

## Anisotropic relaxation in liquid crystals

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We describe phenomenologically the sound propagation at ultrasonic and hypersonic frequencies in nematic and smectic liquid crystals and corresponding isotropic liquid phases. To account for the experimentally well-established effects the standard hydrodynamic equations should be completed by two nonhydrodynamic relaxation mechanisms. More specifically, we prove that by a single nonhydrodynamic relaxation mechanism it is impossible to explain simultaneously both the anisotropy of the sound propagation in the hypersonic regime and the increase of the elastic constants compared to their values in the ultrasonic range. The possible origins of these two nonhydrodynamic relaxation mechanisms are briefly discussed. [S1063-651X(98)14102-8]

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### I. INTRODUCTION

The dynamics of collective propagation modes at ultrasonic [1–6] and hypersonic [7–13] frequencies has been studied experimentally in a large number of compounds. Qualitatively, there are two important general features that characterize the sound propagation in nematic and smectic liquid crystals: (i) There is a significant increase of the sound velocity in the hypersonic regime compared to the ultrasonic one both in isotropic liquids and in nematic and smectic phases of liquid crystalline materials and (ii) there is a large anisotropy in the propagation of acoustic modes in mesophases at hypersonic frequencies. On the other hand, at ultrasonic frequencies, this anisotropy is usually significantly reduced in smectics and practically disappears in nematics. Formally, the sound anisotropy can be well described by a stiffness tensor with effective values of elastic constants [7,12,13]. Due to the uniaxial symmetry of both nematic and smectic-*A* phases, and due to vanishing of the shear components of the strain, only three constants are needed, namely,  $C_{11}$ ,  $C_{13}$ , and  $C_{33}$ .

It is well known [12,14] that the hydrodynamic procedure alone cannot account quantitatively for the observed anisotropy in liquid crystals and, in addition, the differences between ultrasonic and hypersonic propagation clearly suggest the existence of relaxation mechanisms in between the ultrasonic and hypersonic frequency ranges. A relaxation mechanism of quite general nature has to play a significant role; in particular, it should not be related to either any structural relaxation process or molecular ordering in liquid crystals as pointed out in a number of papers [11,15,16]. Actually, such a relaxation mechanism is well known [17] and consists in the transfer of the kinetic energy of molecules due to collisions into their vibrational degrees of freedom. This relaxation mechanism was studied very thoroughly in liquids and in isotropic phases of materials showing various mesophases (see e.g., Refs. [11, 15, 16, 18]). Liu [19] has generalized the hydrodynamic theory by including slowly relaxing quantities when treating the nematic–smectic-*A* phase transition and has remarked that a molecular relaxation would introduce anisotropic dispersion in liquid crystals but he has not inves-

tigated this problem in detail. Other relaxation processes have been considered in order to account for or to predict experimental behaviors of some mesophases. For example, in the case of nematic side-chain polymers, Pleiner and Brand [20] introduced the nematic order parameter itself as an efficient relaxator for providing sound anisotropy in such compounds.

To our knowledge, a deeper discussion of a general scheme, which is supposed to explain the very similar elastic properties of low-molecular-weight nematics and smectics, is still lacking. In this paper we intend to fill the gap between previously considered mechanisms and actual properties of a rather large class of materials. We do not restrict the solution of the problem to two particular directions (propagation parallel or perpendicular to the director), which is usually the case, and we show that the behavior of  $C_{13}$  (which does not contribute to the propagation velocity in these simple directions) is crucial for understanding the whole process. In order to obtain satisfactory agreement with experimental results we show that it is necessary to introduce two nonhydrodynamic relaxation processes into the system of usual hydrodynamic equations describing the sound propagation; one of the relaxators will be identified with the kinetic-energy transfer, as usual, and the nature of the second one will be discussed and tentatively attributed to the above-mentioned relaxation of the nematic order parameter.

We devote the next section of this paper to clarifying the process by which a relaxator provides anisotropy using an elementary model. In Sec. III, comparing with typical experimental data, we show that some important questions remain unsolved using this model. Finally, in the last section we show that an appropriate description of the dynamics of liquid crystals has to involve the participation of a second relaxator closely related to the isotropic-mesophase transition.

### II. ANISOTROPY INDUCED BY A SINGLE RELAXATOR

#### A. Nematics

Let us first consider the nematic case. As is known [14], up to the first order in the wave-vector components, the velocity field  $\vec{v}$  is not coupled to the director orientation. Then,

in the absence of any relaxation process, the equations of motion for  $\vec{v}$  and for the density fluctuation  $\rho$  can be obtained from the mass and momentum conservation laws [21].

We introduce a relaxator  $R$ . Its physical meaning has been mentioned in the introduction, thus  $R$  describes the deviation from the equilibrium value of the population of the excited molecular vibrational state. Let us denote  $Q$  the generalized force conjugate to  $R$ . The rate of change of  $R$  consists of the dissipative (irreversible) flux that is proportional to  $Q$  and of the reactive (reversible) flux. The leading term in the latter is proportional to the gradient of the velocity field, since  $R$  is even in time and since a uniform translation does not alter the nematic state [19]. Hence, the equation of motion for  $R$  reads

$$\dot{R} = -aQ + b_{ij} \frac{\partial v_i}{\partial x_j}. \quad (1)$$

Since the reactive flux should not contribute to the entropy production, it has to be compensated by an additional reactive stress ( $-b_{ij}Q$ ) in the conservation equation of momentum. Finally, the equations of motion are

$$\begin{aligned} \dot{\rho} + \rho_0 \frac{\partial v_i}{\partial x_i} &= 0, \\ \rho_0 \dot{v}_i + \frac{\partial P}{\partial x_i} - b_{ij} \frac{\partial Q}{\partial x_j} &= 0, \end{aligned} \quad (2)$$

$$\dot{R} + aQ - b_{ij} \frac{\partial v_i}{\partial x_j} = 0,$$

where  $\rho_0$  is the equilibrium mass density and where  $P$  stands for the pressure fluctuation. We do not take into account the viscous properties of the fluid; in the first order they do not contribute to the sound velocity anisotropy.

Due to the uniaxial symmetry,  $b_{ij}$  has only two independent components:  $b_{11} = b_{22} = b_{\perp}$  and  $b_{33} = b_{\parallel}$ . The linearized expressions of  $P$  and  $Q$  are written in terms of the three independent derivatives:  $p_{\rho} = \partial P / \partial \rho$ ,  $q_R = \partial Q / \partial R$ , and  $p_R = \partial P / \partial R$  (notice that, due to thermodynamical identities,  $\partial P / \partial R = \rho_0 \partial Q / \partial \rho$ ).

With the above assumptions, it can be easily shown that the frequencies of the eigenmodes are solutions of the following equation:

$$\begin{aligned} \omega^4 + \frac{i}{\tau} \omega^3 - (v_{1\infty}^2 \sin^2 \theta + v_{3\infty}^2 \cos^2 \theta) k^2 \omega^2 - \frac{i v_0^2}{\tau} k^2 \omega \\ + v_0^2 (\delta v)^2 \sin^2 \theta \cos^2 \theta k^4 = 0, \end{aligned} \quad (3)$$

with

$$\begin{aligned} \tau = \frac{1}{q_R a}, \quad v_0^2 = p_{\rho} - \frac{p_R^2}{\rho_0 q_R}, \quad (\delta v)^2 = \frac{(b_{\parallel} - b_{\perp})^2 q_R}{\rho_0}, \\ v_{1\infty}^2 = v_0^2 + \frac{q_R}{\rho_0} \left( b_{\perp} - \frac{p_R}{q_R} \right)^2, \quad v_{3\infty}^2 = v_0^2 + \frac{q_R}{\rho_0} \left( b_{\parallel} - \frac{p_R}{q_R} \right)^2, \end{aligned} \quad (4)$$

where  $k \cos \theta$  and  $k \sin \theta$  are the parallel and perpendicular components of the wave vector, respectively.

For  $\omega \tau \ll 1$ , the solutions consist of a pair of propagation modes showing an isotropic velocity  $v_0$  ( $\omega = \pm v_0 k$ ) and of a diffusive mode [ $\omega = -i \tau (\delta v)^2 \sin^2 \theta \cos^2 \theta k^2$ ] which will not be considered in the following.

For  $\omega \tau \gg 1$ , the solutions consist of two pairs of propagation anisotropic modes, corresponding to the quasilongitudinal (+) and to the quasitransverse (−) waves, respectively. It is convenient to introduce effective elastic constants such as

$$\begin{aligned} C(\theta) \equiv \rho_0 v_{\pm}^2 &= \frac{1}{2} [C_{11} \sin^2 \theta + C_{33} \cos^2 \theta \\ &\pm \sqrt{(C_{11} \sin^2 \theta - C_{33} \cos^2 \theta)^2 + C_{13}^2 \sin^2 2\theta}], \end{aligned} \quad (5)$$

where

$$\begin{aligned} C_0 &= \rho_0 v_0^2, \\ C_{11} &\equiv \rho_0 v_{1\infty}^2 = C_0 (1 + x^2), \\ C_{33} &\equiv \rho_0 v_{3\infty}^2 = C_0 (1 + y^2), \\ C_{13} &= C_0 (1 + xy), \end{aligned} \quad (6)$$

$$\text{with } x = \sqrt{\frac{q_R}{C_0}} \left( b_{\perp} - \frac{p_R}{q_R} \right), \quad y = \sqrt{\frac{q_R}{C_0}} \left( b_{\parallel} - \frac{p_R}{q_R} \right).$$

Notice that if  $x$  and  $y$  have the same sign,  $C_{13}$  lies between  $C_{11}$  and  $C_{33}$ , but  $C_{13}$  is smaller than both  $C_{11}$  and  $C_{33}$  when  $x$  and  $y$  have opposite signs. In the first case the angular variation of the velocity shows only two extrema, at  $\theta = 0$  ( $v_{3\infty}$ ) and  $\theta = \pi/2$  ( $v_{1\infty}$ ). In the second case,  $\theta = 0$  and  $\theta = \pi/2$  correspond to velocity maxima and for some value  $\theta_0$  in the  $[0, \pi/2]$  interval there is a minimum equal to  $C(\theta_0)$ ; it can be shown that  $C(\theta_0) = C_0$ .

When  $\omega \tau$  decreases it follows from Eq. (3) that the quasilongitudinal pair of modes always exists: the angular variation of the velocity keeps the same behavior but its amplitude is continuously reduced down to zero; in addition, there is an imaginary damping term, which shows a similar angular variation; its amplitude reaches a maximum around  $\omega \tau = 1$ . Concerning the quasitransverse mode, it rapidly becomes overdamped when  $\omega \tau$  decreases; its significant broadening when  $\omega \tau$  is not very large may explain that it often cannot be detected experimentally; thus, the frequent absence of the quasitransverse line in the spectra, which was interpreted as a consequence of its dramatic angular intensity variation [13], can also be partly related to the finite value of  $\omega \tau$ .

The contribution of viscosity can be added to Eqs. (2); it does not significantly modify the characteristics of propagation modes and provides an additional damping.

## B. Smectics

Even in the absence of any relaxator contribution the smectic hydrodynamic variable  $u$  (displacement of the smectic layers) induces an effective non-negligible stress  $\sigma_{zz}^{(s)}$

$[\sigma_{zz}^{(s)} = \sigma_\rho \rho + \Phi_3(\partial u / \partial z)]$ , where  $\sigma_\rho$  and  $\Phi_3$  have been defined previously [13,22]. When the relaxator is introduced and when the slow dissipative hydrodynamic contributions are neglected, the equations of motion are written as

$$\begin{aligned} \dot{\rho} + \rho_0 \frac{\partial v_i}{\partial x_i} &= 0, \\ \rho_0 \dot{v}_i + \frac{\partial P}{\partial x_i} - \delta_{iz} \frac{\partial \sigma_{zz}^{(s)}}{\partial z} - b_{ij} \frac{\partial Q}{\partial x_j} &= 0, \\ \dot{u} - v_z &= 0, \\ \dot{R} + aQ - b_{ij} \frac{\partial v_i}{\partial x_j} &= 0. \end{aligned} \quad (7)$$

The propagative solutions do not differ from those concerning the nematics given in the preceding subsection. But now  $C_{13}$  and  $C_{33}$  show an additional frequency-independent term, respectively equal to  $-\sigma_\rho$  and  $\Phi_3 - 2\sigma_\rho$ , which preserves a nonzero anisotropy when  $\omega \rightarrow 0$ . Namely,

$$\begin{aligned} \omega \rightarrow 0: \quad C_{11} &= C_0, \quad C_{13} = C_0 - \sigma_\rho, \\ C_{33} &= C_0 - 2\sigma_\rho + \Phi_3, \\ \omega \rightarrow \infty: \quad C_{11} &= C_0(1 + x^2), \quad C_{13} = C_0(1 + xy) - \sigma_\rho, \\ C_{33} &= C_0(1 + y^2) - 2\sigma_\rho + \Phi_3. \end{aligned}$$

### III. COMPARISON WITH EXPERIMENTAL DATA

As pointed out in the Introduction, most of the published hypersonic data are well fitted using a tensor of elastic constants with three independent components. In the case of nematic liquid crystals, one easily calculates  $|y-x|$  and  $C_0$  from the obtained values of  $C_{11}$ ,  $C_{13}$ , and  $C_{33}$  as

$$\begin{aligned} C_0 &= C_{11} - \frac{(C_{11} - C_{13})^2}{C_{11} + C_{33} - 2C_{13}}, \\ |y-x| &= \sqrt{\frac{q_R}{C_0}} |b_{\parallel} - b_{\perp}| = \sqrt{\frac{C_{11} + C_{33} - 2C_{13}}{C_0}}, \end{aligned} \quad (8)$$

when we assumed that  $\omega\tau \gg 1$ .

Generally, when a compound does exist in both nematic and smectic phases only very weak anomalies are observed at the transition between these two phases, which indicates that the hydrodynamic contributions are not predominant. Thus, for the sake of simplicity, we shall neglect them in the following discussion which will be based on Eqs. (6) and (8).

When assuming that the general relaxation process lies between ultrasonic and hypersonic frequency ranges (as shown experimentally by Grammes *et al.* [11]) one understands some differences between the propagation velocities in these two regimes: at ultrasonic frequencies the velocity is isotropic and, as it clearly appears in Eq. (4), the velocities for  $\theta=0$  (wave vector parallel to the director) and  $\theta=\pi/2$  (wave vector perpendicular to the director) are obtained by adding positive different terms to the low-frequency veloc-

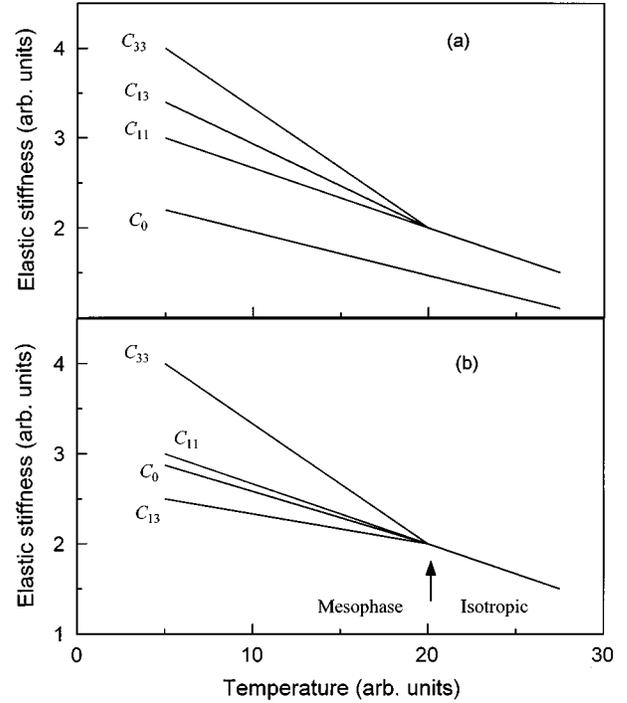


FIG. 1. Two possible types of behavior of the elastic constants in a one-relaxator model, assuming that the splitting at the mesophase-isotropic phase transition is continuous (as always observed). (a) First-type relaxator:  $x$  and  $y$  have the same signs; (b) second-type relaxator:  $x$  and  $y$  have opposite signs.

ity. This accounts for hardening and anisotropy. The difficulties arise when one tries to interpret this anisotropy in more detail.

Two qualitatively different behaviors are predicted by the model depending on the signs of  $x$  and  $y$ . If  $x$  and  $y$  have the same sign [Fig. 1(a)],  $C_0$  is smaller than  $C_{11}$ ,  $C_{13}$ , and  $C_{33}$  in the mesophase (i.e., the Brillouin and ultrasonic data can differ significantly) but, on the other hand,  $C_{13}$  has to lie between  $C_{11}$  and  $C_{33}$  [cf., Eq. (6)]. The larger the difference between ultrasonic and hypersonic data, the closer the value of  $C_{13}$  to its upper limit  $\sqrt{C_{11}C_{33}}$ . If  $x$  and  $y$  have opposite signs [Fig. 1(b)]  $C_{13}$  in the mesophase is smaller than both  $C_{11}$  and  $C_{33}$  but it is also inferior to  $C_0$ , which has to be equal to  $C(\theta_0) (\geq C_{13})$  as shown in the preceding section. If the splitting of the elastic constants is continuous at the isotropic-mesophase transition, then the difference between hypersonic and ultrasonic data should necessarily vanish in the isotropic phase. In the following, we will refer to the relaxators leading to temperature dependencies shown in Figs. 1(a) and 1(b) as the relaxators of first and second type, respectively.

Experimentally, in every case where both ultrasonic and hypersonic data exist for the same compound, the ultrasonic velocity is smaller than the hypersonic one in both isotropic and liquid crystalline phases, whatever the direction of propagation is. It means that  $x$  and  $y$  do have the same signs and, consequently, that  $C_{13}$  should lie between  $C_{11}$  and  $C_{33}$ , in contradiction with most of the experimental results.

Concerning the value of  $C_{13}$ , two situations are found in the experiments. (i)  $C_{13}$  is inferior both to  $C_{11}$  and to  $C_{33}$  as,

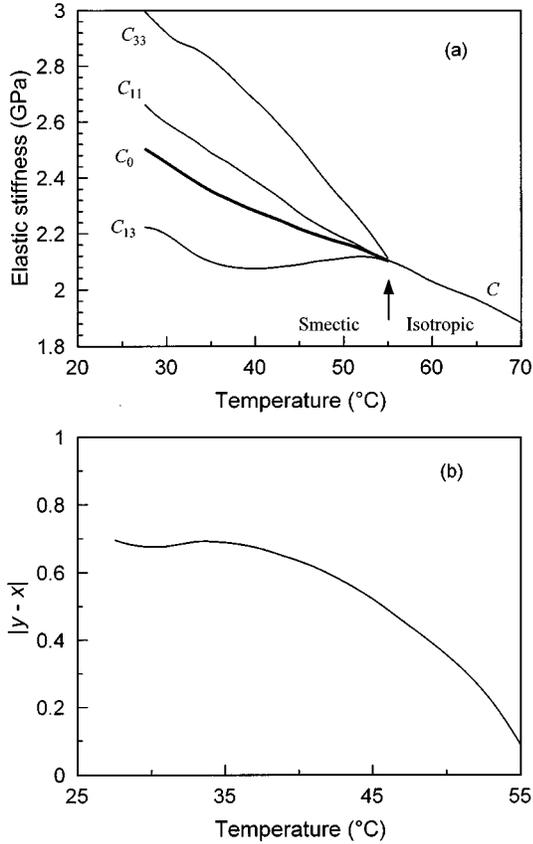


FIG. 2. (a) Experimental data taken in OBAMBCPE by Brillouin scattering;  $C_0$  is the calculated value of the low-frequency elastic constant using Eq. (8) in the one-relaxator model. (b) Calculated temperature dependencies of the parameter  $x$  and  $y$  (both tend to zero when the phase transition is approached from below).

for example found in CBOOA [9], in  $\beta$ -methyl butyl  $p$ [( $p$ -methoxy benzylidene) amino] cinnamate [7], in 8CB [23], and in OBAMBCPE [13]. Figure 2 illustrates an example of contradiction derived from our recently published Brillouin data concerning OBAMBCPE [13]; in this figure we show the variation of  $|y-x|$  versus temperature assuming that  $\omega\tau$  is significantly larger than one. In this case  $x$  and  $y$  should have opposite signs [see Eq. (6)] and tend to 0 at the mesophase-isotropic phase transition. Such a conclusion is not correct, since it would mean that the difference between the ultrasonic and hypersonic propagation velocities should vary with temperature and progressively vanish when the phase transition is approached; this is in contradiction with the admitted physical interpretation of the relaxator, namely, with the experimentally verified fact [11] that it is a nearly temperature-independent process. (ii)  $C_{13}$  is approximately equal to  $C_{11}$  (which in these cases is always smaller than  $C_{33}$ ), as found for example in 5CB and 6CB by Krüger *et al.* [12]. These authors find a large and temperature-independent difference between the elastic constants measured in ultrasonic and hypersonic frequency ranges. On the other hand,  $C_{13}$  remains practically equal to  $C_{11}$  below the isotropic-nematic transition, which means [cf., Eq. (6)] that either  $x=0$  or  $x=y$ . From  $C_{33} \neq C_{11}$  we deduce that  $x \neq y$  and then necessarily  $C_{11} = C_0$ , in contradiction with the large difference between ultrasonic and hypersonic measurements.

In conclusion, the model developed above fails to simultaneously account for the observed characteristics of the anisotropy in the hypersonic regime and the increase of the elastic constants compared to their values in the ultrasonic range. Note that if one supposes that  $\omega\tau$  is finite (but large enough to allow an anisotropy), the results of the discussion are not qualitatively modified and the above-mentioned difficulties remain.

#### IV. TWO-RELAXATOR MODEL

To overcome the preceding contradiction, we shall introduce into the system another relaxator described by the variable  $R'$  and the conjugate force  $Q'$ . The physical origin of this second relaxator will be discussed later. Obviously, it is not smart to introduce many unknown material constants and therefore, we shall simplify our model as much as possible. First, we neglect the dynamical coupling between two relaxators, i.e., the dissipative flux is proportional to the generalized force of the corresponding relaxator only. Also, we will neglect the cross derivatives in the expansions of  $Q$  and  $Q'$ , i.e., we put  $\partial Q/\partial R' = \partial Q'/\partial R = 0$ .

Since the form of the equation of motion for  $Q'$  is the same as that for  $Q$  we immediately get the following high-frequency results [compare to Eqs. (6)]:

$$C_{11} = C_0(1 + x^2 + x'^2),$$

$$C_{33} = C_0(1 + y^2 + y'^2),$$

$$C_{13} = C_0(1 + xy + x'y'), \quad (9)$$

where  $x'$  and  $y'$  are defined in the same way as  $x$  and  $y$ .

In these expressions there are enough independent parameters to fit the behavior of the elastic constants. However, the only possibility of describing experiments in a general way, i.e., without any accidental mutual compensations of temperature-dependent parameters, is to admit that the involved relaxators are of different types (discussed above)—more specifically, a relaxator of the first type [cf., Fig. 1(a)] characterized by the same signs of  $x$  and  $y$ , which is mainly responsible for the hardening of the elastic constants at hypersonic frequencies, and a relaxator of the second type [cf., Fig. 1(b)] characterized by the opposite signs of  $x'$  and  $y'$ , which is responsible for a significant hypersonic anisotropy in the mesophase. It should be emphasized that the relaxator of the second type becomes ineffective in the isotropic phase: it follows immediately from Eq. (9). The relaxator  $R$ , related to the dynamics of the population of excited vibrational state, is a general only slightly temperature-dependent process that is clearly present in ordinary organic liquids. No drastic changes in its characteristics near mesophase-isotropic phase transition can be expected. Therefore, we identify it with the relaxator of the first type. In addition, it is very likely that the anisotropy introduced by this kind of process is rather small ( $b_{\perp} \approx b_{\parallel}$ ).

Now the relaxator  $R'$  has to be necessarily of the second type. Since it becomes effective just at the isotropic-nematic (or isotropic-smectic) transition, it can be connected with the nematic order parameter  $S_{ij} = S(T)(n_i n_j - \frac{1}{3} \delta_{ij})$ . We have already mentioned that the hydrodynamic fluctuations of the director components  $n_i$  practically do not influence the

propagation of sound in nematics. However, one should also consider the fluctuations of magnitude  $S$  of the order parameter, which is not a hydrodynamic variable [24], and in fact we identify it with the second relaxator  $R'$ . Although far away from the isotropic-nematic transition the fluctuations of the nematic order parameter are usually discarded [20] because they relax on a microscopic time scale, we believe that near the phase transition they are slow enough to be effective at the hypersonic frequency range. It should be pointed out that just above the isotropic-nematic transition there exists a semislow motion of nematic clusters that gives rise to several temperature-dependent effects such as pretransitional phenomena. Since even at ultrasonic frequencies no important temperature dependence of elastic constants has been observed, this type of motion is already clamped and does not need to be considered explicitly.

The nematic order-parameter modulus as an additional nonhydrodynamic variable has already been introduced explicitly into macroscopic dynamic equations for the nematic liquid-crystalline side-chain polymer [20]. In this system, far away from any phase transition, the nematic order-parameter fluctuations are slowed down by the hindrance due to the backbone. Our system of macroscopic equations as well as

the derived sound dispersion relations are essentially the same as in Ref. [20], where only one nonhydrodynamic variable has been introduced and the explicit angular dependence of anisotropic properties is not presented.

## V. CONCLUSION

In order to describe qualitatively the anisotropic propagation of sound in liquid crystals as well as the increase of effective elastic constants at hypersonic frequencies, two nonhydrodynamic relaxation mechanisms should be considered in the conventional hydrodynamic treatment of sound propagation. The first mechanism, which is effective also in the isotropic phase, is connected to the energy transfer from kinetic to vibrational motion of molecules. The second mechanism, which is responsible mostly for the anisotropic sound propagation in the hypersonic frequency range, might be related to the fluctuations of the nematic order-parameter magnitude. Both of these mechanisms have been treated independently at different occasions by previous authors [11,15,20]. Our analysis suggests that they should be considered simultaneously.

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