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An Assessment on Dynamic Physical Changes in Dielectric Thin Films: A View Toward the Mechanism Behind The Resistive Switching

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Abstract

Sheer two-terminal devises – often referred to as memristive device – that make use of a dielectric thin film sandwiched by a pair of metallic electrodes routinely exhibit distinctive electrical characteristics – reversible resistive switching (RRS). While a myriad of experimental and theoretical studies intended for acquiring coherent understanding of the physicochemical origin of RRS exist, further assessments are indispensable to substantially improve their practical endurance and adaptably customize their inherent electrical characteristics. In this paper, the phase-field (PF) methodology is implemented in a study of the formation and annihilation of conductive domains often deemed to be the origin of RRS. The study centers the dynamical evolution of conductive domains under the influence of electric potential and a temperature gradient.

1. Introduction

First theoretically discussed more than forty years ago [1], a new type of circuit element, often referred to as memristive device, that simply comprises a thin film of dielectric material inserted between a pair of metallic electrodes was finally commenced in hardware in 2008 [2]. Memristive devices capable of retaining a given electrical state explicitly set by external stimuli prompted a large number of experimental and theoretical studies many of which were intended to gain comprehensive understanding of the physicochemical origin of RSS [3, 4]; however, more complementary – both experimental and theoretical – analyses are certainly imperative to considerably advance their practical implementations in hardware.

Memristive devices offer diverse opportunities in studying ways by which a thin film made of dielectric material responds to such external stimuli as electric potential applied across the film. A collection of memristive devices were found to display two distinctive dynamics – irreversible and reversible; the former is customarily designated as *electroformation* while the latter is commonly regarded as RSS. Although RRS is consistently highlighted in operating memristive devices, the study of electroformation is equally critical from a perspective of practical implementations, especially for optimizing overall performance, enhancing yield, and improving reliability.

In this paper, the phase-field methodology is harnessed in studying both electroformation and RSS by invoking the dynamical evolution of conductive domains under the influence of electric potential and a temperature gradient provided as external stimuli.

2. Approach

An interface that separates two material phases chemically distinctive one another is intrinsically dynamic, often producing unique microstructures via diverse interactions between interface energy, bulk free-energy, electrothermal, and electrochemical phenomena. In our approach, provided the nature of such an interface, incomprehensible electrical behaviors – electroformation and RSS – seen in dielectric thin films are studied as dynamic evolution of complex microstructures emerging at the interface that minimizes the total energy over time. The reduction in the total energy gives rise to the formation of an electrically conductive path (ECP), leading to characteristic transport properties of electric charges as schematically illustrated in Fig. 1. In many existing models, the presence – rather than the formation – of a single ECP is set as the initial condition, and then, the dependence of physical properties of the pre-existing ECP on such parameters as electrical potential and temperature is evaluated to reproduce RSS [5].

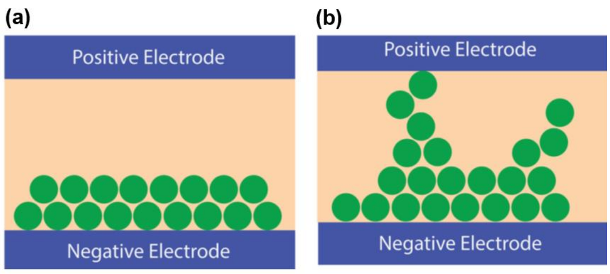

Fig. 1: The conducting channel formalism illustrated by a charged species cluster model. (a) illustrates the pristine preformed state. (b) illustrates a possible formed state, showing one complete conducting channel forming a continuous path between the negative and positive contacts. The green circles represent discrete charged species, hosted by a dielectric, shown in tan [7].

Our study was originally motivated by several key implications obtained by self-consistent continuum electrothermal

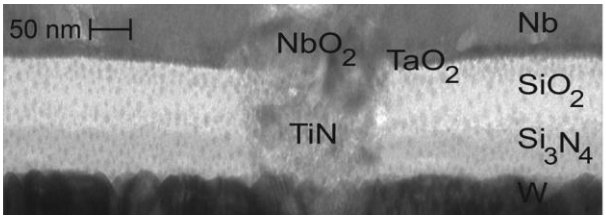

Fig. 2: Analytical TEM cross-sectional image of the self-assembled monolithically integrated NbO_2 selector self-aligned to a TaO_2 memristor after electroforming, with approximate locations of material boundaries. The TiN via is 75nm in diameter and is axially symmetric [6].

simulation and experimental characterization of niobium oxide-tantalum dioxide selector-memristor structures as shown in Fig. 2, focusing specifically on electroformation [6]. Our analysis based on continuum electrothermal formulation suggests that Joule heating produces conditions under which oxidation of niobium by tantalum dioxide and its subsequent insulator-metal transition of niobium oxide takes place. The insulator-metal transition causes changes in resistivity, decreasing local Joule heating and initiating a self-limiting effect on spontaneous electroformation. The continuum formulation naturally admits inhomogeneous mutual interaction, geometry, and interface effects that are central to the transient study of electroforming.

Our next step is to employ the phase-field methodology to assess the dynamical evolution of ECPs. With the phase-field methodology, the assumptions of an a priori ECP and the presence of a specific transport mechanism are abandoned, and our model is instead formulated as a diffuse interface problem subject to a variational principle.

3. Results

In utilizing the phase-field methodology [7], we consider Fig. 3(a) illustrating a pristine dielectric thin-film structure composed of a conducting state (the top region) and a nonconducting state (the bottom region), separated by an interface represented by the dotted horizontal line. The entire thin-film structure measures 50 x 10 nm^2 and the interface is located at 4 nm from the bottom edge. The interface is characterized as a diffuse interface, established by providing an initial normalized charge concentration *c* in the range of 0.1~0.3 for the nonconducting state and 0.7~0.9 for the conducting state, respectively.

The nature of the formation of ECPs is revealed in Fig. 3(b) and Fig. 3(c) – two snapshots representing a transient stage and the final stage, respectively, of irreversible electroformation. A tendency to reduce the total surface area (i.e., the total length of the interface) is clearly pictorialized, being distinguished by the conducting domains shown in red and the nonconducting domains shown in blue. In Fig. 3b, the formation of a number of small-size clusters in conducting states (reddish colors) is seen, in contrast, in Fig. 3c, a single continuous ECP is established. These results suggest that a unique interface microstructure develops from the initial interface under the influence of electrical potential and leading

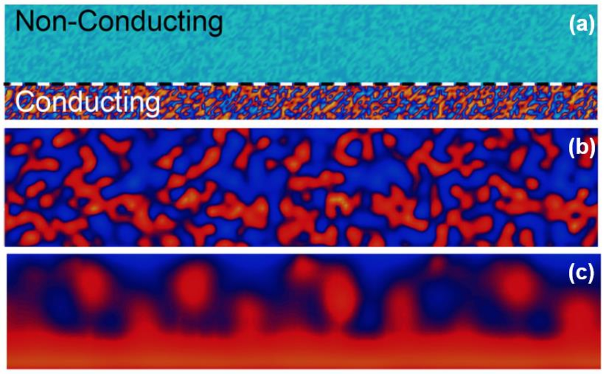

Fig. 3: (a) illustrates the pristine dielectric thin-film structure, (b) and (c) show two snapshots representing a transient stage and the final stage, respectively, of irreversible electroformation [7].

to the birth of a distinctive ECP running through the nonconducting regions. The equilibrium interface formed by the conducting state and the nonconducting state intrinsically describes the morphology of the conducting channel, represented by an envelope of cluster-like domains composed of many discrete charge carriers.

4. Conclusions

Our approach based on the phase-field method successfully predicts the formation and annihilation of ECPs in dielectric thin-film structures equivalent to a range of memristive devices, offering an alternative computational formulation based on metastable states treated at the atomic scale, requiring no pre-existing ECP and its fundamental carrier transport mechanisms.

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