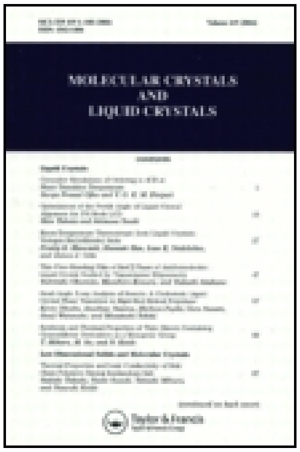

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Theory of Evanescent Light Wave Scattering at the Solid-Nematic Interface

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THEORY OF EVANESCENT LIGHT WAVE SCATTERING AT THE SOLID-NEMATIC INTERFACE

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Abstract The field correlation function of light scattered from the evanescent wave at a solid-nematic interface is deduced. Its perpendicular part has a nonexponential tail falling off as $t^{-3/2}$ and depends on the anchoring energy. Different possible orientations at the surface are discussed.

INTRODUCTION

The interaction of a nematic liquid crystal with an interface is an important problem which is not well understood at the present time[1]. In a phenomenological approach, one can characterize the interaction of the nematic liquid crystal with the surface by a surface anchoring potential, where one assumes that there exists a preferred direction (easy direction) of the nematic director, $\vec{n}_0$ at the surface, which minimizes the surface interaction energy. Usually, the surface potential is assumed to be parabolic around the easy direction, and the corresponding coefficient is called the anchoring energy[2].

The surface anchoring potential determines the boundary conditions for a confined nematic sample, and so affects both the static configuration and the dynamics of the sample. In particular, close to the interface, the thermally excited fluctuations of the nematic director should be influenced by the surface anchoring effects.

The nematic orientational fluctuations are strongly coupled to the optical dielectric tensor and therefore cause strong scattering of light[3]. The total intensity of light scattered in a thin layer of liquid crystal at small angles depends on the anchoring energy and has been studied in ref. [4]. The fluctuations close to the surface can also be specifically probed by the technique of evanescent wave scattering[5,6,7],

*On leave from Dept. of Physics, University of Ljubljana, Slovenia where one detects the light scattered from the exponentially decaying electric field in the region of lower index of refraction when a light beam is totally reflected from the interface. The correlation function of the scattered light, which is commonly measured in light scattering experiments, should contain some information on the surface anchoring energy.

Dynamic light scattering was also used in ref. [8], where the intensity fluctuations in a leaky optical slab wave-guide were measured. The analysis, however, failed to take into account the fact that a given wave- guide mode is excited by scattering from many liquid crystal modes, leading to a nonexponential correlation function.

In this paper we want to present the calculation of the scattered light field correlation function due to the evanescent wave at a solid-nematic interface. We will consider all the possible modes of fluctuations in both pure orientations, one with the director in the plane of the interface and the other with the director perpendicular to the surface.

ORIENTATIONAL FLUCTUATIONS AT THE SURFACE

In our treatment, we will describe the nematic with the director field only and will neglect the coupling to the shear velocity of the nematic material. The introduction of this coupling makes the computation much more involved, and its effect is only to change the effective viscosity entering the relaxation frequency of the fluctuations[3].

1. Parallel Orientation, Splay-bend Mode

First, let $\vec{n_0}$ at the surface be parallel with the surface, along the $\hat{x}$ direction and let the normal to the surface be $\hat{z}$. Two polarizations of the director fluctuations $\delta\vec{n}$ are possible, splay-bend, where $\delta\vec{n}$ lies in the xz plane, and the twist-bend with $\delta\vec{n}$ in the y direction.

The splay-bend mode can be completely specified with the angle θ between the instantaneous local director and the x axis. The elastic free energy of the system can then be written in the form

$$F = \frac{w_1}{2}\int \theta^2(x,0)dx + \frac{1}{2}\int \left[K_3(\frac{\partial\theta}{\partial x})^2 + K_1(\frac{\partial\theta}{\partial z})^2\right] dxdz \ , \qquad (1)$$

where K_1 and K_3 are the splay and bend elastic constants and w_1 is the surface anchoring energy for the rotation of the director perpendicular to the surface. This free energy leads to the equations of motion

$$K_3\frac{\partial^2\theta}{\partial x^2}+K_1\frac{\partial^2\theta}{\partial z^2}=\gamma_1\frac{\partial\theta}{\partial t} \tag{2}$$

where γ_1 is an effective viscosity for the splay-bend mode. We will neglect that, due to the coupling with the velocity field, it is also a function of the direction of the wavevector. The boundary conditions at the surface are

$$\frac{\partial\theta}{\partial z}(x,0)=\lambda_1\theta(x,0) \tag{3}$$

where we have introduced an inverse anchoring length $\lambda = w_1/K_1$.

We seek solutions of 2 and 3 having the form

$$\theta=\eta(z)\exp(iqx)\exp(-\frac{t}{\tau}) \ . \tag{4}$$

By substituting 4 into 2 and taking into account the boundary conditions 3, we get

$$\eta(z)=A_{q\beta}[(\beta+i\lambda)\exp(-i\beta z)+(\beta-i\lambda)\exp(i\beta z)] \ . \tag{5}$$

The relaxation time τ is related to parameters β and q through

$$\frac{1}{\tau}=\frac{1}{\gamma}(K_1\beta^2+K_3q^2) \ . \tag{6}$$

We are interested in the thermally excited fluctuations, so the average square of the amplitudes $A_{q\beta}$ can be calculated with the use of equipartition theorem:

$$\langle|A_{q\beta}|^2\rangle=\frac{Vk_BT}{(K_1\beta^2+K_3q^2)(\beta^2+\lambda_1^2)} \ . \tag{7}$$

In the derivation of eq.7, the surface term in the free energy gives an infinitesimally small contribution, so the presence of the boundary does not affect the magnitude of the fluctuations at given β and q but only causes the phases of the modes with $+\beta$ and $-\beta$ to have a definite relation. The additional factor $(\beta^2+\lambda_1^2)$ in the denominator of eq.7 is the consequence of the definition of $A_{q\beta}$ in eq.5.

2. Twist-bend Mode

In the case of the twist-bend mode, $\delta\vec{n}$ is perpendicular to the xz plane, and can be described by an angle ϕ that the local director makes with the x axis in the xy plane. The relevant free energy is

$$F_{TB}=\frac{w_2}{2}\int\phi^2(x,0)dx+\frac{1}{2}\int\left[K_3(\frac{\partial\phi}{\partial x})^2+K_2(\frac{\partial\phi}{\partial z})^2\right]dxdz \ . \tag{8}$$

Here K_2 is the twist elastic constant and w_2 is the anchoring energy for the rotation of the director parallel to the surface. So the problem is formally the same as in the splay-bend case, with a different inverse anchoring length $\lambda_2 = w_2/K_2$ and with K_2 in place of K_1.

3. Homeotropic Orientation at the Surface

Let us also discuss briefly the case when $\vec{n_0}$ is perpendicular to the surface. Again, both the splay-bend and twist-bend modes are possible. There is just one anchoring energy w_3, and the relevant free energy is

$$F_i = \frac{w_3}{2}\int \theta_i^2(x,0)dx + \frac{1}{2}\int \left[K_i(\frac{\partial\theta_i}{\partial x})^2 + K_3(\frac{\partial\theta_i}{\partial z})^2\right] dxdz \quad ,i = 1,2 \tag{9}$$

where 1 designates the splay-bend mode and 2 the twist-bend mode. The inverse anchoring lenght is $\lambda_3 = w_3/K_3$ and expressions 6 and 8 are replaced by

$$\frac{1}{\tau} = \frac{1}{\gamma}(K_3\beta^2 + K_i q^2) \tag{10}$$

and

$$\langle|A_{q\beta}|^2\rangle = \frac{Vk_BT}{(K_3\beta^2 + K_i q^2)(\beta^2 + \lambda_3^2)} \quad . \tag{11}$$

EVANESCENT WAVE SCATTERING

The director fluctuations give rise to fluctuations of the optical dielectric tensor which causes strong scattering of light. The change in the dielectric tensor is to the first order in the deviations of the director[3]

$$\delta\underline{\epsilon} = \epsilon_a(\vec{n_0} \otimes \delta\vec{n} + \delta\vec{n} \otimes \vec{n_0}) \tag{12}$$

where ϵ_a is the anisotropy of the dielectric tensor. For the splay-bend mode, $\delta\vec{n}$ is in the plane of the unperturbed director $\vec{n_0}$, which is also the direction of the optic axis, and will cause strong scattering when both the incoming and scattered light are extraordinarily polarized. The twist-bend mode causes ordinary-extraordinary scattering.

Let the index of refraction of the solid be larger than the indices of the liquid crystal. Then a light beam impinging upon the boundary at a large enough angle

α in the xz plane and polarized in the incident plane will be totally reflected. In the liquid crystal, the evanescent electric field will be extraordinarily polarized and will have the form

$$E_e(\vec{r},t) = E_0 \exp i(k_0 x \sin\alpha - \omega_0 t) \exp(-\kappa z) \tag{13}$$

where the penetration parameter κ is given by

$$\kappa = k_0(\sin^2\alpha - \sin^2\alpha_c)^{\frac{1}{2}} \ . \tag{14}$$

k_0 is the magnitude of the wavevector in solid and α_c is the angle of total internal reflection for the particular incoming polarization.

Consider first the splay-bend mode with planar orientation. The amplitude of the extraordinarily polarized light scattered in some direction $\vec{k_s} = (k_{||}, 0, k_\perp)$ in the liquid crystal is

$$E_s = C \exp -i\omega_0 t \int E_e(\vec{r}) \exp(-\vec{k_s}\cdot\vec{r})\, \theta(\vec{r},t) d\vec{r} \ . \tag{15}$$

The scattered field correlation function is

$$\begin{aligned} \langle E_s(0)E_s^*(t)\rangle &= C^2E_0^2G(t) = \\ &= C^2E_0^2 \int \exp[-\kappa(z+z')] \exp[i(k_0\sin\alpha - k_{||})(x-x') - ik_\perp(z-z')] \times \\ &\quad \times \langle\theta(\vec{r},0)\theta^*(\vec{r},t)\rangle d\vec{r}d\vec{r}' \ . \end{aligned} \tag{16}$$

The integration over x and x' just selects one spatial Fourier component of the correlation function of θ in the x direction:

$$G(t) = \int_0^\infty \int_0^\infty \exp[-\kappa(z+z') - ik_\perp(z-z')] \langle\theta(z,q,0)\theta^*(z',q,t)\rangle dzdz' \tag{17}$$

where $q = k_0 \sin\alpha - k_{||}$. Using eq.5, we get

$$G(t) = \int \langle|A_{q\beta}|^2\rangle I(\beta) \exp[-t/\tau(\beta)] d\beta \tag{18}$$

where

$$\begin{aligned} I(\beta) &= \int_0^\infty \int_0^\infty \exp[-\kappa(z+z') - ik_\perp(z-z')] \times \\ &\quad \times \left\{(\beta^2+\lambda_1^2)\left[\exp i\beta(z-z') + \exp -i\beta(z-z')\right] + \right. \\ &\quad \left. +(\beta+i\lambda_1)^2 \exp -i\beta(z+z') + (\beta-i\lambda_1)^2 \exp i\beta(z+z')\right\} dzdz' \\ &= \frac{4\beta^2[(\kappa+\lambda_1)^2 + k_\perp^2]}{(\kappa^2+k_\perp^2+\beta^2)^2 - 4\beta^2k_\perp^2} \ . \end{aligned} \tag{19}$$

Putting together eqs. 11, 18, and 19, we finally get

$$G(t) = 4Vk_BT\exp\left(-\frac{K_3}{\gamma}q^2t\right)\times \times\int_0^\infty \frac{\exp(-\frac{K_1}{\gamma}\beta^2t)\beta^2[(\kappa+\lambda_1)^2+k_\perp^2]}{(K_1\beta^2+K_3q^2)(\beta^2+\lambda_1^2)[(\kappa^2+k_\perp^2+\beta^2)^2-4\beta^2k_\perp^2}]d\beta \ . \quad (20)$$

The twist-bend mode contributes to scattering in the ordinary-extraordinary (or e-o) polarizations and the final expression for the field correlation function is very similar, with K_1 replaced by K_2 and λ_1 by λ_2. Similar replacements are to be made in the case of homeotropic orientation of the liquid crystal at the surface.

Expression 20 is rather cumbersome, but can be simplified in the limits of small and large penetration parameter κ. When $\kappa \ll k_\perp$ and λ_i, that is, with large penetration depth very close to the angle of total internal reflection, the main contribution to the field correlation function comes from the pole of $I(\beta)$ at $k_\perp + i\kappa$. $I(\beta)$ can be approximated with

$$I(\beta) \approx \frac{\pi\beta^2[(\kappa+\lambda)^2+k_\perp^2]}{\kappa k_\perp^2}\left[\delta(\beta+k_\perp)+\delta(\beta-k_\perp)\right] \quad (21)$$

so that

$$G(t) \approx \frac{Vk_BT}{\kappa(K_1q^2+K_3k_\perp^2)}\exp\frac{(K_1q^2+K_3k_\perp^2)}{\gamma}t \quad (22)$$

which is essentially the same as in the ordinary bulk scattering.

In the limit of $\kappa \gg k_\perp$ and if also $K_1/\gamma t \gg 1/q^2$ and $1/\lambda^2$, one can make an approximation

$$\beta^2\exp-\frac{K_1}{\gamma}\beta^2t \approx \frac{1}{4}\sqrt{\frac{\pi\gamma}{K_1t^3}}\,\delta(\beta-\sqrt{\frac{\gamma}{K_1t}}) \quad (23)$$

and

$$G(t) \approx \sqrt{\pi}k_BT\frac{K_1}{K_3q^2}\left(\frac{\gamma}{K_1t}\right)^{\frac{3}{2}}\exp(-\frac{K_3}{\gamma}q^2t) \ . \quad (24)$$

Eq. 24 shows that the perpendicular part of the correlation function has a nonexponetial tail at long times.

DISCUSSION

The integral in eq. 20 as a function of t and κ is shown in the Figure. Its nonexponential nature is readily apparent, especially the slowly falling tail at long times that goes as $t^{-3/2}$. This can be compared with the results of ref. [5]; there, however, the complete expression for the scattered field correlation function is considerably simpler and falls off at long times only as $t^{-1/2}$. Our result is different because in the

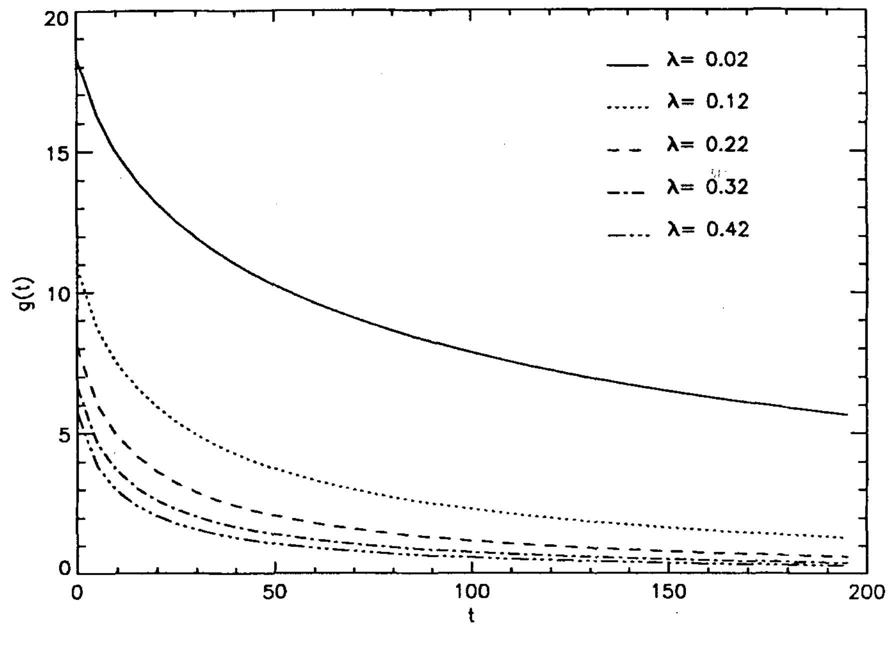

Figure 1: The integral in eq. 20 as a function of time (in units of $\gamma/(K_1 k_0^2)$) at several values of λ. $q = 0.1k_0$, $k_\perp = 0.2k_0$, $\kappa = 0.5k_0$, and $K_1/K_3 = 1$.

case of the fluctuations of the nematic director there exsits an elastic restoring force which is absent in the case of free particle diffusion. Also, the boundary conditions at the surface are different, and give an additional parameter of the problem - the inverse anchoring lenght λ.

The shape of the scattering correlation function depends on the wavevector components q and $k_\perp$, on the ratio of the elastic constants K_1/K_3 and on the anchoring parameter λ_i. q and $k_\perp$ are determined by the incoming and scattering angle and by the indices of refraction. The elastic constants and viscosities are known or can be obtained from other experiments, so in principle, the evanescent wave scattering makes possible the determination of the anchoring parameters λ_i. By choosing different scattering geometries and different surface orientations it is possible to get the anchoring energies for different displacements of the nematic director at the surface in a very direct and unambiguous manner.

The difficulty in doing the analysis of experimentally obtained scattered light correlation function is that the dependence of the integral in eq. 20 on λ_i is not very pronounced so that high quality data would be needed to determine λ_i with a reasonable precision.

To conclude, we have obtained the light field correlation function for the evanescent wave scattering at the interface of a high index of refraction transparent solid and nematic liquid crystal. This kind of scattering is being used to obtain the anchoring energy of the nematic director at the surface[9].

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