Surface oscillations of smectic-*A* liquid crystals

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A theoretical description of free surface motion is presented for a semi-infinite unbounded smectic-*A* (Sm-*A*) liquid crystal. The characteristic equation is analyzed in a wide range of frequencies and wave numbers. It is shown that at low frequencies the surface eigenmotion is a wave of the second sound type. The wave velocity is twice that of the maximum second sound velocity in the bulk phase. This Rayleigh mode may be excited and detected as surface transverse waves. At high frequencies the surface eigenmotion is a damping viscous mode. The spectral intensities of the surface displacement fluctuations are presented in rather simple form for separate areas of wave numbers and frequencies. These results may be used to describe light scattering experiments. It is shown that the surface tension is negligible when describing Sm-*A* surface eigenmotions, in contrast to ordinary liquids. At the same time the surface tension is sufficient for surface displacement fluctuations at low frequencies.

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I. INTRODUCTION

Surfaces of liquid crystals are currently under intensive study both theoretically and experimentally. Considerable attention has been given to studying light and x-ray scattering, ellipsometry, mechanical oscillations of the surface, etc. [1]. Smectic liquid crystals attract special interest due to the possibility of producing freely suspended films ranging from several hundred down to two- on three-layer thickness.

While the static properties of smectic-*A* (Sm-*A*) films have been studied in detail [2–6], their dynamic properties are much less well understood. The theory of Sm-*A* film oscillations was developed in [7–10]. The influence of surface tension and elastic constant B on the spectrum of eigenfrequencies was studied in Ref. [7] within the framework of a model of discrete layers. This method was further developed for investigation of the dynamic behavior of the displacement and density autocorrelation functions in Sm-*A* films [8,9]. The dependences of relaxation times on layer sliding viscosity and surface tension were studied. The results derived were used for analyses of the experimental data obtained by the method of soft-x-ray photon spectroscopy. The problem of Sm-*A* surface oscillations in a sample of infinite thickness was studied in [11]. In this work the spectral intensities of the thermal surface fluctuations were considered for a slowly damping mode. In Ref. [10] freely suspended Sm-*A* films of finite thickness are analyzed in the framework of a continuous model, taking into account the mutual influence of the surfaces. In particular, two surface dynamic modes, undulation and a peristaltic mode, were found. It was shown that a light scattering experiment may be used for the determination of viscosity and elastic coefficients.

Recently, the surface motions have been investigated very intensively in concentrated polymer solutions and soft gels [12–15]. These systems manifest solidlike and liquidlike properties in various conditions as well as Sm-*A* behavior. The most interesting surface motions are in the region of low wave numbers and low frequencies. In recent years the surface hydrodynamics of these materials have been made experimentally accessible thanks to surface laser light scattering and excited surface wave techniques [16]. That is, by these methods the crossover from ordinary capillary wave to Rayleigh elastic mode was observed [12–15]. As far as we know the analogous experiments for Sm-*A* systems have not been carried out, though similar effects may be expected for Sm-*A* surfaces.

In the present work the problem of the existence of surface eigenmotions is analyzed in a wide range of frequencies and wave vectors. The spectral intensities of thermal surface fluctuations are calculated also. The work is organized as follows. In Sec. II all necessary equations and boundary conditions are presented. In Sec. III the problem of the surface eigenmotions is solved based on analysis of the bulk and surface characteristic equations. In Sec. IV the spectral intensities of the thermal surface fluctuations are calculated. A discussion of the results obtained is presented in conclusion.

II. EQUATIONS OF MOTION

Smectic-*A* surface oscillations are considered in the same way as for an isotropic viscous liquid [17]. The free surface is supposed boundless, and a solution of the equations of motion will be derived in the form of plane waves, undergoing damping with depth in a liquid crystal. The general solution and the character of decay of the waves will be found from the equations of motion while the characteristic equation and the wave amplitudes will be obtained from the boundary conditions.

In equilibrium the free surface of a Sm-*A* liquid crystal is flat, and the smectic layers are located parallel to the surface. We use the Cartesian coordinate frame so that the plane xy coincides with the equilibrium position of the free surface; the equilibrium smectic occupies the half space $z \leqslant 0$. We suppose that the amplitude of surface displacement ζ is much less than the wavelength. We represent the solution of the equations of motion as plane waves propagating along the x direction so that along the y axis the system is homogeneous. In this case the density of elastic free energy is given by

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$$F=\frac{1}{2}B\left(\frac{\partial u}{\partial z}\right)^2+\frac{1}{2}K\left(\frac{\partial^2 u}{\partial x^2}\right)^2, \qquad (2.1)$$

where u is the layer displacement from the equilibrium position, and B and K are the layer elastic moduli.

In what follows we suppose the liquid crystal to be incompressible, i.e., the considered velocities of motion are much less than the sound velocity c, and for the circular frequency ω and wave number q the inequality $\omega \ll cq$ is valid. In this approximation the system satisfies the following equations of motion [18–20]:

$$\frac{\partial v_x}{\partial x}+\frac{\partial v_z}{\partial z}=0,$$

$$\rho\frac{\partial v_i}{\partial t}=-\partial_i p+\partial j\sigma'_{ij}+h\delta_{iz}, \quad i=x,z, \qquad (2.2)$$

$$\frac{\partial u}{\partial t}=v_z+\lambda_p h.$$

Here v_x, v_z, and p are the velocity components and pressure, respectively, σ'_{ij} is the viscous stress tensor, λ_p is the permeation constant, and h is expressed as [18–20]

$$h=B\frac{\partial^2 u}{\partial z^2}-K\left(\frac{\partial^2}{\partial x^2}\right)^2 u. \qquad (2.3)$$

In Eq. (2.2) we sum over repeated indices, and the notations $\partial_x \equiv \partial/\partial x$ and $\partial_z \equiv \partial/\partial z$ are used.

A set of equations of motion should be supplemented by boundary conditions. The surface waves have to vanish as $z\rightarrow -\infty$,

$$\lim_{z\rightarrow -\infty} v_{x,z}=0. \qquad (2.4)$$

The tangential component of the stress tensor should be equal to zero,

$$\sigma_{xz}=0, \qquad (2.5)$$

and the jump of the normal component of the stress tensor is compensated by the capillary pressure on the surface,

$$\sigma_{zz}-\sigma_{zz}^{ext}-\gamma\frac{\partial^2\zeta}{\partial x^2}=0. \qquad (2.6)$$

Here γ is the surface tension, and σ_{ij} is the stress tensor,

$$\sigma_{ij}=-p\,\delta_{ij}+\sigma'_{ij}+\sigma^r_{ij}.$$

The viscous stress tensor in incompressible Sm-A has the form [18]

$$\sigma'_{ij}=2\eta_2 v_{ij}+2(\eta_3-\eta_2)(v_{iz}\delta_{jz}+v_{jz}\delta_{iz})+\eta' v_{zz}\delta_{iz}\delta_{jz},$$

where $v_{ij}=\frac{1}{2}(\partial_i v_j+\partial_j v_i)$, and $\eta'=\eta_1+\eta_2-4\eta_3-2\eta_5+\eta_4$ [10,18]. The "reactive part" of the stress tensor, σ^r_{ij}, caused by the inhomogeneous displacement of the smectic layers, has the following components occurring in the boundary conditions [20]:

$$\sigma^r_{zz}=B\frac{\partial u}{\partial z},$$

$$\sigma^r_{xz}=-K\frac{\partial^3 u}{\partial x^3}.$$

Above the surface of the Sm-A liquid crystal the stress tensor is determined only by the external pressure,

$$\sigma^{ext}_{ij}=-p_{ext}\delta_{ij}.$$

The last boundary condition is the impermeability of the surface,

$$\frac{\partial\zeta}{\partial t}=v_z. \qquad (2.7)$$

The equations of motion (2.2) and the boundary conditions (2.4)–(2.7) determine the surface motions in Sm-A liquid crystals.

We shall obtain the solution of the set of equations of motion (2.2) in the form of plane waves where the dependence on space coordinates and time is determined by a factor $\exp(q_z z+iqx-i\omega t)$, where the q value is supposed to be real and positive. In this case we get a system of algebraic equations for the amplitudes:

$$iqv_x+q_z v_z=0,$$

$$[i\omega\rho+\eta_3 q_z^2-(2\eta_2-\eta_3)q^2]v_x-iqp=0,$$

$$[i\omega\rho+(\eta'-2\eta_2+3\eta_3)q_z^2-\eta_3 q^2]v_z + (Bq_z^2-Kq^4)u-q_z p=0,$$

$$v_z+[i\omega+\lambda_p(Bq_z^2-Kq^4)]u=0. \qquad (2.8)$$

For determination of the Sm-A surface eigenmotions we put the determinant of the system (2.8) equal to zero. Thus we obtain the bulk characteristic equation, connecting q_z, q, and ω. It is convenient to introduce the dimensionless variable $S=q_z/q$ and to present the bulk characteristic equation as

$$S^6+A(q,\omega)S^4+B(q,\omega)S^2+C(q,\omega)=0, \qquad (2.9)$$

where

$$A(q,\omega)=i\omega\tau_p-\left(2+\frac{\eta'}{\eta_3}\right),$$

$$B(q,\omega)=\frac{\tau_p}{\tau_M}-i\omega\tau_p\left(2+\frac{\eta'}{\eta_3}\right)-\frac{\omega^2\tau_p}{c_2^2q^2\tau_M}, \qquad (2.10)$$

$$C(q,\omega)=-\frac{\tau_p}{\tau_M}(\lambda q)^2+i\omega\tau_p+\frac{\omega^2\tau_p}{c_2^2q^2\tau_M}.$$

Here $\tau_p=1/\lambda_p Bq^2$ is the permeation characteristic time, $\tau_M=\eta_3/B$ is the Maxwell relaxation time, $c_2=\sqrt{B/\rho}$ is the second sound velocity, and $\lambda=\sqrt{K/B}$ is the characteristic length of a smectic structure.

Equations (2.9) and (2.10) are obtained under conditions $\lambda q \ll 1$ and $\tau_v \ll \tau_p$. The first inequality means that the inhomogeneity length is much greater than the interlayer distances. The second condition means that the shear wave relaxation time $\tau_v=\rho/\eta_3 q^2$ is much smaller than the permeation characteristic time. Using typical numerical values of Sm-*A* material parameters [19] $K \sim 10^{-6}$ dyn, $B \sim 2\times 10^7$ dyn/cm^2, $\lambda_p \sim 10^{-14}$ cm^4/dyn s, $\rho \sim 1$ G/cm^3, $\eta_3 \sim 10$ Pz, we get $q \ll 5\times 10^6$ cm^{-1} from the first condition. As for the condition $\tau_v \ll \tau_p$, it is essentially always satisfied.

It is convenient to consider Eq.(2.9) as the equation determining S as a function of q and ω. This equation has three solutions $S_k(q,\omega)$, $k=1,2,3$, with positive real parts, providing the damping of surface waves with depth, $z\to -\infty$. Using the notation u_1,u_2, and u_3 for the amplitudes of the corresponding displacements we get the general solution of the system (2.8) in the form

$$u=\exp(iqx-i\omega t)\sum_{k=1}^{3} u_k \exp(S_k qz),$$

$$v_x=\exp(iqx-i\omega t)\sum_{k=1}^{3} (\omega - i\Lambda_k)S_k u_k \exp(S_k qz),$$

$$v_z=\exp(iqx-i\omega t)\sum_{k=1}^{3} (-i\omega-\Lambda_k)u_k \exp(S_k qz),$$

$$p=\exp(iqx-i\omega t)\sum_{k=1}^{3} \{i\omega\rho+[\eta_3(S_k^2+1)-2\eta_2]q^2\}$$
$$\times(-i\omega-\Lambda_k)\frac{S_k}{q}u_k \exp(S_k qz), \qquad (2.11)$$

where

$$\Lambda_k=\tau_p^{-1}[S_k^2-(\lambda q)^2].$$

The amplitudes u_1,u_2,u_3 can be obtained from the boundary conditions (2.5)–(2.7) which can be referred to the plane $z=0$ due to the smallness of the oscillations. After substitution of the general solution (2.11) into the boundary conditions we obtain a set of equations for the amplitudes u_1, u_2, and u_3:

$$\sum_{k=1}^{3} [\eta_3(\omega-i\Lambda_k)(S_k^2+1)+iKq^2]qu_k=0, \qquad (2.12)$$

$$\sum_{k=1}^{3}\left(BS_kq+\gamma q^2-[\omega\rho+i\eta_3(3-S_k^2)q^2+i\eta' q^2]\right.$$
$$\left.\times(\omega-i\Lambda_k)\frac{S_k}{q}\right)u_k=-p_{ext}(q,\omega), \qquad (2.13)$$

$$\sum_{k=1}^{3}\Lambda_k u_k=0. \qquad (2.14)$$

Subsequently the function $-p_{ext}(q,\omega)$ will be used as an external force for the calculation of the response function. In order to find the Sm-*A* surface eigenmotions we suppose the external pressure to be equal to zero, $p_{ext}=0$.

For the existence of a nonzero solution of Eqs. (2.12)–(2.14) when $p_{ext}=0$, the system determinant should be equal to zero. This condition is the surface characteristic equation connecting ω with q. In general this equation is very complicated, but it can be simplified using the smallness of the permeation constant λ_p. It should be noted that contributions of various modes to the total displacement u are nonequivalent. First of all we are interested in weakly damped modes, $0<\mathrm{Re}S_k\ll 1$, which have the largest amplitudes [10]. Because of the smallness of the permeation constant λ_p the inequalities $|A(q,\omega)|$, $|B(q,\omega)|$, $|C(q,\omega)|\gg 1$ are valid essentially always. Hence the solution of the bulk characteristic equation Eq. (2.9), which is determined by the permeation $S_3^2\approx -i\omega\tau_p$, satisfies the inequality $|S_3^2|\gg 1$. The rest solutions $|S_1|,|S_2|\ll|S_3|$ may be found from the equation

$$i\omega\tau_M S^4+\left[1-i\omega\tau_M\left(2+\frac{\eta'}{\eta_3}\right)-\frac{\omega^2}{c_2^2q^2}\right]S^2$$
$$-\left(\lambda^2q^2-i\omega\tau_M-\frac{\omega^2}{c_2^2q^2}\right)=0. \qquad (2.15)$$

Then, as was pointed out in [10,11], from the boundary conditions (2.12) and (2.14) it follows that $|u_3|\ll|u_1|,|u_2|$. Therefore we neglect the permeation $\tau_p^{-1}=0$, and we can sum over two indices in the general solution (2.11) and in the boundary conditions (2.12)–(2.14), which transforms them to the equations

$$\sum_{k=1}^{2}[\eta_3\omega(S_k^2+1)+iKq^2]qu_k=0, \qquad (2.16)$$

$$\sum_{k=1}^{2}\left(BS_kq+\gamma q^2-[\omega\rho+i\eta_3(3-S_k^2)q^2+i\eta' q^2]\frac{\omega S_k}{q}\right)u_k$$
$$=0. \qquad (2.17)$$

Equations (2.16) and (2.17) are the boundary conditions for the tangential and normal components of the stress tensor, respectively. The surface impermeability condition (2.14) is fulfilled automatically.

III. ANALYSIS OF CHARACTERISTIC EQUATIONS

To obtain the surface eigenfrequencies ω and the rate of damping S for a given wave number q it is necessary to solve a system including bulk [Eq. (2.15)] and surface characteristic equations that is obtained by equating to zero the determinant of the system (2.16),(2.17). The factors of powers of S in Eq. (2.15) have rather complicated dependences on ω and q. In practice the problem is in determining the characteristic curves in the plane $(|\omega|,q)$ that describe the various types of eigenmotions. The general solution of this problem is rather cumbersome. Therefore it is convenient to separate the plane $(|\omega|,q)$ into regions where it is possible to simplify Eq. (2.15). These regions are presented in Fig. 1.

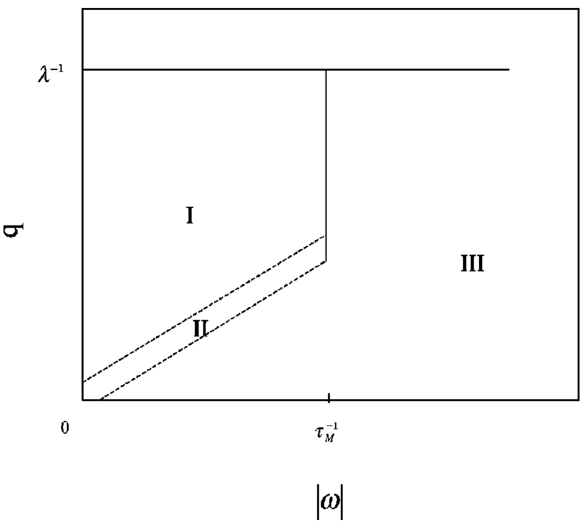

FIG. 1. The ranges in the $|\omega|,q$ plane suitable for analyses of the surface eigenmotions. I, Eq. (3.1); II, Eq. (3.5); III, Eqs. (3.12) and (3.13).

A. Region I

First, we investigate the low-frequency area which is defined by the inequalities

$$|\omega| \ll \frac{1}{\tau_M},$$

$$|\omega| \ll c_2 q. \qquad (3.1)$$

In this area there is only one solution of the bulk characteristic equation (2.15) satisfying the condition $\mathrm{Re}\, S_1 \ll 1$,

$$S_1 = \left((\lambda q)^2 - i\omega\tau_M - \frac{\omega^2}{c_2^2 q^2} \right)^{1/2}, \quad \mathrm{Re}\, S_1 > 0. \qquad (3.2)$$

The second solution may be found from the equality

$$S_2^2 \approx \frac{i}{\omega \tau_M}.$$

This solution obeys the inequality $|S_2^2| \gg 1$. It follows from the tangential boundary condition (2.16) that this mode has a very small amplitude. Thus this mode can be neglected in the boundary condition (2.17) for the normal component of the stress tensor [10,11], and Eqs. (2.16) and (2.17) may have a nonzero solution only when the surface characteristic equation

$$B S_1 q + \gamma q^2 - \frac{S_1}{q}\omega(\omega\rho + 3i\eta_3 q^2 + i\eta' q^2) = 0 \qquad (3.3)$$

is valid.

As follows from conditions (3.1), we may neglect the last term in Eq. (3.3) and it transforms to

$$S_1 = -\frac{\gamma q}{B}, \qquad (3.4)$$

where S_1 is given by Eq. (3.2). Obviously, Eq. (3.4) is insoluble because of the negativity on its right-hand side. Thus the surface eigenmotions are absent in region I.

B. Region II

The region II is defined by the conditions

$$\omega^2 \approx c_2^2 q^2, \quad |\omega| \ll \frac{1}{\tau_M}. \qquad (3.5)$$

Its width is determined by the inequality

$$\left| 1 - \frac{\omega^2}{c_2^2 q^2} \right| \ll |\omega| \tau_M .$$

In this area the bulk characteristic equation has the form

$$S^4 = \frac{i\omega\rho}{\eta_3 q^2}. \qquad (3.6)$$

Both solutions of this equation with positive real parts obey the relation $\mathrm{Re}\, S_{1,2} > 1$.

The surface characteristic equation follows from boundary conditions (2.16) and (2.17). Since in Sm-A liquid crystals inequality $K\rho/\eta_3^2 \ll 1$ is fulfilled it is possible to neglect the term with K in Eq. (2.16). Moreover we can put $\gamma = 0$ in Eq. (2.17) as long as in this area the condition $\gamma q/B \ll 1$ is valid. Thus we have

$$(S_1^2 + 1)u_1 + (S_2^2 + 1)u_2 = 0, \qquad (3.7)$$

$$\left(Bq - \frac{\omega\rho^2}{q} \right)(S_1 u_1 + S_2 u_2) = 0. \qquad (3.8)$$

The determinant of the system (3.7) and (3.8) will be equal to zero for

$$\omega = \pm c_2 q. \qquad (3.9)$$

This means the possibility of existence in Sm-A liquid crystal of a propagating surface Rayleigh wave of the second sound type. This surface wave is formed by two modes whose damping width depth is determined by the factor $\exp(S_{1,2} q z)$, where

$$S_{1,2} = \sqrt[4]{\frac{\omega\rho}{\eta_3 q^2}}\, \frac{\sqrt{2 \mp \sqrt{2}} \mp i\sqrt{2 \pm \sqrt{2}}}{2}. \qquad (3.10)$$

The amplitudes of these modes are connected by condition (3.7).

Taking into account the wave attenuation we obtain the characteristic frequency

$$\omega = \pm c_2 q - i\omega'', \quad \omega'' = \sqrt{\frac{\eta_3 c_2}{8\rho}}\, q^{3/2}. \qquad (3.11)$$

Thus, surface eigenwaves of the second sound type can be detected in Sm-A liquid crystals under conditions

$$\omega \ll \frac{1}{\tau_M} \approx 2\times 10^6 \ \ \mathrm{sec}^{-1},$$

$$q \ll \frac{1}{c_2\tau_M} \approx 4\times 10^2 \ \ \mathrm{cm}^{-1}.$$

It is necessary to note that second sound cannot propagate in bulk along smectic planes because the interlayer distances are invariable in this transverse wave, contrary to the surface Rayleigh wave obtained.

C. Region III

The region III is restricted by fulfillment of either one of inequalities

$$|\omega| \gg \frac{1}{\tau_M}, \tag{3.12}$$

or

$$|\omega| \gg c_2 q. \tag{3.13}$$

Here the bulk characteristic equation has almost the same form as for an isotropic liquid [17],

$$S^4 - \left(2 + \frac{\eta'}{\eta_3} - i\frac{\omega\rho}{\eta_3 q^2}\right)S^2 + 1 - i\frac{\omega\rho}{\eta_3 q^2} = 0. \tag{3.14}$$

The difference is the presence of the viscosity coefficient η', which equals zero in ordinary liquids. This equation has two solutions with $\mathrm{Re}\, S_{1,2} > 0$, $|S_{1,2}| \gg 1$. The boundary conditions on a surface lead to the system of equations

$$\sum_{k=1,2}\left(Bq - \frac{\omega}{q}[\omega\rho + i\eta_3(3 - S_k^2)q^2 + i\eta' q^2]\right)S_k u_k = 0, \tag{3.15}$$

and Eq. (3.7). In Sm-A [21,22] the viscosities obey the inequality $\eta' \ll \eta_3$, and we neglect η' for simplicity. In this case the system (3.7), (3.15) may be easily solved. The roots of Eq. (3.14) are known for the similar problem of surface oscillations in an isotropic liquid [17]:

$$S_1 = 1,$$

$$S_2 = \left(1 - i\frac{\omega\rho}{\eta_3 q^2}\right)^{1/2}. \tag{3.16}$$

By equating to zero the determinant of the system (3.7), (3.15) we get the surface characteristic equation

$$\left(2 - i\frac{\omega}{c_2^2\tau_M q^2}\right)\left(1 - \frac{\omega^2}{c_2^2 q^2} - 2i\omega\tau_M\right) = 2\sqrt{1 - i\frac{\omega}{c_2^2\tau_M q^2}}(1 - 2i\omega\tau_M). \tag{3.17}$$

In the area of ω, q satisfying the condition

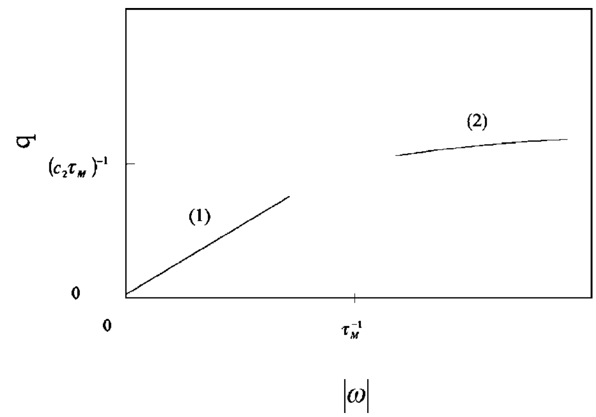

FIG. 2. Solutions of characteristic equation: (1), $\omega = c_2 q$ in region II; (2), $\omega = -i4\eta_3 q^2/3\rho$ in region III.

$$\frac{|\omega|}{c_2^2\tau_M q^2} \ll 1,$$

we may replace the square root in Eq. (3.17) by the first three terms of a power series expansion. In this case the characteristic equation is reduced to a linear one with a solution of the diffusion type,

$$\omega = -i\frac{4\eta_3 q^2}{3\rho}. \tag{3.18}$$

The parameter of the expansion appears not to be small, but accounting for one more term results in only small corrections of numerical factors.

For

$$\frac{|\omega|}{c_2^2\tau_M q^2} \gg 1,$$

Eq. (3.17) has no solutions (see the Appendix). In the area

$$\frac{|\omega|^2}{c_2^2 q^2} \sim |\omega|\tau_M \sim 1$$

the surface characteristic equation can be solved only numerically.

Figure 2 shows the dependence of q on $|\omega|$ obtained from solution of the characteristic equations.

IV. SURFACE FLUCTUATIONS

Surface laser light scattering is the most effective method for studying surface motions [16]. By this method it is possible to measure a power spectrum of thermal fluctuations for low wave numbers, $q \sim 10^2$ cm^{-1}, [12–15]. The spectral intensity, or power spectrum, of surface displacement fluctuations $\langle|\zeta_{\mathbf{q}}|^2\rangle_\omega$ may be obtained using the fluctuation-dissipation theorem [23]

$$\langle|\zeta_{\mathbf{q}}|^2\rangle_\omega = \frac{2k_BT}{\omega}\,\mathrm{Im}\,\chi(\mathbf{q},\omega), \tag{4.1}$$

where the response function $\chi(\mathbf{q},\omega)$ is determined by the relation

$$\zeta = -\chi(\mathbf{q},\omega)p^{ext}. \tag{4.2}$$

Here the frequency ω is real.

The general expression for the response function is rather cumbersome. Therefore we consider expressions for the spectral intensity of surface fluctuations for several regions of q and ω only.

A. Region I

In area I defined by conditions (3.1), together with the inequality $q\lambda \ll 1$, we select three areas Ia, Ib and Ic as shown in Fig. 3. These areas are determined by the following conditions:

$$\omega \ll \frac{Kq^2}{\eta_3}, \qquad \mathrm{Ia},$$

$$\frac{Kq^2}{\eta_3} \ll \omega \ll \frac{\eta_3 q^2}{\rho}, \qquad \mathrm{Ib}, \tag{4.3}$$

$$\frac{\eta_3 q^2}{\rho} \ll \omega \ll c_2 q, \quad \mathrm{Ic}.$$

In area Ia we can obtain from Eq. (3.2)

$$S_1 \approx q\lambda\left(1 - i\frac{\omega\tau_M}{2(q\lambda)^2}\right).$$

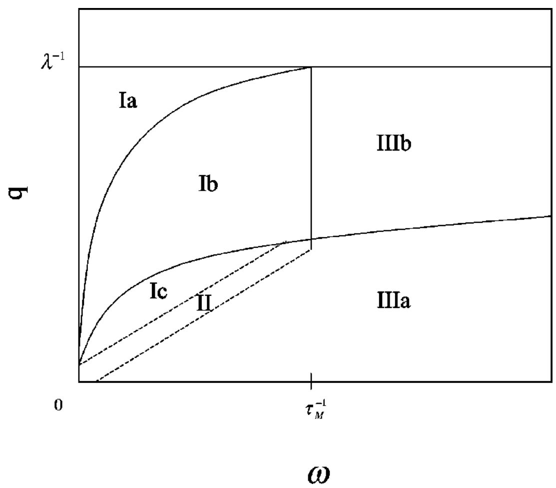

FIG. 3. Regions where the spectral intensities of fluctuations may be written in closed form: Ia, Ib, Ic, Eqs. (4.3); II, Eq. (3.5); IIIa, IIIb, Eqs. (4.8) and (4.10).

Hence, the spectral intensity of surface displacement fluctuations is

$$\langle|\zeta_{\mathbf{q}}|^2\rangle_\omega \approx \frac{4k_BT\lambda}{\eta_3(\omega^2 + 4(K+\gamma\lambda)^2 q^4/\eta_3^2)}. \tag{4.4}$$

In area Ib we get from Eq. (3.2)

$$S_1 \approx (1-i)\sqrt{\frac{\omega\tau_M}{2}}\left(1 + i\frac{(\lambda q)^2}{2\omega\tau_M}\right),$$

$$\langle|\zeta_{\mathbf{q}}|^2\rangle_\omega \approx \frac{2\sqrt{2}k_BT}{\sqrt{B\eta_3}q\omega^{3/2}} \times \frac{1-(\lambda q)^2/2\omega\tau_M}{[1+\sqrt{2\omega\tau_M}\gamma q/\eta_3\omega + (\lambda q)^2/2\omega\tau_M]^2 + [1-(\lambda q)^2/2\omega\tau_M]^2}. \tag{4.5}$$

In area Ic the damping of the surface wave is determined by the factor

$$S_1 = \frac{\tau_M c_2 q}{2} - i\frac{\omega}{c_2 q},$$

where $|\mathrm{Im}\, S_1| \gg \mathrm{Re}\, S_1$. For $\langle|\zeta_{\mathbf{q}}|^2\rangle_\omega$ we obtain in this area

$$\langle|\zeta_{\mathbf{q}}|^2\rangle_\omega = \frac{2k_BT}{\sqrt{B\rho}(\omega^2 + \eta_3^2 q^4/4\rho^2)}. \tag{4.6}$$

B. Region II

In region II, restricted by conditions (3.5), we get

$$\langle|\zeta_{\mathbf{q}}|^2\rangle_\omega = k_BT\left(\frac{\eta_3 q}{c_2^5\rho^5}\right)^{1/4}\frac{a_1(c_2^2q^2-\omega^2)+a_2\omega\omega''}{(\omega^2-c_2^2q^2)^2+4\omega^2\omega''^2}, \tag{4.7}$$

where

$$a_{1,2} = \sqrt{2+\sqrt{2}} \mp \sqrt{2-\sqrt{2}},$$

and ω'' is given by Eq. (3.11). The expression (4.7) for the spectral intensity of fluctuations is valid for low frequencies $|\omega| \ll \tau_M^{-1}$ and in a rather narrow region only,

$$|\omega - c_2 q| < \omega''.$$

C. Region III

In the area IIIa determined by the inequalities

$$\omega\tau_M \ll \frac{\omega^2}{c_2^2 q^2},$$

$$\frac{\omega}{c_2 q} \gg 1, \tag{4.8}$$

it is possible to omit the mode with damping factor S_2, Eq. (3.16). In this case we obtain

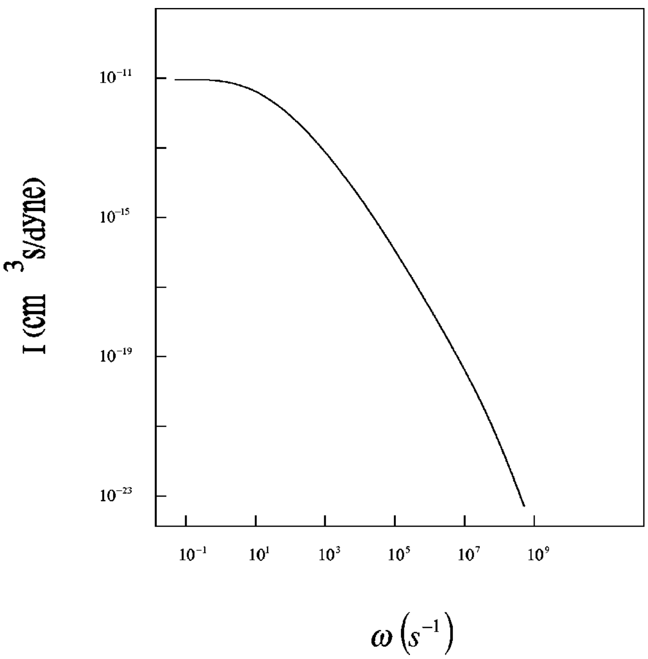

FIG. 4. The frequency dependence of the spectral intensity of surface fluctuations $I=\langle|\zeta_{\mathbf{q}}|^2\rangle_\omega/k_BT$ calculated by Eqs. (4.4), (4.5), and (4.11) for $q=10^4\ \mathrm{cm}^{-1}$.

$$\langle|\zeta_{\mathbf{q}}|^2\rangle_\omega=\frac{4k_BT\eta_3q^3}{\rho^2\omega^4}. \tag{4.9}$$

In the area IIIb determined by the conditions

$$\omega\tau_M\gg 1,$$

$$\omega\tau_M\gg\frac{\omega^2}{c_2^2q^2}, \tag{4.10}$$

it is necessary to keep both modes in Eq. (3.16). Thus, we have

$$\langle|\zeta_{\mathbf{q}}|^2\rangle_\omega=\frac{4k_BTq\Gamma(q)}{3\rho\omega^2[\omega^2+\Gamma^2(q)]}, \tag{4.11}$$

where

$$\Gamma(q)=\frac{1+8c_2^2\tau_M^2q^2}{6\tau_M}.$$

The typical spectral intensities of surface displacement fluctuations are shown in Figs. 4 and 5 as functions of frequency. For wave numbers obeying the inequality $q\gg 1/c_2\tau_M$ there is only one peak, $\omega=0$, which corresponds to the overdamped regime (Fig. 4). For $q\ll 1/c_2\tau_M$ an additional peak appears at $\omega\approx c_2q$ corresponding to the elastic regime (Fig. 5). As q increases the peak moves to higher frequency, accompanied by a considerable spectral broadening. This additional peak is much smaller than the peak at $\omega=0$.

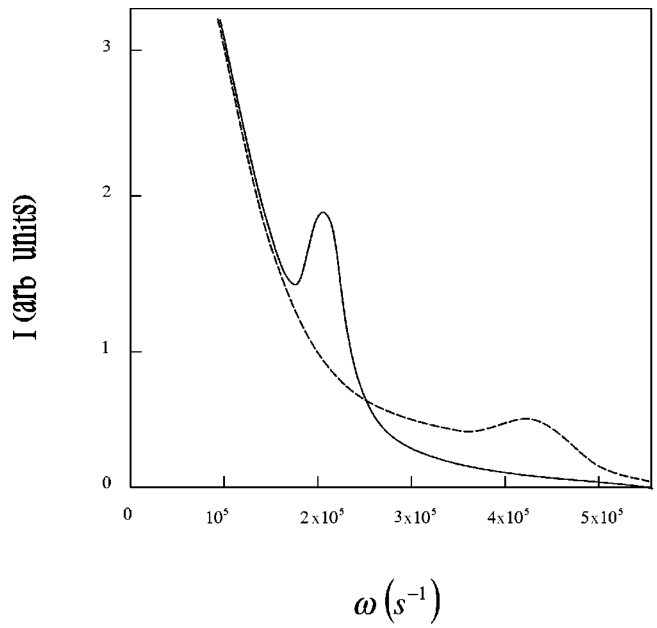

FIG. 5. The frequency dependence of I calculated by Eqs. (4.6), (4.7), and (4.9) for $q=50\ \mathrm{cm}^{-1}$ (solid line); $q=100\ \mathrm{cm}^{-1}$ (dashed line).

V. DISCUSSION

In this paper we analyzed the characteristic equation and showed that at low frequencies $|\omega|\ll\tau_M^{-1}$ the Sm-*A* surface eigenmotions are propagating waves of the second sound type. These Rayleigh waves are caused by the elastic forces arising from inhomogeneous undulation. Surface tension does not affect the surface eigenmotions, in contrast to the case of Sm-*A* thin films [7]. The character of propagation and decay of these waves is described by Eqs. (3.9)–(3.11).

As far as we know these elastic waves have not yet been observed experimentally at the Sm-*A* surface. At the same time, Rayleigh modes have been registered at the surfaces of polymer solutions [12–14] and soft gels [15]. In these systems the crossover from capillary to elastic waves was observed.

At high frequencies $|\omega|\gg\tau_M^{-1}$, the eigenmotions represent damping shear waves similar to waves in isotropic viscous liquids. For these waves the bulk attenuation coefficient is of the same order as the wave number of the surface wave [17]. However, there is an essential difference between the eigenfrequencies of surface motions in Sm-*A* liquid crystals and in viscous isotropic liquids [17]. It is connected with the considerable difference between the boundary condition for the normal component of the stress tensor in Sm-*A* liquid crystals and isotropic liquids. This occurs because Sm-*A* elastic properties are mainly determined by the coefficient *B*. We may neglect surface tension for Sm-*A* liquid crystals because of the validity of the inequality $\gamma q/B\ll 1$. One would expect the surface tension to be important in the vicinity of the transition to the nematic phase T_{NA}, where the elastic modulus *B* tends to zero. But this decrease is rather slow [24], namely, $B\sim\tau^\phi$, where $\tau=(T_{NA}-T)/T_{NA}$, $\phi\sim 0.3$–0.4. Therefore the surface tension is important in an extremely narrow thermal interval.

Low frequency motions may be investigated by mechanical excitation of surface waves in such a way as was done for soft gels [15]. It would be interesting to measure the surface second sound velocity which according to Eq. (3.9) should be twice the maximum velocity of second sound in the bulk.

The calculated spectral intensities of surface displacement fluctuations characterize thermal Sm-*A* surface oscillations. The results obtained are suitable for description of the light scattering spectrum in optical experiments. A contour of Lorentz type should be exhibited for low frequencies in accordance with Eq. (4.4). In the intermediate region of frequencies the form of the contour, Eq. (4.5), is sensitive to surface tension, in contrast to the surface eigenmotion spectrum. For higher frequencies the spectral intensities of surface fluctuations, Eqs. (4.9)–(4.11), decrease more rapidly, i.e., $\sim\omega^{-4}$.

APPENDIX

Under the condition

$$|\omega|\gg c_2^2\tau_M q^2, \tag{A1}$$

Eq. (3.17) reduces to

$$\sqrt{-i\frac{\omega}{c_2^2\tau_M q^2}}\frac{\omega}{c_2^2 q^2}=4i\omega\tau_M-2. \tag{A2}$$

For $|\omega|\ll\tau_M^{-1}$ Eq. (A2) has no solutions obeying the inequalities (A1) and $|\omega|\gg c_2 q$.

When $|\omega|\gg\tau_M^{-1}$, Eq. (A2) transforms to

$$\left(\frac{\omega}{c_2^2\tau_M q^2}\right)^{3/2}=-4\sqrt{-i}. \tag{A3}$$

The solution of Eq. (A3) contradicts Eq. (A1). Thus we may conclude that the surface characteristic equation (3.17) has no solutions under condition (A1).

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