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Stable ZnO thin film transistors by fast open air atomic layer deposition

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We report stable, high performance zinc oxide thin film transistors grown by an atmospheric pressure atomic layer deposition system. With all deposition and processing steps kept at or below 200 °C, the alumina gate dielectric shows low leakage (below 10^{-8} A/cm^2) and high breakdown fields. Zinc oxide thin film transistors in a bottom gate geometry yield on/off ratios above 10^8, near zero turn-on voltage, little or no hysteresis, and mobility greater than 10 cm^2/V s. With alumina passivation, shifts in threshold voltage under gate bias stress compare favorably to those reported in the literature. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2924768]

Oxide-based thin film transistors (TFTs) have emerged as a promising technology, particularly for display applications.[1] ZnO-based devices have been produced by sputtering,[2,3] pulsed laser deposition,[4] solution process methods,[5,6] and atomic layer deposition (ALD).[7] Of these methods, only the solution methods can be done in an open atmosphere, although the resulting devices typically exhibit low mobility. Following initial interest in ZnO semiconductors, some groups have turned to ternary and quaternary zinc compounds to ensure an amorphous semiconductor for performance and stability.[8–10]

In this work, we describe ZnO TFTs with device characteristics comparable to the best in the literature and with improved stability, grown by an open atmospheric pressure system. This growth system uses a fast ALD method that could change the manufacturing methods and economics for oxide-based electronics. While potentially all the electrical layers in a TFT could be grown by the same system, this paper focuses on devices with the gate dielectric and semiconductor layer deposited by this system.

Atomic layer depositions are typically done in an enclosed system, where the reactive gases are sequentially cycled through the chamber in time. The alternative used here is an ALD process employing spatial isolation of reactive gases (spatial ALD). In this process, gases are isolated to regions of the coating head, and the cyclic ALD process results from movement of these regions over the substrate. Figure 1(a) depicts the stainless steel ALD coating head positioned approximately 30 μm above a substrate. The head contains recessed channels facing the substrate that are closed at one end (not shown), and gas introduced near the closed end flows along its channel as shown in the figure. An inert gas (I) and two reactive gas streams (A: oxidizer gas) and (B: metal source) flow in these channels, as shown by the designations in Fig. 1(a). In our case, the inert gas is nitrogen, which also acts as a carrier gas for the metal and oxidizer precursors. The channel shape and proximity to the substrate prevent mixing between neighboring channels and exclude the surrounding air.

If the head is stationary, no deposition occurs because each part of the substrate is contacted by only one gas. However, as the substrate moves relative to the head, regions of the substrate see the alternating ALD gas cycles. We oscillate the sample to produce alternating gas cycles to achieve the desired layer thickness. In situations where speed of deposition is critical, a larger deposition head could enable a single pass of the substrate to deposit the full thickness. Our current laboratory transport rates would give an impressively fast substrate throughput of 3 m/min, which is distinct from the batch-processing times normally associated with ALD.

To compare with conventional ALD, the film growth saturation behavior of Al_2O_3 was studied. Films were grown on bare silicon wafers at substrate speeds corresponding to channel residence times of 37, 56, 112, and 448 ms. The oxidizer channels (A) contained 0.31 mol % water, 0.89 mol % oxygen, and 3.1×10^{-3} mol % H_2O_2, while the metal source channels (B) contained 0.062 mol % trimethylaluminum. From film thickness measured by ellipsometry as

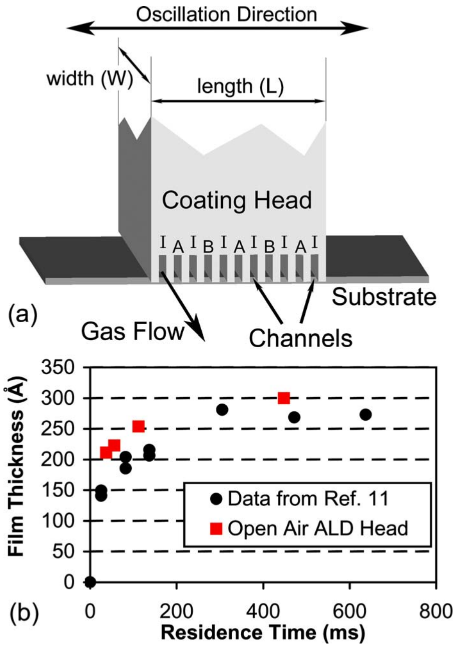

FIG. 1. (Color online) (a) Schematic of the ALD coating head showing the gas channels and gas flow. The channels are 0.7 mm wide with a spacing of 1.4 mm. The coating width W is approximately 50 mm. A is the oxidizing reactant, B is the metal precursor, and I is nitrogen. (b) Al_2O_3 film thickness for 250 ALD cycles as a function of residence time. The squares represent data from the spatial ALD system operated at 177 °C. The circles represent data extracted from Ref. 11.

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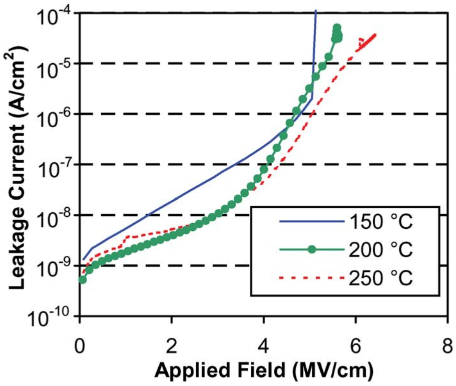

FIG. 2. (Color online) Leakage current as a function of applied electric field for Al_2O_3 films grown at several temperatures. The 200 and 250 °C films show low leakage and are distinctly better than the 150 °C film.

a function of residence time, we find that the monolayer saturation behavior is comparable to chamber-based ALD,[11] with a maximum growth rate of approximately 1.2 Å/cycle [Fig. 1(b)].

Current leakage and dielectric breakdown were measured for Al_2O_3 films deposited on chromium at 150, 200, and 250 °C, with thicknesses of 987, 1243, and 1229 Å, respectively. Aluminum metal contacts measuring 90×130 μm were deposited by shadow mask evaporation onto the Al_2O_3 surface to form test structures.

Figure 2 shows a plot of applied electric field versus area-corrected leakage current for these devices. The leakage density at a field of 1 MV/cm is 5.4×10^{-9}, 1.89×10^{-9}, and 2.78×10^{-9} A/cm^2 for the 150, 200, and 250 °C samples, respectively. Furthermore, these devices always show breakdown voltages in excess of 5 MV/cm. The leakage currents are within an order of magnitude of the best of those summarized in Ref. 12 and also compare favorably to ALD alumina films grown as gate dielectrics for zinc oxide TFTs.[13] The dielectric constant of the alumina was determined from capacitance-voltage measurements to be between 7.5 and 8.5, within the range of published values for Al_2O_3 deposited by ALD.[12]

The TFT devices were made on a glass substrate coated with chromium or ITO as the gate contact. An Al_2O_3 film of approximately 1100 Å was deposited at 200 °C, followed by a 250 Å zinc oxide layer produced using 21 mol % NH_3, 0.64 mol % water, and 0.74 mol % oxygen in the *A* channels, and a 0.52 mol % diethylzinc in the *B* channels. The microcrystalline nature of the ZnO layer has been determined by cross-sectional transmission electron microscopy (TEM). Lattice images (not shown) of the ZnO layer show a variety of orientations of sub-100-nm-sized grains. X-ray diffraction (XRD) from a 330-nm-thick ZnO (Fig. 3) shows primarily (100) orientation, while XRD from thinner layers show about equal intensity in the (100) and (002) peaks (Fig. 3 inset, for a 60-nm-thick ZnO layer.) Grazing incidence XRD confirms a similar structure in the thin (~25 nm) layers used for TFTs. This suggests that (100) is the preferred orientation for this deposition method,[14] although interactions with the surface influence the initial grain orientation.

For TFTs, top aluminum or titanium/gold contacts were patterned by lift off, and devices were isolated using photolithographically patterned poly(methyl methacrylate) to protect the channel. Figure 4(a) shows the double-sweep transfer characteristics of a typical ZnO TFT in the linear regime (V_d=0.25 V) and in saturation (V_d=20 V). There is no visible hysteresis. The turn-on voltage of this device is −0.5 V, with saturation threshold voltage of 4.8 V. The saturation mobility, extracted by a fit to the square root of drain current versus gate voltage from the V_d=20 V, sweep is 12.9 cm^2/V s, while the linear mobility extracted from the 0.25 V_d curve is 12.5 cm^2/V s. The close match between linear and saturation values indicates that excellent contact to these devices is possible with a simple evaporation process. The highest mobility we have reproducibly seen is about 20 cm^2/V s. Mobility of thin film epilayers of ZnO (Ref. 15) is typically below 100 cm^2/V s, which can be taken as an upper limit for these polycrystalline layers. In addition to the dc performance mentioned above, devices produced by this method exhibit the highest frequencies to date for oxide based ring oscillator circuits.[16]

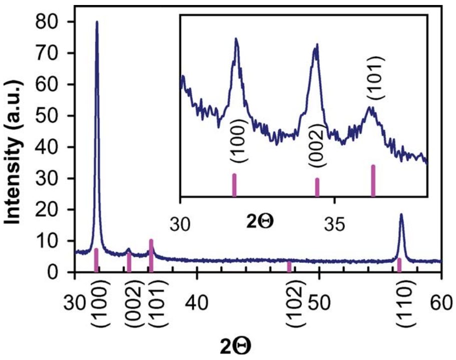

FIG. 3. (Color online) XRD of 330-nm-thick ZnO layer with inset of 60 nm ZnO. For the thicker sample, the primary peak is found at (100), while in thinner samples, the (002) peak becomes equally intense.

Changes in threshold voltage with bias stress were measured. Gate bias was held at 20 V during stress, with inter-

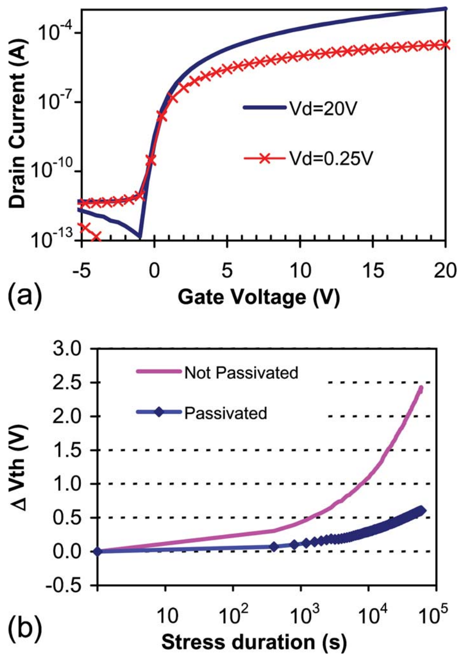

FIG. 4. (Color online) (a) Double sweep transfer characteristics of a zinc oxide TFT showing no hysteresis at V_d=0.25 V and V_d=20 V. Linear and saturation mobility are 12.5 and 12.9 cm^2/V s respectively. (b) Threshold voltage shift under gate bias stress of 20 V for an unpassivated TFT, and one passivated with 100 nm Al_2O_3. The change in mobility over the 16 h is less than 5% for both samples.

ruptions every 400 s for a linear region I_d-V_g sweep. With no passivation of the ZnO, the threshold voltage shift is 2.4 V in 16 h. However, a passivation layer of 100 nm of Al_2O_3, deposited using the spatial ALD process, dramatically improves the bias stability. Passivated devices show positive threshold voltage shifts of 0.6 V for 16 h stress [Fig. 4(b)], and less than 5% change in mobility. These shifts are some of the lowest reported for ZnO based TFTs and compare favorably with values obtained from amorphous multication systems[17,18] while maintaining the simplicity of a pure ZnO semiconductor. Further work to understand the origin of the shifts is underway.

In summary, we have demonstrated promising ZnO TFTs grown in an open air ALD deposition system. The alumina dielectric layer produced has comparable performance to known dielectrics produced by ALD, while resulting ZnO TFTs exhibit excellent mobility and stability characteristics. We believe that this deposition system will be adaptable to very large format, continuous, high throughput processing.

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