Critical absorption of first sound near the smectic *A*–smectic *C* phase transition

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(Submitted 25 November 1988)
Pis'ma Zh. Eksp. Teor. Fiz. **49**, No. 1, 30–33 (10 January 1989)

The absorption of first sound in the smectic *A* phase and smectic *C* phase in the frequency interval 3×10^5 s^{-1}–3×10^7 s^{-1} was studied. The critical behavior of the viscosity coefficients was analyzed. The dynamic critical indices were estimated. The component of the damping of sound associated with the fluctuations of the smectic layers was determined. The results obtained experimentally are compared with the theoretical predictions. {G. F. Mazenko, S. Ramaswamy, and J. Toner, Phys. Rev. **A28**, 1618 (1983); E. I. Kats and V. V. Lebedev, Zh. Eksp. Teor. Fiz. **85**, 2019 (1983) [Sov. Phys. JETP **58**, 1172 (1983)]; E. V. Gurovich, E. I. Kats, and V. V. Lebedev, Zh. Eksp. Teor. Fiz. **94**, 167 (1988) [Sov. Phys. JETP **67**, 741 (1988)]}.

We report here the results of an experimental study of the absorption of first sound in smectic *A* and smectic *C* phases in the frequency interval 3×10^5 s^{-1}–3×10^7 s^{-1} and of the critical behavior of the viscosity near the smectic *A*–smectic *C* phase transition. We have estimated the dynamic critical indices in the hydrodynamic and fluctuation regions and determined the extent to which the fluctuations of smectic layers affect the damping of sound. The results obtained experimentally are compared with theoretical predictions.[1-3]

The smectic liquid crystals are characterized by the presence of a layered structure. In the smectic A phase the mean direction of the major axes of molecules, which is determined by the director $\mathbf{n}$, is the same as that of the unit vector of the normal to the layer, $\vec{\nu}$. In the smectic C phase the director forms an angle with the vector $\vec{\nu}$. Intermolecular forces in this case set only the projection of the director onto the normal to the layer. The vector

$$\vec{\psi} = [\mathbf{n}\cdot\vec{\nu}] \qquad (1)$$

vanishes identically in the A phase and is nonvanishing in the C phase.

The smectic A–smectic C phase transition is a second-order phase transition. The vector (1) is a natural order parameter. For this transition the broad range of developed fluctuations is determined experimentally. Virtually all experimental studies have dealt until recently with the critical thermodynamics of the phenomenon: the heat capacity, susceptibility, and the slope angle of molecules in the smectic C phase were measured as a function of temperature. The dynamic effects associated with the critical fluctuations of smectic layers have, however, received little attention.

In the present letter we report original results of an experimental study of the absorption of longitudinal (first) sound in the smectic A and smectic C phases of n-(hexyloxy) phenyl ether of the n-(decyloxy) benzoic acid. We have obtained for the first time the temperature-frequency curves for the absorption of ultrasound near the smectic A–smectic C phase transition.

The compound we have studied has the following phase-transition scheme:

$$S_3 \xleftarrow{317\text{ K}} S_C \xleftrightarrow{350.8\text{ K}} S_A \xleftrightarrow{356.7\text{ K}} N \xleftrightarrow{362.3\text{ K}} I,$$

where S_3 is the unidentified smectic phase which can be observed only when it is cooled, N is the nematic phase, and I is the isotropic phase.

The sample was oriented as it was cooled from the isotropic phase in a 0.3-T magnetic field. The absorption coefficient α of ultrasound was measured by the resonator method in the frequency range 0.35–1.3 MHz and by the pulse-phase method in the frequency range 3–27.7 MHz. These measurement methods are described in Refs. 4 and 5. The absolute value of α was measured within 10% and the temperature was held constant within 0.01 K.

The results of measurements of the absorption of ultrasound in the perpendicular and longitudinal directions with respect to the plane of the smectic layers are shown in Fig. 1. We see, most notably, that the critical absorption of sound depends strongly on the angle between the wave vector and the normal to the layer, $\vec{\nu}$. The universal models, which are routinely used in the theory of critical phenomena and in which the order parameter is specified in the "isotopic" space, reveal that the isotropic component of the absorption of ultrasound comes from the critical fluctuations.

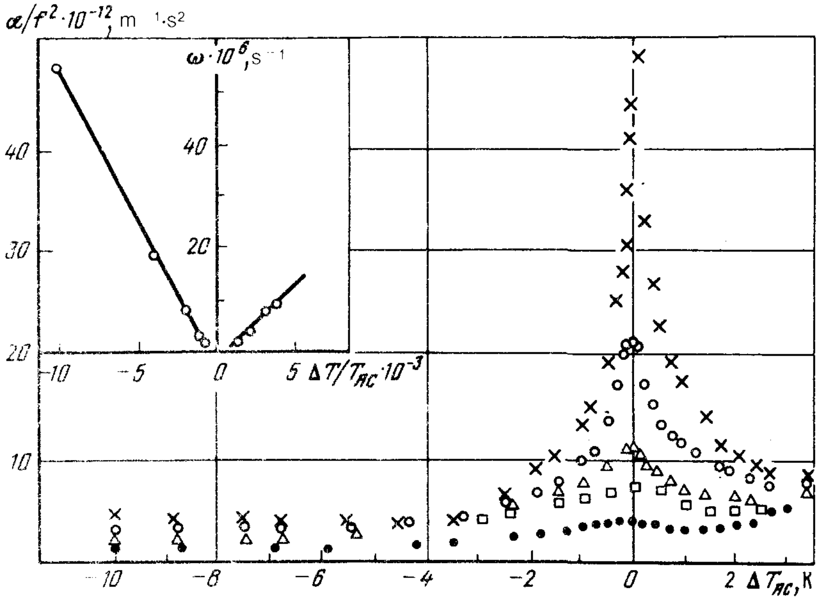

FIG. 1. Temperature dependence of the absorption coefficient of ultrasound: for waves propagating along the normal to the smectic layers ($\theta = 0°$) ×—0.36 MHz; ○—0.69 MHz; △—1.2 MHz; □—3 MHz and for waves propagating perpendicular to the normal to the smectic layers ($\theta = 90°$) ●—0.36 MHz.

Since the physical properties of the smectic phases are strongly anisotropic, the universal models are inapplicable to a smectic *A*–smectic *C* phase transition. The orientational dependence of the critical absorption of first sound clearly demonstrates this fact.

In the critical dynamics of the smectic *A*–smectic *C* phase transition constructed in Ref. 3 the order parameter (1) was prescribed in real space rather than isotopic space. The critical fluctuations of such an order parameter lead to anisotropic corrections in the dilatational-viscosity coefficients:

$$\delta\eta_2 = D_\rho^2\,\mathcal{K}'(\omega,\tau)$$
$$\delta\eta_5 = (D_\rho + D_u)^2\,\mathcal{K}'(\omega,\tau). \qquad (2)$$

The notation for the viscosity coefficients was taken from Ref. 6. The viscosity coefficients η_2 and η_5 determine the absorption of ultrasound which propagates respectively perpendicular to ($\theta = 90°$) and parallel to ($\theta = 0$) the normal to the plane of the smectic layer. The quantities D_ρ and D_u, which have the dimensionality of the energy density, characterize the contribution to the free energy from the interaction of the order parameter (1) with respectively the density fluctuations and the deviations of the smectic layers from the equilibrium position. The dimensionless parameter is $\tau = \Delta T_{AC}/T_{AC}$, where $\Delta T_{AC} = T - T_{AC}$, and T_{AC} is the temperature of the smectic *A*–smectic *C* phase transition.

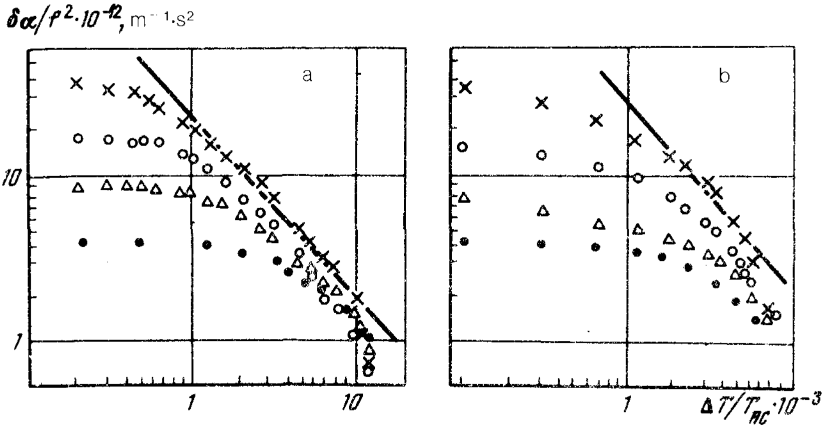

FIG. 2. Temperature dependence of the critical part of the absorption coefficient of ultrasound $\delta\alpha/f^2 \sim \eta_5$ in (a) the smectic C phase and (b) the smectic A phase. ×—0.36 MHz; ○—0.46 MHz; △—1.2 MHz; ●—3 MHz.

Using the experimental results of Fig. 1, we calculated the ratio of the critical corrections to the viscosity coefficients η_5 and η_2. We found that $\delta\eta_5/\delta\eta_2 \approx 30$ and that this ratio does not depend on τ or ω. We have thus shown experimentally that the critical fluctuations of the order parameter interact with the fluctuations of the smectic layers more strongly than they do with the density fluctuations. A qualitative manifestation of this situation is the fact that $D_u > D_\rho$.

Gurovich *et al.*[3] showed that near the phase transition which we are investigating the quantity $\mathscr{H}'(\omega,\tau)$ behaves critically in the hydrodynamic region, $\mathscr{H}' \sim \tau^{-x}$, and in the fluctuation region, $\mathscr{H}' \sim \omega^{-y}$, where $x \approx y \approx 1.1$. Analysis of the temperature-

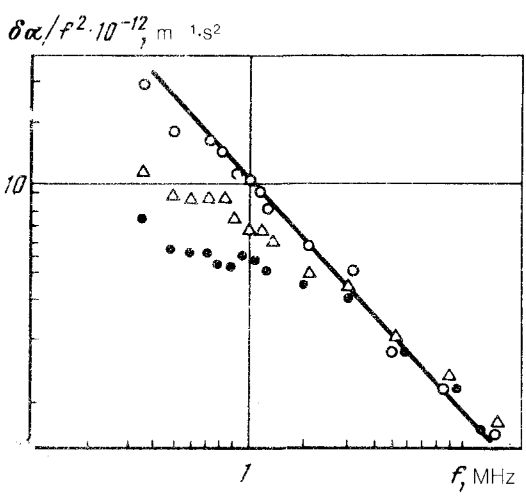

FIG. 3. Frequency dependence of the critical part of the absorption coefficient α/f^2 of ultrasound in the smectic C phase. ○—$\Delta T_{AC} = -0.2$ K; △—$\Delta T_{AC} = -0.5$ K; ●—$T = 2 - 1.2$ K.

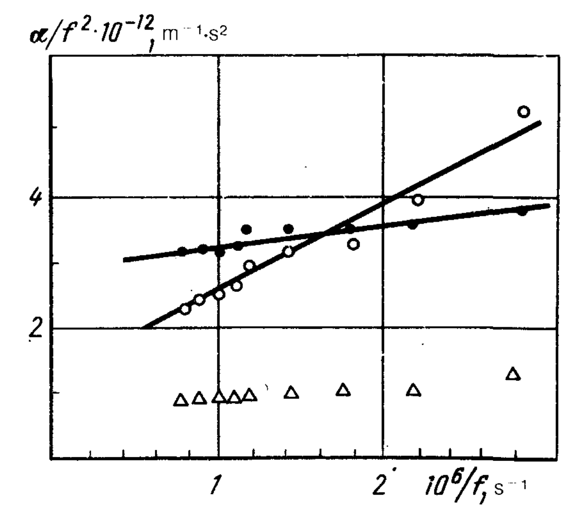

FIG. 4. α/f^2 vs the reciprocal frequency in the smectic C phase. $\theta = 0°$: O—$\Delta T_{AC} = -5.6$ K; ●—$\Delta T_{AC} = -13.5$ K. $\theta = 90°$: Δ—$\Delta T_{AC} = -5.6$ K.

frequency dependence of the critical component of the absorption of ultrasound $\delta\alpha(\theta = 0°)$ in the coordinates shown in Fig. 2 (a and b) and Fig. 3 has made it possible to determine the boundaries of the hydrodynamic and fluctuation regions (see the inset in Fig. 1). The slope of the lines in Figs. 2 and 3 corresponds to $x = y = 1.1$, which was theoretically predicted by Gurovich *et al.*[3]

The frequency dependence of the absorption of ultrasound, which is governed near the phase transition by the critical fluctuations, is also applicable outside the region of the phase transition (Fig. 1), because fluctuations of the smectic layers lead to a highly nontrivial contribution to the dilatational-viscosity coefficients[3]

$$\delta\eta_{2.5} \sim B^{3/2}/\omega , \tag{3}$$

where B is the bulk modulus of the smectic layers, which decreases as the smectic A–smectic C phase-transition point is approached.[3]

Figure 4 is a plot of the dilatational-viscosity coefficient, $\eta_5 \sim \alpha(0)/f^2$, as a function of the reciprocal frequency. We see that this plot is in good agreement with expression (3). A decrease in the slope of the lines as the smectic A–smectic C phase-transition point is approached corresponds to a decrease of the modulus B as a result of critical fluctuations of the order parameter.

We wish to thank V. V. Lebedev and E. I. Kats for a discussion and for valuable remarks made in connection with the analysis of the experimental results.

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Translated by S. J. Amoretty