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Defect Passivation by Hydrogen in Zinc Oxide Films Grown by MOCVD

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I. Introduction

Zinc oxide (ZnO) has attracted considerable attention for its utilization in display and optoelectronic devices due to high mobility and wide bandgap. Sputtering, laser-ablation, and metalorganic chemical vapor deposition (MOCVD) have been used to grow ZnO. Although sputtering showed better on/off characteristics, MOCVD has certain advantages, such as the size of the substrate. The main obstacle at this point for MOCVD is that the leakage current in ZnO thin-film transistor (TFT) is larger than that grown by sputtering. It appears that the gas incorporated during MOCVD growth acts as source of the leakage current.

ZnO usually shows n-type behavior, and it is believed that hydrogen and other defects are the origin of n-type behavior [1]. The exact nature of hydrogen in ZnO is still under debate. In our previous work [2], we showed that MOCVD ZnO films have significant amount of hydrogen and carbon concentrations, even without any additional hydrogen flow. The origin of hydrogen and carbon in our previous work must be diethylzinc (DEZ), because it is the only material containing them. In this work, we studied the effect of hydrogen intentionally added during MOCVD growth. By controlling growth parameters, we obtained $2x10^6$ on/off ratio in ZnO TFT with 10 cm^2/Vsec mobility. We explain that the high on/off ratio is related to hydrogen passivation.

II. Experimental Results

Our MOCVD system has a horizontal reactor operating at atmospheric pressure. ZnO films were grown at temperatures between 200 and 400℃, because we wanted to develop technologies applicable for glass substrates. For ZnO sources, DEZ and O_2 were used. DEZ was fed through a bubbler kept at -10℃, with a N_2 flow of 20 sccm. N_2 was employed as a carrier gas with a flow of 2000 sccm, and O_2 flow was 160 sccm. H_2 flow of 300 sccm was added to the carrier gas. ZnO films were grown on silicon substrates, which have 1100Å thick oxide. Figure 1 shows photoluminescence data measured at room temperature. The sharp bandgap peak and lack of defect peaks indicate that our ZnO film has good quality. After MOCVD growth, we fabricated TFT structure (channel length= 15 μm, width= 500 μm) by using Al evaporation and shadow mask. Figures 2 and 3 are current-voltage characteristics of TFT grown at 300℃.

We investigated if the additional hydrogen changes composition in our ZnO films. Secondary Ion Mass Spectroscopy (SIMS) and x-ray Photoelectron Spectroscopy (XPS) did not show any change in zinc or oxygen composition (not shown here). Therefore, any changes observed in transport properties are from microscopic origins. We compared the effect of the added hydrogen in many samples. In general, the hydrogen increased TFT mobility, and we think that defect passivation is the main reason for this increase. However, the most important effect of hydrogen appeared in the on/off ratio.

Figure 2 shows gate voltage dependence of drain currents measured in two samples, (a) one grown without hydrogen, and (b) the other grown with hydrogen. In fresh samples, mobility and on/off ratio in the two samples were comparable. After 20 days in air, the one grown without hydrogen degraded severely. It looks like that the grain boundary in ZnO film adsorbs oxygen, to make current transport more difficult. In contrast, the one grown with hydrogen showed much smaller degradation, as shown in Fig. 2b. Oxidation decreased leakage current, and on/off ratio increased to $2x10^6$ as a result.

Figure 3 is currents measured in the sample used in Fig. 2b. It shows mobility of 10 cm^2/Vsec, with threshold voltage of -5 V. The fact that oxidation decreases leakage current suggests that the extra zinc in our film is the reason of leakage. (Our XPS results showed that the Zn:O ratio in our films were around 55:45). At low growth temperatures of 200 to 400℃, increasing oxygen flow did not improve TFT performance, since the increased oxygen also contributed as defect sources. With these restrictions, adding hydrogen during MOCVD growth can be a good technology to grow ZnO films on glass substrates.

III. Conclusions

We demonstrated that high on/off ratio ZnO film can be grown by MOCVD, with help of additional hydrogen. The exact nature of hydrogen incorporation is not understood yet, but it appears that the hydrogen passivates defects during MOCVD growth. With this method, our TFT showed $2x10^6$ on/off ratio and 10 cm^2/Vsec mobility.

References

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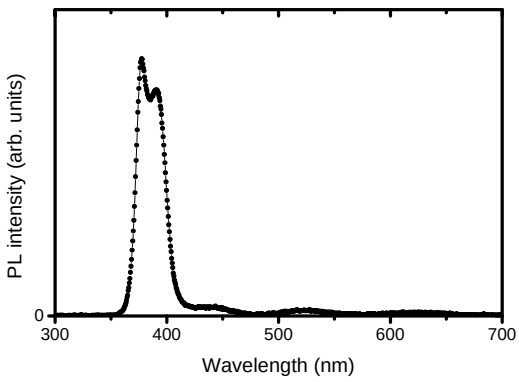

Fig. 1. Room-temperature photoluminescence data measured in ZnO film grown at 400°C.

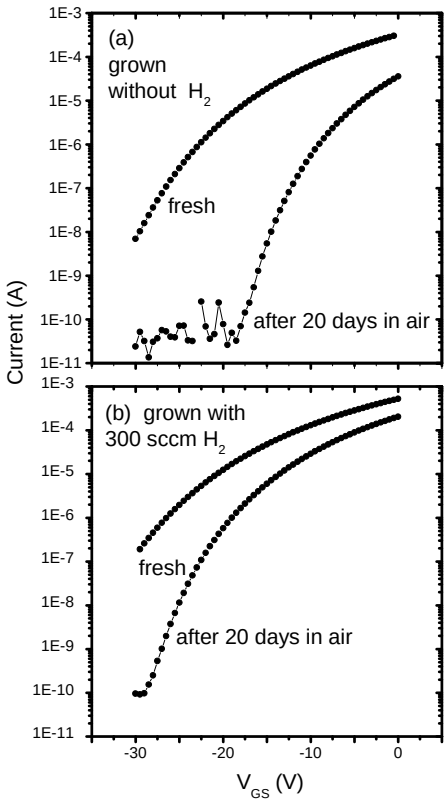

Fig. 2. Gate-voltage dependence of drain current (V_{DS} = 5 V) in samples grown without and with hydrogen. The sample grown with hydrogen shows stronger resistance against degradation.

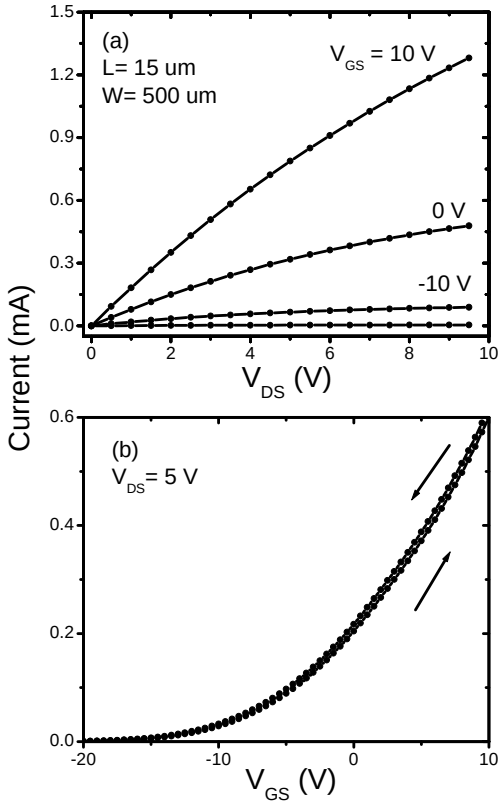

Fig. 3. Current-voltage characteristics of ZnO TFT, used in Fig. 2b. These data were measured after 20 days of air exposure. The lowest curve in (a) is for gate voltage of -20 V.