Polymer-Layer-Free Alignment for Fast Switching Nematic Liquid Crystals by Multifunctional Nanostructured Substrate

*Woo-Bin Jung, Hyeon Su Jeong, Hwan-Jin Jeon, Yun Ho Kim, Jeong Yeon Hwang, Jae-Hoon Kim, and Hee-Tae Jung**

Current liquid-crystal-display (LCD) technology relies on the use of an alignment layer[1] along with polymer rubbing and photoalignment[2–4] to align liquid-crystal (LC) molecules. However, future designs of high performance, 3D, low energy consuming displays will not rely on alignment layers to bring about uniform alignment and fast response of LC molecules. The major reason for the need of a new design strategy is that alignment layers in these displays cause transmittance degradation, refractive index mismatching, undesired capacitance, and low thermal and physical stability.[5–8] As a result, techniques must be uncovered to rapidly align LC molecules on electrode substrates, without any defects over large areas, without utilizing polymer layers.[9,10] Although the potential exists that asymmetric nonnematic LCs can be used for this purpose,[11,12] it is difficult to design the proper LC molecules because of their symmetric breaking characteristics, the narrow temperature ranges over which they can be used, and defects that are typically generated.[13–15]

W.-B. Jung, Prof. H.-T. Jung
Department of Chemical and Biomolecular Engineering (BK-21 plus)
National Laboratory for Organic Opto-Electronic Materials/Korea Advanced Institute of Science and Technology
Daejeon 305-701, South Korea
E-mail: heetae@kaist.ac.kr

Dr. H. S. Jeong
Soft Innovative Materials Research Center
Korea Institute of Science and Technology
Eunha-ri San 101, Bongdong-eup, Wanju-gun
Jeollabuk-do 565-905, South Korea

Dr. H.-J. Jeon
Department ent of Nano-structured Materials Research
Korea Advanced Institute of Science and Technology
Daejeon 305-701, South Korea

Dr. Y. H. Kim
Division of Advanced Materials
Korea Research Institute of Chemical Technology (KRICT)
141 Gajeongro, Yuseong, Daejeon 305-600, South Korea

Dr. J. Y. Hwang
Beam Engineering for Advanced Measurements Co
809 South Orlando Ave, Suite I, Winter Park, FL 32789, USA

Dr. J. Y. Hwang, Prof. J.-H. Kim
Department of Electronic Engineering
Hanyang University
Seoul 133-791, South Korea

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In the study described below, we demonstrated that nematic LCs can be efficiently aligned without employing a polymer layer by a process that uses an electrode with a patterned surface. This technique causes ultrafast switching over a large area without the creation of defects. A schematic describing the overall process envisaged for creating line patterned electrodes (indium tin oxide, ITO) with various shapes and angles is illustrated in **Figure 1**a. In this approach, the line pattern on the ITO surface is created by using a simple modification of the conventional secondary sputtering technique[16–18] (see the Supporting Information for conventional secondary sputtering technique, Figure S1). Secondary sputtering lithography is comprised of etching bottom materials to a depth of 10–20 nm by using Ar^+ ion bombardment, followed by resputtering materials on sidewalls of the polymer prepattern to heights of 300–400 nm. In LCD research field, Oleg Lavrentovich group suggested plasma ion beam sputtering on organic/inorganic surface for planar and tilted alignment.[19] Our technique has precisely controlled nanopattern and used irradiated target material as alignment layer and electrode at the same time. Therefore, this technique suggests new method for precise controlling of LC molecules. The pattern generated in this manner has high resolution (≈10 nm) and a high aspect ratio (>10). It should be mentioned that electrodes other than ITO as well as other lithographic techniques can be used if the electrode surface is precisely patterned.

As will be described below, the presence of tilted and folded line patterns on electrode surfaces is important in order to cause perfect alignment of LC molecules without disclinations. The patterned ITO electrode with a tilted line structure that is inclined with respect to substrate surface can be fabricated by using a trapezoid-shaped polystyrene (PS) prepattern that is created by simply controlling the incident angle during the anisotropic etching step. For example, a tetragonal-shaped prepattern for generating a vertical ITO line pattern can be prepared without using anisotropic etching. In contrast, a trapezoid shape for producing a tilted line pattern is fabricated by employing anisotropic etching with an Ar^+ ion incident angle of 40° with respect to the tetragonal shape. Similarly, a folded line pattern can be formed by using a polygonal-shaped prepattern prepared by anisotropic etching with an Ar^+ ion incident angle of 60° (see Figure S3 in the Supporting Information).

In the current study, target materials sputtered on the inclined sidewall of a trapezoid prepattern were prepared by using Ar^+ ion irradiation at 15° relative to a trapezoid-shaped PS prepattern. Because the vertical sidewalls of the trapezoid

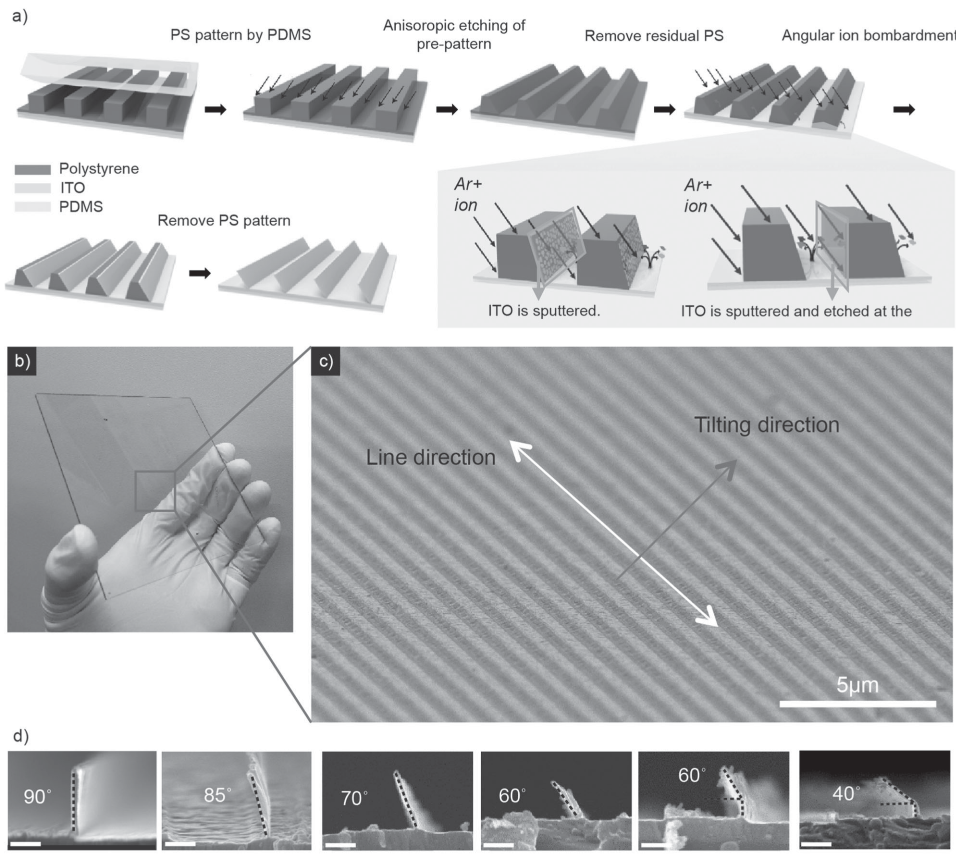

Figure 1. Scheme for fabrication and SEM results. a) Schematic representation of a deformed line pattern. By controlling shapes of prepattern, tilted and folded line patterns are obtained. b) Picture and c) SEM image of large-area tilted line pattern. d) SEM images of different line patterns with each shapes and angles (scale bar is 200 nm).

prepattern are simultaneously sputtered and etched, the ITO line pattern does not remain on the vertical sidewalls. On the other hand, the inclined sidewalls of the trapezoid-shaped prepattern is sputtered but not etched. The resulting ITO line pattern after removal of the PS prepattern is tilted with respect to ITO substrate.

An optical image of a representative ITO electrode surface with a tilted line pattern over a large area (4 in × 4 in) is displayed in Figure 1b. The corresponding scanning electron microscope (SEM) image shows that the 40° inclined ITO lines are highly periodic over the entire substrate surface and that they have a 1 μm pitch, 15 nm thickness, ≈300 nm height and aspect ratio of ≈20 (Figure 1c). Employing this lithographic technique, tilted ITO lines with inclined line angles of 85°, 70°, and 60° can be fabricated (Figure 1d) down to a minimum of ≈60° below which the ITO lines become unstable. Moreover, patterned ITO electrodes with highly periodic vertical and folded lines can be prepared without defects over large areas. The folded ITO lines consist of two parts including a lower region that is perpendicular to the surface and an upper part with angles that depends on the etching time. Folded lines with upper line inclined angles of 60° and 40° were prepared in this investigation.

In this effort, the common nematic LC, 4-cyano-4′-pentylbiphenyl (5CB, Aldrich), was introduced into twisted-nematic (TN) cells containing the patterned ITO electrode (**Figure 2**a). LC cells for electro-optic measurements were prepared by using two substrates possessing ITO line patterns on their inner surfaces, like that used for TN cell construction, where the direction of the ITO patterns on the top and bottom substrates are aligned in a perpendicular fashion. The cell gap was varied in the range of 2–15 μm by using PS beads. To compare display performances, a conventional polyimide (PI) alignment layer, produced by polymer rubbing on a flat electrode, and a surface nanostructured electrode without a polymer alignment layer were prepared (see Figure S7 in the Supporting Information).

To uncover ideal electrode patterns for bringing about uniform alignment without defects, the nature of nematic LCs produced as a function of tilt angle of the patterned ITO lines was determined. It is especially important not to have defects associated with disclination lines, which lead to deterioration of contrast ratios (CR) of devices.[20,21] In the absence of an electric field, TN cells possessing both vertical and tilted ITO line patterns were found to display a uniform white color. This observation indicates that the Mauguin's condition is reached in these TN cells (inset image of Figure 2b). However, when 5 V is applied to a cell with the vertical line pattern, disclination lines (white curved lines, Figure 2b) appear showing that directional alignment of the LCs is not taking place. In contrast, TN

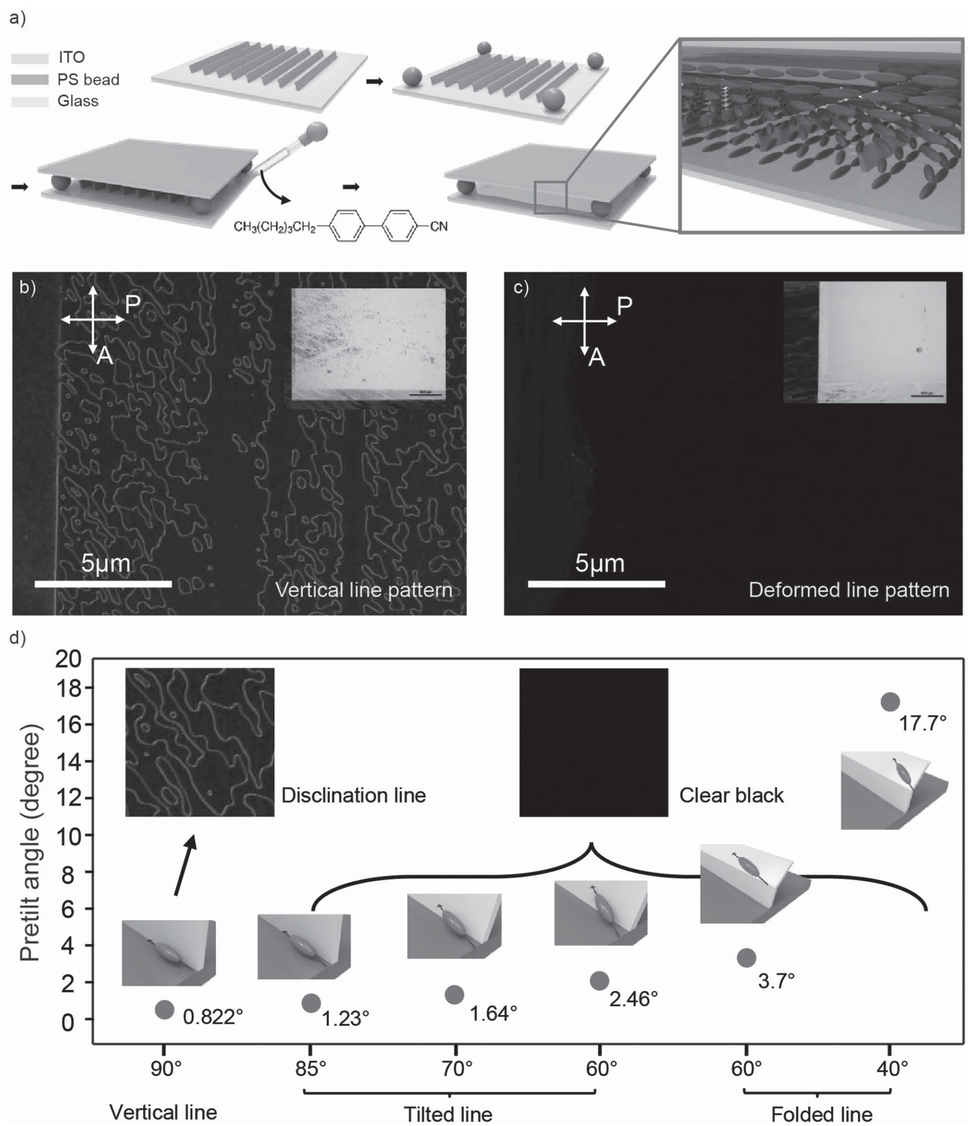

Figure 2. Pretilt angle control results. a) Fabrication of twisted nematic mode (TN mode) cells with tilted line patterns. b) POM images of vertical line pattern and c) tilted line pattern in the voltage-on state. Inset images in (b) and (c) are in the voltage-off state (white). d) Pretilt angle versus angle of line pattern. Inset POM images show the existence of a disclination line in the cell with a vertical line pattern and clear black in cells with tilted line patterns.

cells possessing tilted or folded ITO lines remain completely dark when a voltage is applied, indicating that no disclination lines are generated (Figure 2c). This observation demonstrates that the LC molecules are pretilted under the influence of the patterned ITO electrodes and that the direction of the pretilt angle is determined by the directions of asymmetric ITO line patterns.

Variations of the pretilt angles of 5CB molecules as a function of tilt angles of ITO lines are shown in Figure 2d. The pretilt angles were determined by using the polarizer rotation method.[22] The LC cell gap and line pattern period were 15 µm and 1000 nm. The results show that the pretilt angle of the 5CB molecules can be varied in a controlled manner from 0.822° to 17.7°, depending on the angle of inclination of the tilted and folded ITO lines. This finding indicates that LC molecules adopt out-of-plane configurations in a manner that is determined only by the topology of the electrode surface. When the ITO lines are vertical (not tilted), the pretilt angle of 5CB molecules is 0.822°, which is not large enough to prevent the generation of disclination lines when an electrical field is applied. However, the pretilt angle of 5CB molecules increases in a manner that is proportional to the inclined angle of tilted ITO lines. For example, the respective 5CB pretilt angles are 1.23°, 1.64°, and 2.46°, the latter being the maximum possible value, when the inclined angles are 85°, 70°, and of the 60°. When folded lines are utilized, larger pretilt angles can be generated reaching a maximum value at 17.7°. 5CB containing cells, fabricated using a unidirectional ITO with tilted and folded lines, have a uniform black texture and they do not produce disclination lines in the presence of an electric field (Figure S8, Supporting Information). Therefore, unidirectional, defect free pretilted LCs can be produced by using patterned ITO electrodes. Moreover, the magnitude of the pretilt angle is controlled by the inclination angle of the patterned ITO lines.

The remarkable dependence of the pretilt angle of 5CB molecules on the inclination angle and shape of the patterned ITO lines can be attributed to symmetric breaking of 5CB within the asymmetric lines. The LC director is preferentially aligned along the direction of low roughness so that the total elastic free energy of the LC is minimized.[23–26] Although not entirely clear, the main driving force for generating a pretilt angle of the nematic LCs on tilted and folded line patterns might be associated with the physical anisotropy of ITO lines.[20] In other words, creation of a pretilt angle of the nematic LC molecules minimizes elastic free energy.

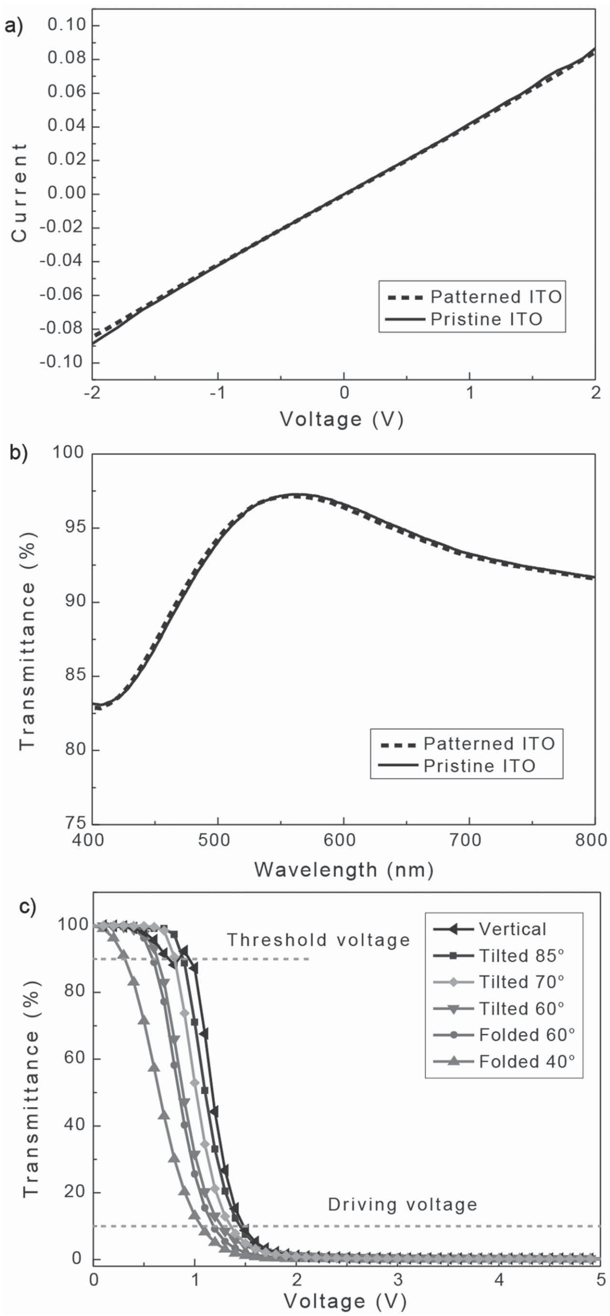

Figure 3. Electro-optic properties of ITO patterns. a) Voltage–current graphs of patterned and pristine ITO glass. b) Wavelength–transmittance graphs of patterned and pristine ITO glass. c) Voltage–transmittance graphs of samples.

It is important to note that pretilt angle control of nematic LC molecules using surface nanostructures on electrode is a new phenomenon. In all previous studies, pretilt angle control was accomplished by using physical and chemical methods such as rubbing and photoalignment.[27,28] In these procedures, which require the use of polymer alignment layers, control of pretilt angles is achieved by changing the length of the polymer side chain, the strength and depth of rubbing, surface hydrophobicity, the curing temperature and film thickness.[29,30] In contrast, in the new approach described above the nature of nanostructures on the ITO electrode surface control the pretilt angle of LC molecules. Consequently, in this system the surface patterned ITO electrode simultaneously functions as an electrode, alignment layer, and pretilt angle controller.

The resolution and aspect ratio of an ITO layer directly govern its electro-optical performance (transmittance, alignment of LCs, and electrical conductivities and electro-optic response). Because of this issue, we were interested in determining if the presence of surface patterns on ITO electrodes influences these properties. The results show that the presence of a unidirectional line-pattern does not deleteriously affect the electrical (**Figure 3**a) and optical properties (Figure 3b) of the ITO. The reason for this is that only a small portion of the ITO layer is lost during creation of walls with heights of a few hundred nanometers and a width of ≈10 nm.

The electro-optical response of TN cells to an electric field is displayed in Figure 3c. Cells with all of the patterned ITO electrodes were found to exhibit typical voltage–transmittance (*V–T*) properties that are characteristics of a normal white mode TN cell. As the applied voltage increases, the LC molecules change from a twisted to a homeotropic state, which results in a decrease of optical transmittance. However, in the case when a vertical line patterned electrode was used, the *V–T* curve contains an inflection point at 1 V (Figure 3c), indicating that behavior of the LC molecules is not controlled in one direction and, instead, they have random pretilt angles. This phenomenon is consistent with the disclination results displayed in Figure 2b,c. On the other hand, the use of unidirectional, angled line patterns on the ITO leads to generation of smooth *V–T* curves as a consequence of the unidirectional pretilt angle of the LC molecules. Also, the threshold, driving and effective switching voltages in these cases are similar to those applied to PI rubbing cell.

Surprisingly, the new approach not only results in perfect alignment of the LC molecules with desirable pretilt angles but it leads to remarkably enhanced response times of the nematic LCs. Response times and corresponding anchoring energies of TN cells created by using PI rubbing; a vertical ITO line pattern, a tilted ITO line pattern with inclined angles of 85°, 70°, and 60°; and a folded ITO line pattern with inclined angles in the upper part of 60° and 40° are given in **Figure 4**. The results demonstrate that both rising and falling times of the 5CB molecules (Figure 4a,b) are strongly influenced by the inclination angle of the tilted and folded line patterns. In particular, the

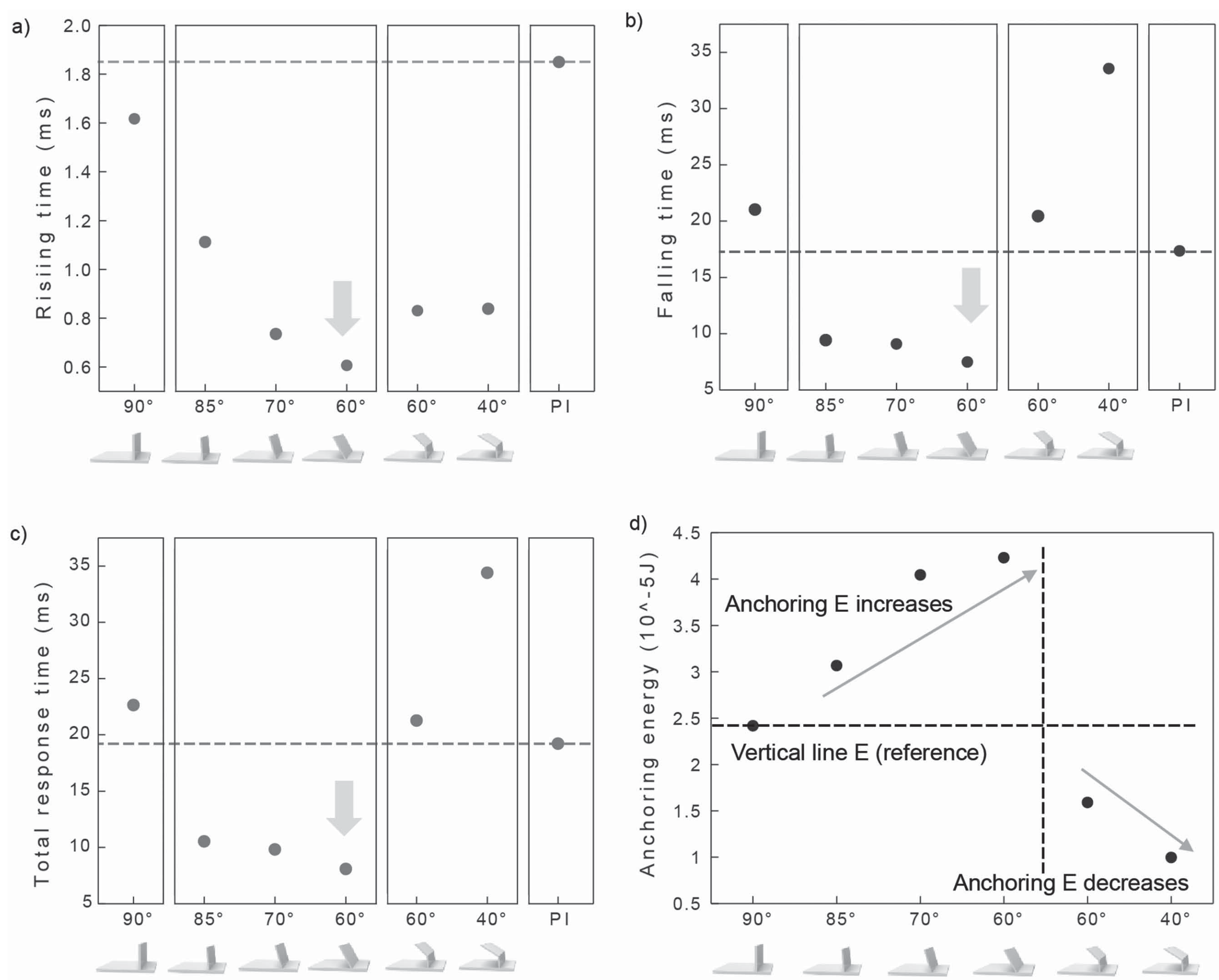

Figure 4. Response time and anchoring energy data. a) Rising and b) falling times of cells with angle line patterns. c) Total response times (sum of rising and falling time). d) Anchoring energy of LCs on cells with line patterns as a function of line angle and shape.

response time is significantly reduced by utilizing the surface tilted line pattern approach.

The influence of inclination angle of the line patterns on the rising time of 5CB molecules in the sandwiched LC cells was evaluated by adjusting the angle from 40° to 90° (Figure 4a). A 5 V bias was applied to each sandwiched LC cell assembled using a patterned ITO substrate in order to modulate the orientation of 5CB molecules from the planar to the vertical direction. The rising time of 5CB molecules was found to decrease when surface line patterns on ITO substrate were used in comparison to utilization of PI rubbing (1.85 ms). In addition, the rising times were observed to markedly decrease from 1.6 to 0.6 ms as the inclination angle is changed from 90° to 60°. However, the rising time does not change from 0.8 ms when the inclined angles of electrodes with folded line patterns are altered. Interestingly, the falling time is dramatically reduced from 21.03 to 7.485 ms as the inclination angle is lowered from 90° to 85°, 70°, and 60°. This enhancement of falling time should be compared to the long falling time of 17.36 ms for LC molecules aligned by using the PI technique. Finally, the falling time is greatly increased when cells with folded line patterns are employed (Figure 4b).

Overall response times, which are sums of rising and falling times, are displayed in Figure 4c. The overall response times of cells with tilted line patterns are faster than those of the cells formed by using PI (19.21 ms), and cells with vertical (22.647 ms) and folded line patterns (21.27 ms). It is significant that the overall response time (8.091 ms) of the cell with a 60° tilted line pattern is approximately 2.5 times faster than that of cell created by PI rubbing. It is important to mention that much faster response times (approximately hundred microseconds) can be attained if nematic LC molecules with faster response times are utilized in place of 5CB. This can be easily accomplished because the switching time of nematic LC molecules in present LC displays are ≈5 ms.[31] Also, a possibility exists that response times can be improved further if dimension features such as line spacing is optimized.

Analysis of the response time results clearly shows that the falling response time is a more important factor determining the overall response times of each substrate. Anchoring energies strongly affect falling times because nematic LC molecules return to planar states more readily when anchoring stronger.[32] In other words, strong anchoring energies cause fast falling times of LC molecules. To gain an understanding about whether or not they govern the response time behavior of the surface patterned ITOs, polar anchoring energies of the cells with different pattern geometries were determined (Figure 4d). To accomplish this, an ECB mode cell with 15 µm gap was fabricated and anchoring energies were measured by using the high field method.[33] The anchoring energy of the cell with the vertical line pattern was found to be 2.417×10^{-5} J m^{-2}. Furthermore, cells with tilted line patterns exhibit much higher anchoring energies than those with folded line patterns and their anchoring energies increase as the tilt angle increases. A maximum anchoring energy of 4.228×10^{-5} J m^{-2} was found for the 60° tilted ITO. However, anchoring energies of cells containing folded line patterns were observed to decrease as the folded line angle increases, and this phenomenon results in relatively slow response times.

In the investigation described above, a new design strategy was explored for creating polymer-free and ultrafast-responsive nematic LCs. The results of an effort exploring this approach show that these goals can be accomplished by employing surface nanostructured electrodes. Specifically, ITOs line patterned with tilted lines of nanoscale widths (10 nm) and high aspect ratios (≈40) can be used to produce perfectly aligned LC molecules that do not have disclinations and that undergo much faster switching than cells created by PI rubbing. Consequently, the surface patterned ITOs simultaneously play three roles including electrodes, pretilt angle controllers and perfect alignment layers. Furthermore, this technique can be applied on any other materials including ITO alternative materials such as ZnO, so it has versatility of material choice. Also, we expected that other LCD modes such as IPS and FFS without polymer layer can be achieved by proposed technique. We believe that the results of this effort will be of great interest to nanotechnology and LC researchers, and that they will demonstrate the potentially general concept of "surface nanostructure based perfect alignment and ultrafast response".

Experimental Section

Fabrication of ITO Line Pattern for Alignment Layer: To fabricate ITO line pattern, polystyrene (8 wt% of PS (molecular weight = 18000 g mol^{-1}) in anhydrous toluene is spin-coated on the ITO glass. Then, the poly (dimethylsiloxane) (PDMS) mold was placed on the PS film and heated above the glass transition temperature in a vacuum oven to move the PS polymer into the mold through capillary force. The mold is strip patterned with a 1:1 width to spacing ratio (w, s = 500 nm, depth = 600 nm). To obtain anisotropic PS patterns, it is etched by using ion milling in certain angles (angles = 45° and 60°). In order to form line pattern on only the ion milling etched side of the PS pattern, the exposed ITO plane is etched by ion milling at an angle of 20°. During this process, the ITO line pattern forms only on one side of PS pattern. Finally, the PS film is removed by using reactive ion etching with oxygen gas (100 sccm).

Liquid-Crystal Cell Fabrication: To observe electric-optic properties of LCs, two kinds of LC cells were fabricated. One is in the TN mode where strip pattern directions of two substrates are perpendicular, and the other is an electrically controllable birefringence (ECB) cell where the substrates are parallel. Cells having the TN mode are used to measure response times and those with the ECB mode are used for observing pretilt angles. The threshold, driving and effective switching voltages used are similar to those applied to the PI rubbing cell. To compare with PI rubbed cell, we used Nissan SE 7294 which is used in the study of LCD research and is commercially available PI precursor.

The polarizer rotation method using a cell with a gap of 5 µm and laser beam at 633 nm wavelength was employed to determine tilt angles. The cell is located between the polarizer and analyzer, and the polarizer and analyzer are rotated during transmittance measurements giving a pretilt angle that corresponds to a transmittance minimum point. For the measurements of the electro-optical properties (voltage–transmittance and response time), the cell gap was maintained at 15 µm. To investigate effects of "bidirectional" and "unidirectional ITO lines on pretilt angles, TN cells with controlled cell gap of 5 µm, were prepared and then filled with 5CB. The electro-optical texture of the LCs upon application of an electric field was recorded.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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