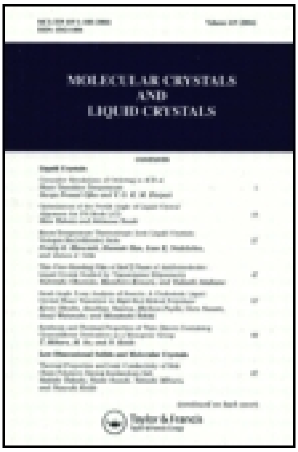

Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl19

Deformed Surface-Induced Smectic A-Structure in Nematic Liquid Crystal

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Version of record first published: 04 Oct 2006

To cite this article: L. V. Mirantsev (1997): Deformed Surface-Induced Smectic A-Structure in Nematic Liquid Crystal, Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals, 301:1, 137-150

To link to this article: http://dx.doi.org/10.1080/10587259708041759

Mol. Cryst. Liq. Cryst., 1997, Vol. 301, pp. 137–150
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DEFORMED SURFACE - INDUCED SMECTIC A - STRUCTURE IN NEMATIC LIQUID CRYSTAL

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Abstract

In the present paper the smectic A - phase induced by the deformed substrate surface in a nematic liquid crystal is considered. The surface microrelief is described by one - dimensional harmonic function. In framework of Landau - de Gennes model the dependences of the surface smectic order parameter and the depth of penetration of the interfacial smectic structure into the nematic bulk on the amplitude and the period of the surface microrelief are obtained. It is shown that the deformation of the substrate surface suppresses the interfacial smectic structure. It is also investigated the effect of the interfacial smectic structure on the depth of penetration of the surface microrelief - induced deformation into the sample bulk. The possibility of experimental verification of the results obtained is discussed.

1. INTRODUCTION

It is known that the interfacial properties of liquid crystals (LC) are significantly different from their bulk properties. For example, the boundary surfaces, such as a solid substrate and a free surface, induce the smectic A (Sm A) - structure in the interfacial region of nematic liquid crystal (NLC). There are a number of experimental [1- 5] and theoretical [6 - 9] papers devoted to the study of the interfacial smectic structures in NLC. However, in these theoretical papers the boundary surface is assumed to be perfectly flat, whereas actually the substrate surface always possesses a certain microrelief. Barbero and Durand have shown [10] that when this microrelief, such as an undulation, is sufficiently sharp, it induces too-large distortional contribution to the nematic free energy that, in turn, gives rise to decrease of the nematic order parameter. It is well known that the elastic constants of the smectic A - phase are much larger than those of the nematic phase [11]. Therefore, one can expect a significant negative effect of the substrate surface microrelief on the interfacial smectic A - structure. On the other hand, in present theoretical papers on the penetration of the surface microrelief - induced deformation into the LC bulk the LC sample is assumed to be in a pure nematic [12] or in pure smectic [11, 13] phase. Consequently, in these papers the interfacial LC region with properties different from those of the bulk phase is ignored.

We propose here a theoretical description of the Sm A -phase induced by the substrate surface with microrelief in NLC near the second order nematic (N) - Sm A phase transition point. For simplicity the microrelief is assumed to be described by the one - dimensional harmonic function with the period of order of one micron. In framework of Landau - de Gennes model the dependences of the surface smectic order parameter and the depth of penetration of the interfacial smectic structure into the nematic bulk on the amplitude and the period of the surface microrelief are obtained. It is also investigated the effect of the interfacial smectic order on the depth of penetration of the surface microrelief - induced deformation into the sample bulk. In conclusion the possibility of experimental verification of the results obtained is briefly discussed.

2. BASIC EQUATIONS OF MODEL

Let us consider the nematic layer in contact with the solid substrate surface having a microrelief described by one - dimensional harmonic function, for example

$$U_0(x) = U_0 \cos((2\pi/d)x), \tag{1}$$

where U_0 and d are the amplitude and period of the surface microrelief, respectively. The NLC layer is assumed to be homeotropically aligned (in the sample bulk the director $\vec{n}$ is parallel to z - axis normal to the substrate surface). Let us also assume that our LC sample is in a vicinity of the second order N - Sm A phase transition (the NLC temperature is slightly above the transition point) and the orientational order in it is perfect (the orientational order parameter $S = 1$, i.e. the long axes of all molecules are parallel to the director $\vec{n}$). The latter assumption is reasonable enough because the most of LCs undergo the second order N - Sm A phase transition sufficiently far from the clearing point and the actual orientational order in them is very close to the perfect one.

Let us assume that due to the interaction between mesogenic molecules and the substrate surface the latter induces the positionally ordered Sm A - structure with the period equal to the molecular length l. If the molecules within the first interfacial smectic layer are assumed to be hardly stacked on the substrate surface then this layer should be distorted in tact with the substrate sinusoidal relief and this distortion due to the small smectic layer compressibility can be transmitted to the neighbouring layers. It is clear that the deformation of the interfacial smectic structure should effect on it's translational order, i.e. on the value of the smectic order parameter. On the other hand, the depth of penetration of the surface microrelief - induced deformation depends on the smectic layer compressibility which is determined in turn by the translational order of the interfacial smectic structure. Thus, to describe completely the LC region near the substrate surface with microrelief we must determine simultaneously the interfacial smectic order parameter profile and the penetration of the surface microrelief - induced deformation into the sample bulk.

In order to solve this problem we must know the expression for the free energy density in the interfacial LC layer. This expression should contain two contributions. The first contribution is the free energy density of the deformed nematic liquid crystal [14]

$$f_N = (K_1/2)(div\vec{n})^2 + (K_2/2)(\vec{n} \cdot rot\vec{n})^2 + (K_3/2)(\vec{n} \times rot\vec{n})^2, \tag{2}$$

where $K_{1,2,3}$ are the Frank splay, twist and bend elastic constants, respectively. If the substrate surface wavy deformation is assumed to be weak enough $((2\pi/d)U_0 \ll 1)$, then the director components are related to the one - dimensional wavy deformation $U(x,z)$ by the equations

$$n_x \approx -\partial U/\partial x, n_y = 0, n_z \approx 1 \tag{3}$$

Substituting these relations into equation (2) one can obtain

$$f_N \approx (K_1/2)(\partial^2 U/\partial x^2)^2 + (K_3/2)(\partial^2 U/\partial x \partial z)^2, \tag{4}$$

It should be noticed that since the LC under consideration is in the vicinity of the second order N - Sm A phase transition, the bend elastic constant K_3 in equation (4) is not similar to that in "pure" nematic phase. Near the second order N - Sm A transition point the smectic short - order fluctuations occur in the nematic bulk phase. These smectic order fluctuations are not induced by the substrate surface and must be considered separately from the surface induced smectic structure. As it will be seen below, they give rise to a renormalization of the elastic constant K_3.

The second contribution is the free energy density of the surface - induced Sm A - phase. If $\sigma(z)$ is the smectic order parameter and $U(x,z)$ is the shift of smectic layers due to the substrate surface microrelief then the surface - induced smectic A - phase in the vicinity of the second order N - Sm A phase transition is described by the density wave

$$\rho(x,z) = \rho_0[1 + \sigma(z)\cos(2\pi(z - U(x,z))/l)], \tag{5}$$

where ρ_0 is the average density of the liquid crystal molecules, and the free energy density of the Sm A - phase is given by the following Landau - de Gennes expression:

$$f_{SmA} = (A/2)\sigma^2 + (C/4)\sigma^4 + (L/2)(d\sigma/dz)^2 + (B_0/2)(\partial U/\partial z)^2, \tag{6}$$

where $A = \alpha(T - T_0)$, α and C are the temperature independent constants, T is the temperature of the system, T_0 is the N - Sm A phase transition temperature, L and B_0 are the elastic constants related to each other by the equation [15]

$$B_0 = L(2\pi/l)^2. \tag{7}$$

It should be noted that two gradient terms in equation (6) are due to the coordinate dependence of the absolute value of the smectic order parameter σ and the smectic layer elastic deformation, respectively. Adding the expressions (4) and (6) one can obtain the following equation for the total free energy density of the LC layer near the deformed substrate surface:

$$f = f_N + f_{SmA} = (K_1/2)(\partial^2 U/\partial x^2)^2 + (K_3/2)(\partial^2 U/\partial x \partial z)^2 +$$

$$(A/2)\sigma^2 + (C/4)\sigma^4 + (L/2)(d\sigma/dz)^2 + (B_0/2)(\partial U/\partial z)^2. \quad (8)$$

In order to determine the total free energy of the interfacial region we must integrate the free energy density (8) over the space above the substrate surface and add to the result obtained the energy of direct interaction between the liquid crystal molecules and the substrate. In previous papers on the surface - induced smectic A - phase [7-9] this interaction was simulated by short - range orienting field which acts directly only on the molecules contacting with the boundary surface. The energy of such interaction can be written as

$$G(z, \vartheta) = -G_0(3/2\cos^2\vartheta - 1/2)\delta(z - U_0(x)),$$

where ϑ is the angle between long axes of the LC molecules and the normal to the boundary surface, G_0 is the interaction constant, and $\delta(z - U_0(x))$ is a well known Dirac function. Since the orientational order in the system under consideration is assumed to be perfect ($\cos\vartheta \rightarrow 1$) this potential can be represented as

$$G(z) = -G_0\delta(z - U_0(x)), \quad (9)$$

and the energy of interaction per unit substrate square is equal to

$$F_s = \int_0^\infty G(z)\rho(x, z)\, dz = -G_0\rho_0 - G_0\rho_0\sigma_0, \quad (10)$$

where σ_0 is the value of the smectic order parameter at the substrate surface and G_0 is the interaction constant. Finally, the total free energy of the LC interfacial layer per unit substrate square can be represented as

$$F = \int_0^\infty \bar{f}(z)\, dz - G_0\rho_0 - G_0\rho_0\sigma_0, \quad (11)$$

where $\bar{f}(z)$ is the free energy density of the LC interfacial layer averaged over the XY - plane. If we search the expression for $U(x, z)$ in the form

$$U(x, z) = U(z)\cos((2\pi/d)x),$$

$$U(x, z)\Big|_{z=0} = U_0\cos((2\pi/d)x), \quad (12)$$

then the expression for $\bar{f}(z)$ is the following:

$$\bar{f} = (A/2)\sigma^2 + (C/4)\sigma^4 + (L/2)(d\sigma/dz)^2 + (B_0/4)\sigma^2(dU/dz)^2 + (K_1/4)(2\pi/d)^4 U^2$$

$$+(K_3/4)(2\pi/d)^2(dU/dz)^2. \quad (13)$$

Substituting equation (13) into the expression (11) for the total free energy of the interfacial layer and minimizing latter with respect to $\sigma(z)$ and $U(z)$, we obtain the following Euler - Lagrange equations:

$$L(d^2\sigma/dz^2) - A\sigma - C\sigma^3 - (B_0/2)\sigma(dU/dz)^2 = 0, \tag{14}$$

$$B_0\sigma(d\sigma/dz)(dU/dz)+(B_0/2)\sigma^2(d^2U/dz^2)+(K_3/2)(2\pi/d)^2(d^2U/dz^2)-(K_1/2)(2\pi/d)^4U = 0. \tag{15}$$

These equations can be solved only numerically. However, let us attempt to obtain the approximative analytical solution. We can use the following approach. Even for perfectly flat substrate the surface - induced smectic order should decay with penetration into the nematic bulk at the distance of order of the longitudinal correlation length $\xi = (L/A)^{1/2}$ for smectic fluctuations [6]. According to experimental data [16], at the temperature about 0.1 K higher than that of the second order N - Sm A phase transition this correlation length is of order of 0.1 μm. On the other hand, if the period of the substrate surface wavy relief d is of order of 1 μm, then the depth of penetration of the surface - induced deformation into the nematic bulk should be of the same order. Therefore it is reasonable to assume that $\sigma(z)$ must decay rapidly in comparison with $U(z)$, or $\sigma(z)$ is a rapidly varying function and $U(z)$ is a slowly varying one. Then in equation (14) one can set $dU/dz \approx dU/dz\big|_{z=0}$ In this case we have no difficulty in integration of equation (14) and obtain

$$\sigma(z) = (a/C^*)^{1/2}\left[\left(\frac{q+e^{-2\sqrt{a}z}}{q-e^{-2\sqrt{a}z}}\right)^2 - 1\right]^{1/2} \tag{16}$$

where

$$a = 1/\xi^2 + (2\pi^2/l^2)(dU/dz)^2\big|_{z=0}, \tag{17}$$

$$q = \frac{\sqrt{a + C^*\sigma_0^2} + \sqrt{a}}{\sqrt{a + C^*\sigma_0^2} - \sqrt{a}}, \tag{18}$$

$$C^* = C/2L. \tag{19}$$

It should be noted that the solution (16) has been obtained under condition of complete decay of the interfacial smectic structure with penetration into the bulk nematic phase ($\sigma \to 0$, $d\sigma/dz \to 0$ at $z \to \infty$). In order to calculate the value σ_0 we can use the following relation:

$$\frac{\partial f}{\partial(\partial\sigma/\partial z)}\Big|_{z=0} = -G_0\rho_0. \tag{20}$$

Taking into account that

$$\frac{\partial \bar{f}}{\partial(\partial\sigma/\partial z)}\Big|_{z=0} = L\frac{d\sigma}{dz}\Big|_{z=0},$$

and

$$\frac{d\sigma}{dz}\Big|_{z=0} = -\sqrt{a\sigma_0^2 + C^*\sigma_0^4}, \tag{21}$$

one can obtain the expression for σ_0^2

$$\sigma_0^2 = -(a/2C^*) + [(a/2C^*)^2 + (G_0^2\rho_0^2/L^2C^*)]^{1/2}. \tag{22}$$

Now let us consider equation (15). Multiplying it by dU/dz and assuming again that $\sigma(z)$ is a rapidly decaying function in comparison with $U(z)$, we can obtain the following approximative equation:

$$\frac{d}{dz}\left(B_0\sigma^2(dU/dz)^2\right) + \frac{d}{dz}\left(K_3(2\pi/d)^2(dU/dz)^2\right) - \frac{d}{dz}\left(K_1(2\pi/d)^4U^2\right) = 0. \tag{23}$$

If the interfacial smectic structure and the substrate surface - induced LC deformation are assumed to decay completely with penetration into the nematic bulk ($\sigma(z) \to 0$, $U(z) \to 0$, $dU/dz \to 0$ at $z \to \infty$), then the integration of equation (23) gives

$$(dU/dz) = -\frac{\sqrt{K_1}(2\pi/d)^2U(z)}{\sqrt{B_0\sigma^2(z) + K_3(2\pi/d)^2}}. \tag{24}$$

Setting $z = 0$ and using the relation [11]

$$B_0/K_0 \approx 1/l^2,$$

we obtain

$$(2\pi^2/l^2)(dU/dz)^2|_{z=0} = \frac{8\pi^4(U_0/d)^2}{[\sigma_0^2(d/2\pi)^2 + l^2(K_3/K_1)]}. \tag{25}$$

Substitution of this relation into equations (17) and (22) allows us to determine numerically the interfacial smectic order parameter σ_0 at given values of the parameters ξ, U_0, $(G_0\rho_0/LC^*)$ and K_3/K_1. The ratio of the elastic constants K_3/K_1 in the vicinity of the second order N - Sm A phase transition can be obtained by using de Gennes's equation [17]

$$K_3 = K_3^0 + (K_BT/6)(\pi\xi/l^2), \tag{26}$$

where K_3^0 is the bend elastic constant in "pure" nematic (without smectic fluctuations) and K_B is the Bolzmann constant. If for simplicity we set $K_3^0 \approx K_1$, then

$$K_3/K_1 \approx 1 + (K_BT/6)(\pi\xi/l^2K_1). \tag{27}$$

Finally, we can determine z - dependence of the substrate surface - induced LC deformation. Substituting the expression (16) for $\sigma(z)$ into equation (24), we obtain after integration

$$U(z) = U_0 \exp[-(I_1 + I_2)], \tag{28}$$

where

$$I_1 = \frac{\pi}{d\sqrt{a(K_3/K_1)}} \ln \frac{2q(K_3/K_1)^{1/2} SQ_1 + 2q^2(K_3/K_1)e^{2\sqrt{a}z} + D}{2q(K_3/K_1)^{1/2} SQ_3 + 2q^2(K_3/K_1) + D}, \tag{29}$$

$$I_2 = \frac{\pi}{d\sqrt{a(K_3/K_1)}} \ln \frac{2(K_3/K_1)^{1/2} SQ_2 + 2(K_3/K_1)e^{-2\sqrt{a}z} + D}{2(K_3/K_1)^{1/2} SQ_3 + 2(K_3/K_1) + D}, \tag{30}$$

$$SQ_1 = [q^2(K_3/K_1)e^{4\sqrt{a}z} + De^{2\sqrt{a}z} + K_3/K_1]^{1/2}, \tag{31}$$

$$SQ_2 = [(K_3/K_1)e^{-4\sqrt{a}z} + De^{-2\sqrt{a}z} + q^2(K_3/K_1)]^{1/2}, \tag{32}$$

$$SQ_3 = [(K_3/K_1) + D + q^2(K_3/K_1)]^{1/2}, \tag{33}$$

$$D = 2q[2(a/C^*)(d/2\pi l)^2 - (K_3/K_1)]. \tag{34}$$

3. RESULTS OF NUMERICAL CALCULATION AND DISCUSSION

The relations obtained in previous section of the present paper allow us to determine both the deformed surface - induced smectic order parameter profile and the dependence of the amplitude of the liquid crystal wavy deformation $U(z)$ on the distance from the substrate. The smectic order parameter profiles obtained for various values of the amplitude U_0 of the substrate surface microrelief (the period d of the wavy microrelief is fixed) are shown in Figure 1. These profiles have been obtained at the following values of the parameters involved:

$(T - T_0)/T_0 = 10^{-4}$; $T_0 \approx 300$ K; $\xi = 0.16\mu$m; $K_3^0 \approx K_1 = 10^{-6}$ dyn; $l = 3 \cdot 10^{-7}$ cm (data for 8 CB from ref. [16]); $C/A \approx 30$ [18]; $g = G_0\rho_0/LC^* = 0.1$.

The value of last parameter g is sufficiently arbitrary because we have no detail information on the interaction between LC molecules and the substrate surface. It should be noted, however, that the value of the surface smectic order parameter $\sigma_0 = 0.559$ obtained at this value of g in case of the perfectly flat surface is reasonable enough. It is clearly seen that the substrate surface deformation gives rise to suppression of the interfacial smectic A - structure. Both, the magnitude of the smectic order parameter and the depth of penetration of the smectic order into the nematic bulk decrease with increasing microrelief amplitude U_0. In other words, as sharper is the substrate surface microrelief, the stronger is the deformation of the interfacial smectic layers which ,in turn, gives rise to the suppression of the interfacial smectic A - structure.

Now let us discuss the effect of the interfacial smectic A - structure on the penetration of the surface microrelief - induced deformation into the sample bulk. The dependence of amplitude of the wavy deformation on the distance from the substrate is shown in Figure 2. The curve 1 corresponds to the pure nematic sample ($g = 0$, i.e. the interfacial smectic A - structure is absent) and the curve 2 corresponds to existence of the interfacial smectic A - phase ($g = 0.2$, $\sigma_0 = 0.62$). It is seen that occurrence of the smectic A - structure near the substrate surface promotes to the penetration of the surface - induced deformation into the nematic bulk. This result is obviously demonstrated by Figure 3 according to which the depth l^* of penetration of the surface - induced deformation into the nematic bulk (l^* is the distance from the substrate at which the amplitude of deformation $U(z)$ is e -times smaller than that at the substrate surface) is proportional to the surface smectic order parameter σ_0.

We have also investigated the dependence of the depth of penetration of the substrate surface - induced deformation into the sample bulk on both amplitude U_0 and period d of the substrate surface microrelief. The dependence of the depth of penetration l^* of the surface - induced deformation into the sample bulk on the amplitude U_0 is shown in Figure 4. When the interfacial smectic A - structure is absent the depth l^* is independent of the amplitude U_0 (curve 1 for pure nematic phase). It should be noted that this result is also valid for the penetration of the surface - induced deformation into a pure smectic A sample [11,13]. However, the existence of the substrate surface - induced smectic A - phase gives rise to a significant dependence of the depth of penetration l^* on the amplitude U_0 (curve 2). It is seen that for very small amplitude of the substrate surface microrelief the depth of penetration of the surface - induced deformation into the sample bulk is about 1.6 - 1.7 times larger than that in a pure nematic phase. As the amplitude of the substrate surface microrelief is increased the depth of penetration l^* decreases because of the suppression of the interfacial smectic structure and for $U_0 \geq 0.02\mu m$ this depth coincides with that in pure NLC.

Finally, let us discuss briefly the possibility of an experimental verification of the results obtained. The experimental study of the Freedericksz transition in thin nematic cells performed by Rosenblatt [4] revealed the anomalous growth of the Freedericksz critical field in the vicinity of the second order N - Sm A phase transition. This growth was attributed to the effect of the surface - induced smectic A - structure. The point is that the penetration of such a structure into the sample bulk gives rise to decrease of the effective thickness of the nematic layer sensitive to the external magnetic field that results, in turn, in growth of the Freedericksz critical field. One can measure the Freedericksz critical field in a vicinity of the second order N - Sm A transition for two thin nematic cells of the same thickness. If the first cell has the substrates with perfectly flat surfaces and in second cell the substrate surfaces possess a sinusoidal microrelief with period d and amplitude U_0 sufficient for suppression of the surface - induced smectic structure, then the Freedericksz critical field for first cell should be larger than that for second one. Numerical estimation based on the Rosenblatt's experimental data for 8 CB [4] and our theoretical results shows that at the temperature about 0.2 K higher than the second order N - Sm A transition point and cell thickness of order of $\sim 2.5\mu m$ the Freedericksz critical field for the cell with perfectly flat substrates can be about 10 % higher than that for cell with substrates having the microrelief with the amplitude $U_0 \approx 0.02\mu m$ and the period $d \approx 0.5\mu m$. We suppose that such experimental verification could be rather interesting.

4.CONCLUSION

In the present paper the theoretical description of the Sm A -phase induced by the substrate surface with microrelief in NLC near the second order N - Sm A phase transition point is proposed. For simplicity the microrelief is assumed to be described by the one - dimensional harmonic function. In framework of Landau - de Gennes model the dependence of the surface smectic order parameter and the depth of penetration of the interfacial smectic structure into the nematic bulk on amplitude and period of the surface microrelief are obtained. It is shown that the substrate surface deformation gives rise to the suppression of the interfacial smectic A - phase. Both, the magnitude of the smectic order parameter and the depth of penetration of the smectic order into the nematic bulk decrease with increasing microrelief amplitude U_0. When the substrate surface microrelief is sufficiently sharp, the interfacial smectic structure is completely suppressed.

The effect of the interfacial smectic order on the depth of penetration of the surface microrelief - induced deformation into the sample bulk is also investigated. It is shown that occurrence of the smectic A - structure near the substrate surface promotes to the penetration of the surface - induced deformation into the nematic bulk. The dependence of the depth of penetration on amplitude and period of the substrate surface microrelief is revealed. It is found that the presence of the surface - induced smectic A order gives rise to dependence of this depth on the amplitude of the substrate surface microrelief, which is absent in case of pure nematic phase [12].

Finally, it is proposed to use the experimental study of the Freedericksz transition in thin nematic cells for verification of the results obtained.

AKNOWLEDGMENTS

Author is grateful to Prof. E. L. Aero for very useful discussions.

This work has been supported by Russian Fund of Fundamental Investigations (Project 96 - 03 - 32417 - a).

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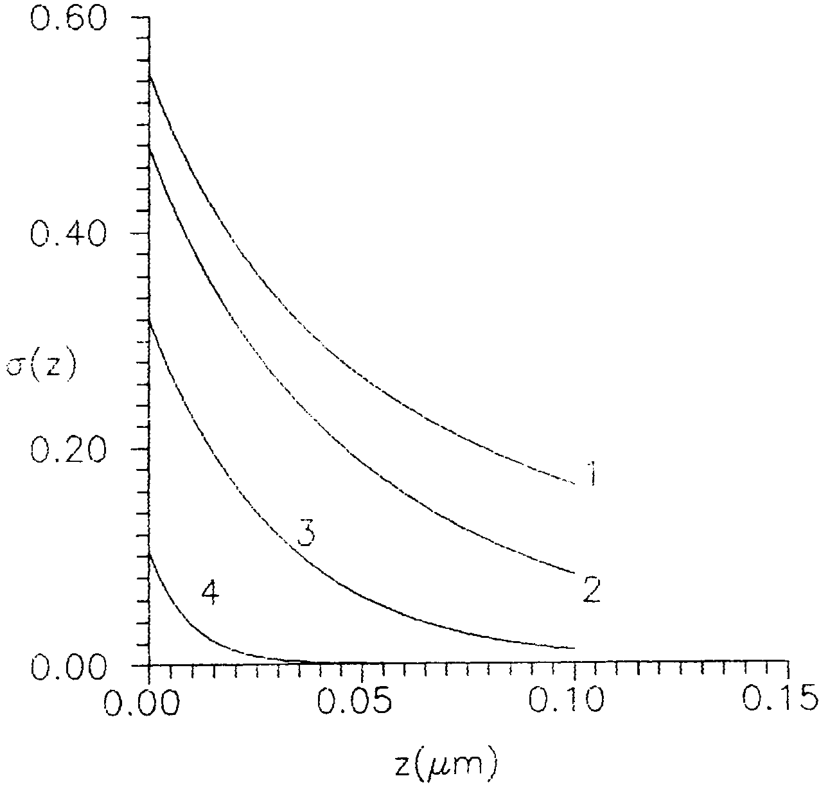

Figure 1. The interfacial smectic order parameter profiles for various amplitudes of the substrate surface microrelief. $d = 0.5\mu m$. 1 - $U_0 = 0$; 2 - $U_0 = 0.01\mu m$; 3 - $U_0 = 0.015\mu m$; 4 - $U_0 = 0.02\mu m$.

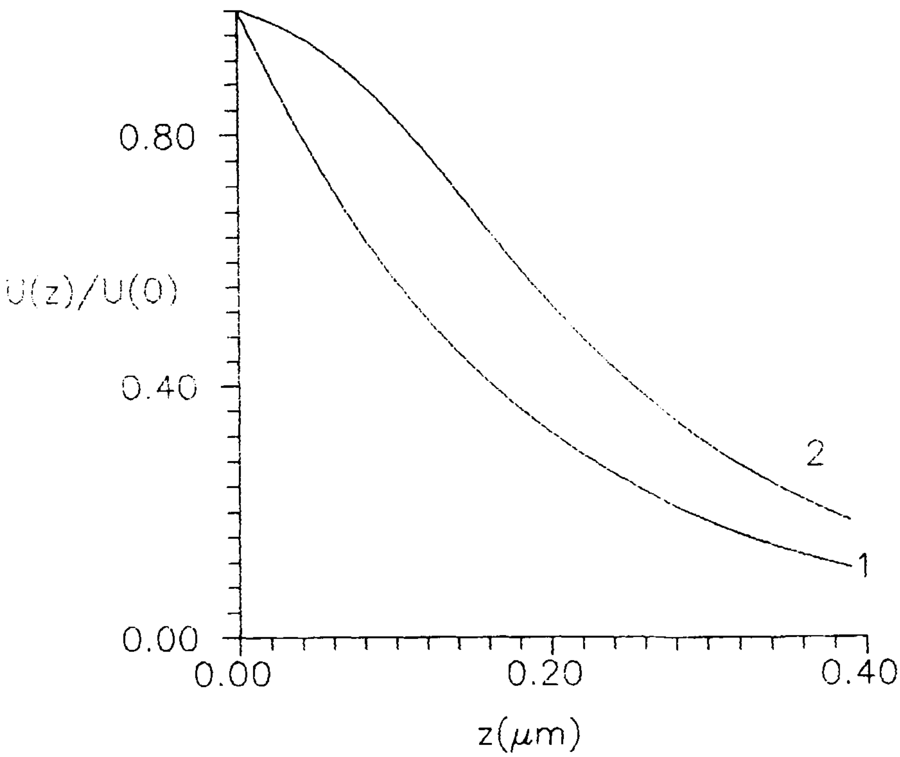

Figure 2. The dependence of amplitude of the wavy LC deformation on the distance from the substrate surface. $U_0 = 0.01\mu m$, $d = 0.5\mu m$. 1 - the interfacial Sm A - structure is absent ($\sigma_0 = 0$); 2 - the interfacial Sm A - phase exists ($\sigma_0 = 0.62$).

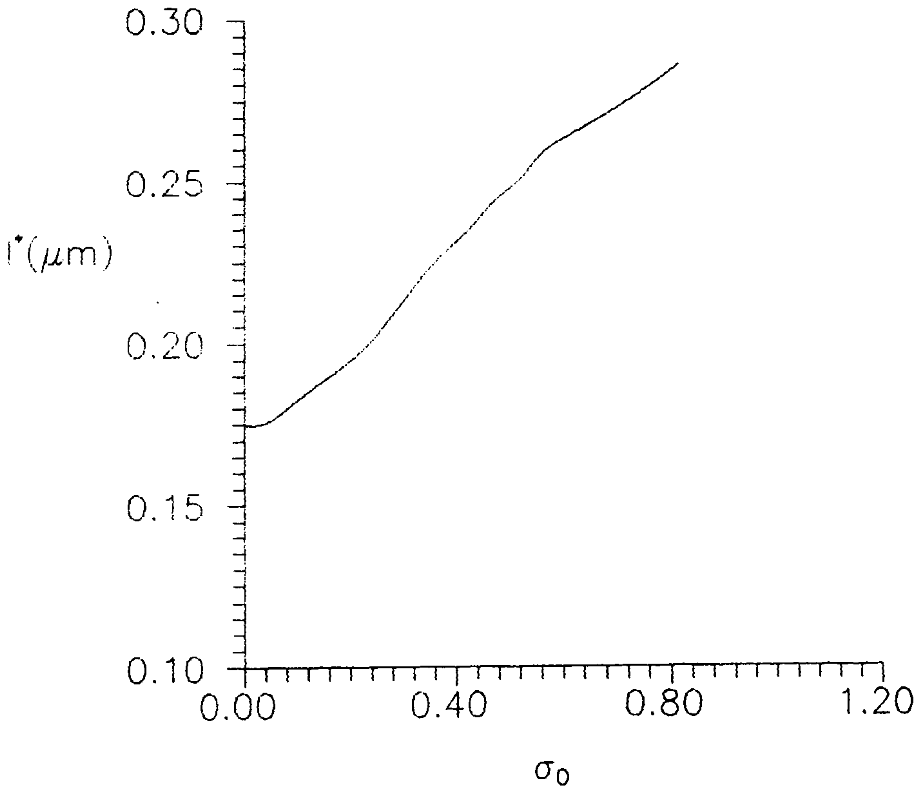

Figure 3. The dependence of the depth of penetration of the substrate surface microrelief - induced deformation into LC bulk on the interfacial smectic order parameter. $U_0 = 0.01\mu m$, $d = 0.5\mu m$.

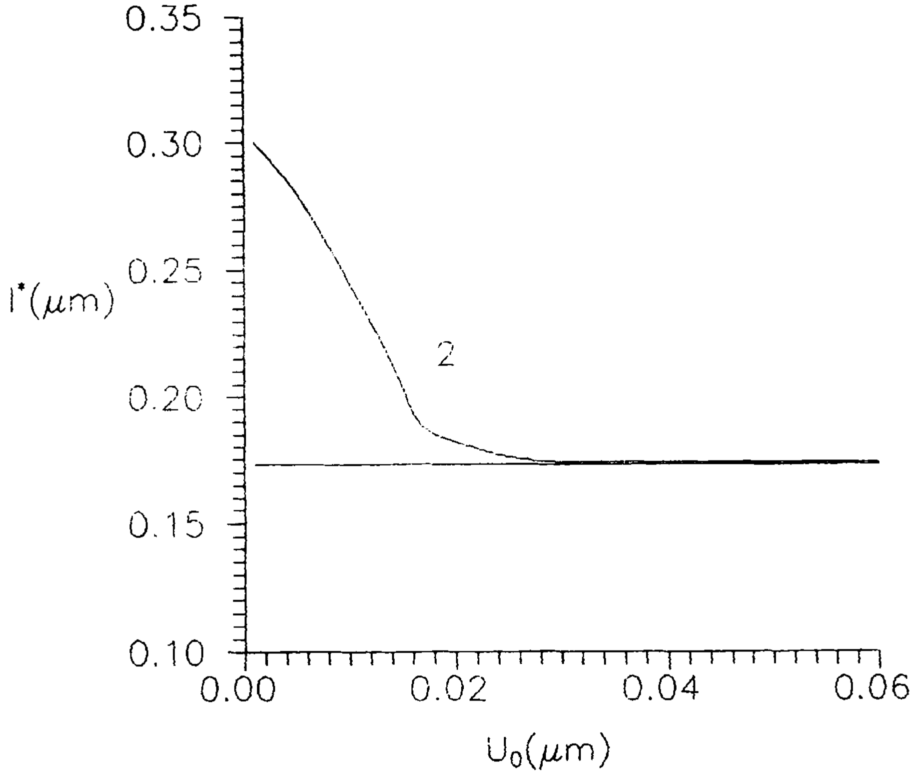

Figure 4. The dependence of the depth of penetration of the substrate surface microrelief - induced deformation into LC bulk on the amplitude of the microrelief. $d = 0.5\mu m$. 1 - the interfacial Sm A - phase is absent ($g = 0$); 2 - the interfacial Sm A - phase exists ($g = 0.1$).