

Dedicated to the memory of B.K. Vainshtein

Structure and Thermal Fluctuations in Thin Films of Smectic Liquid Crystals

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Abstract—Thermal fluctuations of the smectic layers in freely suspended thin liquid crystalline films have been studied by the methods of X-ray diffraction and reflectometry. The fluctuation dynamics is studied by the methods of photon correlation spectroscopy with the use of a source of coherent synchrotron radiation. In freely suspended smectic films, the typical relaxation times are of the order of several microseconds. In thin liquid crystalline films, the simultaneous damped and oscillatory behavior was observed for the layer undulations. © 2001 MAIK “Nauka/Interperiodica”.

INTRODUCTION

Although liquid crystals (LCs) have already been known for more than a century, they were long considered as an amusing scientific incident having no direct relation to the development of physics of condensed matter. The dramatic change in the attitude to liquid crystals occurred in the early 1960s. At that time, it became evident that liquid crystals form a large variety of phases with unusual properties and fill the gap between solid crystals and isotropic liquids. Vainshtein was among the first scientists who started working along this direction. The systematic study of the structure of liquid crystals also started in the 1960s and became a natural continuation of the fundamental studies of the structure of chain molecules such as proteins, nucleic acids, rubbers, and other natural and synthetic polymers. The tendency to parallel packings of chain molecules stems from their geometry and results in anisotropy in their properties along and normally to the axis of their preferred orientation [1]. This opens the possibility to the study of the structure of liquid crystals proper.

In 1963, B.K. Vainshtein and I.G. Chistyakov published a series of articles on liquid crystals [2]. Unlike conventional three-dimensional crystals, whose structure can be described by the periodic density function $\rho(\mathbf{r})$, the structure of partly ordered systems such as liquid crystals requires the consideration of mutual correlations of the position and orientations of the mole-

cules, $\langle \rho(\mathbf{r})\rho(\mathbf{r}') \rangle$ [3, 4]. To describe the structure of a liquid crystal (LC), the so-called function of the intermolecular distances was introduced.¹ The methods for reconstructing this function from the X-ray scattering data in application to liquid crystals were developed in [2].

First, the structural studies of smectic phases were performed. In smectics, there is no translational invariance along the long axes of the molecules, which manifests itself in the one-dimensional density modulation in three-dimensional liquids. These systems exhibit strong thermal fluctuations. Ideal three-dimensional crystals are characterized by a true long-range order in the atomic (molecular) arrangement, whereas in the systems with the one- or the two-dimensional periodicity this order is destroyed by thermal fluctuations [3]. In the mid-1930s, Landau and Peierls showed that thermal fluctuations in the layer positions “smear” the long-range order in smectic LCs. However, this “smearing” is weak (logarithmic) so that the smectic samples of finite size remain stable. Nevertheless, there are differences between smectics and systems with the true long-range order. These differences can be seen from the decrease of the positional correlations at long distances by a power law. The relevant phenomena were thor-

¹ The function of the intermolecular distances, or the Patterson function, has maxima at the points corresponding to the most frequently repeated intermolecular distances. The physical sense of this function is analogous to that of the density–density correlation function $g_2(\mathbf{r})$, used in physics of condensed matter.

oughly studied in the precision X-ray experiments on the measurement of the structure factor of the smectic LCs [5–7]. However, these experiments provided the data solely on the static properties of the density correlation function. At the same time, almost no information on the dynamics of thermal fluctuations in the positions of the smectic layers was known until recently. The situation started improving only in the last few years, when the new methods of studying the dynamics of the disordered systems by means of the coherent X-rays were developed. If a coherent radiation is incident onto the fluctuating medium, the scattered radiation forms the so-called speckle pattern reproducing the instantaneous scatter location with regard for the phase relationships between them. The scatter motion gives rise to the corresponding changes in the speckle pattern, which thus can provide an information on the system dynamics. The photon correlation spectroscopy allows one to measure the time autocorrelation function of the intensity of the scattered radiation. For the visible light, the above technique (the dynamical light scattering) is well known [8]. For X-rays, the study of coherent dynamical scattering became possible only with the advent of the powerful sources of synchrotron radiation of the third generation [9, 10]. Unlike the dynamical light scattering, the X-ray correlation spectroscopy allows the study of the dynamical scattering on the molecular scale. These experiments provide valuable information on the characteristics of the relaxation processes in molecular ordered media and complement the data obtained in the structural studies.

Below, we report the results of the recent studies of the static and dynamic properties of freely suspended smectic films. Unlike the bulk samples, the smectic films are characterized by discrete spectra of relaxation modes of the layer displacements, which depend on the film thickness and surface parameters. In freely suspended smectic films, the characteristic relaxation times are on the order of several microseconds. In thin smectic films, the simultaneous damping and oscillatory of behavior was observed. The latter phenomenon is associated with the contribution provided by the surface to the free energy of the system. This contribution is most pronounced for thin films. We also consider the structure and the properties of polymer liquid crystalline films, and the prospects of the use of the dynamical X-ray scattering for studying polymers and biological and lipid membranes.

STATIC PROPERTIES OF SMECTIC FILMS

The formation of a smectic phase is associated with the appearance of the one-dimensional layer order in a liquid (Fig. 1). Such a structure can be obtained by translation of a layer along the z -axis for a distance corresponding to the layer period d (~ 2 – 3 nm). Now the question as to whether the one-dimensional order can be extended for an infinitely large distance arises. It is well known that even in solids, the thermal motion of

atoms gives rise to their displacement from the equilibrium positions. In smectics, thermal fluctuations result in much larger layer displacements from the given positions. These deviations are described with the use of the parameter $u_z = u(\mathbf{r})$, which characterizes the layer displacement along the layer normal (z -axis). In the harmonic approximation, the functional of the free energy of the smectic corresponding to the layer distortion has the form [11, 12]

$$F = \frac{1}{2} \int d^3 r \left[B \left(\frac{\partial u(r)}{\partial z} \right)^2 + K (\Delta_{\perp} u(r))^2 \right] + \frac{1}{2} \gamma \int d^2 r (\nabla_{\perp} u(r_{\perp}, z = \pm L/2))^2. \quad (1)$$

Here, the subscript “ \perp ” designates the directions lying in the smectic-layer plane; K and B are the volume elastic constants corresponding to bending and compression of the smectic layers, respectively; and γ is the surface-tension coefficient. The last term in Eq. (1) represents the surface contribution to the free energy of the smectic film of the thickness L [13, 14]. To obtain the spectrum of the eigenmodes of the bulk smectic sample, we have to pass to the Fourier components of displacements $u(\mathbf{r})$ and apply the equation

$$\langle u(\mathbf{q})^2 \rangle = k_B T / (Bq_z^2 + Kq_{\perp}^4). \quad (2)$$

Here, the term Bq_z^2 is associated with the mode of compression–dilation along the normal to smectic layers, while Kq_{\perp}^4 is associated with the bending mode (Fig. 1). We should like to draw attention to the absence of the term $\sim q_{\perp}^2$ in the denominator of Eq. (2). Physically, the reason is the absence of the crystalline ordering within the smectic layer, which permits sliding of the liquid layers. The characteristic features of the mode structure of the smectic described by Eq. (2) pro-

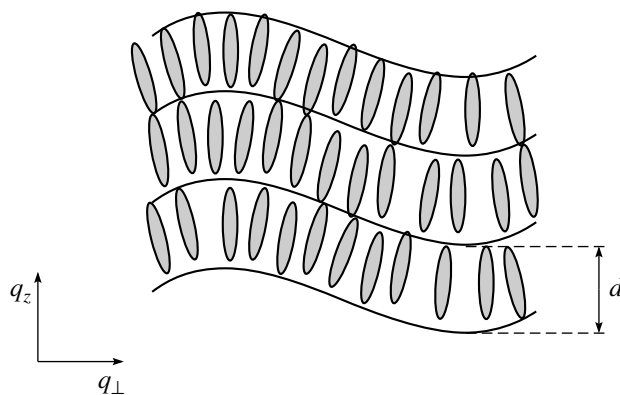


Fig. 1. The smectic A phase of liquid crystals. The bending mode is shown in which the layer period remains constant; q_z and q_{\perp} are the components of the wave vector along the layer normal and in the layer plane, respectively.

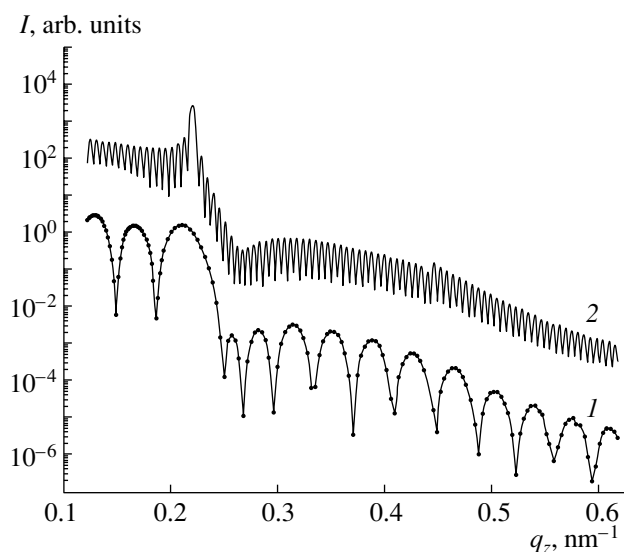


Fig. 2. The reflection curves of (1) 6- and (2) 43-layer-thick freely suspended films of the smectic liquid crystal 40.8. For clearer presentation, the curves are shifted by two decades.

vide instability of the smectic layers at long distances: at $\mathbf{r} - \mathbf{r}' \rightarrow \infty$, the correlator of the layer displacements $\langle u(\mathbf{r})u(\mathbf{r}') \rangle$ is divergent [3]. This signifies that the fluctuations in the positions of smectic layers smear the long-range order of an infinite system. Thus, the mean-square layer displacement,

$$\langle u^2(\mathbf{r}) \rangle = \frac{k_B T}{(2\pi)^3} \int \frac{d^3 q}{Bq_z^2 + Kq_\perp^4} = \frac{k_B T}{8\pi(KB)^{1/2}} \ln \frac{L}{d}, \quad (3)$$

diverges logarithmically with an increase of the sample dimension L . However, this divergence is weak and the amplitude of the layer displacement for laboratory samples does not exceed $\sigma = \langle u^2(r) \rangle^{1/2} \approx 0.4\text{--}0.7$ nm. In terms of the relative displacements, $\sigma/d \approx 15\%$; in other words, the layers do exist.

The characteristics of the static structure factor of smectic LCs can be studied directly in the X-ray experiments on freely suspended films of various thicknesses. In freely suspended smectic films, the layers are parallel to the plane of the opening which bounds the film [15]. The film is not broken down due to the elastic response to dilation (compression) of the layers along the surface normal to the film. Such films are highly homogeneous, although their thickness can vary from two to several hundreds of molecular layers.

Studying freely suspended smectic films, one uses the geometry of reflection at glancing angles (X-ray reflectometry). Within the range of the X-ray wavelengths used ($\lambda \approx 0.1$ nm), the refractive index n is less than unity: $n = 1 - \delta$ (for organic materials, $\delta \approx 3 \times 10^{-6}$). Despite the smallness of δ , the classical phenomena of reflection and interference at the air-LC interface are observed in the small-angle range. The reflection

curves $R(q_z)$ for freely suspended smectic films from a *N*-(4-*n*-butoxybenzylidene)-4-*n*-octylaniline (40.8) are shown in Fig. 2.² The reflection curve $R(q_z)$ falls down by the law $\sim q_z^{-4}$, determined by the Fresnel formulas. In addition, one also observes in freely suspended smectic films the simultaneous interference of the rays reflected from the front and the back surfaces (isoclinic fringes) and diffraction from the smectic layers. At a large number of the layers, one observes intense Bragg maxima at the reciprocal-lattice points $q_{zn} = 2\pi n/d$. The final reflection pattern from the freely suspended smectic film is the result of the interference of the isoclinic fringes and the Bragg peaks.

The positions of the interference maxima allow one to determine the total thickness of the freely suspended smectic film. The analysis of the reflection curves is based on the exact (matrix) solution of the Fresnel equations for reflection from a multilayer system [16]. The electron density of each smectic layer is determined by the convolution of the projections of the electron density of molecules onto the z -axis with the Gaussian of width σ_i , which describes the smearing of the centers of gravity of the molecules in the i th layers caused by thermal fluctuations of the layer positions [17–19]. The resulting distribution of the rms displacement amplitudes σ_i across the freely suspended films of different thicknesses in the smectic-*A* and crystalline-*B* phases of the 40.8 compound are shown in Fig. 3 (only one half of the centrosymmetric profile is presented). The fluctuations in the layer displacements at the free surfaces are suppressed irrespectively of their thicknesses. This is the consequence of the fact that the surface contribution to the free energy of the system prevails in the vicinity of the interface. The theoretical calculations made for a smectic film of finite dimensions show that the layer fluctuations at the free surface of the smectic are suppressed if $\nu = \gamma/(KB)^{1/2} > 1$ [13, 14]. For a 40.8 liquid crystal, the use of the elastic constants $K \approx 10^{-11}$ N and $B \approx 10^7$ N/m² and the surface-tension coefficient $\gamma \approx 0.02$ N/m yields $\nu = 2$, in close agreement with the experimental data. In the smectic *A* phase, the amplitude of thermal fluctuations at the film center increases with its thickness, which is the direct confirmation of the Landau–Peierls instability (3). Vice versa, upon the transition into the low-temperature crystalline *B* phase, the fluctuation amplitude in the center of the freely suspended smectic film stops being dependent on the thickness. It is consistent with the appearance of the term $C_{44}q_\perp^2$ in the dispersion law for the elastic modes of the three-dimensional crystal, which excludes the Landau–Peierls instability [3] (C_{44} is the shear elastic modulus in the xy plane).

² With lowering of the temperature, a 40.8 liquid crystal shows the following sequence of phases: I (78°C) N (63.5°C) SmA (48.5°C) CrB, where I, N, SmA, and B indicate isotropic, nematic, smectic A, and crystalline B phases, respectively.

DYNAMICS OF THERMAL FLUCTUATIONS

The above results concerned the static properties of the density correlation function in smectic films. What can be stated about the dynamics of thermal fluctuations of the smectic layers? The related problems can be solved with the use of the X-ray photon correlation spectroscopy in the X-ray wavelength range. Prior to the discussion of the experiment, consider the dynamics of the layer systems. A set of the hydrodynamic equations describing the dynamics of the smectic phases is considered by Kats and Lebedev [20] and de Gennes and Prost [11]. For the case of thermal fluctuations in the layer positions, the linearized hydrodynamic equation is written as

$$\rho_0 \frac{\partial^2 u(r)}{\partial t^2} = \eta_3 \frac{\partial}{\partial t} \nabla_{\perp}^2 u(r) + (B \nabla_z^2 - K \Delta_{\perp}^2) u(r), \quad (4)$$

where ρ_0 is the LC density and η_3 is the viscosity coefficient corresponding to shear of smectic layers with respect to one another. Equation (4) is, in fact, the second Newton law written for the layer displacements u along the layer normal (z -axis). The left-hand side of the equation describes inertia; the right-hand terms describe the viscous friction and the elastic force acting onto the smectic layers. In discussing the layer dynamics of the bulk LC samples, the term describing inertia can be omitted because of the smallness of the dimensionless parameter $\rho_0 K / \eta_3^2 \approx 10^{-4} - 10^{-6}$. As a consequence, the equation of smectic-layer motion is essentially simplified, while the time-dependent layer displacement correlation function proves to be overdamped and is described by a simple relaxation equation having the form [11, 20]

$$\langle u(\mathbf{q}, t) u^*(\mathbf{q}, 0) \rangle \sim \exp(-t/\tau_q) \quad (5)$$

with the characteristic relaxation times

$$\tau_q = \eta_3 q_{\perp}^2 / (B q_z^2 + K q_{\perp}^4). \quad (6)$$

According to Eq. (6), the spectrum of the relaxation modes of a bulk smectic is continuous. Considering the dynamic properties of the smectic films of finite dimensions one also has to take into account the appropriate boundary and initial conditions. Then the spectrum of the relaxation modes becomes discrete. In the long-wavelength limit ($q_{\perp} \rightarrow 0$), it has the only relaxation time [21, 22]

$$\tau_1 = \eta_3 L / 2\gamma. \quad (7)$$

At the typical values of the quantities in Eq. (7), $\eta_3 \approx 0.04$ kg/m s and $\gamma \approx 0.02$ N/m, and the film thickness L of the order of a micron (10^{-6} m), we arrive at the characteristic relaxation times of the order of several microseconds. Equation (7) was verified for thick freely suspended smectic films in experiments on the dynamical scattering of light in the range of soft X-rays [23].

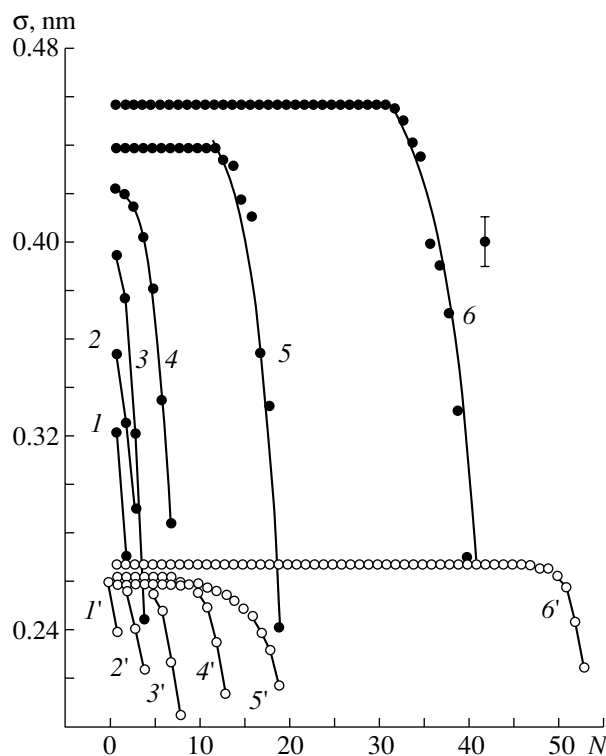


Fig. 3. Fluctuation profiles of the smectic *A* phase films at $\sim 63^\circ\text{C}$ (filled circles) consisting of (1) 5, (2) 6, (3) 8, (4) 15, (5) 38, and (6) 80 layers and in the crystalline *B* phase films of the compound 40.8 at $\sim 47^\circ\text{C}$ (open circles) consisting of (1') 3, (2') 8, (3') 16, (4') 26, (5') 38, and (6') 107 layers, obtained by simulation of the reflection curves [N is the layer position (its number)].

In thin freely suspended smectic films, the dynamics of layer fluctuations was studied by means of the photon correlation spectroscopy with the use of hard (8 keV) X-ray radiation [24]. The measurements were made on the undulator beam line ID10A (Troika I) at the European Synchrotron Radiation Facility (ESRF, Grenoble). To obtain the coherent radiation from the undulator (a partly coherent source), one has to provide the validity of the monochromatic plane-wave approximation at the site of sample location. Therefore, the size of the first slit placed at a distance of $R = 46$ m from the sample did not exceed $s = 0.9$ mm. This provided the so-called transverse coherence length (along the wave front) $\xi_{th} = (\lambda R / 2s) \approx 4$ μm in the horizontal (scattering) plane [25]. In the vertical direction, the transverse coherence length ξ_{tv} was limited by the collimating aperture 12 μm in diameter (Fig. 4a). An analyzing 40×40 μm^2 -large aperture was placed in front of the detector at a distance of 1.2 m from the sample. The monochromator was a Si (111) crystal with the transmission $\Delta\lambda/\lambda \approx 1 \times 10^{-4}$. The $\sim 15\%$ coherence was determined from contrast measurements of the Fraunhofer diffraction pattern from the collimating aperture taken in the absence of the sample.

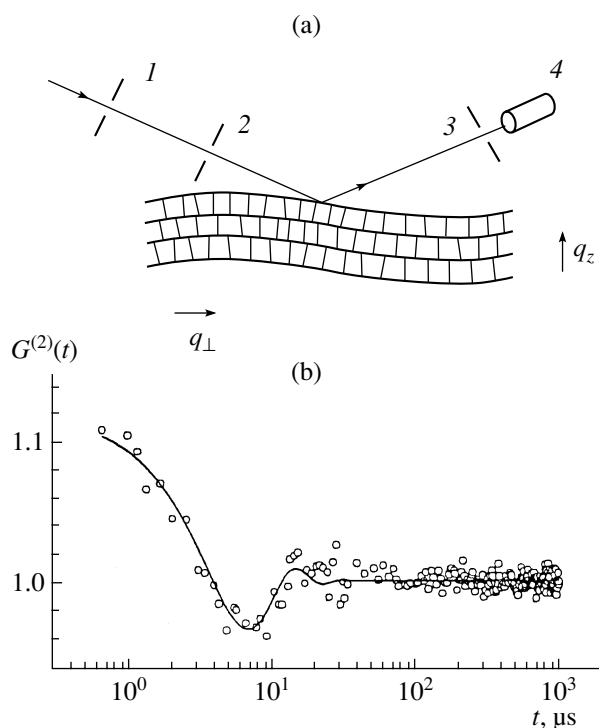


Fig. 4. (a) Scheme of the experiment for measuring dynamical structure factor of smectic LCs with the use of coherent X-ray radiation: (1) the collimating aperture providing the coherent component of the beam, (2) the guard slit protecting from the parasitic scattering from the edges of the previous slit, (3) the detector or “analyzing” aperture, and (4) the detector. (b) The normalized autocorrelation time function for the freely suspended smectic liquid-crystal film consisting of 95 layers (~ 0.3 nm) at $T \approx 50^\circ\text{C}$; the solid line is the approximation by model (8).

The intensity–intensity time correlation function $G^{(2)}(q, t) = \langle I(\mathbf{q}, 0)I(\mathbf{q}, t) \rangle$ was measured with the aid of a fast digital correlator at the reciprocal-space point $q_z = 2\pi/d \approx 2.2$ nm $^{-1}$ corresponding to the first Bragg peak of the layer structure. Under the conditions of our experiment, the function $G^{(2)}(q, t)$ is specified by the time-dependent layer displacement–displacement correlation function $G(\mathbf{q}, z, z', t) = \langle u(\mathbf{q}_\perp, z, 0)u^*(\mathbf{q}_\perp, z', t) \rangle$. The exact expression for the function $G(\mathbf{q}, z, z', t)$ is given in [22]. Despite the considerable losses of intensity associated with the beam collimation, the coherent-scattering volume $(\xi_{\text{coh}}/\sin\theta_B) \times \xi_{\text{sv}} \times L \approx 500$ μm^3 was sufficient for the reliable recording of the time autocorrelation function to the microsecond time range (θ_B is the Bragg angle of beam incidence onto the sample $\sim 1.5^\circ$).

The time autocorrelation function $G^{(2)}(t)$ measured on the freely suspended film of the 40.8 compound in the smectic A phase is shown in Fig. 4b. The function $G^{(2)}(t)$ shows the decay of the fluctuations in the layer displacements on the microsecond time scale and, at the same time, the oscillatory behavior with the typical

times on the order of ten microseconds. The measurements are well described by the formula

$$G^{(2)}(t) = 1 + A \cos(\omega t) \exp(-t/\tau), \quad (8)$$

where the amplitude A is determined by the degree of coherence, whereas the relaxation and the oscillation times τ are equal to ≈ 6 and $2\pi/\omega \approx 16$ μs , respectively.

It should be emphasized that in our experiments, the projection of the wave vector onto the layer normal was equal to $q_z = 2\pi/d \approx 2.2$ nm $^{-1}$, which exceeds by a factor of 100 the range of the wave vectors in the experiments on the dynamic scattering of visible light. At the same time, the range of the wave vectors $q_\perp = 2\pi/r_\perp$ corresponding to the layer-bending mode was much longer—from r_\perp close to the molecular dimensions to the macroscopic values of the order of tens of micrometers and was determined mainly by the diffractometer resolution along q_\perp ($\approx 10^{-4}$ nm $^{-1}$).

At first glance, the oscillatory mode in the dynamics of thermal fluctuations of the smectic layers in the freely suspended films seems to be inconsistent with the theoretical conclusions and the experimental results [23]. However, this is only an apparent contradiction. The point is that the neglect of the term describing inertia in Eq. (4) is justified only if $\rho_0 K/\eta_3^2 \ll 1$. For bulk samples, this parameter is really very small ($\sim 10^{-4}$ – 10^{-6}), because the expression of free energy has no term proportional to q_\perp^2 . However, for thin freely suspended films, the situation is different because of the presence of the additional surface contribution, γq_\perp^2 [13, 14]. In the static case, this leads to suppression of the amplitudes of the layer fluctuations at the free surface of liquid crystals. In dynamics, the surface contribution prevails over the energy of the bending mode Kq_\perp^4 in the long-wavelength limit ($q_\perp \rightarrow 0$), and the inertia term becomes more important [21, 22]. The estimations show that the transition from the surface to the bulk mode takes place at the limiting value $q_{\perp c} \approx 2 \times 10^{-3}$ nm $^{-1}$. With due regard for the inertia term in Eq. (4), the reciprocal relaxation time corresponding to the fundamental mode of the freely suspended smectic film can be written as follows [22]:

$$\frac{1}{\tau_1} = \pm i \left(\frac{2\gamma}{\rho_0 L} \right)^{1/2} q_\perp + \frac{\eta_3}{2\rho_0} q_\perp^2. \quad (9)$$

Thus, the relaxation time of the layer fluctuations becomes complex for $q_\perp < q_{\perp c}$, with its real part providing attenuation and the imaginary part being responsible for oscillations of the time correlation function. Since the instrumental resolution in the experiment is of the same order of magnitude as $q_{\perp c}$, the oscillations of the $G^{(2)}(t)$ function should be observable for rather thin films as well (Fig. 4b).

THE PROSPECTS FOR FURTHER RESEARCH

The experiments performed showed that the methods of the photon correlation spectroscopy based on the coherent radiation sources can be extended to the X-ray wavelengths. This offers the unique opportunity to study the comparatively fast dynamic processes in the structured organic films on the molecular distance scale. It should be remembered that Raman and Mandelstam–Brillouin scattering in the visible range provide an information on the long-wavelength ranges of the phonon spectra of the crystals. These methods are used to study fast processes on the atomic and molecular level (10^7 – 10^{14} Hz). To study comparatively slow processes (1 – 10^6 Hz) with the characteristic scale of the order of the wavelength of the visible light, the dynamical light scattering is widely used. In particular, it was used in the study of thermal fluctuations of the director orientation in nematic liquid crystals [11]. To study fast processes in the condensed media (10^9 – 10^{12} Hz) on the scale of molecular length, one traditionally uses inelastic neutron scattering. The dynamical X-ray scattering allows one to study rather slow processes (10^{-2} – 10^7 Hz) with the characteristic inhomogeneity scale of the molecular length (10^{-1} – 10^3 nm).

The promising objects for studying the dynamic processes on a molecular scale are polymer liquid crystals in which the LC fragments are attached to the main polymer chain via flexible spacers. These phases are the unique combinations of the liquid-crystal order with specific polymer properties. The flexible main polymer chain and the spacers provide the necessary degree of freedom for LC fragments for the self-assembly into the LC phases of various symmetry, including the smectic phases [26]. At present, little is known about the dynamics of such systems, although it can provide an extremely important information on the effect of the conformation of the main polymer chain on the liquid crystalline ordering. The studies of the dynamics of the surfactant solutions forming lyotropic lamellar phases are also of great interest. These are model membranes based on the aqueous phospholipid solutions forming the lipid bilayers alternating with aqueous interlayers [27, 28]. Another important direction is the dynamics of lipid membranes with the imbedded DNA molecules. In particular, linear DNA molecules and certain mixtures of the cationic liposomes can be self-assembled into two-dimensional smectic phases imbedded into lipid bilayers [29]. These systems allow the simulation of DNA transport via cellular membranes. There are some other types of self-assembly of molecules, including those resembling smectic membranes, which exist in amphiphilic associating polyelectrolytes [30, 31]. These substances are characterized by mutually excluding properties provided by the presence of the antagonistic hydrophilic and hydrophobic groups in one polymer. Thin films of such polymers are obtained from the aqueous solutions of polyelectrolytes.

Concluding the article, we should like to emphasize that our knowledge about the dynamic behavior of comparatively simple smectic films is still far from exhaustive. With the enrichment of our knowledge about these films, the focus of such studies would be shifted toward more complicated membrane systems important for biology.

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