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High performance homo-junction field-effect transistor memory devices were prepared using solution processed transparent lithium-doped zinc oxide thin films for both the ferroelectric and semiconducting active layers. A highest field-effect mobility of $8.7 \text{ cm}^2/\text{Vs}$ was obtained along with an $I_{\text{on}}/I_{\text{off}}$ ratio of 10^6 . The ferroelectric thin film transistors showed a low sub-threshold swing value of 0.19 V/dec and a significantly reduced device operating voltage ($\pm 4 \text{ V}$) compared to the reported hetero-junction ferroelectric transistors, which is very promising for low-power non-volatile memory applications. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4729787>]

Ferroelectric thin film transistor (Fe-TFT) based memory devices have attracted much attention in recent years because of their potential advantages: non-volatility, non-destructive readout, low-power consumption, fast switching speeds, and the ability to operate without an access transistor which makes high density memories possible. The information storage in a Fe-TFT is accomplished by inducing a polarization in a ferroelectric film which in turn modulates the conductivity of the channel in a semiconductor substrate. Recently, hetero-structure Fe-TFTs have been reported using zinc oxide,^{1–6} indium tin oxide,^{7,8} indium zinc oxide,⁹ zinc tin oxide,¹⁰ and indium gallium zinc oxide^{11,12} based thin films as the active layers. In all these cases inorganic or organic materials such as lead zirconate titanate, barium titanate, and poly(vinylidene fluoride-trifluoroethylene) [P(VDF-TrFE)] have been used as the ferroelectric layer. Although P(VDF-TrFE) has relatively high remanent polarization and fast switching time, it requires large voltage for electrical switching because of its large coercive field of approximately $0.5\text{--}1 \text{ MV/cm}$. Further, the reported hetero-junction Fe-TFTs operate at a comparatively high voltage, which is not suitable for low-power non-volatile memory applications. One of the key challenges for Fe-TFTs is their poor retention performance. The poor retention can result from two dominant effects (1) the depolarization field which opposes the polarization vector and (2) interfacial defects. Because the ferroelectric material is different from the semiconductor material in all reported ferroelectric field transistors, interfacial defects often form at the interface between the ferroelectric and semiconductor layers. These defects can include interfacial reactions, elemental diffusions, and dislocations that originate from interfacial stresses (from lattice constant or thermal expansion coefficient mismatch). One approach to reduce these interfacial defects is to form a homo-junction Fe-TFT using the same material as the ferroelectric and semiconductor layer. Such a device has not been previously reported.

Transparent zinc oxide (ZnO) based thin film transistors (TFTs) have been demonstrated recently as an alternative to the low-mobility silicon-based TFTs for flat-panel display applications.^{13–15} Lithium has been proven to be a potential dopant to improve the performance and environmental stability of ZnO TFTs using simple solution process techniques.^{16,17} Moreover, the existence of spontaneous ferroelectric polarization in Li-doped ZnO (LZO) thin films has also been demonstrated.^{18–22} It is believed that ferroelectricity in LZO arises due to the large difference in ionic radii between the host Zn (0.74 Å) and the dopant Li (0.6 Å). The combination of good transistor performance and ferroelectric behavior of Li-doped ZnO oxide thin films motivated us to fabricate a homo-junction Fe-TFT that uses LZO for both the ferroelectric and active layers.

Zinc acetate dihydrate (85 at. %) and lithium chloride (15 at. %) were dissolved in 2-methoxyethanol to prepare the precursor solution for the spin coating of ferroelectric Li-doped ZnO thin films. Total concentration of the metal ions in the precursor solution was fixed at 0.5 M . Equal molar of ethanolamine was added for the stabilization of the precursor solution and the solution was stirred at 50°C in air for 1 h. Fresh solutions were used to make devices within two days of the solution preparation. Heavily doped p-type silicon ($\text{p}^+\text{-Si}$) wafers were used as the substrate as well as the bottom gate contact for the fabrication of ferroelectric capacitors and Fe-TFTs. The $\text{p}^+\text{-Si}$ substrates were ultrasonically cleaned using acetone, iso-propanol and then dried by nitrogen gas. The cleaned and dried $\text{p}^+\text{-Si}$ substrates were exposed to oxygen plasma for 2 min to increase the hydrophilicity and better adhesion of the precursor solution. The precursor solution was spun on cleaned $\text{p}^+\text{-Si}$ substrates at a speed of 3000 rpm for 30 s in air. After spin coating, the wet films were placed on a preheated hot plate at 80°C in air for 5 min and then dried at 300°C in air for 5 min. The spin coating and drying process were repeated several times to get films with desired thickness and finally the samples were subjected to slow thermal annealing at 500°C for 10 min in air. For the active layer, one layer of the LZO film was spin coated on top of the aforementioned slow thermally annealed

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LZO films under the same deposition conditions and then the LZO films were subjected to rapid thermal annealing at 500 °C for 1 h. Circular shape silver contacts (80 nm) with diameter of 100 μm for ferroelectric and capacitance measurements and source and drain electrodes with channel width and length of 250 and 40 μm , respectively, for the transistor characteristic measurements were deposited using shadow mask and e-beam evaporation. Thickness of the LZO films was measured by a Veeco Dektak 150 surface profilometer. The average thickness of the LZO film was found to be 210 nm. Crystallinity of the LZO films was studied by x-ray diffraction (XRD) diffractometer (Bruker D8 Discover) using CuK α radiation. The morphology of the LZO films was studied by an Agilent 5400 atomic force microscope (AFM) under tapping mode. Polarization hysteresis behavior was studied using a Premier Precision II ferroelectric tester (Radiant Technologies Inc.). Capacitance of the LZO films was measured by a Precision LCR meter (Agilent E4980A). The current-voltage characteristics of the LZO Fe-TFTs were performed using a semiconductor characterization system (Keithley 4200-SCS) and a Cascade Microtech (Summit-11600 AP) microprobe station.

The XRD pattern of the LZO film deposited on p⁺-Si substrate is shown in Fig. 1(a). The observed intense and

weak peaks corresponds to the (002) and (004) reflections of wurtzite phase of ZnO, respectively. The presence of intense (002) peak and the absence of other reflection peaks of ZnO indicates the preferential orientation of crystallites along the *c*-axis. The AFM image (inset of Fig. 1(a)) of the LZO film showed a root mean square surface roughness and average particle size of 9 and 78 nm, respectively. The UV-VIS transmittance spectra of the LZO film deposited on glass substrate (Fig. 1(b)) showed more than 80% transparency in the visible range of the electromagnetic spectrum. The optical band gap was determined by extrapolation of the linear region from the α^2 versus $h\nu$ plot (inset of the Fig. 1(b)) near the onset of the absorption edge to the energy axis. The band gap of the LZO film was found to be ~ 3.3 eV, which agrees well with the bulk band gap of ZnO.²³ The high transparency of the LZO film will be suitable for the realization of transparent memory devices.

The room temperature ferroelectric hysteresis curves measured at different applied voltages are shown in Fig. 2(a). The polarization curve exhibited a symmetric and well saturated curve with an applied voltage of ≥ 3 V, which confirmed the ferroelectric behavior of LZO film.²¹ A remanent polarization of 0.9 $\mu\text{C}/\text{cm}^2$ was obtained with a coercive field

FIG. 1. (a) XRD pattern of the LZO film deposited on p⁺-silicon substrate obtained in Bragg-Brentano geometry, and the inset shows the AFM image of the corresponding LZO film and (b) UV-VIS transmittance spectra of the LZO film deposited on glass substrate.

FIG. 2. (a) Hysteresis loops measured at different applied voltages and (b) capacitance vs voltage measured at a frequency of 1 kHz of the p⁺-Si/LZO/Ag device structure. The inset shows the leakage current density of the p⁺-Si/LZO/Ag device structure.

of $\sim \pm 0.07$ MV/cm. The coercive field obtained in the present work is significantly lower than reported values for inorganic oxide²⁴ and organic²⁵ based ferroelectric thin films; hence, LZO based ferroelectric thin films have potential for low-voltage applications. The capacitance vs voltage (C-V) curve measured at a frequency of 1 kHz is shown in Fig. 2(b). The presence of hysteresis in the C-V curve further confirms the ferroelectric behavior of the LZO film. A memory window of ~ 0.8 V was found from the CV curve which is very promising for memory applications. It is clearly seen from the C-V curve that at negative gate biasing voltages, the capacitance increases due to the accumulation of the substrate's majority carriers at the interface between the ferroelectric film and the p^+ -Si. At positive biasing voltages on the gate, the ferroelectric film is at higher potential than the silicon and substrate which depletes the silicon substrate from carriers near the interface, resulting in low capacitance. The choice of gate dielectric and the leakage current of the gate oxide are very important for TFT performances. The inset of the Fig. 2(b) shows the leakage current density of the p^+ -Si/LZO/Ag structure. A leakage current density of $\sim 10^{-7}$ A/cm² was observed for voltages below 4 V, thus making LZO as a promising candidate for low-voltage applications.

A schematic of the device structure, output, and transfer characteristics of the homo-junction Fe-TFT using LZO as ferroelectric and active layer are shown in Figs. 3(a)–3(c). The output characteristics curves showed a typical field-effect transistor behavior working in the enhanced mode of operation. The transfer characteristics curve of the LZO Fe-TFT devices was measured in dual sweep mode of gate-to-source voltage (V_{GS}) with a constant drain-to-source voltage (V_{DS}) of 1 V. When a bias voltage greater than the coercive voltage is applied onto the gate of the Fe-TFT, the dipoles

within the LZO ferroelectric layer are aligned to attract or to deplete the electrons in the LZO active layer, depending upon the gate bias polarity. The counterclockwise hysteresis in forward and reverse sweep of V_{GS} from the transfer characteristic suggests that the shift of the turn-on voltage of the Fe-TFTs is caused by the ferroelectric nature of the LZO thin film. The field-effect mobility (μ_{FE}) induced by the transconductance in the linear operation regime ($V_{DS} \leq 1$ V) was estimated using the following equation

$$\mu_{FE} = \frac{g_m}{WC V_{DS}}, \quad (1)$$

where g_m is the transconductance, L and W are the channel length and width, respectively, and C is the capacitance (37 nF/cm² measured at 1 MHz) per unit area of the LZO ferroelectric film. A field-effect mobility of 8.7 cm²/Vs was obtained with an I_{on}/I_{off} ratio of 10^6 . The field-effect mobility obtained in this work is superior to the previously reported results using inorganic/organic ferroelectric materials and ZnO semiconductors.^{1,2,4} It may be mentioned here that, the LZO Fe-TFT operates at a very low voltage (-4 V to $+4$ V) and also showed an I_{on}/I_{off} ratio of 10^4 at $V_{GS} = 0$ V, which is very promising for low-voltage memory applications. The LZO Fe-TFT exhibited a memory window of 0.9 V which is very close to the value of memory window obtained from the C-V characteristic curve of the p^+ -Si/LZO/Ag structure. In general, a low sub-threshold swing value is required for fast switching applications. The LZO Fe-TFT showed a low sub-threshold swing of 0.19 V/dec, which may be attributed to the reduction of defects due to the homo-junction of the LZO ferroelectric and the active layer interface. The retention curves of the homo-junction LZO FE-TFTs are shown in Fig. 3(d). A voltage pulse of 3 V

FIG. 3. (a) Schematic of device structure, (b) output characteristics, and (c) transfer characteristics of LZO homo-junction Fe-TFTs using LZO as ferroelectric and active layers; (d) Retention properties of on- and off-states of LZO Fe-TFTs measured under $V_{DS} = 1$ V and $V_{GS} = 0$ V for 2000 s.

or -3 V was applied for 1 s to the gate electrode (V_{GS}) to switch the LZO Fe-TFT to on or off state, respectively. The drain current (I_{DS}) was measured at $V_{DS} = 1$ V and $V_{GS} = 0$ V for both on- and off- states for 2000 s. Our preliminary results showed an on and off-state drain current ratio of ~ 50 throughout the measurement duration which is very close to the value reported using ZnO/P(VDF-TrFE) based devices.⁴ The lower on-off ratio of the on and off-state drain current may be due to the low polarization of the LZO ferroelectric film, resulting the fast decay of the on-current. The retention properties of the LZO Fe-TFTs can be improved by further optimization of the LZO film and/or by inserting of a suitable interfacial layer, which is under investigation. Nevertheless, it may be noted here that the fabricated homo-junction LZO based Fe-TFT has potential to be used in low-voltage nonvolatile memory applications.

In summary, homo-junction oxide based ferroelectric field-effect thin film transistors were prepared by sol-gel spin coating method. The device uses solution-processed, transparent Li-doped ZnO films for both the ferroelectric and active layers. Furthermore, the devices operate at much lower voltage compared to hetero-junction ferroelectric thin film transistors (± 4 V). A highest field effect mobility of $8.7 \text{ cm}^2/\text{Vs}$ was obtained along with an I_{on}/I_{off} ratio and sub-threshold swing of 10^6 and 0.19 V/dec , respectively. The obtained performances of the LZO Fe-TFTs are very promising for low-voltage non-volatile memory applications.

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¹H. Tanaka, Y. Kaneko, and Y. Kato, *Jpn. J. Appl. Phys.* **47**, 7527 (2008).

²M. Brandt, H. Frenzel, H. Hochmuth, M. Lorenz, M. Grundmann, and J. Schubert, *J. Vac. Sci. Technol. B* **27**, 1789 (2009).

³K. H. Lee, G. Lee, K. Lee, M. S. Oh, S. Im, and S. M. Yoon, *Adv. Mater.* **21**, 4287 (2009).

⁴C. H. Park, K. H. Lee, B. H. Lee, M. M. Sung, and S. Im, *J. Mater. Chem.* **20**, 2638 (2010).

⁵Y. Kaneko, Y. Nishitani, H. Tanaka, M. Ueda, Y. Kato, E. Tokumitsu, and E. Fujii, *J. Appl. Phys.* **110**, 084106 (2011).

⁶S. M. Yoon, S. Yang, and S. H. K. Park, *J. Electrochem. Soc.* **158**, H892 (2011).

⁷T. Miyasako, M. Senoo, and E. Tokumitsu, *Appl. Phys. Lett.* **86**, 162902 (2005).

⁸T. Miyasako, B. N. Q. Trinh, M. Onoue, T. Kaneda, P. T. Tue, E. Tokumitsu, and T. Shimoda, *Appl. Phys. Lett.* **97**, 173509 (2010).

⁹S. M. Yoon, S. H. Yang, S. W. Jung, C. W. Byun, S. H. K. Park, C. S. Hwang, and H. Ishiwara, *Electrochem. Solid. State Lett.* **13**, H141 (2010).

¹⁰S. M. Yoon, S. Yang, C. Byun, S. H. K. Park, D. H. Cho, S. W. Jung, O. S. Kwon, and C. S. Hwang, *Adv. Funct. Mater.* **20**, 921 (2010).

¹¹B. H. Kim, C. W. Byun, S. M. Yoon, S. H. Yang, S. W. Jung, M. K. Ryu, S. H. K. Park, C. S. Hwang, K. I. Cho, O. S. Kwon, E. S. Park, H. C. Oh, K. H. Kim, and K. C. Park, *IEEE Electron Device Lett.* **32**, 324 (2011).

¹²S. M. Yoon, S. Yang, M. K. Ryu, C. W. Byun, S. W. Jung, S. H. K. Park, C. S. Hwang, and K. I. Cho, *IEEE Trans. Electron Devices* **58**, 2135 (2011).

¹³P. F. Carcia, R. S. McLean, M. H. Reilly, G. Nunes, *Appl. Phys. Lett.* **82**, 1117 (2003).

¹⁴B. S. Ong, C. Li, Y. Li, Y. Wu, and R. Loutfy, *J. Am. Chem. Soc.* **129**, 2750 (2007).

¹⁵G. Adamopoulos, S. Thomas, D. D. C. Bradley, M. A. McLachlan, and T. D. Anthopoulos, *Appl. Phys. Lett.* **98**, 123503 (2011).

¹⁶P. K. Nayak, J. Jang, C. Lee, and Y. Hong, *Appl. Phys. Lett.* **95**, 193503 (2009).

¹⁷G. Adamopoulos, A. Bashir, S. Thomas, W. P. Gillin, S. Georgakopoulos, M. Shkunov, M. A. Baklar, N. Stingelin, R. C. Maher, L. F. Cohen, D. D. C. Bradley, and T. D. Anthopoulos, *Adv. Mater.* **22**, 4764 (2010).

¹⁸A. Onodera, N. Tamaki, Y. Kawamura, T. Sawada, and H. Yamashita, *Jpn. J. Appl. Phys.* **35**, 5160 (1996).

¹⁹M. Joseph, H. Tabata, and T. Kawai, *Appl. Phys. Lett.* **74**, 2534 (1999).

²⁰X. S. Wang, Z. C. Wu, J. F. Webb, and Z. G. Liu, *Appl. Phys. A* **77**, 561 (2003).

²¹Dhananjay, J. Nagaraju, and S. B. Krupanidhi, *J. Appl. Phys.* **99**, 034105 (2006).

²²Dhananjay, S. Singh, J. Nagaraju, and S. B. Krupanidhi, *Appl. Phys. A* **88**, 421 (2007).

²³P. K. Nayak, J. H. Yang, J. Kim, S. J. Chung, J. Jeong, C. Lee, and Y. Hong, *J. Phys. D: Appl. Phys.* **42**, 035102 (2009).

²⁴T. S. Bösccke, J. Müller, D. Bräuhäus, U. Schröder, and U. Böttger, *Appl. Phys. Lett.* **99**, 102903 (2011).

²⁵M. A. Khan, U. S. Bhansali, and H. N. Alshareef, *Org. Electron.* **12**, 2225 (2011).