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# 1 Resistance controllability and variability improvement in a TaO<sub>x</sub>-based 2 resistive memory for multilevel storage application

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8 In order to obtain reliable multilevel cell (MLC) characteristics, resistance controllability between  
9 the different resistance levels is required especially in resistive random access memory (RRAM),  
10 which is prone to resistance variability mainly due to its intrinsic random nature of defect generation  
11 and filament formation. In this study, we have thoroughly investigated the multilevel resistance  
12 variability in a TaO<sub>x</sub>-based nanoscale (<30 nm) RRAM operated in MLC mode. It is found that the  
13 resistance variability not only depends on the conductive filament size but also is a strong function  
14 of oxygen vacancy concentration in it. Based on the gained insights through experimental  
15 observations and simulation, it is suggested that forming thinner but denser conductive filament may  
16 greatly improve the temporal resistance variability even at low operation current despite the  
17 inherent stochastic nature of resistance switching process. © 2015 AIP Publishing LLC.

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18 Resistive random access memory (RRAM) is one of the  
19 most promising next generation memory candidates espe-  
20 cially for storage class memory applications due to its scal-  
21 ability, structural simplicity, and backend process compatible  
22 stackability.<sup>1-3</sup> In addition, TaO<sub>x</sub> as a RRAM switching  
23 material have been found to be of commercial interest due to  
24 its superior material property as well as memory perform-  
25 ance.<sup>3-6</sup> To fulfill the increasing demand for low cost and  
26 higher density memory, the most obvious way to increase the  
27 storage capacity is by decreasing the physical size of the  
28 device to a nanoscale dimensions, and in RRAM the scalabil-  
29 ity down to 10 nm has been demonstrated.<sup>7,8</sup> Other interesting  
30 approach is to stack the devices 3-dimensionally, for which  
31 two feasible architectures of crossbar and vertical RRAM are  
32 reported.<sup>9</sup> However, both the methods require complex exper-  
33 imental processes and may suffer from other limitations.  
34 Another alternative and simpler way to increase storage  
35 density is to use multilevel cell (MLC) storage technology, in  
36 which more than one bit per cell can be stored without further  
37 decreasing the physical device size.<sup>10,11</sup> However, precise  
38 control over the resistance of the different resistance levels  
39 should be assured for reliable MLC operation especially for  
40 RRAM, which is known to suffer from variability and reli-  
41 ability issues due to certain randomness in its switching  
42 process.<sup>12-14</sup>

43 In RRAM, MLC characteristics can be obtained by vari-  
44 ous methods such as by varying the switching current (or  
45 compliance current), where different levels of low resistance  
46 state (LRS) can be obtained due to change in the total num-  
47 ber of constituent defects of the conductive filament (CF) or  
48 by controlling the maximum reset voltage during reset

operation which can result different levels of high resistance 49  
state (HRS) due to variation in the gap between CF tip and 50  
metal electrode.<sup>10,11</sup> MLC behavior by varying the program/ 51  
erase pulse width is also reported.<sup>11</sup> Although MLC proper- 52  
ties can easily be obtained in RRAM, its successful imple- 53  
mentation is dictated mainly by the ability to precisely 54  
control the resistance margin between the two resistance 55  
levels. One of the most critical factors, which can degrade 56  
this margin, is the switching cycle-to-cycle variability. 57  
Therefore, the feasible approaches to minimize it should be 58  
delved into through the enhanced understanding of the 59  
responsible factors. The origin of this variability is mainly 60  
related to the change in the total content of the oxygen 61  
vacancy defects present in the CF after each switching 62  
event.<sup>12</sup> This compositional variation in the conductive chan- 63  
nel leads to the conductivity fluctuation. More fundamen- 64  
tally, it can be understood in terms of having different 65  
activation energy of defect migration due to the random 66  
location and different surroundings of the defects with in the 67  
conductive channel. 68

69 In this study, we aim to develop enhanced understanding  
70 on the variability issue through experimental observation  
71 and simulation studies and correlate it with the CF size and  
72 defect concentration in it. For this, we fabricated nanoscaled  
73 (<30 nm) RRAM devices with TaO<sub>x</sub> as a switching material.  
74 Based on the analysis of experimental observations and simu-  
75 lation, a possible way to minimize the variability between  
76 resistance levels for reliable MLC operation is proposed.  
77 The results show that the temporal resistance variability may  
78 greatly be improved even at low switching current by form-  
79 ing denser filament with smaller size. Monte-Carlo calcula-  
80 tions show good agreement with the experimental data.

81 The device was fabricated on a patterned substrate with  
82 30 nm TiN bottom electrode (BE) in a planner type device  
83 structure [see the inset of Fig. 1(a)]. The TiN electrode was  
84 deposited by CVD and its sides were protected by 100 nm

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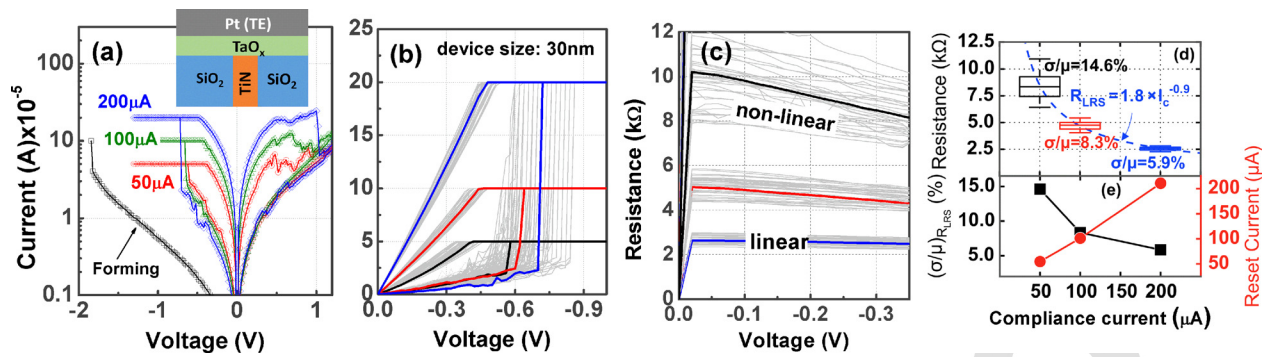


FIG. 1. (a) Forming and resistive switching behavior of fabricated 30 nm sized TaO<sub>x</sub>-based resistive memory device with MLC characteristics. (b) Multiple MLC current-voltage (I-V) curves of all the LRS levels showing stable MLC characteristics. (c) Corresponding resistance-voltage (R-V) plot of all the low resistance levels confirming nonlinear conduction at smaller switching current. (d) Resistance of LRS and its distribution as a function of I<sub>C</sub>. Both decrease with increasing I<sub>C</sub>. (e) Evolution of LRS variability ( $\sigma/\mu$ ) and reset current (I<sub>reset</sub>) with I<sub>C</sub>.

thick SiO<sub>2</sub> layer. Tantalum oxide (TaO<sub>x</sub>) switching layer of ~10 nm was deposited by reactive sputtering using Ta metal target and Ar+O<sub>2</sub> gases at room temperature. The rf power and working pressure were 100 W and 3 mTorr, respectively. Finally, inert (Pt) top electrode (TE) was sputter deposited on TaO<sub>x</sub> switching layer. For electrical measurements, voltage was applied on TE and BE was connected to the ground.

Multilevel cell operation and resistance variability analysis of fabricated 30 nm size RRAM device is presented in Fig. 1. First, the device was formed by applying a negative voltage on Pt TE, as shown in Fig. 1(a). Small voltage of < -2.0 V was sufficient to form the device. After forming, the device exhibits resistive switching behavior and MLC characteristics by varying the switching current, as shown in Figs. 1(a) and 1(b). Multiple cycles for each level have been shown to ensure the stable resistance switching. Three different levels of LRS were obtained with same HRS, which can be used in 2-bit per cell storage. In addition, it was observed that the LRS current-voltage (I-V) gradually shifted from an ohmic to a nonlinear behavior as current compliance (I<sub>C</sub>) was decreased down to 50 μA (Fig. 1(c)) due to decreased number of vacancy defects in the CF leading to the change in the current conduction mechanism. Simultaneously, the resistance margin between the resistance levels is reduced because of cycle-to-cycle resistance fluctuations. The resistance distribution of 50 consecutive cycles for each compliance is shown in Fig. 1(d), where the highest variability of 14.6% is observed for I<sub>C</sub> of 50 μA. The read voltage was 0.25 V. Furthermore, the resistance of LRS (R<sub>LRS</sub>) decreased with increasing I<sub>C</sub> [Fig. 1(d)]. The slope of R<sub>LRS</sub> vs I<sub>C</sub> was found to be -0.9, which is close to the reported value of -1.<sup>12</sup> On the other hand, maximum reset current (I<sub>reset</sub>) increases linearly and coefficient of variation ( $\sigma/\mu$ ) or variability of R<sub>LRS</sub> decreases with increasing I<sub>C</sub>, as shown in Fig. 1(e). The decrease in the variability is due to the increase in the number of oxygen vacancy defects in the CF with increasing I<sub>C</sub>. Considering the dependence of LRS I-V nonlinearity on I<sub>C</sub> and variability behavior, it can be said that by increasing I<sub>C</sub>, both CF size and oxygen vacancy defect density in it may increase. In other words, R<sub>LRS</sub> depends on filament size and its electrical conductivity.

The different electrical conduction behavior (linear/nonlinear) of the LRS I-V curves observed at different

switching currents [Fig. 1(c)] can be understood by the oxygen vacancy concentration (n<sub>V<sub>O</sub></sub>) falling under a threshold value (n<sub>TAC</sub>), which is the minimum concentration of oxygen vacancies below which trap assisted conduction (TAC) dominates and I-V becomes nonlinear, while above this concentration conduction is linear (ohmic).<sup>15-18</sup> This can be accounted by the following equation:

$$\sigma_e(n_{V_o}) = \beta \times n_{V_o} \times \exp\left(-\frac{E_a(n_{V_o})}{kT}\right), \quad (1)$$

$$\text{with } E_a(n_{V_o}) = E_{a_0} \times f(n_{V_o}), \quad (2)$$

where  $\sigma_e$  is the electrical conductivity, E<sub>a</sub> is the activation energy,  $\beta = 1 \times 10^{-23} \text{ S m}^{-1}$ , k = Boltzmann constant, T = absolute temperature, and E<sub>a0</sub> = 0.4 eV. This approach has already been employed to describe resistance switching phenomena or early data-retention failure in RRAM employing either HfO<sub>x</sub><sup>15,16</sup> or TaO<sub>x</sub>.<sup>17,18</sup> The corresponding plot of  $\sigma_e$  as a function of n<sub>V<sub>O</sub></sub> is shown in Fig. 2. In the previous studies,<sup>15-18</sup> a piecewise linear and decreasing function f [Eq. (2)] was employed to account for the decrease of conduction activation energy at increasing defect concentrations which, in turns, increases  $\sigma_e$  in the CF region. Although a proper theoretical justification is still

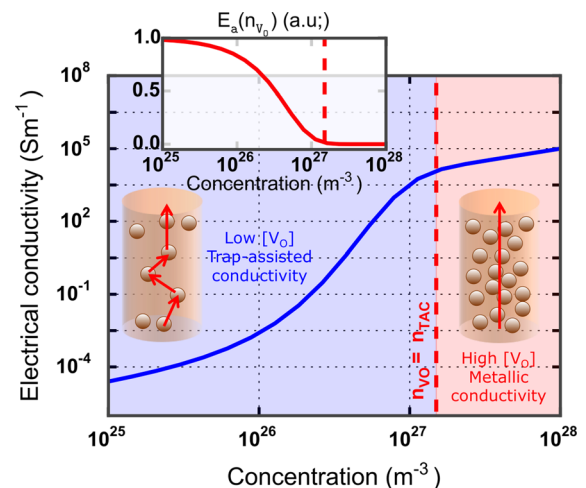


FIG. 2. Calculated electrical conductivity as function of oxygen vacancy concentration [Eq. (1)]. Inset shows the evolution of the activation energy with respect to n<sub>V<sub>O</sub></sub>.

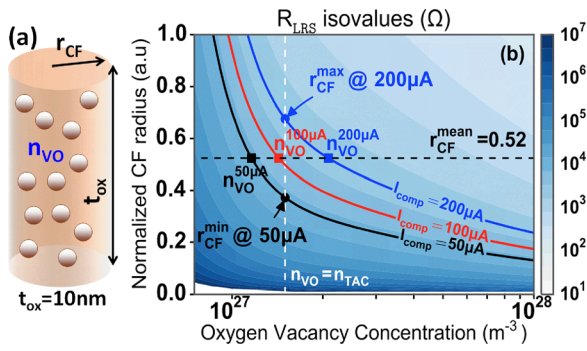


FIG. 3. (a) Schematic description of the conductive filament. (b) Maps of  $R_{LRS}$  as function of conductive filament radius and oxygen vacancy concentration. LRS isolines correspond to experimental mean LRS resistance values at different  $I_C$ .

147 lacking, this first-order approximation showed satisfactorily  
 148 and quantitative agreement with data.<sup>15,17,18</sup> In this work,  
 149 we employed a sigmoid function [see the inset of Fig. 2]  
 150 rather than a piecewise linear function to describe the  
 151 decreasing  $E_a$  with  $n_{VO}$ . Although such a dependency is only  
 152 justified as a piecewise decreasing function, it offers the nu-  
 153 merical advantage of being indefinitely differentiable in the  
 154 whole range of  $n_{VO}$  to conduct further numerical or analyti-  
 155 cal studies. In the following, a value of  $n_{TAC}$  of  
 156  $1.5 \times 10^{27} \text{ m}^{-3}$  was employed (marked by vertical dashed  
 157 line in Fig. 2), which corresponds to an inter-distance below  
 158 1 nm between oxygen vacancies. Considering the  $R_{LRS}$   
 159 equal to the resistance of CF ( $R_{CF}$ ) in LRS and cylindrical  
 160 filament [Fig. 3(a)],  $R_{CF}$  can be written as

$$R_{LRS} \approx R_{CF} = \left( \frac{1}{\sigma_e(n_{VO})} \right) \times \left( \frac{t_{ox}}{\pi \times r_{CF}^2} \right), \quad (3)$$

161 where  $t_{ox}$  is the oxide thickness ( $= 10 \text{ nm}$ ) and  $r_{CF}$  is the  
 162 conductive filament radius. Figure 3(b) shows the  $R_{LRS}$  isovalue  
 163 curves of all the resistance levels as a function of CF radius  
 164 and oxygen vacancy concentration ( $n_{VO}$ ). The  $n_{VO}$  equal to  
 165  $n_{TAC}$  is also shown by a dashed line. The dependence of  
 166  $R_{LRS}$  versus the vacancy concentration is intensified as the

concentration falls below  $n_{TAC}$  (i.e., non-ohmic regime) as  
 167 indicated by sharper  $R_{LRS}$  isovalues [Fig. 3(b)]. As vacancy  
 168 generation and filament formation are intrinsically random in  
 169 RRAM, vacancy concentration change in the filament is in-  
 170 evitable during cycle-to-cycle switching. Consequently,  
 171 small changes in  $n_{VO}$  may introduce large variability in the  
 172 resistance of multilevel, in particular, when  $n_{VO} < n_{TAC}$  as  
 173 observed in the experimental results [Fig. 1]. It is interesting  
 174 to note that LRS variability not only depends on the filament  
 175 radius (size) but also is strongly dictated by vacancy concen-  
 176 tration and fluctuation in the filament. Interestingly, the  
 177 graph also shows that the different topologies of filament  
 178 (i.e.,  $r_{CF}$  and  $n_{VO}$ ) can account for the experimental mean  
 179  $R_{LRS}$  values obtained at different  $I_C$  [Fig. 3]. Given the non-  
 180 linearity (respectively, linearity) of I-V curves at  $50 \mu\text{A}$   
 181 (respectively,  $200 \mu\text{A}$ ),  $r_{CF}$  can be reasonably assumed to be  
 182 higher (respectively, lower) than  $r_{CF}^{min}$  (respectively,  $r_{CF}^{max}$ ) to  
 183 account for non-ohmic (respectively, ohmic) I-V.  
 184

To better understand the effect of vacancy concentration  
 185 on the variability, let us assume a mean filament radius  
 186 ( $r_{CF}^{mean}$ ) in between  $r_{CF}^{min}$  and  $r_{CF}^{max}$ , as shown in Fig. 3(b) by  
 187 horizontal dashed line. Three  $n_{VO}$  can now be obtained on  
 188 the  $R_{LRS}$  isovalue curves corresponding to switching currents  
 189 of  $50, 100,$  and  $200 \mu\text{A}$ . Monte-Carlo calculations have been  
 190 performed to reproduce the experimental LRS resistance distri-  
 191 bution assuming a Gaussian distribution of  $n_{VO}$  in Eq. (3).  
 192 Results are presented in Figs. 4(a)–4(c). A pretty close match  
 193 between the experimental variability data and calculations  
 194 has been achieved using the values given in Figs. 4(d) and  
 195 4(e). It is interesting to note that a small (i.e.,  $< 5\%$ ) varia-  
 196 tion in  $n_{VO}$  can result in a large (14.6%) variability in the resis-  
 197 tance for  $50 \mu\text{A}$   $I_C$  and a 5.9% (smaller) resistance variation  
 198 for  $200 \mu\text{A}$   $I_C$ . This underlines that the resistance variability  
 199 is very sensitive to the  $n_{VO}$  variation in the CF when it oper-  
 200 ates in the nonlinear region where electrical conductivity  
 201 strongly depends on  $n_{VO}$ . Conduction activation energies for  
 202 each computed  $n_{VO}$  are in the range between 0.8 meV (for  
 203  $I_C = 200 \mu\text{A}$ ) and 16.7 meV (for  $I_C = 50 \mu\text{A}$ ) [Fig. 4(e)].  $E_a$   
 204 values are in the same order of magnitude than those already  
 205 reported by Kim *et al.*<sup>17,18</sup> Figure 5 suggests possible way to  
 206

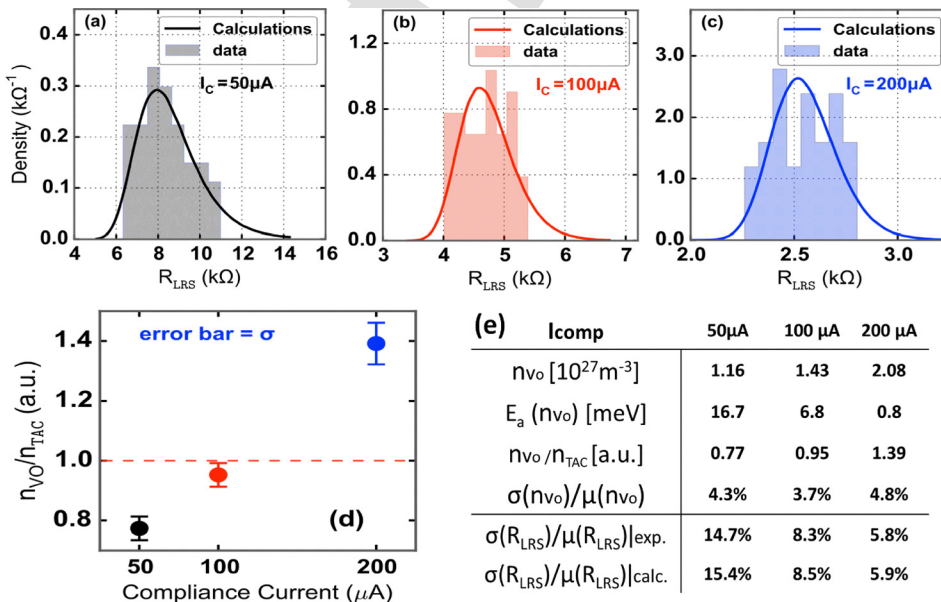


FIG. 4. (a)–(c) Experimental normalized resistance distributions (histograms) and computed density (lines) for each  $I_C$ . Densities were computed using mean values and standard deviation of  $n_{VO}$  presented in (d). Table (e) is a summary of the experimental and calculation results.

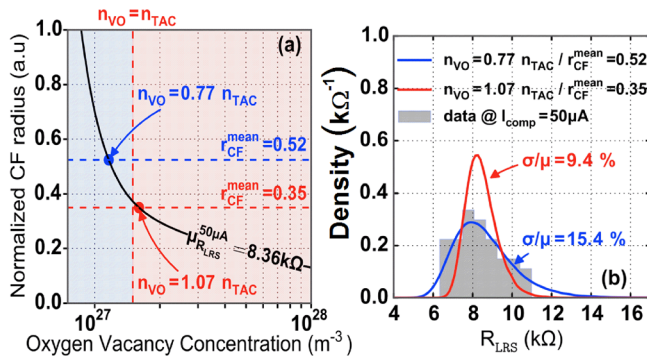


FIG. 5. (a)  $R_{LRS}$  isoline for  $I_C = 50 \mu A$ . Shaded regions correspond to trap-assisted (left side) and ohmic (right side) transport regions. (b) Experimental (histogram) and computed resistance distribution (lines) using the same relative variation of  $n_{vo}$  for different  $n_{vo}/n_{TAC}$  values. It is shown that  $n_{vo}/n_{TAC} > 1$  allows for smaller relative variation of  $R_{LRS}$ .

207 minimize the resistance variations even at low  $I_C$ . It is possible to improve the variations if we can shift to operate in the linear region instead of nonlinear region, as shown in Fig. 5(a). This can be achieved by generating smaller size filament with higher defect concentration. Calculations show that a tighter distribution with 8% variability can be achieved even at low  $I_C$  of  $50 \mu A$  if  $n_{vo}/n_{TAC}$  is increased from 0.77 to 1.07 in conjunction with  $r_{CF}^{mean}$  decreased from 0.52 to 0.35 [Fig. 5(b)]. Therefore, forming denser but thinner filament may remarkably improve the resistance fluctuation even at low operation current. Various techniques such as two step forming, material engineering, and different forming conditions can be exploited to generate different defect density filament.<sup>6</sup>

221 In this work, multilevel resistance variability was investigated on a nanoscale  $TaO_x$ -based RRAM operating in MLC mode by means of a theoretical approach assuming that non-linearity observed at low  $I_C$  is related to trap-assisted conduction resulting from low concentration of oxygen vacancy. It was found that the resistance variability not only depends on the conductive filament size but also is a strong function of oxygen vacancy concentration and fluctuation, which is supported by Monte-Carlo calculations of LRS distributions at various compliance currents. Based on this study, it is suggested that LRS variability can be improved

232 even at low  $I_C$  by forming denser CF in terms of oxygen vacancy by means of material engineering or dedicated forming techniques.

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