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LIGHT SCATTERING BY THIN NEMATIC LIQUID CRYSTAL FILMS

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Abstract The scattering of light by thin nematic liquid crystal films is studied. The distribution of fluctuation wavevectors relevant to the scattering is peaked around the difference between the wavevectors of the scattered and the incident light ray. The width is inversely proportional to the film thickness. Furthermore, only a discrete set of fluctuation wavevectors is allowed due to restrictions imposed by the boundary conditions. In the case of weak anchoring the relaxation time is a linear function of the thickness for sufficiently small film thicknesses. Similar results may be expected for other confined nematic systems, e.g. for nematics confined to porous media.

INTRODUCTION

Light scattering techniques are very important for the research of liquid crystals in thin films[1–4] and in porous media[5–12]. Finite size effects may be quite important for the description of light scattering by nematic liquid crystals in a confined geometry. In order to account for these finite size effects we propose some modifications of the theory of light scattering for bulk nematics[13,14]. In this paper we present a discussion of our results which is predominantly qualitative. A more detailed treatment has been published elsewhere recently[15].

SCATTERING CONDITION

The strong scattering of light by nematic liquid crystals is due to the thermal fluctuations of the dielectric susceptibility tensor. Consider the scattering of incoming light with wavevector $\underline{k}_i$ to scattered light with wavevector $\underline{k}_f$ by a fluctuation with wavevector $\underline{q}$. We approximate the shape of the liquid crystal film of thickness d by that of a rectangular box with length, width and height $\sqrt{V/d}$, $\sqrt{V/d}$, and d, respectively, where V is the volume of the liquid crystal. In the Born approximation, the amplitude of the scattered wave is then proportional to the integral

$$\int \mathrm{d}^3 r\, e^{i(\underline{q}+\underline{k}_i-\underline{k}_f)\cdot\underline{r}} = V \frac{\sin(k_x\sqrt{V/d}/2)}{k_x\sqrt{V/d}/2} \frac{\sin(k_y\sqrt{V/d}/2)}{k_y\sqrt{V/d}/2} \frac{\sin(k_z d/2)}{k_z d/2}, \tag{1}$$

with $\underline{k} \equiv \underline{q} + \underline{k}_i - \underline{k}_f$. This integral differs significantly from zero for values of $\underline{k}$ close to zero. The width is proportional to $\sqrt{d/V}$ for k_x and k_y and to $1/d$ for k_z. If V is sufficiently large it follows that only those fluctuations contribute to the scattering that satisfy

$$\left.\begin{aligned} q_x &= k_{fx} - k_{ix}, \\ q_y &= k_{fy} - k_{iy}, \\ |q_z + k_{iz} - k_{fz}| &\leq \pi/d. \end{aligned}\right\} \tag{2}$$

Clearly, the scattering condition $\underline{q} = \underline{k}_f - \underline{k}_i$ is modified for thin liquid crystal films.

ORIENTATIONAL FLUCTUATIONS

The fluctuations of the dielectric susceptibility tensor are dominated by the orientational fluctuations of the liquid crystal. In de Gennes' theory orientational fluctuations are represented by small perturbations of the equilibrium director field.[13,14] According to the hydrodynamic equation for the director field these small perturbations can be expressed as

$$\delta\underline{n} = \underline{A}\, e^{i\underline{q}\cdot\underline{r}}\, e^{-t/\tau}, \tag{3}$$

with $\underline{A}$ the (small) amplitude, $\underline{q}$ the wavevector, and τ the relaxation time. The last two quantities are related by

$$\tau = \gamma/K\, q^2, \tag{4}$$

where K is an effective elastic constant and γ an effective viscosity. For infinitely large liquid crystals $\underline{q}$ can take any value, for confined liquid crystals $\underline{q}$ is restriced to a set of discrete values. This set is determined by the boundary conditions for the director field. This means that orientational fluctuations in confined liquid crystals are standing waves rather than traveling waves. The discrete modes have wavevectors depending on the size and shape of the confined liquid crystal. Consequently, also the relaxation times of the different modes depend on size and shape. For thin films only the component q_z is quantized. In the case of strong anchoring [4] (the director is fixed to a preferred orientation at the boundaries) the allowed values of q_z are multiples of π/d. The relaxation time of the mode with lowest q_z is then given by

$$\gamma\tau^{-1} = K\left(q_x^2 + q_y^2\right) + \pi^2\frac{K}{d^2}. \tag{5}$$

This fluctuation mode can be probed in a scattering experiment with $k_{iz} = k_{fz}$. When d is sufficiently small the relaxation time is quadratic in d. A different dependence on film thickness is found when weak anchoring is assumed (the director is allowed to deviate from the preferred orientation at the boundaries). This dependence can be estimated as follows. Small perturbations of the equilibrium orientation lead to an increase in elastic free energy of the order $K\, q_z^2 d/2$ and to an increase in surface energy of the order of the anchoring constant C. Assuming that these two contributions are balanced leads to

$$K\, q_z^2 \approx \frac{2C}{d}, \tag{6}$$

which results in a relaxation time given by

$$\gamma\tau^{-1} \approx K\left(q_x^2 + q_y^2\right) + 2\frac{C}{d}. \tag{7}$$

For sufficiently small d the relaxation time is linear in d, in contrast to the case of strong anchoring. Experiment[16] indicates a linear dependence for small d, which supports the weak anchoring assumption. Weak anchoring effects are important in the regime of thicknesses smaller than the so-called extrapolation length K/C. For that reason a cross-over from the C/d term to a K/d^2 term for higher values of d may be expected. Probably the effective relaxation time of nematics confined to random porous media depends on the average pore size in a similar way.[7,11,12]

Qualitatively, orientational fluctuations in nematics that are deformed in equilibrium behave similar to nematics that are not deformed in equilibrium. Quantitatively, however, there are some differences. We compared a nematic film with two planarly anchoring substrates and a nematic film with one planarly anchoring substrate and one homeotropically anchorig substrate. Our model[15] is based on the one-constant approximation for the elastic free energy and the Rapini-Papoular approximation for the anchoring free energy. It appears that fluctuations in the plane of the equilibrium director field (tilt) and fluctuations perpendicular to this plane (twist) are described by the same equations for the planarly aligned film, whereas they are described by quite different equations for the hybridly aligned film. The competition between the elastic energy and the anchoring energy in hybridly aligned films leads to another difference. A director profile that has the preferred orientation at the boundaries gives rise to an elastic free energy contribution of the order K/d, whereas a uniformly planar director profile gives rise to an anchoring free energy contribution of the order C_2 (the anchoring constant of the homeotropically anchoring substrate). When d is sufficiently small a uniformly planar director field is favoured (assuming, of course, that C_2 is maller than the anchoring constant C_1 of the planarly anchoring substrate). In fact, at $d = d_c = K/C_2 - K/C_1$ a second order transition to the uniformly planar director field takes place[17]. This non-analytical behaviour introduces a kink at $d = d_c$ in the relaxation times of the different fluctuation modes. Figure 1 shows the relaxation times of the fluctuation mode with smallest q_z for the planarly aligned film and the hybridly aligned film as a function of the film thickness according to the theory of Stallinga and co-workers[15].

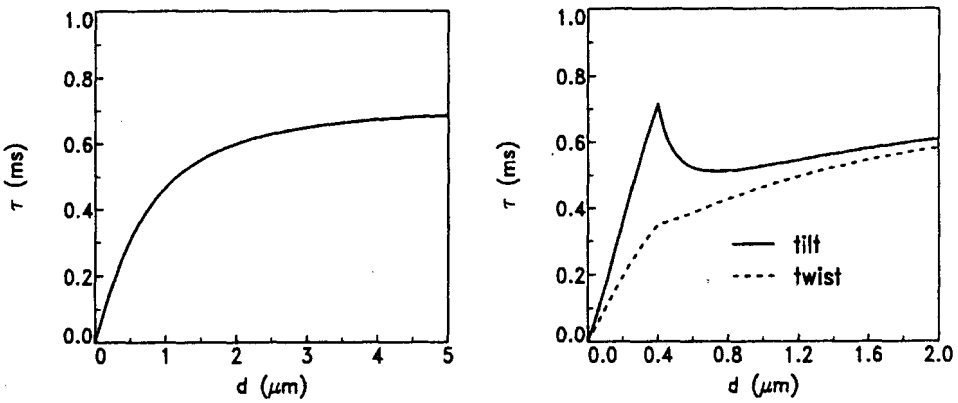

FIGURE 1 The relaxation time of the tilt and the twist mode with smallest q_z of the planarly aligned film (left figure) and the hybridly aligned film (right figure) as a function of thickness according to the model of Stallinga and co-workers[15] (K = 3 pN, γ = 0.015 Pa sec, C_1 = 15 μJ/m^2 and C_2 = 5 μJ/m^2).

CONCLUSION

The scattering of light by thin nematic liquid crystal films differs from the scattering of light by bulk liquid crystals in two important respects. First, the scattering condition, i.e. the relation between the wavevectors of the incoming light, the scattering fluctuation and the scattered light is changed to (2). Second, only a discrete set of fluctuation wavevectors

is allowed due to restrictions imposed by the boundary conditions. As a result, the relaxation times of these discrete fluctuation modes depend on the film thickness. Under the assumption of weak anchoring, the relaxation time varies linearly with film thickness for sufficiently small thicknesses.

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