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APPLIED PHYSICS LETTERS **87**, 043509 (2005)

Low-voltage ZnO thin-film transistors with high-*K* $Bi_{1.5}Zn_{1.0}Nb_{1.5}O_7$ gate insulator for transparent and flexible electronics

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We report on the fabrication of field-effect transistors with transparent oxide semiconductor ZnO serving as the electron channel and high-*K* $Bi_{1.5}Zn_{1.0}Nb_{1.5}O_7$ (BZN) as the gate insulator. The devices exhibited very low operation voltages (<4 V) due to high capacitance of the BZN dielectric. The field effect mobility and the current on/off ratio were 0.024 $cm^2/V\,s$ and 2×10^4, respectively, at an operating voltage of 4 V. The threshold voltage and subthreshold swing were 2 V and 0.25 V/dec, respectively. The high optical transparency (>80% for wavelength >400 nm), low-temperature processing, and low operation voltage of ZnO-based thin-film transistors with integrated BZN dielectric offer a promising route for the development of transparent and flexible electronics. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1993762]

Transparent electronic circuits offer the opportunity to create new optoelectronic devices and applications.[1] The transparent field-effect transistor (FET) is a key device for realizing transparent circuits. Compound semiconductors with wide band gaps (>3.1 eV) are transparent in the visible spectrum and can therefore serve as the channel layer of transparent FETs. Most recent research has focused on improving the field-effect mobility of large gap compound semiconductors, by improving the quality of the channel materials going as far as using single crystals.[2,3] Generally, these approaches require high-temperature processes which go contrary to another important research trend, i.e., near room temperature processing to enable fabrication of active devices (e.g., FETs) on flexible polymer substrates. Organic semiconductors are attractive as the channel layer from this standpoint. However, organic transistors show poor performance and degradation when exposed to air or light illumination. In contrast, inorganic semiconductors, including transparent compound semiconductors, tend to remain stable even when exposed to either high temperatures or illumination.

Zinc oxide (ZnO) based transparent thin film transistors (TFTs) have been studied intensively as transparent FETs due to their potential of replacing hydrogenated amorphous or polycrystalline silicon (*a*-Si:H or poly-Si) TFTs that now serve as the backplane for active matrix displays such as liquid crystal displays and organic light emitting diodes. ZnO is a transparent compound semiconductor with a wide band gap (3.37 eV) which can be grown as a polycrystalline film at low or even room temperature.[4] ZnO is therefore considered to be an ideal material for serving as the channel layer in transparent and flexible FETs. Recently, ZnO films were formed at low temperature, without the need for vacuum environment, by spin coating or printing ZnO nanoparticles, which could lead to considerable reductions in fabrication costs.[5,6] However, the operating voltages of the ZnO TFTs fabricated at a reduced temperature were high.[4–7] High operating voltage results in high power consumption, a potential critical barrier for portable, battery-powered applications.

The operating voltage can be reduced by increasing the gate capacitance, which, in turn, increases the coupling between the gate electrode and channel layer. Some combination of higher gate dielectric constant and reduced film thickness leads to lower voltage operation. The use of a very thin gate insulator is not a suitable approach, given the relatively rough surfaces characteristic of polymer substrates. To ensure pinhole-free coverage, the film should be much thicker than the roughness of the substrate. The use of a high dielectric constant material is therefore the optimum approach for reducing the operating voltage on polymer substrates. The authors recently developed room-temperature deposited $Bi_{1.5}Zn_{1.0}Nb_{1.5}O_7$ (BZN) gate insulator with low leakage current and high dielectric constant and could thereby successfully fabricate low-voltage pentacene TFTs and inverters.[8] In this letter, the successful fabrication of low voltage (<4 V) ZnO TFTs with a BZN gate insulator is reported. This success could serve as a milestone in the realization of transparent and flexible electronics.

Disk-shaped BZN and ZnO targets were prepared by conventional mixed oxide methods. Pyrochlore structured BZN films were grown by pulsed laser deposition (PLD) onto Pt-coated Si wafers with a KrF excimer laser (248 nm) operating at a repetition rate of 25 Hz and a fluence of 2.0 J/cm^2. A 100-nm-thick Pt film (area$=3.5 \times 10^{-3}$ cm^2) was deposited as a top electrode through a shadow mask on the BZN film by dc magnetron sputtering. The dielectric constant was measured between 1 MHz and 1 kHz with an HP4192A impedance analyzer. The dielectric properties were examined with an applied electric field up to 100 kV/cm. *I–V* characteristics were measured with a HP4156A analyzer. During *I–V* measurements, the voltage step and delay time were set to 0.1 V and 0 s, respectively. The transmittance of the indium tin oxide (ITO)/ZnO/BZN structure was measured by an optical spectrometer (Perkin Elmer, UV/VIS/NIR Labmda 19) in the range from 200 to 900 nm. For the fabrication of flexible ZnO TFTs, polyimide was used as the substrate. A 50-nm-thick Cr gate electrode was deposited through a shadow mask by electron-beam evaporation. Then,

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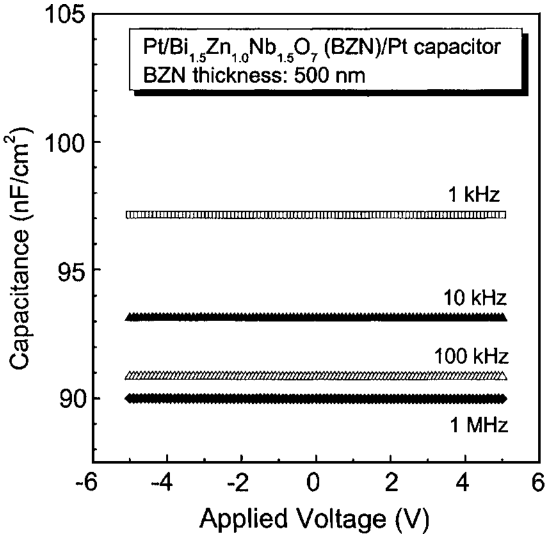

FIG. 1. Capacitance of a BZN film as a function of applied voltage at indicated frequencies.

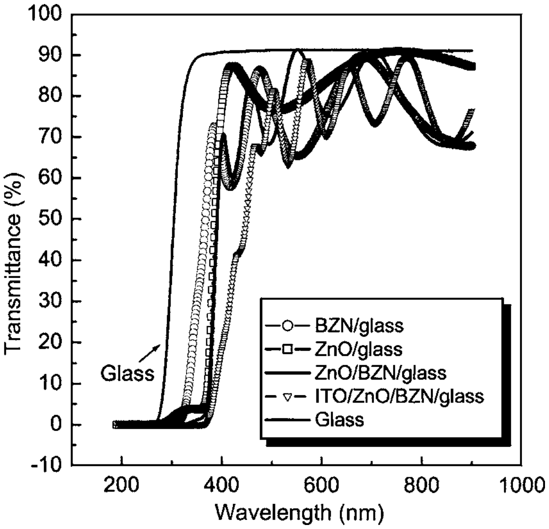

FIG. 2. Optical transmittance of the indicated structures as a function of wavelength in ITO (150 nm)/ZnO (100 nm)/BZN (200 nm)/glass structures.

a 200-nm-thick BZN gate dielectric film was deposited onto the Cr-coated polyimide substrate by PLD. A 100-nm-thick ZnO film was then deposited by PLD. The fabrication of the transistors was completed by the evaporation of gold top contacts through shadow masks to obtain channel lengths of 50–200 μm and widths of 2000 μm. This was followed by annealing at 200°C for 1 h in a forming gas ambient ($5\% H_2 + 95\% N_2$).

Figure 1 shows the capacitance of the BZN film as a function of applied voltage, measured at a series of frequencies ranging from 1 kHz to 1 MHz. The capacitance, for a BZN film of 500 nm thickness, ranged from 97.2 nF/cm^2 at 1 kHz to 90 nF/cm^2 at 1 MHz. The capacitance corresponds to a relative dielectric constant of 55 at 1 kHz and 51 at 1 MHz. These represent the highest values, so far obtained, for the dielectric constant of films prepared by room-temperature deposition. These values are approximately three times larger than the previously reported values for (Ba,Zr)TiO$_3$ and (Ba,Sr)TiO$_3$ films.[9] The high dielectric constant of these BZN films can be related to the ease with which the pyrochlore crystal structure forms at low temperature. This leads to a higher degree of short range order than that achieved by, e.g., perovskite-based oxides, and hence, higher dielectric constants.[8] No measurable tunability was observed up to voltages of 5 V insuring a voltage independent oxide capacitance. Interestingly, the low dielectric loss tangent ($\tan \delta$) of 5×10^{-4}, and the high resistivity ($\sim 3 \times 10^{13}\ \Omega$ cm) would normally make BZN films highly suitable for microwave regime applications, except for the fact that they exhibit low tunability.[10,11] However, for TFT operation, the low-voltage dependence of the dielectric constant ensures more predictable operation.

Figure 2 shows the optical transmission spectra of the indicated structures containing the BZN film in the wavelength range between 200 and 900 nm. To isolate the relative contributions to the optical loss of the overall structure, the transmittance of the glass substrate, BZN/glass, ZnO/glass, ZnO/BZN/glass, and ITO/ZnO/BZN/glass structures were investigated. The BZN/glass structure showed an absorption edge at 310 nm, which is shorter than that of the ZnO/glass structure (360 nm) but higher than that of the glass (280 nm). The transmittance at higher wavelengths than the absorption edge exhibited oscillations due to interference effects. The average transmittance value was on the order of 80% in all structures measured. Given that the transmittance of the BZN film was higher than or at least comparable to that of the ZnO and ITO films, one can conclude that transmission losses due to the BZN gate dielectric in TFTs can be largely ignored.

Figure 3(a) shows the drain-to-source current (I_{DS}) as a function of drain-to-source voltage (V_{DS}) of the ZnO TFT with BZN gate insulator. The TFTs, normally off, operate via the accumulation of carriers. Given that I_{DS} becomes nonzero for positive V_{GS}, one concludes that the carriers are electrons and the channel is n type. I_{DS} at a V_{GS} of 0 V is very small and the TFTs operates in the enhancement mode with excellent current saturation at higher V_{DS}. The large output impedance, achieved at saturation, is desirable in most electronics applications. Figure 3(b) shows the transfer curve of the ZnO TFT. The threshold voltage (V_{th}) was calculated from x-axis intercept of the square root of I_{DS} vs V_{GS} plot. Field effect mobility (μ_{FE}) modeled by the equation, $I_D = (WC_i/2L)\mu_{FE}(V_{GS} - V_{th})^2$, can be calculated from the slope of the plot of $|I_{DS}|^{1/2}$ vs V_{GS} in the saturation region ($V_{GS} = 4$ V), where L is the channel length, W is the channel width, Ci is the capacitance per unit area of the insulating layer, V_{th} is the threshold voltage, and μ_{FE} is the field effect mobility. The measured V_{th} and μ_{FE} were 2 V and 0.024 cm^2/V s, respectively. The subthreshold swing of the TFT was 0.25 V/dec. Importantly, the device was able to operate at a low voltage, below 4 V, due to the high gate capacitance. On-current and off-current at the operating voltage of 4 V were 0.3×10^{-6} A and 1.5×10^{-11} A, respectively, giving an on/off current ratio of 2×10^4. While the TFT operated at low voltage (<4 V), it had relatively low μ_{FE} and, hence, low on-current and on/off ratio. The field effect mobility of ZnO TFTs reported in the literature ranged from 0.01 to 3 cm^2/V s and the on/off current ratio in the range of 10^2–10^7.[7] The performance recorded here can be expected to improve by optimization of ZnO film growth.

The device performance, especially μ_{FE}, is mainly affected by the quality of the ZnO channel layer. The device

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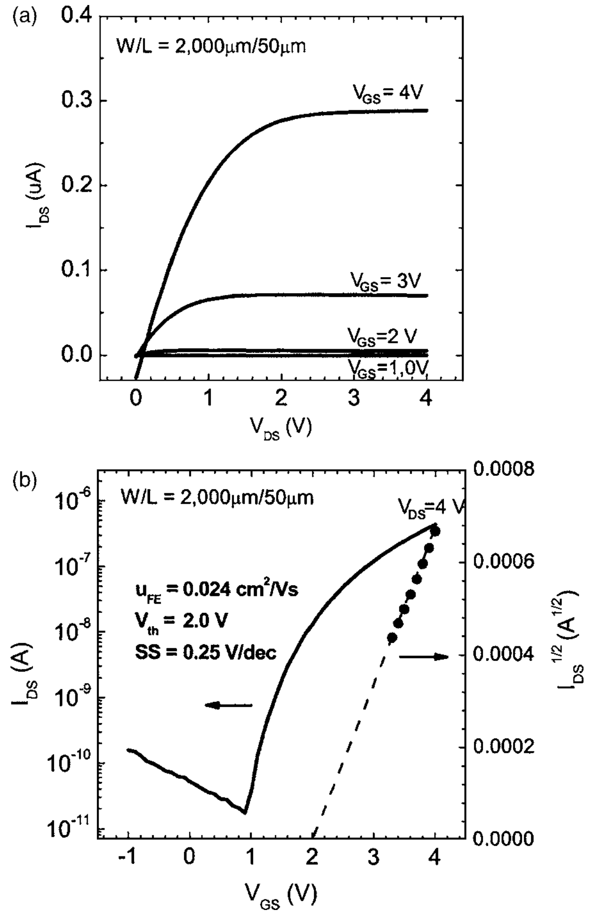

FIG. 3. Electrical characteristics of the ZnO TFT with a BZN gate insulator. (a) Drain-to-source current (I_{DS}) as a function of drain-to-source voltage (V_{DS}) at indicated gate-to-source voltages (V_{GS}). (b) Log drain-to-source current (I_{DS}) and square root of drain-to-source current (I_{DS}) as a function of gate-to-source voltage (V_{GS}) at drain-to-source voltage (V_{DS}) of 4 V. Width and length of the TFT were 2000 and 50 μm, respectively.

showed no field effect characteristics before being annealed in a forming gas ambient. Polycrystalline and amorphous films, in contrast to single crystals, tend to contain a higher density of defects. In particular, defects in the channel layer and/or at the channel/gate insulator interface act as trap sites and can significantly degrade device performance. It is well known that hydrogen treatments, such as a forming gas anneal or hydrogen plasma treatment, passivate defects, mainly dangling bonds, and improve device performance.[12] The finding that a forming gas anneal improves device performance in this work suggests that the ZnO films grown by PLD at room temperature exhibit an elevated defect density.

Usually, the mobility of devices with an active layer having many traps increases with increasing gate electric field. The μ_{FE} of pentacene TFTs with a BZN gate insulator, for example, increased from 0.05 cm^2/V s at 0.1 MV/cm to 0.5 cm^2/V s at 0.37 MV/cm. One would expect, likewise, that the μ_{FE} of the ZnO TFT could be increased by increasing the gate electric field. Unfortunately, the gate leakage current began to significantly increase above V_{GS}=4 V, leading to device degradation. The higher leakage current is likely related to the ZnO film, given that the leakage current of pentacene TFTs with a BZN gate insulator remained low even above an electric field of 0.5 MV/cm. Further study is needed to confirm the source of the leakage current in this system.

In conclusion, ZnO TFTs operating at low voltage (<4 V) were successfully fabricated using the high-*K* BZN gate insulator. Threshold voltage and subthreshold swing were 2 V and 0.25 V/dec, respectively. The field effect mobility and on/off current ratio, at operating voltage of 4 V, were 0.024 cm^2/V s and 2×10^4, respectively. The optical transmittance of the BZN film was better than the other films used in transparent TFTs such as ZnO and ITO. The success of low-voltage operation of ZnO TFTs may serve as a milestone for realizing transparent and flexible electronics and their use in portable and battery-powered applications.

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