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Performance Enhancement of Transparent Amorphous IGZO Thin-Film Transistor Realized by Sputtered Amorphous AlO_x Passivation Layer

Yuanbo Li,^{1,z} Jianxun Sun,¹ Teddy Salim,² Rongyue Liu,¹ and Tupei Chen^{1,z}

¹School of Electrical and Electronic Engineering, Nanyang Technological University, 639798 Singapore

²School of Materials Science and Engineering, Nanyang Technological University, 639798 Singapore

We report a high-mobility transparent Indium-Gallium-Zinc-Oxide (IGZO) thin-film transistor (TFT) with sputtered AlO_x passivation layer. The interfacial region between the IGZO layer and the AlO_x layer played a crucial role in improving the field-effect mobility (the maximum field-effect mobility increased from 6.292 cm² Vs⁻¹ for the TFT without the AlO_x layer to 69.01 cm² Vs⁻¹ for the TFT with the passivation layer) and the on/off current ratio (from ~10⁷ without the layer to ~10⁸ with the layer). The driving current of IGZO TFT was also significantly enhanced. The formation of the interfacial layer has been investigated and verified. The ion bombardment during the AlO_x deposition broke the In-O bond in IGZO, generating oxygen ions (O²⁻). The segregation of the O²⁻ was facilitated by the sputtered amorphous AlO_x. A metallic In-rich layer with high oxygen vacancy concentration was formed at the interface, leading to an increase in the carrier concentration in the interfacial layer. Besides the electrical performance, the reliability tests, including long-term exposure in the ambient environment and positive bias illumination stress (PBIS), showed improved results as well.

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Amorphous IGZO thin-film transistor (TFT) has attracted much attention for transparent flat-panel display (FPD) applications like AMOLED^{1,2} and AMLCD since Nomura et al. reported in Nature, 2004.³ Due to the high transparency, high field-effect mobility (μ_{FE} up to 10 cm² Vs⁻¹), good uniformity, small threshold voltage, small subthreshold swing and low-temperature fabrication process, a-IGZO has gradually replaced hydrogenated amorphous Si (a-Si:H) and low-temperature polycrystalline Si (LTPS) as the channel semiconductor of TFT.⁴⁻⁶ Nevertheless, for the next generation FPD, uniform brightness, larger area, higher resolution, and higher frame rates are in demand. Improving the driving capability and reliability of a-IGZO TFT becomes one of the critical concerns for many researchers.⁷

One of the most important reliability issues is the threshold voltage shift of the TFT, which causes brightness variation of the display pixel.⁸ This instability could originate from ambient molecules penetration, positive or negative bias stresses and illumination stress, as explained in the following. For the as-deposited bottom-gate-top-contact TFT, moisture or oxygen could be absorbed into the back surface of IGZO. The absorbed H₂O (O₂) could act as donor (acceptor), changing the carrier concentration and thus the threshold voltage.⁹ Under positive bias, electrons could be trapped at the IGZO/dielectric interface or in the bulk of dielectric, leading a shift in the threshold voltage.¹⁰ Illumination stress can create electron-hole pairs due to electron excitation from the valence band to the conduction band or excite electrons from the valence band to the mid-gap states, both generating holes in the valence band. The generated holes are attracted to the bulk of the dielectric under negative bias, cause a shift in the threshold voltage.¹¹ To solve the reliability issue, a useful method is to deposit a passivation layer on the back surface of the channel layer,^{10,11} which could effectively prevent the ambient gases (hydrogen and oxygen) and moisture from penetrating into the channel region. To date, silicon oxides (SiO_x) or silicon nitrides (SiN_x) deposited by plasma-enhanced chemical vapor deposition (PECVD) and aluminum oxides (Al₂O₃) deposited by atomic layered deposition (ALD) are common materials used for passivation.¹²⁻¹⁴ The above options could effectively reduce the shift of the threshold voltage under light illumination, high temperature

and positive or negative bias. In the meantime, both ALD and PECVD processes could introduce hydrogen into the channel region, which slightly improves the electron mobility.¹⁴

Although the TFT reliability has been improved significantly by the passivation, the mobility and driving current of the TFT may be not large enough for certain applications, such as virtual/augmented reality, micro-LED, biometric sensing applications, etc.¹⁵ In this work, we reported an amorphous aluminum oxide (AlO_x) passivation layer deposited by RF magnetron sputter at room temperature. After post-passivation annealing, the passivated TFT showed much higher mobility, larger driving current and better reliability compared with the TFT without the passivation layer.

Device and Experiment

TFTs with bottom-gate-top-contact (BGTC) structure were fabricated, as shown in Figures 1a–1c. A 35 nm-thick Al₂O₃ layer was deposited on ITO (200 nm) glass by ALD process at 250 °C with Trimethylaluminum (TMA) and water (H₂O) as the precursors, followed by a 5 nm-thick SiO₂ layer deposition by PECVD process at 300 °C. This bilayer dielectric structure aims to lower the off current and decrease the interface defect states.¹⁶ The breakdown electric field of the bilayer dielectric structure was tested and shown in Fig. S1 (available online at stacks.iop.org/JSS/10/045006/mmedia) in the supporting information. After the formation of bilayer gate dielectric, a 45 nm-thick amorphous IGZO (In: Ga: Zn = 1:1:1) was deposited by RF magnetron sputtering with RF power of 100 W and Ar flow rate of 50 sccm. Then, a 250 nm-thick ITO layer, which serves as the source and drain contacts, was deposited by RF magnetron sputtering with RF power of 100 W and Ar flow rate of 50 sccm. Subsequently, we deposited amorphous AlO_x passivation layer at the top of the device by RF magnetron sputtering. The target used for sputtering deposition of the AlO_x passivation layer is an Al₂O₃ target with purity of 99.99% purchased from ACI Alloy. The RF sputtering was conducted with RF power of 100 W at process pressure of 8 mTorr with 100% Ar flow. All the sputtering processes for deposition of the IGZO, ITO and AlO_x layers were performed at room temperature. The patterning of the IGZO, ITO, and AlO_x layers was carried out by using the UV lithography and lift-off processes. TFTs with various channel widths (W) and channel lengths (L) were fabricated. Finally, the as-fabricated TFTs were annealed at 300 °C in N₂ atmosphere for

^zE-mail: yuanbo001@e.ntu.edu.sg; echentp@ntu.edu.sg

Figure 1. Schematic illustration of the TFT structure (a), top view of the TFT structure ($W/L = 800 \mu\text{m}/40 \mu\text{m}$) observed through an optical microscope (b), photo image of the TFT structure (c), schematic illustration of the stack-layered structure for optical transmittance measurement (d), and photo image of the stack-layered structure (e).

1 h. Schematic illustration of the TFT structure, top view of the structure observed through an optical microscope, and photo image of the structure are shown in Figs 1a–1c, respectively. In the meantime, a stack-layered structure was fabricated for the optical transmittance measurement of the TFT structure. Schematic illustration and photo image of the stack-layered structure are shown in Figs. 1d and 1e, respectively. In addition, for comparison of electrical performance, TFT without the AlO_x passivation layer was fabricated also.

Optical transmittance measurement of the stack-layered structure was conducted with LAMBDA 365 UV/Vis Spectrophotometer (PerkinElmer). The light wavelength for the measurement was swept from 400 nm to 700 nm, namely in the visible light range. The electrical characteristics of TFT were measured with a Keithley 4200 SCS parameter analyzer in dark environment. For the measurement of the transfer curves, the gate voltage was swept from -7 V to 10 V , while the drain voltage was fixed at 0.1 V , 1 V and 3 V , respectively. For the measurement of the output curves, the drain voltage was swept from 0 to 7 V and the gate voltage was fixed at 5 V , 7 V and 10 V , respectively. The transfer curves and output curves are shown in Figs. S2 and S3 of Supporting Information, respectively. To investigate the structure of the passivated TFT, High-Resolution Transmission Electron Microscopy (HRTEM) (JEOL 2010 HR) was used to observe the IGZO/ AlO_x interface. Likewise, X-ray Photoelectron Spectroscopy (XPS) and Auger Electron Spectroscopy (AES) depth profiling were used to track the shift of the binding energy for the elements of interest, usually attributed to the change in the oxidation state and chemical environment, with depth. Firstly, AES depth profiles were obtained using a field-emission Auger microprobe (JEOL JAMP-7830F) with a primary electron beam having an accelerating voltage of 10 keV

and a probe current of 10 nA . The sample was tilted at 30° throughout the analysis and the analysis area was approximately $15 \times 15 \mu\text{m}^2$. The analyzer was operated at a constant retarding ratio (CRR) mode (energy resolution $< 0.5\%$). Floating micro-ion etching device (FMIED) delivering an ion beam of 500-eV Ar^+ was used to sputter the thin-film samples over an area of $1.5 \times 1.5 \text{ mm}^2$ from the AlO_x surface onwards for depth profiling. The AES spectra were recorded at an etching rate of 30 s/cycle .

XPS analysis was performed using an AXIS Supra spectrometer (Kratos Analytical Inc., UK) equipped with a hemispherical analyzer and a monochromatic Al K-alpha source (1487 eV) operated at 25 mA and 15 kV . XPS depth profiling experiments were carried out using an Ar Gas Cluster Ion Source (GCIS, Kratos Analytical Inc. Minibeam 6) operated at 5 keV Ar^+ with a raster size of $1 \times 1 \text{ mm}^2$. Selected-area XPS spectra were acquired after each etching cycle (20 s) from an area defined by a $110\text{-}\mu\text{m}$ aperture at a take-off angle of 90° . A 3.1-volt bias was applied to the sample to neutralize charge built up on the sample surface. The high-resolution scans were recorded at 40 eV pass energy. The binding energies (BEs) were charge-corrected based on the C 1 s at 284.8 eV .

To verify the crystalline state of sputtered AlO_x , X-ray diffraction (XRD) was performed in the 2θ scan mode with Bruker D8 Discover XRD system operated with Cu-K α radiation (40 V , 40 mA) at an incidence angle of 0.5° . The grazing-incidence wide-angle scattering (GiWAXS) pattern was obtained with Nano-inXider (Xenocs) with Cu-K α microsource (30 W) at an incidence angle of 0.2° . The measurement was done at the rotation angle $\Psi = 0^\circ$ to capture the out-of-plane scattering pattern from the sample.

Results and Discussions

Optical transmittance measurement result of the stack-layered structure is shown in Fig. 2a. In the visible light range (400 nm ~ 700 nm), the structure showed a transmittance of around 80%, indicating that the fabricated device is highly transparent.

Transfer curve of an TFT with the AlO_x passivation layer without post-annealing is shown in Fig. 2b. It can be observed from Fig. 2b that the device was at a normally on state and could not be turned off. Figure 2c shows the comparison of the transfer characteristic measured at $V_{\text{DS}} = 1$ V between the TFTs with and without the AlO_x passivation after annealing, while Fig. 2d shows the comparison of the output characteristic measured at $V_{\text{GS}} = 10$ V between the TFTs with and without the AlO_x passivation after annealing.

From the transfer curves, output current on/off ratio, threshold voltage V_{th} , subthreshold swing S.S. and field effect mobility μ_{FE} were extracted and calculated. The on/off ratio was extracted straightforward from the semi-logarithm transfer curve. From the linear transfer curve, V_{th} was extracted from the intercept of the linear extrapolation at low V_{DS} , as shown in Fig. 2c. S.S. was calculated by Eq. 1,¹⁷

$$\text{S.S.} = \left(\frac{d \log I_{\text{D}}}{d V_{\text{GS}}} \Big|_{\text{max}} \right)^{-1} \quad [1]$$

and the field-effect mobility was calculated by Eq. 2,¹⁷

$$\mu_{\text{FE}} = \frac{\frac{g_{\text{m}}}{W}}{C_{\text{ox}} \frac{L}{V_{\text{DS}}}}, \quad [2]$$

where C_{ox} is the dielectric layer capacitance per unit area; and g_{m} is the transconductance at low V_{DS} , which could be derived from the slope of the linear extrapolation of the transfer curve. The value of C_{ox} is obtained from the following calculation by taking the 35 nm Al_2O_3 and 5 nm SiO_2 layers into account:

$$C_{\text{ox}} = \frac{1}{\frac{35 \times 10^{-7} \text{cm}}{8.854 \times 10^{-14} \text{F/cm} \times 9} + \frac{5 \times 10^{-7} \text{cm}}{8.854 \times 10^{-14} \text{F/cm} \times 3.9}} = 1.712 \times 10^{-7} \text{F/cm}^2$$

For the TFT without passivation, the on/off ratio was 1.3×10^7 , threshold voltage V_{th} was 3.8 V, subthreshold swing S.S. was 0.25 V/decade and maximum field effect mobility μ_{FE} was $6.292 \text{ cm}^2 \text{ Vs}^{-1}$ (with V_{GS} swept from -7 V to 10 V and $V_{\text{DS}} = 1$ V). For the TFT with AlO_x passivation, the on/off ratio was 1.87×10^8 , threshold voltage V_{th} was 2.7 V, subthreshold swing S.S. was 0.58 V/decade, maximum field effect mobility μ_{FE} was $69.01 \text{ cm}^2 \text{ Vs}^{-1}$ (with V_{GS} swept from -7 V to 10 V and $V_{\text{DS}} = 1$ V). The performance comparison is summarized in Table I. The field effect mobility versus applied gate voltage curves of these two type of devices (i.e. the TFTs without and with the AlO_x passivation layer) are shown in Figs. S4a and S4b in Supporting Information. Note that the calculation of the mobility is based on the assumption that the gate capacitance does not change significantly during the deposition of the passivation layer (PVL). The assumption is supported by the C-V measurement of the gate capacitance of the TFTs with and without the passivation layer. A comparison of TFT electrical performance between this work and other recent works that reported high performance is shown in Table II.

Besides the above measurements of electrical characteristics, transmission line method (TLM) was conducted to measure the contact characteristics between the ITO source (drain) electrode and the IGZO layer and investigate the effect of the sputtered AlO_x passivation on the contact resistance and resistance of the IGZO layer. In the TLM model, the total resistance of the conduction path can be divided into two parts, the resistance of the IGZO layer and the contact resistance between the ITO source (drain) electrode and the IGZO layer, as described by Eq. 3,²⁰

Figure 2. (a) Optical transmittance measurement of the stack-layered structure, (b) transfer curve measured at $V_{\text{DS}} = 1$ V of the TFT with the AlO_x passivation layer, with W/L of $800/40 \mu\text{m}$, and without post-annealing, (c) comparison of the transfer characteristic measured at $V_{\text{DS}} = 1$ V between the TFTs with and without the AlO_x passivation after annealing, and (d) comparison of the output characteristic measured at $V_{\text{GS}} = 10$ V between the TFTs with and without the AlO_x passivation after annealing.

Table I. Comparison of electrical performance between TFTs with and without AlO_x passivation layer (PVL).

	On/off ratio	V _{th}	S.S. (V/decade)	max μ _{FE} (cm ² Vs ⁻¹)	L _T (μm)
TFT with AlO _x passivation	1.87 × 10 ⁸	2.7	0.58	69.01	8.0
TFT without passivation	1.30 × 10 ⁷	3.8	0.25	6.292	11.4

Table II. Comparison of TFT electrical performance between this work and other recent works.

	On/off ratio	V _{th}	S.S. (V/decade)	μ _{FE} (cm ² Vs ⁻¹)
TFT in Ref. 15	8.90 × 10 ⁸	-1.3	0.26	65.50
TFT in Ref. 18	1.00 × 10 ⁴	-0.1	0.46	68.20
TFT in Ref. 19	2.40 × 10 ¹⁰	1.5	0.20	11.50
TFT in this work	1.87 × 10 ⁸	2.7	0.58	69.01

$$R_{\text{total}} = \frac{V_{\text{DS}}}{I_{\text{DS}}} = r_{\text{IGZO}} \frac{L}{W} + (R_{\text{S}} + R_{\text{D}}) = \frac{r_{\text{IGZO}}}{W} L + 2R_{\text{C}} \quad [3]$$

$$R_{\text{total}} = \frac{r_{\text{IGZO}}}{W} L + 2R_{\text{C}} = \frac{r_{\text{IGZO}}}{W} (L + 2L_{\text{T}}), \quad [4]$$

where $\frac{r_{\text{IGZO}}}{W}$ denotes the IGZO layer resistance per unit channel length and $2R_{\text{C}}$ denotes the contact resistance between the ITO and IGZO layers of the source and drain contacts.

In this case, TFTs with fixed channel width of 80 μm and various channel lengths of 5 μm, 10 μm, 20 μm, 40 μm and 100 μm were selected to construct the total resistance versus channel length curves at different gate voltages, as shown in Fig. 3.

As Fig. 3 shows, for the TFT without passivation, with increasing gate voltage from 6 V to 10 V, the slope of the curves, i.e. r_{IGZO}/W , decreased from $2.5 \times 10^3 \Omega/\mu\text{m}$ to $1.0 \times 10^3 \Omega/\mu\text{m}$, and at the same time the intercept at $L = 0$, i.e. $2R_{\text{C}}$, also decreased from $3.5 \times 10^5 \Omega$ to $1.6 \times 10^5 \Omega$. In contrast, for the TFT with sputtered AlO_x passivation, r_{IGZO}/W was much smaller and maintained at around $30 \Omega/\mu\text{m}$, and the contact resistance decreased from $8.6 \times 10^3 \Omega$ to $3.6 \times 10^3 \Omega$. In Table III, r_{IGZO}/W and $R_{\text{C}}W$ under different gate voltage were summarized. These results clearly show that the AlO_x passivation layer reduced both r_{IGZO}/W and $R_{\text{C}}W$. The reduction of sheet resistance and contact resistance concurrently enhanced the TFT performance.

In the TLM model, transfer length L_{T} is also an important parameter, representing the distance over which most of the current transfers from source/drain into IGZO or vice versa. We can obtain the L_{T} value based on the following equation²⁰:

Based on Eq. 4, the value of $2L_{\text{T}}$ can be extracted from the intercept at $R_{\text{total}} = 0$ in Fig. 3. At $V_{\text{GS}} = 10$ V, for the TFT without passivation, $L_{\text{T}} = 8.0 \mu\text{m}$; for the TFT with AlO_x passivation, $L_{\text{T}} = 11.4 \mu\text{m}$. With the passivation, the length of current transfer path was increased. All the fabricated TFTs had the ITO(source/drain)/IGZO overlap distance $d = 20 \mu\text{m}$, which is significantly larger than the above L_{T} values of both un-passivated and passivated TFTs. In the above approach, we neglected the difference between the IGZO sheet resistance under the contact and the one in the channel. It should be considered in a more precise calculation.

The conduction mechanism of a-IGZO TFT can be explained by the percolation model.²¹ In the percolation model, there is a distribution of potential barrier height (~ 0.1 eV) over the extended states, largely influencing the carrier transport. The potential barrier heights could originate from the random distribution of Ga³⁺ and Zn²⁺.²² At room temperature, carriers would take a lower-barrier conduction path. When carrier concentration increases, the potential barrier heights are lowered, thus mobility and on-current are enhanced.²¹ The electronic structure and percolation model are shown in Figs. S5a and S5b, respectively, in Supporting Information.

As shown in Tables I and III, the TFT with sputtered AlO_x passivation layer showed higher on/off ratio by nearly one order, smaller V_{th} , much higher field effect mobility by over one order, much lower IGZO layer resistance and lower contact resistance. Nevertheless, at the same time, S.S. slightly increased for the TFT with passivation and a “hump” was also observed in the transfer curve. These obvious performance changes suggest that there could be donor-like defects at the IGZO/AlO_x interface (i.e. interface 2 in Fig. 1a). As shown in Fig. 2b, the as-fabricated TFT without post-

Figure 3. TFT TLM curves under different applied gate voltages: (a) without passivation layer (PVL) and (b) with AlO_x passivation layer (PVL).

Table III. Comparison of $\frac{\eta_{\text{IGZO}}}{W}$ and R_{CW} (channel width $W = 80 \mu\text{m}$) between the TFTs with and without the AlO_x passivation layer (PVL).

		6 V	7 V	8 V	9 V	10 V
$\frac{\eta_{\text{IGZO}}}{W} (\Omega\mu\text{m}^{-1})$	without PVL	2.5×10^3	1.9×10^3	1.5×10^3	1.2×10^3	1.0×10^3
	with PVL	36.6	32.1	30	28.7	27.8
$R_{\text{CW}} (\Omega \text{ cm})$	without PVL	282	218	178	151	132
	with PVL	68.8	51.2	40.8	33.6	28.8

deposition annealing could not be switched off, indicating existence of excessive free carriers due to the donor-like defects at the interface. Thus, post-deposition annealing was necessary for reducing the concentration of the donor-like defects to a reasonable level. The mechanism for the lower contact resistance of the TFT with the passivation layer is not clear yet. One possible explanation is given in the following. As demonstrated in Ref. 23, lateral free carrier diffusion occurring due to the gradient of carrier concentration in the IGZO layer in the edge region between the source/drain contact and the channel significantly affects the contact resistance. In the present study, the deposition of the passivation layer produced excessive free carriers in the exposed region of the IGZO layer. During the post-deposition annealing, the free carriers could laterally diffuse into the source/drain contact region of the IGZO layer, leading to reduction in the contact resistance.²³

The curves of the field effect mobility μ_{FE} vs. applied gate voltage V_{GS} for the TFTs without the AlO_x passivation layer and with the passivation layer are shown in Figs. S4a and S4b in Supporting Information, respectively. Without passivation, the mobility of the TFT gradually increased with applied gate voltage and became saturated at V_{GS} of $\sim 8 \text{ V}$ (a slight decrease can be observed at V_{GS} of $\sim 10 \text{ V}$), which can be well explained by the percolation model. Whereas in the passivated TFT, the mobility rapidly increased following by a gradual decrease with increasing V_{GS} with the maximum mobility occurring at V_{GS} of $\sim 4.5 \text{ V}$. It is suspected that the gradual decrease in the mobility of the passivated TFT might be related to the change in contact resistance. As the gate voltage increased, the actual voltage drop across the channel region might change. According to Eq. 2, substituting the fixed V_{DS} by an actual channel voltage drop could change the trend of mobility at high gate-field. To verify the impact of contact resistance, we chose five feature points ($V_{\text{GS}} = 6, 7, 8, 9$ and 10 V) to calculate the actual voltage drop across the channel and then recalculated the mobility. The calculated mobilities corresponding to the V_{GS} values are $162.67, 140.80, 117.66, 108.64$ and $94.56 \text{ cm}^2 \text{ Vs}^{-1}$, respectively.

There was still a large decrease in the mobility. It was found that the decrease of transconductance g_{m} had a more significant impact than the change of actual channel voltage drop. The mechanisms responsible for the decrease of transconductance (which means decrease of actual mobility of the passivated device at high V_{GS}) are not clear yet. Two possible mechanisms are explained in the following. As shown in Fig. 1a, the passivated TFT could have two conduction channels, i.e., the channels at interface 1 and interface 2, respectively; and both channels contributed to the drain current. Deposition of the AlO_x layer by sputtering process could induce charges at interface 2 or other charged defects in the IGZO layer, which might cause decrease of the mobility in the conduction channel at interface 1 at high V_{GS} for the passivated device. On the other hand, the sputtering process could increase the roughness of the IGZO back surface, which would enhance the surface scattering. The enhanced surface scattering at interface 2 at high gate fields could lead to a decrease of the mobility in the conduction channel at interface 2.^{24,25} Decrease of mobility of one of the channels or both channels would lead to decrease of the mobility of the passivated device at high V_{GS} .

As both channels contributed to the drain current, the passivated TFT had lower IGZO layer resistance as compared to the TFT without passivation. The channel at interface 2 had a lower turn-on voltage compared to the channel at interface 1 because the channel region at interface 2 had excess free carriers. Thus, the threshold voltage of the passivated TFT was lower than that of the TFT without passivation. The existence of the two conduction channels could also explain the hump behavior observed in the transfer curve,^{26,27} and the defect states at interface 2 could be responsible for the increase in the subthreshold swing.

Previous studies have proposed that ion bombardment during the AlO_x sputtering process could break the In–O bond in the IGZO.^{28,29} Ar plasma has also been shown to effectively break the In–O bond with the Ar ion bombardment in high sputtering field.³⁰ In order to further investigate the ion bombardment effect, the TFTs passivated

Figure 4. (a) Transfer curves measured at $V_{\text{DS}} = 1 \text{ V}$ of the TFTs passivated by AlO_x sputtered at different Ar gas flow rates (Note: The transfer curve at 120 sccm is the same as that in Fig. 2c); (b) HR-TEM image of the cross sections of the TFT passivated by AlO_x .

by 20 nm sputtered AlO_x layer with different Ar flow rate (25 sccm, 50 sccm, 75 sccm, 100 sccm and 120 sccm) were fabricated. Transfer curves of these devices were measured at $V_{\text{DS}} = 1$ V and shown in Figs. 4a. Thereinto, the TFT passivated by AlO_x sputtered at 120 sccm Ar flow rate is identical to the one shown in Fig. 2c. Figure 4a clearly shows that ion bombardment had a significant impact on the electrical performance. As shown in Fig. 4a, no “hump” is observed in the transfer curves of those passivated by the AlO_x deposited at low Ar flow rates. Adversely, the devices passivated by the AlO_x deposited at the low Ar flow rates of 25, 50 and 75 sccm showed a lower on-current compared with the unpassivated device. A low Ar flow rate could facilitate the oxygen ions from AlO_x to penetrate into the IGZO layer, and the oxygen ions reduced the free carrier (or oxygen vacancy V_{O}) concentration in the IGZO layer. On the other hand, as discussed later, deposition of AlO_x onto IGZO by sputtering could produce oxygen vacancies in the IGZO layer due to the ion bombardment. A low Ar flow rate led to a low level of ion bombardment, which means a low free carrier concentration at interface 2. Thus, the TFTs fabricated with low Ar flow rates had lower on-currents.

The existence of the interfacial layer at interface 2 induced by the ion bombardment could be proven from the HR-TEM (JEOL 2010) cross-section imaging, as shown by the arrow-highlighted region in Fig. 4b.

In Fig. 4b, the sputtered AlO_x layer was not as smooth as other amorphous layers. Thus, we conducted an X-ray Diffraction (XRD) for a layer of sputtered AlO_x on Si substrate, aiming to clarify the crystalline state of the sputtered AlO_x . The results are shown in Figs. S6 and S7. The XRD pattern in Fig. S6 shows that the AlO_x film has an amorphous nature due to the lack of sharp diffraction peak. At the same time, the absence of long-range order in the sample can also be verified from the grazing-incidence wide-angle scattering (GiWAXS) pattern, in which neither ring nor spot, respectively associated with polycrystalline and single crystalline system, could be observed in the 2D diffractogram (Fig. S7). Hence, the sputtered AlO_x was at amorphous state. The grainy structure of the AlO_x film in TEM image could be resulted from the electron beam induced crystallization during the lamella preparation in focused ion beam microscope (FIB).³¹ Some studies showed that oxygen species could easily diffuse into the amorphous alumina.^{32,33}

In order to further analyze the formation of an intermixing region between AlO_x and IGZO, we also utilized a combination of both Auger Electron Spectroscopy (AES) and X-ray Photoelectron Spectroscopy (XPS) depth profiling techniques to study the change in the elemental composition and the chemical states with depth.

As shown in the AES depth profile (Fig. 5), there was an obvious hump for the indium profiling in the interfacial layer, which suggests that the AlO_x -IGZO interface was indeed rich in In. Besides, the O/Al ratio increased compared with the bulk AlO_x , indicating both oxides could have changed chemically within the interfacial layer.

To further investigate the change in the chemical states with depth, Fig. 6 shows the In 3d, O 1s and Al 2p XPS spectra from the AlO_x /IGZO stacks. As seen in the In 3d spectra (Fig. 6a), with the etching cycle increasing, two additional minor peaks positioned at around 444 eV and 451.5 eV, respectively, (i.e., the regions highlighted by the red dotted lines), emerge; and they disappear when the etch cycle further increases. They are attributed to the metallic indium generated at the AlO_x /IGZO interfacial region during the sputtering deposition of AlO_x .^{34,35} This phenomenon agrees with what TEM image (Fig. 4b) shows. The transition region is also characterized by a slight increase in the concentration of the oxygen vacancy (V_{O}), which was obtained via deconvolution of the O 1s spectra, compared to the bulk of the IGZO layer. The detailed fitting parameters like binding energy (BE) peak and FWHM for O1s depth profiling are shown in table S2 in Supporting Information. The higher concentration of V_{O} correlates with higher charge-carrier concentration, giving rise to the improvement in the device performance. At the same time, the O 1s BE peak of AlO_x slightly shifted in the negative direction relative to that of the interface, as

shown by the two vertical red lines in Fig. 6c. It has been reported that O 1s peak would negatively shift when the O/Al ratio increases in AlO_x .³⁶ The Al 2p XPS spectra for the AlO_x /IGZO stack for various etching cycles are shown in Fig. 6b. When the etching cycle goes to 14, the detection goes into the bulk of IGZO, and thus the Al 2p peak disappears. For the Al 2p, as shown in Fig. 6b, the XPS peak energy positively shifted by around 1 eV. For comparison, a layer of 20 nm AlO_x was directly sputtering-deposited on Si substrate. In this AlO_x -Si stack, the Al 2p spectra showed nearly no shift, as shown in Fig. S8 in Supporting Information, due to the absence of the IGZO- AlO_x interface region. Positive shift of Al 2p BE indicates a higher oxidation level,³⁷ and a transition from metastable to stable state.³⁸

Thus, it could be concluded that when depositing amorphous AlO_x onto IGZO by sputtering, oxidation/reduction reactions happened at the interface. Since Al_2O_3 has lower enthalpy of formation (-1675.7 kJ mol⁻¹) than In_2O_3 (-925.9 kJ mol⁻¹), Al ion from the Al_2O_3 target captured oxygen from IGZO, which was encouraged by the ion bombardment. Thus, In-O bond in the IGZO layer was broken, generating metallic indium (i.e. oxygen vacancies) in the IGZO layer. Simultaneously, the deposited amorphous AlO_x might accommodate the free O^{2-} diffusing from the IGZO layer. The metallic-In-rich layer was formed at the interface and provided more mobile carriers. In the percolation model, higher carrier concentration lowered the potential barrier heights over the mobility edge, leading to higher carrier mobility for the IGZO TFT.

Besides the electrical performance, reliability of the TFT passivated by sputtered AlO_x was also investigated. Firstly, the AlO_x passivated TFT was stored in air in a lab with well-controlled air conditioning at room temperature with humidity of 70% ~ 71% for 6 m. The transfer curves were measured and shown in Fig. 7b. The maximum mobility of the device slightly decreased to 54.09 cm² Vs⁻¹ as shown in Fig. S4c in Supporting Information. The V_{th} of the TFT had nearly no shift after exposing in air for 6 m (Fig. 7b), which is in contrast to the situation of the TFT without passivation (Fig. 7a). It indicates that the AlO_x passivation layer has effectively suppressed the penetration of the ambient molecules (water vapor and O_2 , etc.). As illustrated in Fig. 8a, with existence of the passivation layer, oxygen, moisture, hydrogen, and other gases failed to diffuse into the channel and affect the channel property. There is still a hump in the semi-log $I_{\text{DS}}-V_{\text{GS}}$ curve but not so remarkable compared with the initial curve. During the 6-month exposure in air, some free metallic In and oxygen ions recombined with each other in the interface layer at interface 2. The contribution of the channel at interface 2 was suppressed such that the hump was slightly alleviated. To investigate the variation of the TFT performance, the transfer curves of 10 passivated TFTs after exposing in air for 6 m were measured. The deviations of the on current, field-effect mobility, threshold voltage and subthreshold swing were summarized in table S1 in Supporting Information.

Subsequently, tests under positive bias illumination stress (PBIS) and negative bias illumination stress (NBIS) were carried out. The light source used for both PBIS and NBIS measurements was a 21 W 150 W tungsten halogen lamp (GX5.3, OSRAM SYLVANIA). The wavelength range of the lamp is 360–2500 nm. For PBIS, the gate of the device was biased at 7 V with illumination of optical power of 2.65 mW from a halogen light source for various durations. Figures 9a and 9b show the transfer curves (in semi-log scale) after PBIS with various stress durations for the devices without the passivation layer and with the passivation layer, respectively. The corresponding linear transfer curves under PBIS are shown in Figs. S9a and S9b in Supporting Information, respectively. V_{th} shift versus stress time under PBIS is shown in Fig. S10 in Supporting Information. As can be observed from Fig. S10, the passivation layer greatly reduced the V_{th} shift. For example, for the stress duration of 5000 s, the V_{th} of the TFT without passivation layer positively shifted by 1.02 V; in contrast, the V_{th} of the TFT passivated by the AlO_x passivation layer positively shifted by only 0.12 V. It was suggested that the adsorbed oxygen molecules could act as acceptor-

Figure 5. AES depth profiling of In, O, Zn, Ga and Al atoms in the TFT passivated by the sputtered AlO_x layer.

like states, which capture electrons leading to a positive V_{th} shift.^{39,40} As shown in Fig. 8b, the passivation can effectively prevent adsorbing O_2 molecules from the ambient, and thus the PBIS reliability is improved.

For NBIS, the gate of the device was biased at -7 V with illumination of optical power of 2.65 mW for various durations. Figure 9c and 9d show the transfer curves (in semi-log scale) after NBIS of the device without the passivation layer and the device with the passivation layer, respectively. The corresponding linear transfer curves under NBIS are shown in Figs. S9c and S9d in Supporting Information, respectively. V_{th} shift versus stress time under NBIS is shown in Fig. S11 in Supporting Information. For the stress duration of 5000 s, the V_{th} of the TFT without passivation layer showed a negative shift of 3.8 V, while the V_{th} of the passivated TFT showed a negative shift of 3.96 V. It can be observed from Fig. S11 that NBIS could cause a large negative V_{th} shift for the TFTs with and without the passivation layer but there was no large difference in V_{th} shift caused by NBIS between the passivated TFT and non-passivated TFT. We also conducted NBS test (i.e., -7 V was applied to the bottom gate for various durations without illumination) on the TFTs with and without the passivation layer. The results are shown in Fig. S12 in the Supporting Information. It is obvious from Fig. S12 that both TFTs had a negative V_{th} shift but the value was much smaller than that under NBIS. Thus, we could conclude that the illumination was the main cause of the large V_{th} shift. It was suggested that under NBIS, the oxygen vacancies (V_o) in the interface region at the gate insulator/IGZO channel layer interface (i.e., interface 1 in Fig. 1a) could be photon-excited and become V_o^{2+} , and the created V_o^{2+} states could cause a large negative threshold voltage shift.¹¹ As the passivation layer would not have a large impact on the oxygen vacancies at interface 1, there should be no large difference in V_{th} shift after NBIS between the passivated TFT and non-passivated TFT. In other words, the passivation layer was not able to reduce the NBIS effect. To suppress the NBIS effect, a light shielding layer could be used. Besides, in order to reduce the NBIS instability, the channel layer should be modified to lower the valence band maximum (VBM) and maintain the large conduction band

dispersion, aiming to prevent the generation of the photon-excited electron-hole pairs and keep high mobility at the same time.⁴¹

Conclusion

In this work, a transparent a-IGZO TFT passivated by the sputtering-deposited AlO_x layer was fabricated. The mobility and on/off current ratio of the TFT were enhanced by nearly one order, i.e., the maximum electron mobility increased from 6.292 $\text{cm}^2 \text{Vs}^{-1}$ without the passivation layer to 69.01 $\text{cm}^2 \text{Vs}^{-1}$ with the passivation layer; and the on/off current ratio increased from 1.3×10^7 without the passivation layer to 1.87×10^8 with the passivation layer. Such a large improvement resulted from the interfacial layer formation at the IGZO/ AlO_x interface. The origin of the interfacial layer was further verified. The ion bombardment during the sputtering deposition of the AlO_x layer could prompt Al ion to capture oxygen from the IGZO layer and broke the In–O bonds; and at the same time, the deposited AlO_x layer might accommodate the free O^{2-} diffusing from the IGZO layer. Then a metallic-In-rich layer was formed at the interface with high carrier concentration. Besides, the passivated TFT showed better reliability under exposure in air for 6 m, and much smaller V_{th} shifts under PBIS. However, the passivation layer had no significant influence on the NBIS instability.

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ORCID

Yuanbo Li <https://orcid.org/0000-0002-0582-4290>
Rongyue Liu <https://orcid.org/0000-0002-0329-2411>

Figure 6. (a) Waterfall plot of In 3d XPS spectra for the AlO_x/IGZO stack with etching cycles in 2D format (Intensity vs Etching cycles); (b) Waterfall plot of Al 2p XPS spectra for the AlO_x/IGZO stacks with etching cycles in 2D format; (c) O 1s XPS peaks measured in the bulk of the AlO_x layer, IGZO-AlO_x interface region, and bulk of the IGZO layer, respectively. The two red vertical lines show the slight shift of O 1s peak of the interface region relative to the bulk of the AlO_x layer; and (d) depth profiling of O-related species.

Figure 7. Transfer curves (measured at $V_{\text{DS}} = 1 \text{ V}$) of the TFTs without passivation (a) and with the AlO_x passivation layer (b) after 6 m of exposure in air. Note: The initial curves were both the same as the transfer curves shown in Fig. 2c.

Figure 8. Schematic illustration of the mechanisms of reliability improvement by the passivation layer for the two scenarios: (a) exposure to air (O_2 , H_2 , water molecules, etc.); and (b) under PBIS.

Figure 9. Transfer curves after PBIS for the TFT without passivation layer (PVL) (a) and the TFT with passivation layer (PVL) (b); and transfer curves after NBIS for the TFT without passivation layer (PVL) (c) and the TFT with passivation layer (PVL) (d).

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