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Propagation and attenuation of sound near the smectic-*A* — smectic-*C* phase transition in liquid crystals

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Anomalous contributions to the sound attenuation on the smectic-*A* side of the smectic-*A* — smectic-*C* phase transition are found by generalizing the Landau-Ginzburg theory. Terms coupling the smectic-*C* order parameter to the density and to the gradient of the layer spacing are incorporated and are found, to second order near the transition, to enhance certain of the viscosities which determine the acoustic attenuation. The anomalous attenuation depends on the angle between the propagation direction and layer normal, with the degree of anisotropy governed by the sensitivity of the transition temperature to changes in the layer spacing for constant density. The dispersion in the sound velocity near this transition is also considered.

I. INTRODUCTION

Recently, Bhattacharya, Cheng, Sarma, and Ketterson[1] have experimentally investigated the behavior of sound attentuation and dispersion in a liquid crystal near the smectic-*A* — smectic-*C* (*AC*) phase transition. This work has motivated our theoretical study of the acoustic attenuation and dispersion near this transition.

In their earlier experiments near the nematic — smectic-*A* transition, Bhattacharya and co-workers[2] found the attentuation anomaly to be dominated by an *isotropic* contribution, even though the symmetry of the high-temperature state was uniaxial. This was theoretically explained[3,4] by calculations based on a coupling between the order parameter and the density, a scalar hydrodynamic variable, in the nematic phase.

In the smectic-*A* phase *two* hydrodynamic variables, the gradient of the layer spacing as well as the density, enter the sound wave. We allow for couplings between order-parameter fluctuations and density *and* between order-parameter fluctuations and the gradient of the layer spacing. Owing to the coupling to this extra variable, our results imply that the anomaly in the attentuation on the smectic-*A* side of the *AC* transition is, in general, *anisotropic*. The degree of anisotropy depends on the coupling constants between the order parameter and hydrodynamic variables.

In Sec. II we discuss the order parameter for the *AC* transition and the appropriate Landau-Ginzburg free energy. In Sec. III we write down Langevin equations of motion for the order parameter and the hydrodynamic variables. In Sec. IV a mode-coupling solution for the enhancement of the viscosities which give the major contribution to the attenuation is presented. The attenuation for propagation parallel and perpendicular to the layer normal in terms of these viscosities is derived in an appendix. Finally, in Sec. V we summarize and discuss our results for the acoustic attenuation and dispersion in the smectic-*A* phase as the smectic-*C* phase is approached.

II. GENERALIZED LANDAU-GINZGURG THEORY

de Gennes[5] has noted the similarity between the *AC* transition and the superfluid transition in ^{4}He and has proposed a complex order parameter $\psi = \sin\theta e^{i\phi}$ to describe the *AC* transition, where θ is the angle between the director and the normal to the smectic layers and ϕ is the azimuthal angle describing the orientation of the tilted director. Based on a generalized Maier-Saupe[6] theory we find the magnitude of the order parameter to be $\sin\theta\cos\theta$ (rather than $\sin\theta$). We choose to write the smectic-*C* order parameter as $\psi = Q_{13} + iQ_{23}$, where $Q_{\alpha\beta}$ is the symmetric, traceless, second-rank tensor used to describe the nematic liquid crystal,[7] but here the director makes an angle with the 3-direction, the 3-direction being defined by the normal to the smectic layers. The details of the functional dependence of ψ on the polar angle are of no consequence in the discussion which follows. The fluctuations of the smectic-*C* order parameter contribute via an anisotropic Landau-Ginzburg free energy:

$$F_\psi=\tfrac{1}{2}\int d^3r[a\,|\psi|^2+(2m_\perp)^{-1}|\vec{\nabla}_\perp\psi|^2+(2m_3)^{-1}|\nabla_3\psi|^2+(b/2)|\psi|^4], \quad (1)$$

where $\perp$ refers to the plane of the layers, $m_\perp$, m_3, and b are positive constants, and $a\sim T-T_c$, T_c being the mean-field theory transition temperature.

A second contribution to the free energy comes from the elastic properties of the smectic-A liquid crystal[5,7]:

$$F_{\rm el}=\int d^3r[\tfrac{1}{2}(\partial p/\partial\rho)^0_{\nabla_3 u}(\Delta\rho)^2+\tfrac{1}{2}(\partial\phi_3/\partial\nabla_3 u)^0_\rho(\nabla_3 u)^2+(\partial p/\partial\nabla_3 u)^0_\rho(\Delta\rho)(\nabla_3 u)+\tfrac{1}{2}K_1^0(\nabla_1^2 u+\nabla_2^2 u)^2], \quad (2)$$

where $\Delta\rho$ is the deviation of the density from its equilibrium value, u is the displacement in the 3-direction of the smectic layers from their equilibrium position, K_1^0 is the Frank elastic constant, p is the pressure, and ϕ_3 is the conjugate field associated with $\nabla_3 u$. The thermodynamic derivatives are taken at constant entropy here and throughout the paper. The equilibrium density ρ has been set equal to unity. The superscript zero indicates the bare value of the particular quantity.

A kinetic-energy contribution arises due to the velocity of the center of mass $\vec{v}$,

$$F_v=\tfrac{1}{2}\int d^3r\,|\vec{v}|^2 . \quad (3)$$

Finally, we introduce terms in the free energy which couple the density and layer-spacing fluctuations to the smectic-C order parameter. These are

$$F_{\psi\rho}=(\gamma_\rho/2)\int d^3r\,\Delta\rho\,|\psi|^2 \quad (4a)$$

and[8]

$$F_{\psi u}=(\gamma_u/2)\int d^3r\,\nabla_3 u\,|\psi|^2 . \quad (4b)$$

Near the AC transition then, the consequential contributions to the free energy are given by

$$F=F_\psi+F_{\rm el}+F_v+F_{\psi\rho}+F_{\psi u} . \quad (5)$$

III. EQUATIONS OF MOTION

The dispersion relation describing propagation and attenuation of sound results from the solution of the set of equations of motion for the hydrodynamic variables characterizing the system. Details of this dispersion relation in the smectic-A phase have been discussed by Martin, Parody, and Pershan[9] (MPP) and by Miyano and Ketterson.[10] As the AC transition is neared, fluctuations of the smectic-C order parameter become important and renormalize the dissipative and elastic coefficients occurring in the hydrodynamic equations of motion. We allow for these fluctuations by writing down equations of motion for density, layer displacement, velocity, and order parameter in the standard Langevin form.[11]

For the real and imaginary parts of ψ, i.e., Q_{13} and Q_{23}, we have

$$\partial Q_{\alpha 3}(\vec{r},t)/\partial t=\mu_{kj\alpha}(\vec{r},t)\nabla_j v_k(\vec{r},t)-(1/\gamma^0)\delta F/\delta Q_{\alpha 3}(\vec{r},t)+\sigma_\alpha(\vec{r},t)/\gamma^0 , \quad (6a)$$

where the Greek indices take on values 1 and 2, the Cartesian indices take on values 1, 2, and 3, and repeated indices are summed over.

For u,

$$\partial u(\vec{r},t)/\partial t=v_3(\vec{r},t)+\zeta^0\nabla_3(\delta F/\delta\nabla_3 u(\vec{r},t))+\theta(\vec{r},t) . \quad (6b)$$

For $\Delta\rho$,

$$\partial\Delta\rho/\partial t+\nabla_i v_i=0 . \quad (6c)$$

For $\vec{v}$,

$$\partial v_i(\vec{r},t)/\partial t=-\nabla_i(\delta F/\delta\Delta\rho(\vec{r},t))+\mu_{ij\alpha}(\vec{r},t)\nabla_j(\delta F/\delta Q_{\alpha 3}(\vec{r},t))-\nabla_j\sigma^D_{ij}(\vec{r},t)+\delta_{i,3}\nabla_3(\delta F/\partial\nabla_3 u(\vec{r},t))+\zeta_i(\vec{r},t) . \quad (6d)$$

μ_{ija} is given formally by the Poisson bracket of velocity with the microscopic analog of $Q_{\alpha 3}$. Its form can be determined phenomenologically by imposing the constraints of the smectic-A symmetry. For the purposes of this paper the explicit form of $\mu_{ij\alpha}$ is not needed.

In Eq. (6d) σ_{ij}^D, the dissipative part of the stress tensor, also has the symmetry of the smectic-A phase:

$$\sigma_{ij}^D = -\eta_{ijkl}^0 \nabla_l (\delta F/\delta v_k) , \tag{7}$$

where η_{ijkl}^0 is the bare viscosity matrix. We use the notation of MPP for the viscosities. The viscosities are given in their equation (3.5).

In (6) σ_α, θ, ζ_i, the noise sources, are related via the fluctuation-dissipation theorem to γ^0, the bare coefficient describing the rate of relaxation of $Q_{\alpha 3}$, ζ^0, the bare transport coefficient associated with u, and η_{ijkl}^0, respectively, in the usual way.[11]

IV. MODE COUPLING CALCULATION

We find the order-parameter fluctuation contributions to the viscosities and elastic constants by means of the usual mode-coupling method.[12,13] We are looking for the largest contribution to the sound attenuation in this paper. In the calculation[4] of sound damping near the nematic–smectic-A transition, this arose from a dissipative coupling of the square of the order parameter to the density in the free energy. Other terms, such as the reactive coupling of the order parameter to the velocity in Eq. (6a), gave a smaller contribution to the sound attenuation.[14] Therefore, in this paper, we concentrate on the coupling of the density and the gradient of the layer spacing to the order parameter as described in (4a) and (4b). The contributions to the viscosities due to these terms will be found by perturbation theory in γ_ρ and γ_u, correct to second order. The renormalized, frequency-dependent viscosities then determine the anomalous acoustic attenuation and dispersion near the phase transition.

Equations (6a)–(6d) are best solved in Fourier space. The derivatives of the free energy can then be easily found, e.g.,

$$\delta F/\delta Q_{\alpha 3}(-\vec{k},-\omega) = \chi^{-1}(\vec{k}) Q_{\alpha 3}(\vec{k},\omega) + \gamma_\rho \int_{q,\omega'} \Delta\rho(\vec{q},\omega') Q_{\alpha 3}(\vec{k}-\vec{q},\omega-\omega') + i\gamma_u \int_{q,\omega'} q_3 u(\vec{q},\omega') Q_{\alpha 3}(\vec{k}-\vec{q},\omega-\omega') , \tag{8}$$

where

$$\chi^{-1}(\vec{q}) = \gamma^0 \Gamma(\vec{q}) = a + q_\perp^2/2m_\perp + q_3^2/2m_3 \tag{9}$$

and

$$\int_{q,\omega'} \equiv \int \frac{d^3 q}{(2\pi)^3} \int \frac{d\omega'}{2\pi} .$$

An additional term, of order bQ^3, has not been written explicitly. The formal solutions for (6a)–(6c) are then

$$Q_{\alpha 3}(\vec{k},\omega) = G_Q^0(\vec{k},\omega) \Big[\eta_\alpha(\vec{k},\omega)/\gamma^0 + i \int_{q,\omega'} q_j \mu_{kj\alpha}(\vec{k}-\vec{q},\omega-\omega') v_k(\vec{q},\omega') - (\gamma_\rho/\gamma^0) \int_{q,\omega'} \int_{q,\omega'} \Delta\rho(\vec{q},\omega') Q_{\alpha 3}(\vec{k}-\vec{q},\omega-\omega') - (i\gamma_u/\gamma^0) \int_{q,\omega'} q_3 u(\vec{q},\omega') Q_{\alpha 3}(\vec{k}-\vec{q},\omega-\omega') \Big] , \tag{10a}$$

$$u(\vec{k},\omega) = G_u^0(\vec{k},\omega) \Big[\theta(\vec{k},\omega)/\gamma_1^0 + v_3(\vec{k},\omega) + (i/\gamma_1^0) k_3 (\partial p/\partial \nabla_3 u)_\rho^0 \Delta\rho(\vec{k},\omega) + (i/2)(\gamma_u/\gamma_1^0) k_3 \int_{q,\omega'} Q_{\alpha 3}(\vec{q},\omega') Q_{\alpha 3}(\vec{k}-\vec{q},\omega-\omega') \Big] , \tag{10b}$$

$$\Delta\rho(\vec{k},\omega) = k_i v_i(\vec{k},\omega)/\omega , \tag{10c}$$

and Eq. (6d) written out in more detail is

$$
\begin{aligned}
-i\omega v_i(\vec{k},\omega) = {} & \zeta_i(\vec{k},\omega) - (i/2)k_i\gamma_\rho \int_{q,\omega'} Q_{\alpha 3}(\vec{q},\omega')Q_{\alpha 3}(\vec{k}-\vec{q},\omega-\omega') \\
& - ik_i(\partial p/\partial\rho)^0_{\nabla_3 u}\Delta\rho(\vec{k},\omega) + k_i k_3(\partial p/\partial\nabla_3 u)^0_\rho u(\vec{k},\omega) \\
& + i\int_{q,\omega'} q_j\mu_{ij\alpha}(\vec{k}-\vec{q},\omega-\omega')[\chi^{-1}(\vec{q})Q_{\alpha 3}(\vec{q},\omega') + \gamma_\rho\int_{q',\omega''}\Delta\rho(\vec{q}\,',\omega)Q_{\alpha 3}(\vec{q}-\vec{q}\,',\omega'-\omega'') \\
& \qquad + i\gamma_u\int_{q',\omega''} q_3' u(\vec{q}\,',\omega)Q_{\alpha 3}(\vec{q}-\vec{q}\,',\omega'-\omega'')] \\
& - ik_j\sigma^D_{ij}(\vec{k},\omega) + \delta_{i,3}\Big[-\gamma_1^0 W(\vec{k})u(\vec{k},\omega) + ik_3(\partial p/\partial\nabla_3 u)^0_\rho\Delta\rho(\vec{k},\omega) \\
& \qquad + (i/2)k_3\gamma_u\int_{q,\omega'} Q_{\alpha 3}(\vec{q},\omega')Q_{\alpha 3}(\vec{k}-\vec{q},\omega-\omega')\Big] ,
\end{aligned}
\tag{10d}
$$

where

$$G^0_Q(\vec{k},\omega) = [-i\omega + \chi^{-1}(\vec{k})/\gamma^0]^{-1} = [-i\omega + \Gamma(\vec{k})]^{-1} , \tag{11}$$

$$G^0_u(\vec{k},\omega) = [-i\omega + W(\vec{k})]^{-1} , \tag{12}$$

$$W(\vec{k}) = (1/\gamma_1^0)[k_3^2(\partial\phi_3/\partial\nabla_3 u)^0_\rho + K_1^0(k_1^2+k_2^2)^2] , \tag{13}$$

and repeated indices are summed over. Equation (10) can be solved to the desired order in γ_ρ and γ_u (or other couplings) by iteration. We substitute the formal solution (10a) for $Q_{\alpha 3}$ for the factors of $Q_{\alpha 3}$ multiplying γ_ρ and γ_u in (10d). We isolate those terms of second order in γ_ρ amd γ_u. The $Q_{\alpha 3}$'s are then replaced by their zeroth-order expressions and the noise source can be averaged using

$$\langle \eta_i(\vec{k},\omega)\eta_j(\vec{q},\omega')\rangle = 2\gamma^0(2\pi)^4\delta(\vec{k}+\vec{q})\delta(\omega+\omega')\delta_{i,j} . \tag{14}$$

This yields for the equation of motion for the average of v_i

$$
\begin{aligned}
-i\omega\langle v_i(\vec{k},\omega)\rangle = (1/\gamma^0)\int \frac{d^3q}{(2\pi)^3} & \{[\chi(\vec{q})+\chi(\vec{k}-\vec{q})]/[-i\omega+\Gamma(\vec{q})+\Gamma(\vec{k}-\vec{q})]\} \\
& \times\{ ik_i\gamma_\rho[\gamma_\rho\langle\Delta\rho(\vec{k},\omega)\rangle + ik_3\gamma_u\langle u(\vec{k},\omega)\rangle] \\
& \quad - \delta_{i,3} ik_3\gamma_u[\gamma_\rho\langle\Delta\rho(\vec{k},\omega)\rangle + ik_3\gamma_u\langle u(\vec{k},\omega)\rangle]\} + (\text{other terms}) ,
\end{aligned}
\tag{15}
$$

where "other terms" represent bare terms and other renormalizations not expected to give contributions to the sound attenuation as large as those we have retained. We now take the limit of small wave vectors, $\vec{k}\to 0$, and separate the integral over wave vectors $\vec{q}$ into its real and imaginary parts. We eliminate $\langle\Delta\rho(\vec{k},\omega)\rangle$ via (10c) and since we are working with small wave vectors we may neglect the disspative terms in the equation of motion for $\langle u(\vec{k},\omega)\rangle$ and eliminate $-i\omega\langle u(\vec{k},\omega)\rangle$ via

$$-i\omega\langle u(\vec{k},\omega)\rangle = \langle v_3(\vec{k},\omega)\rangle . \tag{16}$$

We then have (writing out the bare terms this time)

$$
\begin{aligned}
-i\omega\langle v_i(\vec{k},\omega)\rangle = {} & -ik_\alpha k_i c^0_{\alpha i} v_i/\omega - ik_i k_3 c^0_{i3} v_3/\omega \\
& - k_j\{\eta_2^0(k_i v_j + k_j v_i) + (\eta_3^0-\eta_2^0)[\delta_{j,3}(k_i v_3 + k_3 v_i) + \delta_{i,3}(k_j v_3 + k_3 v_j)] \\
& \quad + (\eta_4^0-\eta_2^0)\delta_{i,j}k_l v_l + (\eta_1^0+\eta_2^0-4\eta_3^0-2\eta_5^0+\eta_4^0)\delta_{i,3}\delta_{j,3}k_3 v_3 \\
& \quad + (\eta_5^0-\eta_4^0+\eta_2^0)(\delta_{i,j}k_3 v_3 + \delta_{i,3}\delta_{j,3}k_1 v_1)\} \\
& + (I_1+iI_2)[ik_i\gamma_\rho^2 k_l v_l/\omega - ik_i k_3\gamma_\rho\gamma_u v_3/\omega + \delta_{i,3}(k_3^2\gamma_u^2 v_3/\omega - k_3\gamma_\rho\gamma_u k_l v_l/\omega)] \\
& + (\text{higher-order terms}) ,
\end{aligned}
\tag{17}
$$

where

$$c_{11}^0=c_{12}^0=c_{22}^0=(\partial p/\partial\rho)_{\nabla_3 u}^0\ , \tag{18a}$$

$$c_{13}^0=c_{23}^0=(\partial p/\partial\rho)_{\nabla_3 u}^0-(\partial p/\partial\nabla_3 u)_\rho^0\ , \tag{18b}$$

$$c_{33}^0=(\partial p/\partial\rho)_{\nabla_3 u}^0-2(\partial p/\partial\nabla_3 u)_\rho^0+(\partial\phi_3/\partial\nabla_3 u)_\rho^0\ , \tag{18c}$$

and the MPP convention for the η's is used. (See Appendix A of Ref. 10.) Also

$$I_1=(2/\gamma^0)\int\frac{d^3q}{(2\pi)^3}\chi(\vec{q})/[\omega^2+4\Gamma^2(\vec{q})]\ , \tag{19a}$$

$$I_2=[4/(\gamma^0)^2]\int\frac{d^3q}{(2\pi)^3}1/[\omega^2+4\Gamma^2(\vec{q})]\ . \tag{19b}$$

Equation (17) then gives, by inspection,

$$\eta_1=\eta_1^0+(\gamma_\rho-\gamma_u)^2I_1\ , \tag{20a}$$

$$\eta_2=\eta_2^0\ , \tag{20b}$$

$$\eta_3=\eta_3^0\ , \tag{20c}$$

$$\eta_4=\eta_4^0+\gamma_\rho^2I_1\ , \tag{20d}$$

$$\eta_5=\eta_5^0+\gamma_\rho(\gamma_\rho-\gamma_u)I_1\ , \tag{20e}$$

where the η's are the true viscosities. For the contributions to the reactive parameters that determine the dispersion we find

$$(\partial p/\partial\rho)_{\nabla_3 u}=(\partial p/\partial\rho)_{\nabla_3 u}^0-\gamma_\rho^2I_2\ , \tag{21a}$$

$$(\partial p/\partial\nabla_3 u)_\rho=(\partial p/\partial\nabla_3 u)_\rho^0-\gamma_\rho\gamma_uI_2\ , \tag{21b}$$

$$(\partial\phi_3/\partial\nabla_3 u)_\rho-(\partial\phi_3/\partial\nabla_3 u)_\rho^0-\gamma_u^2I_2\ . \tag{21c}$$

In order to predict the size of the anisotropy in the speed or attenuation of sound we need more information about the bare coupling constants γ_ρ and γ_u. We eliminate these in terms of the true coupling constants by using the fact that $(\partial F/\partial\rho)_{\nabla_3 u}=p$ and $(\partial F/\partial\nabla_3 u)_\rho=\phi_3$ and expanding out these relations to second order. We obtain for γ_ρ and γ_u

$$\gamma_\rho=\chi^{-2}(\vec{q}[\partial\chi(\vec{q})/\partial\rho]_{\nabla_3 u}\ , \tag{22a}$$

$$\gamma_u=\chi^{-2}(\vec{q})[\partial\chi(\vec{q})/\partial\nabla_3 u]_\rho\ . \tag{22b}$$

If we substitute (22a) into (20d), for example, we finally obtain for η_4

$$\eta_4=\eta_4^0+2\int\frac{d^3q}{(2\pi)^3}\{\chi^{-1}(\vec{q})[\partial\chi(\vec{q})/\partial\rho]_{\nabla_3 u}\}^2\Gamma(\vec{q})/[\omega^2+4\Gamma^2(\vec{q})]\ , \tag{23}$$

where we used $\Gamma(\vec{q})=[\gamma^0\chi(\vec{q})]^{-1}$. In Eq. (23) $\chi(\vec{q})$ and $\Gamma(\vec{q})$ are the true, remormalized expressions. Similar expression hold for the anomalous parts of η_1 and η_5 and the contributions to the elastic constants in Eqs. (21a)–(21c).

V. RESULTS AND DISCUSSION

The critical behavior of the viscosities and dispersion, which in turn determine the behavior of the sound attenuation and velocity, is contained in the integrals I_1 and I_2 of Eq. (19). The dependence of the viscosities and dispersions on the distance from the critical temperature and on frequency can be found from scaling arguments in two regimes of interest.[4,12] The dynamic critical exponent which describes the relaxation rate of the order parameter is z:

$$\Gamma(\vec{k}=0)\sim\xi^{-z}\ , \tag{24}$$

where ξ is the correlation length. We follow here de Gennes's analogy between the *AC* transition and the superfluid ^{4}He transition.[15] Defining the reduced temperature, $t=(T-T_c)/T_c$, we can write the scaling behavior of ξ for $\omega<\Gamma(\vec{k}=0)$ as $\xi\sim t^{-\nu}$ above the transition. Applying the usual scaling techniques to (14) and using the scaling relation for the specific-heat exponent $\alpha=2-\nu d$, we find, for $\omega=0$,

$$\eta_1\sim\eta_4\sim\eta_5\sim t^{-\alpha}\xi^z\ . \tag{25}$$

The relationships between the elastic constants of Eq. (21) and the velocities of sound for propagation directions perpendicular or parallel to the normal to the smectic layers are given in Eq. (A6) and (A11) of the Appendix. Using these relations in conjunction with the critical elastic constants [Eqs. (21) and (19b)] we can write, for small ω, a scaling relation for ΔV^2, the deviation from the $\omega\doteq0$ result for the correction to the square of the sound velocity $[V^2=(V^0)^2-\Delta V^2]$. For either propagation direction we find

$$\Delta V_\Theta^2(\omega)-\Delta V_\Theta^2(\omega=0)\sim -t^{-\alpha}(\omega\xi^z)^2\ . \tag{26}$$

For $\omega>\Gamma(\vec{k}=0)$ the frequency dependence dominates the scaling and temperature dependence enters only through the derivative of a with respect to ρ or $\nabla_3 u$. Using Kawasaki's scaling assumptions[12] in this regime we find

$$\eta_1 \sim \eta_4 \sim \eta_5 \sim t^{-2\alpha}\omega^{-1+\alpha/z\nu} . \tag{27}$$

In both frequency regimes we find the behavior of the viscosities and velocity corrections to be singular as T approaches T_c.

We now want to use the viscosities we have found to determine anomalies in the attenuation of sound and the sound velocity. The solution of the eigenvalue problem for these quantities for a general angle Θ between the sound propagation direction and layer normal is quite complicated.[9,10] (The angle Θ should not be confused with the angle θ which enters the order parameter). To exhibit the features of our result, i.e., anisotropy in the sound damping, we present the answers for two special angles, $\Theta=0°$ and $\Theta=90°$. The solution for the hydrodynamic modes for these angles is presented in the Appendix. In discussing the sound damping we take into account only the anomalous terms in the viscosities which we have calculated in Sec. IV.

The anomalous attentuation coefficient for $\Theta=90°$, $\alpha_{90°}$, is then, from (A13) and (A11),

$$\alpha_{90°} = \tfrac{1}{2}\eta_4 k^2[(\partial p/\partial \rho)_{\nabla_3 u}]^{-1/2}$$
$$= \tfrac{1}{2}\eta_4 k^2/V_{90°} , \tag{28}$$

where η_4 is given by (23).

For frequencies less than $\Gamma(\vec{q}=0)$ we may use the small wave-vector form of $\chi(\vec{q})$ which is given in (9). Assuming the primary dependence of $\chi(\vec{q})$ on density and layer displacement is in the transition temperature, i.e., in the parameter a of Eq. (9), we can write

$$\gamma_\rho = -(\partial a/\partial \rho)_{\nabla_3 u} , \tag{29a}$$

$$\gamma_u = -(\partial a/\partial \nabla_3 u)_\rho , \tag{29b}$$

where a is the true inverse susceptibility evaluated at zero wave vector. Then (29a), (28), (20d), and (19a) yield

$$\alpha_{90°} = [(\partial a/\partial \rho)^2_{\nabla_3 u}/V_{90°}](k^2/\gamma)\int \frac{d^3q}{(2\pi)^3}\chi(\vec{q})/[\omega^2+4\Gamma^2(\vec{q})] , \tag{30a}$$

where γ is the true viscosity which determines the rate of decay of the order parameter. Similarly, using (A8), (A6), (29b), (20a), and (19a) we find

$$\alpha_{0°} = \{ [(\partial a/\partial \rho)_{\nabla_3 u} - (\partial a/\partial \nabla_3 u)_\rho]^2/V_{0°} \}(k^2/\gamma)\int \frac{d^3q}{(2\pi)^3}\chi(\vec{q})/[\omega^2+4\Gamma^2(\vec{q})] . \tag{30b}$$

Aside from the inverse of the speed of sound in the two directions, which occur in the definition of α, the anisotropy in the damping of sound comes from the derivative of a with respect to ρ and $\nabla_3 u$. Physically we expect molecular interactions and hence T_c to increase as ρ increases so that $(\partial a/\partial \rho)_{\nabla_3 u} < 0$. On the other hand, a negative $\nabla_3 u$ corresponds to a smaller layer spacing, hence we expect T_c to increase with decreasing $\nabla_3 u$,[16] making $(\partial a/\partial \nabla_3 u)_\rho > 0$. Therefore, we expect $(\partial a/\partial \rho)_{\nabla_3 u}$ and $-(\partial a/\partial \nabla_3 u)_\rho$ to have the same sign, which means that the attenuation at 0° would be larger than that at 90°, aside from the speed of sound factors. Since the anomaly for the speed of sound is a weaker function of reduced temperature than for the viscosities, at least for small ω [see Eqs. (25) and (26)], we would expect the experimental results to show $\alpha_{0°}$ to be greater than $\alpha_{90°}$ very near the transition.

This is in contrast to the nematic—smectic-A transition where the attenuation was experimentally shown to be dominated by an *isotropic* contribution.[2] This feature was explained[3,4] by coupling the order parameter to the density. Coupling of the order parameter to the gradient of the layer spacing as well as the density predicts an *anisotropic* attenuation near the AC transition.

For arbitrary Θ between 0° and 90° the behavior of the attenuation and dispersion can again be determined by knowing the shift in the viscosities, Eq. (20), and the elastic constants, Eq. (21). The solutions, however, are considerably more complicated.[10]

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APPENDIX

The linearized equations of motion of the hydrodynamic variables in the smectic-A phase for arbitrary propagation direction are,[9,10] in the notation of MPP (note—ξ is a transport coefficient in MPP),

$$-i\omega\delta\rho+ik_iv_i=0\ , \tag{A1a}$$

$$-i\omega u-v_3-i\zeta k_3\delta\phi_3-i\xi T^{-1}k_3\delta T=0\ , \tag{A1b}$$

$$-i\omega\delta Q+\xi k_3^2\delta\phi_3+\kappa_\perp k_ik_i\delta T+(\kappa_{||}-\kappa_\perp)k_3^2\delta T=0\ , \tag{A1c}$$

$$-i\omega v_i+ik_i\delta p-i\delta_{i,3}k_3\delta\phi_3+\eta_{ijkl}k_jk_lv_k=0\ . \tag{A1d}$$

Choosing ρ, s, and $\nabla_3 u$ to be the independent variables we can expand the remaining quantities:

$$\delta p=(\partial p/\partial\rho)_{s,\nabla_3 u}\delta\rho+(\partial p/\partial s)_{\rho,\nabla_3 u}\delta s+(\partial p/\partial\nabla_3 u)_{s,\rho}\nabla_3 u\ , \tag{A2a}$$

$$\delta T=(\partial T/\partial\rho)_{s,\nabla_3 u}\delta\rho+(\partial T/\partial s)_{\rho,\nabla_3 u}\delta s+(\partial T/\partial\nabla_3 u)_{s,\rho}\nabla_3 u\ , \tag{A2b}$$

$$\delta\phi_3=(\partial\phi_3/\partial\rho)_{s,\nabla_3 u}\delta\rho+(\partial\phi_3/\partial s)_{\rho,\nabla_3 u}\delta s+(\partial\phi_3/\partial\nabla_3 u)_{s,\rho}\nabla_3 u\ . \tag{A2c}$$

For an angle of $\Theta=0^\circ$ between the direction of propagation and the normal to the smectic layers we have $\vec{k}=k\hat{e}_3$. In this case the equations of motion become

$$-i\omega\delta\rho+i\rho kv_3=0\ , \tag{A3a}$$

$$\begin{aligned}-i\omega u-v_3-i\zeta k[(\partial\phi_3/\partial\rho)\delta\rho+(\partial\phi_3/\partial s)\delta s+(\partial\phi_3/\partial\nabla_3 u)\nabla_3 u]\\ -(i\xi k/T)[(\partial T/\partial\rho)\delta\rho+(\partial T/\partial s)\delta s+(\partial T/\partial\nabla_3 u)\nabla_3 u]=0\ ,\end{aligned} \tag{A3b}$$

$$\begin{aligned}-i\omega T\delta s+\xi k^2[(\partial\phi_3/\partial\rho)\delta\rho+(\partial\phi_3/\partial s)\delta s+(\partial\phi_3/\partial\nabla_3 u)\nabla_3 u]\\ +\kappa_{||}k^2[(\partial T/\partial\rho)\delta\rho+(\partial T/\partial s)\delta s+(\partial T/\partial\nabla_3 u)\nabla_3 u]=0\ ,\end{aligned} \tag{A3c}$$

$$-i\omega v_1+\eta_3k^2v_1=0\ , \tag{A3d}$$

$$-i\omega v_2+\eta_3k^2v_2=0\ , \tag{A3e}$$

$$\begin{aligned}-i\omega v_3+ik[(\partial p/\partial\rho)\delta\rho+(\partial p/\partial s)\delta s+(\partial p/\partial\nabla_3 u)\nabla_3 u]\\ -ik[(\partial\phi_3/\partial\rho)\delta\rho+(\partial\phi_3/\partial s)\delta s+(\partial\phi_3/\partial\nabla_3 u)\nabla_3 u]+\eta_1k^2v_3=0\ .\end{aligned} \tag{A3f}$$

There are two dissipative modes arising from the v_1 and v_2 equations, each characterized by

$$\omega=-ik^2\eta_3\ . \tag{A4}$$

Solving the remaining four equations will give the other four modes. First we find the zeroth-order solutions by neglecting all dissipative terms. This is easily done by substituting for $\delta\rho$ and $\nabla_3 u$ in the v_3 equation. We find two additional dissipative solutions (Re $\omega=0$) and a pair of propagating solutions with

$$\omega^2=k^2c_{33}\ , \tag{A5}$$

where c_{33} is given by Eq. (17c). The speed associated with these modes is

$$V_{0^\circ}=\omega/k=\pm c_{33}^{1/2}\ . \tag{A6}$$

The attenuation is found by including the dissipative terms to first order. The correction to the angular frequency is

$$\begin{aligned}\Delta\omega=(i/2c_{33})(\,[(\partial p/\partial s)-(\partial\phi_3/\partial s)]\{\,\xi k^2(c_{33}-c_{13})+\kappa_{||}k^2[(\partial T/\partial\nabla_3 u)-(\partial T/\partial\rho)]\,\}\\ +(c_{33}-c_{13})\{\,\zeta k^2(c_{13}-c_{33})+(\xi k^2/T)[(\partial T/\partial\rho)-(\partial T/\partial\nabla_3 u)]\,\}-c_{33}k^2\eta_1)\ ,\end{aligned} \tag{A7}$$

where c_{13} is given by Eq. (17b). Since the most divergent dissipative parameters near the AC transition are the η's, we find for the adiabatic sound attenuation

$$\alpha_{0^\circ}=i\Delta\omega/V_{0^\circ}\cong k^2\eta_1/2c_{33}^{1/2}\ . \tag{A8}$$

For $\Theta=90^\circ$ (propagation parallel to the layers) $\vec{k}=k\hat{e}_1$ without loss of generality. In this case the equations of motion are

$$-i\omega\delta\rho+ikv_1=0\ , \tag{A9a}$$

$$-i\omega u-v_3=0\ , \tag{A9b}$$

$$-i\omega T\delta s+\kappa_\perp k^2[(\partial T/\partial\rho)\delta\rho+(\partial T/\partial s)\delta s+(\partial T/\partial\nabla_3 u)\nabla_3 u]=0\ . \tag{A9c}$$

$$-i\omega v_1+ik[(\partial p/\partial\rho)\delta\rho+(\partial p/\partial s)\delta s+(\partial p/\partial\nabla_3 u)\nabla_3 u]+(\eta_2+\eta_4)k^2v_1=0\ , \tag{A9d}$$

$$-i\omega v_2+\eta_2k^2v_2=0\ , \tag{A9e}$$

$$-i\omega v_3+\eta_3k^2v_3=0\ . \tag{A9f}$$

Two dissipative modes are described by the last two equations and have

$$\omega=-i\eta_2k^2 \tag{A10a}$$

and

$$\omega=-i\eta_3k^2\ . \tag{A10b}$$

From the other four equations we again find two additional dissipative modes and a pair of propagating modes with

$$V_{90^\circ}=\pm(\partial p/\partial\rho)^{1/2}_{\nabla_3 u}\ . \tag{A11}$$

Doing first-order perturbation theory gives

$$\Delta\omega=-[i/2(\partial p/\partial\rho)][(\partial p/\partial s)\kappa_\perp k^2(\partial T/\partial\rho)+(\partial p/\partial\rho)k^2(\eta_2+\eta_4)] \tag{A12}$$

or

$$\alpha_{90^\circ}\cong k^2(\eta_2+\eta_4)/2(\partial p/\partial\rho)^{1/2}\ . \tag{A13}$$

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