

High-performance and Low-cost Aluminum Zinc Oxide and Gallium Zinc Oxide Electrodes for Liquid Crystal Displays

Andrii Varanytsia, Libo Weng, Tzu-Chieh Lin, Jing Yang, and Liang-Chy Chien

Abstract—The majority of electro-optical devices use transparent conductive oxide (TCO) electrodes such as indium tin oxide (ITO) in their structure. In this study we manufactured aluminum zinc oxide (AZO) and gallium zinc oxide (GZO) TCO films on glass substrates and demonstrate suitability of these materials for application as transparent electrodes of liquid crystal displays (LCD). We have established deposition process for fabrication of AZO and GZO coated glass substrates with average transmittance of 93% and 92% in the visible spectrum, and the sheet resistivity without annealing of ~120 Ohms/square. Using photolithographic patterning and wet etching of AZO and GZO films, we have successfully fabricated laboratory prototypes of twisted nematic (TN), vertical alignment (VA), in-plane switching (IPS), and fringe-field switching (FFS) LCDs. Electro-optical performance of AZO and GZO prototypes is comparable to those of the ITO prototypes of identical design. The AZO and GZO have demonstrated great potential to compete with ITO as low-cost TCOs for display applications. Our findings validated by numerical analyses will be useful for development of TCOs alternative for display and other optical or electro-optical applications.

Index Terms—Aluminum Zinc Oxide, Fringe-Field Switching, Gallium Zinc Oxide, In-plane Switching, Liquid Crystal Display, Transparent Conductive Oxide, Twisted Nematic, Vertical Alignment.

I. INTRODUCTION

ALL modern information displays are designed using one or two substrates with transparent conductive electrodes, usually made from a thin layer of transparent conductive oxide (TCO) deposited on the surface of rigid or flexible substrates. The most widespread TCO is the Indium Tin Oxide (ITO), which consists of large amount of indium oxide doped with small amount of tin oxide. Typically, commercial ITO for display applications has fractions of InO and SnO close to 9:1 [1]. Indium is a rather expensive material affecting the cost of final product. Alternative lower-cost TCO materials for display and other optical applications such as Zinc Oxide based TCOs are greatly attractive. Aluminum zinc oxide (AZO) and gallium

zinc oxide (GZO) draw extra attention due to their relatively low electrical resistivity and good transmission in the visible range of optical spectrum.

Fundamental optical, physical and electrical properties of AZO and GZO have been well studied [2-5]. Potential applications for these materials include electronic sensors, electrodes of solar cells and flat panel displays. Desired transmittance of the TCO films and electrical conductivity can be achieved by optimization of dopants concentration and carrier mobility in the TCO layer. At the same time, physical properties of AZO and GZO films strongly depend on the deposition condition, technique and post deposition treatment [1]. Techniques that have been used for preparing AZO and GZO thin films include sputtering, spray pyrolysis, pulsed laser ablation, reactive evaporation, chemical vapor deposition, solution growth, sol-gel, and cathodic vacuum arc techniques [1].

The objective of this work is to compare optical properties and electrical performance of AZO and GZO layers with ITO in a role of transparent conductive electrodes for application in twisted nematic (TN) [6], vertical alignment (VA) [7, 8], in-plane switching (IPS) [9, 10] and fringe-field switching (FFS) [11-13] liquid crystal displays (LCDs). We have fabricated AZO and GZO coated glass substrates with good transmittance in the visible range of optical spectrum, and the sheet resistivity similar to ITO. The AZO and GZO coated glass substrates were used to manufacture a single pixel laboratory prototype of TN and VA LCDs. High-resolution photolithography processing was successfully performed to pattern AZO and GZO TCOs into interdigitated electrodes. Patterned substrates were used to manufacture an IPS and FFS prototype devices. Optical and electro-optical performance of all AZO and GZO prototype display devices has been tested and confirmed to be comparable to properties of ITO prototypes with identical design.

II. MATERIALS AND METHODS

Optical and electrical properties of AZO and GZO layers deposited on the 1.1 mm-thick soda-lime glass substrates were

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TABLE I
THICKNESS AND SHEET RESISTANCE OF COMPARED TCO LAYERS

TCO	Layer thickness, nm	Sheet resistance, Ω/\square
ITO	25	75 - 85
AZO	200 - 240	110 - 120
GZO	100 - 140	120 - 130

compared with commercially available 1.1 mm-thick ITO coated soda-lime glass. The TN, VA and IPS prototype LCD devices were manufactured from glass substrates coated with single AZO, GZO layer or commercial ITO coated glass. The FFS LCD prototypes were made using glass substrates coated with two layers of AZO or ITO which were separated by ~ 300 nm thick electrically insulating SiO_2 passivation layer. The ITO-FFS prototypes were made from commercial ITO coated glass which was coated with a SiO_2 passivation layer and top ITO layer. For each type of tested LCD prototypes, the AZO or GZO devices were compared with a reference ITO device of an identical design.

The AZO and GZO layers were deposited on uncoated soda-lime glass substrates using a DC magnetron sputtering method without post annealing. Desired properties of sputtered AZO and GZO films were controlled by applied power, and the Ar gas pressure in the sputtering chamber. The SiO_2 passivation layer was deposited by AC magnetron sputtering method. The top ITO layer was deposited by pulsed DC magnetron reactive sputtering process.

Prepared substrates for TN, IPS and FFS prototypes were coated with polyimide alignment layer PI2555 (from HD Microsystems) for planar alignment, then mechanically buffed to define alignment direction for liquid crystal (LC) molecules on the surface. Substrates for VA prototypes were coated with polyimide alignment layer SE1211 (from Nissan Chemical) for homeotropic alignment, and then mechanically buffed to create small pretilt of LC molecules from the normal to the surface. Electro-optic cells of VA, IPS, and FFS LCD prototypes were assembled with rubbing directions antiparallel on opposite substrates, and at angle of 90° for TN prototypes. In IPS and FFS prototypes, the rubbing direction was chosen to be at an angle of 10° with respect to electrode lines. All cells with planar alignment were filled with a nematic liquid crystal (NLC) mixture TMS83700 (from HCCH) which has positive dielectric anisotropy ($\Delta\epsilon = 10.6$) and low birefringence ($\Delta n = 0.1$). Cells with homeotropic alignment were filled with a NLC mixture (from Merck) which has negative dielectric anisotropy ($\Delta\epsilon = -3.4$) and low birefringence ($\Delta n = 0.098$). Cell gap thickness was selected to be $\sim 4.9 \mu\text{m}$ in TN prototypes and $\sim 3.5 \mu\text{m}$ in VA, IPS and FFS prototypes, and was controlled by a particle spacer beads dispersed in the glue seal lines at the edges of the test cells. Performance of manufactured prototypes was evaluated by electro-optical measurements on the optical bench, and observations between crossed polarizers under polarizing optical microscope (POM) and on the light box. Electro-optical measurements were performed by recording the light transmission through the LC cell placed between a pair of crossed polarizers as a function of applied voltage. The rubbing

TABLE II
AVERAGE RESPONSE TIME AND MAXIMUM CR OF COMPARED PROTOTYPES

Cell No.	LCD prototype	Turn-on time, ms	Turn-off time, ms	Maximum CR
1	TN ITO	6.2	15.1	750:1
2	TN AZO	6.1	15.5	750:1
3	VA ITO	400.0	9.4	3000:1
4	VA AZO	440.0	10.1	3000:1
5	IPS ITO	20.7	19.6	1300:1
6	IPS GZO	17.5	18.0	1100:1
7	IPS AZO	19.2	20.6	800:1
8	FFS ITO	28.9	19.4	1500:1
9	FFS AZO	28.3	20.7	500:1

directions in samples with planar alignment were parallel to one optical axes of polarizers, and was aligned at 45° between optical axes of a pair of crossed polarizers in VA samples. Both alignment approaches of the rubbing direction provide the best contrast ratio (CR) in TN, IPS, FFS and VA LCD prototypes. The He:Ne laser (633 nm) was used as a light source for electro-optical measurements.

III. RESULTS AND DISCUSSION

The resistivity of AZO and GZO is larger than in ITO. The film thickness of deposited TCO layers was selected to match approximately the sheet resistance of commercial ITO coated glass. Film thickness and sheet resistance of compared TCO substrates are shown in Table 1. Sheet resistance of AZO and GZO films can be further decreased by fine optimization of deposition condition and post-annealing process, in order to

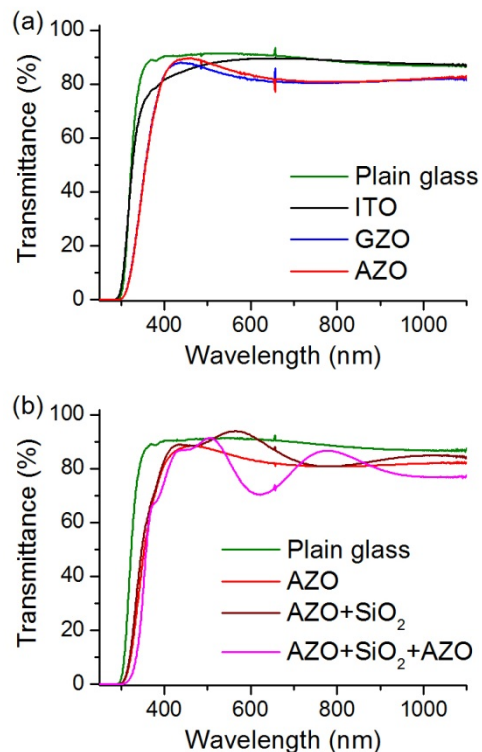


Fig. 1. Transmission spectra of: (a) uncoated soda-lime glass, single TCO layer ITO, AZO, or GZO coated glass substrates used for IPS prototypes, (b) three-layered substrate used for AZO FFS prototypes.

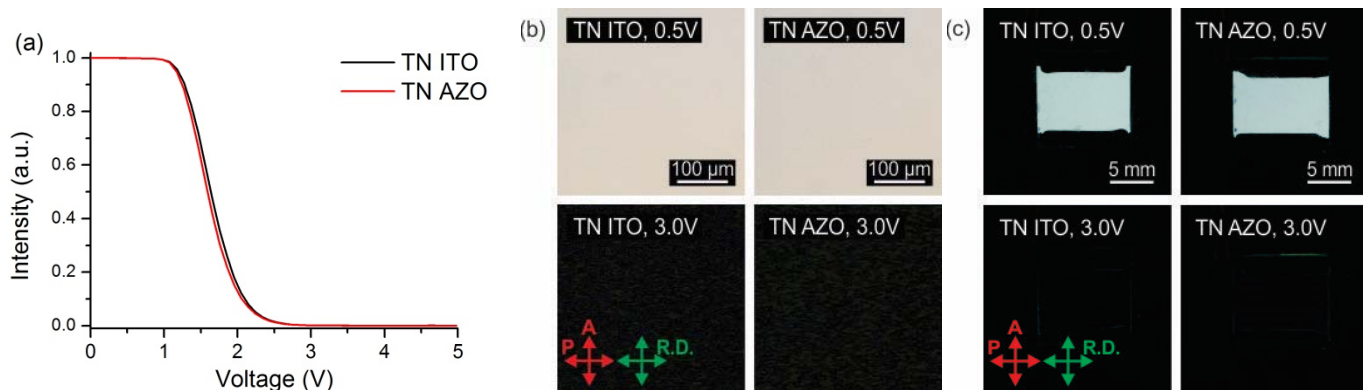


Fig. 2. (a) Static voltage response of TN AZO, and TN ITO prototype samples. Appearance of TN AZO and TN ITO prototype cells (b) under the POM, and (c) on the light box.

obtain desired microscopic structure or structural relaxation of deposited film promoting better carrier mobility.

Fabricated AZO and GZO films are highly transparent in the whole visible range of spectrum. Typical transmission spectra of fabricated AZO, GZO and commercial ITO-coated glass substrates are shown on Fig. 1. In general, AZO and GZO films deposited on glass substrates are slightly more reflective comparing to commercial ITO-coated glass due to their larger film thickness. With respect to uncoated glass the transmittance of deposited AZO and GZO films averaged in the range of wavelength from 380 nm to 780 nm is 92.8% and 91.6%, respectively. In comparison, an average transmittance of ITO-coated soda-lime glass substrate is 96.8%. Due to the smaller band gap of ZnO ($E_{\text{gap}} \sim 3.4\text{eV}$), the UV cut-off wavelengths of AZO and GZO substrates are typically at slightly larger wavelength comparing to ITO ($E_{\text{gap}} \sim 3.6\text{eV}$) or uncoated glass, but are always below 350 nm.

The electro-optic performance of TN ITO (cell 1) and TN AZO (cell 2) prototypes is compared on Fig. 2. In normally-bright configuration of polarizers the TN device is transparent in the Off state, and is switched to a electric field induced dark On state. Normalized static voltage responses of prototype cells 1&2 are identical. The bright and dark states have very similar light transmittance, providing an electro-optic contrast ratio (CR) of $\sim 750:1$ in both types of compared TN devices, as shown on Fig. 2 b&c. The electro-optic switching response times of prototype cells 1&2 are different by a value comparable to an

error of measurement and therefore considered identical, as listed in Table 2.

The electro-optic performance of VA ITO (cell 3) and VA AZO (cell 4) prototypes is compared in Fig. 3. The TV curves of VA prototypes of both types have the same switching threshold and well matched with each other with minor deviations due to small variation of the cell gap thickness of tested electro-optic cells. With homeotropic LC alignment, it is not necessary to match very precisely orientation of polarizers with alignment direction of LC like in the case of planar alignment. Therefore, dark state and CR of VA LCD mode is typically better compared to modes with planar alignment. As expected, both cells 3&4 prototypes have very good and same maximum CR of up to 3000:1. However, a practical challenge for VA LCD mode is to create an effective pretilt angle of LC molecules with respect to vertical direction in order to define orientation of LC during switching and decrease response time. In this study pretilt angle was induced only by mechanical rubbing of the surface of polyimide. Obtained pretilt angle of LC in ITO and AZO VA prototypes was verified by crystal rotation method [14] to be $\sim 89.0^\circ$. Limited ability to affectively generate pretilt angle in compared VA LCD prototypes is the main reason for turn-on switching time to be much larger comparing to prototypes with planar alignment. Additionally, average turn-on switching time of prototype cell 4 was $\sim 10\%$ longer comparing to prototype cell 3, as shown in Table 2. Such behavior is explained by rougher surface of not annealed AZO

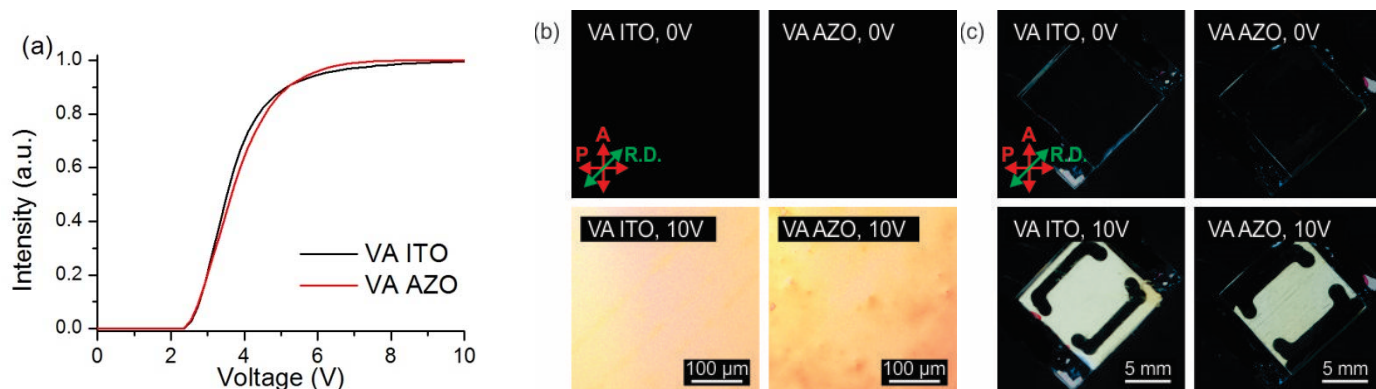


Fig. 3. (a) Static voltage response of VA AZO, and VA ITO prototype samples. Appearance of VA AZO and VA ITO prototype cells (b) under the POM, and (c) on the light box.

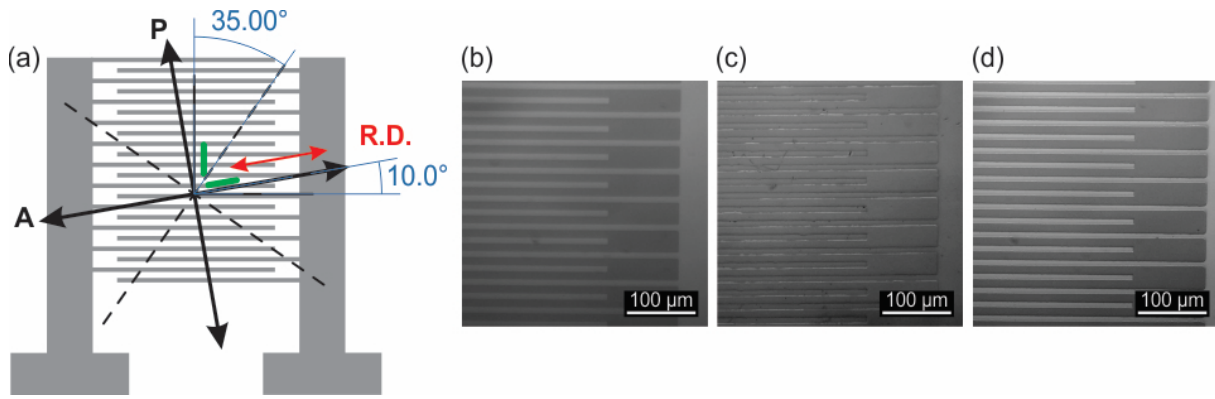


Fig. 4. (a) Interdigitated electrode pattern for IPS and FFS pixels. The drawing is not to scale. Micrographs of photolithographically patterned electrodes produced on: (b) commercial ITO coated glass, (c) manufactured GZO substrate, (d) manufactured AZO substrate. Width and gaps between electrodes are 10 μ m.

compared to commercial ITO, which decreases efficiency to generate pretilt of LC molecules using a mechanical rubbing method. A small difference between average turn-off switching

time of cells 3&4 can arise from cell gap thickness difference from cell to cell and measurement error.

The photolithography of AZO, GZO and ITO layers was performed using positive photoresist S1818 (Dow Chemical) with UV light exposure through high-resolution chromium shadow mask placed in a direct contact with the substrate. Exposed substrates were developed in a weak base solution, open areas of TCO layer were etched away using acid solution, and substrates were striped from photoresist using strong base solution or acetone. Acid solution was a mixture of hydrochloric and nitric acids. The same type of acid solution was used to etch AZO, GZO and ITO TCOs. However, AZO and GZO films are significantly more reactive with the acid. Therefore, etching solution was diluted to 0.5 vol.% and etching time was decreased 24 times compared to used for etching of commercial ITO. The AZO and GZO layers were patterned into a fine electrode pattern with an active area of both IPS and FFS pixels made of interdigitated electrodes having the width and spacing of 10 μ m. Schematic drawing of an IPS electrode pattern and micrographs of produced pixels are shown in Fig. 4.

Electro-optical performance of AZO, GZO and ITO IPS prototype samples is compared in Fig. 5. Normalized TV curves of AZO IPS and GZO IPS prototypes are identical to reference sample made from commercial ITO coated glass. Small horizontal separation of TV curves visible on Fig. 5a is caused by a combined contribution of practical limitation to reproduce the exact dimensions of IPS electrode pattern, and random electrical imperfections creating small electrical shortenings of right and left IPS electrodes. As verified by experimental observations and confirmed by numerical analyses, shorted IPS or FFS electrodes are able to increase operating voltage significantly.

The TV curves of samples with a FFS structure of electrodes show that electro-optic performance of AZO prototypes is indistinguishable from reference ITO prototypes as well, as shown in Fig. 6. The electrical quality of FFS substrates is controlled by the thickness of passivation layer which separates top and bottom electrodes and is not sensitive to photolithographic defects of the top layer. Therefore, experimental TV curves of FFS AZO and FFS ITO prototypes match better compared to IPS prototypes. The appearance of

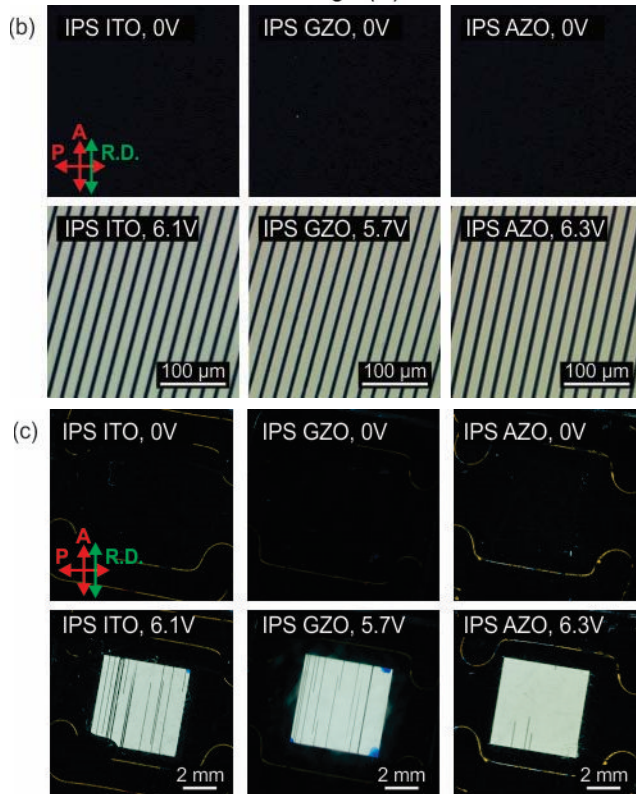
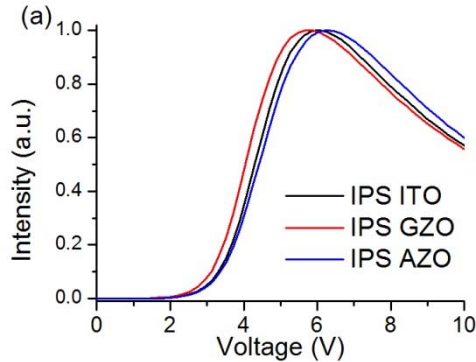


Fig. 5. AZO, GZO, and ITO prototype samples with IPS electrode structure: (a) static voltage response, (b) appearance under the POM, and (c) appearance on the light box. Electrode width and gaps are 10 μ m.

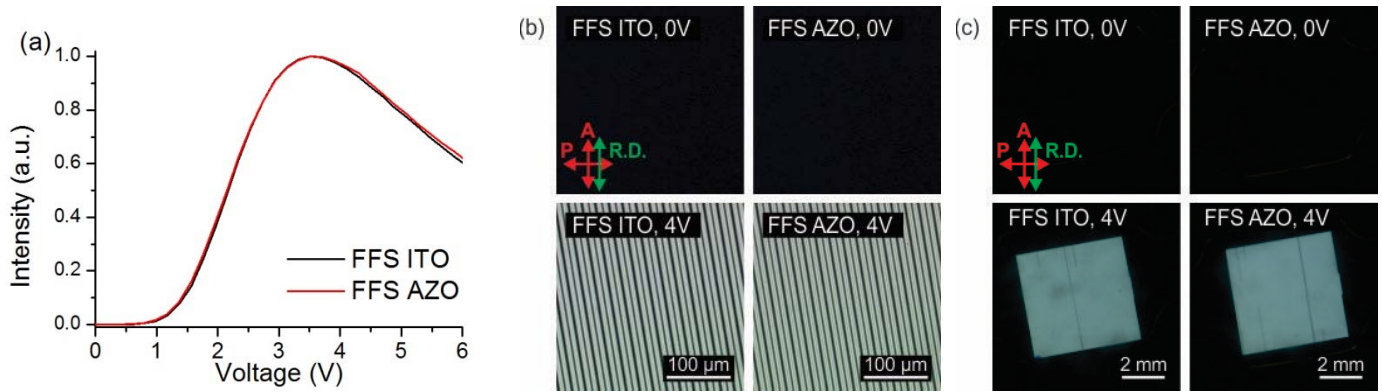


Fig. 6. AZO, GZO, and ITO prototype samples with FFS electrode structure: (a) static voltage response, (b) appearance under the POM, and (c) appearance on the light box. The width and gaps between electrodes are 10 μ m.

compared prototype cells between crossed polarizers under a POM and on the light box demonstrate few dark stripes from inactive electrode lines due to imperfections of photolithography in both IPS and FFS prototypes, as shown on Fig. 5c, and Fig 6c, respectively.

An average turn-on and turn-off switching response time of all AZO, GZO and ITO IPS prototypes is \sim 20 ms. An average response time of FFS prototypes samples is \sim 29 ms for turn-on switching, and \sim 20 ms for turn-off switching. The response time was measured for switching between 0V and V_{on} states corresponding to maximum brightness and was calculated between the points of 10% from the minimum and 90% from the maximum of the transmitted light intensity through the sample. From a point of view of the response time AZO and GZO prototypes are indistinguishable from reference ITO prototypes. The maximum CR is 1300:1 in IPS ITO prototypes, 800:1 in IPS AZO prototypes, and 1100:1 in IPS GZO prototypes. Smaller maximum CR in IPS AZO and IPS GZO prototypes is a result of larger thickness of AZO and GZO TCO layers comparing to ITO. Higher surface profile of electrode pattern comparing to open areas makes patterned AZO and GZO substrates more susceptible to mechanical buffing induced alignment imperfections creating minor light leakage in the dark state. Similarly, the maximum CR of FFS ITO prototypes is 1500:1, and in FFS AZO prototypes is 500:1. This potential problem is expected to be easily eliminated by optimization of conditions for mechanical buffing or using non-contact alignment techniques such as photoalignment.

Experimentally obtained TV curves were validated with results of numerical simulation performed using a TechWiz

LCD software (SANAYi System Co.). The TV curves of IPS and FFS devices with the same electro-optic cell design and NLC material properties as in manufactured prototypes were calculated for the thickness of TCO layer equal to 25 nm, 140 nm and 220 nm. According to simulation results, as the TCO thickness increases by 8.8 times the maximum transmittance from TV curves of the IPS device decreases by 2.9%, and at the same time driving voltage decreases by 1.8%, as shown on Fig. 7a. For the FFS device, an 8.8 times increase of TCO layer thickness decreases transmittance by 3.3% and decreases driving voltage by not more that 1.3%, as shown on Fig. 7b. Detailed analysis of simulated TV curves and transmittance profiles at different applied voltage reveals that thicker TCO layer increases area fraction of dark stripes above electrodes and at the same time creates stronger horizontal component of electric field in the bulk of the LC layer in-between electrodes, resulting in a small decrease of both driving voltage and maximum transmittance. Calculated changes are comparable to experimental error coming from fluctuations of the intensity of laser beam and accuracy to reproduce sample cells and therefore cannot be reliably observed in our experiments. Performed numerical simulation demonstrates that selected cell design provides slightly smaller brightness of FFS device compared to IPS, as shown on Fig. 7c. The electro-optic performance of both IPS and FFS devices can be improved by further modification of cell design [15-17], selection of properties of LC material [18, 19], which is out of scope of current study.

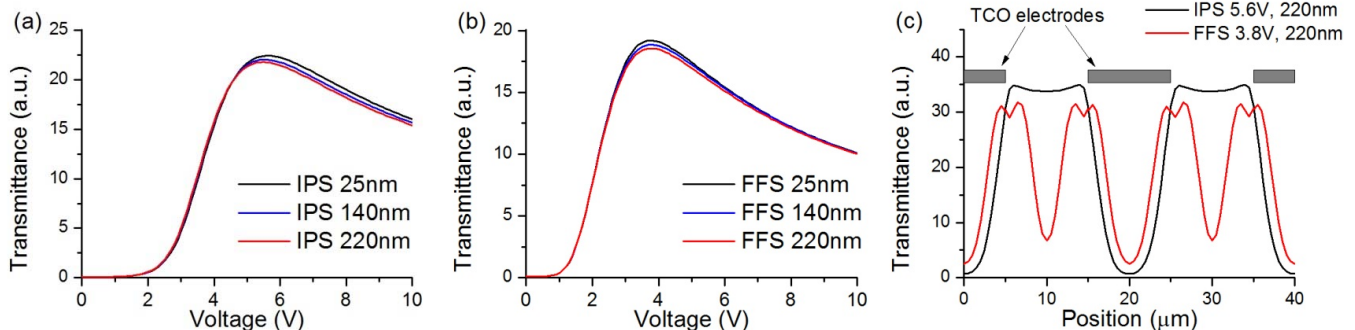


Fig. 7. Numerically simulated TV curves with different thickness of TCO layer for: (a) IPS prototypes (b) FFS prototypes, and (c) numerically simulated transmittance profiles corresponding to a state of maximum transmittance in IPS and FFS cells. The width and gaps between electrodes are 10 μ m.

IV. CONCLUSION

We have fabricated prototype TN, VA, IPS and FFS LCD devices using AZO and GZO films. The AZO and GZO films are demonstrated to have electrical and optical properties equivalent to ITO-based devices of identical design. We have established photolithographic process for AZO and GZO films and demonstrated its scalability and suitability for large area high resolution displays. Electro-optical performance of all types of fabricated AZO and GZO prototype devices is demonstrated to be equivalent to that of the reference ITO prototypes of identical design. The response time of all compared TN, IPS and FFS prototype devices is demonstrated to be equivalent and independent on the type of TCO material. Due to different surface roughness the response time of VA AZO prototypes was observed to be larger compared to VA ITO prototypes. Larger thickness of TCO slightly reduces the maximum CR via alignment nonuniformities produced by mechanical rubbing. Performed numerical simulation has demonstrated that AZO due to its larger film thickness compared to ITO changes distribution of electric field in the bulk of LC layer decreasing both maximum transmittance and driving voltage by 1% - 3%. Regardless noticeable small difference of transmission spectra of AZO and GZO films compared to ITO, all of LCD prototypes compared in this study between crossed polarizers appear identically colored when viewed with a white light backlight. Our findings will be useful for development of TCOs which are competitive to ITO as inexpensive alternatives for displays, smart windows and other electro-optical applications.

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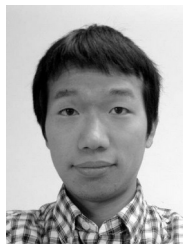
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Tzu-Chieh Lin, photograph and biography not available at time of publication.

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