Dynamics of free-standing smectic-*A* films

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The eigenvibrations and time-dependent layer displacement-layer displacement correlation functions are analyzed in a free-standing thin smectic-*A* films with the help of a discrete layer model. The film motions are described using the Chebyshev polynomials of second kind, $U_n(x)$. The eigenfrequencies problem is essentially simplified within the framework of this approach since the numerical solution of the high degree algebraic characteristic equation is replaced by the analytical solution of a rather simple trigonometrical equation. The dependences of eigenmodes on wave number $q_\perp$ were analyzed. For small $q_\perp$ one mode is a low frequency acoustic wave and other modes are high frequency optical oscillations. As the wave number $q_\perp$ increases all modes successively turn into relaxation when starting with the acoustic mode. The rather simple expression for susceptibility matrix and for spectral densities of layer displacement correlation functions were obtained using the Chebyshev polynomials. It was shown that the frequency dependences of spectral densities are sensitive to wave number $q_\perp$. For small $q_\perp$ the spectral densities of displacement-displacement correlation functions have a sharp peak and for large $q_\perp$ they turn into a contours of Lorentzian type.

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I. INTRODUCTION

In recent years considerable interest has been aimed at studying free-suspended smectic-*A* (Sm-*A*) films both theoretically and experimentally [1–13]. There are the continual and discrete approaches for the theoretical description of static and dynamic properties of Sm-*A* films. In the first model Sm-*A* films are considered as a continuous media [4–6,10–12]. This description is suitable for specimens with thickness much greater than the interlayer distance. The discrete model [1–3,7,8] is effectively used for films containing a few layers. Within the framework of this model it was possible to construct the equations of motion, to calculate the spatial correlation function of layer displacements, to describe the x-ray scattering, to estimate the relaxation times of the layer motions, etc.

Smectic-*A* films have been successfully investigated by light scattering [14] and x-ray scattering experiments [6,7,9,13]. The equilibrium structure of free-standing films have been studied in [6]. Recently the dynamic properties of films were investigated by coherent soft-x-ray [7,9] and hard-x-ray [13] dynamic scattering. For the description of experiments it is necessary to know the time-dependent layer displacement-layer displacement correlation functions.

The solution of most problems related to characteristic frequencies and displacement–displacement correlation functions of Sm-*A* films require cumbersome numerical calculations. This is associated with the absence of convenient methods for determination of eigenfrequencies and eigenmodes of Sm-*A* films. In discrete approach the central problem is associated with the necessity of a definition of symmetric $N\times N$ matrix eigenvalues where N is the number of smectic layers.

On the other hand Sm-*A* films possess one-dimensional periodic structure. For the analysis of dynamic properties of such systems the approach based on the Chebyshev polynomials of the second kind $U_n(x)$ was successfully used [15]. In the present work this approach is applied for description of Sm-*A* films. It enables one to get the analytical solutions of the most interesting problems and also to simplify the procedure of numerical computations significantly. We calculate the characteristic frequencies of the N-layer system and describe the motions of separate layers. The spectral densities of displacement–displacement correlation functions are calculated using the fluctuation–dissipation theorem. The rather simple expressions were obtained for the time-dependent correlation function of layer displacements.

The work is organized as follows. In Sec. II within the framework of the discrete model for freely suspended film the set of equations of motion is presented and the characteristic equation is expressed through the Chebyshev polynomials. Section III is devoted to the solution of the characteristic equation and to the analysis of eigenfrequencies and relaxation times. In Sec. IV the static and dynamic displacement–displacement correlation functions are calculated. In Sec. V the results obtained are discussed.

II. EQUATIONS OF MOTION

We consider the freely suspended film of Sm-*A* liquid crystal consisting of N layers. Its motion can be described within the framework of the discrete model suggested in Refs. [1–3,7,8]. In this model it is supposed that in an equilibrium state the smectic layers are equidistant planes separated by the distance d. We introduce the Cartesian coordinate frame so that the xy plane coincides with the equilibrium position of the first layer; the film is located in the region $z\geqslant 0$.

We use the notation $u_n(\mathbf{r}_\perp,t)$ for displacement of the nth smectic layer in the z direction in the $[\mathbf{r}_\perp, z=(n-1)d]$

point. The contribution to the free energy of a film caused by inhomogeneous displacements of layers has the form

$$F=\tfrac{1}{2}\int d\mathbf{r}_\perp\left\{\frac{B}{d}\sum_{n=1}^{N-1}(u_{n+1}-u_n)^2+dK\sum_{n=1}^{N-1}(\Delta_\perp u_n)^2+\gamma[(\nabla_\perp u_1)^2+(\nabla_\perp u_N)^2]\right\},\tag{2.1}$$

where B and K are the layer compression and layer bend elastic constant and γ is the surface tension. We consider that the coefficient K is the same for the surface and intrinsic layers. The force acting on the nth layer consists of elastic, $-d^{-1}(\delta F/\delta u_n)$, and viscous, $\eta_3\Delta_\perp(\partial u_n/\partial t)$, contributions, where η_3 is the layer sliding viscosity.

We assume the external forces to be equal to zero in studies of characteristic oscillations. In linear approximation the set of equations of motion has the form:

$$\rho\frac{\partial^2u_1(\mathbf{r}_\perp,t)}{\partial t^2}=B\frac{u_2(\mathbf{r}_\perp,t)-u_1(\mathbf{r}_\perp,t)}{d^2}-K\Delta_\perp^2u_1(\mathbf{r}_\perp,t)+\frac{\gamma}{d}\Delta_\perp u_1(\mathbf{r}_\perp,t)+\eta_3\Delta_\perp\frac{\partial u_1(\mathbf{r}_\perp,t)}{\partial t},$$

$$\rho\frac{\partial^2u_n(\mathbf{r}_\perp,t)}{\partial t^2}=B\frac{u_{n+1}(\mathbf{r}_\perp,t)-2u_n(\mathbf{r}_\perp,t)+u_{n-1}(\mathbf{r}_\perp,t)}{d^2}-K\Delta_\perp^2u_n(\mathbf{r}_\perp,t)+\eta_3\Delta_\perp\frac{\partial u_n(\mathbf{r}_\perp,t)}{\partial t},$$

$$n=2,3,\dots,N-1,\tag{2.2}$$

$$\rho\frac{\partial^2u_N(\mathbf{r}_\perp,t)}{\partial t^2}=B\frac{u_{N-1}(\mathbf{r}_\perp,t)-u_N(\mathbf{r}_\perp,t)}{d^2}-K\Delta_\perp^2u_N(\mathbf{r}_\perp,t)+\frac{\gamma}{d}\Delta_\perp u_N(\mathbf{r}_\perp,t)+\eta_3\Delta_\perp\frac{\partial u_N(\mathbf{r}_\perp,t)}{\partial t}.$$

We represent the solution of the problem as a plane wave

$$u_n(\mathbf{q}_\perp,\omega)e^{i\mathbf{q}_\perp\cdot\mathbf{r}_\perp-i\omega t}.$$

Then the set of equations of motion turns into a system of linear algebraic equations with respect to the components $u_1(\mathbf{q}_\perp,\omega),\dots,u_N(\mathbf{q}_\perp,\omega)$. In what follows the arguments $\mathbf{q}_\perp,\omega$ in these components are omitted. So, we have

$$\left(\rho\omega^2+i\omega\eta_3q_\perp^2-\frac{B}{d^2}-Kq_\perp^4-\frac{\gamma}{d}q_\perp^2\right)u_1+\frac{B}{d^2}u_2=0,$$

$$\left(\rho\omega^2+i\omega\eta_3q_\perp^2-2\frac{B}{d^2}-Kq_\perp^4\right)u_n+\frac{B}{d^2}u_{n-1}+\frac{B}{d^2}u_{n+1}=0,\quad n=2,3,\dots,N-1,\tag{2.3}$$

$$\left(\rho\omega^2+i\omega\eta_3q_\perp^2-\frac{B}{d^2}-Kq_\perp^4-\frac{\gamma}{d}q_\perp^2\right)u_N+\frac{B}{d^2}u_{N-1}=0.$$

Equating to zero the determinant of this system we get the equation for determination of eigenfrequencies of Sm-*A* film.

Within the aim of obtaining an analytical solution of Eq. (2.3) it is convenient to rewrite this set of equations in the matrix form

$$\hat{A}\mathbf{u}=0,\tag{2.4}$$

where the column-vector $\mathbf{u}$ and tridiagonal symmetric matrix $\hat{A}$ are

$$\mathbf{u}=\begin{pmatrix}u_1\\u_2\\\vdots\\u_N\end{pmatrix},$$

$$\hat{A}=\begin{pmatrix}(2x+1-\alpha)&1&0&\dots&0&0&0\\1&2x&1&\dots&0&0&0\\0&1&2x&\dots&0&0&0\\\vdots&\vdots&\vdots&\ddots&\vdots&\vdots&\vdots\\0&0&0&\dots&2x&1&0\\0&0&0&\dots&1&2x&1\\0&0&0&\dots&0&1&(2x+1-\alpha)\end{pmatrix}.\tag{2.5}$$

Here we introduced the notations:

$$x=-1+\frac{d^2}{2B}(\rho\omega^2+i\omega\eta_3q_\perp^2-Kq_\perp^4),\qquad \alpha=\frac{d\gamma q_\perp^2}{B}.\tag{2.6}$$

In order to solve the characteristic equation $\det\hat{A}=0$ it is convenient to represent the determinant of the $\hat{A}$ matrix in form:

$$\det\hat{A}=U_N(x)+2(1-\alpha)U_{N-1}(x)+(1-\alpha)^2U_{N-2}(x),\tag{2.7}$$

where the following notation for tridiagonal determinant of the nth order is introduced:

$$U_n(x)=\begin{vmatrix}2x&1&0&\dots&0&0&0\\1&2x&1&\dots&0&0&0\\0&1&2x&\dots&0&0&0\\\vdots&\vdots&\vdots&\ddots&\vdots&\vdots&\vdots\\0&0&0&\dots&2x&1&0\\0&0&0&\dots&1&2x&1\\0&0&0&\dots&0&1&2x\end{vmatrix}.\tag{2.8}$$

The following properties of this determinant are valid:

$$U_1(x)=2x,\quad U_2(x)=4x^2-1,\quad U_n(x)-2xU_{n-1}(x)+U_{n-2}(x)=0. \tag{2.9}$$

These equalities are the recursion relations for the Chebyshev polynomials of the second kind $U_n(x)$ [16,17]. Brillouin and Parodi [15] have successfully used Eq. (2.8) for the description of oscillations in one-dimensional periodic structures. Note $U_n(x)=C_n^1(x)$, where $C_n^\nu(x)$ is the Gegenbauer polynomial [16,17].

Using the recursion relation (2.9) the characteristic equation $\det\hat{A}=0$ can be written as an equation with respect to x:

$$[x+1-\alpha]U_{N-1}(x)-\alpha\left(1-\frac{\alpha}{2}\right)U_{N-2}(x)=0. \tag{2.10}$$

Its left-hand side is the polynomial of the nth power and generally Eq. (2.10) has n solutions. The characteristic frequencies are calculated from the quadratic equation (2.6) with respect to ω. Its solutions

$$\omega_\pm^{(l)}=-i\frac{\eta_3 q_\perp^2}{2\rho}\pm\sqrt{\frac{2B}{\rho d^2}(1+x^{(l)})+\frac{Kq_\perp^4}{\rho}-\frac{\eta_3^2 q_\perp^4}{4\rho^2}},\quad l=1,2,\ldots,N \tag{2.11}$$

correspond to each root $x^{(l)}$ of Eq. (2.10).

The problem of obtaining eigenfrequencies reduces to the solution of Eq. (2.10). These solutions depend on the $\alpha=d\gamma q_\perp^2/B$ parameter. For $0\leqslant\alpha\leqslant 2$ all solutions of this equation are in the interval $-1\leqslant x\leqslant 1$. If the α parameter is in the interval $2<\alpha<2+2/(N-1)$ then $N-1$ solutions are in the region $-1\leqslant x\leqslant 1$ and one solution is in the region $x>1$. For $\alpha>2+2/(N-1)$ there are $N-2$ solutions in the interval $-1\leqslant x\leqslant 1$ and two solutions are more than the unit, and for $\alpha\gg 1$ these two solutions coincide.

It is convenient to use the trigonometric representation of the Chebyshev polynomials to get the solution in the interval $-1\leqslant x\leqslant 1$ [15–17]

$$x=\cos\theta,\quad U_n(\cos\theta)=\frac{\sin[(n+1)\theta]}{\sin\theta}. \tag{2.12}$$

Then Eq. (2.10) has the form

$$(1-\alpha+\cos\theta)\frac{\sin(N\theta)}{\sin\theta}=\alpha\left(1-\frac{\alpha}{2}\right)\frac{\sin[(N-1)\theta]}{\sin\theta}, \tag{2.13}$$

where $0\leqslant\theta\leqslant\pi$.

In the interval $|x|>1$ the substitution $x=\cosh\theta$ can be used. Then the Chebyshev polynomials are [15]

$$U_n(\cosh\theta)=\frac{\sinh[(n+1)\theta]}{\sinh\theta}. \tag{2.14}$$

Now we can obtain the solution of the set of equations (2.4). For the lth root of the characteristic equation (2.10) this set of equations can be written as

$$2x^{(l)}\begin{pmatrix}u_1\\u_2\\u_3\\\vdots\\u_{N-1}\\u_N\end{pmatrix}+\begin{pmatrix}u_2\\u_3\\u_4\\\vdots\\u_N\\0\end{pmatrix}+\begin{pmatrix}0\\u_1\\u_2\\\vdots\\u_{N-2}\\u_{N-1}\end{pmatrix}+(1-\alpha)\begin{pmatrix}u_1\\0\\0\\\vdots\\0\\u_N\end{pmatrix}=0. \tag{2.15}$$

If we assume the amplitude of displacement of the first layer to be $u_1=1$ then from Eq. (2.15) we obtain the displacement amplitudes of all layers sequentially. The solution of Eq. (2.15) has the form:

$$u_n^{(l)}(\mathbf{q}_\perp)=(-1)^{n-1}[U_{n-1}(x^{(l)})+(1-\alpha)U_{n-2}(x^{(l)})],\quad n,l=1,2,\ldots,N. \tag{2.16}$$

Here we formally assumed that $U_{-1}(x)=0$.

The displacement of the nth layer of a film can be written as

$$u_n(\mathbf{r}_\perp,t)=\mathrm{Re}\int\frac{d\mathbf{q}_\perp}{(2\pi)^2}e^{i\mathbf{q}_\perp\cdot\mathbf{r}_\perp}\sum_{l=1}^{N}[a_+^{(l)}(\mathbf{q}_\perp)e^{-i\omega_+^{(l)}t}+a_-^{(l)}(\mathbf{q}_\perp)e^{-i\omega_-^{(l)}t}]u_n^{(l)}(\mathbf{q}_\perp),\quad n=1,2,\ldots,N, \tag{2.17}$$

where $a_\pm^{(l)}(\mathbf{q}_\perp)$ are the complex amplitudes of eigenmodes.

III. SPECTRUM OF FUNDAMENTAL FREQUENCIES

The characteristic equation $\det\hat{A}=0$ is an algebraic equation of $2N$ power with respect to ω. Its roots are eigenfrequencies of the Sm-*A* film. We reduce the problem of obtaining them to the solution of the system of Eqs. (2.6) and (2.10). Equation (2.6) is a quadratic one with respect to ω and a linear one with respect to x. Equation (2.10) contains the variable x only. This equation can be transformed to a rather simple trigonometric equation (2.13) with the help of a change of variables given by Eq. (2.12). This approach essentially simplifies the eigenfrequencies problem.

First, we obtain the auxiliary variable x from Eq. (2.10). This equation can be solved exactly for α equal to 0, 1, 2, and $\alpha\to\infty$. For $\alpha=0$ the solutions of this equation are

$$x^{(l)}=-\cos\frac{(l-1)\pi}{N},\quad l=1,2,\ldots,N. \tag{3.1}$$

For $\alpha=1$ we obtain

$$x^{(l)}=-\cos\frac{l\pi}{N+1},\quad l=1,2,\ldots,N. \tag{3.2}$$

If $\alpha=2$, then

$$x^{(l)}=-\cos\frac{l\pi}{N},\quad l=1,2,\ldots,N. \tag{3.3}$$

For $\alpha\to\infty$ two solutions of Eq. (2.10) turn into infinity and for the remaining roots we have

$$x^{(l)}=-\cos\frac{l\pi}{N-1},\quad l=1,2,\ldots,N-2. \tag{3.4}$$

If $\alpha\ll1$ then Eq. (2.10) can be solved by iteration. In linear approximation over α we have

$$(x+1)U_{N-1}(x)=\alpha[U_{N-1}(x)+U_{N-2}(x)].$$

This equation may be solved by using the substitution $x=\cos\theta$. As a result we have

$$x^{(1)}=-1+\frac{\alpha}{N},\quad x^{(l)}=-\cos\frac{(l-1)\pi}{N}+2\frac{\alpha}{N}\cos^2\frac{(l-1)\pi}{2N},\quad l=2,3,\ldots,N. \tag{3.5}$$

For $\alpha\gg1$ it is convenient to introduce a small parameter $\epsilon=\alpha^{-1}$. Then Eq. (2.10) reduces to

$$U_{N-2}(x)=2\epsilon[U_{N-1}(x)+U_{N-2}(x)]-2\epsilon^2(x+1)U_{N-1}(x).$$

From this equation we obtain

$$x^{(l)}=-\cos\frac{l\pi}{N-1}-\frac{2}{\alpha(N-1)}\sin^2\frac{l\pi}{N-1},\quad l=1,2,\ldots,N-2. \tag{3.6}$$

The remaining two roots in the given approximation coincide and are equal to

$$x^{(N-1)}=x^{(N)}=\frac{\alpha}{2}. \tag{3.7}$$

Note that the solutions (3.6) and (3.7) can be used for numerical estimates even for not too large α. So for $\alpha=2$ and $N>15$ the relative error for the value $1+x^{(l)}$ entering Eq. (2.11) does not exceed 5% for any mode.

If we use the typical parameters of Sm-*A* liquid crystal: $B\sim2.5\times10^7$ dyn/cm^2, $\gamma\sim30$ dyn/cm, $d\sim3\times10^{-7}$ cm, then the condition $\alpha\ll1$ results to inequality $q_\perp\ll10^6$ cm^{-1}. This range of wave numbers is studied by methods of excitation of mechanical oscillations [18] and light scattering experiments. The condition $\alpha\gg1$ corresponds to the wave numbers $q_\perp\gg10^6$ cm^{-1} which are studied by methods of dynamic x-ray scattering.

Now we analyze the behavior of eigenfrequencies by increasing the wave number. For each value of the auxiliary variable $x^{(l)}$ two eigenfrequencies $\omega_\pm^{(l)}$ are obtained from Eq. (2.11). We consider the cases $\alpha\ll1$ and $\alpha\gg1$ separately.

A. $\alpha\ll1$, i.e., $q_\perp\ll\sqrt{B/\gamma d}$

(1) For very long waves,

$$q_\perp^2\ll\frac{8\gamma\rho}{\eta_3^2dN}, \tag{3.8}$$

all modes are oscillations with the following set of eigenfrequencies

$$\omega_\pm^{(1)}=\pm c^{(1)}q_\perp-i\omega'', \tag{3.9}$$

$$\omega_\pm^{(l)}=\pm\frac{c^{(l)}}{d}-i\omega'',\quad l=2,3,\ldots,N,$$

where

$$c^{(1)}=\sqrt{\frac{2\gamma}{\rho dN}},$$

$$c^{(l)}=2\sqrt{\frac{B}{\rho}}\sin\frac{(l-1)\pi}{2N},\quad l=2,3,\ldots,N, \tag{3.10}$$

$$\omega''=\frac{\eta_3q_\perp^2}{2\rho}.$$

The low-frequency oscillating mode $\omega_\pm^{(1)}$ was obtained in [3]. The expression for velocity $c^{(1)}$ has been shown to be consistent with the experimental data. This mode describes the film motion caused by the surface tension. Interlayer distances are constant in this motion. The other eigenfrequencies $\omega_\pm^{(l)}$, $l=2,3,\ldots,N$, are generated by the elastic forces arising from the variation of the interlayer distances. For these modes the eigenfrequencies are independent of the wave number.

(2) In the region of wave numbers

$$\frac{8\gamma\rho}{\eta_3^2dN}<q_\perp^2<\frac{2\pi\sqrt{B\rho}}{\eta_3dN} \tag{3.11}$$

the low-frequency mode turn into a relaxation one

$$\tau_\pm^{(1)}=-\frac{i}{\omega_\pm^{(1)}}.$$

If $q_\perp^2\gg8\gamma\rho/\eta_3^2dN$, than the slow relaxation time is equal to

$$\tau_+^{(1)}=\frac{\eta_3Nd}{2\gamma}. \tag{3.12}$$

This expression coincides with one obtained in [7,12]. For the relaxation time $\tau_-^{(1)}$, the inequality $\tau_-^{(1)}\ll\tau_+^{(1)}$ is valid. The remaining modes, $\omega_\pm^{(l)}$, $l=2,3,\ldots,N$, remain oscillating as well as for the previous interval of the wave numbers.

In the interval

$$\frac{2\pi\sqrt{B\rho}}{\eta_3dN}<q_\perp^2<\frac{4\sqrt{B\rho}}{\eta_3d}$$

the oscillating modes convert into relaxation ones in turn.

(3) In the region

$$\frac{4\sqrt{B\rho}}{\eta_3 d} < q_\perp^2 \ll \frac{B}{\gamma d} \tag{3.13}$$

all modes are relaxation

$$\tau_\pm^{(l)} = -\frac{i}{\omega_\pm^{(l)}}, \quad l=1,2,\ldots,N.$$

The relaxation times can be calculated from Eqs. (2.11) and (3.5)

$$\tau_+^{(1)} = \frac{\eta_3}{(2\gamma/dN)+Kq_\perp^2}, \tag{3.14}$$

$$\tau_+^{(l)} = \frac{\eta_3 q_\perp^2}{(4B/d^2)(\sin^2[(l-1)\pi/2N]+(d\gamma q_\perp^2/NB)\cos^2[(l-1)\pi/2N])+Kq_\perp^4}, \quad l=2,3,\ldots,N.$$

B. $\alpha \gg 1$, i.e., ($q_\perp \gg \sqrt{B/\gamma d}$)

In this region the eigenmodes decay with relaxation times

$$\tau_+^{(l)} = \frac{\eta_3 q_\perp^2}{(4B/d^2)(\sin^2[l\pi/2(N-1)]-[B/(N-1)d\gamma q_\perp^2]\sin^2[l\pi/(N-1)])+Kq_\perp^4}, \quad l=1,2,\ldots,N-2,$$

$$\tau_+^{(N-1)} = \tau_+^{(N)} = \frac{\eta_3 q_\perp^2}{(\gamma q_\perp^2/d)+(2B/d^2)+Kq_\perp^4}. \tag{3.15}$$

For every mode we present the larger of the two relaxation times. It is not difficult to calculate $\tau_-^{(l)}$ from Eq. (2.11).

To illustrate the obtained results we consider the motion of a six layer film. For various modes the layer motions are shown in Fig. 1. The calculations were provided by Eqs. (2.16) and (3.5) with the typical parameters of Sm-*A*: $K \sim 10^{-6}$ dyn, $\eta_3 \sim 1$ Pz, $\rho \sim 1$ g/cm^3. The calculations are provided for wave numbers $q_\perp \leqslant 10^5$ cm^{-1} which correspond to the light scattering or mechanical oscillation problems. As is seen from Fig. 1 the layers equally spaced from the surfaces move either in phase or in opposite phase. This result is consistent with the prediction of Ref. [7] for a discrete model and Refs. [10–12] for a continual model. As the wave number $q_\perp$ increases the character of motions varies. As it follows from Eqs. (2.16) and (3.6) for $q_\perp \geqslant 10^7$ cm^{-1} the surface layers of the film are practically immobile and two modes disappear in the spectrum.

The dependences of the real and imaginary parts of the eigenfrequencies on $q_\perp$ for the small values of $q_\perp$ are shown in Fig. 2. The calculations were provided by Eqs. (2.11) and (3.5). As is seen the first mode is acoustic and the remaining modes are optic with $\omega \neq 0$ for $q_\perp = 0$. As is evident from Fig. 1 the center of gravity displaces in the first mode only. For the small $q_\perp$ this mode is the transverse sound wave with the sound velocity depending on the surface tension and the mass per the unit of the film surface. For the large $q_\perp$ all eigenmodes are relaxation with the characteristic times shown in Fig. 3. Every mode decays with two sharply differed relaxation times. According to Eq. (2.11) these times have the form

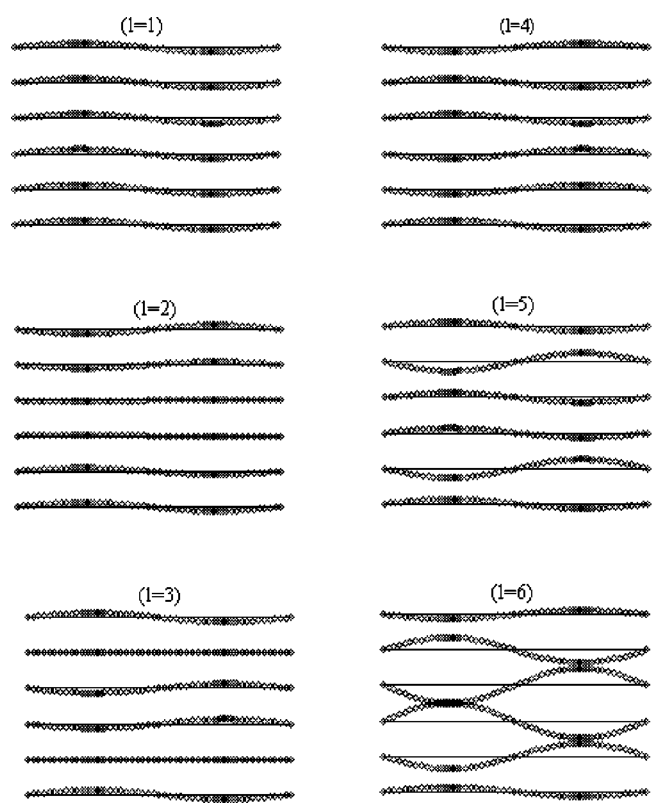

FIG. 1. Types of eigenmodes of six layer free-standing Sm-*A* film. ($l=1$), acoustical mode; ($l=2,3,\ldots,6$), optical modes.

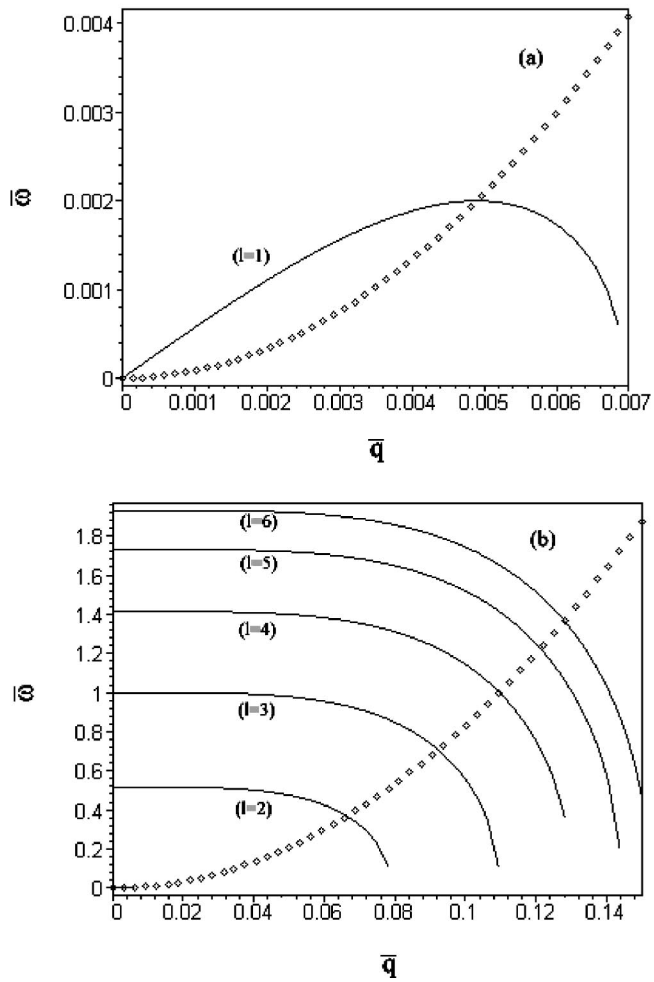

FIG. 2. Dependences of the real part and absolute value of the imaginary part of eigenfrequencies on the wave number for six layer Sm-*A* film. (a) acoustical mode; (b) optical modes. Solid lines correspond to the real parts and point lines are the imaginary parts. The dimensionless frequency $\overline{\omega}=\omega d\sqrt{\rho/B}$ and dimensionless wave number $\overline{q}=q_{\perp}\sqrt{d\gamma/B}$ are used.

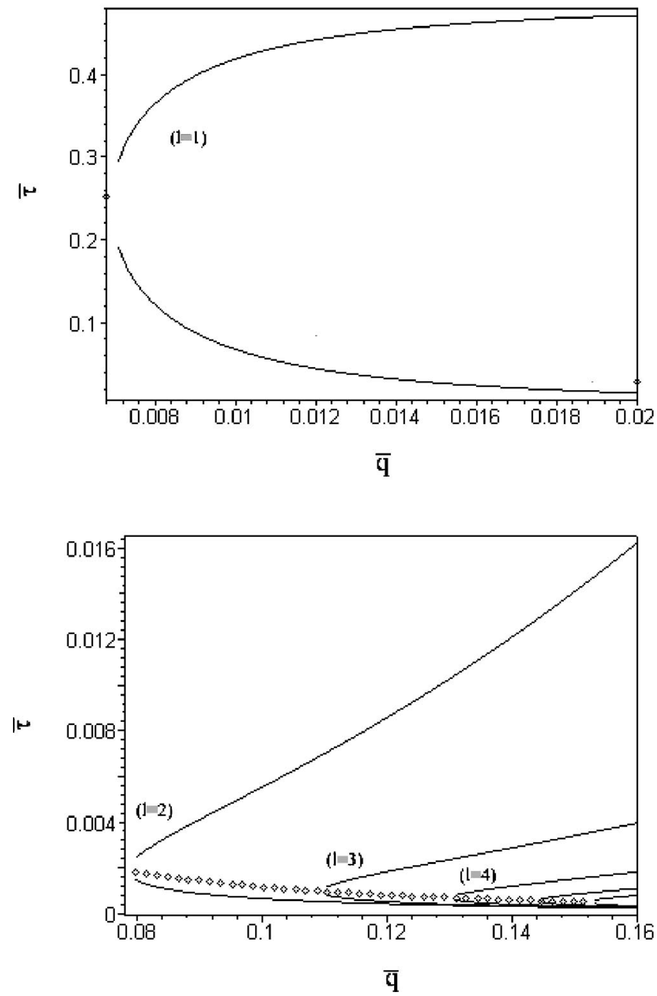

FIG. 3. The appearance in turn of two branches of relaxation times in six layer Sm-*A* film. The branch points appear when relevant real parts of the eigenfrequencies vanish. Upper and lower solid lines correspond to $\tau_{+}^{(l)}$ and $\tau_{-}^{(l)}$, respectively. The point line corresponds to the absolute value of the inverse imaginary part of eigenfrequency. Here $\overline{\tau}=\tau(\sqrt{KB}/\eta_3 d)$ is dimensionless time. The dimensionless wave number $\overline{q}$ is defined in the caption of Fig. 2.

$$\tau_{\pm}^{(l)}=\frac{2\rho}{\eta_3 q_{\perp}^2}\times\left[1\mp\sqrt{1-\left(\frac{2\rho}{\eta_3 q_{\perp}^2}\right)^2\left(\frac{2B}{\rho d^2}(1+x^{(l)})+\frac{Kq_{\perp}^4}{\rho}\right)}\right]^{-1},\quad l=1,2,\ldots,N, \tag{3.16}$$

where $x^{(l)}$ is given by Eq. (3.5). With the increase of $q_{\perp}$ the time $\tau_{+}^{(1)}$ tends to the constant value given by Eq. (3.12), which is valid up to $q_{\perp}\sim\sqrt{B/\gamma d}$. For larger $q_{\perp}$ the relaxation times $\tau_{+}^{(l)}$ are plotted in Fig. 4. The calculations were provided by Eqs. (3.6), (3.7), and (3.16). The dependences of $\tau_{+}^{(l)}$ on $q_{\perp}$ essentially coincide with the ones obtained in Ref. [7] by numerical calculations.

The dynamic characteristics of Sm-*A* films have been studied in recent coherent x-ray scattering experiments [7,9,13]. This scattering is caused by the thermally driven layer fluctuations. The intensity–intensity time correlation functions were recorded and the decay times were obtained for various films. The dynamic properties of the thick Sm-A films ($N\sim 10^3-10^4$) have been studied by the soft-x-ray photon correlation spectroscopy [7,9]. In these experiments x-ray scattering was mainly caused by the layer displacement fluctuations with $q_{\perp}\sim 2\times 10^3$ cm^{-1} ($\lambda\sim 35$ μm). For these values of $q_{\perp}$ according to Eq. (3.12) the largest relaxation time $\tau_{+}^{(1)}$ is independent of $q_{\perp}$ and grows linearly with the film thickness. This effect was observed in Refs. [7,9].

The thin films with $N=95$ [13] were studied by the coherent dynamic hard-x-ray scattering experiment. The oscillation exponential decay of the intensity–intensity temporal correlation function has been observed. This experiment is in qualitative agreement with Eq. (3.9) which predicts the oscillating character of motion in thin films.

It should be pointed out that according to Eq. (3.11) there is a marginal value

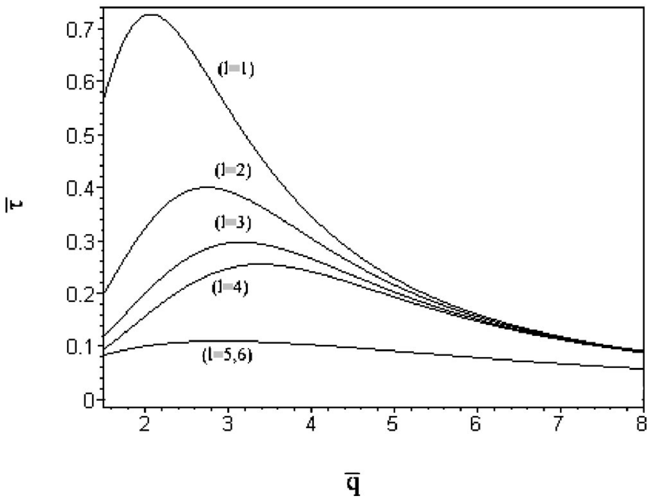

FIG. 4. The dependences of dimensionless relaxation times $\tau_+^{(l)}$ on the dimensionless wave number for six layer Sm-A film. The dimensionless variables are defined in the captions of Figs. 2 and 3.

$$q_{\perp c} \sim \sqrt{\frac{8\gamma\rho}{\eta_3^2 N d}}$$

which separates the oscillating and relaxational regimes for the acoustic mode. This value increases with the decreasing of the film thickness. Therefore for the fixed $q_\perp$ layer displacements may be oscillating or relaxational depending on the film thickness.

IV. THERMAL FLUCTUATIONS

The spectral densities of the Sm-*A* film thermal fluctuations can be found using the fluctuation-dissipation theorem (FDT) [19]. For this purpose we include into the free energy the term F_{ext} connected with the external forces $f_n(\mathbf{r}_\perp, t)$ of the pressure dimensionality acting on every layer. This term has the form

$$F_{\mathrm{ext}} = -\int d\mathbf{r}_\perp \sum_{n=1}^{N} u_n(\mathbf{r}_\perp, t) f_n(\mathbf{r}_\perp, t). \tag{4.1}$$

The elastic forces will contain an additional contribution $-(\delta F_{\mathrm{ext}}/\delta u_n) = f_n$. To use FDT the susceptibility matrix $\hat{\chi}$ determined by the relation $\mathbf{u} = \hat{\chi}\mathbf{f}$ had to be calculated. The equations of motion (2.3) containing the external forces in Fourier representation have the form

$$\left(\rho\omega^2 + i\omega\eta_3 q_\perp^2 - \frac{B}{d^2} - K q_\perp^4 - \frac{\gamma}{d} q_\perp^2\right) u_1 + \frac{B}{d^2} u_2 = -\frac{1}{d} f_1,$$

$$\left(\rho\omega^2 + i\omega\eta_3 q_\perp^2 - \frac{2B}{d^2} - K q_\perp^4\right) u_n + \frac{B}{d^2} u_{n-1} + \frac{B}{d^2} u_{n+1}$$

$$= -\frac{1}{d} f_n, \quad n = 2,3, \ldots, N-1, \tag{4.2}$$

$$\left(\rho\omega^2 + i\omega\eta_3 q_\perp^2 - \frac{B}{d^2} - K q_\perp^4 - \frac{\gamma}{d} q_\perp^2\right) u_N + \frac{B}{d^2} u_{N-1} = -\frac{1}{d} f_N.$$

This system may be presented in the matrix form

$$\hat{A}\mathbf{u} = -\frac{d}{B}\mathbf{f}, \tag{4.3}$$

where the matrix $\hat{A}$ is determined by Eqs. (2.5) and (2.6). Since

$$\hat{\chi} = -\frac{d}{B}\hat{A}^{-1}$$

the determination of the susceptibility matrix requires the calculation of the inverse matrix $\hat{A}^{-1}$. The matrix elements $(\hat{A}^{-1})_{nm}$ may be obtained with the help of the relation

$$(\hat{A}^{-1})_{nm} = \frac{1}{\det \hat{A}} A_{mn}, \quad m,n = 1,2, \ldots, N, \tag{4.4}$$

there A_{mn} is the cofactor of the matrix element $(\hat{A})_{mn}$ and the determinant $\det \hat{A}$ is given by Eq. (2.7). Thus the elements of the susceptibility matrix are

$$\chi_{nm} = \chi_{mn} = (-1)^{n+m+1} \frac{d}{B} \frac{[U_{m-1}(x) + (1-\alpha)U_{m-2}(x)][U_{N-n}(x) + (1-\alpha)U_{N-n-1}(x)]}{U_N(x) + 2(1-\alpha)U_{N-1}(x) + (1-\alpha)^2 U_{N-2}(x)}, \tag{4.5}$$

where

$$m,n = 1,2, \ldots, N; \quad n \geqslant m,$$

and the x value is given by Eq. (2.6).

The spectral densities of the layer displacement correlation functions are determined by the equality:

$$[u_n(\mathbf{q}_\perp) u_m(-\mathbf{q}_\perp)]_\omega = \int_{-\infty}^{\infty} \langle u_n(\mathbf{q}_\perp, t) u_m(-\mathbf{q}_\perp, 0)\rangle e^{i\omega t} dt, \tag{4.6}$$

where the statistical averaging is provided over all layer displacements at time $t = 0$. Using the FDT we get

$$[u_n(\mathbf{q}_\perp)u_m(-\mathbf{q}_\perp)]_\omega = i\frac{k_BT}{\omega}[\chi^*_{mn}(\mathbf{q}_\perp,\omega)-\chi_{nm}(\mathbf{q}_\perp,\omega)]. \tag{4.7}$$

For the static case the fluctuations of the layer displacements $\langle u_n^2(\mathbf{r}_\perp)\rangle$ and correlation functions $\langle u_n(\mathbf{r}_\perp)u_m(\mathbf{0})\rangle$ may be expressed via the Chebyshev polynomials too. Using the Fourier transformation with respect to the $\mathbf{r}_\perp$ variable we represent the free energy, Eq. (2.1), in the form, [2]:

$$F=\frac{1}{2}\int\frac{d\mathbf{q}_\perp}{(2\pi)^2}\sum_{n,m=1}^{N}u_n(\mathbf{q}_\perp)M_{nm}u_m(-\mathbf{q}_\perp), \tag{4.8}$$

where the symmetric tridiagonal matrix $\hat{M}$ is equal to

$$\hat{M}=-\frac{B}{d}\begin{pmatrix}(2y+1-\alpha) & 1 & 0 & \dots & 0 & 0 & 0\\ 1 & 2y & 1 & \dots & 0 & 0 & 0\\ 0 & 1 & 2y & \dots & 0 & 0 & 0\\ \vdots & \vdots & \vdots & \ddots & \vdots & \vdots & \vdots\\ 0 & 0 & 0 & \dots & 2y & 1 & 0\\ 0 & 0 & 0 & \dots & 1 & 2y & 1\\ 0 & 0 & 0 & \dots & 0 & 1 & (2y+1-\alpha)\end{pmatrix}. \tag{4.9}$$

Here y coincides with the x variable at $\omega=0$,

$$y=-1-\frac{d^2Kq_\perp^4}{2B}.$$

The layer displacement fluctuations and correlation functions may be expressed through the elements of the inverse matrix $\hat{M}^{-1}$ [2],

$$\langle u_n(\mathbf{r}_\perp)u_m(\mathbf{0})\rangle=\frac{k_BT}{(2\pi)^2}\int d\mathbf{q}_\perp(\hat{M}^{-1})_{nm}e^{i\mathbf{q}_\perp\cdot\mathbf{r}_\perp}, \tag{4.10}$$

where the integration is performed within the limits $2\pi/L<q_\perp<2\pi/a$, where L is the transverse size of the film and a is the molecular diameter.

The elements of the inverse matrix $\hat{M}^{-1}$ may be found in a similar way as it was done for the $\hat{A}^{-1}$ matrix. Finally the inverse matrix elements $(\hat{M}^{-1})_{nm}$ are given by Eq. (4.5) with $\omega=0$, i.e., with the replacement x by y in Eq. (4.5). Thus we have

$$\hat{M}^{-1}=\hat{\chi}(\omega=0).$$

The elements $(\hat{M}^{-1})_{nm}$ were obtained earlier in another form [2,7]. In the limit $q_\perp\to 0$ all elements of the inverse matrix $\hat{M}^{-1}$ are identical:

$$(\hat{M}^{-1})_{nm}\approx\frac{1}{2\gamma q_\perp^2}.$$

Now we consider the time-dependent layer displacement correlation functions. They may be found from the relation:

$$\langle u_n(\mathbf{q}_\perp,t)u_m(-\mathbf{q}_\perp,0)\rangle=\frac{ik_BT}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega}{\omega}[\chi^*_{mn}(\mathbf{q}_\perp,\omega)-\chi_{nm}(\mathbf{q}_\perp,\omega)]e^{-i\omega t}, \tag{4.11}$$

where the matrix elements $\chi_{nm}(\mathbf{q}_\perp,\omega)$ are given by Eq. (4.5). The integration in Eq. (4.11) is a rather complicated procedure and therefore we perform the calculations another way. For this purpose we diagonalize the free-energy expression Eq. (4.8) by transition to the normal coordinates. The eigenvalues $\lambda^{(l)}$ and eigenvectors $\mathbf{u}^{(l)}$ of the $\hat{M}$ matrix are calculated as well as the eigenfrequencies and eigenmodes in Sec. II. The nonzero solution condition of the equation

$$(\hat{M}-\lambda\hat{I})\mathbf{u}=0$$

leads to the same characteristic equation (2.10). The eigenvalues of the $\hat{M}$ matrix are expressed through the roots $x^{(l)}$, $l=1,2,\dots,N$ of this characteristic equation

$$\lambda^{(l)}=\frac{2B}{d}(1+x^{(l)})+Kdq_\perp^4,\quad l=1,2,\dots,N. \tag{4.12}$$

The eigenvectors components $u_n^{(l)}$ are given by Eq. (2.16). It should be noted that all eigenvalues $\lambda^{(l)}$ are positive. The N-dimensional vector of the layer displacements $\mathbf{u}(\mathbf{q}_\perp)$ may be expanded over the system of the normalized eigenvectors $\mathbf{v}^{(l)}(\mathbf{q}_\perp)=[\mathbf{u}^{(l)}(\mathbf{q}_\perp)]/\|\mathbf{u}^{(l)}(\mathbf{q}_\perp)\|$, where

$$\|\mathbf{u}^{(l)}(\mathbf{q}_\perp)\|=\sqrt{\sum_{n=1}^{N}[u_n^{(l)}(\mathbf{q}_\perp)]^2}.$$

Thus we have

$$u_n(\mathbf{q}_\perp,t)=\sum_{l=1}^{N} A^{(l)}(\mathbf{q}_\perp,t)v_n^{(l)}(\mathbf{q}_\perp), \quad n=1,2,\ldots,N,$$

where the functions $A^{(l)}(\mathbf{q}_\perp,t)$, $l=1,2,\ldots,N$ are the normal coordinates of the vector $\mathbf{u}(\mathbf{q}_\perp,t)$. The diagonalized expression for the free energy has the form

$$F=\frac{1}{2}\int \frac{d\mathbf{q}_\perp}{(2\pi)^2}\sum_{l=1}^{N}\lambda^{(l)}|A^{(l)}(\mathbf{q}_\perp,t)|^2. \tag{4.13}$$

Going over to the normal coordinates in the layer displacement correlation functions we have

$$\langle u_n(\mathbf{q}_\perp,t)u_m(-\mathbf{q}_\perp,0)\rangle =\sum_{l,p=1}^{N}\langle A^{(l)}(\mathbf{q}_\perp,t)A^{(p)}(-\mathbf{q}_\perp,0)\rangle v_n^{(l)}(\mathbf{q}_\perp)v_m^{(p)}(-\mathbf{q}_\perp). \tag{4.14}$$

All modes evaluate in time independently with eigenfrequencies obtained in Sec. II. Hence we get

$$\langle A^{(l)}(\mathbf{q}_\perp,t)A^{(p)}(-\mathbf{q}_\perp,0)\rangle =\langle A_+^{(l)}(\mathbf{q}_\perp,0)A^{(p)}(-\mathbf{q}_\perp,0)\rangle \times e^{-i\omega_+^{(l)}t}+\langle A_-^{(l)}(\mathbf{q}_\perp,0)A^{(p)}(-\mathbf{q}_\perp,0)\rangle e^{-i\omega_-^{(l)}t}. \tag{4.15}$$

The equilibrium correlation functions, entering the right-hand side of Eq. (4.15), may be found (Ref. [8]) using the equipartition theorem

$$\langle A^{(l)}(\mathbf{q}_\perp,0)A^{(p)}(-\mathbf{q}_\perp,0)\rangle=\frac{k_BT}{\lambda^{(l)}}\delta_{lp}, \quad l,p=1,2,\ldots,N, \tag{4.16}$$

and the condition of the statistical independence of the quantities $A^{(p)}(-\mathbf{q}_\perp,0)$ and $[(\partial A^{(l)}/\partial t(\mathbf{q}_\perp,t))]_{t=0}$:

$$\left\langle \left(\frac{\partial A^{(l)}}{\partial t}(\mathbf{q}_\perp,t)\right)_{t=0} A^{(p)}(-\mathbf{q}_\perp,0)\right\rangle=0, \quad l,p=1,2,\ldots,N. \tag{4.17}$$

Thus, we have

$$\langle A_\pm^{(l)}(\mathbf{q}_\perp,0)A^{(p)}(-\mathbf{q}_\perp,0)\rangle=\frac{\omega_\mp^{(l)}k_BT}{\lambda^{(l)}(\omega_\mp^{(l)}-\omega_\pm^{(l)})}\delta_{lp}, \quad l,p=1,2,\ldots,N. \tag{4.18}$$

Substituting Eqs. (4.15) and (4.18) into Eq. (4.14) we get the expression for the time-dependent layer displacement correlation functions

$$\langle u_n(\mathbf{q}_\perp,t)u_m(-\mathbf{q}_\perp,0)\rangle =\sum_{l=1}^{N}\frac{k_BT}{\lambda^{(l)}(\omega_-^{(l)}-\omega_+^{(l)})}(\omega_-^{(l)}e^{-i\omega_+^{(l)}t} -\omega_+^{(l)}e^{-i\omega_-^{(l)}t})v_n^{(l)}(\mathbf{q}_\perp)v_m^{(l)}(-\mathbf{q}_\perp), \quad n,m=1,2,\ldots,N. \tag{4.19}$$

Here the eigenvalues $\lambda^{(l)}$ and the eigenfrequencies $\omega_\pm^{(l)}$ are given by Eqs. (4.12) and (2.11).

A displacement–displacement correlation function in the $(\mathbf{r}_\perp,t)$ representation is calculated from the Fourier transform

$$\langle u_n(\mathbf{r}_\perp,t)u_m(\mathbf{0},0)\rangle =\frac{1}{(2\pi)^2}\int d\mathbf{q}_\perp\langle u_n(\mathbf{q}_\perp,t)u_m(-\mathbf{q}_\perp,0)\rangle e^{i\mathbf{q}_\perp\cdot\mathbf{r}_\perp}. \tag{4.20}$$

The integration is performed within the same limits as in Eq. (4.10). Since the correlation function $\langle u_n(\mathbf{q}_\perp,t)u_m(-\mathbf{q}_\perp,0)\rangle$ is independent of the wave vector $\mathbf{q}_\perp$ direction we may perform the integration over the angle between $\mathbf{r}_\perp$ and $\mathbf{q}_\perp$. Thus we have

$$\langle u_n(\mathbf{r}_\perp,t)u_m(\mathbf{0},0)\rangle=\frac{1}{2\pi}\int dq_\perp q_\perp J_0(q_\perp r_\perp) \times\langle u_n(\mathbf{q}_\perp,t)u_m(-\mathbf{q}_\perp,0)\rangle, \tag{4.21}$$

where J_0 is the zeroth-order Bessel function. The integration in Eq. (4.21) represents a rather simple numerical procedure.

V. DISCUSSION

In conclusion we analyze the layer displacement correlation functions. All eigenmodes of the free standing Sm-*A* films are oscillations for the small wave numbers defined by the condition Eq. (3.8). These modes manifest themselves as peaks in the frequency spectra. The spectrum density of the third layer displacement fluctuations $[u_3(\mathbf{q}_\perp)u_3(-\mathbf{q}_\perp)]_\omega$ in the free-standing six layer film is shown in Fig. 5. The calculations were performed by Eq. (4.7). The sharp peak arises due to the acoustic mode. The peak position is determined by the relation $\omega_{\max}\approx c^{(1)}q_\perp$, where the velocity $c^{(1)}$ is defined by Eq. (3.10). The width and location of this peak allows one to estimate the viscosity coefficient η_3 and the surface tension γ. Figure 6 shows the peak transformation with the increasing of the wave number. When the acoustic vibrations transform into relaxation process the frequency spectrum turns into Lorenzian. It should be noted that for small wave numbers, $q_\perp\ll\sqrt{B/\gamma d}$, and low frequencies, $\omega\ll 2/d\sqrt{B/\rho}\sin(\pi/N)$, the forms of spectrum densities are the same for all layer displacement correlation functions.

At high frequencies there are $N-1$ peaks, arising due to the optical oscillations, Eq. (3.9). The heights of these peaks

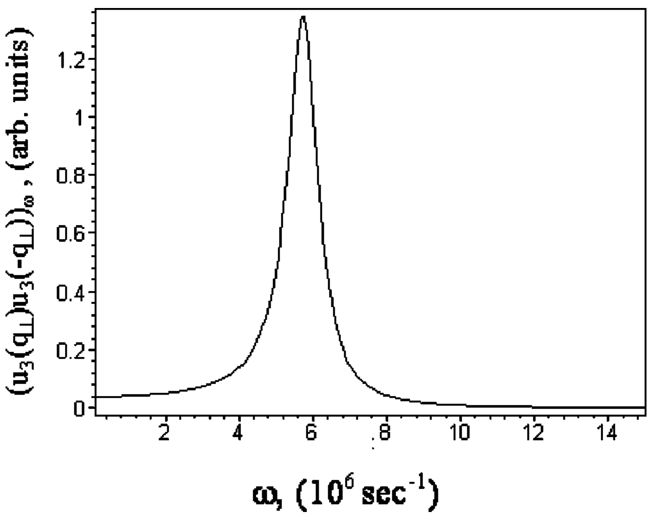

FIG. 5. The frequency dependence of spectral density of layer displacement fluctuations $[u_3(\mathbf{q}_\perp)u_3(-\mathbf{q}_\perp)]_\omega$ for $q_\perp = 10^3$ cm^{-1}.

are much less than the first one. The optical oscillations may exist if the wave numbers obey the inequality

$$q_\perp^2 < \frac{4\sqrt{B\rho}}{\eta_3 d}.$$

Using the typical Sm-*A* parameters we get $q_\perp < 2.5 \times 10^5$ cm^{-1}. The modes with these wave numbers are studied in light scattering experiments. According to Eq. (3.9) the characteristic frequencies of the optical modes are $\omega/2\pi \sim 2\times 10^9$ Hz. These frequencies correspond to the Mandel'shtam–Brillouin doublet produced by sound modes in condensed matters. The main difficulty connected with detecting this doublet in Sm-*A* films is in the smallness of the oscillating peaks compared to the central Lorenzian relaxation contour. As the wave number $q_\perp$ increases the oscillating regime of the optical modes is replaced by the relaxation one. For the wave numbers determined by Eq. (3.13) the

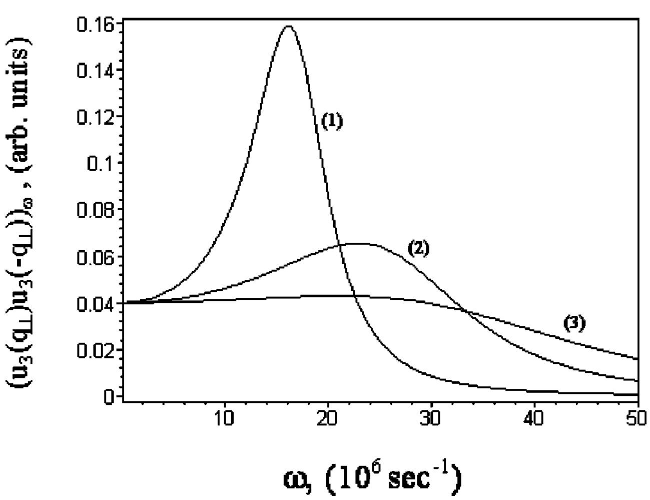

FIG. 6. Transformation of spectral density $[u_3(\mathbf{q}_\perp)u_3(-\mathbf{q}_\perp)]_\omega$ with increasing wave number. (1), $q_\perp = 3\times 10^3$ cm^{-1}; (2), $q_\perp = 5\times 10^3$ cm^{-1}; (3), $q_\perp = 7\times 10^3$ cm^{-1}.

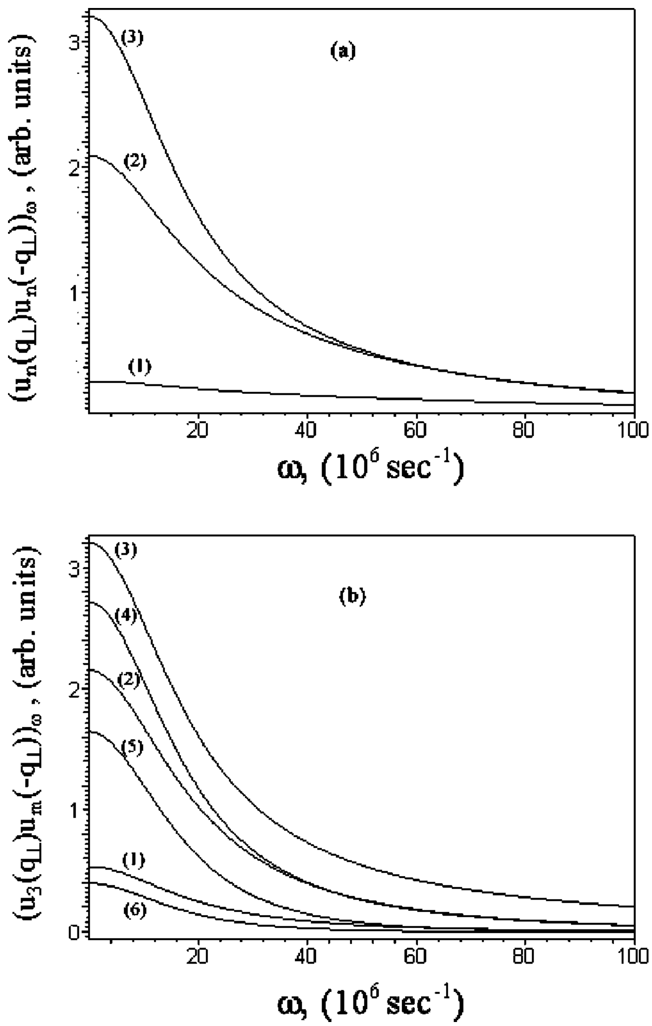

FIG. 7. Frequency dependence of spectral density $[u_n(\mathbf{q}_\perp)u_m(-\mathbf{q}_\perp)]_\omega$ for wave number $q_\perp = 3\times 10^6$ cm^{-1}. (a) Layer fluctuations: (1) $n=m=1$; (2) $n=m=2$; and (3) $n=m=3$. (b) Layer displacement–layer displacement correlation functions for $n=3$: (1) $m=1$; (2) $m=2$; (3) $m=3$; (4) $m=4$; (5) $m=5$; and (6) $m=6$.

inequality $\tau_+^{(1)} \gg \tau_+^{(l)}$, $l = 2,3,\dots,N$ is valid, i.e., the relaxation time $\tau_+^{(1)}$ determined by the surface tension is much more than the remaining relaxation times.

On further increasing of the wave number, $q_\perp \gg \sqrt{B/\gamma d}$, the relaxation times of various modes approach each other. In this region all eigenmodes pay the significant contribution to the time-dependent layer displacement correlation functions. Therefore the spectrum densities for various layers sufficiently differ from each other. These frequency dependencies are shown in Fig. 7. It is obvious that the spectrum densities have the maximal value for the inner layers. For the layers close to the film surface the spectrum densities decrease. This is connected with the decrease of the surface tension influence on the inner layer motion.

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