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A novel structure ZnO-Fe-ZnO thin film memristor

Y.P. Santos*, E. Valença, R. Machado, M.A. Macêdo

Physics Department, Federal University of Sergipe, 49100-000 São Cristóvão, SE, Brazil

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ABSTRACT

The memristor behavior of thin films having a multilayer Pt/ZnO/Fe/ZnO/ITO structure, deposited using RF/DC magnetron sputtering, was studied. The iron layer between the ZnO layers facilitates the change in the resistance of the device through the oxidation of the iron at the ZnO/Fe interface, thus generating oxygen vacancies and providing electrons from the redox reaction between γ-Fe_2O_3 and Fe_3O_4. The main mechanisms of conduction include Poole–Frenkel emission and Fowler–Nordhein tunneling with the ion migration, oxygen vacancies, and redox reactions of iron oxides (γ-Fe_2O_3 and Fe_3O_4). The response of the device to sequential voltage pulses in terms of variation in resistance, R_{ERASE}/R_{WRITE} ratio, retention time, and control of the resistance state, through control of the applied voltage, was also evaluated.

1. Introduction

Initially proposed by Chua [1], based on a theory that considers symmetry in relation to other elements of electric circuits, the "memristor"—an abbreviation for "resistive memory"—has been defined as the fourth fundamental element of circuits, capable of establishing a relationship between magnetic flux and an electric charge [1]. An actual device was realized a few decades after the theoretical proposition by Chua. In 2008, researchers at Hewlett-Packard Labs first observed memristor behavior in a metal-insulator-metal (MIM) structure. It was composed of a thin film of titanium dioxide functioning as an insulating layer, with platinum as a metallic layer [2].

Owing to its potential as a viable alternative to microprocessors with a complementary metal-oxide-semiconductor (CMOS) architecture, memristor devices have attracted great interest in the area of microelectronics and nanoelectronics in that they offer, among other features, an excellent opportunity to miniaturize devices, an ever-present technological goal. Furthermore, they offer great potential for scalability and low power consumption in memory-type applications [3]. Recently, efforts have also been directed towards the development of hybrid memristor-CMOS structures [4].

The possible applications of memristors are diverse, covering areas such as neuromorphic systems [5], neural networks [6,7], optoelectronics [8], radiation sensors [9], and non-volatile memory [3]. In general, memristors have some kind of MIM structure, capable of simultaneously exhibiting conductive and insulating behavior [3].

There have been very few studies of structures in which a metal layer is inserted between two insulating layers, although there have been a few studies in the area of resistive switching that explore this structural configuration [10–12]. However, for memristor applications, to the best of the authors' knowledge, no studies have addressed this configuration. The insertion of a metallic layer into an oxide layer allows the performance of a device to be optimized [10]. In the present study, to improve the device's performance as a memristor, a Fe layer was inserted between two layers of ZnO to facilitate a change in the resistance of the device through the supply of oxygen vacancies (iron oxidation at the ZnO/Fe interface). Indium-tin-oxide (ITO) was selected for the electrodes, given their high conductivity, chemical stability, and high optical transmission in the visible region.

The present study investigated the memristor properties of a thin film of ZnO/Fe/ZnO using structural and electrical characterizations, its conduction mechanism using I × V curves, and its retention efficiency and applicability to microelectronics.

2. Experimental details

Thin films of ZnO/Fe/ZnO were grown on ITO substrates (having a nominal thickness of 100 nm, deposited on glass (Asahi Glass)) using radio-frequency (RF) magnetron sputtering system (AJA International). A ZnO target (99.99%, Macashew Technology) and a Fe target were sputtered at a target power of 100 W under an argon pressure of 20 mTorr in the absence of oxygen. The deposition time of both the top and the bottom ZnO layers was 180 min, whereas that of the intermediate Fe layer was 400 min. The film was characterized by X-ray diffraction (XRD) and X-ray reflectance (XRR) using a D8-ADVANCE diffractometer (Bruker-AXS) with a typical copper tube as a target

* Corresponding author.
E-mail address: yvensp.santos@gmail.com (Y.P. Santos).

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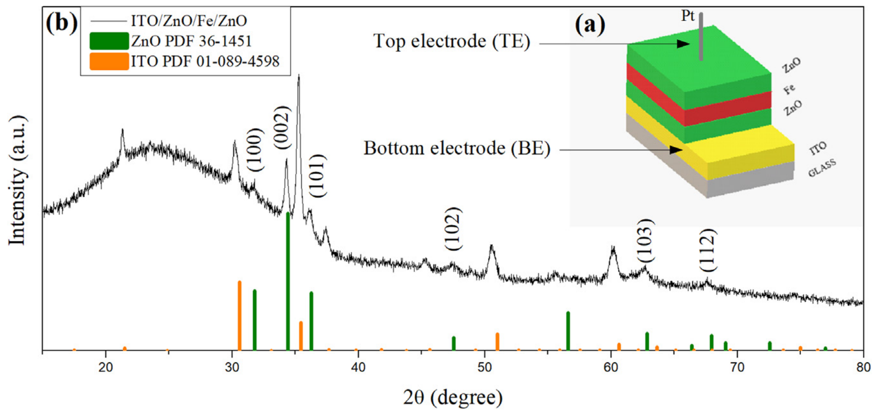

Fig. 1. (a) Schematic illustration of ITO/ZnO/Fe/ZnO sample and positioning of electrodes. (b) X-ray diffraction pattern of ITO/ZnO/Fe/ZnO film deposited by magnetron sputtering.

operating at a voltage of up to 40 kV and a current of up to 40 mA, adjusted according the implemented technique (XRD or XRR). The electrical characterization was performed using a metal-tipped Pt electrode with a diameter of approximately 200 µm. To prevent damage to the top ZnO layer, the platinum tip was connected to an optical microscope, thus ensuring fine control of the clearance between the Pt electrode and the film. A current/voltage source (Keysight Agilent B2901) was used to perform the I/V measurements. All the operating voltages were applied to the upper Pt electrode at 293 K, while the lower ITO electrode was kept grounded, in a free atmosphere and subject to ambient light.

3. Results and discussion

Fig. 1(a) is a schematic of the stacking sequence of the thin film layers in the ITO/ZnO/Fe/ZnO structure. In the resulting configuration, the top electrode (TE) is a platinum tip, while the bottom electrode (BE) is ITO itself. The literature contains several studies of MIM-type structures for application to memristors, wherein the insulating layer consists of oxides, such as ZnO [13], TiO_2 [2], NiO [14], or ZrO_2 [15], or a combination of oxides acting as the active layer [16,17]. Although various new thin-film structures are being developed for memristor devices, structures consisting of a metal layer inserted between two layers of oxide are novel and their potential remains unexploited.

Fig. 1(b) shows the XRD pattern for the ITO/ZnO/Fe/ZnO film which points to the presence of ITO and ZnO. The presence of the main crystalline ZnO planes present in the reference diffraction pattern indicates that the sample did not exhibit any preferential orientation in any direction (the main crystalline planes of ZnO are shown in Fig. 1), which differs from the structure of ZnO reported in other studies using the same deposition technique [18,19]. Although there are no traces of Fe in the XRD pattern, this would be expected because it is an inhomogeneous layer with a reduced thickness (~ 30 nm), and presents high roughness which may have contributed to the formation of an amorphous layer, as demonstrated in XRR results. The absence of any preferential orientation in the ZnO also indicates the presence of Fe in the sample. This is because the stress caused by the Fe atoms during the deposition of the film under a layer of ZnO of reduced density (the production environment of the films is lacking in oxygen, and therefore favorable to the appearance of vacancies) would destabilize the crystalline network of ZnO, and consequently, prevent the growth of the ZnO film in a preferential direction.

Fig. 2 shows the results obtained by XRR. By adjusting the experimental result, the thicknesses of the different layers were estimated using home-made software (IILXRR2013 [20]) based on the Parratt [21] and Nevot-Crocê [22] formalism. The results are listed in Table 1. The ITO thickness was in good agreement with the nominal value

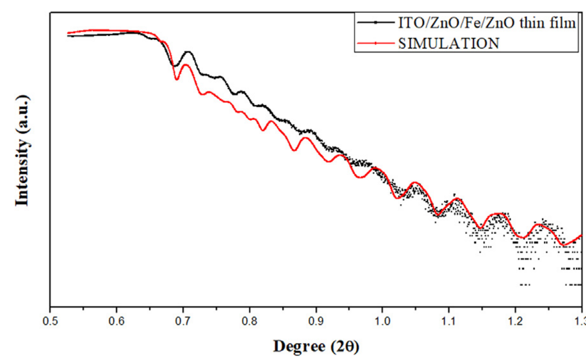

Fig. 2. XRR experimental and simulated pattern of ITO/ZnO/Fe/ZnO film.

Table 1
Results obtained by XRR.

Formula/ layer	Thickness (nm)	Roughness (nm)	density(g/ cm3)	Electronic density(Å^{-3})
ZnO-top	85.0	2.8	3.953	1.068
Fe	30.8	16.1	5.608	1.504
ZnO-bottom	86.5	2.15	4.822	1.302
ITO	100.0	6.5	6.459	1.609

provided by the substrate supplier (100 nm); hence, the thicknesses of the other layers is believed to be a good estimate of the actual value. The ZnO layer deposited on the ITO layer (ZnO-bottom) is not as rough and exhibits a higher specific and electron density because of the homogeneity of the ITO substrate. On the other hand, the ZnO layer deposited on the Fe layer (ZnO-top) exhibits a greater roughness and a lower specific density and electron density than the ZnO-bottom layer. This disparity is directly related to the presence of the Fe layer; the greater roughness of the Fe layer favors the more disordered growth of the ZnO-top layer and consequently forms a structure that is rougher and more porous.

Fig. 3(a, b) shows the current measured in the sample as a function of the voltage applied to the device. Five positive consecutive sweeps (0–5 V) and five consecutive negative sweeps (0 to − 5 V) were performed to study the electrical behavior of the device. A continuous increase in the conductance is observed during the application of positive sequential sweeps, whereas the reverse trend is observed for the opposite polarity. This progressive increase and decrease in the conductance is a characteristic behavior of memristor devices, unequivocally pointing to the film being classifiable as a memristor.

Fig. 3(c) shows the current and voltage applied to the device in the scans performed in (a) and (b) as a function of time, clearly showing the