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## Four-domain twisted nematic structure with enhanced liquid crystal alignment stability and fast response time

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In this paper, we propose a stable four-domain twisted nematic structure with wide viewing angle characteristics and a fast response time. A stable structure was realized by introducing a high pre-tilt angle approach incorporating stacked planar and vertical alignment layers. The response time and thermal stability of the proposed device were dramatically improved by the addition of UV-curable reactive mesogen (RM) mixed with a vertical alignment material. Polymerized RMs along the liquid crystal (LC) director increased the polar anchoring energy and reduced back-flow phenomena. A wide viewing angle and fast response time characteristics were achieved over all gray levels. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4733998>]

### I. INTRODUCTION

Liquid crystal displays (LCDs) are extensively studied and used for a wide range of display applications including mobile phones, monitors, televisions, and other information technologies because of their high image quality and low power consumption. The display performance characteristics of LCDs such as response time, transmittance, and viewing angle depend on the collective behavior of LC molecules, which is determined by the initial LC alignment layer and the external electric field. Therefore, various combinations of LC alignments and electrode structures have been proposed for better electro-optic performances including twisted nematic (TN),<sup>1</sup> in-plane switching (IPS),<sup>2</sup> and patterned vertical alignment (PVA).<sup>3</sup> Among these options, the TN mode is the most commonly used in LCDs due to its simple fabrication process, fast response time, and high transmittance.<sup>1</sup> However, it is difficult to obtain symmetric viewing angle characteristics when utilizing TN because the optic axis in the midplane of the cell is uniformly tilted in one direction.

Four-domain TN (4-D TN) structures have been extensively studied to overcome these asymmetric optical characteristics. The major hurdle to achieving 4-D TN structures is the difficulty of producing multi-domain LC twist alignment within a single pixel. Numerous techniques have been developed including multiple rubbing, multiple UV exposure, and multiple deposition of SiO<sub>2</sub> among other approaches.<sup>4-8</sup> Since most of these techniques require a chiral dopant to stabilize the 4-D TN structure, there can be only one twist sense in a pixel. Thus, while a 4-D TN structure can be achieved using the techniques described above, current fabrication methods are cumbersome and are not readily applicable for mass production.

Recently, Chen *et al.*<sup>5</sup> suggested a relatively simple 4-D TN structure consisting of two left-handed and two right-handed twisted subpixels. Further, since a dopant is not

utilized, two rubbing processes are required at each substrate. However, this technique requires a high pretilt angle to stabilize the 4-D TN structure at 0V, which results in degraded optical performance in terms of the transmittance and response time. Moreover, it is difficult to generate a high pretilt angle using the conventional alignment layers applied in mass production.

In the present study, we fabricated a stable 4-D TN structure at room temperature by a stacked alignment method that achieves high pretilt angles using conventional alignment layers. Secondly, we obtained enhanced thermal stability and response time of the 4-D TN structure by introducing photo-curable monomers in the alignment layers.

### II. EXPERIMENTAL

In order to control the pre-tilt angle, we relied on stacking a vertical alignment layer on a planar alignment layer, as reported previously.<sup>9</sup> The pre-tilt angle was controlled by varying the thickness of the vertical alignment layer on the planar alignment layer. We used SE7492 (Nissan Chemical Industries, Japan) and AL60101 (Japan Synthetic Rubber, Japan) polyimides (PIs) for the planar and vertical alignment layers, respectively. The detailed experimental conditions were described previously.<sup>9</sup> In this experiment, for controlling the pretilt angle, vertical alignment material solution (AL60101) was diluted with 1-3 wt. % to their own solvents which consist of methyl-pyrrolidone, butyrolactone, and butoxyethanol.

In order to produce the 4-D TN structure, reverse rubbing processes were carried out on stacked alignment layers with rubbing masks (SUS, 30 μm thickness and 100 μm spacing). The rubbed glasses were assembled such that the rubbing directions of each glass were perpendicular to each other for a 90° twist structure, as shown in Fig. 1(a). As a result, the sub-pixels consisted of two left-handed and two right-handed sub-pixels (Fig. 1(b)). The cell was made with a 5 μm thick cell gap and filled with nematic LC MLC-6875

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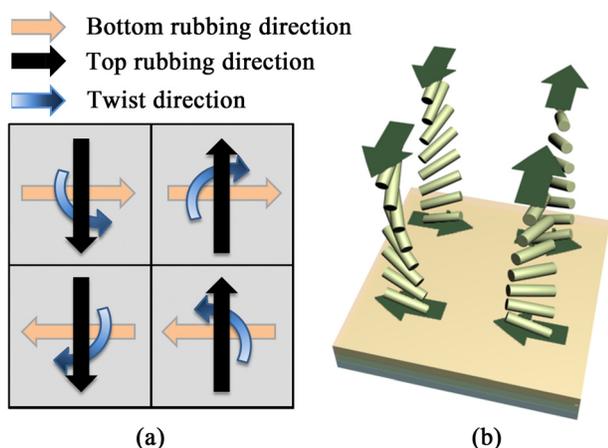


FIG. 1. (a) Rubbing and twist directions for each sub-pixel of the 4-D TN structure. (b) Schematic view of the liquid crystal molecule configuration of each sub-pixel.

( $\Delta n = 0.1114$ ,  $\Delta \epsilon = 7.8$ , Merck Co.), which has positive dielectric anisotropy. The pre-tilt angles were measured using the polarizer rotation method with single domain cells.<sup>10</sup>

To enhance the thermal stability and increase the response time characteristics, we mixed 1 wt. % UV curable reactive mesogen (RM) monomers with the each diluted vertical alignment materials. The mixture of diluted vertical alignment materials and RMs were spin-coated on substrates and cured at 180 °C for 1 h. After reverse rubbing process, two substrates were assembled and LC (MLC-6875) was injected in the cell. For polymerization of RM monomers, we expose the UV light for 30 min under applied voltage (10 V). Under the applied electric field, RM monomers were aligned along the LC molecules due to the liquid crystalline property of RM and these aligned RM monomers were polymerized by the UV curing process.<sup>11</sup>

### III. RESULTS AND DISCUSSION

We first fabricated 4-D TN LCDs with different vertical alignment layer thicknesses (i.e., different pre-tilt angles) without RM mixing. Figure 2 shows polarizing optical microscope images of each 4-D TN LCD obtained with various applied voltages, while Figure 2(a) shows the 4-D TN structure with a pre-tilt angle of 4°, which was produced by planar PI only. At 10 V, the LC molecules were vertically aligned to the surfaces due to the positive dielectric anisotropy of LC. With decreasing voltage, the tilt angle of the LC molecules decreases and generated the 4-D TN structure. However, the 4-D TN structure started to collapse at 1.2 V and was a mono-domain at 0 V. According to a previous report, the stability condition of a 4-D TN structure can be given by  $F_s/F_d \sim \theta^2 L/d\pi \geq 1$ , where  $F_s$  is the splay distortion energy of two subpixels,  $F_d$  is the twist disclination energy which is proportional to  $\zeta$ ,<sup>5</sup> and  $L$ ,  $\theta$ ,  $d$ , and  $\zeta$  are the subpixel size, pretilt angle, sample thickness, and correlation length, respectively. Since the 4-D TN structure has no chiral dopant, any two subpixels should have different handedness. Therefore, disclination lines are formed at the boundaries between the subpixels in order to maintain the stable twisted

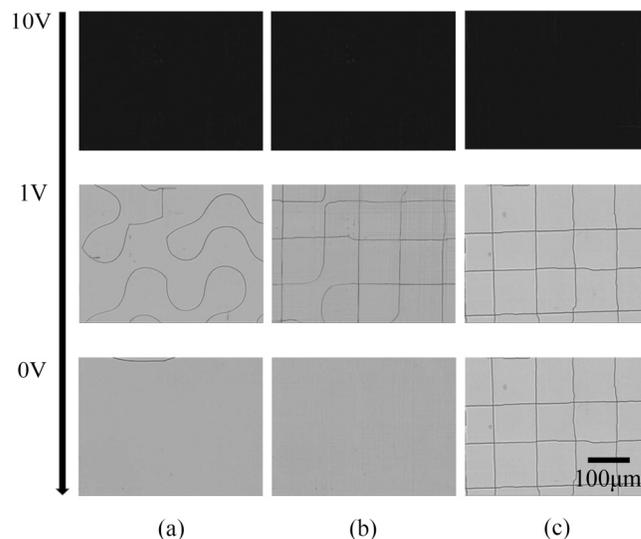


FIG. 2. Images of the 4-D TN structure at pretilt angles of (a) 4°, (b) 7°, and (c) 18°.

structure at each domain. However, if the splay distortion energy is smaller than the twist disclination energy (i.e., the splay distortion is more stable), the disclination lines would disappear and as a result, the cells would become mono-domain. Therefore, for stable 4-D TN structures, a large splay distortion energy is required with a high pretilt angle. In this study, the sample with a pre-tilt angle of 7° exhibited similar behavior, as shown in Fig. 2(b), but started to collapse after the voltage was decreased to 1.0 V. However, we were able to achieve a stable 4-D TN structure over the entire voltage range with a high pre-tilt angle of 18°, as shown in Fig. 2(c). Based on these results, we determined that 18° was the lowest pretilt angle possible to obtain a stable 4-D TN structure at 0 V for use with nematic LC. Further, while increasing the pretilt angle above 18° may result in enhanced stability, the transmittance of the sample was decreased.

Figures 3(a) and 3(b) show iso-contrast maps of a conventional 1-D TN cell and our stable 4-D TN cell, respectively. As expected, our 4-D TN structure had similar symmetrical viewing angle characteristics as the conventional 1-D TN cell. Figures 3(c) and 3(d) show the gray inversion characteristics of the 1-D and 4-D TN cells, respectively. In the conventional 1-D TN structure, gray scale inversion occurred at lower viewing directions. However, in the 4-D TN cell, gray inversion was not observed anywhere over the entire range.

Owing to the different twisting senses at adjacent subpixels in the 4-D TN structure, the stability of the multi-domain structure was weakened by both mechanical and thermal shock. Specifically, we first tested the thermal stability of the 4-D TN sample with a pretilt angle of 18°, as shown in Fig. 4, in which the sample was maintained at 100 °C and the texture was observed at 0 V. The stable 4-D TN structure started to collapse after annealing for 30 min (Figs. 4(a) and 4(b)). The weak thermal stability was accelerated at higher temperatures. After 30 min of thermal annealing at 120 °C, the 4-D TN structure collapsed to a 1-D TN structure in

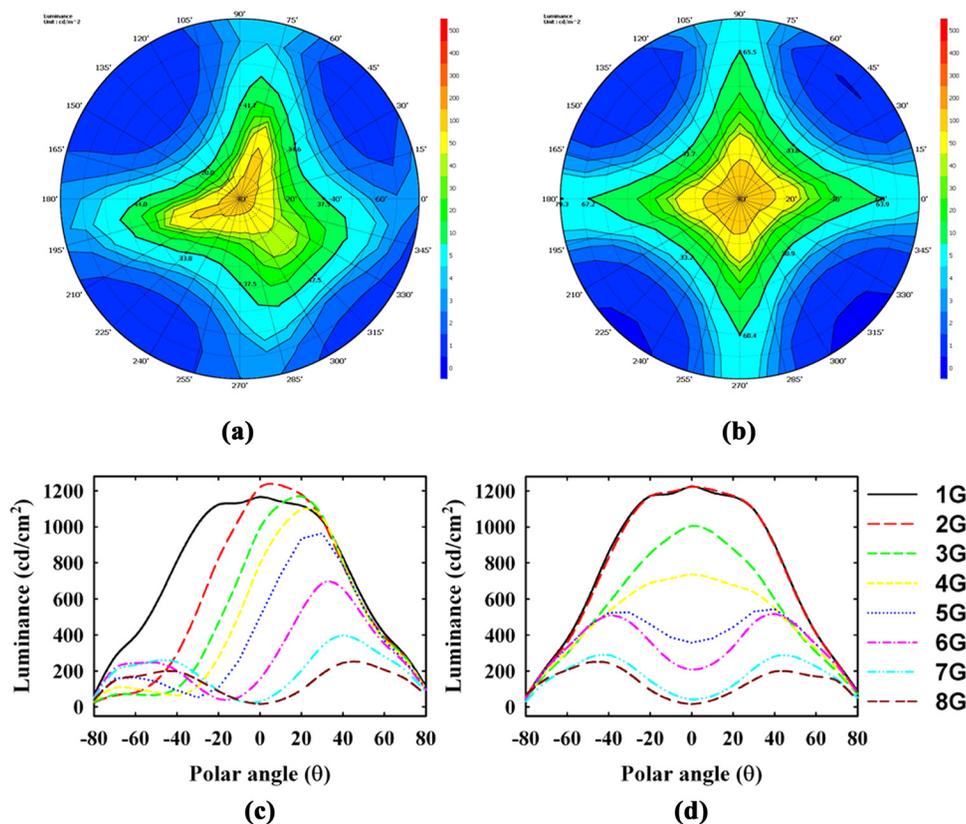


FIG. 3. Iso-contrast map of a (a) conventional 1-D TN cell and (b) stable 4-D TN cell. Gray inversion characteristics of (c) conventional mono-domain TN cells and (d) stable 4-D TN cells.

almost all areas (Figs. 4(c) and 4(d)). The structural stability can be improved by applying a higher pre-tilt angle, but this degrades the optical performance in terms of both transmittance and response time. Therefore, an alternative method was pursued to enhance the stability and optical performance of the 4-D TN structure with a lower pretilt angle.

Recently, we described a method for surface modification by mixing UV-curable RM monomers with a vertical alignment layer, which results in enhanced response time and improved anchoring energy of PVA-LCD.<sup>11</sup> In this

study, we applied the same technique to the vertical alignment layer using the stacked alignment method. In our previous study, we found that the pretilt angle is influenced by the mixed RM. Figure 5 shows the pre-tilt angle with increasing concentrations of the vertical alignment layer with and without RM mixing. We measured the pretilt angle of the sample with mixed RM after 10 min of UV exposure at 10 V. Our results show that the pretilt angle slightly increased with the RM mixed alignment layer at the same concentration of the vertical alignment layer. Specifically, the pretilt angle was approximately 18° with 2.8 wt. % of the vertical alignment layer with RM mixing. A pretilt angle of 18° was achieved at 3 wt. % without RM.

Figure 6 shows the thermal stability of the 4-D TN sample using 1.0 wt. % of RM mixed with vertical alignment layer of 2.8 wt. %. It is very clear that the 4-D TN structure

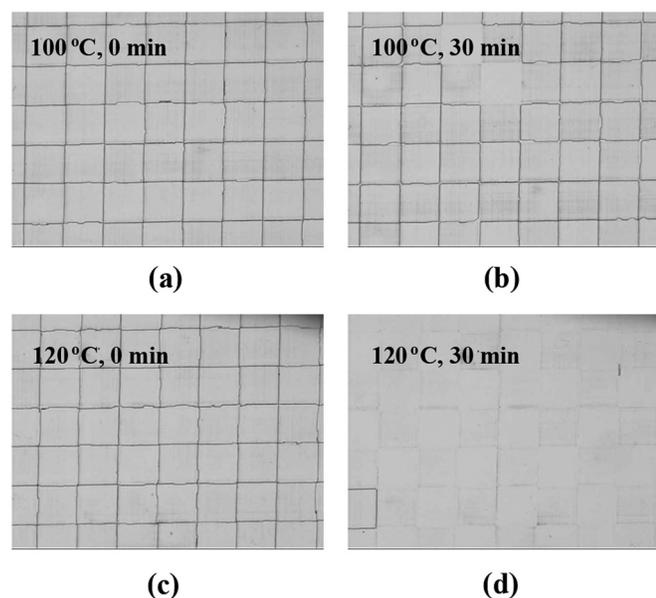


FIG. 4. Thermal stability of a 4-D TN cell with an 18° pretilt angle.

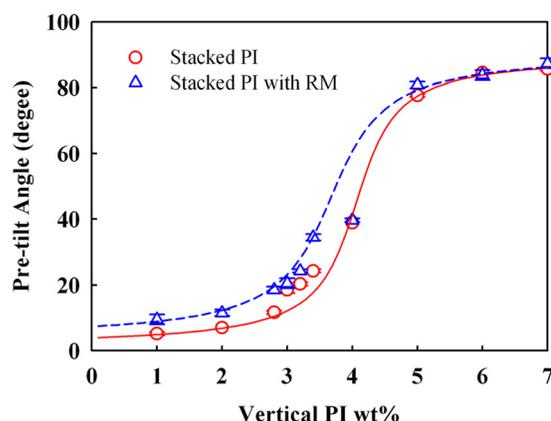


FIG. 5. Pretilt angle as a function of vertical alignment layer concentration.

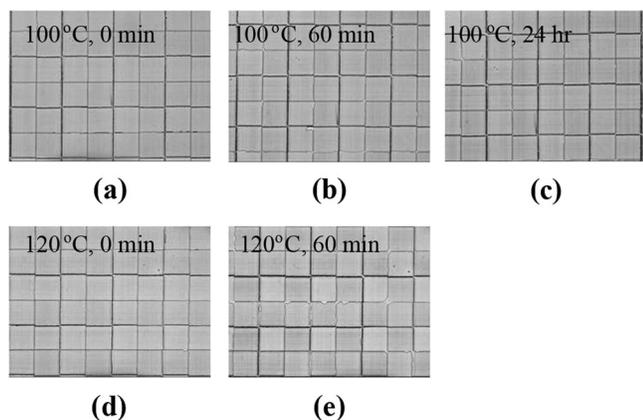


FIG. 6. Thermal stability of a 4-D TN sample with an RM mixed vertical alignment layer of 2.8 wt. %.

was maintained even after 24 h of annealing at 100 °C. Moreover, the structure remained stable after annealing for 60 additional minutes at 120 °C. We hypothesize that this improved stability is the result of increased anchoring energy due to the directionally polymerized RM monomers by the LC alignment, as discussed below.

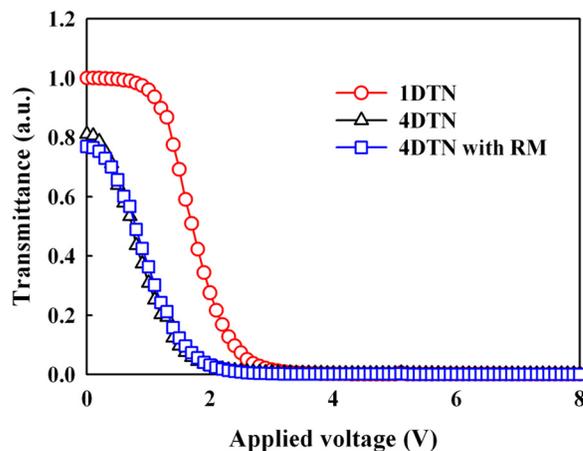


FIG. 7. Transmittance characteristics as a function of applied voltage for 1D TN and 4D TN with or without RM cells.

Figures 7 and 8 show the electro-optic performances of the 4-D TN samples with and without RM monomers with the same pretilt angle of 18°. The results obtained with the conventional 1-D TN sample are also shown for reference. The textures and transmittances of the 4-D TN sample with and

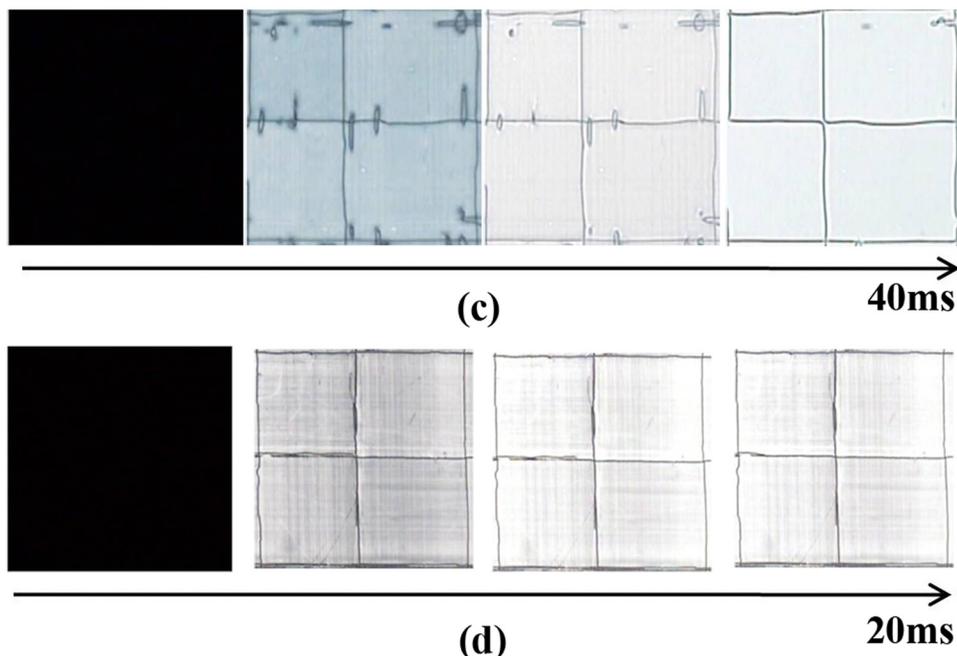
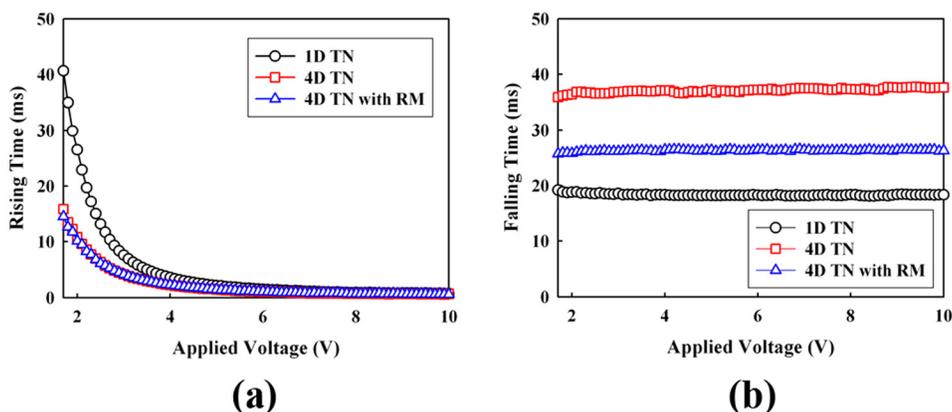


FIG. 8. (a) Rise and (b) fall characteristics for 1D TN and 4D TN cells with or without RM. Time resolved textures for 4D TN cells (c) without RM and (d) with RM.

without RM as a function of the applied voltage were almost the same when the maximum transmittance of the 1-D TN sample was approximately 80% (Fig. 7). The slight decrease in transmittance was a result of the higher pre-tilt angle of the 4-D TN sample than the 1-D TN structure and disclination lines between different twisting senses within the pixels.

Importantly, the response time was also affected by the pre-tilt angle as well as the stability of the disclination line between two sub-pixels with different twisting senses. As shown in Fig. 8(a), the field driven rise times of the 4-D TN samples were shorter than that of the 1-D TN samples due to the higher pre-tilt angle. However, the relaxation time increased more than two-fold for the 4-D TN sample without RM compared to the 1-D TN sample (Fig. 8(b)). The higher pre-tilt angle was one of the reasons for the slower observed response with the other important reason being the stability of the twisting senses between the left- and right-handed twisting directions. Figure 8(c) shows the textures of the samples as a function of time when 10 V was applied for the 4-D TN sample without RM. A transient change in the texture from a black state at 10 V to a white state at 0 V indicates the presence of complicated disclination lines, even for sub-pixels. For the 4-D TN sample with RM, the relaxation time was improved by about 30% compared to the sample without RM (Fig. 8(b)).

The transient textures of the samples with RM are shown in Figure 8(d), which clearly demonstrates that the texture was very uniform and the disclination between sub-pixels remained black during switching. This result indicates that each sub-pixel had a very stable TN structure with different twisting senses. We believe that this stability was the result of improved anchoring energy due to the presence of polymerized RM mixed with the alignment layer.

In order to test our hypothesis, we measured the polar anchoring energy of the samples with and without RM in stacked alignment system using a high field method. We generated a single domain sample with parallel rubbing on both substrates. The polar anchoring energy of the sample with RM ( $5.6 \times 10^{-4} \text{ J/m}^2$ ) was about 50% higher than that of the sample without RM ( $3.6 \times 10^{-4} \text{ J/m}^2$ ). Generally, the falling time of LC ( $\tau_o$ ) is governed by the rotational viscosity ( $\gamma_l$ ), cell gap ( $d$ ), and elastic coefficient ( $K$ ) as shown in the following relationship,  $\tau_o = \gamma_l d^2 / \pi^2 K$ .<sup>12</sup> This mathematical relationship was derived under the assumption of strong surface anchoring, i.e., an infinite surface anchoring energy ( $W \rightarrow \infty$ ). However, surface anchoring energies are close to finite, and larger anchoring energies have an important role in reducing the LC response time.<sup>13,14</sup> In our case, the polymerized RMs on the surface enhanced the surface anchoring energy and thus, the response time improved, especially with respect to the falling time.

An additional role of RMs in alignment layers is to reduce back-flow phenomenon. In a TN mode, when the applied voltage is removed, the transmittance does not increase monotonically due to back-flow phenomenon, which makes the relaxation time slower.<sup>12</sup> In the case of the 4-D TN structure, this phenomenon is more severe than in a mono-domain TN because the 4-D TN structure has two different types of twist sense within each given pixel. We

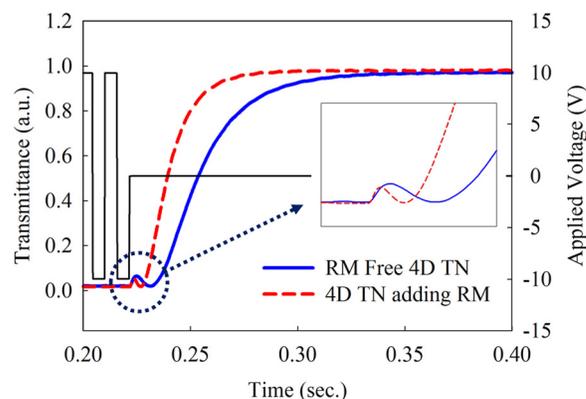


FIG. 9. Response time characteristics of conventional (RM free) 4D TN and RM added 4D TN.

compared the characteristics of the 4-D TN cells with and without RMs and the results are shown in Fig. 9. In this figure, the small bump in the optical transmittance is attributed to the back-flow phenomenon of the RM added to the 4-D TN cell, which was smaller than in the RM-free 4-D TN cell, thereby allowing for faster response time characteristics. This result may be due to the fact that polymerized RMs on the alignment surface memorized the twist direction at each sub-pixel and the LC molecules could subsequently be moved directly down to the substrate upon removal of a higher applied voltage.

#### IV. CONCLUDING REMARKS

In summary, we fabricated a 4-D TN structure that is stable over the entire driving voltage range with a high pre-tilt angle by stacking both planar and vertical alignment layers. As a single pixel has two kinds of twist sense and azimuthal directions for each sub-pixel, this approach successfully resulted in wide viewing angle characteristics. Furthermore, we improved the response time by introducing RM materials within the alignment layer. The polymerized RMs increased the polar anchoring energy and reduced the back-flow phenomenon when switching, which resulted in faster response time characteristics that are sufficient for moving pictures.

#### ACKNOWLEDGMENTS

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