

Viscosity coefficients near the nematic–smectic-*A* phase transition

V. P. Romanov and S. V. Ul'yanov

*Department of Physics, St. Petersburg State University, Petrodvorets, St. Petersburg 198904, Russia
and Department of Higher Mathematics, Institute of Trade and Economics, St. Petersburg 194018, Russia*

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The equations of motion of nematic liquid crystals are complemented by the nonlinear Langevin equation for the smectic order parameter fluctuations. The nonlinear terms of this equation describe coupling of the order parameter to velocity and density. An expression for the reactive part of the stress tensor is obtained. The complex fluctuation inputs into viscosity coefficients are calculated in the frame of the Landau–de Gennes model near the nematic–smectic-*A* (*N-A*) phase transition. The sound velocity dispersion and the attenuation dependences on temperature, frequency, and the direction of the sound wave propagation are calculated. The results obtained are sensitive to values of two critical exponents for correlation lengths. [S1063-651X(97)09404-X]

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I. INTRODUCTION

Investigation of the nematic–smectic-*A* (*N-A*) phase transition has attracted considerable interest for a long time [1–4] and various theoretical models for this transition have been analyzed [2]. The emphasis is on the investigation of the nature of the *N-A* transition, the construction of relevant models, and the determination of critical exponents for two correlation lengths. The basic experimental methods applied in this system are calorimetrics and light scattering. These methods enable one to study the equilibrium properties of *N-A* transitions in detail. For study of the kinetic properties acoustic measurements are most effective since they give information on the mechanism of the phase transition. Experiments on the propagation of longitudinal sound waves have shown an anomalous increase of the attenuation and anisotropy of sound velocity dispersion [3,5–10]. The existence of two correlation lengths [2,4,11] results in complicate dependences of the viscosity coefficients on temperature and frequency and this in turn produces the complicated behavior of the acoustic parameters.

Unfortunately, the acoustic properties are sensitive to the type of liquid crystal. Therefore near the *N-A* phase transition some systems show angular dependences [5,8,10] whereas other systems do not exhibit this behavior [6,7,9].

The complicated point in developing the theory of acoustic properties for *N-A* transition is in the necessity of accounting for various contributions to the critical sound behavior. In particular, Kiry and Martinoty [5] and Swift and Mulvaney [12] have considered in detail the input of the smectic order parameter fluctuations into the bulk viscosity coefficient. They calculated the temperature and frequency dependences of this contribution which causes an isotropic anomalous attenuation of sound and also gives an input to the speed of sound near the phase transition point.

To describe the anisotropy of the acoustic properties it is necessary to calculate the contributions to other viscosity coefficients. McMillan [13] and Jahnig and Brochard [14] studied the coupling of the smectic order parameter fluctuations to the director near the phase transition point. They calculated the fluctuation input into the α_1, α_2 Leslie coeffi-

cients. Using the NAC model Swift and co-workers [15,16] considered the coupling of the order parameter fluctuations to the velocity. They calculated the complex input into the α_1 Leslie coefficient and analyzed its behavior near phase transition point. It was found that in acoustic problems the bulk viscosity dominates and the coupling of the smectic order parameter fluctuation to the velocity is more important compared with the coupling to the director. Actually, five complex viscosity coefficients determine the acoustic properties of the system. The sound velocity dispersion is determined by the imaginary parts of these coefficients and the sound attenuation is a linear function of the real parts.

The aim of this work is to calculate the frequency dispersion of all five viscosity coefficients caused by the interaction of the sound wave with the smectic order parameter fluctuations. The viscosity coefficients are contained in the dissipative part of the stress tensor. To calculate the fluctuation corrections to the stress tensor in the anisotropic uniaxial medium we add the nonlinear Langevin equation describing the variation of the smectic order parameter. By solution of the system equations of motion by iteration after statistical averaging we get the fluctuation contribution to the stress tensor. This input satisfies the symmetry conditions and provides the fluctuation corrections to all viscosity coefficients. Finally, the nematic liquid crystal acoustical properties are calculated. It turns out that they are strongly dependent on two critical exponents of the correlation lengths. Final results admit comparison of the theory and the experiment.

The article is organized as follows. In Sec. II we construct a set of equations of motion for nematic liquid crystals which include the equation for an additional scalar variable. They are used in Sec. III for the calculation of fluctuation inputs into all viscosity coefficients. Sound damping and velocity dispersion are derived. We discuss the obtained results in Sec. IV.

II. BASIC EQUATIONS

The set of hydrodynamic equations for nematic liquid crystals includes the continuity equation [1,17]

$$\frac{\partial \rho}{\partial t} + \nabla(\rho \mathbf{v}) = 0, \quad (1)$$

the equation of motion for the director,

$$\frac{\partial n_j}{\partial t} + v_k \partial_k n_j = \frac{1}{2} (\partial_k v_j - \partial_j v_k) n_k + \lambda (\delta_{jl} - n_j n_l) n_k v_{lk} + \frac{h_j}{\gamma}, \quad (2)$$

the Navier-Stokes equation

$$\rho \left(\frac{\partial v_j}{\partial t} + v_k \partial_k v_j \right) = \partial_k \sigma_{jk}, \quad (3)$$

$$\sigma_{jk} = -p \delta_{jk} + \sigma_{jk}^{(n)} + \sigma'_{jk}, \quad (4)$$

$$\begin{aligned} \sigma_{jk}^{(n)} = & \frac{1}{2} [\partial_l [\pi_{kl} n_j + \pi_{jl} n_k - (\pi_{jk} + \pi_{kj}) n_l] \\ & - (\pi_{kl} \partial_j n_l + \pi_{jl} \partial_k n_l) - \lambda (n_j h_k + n_k h_j)], \end{aligned}$$

and the equation for the variation of the entropy (per unit volume),

$$T \left(\frac{\partial S}{\partial t} + \nabla(\mathbf{v}S) \right) = 2R, \quad (5)$$

where R is the dissipative function,

$$2R = \sigma'_{jk} v_{jk} + \frac{h^2}{\gamma}. \quad (6)$$

The heat conductivity input into acoustic parameters is negligible and hence we ignore this process. Here and below we sum over repeated indices; ρ is the density, \mathbf{v} is the velocity, \mathbf{n} is the director, p is the pressure, $\partial_j = \partial / \partial x_j$, $v_{jk} = \frac{1}{2} (\partial_j v_k + \partial_k v_j)$, $h_j = (\delta_{jk} - n_j n_k) H_k$, $H_k = \partial_m \pi_{mk} - \partial F^{\text{Fr}} / \partial n_k$, and $\pi_{mk} = \partial F^{\text{Fr}} / \partial (\partial_m n_k)$; F^{Fr} is the Frank free energy density for director distortions,

$$F^{\text{Fr}} = \frac{1}{2} [K_{11}(\text{div} \mathbf{n})^2 + K_{22}(\mathbf{n} \text{curl} \mathbf{n})^2 + K_{33}[\mathbf{n} \text{curl} \mathbf{n}]^2], \quad (7)$$

where K_{11}, K_{22}, K_{33} are the Frank elastic constants, λ and the twist viscosity γ are phenomenological coefficients, σ'_{jk} is the dissipative part of the stress tensor. The latter is symmetric in the j and k indices and has uniaxial symmetry in the nematic state. It contains five independent viscosity coefficients [17]

$$\begin{aligned} \sigma'_{jk} = & 2 \eta_1 v_{jk} + \left(\zeta_1 - \frac{2}{3} \eta_1 \right) \delta_{jk} v_{ll} + \zeta_2 (\delta_{jk} n_l n_m v_{lm} + n_j n_k v_{ll}) \\ & + \eta_2 (n_j n_l v_{kl} + n_k n_l v_{jl}) + \eta_3 (n_j n_k n_l n_m v_{lm}). \end{aligned} \quad (8)$$

The coefficients η_1 and ζ_1 are similar to the shear and bulk viscosities in the ordinary isotropic fluids. The viscosities ζ_2 , η_2 , and η_3 are specific for fluids with uniaxial symmetry. The coefficient ζ_2 is the second bulk viscosity and it vanishes for incompressible nematics. The coefficients η_2 and η_3 are two additional shear viscosities. We select this set of viscosity coefficients due to its convenience for acoustical problems. It is related to the set of viscosity coefficients $\eta'_1, \eta'_2, \eta'_3, \eta'_4, \eta'_5$ used in [17] by the relations

$$\eta_1 = \eta'_1, \eta_2 = \eta'_3 - 2 \eta'_1,$$

$$\eta_3 = \eta'_5 + \eta'_1 + \eta'_2 - 2 \eta'_3 - 2 \eta'_4,$$

$$\zeta_1 = \eta'_2 - \frac{1}{3} \eta'_1, \zeta_2 = \eta'_4 + \eta'_1 - 2 \eta'_2. \quad (9)$$

In addition, we present the relation of shear viscosity coefficients η_1, η_2, η_3 and parameters λ, γ of Eq. (2) with the Leslie coefficients $\alpha_1, \alpha_2, \dots, \alpha_6$, describing an incompressible nematic,

$$\gamma = \alpha_3 - \alpha_2,$$

$$\lambda = \frac{\alpha_3 + \alpha_2}{\alpha_2 - \alpha_3},$$

$$\eta_1 = \frac{\alpha_4}{2}, \quad (10)$$

$$\eta_2 = \alpha_6 + \alpha_3 \lambda = \alpha_5 + \alpha_2 \lambda,$$

$$\eta_3 = \alpha_1.$$

Note that some of coefficients $\eta_1, \eta_2, \eta_3, \zeta_1, \zeta_2$ may be negative. The stability condition requires the positiveness of five combinations composed of these coefficients only [17]. In particular, the coefficient η_2 may be negative, but the linear combination $2 \eta_1 + \eta_2$ must be positive, etc.

The solution of the linearized equations of motion yields the following expressions for the sound velocity c and the sound absorption coefficient α in nematics [1,17]:

$$c = \sqrt{\left(\frac{\partial p}{\partial \rho} \right)_s}, \quad (11)$$

$$\alpha = \frac{\omega^2}{2 \rho c^3} \left[\zeta_1 + \frac{4}{3} \eta_1 + 2(\zeta_2 + \eta_2) \cos^2 \theta + \eta_3 \cos^4 \theta \right], \quad (12)$$

where ω is the circular frequency and θ is the angle between the direction of the sound wave propagation and the direction of the average molecular alignment \mathbf{n}_0 .

In the vicinity of the N - A phase transition point the fluctuations of the smectic order parameter are essential. Their input into the free energy density in the nematic phase is [1,18]

$$F^\Psi = \frac{1}{2} (A |\Psi|^2 + L_{\parallel} |\nabla_{\parallel} \Psi|^2 + L_{\perp} |\nabla_{\perp} \Psi|^2). \quad (13)$$

The coefficient A turns to zero at the N - A phase transition temperature T_{NA} and as usual $A = a_0 (T - T_{NA})^\gamma$ with $\gamma = \frac{4}{3}$ using the helium analogy, or $\gamma = 1$ in the mean-field approximation, and L_{\parallel} and L_{\perp} are functions weakly dependent on temperature. Here we neglect the coupling of the order parameter to the director fluctuations. The effect of this coupling is the renormalization of the Frank elastic constants K_{22} and K_{33} , and it explains their divergence near the N - A phase transition [18,19].

We consider the interaction of the sound wave with the fluctuations of the smectic order parameter. To take into account this interaction it is necessary to include an additional equation for the variation of the smectic order parameter to

the equations of motion of the nematic. This equation may be constructed on the basis of the system's symmetry. If each liquid particle is driven by a homogeneous velocity field with its own value of the order parameter, we have

$$\frac{d|\Psi|}{dt} = \frac{\partial|\Psi|}{\partial t} + \mathbf{v} \cdot \nabla|\Psi| = 0. \quad (14)$$

If we take into account the relaxation process and the nonhomogeneity of the velocity field, we should write on the right-hand side of Eq. (14) the relaxation term $-b\delta F^\Psi/\delta\Psi$ and terms describing the interaction between the smectic order parameter and the velocity field in the uniaxial media. Therefore, the total equation describing variation of the order parameter has the form

$$\begin{aligned} \frac{\partial|\Psi|}{\partial t} + \mathbf{v} \cdot \nabla|\Psi| = & -b \frac{\delta F^\Psi}{\delta|\Psi|} + Wv_{ll}|\Psi| + Zn_l n_k v_{lk}|\Psi| \\ & - Xn_l v_l n_k \nabla_k|\Psi|, \end{aligned} \quad (15)$$

where X , W , and Z are phenomenological coefficients. Equation (15) describes the dynamics of the smectic order parameter in the second-order approximation when the nematic is fixed as whole. The last three terms are responsible for the interaction with the inhomogeneous velocity field. The additional variable produces additional terms to the stress tensor due to the order parameter interaction with the velocity field. These terms may be obtained by the method described in [17]. It is based on the energy conservation law in the local form

$$\frac{\partial}{\partial t} \left(\frac{\rho v^2}{2} + E \right) = -\text{div} \mathbf{Q}, \quad (16)$$

where E is the internal energy density and \mathbf{Q} is the energy flux density. This equation provides us with the ability to obtain the dissipative function (6) and the stress tensor. If we differentiate the left-hand side of Eq. (16) in the explicit form, we get

$$\begin{aligned} \frac{\partial}{\partial t} \left(\frac{\rho v^2}{2} + E \right) = & \frac{v^2}{2} \frac{\partial \rho}{\partial t} + \rho \mathbf{v} \cdot \frac{\partial \mathbf{v}}{\partial t} + \mu \frac{\partial \rho}{\partial t} + T \frac{\partial S}{\partial t} \\ & + \left(\frac{\partial E^{\text{Fr}}}{\partial t} \right)_{\rho, S, |\Psi|} + \left(\frac{\partial E^\Psi}{\partial t} \right)_{\rho, S, \mathbf{n}}, \end{aligned} \quad (17)$$

where μ is the chemical potential. The term $(\partial E^{\text{Fr}}/\partial t)_{\rho, S, |\Psi|}$ was analyzed in [17]. It produces the expression (4) for the stress tensor σ_{jk} . We consider the last term of Eq. (17) in a similar way. The derivative $(\partial E^\Psi/\partial t)_{\rho, S, \mathbf{n}}$ may be presented in the form

$$\left(\frac{\partial E^\Psi}{\partial t} \right)_{\rho, S, \mathbf{n}} = \left(\frac{\partial F^\Psi}{\partial|\Psi|} - \partial_k \theta_k \right) \frac{\partial|\Psi|}{\partial t} + \partial_k \left(\theta_k \frac{\partial|\Psi|}{\partial t} \right), \quad (18)$$

where

$$\begin{aligned} \theta_k = & \frac{\partial E^\Psi}{\partial(\partial_k|\Psi|)} = L_\perp \partial_k|\Psi| + (L_\parallel - L_\perp) n_k n_l \partial_l|\Psi| \\ = & L_\perp \left[\delta_{kl} + \left(\frac{L_\parallel}{L_\perp} - 1 \right) n_k n_l \right] \partial_l|\Psi|. \end{aligned} \quad (19)$$

Here ‘‘the small additions theorem’’ [20] is used. In our case it has the form $F^{\text{Fr}} + F^\Psi = E^{\text{Fr}} + E^\Psi$ for the corresponding choice of external conditions.

If we substitute the equations of motion (1), (3), and (5) and Eq. (18) into the right-hand side of Eq. (17), we can form a term of the divergence type. Therefore, Eq. (17) can be written as

$$\frac{\partial}{\partial t} \left(\frac{\rho v^2}{2} + E \right) = -\text{div} \mathbf{Q} + 2R - \left[\sigma'_{lk} v_{lk} + \frac{h^2}{\gamma} + b \left(\frac{\delta F^\Psi}{\delta|\Psi|} \right)^2 \right], \quad (20)$$

where

$$\sigma'_{jk} = \sigma_{jk} + p \delta_{jk} - \sigma_{jk}^{(n)} - \sigma_{jk}^{(\Psi)}, \quad (21)$$

$$\begin{aligned} \sigma_{jk}^{(\Psi)} = & -\theta_k \partial_j|\Psi| - Xn_j n_m \theta_k \partial_m|\Psi| + Xn_j n_k E^\Psi \\ & + \left(\frac{\partial F^\Psi}{\partial|\Psi|} - \partial_l \theta_l \right) |\Psi| (W \delta_{jk} + Zn_j n_k), \end{aligned} \quad (22)$$

where $p = \mu\rho + TS - E$ is the pressure. The reactive part of the stress tensor $\sigma_{jk}^{(\Psi)}$ is calculated for $\mathbf{n} = \mathbf{n}_0$. The expression for the energy flux density \mathbf{Q} is not presented here since it is too cumbersome and it is not used in the following calculations.

Thus, the conservation law is valid under the condition

$$2R = \sigma'_{lk} v_{lk} + \frac{h^2}{\gamma} + b \left(\frac{\delta F^\Psi}{\delta|\Psi|} \right)^2. \quad (23)$$

This relation estimates the form of the dissipative function and the form of the reactive part $\sigma_{jk}^{(\Psi)}$ of the stress tensor. The stress tensor σ_{jk} and the tensor $\sigma_{jk}^{(\Psi)}$ should be symmetric. Therefore, the phenomenological coefficient X in Eq. (15) and coefficients L_\parallel, L_\perp in Eq. (13) should satisfy the relation

$$X = \frac{L_\parallel - L_\perp}{L_\perp}. \quad (24)$$

In this case the reactive part $\sigma_{jk}^{(\Psi)}$ of the stress tensor has the symmetric form

$$\begin{aligned} \sigma_{jk}^{(\Psi)} = & -L_\perp (\delta_{jl} + Xn_j n_l) (\delta_{km} + Xn_k n_m) (\partial_l|\Psi|) (\partial_m|\Psi|) \\ & + Xn_j n_k F^\Psi + (W \delta_{jk} + Zn_j n_k) \left(\frac{\partial F^\Psi}{\partial|\Psi|} - \partial_l \theta_l \right) |\Psi|. \end{aligned} \quad (25)$$

Thus, the presence of the additional variable $|\Psi|$ supplements the equations of motion (1)–(6) by the additional equation (15) for the variable $|\Psi|$. The stress tensor has the form

$$\sigma_{jk} = -p\delta_{jk} + \sigma_{jk}^{(n)} + \sigma_{jk}^{(\Psi)} + \sigma'_{jk} \quad (26)$$

$$\chi^{-1}(\mathbf{q}) = A + L_{\perp}q_{\perp}^2 + L_{\parallel}q_{\parallel}^2, \quad (30)$$

where the additional reactive term is given by Eq. (25)

$$G_{\mathbf{q},\omega}^0 = [-i\omega + b\chi^{-1}(\mathbf{q})]^{-1}, \quad (31)$$

III. VISCOSITY COEFFICIENTS

In the ordered nematics near the N - A phase transition point the fluctuating inputs to the viscosity coefficients can be calculated using the equations of motion which were obtained in the previous section. To this end we use an approach based on the description of the dynamics of the smectic order parameter fluctuations by the Langevin equation. This equation includes the stochastic force $f(\mathbf{r},t)$ noncorrelated in space and time.

It is convenient to use the Fourier transform

$$\Psi_{\mathbf{q},\omega} = \frac{1}{\sqrt{V}} \int d\mathbf{r} \int_{-\infty}^{\infty} dt \Psi(\mathbf{r},t) e^{-i\mathbf{q}\cdot\mathbf{r} + i\omega t}, \quad (27)$$

$$\Psi(\mathbf{r},t) = \frac{1}{\sqrt{V}} \sum_{\mathbf{q}} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \Psi_{\mathbf{q},\omega} e^{i\mathbf{q}\cdot\mathbf{r} - i\omega t}, \quad (28)$$

where V is the volume. We introduce the Cartesian coordinates (x_1, x_2, x_3) with the axis x_3 directed along the equilibrium director \mathbf{n}_0 .

The Langevin equation obtained from Eq. (15) in the \mathbf{q}, ω presentation can be written in the form

$$\begin{aligned} \Psi_{\mathbf{q},\omega} = & G_{\mathbf{q},\omega}^0 \left\{ f_{\mathbf{q},\omega} - \frac{1}{\sqrt{V}} \sum_{\mathbf{q}'} \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \left[i[(q_l - q'_l) \right. \right. \\ & + X\delta_{l3}(q_3 - q'_3) - Wq'_l - Z\delta_{l3}q'_3] v_{l,\mathbf{q}',\omega'} \\ & \left. \left. + b \left(\frac{\partial \chi^{-1}(0)}{\partial \rho} \right)_s \rho_{\mathbf{q}',\omega'} \right] \Psi_{\mathbf{q}-\mathbf{q}',\omega-\omega'} \right\}, \quad (29) \end{aligned}$$

where

and $f_{\mathbf{q},\omega}$ is the Fourier component of the stochastic force with the correlation function

$$\langle f_{\mathbf{q},\omega} f_{\mathbf{q}',\omega'} \rangle = \frac{2(2\pi)^4 b k_B T}{V} \delta(\mathbf{q} + \mathbf{q}') \delta(\omega + \omega'). \quad (32)$$

Besides the Langevin equation (29) it is necessary to use the continuity equation in the linear form

$$\delta \rho_{\mathbf{q},\omega} = \frac{\rho}{\omega} q_l v_{l,\mathbf{q},\omega} \quad (33)$$

and the equation of motion

$$-i\omega \rho v_{j,\mathbf{q},\omega} = -iq_k p_{\mathbf{q},\omega} \delta_{jk} + iq_k (\sigma_{jk,\mathbf{q},\omega}^{(n)} + \sigma_{jk,\mathbf{q},\omega}^{(\Psi)} + \sigma'_{jk,\mathbf{q},\omega}). \quad (34)$$

In Eq. (34) the pressure p consists of two parts [17] $p = p_0 + p^{(\Psi)}$, where p_0 is the thermodynamic pressure and $p^{(\Psi)} = \rho(\partial E^{\Psi}/\partial \rho)_s - E^{\Psi}$ is the smectic order parameter fluctuation contribution. Using Eq. (13) we get the expressions for $p_{\mathbf{q},\omega}^{(\Psi)}$ and $\sigma_{jk,\mathbf{q},\omega}^{(\Psi)}$:

$$p_{\mathbf{q},\omega}^{(\Psi)} = \frac{1}{\sqrt{V}} \sum_{\mathbf{q}'} \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \frac{1}{2} \left[\rho \left(\frac{\partial \chi^{-1}(0)}{\partial \rho} \right)_s - \chi^{-1}(0) + L_{\perp} [q'_l(q_l - q'_l) + Xq'_3(q_3 - q'_3)] \right] \Psi_{\mathbf{q}',\omega'} \Psi_{\mathbf{q}-\mathbf{q}',\omega-\omega'}, \quad (35)$$

$$\begin{aligned} \sigma_{jk,\mathbf{q},\omega}^{(\Psi)} = & \frac{1}{\sqrt{V}} \sum_{\mathbf{q}'} \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} (L_{\perp} (\delta_{jm} + X\delta_{j3}\delta_{m3}) (\delta_{kl} + X\delta_{k3}\delta_{l3}) q'_l (q_m - q'_m) + \frac{1}{2} X\delta_{j3}\delta_{k3} \{ \chi^{-1}(0) + L_{\perp} [q'_l(q_l - q'_l) \\ & + Xq'_3(q_3 - q'_3)] \} + [\chi^{-1}(0) + L_{\perp} (\delta_{lm} + X\delta_{l3}\delta_{m3}) q'_l q'_m] (W\delta_{jk} + Z\delta_{j3}\delta_{k3})) \Psi_{\mathbf{q}',\omega'} \Psi_{\mathbf{q}-\mathbf{q}',\omega-\omega'}. \quad (36) \end{aligned}$$

In what follows we omit the Eqs. (2) and (5) describing the variations of the director and the entropy, since we are interested in the coupling between the order parameter fluctuations and the sound wave only.

The Langevin equation is solved by an iteration procedure in the powers of the velocity \mathbf{v} [21,22]. After the second iteration we have

$$\begin{aligned} \Psi_{\mathbf{q},\omega} = & G_{\mathbf{q},\omega}^0 f_{\mathbf{q},\omega} - \frac{1}{\sqrt{V}} \sum_{\mathbf{q}'} \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \left[i[(q_l - q'_l) + X\delta_{l3}(q_3 - q'_3) - Wq'_l - Z\delta_{l3}q'_3] \right. \\ & \left. + b \left(\frac{\partial \chi^{-1}(0)}{\partial \rho} \right)_s \frac{\rho}{\omega'} q'_l \right] v_{l,\mathbf{q}',\omega'} G_{\mathbf{q},\omega}^0 G_{\mathbf{q}-\mathbf{q}',\omega-\omega'}^0 f_{\mathbf{q}-\mathbf{q}',\omega-\omega'}. \quad (37) \end{aligned}$$

The first term on the right-hand side describes the fluctuations of the unperturbed system and the second term presents the interaction of the order parameter fluctuations with the external velocity field. We substitute the solution (37) into the equations of motion (34)–(36) and then average over all realizations of the stochastic forces. Using Eq. (32) after integrating on frequency we get

$$\begin{aligned}
-i\omega\rho v_{j,\mathbf{q},\omega} = & -iq_j \left(\frac{\partial p}{\partial \rho} \right)_s \delta\rho_{\mathbf{q},\omega} + iq_k (\sigma_{jk,\mathbf{q},\omega}^{(n)} + \sigma'_{jk,\mathbf{q},\omega}) + iq_k \int \frac{d\mathbf{q}'}{(2\pi)^3} k_B T \left\{ \frac{1}{2} \left[\rho b \left(\frac{\partial \chi^{-1}(0)}{\partial \rho} \right)_s - \chi^{-1}(0) + L_{\perp} [q'_l (q_l - q'_l) \right. \right. \\
& \left. \left. + Xq'_3 (q_3 - q'_3) \right] \delta_{jk} - L_{\perp} [q'_k (q_j - q'_j) + X(\delta_{k3}q'_j + \delta_{j3}q'_k)(q_3 - q'_3)] - \left[\frac{X}{2} \{ \chi^{-1}(0) - L_{\perp} [q'_l (q_l - q'_l) \right. \right. \\
& \left. \left. - Xq'_3 (q_3 - q'_3) \} \right] + Z[\chi^{-1}(0) + L_{\perp} (q'^2 + Xq'^2_3)] \delta_{j3} \delta_{k3} \right\} \{-i\omega + b[\chi^{-1}(\mathbf{q}') + \chi^{-1}(\mathbf{q} - \mathbf{q}')] \}^{-1} \\
& \times \left\{ \left[b \left(\frac{\partial \chi^{-1}(0)}{\partial \rho} \right)_s \frac{\rho}{\omega} q_l v_{l,\mathbf{q},\omega} - iZq_3 v_{3,\mathbf{q},\omega} \right] [\chi(\mathbf{q}') + \chi(\mathbf{q} - \mathbf{q}')] - i(q'_l v_{l,\mathbf{q},\omega} + Xq'_3 v_{3,\mathbf{q},\omega}) \chi(\mathbf{q}') \right. \\
& \left. - i[(q_l - q'_l) v_{l,\mathbf{q},\omega} + X(q_3 - q'_3) v_{3,\mathbf{q},\omega}] \chi(\mathbf{q} - \mathbf{q}') \right\}. \tag{38}
\end{aligned}$$

Here we substitute the summation over wave vectors \mathbf{q}' by the integration

$$\sum_{\mathbf{q}'} \rightarrow V \int \frac{d\mathbf{q}'}{(2\pi)^3}.$$

Also we omit the terms containing the coefficient W since it is a small correction to the coefficient

$$\frac{\rho}{\chi^{-1}(0)} \left(\frac{\partial \chi^{-1}(0)}{\partial \rho} \right)_s,$$

diverging as $T \rightarrow T_{NA}$. We perform the integration over \mathbf{q}' in Eq. (38) and compare the results obtained with the expression for the viscous part of the stress tensor:

$$\begin{aligned}
\sigma'_{jk,\mathbf{q},\omega} = & i \{ \eta_1 (q_j v_k + q_k v_j) + (\zeta_1 - \frac{2}{3} \eta_1) \delta_{jk} q_l v_l + \zeta_2 (\delta_{jk} q_3 v_3 + \delta_{j3} \delta_{k3} q_l v_l) + \frac{1}{2} \eta_2 [\delta_{j3} (q_3 v_k + q_k v_3) + \delta_{k3} (q_3 v_j + q_j v_3)] \\
& + \eta_3 \delta_{j3} \delta_{k3} q_3 v_3 \}. \tag{39}
\end{aligned}$$

This comparison allows calculating complex corrections $\zeta_1, \zeta_2, \eta_1, \eta_2, \eta_3$ to the viscosity coefficients. Details of this procedure are given in the Appendix. The frequency dispersions of the viscosity coefficients $\eta_i(\omega) - \eta_i(0)$ and $\zeta_j(\omega) - \zeta_j(0), i=1,2,3, j=1,2$, are calculated without cutoff parameter q_m . So we get

$$\text{Re}[\zeta_1(0) - \zeta_1(\omega)] = \frac{\beta k_B T^2 (\gamma_0 - 1) \rho c^2}{128 \pi C_p} \left(1 - \frac{dT_{NA}}{dp} \frac{C_p}{T \alpha_T} \right)^2 \frac{\tau_{\psi}}{r_{\perp}^3} \left[\frac{1}{\chi(0)} \left(\frac{\partial \chi(0)}{\partial T} \right)_p \right]^2 G_1(\omega \tau_{\psi}) \equiv A_{\zeta_1}(T) G_1(\omega \tau_{\psi}), \tag{40}$$

$$\begin{aligned}
\text{Re}[\zeta_2(0) - \zeta_2(\omega)] = & \frac{\beta k_B T^2 \alpha_T \rho c^2}{8 \pi C_p} \left(1 - \frac{dT_{NA}}{dp} \frac{C_p}{T \alpha_T} \right) \frac{\tau_{\psi}}{r_{\perp}^3} \frac{1}{\chi(0)} \left(\frac{\partial \chi(0)}{\partial T} \right)_p \left[ZG_2(\omega \tau_{\psi}) + \frac{X}{6} \left(\frac{1}{4} G_1(\omega \tau_{\psi}) + G_2(\omega \tau_{\psi}) \right) \right] \\
\equiv & A_{\zeta_2}(T) \left[ZG_2(\omega \tau_{\psi}) + \frac{X}{6} \left(\frac{1}{4} G_1(\omega \tau_{\psi}) + G_2(\omega \tau_{\psi}) \right) \right], \tag{41}
\end{aligned}$$

$$\text{Re}[\eta_1(0) - \eta_1(\omega)] = \frac{\beta k_B T}{480 \pi} \frac{\tau_{\psi}}{r_{\perp}^3} [G_1(\omega \tau_{\psi}) - 16G_2(\omega \tau_{\psi}) + 16G_3(\omega \tau_{\psi})], \tag{42}$$

$$\text{Re}[\eta_2(0) - \eta_2(\omega)] = 2X \text{Re}[\eta_1(0) - \eta_1(\omega)], \tag{43}$$

$$\text{Re}[\eta_3(0) - \eta_3(\omega)] = \frac{\beta k_B T}{6 \pi} \frac{\tau_{\psi}}{r_{\perp}^3} \left[\frac{3}{80} X^2 G_1(\omega \tau_{\psi}) + \left(XZ - \frac{X^2}{10} \right) G_2(\omega \tau_{\psi}) + \left(3Z^2 + XZ + \frac{7}{20} X^2 \right) G_3(\omega \tau_{\psi}) \right], \tag{44}$$

$$\text{Im}\{\omega\zeta_1(\omega) - [\omega\zeta_1(\omega)]_{\omega=0}\} = \frac{\beta k_B T^2 (\gamma_0 - 1) \rho c^2}{16 \pi r_{\perp}^3 C_p} \left(1 - \frac{dT_{NA}}{dp} \frac{C_p}{T \alpha_T} \right)^2 \left[\frac{1}{\chi(0)} \left(\frac{\partial \chi(0)}{\partial T} \right)_p \right]^2 G_2(\omega \tau_{\psi}) \equiv B_{\zeta_1}(T) G_2(\omega \tau_{\psi}), \quad (45)$$

$$\text{Im}\{\omega\zeta_2(\omega) - [\omega\zeta_2(\omega)]_{\omega=0}\} = \frac{\beta k_B T^2 \alpha_T \rho c^2}{4 \pi r_{\perp}^3 C_p} \left(1 - \frac{dT_{NA}}{dp} \frac{C_p}{T \alpha_T} \right) \left[\frac{1}{\chi(0)} \left(\frac{\partial \chi(0)}{\partial T} \right)_p \right] \left[\frac{X}{6} G_2(\omega \tau_{\psi}) + \left(Z + \frac{X}{6} \right) G_3(\omega \tau_{\psi}) \right], \quad (46)$$

$$\text{Im}\{\omega\eta_1(\omega) - [\omega\eta_1(\omega)]_{\omega=0}\} = \frac{\beta k_B T}{60 \pi r_{\perp}^3} [G_2(\omega \tau_{\psi}) - 4G_3(\omega \tau_{\psi}) + G_4(\omega \tau_{\psi})], \quad (47)$$

$$\text{Im}\{\omega\eta_2(\omega) - [\omega\eta_2(\omega)]_{\omega=0}\} = 2X \text{Im}\{\omega\eta_1(\omega) - [\omega\eta_1(\omega)]_{\omega=0}\}, \quad (48)$$

$$\text{Im}\{\omega\eta_3(\omega) - [\omega\eta_3(\omega)]_{\omega=0}\} = \frac{\beta k_B T}{\pi r_{\perp}^3} \left[\frac{X^2}{20} G_2(\omega \tau_{\psi}) + \frac{1}{3} \left(XZ - \frac{X^2}{10} \right) G_3(\omega \tau_{\psi}) + \frac{1}{12} \left(3Z^2 + XZ + \frac{7}{20} X^2 \right) G_4(\omega \tau_{\psi}) \right], \quad (49)$$

where

$$\beta = (1 + X)^{-1/2}, \quad r_{\perp} = \sqrt{\frac{L_{\perp}}{A}}, \quad (50)$$

$$\tau_{\psi} = (2bA)^{-1}, \quad (51)$$

$$G_1(x) = x^2 (\sqrt{1+x^2} + 9) (\sqrt{1+x^2} + 3)^{-1} (\sqrt{1+x^2} + 1)^{-2},$$

$$G_2(x) = x^2 (\sqrt{1+x^2} + 1)^{-3/2} [(\sqrt{1+x^2} + 1)^{1/2} + \sqrt{2}]^{-1},$$

$$G_3(x) = \frac{x^2}{\sqrt{2}} (\sqrt{1+x^2} + 1)^{-1} [(\sqrt{1+x^2} + 1)^{1/2} + \sqrt{2}]^{-1}, \quad (52)$$

$$G_4(x) = \sqrt{2} x^2 (\sqrt{1+x^2} + 1)^{-1/2}.$$

In these equations the thermodynamic derivative $(\partial \chi^{-1}(0)/\partial \rho)_s$ is expressed via experimentally measured values. Using the properties of Jacobians [20] we can write

$$\begin{aligned} \rho \left(\frac{\partial \chi^{-1}(0)}{\partial \rho} \right)_s &= - \frac{\alpha_T (\partial \chi^{-1}(0)/\partial T)_p + (C_p/T) (\partial \chi^{-1}(0)/\partial p)_T}{\alpha_T^2 - C_p^2 / \rho T C_V c^2}, \\ & \quad (53) \end{aligned}$$

where C_p and C_V are the regular parts of the heat capacities per unit volume,

$$\alpha_T = \frac{1}{V} \left(\frac{\partial V}{\partial T} \right)_p$$

is the coefficient of the bulk expansion. The value $(\partial \chi^{-1}(0)/\partial p)_T$ may be expressed through the derivative with respect to the temperature since the function $\chi^{-1}(0)$ depends on the difference $[T - T_{NA}(p)]$. Thus we have

$$\left(\frac{\partial \chi^{-1}(0)}{\partial p} \right)_T = - \left(\frac{\partial \chi^{-1}(0)}{\partial T} \right)_p \frac{dT_{NA}}{dp}. \quad (54)$$

Substituting the Eq. (54) into Eq. (53) we get

$$\rho \left(\frac{\partial \chi^{-1}(0)}{\partial \rho} \right)_s = \frac{\rho T c^2 \alpha_T}{C_p} \left(1 - \frac{dT_{NA}}{dp} \frac{C_p}{T \alpha_T} \right) \left(\frac{\partial \chi^{-1}(0)}{\partial T} \right)_p. \quad (55)$$

Here we use the thermodynamic identity

$$C_p - C_V = \frac{\rho T \alpha_T^2 c^2}{\gamma_0}, \quad \gamma_0 = \frac{C_p}{C_V}. \quad (56)$$

Finally from Eqs. (55) and (56) we have

$$\begin{aligned} \rho^2 \left(\frac{\partial \chi^{-1}(0)}{\partial \rho} \right)_s^2 &= \frac{\rho T c^2 (\gamma_0 - 1)}{C_p} \left(1 - \frac{dT_{NA}}{dp} \frac{C_p}{T \alpha_T} \right)^2 \left(\frac{\partial \chi^{-1}(0)}{\partial T} \right)_p^2. \\ & \quad (57) \end{aligned}$$

In Eqs. (40), (41), (45), and (46) we retain the terms with the nearest to singular behavior near the phase transition point, only.

IV. DISCUSSION

Let us analyze the frequency and temperature dependences of the viscosity coefficient dispersions which are determined by the functions G_1, \dots, G_4 of Eq. (52). For low frequencies $\omega \tau_{\psi} \ll 1$, these functions have the asymptotic behavior

$$\begin{aligned} G_1(\omega \tau_{\psi}) &\sim \frac{5}{8} (\omega \tau_{\psi})^2, \\ G_2(\omega \tau_{\psi}) &\sim \frac{1}{8} (\omega \tau_{\psi})^2, \\ G_3(\omega \tau_{\psi}) &\sim \frac{1}{8} (\omega \tau_{\psi})^2, \\ G_4(\omega \tau_{\psi}) &\sim (\omega \tau_{\psi})^2. \end{aligned} \quad (58)$$

Note that all dispersion parts of the complex viscosities are proportional to ω^2 .

For high frequencies, $\omega \tau_{\psi} \gg 1$, we have

$$\begin{aligned}
G_1(\omega\tau_\psi) &\sim 1, \\
G_2(\omega\tau_\psi) &\sim 1, \\
G_3(\omega\tau_\psi) &\sim \sqrt{\frac{\omega\tau_\psi}{2}}, \\
G_4(\omega\tau_\psi) &\sim \sqrt{2}(\omega\tau_\psi)^{3/2}.
\end{aligned} \tag{59}$$

Equation (59) shows that for $\omega \rightarrow \infty$ the functions G_1 and G_2 approach constant levels whereas the functions G_3 and G_4 tend to infinity as $\omega^{1/2}$ and $\omega^{3/2}$, respectively. Hence the inputs $\text{Re}[\zeta_i(0) - \zeta_i(\omega)]$, $i=1,2$, and $\text{Im}\{\omega\zeta_1(\omega) - [\omega\zeta_1(\omega)]_{\omega=0}\}$ are finite and may be calculated for all frequencies. The remaining contributions depend on the functions $G_3(\omega\tau_\psi)$ and $G_4(\omega\tau_\psi)$ and they are divergent for $\omega\tau_\psi \rightarrow \infty$. These divergences are reduced by introducing the cutoff parameter q_m . It means that Eqs. (42)–(44) and (46)–(49) are suitable for $\omega \ll r_\perp^2 q_m^2 / \tau_\psi$ only. Using typical liquid crystal values $r_\perp \sim 2 \times 10^{-8} [(T - T_{NA}) / T_{NA}]^{-0.5}$ cm, $\tau_\psi \sim 10^{-9} [(T - T_{NA}) / T_{NA}]^{-1}$ s, and $q_m \sim 2 \times 10^7$ cm $^{-1}$ [23–26] we obtain the condition $\omega \ll 10^8$ s $^{-1}$.

The dispersion inputs into the viscosities $\zeta_1(\omega)$ and $\text{Re}\zeta_2(\omega)$ have the following asymptotic behavior for high frequencies ($\omega\tau_\psi \gg 1$):

$$\text{Re}[\zeta_1(0) - \zeta_1(\omega)] = 2A_{\zeta_1}(T)[1 - 4\sqrt{2}(\omega\tau_\psi)^{-3/2}], \tag{60}$$

$$\begin{aligned}
&\text{Re}[\zeta_2(0) - \zeta_2(\omega)] \\
&= A_{\zeta_2}(T) \left[Z + \frac{X}{4} - \left(\sqrt{27} + \frac{X}{3\sqrt{2}} \right) (\omega\tau_\psi)^{-1/2} \right], \tag{61}
\end{aligned}$$

$$\text{Im}\{\omega\zeta_1(\omega) - [\omega\zeta_1(\omega)]_{\omega=0}\} = B_{\zeta_1}(T)[1 - \sqrt{2}(\omega\tau_\psi)^{-1/2}]. \tag{62}$$

For the thermodynamic derivative $(\partial\chi(0)/\partial T)_p$ the following relation is valid in the vicinity of the phase transition point:

$$\frac{1}{\chi(0)} \left(\frac{\partial\chi(0)}{\partial T} \right)_p \sim \frac{1}{T - T_{NA}}.$$

Therefore the coefficients ζ_2 and especially ζ_1 increase more rapidly than the shear viscosity coefficients if $T \rightarrow T_{NA}$. Note that the expression for ζ_1 agrees with the bulk viscosity coefficient obtained in [5,12].

The absorption coefficient complex correction is expressed via the complex inputs to the viscosities in the form

$$\begin{aligned}
\tilde{\alpha}(\omega, \theta) &= \frac{\omega^2}{2\rho c^3} \left\{ \zeta_1(\omega) + \frac{4}{3}\eta_1(\omega) + 2[\zeta_2(\omega) + \eta_2(\omega)] \right. \\
&\quad \left. \times \cos^2\theta + \eta_3(\omega)\cos^4\theta \right\}. \tag{63}
\end{aligned}$$

Therefore the fluctuation corrections to the absorption coefficient $\Delta\alpha(\omega, \theta)$ and to the sound velocity dispersion $c(\omega, \theta) - c$ have the form

$$\Delta\alpha(\omega, \theta) = \text{Re}\tilde{\alpha}(\omega, \theta),$$

$$c(\omega, \theta) - c = \omega c^2 \text{Im}\tilde{\alpha}(\omega, \theta). \tag{64}$$

Equations (63) and (64) show that in the vicinity of the phase transition point the complex correction ζ_1 describes the sound damping and the velocity dispersion whereas the correction ζ_2 describes the anisotropy of acoustic parameters.

If $\eta_3 \neq 0$, then Eqs. (63) and (64) predict the deviation of the angle dependences of the acoustic properties from the form $A + B\cos^2\theta$. This deviation was observed in Refs. [8,10].

Note that the contributions to the shear viscosity η_2 and, hence, to the Leslie coefficients α_5 and α_6 are proportional to

$$X = \frac{L_\parallel}{L_\perp} - 1 = \frac{r_\parallel^2}{r_\perp^2} - 1,$$

where $r_\parallel = \sqrt{L_\parallel/A}$. Therefore these coefficients depend on the correlation length's difference only.

Equations (40)–(49) are suitable for calculating the critical corrections to all viscosity coefficients. Therefore there is a possibility to calculate the values

$$\Delta_1(\omega, \theta) = \frac{\alpha(\omega=0, \theta) - \alpha(\omega, \theta)}{[\omega/(2\pi)]^2}, \tag{65}$$

$$\Delta_2(\omega, \theta) = \frac{c(\omega, \theta) - c(\omega=0, \theta)}{c}. \tag{66}$$

The latter may be measured in acoustical experiments. The total expressions for Δ_1 and Δ_2 may be easily obtained from Eqs. (63), (64), and (40)–(49). They are not presented here due to their awkwardness.

For low frequencies, $\omega\tau_\psi \ll 1$, it is possible to describe the character of temperature, frequency, and angular dependences of Δ_1 and Δ_2 in a rather simple form. For this purpose we retain the most divergent with $T \rightarrow T_{NA}$ terms only in the expressions for complex viscosities (40)–(49). Thus we have

$$\begin{aligned}
\Delta_1 &= \omega^2 [A_1 \tau^{-(5-\nu_\parallel-2\nu_\perp)} + B_1 \tau^{-(4+\nu_\parallel-4\nu_\perp)} \cos^2\theta \\
&\quad + C_1 \tau^{-(3+3\nu_\parallel-6\nu_\perp)} \cos^4\theta], \tag{67}
\end{aligned}$$

$$\begin{aligned}
\Delta_2 &= \omega^4 [A_2 \tau^{-(4-\nu_\parallel-2\nu_\perp)} + B_2 \tau^{-(3+\nu_\parallel-4\nu_\perp)} \cos^2\theta \\
&\quad + C_2 \tau^{-(2+3\nu_\parallel-6\nu_\perp)} \cos^4\theta], \tag{68}
\end{aligned}$$

where $\tau = (T - T_{NA}) / T_{NA}$, and A_i, B_i , and C_i , $i=1,2$, are constants in the approximation used. Here we take into account that the correlation lengths have the temperature dependences $r_\parallel = r_{0\parallel} \tau^{-\nu_\parallel}$, $r_\perp = r_{0\perp} \tau^{-\nu_\perp}$, and for small τ we omit the unit in the expression for $X = (r_{0\parallel} / r_{0\perp}) \tau^{-2(\nu_\parallel - \nu_\perp)} - 1$ since $\nu_\parallel > \nu_\perp$ [2,4,11].

Equations (67) and (68) show that the results of acoustical experiments are sensitive to the values of the critical indices ν_\parallel and ν_\perp . The coefficients A_i, B_i , and C_i , $i=1,2$, may be calculated from the angular dependences of the sound attenuation and velocity dispersion. It gives information about the behavior of the viscosity coefficients near N - A transitions.

The angular variation of Δ_1 and Δ_2 may be quite complicated if the signs of the B_i and C_i coefficients are opposite.

Namely, such behavior with $B_2 < 0$ and $C_2 > 0$ was observed in [8] for the sound velocity dispersion.

Besides acoustic methods, it is possible to use experiments on the reflection of the shear wave [27] to measure the critical behavior of the shear viscosity coefficients separately.

APPENDIX

In the integral in Eq. (39) we change the variables \mathbf{q}' , \mathbf{v} to $\tilde{\mathbf{q}}'$, $\tilde{\mathbf{v}}$ with components

$$\tilde{q}'_j = q'_j, \quad \tilde{v}_j = v_j, \quad j = 1, 2,$$

$$\tilde{q}'_3 = \sqrt{1+X}q'_3, \quad \tilde{v}_3 = \sqrt{1+X}v_3. \quad (\text{A1})$$

This integral term in Eq. (38) presents the contribution of the smectic order parameter fluctuations to the stress tensor for which we use the notation $iq_k \tilde{\sigma}'_{jk, \tilde{\mathbf{q}}, \omega}$. The integral is transformed to the form

$$\begin{aligned} iq_k \tilde{\sigma}'_{jk, \tilde{\mathbf{q}}, \omega} = & iq_k \beta k_B T \int \frac{d\tilde{\mathbf{q}}'}{(2\pi)^3} \left(\frac{1}{2} \left[\rho_0 \left(\frac{\partial \chi^{-1}(0)}{\partial \rho} \right)_s - \chi^{-1}(0) + L_\perp \tilde{q}'_l (\tilde{q}_l - \tilde{q}'_l) \right] \delta_{jk} - L_\perp \{ \tilde{q}'_k (\tilde{q}_j - \tilde{q}'_j) + (\sqrt{1+X} - 1) [\delta_{k3} \tilde{q}'_3 (\tilde{q}_j \right. \\ & - \tilde{q}'_j) + \delta_{j3} \tilde{q}'_3 (\tilde{q}_k - \tilde{q}'_k) \} \right) - \left\{ \frac{X}{2} \left[\chi^{-1}(0) - L_\perp \tilde{q}'_l (\tilde{q}_l - \tilde{q}'_l) - 2L_\perp \left(2 \frac{\sqrt{1+X} - 1}{X} - 1 \right) \tilde{q}'_3 (\tilde{q}_3 - \tilde{q}'_3) \right] + Z [\chi^{-1}(0) \right. \\ & \left. + L_\perp \tilde{q}'^2] \right\} \delta_{j3} \delta_{k3} \left\{ -i\omega + b [\chi_\perp^{-1}(\tilde{q}'^2) + \chi_\perp^{-1}((\tilde{\mathbf{q}}' - \tilde{\mathbf{q}})^2)] \right\}^{-1} \left(\left\{ \frac{b\rho}{\omega} \left(\frac{\partial \chi^{-1}(0)}{\partial \rho} \right)_s \tilde{q}_l \tilde{v}_l - \left[\frac{X}{1+X} \frac{b\rho}{\omega} \left(\frac{\partial \chi^{-1}(0)}{\partial \rho} \right)_s \right. \right. \right. \\ & \left. \left. \left. + i \frac{Z}{1+X} \right] \tilde{q}_3 \tilde{v}_3 \right\} [\chi_\perp(\tilde{q}'^2) + \chi_\perp((\tilde{\mathbf{q}} - \tilde{\mathbf{q}}')^2)] - i \tilde{q}'_l \tilde{v}_l \chi_\perp(\tilde{q}'^2) - i(\tilde{q}_l - \tilde{q}'_l) \tilde{v}_l \chi_\perp((\tilde{\mathbf{q}} - \tilde{\mathbf{q}}')^2) \right), \end{aligned} \quad (\text{A2})$$

where the notation

$$\chi_\perp(\tilde{q}'^2) = (A + L_\perp \tilde{q}'^2)^{-1} \quad (\text{A3})$$

is used. If we retain in Eq. (A2) the terms of the order of q^2 we get

$$\begin{aligned} iq_k \tilde{\sigma}'_{jk, \tilde{\mathbf{q}}, \omega} = & iq_k \beta k_B T \int \frac{d\tilde{\mathbf{q}}'}{(2\pi)^3} \left(\frac{1}{2} \left[\rho \left(\frac{\partial \chi^{-1}(0)}{\partial \rho} \right)_s - \chi_\perp^{-1}(\tilde{q}'^2) \right] \delta_{jk} + L_\perp [\tilde{q}'_k \tilde{q}'_j + (\sqrt{1+X} - 1) (\delta_{k3} \tilde{q}'_3 \tilde{q}'_j + \delta_{j3} \tilde{q}'_3 \tilde{q}'_k)] \right. \\ & \left. - \left\{ \frac{X}{2} \left[\chi_\perp^{-1}(\tilde{q}'^2) + 2L_\perp \left(2 \frac{\sqrt{1+X} - 1}{X} - 1 \right) \tilde{q}'^2 \right] + Z \chi_\perp^{-1}(\tilde{q}'^2) \right\} \delta_{j3} \delta_{k3} \right) [-i\omega + 2b \chi_\perp^{-1}(\tilde{q}'^2)]^{-1} \\ & \times \left(2 \left\{ \frac{b\rho}{\omega} \left(\frac{\partial \chi^{-1}(0)}{\partial \rho} \right)_s \tilde{q}_l \tilde{v}_l - \left[\frac{X}{1+X} \frac{b\rho}{\omega} \left(\frac{\partial \chi^{-1}(0)}{\partial \rho} \right)_s + i \frac{Z}{1+X} \right] \tilde{q}_3 \tilde{v}_3 \right\} - i \tilde{q}_l \tilde{v}_l + 2i L_\perp \tilde{q}'_m \tilde{q}'_l \tilde{q}'_m \tilde{v}_l \right). \end{aligned} \quad (\text{A4})$$

The integral terms in Eq. (A4) may be presented in the form

$$\begin{aligned} \int \frac{d\tilde{\mathbf{q}}'}{(2\pi)^3} \tilde{q}'^2 f(\tilde{q}'^2) &= \frac{1}{3} \int \frac{d\tilde{\mathbf{q}}'}{(2\pi)^3} \tilde{q}'^2 f(\tilde{q}'^2), \\ \int \frac{d\tilde{\mathbf{q}}'}{(2\pi)^3} \tilde{q}'_3 \tilde{q}'_j f(\tilde{q}'^2) &= \frac{1}{3} \delta_{3j} \int \frac{d\tilde{\mathbf{q}}'}{(2\pi)^3} \tilde{q}'^2 f(\tilde{q}'^2), \\ \int \frac{d\tilde{\mathbf{q}}'}{(2\pi)^3} \tilde{q}'_k \tilde{q}'_j \tilde{q}'_m \tilde{q}'_l \tilde{q}'_m \tilde{v}_l f(\tilde{q}'^2) &= \frac{1}{15} (\tilde{q}_l \tilde{v}_l \delta_{jk} + \tilde{q}_j \tilde{v}_k + \tilde{q}_k \tilde{v}_j) \int \frac{d\tilde{\mathbf{q}}'}{(2\pi)^3} \tilde{q}'^4 f(\tilde{q}'^2). \end{aligned} \quad (\text{A5})$$

Using the presentation (A5) in Eq. (A4) we can see that the value of the $\tilde{\sigma}'_{jk, \tilde{\mathbf{q}}, \omega}$ tensor depends on the vector components of $\tilde{\mathbf{q}}, \tilde{\mathbf{v}}$ in a similar way as the viscous part of the stress tensor $\tilde{\sigma}_{jk, \mathbf{q}, \omega}$ in Eq. (8) depends on the vector components of \mathbf{q}, \mathbf{v} . Then we replace the variables $\tilde{\mathbf{q}}, \tilde{\mathbf{v}}$ by \mathbf{q}, \mathbf{v} using Eq. (A1). Thus Eq. (A4) transforms into the form

$$\begin{aligned} \sigma'_{jk, \mathbf{q}, \omega} = & i \left[\eta_1 (q_j v_k + q_k v_j) + (\zeta_1 - \frac{2}{3} \eta_1) \delta_{jk} q_l v_l + \zeta_2^{(a)} \delta_{jk} q_3 v_3 + \zeta_2^{(b)} \delta_{j3} \delta_{k3} q_l v_l + \frac{1}{2} \eta_2 [\delta_{j3} (q_3 v_k + q_k v_3) \right. \\ & \left. + \delta_{k3} (q_3 v_j + q_j v_3)] + \eta_3 \delta_{j3} \delta_{k3} q_3 v_3 \right], \end{aligned} \quad (\text{A6})$$

where

$$\begin{aligned}
\eta_1 &= \frac{2\beta k_B T L_\perp^2}{15} \int \frac{d\mathbf{q}'}{(2\pi)^3} \frac{q'^4 \chi_\perp^2(q'^2)}{[-i\omega + 2b\chi_\perp^{-1}(q'^2)]}, \\
\left(\zeta_1 - \frac{2}{3}\eta_1\right) &= -\frac{ib\beta k_B T \rho^2}{\omega} \left(\frac{\partial \chi^{-1}(0)}{\partial \rho}\right)_s^2 \int \frac{d\mathbf{q}'}{(2\pi)^3} \frac{\chi_\perp(q'^2)}{[-i\omega + 2b\chi_\perp^{-1}(q'^2)]}, \\
\zeta_2^{(a)} &= -\beta k_B T \rho \left(\frac{\partial \chi^{-1}(0)}{\partial \rho}\right)_s \left[\left(Z + \frac{X}{2}\right) \int \frac{d\mathbf{q}'}{(2\pi)^3} \frac{\chi_\perp(q'^2)}{[-i\omega + 2b\chi_\perp^{-1}(q'^2)]} - \frac{XL_\perp}{3} \int \frac{d\mathbf{q}'}{(2\pi)^3} \frac{q'^2 \chi_\perp^2(q'^2)}{[-i\omega + 2b\chi_\perp^{-1}(q'^2)]} \right], \\
\zeta_2^{(b)} &= \frac{2ib\beta k_B T \rho}{\omega} \left(\frac{\partial \chi^{-1}(0)}{\partial \rho}\right)_s \left[\left(Z + \frac{X}{2}\right) \int \frac{d\mathbf{q}'}{(2\pi)^3} \frac{1}{[-i\omega + 2b\chi_\perp^{-1}(q'^2)]} - \frac{XL_\perp}{3} \int \frac{d\mathbf{q}'}{(2\pi)^3} \frac{q'^2 \chi_\perp^2(q'^2)}{[-i\omega + 2b\chi_\perp^{-1}(q'^2)]} \right], \\
\eta_2 &= 2X\eta_1, \\
\eta_3 &= \beta k_B T \left[\left(2Z^2 + \frac{2}{3}XZ + \frac{7}{30}X^2\right) \int \frac{d\mathbf{q}'}{(2\pi)^3} \frac{1}{[-i\omega + 2b\chi_\perp^{-1}(q'^2)]} + \frac{4}{3}AX \left(Z - \frac{1}{10}X\right) \int \frac{d\mathbf{q}'}{(2\pi)^3} \frac{\chi_\perp(q'^2)}{[-i\omega + 2b\chi_\perp^{-1}(q'^2)]} \right. \\
&\quad \left. + \frac{2}{5}A^2X^2 \int \frac{d\mathbf{q}'}{(2\pi)^3} \frac{\chi_\perp^2(q'^2)}{[-i\omega + 2b\chi_\perp^{-1}(q'^2)]} \right]. \tag{A7}
\end{aligned}$$

Here we leave the terms closest to begin singular.

Note that the coefficients $\zeta_2^{(a)}$ and $\zeta_2^{(b)}$ have equal real parts $\text{Re}\zeta_2^{(a)} = \text{Re}\zeta_2^{(b)}$. It is a fluctuation correction to the bulk viscosity coefficient ζ_2 . For the imaginary parts of $\zeta_2^{(a)}$ and $\zeta_2^{(b)}$ the following identity is valid: $\text{Im}[\omega\zeta_2^{(a)} - (\omega\zeta_2^{(a)})_{\omega=0}] = \text{Im}[\omega\zeta_2^{(b)} - (\omega\zeta_2^{(b)})_{\omega=0}]$. This expression presents a part of the fluctuation contribution into the sound velocity dispersion. In Eqs. (A7) the expressions for the coefficients $\eta_1, \zeta_2^{(b)}, \eta_2, \eta_3$ contain integrals depending on the cutoff parameter q_m while the most important variables

$$\text{Re}[\zeta_j - (\zeta_j)_{\omega=0}], \quad \text{Im}[\omega\zeta_j - (\omega\zeta_j)_{\omega=0}], \quad j=1,2,$$

$$\text{Re}[\eta_j - (\eta_j)_{\omega=0}], \quad \text{Im}[\omega\eta_j - (\omega\eta_j)_{\omega=0}], \quad j=1,2,3,$$

may be calculated without it. Performing the integration we get Eqs. (40)–(49).

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