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Effect of electrode material on resistive switching memory behavior of solution-processed resistive switches: Realization of robust multi-level cells

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ABSTRACT

Sol-gel processed ZrO_2 was used as the main active channel material for a resistive switching memory device implemented on Indium Tin oxide coated glass substrates. The memory properties of the deposited ZrO_2 layers depended on the top electrode material. Inert Au top electrodes did not yield resistive switching memory properties, while Ag top electrodes provided conventional resistive switching memory properties, with sharp on and off switching operation. In contrast, Ti top electrodes provided smooth on and off switching operation, and modifying the pulse widths and voltages allowed good control over the resistivity. The fabricated Au/Ti/ZrO_2/ITO systems exhibited four different resistance levels, and good multi level storage characteristics were observed for at least 10^4 cycles without degradation.

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1. Introduction

Significant progress in solution processed electronics has occurred in recent years, with advances in transistors, mechanical switches, and sensors having taken place [1–4]. Memory is one of the most critical components in modern electronic devices, such as RFID tags, sensors, and smart packages since memory is required for most communication and computation functions. There have been some demonstrations of printed memories over the years; however, low-power performance, reliability and bit density must be increased to match application requirements. Two-terminal resistive random access memories (RRAM) are considered promising means of achieving these goals. These types of memory have many attractive properties, such as low power consumption, fast switching time, high endurance, good retention, and simple structure [5–10]. For this application, several different types of inorganic switching layers have been introduced [11–15]. In particular, several metal oxides have been investigated. Among these metal oxides, ZrO_2 insulator is an important material for use as a gate dielectric in high-performance transparent thin-film transistors (TFTs) [1,16] and has been shown to exhibit resistive switching behavior as well [17,18]. Using ZrO_2 as both a TFT insulator and the primary material for resistive switching memory will decrease the number of process steps and thereby manufacturing cost, especially for the 1-transistor - 1-RRAM memory cell structure [19,20]. In the conventional flash memory field, multi-level cell technology has been introduced to increase bit density and decrease manufacturing cost per bit. Some multi-level RRAM cell demonstrations have been made using current-limited programming, which is difficult to implement in realistic large-area circuit applications [21]. Only a few studies have presented multi-level memory characteristics based on the active material properties alone [22,23].

In this work, we introduce another new memory structure, by using intermediate Ti electrodes, for multi level resistance memory unit cells. We use sol-gel processed ZrO_2 as the main active material in a resistive switching memory device and demonstrated that Ag and Ti top electrical contacts on ZrO_2 layers can be engineered to realize either single-level cell functionality using a top Ag electrodes or unique multi-level memory functionality using top Ti electrodes. We also briefly explain the physical processes involved. Robust resistive switching functionality was achieved using simple pulse-based programming schemes, as would be required for use in realistic circuit applications. The resulting devices exhibit no significant retention loss after 10^4 s and no endurance degradation after 10^2 switching cycles.

2. Experimental procedures

All regents were purchased from Sigma-Aldrich and used as received. ZrO_2 liquid precursor was prepared using a sol-gel process. First, 0.487 g of zirconium (IV) acetylacetone was dissolved in 10 ml ethanol with 0.1 mol mono-ethanolamine as the stabilizer. The ZrO_2

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liquid precursor was ultra-sonicated for 1 h and then deposited by spin-coating on UV/ozone-treated Indium Tin oxide (ITO)-coated glass. Multiple spin coating steps were performed to produce 50-nm thick ZrO_2 films. Between each spin-coating process, the samples were dried at 300 °C for 1 min in air. A sintering process was then conducted at 500 °C for 1 h in O_2. Next, 50-nm thick Au or Ag electrodes or 100-nm thick Ti (50 nm)/Au (50 nm) electrodes were deposited onto the sintered ZrO_2 layer by thermal evaporation through 100 μm × 200 μm metal shadow masks. The structural properties and phase of the sol-gel processed ZrO_2 films were confirmed by X-ray diffraction (XRD:SIEMENS D5000 X-ray diffractometer). The surface morphology of sol-gel processed ZrO_2 was examined by atomic force microscopy (AFM:DI AFM Nanoscope Dimension 3100). The dielectric constant dispersion was characterized using a capacitor, consisting of ITO, sol-gel processed ZrO_2 layers and thermally evaporated Au electrodes and directly measured with 4285A LCR meter. The resistive switching memory characteristics of the fabricated devices were measured using an Agilent 4155 semiconductor parameter analyzer at room temperature in air.

3. Results and discussion

Fig. 1(a) shows the XRD spectra of the sol-gel processed films. The formed film was a ZrO_2, well matching with the JCDPS (811554) of the tetragonal ZrO_2 phase. The crystallite size of sol-gel processed ZrO_2 was calculated from the Scherrer equation, $D = K\lambda/(\beta\cos\theta)$, where K is the shape factor, λ is the wavelength of X-ray, β is the line broadening at half the maximum intensity (FWHM), and θ is the Bragg angle. The calculated crystallite size is 5 nm. Fig. 1(b) illustrated the variation of the dielectric constant as a function of frequency. The dielectric constant remains relatively constant at low frequencies (up to 10 kHz) and over 10 kHz, the dielectric constant show decreasing trend behavior with increasing frequency. The obtained dielectric constant at low frequency is around 22. The inset shows the typical leakage current and the break down voltage. A sudden increase in current at approximately + 9.0 V was observed during the positive sweep, and the device did not return to the HRS in the subsequent negative and positive sweeps. The ITO/ZrO_2/Au structure devices thus did not exhibit resistive

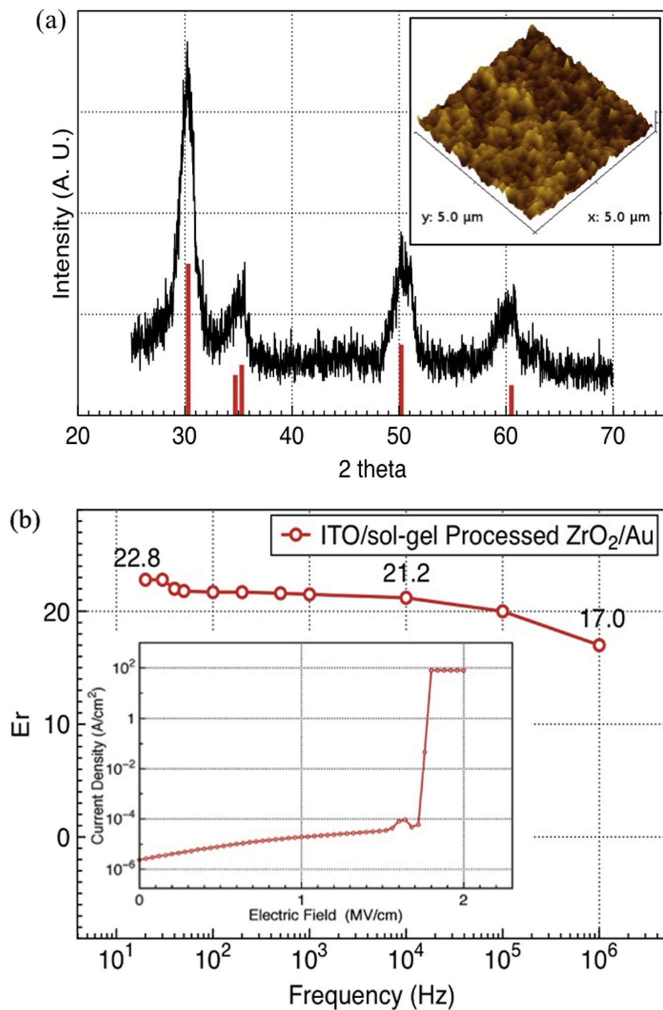

Fig. 1. (a) XRD data of the sol-gel processed ZrO_2 layers. The inset shows the AFM image of sol-gel processed ZrO_2 layers. (b) The dielectric constant variance of sol-gel processed ZrO_2 layers, as a function of frequency. The inset shows the typical leakage current and the break down voltage.

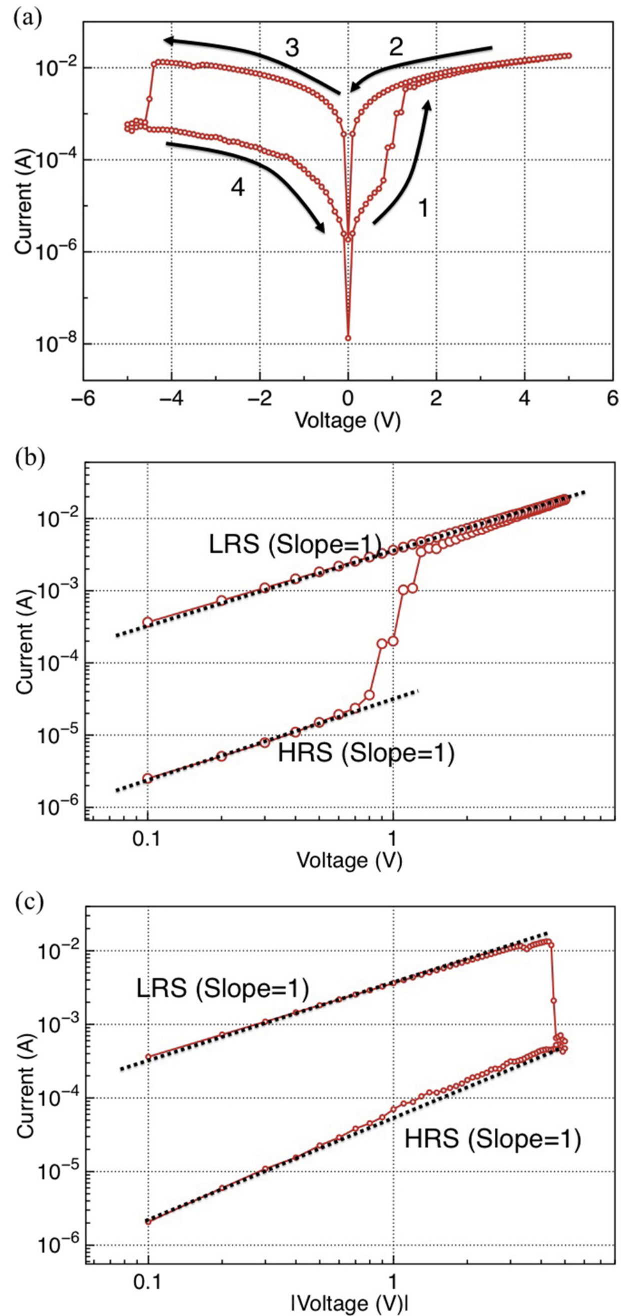

Fig. 2. (a) Representative IV curves obtained from the ITO/ZrO_2/Ag RRAM devices. The arrows and numbers indicate the voltage sweep directions. Double logarithmic plots of the Ohmic transport mechanisms of the high- and low-resistance states of the ITO/ZrO_2/Ag devices: (b) positive and (c) negative voltage regions.

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