Submicron liquid crystal pixels on a nanopatterned indium tin oxide surface

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We have prepared a grooved indium tin oxide (ITO) surface with groove widths of ~40–90 nm and a variable groove separation up to 36 μm using atomic force microscopy nanolithography. Twisted nematic pixels with 4-*n*-pentyl-4-cyanobiphenly (5CB) liquid crystal were prepared using a rubbed polyimide counter plate. The 5CB molecules align along the direction of the grooves, but no pretilt angle was observed on the ITO surface. The surface anchoring energy appeared not to depend on the groove separation nor on the scan force. The lateral correlation length of 5CB on ITO was measured to be 2.2 μm. If a grooved surface is scanned in another direction, liquid crystal molecules follow the direction of the last scan. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1484556]

In liquid crystal displays (LCDs) liquid crystal molecules should be aligned in order to have a collective response to an electric field over a large area. Presently, the most common method of alignment is accomplished by the rubbing of a thin polymer film that is coated on a thin conductive electrode on a glass substrate. The detailed structure of the polymer alignment layer determines the anchoring strength and the pretilt angle of the liquid crystals.[1,2]

The question with large practical implications is: would it be possible to merge two separate layers that are used for alignment and electrode into one conductive alignment layer? To achieve this, one might consider alignment on conductive polymer films or using conductive oxides. In this letter we will discuss the second approach.

Indium tin oxide (ITO) is the most popular electrode for liquid crystal displays due to its high optical transparency and relatively good electrical conductivity. Depending on the method of preparation, ITO films can have different surface structures, conductivity and transparency. If ITO is prepared by rf sputtering, the resulting film usually will have a granular structure. The ITO surface is much harder than the polymer surface. Therefore, conventional methods that are used for rubbing polymer films can hardly deform the surface of ITO. Attempts to align liquid crystal molecules on rubbed ITO films lead to a very weak alignment.[3]

Following our former approach to study the alignment of LC molecules on nanopatterned polymer surfaces,[4] we have performed a series of experiments to study the alignment of LC molecules on nano patterned ITO surfaces using the tip of an atomic-force microscope (AFM). We have found that a good alignment of LC molecules on submicron patterned ITO surfaces can be obtained, with an azimuthal anchoring energy that is comparable to that of rubbed polymer films like polyimide (PI). However, in contrast to rubbing no pretilt angle is found by the AFM technique.

Indium tin oxide coated glass was purchased from Applied Film Corporation[5] and consisted out of 19 nm ITO coated by rf sputtering over a polycrystalline SiO_2 (20 nm) layer on a 1.1 mm sodalime glass substrate. The ITO layer had a granular structure with grain size of the order of 5–10 nm and was polished by the manufacturer to obtain optical quality for LCD application. The average surface resistance was 125 $\Omega/\square$.

Patterns of parallel lines (grooves) were engraved by using a Dimension 3100 AFM[6] in contact mode lithography operation. We used tips with high spring constants ($K = 30-110$ N/m) to apply a large force on the surface of ITO. Square patterns with dimensions of the order of 12.8–116 μm with different scan line separations and different scan forces were prepared. Tapping mode AFM was used to image the patterned areas. Some of the patterned areas were imaged also by scanning tunneling microscopy (STM). (Note: in recent works, Sheng[7] and Clark[8] address the situation of micropatterned surfaces where a competition between homeotropic and planar alignment is created, whereas we discuss the creation of an uniaxial planar anisotropy by creating submicron groves.)

Twisted nematic (TN) cells consisting of a patterned ITO glass on one side and a rubbed PI glass on the other were made with thicknesses of 2.5–6 μm and were filled by 4-*n*-pentyl-4-cyanobiphenly 5CB at $T = 40\,^{\circ}$C in its isotropic

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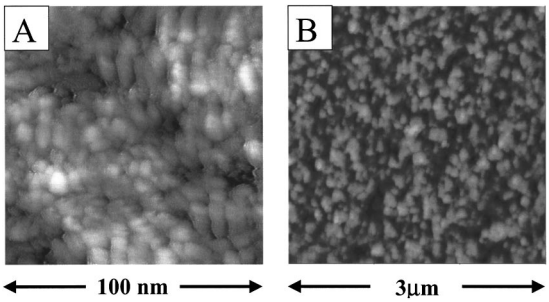

FIG. 1. (A) STM image of ITO surface, Scan size 100 nm×100 nm and scan height 5 nm. (B) AFM image of ITO surface, scan size 3 μm×3 μm and scan height 7 nm.

phase. Cells were then slowly cooled down to room temperature (23 °C). The liquid crystal alignment was studied by video microscopy with a Leica[9] polarizing microscope and a digital Matrox 12 bit camera. Twist angles were measured by pixel intensity analysis in every TN micropixel. The cell rotation method[10] was used to calculate the surface anchoring energies, which was measured as function of the scan line separation and scan force.

Figure 1 shows a STM and an AFM image of an ITO surface prior to patterning. The granular structure of the ITO film can be clearly seen in the STM image as well as pinhole defects and height and depths variations of the order of 15 nm. The ITO grains are cigar shaped with a width of ~5 nm and a length of ~20 nm. Due to the presence of the voids and rough surface topography, the ITO surface is very sensitive to pollution. In particular, organic pollution like fat, cannot be removed easily by conventional cleaning and rinsing methods. Hence, the rubbing of such a surface will align the pollution molecules which in turn may lead to the alignment of LC molecules. Wet chemical etching of ITO caused a considerable increase of the roughness of the surface. Therefore, we used oxygen plasma sputtering to remove the first few surface layers to get rid of the organic pollution. The ITO surface before and after cleaning by oxygen plasma was scanned by STM. No recognizable change of the surface structure was observed. However, the optical reflection showed a change of color indicating that the ITO film thickness was slightly reduced.

The surface of ITO was patterned with a large scan force of the order of 10^{-3} N, by using the contact mode. Figure 2 shows STM [Fig. 2(A)] and AFM [Fig. 2(B)] images of the surface of ITO after patterning. Under this large scan force,

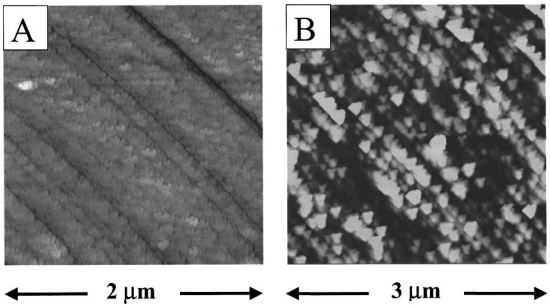

FIG. 2. Image of patterned ITO surface (A) STM image with scan size 2 μm×2 μm and scan height 10 nm, Scan angle: 45° and line separation 200 nm. (B) AFM image, scan size 3 μm×3 μm and scan height 12 nm, Scan angle: 45° and line separation 200 nm.

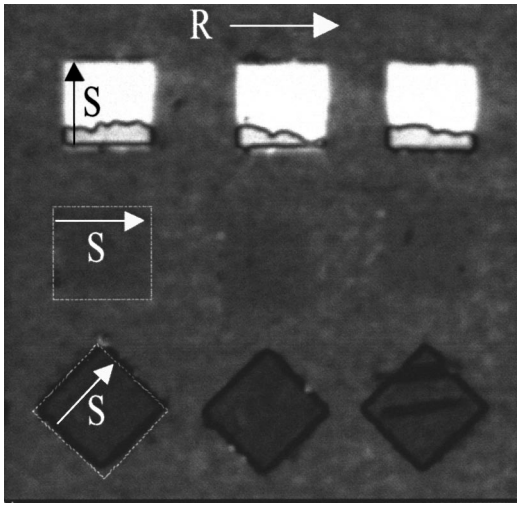

FIG. 3. Micrograph of the TN pixels. The scan direction (*S*) in the upper row is normal, in the middle row is parallel and in the lowest row is under on angle of 45° with respect to the direction (*R*) of the rubbed PI. The analyzer was set parallel to the rubbing direction of the PI. The polarizer is under 60° with respect to the analyzer.

the tip creates grooves in the ITO layer, that form an unaxial potential for the LC molecules.[11] The depth of the grooves is a function of the applied force and increases from 1.5 to 5 nm when increasing the force from 1.5 to 8.15 mN. As one expects, the average groove width also increases by increasing the scan force.

To study the alignment of LC molecules on the ITO surface, TN cells of a patterned ITO surface and a rubbed PI counter surface were prepared. Rubbed PI is known to strongly anchor the LC molecules in the direction of rubbing and therefore can be used as a reference surface. The direction of the grooves was oriented normal to the direction of the rubbed PI. Figure 3 shows optical micrographs of a series of TN pixels. Liquid crystals are well aligned in the scanned area and form TN pixels. It can clearly be seen that liquid crystal molecules on the nonscanned area are not aligned (as a comparison: a TN cell prepared by rubbing ITO with a standard velvet cloth did not show any alignment).

Different LC domains can be clearly seen in most pixels of Fig. 3. Formation of these domains indicates that there is no pretilt angle on the patterned ITO surface,[1] which is consistent with our former work on the alignment on an AFM patterned PI surface.[4] Therefore, the LC molecules can form either a left- or a right-handed twist that energetically are degenerate. Note that these cells have been filled in the isotropic phase of 5CB and therefore there is no flow alignment on the patterned surface to remove this degeneracy.

The lateral correlation length of liquid crystals on a surface can be measured directly by AFM patterning, by using grooves with larger separation (e.g., $d>3$ μm). In TN cells prepared with these larger groove separations, the LC molecules do not form a homogeneous alignment on the whole area of the pixel, but they are only aligned in the vicinity of the scanned area (see Fig. 4). The long-range order of the molecules extends the alignment over a distance ξ (lateral correlation length) from the anchored molecules. Therefore, the single line TN structure that is depicted in Fig. 4 has a width of 2ξ. In this way we found for the ITO surface $\xi = 2.2$ μm. In general the lateral correlation length ξ depends

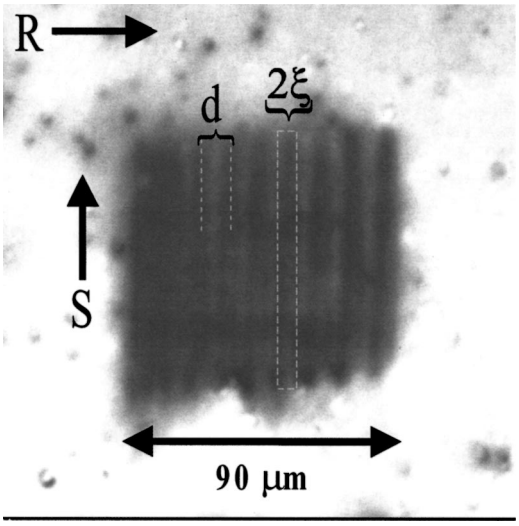

FIG. 4. Micrograph of TN pixels when the line separational is much larger than the LC transverse correlation length ξ, leading to separated and well oriented areas of width 2ξ. *S* and *R* indicate scan and rubbing direction of the ITO and PI surface, respectively.

on the nature of the liquid crystal molecules and their interaction with the surface.

As another measure of the quality of the anchoring of liquid crystal molecules on the ITO surface, we measured the surface anchoring energy W_φ of 5CB on patterned ITO surfaces by using the cell rotation method. We found $W_\varphi = 1.9 \times 10^{-5}$ J/m^2, which is similar to that observed on rubbed PI surfaces. The surface anchoring energy did not depend on the groove separation and the scan force. Decreasing the groove separation does increase the pixel contrast but this is due to an increase in the number of molecules that are strongly anchored to the surface. Increasing the scan force resulted in an increase in the groove depth and width but did not affect the surface anchoring energy.

Figure 5 shows a micrograph of ITO TN pixels. Here first a large patterned surface was scanned on the ITO surface and then a smaller square pattern was scanned on the formerly scanned area in a direction of 45° with respect to the last scan. As one can see the LC molecules align in the direction of the last scan. Although this has been explained in the case of PI by polymer chain realignment,[4] in the case of ITO such a mechanism cannot exist. We have tried by tapping mode scanning to see the second series of the grooves on the top of the first ones. However, due to the rough ITO surface we could not see any trace of any grooved structure

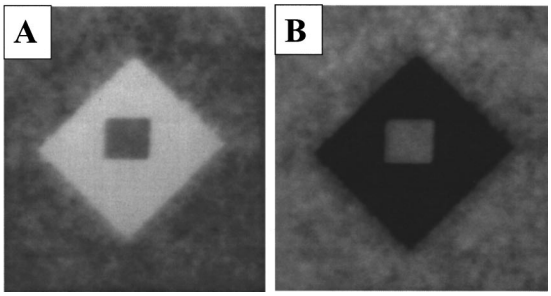

FIG. 5. Polarizing microscope micrograph of TN pixels on the ITO surface. The analyzer was set parallel to the rubbing direction of PI (0°) and the polarizer is at 45° (A) and 135° (B). The larger squares (90 μm$\times$90 μm) are scanned with 512 scan lines in 45° direction and the smaller squares (25 μm$\times$25 μm) were scanned with 512 lines in a direction of 0°.

in the doubled scanned area and a clear explanation is still missing.

In summary we have shown that an ITO surface patterned by contact mode AFM can be used to directly align LC molecules, in this way avoiding the use of an additional polymer alignment layer. The anchoring energy on nano patterned ITO was found to be equally strong as that on rubbed PI and led to very stable alignment. The major challenge for future applications will be the introduction of a pretilt angle, which is zero in the present situation.

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