement with the value (see equations (4) and (5)) obtained in [1]. We recall that the components D_u and D_ρ characterize the contribution to the energy of the interaction of the order parameter fluctuations with the density fluctuations $\delta\rho$ and with the smectic layer shift u. The value in equation (36) proves that the interaction of the order parameter with the density fluctuations is stronger than its interaction with the layer fluctuations.

The fluctuational decrease in the elastic moduli begins (see figures 5–6) in the smectic A phase. Until the corrections are small, they are described by equation (29). With an approach to the phase transition we find ourselves in the area of advanced fluctuations (see equation (24)).

It is interesting to investigate the universal relation (30). This calculated from the value of the critical corrections to the elastic moduli $g_{\rho u}^{-}$, g_{uu}^{-} is shown in figure 10;

$$\frac{\delta g_{uu}^{-}}{\delta g_{u\rho}^{-}}=\frac{\delta g_{u\rho}^{-}}{\delta g_{\rho\rho}^{-}}=\frac{D_u}{D_\rho}. \tag{37}$$

We can see that this value is frequency and temperature independent. Because equation (37) is approximately equal to 4, the critical contribution to the elastic module $g_{\rho\rho}^{-}$ (see figure 6) is one order less than the corresponding contribution to g_{uu}^{-} and $g_{\rho u}^{-}$. In addition, the correction to the $g_{\rho\rho}^{-}$ lies at the border of the experimental accuracy.

5. Conclusion

The analysis shows that the frequency–temperature dependences of the first sound velocity gives the major possibility for investigating the elastic properties of smectics. These properties are determined by three moduli (8), whose behaviour is shown in figures 5 and 6. For the fluctuational corrections to the elastic moduli of a smectic the universal relations (30) and (37) are fulfilled; this fact is proved by figure 10. However, it is impossible to describe the critical correction by means of a simple power law. This is due to the fact that in the temperature–frequency region investigated the corrections are of the same order as the bare values. In this case the smectic elastic moduli, described by equations (25)–(27), do not necessarily possess the minimum in the critical point. The shift of the minimum is a purely dynamic effect, and in the thermodynamic limit it vanishes (see figures 5 and 6). In general, it is impossible to reconstruct the universal function F_1, F_2, using only the single temperature–frequency dependence of the smectic elastic moduli. To constuct these functions we must know the behaviour of the viscosity [1] and the elastic moduli $g_{u\sigma}^-$, $g_{\rho\sigma}^-$ and $g_{\sigma\sigma}^-$. Nevertheless, the low frequency behaviour of the ultrasound velocity anisotropy appears to be informative.

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