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A Review of Nanostructured Alignment Layers for Liquid Crystals

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We review some recent investigations of nanostructured alignment surfaces for liquid crystals. These surfaces are heterogeneous and yet can give uniform alignment of the liquid crystal and are able to give any pretilt angles in the range of 0–90° reliably and controllably. The conditions for having such uniform alignment properties are studied. Three techniques for the generation of such nanostructured surfaces are reviewed.

Keywords Liquid crystals, nanostructures, alignment surfaces, displays, anisotropy

1. Introduction

A long standing problem in liquid crystal physics is how to obtain intermediate pretilt angles of the liquid crystal (LC) molecular director on the substrate surfaces [1–9]. In many applications, such as in liquid crystal displays (LCD), it is desirable to have high pretilt angles in the range of 10°–80°. In fact, many new LCD operating modes can be designed with such high pretilt angles. Having one additional adjustable parameter will allow better optimization of the LCD. High speed operations such as the no-bias bend mode (NBB) [10] and the stressed splay bend (SST) mode [11,12] can be made. Additionally, nematic LC based bistable displays can be fabricated with high pretilt angles. The bistable bend-splay (BBS) [13], bistable bend-twist (BBT) [14], bistable splay-twist (BST) [15] modes are all based on high pretilt angles. Liquid crystal polymer retardation films such as O-plates can also be made if high pretilt angle is available [16].

Alignment of liquid crystal molecules is most commonly achieved by rubbing of polyimide thin films coated on glass substrates. Unfortunately common polyimide alignment layers either provide planar alignment (PA) in the range of 0–5° or vertical alignment (VA) in the range of 85°–90° to the x-y plane. There have been quite a few obtaining large pretilt angles is by SiO₂ evaporation [17]. However this technique is not amenable to large area substrates or for mass production. Photoalignment has been proposed as an alternative where the pretilt angle can be controlled by the exposure time [18,19]. However, reliability and reproducibility are in doubt.

By far, the most common approach is to mix VA and PA alignment materials together. Intuitively, this should give an intermediate pretilt angle if the mixing is uniform. Filas et al attempted to mix a VA surfactant and a PA polyimide together in order to obtain intermediate angles [20]. The results were not too satisfactory in providing controllable

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pretilt angles. Many groups tried to mix the VA and PA components in a polymer blends and in co-polymers [21–23]. There are some reported results but continuously controllable pretilt angles could not be obtained. None of the mixing approaches has proven to be useful so far.

The difficulty with all previous approaches is that they all aimed at fabricating a homogeneous mixture of VA and PA materials in a single layer. This goal is first of all difficult to achieve, since different materials may not blend too well together in the solid phase. Moreover, we shall show in the next section that, even if such a homogeneous mixture can be made, it will not have stable aligning properties and the process window is very narrow. In fact, “a winner takes all” situation will occur and the resulting pretilt angle will either be planar or vertical.

Instead of a homogeneous and uniform alignment layer, we proposed and showed that a heterogeneous alignment layer consisting of nanodomains of PA and VA materials can deliver any pretilt angles between 0–90° [1–3]. Actually nanodomains of different alignment materials have been studied before [24,25]. Yokoyama et al used direct molecular writing to obtain regular nanodomains with different planar alignment directions [24]. Interesting tristable LCD could be made. Tsui et al have also demonstrated that the pretilt angle of such PA nanodomains shows a “phase transition” from low pretilt to high pretilt as the size of the domains decreases. However these studies were limited to planar aligned domains in different planar directions. Also the high pretilt angle obtained is not controllable to arbitrary values.

In this paper, we shall review the theory of heterogeneous alignment surfaces consisting of VA and PA domains. Experimental results will also be shown. Three different methods have been demonstrated. The first two methods rely on self assembly of random nanodomains. The newest method, nanostructures of the third kind, does not involve any random formation of nanostructures and can be made in large areas uniformly. It is a promising technique to make large area LC cells with large pretilt angles for various applications.

2. Principle of Nanostructured Alignment Surfaces

Assume that the alignment layer consists of nanodomains of VA and PA materials. If the typical size of the nanodomain λ is small, a uniform pretilt angle θ_{AV} can be obtained inside the bulk of the liquid crystal cell. The value of θ_{AV} obtained is determined by p , the fraction of the area that is VA. For $p = 1$, the surface consists totally of VA materials. The pretilt angle is that of the VA alignment with value θ_1 , which is typically a few degrees or less from the z-axis. For $p = 0$, PA alignment is obtained with an angle of θ_2 , which is typically a few degrees or less from the x-y plane.

The behavior of the θ_{AV} as a function of p is determined by the extrapolation lengths $\ell_{ei} = K_{11}/W_{\theta i}$ where K_{11} is the splay elastic constant of the liquid crystal, $W_{\theta i}$ is the polar anchoring energy of the alignment material, and $i = 1$ for VA domains and $i = 2$ for PA domains. If $\lambda \ll \ell_{ei}$, then bulk elastic energy dominates over surface energy and the behavior of θ_{AV} is quite nonlinear in p . It can be shown that for this case of very small domains or for very weak anchoring, the pretilt angle is given by [1,7]

$$\tan 2\theta_{AV} = \frac{pW_{\theta 1}\sin 2\theta_1 + (1-p)W_{\theta 2}\sin 2\theta_2}{pW_{\theta 1}\cos 2\theta_1 + (1-p)W_{\theta 2}\cos 2\theta_2} \quad (1)$$

In the opposite limit of $\lambda \gg \ell_{ei}$, corresponding to very strong polar anchoring, θ_{AV} is almost linear in p , and given by

$$\theta_{AV} \approx p\theta_1 + (1 - p)\theta_2 \quad (2)$$

Now we can examine the case of a homogeneous mixture of VA and PA materials and show that it will not work. The case of a homogeneous mixture can be regarded as the limiting case of $\lambda \rightarrow 0$ for a heterogeneous mixture. Thus Eq. (1) can be applied. In this case, the dependence of θ_{AV} on p is highly nonlinear. Figure 1 shows θ_{AV} versus p for four cases of $\theta_1 = 85^\circ, \theta_2 = 5^\circ$; $\theta_1 = 87^\circ, \theta_2 = 3^\circ$; $\theta_1 = 88^\circ, \theta_2 = 2^\circ$ and $\theta_1 = 89^\circ, \theta_2 = 1^\circ$. Here we assume that $W_{\theta_1} = W_{\theta_2}$ for the sake of simplicity. It is obvious from eq. (1) that $\theta_{AV} = 45^\circ$ at $p = 0.5$ for all cases. Moreover, it is easy to see that the θ_{AV} versus p curve becomes a step function when $\theta_1 = 90^\circ, \theta_2 = 0^\circ$. Thus, from Fig. 1, it can be seen that the process window for obtaining the desired pretilt angle is very small as the domain size approaches zero, i.e. for a homogeneous mixture of VA and PA materials. In the extreme case of $\theta_1 = 90^\circ, \theta_2 = 0^\circ$, the alignment is either PA or VA. The situation can be described as “winner takes all.”

It can be argued that the surface energy term used in deriving eq. (1) assumes only second order terms in the Rapini expansion [6,7]. If higher order terms are included, the slope of the θ_{AV} versus p curve may remain finite even for homogeneous mixtures. But it can be shown that the process window is still small. Moreover the anchoring energy is very small as well for such homogeneous mixtures. Thus in general, for the present nanostructured alignment surface to work properly, it is required that $\lambda \geq \ell_{ei}$. In fact, we should require $\lambda \gg \ell_{ei}$ in order that the linear relationship in Eq. (2) applies. However, λ should not be too large either. It is obvious that if λ is too large, the optical properties of the LC cell will become heterogeneous as well. It is not good since we want the optical properties of the LC

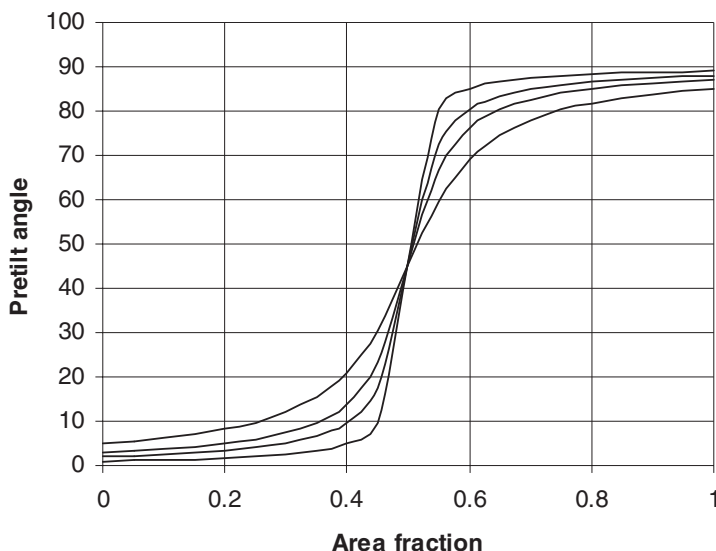


Figure 1. Calculated dependence of pretilt angle on the area fraction (V/V+H), for (θ_1, θ_2) of $(85^\circ, 5^\circ)$, $(87^\circ, 3^\circ)$, $(88^\circ, 2^\circ)$ and $(89^\circ, 1^\circ)$ respectively for the 4 curves. The curve for $(90^\circ, 0)$ will be a step function.

cell to be homogeneous even though the alignment surface is heterogeneous. The largest value of λ is determined by the LC cell gap d . As shown in the simulation in reference [1], the LC director achieves a uniform value in a distance of $\lambda/3$ into the bulk. Thus it is reasonable to require that $\lambda/3 \ll d$. Thus the condition on the nanodomain in the present structure is

$$\ell_{ei} \ll 1 \ll 3d. \quad (3)$$

Fortunately this condition is easily satisfied in most cases. For typical values of K_{11} and $W_{\theta I}$, the value of ℓ_{ei} is ~ 15 nm. Thus the optimal domain size is $0.15\text{--}0.3\ \mu\text{m}$ for the nanostructure, even for LC cell gaps as small as $1\ \mu\text{m}$. The nanostructure can be even larger for larger cell gaps. If eq. (3) is satisfied, then the dependence of θ_{AV} on p is describable by the linear relationship in eq. (2). Thus the process window for the pretilt angle is large and easily controllable. We shall show that this is indeed the case experimentally.

3. Nanostructure of the First Kind

Three methods have been proposed and demonstrated to fabricate such inhomogeneous alignment surfaces reliably. In the first method, which we call nanostructure of the first kind, the inhomogeneity is obtained by differential precipitation of the vertical and planar alignment polyimide from a common solution [2,4,6,7]. A common solvent is used to dissolve both the PA and VA polyimides. The solution is coated onto the substrate either by spin coating or by doctor blade. Upon drying of this mixture, differential precipitation will occur due to the difference in solubility. Figure 2 shows the atomic force (AFM) micrograph of such a nanostructure. It can be seen that the PA material first comes out of the solution and forms a thin film. The VA material forms a network on top of the PA materials. The nanostructure feature size is about $0.2\ \mu\text{m}$, which is just right. The preferred direction can be obtained by mechanical rubbing of the cured polyimide in the usual manner. In reference [4], a variation of this method was proposed. In this variation, photoalignment was used to provide the preferred azimuthal direction [4]. In reference [8], another variation was proposed where ion beam irradiation was used to provide the alignment direction.

Figure 3 shows the measured pretilt angle as a function of the fractional VA area. It can be seen that the curve is approximately linear indicating that eq. (3) is satisfied. There is a large processing window. Indeed this method is superior in making LCD with any pretilt angles. Many studies of new LCD modes [10–12] and bistable LCD [13–15] were accomplished using this method. It has been shown that this nanostructured alignment layer is robust against high process and operating temperature, and the anchoring energies are as large as the individual polyimides [5].

The major drawback of these methods is that the domains are formed by a random process. Even though experimentally it was verified that they are reproducible and reliable over a small area, the same cannot be said of larger substrates. The randomness in the formation of the nanostructure is also somewhat undesirable from a manufacturing point of view.

4. Nanostructure of the Second Kind

In the second method, vertical and planar materials are coated separately [26–30]. The idea of this nanostructure of the second kind is not to improve the processability of the alignment layers. Rather, it provides an additional control of the alignment properties by

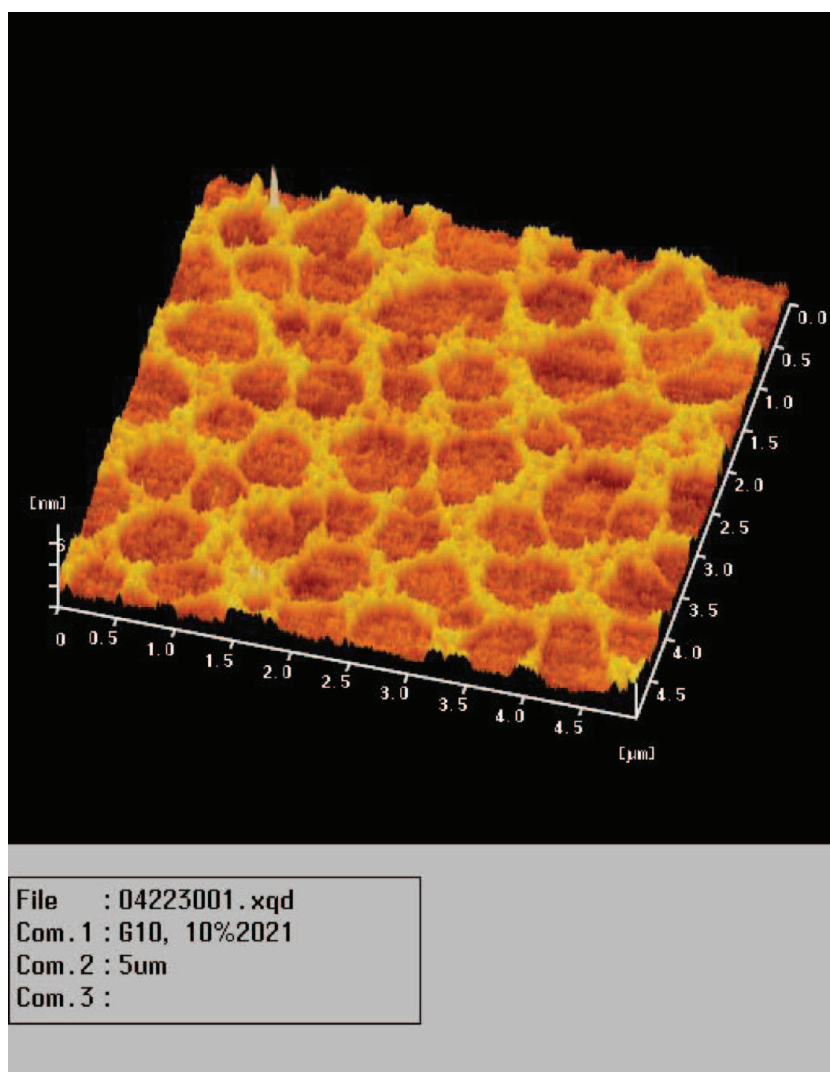


Figure 2. Typical AFM picture of nanostructure of the first kind.

using a photoalignable material. In this second method, a layer of VA polyimide is first applied to the substrate. It is cured and allowed to cover the substrate continuously. Then a second layer of photoalignment material, such as commercial photopolymers, is applied on this first surface. The interface energy of the two materials should be such that there is dewetting or droplet formation. Again the droplet size and area fraction of the droplet is important for the heterogeneous alignment mechanism.

Zhang et al. [29] actually studied this stacked layer process for the cases of complete wetting and dewetting. They were able to demonstrate that if complete wetting occurred, the second layer was continuous over the first layer. Then the pretilt angle obtained goes from 90° to 0 abruptly as a function of the thickness of the second layer. If dewetting occurred, the second layer broke down into small domains on top of the first layer. In this case the pretilt angle obtained is more or less a linear function of the “average thickness” of the

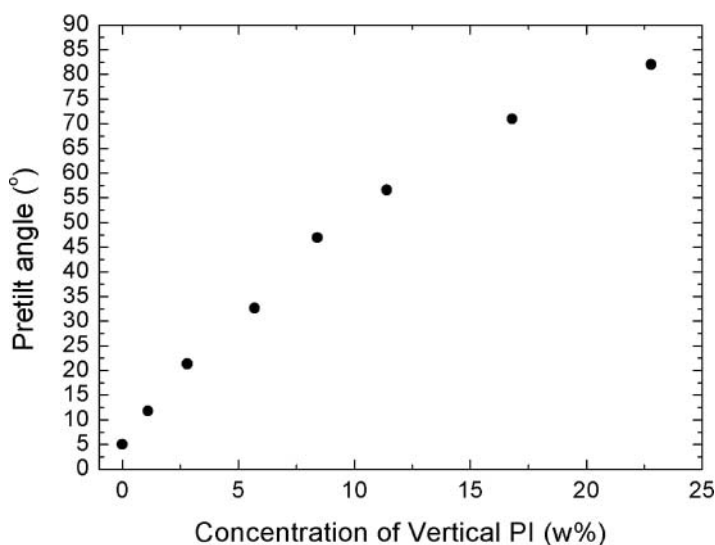


Figure 3. Measured pretilt angle as a function of the area fraction.

second layer. This observation is entirely in agreement with the analysis given in section 2. If the second layer of PA material is continuous, then it is in competition with the bottom layer of VA material. It is because if the top layer is thin, the bottom layer will dominate in terms of determining whether PA or VA is achieved, due to extended effect of the potential field [31]. This situation is physically equivalent to nanostructure of the first kind with infinitely small domains. We have shown previously that this situation will lead to a highly nonlinear step function like behavior of the pretilt angle as a function of the fractional area of the VA or PA material. In this case, the fractional area is dependent on the extended field effect of the bottom layer and is quite complicated to determine. Nevertheless, there is a critical thickness of the top layer which will give equal aligning effect of the top layer and the bottom layer. This is equivalent to $p = 0.5$ for the nanostructured surface.

For the opposite case of having nanodomains of the second “layer” on top of the first layer, the competition between the two alignment materials will be such that a gentle linear change in pretilt angle is obtained. This is the more desirable case. Figure 4 shows an AFM picture of the resulting surface obtained. Islands of the second material are clearly seen on top of the bottom layer. The measured pretilt angle as a function of the concentration of the second material is plotted in Fig. 5. The concentration is directly proportional to the area fraction ($1-p$) of the second PA material. The exact dependence may not be linear and can be a rather complicated function of the dewetting process. We shall not attempt to model that process here even though it can be done [26].

An additional feature of this “nanostructure of the second kind” is that the second layer of PA material can be photoalignable polymer. Since the degree of polymerization of the photopolymer depends on light dosage, the fraction of the second layer that will remain after washing will depend on the exposure light dosage. If the light beam has spatial dependence, then the fractional area will be spatially dependent. Mathematically, the area fraction is dependent on (x, y) through the relationship

$$p = p(I) = p(I(x, y)) \quad (4)$$

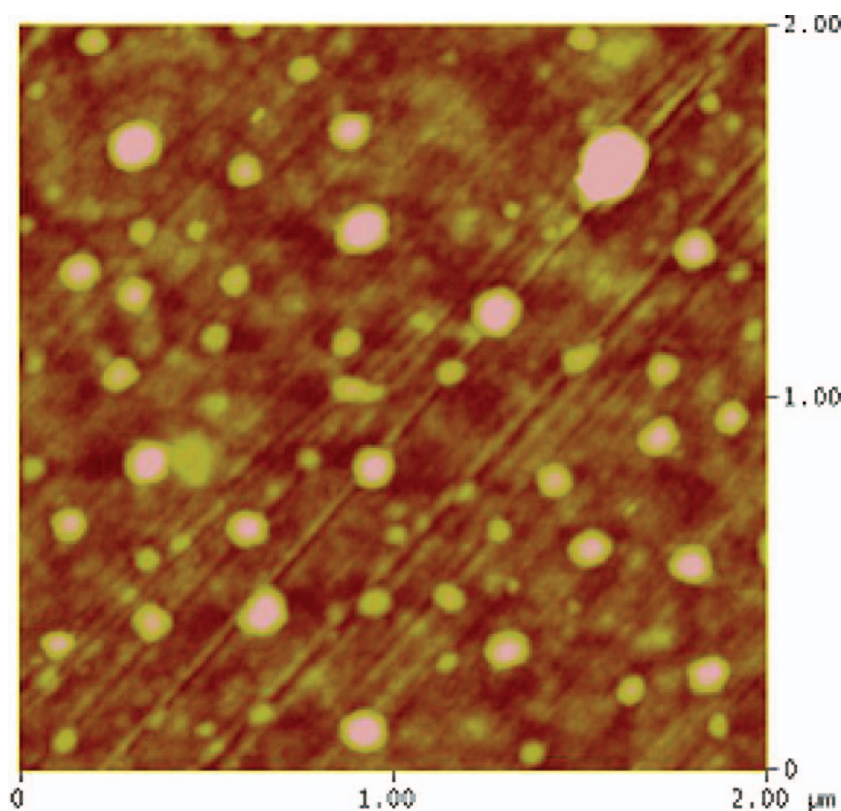


Figure 4. Typical AFM picture of nanostructure of the second kind.

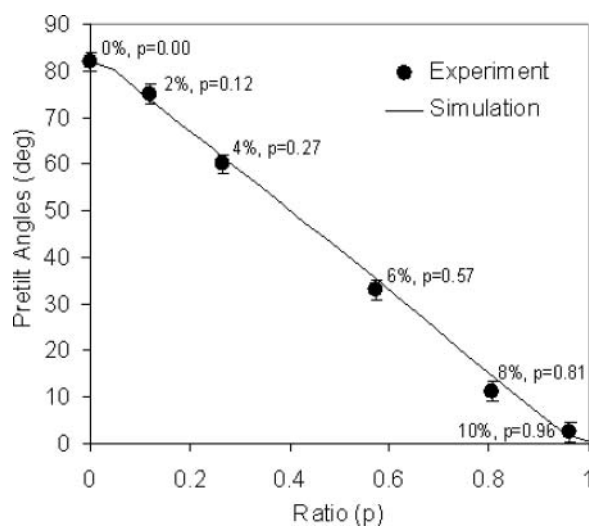


Figure 5. Measured pretilt angle as a function of the area fraction. A nearly linear dependence is obtained. The numbers correspond to concentration of the photoalignment material in the top layer.

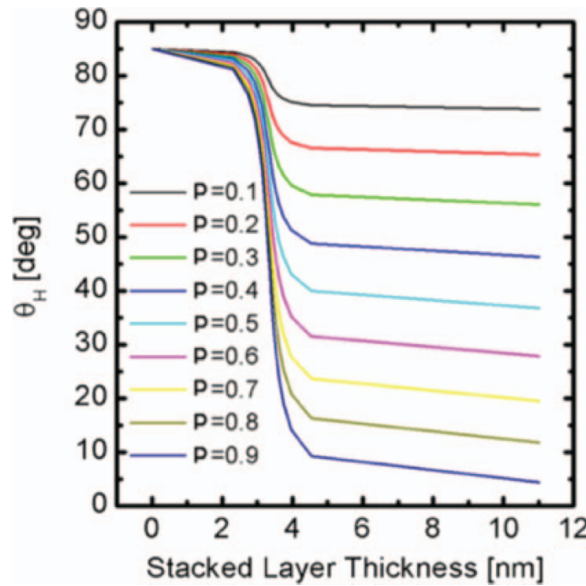


Figure 6. Calculated pretilt angle obtained as a function of light dosage.

This is an important feature of this nanostructure of the second kind. The pretilt angle obtained can have a designed spatial (x, y) dependence by controlling the light intensity [32]. Figure 6 shows the calculated pretilt angle as a function of both concentration of the second layer and the photoalignment light dosage. The x -axis is the average thickness of the second photoalignment layer. It is a measure of the light dosage used to polymerize the second layer. The parameter p is the area fraction if the second layer is fully polymerized. It can be seen that if the light intensity is infinitely large, the pretilt angle will be given by the concentration of the second layer only, as seen in Fig. 5. However if the light dosage is zero, all of the second layer will be washed away and the pretilt angle will be that of the bottom layer which is VA. At intermediate light dosages, an intermediate pretilt angle between these two extremes will be obtained. If the light intensity is dependent on (x, y), then the pretilt angle will have spatial dependence $\theta_{AV}(x, y)$. This is something that no other technique can achieve. We have successfully made a variable focal length liquid crystal lens using this technique [32]. Presumably other electrically controlled liquid crystal optical elements, such as a beam steering device, can be made using this nanostructure of the second kind.

5. Nanostructure of the Third Kind

In principle nanofabrication techniques can be used to pattern the desired nanostructures [33]. However, it has been shown that the feature size has to be $0.15\text{--}0.3\ \mu\text{m}$ for the nanostructured alignment layer. To perform photolithography with such feature size over large areas, such as those needed for LC display production, is impractical. Moreover, it is difficult to find an etchant for one kind of polyimide (VA or PA) that does not affect the other kind.

In “nanostructure of the third kind,” a patterning technique is used to produce the nanostructure in a single step. It is based on the observation that a vertical alignment

polyimide can be turned into planar alignment by exposing the surface to an oxygen plasma or by deep UV irradiation. It is because that a vertical alignment polyimide usually consists of a planar alignment polyimide with the addition of a long alkyl side chain [7,21–23]. If the side chain is destroyed without destroying the main chain, then the vertical alignment polyimide becomes a planar alignment polyimide.

Figure 7 shows the verification of this observation. The pretilt angle of an anti-parallel rubbed cell is measured as a function of irradiation time of the VA polyimide by an oxygen plasma. The VA material used is a commercial one. It can be seen that the pretilt angle measured changes from 90° to 0° as the plasma dosage is increased. The transition is quite sharp. In principle any pretilt angle can be obtained by simply using this irradiation method. But it is not certain that the process window can be reasonably wide. But in terms of converting VA to PA alignment, the use of O_2 plasma irradiation method is certainly a good one.

Using this observation, an inhomogeneous alignment surface can be obtained if we use photolithography to protect parts of the surface during O_2 plasma irradiation. As mentioned above, we need to have a technique that can process substrates of over 1 m in size with fine features of 150–300 nm. It is not practical to use normal type of light exposure photolithography. Fortunately in the present case, the pattern required can be accomplished by nanoimprinting.

One very simple structure that can be used is a subwavelength grating. Such a nanoimprint mold can be made using laser beam interference over a large area. In this technique, the VA polyimide layer was first coated onto a glass substrate, followed by baking and curing. Then a photoresist can be coated on it and nanoimprinting performed. The imprinted substrate was placed in a reactive ion etching (RIE) machine for O_2 plasma exposure. The

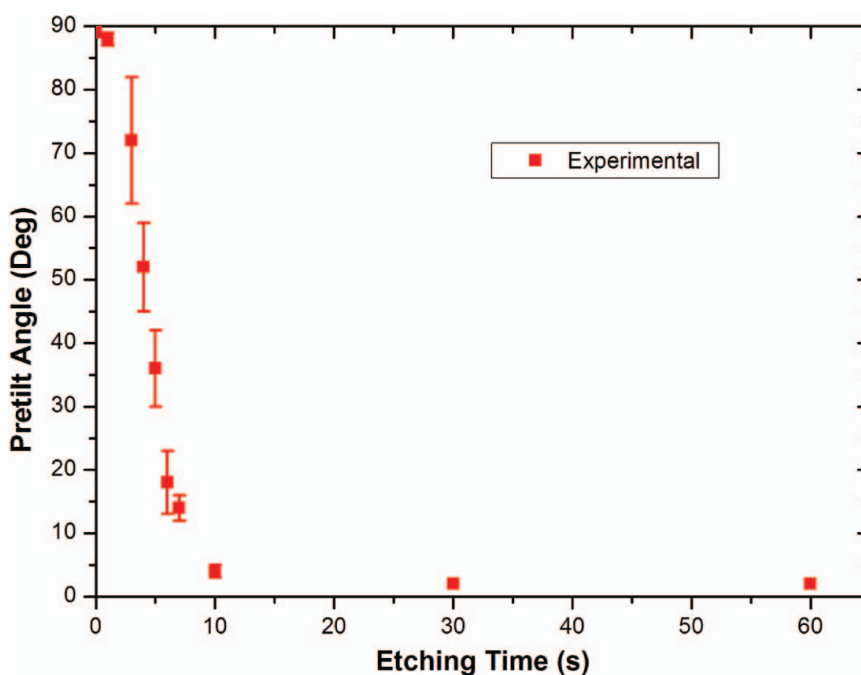


Figure 7. Transformation of a VA alignment to a PA alignment by O_2 plasma modification.

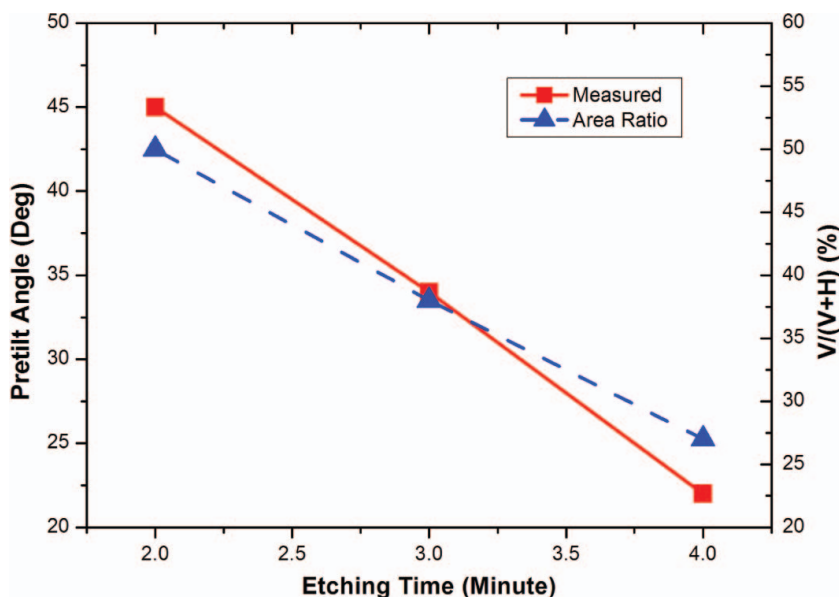


Figure 8. Measured pretilt angle as a function of O_2 plasma exposure time.

exposure time was used as a parameter for optimization of the pretilt angles. After RIE, the photoresist was removed, leaving behind a grating structure of alternating lines of PA and VA polyimides.

Preferential alignment direction of the nanostructure can be provided by rubbing of the final surface or by rubbing before photoresist removal. The procedure is the same as in conventional polyimide alignment. The alignment property of this surface was tested by assembling an anti-parallel rubbed homogeneous alignment liquid crystal cell using positive liquid crystal. Figure 8 shows the measured pretilt angle as a function of the O_2 plasma etching time. It can be seen that the pretilt angle changed from 45° to 25° from an exposure time of 2 to 4 min. Since in the present experiment, the aperture ratio of the grating was 1:1, the area ratio of the PA and VA alignment regions should also be 1:1 and the pretilt angle obtained should be 45° . The decrease in pretilt angle observed is due to the over-etching of the VA polyimide as covered by the photoresist. A longer exposure time will over-etch the VA polyimide leading to a large PA/VA area ratio and hence a smaller pretilt angle, as predicted by Eq. (2).

The range of pretilt angles obtained using this method is not as large as those methods investigated before [1–4]. The smallest value of VA polyimide area coverage is 50% unless a different grating mask is used. Thus in principle the pretilt angle goes only from 45° to 0° . Larger angles require grating with aperture ratio smaller than 1:1. Thus the present technique is only good for making LC cells with pretilt angles of near 45° . This is fine for making NBB cells and BBS bistable displays [10,13].

6. Summary and Conclusion

In summary, a new kind of heterogeneous alignment surface for liquid crystals has been successfully developed. Provided the sizes of the nanodomains are properly chosen, a uniform pretilt angle over a large area can be reliably produced. The range of angles

obtained is very large, from 0 to 90°. We have introduced three methods to fabricate such nanodomains. In the first method, a random precipitation process is used to obtain the nanostructure. It is a quick and easy method, many experiments and LCD mode designs can be accomplished using this method. However, the large area capability of this approach is limited.

In the second method, a two-layer deposition procedure is introduced. It is interesting because a photoalignment material can be used as the second layer so that there is an additional degree of control over the pretilt angle. Spatially dependent pretilt angles can be obtained, which can be useful for some applications, such as in tunable optical elements.

In the third method, nanoimprinting of the nanostructure is introduced. Randomness is taken out of the picture totally. This method is robust and amenable to large area processing. It can also be very uniform and reproducible. As such, this nanostructure of the third kind can be applied to large substrates in LCD manufacturing. We believe this technique will have great potentials for applications in the fabrication of fast LCD modes as well as in making bistable devices.

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