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# Parameters controlling microstructures and resistance switching of electrodeposited cuprous oxide thin films

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ABSTRACT

Cuprous oxide ($Cu_2O$) thin films were electrodeposited cathodically from a highly alkaline bath using tartrate as complexing agent. Different microstructures for $Cu_2O$ thin films were achieved by varying the applied potential from −0.285 to −0.395 V versus a reference electrode of Ag/AgCl at 50 °C in potentiostatic mode, and separately by changing the bath temperature from 25 to 50 °C in galvanostatic mode. Characterization experiments showed that both grain size and orientation of $Cu_2O$ can be controlled by changing the applied potential. Applying a high negative potential of −0.395 V resulted in smaller grain size of $Cu_2O$ thin films with a preferred orientation in [111] direction. An increase in the bath temperature in galvanostatic electrodeposition increased the grain size of $Cu_2O$ thin films. All the films in Au/$Cu_2O$/Au-Pd cell showed unipolar resistance switching behavior after an initial FORMING process. Increasing the grain size of $Cu_2O$ thin films and decreasing the top electrode area increased the FORMING voltage and decreased the current level of high resistance state (HRS). The current in low resistance state (LRS) was independent of the top electrode area and the grain size of deposited films, suggesting a filamentary conduction mechanism in unipolar resistance switching of $Cu_2O$.

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## 1. Introduction

Cuprous oxide ($Cu_2O$) is a p-type semiconductor with a direct band gap of 2.0 eV [1–3], which has recently received much attention due to its potential application in resistance switching random access memory (RRAM) [4–8]. A RRAM is a nonvolatile memory with a capacitor-like structure composed of an insulating or oxide material between top and bottom metal electrodes [9–12]. Reversible switching of the oxide material between high and low resistance states is the principle characteristic of a RRAM device [13,14].

Typically a high electric voltage with compliance current is necessary for most of metal oxides, such as $Cu_2O$, to initiate the reversible resistance switching [15–18]. This initial process in resistance switching is called FORMING [5,10,19]. After FORMING, the material can be switched from a low resistance state (LRS) to a high resistance state (HRS) by the RESET step, and from the HRS to LRS by the SET step [20,21]. In spite of the importance of the FORMING step in resistance switching, there is a lack of information about the parameters that can control it. Generally, a lower FORMING voltage is favorable to have less power consumption and to avoid fatal damage during practical application [22,23].

Based on the current-voltage (I–V) characteristics, resistance switching can be classified into two groups, unipolar and bipolar switching [24–26]. In unipolar switching, the required voltages in SET and RESET steps are in the same polarity with different magnitudes, while in bipolar switching they are in opposite polarity [5,18,27]. These different switching behaviors are dependent on the involved materials in RRAM, including the intermediate oxide layer and the top and bottom electrodes [26,28].

Although different models have been proposed to clarify the resistance switching mechanism [9,13,19,29], the origin of resistance switching is still in controversy. Generally, resistance switching mechanism can be categorized according to the conducting path in the LRS, and two different mechanisms are suggested in the literature, a filamentary conduction path and an interface-type conduction path [5,9,30]. In the filamentary conduction path, the formation and rupture of conductive filaments result in resistance switching of oxide materials [8,31–33]. In the interface-type conducting path, the resistance switching takes place over the entire area of the interface between the metal electrodes and the oxide material [34–36].

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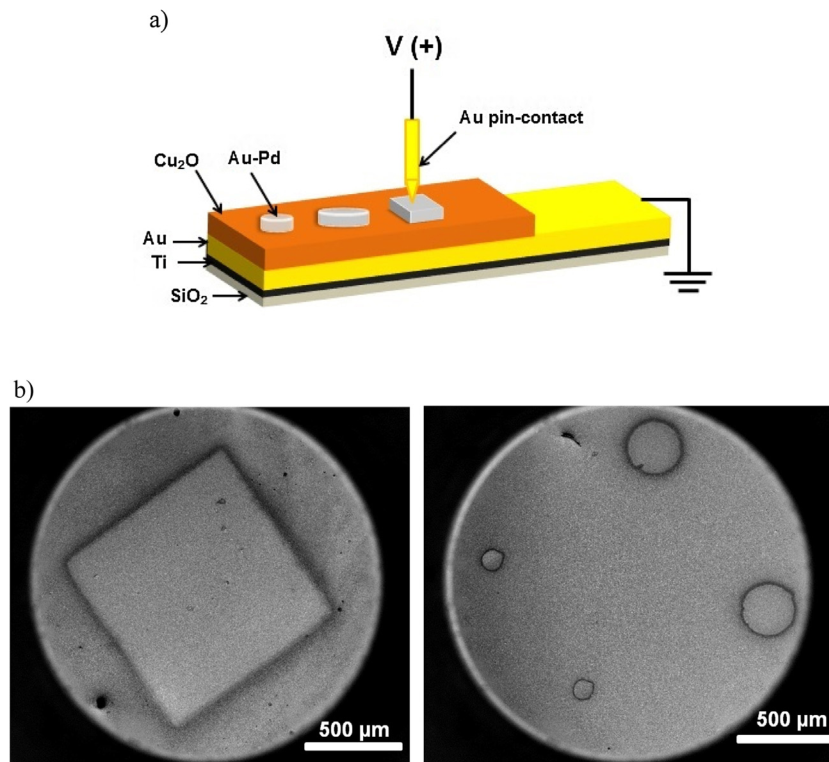


**Fig. 1.** a) Schematic of resistance switching cell with different top electrode area. b) SEM images of different top electrode area.

Microstructures of the oxide materials are believed to affect the resistance switching of RRAMs, and there are only few works in the literature those studied the effects of microstructures on resistance switching of oxide metals [24,37–39]. For example, Park et al. [24] compared the resistance switching behavior of polycrystalline and epitaxial NiO layers in Pt/NiO/Pt cell structure, and they reported a poor reproducible resistance switching in epitaxial NiO layer because of the crucial effect of microstructural defects, such as grain boundaries, on switching properties.

Since one of the most efficient methods to produce thin films of oxide materials for RRAM applications is the electrodeposition process, in this work, we attempt to find the relationships between the electrodeposition parameters, developed microstructures, and resistance switching properties of $Cu_2O$. First we studied the parameters controlling the microstructures of electrodeposited $Cu_2O$ thin films, and then we investigated the effects of $Cu_2O$ microstructures and the top electrode area on the resistance switching characteristics of $Cu_2O$ thin films. To understand the resistance switching mechanism, we studied the role of top electrode area on the current levels in both LRS and HRS.

## 2. Experimental procedure

$Cu_2O$ thin films with a thickness of about 3.8 $\mu$m were electrodeposited on gold coated glass substrates by cathodic reduction of Cu(II) tartrate solution. The solution contained 0.2 M L(+)-tartaric acid, 0.2 M copper (II) sulfate pentahydrate, and 3 M sodium hydroxide [40]. To obtain different microstructures of $Cu_2O$ thin films, both potentiostatic and galvonstatic electrodeposition were performed. Ag/AgCl/KCl(sat.) was used as the reference electrode for electrodeposition of $Cu_2O$ thin films. In potentiostatic mode, the electrodeposition was performed at different potentials between −0.285 and −0.395 V at 50 °C. In galvanostatic mode, different microstructures were achieved at a cathodic current density of 1 mA/cm$^2$ and different temperatures from 25 to 50 °C. The structure and phase composition of electrodeposited films were studied by a high resolution Philips X-Pert MRD X-ray diffractometer (XRD) using Cu K$\alpha$ radiation. The surface and cross section morphologies were observed using scanning electron microscope (SEM). To measure the resistance switching properties of $Cu_2O$ thin films, Au-Pd top electrodes with same thickness of 100 nm and different areas of $7.8 \times 10^3$ $\mu$m$^2$, $5.4 \times 10^4$ $\mu$m$^2$, and $11 \times 10^5$ $\mu$m$^2$ were sputtered on the films using a shadow mask. Fig. 1a and b show the schematic of resistance switching cell and SEM images of three different areas of top electrodes, respectively. Sharp Au spring pin contact was used for electrical connection to the top electrodes. During resistance switching measurements, a positive bias voltage was applied to the top electrode while the bottom electrode was grounded. Resistance switching measurements were performed using Keithley 2400 current source at room condition. To study the effect of $Cu_2O$ microstructure on FORMING voltage, a compliance current of 5 mA was applied on contact area of $7.8 \times 10^3$ $\mu$m$^2$ for all of the samples. To elucidate the resistance switching mechanism, the effect of top electrode area was studied on current levels of LRS and HRS for the sample electrodeposited at cathodic current density of 1 mA/cm$^2$ and 40 °C.

## 3. Results and discussion

Fig. 2a shows X-ray diffraction (XRD) patterns of electrodeposited thin films at different potentials. Pure $Cu_2O$ thin films are obtained at all the applied potentials. At low negative potentials of −0.285 and −0.295 V, $Cu_2O$ is not oriented, while a highly [111]-oriented $Cu_2O$ thin films are obtained at high negative potential of −0.395 V. Fig. 2b shows the cross section images of $Cu_2O$ thin films electrodeposited at −0.395 and −0.285 V. Electrodeposited films at negative potential of −0.395 V show columnar microstructure with oriented grains along [111] direction while non-oriented grains are

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