



Research articles

Non-equilibrium character of resistive switching and negative differential resistance in Ga-doped Cr_2O_3 system

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ARTICLE INFO

Article history:

Received 9 November 2017

Received in revised form 4 February 2018

Accepted 21 February 2018

Available online 21 February 2018

Keywords:

Resistive switching

Negative differential resistance

Charge relaxation

Bi-stable electronic states

ABSTRACT

The samples of Ga-doped Cr_2O_3 system in rhombohedral crystal structure with space group $R\bar{3}C$ were prepared by chemical co-precipitation route and annealing at 800 °C. The current-voltage (I–V) curves exhibited many unique non-linear properties, e.g., hysteresis loop, resistive switching, and negative differential resistance (NDR). In this work, we report non-equilibrium properties of resistive switching and NDR phenomena. The non-equilibrium I–V characteristics were confirmed by repetitively measuring and time relaxation of current. The charge conduction process was understood by analysing the I–V curves using electrode-limited and bulk-limited charge conduction mechanisms, which were proposed for metal electrode/metal oxide/metal electrode structure. The I–V curves in the NDR regime and at higher bias voltage regime in our samples did not obey Fowler-Nordheim equation, which was proposed for charge tunneling mechanism in many thin film junctions. The non-equilibrium I–V phenomena were explained by considering the competitions between the injection of charge carriers from metal electrode to metal oxide, the charge flow through bulk material mediated by trapping/de-trapping and recombination of charge carriers at the defect sites of ions, the space charge effects at the junctions of electrodes and metal oxides, and finally, the out flow of electrons from metal oxide to metal electrode.

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1. Introduction

Recently, different types of random access memory (RAM) devices, e.g., ferroelectric random access memory (FeRAM), magnetoresistive random access memory (MRAM), and phase change random access memory (PRAM), have been introduced in spintronics [1]. There is an attempt to develop non-volatile memory (NVM) devices where data can be stored/written by magnetic process and stored information can be retrieved by electrical process. MRAM is one of the novel approaches that can store data in magnetic storage element, called magnetic tunnel junction (MTJ). MRAM has almost all the criteria of becoming the next generation memory devices (high-speed reading and writing capacity with additional non-volatility). These NVM devices utilize charge-spin coupling in the material and such materials are not easily available in nature. In an attempt to search alternative materials for non-volatile memory device applications, insulator-magnetic metal oxides with high resistive switching have drawn a world-wide attention [1,2]. This is due to the fact that resistive Random Access Memory (ReRAM) devices are based on resistive switching (RS) process; resistive

state is controlled by sweeping electric voltage or current pulses and retains data even at power shut down condition [3–5]. ReRAM is believed to be a potential candidate for the applications in next-generation NVM devices due to its low power consumption, high switching speed and high density [6,7]. The thin films of transition metal oxides, e.g., Cr_2O_3 [7], Ga_2O_3 [8,9], Al_2O_3 [10], Fe_2O_3 [11], Ga-doped Fe_2O_3 [12], $\text{Ba}_{0.7}\text{Sr}_{0.3}\text{TiO}_3$ [13], NiO [14], TiO_2 [15,16], and SiO_2 [17] have shown different types of resistive switching (RS); unipolar, bipolar or abnormal nature. The unipolar switching is polarity-independent (symmetric) and resistance state of the device is switched between low resistance state (LRS) and high resistance state (HRS) ($\text{LRS} \leftrightarrow \text{HRS}$) by consecutive application of electric field either in same or opposite directions. In bipolar switching, resistance state of the device is polarity-dependent (antisymmetric). It switches $\text{LRS} \rightarrow \text{HRS} \rightarrow \text{HRS} \rightarrow \text{LRS}$ by successive electric field application with alternate polarity, i.e., LRS at one voltage polarity and HRS on reversing the voltage polarity. In abnormal bipolar RS, resistance state is switched from LRS to HRS during positive polarity of electric field, and LRS to HRS during negative polarity of electric field. The abnormal RS phenomenon has been observed in devices like $\text{Pt/GaO}_3/\text{Pt}$ [8] and $\text{Pt/TiO}_2/\text{Pt}$ [18]. Recently, thin films of BaTiO_3 [19] and ZnO [20] have shown the coexistence of resistive switching and negative differential

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resistance (NDR) ($\frac{dV}{dI} < 0$). The materials with NDR and resistive switching at room temperature are of increasing interest for applications in analog and digital circuits, including logic gates, voltage-controlled oscillator, and flip-flop circuit [21]. The additional feature of I–V loop is also important for applications of the materials in switching, low power memory, dynamic random access memory, static random access memory, and high storage density with long retention [22].

The magnetic study [23] showed Ga doped α -Cr₂O₃ system as a diluted antiferromagnet with ordering temperature at about 50 K and a typical paramagnet at room temperature. The incorporation of non-magnetic Ga atoms in antiferromagnetic α -Cr₂O₃ has shown resistance switching and NDR phenomena. The present Ga doped samples are electrically high resistive semiconductor; where electric field induced migration and concentration gradient induced diffusion of oxygen ions/vacancies can play a crucial role on electronic properties. Hence, the I–V study for such material under high electric voltage will be useful to get insight of the role of microscopic level lattice defects on charge conduction mechanism. This work will study the non-equilibrium I–V properties in Ga doped α -Cr₂O₃ system using Pt/CrGaO/Pt structure. The non-equilibrium properties of I–V characteristics will be understood using experimental results of bias voltage cycling and time relaxation effects on current measurements.

2. Experimental

2.1. Material preparation and characterization

Details of the preparation of Ga doped Cr₂O₃ samples by chemical co-precipitation and structural phase stabilization have been described in earlier work [23]. In the present work, we used the samples with chemical compositions Cr_{1.45}Ga_{0.55}O₃ and Cr_{1.17}Ga_{0.83}O₃. The X-ray diffraction pattern confirmed single phased rhombohedral structure with profile fitted cell parameters ($a = 4.955$ Å, $c = 13.572$ Å, $V = 288.5$ Å³ for Cr_{1.45}Ga_{0.55}O₃, $a = 4.955$ Å, $c = 13.573$ Å, $V = 288.7$ Å³ for Cr_{1.17}Ga_{0.83}O₃). The grain size of the samples (~ 50 nm and 44 nm for Cr_{1.45}Ga_{0.55}O₃ and Cr_{1.17}Ga_{0.83}O₃, respectively) was found in same range. Raman spectra of the Ga doped samples were matched to the structure of α -Cr₂O₃. This provided microscopic evidence of the occupancy of Ga atoms into the sites of Cr atoms in α -Cr₂O₃.

2.2. Measurement of I–V characteristics

Current-Voltage (I–V) characteristics of the samples were measured using Keithley dual Source Meter (2410-C, USA) and high resistance meter (6517B, Keithley, USA). The pellet shaped samples ($\phi \sim 13$ mm, $t \sim 0.3$ – 0.5 mm) were sandwiched between two Pt electrodes using pressure in home-made sample holder to make a vertical Pt/CrGaO/Pt device structure. Two modes (P and N) with 4 segments were adopted to measure the I–V curves by sweeping bias voltage from 0 to suitable voltage limit across the sample. In P mode, voltage was swept in positive side with step sequences $0 \rightarrow +V_{\max}(S1) \rightarrow +V_{\max} \rightarrow 0(S2) \rightarrow 0 \rightarrow -V_{\max}(S3) \rightarrow -V_{\max} \rightarrow 0(S4)$. In N mode, bias voltage was swept in negative side with sequences $0 \rightarrow -V_{\max}(S1) \rightarrow -V_{\max} \rightarrow 0(S2) \rightarrow 0 \rightarrow +V_{\max}(S3) \rightarrow +V_{\max} \rightarrow 0(S4)$. Reproducibility of the I–V features and charge relaxation effect was tested by additional protocols, (1) 20 times repetition (20P or 20N) of loop measurement; (2) sweeping the voltage with sequences $+V_{\max} \rightarrow 0 \rightarrow -V_{\max}$ and $-V_{\max} \rightarrow 0 \rightarrow +V_{\max}$; (3) varying the delay time between two set voltage points; and (4) time dependent current (I) measurement at constant voltage.

2.3. Method of I–V curve analysis

The I–V characteristics have been studied by placing the metal oxide (MO) in between two metal electrodes (M) to form the M/MO/M structure and the characteristics depend on two mechanisms [24,25]. First one is the electrode-limited conduction (ELC) mechanism, which includes Schottky emission and Fowler-Nordheim tunnelling. Second one is the bulk-limited conduction (BLC) mechanism, which includes space charge limited conduction (SCLC) and Pool-Frenkel (P-F) emission. The ELC mechanism depends on electrical nature at electrode-material interfaces, whereas BLC mechanism relies upon electrical properties of the material itself. The Schottky emission mechanism is described by following equation.

$$J = A * T^2 \exp \left[\frac{-q \left(\phi_B - \sqrt{\frac{qE}{4\pi\epsilon}} \right)}{k_B T} \right] \quad (1)$$

J is the current density, $E (= V/t; t$ is the sample thickness and V is applied voltage) is the electric field, T is measurement temperature (300 K), A is the Richardson constant (typical value $\sim 8 \times 10^4$ A/m²/K² for Metal Oxide structure [24]), ϕ_B is barrier height, $\epsilon (= \epsilon_0 \epsilon_r)$ is the dynamic dielectric constant, and k_B is Boltzmann constant. The validity of Schottky emission can be tested from a linear fit in the $\ln J$ vs. $\sqrt{E}$ (or $\ln I$ vs. $\sqrt{V}$) plot. If ϕ_B at the M/MO junction is much higher than kinetic energy of incoming electrons, tunnelling of electrons through barrier is controlled by Fowler-Nordheim equation [26].

$$J = AE^2 e^{\left(-\frac{B}{E}\right)} \quad \text{where } A = \frac{q^3}{8\pi h \phi_B}, \quad B = \frac{8\pi \sqrt{2m^*} q (\phi_B)^{\frac{3}{2}}}{3h}; \quad E = V/t \quad (2)$$

In case of SCLC mechanism, the Power law: $I \sim V^m$ with exponent $m = 2$ is applicable for the material without any charge trapping [27]. The trapping and de-trapping of the electrons at defect sites, while transporting through metal oxide, are controlled by P-F emission that satisfies the following equation.

$$J = qn_c \mu \left(\frac{V}{t} \right) \exp \left[\frac{-q\phi_B}{k_B T} + \frac{q\sqrt{\frac{qV}{\pi\epsilon t}}}{rk_B T} \right] \quad (3)$$

n_c is the density of charge carriers in the conduction band, μ is the electronic drift mobility and r is the coefficient (range: 1–2). The validity of P-F emission gives a linear fit in the plot of $\ln(J/V)$ vs. $\sqrt{V}$. In P-F emission mechanism, the positively charged oxygen vacancies act as trapping centres. At higher voltage, energy barrier for the transport of electrons is reduced and the oxygen vacancies migrate towards the negative electrode. The electric field induced de-trapping of the electrons from traps into the conduction band increases n_c and current.

3. Experimental results

3.1. Analysis of basic I–V characteristics

We have measured the I–V curves within three bias voltage limits ± 50 V, ± 100 V and ± 200 V and analyzed the data using existing models. The plots and data analysis of the samples are not shown for all the voltage ranges to optimise the number of figures. The I–V characteristics of α -Cr_{1.45}Ga_{0.55}O₃ sample have been analyzed for voltage limits ± 50 V and ± 200 V. Fig. 1 has shown I–V data for P (a), N (b) and 20P (c) modes for $V_{\max} \pm 50$ V. In the measurement sequences (S1 $\rightarrow$ S2 $\rightarrow$ S3 $\rightarrow$ S4) of P mode, the I–V curve initially

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