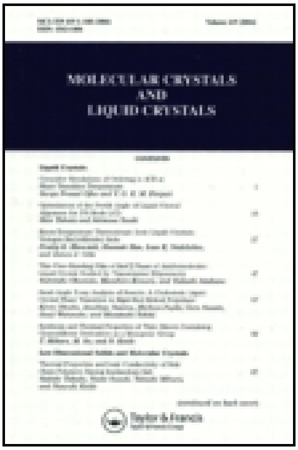


Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl20

# Anisotropic Spreading of Liquid Crystals and Isotropic Fluids on Anisometric Surface of DVD Discs

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Version of record first published: 30 Jan 2009

*Mol. Cryst. Liq. Cryst.*, Vol. 495, pp. 212/[564]–220/[572], 2008

ISSN: 1542-1406 print/1563-5287 online
DOI: 10.1080/15421400802432667

# Anisotropic Spreading of Liquid Crystals and Isotropic Fluids on Anisometric Surface of DVD Discs

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*The first experimental data on spreading of liquid crystal droplets over the surface with a regular submicron relief are presented. It is established that such relief produces a motion of a precursor film in the direction of grooves. The dynamics of motion of a precursor film and a contact line is investigated via polarizing microscopy. The possible influence of anisotropic near-surface viscosity on spreading of liquid crystal droplets is discussed.*

**Keywords:** anisometric surface; nematic phase; spreading



## 1. INTRODUCTION

The behaviour of strongly confined liquid crystals is of interest for both fundamental science and practical applications. A number of experiments, fulfilled in irregular 3D nanostructures like porous glasses, aerogels, aerosils have shown that strong confinement leads to essential modification of thermodynamic properties of liquid crystals [1], for example, to the temperature shifts of transition points. It was shown that not only equilibrium characteristics but the non-equilibrium ones like dielectric relaxation times, diffusion coefficients, shear viscosity coefficients can be modified by strong irregular and

This research was partially supported by grants: RFBR 07-02-08394-z, 07-01-13523-ofi; Grant of President of RF for young scientists MK-704.2007.2.

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regular confinement [2–6]. In the latter case it takes place due to essential slowing down of both the translational molecular motions and the orientational ones in near-surface layers of liquid crystals [5]. Such effects existing in isotropic liquids [2,3,6–8] are usually attributed to extremely thin (about some nanometers) surface layers whereas in liquid crystals the near-surface layers with modified non equilibrium properties can be essentially thicker, about 100 nm [4], for example. So, surface layers can provide the important contribution in the total response of relatively thin layers of LC (1, ..., 10 μm) on the action of shear flows [9] and electric fields which is used in the modern display industry.

It is well known that surface phenomena play a key role at spreading of isotropic droplets over solid surfaces of different types [10]. Contrary to the case of isotropic liquids only rare experiments on spreading of liquid crystal droplets [11,12] were carried out up to now. The strict theoretical description of such phenomena is under elaboration [13] even for the case of nematic droplets.

In this article we concentrate our attention on liquid crystal droplets interacting with a strongly anisotropic surface induced by a regular relief. The usual polarizing microscopy and processing of digital images were used to obtain the results described below. We hope that the new experimental data presented in our paper will stimulate a theoretical description of complicated irreversible processes existing at spreading of anisotropic liquids over anisotropic surfaces.

## 2. EXPERIMENT

In our experiments we used the substrates with a well defined periodical surface shown in Figure 1. It was formed by a special punch in a photopolymer layer which covered a polycarbonate plate of thickness about 1 mm. We also used the samples with a smooth surface made of the same materials with a thickness of a photopolymer layer equal to 0.2 μm.

A number of liquid crystals materials: 5CB, nematic mixtures ZhK 440, ZhK 616, ZhK1289 (NIOPiK production) with different material parameters [14] were studied to establish the general features of spreading of anisotropic liquids. Some additional experiments were performed with different isotropic liquids like water and glycerol too.

In experiments a small droplet of a liquid crystal (or isotropic liquid) was placed on the relief or smooth substrate. The process of spreading was registered by taking snapshots via a polarizing microscope and a digital camera. Observations were performed between parallel or crossed polarizers (at 45 degrees respectively to the direction of a

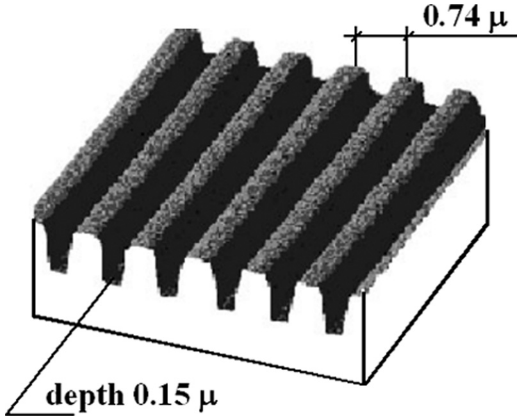


**FIGURE 1** The substrate with a periodical surface relief.

relief). A sequence of digital photos was processed to extract information about dynamics of spreading.

## 3. RESULTS AND DISCUSSION

### Spreading of Liquid Droplets Over an Isotropic Surface

Isotropic and LC droplets have shown analogous behavior on pure isotropic surfaces. A shape of droplets was close to a segment of a sphere. Under observation in polarized light isotropic droplets did not show interference rings, while the latter were visible in LC droplets. The similar interference rings were previously described for the droplets of 5 CB on a smooth surface of silicon oxide wafers [12]. Contrary to the case of Newtonian rings they can be attributed to the changes of birefringence of LC. We also observed some deformation of a shape of a contact line, which can be explained by a hydrodynamic instability of a liquid crystal [12].

### Spreading of a Liquid Crystal Droplet Over Anisotropic Surface

Liquid crystal and isotropic droplets were quickly stretched in the direction of grooves after set down (Fig. 2). Such behaviour can be referred to the better wetting of a surface in the direction of a relief. Afterwards in the case of a pure surface the flow inside of separate grooves was observed. The continuous thin film covered all grooves on the next stage (Fig. 3).

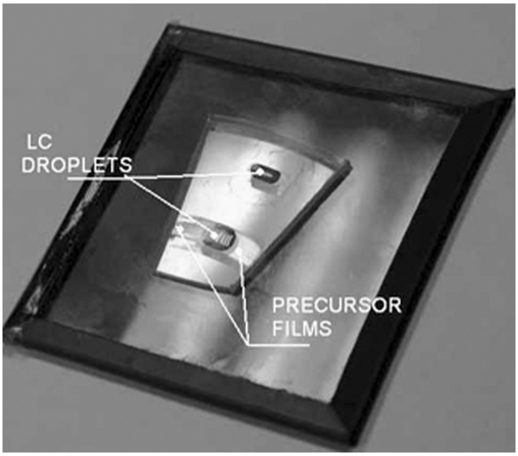


**FIGURE 2** The photo of spreading of two droplets of ZhK616 at natural illumination. A precursor film is well visible on the low (clean) part of the substrate and is practically invisible on the upper (previously treated) part.

The character and the rate of LC spreading were essentially dependent on the previous treatment of the surface. In particular, usage of homeotropic surfactant reduced spreading: a droplet was stretched and spreading was stopped rather quickly. At the same time on pure surfaces a spreading was limited only by size of substrates in the direction of spreading and was observed for one hour. As a result the large area of the substrate was covered by a thin film perfectly visible on the

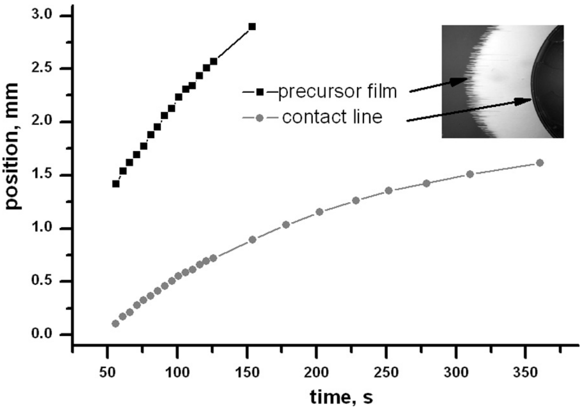


**FIGURE 3** Position of a precursor film (squares) and of a contact line (circles) for different moments of time and a microphotography of a LC droplet between crossed polarizers.

relief surface at natural illumination (Figs. 2, 3). At the same time the overall shape of the stretched droplet was not changed essentially for a long period, which is quite different in the comparison with spreading of isotropic liquids over isotropic surfaces at good wetting [10].

The spreading process of LC droplet is well visualised between crossed polarizers due to optical anisotropy of liquid crystal. In this case the image of the thin precursor film includes the sequence of non-circular interference stripes (Fig. 4). It is obvious that in both cases of isotropic and anisotropic spreading of LC the interference stripes correspond to the same phase difference Φ between the extraordinary (e) ray and the ordinary (o) one. One can restore the approximate profile of the surface with reasonable assumptions about orientational structure inside the film [12].

We made some experiments, which showed that such type of a relief provided a surface orientation of LC in the direction of grooves. In particular, cells made of two substrates with periodical relief had shown planar or twist orientation at case of parallel or perpendicular disposition of grooves respectively.

The analogous structures were obtained in combined cells when one of the relief substrates was replaced by a glass plate treated by rubbing for planar surface anchoring. So we conclude that the strongly anisotropic relief used in our experiments really provides a planar surface orientation.

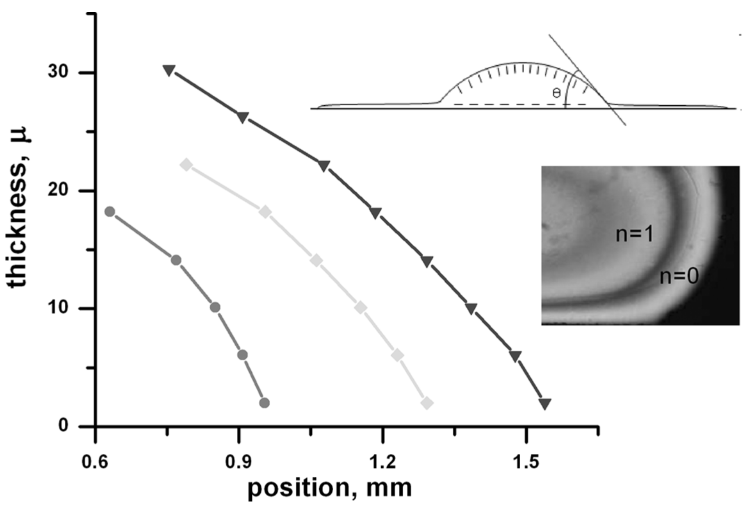


**FIGURE 4** The profile of a LC droplet in the direction of spreading for different moments of time (circles – 178 s, $\theta = 5.0°$, squares – 252 s, $\theta = 3.4°$, triangles – 360 s, $\theta = 2.2°$) and a microphotography used for calculation.

The orientation on the second boundary (LC-air) is unknown for most LC materials, so we will adopt a normal orientation, as in the case of 5 CB. In this case the angle $\beta$ between the local director and the normal to the plate is expressed in one constant approximation as a linear function of a coordinate $z$:

$$\beta(\mathrm{z}) = (\pi/2)(1 - \mathrm{z}/d) \tag{1}$$

One can extract the local thickness $d$ of the film as a function of coordinate $x$ by analyzing a light intensity $I$ distribution in interference stripes which is described by a well known expression:

$$I = I_0 \sin^2 \frac{\Phi}{2} \tag{2}$$

where the phase difference $\Phi$ is expressed as:

$$\Phi = \frac{2\pi}{\lambda} \int_0^d \Delta n dz \tag{3}$$

$$\Delta n = n(\beta) - n_0 \tag{4}$$

$$n(\beta) = n_0 \left[ 1 - \frac{n_e^2 - n_0^2}{n_e^2} \cos^2 \beta \right]^{-\frac{1}{2}} \tag{5}$$

($n_\mathrm{e}$ and $n_0$ – are refractive indexes for the extraordinary ray and the ordinary one, $\lambda$ – is the light wavelength).

Using Eq. (1–5) one can calculate the local thickness $d$ via numerical methods.

It is possible to obtain simple analytical expressions for low values of optical anisotropy:

$$\Delta n = n_e - n_o \ll n_e \tag{6}$$

In this case the phase difference is approximately twice lower than corresponding value for total reorientation of LC from the planar state to the homeotropic one and can be written as:

$$\Phi = (\pi d/\lambda) \left[ n_0 \left( n_e^2 - n_0^2 \right) / n_e^2 \right] \tag{7}$$

It provides the simple calculations of the local thickness corresponding to the maximal (minimal) intensity of interference stripes:

$$d_k = [\lambda/(2\,\Delta n)](2k + 1) \quad \text{for maximal intensity} \tag{8}$$

$$d_k = [\lambda k/\Delta n] \quad \text{for maximal intensity} \tag{9}$$

where $k$-order of interference stripes.

The examples of profiles *d(x)* obtained by usage of Eq. (8) for different moments of time are shown in Figure 4. In principle extrapolation of these curves up to intersection with the $x$ axes provides the calculation of both the contact angle and the position of the contact line as functions of time. Such information seems to be important for an experimental checking of different theoretical models applied to the spreading phenomena [10].

The most drastic difference between our experiments and those mentioned above [12] is connected with a precursor film moving ahead of a contact line. It's overall distance from the contact line (Fig. 3) can exceed some millimeters contrary to the case of LC droplet on the isotropic surface where such film is located at a short distance (about 50 μm) near the contact line.

The results presented above show that overall picture of anisotropic spreading of LC droplets is rather complicated and will demand a serious theoretical work for an adequate description which is beyond this paper. Nevertheless it would be useful to discuss the perspectives of such work.

Firstly, it would be interesting to consider the problem of a flow in individual grooves as in open channels. In general, the thickness of LC film $d_0$ in the centre of such channel can differ from the depth $h$ of a groove due to menisci. At good wetting as in our case $d_0 < h$, so in the first approximation it seems to be reasonable to consider such motion as the Poiseulle flow at nanoscaled confinement. It is questioned whether the flow dynamics in this case is mostly determined by the process of a structural relaxation characteristic for a nanometer precursor film in isotropic liquids or by a balance of usual viscous losses and surface tension [12]. If viscous losses play the essential role one can hope that the computer simulation of such flow will provide the information about anisotropic shear viscosities in nanometer near-surface layers. Such viscosities may contribute to the motion of the contact line too. The tentative analysis of this problem for more simple case of isotropic spreading of LC droplet [12] over smooth surface shows that both mechanisms of the energy dissipation mentioned above can be of importance. The latter is reflected in the balance equation for forces acted on the contact line:

$$(\eta/\theta + A)U = \gamma\theta^2 + B \tag{10}$$

where $\eta$ – the shear viscosity coefficient, $\theta$ – the contact angle, $\gamma$ – the coefficient of a surface tension. The parameter $A$ is referred to an additional term independent on the contact angle and arising due to structural relaxation in a precursor film. At the same time the parameter $B$ describes a contribution to the driven force existing due to not fully developed precursor film. In the case $A = B = 0$ the expression (10) is reduced to the well known Tanner's law [10] for well wetting isotropic liquids. It seems to be interesting to get the expression analogous to (10) for the case of anisotropic spreading of LC droplets realized in our experiments to estimate the viscous losses and shear viscosity coefficient in precursor film of submicron thickness.

So the further experimental studies and theoretical consideration of anisotropic spreading mentioned above may be of importance for an elaboration of the general dynamic theory of liquid crystals at strong confinement.



## 4. CONCLUSION

The first experimental study of spreading of LC droplets over the surface with a regular relief was performed. The main results can be summarized as follows:

1. The submicron relief provides both well-defined surface orientation and well-defined direction of spreading. It is of importance for theoretical description of spreading as material parameters of nematics (shear viscosity and surface tension coefficient) involved in such process are anisotropic.
2. The first stage of the precursor film motion looks like a shear flow of a liquid crystal along a number of separate channels of sub micron sizes. One can hope that computer simulation of such flow will result in new information about anisotropic shear viscosities in near-surface layers of LC.
3. The overall spreading of LC droplet can be considered in terms of motion of a contact line as in the case of isotropic substrates. So the effective anisotropic coefficient of the surface tension can be introduced and determined from experimental results after some modification of existing models. Such work is under progress now.
4. We have established that anisotropic spreading essentially depends on the preliminary treatment of the relief surface. It means that important information about molecular interaction of LC with nanometer surface layers can be extracted from experiments of such type.

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