Description of dynamic x-ray scattering from freely standing smectic-*A* films

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The consequent description of the x-ray scattering from the free standing smectic-*A* films is suggested. Calculations are based on the discrete model for the film dynamics. The scattering intensity temporal autocorrelation function $\langle I(t)I(0)\rangle$ is obtained within the framework of this approach neglecting the multiple scattering and refraction effects. It is shown that the behavior of this function crucially depends on the film thickness. In particular, in thin films containing less than 10^3 layers the time dependence of $\langle I(t)I(0)\rangle$ has damping oscillation character. This behavior is determined by an acoustic mode that describes the film motion caused by the action of the surface tension. For thick films containing more than 10^4 layers the dynamics of the intensity temporal autocorrelation function is determined either by an acoustic mode or by a wide spectrum of modes depending on the x-ray geometry. In both the cases the autocorrelation function is a relaxation one. The results obtained are compared with the experiments on the coherent soft and hard x-ray scattering.

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I. INTRODUCTION

The layer displacements and the correlation functions in a freely standing smectic-*A* (Sm-*A*) films are intensively studied both experimentally and theoretically [1–12]. The basic experimental methods of studying the structure and the thermal fluctuations in these systems are the analysis of the specular and the diffusion x-ray scattering data [1,4–6,8,11]. The static properties of Sm-*A* films have been studied in detail [1,4,5]. In recent works the attention is paid mostly to dynamic properties. A major progress was obtained in the experiments on the coherent soft and hard x-ray scattering from the freely standing Sm-*A* films [6,8,11]. In these studies the scattering intensity temporal autocorrelation function $\langle I(t)I(0)\rangle$ has been measured. The experiments were performed in the vicinity of the first Bragg peak. It was found that in thick films [6,8] with $N\sim 10^4$, where N is the number of smectic layers, the autocorrelation function $\langle I(t)I(0)\rangle$ is described by a model with a single relaxation time associated with the film motion as a whole provided by the surface tension. In Ref. [11] the dynamic correlation function for thin films with $N\sim 10^2$ was studied and the damping oscillations have been found. Its behavior can be described by a simple relation

$$\langle I(t)I(0)\rangle = A + B\cos\omega t\,\exp(-t/\tau),$$

where the frequency ω and the relaxation time τ are fitting parameters. The analysis performed in Ref. [11] showed that the order of the magnitude of these parameters are the same as those describing the film motion as a whole under the action of the surface tension.

The question arises of the consistent description of the correlation function $\langle I(t)I(0)\rangle$ based on the Sm-*A* film dynamics equations. In this work we present calculations of $\langle I(t)I(0)\rangle$ in the framework of a discrete model. The solution of the problem is based on the frequency spectrum and the dynamic displacement-displacement correlation functions, which had been obtained in the previous study [12]. We show that the experimental results on the coherent soft and hard x-ray scattering from Sm-*A* films may be described by the simple analytical expressions both for thin and thick films.

The paper is organized as follows. In Sec. II we present the general relations for the scattering intensity temporal autocorrelation function. Section III is devoted to the analysis of the obtained results on coherent soft and hard x-ray scattering taking into account the experiment geometries. In conclusion, possible experimental facilities are discussed.

II. BASIC EQUATIONS

In the first Born approximation the intensity temporal autocorrelation function $\langle I(t)I(0)\rangle$ of the x-ray scattering is proportional to the square of the Fourier-transform of the electron density autocorrelation function

$$\langle I(t)I(0)\rangle \sim |S(t)|^2, \tag{2.1}$$

where

$$S(t) = \langle \rho(\mathbf{q},t)\rho(-\mathbf{q},0)\rangle. \tag{2.2}$$

Here $\rho(\mathbf{q},t)$ is the electron density Fourier transform and $\mathbf{q} = (\mathbf{q}_\perp, q_z)$ is the scattering vector. Due to the layered structure in the Sm-*A* films the electron density has a one-dimensional periodicity. We introduce the Cartesian coordinate frame with the z axis directed normally to the film surface. In the equilibrium the smectic layers are equidistant planes separated by the distance d. The first layer coincides with the xy plane, and the film is located in the region $z \geqslant 0$. In the N-layer Sm-*A* film the electron density can be expressed as

$$\rho(\mathbf{r}_\perp,z,t)=\rho_s\sum_{n=1}^{N}\rho_M[z-nd-u_n(\mathbf{r}_\perp,t)]=\rho_s\sum_{n=1}^{N}\int dz_1\rho_M(z_1)\delta(z_1-z+nd+u_n(r_\perp,t)),\tag{2.3}$$

where ρ_s is the surface molecular density in a layer, $u_n(\mathbf{r}_\perp,t)$ is the nth layer displacement from the equilibrium position along the z axis, and ρ_M is the linear electron density in molecule.

Substituting Eq. (2.3) into Eq. (2.2) we get the correlation function $S(\mathbf{q}_\perp,q_z,t)$ in the form

$$S(q_\perp,q_z,t)=\rho_s^2\sum_{n,m=1}^{N}\int d\mathbf{r}_\perp\exp(-i\mathbf{q}_\perp\cdot\mathbf{r}_\perp)\times\int dz_1\rho_M(z_1)\int dz_2\rho_M(z_2)\times\int dz'\exp(-iq_zz')\int dz''\exp(iq_zz'')\delta(z'-z_1-nd-u_n(r_\perp,t))\delta(z''-z_2-md-u_m(0,0)).\tag{2.4}$$

After integration of the δ functions we have

$$S(\mathbf{q}_\perp,\mathbf{q}_z,t)=\rho_s^2\sum_{n,m=1}^{N}\int d\mathbf{r}_\perp\exp(-i\mathbf{q}_\perp\cdot\mathbf{r}_\perp)\times\int dz_1\rho_M(z_1)\int dz_2\rho_M(z_2)\langle\exp[-iq_z\{z_1-z_2+(n-m)d+u_n(\mathbf{r}_\perp,t)-u_m(0,0)\}]\rangle.\tag{2.5}$$

Supposing the displacement fluctuations are Gaussian and integrating over z_1 and z_2 we get [6]

$$S(\mathbf{q}_\perp,q_z,t)=\rho_s^2|\rho_M(q_z)|^2\sum_{n,m=1}^{N}\exp[-iq_z(n-m)d]\times\int d\mathbf{r}_\perp\exp(-i\mathbf{q}_\perp\cdot\mathbf{r}_\perp)\times\exp\left(-\frac{q_z^2}{2}\langle[u_n(\mathbf{r}_\perp,t)-u_m(0,0)]^2\rangle\right).\tag{2.6}$$

Since the Sm-A film is isotropic in the layers plane we can perform the integration over the angle between $\mathbf{r}_\perp$ and $\mathbf{q}_\perp$. Therefore the correlation function can be written as

$$S(\mathbf{q}_\perp,q_z,t)=2\pi\rho_s^2|\rho_M(q_z)|^2\sum_{n,m=1}^{N}\exp[-iq_z(n-m)d]\times\exp\left(-\frac{q_z^2}{2}[\langle u_n^2(0,0)\rangle+\langle u_m^2(0,0)\rangle]\right)G_{nm}(q_\perp,q_z,t),\tag{2.7}$$

where

$$G_{nm}(q_\perp,q_z,t)=\int_0^\Lambda r_\perp dr_\perp J_0(q_\perp r_\perp)\exp[q_z^2\langle u_n(r_\perp,t)u_m(0,0)\rangle].\tag{2.8}$$

Here Λ is the linear size of the film, $J_0(x)$ is the zeroth-order Bessel function, $r_\perp$ is the distance from the z axis. The dependence of the correlation function $S(t)$ on time is determined by the $G_{nm}(q_\perp,q_z,t)$ function.

The dynamic displacement-displacement correlation function may be presented in the following form [6,12]:

$$\langle u_n(r_\perp,t)u_m(0,0)\rangle=\frac{1}{2\pi}\int_{2\pi/\Lambda}^{2\pi/u}dq_\perp q_\perp J_0(q_\perp r_\perp)\times\langle u_n(\mathbf{q}_\perp,t)u_m(-\mathbf{q}_\perp,0)\rangle,\tag{2.9}$$

where a is the molecular transverse size.

Formally Eqs. (2.7)–(2.9) describe the intensity-intensity temporal correlation function. It depends on the layer displacement—layer displacement correlation functions. To obtain these functions we start from the well-known expression for the free energy of the free standing Sm-A film

$$F=\frac{1}{2}\int d\mathbf{r}\left[B\left(\frac{\partial u}{\partial z}\right)^2+K(\Delta_\perp u)^2\right]+\frac{\gamma}{2}\int dr_\perp(|\boldsymbol{\nabla}_\perp u_1|^2+|\boldsymbol{\nabla}_\perp u_N|^2),\tag{2.10}$$

where B and K are the layer compression and layer bend elastic constants, γ is the surface tension. This expression consists of the bulk and the surface contributions. Describing the x-ray scattering it is convenient to use a discrete model for the free energy [2,6,12]

$$F=\frac{1}{2}\int d\mathbf{r}_\perp\left\{\frac{B}{d}\sum_{n=1}^{N-1}(u_{n+1}-u_n)^2+dK\sum_{n=1}^{N}(\Delta_\perp u_n)^2+\gamma[(\boldsymbol{\nabla}_\perp u_1)^2+(\boldsymbol{\nabla}_\perp u_N)^2]\right\}.\tag{2.11}$$

The motion of each smectic layer depends on the elastic, $-d^{-1}(\delta F/\delta u_n)$, and viscous, $\eta_3\Delta_\perp(\partial u_n/\partial t)$, forces. Here η_3 is layer sliding viscosity. The set of equations of motion of smectic layers for free standing film in ω, $q_\perp$ presentation has the form [6,12]

$$\left(\rho\omega^2+i\omega\eta_3 q_\perp^2-\frac{B}{d^2}-Kq_\perp^4-\frac{\gamma}{d}q_\perp^2\right)u_1+\frac{B}{d^2}u_2=0,$$

$$\left(\rho\omega^2+i\omega\eta_3 q_\perp^2-2\frac{B}{d^2}-Kq_\perp^4\right)u_n+\frac{B}{d^2}u_{n-1}+\frac{B}{d^2}u_{n+1}=0,\quad n=2,3,...,N-1, \tag{2.12}$$

$$\left(\rho\omega^2+i\omega\eta_3 q_\perp^2-\frac{B}{d^2}-Kq_\perp^4-\frac{\gamma}{d}q_\perp^2\right)u_N+\frac{B}{d^2}u_{N-1}=0.$$

Note that to describe the film dynamics in the case of solid supported films we had to replace the last equation in the set (2.12) by a flat boundary condition

$$u_N=0.$$

For the calculation of the layer displacement-layer displacement temporal correlation functions we can include into the free energy the term $F_{\rm ext}$ connected with the external forces. This term has the form

$$F_{\rm ext}=-\int d\mathbf{r}_\perp\sum_{n=1}^{N}u_n(\mathbf{r}_\perp,t)f_n(\mathbf{r}_\perp,t). \tag{2.13}$$

In this case the set of equations of motion (2.12) becomes nonhomogeneous with the solution

$$\mathbf{u}=\hat{\chi}\mathbf{f},$$

where $\hat{\chi}$ is the susceptibility matrix, and

$$\mathbf{u}=\begin{pmatrix}u_1\\u_2\\\vdots\\u_N\end{pmatrix},\quad \mathbf{f}=\begin{pmatrix}f_1\\f_2\\\vdots\\f_N\end{pmatrix}.$$

The spectral densities of the layer displacements correlation functions can be found by using the fluctuation-dissipation theorem,

$$\langle u_n(\mathbf{q}_\perp,t)u_m(-\mathbf{q}_\perp,0)\rangle=\frac{ik_BT}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega}{\omega}[\chi_{mn}^*(\mathbf{q}_\perp,\omega)-\chi_{nm}(\mathbf{q}_\perp,\omega)]e^{-i\omega t}, \tag{2.14}$$

where the elements of the susceptibility matrix are [12]

$$\chi_{nm}=\chi_{mn}=(-1)^{n+m+1}\frac{d}{B}\frac{[U_{m-1}(x)+(1-\alpha)U_{m-2}(x)][U_{N-n}(x)+(1-\alpha)U_{N-n-1}(x)]}{U_N(x)+2(1-\alpha)U_{N-1}(x)+(1-\alpha)^2U_{N-2}(x)}, \tag{2.15}$$

for

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

Here $U_n(x)$ are the Chebyshev polynomials of the second kind, [13,14], the α and x values are given by relations

$$\alpha=\frac{d\gamma q_\perp^2}{B},$$

$$x=-1+\frac{d^2}{2B}(\rho\omega^2+i\omega\eta_3 q_\perp^2-Kq_\perp^2). \tag{2.16}$$

The roots of denominator in Eq. (2.15) determine the eigenfrequencies of the free standing Sm-A film,

$$\omega_\pm^{(l)}=-i\frac{\eta_3 q_\perp^2}{2\rho}\pm\left(\frac{2B}{\rho d^2}(1+x^{(l)})+\frac{Kq_\perp^4}{\rho}-\frac{\eta_3^2 q_\perp^4}{4\rho^2}\right)^{1/2},\ l=1,2,\ldots,N, \tag{2.17}$$

where $x^{(l)}$ are the roots of the characteristic equation [12]

$$[x+1-\alpha]U_{N-1}(x)-\alpha\left(1-\frac{\alpha}{2}\right)U_{N-2}(x)=0. \tag{2.18}$$

For the case of a solid supported films the equations of motion have the form

$$\hat{A}_1\mathbf{u}=0, \tag{2.19}$$

where

$$u=\begin{pmatrix}u_1\\u_2\\\vdots\\u_{N-1}\end{pmatrix},$$

$$\tilde{A}_1=\begin{pmatrix}(2x+1-\alpha)&1&0&\cdots&0&0&0\\1&2x&1&\cdots&0&0&0\\0&1&2x&\cdots&0&0&0\\\vdots&\vdots&\vdots&\ddots&\vdots&\vdots&\vdots\\0&0&0&\cdots&2x&1&0\\0&0&0&\cdots&1&2x&1\\0&0&0&\cdots&0&1&2x\end{pmatrix}.$$

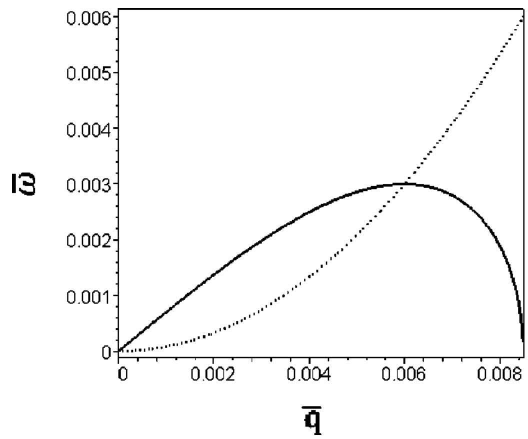

FIG. 1. Dependencies of the real part (solid line) and absolute value of the imaginary part (dotted line) of eigenfrequency on the wave number for the acoustical mode for free standing four-layer Sm-*A* film. The dimensionless frequency $\overline{\omega}=\omega d\sqrt{\rho/B}$ and dimensionless wave number $\overline{q}=q_\perp\sqrt{d\gamma/B}$ are used.

The existence of a nonzero solution of Eq. (2.19) results in the characteristic equation

$$U_{N-1}(x)+(1-\alpha)U_{N-2}(x)=0. \tag{2.20}$$

For each root of this equation the characteristic frequencies can be obtained from Eq. (2.17). The sufficient difference between the dynamics of freely standing and solid supported films is in the existence of low frequency acoustic mode with $\omega\sim q_\perp$ in the freely standing films for small $q_\perp$. All eigenmodes of the solid supported films are optical. Figures 1 and 2 show the dependencies of eigenfrequencies on $q_\perp$ in forth layer film for both cases.

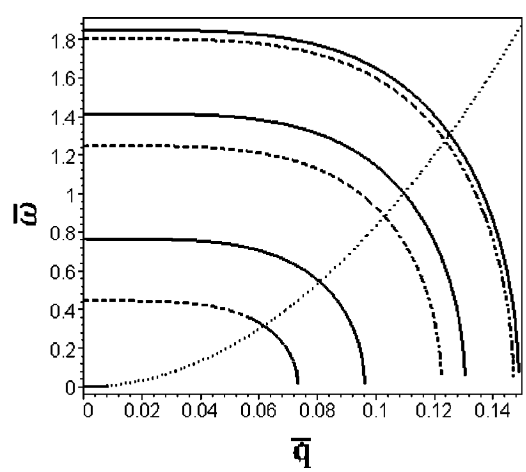

FIG. 2. Dependencies of the real parts (solid and dashed lines) and absolute values of the imaginary parts (dotted line) of eigenfrequencies on the wave number for the optical modes. Solid lines correspond to free standing four-layer Sm-*A* film and dashed lines correspond to the solid supported one. The dimensionless frequency $\overline{\omega}$ and dimensionless wave number $\overline{q}$ are defined in caption to Fig. 1.

III. ANALYSIS OF X-RAY SCATTERING

We apply the obtained results for the description of experiments on the coherent x-ray scattering from freely standing Sm-*A* films [6,8,11]. Performing the integration in Eq. (2.14) we can obtain the temporal correlation function of layer displacements in the form [12]

$$\langle u_n(\mathbf{q}_\perp,t)u_m(-\mathbf{q}_\perp,0)\rangle = \sum_{l=1}^{N}\frac{k_BT}{\lambda^{(l)}(q_\perp)}v_n^{(l)}(q_\perp)v_m^{(l)}(q_\perp) \times\frac{\omega_-^{(l)}\exp(-i\omega_+^{(l)}t)-\omega_+^{(l)}\exp(-i\omega_-^{(l)}t)}{\omega_-^{(l)}-\omega_+^{(l)}}, \tag{3.1}$$

where k_B is the Boltzmann constant, T is the temperature, and the value $\lambda^{(l)}(q_\perp)$ is given by the relation [12]

$$\lambda^{(l)}(q_\perp)=\frac{2B}{d}(1+x^{(l)})+Kdq_\perp^4. \tag{3.2}$$

The components of the normalized eigenvector $\mathbf{v}^{(l)}(q_\perp)$ corresponding to the lth eigenvibration are

$$v_n^{(l)}(q_\perp)=\frac{u_n^{(l)}(q_\perp)}{\sqrt{\sum_{n=1}^{N}[u_n^{(l)}(q_\perp)]^2}}, \tag{3.3}$$

$$u_n^{(l)}(q_\perp)=(-1)^{n-1}[U_{n-1}(x^{(l)})+(1-\alpha)U_{n-2}(x^{(l)})]. \tag{3.4}$$

Here n and l are the layer and mode numbers, respectively. Note, that for $t=0$ Eq. (3.1) describes the static correlation function

$$\langle u_n(\mathbf{q}_\perp)u_m(-\mathbf{q}_\perp)\rangle=\sum_{l=1}^{N}\frac{k_BT}{\lambda^{(l)}(q_\perp)}v_n^{(l)}(q_\perp)v_n^{(l)}(q_\perp), \tag{3.5}$$

which is written as a series of eigenmodes. This static correlation function agrees with those obtained previously in Refs. [2,5]. This function was analyzed in detail in Refs. [2,5] for free standing films and in Ref. [15] for solid supported films.

Usually the measurements are performed in the vicinity of the first Bragg peak with $q_z=2\pi/d$. The $q_\perp$ values are dependent on the experiment geometry and typical $q_\perp$ are within the limits 10^3–10^4 cm^{-1} [6,8,11]. It is essential that for these values of the scattering vector the expression for $\langle I(t)I(0)\rangle$ can be simplified significantly. The film size is of the order of $\Lambda\sim1$ cm.

For the typical Sm-*A* parameters the exponent in Eq. (2.8) is sufficiently small even at $t=0$. Thus for $q_z\sim10^7$ cm^{-1} and $\langle u_m(0,0)u_n(0,0)\rangle\sim20$ Å, [2], the exponent in Eq. (2.8) is of the order of 0.2, and it decreases rapidly when the time t in the correlation function increases. Therefore, we expand the exponential in Eq. (2.8) in the Taylor series and account for two first terms only

$$\exp[q_z^2\langle u_n(r_\perp,t)u_m(0,0)\rangle]\sim 1+q_z^2\langle u_n(r_\perp,t)u_m(0,0)\rangle. \tag{3.6}$$

Taking into account Eq. (3.6) we present the function $G_{nm}(q_z,q_\perp,t)$ as a sum of two terms

$$G_{nm}(q_z,q_\perp,t)=G_{nm}^{(0)}(q_z,q_\perp)+G_{nm}^{(1)}(q_z,q_\perp,t).$$

The time-dependent second term $G_{nm}^{(1)}(q_z,q_\perp,t)$ has the form

$$G_{nm}^{(1)}(q_z,q_\perp,t)=q_z^2\int_0^\Lambda dr_\perp r_\perp J_0(q_\perp r_\perp)\langle u_n(r_\perp,t)u_m(0,0)\rangle. \tag{3.7}$$

Using Eq. (2.9) we get

$$G_{nm}^{(1)}(q_z,q_\perp,t)=\frac{q_z^2}{2\pi}\int_{2\pi/\Lambda}^{2\pi/a}dq'_\perp q'_\perp\langle u_n(\mathbf{q}'_\perp,t)u_m(-\mathbf{q}'_\perp,0)\rangle\times\int_0^\Lambda dr_\perp r_\perp J_0(q_\perp r_\perp)J_0(q'_\perp r_\perp). \tag{3.8}$$

The integral over $r_\perp$ is the Lommel integral. It is equal to [13,14]

$$\int_0^\Lambda dr_\perp r_\perp J_0(q_\perp r_\perp)J_0(q'_\perp r_\perp)=\begin{cases}\Lambda\dfrac{q_\perp J_0(q'_\perp\Lambda)J_1(q_\perp\Lambda)-q'_\perp J_0(q_\perp\Lambda)J_1(q'_\perp\Lambda)}{q_\perp^2-q'^2_\perp} & \text{for } q_\perp^2-q'^2_\perp\neq 0\\[2mm] \dfrac{\Lambda^2}{2}[J_0^2(q_\perp\Lambda)+J_1^2(q_\perp\Lambda)] & \text{for } q_\perp=q'_\perp\end{cases} \tag{3.9}$$

This expression is the continuous function of the difference $q_\perp-q'_\perp$. Therefore we get

$$G_{nm}^{(1)}(q_z,q_\perp,t)=\frac{q_z^2\Lambda}{2\pi}\int_{2\pi/\Lambda}^{2\pi/a}dq'_\perp f(q'_\perp,q_\perp,\Lambda)q'^2_\perp\times\langle u_n(\mathbf{q}'_\perp,t)u_m(-\mathbf{q}'_\perp,0)\rangle, \tag{3.10}$$

where

$$f(q'_\perp,q_\perp,\Lambda)=\frac{q_\perp J_0(q'_\perp\Lambda)J_1(q_\perp\Lambda)-q'_\perp J_0(q_\perp\Lambda)J_1(q'_\perp\Lambda)}{q'_\perp(q_\perp^2-q'^2_\perp)}. \tag{3.11}$$

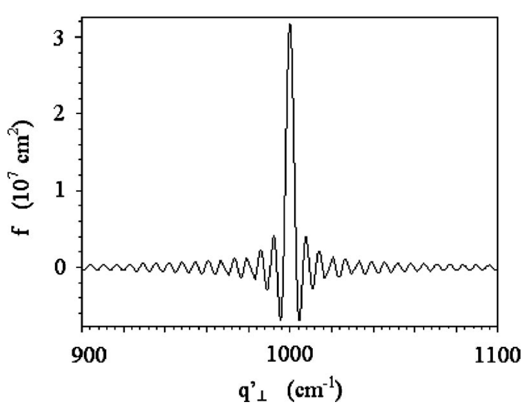

FIG. 3. Dependence of the function $f(q'_\perp,q_\perp,\Lambda)$ defined by Eq. (3.11) on the wave number $q'_\perp$ for $\Lambda=1$ cm and $q_\perp=10^3$ cm^{-1}.

Since the displacement-displacement correlation function behaves as $1/q'^2_\perp$ for small $q'_\perp$, the product

$$q'^2_\perp\langle u_n(\mathbf{q}'_\perp,t)u_m(-\mathbf{q}'_\perp,0)\rangle$$

is the regular function of $q'_\perp$. At the same time $f(q'_\perp,q_\perp,\Lambda)$ is a rapidly oscillating function of $q'_\perp$ variable, and it has a sharp peak at $q_\perp=q'_\perp$. Figure 3 shows the behavior of the function $f(q'_\perp,q_\perp,\Lambda)$.

Note that the upper integrating limit in Eq. (3.10) may be extended to infinity while the low limit, $q_{\min}=2\pi/\Lambda$, is introduced due to the Landau-Peierls instability that leads to the logarithmic divergence of the integral with the increasing of the film size. So Eq. (3.10) contains two contributions. The first of them is formed in the vicinity of the function $f(q'_\perp,q_\perp,\Lambda)$ peak, and the second one results from the lower integration limit. In what follows the second input will be omitted since it is noticeably less than the peak contribution. In the intermediate region the function $f(q'_\perp,q_\perp,\Lambda)$ oscillates rapidly and has a small amplitude. As long as the function

$$q'^2_\perp\langle u_n(\mathbf{q}'_\perp,t)u_m(-\mathbf{q}'_\perp,0)\rangle$$

is smooth, it may be carried out from the integral in Eq. (3.10) in the point $q'_\perp=q_\perp$. Accordingly we have

$$G_{mn}^{(1)}(q_z,q_\perp,t)=\langle u_n(\mathbf{q}_\perp,t)u_m(-\mathbf{q}_\perp,0)\rangle\frac{q_z^2q_\perp^2\Lambda}{2\pi}\times\int_{2\pi/\Lambda}^{2\pi/a}f(q'_\perp,q_\perp,\Lambda)dq'_\perp. \tag{3.12}$$

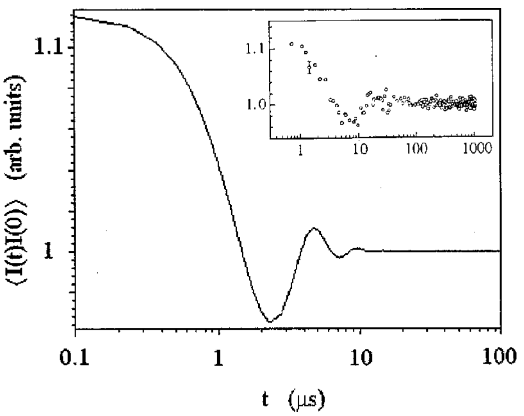

FIG. 4. Calculated time dependence of the scattering intensity autocorrelation function for 100-layer-thick film, $q_\perp = 10^3$ cm^{-1}. The inset shows the experimental results for 95-layer-thick film from [11]. Units are the same in the inset and in the environment.

The time-dependent correlation function

$$\langle u_n(\mathbf{q}_\perp,t)u_m(-\mathbf{q}_\perp,0)\rangle$$

can be calculated with the aid of Eq. (3.1), which contains summation over all modes. For the typical values of $q_\perp \sim 10^3 - 10^4$ cm^{-1}, the summation may be simplified. Actually, the amplitudes of various mode fluctuations, $k_BT/\lambda^{(l)}$, where $l=1,\ldots,N$, are significantly different. In order to illustrate this we consider the relation between fluctuation amplitudes of the lth and the first mode. Using Eq. (3.2) we can write

$$\frac{k_BT/\lambda^{(l)}}{k_BT/\lambda^{(1)}} = \frac{\lambda^{(1)}}{\lambda^{(l)}} = \frac{1+x^{(1)}+Kd^2q_\perp^4/(2B)}{1+x^{(l)}+Kd^2{q_\perp}^4/(2B)} \approx \frac{1+x^{(1)}}{1+x^{(l)}}, \tag{3.13}$$

where [12]

$$x^{(1)} = -1+\frac{\alpha}{N}, \tag{3.14}$$

$$x^{(l)} = -\cos\frac{(l-1)\pi}{N} + \frac{2\alpha}{N}\cos^2\frac{(l-1)\pi}{2N}.$$

In particular, for $l=2$ and $q_\perp = 10^3$ cm^{-1} we get

$$\frac{\lambda^{(1)}}{\lambda^{(2)}} \approx \frac{\alpha N}{2\pi^2} = \frac{d\gamma q_\perp^2 N}{2B\pi^2} \sim 2N\times 10^{-8}. \tag{3.15}$$

For the modes with $l>2$ the ratio (3.13) is even less.

As it follows from Eqs. (3.2) and (3.13)–(3.15) the mode with $l=1$ has the largest amplitude and either the smallest characteristic frequency or the largest relaxation time. Therefore, in Eq. (3.1), we may take into account the first mode only for not too thick films. This is an acoustic mode that describes the synchronous motion of all smectic layers when the interlayer distances are constant. In this case the correlation functions $\langle u_n(\mathbf{q}_\perp,t)u_m(-\mathbf{q}_\perp,0)\rangle$ are equal for any layer numbers n and m. Thus the value $S(t)$, which according to Eq. (2.1) determines the intensity autocorrelation function $\langle I(t)I(0)\rangle$, has the form

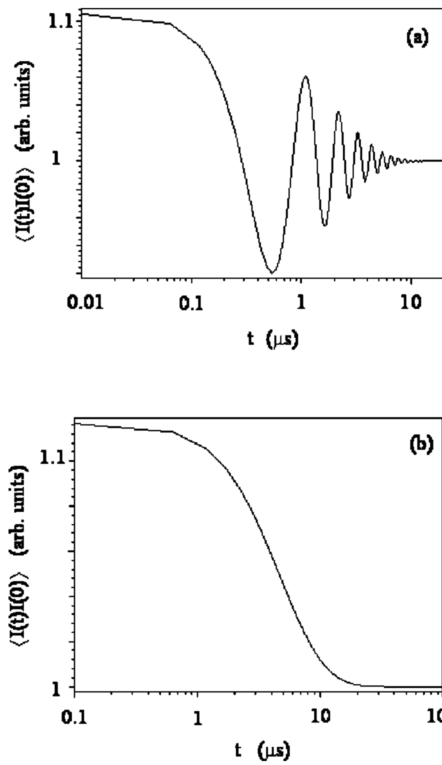

FIG. 5. Calculated time dependence of the scattering intensity autocorrelation function for $q_\perp = 10^3$ cm^{-1}, for the films of various thickness: (a) $N=6$, (b) $N=1000$.

$$S(t) = C(q_z)\Bigg[\int_0^\Lambda r_\perp J_0(q_\perp r_\perp)dr_\perp + \frac{k_BT\Lambda q_z^2}{4\pi\gamma}\frac{\omega_-^{(1)}\exp(-i\omega_+^{(1)}t)-\omega_+^{(1)}\exp(-i\omega_-^{(1)}t)}{\omega_-^{(1)}-\omega_+^{(1)}} \times \int_{2\pi/\Lambda}^{2\pi/a} f(q_\perp',q_\perp,\Lambda)dq_\perp'\Bigg], \tag{3.16}$$

where

$$C(q_z) = 2\pi\rho_s^2|\rho_M(q_z)|^2\sum_{n,m=1}^{N}\exp[-iq_z(n-m)d] \times\exp\left(-\frac{q_z^2}{2}[\langle u_n^2(0,0)\rangle+\langle u_m^2(0,0)\rangle]\right). \tag{3.17}$$

And the frequencies $\omega_\pm^{(1)}$ are equal to

$$\omega_\pm^{(1)} = \pm\left[\frac{2\gamma q_\perp^2}{\rho dN} - \left(\frac{n_3q_\perp^2}{2\rho}\right)^2\right]^{1/2} - i\frac{\eta_3 q_\perp^2}{2\rho}. \tag{3.18}$$

The value

$$q_\perp^{(M)} = \sqrt{(8\gamma\rho/\eta_3^2 dN)}$$

separates the regions of oscillating and relaxation motions for the acoustic mode. For the value $q_\perp \sim 10^3$ cm^{-1} in thin films with $N < (8\gamma\rho)/(\eta_3^2 d q_\perp^2) \sim 800$ the intensity temporal autocorrelation function in x-ray scattering experiments has to be oscillating whereas in thick films its behavior has to be relaxational.

IV. DISCUSSION

Figures 4 and 5 shows the correlation function $\langle I(t)I(0)\rangle$ for films of various thickness. Calculations have been performed according to Eqs. (2.1) and (3.16). Note that the first term in Eq. (3.16), $\int_0^\Lambda r_\perp J_0(q_\perp r_\perp) dr_\perp$, is very sensitive to the value of $\Lambda q_\perp$ contrary to the second term in Eq. (3.16). Since in the x-ray experiments the sum of the waves in the interval $q_{\perp\,\min} \leqslant q_\perp \leqslant q_{\perp\,\max}$ is recorded this term may be used as a fitting parameter in the experimental data processing. The second term in Eq. (3.16) describing the temporal dependence does not contain any fitting parameters. In Fig. 4 the fitting parameter is chosen to make consistent the numerical results with the experimental data for the 95-layer film [11]. Figure 5(b) shows the correlation function in the thick film. This function is a relaxation one and is consistent with the experimental results [6,8].

The essential role of the acoustic mode for the coherent x-ray scattering description was established. The estimates (3.13) and (3.15) for the ratio of amplitudes of various modes indicate that the possibility to restrict our consideration to an acoustic mode is justified only for $q_\perp$ being not too large,

$$q_\perp \ll \left(\frac{\gamma}{KdN}\right)^{1/2} \sim \frac{10^7}{\sqrt{N}}\,\mathrm{cm}^{-1}.$$

In particular this estimate shows that taking into account the acoustic mode only is valid for thin films. In this relation it is interesting to investigate the x-ray scattering from thin films with $N \sim 10-20$ where according to our calculations, the autocorrelation function $\langle I(t)I(0)\rangle$ should have an oscillating character with large amplitudes.

At the same time for thick films this approach is valid for rather small $q_\perp$. If $q_\perp \sim 10^4-10^5$ cm^{-1} the contribution of the remaining modes is essential. In this case one may expect that for such films the decay of the dynamic correlation function should be described by the wide relaxation spectrum.

The rather simple description of the dynamic x-ray scattering is based on the expansion (3.6). It is valid for not too large $q_z, q_z \sim 10^7$ cm^{-1}. If q_z considerably exceeds this value, the expansion (3.6) may be used for the times at which the correlation function $\langle u_n(r_\perp, t) u_m(0,0)\rangle$ is noticeably decreased. The initial behavior of the function $\langle I(t)I(0)\rangle$ may be numerically calculated by the general Eqs. (2.1), (2.6)–(2.9), and (3.1).

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