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Trap characterization and conductance quantization in phosphorus-doped ZnO memory devices

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ABSTRACT

This work addresses the trap characterization and conductance quantization of bipolar resistive switching devices using transparent phosphorus-doped ZnO (ZnO:P) films. The average transmittance in the visible light region and optical band-gap of the ZnO:P films are around 95.6% and 3.4 eV, respectively. Based on the analysis of current-voltage characteristics at high resistance state (HRS) in the structure of Ni/ZnO:P/Ni, the current conduction mechanism is dominated by hopping conduction. Accordingly, the trap spacing and trap energy level in ZnO:P films are extracted to be around 1.5 nm and 280–370 meV, respectively. The trap spacing remains about constant during set/reset cyclic switchings. Meanwhile, the trap energy level is decreased as the set/reset switching increases, which leads to the raised leakage current at HRS. In addition, the conductance values read at small voltages exhibit quantized effect during the set processes. The histogram of normalized conductance reveals that the conductance levels and peaks initially appear at $1 G_0$ and $2 G_0$, and shift to $5.5 G_0$, $7 G_0$, and $8 G_0$ after the set/reset switching cycle is more than about 100 times, where G_0 ($=77.5 \mu S$) is the quantum of conductance.

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1. Introduction

ZnO is a direct and wide band-gap (E_g) semiconductor that typically crystalizes in the hexagonal wurtzite structure. The E_g value is around 3.37 eV at 300 K and increases to be around 3.44 eV at 4.2 K. Thus ZnO is transparent in visible light and is suitable for blue and ultraviolet photonic devices [1,2]. It would be very attractive to the transparent electronics integrated with a resistive random access memory (RRAM) structure in which the switching layer possesses high transmittance in visible light. In addition, ZnO is able to be adopted to fabricate flexible RRAM devices because of its low synthetic temperature [3]. There are several attempts reported to improve ZnO-based material as a switching layer in RRAM devices, such as stacked with various metal electrodes, controlled its deposition process, doped with various elements, and embedded/multilayered with various metal oxides [4]. For the effect of electrodes, high work function may create high interfacial barrier to induce resistive switching (RS) properties with high ON/OFF ratio [5]. Oxidizable metal electrodes may have the advantage of having oxygen reservoir behavior leading to long

endurance in ZnO-based memory devices. Furthermore, the use of low electronegativity and small ionic size of the active electrodes may result in easier formation and rupture of conductive filaments (CFs) leading to lower set and reset voltages. The major parameters of ZnO-based RRAM devices as a function of different metal electrodes are tabulated in Ref. [4]. Because microstructural properties and defects in ZnO film will strongly affect the switching characteristics, controlled ZnO film growth is required to fabricate good quality film having highly oriented growth and less native defects. For example, ZnO film properties can simply controlled by modulating Ar/O₂ flow ratio during sputtering. In some ZnO-based devices, the ON/OFF ratio tends to increase as oxygen flow ratio increase [4]. Meanwhile, the increase of oxygen flow may result in device instability because of smaller grains grown in the ZnO film that leads to higher number of grain boundaries and multiple conducting paths [6]. Besides, post-thermal treatment after deposition can be employed to modulate the crystallinity and defect concentration in ZnO film, which alters the switching properties in ZnO-based RRAM devices [4]. Multilayered and embedded structure can also be used to improve the switching characteristics in ZnO-based RRAM devices [4]. By employing proper electrical programming, the formation and rupture of CFs can be well controlled at the particular switching layer and then the complementary switching characteristics can be achieved. The complementary

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switching characteristics can be used to solve the problem of read operation error arising from the sneak current issue in the three-dimensional crossbar array architecture [7–9]. Compare to deposition process and thermal treatment on the RS layer, the doping technique may be more effective to control defect concentration. Various dopant elements have been employed to achieve proper switching characteristics in ZnO-based RRAM devices [4]. However, excessive dopant may degrade the cycling endurance and stability performance because of the weakening of *c*-axis-textured structure [10]. ZnO film with *c*-axis texture can enhance the confinement of conductive filaments. However, there is a trade-off between reducing native defects and maintaining *c*-axis-textured structure in ZnO-based RRAM devices according to the study of relationship between CF development and dopant concentration in RS layer [10]. In general, CFs are known to be easily formed and branched along the grain boundaries. Hence, employing an amorphous switching layer by doping technique can achieve highly uniform RS properties in ZnO-based RRAM devices because of its lack of grain boundary structure [11,12].

Datta pointed out that the conductor will show conductance quantization behavior if any of the three dimensions of a conductor is smaller than one of the following three characteristic length scales [13]: (1) the de Broglie wavelength which is associated with the kinetic energy of the electrons; (2) the mean free path which is associated with the distance that an electron travels before its initial momentum is destroyed; and (3) the phase-relaxation length which is associated with the distance that an electron travels widely from one material to another. In the RRAM devices with atomic-scale CF, the current transport through the CF can be carried along the discrete conductance channels. In this case, the electrons are not scattered and the atomic-scale CF behaves like a waveguide for electrons. The “electron-waveguide” could be considered as a ballistic transportation path made up a bundle of discrete conductance channels. Each conductance channel contributes a maximum amount of one G_0 to the total conductance, namely, one G_0 could be simply considered as a single atomic contact or a nanowire in a CF. Landauer theory for ballistic electron transport predicts $G_0 = 2e^2/h$ as the quantum of conductance, with the value of $77.5 \mu\text{S}$ or $12.9 \text{ k}\Omega^{-1}$, where e is the elementary charge, h is the Planck's constant and the factor 2 accounts for spin degeneracy [14–16]. Recently, the quantized conductance (QC) effect in RRAM has attracted a lot of attention, because it can be applied to realize the multi-level storage for ultra-high-density memory applications as well as the interesting physics behind [17,18]. The QC effect has been observed in various RRAM devices with the structures of sandwich and crossbar. The sandwich structure can be a stack of bottom electrode and RS layer using a patterned top electrode or a conductive atomic force microscope (CAFM) tip as top electrode. The RS materials involving QC effect include amorphous silicon, silicon oxide, polymer, solid-state electrolytes, transition metal oxides, etc. There are various material systems with the QC effect are tabulated in Ref. [17], involving different RS mechanisms, RS polarities (unipolar or bipolar), conductive filament types, set/reset processes, and conductance quantization levels. In addition, using an appropriate operating method is the key to precisely control the size of CFs for successful observation of the QC effect in RRAM devices. The reported operating methods include fresh device operation, voltage sweeping, current sweeping, voltage pulse operation, and constant voltage bias operation [17]. Recently, first principle calculations based on the density-functional theory were performed to obtain quantized conductance of conductive path in a crystalline oxide matrix [19]. Furthermore, an equivalent circuit model has been built to account for the QC effect in ECM devices [20].

In this work, transparent RS layer of phosphorus-doped ZnO

(ZnO:P) film and high work-function metal electrode Ni are adopted to study the resistive switching mechanism and quantized conductance effect in the RRAM structure of Ni/ZnO:P/Ni. Using the operating method of voltage sweeping, the conduction mechanism and the conductance quantization in the RS layer are identified. Based on the analyses of the conduction mechanism, the defect trap properties in ZnO:P film are explored, including trap spacing and trap energy levels. Furthermore, the discrete conductance phenomena are found in the phosphorus-doped RRAM devices in which the quantized conductance steps with multiples of G_0 in the set processes.

2. Experiment

In this work, metal-oxide-metal capacitors were fabricated and investigated using the resistive switching film of ZnO:P. The ZnO:P films with 25 nm-thick were deposited by *rf* magnetron sputtering at room temperature using a ceramic ZnO:B target with P_2O_5 serving as the doping agent. The phosphorus doping concentration of ZnO:P films was about 4 wt%. The sputtering power is 60 W. The working pressure is 0.533 Pa in Ar ambient with a flow rate of 20 sccm. Nickel (Ni) works as top and bottom electrodes, which were deposited by thermal evaporation. The top Ni electrode with a round area of $3.14 \times 10^{-4} \text{ cm}^2$ was patterned by the metal mask process. The electrical properties of the Ni/ZnO:P/Ni memory devices were characterized under dark condition using Agilent 4156C semiconductor parameter analyzer.

3. Results and discussion

The inset of Fig. 1 shows the transmittance spectra of phosphorus-doped ZnO (ZnO:P) films in the wavelength ranging from 340 to 700 nm. The average transmittance of ZnO:P film is about 95.6% in the visible region (400–700 nm). In this work, Lambert's law was used to identify the optical absorption coefficient in ZnO:P films [21].

$$\alpha = (1/d)\ln(1/T_{TM}) \quad (1)$$

where α is the optical absorption coefficient, d is the film thickness and T_{TM} is the transmittance. For a direct band-gap semiconductor, the optical absorption coefficient α is associated with light frequency by the following equation [21].

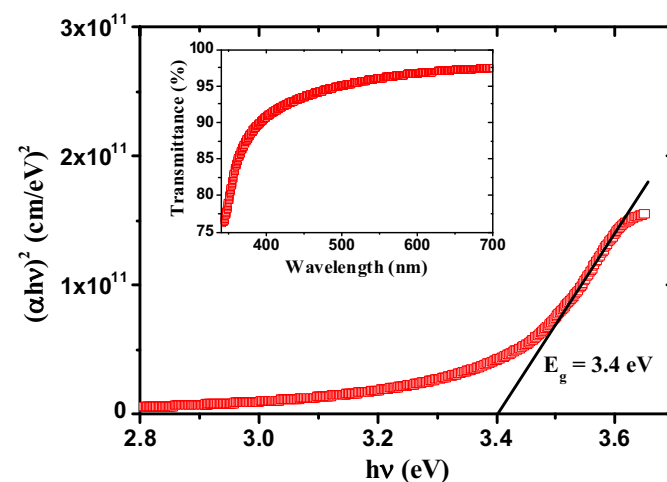


Fig. 1. Optical band-gap calculated by the $(\alpha hv)^2$ -energy of the transmittance curve. Inset shows the optical transmittance of phosphorus-doped ZnO films.

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