Discontinuous Alignment Thin-Film Formation by Self-Organized Dewetting

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Abstract

A new discontinuous alignment layer is developed. The formation of such discontinuous structure is created by self-organized dewetting. Different dewetting mechanisms have been investigated. Such a discontinuous alignment layer can be fabricated on top of a continuous alignment layer. This heterogeneous surface for liquid crystal alignment can be used to produce arbitrary pretilt angles for the liquid crystal cell. Experiments using photo-aligned polymer have been done to verify the dewetting theory. The alignment layers thus produced are proved to be robust. Moreover, the dewetting step is a fully controllable process and is compatible with existing manufacturing techniques.

1. Introduction

Recently, nanostructured alignment layers have been successfully fabricated. Unlike homogeneous alignment layers, these new heterogeneous alignment layers can provide arbitrary pretilt angles [1-3]. These nanostructured alignment layers can either be formed by phase segregation, or by stacking a nanonetwork on top of another homogeneous alignment layer [4]. In this paper, we study the formation of a discontinuous alignment film by a dewetting technique. This is a self-organized approach without the need of any mask. This discontinuous alignment layer can be formed on another continuous alignment layer and forming a stacked structure [5-6]. The self-organized islands produced form nanostructure domains of one alignment layer on top of the other. In addition, by varying the dewetting conditions, precise control of the domain ratio becomes possible, allowing any pretilt angle to be possible for the liquid crystal (LC) molecules [7].

Here we show that this alignment layers are able to fill the technology gap for arbitrary pretilt angles on the alignment substrate, especially in the range of 30°-60°. Many applications can be made possible if such pretilt angles are available, such as bistable display devices [8] and no-bias-bend fast switching display devices [9]. By using a photoalignment material for the second layer, spatially varying pretilt angles can also be produced. The alignments produced are robust. Moreover, the processing window is also maximized.

2. Modeling of Discontinuous Thin-Films

Liquid surfaces are never completely flat. Due to temperature fluctuations, there is always a spectrum of capillary wave present. The stability of the film is determined by the transient behavior of fluctuations on its surface. The schematic drawing of the instabilities of the thin film is sketched in Figure 1.

Figure 1. An overview of the proposed stacked alignment structure.

The instabilities of thin films are basically some kind of hydrodynamics problem. For the motion of Newtonian fluid with viscosity $\eta$, density $\rho$, it can be described by Navier-Stokes equation:

$$\rho \left( \frac{\partial v}{\partial t} + (v \cdot \nabla)v \right) = -\nabla p + \eta \nabla^2 v + \rho g$$

(1)

where $p$ is the pressure within the film. If the viscosity of the applied fluid such as polymer is very high, the convective term $(v \cdot \nabla)v$ can be ignore. Furthermore, since the resulting dynamics is very slow, velocity profile can always be considered to be $\partial v / \partial t \to 0$. Therefore, the gravity is also not important at this case as well. With the above considerations, equation (1) can be rewritten as

$$\nabla^2 v = \frac{1}{\eta} \left( \nabla p \right)$$

(2)

It can be seen that the velocity only depends on the $z$-coordinate. The corresponding integration with the boundary conditions give velocity:
Hence, the mean velocity can be obtained:

\[ \mathbf{v} = \frac{1}{2\eta} (\nabla p) \mathbf{z} - \frac{h}{\eta} (\nabla p) \mathbf{z} \]  

(3)

Consider that the liquid is incompressible, the change in volume must simply be due to a variation in the level of the fluid. It can be expressed as \( \partial_t h = \nabla \cdot (h \mathbf{v}) \). Therefore,

\[ \partial_t h + \frac{1}{3\eta} \nabla \cdot \left( h^2 (\nabla p) \right) = 0 \]  

(4)

For the pressure within the film, it is actually depends on the film thickness. The pressure may come from several contributions, atmospheric pressure, surface tension pressure and excess intermolecular force. The importance of the various terms depends on the experimental conditions. Here we only consider the most important term, the excess intermolecular force, which can be written as:

\[ p_{vdW} (h) = \frac{B}{h^8} - \frac{A}{12 \pi h^3} \]  

(6)

where \( A \) is the effective Hamaker constant to describe the long range van der Waals force and \( B \) describes the short range repulsive interactions. Assuming only non-retarded van der Waals forces in the thin film geometry, therefore, there are only two possible scenarios for the film. The first scenario, Hamaker constant is negative, which means that the van der Waals forces are repulsive, pushing the film apart. Hence, the film can minimize the free energy by increasing its thickness. In other words, the film is wetting the surface and is stable. However, if the Hamaker constant is positive, which means the van der Waals forces are attractive when \( \langle h \rangle > h_c \), the film reduces its free energy by thinning. The film is hence unstable and dewets. Such dewetting is called spinodal dewetting.

By substituting equation (6) into equation (5), we can obtain the final equation to describe the dynamics of the thin film on the substrate:

\[ \partial_t h + \frac{1}{3\eta} \nabla \cdot \left[ h^2 \left( \nabla h + \nu \nabla \cdot (h \mathbf{v}) \right) + \partial_z p_{vdW} (h) \nabla h \right] = 0 \]  

(5)

Numerical simulation using time iteration method on 250x250 lattices with periodic spacing 2.5nm and time step 0.1ns has been done. We assume the mean film thickness is 7nm with initial small amplitude ±1% fluctuation. The surface tension is 30mNm\(^{-1}\), viscosity is 30mPas, short range repulsive interactions 6x10\(^{-72}\)Jm\(^6\), the effective Hamaker constant 5.3x10\(^{-29}\)J. Based on the numerical model, we are able to investigate the dependence of the mean thin film thickness as shown in Figure 2.

![Figure 2. The dynamics of the thin film with mean thickness 7nm at time (a) 0\(\mu\)s, (b) 0.06\(\mu\)s, (c) 4\(\mu\)s, (d) 8\(\mu\)s, (e) 40\(\mu\)s, (f) 81\(\mu\)s, (g) 162\(\mu\)s and (h) 292\(\mu\)s.](image-url)
fluctuation. The corresponding transition range is about 3 times wider than the continuous stacked alignment film situation.

![Graph](image)

Figure 3. (a) The roughness of the thin film. (b) Pretilt angle of a dewetted film versus wetted film.

3. Experimental Results

In order to verify the theory, several experiments have been done. Firstly, an ITO glass substrate is prepared. Figure 4 shows the AFM picture of the ITO crystalline pattern.

![AFM Picture](image)

Figure 4. AFM picture of the ITO crystalline pattern.

The substrate is then spin-coated with different concentrations of a photo alignment polymer, ROP-103, provided by Rolic. Ltd. The viscosity of the ROP-103 can be adjusted by the solvent Cyclohexagone. Since low viscosity solvent is applied, the thin polymer film will be formed during spin coating. Dewetting is expected to occur as shown in Figure 5(a) and (b). It can be seen that the alignment layers are indeed discontinuous and the “valley” and “hill” are uniformly distributed within the substrate. The average domain size is about 400nm to 580nm. However, when the concentration of the ROP-103 increases, the viscosity of such mixture will also increases. Thus, thicker film will be obtained after spin coating. Eventually, the solution cannot undergo dewetting. Hence, a continuous alignment layer is formed as shown in Figure 5(c).

![AFM Pictures](image)

Figure 5. AFM pictures of the photo alignment layers with different concentrations. (a) 6% (b) 8% (c) 10%.

The effect of the surface tension has also been verified by experiments. The Two substrates are coated with same vertical polyimide called S1 and S2, which are prepared by applying different cycling temperature. S1 is baked with 180°C while S2 is baked with 230°C. The substrates are then exposed under DMF solvent vapor in dry box for 12 hours. The corresponding surface contact angles are then measured. Figure 6 shows the measured result. It can be seen that S1 has a larger contact angle then S2.
Then the substrates are coated with different concentration of ROP-103 solutions to form stacked alignment surface. The roughness and pretilt angle of the films have been investigated. It is found that the stronger surface tension alignment layer, such as substrate S1 will induce thicker and larger domains. The corresponding pretilt angles will change in a more gradual way. On the other hand we found abrupt drop of pretilt angle for substrate S2 which has lower surface tension. Such findings are agreed with our simulations in the previous section.

4. Conclusion

We have demonstrated a new discontinuous alignment surface that is produced by dewetting. The dewetting technique can precisely produce different domain size of a heterogenous liquid crystal alignment surface. This alignment surface is capable generating arbitrary pretilt angle. And the results are highly repeatable due to the promising dewetting process. Different pretilt angle profiles can also be achieved by varying different dewetting conditions. Such alignment layer is particularly useful for liquid crystal switchable lens, bistable displays, multi-domain displays and wave-guide devices.

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6. References


Figure 6. Contact angles of substrates S1 (left) and S2 (right).

Figure 7. (a) Roughness and (b) pretilt angles for substrate S1 (180°C) and S2 (230°C).