Photoaligned Vertical Aligned Nematic Mode in Liquid Crystals

Viktor KONOVALOV^{*}, Vladimir CHIGRINOV[†], Hoi Sing KWOK, Hirokazu TAKADA¹ and Haruyoshi TAKATSU¹

The Hong Kong University of Science and Technology, Clear Water Bay, Kowloon, Hong Kong, R.O.C. ¹Dainippon Ink & Chemicals Inc., 631, Sakado, Sakura, Chiba 285-8668, Japan

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The photoaligned vertical aligned nematic (VAN) mode in liquid crystals (LC) was developed using the photosensitive polyimide (PI) JALC 2021-R2, illuminated by obliquely incident nonpolarized light. It has been found that the combination of PI with water soluble sulfonic azo-dye (SD) yields a perfect electro-optical performance of a liquid crystal in the VAN mode. The polar and the azimuthal anchoring strengths of LC in VAN mode were measured both for rubbed and photoaligned surfaces. Increasing the illumination time results in the exponential growth of the contrast ratio in the VAN LC cell. Analysis of anchoring strength and electrooptic behaviour of the photo-aligned LC cells confirm that the mechanism of photo-alignment in our materials includes not only the photodegradation, but mostly the reorientation of both SD-1 and PI molecules to get the average absorption oscillator almost parallel to the direction of the obliquely incident UV light. [DOI: 10.1143/JJAP.43.261]

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1. Introduction

The "rubbing" technique of liquid crystal (LC) aligning in liquid crystal display (LCD) cells is widely used and provides the perfect electrooptic performance of LC cells over a large area of the surface. However the corresponding impurities, electrostatic charges and mechanical damage may result in the deterioration of the quality of LC switching, particularly when active matrix (AM) elements are used for the purpose.¹⁾ Therefore, an alternative photoaligment technique has been proposed to provide a homogeneous and oblique LC alignment.²⁻⁴⁾ There are several mechanisms involved in the photoaligment phenomena: reversible cis-trans isomerization, photodimerization or crosslinking and photodegradation.⁵⁾ The photosensitive polyimide (PI) is known to produce a homogeneous or tilted alignment of the LC director perpendicular to the UVlight polarization vector as a result of an irreversible chemical reaction of photodegradation.⁶⁻¹²⁾ Linearly polarized UV light would preferentially break the PI backbones lying parallel to the UV polarization at the surface, thus yielding the photoalignment effect. The analysis of both UVvisible and IR dichroism in the polarized UV exposed light^{8–11} shows that the alignment in PI is produced by the photoreaction of the O=C-N bonds of the imide rings. The infrared-visible sum-frequency generation (SFG) vibration spectroscopy of rubbed and photoaligned PI reveals that UV illumination breaks the C=O and C-C bonds in the PI main chains.¹²⁾ The resulting surface anisotropy that arises from the preferential bond breaking by a linearly polarized light parallel or perpendicular to the PI backbones is small. Consequently, the azimuthal anchoring energy of the LC layer attached to this surface would also be small.¹²⁾

Vertical aligned nematic (VAN)-LCD became the most popular LCD because of its high contrast ratio and wide viewing angle.¹⁾ VAN-LCD can be aligned even with a slantwise unpolarized light that makes this technique very promising for mass production application.^{13–19)} At first, the procedure of obtaining VAN mode by a slantwise nonpolarized UV light using azobenzene side chain polymer with a subsequent annealing was suggested by Furumi et al.¹³⁾ The order parameter increases after the annealing procedure, however, it remains very small.¹³⁾ A similar irradiation method using obliquely incident nonpolarized light enabled obtaining slightly pretilted homeotropic alignment using polyimide films. The photodegradation of the commercially available polyimide aligning materials^{15,16)} or crosslinking of photopolymers^{14,18,19} during the exposure of obliquely incident unpolarized light is believed to be the main process, which is responsible for the alignment in this case. Unfortunately, the former process results in a decrease of the voltage holding ratio (VHR), which is a very important parameter especially for thin film transistor (TFT)-LCD fabrication. Moreover, notwithstanding the efforts the VAN-LCDs prepared by photoaligning technique have not yet reached an appropriate quality (response time, contrast ratio) in comparison with conventional LCDs prepared by rubbing technology.¹³⁻¹⁹⁾ Certain success has been achieved in the development of multidomain LCDs, utilizing homeotropic orientation. A four domain hybrid twisted nematic (HTN) LC cell was fabricated with a small pretilt angle on one substrate and high pretilt angle on the other,²⁰⁾ showing uniform transmission characteristics for wide viewing angles up to 40° . Two-domain²¹⁾ and fourdomain²²⁾ photo-aligned VAN-LCDs were also fabricated using polymeric liquid crystals-linear photopolymerized (LCP-LPP) films.

We suggest that the application of modern commercial VAN LC materials with high resistivity to UV light in combination with some newly developed photosensitive materials is the key factor in overcoming the abovementioned drawbacks.²³⁾ Our study is devoted to the development of the photo-aligned VAN mode in liquid crystals having a high contrast ratio and fast response time together with a high VHR value. The effect of UV light exposure time on the anchoring strength, pretilt angle and VAN electrooptic response will also be considered.

2. Experimental

In the experiment we used unpolarized UV light to align the LC molecules on the surfaces. The incidence angle of the activating UV light and the exposure time was varied. The

^{*}On leave from the Institute of Applied Physics Problems, Kurchatova 7, Minsk 220064, Belarus.

[†]E-mail address: eechigr@ust.hk



Fig. 1. Nonpolarized UV-illumination of the photosensitive SD layer at the angle θ . The chemical formula of photosensitive derivative of sodium salt of benzidine -3,3'-disulfonic acid (SD) is shown below.

incidence light angle θ was defined as the deviation of the incident light from the normal to the surface and is shown in Fig. 1.

The testing cells were assembled with the thickness of the LC layer of $3.5 \pm 0, 1 \,\mu\text{m}$ and with opposite directions of the substrate illumination. MLC-6609 (Merck) with a negative dielectric anisotropy and without a chiral dopant was used as the LC mixture. To measure the azimuthal anchoring strength the cells with a 6µm LC thickness layer were made. A collimated UV light beam source with the wavelength of 365 nm and power of 12-14 mW/sm² was chosen for substrate illumination. The substrates were coated with JALS-2021-R2 (PI) (Japan Synthetic Rubber Co., Ltd) as aligning layers with a thickness of 30 nm by a spin coating method at 3000 rpm. After this the PI films were cured at 180°C for 1.5 h. It was shown earlier that the derivative of the sodium salt of benzidine -3,3'-disulfonic acid (SD) can be an effective reagent for the homogeneous photoalignment of the LC layer.^{23,24)} SD was used as a dopant to JALS 2021-R2 for varying the pretilt angle. The pretilt angle was measured by a crystal rotation method¹) changing the retardation of the LC layer between -20° and $+20^{\circ}$ and the subsequent fitting with an accuracy of $\pm 0, 1^{\circ}$. The contrast ratio and response time were measured at $\lambda = 632 \text{ nm}$ (He–Ne laser) and registered by HP "Infinum" oscilloscope at the square driving voltage $\pm 5 V$ with the frequency of 1 kHz. The actual twist angle in the cells was measured by the spectroscopic ellipsometry method.²⁵)

3. Results and Discussion

The uniformity of the alignment of VAN mode with unpolarized light depends on the exposure time of the UV light and the exposure angle (Fig. 1). At first the substrates with deposited aligning JALS 2021-R2 films were illuminated at the exposure angles varied from 10° to 80° for 20 min and their alignment properties were estimated by the electrooptic response of the corresponding assembled LC cells. The increase in the incident light angle up to 60° results in the faster response time of VAN mode (Fig. 2). This means that the uniformity of the alignment is also improving. At the incidence angle higher than this value the response time drastically increases. This takes place probably due of the lower UV light energy that reaches the



Fig. 2. Switching "on" time of VAN mode in LC cell with PI aligning layer versus the angle of UV-light illumination. Exposure time is 20 min.



Fig. 3. Dynamics of switching "on" process in VAN mode is presented. On the left the chaotic structure appeared immediately after application of ± 6 V driving voltage. On the right, the photograph of the same structure after 1 s is shown. The pitch of dark lines in the patch is 8 μ m. The direction of the illumination is from left to right and coincides with one of the axes of the crossed polarizers.

photoaligning substrate. The cell with the substrates illuminated at 60° for 20 min has a rather low pretilt angle of 0.18°. In this case the pretilt angle is not high enough to switch the LC molecules to the plane of the substrate in a unidirectional way in an electric field, which was confirmed by the direct observation in a polarizing microscope (Fig. 3). At the beginning of this process, a numerous small areas with different direction of switching appeared and only after a sufficiently long period of time, the uniform "on" state occurred. This phenomenon is known as "umbrella" defects of the VAN texture in an electric field, which occurred when the director pretilt from the pure homeotropic state is not sufficiently high.¹⁾ The increase in the exposure time to 50 min and then to 60 min at 60° illumination angle enlarges the LC pretilt angle up to 0.3° . In this case the VAN mode response time $\tau_{\rm res} = (\tau_{\rm on} + \tau_{\rm off})$ decreases to 36.4 ms and 22.9 ms accordingly. A more uniform alignment results in the faster response time, as the switching energy of the applied electric field is the most effective in this case.

It is well known that LC polar anchoring energy strongly effects the switching "off" time in LC electrooptical modes.¹⁾ The fact that the VAN mode switching "off" time remains the same for all the tested cells ($\tau_{off} \approx 8-8, 2 \text{ ms}$) can be understood, if we suggest that the polar part of the anchoring energy remains almost the same. The estimation of the polar anchoring energy from the measured transmission-voltage curve (TVC) for the three VAN cells prepared by photoalignment (Fig. 4) was carried out by



Fig. 4. Transmission-voltage curve for: 1 — cell with the rubbed substrates covered by JALS 2021-R2; 2 — cell with illuminated substrates covered by JALS 2021-R2 having 1 wt % of SD; 3 — the cell with illuminated surfaces covered by the pure JALS 2021-R2. The values of the polar anchoring energy W_{θ} , estimated using our MOUSE-LCD software were as follows: 1 — $W_{\theta} = 2.2 \times 10^{-5} \text{ J/m}^2$, 2 — $W_{\theta} = 3 \times 10^{-5} \text{ J/m}^2$ and 3 — $W_{\theta} = 2.9 \times 10^{-5} \text{ J/m}^2$.

modeling of the LC director deformation with a finite polar energy using MOUSE-LCD software.²⁶⁾ The values of the polar anchoring energy were as follows (Fig. 4): 1—2.2× 10^{-5} J/m², 2—3×10⁻⁵ J/m² and 3—2.9×10⁻⁵ J/m², which are almost the same and rather low. This probably means that the polar part of the anchoring energy is not very important and the most significant contribution to the decrease of switching "on" time τ_{on} of VAN mode is made by the azimuthal part of the anchoring energy that increases during the exposure time. If the azimuthal anchoring energy is sufficiently high the LC director can hardly deviate from the incidence plane of UV light and the switching properties of VAN mode are improved.

From this point of view it is interesting to compare the "effective" contrast ratio in the "on" state of the VAN mode between the two positions of the cell in the crossed polarizers at 0° and 45° with respect to the plane of incidence of the slantwise activating UV light, where the LC director deformation takes place (Fig. 5). If the LC director is uniformly aligned by UV light in the plane of exposure, then the perfect dark LC cell can be observed between the crossed polarizers in the "on" state of the VAN mode when one of the polarizers is parallel to the plane (Fig. 5, left), while the rotation of the polarizers at the angle of 45° with

respect to the plane will result in the corresponding "bright" state in this case (Fig. 5, right). The higher the "effective" contrast ratio the smaller the deviation of the LC director from the exposure plane during switching in an electric field and consequently the smaller the response time of the VAN mode.

Indeed the increase in the exposure time from 20 min to 50 min and than to 60 min leads to the growth of the "effective" contrast ratio from 5.5:1 to 20.4:1 and finally to 96:1. It should be kept in mind that this "effective" contrast ratio is also defined by the presence of defects in the aligning film that can drop its value. It should be mentioned that the measured value of the contrast ratio between "off" and "on" states in VAN mode (the polarizer angle is 45° with respect to the exposure plane) exceeds 1000:1 in the monochromatic light (He–Ne laser, λ =632.8 nm).

Unfortunately the oblique illumination of a pure PI layer by unpolarized UV light proceeds through the photodegradation of the main PI chains^{16,17)} and results in a low VHR value of the VAN mode. The measured values of VHR for the cells irradiated for 50 min and 60 min were markedly small, i.e., 71.3% and 63% accordingly. The VHR measurements¹⁾ were conducted by a standard technique, when the voltage pulse of V_0 =5 V was applied during 64 µs and the voltage V(t) on the LC cell dropped down during τ =16 µs after the pulse was switched off at *t*=0.

To provide the pretilt angle we doped PI with the photochemical stable azodye SD (Fig. 6). The photoaligning of azo-dye molecules takes place due to the pure reorientation of the molecular absorbtion oscillators perpendicular to the UV-light polarization.²⁴⁾ The compositions of different concentrations of SD in solution of JALS 2021-R2 (PI) were prepared and the photoaligning films were treated as described above. It has been found that the increase of the illumination time results in higher pretilt angles up to 0.53° for the 1 wt % of SD in PI, which is better than those for the pure PI layers (0,3°) under the same conditions of UV exposure. It should be mentioned that within the range of accuracy (\pm 0.1°) the measured pretilt angle does not depend on the concentration of SD and saturates at a 60 min exposure time.

In contrast to the changing of pretilt angle, the dependence of "effective" contrast from the concentration of SD in PI at the same illumination time is more pronounced. The "effective" contrast ratio in the "on" state of VAN mode was increased up to 171 at the exposure time of 60 min



Fig. 5. Photo-aligned VAN LC cell exposed for 20 min in the "on" state (5 V) between the two crossed polarizers. The arrows in the pictures show the direction of UV light exposure. The arrows under the photographs show the directions of crossed polarizers. "Effective" contrast ratio is 5.5:1. The pitch of the dark lines in the patch is $8 \mu m$.



Fig. 6. Pretilt angle of the compositions with different concentration of SD in PI in versus the illumination time of UV light with the power of 12 mW/sm^2 .



Fig. 7. Dependence of the "effective" contrast ratio on the concentration of SD in PI for different values of exposure time.



Fig. 8. VAN mode response time versus the exposure time for various concentrations of SD in PI and various exposure times. The discrete line is the response time of the VAN cell obtained by rubbing.

 (12 mW/sm^2) for the films based on 1% of SD in PI (Fig. 7).

The VAN mode response rate was strongly dependent on the exposure time. The fastest response time is observed when the exposure period reaches 60 min (Fig. 8). Probably the value of pretilt angle of 0.53° is sufficient for the reliable dynamic switching of the VAN mode. Figure 8 provides the VAN mode response time for the PI doped with SD in the test cell.

The "effective" contrast exponentially increases with exposure time, which means, that in our case the azimuthal energy for the photoaligned VAN LC cell is becoming considerably higher (Fig. 9).

To compare the electrooptic properties of the VAN mode



Fig. 9. Dependence of "effective" contrast on the exposure time for the VAN cells based on photo-aligned layer with 1 wt % SD in PI.

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prepared by photoalignment and conventional rubbing, the test cell was prepared. The aligning film of pure JALS 2021-R2 was deposited by spin coating on the glass plates covered with indium tin oxide (ITO) and prepared by the thermal treatment described above. Instead of illumination the rubbing technique was used to align the LC molecules to the homeotropic (VAN) state with some pretilt angle. The measured value of the pretilt angle from the vertical LC director state was approximately 1.4°, which is higher than in the photoaligned VAN cell (0.53°) . However the response time of the photoaligned and conventional VAN LCD was almost the same $\tau_{on} + \tau_{off} = 7.8 \text{ ms} + 9 \text{ ms}$ and $\tau_{on} + \tau_{off} =$ $8.1 \,\mathrm{ms} + 8.9 \,\mathrm{ms}$ respectively. This probably means that SD as a dopant in PI further increased the azimuthal anchoring strength in photo-aligned cells. Moreover this increase takes place not only due to the photodegradation of PI in the UV light, but also due to the pure reorientation of both SD and PI molecules in such a manner that their average absorption oscillator becomes perpendicular to the polarization of the activating UV light.

To measure the electrooptic response a square wave driving voltage with the amplitude of ± 5 V and the duration of 200 ms was applied (bottom part of each part in Fig. 10) with the following delay of 300 ms. The VAN LC cells were placed at 45° (middle curves) and 0° relative to the cross polarizers (upper lines, we mark them with a cross "x"). Similar behaviour has been observed for the VAN mode prepared by both photo-aligning and rubbing techniques.

We obtain the high values of VHR for the photo-aligned VAN-LCD using SD/PI composition of 94–96% (in contrast to the conventional rubbed VAN-LCD with VHR=88%). The contrast ratio for both cells between "on" and "off" states was more then 1000:1 ($\lambda = 632.8$ nm) at normally



Fig. 10. Electrooptic response of the photo-aligned VAN mode (on the top) and VAN mode prepared by conventional rubbing (bottom).



Fig. 11. Orientation of LC director for the LC mixture with negative dielectric permittivity (MLC 6609) in VAN mode under electric fields. The modeling was performed by MOUSE-LCD software.²⁶⁾ We used the parameters of MLC 6609 in our calculations, cell thickness was $d=3.5\mu$, and polar anchoring was $W_{\theta}=3\times10^{-5} \text{ J/m}^2$. Pretilt angles on the boundaries were 0.5° from the normal direction, and the twist angle was 85°.

incident light, as we mentioned before.

The properties of the LCD strongly depend on the interaction between LC molecules and the surface. The surface anchoring for a nonpolar LC is generally characterized by two parts: an azimuthal anchoring component which governs the anchoring with respect to in-plane twist of the NLC and a polar anchoring term which controls the out-of-plane tilt. The torque balance equation helps to estimate the azimuthal anchoring strength W_{φ}^{27} as the actual twist angle should be less than that prepared in the experiment:

$$W_{\varphi} = 2K_{22}\Delta\varphi/d\sin 2(\Phi - \Delta\varphi) \tag{1}$$

where Φ , $\Delta \varphi$, K_{22} , and d are the twist angle prepared in the experiment, the actually measured twist angle, the twist elastic constant of a liquid crystal, and the cell gap, respectively.

In the initial state LC molecules in the VAN mode aligned perpendicular to the surfaces with some small ($\approx 0.5-1^{\circ}$) pretilt angle. Under an electric field the LC director realigned to the in-plane of substrates (Fig. 11). The LC director on the substrates also tends to the in-plane state, as the polar anchoring energy is not sufficiently strong (Fig. 11).

If the projections of LC directors on the two substrates, obtained by rubbing or UV illumination, are almost perpendicular to each other, the quasi TN structure can be reached. As seen from Fig. 11 for a sufficiently high voltage (>6 V in our case), the LC director mostly becomes almost parallel to the substrates and the apparent LC configuration is very close to a pure twist LC cell everywhere, except at the boundary regions. It has been found that the pretilt angle on the surface is strongly dependent on the driving voltage and reaches 18° from the normal at 6 V.

To measure the azimuthal anchoring coefficients W_{φ} , a number of the VAN LC cells were assembled with twist angle equals to $\Phi \approx 85^{\circ}$ by the photoalignment method to avoid the regions with different twisting and then thoroughly controlled using a microscope. The actual twist angle $\Delta\varphi$ defined by the value of the azimuthal anchoring energy W_{φ} (1) was measured by spectroscopic ellipsometry at different driving voltages with an accuracy of $\pm 0.5^{\circ}$ and was almost independent of voltage in the range 4–11 V (Fig. 12). The relative azimuthal anchoring strength $W_{\varphi}d/K_{22}$ calculated from eq. (1) using $d = 3.5\mu$ and $K_{22}=0.6 \times 10^{-6}$ dyne for the



Fig. 12. Actual twist angle of the VAN mode in LC cell versus the applied voltage.



Fig. 13. Relative azimuthal anchoring strength $W_{\varphi}d/K_2$ for the photoaligned VAN mode as a function of exposure time. The solid line presents the data, when the LC pretilt on the substrates in VAN mode was obtained by a conventional rubbing. We have taken the values of $d=3.5 \mu$ and $K_{22}=0.6 \times 10^{-6}$ dyne.

quasi TN cells in "on" state of the VAN mode as a function of the UV light exposure time is presented in Fig. 13.

The values of $W_{\varphi}d/K_2$ are found to be within the range of 4–43 (Fig. 13) and correspond to a rather strong azimuthal anchoring strength.¹⁾ After 40 min of illumination, they even exceed these ones for the VAN cell with rubbed PI surfaces. Thus, the addition of the azodye SD to PI leads to the higher values of the azimuthal anchoring strength and a more appropriate electrooptic response of the VAN mode in general. We can see that the higher azimuth anchoring provides the larger values of the "effective" contrast



Fig. 14. "Effective" contrast of photo-aligned VAN mode versus the relative azimuthal anchoring strength $W_{\phi}d/K_{22}$. We have taken the values of $d=3.5\mu$ and $K_{22}=0.6\times10^{-6}$ dyne.

(Fig. 14). The LC director cannot easily get out from the plane of the oblique quasi-homeotropic alignment, which provides perfect switching of the VAN mode.

4. Conclusions

Photo-aligned VAN mode was developed using the commercially available and UV-light-stable-aligning polyimide (PI) material JALC 2021-R2. To provide the photoaligning of liquid crystal, the polyimide was illuminated by oblique nonpolarized UV light. The electrooptic performance of the developed VAN mode strongly depends on exposure time and illumination angle of the UV light. It has been shown that doping the PI with water soluble sulfonic azodye (SD) yields even more reliable and perfect electrooptical performance of the VAN mode. This PI/SD mixture, used for the preparation of the photoaligning layer, provides the same parameters of VAN mode response as the conventional rubbing technique and maintains a high VHR ratio. The polar and azimuthal anchoring strength were estimated for both rubbed and photoaligned surfaces of the VAN LC cell, which exponentially increase with the exposure time of the UV light. For a certain value of the exposure time the anchoring strength of the photoaligned LC cells reaches even higher values than those obtained in the conventional rubbing method. The electrooptic response of the photoaligned VAN mode and VAN mode obtained by rubbing are very similar despite a certain difference in the LC pretilt angles on the substrate. We believe that the photo-alignment mechanism in our materials includes not only photodegradation, but mostly the pure reorientation of PI/SD average molecular absorption oscillators almost parallel to the direction of the obliquely incident UV light.

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